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Growth and modification of Ag islands on hydrogen terminated Si(100) surfaces

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The interaction of Ag with the Si(100)-2×1:H surface has been studied using a scanning tunneling microscope (STM). By comparing surfaces with various dangling bond defect densities, it is shown that such sites nucleate the growth of Ag islands. The critical Ag coverage above which coalescence of islands occurs, together with the mean height and width of islands are all determined by the initial dangling bond density on the Si(100)-2×1:H surface. We have also observed that the surface may be modified by scanning with a STM resulting in the transfer of Ag from the sample to the tip.

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I. INTRODUCTION

The formation of metal wires and contacts with nanometer dimensions represents an important step in the fabrication of atomic and molecular scale devices. One promising approach relies on the hydrogen termination of Si(100) to render a chemically inert or “passivated” surface. The particular attraction of this surface lies in its capacity for modification using a scanning tunneling microscope (STM). It has been demonstrated that by using high positive sample bias voltages or large tunneling currents desorption of hydrogen from the surface is possible over nanometer scale regions close to the tip. The dangling bonds which are formed in this process subsequently act as preferential adsorption sites for atoms or molecules which are subsequently deposited on the surface. Thus, the patterned surface acts as a template for metal adsorption. It has previously been shown that Al, Ga, Fe, and Co are preferentially adsorbed onto depassivated regions on the surface allowing the formation of nanoscale metallic patterns.

We have recently attempted to fabricate Ag wires using this method. The wires in our work straddle two conducting tracks formed by ion implantation. In these experiments, we have found that the adsorption of sufficient silver for the wire to remain conducting following the exposure to atmosphere leads to parallel conduction due to the coalescence of Ag islands nucleated at residual dangling bonds on the surrounding surface. Consequently, our attempts to measure the conductance of such a wire has met with limited success.

In this article, we report a study of the growth of Ag islands which may be used to determine the maximum Ag coverage which may be deposited while avoiding island coalescence. This is found to depend on the density of dangling bonds present on the surface prior to patterning. In addition, we present evidence to show that the deposited Ag films may be modified by the STM tip.

II. EXPERIMENTAL METHOD

The experiments were carried out using a commercially supplied ultrahigh vacuum (UHV) STM. Si(100) samples with dimensions 3×7 mm² were cut from a n-type wafer and loaded into the UHV system (base pressure ~7×10⁻¹¹ Torr). The Si sample was outgassed at 650°C overnight, flash annealed at 1200°C for 60 s, held at 800°C for 4 min, and then allowed to cool to room temperature. This annealing procedure consistently results in well-ordered (2×1) reconstructed surfaces with low defect densities.

Monohydride termination of the Si surface was achieved by the exposure to atomic hydrogen which was generated by cracking molecular hydrogen (99.999% purity) over a hot tungsten filament (~1500°C). The sample was held at ~400°C during this process. By adjusting the hydrogen dose, the resulting dangling bond defect density of the Si(100)-2×1:H surface could be varied, and subsequently determined from STM images.

Ag was sublimed at 830°C from a Knudsen cell at a deposition rate of 1.7±0.2 monolayers/min and a pressure of 1×10⁻¹⁰ Torr, while the Si(100)-2×1:H surface was held at room temperature [a monolayer (ML) is taken to be 6.8×10¹² atoms/cm², which is the number of Si atoms on an ideal Si(100) surface]. STM images were recorded in constant current mode using W tips fabricated by electrochemically etching and then heating in UHV by electron beam bombardment.

III. RESULTS

Figures 1(a)–1(c) show three hydrogen terminated Si(100)-2×1 surfaces prior to Ag deposition. The dimer rows are clearly visible, and defects including unsaturated dangling bonds and dimer vacancies are identified in Fig. 1(a). The average number of dangling bonds per unit area has been determined for each sample from 25 images of different regions on each surface (see Table I).

Figures 1(d)–1(f) correspond to area on samples (a)–(c), respectively, after the deposition of 1.1±0.1 ML of Ag. The bright topographic features observed in both filled and unfilled state STM images correspond to islands of silver atoms as reported previously for lower Ag coverage. At this coverage, the islands do not coalesce significantly and the majority of them appear circular in shape. By counting the islands for each surface, the mean densities have been
determined (see Table I) and are found to be very close to the dangling bond density. This confirms that the dangling bonds act as nucleation sites and also implies that further defects in the H termination are not generated by metal deposition. Note that for sample 3, the cluster density is slightly less than the dangling bond defect density. The high dangling bond density of this surface leads to some coalescence of the islands at this coverage. No correlation was found between the dimer vacancy defects and the cluster density.

From Fig. 1 it is clear that for a given coverage, the height and width of the islands are different for each surface. Measurements of full width at half maximum (FWHM) and height were taken for 200 islands on each sample and the mean values are presented in Table I. Histograms of island height for each sample are plotted in Fig. 2. Our results show that for a given Ag coverage, the mean values for island height and FWHM decrease with increasing surface dangling bond density. This in turn means that the fraction of the Si(100)-2×1:H surface which is covered by Ag is lower for surfaces with lower dangling bond densities.

