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

Jürgen Henk
Theory Department
Max Planck Institute of Microstructure Physics
Halle (Saale), Germany

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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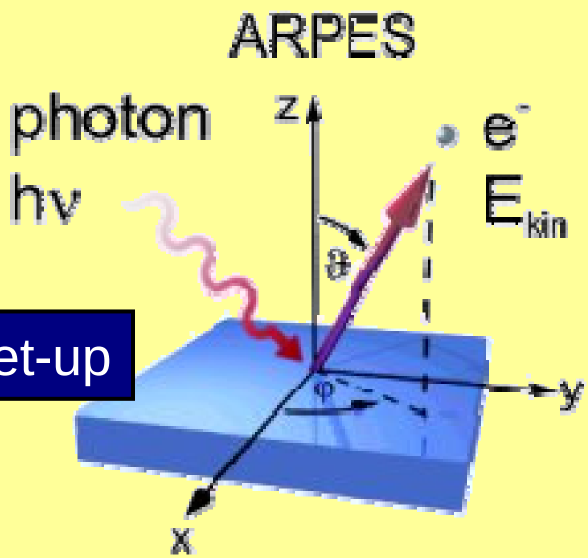
Photoelectron spectroscopy

Spin- and angle-resolved photoelectron spectroscopy (SPARPES)

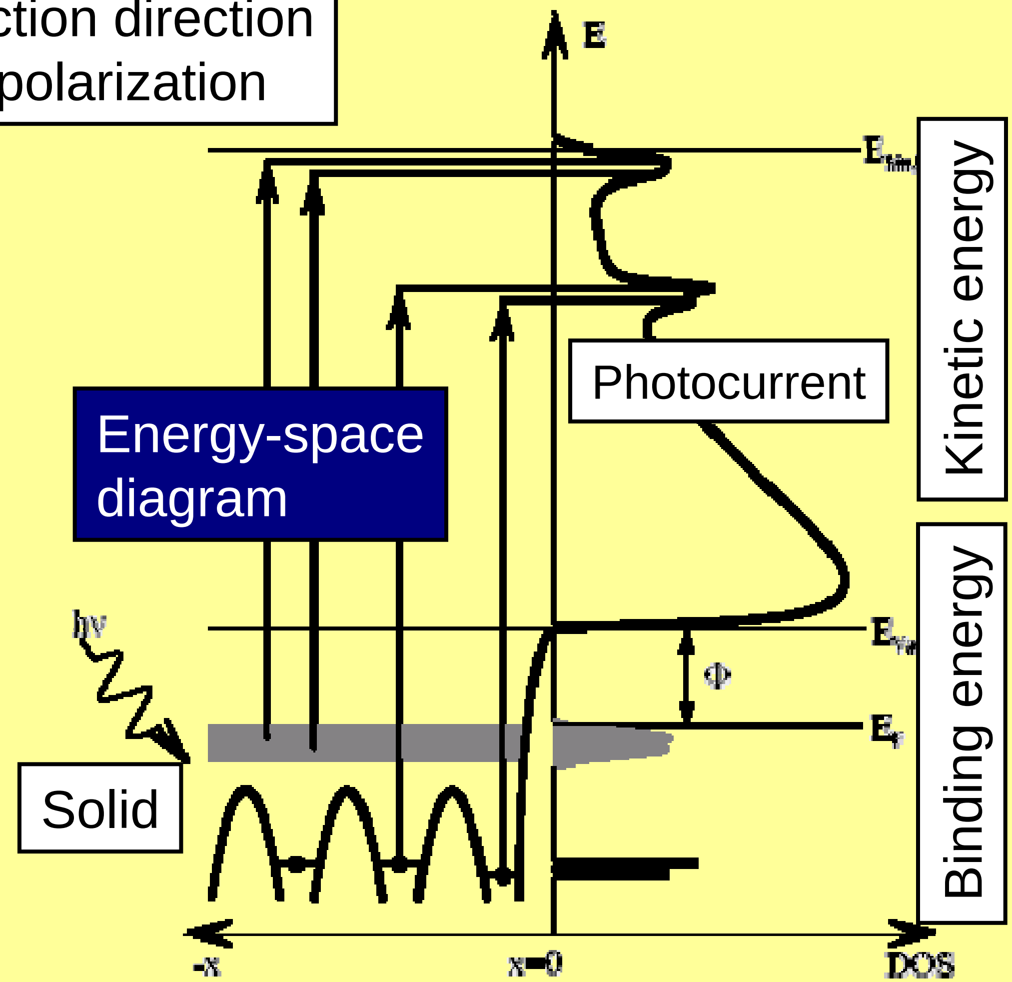
Incident photon
- Energy
- Polarization
- Incidence direction

Photoelectron
- Energy
- Detection direction
- Spin polarization



Set-up

Solid
- Geometric structure
- Electronic structure
- Magnetic structure



One-step model for SPARPES

Excitation and transport = coherent process

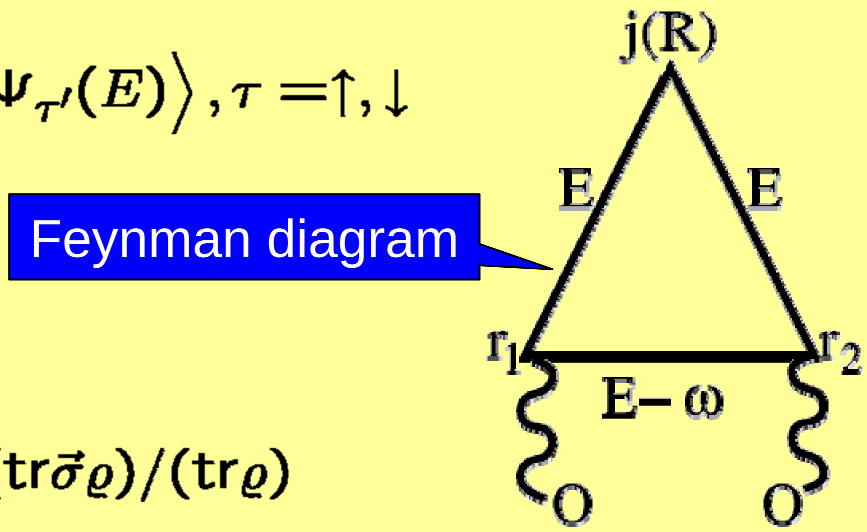
Relativistic description (Dirac equation)

Orbital and spin degrees of freedom treated on equal footing

Spin density matrix

$$\varrho_{\tau\tau'} \propto -\mathrm{Im} \left\langle \Psi_\tau(E) | \Delta G^+(E-\omega) \Delta^\dagger | \Psi_{\tau'}(E) \right\rangle, \tau = \uparrow, \downarrow$$

Ψ_τ time-reversed (SP)LEED state



Photocurrent $j = \mathrm{tr}\varrho$

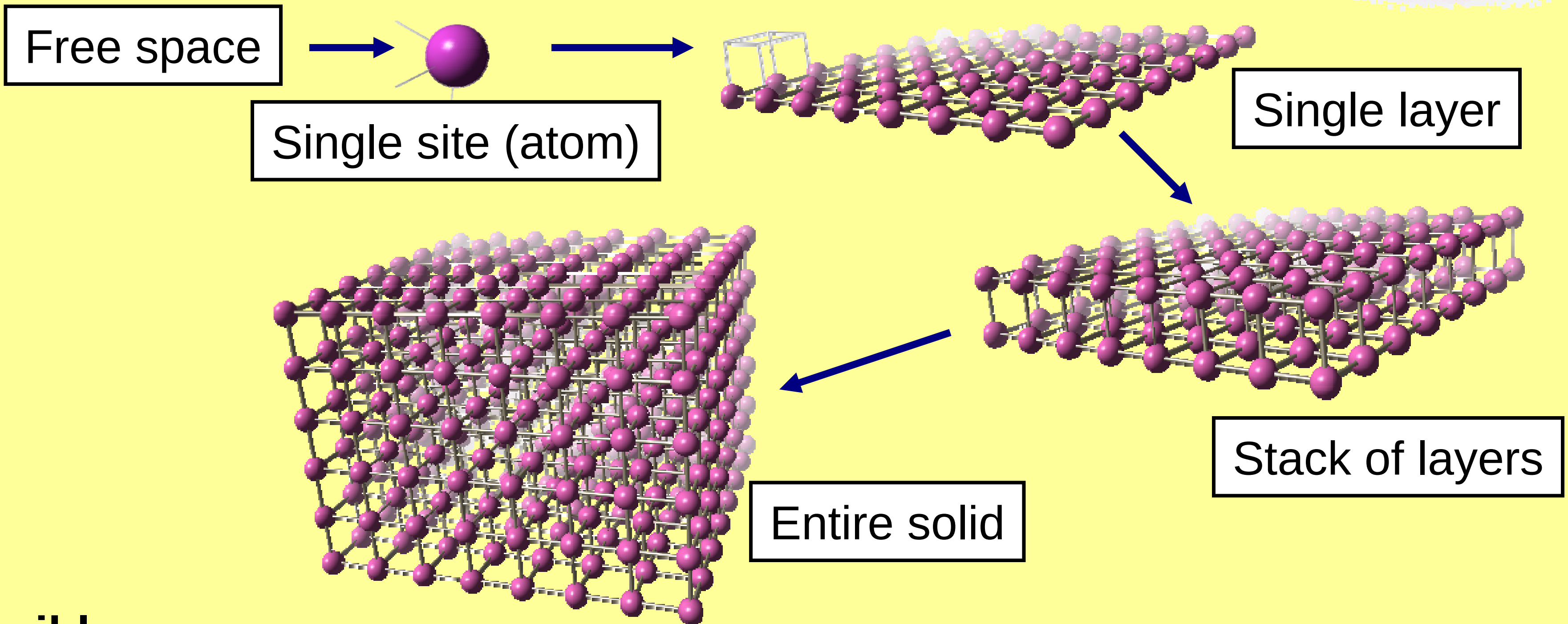
Photoelectron spin polarization $\vec{P} = (\mathrm{tr}\vec{\sigma}\varrho)/(\mathrm{tr}\varrho)$

