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# INITIAL STAGES OF METAL/SEMICONDUCTOR INTERFACE FORMATION: Au AND Ag ON Si(111)

by

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We have studied the atomic structures formed by monolayer coverages of Au and Ag on the Si(111) surface using primarily the technique of the Impact-Collision Ion Scattering Spectroscopy (ICISS). For the case of Au films annealed at 700°C, three different types of LEED patterns are formed depending on the fractional monolayer coverage: 5×1, $\sqrt{3} \times \sqrt{3}$, and 6×6. The ICISS data reveal that all the three surfaces are structurally similar: the Au atoms reside above the Si(111) plane, most likely in threefold-hollow sites, and the different surfaces appear to be characterized by rows (5×1) or a honeycomb network ($\sqrt{3} \times \sqrt{3}$ and 6×6). In constrast, the Ag films deposited at elevated substrate temperature (480°C) display only a $\sqrt{3} \times \sqrt{3}$ LEED pattern for coverages ranging from 0.25 to 35 monolayers. A trimer model appears to be more consistent with the low-coverage Ag ICISS data rather than a honeycomb arrangement of the Ag atoms.

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## Abstract

We have studied the atomic structures formed by monolayer coverages of Au and Ag on the Si(111) surface using primarily the technique of Impact-Collision Ion Scattering Spectroscopy (ICISS). For the case of Au films annealed at 700° C, three different types of LEED patterns are formed depending on the fractional monolayer coverage: 5x1, $\sqrt{3}$x$\sqrt{3}$, and 6x6. The ICISS data reveal that all the three surfaces are structurally similar: the Au atoms reside above the Si(111) plane, most likely in threefold-hollow sites, and the different surfaces appear to be characterized by rows (5x1) or a honeycomb network ($\sqrt{3}$x$\sqrt{3}$ and 6x6). In contrast, the Ag films deposited at elevated substrate temperature (480° C) display only a $\sqrt{3}$x$\sqrt{3}$ LEED pattern for coverages ranging from 0.25 to 35 monolayers. A trimer model appears to be more consistent with the low coverage Ag ICISS data rather than a honeycomb arrangement of the Ag atoms.

## 1. Introduction

The reconstructions induced on the Si(111) surface by the noble metals Au and Ag have been the subjects of many scientific investigations for a great many years [1,2], but as yet there is no consensus on their detailed atomic structures. If anything, the number of proposed structural models has increased in proportion to the number of studies. This is principally because each of the currently utilized surface structure techniques is sensitive to a different aspect of the surface structure. One might think that the most direct real-space technique, Scanning Tunneling Microscopy (STM), would resolve any ambiguities. However, as demonstrated last year by two consecutive papers in Physical Review Letters on the Ag-induced $\sqrt{3}x\sqrt{3}$ Si(111) surface [3,4], STM is not yet capable of elemental recognition. Although both papers showed essentially identical honeycomb patterns, one group concluded that the STM features were caused by Si adatoms atop Ag trimers [3] and the other decided that the STM features were individual Ag adatoms [4]. A recent X-ray diffraction study concluded that this surface was characterized by Ag trimers, but without Si adatoms above the trimers [5]. An STM study of a surface that might be structurally similar, that is the Au-induced $\sqrt{3}x\sqrt{3}$ Si(111) surface, revealed triangular shaped features that were interpreted to be Au trimers [6], although later work showed the features to be essentially circular [7]. Our previous Impact-Collision Ion Scattering Spectroscopy (ICISS) [8] studies of Au:$\sqrt{3}x\sqrt{3}$ Si(111) were shown to be consistent with a honeycomb structure [9].

Ion scattering techniques do not directly image a surface, but they do have several advantages for structural analysis: the data provide real-space information (*i.e.* no diffraction), the scattering features are determined by the nuclear positions (not the valence electrons), and the scattered-ion energy depends on the mass of the scattering atom (elemental specificity). In this paper, we will review the bases of our conclusions regarding the structure of the Au-induced Si(111) reconstructions and argue that a honeycomb model is consistent with the published STM images [6]. We also present ICISS data for the Ag-induced $\sqrt{3}x\sqrt{3}$ Si(111) surface, and conclude that the local atomic structures of the Au- and Ag-covered Si(111) surfaces are probably different.

The ICISS data from the Ag covered surface is more consistent with a trimer arrangement [5] than a honeycomb, but the latter cannot be strictly ruled out based on our data alone.

