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Nanoscale growth of silver on prepatterned hydrogen-terminated Si(001) surfaces

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Silver (Ag) growth on H-terminated Si(001) surfaces with prepatterns of silicon (Si) dangling bonds has been studied at room temperature using a scanning tunneling microscope (STM). The patterns were fabricated by extraction of hydrogen (H) atoms from a H-terminated Si(001)-(2×1) surface with a STM tip. The study covers four different patterns: growth on H-terminated Si(001) surfaces with (1) an isolated single dangling bond site, (2) a dangling bond wire (~1 nm wide), (3) a dangling bond area (20×20 nm^2), and (4) a dangling bond array. Ag adatoms readily diffuse on the surface and preferentially stick to patterns of dangling bonds. Ag adatoms are not spontaneously substituted for H atoms around the Ag adsorption sites. The behavior of Ag growth is different in each case. An isolated single dangling bond site can be occupied by an isolated single Ag atom or a Ag cluster. The dangling bond wire is covered by an Ag island of 1 monolayer (ML) in height. On the dangling bond area, Ag atoms preferentially stick to the edge of the area and a Ag island 1 ML in height grows in the direction perpendicular to the dimer rows. The behavior of each growth pattern is explained by the difference in binding energy of diffusing adatoms on H-terminated areas and Si dangling bond sites.

I. INTRODUCTION

The innovation of the scanning tunneling microscope (STM) enables us to observe surface structures[1] and to modify surfaces on an atomic scale.[2] Recently, atomic-scale extraction of hydrogen (H) atoms from H-terminated Si(001) surfaces was studied in order to understand the extraction mechanism[3] and to develop nanoscale electronics and optical devices. A monolayer of H can be used as a mask in a lithographic process, because H-terminated areas are less reactive to adatoms than the dangling bond sites. Subsequent deposition of adatoms onto the surface leads to selective growth on the dangling bond sites. The deposition of oxygen,[4] gallium,[5] iron,[6] and aluminum[7] onto prepatterned Si(001)-(2×1)-H surfaces has been reported in the literature. Adsorption and diffusion of adatoms on a H-terminated Si(001) surface have also attracted considerable interest in the study of homoepitaxial and heteroepitaxial growth,[8,9] because H atoms can change surface properties and lead to different growth modes.

Deposition of atoms onto a surface leads to diffusion, nucleation, and growth of adatoms. Creation of dangling bonds by a STM tip corresponds to the arrangement of nucleation sites on an atomic scale. A surface with nucleation sites arranged atomically is ideal for the study of growth processes, and understanding the processes on an atomic scale is important for fabrication of atomic-scale structures and nanoscale devices. However, growth processes on a prepatterned surface have not yet been studied in detail.

The present study focuses on silver (Ag) nanogrowth on a prepatterned Si(001)-(2×1)-H surface by comparing STM images of the same area before and after deposition. It is found that Ag adatoms grow on exposed Si dangling bond sites rather than on defects and step edges of Si substrates. An isolated dangling bond site can be occupied by an isolated single Ag atom or a Ag cluster. On the dangling bond wire, Ag grows row-by-row along the wire. On the dangling bond area, Ag atoms preferentially stick to the edge of the area and islands 1 monolayer (ML) in height grow layer-by-layer in the direction perpendicular to the dimer rows. On the dangling bond array, each dot of the array is covered by a monolayer of Ag atoms and then three-dimensional Ag clusters are formed. Ag adatoms are not spontaneously substituted for H atoms around Ag adsorption sites. Three-dimensional clusters form on an H-terminated Si(001) surface [the Volmer-Weber (VW) mode] and the growth is different from that on a Si(001)-(2×1) surface, where Ag grows in the Stranski-Krastanov (SK) mode.[10–13] The behavior of the growth is explained in terms of the difference in diffusion and adsorption of Ag adatoms on H-terminated areas and Si dangling bond sites.

This paper is organized as follows. Section III A describes the growth of Ag on a Si(001)-(2×1)-H surface with a few isolated dangling bond sites. Sections III B and III C show results of Ag deposition onto a Si(001)-(2×1)-H surface with a dangling bond wire and a dangling bond area of 20×20 nm^2, respectively. Finally, in Sec. III D, Ag growth on a dangling bond array is described.

II. EXPERIMENT

The experiments were performed in an ultrahigh vacuum (UHV) chamber with a base pressure of $\sim1\times10^{-8}$ Pa by a

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commercial STM (JEOL-JSTM 4000XV). Electrolytically sharpened tungsten (W) tips and antimony-doped (1×10^{18} cm^{-3}) *n*-type Si(001) samples were used. After cleaning the sample in UHV by repeated heating at 1500 K, it was exposed to atomic H, produced by dissociating H_2 using a hot tungsten filament. During deposition of H, the sample temperature (T_s) was kept at 600 K to obtain the 2×1 reconstructed surface.[14] In order to prepattern the structure, the STM tip was scanned on the surface under high sample bias voltage (V_s) and tunnel current (I_t) conditions and H atoms were extracted along the line scan. At a rate of $\sim$0.035 ML/min, Ag was evaporated onto the prepatterned sample which was kept at room temperature. During the evaporation, the STM tip was retracted from the imaging area by $\sim$100 nm in the *z* direction. The study of Ag growth was performed at a low coverage (less than 0.3 ML). At a high coverage, Ag atoms are adsorbed on the STM tip during the deposition and they disturb the interpretation of the STM images. All STM images in this paper were obtained in the topographic mode using $V_s=-2$ V and $I_t=0.2$ nA.

