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## Interplay between Sb flux and growth temperature during the formation of GaSb islands on GaP

S. El Kazzi,<sup>1</sup> L. Desplanque,<sup>1</sup> X. Wallart,<sup>1</sup> Y. Wang,<sup>2</sup> and P. Ruterana<sup>2</sup>

<sup>1</sup>*Institut d'Electronique, de Microélectronique et de Nanotechnologie, UMR-CNRS 8520, BP 60069, 59652 Villeneuve d'Ascq Cedex, France*

<sup>2</sup>*CIMAP UMR 6252 CNRS-ENSICAEN-CEA-UCBN, 6, Boulevard du Maréchal Juin, 14050 Caen Cedex, France*

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We investigate the influence of the Sb flux on the growth of GaSb islands on a highly mismatched (001) GaP substrate. Between low and medium Sb flux values, standard kinetics drives the GaSb island formation and their relaxation is progressively favored by 90° misfit dislocations at the GaSb/GaP interface. However, under high Sb flux, the GaSb islands are elongated in the [110] direction and their density decreases. Further experiments varying the growth temperature at fixed Sb flux confirm this finding. We relate this observation to an enhancement of Ga diffusion when the effective Sb flux on the surface is increased. This behavior is qualitatively explained by the large cohesive energy of Sb-Sb bonds present on the surface, which impede the Ga adatom incorporation. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4729548>]

### INTRODUCTION

Heteroepitaxial growth of III-Sb semiconductors has been extensively studied in recent years, and many efforts have been made to obtain high quality antimony-based heterostructures for electronic and optoelectronic applications. For example, high electron mobility InAs/AlSb heterostructures grown on semi-insulating GaAs substrates are good candidates for high frequency and low power consumption devices.<sup>1,2</sup> To reach good material quality with a low density of threading dislocations, the optimization of the (Al)GaSb buffer layer required to accommodate the 7.8% lattice mismatch is needed and has attracted much attention during the last years. In the case of GaSb/GaAs, the strain relaxation can occur via the formation of 90° misfit dislocations (MDs) at the interface with the substrate.<sup>3-5</sup> After optimization, this nucleation process can lead to a very low density of threading dislocations in the GaSb layer.<sup>6</sup> Depending on the Sb/Ga flux ratio at the initial stages of the growth, either Volmer-Weber (V-W) or Stranski-Krastanov (S-K) growth mode can occur.<sup>7-11</sup> Furthermore, it has been reported that a high Sb/Ga flux ratio leads to elongated GaSb islands either in the [110] or the [1-10] directions.<sup>12,13</sup> However, the explanation for this behavior is still unclear since it has been sometimes attributed either to anisotropy in the diffusion length of Ga and Sb adatoms or to different growth rates for the various facets of the islands.<sup>4,7</sup>

In this paper, we report on the growth of GaSb islands on a (001) GaP substrate. Since GaP and Si have nearly the same lattice constant (0.37% mismatch), optimizing the relaxation process for (Al)GaSb on GaP would open the way to the integration of antimonides on silicon, thanks to the GaP/Si antiphase domain free templates demonstrated recently.<sup>14</sup> Considering the very large lattice mismatch between GaSb and GaP (11.7%), one can expect a 3D V-W growth mode during the nucleation of GaSb on GaP with the formation of relaxed islands, which subsequently coalesce to

give rise to the epilayer.<sup>15</sup> In previous works, we have shown that the GaP surface treatment and the growth temperature are very crucial in the relaxation of GaSb islands at the initial step of growth.<sup>16</sup> In this work, we focus on the influence of the Sb flux on the formation and elongation of GaSb islands on GaP. To explain our observations, we propose a qualitative model relying on the variations of the Ga adatom diffusion length with the Sb flux and the Sb surface coverage.

