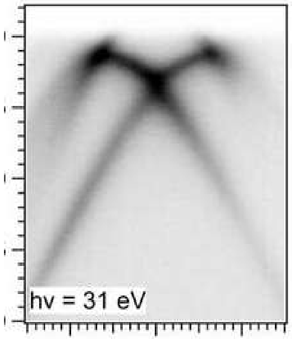
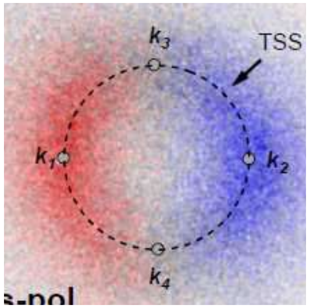
Unscrambling orbital character and spin: The importance of the final state

CORPES17

Hiroshima



6. July 2017



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- **Hendrik Bentmann**
  with **Henriette Maaß, Christoph Seibel, Thiago Peixoto**

Photoemission theory:
- **E. Krasovskii (San Sebastian)**
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- **Christian Tusche**

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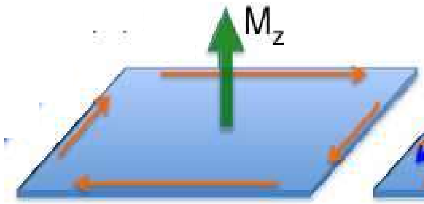
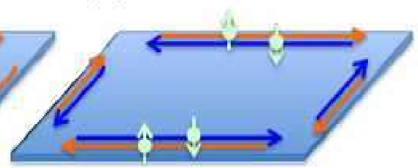
SFB1170

Motivation: Spin-split electronic structure

Quantum Anomalous Hall effect

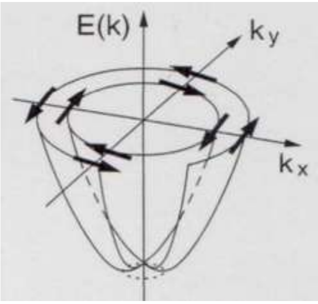


Quantum Spin Hall effect

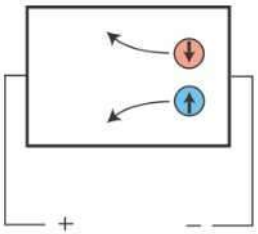


2D

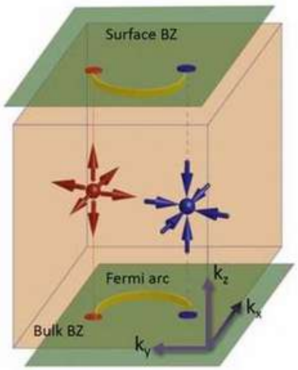
Rashba effect



Spin-Hall effect



Weyl semimetal

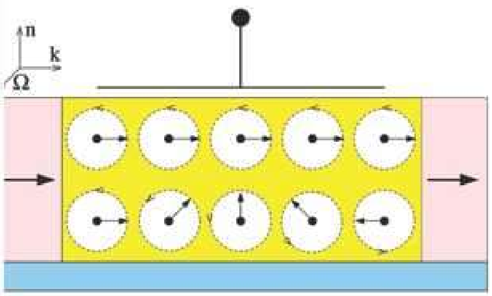


Spin-Orbit Coupling (SOC)

spin-polarization of electronic states

→ origin of non-trivial **topology**

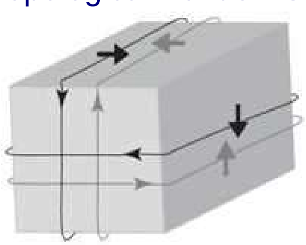
direct spectroscopy by **spin- and angle-resolved photoemission**



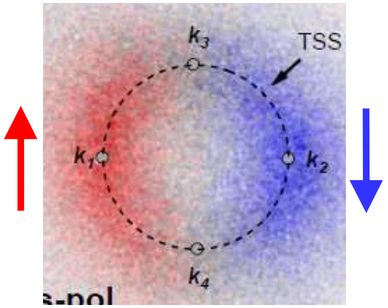
Spin transistor

Topological insulator

Topological Kondo insulator

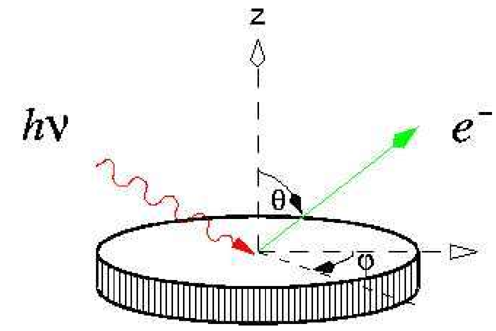


3D

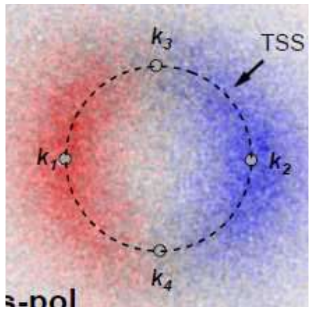


Outline

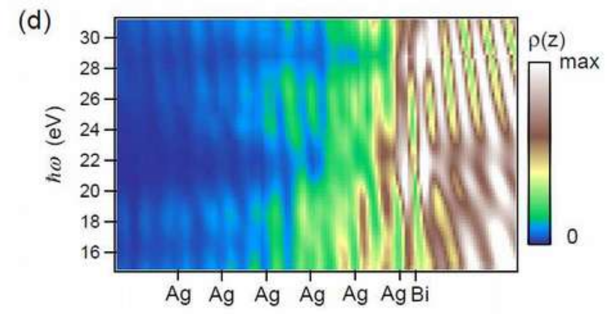
ARPES on systems with strong spin-orbit coupling



Complications by initial state
→ linear dichroism

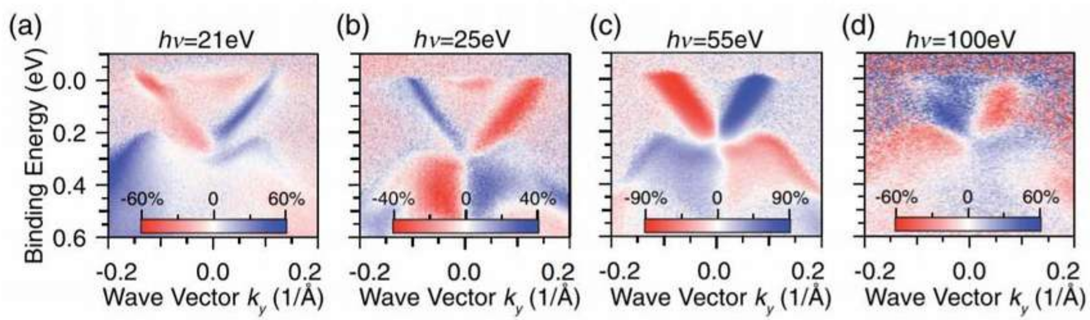


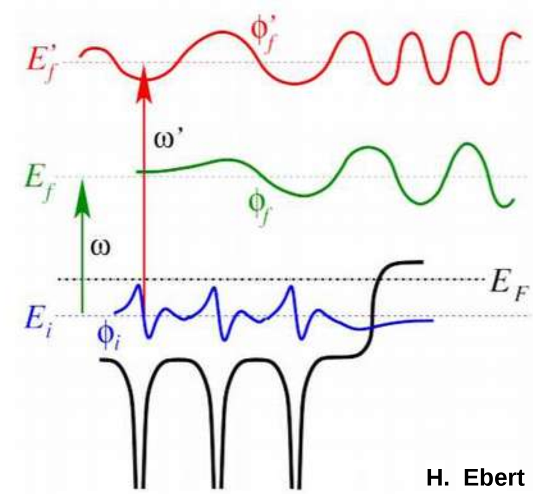
Importance of the final state
→ photoelectron polarization



How to address intrinsic spin properties by photoemission?

Circular dichroism (indirectly)

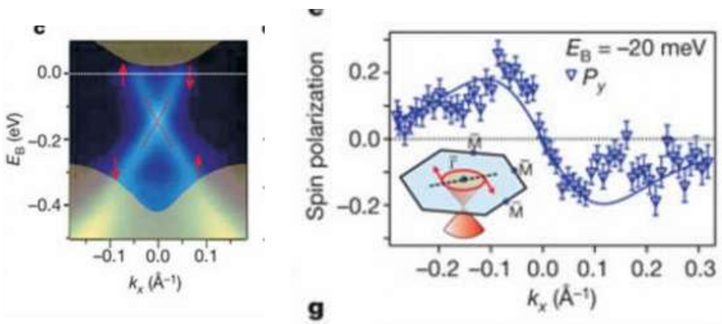


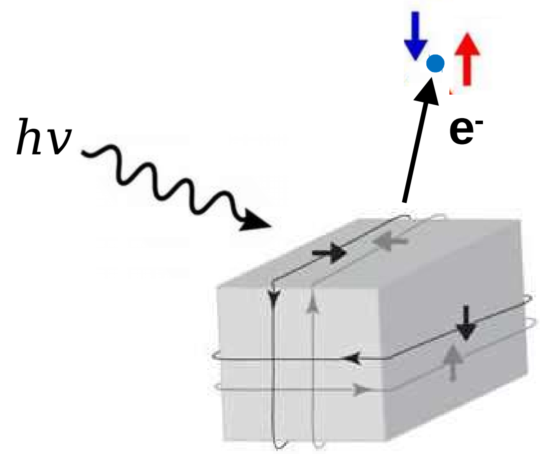


Scholz *et al.*, Phys. Rev. Lett. **110**, 216801 (2013)

→ **„final state effect", dichroism depends on excitation energy**

Photoelectron spin polarization





Hsieh et al., Nature **460**, 1101-1105 (2009)

→ **also dependence on photoemission process?**

IOP PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER
J. Phys.: Condens. Matter **24** (2012) 171001 (3pp) doi:10.1088/0953-8984/24/17/171001

VIEWPOINT

Can spin-polarized photoemission measure spin properties in condensed matter?

