Detecting Bands and Fermi Surfaces by State-of-the-Art ARPES

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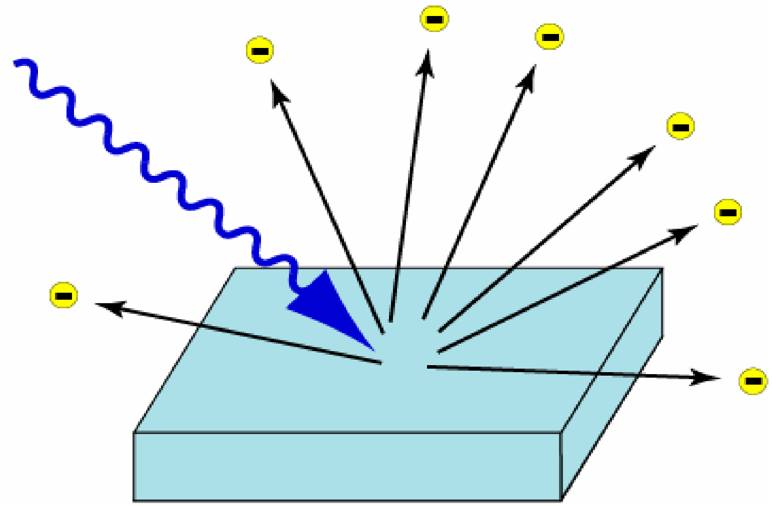
Outline: Part I

State-of-the-Art ARPES: the essentials

- ▶ **Motivation and potential**
- ▶ **Formal description**
  - One-step vs three-step model
  - The sudden approximation
  - Kinematics of photoemission
  - One-particle spectral function
- ▶ **Experimental**
  - State-of-art ARPES
  - Surface vs bulk sensitivity
- ▶ **Summary and discussion**

History of Photoemission

The Photoelectric Effect



- First experimental work performed by H. Hertz (1886), W. Hallwachs (1888), von Lenard (1900)

- Theoretical explanation by Einstein (1905)

FIRST EXPERIMENTAL EVIDENCE FOR QUANTIZATION OF LIGHT!

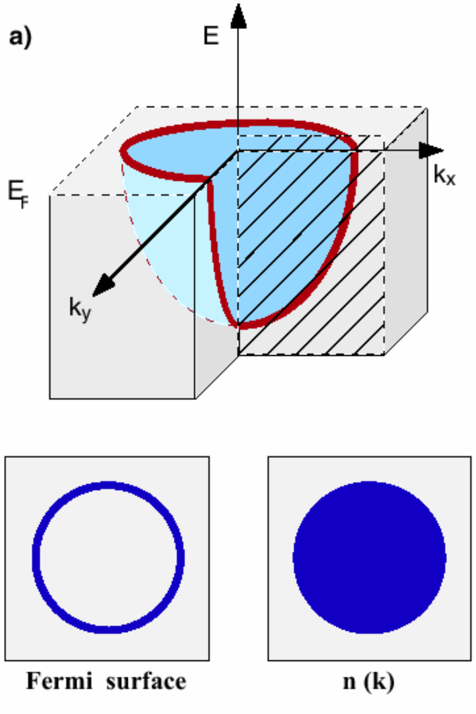
Is there anything else we can learn from the photoelectric effect?

Insights into the solid-state!

Courtesy of Kyle Shen

Understanding the Solid State: Electrons in Reciprocal Space

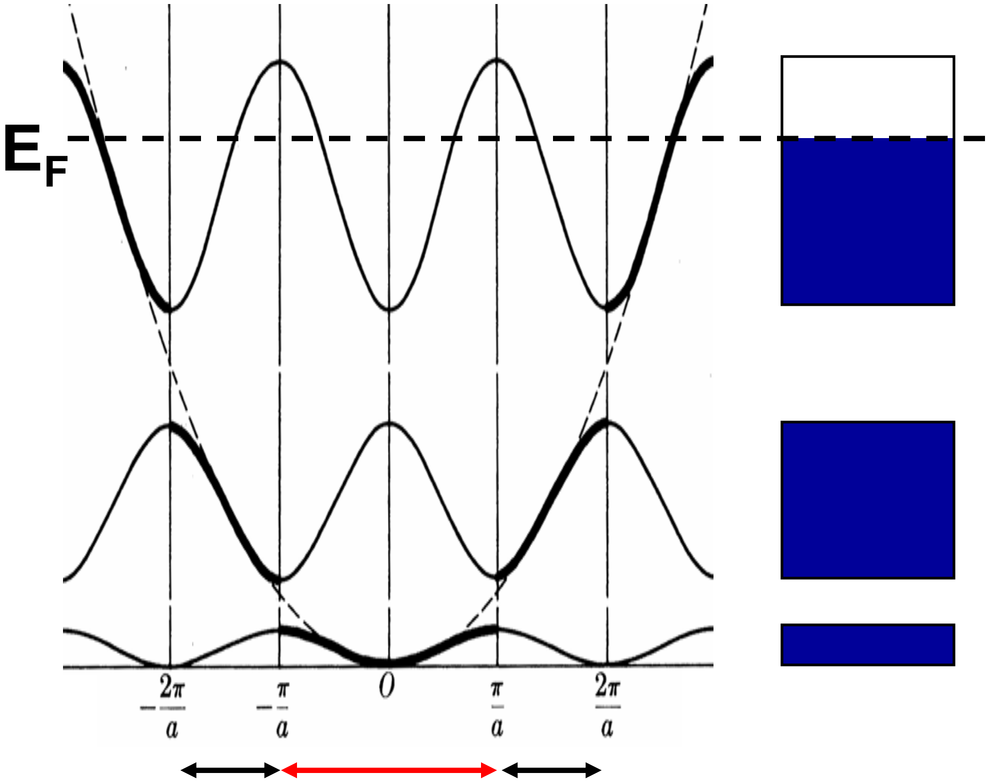
Many properties of a solids are determined by electrons near E_F (conductivity, magnetoresistance, superconductivity, magnetism)



Only a narrow energy slice around E_F is relevant for these properties (kT=25 meV at room temperature)

Allowed electronic states

Repeated-zone scheme



Second Brillouin zone

First Brillouin zone

Second Brillouin zone

Interaction Effects between Electrons : “Many-body Physics”

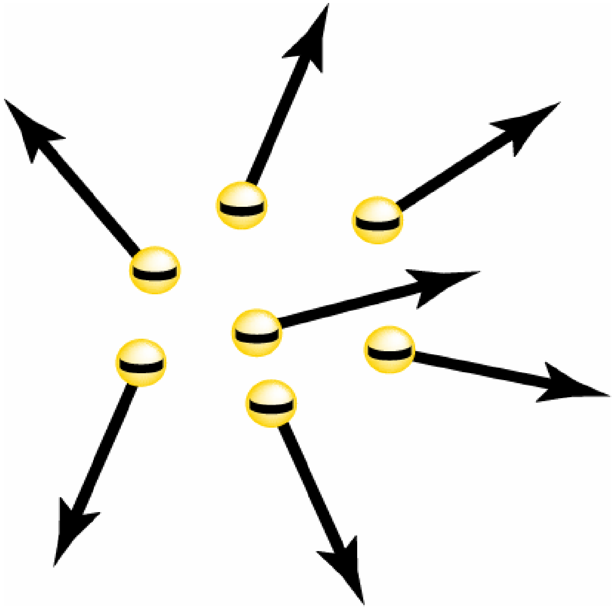
Many-body effects are due to the interactions between the electrons and each other, or with other excitations inside the crystal :

1) A “many-body” problem : intrinsically hard to calculate and understand

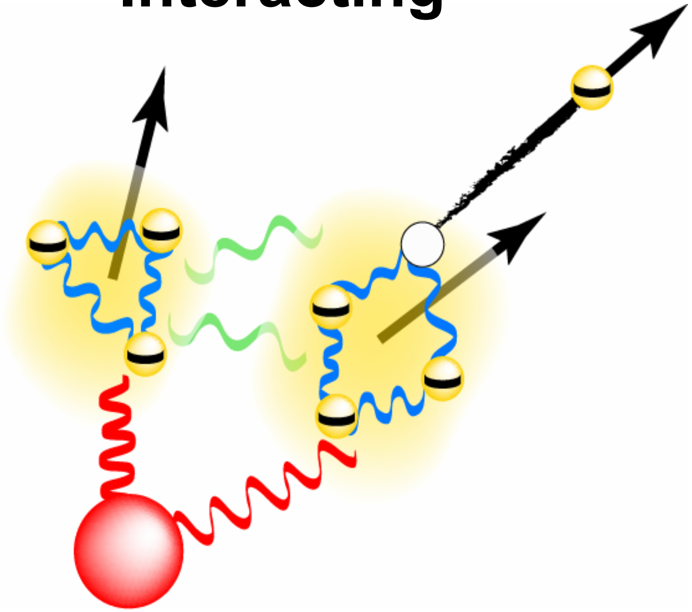
2) Responsible for many surprising phenomena :

Superconductivity, Magnetism, Density Waves, ...

Non-Interacting

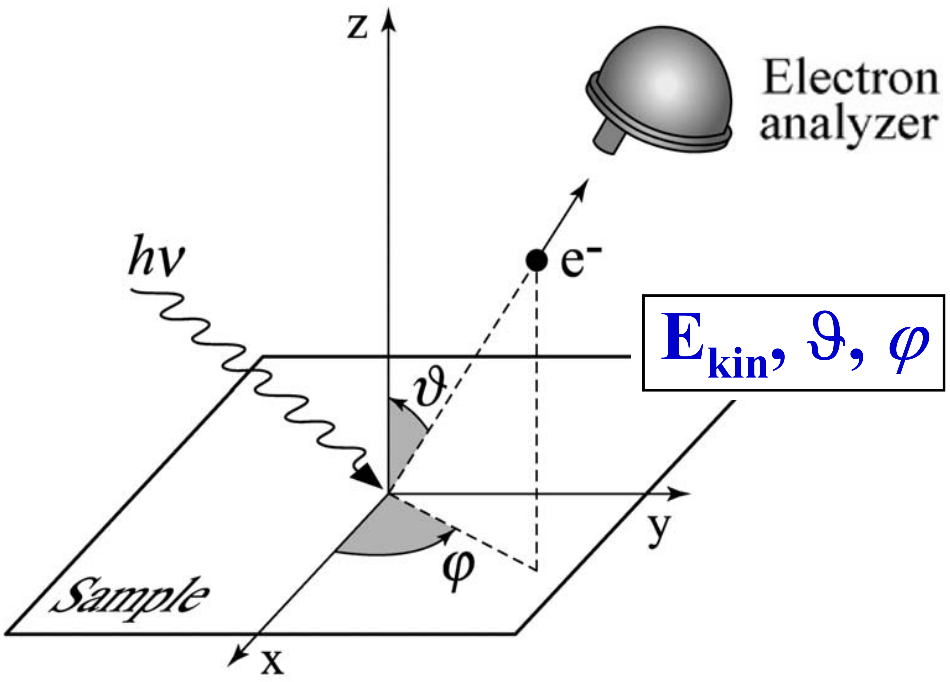


Interacting



Courtesy of Kyle Shen

Angle-Resolved Photoemission Spectroscopy



E_{kin}, ϑ, φ →

$$\mathbf{K} = \mathbf{p}/\hbar = \sqrt{2mE_{kin}}/\hbar$$

$$K_x = \frac{1}{\hbar}\sqrt{2mE_{kin}}\sin\vartheta\cos\varphi$$

$$K_y = \frac{1}{\hbar}\sqrt{2mE_{kin}}\sin\vartheta\sin\varphi$$

$$K_z = \frac{1}{\hbar}\sqrt{2mE_{kin}}\cos\vartheta$$

Vacuum

E_{kin}

$\mathbf{K}$

Conservation laws

$$E_f - E_i = h\nu$$

$$\mathbf{k}_f - \mathbf{k}_i = \cancel{\mathbf{k}_{h\nu}}$$

Solid

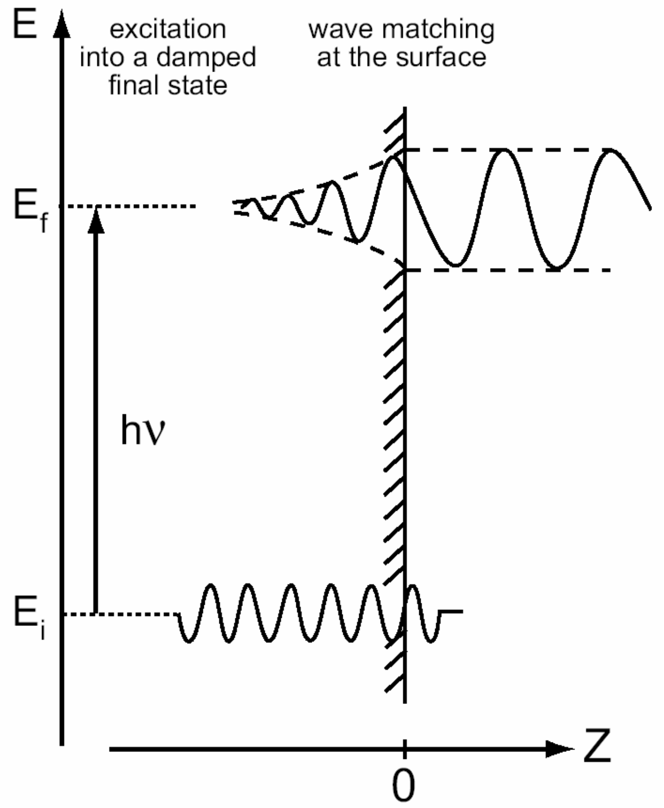
E_B

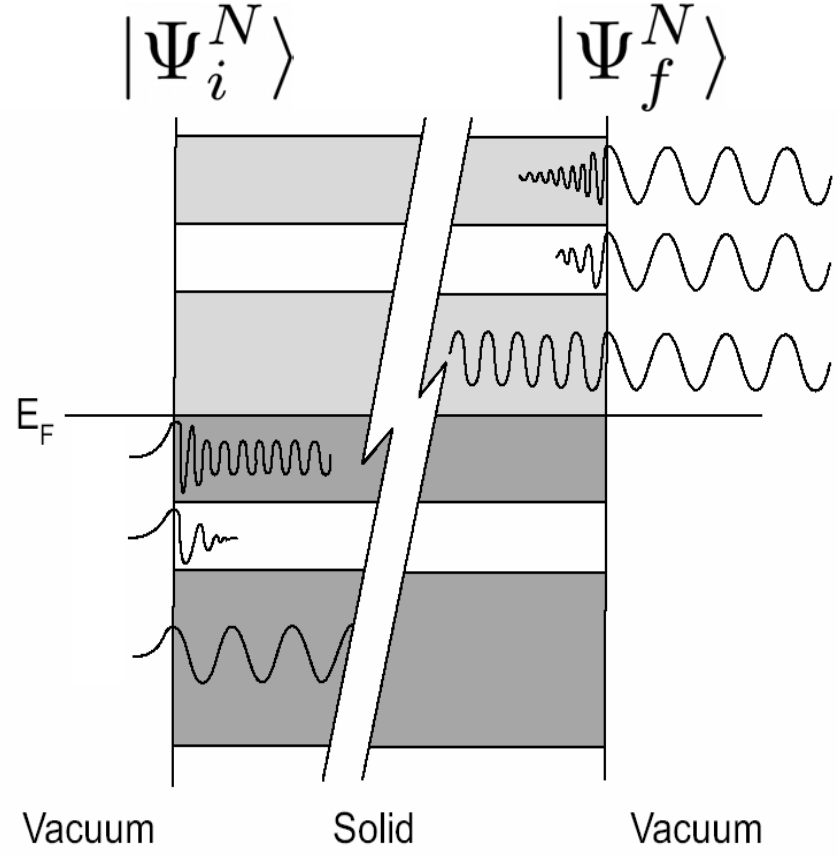
$\mathbf{k}$

ARPES: One-Step vs Three-Step Model

Photoemission Intensity $I(k,\omega)$ $\Big\}$ $$w_{fi} \propto |\langle\Psi_f^N|\mathbf{A}\cdot\mathbf{p}|\Psi_i^N\rangle|^2\delta(E_f^N - E_i^N - h\nu)$$

One-step model



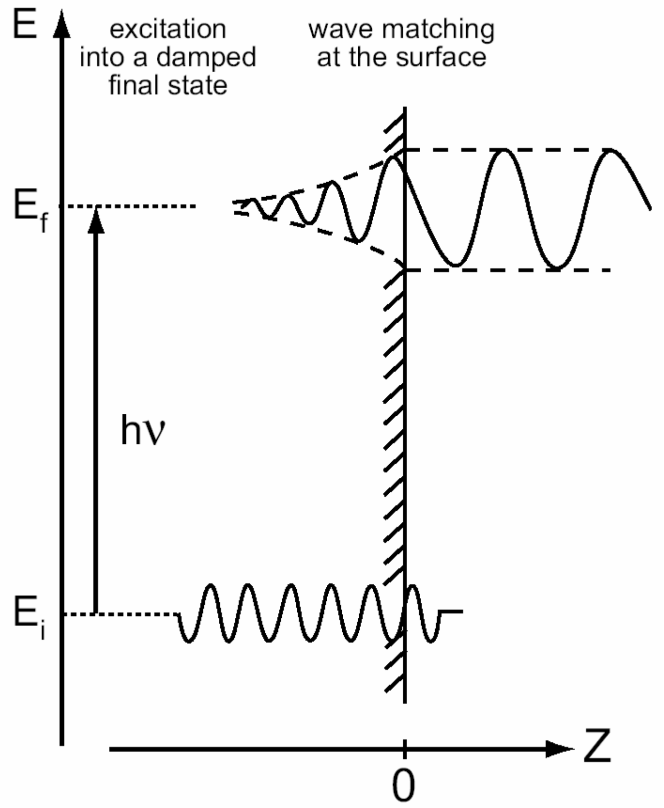


ARPES: One-Step vs Three-Step Model

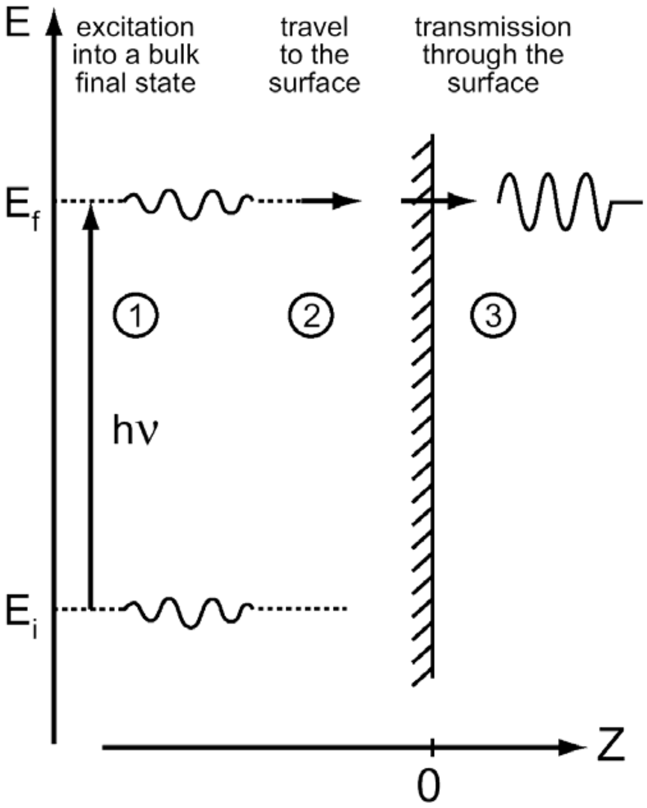
Photoemission Intensity *I(k,ω)*

$$\left.\begin{array}{l}\end{array}\right\} w_{fi} \propto |\langle \Psi_f^N | \mathbf{A}\cdot\mathbf{p} | \Psi_i^N \rangle|^2 \delta(E_f^N - E_i^N - h\nu)$$

