APPLIED PHYSICS LETTERS VOLUME 83, NUMBER 25 22 DECEMBER 2003

Growth of quantum fortress structures in $Si_{1-x}Ge_x/Si$ via combinatorial deposition

Thomas E. Vandervelde
Department of Physics, University of Virginia, Charlottesville, Virginia 22904
Piyush Kumar and Takeshi Kobayashi
Department of Electrical & Computer Engineering, University of Virginia, Charlottesville, Virginia 22904
Jennifer L. Gray
Department of Materials Science & Engineering, University of Virginia, Charlottesville, Virginia 22904
Tim Pernell
Department of Electrical & Computer Engineering, University of Virginia, Charlottesville, Virginia 22904
Jerrold A. Floro
Sandia National Laboratories, Albuquerque, New Mexico 87185
Robert Hull
Department of Materials Science & Engineering, University of Virginia, Charlottesville, Virginia 22904
John C. Bean[a]
Department of Electrical & Computer Engineering, University of Virginia, Charlottesville, Virginia 22904

(Received 2 July 2003; accepted 31 October 2003)

This study details the evolution of morphologies in the $Si_{1-x}Ge_x/Si$ system, under kinetically controlled conditions of 550 °C growth temperature and 1 Å/s growth rate. We find that, with increasing film thickness and Ge fraction, a series of three-dimensional structures develop, starting from pits, and leading to quantum fortresses and ridges. The quantum fortress structures are of special significance because of their potential application in quantum cellular automata. We establish approximate boundaries in the parameter space of film thickness and Ge fraction, in which these structures form. We present a simple model, based on kinetics and strain, to explain the observed structures. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1636268]

The SiGe/Si system has been the subject of intense study over the past decade. When $Si_{1-x}Ge_x$ is deposited on Si under conditions of sufficiently high strain and/or adatom mobility, it has been observed that a planar wetting layer appears, followed by roughening in the form of islanding. The three-dimensional (3-D) islanding arises due to strain relief considerations. The growth of $Si_{1-x}Ge_x$ on Si has been described as a classic Stranski–Krastanov (SK) process.[1] However, the actual progression of morphologies for SiGe on Si is considerably more complex than a simple SK model. As the thickness of the deposited layer increases, a series of metastable morphologies, including ‘‘hut clusters,’’ domes, and finally large dislocated islands are observed.[2–6]

Three-dimensional islands in the SiGe/Si system afford the possibility of quantum confinement, and offer unique optical and electronic properties. While quantum dots (QDs) have many promising properties, it is difficult to construct useful devices from them at this stage of their technological evolution. For device applications, QDs should be uniform in size and small enough so that quantum confinement occurs, and one should be able to arrange them in well-defined arrays. In this letter, we will discuss research that could offer solutions to some of the problems just discussed.

Deng and Krishnamurthy first reported growth of a regular array of four islands on the four corners of a pit.[7] However, they achieved this by depositing submonolayers of carbon before the SiGe deposition. In contrast, Gray *et al.* later grew these self-organized arrays of four QDs for certain Ge compositions, by depositing $Si_{1-x}Ge_x$ without using C.[8] Based on their four-walled shape, they called these structures ‘‘quantum fortresses’’ (QFs) and suggested their application as the unit cell for quantum cellular automata (QCA).[9] Since QFs have {105} facets, their shape is self-limiting. In addition, QFs have a natural self-organization of four QDs around a pit, similar to the structure of a QCA cell.

In our work, we explored the different structures that are formed when $Si_{1-x}Ge_x$ is deposited on Si (001), for a wide range of Ge compositions and thicknesses. We found that as growth progressed, a series of surface morphologies appeared, including pits, QFs, and ‘‘ridges.’’ [2] We observed the effect of varying the parameter space (thickness, Ge composition, temperature) on the evolution of these structures. We find that, even for small changes in these parameters, there can be sudden and complete transitions from one morphology to another.

