Mechanism of organization of three-dimensional islands in SiGe/Si multilayers

E. Mateeva[a] and P. Sutter
University of Wisconsin—Madison, Madison, Wisconsin 53706

J. C. Bean
University of Virginia, Charlottesville, Virginia 22903

M. G. Lagally[b]
University of Wisconsin—Madison, Madison, Wisconsin 53706

(Received 31 July 1997; accepted for publication 3 October 1997)

The organization of coherent three-dimensional islands during the growth of SiGe/Si multilayers on Si(100) was investigated with cross-sectional transmission electron microscopy. Merging of islands of different initial size is found to be the dominant mechanism leading to a uniform size distribution. Upon overgrowth with Si, we observe a change of the shape of the islands from the {105}-faceted ''hut'' to a boxlike shape bounded on top by a (100) facet. © *1997 American Institute of Physics.* [S0003-6951(97)02548-5]

Self-organized formation of quantum structures in the SiGe/Si system has attracted considerable interest, as this system is expected to play an important role in the monolithic integration of Si-based micro- and optoelectronic quantum devices SiGe/Si is a Stranski–Krastanov system in which heteroepitaxial growth starts with the formation of a two-dimensional wetting layer followed by the nucleation of small three-dimensional (3D) islands. These coherently strained islands evolve into large 3D islands, in which strain relaxation occurs via introduction of dislocations. The small 3D islands, the so-called ''hut clusters''[1] with shallow {105} facets, are of interest, as they are expected to exhibit the carrier confinement properties of quantum dots. Although these 3D islands have a well-defined shape, their random nucleation in a single SiGe alloy layer results in a broad distribution of island sizes. Because arrays of islands of the same size and shape are required for practical applications, the identification of growth mechanisms that lead to a narrowing of the size distribution is of great importance.

Lateral and vertical ordering of 3D islands in multilayers has been studied in both the SiGe/Si[2–4] and the InAs/GaAs[5,6] systems and was probably present even in some of the first superlattices grown by molecular beam epitaxy (MBE).[7] For the SiGe/Si(100) system, it has been shown by atomic force microscopy (AFM) that coherent 3D islands undergo a transition from a broad size distribution in a single SiGe layer to intraplanar size equalization in a SiGe/Si multilayer structure.[2] This transition has been referred to as ''self-organized'' growth. On the basis of the AFM investigation of the surface morphology, a model was proposed to account for the vertical correlation of the islands in successive layers.[8] Using elastic continuum theory, this model establishes that the inhomogeneous strain field in Si due to a buried SiGe island can cause the self-organization. Here we provide a direct observation by cross-sectional transmission electron microscopy (XTEM) of the vertically correlated arrangement of coherent islands in $Si_{1-x}Ge_x$/Si multilayers, and of their evolution to a uniform size distribution. Moreover, we observe a change of shape of the islands from a {105} faceted pyramid to a rounded, boxlike shape with a [100] top facet upon overgrowth with Si.

The $Si_{1-x}Ge_x$/Si multilayers used in this study were grown by MBE on vicinal Si(100) substrates at a temperature of 550 °C and growth rate of 0.055 nm/s. Deposition of a 100 nm Si buffer layer was followed by the growth of up to 20 bilayers, in which SiGe alloy layers with Ge concentration $x=0.75$ and nominal thickness 2.5 nm were separated by 10-nm-thick Si spacer layers. The structures were terminated by a SiGe alloy layer. The AFM study of the surface after the first $Si_{0.25}Ge_{0.75}$ layer shows that the islands formed have mostly rectangular bases and are bounded by {105} facets.[2] The structure of the multilayers was investigated by XTEM using a Philips CM200 microscope at 200 kV. TEM samples were prepared by tripod polishing followed by brief ion milling.