### EXPERIMENT

The samples are grown by Molecular Beam Epitaxy (MBE) on exactly oriented (001)  $\pm 0.1^\circ$  GaP substrates in a Riber 32P chamber equipped with group III and V sources. The phosphorus flux is obtained by cracking phosphine (PH<sub>3</sub>) through a high-temperature injector, whereas a valve-cracker cell is used to produce Sb<sub>2</sub>. Ga and Sb fluxes are calibrated via reflection high energy electron diffraction (RHEED) specular beam intensity oscillations on GaAs and GaSb. After deoxidization of the GaP substrate under phosphorus at 700 °C, a 450 nm thick GaP buffer layer is grown at 630 °C to smooth the surface exhibiting a sharp (2 × 4) RHEED pattern. The phosphine flux is then interrupted, and the substrate is cooled down under a constant Sb<sub>2</sub> flux at 2.5 ML/s for the growth of 10 GaSb monolayers (MLs). While the exposure of Sb<sub>2</sub> begins, the RHEED pattern turns from a (2 × 4) to a (2 × 8) reconstruction. A first set of samples is used to investigate the influence of the Sb flux during GaSb deposition at 510 °C. Three samples A, B, and C are grown with an Sb flux of 1 ML/s, 2.5 ML/s, and 7.3 ML/s, respectively, with a constant Ga growth rate of 0.7 ML/s. Two other samples (D, E) are grown at 480 °C and 450 °C to investigate the growth temperature influence, the Sb flux being fixed at 2.5 ML/s during GaSb deposition. For all growth conditions, the RHEED exhibits a spotty pattern at the opening of the Ga shutter characteristic of a 3D V-W growth mode. The surface morphology of the samples is

examined by atomic force microscopy (AFM) using a Digital Nanoscope III system, working in the tapping mode. The topology of AFM images were analyzed using Mountains Map software.<sup>17</sup> The structural properties are investigated by transmission electron microscopy (TEM) using a JEOL 2010 FEG instrument operated at 200 kV.

## RESULTS

Figure 1 displays AFM images of GaSb islands on GaP within a ( $1 \times 1 \mu\text{m}^2$ ) scan area of the samples A, B, and C. In sample A, the formation of two kinds of GaSb islands (L and S) can be attributed to the large Ga adatom diffusion length under low Sb flux. Larger islands (L) grow due to the coalescence of smaller ones (S).<sup>9</sup> Figure 1(d) displays the density and the mean aspect ratio  $L_{[110]}/L_{[1-10]}$  (where  $L_{[110]}$  and  $L_{[1-10]}$  are the island length along each direction) of GaSb islands in function of the Sb flux. For the first two samples, the density increases with the Sb flux, which is a common behavior observed during III–V MBE growth where a high group-V element flux reduces the group-III element diffusion length.<sup>11</sup> However, for very high Sb fluxes (sample C), this behavior is completely reversed, and a lower density of large islands is observed. GaSb islands are elongated in the [1-10]

direction at low Sb flux and in the [110] direction when this flux is increased. An aspect ratio near unity corresponds to the maximum island density.

Cross-sectional high-resolution TEM images of samples A, B, and C are shown in Figure 2. Sample A exhibits islands with a nearly elliptical shape, with stacking faults, as indicated by the pink arrows in Figure 2(a). As can be seen in Figure 2(b), in addition to the interface misfit dislocations, stacking faults form inside the two (111) families of lattice planes and run over to the surface. This may indicate that the islands reach a complete strain relaxation by the formation of such high densities of defects, at the interface and in their volume. In sample B (Fig. 2(c)), the shape of the island evolves with facets that start to appear. As shown in Figure 2(d), an array of misfit dislocations also forms at the interface, and in this case, very few stacking faults can be seen (arrow). Figures 2(e)–2(h) show the islands in sample C seen along the [110] (Figs. 2(e) and 2(f)) and [1-10] (Figs. 2(g) and 2(h)) directions. These islands are flat with a tetragonal shape and are limited by well defined {111} and {001} facets with no stacking faults. In these HRTEM images, the  $90^\circ$  dislocation array forms in both [110] and [1-10] directions, revealing a good strain relaxation.<sup>6,15,16</sup>

