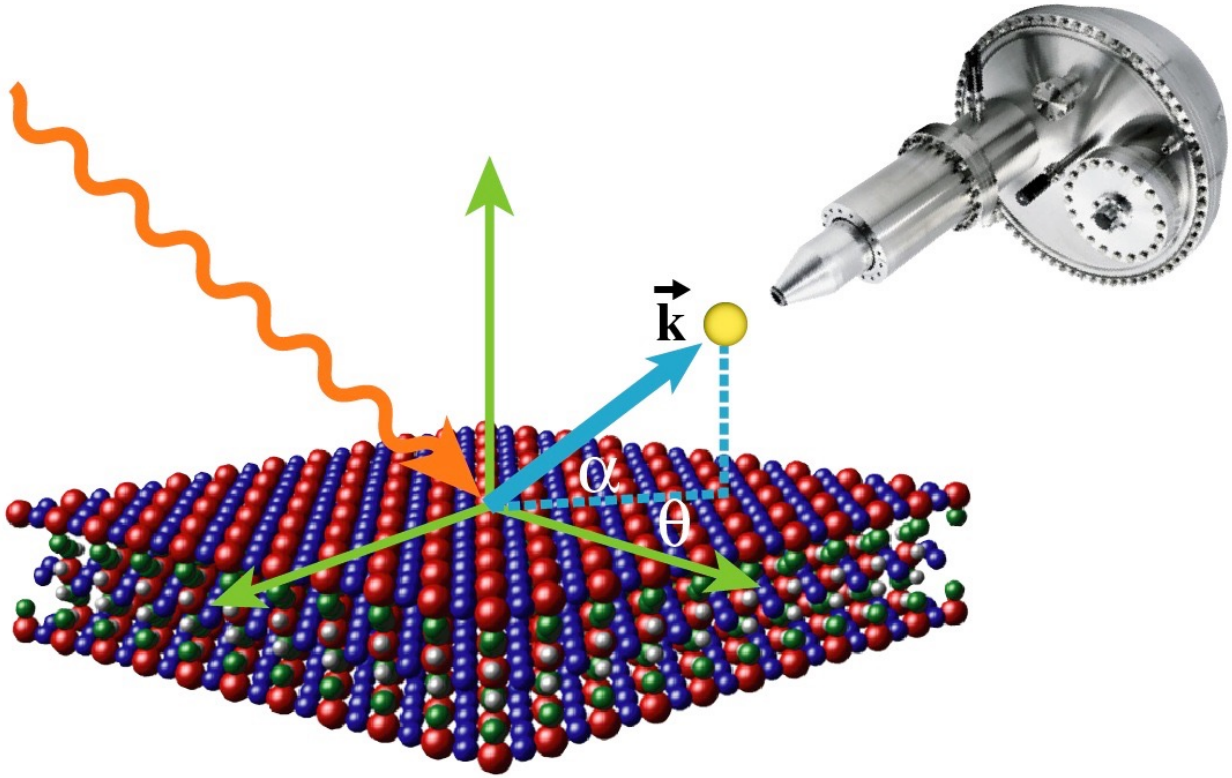
From Particles to Quasiparticles



Kyle Shen

PARADIM Summer School

June 16, 2022

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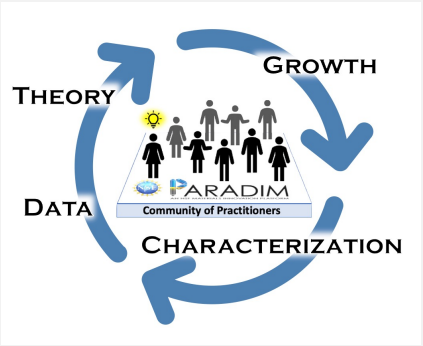
PLATFORM FOR THE ACCELERATED REALIZATION, ANALYSIS & DISCOVERY OF INTERFACE MATERIALS



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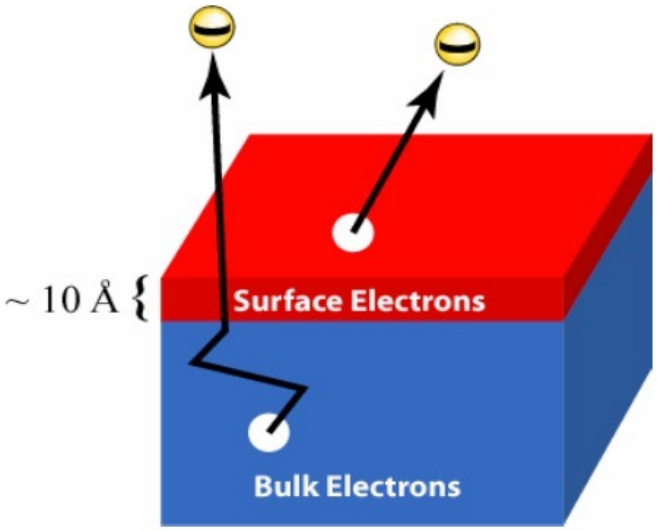
a few things to keep in mind about ARPES

• mean free path (mfp) of photoelectrons in solids is ~ **1 nanometer**. Measurements are sensitive to top unit cells & monolayers

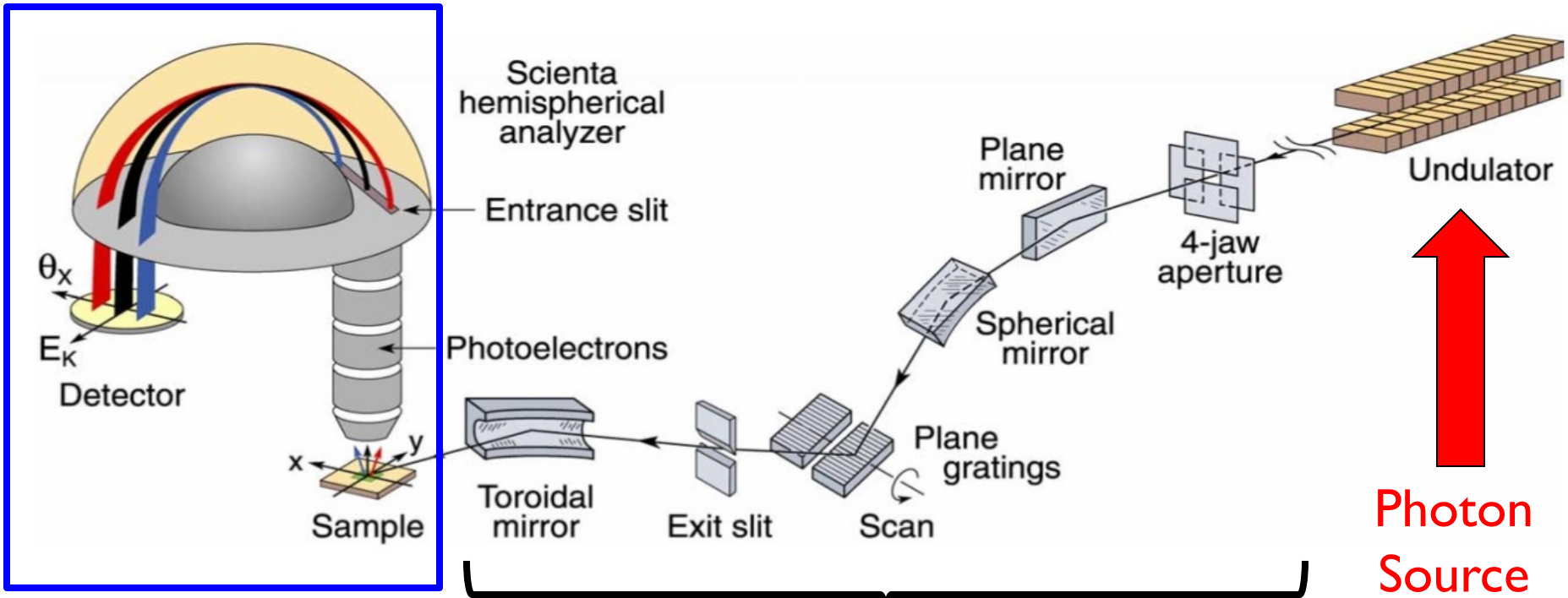
• short mfp means sample surfaces must be kept pristine (no adsorbed gases) in ultrahigh vacuum (UHV - 10^{-10} torr)

• need to reduce stray electromagnetic fields (deflection)

• infeasible to measure samples *in vivo* or in solution



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 ~ 3000 : 1

synchrotron facilities

approximately 50 synchrotrons worldwide

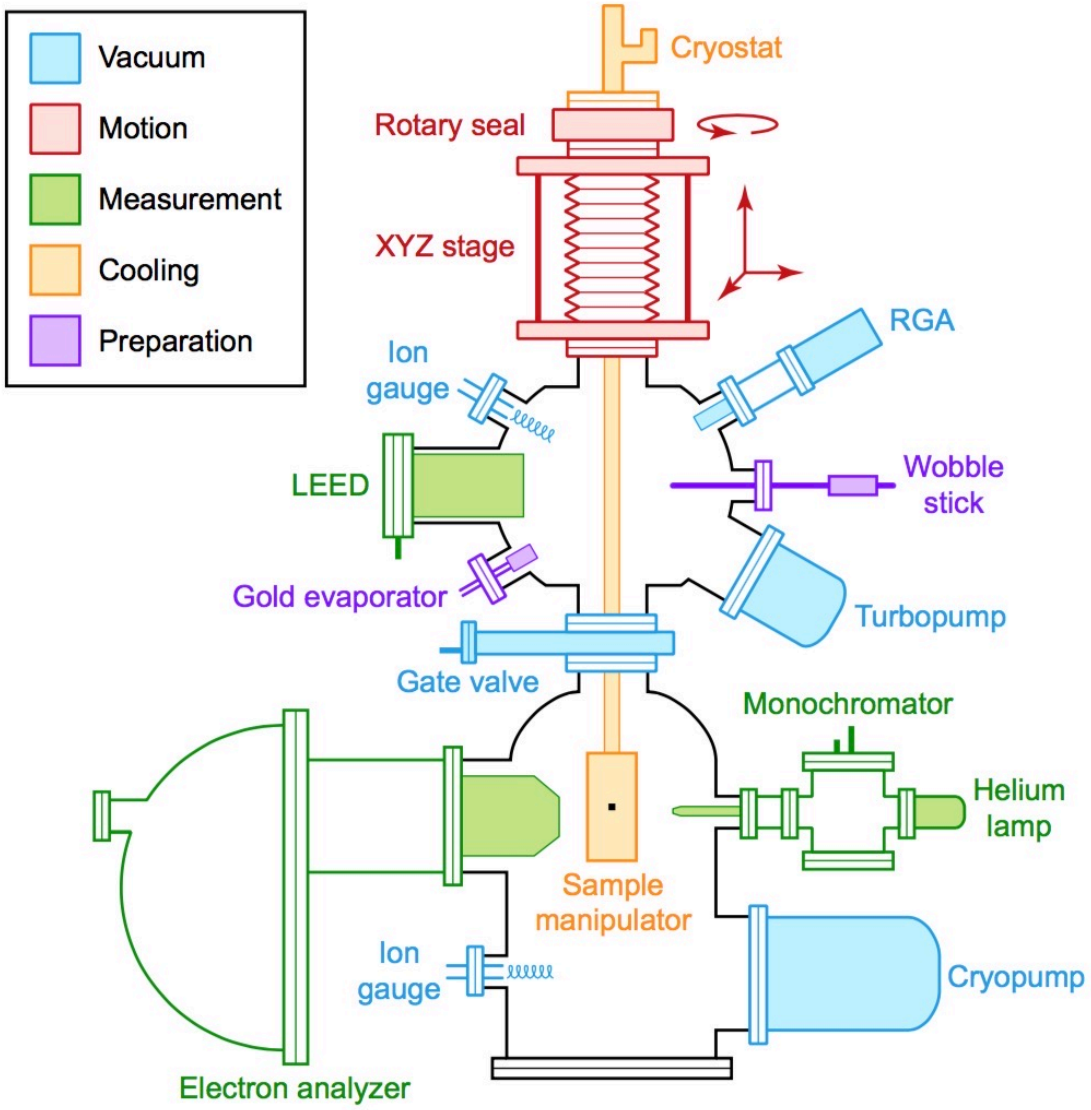
Beamtime at synchrotrons is free of charge – granted based on merit of proposal

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

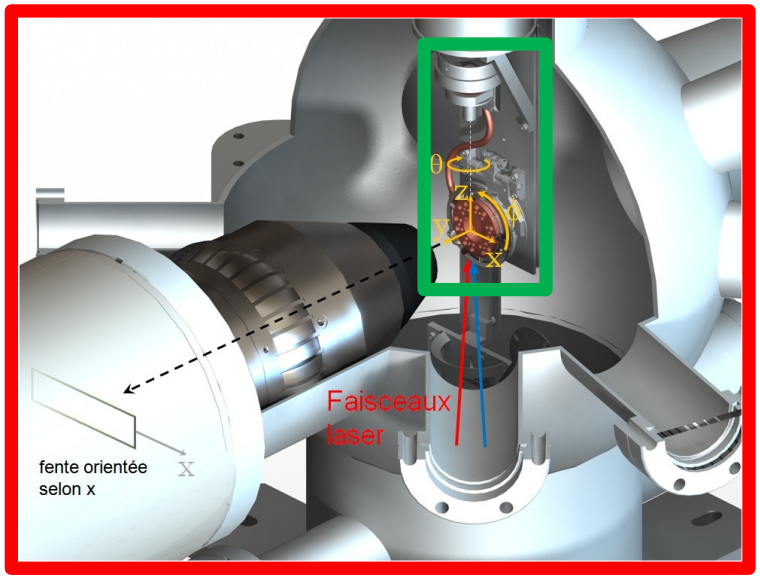
graphic from Argonne National Laboratory

