Positioning of Strained Islands by Interaction with Surface Nanogrooves

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When strained Stranski-Krastanow islands are used as ''self-assembled quantum dots,'' a key goal is to control the island position. Here we show that nanoscale grooves can control the nucleation of epitaxial Ge islands on Si(001), and can drive lateral motion of existing islands onto the grooves, even when the grooves are very narrow and shallow compared to the islands. A position centered on the groove minimizes energy. We use as prototype grooves the trenches which form naturally around islands. During coarsening, the shrinking islands move laterally to sit directly astride that trench. In subsequent growth, we demonstrate that islands nucleate on the ''empty trenches'' which remain on the surface after complete dissolution of the original islands.

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In many semiconductor systems, strained layers grow as islands in the Stranski-Krastanow growth mode [1]. This has attracted great interest as a way to fabricate defect-free ''self-assembled quantum dots'' without the inherent limitations of lithography. Many applications require some control over the position of the islands, and surface patterning has been studied intensely as a method for position control [2–7]. For example, a one-dimensional string of islands grows spontaneously at the edge of a lithographically defined mesa [8], with the island size and spacing not constrained by the lithography. More complete control over island position can be obtained by patterning the surface with a focused ion beam [9,10]. However this process may introduce defects or impurities, and it is not clear what role those play in controlling nucleation. Lithographic patterning of the substrate surface has also been used to obtain perfect ordering of islands in two dimensions [4,5,11,12]. This involves etching pits that are relatively large and deep compared to the island size, which limits the density and can be problematic for device integration.

Here we show that extremely small and gentle surface grooves can effectively control the island position. Unlike typical lithographic patterning, such mild patterning allows very close spacing of the islands while keeping an essentially planar substrate. We study Ge on Si (001), which has become the paradigm system due to its relative simplicity. During growth at 620 °C, nanoscale grooves as small as 50 nm wide and 0.5 nm deep can effectively guide growth of either individual quantum dots or one-dimensional strings of closely spaced dots. The grooves do not need to be straight—the string of dots follows the groove. During annealing at high substrate temperature (740–840 °C), we also observe nearby islands to move laterally onto the groove. The position control is thermodynamically driven, as proper positioning minimizes the energy. Presumably dimples of comparable dimensions could position individual dots at specific sites.

A unique feature of our experiment is the use of nanoscale grooves formed *in situ* without introducing damage or impurities. Rather than attempt to fabricate grooves directly, we use the trenches that form spontaneously around large islands. During annealing and coarsening, many smaller islands shrink and disappear [13]. They leave behind empty trenches, which serve as our prototype for gentle nanoscale patterning. To extend this to technological applications will require new patterning techniques [14,15].

The samples used in this study were grown by solid source molecular beam epitaxy. Ge was deposited at a rate of 0.04 ML/s while the substrate was kept at 740 °C or 840 °C. At high temperature there is significant intermixing during growth, so the islands are actually a Si-Ge alloy [16]. The morphology of the islands was investigated by ultrahigh vacuum scanning tunneling microscopy (STM) and by atomic force microscopy (AFM) in tapping mode under ambient conditions.

Figures 1(a) and 1(b) show AFM images of the sample obtained upon deposition of 5 monolayers (ML) of Ge on Si(001) at 740 °C. Under these conditions, there is significant coarsening taking place during growth or while the sample is cooled to room temperature. The larger and more steep-sided ''dome'' islands (D) continue to grow, while smaller shallower islands such as {105} facetted ''pyramids'' (P) and ''truncated pyramids'' (TP) shrink [13]. There are also empty trenches (ET) where an island has shrunk away entirely. Figures 1(c)–1(e) show higher resolution STM images of a dome, pyramid, and truncated pyramid, respectively.