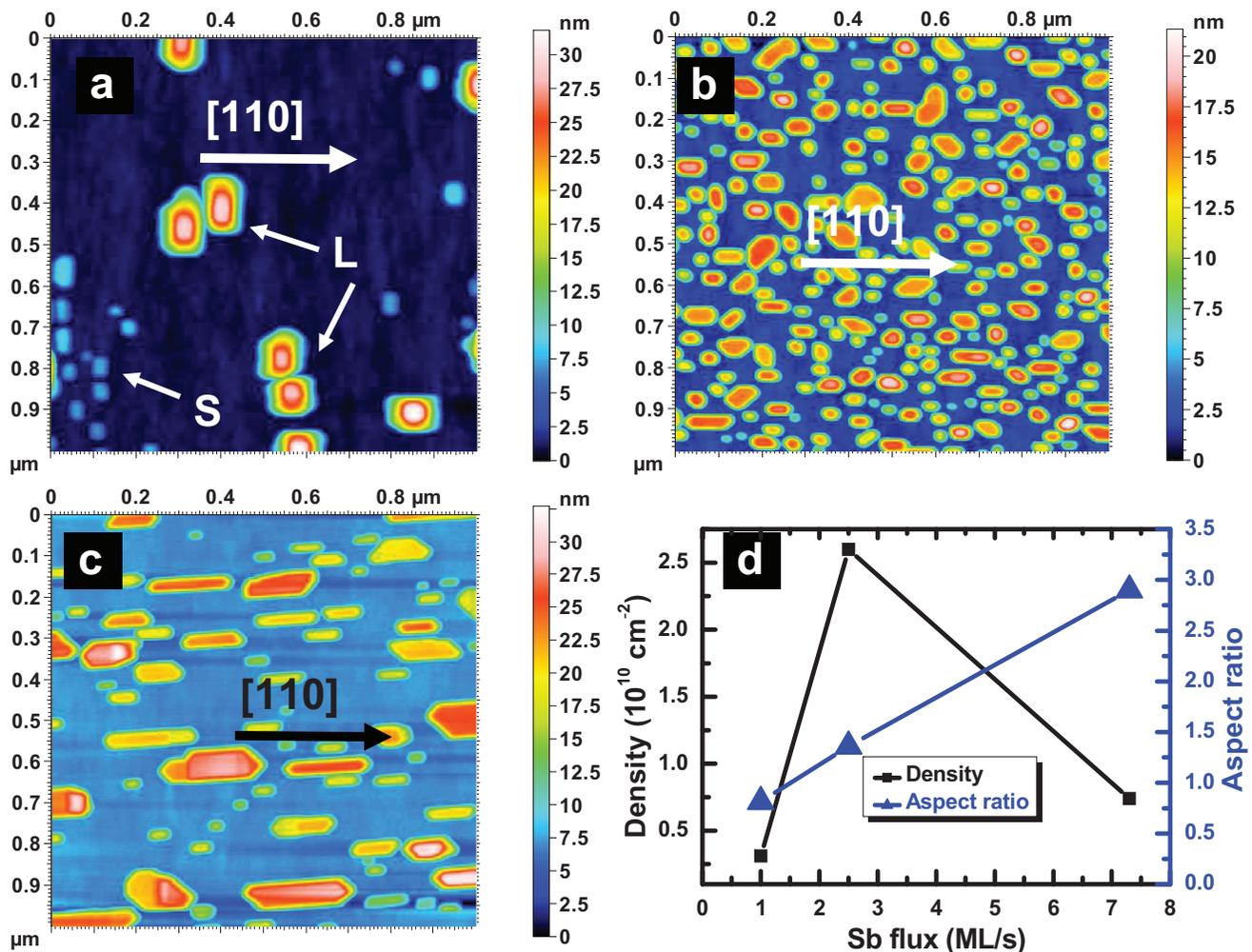


FIG. 1. AFM images of samples A, B, and C: 10 ML GaSb grown at  $510^\circ\text{C}$  with a Sb flux of 1 ML/s (a), 2.5 ML/s (b) and 7.3 ML/s (c), respectively. (d) Density (black squares) and mean aspect ratio (blue triangles) of GaSb islands in function of the Sb flux during GaSb island growth.