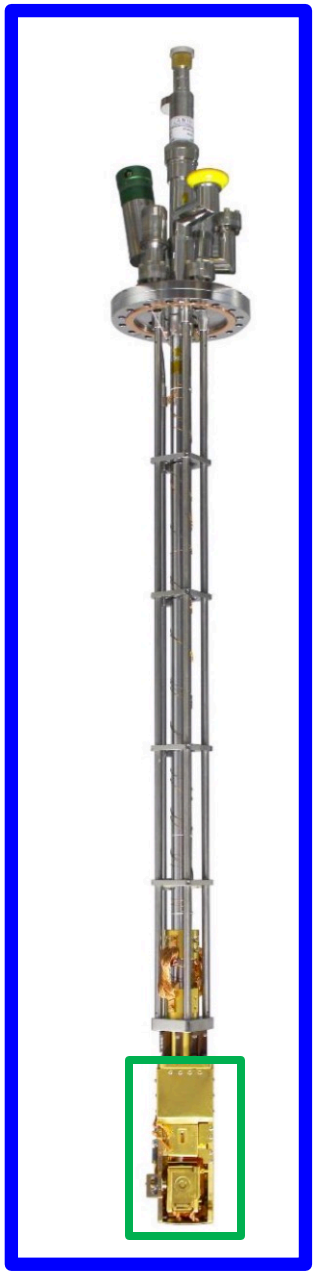
ARPES chamber : Block Diagram

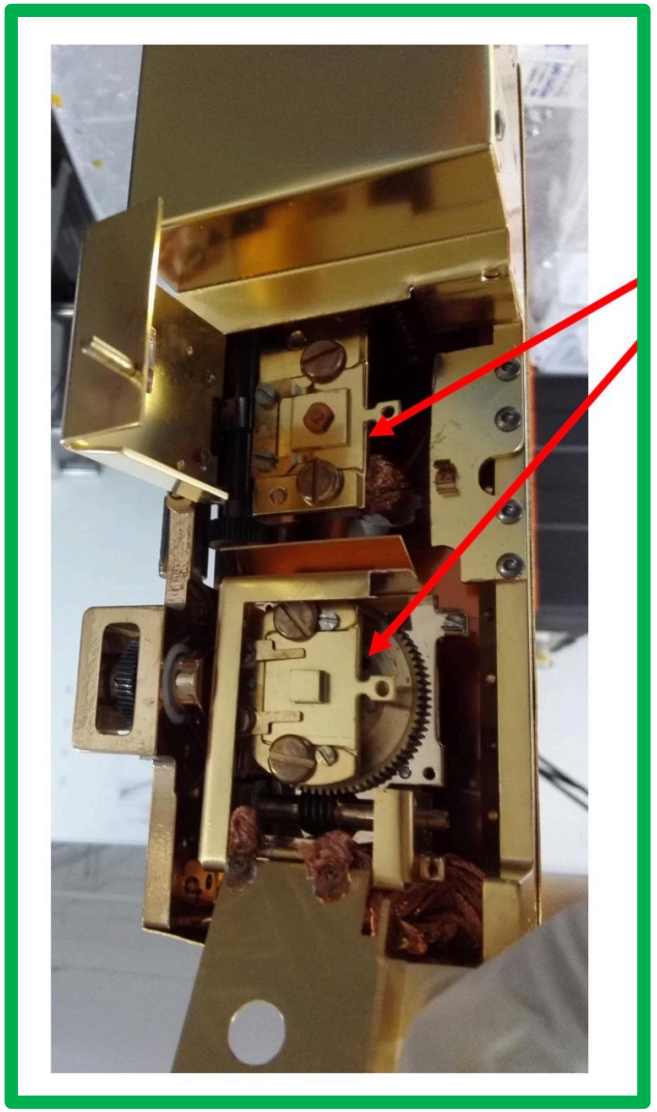


From the thesis of John Harter

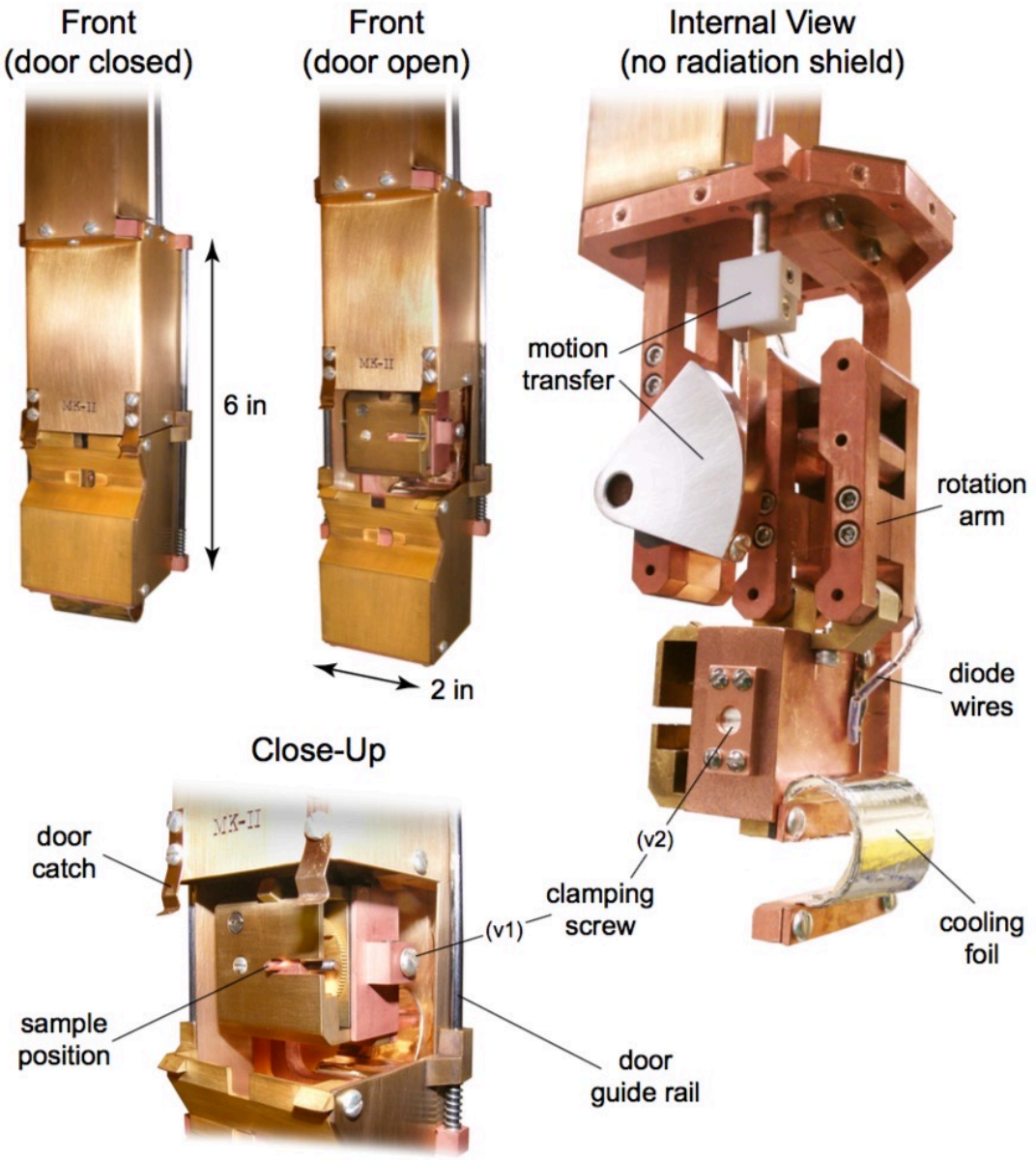
A typical ARPES chamber







ARPES sample manipulator



From the thesis of John Harter

advantages & disadvantages of ARPES

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Advantages

- surface sensitive (nanometer probe depth)
- high elemental sensitivity / specificity
- sensitive to oxidation state & chemical bonding
- uniquely sensitive probe of electronic structure (can determine electron energy & momentum)
- photoemission "cross-section" is usually quite high (strong signals)

Disadvantages

- bulk insensitive (nanometer probe depth)
- ultrahigh vacuum necessary (can limit types of samples studied)
- cannot study materials under high pressures, under magnetic fields, in solution / in *vivo*
- limited biological applications
- limited spatial resolution (some exceptions)
- limited temporal resolution (some exceptions)

typical energy scales in photoemission

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$1\ \text{eV} = 1.24\ \mu\text{m}$ $0.001\ \text{eV} = 12\ \text{K}$

- Ionization energy of a 1s electron in copper : **9000 eV**
- Ionization energy of hydrogen atom : **13.6 eV**
- Ferromagnetic exchange splitting in iron : **1.6 eV**
- Lattice vibrations in Si (optical phonon) : **0.06 eV**
- Room temperature (300 K) : **0.025 eV**
- Superconducting gap in niobium (9.2 K) : **0.0013 eV**
- Boiling point of liquid He (4.2 K) : **0.0004 eV**

Concept Question #2

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

Concept Question #3

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.

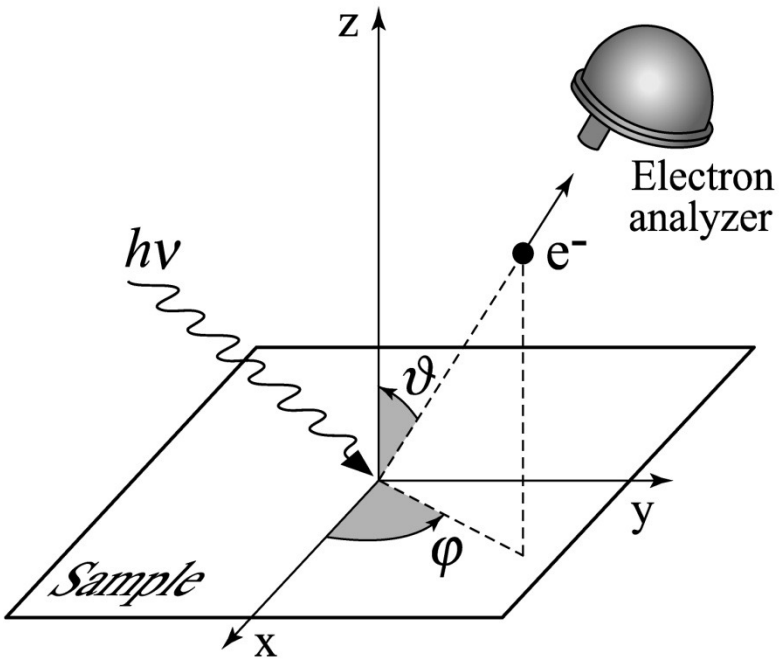
Is this a complete waste of time (and money)?