## II. Experimental Procedure

The substrates used in this study were mirror-smooth Si(111) wafers measuring 1.5x0.7 cm. The surfaces were prepared by resistively heating the samples in an ultra-high vacuum chamber to alternately anneal and flash them to 1050° C until sharp 7x7 LEED patterns were observed. No surface contaminates could be detected by Auger spectroscopy. The noble metal films were deposited by evaporating either Au or Ag from a tungsten filament onto a room temperature substrate and then annealing to 700° C (Au) or onto a substrate maintained at 480° C (Ag). The deposition thickness in monolayers (ML) was monitored with a quartz crystal oscillator. The Au films were much more stable than the Ag films, since the Au could not be removed by heating, but the Ag could be completely removed to recover a clean 7x7 surface by annealing to 800° C.

The ICISS experiments were performed as described previously [9,10] using 5 keV Li ions as projectiles and a constant scattering angle of 157° (Au) or 155° (Ag). No ion-bombardment damage was observed for the Li ion dose required to accumulate an ICISS scan from the Au-covered surfaces, but some systematic reduction of the scattering intensity did occur for the Ag-covered surfaces.

## III. Results and Discussion

The ICISS data for scattering from the Au adatoms of the three Au-covered surfaces that produced the sharpest LEED spots (and thus presumably the most well ordered reconstructions) are shown in Fig. 1 for the 5x1 (0.4 ML), $\sqrt{3}$x$\sqrt{3}$ (0.8 ML), and 6x6 (1.1 ML) reconstructions. The data for the different surfaces are all similar, which implies that the local atomic structures of the three reconstructions are similar. Each ICISS scan reveals an intense surface flux peak, with no intensity modulations at higher polar angles. This is firm evidence that for all three surfaces the Au atoms reside above the outermost Si plane, for otherwise there should be orientations at which Si could block incident Li ions from hitting Au atoms and cause a local minimum in the

scattering yield. The ICISS data along the four major azimuths in the Si(111) plane, that is the $[1\bar{1}0]$, $[\bar{1}10]$, $[11\bar{2}]$, and $[\bar{1}\bar{1}2]$, are consistent with the only model so far proposed for the 5x1 surface [1], as reported previously [10], so this reconstruction will not be discussed further.

For the cases of the $\sqrt{3}$x$\sqrt{3}$ and 6x6 reconstructions, many variations of all the previously proposed models have been investigated [9] by comparing the experimentally determined ICISS angular distributions to computer simulations [11]. The models most consistent with the experimental data were the honeycomb model shown in Fig. 2a for the $\sqrt{3}$x$\sqrt{3}$ surface and the centered-hexagon structure (a honeycomb with an additional Au atom in the center of each hexagon but displaced vertically downward from the honeycomb plane by 0.3 Å) for the 6x6 surface. Calculated ICISS distributions for these models are shown as dashed lines in Fig. 1. Although the agreement between the model calculations and the experimental data was reasonable, it could be improved, as shown by the solid lines of Fig. 1, by considering each surface to be a combination of both the honeycomb and centered-hexagon structures. Such a model is reasonable for the $\sqrt{3}$x$\sqrt{3}$ surface, since an ideal honeycomb structure would consist of 0.67 ML Au, whereas the experimentally determined Au coverage was 0.8 ML. Placing the equivalent of 0.13 ML of Au atoms in the centers of the hexagons and displacing them downward 0.3 Å significantly improved the agreement between the calculated and experimental ICISS scans. In the case of the 6x6 structure, the agreement between the experimental data and the simulations for the centered-hexagon model was improved by removing the equivalent of 25% of the Au atoms from the centers of the hexagons. An arrangement of centered- and empty-hexagons that would produce a 6x6 LEED pattern is shown in Fig. 2b. Therefore, the experimentally observed Si(111) surface with Au coverages greater than 0.4 ML is probably always a mixture of honeycomb and centered-hexagon subunits (both give rise to a $\sqrt{3}$x$\sqrt{3}$ unit cell), since it is likely that the difference in adsorption energy between the two reconstructions is small [12]. At higher coverages, long range order in the arrangement of the empty hexagons, reminiscent of the 7x7 structure of the clean Si surface, produces the 6x6 structure. The fact that the STM images of this surface did not show a honeycomb pattern [6] may be the result of the inability of the STM to resolve Au-atom pairs

along the <110> azimuths (the short Au-Au distance in the honeycomb). Analysis of 5 keV Li scattered from Si atoms for the $\sqrt{3}x\sqrt{3}$ and 6x6 structures [9] shows the threefold-hollow site to be preferred over the threefold-atop for Au adsorption.