Figure 1 shows a representative [110] cross-section bright-field micrograph of a 20 bilayer $Si_{0.25}Ge_{0.75}$/Si multilayer film. Regions of light contrast correspond to the Si spacer layers, while contrast in the dark regions stems from the islanded SiGe alloy and the thin wetting layer. The figure shows that the Si spacer layer is thick enough to flatten the growth front prior to deposition of the subsequent alloy layer. SiGe islands in the lowest alloy layers are small, with average base size[9] between 50 and 70 nm, and become larger and less numerous with an increasing number of bilayers. Most importantly, Fig. 1 shows that islands in subsequent alloy layers are not simply located on top of each other: their behavior is more complicated. In the first 10–15 bilayers, small islands move towards each other, preserving their initial size. Upon reaching a certain critical value of island spacing, only one island of larger size nucleates on top of each pair. These larger islands then show direct vertical correlation, i.e., nucleate directly on top of each other. The second important observation in Fig. 1 concerns the shape of the 3D islands. Buried islands are rounded, boxlike mounds and are bounded on top by a large (100) facet. Since the un-

[a]Corresponding author: Electronic mail: mateeva@mrgcvd.engr.wisc.edu; fax: (608) 265-4118.
[b]Electronic mail: lagally@engr.wisc.edu

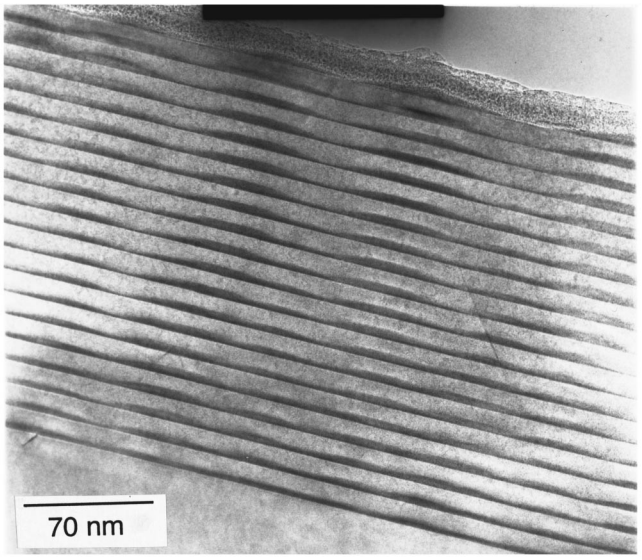

FIG. 1. Representative [110] cross-section bright-field micrograph of an uncapped 20 bilayer $Si_{0.25}Ge_{0.75}$/Si multilayer structure showing the vertical ordering of the SiGe islands. The thickness of the Si spacer layer is 10 nm.

capped islands in the topmost alloy layer are seen to be pyramids with well-defined {105} facets, these hut clusters apparently undergo a significant change of shape upon overgrowth with Si.

Figure 2 shows in more detail the merging of islands, which we find to be the dominant mechanism leading to a uniform size distribution in our SiGe/Si multilayers. Figure 2(b) is a schematic representation of the change of size and spacing of two islands in the part of a multilayer shown in the micrograph of Fig. 2(a). The two islands have a similar initial base size of about 60 nm. With increasing bilayer number, they nucleate closer to each other with almost unchanged island size. When the island spacing[10] becomes smaller than 40 nm, the islands merge to form a single island, which almost covers the two underlying ones (base size 100 nm). Once this large island is formed, no further change in island position is observed, and islands in consecutive layers nucleate exactly on top of each other. Moreover, their size assumes a stable value of 90 nm, slightly less than the value of the initial merged island.

The one-to-one vertical correlation of islands of stable size is also seen in Fig. 1, as well as in the high-resolution micrograph shown in Fig. 3. This micrograph depicts the topmost 5 bilayers of the uncapped SiGe/Si multilayer sample, and allows a direct comparison of the shape of overgrown and uncapped islands. The uncapped island is bounded by {105} facets. The buried islands are more rounded, and have a large flat (100) terrace on top. The regions of dark contrast are also larger for the buried islands, indicating an increase in island volume upon overgrowth.