A. Yes

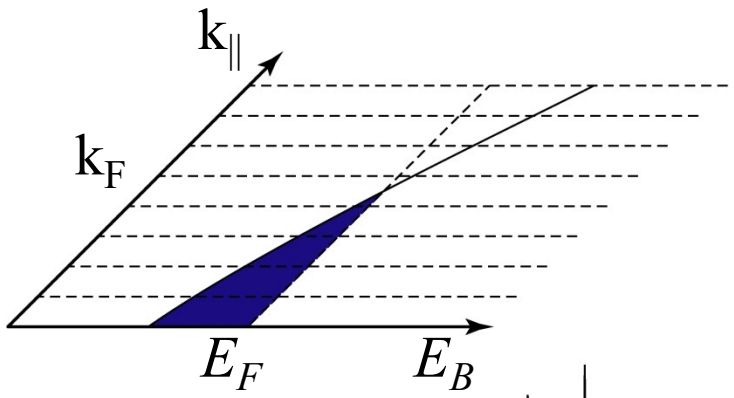
B. No

C. All of the above

Kinematics of the ARPES process : Momentum Conservation



Electrons in Reciprocal Space

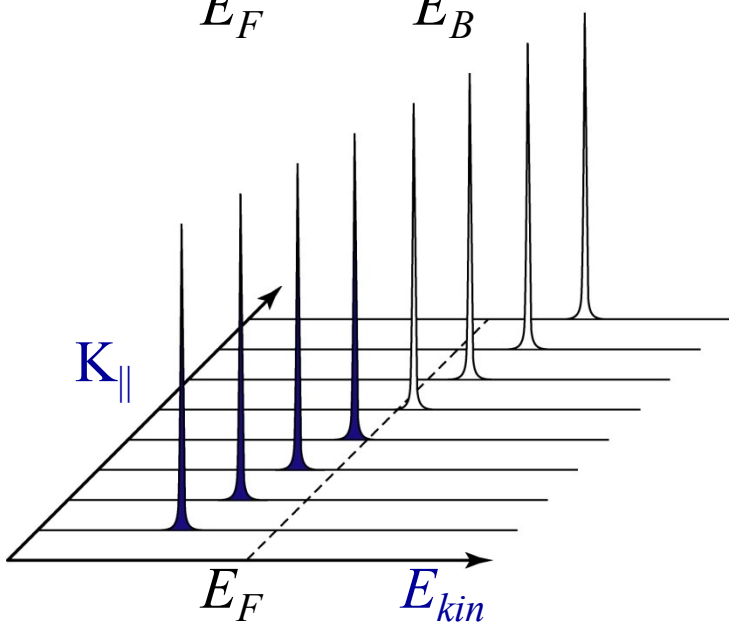


Energy Conservation

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

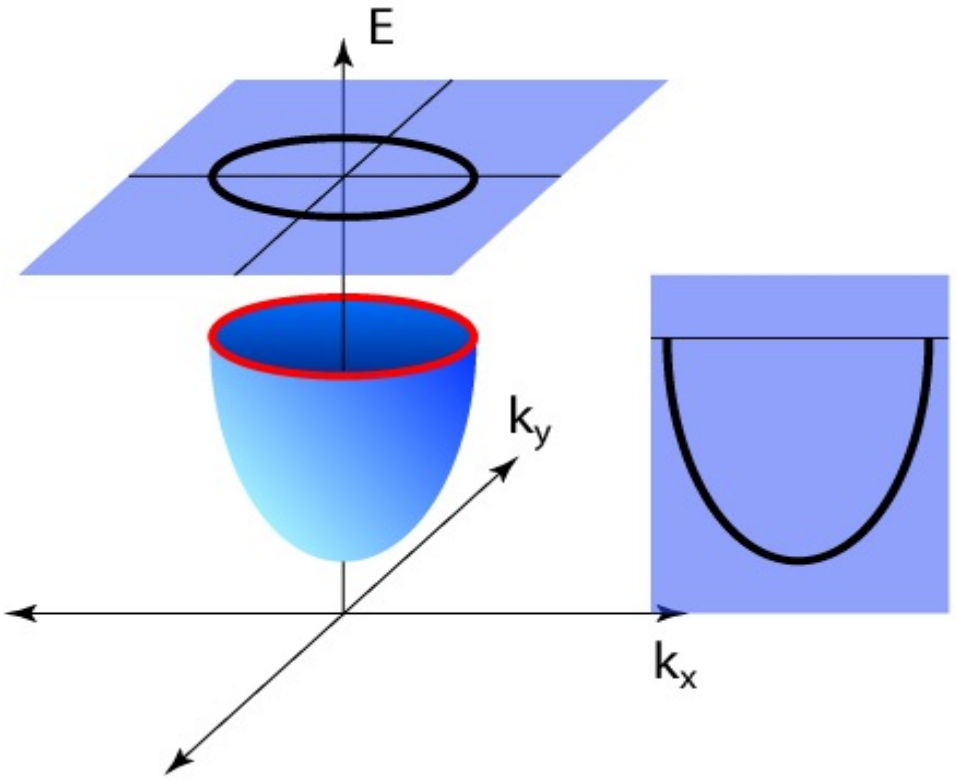
Momentum Conservation

$$\hbar k_{||} = \hbar K_{||} = \sqrt{2mE_{kin}} \cdot \sin\vartheta$$



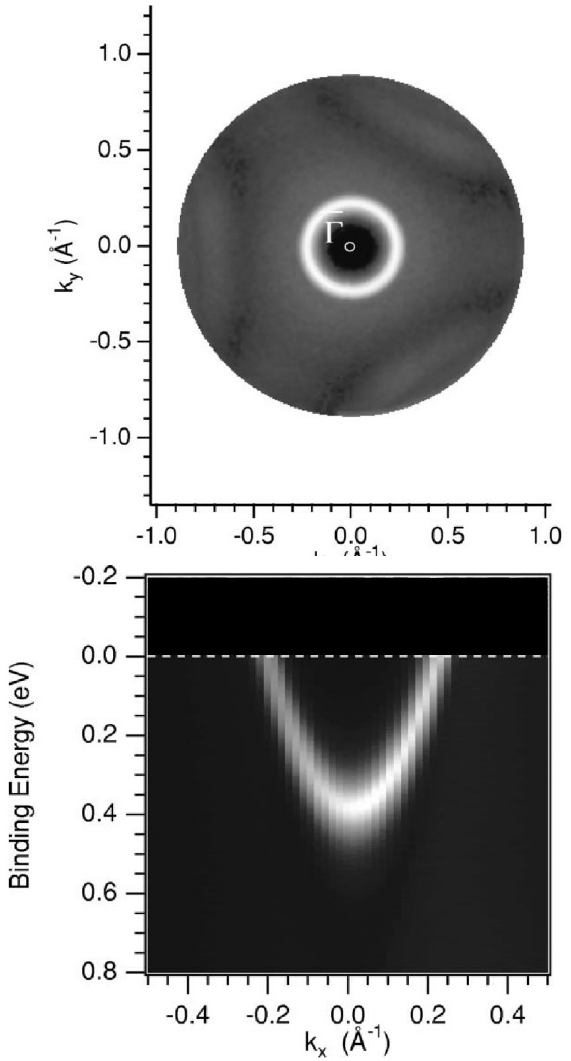
Courtesy of Andrea Damascelli

a simple example of ARPES : 2D free electron gas on Cu (111)



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

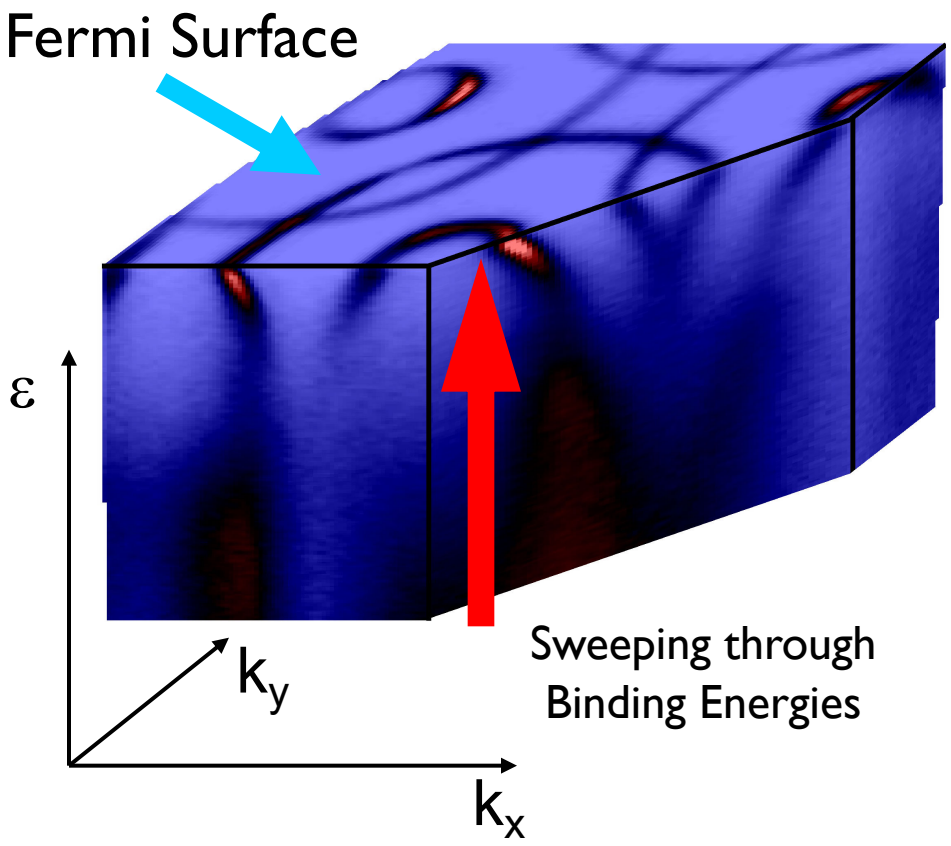
Cu (111)

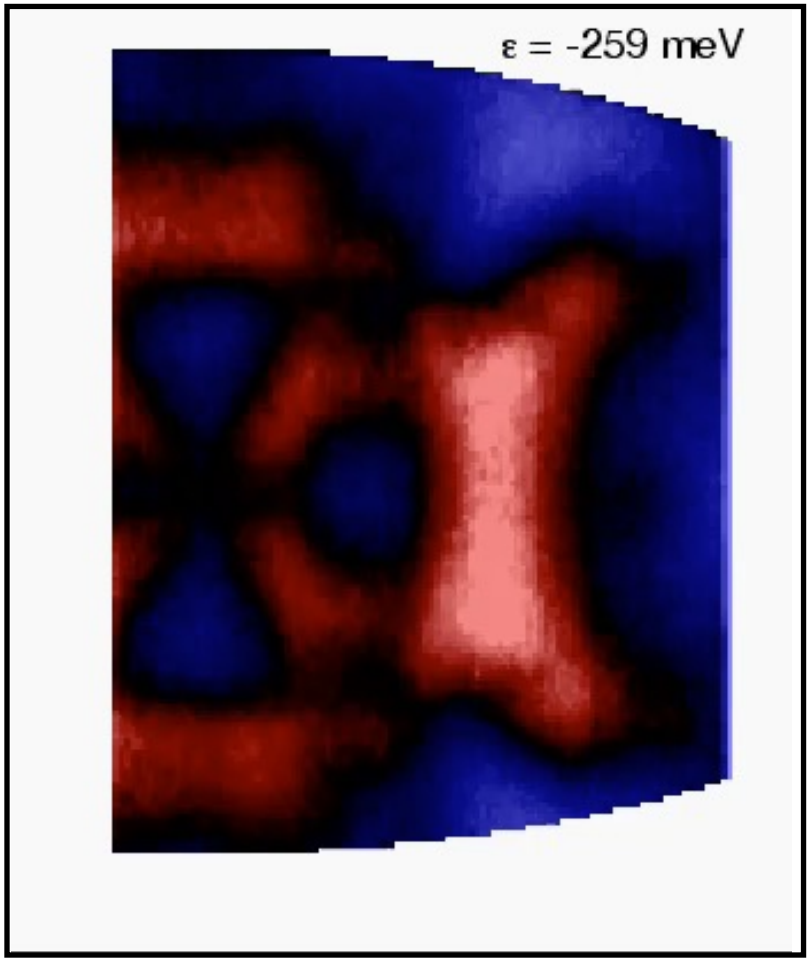


F. Baumberger et al., PRB 64, 195411 (2001)

PARADIM

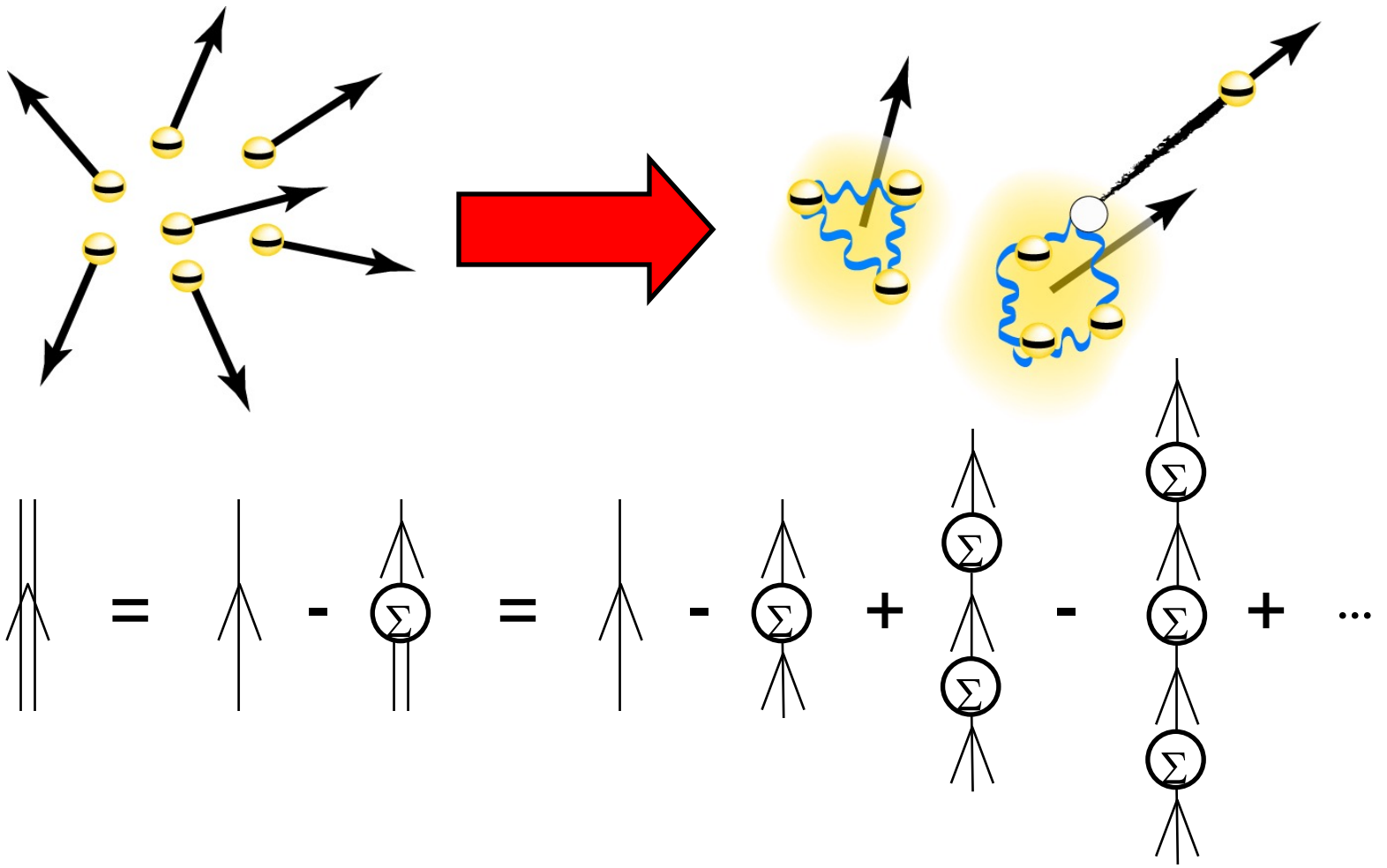
Sr_2RhO_4 : A *4d* transition metal oxide with strong spin-orbit coupling





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

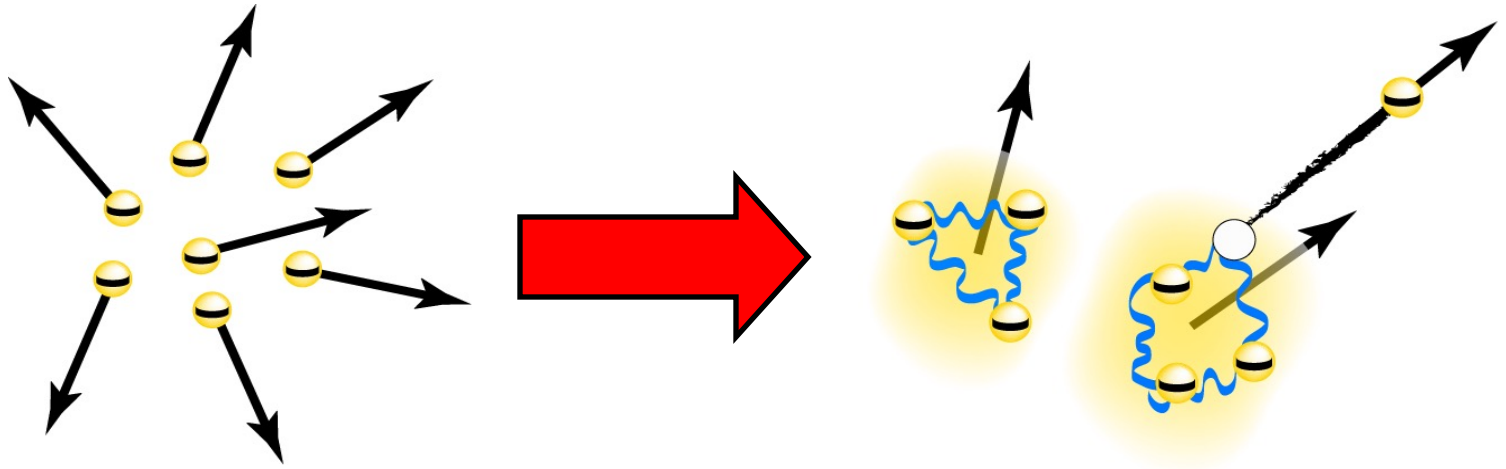
the quasiparticle concept : Landau Fermi liquid theory

