APPLIED PHYSICS LETTERS **89**, 253105 (2006)

Evolution of buried semiconductor nanostructures and origin of stepped surface mounds during capping

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(Received 22 August 2006; accepted 9 November 2006; published online 18 December 2006)

The authors apply selective wet chemical etching and atomic force microscopy to reveal the three-dimensional shape of SiGe/Si(001) islands after capping with Si. Although the "self-assembled quantum dots" remain practically unaffected by capping in the temperature range of 300–450 °C, significant morphological changes take place on the Si surface. At 450 °C, the morphology of the capping layer (Si matrix) evolves toward an intriguing semifacetted structure, which we call a "ziggurat," giving the misleading impression of a stepped SiGe island shape. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2405876]

Semiconductor quantum dot (QD) structures have attracted intense interest as promising candidates for future nanoscale devices.[1–4] For most applications, these nanostructures must be capped, i.e., epitaxially embedded in a host matrix. However, during the capping process, dramatic morphological and compositional changes may occur,[5–9] altering the electronic properties of the QDs. Therefore, an understanding of the capping process is of fundamental importance. Understanding the surface morphology of the cap layer is also important, because many applications require further overgrowth steps.

Various techniques have proven useful to determine the morphology of nanostructures after capping, but all have their limitations, and a complete picture of the evolution is still lacking. Cross sectional transmission electron microscopy (TEM) and scanning tunneling microscopy (STM) provide information only along a randomly selected two-dimensional plane. X-ray scattering measurements[8] average over a statistical ensemble of islands. Selective wet chemical etching techniques[10–12] have not yet been able to give much information about the morphological changes of islands upon capping. The primary source of information on the capping process itself has been atomic force microscopy (AFM) and STM imaging of the surface at successive stages of capping,[7,9,13–15] but these techniques do not distinguish between island and capping materials.

Here we focus on the paradigm system of Ge on Si(001), using the resulting SiGe islands as self-assembled quantum dots. We use a combination of selective wet chemical etching and AFM to extract quantitative information about the evolution of the morphology during the capping process. We find that the surface morphology can be completely different from that of the buried islands. Thus measurements of surface morphology alone,[7,9,13–15] even for quite thin capping layers, do not provide a reliable measure of the evolution of the buried islands and can even lead to serious misunderstandings regarding their structure.

The samples were grown by solid source molecular beam epitaxy (MBE). After deoxidation and Si buffer growth, 5.9 ML (monolayer) of Ge were deposited at a rate of 0.04 ML/s while the substrate temperature was kept at 580 °C. This results in the spontaneous formation of strained three-dimensional (3D) islands. Subsequently, 20 nm of Si were deposited at a rate of 0.7 ML/s at 300, 450, and 580 °C. Some samples were etched in a $2M$ potassium hydroxide (KOH) solution at room temperature (RT). The morphology of islands and Si surfaces after capping was investigated by AFM in tapping mode and by an ultrahigh vacuum STM operating at RT.

Figure 1 displays AFM images of the as-grown islands before capping (reference sample) and after 20 nm Si capping at different temperatures (300, 450, and 580 °C). The reference sample [Fig. 1(a)] exhibits a distribution of elongated hut clusters, square pyramids, and dome islands. For Si capping at 300 °C [Fig. 1(b)], surface diffusion is largely suppressed. The resulting Si surface closely resembles the original islands, especially on the larger scale of dome islands.[13,16]

In contrast, after Si capping at 450 °C [Fig. 1(c)] the surface consists of truncated pyramids with edges aligned along the ⟨110⟩ directions. They are rotated by 45° as compared with the edges of Ge pyramids on the reference sample. Such structures are actually seen for a wide range of temperatures (450–550 °C) (Ref. 13) and for different growth techniques, such as MBE,[13] chemical vapor deposition,[17] and magnetron sputtering epitaxy (MSE).[7,14] Due to their similarity to the terraced pyramidal temples of ancient Mesopotamia (as will be seen clearly later), we call them for simplicity *ziggurats*. Finally, Si capping at 580 °C [Fig. 1(d)] restores a flat surface.