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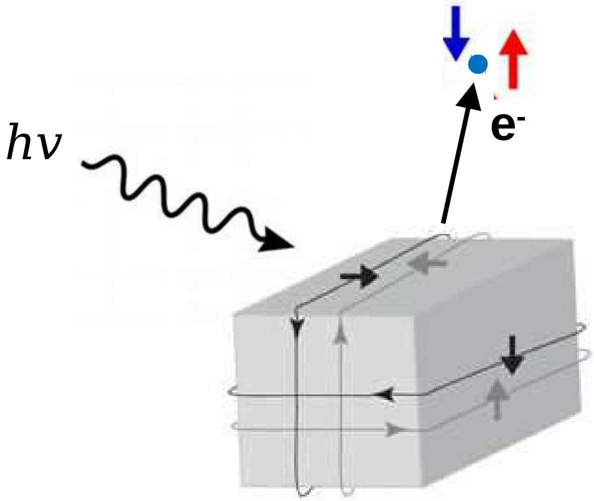
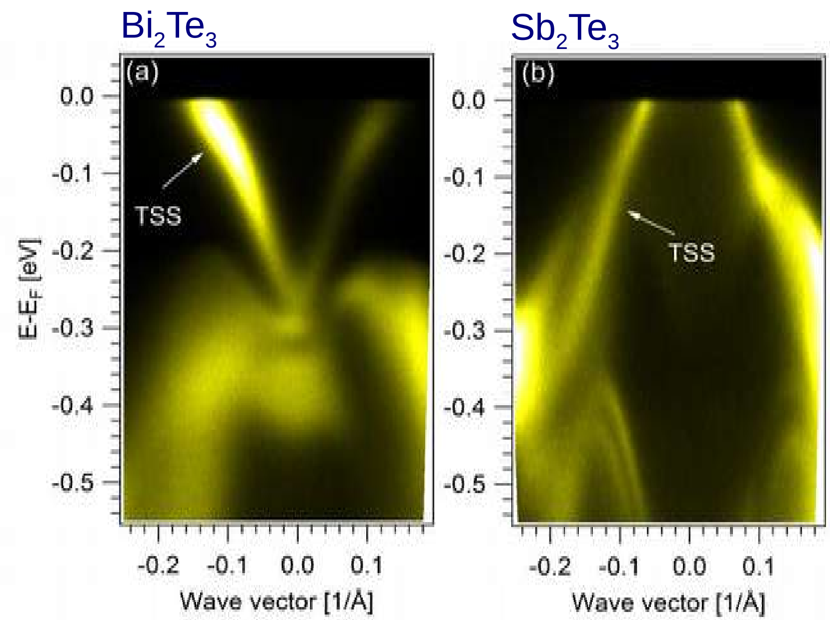
The question posed in the title of this viewpoint article cannot be answered conclusively at this point.

these 'intrinsic' spin polarizations can be expected to be stable with respect to variations in the photon energy and photon polarization direction

Spin- and angle-resolved photoelectron spectroscopy



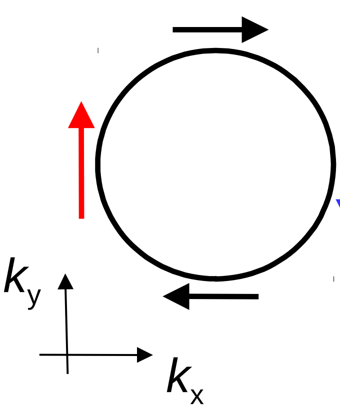


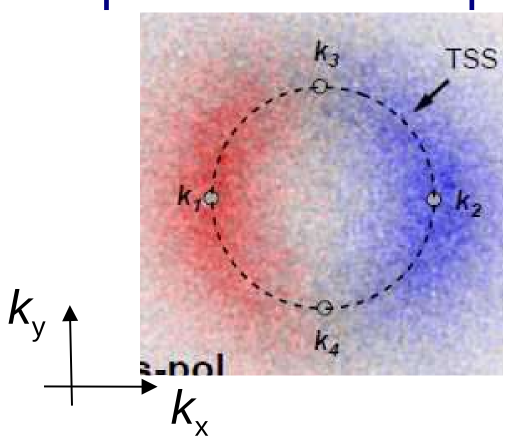
PRL **114** (2015) 066802

?

intrinsic spin

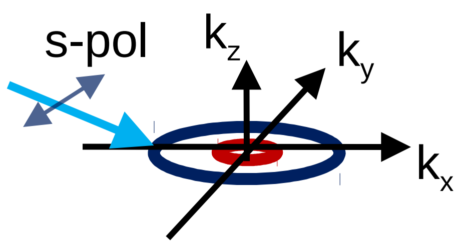
photoelectron spin

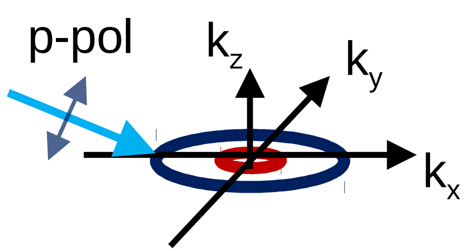




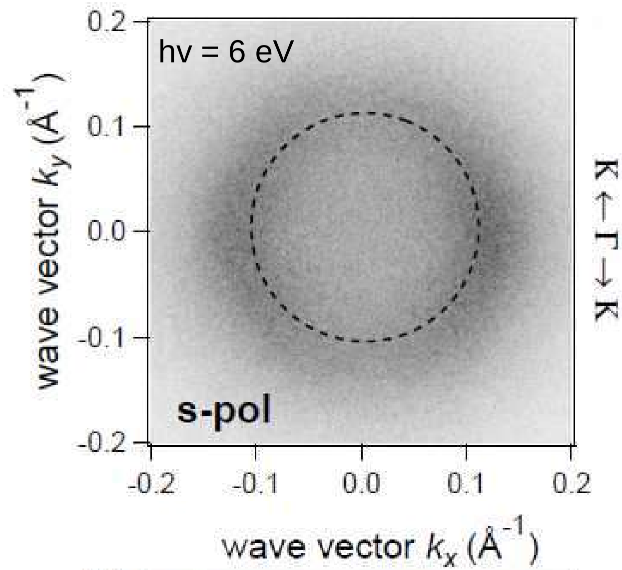
- electronic structure probed with simultaneous energy, momentum, and spin sensitivity
- relation between intrinsic and photoelectron spin texture?

