« ARPES and topology »

Véronique Brouet

Laboratoire de Physique des Solides d’Orsay
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....

- 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!!

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

\[ E_{\text{kin}} = h \nu - W - |E_B| \]

Conservation of momentum parallel to surface

\[ \hbar k_\parallel = \sqrt{2mE_{\text{kin}}} \sin \theta \]
Angle-resolved photoemission

1- Energy conservation:

\[ E_{\text{kin}} = h\nu - W - |E_B| \]

2- Momentum conservation

\[ \hbar k_\parallel = \sqrt{2mE_{\text{kin}}} \sin \theta \]

⇒ \( E_{\text{kin}} \) vs \( k_\parallel \): Structure de bandes

NB: \( k_{\text{perp}} \) is not conserved
Experimentally...

CASSIOPEE beamline, SOLEIL synchrotron

Photons from: Synchrotrons: 10-1000eV
He lamp: 21 eV
Laser: 6-7eV (extending...)

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

$\hbar k_{||} = \sqrt{2mE_{\text{kin}}} \sin \theta$

Surface sensitive !!

=> Ultra-high vacuum, good surface!

$E_{\text{kin}} \sim h\nu$

"universal" curve
3D data set \( I(E, k_x, k_y) \)

**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

LiFeAs

free electron final state

42eV

30eV

initial state, in bulk

Bulk dispersion perpendicular to surface

\[ V_0 - W \]
Example of 3D dispersion

Quasi-2D Fermi Surface

LiFeAs

dxz   dxy   dyz

(a2)   (a1)

Energy (eV)

k axis (π/a units)

Photon energy (eV)

k axis (π/a units)

=> 2D band

=> Significant warping
Perpendicular momentum?

\[ \delta k_{\text{perp}} \sim \frac{1}{\lambda} \]

Steps of the photoemission process

Moser, J. electron spectroscopy 2016
Perpendicular momentum?

\[ \delta k_{\text{perp}} \sim \frac{1}{\lambda} \]

- Free electron final state
- Initial state, in bulk
- Bulk dispersion perpendicular to surface

\[ E - E_F \]

\[ k_{\text{perp}} \text{ (Å}^{-1} \text{)} \]

- 3D Fermi Surface
- Quasi-2D Fermi Surface
- LiFeAs

42 eV
30 eV

\[ \frac{1}{\lambda} \]
How to increase the definition of $k_{\text{perp}}$?

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\textsuperscript{1,2}, D. Qian\textsuperscript{1,3}, D. Hsieh\textsuperscript{1,2}, L. Wray\textsuperscript{1}, A. Pal\textsuperscript{1}, H. Lin\textsuperscript{4}, A. Bansil\textsuperscript{4}, D. Grauer\textsuperscript{5}, Y. S. Hor\textsuperscript{5}, R. J. Cava\textsuperscript{5} and M. Z. Hasan\textsuperscript{1,2,6} \textsuperscript{*}

$\text{Bi}_2\text{Se}_3$

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

Xia et al. Nature Physics 2009
Topological surface states

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

Y. Xia\textsuperscript{1,2}, D. Qian\textsuperscript{1,3}, D. Hsieh\textsuperscript{1,2}, L. Wray\textsuperscript{1}, A. Pal\textsuperscript{1}, H. Lin\textsuperscript{4}, A. Bansil\textsuperscript{4}, D. Grauer\textsuperscript{5}, Y. S. Hor\textsuperscript{5}, R. J. Cava\textsuperscript{5} and M. Z. Hasan\textsuperscript{1,2,6,*}

\textbf{Bi}_2\textbf{Se}_3

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

\textit{Xia et al. Nature Physics 2009}
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

Hsieh, Hasan Nature 09

$\text{Bi}_2\text{Te}_3$
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:
\[ \text{CD} = I(\text{RCP}) - I(\text{LCP}) \]
\[ \text{CD} \sim \vec{l} \cdot \vec{k}_{ph} \]

Orbital Angular Momentum
Related to Berry curvature:
\[ \ell_{\nu\sigma}^z (\vec{k}) = -\frac{m}{\hbar} (\varepsilon_{k\nu} - \varepsilon_{k\sigma}) \Omega_{\nu\sigma} (\vec{k}) \]

Schuler Science Adv. 2020

Park PRL12
Circular dichroism in Bi$_2$Te$_3$ 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$_2$Se$_3$

Can we go deeper into the nature of the observed states?
Deeper into the photoemission process

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

- N-1 electrons in interaction
- + photoemitted electron (\(\hbar\omega, k\))
- N electrons in interaction
Deeper into the photoemission process

Assumes independent particles

$$I(k, \omega) = \sum_{i,f} \frac{2\pi}{\hbar} |\langle \phi_{f}^{k} | H_{int} | \phi_{i}^{k} \rangle|^2 \delta(E_{f} - E_{i} - \hbar \nu)$$

« One electron » wave function
One electron matrix element

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

- Final state = plane wave?

- Interaction Hamiltonian?

\[
\hat{p} \rightarrow \hat{p} - e\hat{A}
\]

\[ H_{int} = \frac{e}{m} \hat{A} \cdot \hat{p} \quad \text{if} \ \nabla \hat{A} = 0 \]

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

\[ M_{i,f} = \left| \langle \phi_f^k | H_{\text{int}} | \phi_i^k \rangle \right|^2 \]

\[ H_{\text{int}} = \frac{e}{mc} \vec{A} \cdot \vec{p} \]

\( M_{i,f} \neq 0 \) if \( |\phi_f|^2 \neq 0 \) on detector,

i.e. \( \phi_f \) is even / symmetry plane.

