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Brief Reports

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Domain structure of the clean reconstructed Au(110) surface

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We draw together the results of two different types of experiments on the Au(110) surface and show they are mutually consistent. Glancing incidence x-ray diffraction showed surface Bragg peaks displaced along [100], indicating that the surface has an average spacing different from the bulk in that direction. The local structure was found to be a 2×1 "missing-row" morphology. Rutherford-backscattering (He$^+$) measurements of the Au(110)-⟨110⟩ surface peak were significantly larger than expected for a simple missing-row model. Lateral displacements in the second layer of the solid are necessary for agreement with both sets of measurements, and the two derived magnitudes (0.12 Å) are the same. Evidence for a large vertical displacement of the top layer is also obtained by both techniques. The inclusion of a specific domain structure along [100] brings this model into agreement with all experimental evidence.

The ability to determine the atomic structure of a crystal surface is greatly enhanced when different types of experimental data are available. Short-range probes such as surface extended x-ray absorption fine structure and ion scattering determine the local *geometry* of surface atoms and their neighbors; the extraction of a complete structure then requires triangulation of distances which can be ambiguous. Diffraction methods (low-energy-electron diffraction and x-ray scattering) provide primarily long-range information about the surface periodicity; *average* atomic positions are derived by analysis of scattering intensities. When techniques of both kinds are applied to the same problem, not only is there a cross check, but also a new level of information attainable that neither technique alone could provide. We attempt to demonstrate this last point here.

Our x-ray results for the clean reconstructed Au(110) surface have been published previously.[1] We measured 12 integrated superlattice intensities using the glancing incidence x-ray technique[2] in a vacuum system with beryllium windows at 10^{-9} Torr, and four-circle diffractometer. Crystallographic analysis showed the structure to be locally a 2×1 "missing-row" arrangement in which every second [110] chain of top-layer atoms is absent.[1] This settled a debate that had continued for a number of years.[3–5] The missing-row description is now supported by direct surface imaging in the high-resolution transmission electron microscope[6] and electron tunneling microscope.[7] Further[1] refinement of the x-ray data revealed that the second-layer atoms were paired symmetrically by a lateral [100] displacement of 0.12 ± 0.02 Å from bulklike positions, and the surface had an enhanced lateral thermal vibration amplitude of 0.13 Å in the first two layers, compared with 0.084 Å in the bulk at 300 K.

The crystallographic analysis[1] assumed that the measured intensities belonged to half-integral positions in reciprocal space of the form $(h+\frac{1}{2}, k, k)$, h, and k integral. For this reason, the interpretation was implicitly 2×1 with a doubled unit cell along the surface [100] direction and the atomic positions derived were relative to this assumption. All the information about the long-range structure of the surface, however, is contained in the x-ray line shapes and positions. As was originally observed,[1] and as shown in Fig. 1, the actual peak positions were systematically displaced by an amount δ according to the rule

$$(h+\tfrac{1}{2}+\delta,\ k,\ k),\ h+k \text{ odd}\ ,$$

$$(h+\tfrac{1}{2}-\delta,\ k,\ k),\ h+k \text{ even}\ ,$$

where δ is a small positive quantity. Because a sharp peak at a displaced position would indicate an incommensurate surface, δ was called the incommensurability. As δ was found to vary with the sample preparation conditions, and since there appears to be a distribution of values in each case (the line is broader than the instrumental resolution and overlaps the commensurate position), the surface does not have a unique new periodicity and should not be called incommensurate. The shifts of the crystallographically different reflections for a single preparation were all the same, however, in accordance with the rule above. There was no evidence of peaks shifted by multiples of δ or peaks shifted in any other direction, which led to the interpretation of a distribution of localized domain walls perpendicular to [100] separating islands of commensurate 2×1 structure.[1] This picture is also consistent with interruptions of the 2×1 missing-row repeat along [100] seen in tunneling microscope images of Au(110).[7]

High-energy ion channeling measurements (He$^+$) were made on the same Au(110) sample.[8] The energy dependence of the surface peak along the normal [110] direction

