When Independent Particle Methods Fail: The Effect of Correlations

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Approaches to the Many-Electron Problem

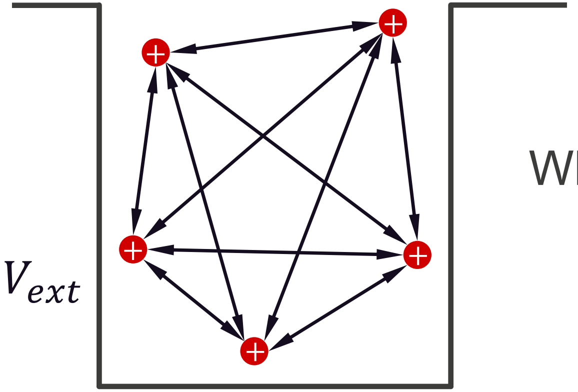
Many-body Hamiltonian

$$\hat{H} = \sum_{\alpha\beta}\int d^3r\, \underbrace{\hat{\psi}^{\dagger}_{\alpha}(r)\left(\frac{-\hbar^2\nabla^2}{2m} + v^{\alpha\beta}_{ext}(r)\right)\hat{\psi}_{\beta}\,(r)}_{\text{Electronic Kinetic Energy}} + \sum_{\alpha\beta\gamma\delta}\iint d^3r\, d^3r'\, \underbrace{\hat{\psi}^{\dagger}_{\alpha}(r)\hat{\psi}^{\dagger}_{\beta}(r')v^{\delta\gamma}_{\alpha\beta}(r,r')\hat{\psi}_{\gamma}(r')\hat{\psi}_{\delta}(r)}_{\text{Interactions}}$$

$\alpha\beta\gamma\delta \in$ spin

Wavefunction based approaches

Key Quantity: $\Psi(\{r_j\})$

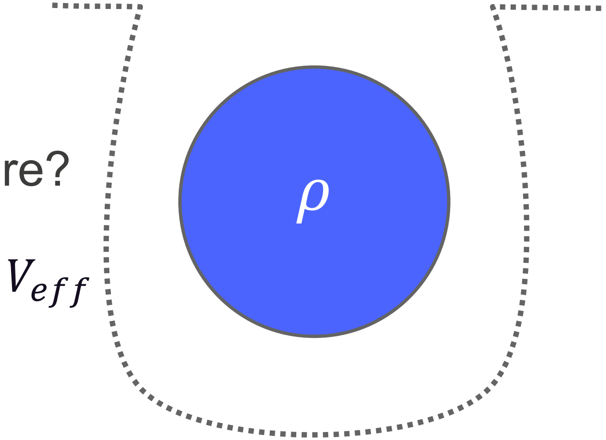


Observables: $\langle\Psi|\hat{O}|\Psi\rangle$

What do we fail to capture?
What is missing?

Reduced quantity based approaches

Key Quantity: Simpler physical quantitiy, e.g., $\rho(r)$



Observables: $\mathrm{F}[\rho]$

Correlations

Independent
electron theory

Weak
Correlations

Strong
Correlations

Correlations

Quantities A and B are *Correlated* if

$$\langle AB\rangle \neq \langle A\rangle\langle B\rangle$$

Measure of Correlation

$$C_{AB} = \langle AB\rangle - \langle A\rangle\langle B\rangle$$

Spatial

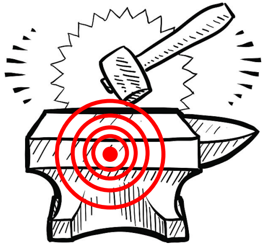
$$\langle A(rt)B(r't)\rangle \neq \langle A(rt)\rangle\langle B(r't)\rangle$$

Temporal

$$\langle A(rt)B(rt')\rangle \neq \langle A(rt)\rangle\langle B(rt')\rangle$$

Strong Correlations

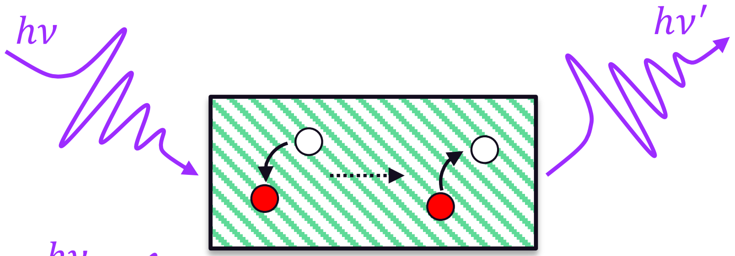
How Do We Measure Correlations?...