One-step model



Three-step model

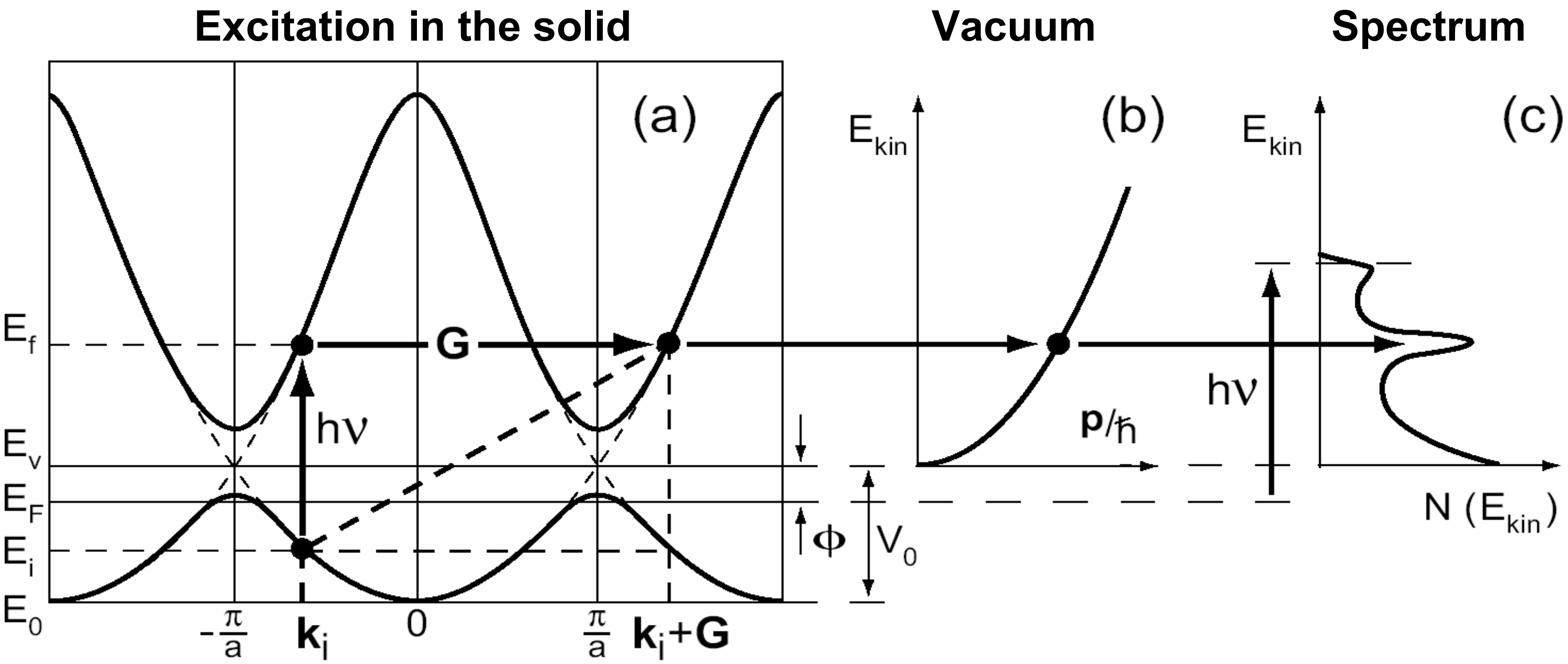


ARPES: The Sudden Approximation

$$\left.\begin{array}{l}\textbf{Photoemission}\\ \textbf{Intensity}\;\; I(k,\omega)\end{array}\right\}\quad w_{fi} \propto |\langle \phi_f^{\mathbf{k}}|\mathbf{A}\cdot\mathbf{p}|\phi_i^{\mathbf{k}}\rangle\langle\Psi_m^{N-1}|\Psi_i^{N-1}\rangle|^2\delta(\omega-h\nu)$$

Ψ_f^N: **Sudden approximation** → $\Psi_f^N = \mathcal{A}\,\phi_f^{\mathbf{k}}\,\Psi_f^{N-1}$

Ψ_i^N: **One Slater determinant** → $\Psi_i^N = \mathcal{A}\,\phi_i^{\mathbf{k}}\,\Psi_i^{N-1}$

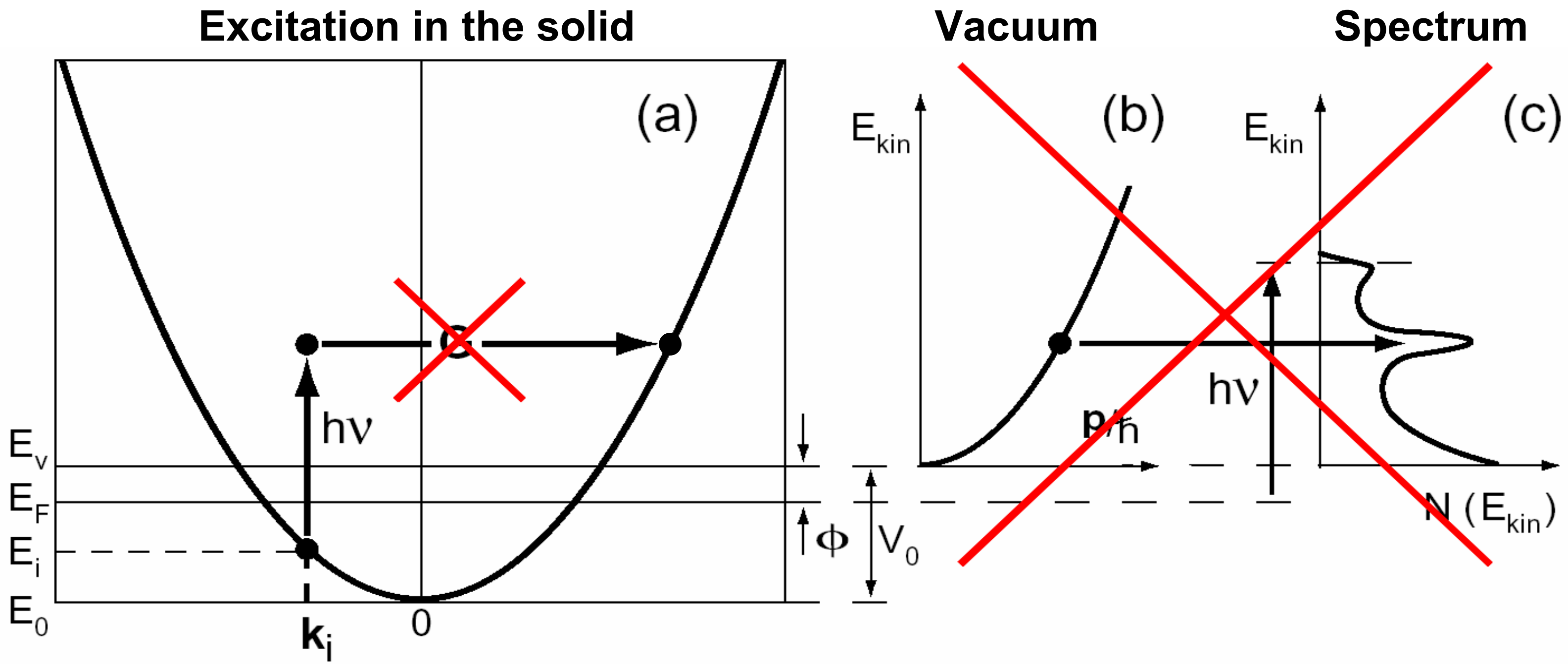


ARPES: Role of the Crystal Potential

$$\left.\begin{array}{l}\textbf{Photoemission} \\ \textbf{Intensity}\;\; I(k,\omega)\end{array}\right\} \; w_{fi} \propto |\langle \phi_f^{\mathbf{k}} | \underline{\mathbf{A}\cdot\nabla V} | \phi_i^{\mathbf{k}} \rangle \langle \Psi_m^{N-1} | \Psi_i^{N-1} \rangle|^2 \delta(\omega - h\nu)$$

Ψ_f^N: **Sudden approximation** → $\Psi_f^N = \mathcal{A}\,\phi_f^{\mathbf{k}}\,\Psi_f^{N-1}$

Ψ_i^N: **One Slater determinant** → $\Psi_i^N = \mathcal{A}\,\phi_i^{\mathbf{k}}\,\Psi_i^{N-1}$



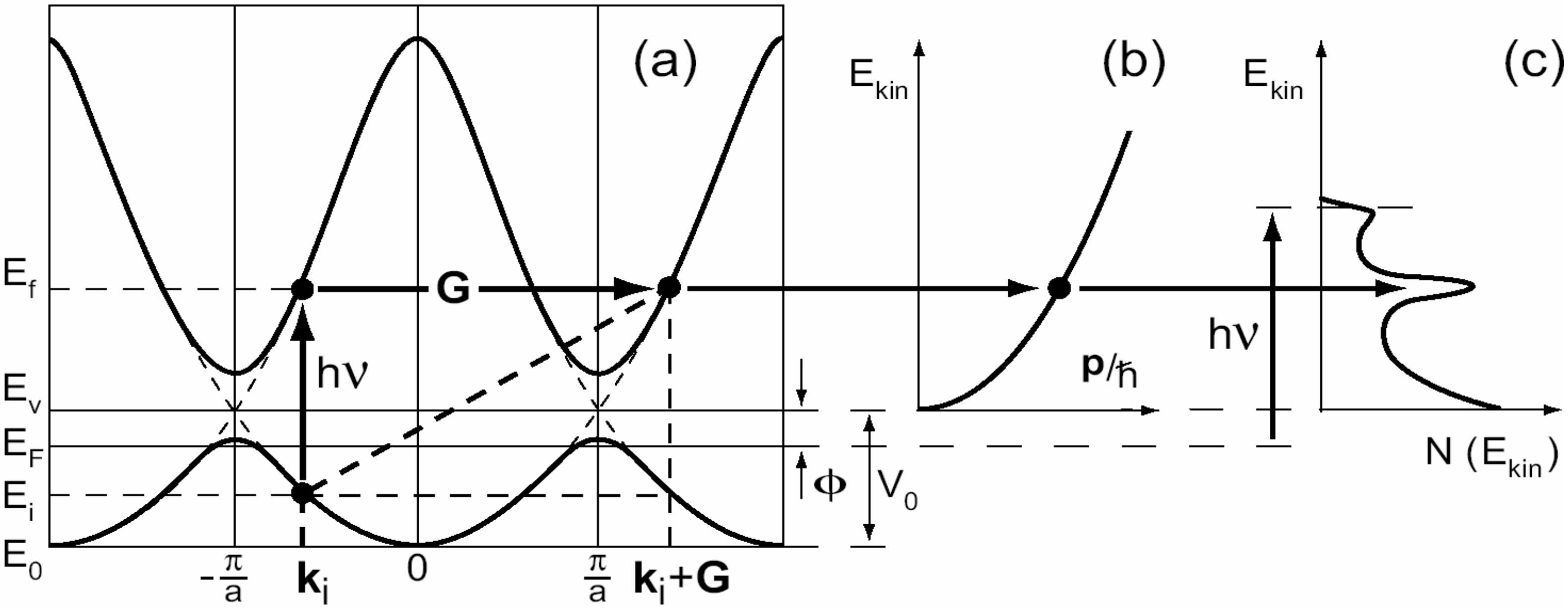
ARPES: Inner Potential and Determination of k_z

Free-electron final state $$E_f(\mathbf{k}) = \frac{\hbar^2\mathbf{k}^2}{2m} - |E_0| = \frac{\hbar^2(\mathbf{k}_{\|}{}^2 + \mathbf{k}_{\perp}{}^2)}{2m} - |E_0|$$

because $\hbar^2\mathbf{k}_{\|}^2/2m = E_{kin}\sin^2\vartheta$ $E_f = E_{kin}+\phi$ $V_0 = |E_0|+\phi$

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

Excitation in the solid **Vacuum** **Spectrum**

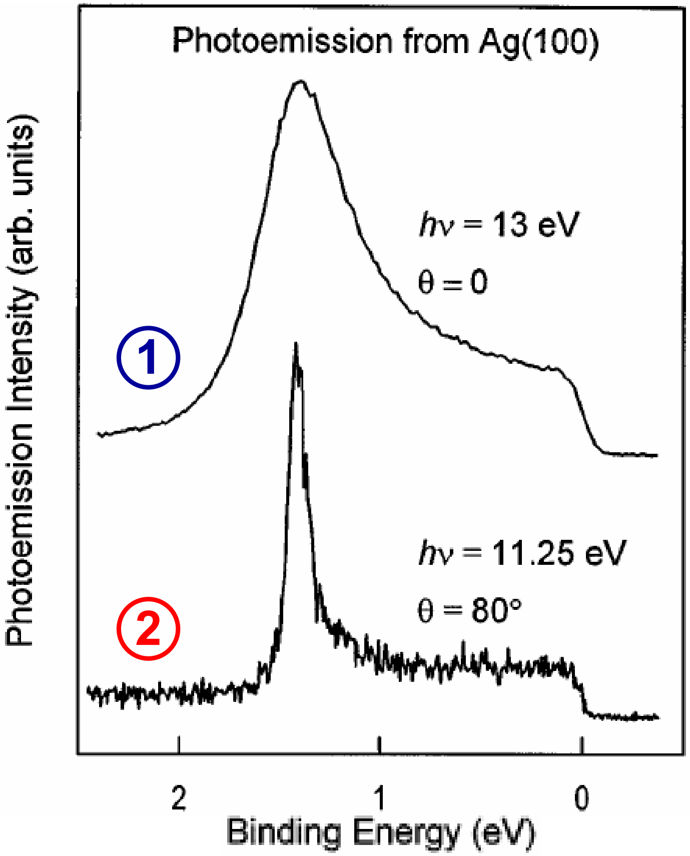


ARPES: FWHM and Inverse Lifetime

FWHM of an ARPES peak

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

Hansen *et al.*, PRL **80**, 1766 (1998)



① if $E_i \simeq E_F$

$\rightarrow \Gamma_i \longrightarrow 0 \rightarrow \boxed{\Gamma \propto \Gamma_f}$

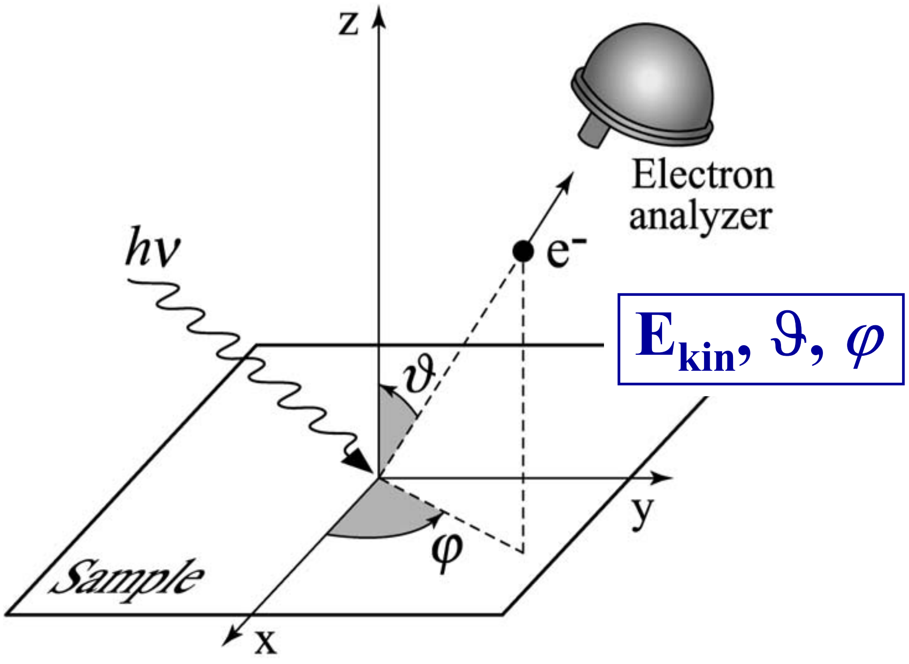
② if $|v_{i\perp}| \simeq 0$

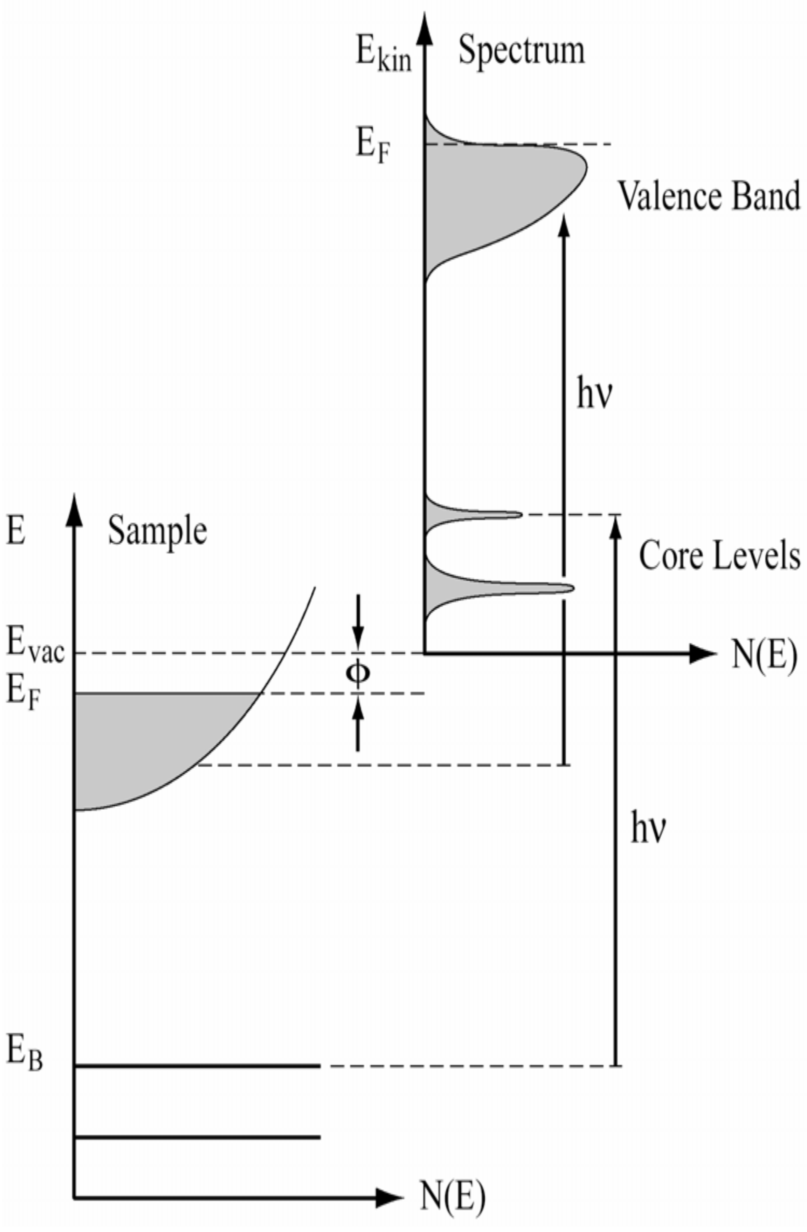
$$\rightarrow \Gamma = \frac{\Gamma_i}{\left| 1 - \dfrac{m v_{i\|} \sin^2 \vartheta}{\hbar k_\|} \right|} \equiv C\,\Gamma_i$$

