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Andrea Damascelli

UBC-MPI Quantum Matter Institute

ARPES on Correlated Electron Systems

CUSO Lecture – Lausanne 02/2011

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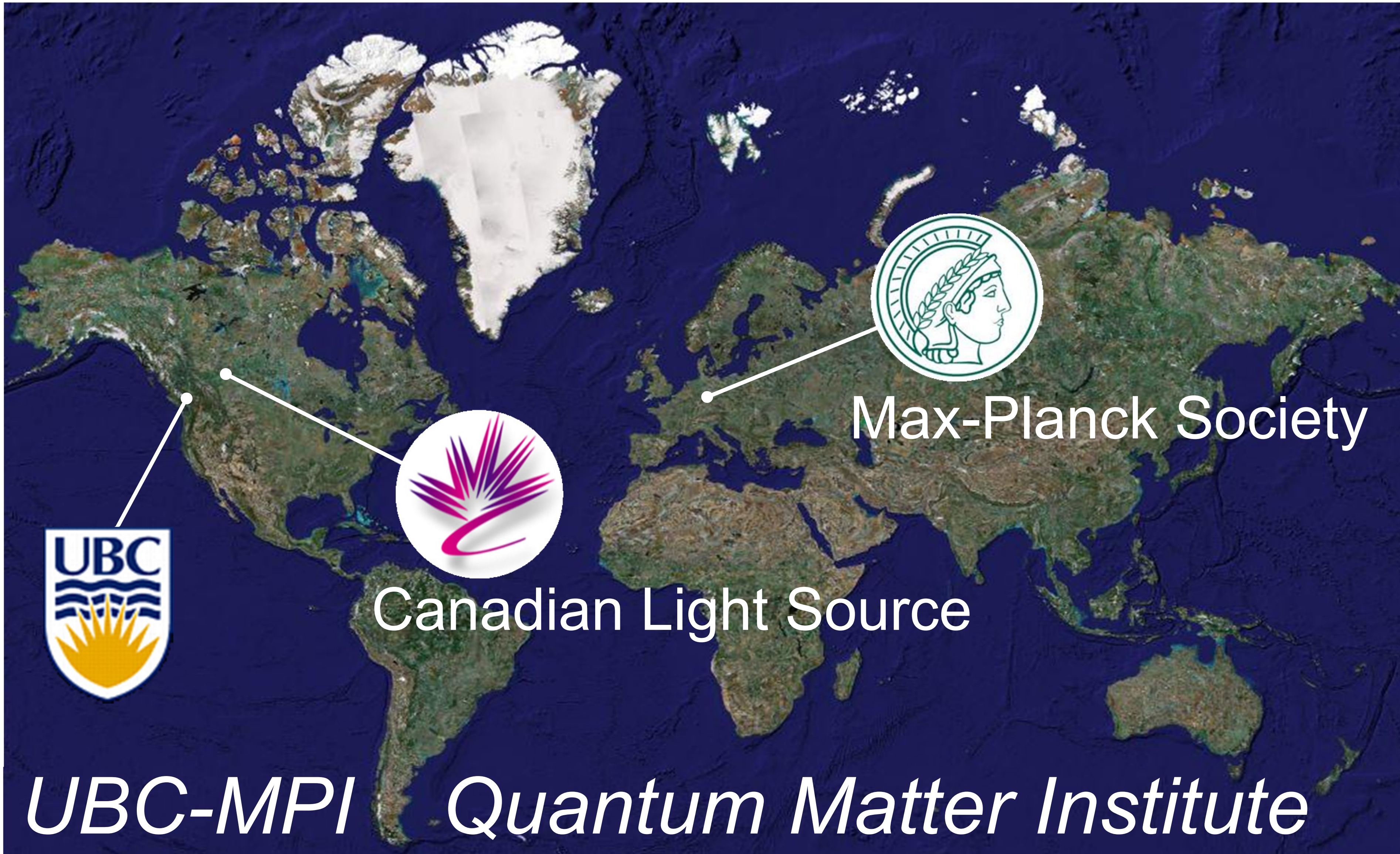

Outline Part I

- Introduction: Transition metal oxides
- Birth and history of photoemission
- ARPES: Fundamentals and spectral function
- ARPES: Technique and developments
- Bulk, surface, and Fermi surfaces: Sr_2RuO_4
- Superconducting gap: BCS and HTSC

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http://www.physics.ubc.ca/~quantmat/ARPES/RESEARCH/ARPES_intro.html

ARPES ON COMPLEX SYSTEMS

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Electronic and Magnetic Interactions in Novel Complex Systems

Angle-resolved photoemission spectroscopy (ARPES) is one of the most direct methods of studying the electronic structure of solids and is the only truly momentum-resolved probe, which is essential for the investigation of low dimensional and strongly anisotropic systems. By measuring momentum and kinetic energy of the electrons photoemitted from a sample illuminated with radiation of energy larger than the material work function, it is possible to gain information on both energy and the momentum of the electronic excitations inside the solid.

As the intensity measured in photoemission experiments is proportional to the single-particle spectral function $A(\mathbf{k},\omega)=-(1/\pi)\mathrm{Im}G(\mathbf{k},\omega)$, ARPES provides direct insights on the Green's function $G(\mathbf{k},\omega)$ which describes the propagation of an electron in a many-body system. This is of vital importance in elucidating the connection between electronic, magnetic, and chemical structure of solids, in particular for those complex systems which cannot be described within the independent-particle picture.

The last decade witnessed significant progress in this technique and its applications, thus ushering in a new era in photoelectron spectroscopy. Today, ARPES experiments with 2 meV energy resolution and 0.2 degree angular resolution are a reality even for photoemission on solids, providing detailed information on band dispersion and Fermi surface, as well as on the strength and nature of those many-body correlations which may profoundly affect the one-electron excitation spectrum and, in turn, determine the macroscopic physical properties.

The Photoelectric Effect

Einstein's Photoelectric Effect

From Light Quanta to Quantum Phenomena in Solids

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ARPES: Introduction

ARPES on HTSC's

Reviews of Modern Physics

ARPES: Viewgraphs

Probing the Electronic Structure of Complex Systems by State-of-the-Art ARPES

Andrea Damascelli

http://www.physics.ubc.ca/~quantmat/ARPES/RESEARCH/ARPES_intro.html

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Probing the Electronic Structure of Complex Systems by ARPES

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Abstract

Angle-resolved photoemission spectroscopy (ARPES) is one of the most direct methods of studying the electronic structure of solids. By measuring the kinetic energy and angular distribution of the electrons photoemitted from a sample illuminated with sufficiently high-energy radiation, one can gain information on both the energy and momentum of the electrons propagating inside a material. This is of vital importance in elucidating the connection between electronic, magnetic, and chemical structure of solids, in particular for those complex systems which cannot be appropriately described within the independent-particle picture. The last decade witnessed significant progress in this technique and its applications, thus ushering in a new era in photoelectron spectroscopy; today, ARPES experiments with 2 meV energy resolution and 0.2° angular resolution are a reality even for photoemission on solids. In this paper we will review the fundamentals of the technique and present some illustrative experimental results; we will show how ARPES can probe the momentum-dependent electronic structure of solids providing detailed information on band dispersion and Fermi surface as well as on the strength and nature of many-body correlations, which may profoundly affect the one-electron excitation spectrum and in turn the macroscopic physical properties.

photoemission event is decomposed in three independent steps: optical excitation between the initial and final *bulk* Bloch eigenstates, *travel* of the excited electron to the surface, and escape of the photoelectron into vacuum after transmission through the *surface* potential barrier. This is the most common approach, in particular when photoemission spectroscopy is used as a tool to map the electronic band structure of solids. However, from the quantum-mechanical point of view photoemission should not be described in terms of several independent events but rather as a *one-step* process (Fig. 1(b)): in terms of an optical transition (with probability given by Eq. (12)) between initial and final states consisting of many-body wave functions that obey appropriate boundary conditions at the surface of the solid. In particular (see Fig. 2), the initial state should be one of the possible N-electron eigenstates of the semi-infinite crystal, and the final state must be one of the

Outline Part I

Introduction:

Transition metal oxides

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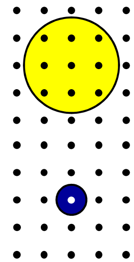
Strongly Correlated Electron Systems

d - f
open shells materials

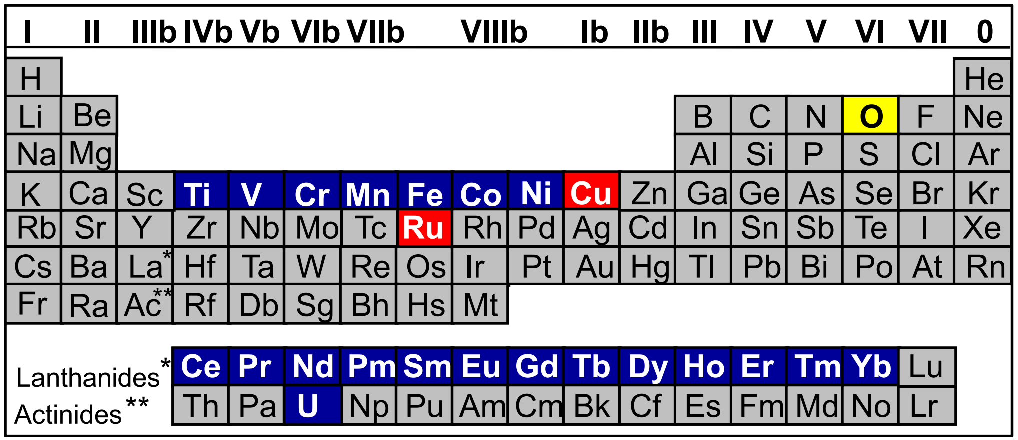


U<<W
Charge fluctuations

U>>W
Spin fluctuations

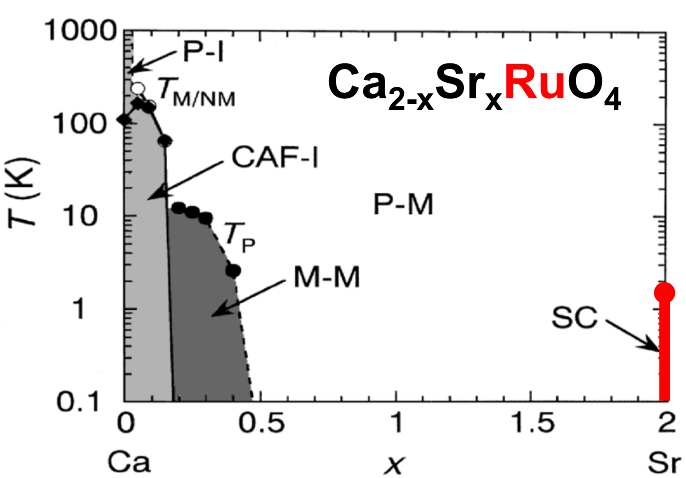
Control parameters

Bandwidth (U/W)
Band filling
Dimensionality

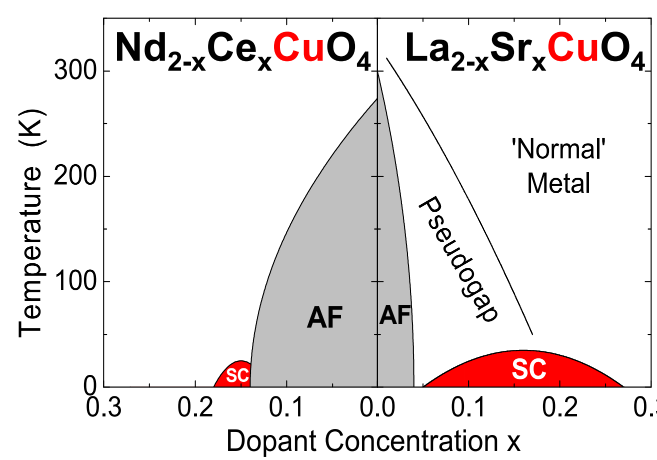


Degrees of freedom

Charge / Spin
Orbital
Lattice

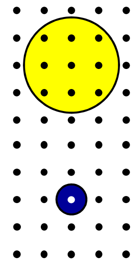


- **Kondo**
- **Mott-Hubbard**
- **Heavy Fermions**
- **Unconventional SC**
- **Spin-charge order**
- **Colossal MR**



Strongly Correlated Electron Systems

d - f
open shells materials

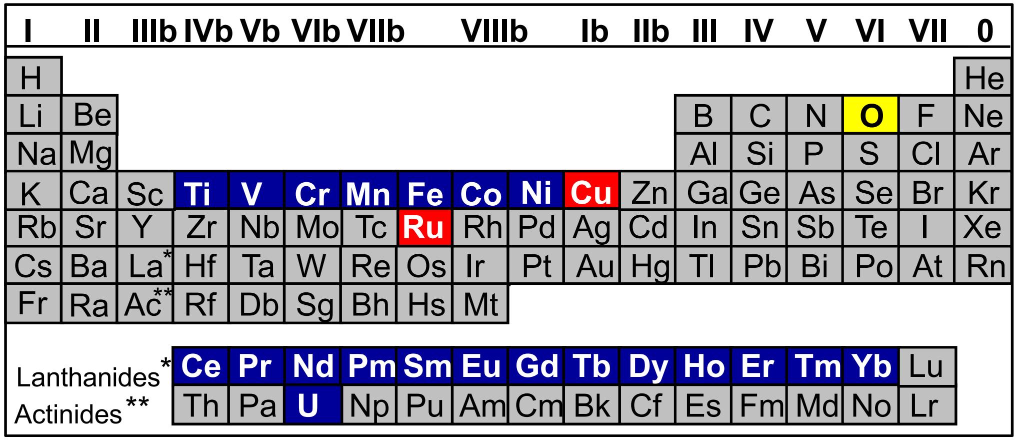


U<<W
Charge fluctuations

U>>W
Spin fluctuations

Control parameters

Bandwidth (U/W)
Band filling
Dimensionality



Degrees of freedom

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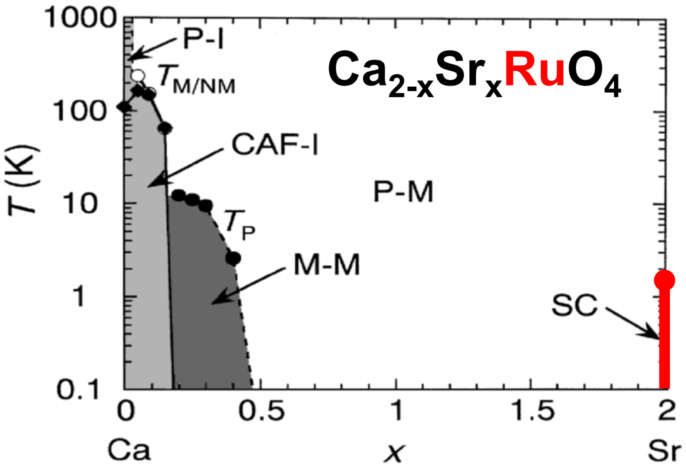
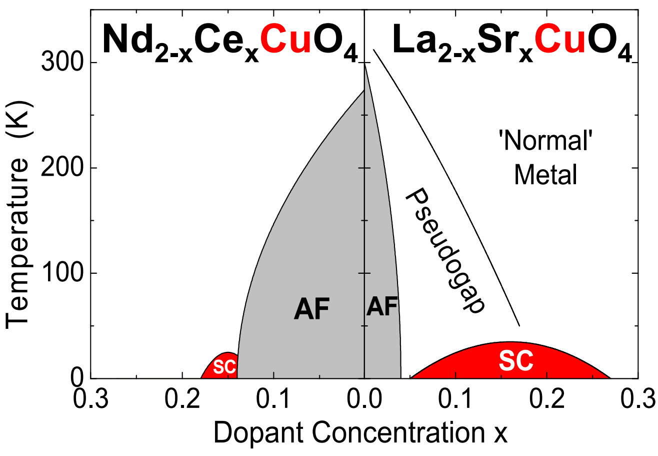


d-orbitals radial extent

↔

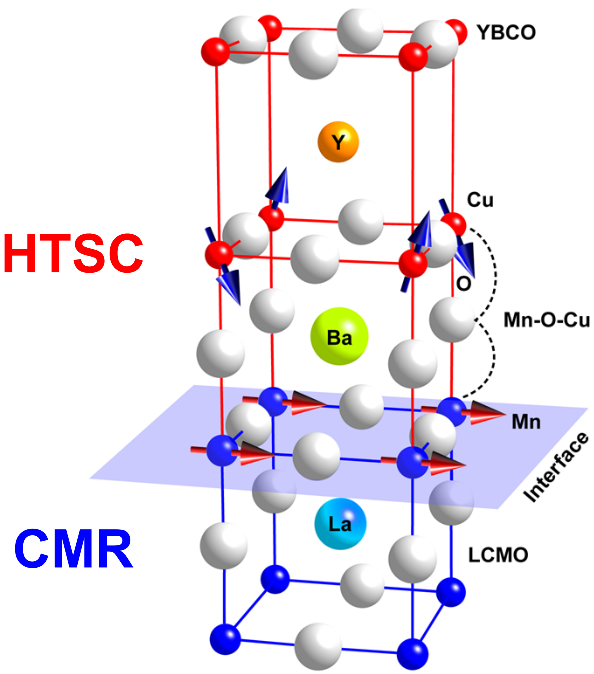
Spin-orbit coupling



Novel Complex Materials and Functionalities

Tune the physical properties

Chakhalian et al., Nature Physics 2006



- Modern synthesis methods

  Single crystals, multilayers, nanostructures

- Sophisticated structural tools

  Physical, chemical, and magnetic structures

- Novel probes of intrinsic susceptibilities

  Lattice, magnetic, and electronic excitations

$$\varepsilon(q,q',\omega) \quad \chi(q,q',\omega)$$
$$N(\vec{r},E) \quad A(\vec{k},E)$$

Interface-tuned magnetism in oxide multilayers ⟷ Understand interplay of **lattice**, **spin**, **charge**, **orbital** degrees of freedom

Outline Part I

Birth and history of photoemission

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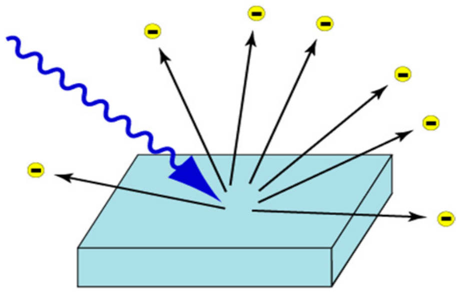
Probing Electrons in Reciprocal Space



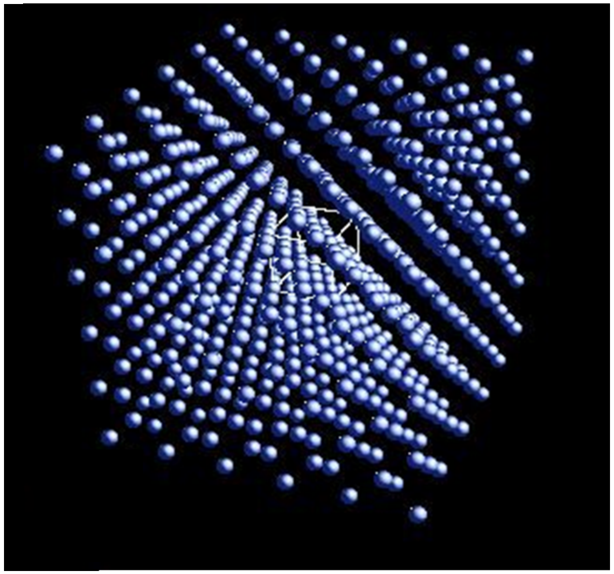
Angle Resolved PhotoElectron Spectroscopy

FIRST EVIDENCE FOR THE QUANTIZATION OF LIGHT!

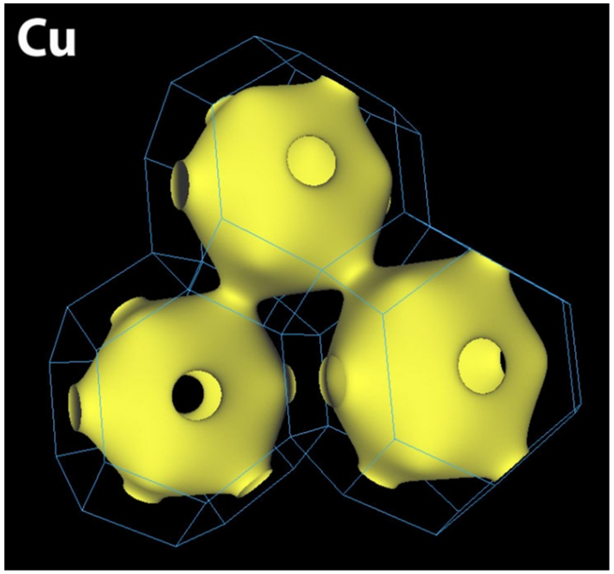
Velocity and direction of the electrons in the solid

Low-energy Electronic Structure → Macroscopic Physical Properties

Superconductivity, *Magnetism*, *Density Waves*, ...



X-ray diffraction



Photoemission

Interaction Effects between Electrons : “Many-body Physics”

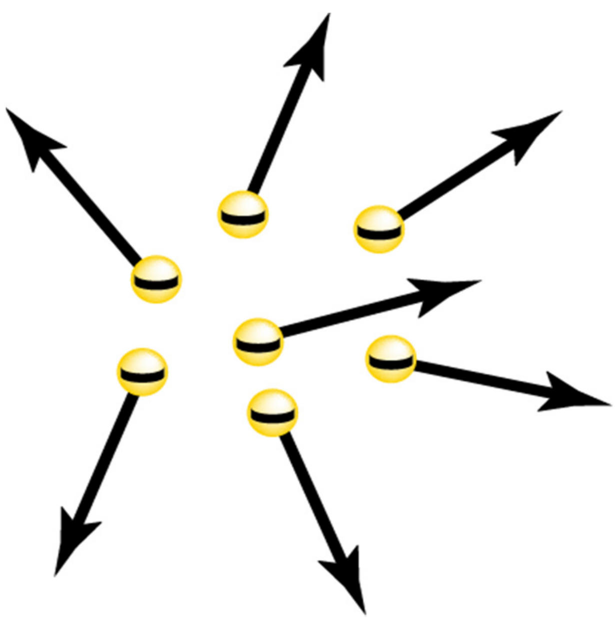
Many-body effects are due to the interactions between the electrons and each other, or with other excitations inside the crystal :

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

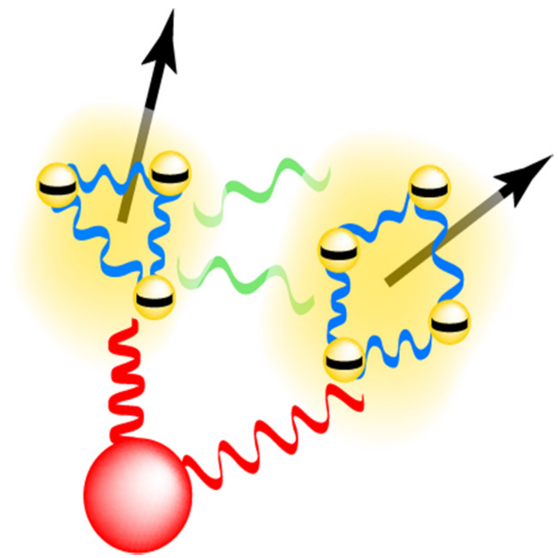
2) Responsible for many surprising phenomena :

Superconductivity, Magnetism, Density Waves, ...

Non-Interacting



Interacting



Courtesy of Kyle Shen

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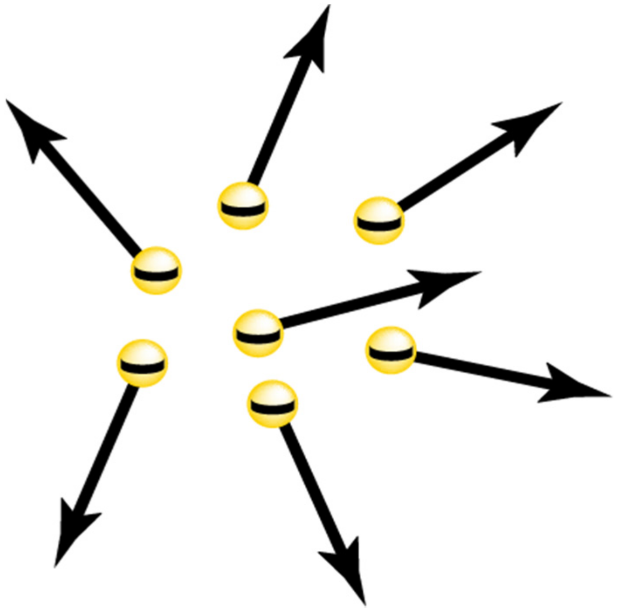
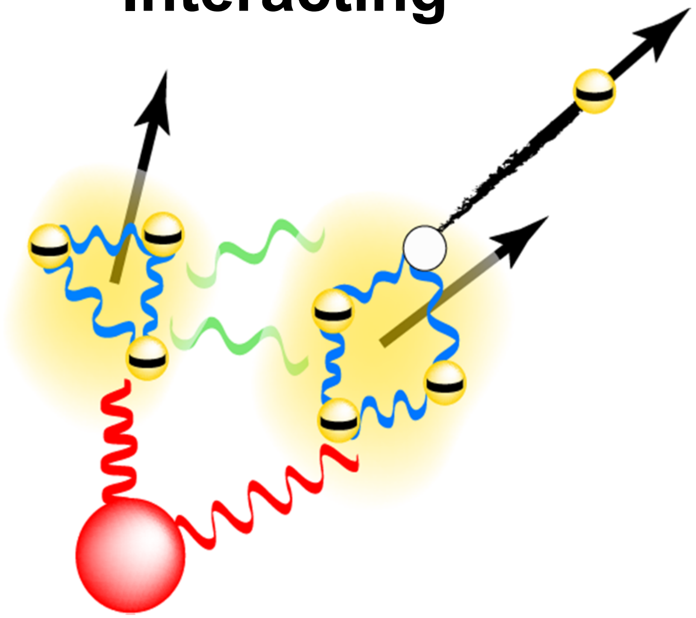
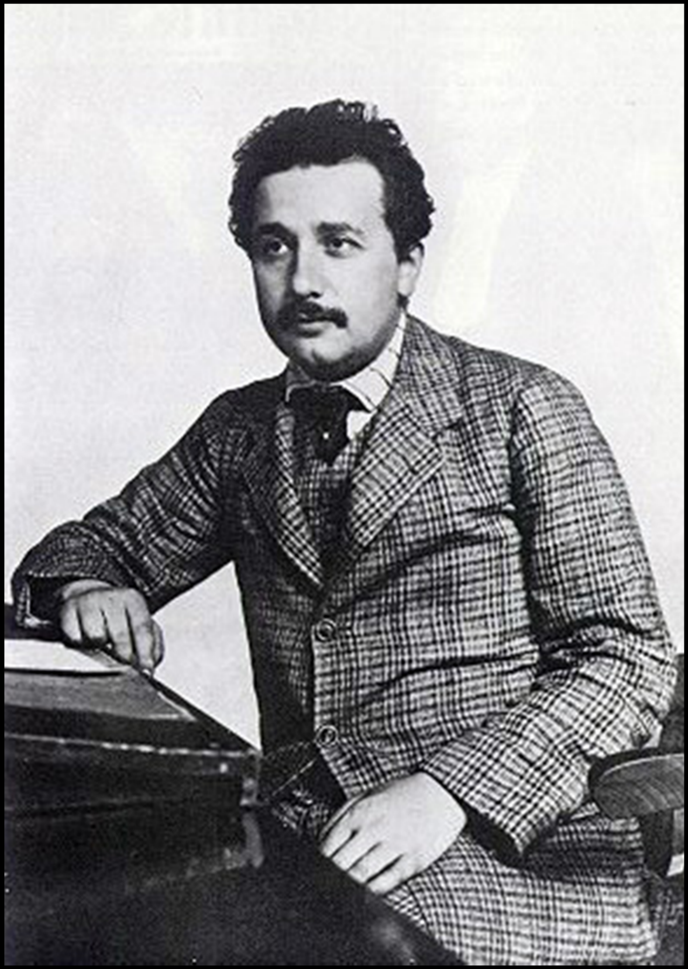


Interacting



Courtesy of Kyle Shen

Einstein's Annus Mirabilis: 1905



- **The Brownian motion**

  "On the motion of small particles suspended in liquids atrest required by the molecular-kinetic theory of heat."
  Annalen der Physik, **17** (1905), pp. 549-560.

- **The photoelectric effect**

  "On a heuristic viewpoint concerning the production and transformation of light"
  Annalen der Physik, **17** (1905), pp. 132-148.

- **The special theory of relativity**

  "On the electrodynamics of moving bodies"
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- **Mass-energy Equivalency $E=mc^2$**

  "Does the inertia of a body depend on its energy?"
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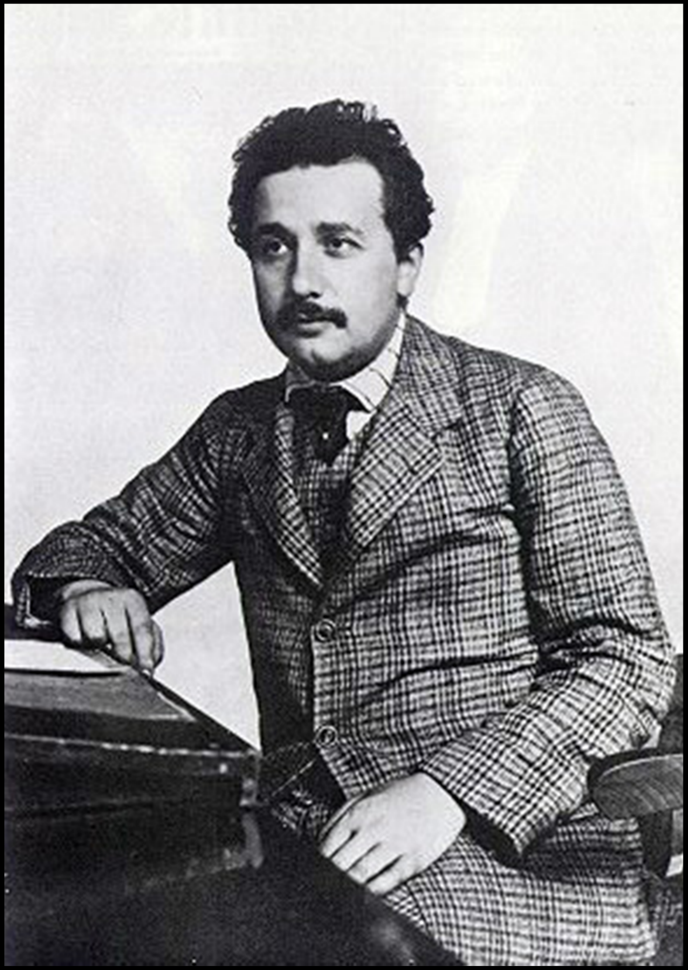
Einstein's Annus Mirabilis: 1905



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1921

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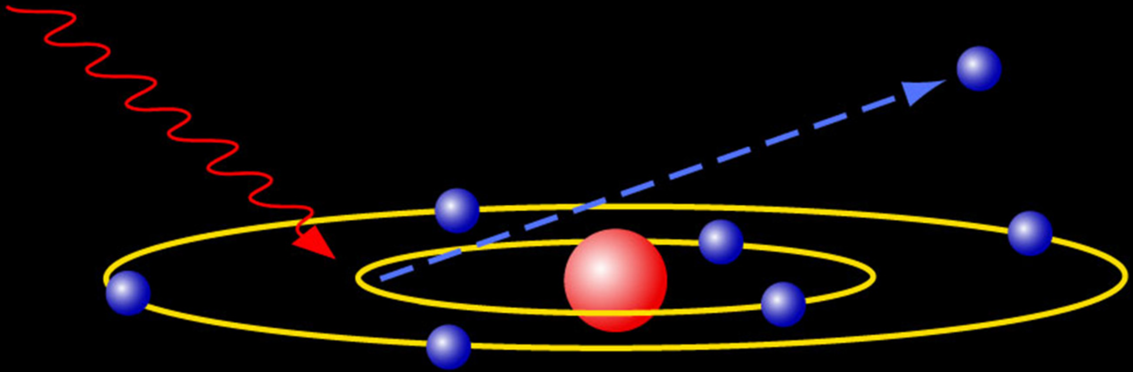
The Photoelectric Effect: Intro

Emission of an electron due to the absorption of light



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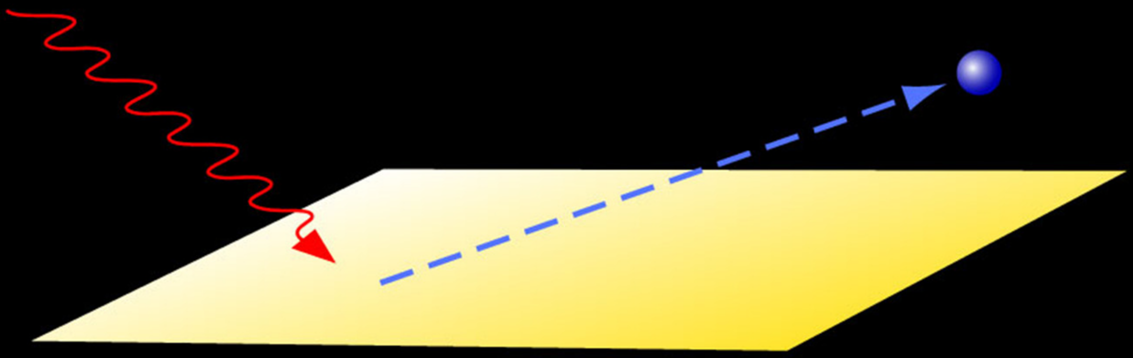
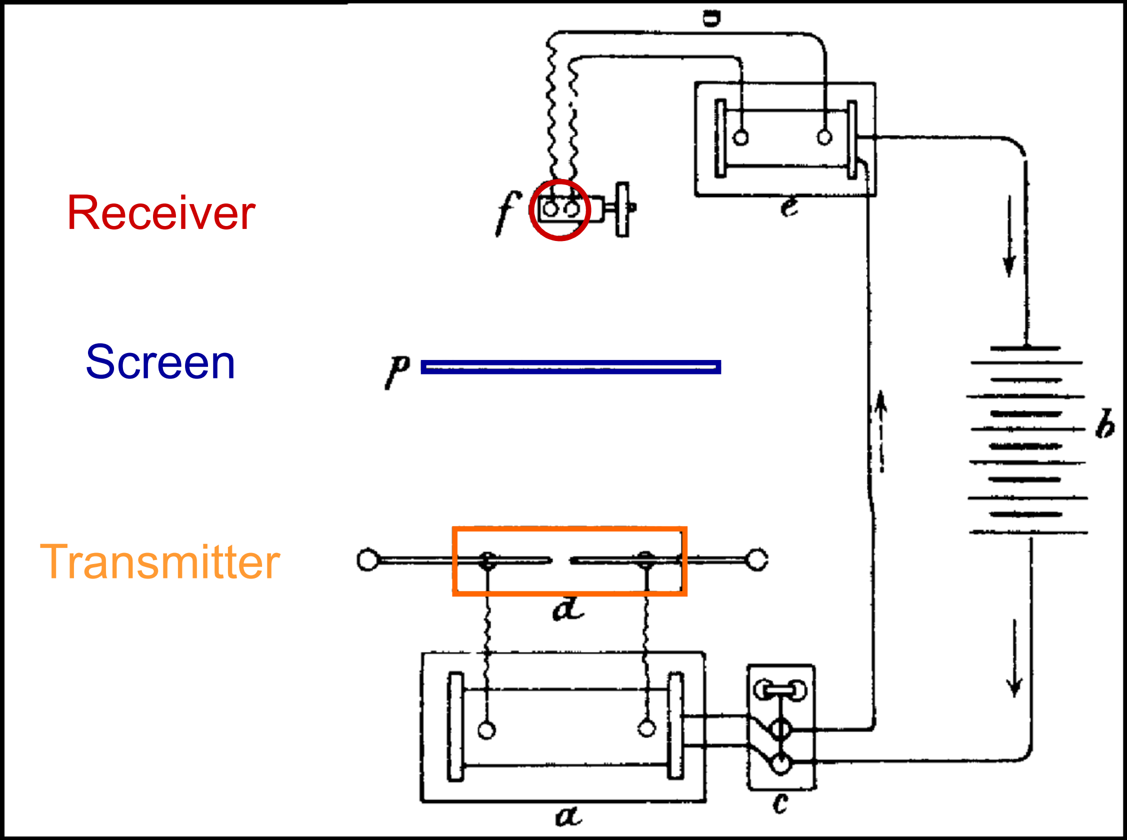


First experimental evidence
for the quantization of light

The Photoelectric Effect: History

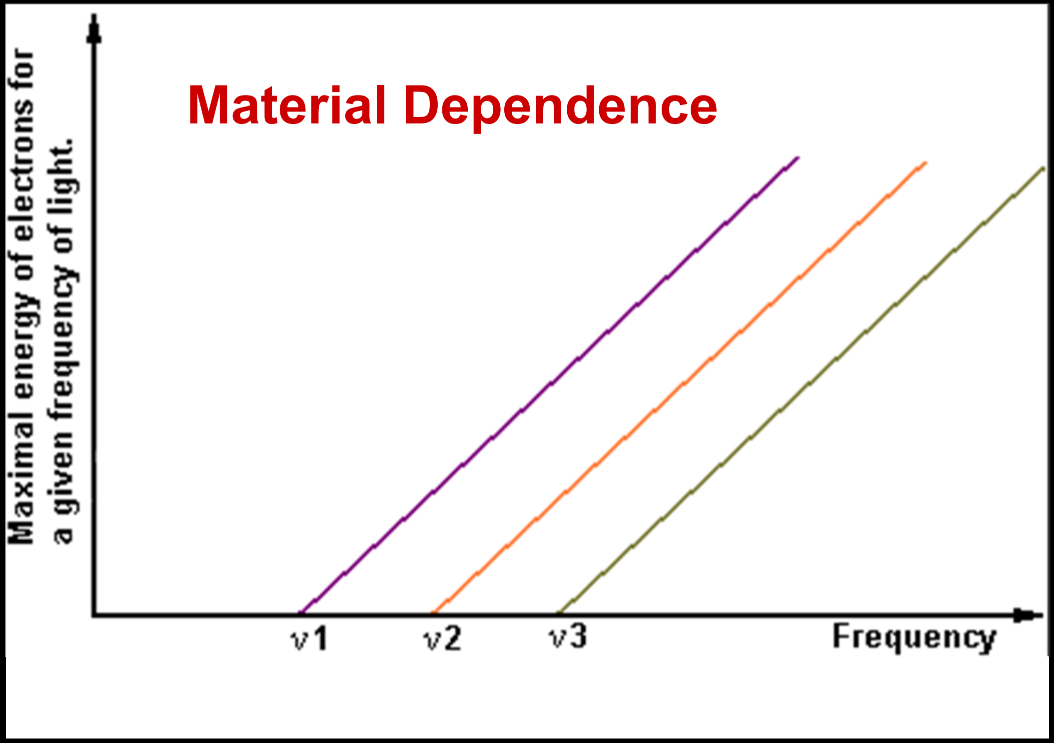
1887 Hertz finds Maxwell's waves; and something else



The small RECEIVER SPARK was more vigorous when the receiver was exposed to the ultraviolet light form the TRANSMITTER SPARK

The Photoelectric Effect: History

1902 von Lenard varies the intensity and color of the light

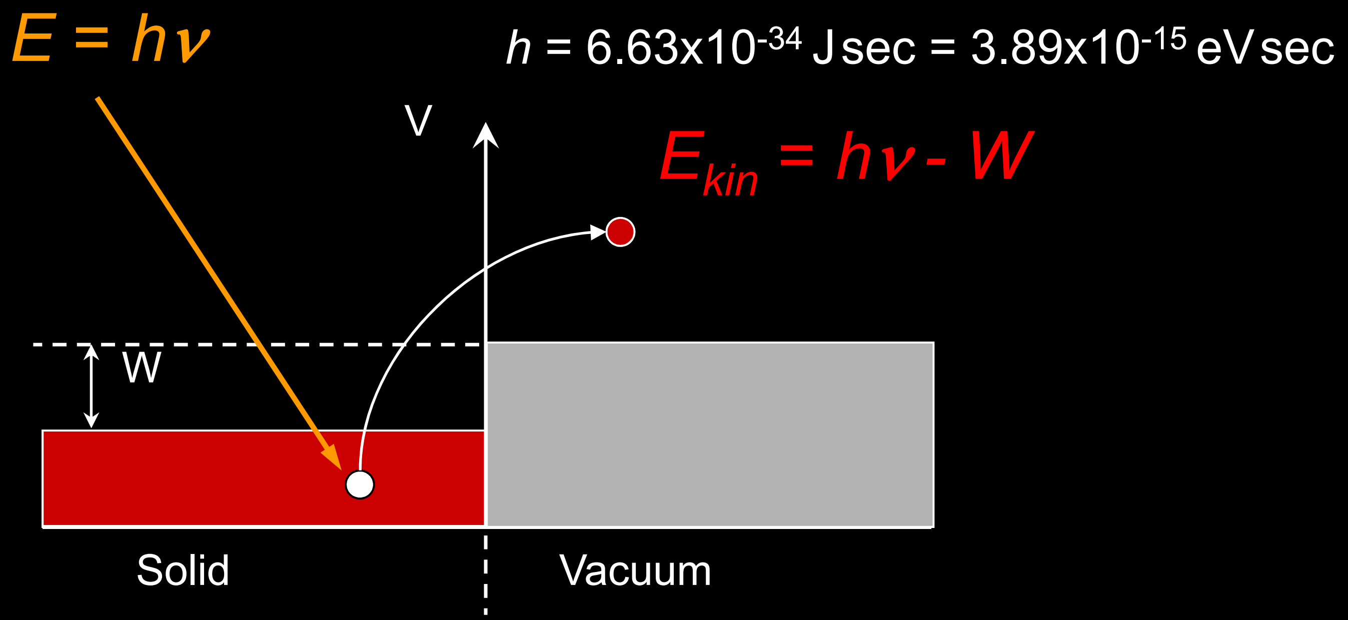


The NUMBER of electrons is proportional to the INTENSITY

The maximum $E_{kin}=\frac{1}{2}mv^2$ is proportional to the FREQUENCY

The Photoelectric Effect: History

1905 Einstein' hypothesis: light quanta with $E = h\nu = hc / \lambda$



The maximum $E_{kin} = \frac{1}{2} mv^2$ is proportional to the FREQUENCY but depends also on the material work function W

The NUMBER of electrons is proportional only to the INTENSITY

The Photoelectric Effect: History

1887 Heinrich Hertz

1897 Joseph Thomson: "for the theoretical and experimental investigations on the conduction of electricity by gases"

1906

1888 Wilhelm Hallwachs

1902 Philipp von Lenard: "for his work on cathode rays"

1905

1905 Albert Einstein: "for his services to Theoretical Physics, and …. for his discovery of the law of the photoelectric effect"

1921

1916 Robert Millikan: "for his work on the elementary charge of electricity and on the photoelectric effect"

1923

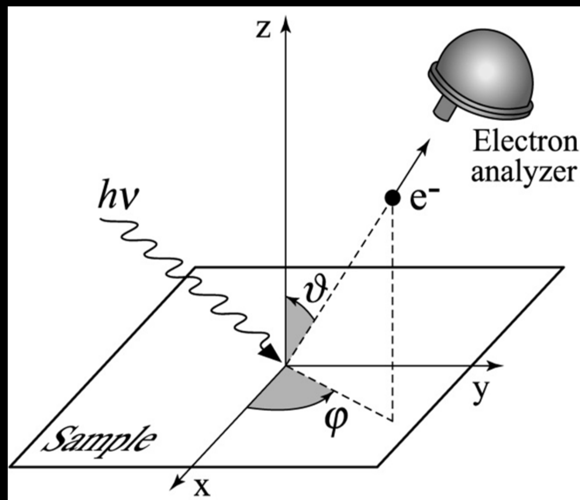
Einstein's hypothesis is too revolutionary

In 1913 Einstein was elected to the Prussian Academy of Sciences and appointed to a research position in Berlin. In his nomination speech to the Prussian Academy, Planck says:

"Summing up, we may say that there is hardly one among the great problems in which modern physics is so rich, to which Einstein has not made an important contribution."

