Oblique stacking of three-dimensional dome islands in Ge/Si multilayers

P. Sutter[a)] and E. Mateeva-Sutter
Department of Physics, Colorado School of Mines, Golden, Colorado 80401

L. Vescan
Institut für Schicht und Ionentechnik, Forschungszentrum Jülich GmbH, D-52425 Jülich, Germany

(Received 2 October 2000; accepted for publication 26 January 2001)

The organization of Ge ''dome'' islands in Ge/Si multilayers has been investigated by cross-sectional transmission electron microscopy. Ge domes are found to spontaneously arrange in oblique stacks, replicating at a well-defined angle from one bilayer to the next. The formation of oblique island stacks is governed by a complex interplay of surface strain, generated by the already buried islands, and surface curvature, caused by the inherent tendency of large domes to carve out material from the surrounding planar substrate. © *2001 American Institute of Physics.*
[DOI: 10.1063/1.1357214]

Semiconductor quantum dots (QDs)—nanostructures in which charge carriers are confined in three spatial dimensions—promise to become the building blocks of future electronic and optoelectronic devices with enhanced functionality, and have been proposed recently for the implementation of solid state quantum computing.[1] Most of the proposed technological applications require ordered ensembles of densely packed uniformly spaced QDs, which should be identical in size and shape. Meeting these requirements remains a formidable challenge for all of the techniques used to fabricate QDs, which range from high-resolution lithographic patterning[2] to colloidal chemistry[3] and heteroepitaxial growth.[4] Particularly interesting among these techniques are those in which quantum dot arrays are formed through nanoscale self-assembly processes, i.e., essentially fabricate themselves. In strained-layer heteroepitaxy, for instance, common pathways of strain relaxation involve the spontaneous self-assembly of small, faceted three-dimensional (3D) islands at the growth front, which can be converted into QDs by epitaxial embedding in a matrix material. In a prototype system used to study the formation of epitaxial dots, Ge/Si(100), lattice mismatch strain is initially relaxed via the formation of small pyramidal ''hut'' islands bounded by shallow {105} facets.[5] Growing beyond a critical size, these huts transform into ''dome''-shaped islands with additional higher angle facets that allow more complete strain relaxation.[6]

Growth of single layers of epitaxial 3D islands typically produces island ensembles that are disordered and show significant fluctuations in island size. Multilayer films, consisting of layers of 3D islands separated by thin spacers of the substrate material, were found to give improved control over both the spatial ordering and island size distribution.[4,7] In SiGe/Si multilayers containing hut islands we have identified an organization mechanism that is based on the inhomogeneous surface strain field due to buried islands steering the formation of the next layer of islands at the growth front.[8] The strain field causes small neighboring huts to grow closer in subsequent layers, such that they eventually merge and are replaced by a single larger hut island. The surface strain field of these larger islands, in turn, is of much shorter range relative to the island size. Instead of continuing to merge, the larger huts thus replicate, with constant size, vertically from one layer to the next.

In this letter, we discuss the ordering of dome islands in multilayer structures. Based on the results of Ref. 8, one would expect these large islands to organize in straight stacks along the growth direction. We find, however, that the transition from the small {105}-faceted huts to the larger multifaceted domes causes surprising qualitative changes in the mechanisms of island organization. Strikingly, we observe the formation of *oblique stacks* of domes in these multilayers. We demonstrate that the oblique stacking, which introduces the island stacking angle as a new variable for modifying the interaction of electronically coupled dots, is caused by a complex interplay of surface strain (as in hut multilayers) and surface curvature, with the latter not being introduced deliberately, but caused by the inherent tendency of large domes to carve out material from the surrounding planar substrate.