For this study, $Si_{1-x}Ge_x$ films were grown via molecular-beam epitaxy (MBE) in a custom-built VG 90S double-chamber system. The base pressure in the chamber prior to growth was typically 2×10^{-10} Torr. The samples were grown on 100 nm (4 in.) Si (001) wafers. Because of the relative placement of the Si and Ge sources, the composition of the deposited film varies progressively across the

---

[a] Electronic mail: john-bean@virginia.edu

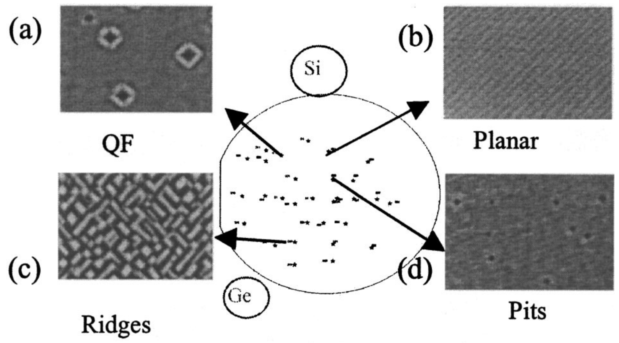

FIG. 1. Variation of morphologies across a 4 in. wafer with 30% Ge and 50-nm-thick film at the center, grown at 550 °C. Different structures are observed, because the film thickness and Ge composition vary across the wafer. The relative positions of the Si and Ge sources are pointed out. At the four AFM locations, the effective layer thickness and alloy fractions are approximately (a) QFs: 54 nm/30% Ge, (b) Planar: 57 nm/23% Ge, (c) Ridges: 45 nm/42% Ge, and (d) Pits: 50 nm/26% Ge. (The slight waviness observed in the background of areas (b) and (d) is believed to be an AFM artifact.)

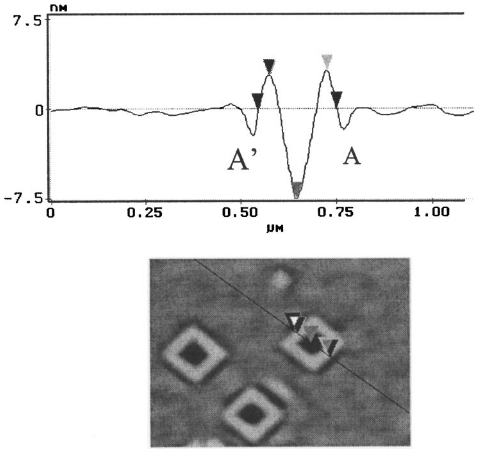

FIG. 2. Cross-sectional AFM analysis of a QF structure. Depressions can be seen at the edges of the QFs at points A and A′. Effective layer thickness is 49 nm with 31% Ge.

wafer. This also causes a variation in the thickness of the film across the wafer. (However, these variations can be made to disappear, if the substrate is rotated during growth.) By performing atomic force microscopy (AFM) scans over different parts of the wafer after growth, we were able to observe morphological evolution as a function of thickness and Ge percentage. In effect, we were able to obtain results from single wafers, which would otherwise have been possible only by doing scores of growths. The thickness and Ge fraction at any point on the wafer can be derived from the cosine law of emission for a finite-area source.[10] For example, on a sample with 30% Ge and 50 nm thickness at the center, the Ge composition varied from 20% to 45%, while the thickness ranged from 400 to 575 nm over the wafer. Our theoretical calculations were compared with the actual thickness profile of our wafers, and we found a maximum error of around 10%.

To understand the evolution of morphologies, we grew a series of 15 samples at 550 °C with 30% Ge at the center of the wafer, while varying the thickness of the SiGe layer from 5 to 175 nm. A deposition rate of 1 Å/s was used for all samples. Additional samples were grown at 20% and 40% Ge, to expand the parameter space. We also studied the effect of temperature on the growth, by using different growth temperatures for 30% Ge samples.[11] After depositing $\mathrm{Si}_{1-x}\mathrm{Ge}_x$, we analyzed the surface by AFM.