The ICISS data for 5 keV Li ions scattered from Ag atoms of the Ag-induced Si(111) $\sqrt{3}x\sqrt{3}$ surface looks considerably different from that for the corresponding Au-covered surface along the $[\bar{1}10]$ azimuth, but data along the $[11\bar{2}]$ azimuth is similar, as shown in Fig. 3 for a 0.25 ML coverage of Ag. For higher coverages ($\geq$ 1.0 ML) the flux peaks along both azimuths begin to broaden; the additional scattering trajectories may arise because of the initiation of Ag island formation [1]. The surface flux peak for the Ag-induced $\sqrt{3}x\sqrt{3}$ surface along the $[\bar{1}10]$ azimuth is considerably narrower and does not have the pronounced shoulder seen for the Au-induced reconstruction. Indeed, the Ag $[\bar{1}10]$ ICISS angular distribution is similar to that calculated previously for a Au-trimer model [9]. In addition, the progression in LEED patterns as a function of coverage from 5x1 through $\sqrt{3}x\sqrt{3}$ to 6x6 observed for the Au:Si(111) surfaces were not present in the Ag:Si(111) system. All Ag coverages from 0.25 to 35 ML exhibited a $\sqrt{3}x\sqrt{3}$ LEED pattern. Also, the Ag-covered Si surface was much more sensitive to ion-bombardment and heating than the Au-covered surface.

These observations are evidence that the atomic structures of the Au- and Ag-induced $\sqrt{3}x\sqrt{3}$ reconstructions of Si(111) are structurally different. Figure 3 shows calculations that have been least squares fit to the data for the honeycomb model and the trimer model of Fig. 4, which is based on the chemically intuitive bonding arrangement shown in the inset. This set of Ag ICISS data is only sensitive to the displacements of the Ag atoms relative to each other; the Ag-Si bonding geometry shown was constructed to be consistent with the interplaner distances reported by Takahashi [5] and our chemical intuition. However, Vlieg and van der Veen [13] and Ichimiya *et al.* [14] have presented convincing data that shows that the actual structure is not a simple silyl group termination as shown in the inset of Fig. 4, but probably involves a trimer of Si atoms below each Ag trimer. The agreement with the ICISS data for the trimer model is somewhat better than that for the honeycomb, although the honeycomb model cannot be completely ruled out based

on our data alone. The 180° backscattering ICISS data of Aono *et al.* [15] appears to distinguish more clearly between the honeycomb and trimer models, which suggests that ICISS data collected at different scattering angles can resolve ambiguities that may arise for a particular experimental orientation. At the present time, ICISS data collected along other azimuths and also with He ions as the projectile are being analyzed to better distinguish between the two different types of models for the Ag-induced reconstruction.

## IV. Conclusions

The ICISS data for the Au- and Ag-induced reconstructions of Si(111) show conclusively that in both cases the metal atoms reside above the outermost Si layer, and that there are no Si adatoms above the metal atoms. For the Au covered surfaces, the ICISS data are consistent with primarily honeycomb and centered-hexagon models for the $\sqrt{3}x\sqrt{3}$ and 6x6 surfaces, respectively, with the Au adatoms residing above the threefold-hollow sites. However, for the Ag-induced $\sqrt{3}x\sqrt{3}$ surface, the data slightly favor a trimer arrangement of the Ag atoms, with a best fit yielding a Ag-Ag interatomic distance within the silyl group of 4.60 Å. This model is in close agreement with the models of Refs. [5,13-15] for the arrangement of the Ag atoms.

