INELASTIC EFFECTS IN LOW-ENERGY ELECTRON DIFFRACTION

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The systematic inclusion of inelastic effects in low-energy electron diffraction leads to (1) broad (10- to 20-eV) peak widths, (2) asymmetric peak shapes, and (3) low reflectivities. A connection between the "dynamical" and "kinematic" approaches to diffraction theory is developed.

The diffraction of low-energy electrons by crystal surfaces is a widely used experimental technique for studying surface properties and has recently stimulated considerable theoretical attention.¹⁻⁴ Of particular interest is the spectrum of the elastically reflected beam intensity as a function of the incident-electron energy. Previous approaches to this problem generally have considered elastic scattering alone, solving the boundary value problem by multiple-scattering or wave-matching approaches. These intensity calculations predict peak widths which correspond to the band gaps in the bulk material, i.e., of the order of 1 eV. These predictions are in marked disagreement with experimentally observed peak widths⁵ (10-20 eV) and the discrepancy has remained in spite of ad hoc inclusion of inelastic-scattering effects by the addition of small imaginary parts to the phase shift¹ or to the Fourier components of the crystal potential. This Letter reports model calculations of electron reflectivities which include, for the first time, the effect of inelastic processes in a systematic way. The absolute reflectivity appears as a quantity of the first importance, and our conclusions are consistent with reflectivities of the order of a few percent or less. The results explain the large discrepancies in linewidth between experiment and current theories and suggest that appropriate modifications to the latter will remove this variance.

A formalism derived by Yoshioka⁶ in a calculation of inner-potential corrections in high-energy electron diffraction may be adapted to the present problem.

The interaction $H'(\mathbf{\tilde{r}})$ between an electron at position $\mathbf{\tilde{r}}$ and the system of electrons and ions constituting the crystal is

$$H'(\mathbf{\hat{r}}) = \sum_{j} \frac{e^2}{|\mathbf{\hat{r}} - \mathbf{\hat{r}}_j|} - \sum_{k} \frac{Z_k e^2}{|\mathbf{\hat{r}} - \mathbf{\hat{R}}_k|},$$
(1)

where $\mathbf{\tilde{r}}_j$ is the coordinate of the *j*th electron and $\mathbf{\tilde{R}}_k$ that of the *k*th nucleus. The effective Hamiltonian for the incident electron may then be written

$$H = T + H_{00}'(\tilde{\mathbf{r}}) + \Sigma, \qquad (2)$$

where T is the kinetic-energy operator and $H_{00}'(\mathbf{\bar{r}})$ is the effective one-particle periodic potential obtained by averaging over the ground state of the crystal. The additional term Σ is a nonlocal potential or integral operator, whose Fourier transform may be written, to second order, by

$$\Sigma_{\widehat{\mathbf{h}}\widehat{\mathbf{g}}} = -\frac{1}{\Omega} \sum_{n \neq 0} \int \frac{d^3 \widehat{\mathbf{q}}}{(2\pi)^3} \frac{H_{n0}'[\widehat{\mathbf{k}} + \widehat{\mathbf{h}} - \widehat{\mathbf{q}}]H_{0n}'[\widehat{\mathbf{q}} - \widehat{\mathbf{k}} - \widehat{\mathbf{g}}]}{(q - i\delta)^2 - (E - E_n)}$$

In this equation $H_{n0}'[K] = \int H_{n0}'(\mathbf{r}) \exp(-i\mathbf{K}\cdot\mathbf{r}) d^3\mathbf{r}$, $\mathbf{\tilde{g}}$ and $\mathbf{\tilde{h}}$ are reciprocal lattice vectors, and $\mathbf{\tilde{q}}$ is the wave vector of an intermediate electron state. Matrix elements of the effective one-electron periodic potential are taken between the ground state $|0\rangle$ and excited states $|n\rangle$ of the crystal, and the integration is over all values of \mathbf{q} . The real and imaginary parts of this potential correspond to virtual and real inelastic processes, respectively. If the interaction between the incident electron and the ions is neglected in Σ , Eq. (3) is then the self-energy of an electron interacting with a uniform electron gas.⁷ Bandstructure effects in many-particle systems present severe computational difficulties and approximations in the present work have utilized recent advances in calculations of electron self-energies in an interacting uniform electron gas.

