# « ARPES and topology »


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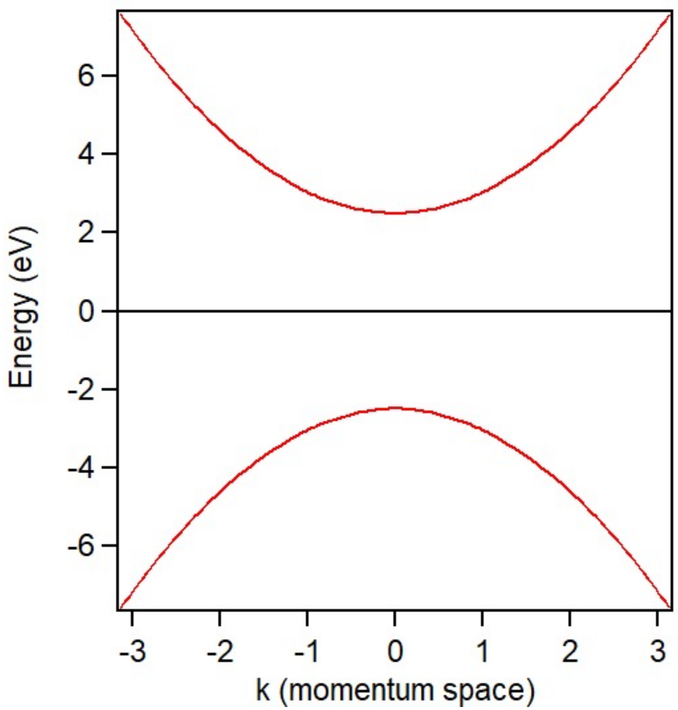
# « ARPES and topology »

ARPES is one of the most « direct » to visualize electron bands
But topology is in the wave function (not the dispersion)…

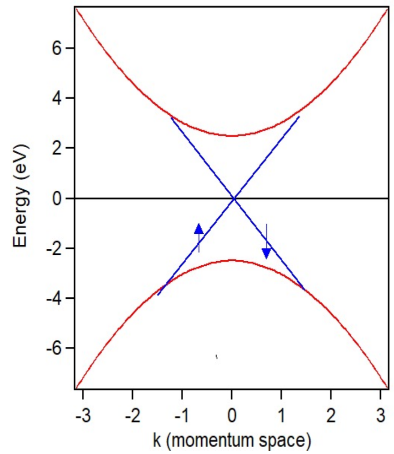
A gap….




Is it topological or not ?
=> E(k) can be measured by ARPES but it is not sufficient

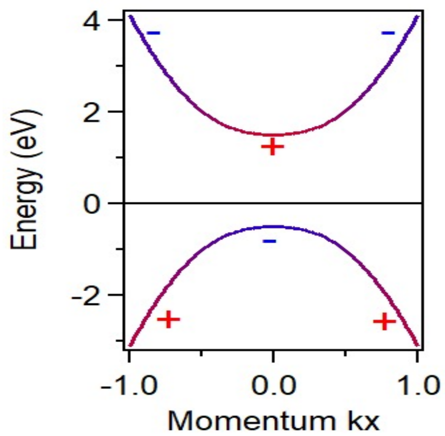
- In Topological insulator, Topological Surface state inside the gap

⇒ Dirac cone with spin-momentum locking can be imaged by ARPES




- Direct probe of band inversion ?

⇒ Somewhat possible using polarization, but not always easy to interpret !!



# Outline

- Photoemission principle

  Photoelectric effect + conservation laws (energy, momentum)
  
  => 3D electronic structure + surface states
  
  *Topological surface states, spin resolution*
  
  *Weyl semimetal, Fermi arcs*

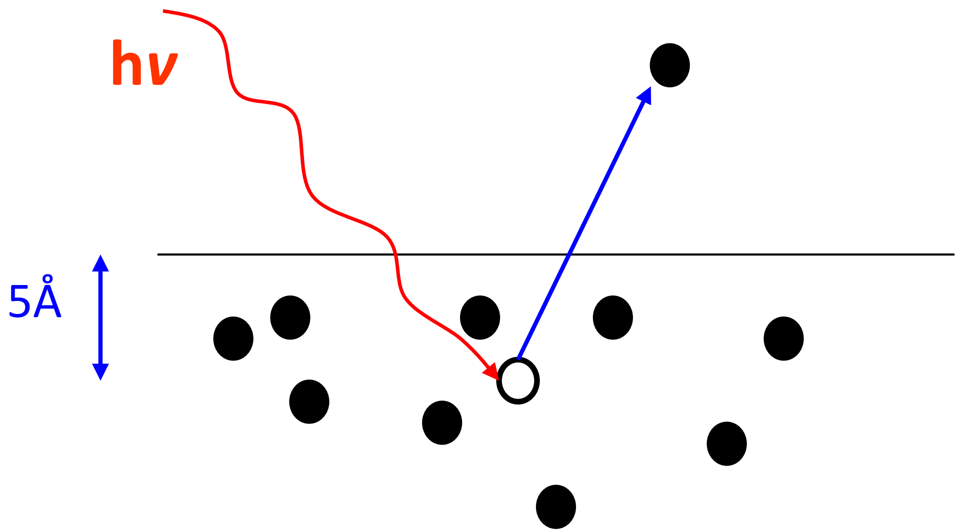
- Beyond band dispersion : role of light polarization

  Light-matter interaction
  
  Band parity
  
  Berry phase (graphene)
  
  Pb of photoelectron final state !

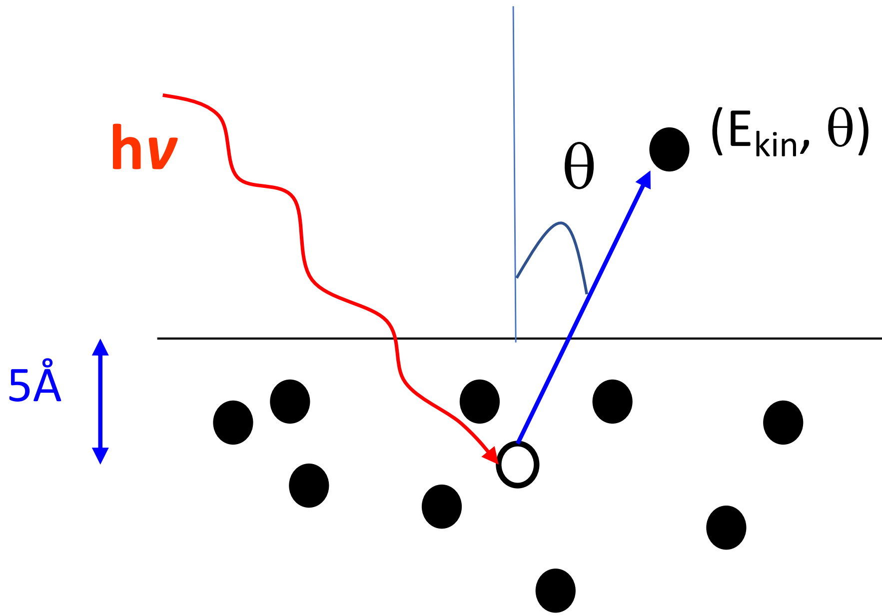
- How can it be useful ?

  Topological transition between trivial and non-trivial insulators
  
  Correlated cases : Kondo insulators, kagome metals

# A photoemission experiment



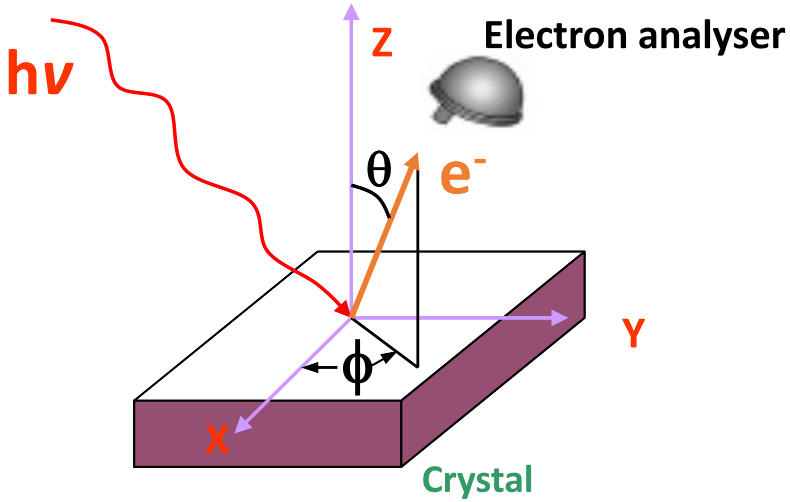
# A photoemission experiment



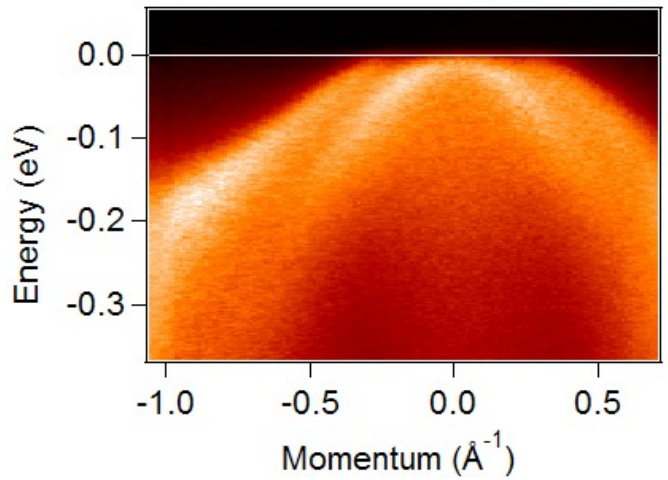

Conservation of energy $$E_{kin} = h\nu - W - |E_B|$$

Conservation of momentum parallel to surface $$\hbar \mathbf{k}_{\parallel} = \sqrt{2mE_{kin}}\,\sin\theta$$

# Angle-resolved photoemission

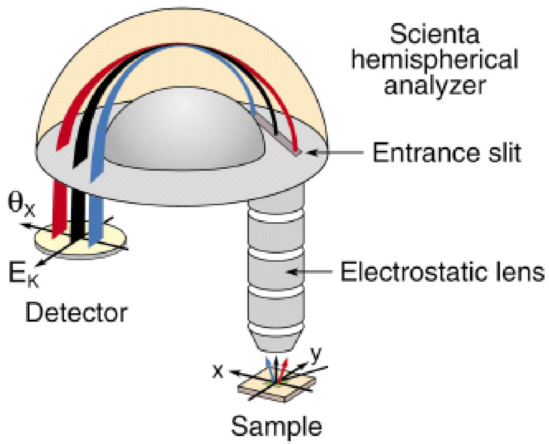



*Hole pockets in $Ba(Fe_{0.92}Co_{0.08})As_2$*




## 1- Energy conservation:

$$E_{kin} = h\nu - W - |E_B|$$




## 2- Momentum conservation

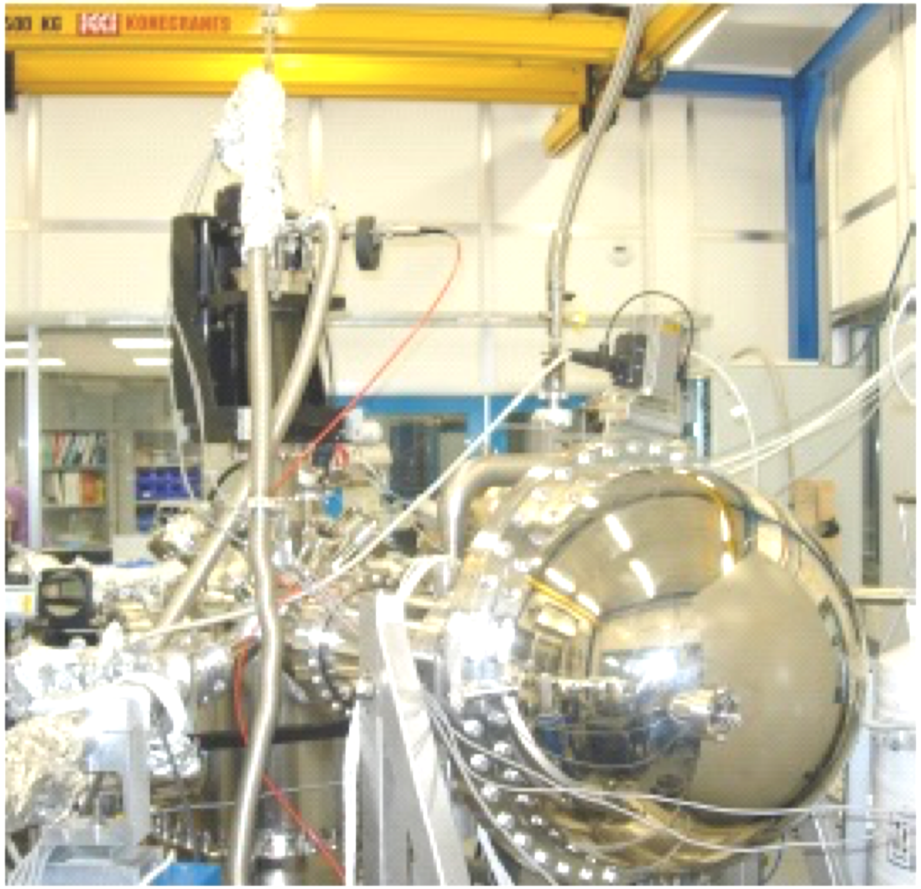
$$\hbar \mathbf{k}_{\|} = \sqrt{2mE_{kin}} \sin\theta$$

$\Rightarrow E_{kin}$ vs $k_{//}$ : Structure de bandes

NB : $k_{perp}$ is not conserved

# Experimentally…

CASSIOPEE beamline, SOLEIL synchrotron

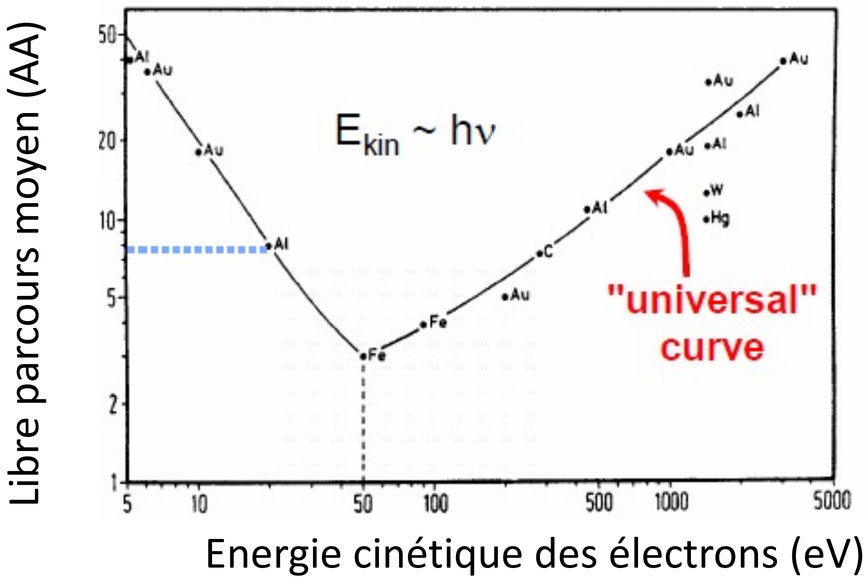



$$E_{kin} = h\nu - W - |E_B|$$

$$\hbar \mathbf{k}_{\|} = \sqrt{2mE_{kin}} \sin\theta$$

**Surface sensitive !!**

=> Ultra-high vacuum, good surface !




Photons from : Synchrotrons : 10-1000eV

He lamp : 21 eV

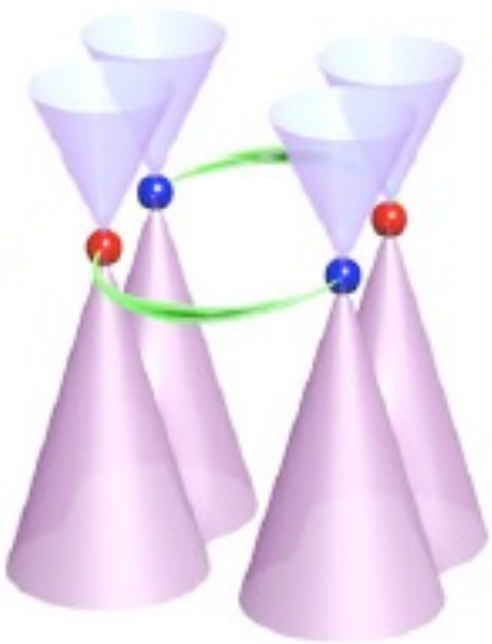
Laser : 6-7eV (extending…)

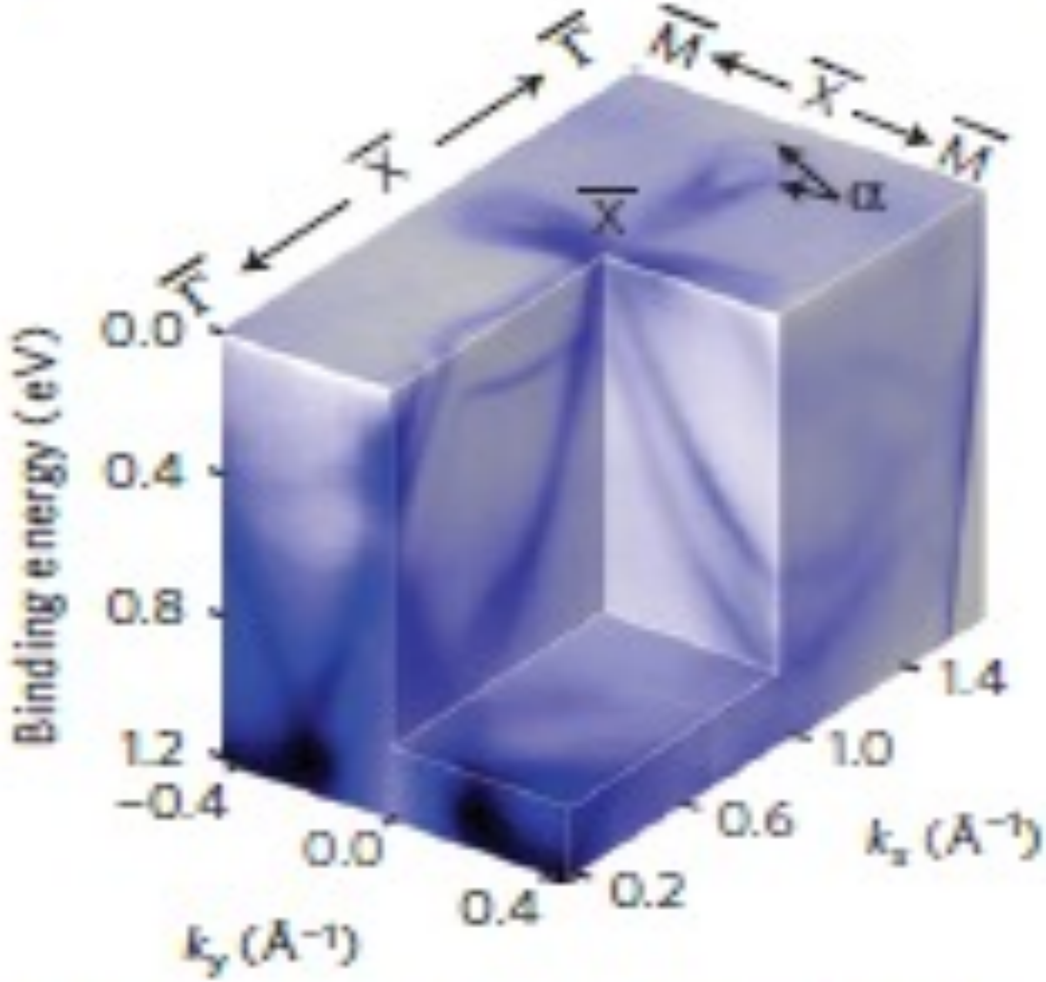
# 3D data set I(E, kx, ky)

