Surface Scattering from W Single Crystals by MeV He^+ Ions

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The surface peak, observed in backscattering-channeling experiments, has been measured as a function of projectile (He^+) energy on a clean W(100) crystal. We show that the intensity of the surface peak is in good agreement with classical models of channeling. Furthermore, we deduce an upper limit of 6% for the relaxation of the W(100) surface.

The backscattered energy spectrum of a beam of MeV ions incident along a major axial direction of a single crystal displays a high-energy peak which represents the interaction of the beam with the surface region of the solid. Recently, several authors have reported measurements of this surface peak,[1] under ultrahigh-vacuum (UHV) conditions, which provide information about the relaxation of Pt [2] and Ni [3] surfaces and the reconstruction of Au [4] surfaces. The interpretation of such measurements in terms of surface structure and dynamics depends on an understanding of the absolute magnitude of the surface peak. In this Letter we report on the ion-beam-energy dependence of the surface-peak intensity in W(100) using He projectiles. We show that the results are in good agreement with classical models of channeling using an enhanced surface-vibration amplitude and deduce an upper limit of 6% for the relaxation of the W surface.

Consider an ideal crystal with a principal axis aligned with an incident ion beam. The surface peak will always include scattering from the first monolayer of atoms. It may include additional contributions from atoms further along the axial string if the transverse thermal vibrations allow them to penetrate outside the shadow cone formed by the small-angle scattering from the first atom(s). Thus one way to characterize the surface-peak intensity is in terms of the ratio ρ/R where ρ is the transverse, two-dimensional rms thermal-vibration amplitude and R is the shadow-cone radius at the second atom. This radius is described by Lindhard[5] and represents the minimum distance between the ion beam and the second of a pair of atoms whose internuclear axis is parallel to the incident projectile direction. For Coulomb scattering

$$R = 2(Z_1 Z_2 e^2 d/E)^{1/2},$$

where Z_1 and Z_2 are the atomic numbers of projectile and target atom, respectively; E is the energy of the projectile, and d is the distance between the atoms. As ρ/R increases contributions to the surface peak from successive atoms in the string will increase. The energy region explored in this report allows us to vary ρ/R from 0.3 to 0.9.

The experiments were carried out in an UHV system which was coupled, via differential pumping, to a 3.75-MeV accelerator, permitting MeV ion-beam measurements to be made under clean surface conditions. The chamber was pumped to a base pressure of 1.5×10^{-10} Torr and contained a retarding grid low-energy electron-diffraction (LEED)–Auger device, a residual gas analyzer, and other apparatus conventionally used for surface characterization and modification. A surface-barrier solid-state detector with ~ 14 keV resolution was used for measuring the energy spectra of backscattered particles.

The single-crystal tungsten samples were typically 0.5×1.0 cm and 75–100 μm thick and were mounted in a UHV goniometer[6] with provisions for resistive heating. The sample temperature was measured with an optical pyrometer. Cleaning and decarburization were accomplished by heating the sample to 1300°K in O_2 at 10^{-6} Torr for several minutes and then flashing to 1800°K in UHV. This cleaning procedure produced a well-defined LEED pattern characteristic of clean tungsten (100) surface. The sample was flashed and allowed to cool before the acquisition of each backscattering spectrum. Small carbon and oxygen signals were present in the Auger scan. By use of systematics and procedures described by Davis *et al.*,[7] these signals correspond to ~5% and 10% of a monolayer, respectively. The apparatus remained in the mid-10^{-10}-Torr range for the 3–5 min required for each spectrum.

Typical energy-analyzed particle spectra for the beam incident in (a) a channeling direction and (b) a nonchanneling direction are shown in Fig. 1. The significant features of such spectra have been discussed before.[8] Of particular concern to this class of experiments is the subtraction of the background under the surface peak. Using Monte Carlo simulations as a guide we subtracted the background in a simple triangular approximation as indicated. The background represents ~8% of the surface peak.

In order to extract the number of monolayers, or surface density, represented by the surface peak it is useful to compare to a nonchanneling spectrum taken with the beam incident in a direction not corresponding to any major crystallographic axis or plane. In this case, we took many different nonchanneling spectra at one energy (2.0 MeV) and averaged them in order to arrive at a resonable value. An example of such a spectrum is shown in Fig. 1. The intensity of the surface peak at other energies was then deduced by comparison with the 2.0-MeV surface-peak value, scaled with the use of the Rutherford scattering formula. Thus we believe the relative values of the surface-peak intensities presented here are quite reliable; the absolute values have an uncertainty associated with the background subtraction (~5%), the value of the averaged nonchanneling spectrum (~5%), and the stopping power of He$^+$ in W at 2.0 MeV (~5%) which is used in the normalization. The latter quantity was taken from the tables of Ziegler and Chu.[9] We have placed an error of ±10% on the quoted surface-peak intensities.