III. RESULTS AND DISCUSSION

A. Ag growth on an isolated dangling bond site

Figure 1(a) shows an occupied-state STM image of a H-terminated Si(001) surface. Most of the surface consists of monohydride rows in which each Si atom is bonded to one H atom. Bright spots are due to exposed Si dangling bonds having a concentration of 0.04–0.2 %. The area marked by the white arrow in the figure corresponds to dihydride rows in which each Si atom forms bonds with two H atoms. Figure 1(b) is a STM image of the same area taken after deposition of Ag atoms, suggesting that Ag adatoms preferentially stick to Si dangling bond sites and that three-dimensional Ag clusters are formed on the sites. Note that Ag atoms only grow on dangling bond sites and not on defects, the step edge, or dihydride sites.

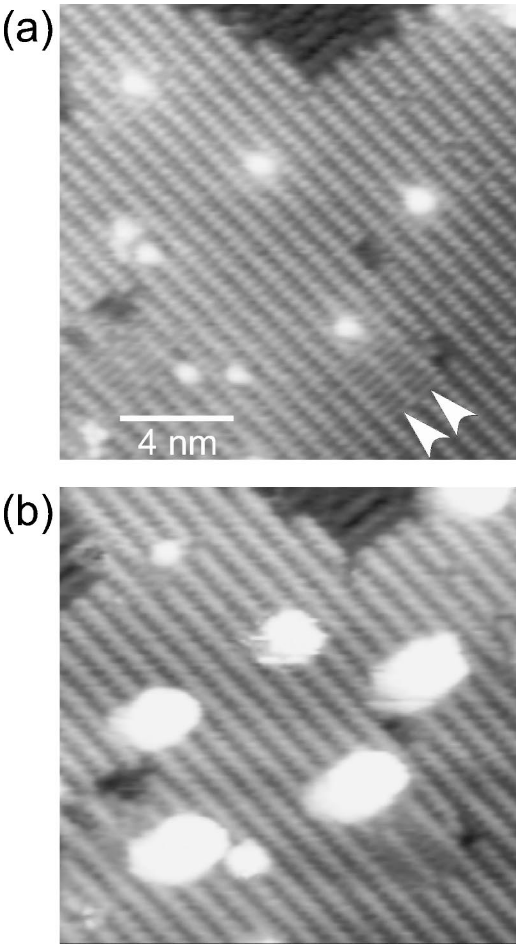

FIG. 1. STM image of a Si(001)-(2×1)-H surface taken (a) before and (b) after deposition of Ag. Bright spots in (a) are due to Si dangling bonds. The area marked by the white arrow is terminated by dihydride Si.

Figure 2(a) shows an occupied-state STM image of a Si(001)-(2×1)-H surface obtained before deposition of Ag. The surface consists of monohydride rows. The bright spots on the surface are exposed Si dangling bond sites and are labeled A–E. Figures 2(b) and 2(c) depict STM images of the same area recorded after 0.14 ML deposition and additional 0.07 ML deposition of Ag, respectively. Ag atoms have been preferentially adsorbed on Si dangling bond sites. In the direction perpendicular to the dimer row, cross-sectional line profiles of the bright spot A in Figs. 2(a) and in 2(b) are plotted in Fig. 2(d) as a broken line and as a solid line, respectively. The maximum point of the broken line is located on the left-hand side of the dimer. This suggests that one H atom of the dimer is missing. This is schematically illustrated at the bottom of Fig. 2(d). The maximum of the solid line, which is obtained after Ag deposition, is larger than the maximum of the broken line, indicating that one Ag atom has been adsorbed on site A. The difference between the two maxima corresponds to a height difference of 0.15 nm. Since sites B, D, and E have similar cross-sectional line profiles to that of site A, a single Ag atom is known to be adsorbed on each site. Sites A, B, D and E in Fig. 2(c) have the same line profiles as in Fig. 2(b). Within the coverage considered in the experiment, these sites are not nucleation sites of Ag growth. On the other hand, the protrusion of site C in Fig. 2(b) has a diameter of 2 nm and a height of 0.4 nm, and a three-dimensional Ag cluster grew on this site. The Ag cluster on site C is larger in Fig. 2(c) than in Fig. 2(b), suggesting that the Si dangling bond site acts as a nucleation site of Ag growth.

On a Si(001)-(2×1)-H surface with a few Si dangling bond sites, all Ag clusters have been formed by more than six Ag atoms. This suggests that there is a threshold size at which Ag clusters stay on a H-terminated Si(001) surface. The spherical shape of the Ag clusters indicates that the cluster is formed by Ag cohesive energy and weak Ag-H bonding. Since a cluster with a diameter larger than the threshold size is stable on a H-terminated surface and cohesive energy of a cluster increases with its size, the cluster grown on a dangling bond site acts as a nucleation site.