…Hit it with a hammer and listen

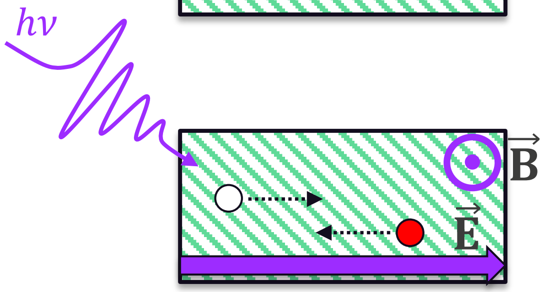
$$\langle n(rt) n(r't') \rangle$$

$$\langle \boldsymbol{S}(rt) \cdot \boldsymbol{S}(r't') \rangle$$



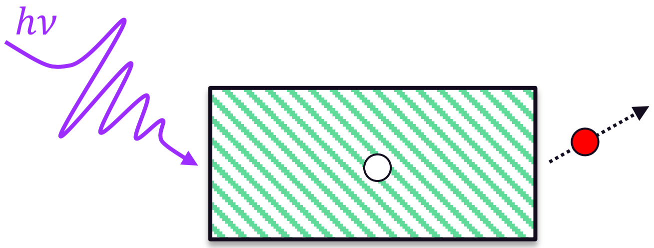
Pair Correlation Function for Electrons and Spin