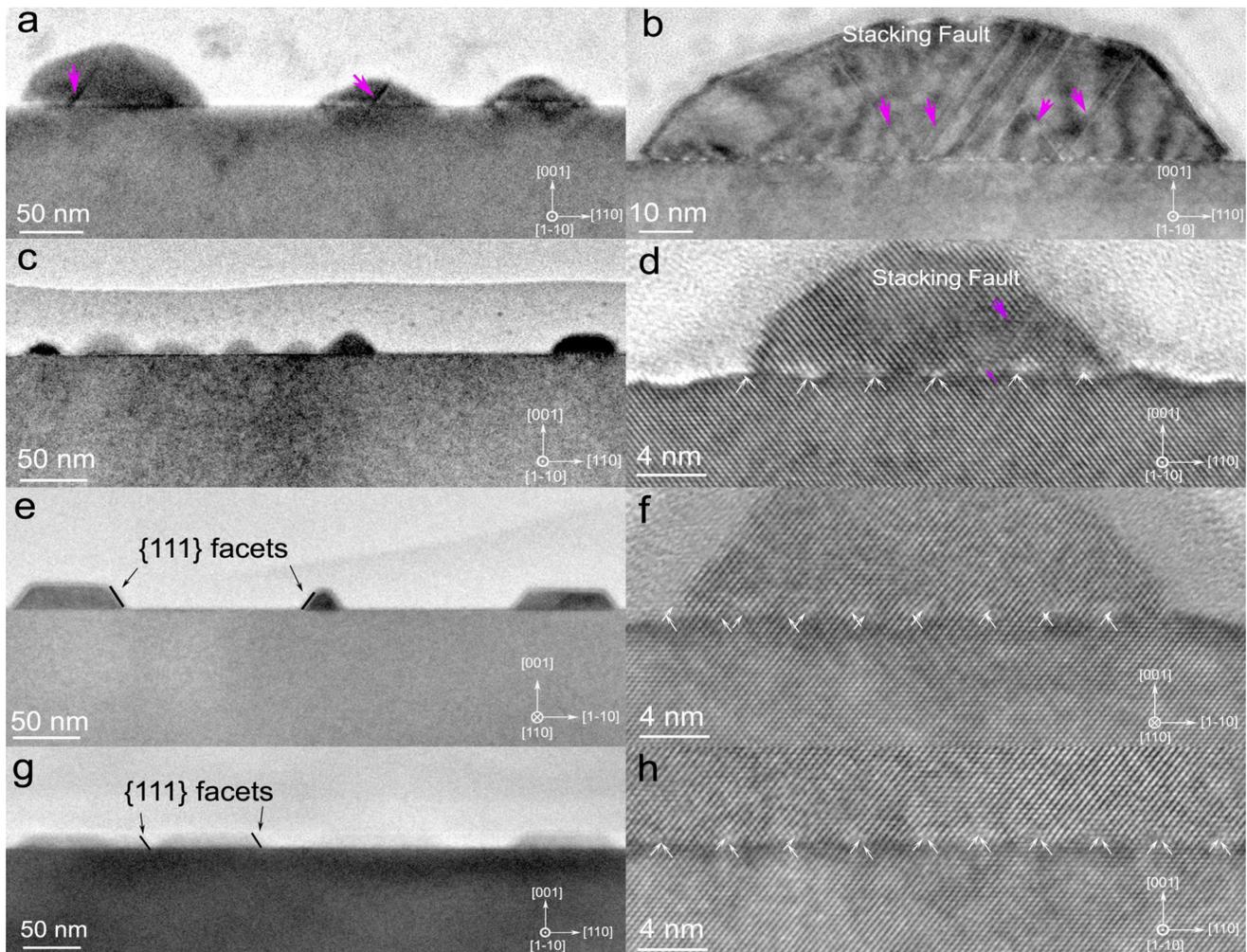


FIG. 2. Cross-sectional high resolution TEM images of samples A (a), (b) and B (c), (d) in the  $[110]$  direction. Cross-sectional high-resolution TEM images of sample C in the  $[1-10]$  (e), (f) and  $[110]$  (g), (h) directions, respectively. White arrows mark the termination of  $\{111\}$  lattice planes in the GaP substrate. Pink arrows on (b) and (d) underline the presence of stacking faults.

Figure 3 displays AFM analysis of samples E, D, and B grown at 450, 480, and 510 °C respectively. In Figure 3(d), the density and the aspect ratio of GaSb islands are given in function of the growth temperature. A meaningful aspect ratio in sample E cannot be established due to a nearly full coalescence in the  $[110]$  direction. The mean heights of samples B, D, and E are 8.6, 4.0, and 1.7 nm, respectively. This evolution seems to agree with common kinetics: at low temperatures, the reduced Ga adatom diffusion length should induce a large density of small GaSb islands. However, Figure 3(d) shows that a low growth temperature limits the island density and increases the island aspect ratio (i.e., the islands are more elongated at 450 and 480 °C than at 510 °C). This would imply an increase of the Ga adatom diffusion length, unexpected when decreasing the growth temperature.<sup>18</sup>

## DISCUSSION

To explain the above depicted behavior, one should take into account the Sb coverage of the surface. Indeed the lower reevaporation of Sb at low temperature results in a larger Sb coverage on the GaP surface. Therefore, two effects are competing, acting conversely on the Ga adatom diffusion length:

lowering the growth temperature reduces this length whereas a larger Sb surface coverage seems to increase it.

At high Sb overpressure, an Sb coverage larger than 1 ML with the formation of Sb-Sb bonds on the surface has been reported for both the (001) GaSb (Ref. 19) and the  $(2 \times 8)$  Sb-terminated (001) GaAs surfaces.<sup>20</sup> In the present study, the observation of a  $(2 \times 8)$  reconstruction under Sb flux is a strong indication of the formation of Sb-Sb bonds at the GaP surface. Since the Ga-Sb and Sb-Sb bond energies are rather similar,<sup>21</sup> the driving force to break a Sb-Sb bond and form a Ga-Sb one is rather low. Then, the Ga incorporation is reduced, and its diffusion is enhanced. As mentioned before, high Sb fluxes promote the formation of fully relaxed islands with well-defined  $\{111\}$  facets. As illustrated in Figure 4, on the Sb-terminated  $\{111\}_b$  facets, the Sb-Sb bonds on the surface will impede Ga adatom incorporation and favors it on Ga-terminated  $\{111\}_a$  facets. These latter advance faster leading both to the observed  $[110]$  island elongation and to the reduced island density.

The key factor is the competing influence of the growth temperature and the Sb flux on the Ga adatom diffusion length as displayed on the sketch of Figure 5. The black line separates two domains of growth conditions, depending on