In Fig. 1(c) the dome island is surrounded by a trench. Such trenches have been extensively discussed in the literature [17–21]. The situation is different for the shrinking

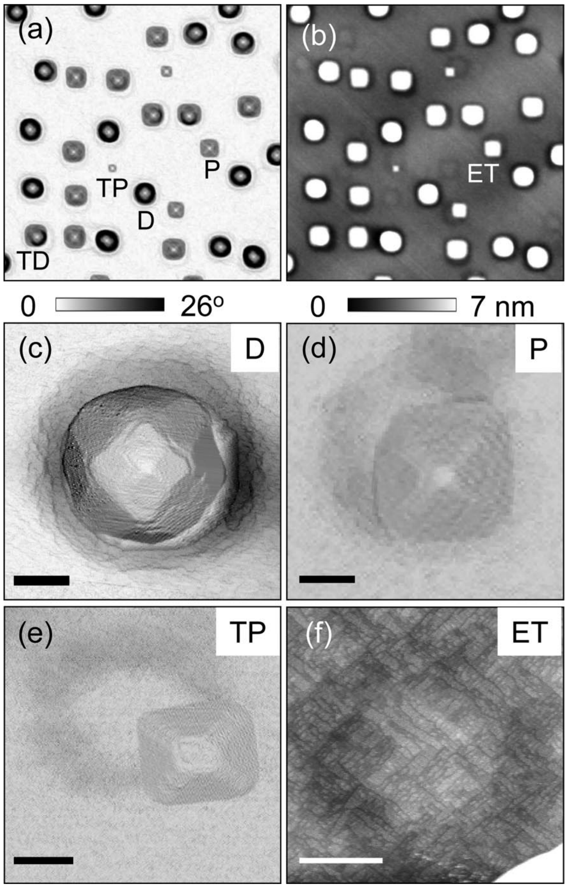

FIG. 1. Surface morphology after deposition of 5 ML of Ge on Si(001) at 740 °C. (a),(b) $2 \times 2\ \mu m^2$ AFM images of the sample. (c)–(f) STM images of a dome (c), a pyramid (d), a truncated pyramid (e), and an empty trench (f). The gray scale reflects the local surface slope with respect to the (001) plane in (a) to highlight the islands; the local height in (b) and (f), to highlight the trenches; and a combination of the local surface slope and height in (c)–(e) to show both the islands and the trenches. The sides of the images are aligned parallel to the [100] directions and the scale bars are 50 nm.

islands. As can be seen in Figs. 1(d) and 1(e), when the island shrinks it also moves laterally onto its own trench.

Figure 1(f) shows an empty trench which remains on the surface after the island has completely disappeared. The empty trenches are narrow and very shallow, with full width at half maximum around 50 nm, and depths around 0.5 nm. The surrounding surface shows an irregular $M \times N$ reconstruction typical for SiGe on Si (001). The average values of M and N are about 15 and 7.5, which are larger than those observed when Ge growth is performed at lower temperatures (see, e.g., Ref. [22]) because of a more pronounced intermixing. We see the same reconstruction inside the empty trenches, and even in the trenches surrounding dome islands, which cut deep into the Si substrate. This shows that all the trenches are lined with at least a thin wetting layer of SiGe [20].

To understand the effect of trenches on island position, we calculate the energetics within a two-dimensional model. Within the small-slope approximation [23], the elastic relaxation energy is

$$E = c\epsilon^2 \iint s(x)s(x') \ln|x - x'| dx dx'. \qquad (1)$$

Here c is an elastic constant; ϵ is the nominal misfit strain; and $s = dh/dx$, where $h(x)$ is the total thickness of the strained material at lateral position x (sum of height above surface and depth below).

We begin with a highly simplified case of triangular islands and trenches, with only three orientations present as shown in Fig. 2(a). For Ge on Si (001) the three orientations represent (001), (105), and $(\bar{1}05)$. But from Eq. (1), the results are independent of the specific misfit and slope, except for an overall scaling of relaxation energy as $(\text{misfit})^2(\text{slope})^2$. We neglect SiGe composition variations for purposes of this discussion, and refer to the SiGe simply as Ge.