## Weyl semi-metal TaAs

*Yang Nature Physics 2015*

Fermi arcs in Weyl semimetal

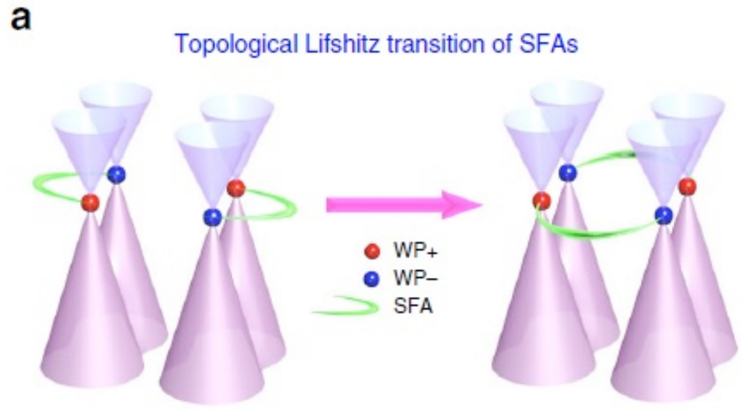


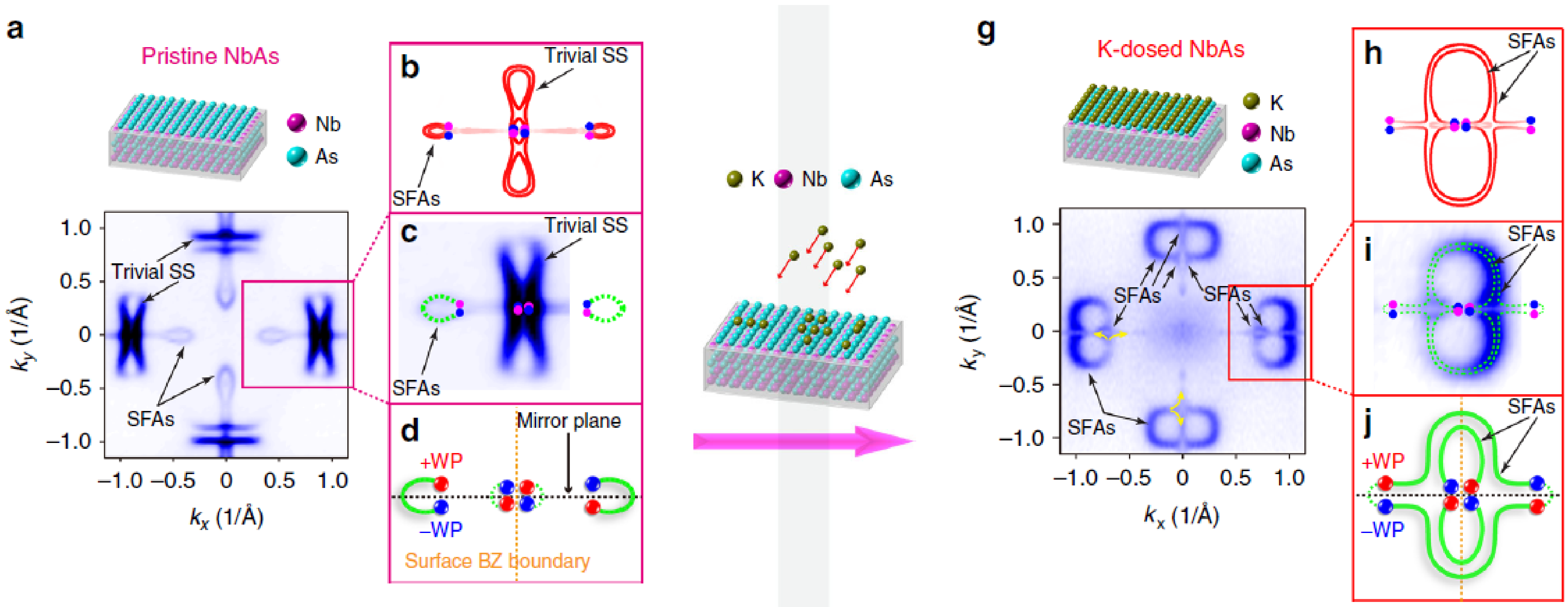


# « Topological Lifshitz transition »

Weyl semimetal NbAs

*H.F. Yang, Y.L. Chen, Nature Com. 2019*



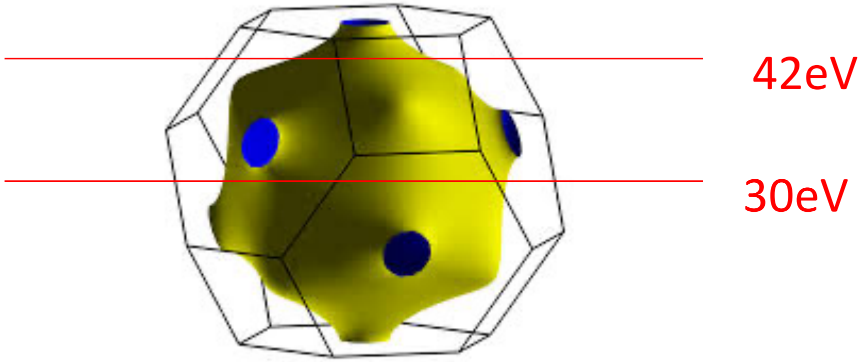




=> Manipulation of connectivity of Fermi arcs by altering surface potential

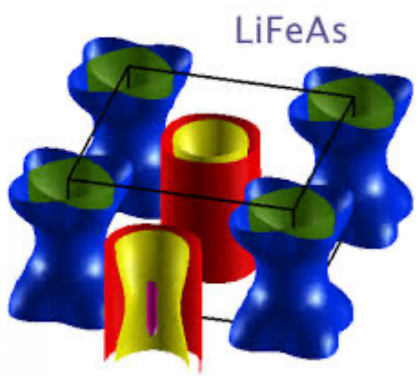
# Perpendicular momentum ?

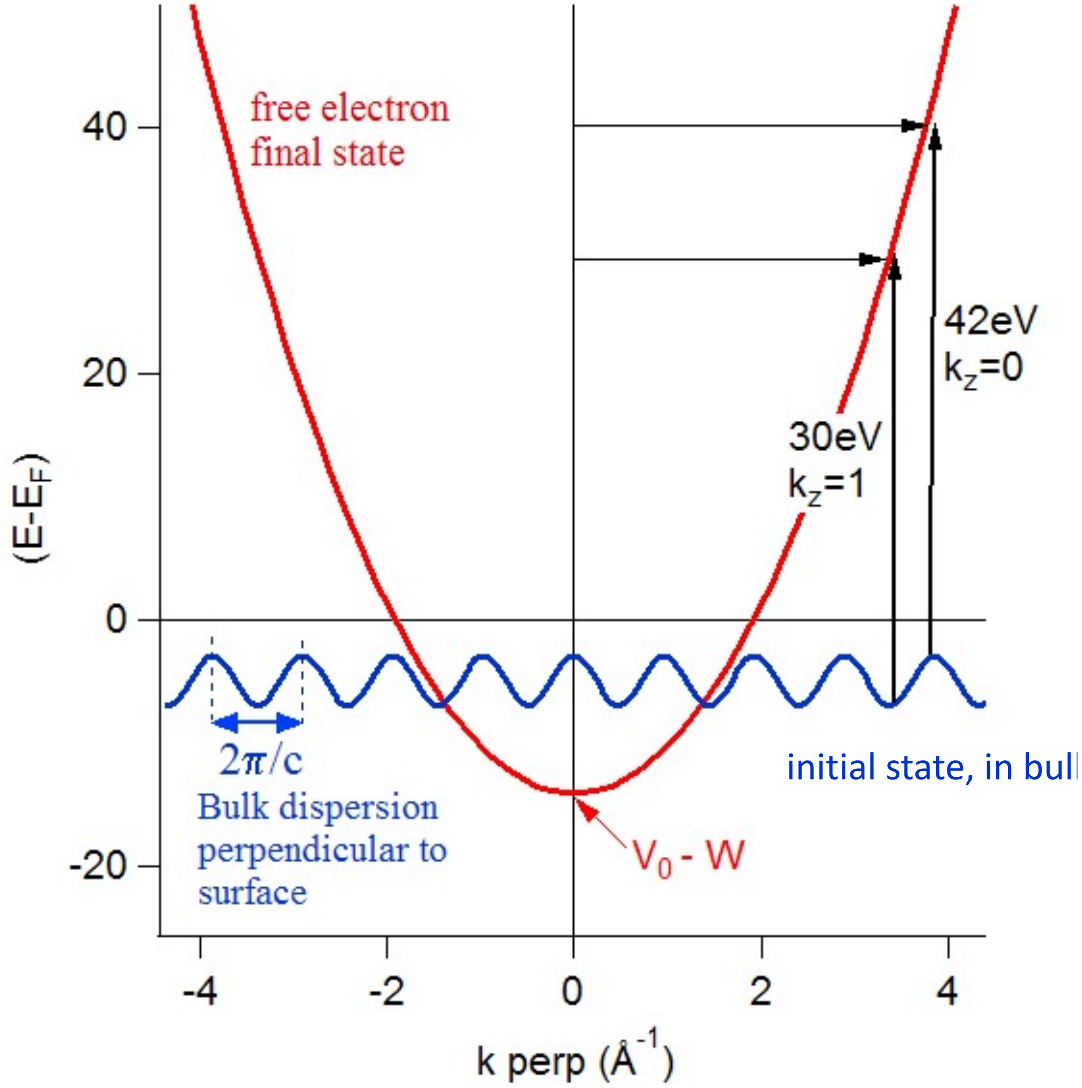
I (E, $k_x$, $k_y$, $k_z$ ?)

3D Fermi Surface



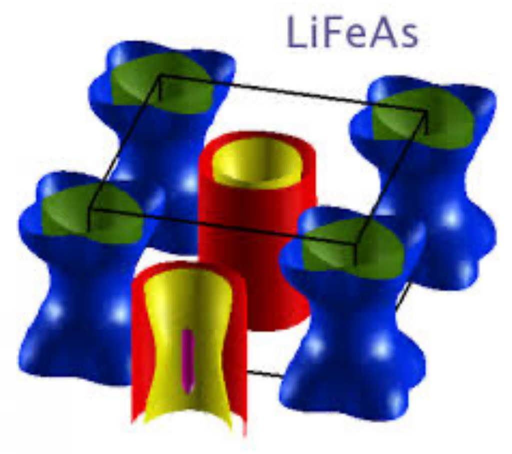

Quasi-2D Fermi Surface



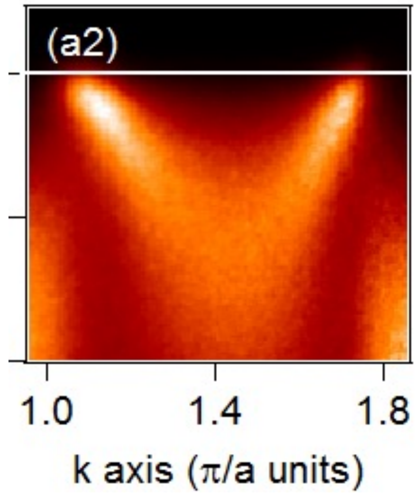



# Example of 3D dispersion

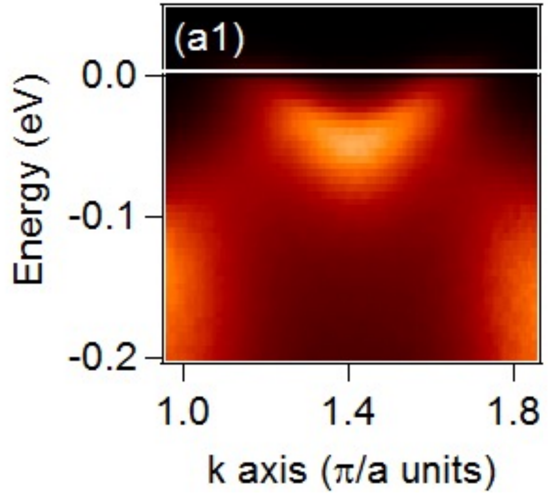
Quasi-2D Fermi Surface



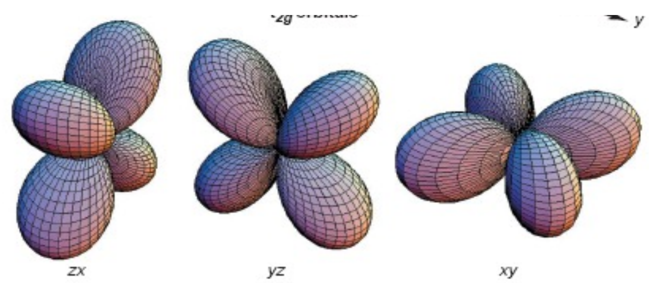

dxy

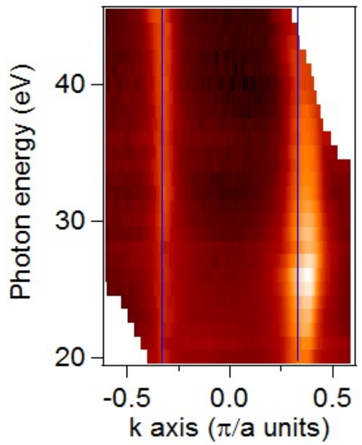



dxz/dyz

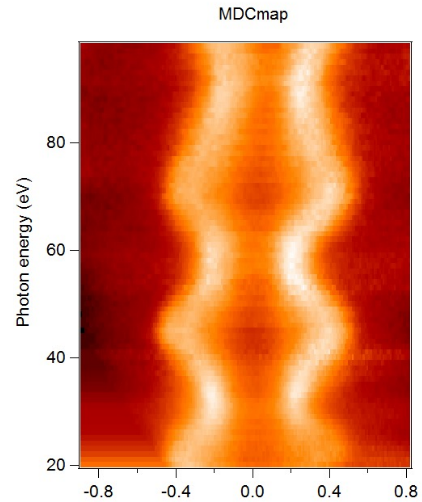



dxz dyz dxy



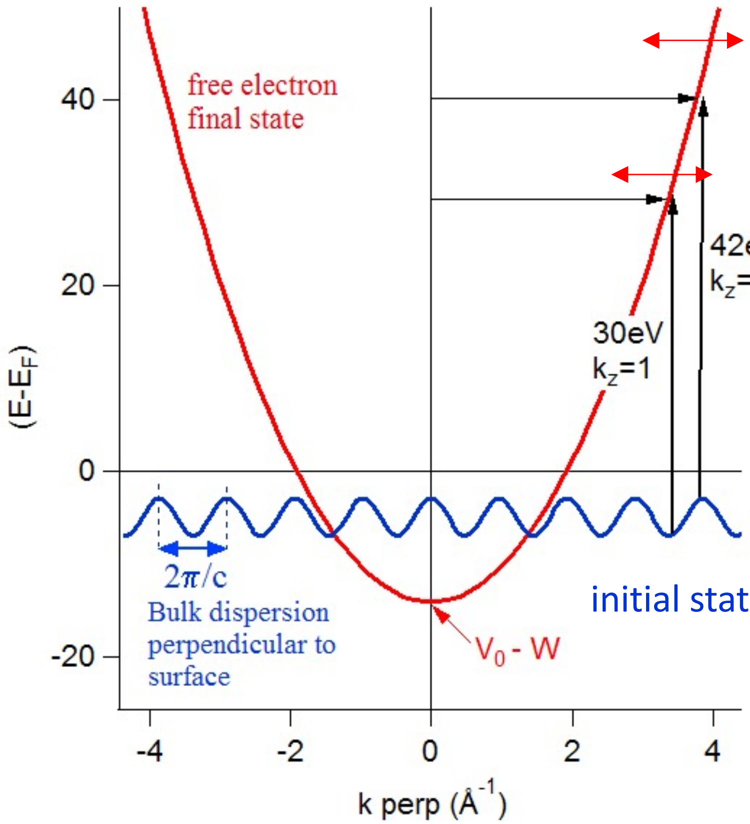

=> 2D band




=> Significant warping

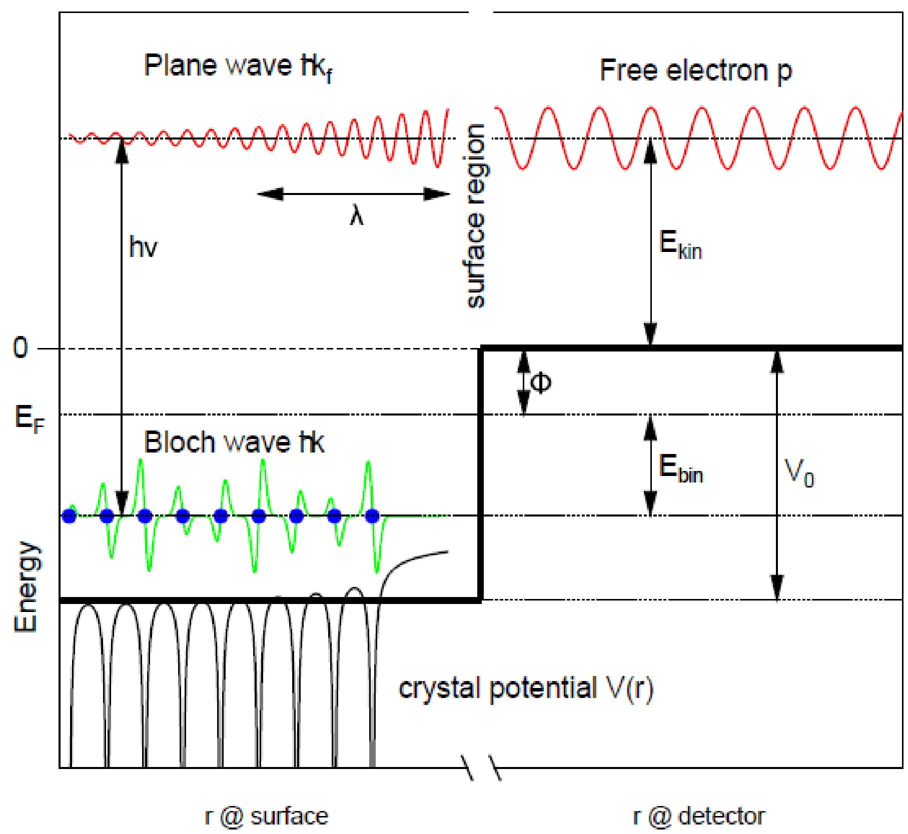
# Perpendicular momentum ?

