Focusing Effects in Low-Energy Ion Scattering from Single Crystal Surfaces

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The angular distribution of low-energy K^+ ions (250 to 1000 eV) scattered from a W(110) surface is studied as a function of polar angles of incidence and exit for various azimuths with a 127° electrostatic energy analyzer. For specific combinations of polar and azimuthal angles a strong enhancement of the intensity of certain peaks in the energy spectrum is observed which is attributed to focusing of ions scattered in the second layer by the atomic configuration of the first layer.

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Recently Winograd, Garrison, and Harrison[1] have shown that the long-known maxima in the angular distribution of sputtered particles from single crystal surfaces are produced mainly by scattering of energetic ejected particles ($E > 20$ eV) through the holes between the atoms in the topmost layer. This phenomenon opens up the possibility of surface-structure analysis as demonstrated for the case of adsorbed oxygen on a Cu(100) surface.[2,3] In this Letter we report results for the scattering—not secondary emission—of low-energy ions which clearly indicate that in this case the topmost layer of atoms not only constrains the path of the scattered atoms but actually causes a focusing which gives rise to abnormally high scattering intensities.

The experiments were performed in an UHV system with a base pressure in the low 10^{-11}-Torr range equipped with an alkali-ion gun, a noble-gas–ion gun, a 127° electrostatic energy analyzer for the ion-scattering measurements, a cylindrical mirror analyzer for Auger electron spectroscopy (AES), low-energy electron-diffraction optics, and a quadrupole mass spectrometer. The crystal manipulator allowed two rotations of the crystal, one about its normal which determined incident and exit azimuths, the other one about an axis in the plane of the crystal which determined the incident polar angle. The exit polar angle was varied by rotating the 127° analyzer about the second rotation axis of the crystal. A detailed description of the apparatus will be given elsewhere.[4] The W single crystal was oriented to within 0.5° from the [110] direction and cleaned in the usual manner so that the residual carbon contamination was less than 0.2% of a monolayer as determined by AES. During the measurements the pressure rose into the middle to high 10^{-11}-Torr range. This caused a CO contamination of less than 1% of a monolayer during the maximum duration of a measurement (10 min). No potassium contamination due to the primary beam could be detected by AES and ion scattering. Measurements were made at 250-, 600-, and 1200-eV primary energy but only the 600-eV data are discussed here, being representative for all energies studied.

Figure 1 shows three typical spectra taken with 600-eV K^+ ions under specular scattering conditions at three glancing angles of incidence ($\psi = 45°$, 65°, and 75°) in the [$\bar{1}$10] azimuth. The origin of the peaks is determined by comparing the spectra obtained over a wide range of energies and angles with the binary collision model: S and S^* are single scattering peaks, D_1 and D^* are linear double scattering peaks, and D_2 and D_3 are zigzag double scattering peaks as discussed elsewhere.[4] In Fig. 2(a) the measured energies of the peaks (relative to the energy of the incident beam) are plotted for specular reflection in the high-scattering-angle (θ) region for 600-eV K^+ ions incident in the [$\bar{1}$10] azimuth

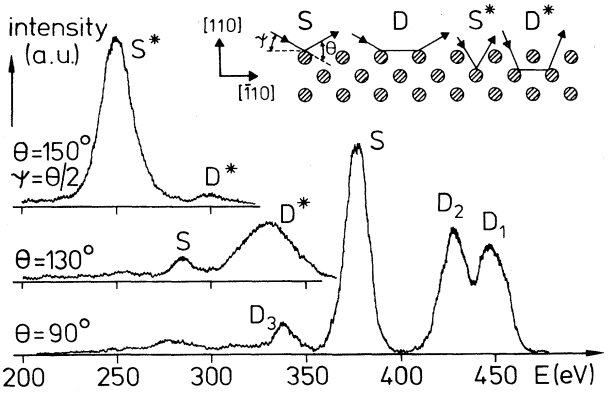

FIG. 1. Typical energy distributions of 600-eV K^+ ions specularly scattered from a clean W(110) surface in the [$\bar{1}$10] azimuth into angles of $\theta = 90°$, 130°, and 150°. The incident current is 7×10^{-11} A. The inset indicates the proposed assignment of the various peaks.