For the geometry shown, the surface area and surface energy are independent of island position, so we need only to calculate the strain energy. The result is shown in Fig. 2(b). The energy is minimized when the island is centered on the trench. In the context of islands, strain is best relaxed for a large aspect ratio (height/width). This remains true for our island/trench geometry, if we measure

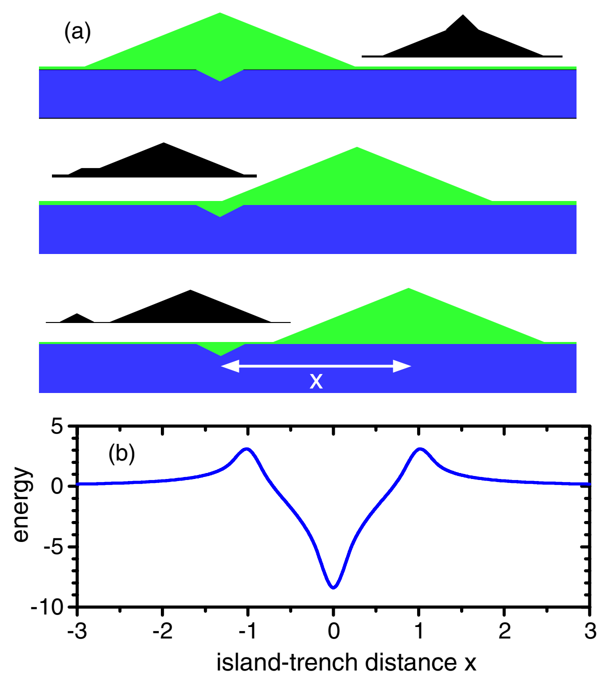

FIG. 2 (color online). (a) Schematic illustration of the geometry used to calculate relaxation energy, for lateral island positions $x = 0$, 1, and 1.5 in units of the island half-width. The trench is $1/5$ as wide as the island here. Black insets illustrate corresponding total thickness $h(x)$ of the strained layer; see text for details. (b) Energy vs lateral island position. The plot is independent of island size or slope—these factors only enter through the energy scale, which is left unspecified to emphasize this generality.

height vertically from the nonplanar Si-Ge interface. To avoid ambiguity, we refer to the strained-layer thickness, which in Fig. 2(a) is the sum of height above the surface and depth below the surface (in the trench). This thickness (projected above the surface level) is illustrated with the black insets in Fig. 2(a). Note that in Eq. (1), $s = dh/dx$ refers to this thickness rather than to the topographic height.

It is clear in Fig. 1(f) that the trench is not entirely filled with Ge as in Fig. 2. We therefore consider the energetics of an empty trench in equilibrium with an island (but not so close that the island induces significant strain at the trench). We denote the chemical potential of a flat epitaxial Ge layer as μ_0. The island serves as a reservoir of Ge, with a chemical potential $\mu_{\rm isl} < \mu_0$ due to elastic strain relaxation outweighing the extra surface energy of the island. (Depending on diffusion lengths, the effective reservoir might actually be some average of many surrounding islands.)

We can neglect strain relaxation in the shallow trench. Previous work suggests that the (001) orientation is not a facet for Ge on Si [24], and this is confirmed by the smoothly varying profile of the trench seen here. Thus the chemical potential $\mu_{\rm tr}$ in the trench can be described as $\mu_{\rm tr} = \mu_0 - \gamma\kappa$, where γ is the surface stiffness around (001), and κ is the curvature (1/radius) of the surface of the Ge in the trench [shown schematically in Fig. 3(d)]. As the trench fills, κ decreases toward zero and $\mu_{\rm tr}$ increases toward μ_0. Equilibrium is reached for a curvature

$$\kappa = (\mu_0 - \mu_{\rm isl})/\gamma. \tag{2}$$

If this curvature is larger than that of the underlying trench in the substrate, the trench will be empty except for a conformal wetting layer. But in equilibrium with a very small shallow island, $\mu_0 - \mu_{\rm isl}$ is very small, so a relatively deep trench would be nearly filled.

