# Angle-resolved photoemission spectroscopy: introduction and applications

**Los Alamos Computational Condensed Matter Summer School 2025**

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**EPI Grant #12957**

# Outline

- Overview and experimental matters
  - Lightsources
  - Surface sensitivity
  - XPS
  - Accessing different parts of momentum space
  - Matrix elements
  - Resolution

- Applications/examples
  - Interactions and lifetimes
  - Gaps
  - Twists on ARPES technique
  - Throughout talk: examples from heavy fermions, cuprates, Weyl semimetals, topological insulators, unconventional superconductors

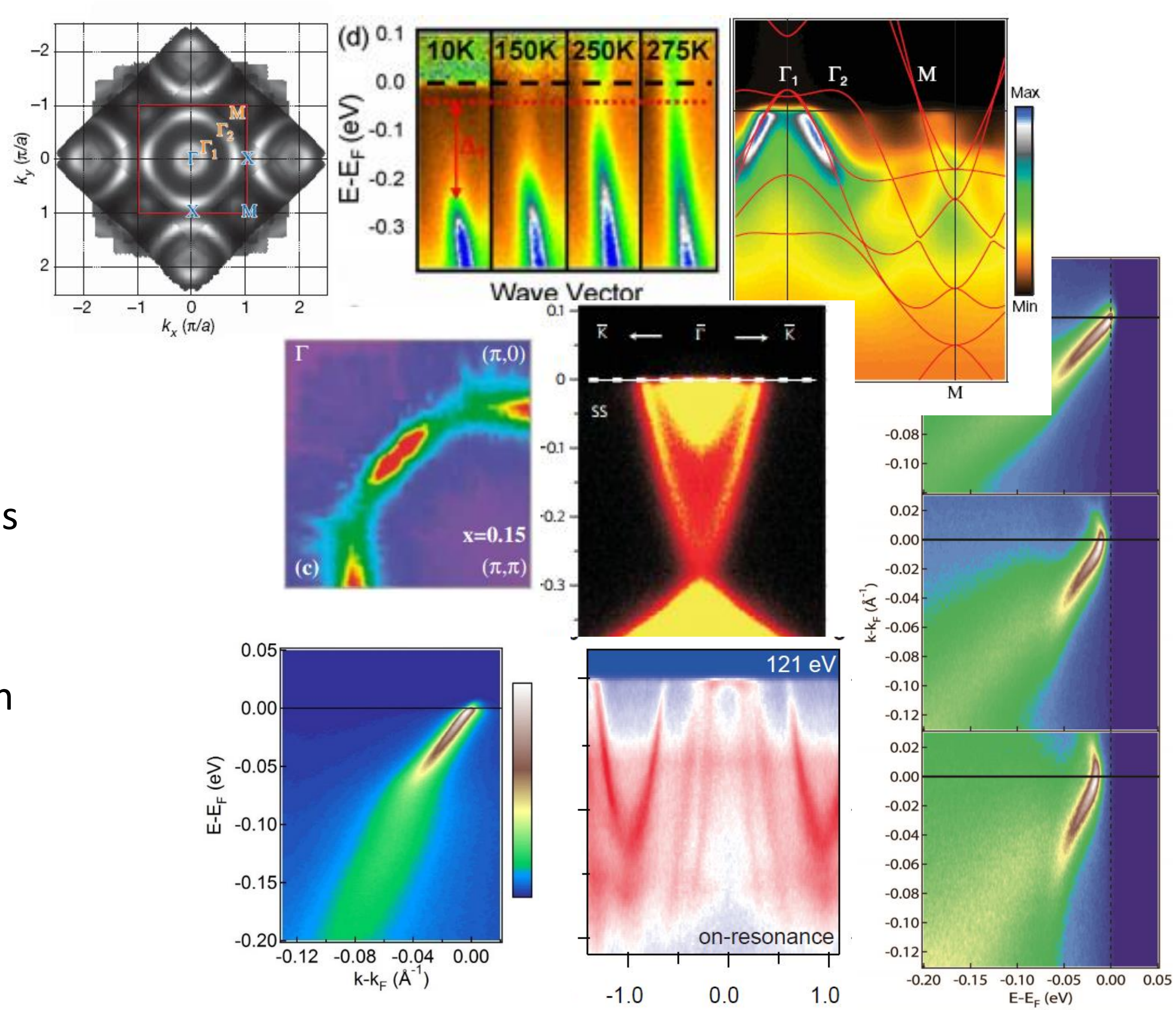
# Structures in momentum space

All crystalline materials

- Brillouin zones
- Fermi surfaces
- Band dispersion

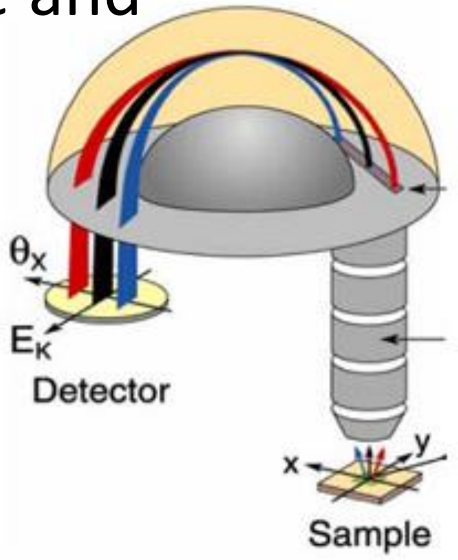
Some crystalline materials

- Charge density wave gaps
- Superconducting gaps
- Spin density wave gaps
- Magnetism that shifts bands or opens gaps
- Spin orbit coupling
- Electron-boson coupling
- Heavy fermion hybridization gaps
- Spin momentum locking
- Dirac dispersions
- Surface states
- …



# Angle-Resolved Photoemission spectroscopy overview

- Purpose: measure electronic band structure and interactions
- Photoelectric effect, conservation laws




$$E_{kin} = h\nu - \phi - \boxed{|E_B}\,|$$

$$\mathbf{p}_{||} = \hbar \boxed{\mathbf{k}}_{||} = \sqrt{2mE_{kin}} \cdot \sin \vartheta$$

Definitions:

$E_{kin} = kinetic\ energy\ of\ photoelectron$ measure

$h\nu = photon\ energy$ Know

$\phi = work\ function$ know/measure (~ 4 eV)

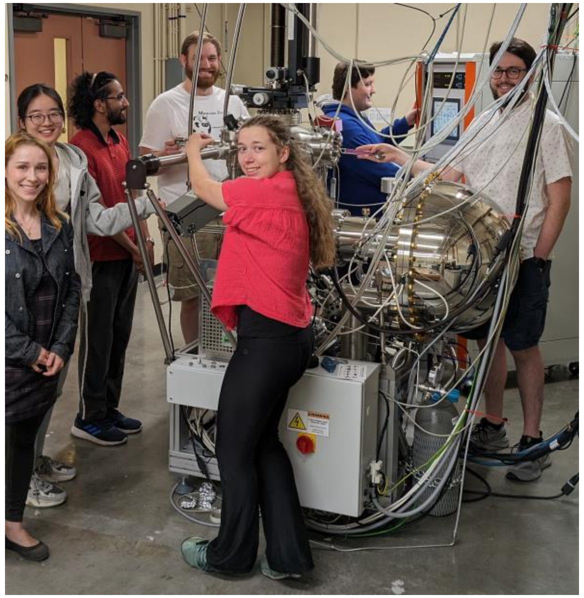
$E_B = electron\ binding\ energy\ inside\ material, relative\ to\ Fermi\ level$ want

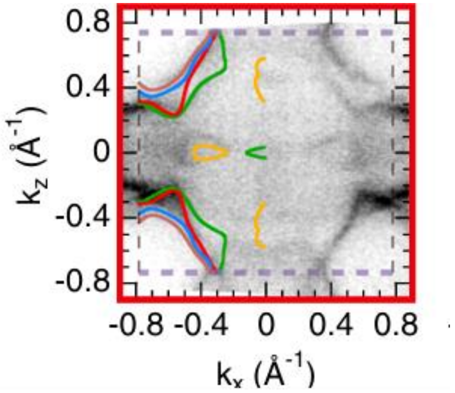
$k_{||} = crystal\ momentum, parallel\ to\ sample\ surface\ plane$ want

$m = mass\ of\ free\ electron$ know

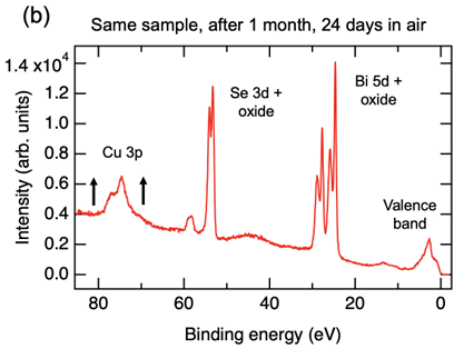
$\vartheta = emission\ angle\ of\ photoelectron$ measure

# ARPES, photoemission, and ultrafast optics at UC Davis

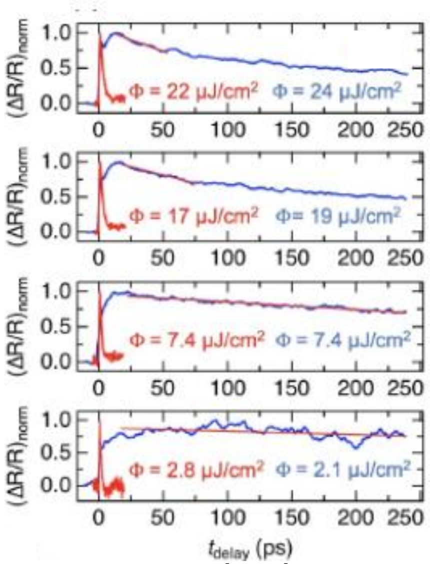


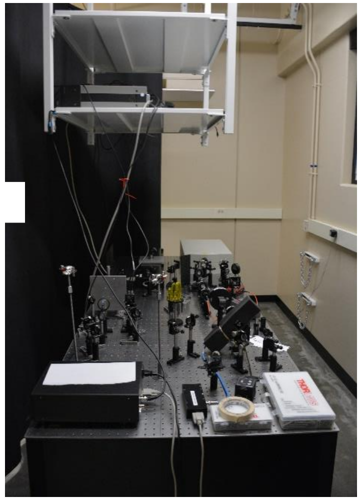



**J. Badger, Y. Quan, M. C. Staab,** *et al*, Communications Physics **5** *(2022)*
**Staab** *et al*, Phys. Rev. B 110, 165115 (2024)
*(topo superconductor $LaNiGa_2$)*

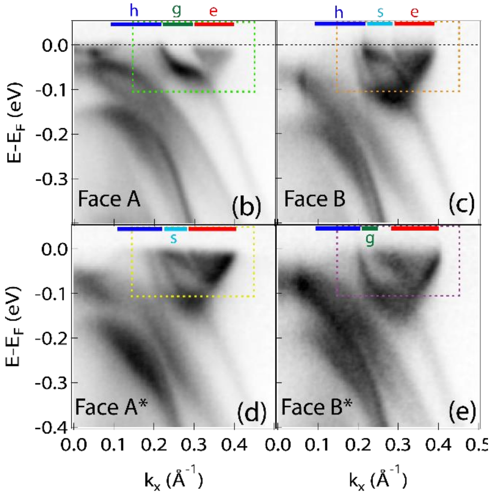



Gross *et al J. Phys. Mater.* **5** 044005 (2022) (Topo SC $Cu_xBi_2Se3$)

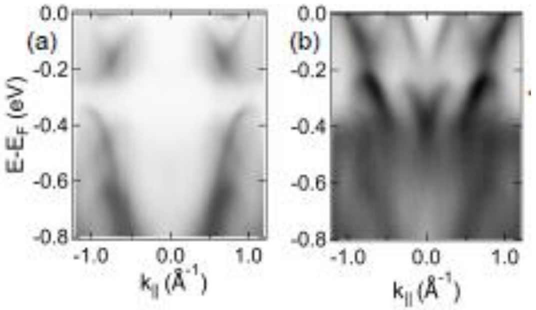


**A. Gross** *et al, Phys. Rev. B 103, L020301 (2021) (TI $Bi_{2-x}Sb_xSe_3$)*

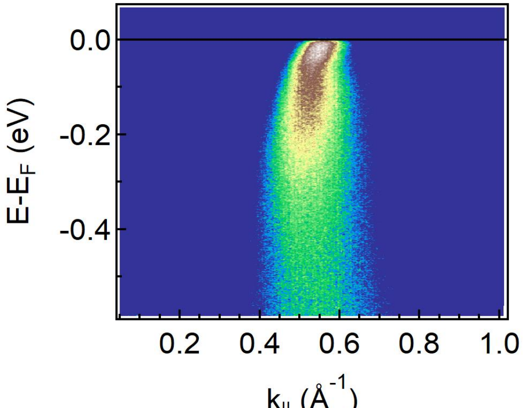



**A. Rossi**, *et al.*, Phys. Rev. B **102** (2020) (2D material $WTe_2$)



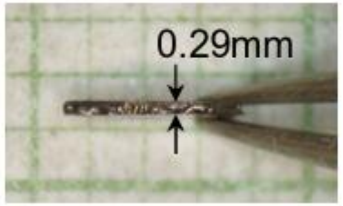

A. Rossi et al, Phys. Rev. B 104, 155115 (2021)
S. Sreedhar et al, *in preparation*
*(Magnetic Weyl semimetal $Co_3Sn_2S_2$)*

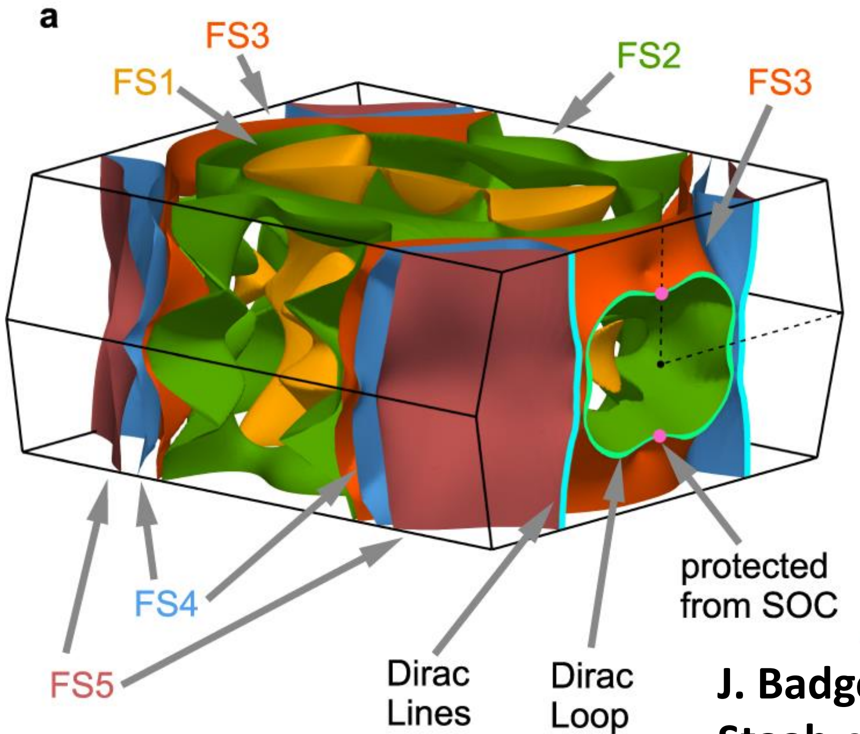
**S. Sreedhar** *et al. Phys. Rev. B 102,* 205109 (2020) (cuprate Hg1201)

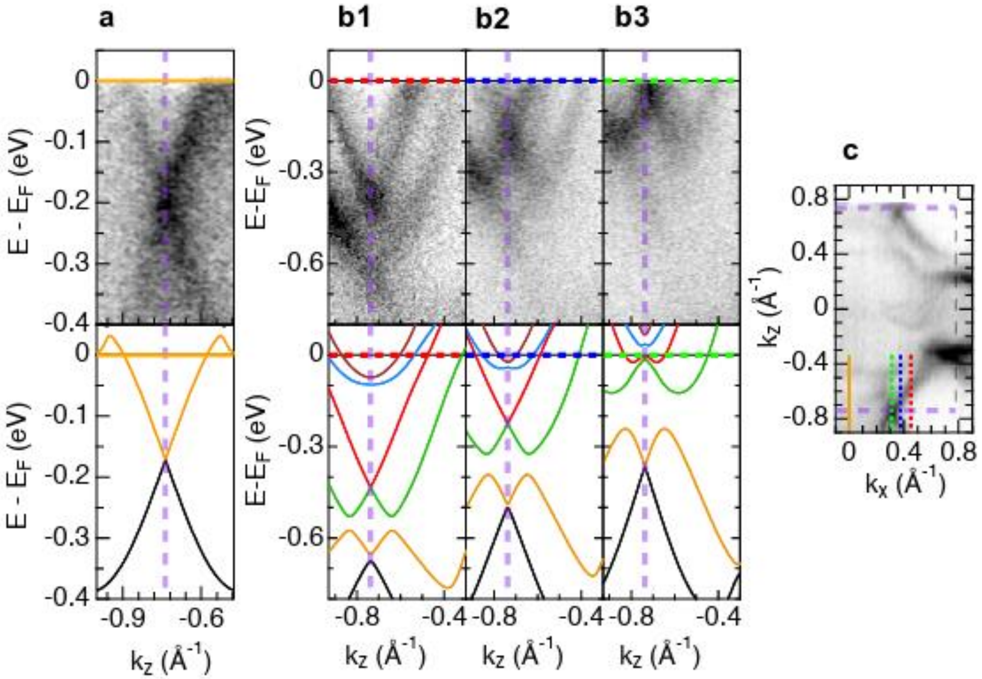


# Meet my friend $LaNiGa_2$

- Possible TRSB superconductor, $T_c$~2K
- 5 Fermi surfaces
- Nodal lines/loop/points on one face of the BZ enforced by non-symmorphic space group
- Near perfect match between DFT and expt




**J. Badger,***et al,* Communications Physics **5** *(2022)*
**Staab** *et al,* Phys. Rev. B 110, 165115 (2024

# ARPES data: Band structure, interactions, and other things

- Interactions (electron-electron, electron-phonon, etc) can change band dispersions and quasiparticle lifetimes
- Single particle spectral function captures these interactions
- Matrix elements, resolution, and sample imperfections modify the spectrum