## V. Acknowledgements

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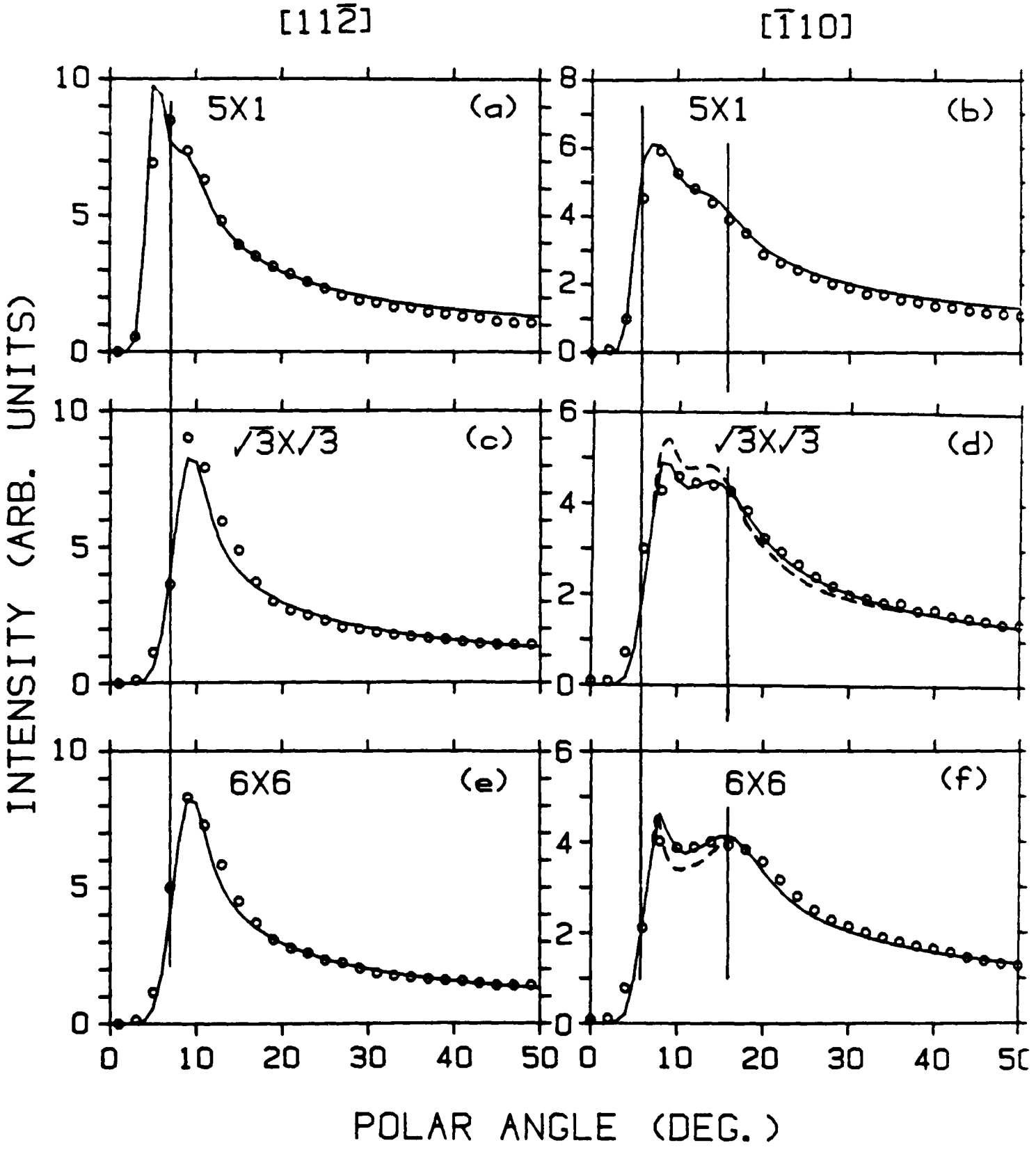
## VII. Figure Captions

1. ICISS angular distributions (detected ion intensity as a function of the angle between the sample surface and the incident ion beam) for 5 keV Li ions scattered at 157° from Au atoms in the 5x1, $\sqrt{3}$x$\sqrt{3}$, and 6x6 reconstructions of Si(111) induced by Au overlayers. The distributions shown are for the $[11\bar{2}]$ and $[\bar{1}10]$ azimuths (the distributions along the $[\bar{1}\bar{1}2]$ and $[1\bar{1}0]$ azimuths, respectively, are essentially identical). The circles indicate the data points, and the lines are the result of calculations for the models described in the text.

2. Drawings of (a) a honeycomb overlayer on Si(111) in which the Au adatoms are located over the threefold-hollows, and (b.) a 6x6 unit cell on Si(111) based on a centered-hexagon array with 25% of the hexagons vacant. Slashed and dotted circles represent Au atoms, and darkly shades circles represent Si.