The XTEM micrographs show that the originally nucleated islands of different size interact with one another. This interaction gives rise to a uniform size distribution involving a typical length scale of the base sizes of the order of 90 nm (for this alloy composition and these layer thicknesses). After this larger value is reached, we observe a pronounced stabilization of the island size. The observed self-organization can be understood qualitatively in the frame-

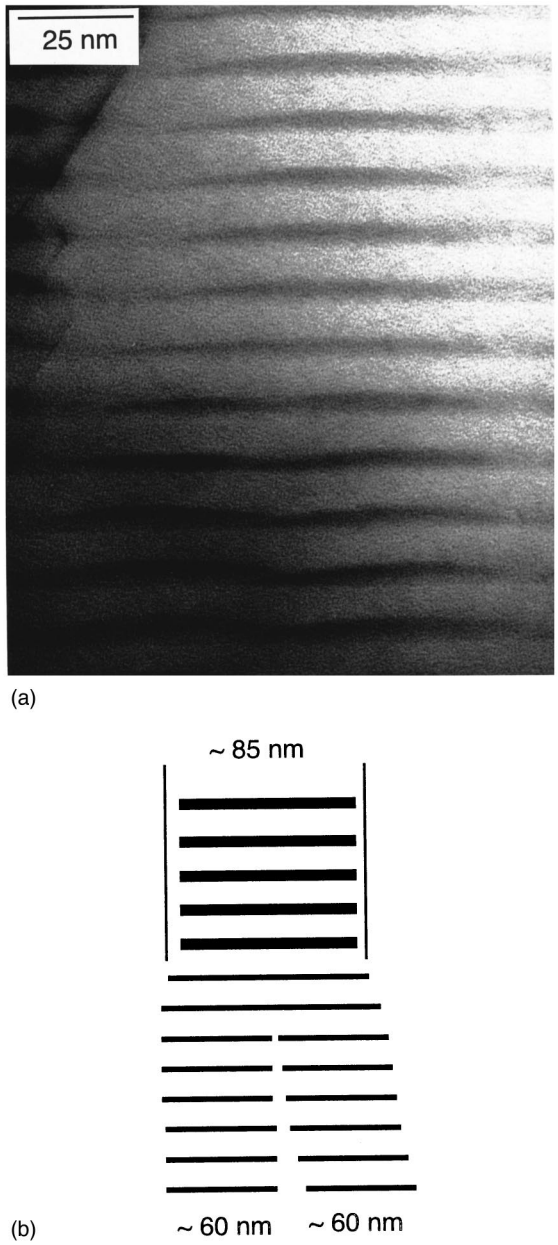

FIG. 2. Island ''coalescence'' in a $Si_{0.25}Ge_{0.75}$/Si multilayer. (a) Bright-field XTEM micrograph showing a part of a $Si_{0.25}Ge_{0.75}$/Si multilayer and (b) a schematic presentation of the change of size and spacing of two islands in the part of the multilayer shown in (a).

work of a model based on continuum elasticity theory. In this model, the islands are treated as spherical inclusions in an isotropic medium.[8,11] Formation of an island on the Si surface is determined by the strain from the buried islands, with nucleation occurring where this strain gives a local minimum of the misfit between the Si spacer layer surface and the new SiGe island. According to this model, the diameter of the surface region under tensile strain, i.e., with enhanced nucleation rate for 3D islands, is given by $\sqrt{2}L$, where L is the distance between the center of the buried island and the top of the Si spacer layer. It is thus independent of the size of the island (radius r_0 in the case of a spherical inclusion).

For a small SiGe island buried under 10 nm Si, the surface region under tensile strain expands beyond the lateral size of the island. For two such islands, the superposition of

FIG. 3. High resolution lattice image of the topmost 5 bilayers of an uncapped $Si_{0.25}Ge_{0.75}$/Si multilayer. The topmost island is bounded with {105} facets. The islands overgrown with Si show more rounded shapes topped with (100) facets.

the strain fields leads to a closer nucleation of the islands in the consecutive layers. Finally, when the spacing between them becomes small enough, the lateral superposition of the strain fields is such that the nucleation of one island on top of the two smaller ones is most probable. The two islands merge and are replaced by an island of larger size. For such a larger island (i.e., island diameter $2r_0 > \sqrt{2}L$), the in-plane tensile strain of the spacer layer surface vanishes within the lateral extent of the island. Thus only one island nucleates and the favored nucleation position is directly above the previous one. This preferred nucleation accounts for the observed stabilization of the island size, as well as for the direct vertical correlation of the islands of stable size.

