Correlated Electrons: Why we need Models to Understand Real Materials?

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In collaboration with

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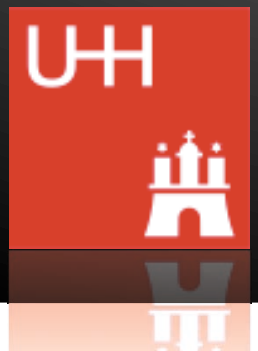
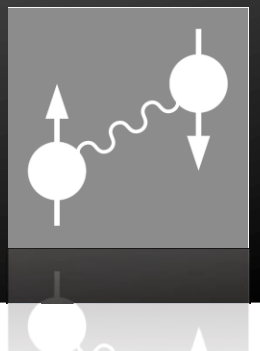


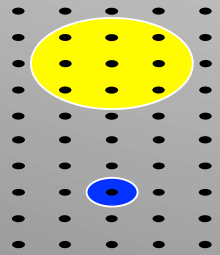


Outline

1. Introduction
2. Functional approach: Route to fluctuations
3. Local correlations and beyond
4. Solving quantum impurity problems
5. From models to real materials
6. Outlook

Strongly Correlated Electron Systems

3d - 4f
open shells
materials



U<<W
Charge fluct.

U>>W
Spin fluct.

- Kondo
- Mott-Hubbard
- Heavy Fermions
- **High-Tc SC**
- Spin-charge order
- **Colossal MR**

I	II	IIIb	IVb	Vb	VIb	VIIb	VIIIb		Ib	IIb	III	IV	V	VI	VII	0
H																He
Li	Be										B	C	N	O	F	Ne
Na	Mg										Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	La*	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra	Ac**	Rf	Db	Sg	Bh	Hs	Mt									

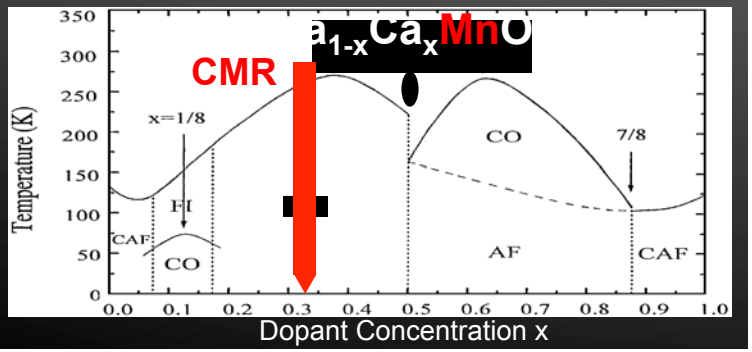
Lanthanides *	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
Actinides **	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr

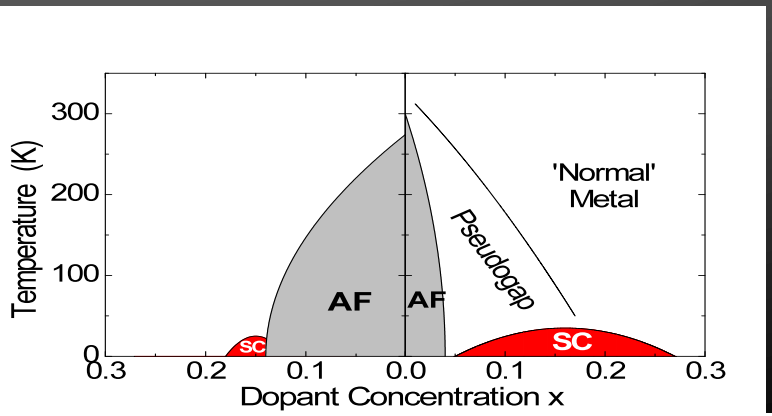
Control parameters

- **Bandwidth (U/W)**
- **Band filling**
- **Dimensionality**

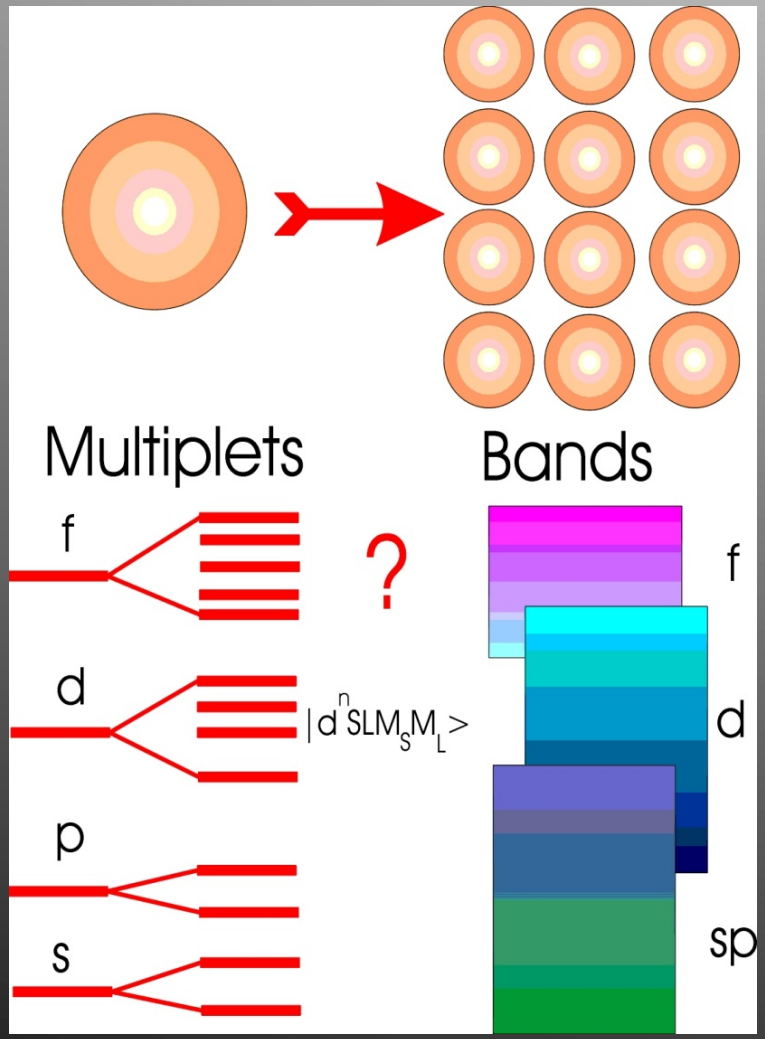
Degrees of freedom

- **Charge / Spin**
- **Orbital**





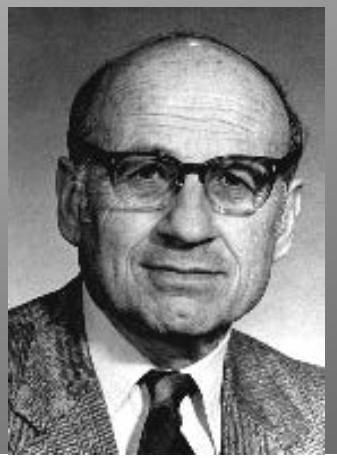
From Atom to Solids

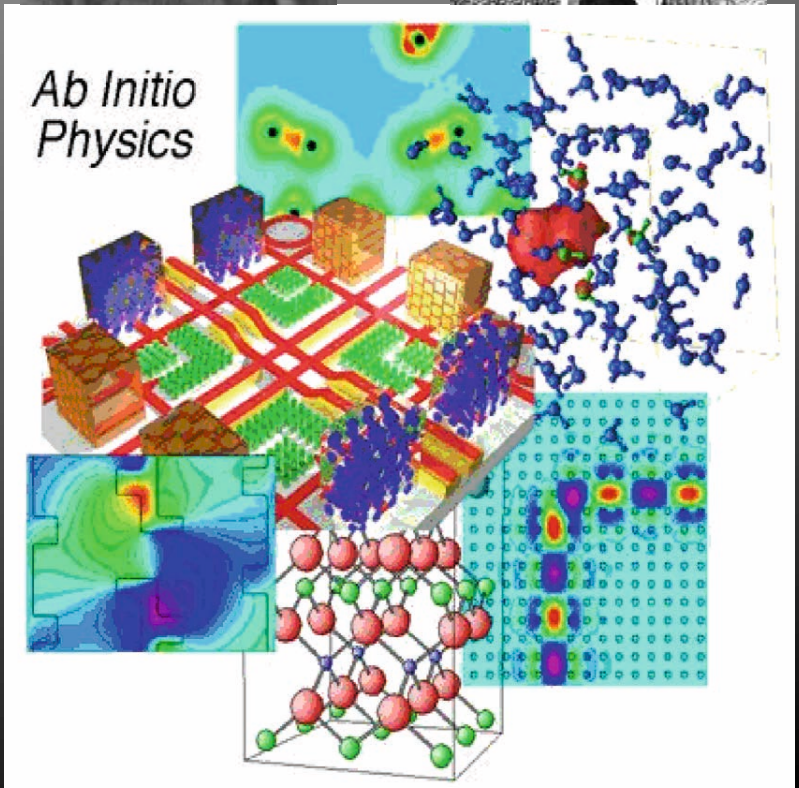


- How to incorporate atomic physics in the band structure ?
- How good is a local approximation ?
- What is a best solution for atomic problem in effective medium ?
- What is different from one band Hubbard model?
- How to solve a complicated Quantum multiorbital problem ?
- What is the best Tight-Binding scheme for realistic Many-Body calculation for solids?