In Fig. 1, we can see the different structures that develop over the different regions of a sample with 30% Ge and 50 nm film thickness at the center. For regions with small Ge fractions (15%–25%), we found that the surface remained planar. However, at around 25% Ge, pits start to appear on the film [Fig. 1(d)]. The formation of pits seems to be a mechanism through which the film relieves strain.[12,13] We found that the edges of the pits always lie along the ⟨100⟩ directions.

On the same sample, when we move to regions of higher Ge content, we observed that islands start to develop at the four edges of each pit. For even higher Ge concentration, we observe four elongated islands (or QDs) around the pit, forming a QF [Fig. 1(a)]. From their facet angles, these islands appear to be huts. Figure 2 shows a cross-sectional analysis of a QF structure. The heights of the QDs which make up the QFs were measured using AFM, and we found that the QF height ranged from around 6 to about 12 nm. After QFs mature, some of them begin to develop outer walls, as shown in Fig. 2. For well separated QFs, double walls are observed roughly 10% of the time.

Around 35% Ge, the surface becomes covered with ridges [Fig. 1(c)]. These interconnected ridges are composed of {105} facets and have lengths much larger than their width. They lie along ⟨100⟩ directions and have been previously reported in the literature.[2] A similar progression of morphologies is seen across the wafer with change in film thickness. If we follow a contour line of constant Ge percent and increasing thickness on the wafer, we find pits followed by QFs, and then ridges at the maximum thicknesses.

This suggests that strain energy plays a strong role in the formation of these structures. In the absence of dislocations, the strain energy per unit area in a deposited layer is given by

$$E_{\mathrm{el}}=2G\varepsilon^2(1+\nu)h/(1-\nu),$$

where, ν is the Poisson ratio of the epilayer, G is the epilayer shear modulus, h is the epilayer thickness, and ε is the epilayer strain. It is clear that increase in Ge fraction as well as increase in thickness, should lead to greater elastic strain energy in the film.

In Fig. 3, we have collated our observations of the different structures, as a function of deposited layer thickness and Ge composition. We have been able to establish approximate boundaries in parameter space, inside which each structure predominates. From Fig. 3, it is clear that increase in strain, whether due to continued deposition or because of a higher Ge fraction, causes the structures to evolve. It is important to note from the figure that QFs appear only over a small range of Ge compositions, for any given thickness. Note that dislocations are expected only for metastable films, at the far right of the diagram in the regions where ridges dominate.

To explain the evolution of these morphologies, we consider the strain profile in the underlying layers. The planar regions will be under compressive strain. The formation of a pit allows the lattice at its edges to expand into its empty

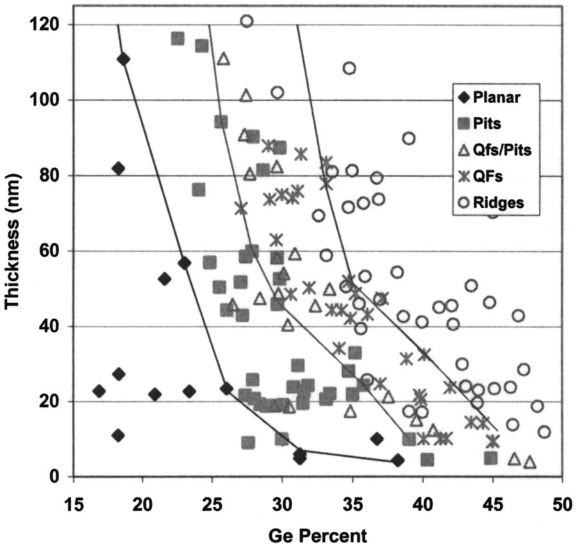

FIG. 3. Appearance of pits, QFs, and ridges as a function of film thickness and Ge composition, for samples grown at 550 °C. The graph shows approximate boundaries for the regions in which a planar surface, pits, QFs, and ridges are found.