To check whether the trenches are partially filled with Ge, we have used selective wet chemical etching. A sample

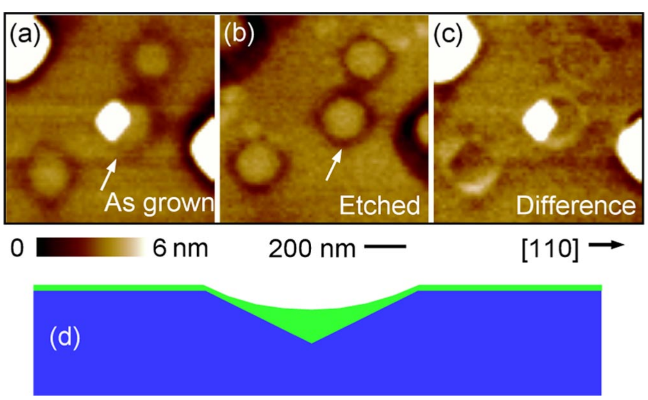

FIG. 3 (color online). (a)–(b) AFM images showing the same surface area (a) before and (b) after etching the sample for 30 s in a BPA solution. (c) Difference of (a) and (b), highlighting that the trenches are partially filled with SiGe. (d) Schematic illustrating the smoothly curved surface expected for the material in the trench.

obtained by deposition of 6 ML Ge at 840 °C was etched for 30 s in a buffered solution of hydrofluoric acid, hydrogen peroxide, and acetic acid (BPA solution) [25]. Figures 3(a) and 3(b) are AFM images of the same surface area prior to and after etching, respectively. Figure 3(c) shows the difference between the two images. We see that trenches on the SiGe surface are wider and slightly less deep (by about 0.2–0.3 nm) than those on the underlying Si substrate. These are clear indications that trenches are partially filled with SiGe. Similar results were obtained for a sample grown at 740 °C.

Thus Fig. 2(b) overestimates the energy for island positions where the trench is not completely covered—some Ge should move from the trench to the island, lowering the total energy (although the surface energy increases). Nevertheless, it remains true that the position directly over the trench minimizes the energy. This position gives the lowest strain energy because it has the highest aspect ratio for $h(x)$. It also has lower surface energy than a geometry with an uncovered, partially filled trench.

The empty trenches here are extremely small and gentle compared with typical lithographic patterning. If they can nevertheless drive motion of an existing island, it is natural to expect that they could also control the positions at which islands form during growth. Indeed, in theoretical simulations even a tiny dimple in an otherwise planar substrate is sufficient to determine the subsequent island position [20].

To test this, we first deposited 6 ML Ge at 840 °C, then annealed for 300 s at 840 °C before cooling down to 620 °C and depositing 6 ML Ge at this temperature. After the growth and annealing step at 840 °C the surface is similar to what observed in Fig. 1, except that the domes have alloyed and transformed into large pyramids [26].

Figure 4(a) shows the result of the two-step growth. Part of the Ge goes onto the existing large islands. The rest goes to form coherent islands, most of which are either (i) centered on the empty (or partially empty) trenches, or (ii) densely packed along the outside edge of the much deeper trenches around large islands.

Case (i) is highlighted in Figs. 4(b)–4(d). In Fig. 4(b), a single island forms centered on the empty trench, much as in Fig. 1(e). In Fig. 4(c), four well-separated islands form along the trench. In Fig. 4(d), a closely spaced string of islands forms along the trench. To verify that trenches can reliably position islands, we have analyzed 150 trenches by counting the number of islands nucleating on them during the second growth step [Fig. 4(f)]. This analysis shows that islands nucleate on all but about 1% of the empty trenches. The number of islands per trench reaches an upper limit at 8–9, which corresponds to an almost complete filling of the largest trenches.