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)

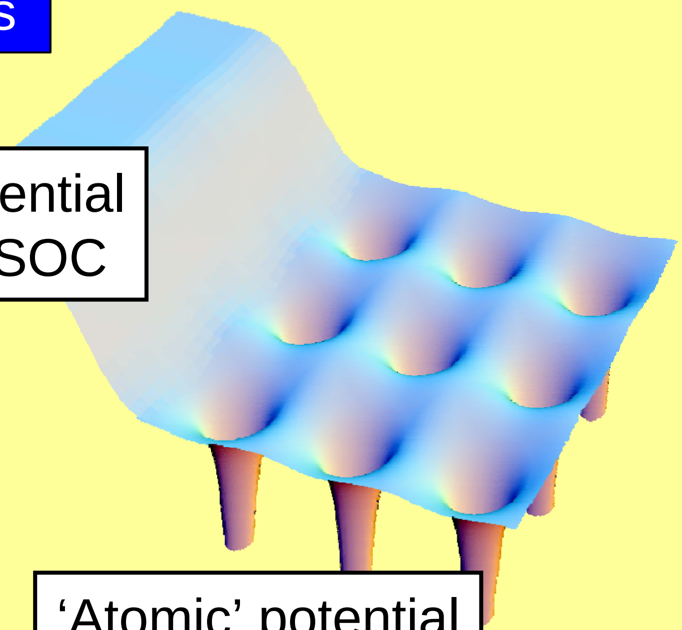
4-spinor

$$\left[c\vec{\alpha}\cdot\vec{p} + \beta m c^2 + V_{\text{eff}} - \epsilon_i\right] \mid \Phi_i\rangle = 0$$

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

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

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



Spin-orbit coupling (SOC)

Pauli form (non-relativistic)

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

Korringa-Kohn-Rostoker method

Green function

Scattering solution (single site)

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

Single-site contribution

Multiple-scattering contribution
Scattering-path operator

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

Spin-angular quantum number

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)
   - $\mapsto$ 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)

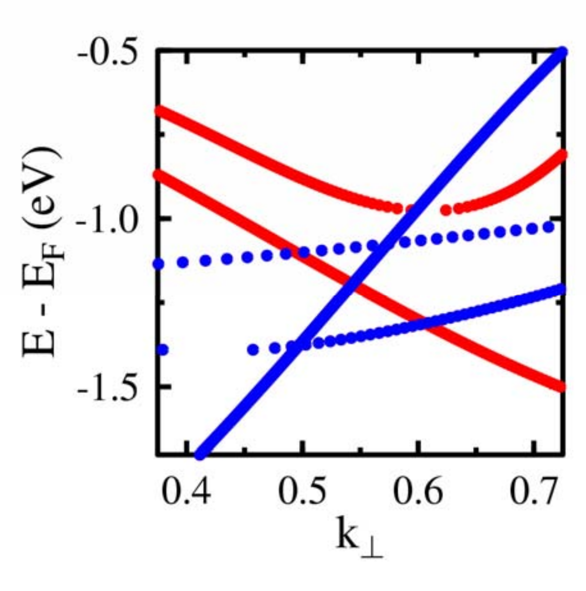
'Free' parameters

- 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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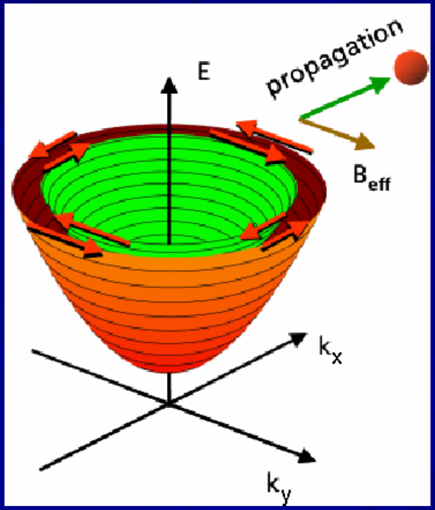

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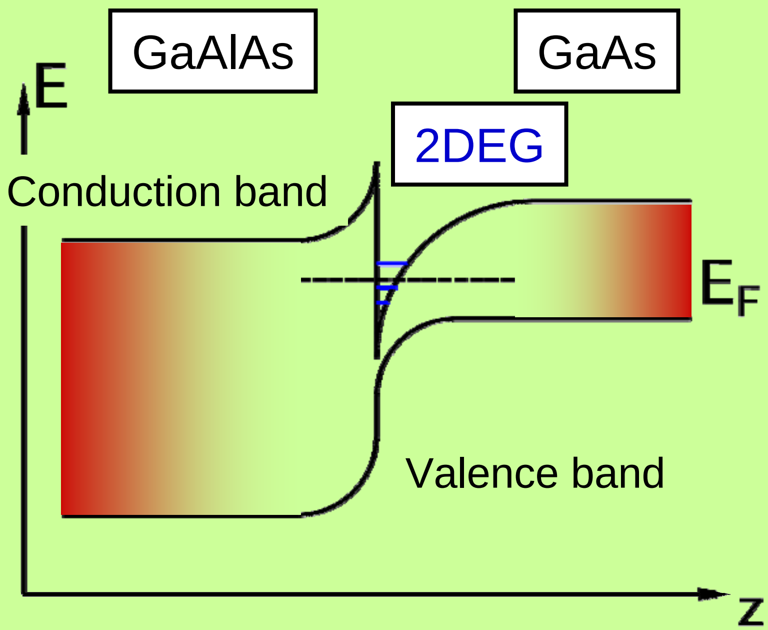
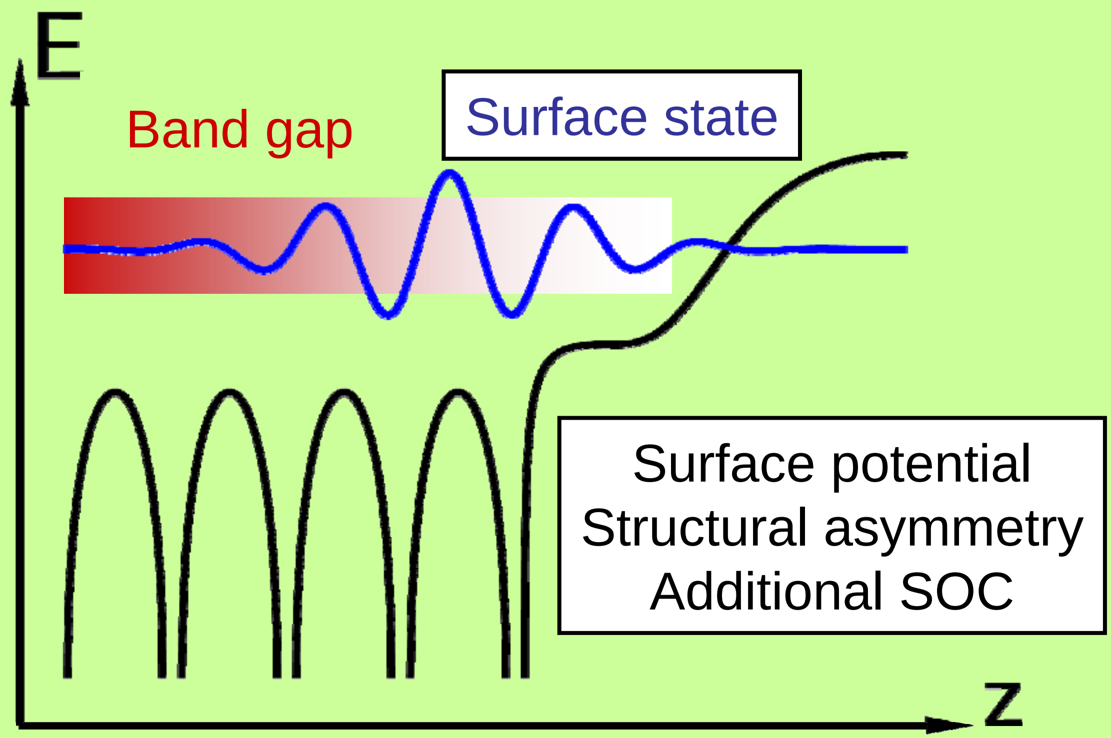
Rashba-Bychkov effect

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

Interface in a semiconductor heterostructure



Metal surface



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

$$H_{SOC} = \frac{e\hbar}{mc}\vec{s}\cdot\vec{B}$$

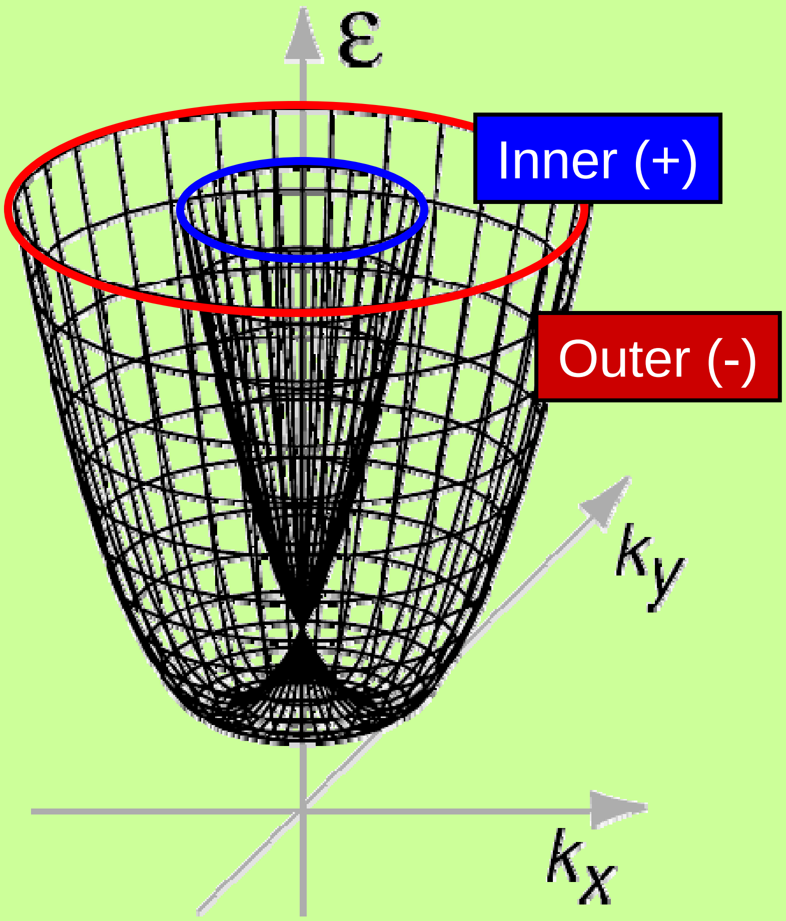
$$H_{SOC} = \frac{\hbar^2}{2m^2c^2}\vec{s}\cdot\left[\vec{\nabla}V(\vec{r})\times\vec{p}\right]$$