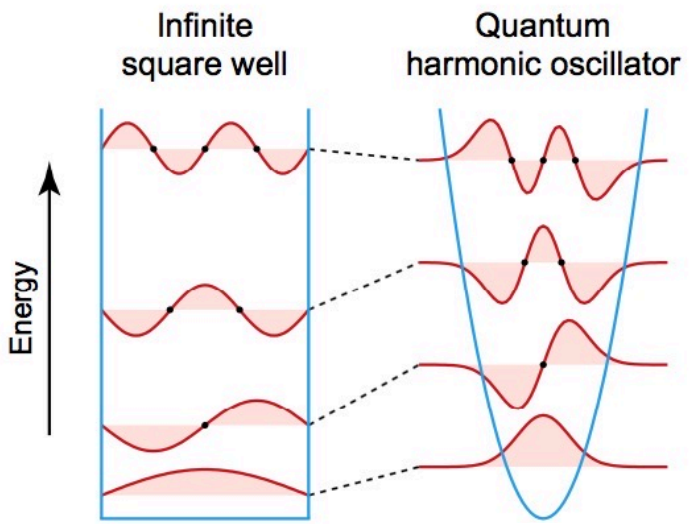


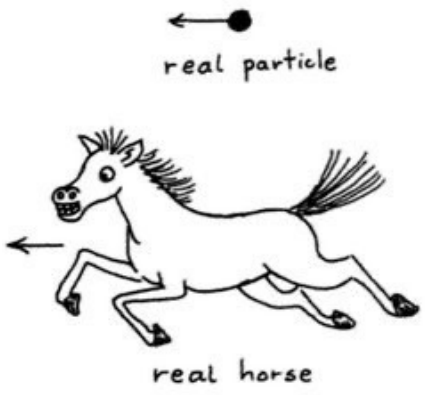
Single-particle spectral function

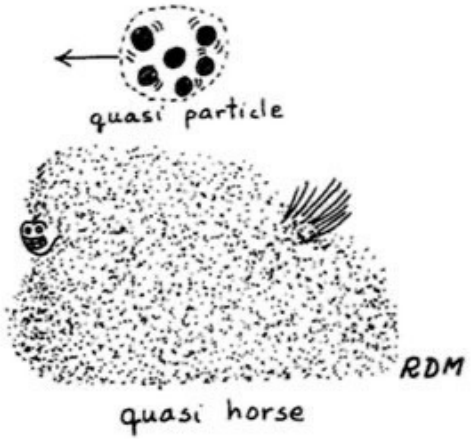
$$A(\mathbf{k},\omega) \propto \frac{\mathrm{Im}\Sigma(\mathbf{k},\omega)}{[\omega - \varepsilon_k - \mathrm{Re}\Sigma(\mathbf{k},\omega)]^2 + [\mathrm{Im}\Sigma(\mathbf{k},\omega)]^2}$$

the quasiparticle concept : Landau Fermi liquid theory









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

Single-particle spectral function

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

Spectral Function Analysis of ARPES data

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

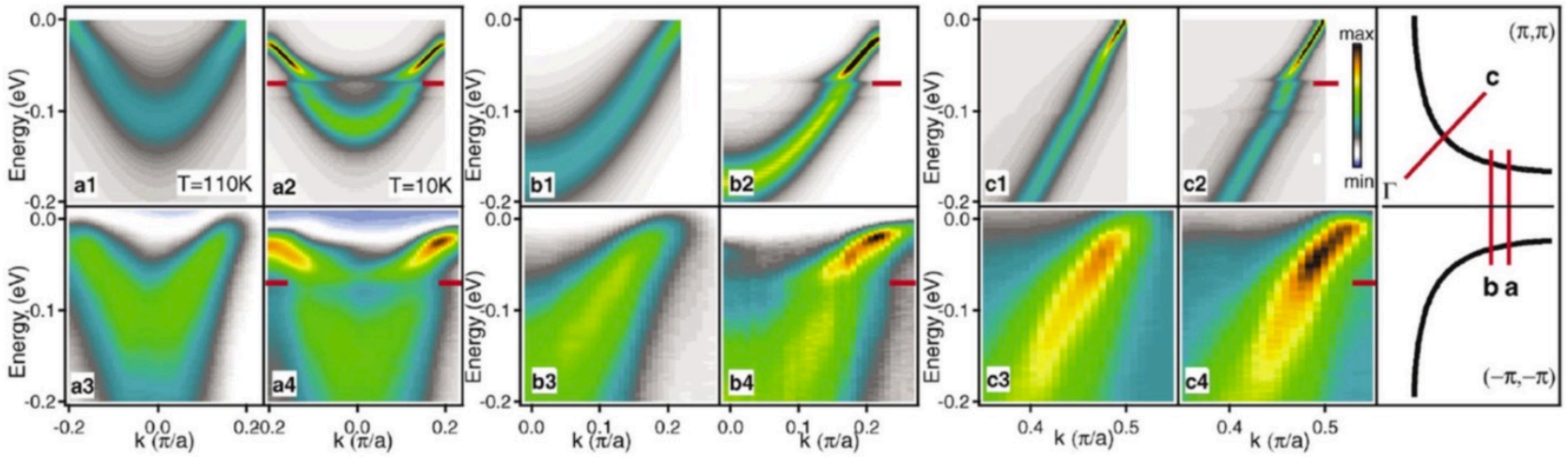


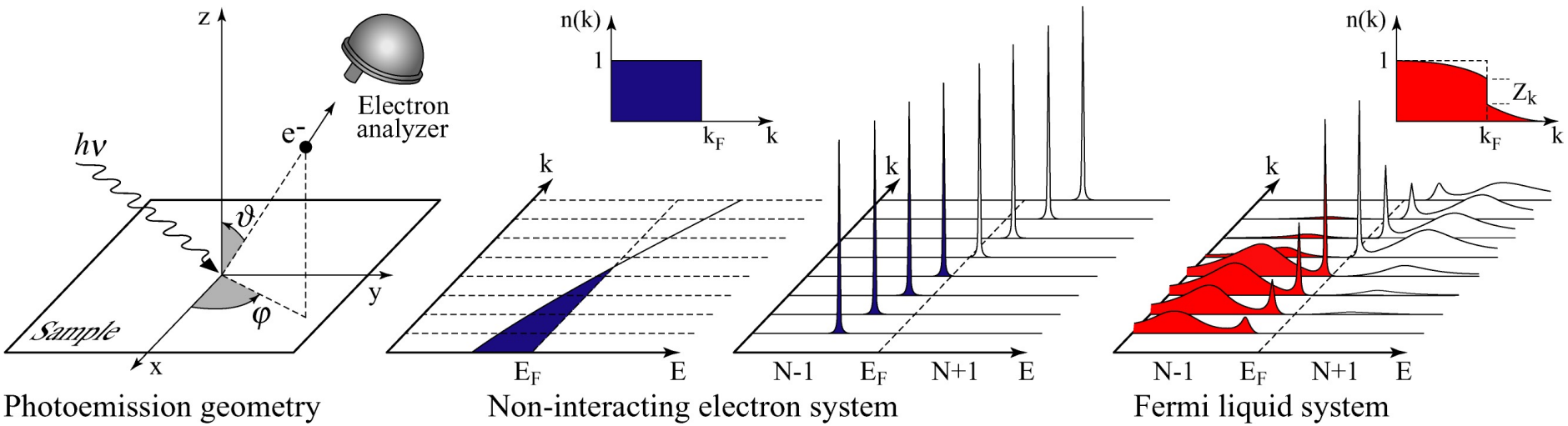
FIG. 3 (color). Image plots of the calculated spectral functions in the normal (a1,b1,c1) and superconducting (a2,b2,c2) states compared to the spectral functions in the normal (a3,b3,c3) and superconducting (a4,b4,c4) states measured in $Bi_2Sr_2Ca_{0.92}Y_{0.08}Cu_2O_{8+\delta}$ (Bi-2212) [6] for momentum 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

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

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 liquid theory as a cornerstone

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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

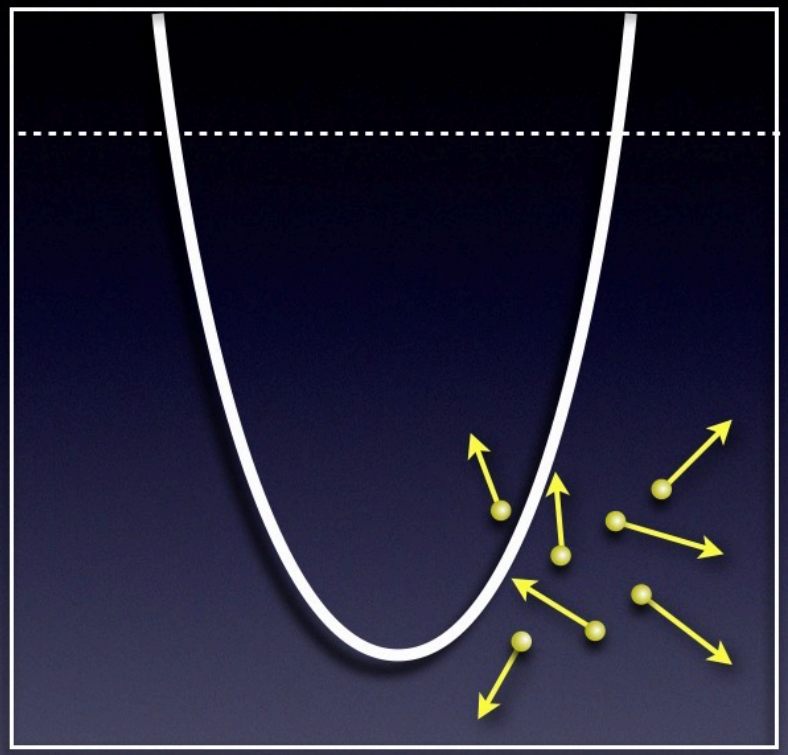
Sharp coherent quasiparticles ($\mathrm{Im}\Sigma\,(\omega) < \varepsilon_k$)

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

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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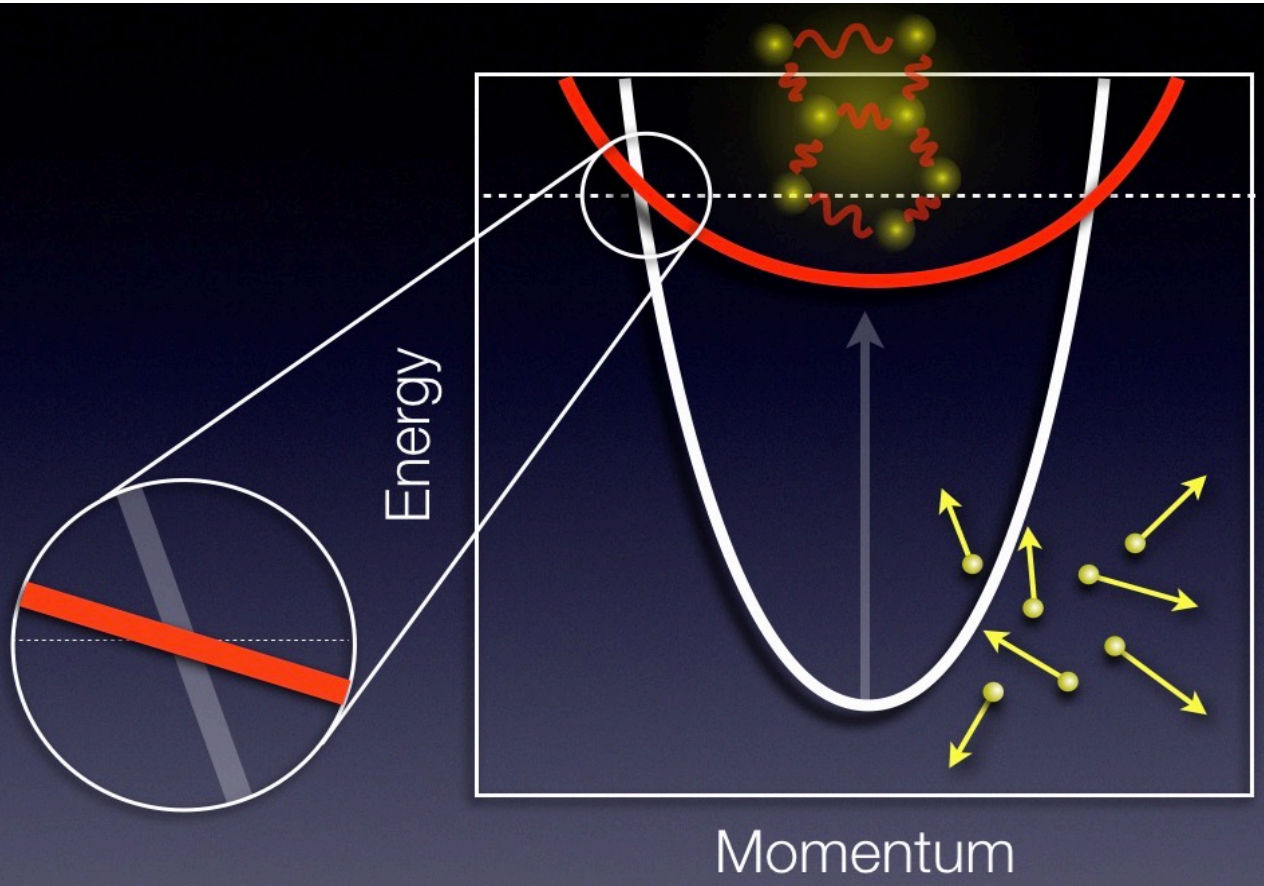
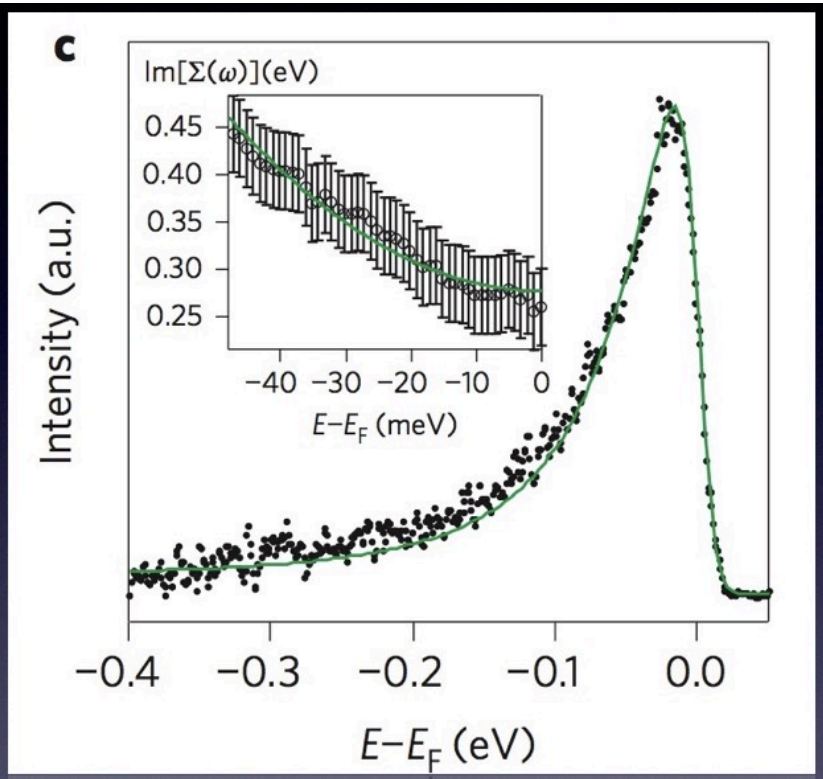