Light-polarization dependence of photoelectron spin

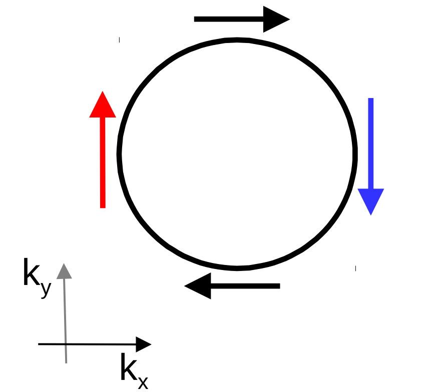


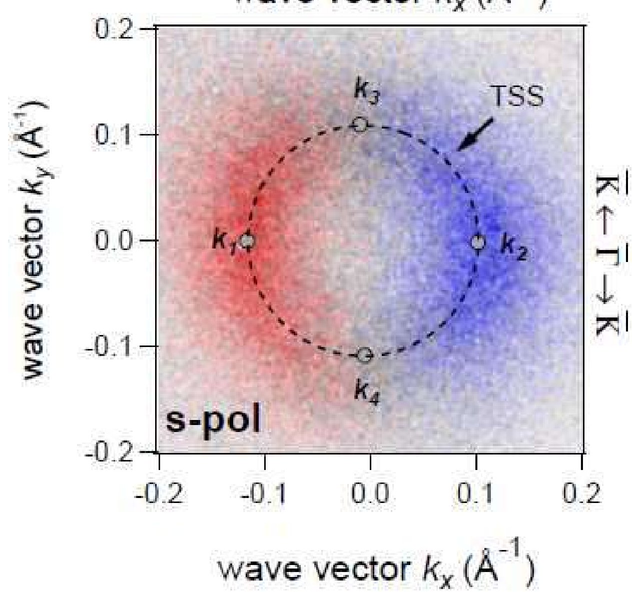


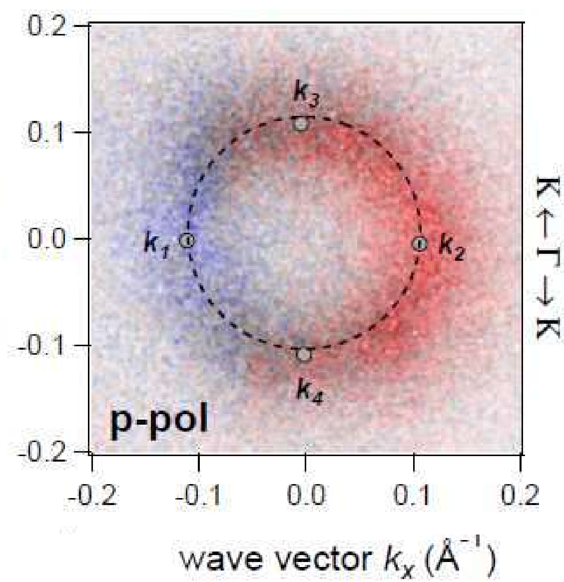
Fermi surface of TSS in Bi_2Te_2Se



expected spin texture

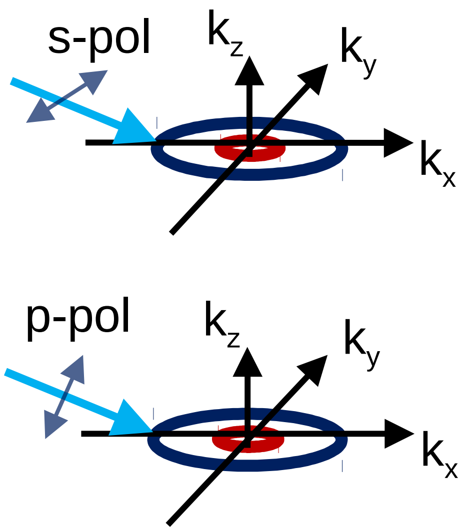


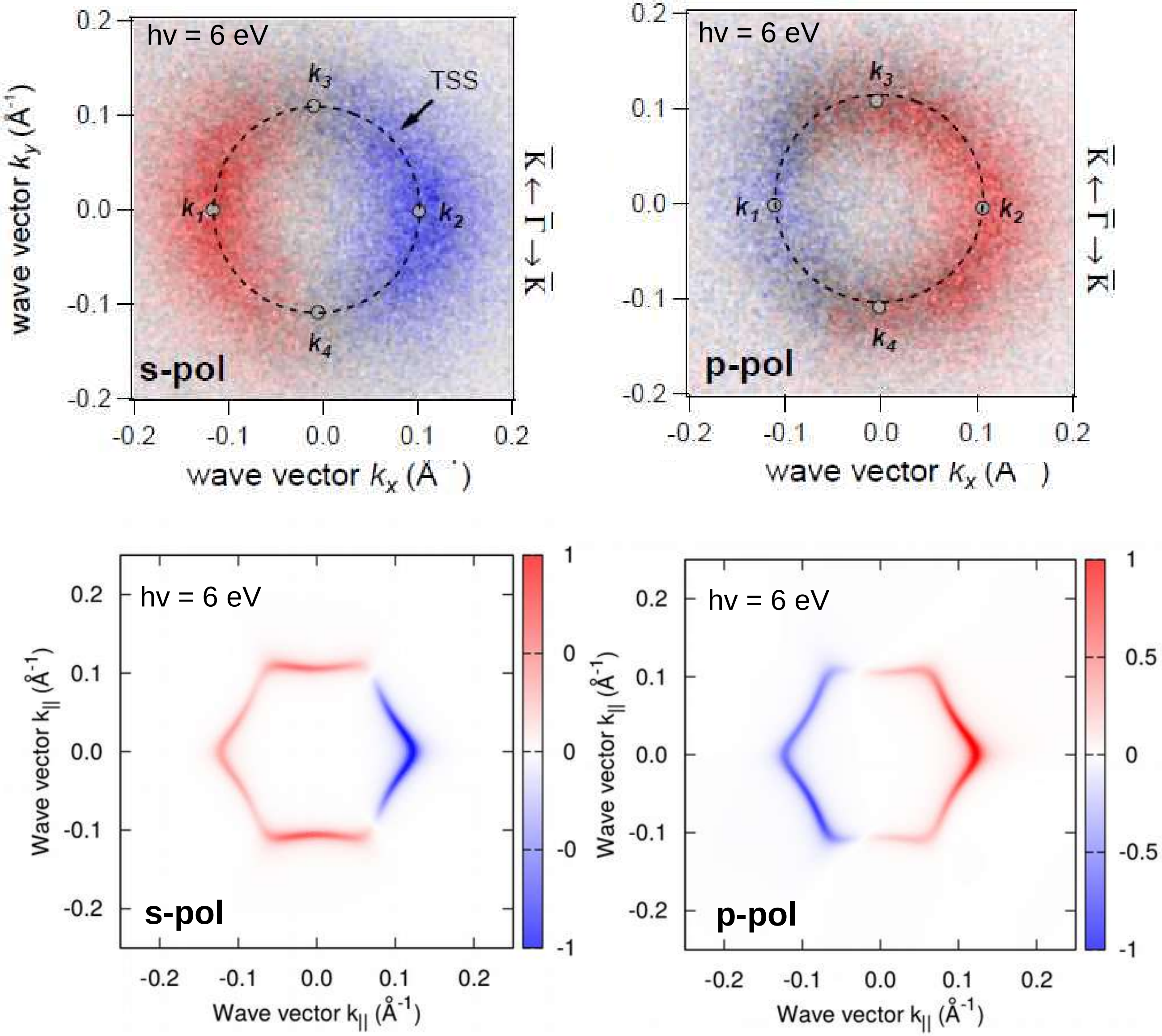




Nature Commun. **7** (2016) 11621

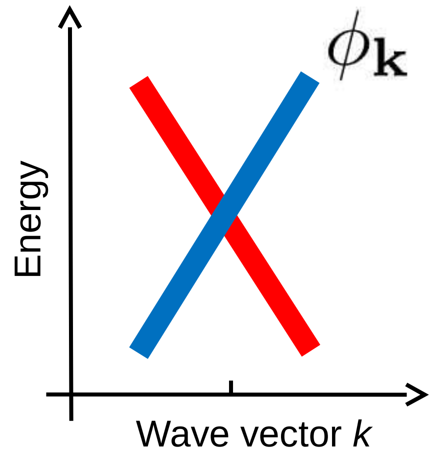
Experiment vs. one-step photoemission theory





Calculations by Jürgen Braun and Jan Minar (LMU, UWB)

Spin-orbit coupling in Bloch wave functions



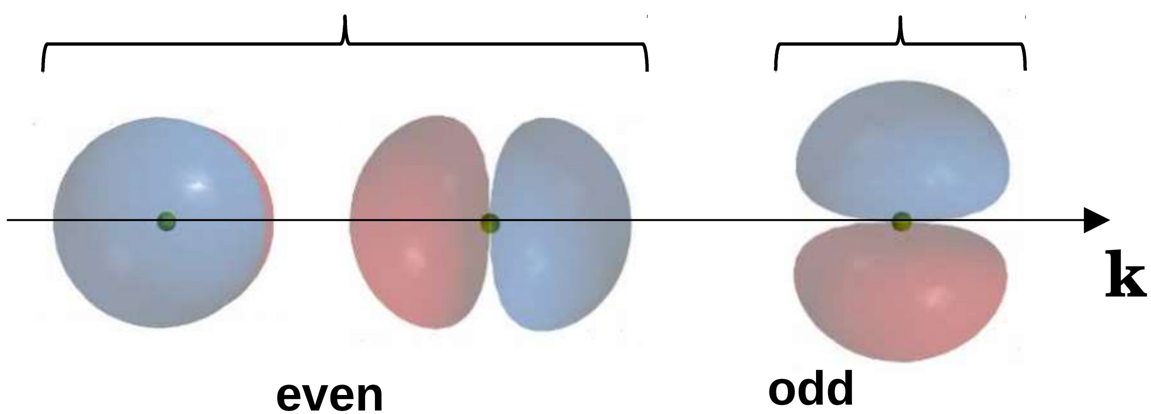
Bloch wave:

$$\phi_{\mathbf{k},\sigma}(\mathbf{r}) = u_{\mathbf{k}}(\mathbf{r})e^{i\mathbf{k}\cdot\mathbf{r}}\chi_\sigma$$

Bloch wave with spin-orbit interaction:

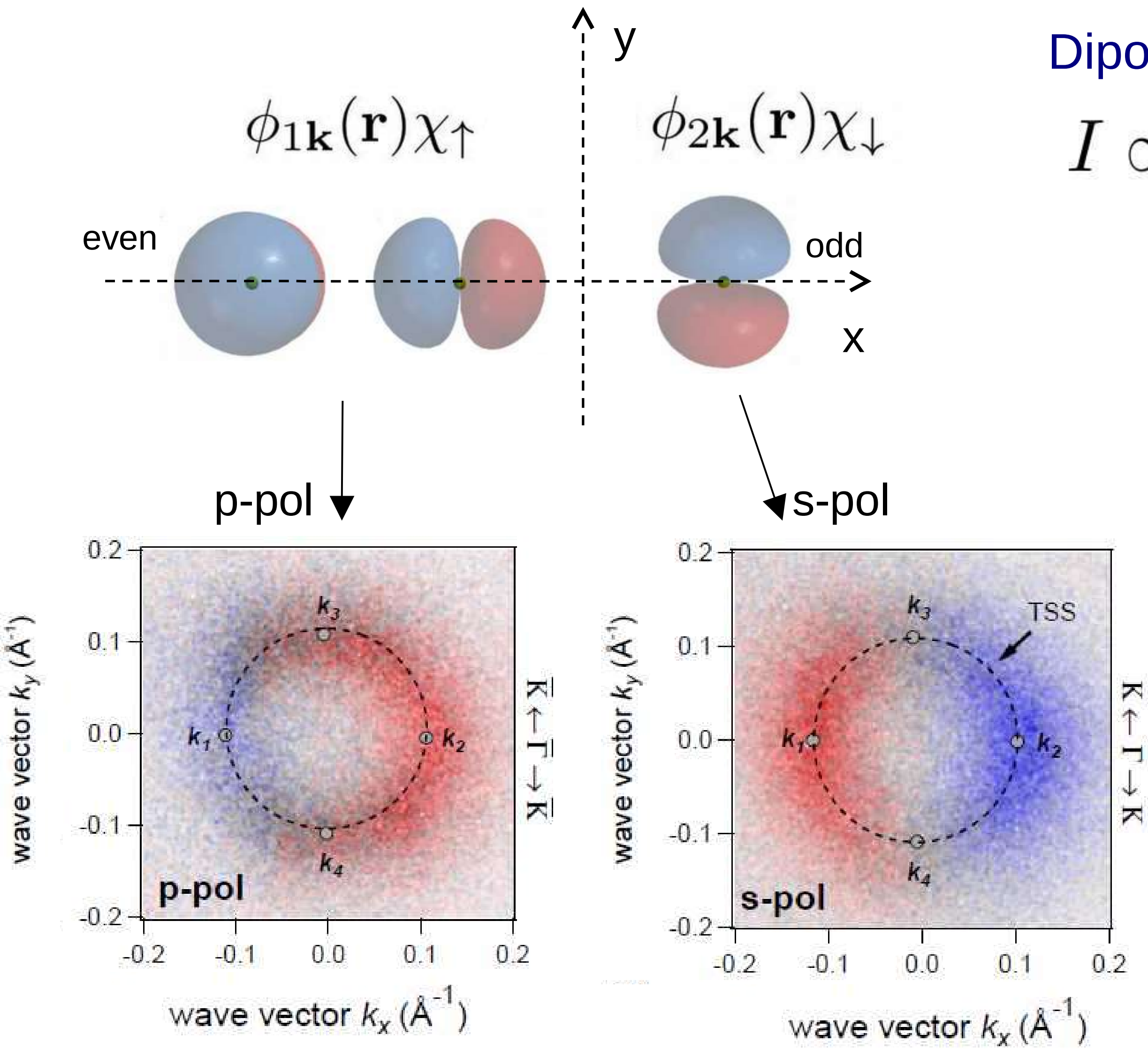
$$\phi_{\mathbf{k}}(\mathbf{r}) = \phi_{1\mathbf{k}}(\mathbf{r})\chi_\uparrow + \phi_{2\mathbf{k}}(\mathbf{r})\chi_\downarrow$$

when $\mathbf{k}$ in mirror plane:



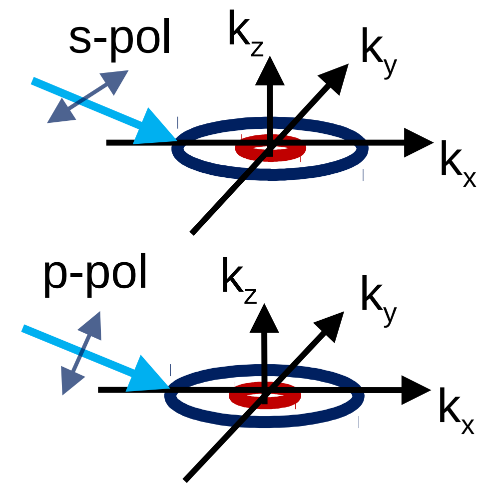
Henk *et al*., PRB**68** (2003) 165416

Dipole selection rules and spin-orbit coupling



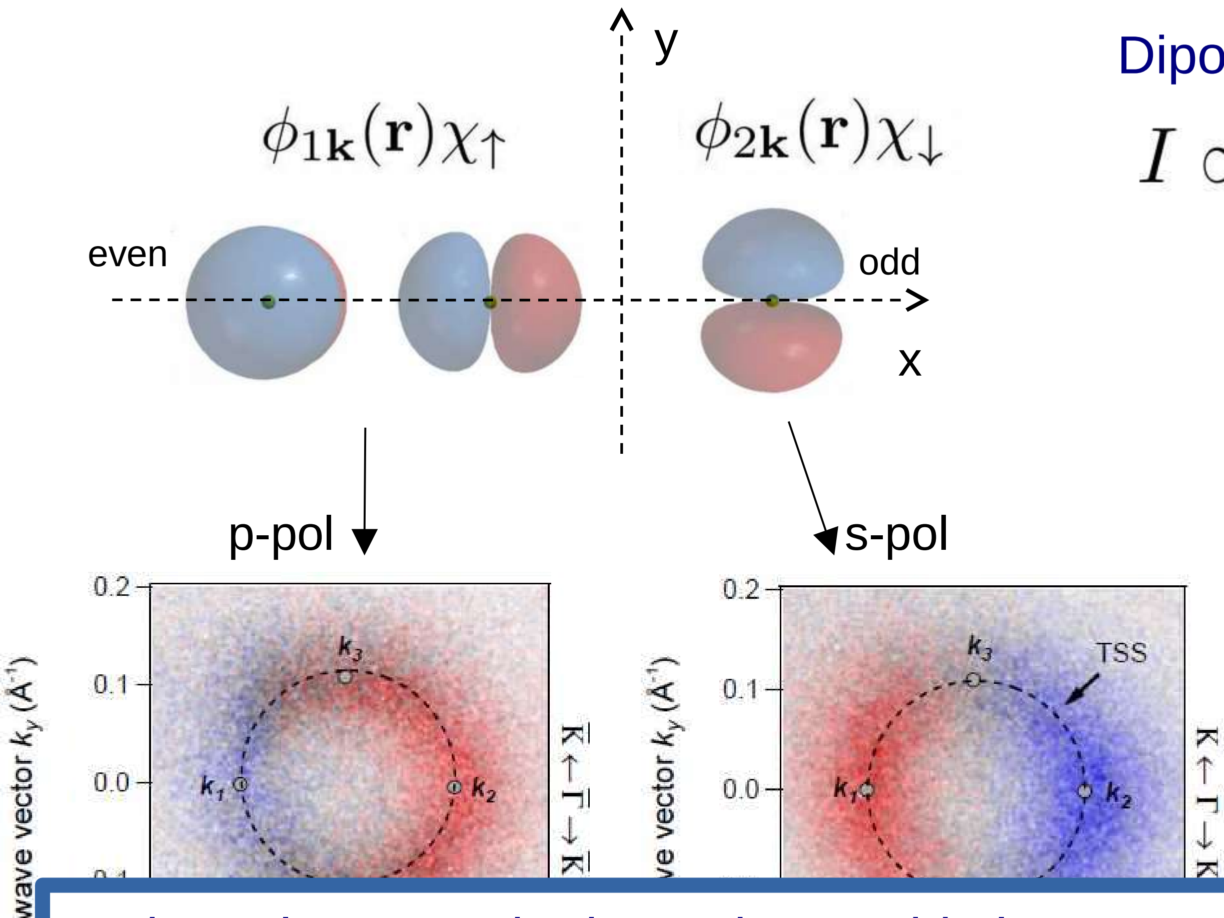
Dipole matrix element:

$$I \propto |\langle \Phi_f | \hat{e} \cdot \mathbf{r} | \phi_{\mathbf{k}} \rangle|^2$$



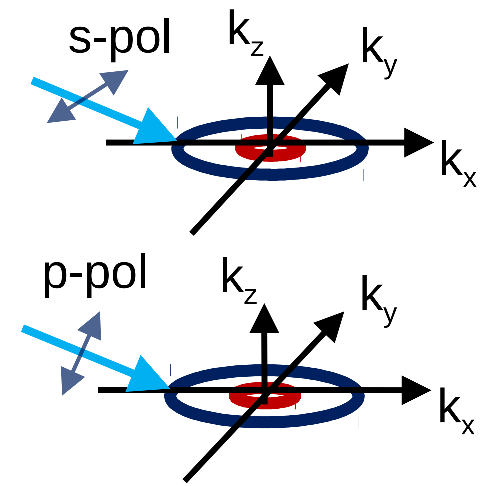
Nature Commun. 7, 11621 (2016)

Dipole selection rules and spin-orbit coupling



Dipole matrix element:

$$I \propto \left| \langle \Phi_f | \hat{e} \cdot \mathbf{r} | \phi_{\mathbf{k}} \rangle \right|^2$$

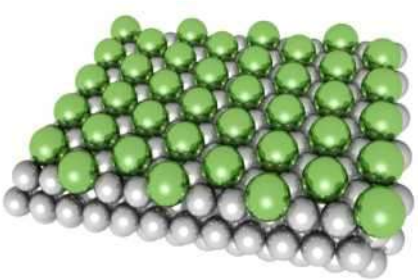


- photoelectron spin depends on orbital symmetry and experimental geometry
- even the observed spin reversion reflects the **intrinsic spin** structure

→ talk by Koji Miyamoto for d-states of W(110)

Nature Commun. 7, 11621 (2016)

Model system: Rashba-split surface states on $BiAg_2$/Ag(111)

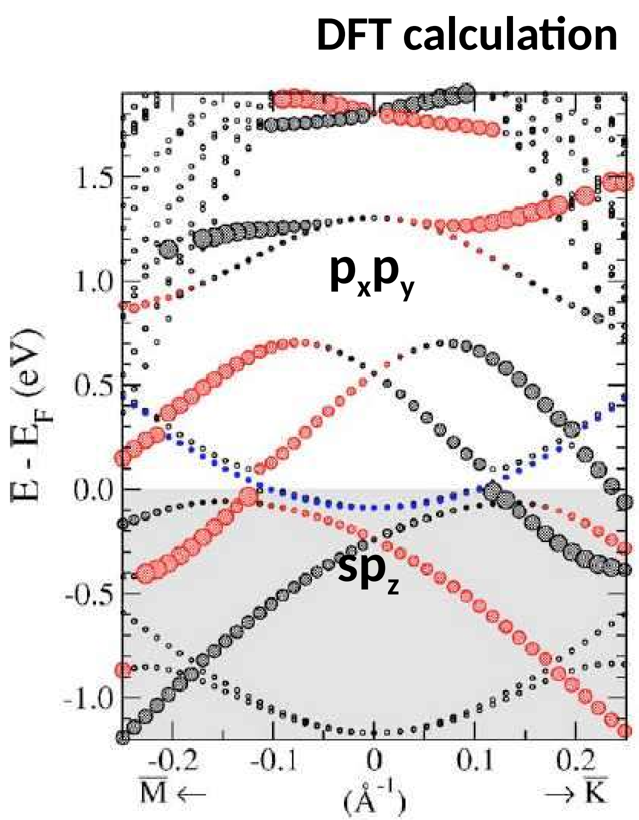


1/3 ML Bi on Ag(111)
(√3x√3)*R*30°-reconstruction

C. Ast *et al.*, PRL **98,** 186807 (2007)

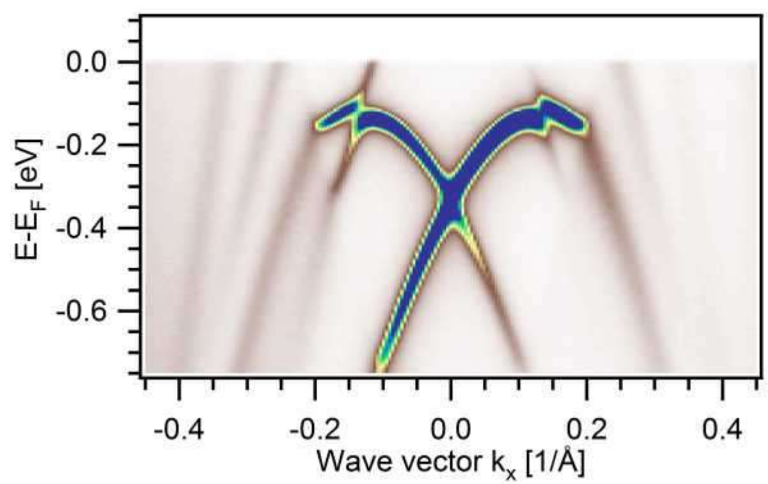
Poster by Ryo Noguchi 1-15

R. Noguchi *et al.*, PRB **95,** 041111(R) (2017)



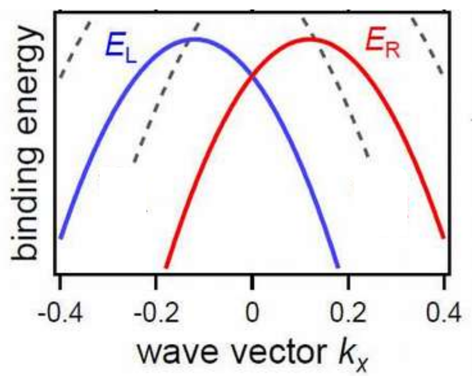
G. Bihlmayer *et al.*, PRB **75,** 195414 (2007)

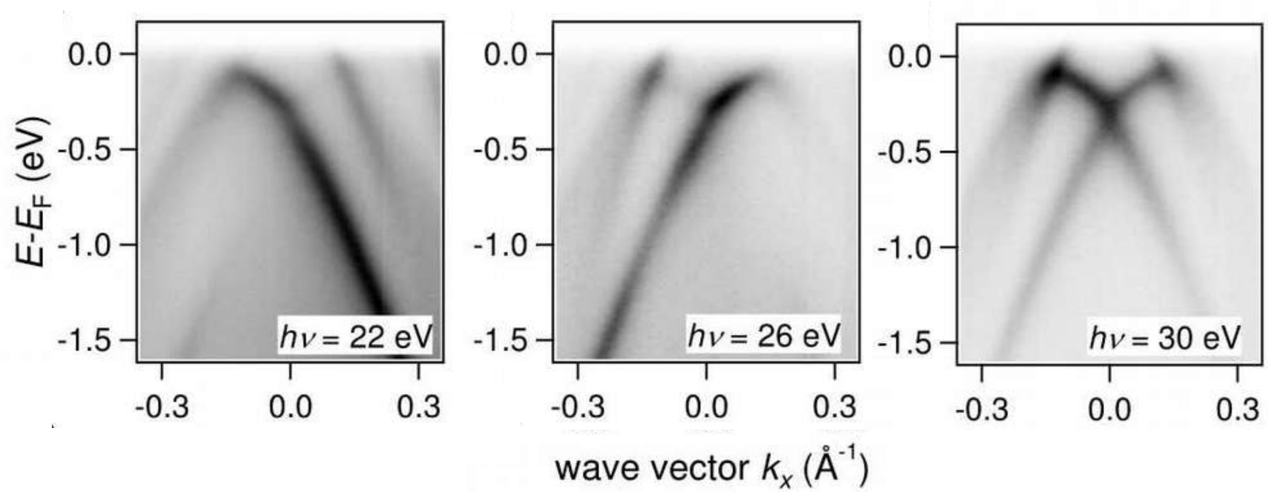
- nearly prefect 2DEG with
- large spin-orbit splitting
  → spin-polarized bands
- occupied states of mainly spz character
- unoccupied states of mainly p_xp_y character

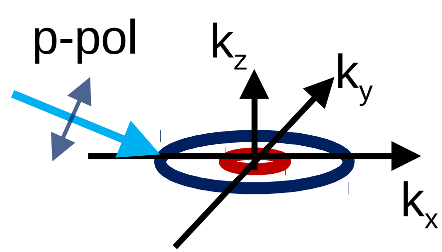


H. Bentmann *et al.*, EPL **87,** 37003 (2009)

Linear dichroism for spin-split states in $BiAg_2/Ag(111)$







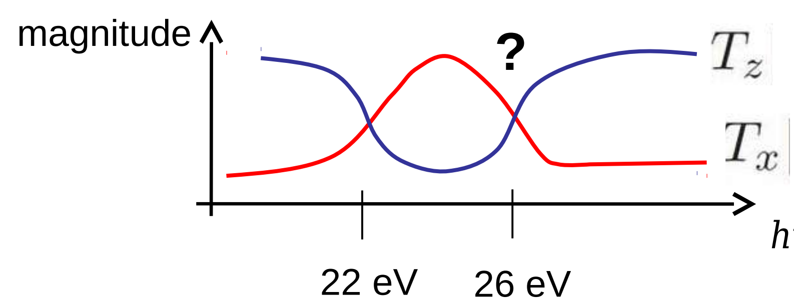
$$I_R \propto |\langle \Phi_f | E_z \hat{z} | \phi_{\mathbf{k}} \rangle + \langle \Phi_f | E_x \hat{x} | \phi_{\mathbf{k}} \rangle|^2$$

$$I_L \propto |\langle \Phi_f | E_z \hat{z} | \phi_{\mathbf{k}} \rangle - \langle \Phi_f | E_x \hat{x} | \phi_{\mathbf{k}} \rangle|^2$$