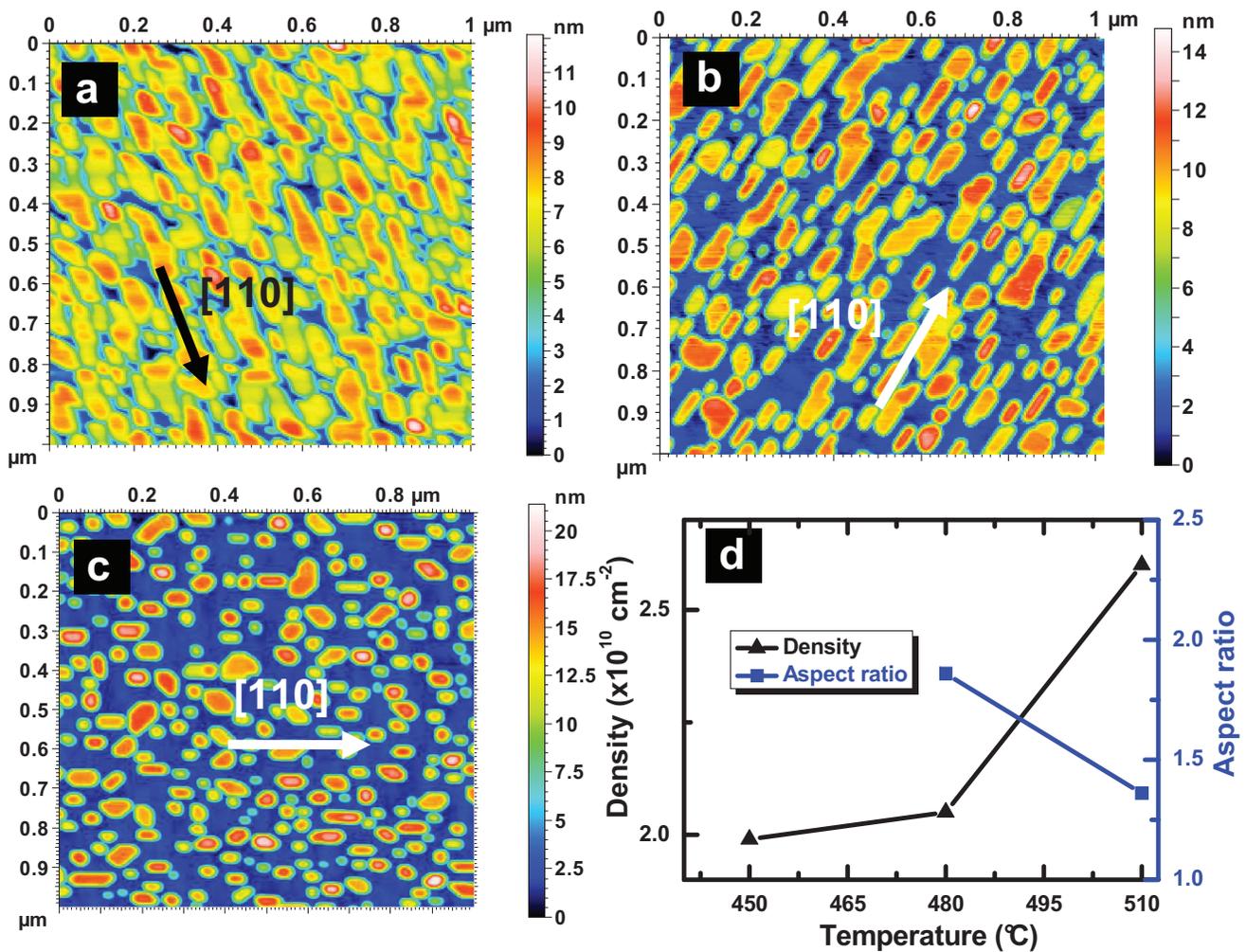


FIG. 3. AFM images of samples E, D, and B: 10 ML GaSb grown with a 2.5 ML/s Sb flux at 450 °C (a), 480 °C (b), and 510 °C (c), respectively. (d) Density (black triangles) and mean aspect ratio (blue squares) of GaSb islands in function of the growth temperature.

whether or not stable Sb-Sb bonds are present on the surface. Below the line, standard kinetics governs the evolution of the Ga adatom diffusion length with both temperature and Sb flux. The evolution observed between samples A (Fig. 1(a)) and B (Fig. 1(b)) enters this domain. Above the

line, stable Sb-Sb bonds impede Ga incorporation and lead to an increase in the Ga diffusion length in agreement with the evolutions observed between samples E, D, and B (Figs. 3(a)–3(c)) and between samples B (Fig. 1(b)) and C

