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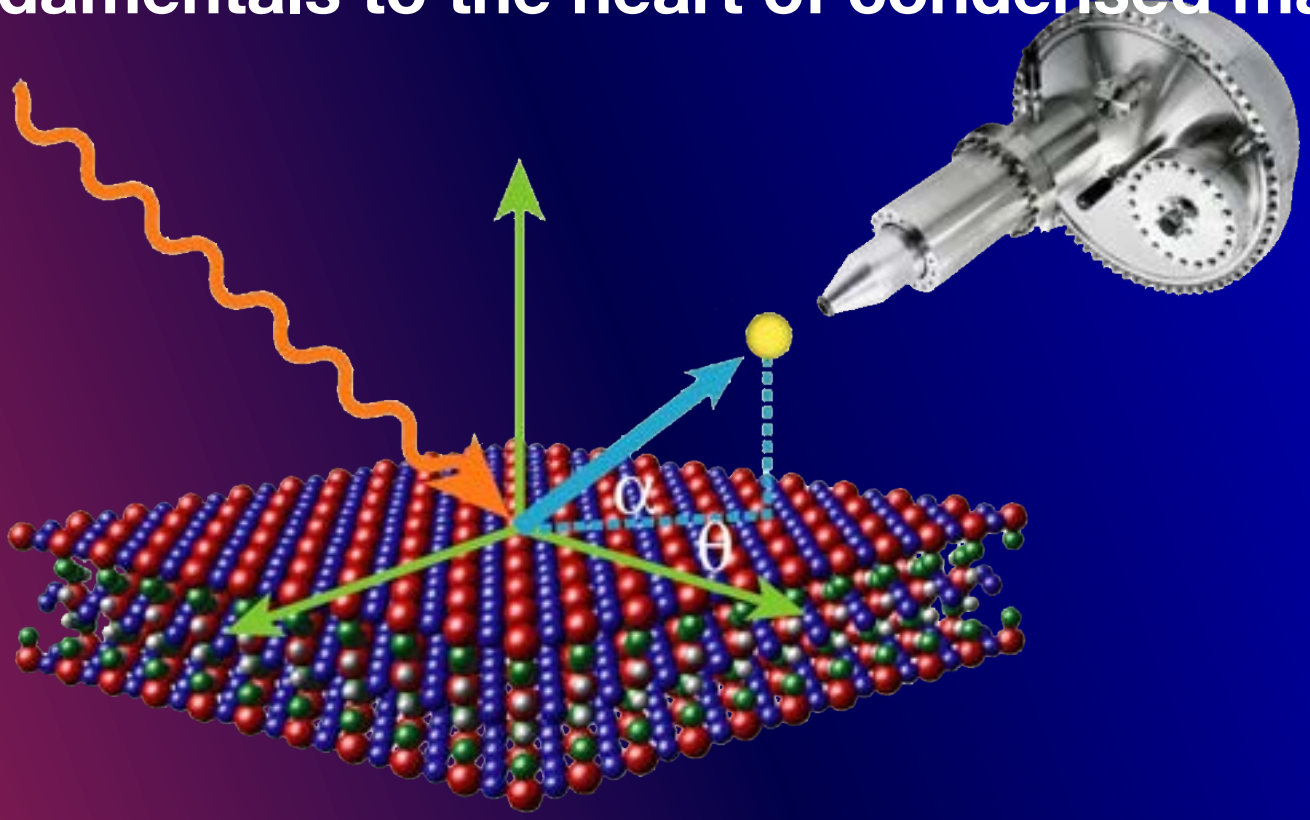
EXCELENCIA SEVERO OCHOA

Angle Resolved Photoemission Spectroscopy

From fundamentals to the heart of condensed matter



6-7 FEBRERO, 2023

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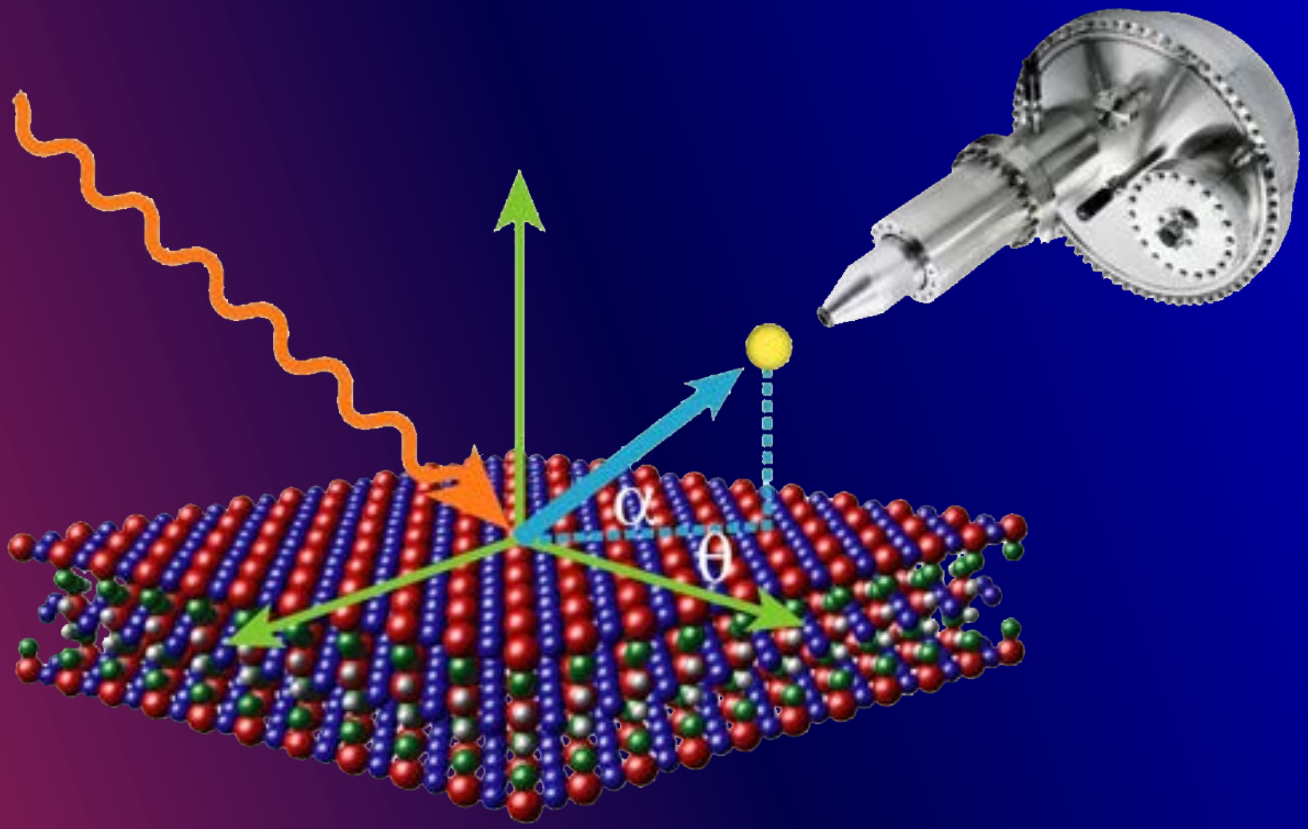
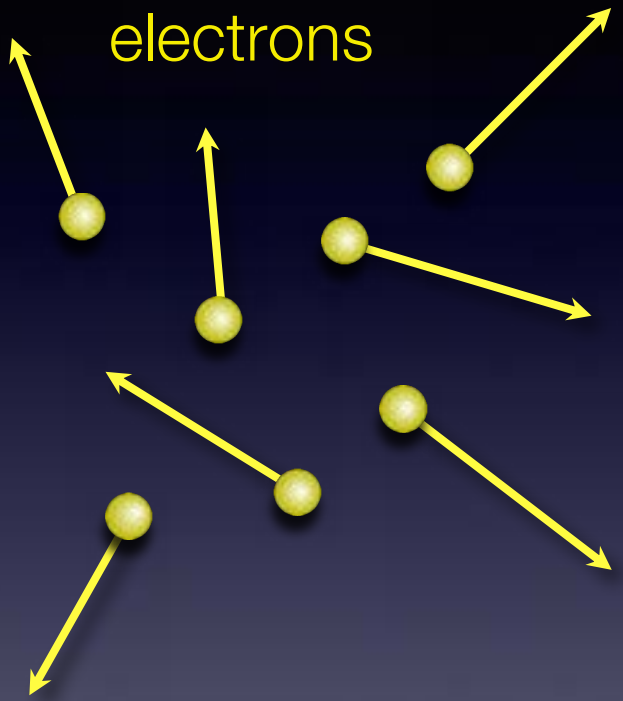
Lecture #2 : From Particles to Quasiparticles

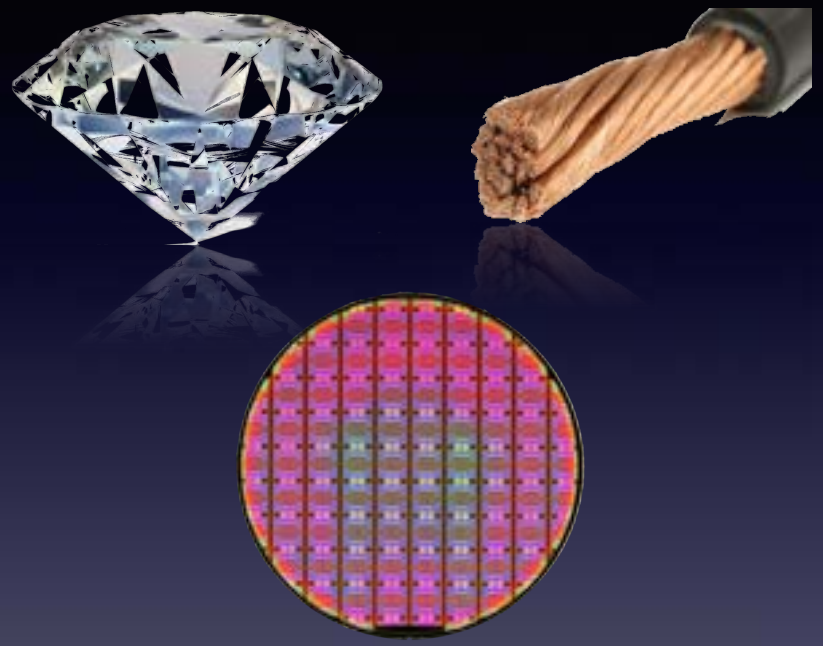


6-7 FEBRERO, 2023

the “independent electron” approximation

independent electrons





electron KE

$$-\sum_{j}^{N_e}\frac{\hbar^2}{2m}\nabla_j^2$$

electron-electron interactions

$$+\sum_{j\ll k}^{N_e}\frac{e^2}{|\vec{r}_j-\vec{r}_k|}$$

nuclei KE

$$-\sum_{\alpha}^{N_i}\frac{\hbar^2}{2M_\alpha}\nabla_\alpha^2$$

electron-nuclei interactions

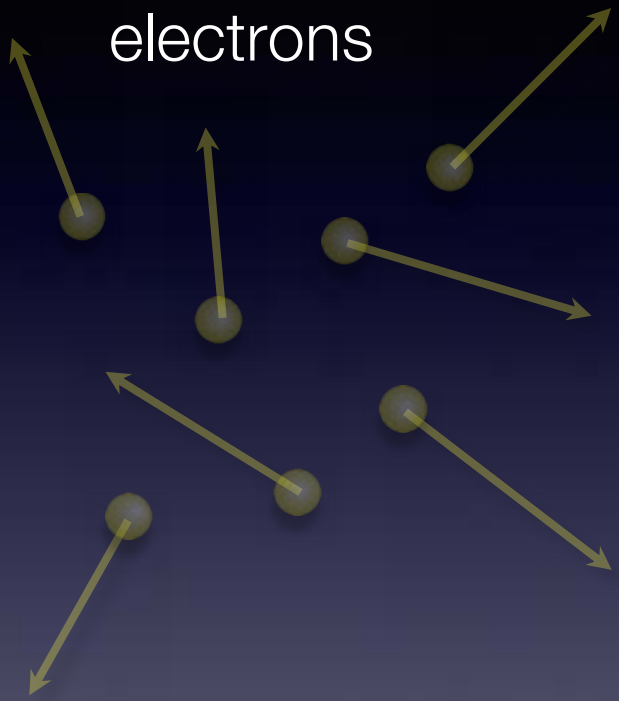
$$-\sum_{j}^{N_e}\sum_{\alpha}^{N_i}\frac{Z_\alpha e^2}{|\vec{r}_j-\vec{R}_\alpha|}$$

nuclei-nuclei interactions

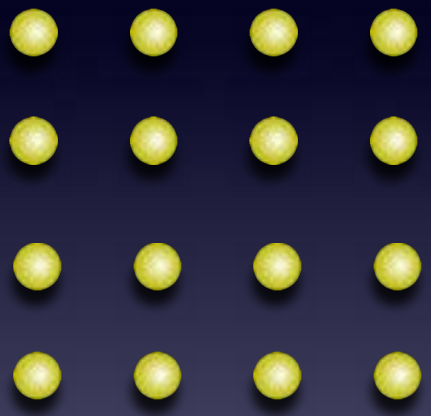
$$+\sum_{\alpha\ll\beta}^{N_j}\frac{Z_\alpha Z_\beta e^2}{|\vec{R}_\alpha-\vec{R}_\beta|}$$

how are “quantum materials” different from conventional ones?

independent electrons



electronic crystal



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electron-nuclei interactions

$$-\sum_{j}^{N_e} \sum_{\alpha}^{N_i} \frac{Z_\alpha e^2}{|\vec{r}_j - \vec{R}_\alpha|}$$

nuclei-nuclei interactions

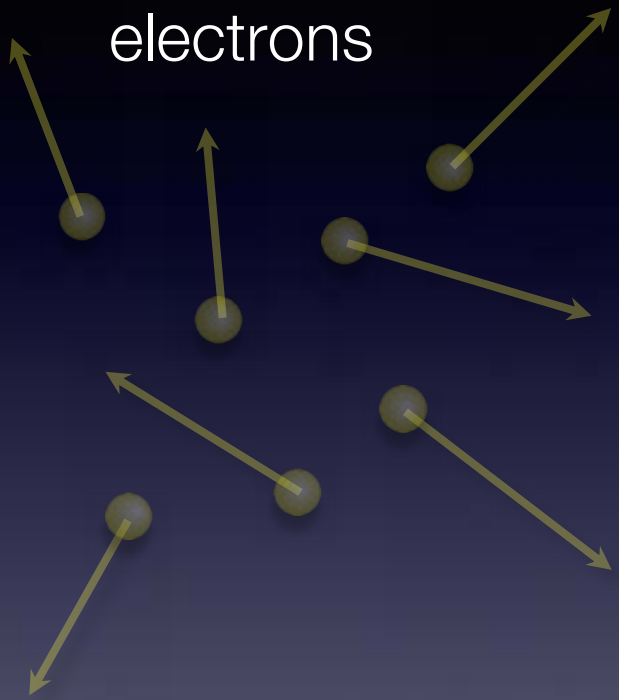
$$+\sum_{\alpha \ll \beta}^{N_j} \frac{Z_\alpha Z_\beta e^2}{|\vec{R}_\alpha - \vec{R}_\beta|}$$

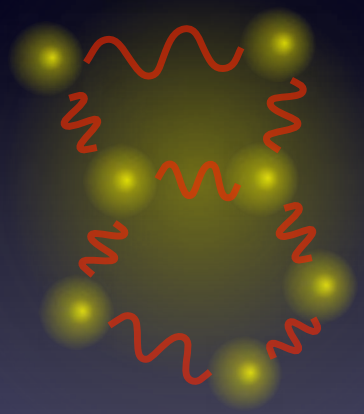
how are “quantum materials” different from conventional ones?

independent electrons

highly entangled many-body state

electronic crystal





electron KE

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electron-electron interactions

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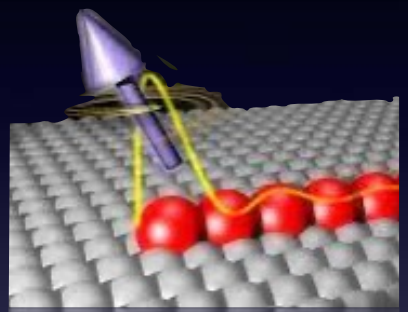
nuclei-nuclei interactions

$$+\sum_{\alpha \ll \beta}^{N_j} \frac{Z_\alpha Z_\beta e^2}{|\vec{R}_\alpha - \vec{R}_\beta|}$$

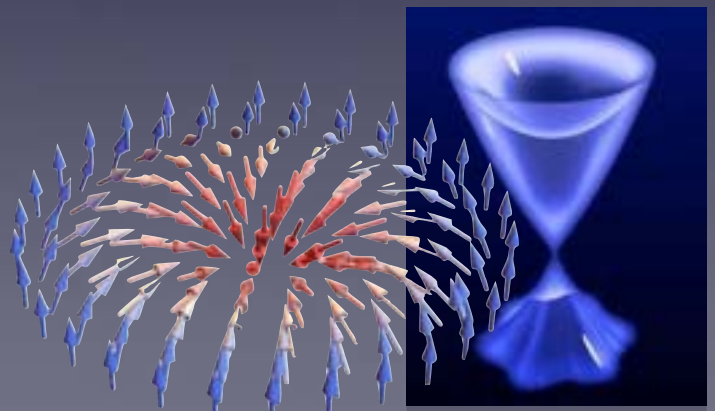
New opportunities presented by quantum materials

higher (room) temperature superconductors?

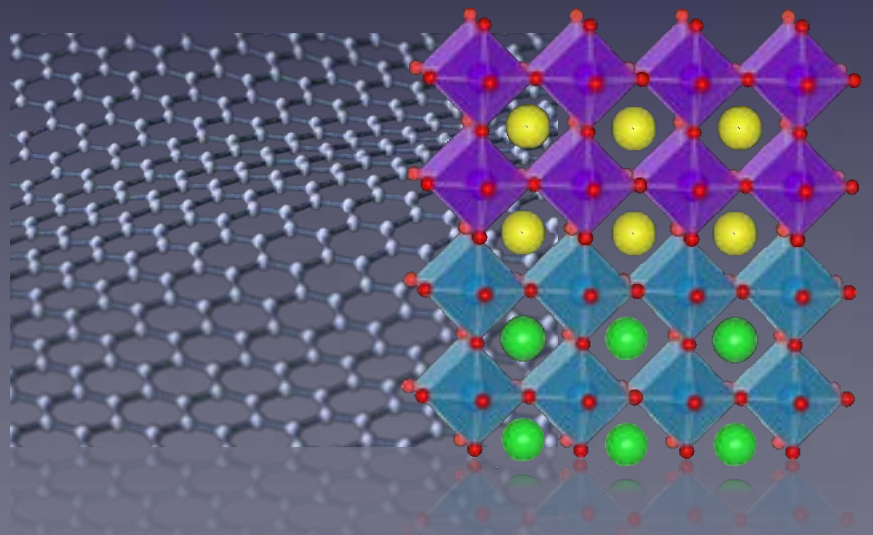
fault-tolerant quantum computing?

