Experimental Verification of a New Spin-Polarization Effect in Photoemission: Polarized Photoelectrons from Pt(111) with Linearly Polarized Radiation in Normal Incidence and Normal Emission

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A theoretical prediction of a new spin effect by Tamura, Piepke, and Feder has been experimentally verified: Photoelectrons can be polarized even if the photoemission is performed with linearly polarized radiation and even if it is studied in the highly symmetrical setup of normal incidence and normal emission. Radiation with energies between 21 and 22.4 eV ejects photoelectrons from Pt(111) with a degree of polarization between 10% and 40%. The spin direction coincides with a plane parallel to the surface and changes its sign when the crystal is rotated by 60° about the surface normal.

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The existence of spin-polarized photoelectrons obtained with circularly polarized radiation from unpolarized targets (free atoms, molecules, adsorbates, and nonferromagnetic solids) has been proved to be a common phenomenon rather than exceptional. The spin-polarization information is an important tool to characterize the symmetry of the states and bands involved, i.e., to perform a symmetry-resolved band mapping of solids or a characterization of quantum numbers, dipole matrix elements, or phase-shift differences of wave functions in the photoionization of free or adsorbed atoms.

That linearly or even unpolarized radiation is able to eject polarized electrons in photoemission of ferromagnetic solids, in which the photoelectron polarization is primarily an effect of the initial states, is well known. In photoionization of free unpolarized atoms and molecules or in photoemission of nonferromagnetic solids it has been found in angle-resolved off-normal photoelectron emission as a final-state effect; in these cases it is a consequence of a quantum-mechanical interference between different photoelectron partial waves in atomic photoionization, or due to spin-dependent photoelectron diffraction or phase-matching conditions at the solid-vacuum interface in photoemission. In spin-resolved photoemission from noncentrosymmetric crystals spin polarization can arise from difference in spin-up and spin-down conduction-band hybridization with valence p states and from surface-transmission effects.

Normal incidence of linearly polarized light along centrosymmetric cubic crystals and normal photoelectron emission was, however, commonly assumed to yield no spin polarization at all. Very recently, Tamura, Piepke, and Feder refuted this belief and predicted normal-emission photoelectron spin polarization by linearly polarized light for (111) surfaces of centrosymmetric cubic crystals. Their prediction is based upon a one-step photoemission theory using a relativistic multiple-scattering formalism and they identify the spin-orbit interaction in the half-space initial states as its main cause. In general, symmetry arguments show that for this special geometry electron spin polarization P can be nonzero. Because of the invariance of the total system (semi-infinite crystal with surface, incident light, electron detection direction) under a symmetry operation, photoelectrons can only be polarized perpendicular to a mirror plane. This implies \( P = 0 \) for surfaces with \( n \)-fold rotation axes associated with the point groups \( C_{nv} \), \( n = 2, 4, \) and 6, because there are two or more mirror planes perpendicular to one another. For \( n = 3 \) there is no such a restriction. For photoexcitation in the bulk of a centrosymmetric crystal, however, space inversion and time reversal imply \( P = 0 \). A nonzero \( P \) appears to be possible therefore in cases in which, for \( n = 3 \), a three-step model is not applicable [like emission from clean surface states or via evanescent states (band-gap emission) from centrosymmetric crystals]. The present Letter is the first experimental evidence that such a spin-polarization effect exists for normal incidence of linearly polarized light and normal photoelectron emission. Along the \( \Lambda \) direction of Pt(111) we find electron spin polarization ranging up to more than 30% for photon energies between 21 and 22.4 eV.

The experiments were performed with linearly polarized synchrotron radiation from the BESSY storage-ring plane and with the 6.5-m normal-incidence monochromator in an apparatus described previously. The Pt crystal surface coincided within 0.5° with the (111) direction and was aligned within 0.3° with the direction of the incident light. Photoelectrons emitted normally to the surface were collected by the electron spectrometer within an angular cone of ± 3°. Phonon effects were minimized by our keeping the crystal at a temperature below 50 K during the measurements. The target preparation was performed as usual and included Ne⁺ bombardment, oxygen heating, and flashing. The surface was characterized by Auger spectroscopy and LEED. LEED was also applied to determine the mirror planes of the Pt(111) crystal.
components were measured simultaneously in the Mott
detector: one parallel to the surface normal (coinciding
with the photon momentum), and the other parallel to
the crystal surface but at 45° with respect to the
storage-ring plane (and thus to the E vector of the
linearly polarized radiation).\textsuperscript{12,13} Influences of experi-
mental asymmetries have been eliminated in the data
presented by the use of four additional detectors in for-
motion scattering directions in the Mott detector,\textsuperscript{12,13} as
well as by the use of count rates in the Mott detector
when unpolarized electrons are spin analyzed there.