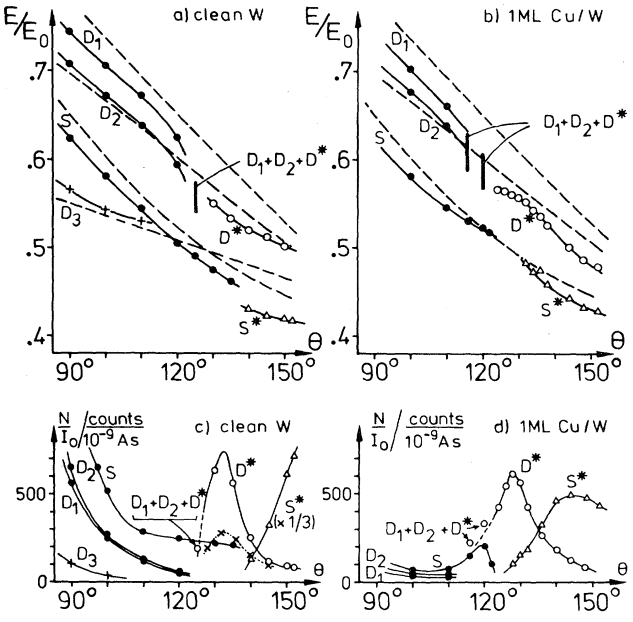

FIG. 2. Specular scattering of 600-eV K^+ ions from a W(110) surface in the $[\bar{1}10]$ azimuth. (a) and (c) Clean surface; (b) and (d) surface covered with one monolayer of Cu. In (a) and (b) the energies E of the scattered ions relative to the energy E_0 of the incident ions are shown as a function of scattering angle. S, S^*, D_1, D_2, D_3, and D^* are the measured energies of the peaks shown in Fig. 1. The dashed lines show the energies calculated from the binary collision model. For the zigzag scattering (D_2, D_3), scattering angle components in the surface plane of 35° and 65° were assumed which are upper limits; for smaller angles the curve is shifted upward. In (c) and (d) the peak heights of the various peaks are shown, normalized to the incident current. The dotted line represents the intensity of the D^* peak in the $[\bar{1}11]$ azimuth. The Cu peaks are not shown in this figure because of their low intensity in the high-θ region. Their energetic positions agree with the binary collision model as well as those of the W peaks.

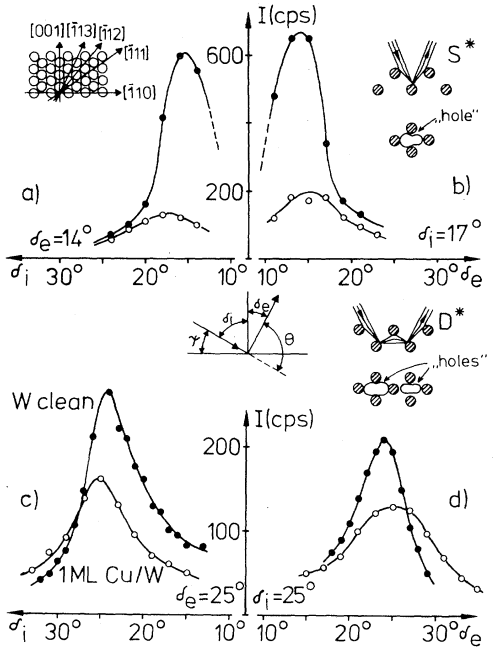

FIG. 3. Polar-angle dependence of the peak heights of [(a) and (b)] the S^* peaks and [(c) and (d)] the D^* peaks for 600-eV K^+ ions scattered from W(110) in the $[\bar{1}10]$ azimuth for a clean surface (solid dots) and a surface covered with one monolayer of Cu (open circles). In (a) and (c) the exit angle was kept constant ($\vartheta_e = 14°$ and 25°, respectively); in (b) and (d) the angle of incidence was kept constant ($\vartheta_i = 17°$ and 25°, respectively). The other polar angle was varied about the specular direction. The insets are referred to in the text.