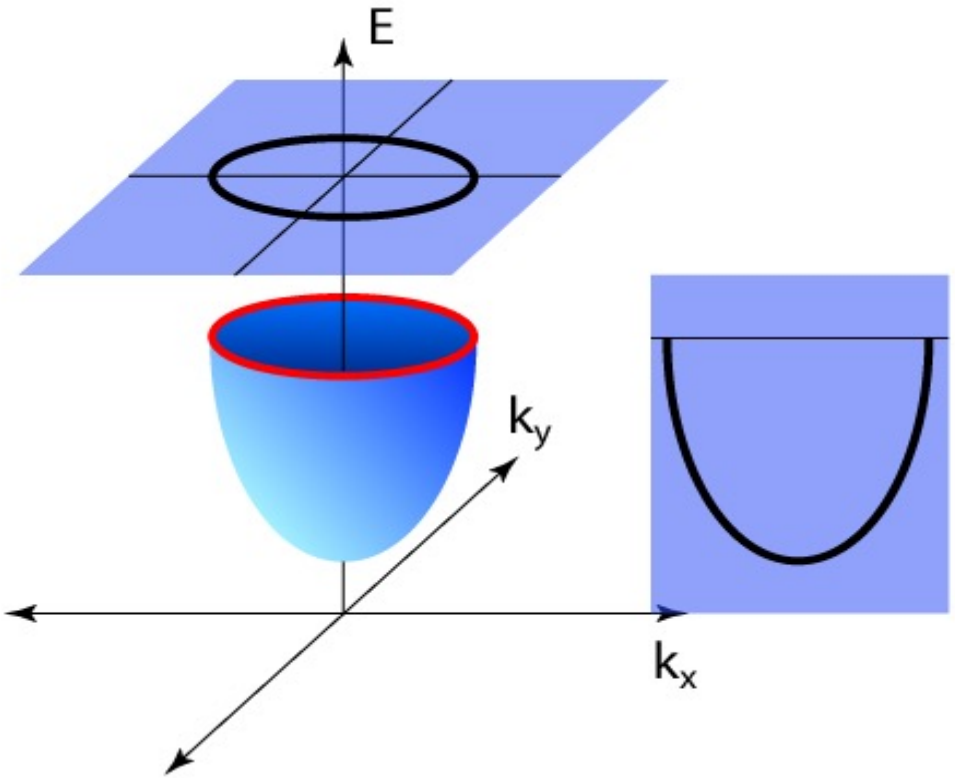


topologically protected states



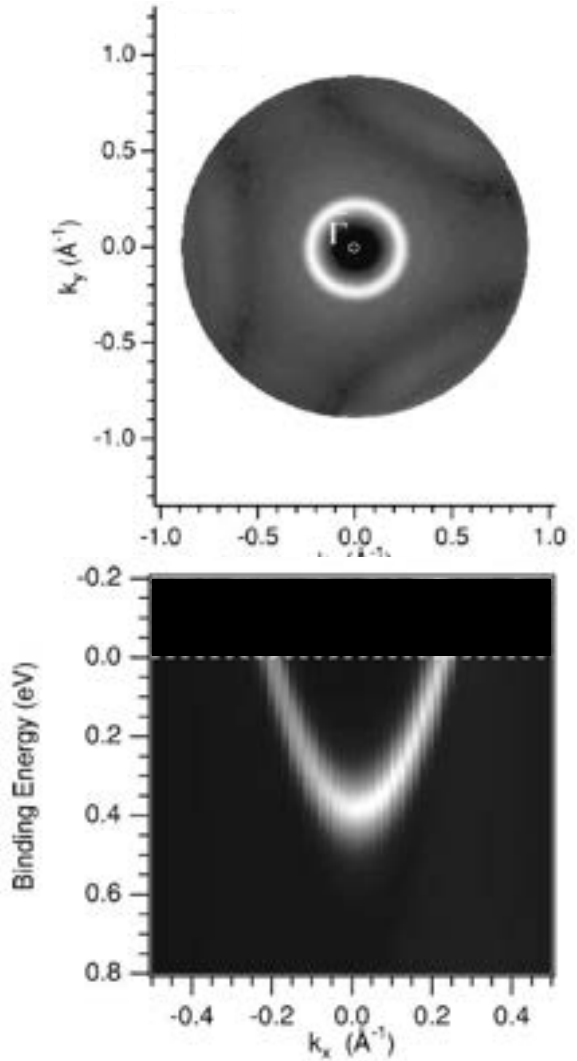
low-dimensional materials





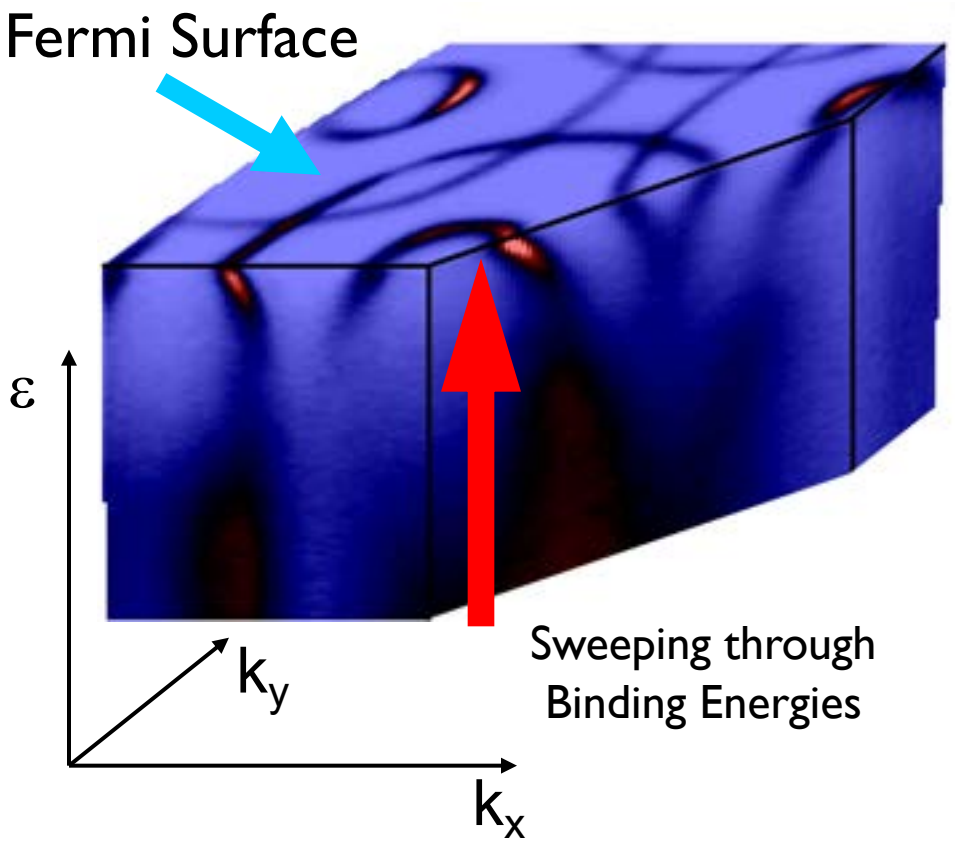
$$E = \hbar^2\mathbf{k}^2 / 2m^*$$

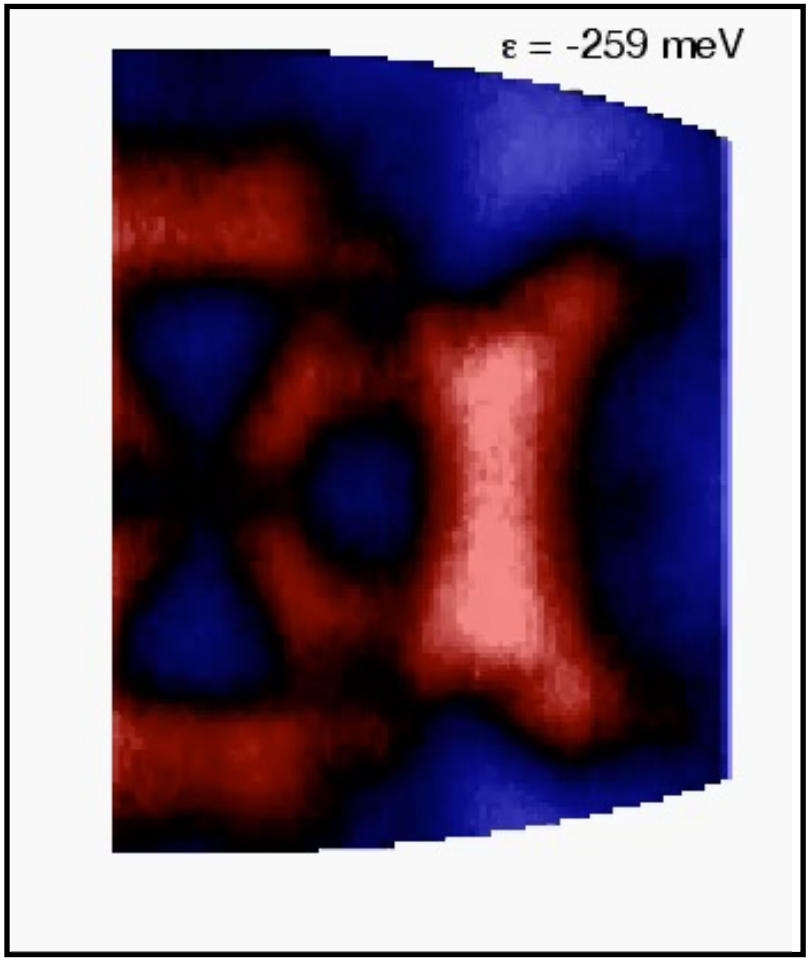
Cu (111)



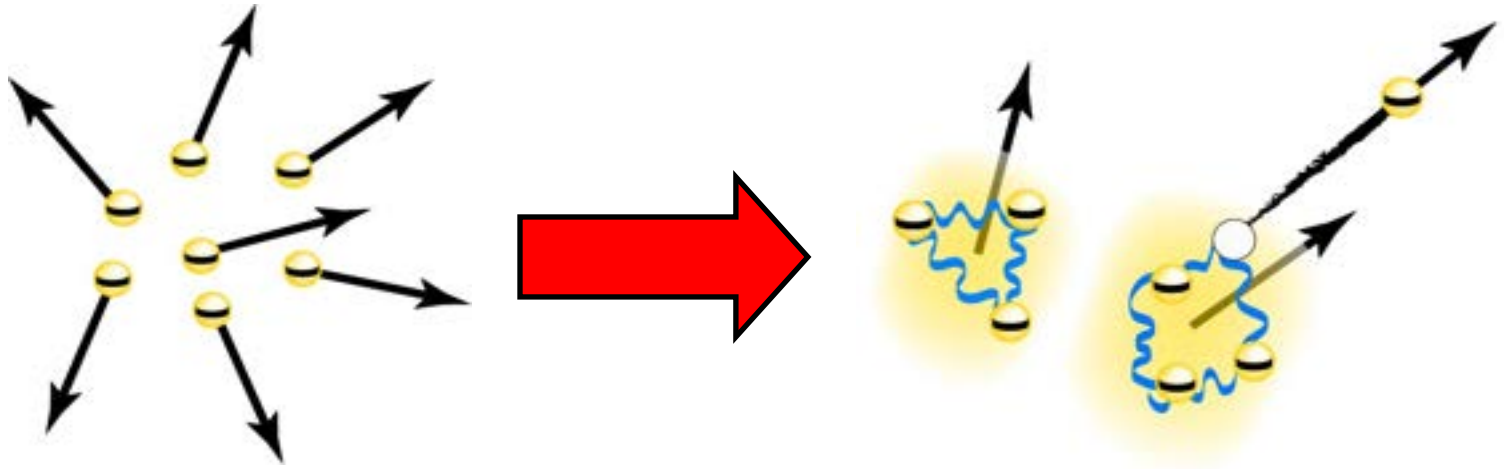
F. Baumberger et al., PRB 64, 195411 (2001)

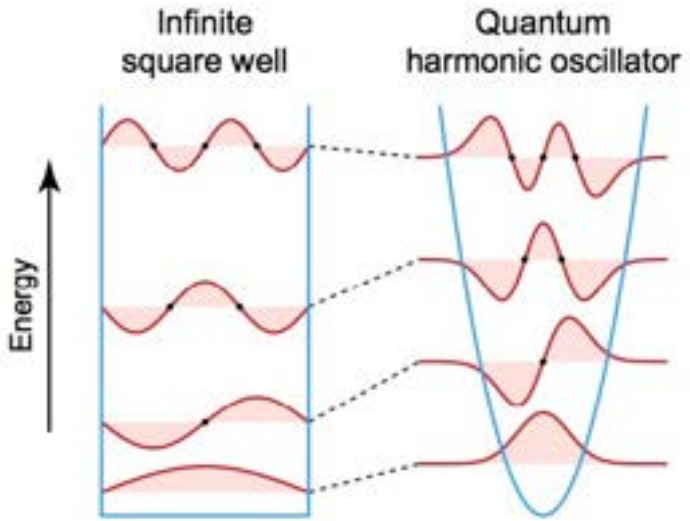
Sr_2RhO_4 : A *4d* transition metal oxide with strong spin-orbit coupling

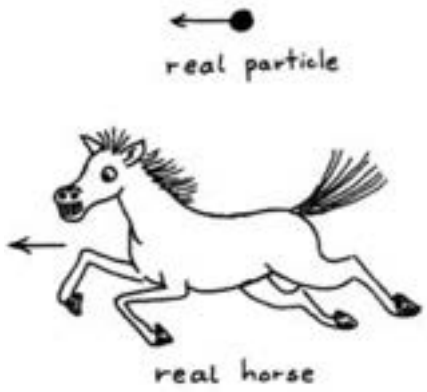


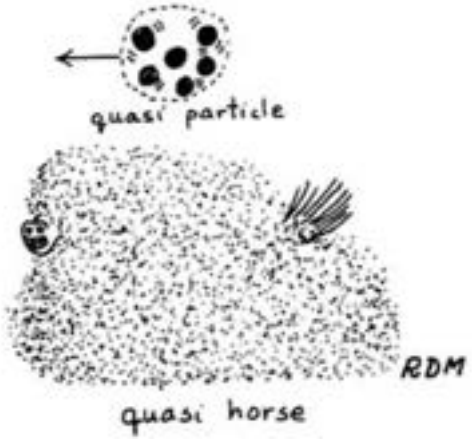


F. Baumberger, N.J.C. Ingle, W. Meevasana, K.M.S., et al. (Phys. Rev. Lett.)





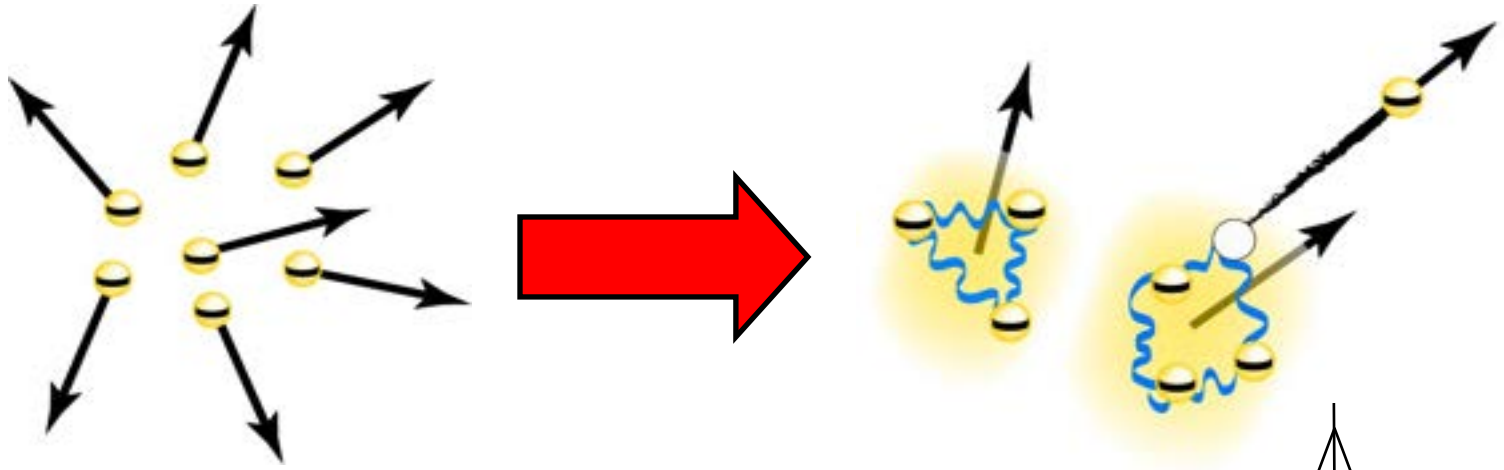


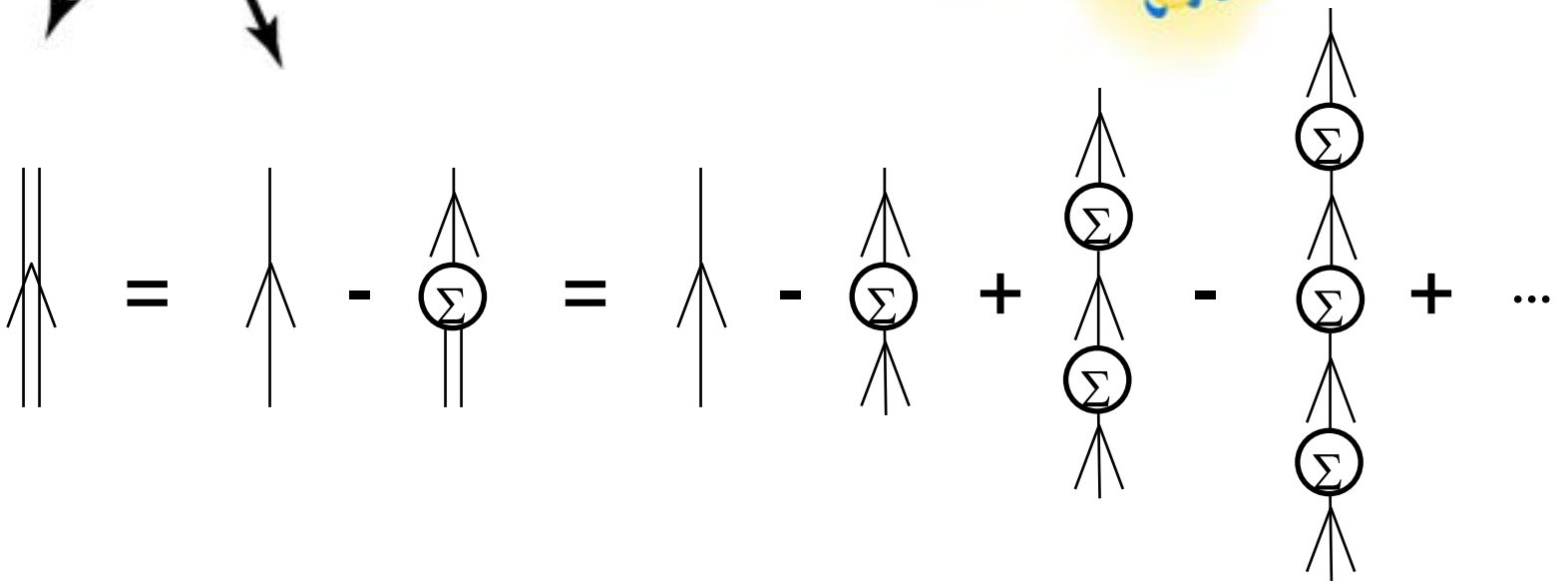


Mattuck "A guide to Feynman diagrams in the Many-Body Problem"

Single-particle spectral function

$$A(\mathbf{k},\omega) \propto \frac{\mathrm{Im}\Sigma(\mathbf{k},\omega)}{[\omega - \varepsilon_k - \mathrm{Re}\Sigma(\mathbf{k},\omega)]^2 + [\mathrm{Im}\Sigma(\mathbf{k},\omega)]^2}$$





