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MULTIPLE-SCATTERING DESCRIPTION OF INTENSITY PROFILES OBSERVED IN LOW-ENERGY ELECTRON DIFFRACTION FROM SOLIDS*

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The dependence on beam energy and lattice geometry of the intensity of electrons diffracted from surfaces is evaluated using the inelastic-collision model. At energies below that of the second primary Bragg peak, qualitative features of the intensity profiles are related to the lattice geometry. The quantitative features depend sensitively on the scattering from the surface layer. The first detailed interpretation of such profiles is proposed for the (100) face of tungsten.

In recent work we have presented a heuristic derivation,[1] a perturbation-theory analysis,[2] and a matrix-inversion analysis[3] of the inelastic-collision model for the evaluation of the intensity versus energy profiles of low-energy electron beams diffracted from crystal surfaces. In this Letter we summarize the major results of our analysis and note their consequences for the interpretation of experimental data.

Interest in this topic is due to the fact that despite numerous experimental measurements of intensity profiles since 1927[4] and extensive theoretical studies of the effects of multiple scattering,[2,5-14] no satisfactory description of very low-energy ($E \lesssim 100$ eV) experimental data has been achieved. Our results, presented in Refs. 1-3 and herein, constitute the first systematic discussion of the influence of electron damping and lattice geometry on the predictions of multiple-scattering models. We also present herein the first detailed interpretation of experimental intensity profiles in the energy region of the first Bragg peak. The only other such interpretation is that of Hirabayashi,[14] which adequately describes the observed profiles of carbon exclusively at higher energies where only primary Bragg peaks occur. Earlier attempted interpretations[15-17] were based on a Darwin kinematical model in which the predicted scattering intensity is not simply related to a solution of the Schrödinger equation describing the electronic motion. Our analysis also reveals the critical importance for the interpretation of low-energy electron-diffraction (LEED) profiles both of strong inelastic-collision damping and of the electronic inequivalence of the "surface" and "bulk" layers of even a chemically clean, unreconstructed surface. Both of these effects usually have been regarded as "unimportant" in the current theoretical literature[7-13,18] on multiple-scattering descriptions of LEED from clean, unreconstructed surfaces.

The essential concept underlying the inelastic-collision model[1] is that the damping of the elastic wave field of an incident electron, due to its excitation of plasmons and incoherent electron-hole pairs, is the dominant feature of its motion in a solid which restricts its (elastic) penetration into the solid to a depth of about 5-10 Å. In this limit,[1,19] the energy widths and maximum intensities of the diffraction peaks are determined primarily by the damping length rather than by the lattice potential of the solid, and the location of the peaks depends primarily on the geometry of the lattice. The analytical formulation of the inelastic-collision model is achieved by using a propagator formalism[13] to describe the multiple scattering by the lattice. The new feature of the model is the use of electron propagators associated with a (uniform) interacting electron fluid,[20]

rather than those associated with noninteracting electrons.[13] The final result[1-3] is an expression for the intensity of a given LEED beam as a function of (1) its energy E, (2) its momentum parallel to the surface of the crystal $k_{\|}$, and (3) the ion-core partial-wave scattering amplitudes and (energy-dependent) electronic proper self-energy characteristic of the target.

In this Letter we consider only the (00) diffracted beam using a simplified, semiphenomenological version[2] of the inelastic-collision model in which the ion-core scattering is described by an s-wave scattering amplitude

$$t(E)=\{\exp[2i\delta(E)]-1\}/2ik(E), \tag{1}$$

and the electronic proper self-energy is taken to be

$$\Sigma(k,E)\equiv V_0(E)-i\Gamma(E), \tag{2}$$

in which V_0 is the electron inner potential and $\Gamma(E)$ is related to a prescribed damping length, λ_{ee}, by

$$\Gamma(E)\equiv(\hbar^2/2m\lambda_{ee})[(2m/\hbar^2)(E+V_0)]^{1/2}. \tag{3}$$

The propagation wave vector in the solid is determined by energy and $k_{\|}$ conservation via

$$E-(\hbar^2/2m)(k_{\perp}^2+k_{\|}^2)-\Sigma(k_{\perp},k_{\|},E)=0. \tag{4}$$

Equation (4) leads to a complex propagation constant, $k_{\perp}=k_{\perp 1}+ik_{\perp 2}$, for motion normal to the surface of the crystal.