Computational Material Science

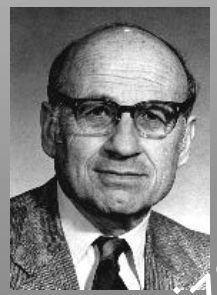
- Starting from Schrödinger?
- Kohn Density Functional Theory (DFT) of inhomogeneous electron gas in solids
- Strongly correlated electron systems ? Dynamical Mean-Field Theory (DMFT)





Quantum Theory & Electronic structure



1900 Planck
'26 Schrodinger
'28 Dirac
'37 APW
'59 Pseudopotential
'64 DFT
'75 LMTO, LAPW
'85 Car-Parrinello
1998 Nobel Prize Kohn

1900
2010

'27 Fermi
'30 Hartree
'51 Slater
'72 LSDA
'86 GGA
'91 LDA+U
'92 DMFT



DFT: KS-equation (1965)

Effective one-electron Schrödinger-like equation:

$$(-\frac{\hbar^2}{2m}\nabla^2 - V_{eff}(\vec{r}))\psi_i(\vec{r}) = \varepsilon_i\psi_i(\vec{r})$$

Charge density:

$$n(\vec{r}) = \sum_i^N |\psi_i(\vec{r})|^2$$

Energy Functional:

$$E[n] = T_s[n] + V_H[n] + \int n(\vec{r})V_{ext}(\vec{r})d\vec{r} + E_{xc}[n]$$

KS-kinetic energy:

$$T_s[n] = \sum_i^N \int d\vec{r}\psi_i^*(\vec{r})(-\frac{\hbar^2}{2m}\nabla^2)\psi_i(\vec{r})$$

Hartree potential:

$$V_H[n] = \frac{e^2}{2}\int d\vec{r}\int d\vec{r}'\frac{n(\vec{r})n(\vec{r}')}{|\vec{r}-\vec{r}'|}$$

Effective potential:

$$V_{eff}(\vec{r}) = V_{ext}(\vec{r}) + e^2\int d\vec{r}'\frac{n(\vec{r}')}{|\vec{r}-\vec{r}'|} + \frac{\delta E_{xc}[n]}{\delta n(\vec{r})}$$

How well performs DFT?

- DFT successful approximations for extended states s, p, electrons…

  Why it is so good?

  - The Fermi Liquid Theory (1957-59):
    - Interactions are weak - quasiparticle
    - Interactions are slowly switched on
    - Energy levels are modified
    - Eigenstate is given by occupation numbers

$$E = \sum_{\sigma,k} n_\sigma(k)\varepsilon(k) + \frac{1}{2}\sum_{\sigma,\sigma',k,k'} f_{\sigma,\sigma'}(k,k')n_\sigma(k)n_{\sigma'}(k')$$

Lev Landau

- DFT – bad for correlated electrons…
  - Mott insulators, Heavy Fermions etc.

Where is a small parameter?

Why DFT-LDA works?

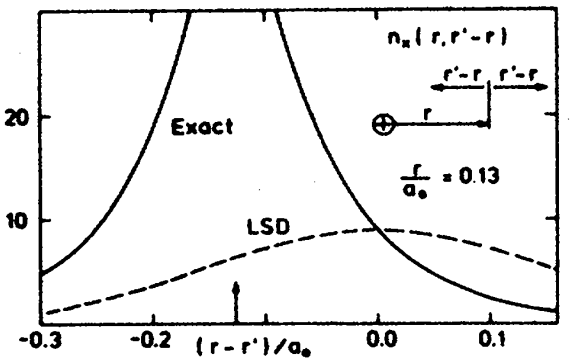
- Errors in the approximation of exchange and correlation cancel

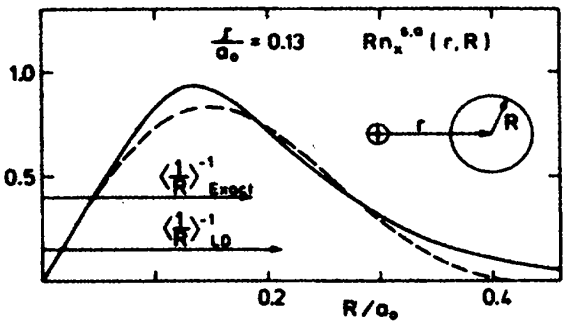
- LDA does fulfill the sum rule for the exchange-correlation hole

$$\int d\mathbf{r}' n_{xc}(\mathbf{r}, \mathbf{r}' - \mathbf{r}) = -1$$

$n_{xc}(\mathbf{r}, \mathbf{r}') = n(\mathbf{r}')[\tilde{g}(\mathbf{r}, \mathbf{r}') - 1]$ = exchange-correlation hole density;

$\tilde{g}(\mathbf{r}, \mathbf{r}')$ = pair correlation function averaged over coupling constant.





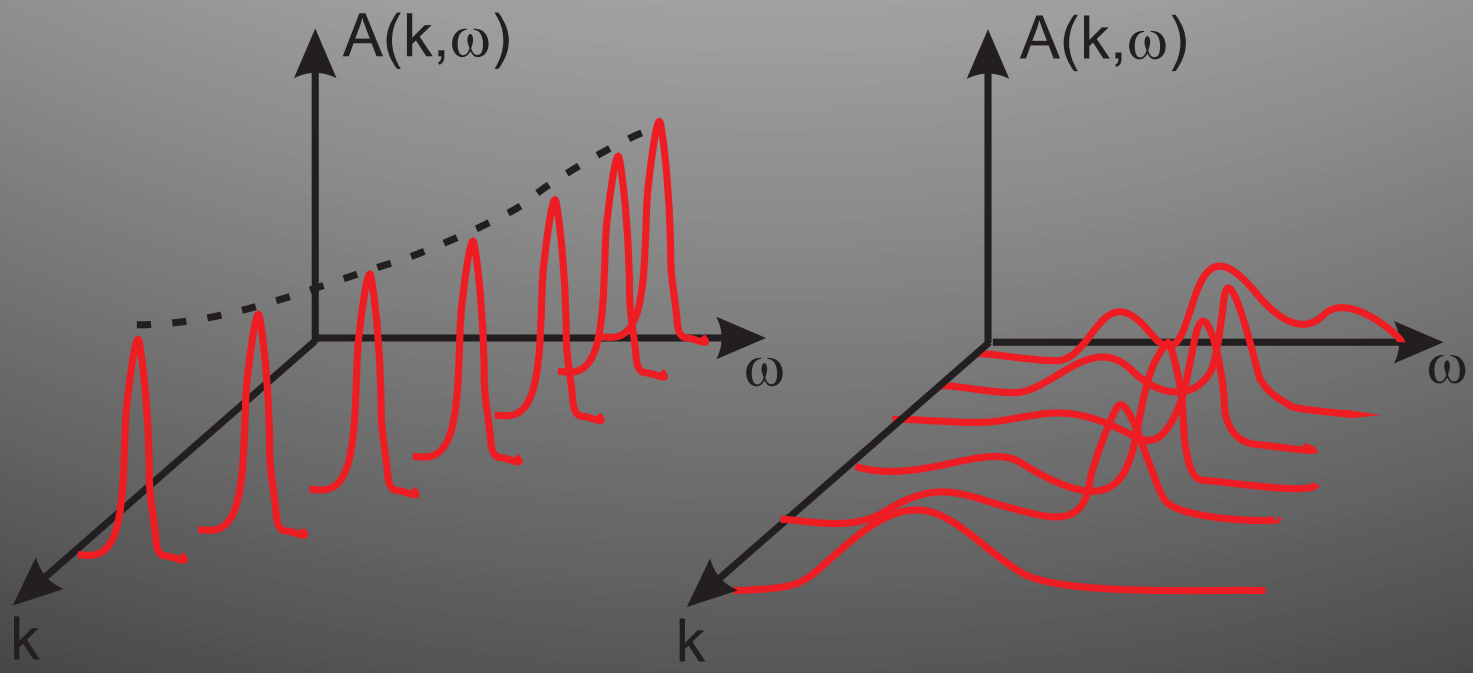
- The exchange-correlation energy depends only on the angle-averaged exchange-correlation hole which is well described in LDA.

