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Symmetry in Low-Energy–Polarized-Electron Diffraction

G.-C. Wang, B. I. Dunlap, R. J. Celotta, and D. T. Pierce
National Bureau of Standards, Washington, D. C. 20234
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The first low-energy–electron-diffraction measurements using a polarized incident electron beam are reported and compared to measurements where an unpolarized incident beam is analyzed after scattering. Whereas, because of multiple scattering, equivalence of the two measurements is not expected in general, excellent agreement is obtained for specular scattering in the (010) plane from W(100). A theoretical argument is presented for the case where the scattering plane is a mirror-symmetry plane of the crystal.

Electron scattering from both free atoms and surfaces is a spin-dependent process; i.e., the cross section for scattering an electron at a given energy and angle will in general be different for electrons with spins aligned up or down relative to the scattering-plane normal. This comes as a consequence of the interaction between the incident electron's spin, $\vec{s}$, and its orbital angular momentum, $\vec{L}$, as it scatters from the atom core. This spin-orbit interaction energy is proportional to $(1/r)(dV/dr)\vec{s}\cdot\vec{L}$, where V is the scattering potential. As a consequence of the different cross sections, an unpolarized beam (made up of equal number of spin-up and -down electrons) will be polarized after scattering, and an initially polarized beam will scatter with different intensity when its spin direction is changed from up to down.

The purpose of this paper is to report the first use of a beam of polarized electrons as a surface probe and to demonstrate that under certain symmetry conditions measurement of the dependence of the scattered intensity on incident-beam polarization, and the measurement of the polarization of an initially unpolarized beam after scattering, are equivalent. In recent polarized low-energy–electron-diffraction (PLEED) studies,[1-5] an unpolarized incident beam was scattered from a crystal, and the polarization $P(E,\theta)$ induced by the crystal in the scattered beam was measured with respect to the scattering plane normal $\hat{n} = (\vec{k}\times\vec{k}')/|\vec{k}\times\vec{k}'|$, where $\vec{k}$ and $\vec{k}'$ are the electron wave vectors before and after scattering.[6] In contrast, we use a polarized incident beam to determine the strength of the spin dependence of the scattering,

$$S(E,\theta) = \frac{1}{P_0}\frac{I(P_0)-I(-P_0)}{I(P_0)+I(-P_0)}, \tag{1}$$

by measuring the change in the intensity of a scattered beam for a given polarization P_0 of the incident beam modulated sinusoidally parallel (P_0) or antiparallel ($-P_0$) to the normal to the scattering plane. $S(E,\theta)$ is a property of the crystal and is independent of the magnitude of P_0.

In scattering from atoms it is well known[7] that $P(E,\theta)=S(E,\theta)$. In electron diffraction from solids, the situation is complicated by multiple scattering and the lower symmetry of the crystal surface and in general $P(E,\theta)\neq S(E,\theta)$. In this work we present the first experimental diffraction measurement of $S(E,\theta)$ at a solid surface and the first comparison to $P(E,\theta)$ measured[2] in the same scattering geometry. We find that $S(E,\theta)=P(E,\theta)$ for scattering of the specular beam on W(100) incident at an azimuth in the (010) plane. We present a theoretical argument which shows that the equality between $P(E,\theta)$ and $S(E,\theta)$ arises in cases where the scattering plane is a mirror-symmetry plane of the crystal.

To measure $S(E,\theta)$ we have developed a new experimental technique in which a polarized-electron gun is added to an otherwise conventional LEED apparatus as shown in Fig. 1. This allows the measurement of the polarization information, $S(E,\theta)$, simultaneously and at the same speed as the usual intensity measurements. The polarized-electron source[8] used in these experiments consists of a GaAs photocathode from which a beam

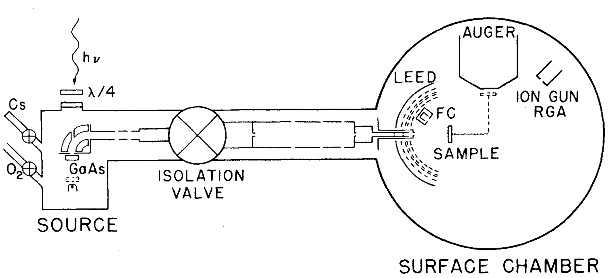

FIG. 1. LEED apparatus in which the conventional electron gun is replaced by the negative-electron-affinity GaAs polarized-electron gun. The polarization of the electron beam is modulated in phase with the changing circular polarization of the light produced by the rotation of the quarter-wave plate ($\lambda/4$). The longitudinal polarization is changed to a transverse polarization by the 90° spherical deflector. The spin-modulated beam which has constant intensity is incident on the sample crystal. Any modulation in the scattered intensity detected by the Faraday collector (FC) results from spin-dependent scattering. (The FC is actually out of the plane of the drawing for measurement of a spin-dependent signal.)

current of 10^{-6} A is extracted with an energy spread of less than 0.2 eV. The beam polarization may be reversed in direction by changing the helicity of the circularly polarized incident radiation. The beam current remains constant while the polarization is modulated, typically at 37 Hz. Thus any modulation observed in the diffracted signal must correspond to a spin-dependent term in the scattering. A Faraday collector can be positioned on a LEED spot and can scan both the polar and azimuthal scattering angle. The signal measured at the Faraday collector has both an average (dc) value and an ac component at the modulation frequency. The dc signal is the spin-averaged LEED intensity, while the size of the phase-locked ac signal directly measures the spin dependence of the scattering.