The intensity of the (00) diffracted beam is a function of the beam parameters, E and $k_{\|}$, and of the material parameters δ_S, δ_B, and λ_{ee}. A subscript S is used to designate the phase shift associated with ion cores in the surface layer and a subscript B to designate the remaining "bulk" ion-core phase shifts. It is given by[1,13]

$$I_{00}(E)=|(-m/2\pi\hbar^2)\textstyle\sum_{\nu}e^{2ik_{\perp}(E)d_{\nu}}T_{\nu}\delta_{k'_{\|},k_{\|}}|^2 \tag{5}$$

in which d_ν denotes the distance from the surface of the layer labeled by ν ($\nu=0$ denotes the surface layer). The self-consistent layer scattering amplitudes T_ν satisfy a set of coupled algebraic equations[1,13] of the form

$$T_{\nu}=\tau_{\nu}+\tau_{\nu}\sum_{\nu'\neq\nu}G_{\nu\nu'}T_{\nu'}, \tag{6a}$$

$$\tau_{\nu}=t_{\nu}[1-t_{\nu}G_{\nu}]^{-1} \tag{6b}$$

in which $G_{\nu\nu'}$ and G_ν are appropriately defined propagators.[1,2,13] Thus τ_ν describes the scattering from an isolated "νth" layer of ion cores. It exhibits resonance structure (for s-wave scattering) when $[1-t_\nu G_\nu]^{-1}\to 0$. These resonances have been designated "multiple-scattering" resonances by McRae.[7] They correspond to a threshold effect in an elastic channel near the threshold for an inelastic process.[2] The conventional "nth-order primary Bragg peaks" result from taking all of the T_ν to be identical constants. Additional resonant peaks and minima in $I_{00}(E)$ occur because $T_\nu\neq\tau$ in the solutions of Eq. (6a). We refer to them as "higher-order" Bragg peaks. The criteria for these higher-order peaks depend both on the Bravais net parallel to the surface and on the geometry of successive scattering planes.[2] Hence, the energy at which they occur is a direct and rather sensitive consequence of the geometry of the ion-core arrangement in the top few layers of the solid.[2]

The first task in an investigation of the consequences of our model is the determination of the dependence of the predicted intensity profile on the values of the model parameters. The theory of the uniform electron fluid[1,20] predicts that $\lambda_{ee}(e)$ decreases from nearly infinity at the Fermi energy in the solid to a value of about 4-6 Å at energies near 10 times the Fermi degeneracy ζ, and then slowly increases with increasing energy. This predicted behavior is consistent[1] with all known data on the inelastic-collision mean free paths of electrons in the energy range $E\lesssim 200$ eV. The inner potential is known to satisfy[21] $V_0(\mu)=-\varphi-\zeta$ for electrons at the Fermi energy μ, where φ is the field-emission work function and ζ is the Fermi degeneracy of the valence electrons. Except near a plasmon-emission threshold, $V_0(E)$ decreases in magnitude with increasing energy.[20,21] The ion-core phase shifts are not known, but in principle can be related to the parameters of models for the ion-core potentials. The sensitivity of the predicted intensity profiles to the values of these phase shifts is illustrated in Fig. 1. From this figure and similar calculations we conclude that the electronic structure of the ion-core factors in the top layers of the solid usually dominate the predicted intensity profiles. Hence LEED intensity profiles do not reflect the band structure of the bulk solid, but rather measure the electronic properties of the surface layers. In metals, the electronic structure of the bulk and surface layers differ even for chemically clean surfaces.[21] The importance of the electronic inequivalence of the surface and bulk layers for the prediction of intensity profiles render "first-principles"

