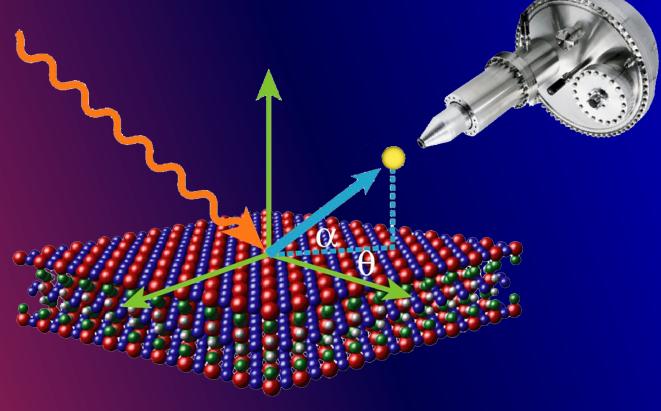




Angle Resolved Photoemission Spectroscopy

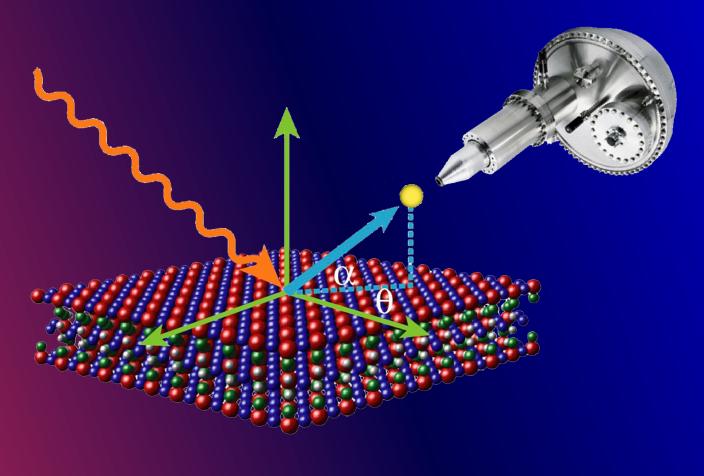
From fundamentals to the heart of condensed matter





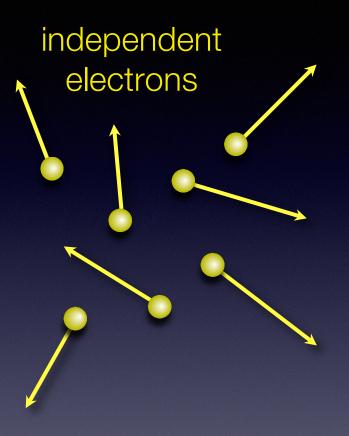


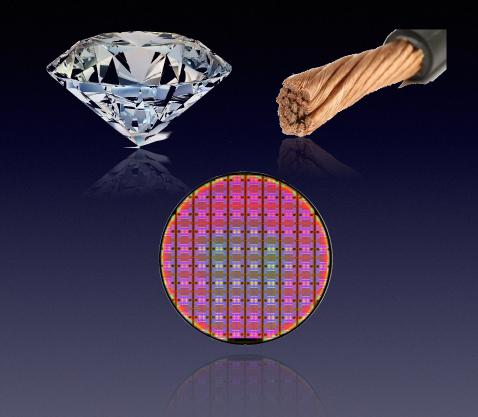
Lecture #2: From Particles to Quasiparticles



6-7 FEBRERO, 2023

the "independent electron" approximation





electron electron-electron interactions KE

nuclei KE

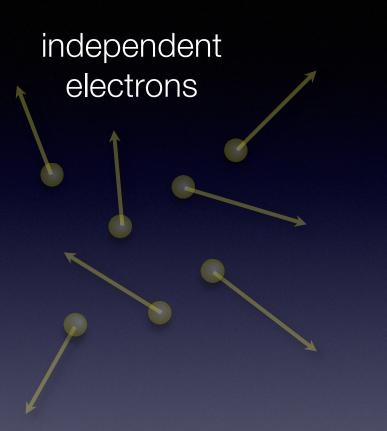
 $-\sum_{i}^{N_e}rac{\hbar^2}{2m}
abla_j^2 + \sum_{i \in I}^{N_e}rac{e^2}{|ec{r}_j-ec{r}_k|} - \sum_{lpha}^{N_i}rac{\hbar^2}{2M_lpha}
abla_lpha^2 - \sum_{i}^{N_e}\sum_{lpha}^{N_i}rac{Z_lpha e^2}{|ec{r}_j-ec{R}_lpha|} + \sum_{lpha \in eta}rac{Z_lpha Z_eta e^2}{|ec{R}_lpha-ec{R}_eta|}$

electron-nuclei interactions

nuclei-nuclei interactions

$$-\sum_{lpha\lleta}^{N_j}rac{Z_lpha Z_eta e^2}{|ec{R}_lpha-ec{R}_eta|}$$

how are "quantum materials" different from conventional ones?



electronic crystal



$$\circ$$
 \circ \circ

$$-\sum_{j}^{N_{e}} \frac{\hbar^{2}}{2m} \nabla_{j}^{2} + \sum_{i \leqslant k}^{N_{e}} \frac{e^{2}}{|\vec{r_{j}} - \vec{r_{k}}|} - \sum_{\alpha}^{N_{i}} \frac{\hbar^{2}}{2M_{\alpha}} \nabla_{\alpha}^{2} - \sum_{j}^{N_{e}} \sum_{\alpha}^{N_{i}} \frac{Z_{\alpha}e^{2}}{|\vec{r_{j}} - \vec{R}_{\alpha}|} + \sum_{\alpha \leqslant \beta}^{N_{j}} \frac{Z_{\alpha}Z_{\beta}e^{2}}{|\vec{R}_{\alpha} - \vec{R}_{\beta}|}$$

nuclei KE

$$+\sum_{lpha}^{N_i}rac{\hbar^2}{2M_lpha}
abla_lpha^2$$

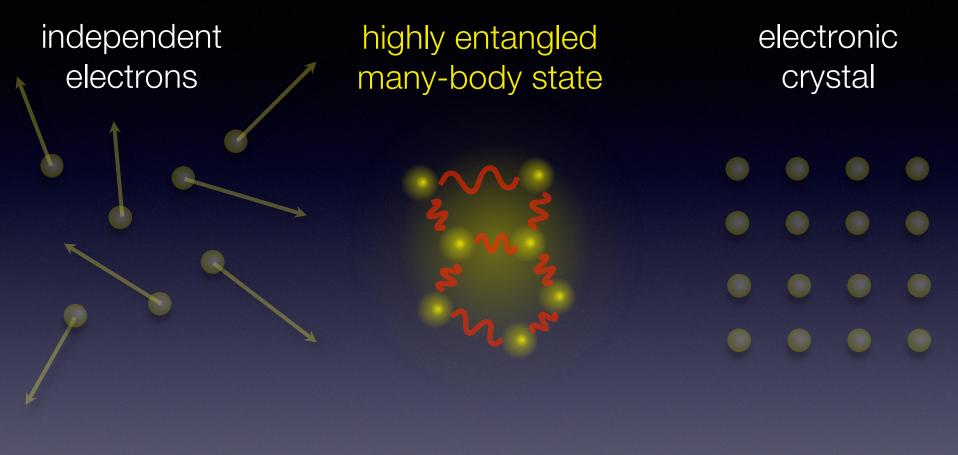
electron-nuclei interactions

$$-\sum_{j}^{N_e}\sum_{lpha}^{N_i}rac{Z_{lpha}e^2}{|ec{r}_j-ec{R}_{lpha}|}.$$

nuclei-nuclei interactions

$$-\sum_{lpha \ll eta}^{N_j} rac{Z_lpha Z_eta e^2}{|ec{R}_lpha - ec{R}_eta|}$$

how are "quantum materials" different from conventional ones?