if $v_{i\|} < 0$, large; θ large; $k_\|$ small

$\rightarrow C < 1$, and $\boxed{\Gamma < \Gamma_i}$

ARPES: Energetics and Kinematics





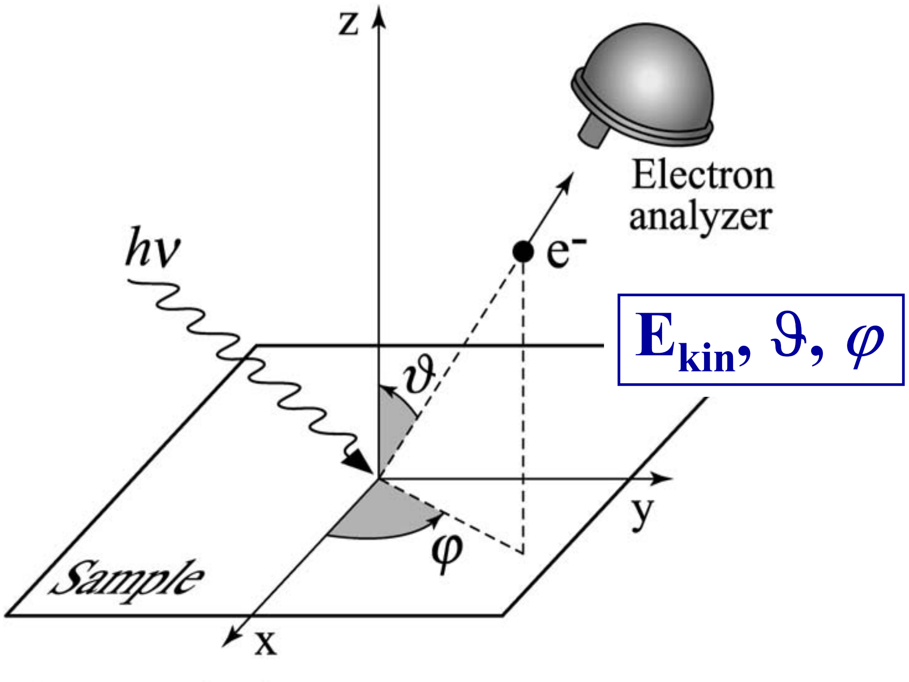
Energy Conservation

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

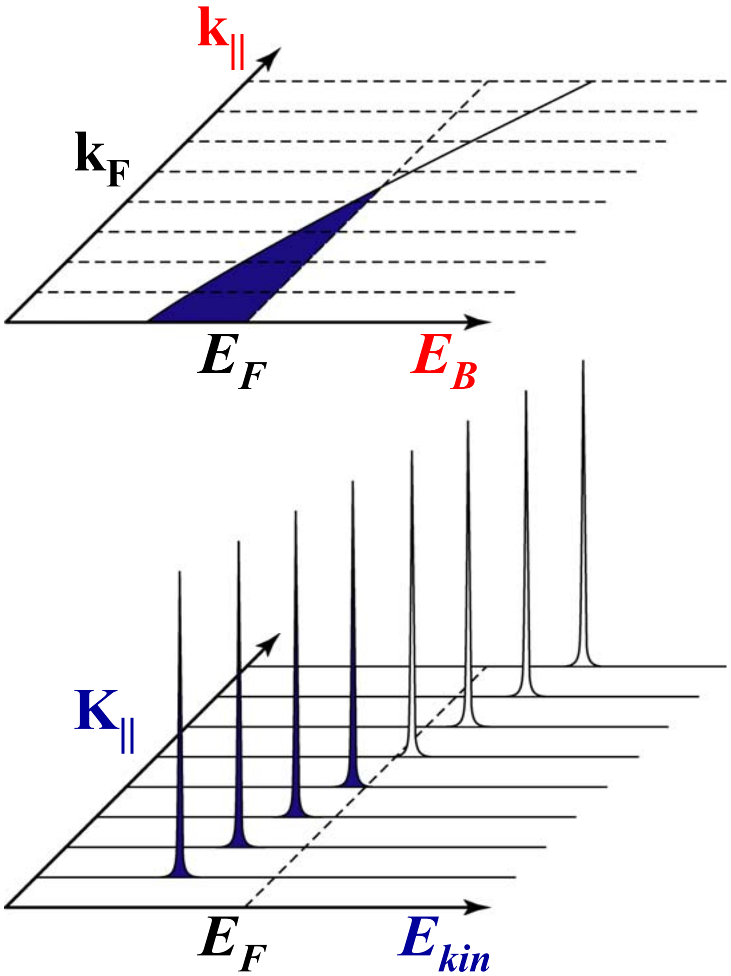
Momentum Conservation

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

ARPES: Energetics and Kinematics



Electrons in Reciprocal Space



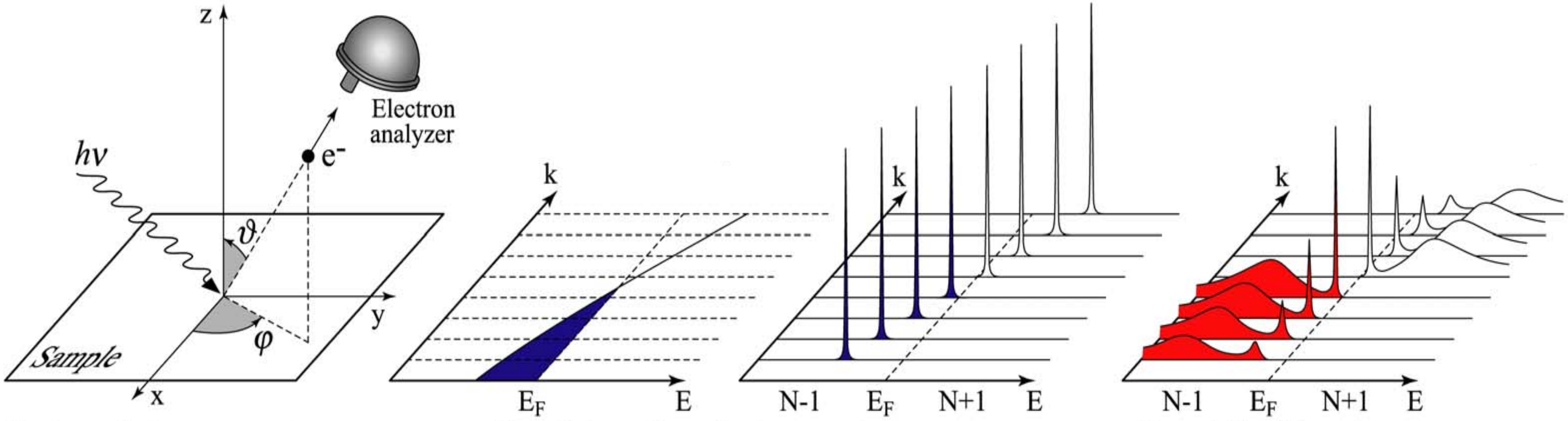
Energy Conservation

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

Momentum Conservation

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

ARPES: Interacting Systems



Photoemission geometry Non-interacting electron system Fermi liquid system

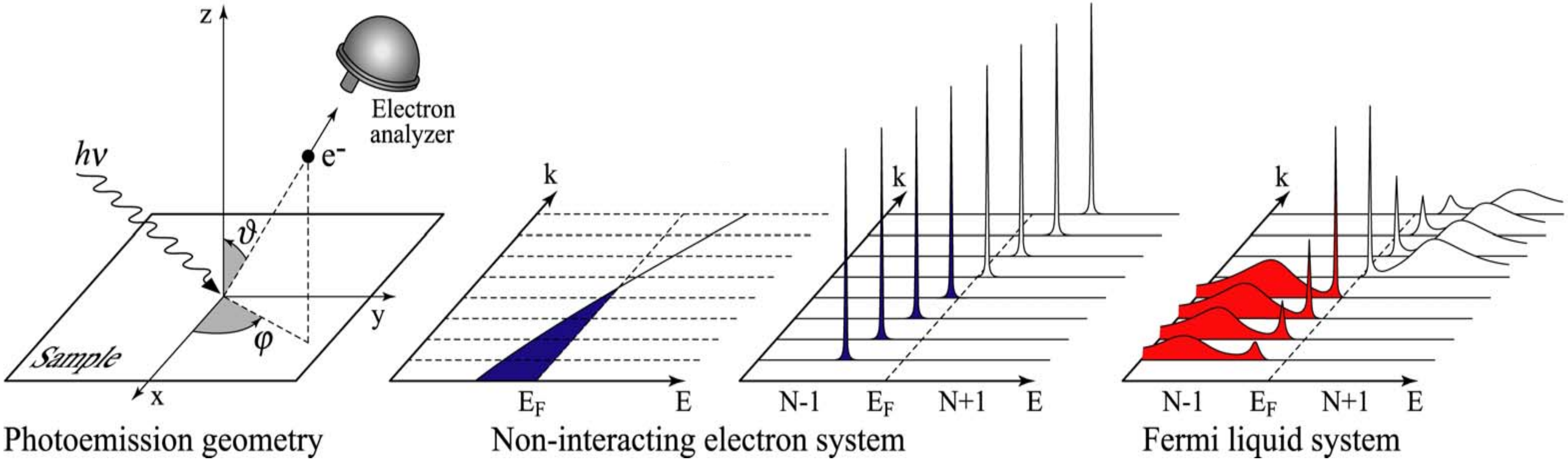
Photoemission intensity: $I(\mathbf{k}, E_{kin}) = \sum_{f,i} w_{f,i}$

$$I(\mathbf{k}, E_{kin}) \propto \sum_{f,i} |M^{\mathbf{k}}_{f,i}|^2 \sum_m |c_{m,i}|^2 \delta(E_{kin} + E^{N-1}_m - E^N_i - h\nu)$$

$$|M^{\mathbf{k}}_{f,i}|^2 \equiv |\langle \phi^{\mathbf{k}}_f | \mathbf{A} \cdot \mathbf{p} | \phi^{\mathbf{k}}_i \rangle|^2 \qquad |c_{m,i}|^2 = |\langle \Psi^{N-1}_m | \Psi^{N-1}_i \rangle|^2$$

In general $\Psi^{N-1}_i = c_{\mathbf{k}} \Psi^N_i$ **NOT orthogonal** Ψ^{N-1}_m

ARPES: The One-Particle Spectral Function



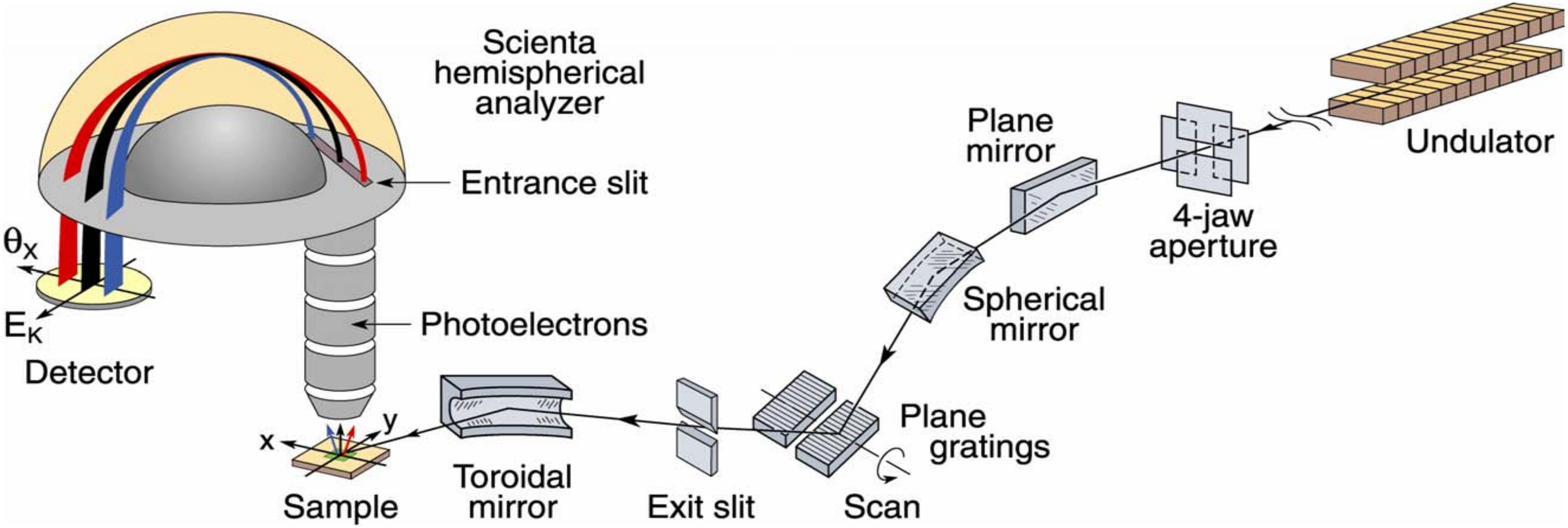
Photoemission intensity: $I(k,\omega)=I_0\,|M(k,\omega)|^2 f(\omega)\,A(k,\omega)$

Single-particle spectral function

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

$\Sigma(k,\omega)$: the “self-energy” captures the effects of interactions

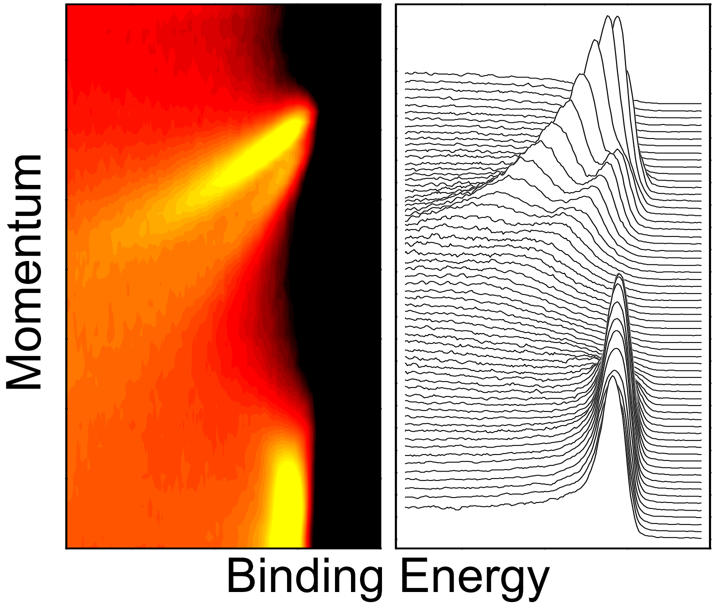
Angle-Resolved Photoemission Spectroscopy



Parallel multi-angle recording

- Improved **energy resolution**
- Improved **momentum resolution**
- Improved **data-acquisition efficiency**