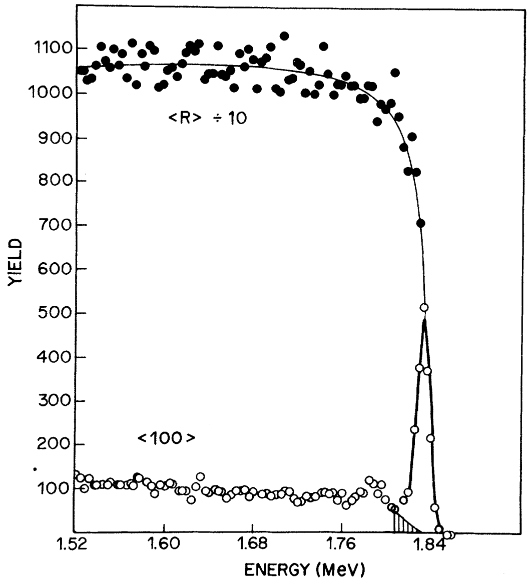

FIG. 1. Energy spectra for 2.0-MeV He incident on W along a ⟨100⟩ axial direction (open circles) and in a nonchanneling direction (closed circles). The method of background subtraction is indicated.

The value of the surface-peak intensity is presented in terms of atoms/string in order to avoid ambiguity in the definition of a monolayer. For example, the W(100) surface exposes two sheets (monolayers) of atoms to the beam, each containing 1×10^{15} atoms/cm^2. Thus a surface-peak intensity of 2×10^{15} atoms/cm^2 corresponds to 1.0 atoms/string.

Figure 2 shows the results for the energy dependence and the axial-direction dependence of the surface peak of W(100) as a function of E/d. In the following, d is the spacing between atoms of the various axial directions (3.16 Å for W⟨100⟩); the scaling with E/d follows naturally from general scattering considerations. The value of ρ_B/R is indicated as well, where we have used a bulk Debye value[10] of 348°K to obtain ρ_B. Also shown are the results of a Monte Carlo simulation of

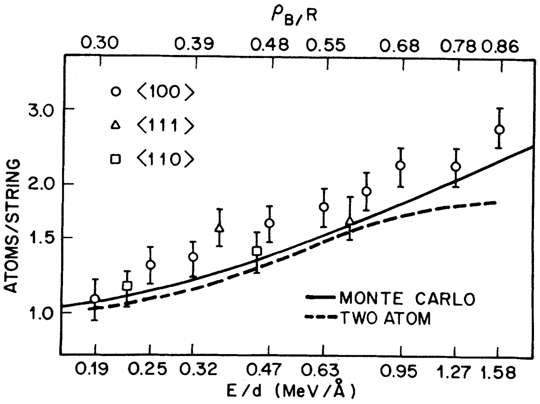

FIG. 2. Intensity of the surface peak vs E/d for various directions in W(100). Results from the Monte Carlo and the two-atom calculation are indicated.

the experiment. The methods used in these Monte Carlo calculations have been previously described.[11] In addition, we have indicated the results of a simple two-atom model for the surface peak. This calculation, based on a Coulomb potential, assumes all particles excluded from the shadow are concentrated in a ring of radius R at the second atom. The contribution from the second atom, I_2, is given by

$$I_2 = e^{-R^2/2\rho^2} + (R^2/2\rho^2)\, e^{-R^2/2\rho^2}.$$

The first term represents the probability that the two atoms have a relative displacement greater than R, allowing the second atom to penetrate the uniform flux. The second term represents the scattering from the enhanced flux at R.

The Monte Carlo simulation is the most complete and accurate calculation available for this type of phenomena within the accuracy of the initial assumptions. The two-atom model is a less accurate but useful approximation which compares well with the Monte Carlo calculation for $\rho/R < 0.6$, within the present limits of experimental accuracy. It is this region of ρ/R which will be the most useful for surface studies. The two-atom model obviously saturates at 2.0 atoms/string and deviates for small ρ/R, we believe, because of the lack of a properly screened potential. Both the two-atom model and the Monte Carlo calculations used a bulk Debye temperature of 348°K. As shown in Fig. 2, the trend of the data is in agreement with the theoretical model; the small discrepancy in absolute magnitude will be discussed later.

A summary of the published surface-peak measurements on well-characterized surfaces is shown in Fig. 3. The horizontal scale, ρ_B/R, is derived from the bulk vibration amplitude. For the limited range of cases studied, the ρ_B/R scaling (suggested originally in Ref. 11) is well verified. This scaling should be generally useful, although surface effects such as enhanced vibration amplitudes and structural changes must be considered for each individual case.