$$2E_{xc}[n] = \int \frac{n(\mathbf{r}) n_{xc}(\mathbf{r}, \mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r} d\mathbf{r}' = \int n(\mathbf{r}) d\mathbf{r} \int \tilde{n}_{xc}(\mathbf{r}, R) dR/R,$$

$$\tilde{n}_{xc}(\mathbf{r}, R) = \int n_{xc}(\mathbf{r}, \mathbf{r} + \mathbf{R}) d\Omega_R / 4\pi.$$

R. O. Jones and O. Gunnarsson, Rev. Mod. Phys. 61, 689 (1989)

Correlated Electrons: ARPES

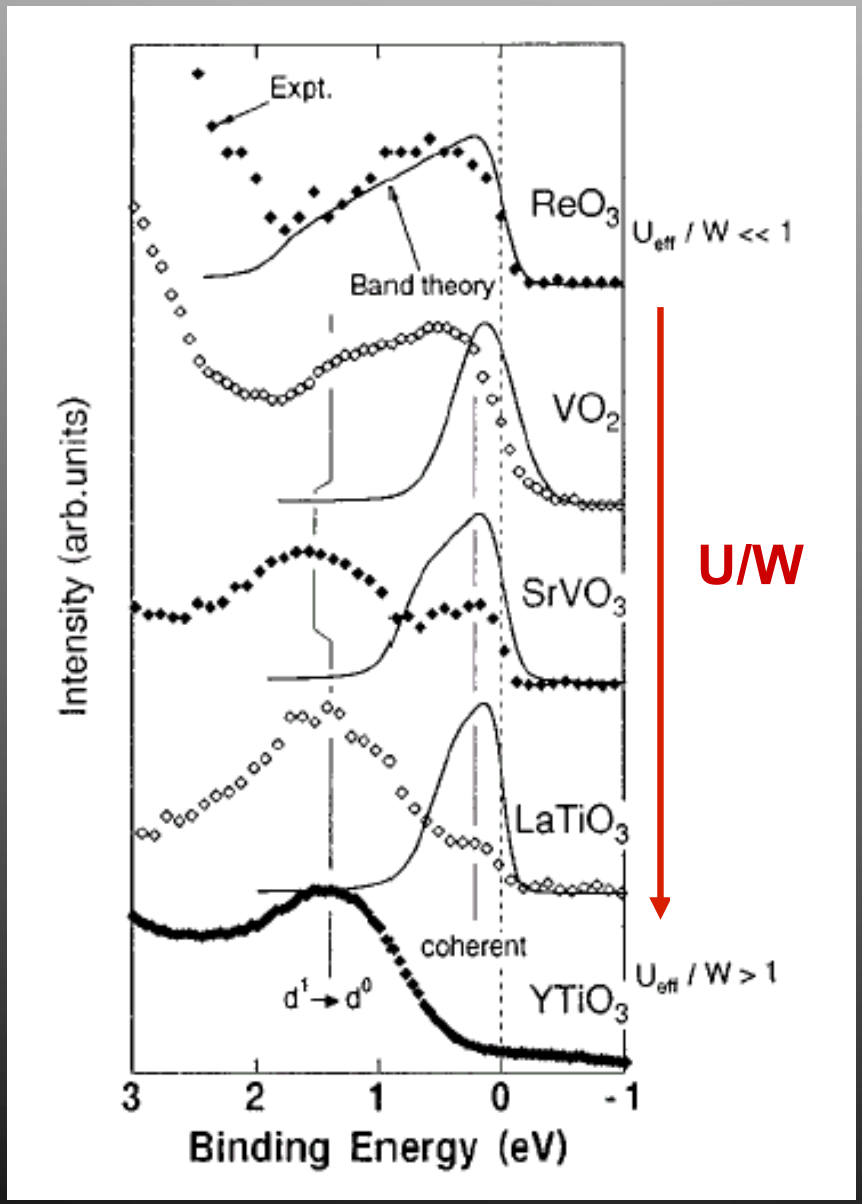


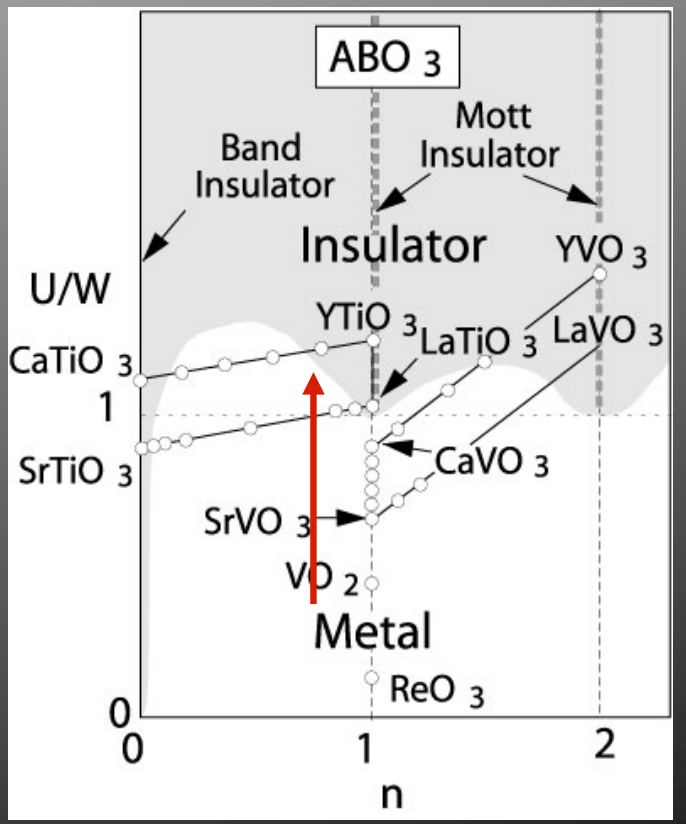
Weak corr.

Strong corr.

Correlation driven MIT

photoemission spectra (DOS)
A. Fujimori et al.

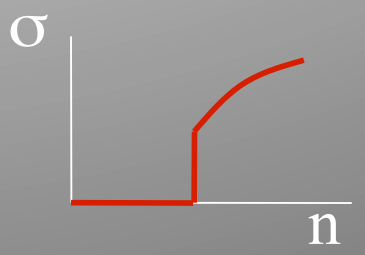




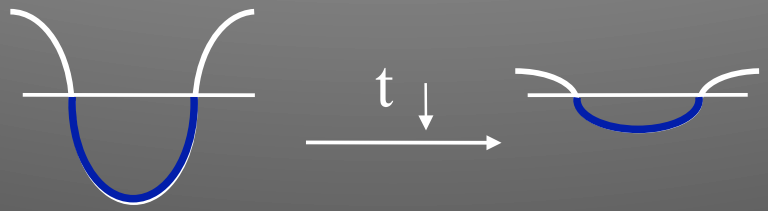
What is the Mott transition?

a correlation driven metal-insulator transition

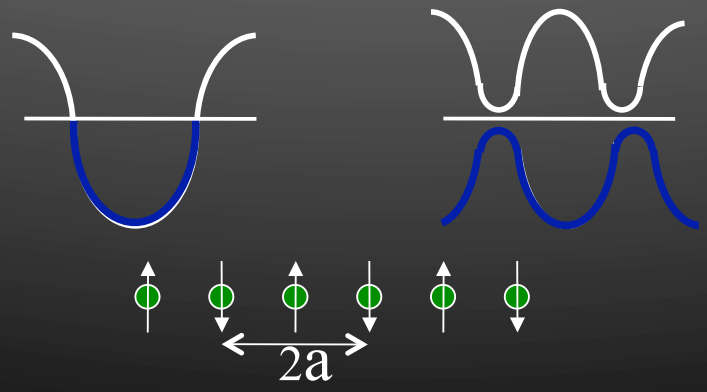
Mott '49

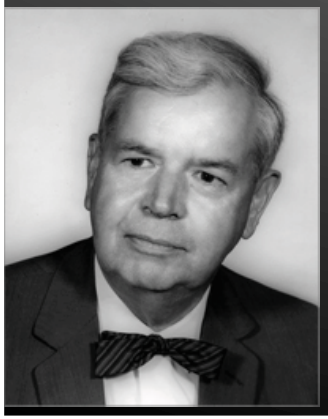


cannot be obtained in band theory:



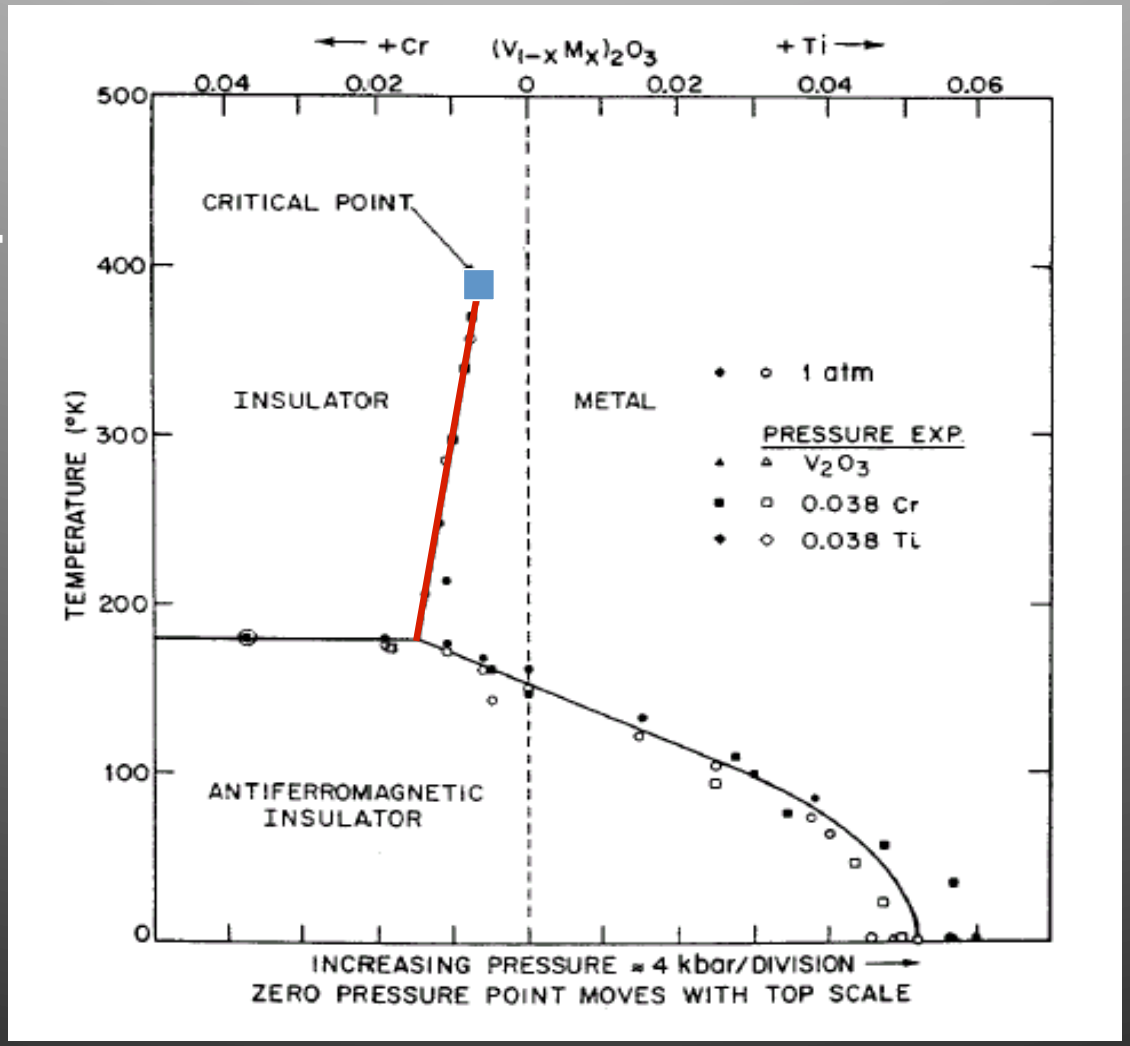
not due to Slater AF (weak coupling effect):





Mott transition in V_2O_3

T



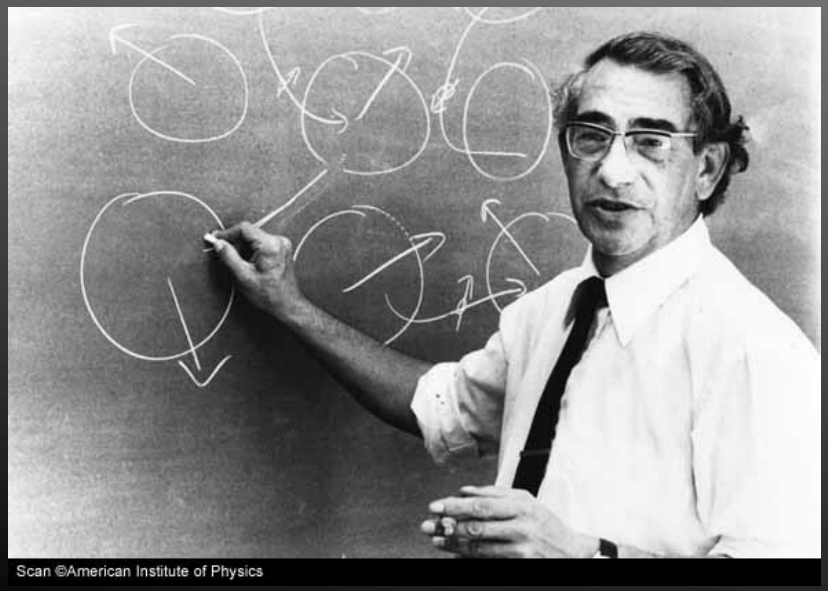
pressure or chemical substitution

McWhan et al

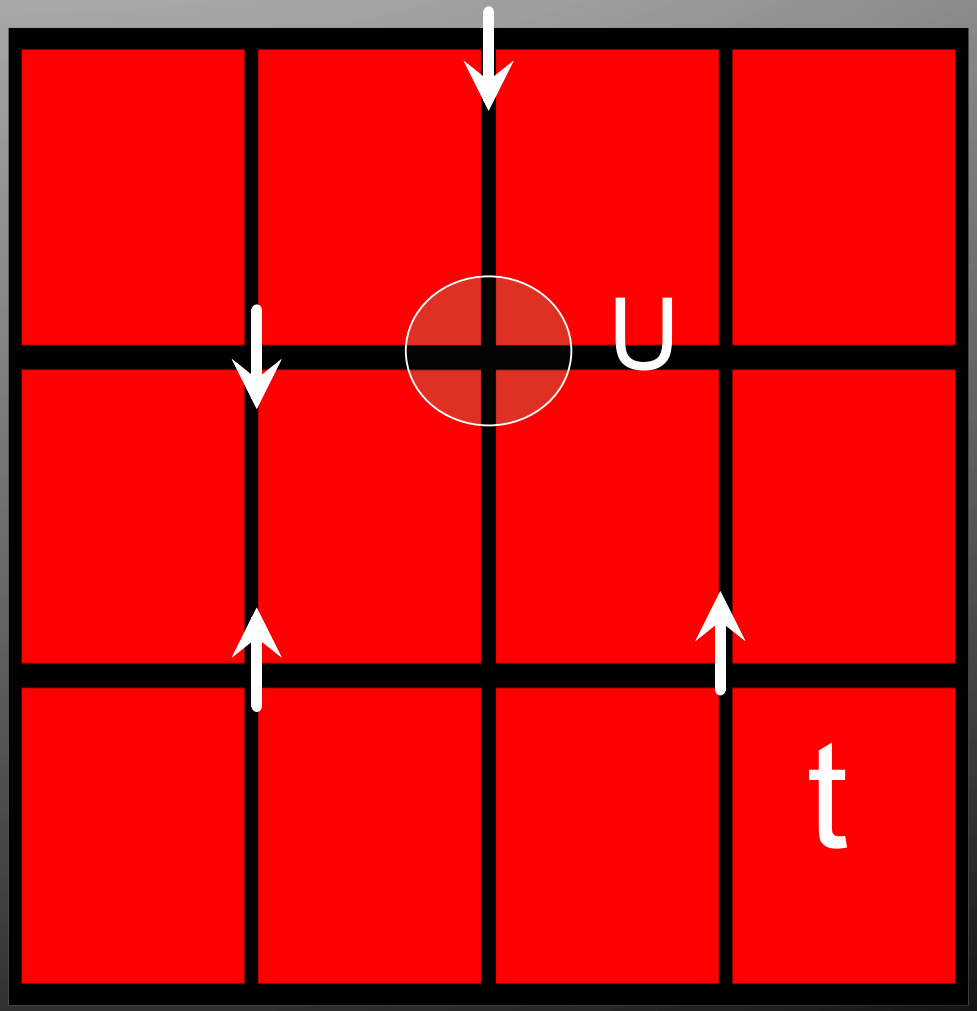
Hubbard model for correlated electrons

$$H = \sum_{ij} t_{ij} c^{+}_{i\sigma} c_{j\sigma} + U \sum_{i} n_{i\uparrow} n_{i\downarrow}$$

