Spin Polarization in Low-Energy Electron Diffraction from W(001)

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A relativistic low-energy electron-diffraction theory has been modified to allow computations for energies up to 200 eV. Its application to W(001) yields intensity profiles in reasonable correspondence with experiment, and pronounced polarization profiles which show encouraging agreement with novel and as yet limited experimental data. A high sensitivity of the polarization profiles to displacement of the topmost layer suggests that spin polarization measurements could be valuable for structure determination of surfaces involving heavy atoms.

Theoretical studies of spin-polarization effects in low-energy electron diffraction (LEED) from the (001) surface of tungsten[1,2] were recently followed by an experiment,[3] in which significant degrees of spin polarization were found for electron energies from 45 to 190 eV at various small angles of incidence. Comparison of these novel data with the theoretical predictions[1,2] is not possible, however, since the latter are confined to energies below about 40 eV. It is the aim of this Letter to report the first theoretical results at higher energies, to compare them with the recent experiment,[3] and to draw conclusions regarding the value of a polarization analysis in surface-structure determination.

The theory underlying the present calculations follows to a large extent a relativistic LEED theory described earlier.[4] In particular, the crystal is assumed to consist of a finite number of monatomic layers, for which the Dirac-equation boundary problem is solved to yield the four-spinor amplitudes and thence the intensities and the spin polarization vectors of the reflected beams as functions of the energy, the polar and azimuthal angles of incidence, and the polarization vector of the (normalized) primary beam. A modification was, however, made in the treatment of the monolayer part of the problem. The Korringa-Kohn-Rostoker-Ziman (KKRZ)[5] type relativistic pseudopotential used in Ref. 4 depends on radii R_l—associated with the spin-up and spin-down atomic phase shifts $\delta_l{}^{\pm}$—which have to be optimized individually in a numerical application of the method. Since calculations up to 200 eV require phase shifts up to about $l=7$ (cf. also Van Hove and Tong),[6] I decided to avoid the resulting radii parameter problem by developing and applying the Dirac-equation–based analog[7] of a KKR-type Schrödinger-equation–based method,[8,9] which has been used with great success for LEED intensity calculations.[10] The effect of thermal lattice vibrations is taken into account by replacing the actual (real) spin-up and spin-down atomic phase shifts $\delta_l{}^{\pm}$ by effective (complex) phase shifts $\tilde{\delta}_l{}^{\pm}$, which are obtained from the $\delta_l{}^{\pm}$ by averaging the corresponding scattering amplitudes over a Debye spectrum.[11]

In the computational application to W(001) the following specific model assumptions have been made. Effective phase shifts $\tilde{\delta}_l{}^{\pm}$, obtained from Mattheiss's muffin-tin potential[12] and with use of the bulk Debye temperature of 380°K, are included up to $l=7$. The number of surface reciprocal lattice vectors (beams) taken into account is 21 below 70 eV and increases successively to 45 at about 180 eV. The number of monolayers ranges from 5 to 9. A real inner potential of 10 eV (cf. Ref. 6) and an imaginary absorption potential of 4 eV were assumed as constant for all energies.[13] The surface barrier is assumed as refracting, but nonreflecting, since the difference between the results from this barrier model and the results from a more realistic smooth barrier can be expected to be generally small above about 40 to 50 eV (see Ref. 8 and Jepsen, Marcus, and Jona, Ref. 9). The topmost interlayer spacing is taken as 100, 95, and 90% of the bulk interlayer spacing.

Computations were carried out for incidence conditions of the recent experiment[3] at energy points separated by 2 eV in general and by 1 eV in regions of rapid change in the spin polarization. For an unpolarized incident beam, the polarization vector of each diffracted beam is found to be parallel or antiparallel to the normal $\vec{n}$ to the scattering plane—defined by the primary beam and the diffracted beam under consideration—if the intersection of the scattering plane with the surface is a mirror symmetry line. The degree of spin polarization is given by the length of the polarization vector with a positive (nega-

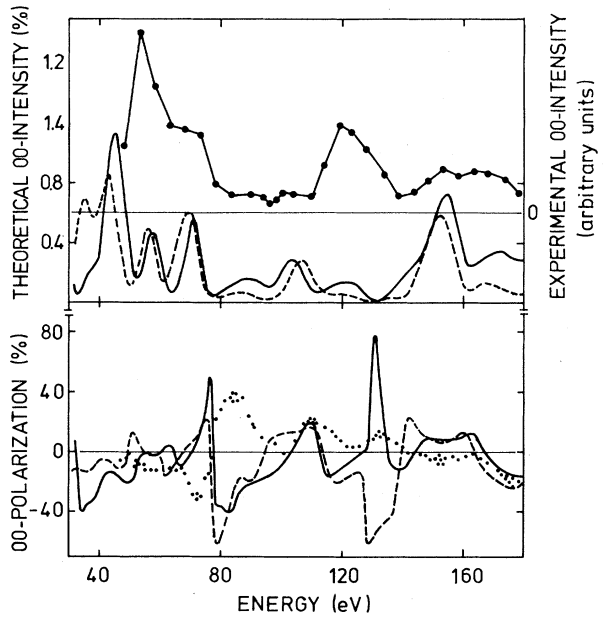

FIG. 1. Intensity and spin-polarization profiles of the (00) beam from W(001) for $\theta = 11°$: theoretical results for no contraction of the top interlayer spacing, dashed line, and 10% contraction, solid line; experimental results (Ref. 3), dotted line. The experimental profiles have been shifted towards higher energies by 3 eV.

