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Kinematic Low-Energy Electron-Diffraction Intensities from Averaged Data: A Method for Surface Crystallography*

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It is demonstrated that the interference of the low-energy–electron multiply scattered amplitudes can be in large part eliminated by averaging data taken with different diffraction geometry but at fixed momentum transfer. This essentially yields a measurement of the kinematic intensities, which are most directly useful for surface crystallography.

The most important potential of low-energy electron diffraction is the unambiguous determination of the atomic structure of the outermost few atomic planes of a crystal, but thus far no structure has been determined using this technique in the sense that bulk structures have been determined using x-ray crystallography. The periodicity of the structure parallel to the surface requires that the parallel component of the electron momentum be conserved to within a reciprocal-net vector. This enables one to determine the size and shape of the unit mesh, but to determine the crystal structure one must be able to interpret the diffracted intensities. Low-energy electron-diffraction intensities are very complicated because of important multiple-scattering contributions to the scattered amplitude. Although recent theoretical results reproduce the features of measured intensities, these require extensive computation and have generally been limited to simple systems.[1] Calculations of the intensities for assumed structures are far more complex in a dynamic than in a kinematic theory, and the inverse problem of determining the autocorrelation function from measured intensities has not been attempted in the framework of the dynamic theory.

Information about the atomic sturucture is most readily obtained from data which can be interpreted properly by a kinematic, or single-scattering, theory: Thus one would like to extract this first-order contribution from the low-energy electron-diffraction data. It is the purpose of this Letter to present an experimental demonstration that by properly averaging the data one can indeed eliminate the interference of the multiply scattered amplitudes and extract what is essentially the kinematic intensity.

The procedure is motivated by the following argument. The scattered intensity, including the multiple scattering explicitly to second order, is

$$I(\vec{S}) = AA^* = \sum_{i,m} f_i f_m^* \alpha_i \alpha_m \exp[i\vec{S}\cdot(\vec{r}_i - \vec{r}_m)] + \sum_{\substack{i \neq j \\ m \neq n}} f_{ij} f_{mn}^* \alpha_{ij} \alpha_{mn} \times \exp\{i[-\vec{k}_0\cdot\vec{r}_{im} + \vec{k}\cdot\vec{r}_{jn} - |\vec{k}|(|\vec{r}_{ij}| - |\vec{r}_{mn}|)]\} + \cdots, \quad (1)$$

where $\vec{k}$ and $\vec{k}_0$ are the propagation vectors of the scattered and incident beams, respectively, and $\vec{S} = \vec{k} - \vec{k}_0$. α_{ij} accounts for the attenuation of the ray scattered successively by atoms i and j, and f_{ij} is the product of the appropriate scattering factors. Equation (1) and the arguments that follow can easily be extended to higher orders. The first sum, $I^{(1)}$, is the single scattering and depends only on

$\vec{S}$. The second sum, $I^{(2)}$, is the double scattering.

In $I^{(2)}(\vec{S})$ those terms in which $i=m$ and $j=n$ give the intensity from independently scattering pairs of atoms and account for the entire angular integral of the double scattering. The remaining terms, for which i, j, m, and n are distinct, give the interference of the twice-scattered amplitudes. This interference confines the intensity to the reciprocal-lattice rods and is responsible for the complicated intensity profiles on the rods. These terms depend explicitly on $\vec{k}$ and $\vec{k}_0$. If the intensity is measured at fixed $\vec{S}$ but over a range of $\vec{k}$ and $\vec{k}_0$, $I^{(1)}(\vec{S})$ remains constant, but the interference terms oscillate and the averaged multiple scattering tends to a smooth background along the rods. This averaging should thus eliminate the interference of the multiple scattering, leaving the kinematic intensity and a slowly varying multiple-scattering background.

The experiment limits the range over which the average may be taken; the question is whether the accessible range is sufficient so that the remaining multiple-scattering interference in the incomplete average is small compared to the single-scattering intensity. This question is to be answered experimentally; however, that the answer should be favorable is suggested by the fact that the single scattering makes a large contribution to the observed intensity.[1,2]

Here we present experimental results for the specular reflection from the (111) face of Ag; more extensive results for both Ag and Ni, as well as details of the arguments motivating the averaging procedure, will be described in a future publication.[3] The experiments were done with apparatus and procedures described previously.[4] The ratio of the specularly reflected elastic intensity to the incident intensity, I/I_0, was measured as a function of the incident-electron energy for a fixed grazing angle of incidence φ and azimuth ζ. (For the specular reflection, $\theta=\varphi$.) This measurement was repeated every 10° for a 60° range of ζ. These six curves were then averaged. An example is shown in Fig. 1. In the average, the Bragg peaks are clearly evident; and, although there is still multiple-scattering structure, the average is simpler than most of the individual curves. This procedure was repeated at each of six angles of incidence, $68°<\varphi<78°$. The resulting azimuthal averages are shown in Fig. 2 plotted as a function of the reduced diffraction vector $S/S_0=\frac{2}{3}\sqrt{3}(a_0/\lambda_0)\sin\theta$,

