Spin-orbit coupling, matrix elements, and scattering effects in angle-resolved photoelectron spectroscopy

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Outline

- One-step model of spin-resolved photoemission
  - Dirac equation, multiple-scattering theory, …

- Spin-orbit coupling and spin polarization
  - Rashba effect: Au(111)

- Magnetic dichroism
  - Probing spin-orbit coupling: Fe(110)

- Photoelectron diffraction
  - Spin-dependent final-state scattering: Fe(001)

- Spin motion of photoelectrons
  - Spin precession in ultra-thin magnetic films: Fe/Pd(001)

- Summary
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Summary
Photoelectron spectroscopy

Spin- and angle-resolved photoelectron spectroscopy (SPARPES)

Incident photon
- Energy
- Polarization
- Incidence direction

Photoelectron
- Energy
- Detection direction
- Spin polarization

Set-up

Solid
- Geometric structure
- Electronic structure
- Magnetic structure

Energy-space diagram

Photocurrent

Kinetic energy

Binding energy

DOS

Solid
One-step model for SPARPES

Excitation and transport = coherent process

**Relativistic description** (Dirac equation)
Orbital and spin degrees of freedom treated on equal footing

Spin density matrix
\[ \rho_{\tau\tau'} \propto -\text{Im} \langle \Psi_{\tau}(E) | \Delta G^+(E - \omega) \Delta \dagger | \Psi_{\tau'}(E') \rangle, \tau = \uparrow, \downarrow \]

\( \Psi_{\tau} \) time-reversed (SP)LEED state

Photocurrent \( j = \text{tr} \rho \)
Photoelectron spin polarization \( \vec{P} = (\text{tr} \vec{\sigma} \rho)/(\text{tr} \rho) \)

Sudden approximation
Multiple-scattering theory

Stepwise building up the entire system
Separation of geometry and single-site scattering properties

Flexible
- Low-dimensional systems: Surfaces, thin films, defects, adatoms, …
- Boundary conditions

Efficient
Accurate
Relativistic theory

**Dirac equation** (instead of the Schrödinger equation)

\[
[c\vec{\alpha} \cdot \vec{p} + \beta mc^2 + V_{\text{eff}} - \epsilon_i] | \Phi_i\rangle = 0
\]

\[
\vec{\alpha} = \begin{pmatrix}
0 & \vec{\sigma} \\
\vec{\sigma} & 0
\end{pmatrix}
\]

\[
\beta = \begin{pmatrix}
1 & 0 \\
0 & -1
\end{pmatrix}
\]

**Spin-orbit coupling** (SOC)

Pauli form (non-relativistic)

\[
H_{\text{SOC}} = \frac{\hbar^2}{2m^2c^2}\vec{s} \cdot \left[\vec{\nabla} V(\vec{r}) \times \vec{p}\right]
\]

Kohn-Sham potential (DFT; LSDA, GGA, SIC)

Self-energy corrections

Surface potential

Additional SOC

‘Atomic’ potential

Strong SOC
Korringa-Kohn-Rostoker method

Green function

$$G^{ij} = -i \kappa \sum_{\Lambda} |Z^i_{\Lambda}\rangle \langle J^j_{\Lambda} | \delta_{ij} + \sum_{\Lambda\Lambda'} |Z^i_{\Lambda}\rangle \tau^{ij}_{\Lambda\Lambda'} |Z^j_{\Lambda'}\rangle$$

Single-site contribution

$$\Lambda = (\kappa, \mu) \equiv (j, m_j) \equiv (l, m, s)$$

Spin-angular quantum number

Multiple-scattering contribution

Scattering-path operator

Dyson equation

$$G = G_0 + G_0 V G$$

Layer KKR

Layer = fundamental object

Change of basis

- Spin-angular basis for single-site properties
- Plane waves for interlayer scattering
omni2k program package

... for electron spectroscopies

- **Spin-polarized relativistic layer-KKR (SPRLKKR)**
- **Systems**
  - Bulk, surfaces, films, adatoms, nanocontacts
- **Modes**
  - Band structure, local spectral density, DOS, magnetic anisotropy
  - SPLEED
  - Photoemission (valence bands and core levels)
  - Spin-dependent transport in nanostructures (STM, …)
- **Green function**
- **Disorder**
  - Coherent potential approximation
  - Adaptive wave-vector mesh for Brillouin zone sampling
  - Object-oriented (C++) and modular
  - User friendly (?!?) and free (henk@mpi-halle.de)
Ab initio SPARPES calculations

Calculation scheme
1. DFT+LSDA calculations (e.g. KKR code of Arthur Ernst)
   • ground-state potentials
2. Compare band structures, LDOS obtained by omni2k with original ones
   • PE- and ab initio calculations on equal footing
3. SPARPES calculations (two modes: GF and PEOVER)

‘Free’ parameters
• Optical potential (local and energy-dependent self-energy)
• Size of basis set (spin-angular functions, plane waves)
• System size (e.g. number of layers contributing to the photocurrent)
• Mesh size for Brillouin sampling
Intrinsic spin-orbit effects