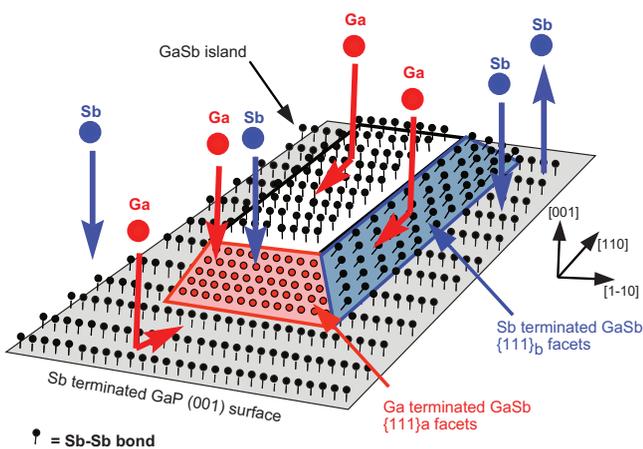


FIG. 4. Schematic illustration of the Ga adatom behavior and growth mechanism of {111}-faceted islands at high Sb flux leading to [110] elongated islands.

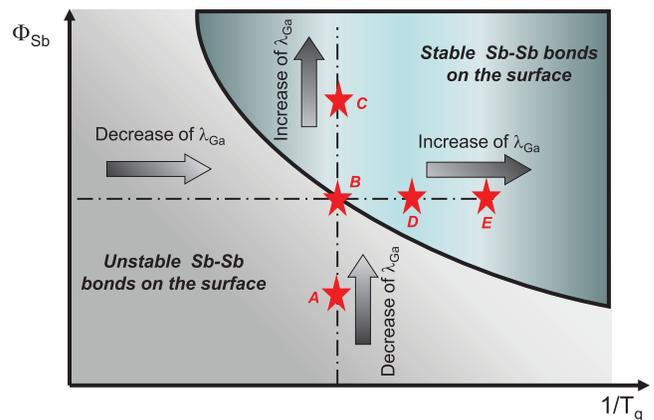


FIG. 5. Qualitative description of the evolution of Ga adatom diffusion length versus growth conditions. The black line separates two domains (stable Sb-Sb bonds and unstable Sb-Sb bonds) where the influence of the growth condition is reversed. The positions in the two domains corresponding to the growth conditions used for the different samples (A, B, C, D, and E) are indicated by red stars.

(Fig. 1(c)). This latter effect is directly related to the relative strength of Sb-Sb and Ga-Sb bonds. For arsenides and phosphides, even if stable V-V bonds are formed on the surface, they do not much affect the group-III element diffusion length since As-As and P-P bonds are significantly weaker than Ga-As and Ga-P ones.

This interpretation can be extended to the GaSb/GaAs system and accounts for most of the previous observations.<sup>12,13,22</sup> The different mismatches and substrate anion atoms, which are involved in the two systems, may however induce shifts in the range of growth temperature and Sb overpressure since these parameters can affect the thermodynamical equilibrium of the interface.

## CONCLUSION

We have investigated the influence of the Sb flux and the growth temperature on the formation of relaxed GaSb islands on GaP substrates. Within the range of low Sb fluxes or high growth temperature, standard kinetics governs the formation of GaSb islands. At very low Sb flux, part of the strain in the islands is plastically relaxed by the misfit dislocations whereas the high density of stacking faults should accommodate the residual strain. By increasing the Sb flux, the {111} facets begin to appear and GaSb islands are relaxed only via a 90° MDs interfacial network. The largest Sb coverage of the Sb-terminated GaP surface leads to the formation of a low density of relaxed {111}-faceted islands elongated in the [110] direction, driven by the larger incorporation of Ga on {111}<sub>a</sub> facets at very high Sb fluxes. This behavior is qualitatively explained considering the interplay between the growth temperature and the Sb flux on the Ga adatom diffusion length. The proposed interpretation is consistent with previous observations reported on the GaSb/GaAs system.

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