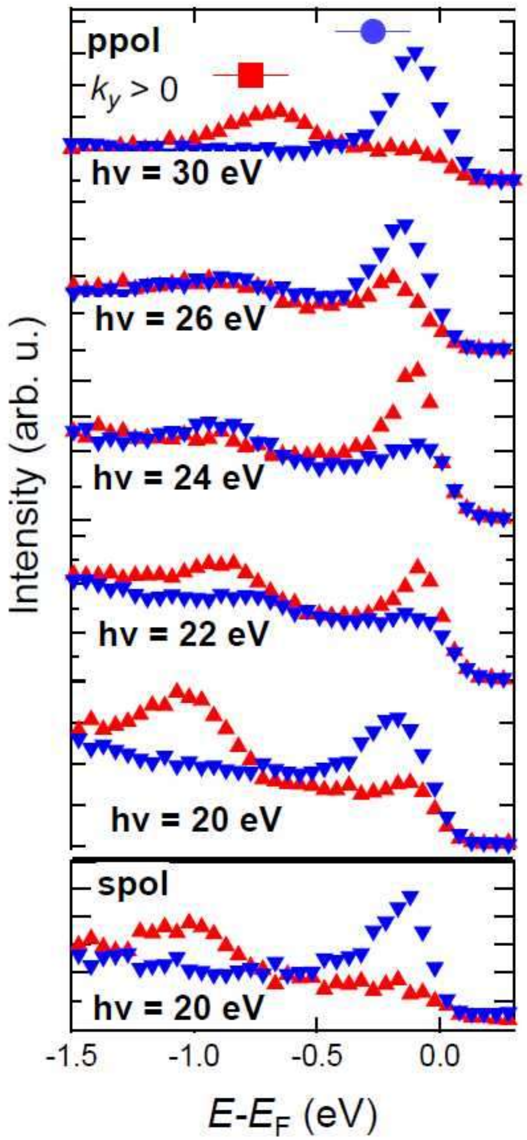
$$\downarrow \qquad\qquad\qquad \downarrow$$

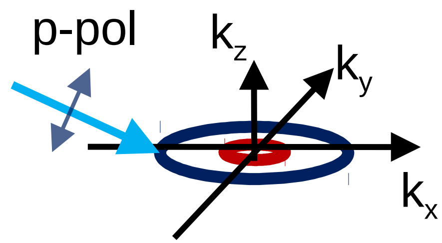
$$T_z \qquad\qquad\qquad T_x$$

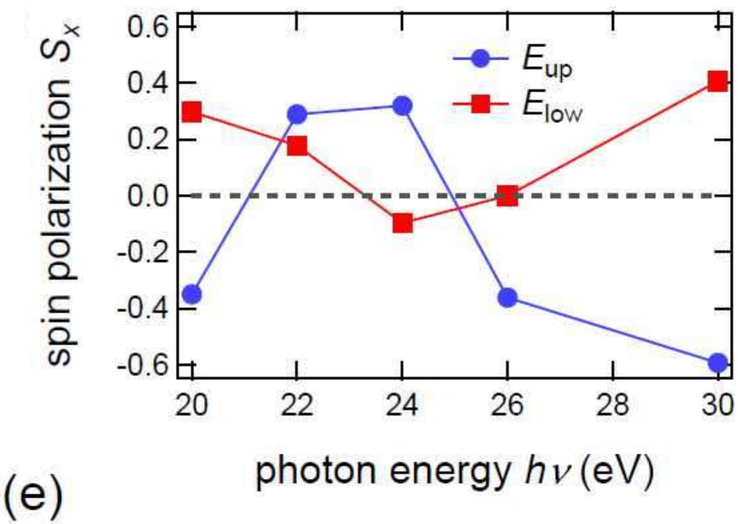
→ T_x and T_z vary with $h\nu$, due to **change of the final state** Φ_f



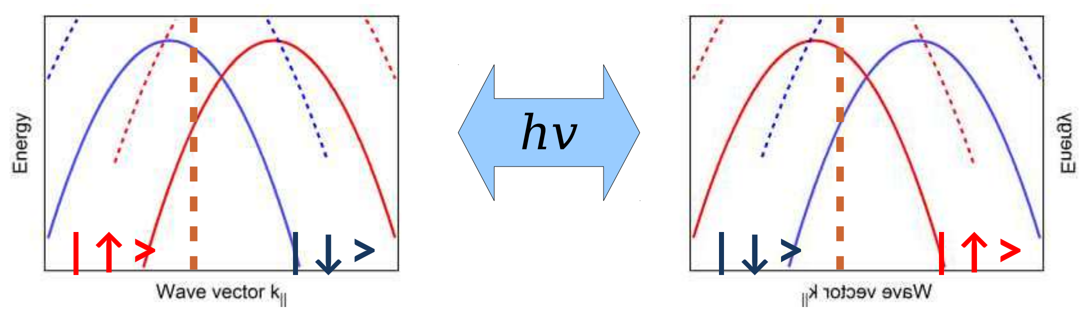
Photon energy dependence of spin-polarization





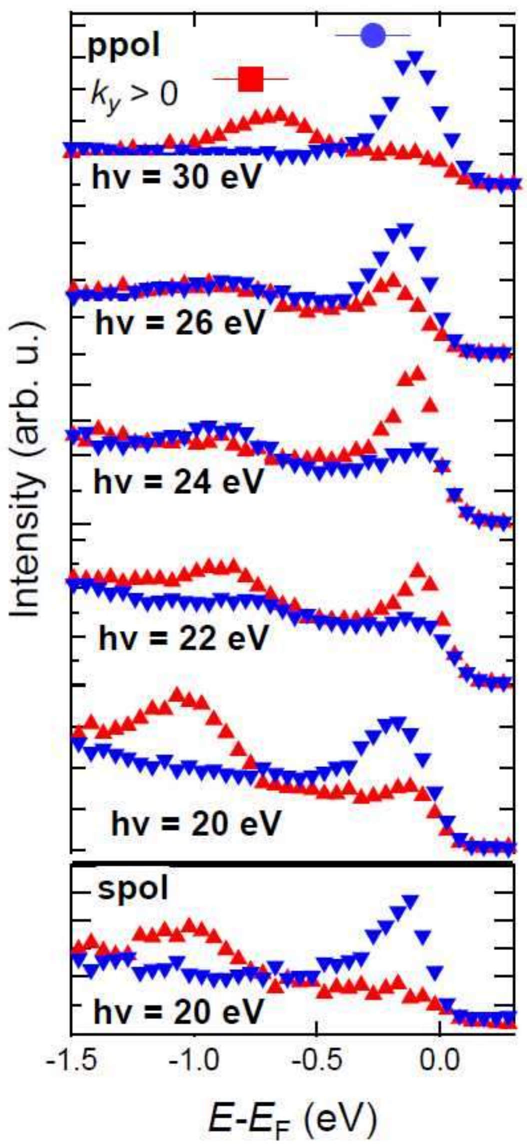


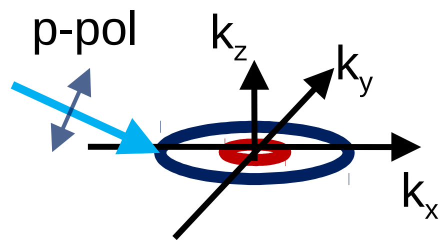
→ measured spin-polarization reverses within a few eV of photon energy!

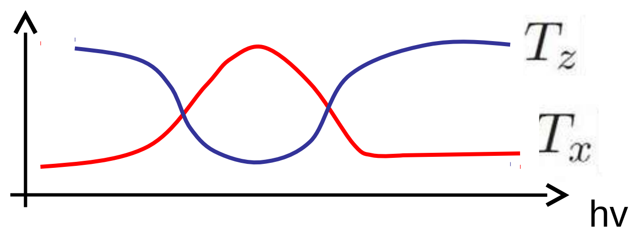


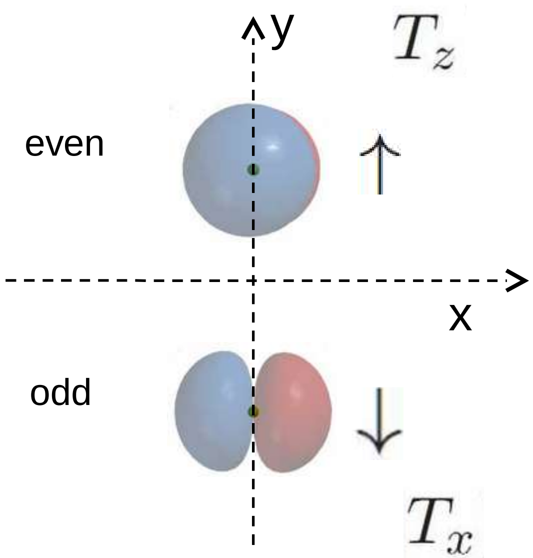
H. Bentman, FR *et al.*, PRL(2017) accepted

Photon energy dependence of spin-polarization





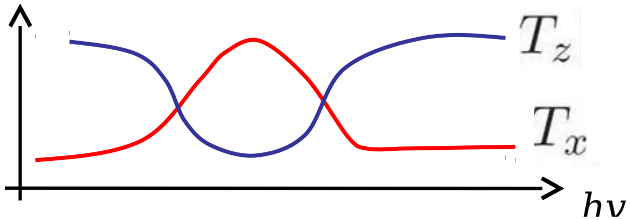


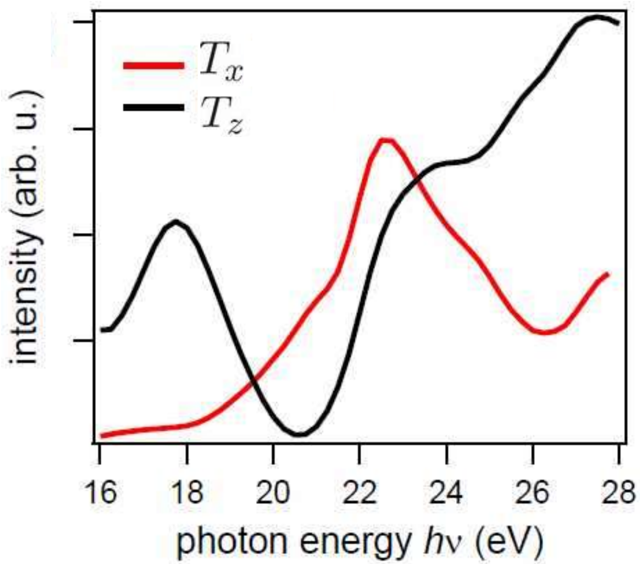


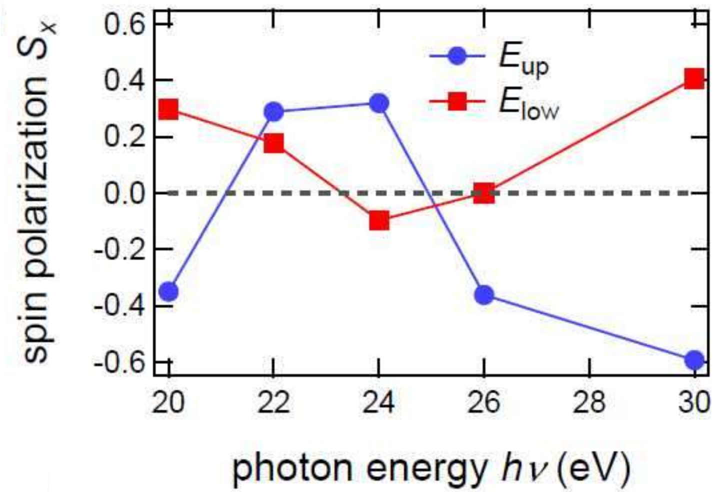
- photoelectron spin flips sign with excitation energy
- T_z produces spin-↑ and T_x spin-↓ photoelectrons