The structure of a single Ag atom being adsorbed on a Si(001)-(2×1)-H surface has been studied. Figure 3(a) is an occupied-state STM image of a Si(001)-(2×1)-H surface. The bright spot on the surface corresponds to one exposed Si dangling bond where one H atom on the right-hand side of the dimer is missing. Figure 3(b) is a STM image taken at the same area after the deposition of Ag. The protrusion of the bright spot in Fig. 3(b) is larger than that in Fig. 3(a), suggesting that a Ag atom has been adsorbed on the site. The

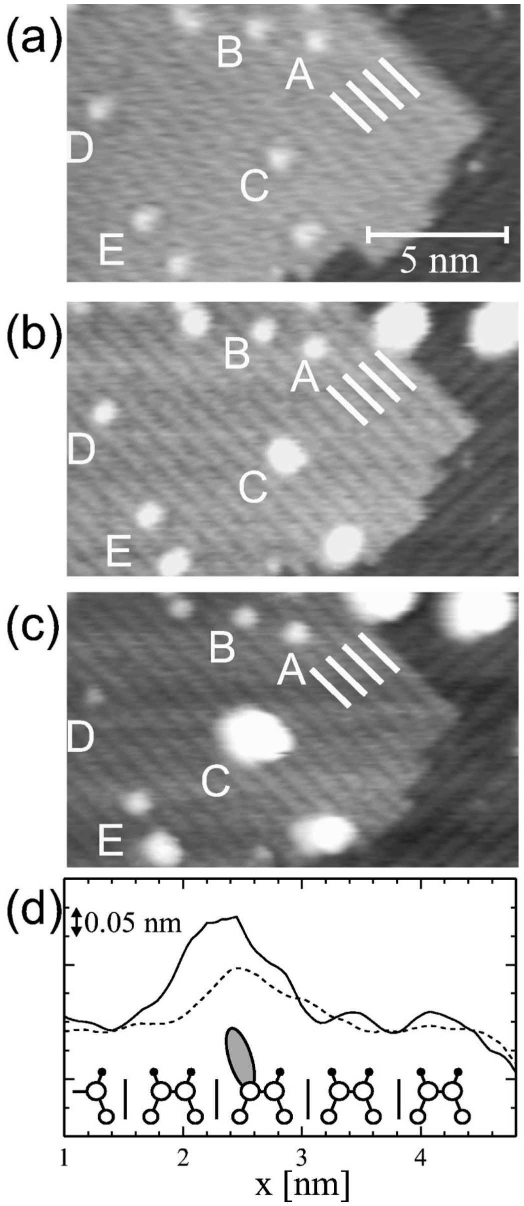

FIG. 2. STM images of a Si(001)-(2×1)-H surface taken (a) before, (b) after 0.14 ML deposition of Ag, and (c) after addition of 0.07 ML of Ag. Each bright spot labeled A–E in (a) corresponds to a single dangling bond. The white lines on the upper terrace in (a)–(c) show the positions between adjacent dimer rows. (d) Broken line: cross-sectional line profiles of bright spot A in (a) in the direction perpendicular to the dimer rows. Solid line: cross-sectional line profiles of spot A in (b). Schematic structure corresponding to the broken line is illustrated in the bottom panel. A white or gray circle and a black dot correspond to a Si atom and a H atom, respectively. A H atom is missing at the left-hand side of the dimer positioned at the center in the figure.

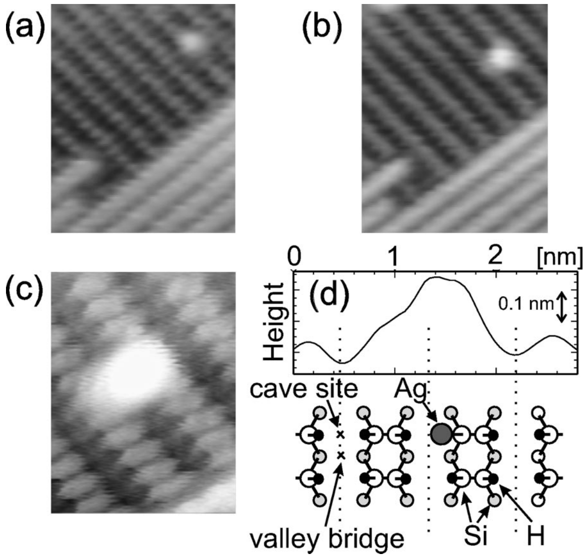

FIG. 3. STM images of (a) an isolated single dangling bond and (b) an isolated Ag atom adsorbed on the site between adjacent dimer rows on a Si(001)-(2×1)-H surface. The scanning area is $5\times7.5\ \mathrm{nm}^2$. (c) A focused STM image of the bright spot in (b). (d) Cross-sectional line profile of the Ag atom in (c) is the direction perpendicular to dimer rows. The corresponding surface structure is illustrated in the bottom panel.

site occupied by a Ag atom is on the left-hand side rather than the right-hand side of the dimer missing one H atom. This is caused by a single dangling bond readily flipping from one side to the other side of a dimer.[15] Figure 3(c) is an enlarged image of Fig. 3(b). In the direction parallel to the dimer row, the protrusion of the bright spot has a maximum on the cave site [see the bottom panel of Fig. 3(d)]. Figure 3(d) is a cross-sectional line profile of the bright spot in Fig. 3(c) in the direction perpendicular to the dimer rows. The line profile of the Ag atom is not symmetrical with respect to the center of adjacent dimer rows. The asymmetric shape suggests that the H atom on the left-hand side of the adsorption site still exists after Ag adsorption. The situation is schematically illustrated in the bottom panel of Fig. 3(d). The Ag atom forms a strong bond with the Si atom on the right-hand side of the adsorption site and a weak bond with the H atom on the left-hand side. The structure is different from that in the case of adsorption of a single Ag atom on a Si(001)-(2×1) surface,[12] where a single Ag atom adsorbed on the cave site forms bonds with neighboring Si atoms and causes buckling of adjacent Si dimers.[16] In Fig. 3(c), buckling of Si dimers is not observed around the Ag adsorption site, supporting the model shown in Fig. 3(d). The Ag atom is located on the right-hand side of the cave site and the Ag-Si bond has a suitable bond length. Thus the Ag adsorption does not induce a deformation of the Si atom forming a bond with the Ag atom.

