Controlled arrangement of self-organized Ge islands on patterned Si (001) substrates

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(Received 23 June 1999; accepted for publication 2 September 1999)

We report the ability to arrange self-organized Ge islands on patterned Si (001) substrates. Selective epitaxial growth of Si is carried out with gas-source molecular beam epitaxy to form Si mesas followed by subsequent Ge growth. Self-aligned and regularly spaced Ge islands are formed on the ⟨110⟩-oriented ridges of the Si stripe mesas. A *mono*-modal size distribution of the islands has been observed on the ridges. Using preferential nucleation sites allows us to place Ge islands at predetermined positions. The controlled arrangement of self-organized nanostructures offers the potential applications of island arrays for the implementation in nanoelectronics and quantum computation. © *1999 American Institute of Physics.* [S0003-6951(99)02644-3]

Since the observation of coherent, self-organized islands in strained heterostructures, such as InAs on GaAs (001)[1] and Ge on Si (001),[2,3] there has been a considerable interest in exploring their fundamental properties and applications in photonics and electronics. For understanding the nature and the mechanism of the formation of the islands, many studies on the size distribution, evolution,[4] and shape transition[5] of the islands have been carried out. Good size uniformity of self-organized dots has been reported[6,7] and self-organized dot based lasers have been demonstrated.[8,9] However, controlled spatial arrangement, which is usually required for electronic and signal processing applications, remains a major problem.

In order to control spatial distribution, many efforts have been devoted using a variety of techniques, such as growth on miscut substrates with surface steps[10] and on relaxed templates with dislocation network[11,12] and stacking growth of multilayers of islands.[13,14] Among them, one of the most effective approaches is using selective epitaxial growth (SEG) mesas as templates for the subsequent Ge growth. This approach shows one-dimensional (1D) ordering of Ge islands along the edges of the Si stripe mesas, formed in patterned windows with large feature size prepared by conventional lithography.[15] However, the control of the top width of the mesas is a critical and difficult issue, and Ge islands are often formed in the central regions of the stripe mesas, leading to the corruption of 1D ordering. Furthermore, arranging self-organized islands at predetermined sites remains a major challenge for the implementations of nanoelectronics and quantum computation.[16]

Here, we report perfect 1D arrays of self-organized Ge islands along the ridges of the Si stripe mesas. These island arrays are formed due to the preferential nucleation at the ridge positions and the repulsive interactions between the neighboring islands through the substrates.[17,18] Furthermore, we also show the control of the arrangement of the Ge islands at the predetermined positions. Other kinds of arrangement may be controlled at will.

The Si (001) substrates were selected as the starting materials. The 400-nm-thick SiO_2 were formed by thermal oxidation. By using conventional photolithography, the square and stripe Si windows were formed with the edges of all the Si windows aligned along the ⟨110⟩ orientations. The patterned Si (001) substrates were chemically cleaned and dipped in a diluted HF solution to form a hydrogen-terminated surface. The growth was carried out in a molecular-beam epitaxy system with a Si_2H_6 gas source and a Ge Knudsen cell source. After thermal cleaning, about 120 nm Si was selectively grown in the exposed Si windows at 660 °C. Si mesas with facets were thus formed. Details on the facet formation in the SEG process can be found in previous publications.[19,20] After the Si growth, Ge was subsequently deposited at a growth temperature of 630 °C and a growth rate of about 0.01 nm/s. After the Ge growth, the samples were removed from the vacuum and the silicon oxide was etched away for atomic force microscopy (AFM) study.

Due to the anisotropy of the growth rate in the SEG process, sidewall facets are formed on patterned Si (001) and evolved from the dominance of the {113} facets at the early stage of the Si selective growth to the dominance of the {111} facets at larger Si thickness.[19,20] Continuous growth of Si will reduce the lateral size of the top (001) surface of the mesas, leading to the shrinkage and finally the full reduction of the top surfaces in small structures. In this particular case, the {113} sidewall facets dominate the mesa sides at the Si thickness of about 120 nm.

Figure 1(a) shows a 3D AFM image of the self-organized Ge islands on the ⟨110⟩-oriented Si stripe mesas, formed in the exposed Si stripe windows with a window width of 0.6 μm and the separation between two stripes of 0.1 μm. Perfectly aligned and regularly spaced 1D arrays of the Ge islands are formed on the ridges of the Si stripe me-

