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Positioning of self-assembled, single-crystal, germanium islands by silicon nanoimprinting

T. I. Kamins,[a] D. A. A. Ohlberg, and R. Stanley Williams
Quantum Structures Research Initiative, Hewlett–Packard Laboratories, Palo Alto, California 94303-0867

W. Zhang and S. Y. Chou
Nanostructures Laboratory, Department of Electrical Engineering, Princeton University, Princeton, New Jersey 08540

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Strain energy from the lattice mismatch of a heteroepitaxial system can create ''self-assembled,'' single-crystal islands irregularly arranged on the surface. Alternatively, features of tens of nanometers can be patterned on a substrate by ''nanoimprinting'' using a mold and etching. When these two techniques are combined, the small patterned features can interact with the self-assembly process, causing the islands to form at the patterned features. The resulting regular array of very small islands may be useful for future devices. The positioning of single-crystal Ge islands by Si mesas formed by nanoimprinting and etching is demonstrated in this letter. © *1999 American Institute of Physics.* [S0003-6951(99)04512-X]

As integrated-circuit technology progresses, forming the smaller features needed for higher transistor density becomes harder. Even if features with small pitch can be achieved, controlling the critical dimensions by patterning and etching is more difficult. On the other hand, small islands can be formed by depositing one material on a substrate with a substantially different lattice constant.[1–4] The strain energy from the lattice mismatch can cause the deposited layer to ''self-assemble'' into small islands, which can potentially be used for electronic and photonic functions. Under controlled deposition conditions, the size of the islands can be quite uniform.[2] However, the position of the islands on the substrate is generally not well controlled, as is usually required if the islands are to be used for electronic functions.

By combining patterning with self-assembled island formation, strain energy can be used to determine the feature size while its position is determined by patterning. Control of the pattern size is then much less critical, allowing finer device dimensions or less-expensive fabrication.

We previously showed that Ge islands preferentially locate along edges formed on a Si surface by patterning and conventional integrated-circuit processing, giving control over the island position in one dimension.[5–7] The size and spacing of the islands can be much less than the lithographic patterning capability. Similar two-dimensional control of the island position can be achieved at points formed by patterning. Although the island size is determined primarily by strain energy, the minimum island spacing is then limited by the capability of the patterning technology.

Although electron-beam lithography can form small, closely spaced features, serial exposure limits its throughput, making it unable to pattern the tens of millions of features on an integrated circuit rapidly. A parallel exposure technique is needed. ''Nanoimprinting'' lithography has been shown to pattern sub-10 nm features over a large area in less than a few minutes.[8,9] The technique employs a reusable mold (equivalent to a mask) to pattern resist, followed by directional dry etching. It has been speculated that the nanometer-scale mesas formed by nanoimprinting can act as nucleation sites, allowing self-assembled, single-crystal Ge islands to form at the sites.[10]

This letter shows that 10–100 nm mesas formed in Si by nanoimprinting can be used to position self-assembled, single-crystal Ge islands, allowing the critical feature size to be determined by the strain energy while the island position is determined by nanoimprinting. Both nanoimprinting and self-assembly are parallel processes with the potential for rapid fabrication of device features, such as quantum cellular automata.[11]

Small Si mesas were formed on heavily doped, *n*-type, Si(001) wafers by nanoimprinting and dry etching. Mesas in the shape of squares and lines with height of 25 nm and nominal lateral dimensions of 10–60 nm were formed. Ge was deposited on these features under conditions that form self-assembled Ge islands on unpatterned Si(001) surfaces. For convenience in this experiment, the Ge was deposited by physical vapor deposition by electron-beam evaporation in an ultrahigh vacuum (UHV) system. (We previously showed that equivalent islands are obtained on unpatterned wafers by physical vapor deposition and by chemical vapor deposition, which is more amenable to large-scale production.)

The substrates were cleaned by a combination of conventional *ex situ* wet cleaning and *in situ* baking at an elevated temperature in UHV with a base pressure of $\sim 3 \times 10^{-10}$ Torr. Each sample was heated by direct current flowing through the sample, and the temperature was monitored with an optical pyrometer. Although unpatterned samples are usually heated briefly to $\sim 1250\,^{\circ}\mathrm{C}$ in UHV to prepare clean and ordered surfaces, the temperature was limited in this experiment to reduce damage to the Si mesas. After baking, the cleanliness of the Si substrate was examined by observing the dimer rows on the Si(001) surface using an *in situ* scanning tunneling microscope. Ge was then

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[a] Electronic mail: kamins@hpl.hp.com

