Los Alamos NATIONAL LABORATORY

When Independent Particle Methods Fail: The Effect of Correlations

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National Nuclear Security Administration

THEORETICAL DIVISION Since 1943

Approaches to the Many-Electron Problem

Many-body Hamiltonian

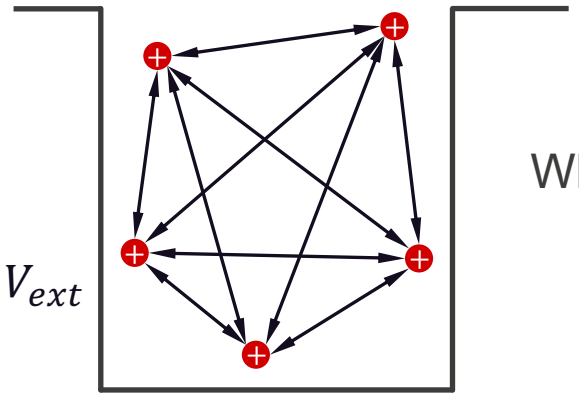
$$\hat{H} = \sum_{\substack{\alpha\beta \\ \alpha\beta\gamma\delta \in \text{spin}}} \int d^3r\, \hat{\psi}_\alpha^\dagger(r)\left(\frac{-\hbar^2\nabla^2}{2m} + v_{ext}^{\alpha\beta}(r)\right)\hat{\psi}_\beta(r) + \sum_{\alpha\beta\gamma\delta} \iint d^3r\, d^3r'\, \hat{\psi}_\alpha^\dagger(r)\hat{\psi}_\beta^\dagger(r')v_{\alpha\beta}^{\delta\gamma}(r,r')\hat{\psi}_\gamma(r')\hat{\psi}_\delta(r)$$

Electronic Kinetic Energy

Interactions

Wavefunction based approaches

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

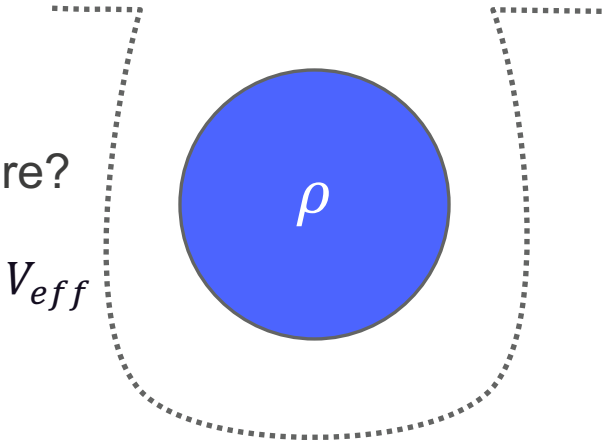


Observables: $\langle\Psi|\hat{O}|\Psi\rangle$

What do we fail to capture?
What is missing?

