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A. A. Baski and C. F. Quate

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This report is a summary of our results during the second year of a research program aimed at studying the epitaxial growth of metals on the (111) and (100) surfaces of silicon using scanning tunneling microscopy (STM). The following three systems have been studied in depth during the past year: Al:Si(100), Sn:Si(100), and Yb:Si(111). The room temperature deposition of Al on Si(100) results in the formation of metal dimer rows, similar to the behavior of other group-III metals on Si(100). At elevated temperatures, Al forms "bow tie" structures which have also been observed on the annealed In:Si(100) surface. In contrast to the group-III metal systems, the annealed Sn:Si(100) system illustrates a complicated phase mixing behavior which involves numerous surface structures. In spite of the complexity of these structures, we believe that many of them can be simply modeled as Sn-Sn dimers. Finally, we have studied the Yb:Si(111) system, which forms two types of reconstructions up to 0.5 ML: (1) a lower coverage 3x1 phase and (2) higher coverage "n x 1" phases (5x1, 7x1, 2x1).

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**Edward L. Ginzton Laboratory
Stanford University
Stanford, CA 94305**

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Introduction

This report is a summary of our results during the second year of a research program aimed at studying the epitaxial growth of metals on the (111) and (100) surfaces of silicon using scanning tunneling microscopy (STM). The following three systems have been studied in depth during the past year: Al:Si(100), Sn:Si(100), and Yb:Si(111).

Our study of the room temperature deposition of Al on Si(100) completes our work on the nucleation and evolution of the 2×2 reconstruction for group-III metals on Si(100). As expected, we found that the Al, Ga and In:Si(100) systems exhibited a common growth mode near room temperature. At elevated temperatures, the Al:Si(100) surface behaved very much like the In:Si(100) surface, forming "bow tie" structures (~ 6 Al atoms) which had widely separated filled and empty electronic states. Our most recent work has involved a comprehensive study of the annealed Sn:Si(100) reconstructions. This system illustrates a complicated phase mixing behavior which can only be observed in real space by scanning tunneling microscopy. The deposition of Sn on the surface results in a series of surface structures, including "trenches", "stripes", "chains", and bright 1×5 overlayer features. Finally, we have examined the Yb:Si(111) system, which forms two types of reconstructions up to 0.5 ML: (1) a lower coverage 3×1 phase and (2) higher coverage " $n\times 1$ " phases (5×1 , 7×1 , 2×1).



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1) The Al:Si(100) System

Room Temperature Dimer Rows

We have studied the room temperature ($T < 150^{\circ}\text{C}$) behavior of the group-III metal Al on the Si(100) 2×1 surface. For metal coverages up to 0.5 ML, the coverage evolution of this surface is similar to previously studied group-III systems [Ga and In:Si(100)].¹⁻³ At very low coverages, the surface mobility of these metals is sufficiently high at room temperature that the metal atoms arrange themselves into rows lying perpendicular to the underlying Si dimer rows. These rows consist of metal overlayer dimers which lie over the trenches between the substrate Si dimer rows. As the coverage is increased, the metal configuration evolves from isolated rows, to areas of local 2×3 and 2×2 structure, to a surface terminated by a 2×2 array of metal dimers at 0.5 ML. In the case of the Al:Si(100) surface, more antiphase boundaries oriented perpendicular to the metal dimer rows appear to develop. As a consequence, the Al:Si(100) 2×2 phase is not quite as ordered as the other group-III systems.

Recently, we have examined more closely the two possible configurations for the metal overlayer dimers. The dimers can be oriented with the dimer bond perpendicular to the Si dimer bonds, as proposed previously.⁴⁻⁷ We believe, however, that the dimers can be rotated 90° so that the dimer bond is oriented parallel to the Si dimer bonds. This parallel orientation might appear less feasible because of the large bond angle distortions and the proximity of the metal atoms to Si atoms in the second subsurface layer. However, a similar adsorption site above second layer Si atoms is energetically favorable for the case of adatoms on the Si(111) surface.⁸ The parallel dimer configuration also provides a natural explanation for the strict $2a$ periodicity observed along each metal dimer row. Detailed calculations are necessary to determine whether the parallel dimer orientation is energetically lower than the presently accepted perpendicular orientation.

Since it is not possible to add more metal dimers once the 2×2 phase is formed, it is likely that the behavior of the group-III metals changes above 0.5 ML. To explore whether layer-by-layer growth continues at higher coverages, we examined the Al:Si(100) system in this coverage regime. Near 0.6 ML, three-dimensional clusters of Al formed on top of the 2×2 phase. At this

coverage, the clusters were still relatively small with an average height of 3 to 5 Å above the 2×2 surface. Further metal deposition resulted in the growth of these clusters, eventually making imaging difficult because of both increased surface roughness and instability of the metal in the clusters under the influence of the STM tip. This type of growth behavior can be described as a Stranski-Krastanov mode, where layer-by-layer growth occurs below 0.5 ML and 3-D growth occurs above 0.5 ML. This system is slightly unusual in that the transition to 3-D growth occurs before the completion of the first monolayer of metal deposition.

