Soft X-ray photoelectron spectroscopy and its applications

Hugo Dil

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- Introduction to photoelectron spectroscopy
- Case studies on BiTeCl and topological insulator
- Spin- and angle-resolved photoemission spectroscopy
- Manipulating spin texture of 2DEG on $SrTiO_3$

  E.B. Guedes et al. Physical Review Research 2, 033173 (2020)
- Operando (S)ARPES on multiferroic (Ge,Mn)Te

  J. Krempasky et al. Physical Review X 8, 021067 (2018)
- Measuring time without a clock

  M. Fanciulli et al. Physical Review Letters 118, 067402 (2017)

Photoelectric effect

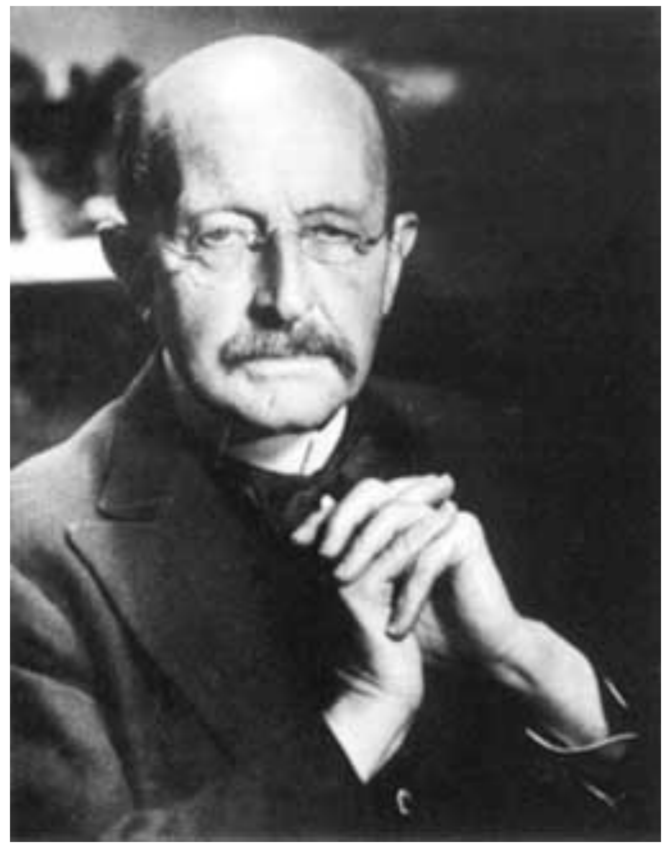
Einstein's Photoelectric Equation

The electron leaves the body with energy

$$\tfrac{1}{2}mv^2 = hv - P,$$

where h is Planck's constant, v is the light frequency and P is the work the electron has to do in leaving the body.

Albert Einstein, 1905



In his 1913 letter nominating Einstein for the membership of Prussian Academy, Max Planck wrote:

"In sum, one can say there is hardly one among the great problems in which modern physics is so rich to which Einstein has not made a remarkable contribution. That he may sometimes have missed the target in his speculations, as, for example, in his hypothesis of light quanta, cannot really be held too much against him, for it is not possible to introduce really new ideas even in the most exact sciences without sometimes taking a risk."

X-ray photoelectron spectroscopy (XPS)

PHYSICAL REVIEW VOLUME 105, NUMBER 5 MARCH 1, 1957

Precision Method for Obtaining Absolute Values of Atomic Binding Energies

CARL NORDLING, EVELYN SOKOLOWSKI, AND KAI SIEGBAHN
Department of Physics, University of Uppsala, Uppsala, Sweden
(Received January 10, 1957)

WE have recently developed a precision method of investigating atomic binding energies, which we believe will find application in a variety of problems in atomic and solid state physics. In principle, the method is an old one: a magnetic analysis of electrons expelled from a substance exposed to x-radiation. Previous attempts in this direction have, however, given considerably less information about atomic structure than ordinary x-ray spectroscopic experiments, and some twenty years ago the method seems to have been definitely abandoned. We have introduced a number of improvements, both regarding the intensity and, in particular, the accuracy (a factor 100), which now enables us to measure atomic binding energies with an accuracy of one single electron volt from microgram quantities. The definition of the lines is essentially limited by the natural line widths of the atomic levels themselves. There is no shift of the lines due to electron scattering or similar causes, which could introduce systematic errors.

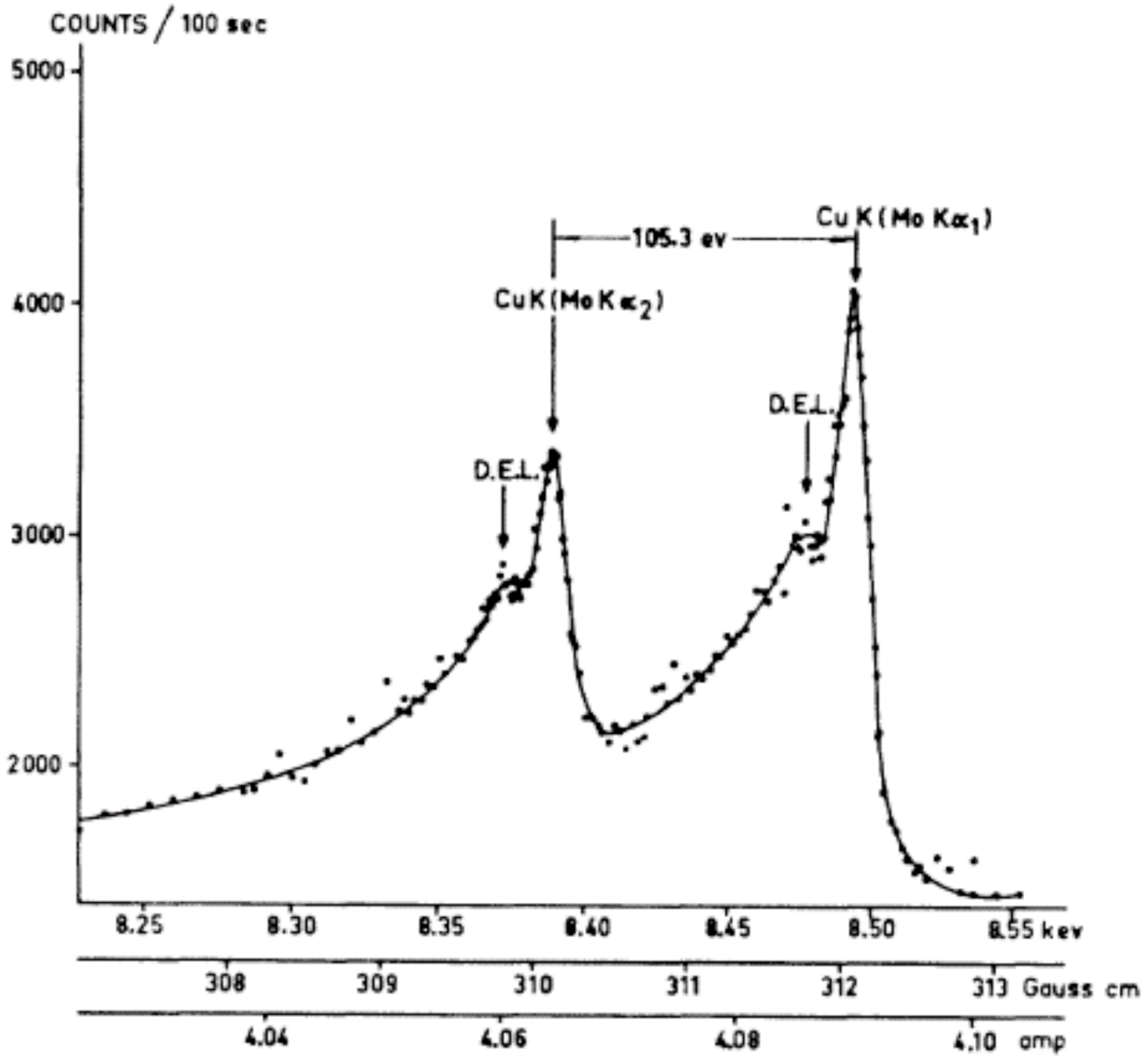
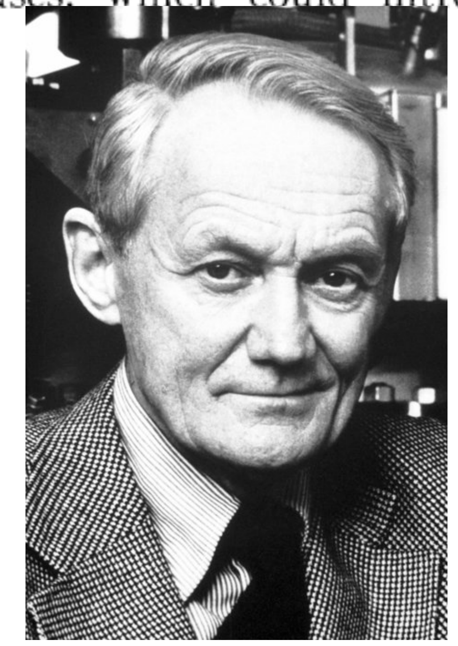
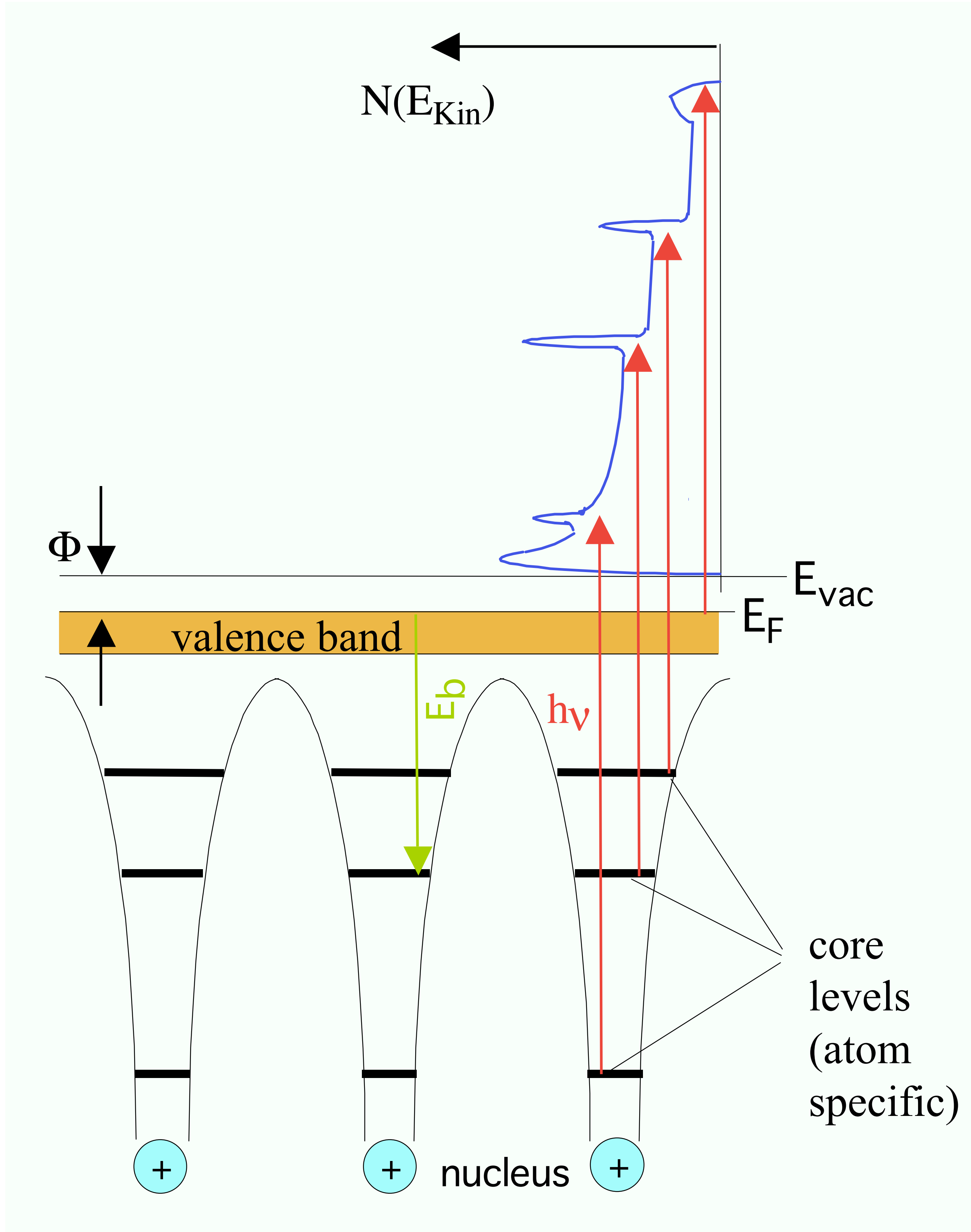


FIG. 1. Lines resulting from photoelectrons expelled from Cu by Mo $K\alpha_1$ and Mo $K\alpha_2$ x-radiation. The satellites marked D.E.L. are interpreted as due to electrons which have suffered a discrete energy loss when scattered in the source.



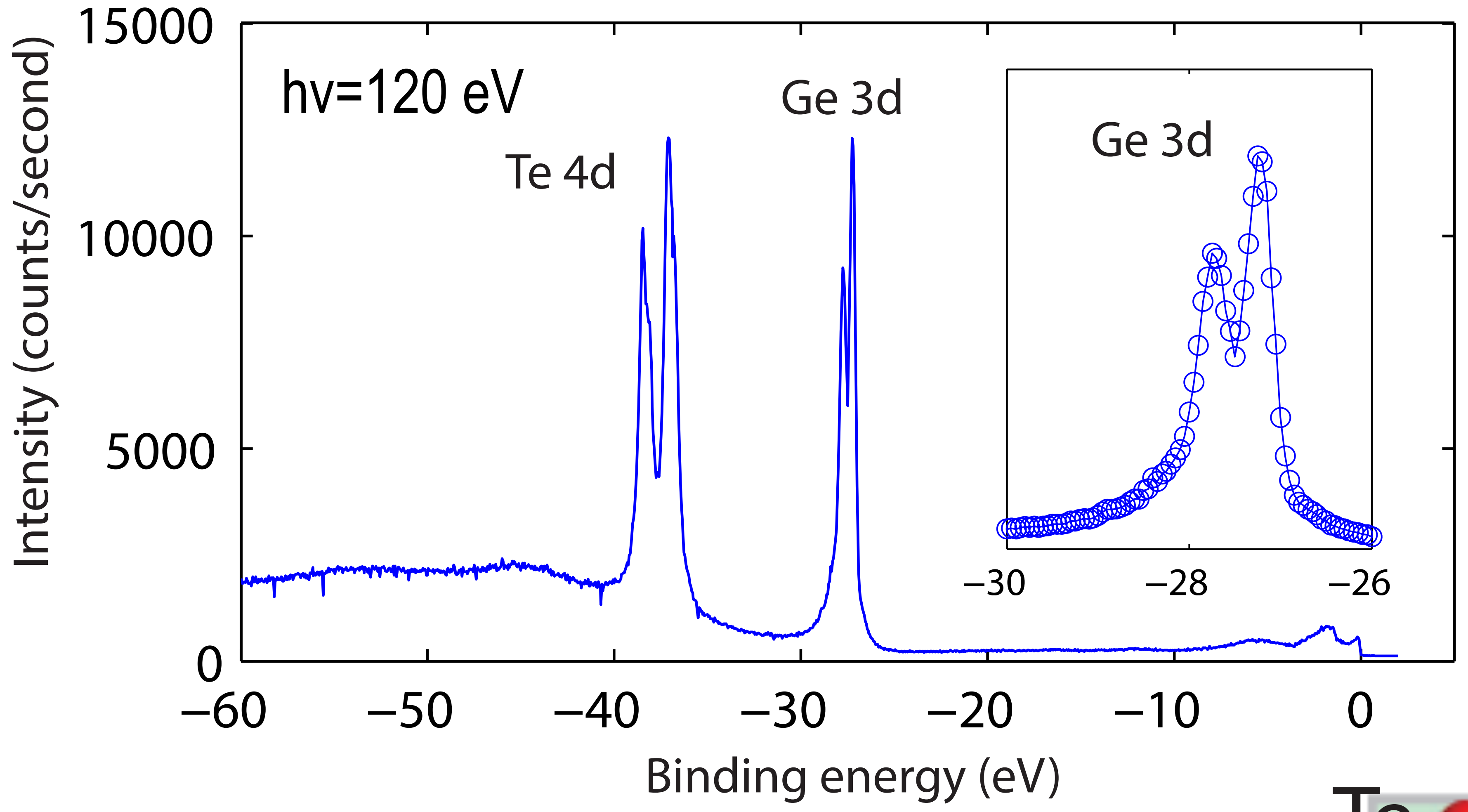


	$1s_{1/2}$	$2s_{1/2}$	$2p_{1/2}$		$2p_{3/2}$	$3s_{1/2}$	$3p_{1/2}$
1 H	14						
2 He	25						
3 Li	55						
4 Be	111						
5 B	188						
6 C	284						
7 N	399						
8 O	532	24					
9 F	686	31					
10 Ne	867	45					
11 Na	1 072	63		31			
12 Mg	1 305	89		52			
13 Al		118	74		73		
14 Si		149	100		99		
15 P		189	136		135		
16 S		229	165		164		
17 Cl		270	202		200		
18 A		320	247		245	25	
19 K		377	297		294	34	
20 Ca		438	350		347	44	26
21 Sc		500	407		402	54	32
22 Ti		564	461		455	59	34
23 V		628	520		513	66	38
24 Cr		695	584		575	74	43
25 Mn		769	652		641	84	49

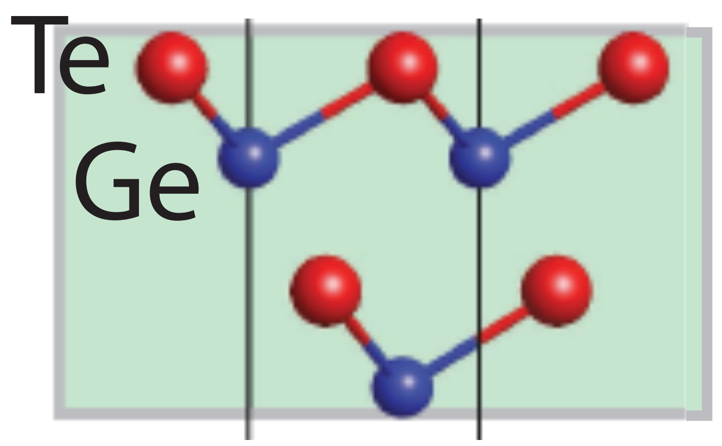
Quantitative aspects: Atomic cross section

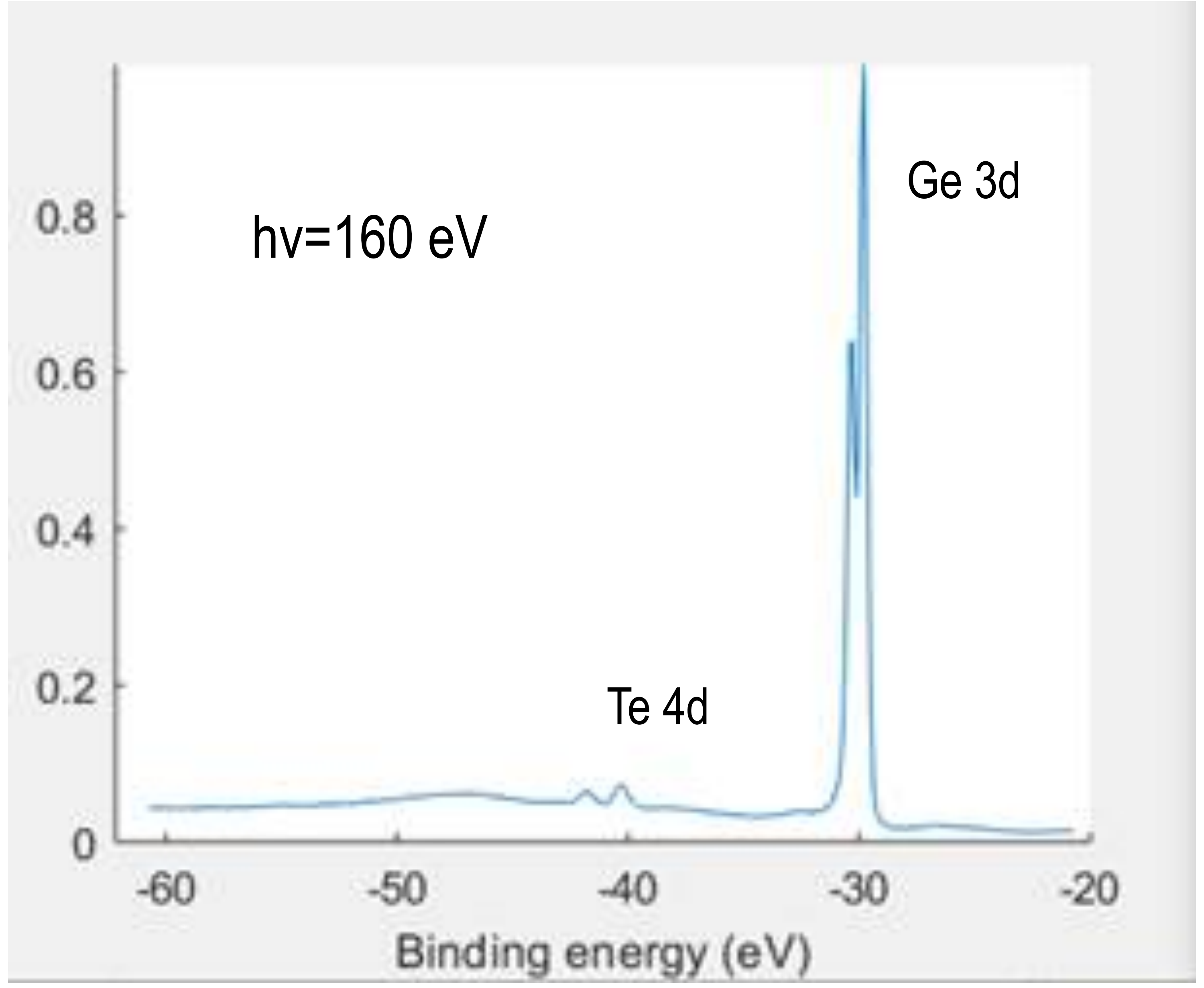
Intensity depends on:

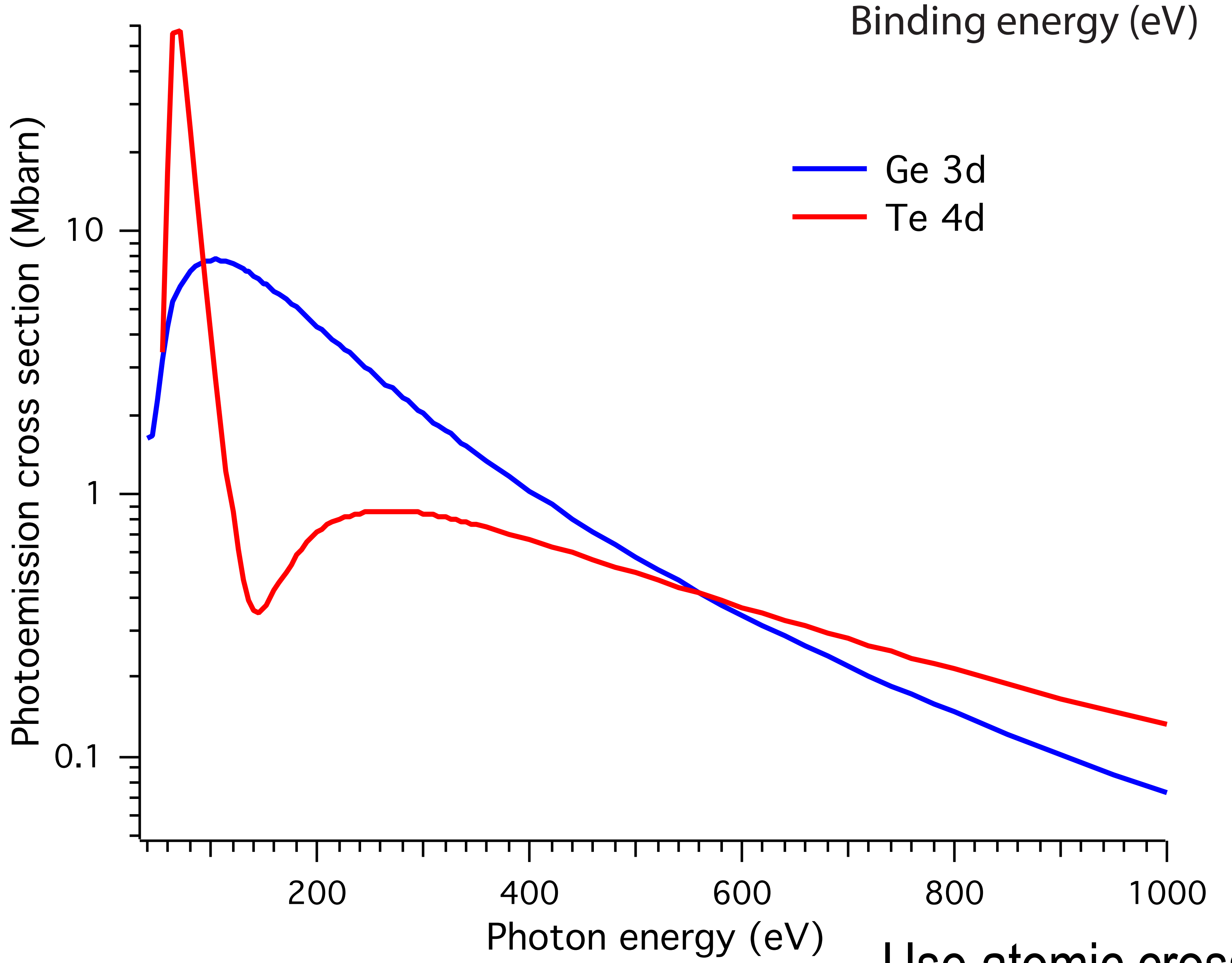
- amount of material
- atomic cross section
- distance from surface
- emission angle

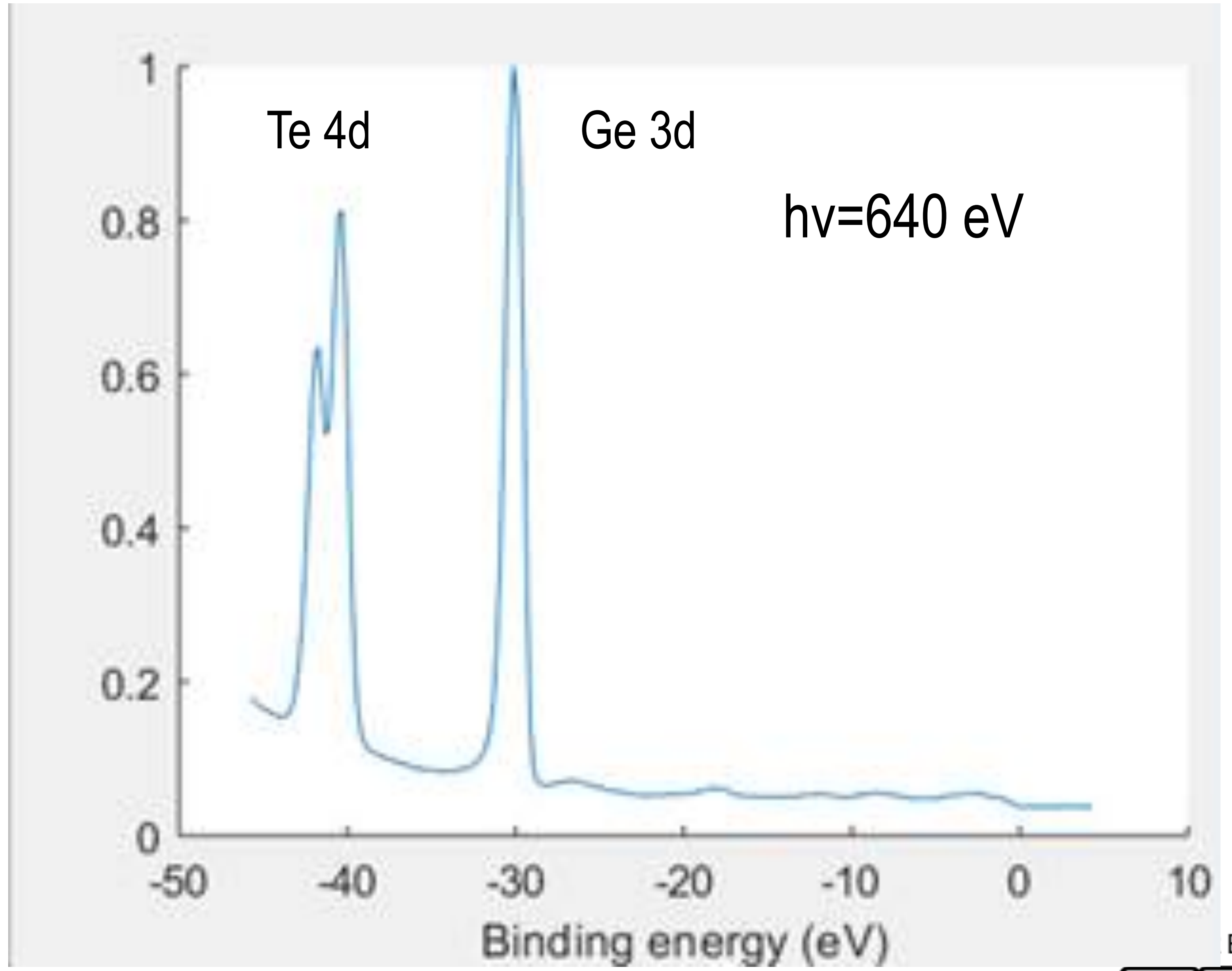


GeTe core levels









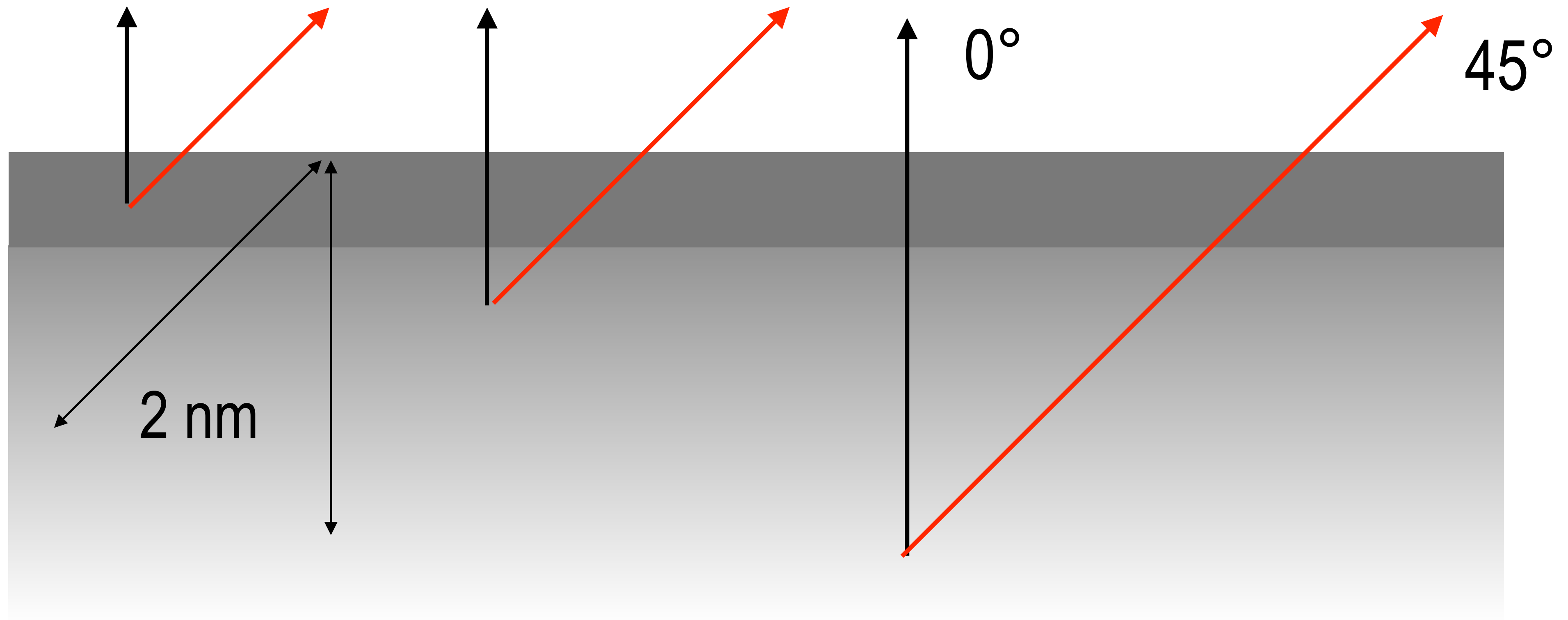
Use atomic cross sections for quantitative results (https://vuo.elettra.eu/services/elements/WebElements.html)