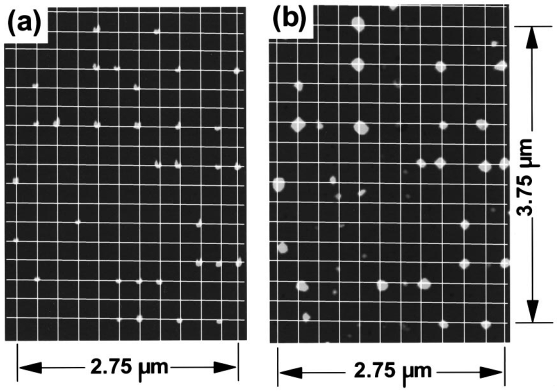

FIG. 1. Atomic-force micrographs of the sparse array of mesas with 250 nm pitch. (a) Before baking and Ge deposition. (b) After Ge deposition. The grid lines correspond to the 250 nm pitch of the array. The full height scale is 30 nm.

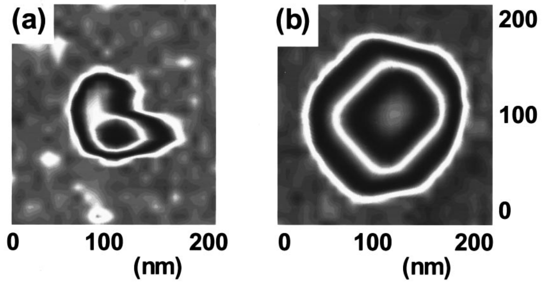

FIG. 2. Atomic-force micrograph of (a) the Si mesa before Ge deposition and (b) the Ge island at the same location, showing the characteristic shape of Ge islands on Si(001). The bands are lines of equal height to emphasize the shape of the island, with each pair of contour lines representing about 20 nm.

deposited at a nominal temperature of 600 °C for 9–11 min. The estimated amount of Ge deposited was about 8–11 eq ML ($1\,\mathrm{eq\,ML} = 6.27\times10^{14}\,\mathrm{Ge\ atoms\ cm^{-2}}$). On unpatterned wafers, this amount of Ge produces the faceted, ‘‘dome’’ structure that tends to a self-limiting size.[2,3]

After removing the samples from the deposition system, the structures were examined by tapping-mode, atomic-force microscopy (AFM) in air. (Selected patterns were also examined by AFM before inserting the substrates into the deposition system to allow correlating Ge islands with the corresponding Si features formed by nanoimprinting. However, changes in the feature sizes during surface preparation limit quantitative comparison of the size of a Ge island with that of the corresponding Si mesa.)

Figure 1(a) shows a portion of a sparse array of Si mesas formed by nanoimprinting small mesas with a 250 nm pitch. A grid is superposed on the AFM image for clarity in viewing. Mesas occupy about 13% of the grid points. Using a sparse array with a regular pitch allows the same locations to be studied before and after deposition and shows that the islands form preferentially on the nanoimprinted patterns. The sample shown in Fig. 1(a) was baked at a nominal temperature of 950 °C before deposition to obtain a Si surface clean enough to exhibit dimer rows and to allow high surface diffusion of the subsequently deposited Ge. Although the mesa dimensions were measured before the sample was inserted into the UHV chamber, baking at 950 °C markedly changed the Si patterns. Before baking, the mesa height was ~25 nm, and its length was about 65 nm; however, the mesa size was significantly reduced by baking at 950 °C before the Ge deposition started. Baking also greatly increased the roughness of the Si surface away from the patterns, possibly retarding subsequent Ge surface diffusion.

Figure 1(b) shows the same portion of the array after Ge deposition. Large Ge islands form on the Si mesas defined by nanoimprinting. However, no islands form at grid locations that had no Si mesas. The mean-surface dimension of the Ge islands associated with grid points is ~165 nm, and the height is about 35 nm. Some smaller Ge islands (mean-surface dimension ~80 nm) form between the filled grid points both because the large spacing used to aid visibility is probably comparable to the surface diffusion length of the Ge and possibly because the nonideal etched Si surface in this experiment limits Ge surface diffusion. Outside the patterned regions, Ge islands form randomly, with a density of about 4 islands/μm^2. The density of small islands within the array is considerably lower, suggesting that most of the deposited Ge diffused to the Si mesas to form Ge islands there. To eliminate islands between defined patterns, the maximum spacing of the features must be much less than the surface diffusion length corresponding to the temperature and time of the deposition and subsequent *in situ* heat treatment.[12] An optimally clean surface will also aid diffusion.