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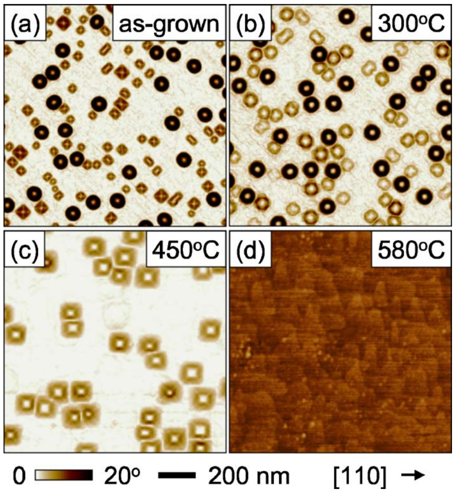

FIG. 1. (Color online) AFM images taken after deposition of 5.9 ML Ge at (a) 580 °C and after subsequent 20 nm Si capping at (b) 300 °C, (c) 450 °C, and (d) 580 °C. The color scale in (a)–(c) represents the local surface slope with respect to the (001) plane, while in (d) the gray scale corresponds to the local surface height.

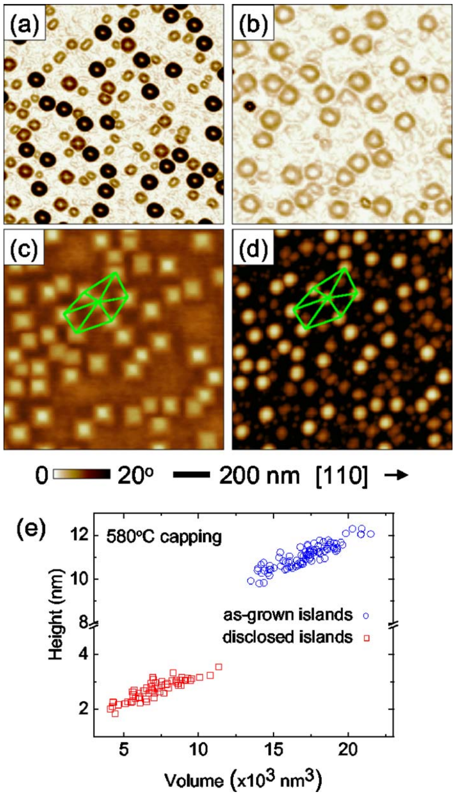

FIG. 2. (Color online) AFM images displaying the morphology of disclosed islands overgrown at (a) 450 °C and (b) 580 °C. (c) and (d) are AFM images showing the same area of the sample capped at 450 °C, before and after removing the Si layers, respectively. There is a clear one-to-one correlation between the ziggurats and the buried dome islands, as illustrated by the polygon in (c) and (d). The color scale in (a) and (b) is related to the local surface slope, while in (c) and (d) it is related to the local surface height. (e) Statistical analysis of the dome islands contained in the reference and the disclosed sample which was overgrown at 580 °C: height vs volume is plotted. A clear reduction of both height and volume of the buried islands can be observed.

To reveal the 3D shape of the buried islands, we etched the overgrown samples in a 2*M* KOH solution for ~2 min. This removes Si with a selectivity of ~100:1 over $Si_{0.8}Ge_{0.2}$.[18] At the growth temperature used in this study, the wetting layer (WL) is Ge rich. Thus, as soon as the WL is reached, the etch rate slows down significantly. Stopping the etching at this point gives a direct 3D view of the *disclosed islands*.

By removing the Si layers from the sample capped at 300 °C, it is seen that the morphology of the islands shows virtually no difference with respect to the as-grown ones (image not shown).[18] Surprisingly, the same is also true for the islands which were overgrown at 450 °C [Fig. 2(a)]. Although after capping the surface looks completely different [Fig. 2(c)], it appears that the buried islands do not experience significant morphological changes. This simple experiment shows that the ziggurats observed after capping do *not* reflect morphological changes of the buried islands.

The dome island density on the reference sample and that of the ziggurats on the capped sample are similar, suggesting a possible correlation. In order to check this point, we analyze the *same surface location* prior to and after Si etching. Figures 2(c) and 2(d) respectively show the same region before and after the Si layer has been removed. While the surface above the hut clusters and pyramids is flat, a one-to-one correlation between the position of the buried dome islands and the ziggurats is evident. This is also confirmed by TEM measurements (not shown here).

In contrast to the capping at 300 and at 450 °C, overgrowing the islands at 580 °C leads to significant morphological changes [Fig. 2(b)]. The hut clusters and pyramids can no longer be clearly resolved. It may be that these islands have partially dissolved and their material has moved to the WL, since a rough surface is observed between the dome islands. The dome islands become shallower and their base area increases, in accordance to what has been reported previously.[6–8] A detailed statistical analysis also reveals that the island volume decreases [Fig. 2(e)]. (The volume refers to the region having Ge content larger than 20%.) Indeed the volume of the buried islands is reduced by more than half compared to that of the as-grown islands. Some of the Ge may redistribute across the surface or dissolve into the wetting layer (as observed in real time by Lang *et al.*[19] and for InAs islands[20]). Ge present as a very dilute alloy is not counted in our analysis, nor does it count as part of the QD in electronic devices.