Quantitative aspects: Dependency on emission angle

Distance travelled in material increases rapidly with angle

Electron mean free path ~2 nm

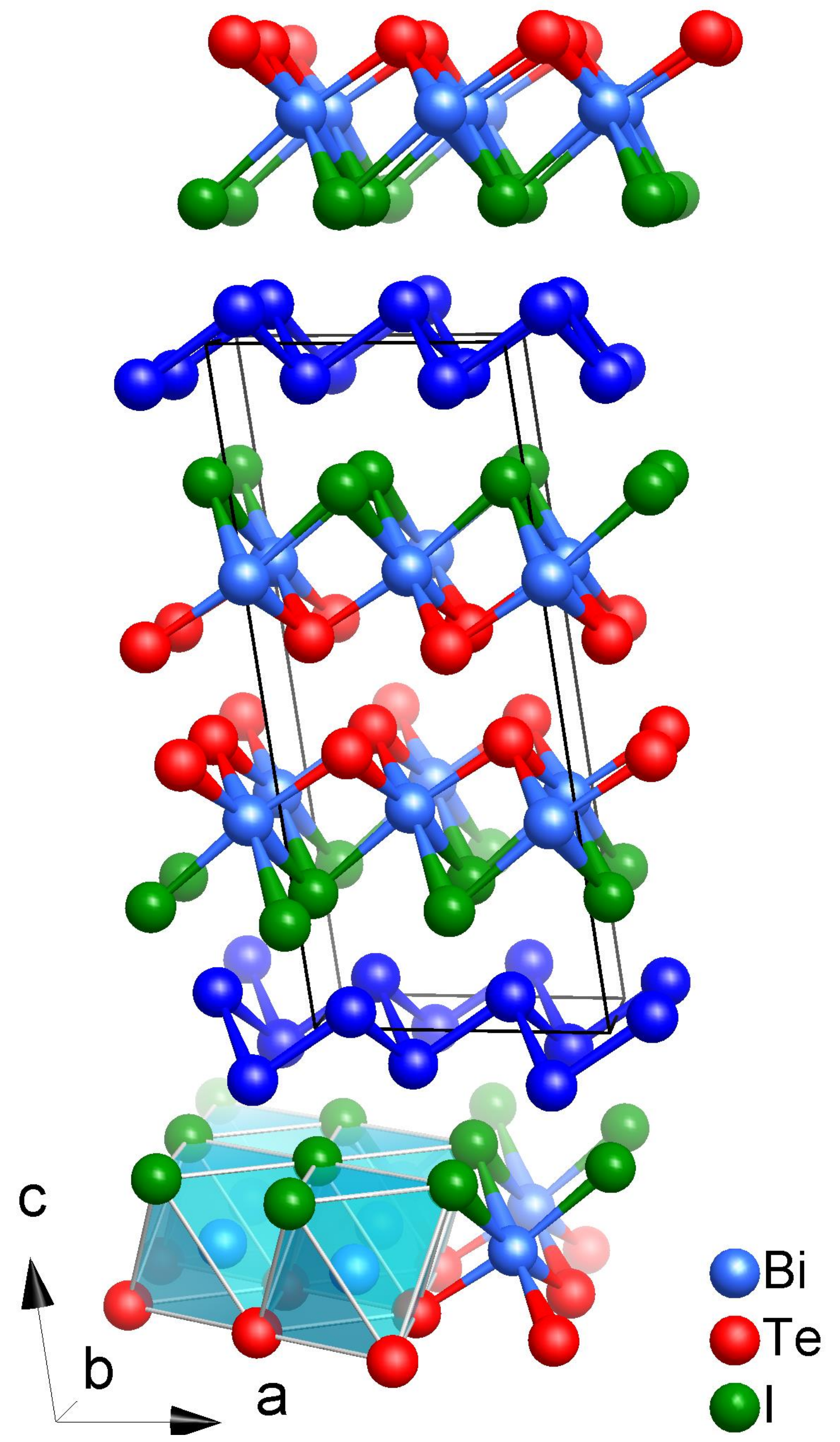
Determine chemical composition of top few atomic layers (surface termination)



Photon penetration depth not relevant here

Bi_2TeI

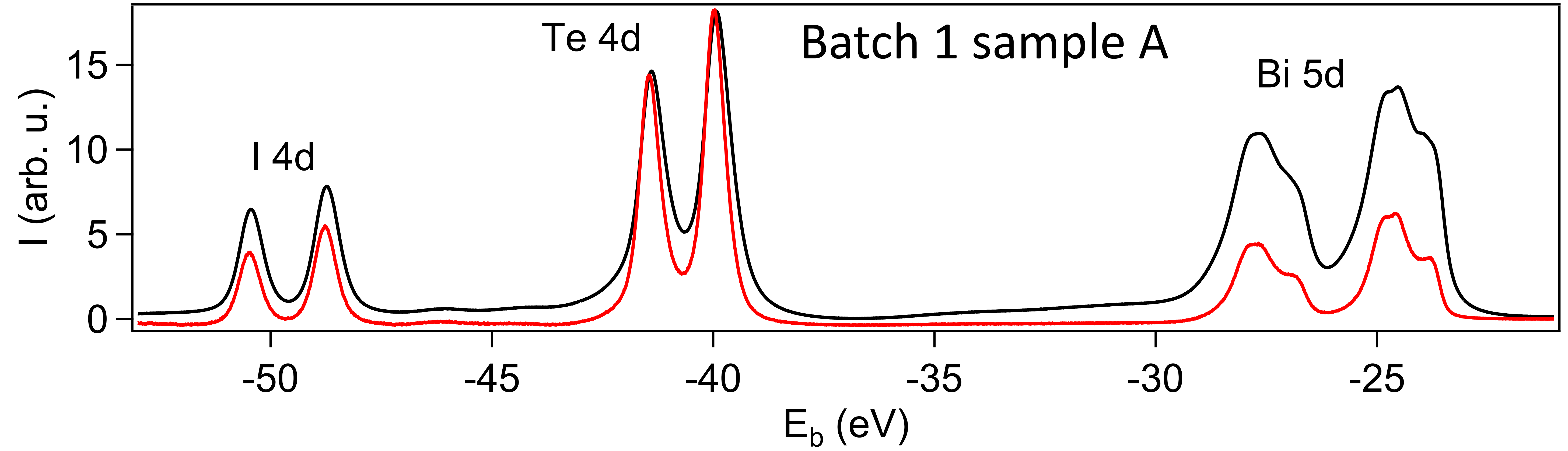
What termination?



0°

45°

Normalised to Te



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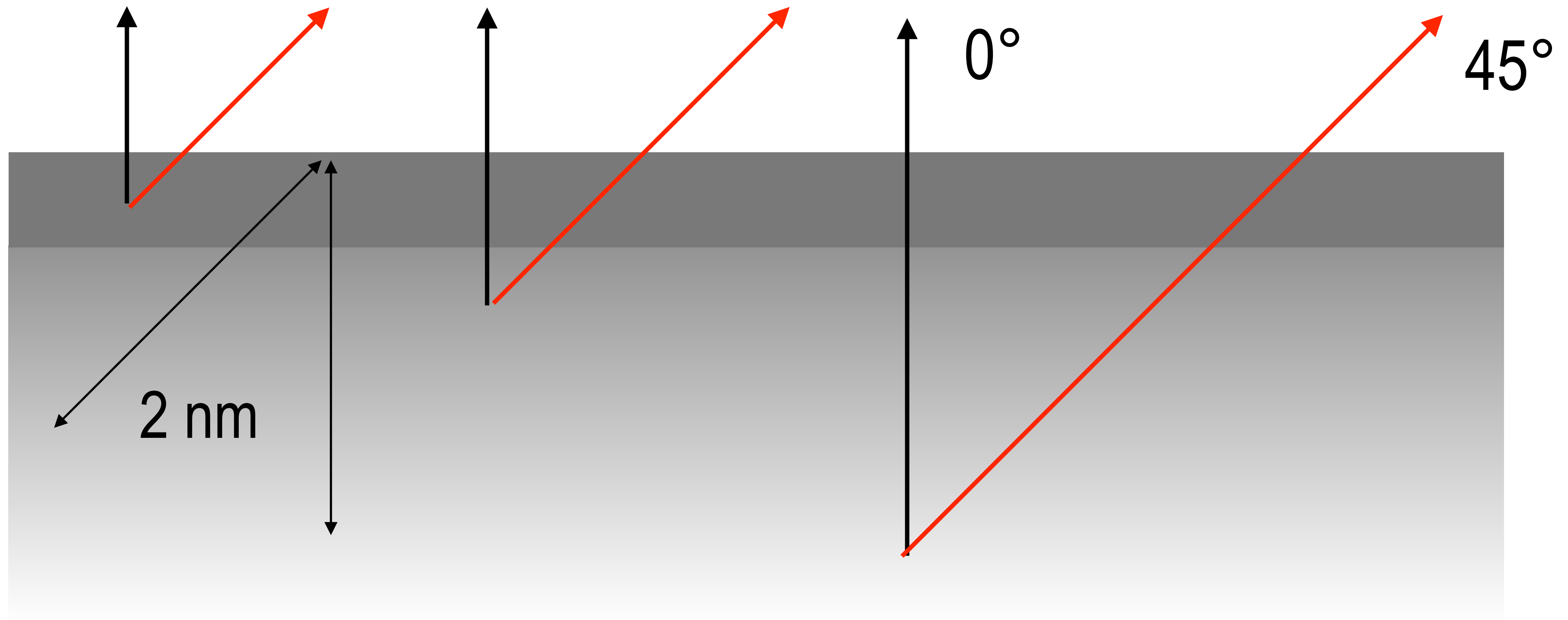

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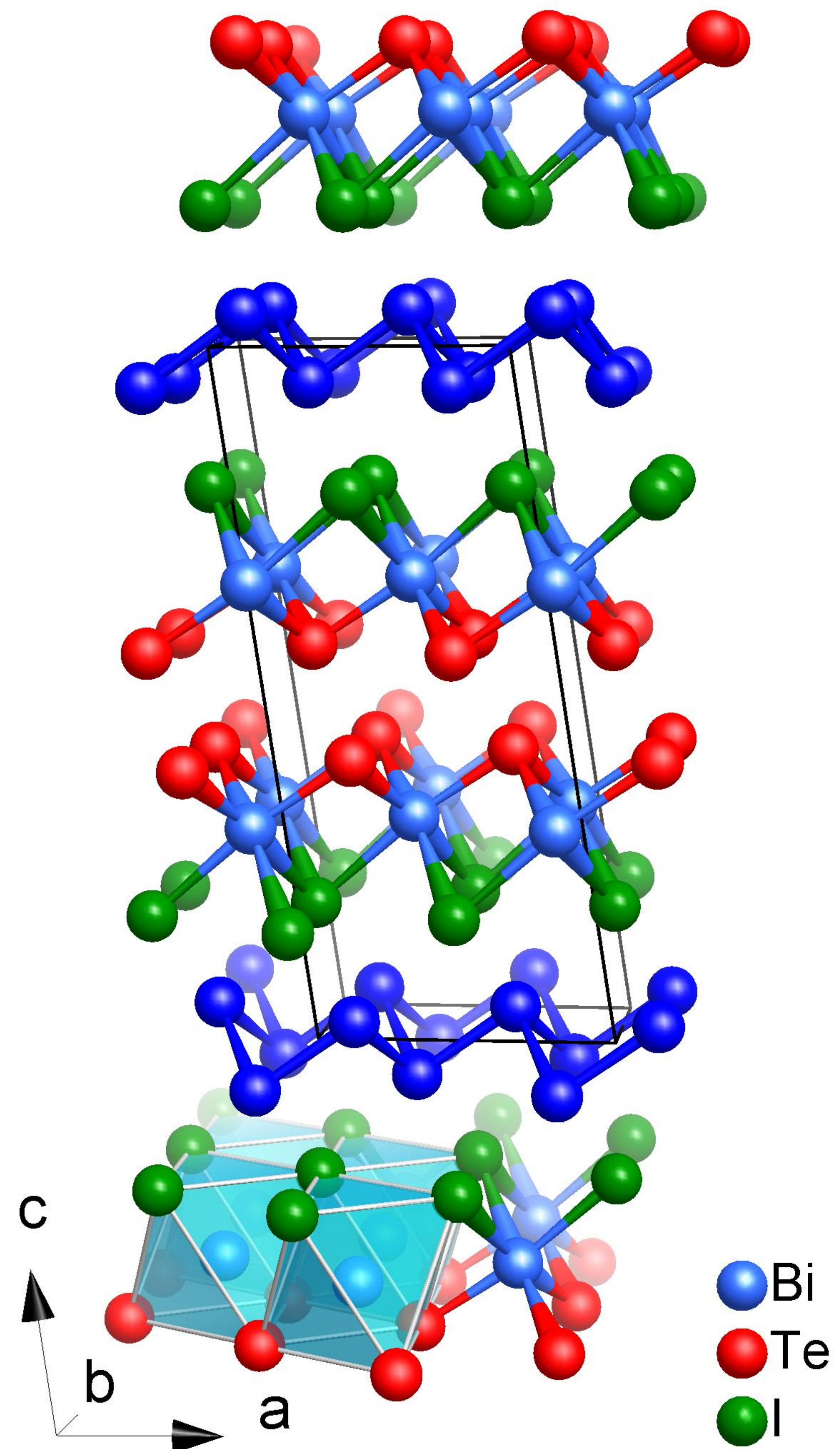
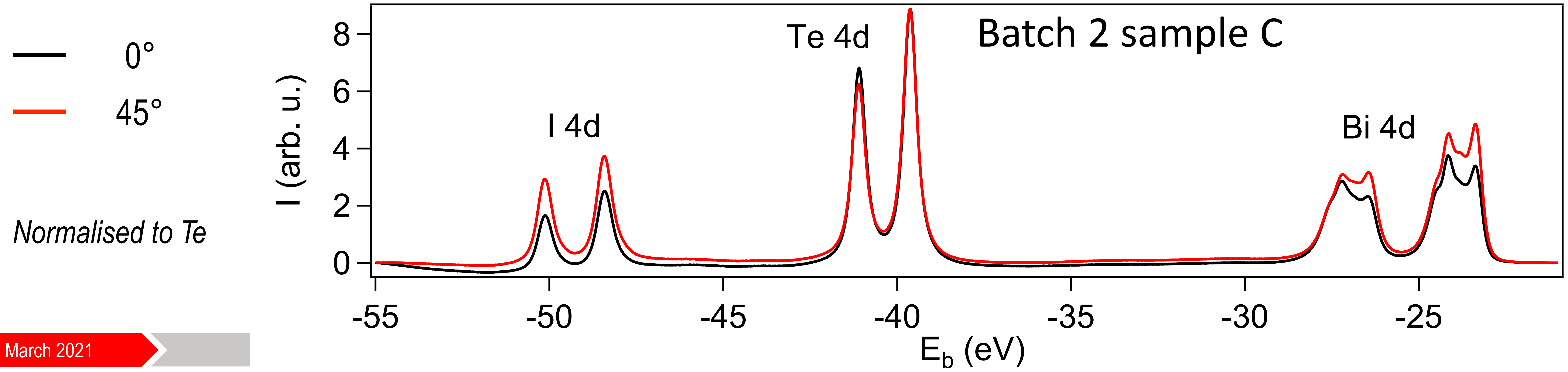
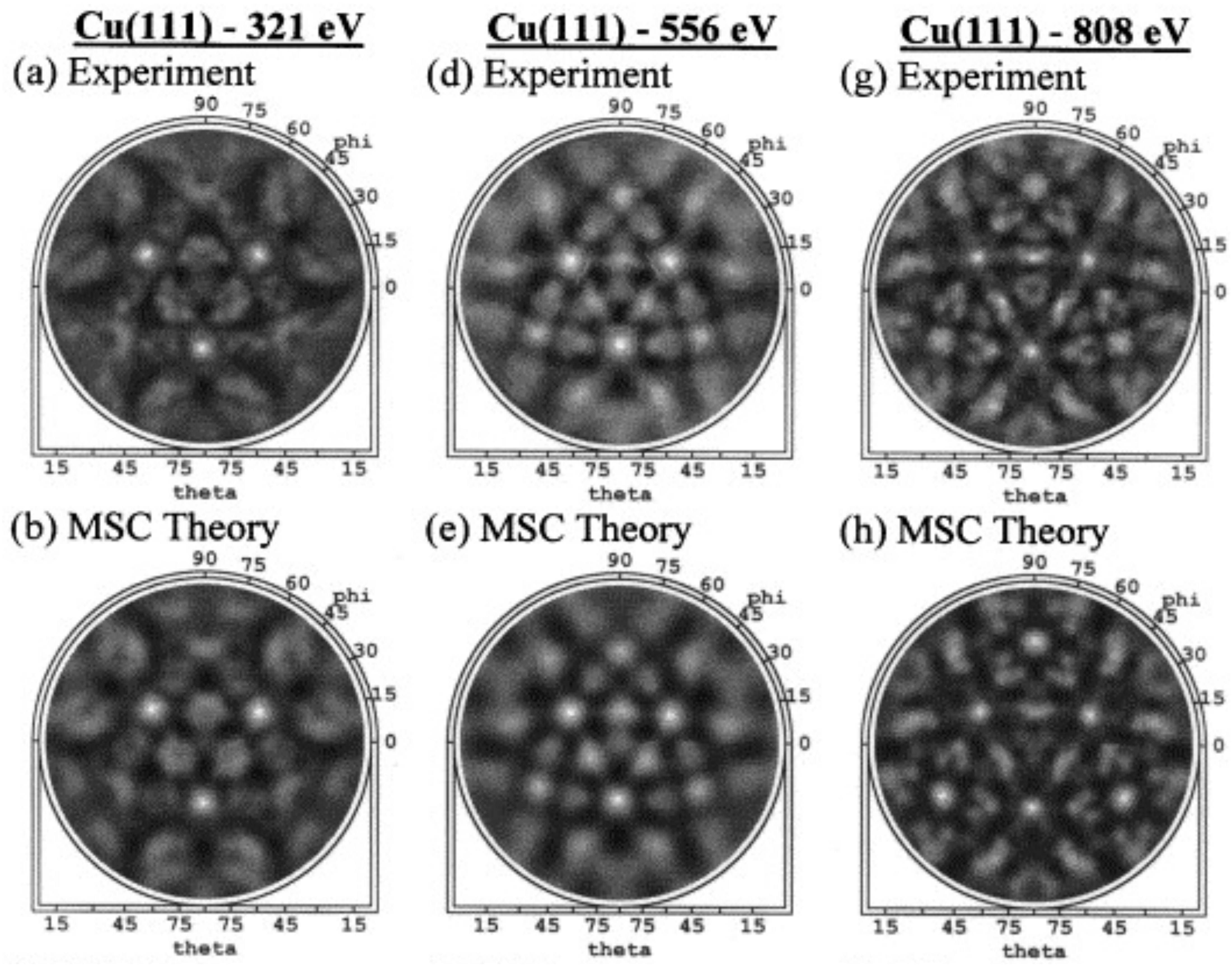




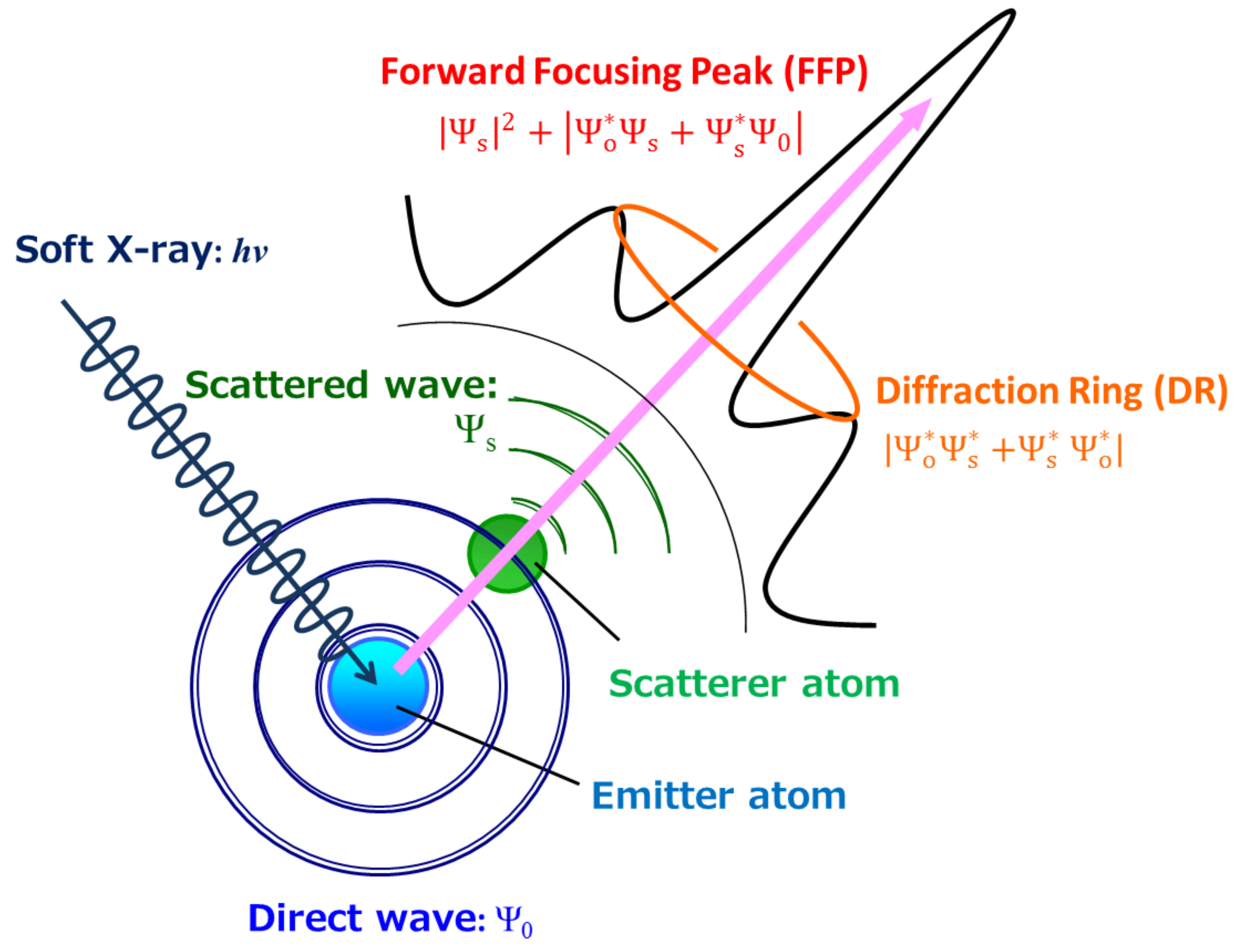
X-ray photoelectron diffraction (XPD)

Matsui, Nara Institute of Science and Technology

Combine XPS and fine angular resolution



S.D. Ruebush et al. Surface Science 421, 205 (1999)



Scattering of photoemitted electron on local environment

Structural determination of surfaces, overlayers, and adsorbates

Chemical and sub-monolayer sensitivity

Recent developments in holography and dichroic imaging

Review: D.P. Woodruff Surface Science Reports 62, 1 (2007)

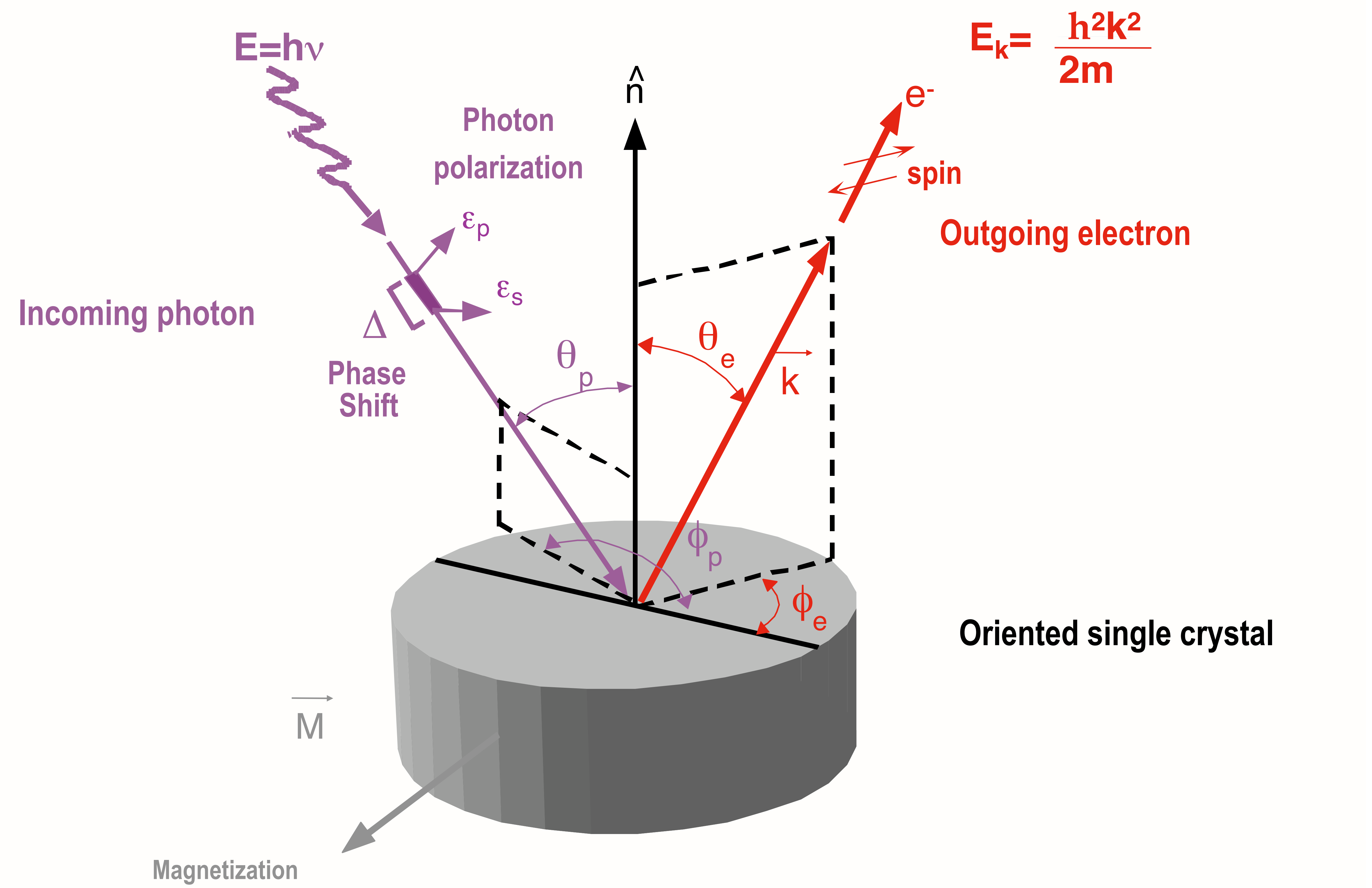
Recent developments: M.V. Kuznetsov et al. J. Phys. Soc. Jpn. 87, 061005 (2018)

Angle-resolved photoemission spectroscopy (ARPES)