$$H_{SOC} = \frac{\hbar^2}{2m^2c^2}\frac{1}{r}\frac{dV}{dr}\vec{s}\cdot\vec{l}$$

Dispersion with SOC

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

Splitting

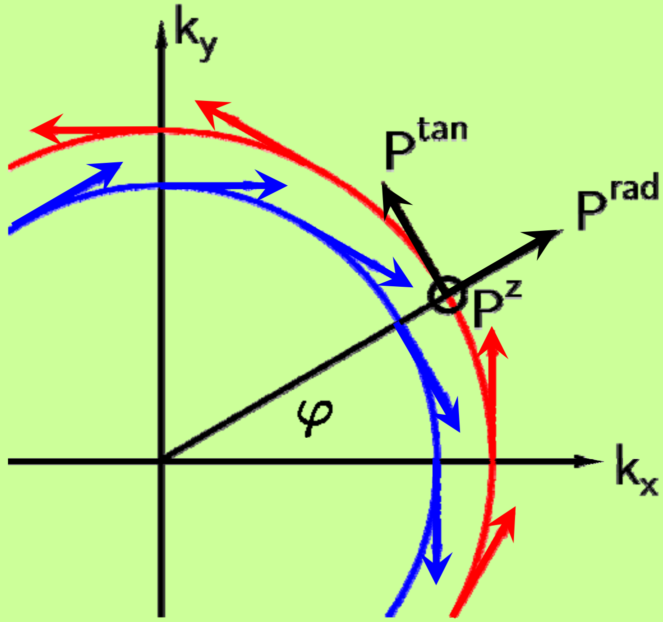


Isotropic 2DEG versus Au(111) surface state

Anisotropic spin polarization

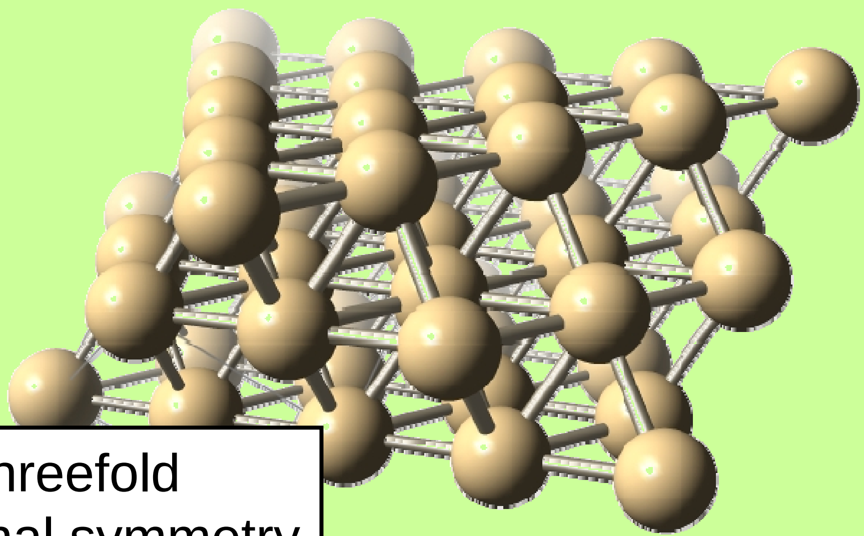
2DEG
$$\begin{cases} P_{\pm}^{\tan} &= \pm 1 \\ P_{\pm}^{\text{rad}} &= 0 \\ P_{\pm}^{z} &= 0 \end{cases}$$

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



Au(111)
$$\begin{cases} P_{\pm}^{\tan} &= \alpha_{\pm} \\ P_{\pm}^{\text{rad}} &= \beta_{\pm} \cos 3\varphi \\ P_{\pm}^{z} &= \gamma_{\pm} \cos 3\varphi \end{cases}$$

Value and sign of *α*, *β* und *γ*?