In Figs. 4(a)–4(d), histograms of the number of clusters are depicted as a function of the lateral diameter of Ag clusters, which is estimated from the STM images recorded on the same area with different Ag coverage. At the initial stage of Ag deposition [see Fig. 4(a)], the diameter of the clusters has a Gaussianlike distribution. The solid curve in Fig. 4(a) is a Gaussian fit to the data with an average (d_A) of 2.27 nm and a standard deviation (σ_A^2) of 0.407 nm^2. Since the process in which a Ag cluster on a Si dangling bond site becomes as large as the threshold size and the site acts as a nucleation site occurs with a low probability (see Fig. 2), the size of clusters is thought to have a Gaussian distribution at the initial stage of Ag growth. The height of each cluster is proportional to its diameter with a proportionality factor of 0.17, the factor being kept almost constant at a different coverage considered in this experiment.

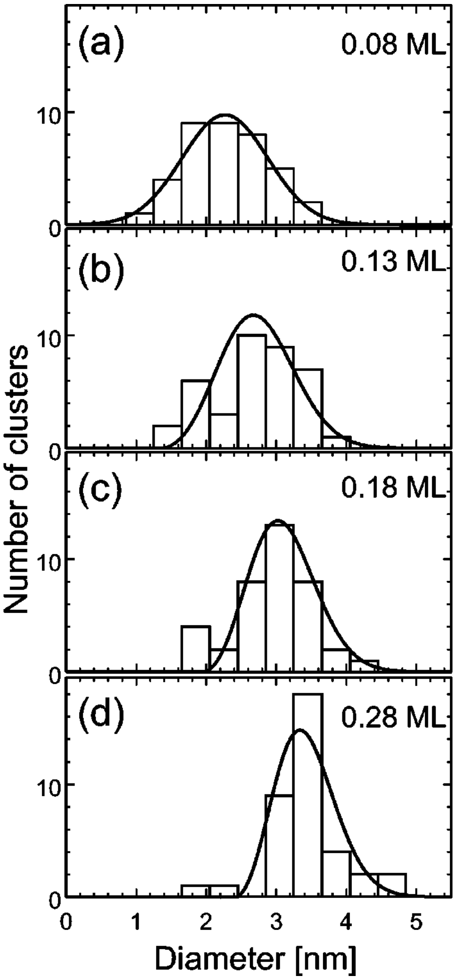

FIG. 4. Histogram of lateral diameter of Ag clusters on the same area with different coverage. Solid lines are given by Eq. (2) of the text.

Figures 4(b)–4(d) suggest that increasing coverage causes a narrowing in the Gaussian distribution. The behavior in the distribution can be explained quantitatively by the following model, which is obtained by simplification of several models for the process of island growth.[17] Ag atoms evaporated onto a H-terminated surface diffuse on the surface and stick to an existing cluster. The growth rate of a cluster is mainly determined by the frequency of arriving adatoms, the frequency being proportional to the circumference of a cluster. The number density of dangling bond sites on the surface is low, and there is a negligible effect of evaporated Ag atoms impinging on existing clusters. The rate equation of the number of atoms within a cluster with diameter d_i is given by

$$\frac{\partial N_i}{\partial t} = \sigma_I \pi d_i , \tag{1}$$

where σ_I is a proportionality constant that corresponds to the sticking probability of adatoms, and πd_i is the circumference of the cluster. The number of atoms within a cluster (N_i) can be approximated by the equation $N_i = \pi \rho k d_i^3/12$, where ρ is the number density of Ag, and the ratio k of height to diameter is 0.17, as observed experimentally. The following relationship can then be obtained: $\partial d_i/\partial t \propto 1/d_i$, which explains why the growth rate of a large cluster is suppressed. A simple calculation gives $d(t) = \sqrt{d_0^2 + At}$, where $A = 8\sigma_I/\rho k$. After substituting d_0 in the solution of the initial distribution $f(d_0, t_0) = C\exp[-(d_0 - d_A)^2/(2\sigma_A^2)]/(\sqrt{2\pi}\sigma_A)$, the distribution of the cluster diameter as a function of time reads

$$f(d,t) = \frac{C}{\sqrt{2\pi}\sigma_A^2}\exp\left[\frac{(\sqrt{d^2 - At} - d_A)^2}{2\sigma_A^2}\right], \tag{2}$$

where the parameters d_A and σ_A are determined by the initial fit in Fig. 4(a) and C is the number of clusters. The function then includes only one fitting parameter A. By setting $A = 0.017\ \mathrm{nm}^2/\mathrm{s}$, the calculated distributions are drawn as solid curves in Figs. 4(b)–4(d). The results are in agreement with the experimental data. This agreement implies that clusters are formed by arriving adatoms and the simple assumption that each cluster has the same sticking probability works within the coverage considered in this experiment.

The present study on a Si(001)-(2×1)-H surface shows that Ag grows in the VW mode, which is different from the situation on a Si(001)-(2×1) surface, where Ag grows in the SK mode. Since growth modes are simply determined by the balance of the free energies of the overlayer-substrate interface, overlayer-vacuum interface, and substrate-vacuum interface,[18] a different binding energy between the adatom and the substrate leads to a different growth mode. Since Ag adatoms on a H-terminated Si(001) surface are not spontaneously substituted for H atoms and the binding energy of a Ag atom on a H-terminated Si(001) surface is smaller than the energy on a Si(001) surface, a three-dimensional Ag cluster grows on a H-terminated area.