Annealed "Bow Tie" Structures

When low coverages of Al are annealed on the Si(100) surface, structures develop which are similar to the "bow tie" structures previously observed for the annealed In:Si(100) system.⁹ Empty state images of the bow tie structure show two maxima separated by roughly two unit cells along the direction of the Si dimer rows. In the filled state images, there is one central maxima positioned between the two empty state maxima. Although we believe each bow tie is associated with six metal atoms, it is difficult to assign a model for these structures because of the strong asymmetry in the empty and filled state images. In the case of the In:Si(100) system, these structures order into rows at higher coverages and form the In(4×3) phase near 0.5 ML.⁵ The annealed Al:Si(100) surface is known to have a 4×5 reconstruction between 0.3 to 0.5 ML,⁴ but we have been unable to verify whether the bow tie structures are the fundamental unit of this phase due to tunneling problems at higher coverages.

2) The Sn:Si(100) System

The behavior of Sn on the Si(100) surface demonstrates an abundance of surface structures and reconstructions. Although this system behaves very similarly to group-III metal:Si(100) systems near room temperature, the annealed phases of Sn:Si(100) appear much more complicated and interesting. For annealing treatments up to 500°C, Sn forms four reconstructions as follows: $c(4\times 4)$ for 0.2 to 0.375 ML, 2×6 for 0.375 to 0.5 ML, $c(4\times 8)$ for 0.5 to 1.0 ML, 1×5 for 1.0 to 1.5 ML and 1×5 plus streaks above 1.5 ML.¹⁰ A number of distinct structures are encountered on

the Sn:Si(100) surface which are associated with each of its many reconstructions. The origin of these structures, as well as their ordering behavior with coverage, were investigated.

Annealed Behavior for 0.05 to 1.0 ML Sn

The coverage evolution of the annealed Sn:Si(100) surface up to 1 ML essentially includes a progression of three steps: 1) development of an array of "trenches", 2) replacement of the trenches by an array of "stripes" [4×6 local order] and 3) growth of second layer islands consisting of "4-chains" [$c(4 \times 8)$ local order]. We will now examine these three structures and their associated surface reconstructions more closely.

At coverages below 0.2 ML, trenches form on the surface which appear as dark lines oriented perpendicular to the Si dimer rows. Initially, these trenches are spaced relatively far apart at low coverages and do not form very straight or long lines. As the Sn coverage increases, they become more well defined and order into two dimensional arrays with inter-trench spacings of approximately $12a$ to $8a$, where a is the unit cell distance $a = a_0/\sqrt{2} = 3.84\text{\AA}$. The simplest structural description of the trenches involves an arrangement of missing Si dimers and possible substitution of Sn for Si along the trench edges.

At coverages above 0.2 ML, another type of surface structure dominates. These structures appear as bright stripes oriented perpendicular to the Si dimer rows, where the inter-stripe spacing decreases as the coverage increases. At a nominal coverage of 0.6 ML, the stripes ($3a$ width) are spaced approximately $6a$ apart. The filled state maxima of each stripe are positioned antisymmetrically along the left and right sides of the stripe with a $4a$ periodicity. Since the periodicity along the stripes is $4a$, a local ordering of 4×6 results. Previous LEED studies have indicated a 2×6 reconstruction in this coverage regime. Most likely, the $4 \times$ periodicity is not evident in LEED because different regions of 4×6 can be out of phase, and the $4 \times$ periodicity along the stripe is not always well developed.

When the coverage is increased further, the surface structure becomes increasingly more complex. Islands form which consist of trenches spaced $4a$ apart with chain-like rows located

between them. These linked structures are referred to as "4-chains", where "4" refers to the chain width. The periodicity along each 4-chain is also $4a$. Since adjacent rows of 4-chains are 180° out of phase, a local $c(4\times 8)$ ordering results. This $c(4\times 8)$ arrangement occurs frequently on the sample and corresponds to the $c(4\times 8)$ reconstruction observed at 1 ML in previous LEED studies. A possible structural description for the 4-chains involves the formation of buckled Sn-Sn dimers. In a simplistic model, the surface is assumed to consist entirely of Sn dimers, where one Sn dimer is removed every four unit cells along each of the Sn dimer rows. These areas of missing Sn dimers correspond to the dark trenches observed between the 4-chains in the images. The remaining Sn dimers are then alternately buckled along each dimer row to form rows of 4-chains which are 180° out of phase with each other.