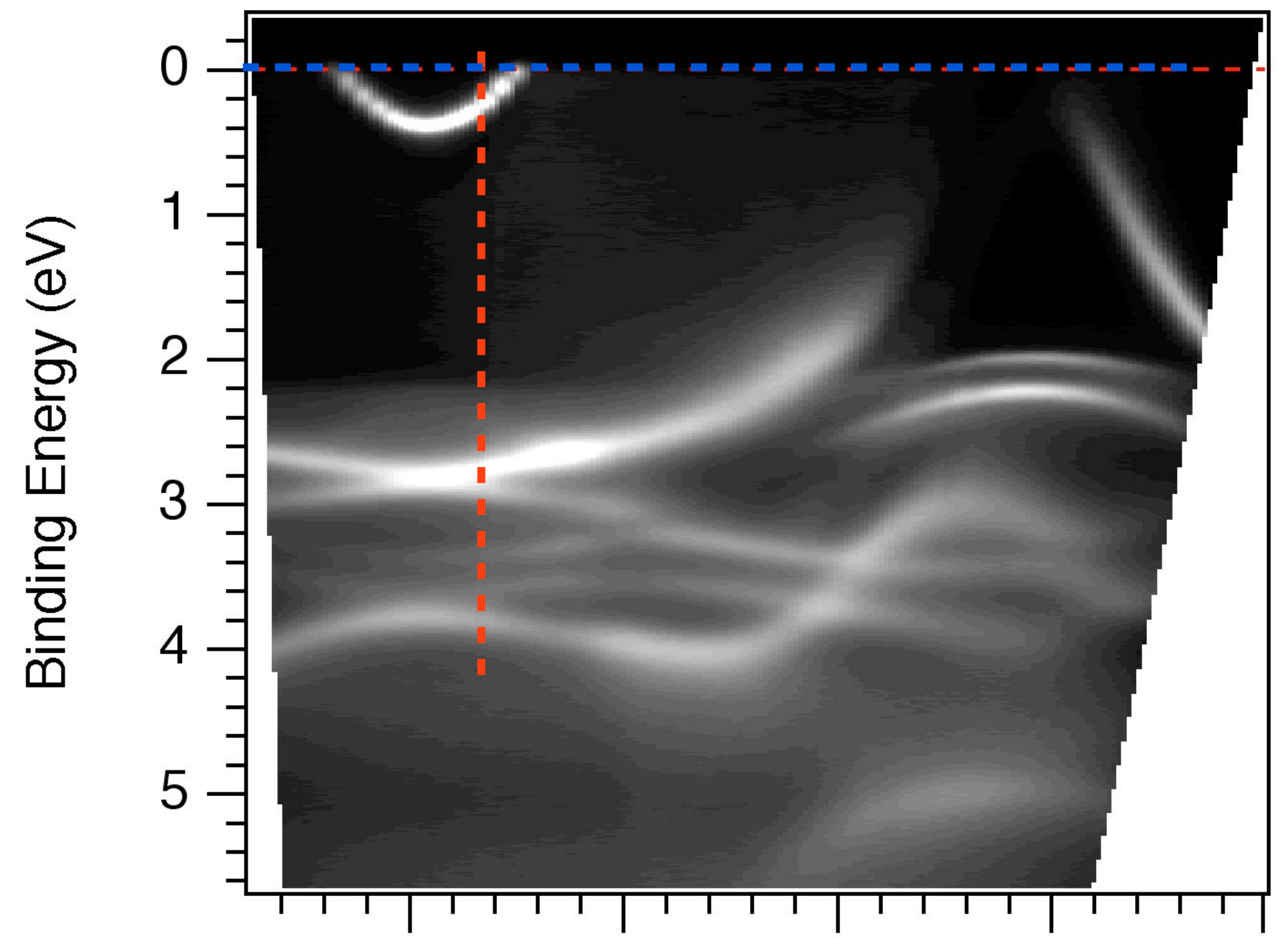
Measurement:

1) **Kinetic energy → Binding energy** $E_b = h\nu - \Phi - E_k$
2) **Emission angle → momentum k** $k_{||} = 0.512\sqrt{E_k}\sin\theta$
3) Asymmetry → Spin polarization vector

Change photon energy to acces 3D momentum



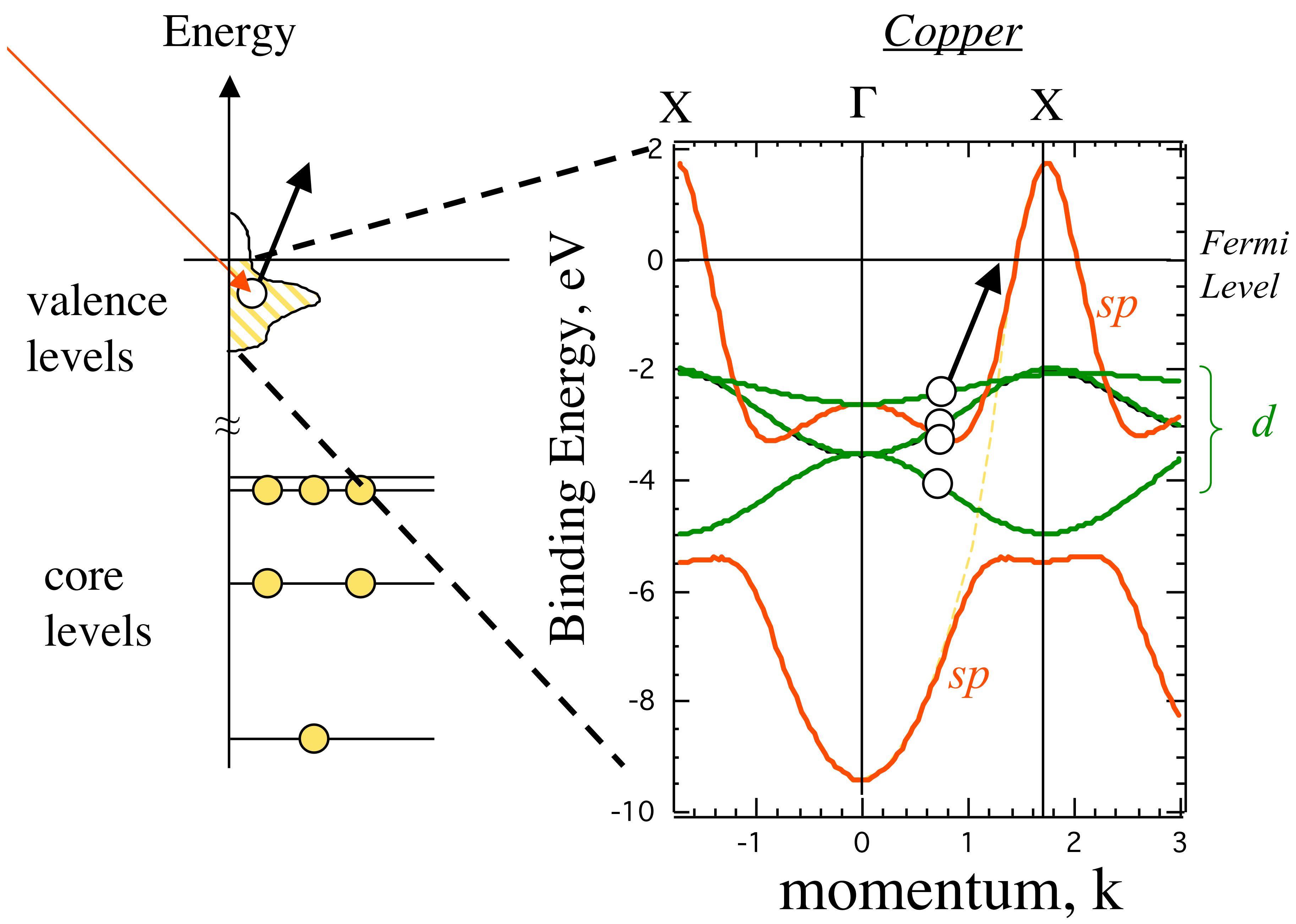
Cu(111)



Band structure

EDC:
energy distribution curve

MDC:
mometum distribution curve



"See how the electrons move in the crystal"

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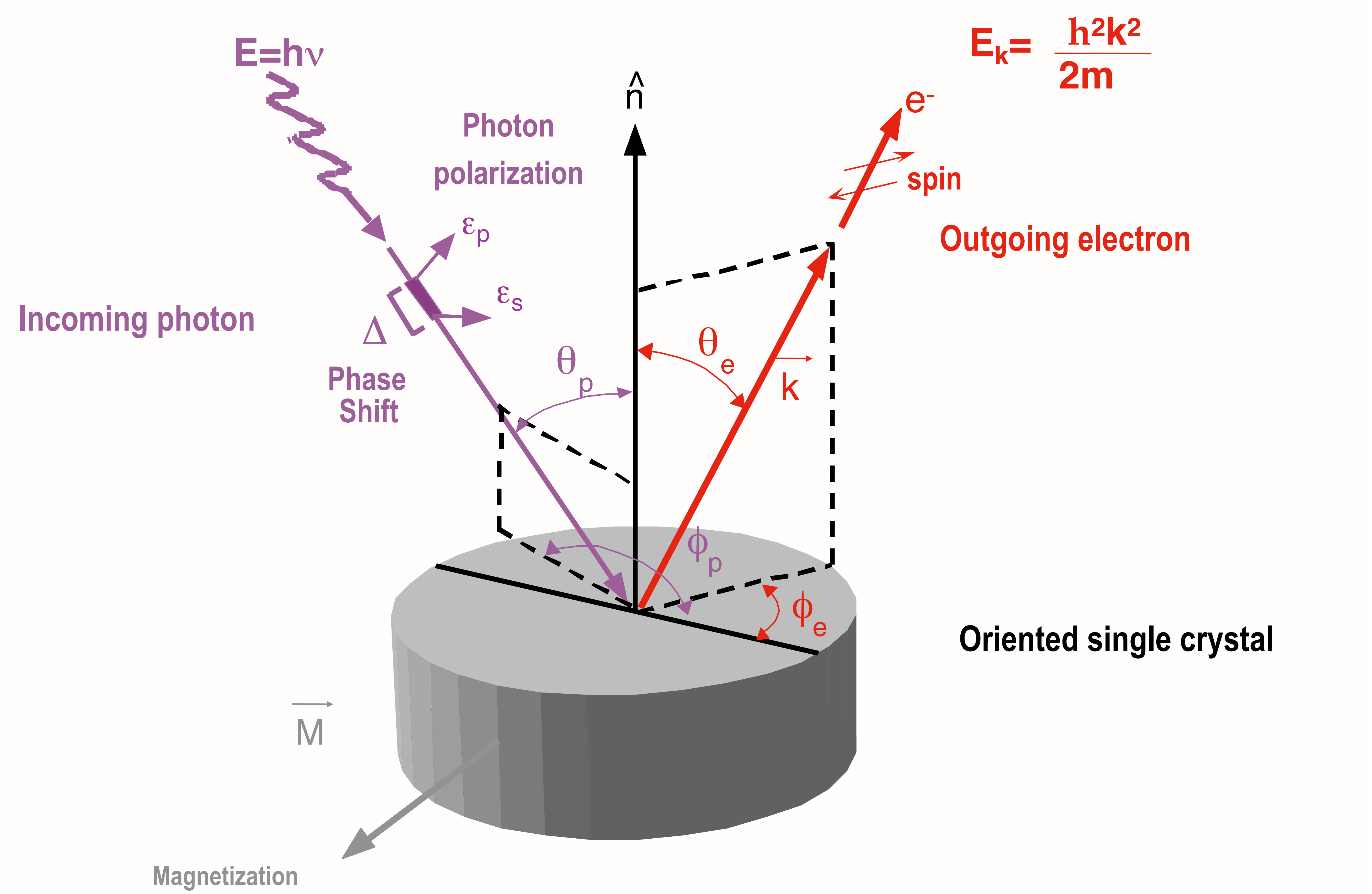

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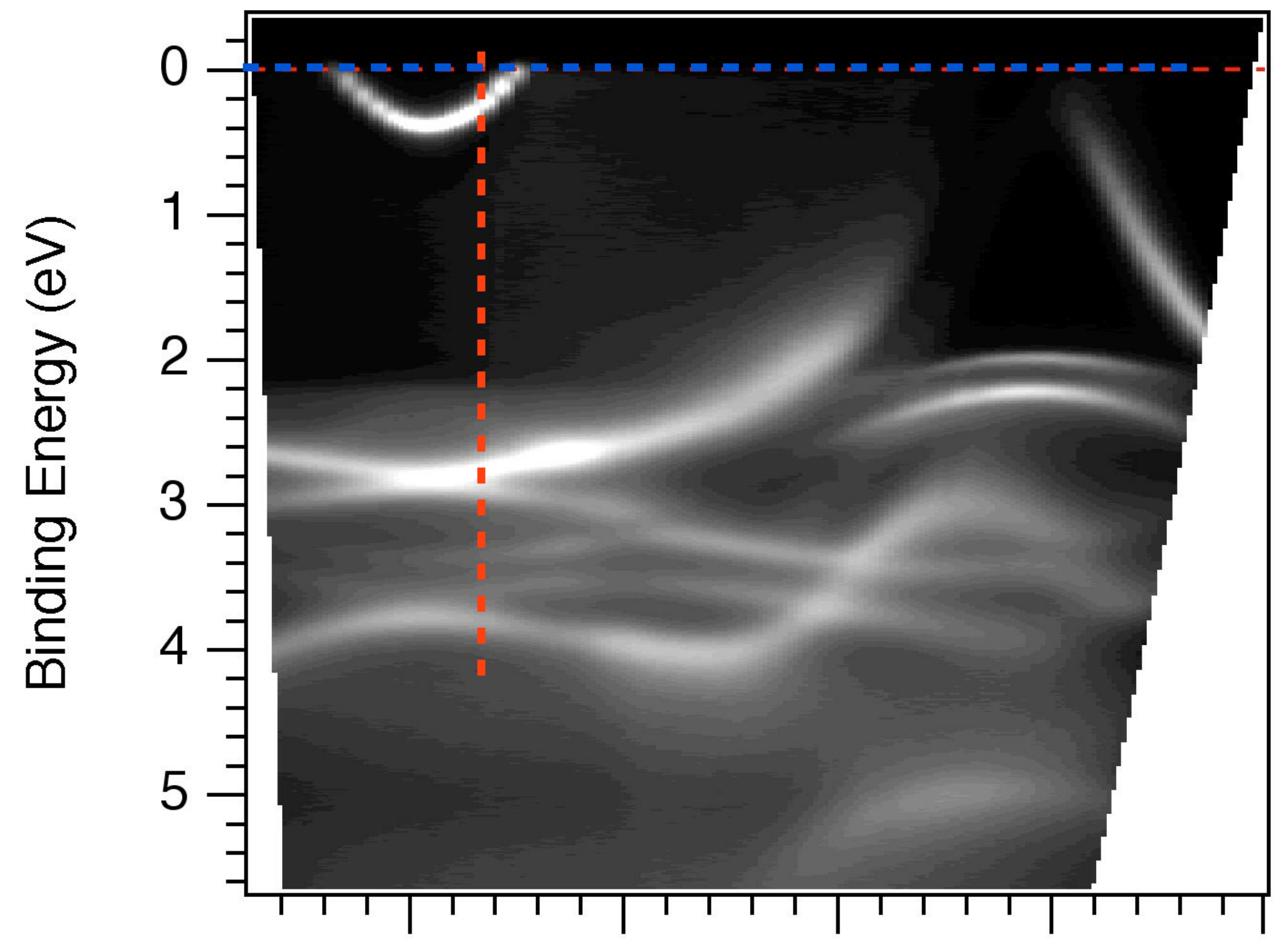

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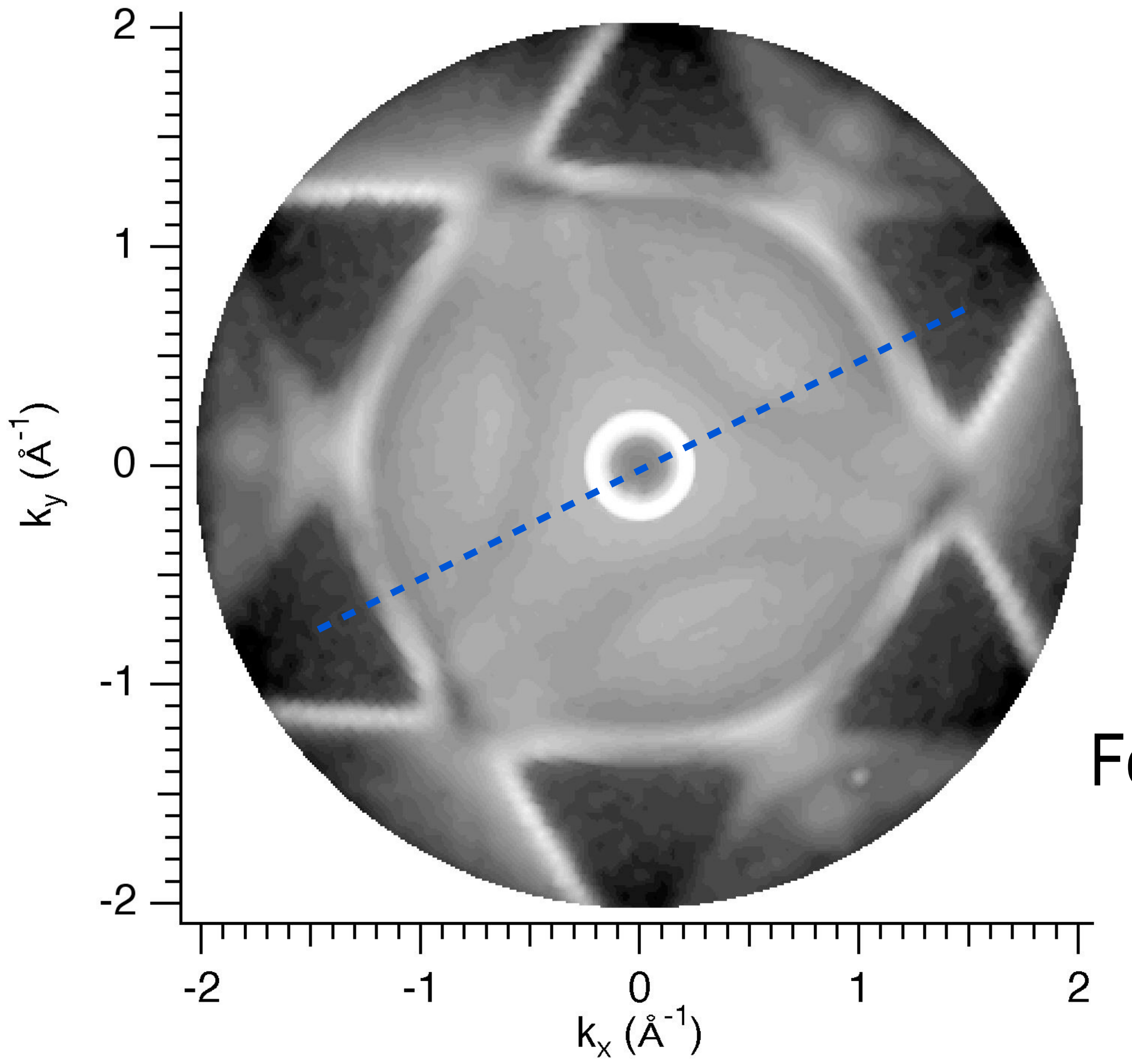


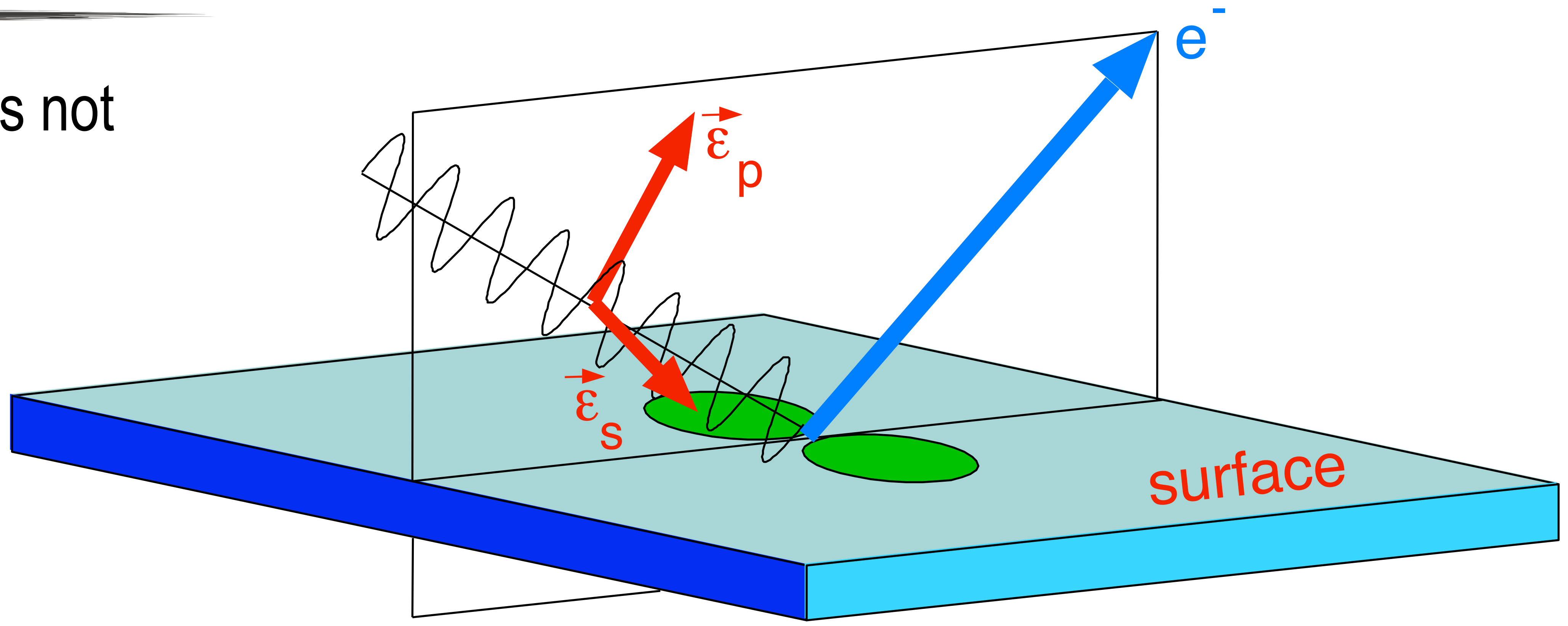
Fermi surface

"See how the electrons move in the crystal"

Quantitative aspect: Matrix element effects

For accessible extensive description:
S. Moser J. Electron Spectroscopy and Related Phenomena 214, 29 (2017)

Relies on **sudden approximation**: photoelectron emitted from N particle system does not interact with N-1 system *after* photo-excitation



$\vec{\varepsilon}_p$ photon polarization in...
$\vec{\varepsilon}_s$ ...perpendicular ("senkrecht") to...scattering plane

Fermi's Golden Rule for transition probability:

$$w = \frac{2\pi}{\hbar}|\langle\Psi_f|H_{int}|\Psi_i\rangle|^2\delta(E_f - E_i - \hbar\omega)$$

For dipole allowed transitions:

$$H_{int} = \frac{e}{mc}A \bullet p$$

Dipole Selection Rules:

$\Delta l = \pm 1;$
$\Delta m = 0, \pm 1.$

$$I \propto |\langle\Psi_f|A \bullet p|\Psi_i\rangle|^2$$

Measured intensity: I; Final state: Ψ_f; Initial state: Ψ_i; Photon E field: A; Electron momentum: p

$$|\psi_f\rangle \quad A \bullet p \quad |\psi_i\rangle$$

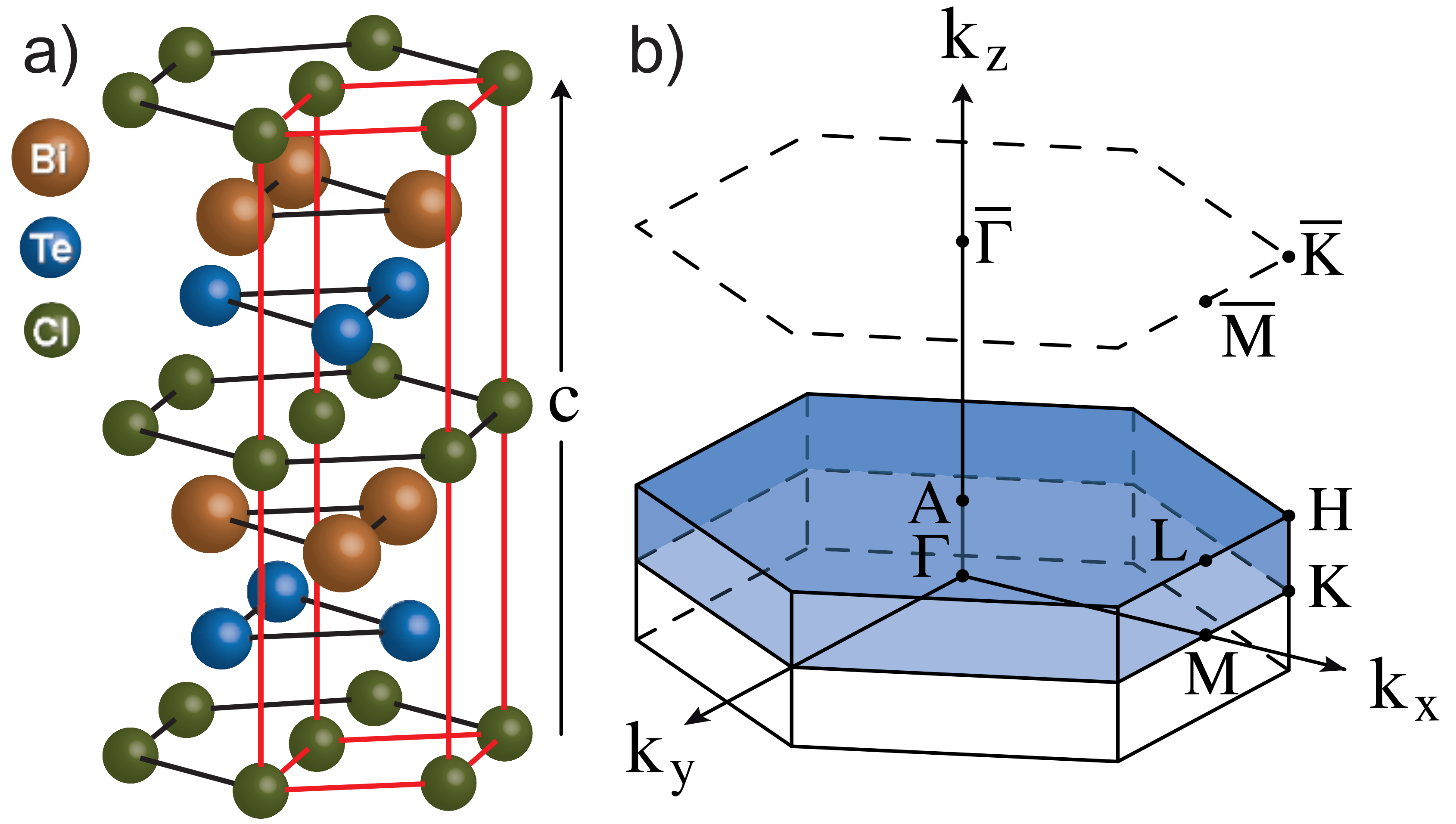
Plane wave always even ($|\psi_f\rangle$); Even for p-pol odd for s-pol ($A \bullet p$); Depends on band symmetry (s, $p_{x,y,z}$) ($|\psi_i\rangle$)

	ψ_f	$A \bullet p$	ψ_i	
p - pol.	+1	+1	-1	0
	+1	+1	+1	max.
s - pol.	+1	-1	-1	max.
	+1	-1	+1	0