B. Ag growth on a dangling bond wire

A wire of exposed Si dangling bonds was fabricated by extraction of H atoms along monohydride dimer rows using the following scanning conditions of the STM tip: $V_s = +3.5$ V, $I_t = 2$ nA, and the scanning velocity $v_t = 2$ nm/sec. Figures 5(a) and 5(b) show occupied-state STM images of a Si(001)-(2×1)-H surface obtained before and after fabrication of a dangling bond wire, respectively. The wire has a width of one or two monohydride rows.

Figures 5(c) and 5(d) show STM images recorded after sequential deposition of Ag onto the prepatterned surface. Ag adatoms have been preferentially adsorbed on the dangling bond wire. In Fig. 5(c), the wire has been partially covered by Ag islands. Each island has a width of 1–2 nm along the wire. It is formed by 2–4 Ag atoms. After additional deposition of Ag [see Fig. 5(d)], the wire is almost entirely covered by Ag islands. Each island has a width of 2–4 nm along the wire. The island formation along the wire is different from that on a single dangling bond site, because the dangling bond sites adjacent to a Ag adsorption site are preferentially occupied by Ag adatoms. The preferential occupation suggests that there is an attractive force between the neighboring Ag atoms via the Si dangling bond wire. The force leads to island formation along the wire.

Cross-sectional line profiles of the wire have been obtained at four different positions, as shown in Fig. 6(a). The broken and solid curves in Fig. 6(b) are line profiles in the direction perpendicular to the wire before and after Ag depo-

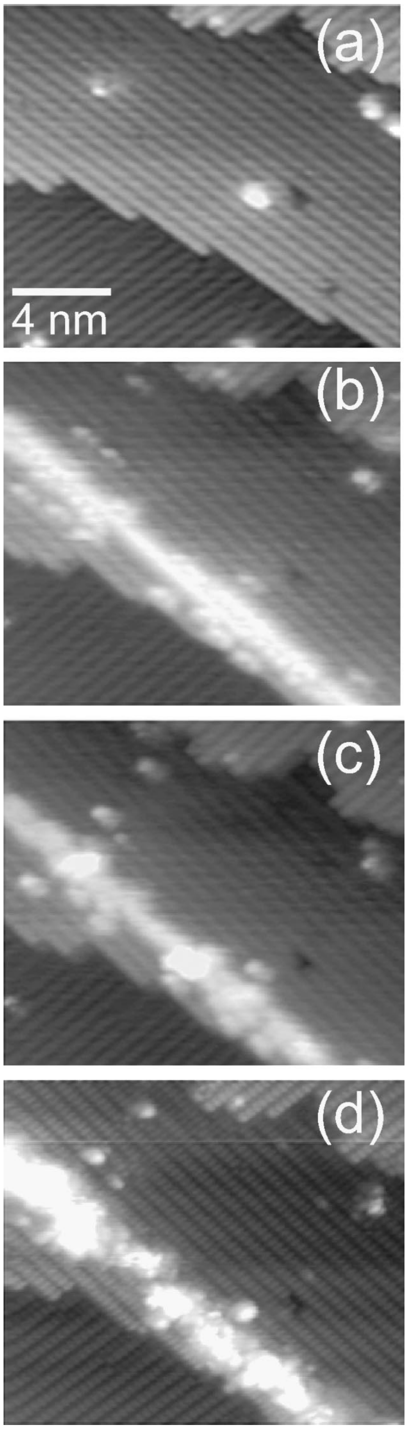

FIG. 5. STM images (a) before and (b) after fabrication of a dangling bond wire. STM images taken (c) after deposition of Ag and (d) after additional deposition.

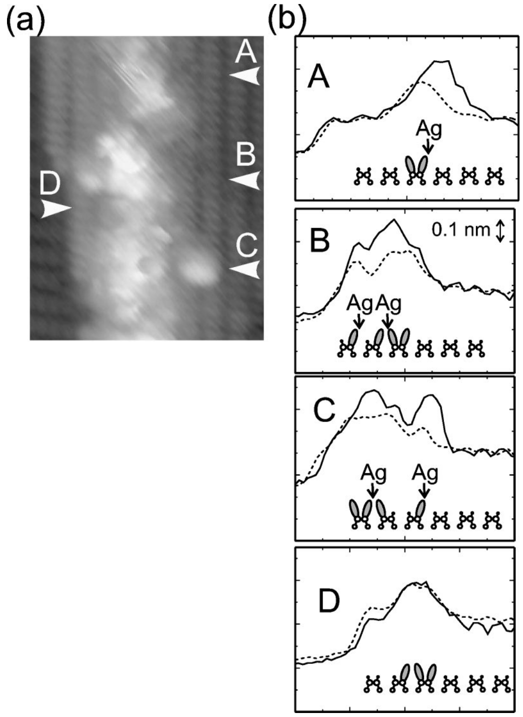

FIG. 6. (a) Focused STM image of the Ag wire shown in Fig. 5(d). (b) Cross-sectional line profiles of the wire at the positions marked in (a). The broken and solid lines are line profiles before and after deposition of Ag, respectively. The corresponding structure is schematically illustrated in the bottom panel of each line profile.

