Electron Spin Polarization in Energy- and Angle-Resolved Photoemission from W(001): Experiment and Theory

J. Kirschner
Institut für Grenflächenforschung und Vakuumphysik, Kernforschungsanlage Jülich, D-5170 Jülich, West Germany

and

R. Feder
Theoretische Festkörperphysik, Universität Duisburg GH, D-4100 Duisburg, West Germany

and

J. F. Wendelken
Solid State Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830

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We report on a new phenomenon in solid-state photoemission, the emission of spin-polarized electrons from nonmagnetic crystals by unpolarized light. It becomes observable in fully energy-, angle-, and electron-spin–resolved measurements, the first of which is presented here. It is explained in terms of "final-state spin-polarization effects," a new theoretical approach which is outlined. Fairly good agreement between experimental and theoretical results is found. These polarization effects also suggest a new way for analyzing photoelectron spin polarization.

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We report on the first observation of spin-polarized photoemission from nonmagnetic crystals by unpolarized light. Within the framework of a three-step model of spin-polarized photoemission, there are three possible causes for the observation of polarized electrons:

(i) Initial-state effects, due to preferential population of one spin state in the ground state of a solid, e.g., photoemission from ferromagnets.[1]

(ii) Matrix-element effects, due to dipole selection rules for interband or intraband transitions excited by circularly polarized light.[2-5] A necessary condition for this effect to occur is sufficient spin-orbit coupling, either in the initial-state or the final-state band. The best-known example is the emission of polarized electrons from GaAs.[6]

(iii) Independently of the above processes, unpolarized electrons excited into the upper Bloch states may acquire spin polarization on their way to the detector outside the solid. These polarization effects are due to the very existence of a crystal surface, separating the "Bloch spinor regime" from the "free-electron spinor regime" in which the detector is placed. Matching these spinors across the boundary then may lead to a net spin polarization of the transmitted electrons, if spin-orbit interaction is present in the solid. This "final-state spin-polarization effect," which has hitherto not yet been observed, is the subject of the present note.

With respect to final-state spin polarization, a photoemission experiment bears some resemblance to a time-reversed low-energy electron-diffraction (LEED) experiment.[7,8] However, unlike in LEED, the "primary" electrons in photoemission may occupy a number of different Bloch states with the same energy. Also, source and detector lie on different sides of the crystal-vacuum interface and consequently refer to electrons in two different eigenstates. A common feature is the strong wave-vector dependence of electron spin polarization. Thus, in order to observe final-state spin polarization of photoelectrons, an energy-, angle-, and spin-resolved photoemission experiment is required.

As final-state effects can occur independently and simultaneously with initial-state and matrix-element effects, they will appear in their pure form only in photoemission from nonmagnetic crystals, with linearly polarized or unpolarized light. Matrix-element effects have been shown, by group-theoretical arguments, to exist only for circularly polarized light.[9] Unpolarized light is preferable in the experiment, as otherwise the complex refractive index of metals might introduce unwanted circular light components. In order to avoid complications due to rapid oscillations of the vector potential and nontransversality of the electromagnetic wave in the near-surface region[10] (those might spoil the assumptions of the

dipole approximation), normal incidence of the light is mandatory.

The experimental setup meeting these requirements is schematically shown in Fig. 1. Unpolarized light from a commercial discharge lamp (H, Ly_α) is normally incident onto a W(001) single-crystal surface. The polar angle θ has to be nonzero, since for normal emission final-state polarization effects vanish for reasons of space and time symmetries (e.g., Kramers degeneracy). The value $\theta \sim 70°$ was determined by geometrical restraints. The angular resolution $\Delta\theta \sim \pm 3°$ was estimated from the electron optics. It was checked by the measurement of the emission from surface resonances strongly peaked in the ⟨100⟩ direction.[11] While these resonances have been observed at near-normal emission, no such surface-state emission has been observed at $\theta \cong 70°$. The crystal temperature during the measurements was between 50 and 100 °C, so that surface reconstruction should be negligible.[12] The electrons leaving the surface are accelerated and focused by a four-element zoom lens into the energy analyzer, which is a section of a cylindrical mirror analyzer (CMA) operating in second-order focusing conditions with a constant pass energy (energy resolution about 0.3 eV). In order to reduce contributions from secondary electrons and inelastically scattered photoelectrons, the energy window was set closely below the Fermi energy as measured from intensity spectra.