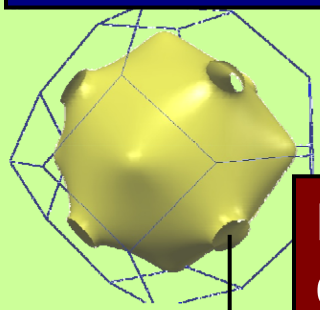
Threefold rotational symmetry

Au(111) surface state

Dispersion and momentum distribution

J. Osterwalder *et al.* (Zürich)

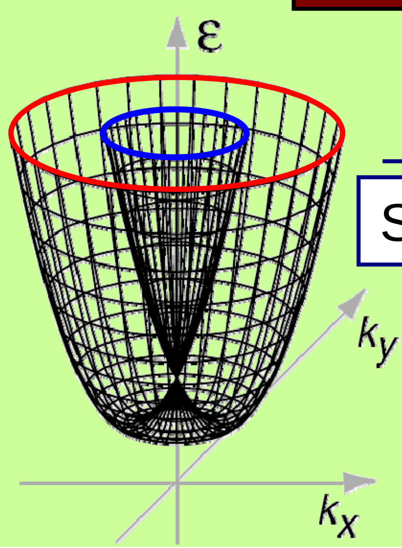
Fermi surface



L point

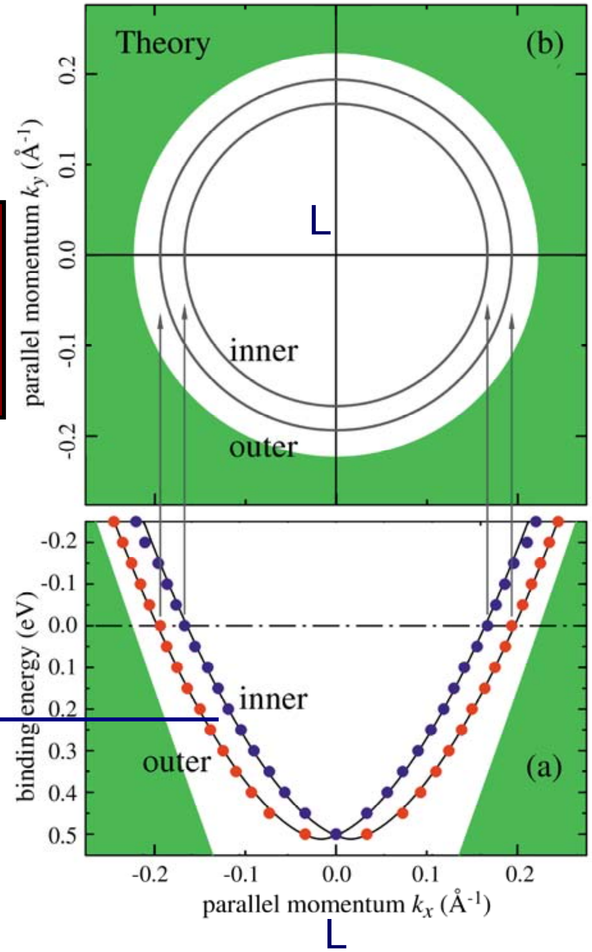
Momentum distribution at E_F

Dispersion

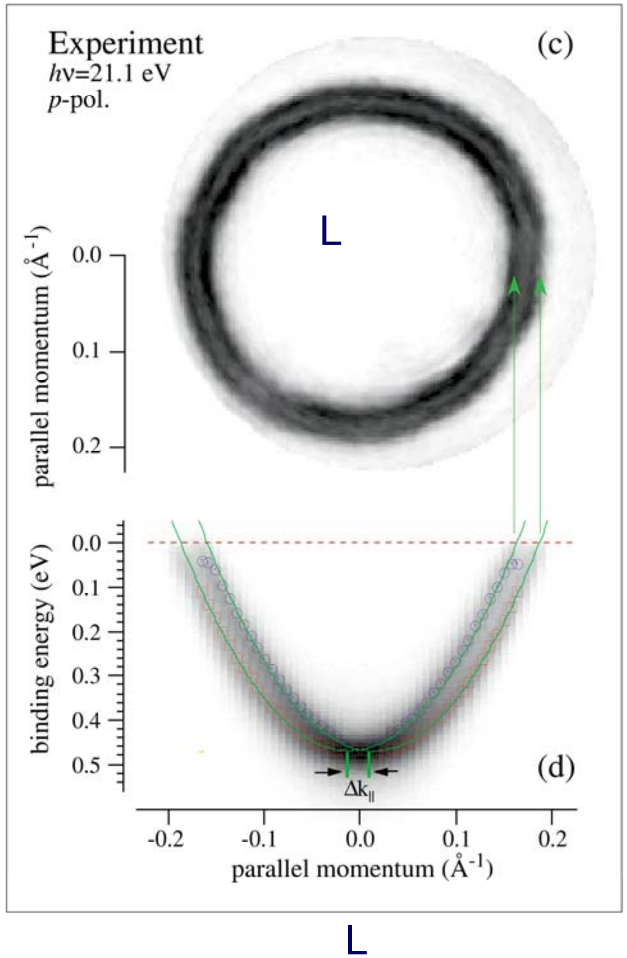


Splitting

Ab initio theory



Experiment

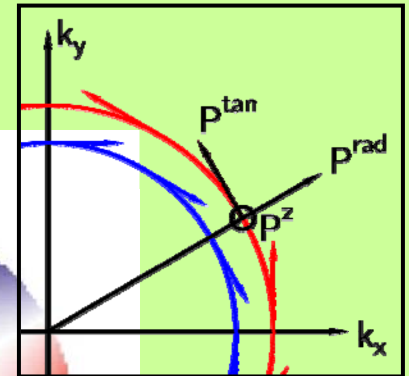


- Parabolic dispersion
- Splitting by SOC

Au(111) surface state

Spin polarization of photoelectrons

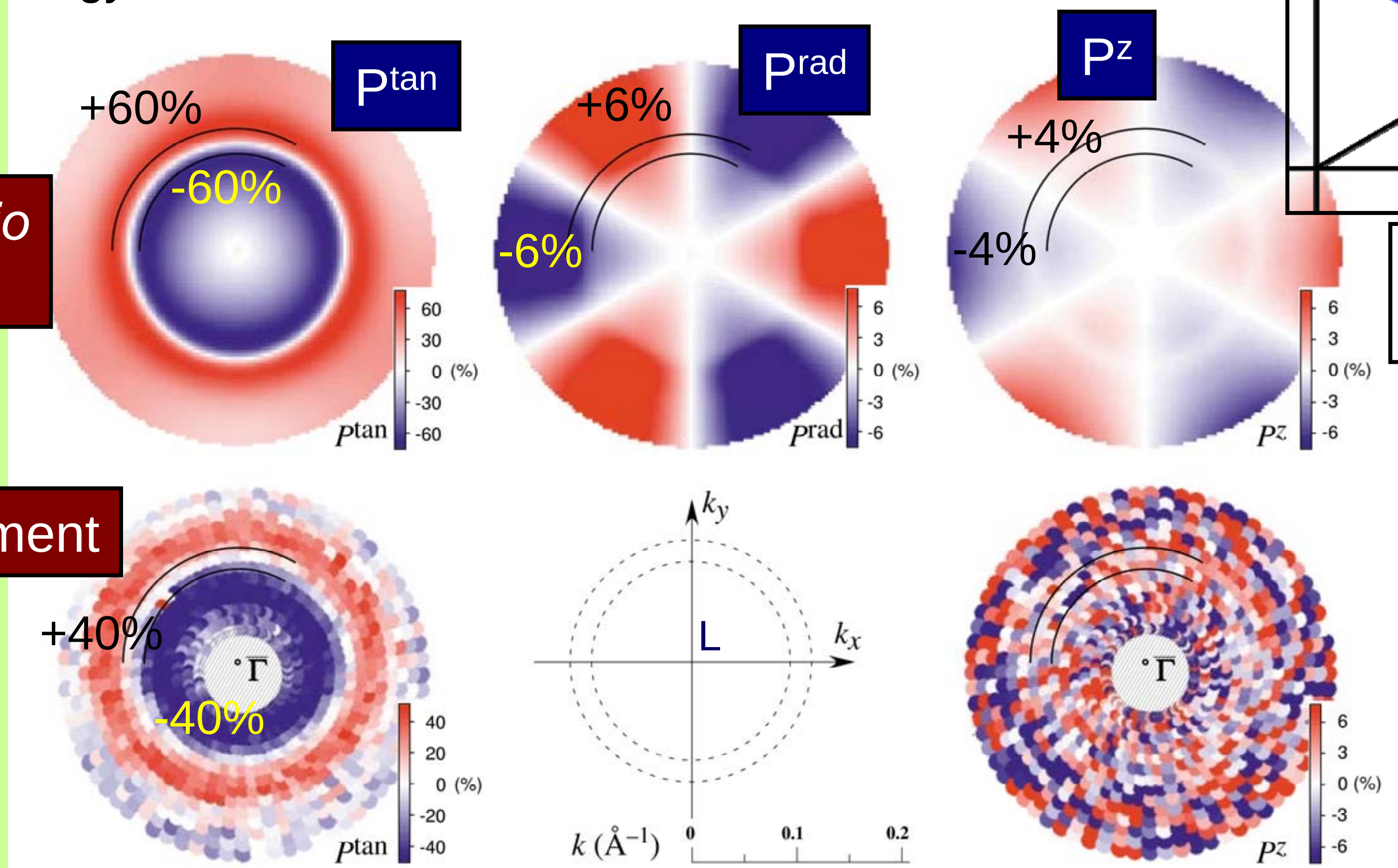
Binding energy 0.17 eV



Threefold symmetry

Ab initio theory

Experiment



- Sign
- Order of magnitude
- Threefold symmetry

$$P^{tan} = \alpha_\pm$$
$$P^{rad} = \beta_\pm \cos 3\varphi$$
$$P^{z} = \gamma_\pm \cos 3\varphi$$

Au(111) surface state

Summary

Agreement of dispersion and splitting

Spin polarization

		P^{tan}	P^{rad}	P^{z}
Ground state	Model calculation	1	0	0
	Ab initio calculation	0.97	0.01	0.014
Photoelectron	PE calculation	0.6	0.06	0.04
	PE experiment	0.4	—	< 0.05

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

JH, A. Ernst, P. Bruno, Phys. Rev. B **68** (2003) 165416; Surf. Sci. **566-568** (2004) 482

JH, M. Hoesch, J. Osterwalder, A. Ernst, P. Bruno, J. Phys. CM **16** (2004) 7581

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Magnetic linear dichroism

Magnetic dichroism

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

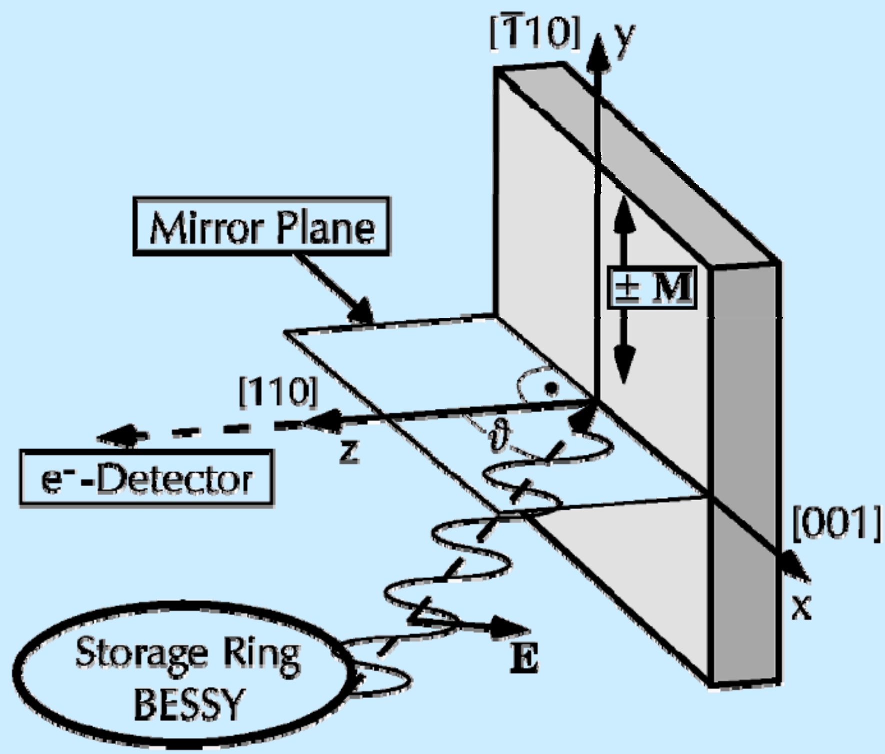
Magnetic *linear* dichroism

Normal emission

p-polarized light

Magnetization in a mirror plane

$\mapsto$ reduction of symmetry (*2mm* → *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')

$$\begin{aligned} j(\pm\vec{M}) &= \sin^2\vartheta \left(|M^{1++}|^2 + |M^{1--}|^2\right) \\ &+ \cos^2\vartheta \left(|M^{3++}|^2 + |M^{3--}|^2\right) \\ &\pm \frac{\sin 2\vartheta}{2} \Im \left(M^{1++*} M^{3++} - M^{1--*} M^{3--}\right) \end{aligned}$$