sition, respectively. The difference between the broken curve and the solid curve corresponds to the profile of the Ag island. The height of each Ag island is 0.15 nm, and it is nearly equal to the height in the case of adsorption of a single Ag atom [see Fig. 2(d)]. The structure of each profile is schematically drawn at the bottom panel of each figure with Ag atoms forming bonds with Si atoms. The schematic structures show that there are two adsorption sites on the dangling bond wire; a Ag atom forms bonds with adjacent Si atoms [see the right Ag atom in Fig. 6(b)C], and a Ag atom forms bonds with an adjacent Si atom and a H atom [see the Ag atom in Fig. 6(b)A]. The line profile at position D is the same as that taken before Ag deposition [Fig. 6(b)D], suggesting that a Ag atom is not adsorbed on that part and that Si dangling bonds still exist. The results show that Ag grows row-by-row along the wire until the wire is fully covered. This is because the Ag-Si bond formation is energetically more stable than cluster formation.[19,20]

The study of growth on a single dangling bond site and a dangling bond wire indicates that a H atom on a H-terminated Si(001) surface does not act as a surfactant. Ag adatoms are not spontaneously substituted for H atoms around Ag adsorption sites. The role of H atoms in the present study causing preferential adsorption on dangling bond sites is different from that in the case of Si or Ge adatoms growing on a H-terminated Si(001) surface, where an adatom spontaneously substitutes for one H atom from a surface Si dimer during adsorption.[8] The mechanism proposed for the substitution of H atoms[9] does not work in the case of Ag adatoms. In the case of Si and Ge adatoms growing on a H-terminated Si(001) surface, the energy barrier of the substitution is small because of the similar chemical properties of Si and Ge. In the case of Ag adatoms, the different chemical properties between Ag and Si probably lead to a large barrier of substitution.

C. Ag growth on a dangling bond area

This section describes the growth of Ag on an area of 20×20 nm^2 comprising Si dangling bonds. The surface consists of two parts: a dangling bond area and a surrounding

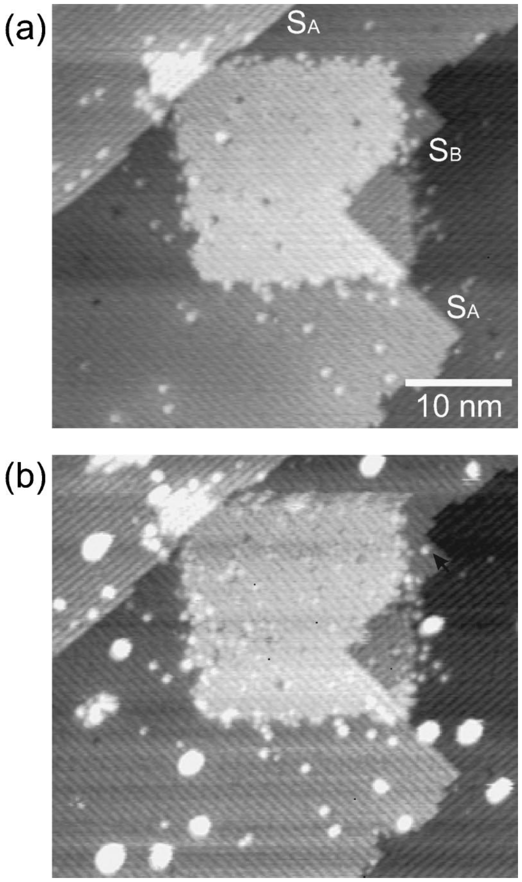

FIG. 7. STM images (a) before and (b) after 0.13 ML deposition of Ag. Two types of monatomic steps of the Si substrate are indicated by S_A and S_B.

area terminated by H atoms. A simple calculation based on a one-dimensional random walk of a Ag adatom suggests that the size of the dangling bond area is small enough for an adatom to diffuse from the center to the edge during the deposition time (~2 min).

Figure 7(a) is an occupied-state STM image of a Si(001)-(2×1)-H surface, in which H atoms on an area of 20×20 nm^2 have been extracted by scanning the STM tip at $V_s = 3.5$ V, $I_t = 2$ nA and $v_t = 300$ nm/s (= 25.6 lines/nm) from the top to the bottom of the area. The 2×1 reconstructed dimer rows are clearly observed on the extracted area. Monatomic steps are seen at the top left (S_A type) and bottom right (S_A and S_B types) of the area. There are a few isolated dangling bond sites on the surrounding area. Figure 7(b) is an STM image taken after 0.13 ML Ag deposition on the surface. Ag atoms have been adsorbed on the dangling bond area. On the surrounding H-terminated area, Ag clusters or single Ag atoms were observed on dangling bond sites as described in Sec. III A.

Figures 8(a)–8(d) are STM images of the dangling bond area with (a) 0.08, (b) 0.13, (c) 0.18, and (d) 0.28 ML Ag deposition. The coverage is estimated from the size of Ag clusters on the H-terminated area and it is almost proportional to the deposition time. Bright spots on the dangling bond area correspond to Ag atoms. They are drawn as filled circles in Fig. 8(a′)–8(d′). At low coverage, as shown in Figs. 8(a) and 8(a′), Ag atoms are adsorbed on the edge of the dangling bond area. Note that Ag atoms on the edge are distributed almost isotropically. On the dangling bond area, some isolated Ag atoms are observed on sites between Si dimer rows. As the coverage increases, Ag islands grow either from the edge or to a lesser extent from an existing Ag island. In Fig. 8(b), a small island has been formed on the top left of the large terrace. The island grows in the direction perpendicular to the dimer rows in Fig. 8(c). In Fig. 8(d), a chain has been formed from one edge to the other. The height of the chain is ~0.12 nm, suggesting that Ag grows layer-by-layer on the area. All islands grow in the direction perpendicular to the Si dimer rows. The situation is similar to the island formation observed on a clean Si(001)-(2×1) surface.[11] At the step edges S_A and S_B on the dangling bond area, Ag islands have grown on the lower terraces as observed in Fig. 8(d).