"That he may sometimes have missed the target in his speculations, as for example, in his hypothesis of light quanta, cannot really be held too much against him, for it is not possible to introduce fundamentally new ideas, even in the most exact sciences, without occasionally taking a risk".

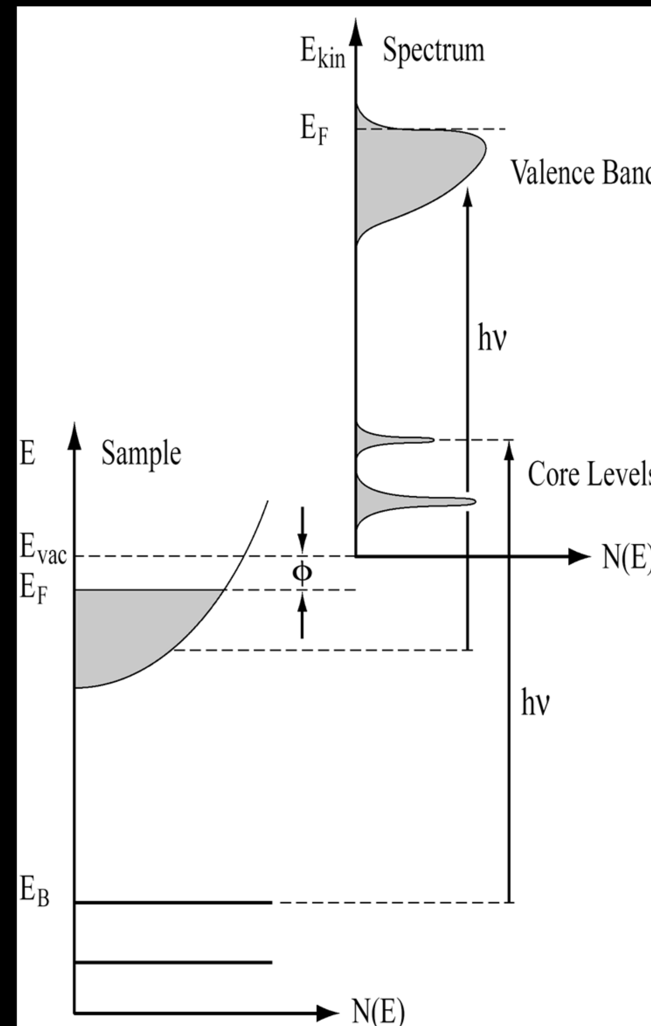
Scientific application: Spectroscopy



Electron Spectroscopy for Chemical Analysis (ESCA)

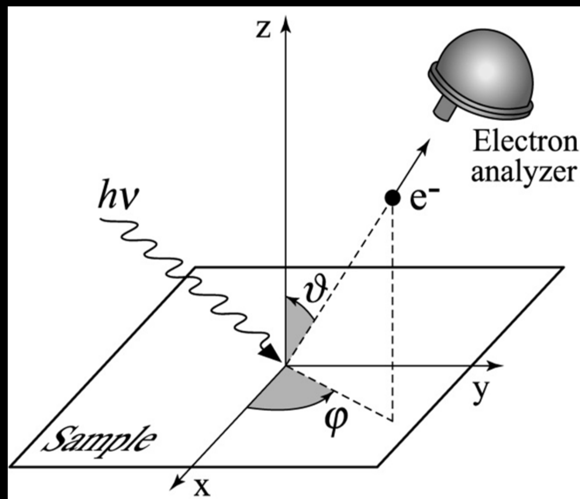
Kai Siegbahn:  1981

"for his contribution to the development of high-resolution electron spectroscopy"



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

Scientific application: Spectroscopy



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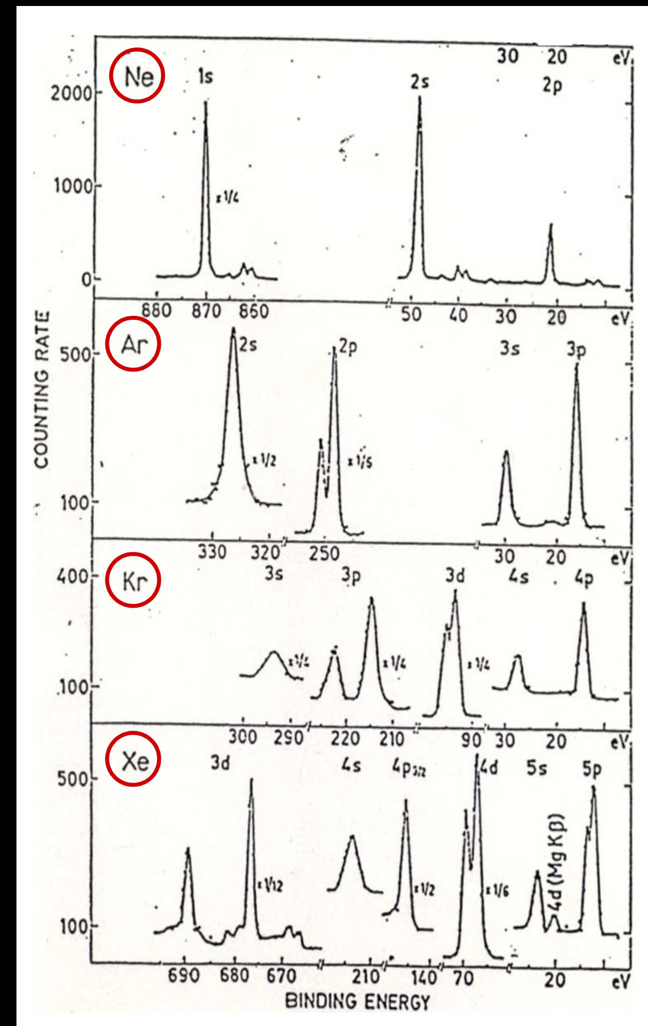
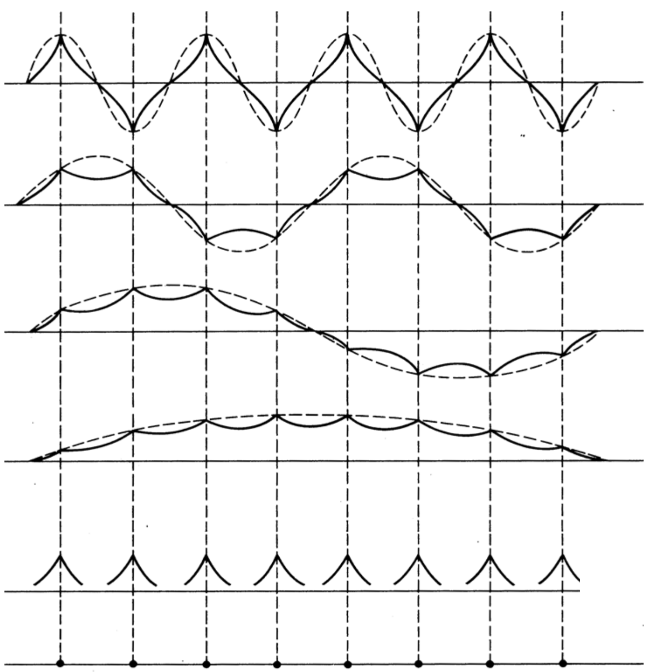


$$E_{kin} = h\nu - \phi - |E_B|$$

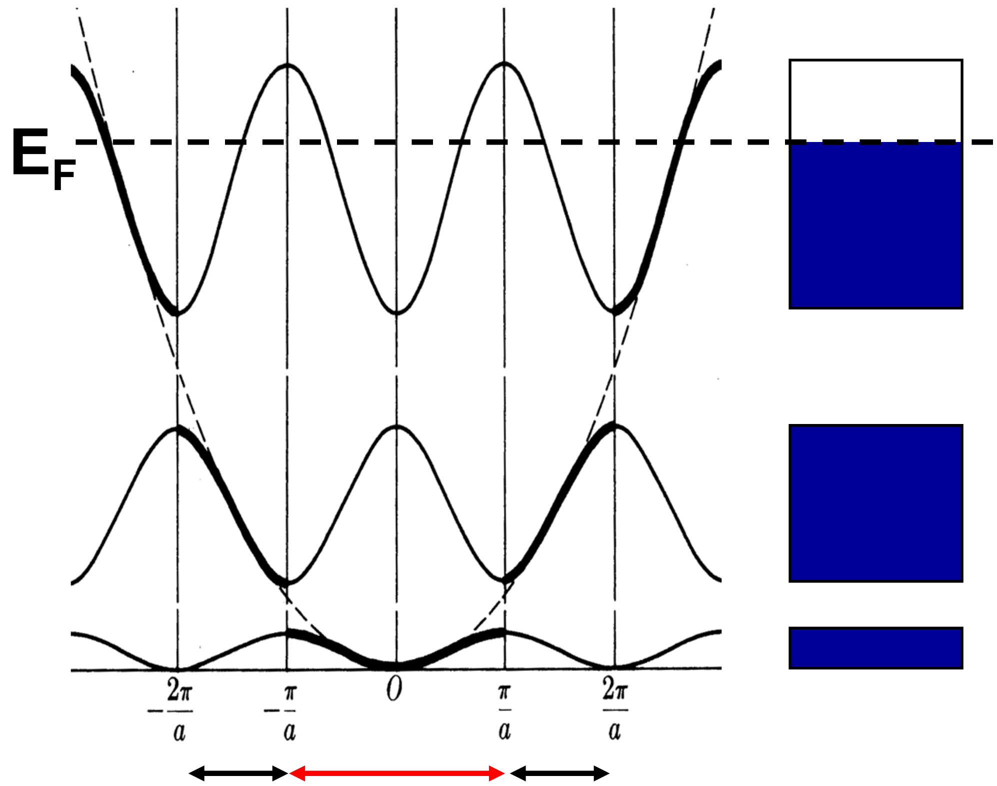
Solid State: Electrons in Reciprocal Space

Wave functions
in a 1D lattice



1D chain of atoms

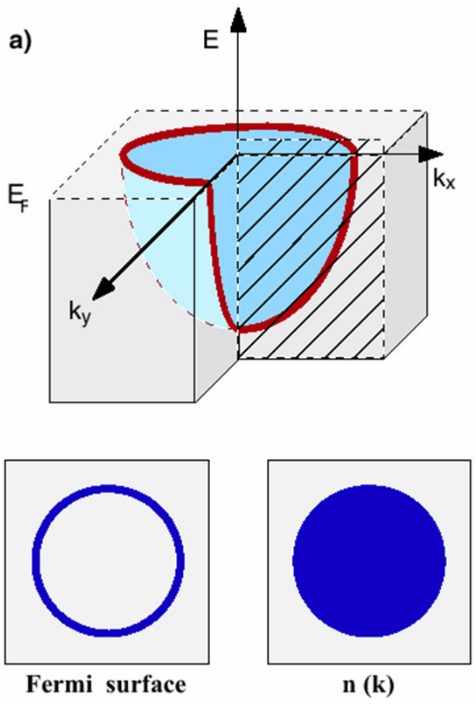
Allowed electronic states
Repeated-zone scheme



Second Brillouin zone | **First Brillouin zone** | **Second Brillouin zone**

Solid State: Electrons in Reciprocal Space

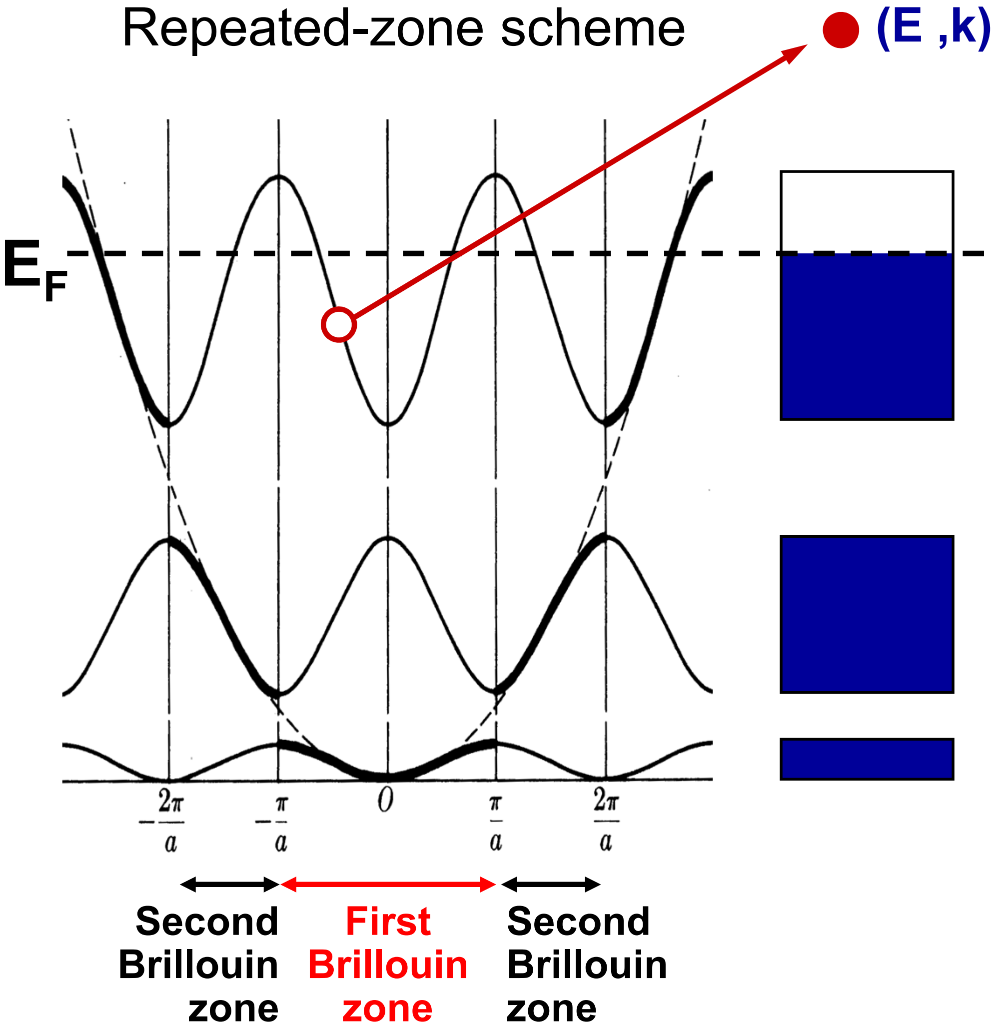
Many properties of a solids are determined by electrons near E_F (conductivity, magnetoresistance, superconductivity, magnetism)



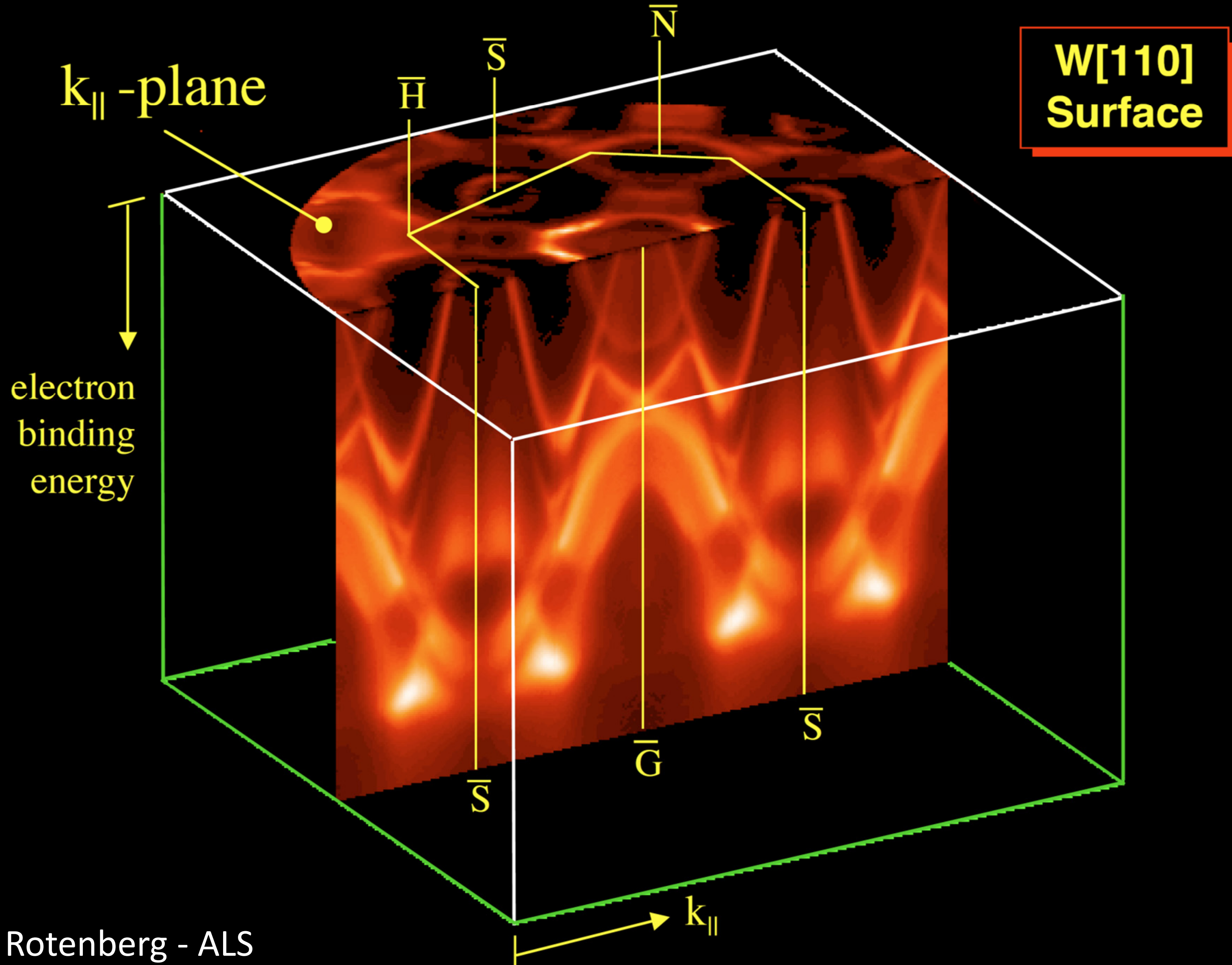
Only a narrow energy slice around E_F is relevant for these properties (kT=25 meV at room temperature)

Allowed electronic states

Repeated-zone scheme



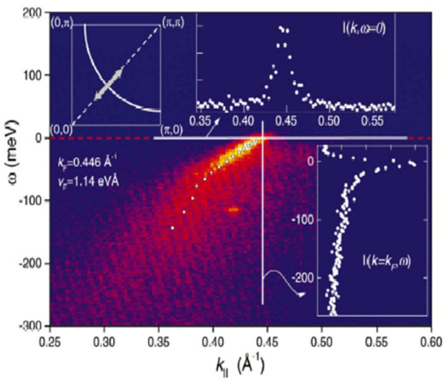
Band Mapping and Fermi Contours



Courtesy of Eli Rotenberg - ALS

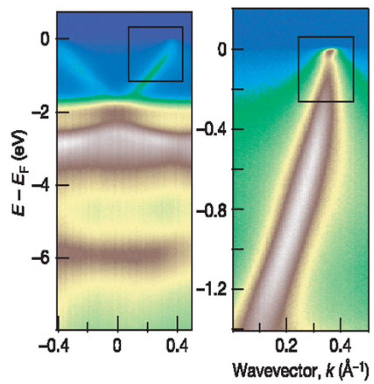
ARPES: Widespread Impact in Complex Materials

HTSC's



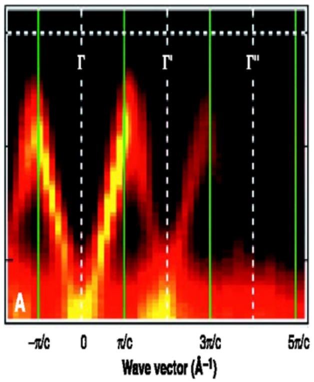
Science 1999

CMR's



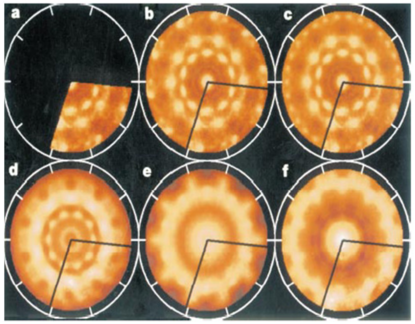
Nature 2005

CDW's



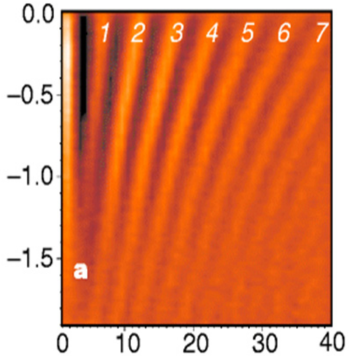
Science 2000

Quasicrystals



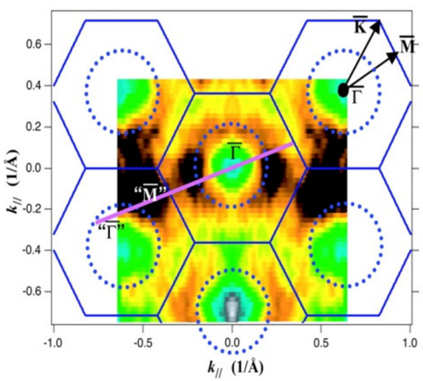
Nature 2000

Quantum Wells



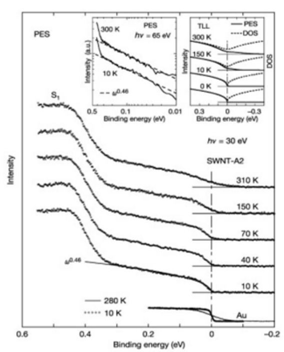
Nature 1999

C_{60}



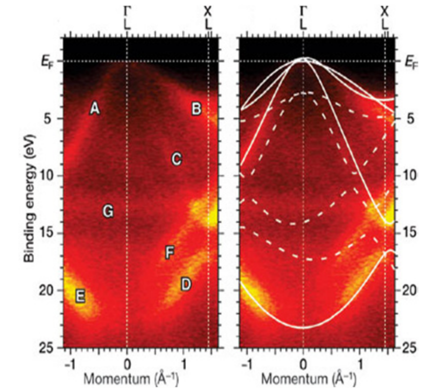
Science 2003

Nanotubes



Nature 2003

Diamond



Nature 2005

Outline Part I

ARPES: Fundamentals and spectral function

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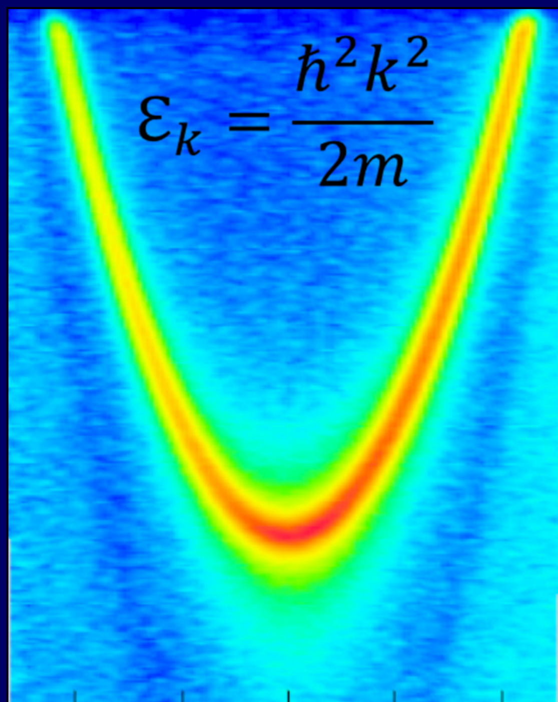
Band Velocity

$$v_k = \frac{1}{\hbar}\frac{\partial \mathcal{E}_k}{\partial k}$$

Cu surface state



Energy

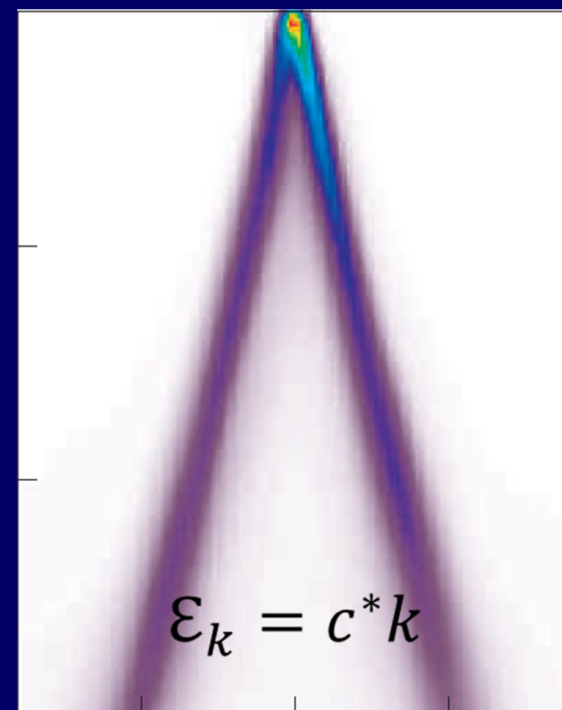
Momentum

Reinert & Hufner, NJP 2005

Band Mass

$$\frac{1}{m_k} = \frac{1}{\hbar^2}\frac{\partial^2 \mathcal{E}_k}{\partial k^2}$$

Graphene

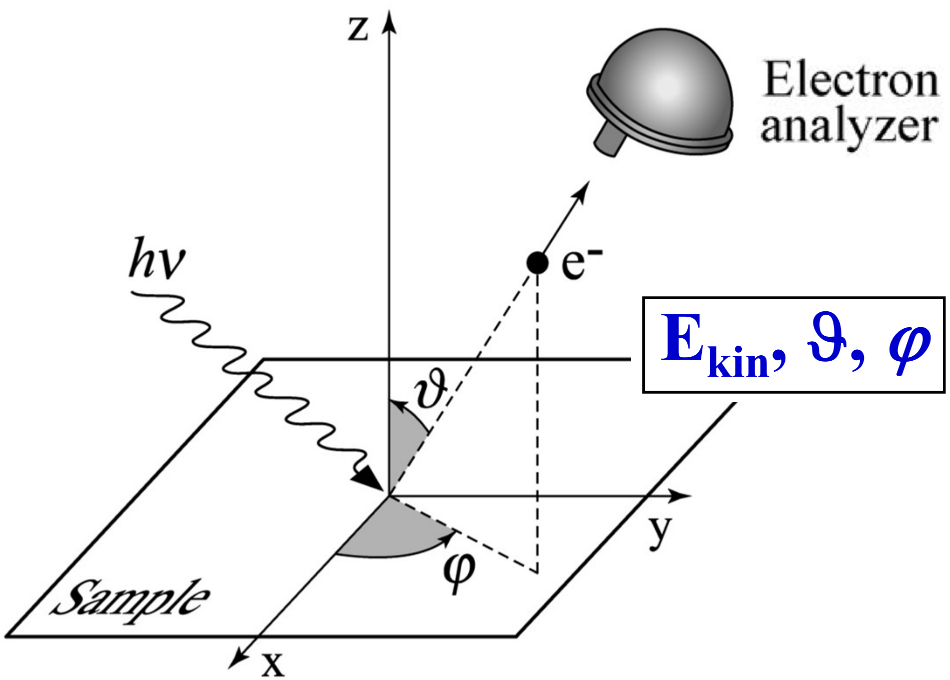


Energy

Momentum

Zhou et al., Nat. Phys. 2006

Angle-Resolved Photoemission Spectroscopy



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

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

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

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

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

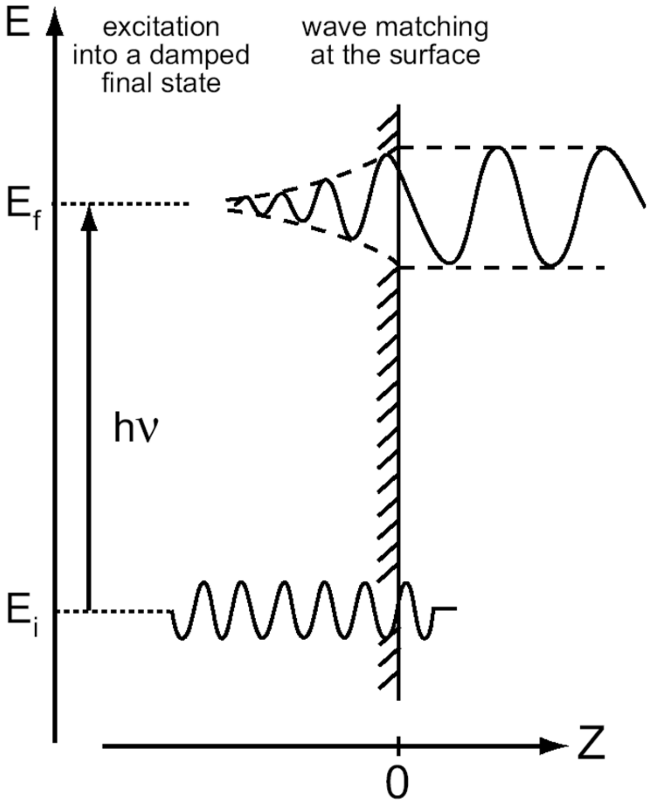
$\mathbf{k}$

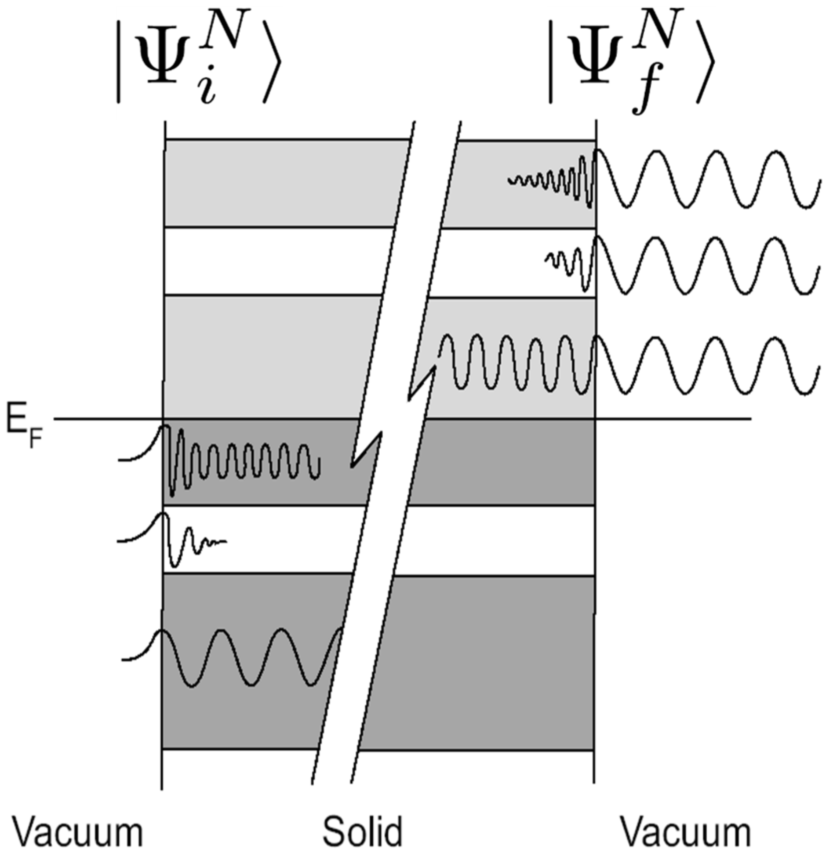
ARPES: One-Step vs Three-Step Model

Photoemission Intensity $I(k,\omega)$

$$w_{fi} \propto |\langle \Psi_f^N | \mathbf{A} \cdot \mathbf{p} | \Psi_i^N \rangle|^2 \delta(E_f^N - E_i^N - h\nu)$$

One-step model

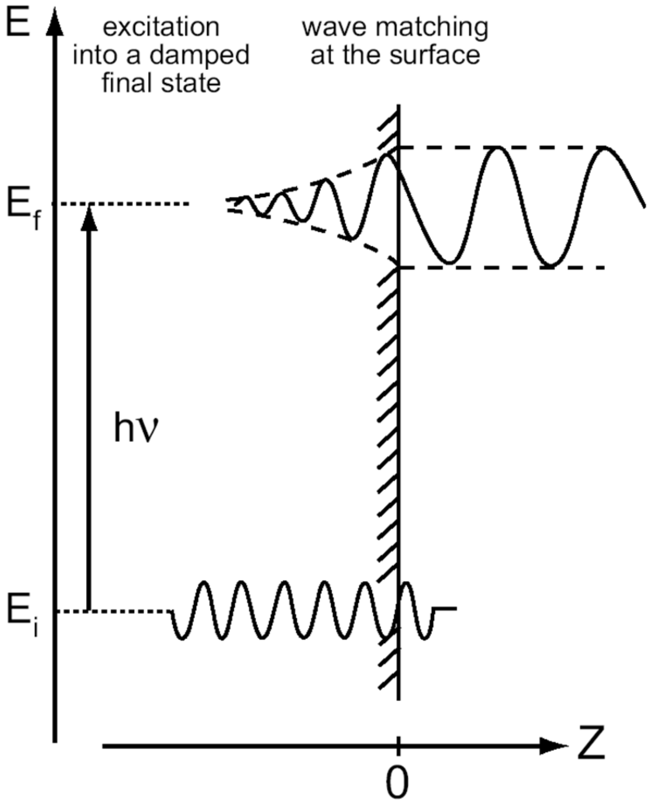




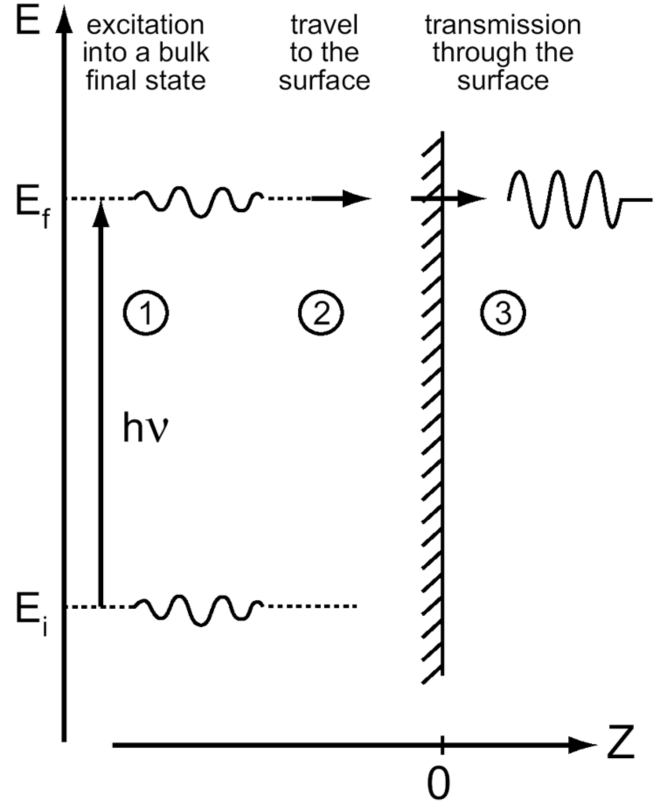
ARPES: One-Step vs Three-Step Model

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

One-step model



Three-step model



ARPES: The Sudden Approximation

Photoemission
Intensity ***I(k,ω)***

$$w_{fi} \propto |\langle \Psi_f^N | \mathbf{A} \cdot \mathbf{p} | \Psi_i^N \rangle|^2 \delta(E_f^N - E_i^N - h\nu)$$

Sudden approximation

One Slater determinant

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

$$\Psi_i^N = \mathcal{A}\, \phi_i^{\mathbf{k}}\, \Psi_i^{N-1}$$

ARPES: The Sudden Approximation

Photoemission Intensity $I(k,\omega)$ $\Big\}$ $w_{fi} \propto |\langle \phi_f^{\mathbf{k}} | \mathbf{A} \cdot \mathbf{p} | \phi_i^{\mathbf{k}} \rangle \langle \Psi_m^{N-1} | \Psi_i^{N-1} \rangle|^2 \delta(\omega - h\nu)$

Sudden approximation

One Slater determinant

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

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ARPES: The Sudden Approximation

Photoemission Intensity $I(k,\omega)$ $\Bigg\}$ $w_{fi} \propto |\langle \phi_f^{\mathbf{k}}|\mathbf{A}\cdot\mathbf{p}|\phi_i^{\mathbf{k}}\rangle\langle\Psi_m^{N-1}|\Psi_i^{N-1}\rangle|^2\delta(\omega-h\nu)$

Sudden approximation

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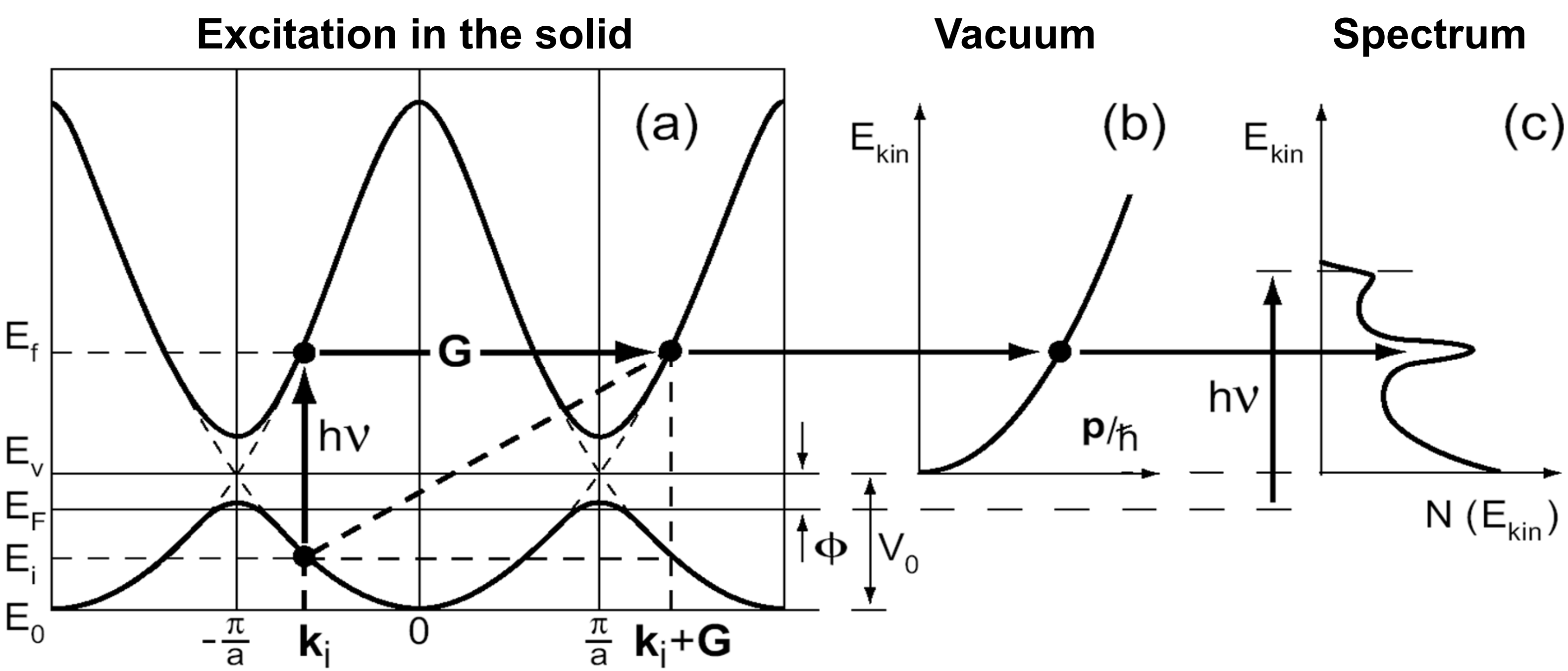