Reduced quantity based approaches

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

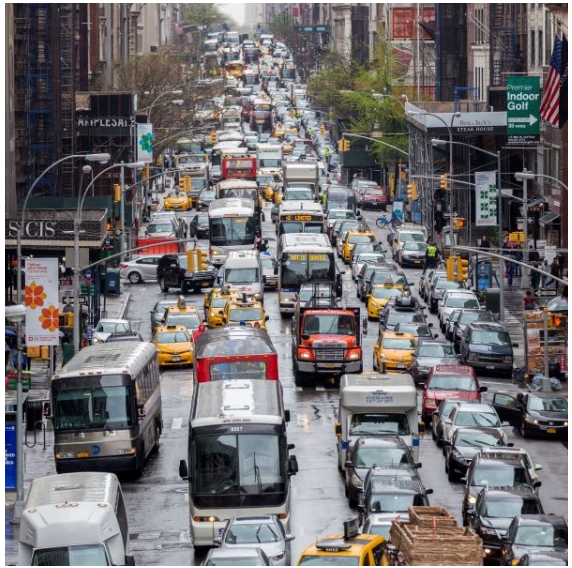


Observables: $\mathrm{F}[\rho]$

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

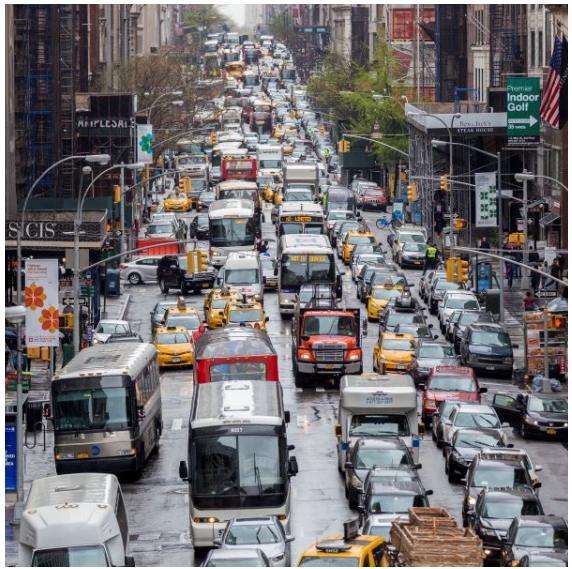
$$C_{AB} = \langle AB \rangle - \langle A \rangle \langle B \rangle$$

Spatial

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

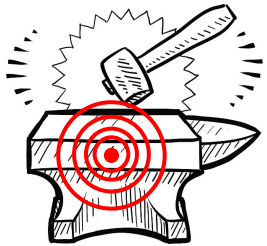
Temporal

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



Strong Correlations

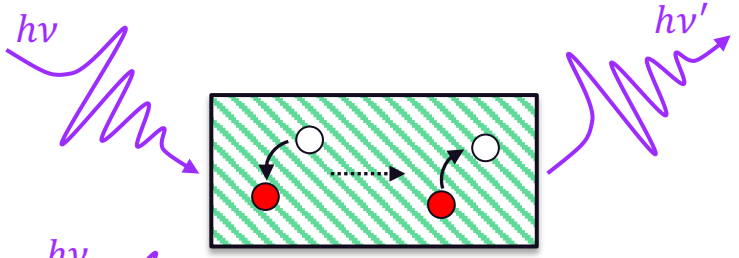
How Do We Measure Correlations?...



…Hit it with a hammer and listen

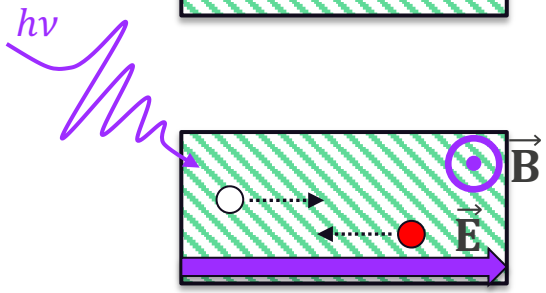
$\langle n(rt)n(r't')\rangle$

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



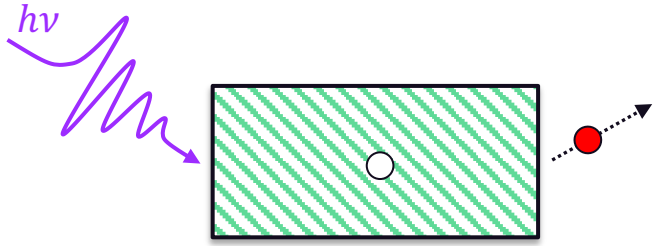
Pair Correlation Function for Electrons and Spin