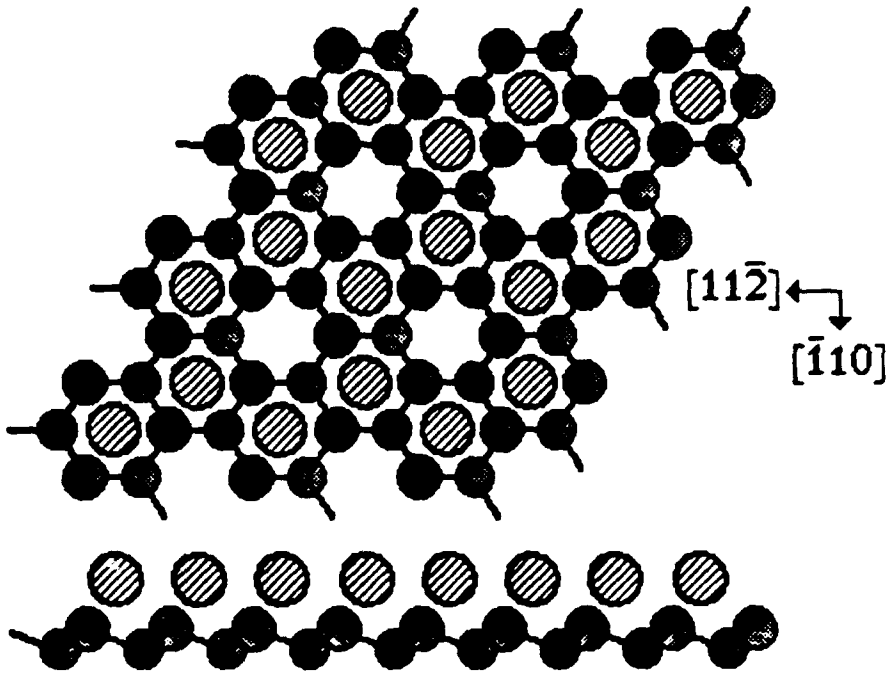
3. ICISS angular distributions for 5 keV Li ions scattered at 155° from the Ag-induced $\sqrt{3}$x$\sqrt{3}$ reconstruction on Si(111). The experimental data (circles) were collected along the $[\bar{1}10]$ and the $[11\bar{2}]$ azimuths, but the scans along the $[1\bar{1}0]$ and $[\bar{1}\bar{1}2]$ azimuths, respectively, were essentially identical. Also shown are best fits (solid lines) to a trimer model in which the surface is terminated with ($SiAg_3$) groups (a, b), and a honeycomb model (c, d).

4. Drawing of the proposed trimer model for the Ag:Si(111)-$\sqrt{3}$x$\sqrt{3}$ surface, with the inset showing the local bonding arrangement that was assumed. Slashed circles represent Ag atoms, and darkly shades circles represent Si.


[11$\bar{2}$]
[$\bar{1}$10]
5X1
(a)
5X1
(b)
$\sqrt{3}$X$\sqrt{3}$
(c)
$\sqrt{3}$X$\sqrt{3}$
(d)
6X6
(e)
6X6
(f)
INTENSITY (ARB. UNITS)
POLAR ANGLE (DEG.)