Then:

- either: \( \phi_i \) even and \( A \) even (\( E_p \))
- or: \( \phi_i \) 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

« Electron pockets »

- dxy (even)
- dxz/dyz (odd)

« Odd » polarization

Odd polarization

« Even » polarization

Even polarization

M_{i,f} = |\langle \varphi_f | \vec{A} \cdot \vec{p} | \varphi_i \rangle|^2
Selecting orbitals of one symmetry

Graphene

\[ |\psi_k(r)> = c_A |p_z(r-R_A)> + c_B |p_z(r-R_B)> \]

Gierz PRB11
Selecting orbitals of one symmetry

Graphene

\[ |\psi_k(r)\rangle = c_A \mid p_z(r-R_A)\rangle + c_B \mid p_z(r-R_B)\rangle \]

\[ \begin{pmatrix} c_A \\ c_B \end{pmatrix} \sim \left( \frac{1}{e^{i\varphi(k)}} \right) \]
Graphene pseudospin orientation

\[ |\psi_k(r)\rangle = c_A |p_z(r-R_A)\rangle + c_B |p_z(r-R_B)\rangle \]

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

\[ \pi - \phi \]

\[ \Gamma K K' \]

Liu PRL 11
Graphene pseudospin orientation

Graphene

\[ |\psi_k(r)\rangle = c_A |p_z(r-R_A)\rangle + c_B |p_z(r-R_B)\rangle \]

\[
\begin{pmatrix}
    c_A \\
    c_B \\
\end{pmatrix} \sim \left( e^{i\varphi(k)} \right)
\]

\[
M_{i,f} = \left| \langle \varphi_f | \hat{A} \cdot \vec{p} | \varphi_i \rangle \right|^2
\]

\[
M_{i,f} = |A_p \langle \varphi_f | \overrightarrow{\epsilon_p} \cdot \vec{p} | \varphi_i \rangle + A_s \langle \varphi_f | \overrightarrow{\epsilon_s} \cdot \vec{p} | \varphi_i \rangle|^2
\]

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

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

\[ T_{s,p}(A) = A_s \langle \varphi_f | \overrightarrow{\epsilon_{s,p}} \cdot \vec{p} | p_z(A) \rangle \]
Graphene pseudospin orientation

\[ |\psi_k(r)\rangle = c_A |p_z(r-R_A)\rangle + c_B |p_z(r-R_B)\rangle \]
\[ \left( \begin{array}{c} c_A \\ c_B \end{array} \right) \sim \left( \begin{array}{c} 1 \\ e^{i\varphi(k)} \end{array} \right) \]

No apparent difference in \(K\) and \(K'\)

Liu PRL 11
Graphene Berry phase

\[ |\psi_k(r)\rangle = c_A |p_z(r-R_A)\rangle + c_B |p_z(r-R_B)\rangle \]

\[
\begin{pmatrix}
C_A \\
C_B
\end{pmatrix} \sim \begin{pmatrix}
1 \\
1
\end{pmatrix} e^{i\varphi(k)}
\]

Linear polarization \( A_p \pm i A_s \)

Circular Polarization

\begin{align*}
I &\sim 2(1+\cos\phi) \\
I &\sim |C_A + C_B|^2 \\
I &\sim 2(1-\cos\phi) \\
I &\sim |C_A - C_B|^2 \\
I &\sim |(1+i)C_A + (1-i)C_B|^2 \\
I &\sim |(1-i)C_A + (1+i)C_B|^2 \\
I &\sim 4(1+\sin\phi) \\
I &\sim 4(1-\sin\phi)
\end{align*}

\textit{Liu PRL 11}
Warning: this reasoning only takes the initial state into account

Changes as a function of photon energy!

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

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

---

GierzPRB11
Alternative way: switching K and K’

Idea: keep experimental geometry identical will keep same matrix elements

\[ W\text{Se}_2 \]

Orbital pseudospin:

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

\[
\sigma(k) = \langle \psi_{k\alpha} | \hat{\sigma} | \psi_{k\alpha} \rangle
\]

Beaulieu PRL 20
Alternative way: switching K and K’

Idea: keep experimental geometry identical will keep same matrix elements

WSe$_2$
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 :

\[
\begin{align*}
M>0 & \quad \varepsilon_0 + M \\
M<0 & \quad \varepsilon_0 - M
\end{align*}
\]

\(\Rightarrow\) 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:

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

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

Dispersion along \(k_x\):

\[\begin{align*}
M < 0 & : & \varepsilon_0 + M \\
M = 0 & : & \varepsilon_0 \\
small A & : & \text{Gap} = 2A_k_c \\
large A & : & \text{Topological insulator}
\end{align*}\]

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

\(\Rightarrow\) 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 ($S_{1-\delta}Se_{\delta}$)$_2$
Transition controlled by strain

1D chains TaSe$_3$

Inverted bands

Strain device

C. Lin, T. Kondo Nature Mat. 21
Kondo insulators: SmB$_6$

Typical example: SmB$_6$

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

- Very narrow band = Strong correlations.
- Tendency to ferromagnetism.
- Possible high temperature superconductivity.
- Wigner crystallization.
- Fractional quantum Hall effect...

Non trivial, due to interference effects between atoms.

Dirac cone

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

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

Van Hove singularities

Nesting, exotic CDW..

Cf AV₃Sb₅
Metallic kagome networks: a way to combine correlations and topology?

Kagome planes are found in many real bulk materials

Fe$_3$Sn$_2$, FeSn, CoSn, Co$_3$Sn$_2$S$_2$, Mn$_3$Sn…

M$_3$Sn plane

AV$_3$Sb$_5$
CoSn : a kagome metal with relatively well defined flat bands

ARPES view (V. Brouet, Cassiopée)
Renormalization by \(\sim 1.4\)

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
Bibliography


Thanks to Frédéric Piéchon, Andrej Mesaros and Mark Goerbig for discussions