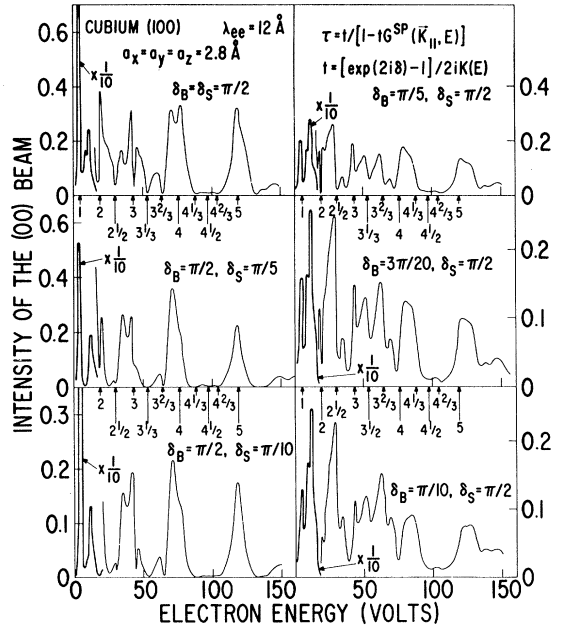

FIG. 1. Intensities of the (00) beam normally incident on the (100) face of a simple cubic lattice calculated in second-order perturbation theory. Scattering from the top layer of ion cores is described by the phase shift δ_S and that from the "bulk" ion cores by δ_B. The intensities are calculated using Eqs. (15), (30), (44), and (45) given by Duke, Anderson, and Tucker (Ref. 2) using the parameters shown in the figure.

calculations of these profiles considerably less reliable than those of bulk electronic band structures because of the difficulty in describing variations in the electronic density (and hence ion-core form factors) in the top layers of the solid.[21]

A detailed discussion of the influence of the lattice geometry on the intensity profiles is given by Duke, Anderson, and Tucker.[2] The important feature of this discussion is its simplicity, which renders the results directly usable by experimentalists. As the simplicity of this analysis relies heavily on the use of double-diffraction perturbation theory,[2] we have compared the intensities thus calculated with those obtained from the numerical solution of Eqs. (6). The results for three faces of aluminum are shown in Fig. 2. The usual consequence of the numerical calculation, evident in the figure, is the smoothing out of some of the fine structure predicted by the double-diffraction approximation. However, the qualitative features of the predicted intensity profiles remain unaltered for s-wave scattering from equivalent layers of ion cores. The case of an inequivalent surface layer is more complicated and is currently under study.[3]

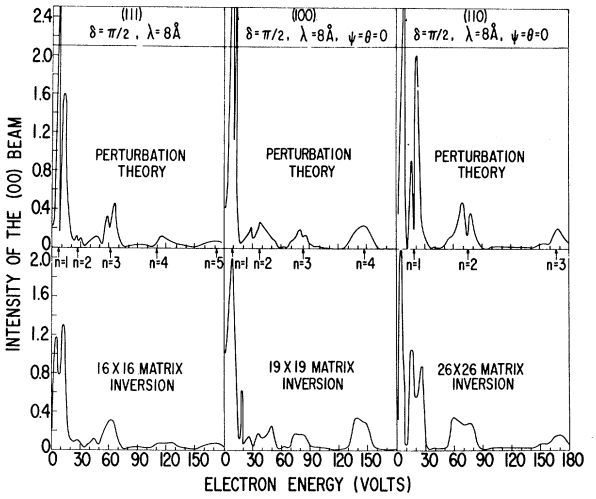

FIG. 2. Comparison of the matrix-inversion and double-diffraction calculations of a (00) beam of electrons normally incident on the various faces of an fcc aluminum lattice; $\delta = \pi/2$, $\lambda_{ee} = 8$ Å, $4 = \theta = 0$. In all cases, the results of the matrix-inversion calculations do not change significantly if the dimensionality of the matrix is increased further.