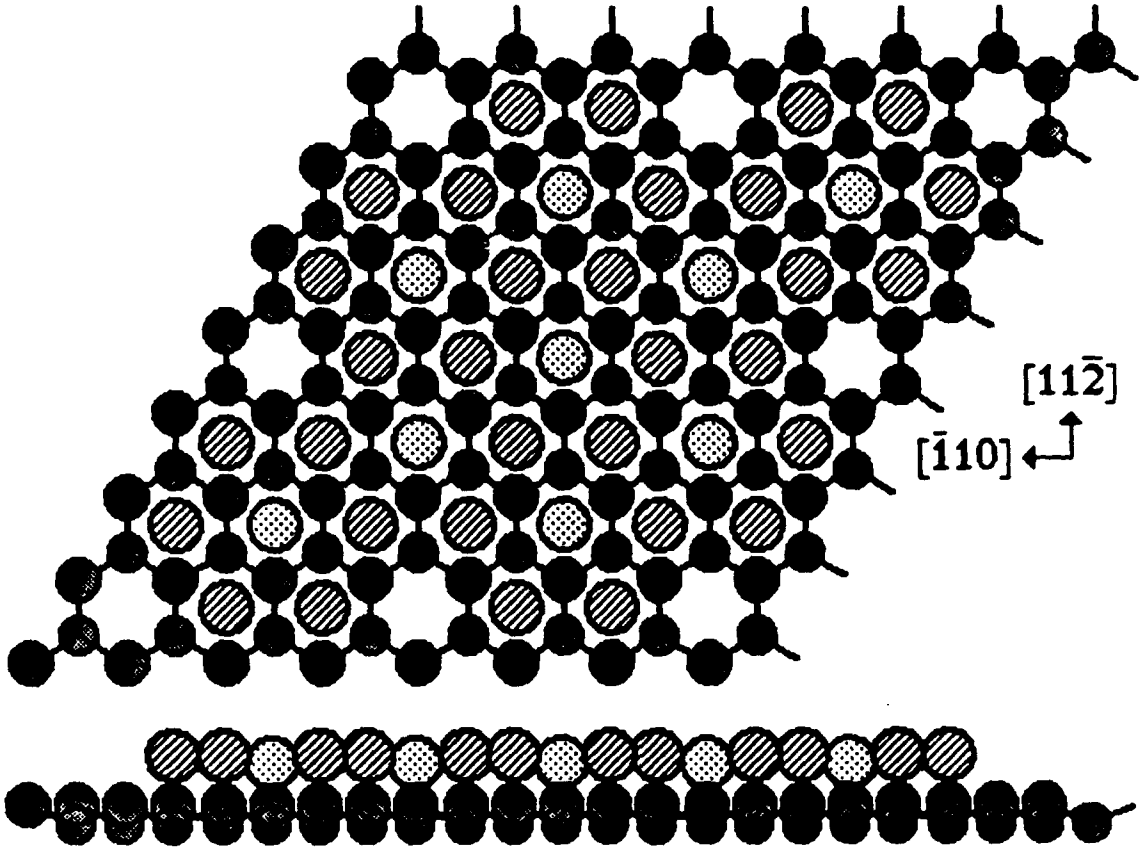
(a)

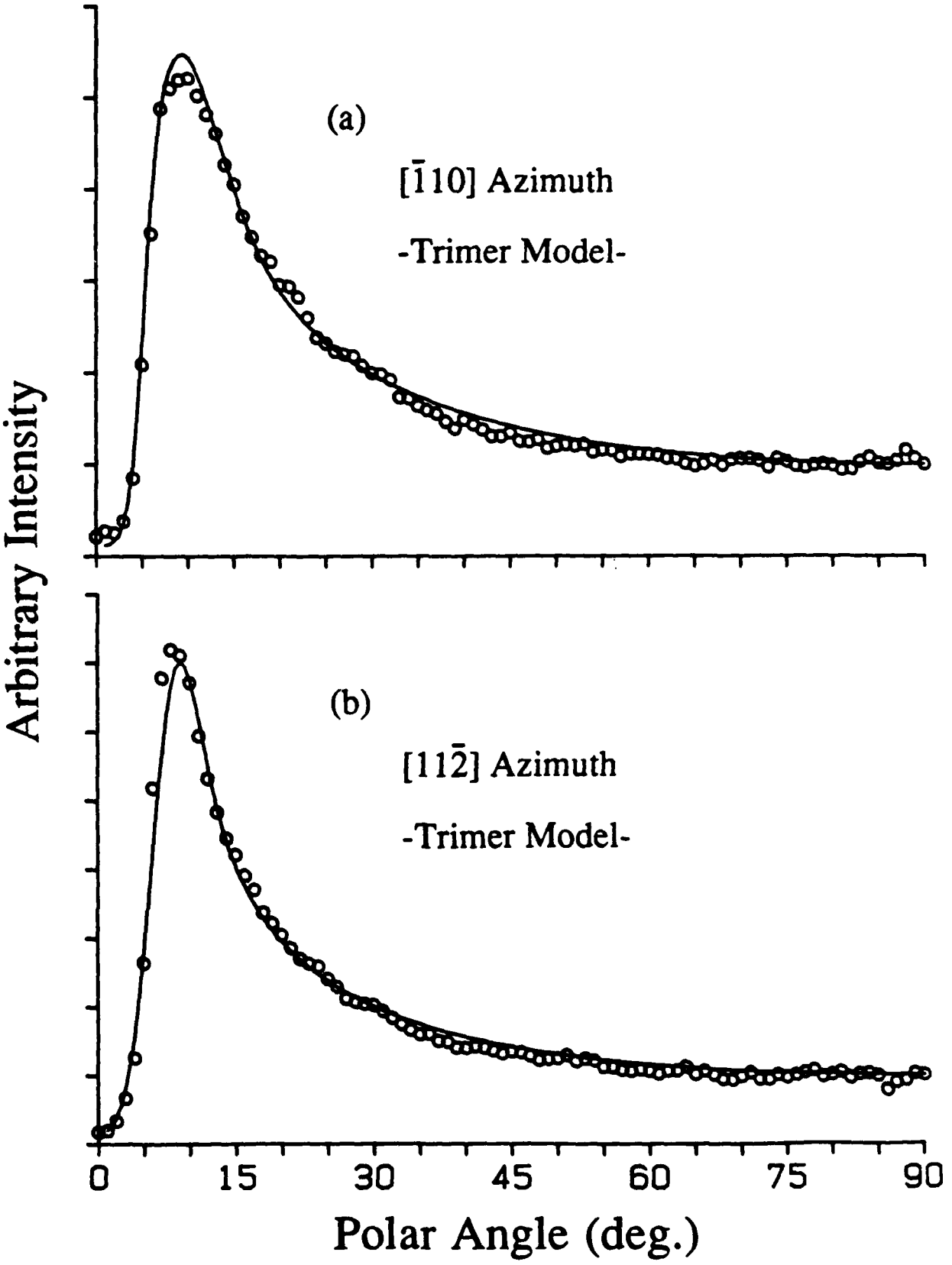
Model for Au-$\sqrt{3}$x$\sqrt{3}$ Surface