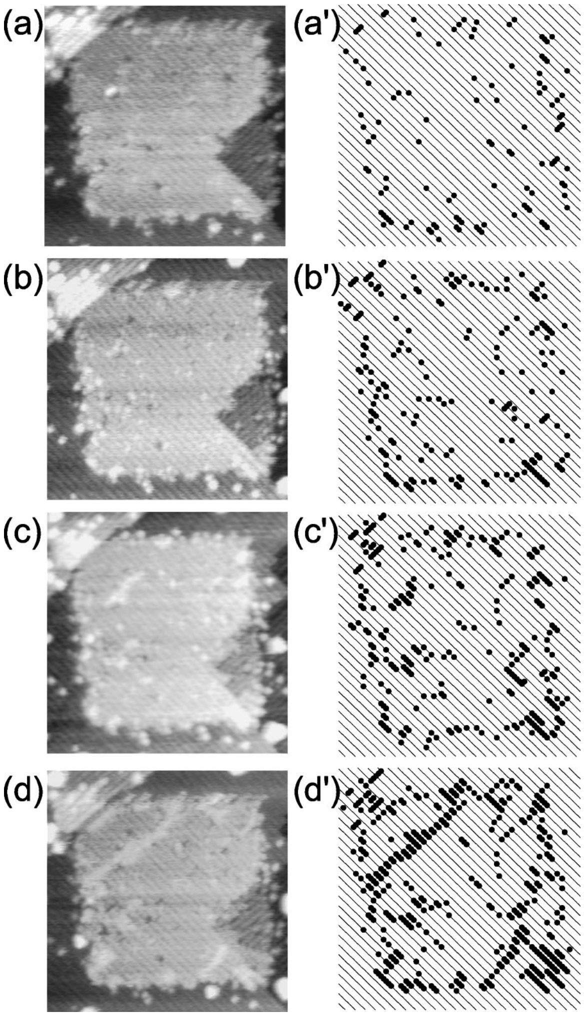

FIG. 8. STM images of Si dangling bond area recorded after sequential deposition of (a) 0.08 ML, (b) 0.13 ML, (c) 0.18 ML, and (d) 0.28 ML. The scanning area is 24×24 nm^2. (a′)–(d′) Corresponding spatial distribution of Ag adatoms on the dangling bond area. Each dot corresponds to a bright spot in (a)–(d).

The result that Ag atoms are adsorbed preferentially on the edge of the dangling bond area is explained by the diffusion barrier formed between the two different areas. Since the binding energy of Ag atoms on Si dangling bond sites is about 1 eV larger than that on a H-terminated area,[20] a diffusion barrier is formed at the boundary of the two areas. The barrier prevents Ag adatoms from diffusing out of the dan-

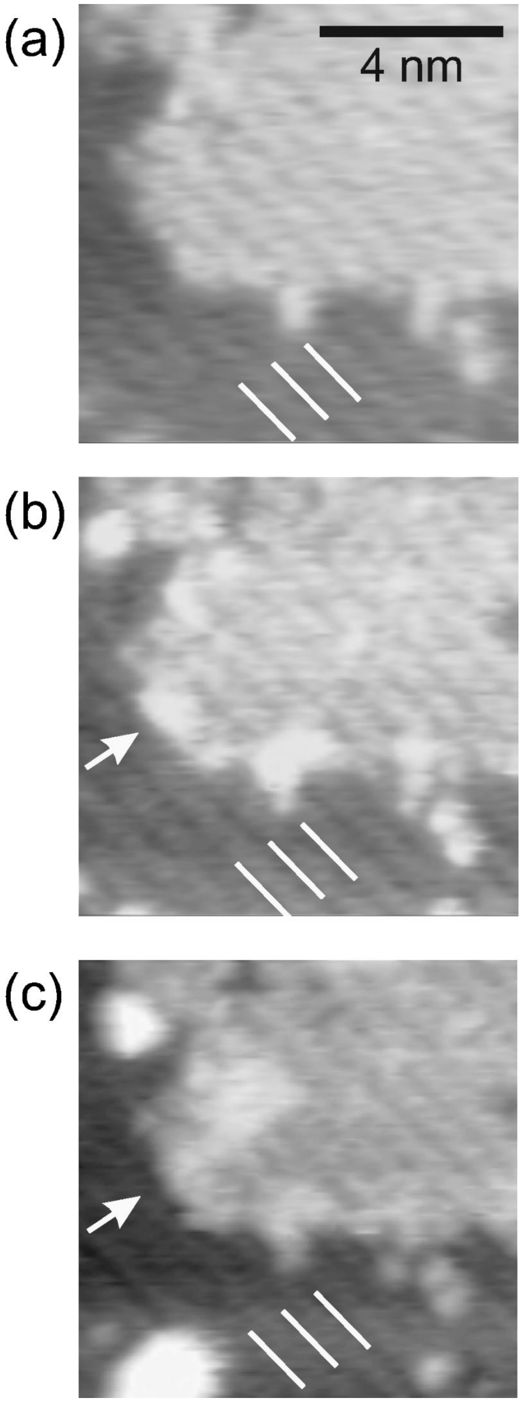

FIG. 9. Focused STM images of the dangling bond area with different Ag coverage. The bright area in (a) comprises Si dangling bonds [corresponding to the bottom left of Fig. 8(a)]. Ag islands are formed on the edge after (b) 0.13 ML [corresponding to Fig. 8(b)] and (c) 0.28 ML [corresponding to Fig. 8(d)] deposition.

gling bond area. Since Ag adatoms can diffuse into the dangling bond area from the surrounding H-terminated area, the imbalance causes nucleation on the edge. The isotropic distribution of Ag adatoms on the edge suggests that the islands on the edge are formed mainly by adatoms arriving from the surrounding H-terminated area with a larger diffusion coefficient. If we initially assume that the origin of atoms sticking to the edge stems solely from adatoms diffusing on the dangling bond area, then the distribution will be anisotropic, and Ag atoms will accumulate mainly at the top right corner and the bottom left corner. The results in Fig. 8 do not support this assumption and the preferential adsorption on the edge is attributed rather to Ag adatoms diffusing from the surrounding H-terminated area with a larger diffusion coefficient.