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



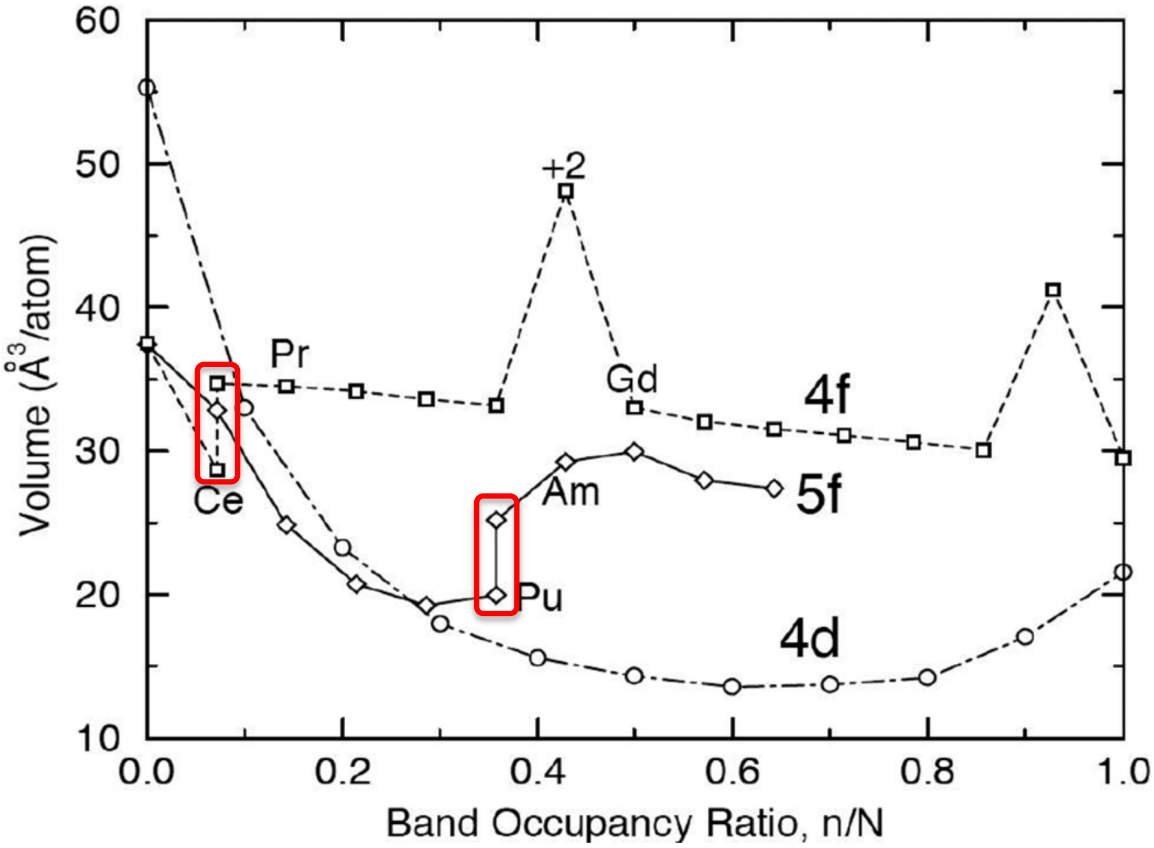
Electrictrical and Optical Properties

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

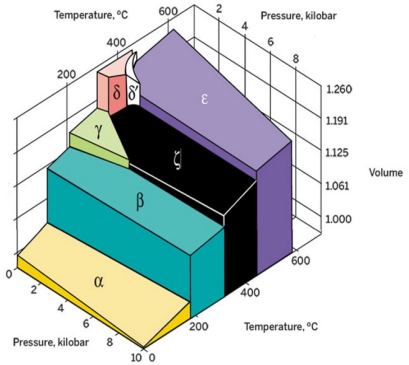


Removing (Adding) Electrons

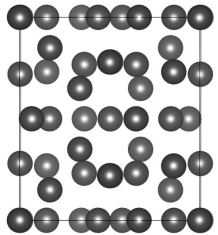
Signatures of Correlations: Ground State / Thermodynamic



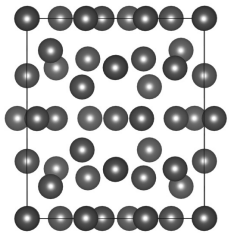
Delocalized → Localized Behavior



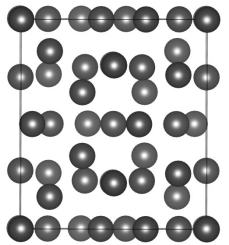
Experimental



VASP Relaxation

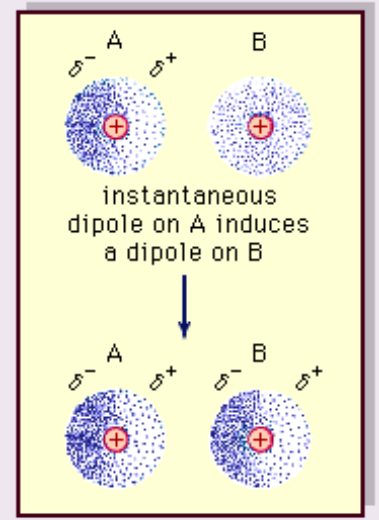


WIEN2k Relaxation

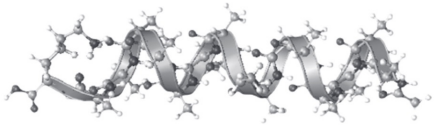


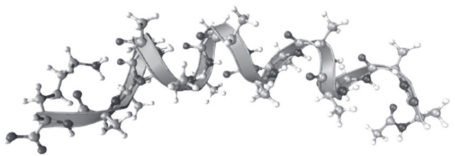
*Los Alamos Sci.***2000,***26* (II), 312
Project# 20210795DI - PI: Hernandez.

