Au-Induced Nanostructuring of Vicinal Si Surfaces

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Abstract. We have investigated the Au–induced nanofaceting of vicinal silicon surfaces tilted from [111] towards [112]. Using samples miscut 3.8°, and 8°, from [111] respectively we have used scanning tunnelling microscopy (STM) to measure the surface morphology as a function of Au coverage (0.04 to 0.44 ML). As expected, Au adsorption produces dramatic changes in surface morphology on both samples. On the 3.8° sample we find that as little as 0.04 ML of Au is sufficient to remove the faceting present on the clean surface. As more Au is deposited 1–dimensional chain structures nucleate at step edges. These chain structures eventually grow to form (775)-Au facets. At 0.17 ML we observe a surface with Si(111)7x7 terraces and (775)–Au nanofacets. With more Au, the (111) terraces transform from a 7x7 to a 5x2 reconstruction and at 0.4 ML the sample consists of Si(111)5x2–Au terraces separated by (775)-Au facets. The morphology of the 8° sample also depends critically on Au coverage. Below 0.32 ML all 8° surfaces include (775)-Au nanofacets. Above 0.32 ML, the (775)–Au facet is no longer stable and Au is accommodated on the surface via the formation of higher angle facets with smaller chain spacing. In both samples, the persistence of the (775)-Au facet reinforces the idea that it represents a low energy facet on these Au modified vicinal surfaces.

1. Introduction

Vicinal Si surfaces have attracted significant interest since they represent natural substrates for the self-assembly of one–dimensional (1–d) nanostructures. The deposition of extremely small amounts of metal onto vicinal semiconductor surfaces can cause dramatic changes in surface morphology on a nanometer scale. Recently, Au deposition onto Si(111) samples tilted either towards or away from the [112] direction has been exploited to produce arrays of atomic chains that exhibit bands with intriguing 1-d metallic behavior [1].

Several groups have investigated the Au–induced morphology of Si surfaces miscut towards [112] [2-4]. In particular, Seehofer et al. [2] deposited 0.44 ML of Au onto samples tilted 2, 4 and 9° from [111], while Shibata et al. [3] deposited 0.3 ML onto a sample tilted 4° towards [112]. Hild et al. [4], measured the Au–induced faceting of a 4.3° sample as a function of coverage. These results suggested that at low coverage (less than 0.45 ML) Au adsorption produces bunched steps which form (775) facets oriented 8.5° from [111]. In this article we expand on these studies to investigate the Au-induced nanofacetting of two silicon surfaces tilted 3.8° and 8° from [111] towards [112] respectively.

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Using scanning tunneling microscopy (STM), we have measured the surface morphology for Au coverages ranging from 0.04 ML up to 0.44 ML. We find that the surface morphology of both samples is exquisitely sensitive to Au coverage. Although the details of the nanofaceting for the two samples are different, they do have important similarities. In agreement with previous results, both surfaces tend to form (775)–Au facets and thus provides further evidence that the (775) represents a low energy facet on these Au–induced vicinal surfaces.

2. Experimental Details

The 3.8°, and 8° samples were resistively heated and the current was applied parallel to the step edges ([110] direction) to avoid electromigration effects. Clean Si surfaces were prepared using a procedure outlined by Lin et al. [5]. Au was deposited from a tungsten basket. During Au deposition the sample was held at 650°C. Following deposition, the sample was annealed at 830°C for 2 minutes.

The Au evaporator was calibrated against the Si(111)5x2-Au reconstruction which occurs at a Au coverage of 0.44 ML (1 ML = 7.8 x 10¹⁴ atoms/cm² [6]). Using Low energy electron diffraction (LEED), we optimized the 5x2-Au reconstruction and correlated this against the Auger electron spectroscopy (AES) ratio between the Si (96eV) and Au (74eV) peak heights. The minimum amount of Au that we could measure on the surface using AES was 0.03 ML.

LEED and AES measurements were performed immediately following annealing, however to reduce thermal drift the samples are allowed to cool for at least 1 hour prior to STM measurements. STM images were obtained using an Omicron MicroSPM [7] and RHK [8] control electronics. All STM images shown in this paper were obtained at room temperature.