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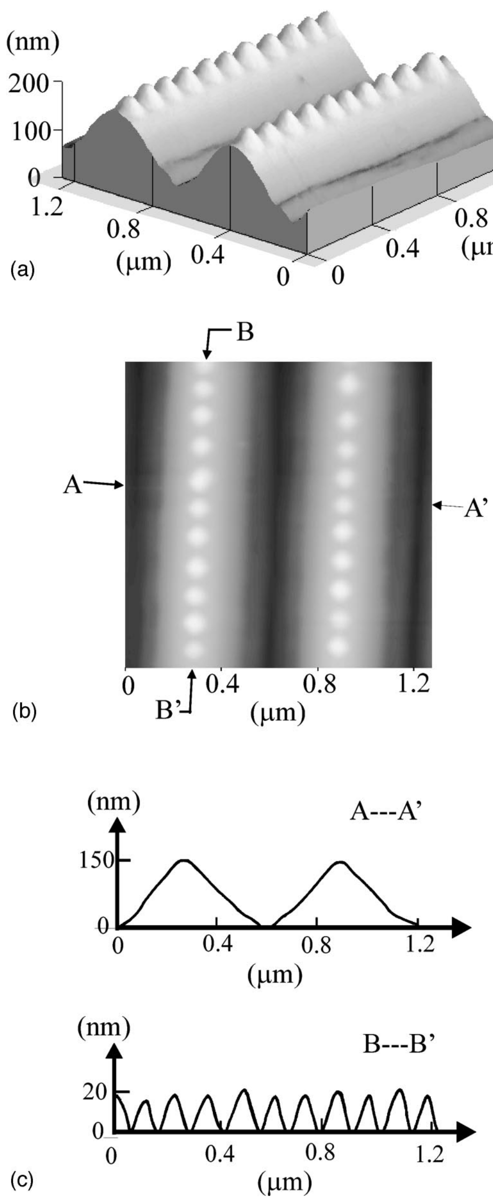

FIG. 1. (a) A 3D AFM image of the self-organized Ge islands on the ⟨110⟩-oriented Si stripe mesas with a window width of 0.6 μm. Self-aligned and well-spaced 1D arrays of the Ge islands are formed on the ridges of the Si mesas after the deposition of 10 ML Ge. (b) The 2D image of the island arrays in (a), along with the cross sections of the mesas (line AA′) and one array of the islands (line BB′), respectively. The sidewall facets are {113} facets.

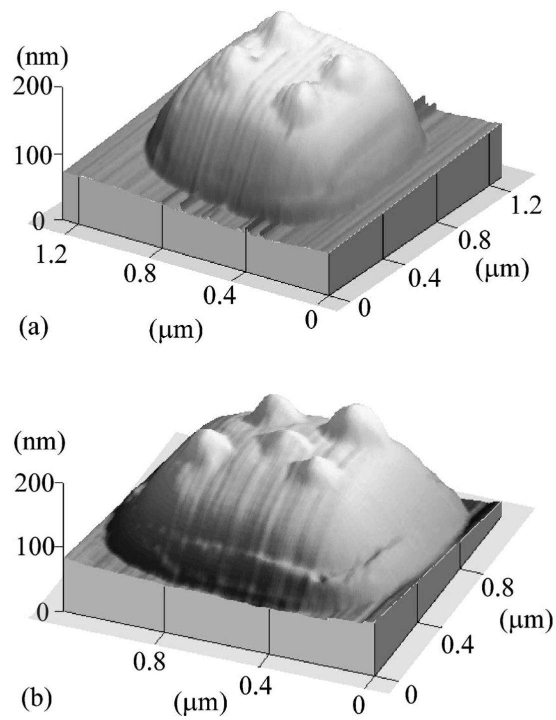

FIG. 2. (a) A 3D AFM image with four Ge islands located at the corners on a square Si mesa with the base lines parallel to the ⟨110⟩ directions. The Ge thickness is 9 ML. (b) A 3D AFM image with five Ge islands with 10 ML Ge. The fifth pyramidal island is formed in the central region. The average base size of the islands is about 140 nm.

sas. The perfect alignment of the islands along the Si stripe mesas is due to the formation of the ridges, which results from the full reduction of the top surface of the stripe mesas. Figure 1(b) depicts the 2D image of the island arrays in Fig. 1(a), along with the cross sections of the mesas and one array of the islands. The sidewall facets of the Si stripe mesas are confirmed to be {113} facets. The dimensions of the Ge islands are about 80 nm wide and 20 nm high, and the period of Ge islands is about 120 nm. The Ge islands with the dimensions are free of dislocations as discussed in a previous paper.[21]

It is interesting that only islands with a *mono*-modal distribution, which means that all the islands are dome shaped and have a close size of 70–90 nm, were observed on the ridges of Si stripe mesas over a large region. A similar result was reported on high index facets of SEG mesas,[22] in contrast with the results usually obtained on bare Si (001) substrates, where a *bi*- or *multi*-modal distribution of the Ge islands was present. The *mono*-modal distribution of the islands may be the result of the strain effect and the quasi-1D-spatial confinement. As seen in the facet formation in a SEG process, mass transfer from sidewalls to the top surface has been observed due to the anisotropic growth rates on different surfaces.[20] Our micro-Raman results (not shown here) indicate the tensile stress at the edges of the SEG Si mesas, which correspond to the energetically favorable nucleation sites. From the energetic point of view, the adatom diffusion along the 1D ridges to pass over the formed islands is limited due to the high energy barrier arising from the formation of the islands. Thus, it can be regarded as a quasi-1D case. This is different from the case, where the Ge adatom diffusion on the surface can be in a 2D plane, thus the Ge islands are randomly distributed on a plane. Therefore, the spatial confinement of the Si stripe mesas confines the diffusion of Ge adatoms from both directions of the two sidewalls, leading to uniform islands, a *mono*-modal distribution.