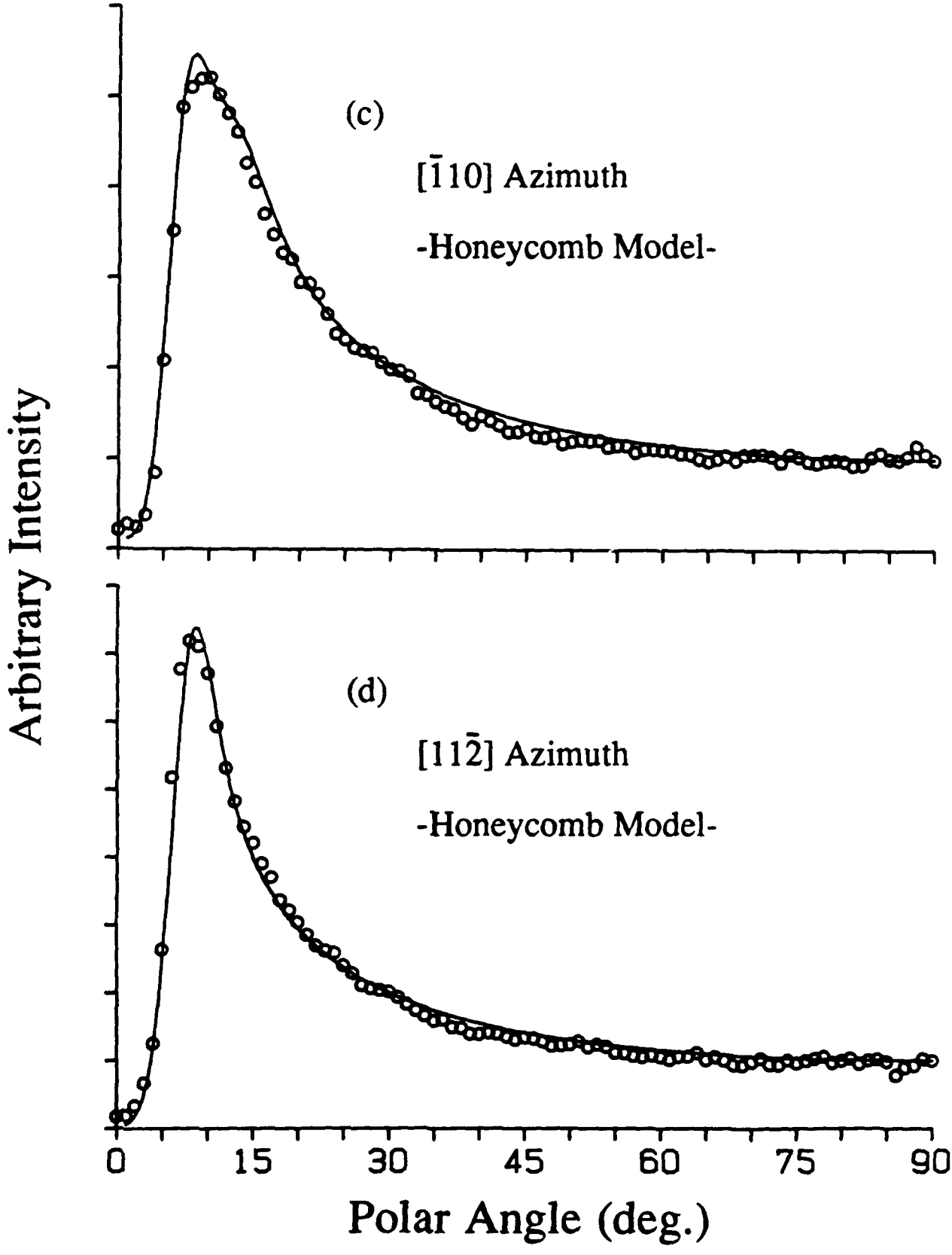
(b)