Single-particle spectral function

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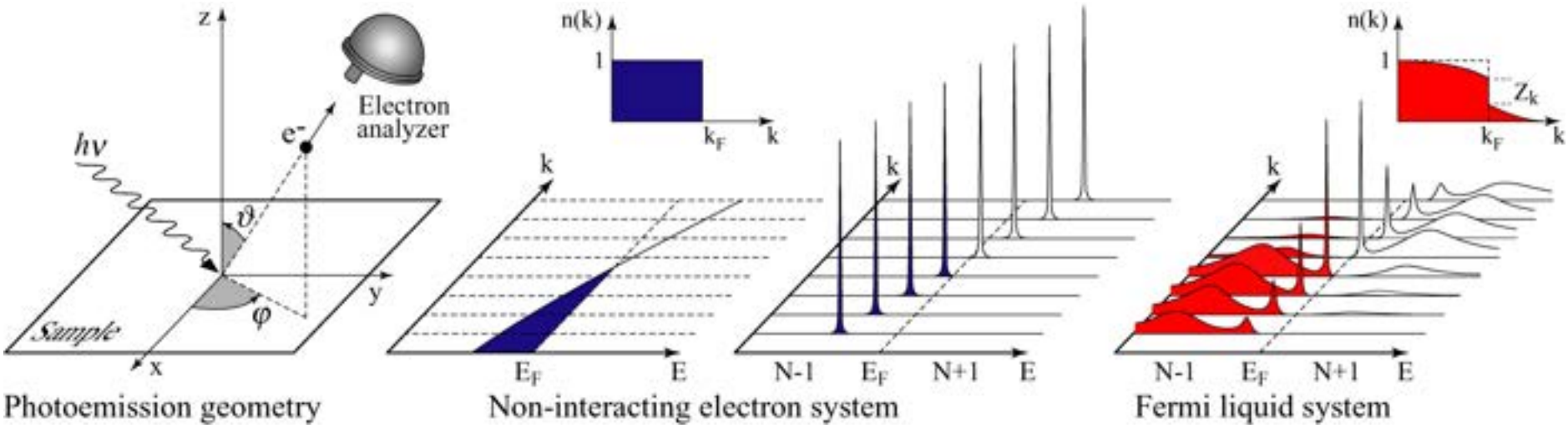
ARPES on Fermi Liquid Quasiparticles



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

Courtesy of Andrea Damascelli

Rev. Mod. Phys. 75, 473 (2003)

Fermi's Golden Rule : Transition probability from state i to f

$$w_{i\to f} = \frac{2\pi}{\hbar} \; |\langle \Psi_f^N | H_{int} | \Psi_i^N \rangle|^2 \;\; \delta(E_f - E_i - \hbar\omega)$$

what is H_{int}?

$$H_{int} = \frac{1}{2m}(\vec{p} + \frac{e}{c}\vec{A})^2 - e\phi - \frac{\vec{p}^{\,2}}{2m} \approx -\frac{e}{mc}\vec{A}\cdot\vec{p} \approx \vec{A}_0 \cdot \vec{r}$$

- This assumes a classical EM wave (not valid for very intense fields, neglects $\mathbf{A}^2$)
- Assumes $[\mathbf{p}, \mathbf{A}] = 0$ (not formally true at surfaces)
- Electric dipole approximation valid (since wavelength of A >> atomic dimensions)

Fermi's Golden Rule : Transition probability from state i to f

$$w_{i\to f} = \frac{2\pi}{\hbar} \left|\langle \Psi_f^N | H_{int} | \Psi_i^N \rangle\right|^2 \delta(E_f - E_i - \hbar\omega)$$

How to express initial & final states?

$$\Psi_f^N = \overbrace{\phi_k}^{\text{photoelectron}} \Psi_f^{N-1}$$

Final state is the outgoing photoelectron & N-1 electron system left behind.

$$\Psi_i^{N-1} = c_k \Psi_i^N \approx \phi_i^k \Psi_i^{N-1}$$

Assume single Slater determinant for simplicity of calculation of matrix element

Fermi's Golden Rule : Transition probability from state i to f

$$w_{i\to f} = \frac{2\pi}{\hbar} \; |\langle \Psi_f^N | H_{int} | \Psi_i^N \rangle|^2 \;\; \delta(E_f - E_i - \hbar\omega)$$

$$\langle \Psi_f | H_{int} | \Psi_i \rangle = \langle \phi_f^k | A_0 \cdot \vec{r} | \phi_i^k \rangle \; \langle \Psi_f^{N-1} | \Psi_i^{N-1} \rangle$$

One-electron dipole matrix element, M_{if}

Overlap integral between initial & final N-1 systems

To calculate the total photoemission intensity at a specific momentum and energy, $I(\mathbf{k},\omega)$, we should sum over all of the possible initial and final states that will contribute at that energy and momentum :

$$I(k, E) = \sum_{i,f} w_{i\to f}$$

From photoemission intensity to spectral functions

For interacting systems, Ψ_f^{N-1} is NOT an eigenstate of the *N-1* system due to interactions

$$I(\vec{k}, E_{kin}) = \sum_{i,f} |M_{i\to f}|^2 \sum_m |\langle \Psi_m^{N-1} | \Psi_i^{N-1} \rangle|^2 \delta(E_{kin} + E_m^{N-1} - E_i^N - h\nu)$$

Dipole matrix element

$\langle \phi_f^k | A_0 \cdot \vec{r} | \phi_i^k \rangle$

Interesting part!

Energy conservation

Dipole matrix element :

- Highly dependent on the orbital character of the wavefunction, polarization of the incoming radiation, and momentum wavevector
- Usually slowly varying as a function of energy and momentum (can be thought of as Fourier transform of the wavefunction)

Overlap integral :

- We are calculating the overlap integral between the *N*-electron initial state wavefunction with one electron "annihilated", with the *N-1* final state wavefunction
- We are expressing the *N-1* final state wavefunction in the basis of the eigenstates, m, of the *N-1* system

Green's functions and spectral functions

One electron Green's function for a many-body system :

$$G^{-}(k,\omega) = \sum_{m} \frac{\left|\left\langle \Psi_m^{N-1} \left| c_k \right| \Psi_i^{N} \right\rangle\right|^2}{\omega - E_m^{N-1} + E_i^{N} - i\eta}$$

For the 'causal' part of the Green's function, it can also be expressed in terms of the "single particle spectral function", $A(k,\omega)$

$$G(k,\omega) = \int_{-\infty}^{\infty} d\omega' \frac{A\left(k,\omega'\right)}{\omega - \omega' - i\eta} \qquad -\frac{1}{\pi}\operatorname{Im} G^{-}(k,\omega) = A^{-}(k,\omega)$$

$$A^{-}(k,\omega) = \sum_{m} \left|\left\langle \Psi_m^{N-1} \left| c_k \right| \Psi_i^{N} \right\rangle\right|^2 \delta\left(w - E_m^{N-1} + E_i^{N}\right)$$

$$I(\vec{k}, E_{kin}) = \sum_{i,f} |M_{i\to f}|^2 \sum_{m} |\langle \Psi_m^{N-1} | \Psi_i^{N-1} \rangle|^2 \delta(E_{kin} + E_m^{N-1} - E_i^{N} - h\nu)$$

Dipole matrix element

Interesting part!

Energy conservation

The photoemission spectrum contains information about the spectral function!

$$I\left(k, E_{kin}\right) \propto \boxed{M_0}\ \boxed{A^{-}(k,\omega)} \cdot \boxed{f_{FD}(\omega, T)}$$

Matrix Element (constant*)

Spectral Function

Fermi-Dirac function

A convenient way of expressing the spectral function is in terms of the "self-energy", which encodes all the information about the electron interactions

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

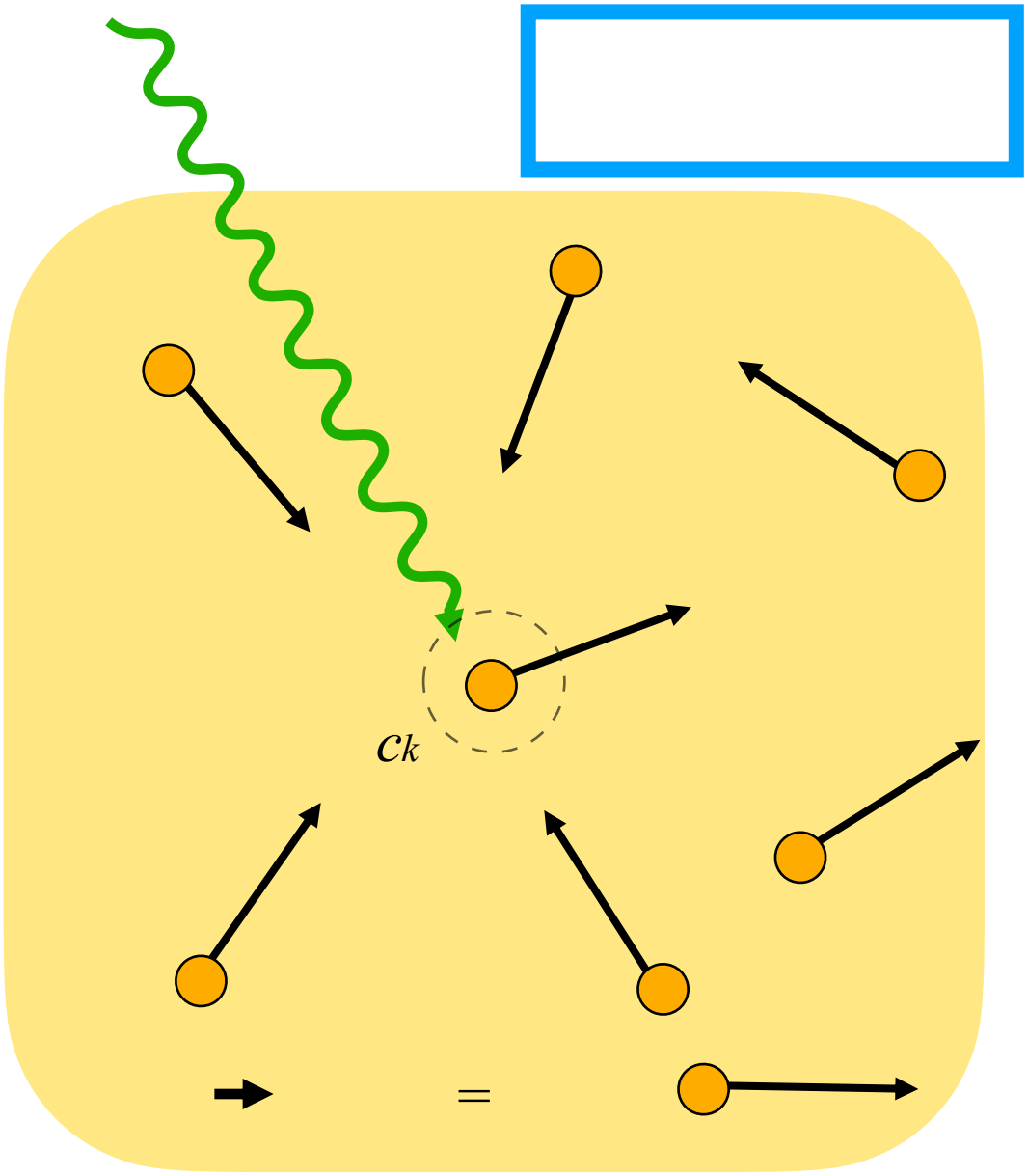
Real part : $\Sigma'(k,\omega)$: energy renormalization

Imaginary part : $\Sigma''(k,\omega)$: quasiparticle lifetime

} *Self-energy is causal; Kramers-Kronig relation*

* Not really, but usually slowly varying as a function of momentum & energy

A graphical interpretation of the photoemission process



For a non-interacting system,

=

i.e. the system does not “relax”

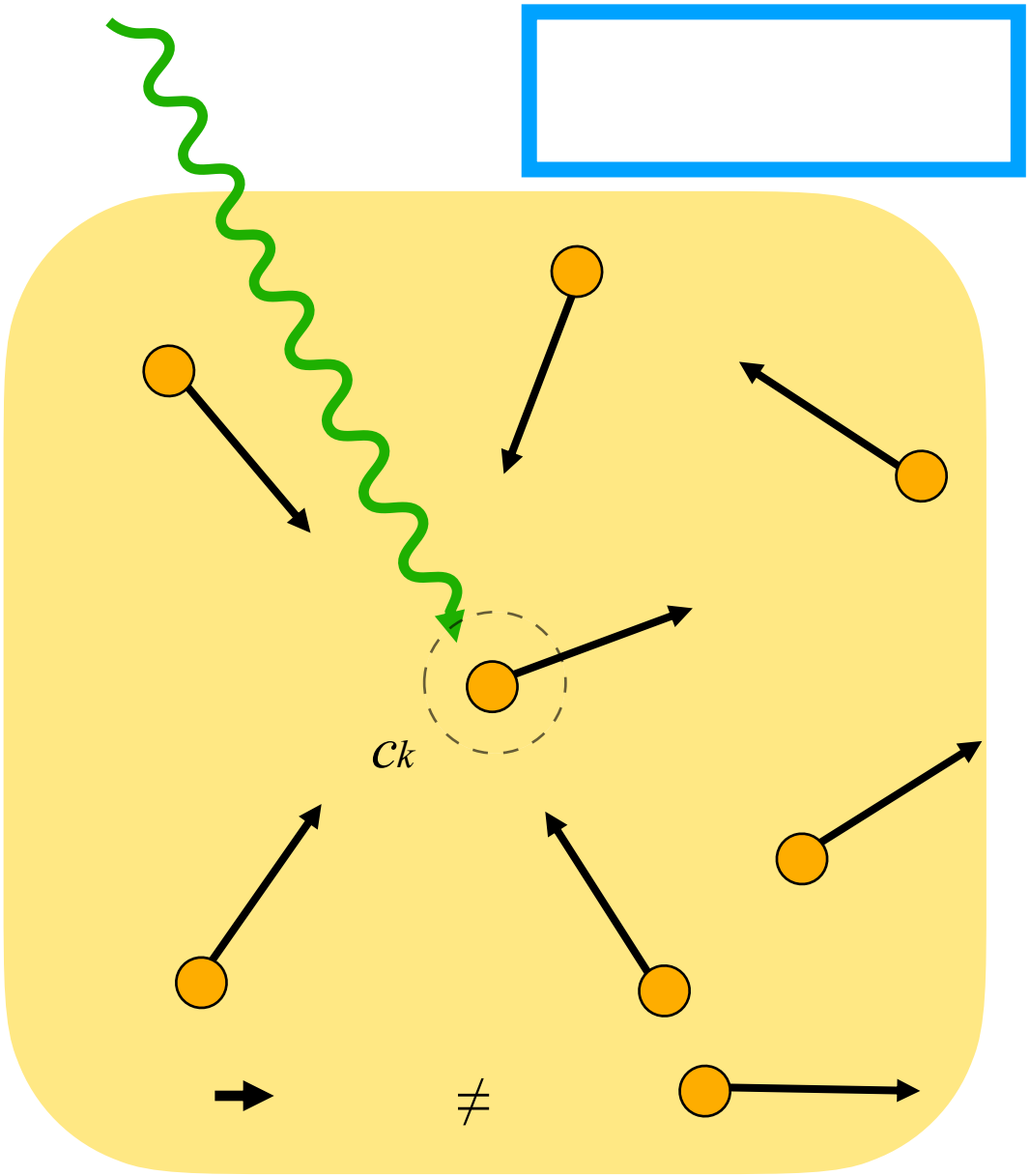
therefore,

is an eigenstate,

of the N-1 system

is a delta-function

A graphical interpretation of the photoemission process



For an interacting system,

$$\neq$$

interactions mean that the eigenstates of the *N-1* system are **NOT** single-electrons

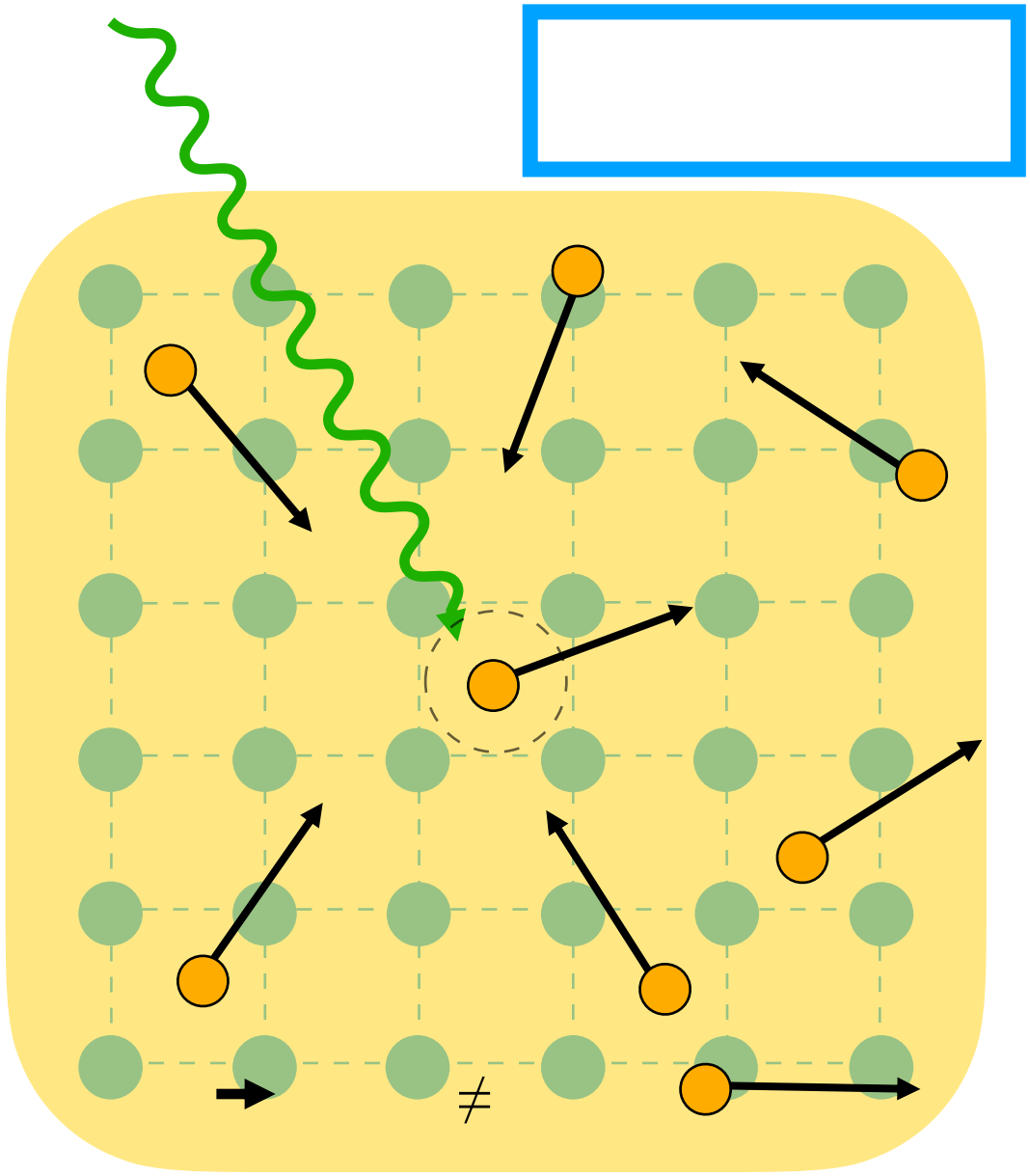
therefore,

is **NOT** an eigenstate,

of the N-1 system

represents the probability that removal of an electron leaves the *N-1* state in eigenstate m

A graphical interpretation of the photoemission process



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Devereaux et al., Phys. Rev. Lett. **93**, 117004 (2004)