Single particle spectral function: $$A(\mathbf{k},\omega) = -\frac{1}{\pi}\frac{\Sigma''(\mathbf{k},\omega)}{[\omega-\varepsilon_{\mathbf{k}}-\Sigma'(\mathbf{k},\omega)]^2+[\Sigma''(\mathbf{k},\omega)]^2}$$

Bare band: $\varepsilon_{\mathbf{k}}$

Self Energy: $\Sigma(\mathbf{k},\omega) = \Sigma'(\mathbf{k},\omega) + i\Sigma''(\mathbf{k},\omega)$

Band position

Linewidth or lifetime

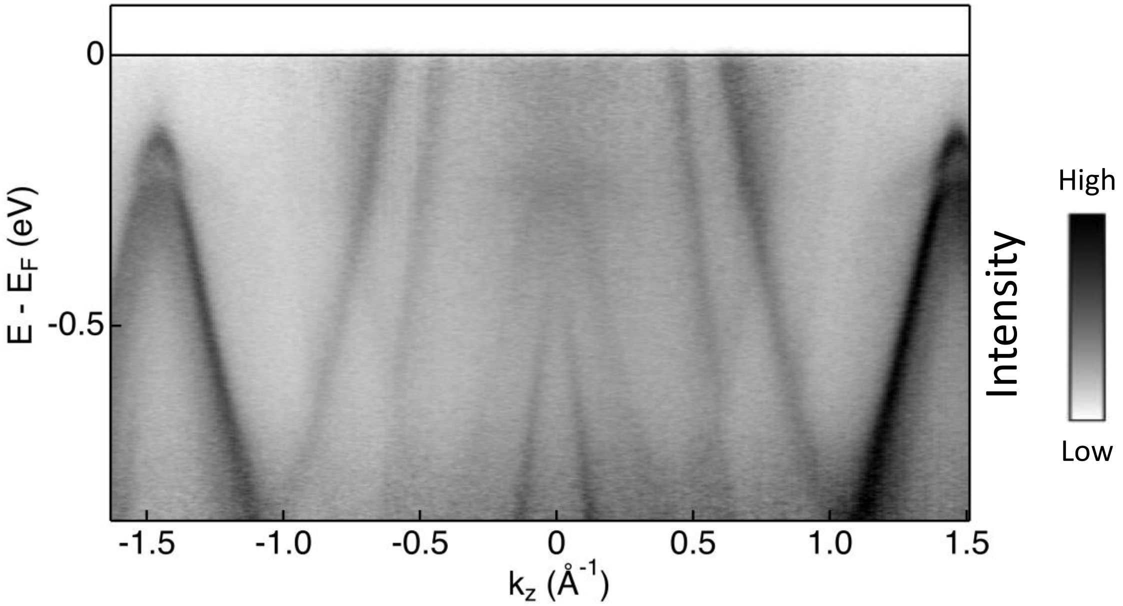
# Intensity in ARPES experiment

$$I(\boldsymbol{k},\omega) = M(\boldsymbol{k},\nu,\boldsymbol{A})A(\boldsymbol{k},\omega)f(\omega)\otimes R(\boldsymbol{k},\omega)$$

“Matrix elements”—depends on experiment details

Fermi-Dirac function: only measure occupied electron states

Resolution: contribution from instrument, experiment, and sample




Coming slides: Discuss how experimental considerations factor into each term

# Anatomy of an ARPES experiment: the lightsource

Need intense UV light for photon-hungry experiments

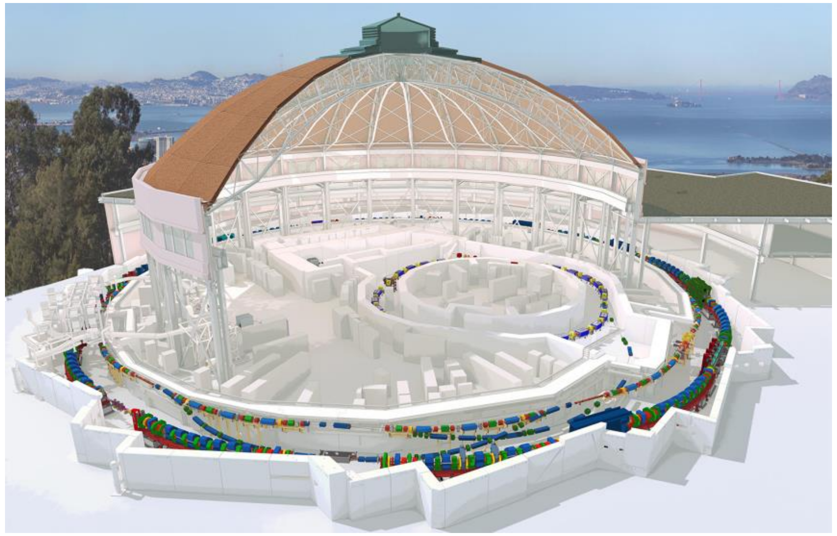
Laser (CW)

$h\nu$ small (<11 eV)

Gas discharge lamp

$h\nu$ discrete
21.2 eV (He) most common

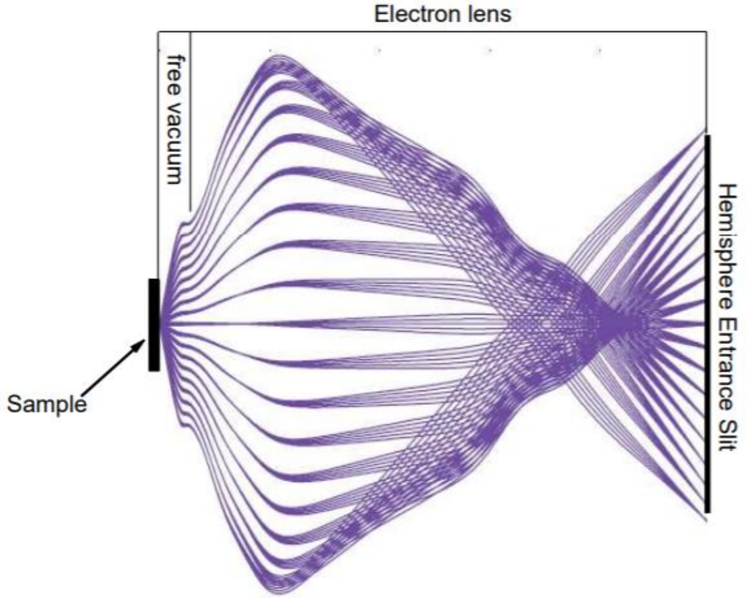
Synchrotron

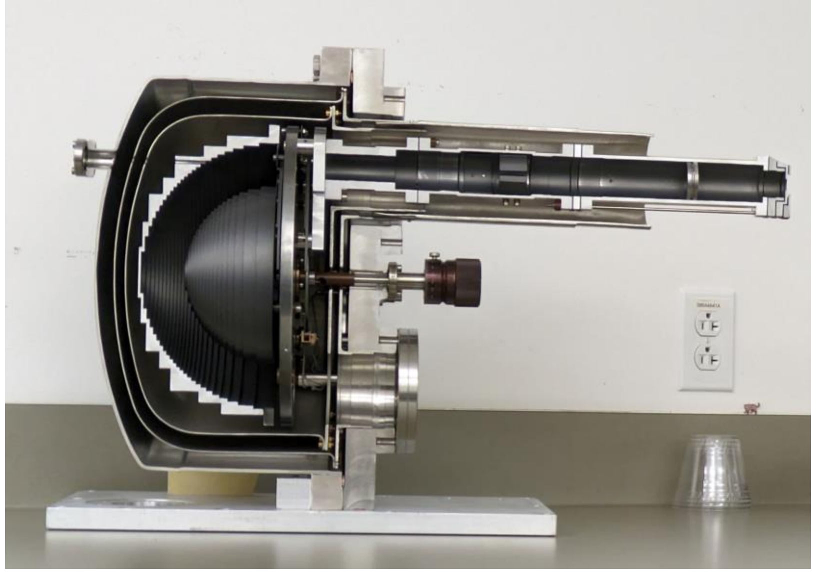


$h\nu$ variable

# Hemispherical ARPES spectrometer/analyzer

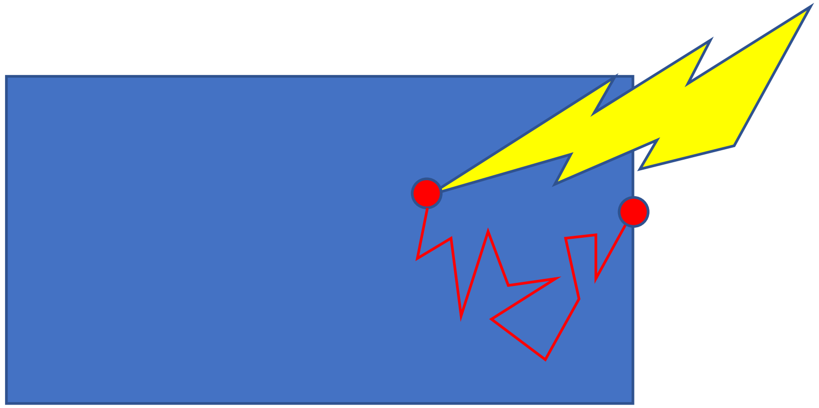
Photos from
Scienta Omicron



- Select 1D trajectory in momentum space via entrance slit
- Electrostatic lens decelerates and focuses electrons onto entrance slit
- Concentric hemispheres kept at potential difference so that electrons of different energy take different trajectory
- 2D detection of electrons, E vs k

# ARPES is a surface sensitive experiment

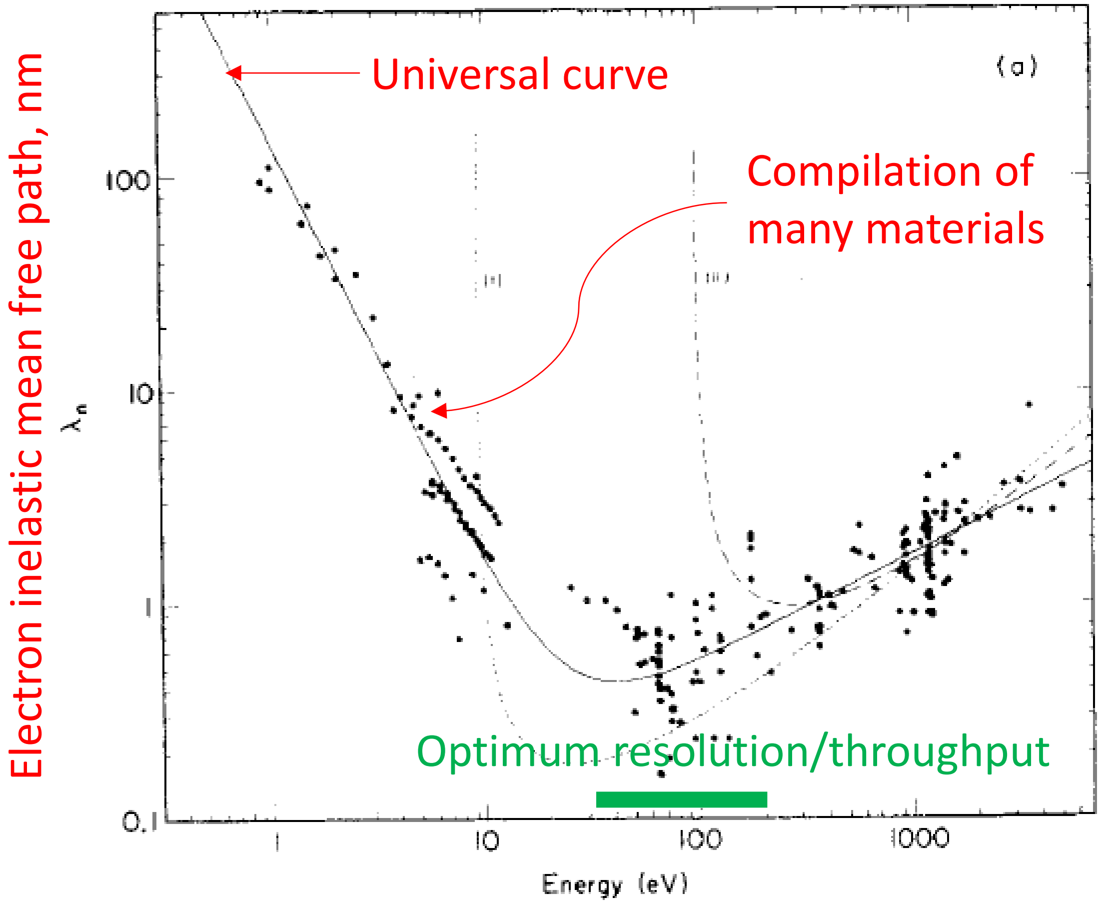


Probability of extracting electron: $\sim e^{-\frac{z}{\lambda}}$

$z = depth\ below\ surface$

$\lambda = electrons'\ inelastic\ mean\ free\ path$

- "information depth" $\approx 2\lambda$
- 98% of photoelectrons $\approx 4\lambda$




Seah and Dench, SURFACE AND INTERFACE ANALYSIS, VOL. 1, NO. 1, 1979

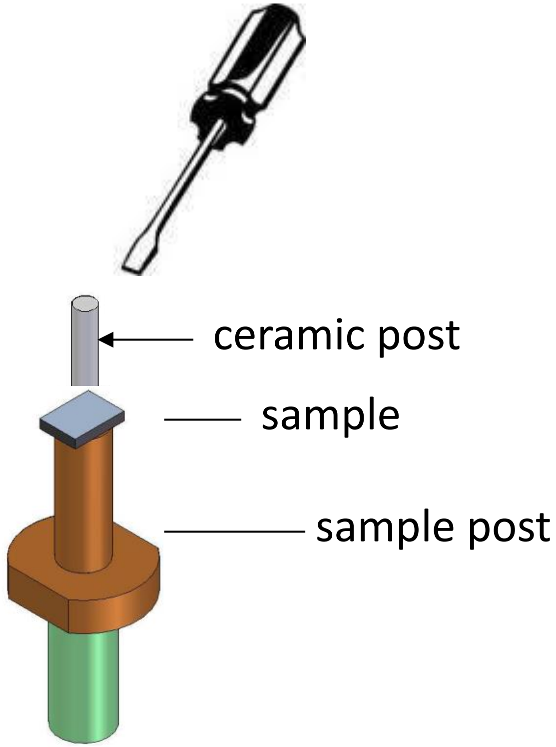
Energy=kinetic energy $\approx h\nu$ for electrons near $E_F$

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

Empirical method for estimating $\lambda$:
S. Tanuma et al, Surf. Sci. 192, L849 (1987)

# Surface sensitivity necessitates fresh surface before each experiment

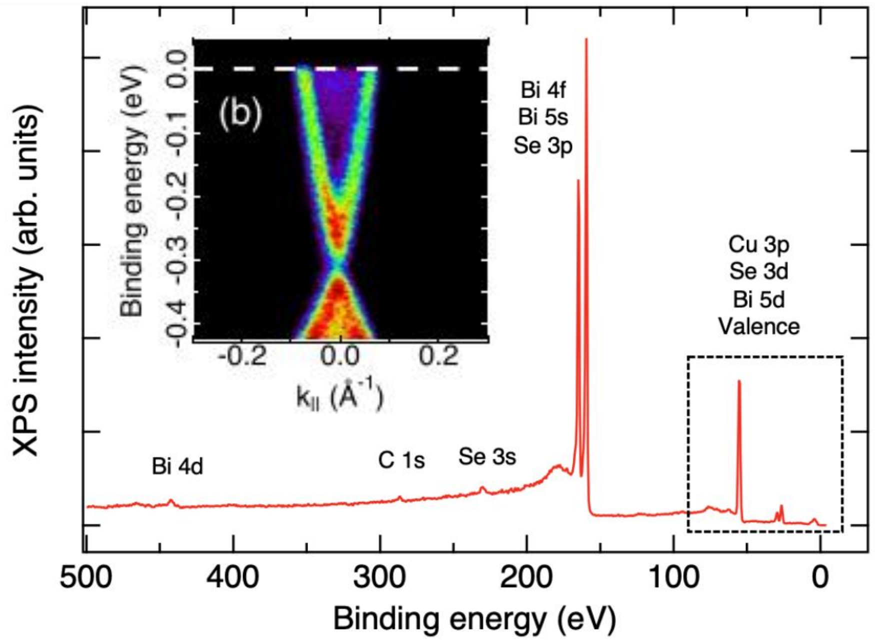
- **Cleaving in-situ**
- Growing material in-situ
- Sputter-and-anneal
- Exfoliation, if there is glove box attached to UHV
- Grow materials in external vacuum chamber and transfer via vacuum suitcase




Sample cleaving

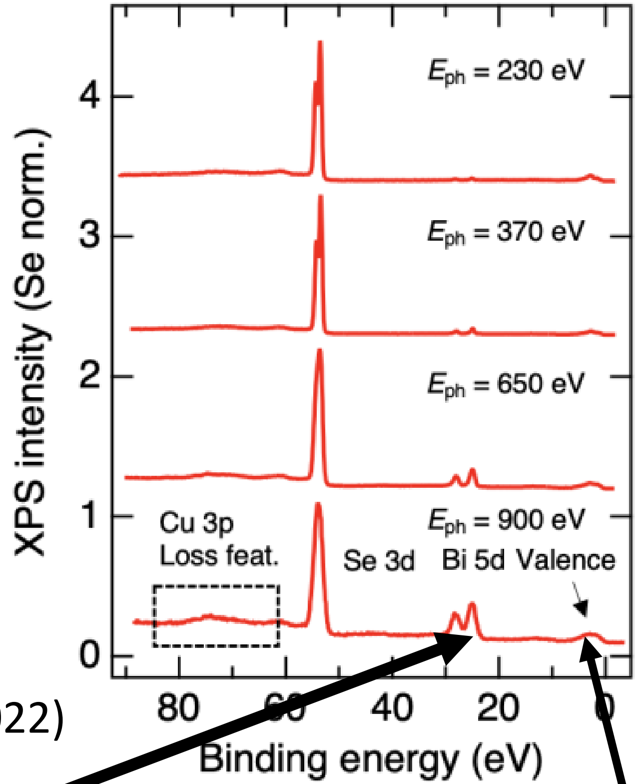
# ARPES and XPS use the same spectrometer

Material: $Cu_xBi_2Se_3$




Adam L Gross *et al J. Phys. Mater.* **5** 044005 (2022)

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




"**Core levels**"