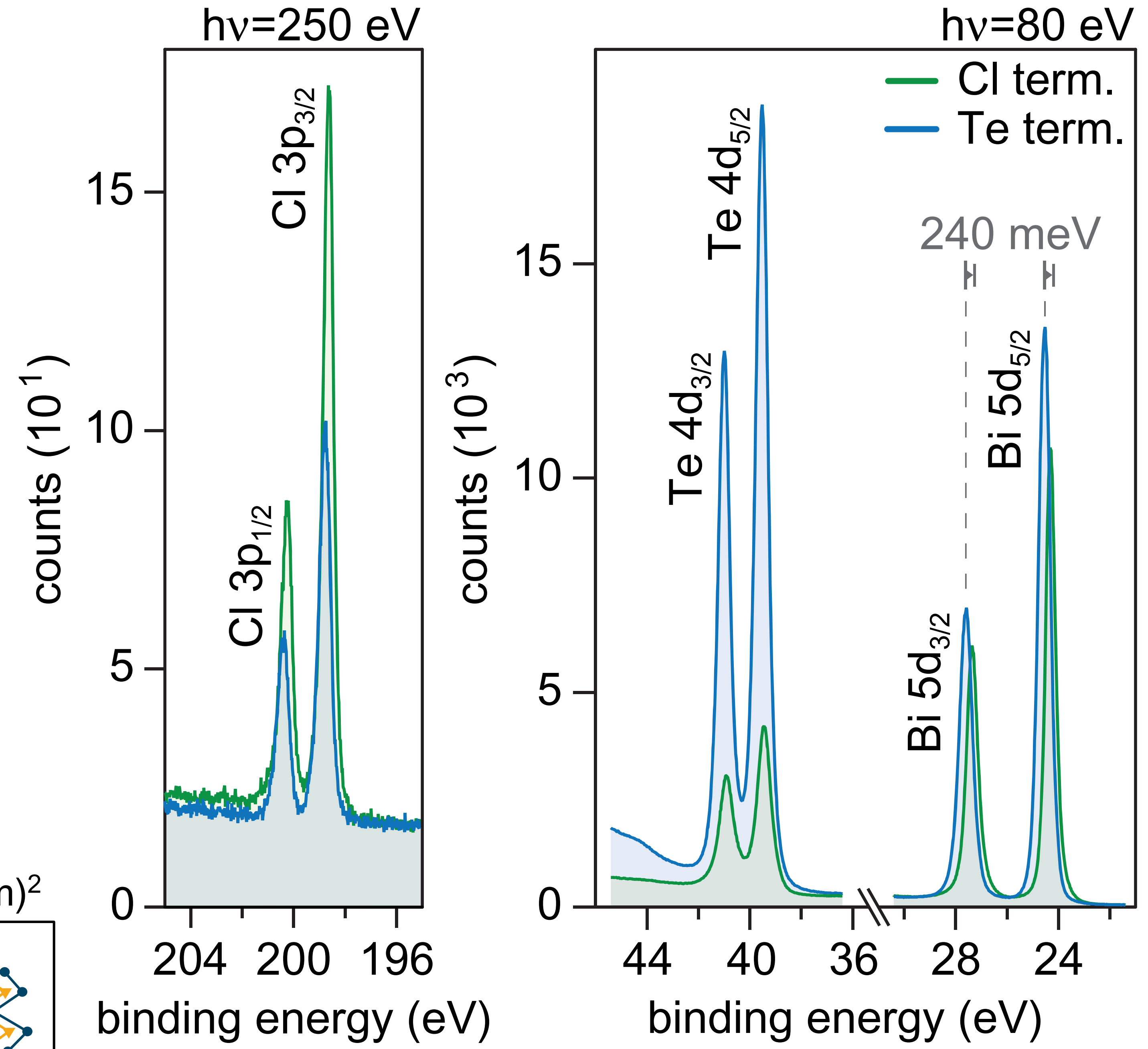
BiTeCl as example

G. Landolt et al. New Journal of Physics 15, 085022 (2013)

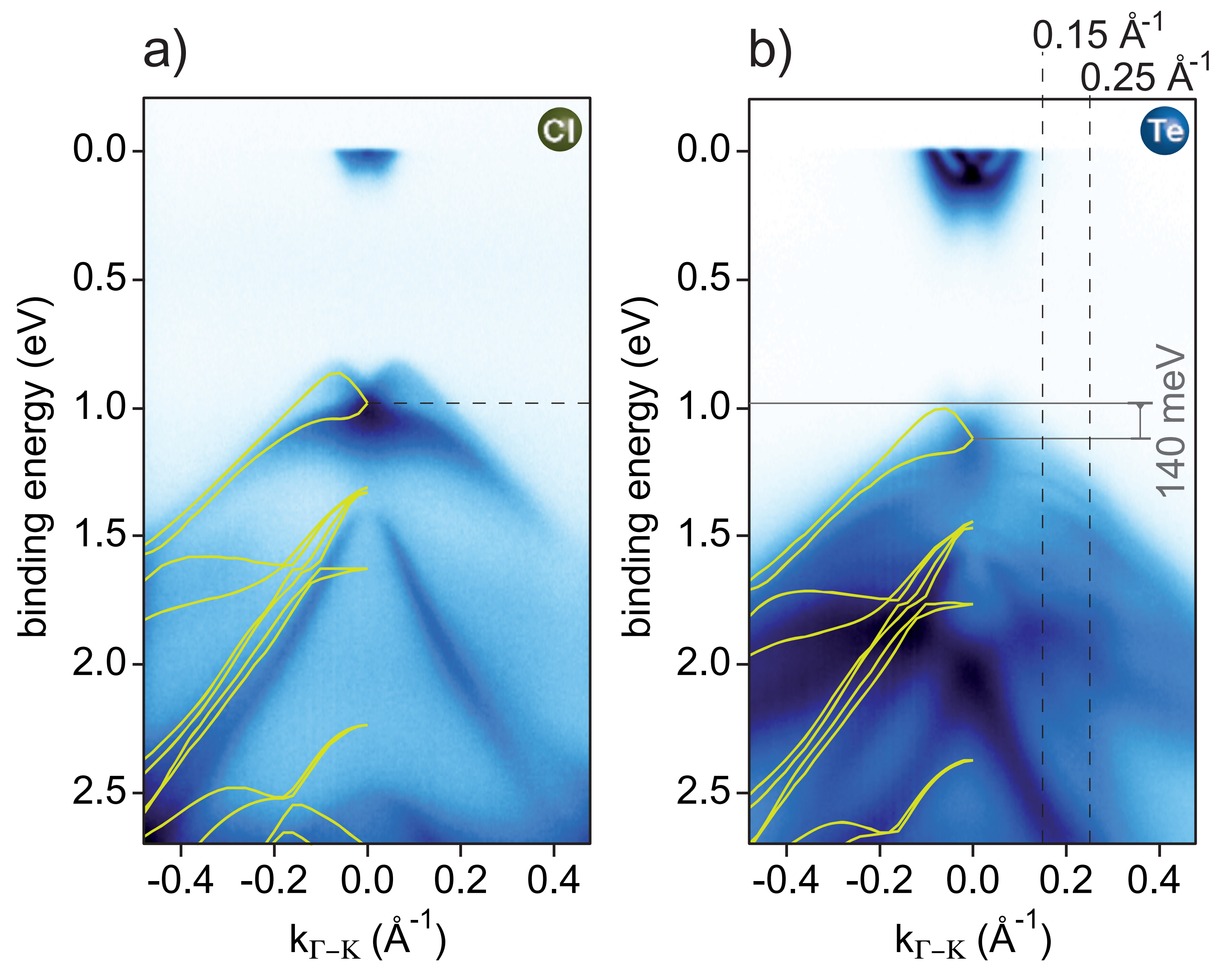
No inversion symmetry and non-symmorphic

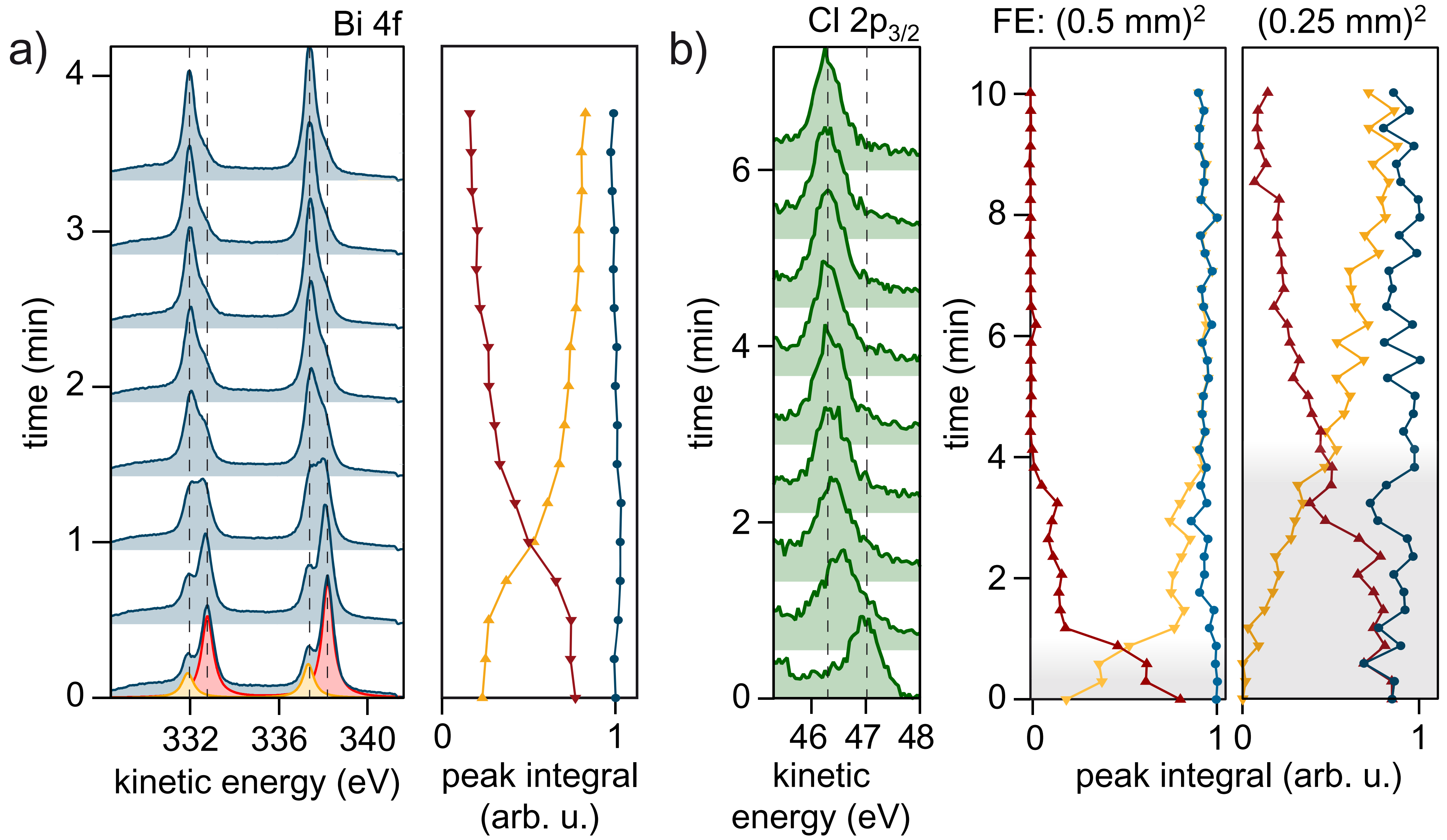


XPS to detrmine surface termination



ARPES to see shift in bands and match to DFT

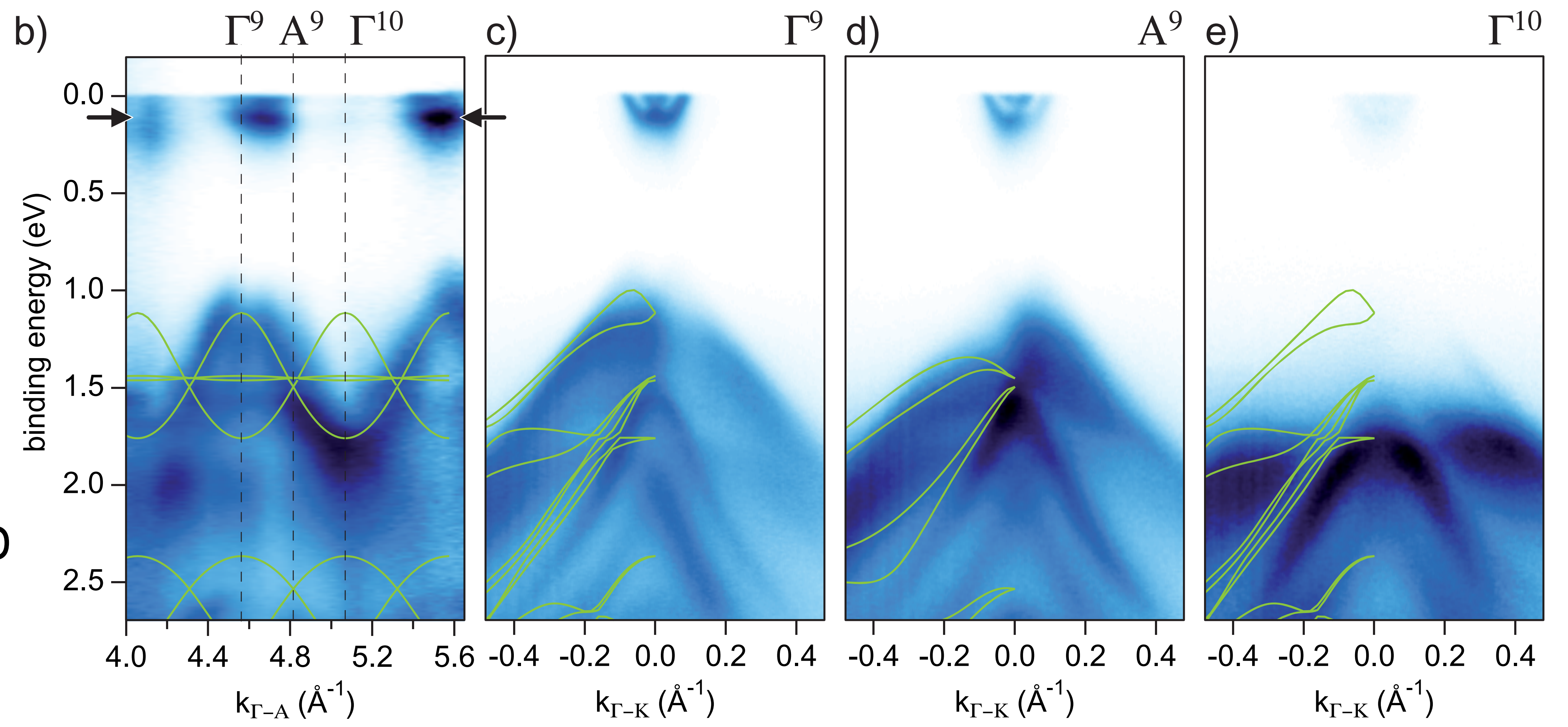




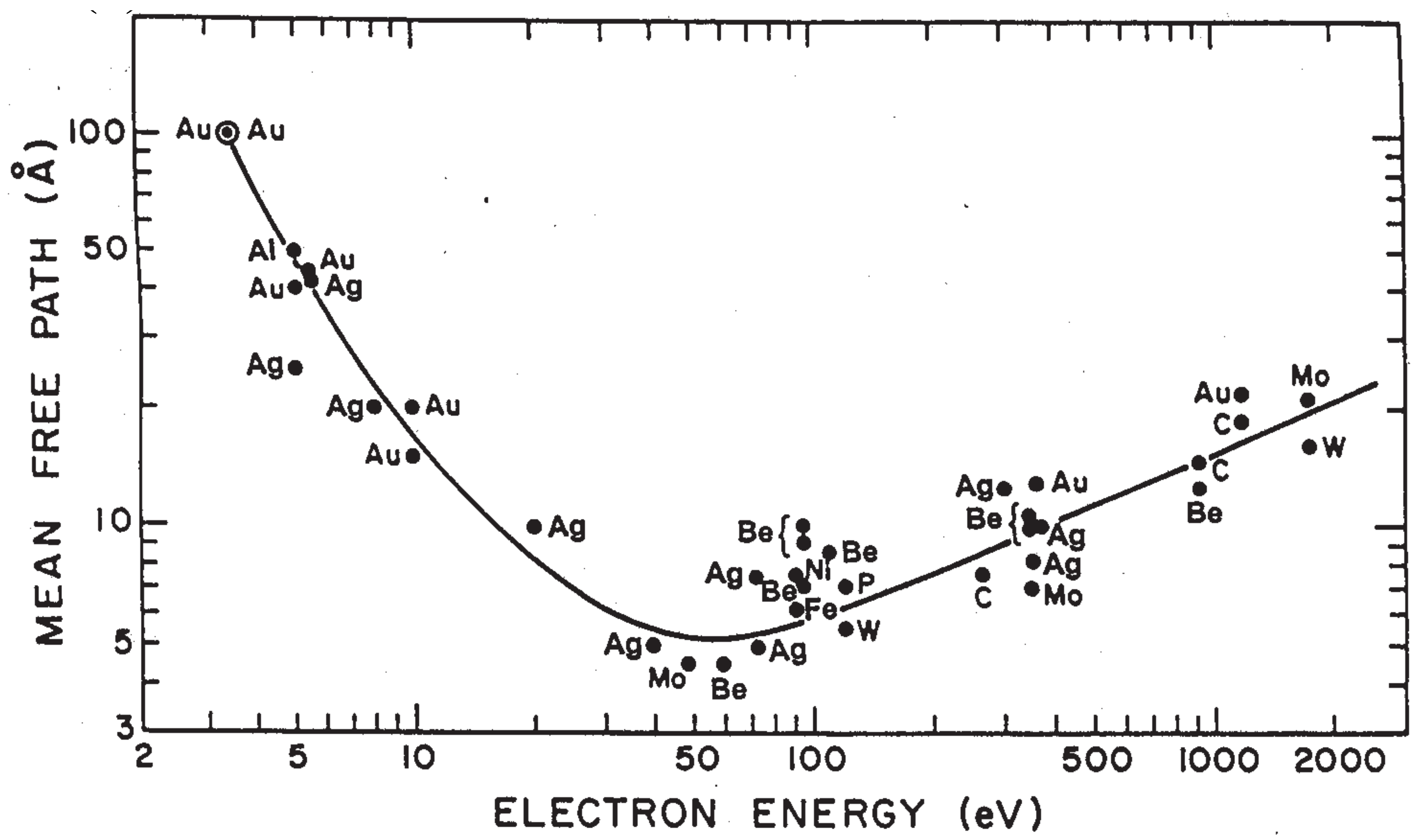
Photon induced chemistry destroys surface states on Cl-termination

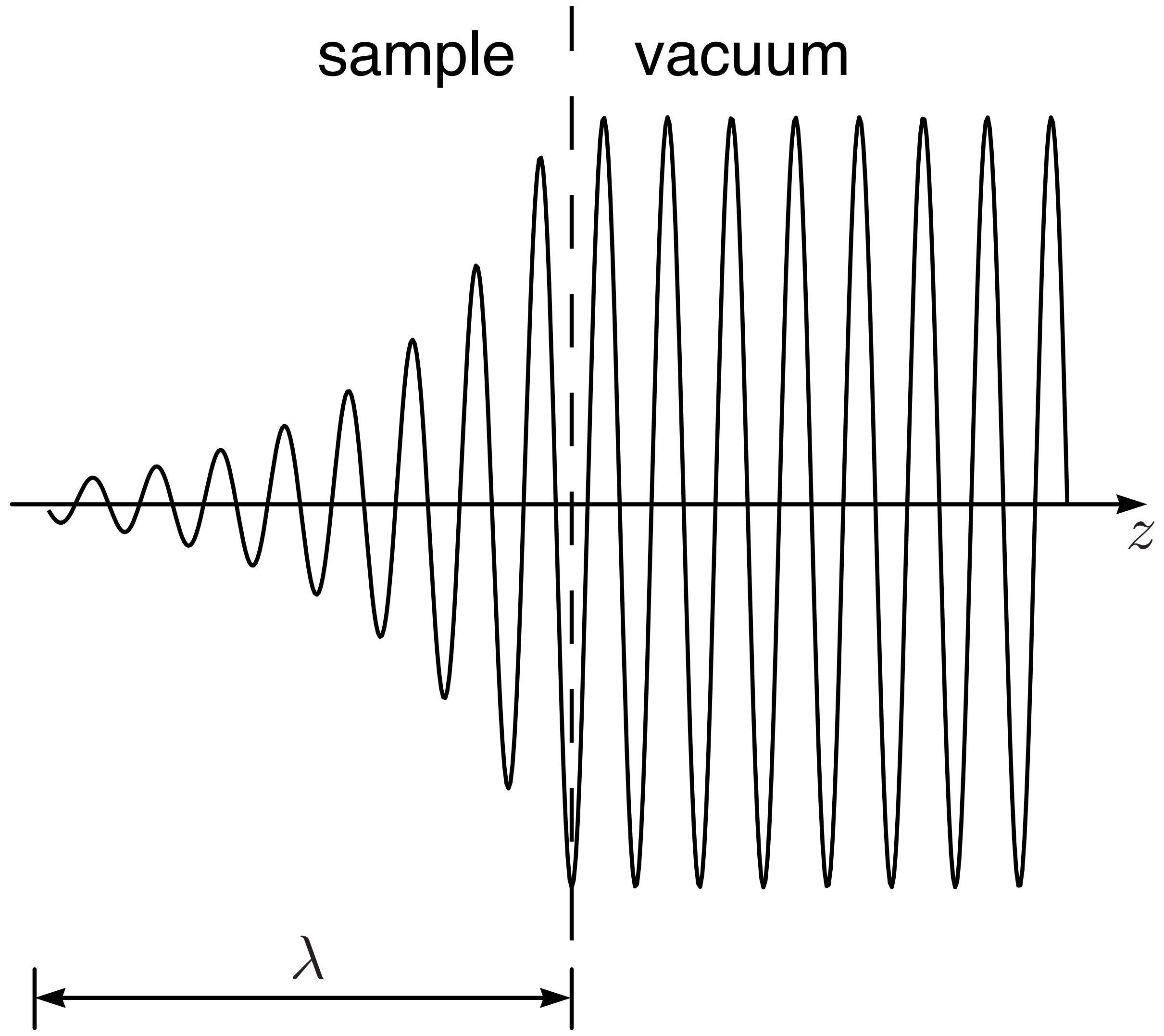
hv dependence to distinguish surface and bulk states (Te-termination)

Band visible only in every 2nd BZ due to interference

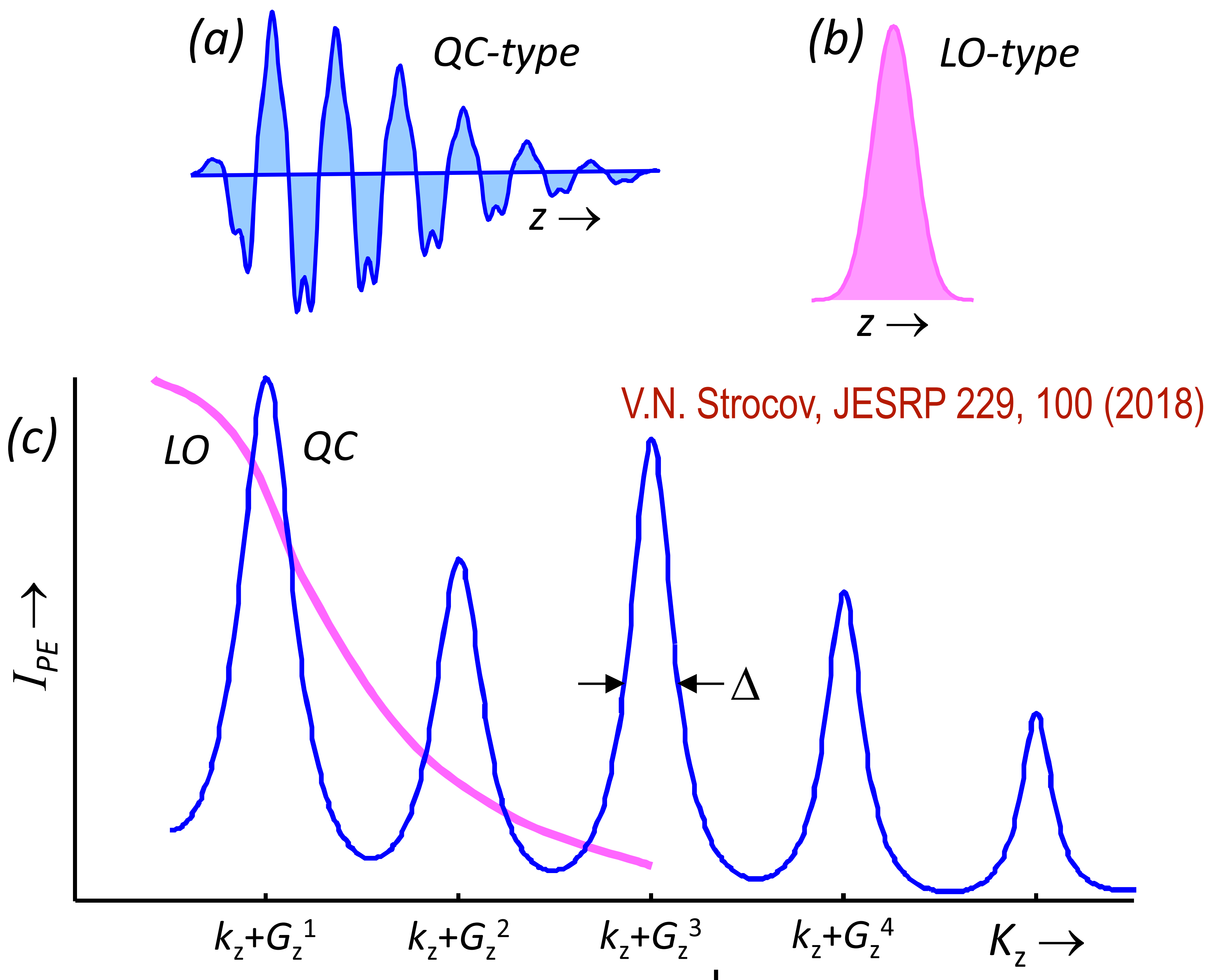


Some remarks on soft X-ray ARPES (SX-ARPES)





Initial state wave function extension



- Larger mean free path increases probing depth (main signal still from surface region)
- Increased damping distance enhances k_z resolution ($\Delta k_z=\lambda^{-1}$): large problem at low hv
- High kinetic energies mean final state feels less of crystal potential: more free electron-like

→ Simplified matrix element and "cleaner" spectrum

Price to pay: less good resolution (40meV vs. 4meV) and lower count rate

QC: quantum confinement

Shockley surface states

Quantum well states

…

LO: local orbital

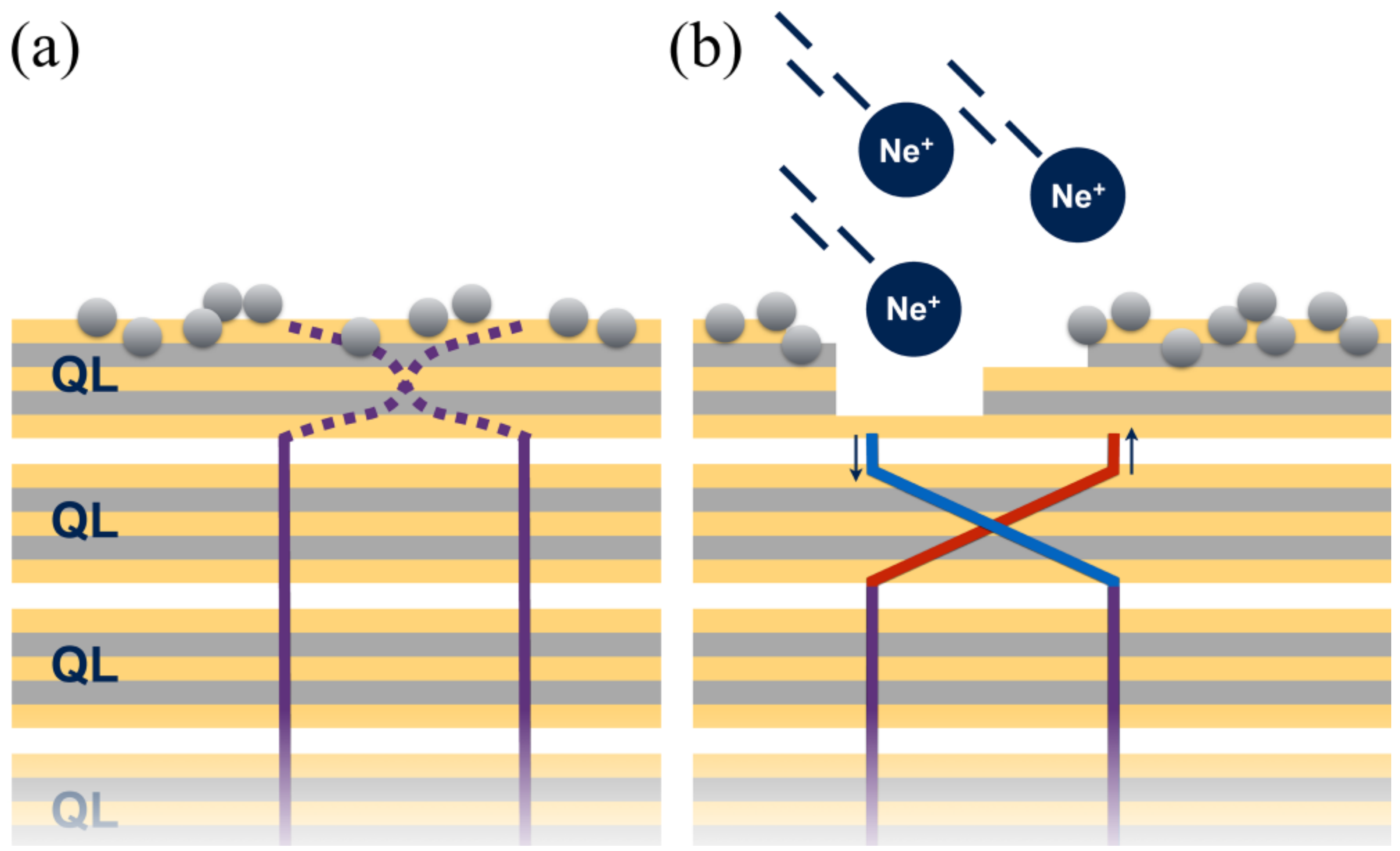
Absorbed molecules

Dangling bond states

Surface reconstructions

…

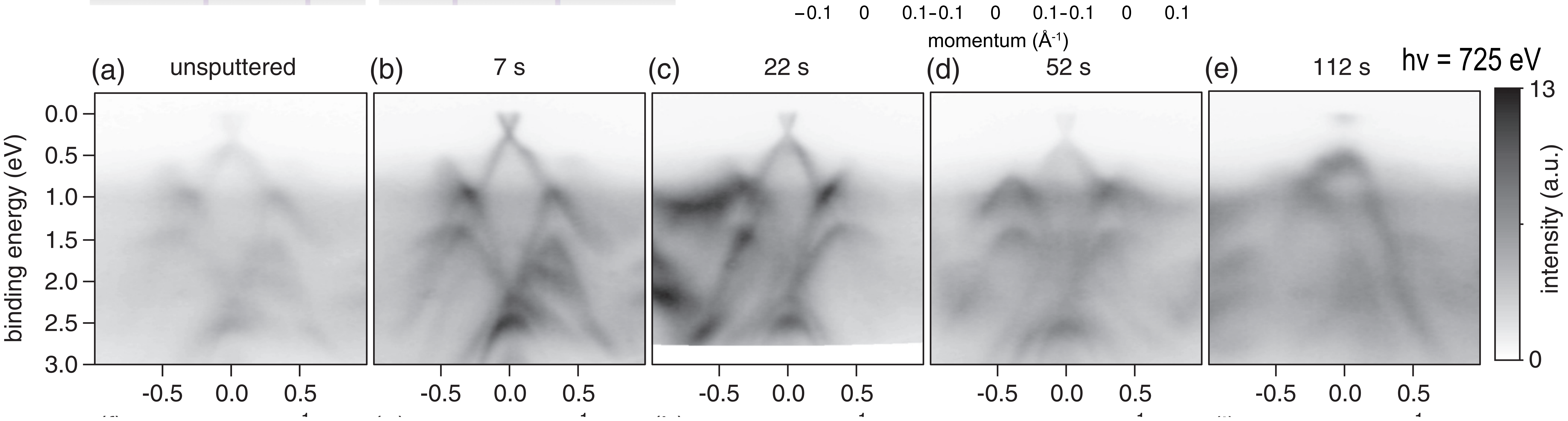
Soft X-ray ARPES on sputtered topological insulator Bi_2Se_3