ARPES: Role of the Crystal Potential

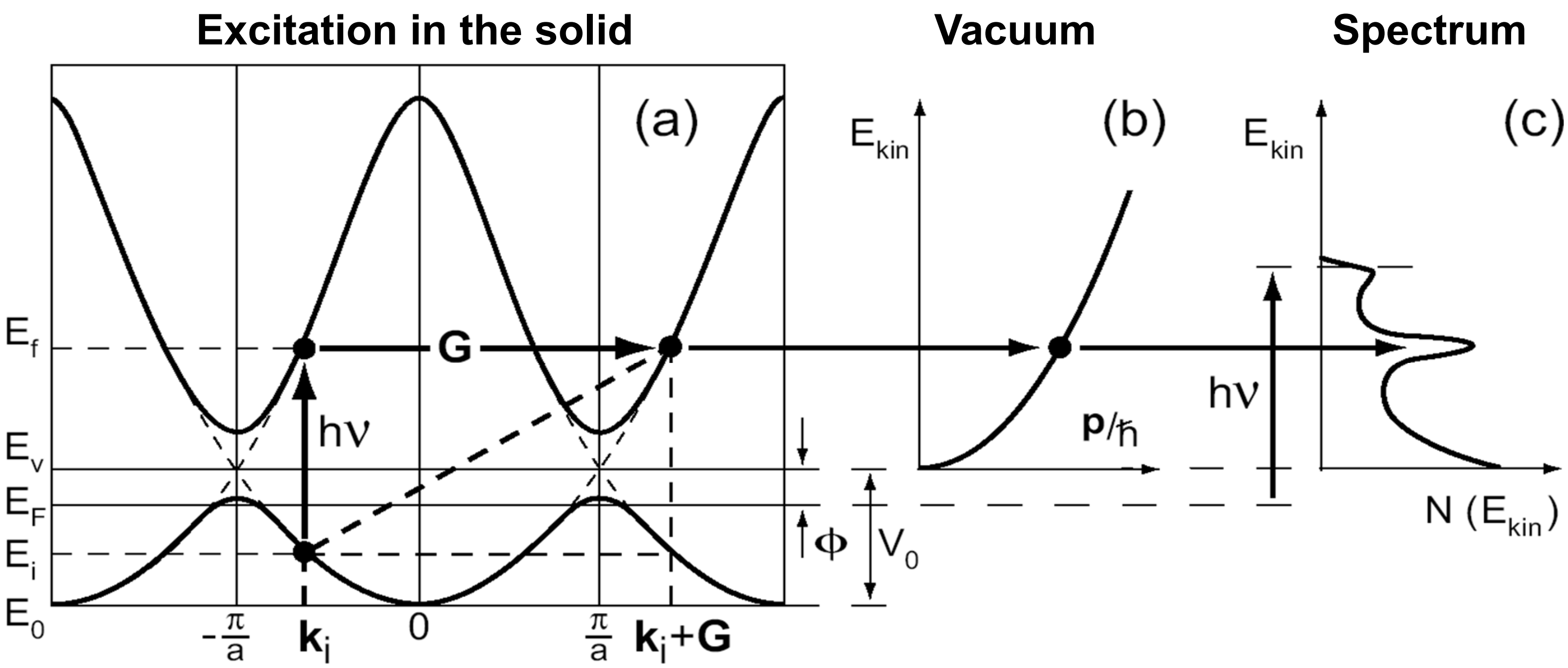
Photoemission Intensity $I(k,\omega)$ $\Big\}$ $w_{fi} \propto |\langle \phi_f^{\mathbf{k}} | \underline{\mathbf{A}\cdot\nabla V} | \phi_i^{\mathbf{k}} \rangle \langle \Psi_m^{N-1} | \Psi_i^{N-1} \rangle|^2 \delta(\omega - h\nu)$

Sudden approximation

One Slater determinant

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

$$\Psi_i^N = \mathcal{A}\, \phi_i^{\mathbf{k}} \Psi_i^{N-1}$$



ARPES: Role of the Crystal Potential

Photoemission Intensity $I(k,\omega)$ $\Big\}$ $w_{fi} \propto |\langle\phi_f^{\mathbf{k}}|\underline{\mathbf{A}\cdot\nabla V}|\phi_i^{\mathbf{k}}\rangle\langle\Psi_m^{N-1}|\Psi_i^{N-1}\rangle|^2\delta(\omega-h\nu)$

Sudden approximation

One Slater determinant

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

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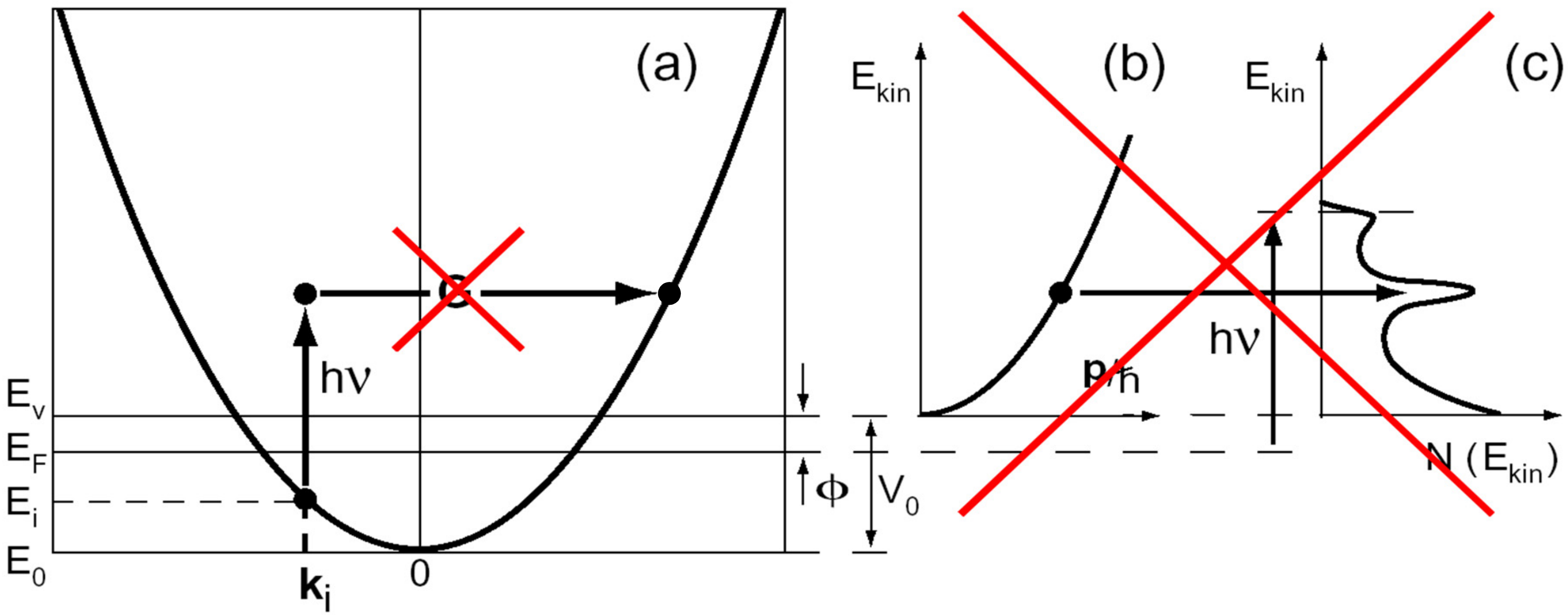


ARPES: Role of the Crystal Potential

Photoemission Intensity $I(k,\omega)$ $\Big\}$ $w_{fi} \propto |\langle\phi_f^{\mathbf{k}}|\underline{\mathbf{A}\cdot\nabla V}|\phi_i^{\mathbf{k}}\rangle\langle\Psi_m^{N-1}|\Psi_i^{N-1}\rangle|^2\delta(\omega-h\nu)$

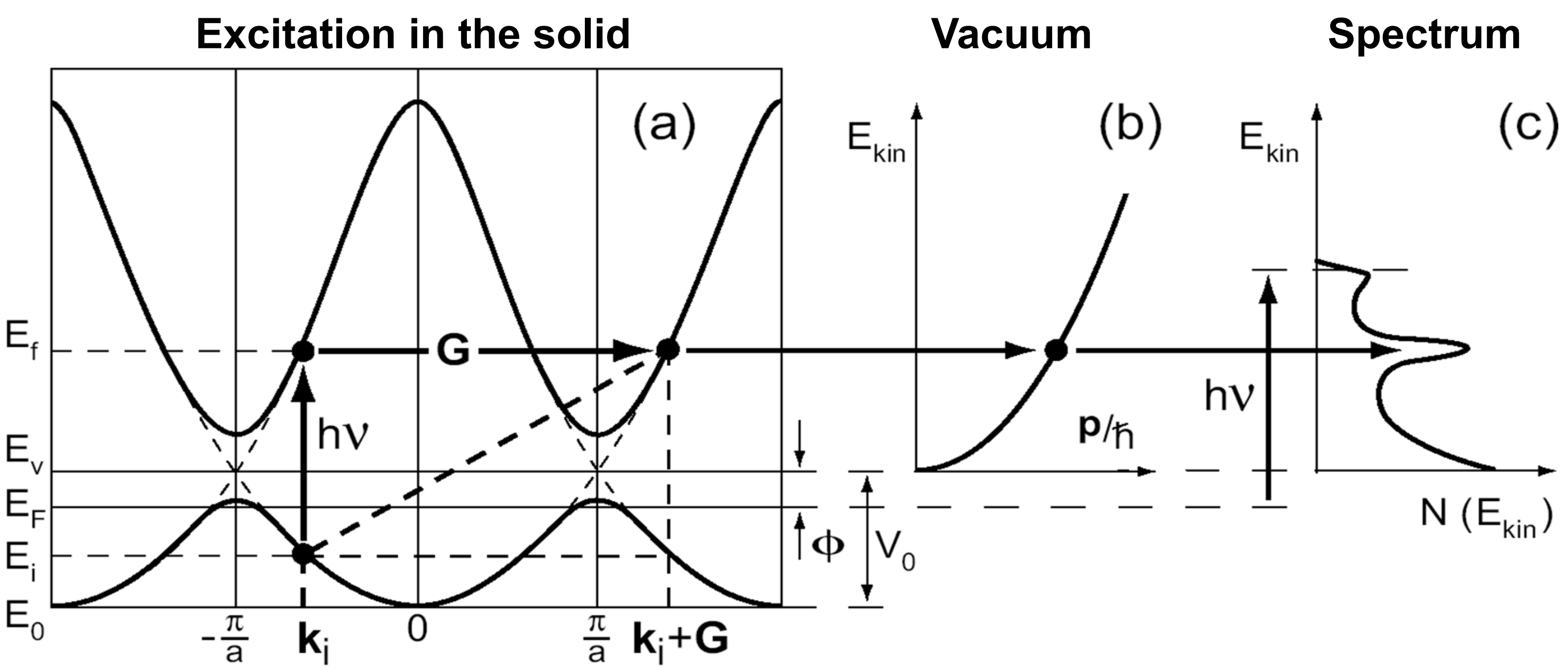
"In a nearly-free electron gas, optical absorption may be viewed as a two-step process. The absorption of the photon provides the electron with the additional energy it needs to get to the excited state. The crystal potential imparts to the electron the additional momentum it needs to reach the excited state. This momentum comes in multiples of the reciprocal-lattice vectors ***G****: So in a reduced zone picture, the transitions are vertical in wave-vector space. But in photoemission, it is more useful to think in an extended-zone scheme."*

G.D. Mahan, Phys. Rev. B **2**, 4334 (1970)

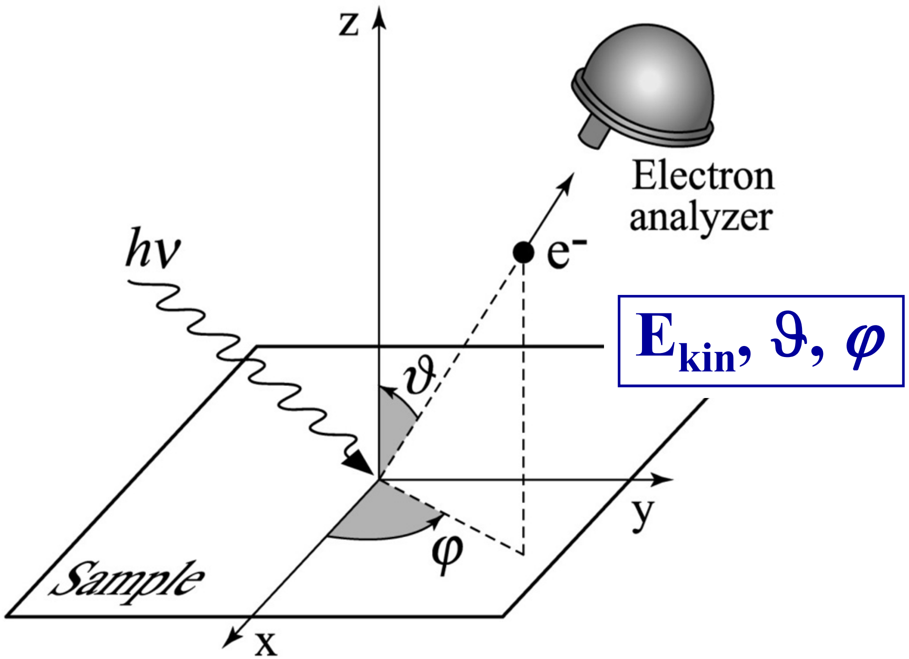
ARPES: Three-step Model & Sudden Approximation

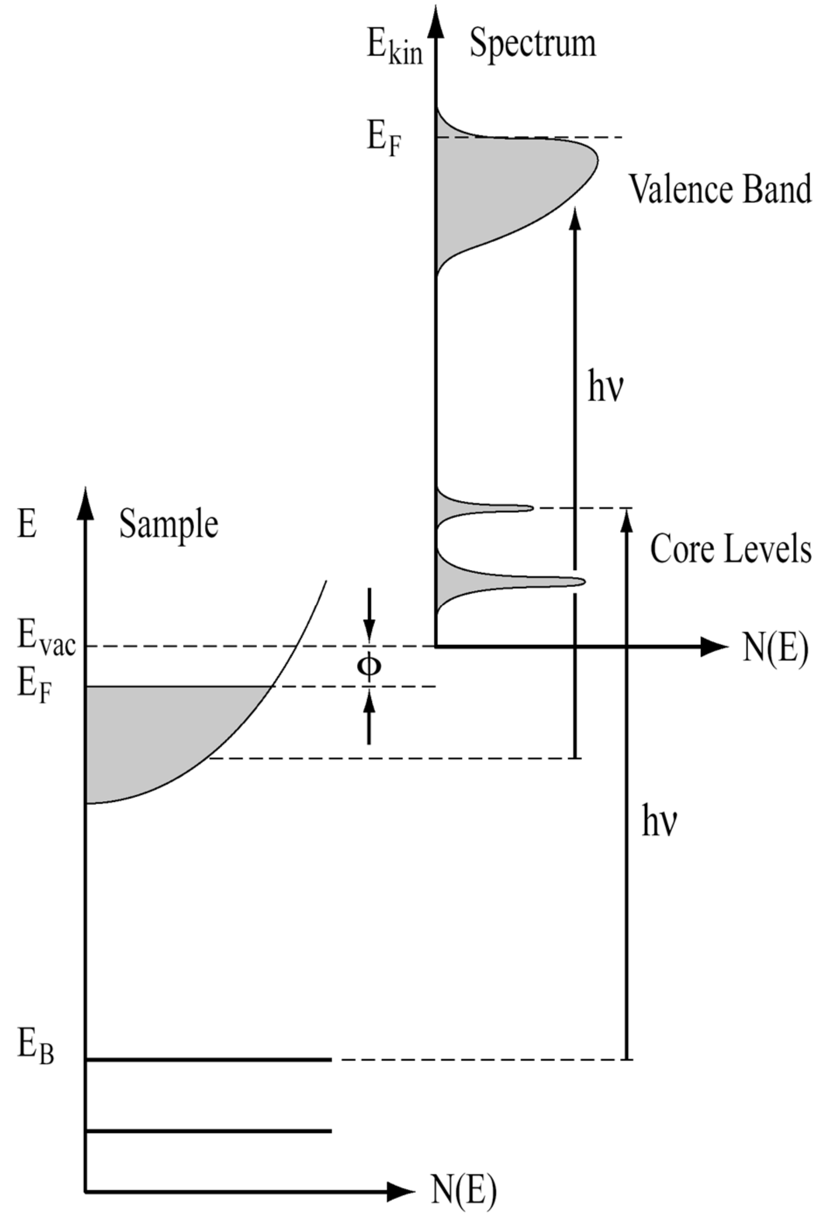
Photoemission Intensity $I(k,\omega)$ $\Big\}$ $w_{fi} \propto |\langle \phi_f^{\mathbf{k}} | \mathbf{A} \cdot \nabla V | \phi_i^{\mathbf{k}} \rangle \langle \Psi_m^{N-1} | \Psi_i^{N-1} \rangle|^2 \delta(\omega - h\nu)$

The photoemission intensity for the SAME band is very DIFFERENT in various Brillouin zones



ARPES: Energetics and Kinematics





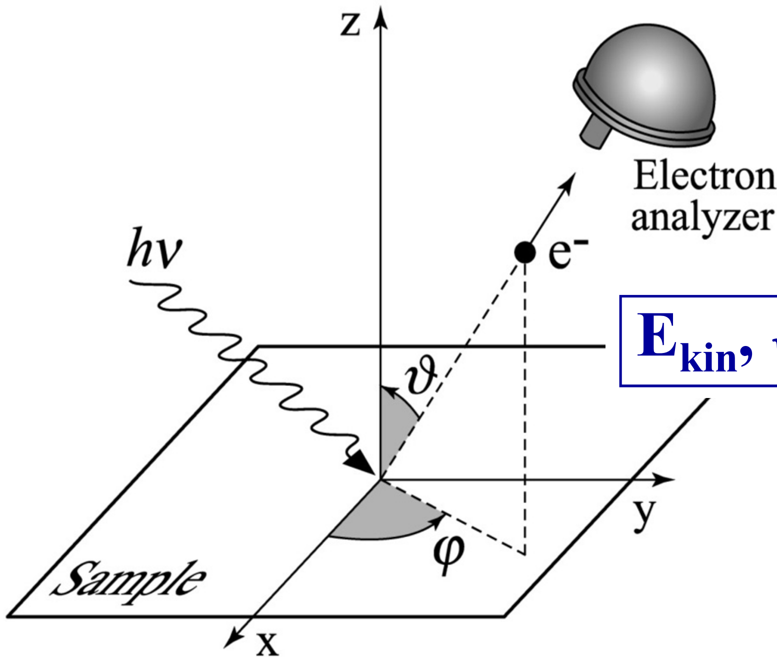
Energy Conservation

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

Momentum Conservation

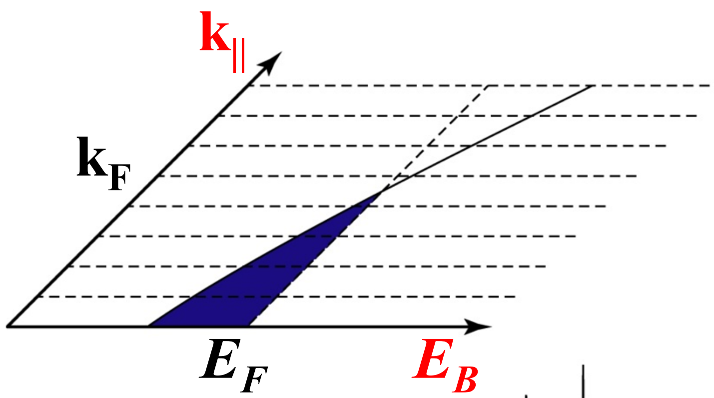
$$\hbar \mathbf{k}_{\|} = \hbar \mathbf{K}_{\|} = \sqrt{2mE_{kin}} \cdot \sin\vartheta$$

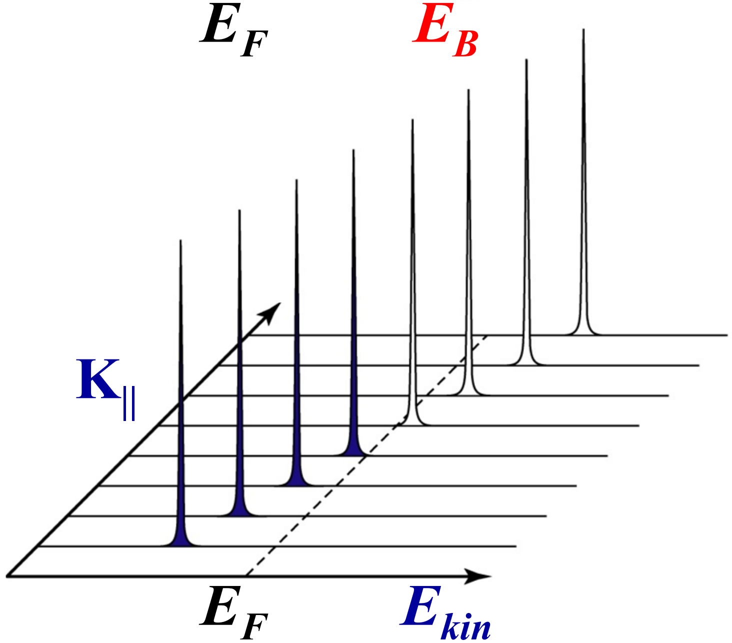
ARPES: Energetics and Kinematics



$E_{kin}, \vartheta, \varphi$

Electrons in Reciprocal Space





Energy Conservation

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

Momentum Conservation

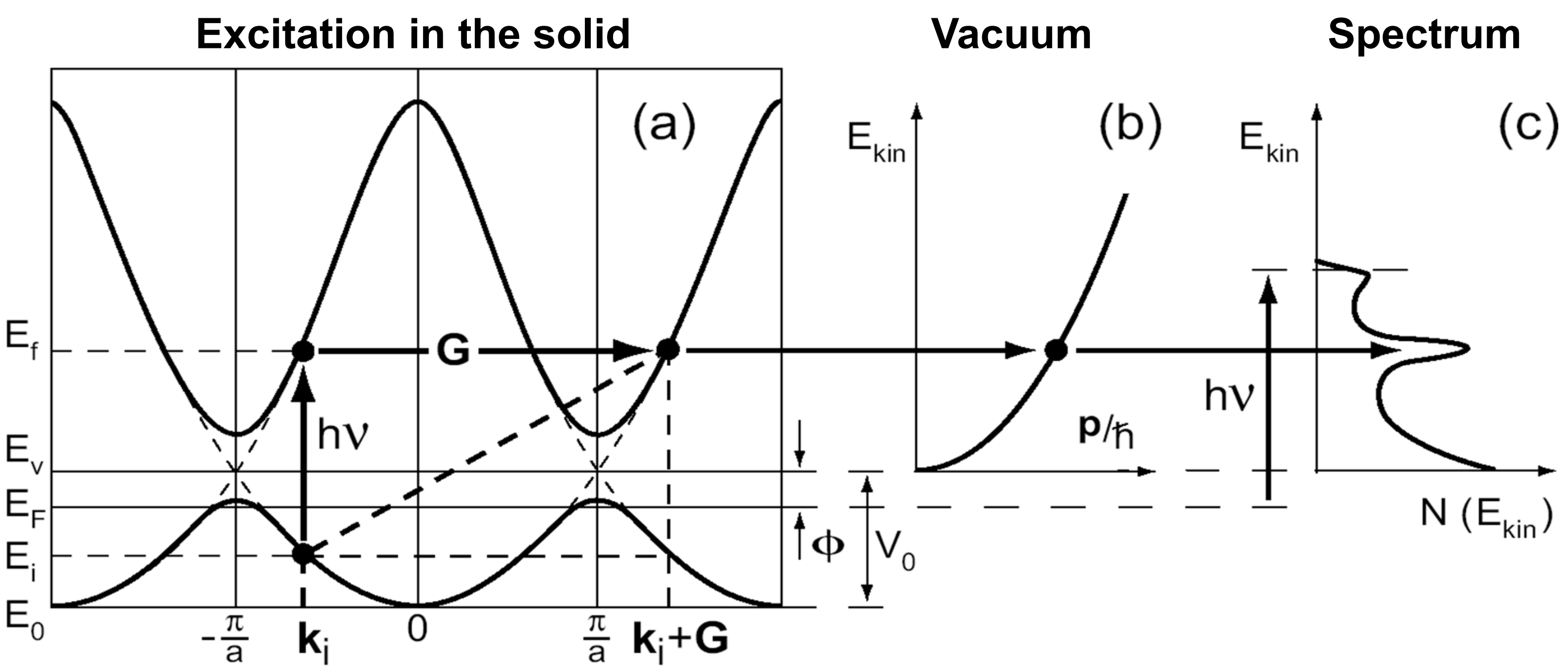
$$\hbar \mathbf{k}_{\parallel} = \hbar \mathbf{K}_{\parallel} = \sqrt{2mE_{kin}} \cdot \sin\vartheta$$

ARPES in 3D: Inner Potential and Determination of k_z

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

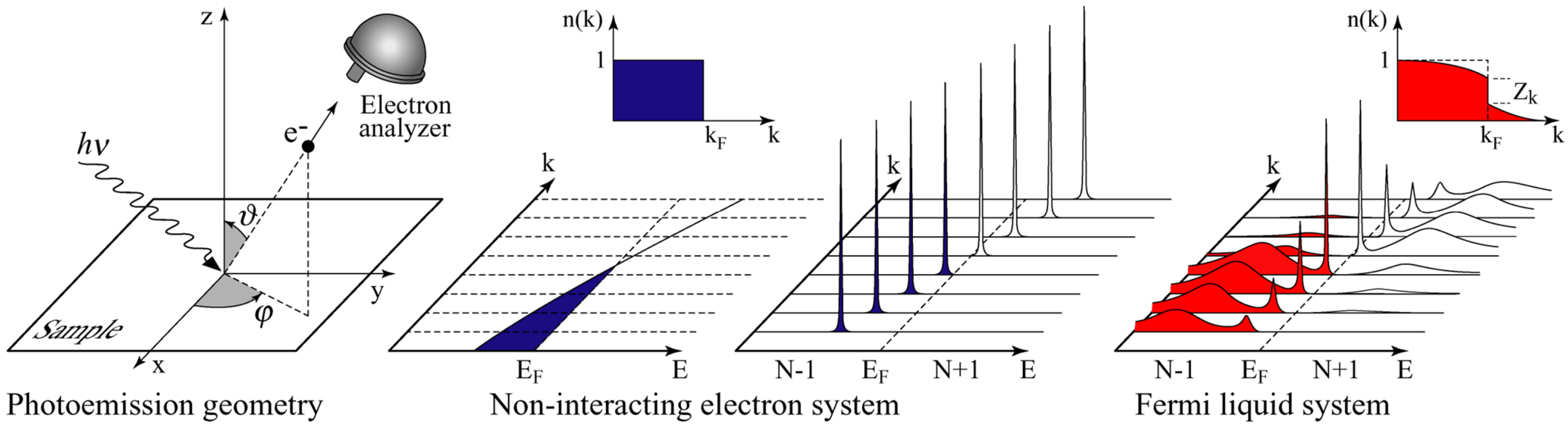
because $\hbar^2 \mathbf{k}_\parallel^2 / 2m = E_{kin} \sin^2 \vartheta$ $\quad E_f = E_{kin} + \phi \quad V_0 = |E_0| + \phi$

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



ARPES: Interacting Systems

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



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

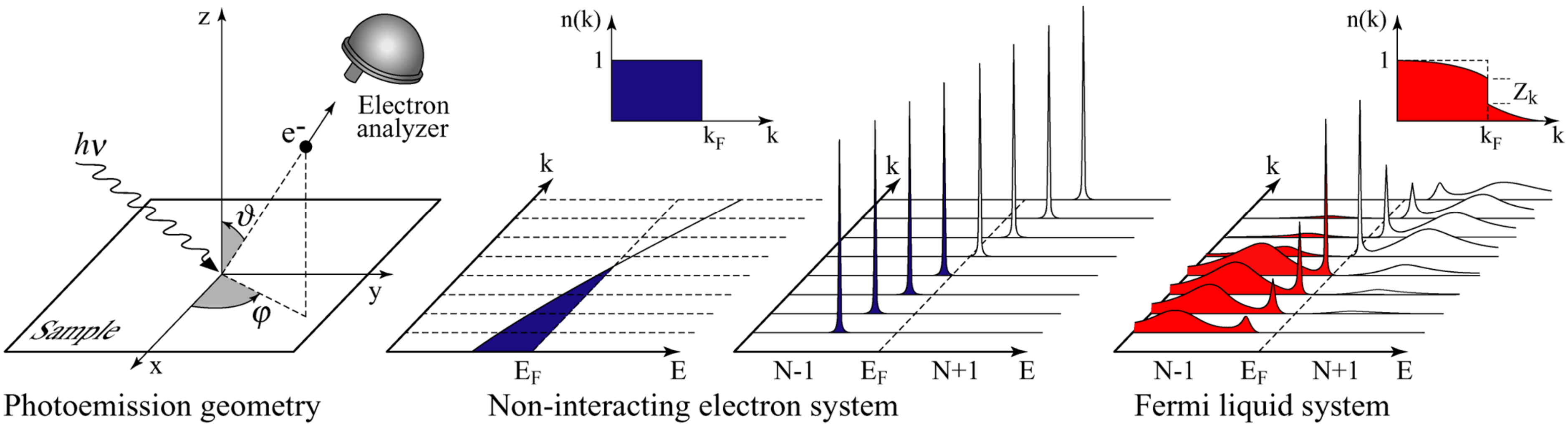
$$I(\mathbf{k}, E_{kin}) \propto \sum_{f,i} |M_{f,i}^{\mathbf{k}}|^2 \sum_m |c_{m,i}|^2 \delta(E_{kin} + E_m^{N-1} - E_i^N - h\nu)$$

$$|M_{f,i}^{\mathbf{k}}|^2 \equiv |\langle \phi_f^{\mathbf{k}} | \mathbf{A} \cdot \mathbf{p} | \phi_i^{\mathbf{k}} \rangle|^2 \qquad |c_{m,i}|^2 = |\langle \Psi_m^{N-1} | \Psi_i^{N-1} \rangle|^2$$

In general $\Psi_i^{N-1} = c_{\mathbf{k}} \Psi_i^N$ **NOT orthogonal** Ψ_m^{N-1}

ARPES: Interacting Systems

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



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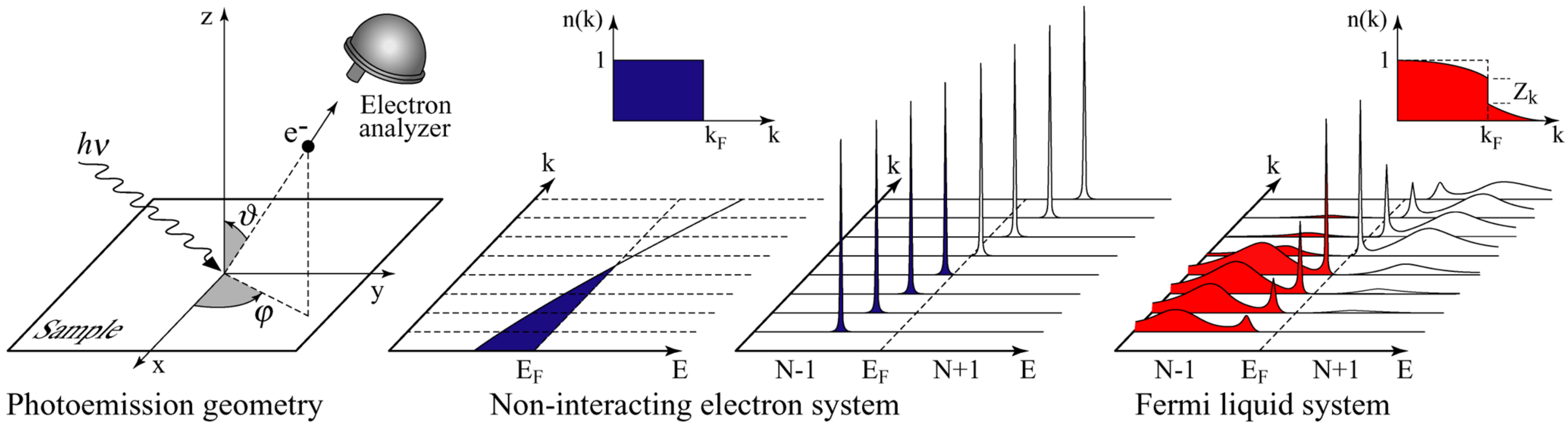
"Like removing a stone from a water bucket"

ARPES: Fermi Liquid Description

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



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

Non-interacting

$$A(\mathbf{k},\omega)=\delta(\omega-\epsilon_k)$$

No Renormalization

Infinite lifetime

Fermi Liquid

$$A(\mathbf{k},\omega) = Z_{\mathbf{k}}\frac{\Gamma_{\mathbf{k}}/\pi}{(\omega-\varepsilon_{\mathbf{k}})^2+\Gamma_{\mathbf{k}}^2} + A_{inc}$$

$$m^* > m \qquad |\varepsilon_{\mathbf{k}}| < |\epsilon_{\mathbf{k}}|$$

$$\tau_{\mathbf{k}} = 1/\Gamma_{\mathbf{k}}$$

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

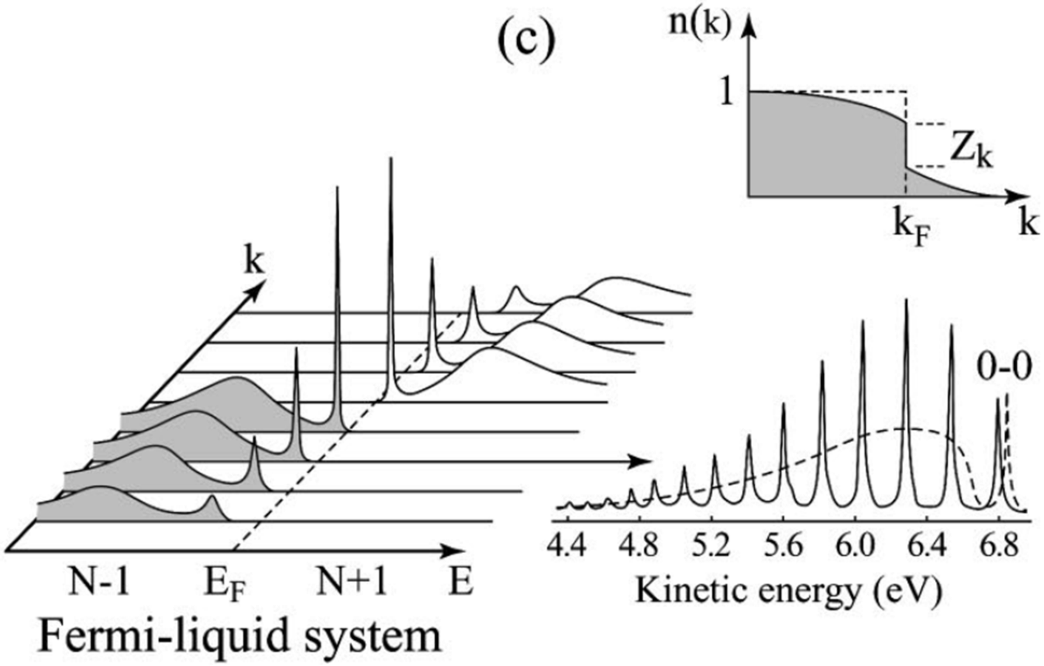
NEWS AND VIEWS

Nature **342**, 480 (1989)

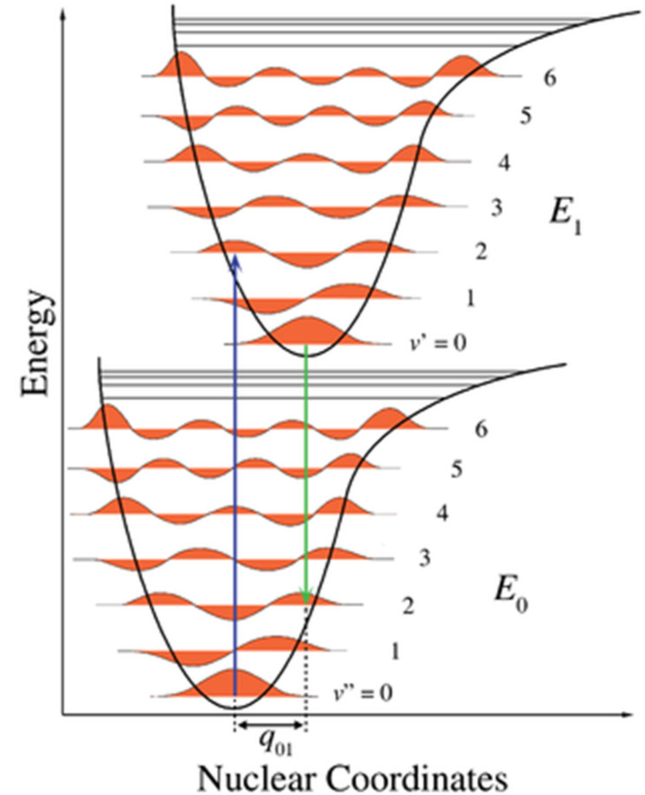
HIGH-TEMPERATURE SUPERCONDUCTIVITY

Testing Fermi-liquid models

G.A. Sawatzky



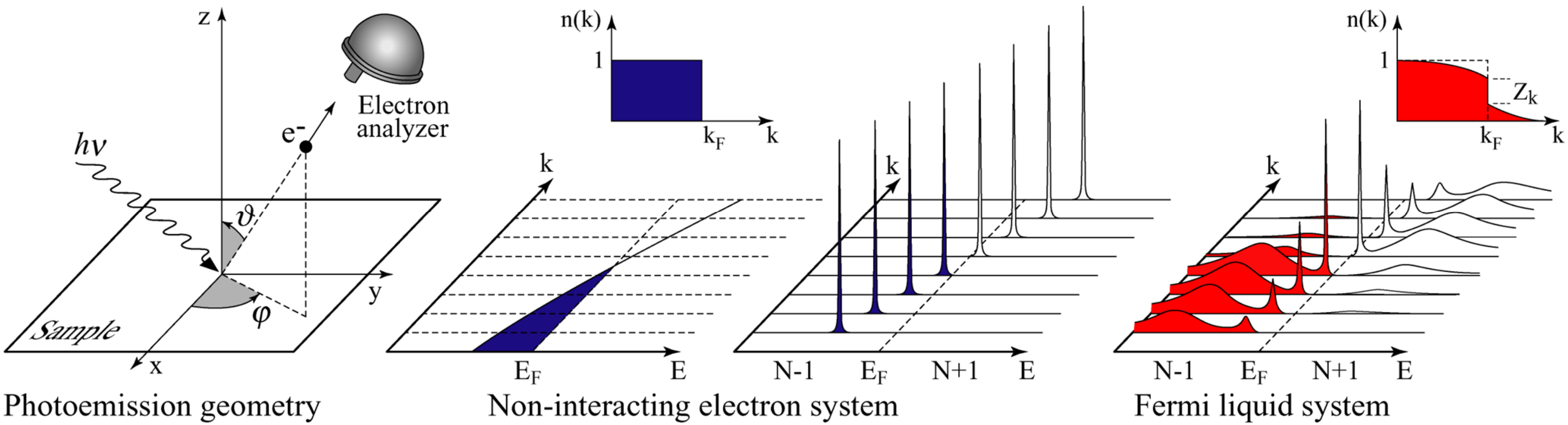
Franck-Condon



"In gaseous hydrogen, the equilibrium bond length is dependent on the degree of occupation of that level. The electrons are dressed by interatomic displacements. The intensities are given by the Franck-Condon factors, the molecular equivalent of the sudden approximation. The ARPES spectrum of solid hydrogen, developed from the molecular spectrum, will be angle dependent but for some angle will resemble the broken line. The fundamental transition (0-0) becomes the solid state quasiparticle peak. The phonon excitations develop into a broad, incoherent quasicontinuum."