$$\langle \boldsymbol{j}(rt) \cdot \boldsymbol{j}(r't') \rangle$$



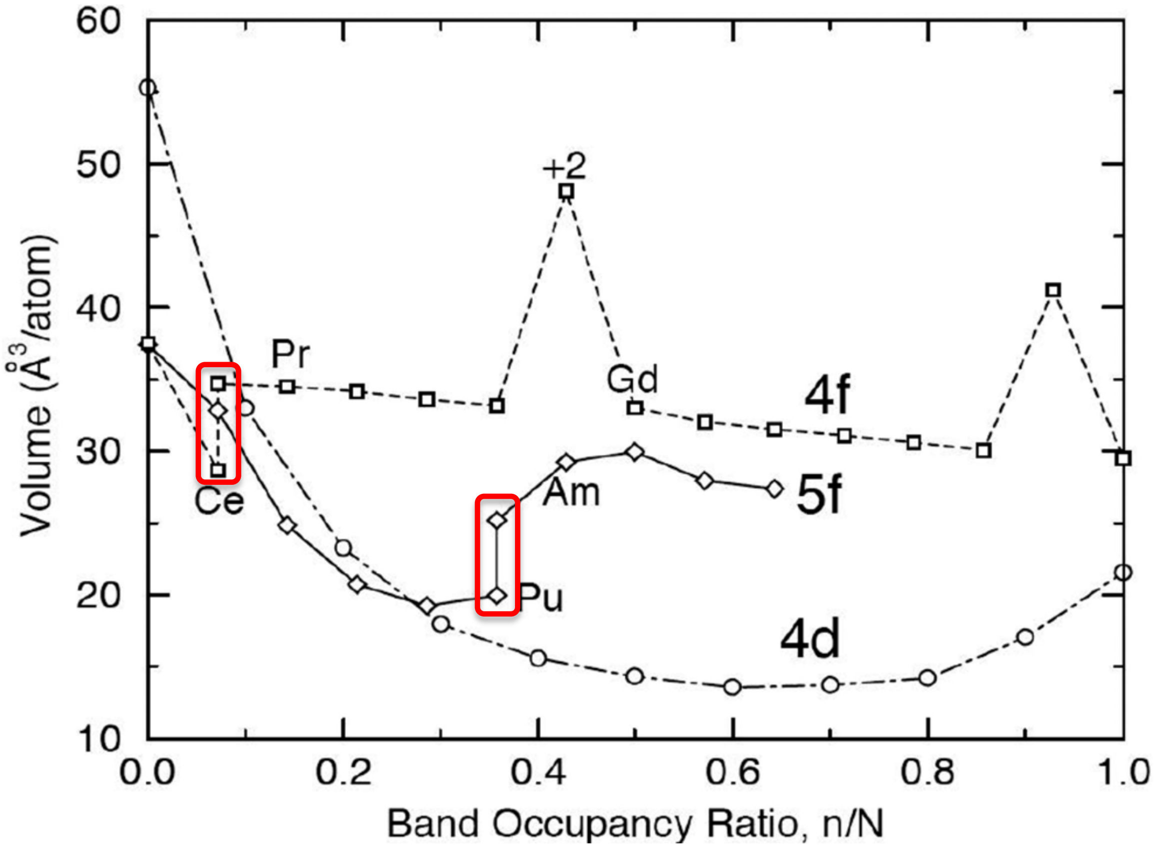
Electrictrical and Optical Properties

$$\langle \{ \psi(rt), \psi^{\dagger}(r't') \} \rangle$$

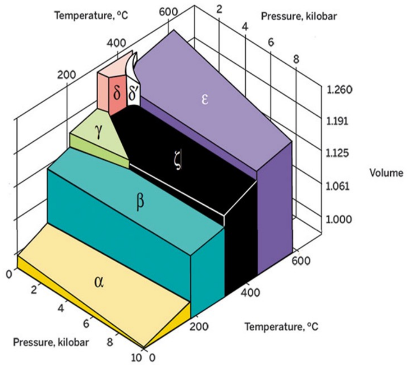


Removing (Adding) Electrons

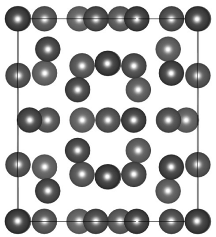
Signatures of Correlations: Ground State / Thermodynamic



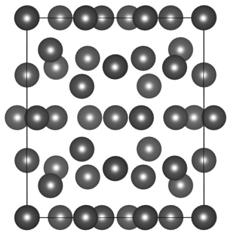
Delocalized → Localized
Behavior



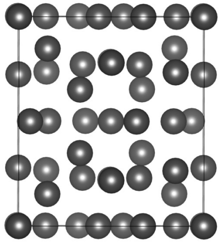
Experimental



VASP Relaxation

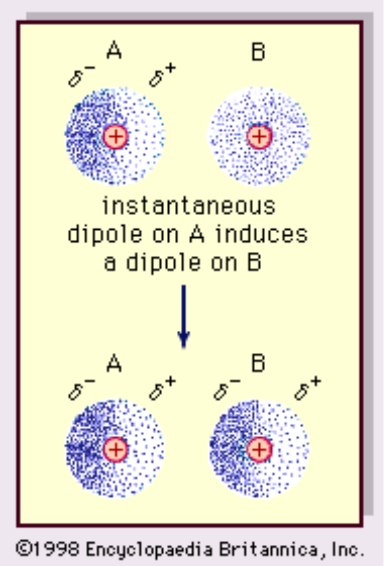


WIEN2k Relaxation

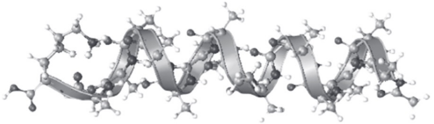


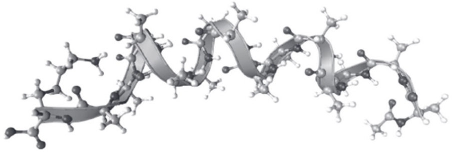
Los Alamos Sci. **2000**, *26* (II), 312
Project# 20210795DI - PI: Hernandez.

