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# Self-ordering of Ge islands on step-bunched Si(111) surfaces

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By using step-bunched Si(111) surfaces as templates, we demonstrate the self-assembly of an ordered distribution of Ge islands *without* lithographic patterning. Initially, islands nucleate and evolve at step edges, up to complete ripening, forming long ribbons. Subsequently, island nucleation takes place at the center of flat terraces. Ge islands appear to be regularly spaced in scanning tunneling microscope images. The exploitation of this effect provides a possible route to grow ordered arrays of semiconducting nanostructures.
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The goal of reducing device dimensions down to the nanometer scale naturally raises the problem of how to control surface patterning with a spatial resolution that is not accessible with today's technology. Several studies have shown that long-range-ordered reconstructions can be used as templates for the growth of nanostructures.[1–4] Surface nanopatterning has been obtained in various ways, by taking advantage of self-organization or *artificially*, by forming patterns via e-beam lithography or focused ion beam. On Si(111) surfaces, dc heating may create bunches of natural surface steps,[5] yielding a simple way to obtain a nanopatterned substrate. Several authors have studied this phenomenon,[6–8] demonstrating that the step configuration at a vicinal surface depends on the direction of the current flowing through the steps, as well as on the miscut angle and on temperature.[6,9] With respect to the temperature dependence,[10] for $T > 1220\,°\mathrm{C}$, step bunching occurs in the step-down direction, while a regular step distribution occurs in the step-up direction. In this letter, we focus on the influence of surface morphology on the Stranski-Krastanov growth of Ge on both regular (R) and step-bunched (SB) Si(111) surfaces kept at 450 °C. Here, we analyze the evolution and distribution of the 3-dimensional islands that form after the completion of the wetting layer (WL). By varying surface preparation, we find an evident self-ordering on SB surfaces. The ordering process is studied by varying substrate patterning, coverage and terrace width.

Si(111) substrates ($n$-type, $\rho = 10^{-3}\ \Omega\,\mathrm{cm}$, miscut angle $< 0.5°$) were flashed for 30–60 s at about 1250 °C by passing a direct current of a few amperes, keeping the pressure below $5 \times 10^{-8}$ Pa. The R and SB surfaces were obtained with a current flow oriented in the step-up and step-down directions, respectively. Ge was deposited at a substrate temperature of $T_S = 450 \pm 20\,°\mathrm{C}$ by physical vapor deposition using a growth rate of about 0.3 monolayers (ML)/min. The samples were cooled down to room temperature and their surface morphology was observed *in situ* by variable temperature scanning tunneling microscope (STM)[11] at a base pressure of $4 \times 10^{-9}$ Pa. In Fig. 1(a), we report an STM image of a R surface. It consists of a staircase of equally spaced, fully reconstructed 7×7 bilayer steps of ~65 nm width and 0.31 nm height. From the image profile [Fig. 1(b)], we measure an average miscut angle $\theta < 0.3°$. By contrast, if we heat in the step-down direction, a SB surface is obtained, as shown in Fig. 1(c), along with a profile taken across the steps [Fig. 1(d)]. In this regime, the terraces have an average width of 1350 nm and are separated by bunches about 8.5 nm high, which corresponds to $N = 27$ atomic steps. From the measured staircase width $L_b = 450$ nm, we obtain a typical inter-

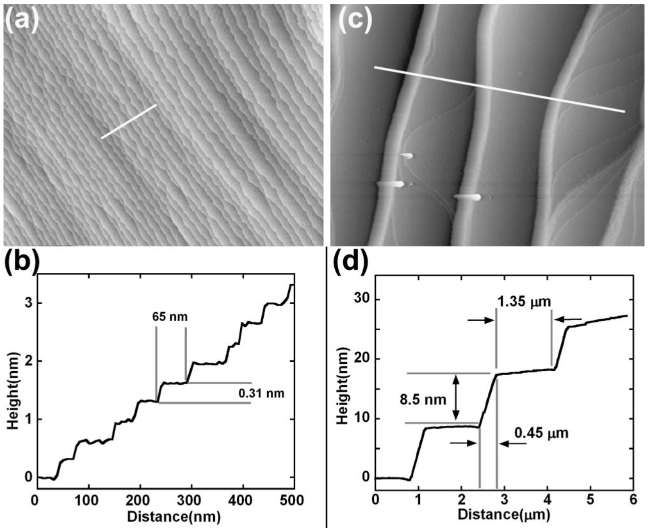

FIG. 1. Different morphologies of Si(111) surfaces after flashing at $T = 1250\,°\mathrm{C}$ by dc heating. (a) STM topography ($1700 \times 1700 \times 10\ \mathrm{nm}^3$) of a R surface obtained by current flowing in the step-up direction. (b) Height profile taken along the white line in (a). (c) STM topography ($7000 \times 7000 \times 36\ \mathrm{nm}^3$) of a SB surface obtained with current flowing in the step-down direction. (d) Height profile taken along the white line.