We measured $S(E,\theta)$ from a W(100) crystal which was cleaned by heating several hours in 1×10^{-6} Torr O_2 at 1800 K with flashes to 2500 K, and then flashing to 2500 K in ultrahigh vacuum before each data run. A single data run accumulated the $S(E,\theta)$ and $I(E,\theta)$ profile over the incident energy range of 50 to 150 eV in 1-eV steps with an integration time of 1 sec per point. The temperature of the crystal was held between 500 and 650 K during the measurement to prevent reconstruction and thus maintain a 1×1 pattern.[9] Scanning of the Faraday cup through the $(\frac{1}{2},\frac{1}{2})$ beam position gives only the background intensity indicating that the surface is truly a 1×1 pattern under our experimental conditions. The reproducibility of the $S(E,\theta)$ profile was excellent with agreement being obtained over periods of many weeks and from two different crystals.

The measured $S(E,\theta)$ for specular scattering from W(100) at an angle of incidence of 15° is shown in Fig. 2. From $S(E,\theta)$ and the average $I(E,\theta)$, we can determine the intensity curves that would result if the incident beam were entirely spin up or spin down; these curves are shown in the bottom of Fig. 2. Spin-dependent effects can be very large; at the 79-eV minimum $I_\downarrow$ is 5 times $I_\uparrow$, and even at the 86-eV maximum $I_\downarrow$ is approximately twice $I_\uparrow$.

In order to determine the relationship between previously measured $P(E,\theta)$ and our $S(E,\theta)$, we measured $S(E,\theta)$ and $I(E,\theta)$ of the (00) beam for polar scattering angles from 160° to 146° where $P(E,\theta)$ data are available. This corresponds to an angle of incidence range of 10° to 17° which we measured in 1° steps. The $I(E,\theta)$ curves are in good agreement[10] with other available data.[11] In Fig. 3 we compare our $S(E,\theta)$ measured with the

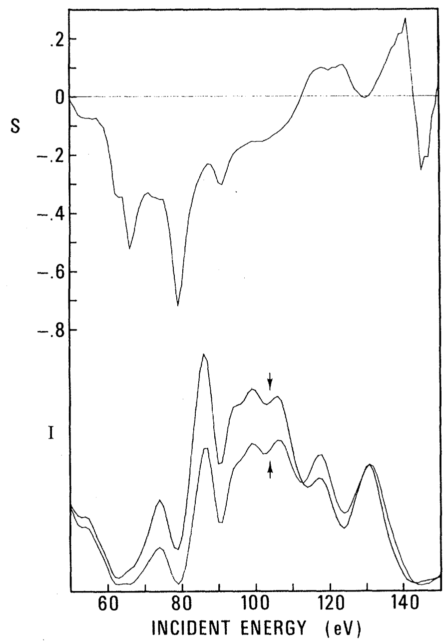

FIG. 2. The spin dependence of the scattering $S(E,\theta)$ is plotted for specular diffraction from W(100) at an angle of incidence of 15°. The scattered intensities resulting from an incident beam consisting of only spin up (↑) or of only spin down (↓) electrons are shown as $I_\uparrow$ and $I_\downarrow$.

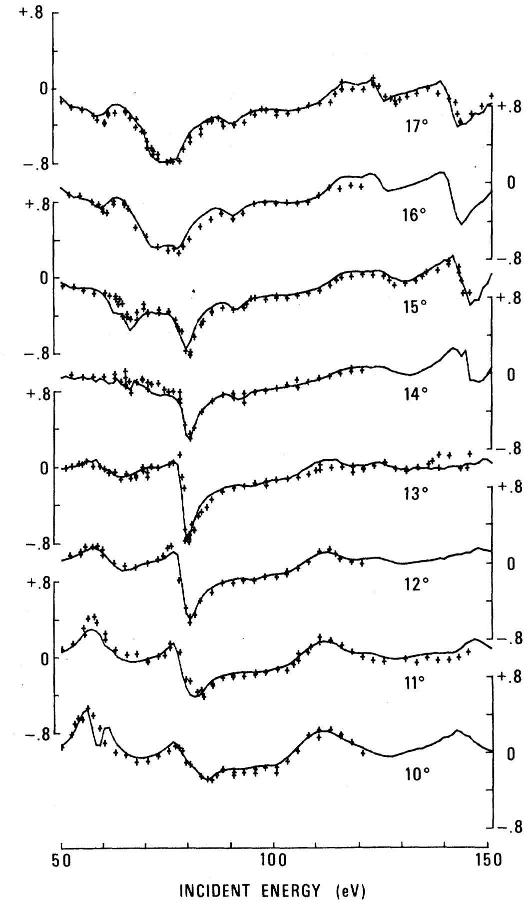

FIG. 3. Our measurements of $S(E,\theta)$ (solid line) are compared to the measurements $\mathbf{P}(E,\theta)$ (crosses) of Ref. 2 of the (00) beam for angles of incidence from 10° to 17°. The scattering plane is in a (010) plane of the crystal. The curves are normalized as described in the text.