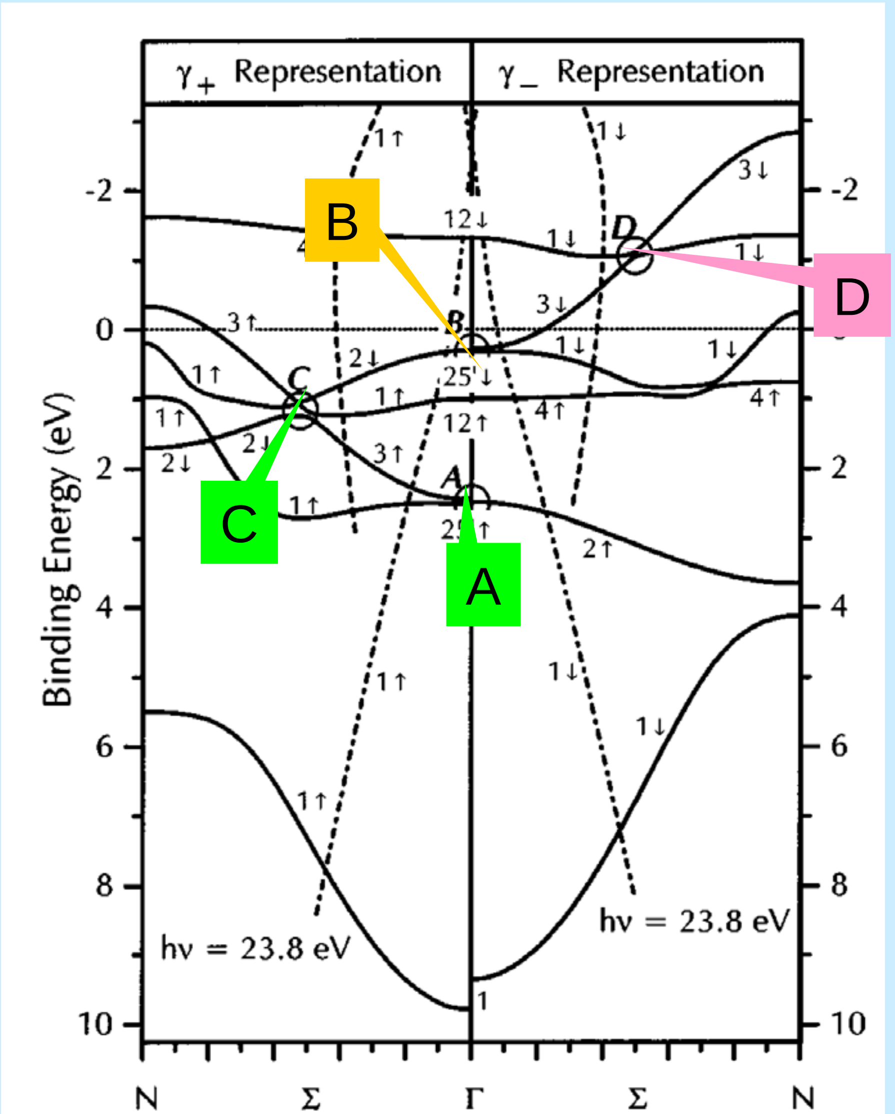
Hybridization by SOC

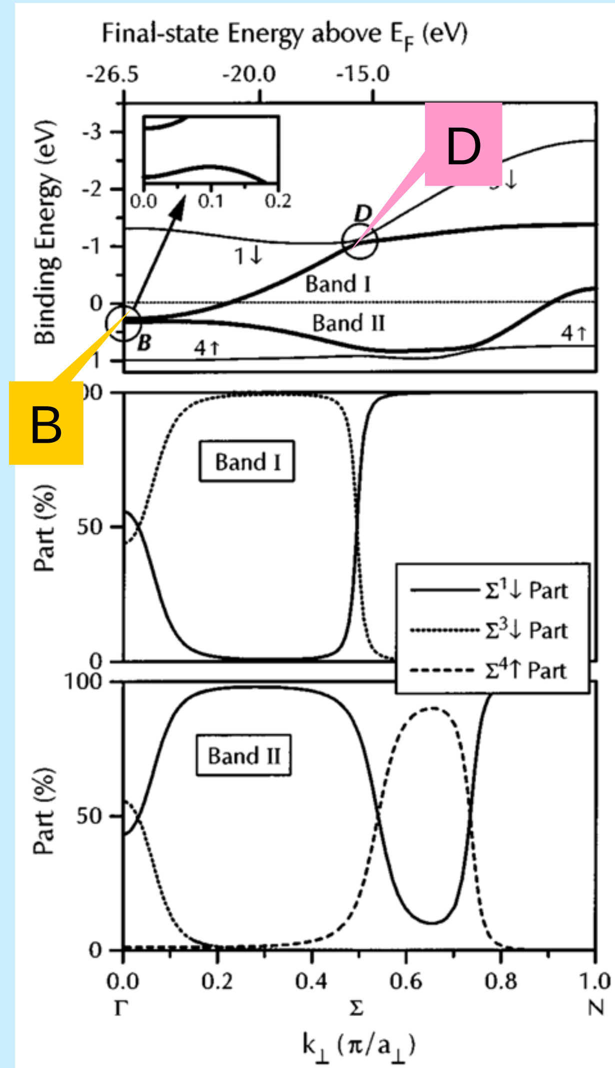
MD does *not* probe areas of large magnetism but of large hybridization

A. Rampe, G. Güntherodt, D. Hartmann, JH, T. Scheunemann, R. Feder, Phys. Rev. B **57** (1998) 14370

Fe(110) – hybridization

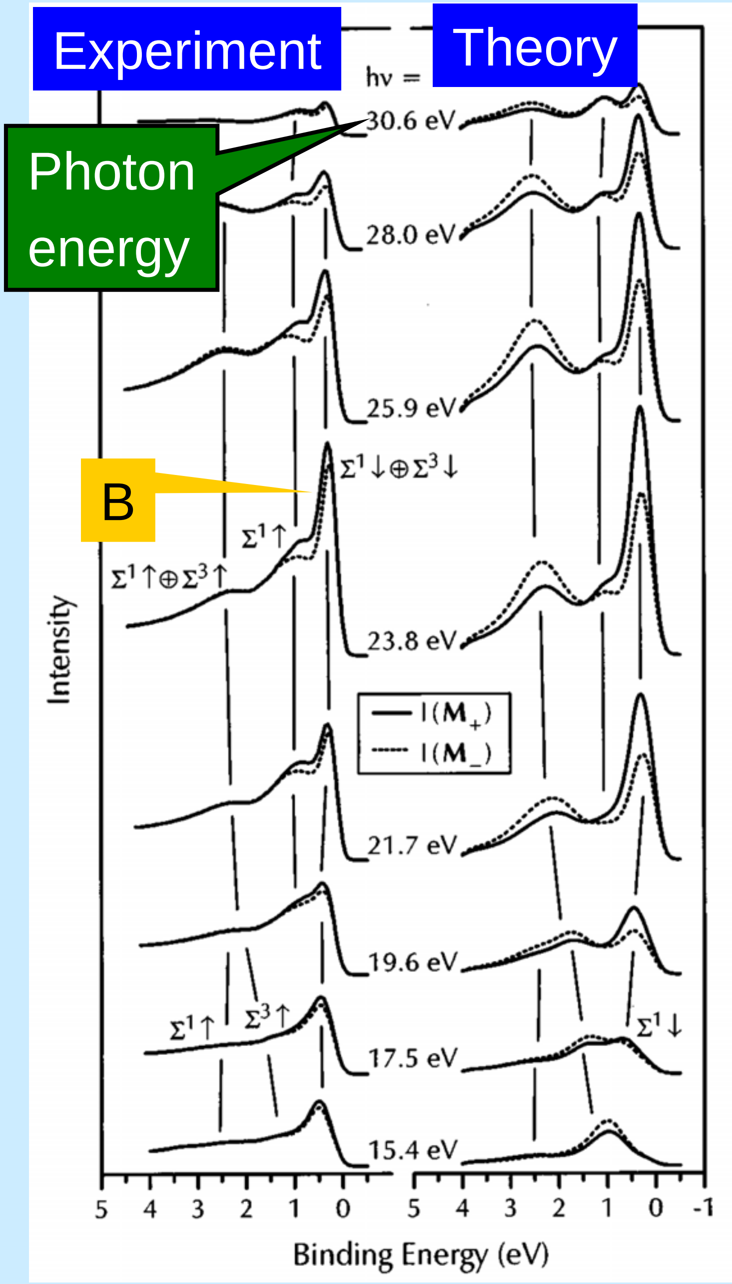
Focus on $\Sigma^{1\uparrow} \oplus \Sigma^{3\uparrow}$ and $\Sigma^{1\downarrow} \oplus \Sigma^{3\downarrow}$