$\delta k_{perp} \sim 1/\lambda$



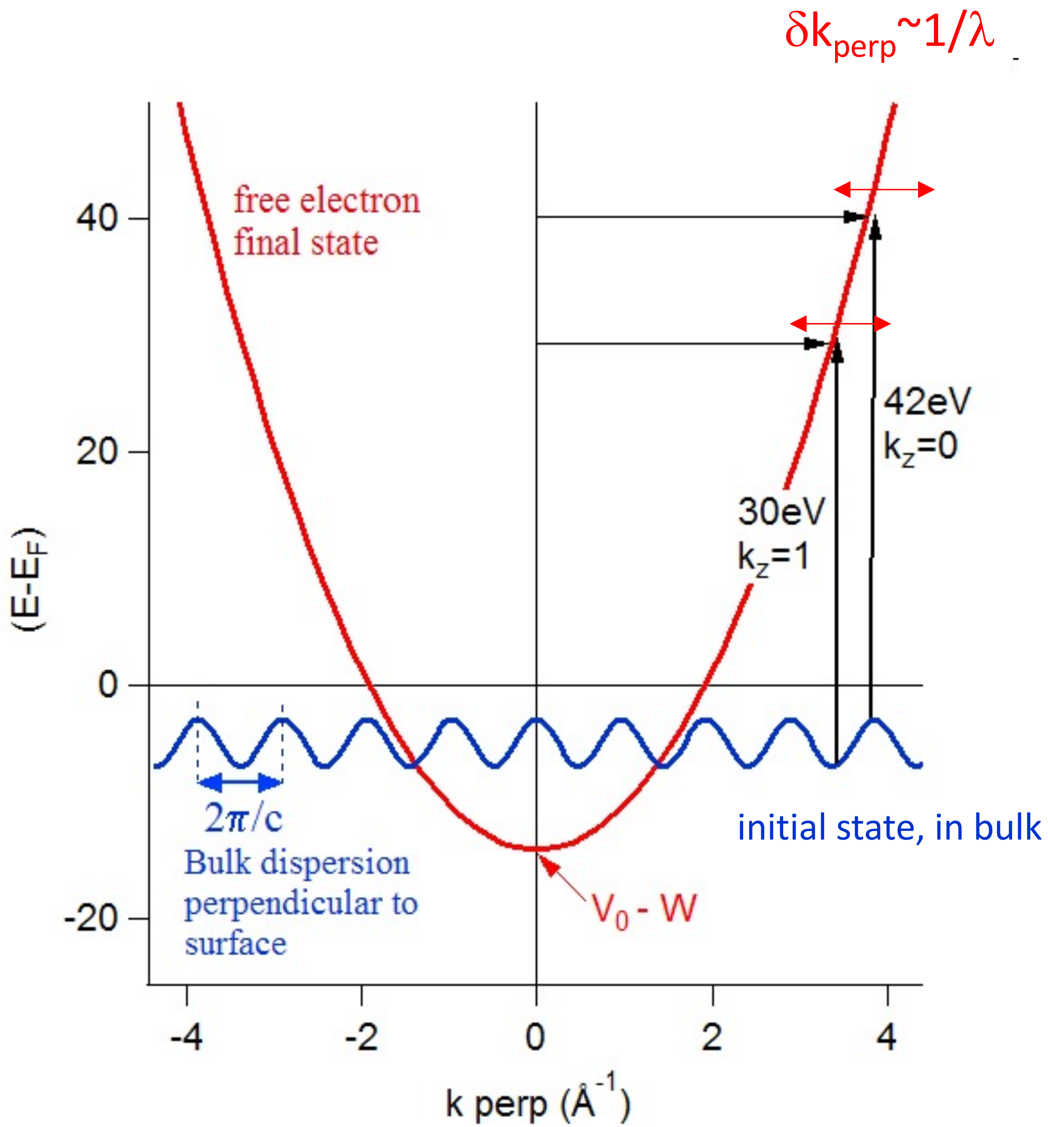

initial state, in bulk

Steps of the photoemission process

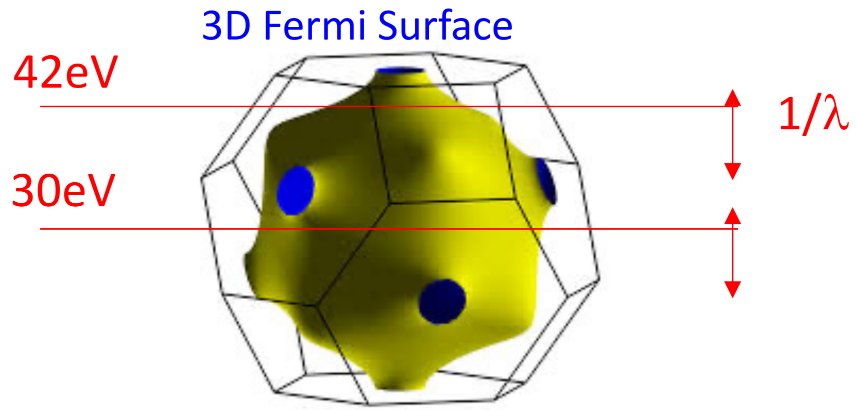


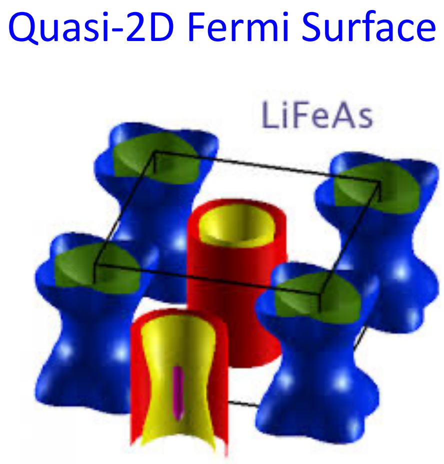

*Moser, J. electron spectroscopy 2016*

# Perpendicular momentum ?



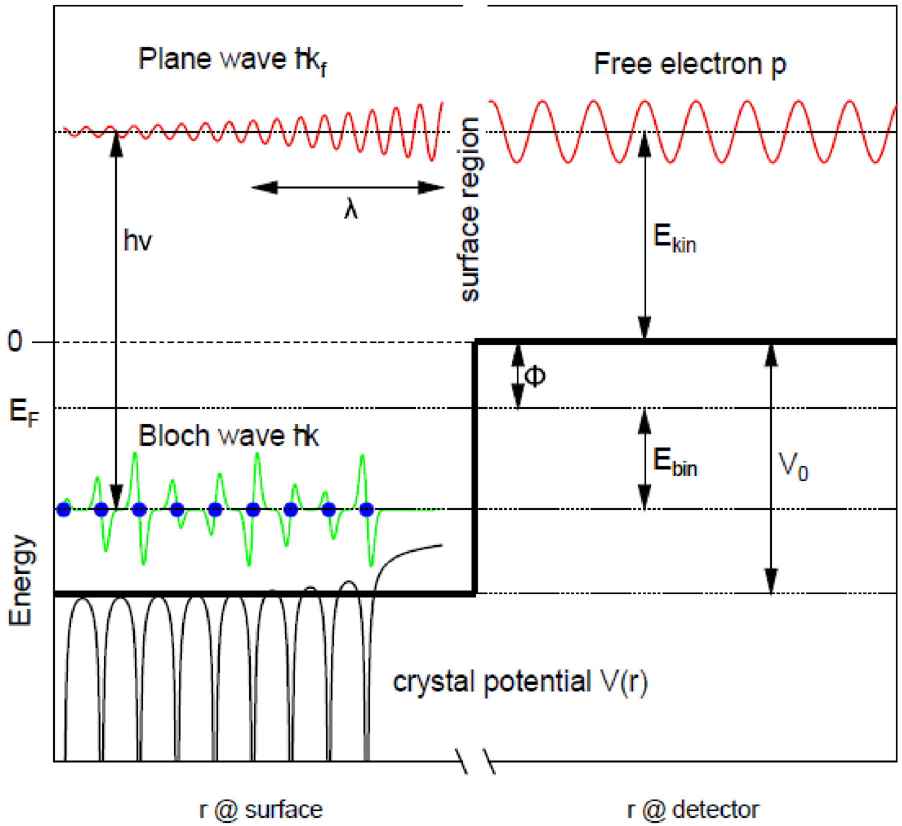

$I\ (E, k_x, k_y, k_z\ ?)$



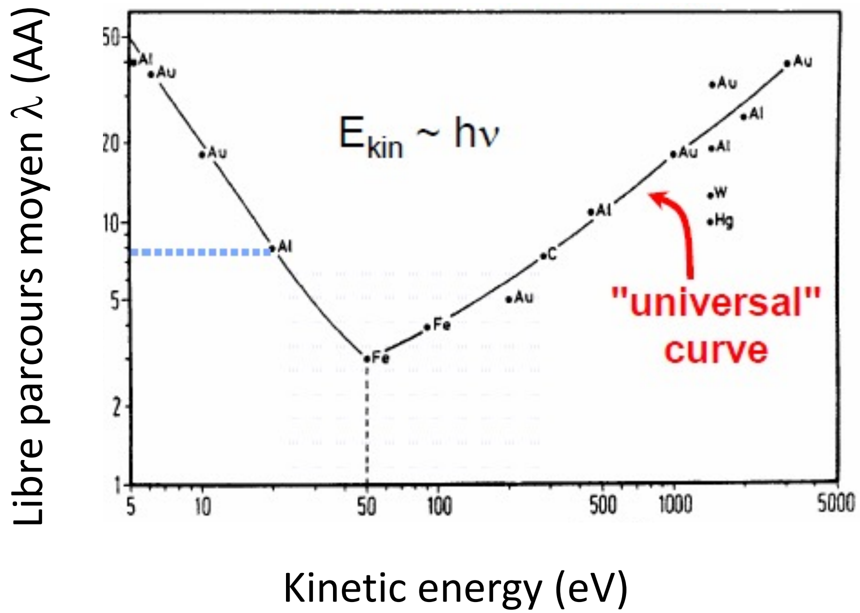
# How to increase the definition of $k_{perp}$ ?

Steps of the photoemission process




**Surface sensitive !!**

=> Ultra-high vacuum

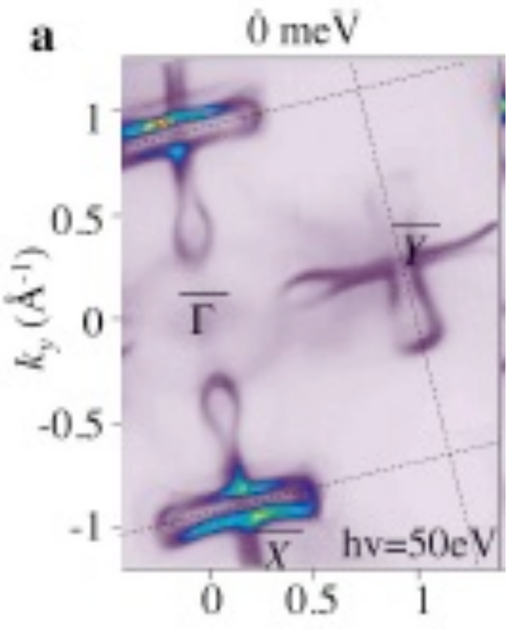



*Moser, J. electron spectroscopy 2016*

# Using different photon energy to increase bulk sensitivity

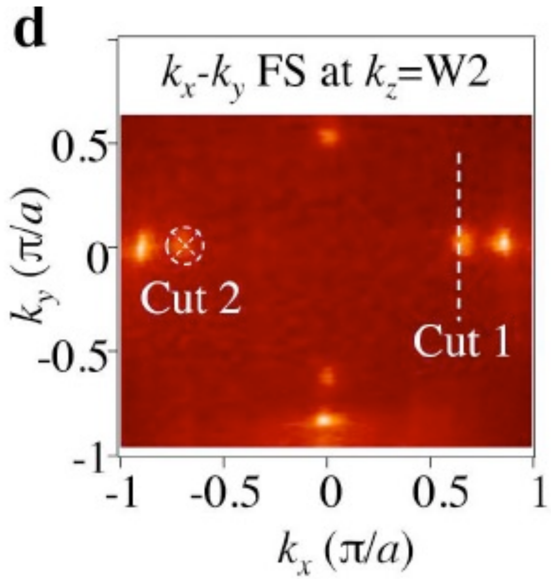
## Example of Weyl semi-metal NbAs

### 50eV : Fermi arcs

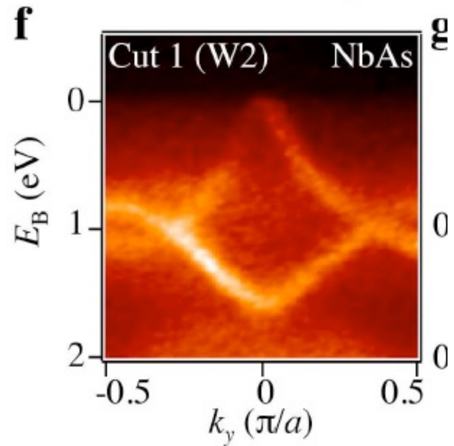



### 651eV : Weyl nodes

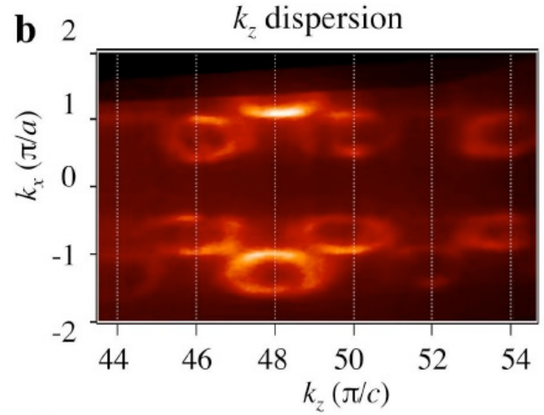
Fermi Surface (kx,ky)




Dispersion along ky




Dispersion along kz



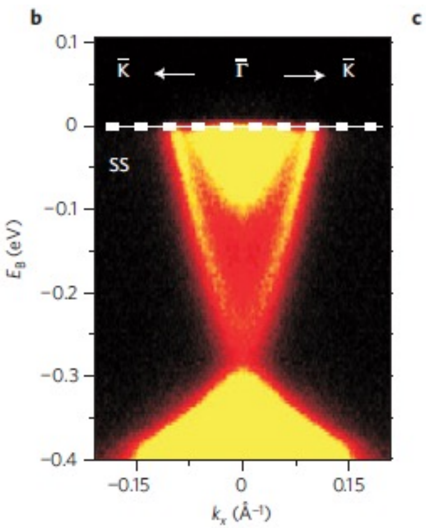

*Xu, Hasan, Cond-mat2015*

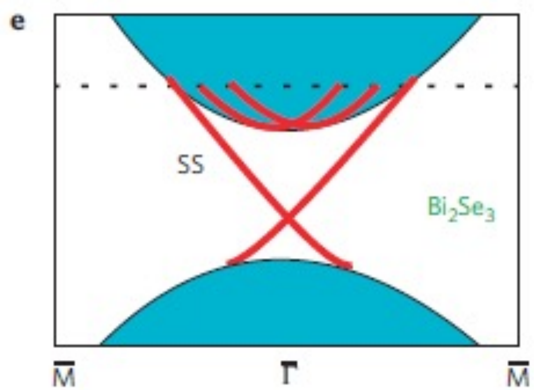
# Topological surface states

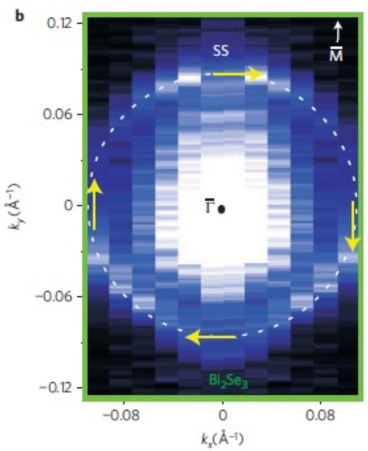
## Observation of a large-gap topological-insulator class with a single Dirac cone on the surface

Y. Xia[1,2], D. Qian[1,3], D. Hsieh[1,2], L. Wray[1], A. Pal[1], H. Lin[4], A. Bansil[4], D. Grauer[5], Y. S. Hor[5], R. J. Cava[5] and M. Z. Hasan[1,2,6]★

$Bi_2Se_3$




*Xia et al. Nature Physics 2009*

Bulk band : strongly 3D, look like continuum
Surface states : much better defined

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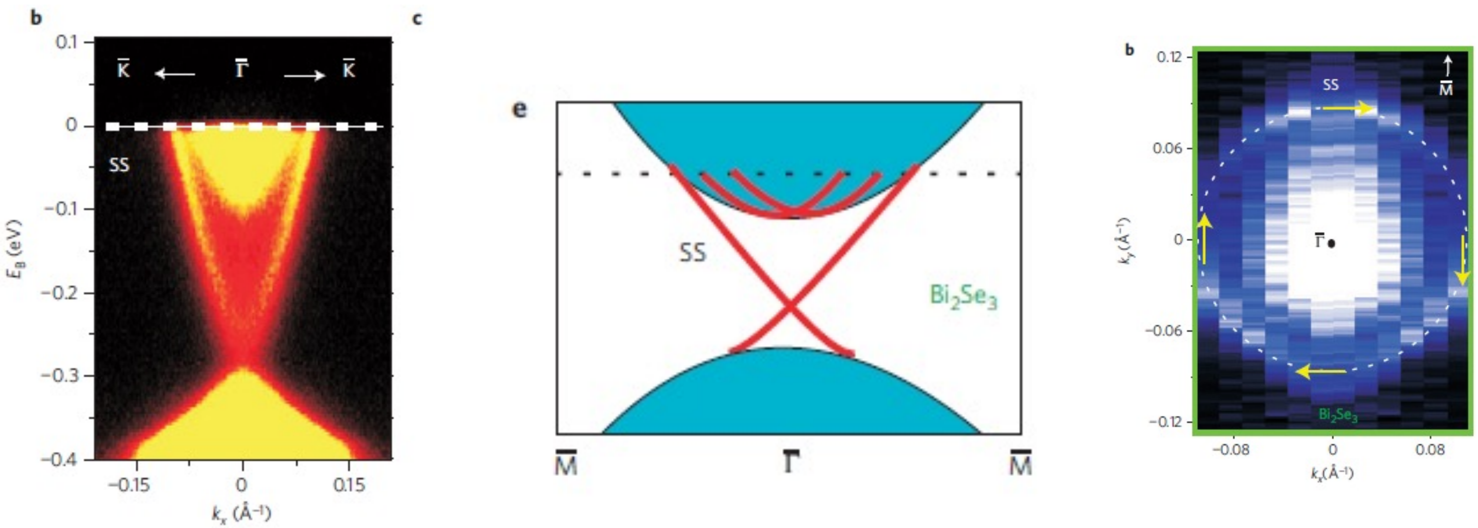
$Bi_2Se_3$




*Xia et al. Nature Physics 2009*

Bulk band : strongly 3D, look like continuum
Surface states : much better defined

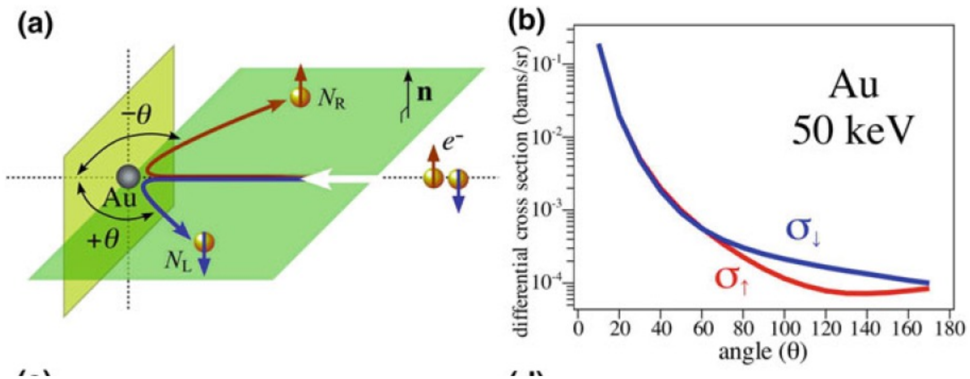
# Spin resolution

Applying a magnetic field would deviate electrons and destroy information about k !
Other means are used to detect spin, but very inefficient.

## Mott scattering

Relativistic effect : anisotropy in reflection depending on spin (efficiency $10^{-4}$)
electrons accelerated to relativistic speed on Au target

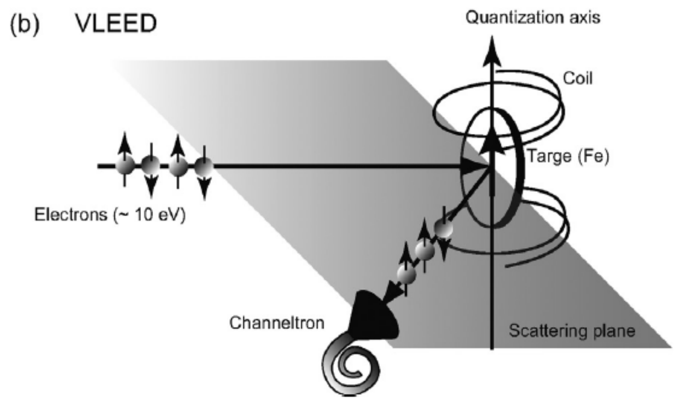
COPHEE (SLS)
Beginning of years 2000




## Exchange scattering (VLEED)

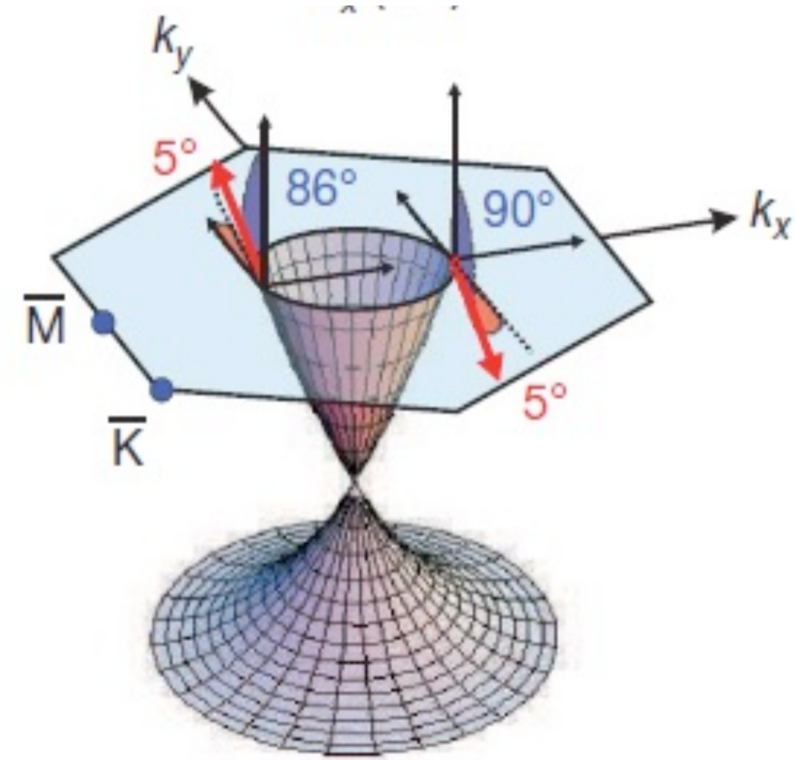
Reflection of low energy electrons on a ferromagnetic target.
More efficient but requires regular maintenance of the target

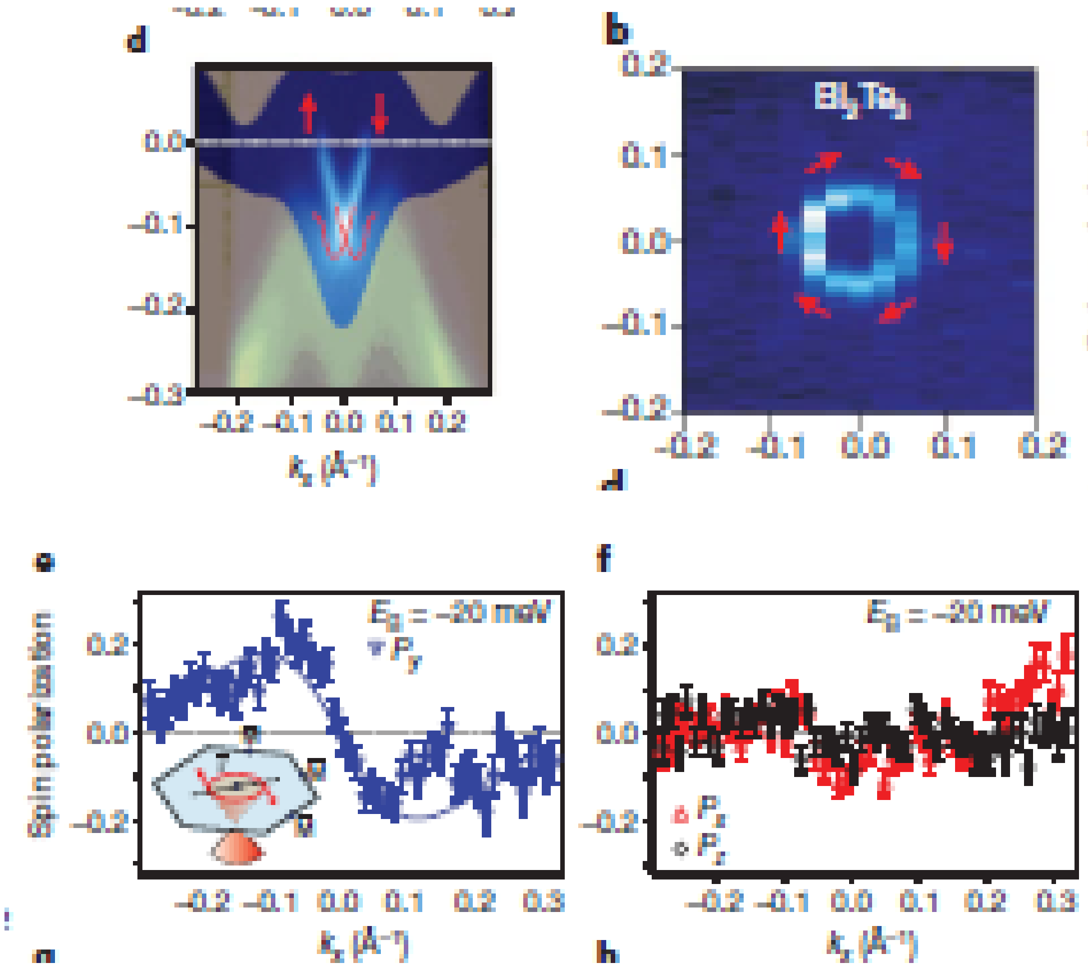
ESPRESSO (Hiroshima)