The reflection coefficient of an electron incident on the surface is found by matching the logarithmic derivative of the electron wave function at the surface to the same quantity inside the crystal. Calculations using this method have

been carried out by Kronig and Penney⁸ and by a number of authors subsequently. The wave function inside the crystal is usually expanded in plane waves and the one-electron crystal potential takes some suitable model form. The first model we discuss, in which the potential is simulated by a linear array of delta functions,⁸ may, however, be solved exactly and is free of truncation errors. Electron-electron scattering processes are included by adding to the crystal potential the energy- and momentum-dependent self-energy for the appropriate value of r_s , as calculated by Lundqvist⁹ for the uniform electron gas within the random-phase approximation. The one-dimensional calculation has been performed using values of r_s and lattice constant appropriate to zinc. Reflectivity behavior for two values of $V_{\overline{g}}$, proportional to the strength of the delta function, are shown in Fig. 1. Several features are immediately apparent: (1) the broad peak widths, (2) the asymmetric peak shapes for small values of $V_{\overline{g}}$, and (3) low reflectivities. The peak widths and reflectivities are comparable with those obtained by Lander and Morrison¹⁰ and by Baker, Blakely, and Strozier.¹¹

In Fig. 2, the width of the 138-eV peak is plotted against $V_{\overline{g}}$. For large values of $V_{\overline{g}}$, the crystal potential dominates the scattering and the peak width varies linearly with the potential ("dynamical region"). For very weak potentials (and correspondingly narrow "dynamical widths"), the peaks are again very broad and take the familiar "cosine-squared" kinematic shape. In the intermediate region between these two limits, where lifetime effects are important, the abso-



FIG. 1. Reflectivity as a function of incident-electron energy for $V_g = 8.2 \text{ eV}$ (full curve) and $V_g = 1.37 \text{ eV}$ (broken curve). Note the change of scale. Upper figures on reflectivity scale refer to full curve.

lute reflectivity is a more sensitive probe of the scattering mechanism than the peak width. In the region of very low reflectivity (below the nose in Fig. 2), the experimental widths (typically 10-20 eV) imply that the corresponding dynamical widths and hence the Fourier coefficients for electron scattering are of the order of an electron volt. This observation shows that the dynamical calculations¹⁻⁴ are consistent with large peak widths, provided that the large inelastic scattering is included. For incident electrons whose energy is less than the plasmon energy (~10 eV), inelastic processes are relatively unimportant. Scattering at these energies is essentially dynamical with correspondingly large reflectivities and widths which more closely reflect the band structure.

The intensity curves always lie inside an envelope which corresponds to purely elastic scattering and, in this sense, our results are consistent with the observation of McRae¹ that the introduction of inelastic scattering does not alter the base widths of the peaks. If appropriate elastic scattering is taken and the imaginary part of the potential increased from zero, the reflectivity decreases uniformly, though the full width at half-maximum changes, at first, very little. For the imaginary parts used in the present calculation and the corresponding very low reflectivities, however, the width so defined is much greater than the dynamical width.

Lifetime effects play a much more significant



FIG. 2. Potential strength V_g plotted against width of 138-eV peak. The straight line through the origin corresponds to the width to be expected from a purely dynamical theory.

role than polarization effects, which produce a weakly \bar{k} -dependent energy shift in the present model. Peak broadening may occur in a dynamical theory as a result of spatial variations in the real part of the self-energy. To estimate this effect, we have computed the Fourier transform of the polarization potential of a periodic array of sodium atoms.¹² For energies usually considered in low-energy electron diffraction the effect is negligible, though for very low energies (<10 eV) it may be important.

The basic results of this calculation also hold for the case of a more realistic three-dimensional potential. In particular, the low-energy electron-diffraction dynamical calculations reported previously¹⁻⁴ may readily be modified by the inclusion of an energy- and momentum-dependent, spatially homogeneous self-energy.¹³ This conclusion is supported by preliminary results from a calculation in which the wave field inside the crystal is expanded in terms of Bloch states of complex momenta and matched to a linear combination of plane waves outside.¹⁴

The emphasis placed in this Letter on <u>absolute</u> reflectivity has important consequences for future experimental work. There are at present very few published data of this nature.

It is a pleasure to acknowledge the generous assistance of Dr. N. W. Ashcroft, Dr. J. M. Baker, Dr. J. M. Blakely, Dr. C. Y. Li, Dr. T. N. Rhodin, and Dr. J. W. Wilkins.

*Work supported by the Advanced Research Projects

Agency through the Materials Science Center at Cornell University, Materials Science Center Report No. 1131.

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