ARPES: The One-Particle Spectral Function

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



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

Single-particle spectral function

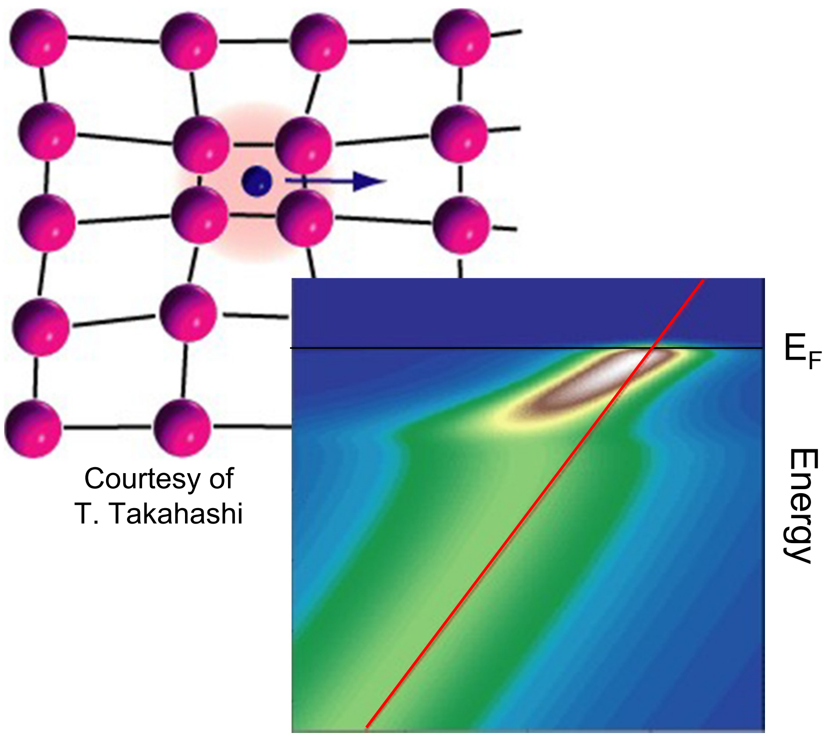
$$A(\mathbf{k},\omega) = -\frac{1}{\pi}\frac{\Sigma''(\mathbf{k},\omega)}{[\omega-\epsilon_{\mathbf{k}}-\Sigma'(\mathbf{k},\omega)]^2+[\Sigma''(\mathbf{k},\omega)]^2}$$

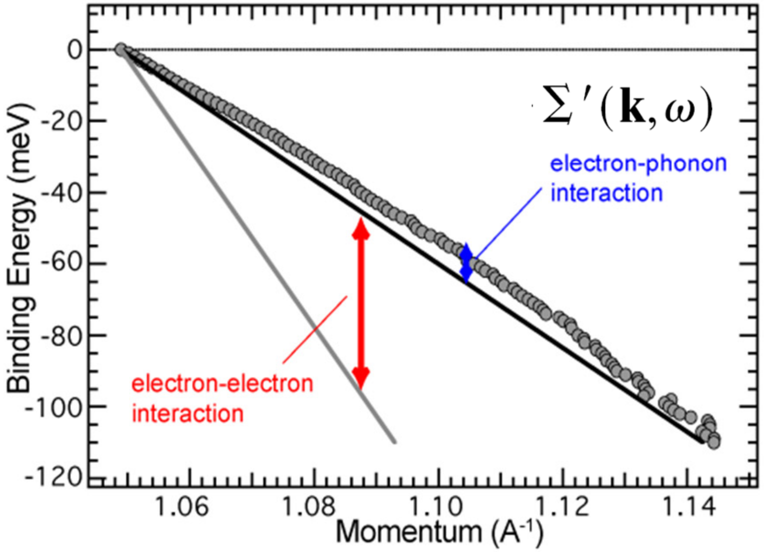
Σ(k,ω) : the "self-energy" captures the effects of interactions

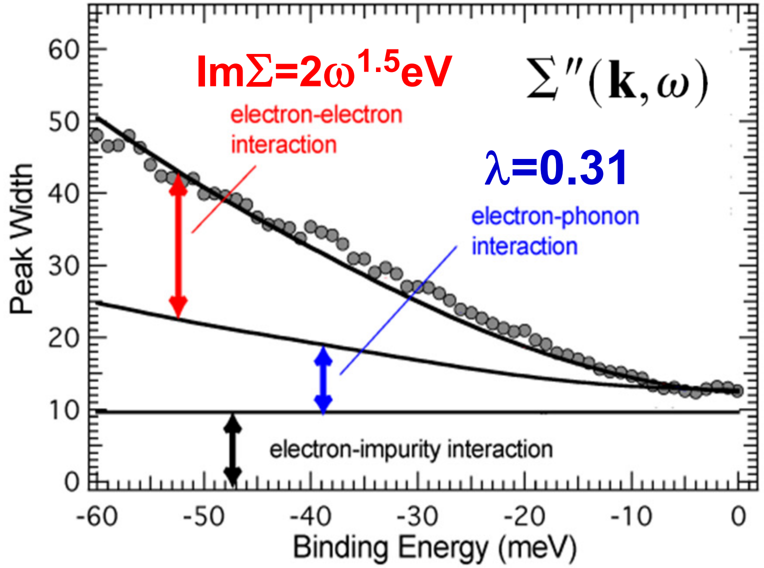
Many-Body Correlation Effects in Sr_2RuO_4

Single-particle spectral function

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







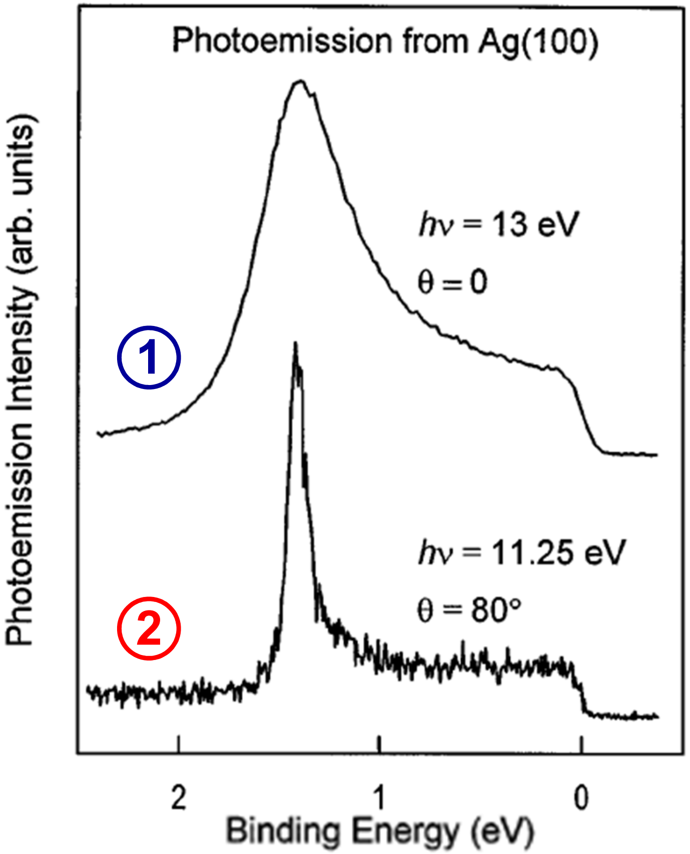
N.J.C. Ingle, K.M. Shen, A. Damascelli *et al.*, PRB **72**, 205114 (2005)

ARPES: FWHM and Inverse Lifetime in 3D

FWHM of an ARPES peak

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

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



① if $E_i \simeq E_F$

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

② if $|v_{i\perp}| \simeq 0$

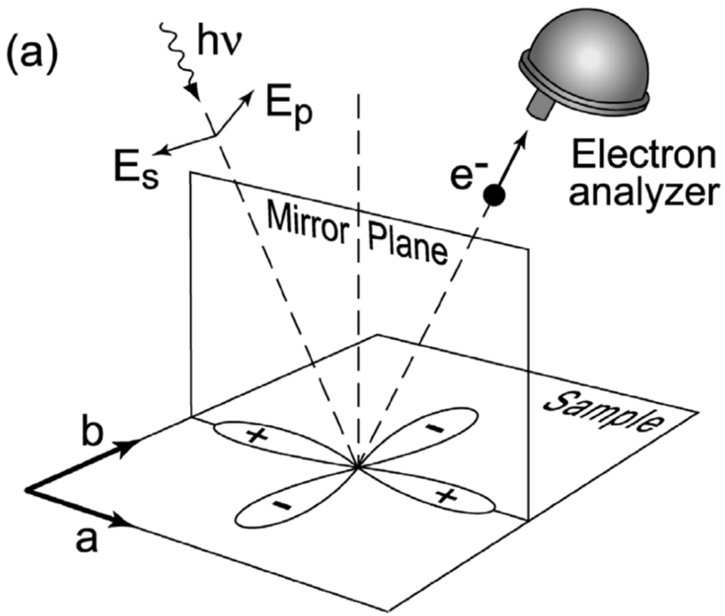
→ $\Gamma = \frac{\Gamma_i}{\left| 1 - \frac{m v_{i\parallel} \sin^2 \vartheta}{\hbar k_\parallel} \right|} \equiv C\,\Gamma_i$

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

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

ARPES: Matrix Elements Effects (polarization + energy)

Photon polarization

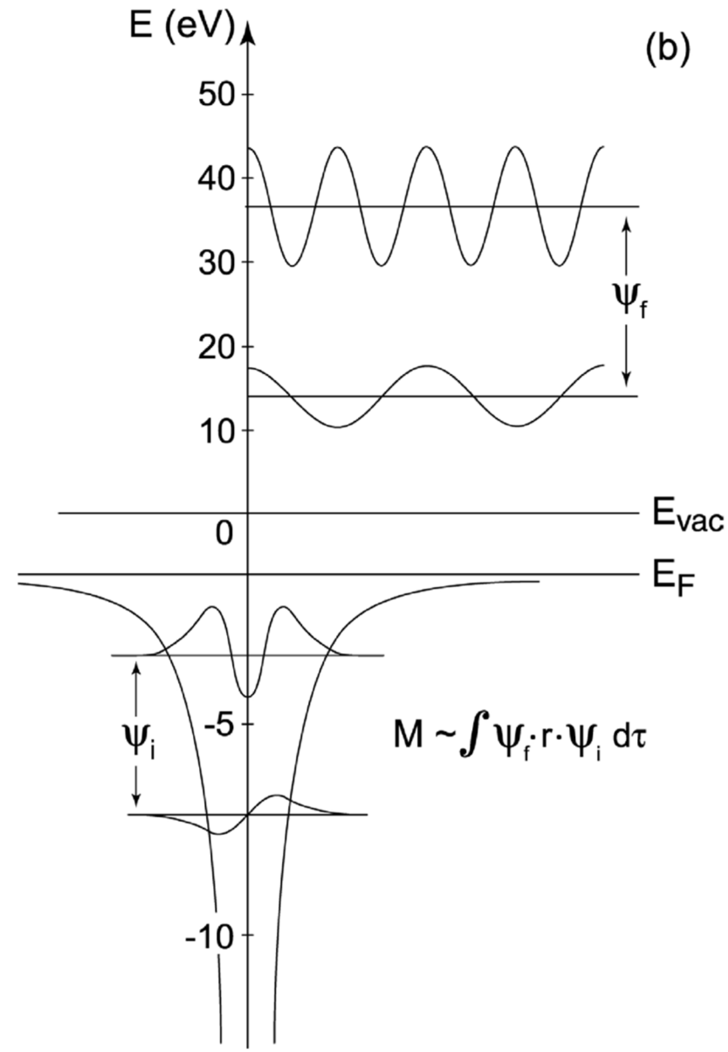


$$w_{fi}=\frac{2\pi}{\hbar}|\langle\Psi_f^N|H_{int}|\Psi_i^N\rangle|^2\,\delta(E_f^N-E_i^N-h\nu)$$

$$\Psi_i^N=\mathcal{A}\,\phi_i^{\mathbf{k}}\,\Psi_i^{N-1} \qquad \Psi_f^N=\mathcal{A}\,\phi_f^{\mathbf{k}}\,\Psi_f^{N-1}$$

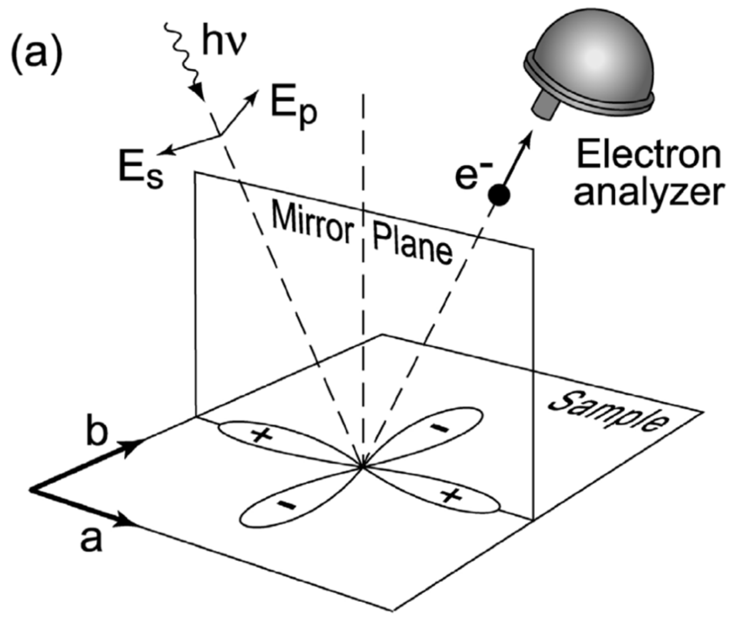
$$\langle\Psi_f^N|H_{int}|\Psi_i^N\rangle=\langle\phi_f^{\mathbf{k}}|H_{int}|\phi_i^{\mathbf{k}}\rangle\langle\Psi_m^{N-1}|\Psi_i^{N-1}\rangle$$

Photon energy



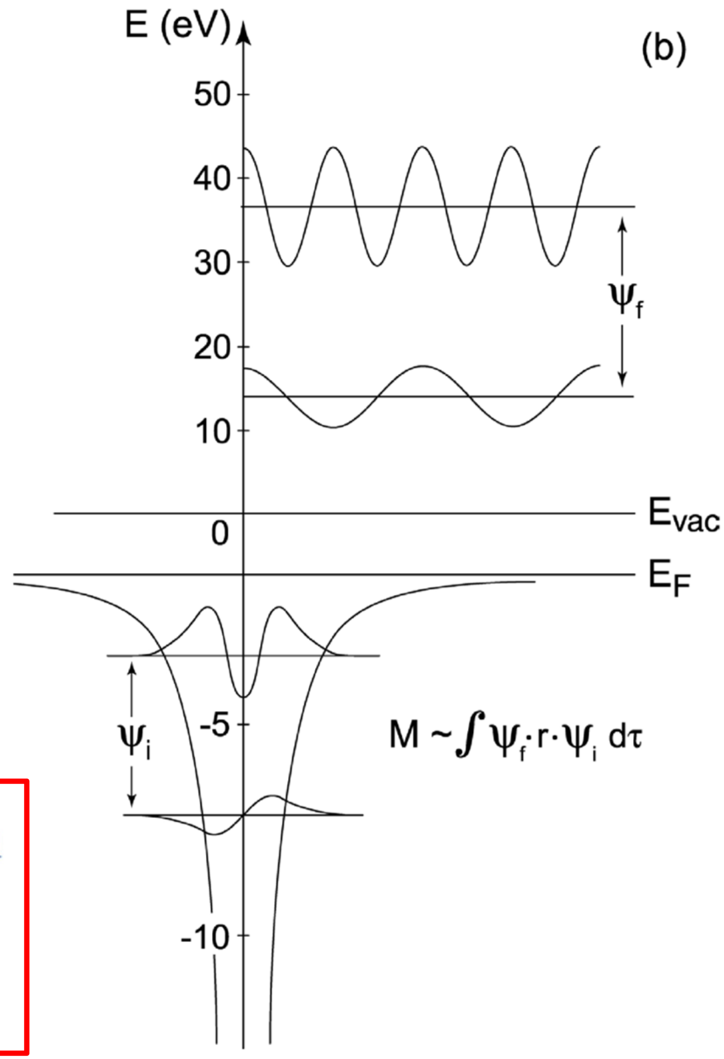
ARPES: Matrix Elements Effects (polarization + energy)

Photon polarization



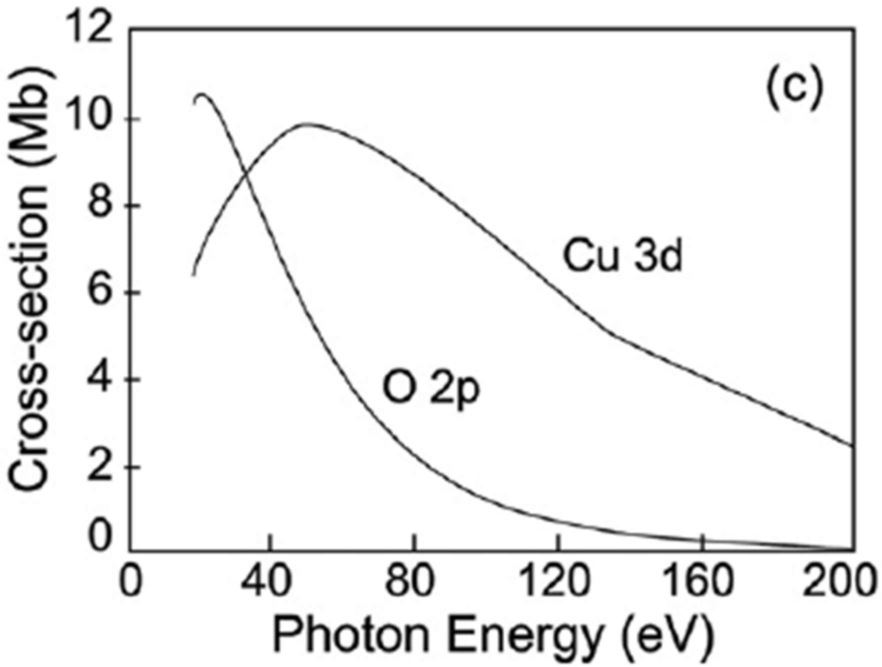
$$\left\langle \phi_f^k | A \cdot p | \phi_i^k \right\rangle \begin{cases} \phi_i^k \text{ even } \langle + | + | + \rangle \Rightarrow A \text{ even} \\ \phi_i^k \text{ odd } \langle + | - | - \rangle \Rightarrow A \text{ odd} \end{cases}$$

Photon energy

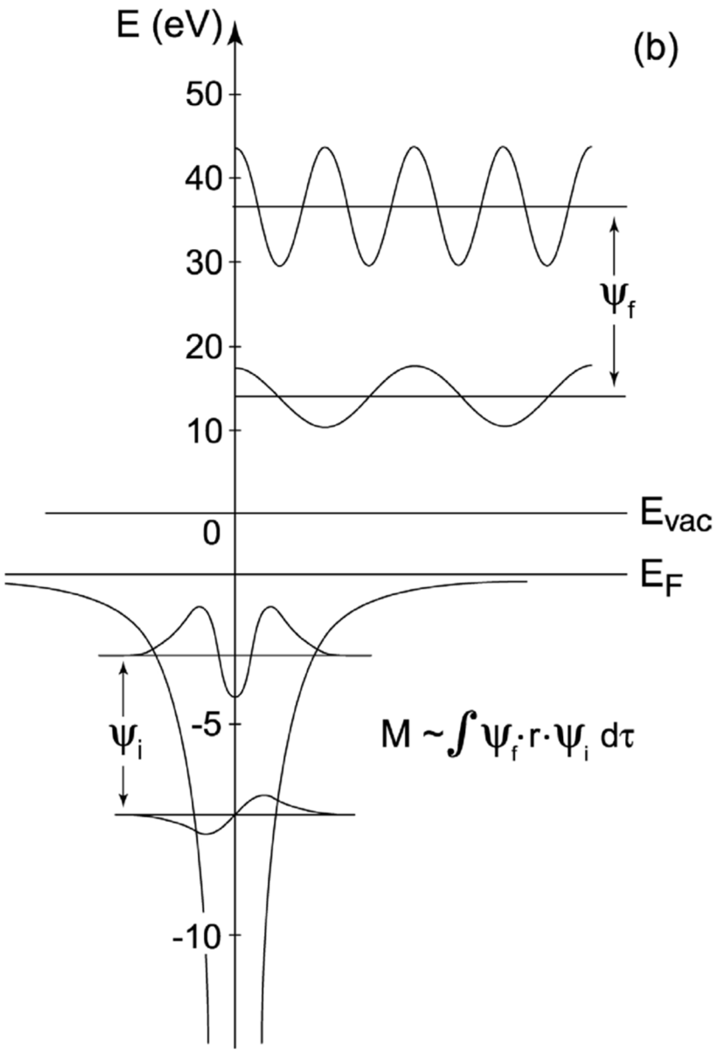


ARPES: Matrix Elements Effects (polarization + energy)

One Cooper minimum in free atoms



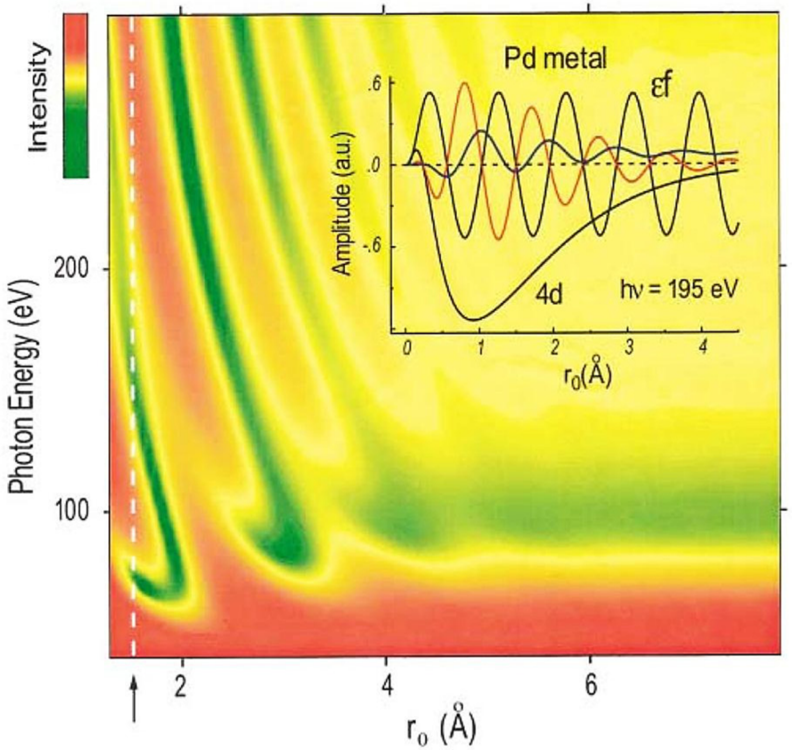
Photon energy



$$\left\langle \phi_f^k | \boldsymbol{A} \cdot \boldsymbol{p} | \phi_i^k \right\rangle^2 \propto |(\varepsilon \cdot \boldsymbol{k}) \langle \phi_i^k | e^{i\boldsymbol{kr}} \rangle|^2$$

ARPES: Matrix Elements Effects (polarization + energy)

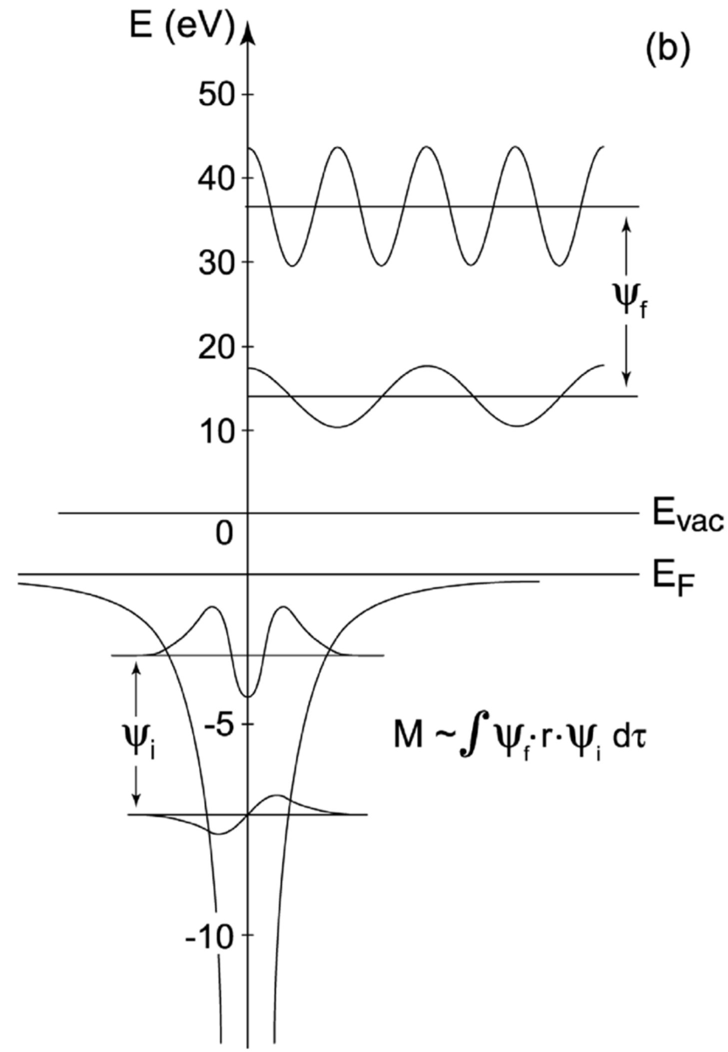
Several Cooper minima in solids



$$\left\langle \phi_f^{\mathbf{k}} | \boldsymbol{A} \cdot \boldsymbol{p} | \phi_i^{\mathbf{k}} \right\rangle^2 \propto |(\varepsilon \cdot \boldsymbol{k}) \langle \phi_i^{\mathbf{k}} | e^{i\boldsymbol{kr}} \rangle|^2$$

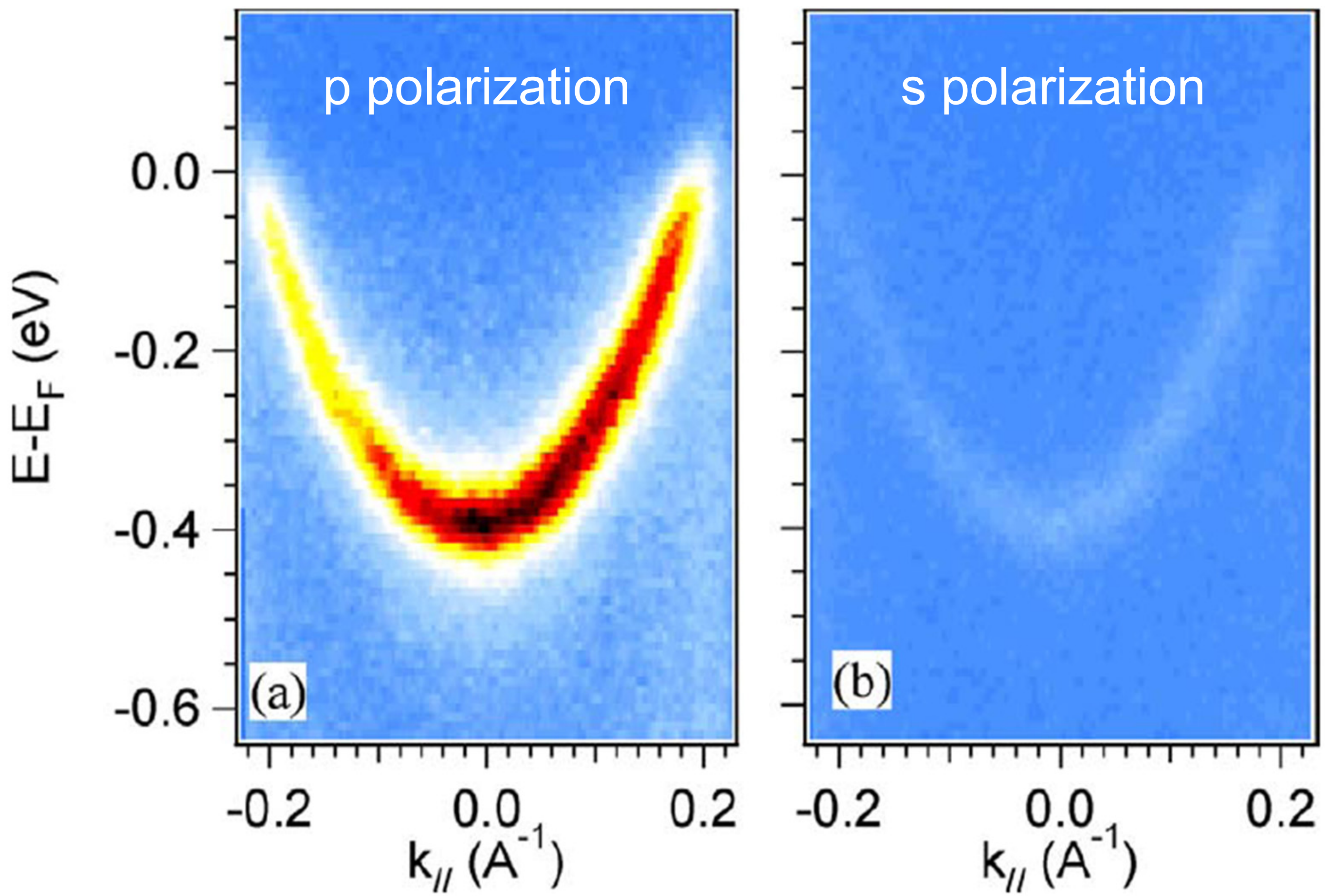
Molodstov et al., Phys. Rev. Lett. **85**, 4184(2000)

Photon energy



ARPES: Polarization Dependence

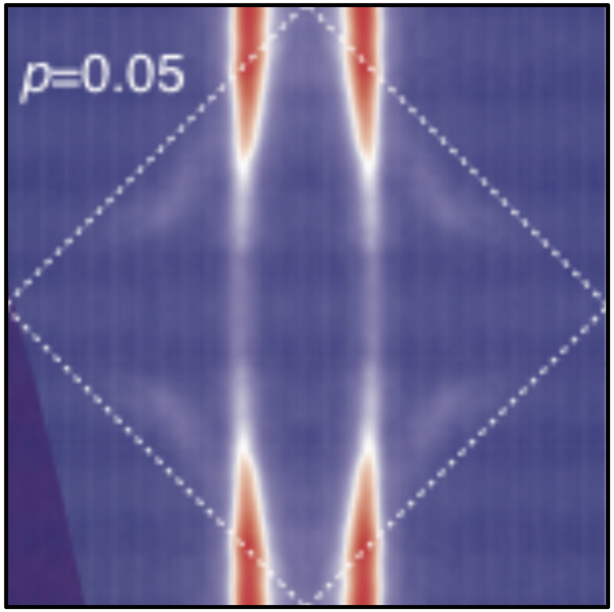
Dispersion of the Cu(111) surface state

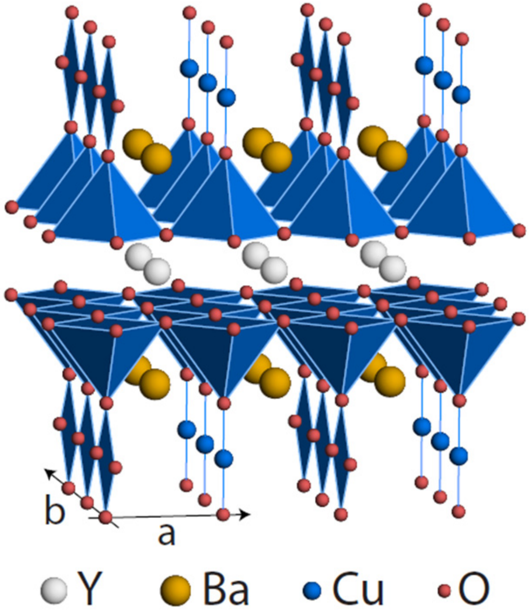


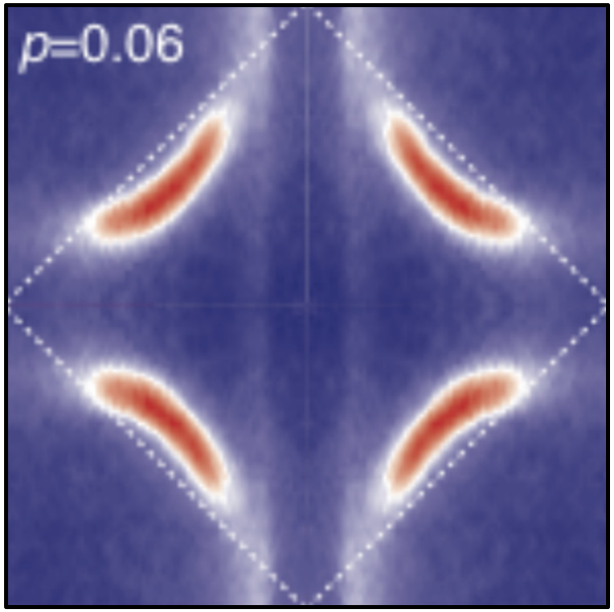
M. Mulazzi et al., Phys. Rev. B **79**, 165421 (2009)

ARPES: Polarization Dependence

Fermi surface of underdoped YBCO





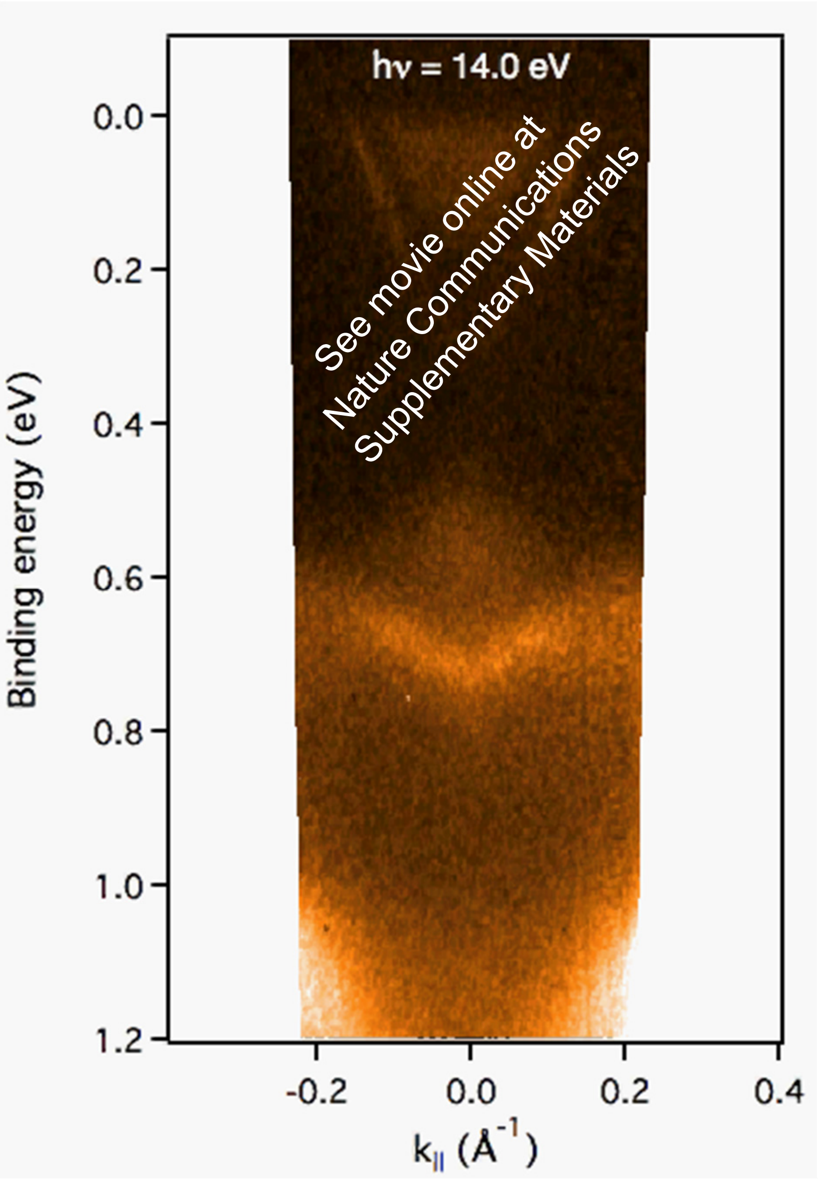


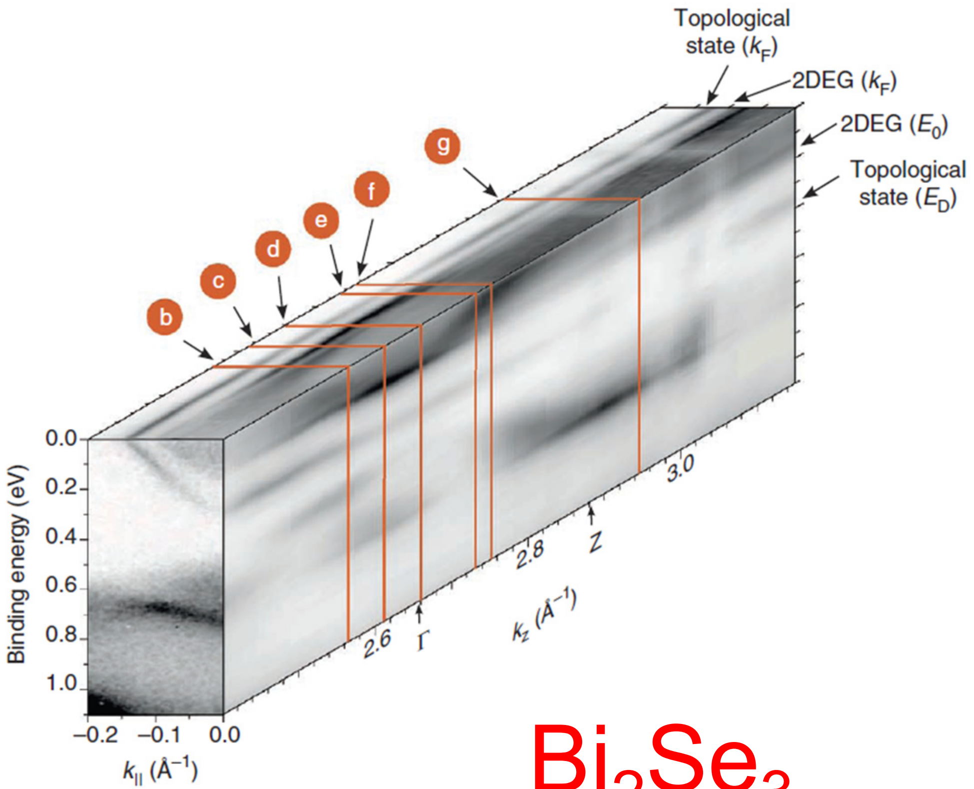
E || CuO chains

E ⊥ CuO chains

Fournier et al., Nature Physics **6**, 905 (2010)

ARPES: Photon Energy Dependence

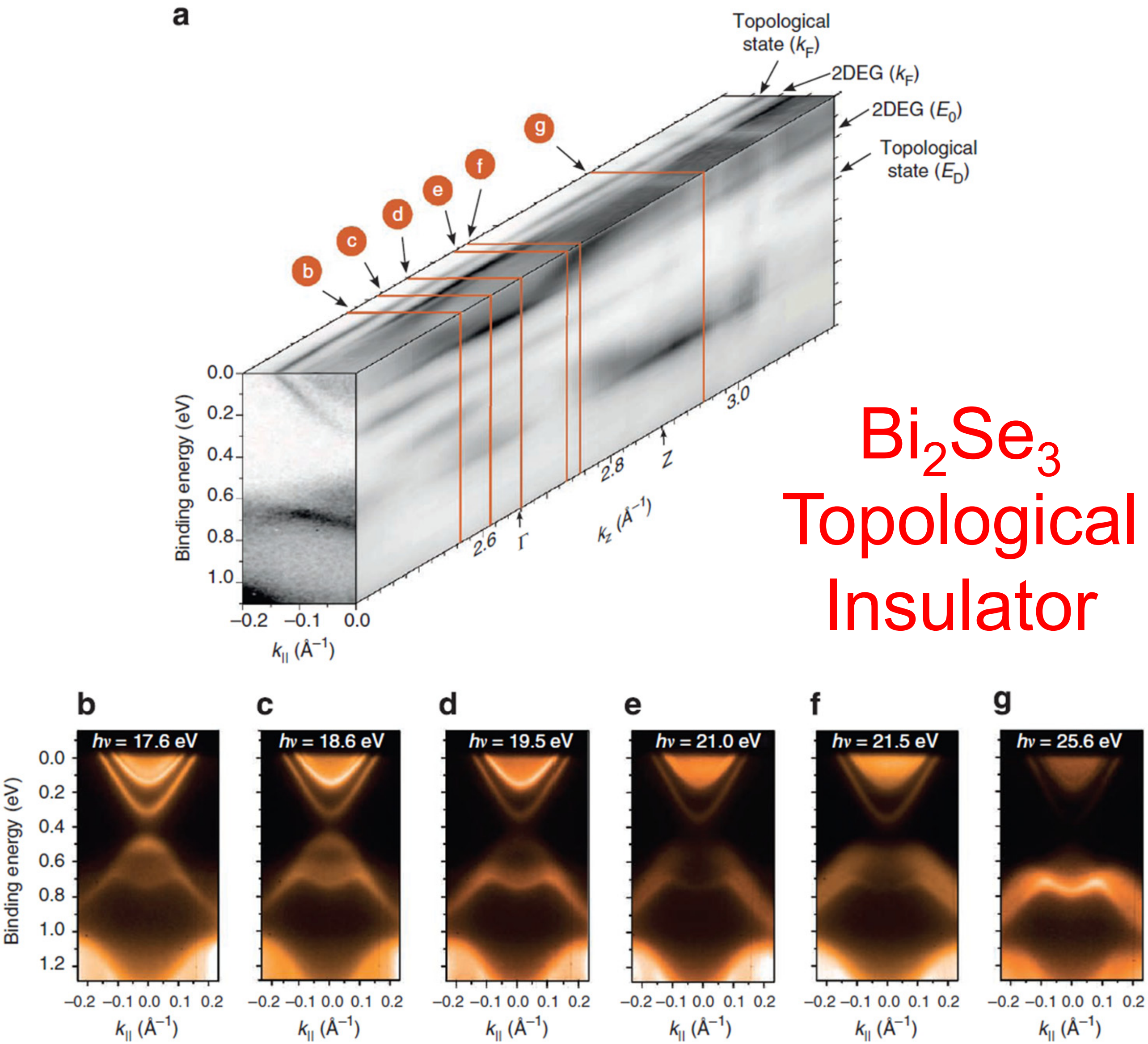




Bi_2Se_3 Topological Insulator

Bianchi et al., Nature Comm. **1**, 128 (2010)

ARPES: Photon Energy Dependence



Bi_2Se_3 Topological Insulator

Bianchi et al., Nature Comm. **1**, 128 (2010)

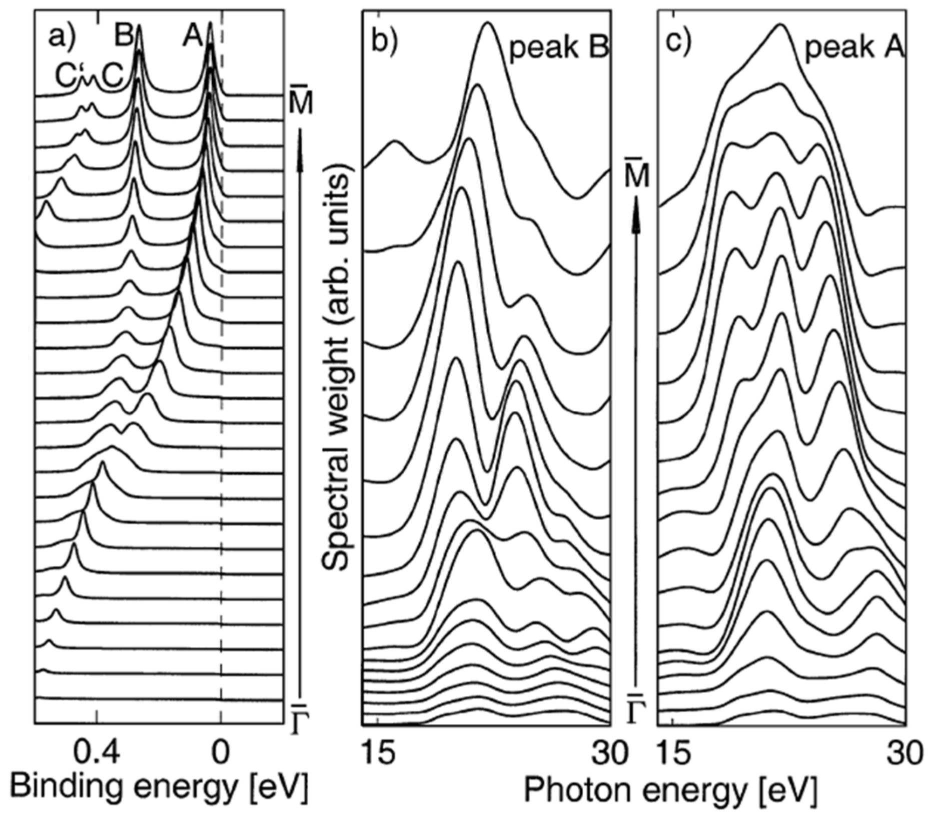
Importance of Matrix Elements in the ARPES Spectra of BISCO