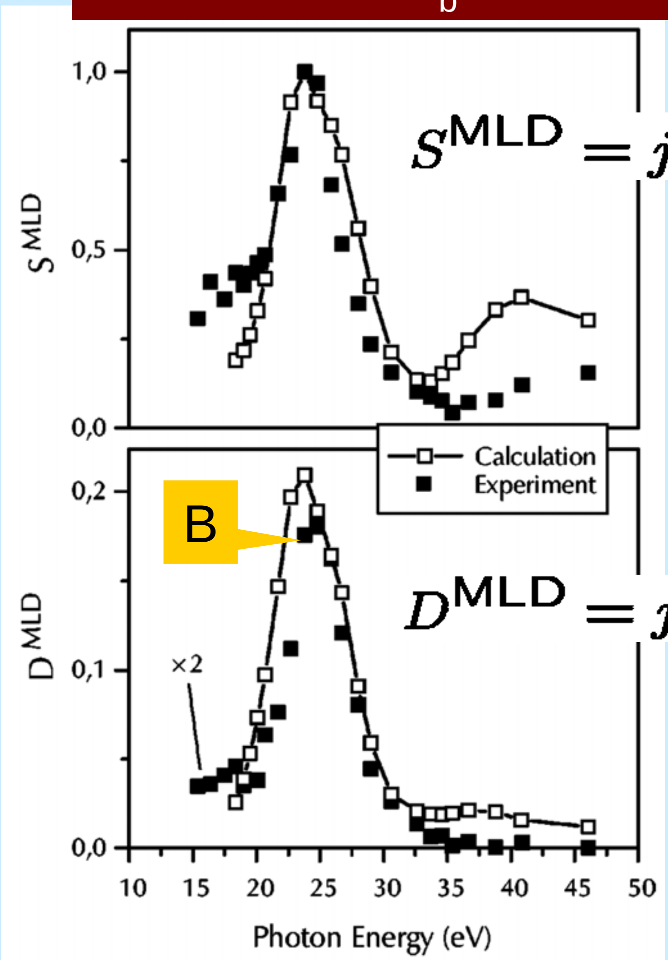


Change of orbital character by SOC

Fe(110) – Magnetic linear dichroism



Dichroism at E_b = 0.5 eV



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

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

MD allows to identify areas of hybridization induced by SOC

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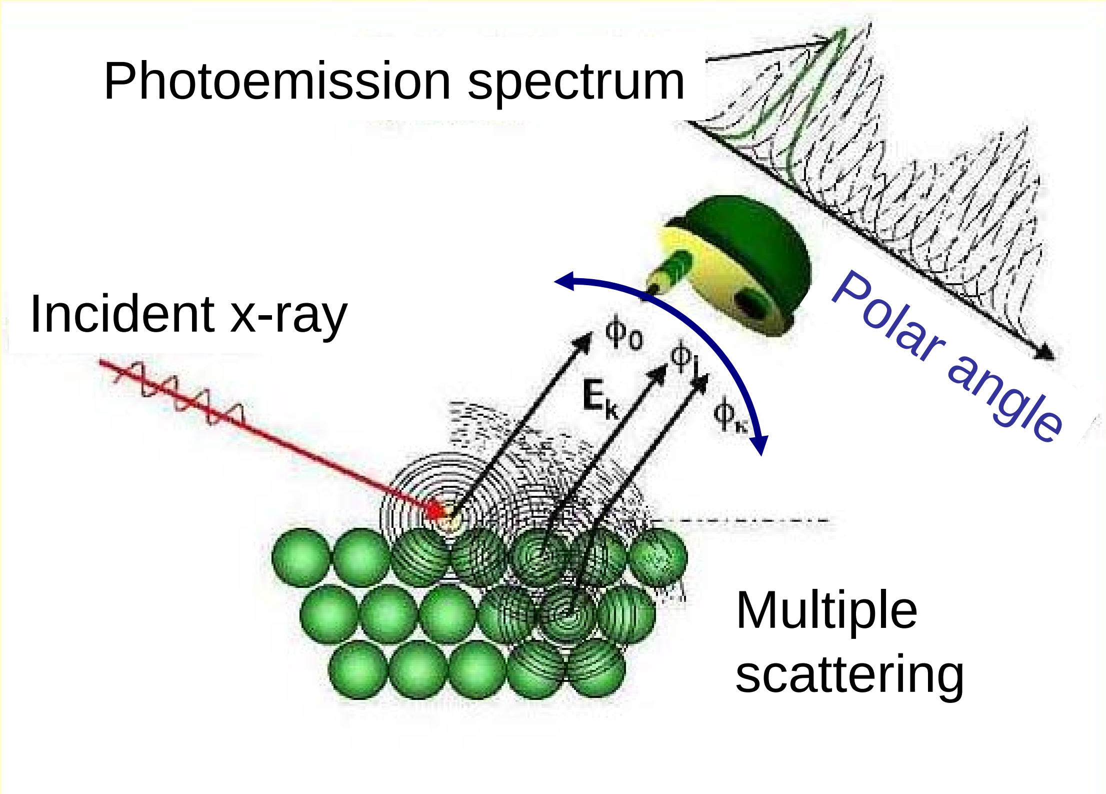
Photoelectron diffraction



Emission from core levels
$\mapsto$ element-specific

Constant kinetic energy
Polar-angle scans

Information on
- Surface geometry
- Surface magnetism

Spin-dependent scattering in the final state (photoelectron)

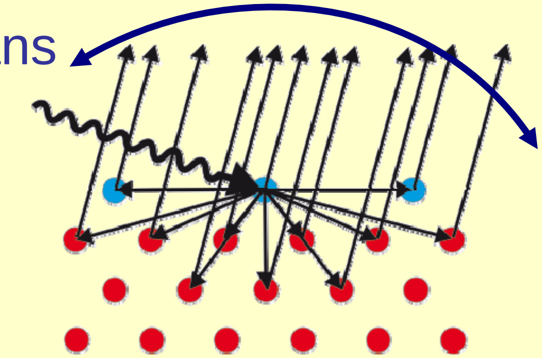
Example: Dichroic PED from 3*p* levels in Fe(001)