Signatures of Correlations: Ground State / Thermodynamic



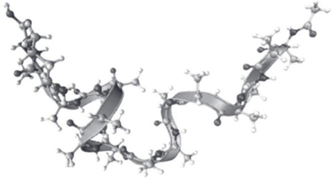
0 ps





30 ps, including vdW

α –helix structure in agreement with experiment

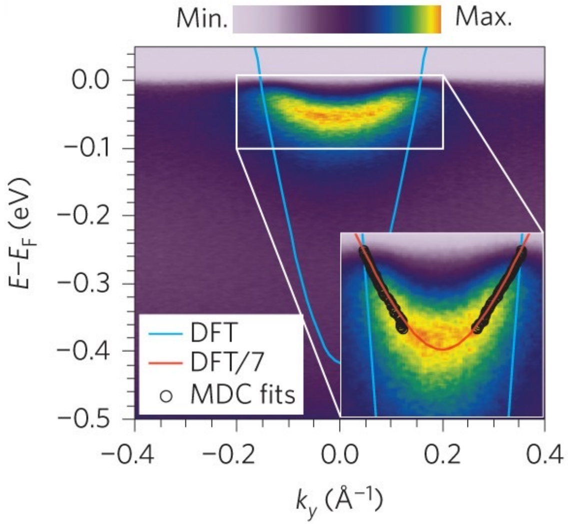


30 ps, no vdW

An open structure

Signatures of Correlations: Electron Addition/Removal

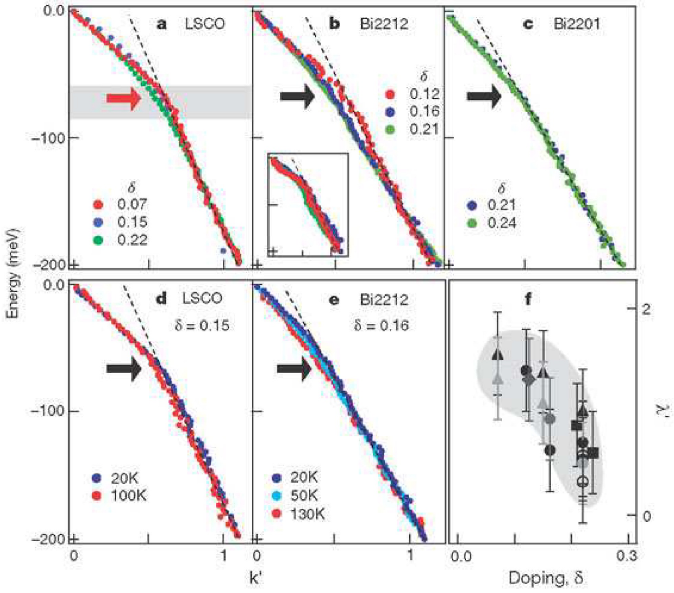
$LiNiO_3$



Renormalization

PDC King, et al. Nature Nanotechnology 9, 443–447 (2014)

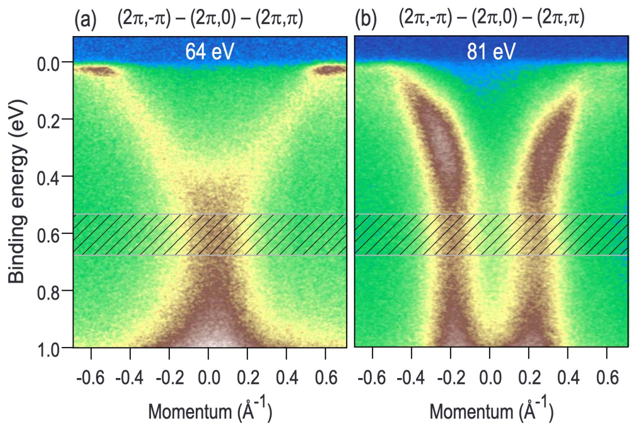
Various Cuprates



Kinks

Zhou, X. J., et al. Handbook of high-temperature superconductivity: theory and experiment 87-144 (2007).