A. Bansil[1] and M. Lindroos[1,2]
[1]*Physics Department, Northeastern University, Boston, Massachusetts 02115*
[2]*Tampere University of Technology, P.O. Box 692, FIN-33101 Tampere, Finland*
(Received 9 July 1999)

$$P(E,\omega) \sim |\langle \Psi_f^{\text{bulk}} | \Delta | \Psi_i^{\text{bulk}} \rangle|^2 A_i(E)$$

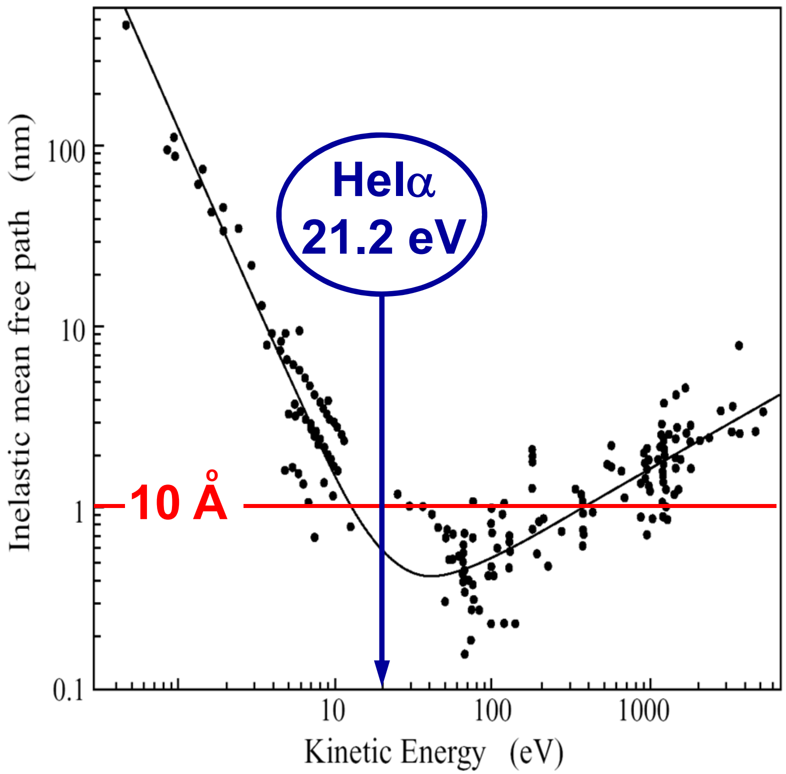
One-step model calculation + LDA

$$I(\mathbf{k}_\parallel, E, \hbar\omega) = \frac{1}{\pi} \operatorname{Im}\langle \mathbf{k}_\parallel | G_2^+(E + \hbar\omega) \Delta G_1^+(E) \Delta^\dagger \times G_2^-(E + \hbar\omega) | \mathbf{k}_\parallel \rangle$$

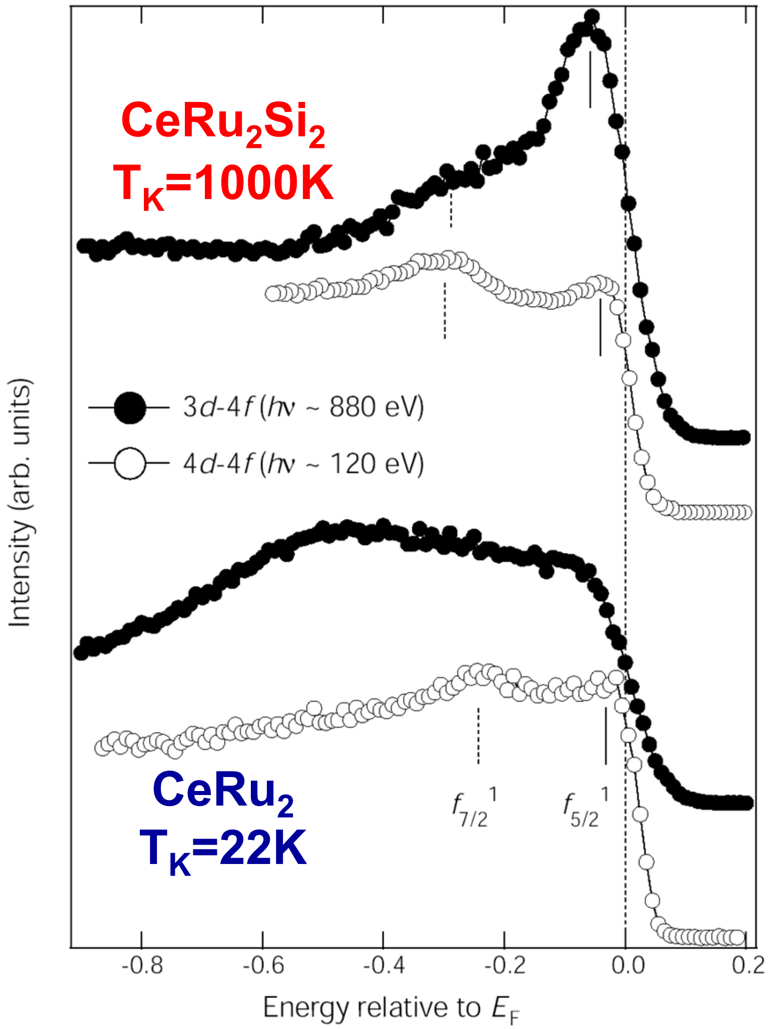


ARPES: Surface vs Bulk Sensitivity

Mean-free path for excited electrons



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



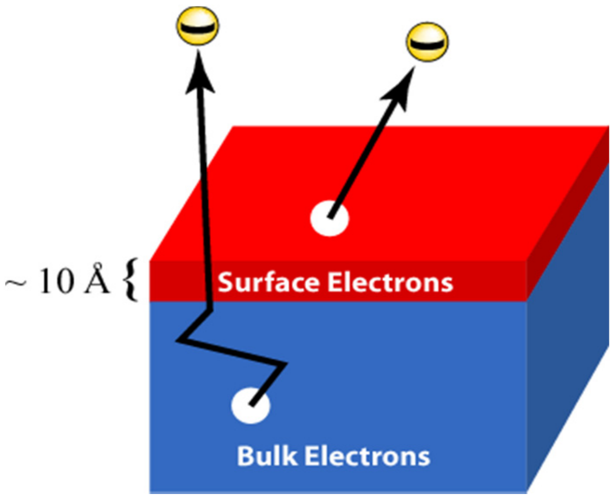
Sekiyama *et al.*, Nature **403**, 396 (2000)

ARPES: 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

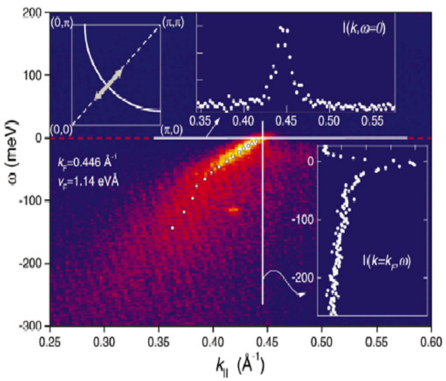


- **Not bulk sensitive**
- Requires clean, atomically flat surfaces in **ultra-high vacuum**
- Cannot be studied as a function of pressure or magnetic field

Courtesy of Kyle Shen

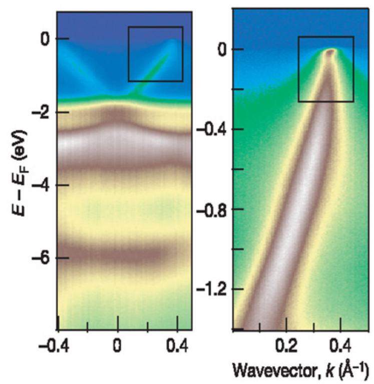
ARPES: Widespread Impact in Complex Materials

HTSC's



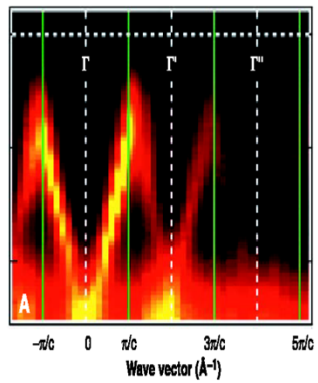
Science 1999

CMR's



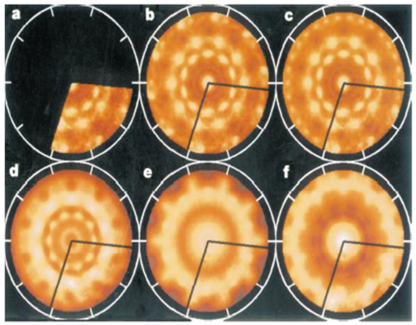
Nature 2005

CDW's



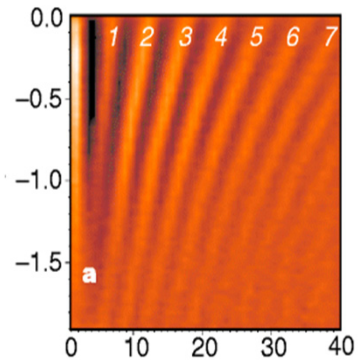
Science 2000

Quasicrystals



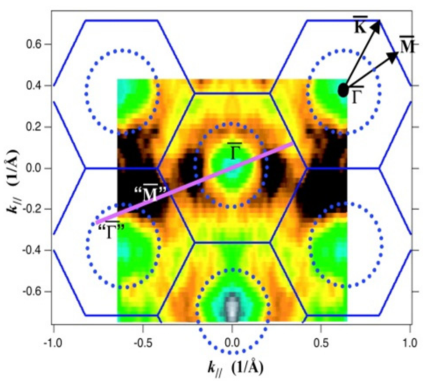
Nature 2000

Quantum Wells



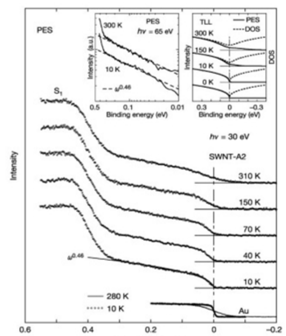
Nature 1999

C_{60}



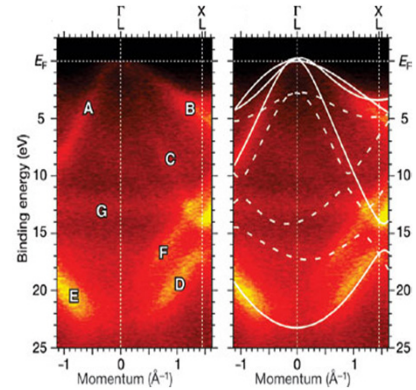
Science 2003

Nanotubes



Nature 2003

Diamond



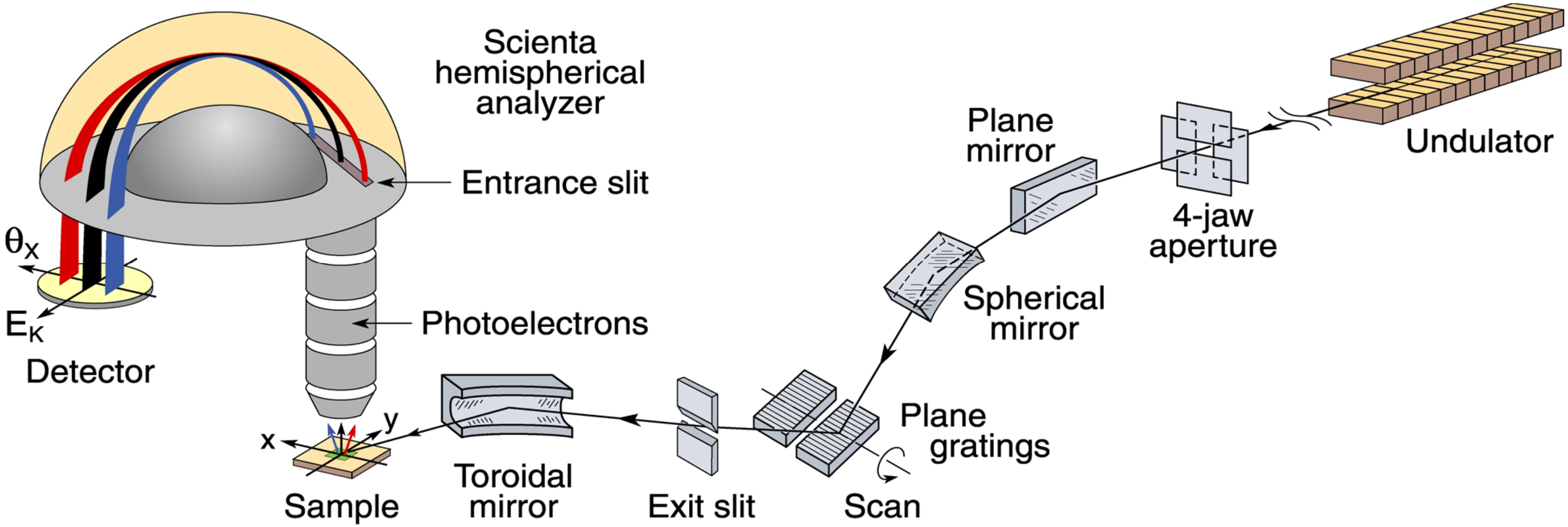
Nature 2005

Outline Part I

ARPES: Technique and developments

CUSO Lecture – Lausanne 02/2011

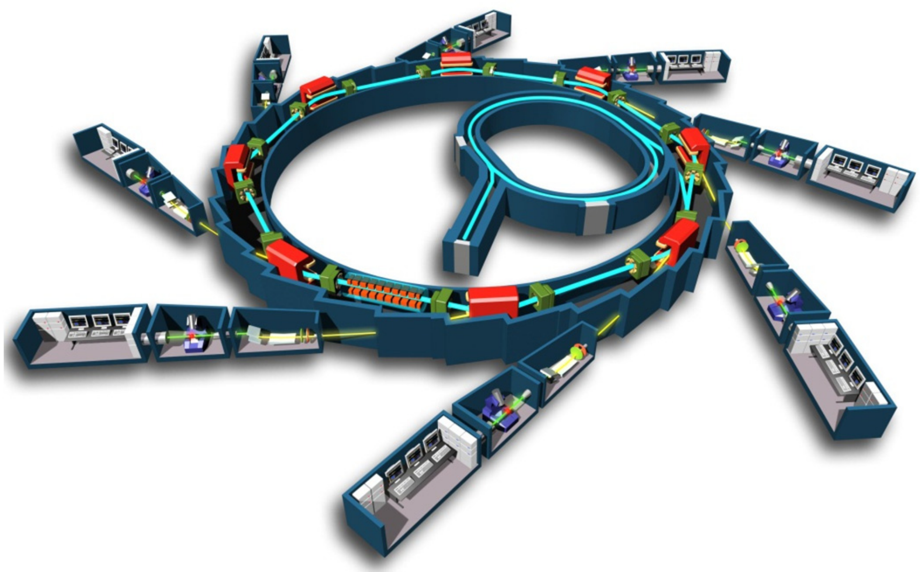
Angle-Resolved Photoemission Spectroscopy



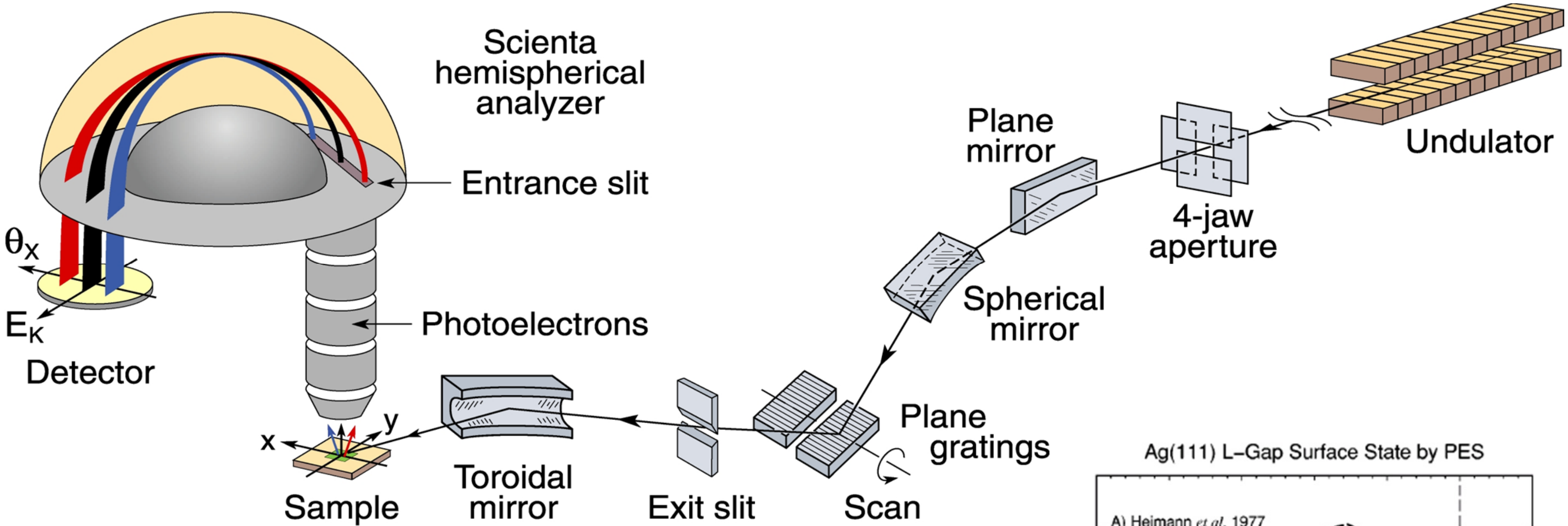
Parallel multi-angle recording

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

	ΔE (meV)	$\Delta\theta$
past	20-40	2°
now	1-10	0.2°



Angle-Resolved Photoemission Spectroscopy



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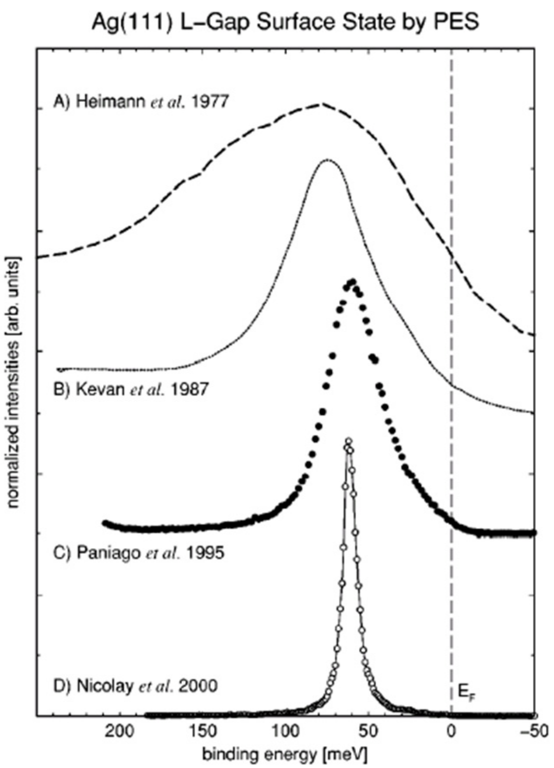
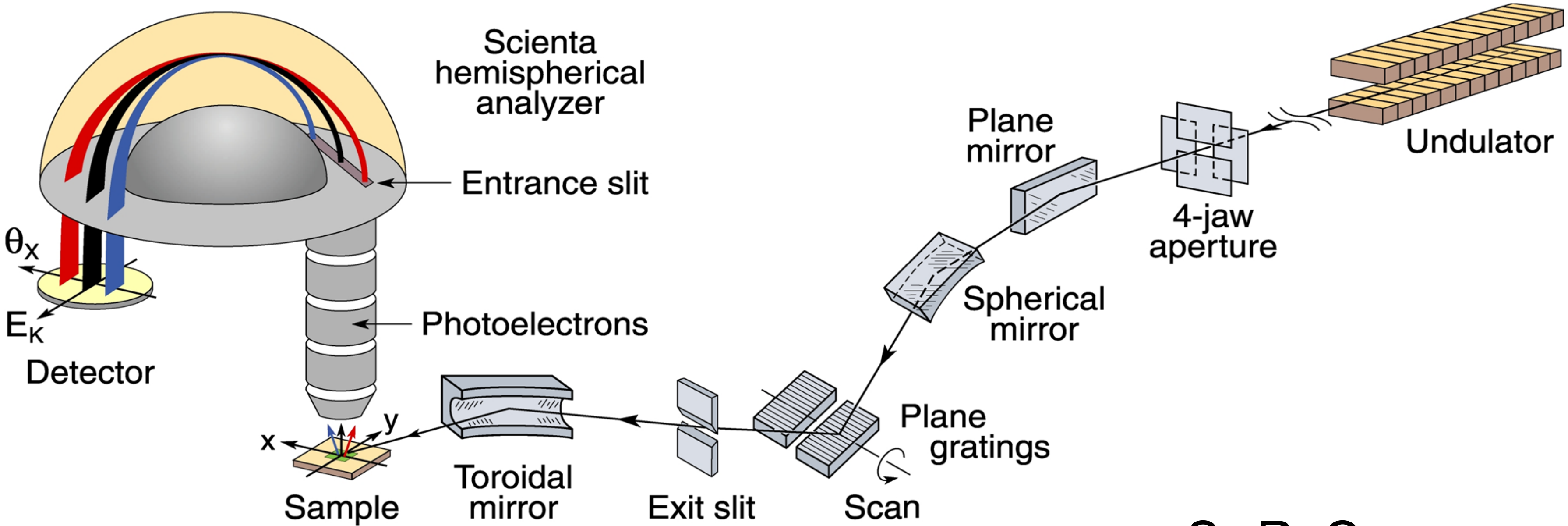
	ΔE (meV)	*Δθ*
past	*20-40*	*2°*
now	*1-10*	*0.2°*



Angle-Resolved Photoemission Spectroscopy



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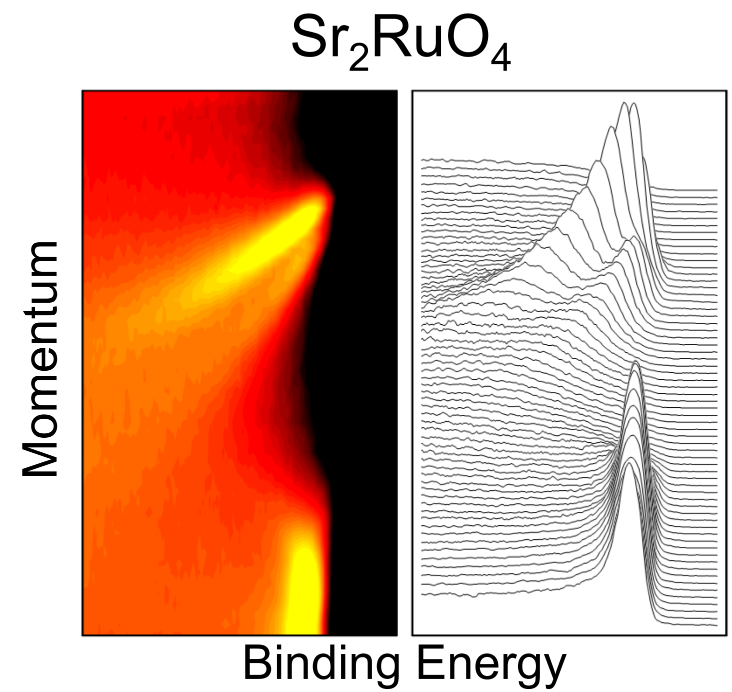
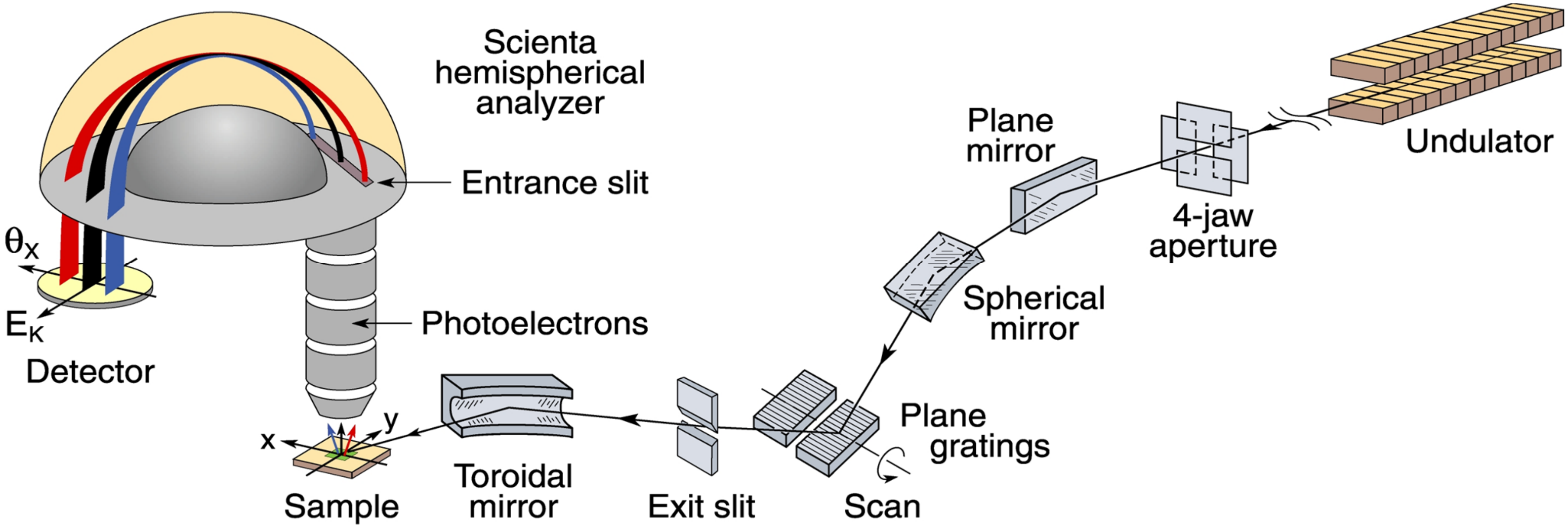


Angle-Resolved Photoemission Spectroscopy



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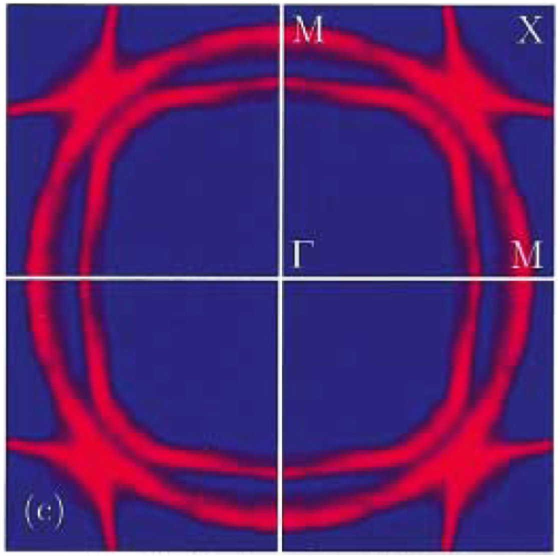
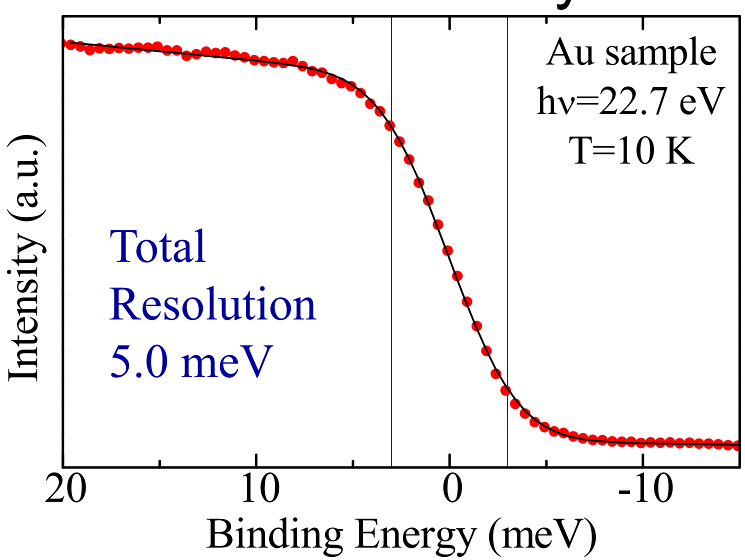


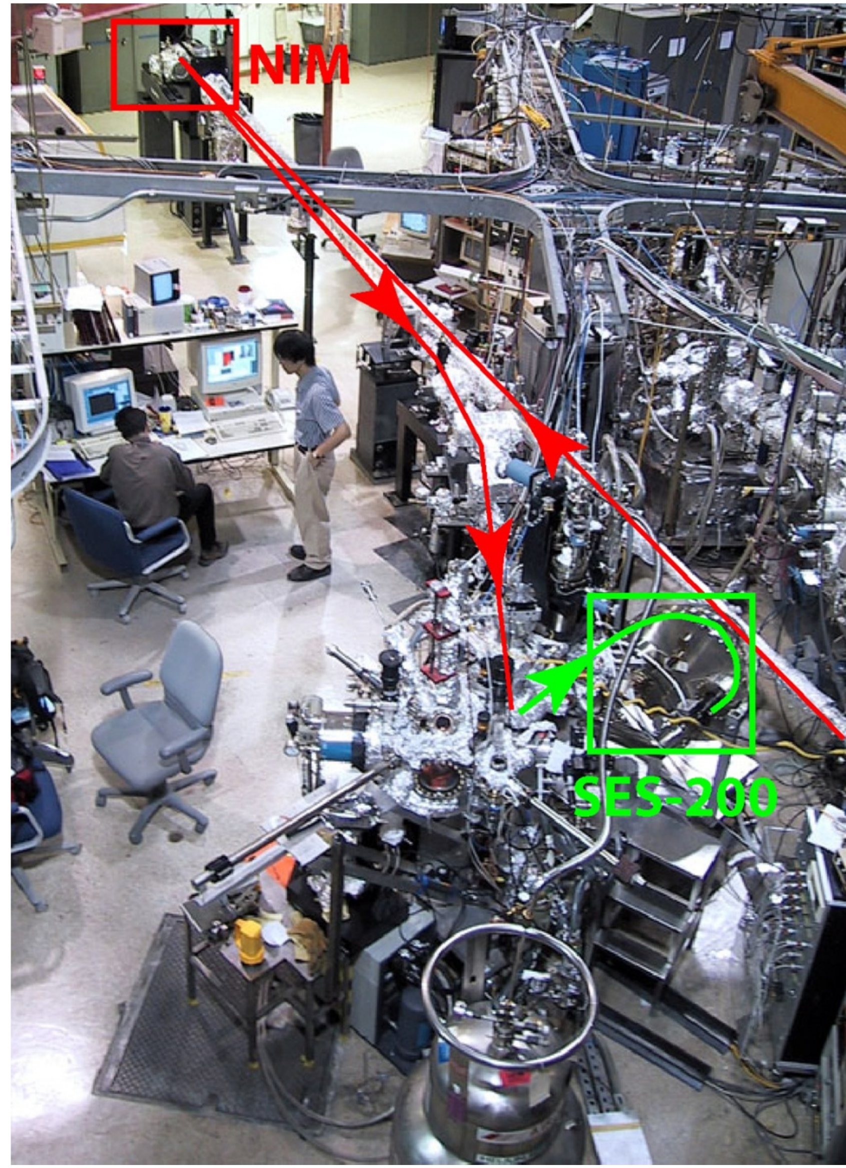
A. Damascelli *et al.*, PRL **85**, 5194 (2000)

Angle-Resolved Photoemission Spectroscopy

ΔE (meV)	*Δθ*
1-10	*0.2°*

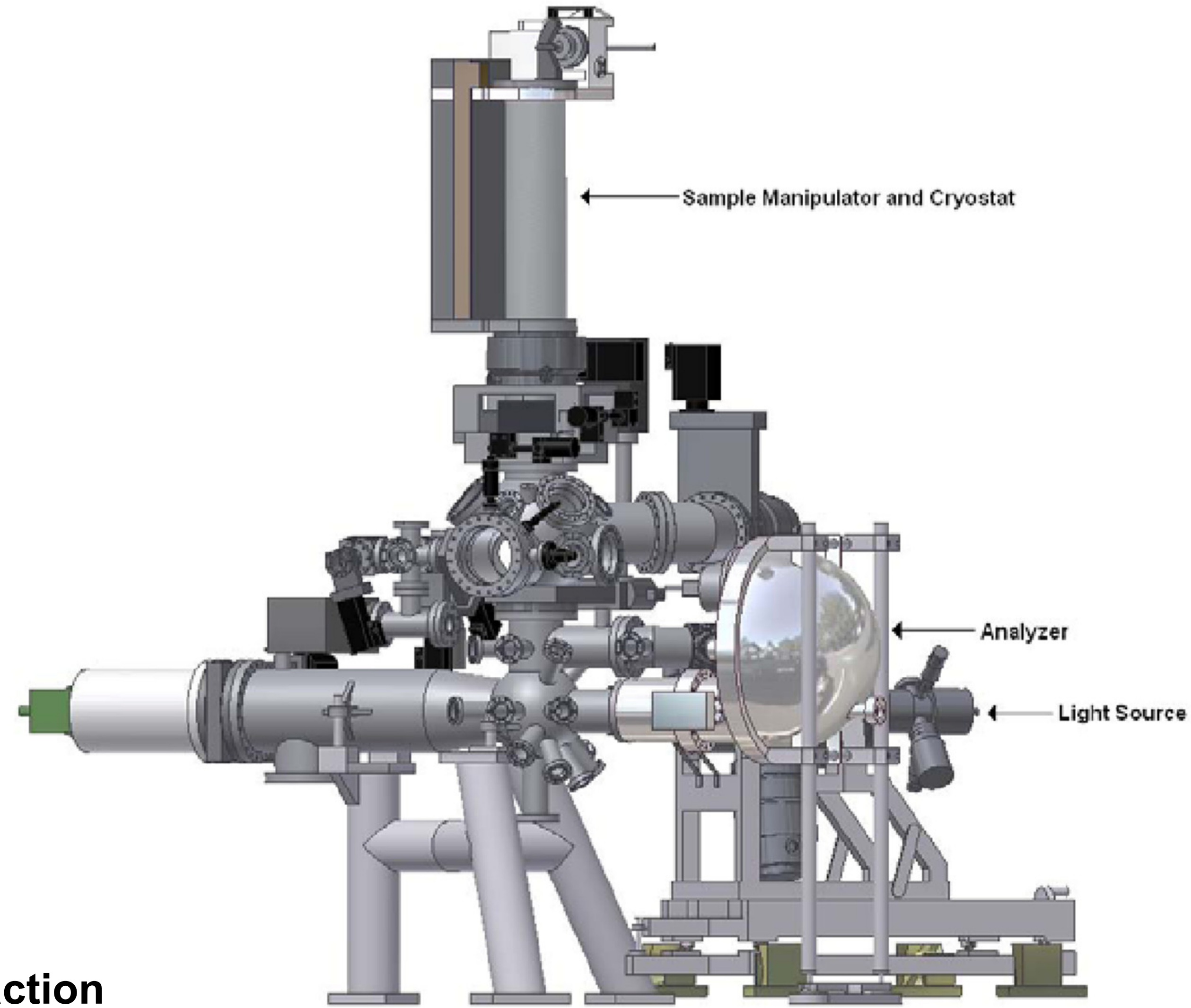
NIM/SCIENTA System





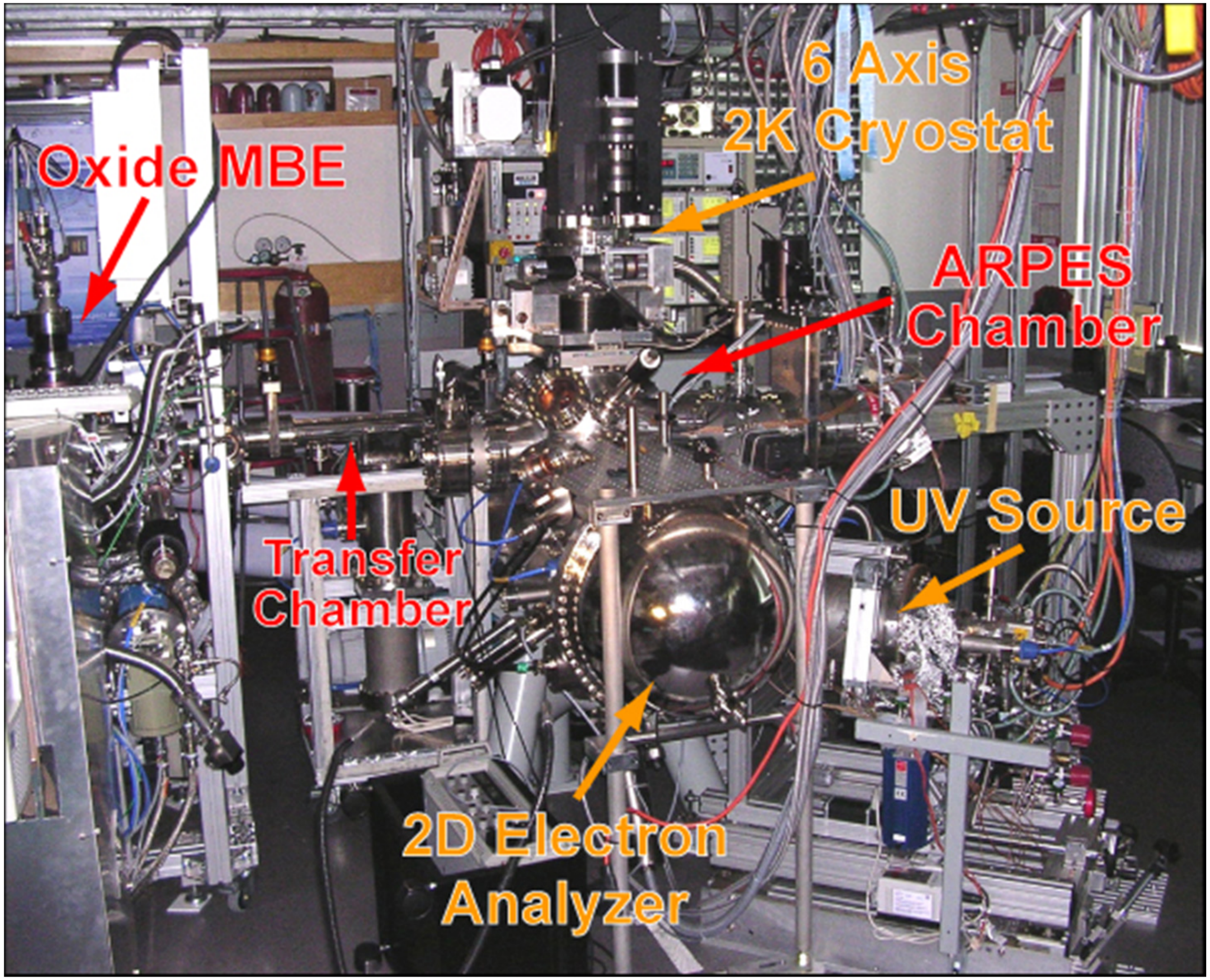
ARPES ON COMPLEX SYSTEMS

- High energy resolution
  $\Delta E < 1 meV$
- High angular precision
  $\pm 0.05^{o}$
- Low base temperature
  ~ 2 K
- Photon energies
  H_2, He, Ne
- Polarization control
  linear
- Ultra-high vacuum
  ~ 10^{-11} torr
- Surface / Thin films
- Low Energy Electron Diffraction



ARPES ON COMPLEX SYSTEMS

- **High energy resolution**
  ΔE<1meV
- **High angular precision**
  ± 0.05º
- **Low base temperature**
  ~ 2 K
- **Photon energies**
  H_2, He, Ne
- **Polarization control**
  linear
- **Ultra-high vacuum**
  ~ 10^{-11} torr
- **Surface / Thin films**
- **LEED**