# Spin-resolved ARPES on surface states of topological insulators

$Bi_2Te_3$



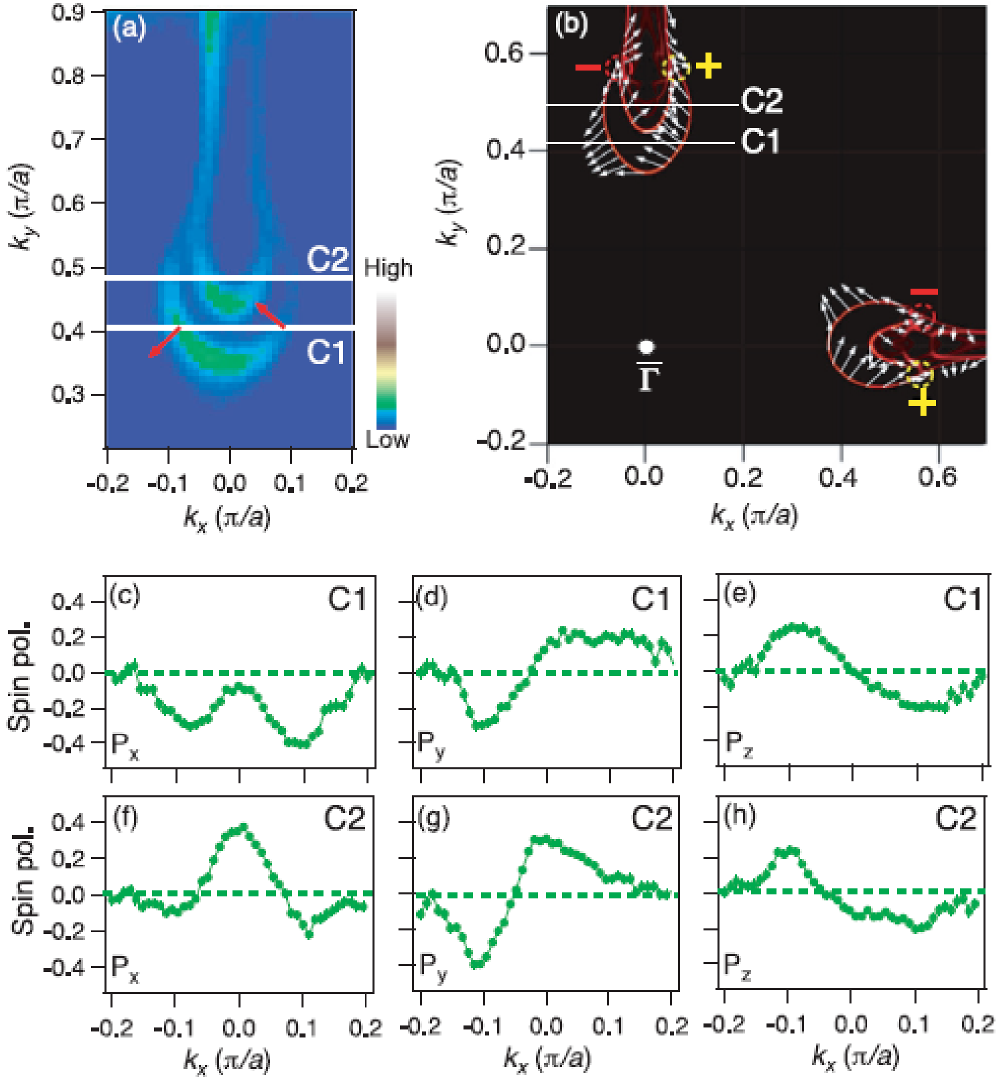




*Hsieh, Hasan Nature 09*

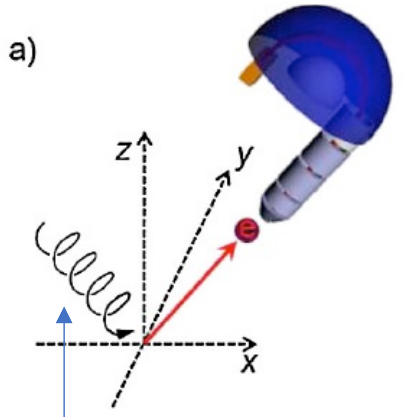
# 3D detection of the spin

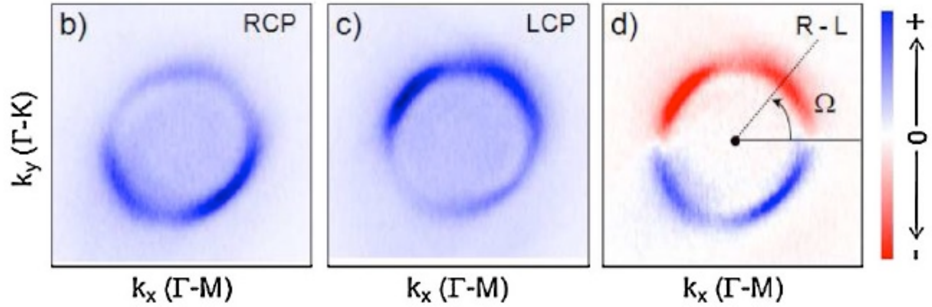
Fermi arcs in TaAs

*Lv PRL15*



# Is circular dichroism a « quick way » to get spin polarization ?




Circular polarisation : LCP or RCP

Circular Dichroism :

CD = I(RCP) – I(LCP)

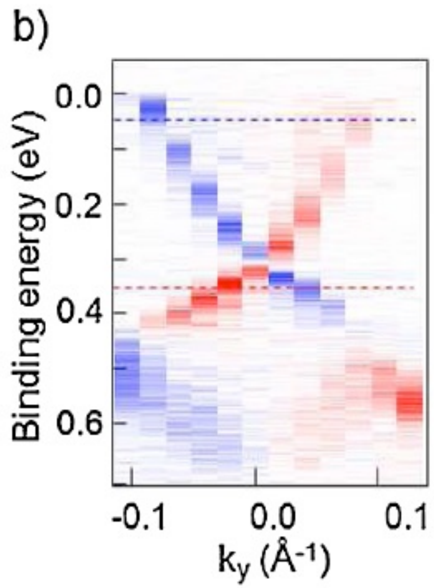
CD ~ $\vec{l}.\ \overrightarrow{k_{ph}}$

Orbital Angular Momentum

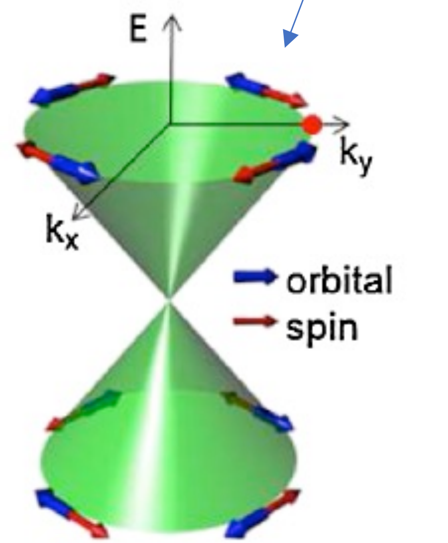
Related to Berry curvature :

$$\ell^z_{v\sigma}(\mathbf{k}) = -\frac{m}{\hbar}(\varepsilon_{\mathbf{k}c} - \varepsilon_{\mathbf{k}v})\,\Omega_{v\sigma}(\mathbf{k})$$

*Schuler Science Adv. 2020*

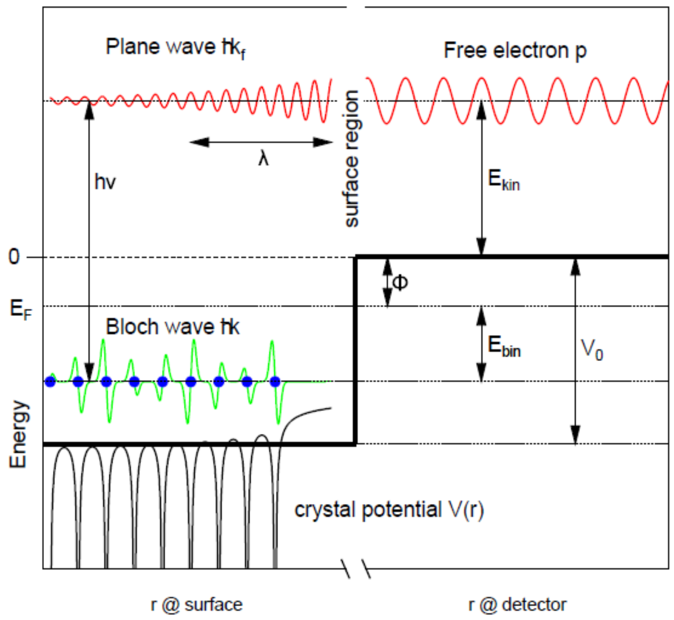



Locked by strong SOC

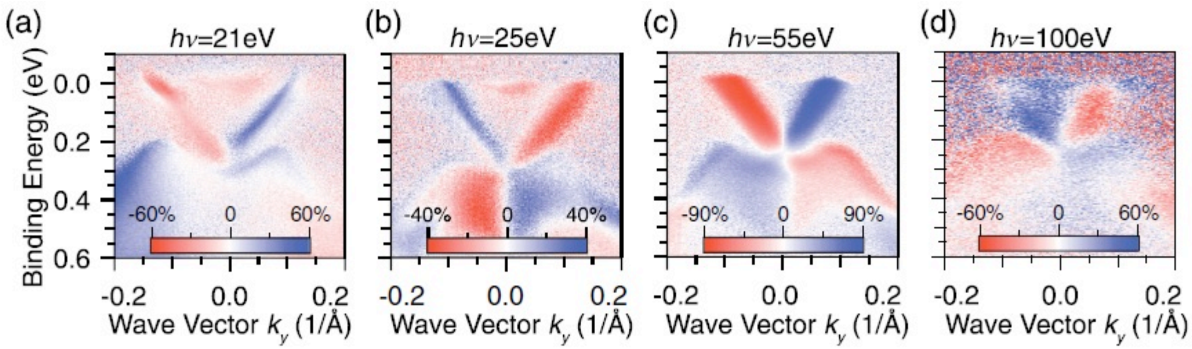



*Park PRL12*

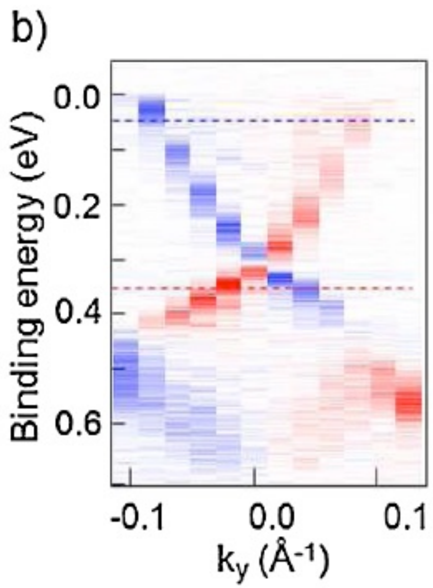
# Warning : this information can be masked by « final states effect »



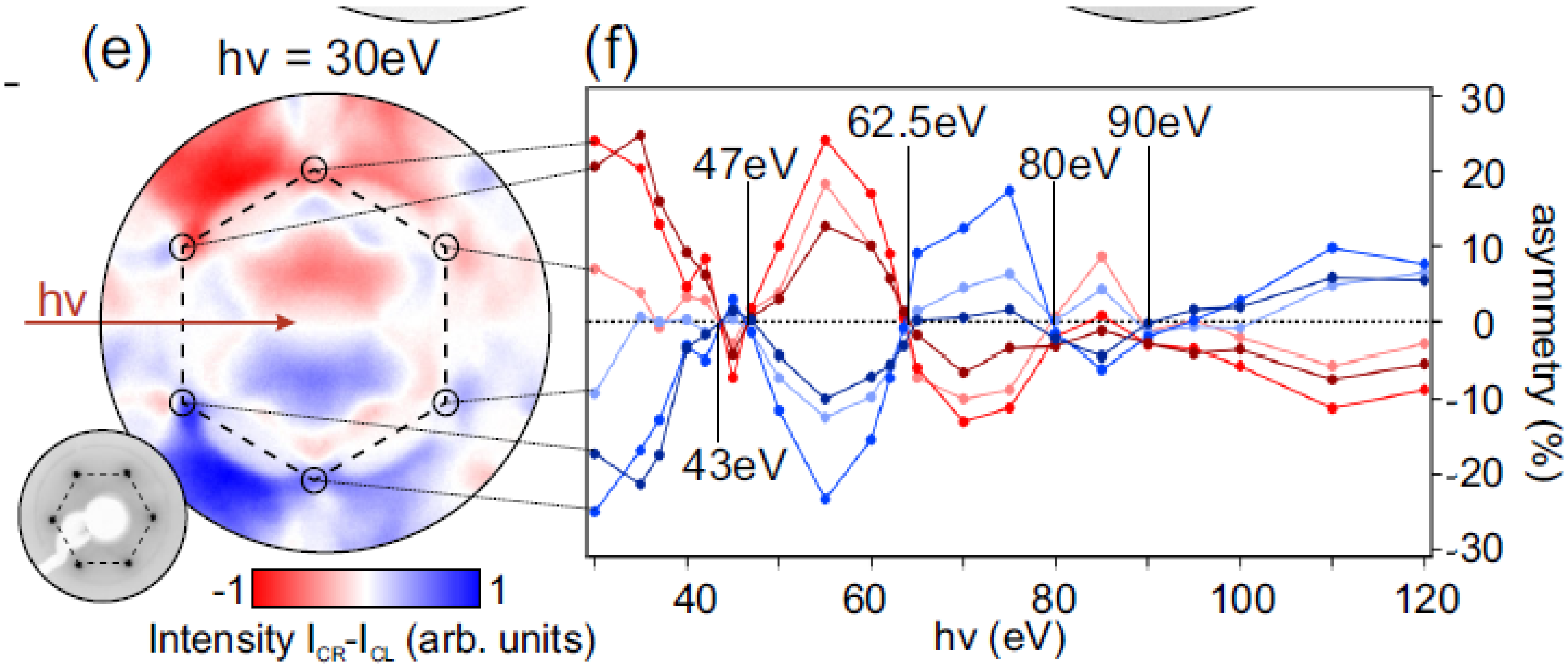

Circular dichroism in $Bi_2Te_3$ as a function of photon energy




*Scholz PRL13*



# Circular dichroism vs photon energy



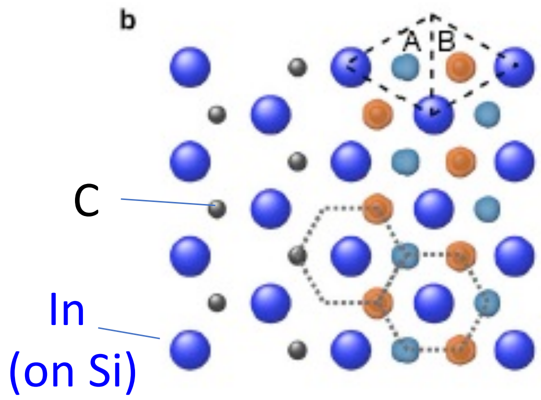

Specific photon energies can be used to probe reliably circular dichroism

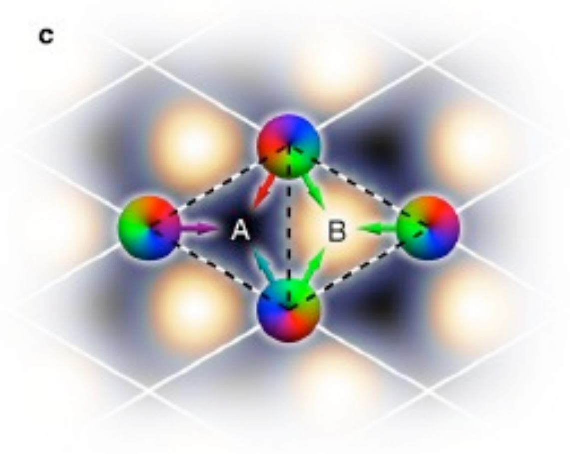
*Erhardt, R. Claessen, S. Moser PRL24*

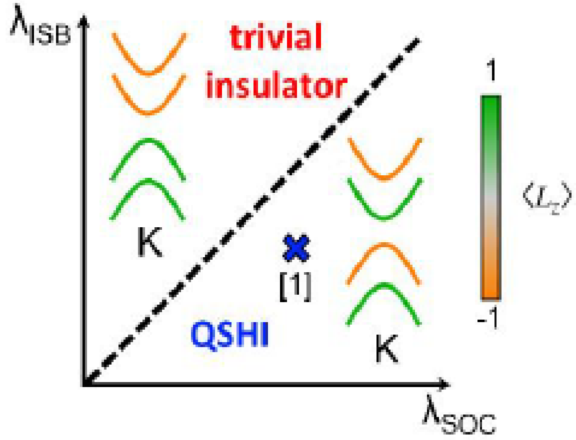
# « Direct » proof of band inversion

Monolayer of In/SiC : Indenene.

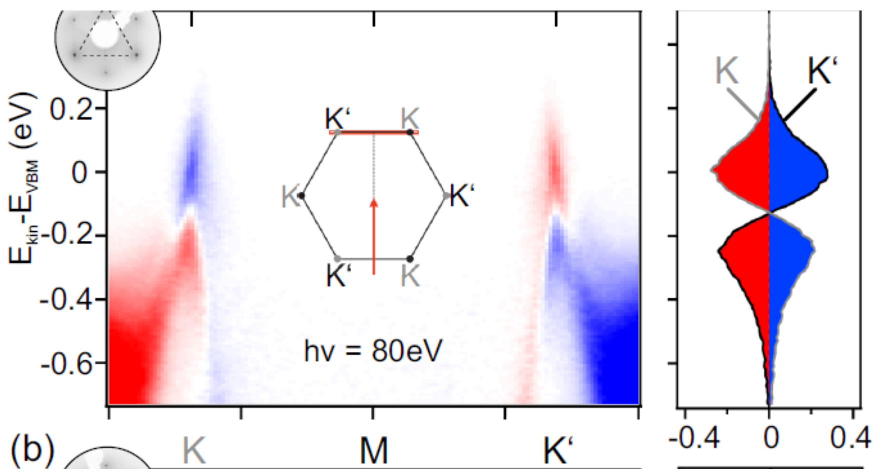
Triangular arrangement, but 2 inequivalent sites A/B : this breaks inversion symmetry.

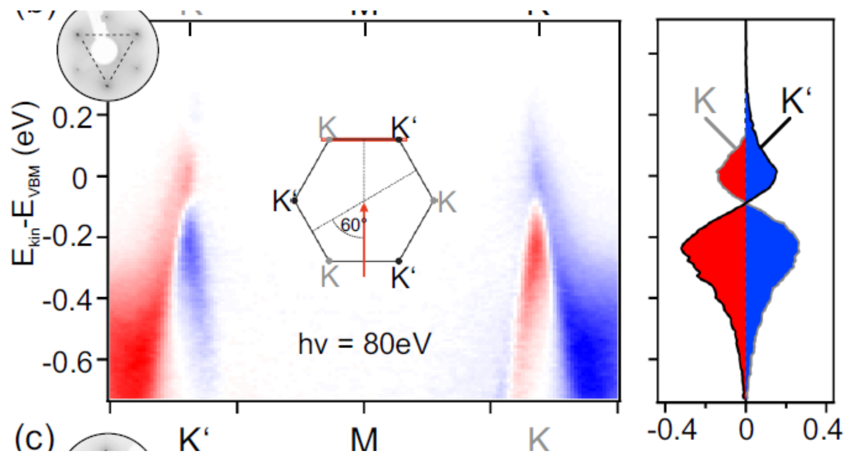



Circular dichroism

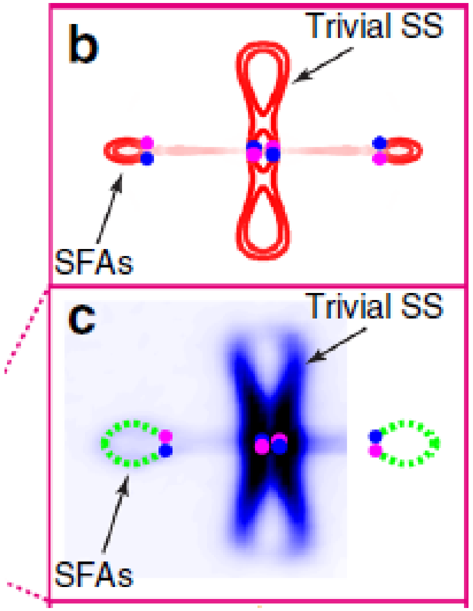


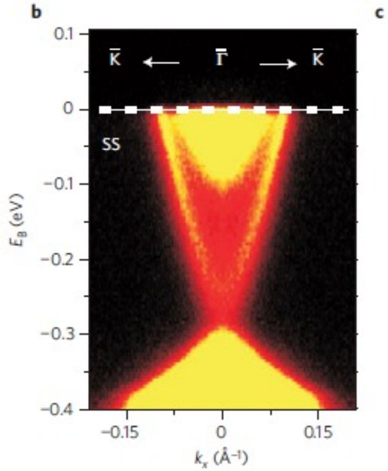
# Summary of part 1

ARPES can map the 3D band structure, including trivial and topological surface states.
High surface sensitivity of ARPES is an advantage to study topological materials !

## Fermi arcs in NbAs



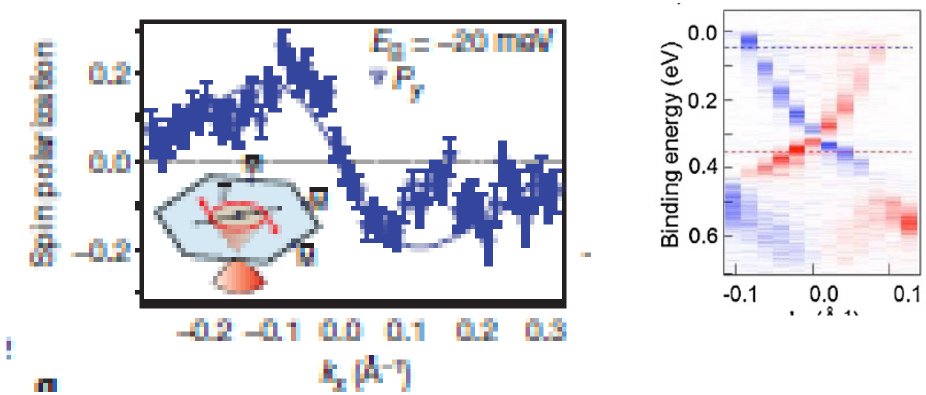

## Topological Surface State in $Bi_2Se_3$




*Evidence for spin-momentum locking*

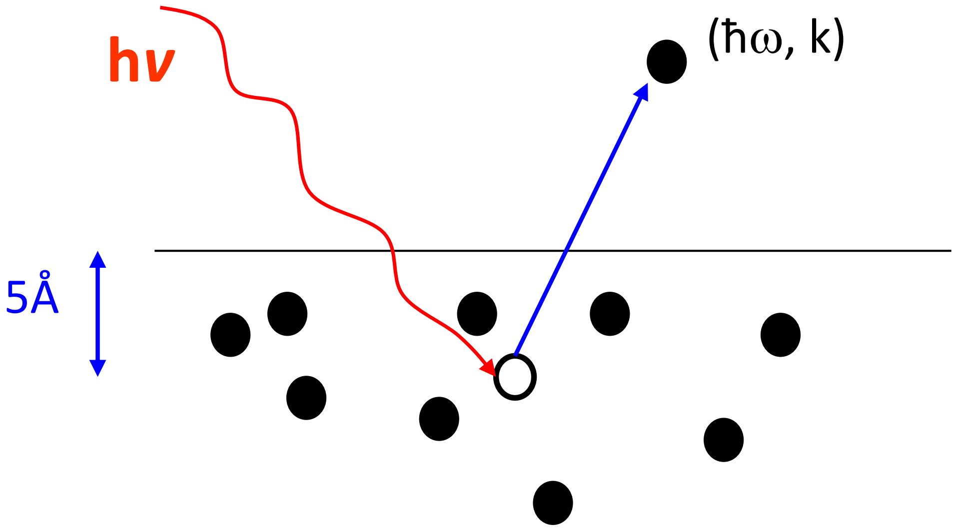
Spin polarization

Circular light




Can we go deeper into the nature of the observed states ?