polarized incident beam to $P(E,\theta)$ measured by Kalisvaart *et al.*[2] using an unpolarized incident beam and Mott detector.[12] The magnitude of our source polarization, expected[8] to be in the range of 40–50%, was taken to be 43% based upon a comparison[13] of our data and that of Kalisvaart *et al.*[2] at an angle of incidence of 13°. The excellent relative agreement in magnitude of $S(E,\theta)$ and $P(E,\theta)$ at different energies and angles demonstrates their equivalence when the scattering plane is a mirror-symmetry plane of the crystal.

In order to understand this result, we use a general formulation to compare the two measurements. Let $M_{\vec{s}'\vec{s}}(\vec{k}',\vec{k})$ be the amplitude that an electron initially specified by spin $\vec{s}$ and propagation vector $\vec{k}$ is scattered by the crystal into a state specified by $\vec{s}'$ and $\vec{k}'$. Since in spin space this amplitude is a 2×2 matrix, it can in general be expanded in terms of the Pauli matrices,

$$M(\vec{k}',\vec{k}) = A(\vec{k}',\vec{k}) + \vec{B}(\vec{k}',\vec{k})\cdot\vec{\sigma}. \quad (2)$$

Here A is the scalar scattering amplitude and the vector $\vec{B}$ indicates spin flipping in the directions perpendicular to $\vec{B}$. Using density matrix methods, $P(E,\theta)$ and $S(E,\theta)$ are readily determined.[14] Relative to any spin quantization axis $\hat{\eta}$, we find

$$P = \frac{A\vec{B}^* + A^*\vec{B} + i\vec{B}\times\vec{B}^*}{AA^* + \vec{B}\cdot\vec{B}}\cdot\hat{\eta} \quad (3)$$

and

$$S = \frac{A\vec{B}^* + A^*\vec{B} - i\vec{B}\times\vec{B}^*}{AA^* + \vec{B}\cdot\vec{B}^*}\cdot\hat{\eta}. \quad (4)$$

For elastic scattering by nonaligned atoms, spherical averaging over all possible atomic orientations ensures that $S = P$ and each is maximal when $\hat{\eta}$ lies along the normal to the scattering plane.[7] However, in general $S \neq P$ as is apparent from the above expressions. For PLEED, the interaction Hamiltonian including exchange and spin-orbit terms is invariant under the point group of the crystal. Thus, independent of the strength of the interaction, the symmetry of the scattering amplitude, Eq. (2), is determined by the combined symmetry of the crystal and the incident electron. For non-normal incidence onto W(100) in the (010) plane, the C_{4v} symmetry is broken to leave only the two-dimensional reflection group C_s.[15] Provided the crystal wave function is initially invariant under reflection in the (010) plane (in particular, there cannot be ferromagnetic or ferrimagnetic order), the elastic scattering amplitude is invariant under reflection.[16] In this case consideration of the matrix elements of the commutator of the scattering T operator [17] with the reflection operator requires that $\vec{B}$ be normal to the scattering plane; hence $\vec{B}\times\vec{B}^*\cdot\hat{\eta}=0$, and consequently $S(E,\theta)$ and $P(E,\theta)$ are equal and maximal when meaaured normal to the reflection (scattering) plane.[18]

The PLEED measurement determines a new observable which, being sensitive to the gradient of the potential, may offer new insight into the diffraction process. The dramatic change in polarization profiles from one diffraction condition to another suggests that this observable is very sensitive to surface structure; measurements are under way to determine the usefulness of PLEED in surface-structure determination. In summary, we have described the first use of a polarized electron beam as a surface probe, and have applied it to reveal a symmetry principle in low-energy–polarized-electron diffraction.

We wish to thank the Rice University spin-polarization group[1,2] for providing their detailed data for comparison. We are grateful to W. N. Unertl, S. R. Mielczarek, and B. J. Waclawski for their assistance in the developmental stage of the experiment and to C. E. Kuyatt and A. Galejs for designing the electron optics. This work was supported in part by the U. S. Office of Naval Research. One of us (B.I.D.) would like to acknowledge receipt of support from the National Bureau of Standards and the National Research Council of Canada.

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[12]The absolute angle was not measured accurately. The nominal incidence angle of our curves is reduced 2° for the comparison in Fig. 3. A reduction of 1° gives the best agreement with the intensity curves of Debe, King, and Marsh, Ref. 10. The estimated precision in the relative measurement was ± 0.2°.

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