Typically estimate mass renormalization by comparing the experimental Fermi velocity (slope) with the predicted velocity *without interactions**

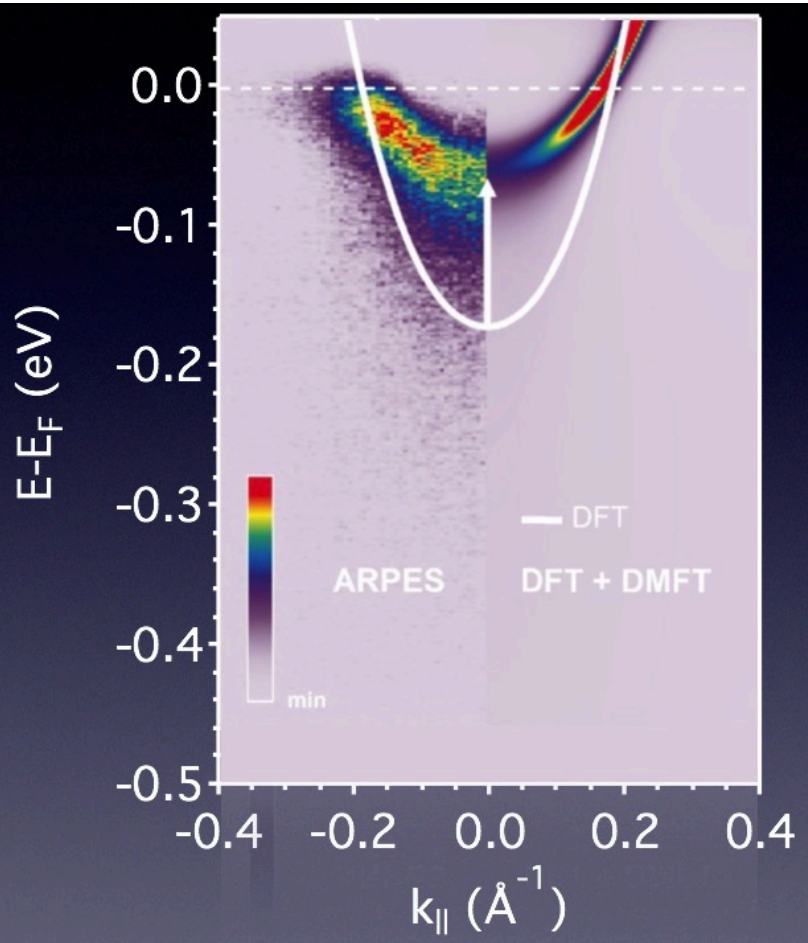
*usually DFT calculations

Electron-electron interactions in $LaNiO_3$



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

in agreement with optical & thermodynamic measurements



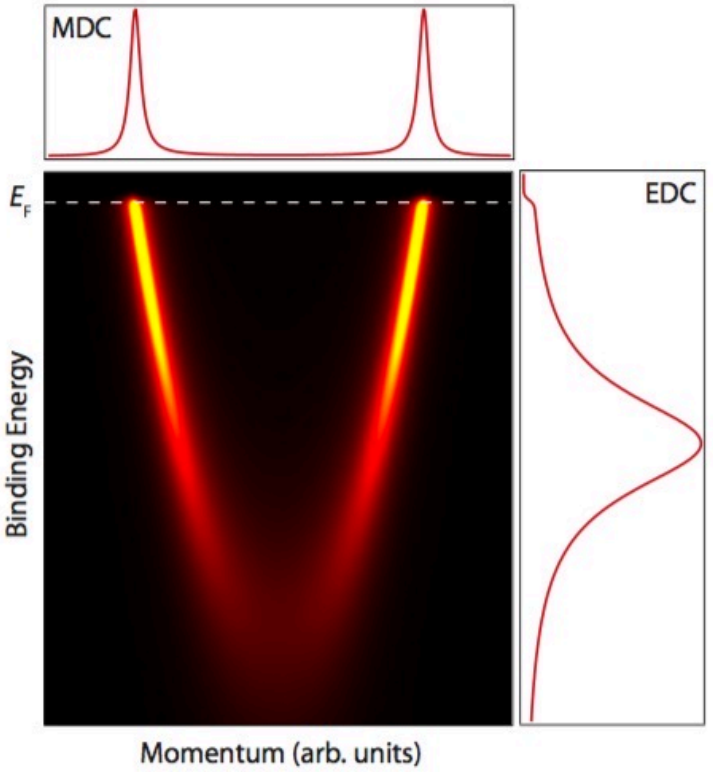
P.D.C. King *et al*, *Nature Nanotechnology* **9**, 443 (2014)

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

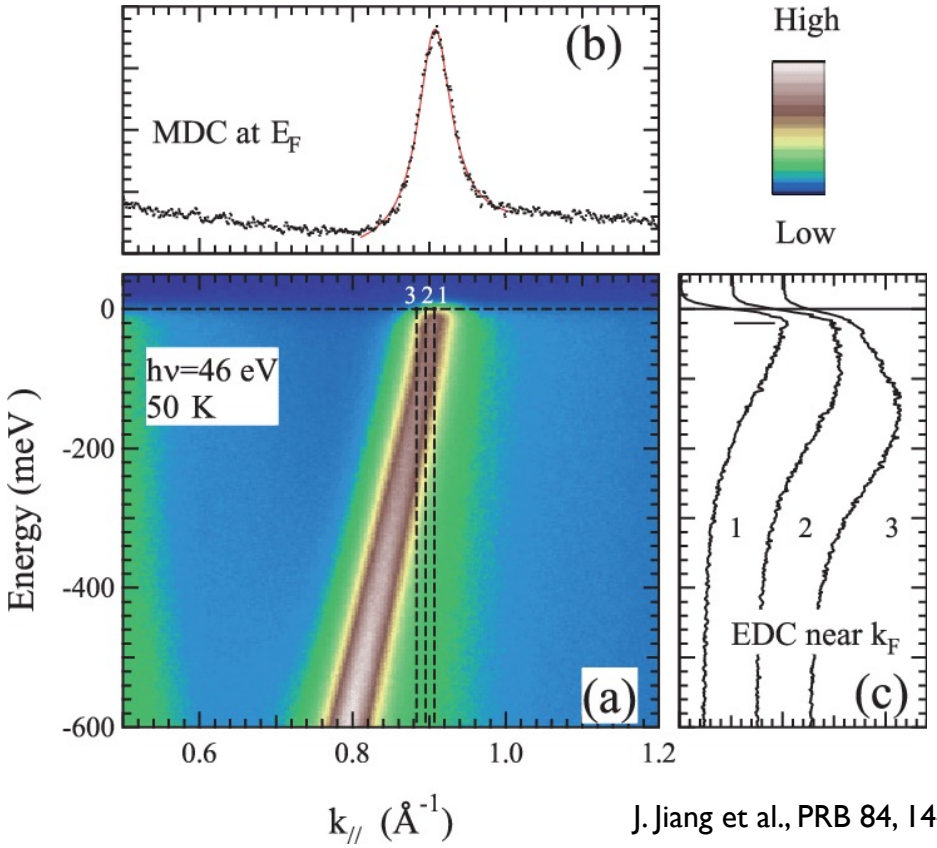
P.D.C. King *et al*, *Nature Nanotechnology* **9**, 443 (2014)

Ways to analyze ARPES data : EDC & MDC analysis

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



From D. Shai's thesis

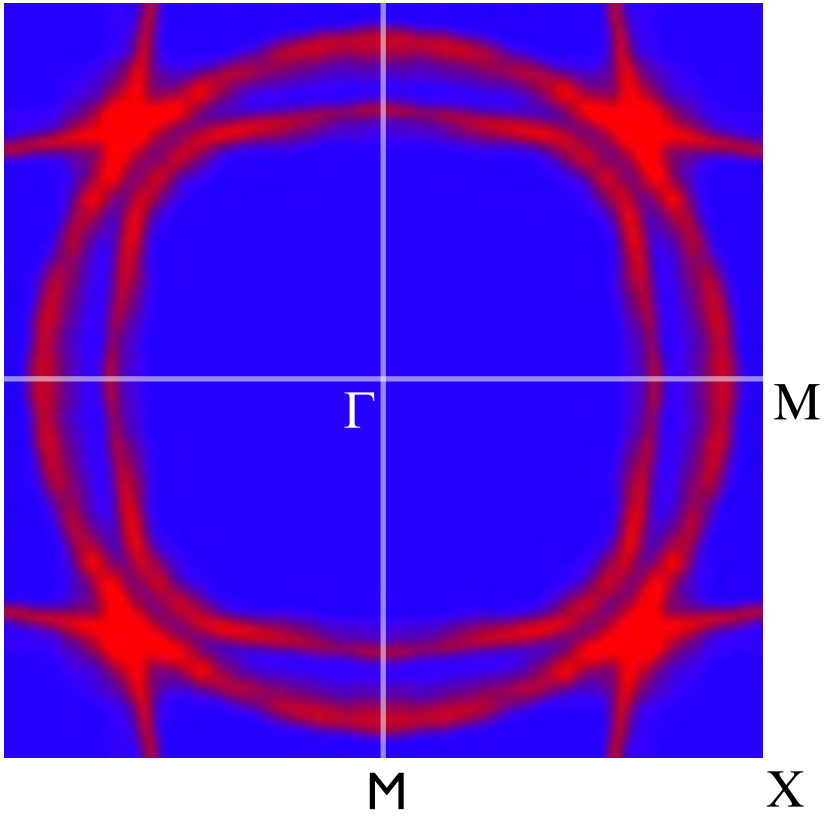


J. Jiang et al., PRB 84, 144124

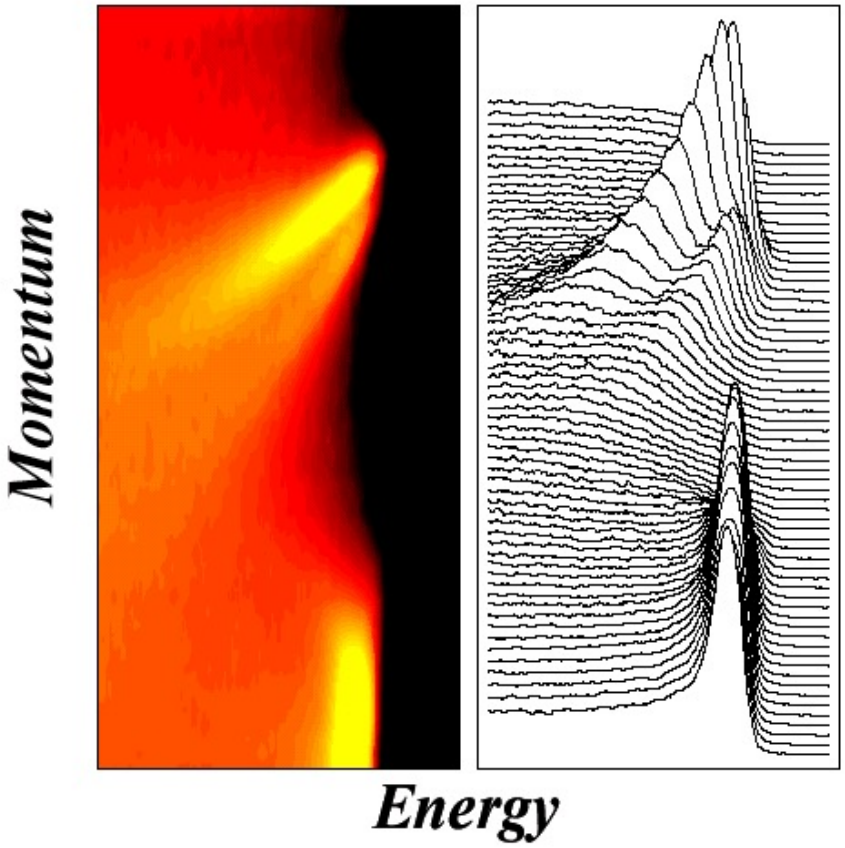
MDCs : Good for dispersion tracking; steeply dispersing bands
EDCs : Good for lineshape analysis; analyzing flat bands

quasiparticle excitations in Sr_2RuO_4

Fermi Surface from ARPES



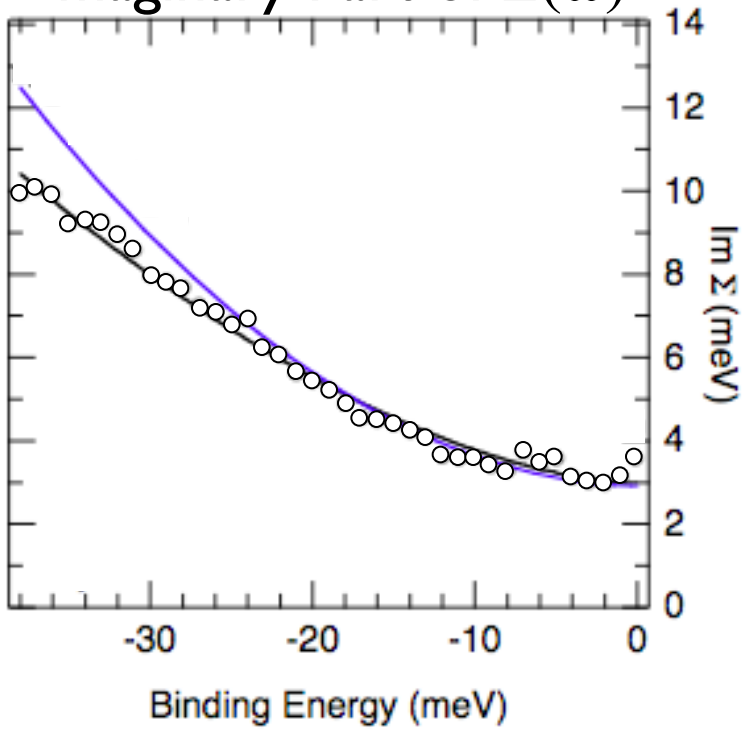
Quasiparticles in Sr_2RuO_4



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)