electron-electron electron interactions

$$+\sum_{i \ll k}^{N_e} rac{e^2}{|ec{r}_j - ec{r}_k|}$$

nuclei KE

$$\sum_{i=1}^{N_i} rac{\hbar^2}{2M_lpha}
abla_lpha^2$$

electron-nuclei interactions

$$-\sum_{j}^{N_{e}} \frac{\hbar^{2}}{2m} \nabla_{j}^{2} + \sum_{j \leqslant k}^{N_{e}} \frac{e^{2}}{|\vec{r_{j}} - \vec{r_{k}}|} - \sum_{\alpha}^{N_{i}} \frac{\hbar^{2}}{2M_{\alpha}} \nabla_{\alpha}^{2} - \sum_{j}^{N_{e}} \sum_{\alpha}^{N_{i}} \frac{Z_{\alpha}e^{2}}{|\vec{r_{j}} - \vec{R_{\alpha}}|} + \sum_{\alpha \leqslant \beta}^{N_{j}} \frac{Z_{\alpha}Z_{\beta}e^{2}}{|\vec{R_{\alpha}} - \vec{R_{\beta}}|}$$

nuclei-nuclei interactions

$$-\sum_{lpha \ll eta}^{N_j} rac{Z_lpha Z_eta e^2}{|ec{R}_lpha - ec{R}_eta|}$$

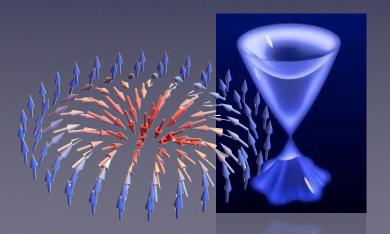
New opportunities presented by quantum materials

higher (room) temperature superconductors?

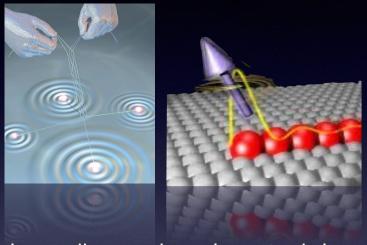




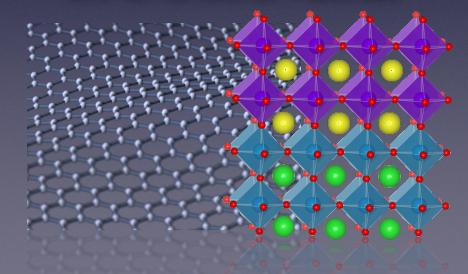
topologically protected states

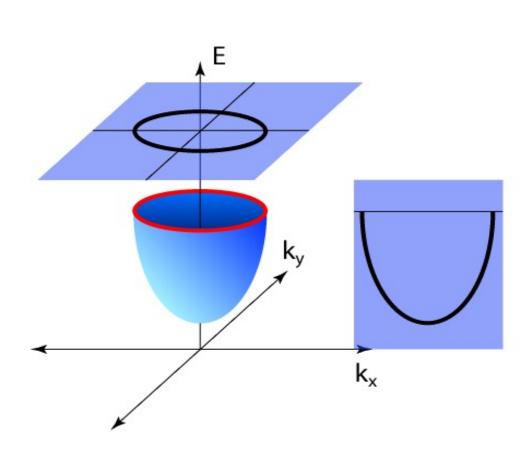


fault-tolerant quantum computing?