- Localized electrons→chemistry

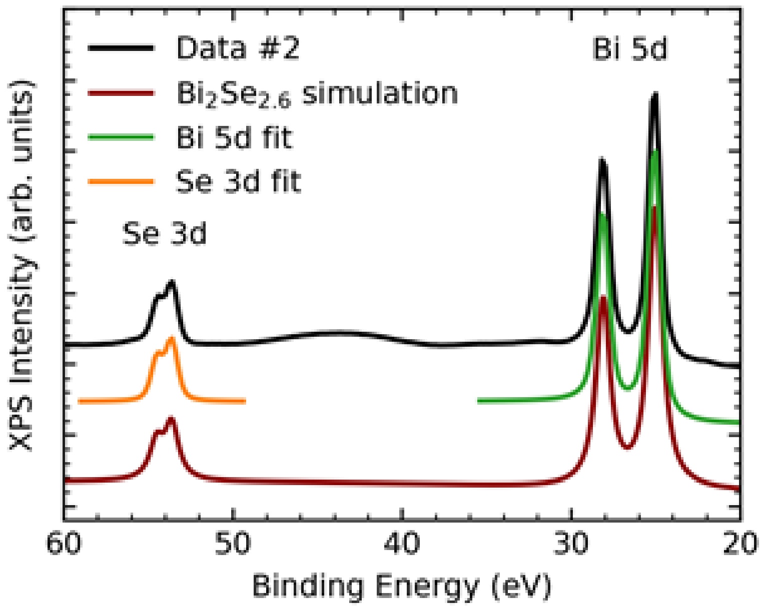
"**Valence band**"

- Occupied and delocalized states
- How electrons 'move'

xps.matthewstaab.com

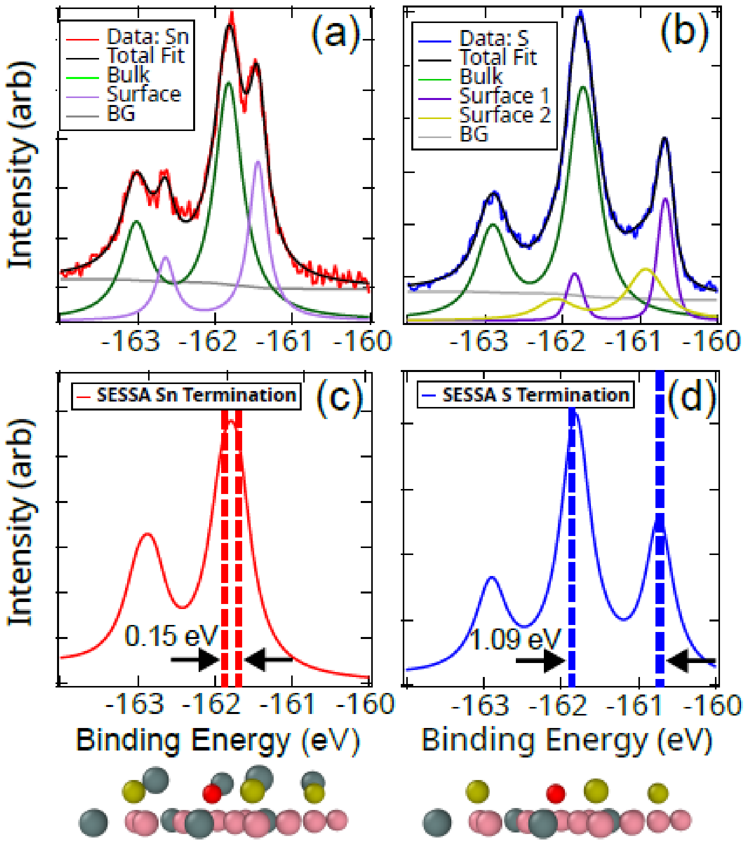
# Composition of near-surface region is knowable via XPS

Example 1: Se deficiency in $Bi_2Se_3$ surface



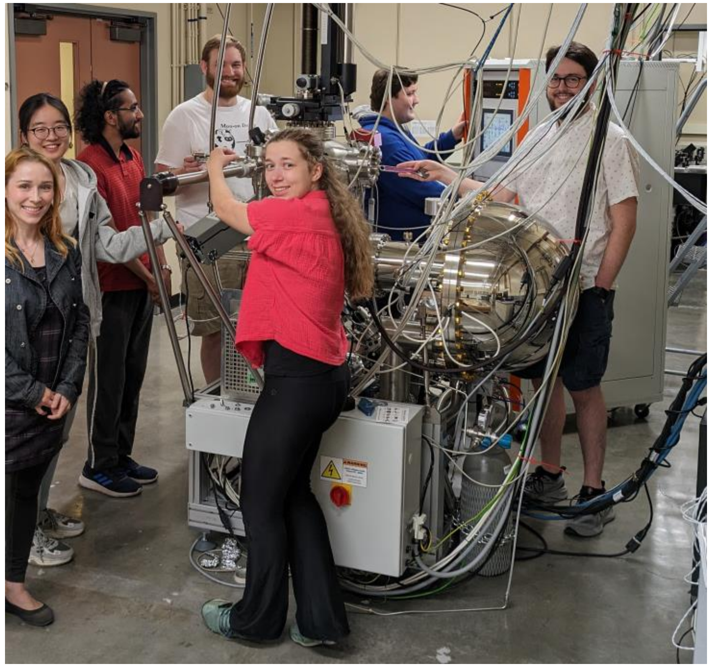

Simulation: Se and Bi core level intensities weighted by photoionization cross section and depth inside sample of each chemical species

Example 2: Sn and S terminations of $Co_3Sn_2S_2$
Differing chemical shifts on different surfaces Expt + DFT



# Ultrahigh vacuum (UHV) chamber ($< 10^{-10} torr$) to maintain surface

| | High vacuum (HV) | Ultrahigh vacuum (UHV) |
|---|---|---|
| **Pressure** | 1e-3 to 1e-9 torr | 1e-9 to 1e-12 torr |
| **Molecular mfp** | 10 cm to 1000km | 1000 to 100,000 km |
| **Amount of time to deposit a monolayer on sample surface*** | .006s to 95 minutes | 95 minutes to 65 days |



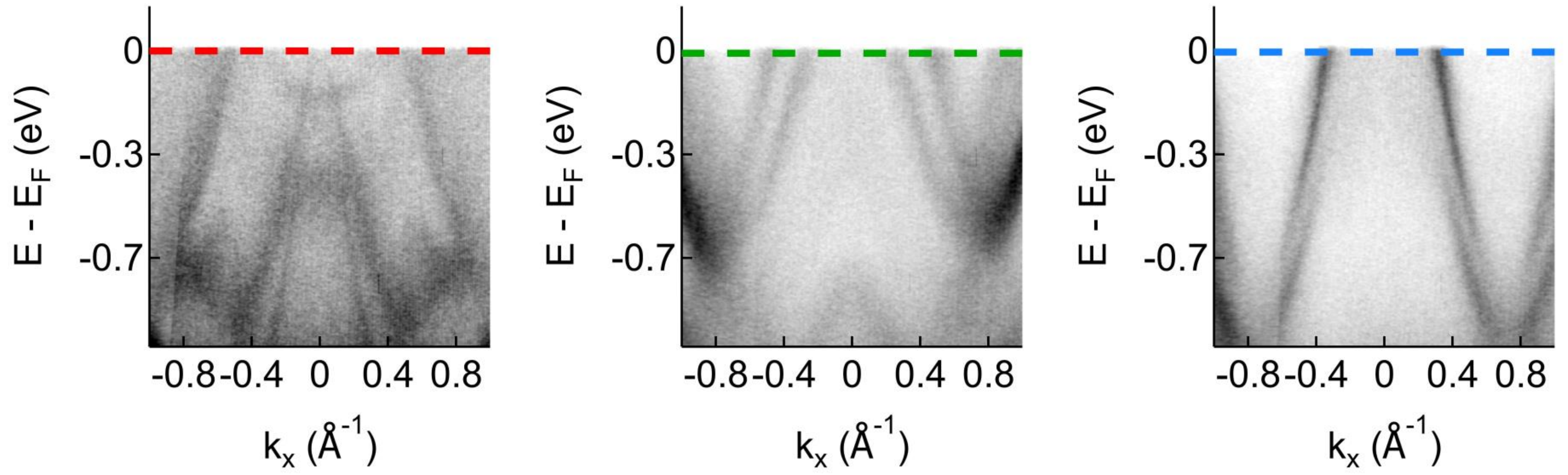
$$*t = \frac{1.7\times10^{-6}}{0.6 * p * S}$$

p=pressure in torr
S=sticking coefficient (between 0 and 1)
Ref: Hufner, *Photoelectron Spectroscopy*

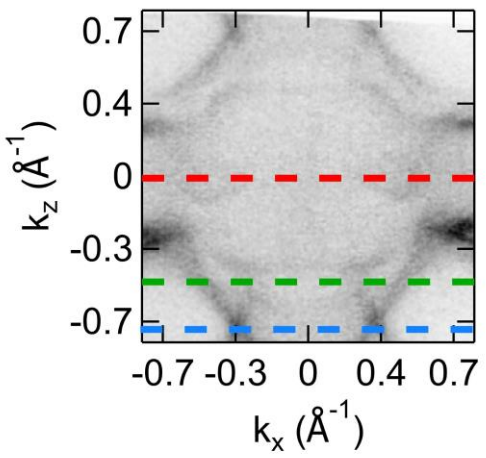
- UV light + surface adsorbates = surface chemistry
- Light elements can move and evaporate
- Fresh surfaces typically survive <48 hours in ARPES experiment

# From two to three dimensional data sets

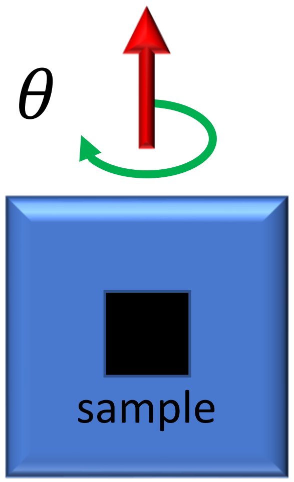
Quantum of ARPES data from hemispherical analyzer is energy vs (1D) momentum "cut"

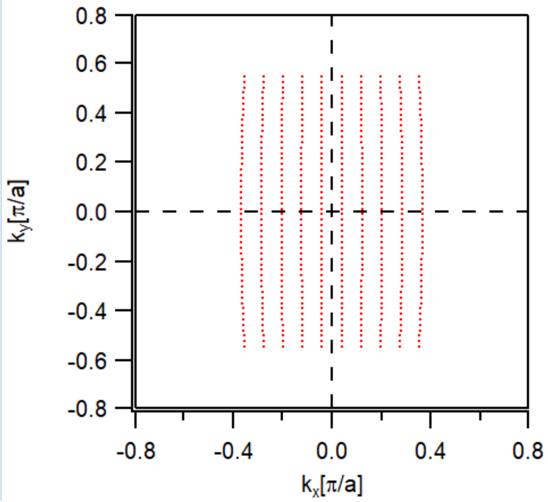



- Get energy vs 2D momentum by pasting many 1D cuts together
- This 3D data usually presented as constant-energy maps ($k_x$ vs $k_y$)
- Constant-energy map at $E_F$ is Fermi surface

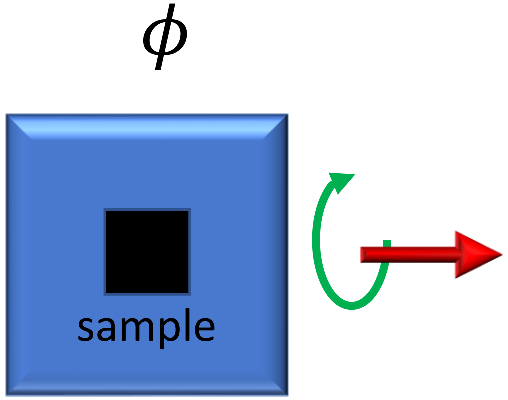


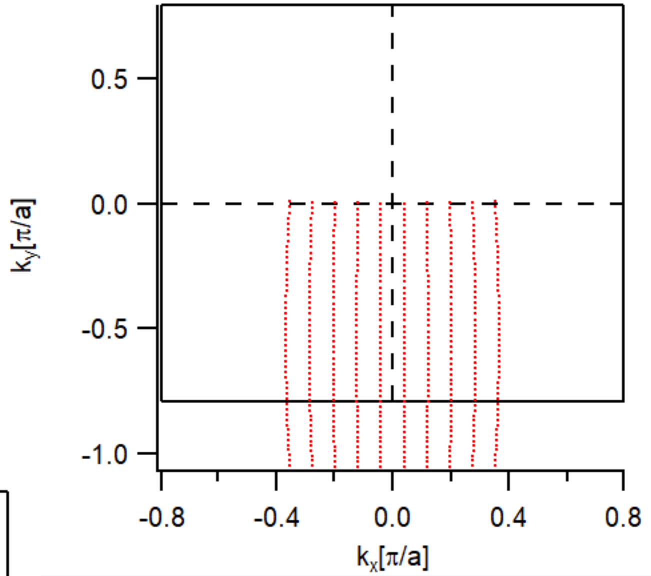
# Accessing different parts of the Brillouin zone (parallel momentum)

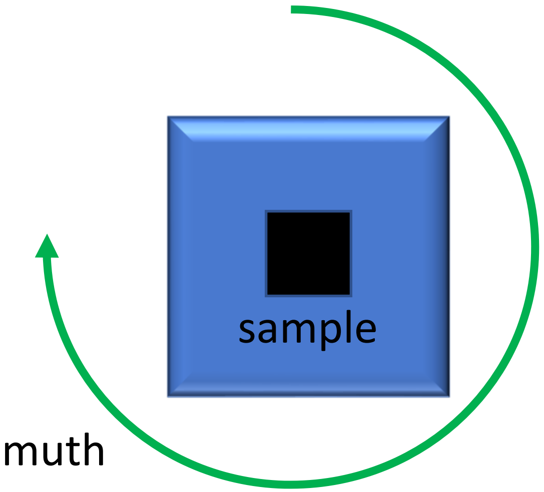


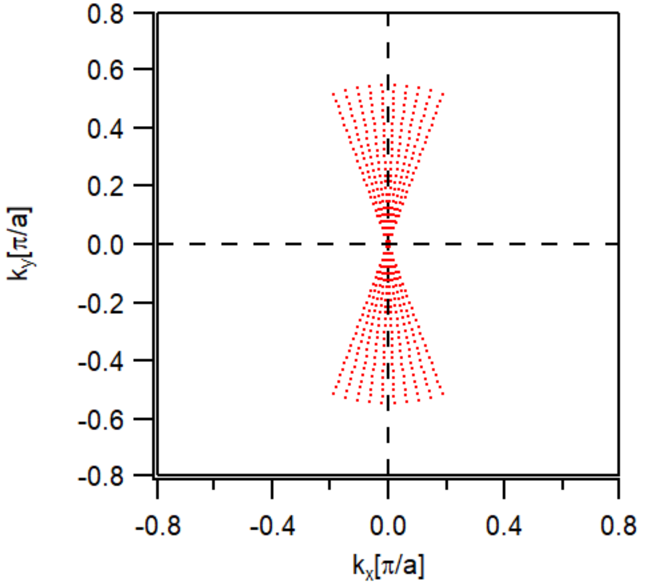

Can also do ‘deflector’ scan to allow equivalent of $\theta$ rotation without moving sample




$\omega$ or azimuth




$$\boxed{\hbar k_{||} = \sqrt{2mE_{kin}}\,\sin\vartheta}$$

# “Final states”

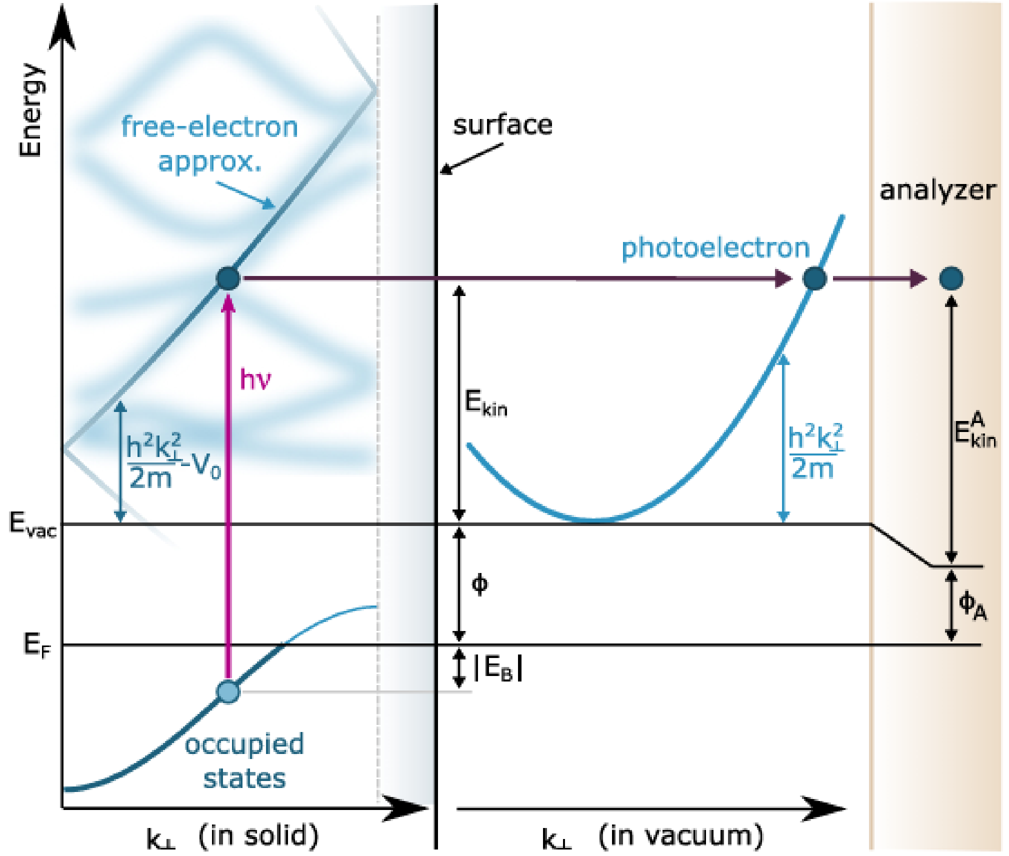



Image source: Sobota *et al,* Rev. Mod. Phys. 93 (2021)