The films used in this study consist of alternating Ge and Si layers grown on Si(100) substrates by low pressure chemical vapor deposition. Following *ex situ* (RCA) and *in situ* (at 950 °C in H_2) substrate cleaning, a 350 nm thick Si buffer layer was deposited to ensure a planar starting surface for the subsequent heteroepitaxy. Ge was grown at a rate of 0.043 nm/s, substrate temperature of 700 °C, and total pressure of 0.12 Torr to a nominal thickness of 1.2 nm, well beyond the transition to the formation of stress-induced 3D islands, and in fact well beyond the transition in island shape from pyramidal huts to multifaceted domes: atomic force microscopy of uncapped samples shows that more than 95% of the Ge islands are multifaceted domes. The islanded Ge layers are separated by 40 nm thick Si spacers to form multilayer structures with 20 Ge/Si bilayers. The arrangement of 3D islands in these multilayer samples was studied by cross-sectional transmission electron microscopy (XTEM) using a Philips CM200 microscope at 200 kV. XTEM samples were prepared by tripod polishing to electron transparency, followed by brief ion milling.

[a)]Author to whom correspondence should be addressed; electronic mail: psutter@mines.edu

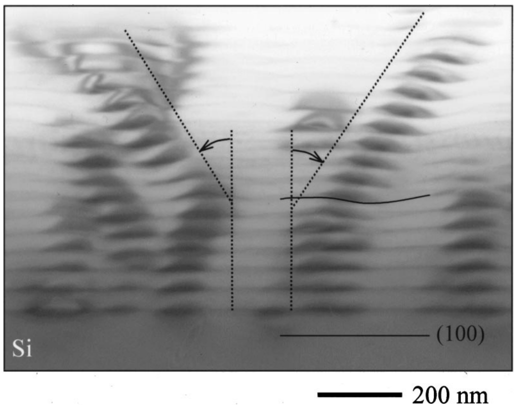

FIG. 1. Representative [110] cross-sectional bright-field TEM micrograph of a 20 bilayer Ge/Si multilayer film, showing the arrangement of Ge islands.

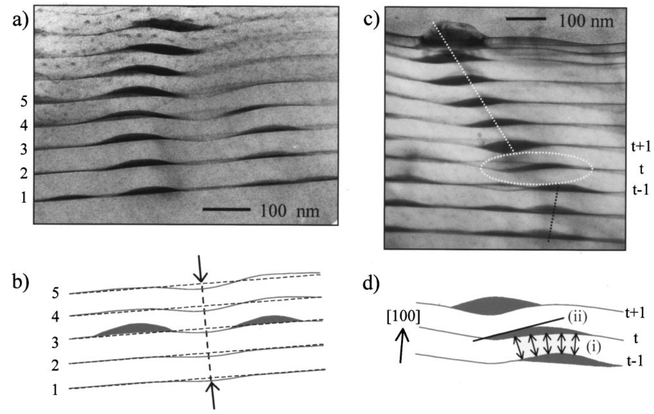

FIG. 2. Cross-sectional TEM images (a), (c) and schematic drawings (b), (d) illustrating the different stages of the formation of oblique island stacks in multilayers with alternating Ge domes and Si spacers. (a), (b) Initial formation of a depression between large Ge dome islands. (c), (d) Transition to oblique stacking.

Figure 1 shows a representative [110] cross-sectional bright-field TEM micrograph of a 20 bilayer Ge/Si multilayer film. Dark contrast stems from the Ge dome islands and from the thin Ge wetting layer. Regions with light contrast correspond to the Si spacer layers. The image shows a strong correlation between the positions of islands in subsequent Ge layers. In the lower part of the multilayer (the first four to six bilayers) the Ge dome islands align roughly vertically, similar to large hut islands in multilayers.[8] After these initial layers, the spatial arrangement changes suddenly and dramatically: instead of replicating vertically, subsequent islands form with significant in-plane displacements, generating oblique island stacks in the multilayer. The stacking angles are about the same in all oblique stacks we observed in this sample, and remain constant throughout the multilayer (as indicated with arrows in Fig. 1). While the islands are symmetric in the lowest bilayers, the island shapes become asymmetric with the onset of oblique stacking. In contrast to hut multilayers the surface of the Si spacers shows significant undulations, which start in the lowest bilayers and whose amplitude is largest in the proximity of large domes. Depressions next to large islands become progressively deeper in consecutive layers and seem to induce the observed in-plane shifts and control the formation of oblique island stacks. The simplest explanation for the observed surface undulations would be that the Si spacers were not thick enough to flatten the growth front and eliminate the height differences due to the underlying islands.[9] Our data, however, show a different and intriguing mechanism for the formation of surface undulations and of the resulting oblique island stacks, which we will discuss in detail below.