The formation of the trenches and the buckling of the Sn dimers are both manifestations of the stress induced by the Sn overlayer. As the proportion of Sn surface atoms increases, the fraction of buckled dimers increases and the spacing between trenches decreases. In the 1 ML $c(4\times 8)$ phase, trenches are spaced $4a$ apart, which corresponds roughly to the 20% lattice mismatch between grey Sn and Si. The larger size of the Sn atoms induces compressive stress at the surface. Given that the clean Si(100) surface is under compressive stress normal to the dimer direction and under tensile stress along the dimers, the compressive stress from the Sn will be primarily normal to the dimers. The formation of trenches is a mechanism to relieve this stress. The buckling of the Sn dimers might also play a role in reducing the surface stress. For clean Si(100), the compressive surface stress normal to the dimers is over two times larger for unbuckled, versus buckled, dimers.¹¹

Annealed Behavior for 1.0 to 1.5 ML Sn

After 1 ML Sn has been deposited and annealed, the $c(4\times 8)$ reconstruction completely covers the surface. Higher depositions of Sn result in the growth of bright features over the $c(4\times 8)$ phase. Near 1.5 ML, these features are arranged into rows which are roughly $5a$ apart and are separated by trenches. Each 1×5 row usually consists of two bright features which are spaced $1a$ or $2a$ apart

along the length of the row. The random positions of these features prevent the development of any global periodicity along the direction of the rows. Therefore, the only global ordering is the $5a$ periodicity perpendicular to the rows. This surface arrangement is consistent with the 1×5 reconstruction observed by LEED for 1 to 1.5 ML Sn.

The most fundamental unit of the 1×5 structure corresponds to a pair of evenly spaced maxima in the empty states, and one central maximum in the filled states which lies either in the center ("symmetric") or at the end ("buckled") of the paired empty state maxima. This arrangement of empty and filled states strongly suggests symmetric or buckled Sn-Sn dimers, where the empty state maxima indicate the positions of the Sn atoms. Coverage calculations also support the formation of dimers for the 1×5 reconstruction. This phase can be viewed as a 0.5 ML layer of Sn dimers dispersed over the full monolayer $c(4\times 8)$ phase. Assuming that the underlying Sn layer is still dimerized, the additional 0.5 ML of dimers has the effect of halving the number of surface dangling bonds.

Annealed Behavior Above 2.0 ML Sn

When more than 2.0 ML Sn is deposited and annealed on the Si(100) surface, large rows oriented along $\langle 011 \rangle$ directions (parallel to the original Si dimer row directions) develop. These rows are typically 15 to 25 Å high, are spaced 50 to 80 Å apart, and have a triangular cross section with sidewalls sloped at $\theta = 28^\circ \pm 4^\circ$ from the surface normal. By measuring the specular reflection from these sidewalls using LEED, we obtain a more precise value of $\theta = 25.3^\circ \pm 1^\circ$. Simple calculations show that these rows are facets which result from a gross rearrangement of the Si(100) surface. Terraces can be observed along the facets which have a width separation of $x = 5.6 \pm 0.3$ Å in the Si(100) plane. The most likely facet structures consistent with our data are $\{311\}$ planes, which intersect the (100) surface plane along $\langle 011 \rangle$ axes, have an angle of 25.2° to the surface normal, and have a projected width in the (100) plane of $x = 5.76$ Å for the 1×1 unit cell.

Presently, we do not know if the facetting behavior of the Sn:Si(100) system is dominated by an equilibrium state or kinetics. It is possible that the surface free energy is lowered by the substitution of Sn into the first few layers of {311} facet planes, versus incorporation of Sn into the original Si(100) surface. The clean Si(311) surface has a 3×2 reconstruction, with the $3\times$ periodicity oriented along a $\langle 011 \rangle$ axis, i.e. parallel to the edge of a faceted row.^{12, 13} The component of the $2\times$ periodicity which is perpendicular to the $\langle 011 \rangle$ axis has a projection onto the (100) plane equal to $3\sqrt{2} a_0 = 11.5 \text{ \AA}$. This value corresponds to the spacing between terraces on the facets and is approximately double the value seen in our STM images. Our result implies that the {311} facets observed for the Sn:Si(100) system are unreconstructed, indicating that Sn could in fact be disrupting the expected clean reconstruction.

In conclusion, we have studied the growth of up to a few monolayers of Sn on the Si(100) surface. Up to 1 ML, the surface typically evolves as follows: 1) development of an array of trenches; 2) formation of stripes $[4\times 6]$; and 3) growth of 4-chains at 1 ML $[c(4\times 8)]$. Beyond 1 ML, we observe the growth of bright overlayer features on the $c(4\times 8)$ phase, evolving into the 1×5 phase near 1.5 ML. Models for some of the structures involved in this growth evolution have been proposed where possible. In spite of the complexity of these structures, we believe that many of them can be simply modeled as Sn-Sn dimers. Above approximately 2 ML, the surface undergoes a gross rearrangement and forms {311} facets.