To avoid the ambiguity caused by Si movement during the *in situ* bake, the pattern on another sample was examined by *ex situ* AFM after baking at 800 °C. After recleaning, it was baked in the UHV chamber only at about 730 °C before Ge deposition. The minimal change of Si mesa dimensions during the 730 °C bake allowed comparison of the shape and size of the structure before and after Ge deposition, although the Ge diffusion on the surface was probably reduced because the surface was not as clean. Figure 2(a) shows an irregular Si mesa observed before Ge deposition, and Fig. 2(b) shows the more regularly shaped Ge island formed on the same Si mesa. Equal-height contours are shown in Fig. 2 to indicate the regular shape of the island. The base of the Ge island is not as well defined as on a smoother surface,[3] but its shape resembles the octagonal base of a Ge ‘‘dome.’’ The upper portions of the island show clearly defined facets with edges aligned along the ⟨110⟩ directions.

Even when the positioning mesa is larger, the shape of the Ge island tends toward its energetically favored dome-like shape. Figure 3(a) shows a Ge island nucleated at an elongated Si mesa about 150 nm long, 80 nm wide, and 25 nm high (dashed lines). Despite the elongated shape of the Si mesa, the Ge island has its characteristic dome shape, only slightly distorted by the Si mesa. The Ge island appears to have nucleated at a high region near one end of the Si mesa. Once nucleated, the island grew to its final shape with only a small effect of the remainder of the Si mesa. The final Ge island is about 60 nm high and 250 nm across.

These observations demonstrate that a small Si mesa can be used to position a single-crystal Ge island, while the general shape of the island is still determined by the energy of the materials system, as on a smooth, unpatterned Si(001) surface.

Ge islands on the unpatterned regions have dimensions

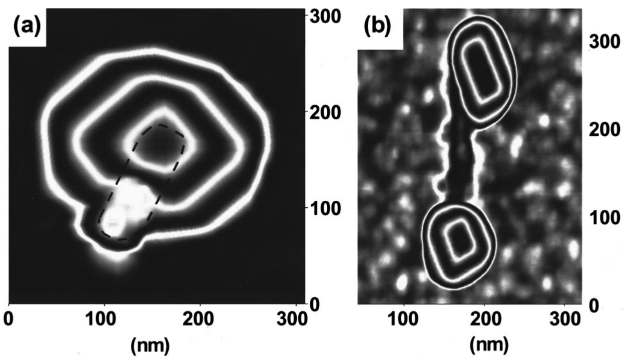

FIG. 3. (a) Atomic-force micrograph of Ge island formed on a larger Si mesa (outlined by the dashed lines); each pair of contour lines represents ~ 20 nm. (b) Atomic-force micrograph of a 40 nm wide Si line with Ge islands on both ends; each pair of contour lines represents ~ 10 nm.

similar to those of Ge islands formed on an unpatterned, atomically clean surface under similar deposition conditions. However, islands nucleating at Si mesas are considerably larger. The stable island size is probably determined largely by strain energy arising from the lattice mismatch between the depositing Ge and the underlying Si. In addition, the interface area between the Ge island and the Si is increased by the mesa. Additional edges and corners are also introduced, and the Ge volume is smaller than that of the island by the volume of the Si mesa. All these factors are likely to change the stable island size. In addition, possible diffusion of Si from the substrate into the Ge island should be considered.[13] In the present experiment, the Si mesa may significantly perturb the stress and the resulting interdiffusion. However, the intermediate shapes sometimes seen during the intermixing process are not observed in this experiment, making alloying less likely as the cause of the larger island size.

Although small Si mesas can accurately position Ge islands, other mesa shapes can also influence their positions. Figure 3(b) shows a narrow Si line (~ 40 nm wide and ~ 10 nm high after baking at 800 °C) with Ge islands formed on the two ends of the line. The edges of the island are again aligned along the expected crystallographic directions. (The line was rotated by about 25° from a $\langle 110\rangle$ crystal direction in this case.) In other cases, the Ge islands formed at different positions along the lines, possibly because of small irregularities at the edges of the lines and variations in the surface cleanliness after limited *in situ* baking.

Although the positioning effect of depressions (contrasted to mesas) is not demonstrated by the nanoimprinted samples in this experiment, other groups[14] and a complementary experiment in this laboratory[15] have demonstrated that depressions, as well as raised mesas, can position self-assembled islands.

In summary, this work demonstrated the ability to form small Ge islands on a Si substrate using strain energy from the lattice mismatch of the heteroepitaxial Ge/Si(001) system, with the positions of the islands determined by artificially formed patterns. The decoupling of the island dimensions from the size of the patterned features relaxes the dimensional control needed for the patterning. Nanoimprinting can form positioning features smaller than can be obtained by conventional lithography, with all the features being formed in parallel. To use the technique, however, an adequately clean Si surface must be obtained without significantly changing the nanoimprinted features by excessive heating.

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