Model for Au-6x6 Surface

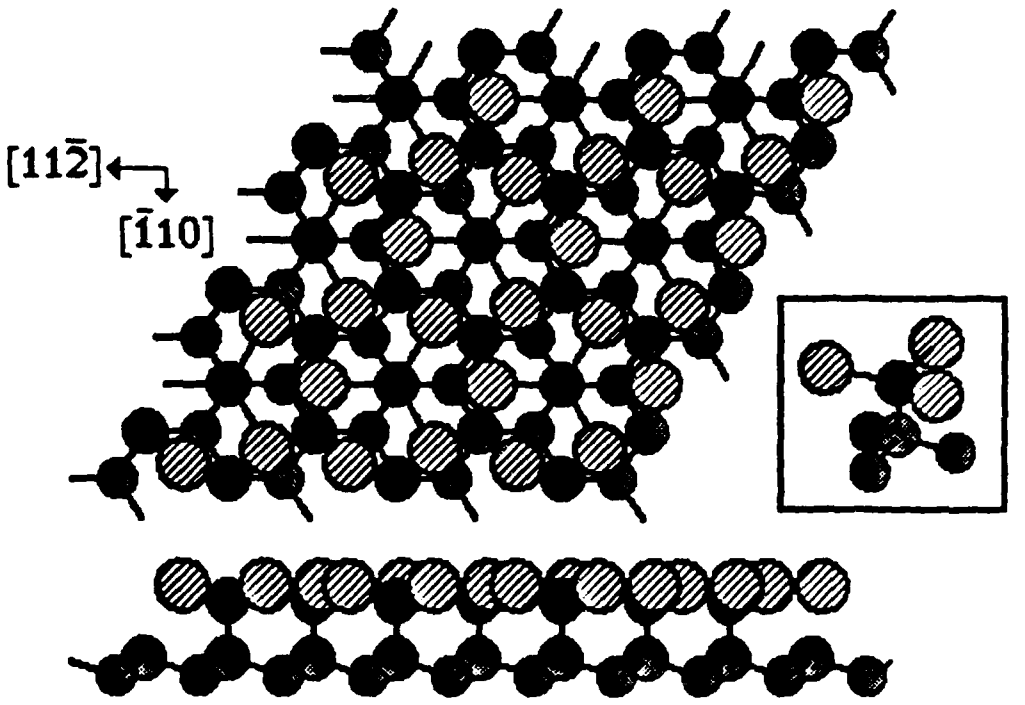



(a)
[$\bar{1}$10] Azimuth
-Trimer Model-
(b)
[11$\bar{2}$] Azimuth
-Trimer Model-
Arbitrary Intensity
0
15
30
45
60
75
90
Polar Angle (deg.)

3a,b


(c)
[$\bar{1}$10] Azimuth
-Honeycomb Model-
(d)
[11$\bar{2}$] Azimuth
-Honeycomb Model-
Arbitrary Intensity
0
15
30
45
60
75
90
Polar Angle (deg.)

# Proposed Model for Ag-$\sqrt{3}$x$\sqrt{3}$ Surface



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