	ΔE (meV)	*Δθ*
past	*20-40*	*2°*
now	*2-10*	*0.2°*

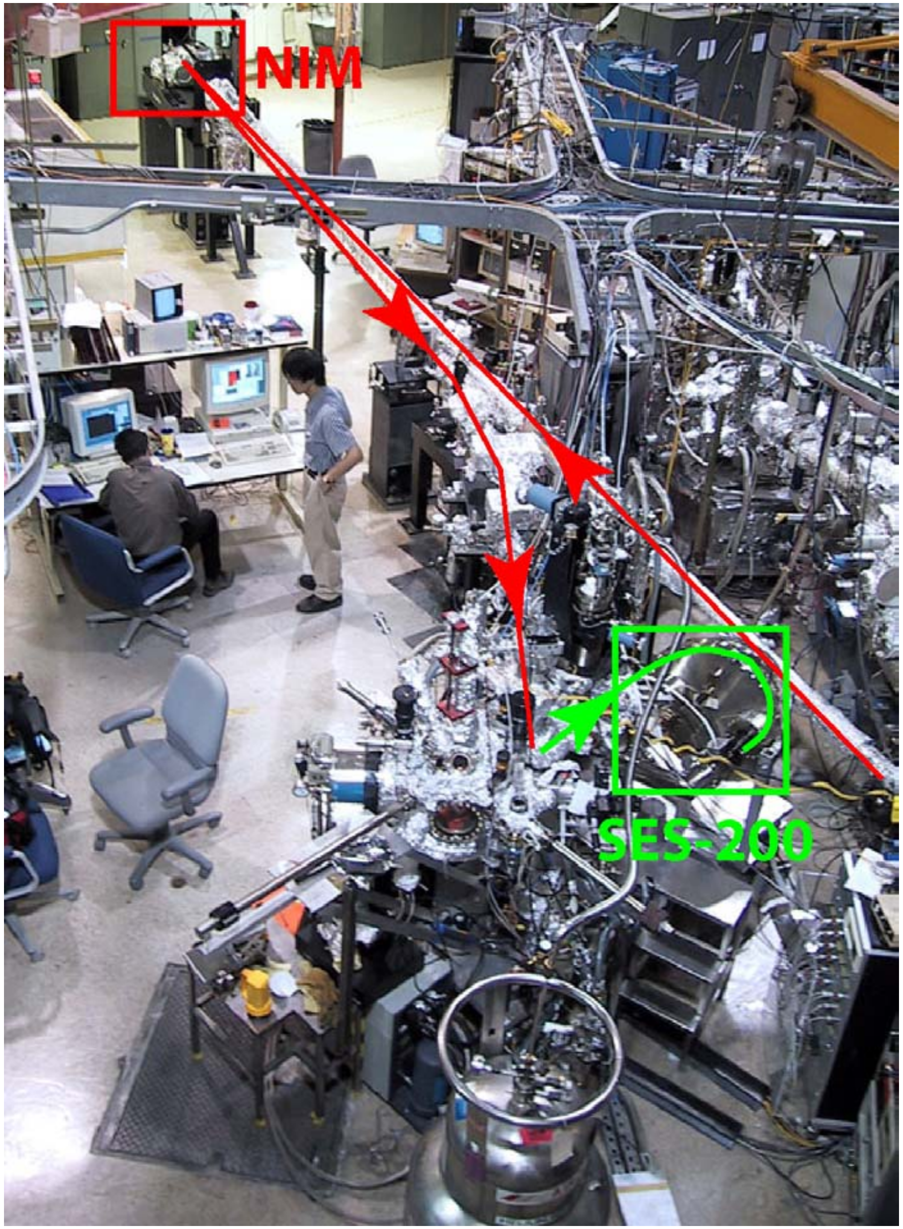


SSRL Beamline 5-4 : NIM / Scienta System

- **High resolution**

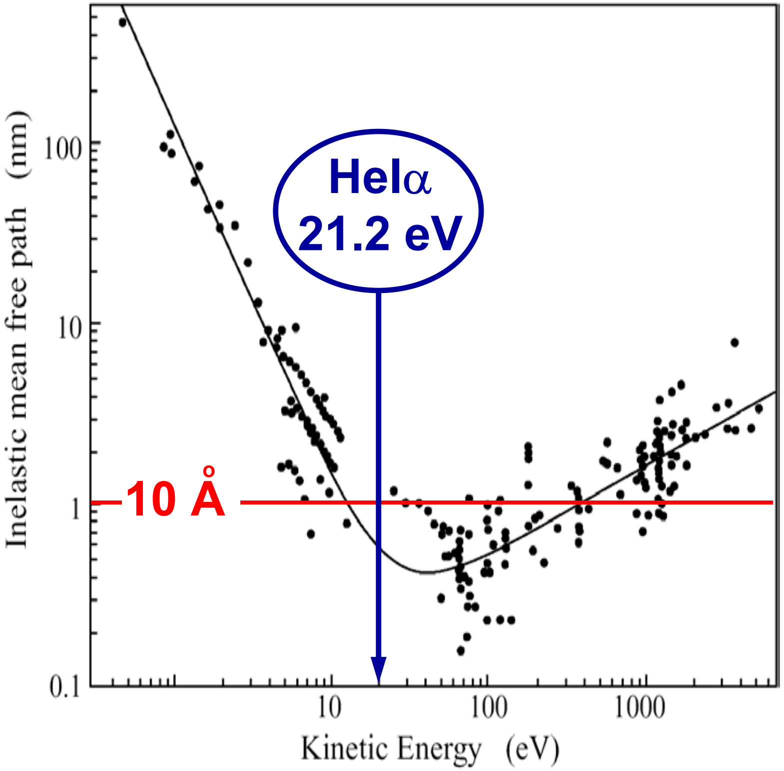
ΔE (meV)	$\Delta\theta$
2-10	0.2°

- Ultra-high vacuum (~ 10^{-11} torr)
- High angular precision (+/- 0.1°)
- Low base temperature (< 10 K)
- Wide temperature range (10-350 K)
- Variable photon energies (12-30 eV)
- Multiple light sources (He lamp)
- Control of light polarization
- Single crystal cleaving tools
- Sample surface preparation & cleaning
- Low-Energy Electron Diffraction (LEED)

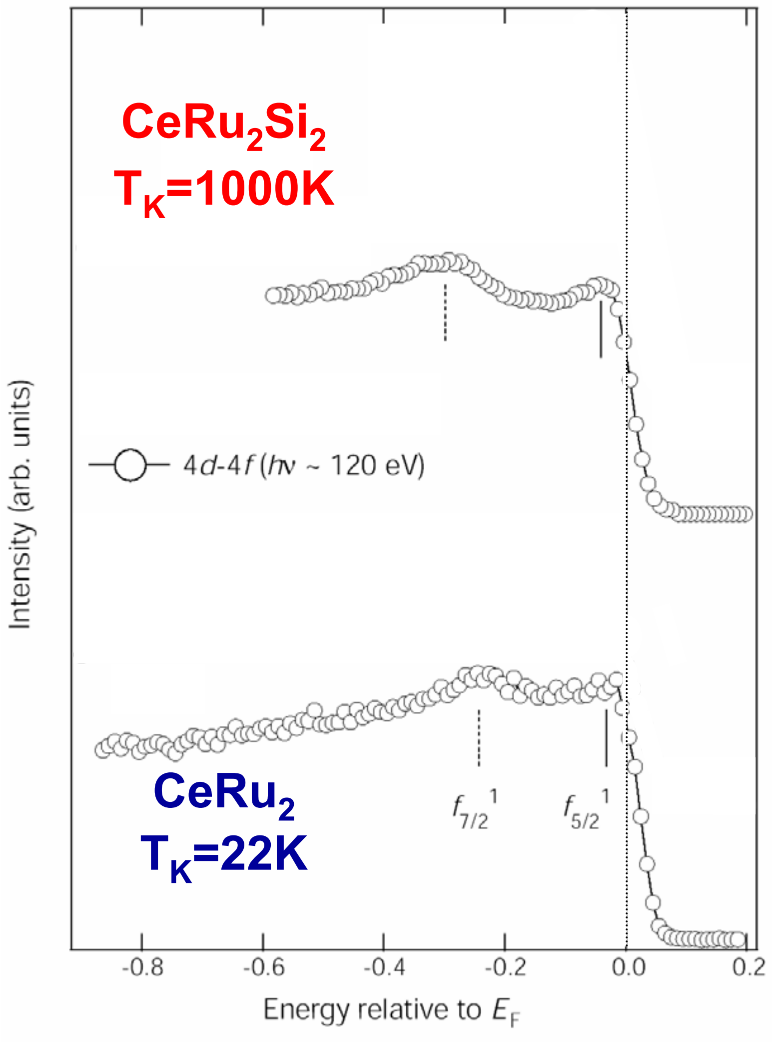


ARPES: Surface vs Bulk Sensitivity

Mean-free path for excited electrons



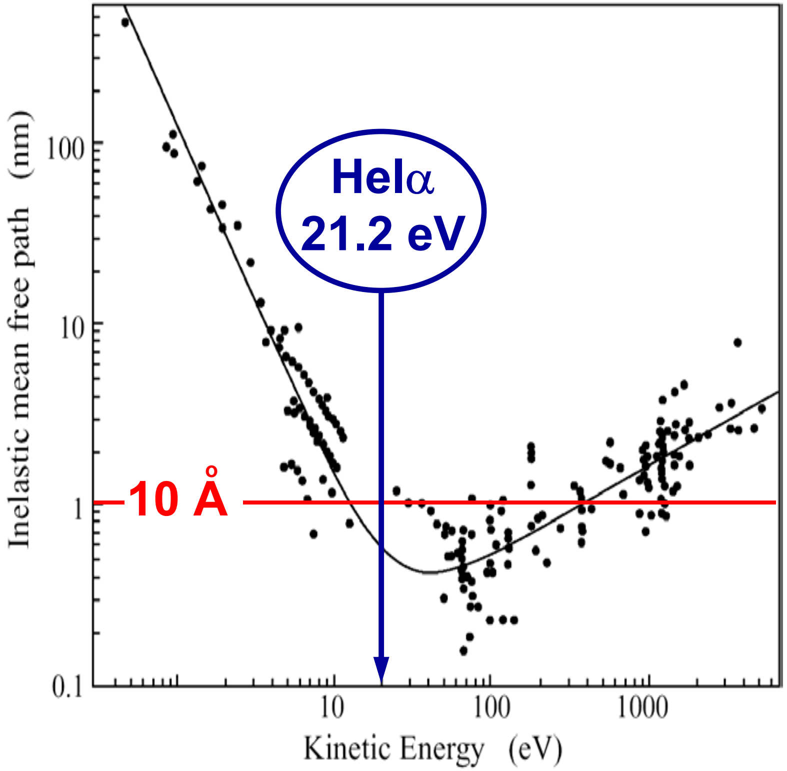
Seah, Dench *et al.*, SIA **1**, 2 (1979)



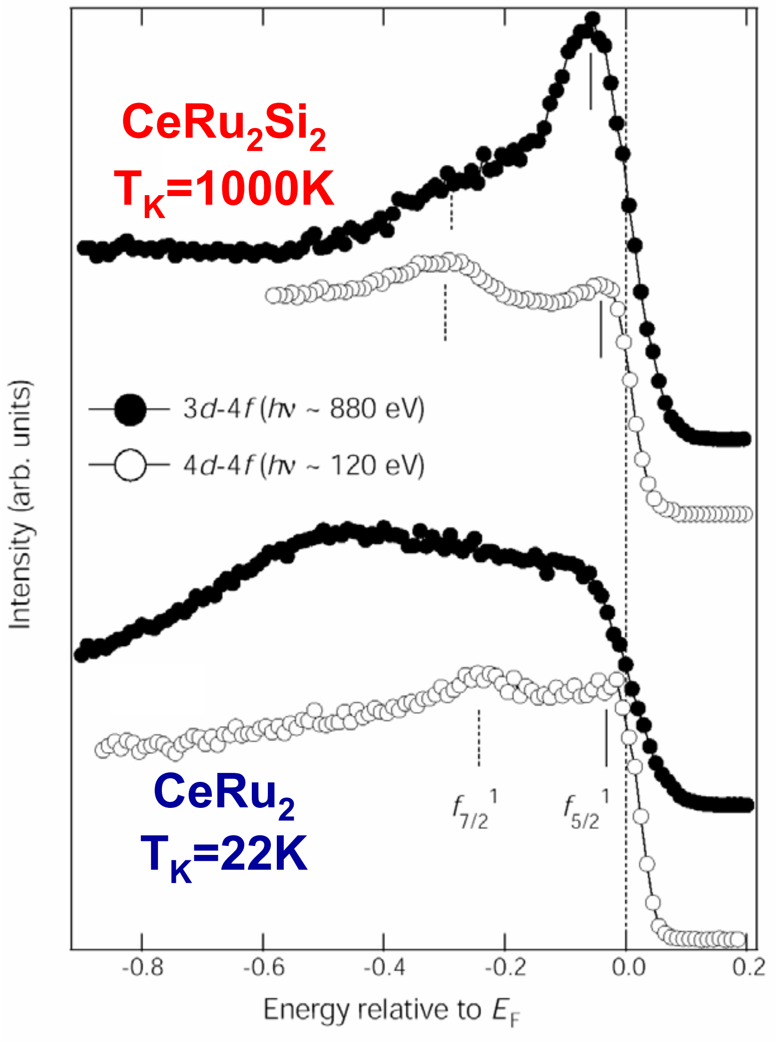
Sekiyama *et al.*, Nature **403**, 396 (2000)

ARPES: Surface vs Bulk Sensitivity

Mean-free path for excited electrons



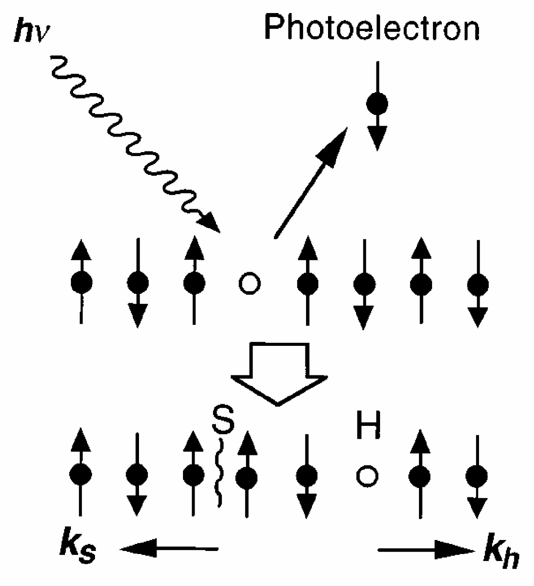
Seah, Dench *et al.*, SIA **1**, 2 (1979)



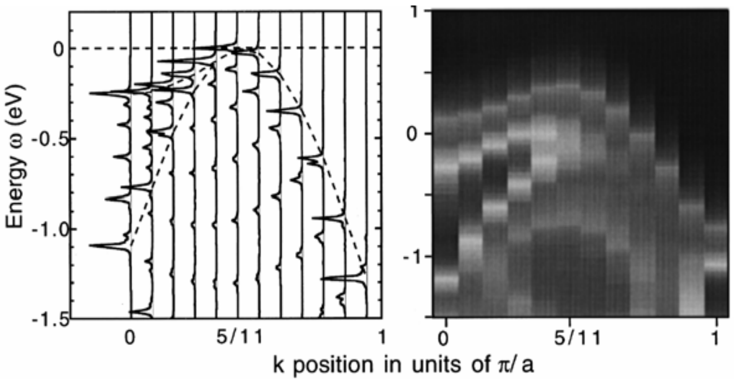
Sekiyama *et al.*, Nature **403**, 396 (2000)

ARPES on 1D Nanostructures: Spin-Charge Separation?

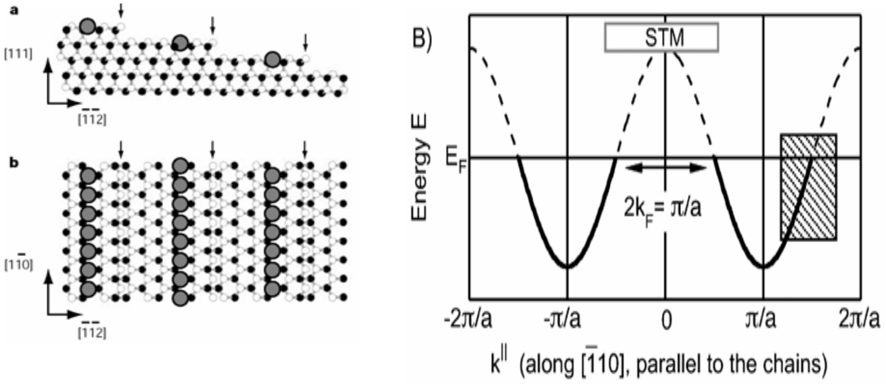
C. Kim *et al.*, PRL **77**, 4054 (1996)



$SrCuO_2$

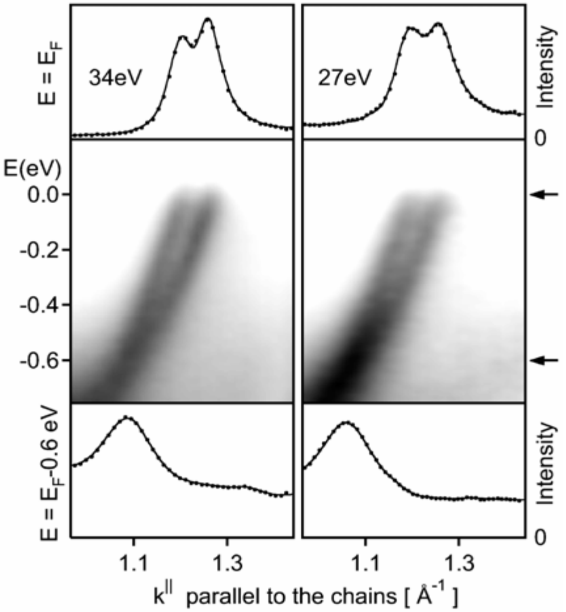


P. Segovia *et al.*, Nature **402**, 504 (1999)



Au chains on Si(557)

Losio *et al.*, PRL **86**, 4632 (2001)

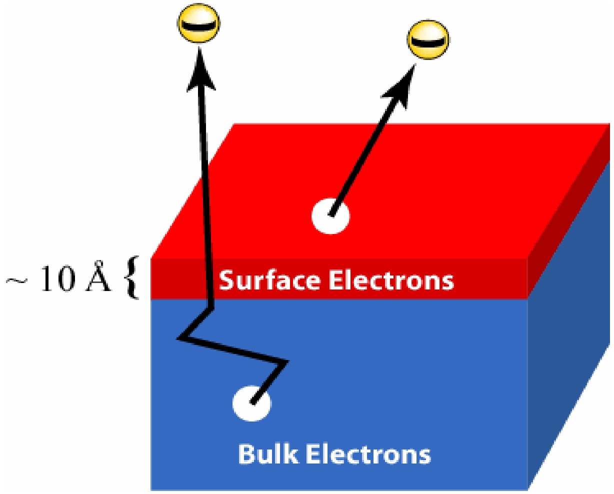


ARPES: Advantages and Limitations

Advantages

- **Direct information about the electronic states!**
- Straightforward comparison with theory - little or no modeling.
- High-resolution information about **BOTH energy** and **momentum**
- **Surface-sensitive probe**
- Sensitive to “**many-body**” effects
- Can be applied to small samples (100 μm x 100 μm x 10 nm)

Limitations



- **Not bulk sensitive**
- Requires clean, atomically flat surfaces in **ultra-high vacuum**
- Cannot be studied as a function of pressure or magnetic field

Courtesy of Kyle Shen

Outline: Part II

Electronic structure of complex systems

- **2*H*-$NbSe_2$ - Sr_2RuO_4**
  - Detecting bands and Fermi surface
  - Bulk & surface electronic structure

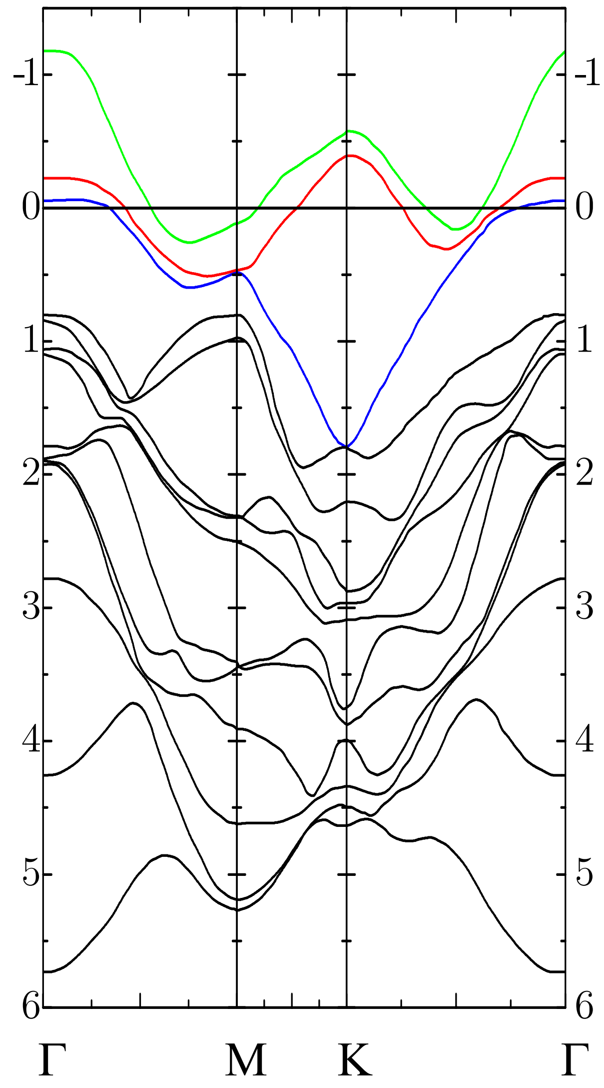
- **Nb - $Bi_2Sr_2CaCu_2O_{8+\delta}$**
  - Superconducting gap: s-wave vs. d-wave
  - Bogoliubov quasiparticles in high-T_c cuprates