ARPES ON COMPLEX SYSTEMS

- High energy resolution

  $\Delta E < 1 meV$

- High angular precision

  $\pm$ 0.05º

- Low base temperature

  ~ 2 K

- Photon energies

  H_2, He, Ne

- Polarization control

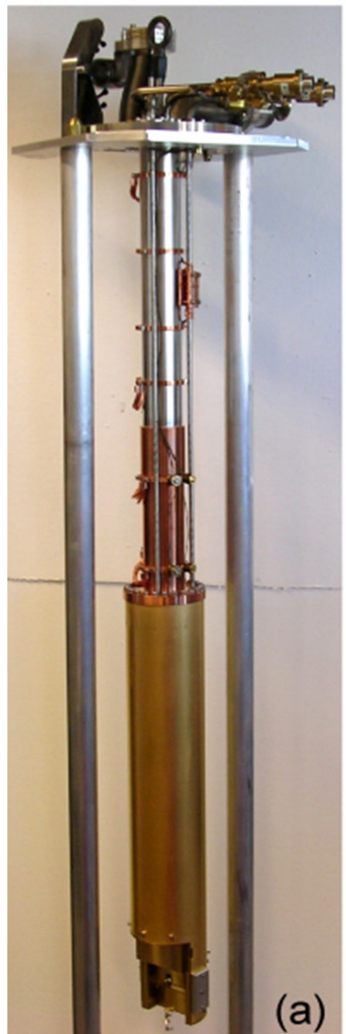
  linear

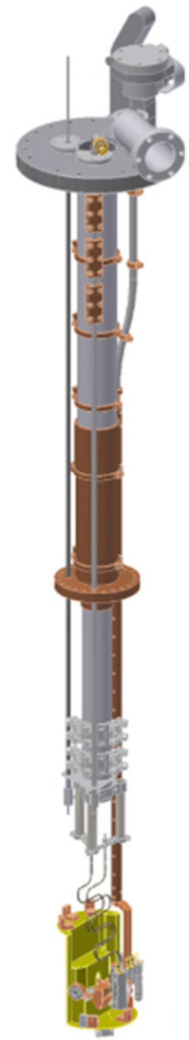
- Ultra-high vacuum

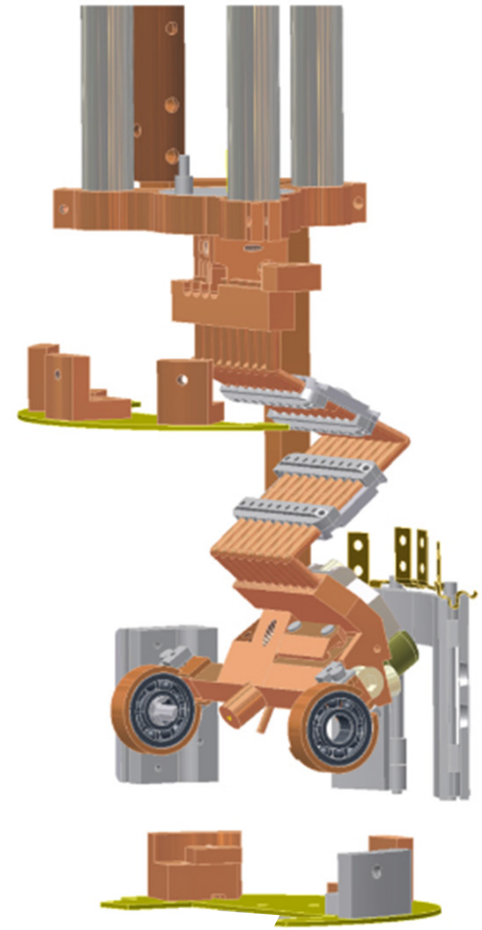
  ~ 10^{-11} torr

- Surface / Thin films

- LEED

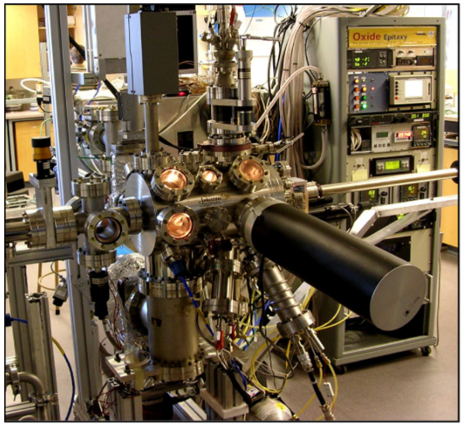


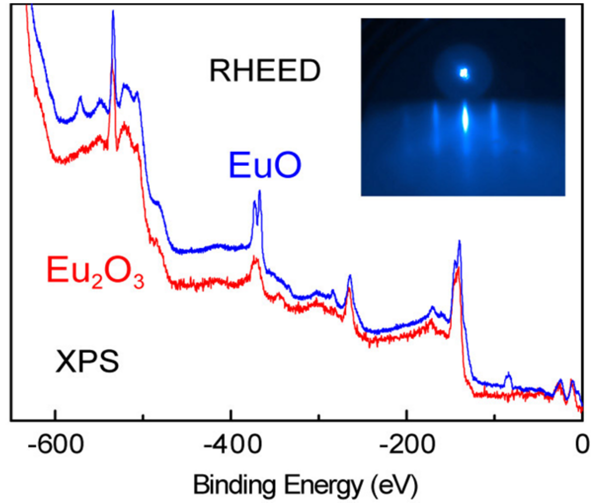




5 ½ axes manipulator
2.7 K on the sample

ARPES ON COMPLEX SYSTEMS

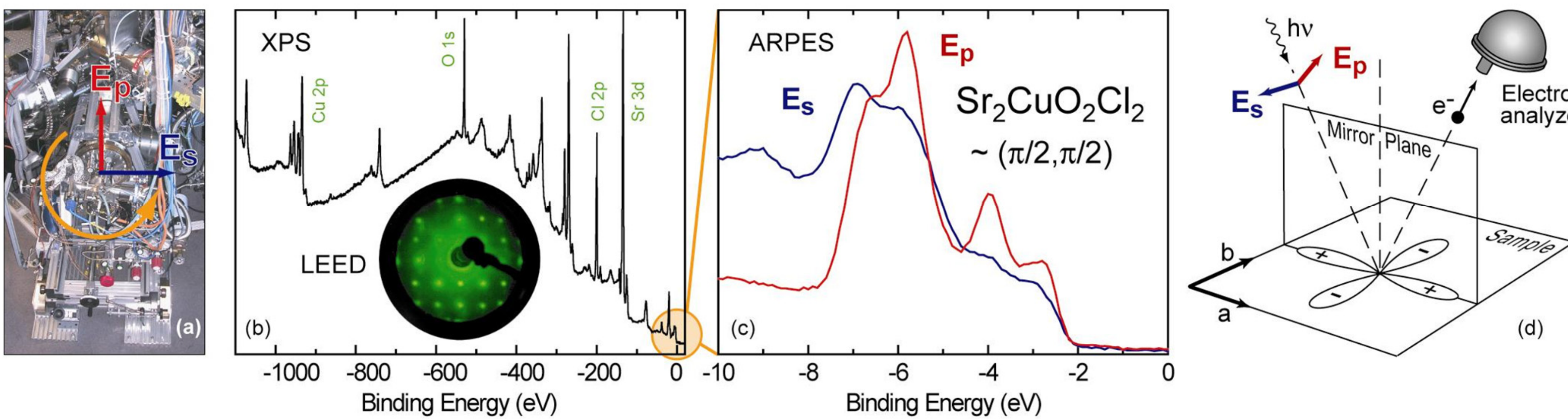




MBE Growth

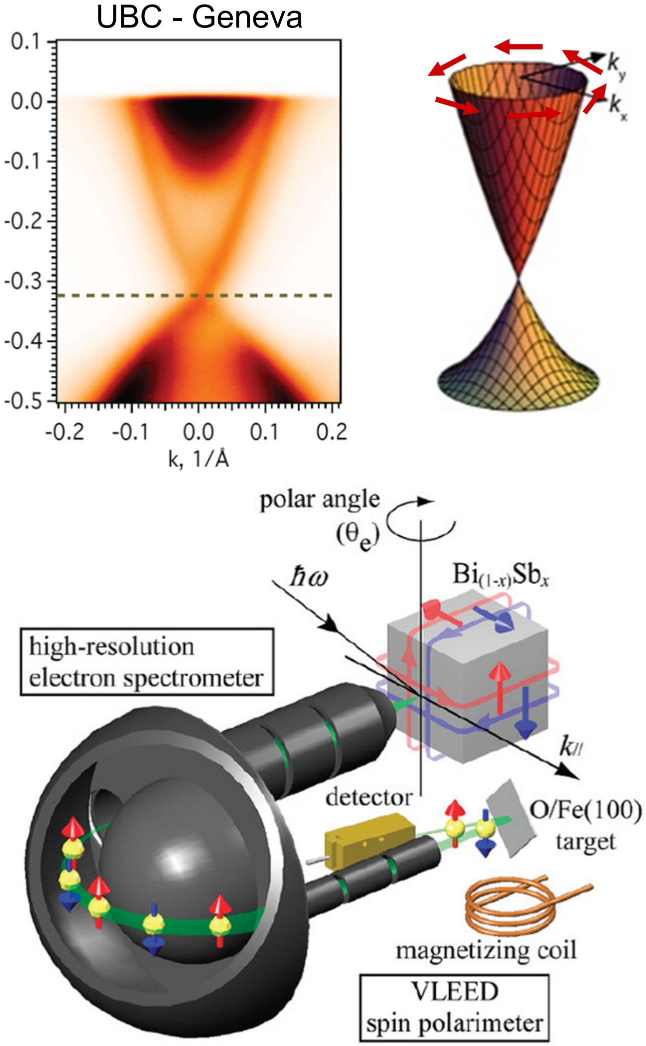
Characterization
Structure – chemistry

ARPES
Polarization - Energy



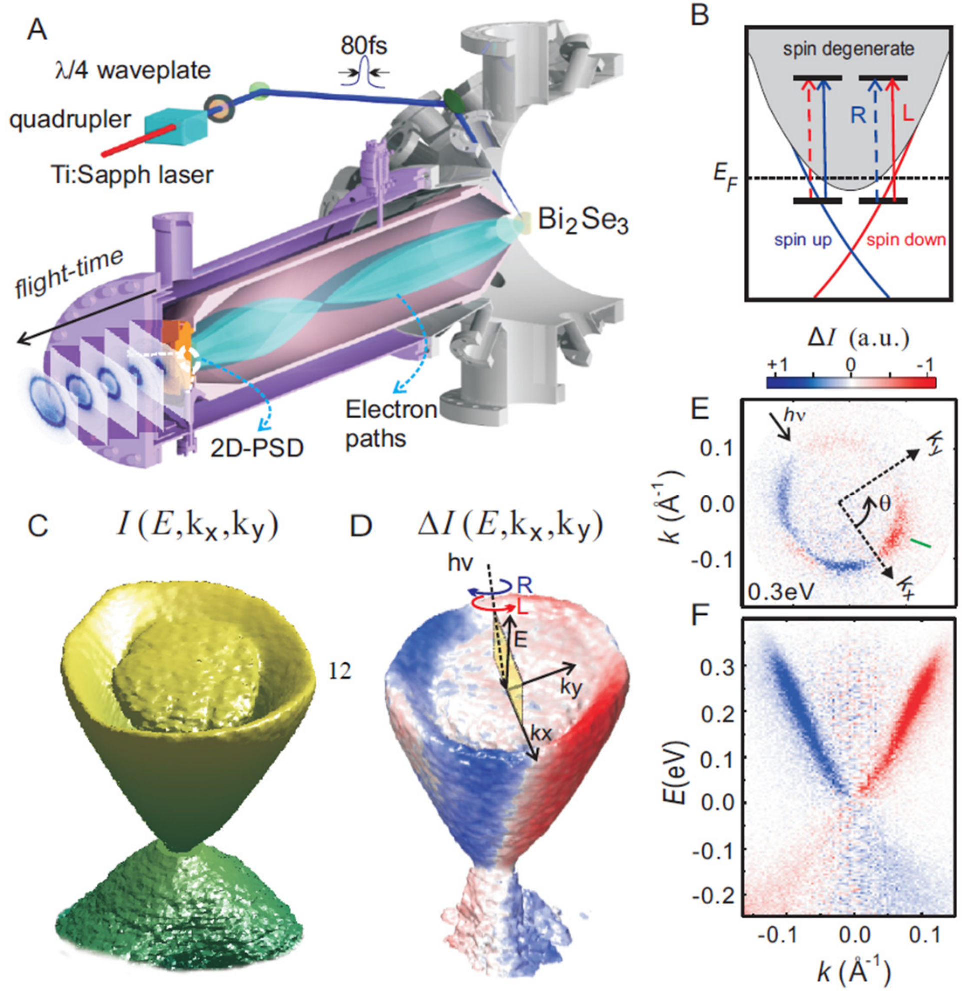
New Developments: ARPES + Spin + Time

ARPES+Spin polarimeter



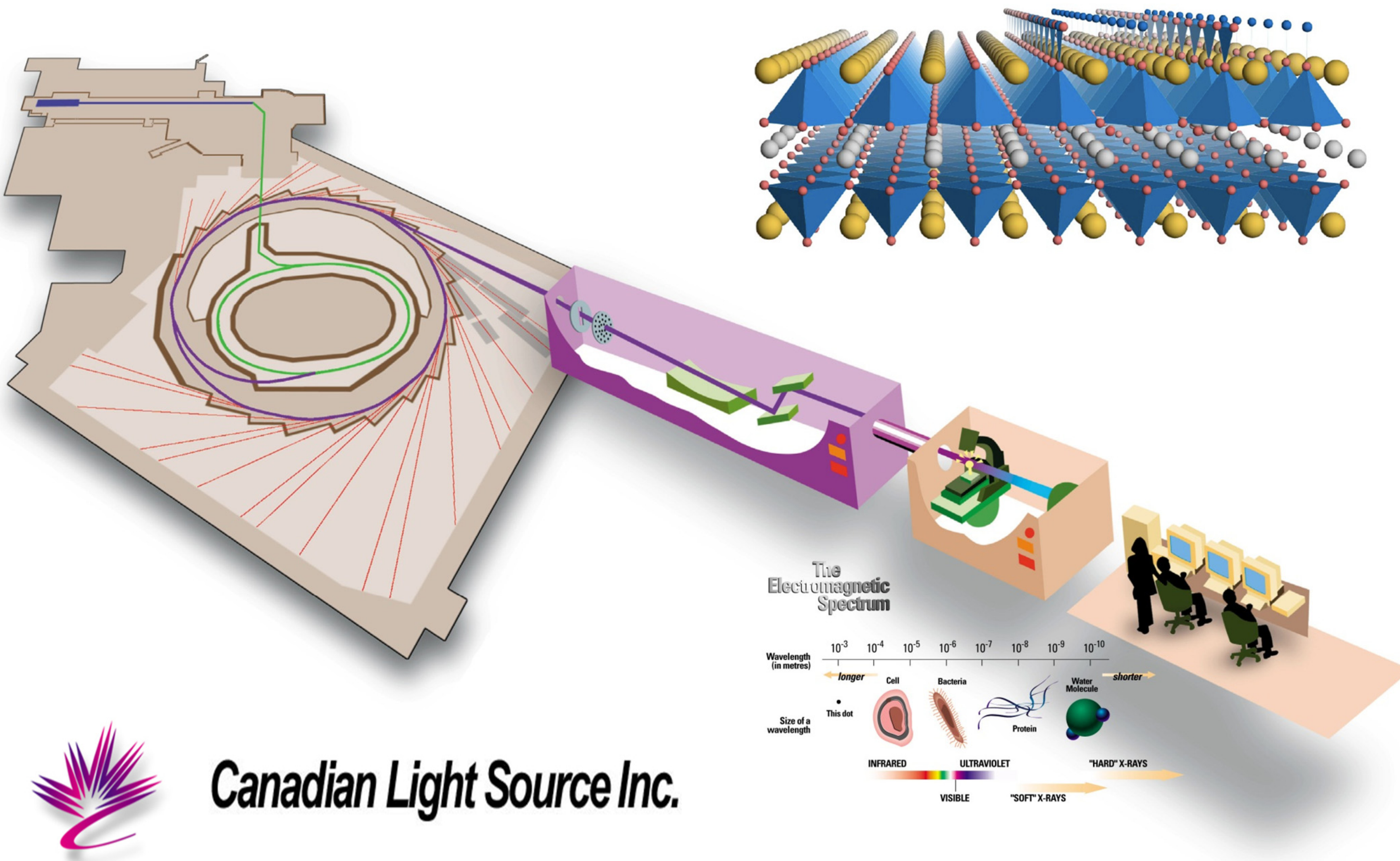
Nishide et a., New J. Phys. 12, 065011 (2010)

ARPES+Time of Fight



Wang et al., arXiv:1101.5636 (2011)

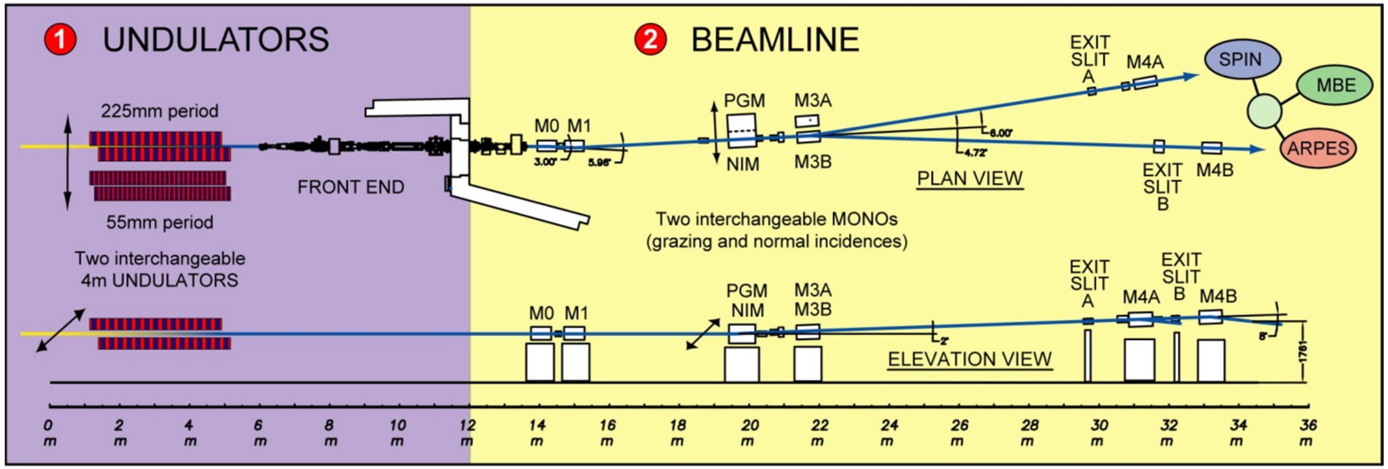
Quantum Materials Spectroscopy Center at CLS



Canadian Light Source Inc.

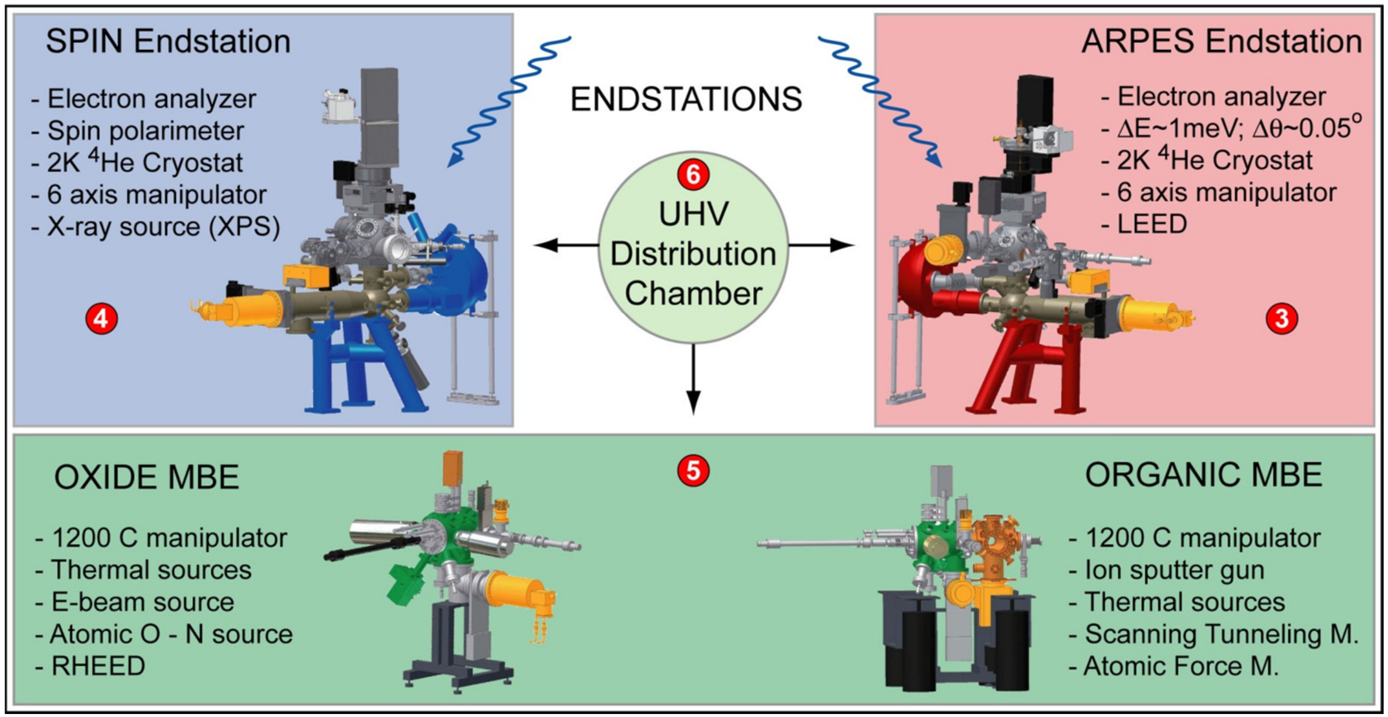
Quantum Materials Spectroscopy Center at CLS

- Broad energy range
- Polarization control
- Resolving power
- Maximum flux

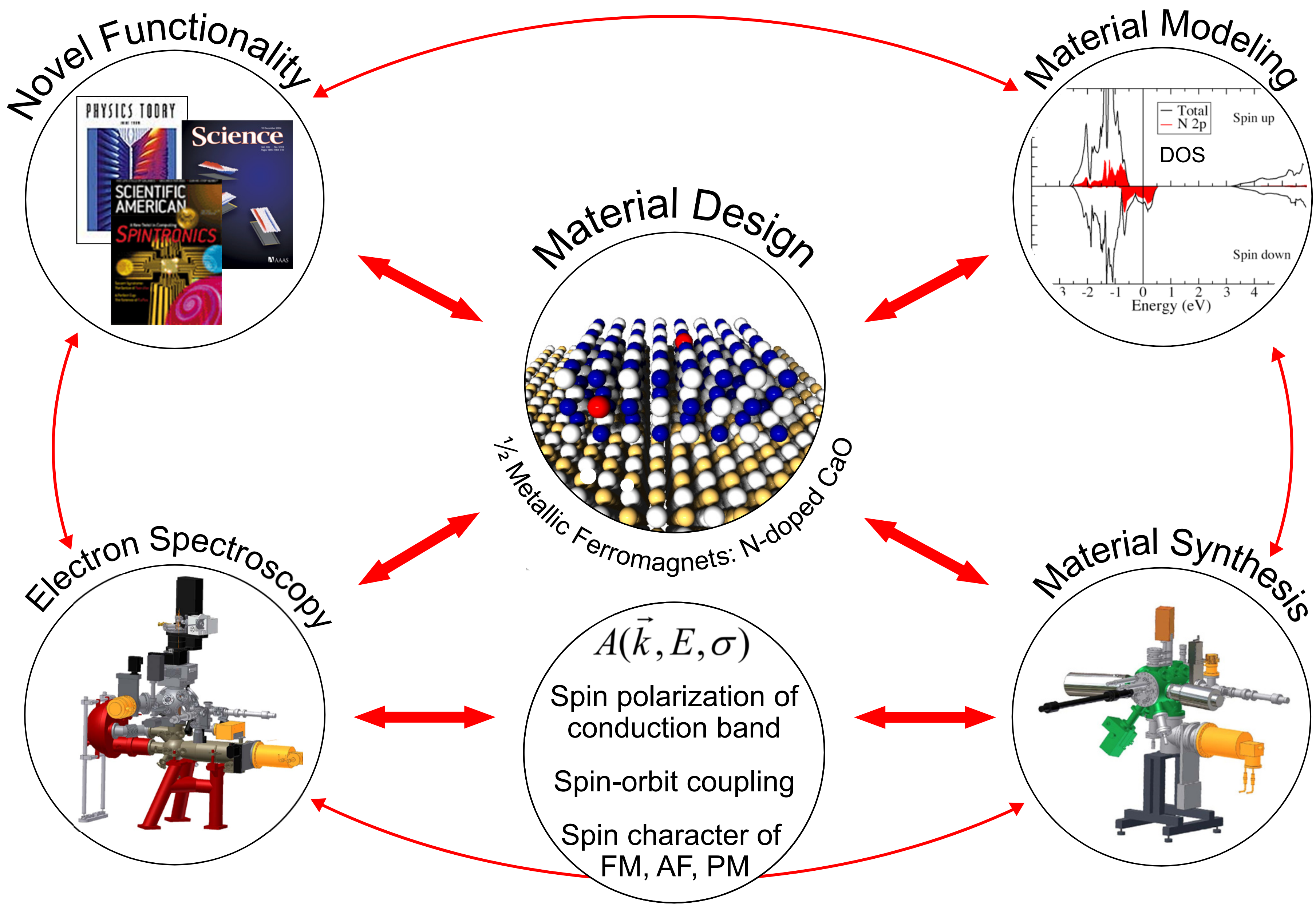


- High-res. ARPES
- Spin polarimeter
- Motion Accuracy
- Low Temperature

- Oxide MBE
- Organic MBE
- LEED / RHEED
- STM / AFM / XPS



Novel Complex Materials and Functionalities



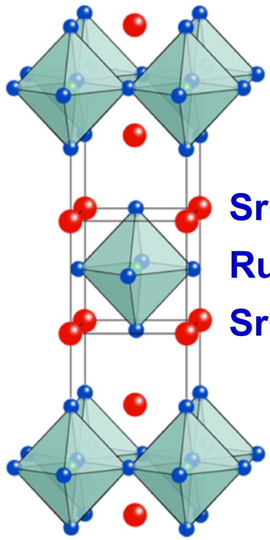
Outline Part I

Bulk, surface, and Fermi surfaces: Sr_2RuO_4

CUSO Lecture – Lausanne 02/2011

Sr_2RuO_4: basic properties

2D perovskite

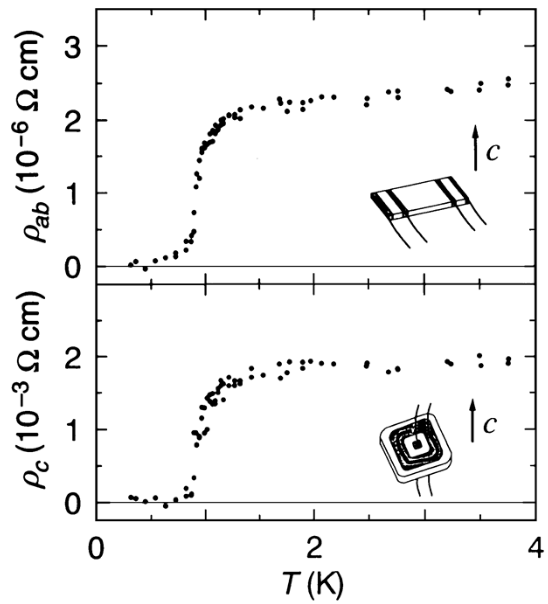


Unconventional superconductivity

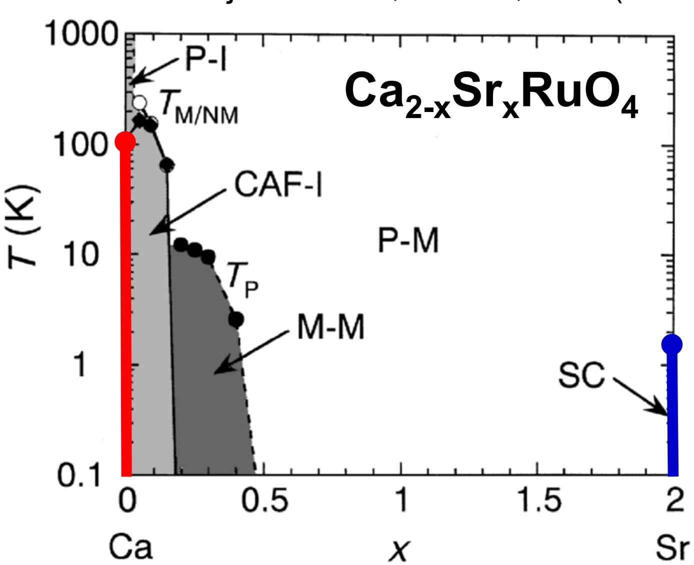
- Pairing mechanism ?
- Order parameter ?
- FM-AF fluctuations ?

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

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



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



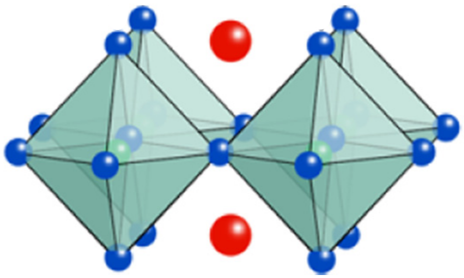
Lattice-magnetism interplay
Orbital degrees of freedom

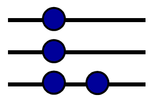
Sr_2RuO_4 : 2D **Fermi Liquid** (ρ_c/ρ_{ab}=850)

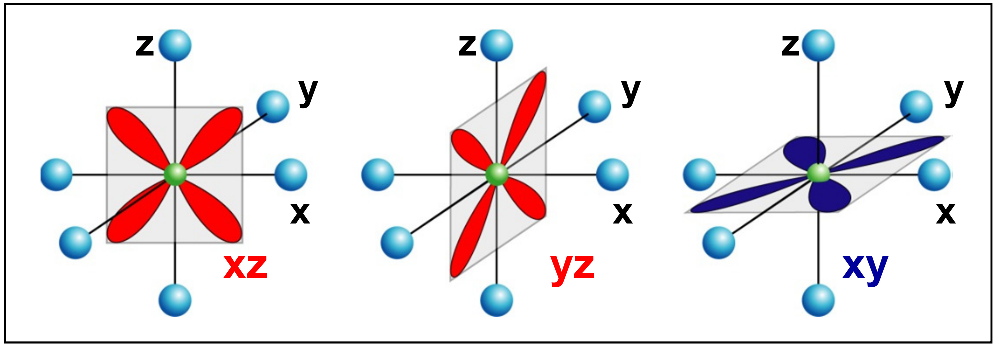
Ca_2RuO_4 : insulating **Anti-FerroMagnet**

$SrRuO_3$: metallic **FerroMagnet**

1D ($d_{xz,yz}$) versus 2D (d_{xy}) Superconductivity ?

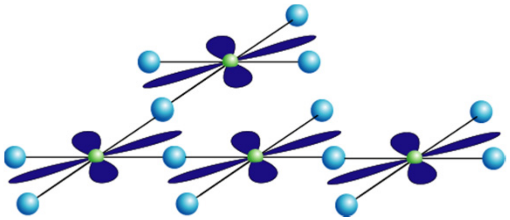


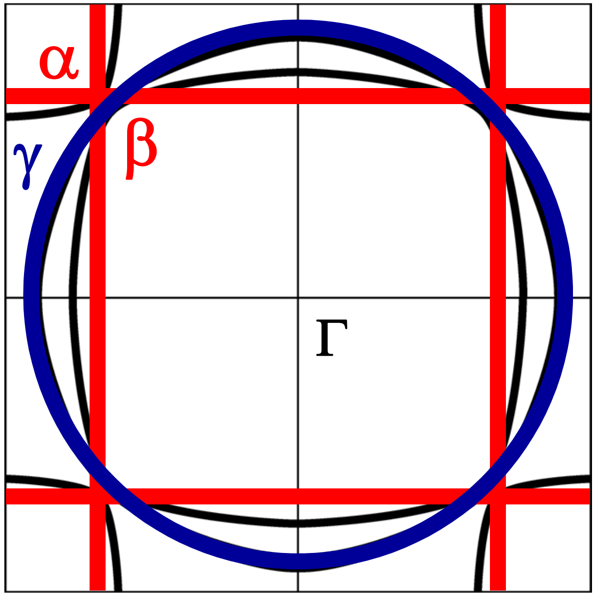




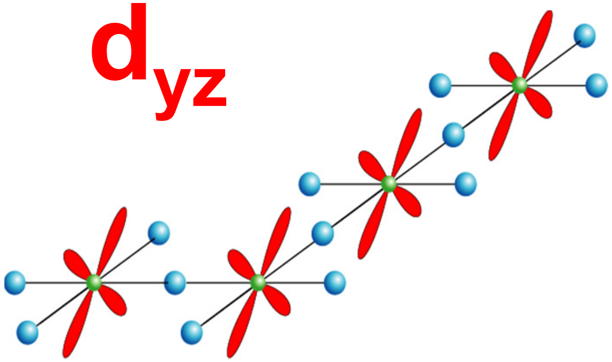
▶ Band structure calculation: **3 t_{2g}** bands crossing $\mathbf{E_F}$

d_{xy}





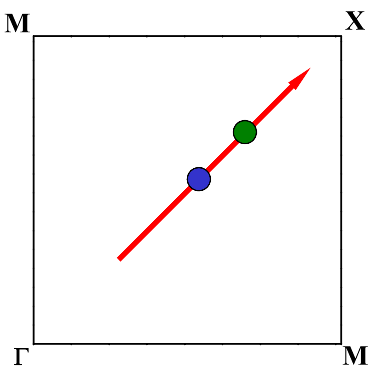
d_{yz}



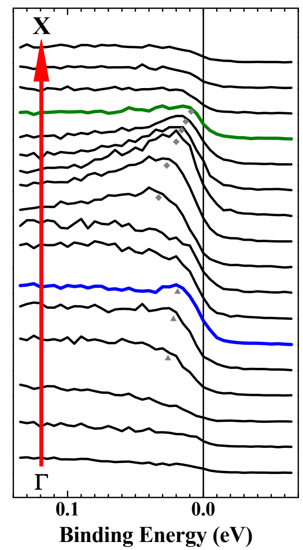
Mazin *et al.*, PRL **79**, 733 (1997)

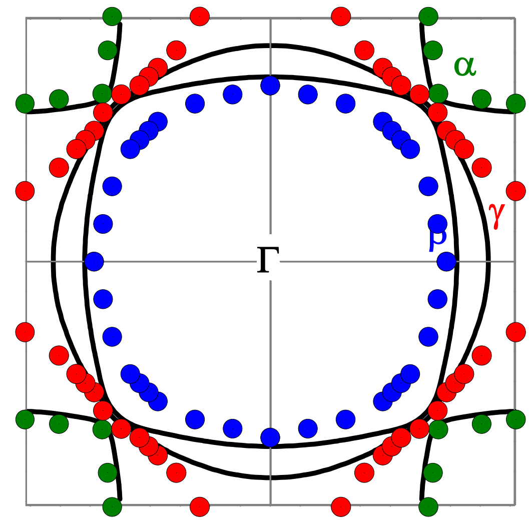
Fermi Surface Topology of Sr_2RuO_4

ARPES : circa 1996



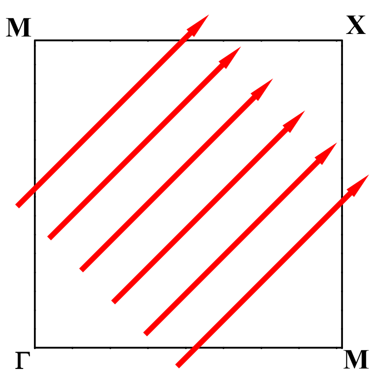
D.H. Lu *et al.*, PRL **76**, 4845 (1996)



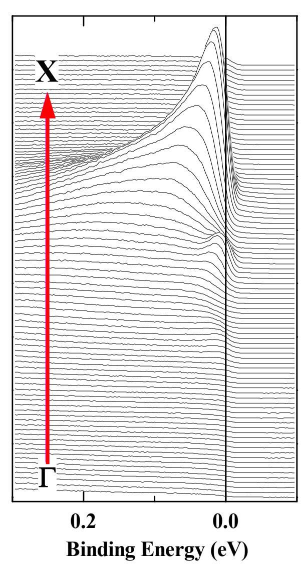


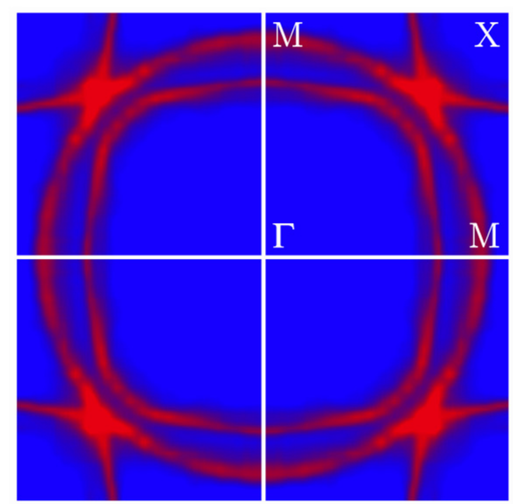
D.J. Singh, PRB **52**, 1358 (1995)

ARPES : present day



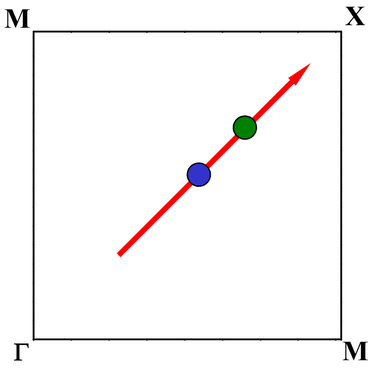
A. Damascelli *et al.*, PRL **85**, 5194 (2000)



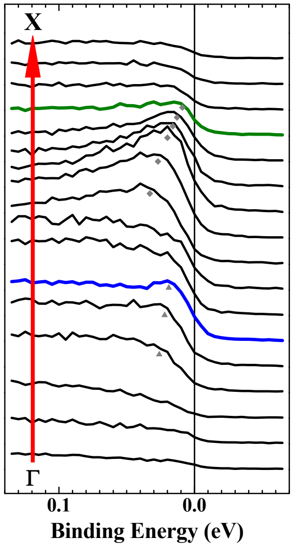


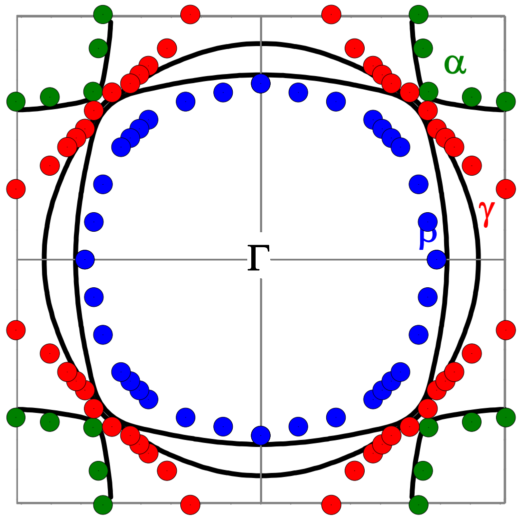
Fermi Surface Topology of Sr_2RuO_4

ARPES : circa 1996



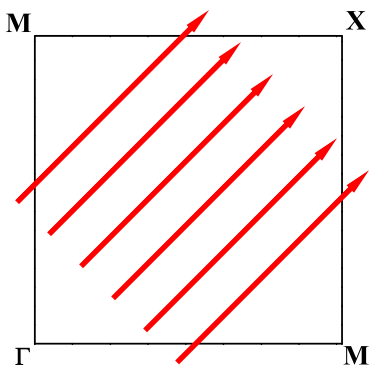
D.H. Lu *et al.*, PRL **76**, 4845 (1996)





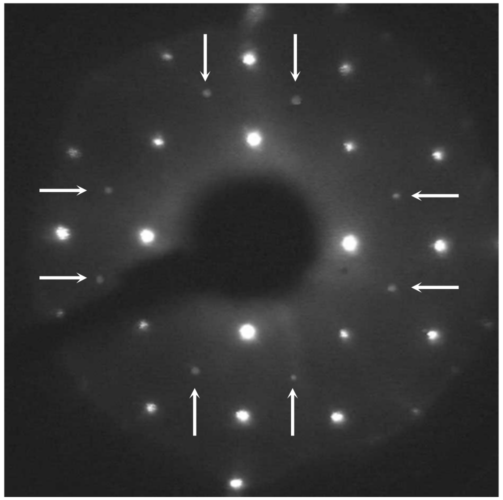
D.J. Singh, PRB **52**, 1358 (1995)

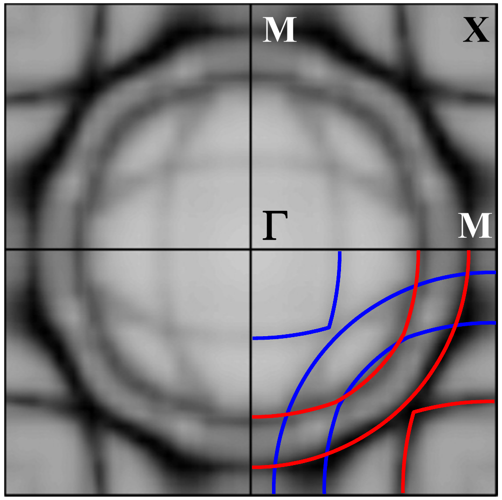
ARPES : present day



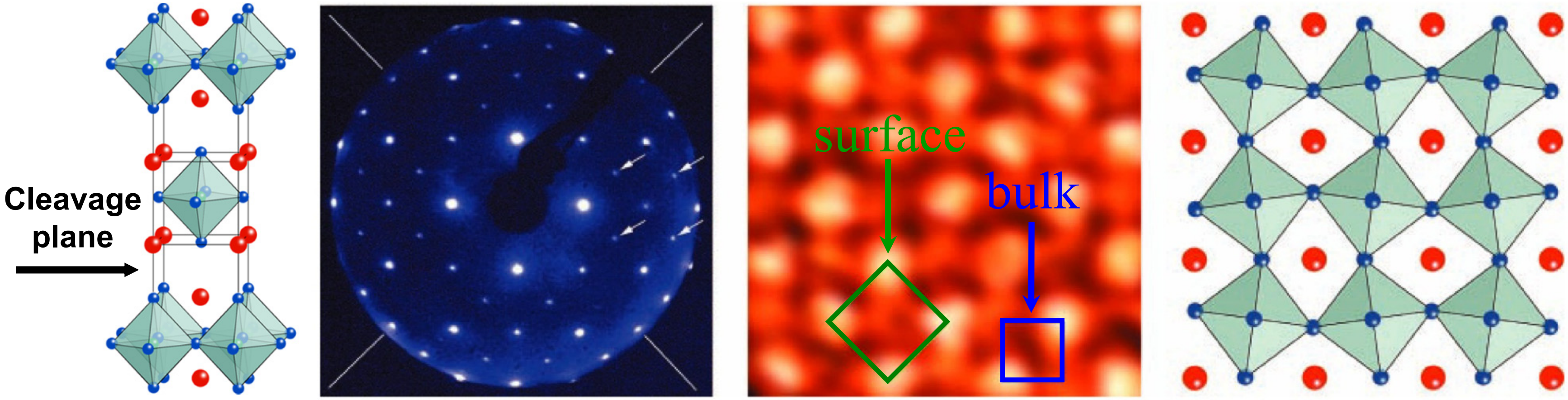
A. Damascelli *et al.*, PRL **85**, 5194 (2000)

Surface instability ⟷ **Band folding**





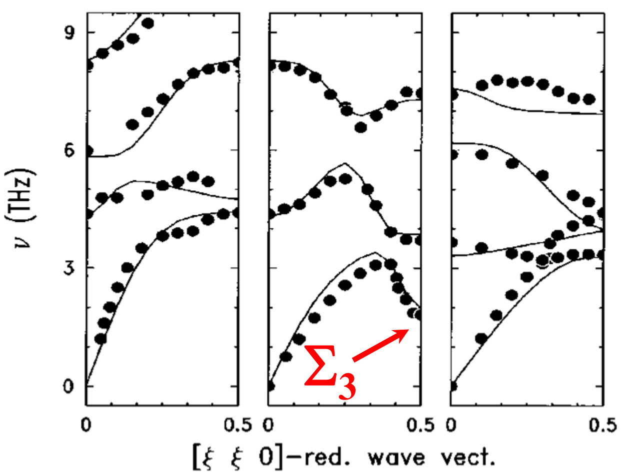
Surface reconstruction of cleaved Sr_2RuO_4