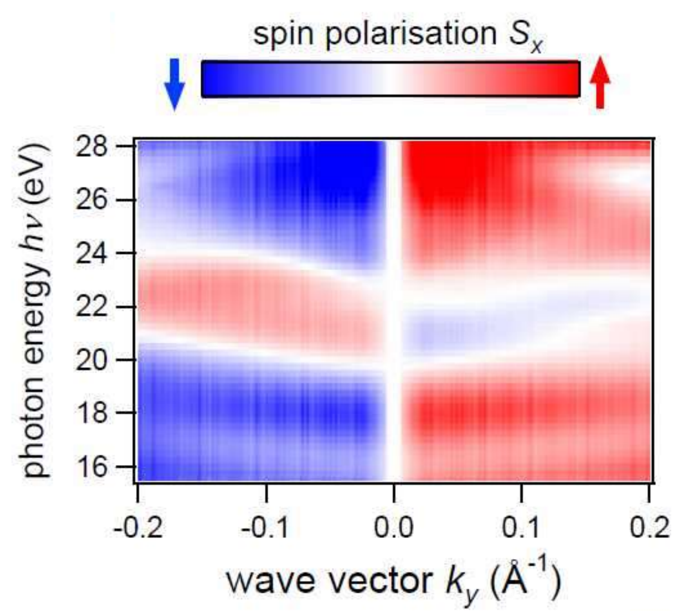
H. Bentman, FR *et al.*, PRL(2017) accepted

Ab initio one-step photoemission theory





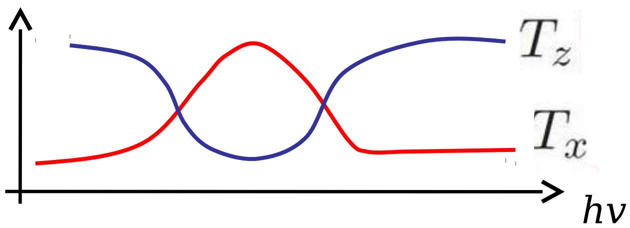


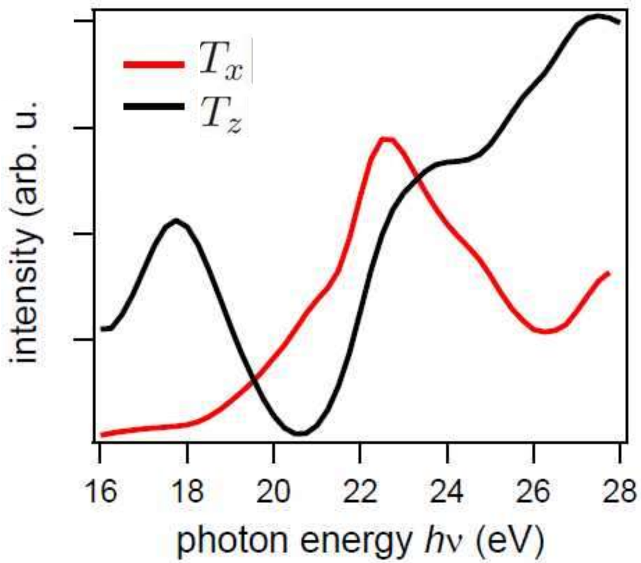


E. E. Krasovskii
DIPC San Sebastian

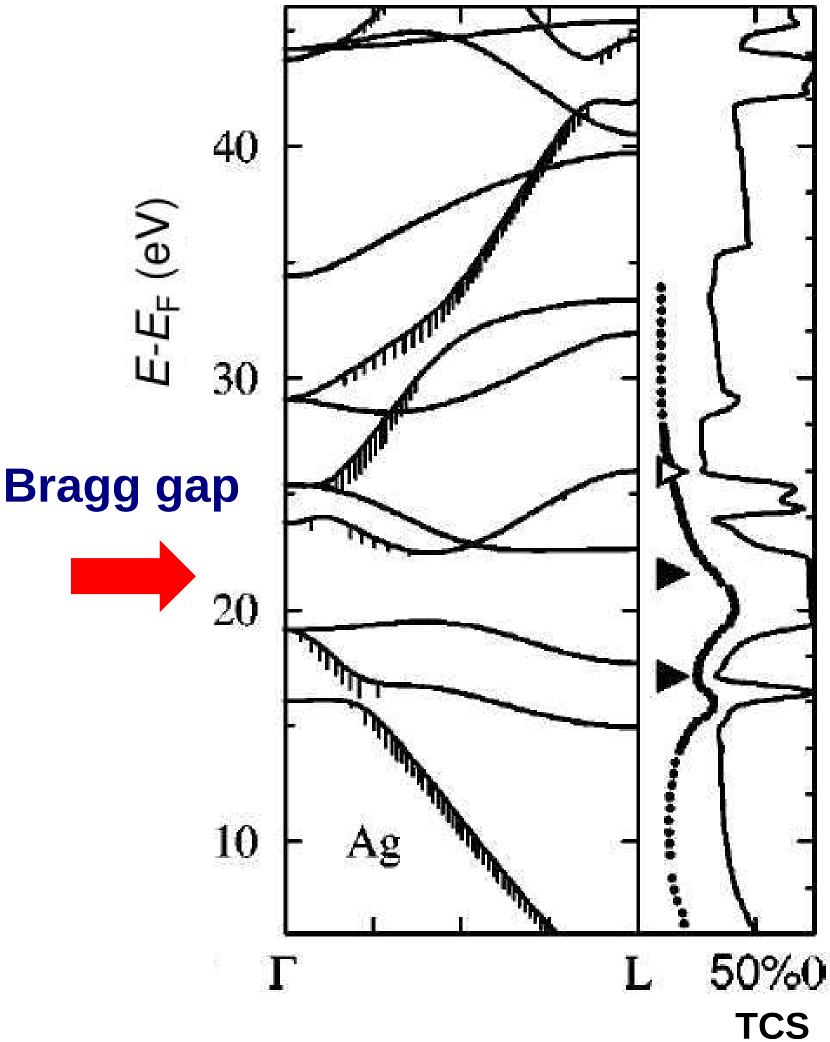
→ $h\nu$-dependence
captured by
ab initio theory

Ab initio one-step photoemission theory





Ag(111) band structure above E_F



Krasovskii, Schattke, PRB **59** (1999) 15609(R)

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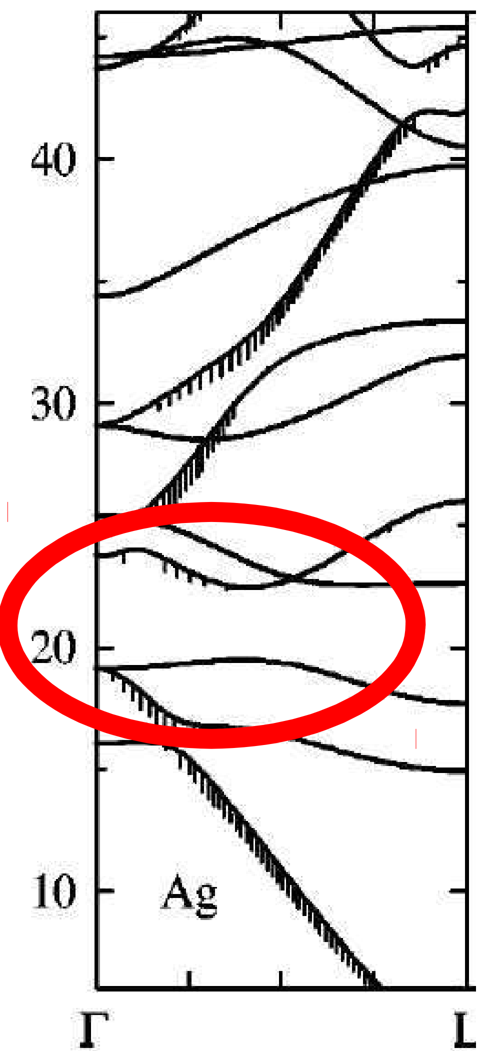
DIPC San Sebastian

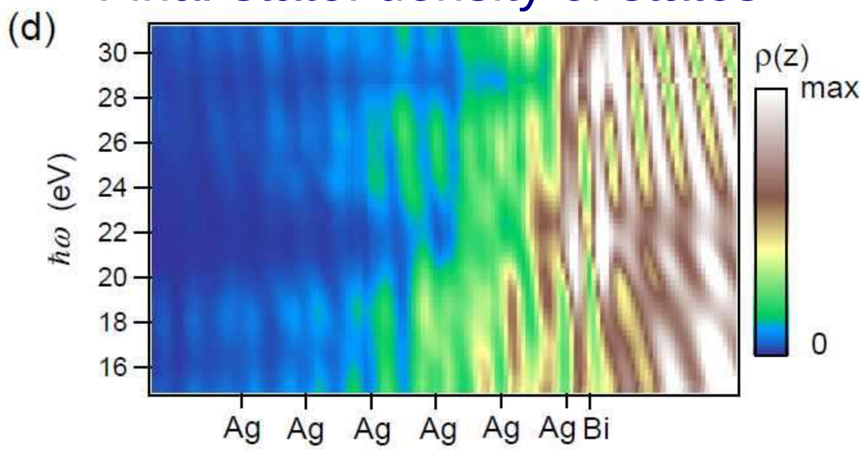
→ *hν*-dependence
captured by
ab initio theory

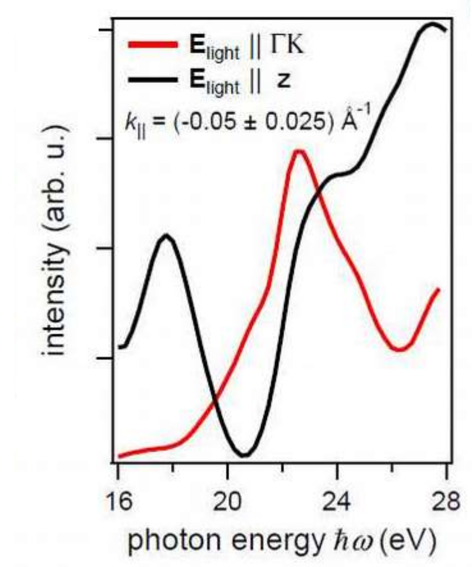
Complex unoccupied band structure: PE Final state