Probability of electron excitation related to Fermi’s golden rule:

$$w_{fi} = \frac{2\pi}{\hbar} \left|< \Psi_f^N \right| - \frac{e}{mc} \boldsymbol{A} \cdot \boldsymbol{p} |\Psi_i^N > \Big|^2 \delta(E_f^N - E_i^N - h\nu)$$

**p**=electron momentum
**A**=vector potential of photon (points in direction of polarization)
$\Psi_{i,f}^N$ =initial,final N-electron wavefunction

One-step model: photon absorption, electron excitation, and electron detection are treated as a single coherent process

Final states are usually not of direct interest, but they have implications for:

- Perpendicular momentum (assessing 3D electronic structure)
- “matrix element effects”: bands being highlighted or diminished based on experimental details

# Accessing different parts of the Brillouin zone (perpendicular momentum)

$$\hbar \boldsymbol{k}_{\parallel} = \sqrt{2mE_{kin}} \cdot \sin\vartheta$$

In photoemission, only in plane momentum is conserved, but electrons have out-of-plane momentum too

$$E_f(\boldsymbol{k}) = \frac{\hbar^2 \boldsymbol{k}^2}{2m} - |E_0| = \frac{\hbar^2\left(\boldsymbol{k}_{\parallel}^2 + \boldsymbol{k}_{\perp}^2\right)}{2m} - |E_0|$$

Use different photon energies to access different $k_{\perp}$

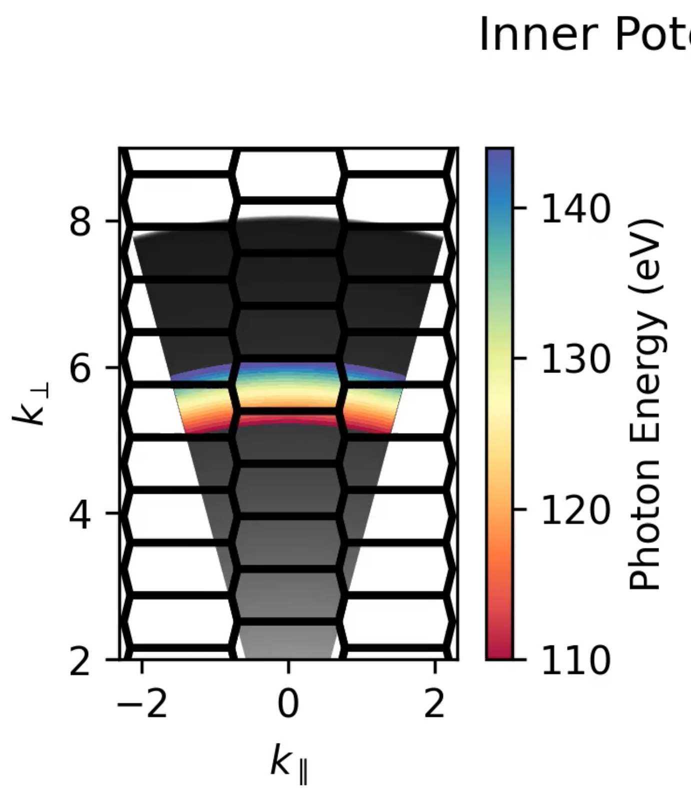
Final state energy
$h\nu - |E_B|$
$E_{kin} + \phi$

~Bottom of valence band

Solve for $\boldsymbol{k}_{\perp}$

$$\boldsymbol{k}_{\perp} = \frac{1}{\hbar}\sqrt{2m(E_{kin}\cos^2\vartheta + V_0)}$$

$V_0 = |E_0| + \phi$ (inner potential)

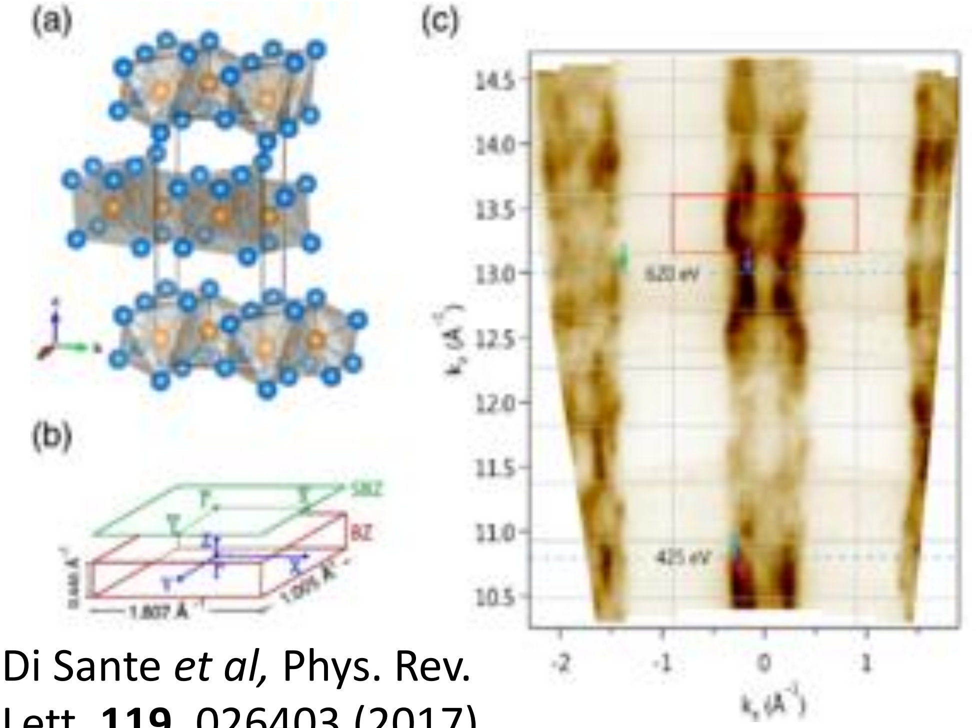



Resource for $k_{\perp}$: Damascelli, Physica Scripta. Vol. T109, 61–74 (2004)

# How to ascertain inner potential

Significance: Many experiments try to target specific planes of BZ (e.g. high symmetry planes, Weyl points), so connection between photon energy and BZ plane must be established

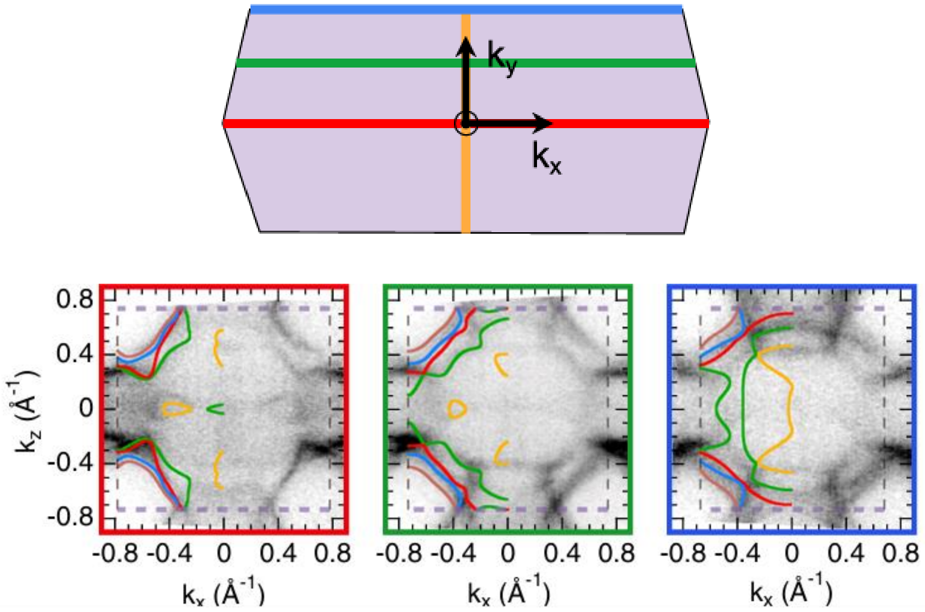
Measure multiple Brillioun zones and find periodicity




Di Sante *et al*, Phys. Rev. Lett. **119**, 026403 (2017)

Match to calculation

$LaNiGa_2$




$V_0 = 15$

Guess a number 10-15 eV

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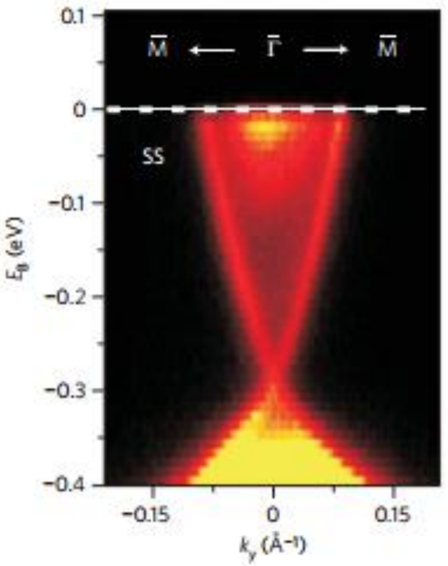
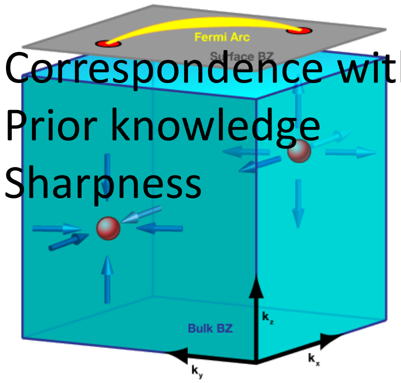
# Distinguishing surface vs bulk electronic structure

e.g. 1: Topological insulator




Xia *et al.* Nat. Phys. **5** May 2009

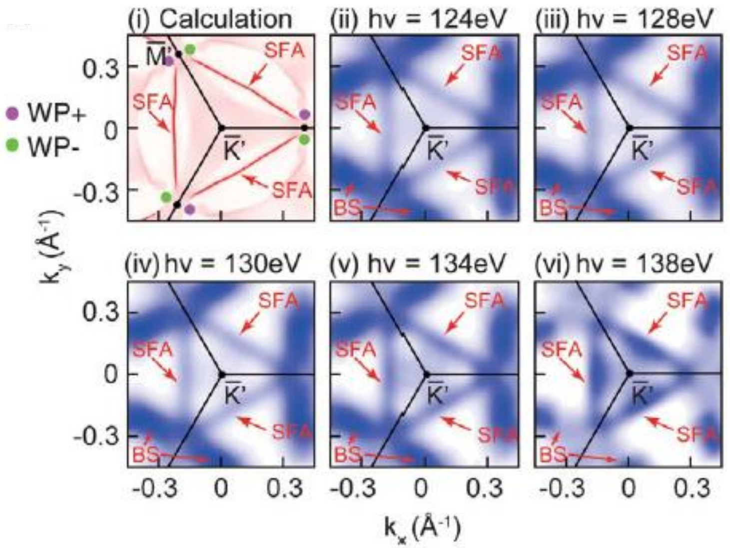
E.g.2 Weyl semimetal

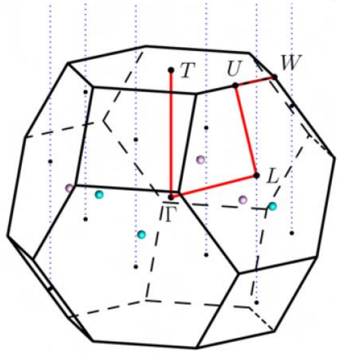



- Correspondence with theory
- Prior knowledge
- Sharpness

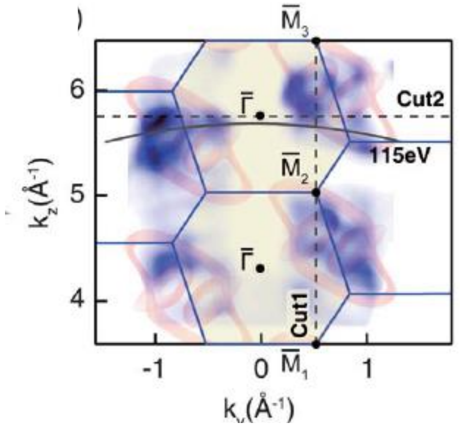
$Co_3Sn_2S_2$: magnetic Weyl semimetal

2D Surface Fermi arcs do not disperse with photon energy

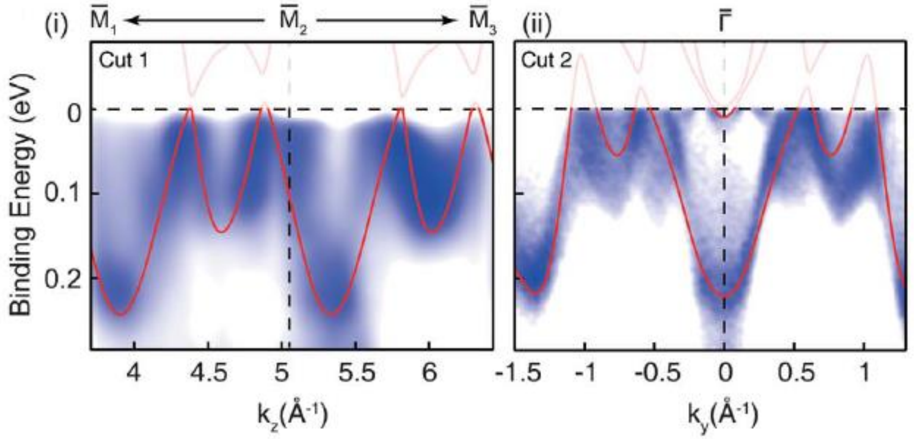



3D Bulk Weyl nodes accessed by varying photon energy




*vary hν*




Liu et al., Science 365, 1282–1285 (2019)

# Matrix element effects

Express many-electron WF as antisymmetric product of 1-electron state and N-1 electron state

$$\Psi_f^N = \mathcal{A}\phi_f^{\boldsymbol{k}}\Psi_f^{N-1}$$

$$<\Psi_f^N \left| -\frac{e}{mc}\boldsymbol{A}\cdot\boldsymbol{p} \right| \Psi_i^N > = < \phi_f^{\boldsymbol{k}} | -\frac{e}{mc}\boldsymbol{A}\cdot\boldsymbol{p} | \phi_i^{\boldsymbol{k}} >< \Psi_m^{N-1} | \Psi_i^{N-1} >$$

$$\equiv M_{f,i}^{\boldsymbol{k}} < \Psi_m^{N-1} | \Psi_i^{N-1} >$$

$M_{f,i}^{\boldsymbol{k}}$= 'ARPES matrix elements'

*m*=index given to N-1-electron **excited** state

Consequence: intensity of bands depends on their orbital character + experimental details (**photon energy, polarization, geometry**)

- Sometimes just incidental fact
- Can be exploited to highlight certain bands and/or discern their orbital character

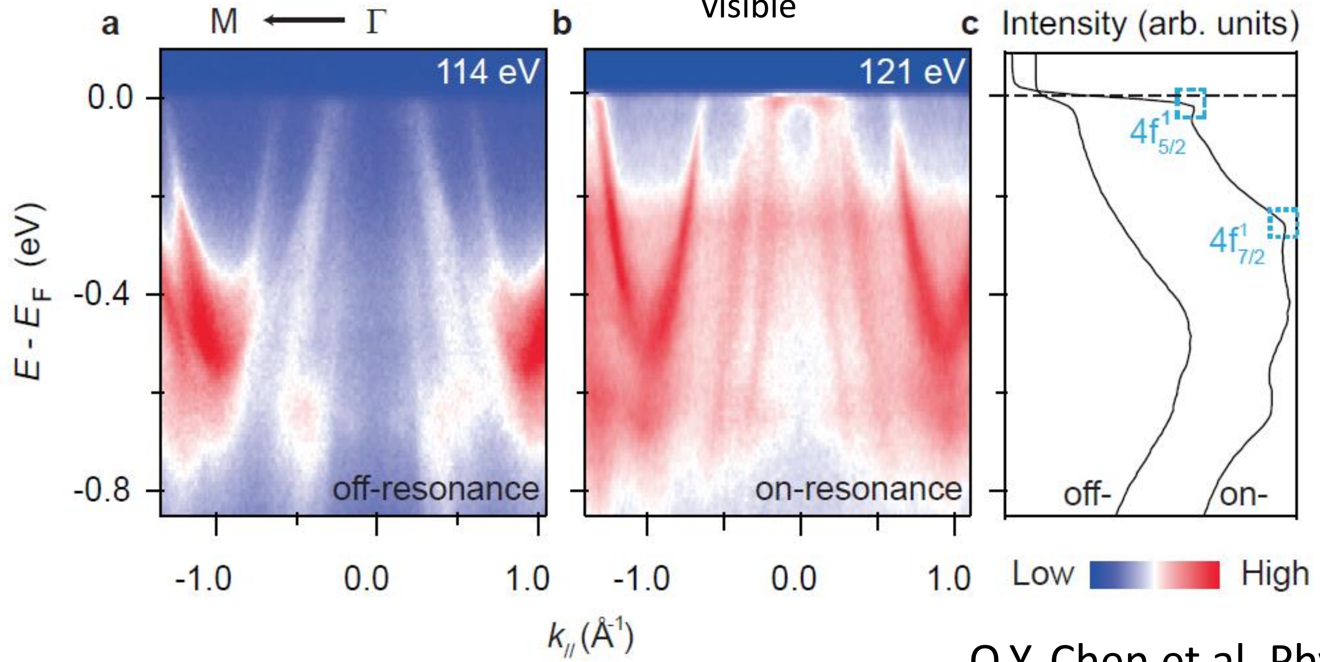
S. Moser, Journal of Electron Spectroscopy and Related Phenomena **214** 29–52 (2017)

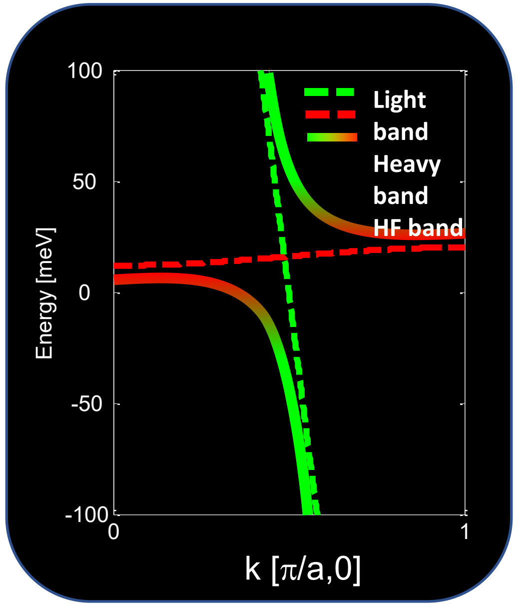
# Example: highlighting ‘heavy’ bands

- Different parts of hybridized band structure originate from different orbitals
- $Ce^{3+}$ corresponds to a $4f^1$ electronic configuration.
- At 121 eV photon energy, there is resonance between a “core” 4d state and the 4f state.