Present in the ground state

- Band gaps and hybridization

Splitting of bulk electronic states
- In non-centrosymmetric solids
- Bulk inversion asymmetry (BIA)
- Dresselhaus effect

Splitting and spin polarization of surface states
- Structural inversion asymmetry (SIA)
- Rashba-Bychkov effect

Examples
- Rashba-Bychkov effect in Au(111)
- Magnetic dichroism in Fe(110)
Extrinsic spin-orbit and scattering effects

‘Matrix element effects’ – due to the measurement

Extrinsic spin-orbit effects
Spin polarization of photoelectrons
• Optical orientation (Fano)
  ➢ Spin polarization with linearly polarized light (Feder, Tamura, JH)
  ➢ Magnetic dichroism

Examples
➢ Rashba effect in Au(111)
➢ Magnetic linear dichroism in Fe(110)

Scattering effects
Examples
➢ Spin-dependent photoelectron diffraction in Fe(001)
➢ Spin precession and relaxation (‘spin motion’) in Fe-Pd(001)
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Rashba-Bychkov effect

Band splitting by spin-orbit coupling in a two-dimensional electron gas (2DEG)

Interface in a semiconductor heterostructure

Metal surface

Conduction band

Valence band

GaAlAs

GaAs

2DEG

Surface state

Band gap

Surface potential

Structural asymmetry

Additional SOC
Rashba-Bychkov effect

Free Electrons in two dimensions (2DEG)

Dispersion without SOC  \[ E_{\pm} = \frac{\hbar^2}{2m} \vec{k}_{\parallel}^2 \]

Spin-orbit coupling

\[ H_{\text{SOC}} = \frac{e\hbar}{mc} \vec{g} \cdot \vec{B} \]

\[ H_{\text{SOC}} = \frac{\hbar^2}{2m^2c^2} \vec{g} \cdot \left[ \vec{\nabla}V(\vec{r}) \times \vec{p} \right] \]

\[ H_{\text{SOC}} = \frac{\hbar^2}{2m^2c^2} \int \frac{1}{r} \frac{dV}{dr} \vec{g} \cdot \vec{l} \]

Dispersion with SOC

\[ E_{\pm} = \frac{\hbar^2}{2m} \vec{k}_{\parallel}^2 \pm \gamma_{\text{SOC}} |\vec{k}_{\parallel}| \]

Splitting
Isotropic 2DEG versus Au(111) surface state

Anisotropic spin polarization

\[
\begin{align*}
2\text{DEG} & \quad \begin{cases} 
P_{\pm}^{\text{tan}} & = \pm 1 \\
P_{\pm}^{\text{rad}} & = 0 \\
P_{\pm}^{z} & = 0 
\end{cases} \\
\text{Au}(111) & \quad \begin{cases} 
P_{\pm}^{\text{tan}} & = \alpha_{\pm} \\
P_{\pm}^{\text{rad}} & = \beta_{\pm} \cos 3\varphi \\
P_{\pm}^{z} & = \gamma_{\pm} \cos 3\varphi 
\end{cases}
\]

- Complete
- Normal to the wave-vector
- Within the surface plane

Value and sign of $\alpha$, $\beta$ und $\gamma$?

Threefold rotational symmetry
Au(111) surface state

Dispersions and momentum distribution

• Parabolic dispersion
• Splitting by SOC

Fermi surface

L point

Momentum distribution at $E_F$

Dispersion

Ab initio theory

Experiment

J. Osterwalder et al. (Zürich)
Au(111) surface state

Spin polarization of photoelectrons

Binding energy 0.17 eV

- Sign
- Threefold symmetry
- Order of magnitude

Ab initio theory

Experiment

\[ P_{\text{tan}} = \alpha \pm \]
\[ P_{\text{rad}} = \beta \pm \cos 3\varphi \]
\[ P_z = \gamma \pm \cos 3\varphi \]
Summary

Agreement of dispersion and splitting
Spin polarization

<table>
<thead>
<tr>
<th>Ground state</th>
<th>Model calculation</th>
<th>$p_{\tan}$</th>
<th>$p_{\text{rad}}$</th>
<th>$p_{z}$</th>
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<tbody>
<tr>
<td>$\text{Ab initio calculation}$</td>
<td>0.97</td>
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<td>PE calculation</td>
<td>0.6</td>
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<tr>
<td>PE experiment</td>
<td>0.4</td>
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<td>$&lt; 0.05$</td>
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</tbody>
</table>

Photoelectron spin polarization

Strongly reduced by matrix element effects
Depends on the set-up
- ‘Good’ set-up: p-polarized light
- ‘Bad’ set-up: circularly polarized light

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Magnetic linear dichroism

Magnetic dichroism
Change of the photocurrent upon magnetization reversal $j(\vec{M}) \neq j(-\vec{M})$

Magnetic linear dichroism
Normal emission
p-polarized light
Magnetization in a mirror plane
$\mapsto$ reduction of symmetry ($2mm \rightarrow m$)

‘Golden rule’ of magnetic dichroism
SOC produces spin polarization of photoelectrons along the magnetization
$\mapsto$ magnetic dichroism
NB: MD does not probe magnetism but spin-orbit coupling
Group-theoretical analysis