$Bi_2Sr_2CaCu_2O_8$

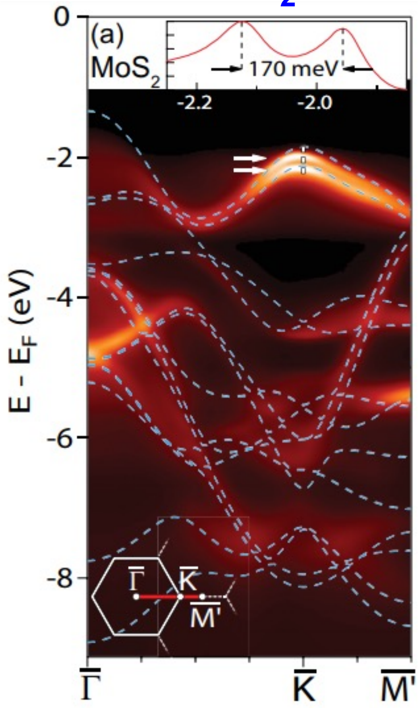


Waterfalls

D. S. Inosov, et al. Phys. Rev. B 77, 212504 (2008)

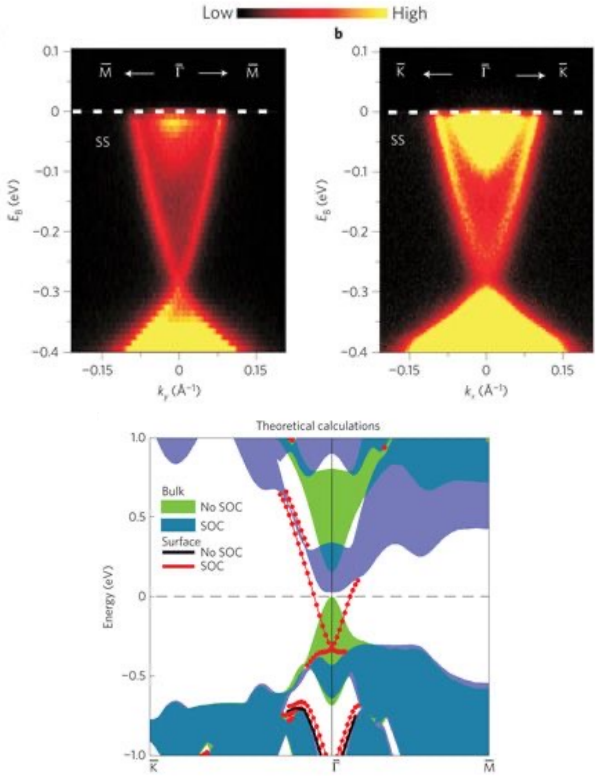
Not all doom and gloom…

MoS_2



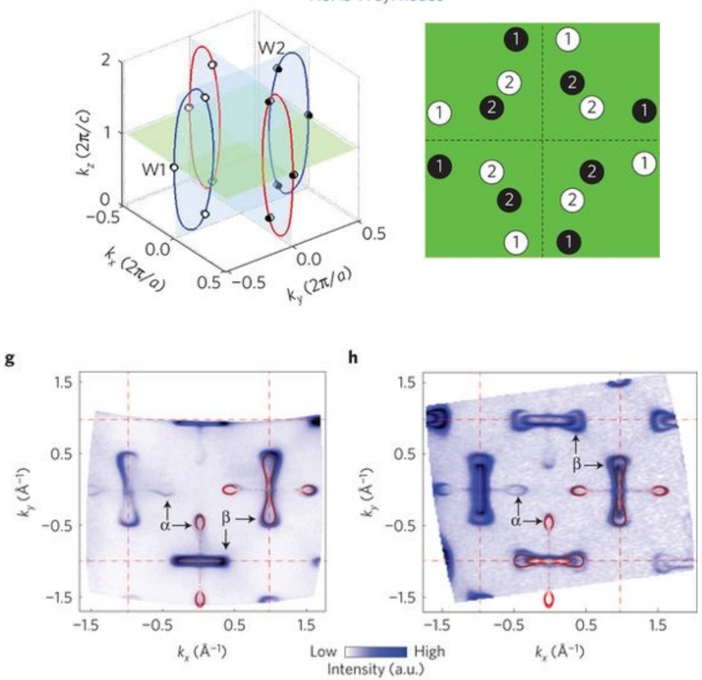
D.W. Latzke et al.
Phys Rev. B 91, 235202 (2015)