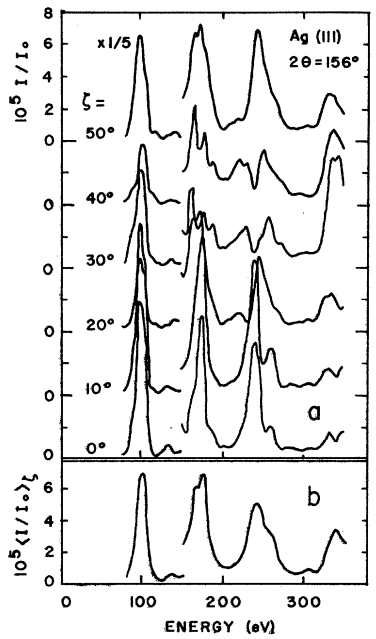

FIG. 1. Azimuth average at fixed angle of incidence. (a) Intensity versus energy in the (00) beam for different azimuths ζ. (b) Average of the curves in (a). 2θ is the scattering angle.

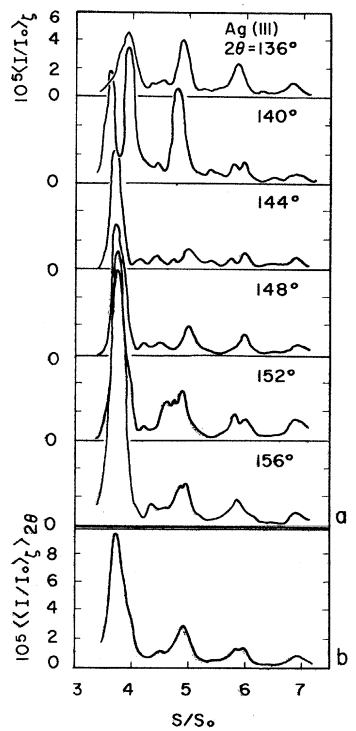

FIG. 2. Grand average. (a) Azimuth averages for different angles of incidence φ. $2\theta=2\varphi$, where φ is measured from the surface. Each curve is the average of six curves, as shown in Fig. 1. (b) Average of the curves in (a) as a function of S/S_0, the reduced diffraction vector.

where a_0 is the Ag lattice constant. We have neglected the inner potential, so λ_0 is the free-space wavelength. Figure 2(b) shows the average of the aximuthal averages; it is clear that in the average the subsidiary structure is essentially gone. Inner-potential corrections of 17, 9.5, 9.2, and 12.3 eV for the (444) through (777) peaks, respectively, would be required to bring them to integral values of S/S_0.

The range of $\vec{k}$ and $\vec{k}_0$ used here[5] suffices to nearly average out the multiple-scattering interference. Experiments with finer increments in ζ at fixed φ and over a larger range of 2θ at fixed ζ indicate that the limited data presented are sufficient to give the features of the averaged curve.

It remains to show that this average is a good approximation to the kinematic intensity by comparing it with the calculated result. Assuming a uniform attenuation of the elastic beam and a scattering factor the same for all atoms, the intensity of the specular reflection at a given $\vec{S}$ is

$$I(\vec{S}) \propto |f|^2 e^{-2M} \mathfrak{L}(E)[1-W(\varphi)]^2 \times (1+\alpha^2-2\alpha\cos\vec{S}\cdot\vec{c})^{-1}, \qquad (2)$$

where $|f|^2$ is the square of the atomic scattering factor, e^{-2M} is the Debye-Waller factor, $\mathfrak{L}(E)$ is the Lorentz factor,[6] and $c = a_0/\sqrt{3}$. $[1-W(\varphi)]^2$ is a correction for the surface losses.[7] The last factor is the interference function along the (00) rod for an absorbing crystal. $\alpha = A_{i+1}/A_i$ is the ratio of amplitudes scattered from successive planes and is related to the linear attenuation coefficient μ by $\alpha = \exp(-\mu c/\sin\varphi)$. To compare the kinematic intensity with the result in Fig. 2(b), it is necessary to average Eq. (2) over 2θ and λ_0 along the line defined by $|S| = (4\pi/\lambda_0)\sin\theta = \text{const}$.

The quantities in Eq. (2) are all known from other sources. We use the $|f|^2$ calculated by Kambe.[8] Values of e^{-2M} were taken from the results of Jones, McKinney, and Webb.[4] We take[6] $\mathfrak{L}(E) \propto E^{-1}$, and neglect its angular dependence. $[1-W(\varphi)]^2$ is calculated from optical constants.[9] μ and its energy dependence have been estimated from various experiments and calculations,[10] but there is considerable uncertainty in these estimates. Therefore for the present purpose we calculate α using $\mu = \mu_0 E^{-1/2}$, where μ_0 is a single energy-independent parameter chosen to fit the widths of the peaks in Fig. 2(b), and then compare this value with previous estimates.