Signatures of Correlations: Ground State / Thermodynamic



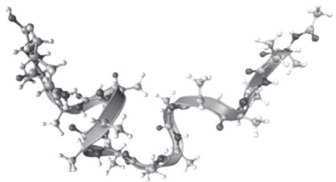
0 ps





30 ps, including vdW

α –helix structure in agreement with experiment

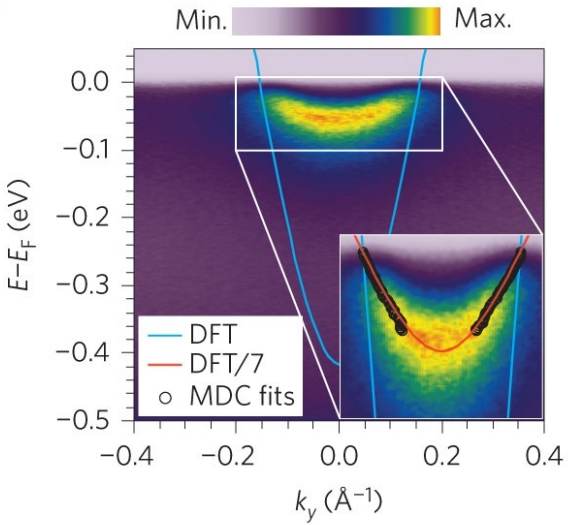


30 ps, no vdW

An open structure

Signatures of Correlations: Electron Addition/Removal

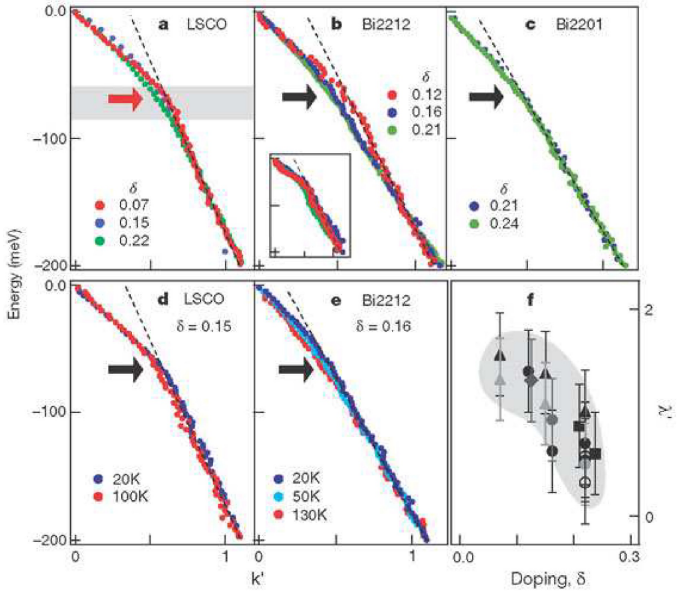
$LiNiO_3$



Renormalization