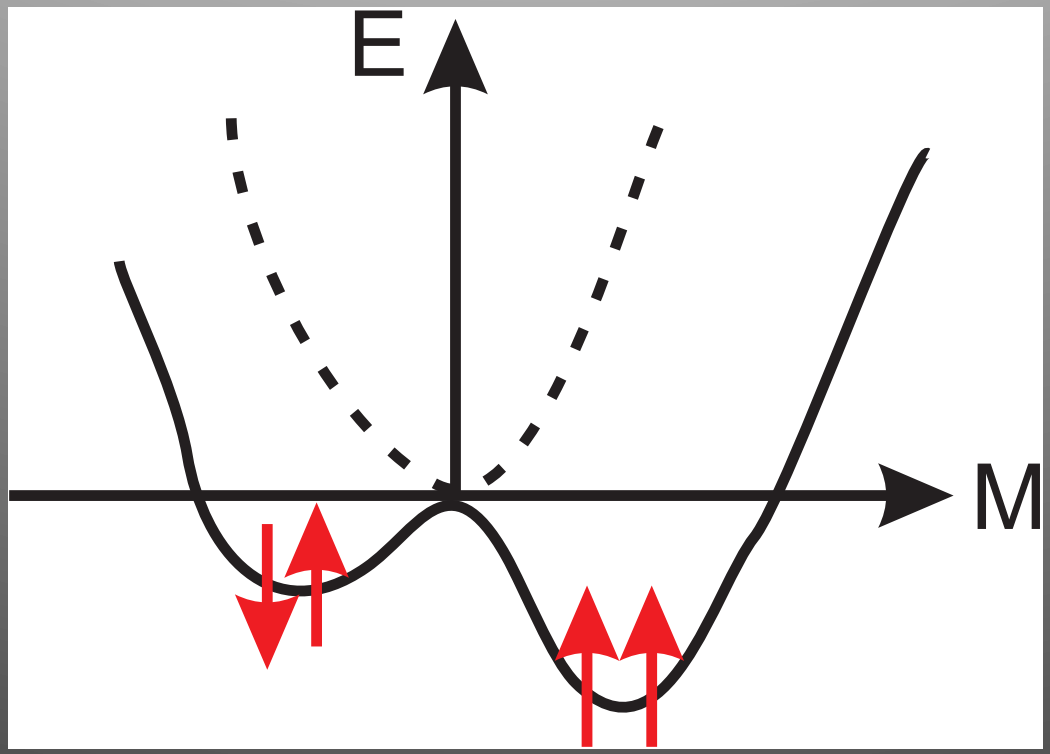
- U/t
- Chemical potential



Scan ©American Institute of Physics

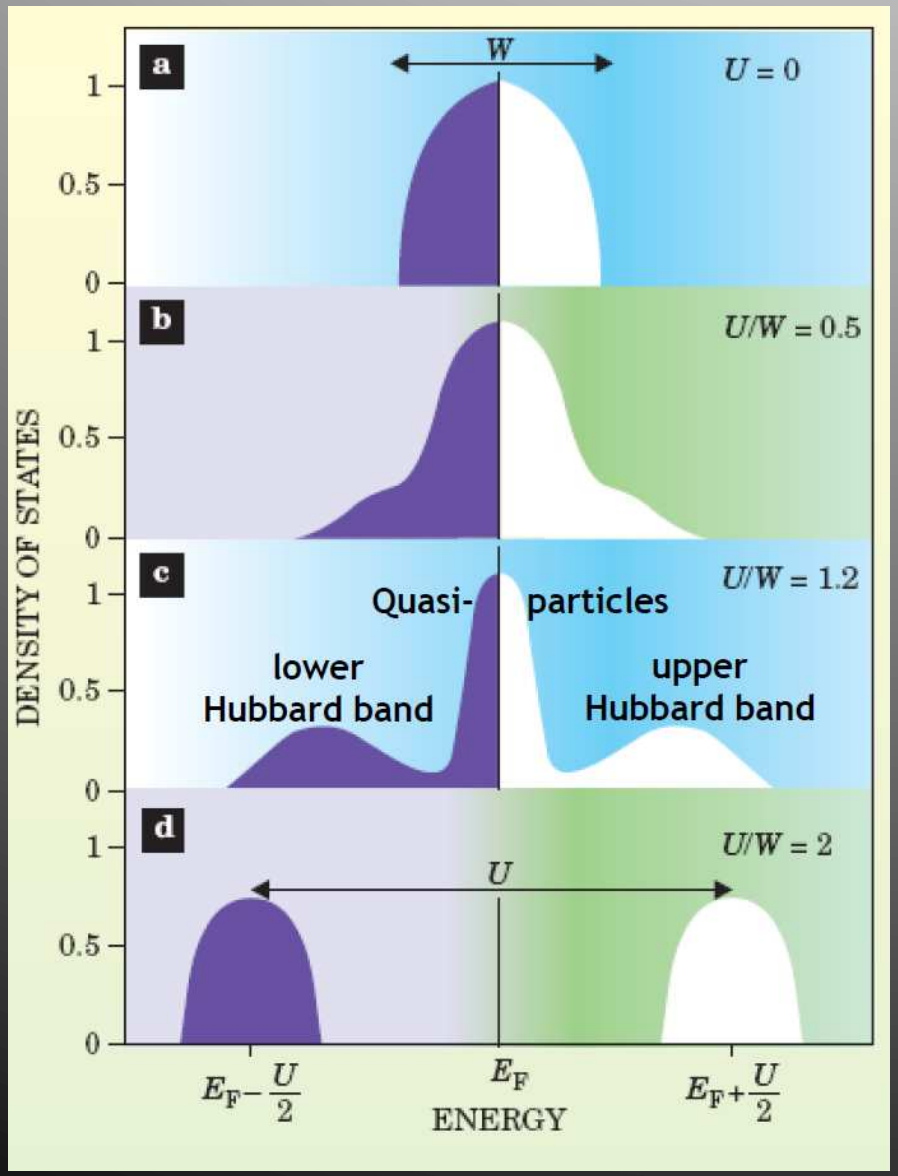


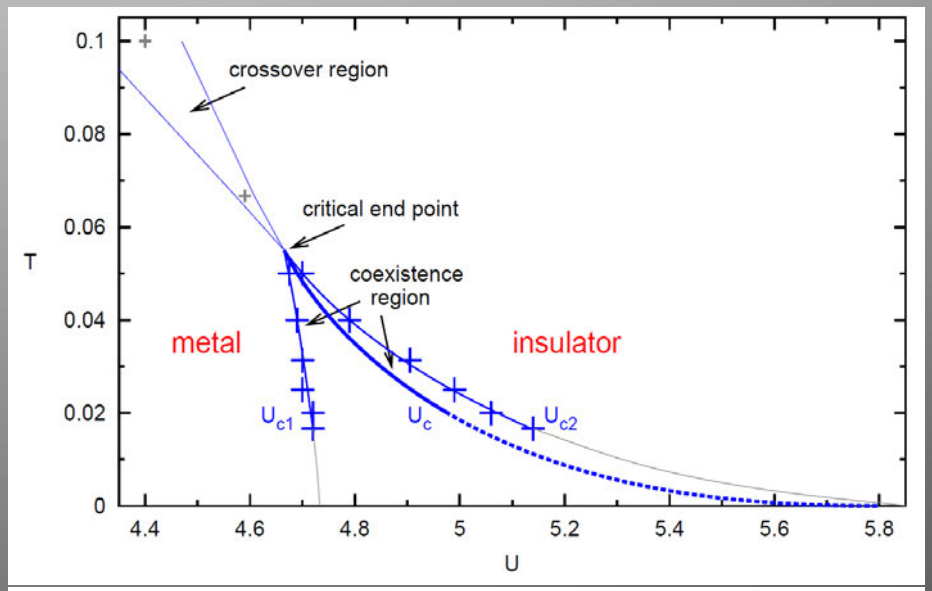
Correlated Electrons: Fluctuations

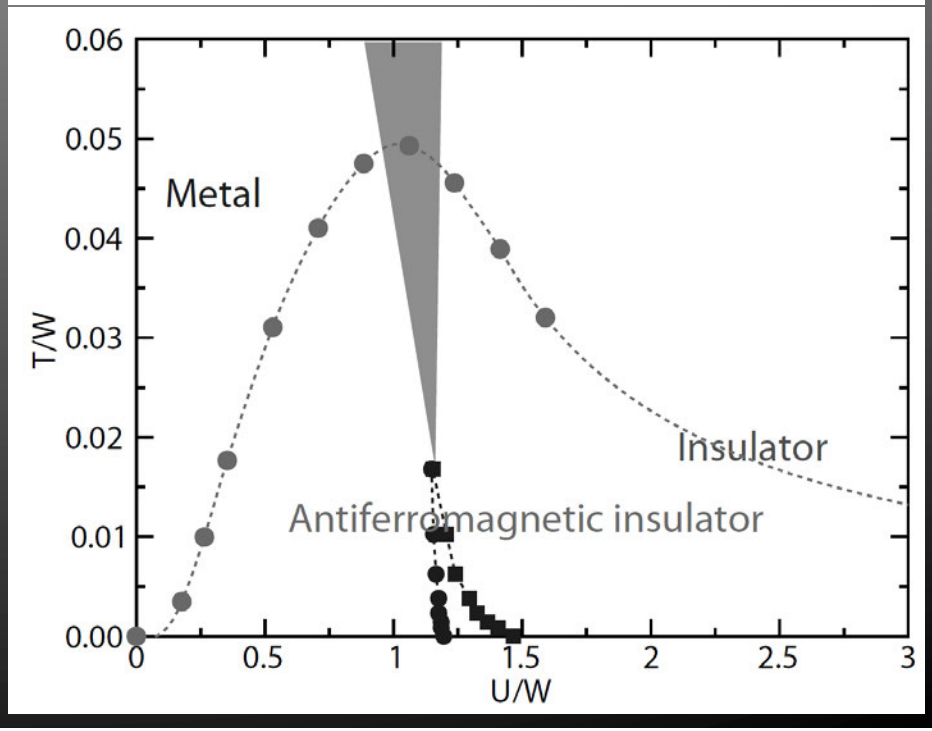


Fluctuation of charge, spin and orbital degrees of freedom related with complex behavior of correlated electronic systems