Topological protection: At interface of change in topology in-gap state **must** exist

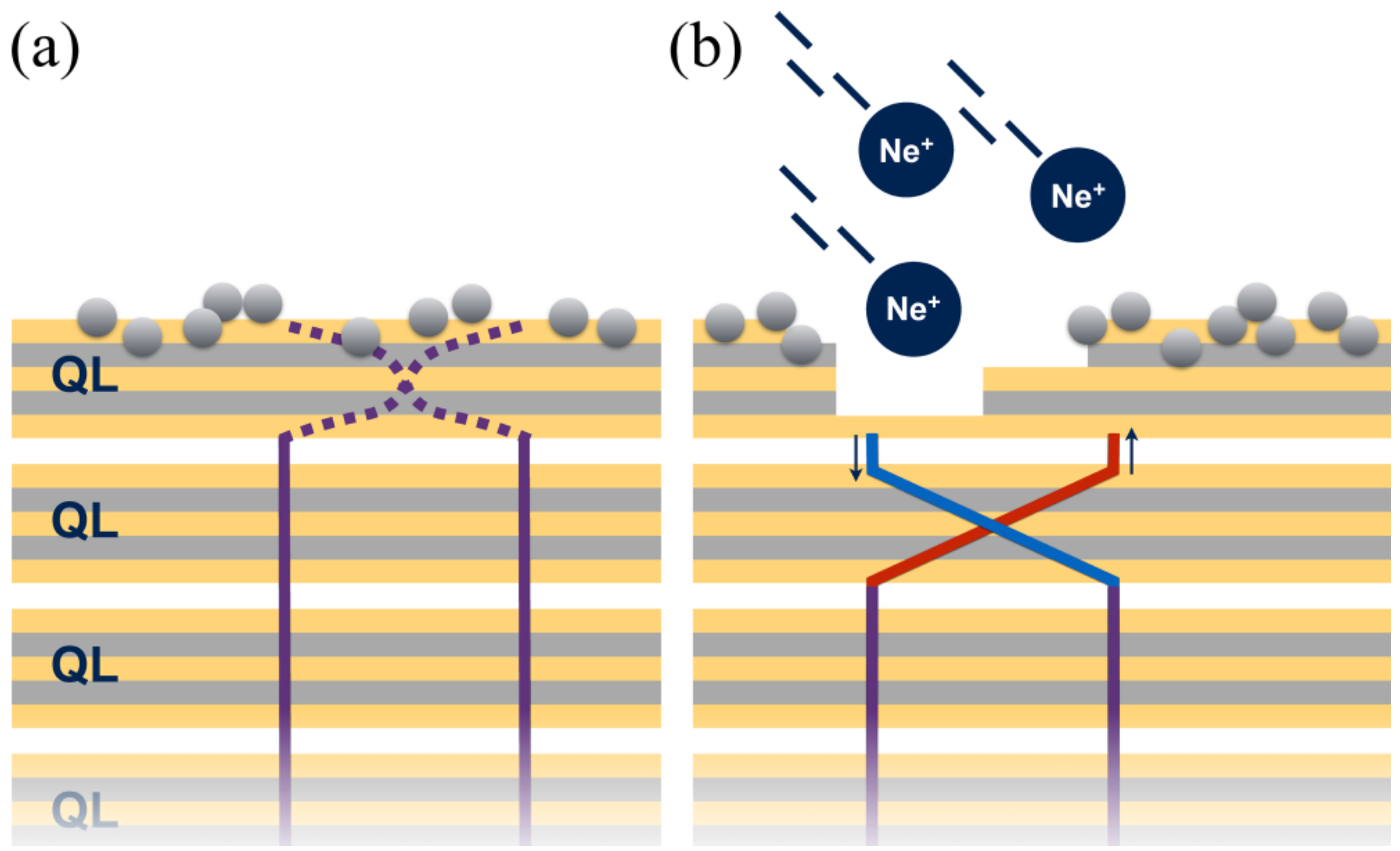
Protection occurs by state moving away from areas with high defects

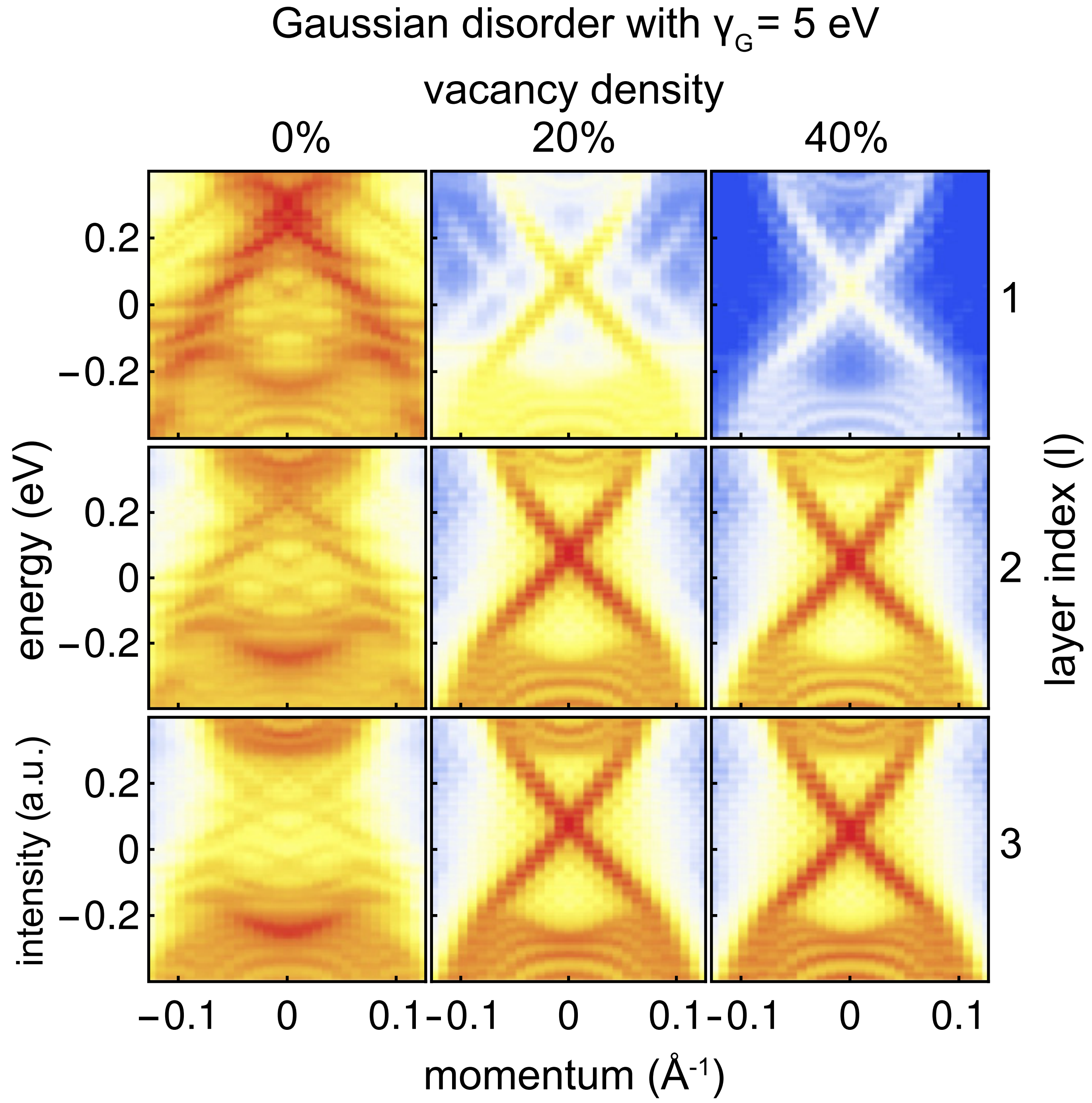
Probed with relatively high hv



R. Queiroz et al. PRB 93, 165409 (2016)

Soft X-ray ARPES on sputtered topological insulator Bi_2Se_3





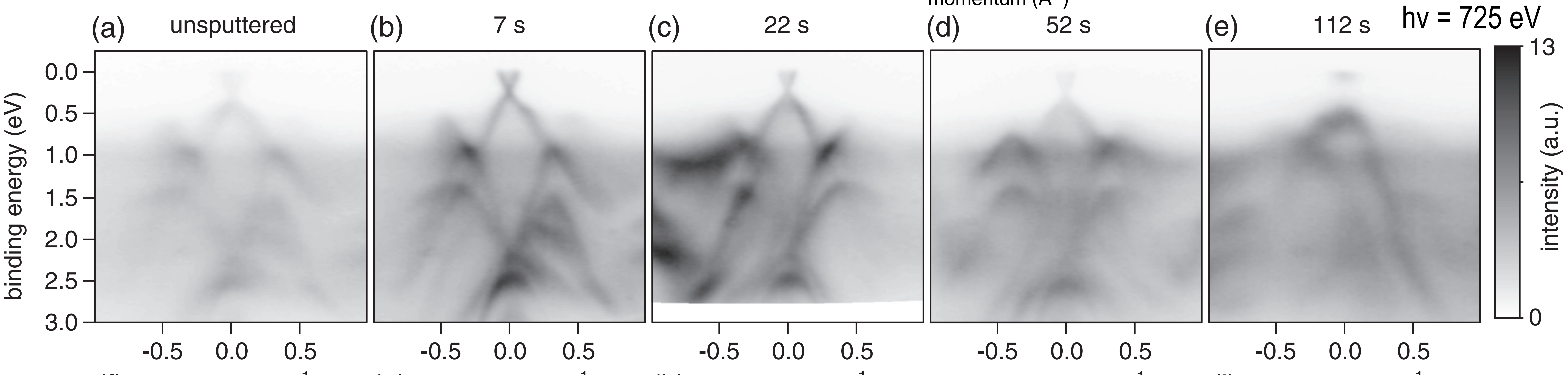
Topological protection: At interface of change in topology in-gap state **must** exist

Protection occurs by state moving away from areas with high defects

Probed with relatively high hv

Gaussian disorder by surface adsorbates reduces overall intensity

Unitary disorder by sputtering reduces influence of Gaussian disorder

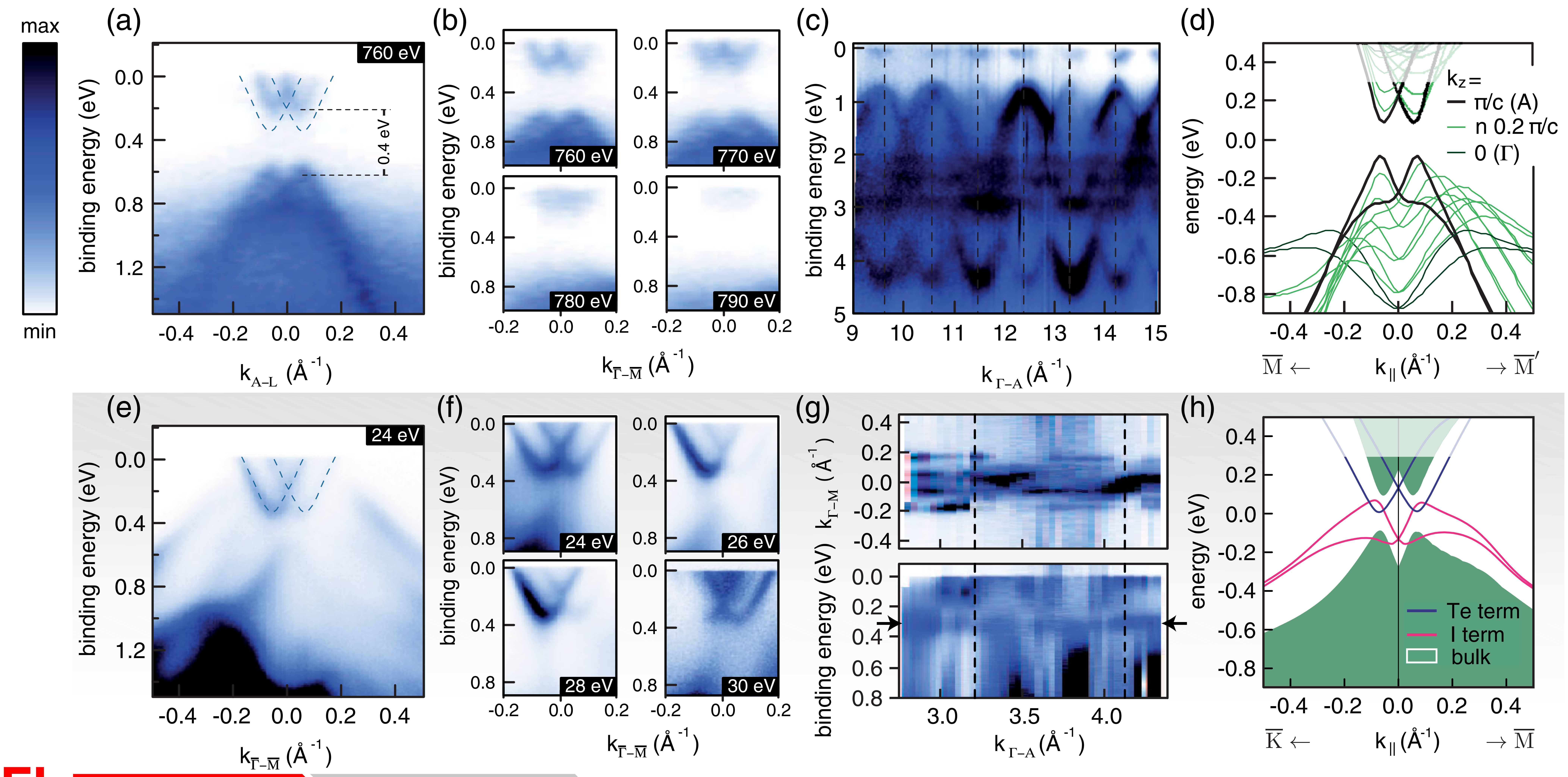


R. Queiroz et al. PRB 93, 165409 (2016)

Localised surface states

PRL 109, 116403 (2012) PHYSICAL REVIEW LETTERS week ending 14 SEPTEMBER 2012

Disentanglement of Surface and Bulk Rashba Spin Splittings in Noncentrosymmetric BiTeI



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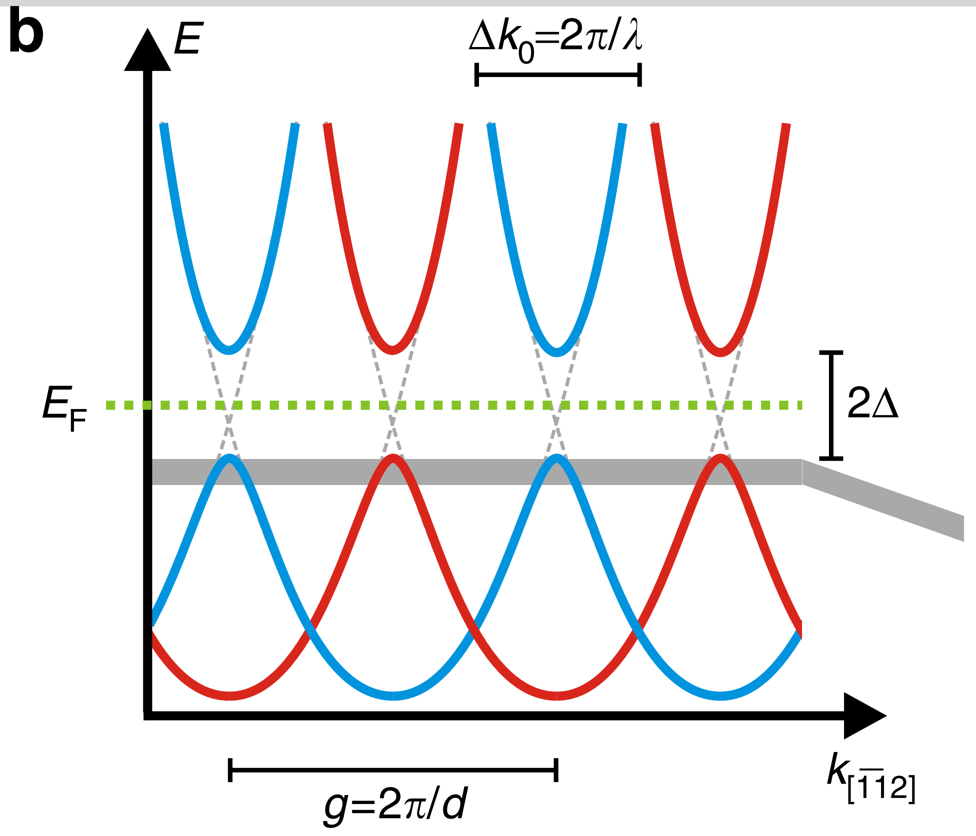
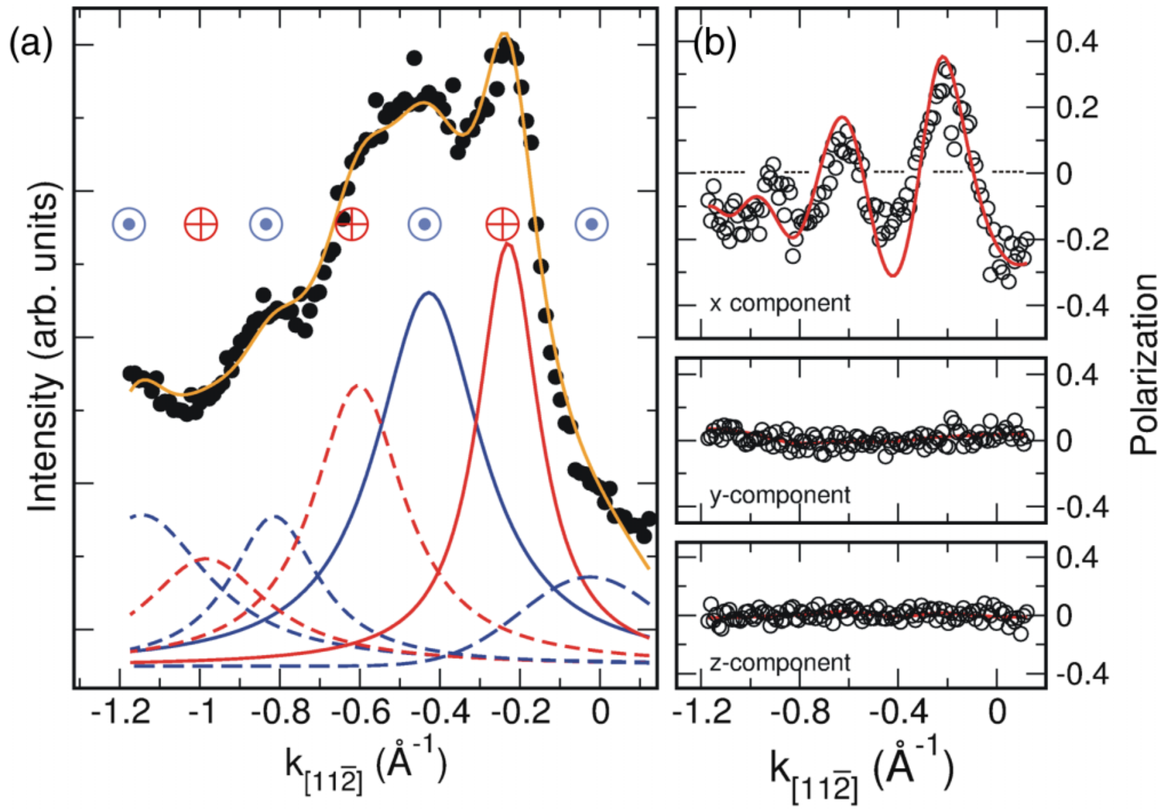
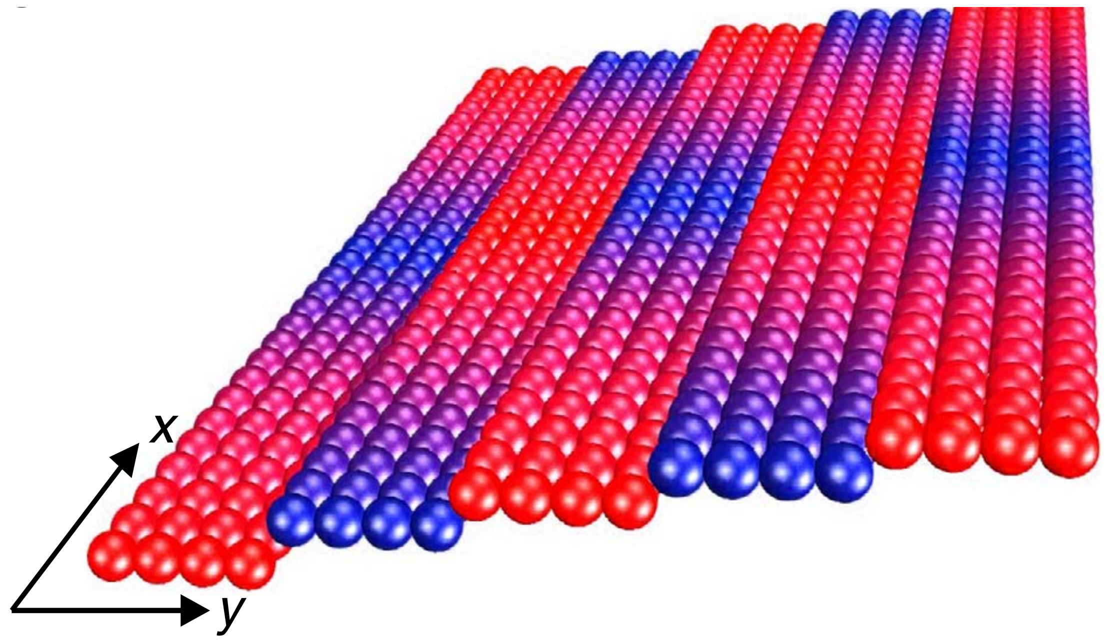
SOIS: Spin-Orbit Interaction Spectroscopy group

part of Laboratory for Topological Matter
www.epfl.ch/labs/ltm/

1. **Spin-orbitronics**: Rashba effect, Spintronics "without" magnetism





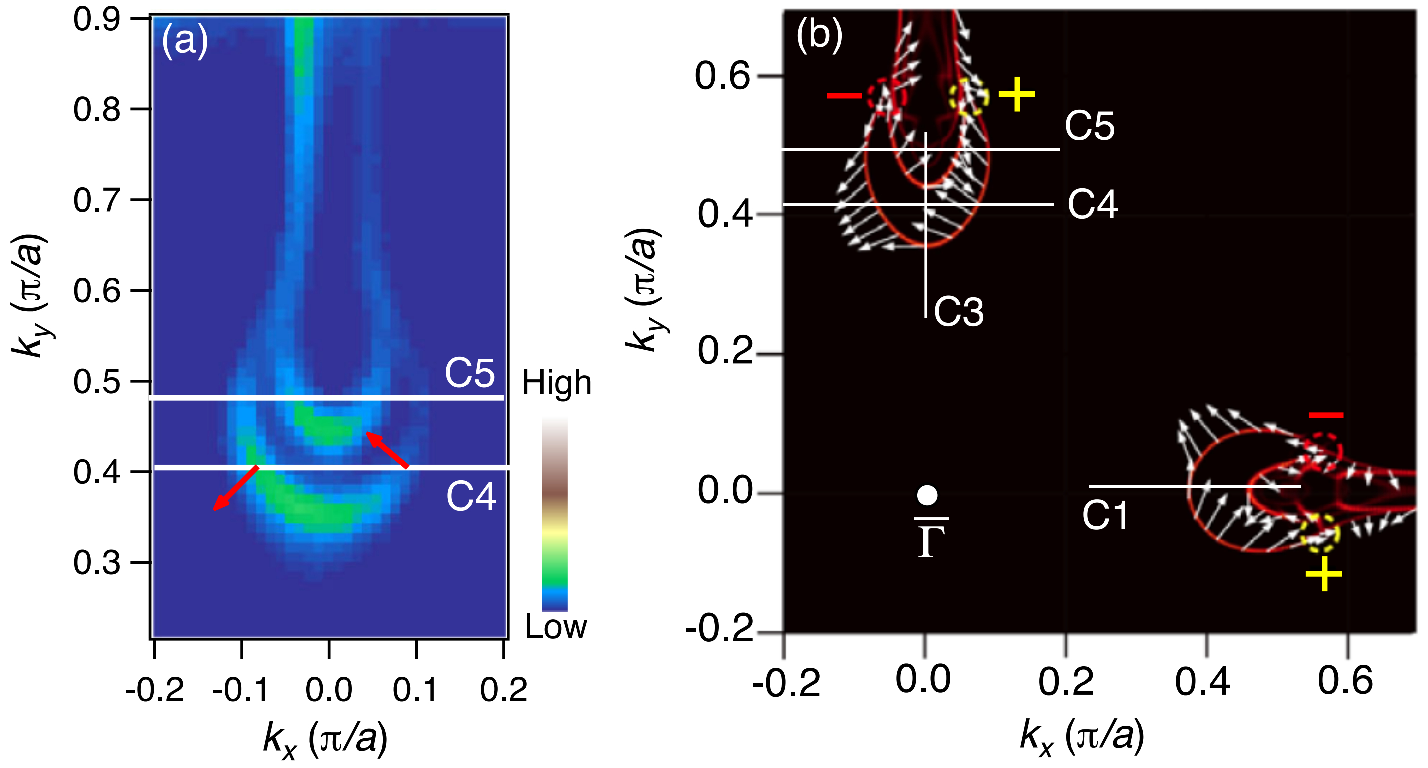


Spin-orbit density wave (SODW) in Pb/Si(557)

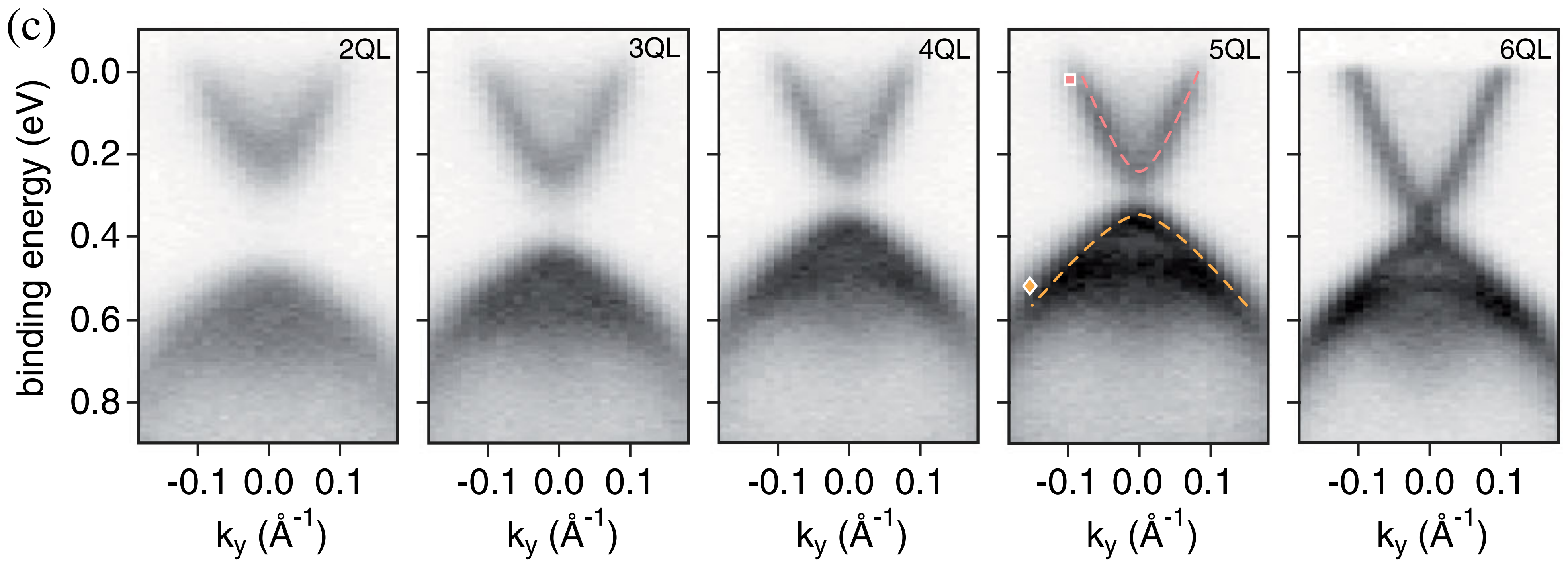
C. Tegenkamp et al. PRL 109, 266401 (2012);
C. Brand et al. Nature Comm. 6, 8118 (2015)

2. **Novel physical phases**: Topological insulators; Topological protection and transition, type-I and type-II Weyl semimetals

Spin texture of Weyl semimetal TaAs



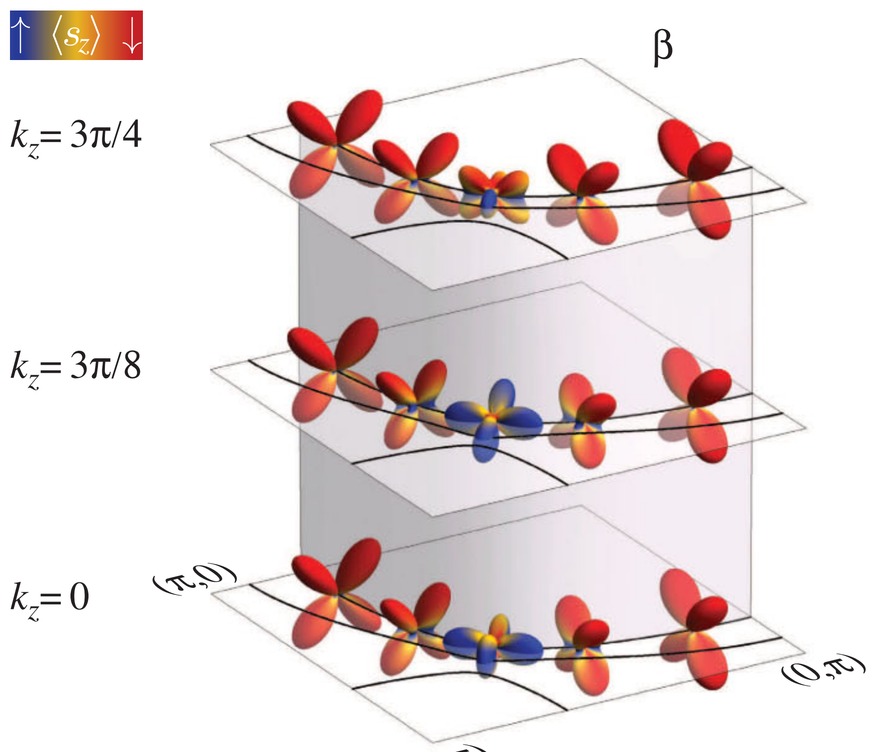
B.Q. Lv, S. Muff et al. PRL 115, 217601 (2015)