Figure 1. 5000 Å x 3570 Å image of the 3.8° sample. Slowly cooling through the transition produces a regular array of bunched steps separated by flat (111) terraces. The image was obtained at a sample bias of 2.3 V and a current of 0.3 nA.

Figure 2. 5000 Å x 5000 Å image following deposition of 0.04 ML of Au onto the 3.8° sample. The large 7° facets present on clean surface are eliminated. Tunneling conditions: 2.4 V sample bias and a current of 0.5 nA.
3. Results

3.1. Clean Si Surfaces

Step bunching occurs on Si samples miscut towards [11\(\overline{2}\)] as they are cooled through the 1x1–7x7 phase transition. Slowly cooling through this transition produces a regular array of bunched steps separated by (111)7x7 terraces. The bunched steps produce 7° nanofacets (figure 1) on the surface in agreement with others [5].

3.2. Au–induced faceting of 3.8° surfaces

Deposition of as little as 0.04 ML of Au onto our 3.8° samples dramatically alters the surface morphology (figure 2). Compared with the clean surface it is apparent that the regular 7° nanofacets are no longer present. On closer inspection we observe the nucleation of 1-d chains adjacent to step edges (figure 3). With increasing Au coverage these regions grow to form small (775) nanofacets. The (775) facets are oriented 8.5° from [111] towards [11\(\overline{2}\)]. The (775)–Au surface consists of a series of Si(111) terraces separated by single silicon steps. The resulting unit cell thus includes atoms from the step edge [2]. In STM images the surface is characterized by 1-d chains spaced 21.3 Å apart running along the [1\(\overline{1}\)0] direction [1]. The Si(111) terrace width determines the interchain spacing. The chains are imaged as bright lines and defects in the chains are imaged as voids (figure 4). Adatoms are also evident between the chains and are imaged as bright protrusions independent of sample bias. We find that these features are observed on all (775)-Au facets independent of the global Au coverage and sample miscut suggesting that the Au stoichiometry of the (775) facet is constant. Previously, we have determined that the local Au coverage on the (775) nanofacets corresponds to 0.24 ML, or 1.5 Au atoms per (1x1) unit cell [9]. This is in contrast to previous models incorporating two atoms per cell [1, 4].

![Figure 3](image3.png) **Figure 3.** 270 Å x 270 Å image obtained on the 3.8° sample following the deposition of 0.04 ML of Au. The nucleation of chains adjacent to the step edge is evident. Tunneling conditions: 2.1 V sample bias and a current of 0.3 nA.

![Figure 4](image4.png) **Figure 4.** 420 Å x 300 Å image of the Si(775)–Au reconstruction obtained at −2.1 V sample bias and a current of 0.5 nA.
At 0.22 ML the (775) regions are fully developed and the sample surface consists of Au free Si(111)7x7 terraces separated by Au–induced (775) terraces. In other words, the surface has transformed from (111) terraces with 7° nanofacets, to (111) terraces with 8.5° Au–induced nanofacets. On further Au deposition the (111) terraces adopt a 5x2–Au reconstruction. The transition to Si(111)5x2–Au + (775) facets is complete by 0.4 ML (figure 5).

3.3. Au–induced faceting of 8° surfaces
As with the 3.8° samples, we find that the morphology of the 8° sample is exquisitely sensitive to Au coverage and that the (775)-Au facet is observed over a wide range of Au coverage. In fact, the (775) facet is present at the onset of Au deposition and is stable up to 0.32 ML of Au.

As the Au coverage increases, the relative extent of the (775)-Au facets on the surface increase causing a corresponding change in the orientation of the non-(775) facets to maintain the overall miscut of the 8° sample. The orientation of the non-(775) facet moves from an initial (111) orientation towards (775) with increasing Au coverage. For example, at 0.06 ML the non-(775) facets are (111)7x7 terraces, whereas at 0.20 ML they are (11 11 9) facets oriented 5.5° from [111].