Figure 2 illustrates the different stages of the formation of oblique island stacks. The first stage of this process, the initial formation of depressions in the vicinity of large Ge dome islands, is shown in detail in the XTEM image and the drawing of Figs. 2(a) and 2(b), respectively. Consider the surface of the Si substrate on which the first layer of Ge islands is grown. Our XTEM observations generally show a planar substrate surface, and indicate that the Si substrate was indeed flat prior to growth of the first layer of Ge islands. After deposition of the first Ge layer, the substrate surface remained flat everywhere, except in the neighborhood of larger domes. In these areas we consistently observe the formation of depressions where substrate material was obviously carved out below the original surface profile. Such depressions are particularly deep in regions between two or more large dome islands. We have shown previously that the shape transition from pyramidal huts to multifaceted dome islands can induce significant mass transfer, causing the spontaneous removal of material surrounding the island and the incorporation of this material into the dome.[10] The microstructure shown in Figs. 2(a) and 2(b), showing depressions in the substrate surface near large dome islands, is consistent with such spontaneous self-embedding of the domes with material from the surroundings. We observe depressions in the substrate surface that are about 5 nm deep. In the bilayers that follow the depressions replicate and become progressively deeper. Their depth roughly increases by a constant increment from one bilayer to the next, indicating that the spacer does not recover a planar surface, and that the next dome island again carves out material. Note that, at this stage, consecutive islands form roughly on top of each other: the positions of the domes and of the neighboring depressions do not shift laterally.

The second stage in the process, the abrupt transition to oblique stacking, is illustrated by the XTEM image and the schematic drawing in Figs. 2(c) and 2(d), respectively. A sudden transition occurs in the fourth bilayer in this particular case, or in bilayer number t in general. The depressions between large domes are now ~40 nm deep, at which point the islands significantly reduce their thickness and spread out laterally. Figure 2(d) (i) illustrates the roughly constant spacer thickness over the entire underlying island in layer $t-1$. The buried island should thus generate constant tensile surface strain over an extended area, rather than be sharply peaked near the apex as in the first layers, and promote the formation of a t-th Ge ''island'' of roughly constant thickness over this entire area. In addition, extending into the adjacent depression, this island can form a large steep facet (ii).

In the bilayers that follow the islands continue to shift toward the depressions, and induce the formation of oblique island stacks. This final stage of the process is shown in detail in Fig. 3. The topmost Ge islands in this image are not

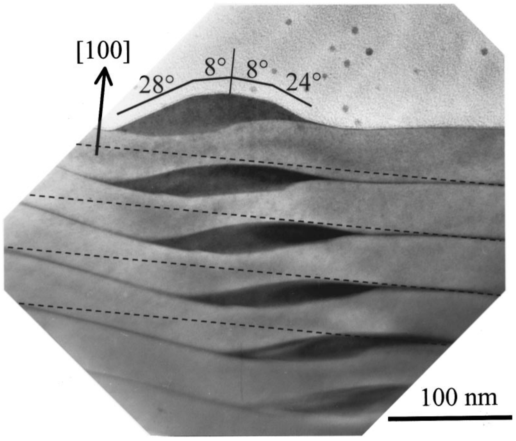

FIG. 3. Cross-sectional TEM image of the topmost six bilayers of an uncapped Ge/Si multilayer. The topmost island is clearly asymmetric, and bounded by several distinct facets: a symmetric pair of shallow (105) facets near the apex (8° facet angle in this [110] projection), and steeper (24°, 28°) facets near the base. The steepest facet is also largest, and extends far into the adjacent depression.

capped with Si, which allows us to determine details of their shape.[11] The islands are clearly faceted. We observe three groups of distinct facets in this [110] projection. The island apex is bounded by a set of symmetric facets with an angle of ~8° relative to [100], consistent with the angle of (105) facets in this perspective.[8] The periphery is bounded by steeper facets, with projected angles of 28° (toward the depression, left) and 24° (away from the depression, right). In contrast to the dome islands observed on planar substrates, our obliquely stacked domes are thus *asymmetric* and bounded by a very long and steep facet toward the neighboring depression.