The novel observation that the SiGe islands undergo a significant change of shape upon overgrowth with Si is not fully understood. The observed larger regions of dark contrast of the buried islands indicate that they are of greater volume than the ones at the growth front, which implies that significant intermixing with Si occurs during overgrowth of the SiGe islands. While intermixing would explain an increase in volume, it cannot account alone for the formation of a large (100) facet bounding the top of the buried SiGe islands. The flattening of the pyramids involves the reduction during overgrowth with Si of the effective volume of the SiGe islands, which destabilizes the tops of the pyramids and eventually favors their transformation into a 2D layer. This mechanism will be discussed in detail elsewhere.

The described mechanism of island merging should lead to self-organization, independent of the detailed shape of the buried islands. We expect, however, that their shape, together with the spacer layer thickness L and the alloy concentration, influences the length scales involved, i.e., the base size of the islands of stable size. Additionally, both the intermixing with Si and the change of shape of the buried SiGe islands will significantly influence the electronic properties of these nanostructures and have to be taken into account, e.g., in the interpretation of photoluminescence spectra.

In summary, we have shown experimental evidence for the vertically self-organized growth of strained SiGe islands in multilayer SiGe/Si films grown on Si(100). We have shown that the main mechanism leading to size uniformity is the merging of small neighboring islands. This merging leads to the controlled formation of much larger islands, which essentially do not interact, thus stabilize in size and spacing, and nucleate directly on top of each other. We also observe a change of the shape of the islands from the {105}-faceted hut cluster to a rounded, boxlike island bounded on top by a large (100) facet upon overgrowth with Si.

This work was supported by Office of Naval Research and by the Swiss National Science Foundation (P.S.).

[1] Y.-W. Mo, D. E. Savage, B. S. Schwartzentruber, and M. G. Lagally, Phys. Rev. Lett. **65**, 1020 (1990).
[2] C. Teichert, M. G. Lagally, L. J. Peticolas, J. C. Bean, and J. Tersoff, Phys. Rev. B **53**, 16 334 (1996).
[3] B. Rahmati, W. Jaeger, H. Trinkaus, R. Loo, L. Vescan, and H. Lueth, Appl. Phys. A: Solids Surf. **62**, 575 (1996).
[4] A. A. Darhuber, P. Schittenhelm, V. Holy, J. Stangl, G. Bauer, and G. Abstreiter, Phys. Rev. B **55**, 15 652 (1997).
[5] Q. Xie, A. Madhukar, P. Chen, and N. Kobayashi, Phys. Rev. Lett. **75**, 2542 (1995).
[6] A. A. Darhuber, V. Holy, J. Stangl, G. Bauer, A. Krost, F. Heinrichsdorff, M. Grundmann, D. Bimberg, V. M. Ustinov, P. S. Kop'ev, A. O. Kosogov, and P. Werner, Appl. Phys. Lett. **70**, 955 (1997).
[7] R. Hull, J. C. Bean, F. Cerdeira, A. T. Fiory, and J. M. Gibson, Appl. Phys. Lett. **48**, 56 (1986).
[8] J. Tersoff, C. Teichert, and M. G. Lagally, Phys. Rev. Lett. **76**, 1675 (1996).
[9] Because the XTEM images are [110] cuts through the islands, the base size corresponds to the diagonal of the ⟨100⟩ aligned base of the pyramid-shaped huts.
[10] The island spacing is considered to be the distance between the center of two islands.
[11] We follow this simplified approach in spite of our observation that the shape of the overgrown islands is boxlike and not well approximated by a single sphere/point source of expansion. But, nevertheless, the physics of the island ''interaction'' can be understood using this simple model.