114 eV: Photoemission dominated by Co 3d and Ce 5d states.

121 eV: 4f states visible



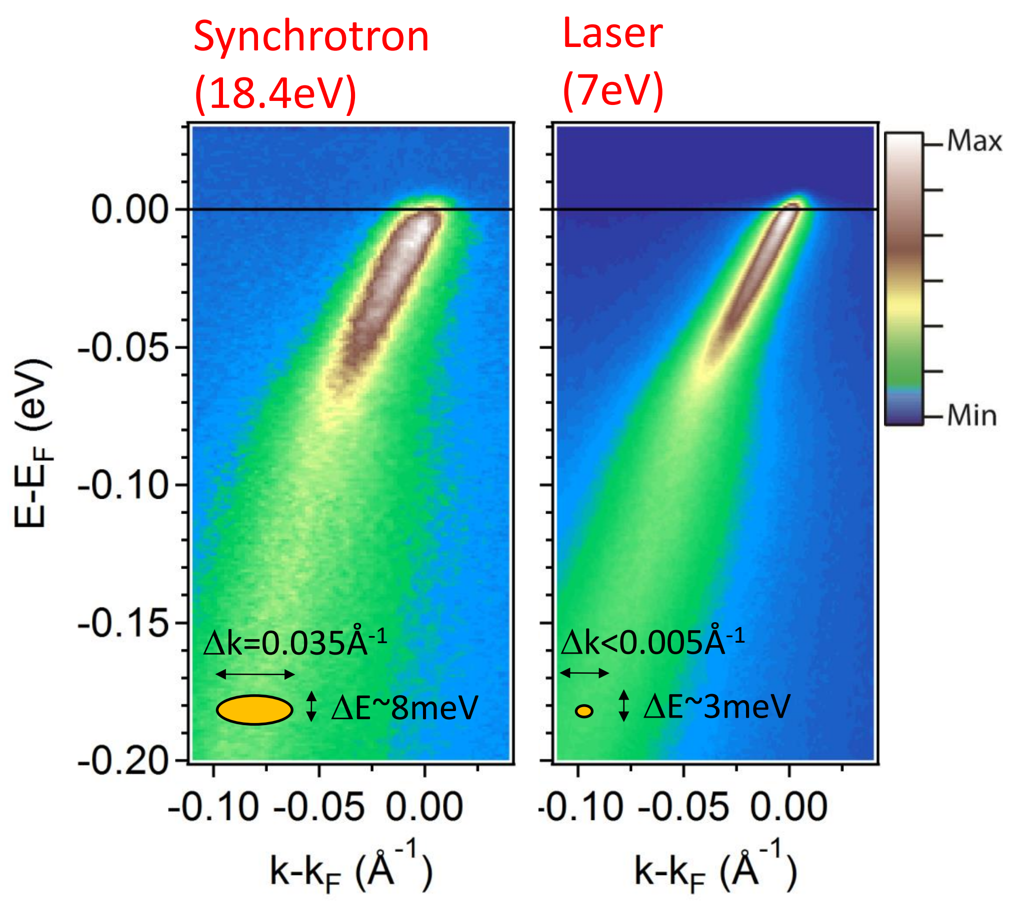



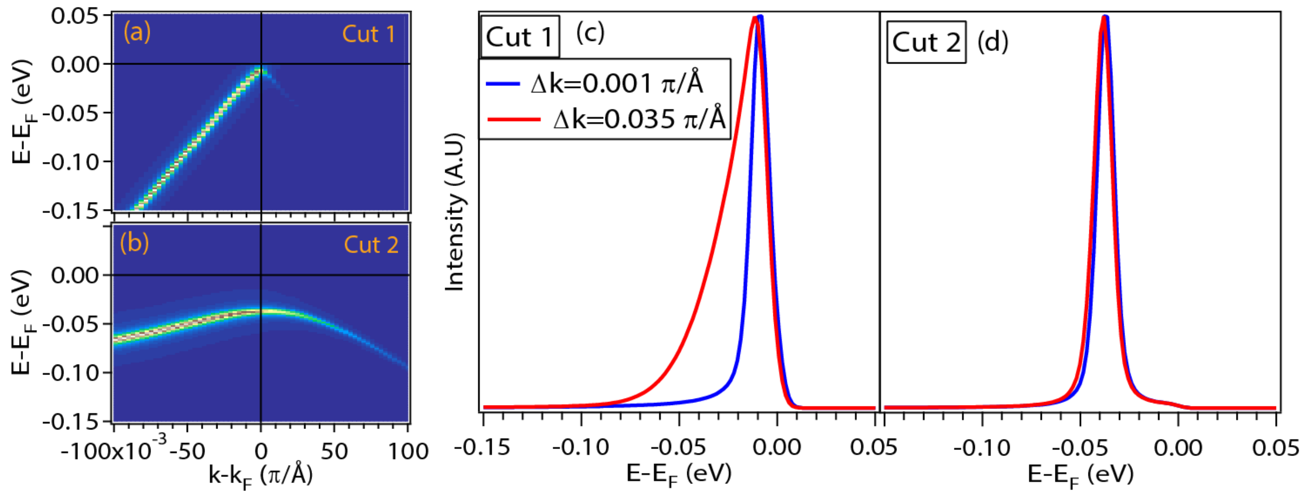



Q.Y. Chen et al. Phys. Rev. B **96**, 045107 (2017)

# Resolution effects: energy and parallel momentum

- Instrument resolution: convolution of spectral function with resolution ellipsoid. It does not represent the smallest energy or momentum scale which can be resolved
- Tradeoffs to achieving better resolution which may be unacceptable for some experiments
- In strongly dispersing bands, momentum resolution can lead to energy broadening
- Most experiments are not operated at optimal/best resolution of instrument



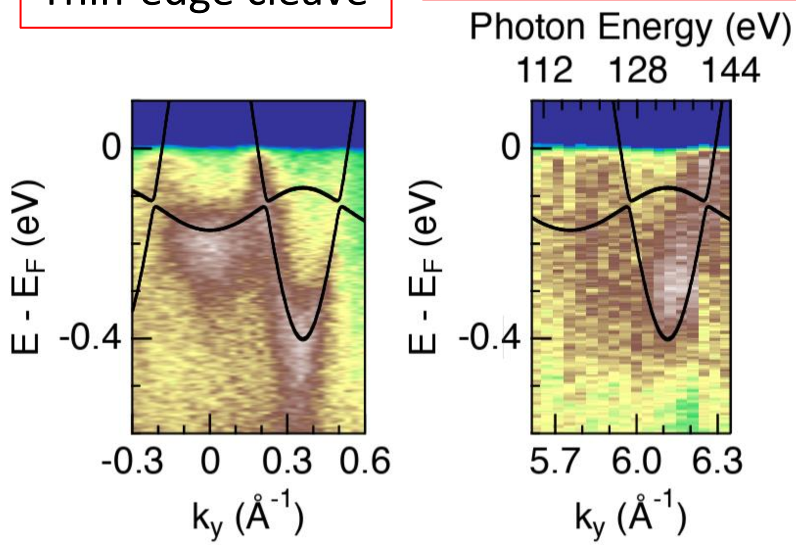



# Resolution effects: perpendicular momentum

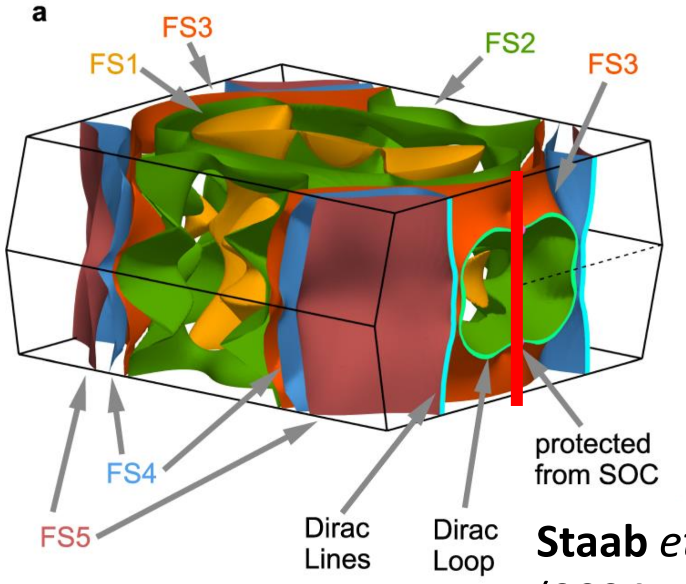
- Photoelectrons are known to originate from a short depth inside the sample (e.g. $\lambda = 5\text{Å}$)
- Short probing depth leads to quantum uncertainty in perpenidular momentum (e.g. $\Delta k_{\perp} = 0.2\ \text{Å}^{-1}$)
- $\Delta k_{\perp}$ can be large fraction of BZ
    - $k_{\perp}$ dispersion becomes changed/muted
    - Precise determination of $V_0$ less critical for very surface sensitive experiments

Thin-edge cleave

Photon energy reconstruction







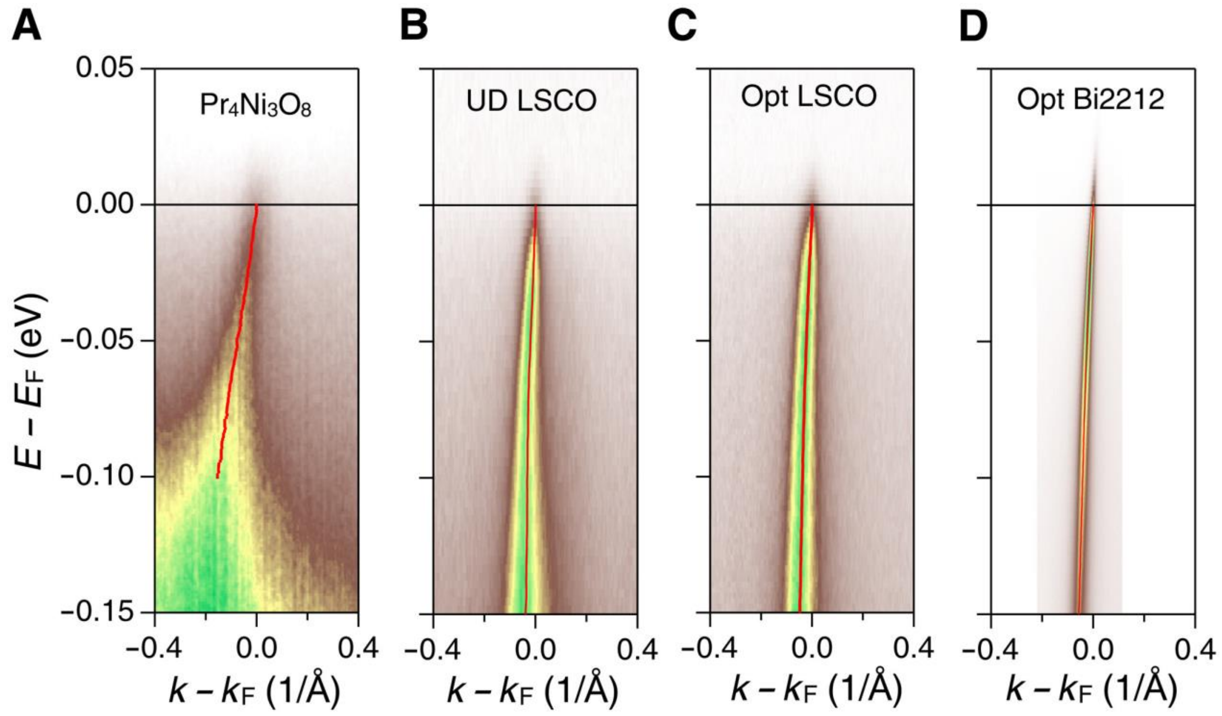

Resource for $k_{\perp}$ broadening
V.N. Strocov Journal of Electron Spectroscopy and Related Phenomena 130 65–78 (2003)

**Staab** *et al,* Phys. Rev. B 110, 165115 (2024

# Resolution effects: other sources

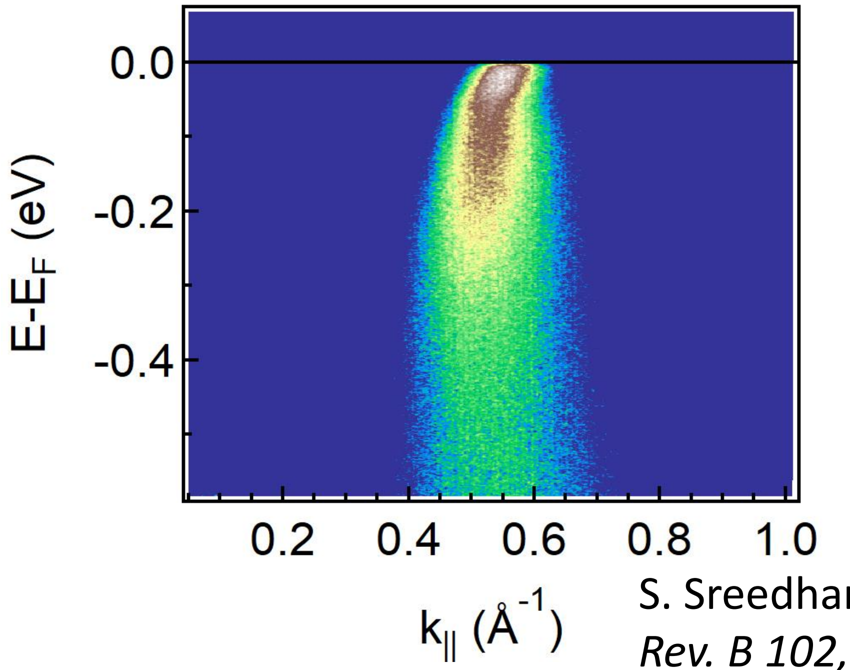
- Sample inhomogeneity
  - Surface curvature or roughness
  - Chemical inhomogeneity
- Experimental un-idealities
  - Larger beam spot size
  - Electrical noise
  - Bandwidth of lightsource
  - Finite width of entrance slit
  - Space charging (electrons repelling each other after photoemission)

# Sometimes the sample limits the precision of information ARPES can extract



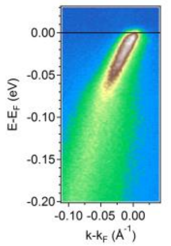

Haoxiang Li *et al.*, *Sci. Adv.* **9**, 4418 (2023)

$HgBa_2CuO_4$: “clean” cuprate




S. Sreedhar *et al. Phys. Rev. B 102,* 205109 (2020)

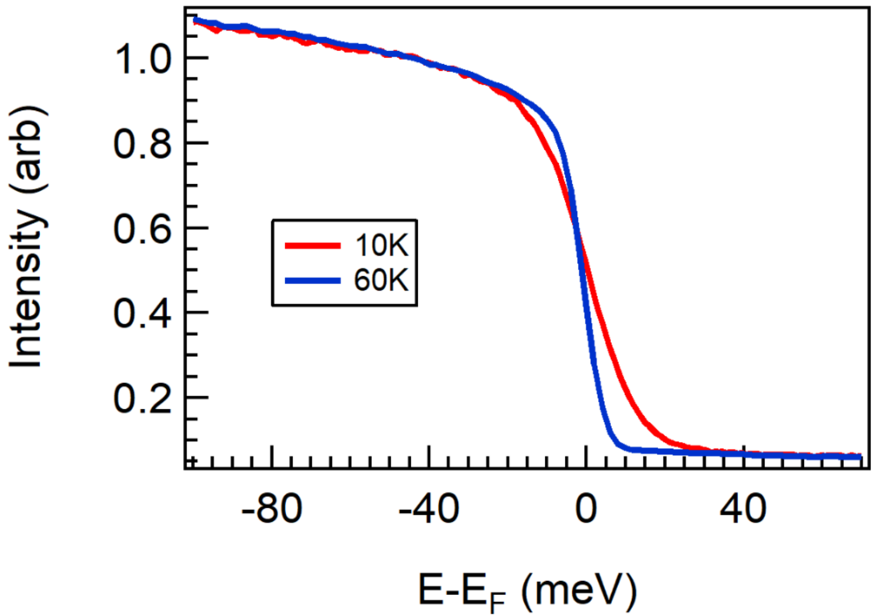
$Bi_2Sr_2CaCu_2O_8$: “dirty” cuprate
(images to scale)



# Temperature

$$I(\mathbf{k},\omega) = I_0(\mathbf{k},\nu,\mathbf{A})\boxed{f(\omega)}\boxed{A(\mathbf{k},\omega)}\otimes R(\Delta k,\Delta\omega)$$

- Fermi-Dirac cutoff gets broader giving access to more unoccupied states
- Spectra get broader, generally following electron lifetime of material system