Our data indicate that nonplanar spacer surfaces with deep depressions promote the asymmetric shape of the Ge islands and that this asymmetry, in particular the existence of a long, steep bounding facet, in turn governs their oblique stacking. According to Fig. 3, the apex of each island in the oblique stack is shifted to the right from the island's center of mass, i.e., away from the depression. On the other hand, the thinnest regions of the spacer layers clearly shift to the left, i.e., toward the depression and the long, steep facet of the underlying island. Theory predicts that steep facets strongly promote geometric strain relaxation in 3D islands.[12] We would thus expect the Ge islands to be most relaxed near the steep facets, and growth of the Si spacers, in turn, to be slowest in areas where the Ge island is most relaxed. As a consequence, the deepest point of the depression should shift to the left from one bilayer to the next, which is indeed observed in Fig. 3. It is well established that new 3D islands in Ge/Si multilayers form preferentially at positions of the spacer layer, where the magnitude of the tensile surface strain due to the underlying islands is largest.[7] In the present case, we would expect this position, and thus the center of mass of the new island, to coincide with the point of minimum spacer thickness. The shifting of the point of minimum spacer thickness, caused by the asymmetric shape of the domes, therefore induces the formation of an oblique island stack.

The striking oblique stacking of dome islands can organize self-assembled quantum dots into unique arrangements. We have observed the formation of branched columns of dots, caused either by a single large dome island producing a pair of distinct oblique stacks, or by the coalescence and merging of two stacks that, originating at different positions, were tilted toward one another. We believe that the observed branching, along with other important parameters such as the stacking angles, can be controlled by a judicious choice of sample properties such as island size and composition, or spacer thickness. We expect these new degrees of freedom to be of great interest for fundamental studies of quantum dot ensembles and for device applications. Tunable stacking angles, for instance, may significantly alter the way adjacent dots couple electronically, which may have impact on potential applications ranging from quantum dot memory and logic structures to quantum dot quantum computers.

In conclusion we have demonstrated an arrangement of Ge dome islands in Ge/Si multilayers: oblique stacking. Our data show that the spontaneous formation and self-organization of oblique island stacks are governed by a complex interplay of surface strain, generated by the already buried islands, and surface curvature, caused by the inherent tendency of large domes to carve out material from the surrounding planar substrate.

This work was supported by the National Science Foundation under Grant No. DMR-0081183.

[1] X. Hu and S. Das Sarma, Phys. Rev. A **61**, 062301 (2000).
[2] M. A. Reed, Sci. Am. **268**, 118 (1993).
[3] O. I. Micic, C. J. Curtis, K. M. Jones, J. R. Sprague, and A. J. Nozik, J. Phys. Chem. **98**, 4966 (1994).
[4] Q. Xie, A. Madhukar, P. Chen, and N. Kobayashi, Phys. Rev. Lett. **75**, 2542 (1995).
[5] Y.-W. Mo, D. E. Savage, B. S. Swartzentruber, and M. G. Lagally, Phys. Rev. Lett. **65**, 1020 (1990).
[6] G. Medeiros-Ribeiro, A. M. Bratkovski, T. I. Kamins, D. A. A. Ohlberg, and R. S. Williams, Science **279**, 353 (1998).
[7] J. Tersoff, C. Teichert, and M. G. Lagally, Phys. Rev. Lett. **76**, 1675 (1996).
[8] E. Mateeva, P. Sutter, J. C. Bean, and M. G. Lagally, Appl. Phys. Lett. **71**, 3233 (1997).
[9] L. Vescan, W. Jäger, C. Dieker, K. Schmidt, A. Hartmann, and H. Lüth, Mater. Res. Soc. Symp. Proc. **263**, 23 (1992).
[10] E. Mateeva, P. Sutter, and M. G. Lagally, Appl. Phys. Lett. **74**, 567 (1999).
[11] P. Sutter and M. G. Lagally, Phys. Rev. Lett. **81**, 3471 (1998).
[12] F. M. Ross, J. Tersoff, and R. M. Tromp, Phys. Rev. Lett. **80**, 984 (1998).