Metal-Insulator Transition





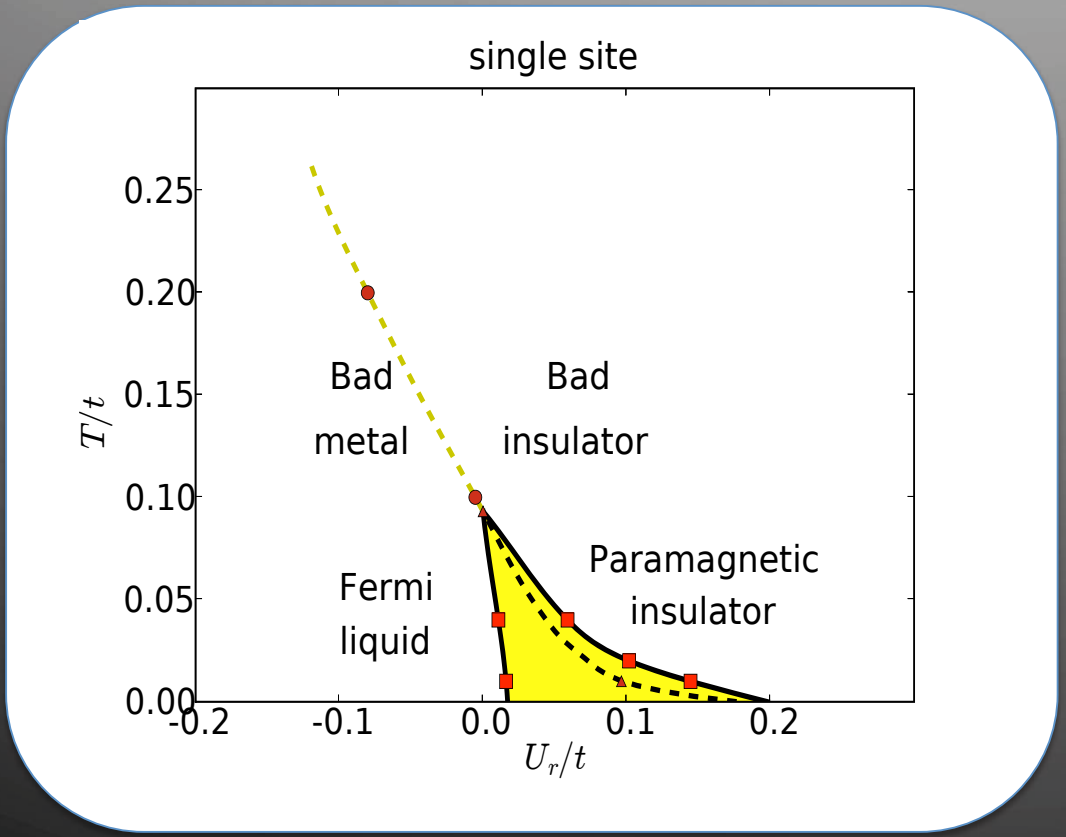


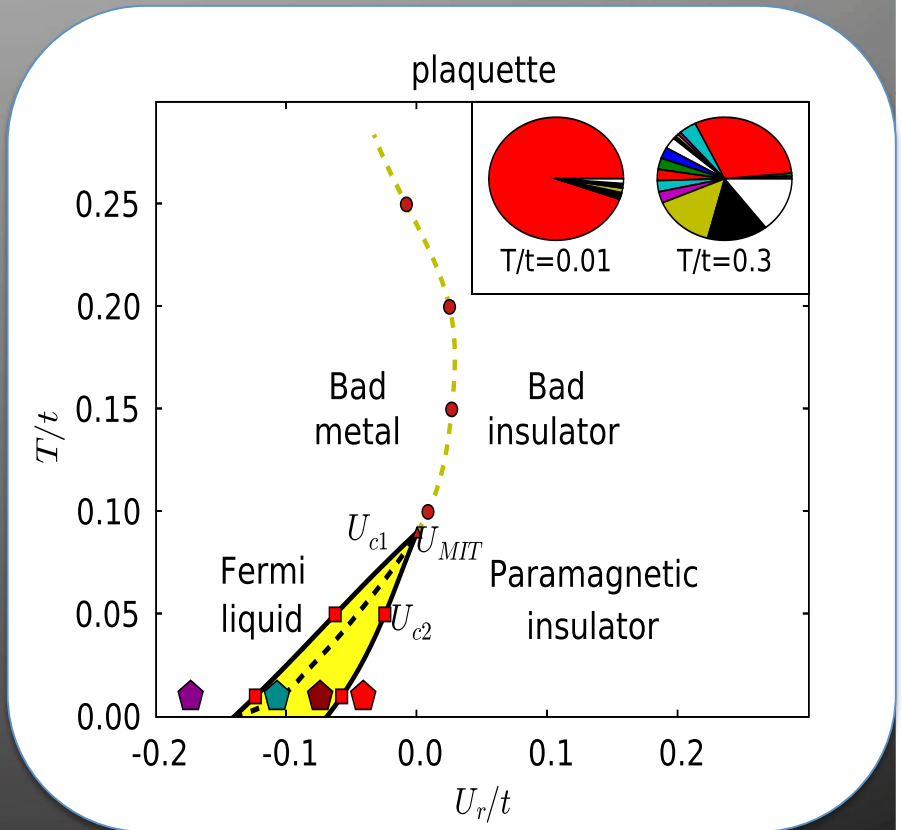
G. Kotliar and D. Vollhardt,
Physics Today **3,** 53 (2004)