JH, A.M.N. Niklasson, B. Johansson, Phys. Rev. B **59** (1999) 13986

Refining the theory

Emission from Fe(001)-3*p*

Polar-angle scans



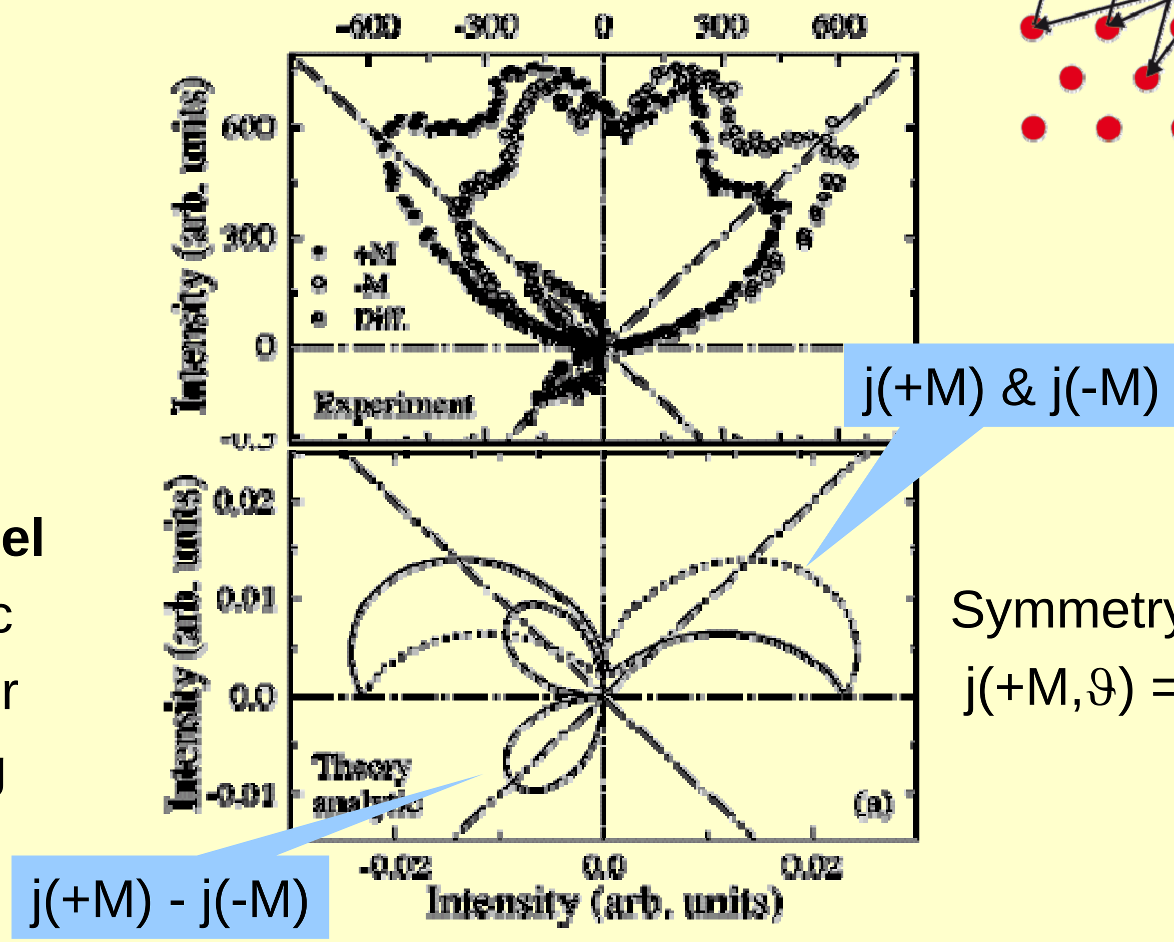
Experiment

Atomic model

Semi-analytic

Single emitter

No scattering



Symmetry

$j(+M,\vartheta) = j(-M,-\vartheta)$

Refining the theory

Experiment

Solid

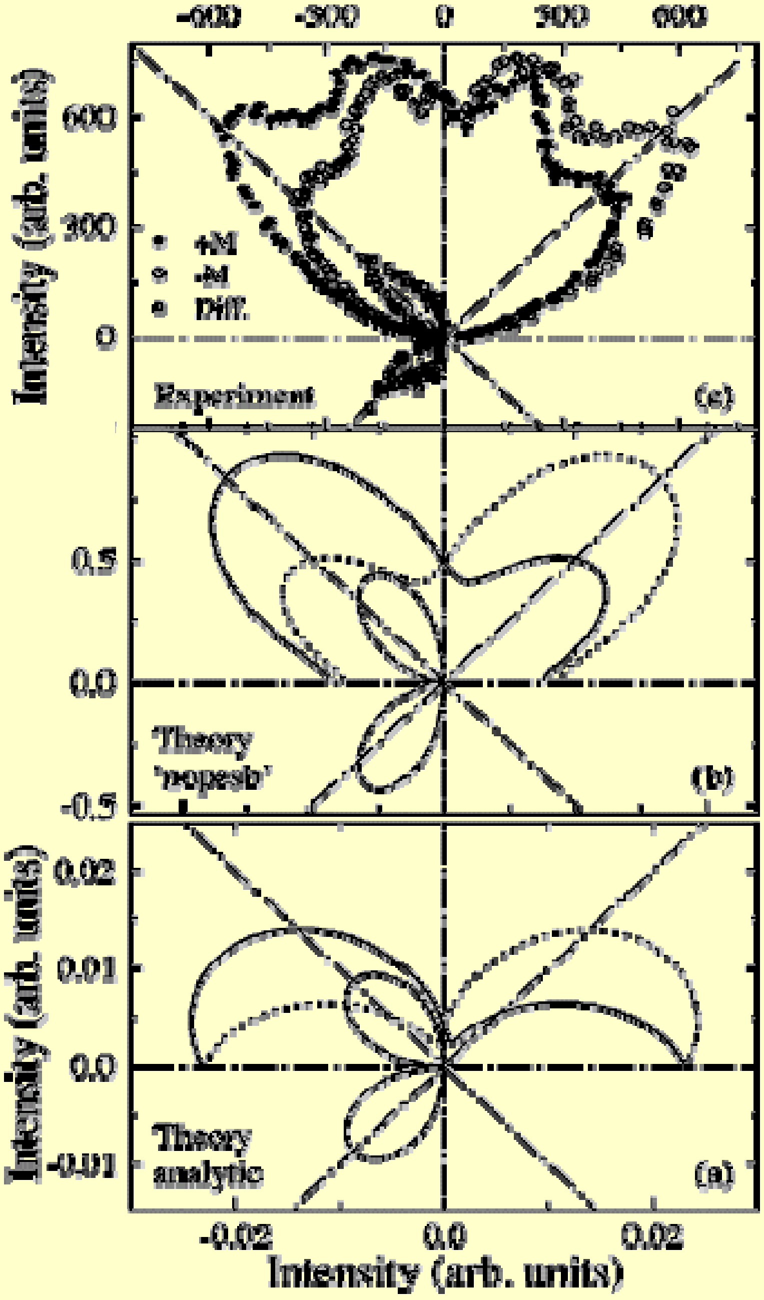
Numerical

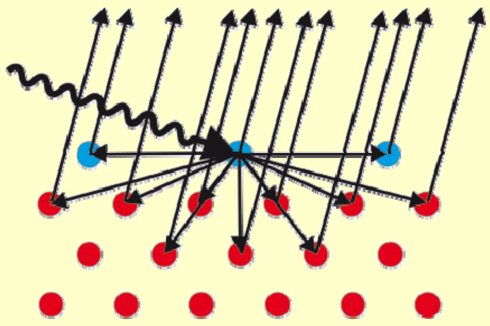
All Fe sites emit

No scattering

No surface barrier

Atomic model

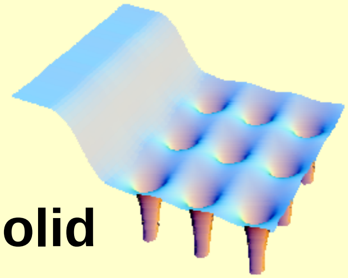




Refining the theory

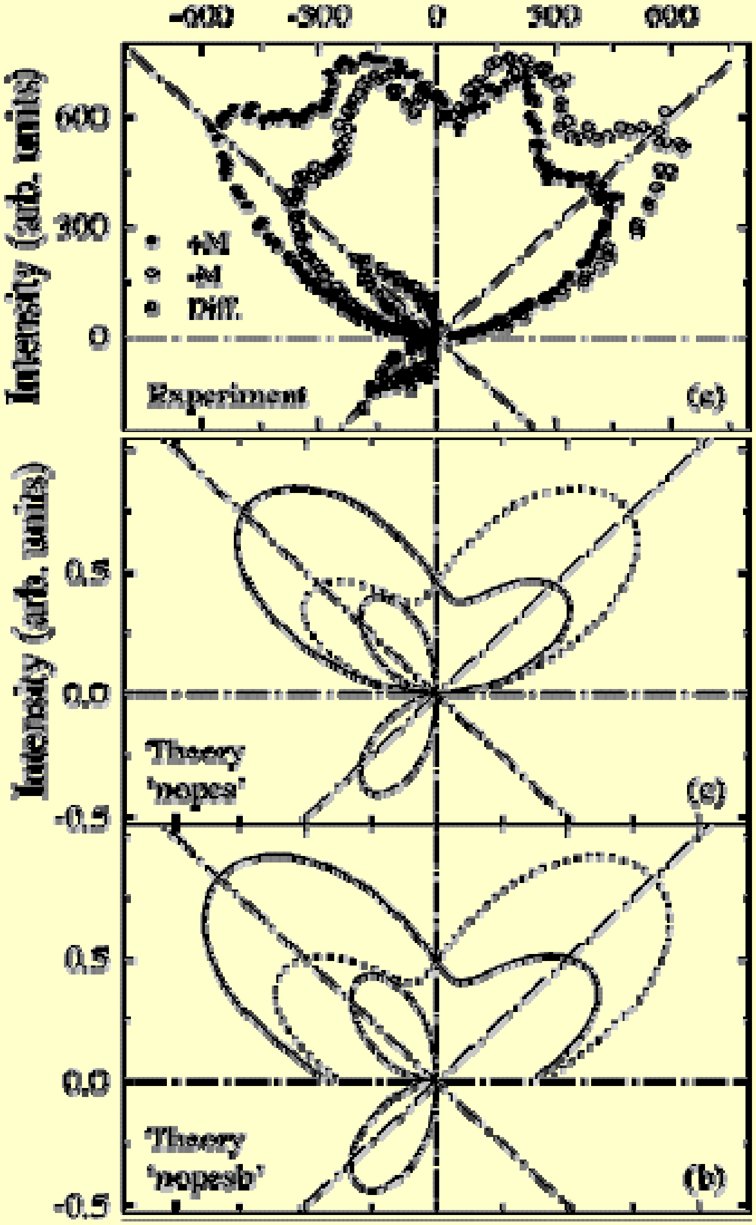
Experiment

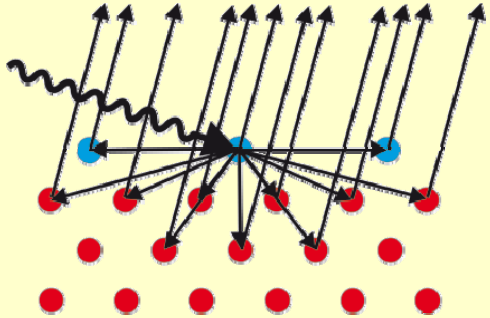
Solid



Surface barrier

Solid



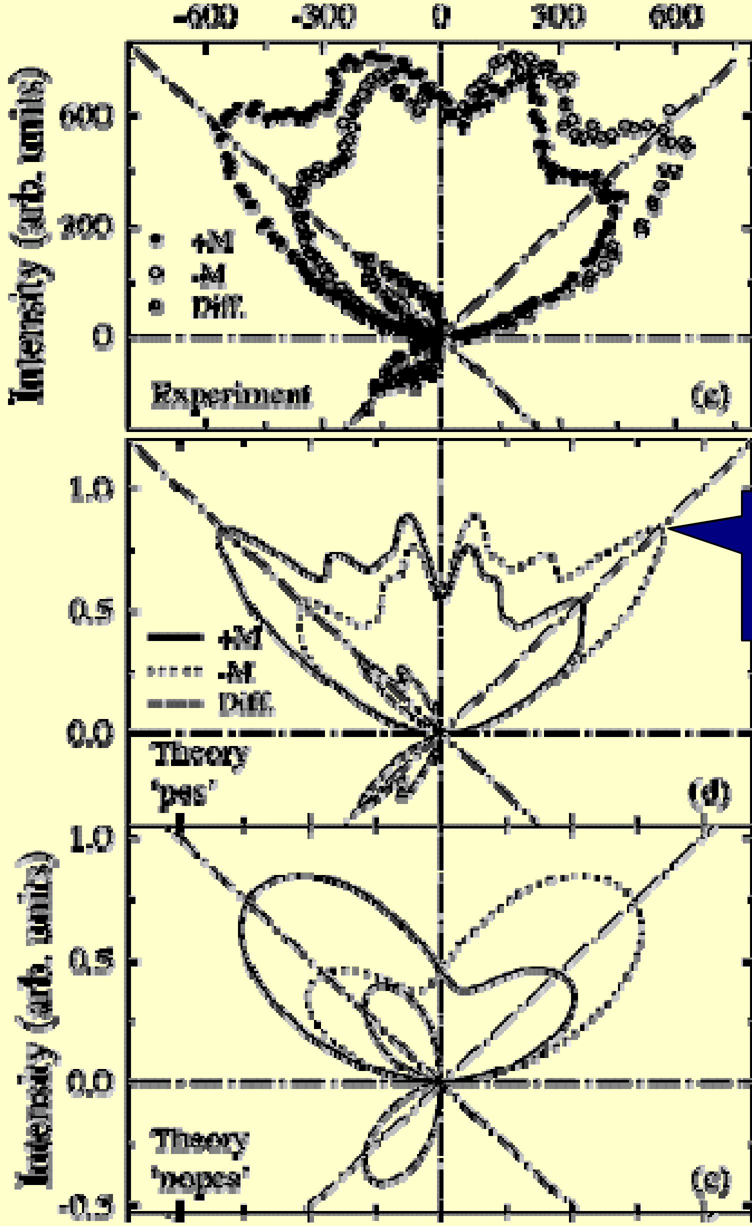


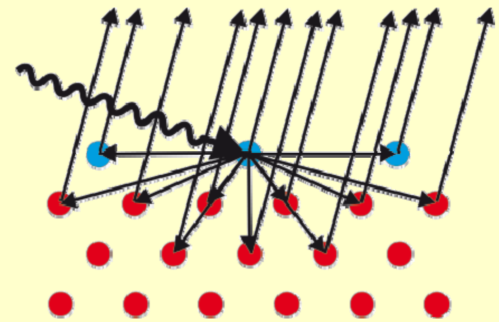
Refining the theory

Experiment

Solid
Multiple scattering

Solid





Increased forward scattering at 45°

Strong spin-dependent multiple scattering in the final state

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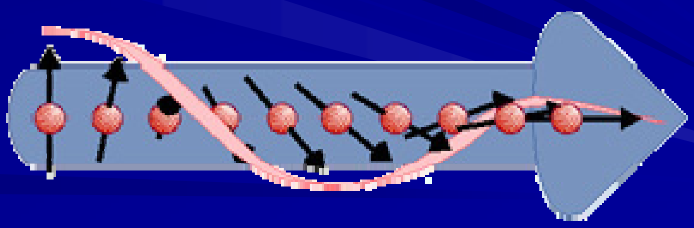
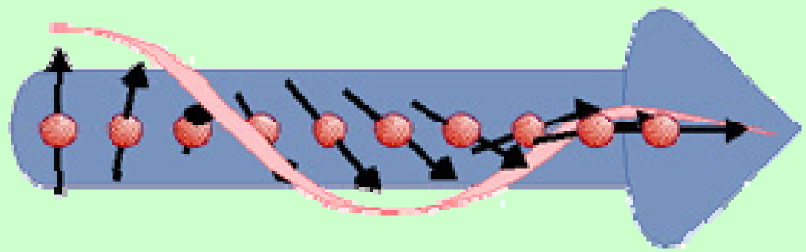


Spin motion

Motion of electrons in a magnetic system

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

Relaxation: Inelastic processes

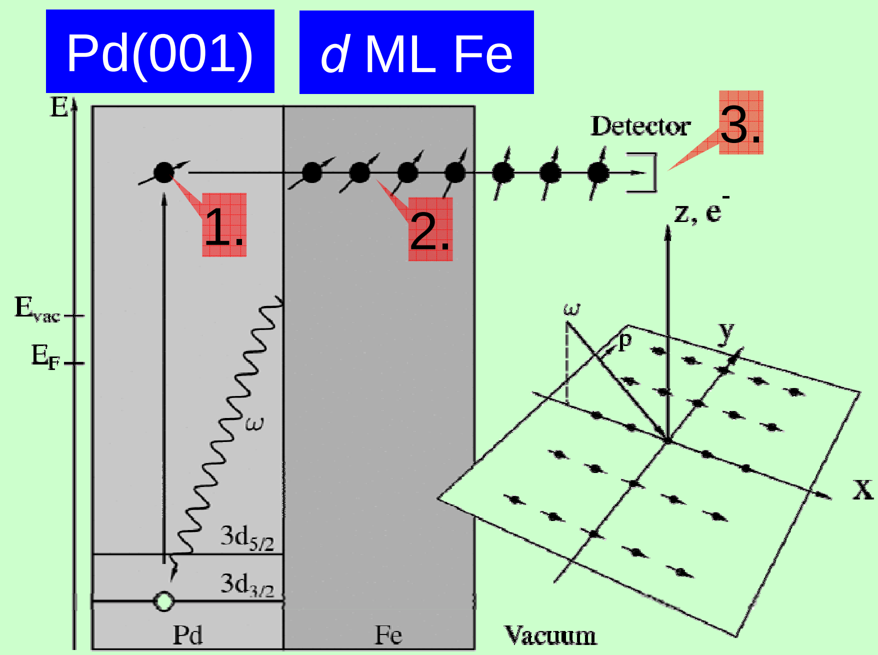


Experiments:

- Transmission through a free-standing magnetic film
- SPLEED

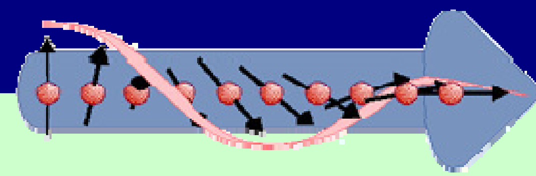
New approach: Spin-resolved ARPES

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

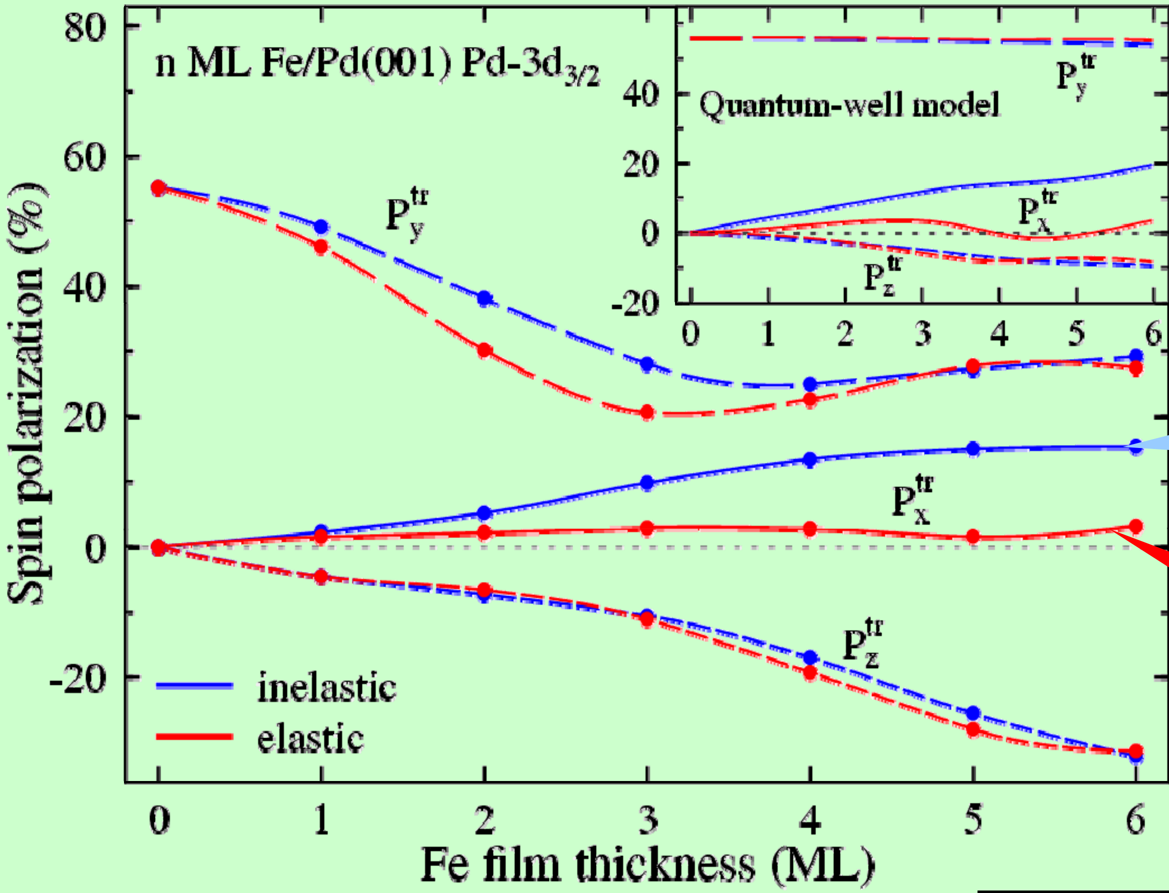


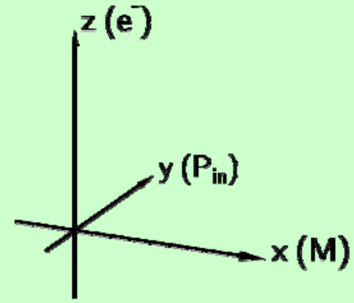
Spin motion = Spin polarization in dependence of film thickness *d*

Spin motion of photoelectrons



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





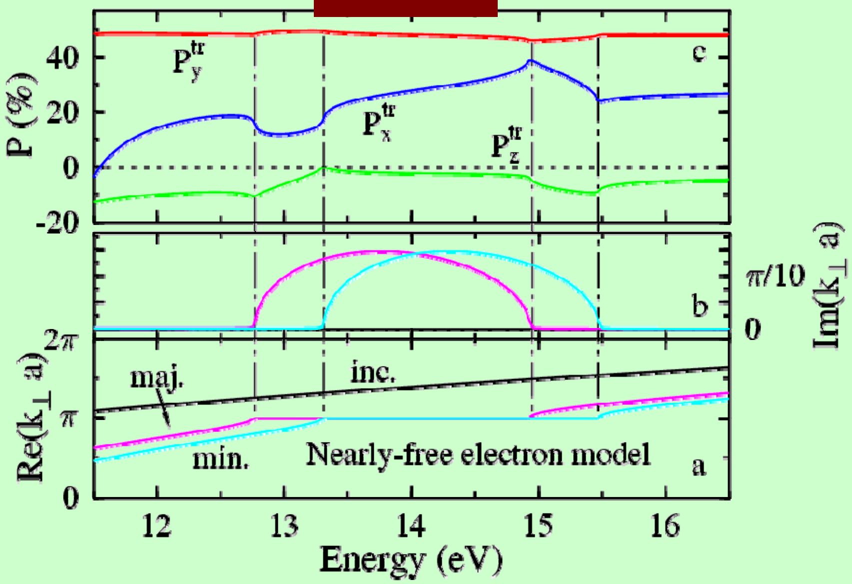
Deviations from model calculations

Effect of the Fe electronic structure

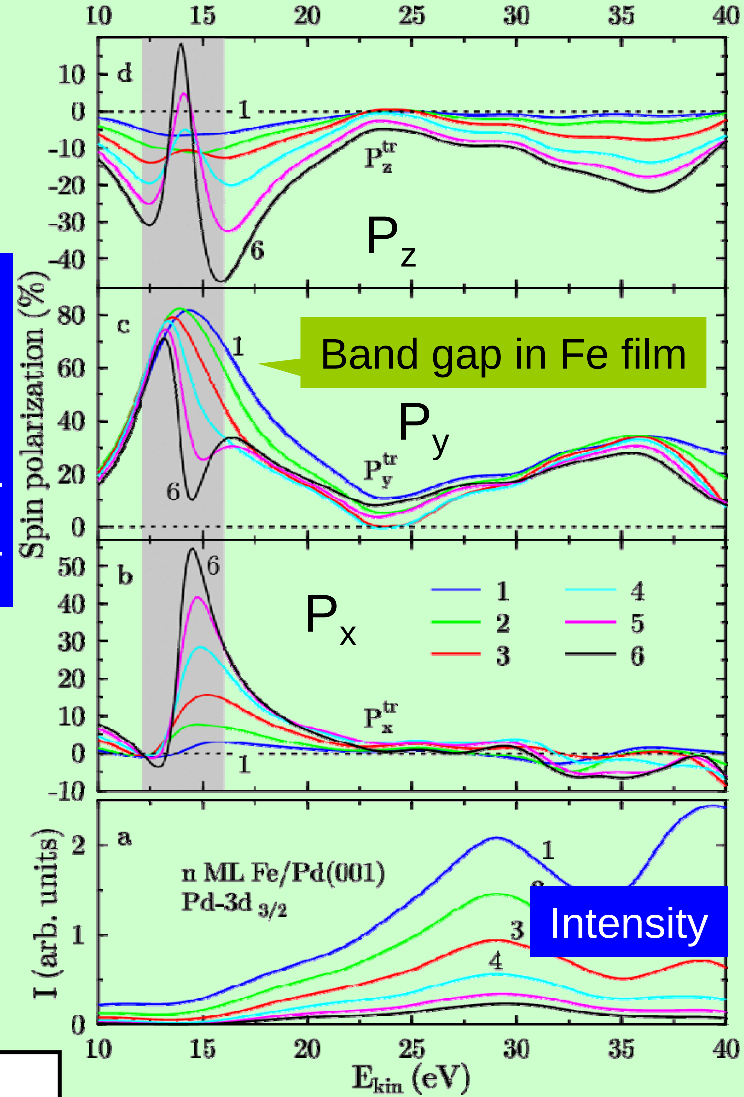
JH, P. Bose, Th. Michael, P. Bruno, Phys. Rev. B **68** (2003) 052403.

Spin motion of photoelectrons

Model



Ab-initio photoemission



Band gaps: rapid change of the spin polarization

Evolution of a band gap with film thickness

Spin motion provides information on unoccupied electronic structure in magnetic films

Summary

Spin-orbit effects

Au(111)

- Intrinsic SOC effect: Rashba effect in the L-gap surface state
- Extrinsic SOC effect: photoelectron spin polarization due to SOC

Fe(110)

- Intrinsic SOC effect: Band gaps and hybridization
- Extrinsic SOC effect: Magnetic dichroism (transition matrix elements)

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

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