together with theoretical curves based on the binary collision model. The deviation of the measured from the calculated curves is of the usual order of magnitude (here about 2% at scattering angles below about 120°). The corresponding peak heights of the various peaks are shown in Fig. 2(c). It is evident that the peaks S, D_1, and D_2 decrease rapidly with increasing θ: S decreases up to $\theta = 135°$. At $\theta = 140°$, however, it increases rapidly but is located at a different energy [see Fig. 2(a)]. D_1 and D_2 decrease to very small values at $\theta = 125°$ but then a new peak with similar energy [D^*, see Fig. 2(a)] rises rapidly to a sharp maximum at about $\theta = 132°$ followed by a rapid decrease.

It is the peaks S^* and D^* which are the subject of this Letter. Their energetic position and the scattering angles at which they occur suggest single and double scattering from W atoms in lower layers, predominantly in the second layer as indicated by the insets in Fig. 3. In order to check this hypothesis a monolayer of Cu was adsorbed on the surface. The first monolayer of Cu on W(110) is known to have the same lateral periodicity as the substrate.[5] If the hypothesis is correct then the peaks S, D_1, and D_2—which originate mainly from the top layer—should be reduced considerably in intensity; the peaks S^* and D^*, however, should not be influenced significantly. Figure 2(d) shows the experimental results, confirming the hypothesis. Inspection of Fig. 2(b) indicates that peak D^* has about the same energy position without and with the Cu overlayer while S^* is shifted considerably from the clean-surface value in the presence of the Cu overlayer, indicating a stronger influence of the Cu atoms.

Intensities as high as those of the peaks S^* and D^* in Figs. 2(b) and 2(d) are incompatible with the general behavior of differential scattered-ion yields which are monotonically decreasing with increasing scattering angle except in systems in which near-resonant neutralization processes occur.[6,7] The system K+W does not belong in this class. The high S^* and D^* intensities are also not due to the anomalously enhanced ion backscattering near 180° scattering angle,[8-11] which does not require the regular atomic arrangement necessary for processes observed here. Rather, the phenomenon is caused by the focusing effect which the "holes" between the atoms of the topmost layer (see insets in Fig. 3) exert on the ions incident on and reflected from the atoms of the second layer. This explanation was checked in two ways: (i) by changing the azimuth which changes the focusing possibilities and (ii) by determining the angular dependence of the peaks S^* and D^* in more detail. Regarding (i), no measurable S^* peak could be seen in any of the other azimuths studied {[001], [$\bar{1}$11], [$\bar{1}$12], and [$\bar{1}$13]; see insets in Fig. 3(a)} and a small D^* peak could be seen only in the [$\bar{1}$11] azimuth [see Fig. 2(c)]. By inspection of the inset in Fig. 3 this is immediately plausible within the hole-focusing model. The S^* peak which is attributed to incidence and exit of the ion through the same hole [see Fig. 3(b)] can occur only if the hole is "long" enough and symmetric. The D^* peak requires two close-by holes and two close-by second-layer atoms in the path of the ion [see Fig. 3(d)].

The "walls" of the holes have apparently sufficient focusing effect that the precise alignment of the ion beam with the [$\bar{1}$10] azimuth is not too critical: As a function of azimuth the S^* and D^* peaks have full width at half maximum (FWHM) values of 16° and 13°, respectively, upon specular scattering of 600-eV K^+ ions from the clean surface by $\theta = 150°$ and $\theta = 130°$, respectively.

The polar angle is much more critical, particularly for the S^* peak. This can be seen in Fig. 3, in which either the polar angle of incidence ϑ_i [Figs. 3(b), 3(d)] or the polar angle of exit ϑ_e [Figs. 3(a), 3(c)] was kept constant close to the optimum angle for maximum specular scattering and the corresponding polar angles of exit or incidence were varied. On the clean surface both the S^* and D^* peaks have polar-angle FWHM values of only about 7°, indicating that the focusing phenomenon can occur only within a narrow polar-angle range.