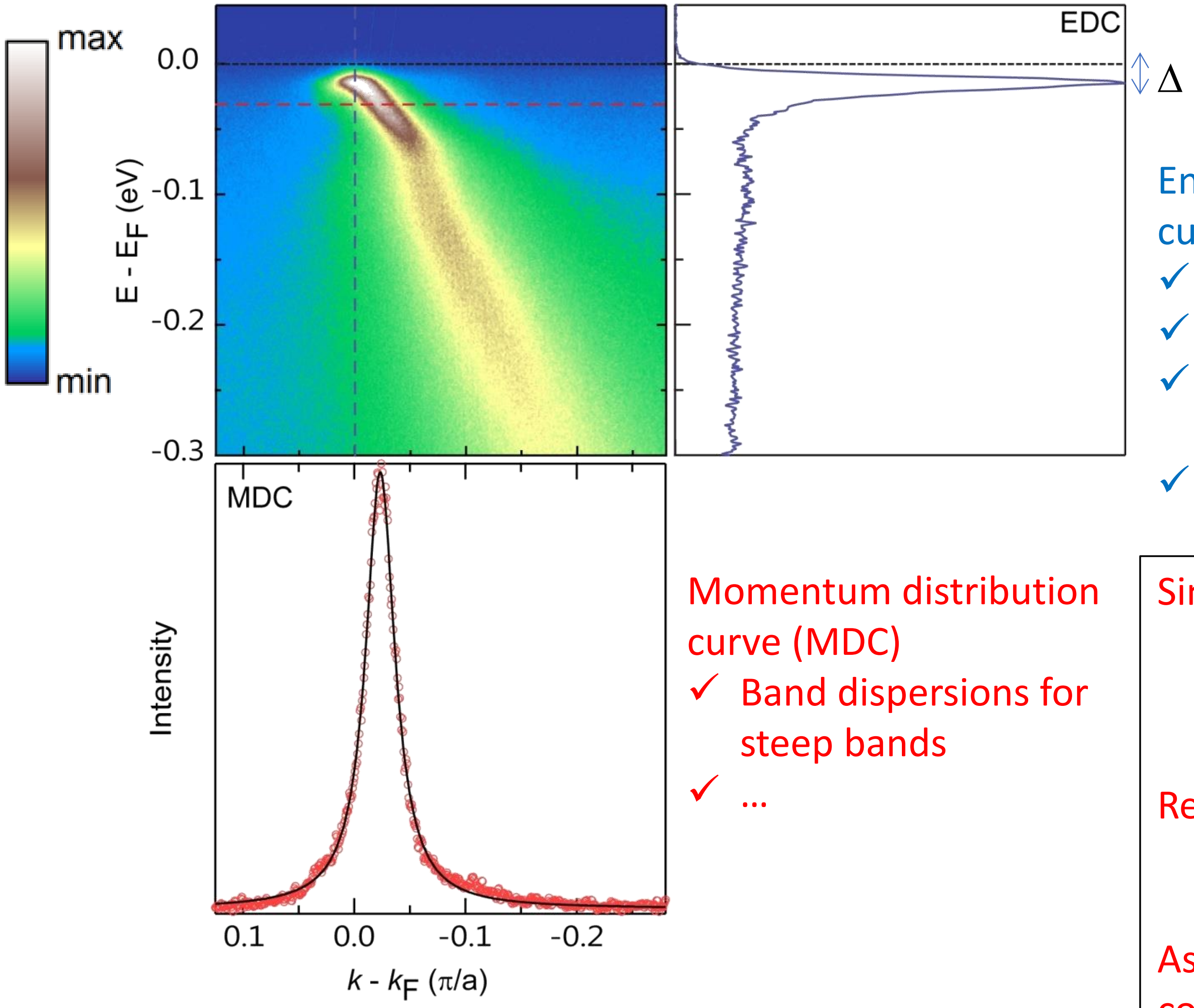



Polycrystalline Au often used as Fermi level reference

Temperature control during experiment:

- Flow cryostat
- Typical minimum temperature: 5-20K
- He-3 systems capable of reaching 1Kor below exist but are rare
- <1K operation also requires energy resolution better than 1 meV, which is challenging

# Slicing up ARPES data: terminology




Energy distribution curve (EDC)

- ✓ Spectral gaps
- ✓ Lineshapes
- ✓ Band dispersion for flatter bands
- ✓ …

Momentum distribution curve (MDC)

- ✓ Band dispersions for steep bands
- ✓ …

Single particle spectral function:

$$A(\boldsymbol{k},\omega) = -\frac{1}{\pi}\frac{\Sigma''(\boldsymbol{k},\omega)}{[\omega-\varepsilon_{\boldsymbol{k}}-\Sigma'(\boldsymbol{k},\omega)]^2 + [\Sigma''(\boldsymbol{k},\omega)]^2}$$

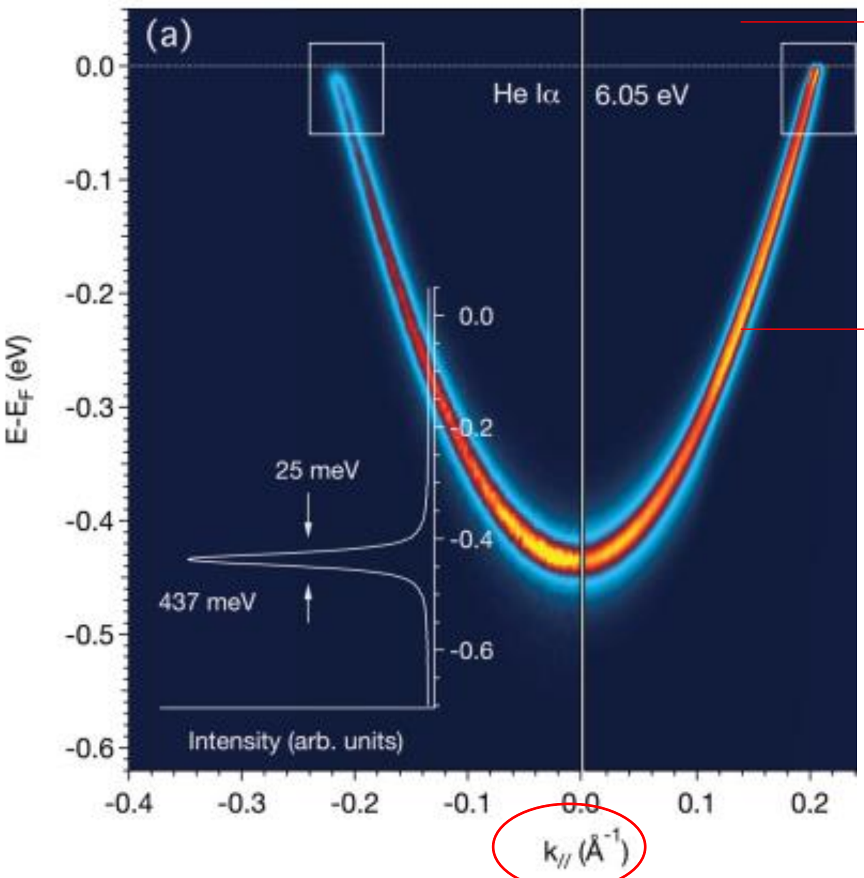
Real and imaginary part of self energy:

$$\Sigma(\boldsymbol{k},\omega) = \Sigma'(\boldsymbol{k},\omega) + i\Sigma''(\boldsymbol{k},\omega)$$

Assume momentum-independent self energy within cut, so constant energy slices (MDC) are Lorentzian

# Putting it all together: Cu (111) surface state

Electron binding energy
$|E_B| = E - E_F$




In-plane momentum

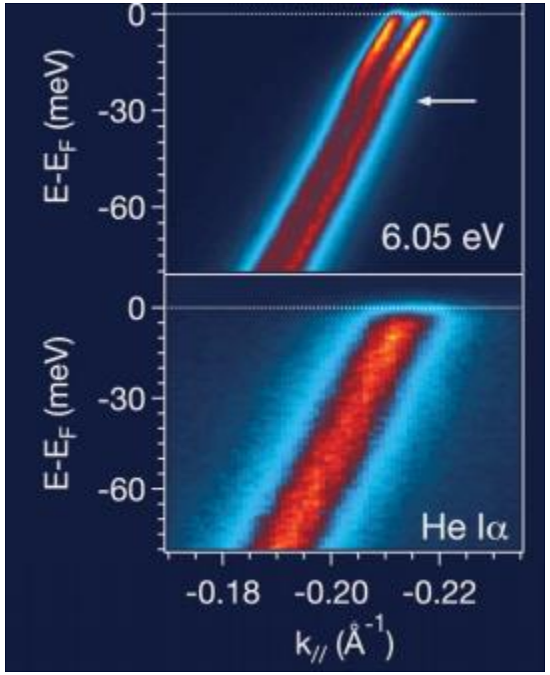
Fermi-Dirac cutoff

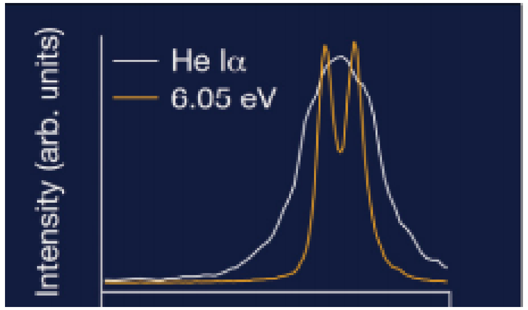
$$F(E) = \frac{1}{e^{(E-\mu)/k_B T} + 1}$$

$$\epsilon_k = E(k) = \frac{\hbar^2 k^2}{2m^*}$$

Tamai et al, PRB **87**, 075113 (2013)

# Interactions in simple system



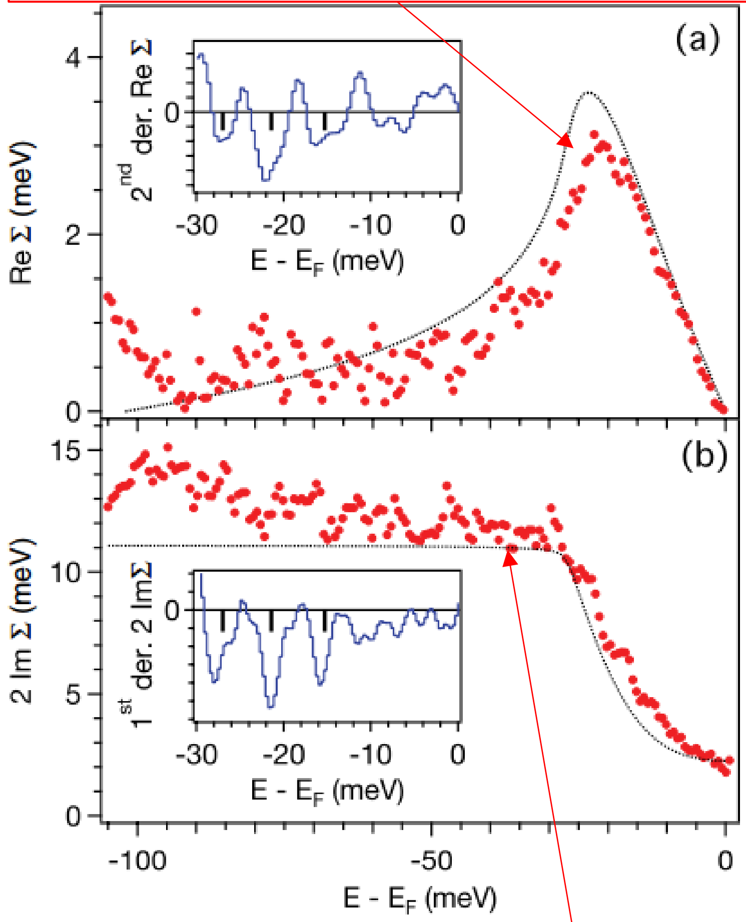




PRB **87**, 075113 (2013)

$$A(\mathbf{k},\omega) = -\frac{1}{\pi}\frac{\Sigma''(\mathbf{k},\omega)}{[\omega-\varepsilon_{\mathbf{k}}-\Sigma'(\mathbf{k},\omega)]^2+[\Sigma''(\mathbf{k},\omega)]^2}$$

$$\Sigma(\mathbf{k},\omega) \rightarrow \Sigma(\omega) = \Sigma'(\omega) + i\Sigma''(\omega)$$

Measured dispersion minus calculated/assumed bare dispersion



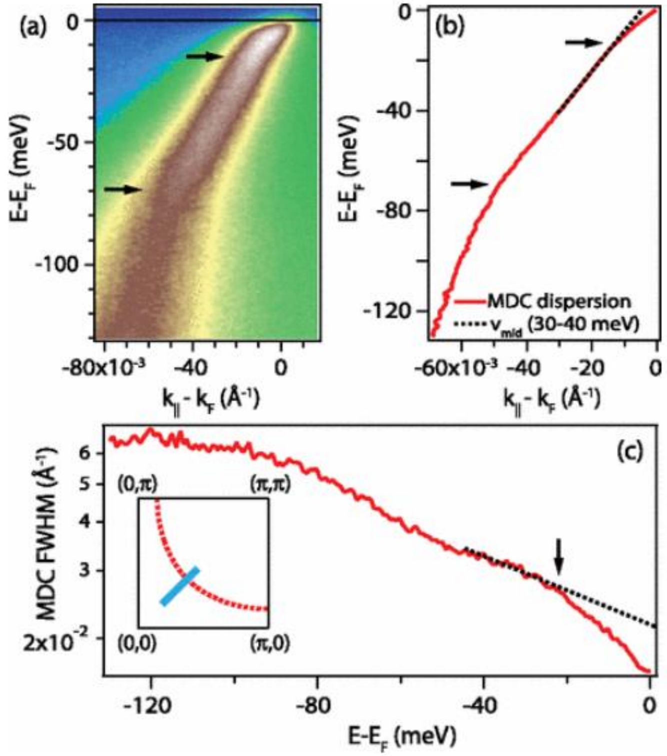

Width of peaks

Electron phonon-coupling!

# Examples: interactions in more complex systems

Kinks in cuprates

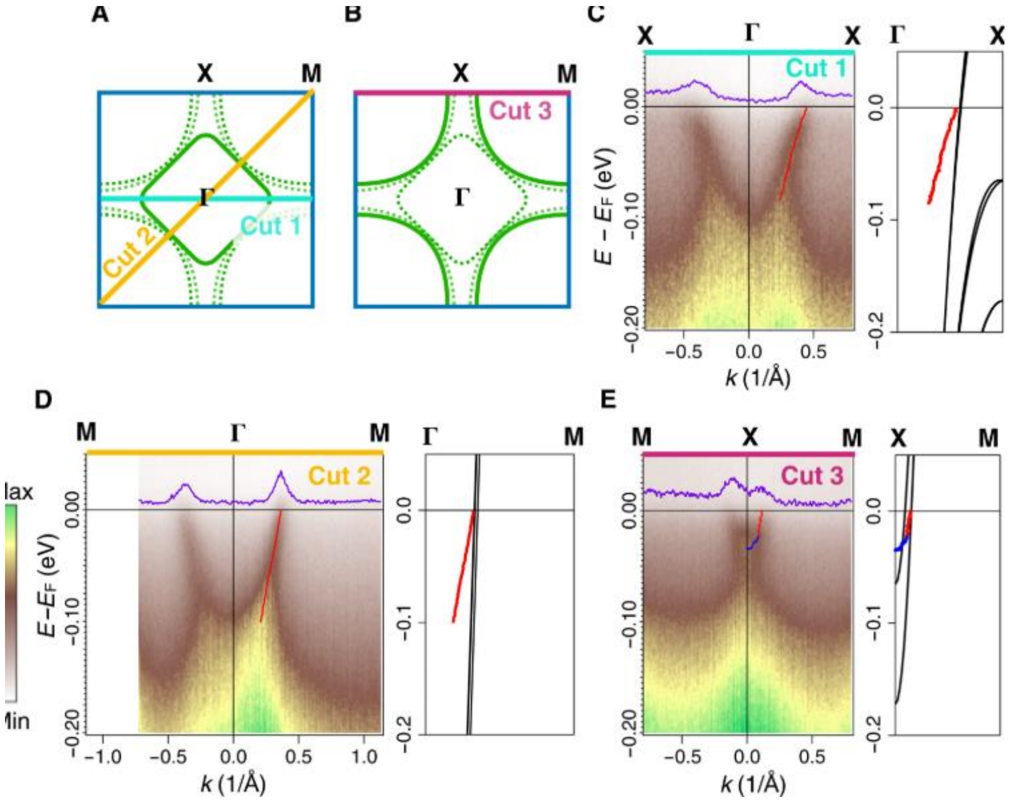
- Establishing origin requires input from other experiments
- Common energy scale in 2 channels attests to veracity of feature




Johnston, Vishik, et al, Phys. Rev. Lett. **108**, 166404 (2012)

Mass enhancement in planar nickelate

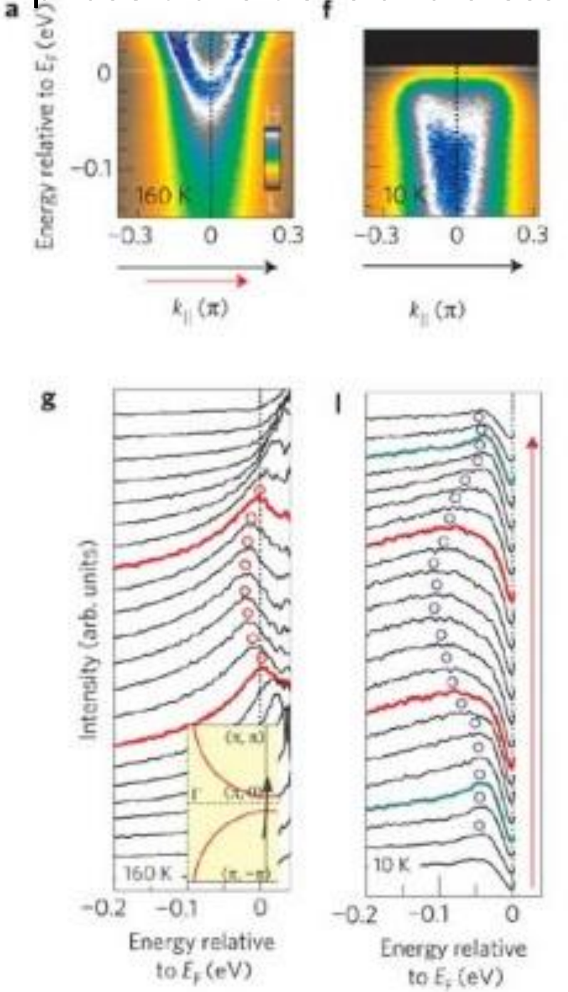
- Measured dispersion with smaller $v_F$ or larger m* is evidence of interactions
- Caution: dispersion can be established more precisely than linewidth




Haoxiang Li *et al.*, *Sci. Adv.* **9**, 4418 (2023)

# What can we accurately say about lineshapes and lifetime?

Sometimes big effects are seen at phase transitions and crossovers




Hashimoto *et al,* Nat. Phys. **6**, 414–418 (2010)

Assessing lifetimes from ARPES linewidths in 3D systems is complicated

$$\Gamma = \frac{\frac{\Gamma_i}{|v_{i\perp}|} + \frac{\Gamma_f}{|v_{f\perp}|}}{\left|\frac{1}{v_{i\perp}}\left[1 - \frac{m v_{i\parallel} \sin^2 \vartheta}{\hbar k_\parallel}\right] - \frac{1}{v_{f\perp}}\left[1 - \frac{m v_{f\parallel} \sin^2 \vartheta}{\hbar k_\parallel}\right]\right|}$$