# Deeper into the photoemission process

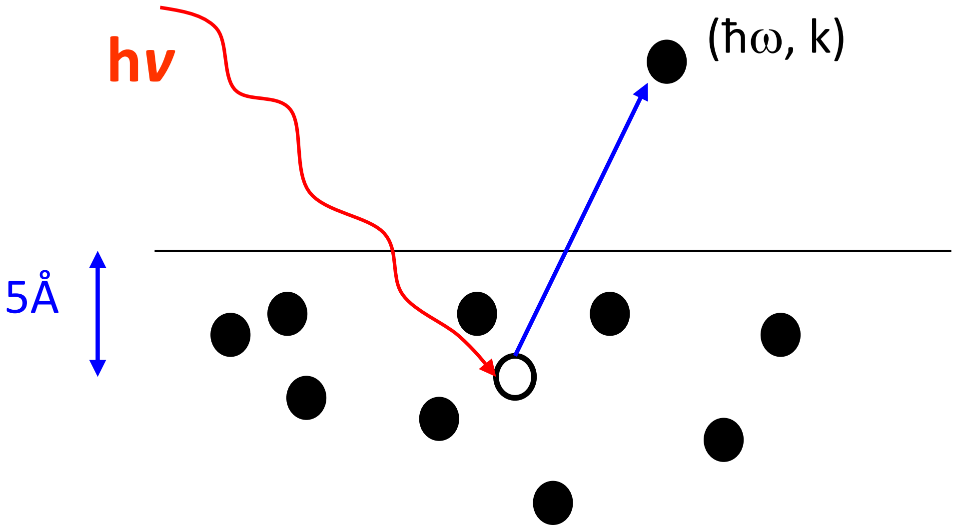



$$I(k,\omega)=\sum_{i,f}\frac{2\pi}{\hbar}\left|\left\langle \psi_f^N \middle| H_{\text{int}} \middle| \psi_i^N \right\rangle\right|^2 \delta(E_f^N - E_i^N - h\nu)$$

N-1 électrons
in interaction
+ photoemitted electron (ħω, k)

N electrons
in interaction

# Deeper into the photoemission process



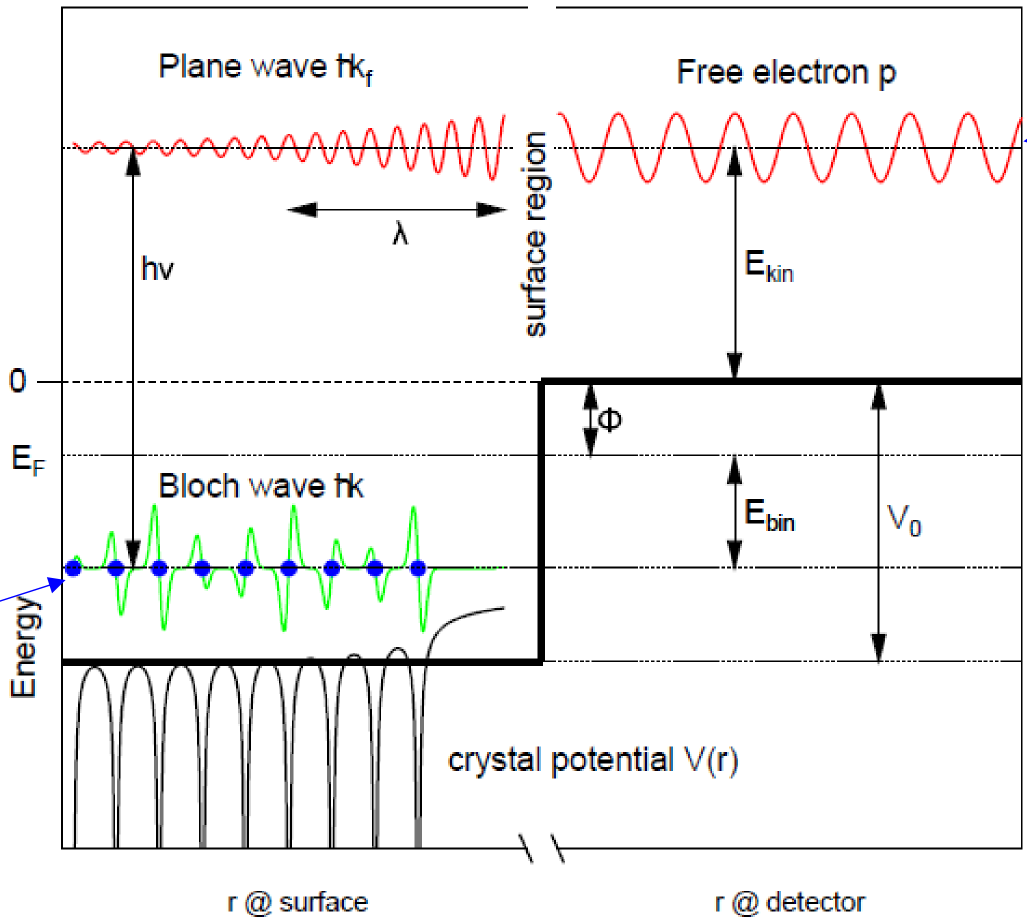

Assumes independent particles

$$I(k,\omega) = \sum_{i,f} \frac{2\pi}{\hbar} |\langle \varphi^{k}{}_{f} | H_{int} | \varphi^{k}{}_{i} \rangle|^2 \, \delta(E_f - Ei - h\nu)$$

« One electron » wave function

# One electron matrix element

$$I(k,\omega) = \sum_{i,f} \frac{2\pi}{\hbar} |\langle \varphi_f | H_{int} | \varphi_i \rangle|^2 \, \delta(E_f - E_i - h\nu)$$




$\phi_f^k$

$\phi_i^k$

- Final state = plane wave ?

- Interaction hamiltonian ?

$$\vec{p} \rightarrow \vec{p} - e\vec{A}$$

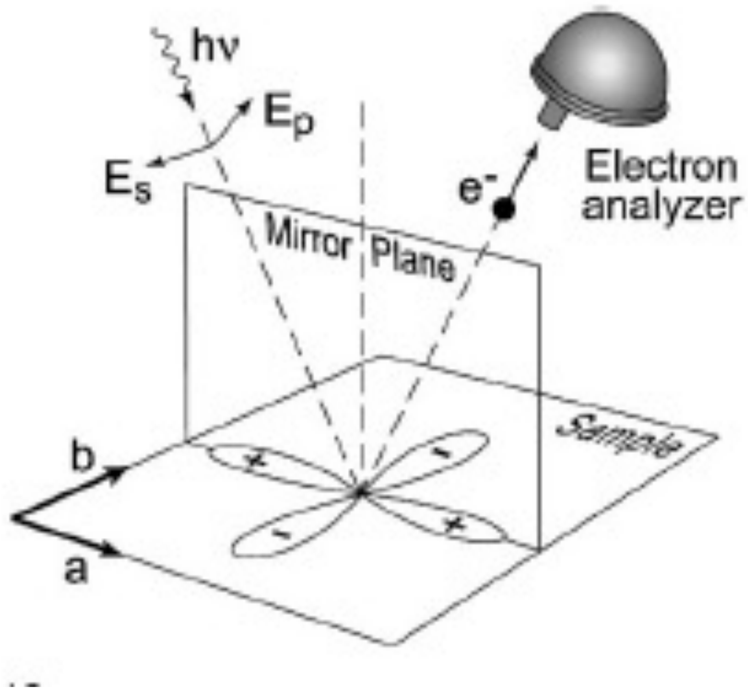
$$H_{int} = \frac{e}{m} \vec{A}.\vec{p} \quad \text{if } \vec{\nabla}\vec{A} = 0$$

Choice of Gauge not trivial when Hamiltonian is non-local or with SOC

*Moser, J. electron spectroscopy 2016*

# Selection rules as a function of light polarization

$$M_{i,f} = \left| \left\langle \varphi_f^k \middle| H_{\text{int}} \middle| \varphi_i^k \right\rangle \right|^2 \qquad H_{\text{int}} = \frac{e}{mc} \vec{A}.\vec{p}$$




$M_{i,f} \neq 0$ if $|\varphi_f|^2 \neq 0$ on detector,

i.e. $\varphi_f$ is even / symmetry plane.

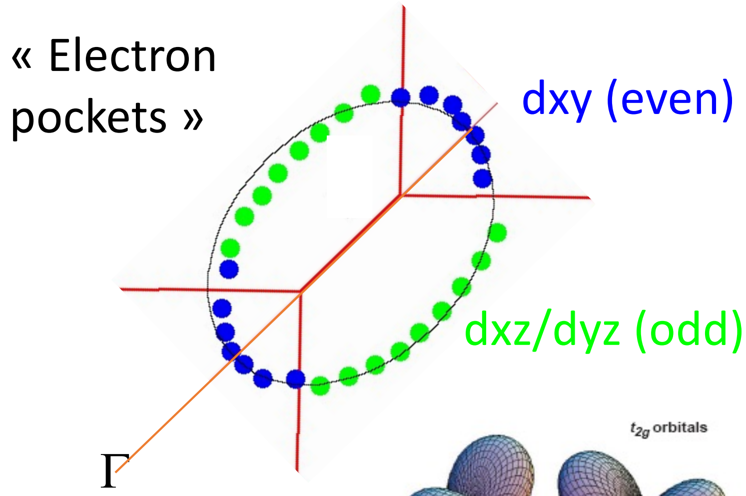
Then : either : $\varphi_i$ even and A even ($E_p$)

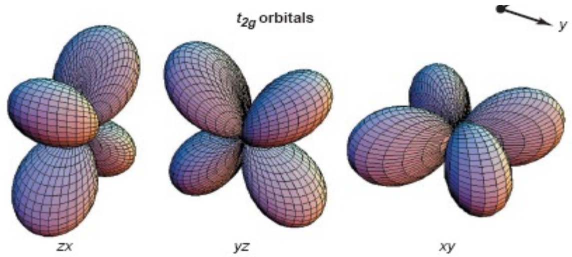
or : $\varphi_i$ odd and A odd ($E_s$)

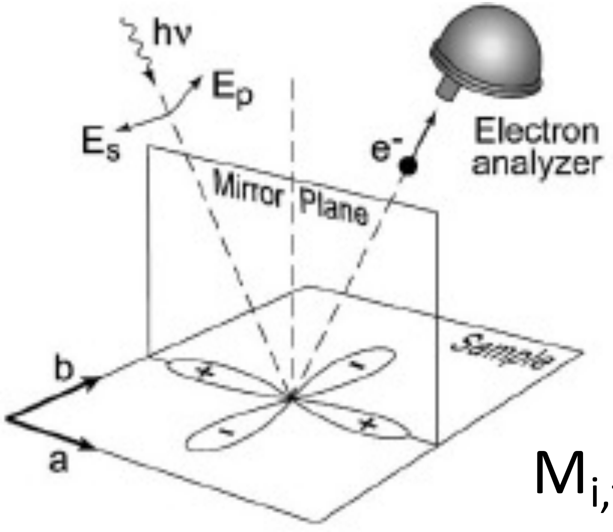
$\Rightarrow$ Polarization selects orbital of a given symmetry with respect with a mirror plane

# Selecting orbitals of one symmetry

## Iron-based superconductors

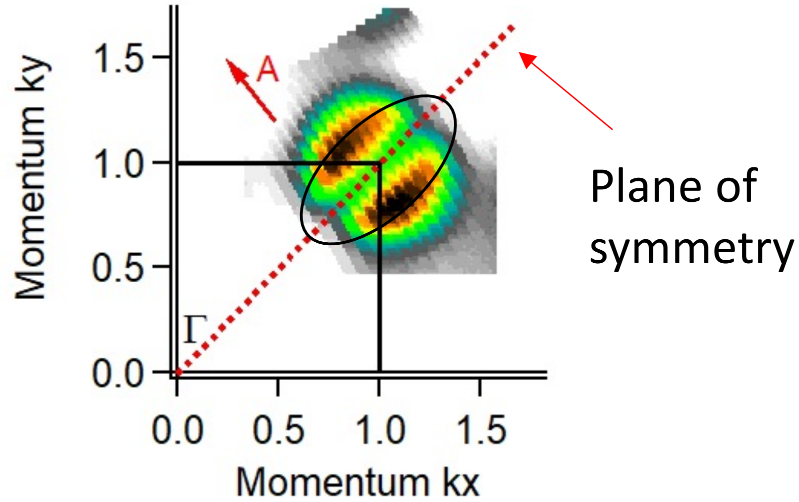


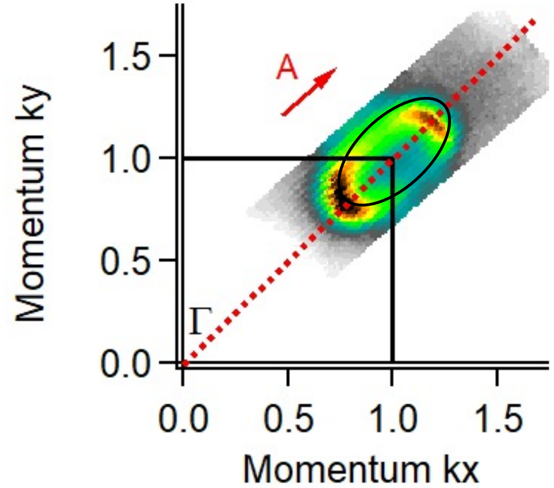




$$M_{i,f} = \left| \langle \varphi_f | \vec{A}.\vec{p} | \varphi_i \rangle \right|^2$$