We have also investigated the Ge islands formed on the square Si mesas. As mentioned before, after the formation of Si square mesas in the exposed Si windows, four corners on the mesa are formed, which are the energetically preferred sites. Therefore, four Ge islands are formed on the square mesas with the base square oriented in the ⟨110⟩ directions [as shown in Fig. 2(a)]. In contrast, the central region is free of Ge islands. This is because the sites in the central region are not preferred and Ge adatoms have a sufficiently long diffusion length to migrate to the preferential corner sites. The preferential positioning enables us to place the Ge is-

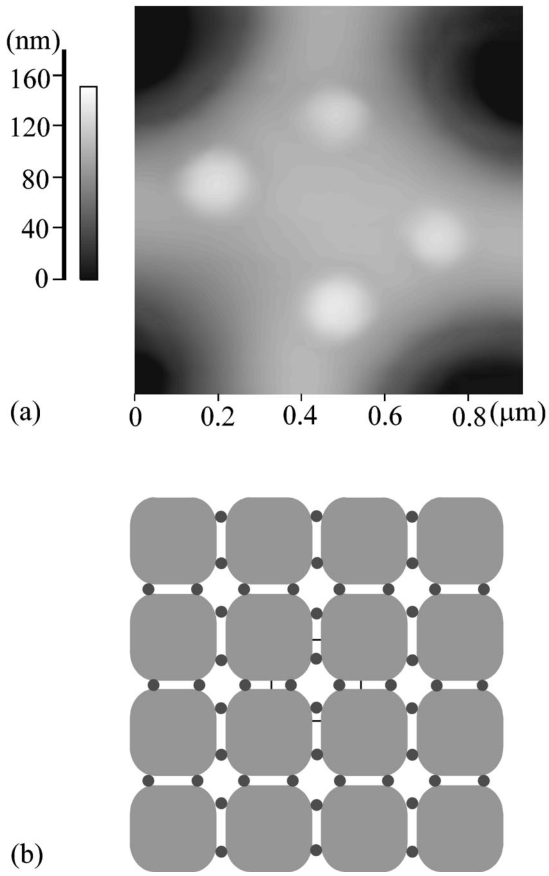

FIG. 3. (a) A 2D AFM image with four Ge islands. One could see four islands located at the predetermined sites based on the idea of the preferential nucleation positions. The average base size of the islands is about 130 nm. (b) A schematic of the pattern structure. The central small square cell with four spots is corresponding to the result in (a). The black spots represent Ge islands, and the white and dark areas stand for the raised Si mesas and the removed SiO_2 regions, respectively.

lands at the predetermined sites—the corners. When increasing the Ge deposition thickness, the fifth island is formed in the central region of the square mesas [shown in Fig 2(b)]. It is found that the central island is a pyramid with a square base, which is different from the other four dome-shaped islands at the corners. The central island is believed to be at the earlier stage of its evolution, i.e. that it has not yet undergone a shape transformation to dome shape. The possible mechanism is that the formation of the four islands at the corners changes the strain distribution, thus leading to the change of the energy distribution on the mesa. Compared with the corners, other sites, such as the center of the mesa become the preferential sites. Then the excessive Ge forms the fifth island in the center.

Figure 3(a) presents a 2D AFM image with four Ge islands on another kind of pattern structure. The schematic of the structure is depicted in Fig. 3(b), where the central cell is corresponding to the structure in Fig. 3(a). The SEG Si mesas were formed around originally raised square SiO_2 mesas oriented in the ⟨110⟩ directions. After the growth, the SiO_2 mesas were etched away for the AFM study. The result demonstrates that the four islands can be placed at the predetermined sites based on the idea of preferential nucleation [in Fig. 3(a)]. Compared with the four islands formed on square mesas shown in Fig. 2, the scheme shown in Fig. 3 is more favorable to make 2D island arrays for the applications of quantum computation.[16]

In summary, controlled arrangement of self-organized Ge islands on patterned Si (001) substrates have been investigated. Self-aligned and well-spaced Ge islands are arranged on the ⟨110⟩-oriented ridges of the Si stripe mesas after the full reduction of the top surfaces of the Si mesas. A *mono*-modal distribution of the islands has been observed on the ridges of Si mesas. Using the idea of preferential nucleation sites, we have demonstrated four Ge islands arranged at the predetermined positions. The success of the controlled arrangement of the self-organized island arrays offers potential implementations of quantum device architectures.

This work was partly supported by the National Science Foundation (NSF) and Semiconductor Research Corporation (SRC).

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