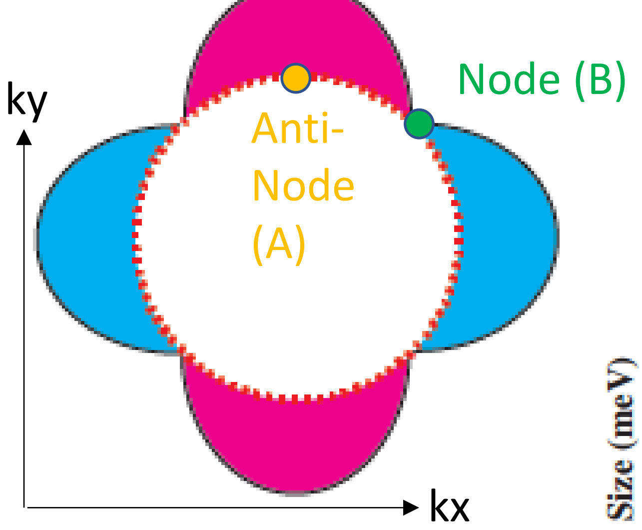
2D non-interacting systems are safer

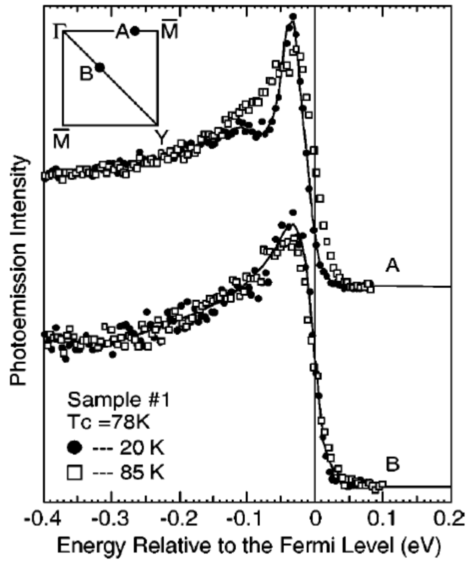
$$\Gamma = \frac{\Gamma_i}{\left|1 - \frac{m v_{i\parallel} \sin^2 \vartheta}{\hbar k_\parallel}\right|} \equiv C\,\Gamma_i$$

Damascelli, Physica Scripta. Vol. T109, 61–74 (2004)

Caution: extrinsic effects usually contribute substantially to linewidth

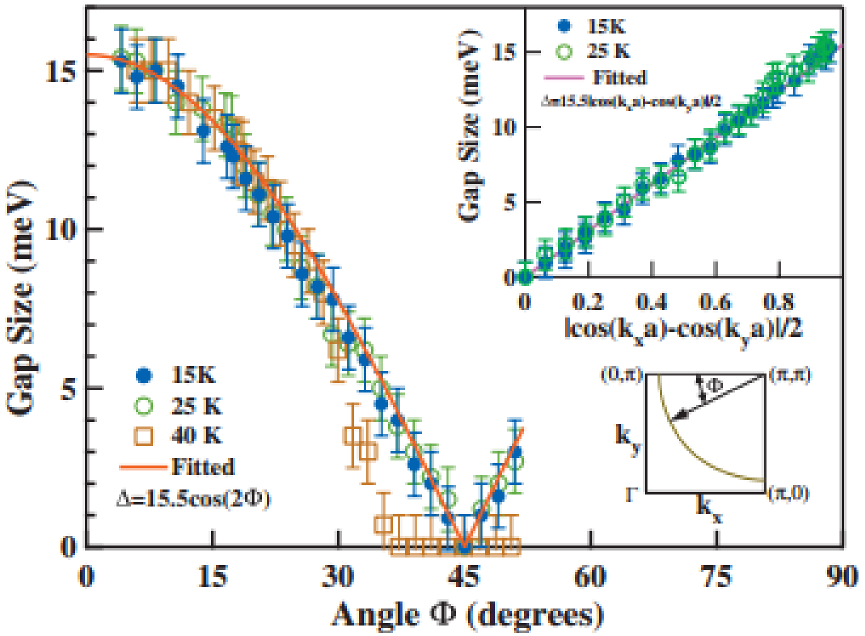
# Quantifying spectral gaps with ARPES



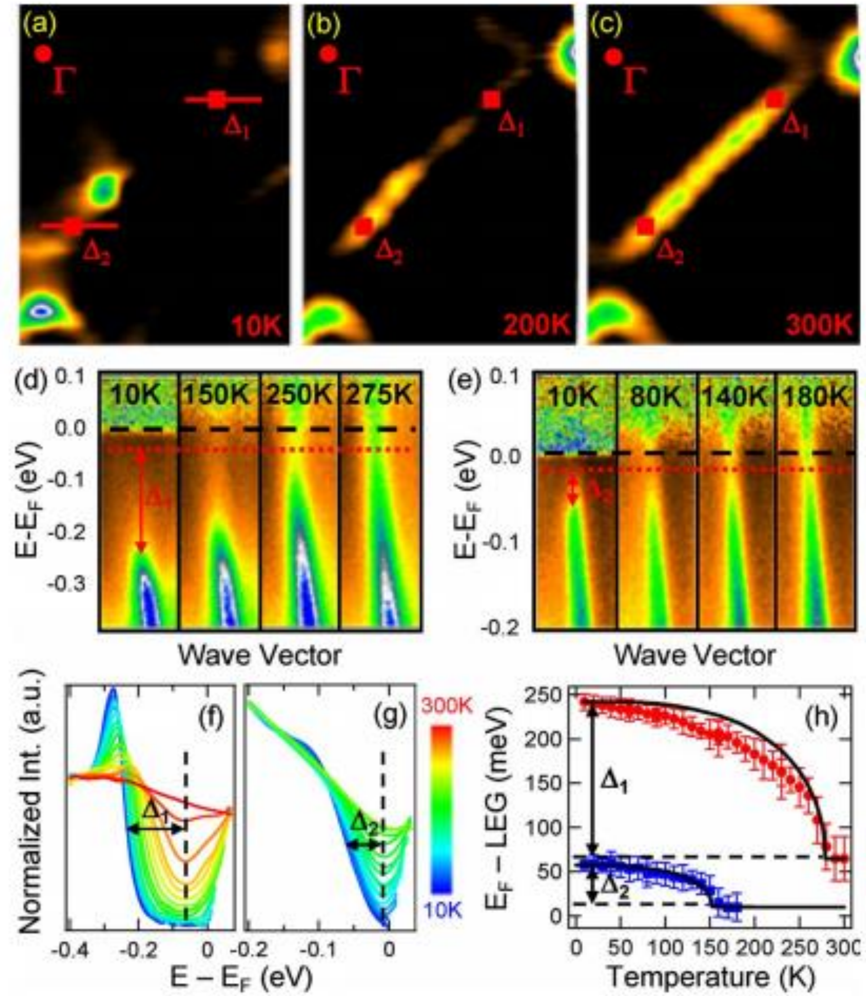

Shen *et al.* PRL **70** (1993)

Superconducting gaps in cuprates




Meng *et al*, PRB **79**, 024514 (2009)

CDW gaps in $ErTe_3$



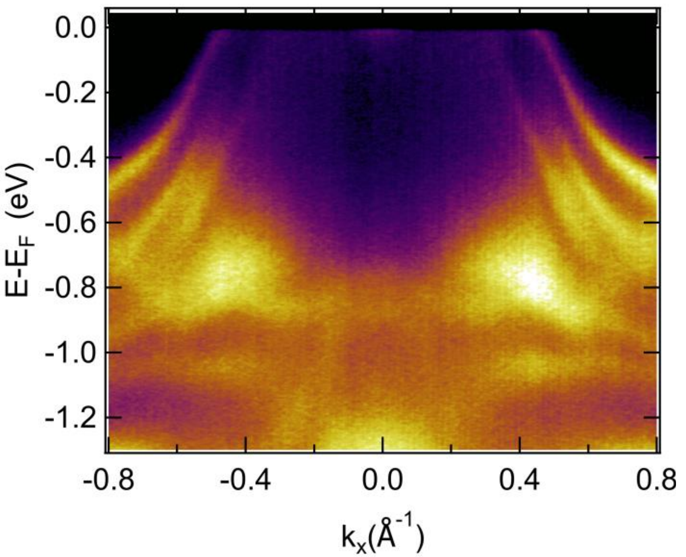

Moore *et al.* PRB **81**, 073102 (2010)

# Twists on the ARPES technique

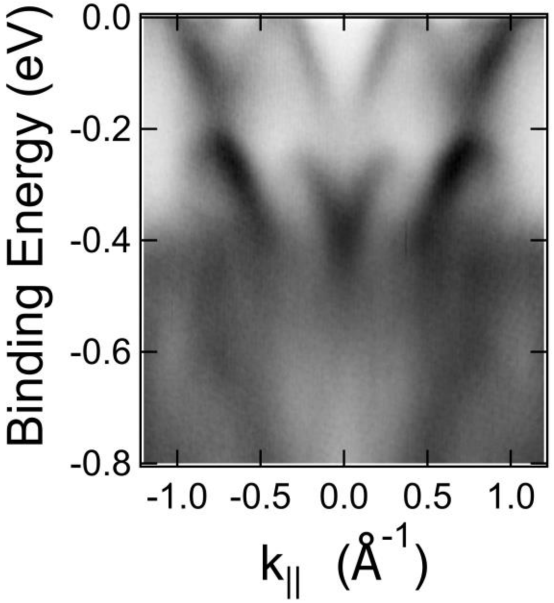
- Time-resolved ARPES
  - Pump-probe or two-photon photoemission
  - Tradeoff with resolution, limiting factor is lightsources
- Spin-resolved ARPES
  - Tradeoff with throughput and resolution
- Circular dichroism ARPES (poor man's spin-resolved)
  - Easier/faster experiment than spin-resolved, harder to interpret
- ARPES while perturbing sample (gate, current, small magnetic field)
  - Rare, challenging
- Micro and nanoARPES
  - No tradeoffs

# ARPES is an information rich technique

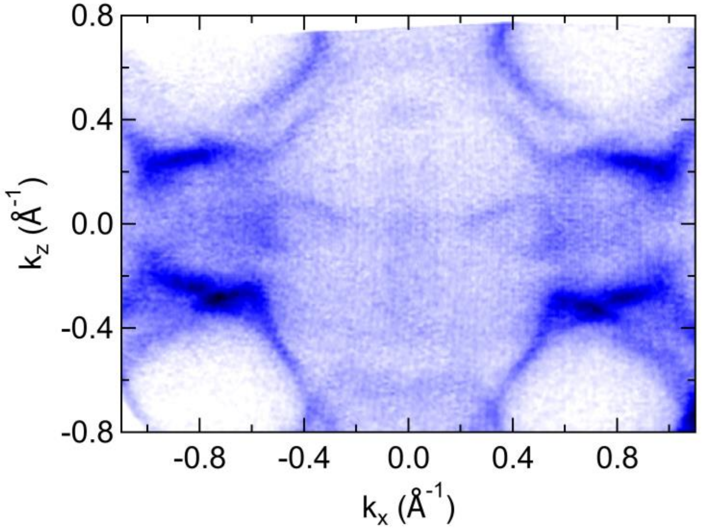
**Volcano**



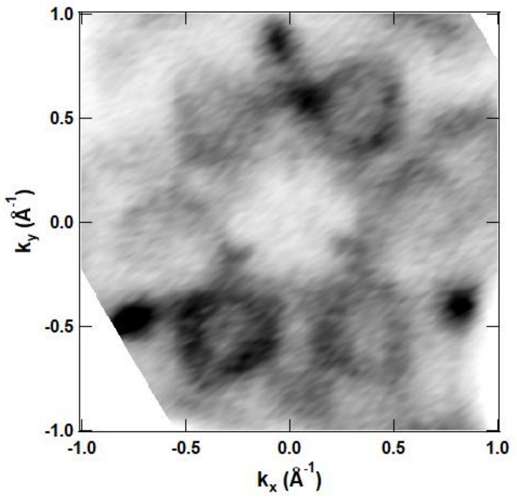

**Bat**




**Person**




**Flower**



# Resources


- Campuzano, Norman, Randeria. *Photoemission in the high-Tc superconductors.* https://arxiv.org/pdf/cond-mat/0209476.pdf
- Damascelli, Hussain, Shen. *Angle-resolved photoemission studies of the cuprate superconductors.* Rev. Mod. Phys. **75** 473 (2003)
- Damascelli. *Probing the Electronic Structure of Complex Systems by ARPES.* Physica Scripta. Vol. T109, 61–74, 2004
- Moser, 'An experimentalist's guide to the matrix element in angle resolved photoemission' Journal of Electron Spectroscopy and Related Phenomena **214** 29–52 (2017)
- Sobota, He, Shen. *Angle-resolved photoemission studies of quantum materials.* Rev. Mod. Phys. **93**, 025006 (2021)
- V.N. Strocov Journal of Electron Spectroscopy and Related Phenomena 130 65–78 (2003)
- E. Rotenberg and A. Bostwick, "microarpes and nanoarpes at diffraction-limited light sources: opportunities and performance gains," Journal of Synchrotron Radiation, vol. 21, no. 5, pp. 1048–1056 (2014)

# Extra slides

# ARPES (CW) light sources: compare/contrast

| Type | Available photon energies | Bandwidth/monochromaticity | Intensity | Polarization | Versatility |
|---|---|---|---|---|---|
| **Laser** | 6-11 eV; not much variation for a given laser; for time-resolved experiments: HHG sources (meh) | Can be <<1 meV | Potentially high | Usually Variable polarization | Low; does not work on every material; usual fixed photon energy gives incomplete info about 3D materials |
| **Gas (He, Xe, Ne, Ar…) discharge lamp** | 21.2, 40.8, 8.4, 9.6, 11.6 eV (and more) | Can be small (<1 meV) with monochromator | Moderate | unpolarized | Low-moderate; fixed photon energy gives incomplete info about 3D materials |
| **Synchrotron** | Variable; different synchrotrons and endstations specialize in different energy ranges | Can be <1meV; tradeoff between bandwidth/ intensity | Often high, sometimes tradeoffs with bandwidth | Usually variable | High; tunable photon energy allows to optimize matrix elements and study 3D materials |

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

$$\mathbf{p}_{\parallel} = \hbar \mathbf{k}_{\parallel} = \sqrt{2mE_{kin}} \cdot \sin\vartheta$$

$$M_{f,i}^{\boldsymbol{k}} \equiv < \phi_f^{\boldsymbol{k}} | -\frac{e}{mc} \boldsymbol{A} \cdot \boldsymbol{p} | \phi_i^{\boldsymbol{k}} >$$

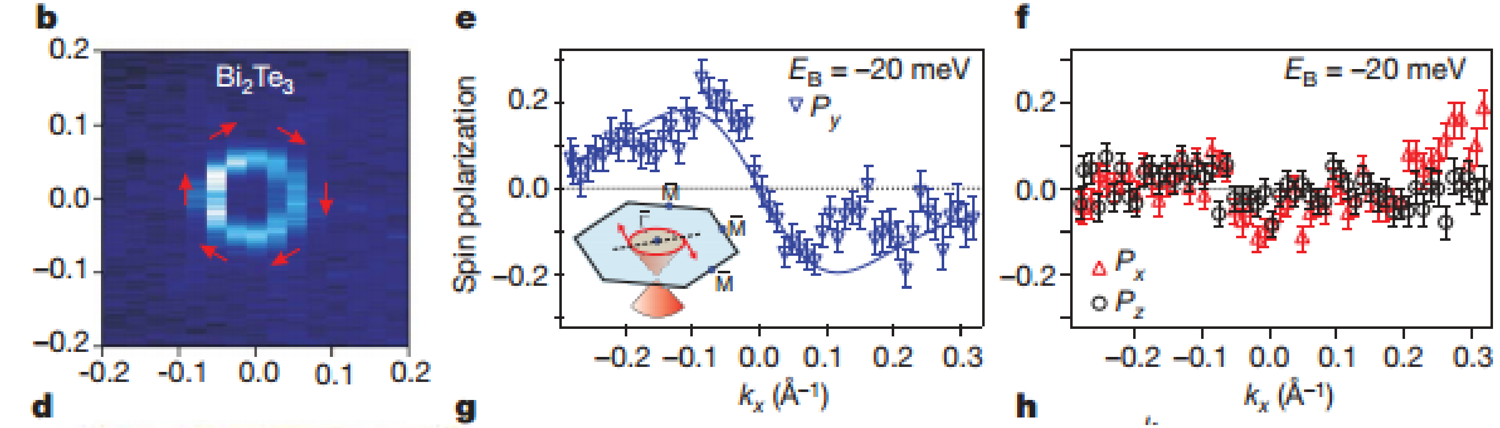
# Spin resolved ARPES

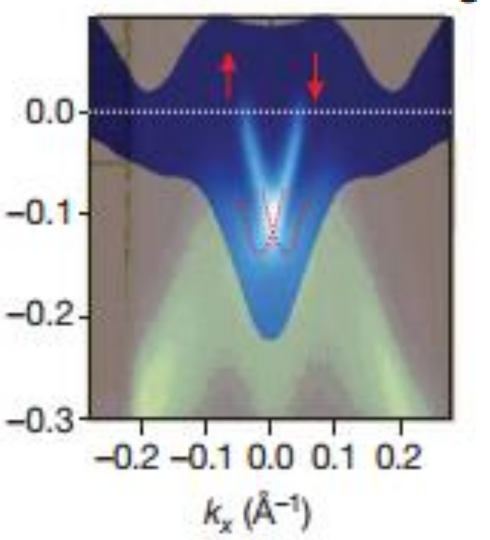
How can we measure electron spin in photoemission experiments?

| Method | Interaction | Operation voltage | $S_{eff}$ | Figure of merit | Target |
|---|---|---|---|---|---|
| Mott | Spin-orbit | 20–100 kV | 0.1–0.2 | $1–5 \times 10^{-4}$ | Au thin film |
| SPLEED | Spin-orbit | 150 V | 0.2–0.3 | $1–2 \times 10^{-4}$ | W single crystal |
| Diffuse scattering | Spin-orbit | 150 V | ~0.2 | $\sim 1 \times 10^{-4}$ | Au thin film |
| VLEED | Spin-exchange | 6–10 V | 0.3–0.4 | $\sim 10^{-2}$ | Fe single crystal |

A. Takayama, *High-resolution spin-resolved photoemission spectrometer and the Rashba effect in Bismuth thin films (2015)*