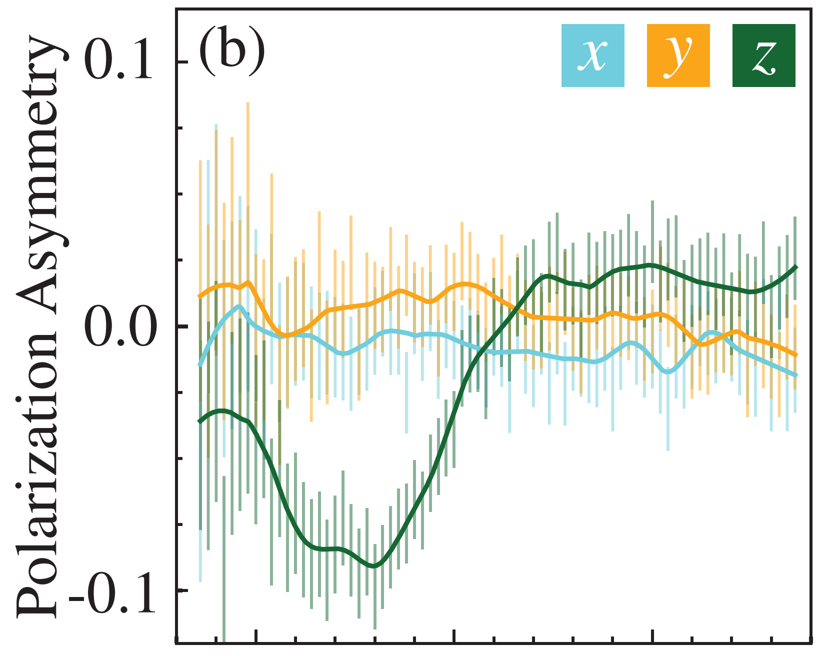


Topological transition in Bi_2Se_3 films

G. Landolt et al. PRL 112, 057601 (2014)

Breakdown of Singlets and Triplets in Sr_2RuO_4





C. Veenstra et al. PRL 112, 127002 (2014)

3. **Extra tag in spectroscopy**: photoelectron time scale, orbital mapping, phase determination, quasiparticle interactions …

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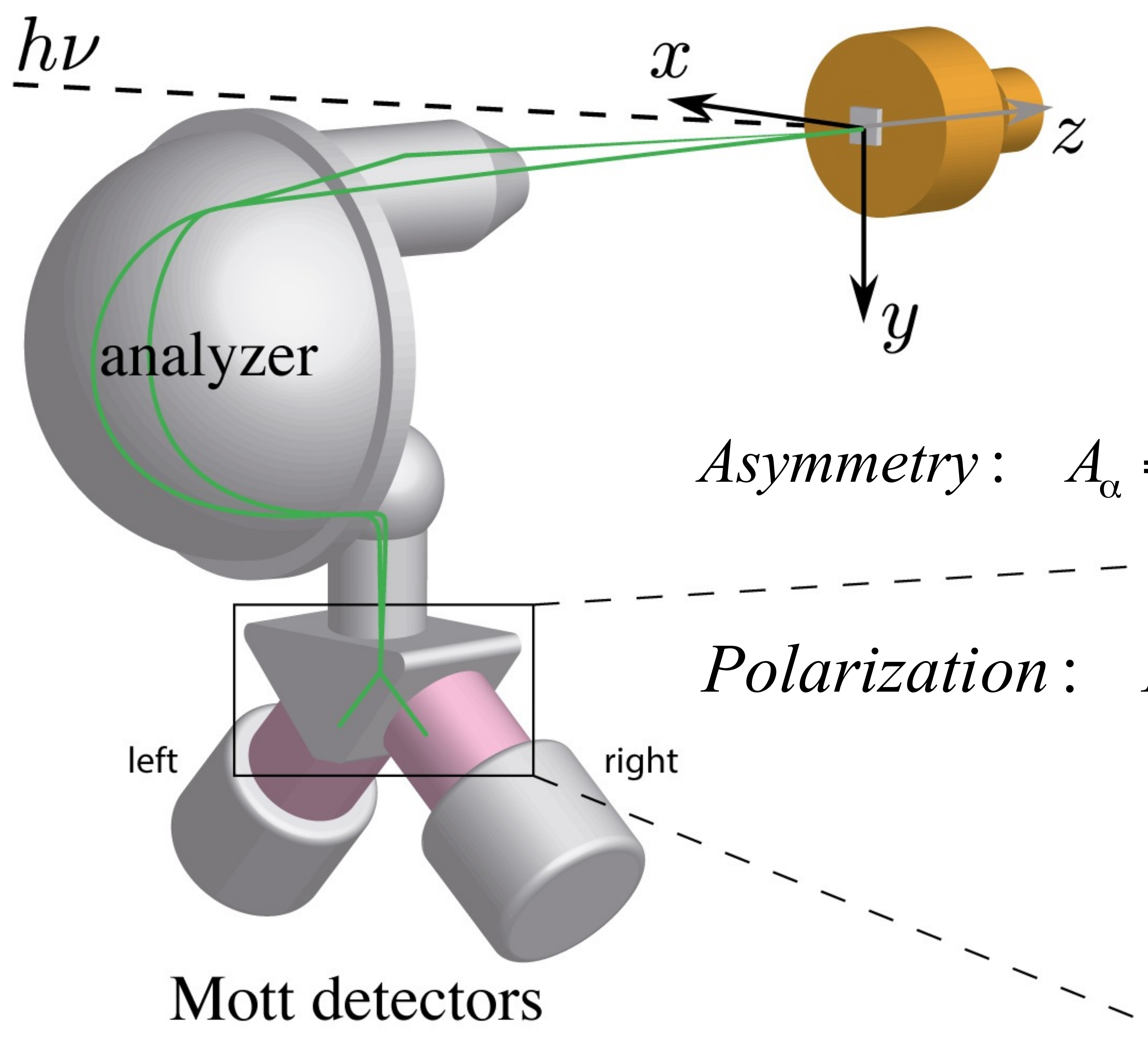
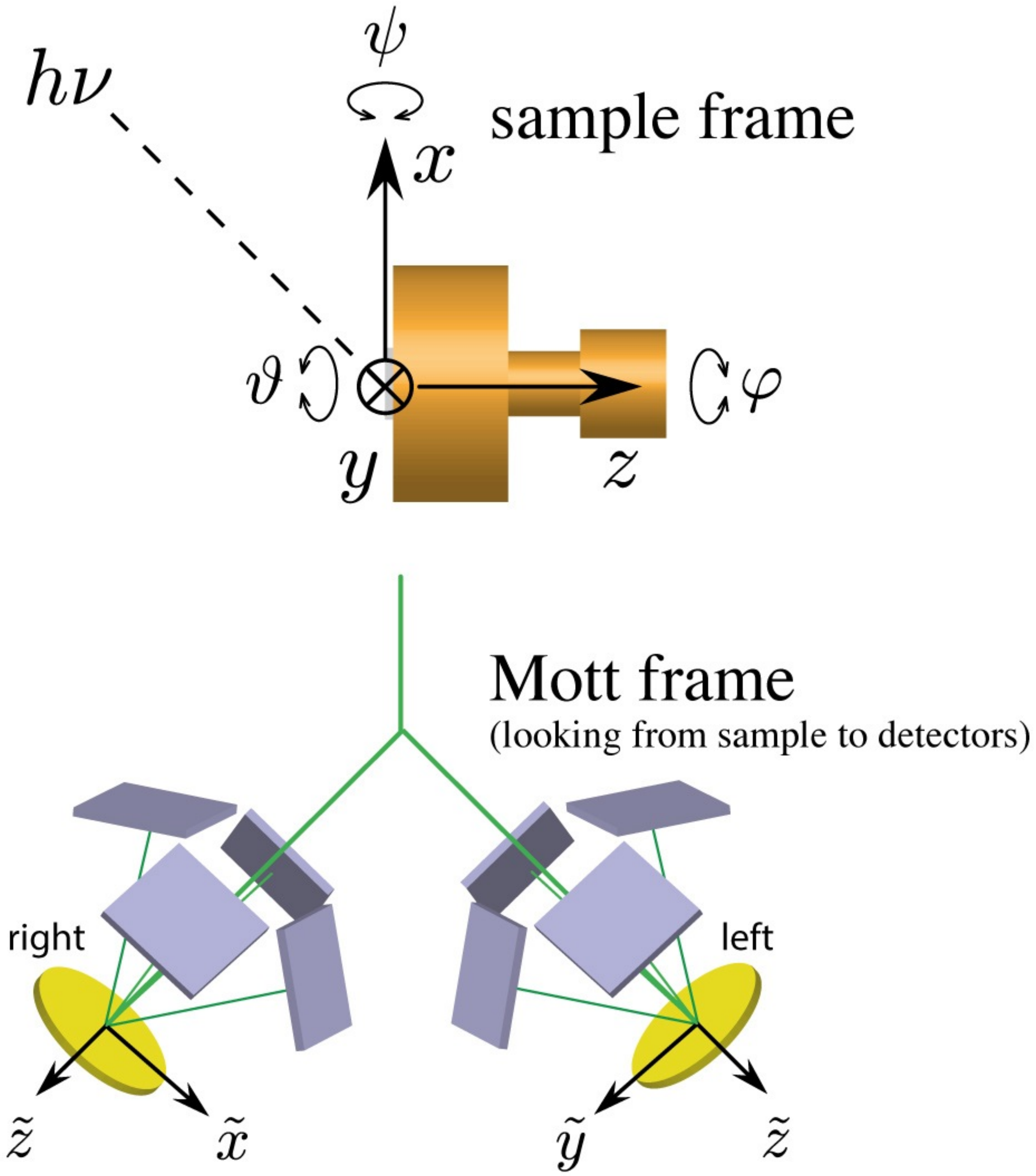
COPHEE at the Swiss Light Source

COPHEE = The COmplete PHotoEmission Experiment



Asymmetry : $A_\alpha = \frac{N_l - N_r}{N_l + N_r}$

Polarization : $P_\alpha = \frac{A_\alpha}{S}$

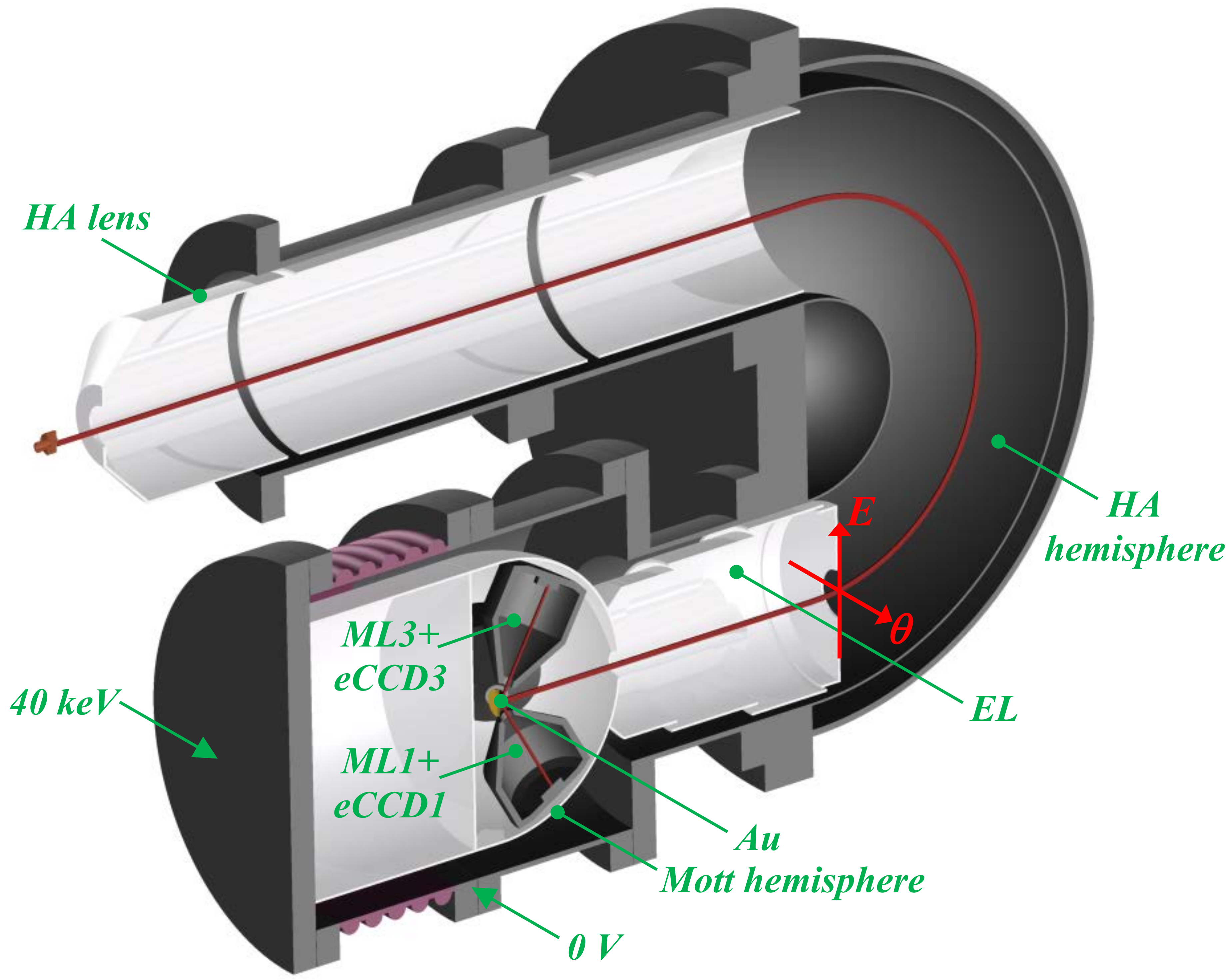


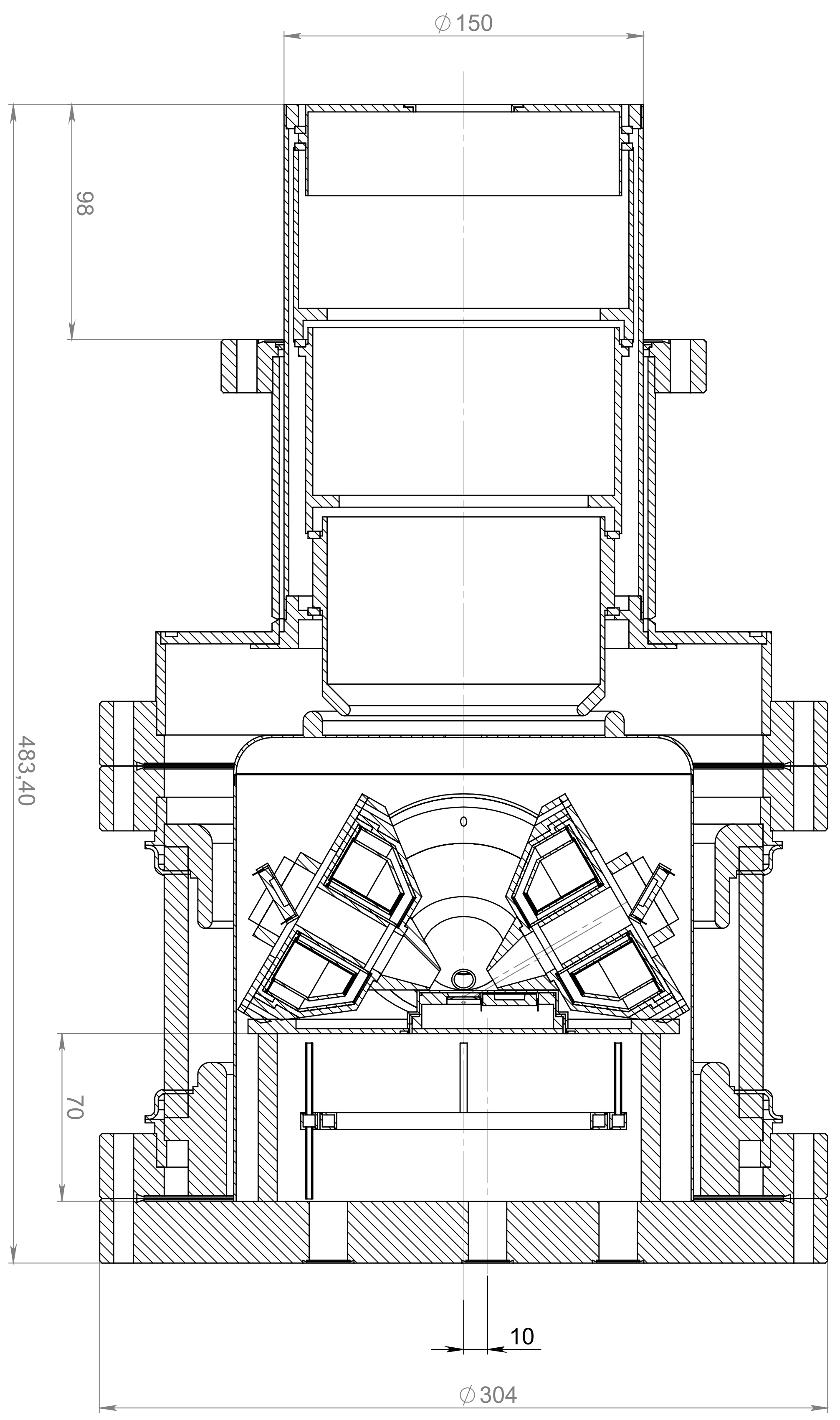
Two orthogonal classical (40 kV) Mott detectors
Access to "all" quantum numbers of the electron:
energy, momentum (3D) and spin (3D)

For reviews on SARPES on SOI systems see: J.H. Dil, J. Phys.: Cond. Matt. 21, 403001 (2009)
and U. Heinzmann and J.H. Dil, J. Phys.: Cond. Matt. 24, 173001 (2012)

For review of SARPES on Topological Materials see: J.H. Dil, Elec. Structure 1 023001 (2019)

Next generation: iMott





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Full spin-resolved 2D (E_k,θ) band structure in one shot

Enhancement of efficiency by ~10^4

Single event detection also very suitable for low intensity (spin integrated) spectroscopy

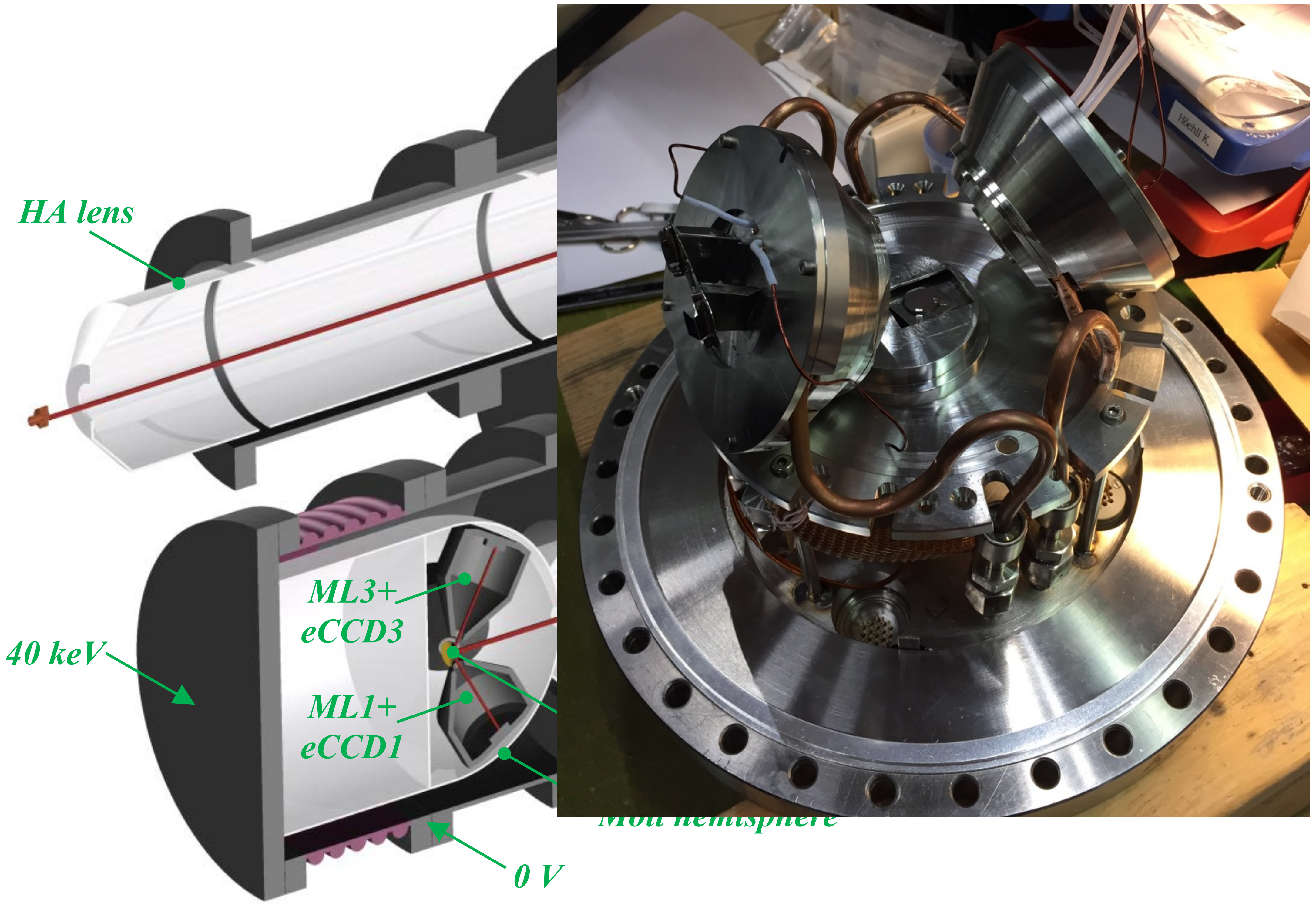
V. Strocov, V. Petrov, and J.H. Dil, J. Synchrotron Radiation 22, 708 (2015)

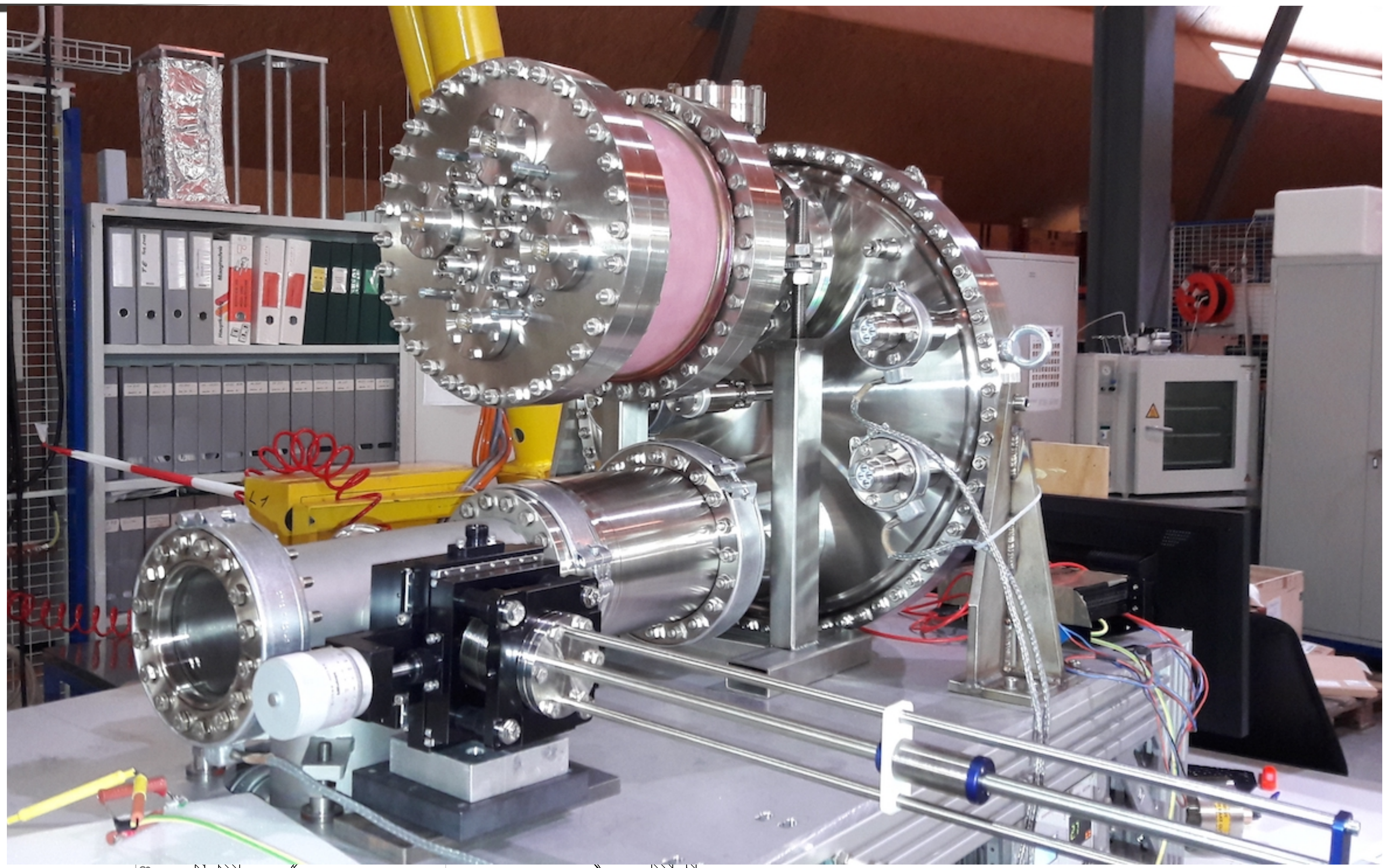
For other approaches see:

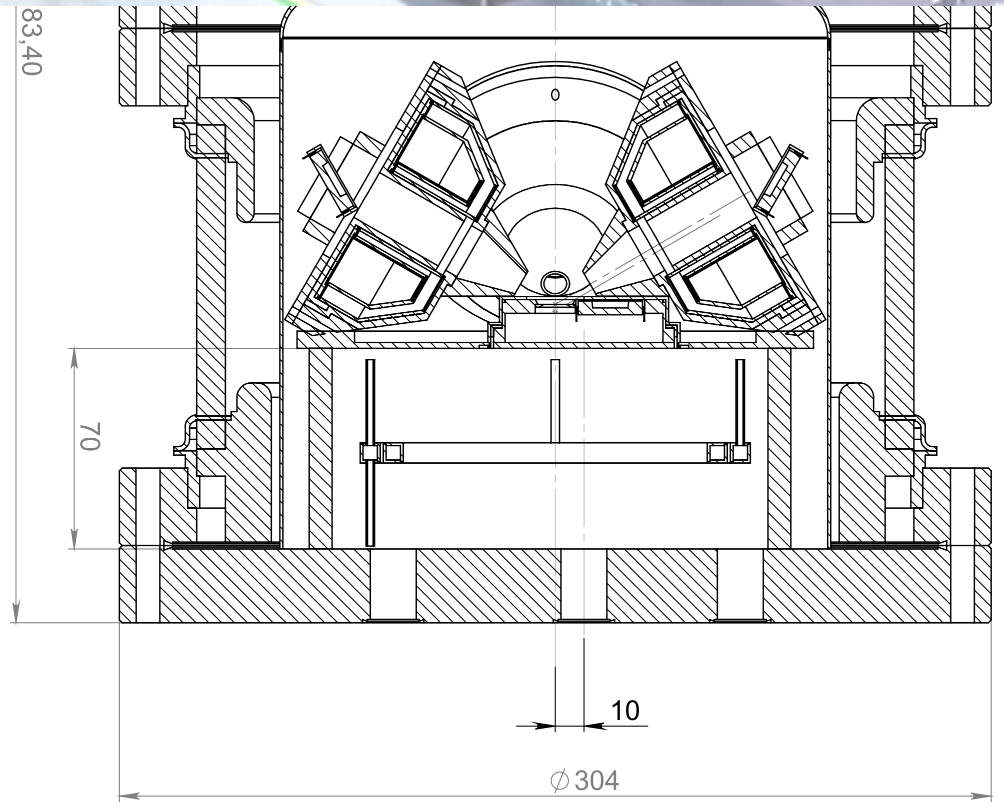
G. Schönhense et al. J. Electron Spectrosc. Relat. Phenom. 200, 94 (2015)