Double group
Two representations

\[ \gamma_+ = \Sigma^{1\uparrow} \oplus \Sigma^{2\downarrow} \oplus \Sigma^{3\uparrow} \oplus \Sigma^{4\downarrow} \]
\[ \gamma_- = \Sigma^{1\downarrow} \oplus \Sigma^{2\uparrow} \oplus \Sigma^{3\downarrow} \oplus \Sigma^{4\uparrow} \]

Photocurrent (Fermi’s `golden rule‘)

\[ j(\pm \vec{M}) = \sin^2 \vartheta \left( |M^{1+++}|^2 + |M^{1---}|^2 \right) \]
\[ + \cos^2 \vartheta \left( |M^{3+++}|^2 + |M^{3---}|^2 \right) \]
\[ \pm \frac{\sin 2\vartheta}{2} \Sigma \left( M^{1+++} M^{3+++} - M^{1---} M^{3---} \right) \]

Hybridization by SOC

MD does not probe areas of large magnetism but of large hybridization

A. Rampe, G. Güntherodt, D. Hartmann, JH, T. Scheunemann, R. Feder,
Fe(110) – hybridization

Focus on $\Sigma^1 \uparrow \oplus \Sigma^3 \uparrow$ and $\Sigma^1 \downarrow \oplus \Sigma^3 \downarrow$

Change of orbital character by SOC
Fe(110) – Magnetic linear dichroism

Dichroism at $E_b = 0.5$ eV

$S_{MLD}^{MD} = j(\vec{M}) + j(-\vec{M})$

$D_{MLD}^{MD} = j(\vec{M}) - j(-\vec{M})$

MD allows to identify areas of hybridization induced by SOC
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Photoelectron diffraction

Emission from core levels $\leftrightarrow$ element-specific

Constant kinetic energy
Polar-angle scans

Information on
- Surface geometry
- Surface magnetism

Spin-dependent scattering in the final state (photoelectron)

Example: Dichroic PED from 3p levels in Fe(001)

Refining the theory

Emission from Fe(001)-3p

Experiment

Atomic model
Semi-analytic
Single emitter
No scattering

Symmetry
\( j(+M, \theta) = j(-M, -\theta) \)

\[ j(+M) - j(-M) \]
Refining the theory

Experiment

Solid
Numerical
All Fe sites emit
No scattering
No surface barrier

Atomic model
Refining the theory

Experiment

Solid
Surface barrier

Solid

Intensity (arb. units)

Experiment

Theory 'ropes'

Theory 'ropes

-0.5 0.5

-0.5 0.5

-0.5 0.5
Refining the theory

Experiment

Solid
Multiple scattering

Increased forward scattering at 45°

Solid

Strong spin-dependent multiple scattering in the final state
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Spin motion

Motion of electrons in a magnetic system

**Precession**: Phase difference between up- & down wave-functions

**Relaxation**: Inelastic processes

**Experiments**:
- Transmission through a free-standing magnetic film
- SPLEED

**New approach**: Spin-resolved ARPES
1. SOC produces spin-polarized photoelectrons in the sample
2. Transmission through the magnetic film
3. Spin-resolved detection

Spin motion = Spin polarization in dependence of film thickness $d$
Spin motion of photoelectrons

Fe/Pd(001) – Variation of the Fe-film thickness

Deviations from model calculations
Effect of the Fe electronic structure

Spin motion of photoelectrons

Spin motion provides information on unoccupied electronic structure in magnetic films

Band gaps: rapid change of the spin polarization

Evolution of a band gap with film thickness

Spin motion of photoelectrons

Model

Ab-initio photoemission

Band gap in Fe film

Spin polarization

Wave-number

Kinetic energy

Intensity
**Summary**

### Spin-orbit effects

<table>
<thead>
<tr>
<th>Material</th>
<th>Intrinsic SOC effect</th>
<th>Extrinsic SOC effect</th>
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</thead>
<tbody>
<tr>
<td>Au(111)</td>
<td>Rashba effect in the L-gap surface state</td>
<td>photoelectron spin polarization due to SOC</td>
</tr>
<tr>
<td>Fe(110)</td>
<td>Band gaps and hybridization</td>
<td>Magnetic dichroism (transition matrix elements)</td>
</tr>
</tbody>
</table>

### Scattering effects

- Photoelectron diffraction in Fe(001)
- Spin-dependent scattering in the final state

- Spin motion in Fe/Pd(001)
- Spin-dependent scattering in the final state

Further...

- Matrix element effects in … from the Ni(111) surface

M. Mulazzi et al, PRB 74 (2006) 035118
Thanks

<table>
<thead>
<tr>
<th>MPI Halle</th>
<th>A. Ernst, P. Bruno</th>
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<tr>
<td>University Halle</td>
<td>and International Max Planck School on Research and Technology of Nanostructures</td>
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<td>P. Bose, Th. Michael</td>
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<td>R. Feder, Th. Scheunemann, E. Tamura (Tsukuba)</td>
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