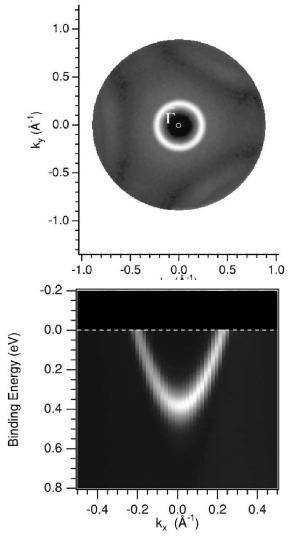
low-dimensional materials





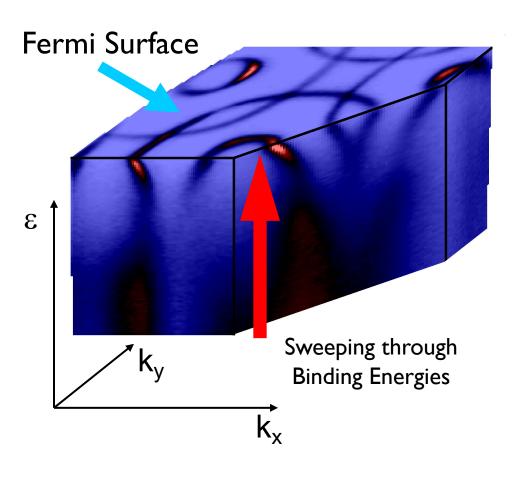
 $E = \hbar^2 \mathbf{k}^2 / 2m^*$

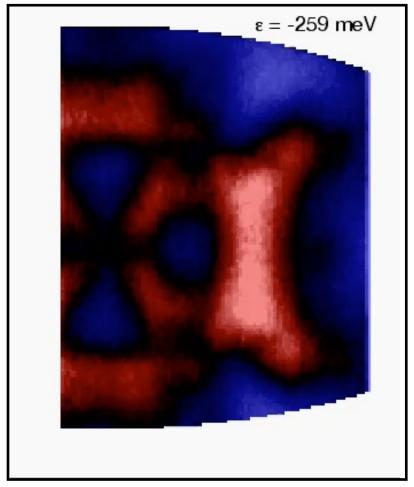


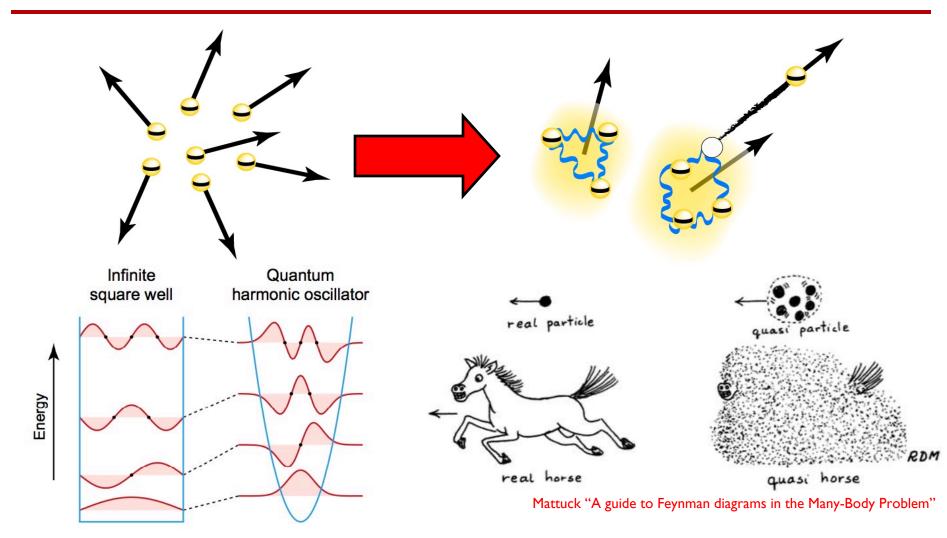


F. Baumberger et al., PRB 64, 195411 (2001)

Sr₂RhO₄: A 4d transition metal oxide with strong spin-orbit coupling

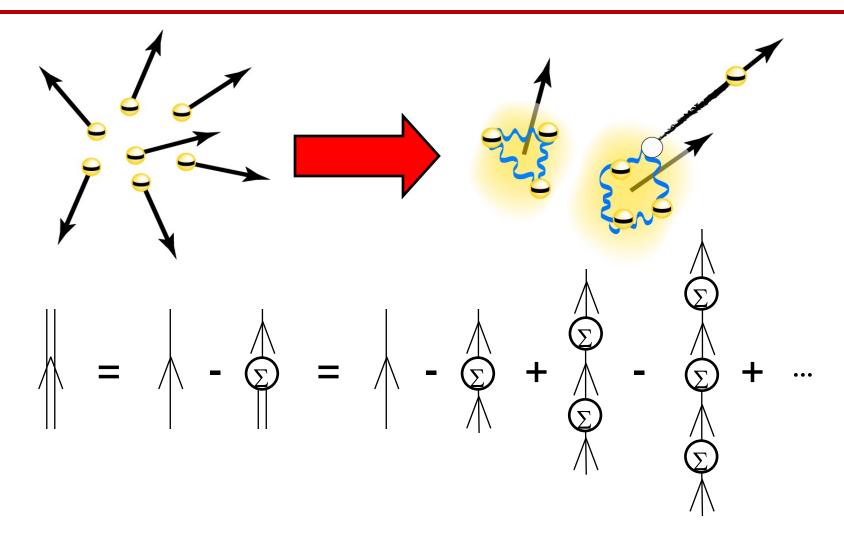






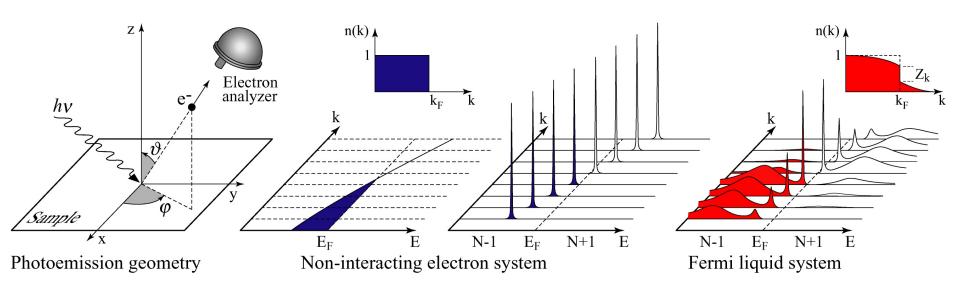
Single-particle spectral function

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



Single-particle spectral function

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



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

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

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

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

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

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 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

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

How to express initial & final states?

$$\Psi_f^N = \overleftarrow{\phi_k} \Psi_f^{N-1}$$

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

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

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

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:

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

For interacting systems, Ψ_f^{N-1} is <u>NOT</u> 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 Interesting part! Energy conservation

<u>Dipole matrix element:</u>

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

- 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

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", $A(k, \omega)$

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

$$A^{-}(k,\omega) = \sum_{m} \left| \left\langle \Psi_{m}^{N-1} \left| c_{k} \right| \Psi_{i}^{N} \right\rangle \right|^{2} \delta\left(w - E_{m}^{N-1} + E_{i}^{N} \right)$$

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

$$\sum_{m} |\langle \Psi_{m}^{N-1} | \Psi_{i}^{N-1} \rangle|^{2}$$

$$\delta(E_{kin} + E_m^{N-1} - E_i^N - h\nu)$$

The photoemission spectrum contains information about the spectral function!

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

Matrix Element (constant*) Spectral Function Fermi-Dirac function

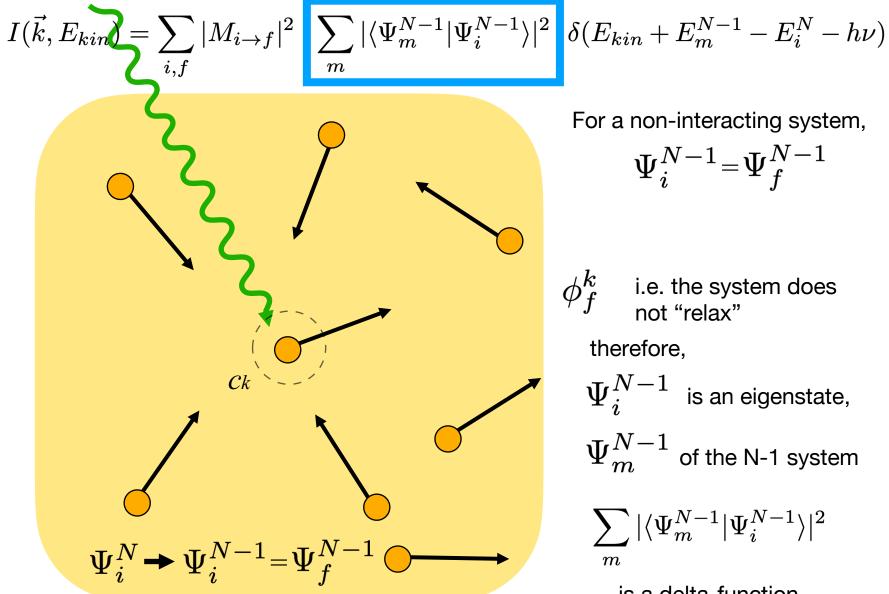
A convenient way of expressing the spectral function is in terms of the "self-energy", which encodes all the information about the electron interactions

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

Real part : $\Sigma'(k,\omega)$: energy renormalization Self-energy is causal; Kramers-Kronig relation Imaginary part : $\Sigma''(k,\omega)$: quasiparticle lifetime

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

A graphical interpretation of the photoemission process ARPES2023



$$\delta(E_{kin} + E_m^{N-1} - E_i^N - h\nu)$$

For a non-interacting system,

$$\Psi_i^{N-1} = \Psi_f^{N-1}$$

 ϕ_f^k i.e. the system does not "relax"