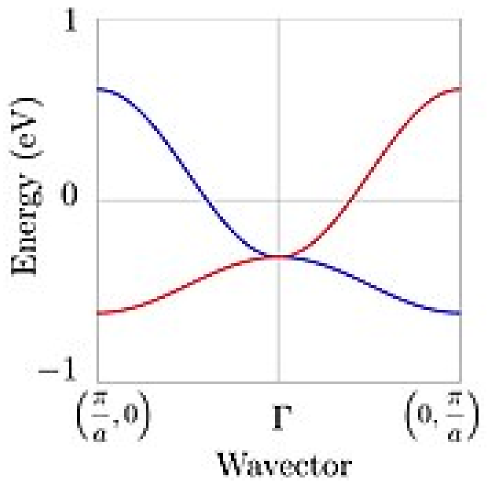
# Spin-momentum locking in topological insulators



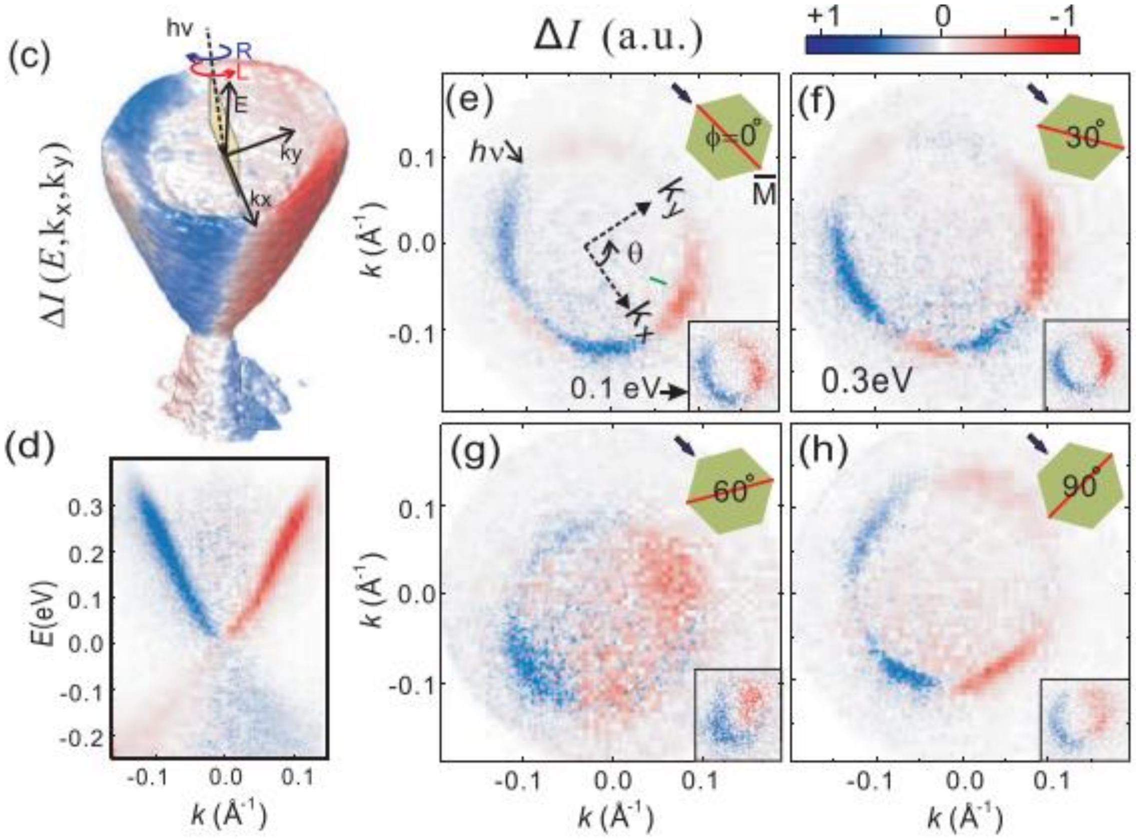

Hsieh *et al.* Nature **460** (2009)

Modern application: Altermagnets



# Spin texture via circular dichroism

1. Measure ARPES spectrum with left-circularly polarized (LCP) light
2. Measure ARPES spectrum with right-circularly polarized (RCP) light
3. $\Delta I(E, k_x, k_y) = I_{LCP} - I_{RCP}$

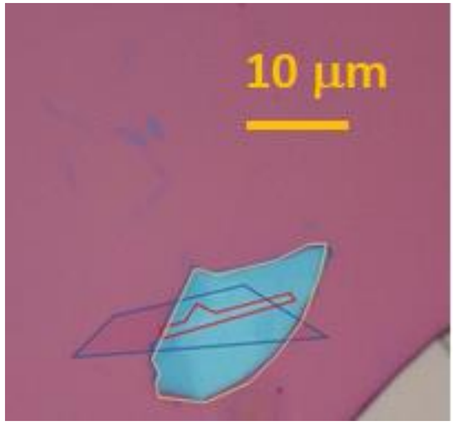



- Benefits:
  - Possible with most lightsources
  - High throughput/resolution
- Challenges:
  - Spin+orbital
  - Final state effects

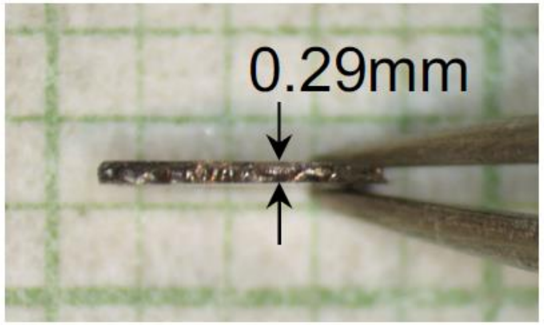
Wang *et al.* PRL **107**, 207602 (2011)

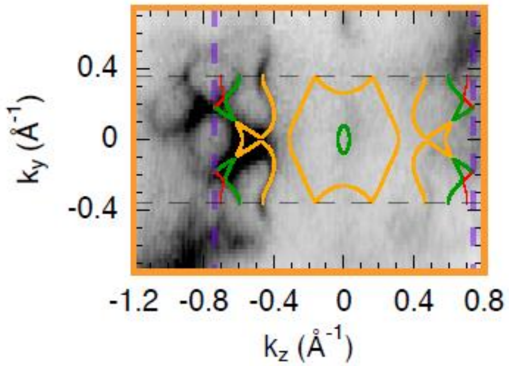
# micro/nanoARPES have enabled more materials to be ARPESed

Samples with small or irregular cleaves

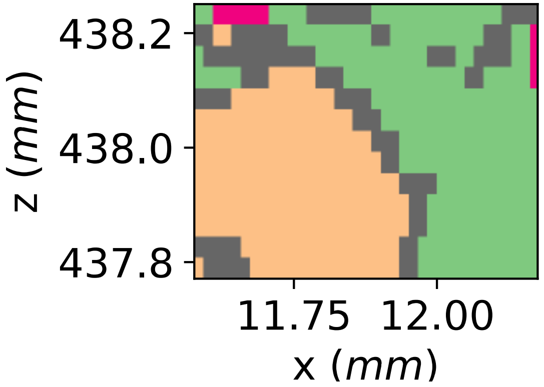



Exfoliated materials and heterostructures




Staab *et al,* Physical Review B 110 (16), 165115 (2024)




Samples with mesoscale chemical or electronic inhomogeneity
A. Rossi et al, Phys. Rev. B 104, 155115 (2021)
S. Sreedhar et al, *in preparation*

# Relationship between ARPES and single particle spectral function

- Photoemission removes an electron and inverse photoemission adds an electron
- Electron removal/addition described by one-electron addition and removal Green's function:

$$G^{\pm}(\boldsymbol{k},\omega) = \sum_m \frac{|\langle \Psi_m^{N\pm1} | c_{\boldsymbol{k}}^{\pm} | \Psi_i^N \rangle|^2}{\omega - E_m^{N\pm1} + E_i^N \pm i\eta}$$

  $c_{\boldsymbol{k}}^{\pm}$ creates/annihilates electron with energy $\omega$ and momentum **k**

  $\eta$ is positive infinitesimal

- Retarded Green's function is related to one-electron spectral function via:

$$G(\boldsymbol{k},\omega) = \int_{-\infty}^{\infty} d\omega' \frac{A(\boldsymbol{k},\omega')}{\omega - \omega' \pm i\eta}$$

- $(x \pm i\eta)^{-1} = \mathcal{P}\left(\frac{1}{x}\right) \mp i\pi\delta(x), \eta \to 0^+$
- $-\left(\frac{1}{\pi}\right) Im\, G(\boldsymbol{k},\omega) = A^+(\boldsymbol{k},\omega) + A^-(\boldsymbol{k},\omega)$ where $\mathrm{G}(\boldsymbol{k},\omega)$ is the retarded Green's function given by $G(\boldsymbol{k},\omega) = G^+(k,\omega) + [G^-(k,\omega)]^*$

# Single particle spectral function (continued)

- $A^{\pm}(\boldsymbol{k},\omega) = \sum_m \left|\left\langle \Psi_m^{N\pm 1}\middle| c_{\boldsymbol{k}}^{\pm} \middle| \Psi_i^N \right\rangle\right|^2 \delta(\omega - E_m^{N\pm 1} + E_i^N)$
- Corrections to Green's function originating from interactions:

$$G(\boldsymbol{k},\omega) = \frac{1}{\omega - \epsilon_{\boldsymbol{k}} - \Sigma(\boldsymbol{k},\omega)}$$

  where $\epsilon_{\boldsymbol{k}}$ is bare band dispersion, and $\Sigma(\boldsymbol{k},\omega) = \Sigma'(\boldsymbol{k},\omega) + i\,\Sigma''(\boldsymbol{k},\omega)$ is the self-energy
- This allows to write the single-particle spectral function in terms of self energies as well:

$$A(k,\omega) = -\frac{1}{\pi}\frac{\Sigma''(\boldsymbol{k},\omega)}{[\omega - \epsilon_{\boldsymbol{k}} - \Sigma'(\boldsymbol{k},\omega)]^2 + [\Sigma''(\boldsymbol{k},\omega)]^2}$$

# Pump-probe experiments

**The pump**

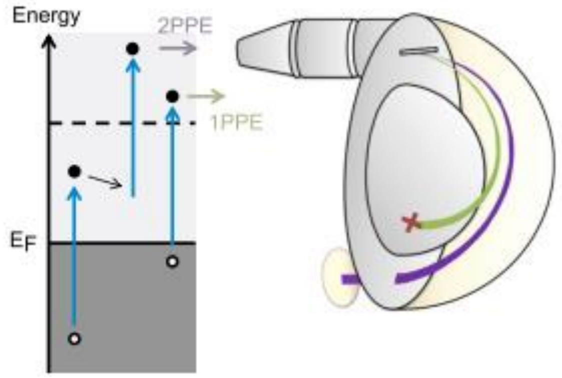
- Purpose (depends on specific experiment)
  - Create specific excitation
  - Whack the electronic system on a timescale faster than lattice response
  - Cause destruction
- Frequency (depends on specific experiment)
  - 1.5 eV (straight out of the Ti-Sapph laser)
  - Mid-IR (70-500 meV—relevant to excitations in solids)

**The probe**

- Ascertains system's response as a function of time delay from pump
- Defines what experiment you are doing
  - Optics (probe measures change in reflectivity or absorption)
  - THz (measures changes in optical conductivity at low frequencies)
  - ARPES (measures changes in band structure)
  - Many others

# 2 photon photoemission (2PPE) as a substitute for inverse photoemission

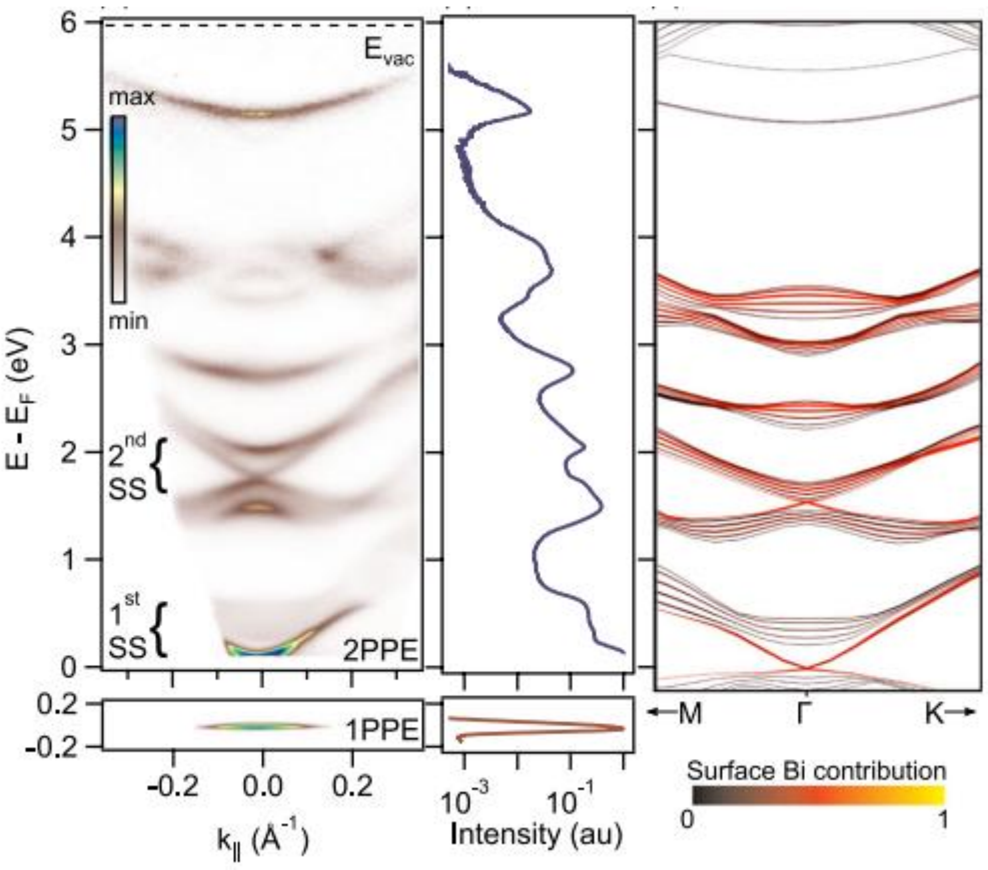
| | | | |
|---|---|---|---|
| **Photoemission** | Photon in, electron out | Measure **occupied** electronic states | Sub-meV resolution common |
| **Inverse photoemission** | Electron in, photon out | Measure **unoccupied** electronic states | ~500 meV resolution |




Use time-resolved ARPES to measure unoccupied states

- Pulse 1 (pump): make excitation into unoccupied state
- Pulse 2 (probe): perform photoemission out of pump-populated unoccupied state
- Time resolution is not very important, but light **intensity** is because this is 2nd order process

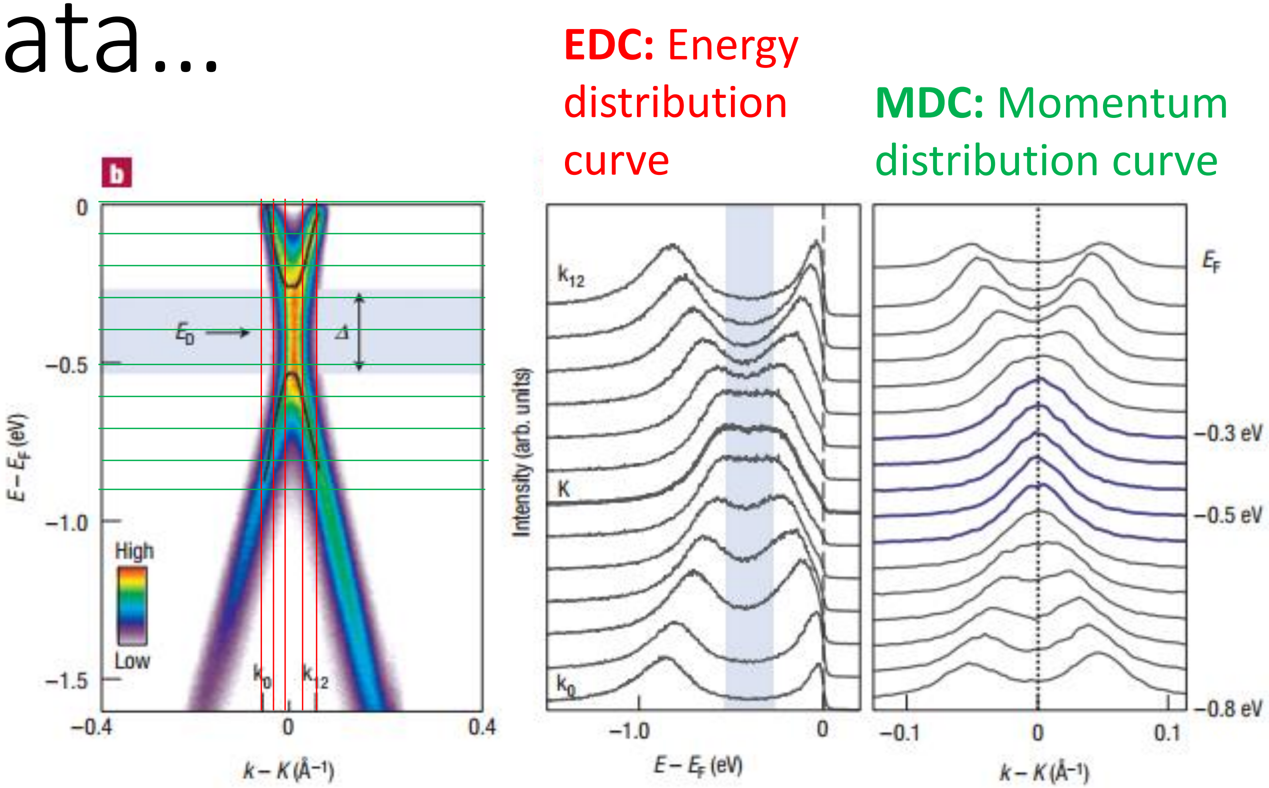
# 2 PPE experiments in $Bi_2Se_3$




- 1.5 eV pump, 5.98 eV probe, $\Delta t \sim 100 fs$
- 2$^{nd}$ surface state observed above $E_F$!
- Applicable to many different materials

Sobota *et al*. PRL **111**, 136802 (2013)

# Looking at data…




Zhou *et al* Nat. Mater **6** 770 (2007)

Main result: substrate (SiC) breaks sublattice symmetry of graphene, opening a gap at the Dirac point

Which analysis (EDC or MDC) illustrates this result better?