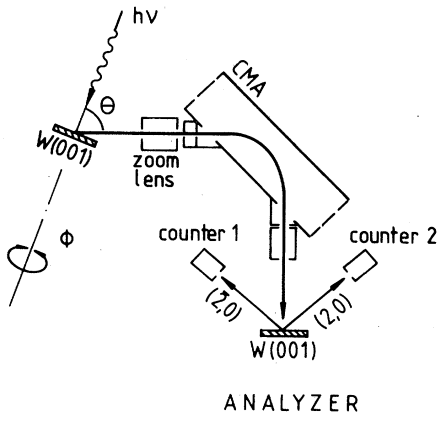

FIG. 1. Schematic of the experimental setup. Unpolarized uv light (H, Ly_α) from a discharge lamp impinges normally onto a W(001) crystal. Photoelectrons are collected at the angle $\theta = 70°$ and energy analyzed in a section of a cylindrical mirror analyzer. The outgoing electrons are spin analyzed by means of electron diffraction from a W(001) surface as described previously (Ref. 13).

After passing the CMA, the electrons are accelerated onto the second W(001) crystal (normal incidence) and the intensity asymmetry in the Channeltron counters 1 and 2 is a measure of the component of the polarization vector normal to the plane of Fig. 1. The analyzer part of the present experiment is identical (except for the zoom lens) to the spin detector part of the double diffraction experiment described previously.[13] It has a measured polarization sensitivity of 0.28 ± 0.02 and an efficiency of 1.1×10^{-3}. The experimental data have been taken in the form of azimuthal scans (rotation diagrams) at fixed photon energy $h\nu = 10.2$ eV, fixed polar angle $\theta \cong 70°$, and kinetic energy $E \sim E_F - 0.5$ eV. In the intensity rotation diagram, structures are to be expected because of the anisotropic emission from the bulk crystal, modified by diffraction at the surface. By contrast, in the absence of final-state spin-polarization effects, the rotation diagram for the polarization, which is the normalized difference of spin-up and spin-down electrons, should show a constant zero, as the intensity variations cancel out. Conversely, the pres-

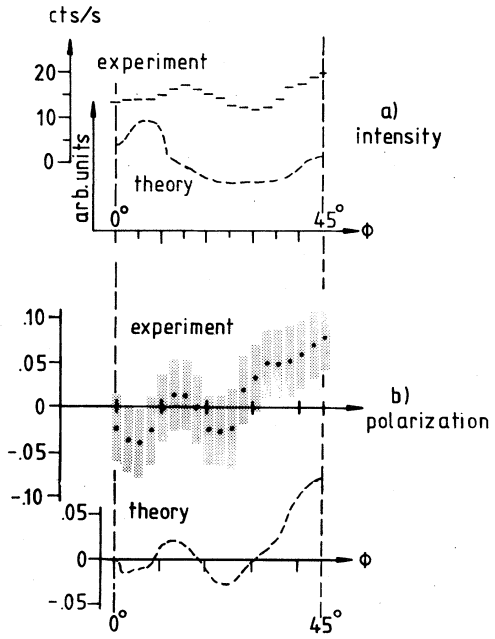

FIG. 2. (a) Experimental and theoretical intensity rotation diagrams (azimuthal scans) at fixed polar angle and kinetic energy. (b) Experimental and theoretical polarization rotation diagrams, showing the existence of final-state spin-polarization effects in photoemission from nonmagnetic solids by unpolarized light.

ence of structure in the polarization rotation diagram, connected to the crystal symmetries, is a direct proof for the existence of final-state spin-polarization effects as postulated above. The experimental data in Fig. 2(b) show that this is indeed the case.

A complete one-step theory of spin-polarized photoemission not yet being at hand (for the spinless case see, e.g., Refs. 14–16), we adopted the following theoretical final-state model: Each (spin) pair of the Bloch spinors corresponding to the values $(E, k_{\parallel})$ ($k_{\parallel}$ modulo reciprocal surface vectors) determined by the detector is assumed to be populated by unpolarized light incoherently, so that its spin polarization is zero. These unpolarized Bloch electrons, incident from the interior of the crystal, are diffracted at the surface, i.e., there are Bloch electrons reflected back into the crystal and plane-wave electrons (beams) going out into the vacuum, out of which one beam propagates in the direction of the detector. (In contrast to LEED, there are no incident plane-wave electrons in this case). If the Bloch spinors involve spin-orbit interaction, this latter beam can be spin polarized as a consequence of the spin-dependent matching conditions. The Bloch spinors are obtained by diagonalizing the bulk single-layer transfer matrix obtained by means of a spin-dependent multiple-scattering formalism.[17,18] Matching each incident Bloch spinor and the corresponding reflected Bloch spinors with the transmitted plane-wave spinor field at the surface then yields the spinor amplitudes, and thereby the particular density matrix, for the beam selected by the detector. From the total density matrix of the beam, obtained by summing—with equal weights—over these particular density matrices, the intensity and spin polarization of the photocurrent accepted by the detector is calculated. Emission from surface states is not included in the model since these states are not excited by s-polarized light.[11] Surface emission is neglected in view of the large mean free path at the present low energies. Numerical calculations have been performed for a truncated bulk crystal with a nonreflecting surface barrier, a real inner potential $V_r = 14$ eV, and an imaginary part $V_i = 0$ or $V_i = 0.25$ eV. We chose an ion-core scattering potential due to Mattheiss, as suggested by a recent elastic reflection coefficient study.[19,20]