Bi_2Se_3



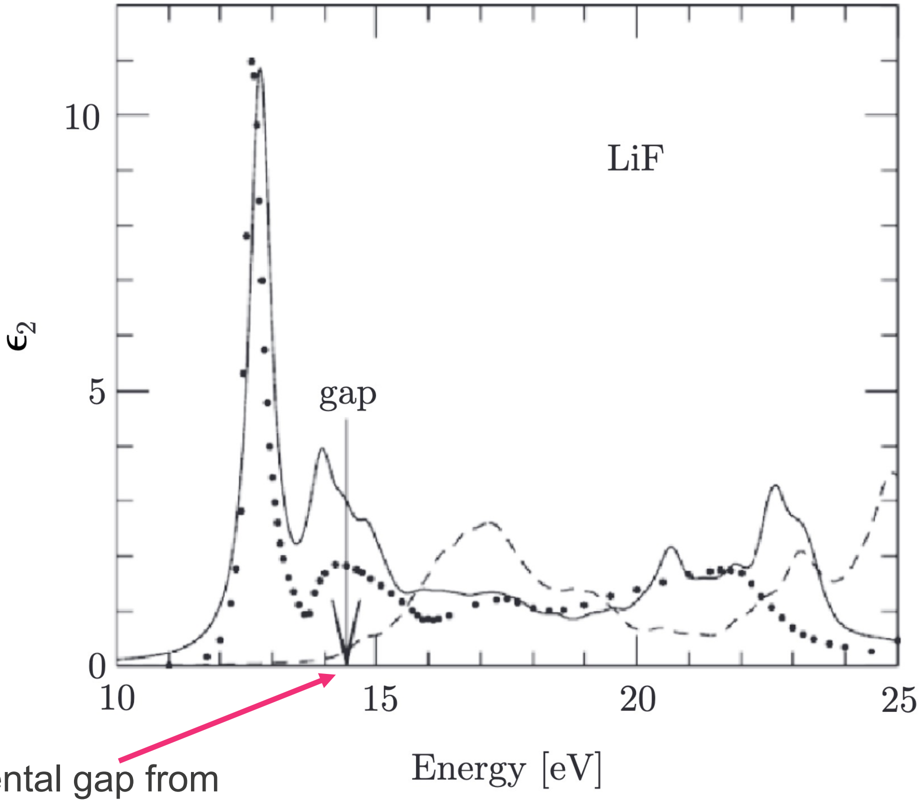
Y. Xia et al.
Nature Physics 5, 398–402 (2009)

NbAs



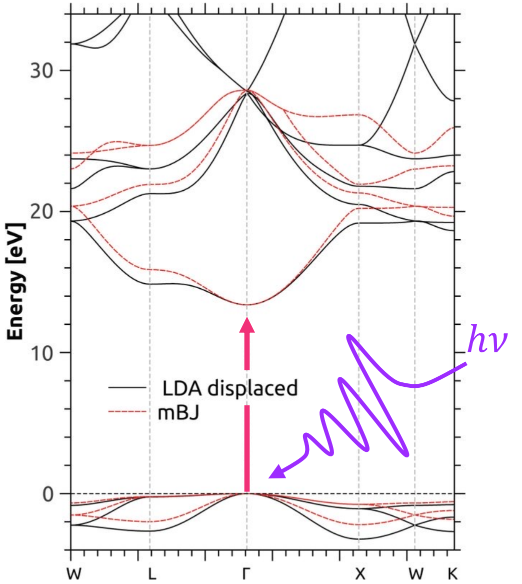
Z.K. Liu et al. Nature Materials 15, 27–31 (2016)
Su-Yang Xu et al. Nature Physics 11, 748–754 (2015)

Signatures of Correlations: Particle-Hole/Collective Excitations



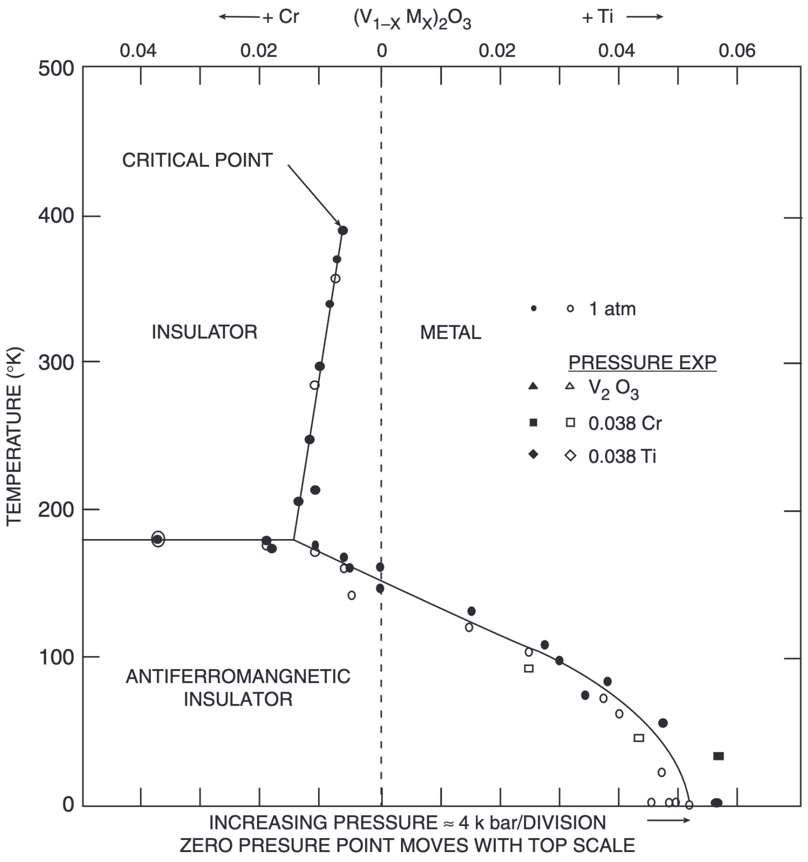
Fundamental gap from photoemission and inverse photoemission