Concluding with the question of the interpretation of experimental data, we first note that due to the occurrence of many internal beams at higher energies, the relation between lattice geometry and the features of the intensity profile is likely to be simple only at energies below that of the $n = 2$ primary Bragg peak (unless the electron ion-core scattering is sufficiently weak that only primary Bragg peaks occur). The most extensive low-energy data on metals have been taken on the (110), (100), and (211) faces of tungsten.[15,22-25] A discussion of all three faces is given by Duke, Anderson, and Tucker.[2] In Fig. 3 we show a comparison between the model calculations and experimental data at two angles of incidence on W(100). The qualitative similarity between the theoretical and experimental profiles is evident from the figure. The interpretation of the maxima in the calculated profiles is noted in the figure. Although small changes in the parameters δ_S, δ_B, and λ_{ee} do not alter drastically the calculated intensities, the suitability of the theoretical description was decreased substantially by the use of other regions of the three-dimensional parameter space which we searched. The deviations evident in Fig. 3 between the experimental data and the model predictions are comparable with deviations between experimental measurements taken on different, "equivalent" crystals.[22-25] Therefore we conclude that the interpretations noted in Fig. 3 are reasonably

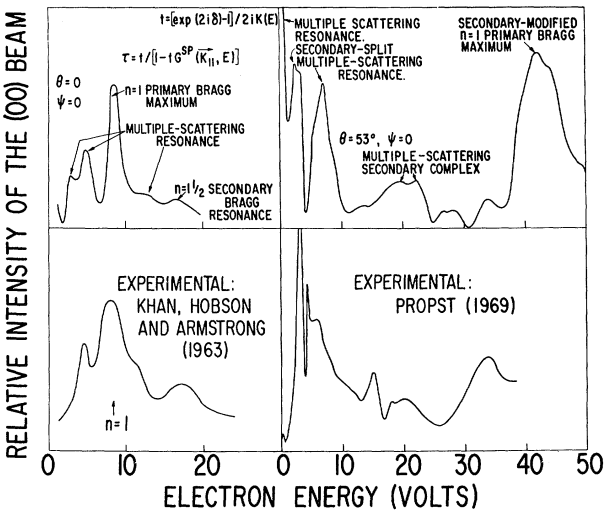

FIG. 3. Comparison of the intensity profiles calculated using the double-diffraction approximation (top panels) with the experimental relative-intensity profiles (lower panels) for a (00) beam incident on the (100) face of tungsten. Parameters for the calculations are $\delta_B = 0.5\ \pi$, $\delta_S = 0.65\ \pi$, $\lambda_{ee} = 14$ Å, and as indicated in the figure. The identification of the various peaks in the calculated profiles is indicated. The values of the phase shifts were adjusted to describe the data. The calculations were performed using Eqs. (15), (30), (44), and (45) in Duke, Anderson, and Tucker (Ref. 2). The normal-incidence experimental data are taken from Khan, Hobson, and Armstrong (Ref. 22), and the 53° incidence data from Propst (Ref. 25).

unique consequences of the double-diffraction analysis of the model, although the associated inner-potential shifts, $V_0 \cong 5$ V, are smaller than we anticipated.

The numerical solutions to Eqs. (6a) and (6b) for a normally incident beam give results[3] similar to those shown in Fig. 3 with the $n = 1$ Bragg peak apparently occurring as a high-energy shoulder on the multiple-scattering structure.

Another large maximum occurs near 28 eV. The only sensible alternative to the interpretation given in Fig. 3 seems to be that obtained by identifying the experimental observations with this higher-energy resonance and using an inner potential $V_0 \cong 20$ eV. Hence the interpretation in Fig. 3 appears to be the more reasonable one of the two alternatives.

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