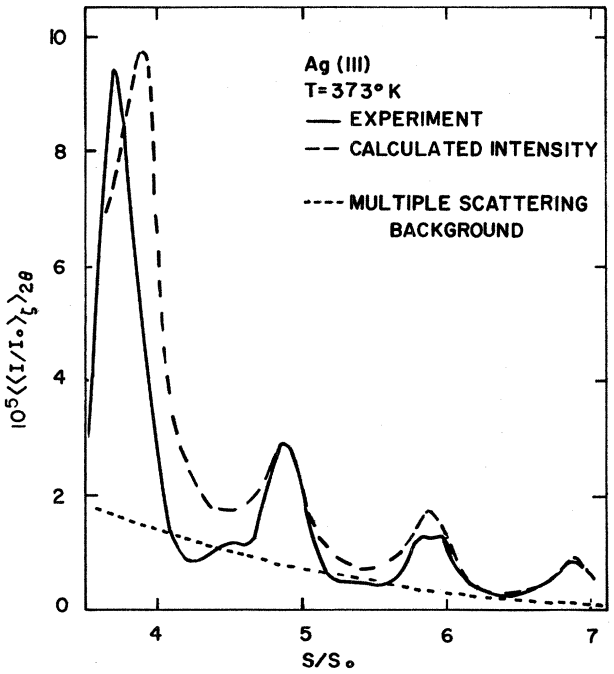

FIG. 3. Comparison of calculated kinematic intensity with experimental average. The multiple-scattering background has been added to the calculated kinematic intensity [Eq. (2)]. The result is normalized to the (555) peak height and shifted by a constant $-0.12S/S_0$.

In addition to the kinematic intensity, the observed average contains the multiple-scattering background, which can be estimated only crudely. Using results for the integral of the multiple scattering[2] and arguments on the temperature dependence of the multiple scattering,[11] we estimate this background as shown by the short dashes in Fig. 3. This intensity is a smooth function of S/S_0; and thus, despite the large uncertainties, it does not importantly affect the determination of the relative intensities or the widths of the Bragg peaks.

The resulting sum of the kinematic intensity and the multiple scattering background is shown in Fig. 3. No correction for the inner potential has been included in the calculation, but the calculated curve has been shifted by -0.12 in S/S_0 to facilitate comparison of peak intensities and widths. $\mu_0 = 4.9$ Å^{-1} V$^{1/2}$ was chosen to fit the widths; earlier estimates[10] of μ_0 have ranged from 2.8 to 4.5 Å^{-1} V$^{1/2}$.

The present experiment recovers the kinematic intensity very well. In fact the agreement is better than can be expected from the uncertainties in presently available values of factors in Eq. (2). Also effects of thermal expansion and

the small relaxation of the interplanar spacing at the surface have been neglected. If inner potential corrections were made before performing the averages over 2θ, the peaks in the experimental average would be about 10% narrower.

It seems clear that the kinematic intensity can be determined sufficiently well so that the modulation of the peak intensities by the crystal-structure factor can be recognized in order to distinguish between proposed models for unknown surface structures. Although this is demonstrated here only for the specular reflection, the procedure is applicable to other beams as well. Structure factors for surfaces which are reconstructed or contain adsorbed atoms will produce large modulations in diffracted intensities because the amplitude scattered by the outermost plane of atoms is of the same order of magnitude as that scattered by the remainder of the crystal. It is from this modulation of indexed reflections, rather than from their detailed position and shape, that structural information will be most readily available since the latter is determined by inner potentials which at present are not well known.

The use of averaged low-energy electron-diffraction intensity data was anticipated by Lander and Morrison.[12] Independently Duke and Tucker[13] have discussed averaging results of their model calculations to extract structural information.

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Multiple Reflections in Gantmakher-Kaner Oscillations*

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Gantmakher-Kaner oscillations in gallium have been observed at 1.3°K at 9.2 GHz in the transmission mode for the field-normal case in addition to angles well away from the normal. A set of Gantmakher-Kaner oscillations with striking properties and a very small period has been associated with electrons on a small "approximately ellipsoidal" Fermi surface piece oriented along k_b. These oscillations have harmonic content which provides direct evidence for multiple reflections in the gallium slab indicating specular surface scattering for these particular electrons. A dramatic disappearance of just the fundamental as the dc field is decreased below the cyclotron resonance field cannot be explained by current theories of Gantmakher-Kaner oscillations.

In the presence of an incident electromagnetic wave under anomalous skin-effect conditions, ineffective (in the Pippard sense) electrons or holes can carry energy in the form of coherent electromagnetic waves well beyond the skin depth δ to a distance of order of the electron or hole mean free path (mfp) λ. When a dc magnetic field $\vec{H}$ is oriented normal to a metal slab (or an angle