In previous experiments [9], we exploited the fact that the off-axis angle of our 8° sample is very close to the angle of the (775) facet (8.5°) in order to determine the stoichiometry of the (775)-Au surface. By maximizing the relative surface area we determined the stoichiometry of the (775) facets to be 0.24 ML, or 1.5 Au atoms per (1x1) unit cell (figure 6).

Up to 0.24 ML the non–(775) facet does not contain Au, however at higher coverage (up to 0.32 ML), the surface morphology consists of (775)-Au and (111)-Au facets. As the Au coverage is further increased, the (775)-Au reconstruction is no longer stable and the extra Au must be accommodated by the formation of higher angle facets with smaller chain spacings. For example, at 0.43 ML the surface consists of two nanofacets with a 7.1° angle between them. The chain spacing on one of the facets is 14.8 Å apart, and consistent with a (553)-Au surface. Given the 7.1° angle, the other facet must be (554).

Figure 5. 700 Å x 430 Å image of a 3.8° sample following deposition of 0.44 ML of Au. The surface exhibits alternating (111)5x2 and (775) nanofacets. The image was obtained at a sample bias of –2.5 V and 0.2 nA.

Figure 6. A 1000 Å x 1000Å image of an 8° sample following the deposition of 0.24 ML of Au. The relative surface area of the (775) facets is optimized. Tunneling conditions: -2.5 V sample bias and a current of 0.2 nA.
4. Discussion
We have shown that the morphology of these vicinal Si surfaces is exquisitely sensitive to Au; 0.04 ML on our 3.8° sample, and 0.06 ML on our 8° sample is sufficient to produce dramatic changes. We also find that Au deposition leads to the formation of (775) facets on both samples.

With increasing Au coverage, the surface of the 3.8° sample transforms from (111) terraces and 6° nanofacets to (111) terraces and 8.5° (775)–Au nanofacets. In agreement with Hild et al. [4] the initial (775) facets are observed with as little as 0.06 ML of Au. Below 0.22 ML the (111) terraces are Au free and (7x7) reconstructed, however with increasing coverage they convert to 5x2–Au. These results are consistent with the earlier experiments which observed (111)7x7 + (775) facets below 0.30 ML, and (111)5x2–Au + (775) facets up to 0.44 ML [2-4]. Shibata et al. [3] argued that the (775) facet is preferred since it amounts to a 5x2–Au reconstruction modified to incorporate a step. The 5x2 is the preferred Au–induced reconstruction on (111) terraces, and thus it is reasoned that (775) should be the preferred on stepped surfaces. Our results confirm this hypothesis and further suggest that initial Au adsorption nucleates (775) facets rather than 5x2–Au regions on otherwise clean (111) terraces. The overall extent of the (775) facets is limited by the miscut of the sample and beyond a critical coverage additional Au nucleates 5x2–Au regions on the (111) terraces.

The evolution of our 8° sample is more complicated [9]. Like the 3.8° sample, Au deposition leads to the formation of (775) facets. However, the (775) nanofacets are accompanied by a series of non-(775) nanofacets which depend on coverage. Initially the extent of the (775)–Au facets grow with increasing Au coverage. To maintain the overall miscut of the sample a change in the orientation of the non-(775) facet is required. Since the local stoichiometry of the (775) nanofacets is 0.24 ML, at a given Au coverage the excess or deficit of Au is accommodated by the secondary nanofacet. Beyond 0.32 ML the (775) facet is no longer stable and Au must be accommodated on nanofacets with shorter chain spacings or higher Au coverage.

5. Conclusions
We have studied the Au–induced nanofaceting of Si samples miscut 3.8 and 8° from [111] towards [110]. On both samples Au deposition leads to the formation of (775) facets. However, the (775) nanofacets are accompanied by a series of non-(775) nanofacets which depend on coverage. Initially the extent of the (775)–Au facets grow with increasing Au coverage. To maintain the overall miscut of the sample a change in the orientation of the non-(775) facet is required. Since the local stoichiometry of the (775) nanofacets is 0.24 ML, at a given Au coverage the excess or deficit of Au is accommodated by the secondary nanofacet. Beyond 0.32 ML the (775) facet is no longer stable and Au must be accommodated on nanofacets with shorter chain spacings or higher Au coverage.

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6. References
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