« Odd » polarization

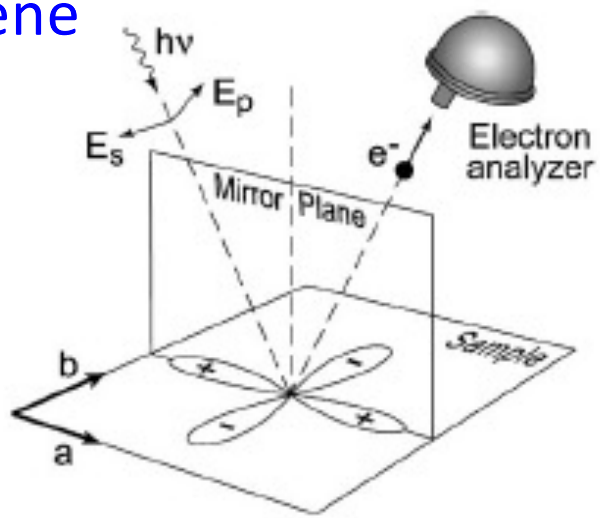


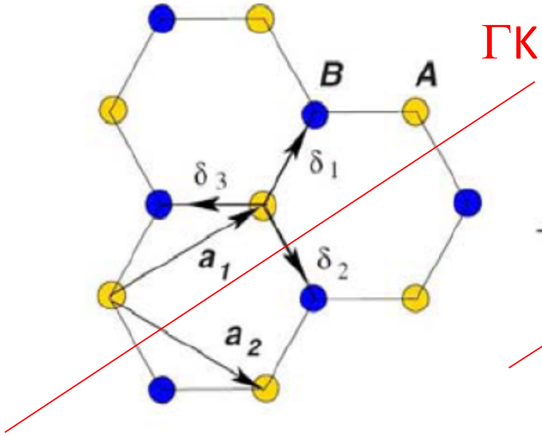

« Even » polarization

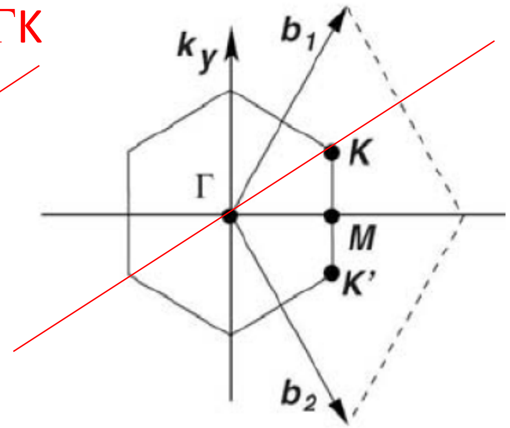


# Selecting orbitals of one symmetry

Graphene

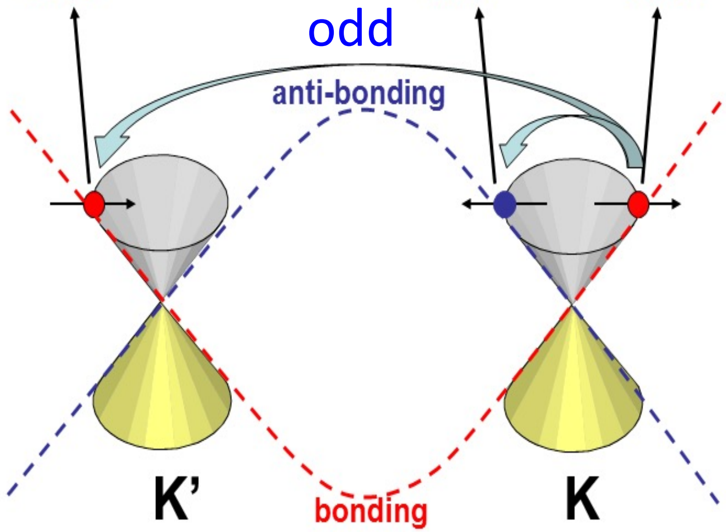


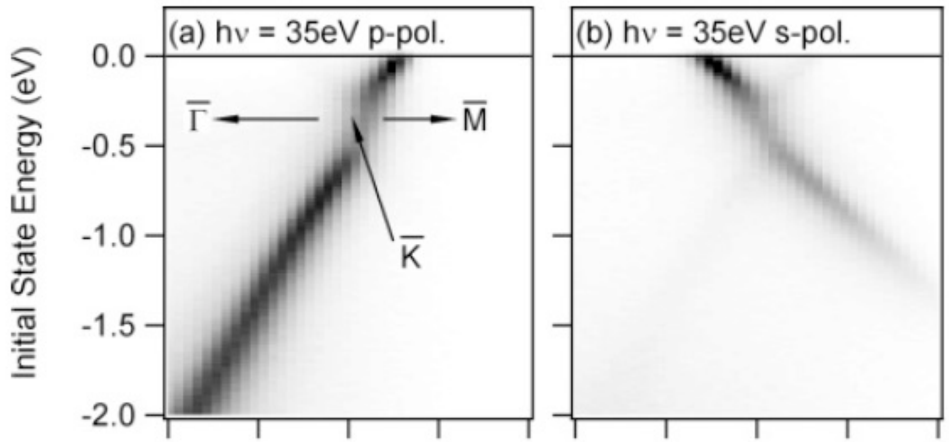

$$|\psi_k(r)\rangle = c_A\,|p_z(r-R_A)\rangle + c_B\,|p_z(r-R_B)\rangle$$

odd




even

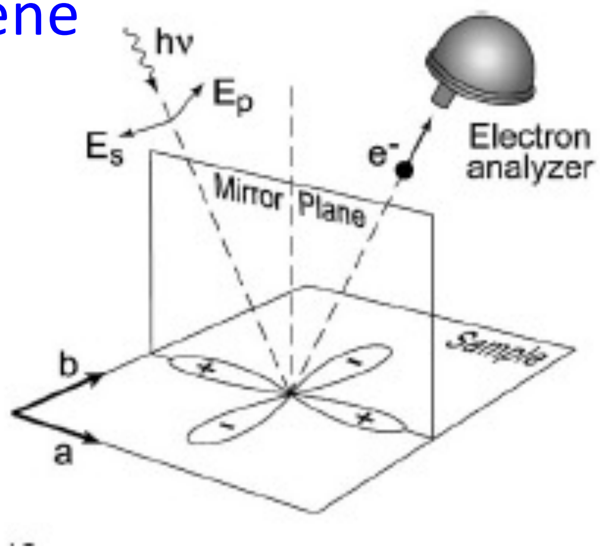
even odd

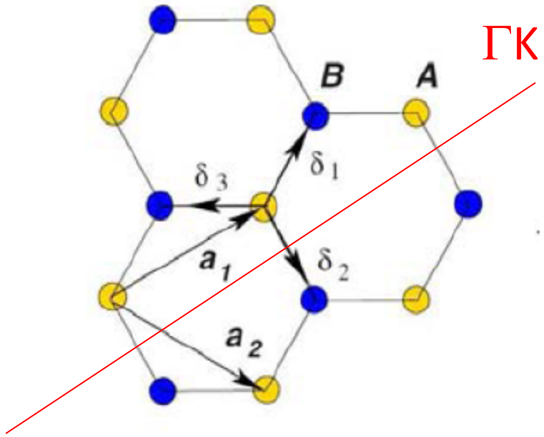



*Gierz PRB11*

# Selecting orbitals of one symmetry

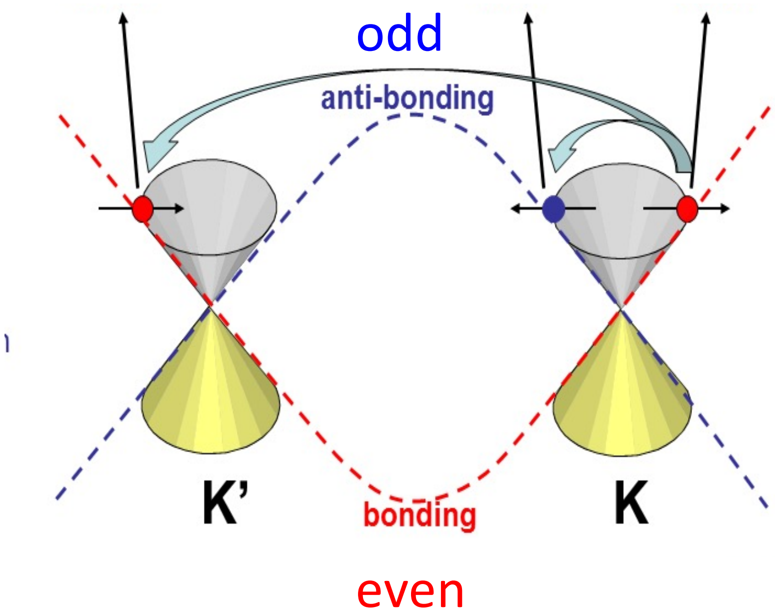
Graphene

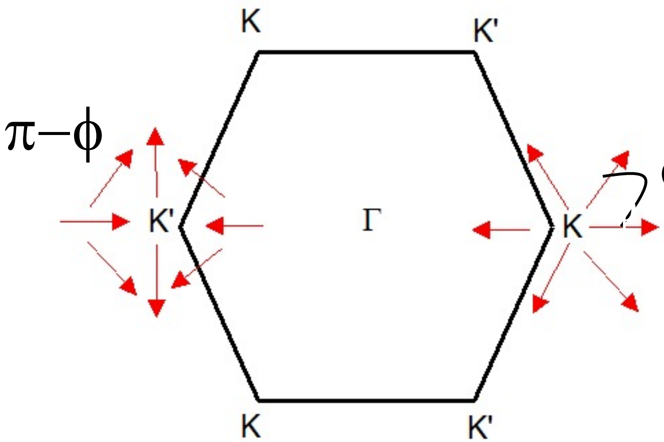


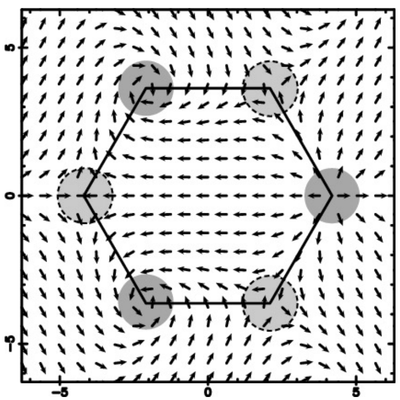




$$|\psi_k(r)\rangle = c_A\, |p_z(r-R_A)\rangle + c_B\, |p_z(r-R_B)\rangle$$

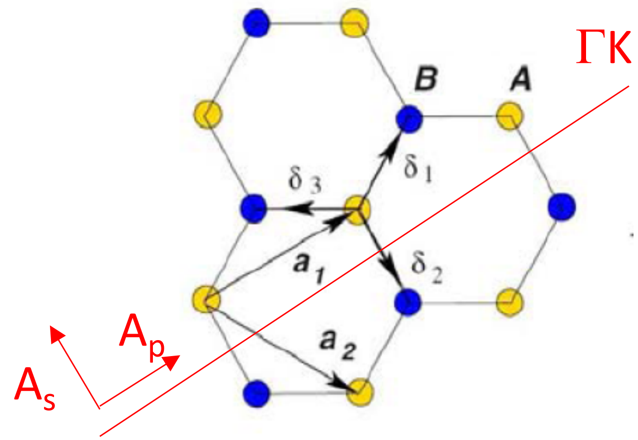
$$\begin{pmatrix} c_A \\ c_B \end{pmatrix} \sim \begin{pmatrix} 1 \\ e^{i\varphi(k)} \end{pmatrix}$$



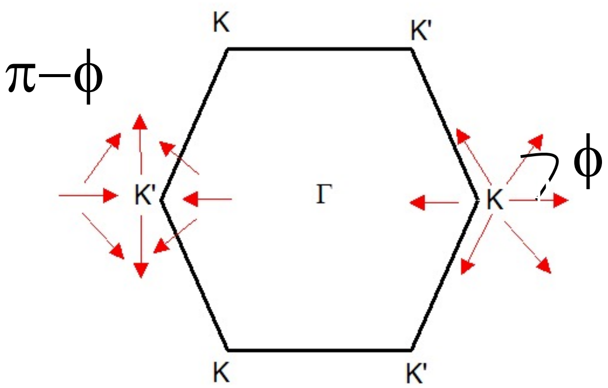



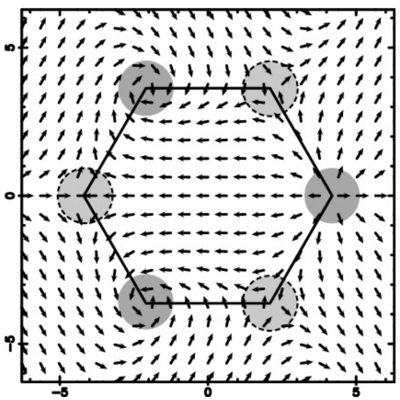
# Graphene pseudospin orientation



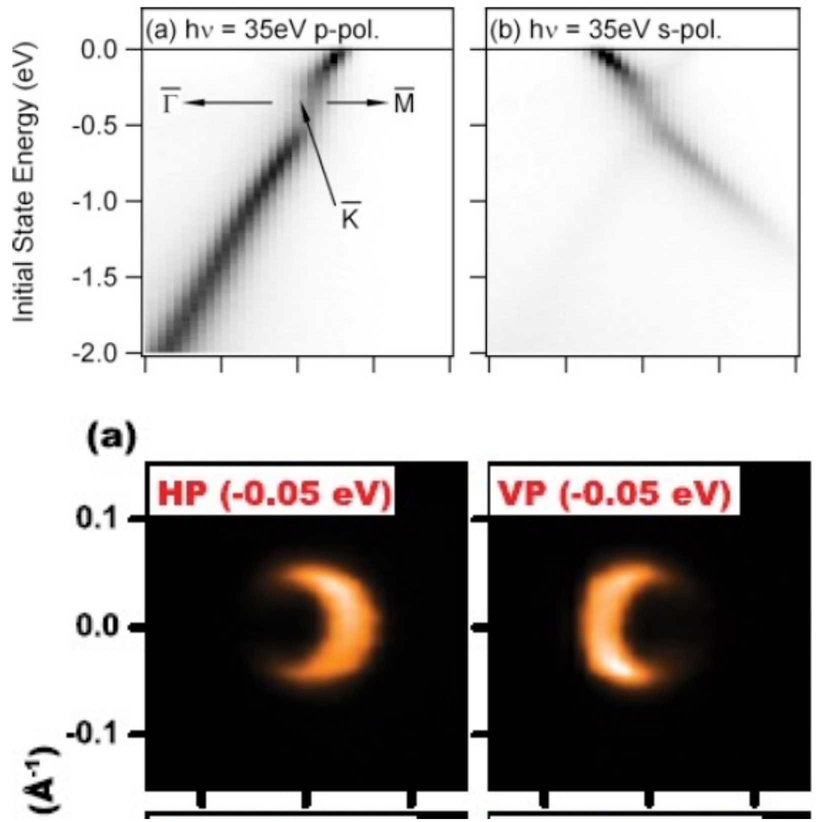

$$|\psi_k(r)\rangle = c_A\,|p_z(r-R_A)\rangle + c_B\,|p_z(r-R_B)\rangle$$

$$\begin{pmatrix} c_A \\ c_B \end{pmatrix} \sim \begin{pmatrix} 1 \\ e^{i\varphi(k)} \end{pmatrix}$$



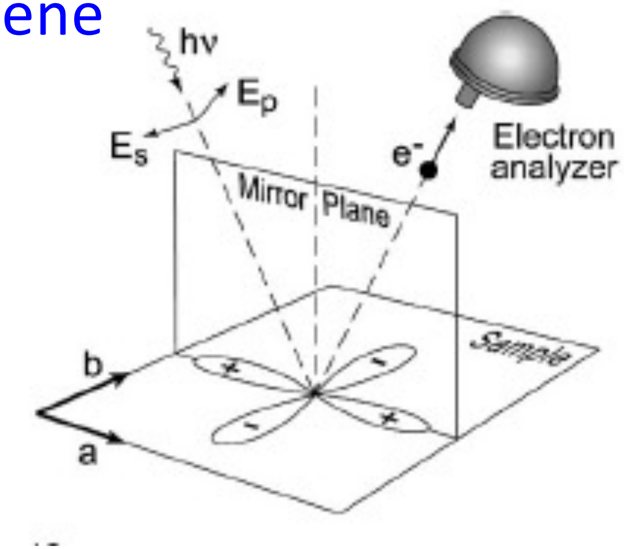
p-pol
s-pol

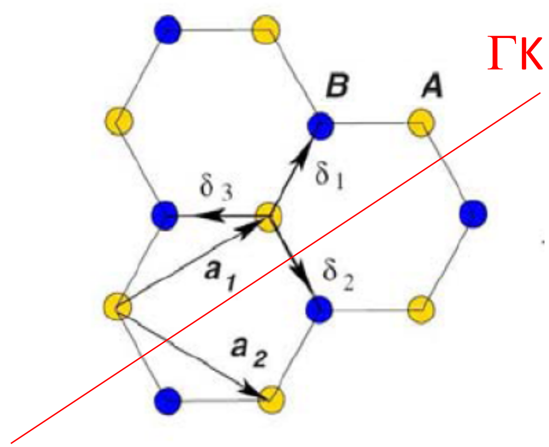



*Liu PRL 11*

# Graphene pseudospin orientation

Graphene




$|\psi_k(r)\rangle = c_A\, |p_z(r-R_A)\rangle + c_B\, |p_z(r-R_B)\rangle$

$$\begin{pmatrix} c_A \\ c_B \end{pmatrix} \sim \begin{pmatrix} 1 \\ e^{i\varphi(k)} \end{pmatrix}$$

$M_{i,f} = \left|\langle \varphi_f | \vec{A}.\vec{p} | \varphi_i \rangle\right|^2$

$M_{i,f} = |A_p\langle\varphi_f|\vec{\varepsilon_p}.\vec{p}|\varphi_i\rangle + A_s\langle\varphi_f|\vec{\varepsilon_s}.\vec{p}|\varphi_i\rangle|^2$

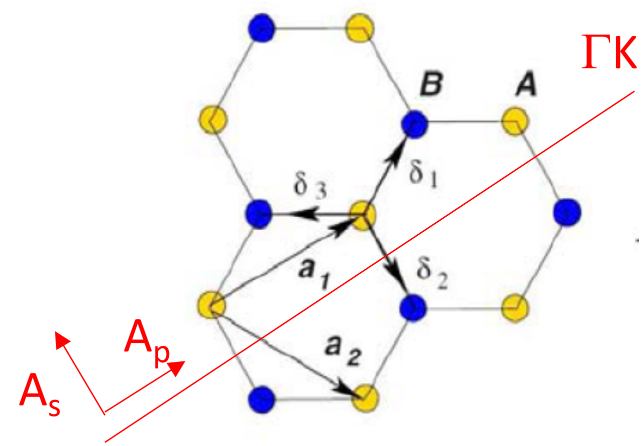
$M_{i,f} = |A_p[c_A T_p(A) + c_B T_p(B)] + A_s[c_A T_s(A) + c_B T_s(B)]|^2$

*with*

$T_{s,p}(A) = As_{,p}\langle\varphi_f|\overrightarrow{\varepsilon_{s,p}}.\vec{p}|p_z(A)\rangle$

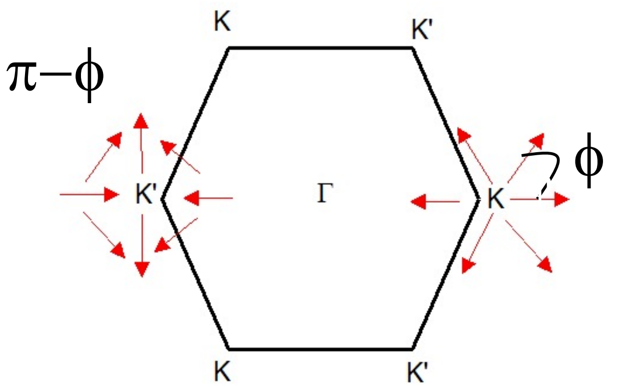
$M_{i,f} = |A_p T_p(c_A + c_B) + AsTs(c_A - c_B)|^2$

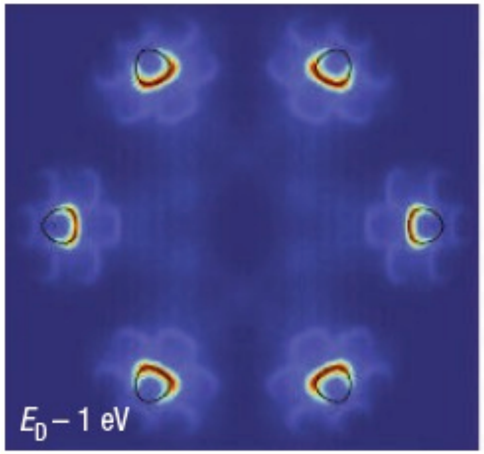
# Graphene pseudospin orientation



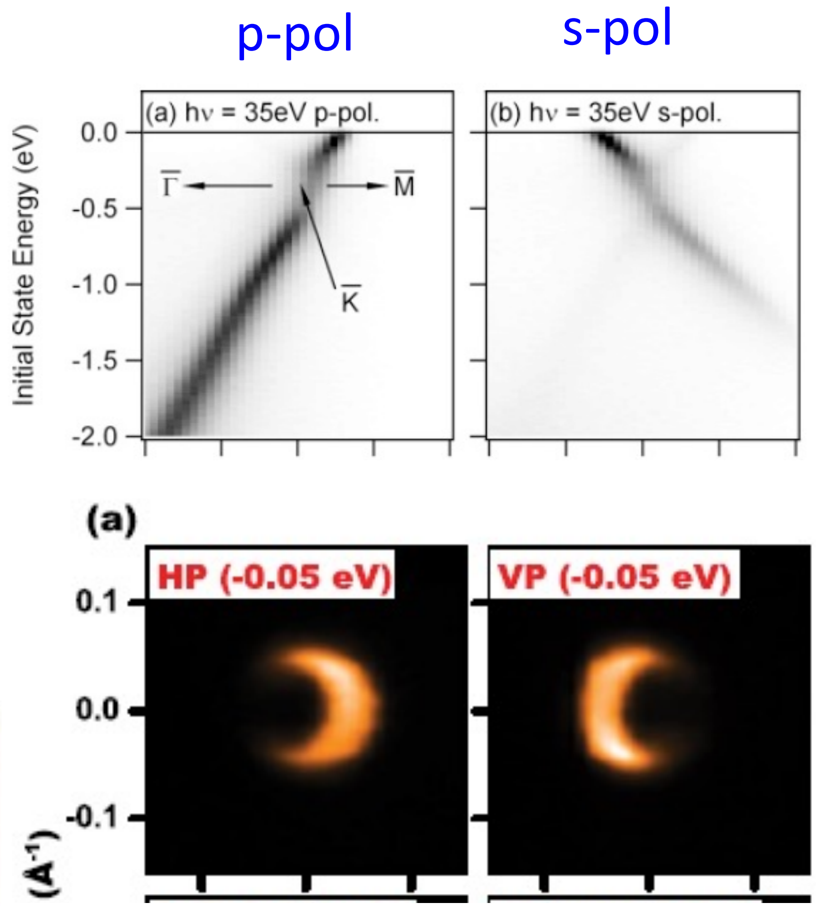

$$|\psi_k(r)\rangle = c_A\,|p_z(r-R_A)\rangle + c_B\,|p_z(r-R_B)\rangle$$

$$\begin{pmatrix} c_A \\ c_B \end{pmatrix} \sim \begin{pmatrix} 1 \\ e^{i\varphi(k)} \end{pmatrix}$$




No apparent difference in K and K'




$I \sim |C_A + C_B|^2$

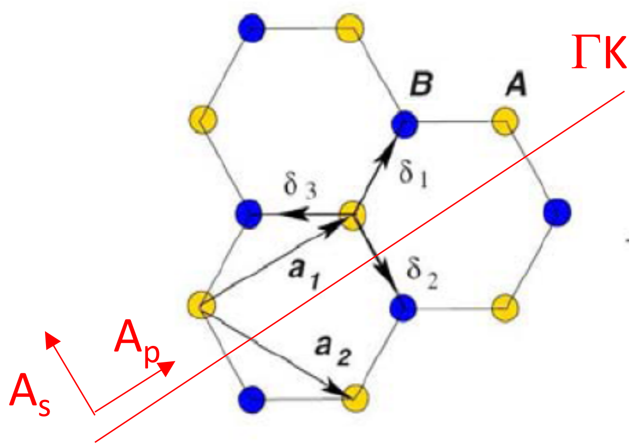
$I \sim 2(1+\cos\phi)$

$I \sim |C_A - C_B|^2$

$I \sim 2(1-\cos\phi)$

*Liu PRL 11*

# Graphene Berry phase




$|\psi_k(r)\rangle = c_A\,|p_z(r-R_A)\rangle + c_B\,|p_z(r-R_B)\rangle$

$$\binom{c_A}{c_B} \sim \binom{1}{e^{i\varphi(k)}}$$

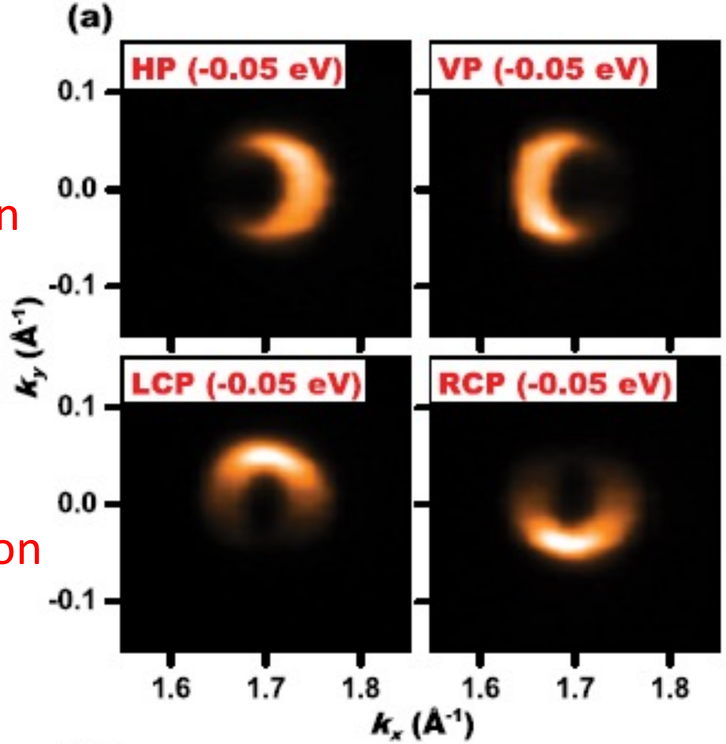
$I \sim 2(1+\cos\phi)$ $\quad$ $I \sim 2(1-\cos\phi)$

$I \sim |C_A+C_B|^2$ $\quad$ $I \sim |C_A-C_B|^2$

Linear polarization

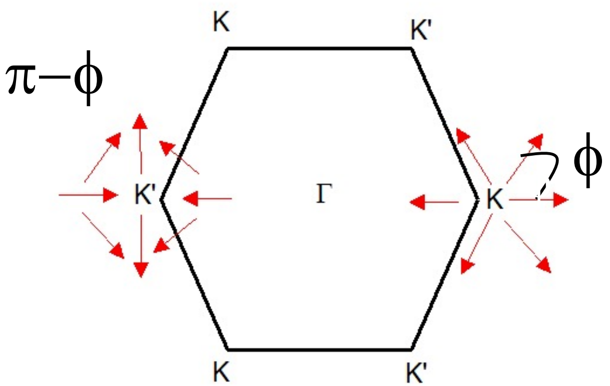
Circular Polarization

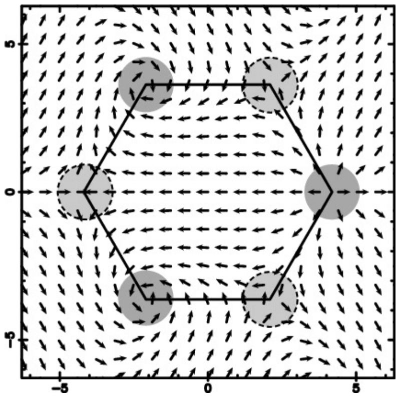
$A_p \pm i\,A_s$




$I \sim |(1+i)C_A + (1-i)C_B|^2$ $\quad$ $I \sim |(1-i)C_A+(1+i)C_B|^2$

$I \sim 4(1+\sin\phi)$ $\quad$ $I \sim 4(1-\sin\phi)$

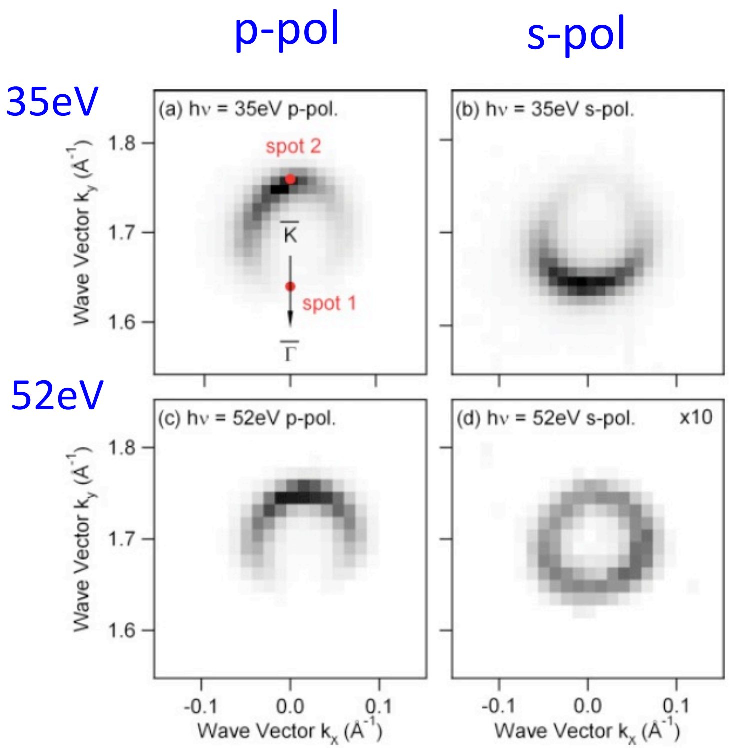



*Liu PRL 11*

# Warning : this reasoning only takes the initial state into account

Changes as a function of photon energy !