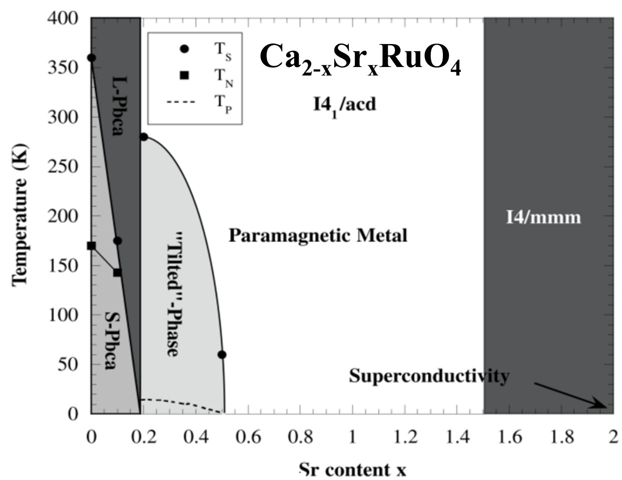
R. Matzdorf *et al.*, Science **289**, 746 (2000)

Rotation of the RuO_6 octahedra around the c axis

Soft S_3 phonon branch → Rotation instability of $Ca_{2-x}Sr_xRuO_4$



M. Braden *et al.*, PRB **57**, 1236 (1998)



O. Friedt *et al.*, PRB **63**, 174432 (2001)

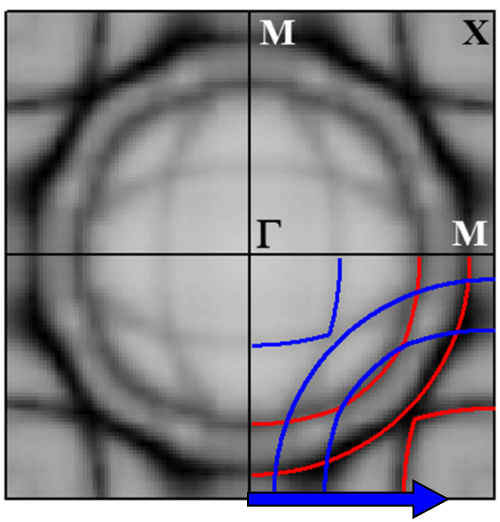
Surface electronic structure of Sr_2RuO_4

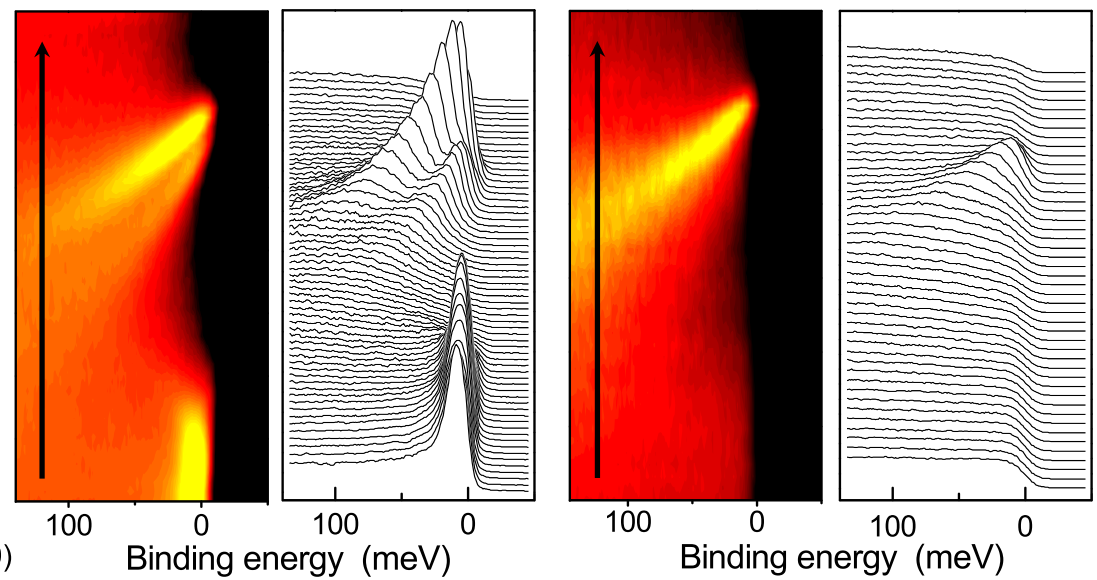
On samples cleaved at **180 K** the **surface**-related features are **suppressed**

E_F mapping ±10 meV

Cold cleave T=10 K

Hot cleave T=180 K



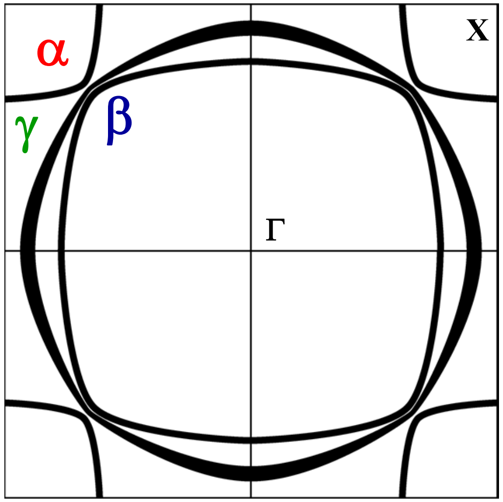


A. Damascelli *et al.*, PRL **85**, 5194 (2000)

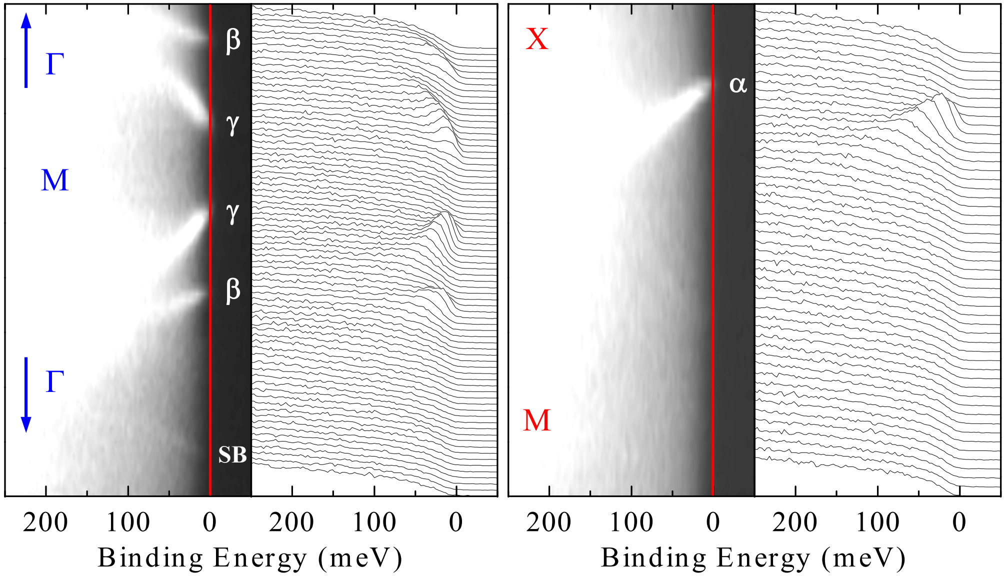
Bulk electronic structure of Sr_2RuO_4

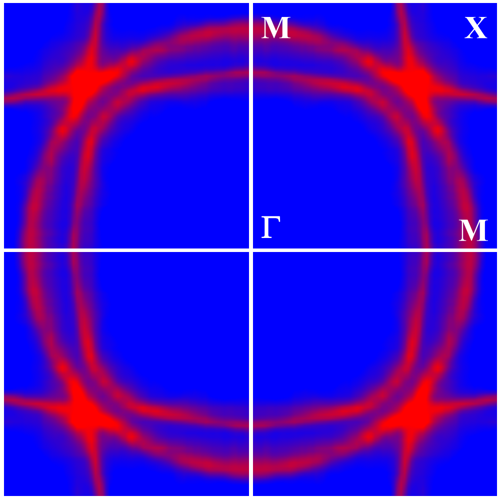
What do we learn about the bulk electronic structure?

- **FS topology**
- **Fermi velocity**
- **Effective mass**



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





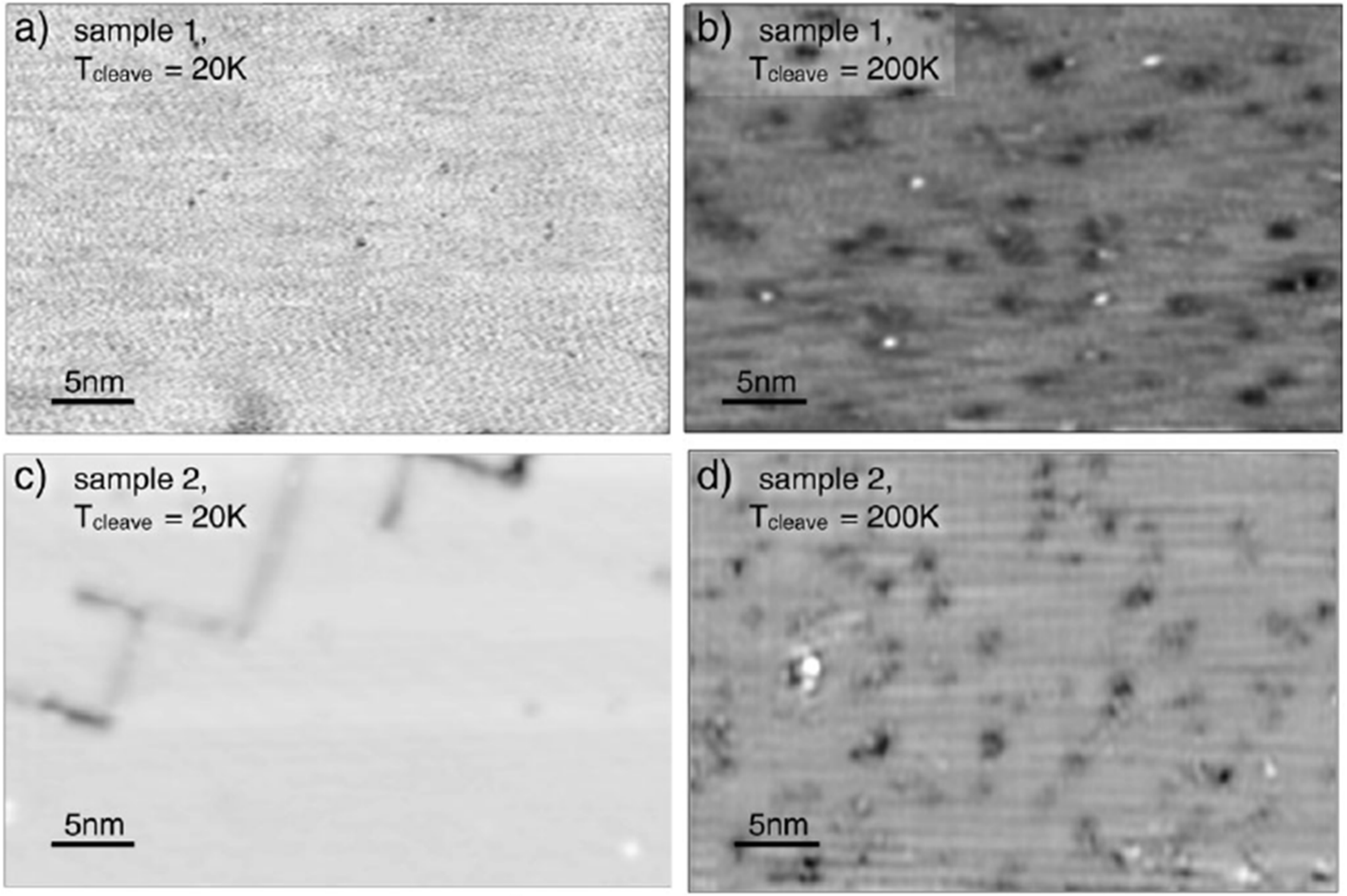
Sr_2RuO_4 cleaved at 180 K

T= 10 K hν=28 eV

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

Cleaving-Temperature Dependence of Sr_2RuO_4 Surfaces

Temperature dependent STM



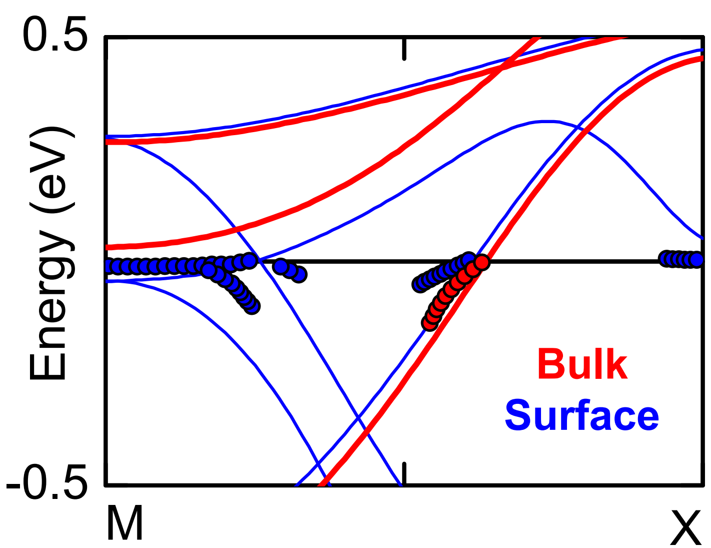
~ 0.002 defects/nm^2 $\quad$ 0.056 ± 0.01 defects/nm^2

Y. Pennec, N.J.C. Ingle, I.S. Elfimov, E. Varene, Y. Maeno, A. Damascelli, J.V. Barth, PRL **101**, 216103 (2008)

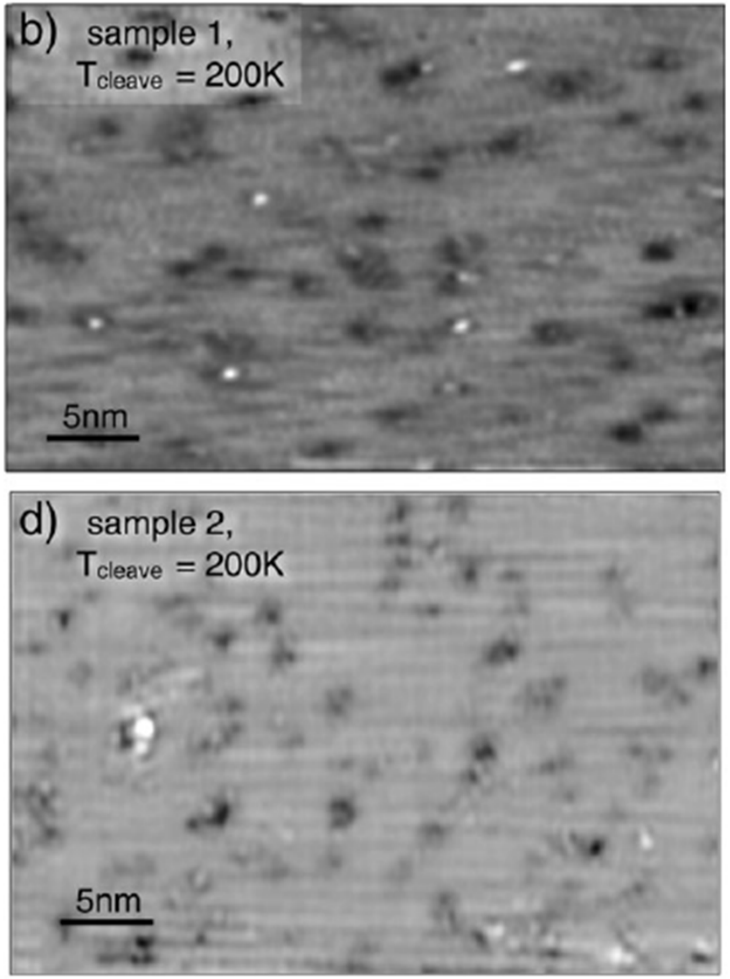
Cleaving-Temperature Dependence of Sr_2RuO_4 Surfaces

Temperature dependent STM

Larger scattering for surface ARPES features



K.M. Shen, A. Damascelli *et al.*, PRB **64**, 180502(R) (2001)

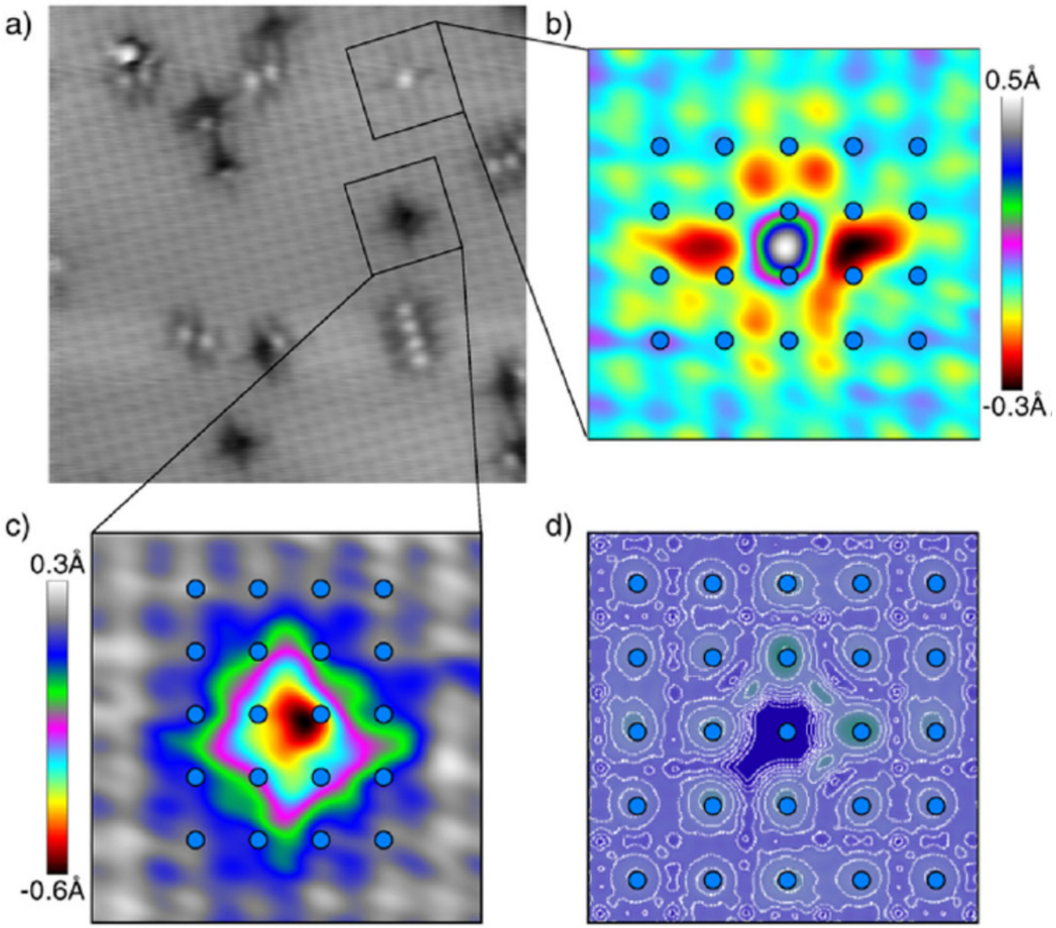


0.056 ± 0.01 defects/nm^2

Y. Pennec, N.J.C. Ingle, I.S. Elfimov, E. Varene, Y. Maeno, A. Damascelli, J.V. Barth, PRL **101**, 216103 (2008)

Cleaving-Temperature Dependence of Sr_2RuO_4 Surfaces

Temperature dependent STM



Two types of defects:

(b) Protrusion

(c) Hole

Blue dots: Sr locations on the SrO surface termination.

(d) Charge density:

DFT calculated contour at a height of 2.13 Å, for a missing charge neutral SrO molecule.

Y. Pennec, N.J.C. Ingle, I.S. Elfimov, E. Varene, Y. Maeno, A. Damascelli, J.V. Barth, PRL **101**, 216103 (2008)

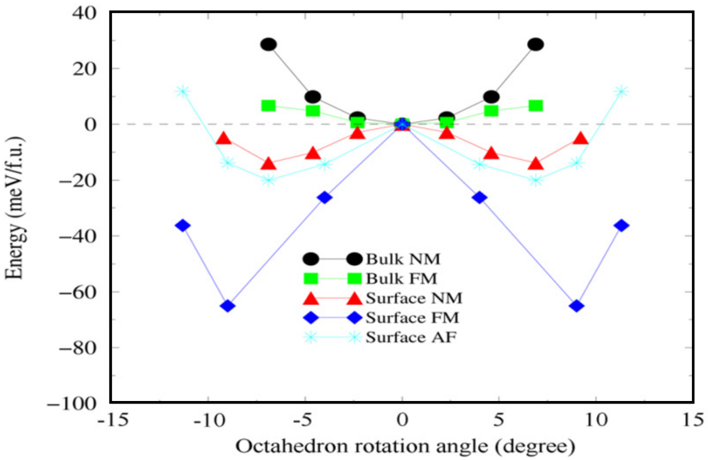
Surface Ferromagnetism in Sr_2RuO_4?

Surface Reconstruction ⟷ Surface Ferromagnetism

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

First principle calculations

- **NM surface**: rotation of RuO_6 (R=6.5º)
- **FM surface**: R= 9.0º, ΔE=-51meV
- GGA favors **FM** in the **bulk** (4meV)



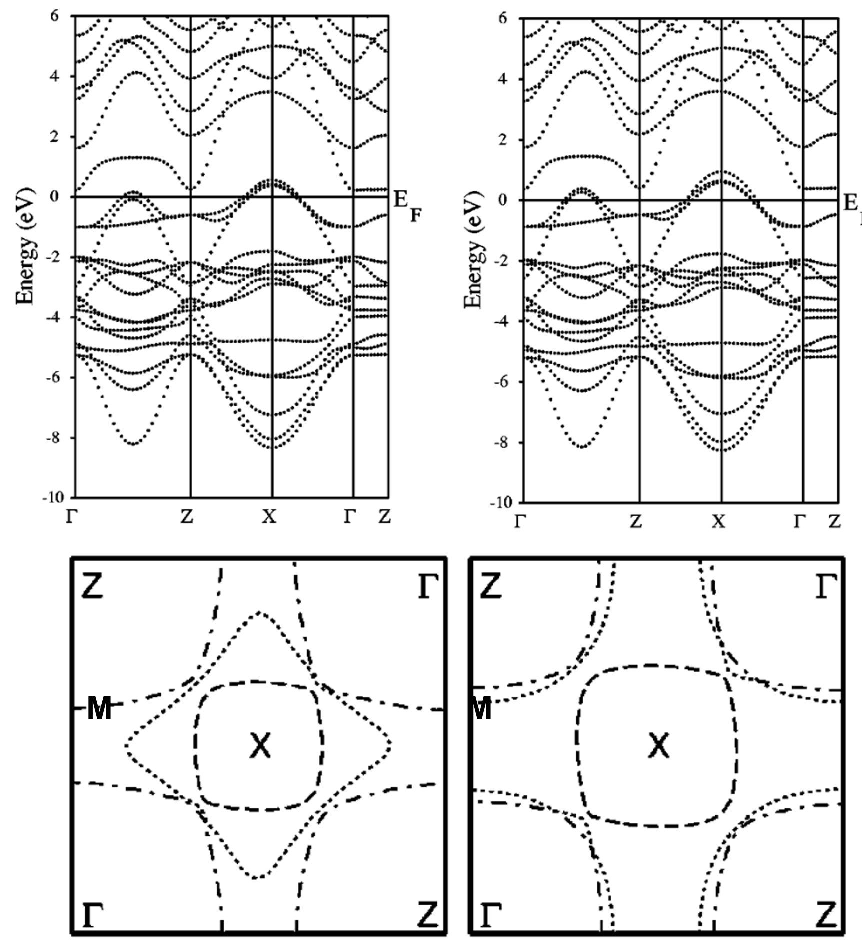
FM surface

Exchange splitting: **500 meV**

Magnetic moment: **1.0 μ_B/ Ru**

Z. Fang, K. Terakura, PRB **64**, 20509 (2001)

Majority spin channel **Minority spin channel**



P.K. de Boer *et al.*, PRB **59**, 9894 (1999)

Superconductivity at the Surface ?

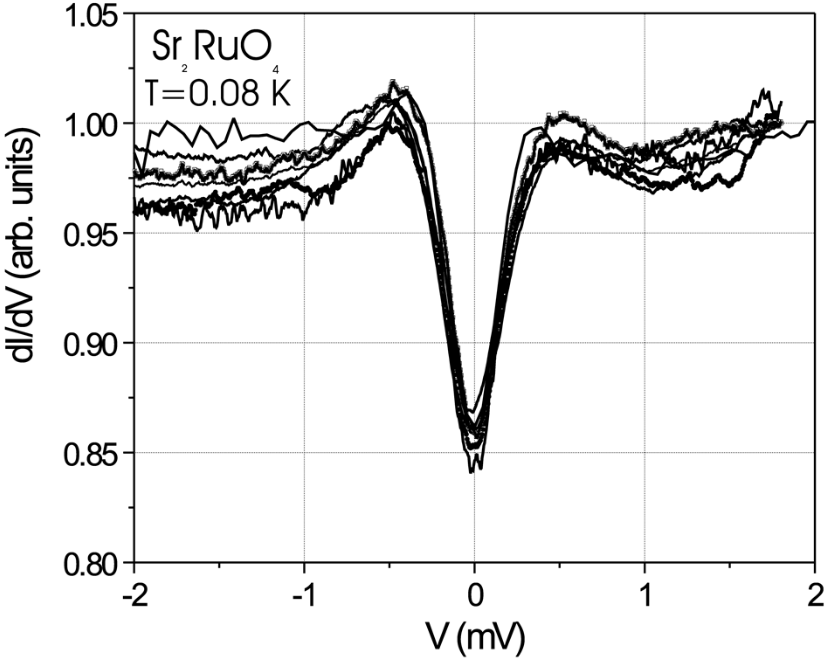
STM results

M.D. Upward *et al.*, PRB **65**, 220512 (2002)

DOS suppression within 500 μV

Gap closes for T>1.5K ; B>700G

$2\Delta_{max} / kT_C \sim 8.0$



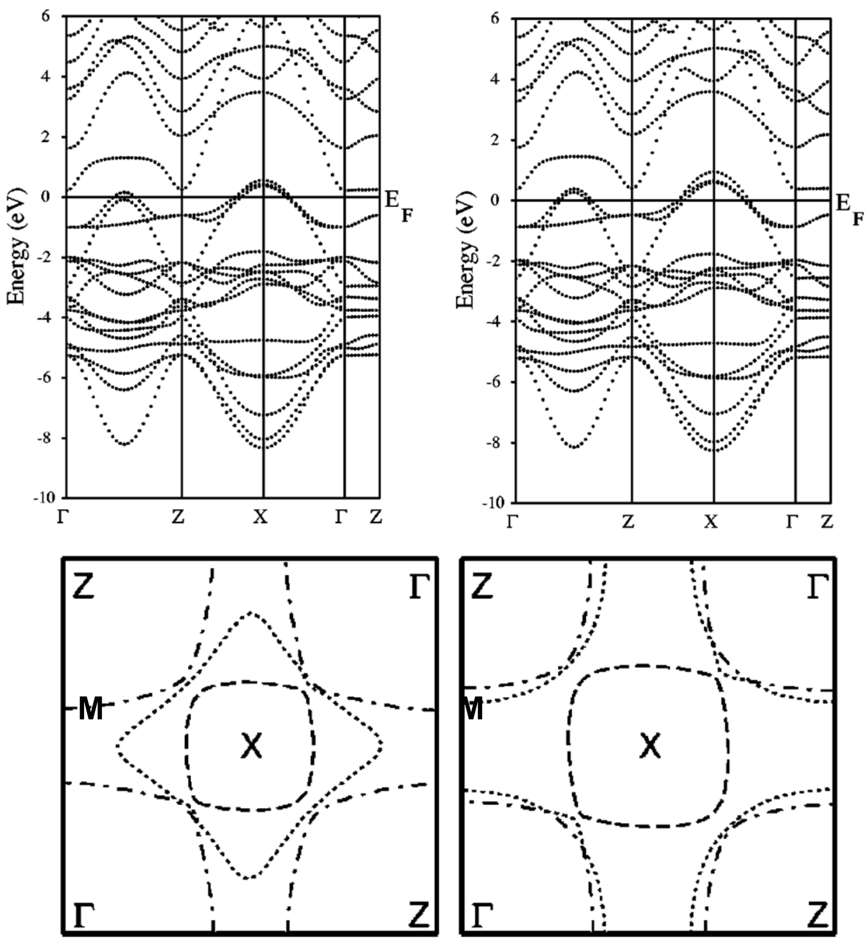
▶ Opening of a SC gap ◀

Surface Ferromagnetism in Sr_2RuO_4?

Surface Reconstruction ⟷ Surface Ferromagnetism

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

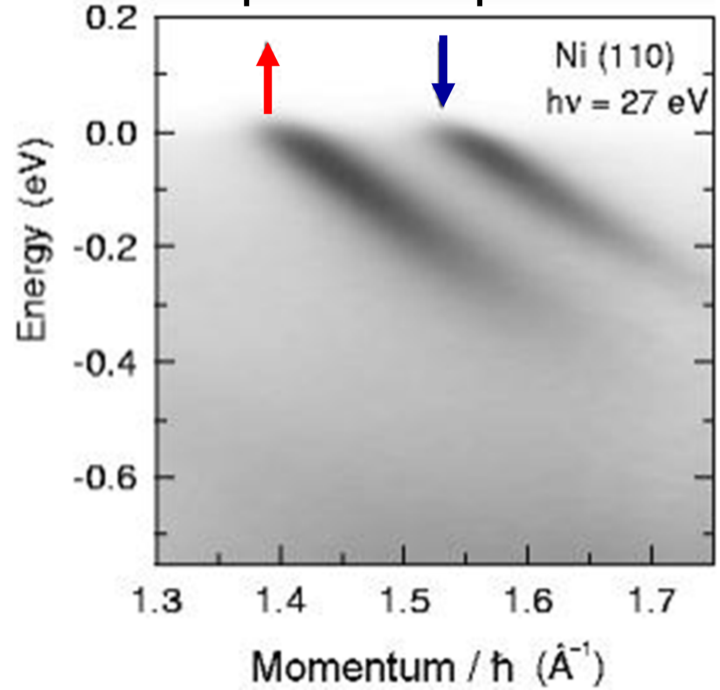
Majority spin channel **Minority spin channel**



P.K. de Boer *et al.*, PRB **59**, 9894 (1999)

Spin-split Fermi-level crossings of the conduction band in Ni

Majority spins **Minority spins**

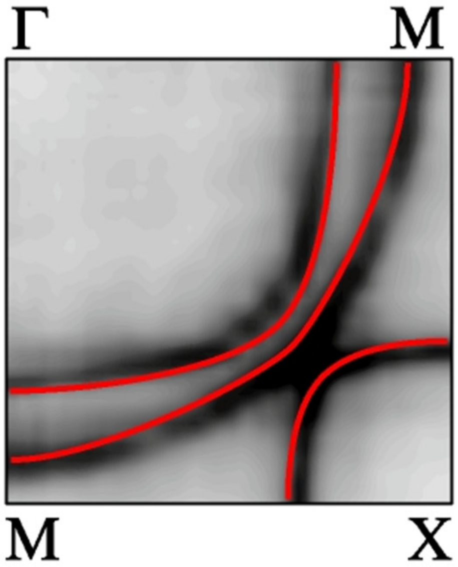


Petrovykh *et al.*, APL **73**, 3459 (1998)

Bulk and Surface FS of Sr_2RuO_4

Hot cleave

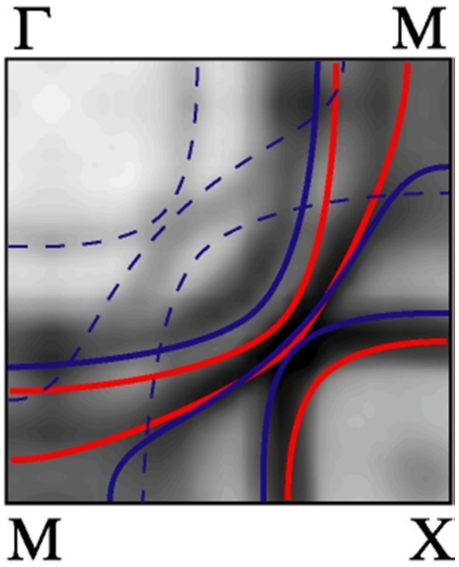
T=180 K



Bulk

Cold cleave

T=10 K



Bulk

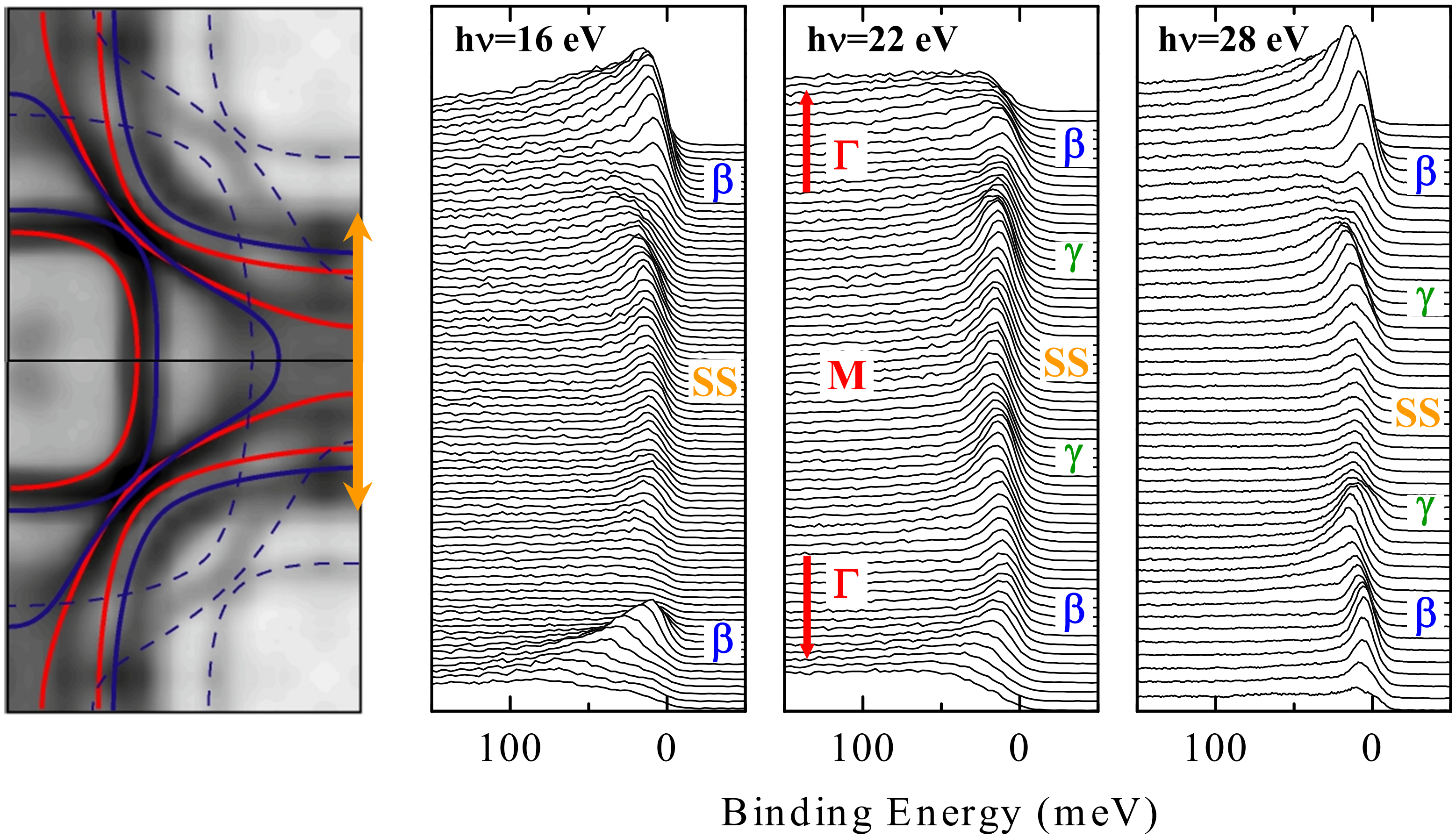
Surface

Bulk and reconstructed surface features

NO evidence for surface ferromagnetism

K. M. Shen, A. Damascelli et al., Phys. Rev. B **64**, 180502 (R) (2001)

Photon Energy Dependence in Sr_2RuO_4



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

What is the role of spin-orbit coupling in Ru-oxides?

In Sr_2RuO_4 it has been effectively mostly ignored!

PRL **101,** 026406 (2008) PHYSICAL REVIEW LETTERS week ending 11 JULY 2008

Strong Spin-Orbit Coupling Effects on the Fermi Surface of Sr_2RuO_4 and Sr_2RhO_4

M. W. Haverkort,[1] I. S. Elfimov,[2] L. H. Tjeng,[1] G. A. Sawatzky,[2] and A. Damascelli[2]

[1]*II. Physikalisches Institut, Universität zu Köln, Zülpicher Straße 77, 50937 Köln, Germany*

[2]*Department of Physics and Astronomy, University of British Columbia, Vancouver, British Columbia, Canada V6T 1Z1*

(Received 29 February 2008; published 11 July 2008)

PRL **101,** 026408 (2008) PHYSICAL REVIEW LETTERS week ending 11 JULY 2008

Coulomb-Enhanced Spin-Orbit Splitting: The Missing Piece in the Sr_2RhO_4 Puzzle

Guo-Qiang Liu, V. N. Antonov, O. Jepsen, and O. K. Andersen.