therefore,

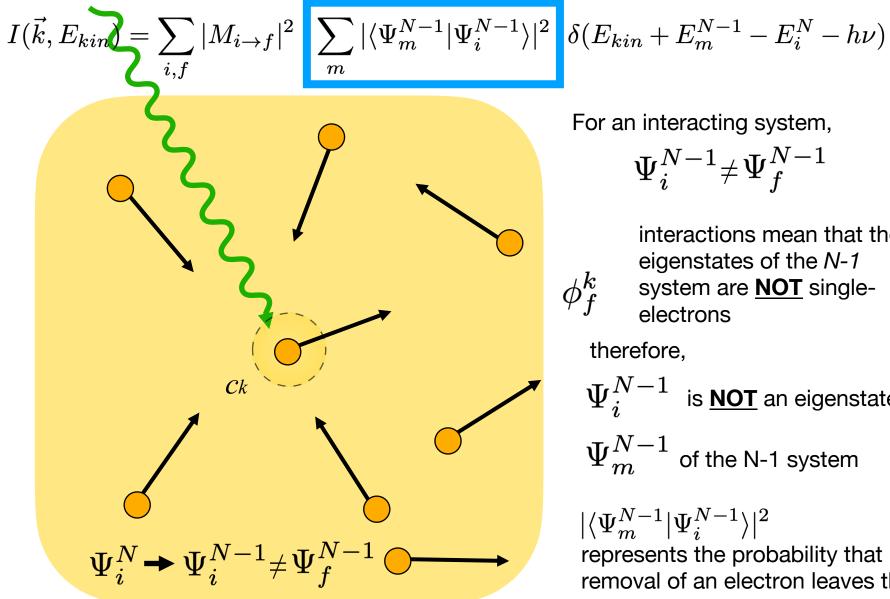
 Ψ^{N-1}_{i} is an eigenstate,

 Ψ_m^{N-1} of the N-1 system

$$\sum_{m} |\langle \Psi_{m}^{N-1} | \Psi_{i}^{N-1} \rangle|^{2}$$

is a delta-function

A graphical interpretation of the photoemission process ARPES2023



$$\delta(E_{kin} + E_m^{N-1} - E_i^N - h\nu)$$

For an interacting system,

$$\Psi_i^{N-1}
eq \Psi_f^{N-1}$$

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

therefore,

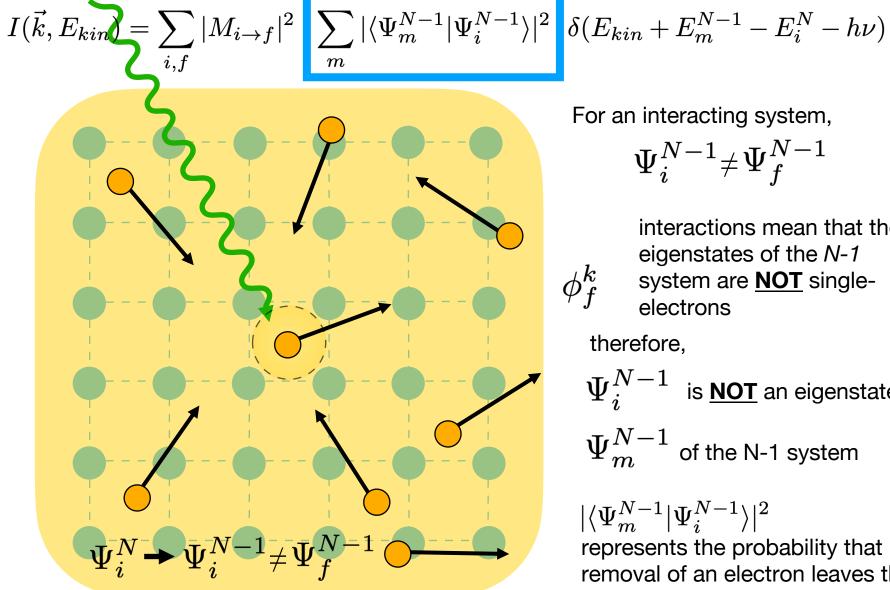
$$\Psi_i^{N-1}$$
 is **NOT** an eigenstate,

$$\Psi_m^{N-1}$$
 of the N-1 system

$$|\langle \Psi_m^{N-1} | \Psi_i^{N-1} \rangle|^2$$
 represents the probability that

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

A graphical interpretation of the photoemission process ARPES2023



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

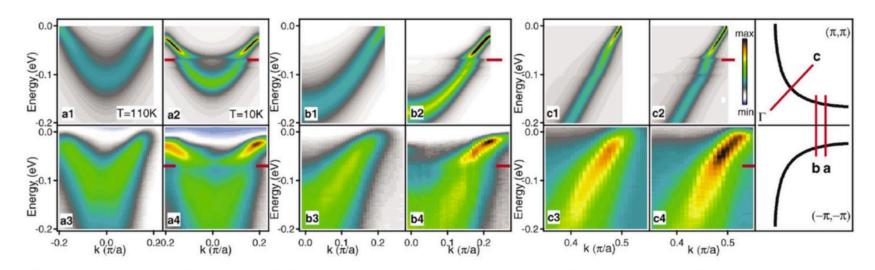


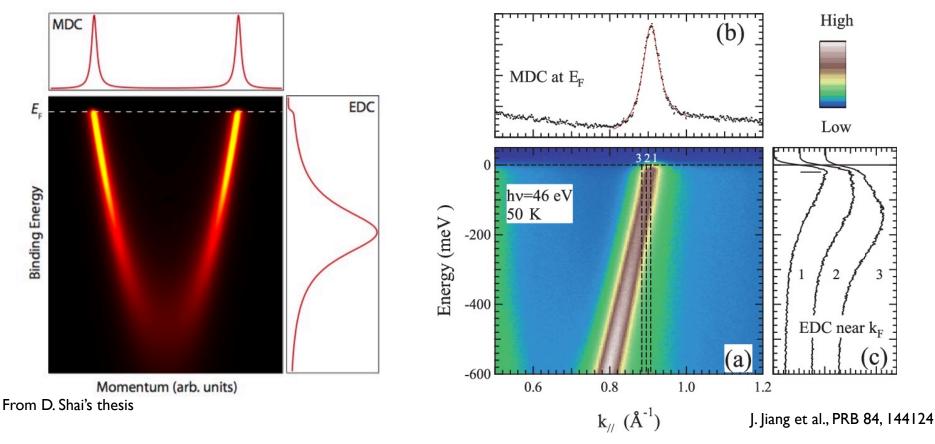
FIG. 3 (color). Image plots of the calculated spectral functions in the normal (a1,b1,c1) and superconducting (a2,b2,c2) states compared to the spectral functions in the normal (a3,b3,c3) and superconducting (a4,b4,c4) states measured in $\text{Bi}_2\text{Sr}_2\text{Ca}_{0.92}\text{Y}_{0.08}\text{Cu}_2\text{O}_{8+\delta}$ (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

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

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



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

Fermi Liquid Theory

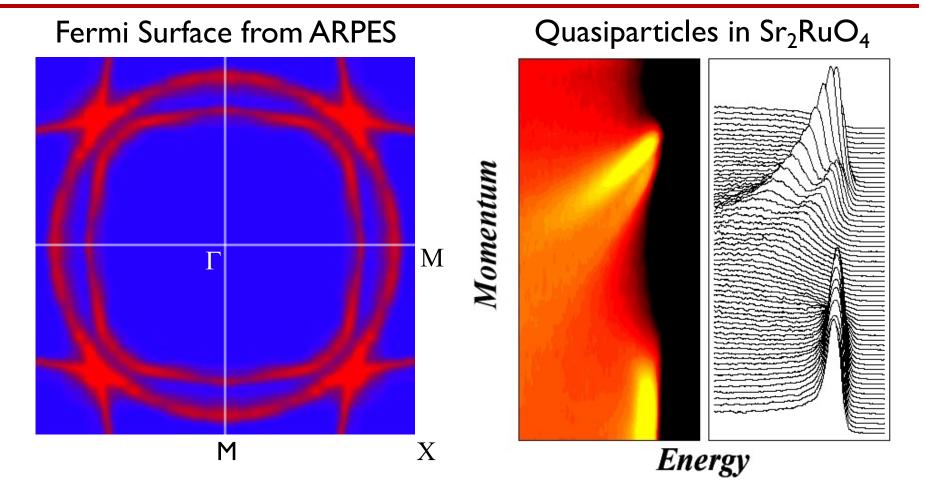


Backbone for Single Particle Approximations

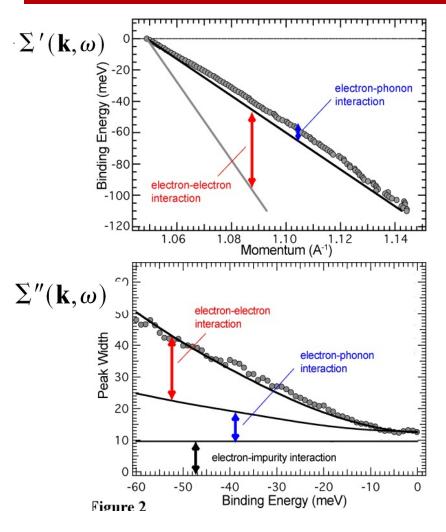
Band Structure of Metals, Semiconductors, Insulators

What can we test in FL Theory by ARPES?