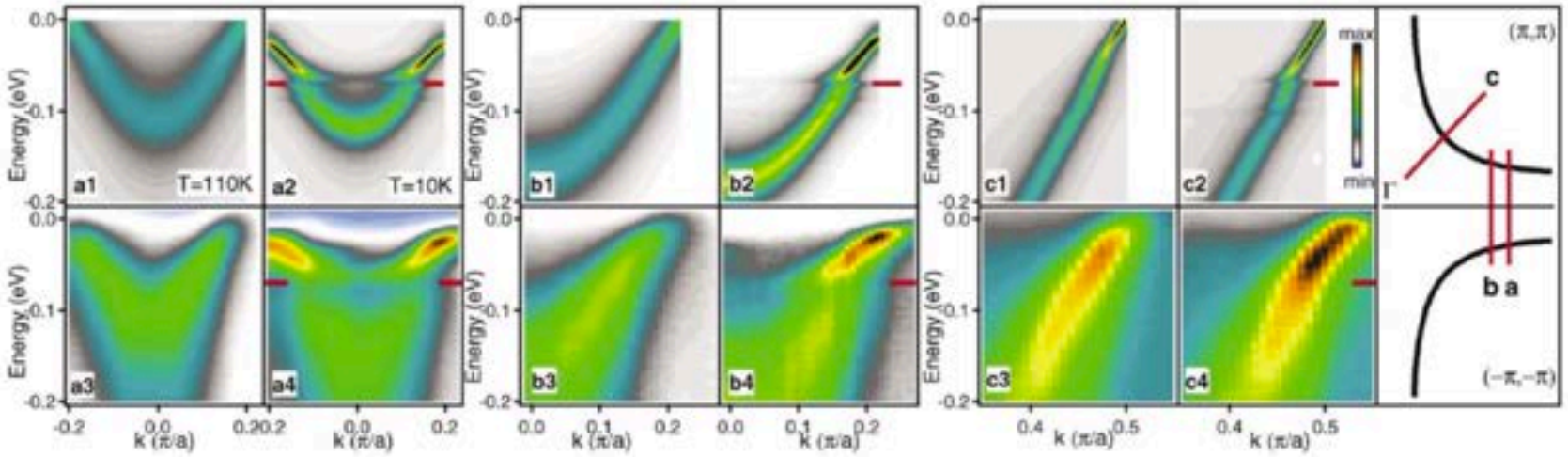


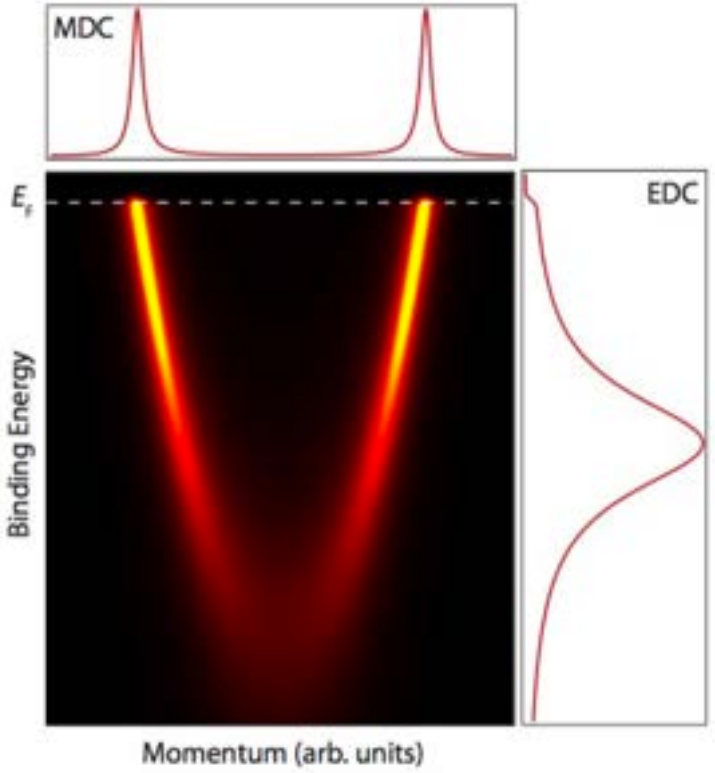
FIG. 3 (color). Image plots of the calculated spectral functions in the normal (a1,b1,c1) and superconducting (a2,b2,c2) states compared to the spectral functions in the normal (a3,b3,c3) and superconducting (a4,b4,c4) states measured in $Bi_2Sr_2Ca_{0.92}Y_{0.08}Cu_2O_{8+\delta}$ (Bi-2212) [6] for momentum cuts *a*, *b*, *c* shown in the rightmost panel and in Fig. 2. The same color scale is used for the normal or superconducting pairs within each cut, but the scaling for the data and the calculation are separate. The red markers indicate 70 meV in the superconducting state.

Many theory groups try to theoretically calculate spectral functions using different approaches & compare with ARPES

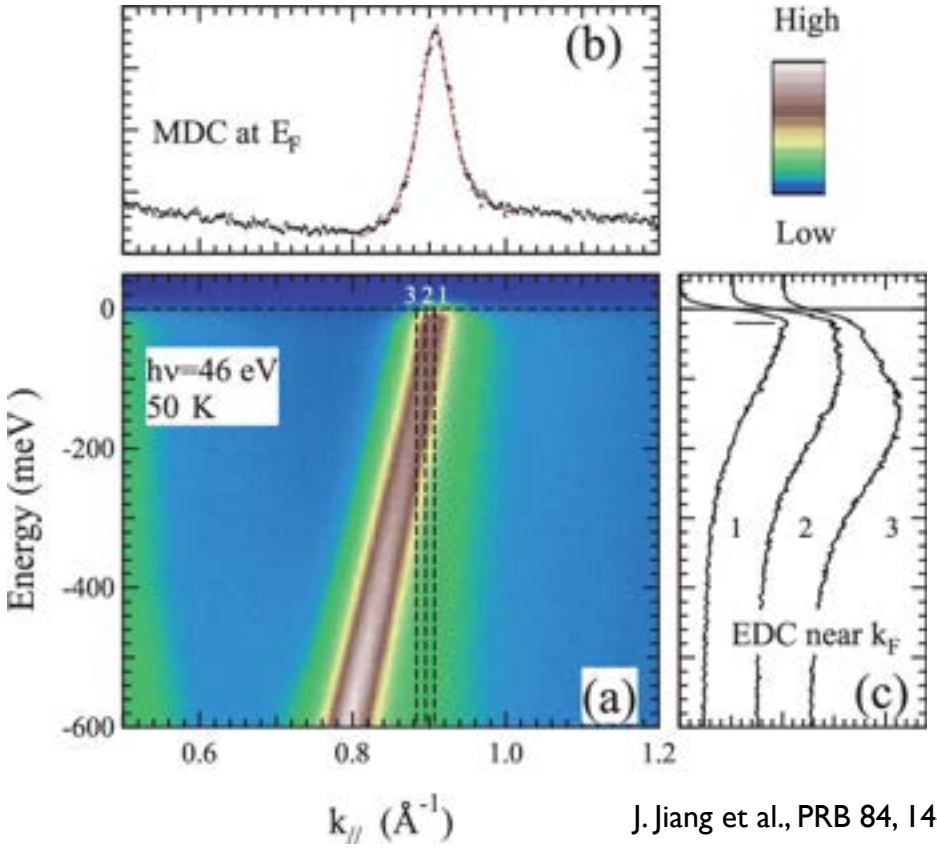
Single-particle spectral function

$$A(\mathbf{k},\omega) \propto \frac{\mathrm{Im}\Sigma(\mathbf{k},\omega)}{[\omega - \varepsilon_k - \mathrm{Re}\Sigma(\mathbf{k},\omega)]^2 + [\mathrm{Im}\Sigma(\mathbf{k},\omega)]^2}$$

"EDC" : Energy Distribution Curve (a vertical slice of the image)
"MDC" : Momentum Distribution Curve (a horizontal slice)



From D. Shai's thesis



J. Jiang et al., PRB 84, 144124

MDCs : Good for dispersion tracking; steeply dispersing bands
EDCs : Good for lineshape analysis; analyzing flat bands

Fermi Liquid Theory

Backbone for Single Particle Approximations

Band Structure of Metals,
Semiconductors, Insulators

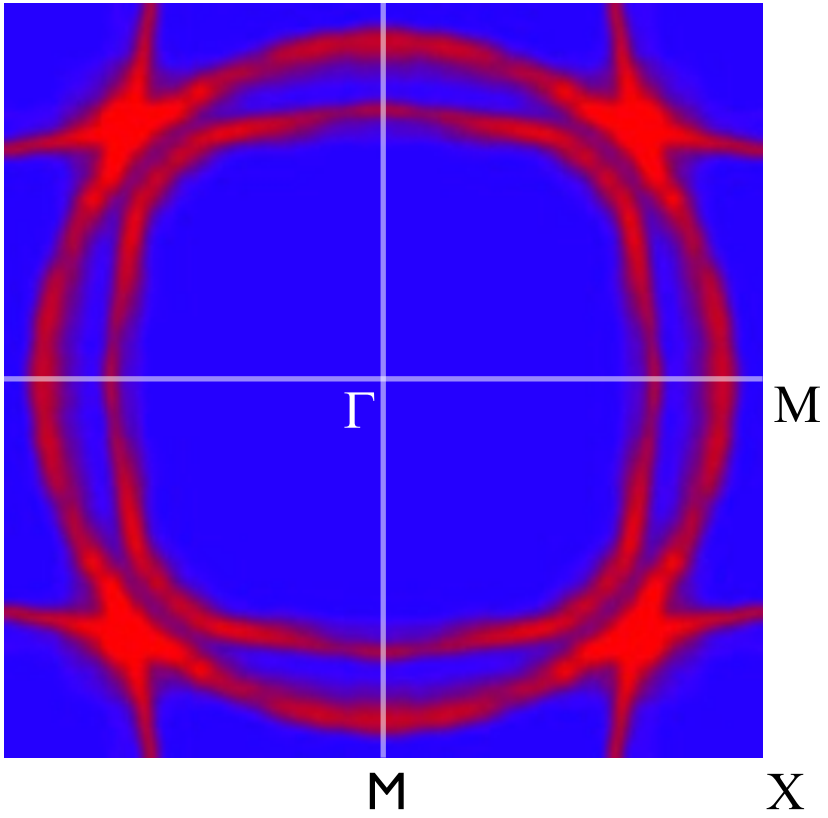
What can we test in FL Theory by ARPES?

Luttinger Volume

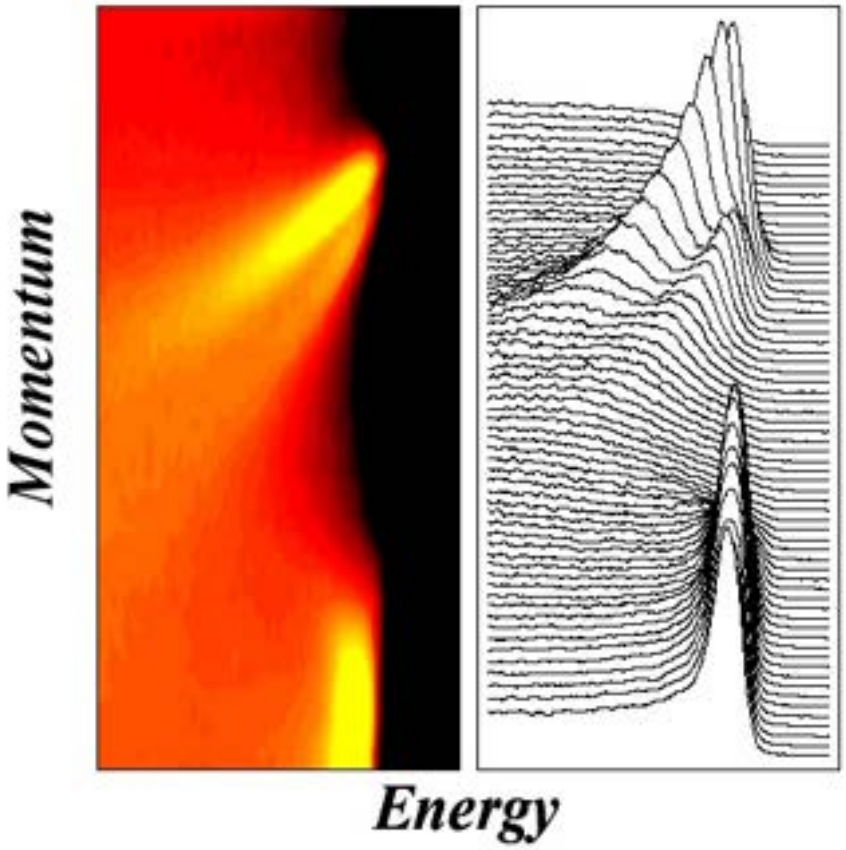
Sharp coherent quasiparticles ($\mathrm{Im}\Sigma\,(\omega) < \varepsilon_k$)

Energy dependence of $\mathrm{Im}\Sigma\,(\omega)$

Fermi Surface from ARPES



Quasiparticles in Sr_2RuO_4



K.M. Shen, A. Damascelli, *et al. Phys. Rev. B* (2001)
A. Damascelli, D.H. Lu, K.M. Shen, *et al., Phys. Rev. Lett.* (2000)

Electron-electron interactions in Sr_2RuO_4

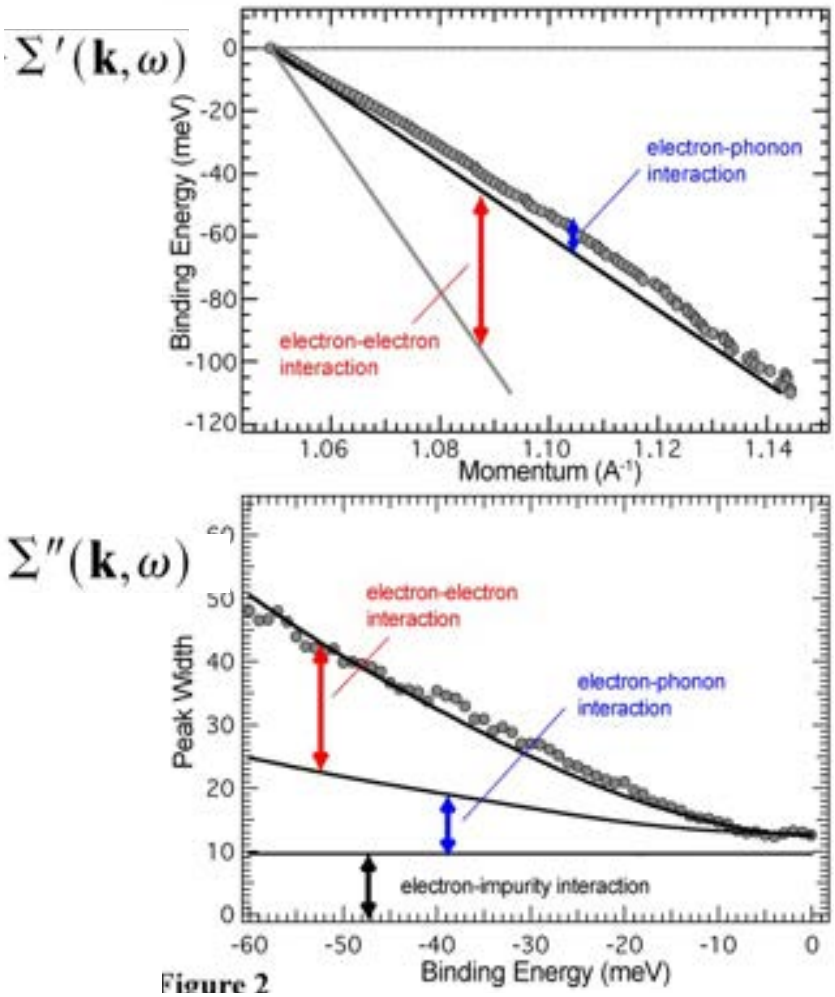
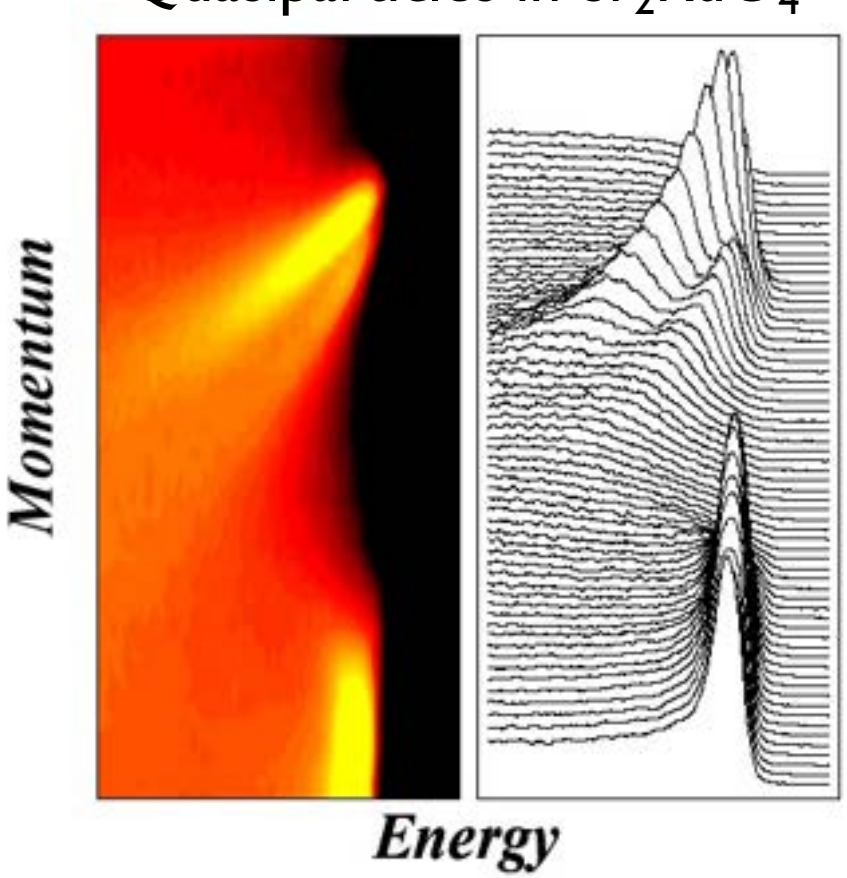


Figure 2

Quasiparticles in Sr_2RuO_4



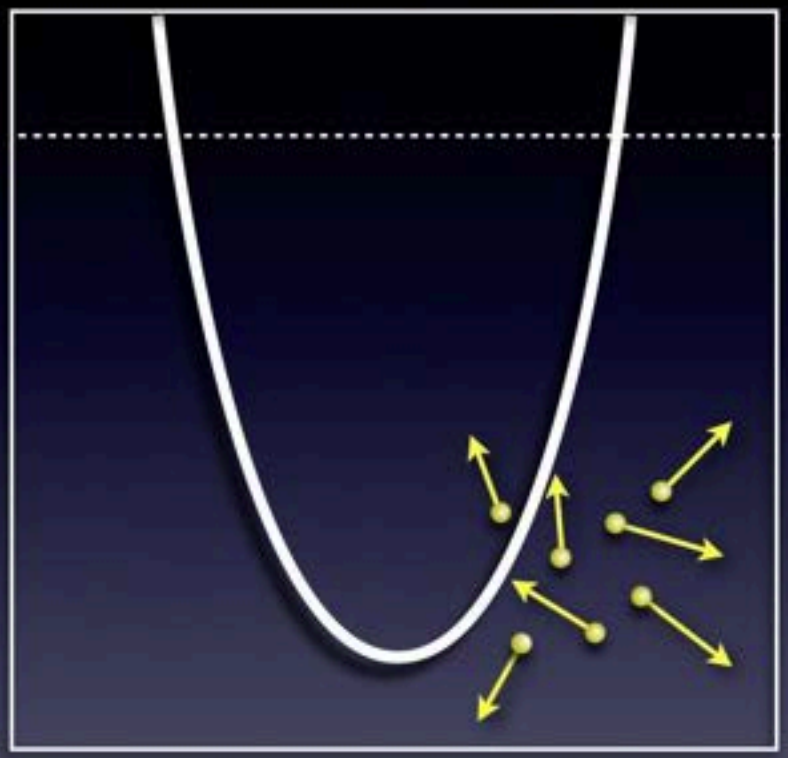
- $\mathrm{Im}\Sigma < |E - E_F|$:Well-defined quasiparticles
- Electron-electron scattering dominates ov ' ron-phonon interactions

N.J.C. Ingle, K.M. Shen, *et al.*, Phys. Rev B (2006)

Mass renormalization due to electron-electron interactions

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

electrons' effective mass can be extracted from band curvature (or velocity & k_F)



Momentum

Typically estimate mass renormalization by comparing the experimental Fermi velocity (slope) with the predicted velocity *without interactions**