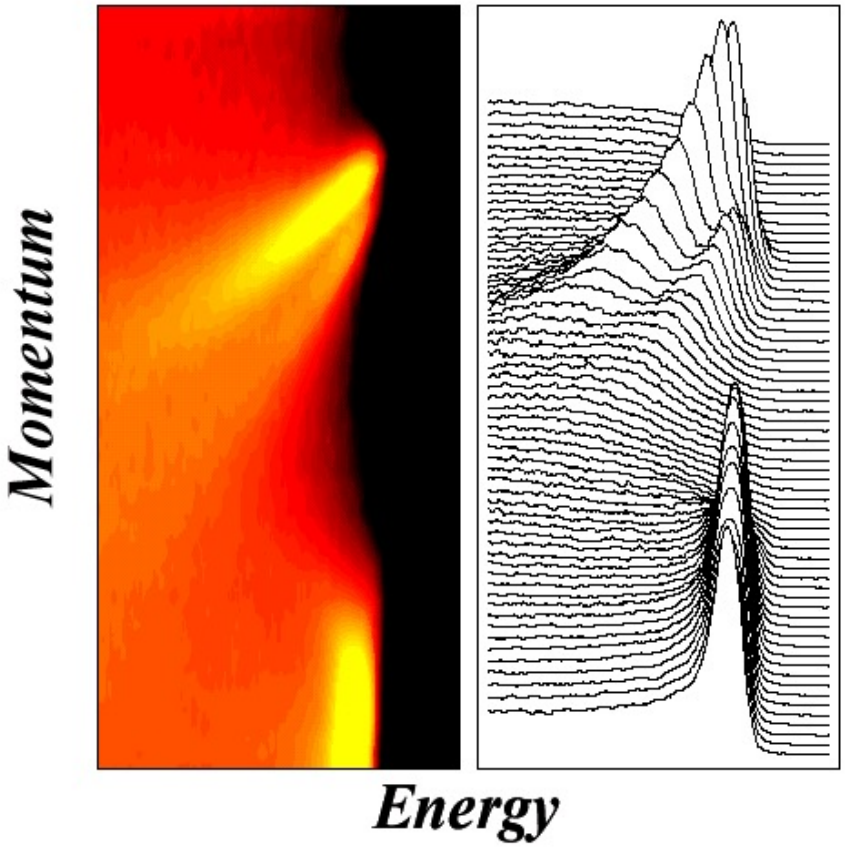
Electron-electron interactions in Sr_2RuO_4

Imaginary Part of $\Sigma(\omega)$



$$\mathrm{Im}\Sigma(\omega) = \beta\omega^2\left[1 + 0.53\,\ln\frac{\omega}{E_F}\right] + c$$

Quasiparticles in Sr_2RuO_4



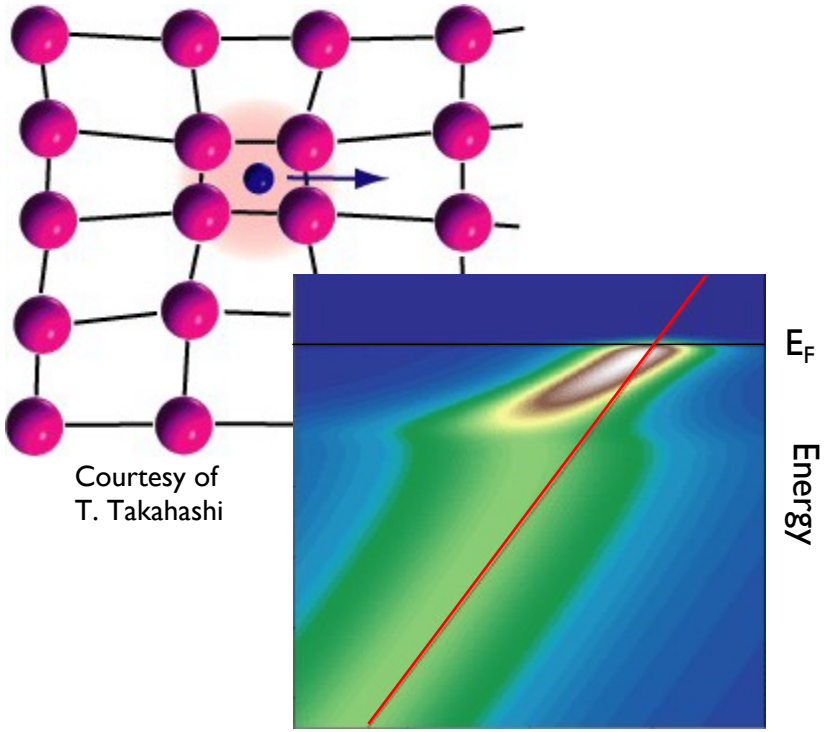
- $\mathrm{Im}\Sigma < |E - E_F|$:Well-defined quasiparticles
- Electron-electron scattering dominates over electron-phonon interactions

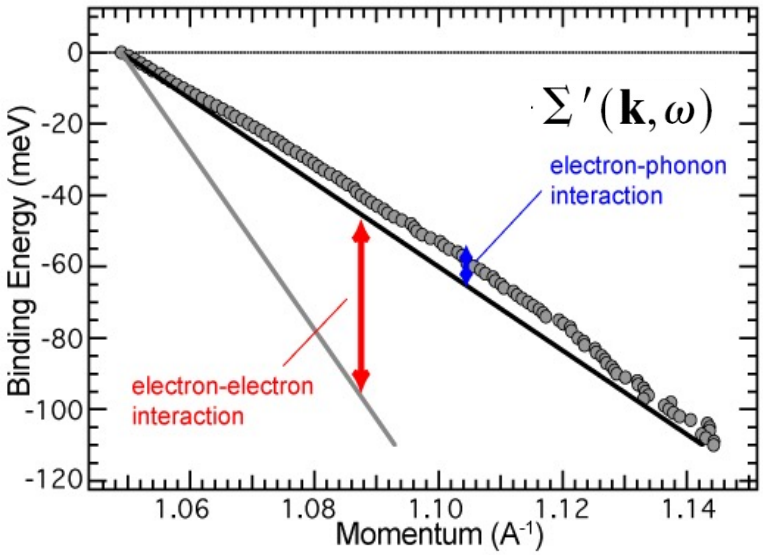
N.J.C. Ingle, K.M. Shen, *et al.*, Phys. Rev B (2006)

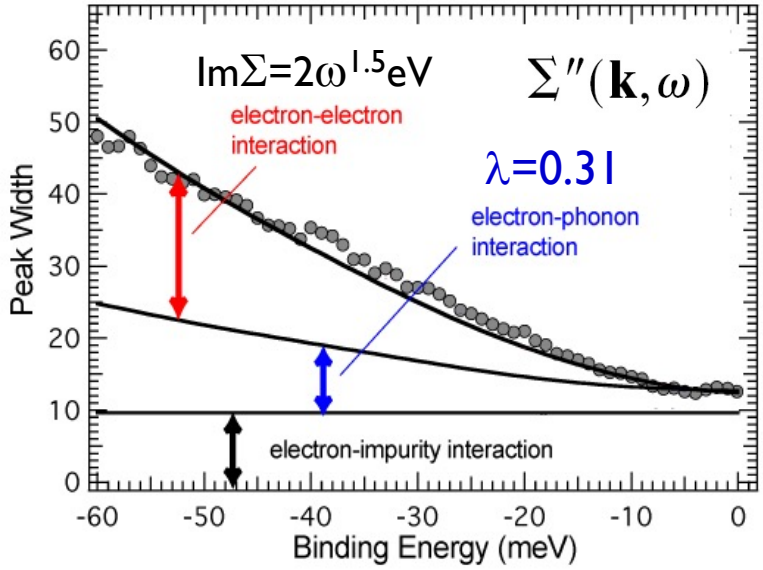
Many body correlation effects in Sr_2RuO_4

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, *et al.*, Phys. Rev B (2006)

Concept Question #4

PARADIM

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

Concept Question #4

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C. 2, 3, 4, and 5

D. 4 and 5

E. All of the above

Electron-Lattice Interactions by ARPES

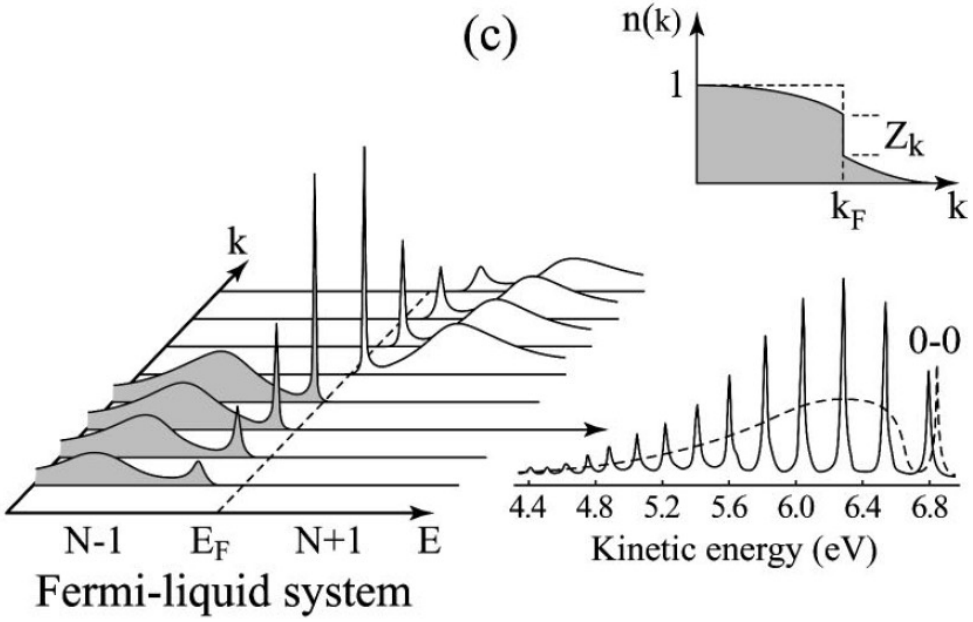
NEWS AND VIEWS

HIGH-TEMPERATURE SUPERCONDUCTIVITY

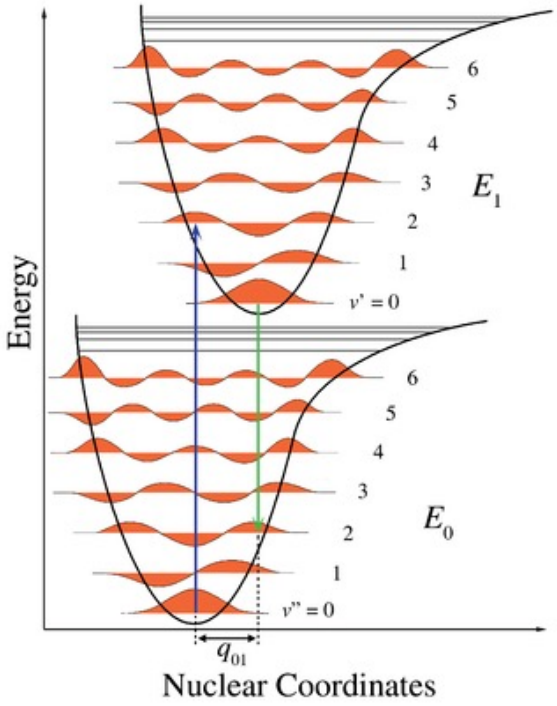
Nature 342, 480 (1989)

Testing Fermi-liquid models

G.A. Sawatzky

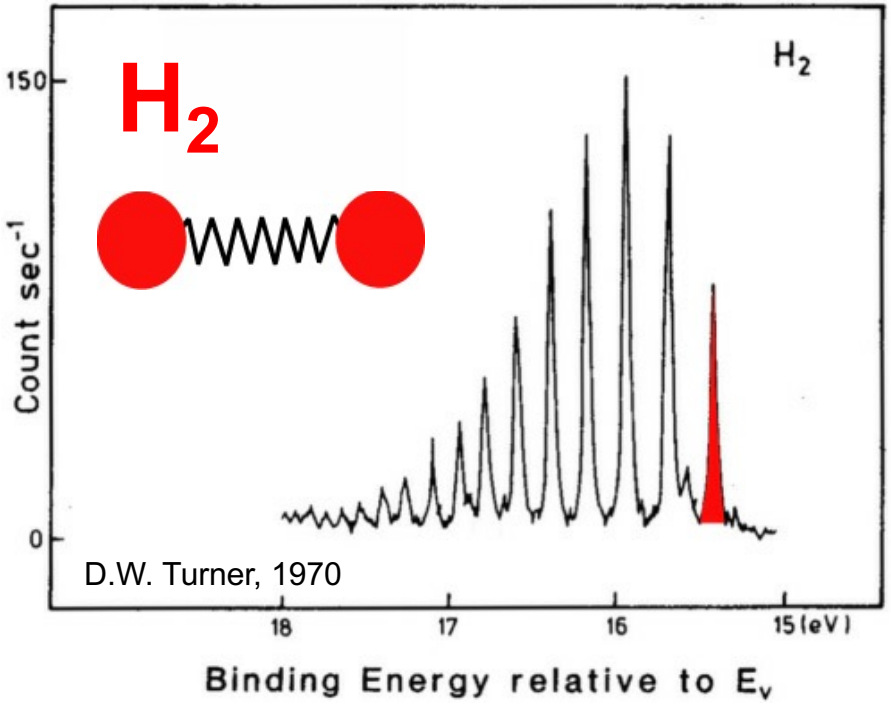


Franck-Condon



"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."