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

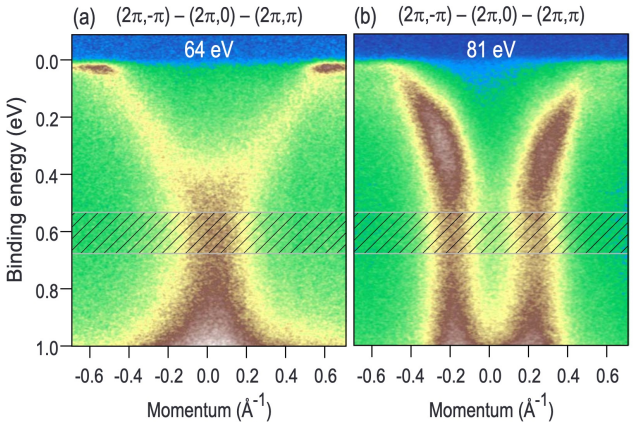
Various Cuprates



Kinks

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

$Bi_2Sr_2CaCu_2O_8$



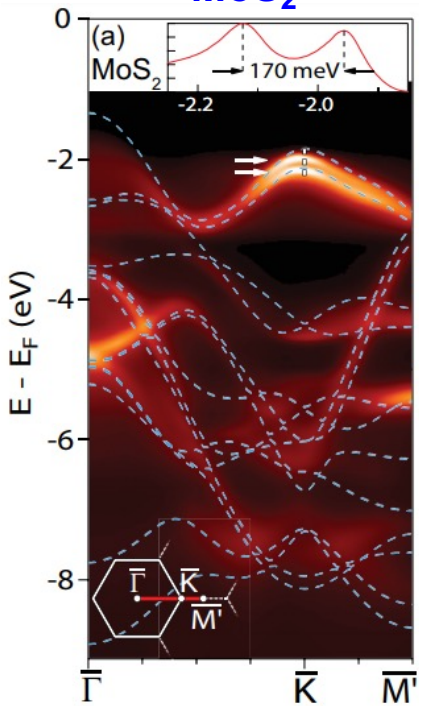
Waterfalls

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

Signatures of Correlations: Electron Addition/Removal

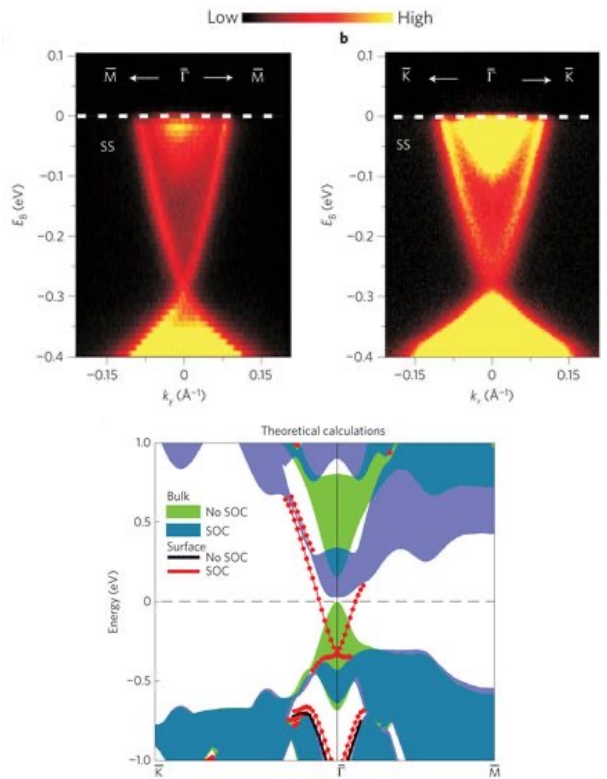