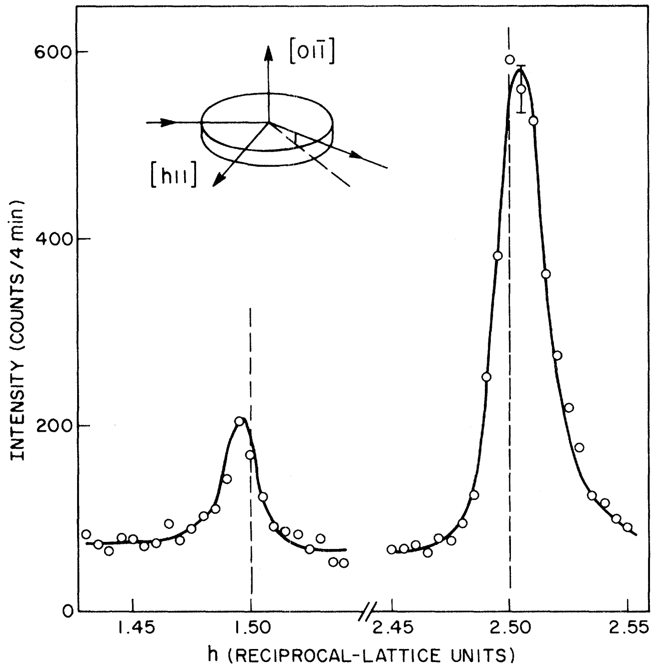

FIG. 1. X-ray diffractometer scans along the [h11] direction, where the absissca h is the fractional reciprocal-lattice coordinate parallel to [100]. The glancing incidence geometry is shown inset. The two profiles shown, corresponding to the $(\frac{3}{2},1,1)$ and $(\frac{5}{2},1,1)$ superlattice reflections, are displaced in opposite directions.

was obtained. The surface peak yield was then calculated by Monte Carlo simulation[9] using the in-plane surface vibration amplitude from the x-ray study[1] assuming an unreconstructed surface. Figure 2(b) shows the difference between the simulated and experimental data (measured at 100 K), showing an excess of measured signal that we attribute to surface reconstruction. In these measurements the shadow cone radius[9] of atoms at the second layer varies from 0.11 Å at high energy to 0.25 Å at the low end of the energy range. An atom displaced laterally by an amount within this range [Fig. 2(a)] would emerge from the cone at a certain point along the energy scan and so explain the step profile observed. The curves superimposed in Fig. 2(b) were generated for a variety of assumed lateral displacements of a whole monolayer of atoms; the best fit to the data yields a value of 0.12 ±0.04 Å for the displacement. The ion scattering measurement provides no direct information about *which atoms* are involved (i.e., first or second layer) or the *direction* of the displacement within the surface plane; however, displacements in the half-filled first layer alone could not explain the data. In view of the excellent agreement, it does seem very likely, however, that this result corresponds to the second-layer pairing displacement seen with x rays.

So far the account for both techniques has been restricted to observations of in-plane atomic rearrangements. X-ray measurements on Au(110) have been reported in which the glancing incidence, glancing exit angle geometry (Fig. 1 insert) is relaxed to allow a normal component to the momentum transfer;[1] considerable variation of scattered x-ray intensity was observed, from which the top-layer spacing was derived. The reported value suggested that the top half-layer was displaced outwards by a large amount. Ion scattering surface peak yields along off-normal directions are also sensitive to such a displacement of atoms from their bulk positions. Measurements were made with the incident beam along the crystal [101] direction;[8] after subtraction of the calculated surface yield of the *reconstructed* surface (i.e., including the lateral displacements and surface-enhanced thermal vibration amplitudes from the x-ray measurements), an extra 0.5 ±0.2 monolayer of scattering remained at all energies. This half-integral value constitutes independent verification that there is a unique half-layer in the reconstruction, an important characteristic of the missing-row model.[5] From shadow cone considerations and the energy dependence of the ion scattering, the magnitude of the normal displacement is greater than 0.25 Å, but there is no information about its sign. A third piece of evidence for a large displacement of the top layer comes from the high-resolution electron microscope images,[6] in which an outward relaxation is clearly seen.

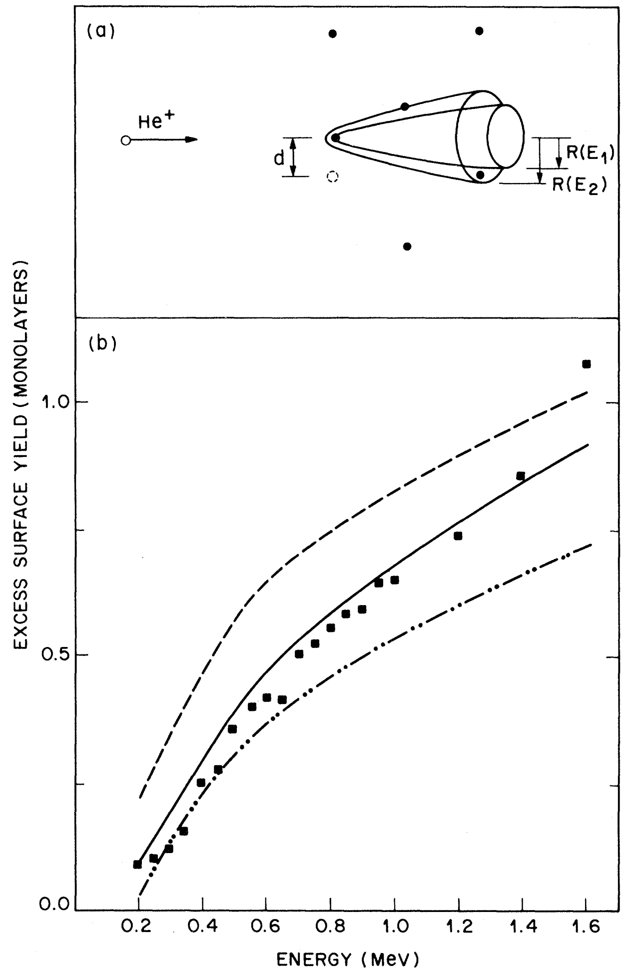

FIG. 2. (a) Schematic illustration of the shadowing of a second-layer atom by a displaced first-layer atom. (b) Energy dependence of the difference between the measured and calculated He$^+$ ion scattering surface peak intensity for Au(110) along the normal ⟨110⟩ direction. Measurements were made at 100 K. Calculations assumed a surface-enhanced vibration amplitude given by the x-ray data and a full monolayer of atoms displaced by 0.10 Å (dot-dash line); 0.12 Å (full line), and 0.15 Å (dashed line).