*usually DFT calculations

Mass renormalization due to electron-electron interactions

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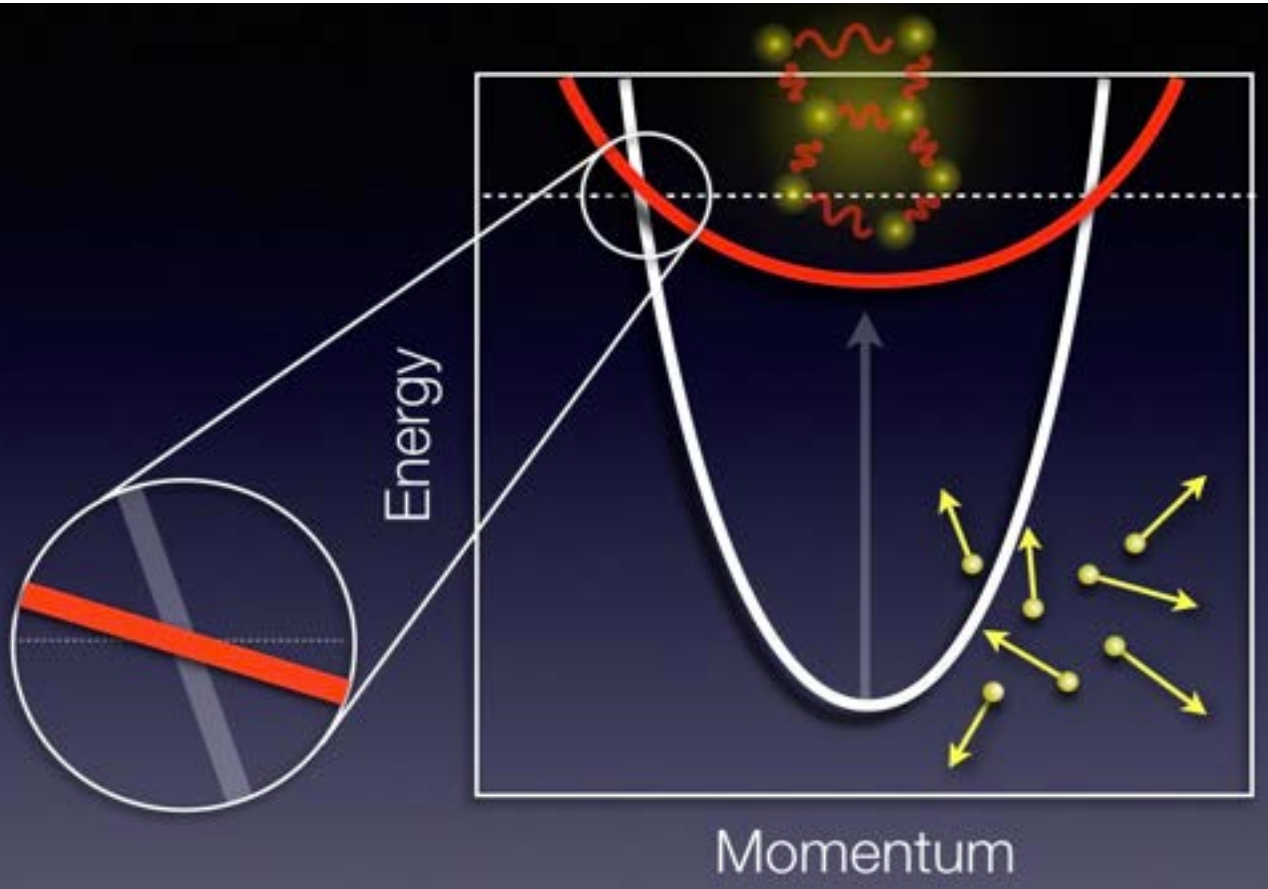

Typically estimate mass renormalization by comparing the experimental Fermi velocity (slope) with the predicted velocity *without interactions**

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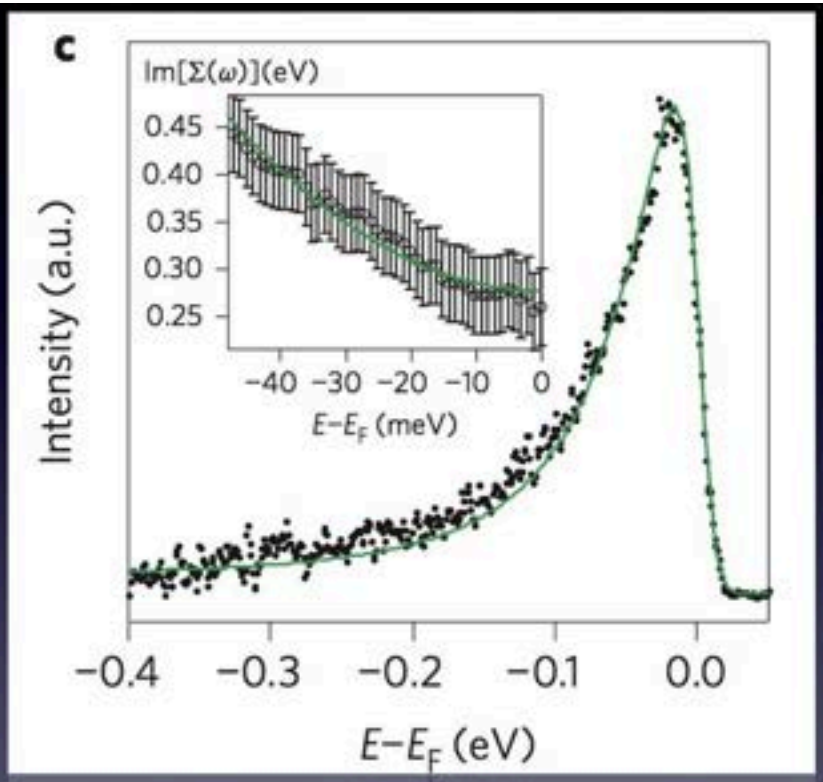
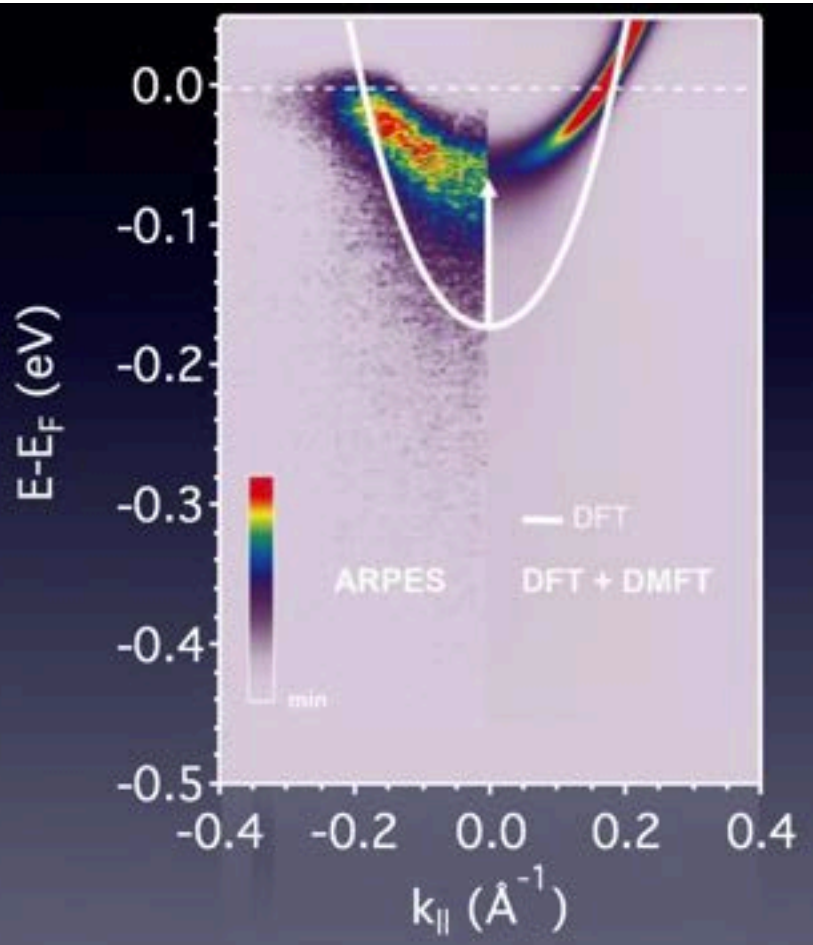
Electron-electron interactions in $LaNiO_3$



$$\frac{m^*}{m} = 3.3 \pm 0.5$$

in agreement with optical & thermodynamic measurements



P.D.C. King *et al*, *Nature Nanotechnology* **9**, 443 (2014)

E.A. Nowadnick, J.P. Ruf et al., *Phys. Rev. B* **92**, 245109 (2015)

P.D.C. King *et al*, *Nature Nanotechnology* **9**, 443 (2014)

You would like to know whether a particular material that you are studying has well-defined coherent "quasiparticle" excitations. You investigate the width, Γ, of the ARPES peaks as a function of binding energy, ω, from the Fermi level (E_F is $\omega = 0$).

Which of the following would be consistent with well-defined, coherent quasiparticles?

1. $\Gamma \alpha \omega^{-1/2}$
2. $\Gamma \alpha \omega^{1/2}$
3. $\Gamma \alpha \omega^{3/2}$
4. $\Gamma \alpha \omega^{2}$
5. $\Gamma \alpha \omega^{3}$

A. 4 only

B. 3, 4 and 5

C. 2, 3, 4, and 5

D. 4 and 5

E. All of the above

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A. 4 only

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Let's assume we are trying to measure a superconducting gap by ARPES in a material whose T_c = 7 K and energy gap Δ = 1 meV.

Our measurement is occurring at T = 4 K, and we are using an energy resolution of ΔE = 5 meV.

Is this a complete waste of time (and money)?

A. Yes

B. No

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A. Yes

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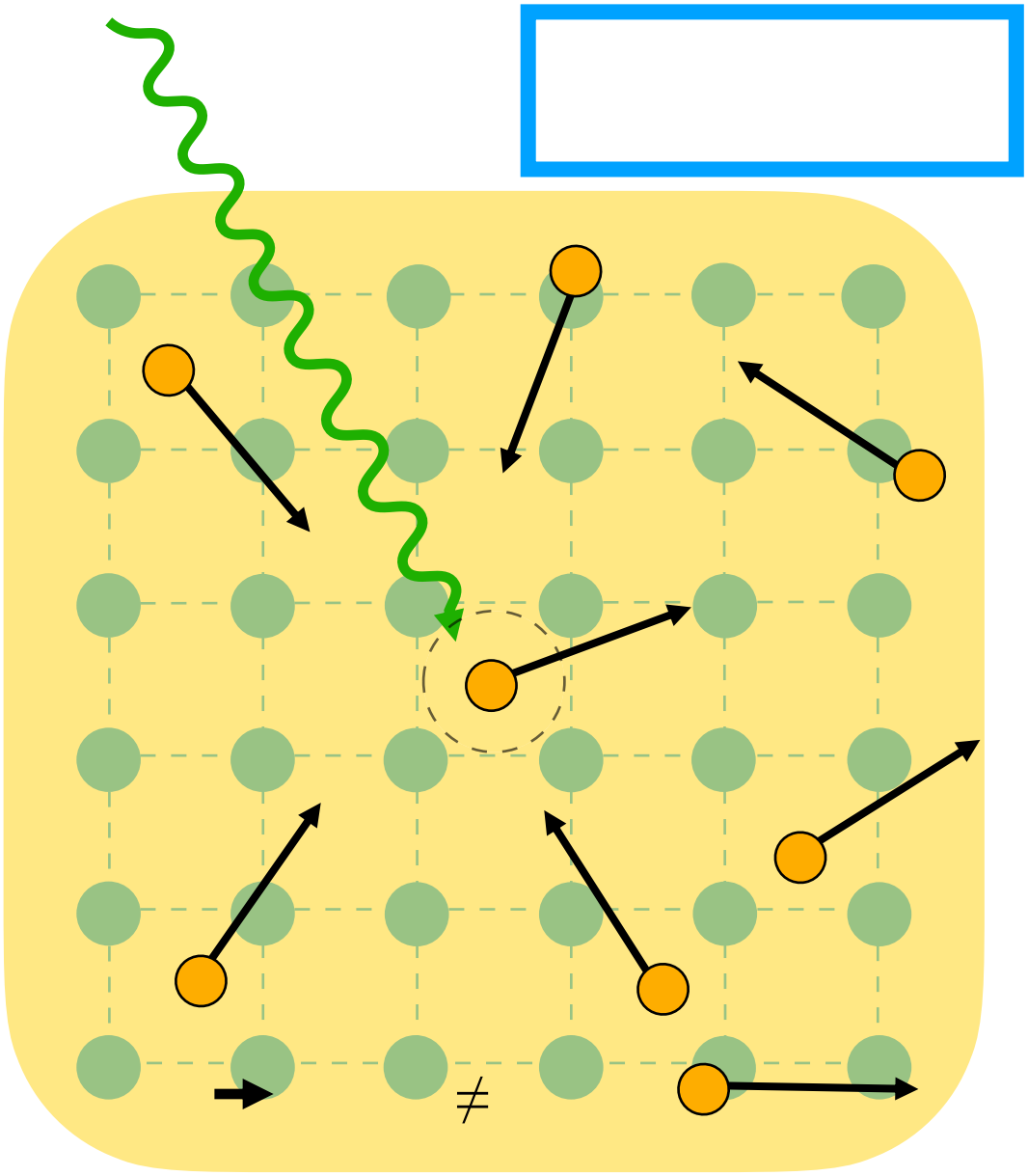
What about electron-lattice interactions?



For an interacting system,

$\neq$

interactions mean that the eigenstates of the *N-1* system are **NOT** single-electrons

therefore,

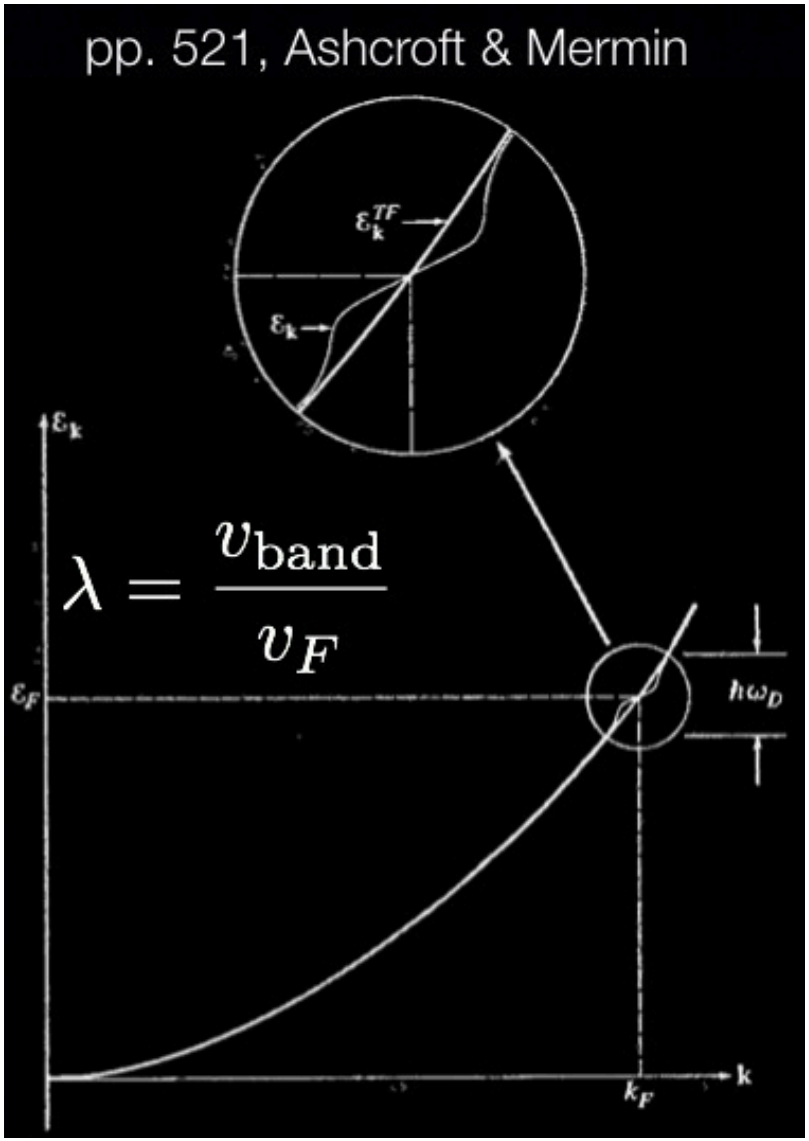
is **NOT** an eigenstate,

of the N-1 system

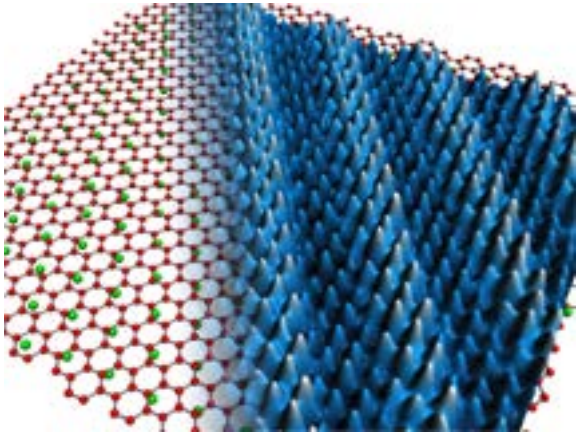
represents the probability that removal of an electron leaves the *N-1* state in eigenstate m