T. Okuda J. Phys.: Condens. Matter 29 483001 (2017)

Next generation: iMott







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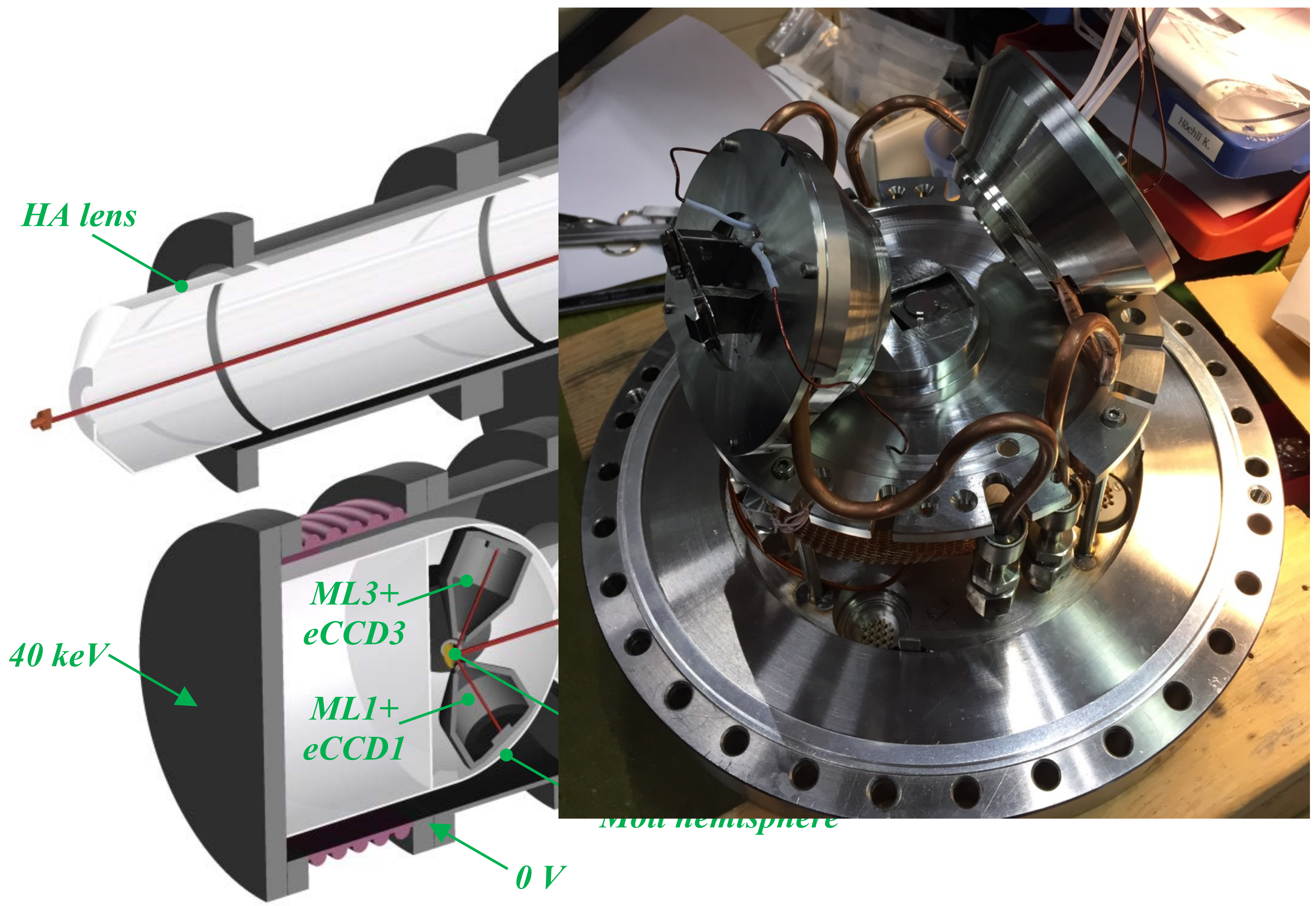

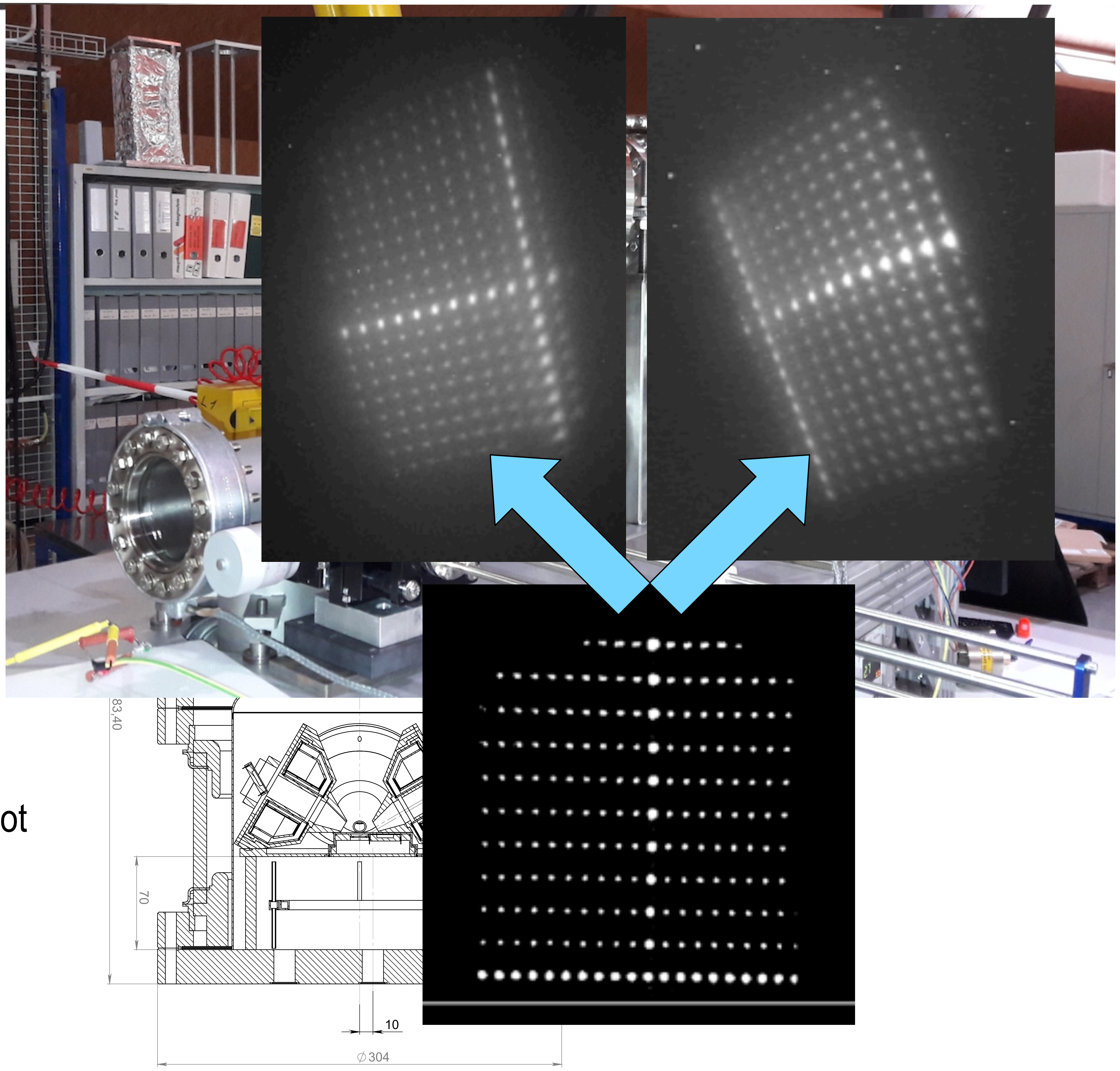

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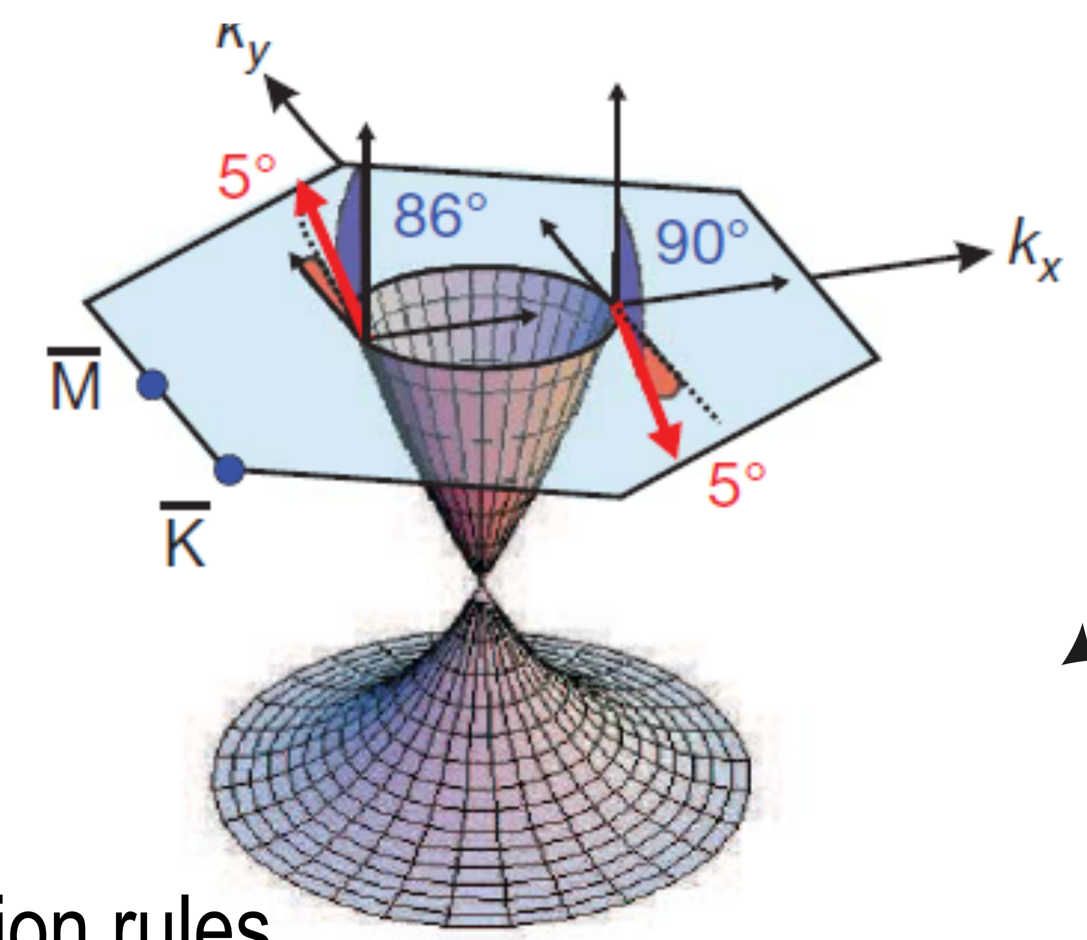
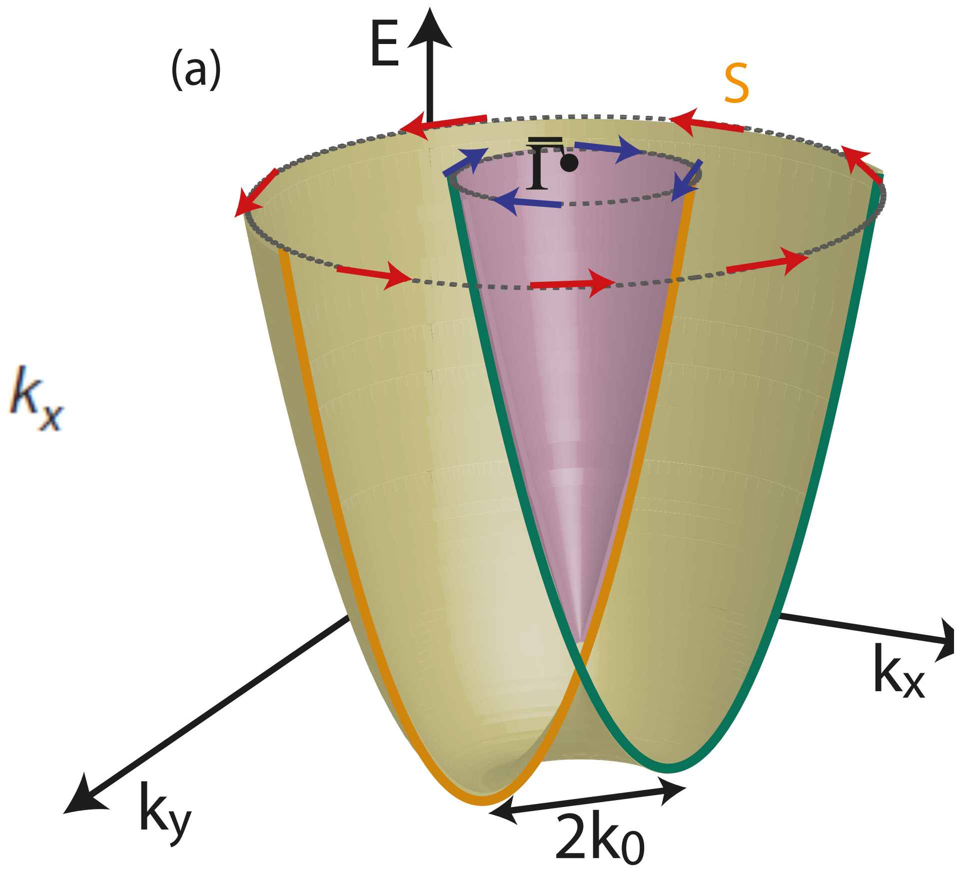
T. Okuda J. Phys.: Condens. Matter 29 483001 (2017)

Sources of spin polarization in SARPES

“All” photoelectrons are highly spin polarised

For a review: U. Heinzmann and J.H. Dil, J. Phys.: Condens. Matter 24, 173001 (2012)





Spin polarized initial states:

- Magnetic systems
- Spin-orbit interaction (Rashba, topological materials)

Photoemission induced effects:

- Change of initial state spin: spin interference, geometry induced effects, …
- Spin induced in spin-degenerate initial state: geometry effects, dipole selection rules, interference of transitions, …

$$\varphi_{x+} = \frac{1}{\sqrt{2}}\begin{pmatrix}1\\1\end{pmatrix}, \quad \varphi_{x-} = \frac{1}{\sqrt{2}}\begin{pmatrix}1\\-1\end{pmatrix}$$

Basis vectors of spin space

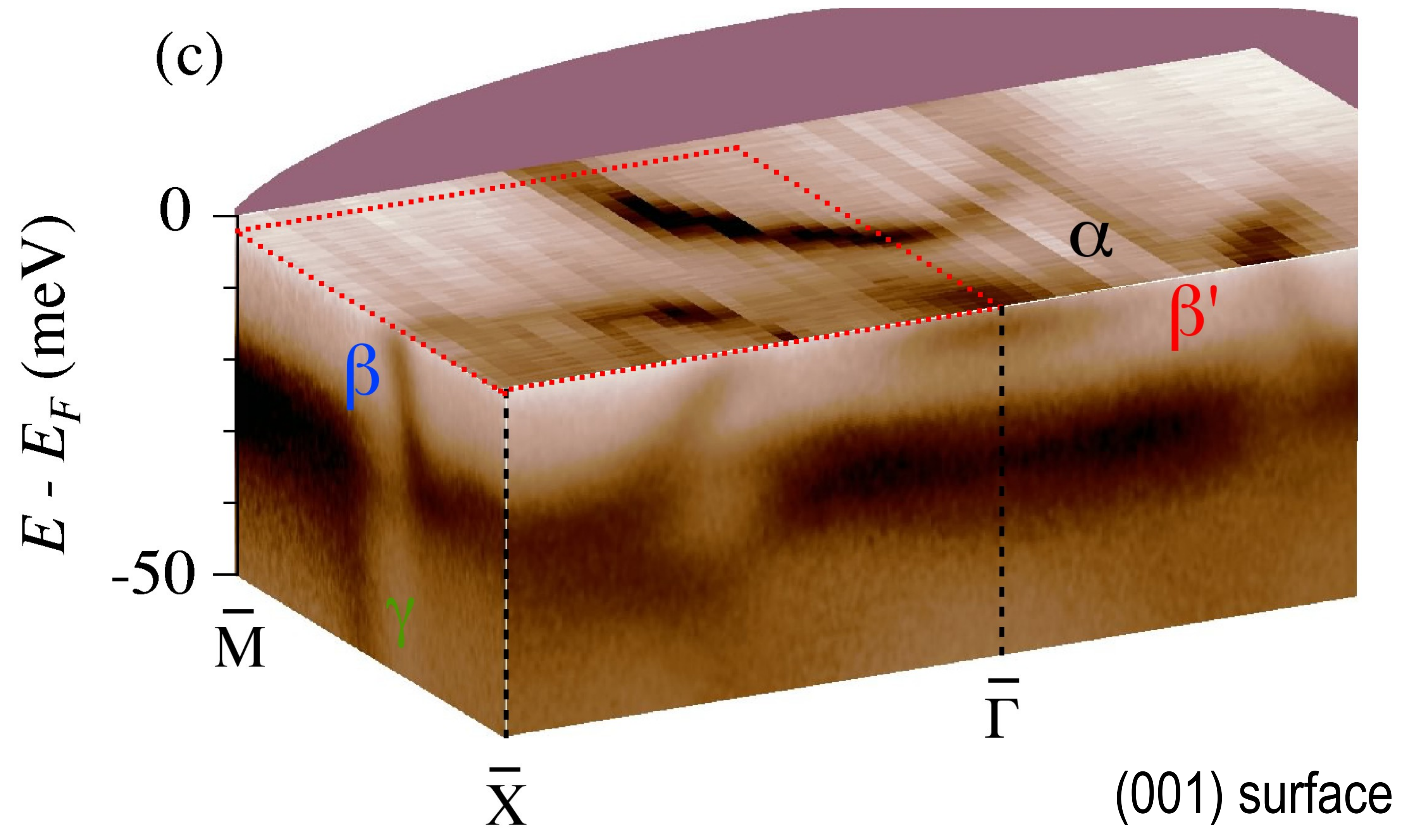
$$\varphi_{y+} = \frac{1}{\sqrt{2}}\begin{pmatrix}1\\i\end{pmatrix}, \quad \varphi_{y-} = \frac{1}{\sqrt{2}}\begin{pmatrix}1\\-i\end{pmatrix}$$

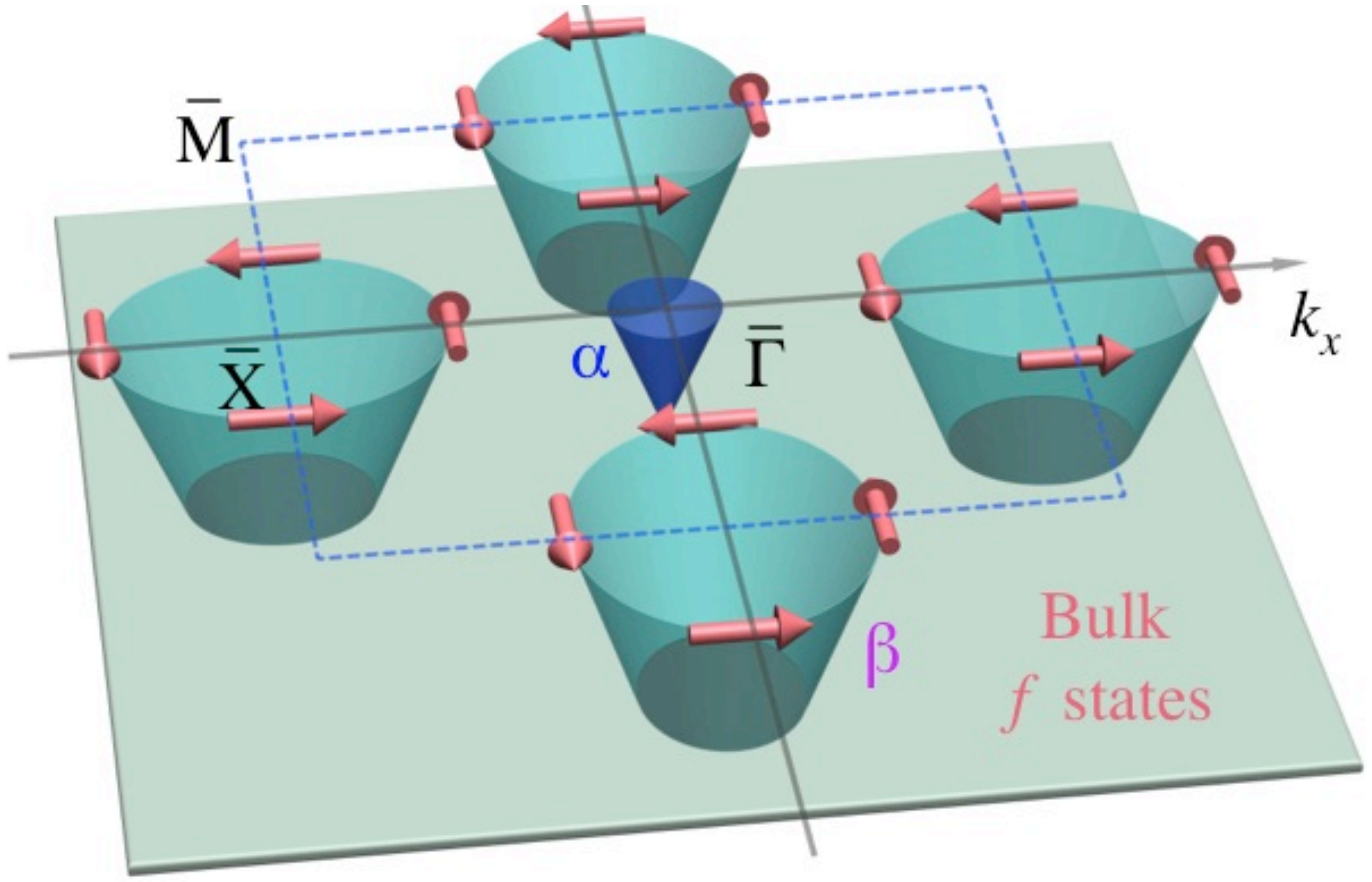
In coherent summation spin rotates

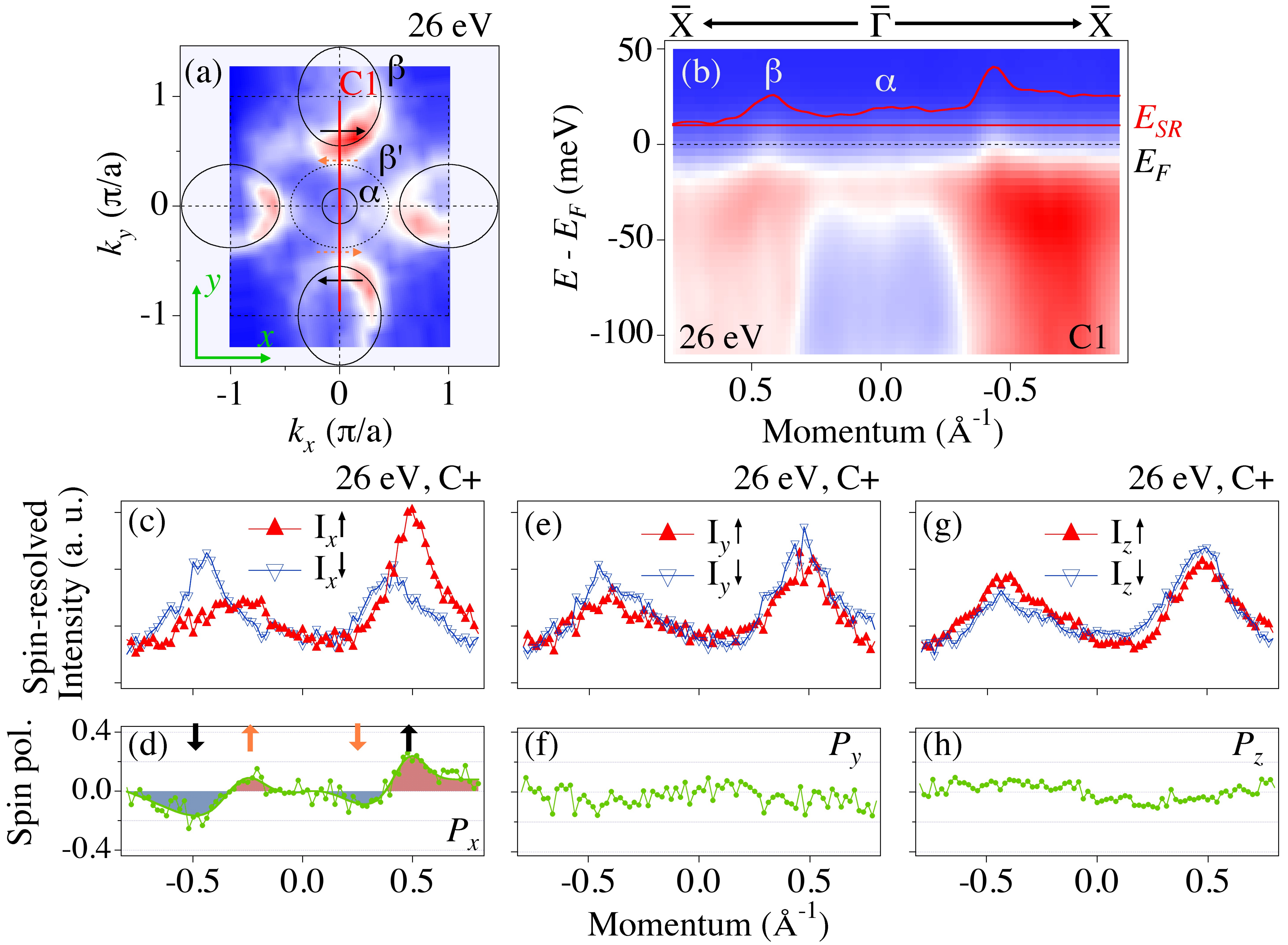
$$\varphi_{z+} = \begin{pmatrix}1\\0\end{pmatrix}, \quad \varphi_{z-} = \begin{pmatrix}0\\1\end{pmatrix}$$

SmB_6 a topological "Kondo" insulator

N. Xu et al. Nature Communications 5, 4566 (2014).







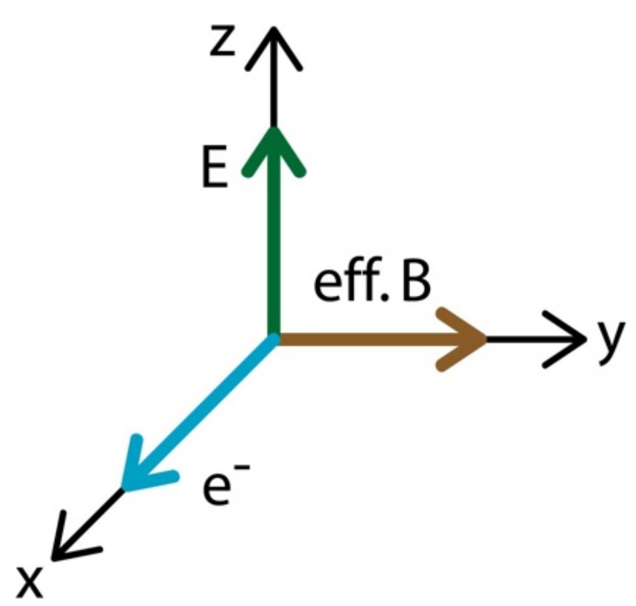
Band gap (40 meV) by definition always around Fermi level due to mixed valence

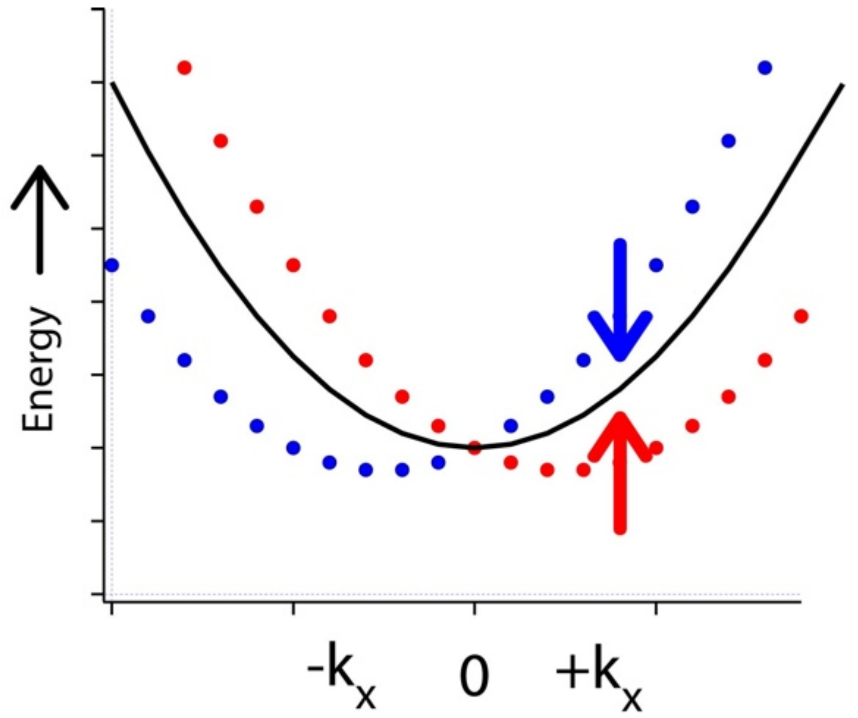
Spin polarised topological surface states in band gap

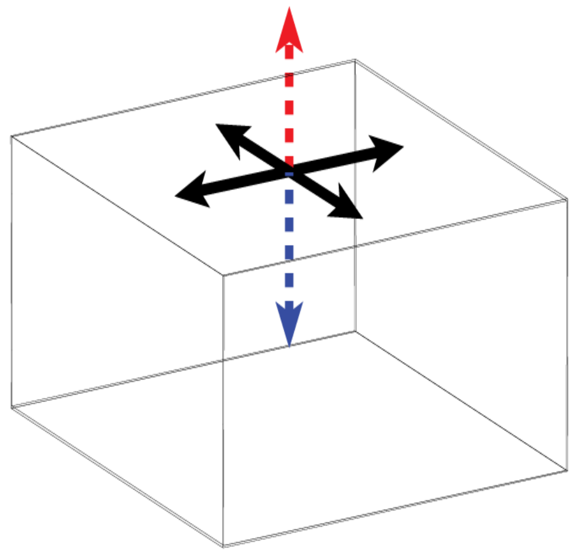
No sign of correlations in the spin structure