3) The Yb:Si(111) System

Ytterbium (Yb) forms several ordered reconstructions on the Si(111) surface. The sequence of phases with increasing coverage is 3×1 , 5×1 , 7×1 , and 2×1 , with the 2×1 phase corresponding to 0.5 ML Yb.¹⁴ The 3×1 phase appears as rows of bright protrusions oriented along $\langle \bar{1}10 \rangle$ directions with a three unit cell ($3a$, where $a = a_0/\sqrt{2} = 3.84 \text{ \AA}$) inter-row spacing. The protrusions along each row are separated by $2a$, corresponding to a $1/6$ ML areal density. Since these protrusions are oftentimes not in phase between adjacent rows, no global $2\times$ periodicity occurs, consistent with the observed 3×1 LEED pattern.

Both the relative height and registration of the 3×1 maxima with respect to the clean Si(111) 7×7 surface have been determined. We measured the height of the 3×1 maxima relative to the 7×7 adatom maxima to be $1.1 \pm 0.2 \text{ \AA}$ above ($1.9 \pm 0.3 \text{ \AA}$ below) the lower (upper) 7×7 terrace along a step edge. Using ion scattering spectroscopy, Kofoed *et al.* found that the height of the Yb atoms relative to the substrate top-layer was $1.9 \pm 0.3 \text{ \AA}$.¹⁴ Given that the Si adatoms lie approximately $1.2 \text{ \AA}$ above the substrate top-layer,¹⁵ our STM data indicates that the height of the Yb atoms relative to the substrate top-layer of the lower terrace is $2.3 \pm 0.2 \text{ \AA}$, consistent with Kofoed's value. The registration of the 3×1 maxima indicates positions equivalent to the adatom maxima of the lower 7×7 terrace. Since the 7×7 adatoms are located on T4 sites⁸, the 3×1 maxima should also lie on T4 sites, assuming that the layer directly beneath the 3×1 phase is not reconstructed. On the basis of the height and registration of the 3×1 maxima, we suggest a simplistic model for the 3×1 phase which assigns each 3×1 maximum to a Yb adatom in a T4 site, corresponding to $1/6$ ML coverage.

As the coverage is increased above that of the 3×1 phase, the higher coverage " $n\times 1$ " phases (i.e. 5×1 , 7×1 , and 2×1) develop. These phases consist of rows that appear different in the STM images from those associated with the 3×1 phase. The " $n\times 1$ " rows are also oriented along $\langle \bar{1}10 \rangle$ directions, but they are not composed of bright protrusions and can pack with a $2a$ inter-row spacing. In addition, the $n\times 1$ rows have a relatively well-defined $1a$ periodicity along each row. This $1a$ periodicity, in contrast to the $2a$ periodicity of the 3×1 rows, indicates that the $n\times 1$ and 3×1 rows are intrinsically different. At low coverages, the majority of the surface consists of 3×1 rows. As the coverage is increased, pairs of the $n\times 1$ rows with a $2a$ inter-row spacing occur. Local regions of the 5×1 phase form when the $n\times 1$ rows are spaced alternately $2a$ ("pairing") and $3a$ apart, resulting in a $5a$ periodicity. As the coverage continues to increase, the 3×1 rows disappear entirely and the $n\times 1$ rows begin to pack more densely, forming "triplets" as well as pairs. Analogous to the pairs of $n\times 1$ rows, the triplets consist of three $n\times 1$ rows which have a $2a$ inter-row spacing. Local regions of the 7×1 phase form when these triplets of $n\times 1$ rows occur next to each other. Close to 0.5 ML, the $n\times 1$ rows reach their maximum packing density with all rows

separated by a $2a$ inter-row spacing. The evolution of the $n \times 1$ phases thus concludes near 0.5 ML with the 2×1 reconstruction.

In conclusion, the Yb:Si(111) system forms two types of reconstructions up to 0.5 ML: (1) a lower coverage 3×1 phase and (2) higher coverage " $n \times 1$ " phases (5×1 , 7×1 , 2×1). The 3×1 phase consists of 3×1 rows which have a $3a$ inter-row spacing and bright protrusions with a $2a$ periodicity along each row. We believe these protrusions correspond to Yb adatoms which are located in T4 sites. The $n \times 1$ phases consist of $n \times 1$ rows which have a $2a$ or $3a$ inter-row spacing and a weak unit cell periodicity along each row. Various packing arrangements of the $n \times 1$ rows result in 5×1 , 7×1 , or 2×1 local ordering.

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