Electron-phonon coupling in solids

pp. 521, Ashcroft & Mermin

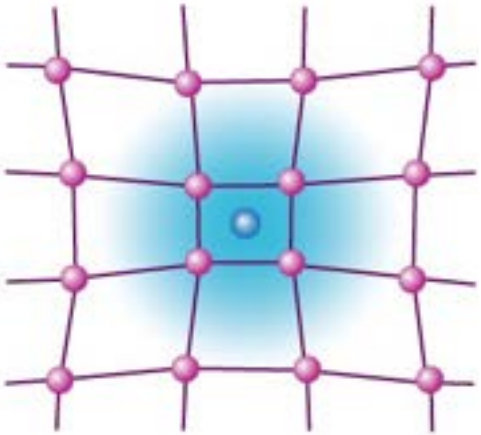


Charge Density Waves

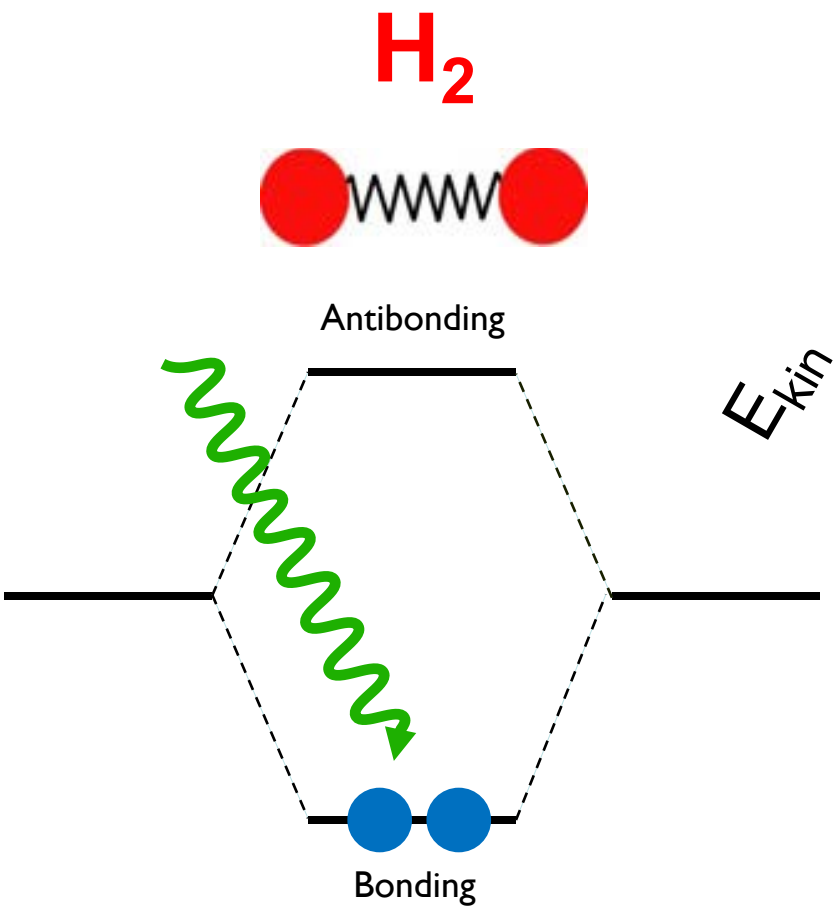


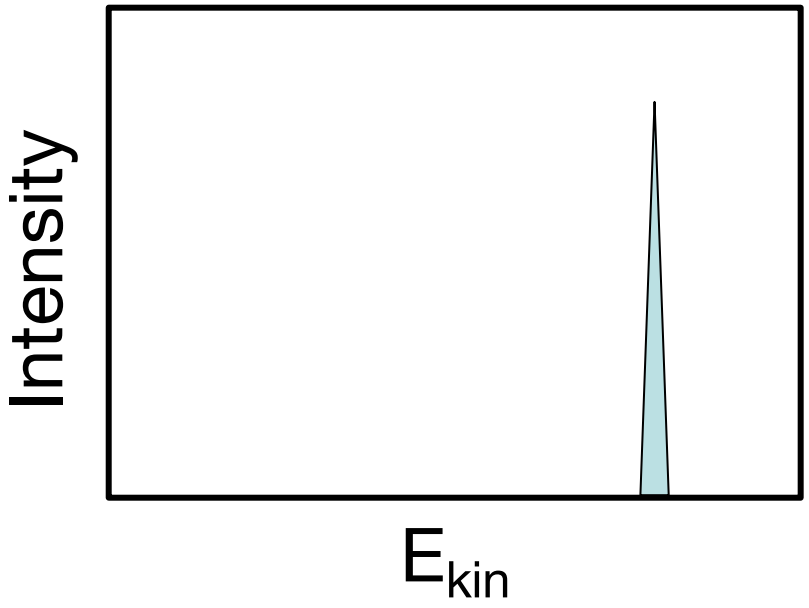
Superconductivity

Polarons



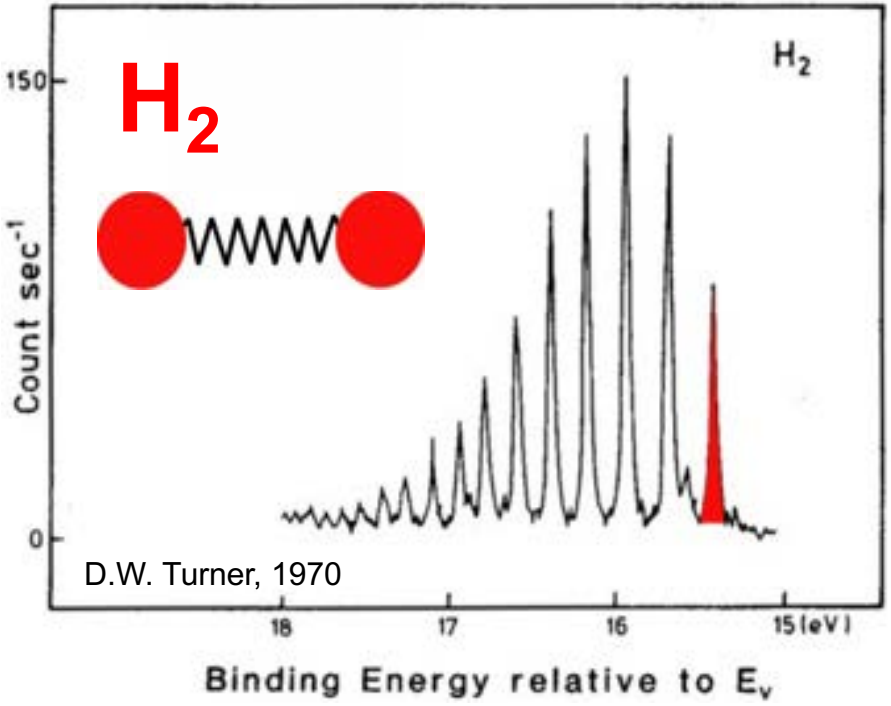
Consider Photoemission from a Hydrogen Molecule





Citrin (1974), Sawatzky (1989), Perebeinos & Allen (2000), Perfetti (2001,2002)

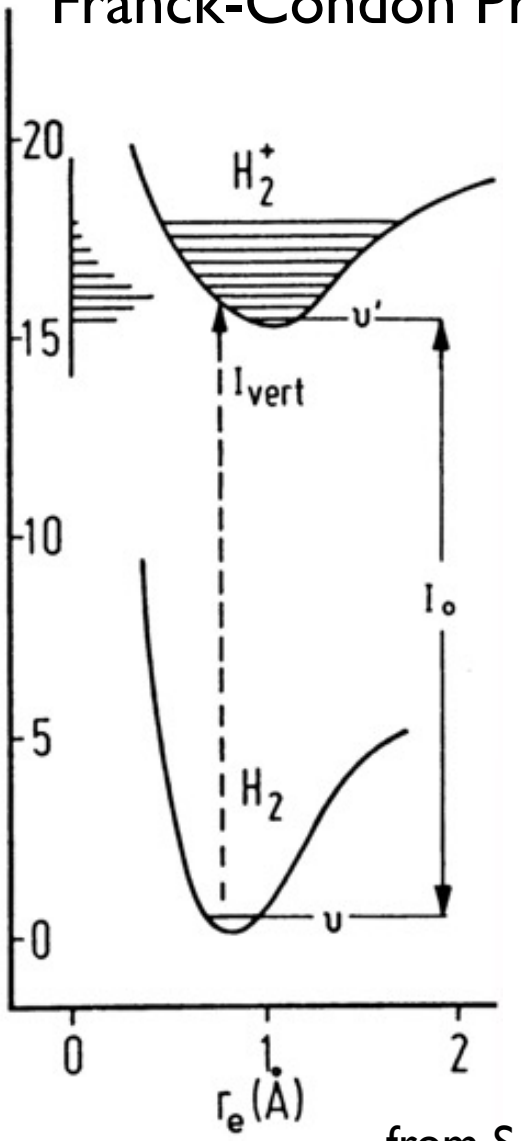
Photoemission from molecular hydrogen



- Most spectral weight in shake-off peaks (*incoherent* part of spectral function)

- Only lowest-energy transition (0-0) analogous to quasiparticle

Franck-Condon Process



from S. Hufner

Citrin (1974), Sawatzky (1989), Perebeinos & Allen (2000), Perfetti (2001,2002)

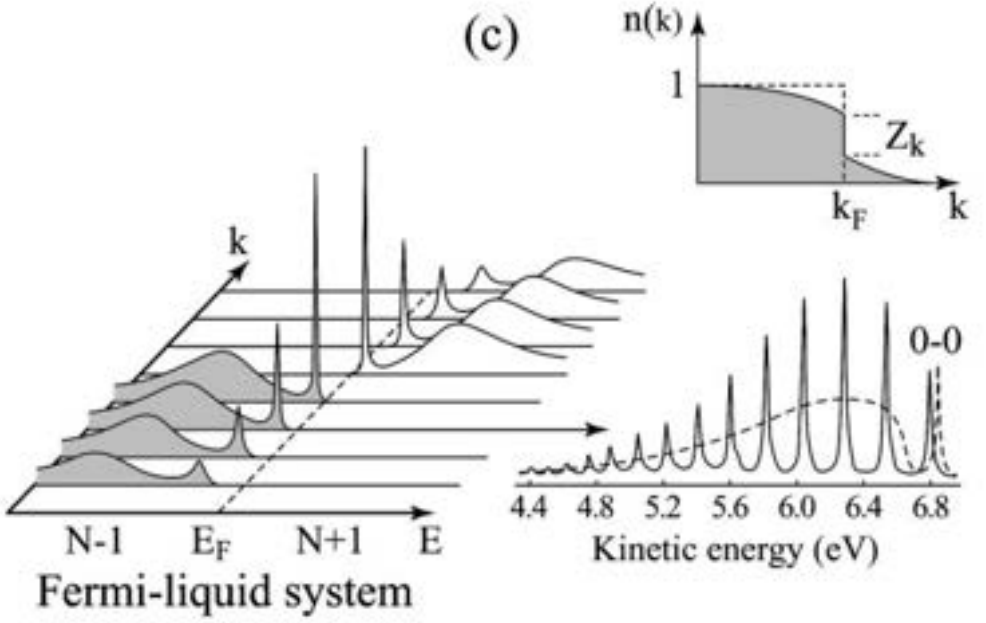
NEWS AND VIEWS

HIGH-TEMPERATURE SUPERCONDUCTIVITY

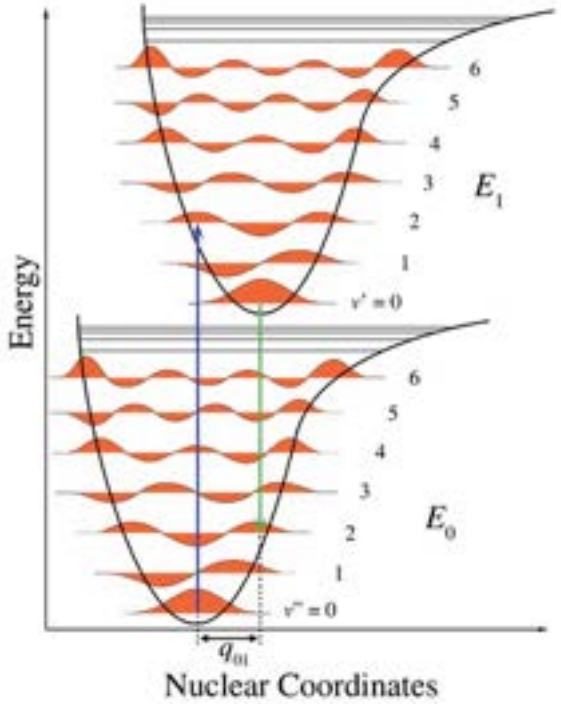
Nature 342, 480 (1989)

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."

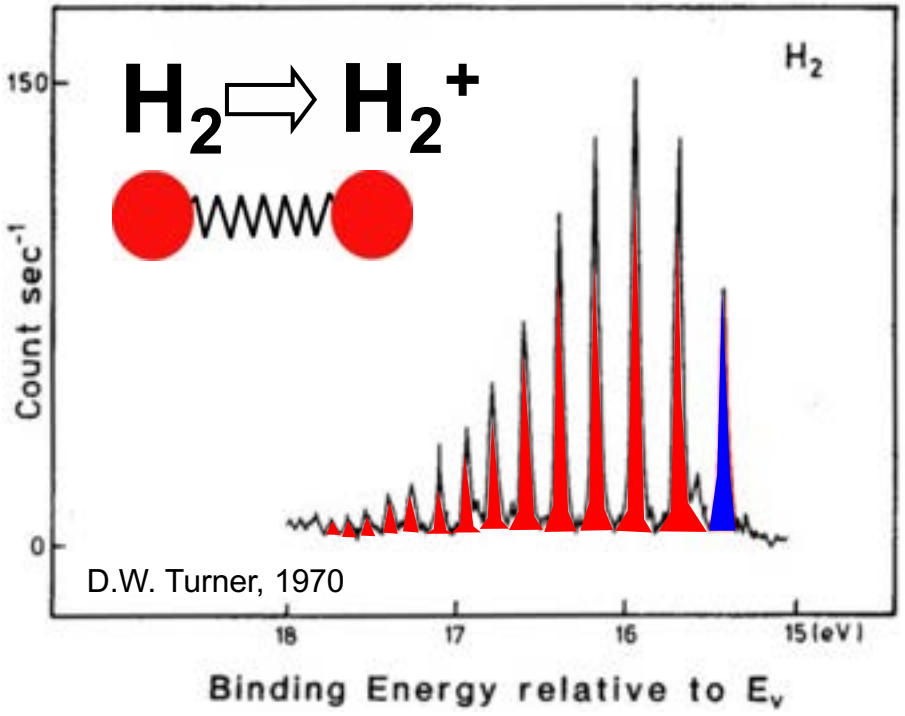
Franck-Condon Effect & electron-phonon coupling

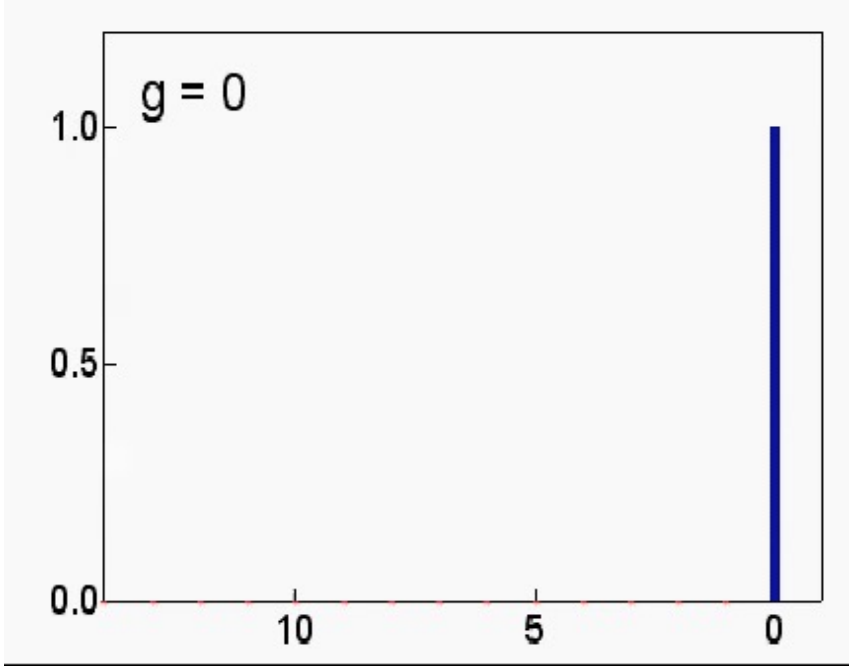
$$\mathcal{H} = \epsilon_0\, c^\dagger c + \sum_{\mathbf{q}} \omega_{\mathbf{q}}\, a^\dagger_{\mathbf{q}} a_{\mathbf{q}} + \sqrt{g\omega_0^2} \sum_{\mathbf{q}} c^\dagger c (a_{\mathbf{q}} + a^\dagger_{-\mathbf{q}})$$