The analysis of the disclosed islands demonstrates that, at least for quite thin Si capping thicknesses, no conclusions about the *shape* of the buried islands should be drawn by studying the Si surface covering them.[7,13,15] The same is true of the island *size*. Surface measurements suggested an increase in size,[7] while etching shows a decrease.

Finally, we analyze in more detail the ziggurat structures seen in Fig. 2(c); a high resolution 3D STM image of such a ziggurat taken from a sample grown by MSE under similar conditions is shown in Fig. 3(a). Understanding their formation is of particular importance since they can influence subsequent overgrowth processes. Moreover, because of their stepped appearance, they might be easily mistaken for stepped islands,[7,21] which have been studied extensively. (We use the term island just for 3D SiGe morphologies, not for 3D structures consisting of pure Si.)

Because the islands are virtually unaffected by capping up to 450 °C, there must be very little intermixing, so the capping layer consists of nearly pure Si. Then, thermodynamically, we would actually expect a *dip* in the surface above a buried island, because of the unfavorable strain

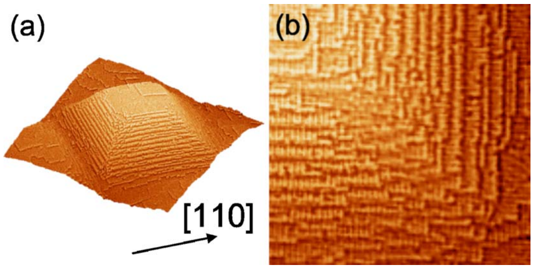

FIG. 3. (Color online) (a) 3D STM image of a ziggurat structure ($160 \times 160 \times 5.1$ nm^3). (b) 40×40 nm^2 2D STM image which reveals the *B*-type double (DB) steps aligned along the [110] directions.

there.[22,23] We therefore believe that the ziggurats reflect the kinetically limited smoothing of the Si cap-layer surface. Apparently, at 450 °C the Si surface kinetics is too slow for complete smoothing or formation of a dip.

The kinetics of smoothing of stepped surfaces is an extremely complex subject,[24] but consideration of the energetics driving the smoothing is sufficient to explain the qualitative features here. Steps on Si (001) have been studied extensively. At very small angles from (001), the surface consists of single-layer steps, or a mixture of single- and double-layer steps.[25] But the sidewalls of our ziggurats are generally 3°–6.5° from (001). In this range, double-layer steps of the [110] oriented *B*-type (DB steps) are energetically favorable.[25] This drives facetting with respect to azimuth (i.e., straight [110] oriented steps), while the slope of the island sidewall (i.e., the step spacing) can vary continuously. For this reason we refer to the ziggurat structure as semifacetted.

At 300 °C there is little Si mobility, and capping is nearly conformal. But at 450 °C, the morphology can start to evolve toward lower energy. We note that 450 °C is far below the roughening temperature for Si, so the surface should be composed of steps on Si (001). [In principle, other facets such as {113} could form as reported in Ref. 13; but apparently the angles here are too shallow to support any other facet orientation besides (001).] This is directly confirmed by the high resolution STM shown in Fig. 3(b). Any deviation from [110] orientation requires having single-layer step pairs, or worse, a mixture of *A*-type double steps with the DB steps. Therefore there is a very strong driving force for formation of four sides having strict [110] azimuth, and the morphology evolves rapidly to the semifacetted structure. On the other hand, since the DB steps have rather low energy,[25] the driving force for lateral spreading of the ziggurats is relatively weak by comparison, so the lateral spreading is a much slower process. Finally, at 580 °C the Si mobility is much higher, and the buried islands are reduced in height, giving a flat cap surface.

In conclusion, we have revealed the 3D shape of buried SiGe islands. Our results show that the Si surface morphology can be radically different from the morphology of the buried islands. In particular, for capping at 450 °C we observe ziggurat structures which do not correspond to shape changes of buried islands. Instead, these surface structures represent the kinetically limited smoothing of the Si cap-layer surface.

The authors acknowledge A. M. Bittner and S. Kiravittaya for fruitful discussions.

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