volume, partially relaxing the compressive strain. Adatoms are then attracted to this dilation, and thus the edges of the pits provide a favorable location for the nucleation of Ge-rich islands.[13] On a (001) surface, this drives the observed formation of a square ring of four islands. As these Ge-rich islands grow, they can overdilate the lattice below. This creates a fringe area of lattice compression[14] from which subsequent Ge atoms are repelled. Island growth may stop here or, in certain instances, the strain can drive the formation of a moat-like ring of trenches around the fortress (points A and A′ in Fig. 2). Subsequent lattice relaxation due to the moat can then attract a second ring of islands, as seen in the bottom double-walled fortress of Fig. 2.

In this letter, we have described the morphologies that develop when $Si_{1-x}Ge_x$ is deposited on Si (001), under kinetically controlled conditions of 550 °C growth temperature and 1 Å/s growth rate. The first 3-D structures that developed to relax the strain in the film were pits. As the thickness and the Ge fraction in the deposited film increased, these pits evolved in size, until four hut-cluster islands start to form at each of the four edges of the pits. With increase in Ge concentration and thickness, these islands elongate until fully developed QF structures appear. We have found approximate values for the thickness and Ge fraction for which these structures form.

Because of the configuration of our MBE system, we were able to obtain a continuum of values for thickness and Ge percent over the wafer, for each growth. Hence, we were able to observe the evolution of 3-D structures as a function of the growth parameters. We showed that the successive transformations of the surface structures, from pits to QFs to ridges are strain driven.

This work was funded by NSF-DMR through the NSF-MRSEC at UVa, ‘‘The Center for Nanoscopic Materials Design’’ and the UVa-UIUC-IBM-Sandia Focused Research Group ‘‘Nanoscale Morphological Evolution of Semiconductor Surfaces’’ (Grant No. 0075116).

[1] D. J. Eaglesham and M. Cerullo, Phys. Rev. Lett. **64**, 1943 (1990).
[2] J. A. Floro, E. Chason, L. B. Freund, R. D. Twesten, R. Q. Hwang, and G. A. Lucadamo, Phys. Rev. B **59**, 1990 (1999).
[3] F. M. Ross, R. M. Tromp, and M. C. Reuter, Science **286**, 1931 (1999).
[4] F. Liu and M. G. Lagally, Surf. Sci. **386**, 169 (1997).
[5] Y.-W. Mo, D. E. Savage, B. S. Swartzentruber, and M. G. Lagally, Phys. Rev. Lett. **65**, 1020 (1990).
[6] M. Tomitori, K. Watanabe, M. Kobayashi, and O. Nishikawa, Appl. Surf. Sci. **76/77**, 322 (1994).
[7] X. Deng and M. Krishnamurthy, Phys. Rev. Lett. **81**, 1473 (1998).
[8] J. L. Gray, R. Hull, and J. A. Floro, Appl. Phys. Lett. **81**, 2445 (2002).
[9] C. S. Lent, P. D. Tougaw, and W. Porod, J. Appl. Phys. **74**, 3558 (1993).
[10] R. Glang, *Handbook of Thin Film Technology*, edited by L. Maissel and R. Glang (McGraw Hill, 1970), pp. 1–55.
[11] T. Vandervelde, P. Kumar, T. Kobayashi, T. Pernell, J. L. Gray, R. Hull, and J. C. Bean (unpublished).
[12] J. Tersoff and F. K. LeGoues, Phys. Rev. Lett. **72**, 3570 (1994).
[13] D. E. Jesson, K. M. Chen, S. J. Pennycook, T. Thundat, and R. J. Warmack, Phys. Rev. Lett. **77**, 1330 (1996).
[14] Z. M. Jiang, X. M. Jiang, W. R. Jiang, Q. J. Jia, W. L. Zheng, and D. C. Qian, Appl. Phys. Lett. **76**, 3397 (2000).