Ag(111)



Final-state: density of states



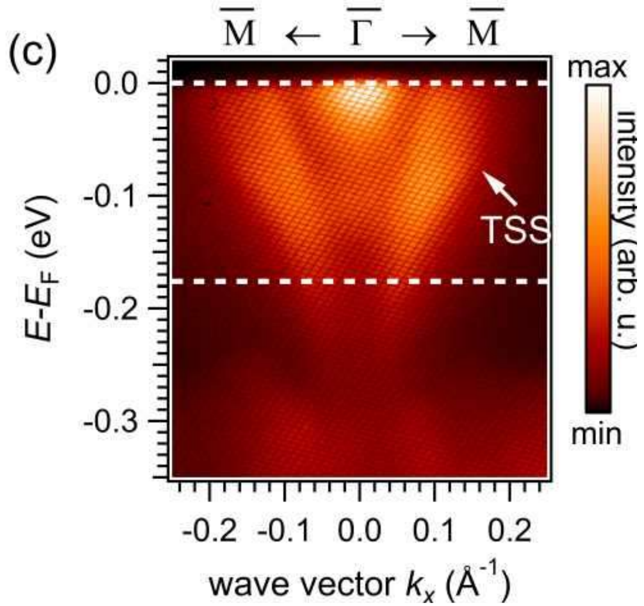


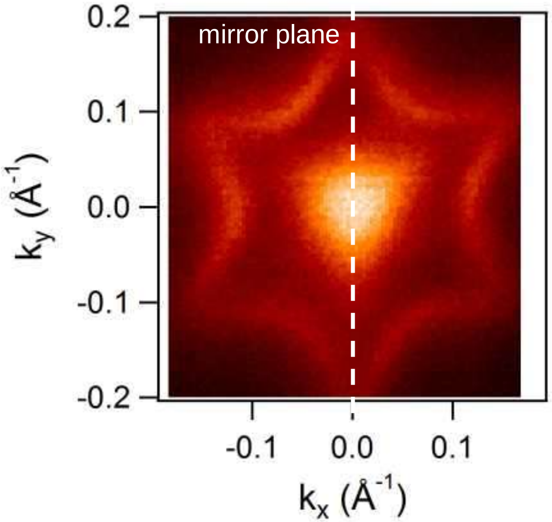
- propagating and evanescent waves
- gap: only evanescent solutions

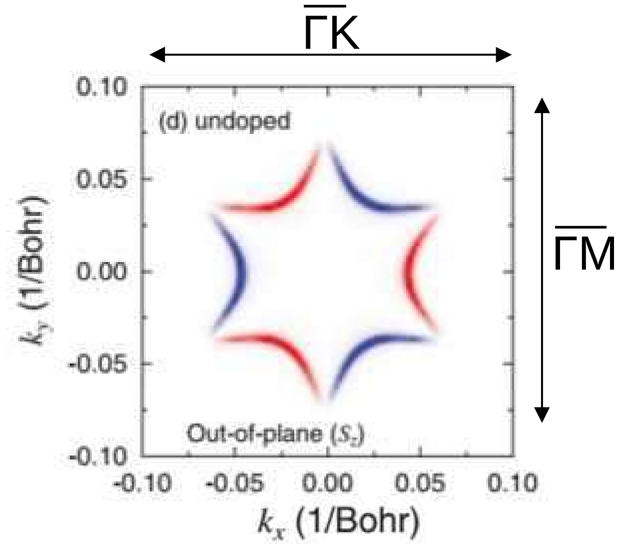
→ rapid change of final-state wave function

Krasovskii, Schattke, PRB **59** (1999) 15609(R)

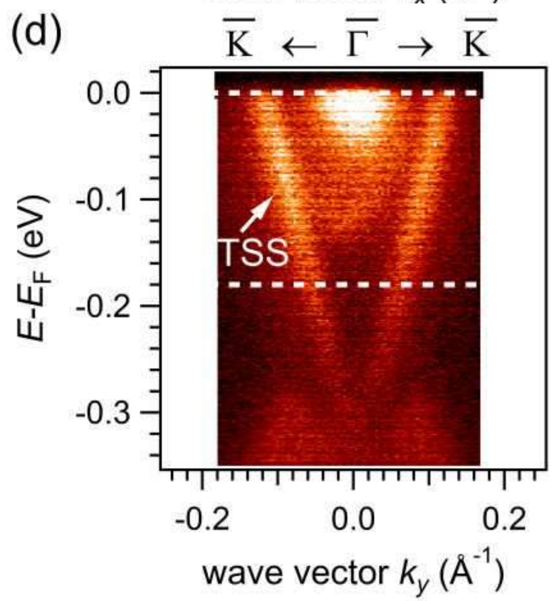
Topological surface state in Bi_2Te_3





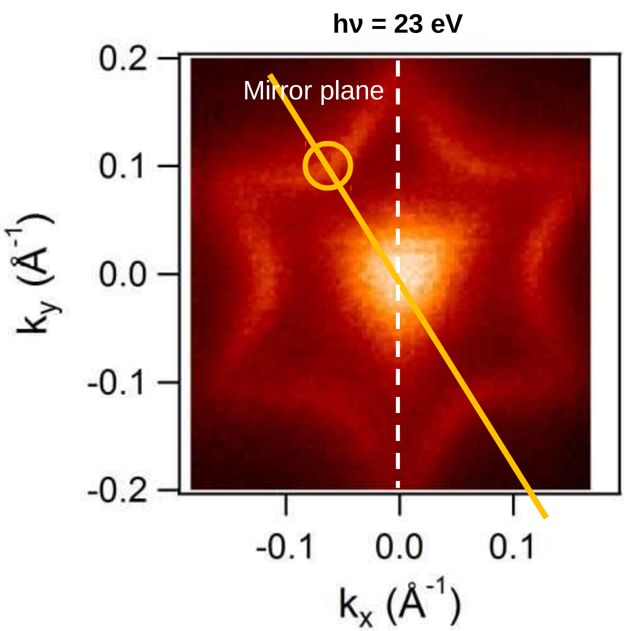


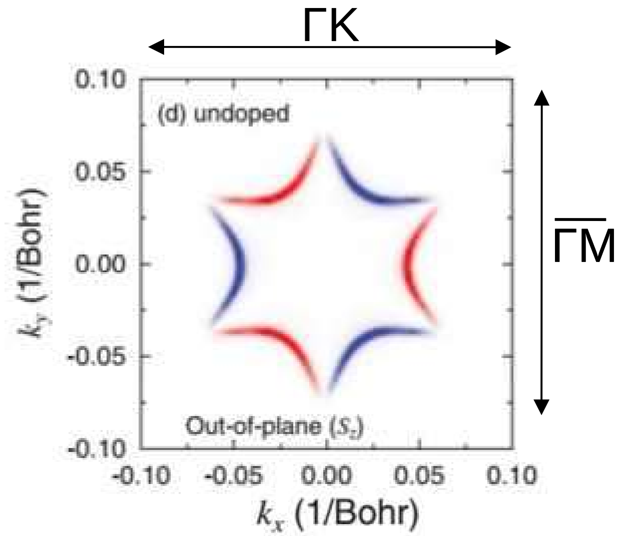
Henk *et al.*, PRL**108** (2012) 206801



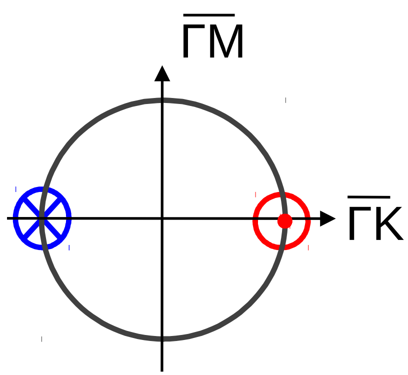
- strong hexagonal warping of surface-state dispersion
- in-plane potential landscape
- spin-polarization acquires **out-of-plane component** off the mirror planes

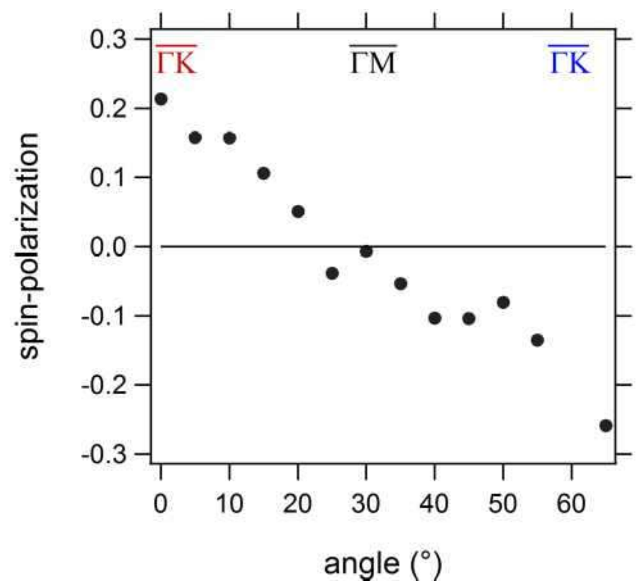
Out-of-plane spin polarization in Bi_2Te_3