Not all doom and gloom…

MoS_2



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

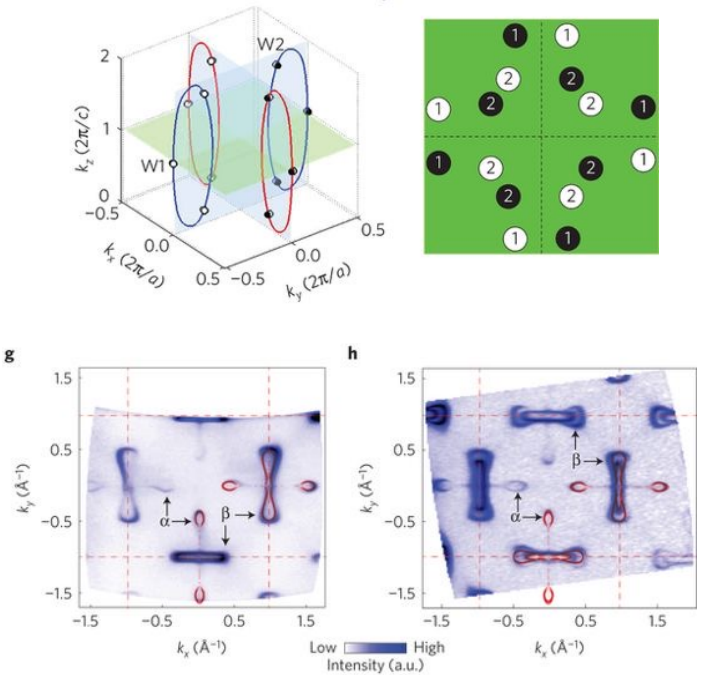
Bi_2Se_3



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

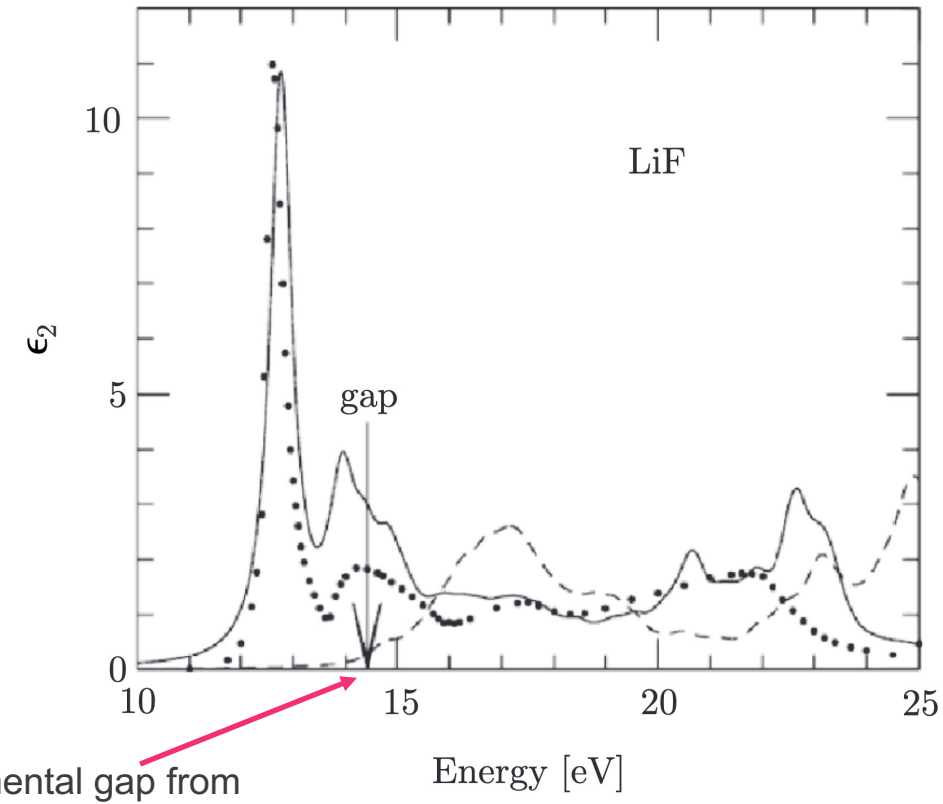
NbAs

NbAs Weyl nodes



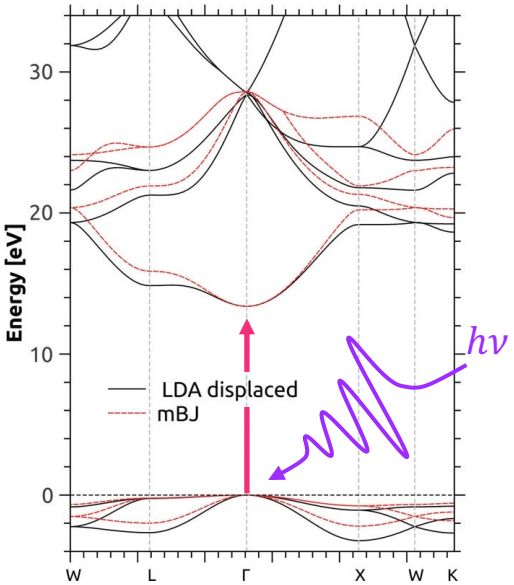
Z.K. Liu et al. Nature Materials 15, 27–31 (2016)
Su-Yang Xu et al. Nature Physics 11, 748–754 (2015)