electron energy

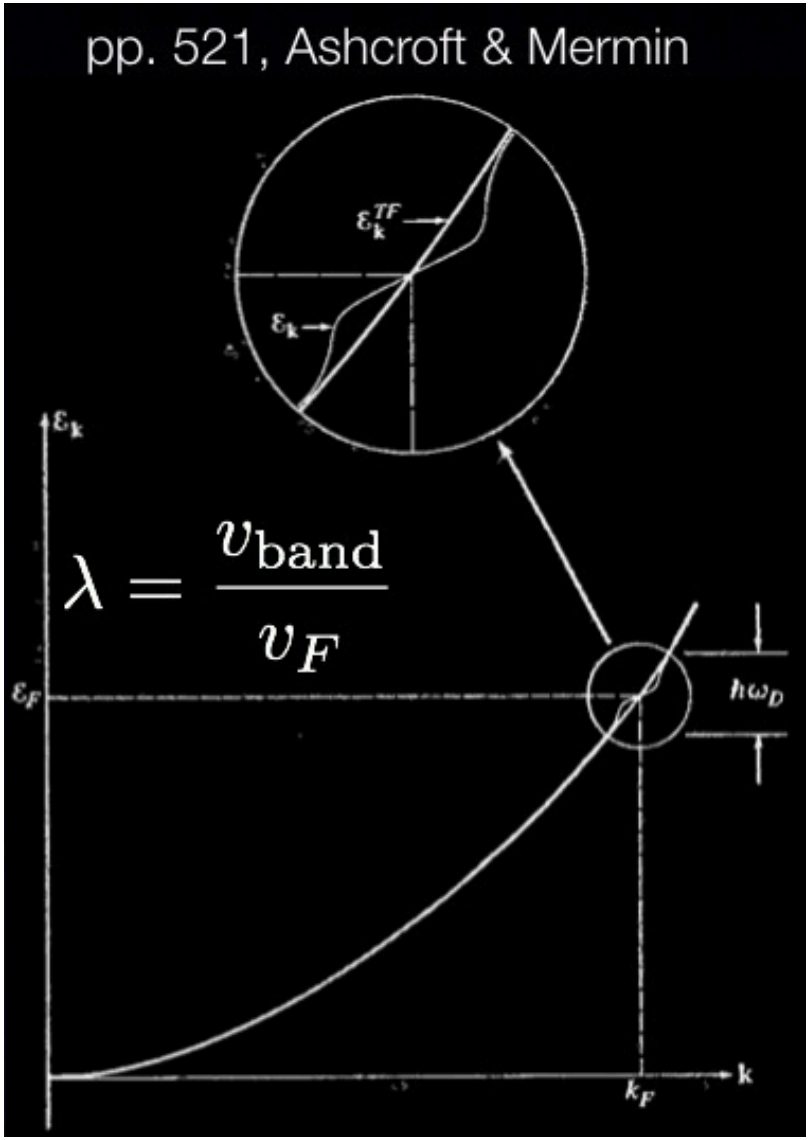
phonon energy

electron-phonon coupling





Electron-phonon coupling in solids : ARPES

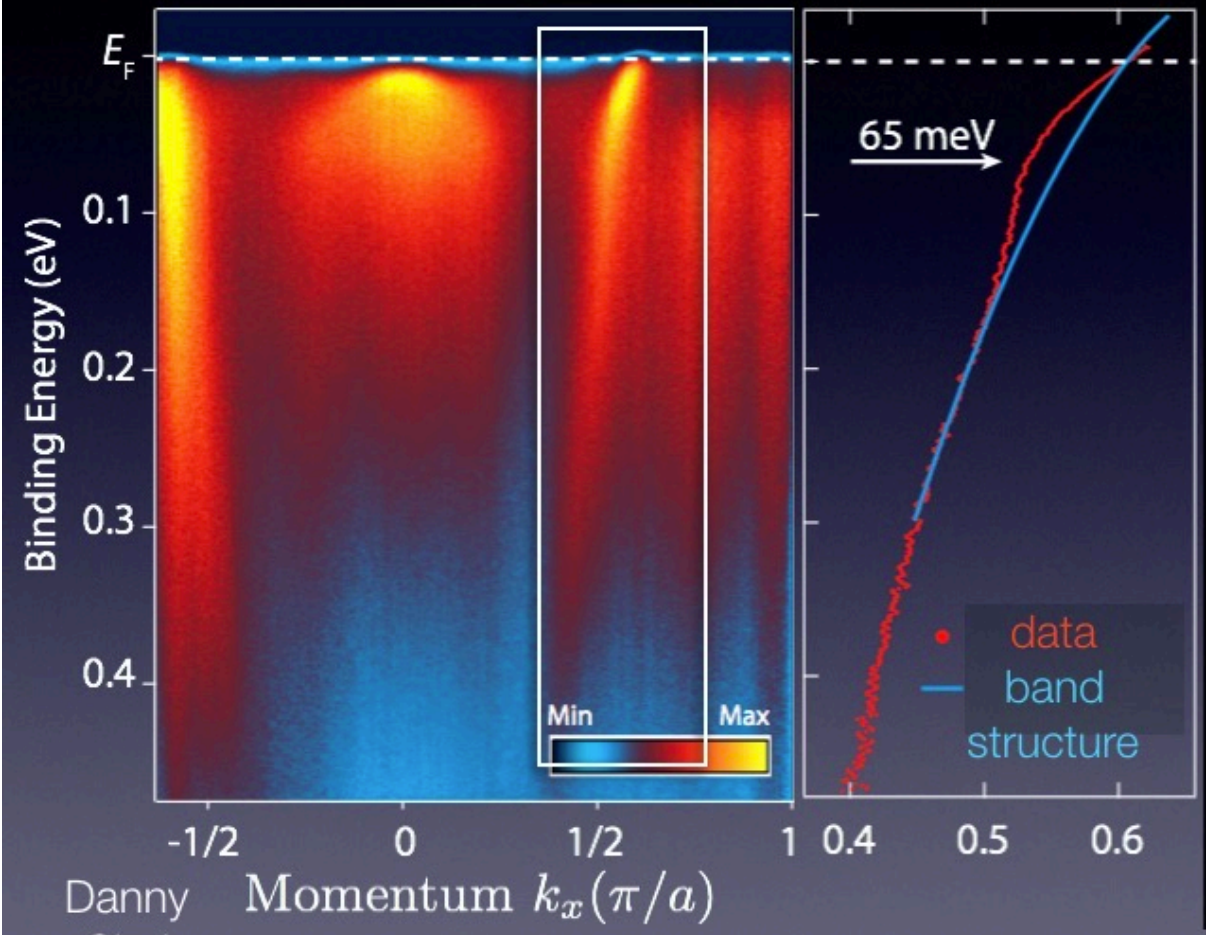


Be (0001) : Electron-phonon

M. Hengsberger *et al.*, PRL ('99)

A real example of electron-phonon interactions

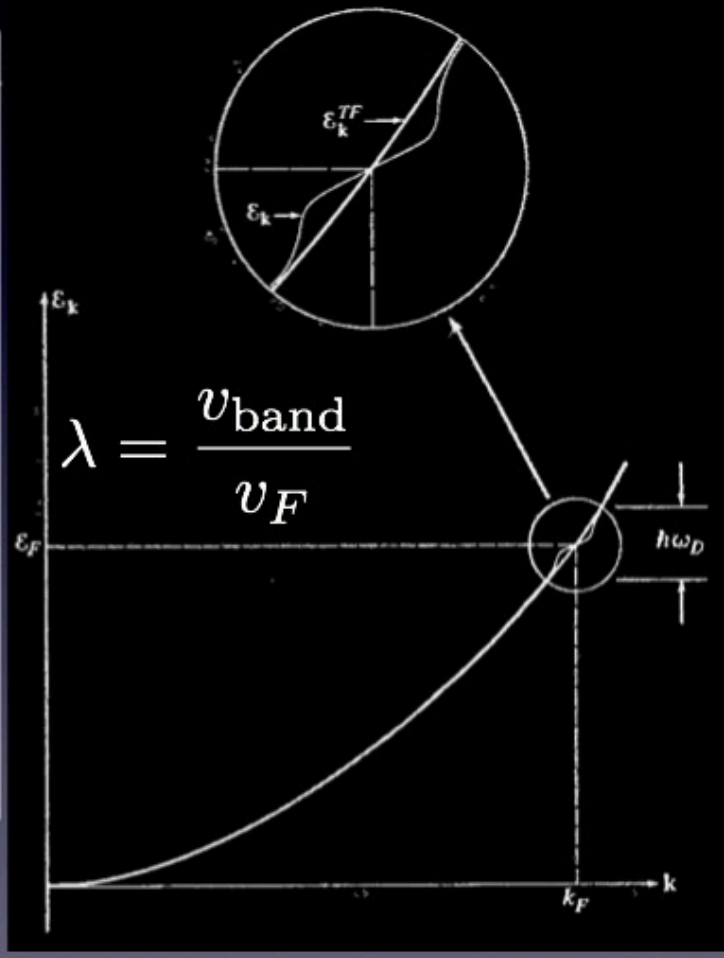
$SrRuO_3$: correlated oxide



Danny Shai

D.E. Shai et al., *Phys. Rev. Lett.* 110, 087004 (2013)

pp. 521, Ashcroft & Mermin

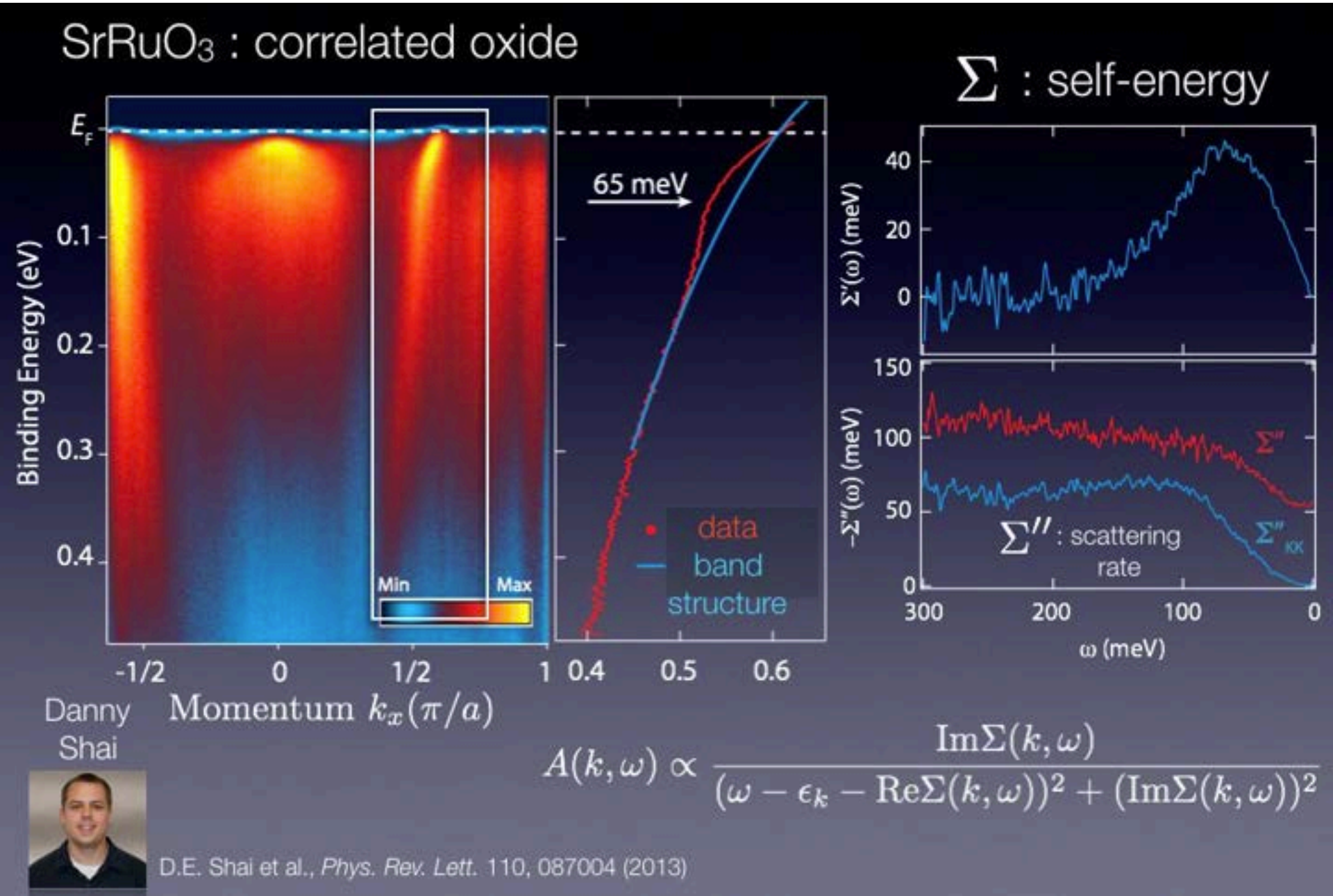


$$\lambda = \frac{v_{\text{band}}}{v_F}$$

$$\frac{m^*}{m_b} = 3.9$$

good agreement with values from transport ($m^*/m_b = 4.1$)

A real example of electron-phonon interactions



Danny Shai

$$A(k,\omega) \propto \frac{\mathrm{Im}\Sigma(k,\omega)}{(\omega - \epsilon_k - \mathrm{Re}\Sigma(k,\omega))^2 + (\mathrm{Im}\Sigma(k,\omega))^2}$$

D.E. Shai et al., *Phys. Rev. Lett.* 110, 087004 (2013)

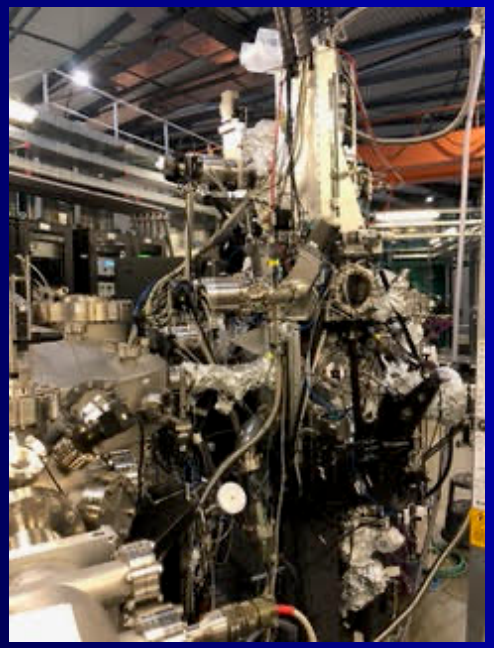
Textbooks

- Photoelectron Spectroscopy : Principles & Applications by Stefan Hufner (Springer-Verlag)

Review articles

- Angle-resolved photoemission studies of the cuprate superconductors. (A. Damascelli, Z. Hussain and Z.-X. Shen. Rev. Mod. Phys. 75, 473, 2003)
- ARPES : A Probe of Electronic Correlations (R. Comin and A. Damascelli, arXiv:1303.1438)

LOREA Beamline @ ALBA



- High spatial resolution (~ 20 microns spot size with microscope)
- Advanced sample preparation capabilities (annealing, vacuum transfer)
- Spin-resolved detection (installed & coming online this spring)
- Wide photon energy range (20 – 500^{+} eV)

approximately 50 synchrotrons worldwide

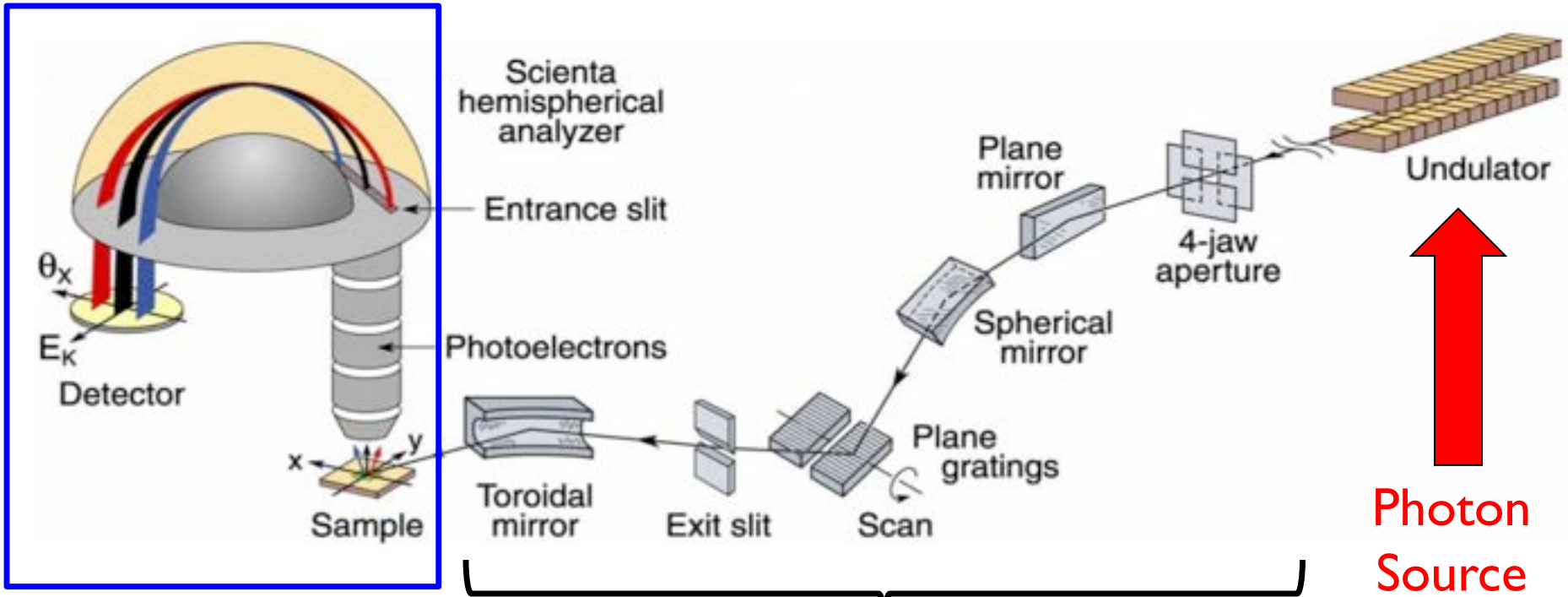
Beamtime at synchrotrons is free of charge – granted based on merit of proposal

Considerations to take into account regarding where to apply :

- what photon energy range, resolution, polarization is needed?
- sample handling capabilities and specialization of endstation
- demand on endstation / beamline
- ease of use / productivity of facility

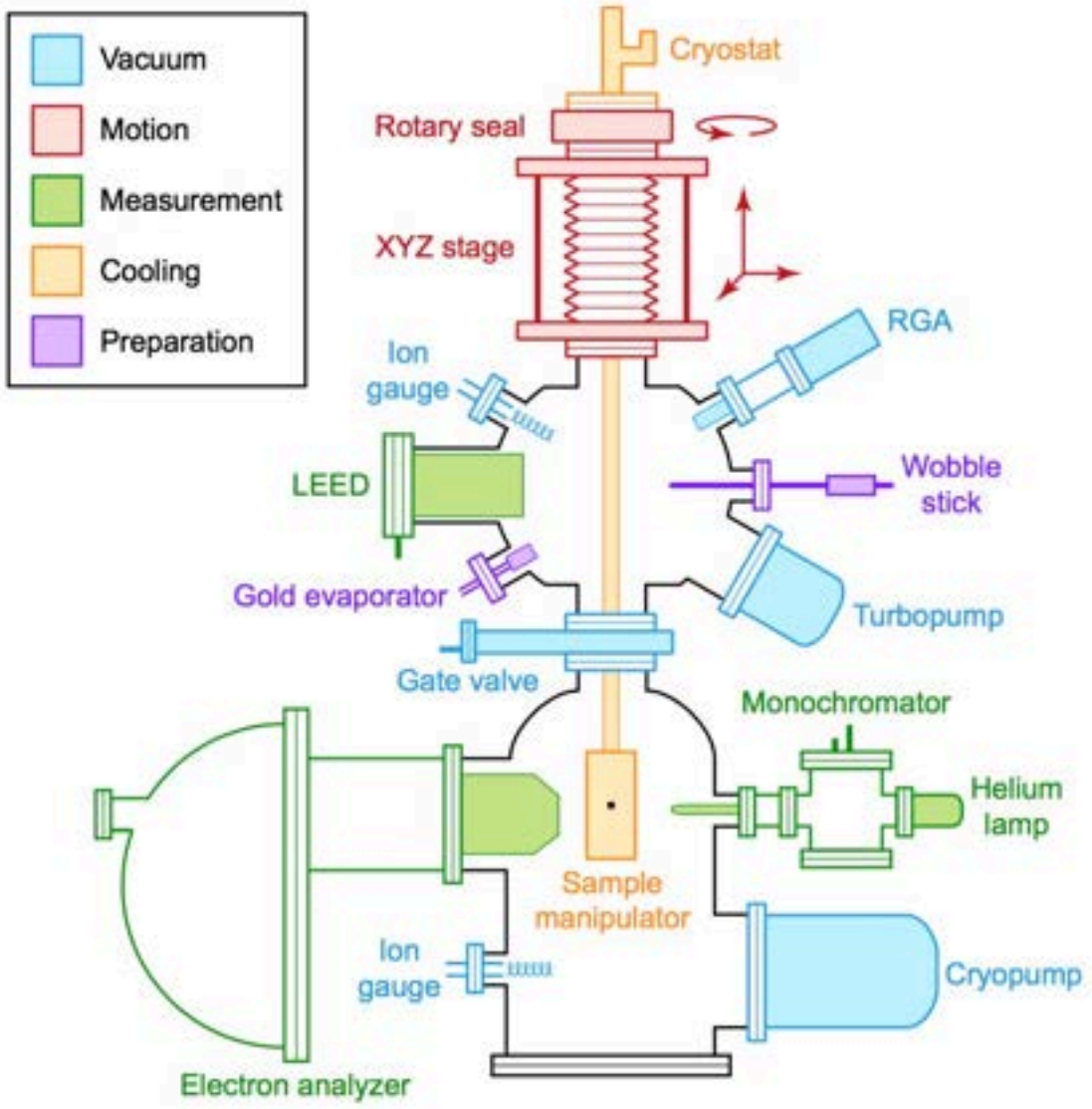
graphic from Argonne National Laboratory

layout of a synchrotron beamline



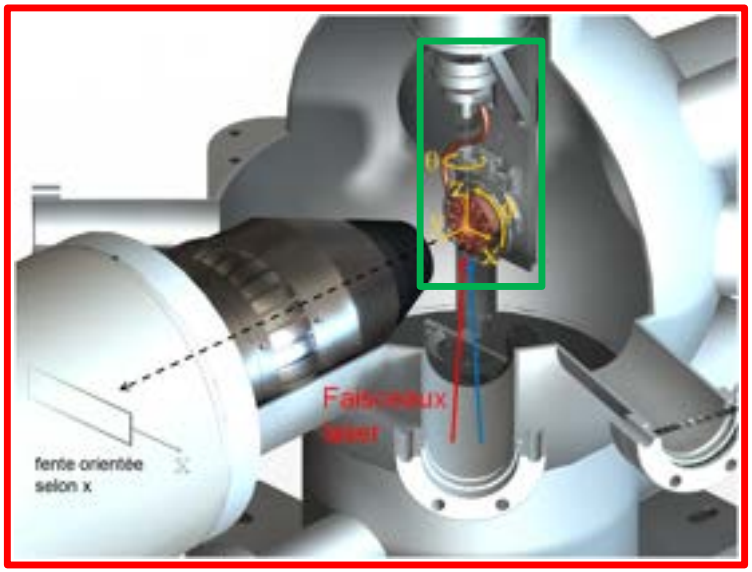
- No good windows for "soft" x-ray, VUV range (10-1000 eV)
- Reflectivities of mirrors in VUV, soft x-ray range of ~ 50%
- Typical resolving powers ($\Delta E / E$) of ~ 5000 : 1