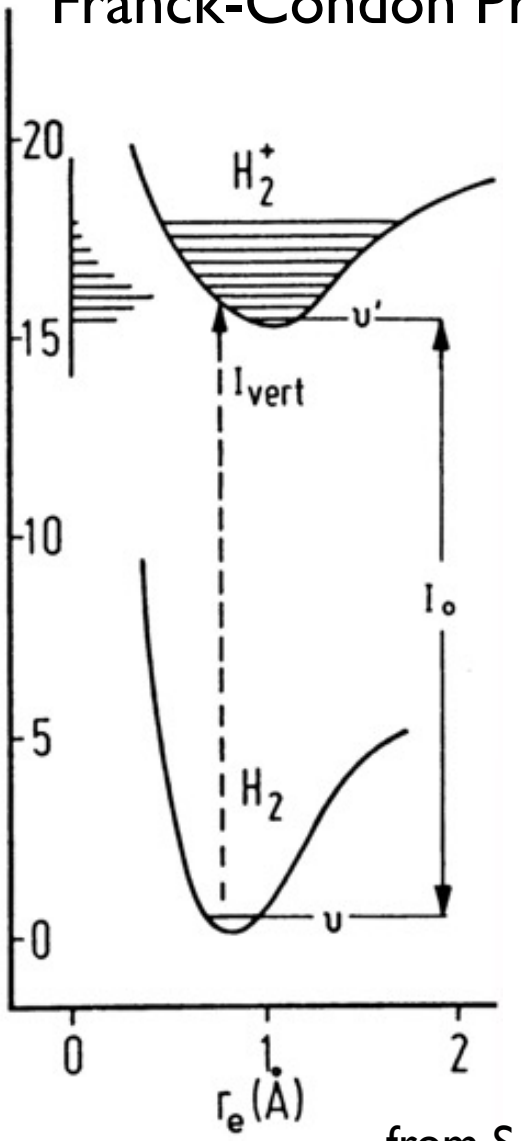
Photoemission from molecular hydrogen



- 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)

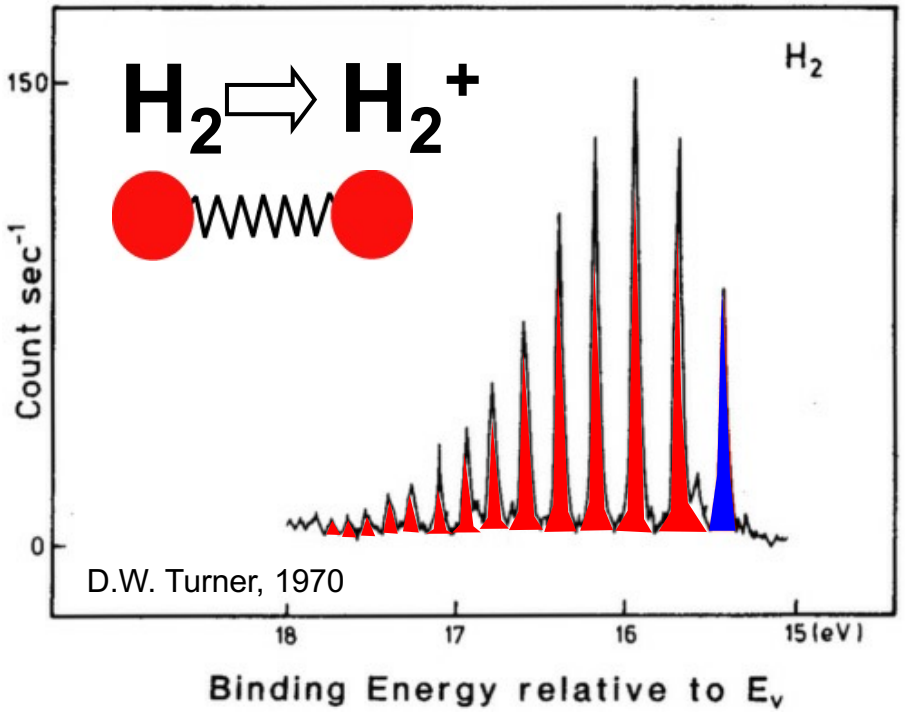
Franck-Condon Effect & electron-phonon coupling

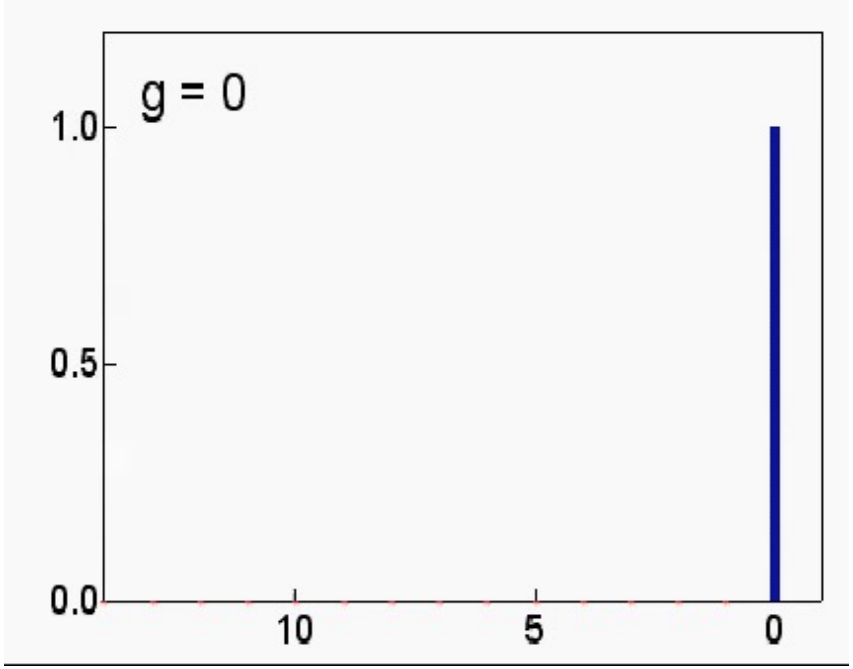
$$\mathcal{H} = \epsilon_0\, c^\dagger c + \sum_{\mathbf{q}} \omega_{\mathbf{q}}\, a^\dagger_{\mathbf{q}} a_{\mathbf{q}} + \sqrt{g\omega_0^2} \sum_{\mathbf{q}} c^\dagger c (a_{\mathbf{q}} + a^\dagger_{-\mathbf{q}})$$