Signatures of Correlations: Particle-Hole/Collective Excitations



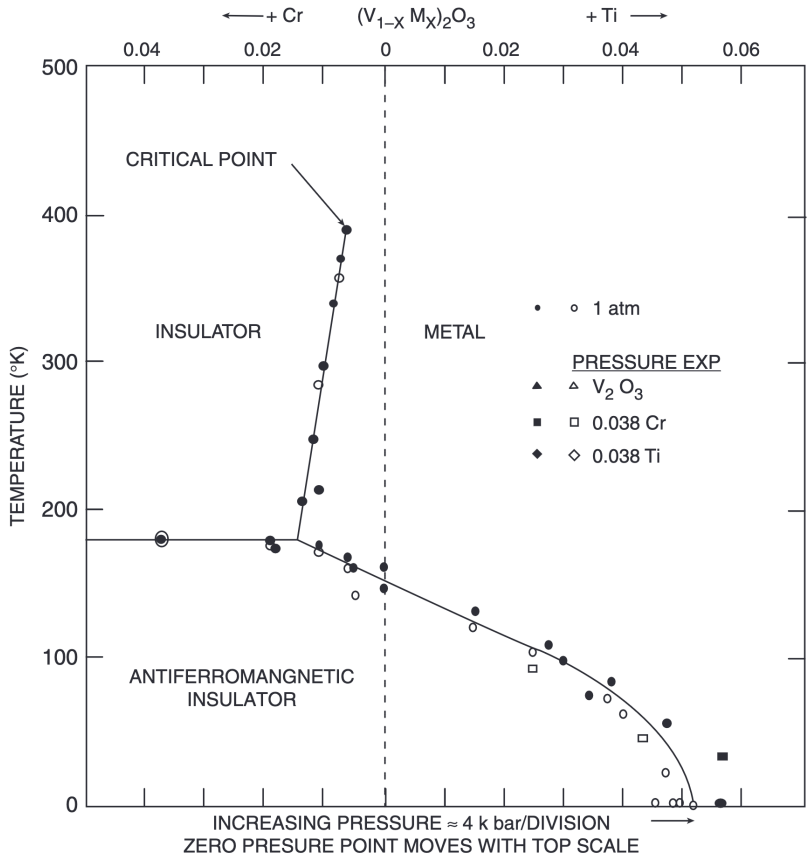
Fundamental gap from photoemission and inverse photoemission

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



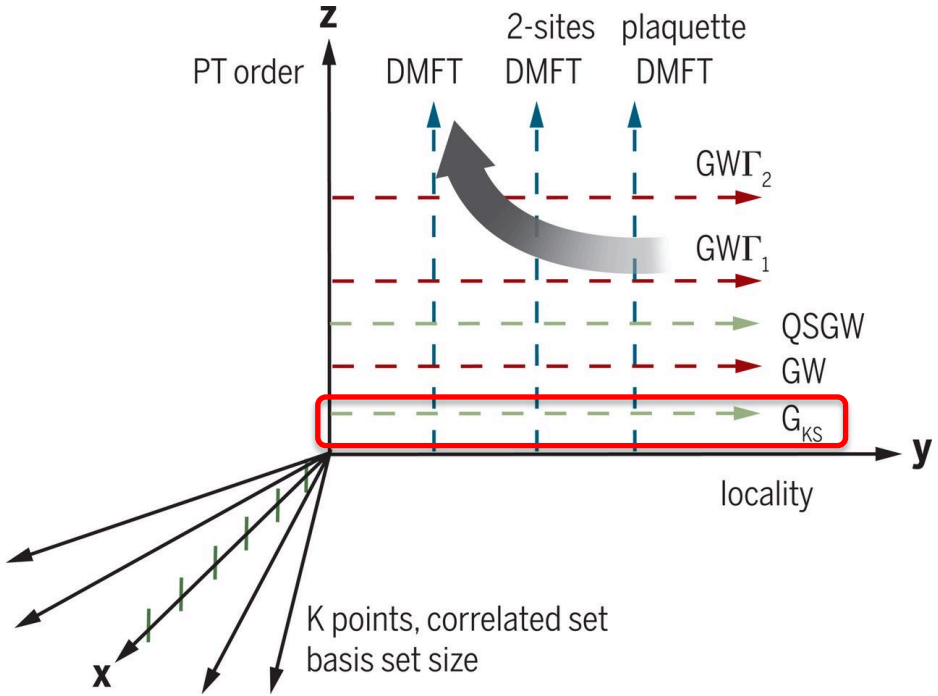
Metal-Insulator Transitions can arise from:

- 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