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step spacing inside the bunch $l_b = L_b/N = 16.4$ nm, in good agreement with previous results.[10]

An estimate of $l_b$ is obtained using the Burton–Cabrera–Franck-like model by Stoyanov and Tonchev in regime III[12]

$$l_b = 0.63 N^{-2/3} (18aA/F)^{1/3}, \tag{1}$$

yielding a value of 10.6 nm.[13] The reasonable agreement of this model with our experimental result confirms the validity of the scaling relation between the average step spacing $l_b$ and the number $N$ of steps in a bunch given by Eq. (1).

Having developed this realistic method to control the morphology of the Si(111) substrate by tuning current intensity and/or current direction, we now tackle the issue of the controlled positioning of Ge islands. As previously reported[14] on Si(111) mesas, atomic steps act as nucleation sites for Ge islands. Thus, in our case as well, we expect preferential nucleation on step bunches. After depositing 17 ML of Ge coverage on a R substrate, islands appear randomly distributed[15–17] [Fig. 2(a)]. By contrast, on SB substrates, islands first nucleate and evolve along step edges, and subsequently on flat terraces, as shown in Fig. 2(b). Islands grown on step bunches undergo complete ripening. They are elongated and they coalesce, forming a continuous ribbon.[18] When the evolution on the step edges is complete, nucleation takes place at the center of terraces [Fig. 2(b)]. As evidenced by the color-equalized image [Fig. 2(c)] and by the zoom [Fig. 2(d)], the supercritically thick WL appears to roughen as a consequence of the metastable strained state before the two-dimensional to three-dimensional transition.[19] In the central part of the terrace, the WL is made of regions of overgrown areas typically one bilayer high [Fig. 2(d)], whereas along the decorated edges, the WL shows a depleted region caused by the material that migrated towards the steps. This morphology is consistent with the idea that, due to intermixing,[20] the compressed Ge is more mobile,[21] even without actually melting.[22]

By increasing Ge coverage, step bunches appear fully decorated, the islands' size on terraces increases [Fig. 2(e)], and their density appears to be constant. Moreover, some flat, narrow terraces are found free of islands [Fig. 2(f)].

Lateral ordering of islands can be quantified by a statistical analysis of island–island and island–step-bunch distances. At a coverage of 8 ML [Fig. 3(a)] the average island–island distance is 360±10 nm. The two distributions of island–step-bunch distances (from upper and lower bunches) are reported in Figs. 3(b) and 3(c). They were fitted using two Gaussian peaks located at 470±20 nm for the island–upper-bunch (b) and at 520±20 nm for the island–lower-bunch distribution (c), respectively. Each peak gives the minimal distance from the step at which island nucleation takes place. As a consequence, by summing distances (b) and (c), we can estimate the maximum width ($w_{\mathrm{depl}} \approx 1$ $\mu$m) for an island-free terrace, caused by the adatom attraction towards step borders. Moreover, in each distance distribution, a smaller peak occurs at 750±20 nm and 860±20 nm for distances (b) and (c), respectively, corresponding to a second row of islands that nucleated on the terrace. The smaller intensity of this peak is caused by the lower number of terraces wide enough to host a double row of islands.[23,24]