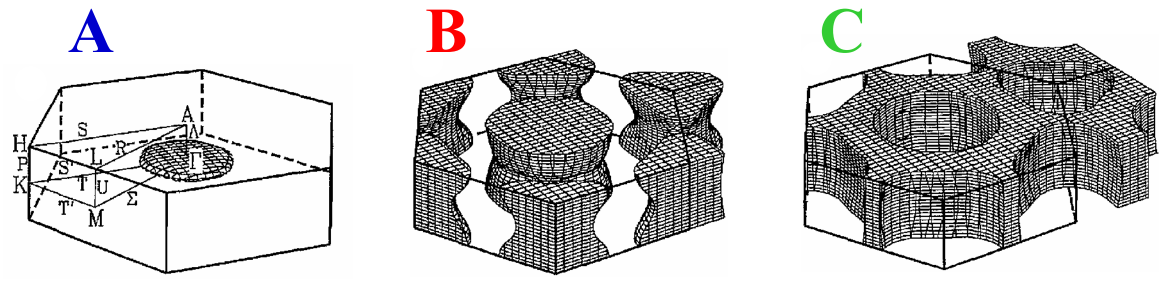
- **Be(0001) - Mo(110) - $Bi_2Sr_2CaCu_2O_{8+\delta}$**
  - Many-body effects in the quasiparticle dispersion

- Conclusions and discussion

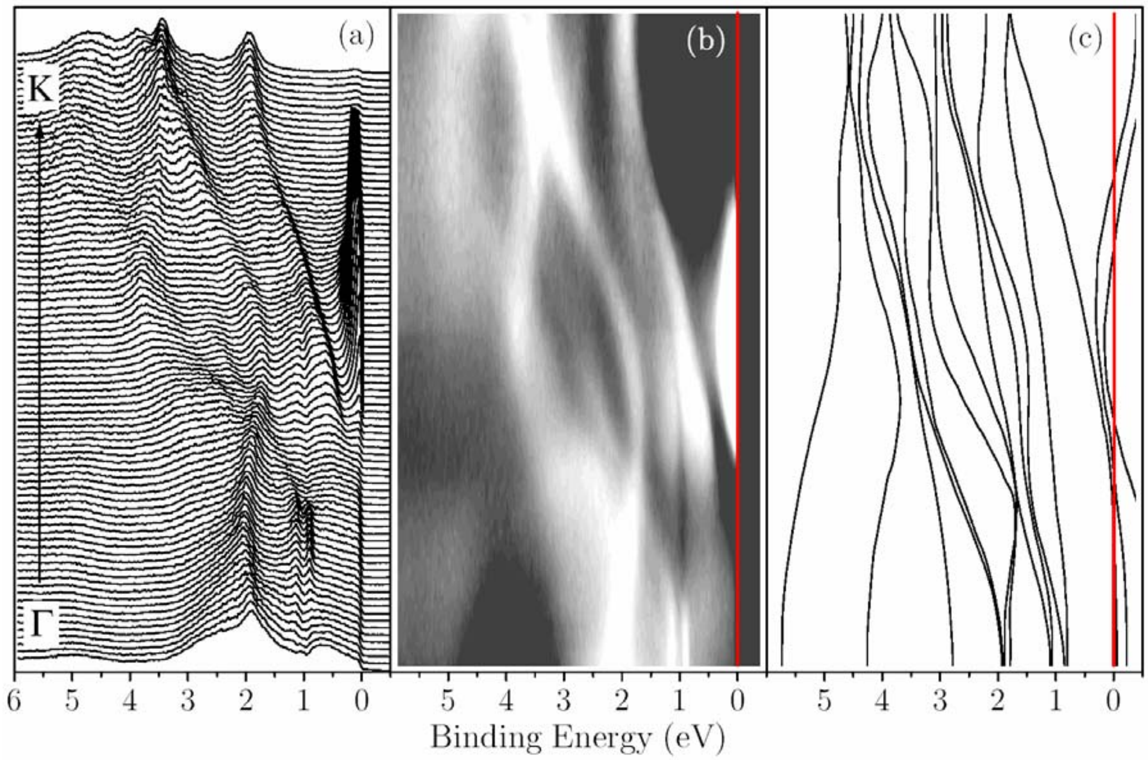
2*H*-NbSe$_2$: Normal State Electronic Structure

Corcoran *et al.*,JPCM **6**, 4479 (1994)



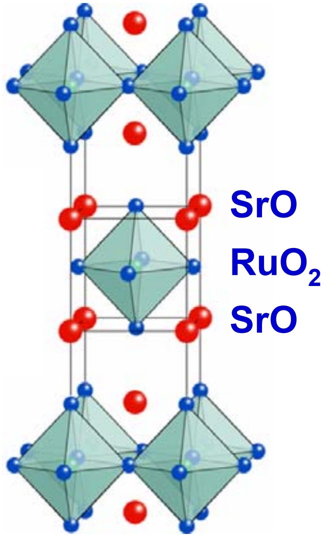


Damascelli *et al*. (2000)



Sr_2RuO_4: basic properties

2D perovskite

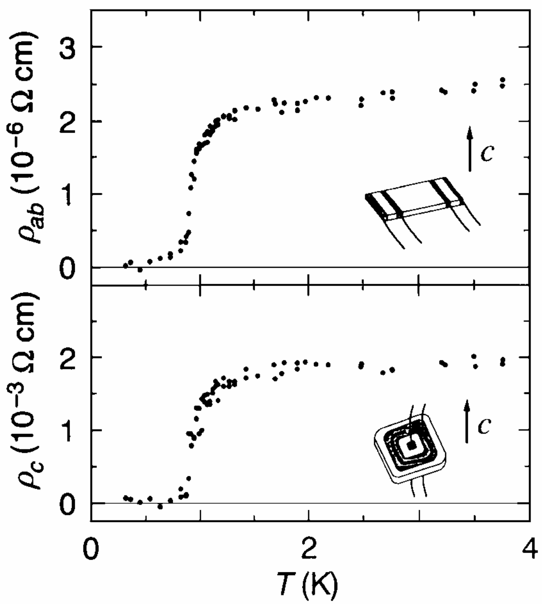


Unconventional superconductivity

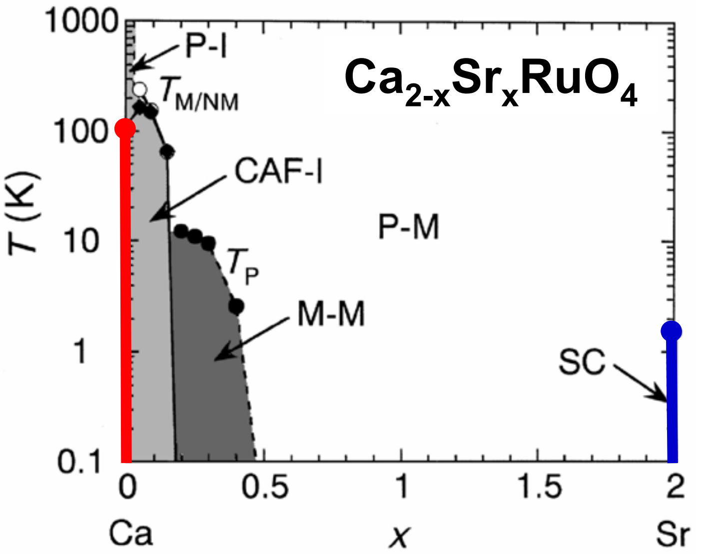
- Pairing mechanism ?
- Order parameter ?
- FM-AF fluctuations ?

Rice & Sigrist, JPCM **7**, L643 (1995)

Maeno *et al.*, Nature **372**, 532 (1994)



Nakatsuji & Maeno, PRL **84**, 2666 (2000)



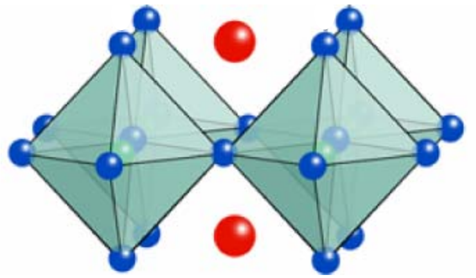
Lattice-magnetism interplay
Orbital degrees of freedom

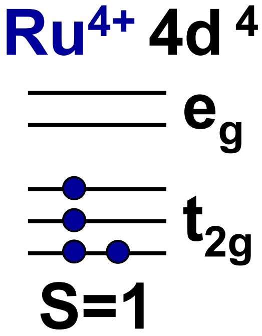
Sr_2RuO_4 : 2D **Fermi Liquid** (ρ_c/ρ_{ab}=850)

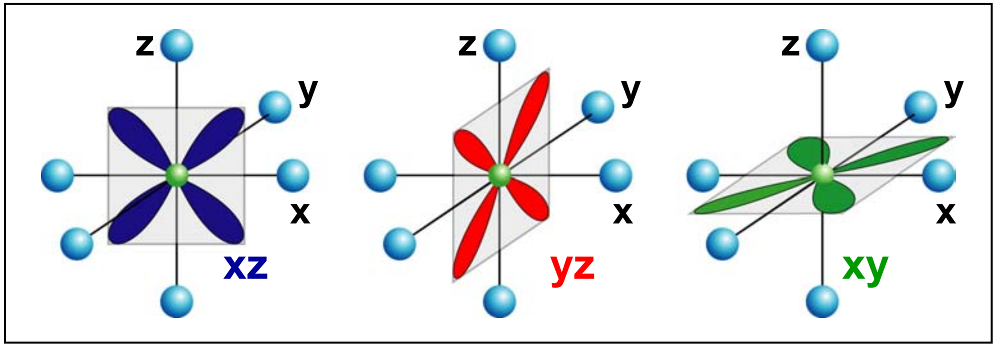
Ca_2RuO_4 : insulating **Anti-FerroMagnet**

$SrRuO_3$: metallic **FerroMagnet**

Low-Energy Electronic structure of Sr_2RuO_4

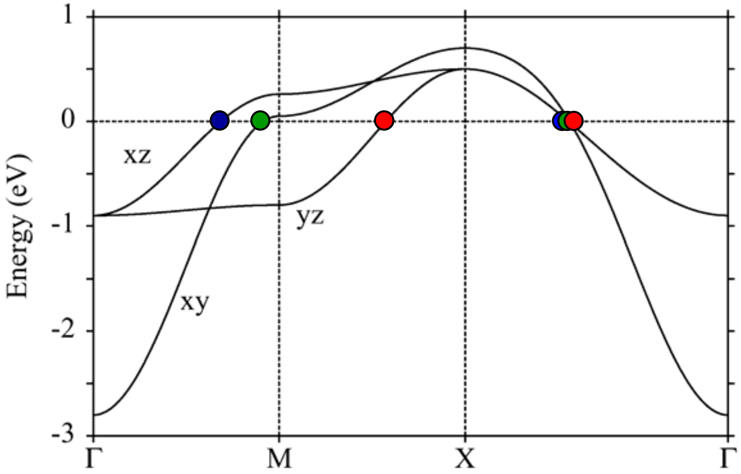




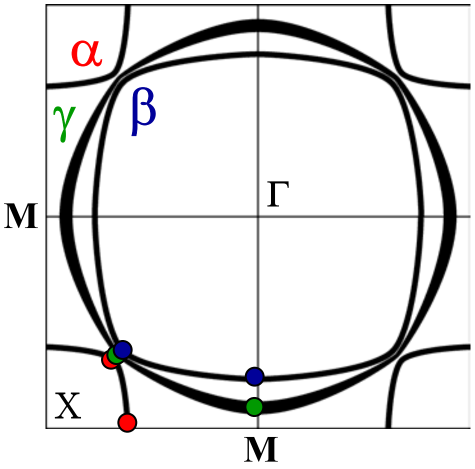


- ▶ Band structure calculation: **3 t_{2g}** bands crossing **E_F**

→ 3 sheets of FS $\begin{cases} \alpha \text{ (hole-like)} \\ \beta \text{ and } \gamma \text{ (electron-like)} \end{cases}$



A. Liebsch *et al*, PRL **84**, 1591 (2000)

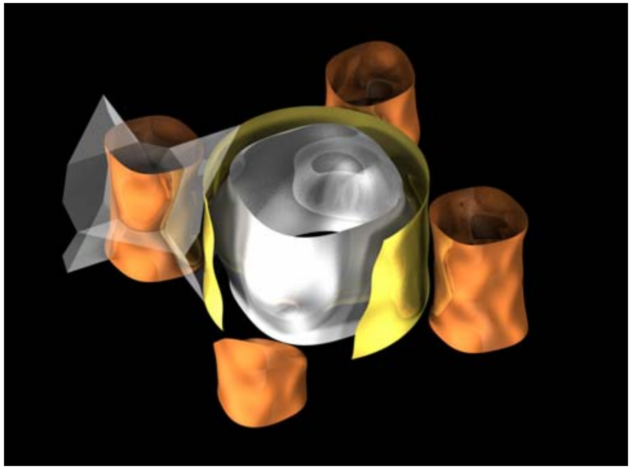


I.I. Mazin *et al*, PRL **79**, 733 (1997)

Fermi Surface Topology of Sr_2RuO_4

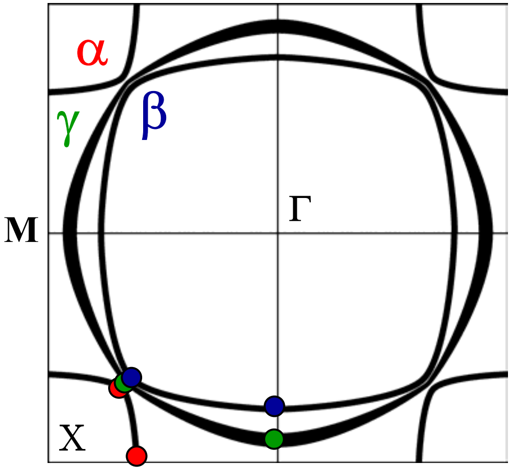
- Early ARPES results gave a different topology

de Haas-van Alphen



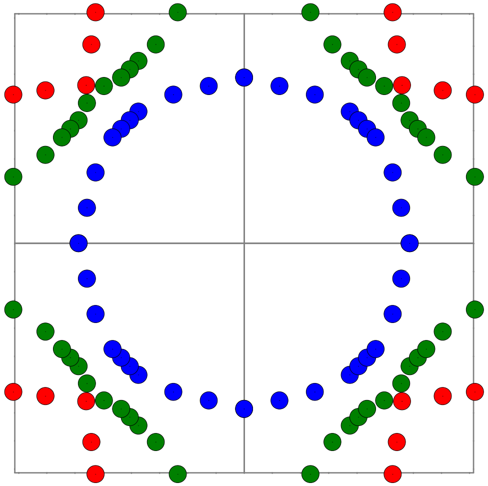
A.P. Mackenzie *et al.*, PRL **76**, 3786 (1996)
C. Bergemann *et al.*, PRL **84**, 2662 (2000)

LDA



I.I. Mazin *et al.*, PRL **79**, 733 (1997)

ARPES



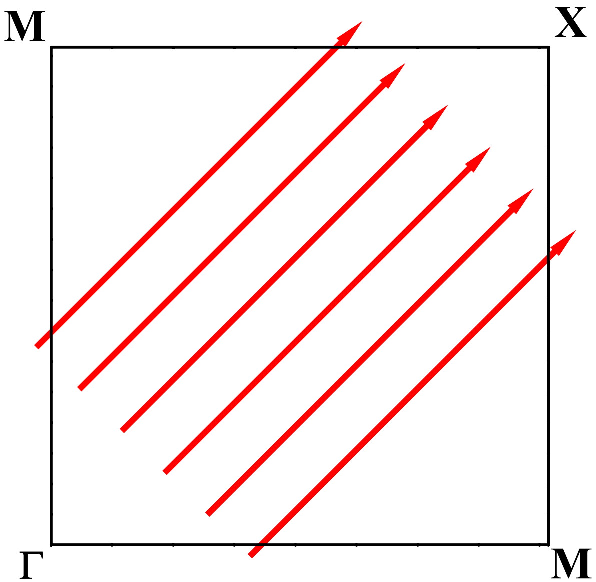
T.Yokoya *et al.*, PRB **54**, 13311 (1996)
D.H. Lu *et al.*, PRL **76**, 4845 (1996)