We now return to the central question of the surface domain structure from the ion scattering viewpoint. A direct consequence of the results in Fig. 2 is that the structure of Au(110) is *locally* commensurate: all the atoms of the displaced (i.e., reconstructed) layers are the same lateral distance away from bulk positions. This follows from the sharpness of the transition in excess surface yield as a function of energy or shadow cone radius. A continuously incommensurate structure with a lattice mismatch between the top layer and the bulk would have only coincidental alignment of atoms and would show an extra monolayer of scattering at *all* energies. The fact that almost all of the top-layer atoms are within ~0.25 Å of lateral alignment with the bulk adds weight to the argument made above and in Ref. 1 that the peak displacements seen in Fig. 1 must be due to a one-dimensional domain-wall array.

A model of the Au(110) surface which reconciles the results of the x-ray, ion scattering, and tunneling microscope experiments is shown in Fig. 3. The model is locally 2×1 as required and uses monatomic steps as domain walls, with all atoms close to bulk locations. The {111} facets exposed are close packed so that this is the most stable step orientation. Such a step is one of four common kinds of defects seen to interrupt the 2×1 regions in electron tunneling images:[7] 3- or 4-unit-cell gaps between adjacent top-layer rows and monatomic or double-height steps (all of which expose {111} facets). Of the four, only monatomic steps can explain the shifted scattering profiles of Fig. 1, as 4-unit-cell gaps do not change the long-range repeat, and 3-unit-cell gaps and double-height steps yield surface antiphase domains that would produce symmetrically broadened or split half-order reflection profiles[10] that we do not observe. The models based on tunneling microscopy[7] have much poorer long-range order than we observe; we would therefore speculate that annealing the surface preferentially removes the 3-unit-cell and higher-order defects to leave a majority of 2-unit-cell repeating units (i.e., 2×1 missing-rows) with occasional steps of the kind described.

The derivation of the scattering profile is also shown schematically in Fig. 3. The result is independent of the actual height of the step (or the amount of top-layer relaxation), except that adjacent domains must scatter in quadrature: a phase slip of $a_0/2$ out of the $2a_0$ repeat achieves this; a phase slip of $3a_0/2$ would shift the lines in the wrong direction. Inclusion of a distribution of domain sizes leads to a predicted profile in which the largest peak is 20 times higher and 3 times sharper than the next highest. This is consistent with the observation of a single shifted peak, given the statistics and background levels prevailing. This argument is analogous to that used to explain the diffraction patterns of magnesium fluorogermanate: occasional plane defects, in which one of four layers in a stacking sequence is omitted, leads to a local contraction of the unit cell and to shifted diffraction peaks.[11]

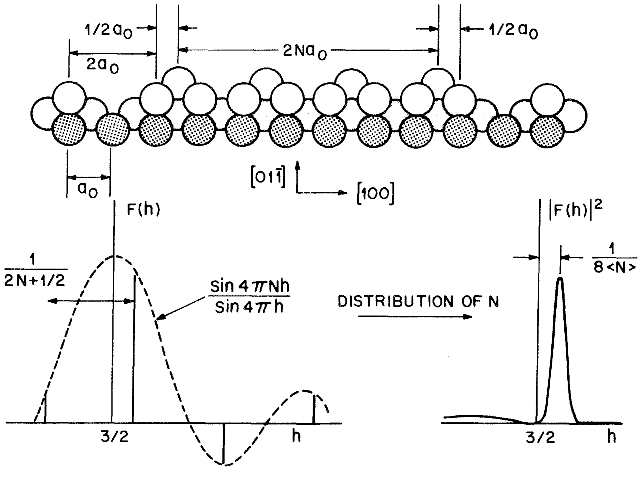

FIG. 3. Model of the Au(110) surface based on single-height atomic steps. The diffraction pattern is constructed as the product of an *N*-slit interference function and the reciprocal lattice of an infinite array of steps.

We have shown how the techniques of x-ray diffraction and ion backscattering are mutually complementary in their probing of long- and short-range order in surfaces. We have seen how results obtained with one technique can greatly strengthen and focus arguments made from results of the other to obtain a clearer understanding of surface reconstruction. Taken together, these experimental findings yield a missing-row model of the Au(110)2×1 surface with second-layer displacements, a first-layer relaxation, and a size distribution of domains bounded by monatomic steps which expose {111} facets.

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