*GierzPRB11*

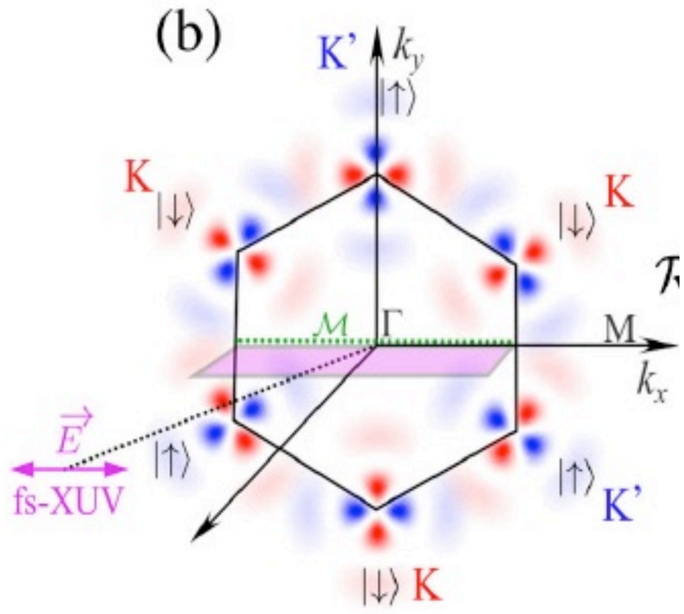
$$M_{i,f} = \left|\langle \varphi_f | \vec{A}.\vec{p} | \varphi_i \rangle\right|^2$$

Complete calculation of ARPES matrix elements are more and more possible, but less intuitive

# Alternative way : switching K and K'

Idea : keep experimental geometry identical will keep same matrix elements

WSe$_2$



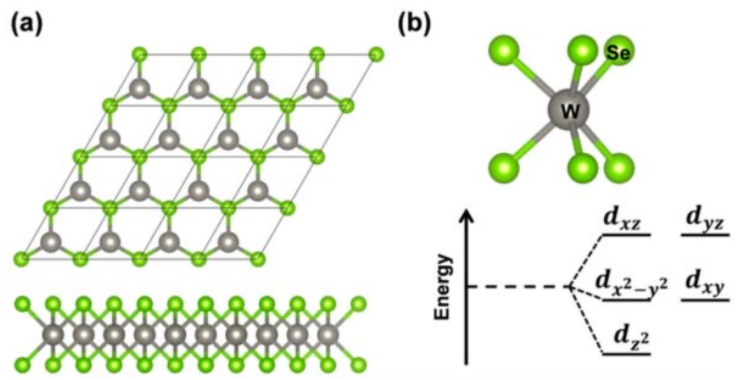

Orbital pseudospin :

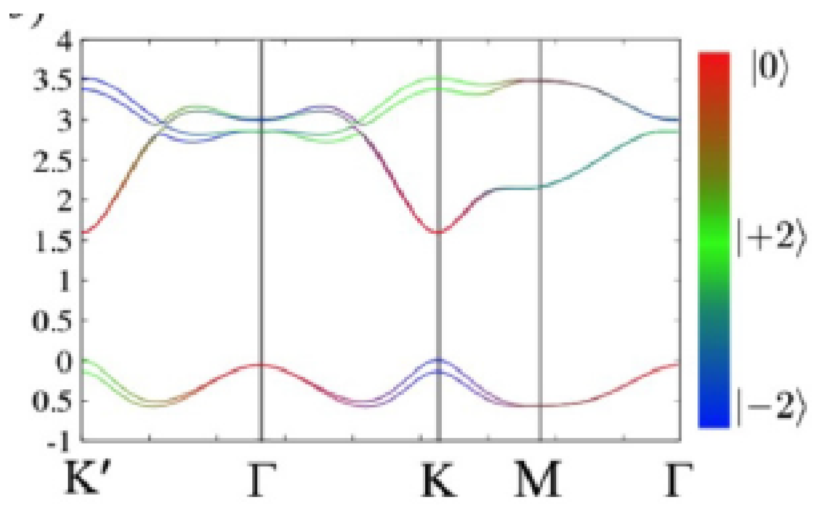
wavefunction

$$|\psi_{\mathbf{k}\alpha}\rangle = C_{\pm 2}(\mathbf{k})|d_{\pm 2}\rangle + C_0(\mathbf{k})|d_{z^2}\rangle$$

pseudospin

$$\sigma(\mathbf{k}) = \langle\psi_{\mathbf{k}\alpha}|\hat{\sigma}|\psi_{\mathbf{k}\alpha}\rangle$$



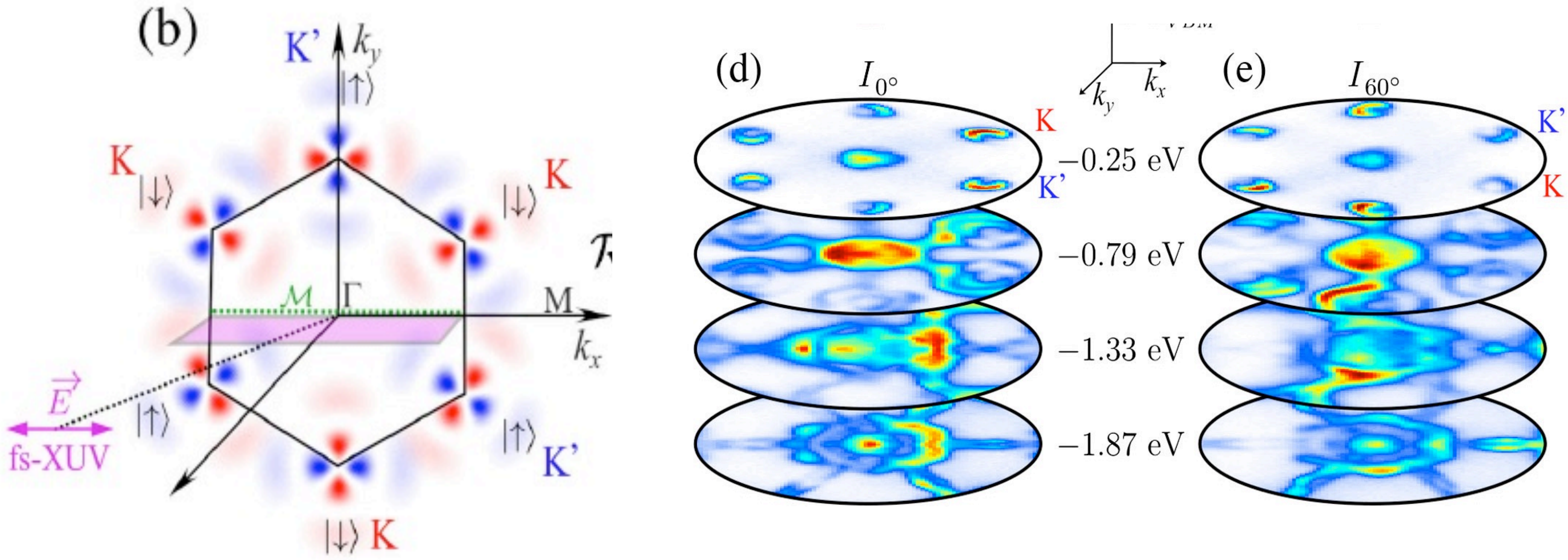




*Beaulieu PRL 20*

# Alternative way : switching K and K’

Idea : keep experimental geometry identical will keep same matrix elements

$WSe_2$



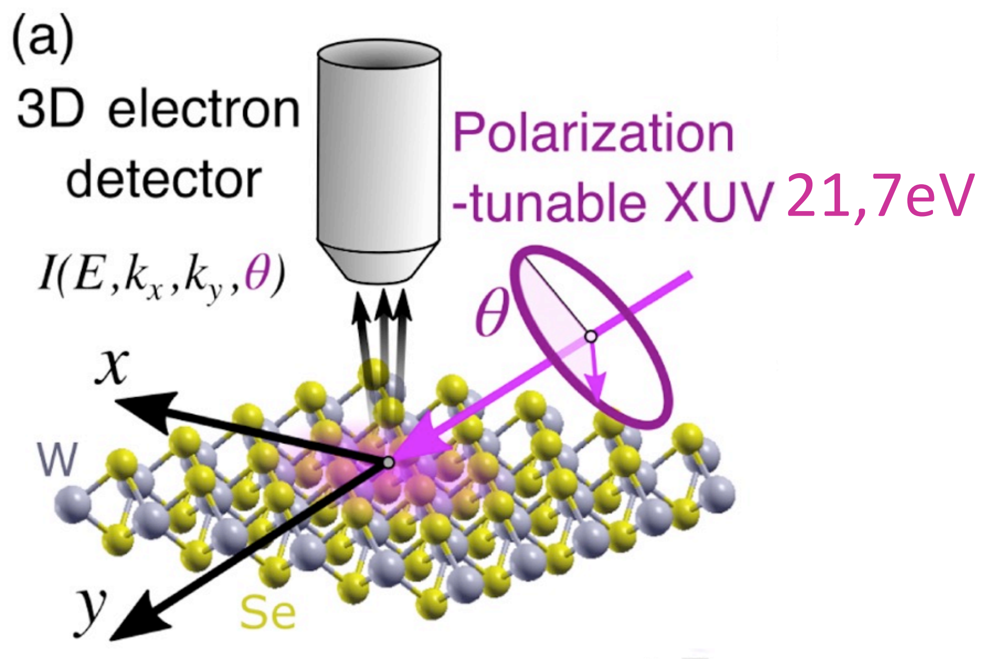

*Beaulieu PRL 20*

# Technological developments

**Polarization-Modulated Angle-Resolved Photoemission Spectroscopy: Toward Circular Dichroism without Circular Photons and Bloch Wave-function Reconstruction**

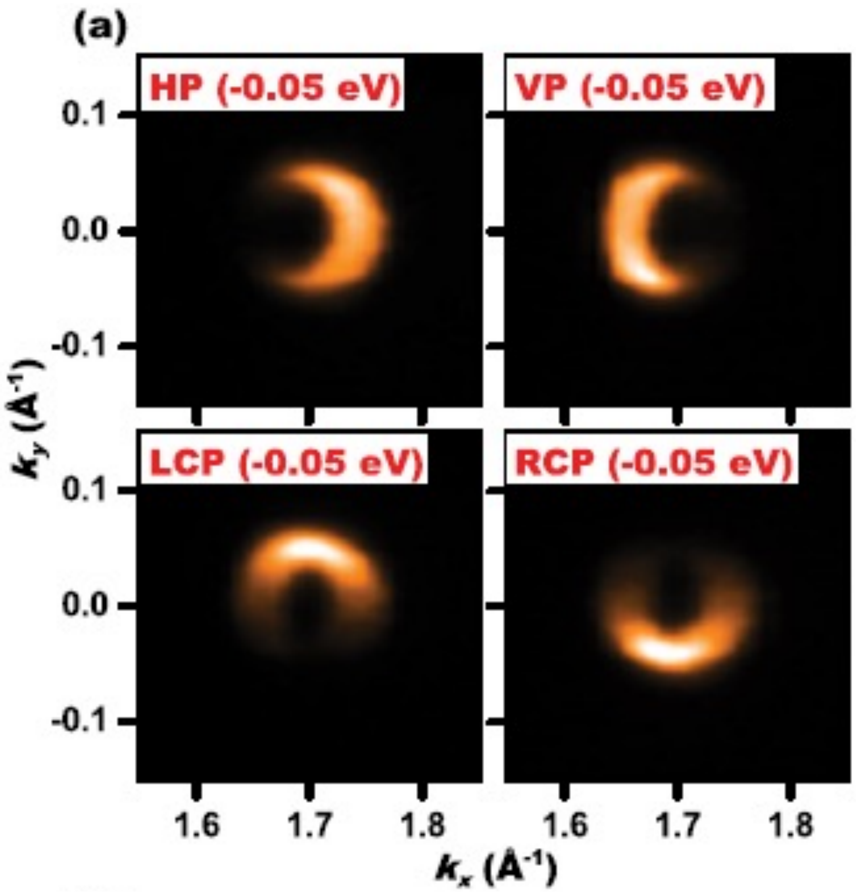
*Schuler et al., PRX 22*

*Samuel Beaulieu, Bordeaux, CELIA*




High-repetition rate (500 kHz) femtosecond XUV source with tunable linear polarization axis direction. Obtained via annular beam HHG scheme.

# Summary of part II




Detailed information on the initial wave function can be obtained from ARPES as a function of polarization, but care has to be taken to separate it from final state effects.

Graphene, *Liu PRL 11*

# When can ARPES be useful for the study of topological materials ?

In situations where DFT may be insufficient

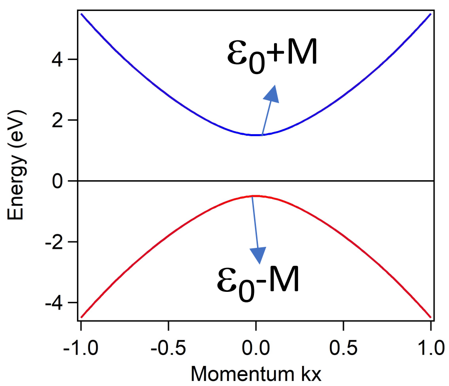
*Example : effective doping of graphene by substrate or TI by impurities*

- Position of the Dirac cones etc. with respect to Fermi level
- Precise knowledge of relative band positions (crossing or not ?)
- Strong correlations (Kondo insulator etc)

# Transition between a trivial and topological insulator

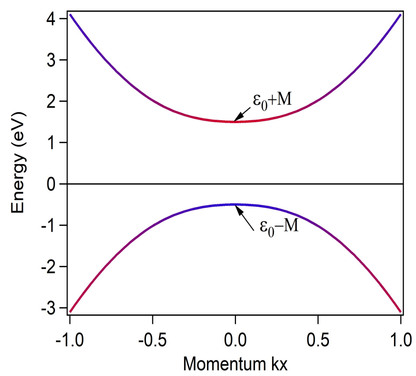
Crossing of 2 bands with opposite symmetry :

M>0




Trivial insulator

M<0




Topological insulator

$\Rightarrow$ DFT may not be accurate enough to predict the value of M

# Transition between a trivial and topological insulator

Crossing of 2 bands with opposite symmetry :

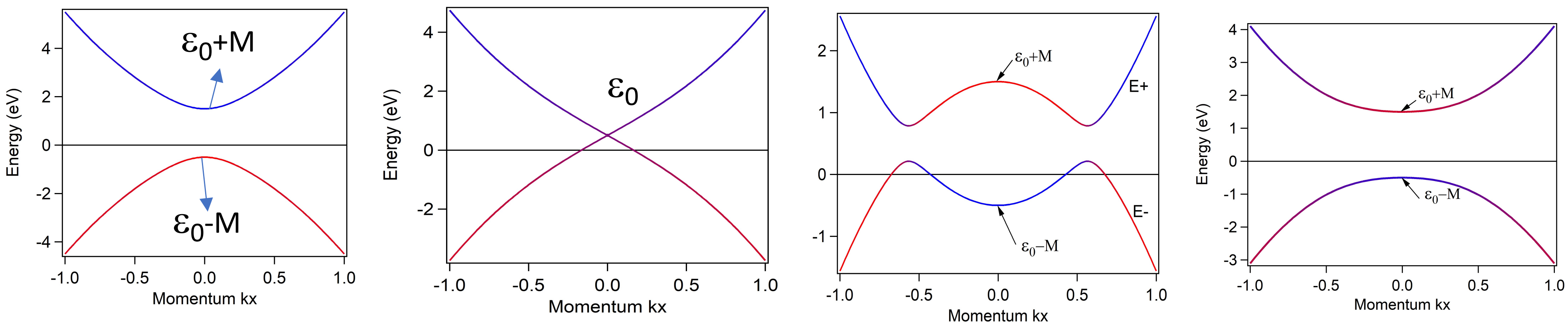
$$H(k) = \begin{bmatrix} \varepsilon_0 & 0 \\ & \varepsilon_0 \end{bmatrix} + \begin{bmatrix} M - Bk^2 & A(k_x + ik_y) \\ A(k_x - ik_y) & -M + Bk^2 \end{bmatrix}$$

$$\varepsilon(k) = \varepsilon_0 \pm \sqrt{M(k)^2 + A^2({k_x}^2 + {k_y}^2)}$$

Dispersion along $k_x$ :