A photoemission spectrum of Pt(111) obtained with
linearly polarized radiation of photon energy $h\nu = 21$ eV
is given in Fig. 1(a). Two peaks were obtained: The
first at 1.5 eV below $E_F$ (peak 1) corresponds to transi-
tions from the upper initial bands of the bulk band struc-
ture close to $\Gamma$; the second one at 4 eV below $E_F$ (peak
2) is correlated with a transition from the lower $\Delta_{\perp,5}$
band (see, e.g., Eyers et al.\textsuperscript{12}; for the bulk band structure
see Fig. 3 of the present Letter). For both peaks a spin
analysis of the photoelectrons was made: The combina-
tion of photoelectron intensity $I$ and polarization $P$
according to $I_{\pm} = I(1 \pm P)^2$ yields the partial intensities
$I_{\pm}$, characterizing the number of photoelectrons with
spin parallel or antiparallel to the Mott-detector analyz-
ing axis. This procedure shows that the spin-polarization
component perpendicular to the Pt(111) surface vanishes
for both peaks within the experimental uncertainty (3%).
The same is true for the component in the Pt(111) sur-
face plane for peak 2 [$I_{\pm} = I_{-}$ in Fig. 1(a)], whereas
peak 1 clearly demonstrates the existence of polarized
electrons ($I_{\pm} \neq I_{-}$) (it corresponds to about 20% spin
polarization). All the findings are reproduced by a re-
cent calculation of Tamura and Feder\textsuperscript{15} given in Fig.
1(b). The results can be compared despite the fact that
they were obtained for photon energies differing by 1 eV,
and the calculation neglects a self-energy correction of
about the same magnitude (0.75 eV, see Wern et al.\textsuperscript{16}).
While we find good qualitative agreement for the total
intensities (a convolution of the calculated spectra with
the experimental resolution of 250 meV will mainly
broaden peak 1), the polarization values in peak 1 and
peak 2 are in excellent agreement with the theoretical re-
results.

Tamura, Piepke, and Feder predicted that the spin-
polarization vector is in a plane parallel to the (111) sur-
face and rotates by an angle $-\alpha$ upon rotation of the
light polarization by $\alpha$ (see Fig. 1 of Ref. 10). In our ex-
perimental geometry the analyzing axis of our spin Mott
detector and the E vector of the synchrotron radiation
are fixed in space, and so the crystal surface is rotated
about the surface normal. For this geometry, the
Tamura-Piepke-Feder prediction means that the spin pol-
larization reverses sign when the crystal is rotated by
60°.

For further elucidation of the effect, the dependence of
the spin polarization on the rotation angle $\omega$ of the crys-
tal about the surface normal has been measured. The
upper part of Fig. 2 shows the angular dependence of the
ratios of count rates $N_B/N_D$ and $N_A/N_C$ directly mea-
sured by the detectors of the Mott detector. $N_B/N_D$
corresponds to the out-of-plane [Pt(111) plane] polariza-
tion, and $N_A/N_C$ to the in-plane polarization. The corre-
sponding detector setup is given in the schematic dia-
gram. While there is no angular dependence of $N_B/N_D$
within the experimental uncertainty, $N_A/N_C$ demon-
strates a clear sinusoidal shape as a function of the angle
$\omega$. Instrument-related asymmetries (due to different
detector efficiencies, scattering solid angles, etc.) are re-
sponsible for the fact that the $\omega$-averaged asymmetry is
different from 1, but they are independent of $\omega$. From
the measured asymmetry data one obtains the spin-
polarization component $P_{\omega}$ in the crystal plane. These
values are presented in the lower part of Fig. 2. For

![FIG. 1. Photoelectron spectrum obtained from normal in-
cidence of linearly polarized light and normal photoelectron
emission. The partial intensities $I_{\pm}$ correspond to spin
directions parallel and antiparallel to a trace of a nonmirror
plane in the Pt(111) surface which is rotated by 30° with
respect to a trace of a mirror plane in the Pt(111) surface and
by 45° with respect to the E vector of the incident light. (a)
Experimental result (energy resolution $\Delta E = 250$ meV). (b)
Calculation of Tamura and Feder (Ref. 15).]
$\omega = 0^\circ$ the crystal mirror plane and the storage-ring plane are parallel. The data show a periodicity of 120° and can be well described by a sinusoidal behavior (continuous line in Fig. 2). We find maximum polarization for $\omega \approx 15^\circ$, i.e., a spin rotation of $3\omega \approx 45^\circ$, which agrees with our experimental setup of the analyzing axis of the Mott detector ($45^\circ$ with respect to $E$) as discussed above.

In addition to these results the measurements were extended to photon energies $h\nu$ between 20 and 22.4 eV. Figure 3 (left-hand side) shows the spin polarization $P_{xy}$ of a constant-initial-state spectrum obtained for the initial energy 1.5 eV below $E_F$. Below $h\nu = 20.8$ eV we do not find a spin polarization within the experimental accuracy, while the polarization seems to increase gradually with increasing photon energy except for a breakdown at 21.8 eV. For a rough interpretation of the results we compare the energy dependence of the spin polarization with a bulk band structure of Tamura and Feder, which is shifted by the self-energy correction value of 0.75 eV (Ref. 16) towards higher energies. From the comparison we find that the polarization onset at 20.8 eV is correlated with an onset of transitions into the narrow band gap between 19.3- and 20.2-eV final energy. The breakdown of the polarization occurs at that point of the band structure where the $\Lambda_3$ bands meet the $\Gamma$ point. All these findings are in agreement with the interpretation that transitions into the band gap produce the spin polarization. The threefold symmetry of Pt(111) and the surface are thus observed, because the electron emission occurs via $\Lambda_3$ evanescent states.\textsuperscript{17}

Summarizing, we have reported experimental evidence of a new spin-polarization effect. Spin-polarized photoelectrons are obtained with linearly polarized light in the highly symmetrical experimental setup of normal light incidence and normal photoelectron emission from Pt(111). The spin-polarization direction is parallel to the surface. We observe polarization up to more than 30%, a sinusoidal behavior with the rotation angle $\omega$ about the Pt(111) surface, and a 120° periodicity according to the threefold symmetry of Pt(111) and the inclusion of the surface. The observed surface effect is obviously of the same nature as that predicted by Tamura, Piepke, and Feder.\textsuperscript{10}
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1For a review see U. Heinzenmann, Phys. Scr. T17, 77 (1987), and references therein.
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