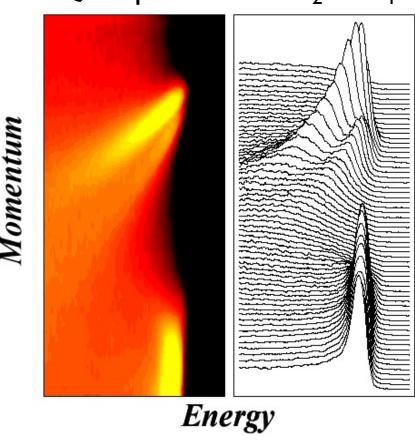
 $\begin{array}{c} \text{Luttinger Volume} \\ \text{Sharp coherent quasiparticles } (\text{Im}\Sigma \ (\omega) < \epsilon_k) \\ \text{Energy dependence of } \text{Im}\Sigma \ (\omega) \end{array}$



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)



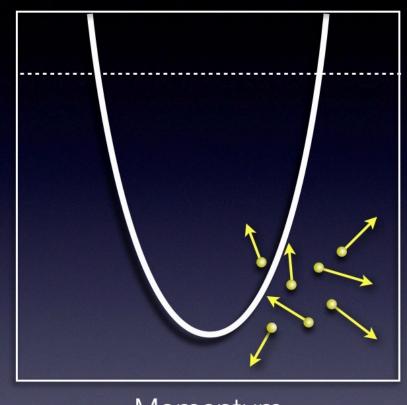
Quasiparticles in Sr₂RuO₄



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

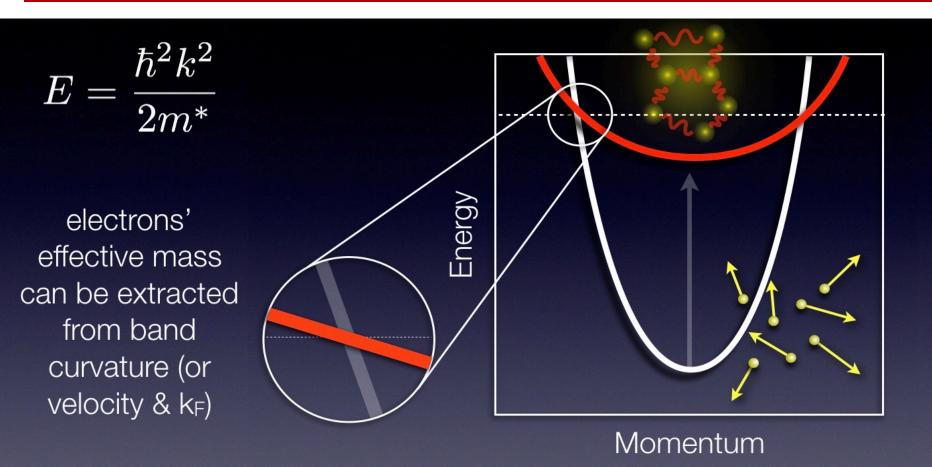
$$E = \frac{\hbar^2 k^2}{2m^*}$$

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

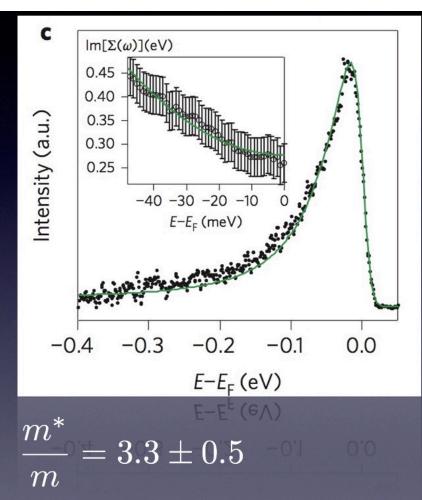


Momentum

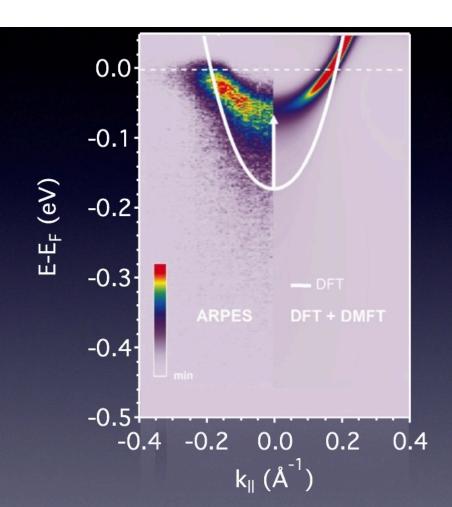
Typically estimate mass renormalization by comparing the experimental Fermi velocity (slope) with the predicted velocity without interactions*



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



in agreement with optical & thermodynamic measurements



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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- D. 4 and 5
- E. All of the above

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

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

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

- A. Yes
- B. No

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$$I(\vec{k},E_{kin}) = \sum_{i,f} |M_{i\rightarrow f}|^2 \sum_{m} |\langle \Psi_m^{N-1}|\Psi_i^{N-1}\rangle|^2 \\ \delta(E_{kin}+E_m^{N-1}-E_i^N-h\nu)$$
 For an interacting system,
$$\Psi_i^{N-1} \neq \Psi_f^{N-1}$$
 interactions mean that the eigenstates of the N-1 system are NOT single-electrons therefore,
$$\Psi_i^{N-1} \text{ is } \underline{\text{NOT}} \text{ an eigenstate}$$

$$\Psi_m^{N-1} \text{ of the N-1 system}$$

$$|\langle \Psi_m^{N-1}|\Psi_i^{N-1}\rangle|^2 \text{ represents the probability that removal of an electron leaves the probability that removal of the leaves the probability that removal of the leaves the probability that removal of the leaves the probability that the leaves the probability that the leaves the lea$$

$$\delta(E_{kin} + E_m^{N-1} - E_i^N - h\nu)$$

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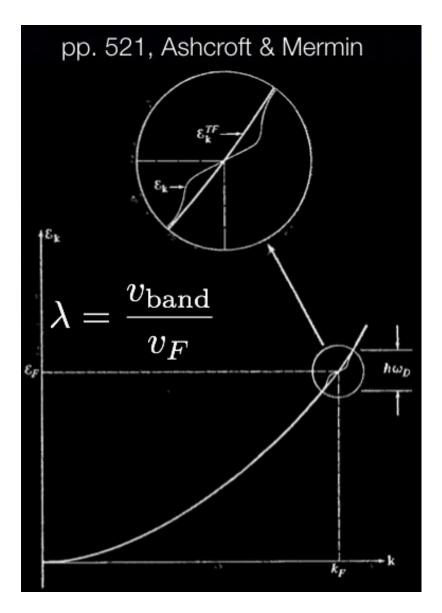
therefore,

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 is **NOT** an eigenstate,

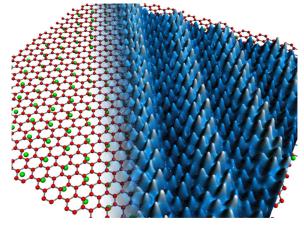
$$\Psi_m^{N-1}$$
 of the N-1 system

$$|\langle \Psi_m^{N-1} | \Psi_i^{N-1} \rangle|^2$$
 represents the probability that