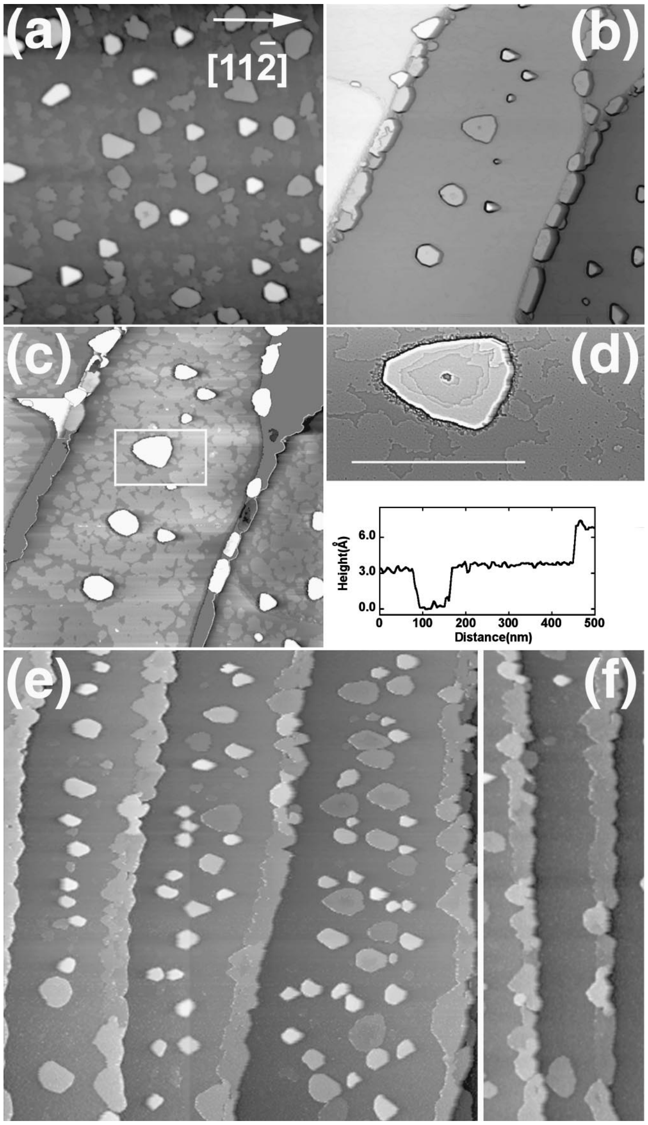

FIG. 2. Formation of the WL and distribution of 3D Ge islands on different Si substrates. (a) STM topography ($3000\times3000\times29$ nm$^3$) on a R surface after 17 ML Ge deposition at $T=450$ °C. (b) STM topography ($2950\times3000\times73$ nm$^3$) on a SB surface after 8 ML Ge deposition at $T=450$ °C. (c) Color equalized image ($2850\times2850\times73$ nm$^3$) to enhance the roughness of the WL in (b). (d) Zoom ($920\times440\times8$ nm$^3$) on the WL of (c) and profile taken along the white line. (e) STM topography ($10000\times10000\times30$ nm$^3$) on a SB surface after 19 ML Ge deposition at $T=450$ °C. (f) STM topography ($2660\times10000\times12$ nm$^3$) of an island-free terrace on the same sample of (e).

On a SB surface covered by 19 ML of Ge and varying terrace widths [Fig. 2(e)], we have measured both the island–island distance and the radial distribution function around each island (on each terrace). Values of 370±10 nm for the island–island distance and of 340±10 nm between nearest-neighbor and next-nearest-neighbor islands are found, showing that the island–island distance is nearly constant. This result implies that the density of ordered islands is constant, as previously reported for Ge deposition on patterned Si(001) mesas.[25] Thus, we expect that few rows of islands should form depending on the terrace width. This is strikingly apparent in Fig. 2(e): a single row forms on a 2.1-$\mu$m-wide terrace, while a double row forms on a wider terrace (2.8 $\mu$m). Below 1.2 $\mu$m, islands do not nucleate on the terrace, as shown in Fig. 2(f). Controlling the positioning

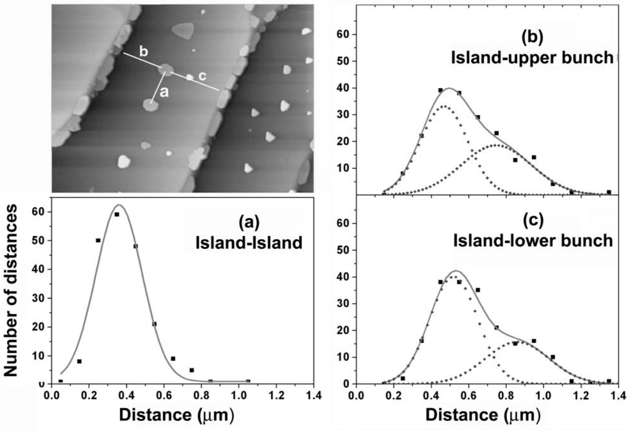

FIG. 3. Lateral ordering of Ge islands on the SB surface visible on the topographic STM image and quantified by the statistical distributions. The analysis is performed on a sample covered by 8 ML of Ge deposited at 450 °C. (a) Island–island distance. (b) Island–upper-bunch distance. (c) Island–lower-bunch distance. The histograms were fitted using Gaussian distributions.

of islands has been previously demonstrated by chemical vapor deposition growth on lithographically patterned substrates.[26] The present results show that the same is possible on Si(111) by using the natural patterning due to step bunching.

In summary, we have demonstrated controlled positioning of Ge islands on Si(111) *without* lithographic patterning. By using *in situ* STM, we have analyzed the island distribution on SB and R surfaces. We find a nearly ordered distribution of equally spaced rows of islands on the wide terraces of SB substrates.

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