When the surface is covered by one monolayer of Cu the FWHM value of the S^* peak is larger by nearly a factor of 2 but the peak intensity is decreased at the same time. This shows that the focusing conditions are relaxed by the Cu atoms on top of the W surface. In the case of the D^* peak only the distribution on the exit side [Fig. 3(d)] is broadened by the Cu top layer. This is attributed to the fact that the incident-angle and impact-parameter range in which this process can occur is rather limited because the incident beam is rather parallel and monochromatic ($\Delta E < 2$ eV). Therefore the replacement of the W atoms in the topmost layer by Cu has little influence on the focusing conditions and reduces mainly the peak height because of the lower scattering cross section of Cu. On the other hand, on the exit side the ions have a broad angular and energy distribution ($\Delta E \approx 40$ eV) after the last second-layer collision so that the situation is similar to that which produces the S^* peak.

Several features of Fig. 3 should be noted: (1) The maxima of the angular distributions on the exit side [Figs. 3(b), 3(d)] are at smaller angles than on the incident side [Figs. 3(a), 3(c)]. This is due to the fact that the scattered ions have lower energies when they pass through the first layer. Therefore the cross sections of these atoms are larger and the holes are somewhat smaller on the exit side. (2) When the surface is covered with one monolayer of Cu, the maxima are shifted to higher values of ϑ_i, ϑ_e. Cu has smaller cross sections and therefore the holes are larger (longer) than in the case of the clean W surface.

It should be noted that the Cu atoms are not necessarily located in the sites required by the bulk periodicity of the bcc lattice (volume sites). Pairwise interaction calculations actually suggest that the Cu atoms are displaced laterally in the [1$\bar{1}$0] direction so that they have approximately three nearest neighbors (surface site).[12] This could cause similar displacements of the peaks, in particular of the S^* peak. A separation of these two possible effects would require a quantitative analysis based on numerical trajectory calculations with realistic scattering potentials.

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Pressure-Driven Commensurate-Incommensurate Transition in Low-Temperature Submonolayer Krypton on Graphite

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By using D_2 gas as a source of two-dimensional spreading pressure, we have studied the commensurate-incommensurate (C-I) transition in submonolayer Kr on *ZYX* graphite at temperatures near 40 K. High-resolution synchrotron x-ray diffraction results show both hysteresis and C-I phase coexistence, clear signatures of a first-order transition. This agrees with theoretical predictions, but apparently gives different behavior from earlier studies at higher temperatures and coverages.

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When a thin film is deposited on a crystalline substrate, the competition between adsorbate-adsorbate interactions and the adsorbate-substrate lateral periodic potential can produce a structure which is either commensurate with the substrate or incommensurate, depending on the misfit between the substrate and the natural adsorbate lattice spacings. The resultant misfit depends on the temperature and the two-dimensional (spreading) pressure. Although there have been detailed structural studies[1-4] of near monolayer films at elevated temperatures, as well as extensive theoretical work on the nature of the commensurate-incommensurate (C-I) transition, the overall situation is far from being understood. To quote Villain[5]: "Conclusion: either the C-I transition is first order or the hexagonal symmetry is destroyed near the transition. There is no well established confirmation of this theoretical prediction. Krypton monolayers on graphite. . . seem to provide a counterexample. . . it would be interesting to have data at low temperatures which are more accessible to theory." We report such a high-resolution study here.

Two experimental ingredients have been crucial in making this possible. The first is a means of continuously adjusting the surface pressure at submonolayer coverages and at temperatures well below the melting point of the Kr film. This has been done by adding D_2 gas, which is insoluble in the Kr film, and whose spreading pressure can be controlled even at constant temperature by adjusting its bulk vapor pressure. It thus acts as a two-dimensional piston pushing on the Kr film. The second ingredient is the use of the synchrotron source at Hasylab, Hamburg, to provide the intense collimated x-ray beam required for very-high-resolution line-shape studies in the transition region.

Three independent tests on an 0.6-monolayer Kr sample at temperatures near 40 K all point to a first-order C-I transition. These are line-shape studies in the C-I coexistence region and hysteresis loops both as a function of surface