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



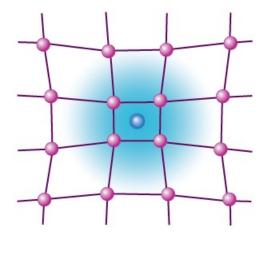
Charge Density Waves

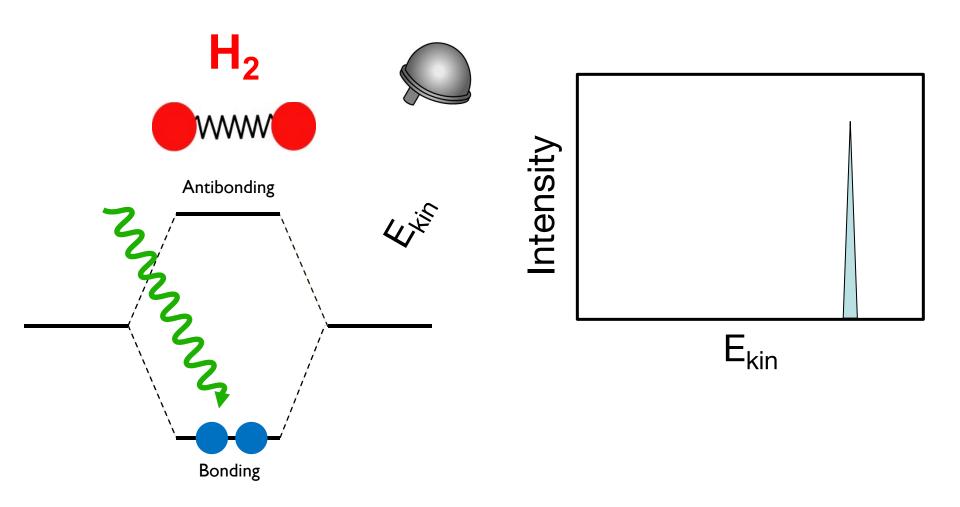


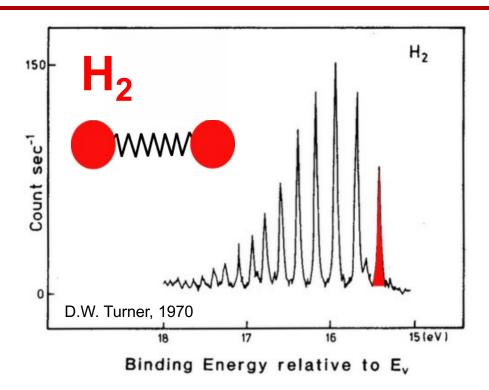
Superconductivity



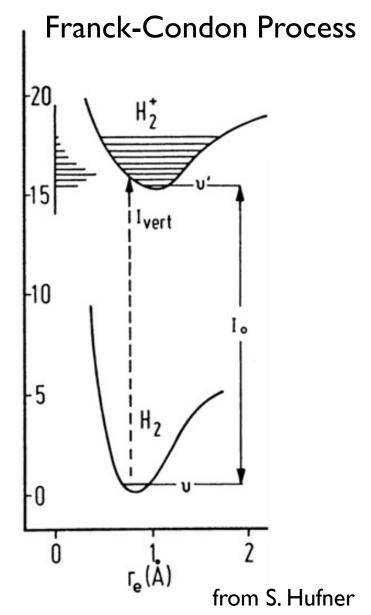
Polarons







- Most spectral weight in shake-off peaks (incoherent part of spectral function)
 - Only lowest-energy transition (0-0) analogous to quasiparticle

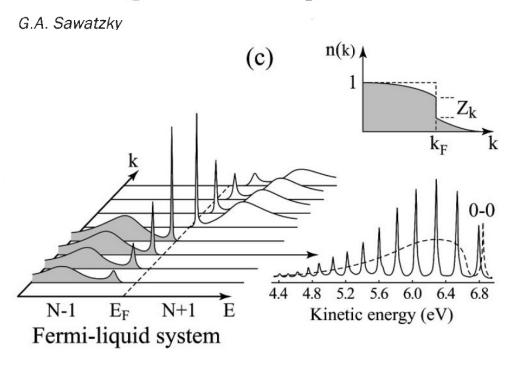


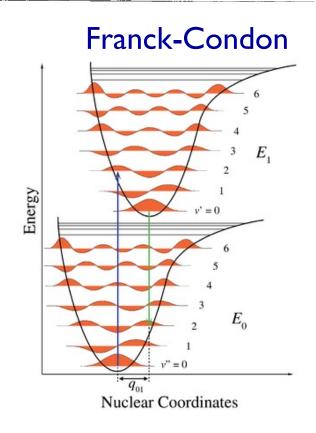
NEWS AND VIEWS

HIGH-TEMPERATURE SUPERCONDUCTIVITY

Nature 342, 480 (1989)

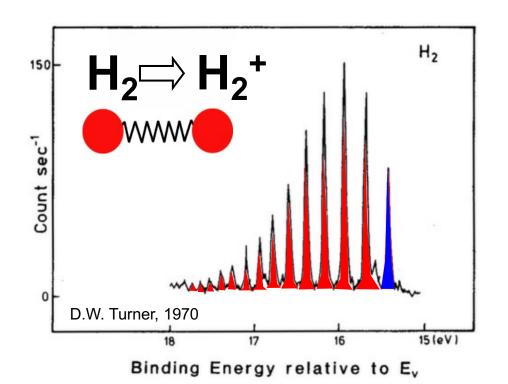
Testing Fermi-liquid models

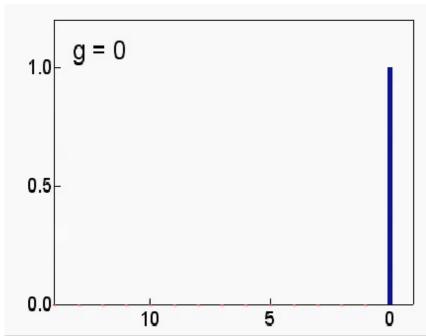


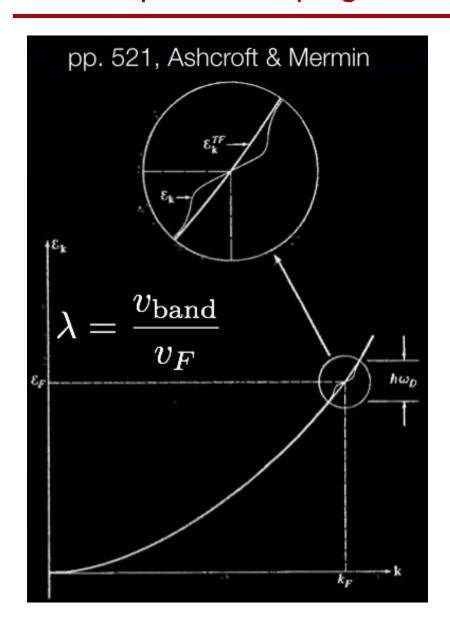


"In gaseous hydrogen, the equilibrium bond length is dependent on the degree of occupation of that level. The electrons are dressed by interatomic displacements. The intensities are given by the Franck-Condon factors, the molecular equivalent of the sudden approximation. The ARPES spectrum of solid hydrogen, 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."

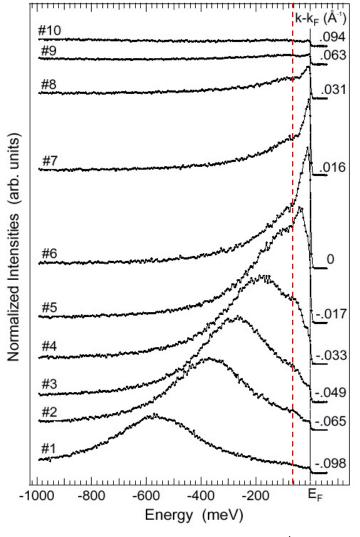
$$\mathcal{H} = \epsilon_0 \, c^\dagger c + \sum_{\mathbf{q}} \omega_{\mathbf{q}} \, a_{\mathbf{q}}^\dagger a_{\mathbf{q}} + \sqrt{g} \omega_0^2 \, \sum_{\mathbf{q}} c^\dagger c (a_{\mathbf{q}} + a_{-\mathbf{q}}^\dagger)$$
 electron phonon energy energy coupling