ARPES: additional information

dHvA: limited set of systems

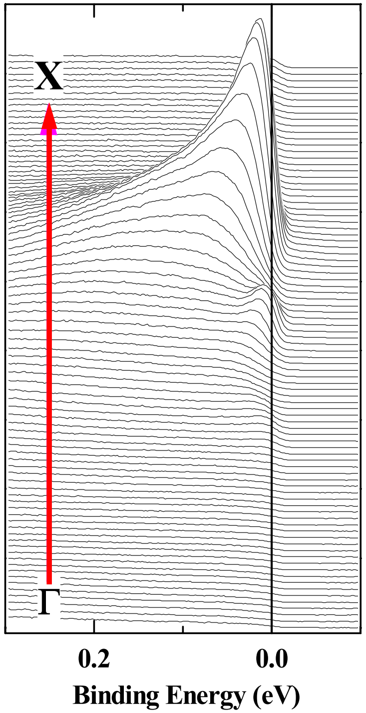
Fermi Surface Topology of Sr_2RuO_4

ARPES : present day

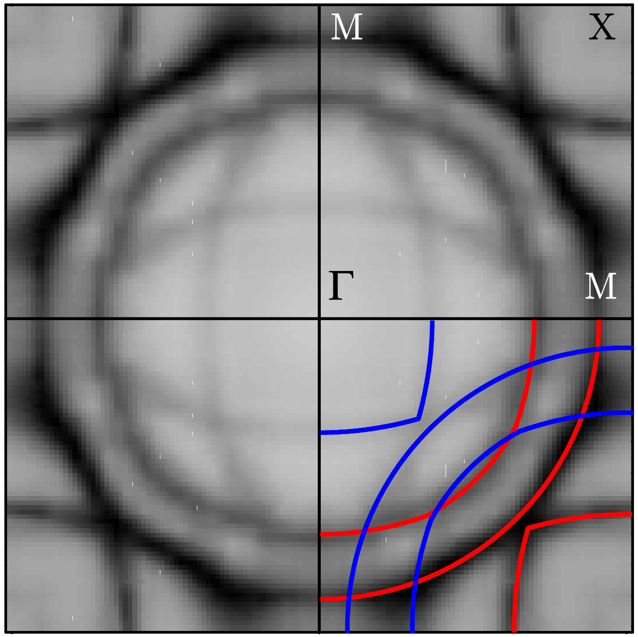


A. Damascelli *et al.*,
PRL **85**, 5194 (2000)

ARPES Spectra

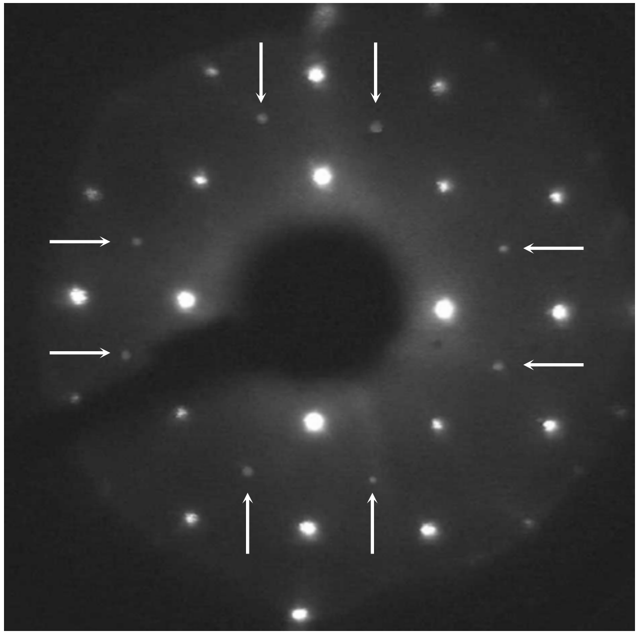


Intensity at E_F



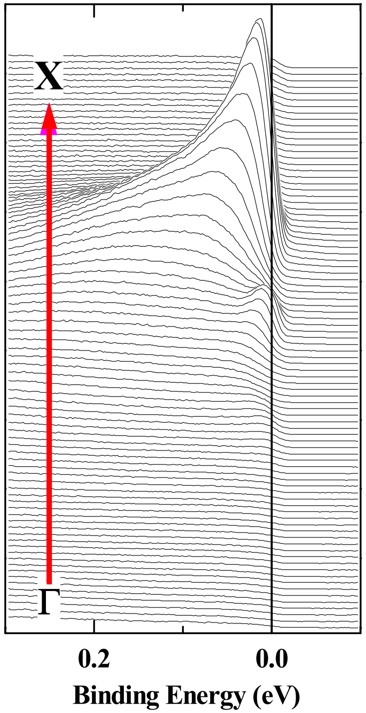
Fermi Surface Topology of Sr_2RuO_4

LEED Pattern

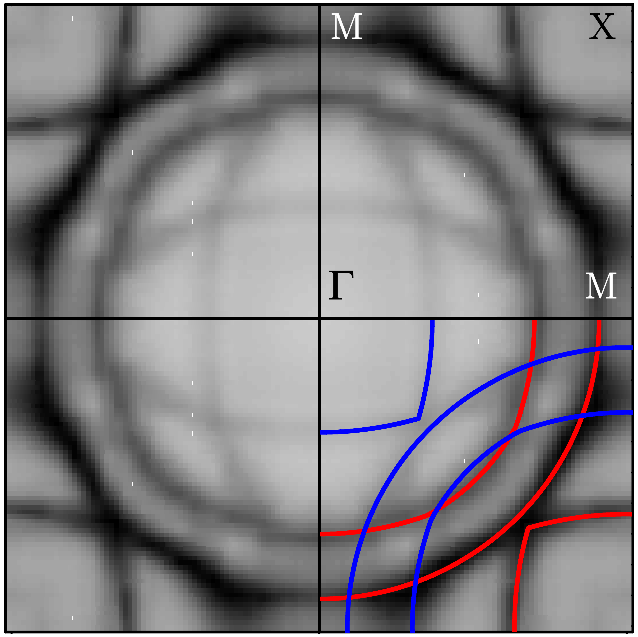


A. Damascelli *et al.*,
PRL **85**, 5194 (2000)

ARPES Spectra

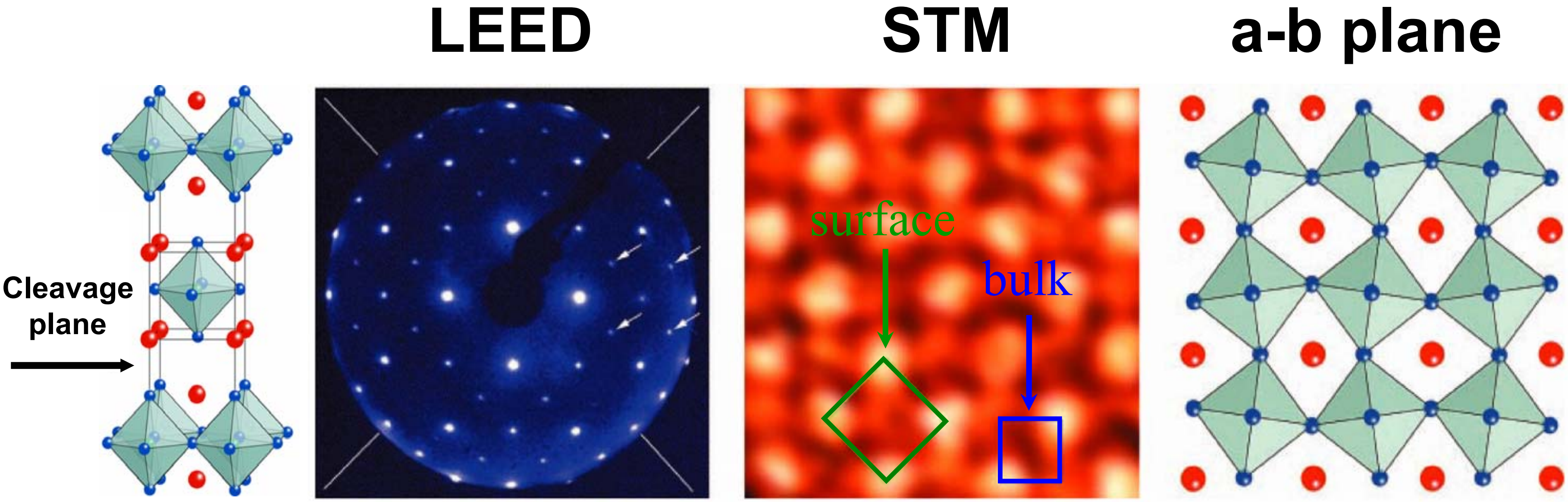


Intensity at E_F



Surface instability ↔ **Band folding**

Surface reconstruction of cleaved Sr_2RuO_4



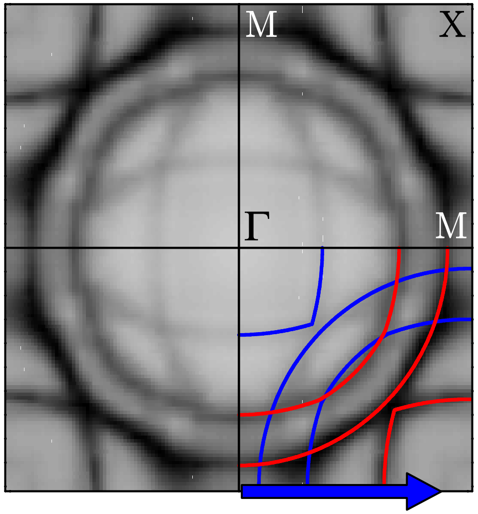
R. Matzdorf *et al.*, Science **289**, 746 (2000)

Rotation of the RuO_6 octahedra around the c axis (9°)

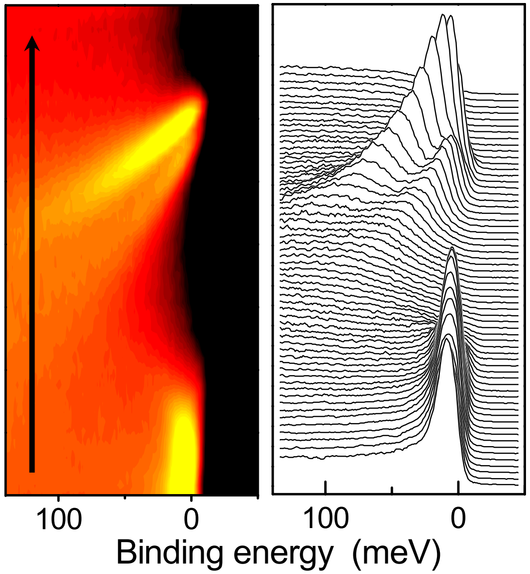
Surface electronic structure of Sr_2RuO_4

On samples cleaved at **180 K** the **surface**-related features are **suppressed**

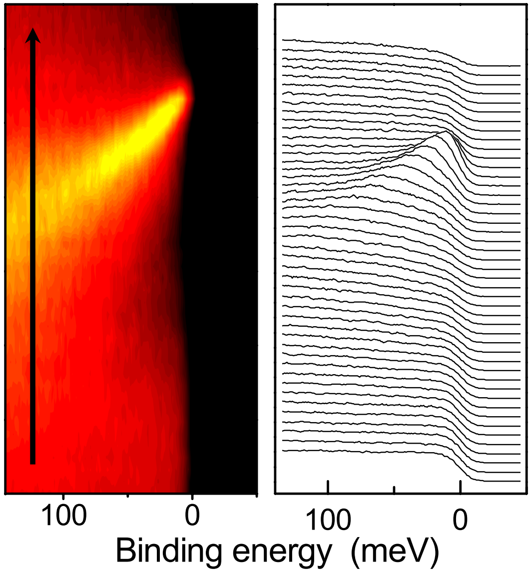
E_F mapping
±10 meV



Cold cleave
T=10 K



Hot cleave
T=180 K

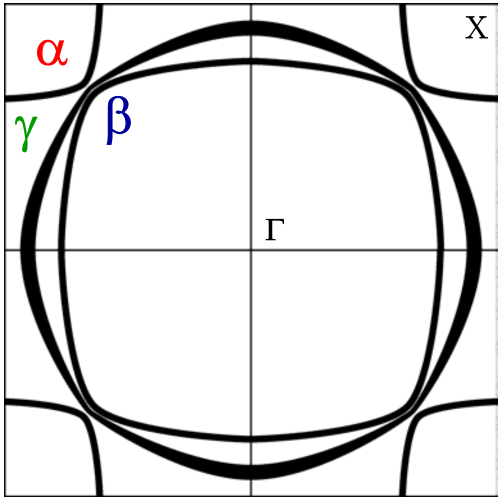


A. Damascelli *et al.,* PRL **85**, 5194 (2000); K.M. Shen, A. Damascelli, *et al.*, PRB **64**, 180502(R) (2001)

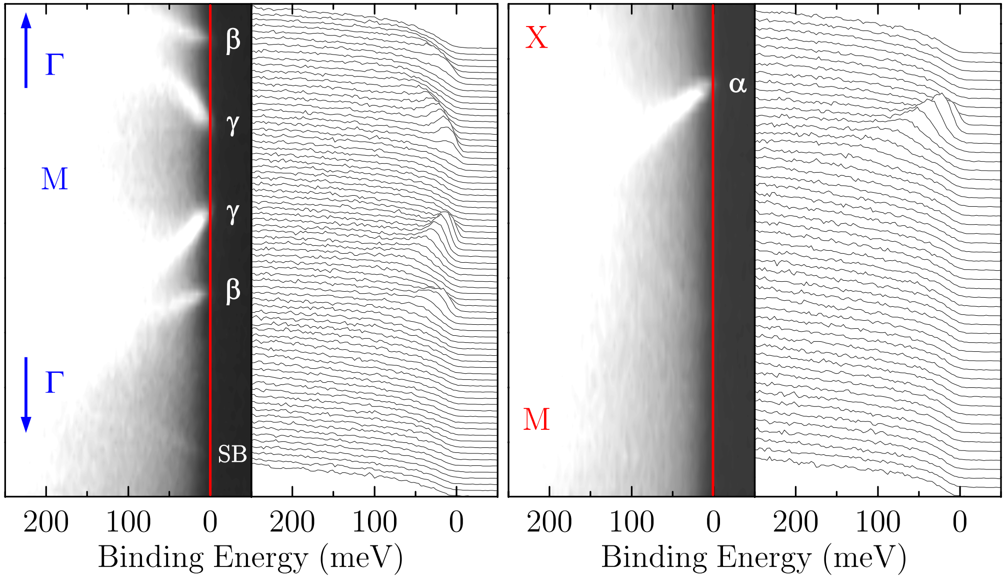
Bulk electronic structure of Sr_2RuO_4

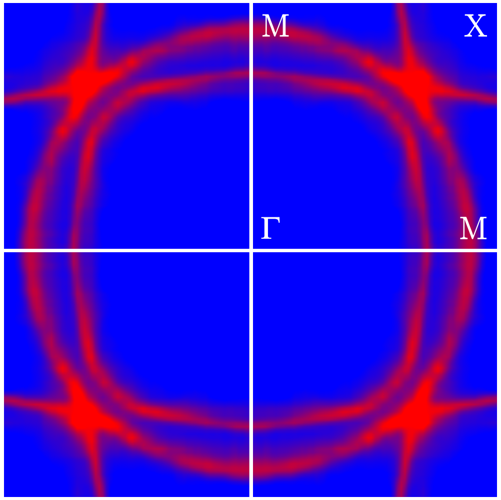
What do we learn about the **bulk electronic structure**?

- **Fermi Surface**
- **Fermi velocity**
- **Effective mass**



I.I. Mazin *et al.*, PRL **79**, 733 (1997)



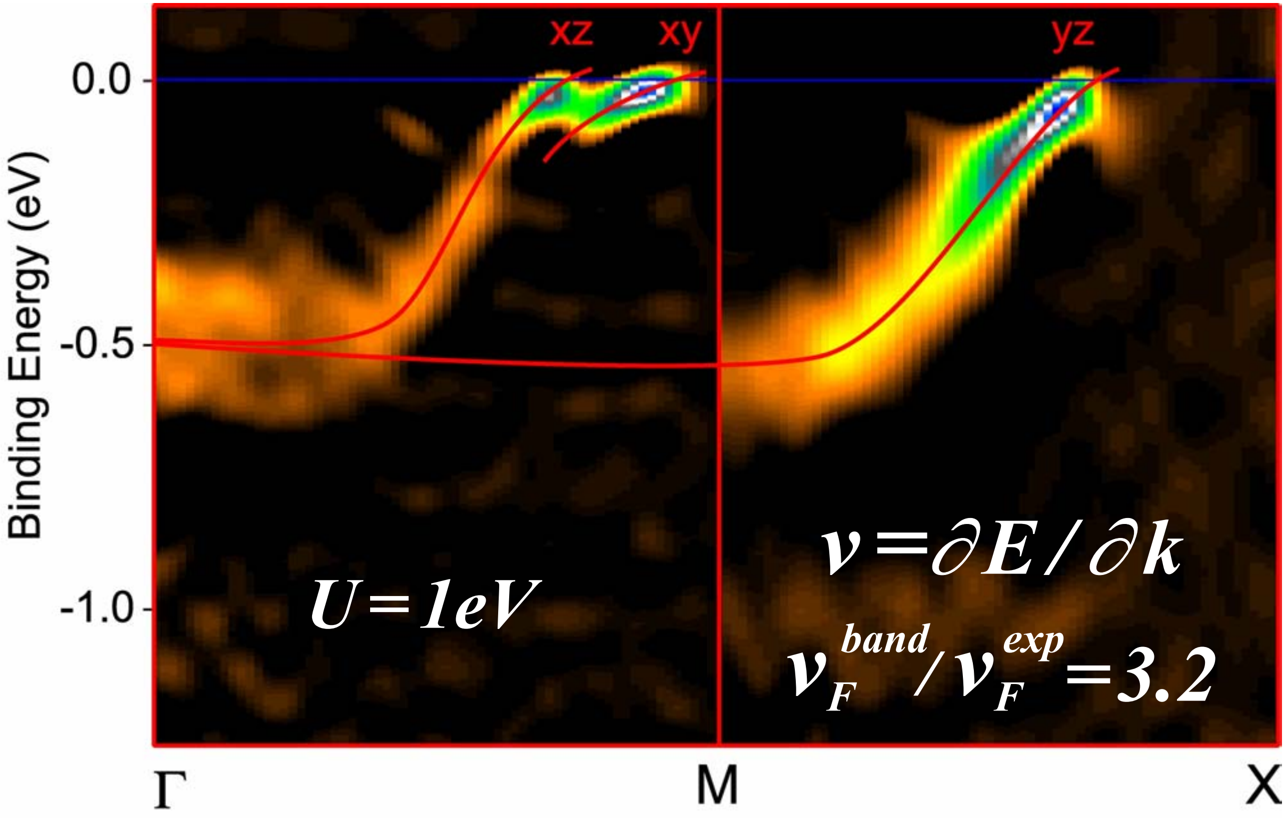


Sr_2RuO_4 cleaved at 180 K

T= 10 K hν=28 eV

A. Damascelli *et al.*, PRL **85**, 5194 (2000)

Dispersion of the bulk electronic bands

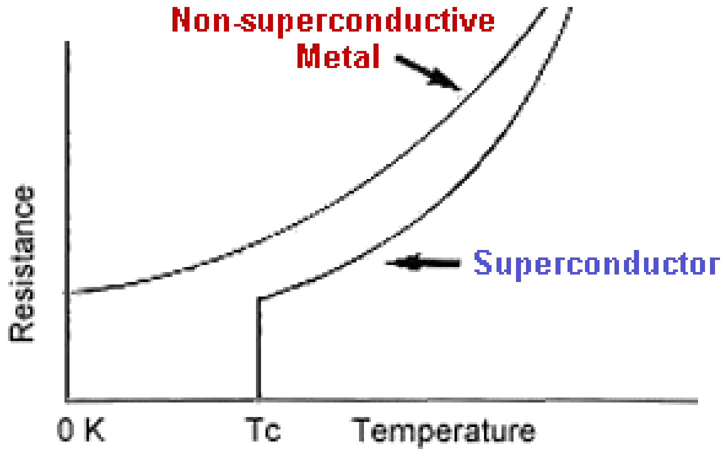


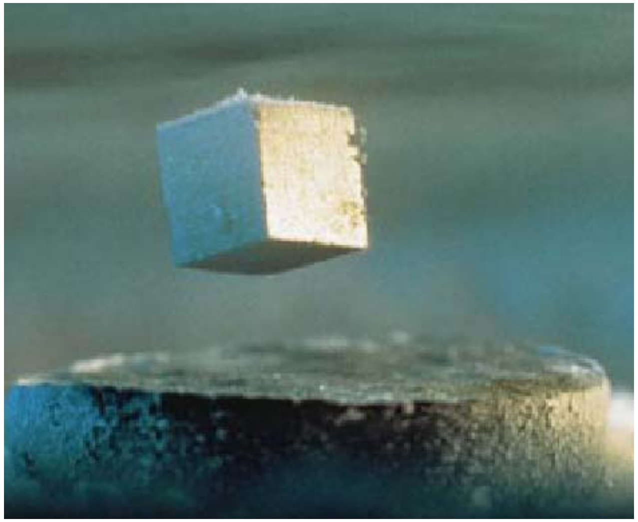
Experiment compares well with LDA+U calculations