In order to fabricate Ag “nanowires” on dangling bond templates, the coverage of Ag on the surface must be high enough to saturate all the dangling bonds and to form a connected metal track. However, the surrounding islands on the surface must have minimal coalescence so that the conduction path is solely through the nanostructure. Experiments were therefore performed on samples 1 and 3 to determine the relationship between coalescence, the density of nucleation sites, and the Ag coverage. The results are shown in Fig. 3 in which the number of islands per unit area is plotted versus the total Ag coverage. Note that the points for zero coverage correspond to the defect dangling bond density. For sample 3, the island density has fallen for the lowest coverage investigated indicating that some coalescence has occurred even at this stage. In contrast, the island density for sample 1 remains constant up to 1.7 ML at which point coalescence starts and the density falls. A comparison of samples at a coverage of 1.7 monolayers shows that for surface 1, the island density is ~95% of the dangling bond density, whereas surface 3 has an island density that is ~67% of the dangling bond density. At a coverage of ~8

**Figure 1.** (A)–(C) STM images of Si(100)-2×1:H surfaces with different dangling bond defect densities (samples 1–3, respectively), prior to deposition of Ag. (A) also shows a dangling bond defect (1) and a dimer vacancy (2); (D)–(F) STM images of the surfaces after deposition of 1.1 ML of Ag (samples 1–3, respectively) (25×25 nm², −2.5 V, 0.2 nA).

**Figure 2.** Histograms of cluster sizes (height) for samples 1, 2, and 3.

**Figure 3.** Cluster density as a function of total coverage for samples (1) and (3). The STM images show the morphology of the islands for sample (3) at different stages of island growth (30×30 nm² images, −2.5 V, 0.2 nA).

**Table I.** Properties of Ag clusters grown on Si(100)-2×1:H.

<table>
<thead>
<tr>
<th></th>
<th>Sample 1</th>
<th>Sample 2</th>
<th>Sample 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dangling bond</td>
<td>4.0±0.1</td>
<td>5.8±0.2</td>
<td>12.7±0.2</td>
</tr>
<tr>
<td>defect density</td>
<td>(×10¹⁶ cm⁻²)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cluster density</td>
<td>4.0±0.1</td>
<td>5.7±0.2</td>
<td>11.3±0.5</td>
</tr>
<tr>
<td>at 1.1 ML</td>
<td>(×10¹⁶ m⁻²)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Mean cluster</td>
<td>1.4±0.4</td>
<td>1.2±0.3</td>
<td>0.9±0.3</td>
</tr>
<tr>
<td>height (nm)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Mean cluster</td>
<td>3.7±0.8</td>
<td>3.4±0.7</td>
<td>2.7±0.6</td>
</tr>
<tr>
<td>FWHM (nm)</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
ML, the two curves merge and coalescence has proceeded so that there is no longer any dependence on initial dangling bond density.

The images in Fig. 3 show the morphology of the islands at different coverages for sample 3. As the growth proceeds, the islands coalesce and appear to become more oblique. At a coverage of ~18 monolayers, the islands are ~20 nm in width and the island density is a small percentage of the underlying surface dangling bond density.

It is possible to modify the Ag/Si(100)-2×1:H surface by transferring Ag to the tip of the STM. Scanning at a large negative tunneling current of ~4 nA (bias voltage ~2.5 V) over a small scan area of 2.4 nm² resulted in the removal of a single island from the surface as shown in Fig. 4. After the removal of this island, the surrounding clusters are not significantly larger, suggesting that the majority of the Ag atoms are adsorbed onto the tip. This assumption of adsorption to the tip is confirmed by the attempts to remove larger areas by scanning at the same parameters. It was found that large amounts of silver would often be redeposited at the center of scans where the tip idles. Changes in image quality in image quality due to changes in the condition of the tip also occurred.

It is also possible to remove the Ag islands by scanning at high positive bias voltages. Figure 5 shows a STM image of Ag clusters (~1 monolayer coverage) which has had a 30 nm² region scanned at +7 V bias and 0.2 nA. The background of the area from which the clusters have been removed is brighter than the surrounding surface background indicating that the hydrogen has been desorbed. One- and two-dimensional structures with an approximate height of 0.25 nm remain on the surface. These features have a close resemblance to the structures observed following the adsorption of Ag on the bare Si(100)-2×1 surface. A lower coverage of Ag (~0.45 monolayers) is observed on the surface after the removal process, and the surrounding area does not show any increase in cluster size, thus indicating that this process also induces transfer of Ag onto the tip.

At this stage, the detailed mechanisms for modification are unclear although in each case, the STM images show clearly that Ag is transferred from the surface to the STM tip.

IV. CONCLUSION

We have shown that the growth of Ag clusters on Si(100)-2×1:H occurs at the dangling bond defects. The coverage of Ag required to promote coalescence of the clusters was found to depend on the dangling bond density. It is therefore possible to conclude that a dangling bond nanowire on the monohydride surface created by the STM tip may be continuously coated with Ag with minimal coalescence of Ag islands on the surrounding surface for a dangling bond density ~4×10¹⁵ m⁻² and a Ag coverage in the range 1–1.7 ML. We have also shown that the clusters can be removed from the surface using the STM tip.

ACKNOWLEDGMENT

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Fig. 4. STM induced removal of a cluster. (A) Prior to removal (B) after cluster removal. The arrow indicates the cluster removed by scanning over 2.4 nm at parameters of ~2.5 V, 4.0 nA (17×17 nm² images, ~2.5 V, 0.2 nA).

Fig. 5. Scan induced fabrication of silver nanostructures. A 30×30 nm² area was scanned at +7 V, 0.2 nA. (A) Indicates a one-dimensional structure, (B) indicates a two-dimensional structure, (C) indicates the direction of the underlying dimer rows (50×50 nm² image, +2.5 V, 0.2 nA).