M>0

M<0 M=0 small A Gap=$2Ak_c$ large A



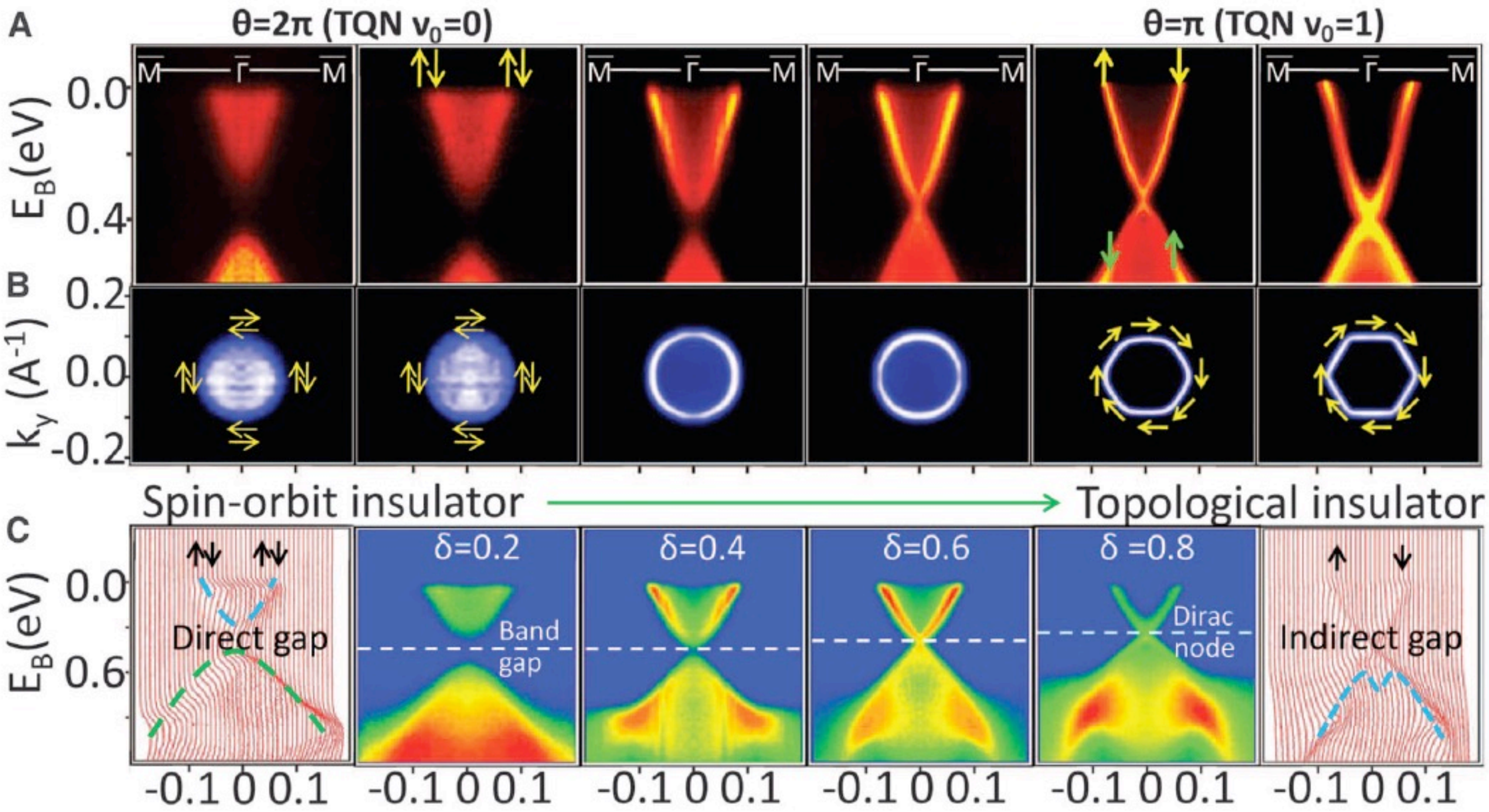

Trivial insulator ⟶ Dirac semimetal ⟶ Topological insulator

⇒ DFT may not be accurate enough to predict the value of M

=> The dispersion and the hybridation are sensitive to the value of coupling A

# Transition controlled by doping

Expected as a function of $\delta$ in Bi Tl $(S_{1-\delta}Se_{\delta})_2$



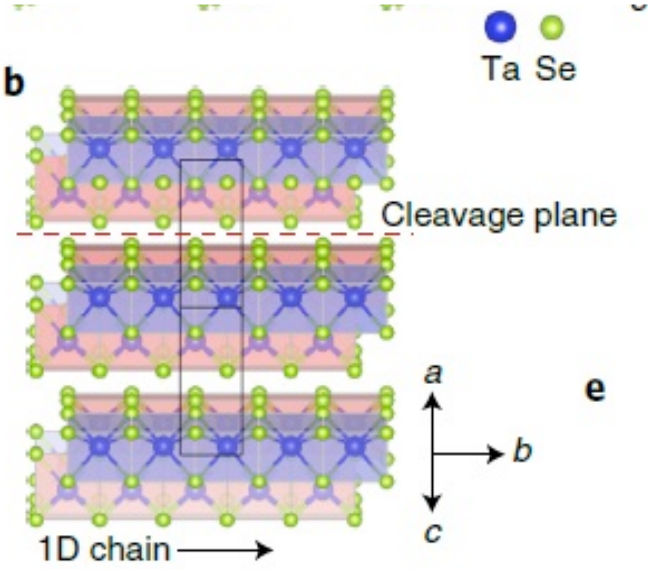

*XuHasanScience11*

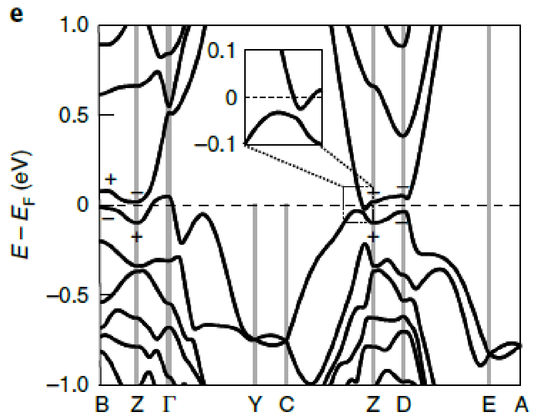
# Transition controlled by strain

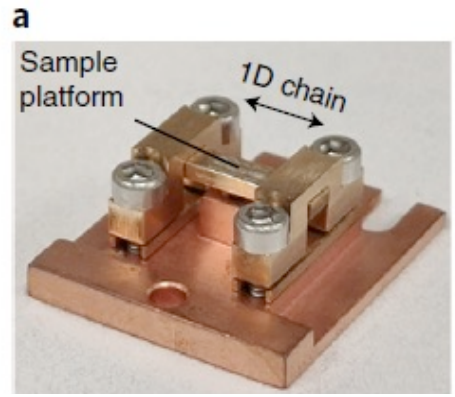
Strain device

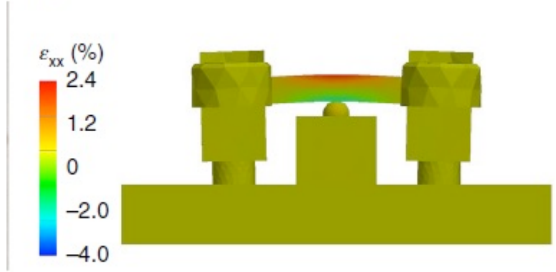
1D chains $TaSe_3$

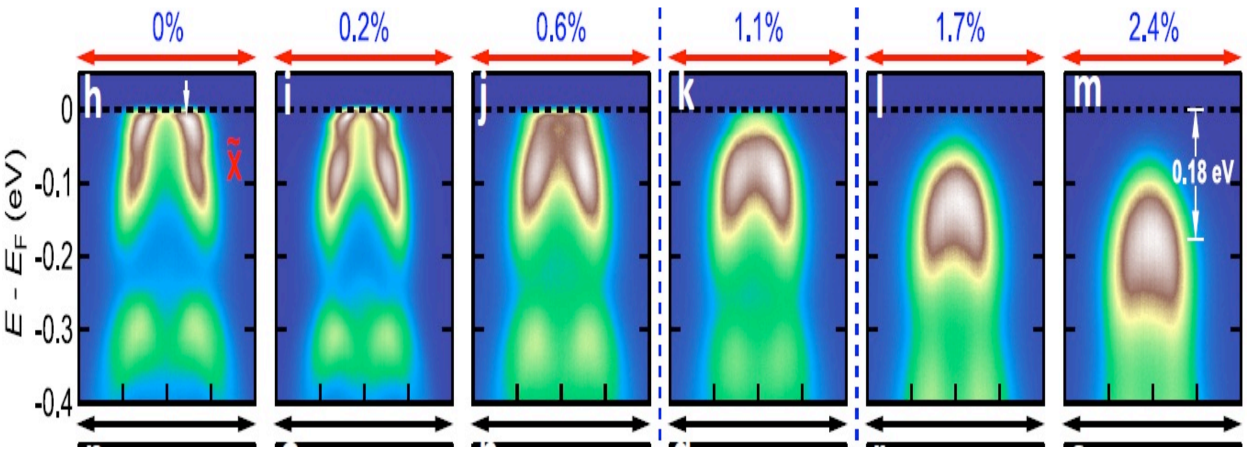
Inverted bands

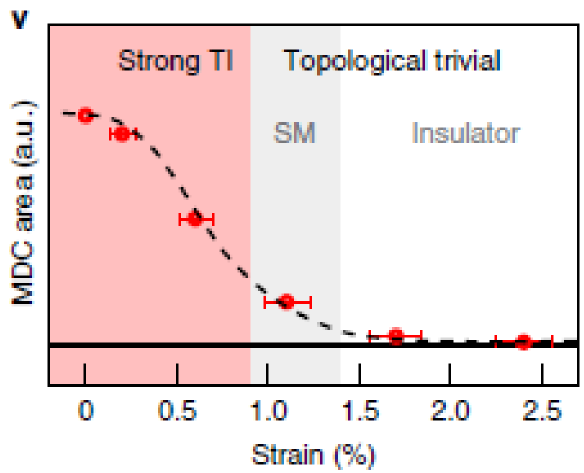


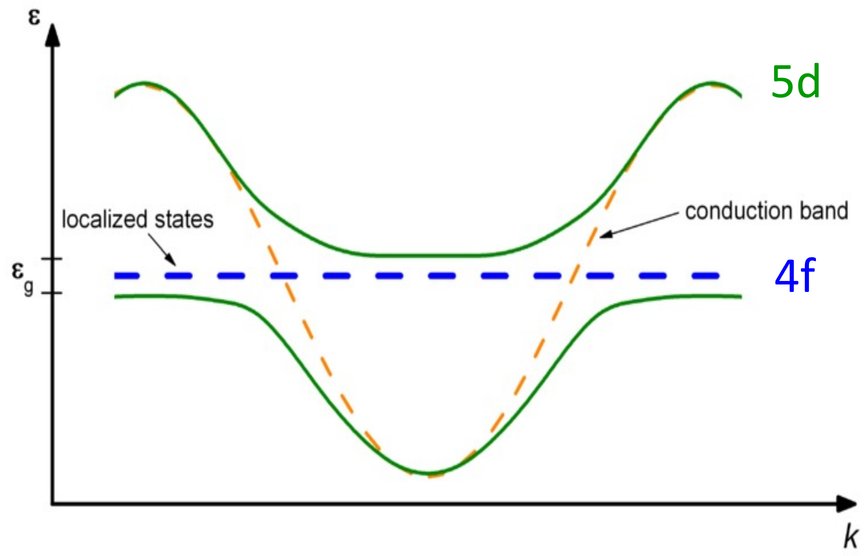



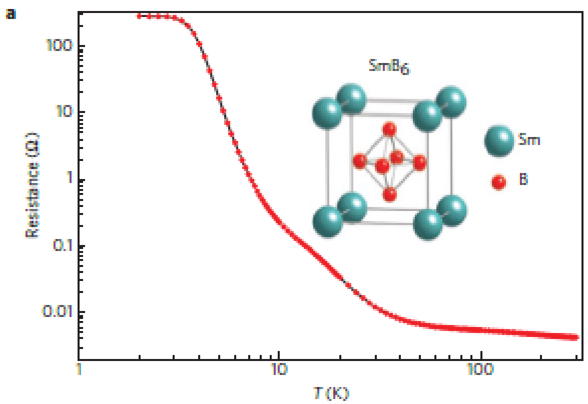

*C. Lin, T. Kondo Nature Mat. 21*

# Kondo insulators: $SmB_6$



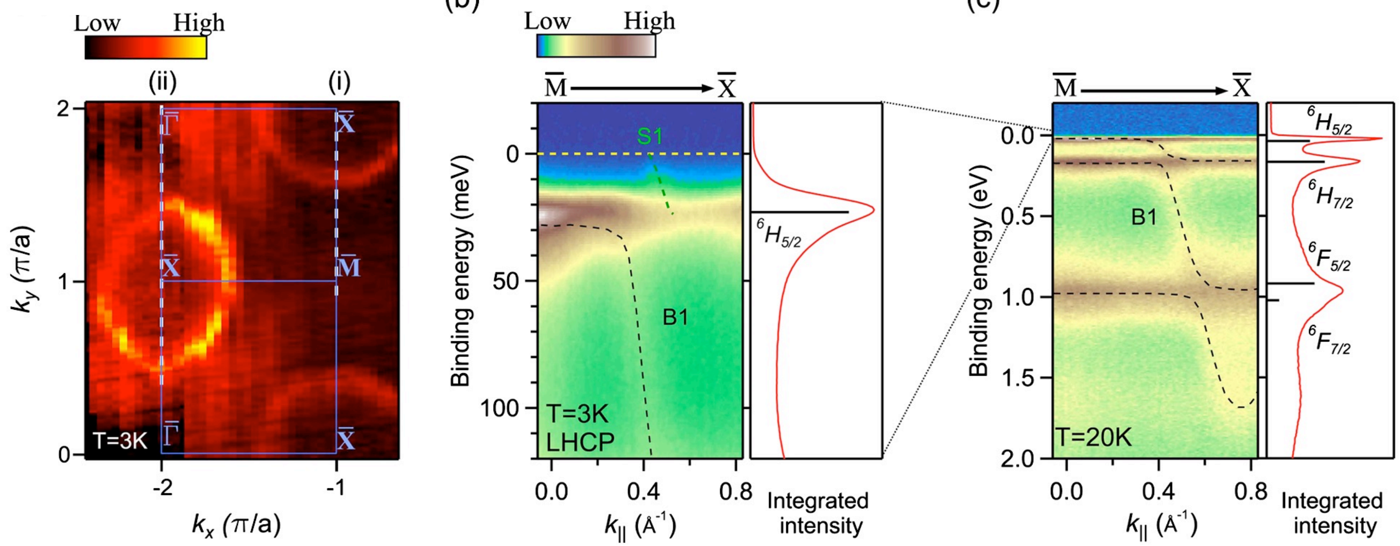

*Dzero, Coleman PRL10*

Typical example : $SmB_6$




*Kim, Fisk, Nature Mat 14*

ARPES



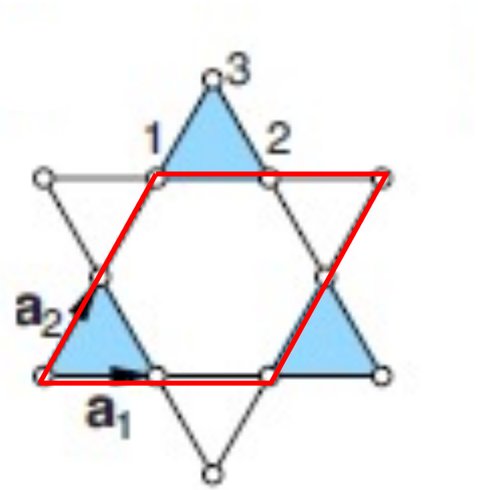

A lot of discussion on whether surface states are trivial or topological ones

*Min, Reinert PRL 14*

# Kagome metals

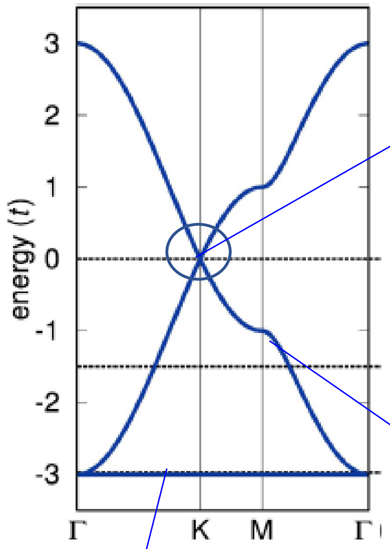
A way to combine topology and correlations/magnetism ?

Kagomé plane => 3 bands with non-trivial topological properties



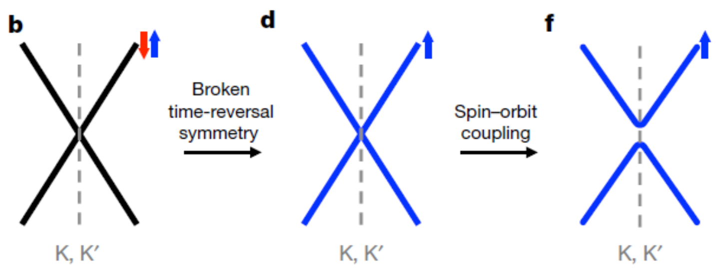

Guo PRL 09
Guterding Scientific Reports 17

Tight-binding model




Dirac cone

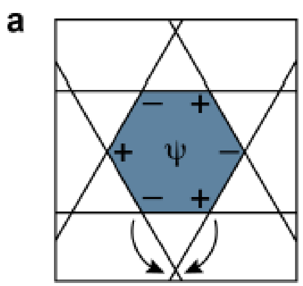
Similar to graphene, but spin polarized Dirac cone, with larger SOC




*Quantum Anomalous Hall state /Chern insulator (if 2D)*

« Flat band »

Non trivial, due to interference effects between atoms




Van Hove singularities

Nesting, exotic CDW..
Cf $AV_3Sb_5$

Very narrow band = Strong correlations.

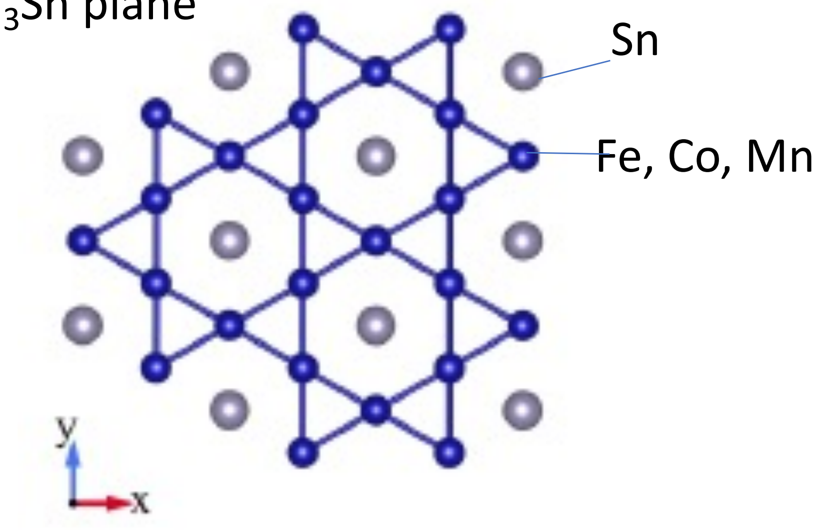
⇒ tendency to ferromagnetism
⇒ Possible high temperature superconductivity
⇒ Wigner crystallization
⇒ Fractional quantum Hall effect…

# Metallic kagome networks :
## a way to combine correlations and topology ?

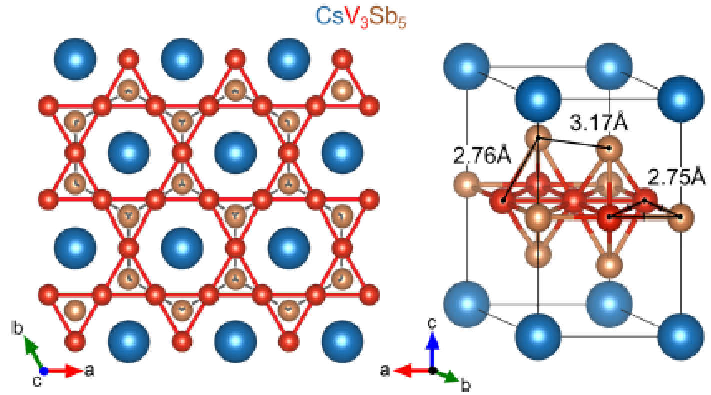
*Kagome planes are found in many real bulk materials*

$Fe_3Sn_2$, FeSn, CoSn, $Co_3Sn_2S_2$, $Mn_3Sn$…

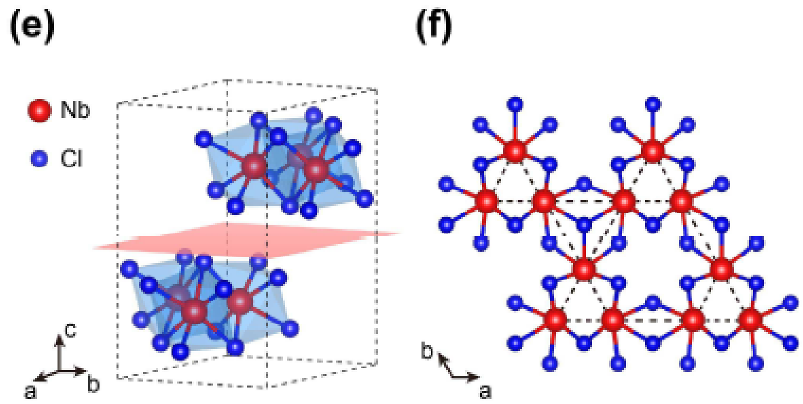
$M_3Sn$ plane



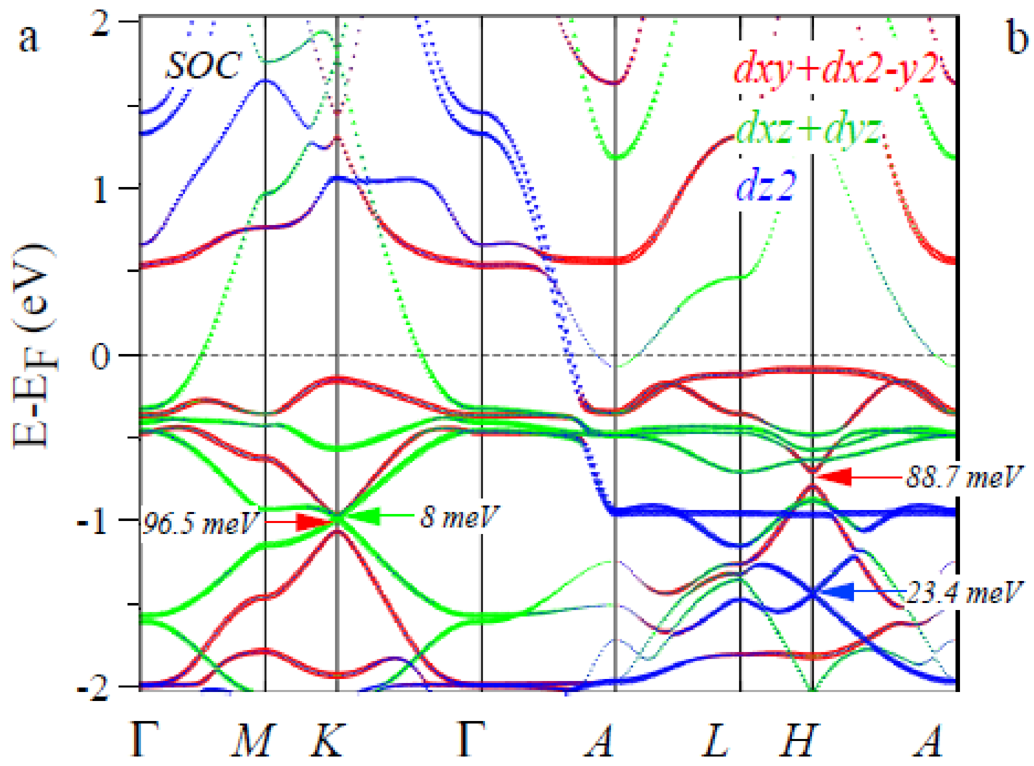

$AV_3Sb_5$




$Nb_3Cl_8$ (Van der Waals)

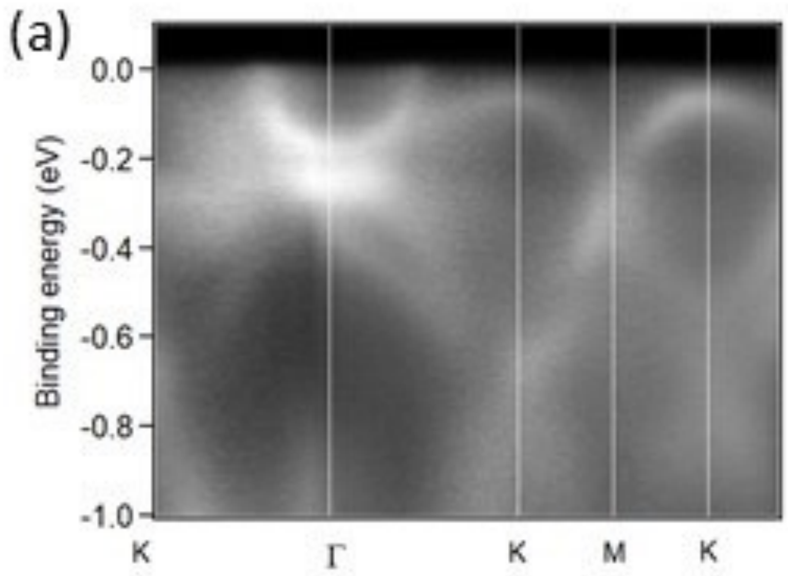


# CoSn : a kagome metal with relatively well defined flat bands




ARPES view (V. Brouet, Cassiopée)

Renormalization by ~1,4




*Can you move these features to $E_F$ ?*

# Conclusion

⇒ Qualitative « view » of band dispersion is easily obtained from ARPES (provided surface quality is good enough).

⇒ Quantitative measurement is often difficult, because of many uncontrolled quantities in ARPES « matrix element »

=> To interpret reliably topological properties

- Detailed and accurate ab-initio calculations
- Careful experiments with good control of polarization, sample alignment, change as a function of energy can give valuable clues

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