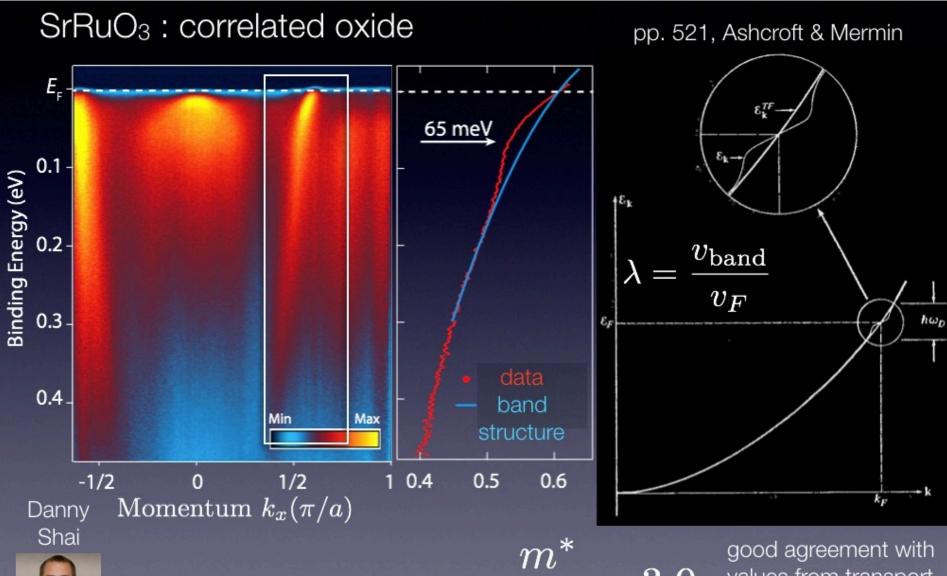




Be (0001): Electron-phonon



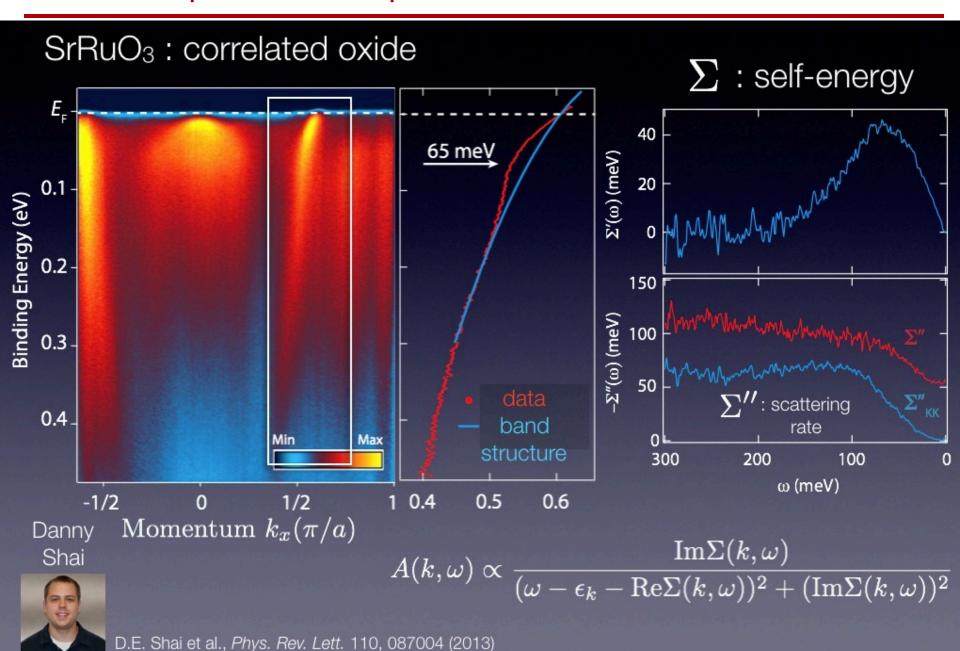
M. Hengsberger et al., PRL ('99)



 \overline{m}_b

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

yalues from transport $(m^*/m_b = 4.1)$



Good references ARPES2023

Textbooks

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

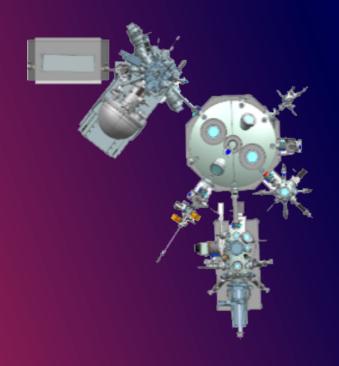
Review articles

- 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)

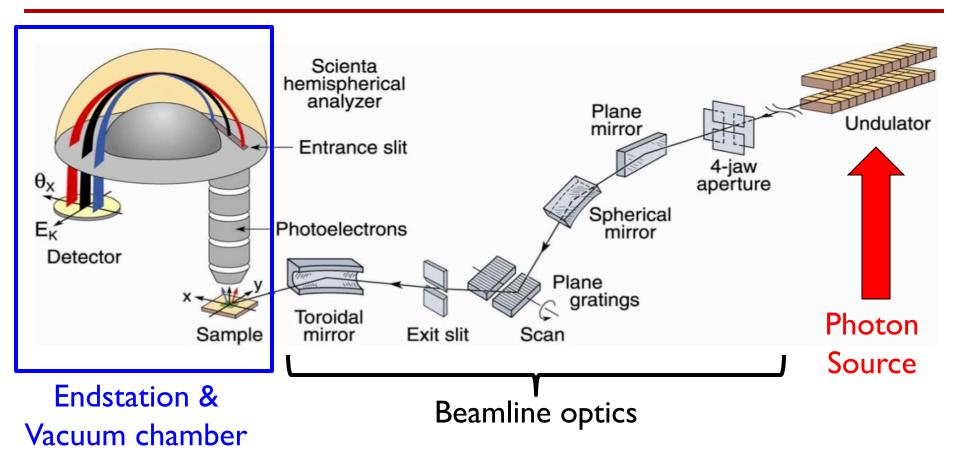
approximately 50 synchrotrons worldwide



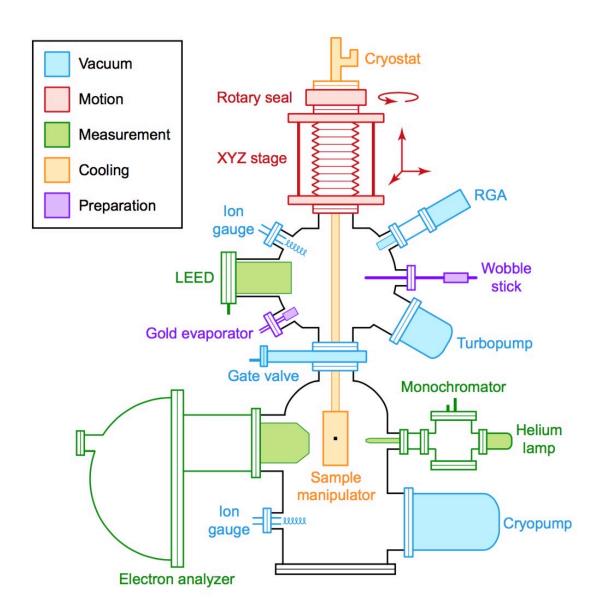
Beamtime at synchrotrons is <u>free of charge</u> – granted based on merit of proposal

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

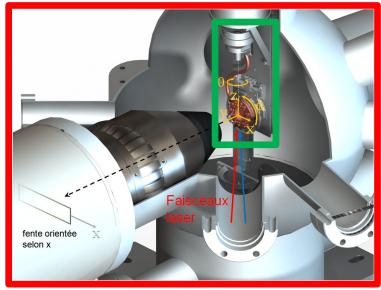


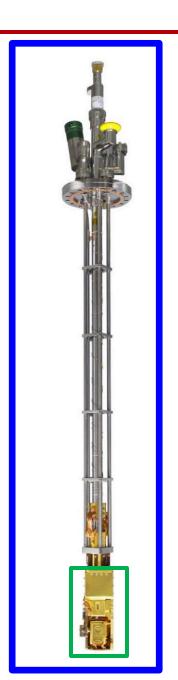
- No good windows for "soft" x-ray, VUV range (10-1000 eV)
- Reflectivities of mirrors in VUV, soft x-ray range of $\sim 50\%$
- Typical resolving powers ($\Delta E / E$) of ~ 5000 : I