Case (ii) is easily seen even in Fig. 4(a). The closely spaced lines of islands at the outer edge of the deep trenches are very similar to what is seen at the edges of large-scale lithographic structures such as mesa edges [8]. This comparison emphasizes that our small gentle trenches give a very different and flexible positioning of islands.

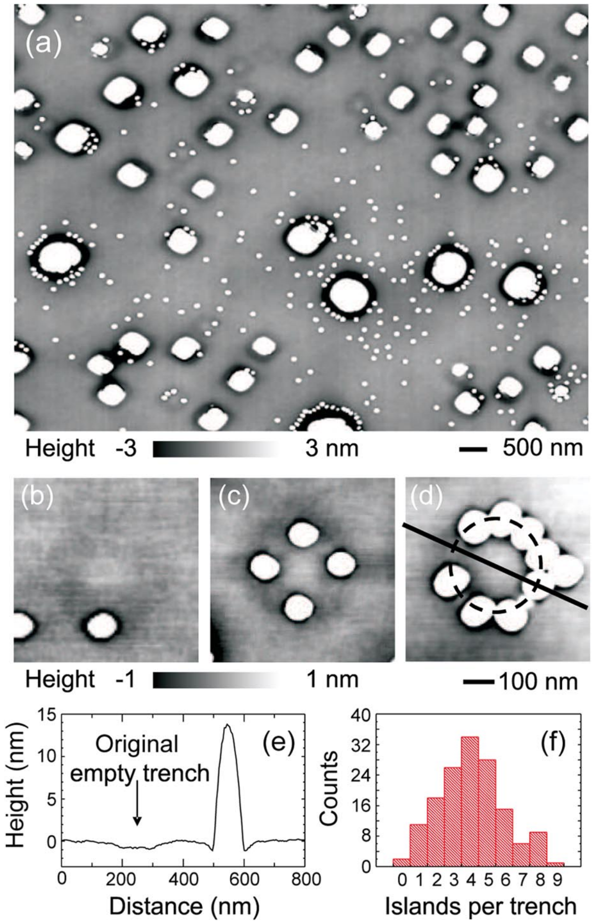

FIG. 4 (color online). AFM image of a sample obtained by growth of 6 ML Ge at 840 °C, followed by 300 s annealing, and then depositing 6 ML Ge at 620 °C. (a) Overview image showing both large islands from first growth, and small islands formed during second growth. (b)–(d) Closeups of (b) one, (c) four, and (d) a continuous string of small islands that formed on empty trenches during the second growth, with each image centered on a circular trench (barely visible due to its shallowness). (e) Line scan taken along the line marked in (d). (f) Histogram of number of islands nucleating on each empty trench in the second growth step.

One can obtain either isolated islands or closely spaced strings of islands, presumably depending on local coverage. This also suggests that single islands could be precisely positioned by gentle dimples or by crossing points of gentle trenches formed by some other technique. Crossing points of a crosshatch pattern in SiGe have been already successfully used to guide island formation [27].

Thus the gentle trenches seem very appealing for device application. The key problem is how to form them in a controlled way. A promising approach, at least for prototype studies, is the STM patterning demonstrated by Salling *et al.* [14,15]. Besides offering atomic-scale precision, such *in situ* patterning under UHV avoids the problems that can arise from surface contamination during *ex situ* treatments.

In conclusion, we have demonstrated that nanoscale grooves can drive lateral motion of islands onto the grooves, even when the grooves are very narrow and shallow compared with the islands. Both strain energy and surface energy are minimized in this way. Such motion occurs spontaneously when large epitaxial Ge islands on Si(001) shrink via coarsening. Exposed portions of empty trenches are partially filled with Ge, due to a competition between strain and surface energy. By applying a two step growth procedure we show that such nanoscale grooves can effectively control the growth positions of SK islands.

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