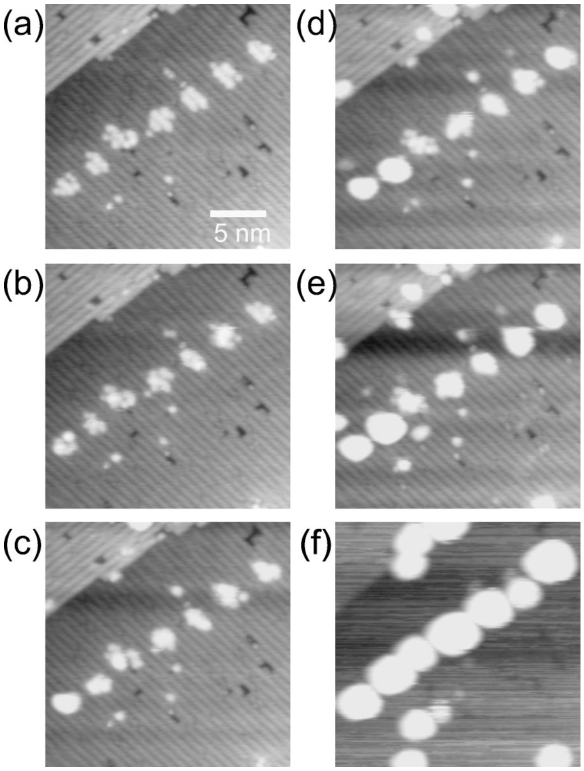

FIG. 10. STM images of a Si dangling bond array recorded (a) before and (b)–(f) after sequential deposition of Ag atoms.

Figures 9(a)–9(c) are STM images focusing on the bottom left of the dangling bond area in Figs. 8(a), 8(b), and 8(d). The white lines in the figures correspond to the positions between adjacent dimer rows. In Figs. 9(b) and 9(c), which are STM images taken after sequential Ag deposition, the position of the Ag island grown on the dangling bond area (marked by the white arrow in the figures) is the same as the edge of the Si dangling bond area in Fig. 9(a). They clearly show that Ag islands are formed solely on the dangling bond sites and that H atoms adjacent to the edge are not spontaneously substituted for Ag atoms.

D. Ag growth on a dangling bond array

In order to form a continuous Ag wire, another pattern of Si dangling bonds different from the one described in Sec. III B is available. Figure 10(a) shows an occupied-state STM image of a Si dangling bond array created by the extraction of H atoms using the STM tip. Each dot contains 8–12 Si dangling bond sites. Figures 10(b)–10(f) depict STM images recorded after sequential deposition of Ag atoms. At the initial stage of Ag deposition, single Ag atoms have been adsorbed on the dots [see Fig. 10(b)]. On some of the dots, two Ag atoms were observed. After subsequent deposition, the dots are almost covered by Ag islands 1 ML in height as shown in Fig. 10(c). The island formation is similar to that on the dangling bond wire in Sec. III B and on the dangling bond area in Sec. III C. As the coverage increases, three-dimensional Ag clusters are formed on the dots [Figs. 10(d)–10(f)]. Note that the size of the Ag clusters located at the two ends of the array is larger than the size of clusters located near the center of the array in Figs. 10(d) and 10(e). Similar features were observed in other experiments. This is because the Ag clusters are formed by diffusing adatoms on the surface. The number of adatoms arriving at Ag clusters located at the two ends of the array is larger than the number arriving at Ag clusters located near the center of the array. Additional deposition leads to further growth of each dot, and merging with adjacent Ag islands produces a continuous Ag wire.

IV. CONCLUSIONS

Ag atoms evaporated onto a prepatterned Si(001)-(2×1)-H surface diffuse on the surface and are adsorbed preferentially on Si dangling bond sites rather than on defects and step edges. Ag atoms grow layer-by-layer on the pattern of dangling bond sites until the pattern is covered by Ag atoms. An isolated dangling bond site can be occupied by an isolated single Ag atom or a Ag cluster. On a dangling bond wire, Ag grows row-by-row along the wire. On a dangling bond area, Ag atoms preferentially stick to the edge of the area and islands of 1 ML in height grow layer-by-layer in the direction perpendicular to the dimer rows. On a dangling bond array, each dot is covered by a monolayer of Ag atoms and then three-dimensional Ag clusters are formed. Ag adatoms are not spontaneously substituted for H atoms around the Ag adsorption site. The width of the structure covered by Ag atoms is almost the same as that of the pattern of dangling bonds, suggesting that a monolayer of H as a mask in a lithographic process works with atomic scale precision. Since island formation depends on the frequency of arriving adatoms, there is some possibility of inhomogeneous growth in the case of complicated patterns. The present study shows that the method based on STM lithography is useful for achieving atomic-scale structures, which is important for future nanoelectronics and nano-optical devices.

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