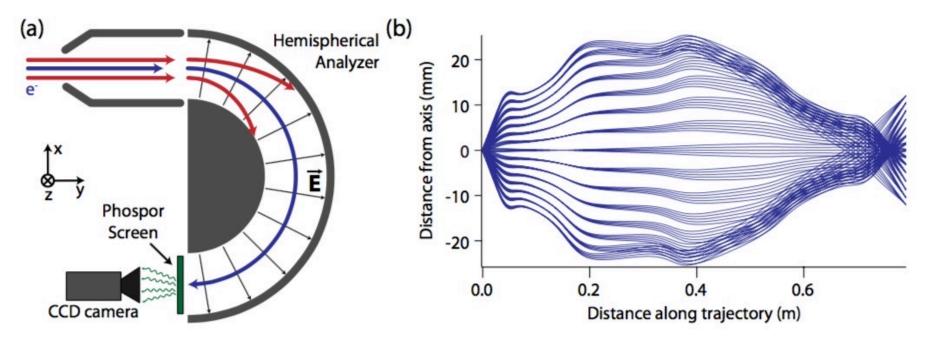




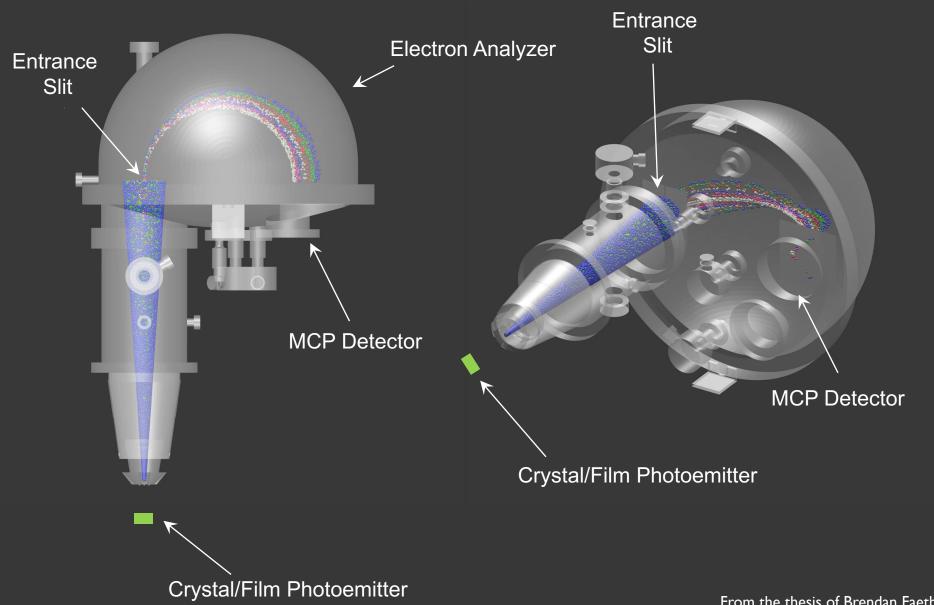






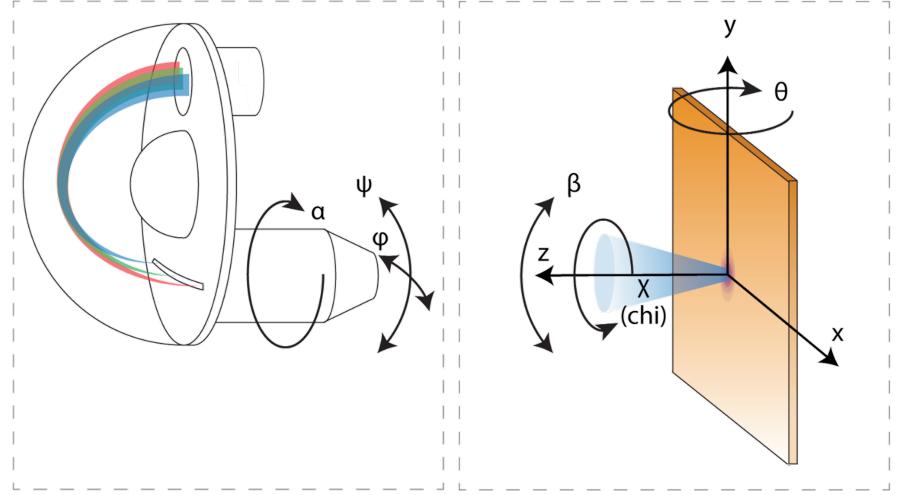




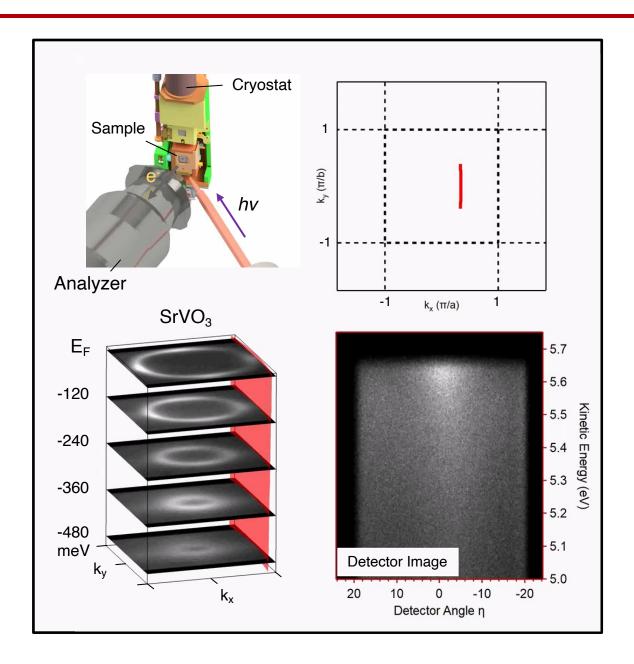


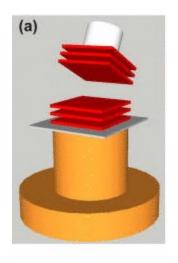
Analyzer Coordinates: (ϕ, ψ, α)

Manipulator Coordinates: $(x, y, z, \theta, \beta, \chi)$



Hemispherical Analyzer Angular Conventions



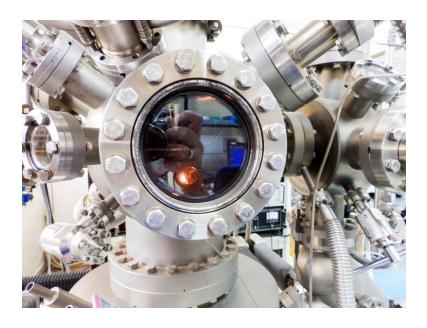






- Bi₂Se₃ crystal should be mounted in a ARPES cryostat prior to start of demo
- Show cleaving procedure with wobble stick, preferably with a nice camera so everyone can see.
- Crystal should be shiny and reflective after cleave, and (ideally) not have any differently oriented fractures

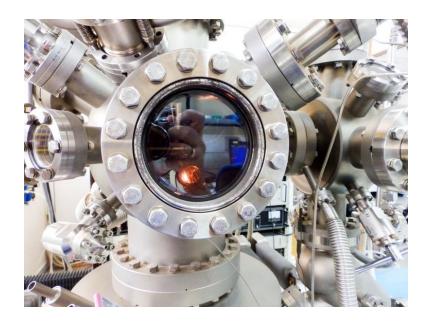
ARPES measurements need to take place in ultrahigh vacuum (10⁻¹⁰ torr or better). Which of the following is the **most important** factor which determines the level of vacuum needed to perform experiments?



- 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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Concept Question #7



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

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

- A. I, 3, 5 & 6
- B. 3 & 5
- C. 2,4 & 5
- D. 2,3 & 5
- E. All of the above

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Consider an ARPES experiment being conducting with an electron analyzer resolution of $\Delta E = 10$ meV and an photon bandwidth of $\Delta E = 2$ meV

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

- A. I0 meV
- B. II meV
- C. I2 meV
- D. I3 meV



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

- A. I0 meV
- B. II meV
- C. I2 meV
- D. 13 meV