A comparison of experimental and theoretical results is shown in Fig. 2. The experimental data were measured several times over a range of more than 100° in φ, and, by virtue of the C_{4v} symmetry of the surface, have been folded back into a 45° segment in order to improve the statistics in the polarization measurement. The horizontal extension of the symbols represents the azimuthal angular uncertainty, while the vertical extension indicates the statistical error (variance σ). The absolute uncertainty of the polarization zero is about ± 0.02. The theoretical data were calculated for a kinetic energy of 5.0 eV, with an imaginary part $V_i = 0.25$ eV. The intensity rotation diagrams in Fig. 2(a) both exhibit azimuthal structure, the agreement between experiment and theory being rather poor. This is no surprise, however, since in the calculations, aimed at spin-polarization effects, the optical-transition matrix element was set constant. A complete agreement would thus be accidental. The spin polarization, on the other hand, is a *normalized* quantity, so that the matrix-element contribution is divided out. For the polarization rotation diagrams in Fig. 2(b) we note fairly good agreement, both in the general structure and in the absolute numbers. This agreement was checked to persist for a variation of the imaginary part of the inner potential within reasonable limits. The polarization vector is in general not normal to the scattering plane, as is well known from spin-polarized LEED[8] [only for $\varphi = 0°$ and $\varphi = 45°$ in the present case, measured from the (10) direction]. The figure therefore shows only its component normal to the emission plane, defined by the surface normal and the direction of the outgoing electrons.

From the experimental and theoretical results above we conclude that a final-state spin polarization of photoelectrons emitted from nonmagnetic solids by unpolarized light does indeed exist, and that it can successfully be described by the theoretical approach outlined. The absolute magnitude of the polarization is not very large in the present case, a systematic search presently being hampered by experimental constraints. Note, however, that in electron diffraction polarization up to almost 80% has been found.[8,21] Large effects in final-state photoelectron spin polarization are thus to be expected, too.

The present results may be considered important in several aspects:

First, they demonstrate the feasibility of fully energy-, angle-, and spin-resolved photoemission experiments when using the LEED spin polarization detector.[13]

Second, as spin polarization in electron diffrac-

tion is known to be sensitive to structural parameters,[21] its analysis may add a further dimension to structure determination via photoelectron diffraction. The main constraint at present, that of low uv light intensities, should be much relaxed by the future use of synchrotron light.

Third, final-state polarization effects have to be taken into account in all energy- and angle-resolved photoemission experiments, including those with magnetic materials, if spin-orbit interaction is present. In the latter case, they may lead to distortions of the polarization vector as measured outside the crystal, both in magnitude and orientation.

Fourth, scattering of polarized electrons gives rise to characteristic intensity asymmetries in the presence of spin-orbit coupling. This is well known in atomic physics,[22] and in LEED has first been observed (and used) by Kirschner and Feder.[13] Thus, if polarized electrons are generated inside the crystal, be they wanted or not (e.g., by a circular component of the electromagnetic wave inside the crystal), the intensity measured outside may show characteristic asymmetries under complementary angles and/or upon reversal of the electron spin orientation. On the other hand, if the final-state polarization effects are known, either by experiment or by theory, the measurement of the *polarization* inside the crystal is reduced to simple *intensity* measurements outside the crystal. In this sense, the crystal to be studied may serve as its own spin-polarization analyzer.

Fifth, as the phenomenon of final-state spin polarization is very general in nature—being due to the spin-dependent matching conditions between a Bloch spinor regime on the one side and a free-electron spinor regime on the other side of the crystal-vacuum interface—it is not restricted to photoemission, but may occur in other electron-emission spectroscopies as well (e.g., field emission).

One of us (J.F.W.) was a guest scientist (1979–1980) at Institut für Grenflächenforschung und Vakuumphysik, Kernforschungsanlage Jülich, Jülich, West Germany.

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