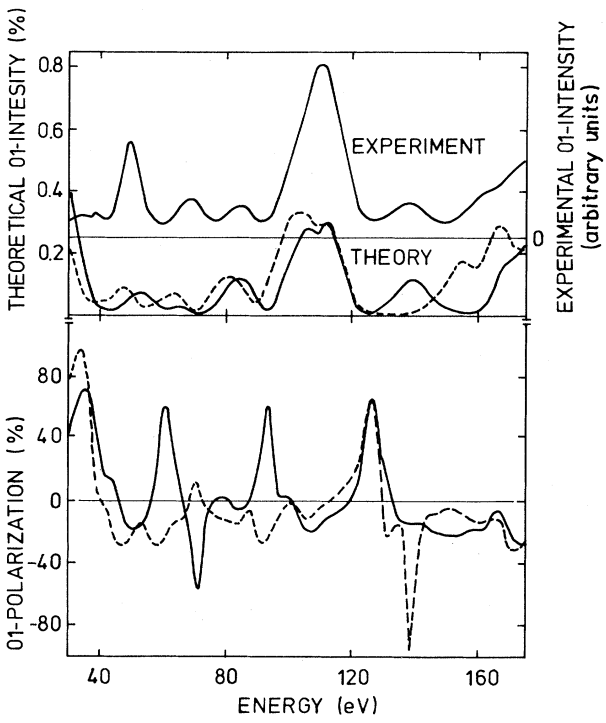

FIG. 2. Intensity and spin polarization profiles of the (01) beam from W(001) for $\theta = 0°$: theoretical results for no contraction, dashed line, and for 10% contraction, solid line; experimental data (Ref. 15), solid line.

tive) sign, if its projection onto $\vec{n}$ is parallel (antiparallel) to $\vec{n}$. Figure 1 shows the intensity and degree of spin polarization versus energy profiles of the (00) beam for an unpolarized primary beam incident along the (10) direction at $\theta = 11°$ with respect to the surface normal. The calculated intensity profiles can be seen to agree with their experimental counterpart with regard to the existence and positions of peaks. As for the relative peak heights, there are some discrepancies, which I ascribe partly to my neglect of the energy dependence of the imaginary part of the inner potential. Some doubt is case on the experimental data of Ref. 3, however, by noting that earlier data[14] give relative peak heights above 110 eV which are in good agreement with our calculated results and at variance with Ref. 3. The intensity profile obtained for a surface with 10% contraction of the top interlayer spacing gives a noticeably better fit than the profile for an uncontracted surface. The polarization profiles for 0% and 10% contraction are qualitatively similar except for a marked discrepancy near 130 eV. Comparison with the experimental polarization profile shows agreement regarding peak positions near 110, 130, and 175 eV. The two polarization peaks near 75 and 85 eV agree very well, if one reverses the sign either of the calculated or of the experimental profile. Bearing in mind the preliminary nature of the experimental data and the theory assumptions concerning the absorption potential and the top layer spacing, I feel that the agreement is very encouraging. In Fig. 2 I present analogous results for the (01) beam for the case of normal incidence. The intensity profile calculated for a 10% contraction of the top interlayer spacing corresponds again better with the experiment.[15] The polarization profiles for 0% and 10% contraction differ drastically from each other near 60, 72, and 92 eV. Together with the polarization results near 130 eV in Fig. 1 this gives rise to the hope that a reliable polarization measurement and calculation would provide a very sensitive means of establishing the top layer spacing. The results obtained for a 5% contraction of the top layer are roughly intermediate between the ones presented above.

The present theoretical results suggest that spin polarization analysis in LEED could be of

use for surface structure determination.[16] An attempt, however, to assess its potential role and to compare it to established surface-structure-determination methods that are based on LEED intensity analysis[17-19] would be premature at this stage. Further calculations and measurements are required.

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Crystal-Electric-Field Effects on the Thermal Expansion of TmSb

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We report a crystal-electric-field effect on the low-temperature thermal expansion of TmSb, manifesting itself as a Schottky-type anomaly. From our results we deduce a volume dependence of the crystal-field level splitting which is in contradiction to point-charge–model expectations.

Because of the crystal-field splitting of the lowest J multiplet of the order of 200 K, rare-earth compounds, such as the rare-earth pnictides, are ideally suited to study effects of the crystal electric field (CEF) on various physical properties in these substances. In particular, TmSb has often been quoted as a model crystal-field-only paramagnetic substance.[1,2] Experimentally determined thermodynamic properties such as the magnetic susceptibility,[1] specific heat, and elastic constants[3] can very well be accounted for by using a crystal-field energy-level scheme determined from neutron-scattering experiments.[2] TmSb crystallizes in the cubic NaCl structure and the Tm^{3+} $J=6$ CEF splitting gives a Γ_1 singlet ground state followed by a Γ_4 triplet at 25 K and a Γ_5 triplet at 56 K, the other excited levels lying higher than 110 K. These higher levels are neglected in our considerations throughout this work.[4]

In this Letter we would like to show the first experimental evidence of a CEF effect on the thermal expansion. We have chosen TmSb because of its properties mentioned above and because elec-