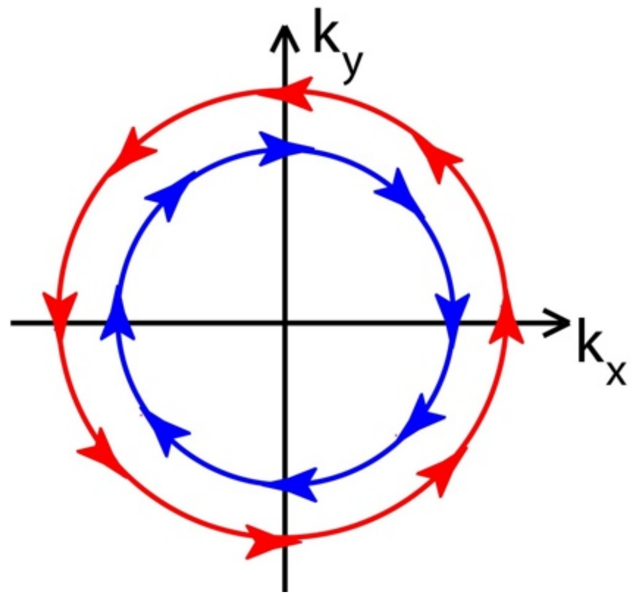
Independent verification for (111) surface: Y. Ohtsubo et al. Nature Comm. 10, 2298 (2019)

Rashba type spin-orbit interaction









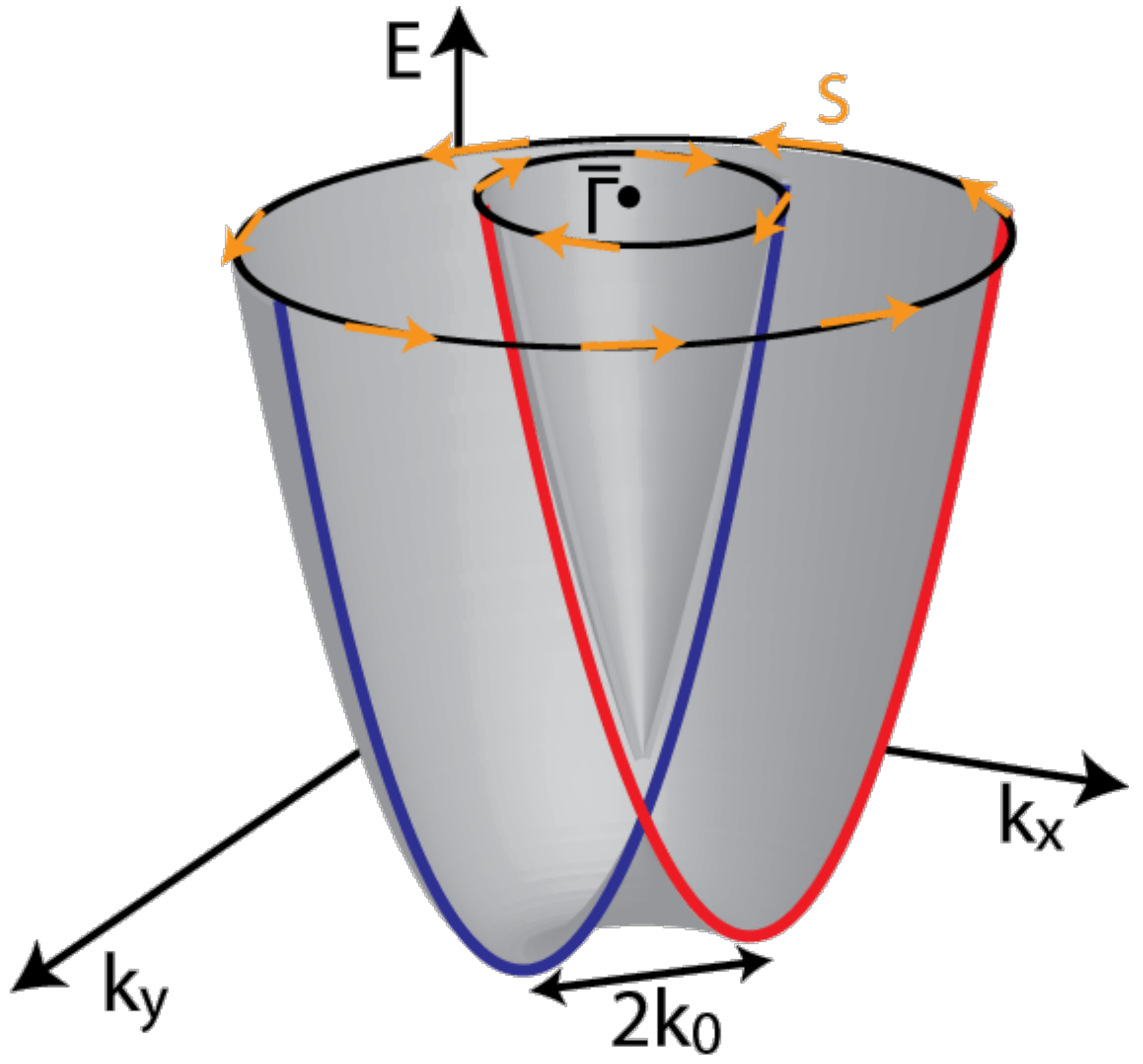
Size of splitting depends on:

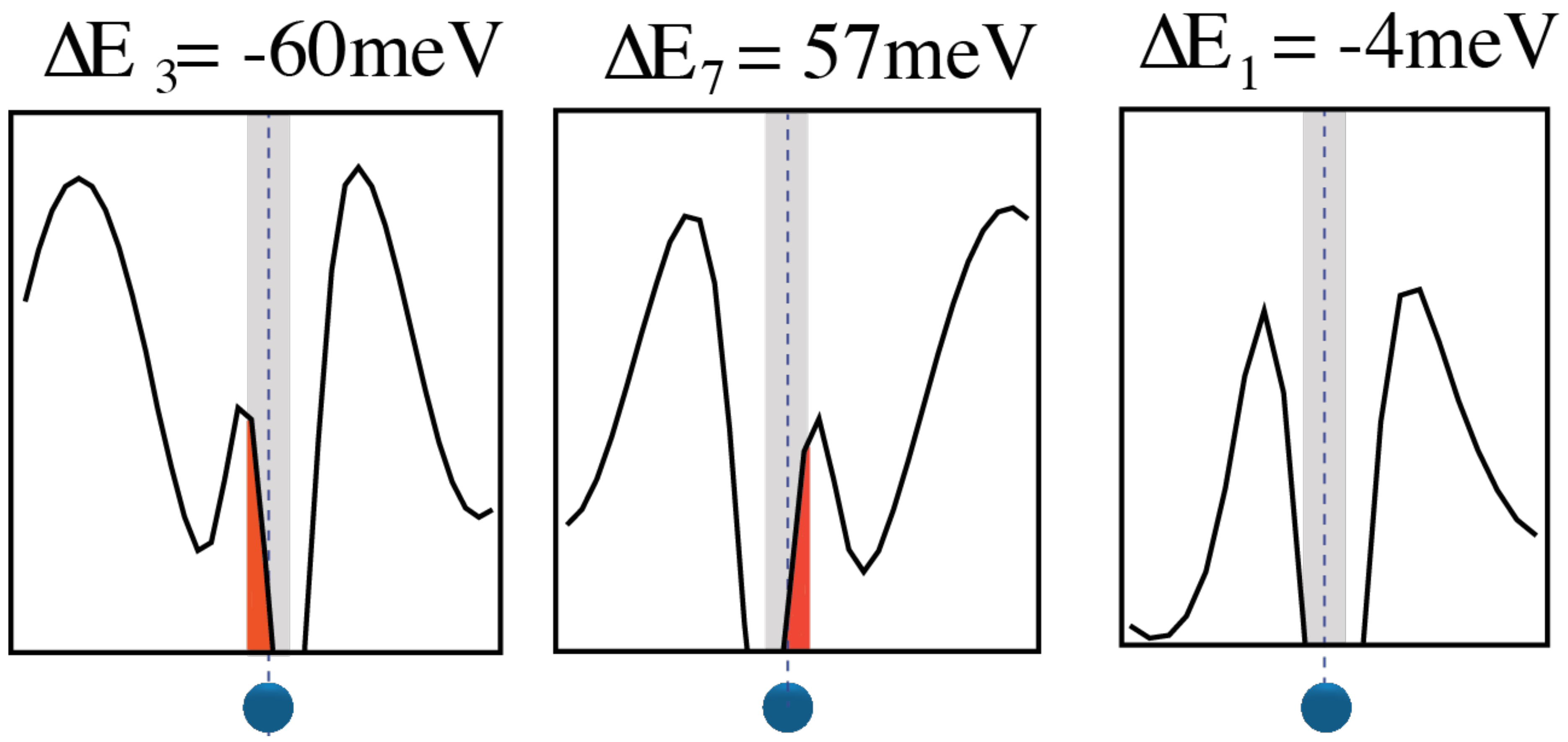
-atomic number

-atomic displacement/corrugation

-orbital overlap

-potential gradient at surface





B. Slomski et al. PRB **84**, 193406 (2011)

Origin in local wave function distribution 0.2 Å around heavy atom core:

$$\Delta E_i \propto k_{\|} \int_{\Delta z} \mathrm{d}z \frac{\partial V}{\partial z} |\psi(z)|^2$$

Total measured Rashba splitting is sum of all contributions

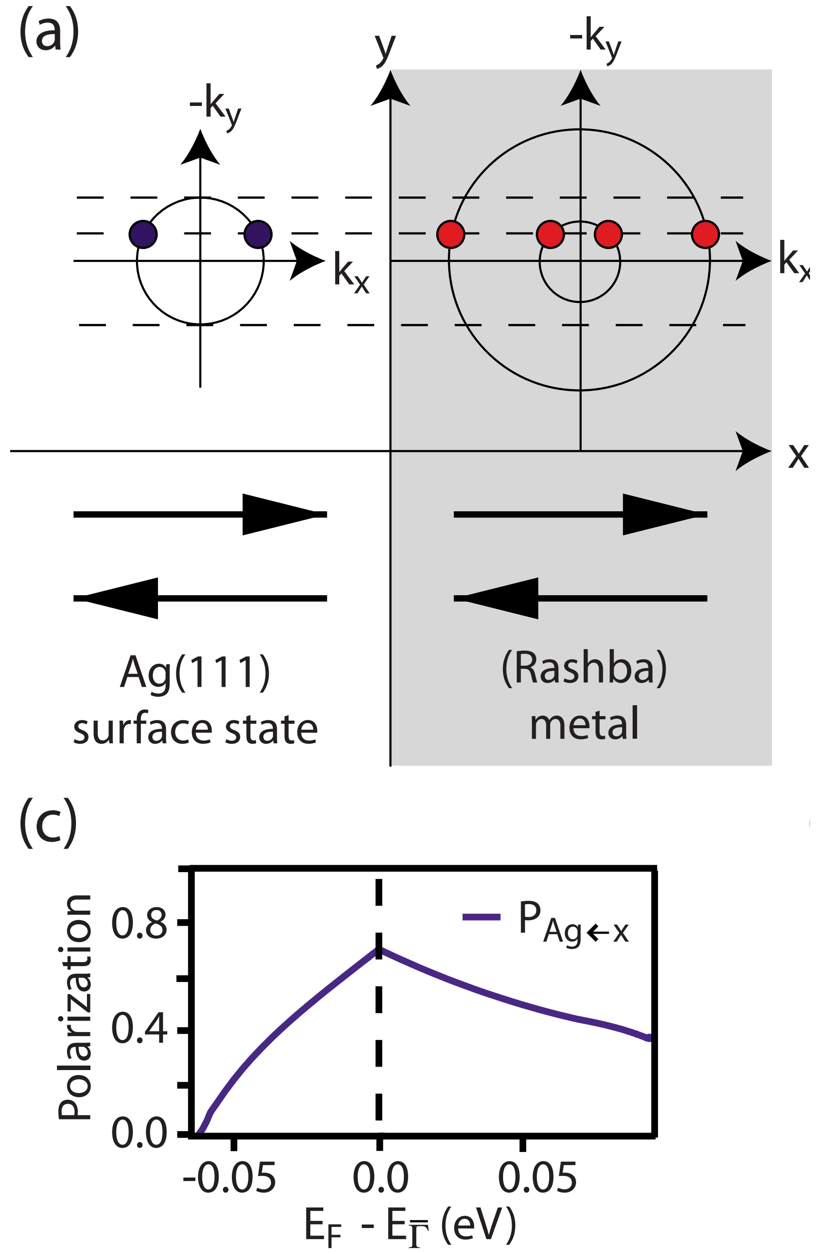
G. Bihlmayer et al. Surf. Sci. **600**, 3888 (2006)

M. Nagano et al. J. Phys.: Condens. Matter 21, 064239 (2009).

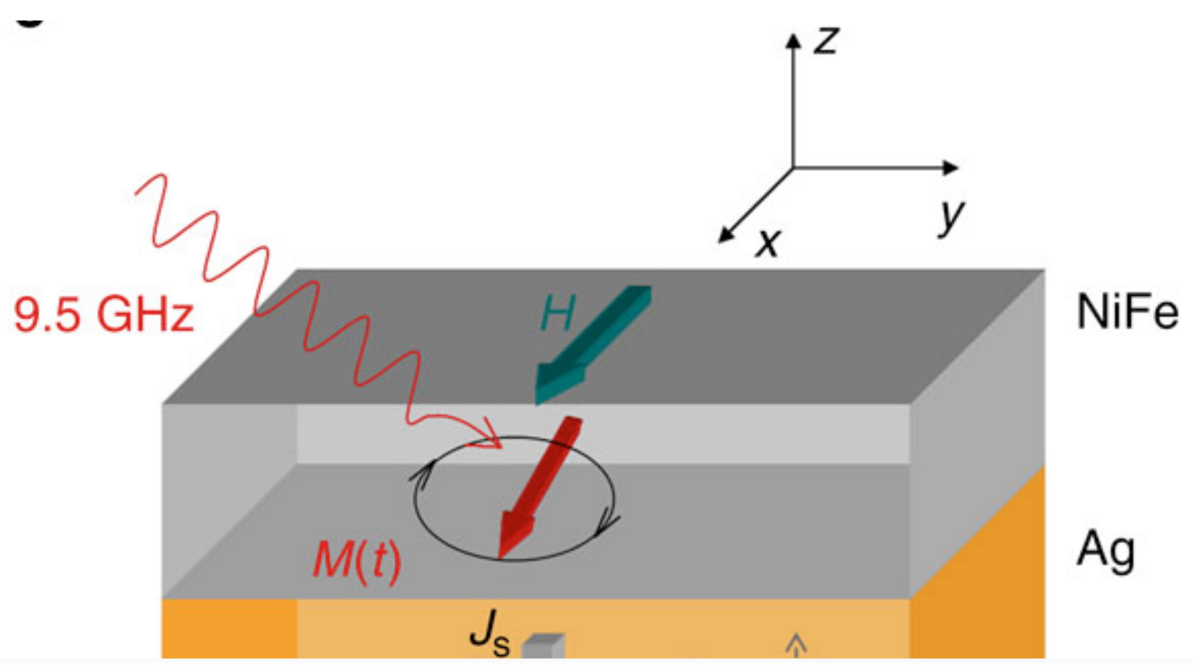
- Original model based on Lorentz transformation
- Six orders of magnitude wrong
- Rashba terminology maintained for historical and symmetry reasons (Rashba-type)

Relevance of the Rashba effect

Spin injection

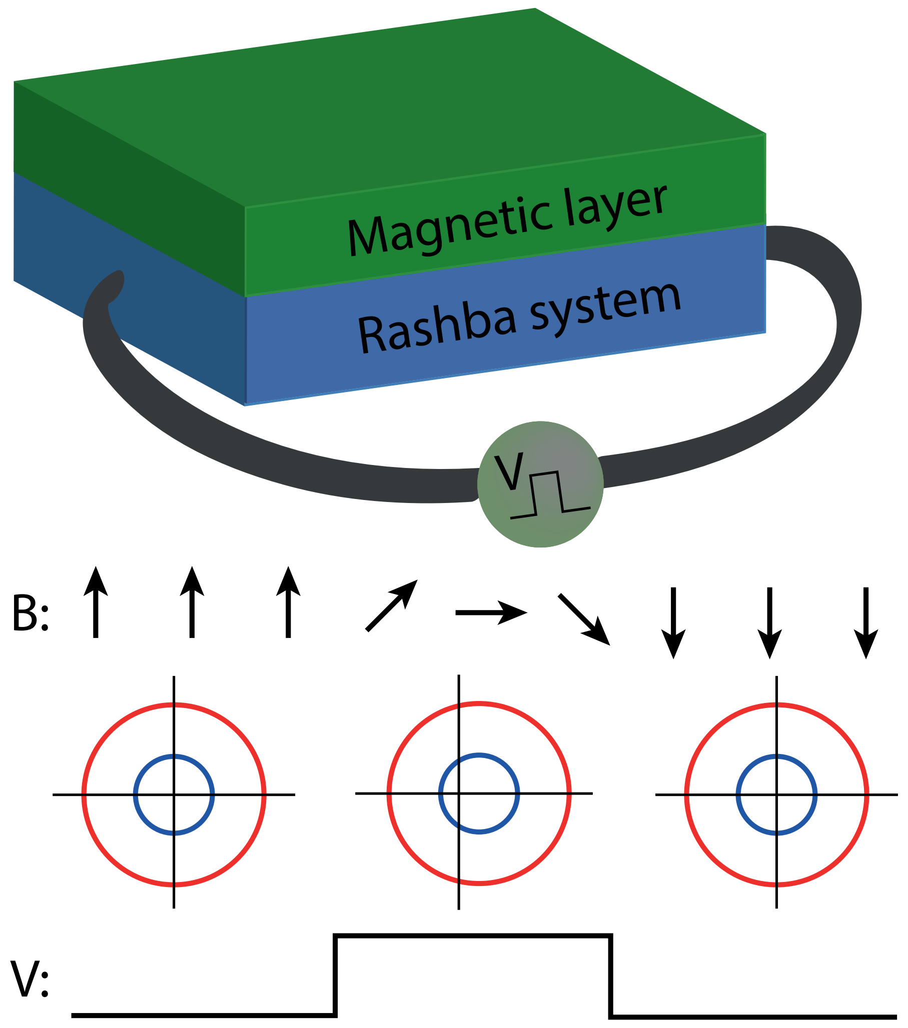


Spin ⟷ charge conversion

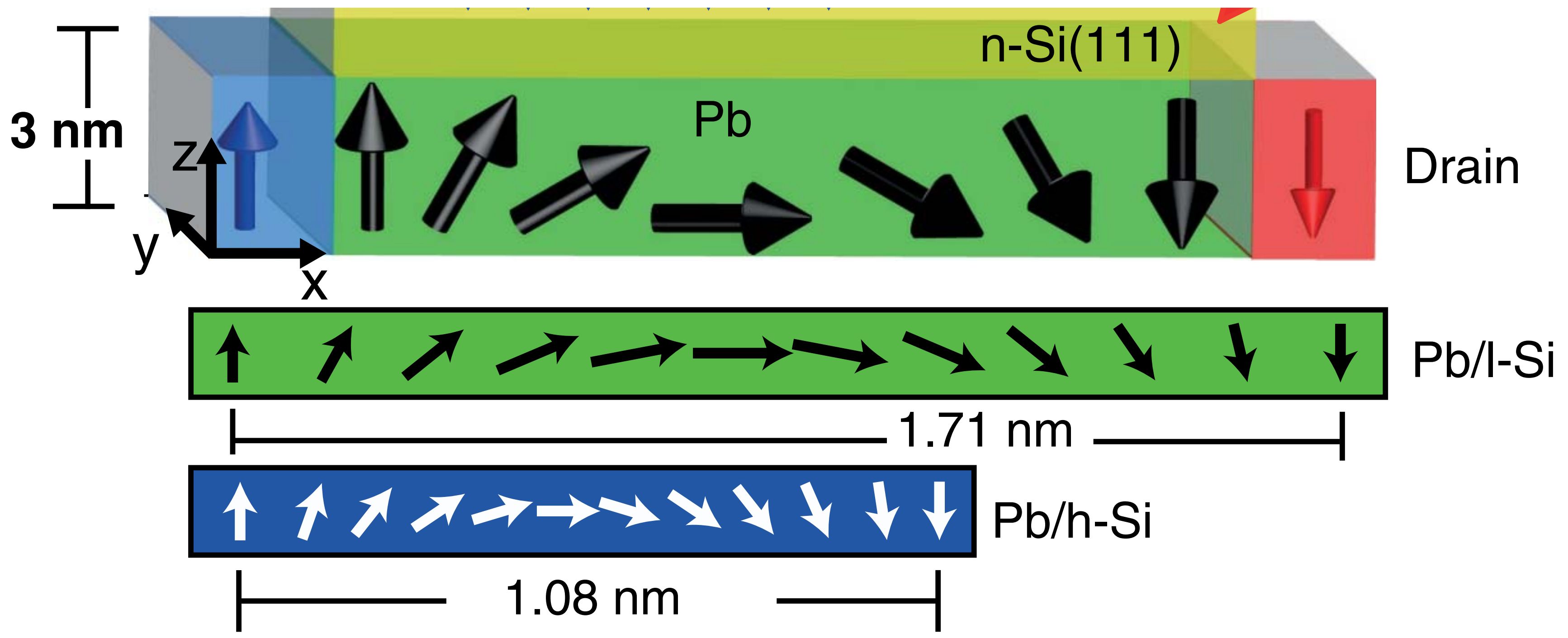


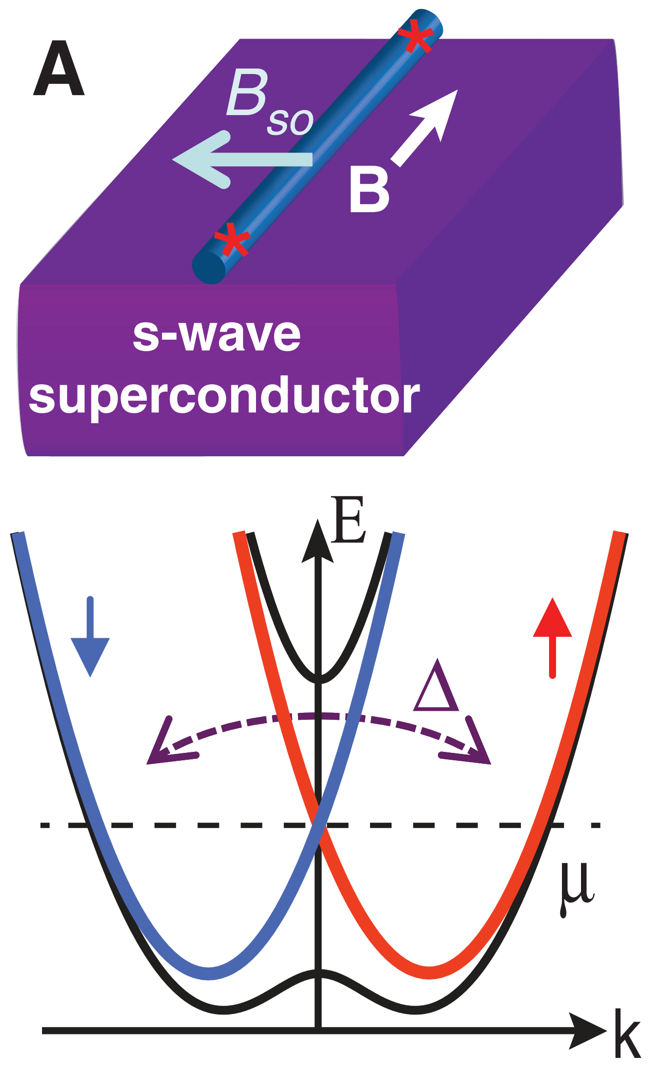
To be used, spurious effects from substrate or bulk have to be negligible and the system tunable

Spin-orbit torque: Magnetic writing by small voltage pulse



Spin manipulation





Majorana fermions

For popular summary:
Nature 539, 509 (2016)

SOIS: Spin-Orbit Interaction Spectroscopy group

Influence of spin-orbit interaction (SOI) in solid state physics

1. **Spin-orbitronics**: Rashba effect; model systems, transition metal oxides, ferroelectrics, multiferroics

F. Meier et al. PRB 77, 165431 (2008)
H. Dil et al. PRL 101, 266802 (2008)
F. Meier et al. PRB 79, 241408(R) (2009)
F. Meier et al. NJP 11, 125008 (2009)
J. Lobo et al. PRL 104, 187602 (2010)
I. Gierz et al. PRB 81, 245430 (2010)
I. Gierz et al PRB 83, 195122 (2011)
P. King et al. PRL 107, 096802 (2011)
B. Slomski et al. B 84, 193406(B) (2011)
P. Höpfner et al. PRL 108, 186801 (2012)
G. Landolt et al. PRL 109, 116403 (2012)
C. Tegenkamp et al. PRL 109, 266401 (2012)
B. Slomski et al. Scientific Reports 3, 1963 (2013)
G. Landolt et al. NJP 15, 085022 (2013)
B. Slomski et al. NJP 15, 125031 (2013)
A. Santander-Syro et al Nature Mat. 13, 1085 (2014)
G. Landolt et al. PRB 91, 081201(R) (2015)
J. H. Dil et al. JESRP 201, 42 (2015)
C. Brand et al. Nature Comm. 6, 8118 (2015)
J. Krempasky et al. Nature Comm. 7, 13071 (2016)
J. Krempasky et al. PRB 94, 205111 (2016)
S. Muff et al. App. Surf. Sci. 432, 41 (2018)
J. Krempasky et al. PRX 8, 021067 (2018)
S. Muff et al. PRB 98, 045132 (2018)
M. Jäger et al. PRB 98, 165422 (2018)
M. Jäger et al. Phys. Status Solidi B, 1900152 (2019)
D. Krieger et al. Crystals 9, 335 (2019)
J. Krempasky et al. PRR 2, 013107 (2020)
G. Kremer et al. PRR 2, 033115 (2020)
E.B. Guedes et al. PRR 2, 033173 (2020)

2. **Novel topological phases**: Topological insulators; Topological protection and transition, type-I and -II Weyl semimetals

D. Hsieh et al. Science 323, 919 (2009)
J.W. Wells et al. PRL 102, 096802 (2009)
D. Hsieh et al. Nature 460, 1101 (2009)
D. Hsieh et al. PRL 103, 146401 (2009)
D. Hsieh et al NJP 12, 125001 (2010)
S.Y. Xu et al. Science 332, 560 (2011)
S.V. Eremeev et al. Nature Comm. 3:635 (2012)
S.Y. Xu et al. Nature Physics 8 616 (2012)
S.Y. Xu et al. Nature Comm. 3, 1192 (2012)
S. Muff et al. PRB 88, 035407 (2013)
N. Xu et al. PRB 88, 121102(R) (2013)
A. Barfuss et al. PRL 111, 157205 (2013)
G. Landolt et al. PRL 112, 057601 (2014)
N. Xu et al. Nature Comm. 5, 4566 (2014)
F. Pielmeier et al. NJP 17, 023067 (2015)
S.Y. Xu et al. Nature Comm. 6, 6870 (2015)
B.Q. Lv et al. PRL 115, 217601 (2015)
R. Queiroz et al. PRB 93, 165409 (2016)
P. Rüssmann et al. PRB 97, 075106 (2018)
N. Xu et al. PRL 121, 136401 (2018)
A. Weber et al. PRL 121, 156401 (2018)
J. Hu et al. PRB 100, 115201 (2019)

3. **Extra tag in spectroscopy:** photoemission time, orbital mapping, phase determination, spin interference

F. Meier et al. JPCM 23, 072207 (2011)
C. Veenstra et al PRL 112, 127002 (2014)
M. Fanciulli et al PRL 118, 067402 (2017)
E. Razzoli et al. PRL 118, 086402 (2017)
M. Fanciulli et al. PRB 95, 245125 (2017)
M. Fanciulli, and J.H. Dil, SciPost Physics 5, 058 (2018).

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J. Lobo et al. ...02 (201...
I. Gierz et al. ... (2010)
I. Gierz et al PRB 83, 195122 (2011)
P. King et al. PRL 107, 096802 (201...
B. Slomski et al. B 84, 193406(B) (2...
P. Höpfner et al. PRL 108, 186801 (2012)


Fabian Meier

Bartosz Slomski

G. La... 109, 116403 (2012)
C. Te... PRL...
B. Sl... ntific Reports 3, 1963 (2013)
G. La... 15, 08...
B. Sl... 15, 12...
A. Santander-Syro et al Na... 5 (2014)
G. La... 91, 0...
J. H. Dil et al. JESRP 201, ...
C. Brand et al. Nature Com... 5)
J. Krempasky et al. Nature Comm. 7, 13071 (2016)


Stefan Muff

Juraj Krempasky

J. Krempasky et al. PRB 94, 205111 (2016)
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...äger et al. ...22 (2018)
...äger et al. ...olidi B, 1900152 (2019)
D. Krieger et al. ...35 (2019)
J. Krempasky ...13107 (2020)
G. Kremer et al. ...15 (2020)
E.B. Guedes et al. PRR 2, 033173 (2020)


Milan Radovic

Eduardo Guedes

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Gabriel Landolt

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G... PRL 112, 057601 (2014)
N... e Comm. 5, 4566 (2014)


Andrew Weber

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Marco Caputo

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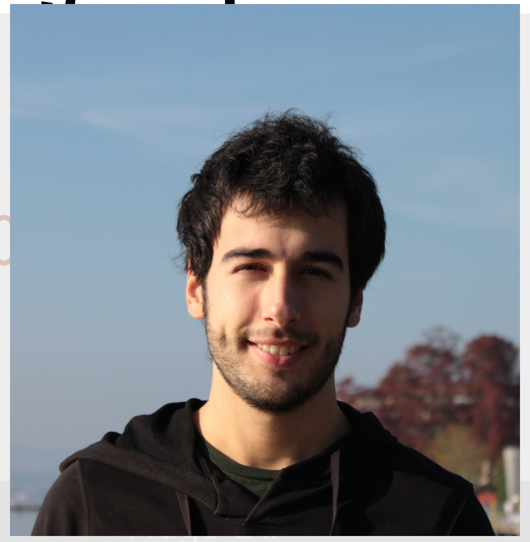
C. Veenstra et al PRL 112, 12700...


Mauro Fanciulli

M. Fanciulli et al PRL 118, 067...
E. Razzoli et al. PRL 118, 0864...

M. Fanciulli et al. PRB 95, 245125 (2017)
M. Fanciulli, and J.H. Dil, SciPost Physics 5, 058 (2018).

Many others…


Unsolicited advise:
don’t aim for best machine, but be versatile
and flexible, especially in sample
preparation and environment