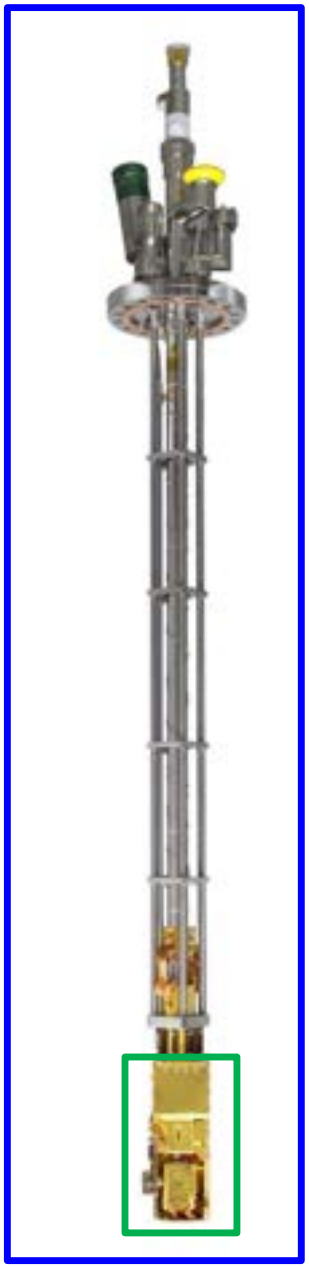
ARPES chamber : Block Diagram



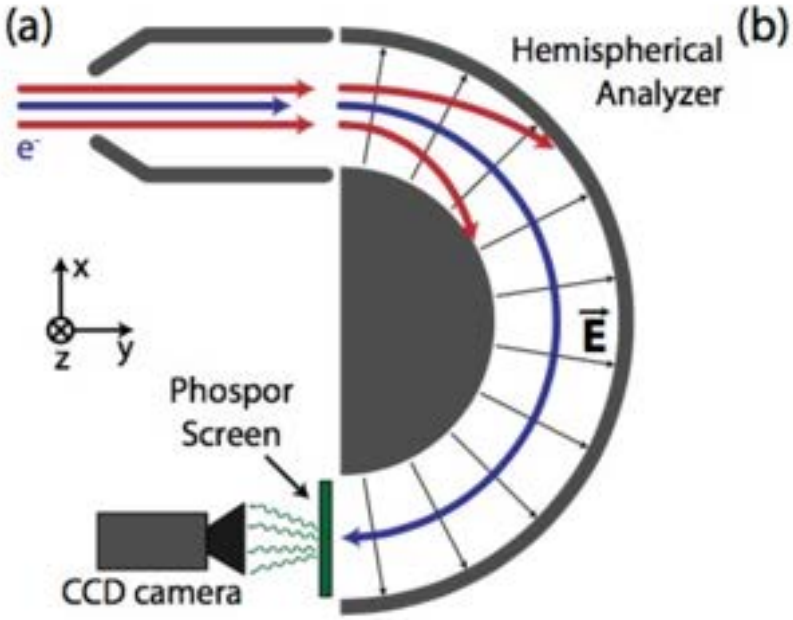
From the thesis of John Harter

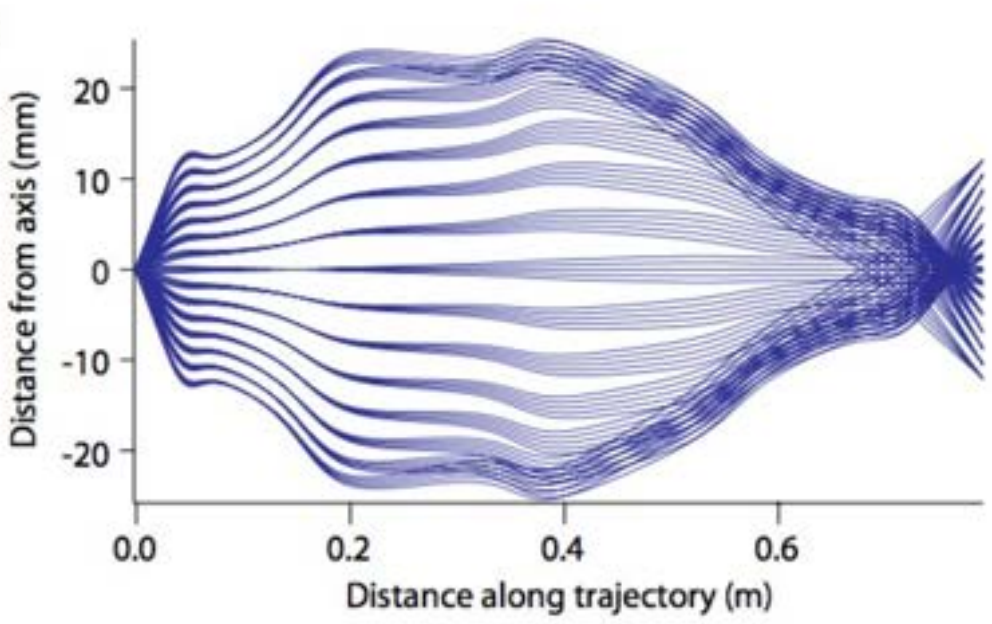
A typical ARPES chamber





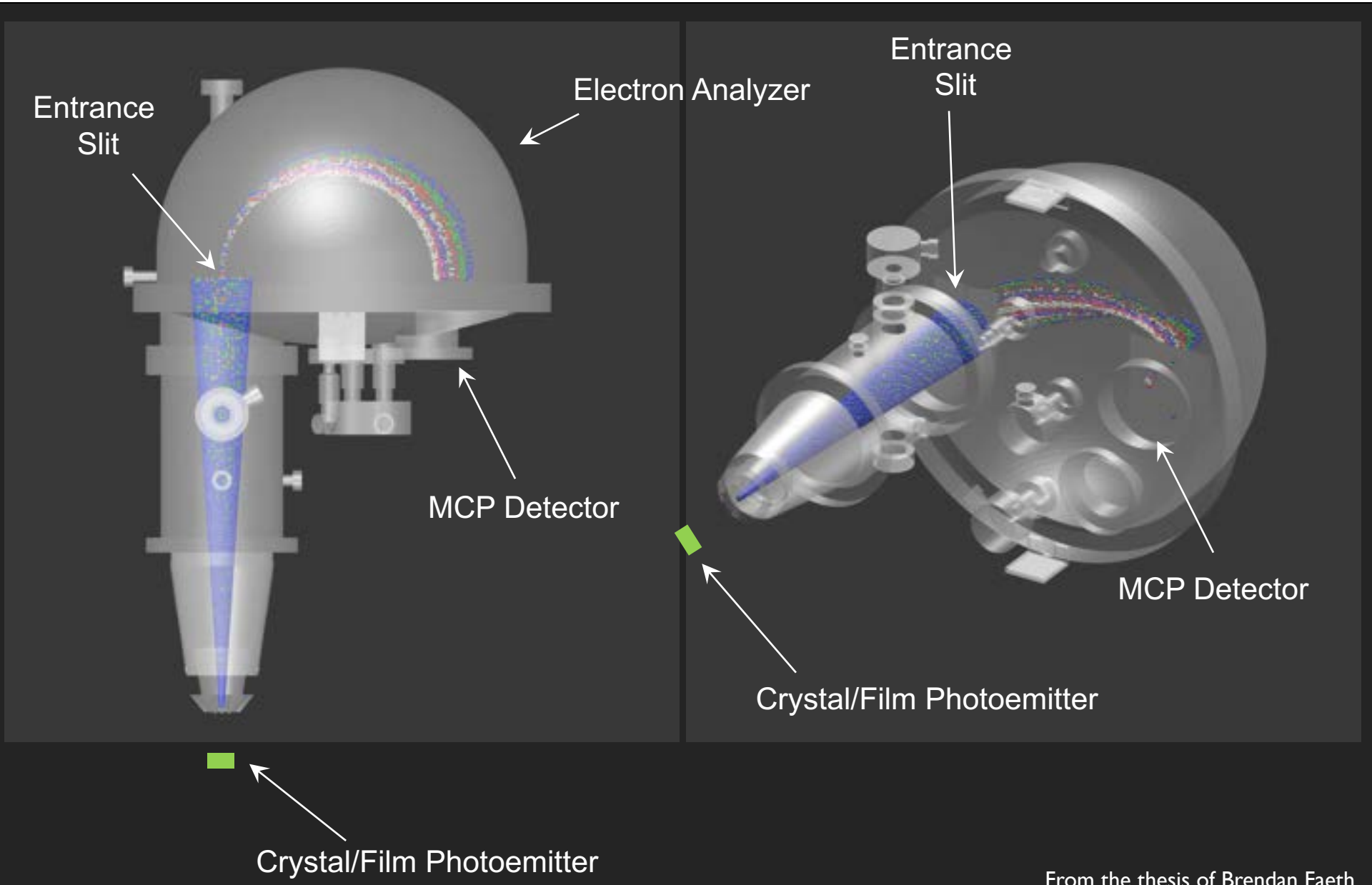
Inside the Electron Analyzer





From the thesis of Haofei Wei

ARPES electron analyzer in action

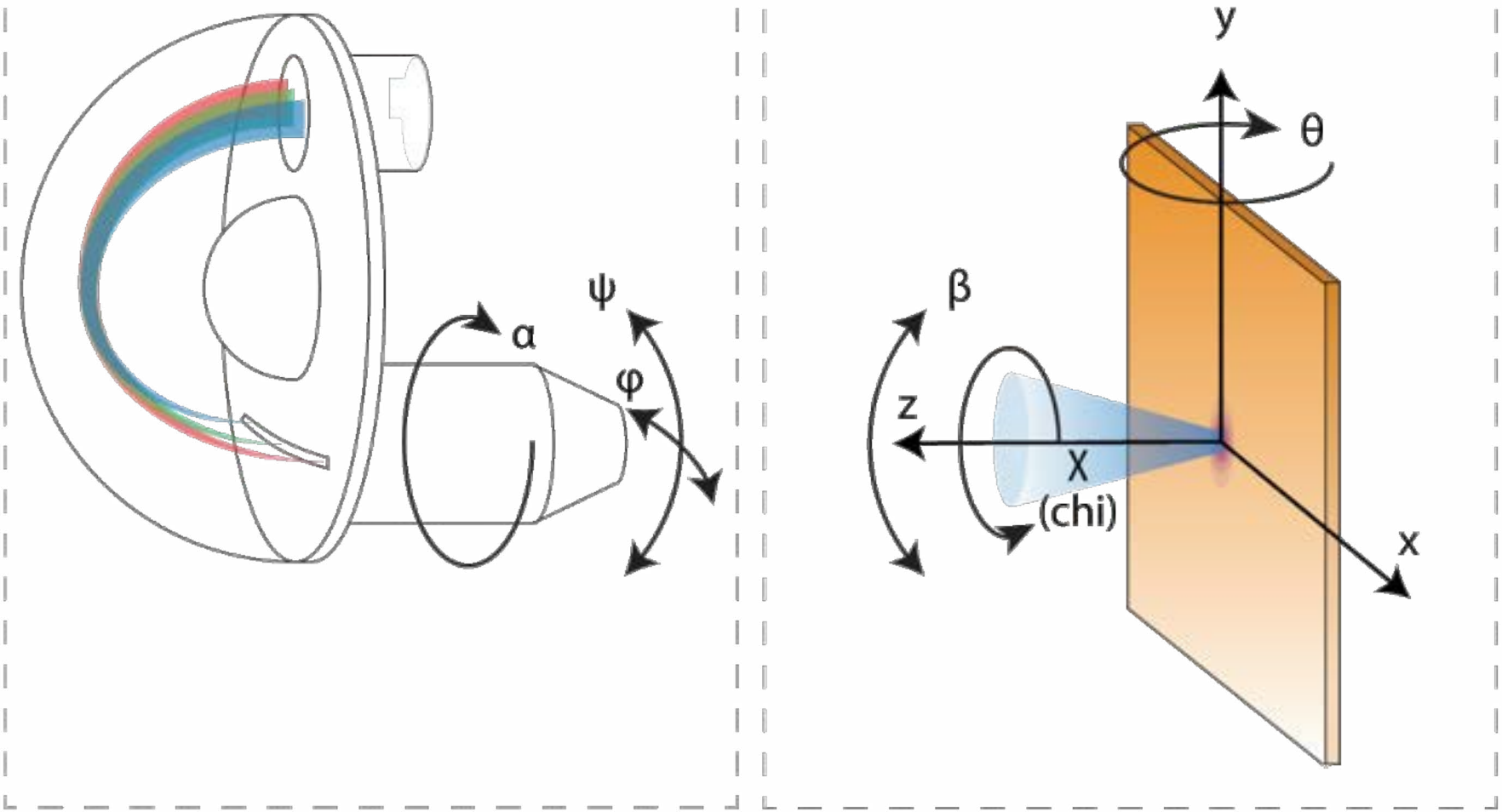


From the thesis of Brendan Faeth

Geometry of an ARPES experiment

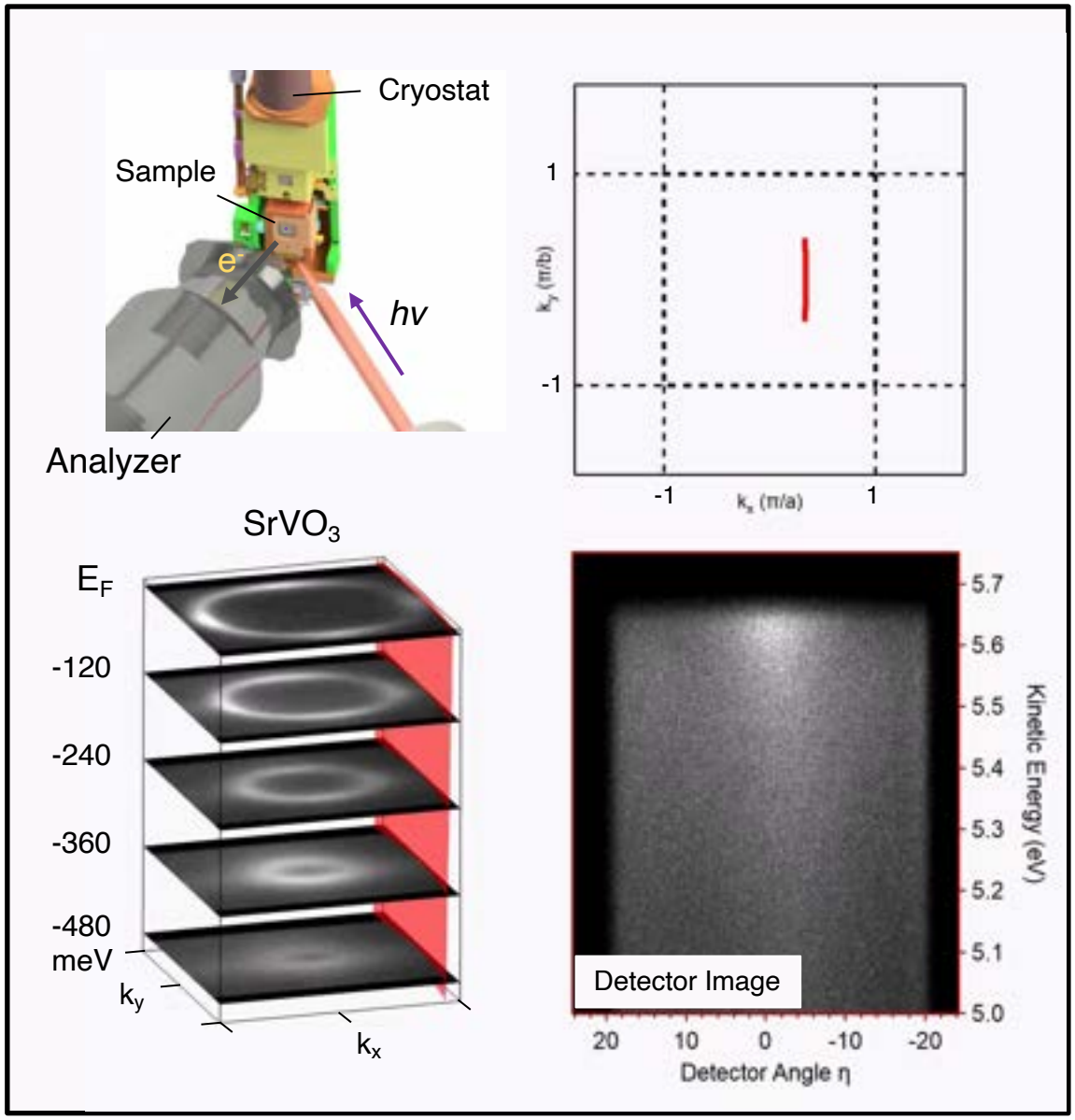
Analyzer Coordinates: (φ, ψ, α)

Manipulator Coordinates: (x, y, z, θ, β, χ)

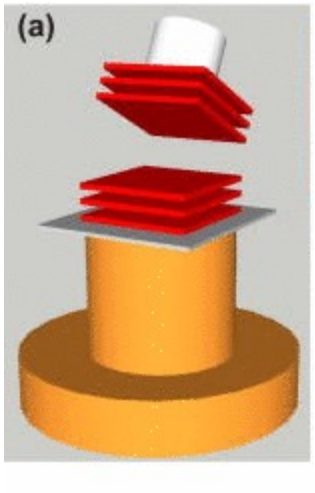


Hemispherical Analyzer Angular Conventions

Example of ARPES measurements in action



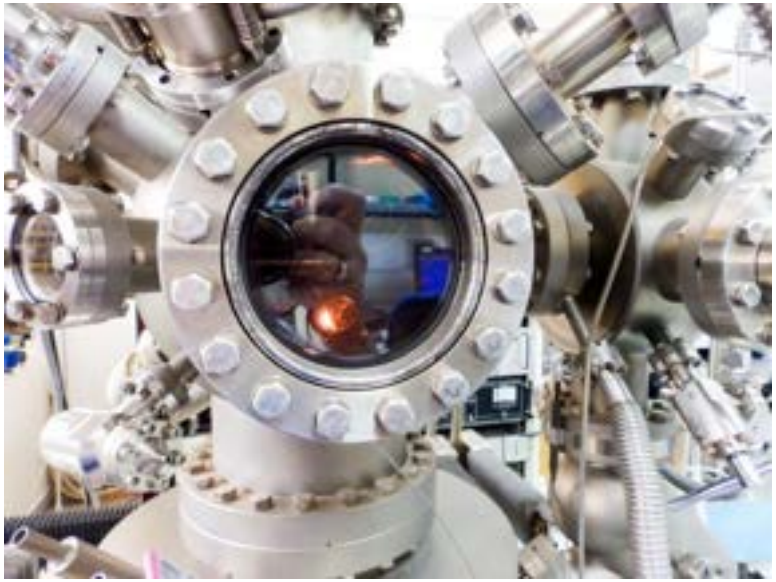
Tomorrow's demo : Cleaving a crystal



- Bi_2Se_3 crystal should be mounted in a ARPES cryostat prior to start of demo
- Show cleaving procedure with wobble stick, preferably with a nice camera so everyone can see.
- Crystal should be shiny and reflective after cleave, and (ideally) not have any differently oriented fractures

Concept Question #6

ARPES measurements need to take place in ultrahigh vacuum (10^{-10} torr or better). Which of the following is the **most important** factor which determines the level of vacuum needed to perform experiments?



A. The scattering / absorption of photoelectrons traveling inside the chamber

B. The operation of the electron analyzer

C. The absorption of vacuum ultraviolet (VUV) photons used for photoemitting the electrons

D. The scattering of electrons from adsorbed molecules at the sample's surface

E. All of the above are equally important

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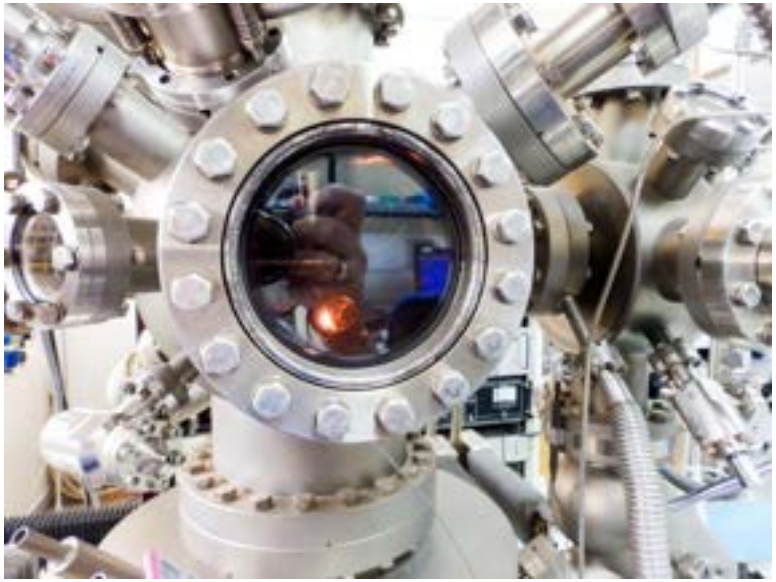

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Concept Question #7

PARADIM

Which of the following material systems could be readily studied by ARPES?

1. Organic materials / compounds
2. Ferromagnetic samples
3. Antiferromagnetic samples
4. Large band-gap insulators
5. Lightly doped semiconductors

A. 1, 3, 5 & 6

B. 3 & 5

C. 2, 4 & 5

D. 2, 3 & 5

E. All of the above

Concept Question #7

PARADIM

Which of the following material systems could be readily studied by ARPES?

1. Organic materials / compounds
2. Ferromagnetic samples
3. Antiferromagnetic samples
4. Large band-gap insulators
5. Lightly doped semiconductors

A. 1, 3, 5 & 6

B. 3 & 5

C. 2, 4 & 5

D. 2, 3 & 5

E. All of the above

Concept Question #8

Consider an ARPES experiment being conducting with an electron analyzer resolution of ΔE = 10 meV and an photon bandwidth of ΔE = 2 meV

What is the closest value of the **TOTAL** "effective" energy broadening in the experiment?

A. 10 meV

B. 11 meV

C. 12 meV

D. 13 meV

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