D. M. Roessler and W. C. Walker, J. Opt. Soc. Am. 57:835–836, 1967.



J. A. Camargo-Martínez and R. Baquero Phys. Rev. B 86, 195106 (2012)

Signatures of Correlations: Metal-Insulator Transitions



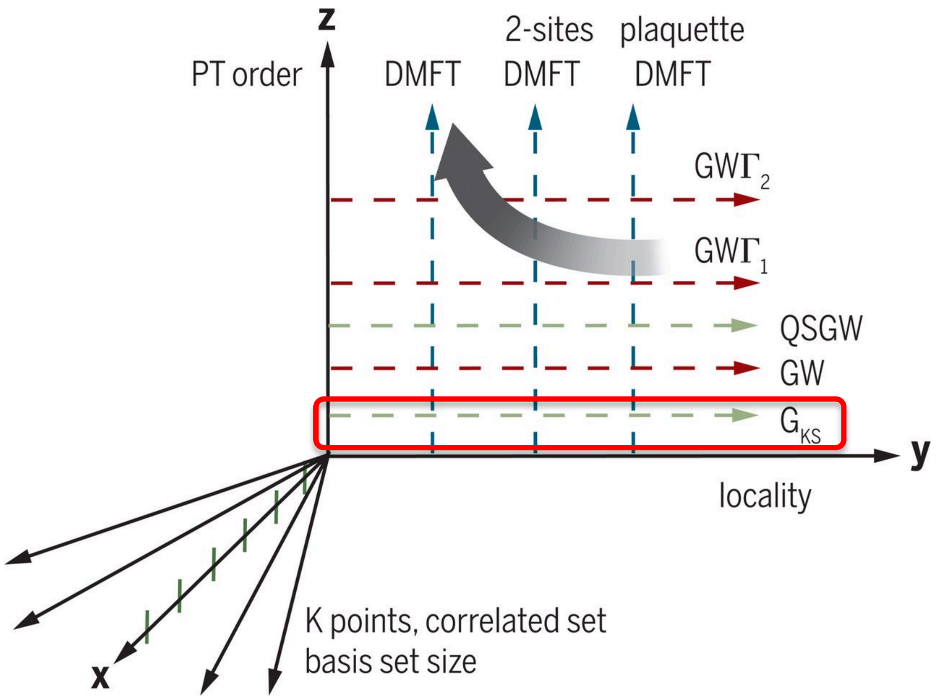
Metal-Insulator Transitions can arise from:

- Mott transition: (The most common transition) Arising from intense electron-electron correlation.
- Peierls transition: Electron-phonon interactions can give rise to a transition.
- Anderson transition: When an insulator behavior in metals arises from distortions and lattice defects.

D. B. McWhan, et al. Phys. Rev. B 7, 1920 (1973)

Paths forward?

Ab initio Methods



P. Kent and G. Kotliar. Science 361.6400, 348-354 (2018)

Low Energy Models

- Exact diagonalization method
  - Exact
  - Limited to finite sizes
- Quantum Monte Carlo method
  - Non-perturbative and thermodynamic limit
  - Negative sign problem for fermions
- Density matrix renormalization group (DMRG) theory
  - Capturing both quantum temporal and spatial fluctuations
  - Limited to one dimensional (1D) or quasi-1D systems