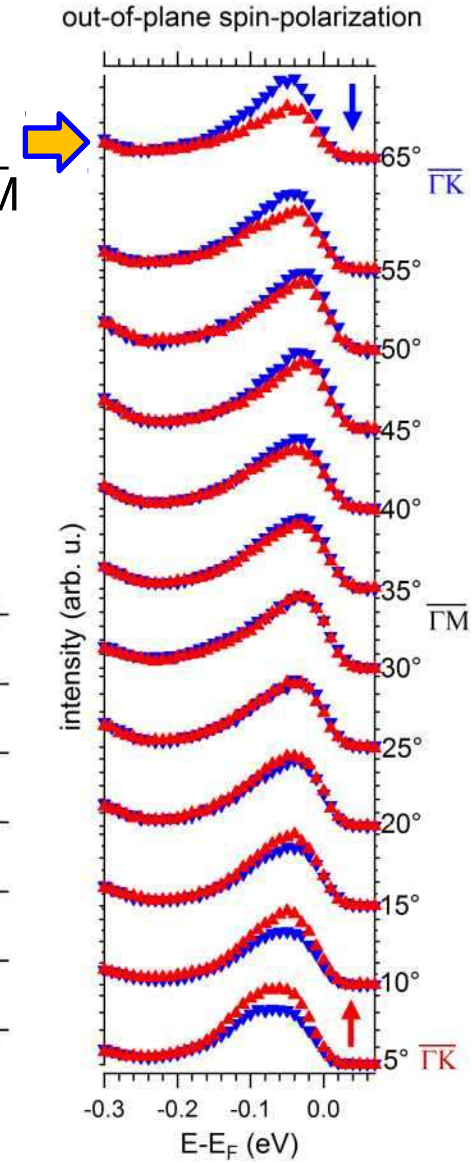




Henk *et al.*, PRL**108** (2012) 206801



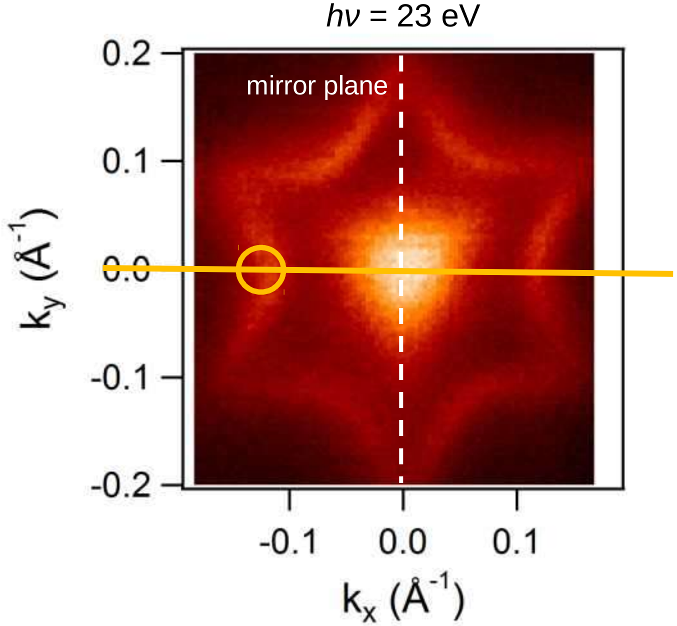


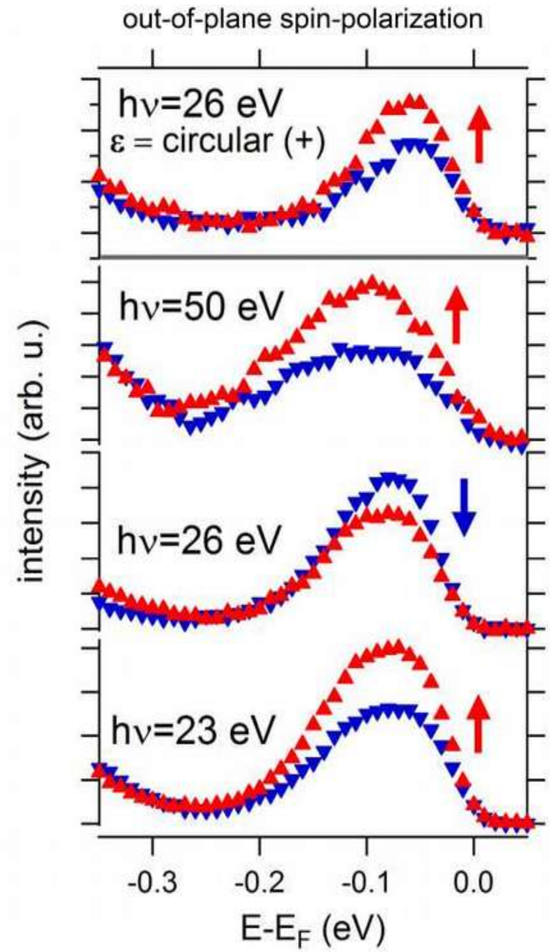


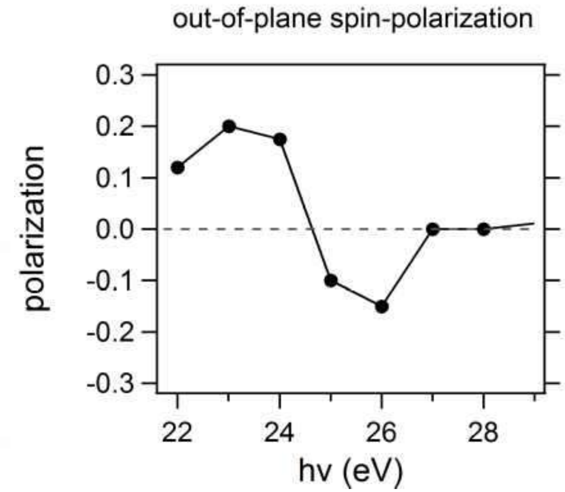
Unpublished: HiSOR (ESPRESSO), Hiroshima

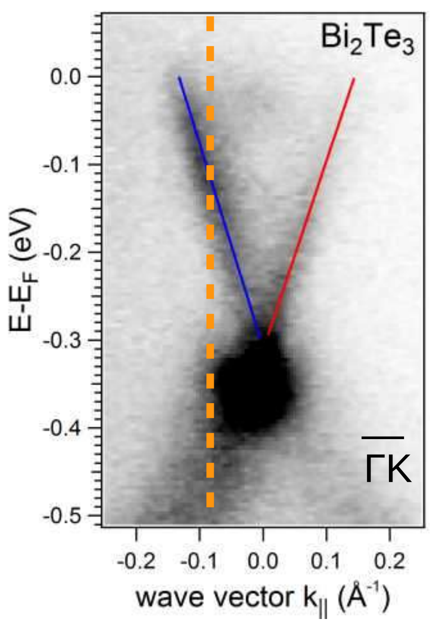
H. Bentman, FR *et al.*, (2017) unpublished

Effect of photon-energy on out-of-plane spin polarization



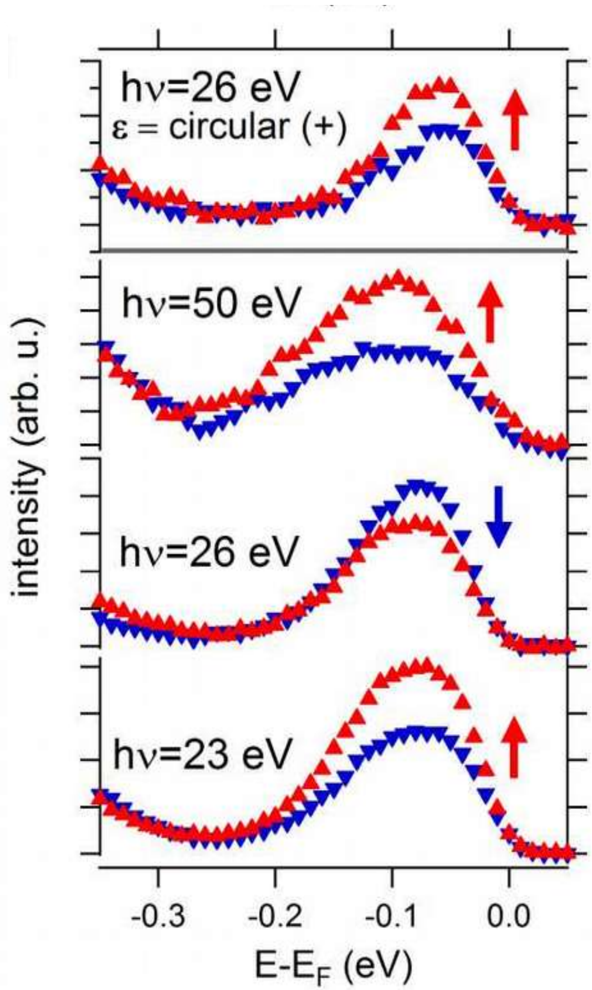


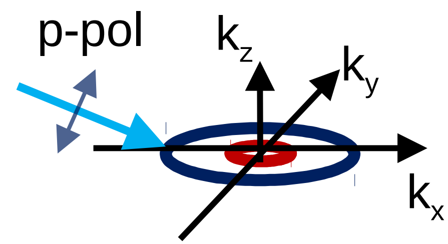




→ measured P_z does **not** reflect intrinsic spin polarization along z (here), but arises mainly from the photoemission process itself

Photon-energy-dependence of out-of-plane spin polarization





Matrix elements:

$$\phi_{\mathbf{k}}(\mathbf{r}) = \phi_{1\mathbf{k}}(\mathbf{r})\chi_{\uparrow} + \phi_{2\mathbf{k}}(\mathbf{r})\chi_{\downarrow}$$

$$\phi_{1\mathbf{k}}(\mathbf{r})\chi_{\uparrow} \xrightarrow{T_1} \Phi_f \xleftarrow{T_2} \phi_{2\mathbf{k}}(\mathbf{r})\chi_{\downarrow}$$

k along ΓK (no mirror plane) $T_1, T_2 \neq 0$

Photoelectron spin:

$$\mathbf{P}_f = \begin{pmatrix} |T_1|^2 - |T_2|^2 \\ |T_1||T_2|cos(\varphi_1 - \varphi_2) \\ |T_1||T_2|sin(\varphi_1 - \varphi_2) \end{pmatrix}$$

H. Bentman, FR *et al.*, (2017) unpublished

Summary

$$I \propto |\langle \Phi_f | \hat{e} \cdot \mathbf{r} | \phi_{\mathbf{k}} \rangle|^2$$

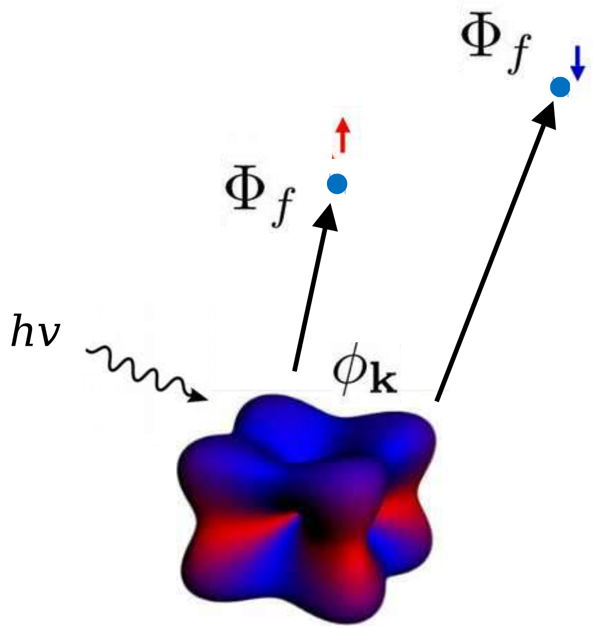
1) SOC couples even and odd WFs differently to the "intrinsic" spin in $\phi_{\mathbf{k}}$

2) Experimental geometry (light polarization) "selects" WF and therewith the (photoelectron) spin (→ linear dichroism)

3) Different contributions do **interfere** in squared matrix element

4) If strong **energy dependence** of Φ_f (e.g. near gap), interference terms might produce changes of photoelectron spin by changing $h\nu$

Conclusions



$$I \propto |\langle \Phi_f | \hat{e} \cdot \mathbf{r} | \phi_{\mathbf{k}} \rangle|^2$$

- Photoelectron spin polarization can strongly depend on experimental parameters
- Spin density of initial state is sampled differently depending on experimental geometry and excitation energy
- In general: comprehensive data and/or additional knowledge from theory or experiment is required
- Outlook: soft X-ray SARPES

  & experimental info about final states

Fig. from Jackeli *et al.*, Phys. Rev. Lett. 102, 017205 (2009)