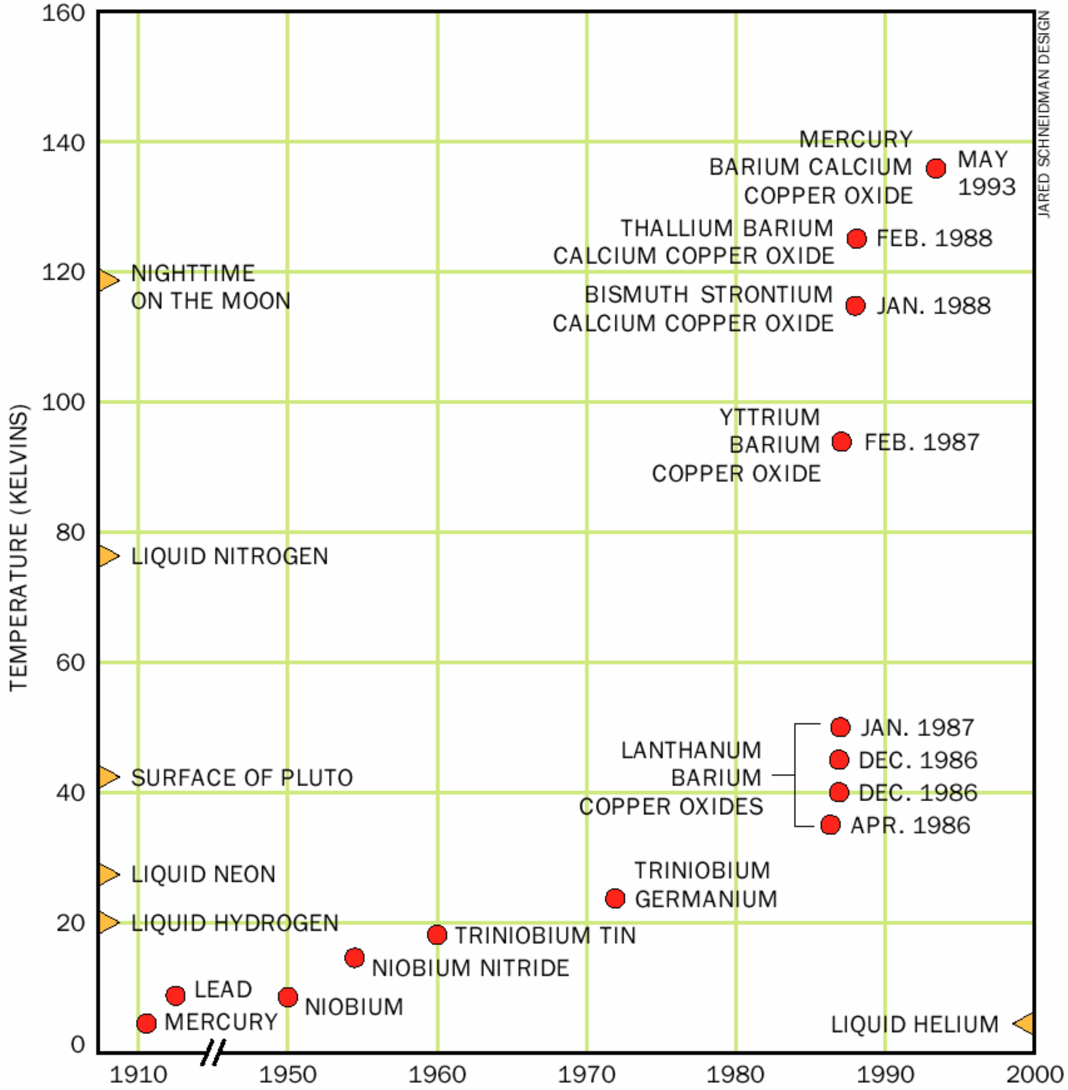
A. Damascelli *et al.,* PRL **85**, 5194 (2000)

A. Liebsch & A. Lichtenstein, PRL **84**, 1591 (2000)

Superconductivity



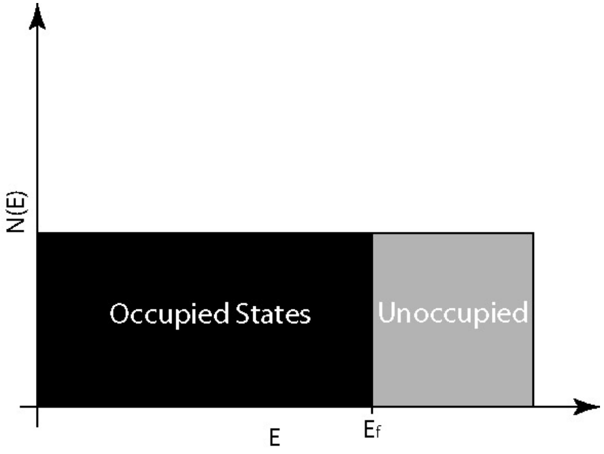




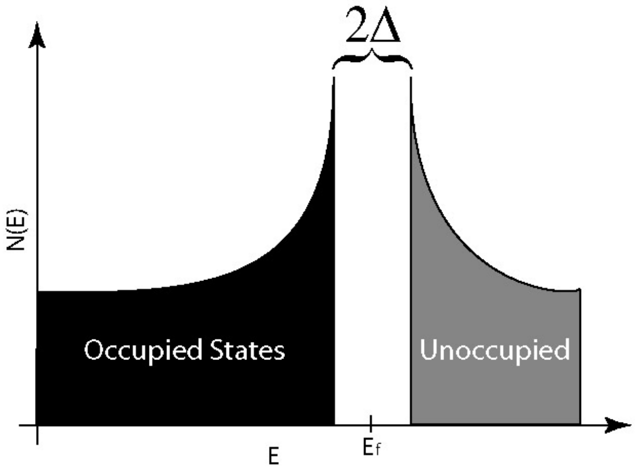
Courtesy of Kyle Shen

“Classic Low-temperature” Superconductors

Metallic Density of States



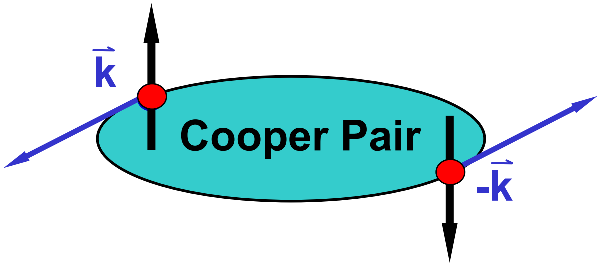
Superconducting Density of States



Superconductivity can only be seen on low energy scales and needs **high resolution**!

Superconductivity

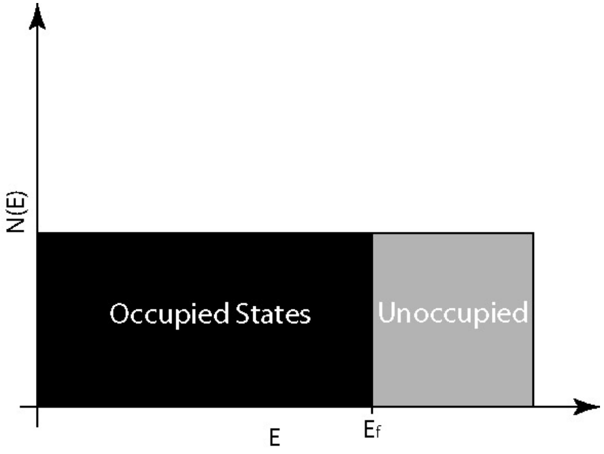
2-electron bound state



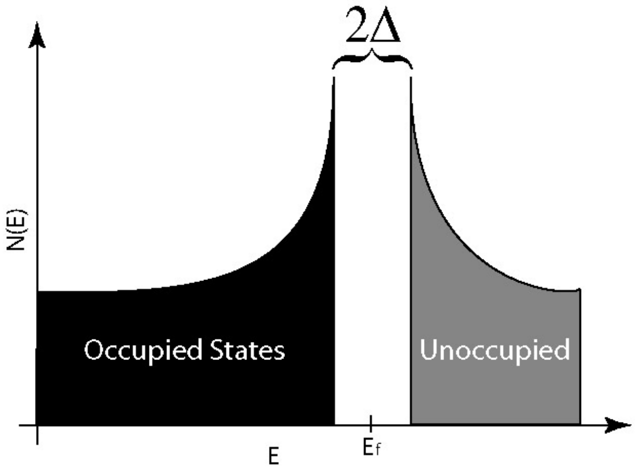
spin-singlet pairing

“Classic Low-temperature” Superconductors

Metallic Density of States

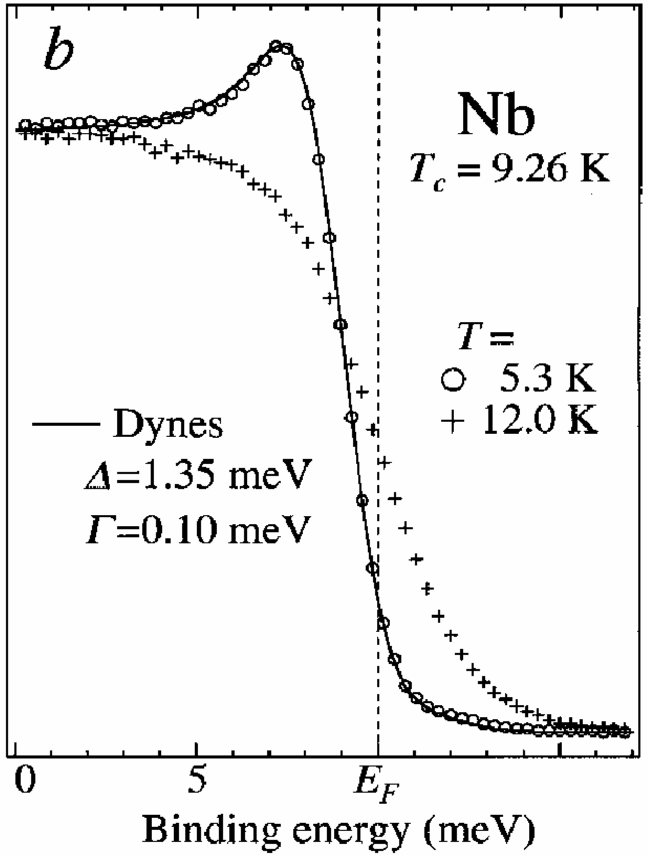


Superconducting Density of States

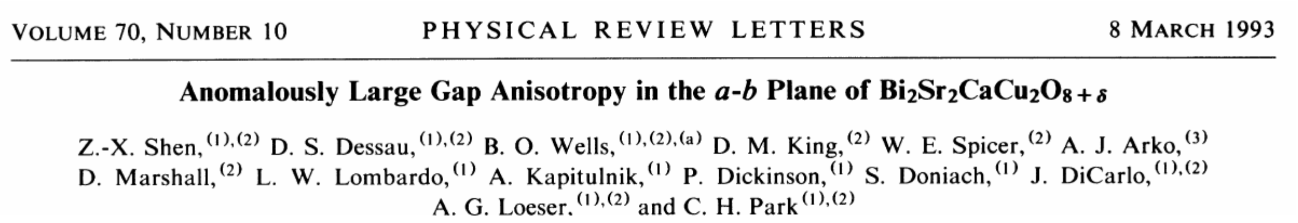


Superconductivity can only be seen on low energy scales and needs **high resolution**!

A. Chainani et al., PRL **85** (2000)



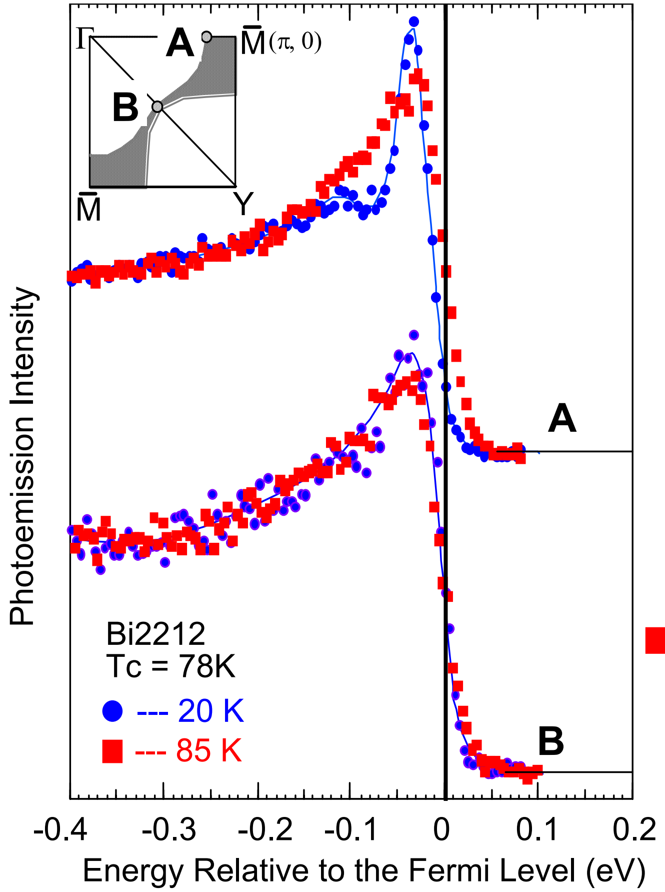
High-Temperature Superconductors: Bi2212



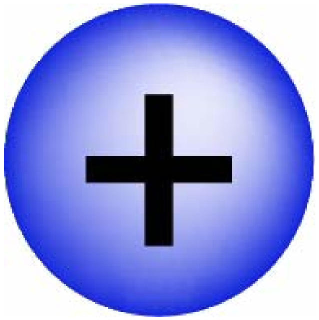
VOLUME 70, NUMBER 10 PHYSICAL REVIEW LETTERS 8 MARCH 1993

Anomalously Large Gap Anisotropy in the *a-b* Plane of $Bi_2Sr_2CaCu_2O_{8+\delta}$

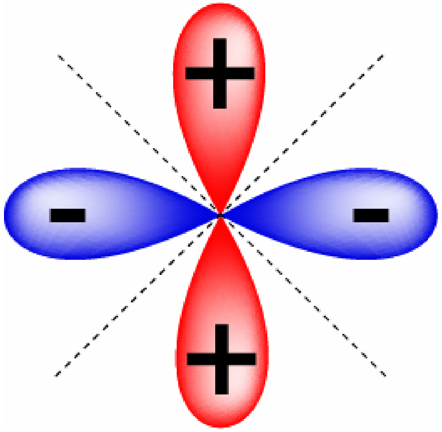
Z.-X. Shen,(1),(2) D. S. Dessau,(1),(2) B. O. Wells,(1),(2),(a) D. M. King,(2) W. E. Spicer,(2) A. J. Arko,(3) D. Marshall,(2) L. W. Lombardo,(1) A. Kapitulnik,(1) P. Dickinson,(1) S. Doniach,(1) J. DiCarlo,(1),(2) A. G. Loeser,(1),(2) and C. H. Park(1),(2)



“*s*-wave”

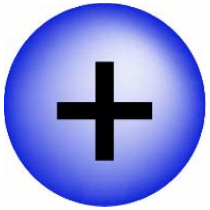


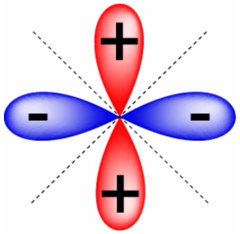
“*d*-wave”