electron energy

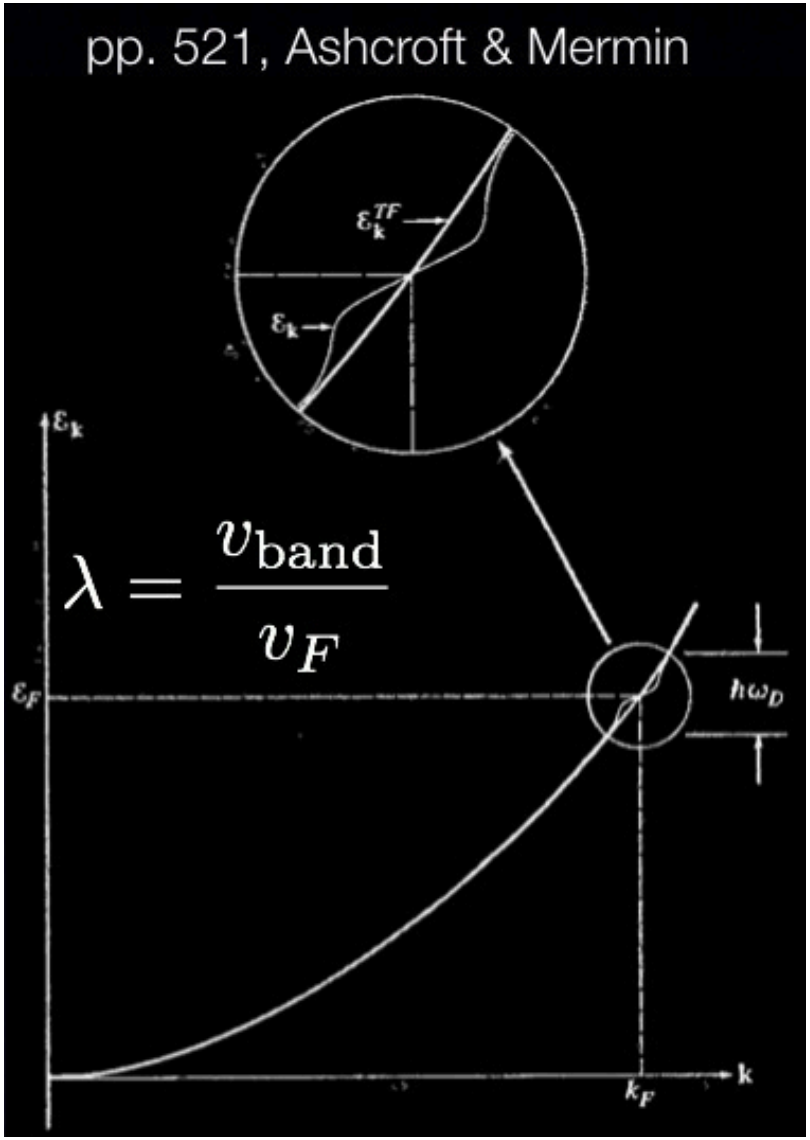
phonon energy

electron-phonon coupling

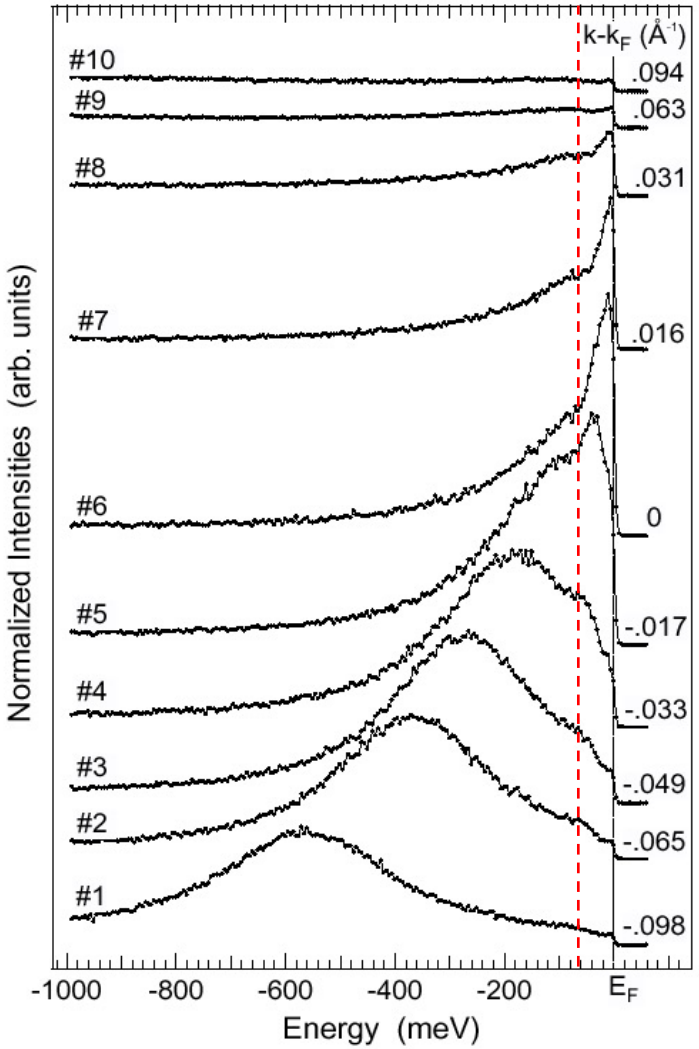




Electron-phonon coupling in solids : ARPES

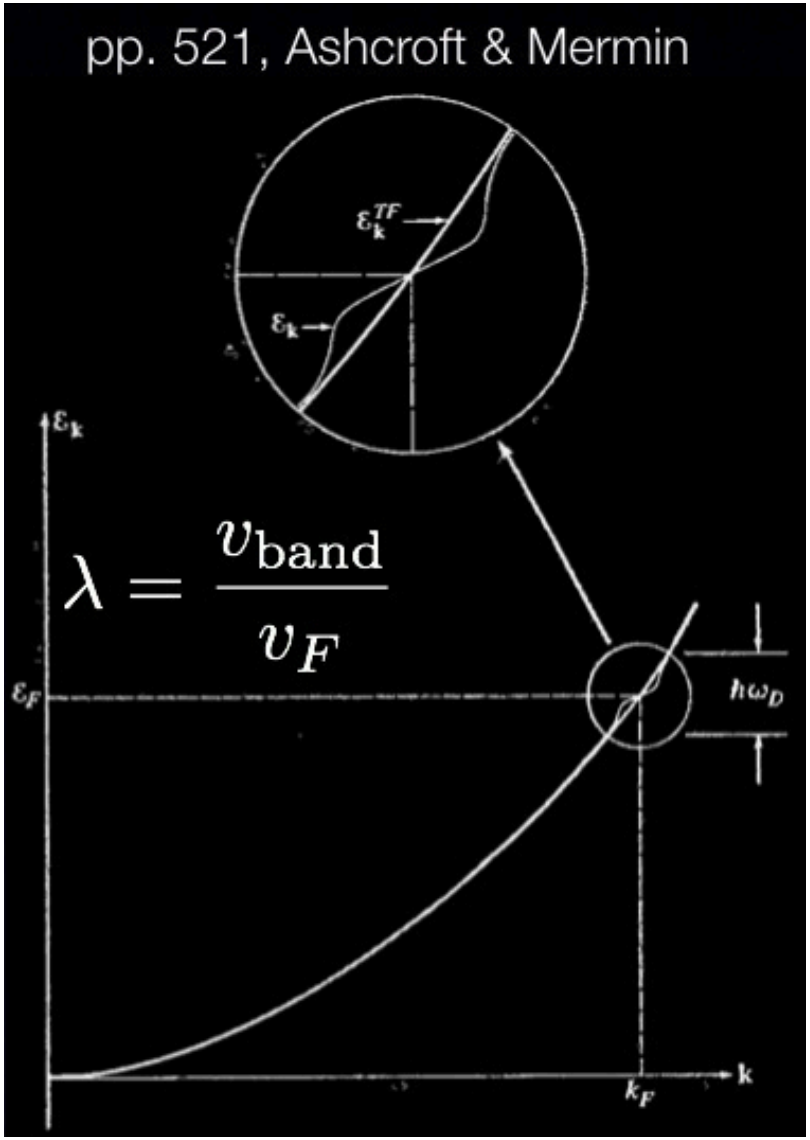


Be (0001) : Electron-phonon

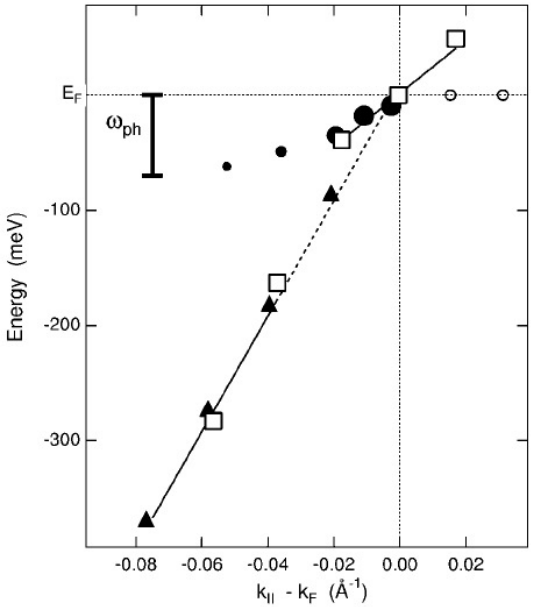


M. Hengsberger *et al.*, PRL ('99)

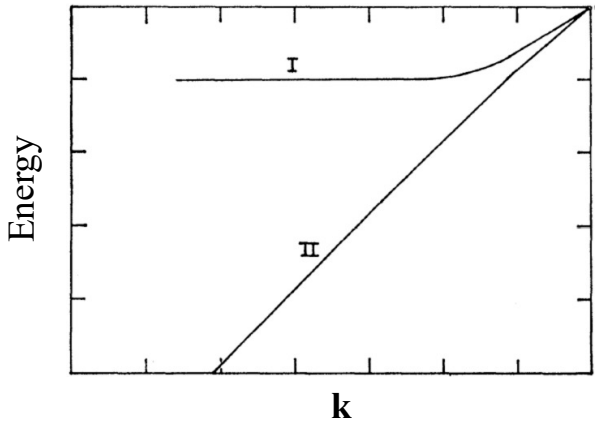
Electron-phonon coupling in solids : ARPES



Be (0001) : Electron-phonon



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

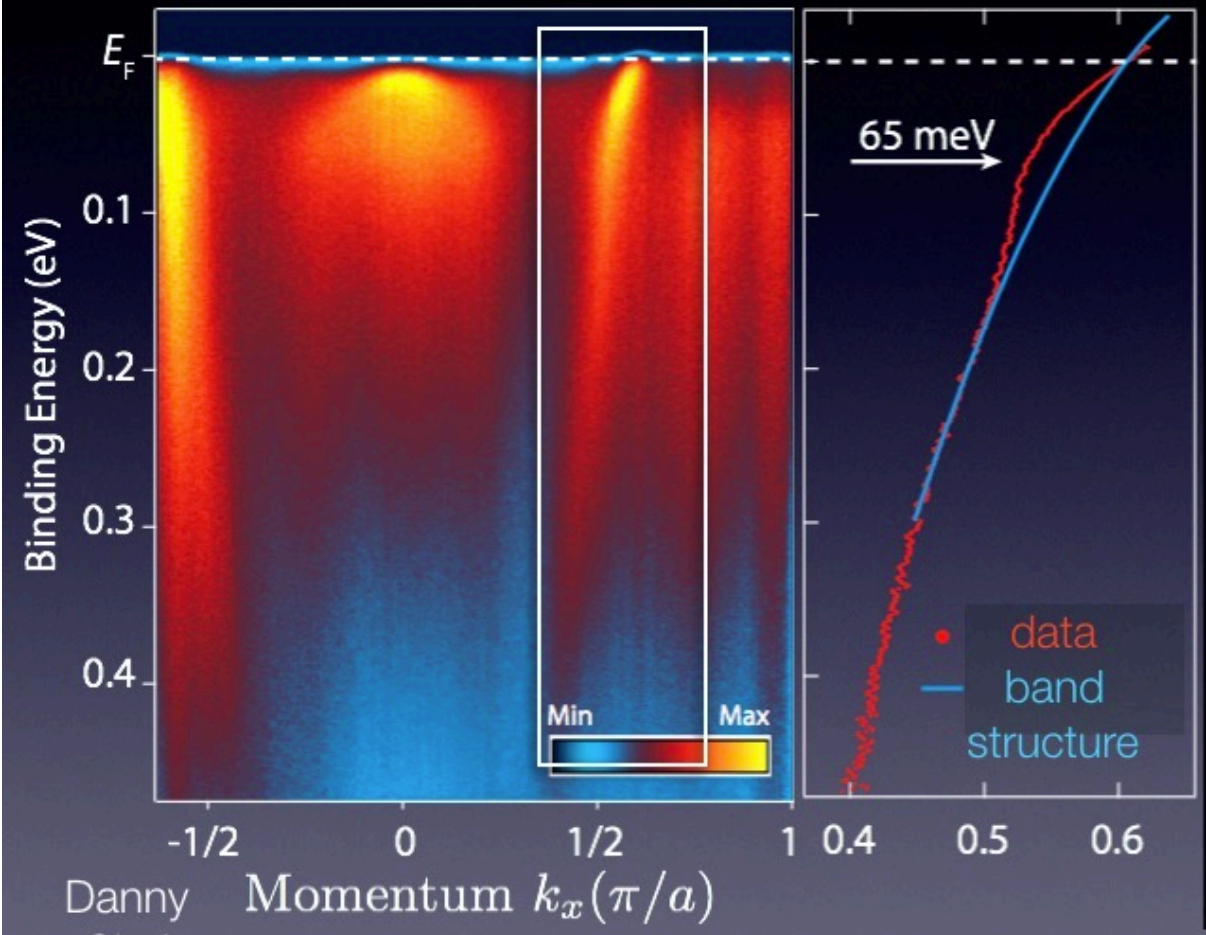


S. Engelsberg and J.R. Schrieffer, Phys. Rev ('63)

A real example of electron-phonon interactions

PARADIM

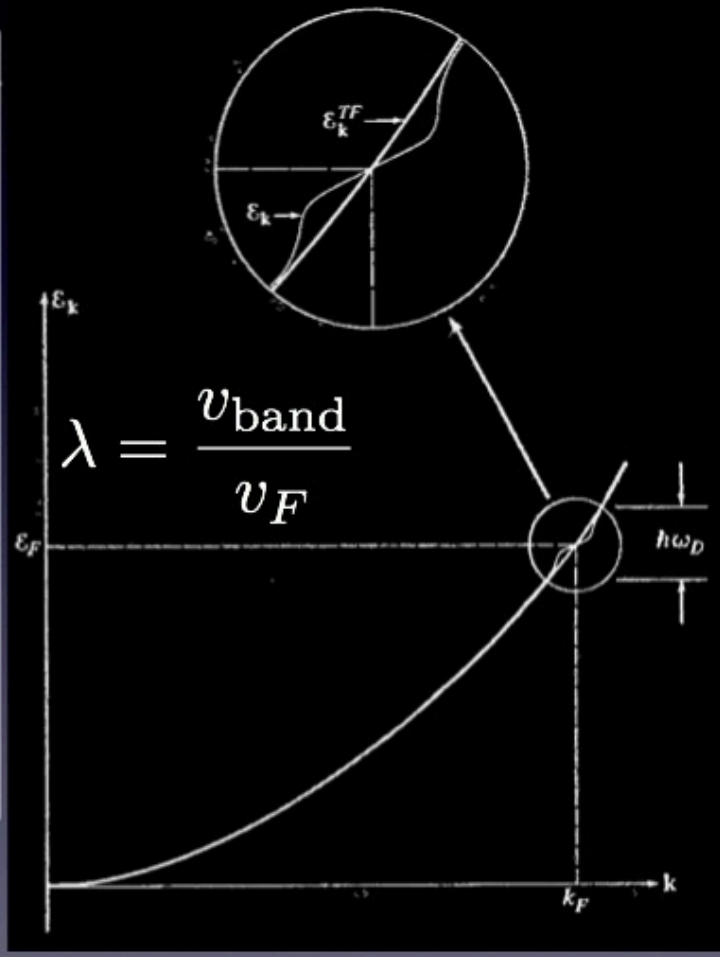
$SrRuO_3$: correlated oxide



Danny Shai

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

pp. 521, Ashcroft & Mermin



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

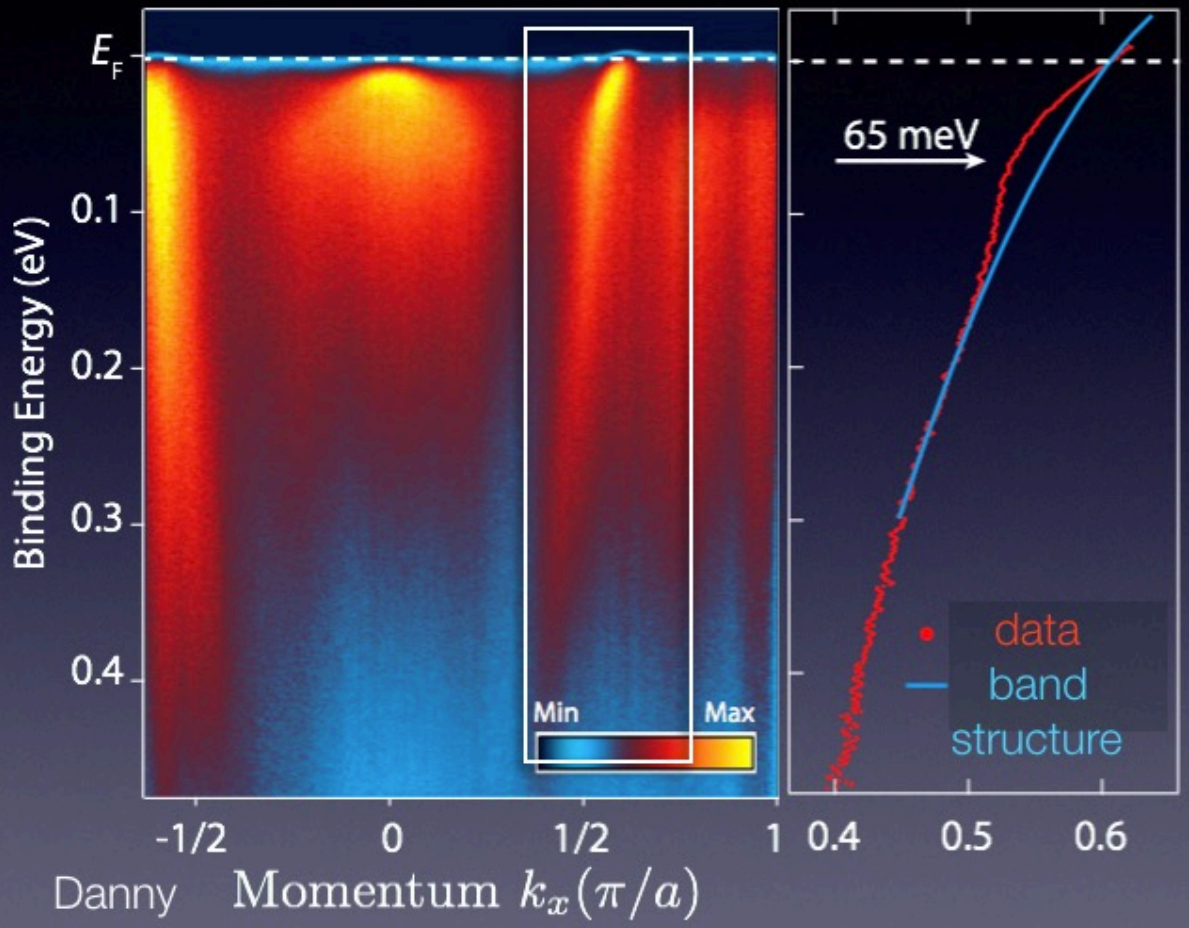
$$\frac{m^*}{m_b} = 3.9$$

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

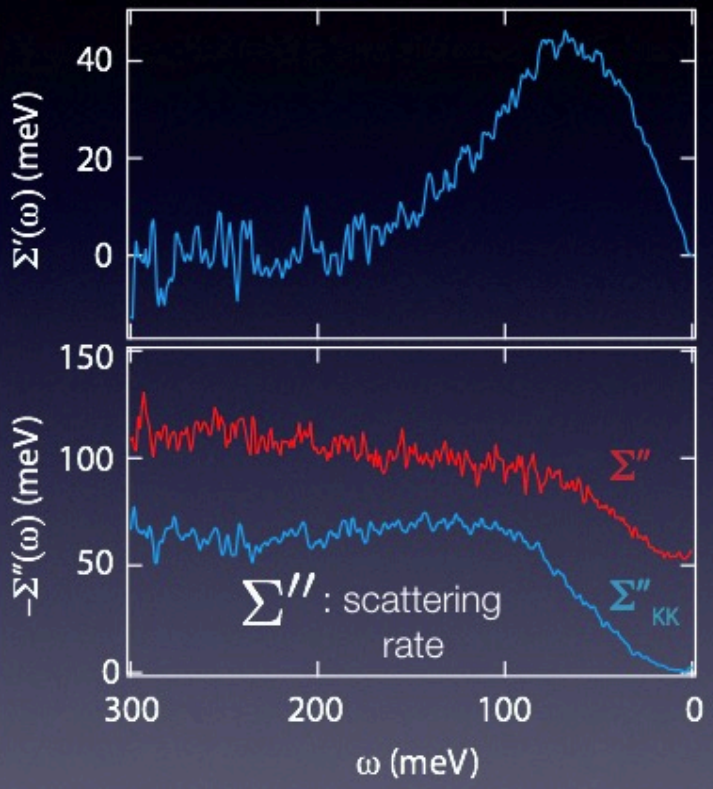
A real example of electron-phonon interactions

PARADIM

$SrRuO_3$: correlated oxide



Σ : self-energy



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)