A number of other measurements and comments are related to this study:

(i) Davies *et al.*[2] have shown that surface relaxation (i.e., differences in the surface monolayer spacing from that of the bulk) can be studied by surface-peak-intensity measurements along axial directions which are not perpendicular to the surface. It is clear that for such directions the surface atoms will not be aligned with the string if relaxation exists; hence the surface-peak intensity will increase. Figure 2 indicates that measurements along the ⟨110⟩ and ⟨111⟩ axes show the proper scaling relative to the ⟨100⟩. This allows us to set a limit on the relaxation of the W(100) surface of ≤6%. This result is in disagreement with the recently reported work of Lee *et al.*[12] in which a contraction of 11% is deduced for W(100) by analysis of LEED data. Calculations show that an 11% contraction would produce a surface-peak intensity of ~1.7 atoms/string for the ⟨110⟩ and ~2.3 atoms/string for the ⟨111⟩ at 1.0 MeV (i.e., $E/d = 0.23$ and 0.37 MeV/Å, respectively). These values are well above our measured results. The discrepancy in measured relaxation may be the result of different methods of surface preparation in the two experiments.

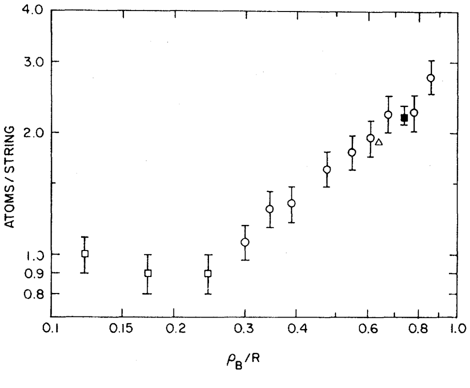

FIG. 3. Intensity of the surface peak vs ρ/R for various atomic systems. Open squares (Ref. 2), He$^+$ on Pt(111) at 40°K; open triangles (Ref. 3), 200-keV H$^+$ on Ni(110); closed squares (Ref. 4), He$^+$ on Au(100); open circles (present results), He$^+$ on W(100).

(ii) We have carried out a more extensive analysis of the surface-peak intensity in which the surface vibration amplitude was treated as a parameter. We find good quantitative agreement with our data for the following models: (a) If both the parallel and perpendicular components of the vibration amplitude of surface atoms in the first monolayer are increased by a factor of ~1.8 over the bulk value. [While large anisotropy is often expected for surface vibrations, this is not the case for (100) surfaces of bcc crystals as predicted by Jackson.[13]] (b) An increase of a factor of ~1.6 over the bulk value for atoms in the first two monolayers. (Note that the first two monolayers contain all the first atoms of the strings). (c) An increase of a factor of 1.5, 1.3, and 1.2 over the bulk value for the first, second, and third monolayers, respectively. This model is more consistent with the expected exponential decay of the enhanced amplitude.

(iii) After exposure of the W surface to ~7 L ($1\ \mathrm{L} = 10^{-6}$ Torr sec) of oxygen, the ⟨100⟩ surface-peak intensity increases by ~20%. This could be due to a lateral translation of the W surface atoms, as suggested by Bauer *et al.*[14] In any case this observation illustrates the importance of clean surfaces for comparison of surface-peak measurements with theory.

In conclusion, we have shown that the intensity of the surface peak behaves predictably as a function of energy or ρ/R. For $\rho/R < 0.4$ in W the interaction is almost entirely with the first exposed monolayer. In general, MeV scattering from single crystals in the region of $\rho/R < 1$ will be a useful surface tool.

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Linear Specific Heat of Disordered Solids at Low Temperatures

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It is shown that a linear specific heat in amorphous systems at low temperatures is a general consequence of the nonapplicability of the third law of thermodynamics. A hydrodynamic formula relating the pair density of states of Anderson *et al.* to diffusive and mechanical coefficients, determinable under favorable conditions by Rayleigh light scattering is also proposed.

Much interest has rightly been aroused by the observation of a low-temperature specific heat which is linear with temperature in amorphous solids[1] and also in spin-glasses.[2] In particular Anderson, Halperin, and Varma[3] have presented a model in which the linear temperature dependence is interpreted in terms of Schottky excitation between two levels; the two-level density of states at zero excitation energy determines the coefficient of this linear term.

In this Letter, we draw attention to the fact that for disordered systems in which the third law of thermodynamics is not obeyed, i.e., the entropy $S(T)$ is nonzero as $T \to 0$, one must have a linear specific heat. The argument utilizes the following thermodynamic equations:

$$C_P - C_V = T\alpha^2 V/K_T, \tag{1}$$

$$\alpha = \frac{1}{V}\left(\frac{\partial V}{\partial T}\right)_P = -\frac{1}{V}\left(\frac{\partial S}{\partial P}\right)_T, \tag{2}$$

$$K_T/K_S = C_P/C_V, \tag{3}$$

where K represents the compressibility and C the specific heat. We see immediately from (1) that if $C_P - C_V \propto T$ as $T \to 0$, then α, the thermal expansion must tend to a constant as $T \to 0$, since