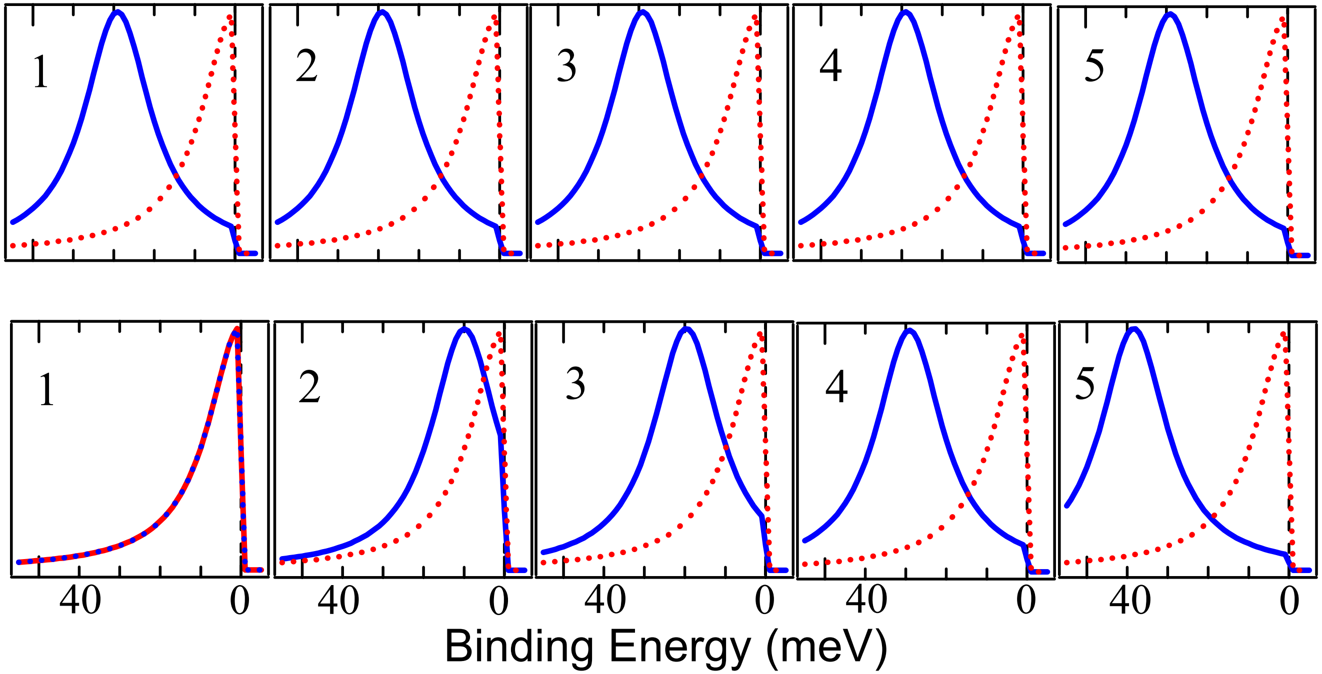


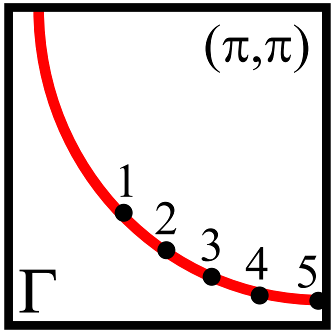
Courtesy of Kyle Shen

High-T_c Superconductors: s-wave vs. d-wave gap

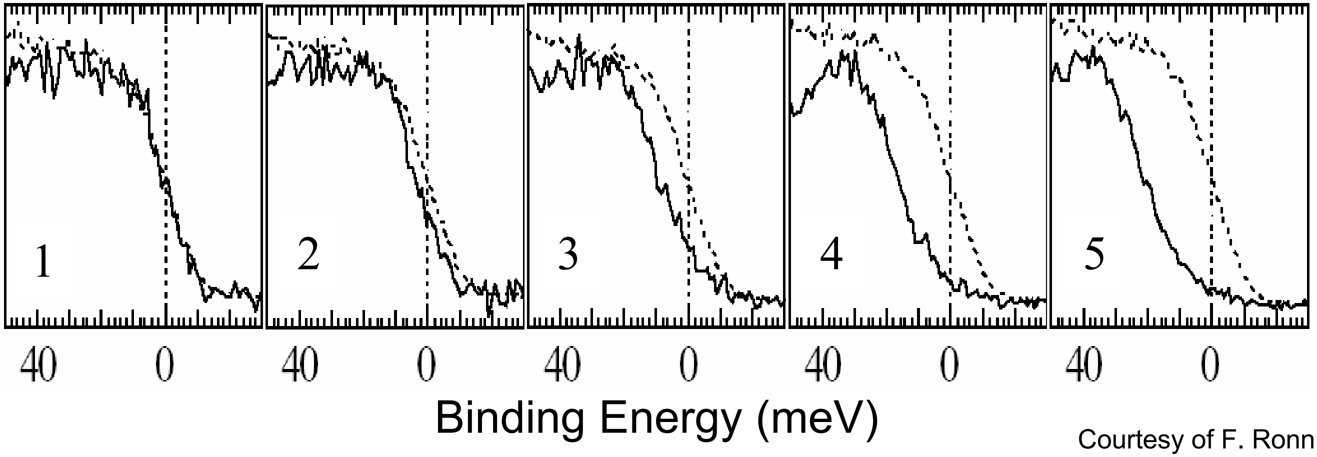






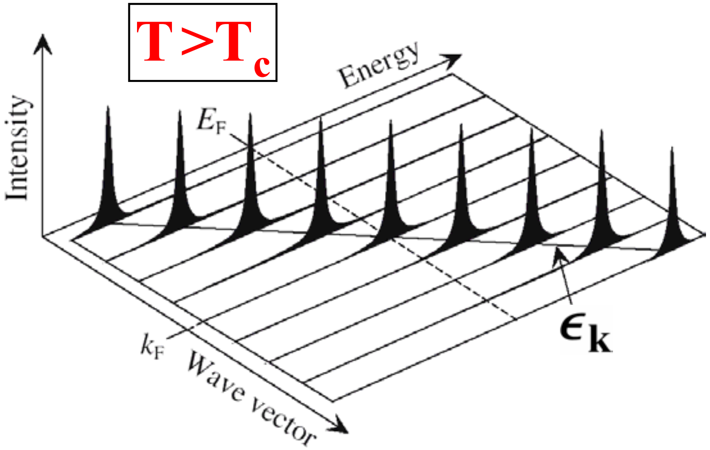


Ding et al. PRB **54**, R9678 (1996)

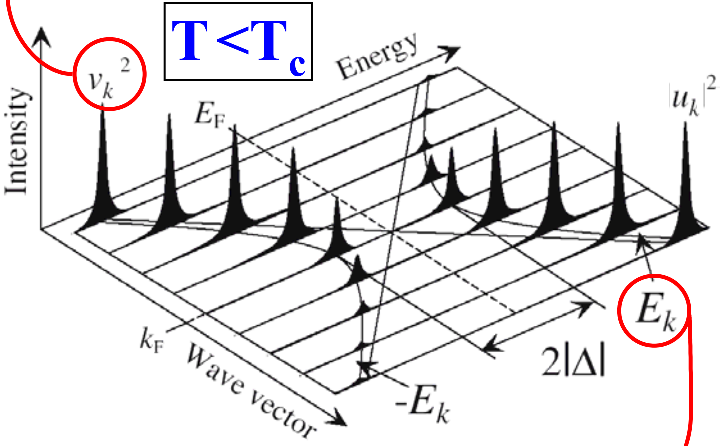


Courtesy of F. Ronning

High-T_c Superconductors: Bogoliubov QP in Bi2223

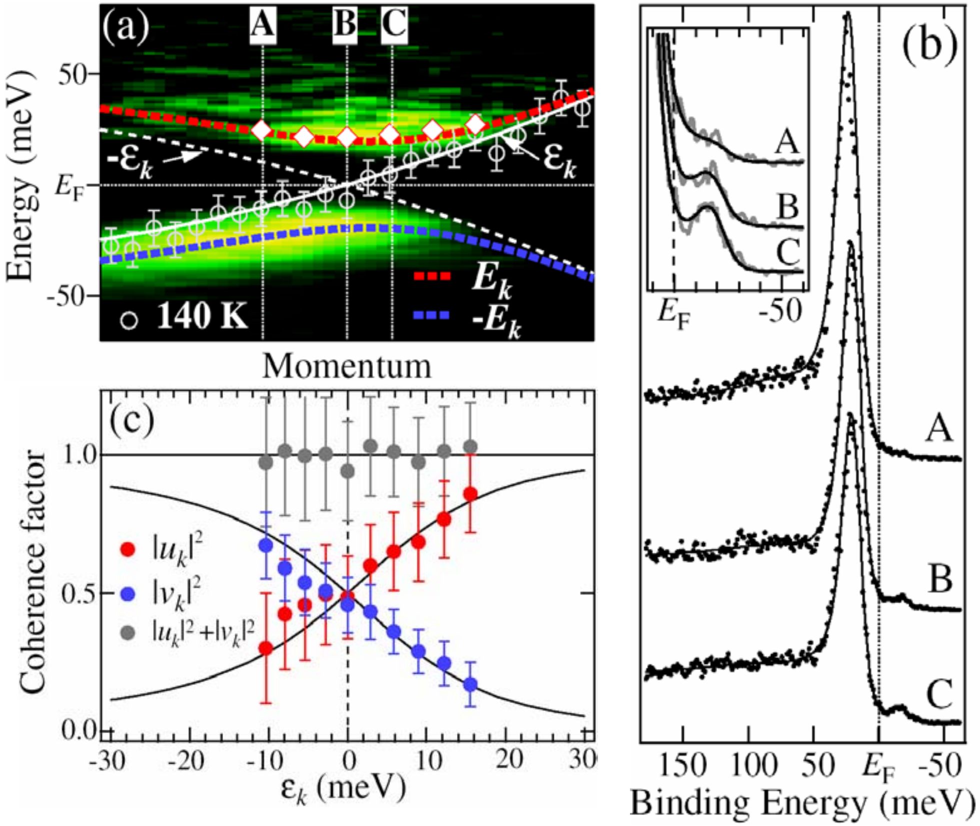


$$v_{\mathbf{k}}^2 = 1 - u_{\mathbf{k}}^2 = \tfrac{1}{2}\left(1 - \epsilon_{\mathbf{k}}/E_{\mathbf{k}}\right)$$



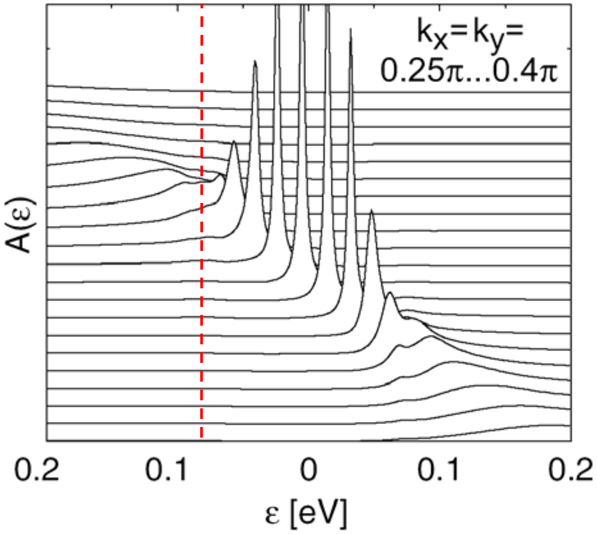
$$E_{\mathbf{k}} = \sqrt{\epsilon_{\mathbf{k}}^2 + |\Delta(\mathbf{k})|^2}$$

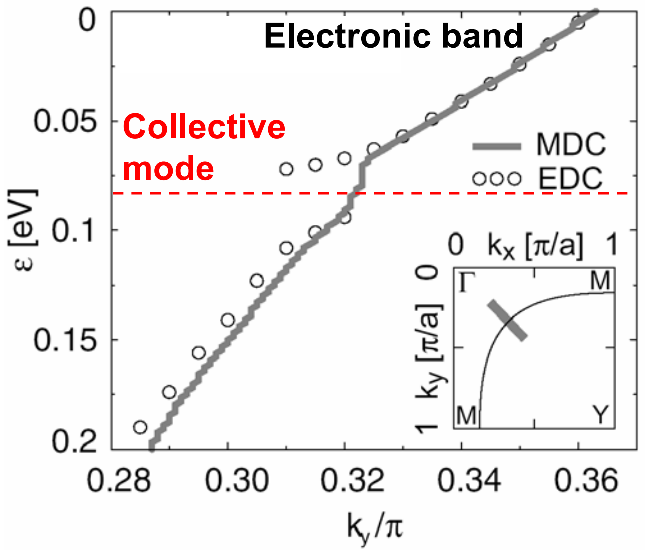
$$A_{\mathrm{BCS}}(k,\omega) = \frac{1}{\pi}\left\{\frac{|u_k|^2\Gamma}{(\omega - E_k)^2 + \Gamma^2} + \frac{|v_k|^2\Gamma}{(\omega + E_k)^2 + \Gamma^2}\right\}$$



Matsui *et al.*, PRL **90**, 217002 (2003)

Many-Body effects: Electron-Phonon Coupling



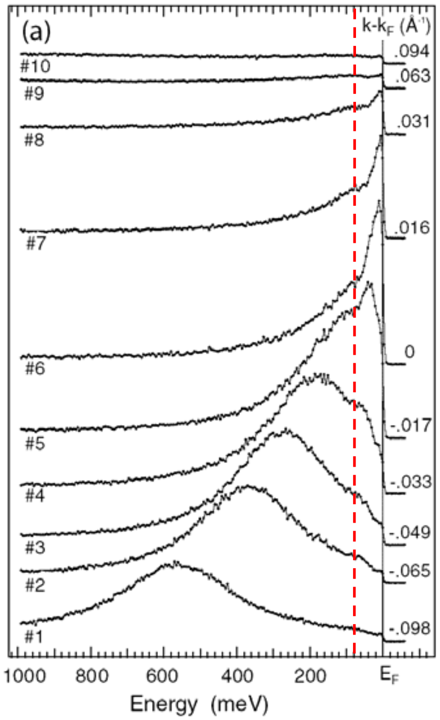


Eschrig, Norman, PRB **67**, 144503 (2003)

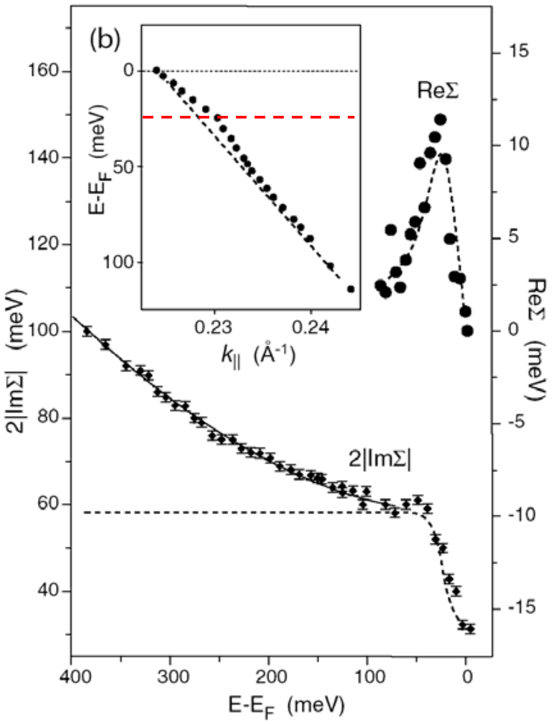
Single-particle spectral function

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

Be(0001)



Mo(110)

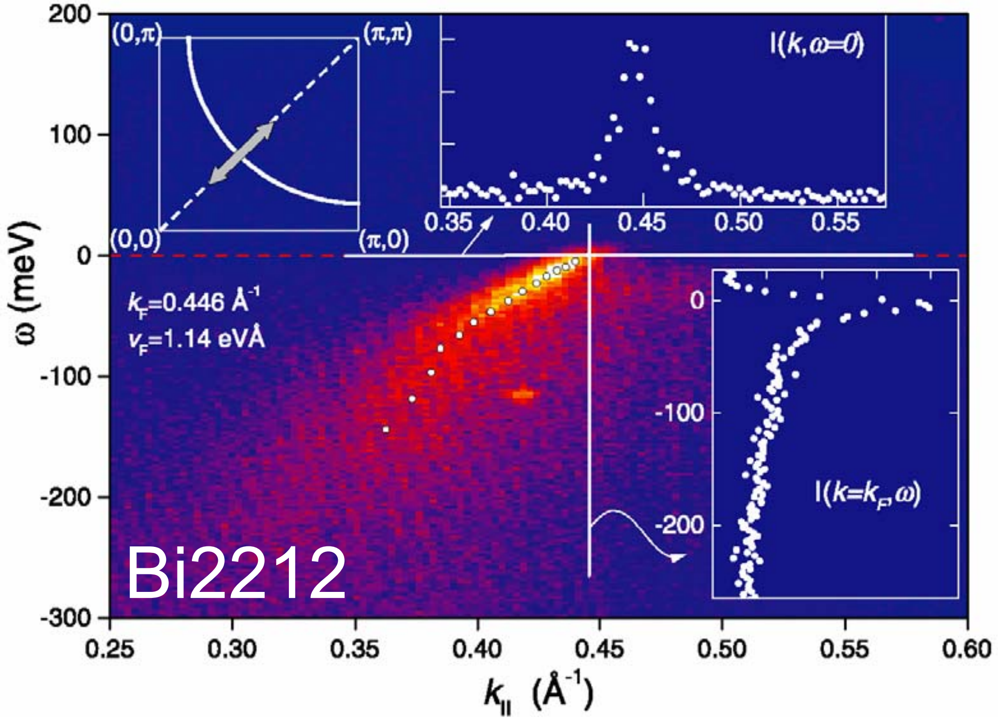


Hengsberger *et al.*, PRL **83**, 592 (1999)

Valla *et al.,* PRL **83**, 2085 (1999)

Many-Body effects in the High-T_C Cuprates

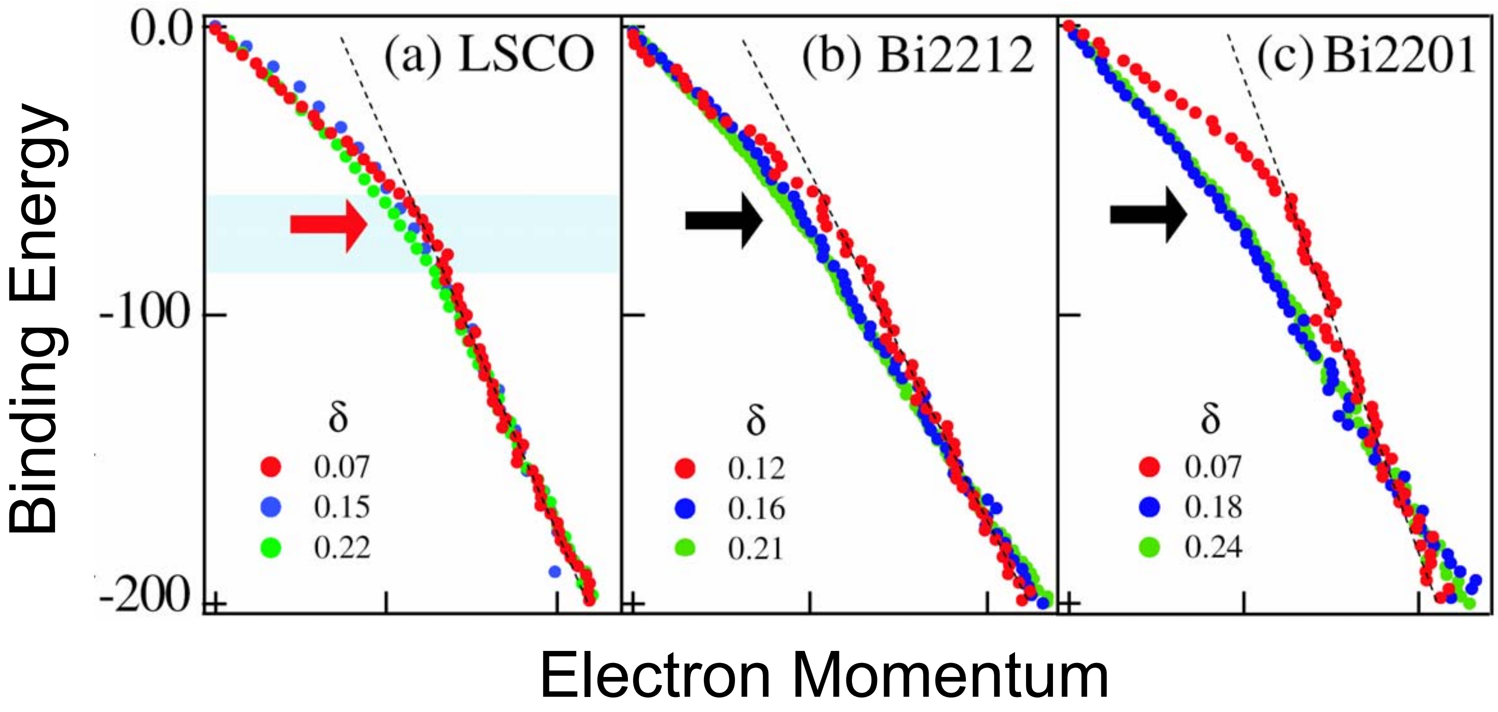
Valla *et al.*, Science **285**, 2110 (1999)



Mechanism for High-T_C { **Magnetic fluctuations ?**
Electron-phonon coupling ?

Many Body effects in the Quasiparticle Dispersion

Lanzara *et al*., Nature **412**, 510



Mechanism for High-T_c { **Magnetic fluctuations ?**
Electron-phonon coupling ?

Conclusions

ARPES results from complex systems

- **Bands and FS in unprecedented detail**
- **Fermi velocity and effective mass**
- **Superconducting (d-wave) gap**
- **Many-body effects in the QP dispersion**
- **Nanostructured materials (surface FM)**

ARPES is a powerful tool for the study of the electronic structure of complex systems

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For a review article see:

A. Damascelli, Z. Hussain, and Z.-X Shen, Rev. Mod. Phys. **75**, 473 (2003)

For additional material see:

http://www.physics.ubc.ca/~damascel/