Max-Planck Institut für Festkörperforschung, D-70569 Stuttgart, Germany

(Received 2 April 2008; published 11 July 2008)

Eigenstates with Spin-Orbit Coupling

Starting from degenerate t_{2g} orbitals

$\sqrt{1/2}\,(-d_{xz}-i\,d_{yz})=d_{1}$

$\sqrt{1/2}\,(d_{xz}-i\,d_{yz})=d_{-1}$

d_{xy}

3-orbitals with orbital momentum 0, +/- 1 like *p*-orbitals

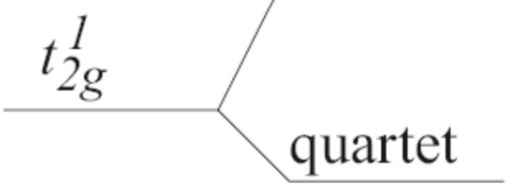
Atomic relativistic SOC

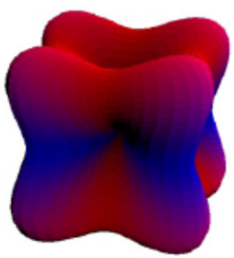
Ru^{4+} ζ=161 meV

Rh^{4+} ζ=191 meV

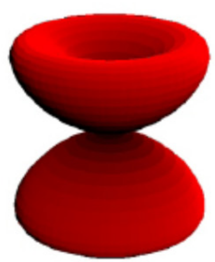
Earnshaw, JCS **601**, 3132 (1961)

$$H = \zeta \sum_i l_i \cdot s_i$$

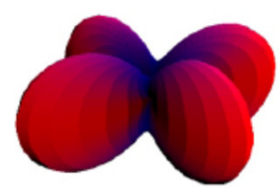




L_z=+-2/3
S_z=+-1/6



L_z=+-1/2
S_z=-+1



L_z=+-1/3
S_z=-+1/6

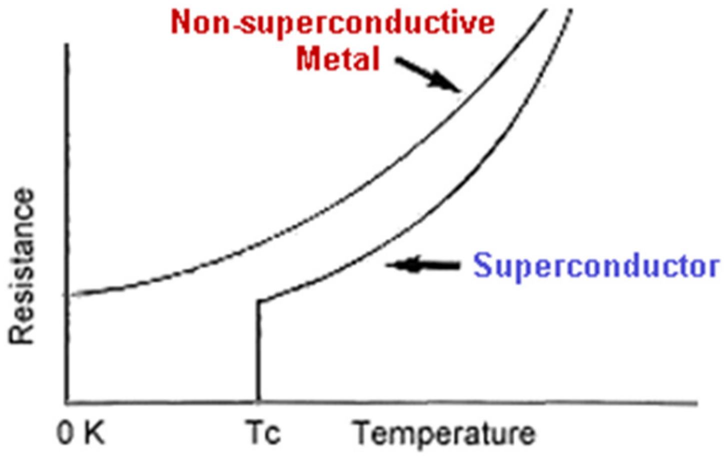
M.W. Haverkort, I.S. Elfimov, L.H. Tjeng, G.A. Sawatzky, A. Damascelli, PRL **101**, 026406 (2008)

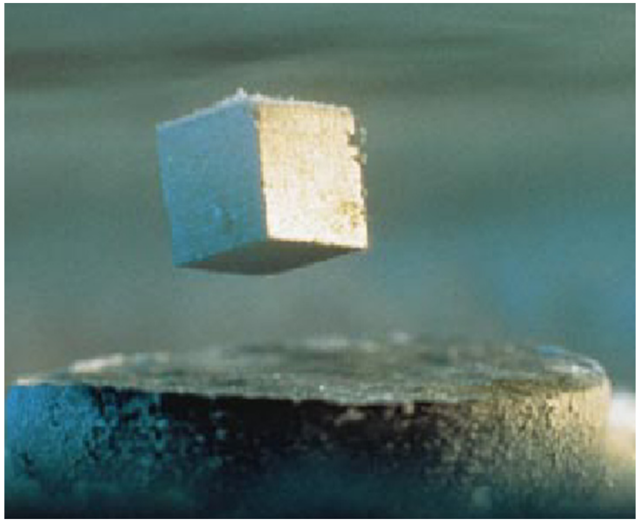
Outline Part I

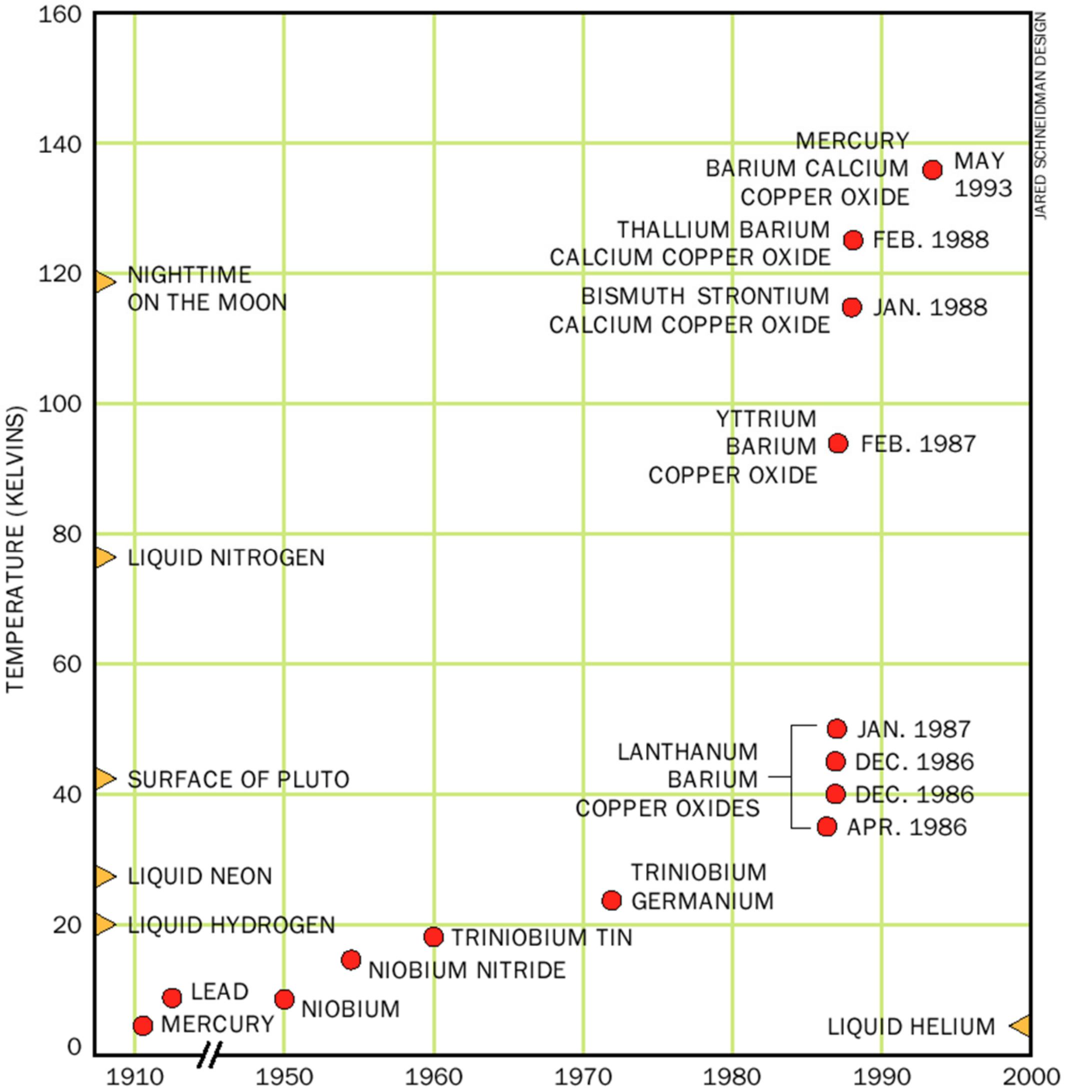
Superconducting gap: BCS and HTSC

CUSO Lecture – Lausanne 02/2011

Superconductivity



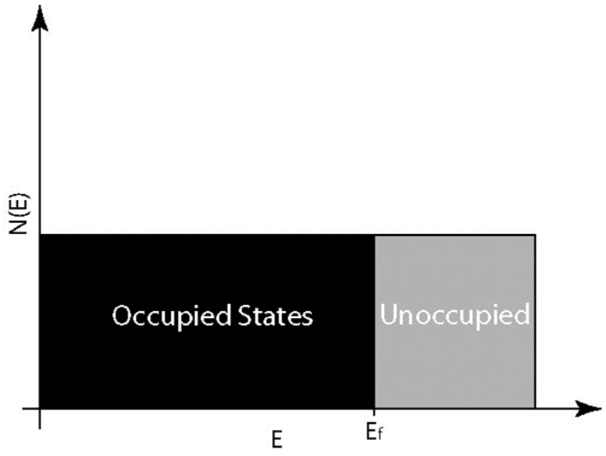




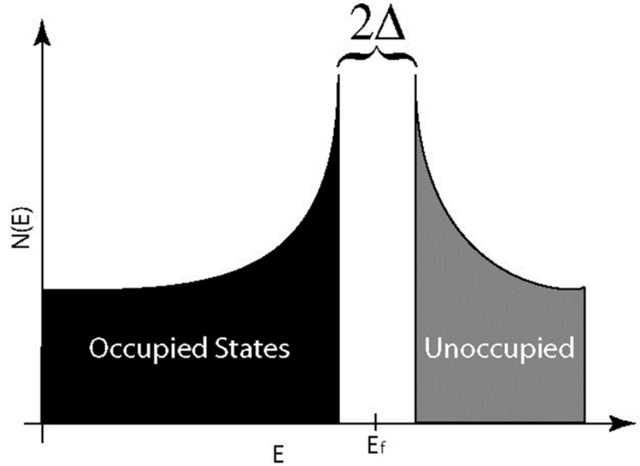
Courtesy of Kyle Shen

"Classic Low-temperature" Superconductors

Metallic Density of States



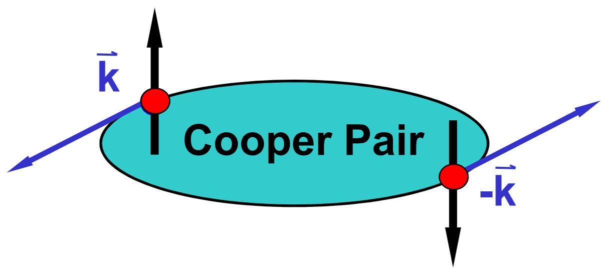
Superconducting Density of States



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

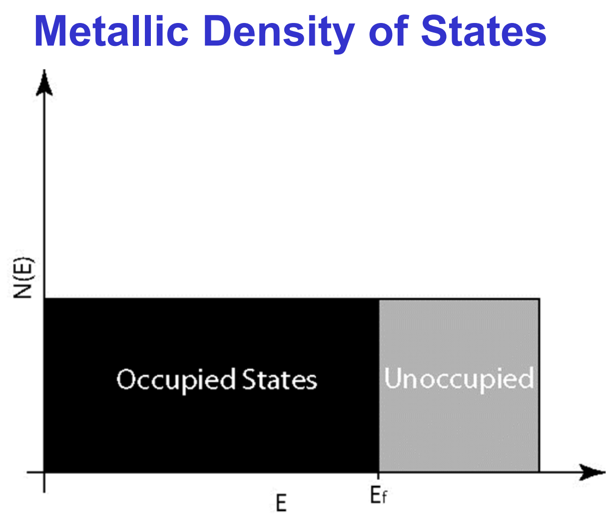
Superconductivity

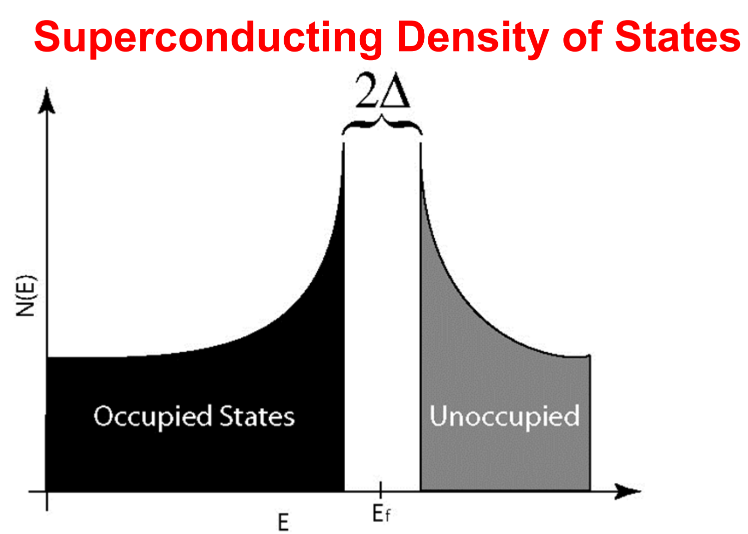
2-electron bound state



spin-singlet pairing

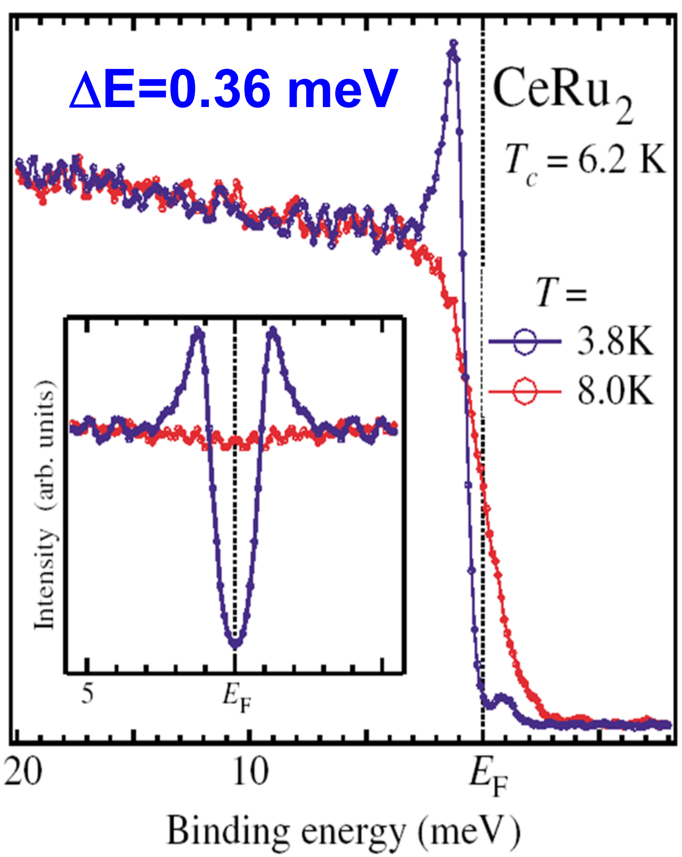
"Classic Low-temperature" Superconductors





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

Kiss et al., PRL **94,** 057001 (2005)

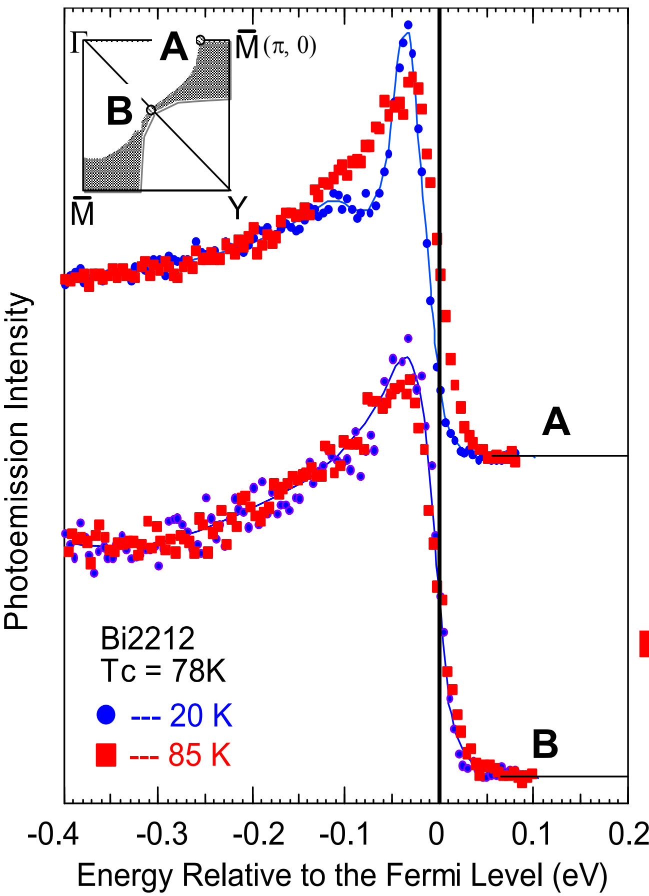


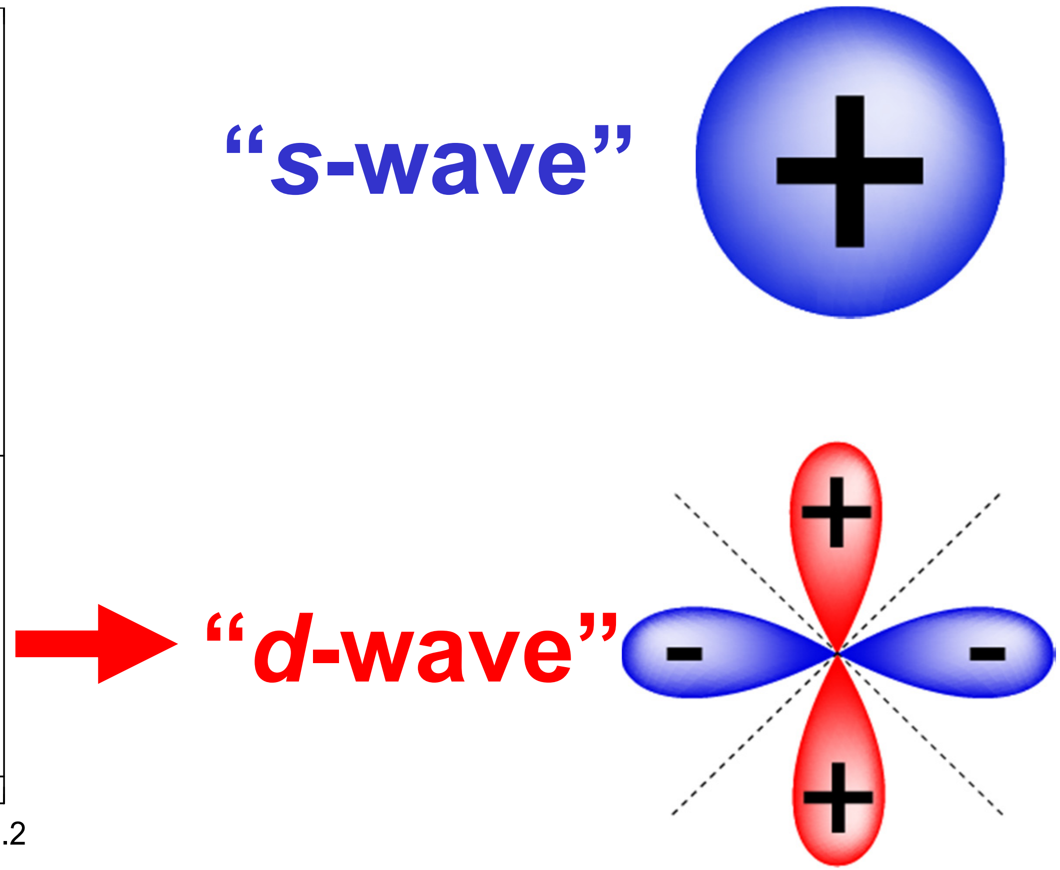
High-Temperature Superconductors

VOLUME 70, NUMBER 10 PHYSICAL REVIEW LETTERS 8 MARCH 1993

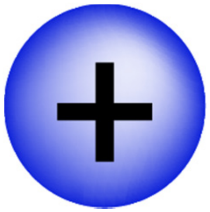
Anomalously Large Gap Anisotropy in the *a*-*b* Plane of $Bi_2Sr_2CaCu_2O_{8+\delta}$

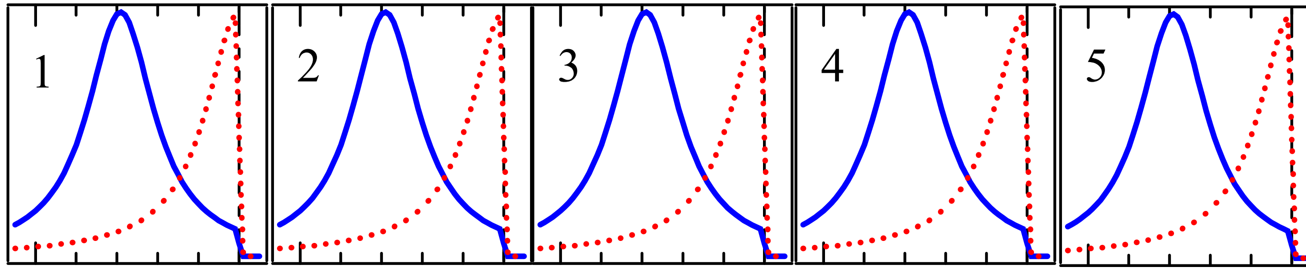
Z.-X. Shen,[(1),(2)] D. S. Dessau,[(1),(2)] B. O. Wells,[(1),(2),(a)] D. M. King,[(2)] W. E. Spicer,[(2)] A. J. Arko,[(3)] D. Marshall,[(2)] L. W. Lombardo,[(1)] A. Kapitulnik,[(1)] P. Dickinson,[(1)] S. Doniach,[(1)] J. DiCarlo,[(1),(2)] A. G. Loeser,[(1),(2)] and C. H. Park[(1),(2)]

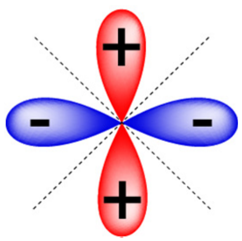


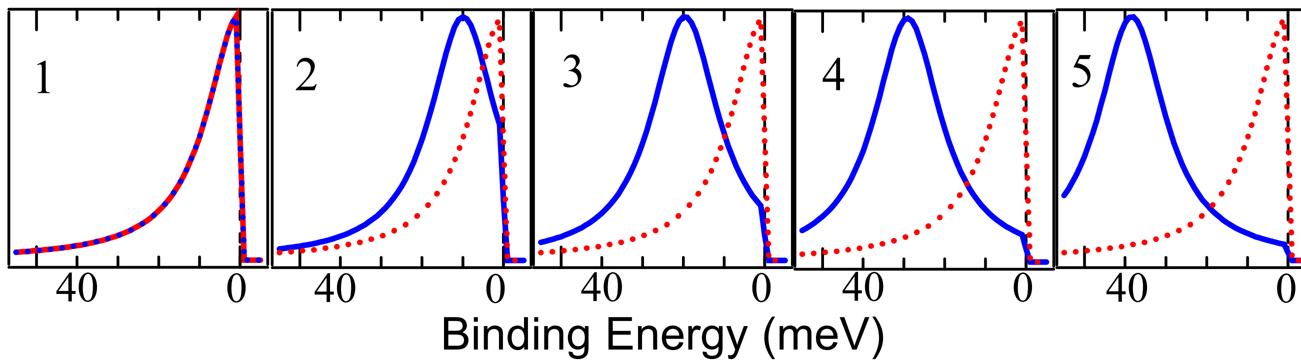


High-Tc Superconductors: s-wave vs. d-wave gap

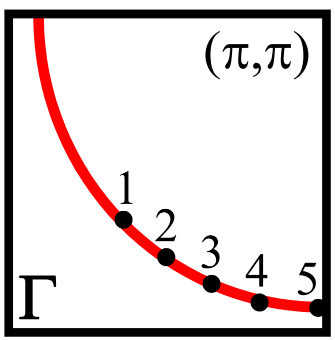


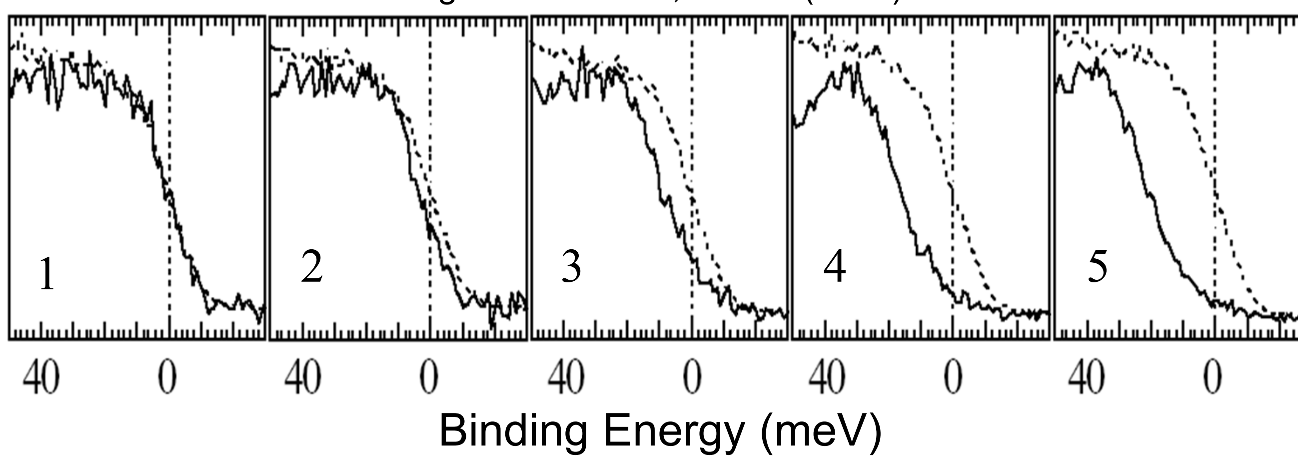




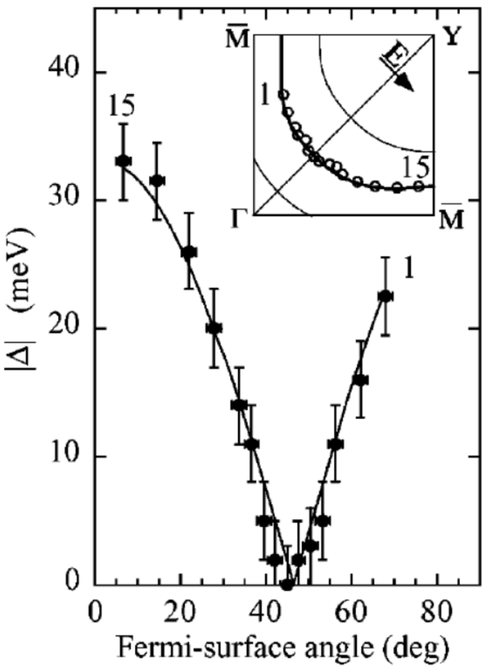


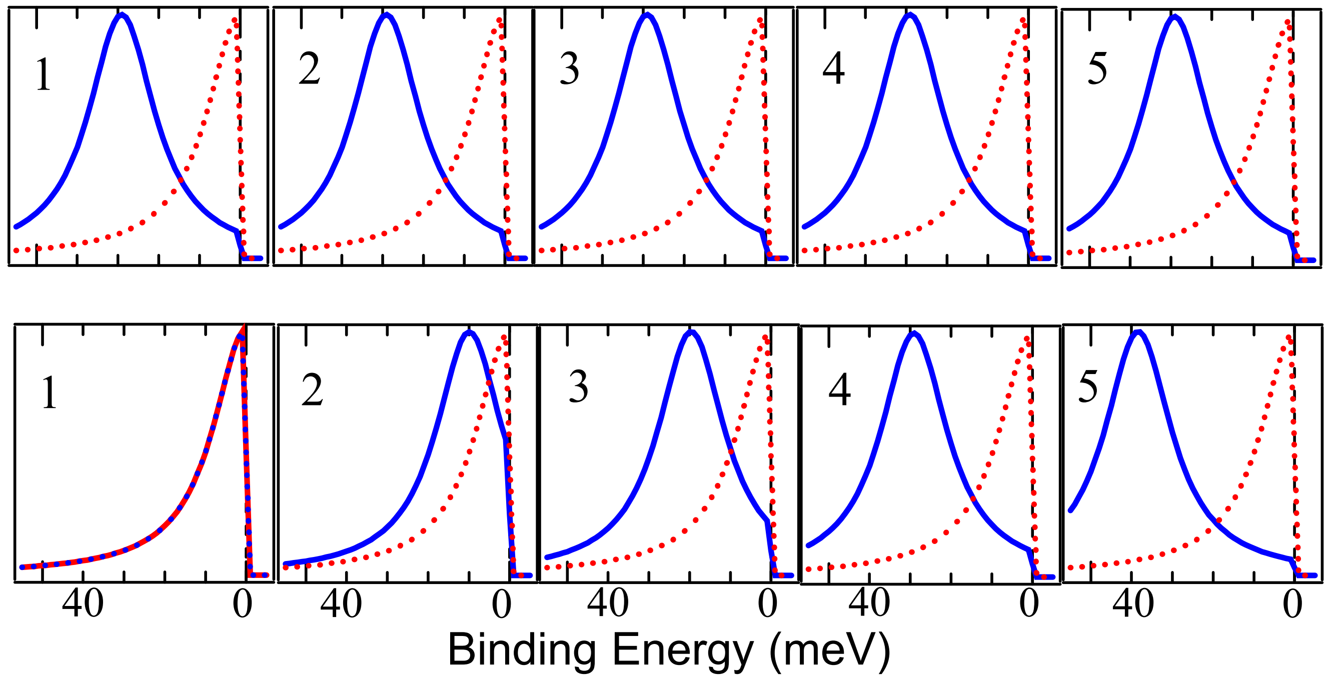
Ding et al. PRB **54**, R9678 (1996)

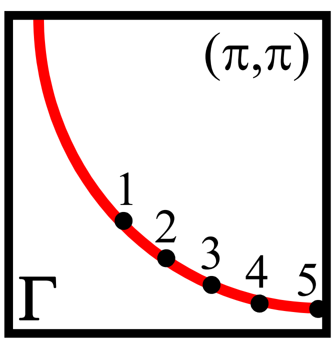




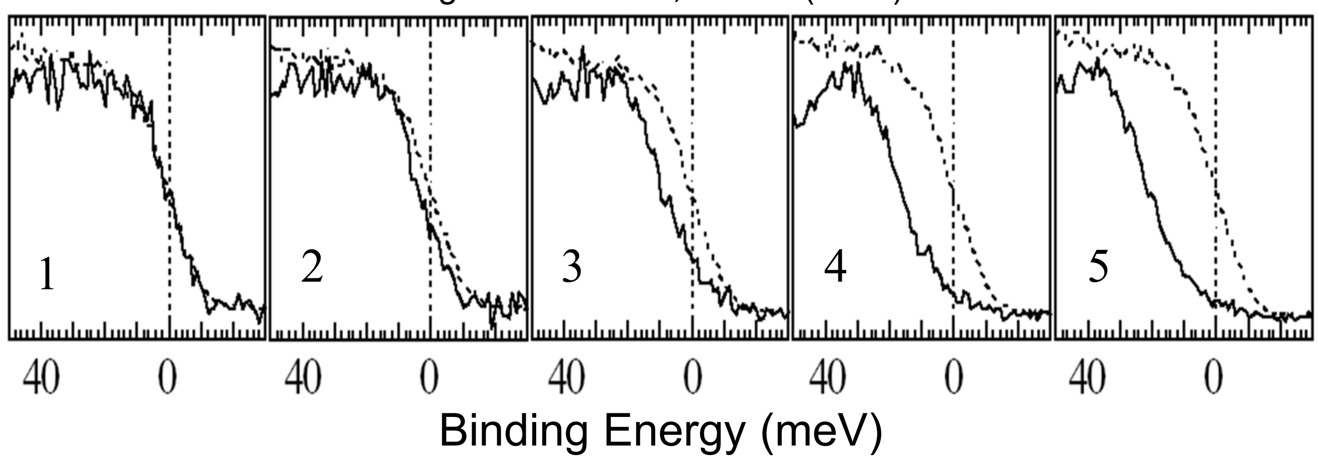
High-Tc Superconductors: s-wave vs. d-wave gap



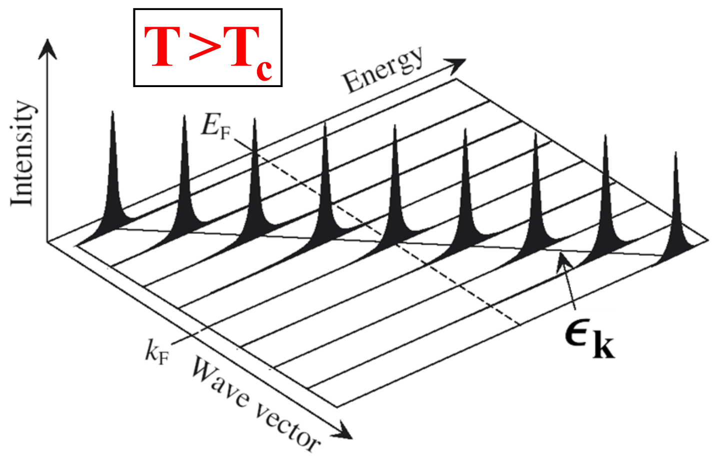




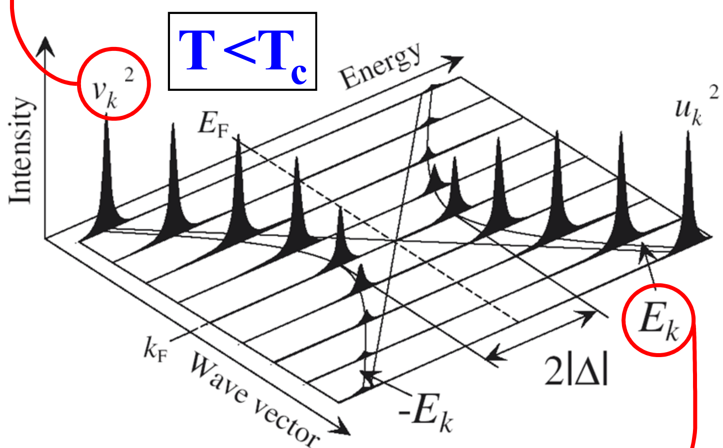
Ding et al. PRB **54**, R9678 (1996)



High-Tc Superconductors: Bogoliubov QP in Bi2223

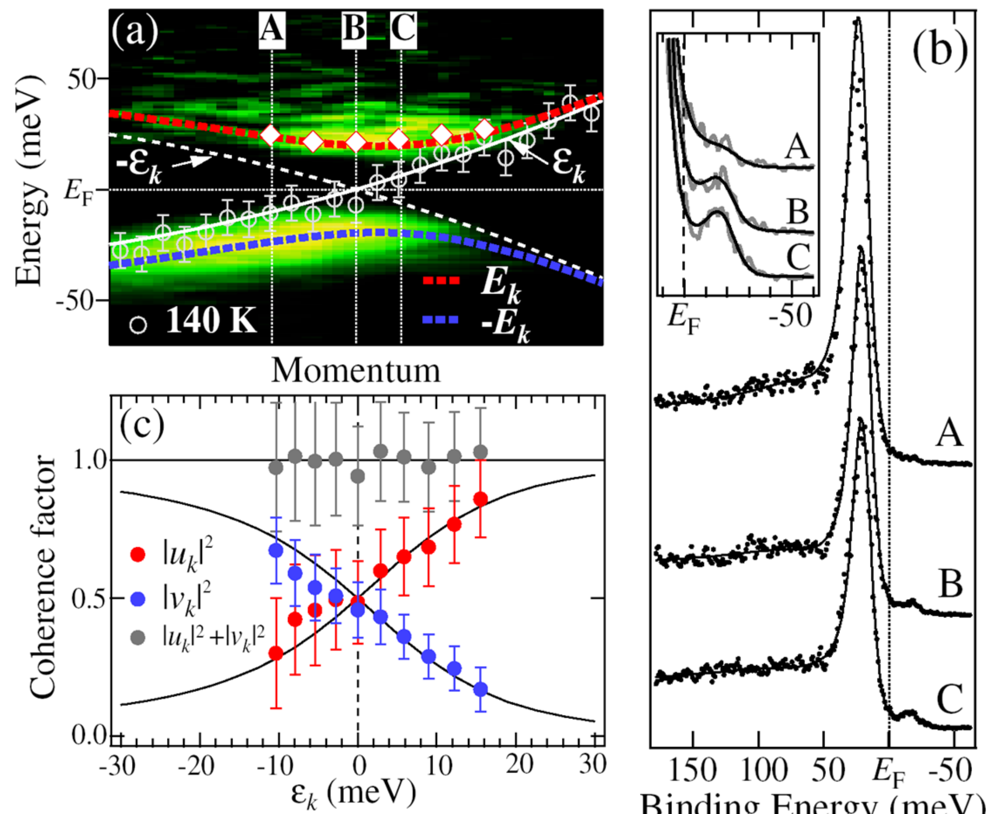


$$v_{\mathbf{k}}^2 = 1 - u_{\mathbf{k}}^2 = \frac{1}{2}\left(1 - \epsilon_{\mathbf{k}}/E_{\mathbf{k}}\right)$$



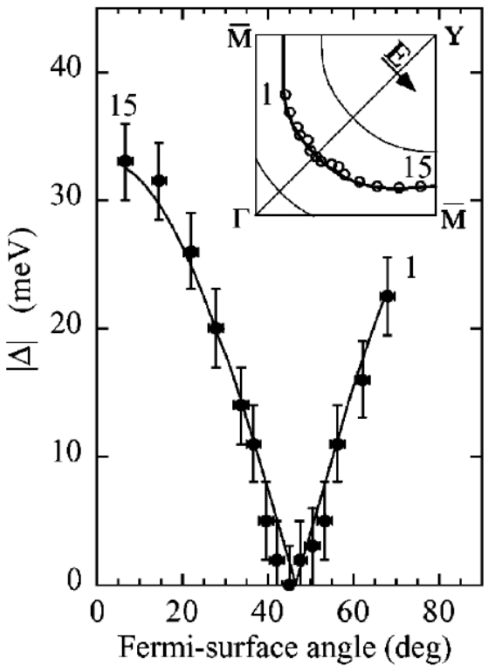
$$E_{\mathbf{k}} = \sqrt{\epsilon_{\mathbf{k}}^2 + |\Delta(\mathbf{k})|^2}$$

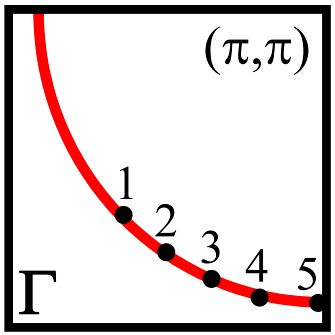
$$A_{\mathrm{BCS}}(k,\omega) = \frac{1}{\pi}\left\{\frac{|u_k|^2\Gamma}{(\omega - E_k)^2 + \Gamma^2} + \frac{|v_k|^2\Gamma}{(\omega + E_k)^2 + \Gamma^2}\right\}$$



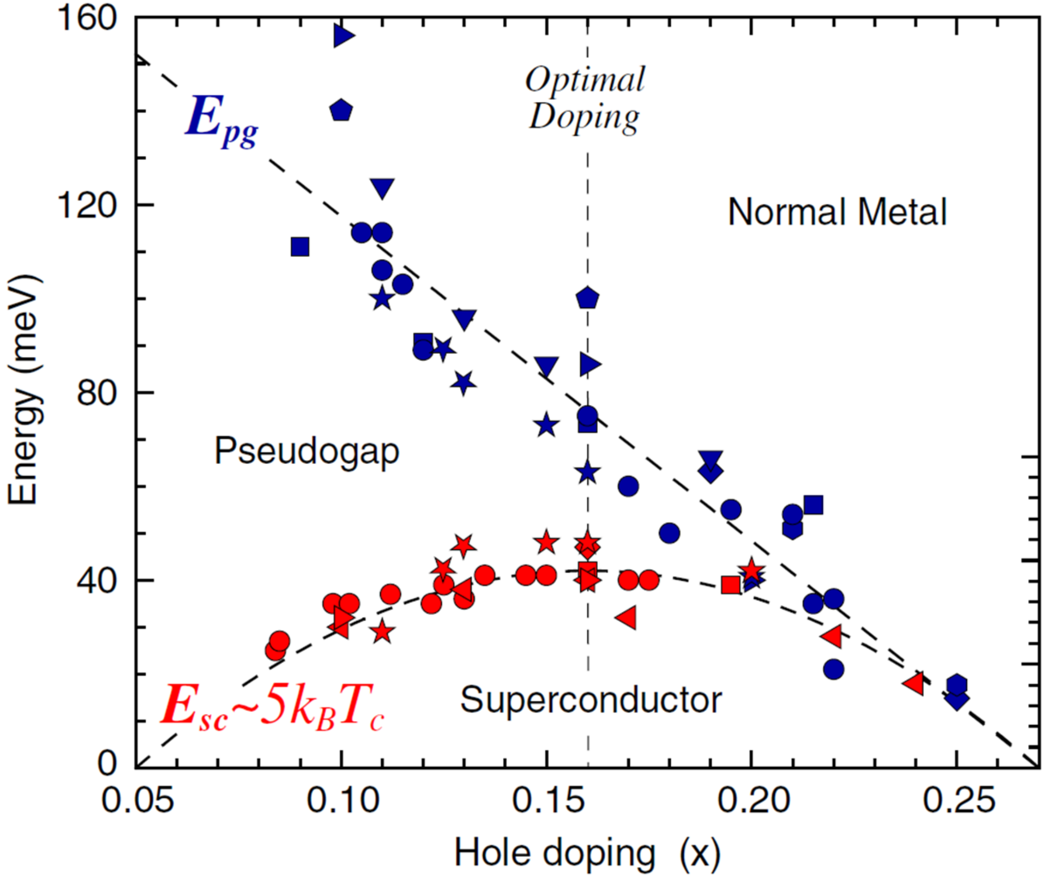
Matsui *et al.*, PRL **90**, 217002 (2003)

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



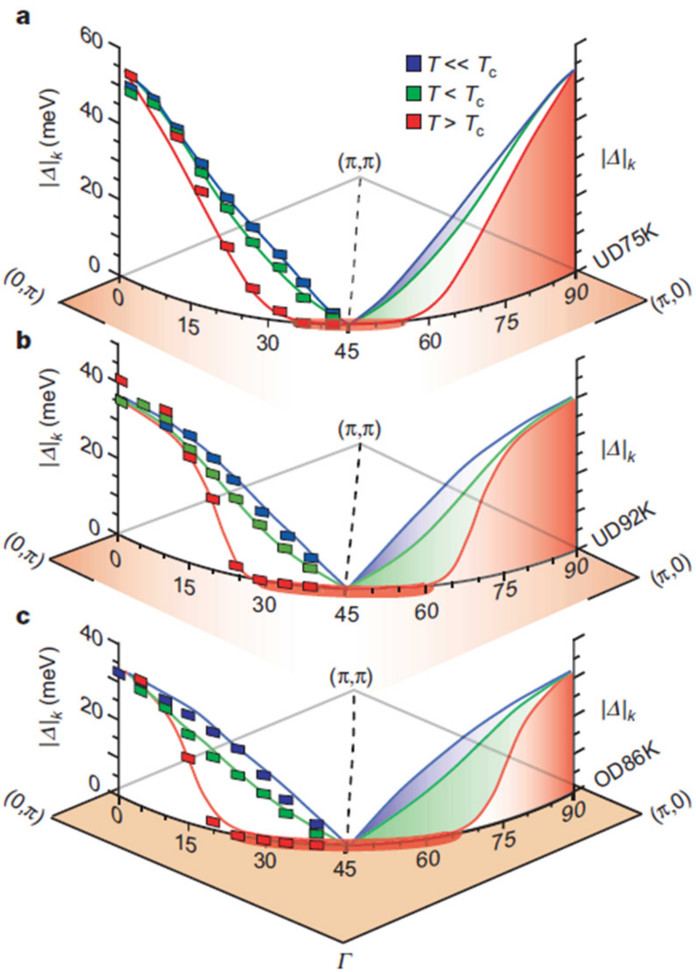


Two gaps make a HTSC?



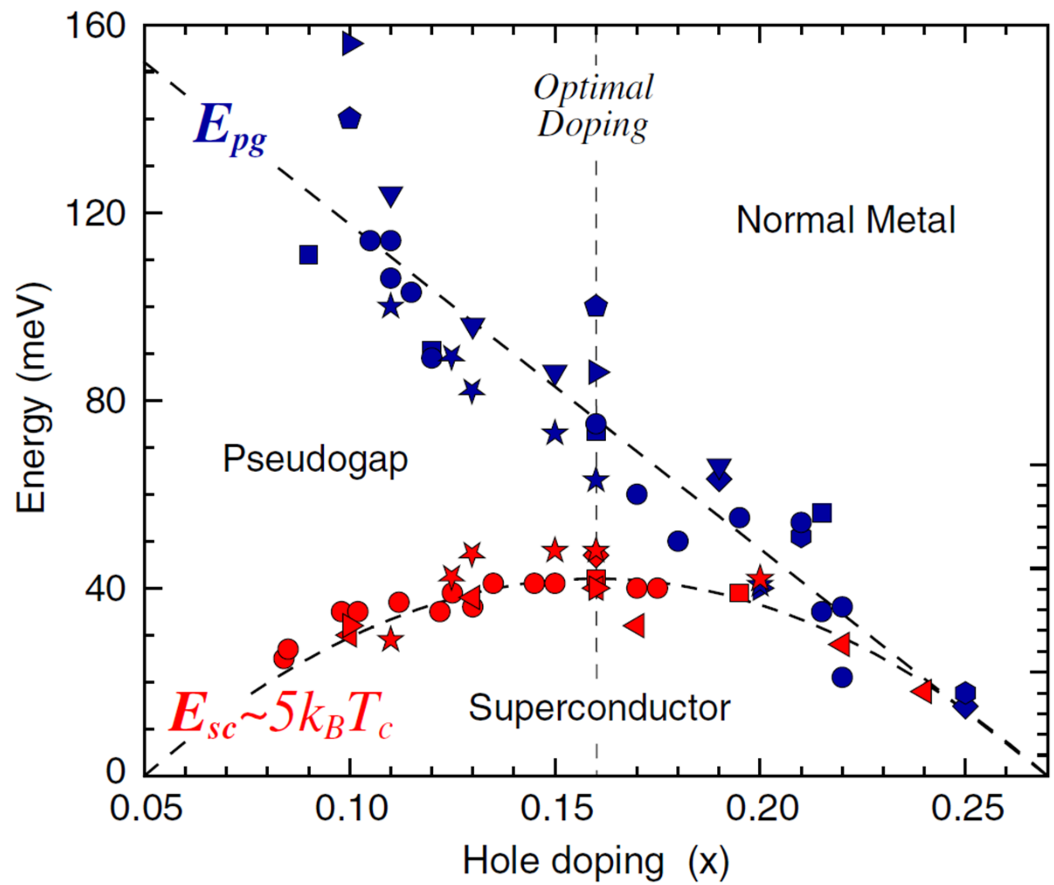
Hufner, Hossain, Damascelli, Sawatzky
Rep. Prog. Phys. 71, 062501 (2008)

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



W. S. Lee et al., Nature 450, 81 (2007)

Two gaps make a HTSC?



Hufner, Hossain, Damascelli, Sawatzky
Rep. Prog. Phys. 71, 062501 (2008)