Phase diagram of Hubbard model

Uc=9.35t

Uc=6.05t

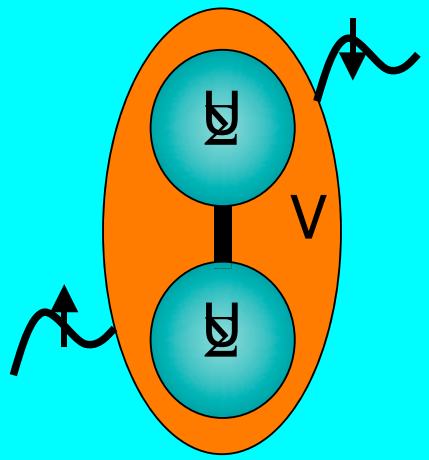




H. Park et al PRL (2008)
C-DMFT with CT-QMC

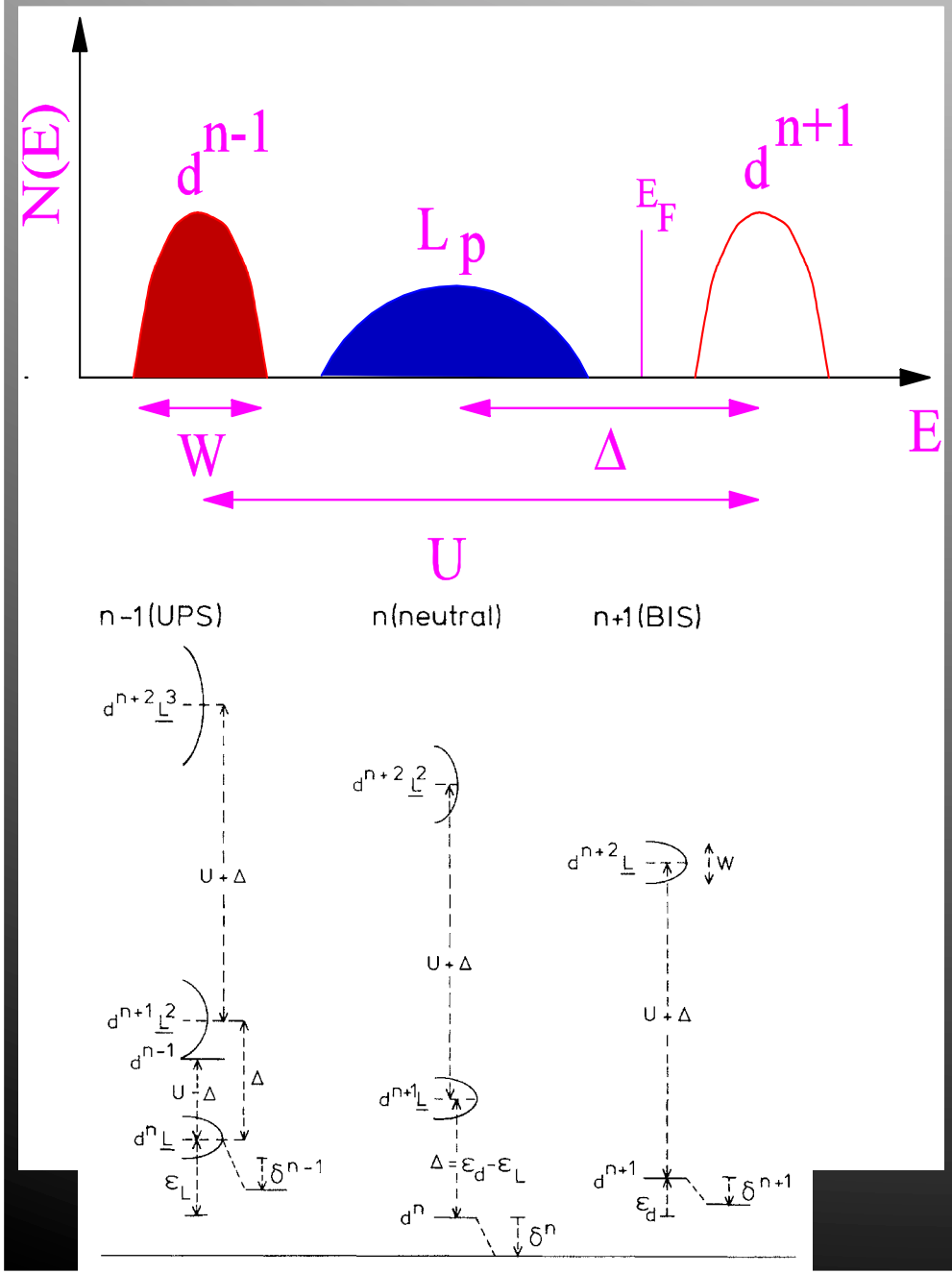
Cluster DMFT (DCA)

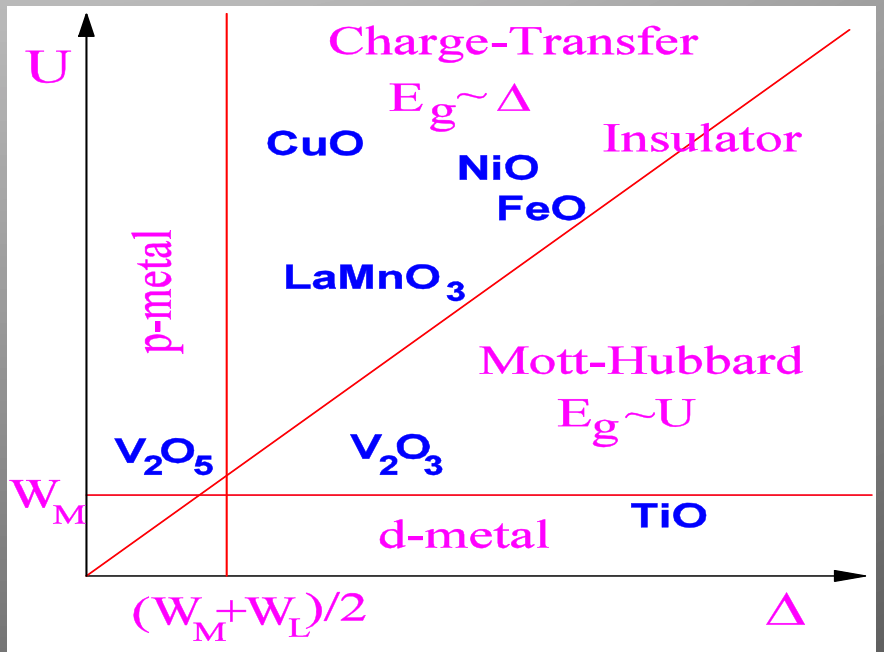
$$\mathcal{G}_0(\tau - \tau')$$



M. Hettler et al, PRB 58, 7475 (1998)
A. L. and M. Katsnelson, PRB 62, R9283 (2000)
G. Kotliar, et al, PRL 87, 186401 (2001)

Charge transfer TMO insulators



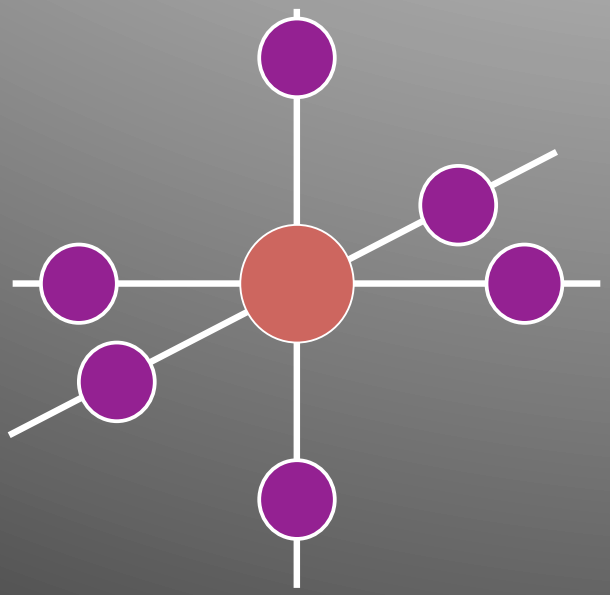


Zaanen-Sawatzky-Allen (ZSA) phase diagram

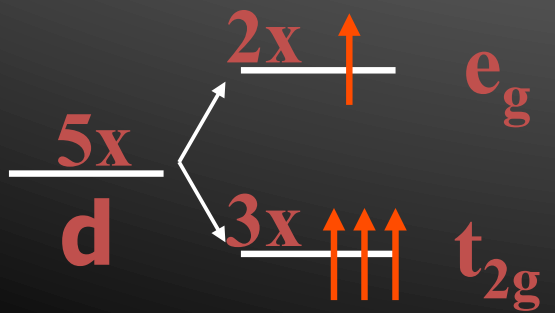
Phys. Rev. Lett. 55, 418 (1985)

Orbital degrees of freedom

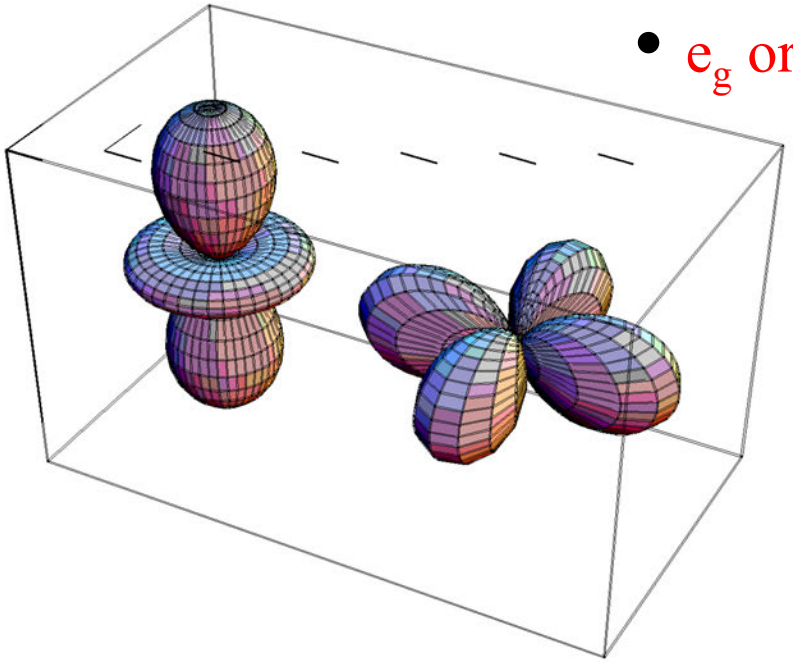
Mn (3+) = $3d^4$



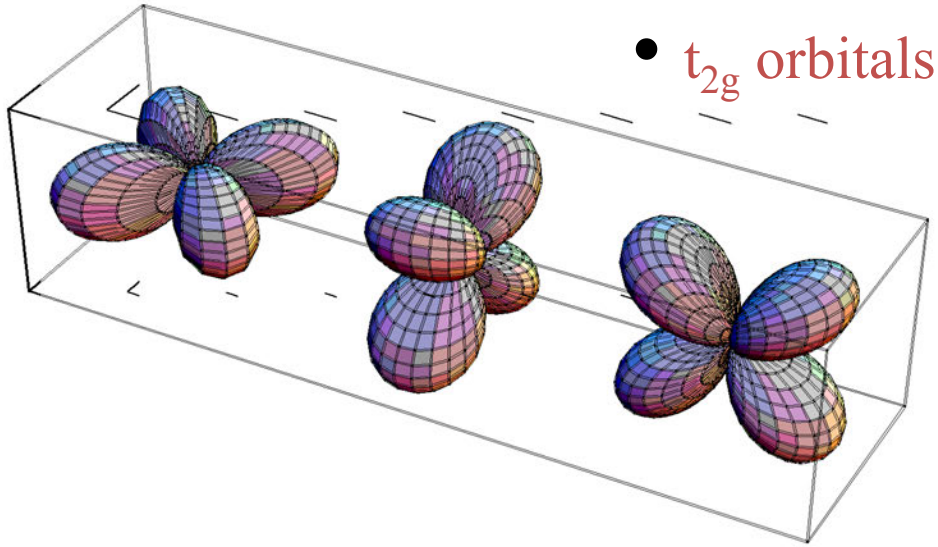
3d-ion in cubic crystal field



- e_g orbitals



- t_{2g} orbitals



J. van der Brink and D. Khomskii

Matrix U_{1234}

Multipole expansion:

$$\frac{1}{|\mathbf{r}-\mathbf{r}'|}=\sum_{kq}\frac{4\pi}{2k+1}\frac{r_<^k}{r_>^{k+1}}Y_{kq}^*(\hat{r})Y_{kq}(\hat{r}')$$

Coulomb matrix elements in Y_{lm} basis:

$$< mm'||m''m''' >=\sum_k a_k(m,m'',m',m''')F^k$$

Angular part – 3j symbols

$$a_k(m,m',m''',m''')=\sum_{q=-k}^{k}(2l+1)^2(-1)^{m+q+m'}\begin{pmatrix} l & k & l \\ 0 & 0 & 0\end{pmatrix}^2\begin{pmatrix} l & k & l \\ -m & -q & m'\end{pmatrix}\begin{pmatrix} l & k & l \\ -m'' & q & m'''\end{pmatrix}$$

Slater integrals:

$$F^k=e^2\int_0^\infty r^2dr|\varphi_d(r)|^2\int_0^\infty (r')^2dr'|\varphi_d(r')|^2\frac{r_<^k}{r_>^{k+1}}$$

Average interaction: U and J

Average Coulomb parameter:

$$U = \frac{1}{(2l+1)^2} \sum_{mm'} U_{mm'} = F^0$$

Average Exchange parameter:

$$J = \frac{1}{(2l+1)^2} \sum_{mm'} J_{mm'} = \sum_{k \neq 0} \begin{pmatrix} l & k & l \\ 0 & 0 & 0 \end{pmatrix}^2 F^k$$

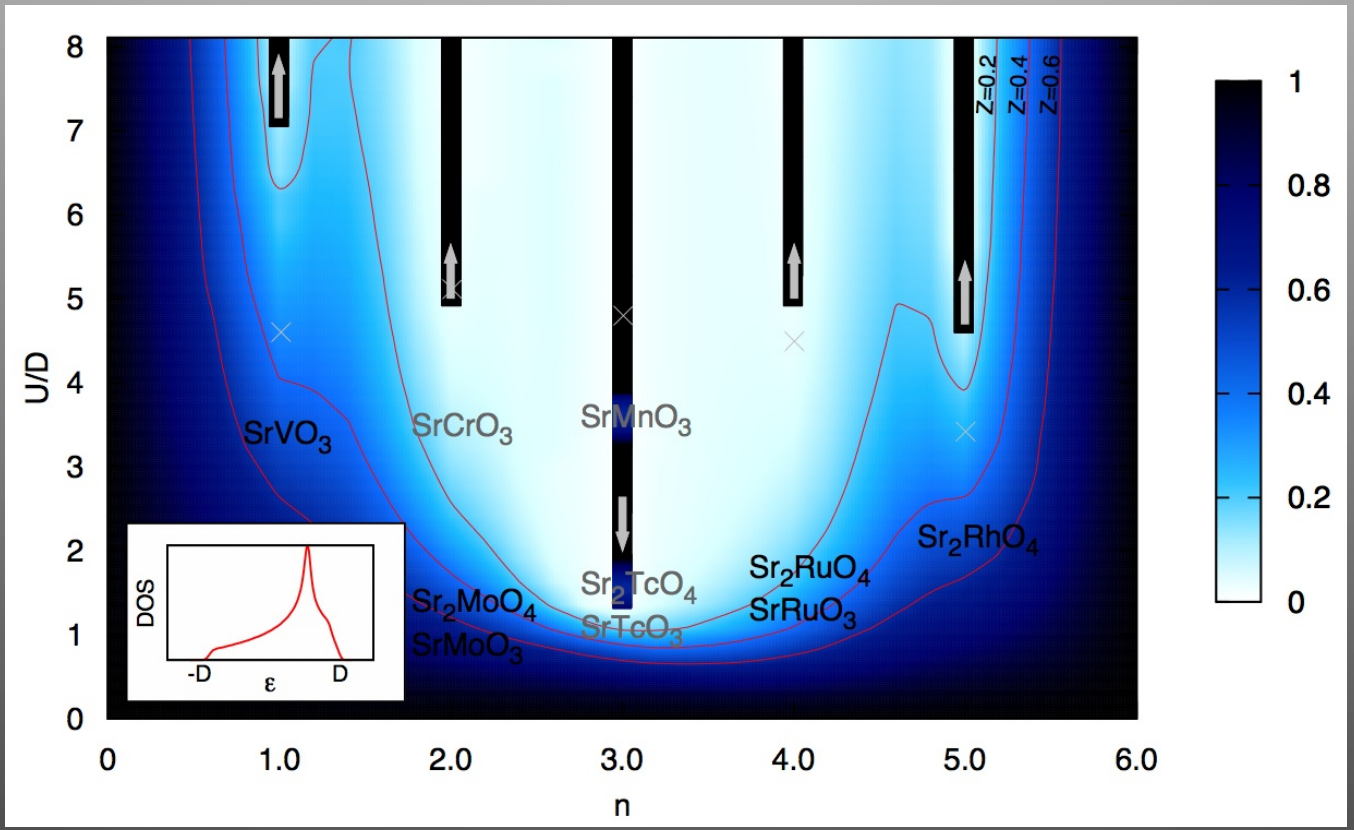
For d-electrons:

$$J_d = \frac{1}{14}(F^2 + F^4)$$

Coulomb and exchange interactions:

$$U_{mm'} = < mm' || mm' >$$
$$J_{mm'} = < mm' || m'm >$$

J - Hund's Materials



$$H_{t_{2g}} = (U - 3J)\,\frac{\hat{N}(\hat{N}-1)}{2} - 2J\,\vec{S}^2 - \frac{J}{2}\vec{L}^2 + \frac{5}{2}J\,\hat{N}$$

A. Georges et.al, arXiv:1207.3033

The Theory of Everything

Hamiltonian for multi-fermionic system in field-operators:

$$H = \sum_{\sigma} \int d\mathbf{r}\, \hat{\psi}^{\dagger}_{\sigma}(\mathbf{r}) \left(-\frac{1}{2}\nabla^2 + V(\mathbf{r}) - \mu \right) \hat{\psi}_{\sigma}(\mathbf{r})$$
$$+\frac{1}{2} \sum_{\sigma\sigma'} \int d\mathbf{r} \int d\mathbf{r}'\, \hat{\psi}^{\dagger}_{\sigma}(\mathbf{r}) \hat{\psi}^{\dagger}_{\sigma'}(\mathbf{r}')\, U(\mathbf{r}-\mathbf{r}')\, \hat{\psi}_{\sigma'}(\mathbf{r}') \hat{\psi}_{\sigma}(\mathbf{r})$$

Atomic Units: $\hbar = m = e = 1$

Coilomb interaction: $U(\mathbf{r}-\mathbf{r}') = 1/|\mathbf{r}-\mathbf{r}'|$

Second quantisation operators in orthonormal basis:

$$\hat{\psi}(\mathbf{r}) = \sum_{n} \phi_n(\mathbf{r}) \hat{c}_n$$
$$\hat{\psi}^{\dagger}(\mathbf{r}) = \sum_{n} \phi^*_n(\mathbf{r}) \hat{c}^{\dagger}_n$$

$n = (im\sigma)$

Wannier Basis: $\phi_n(\mathbf{r})$ with site, orbital andspins quantum numbers

QM-Alphabet

1-Q

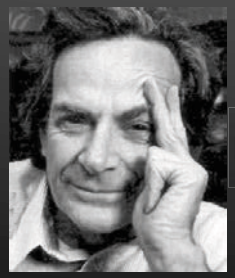
$$\left(-\frac{1}{2}\Delta + V_{eff}(\vec{r})\right)\psi(\vec{r}) = \varepsilon\psi(\vec{r})$$

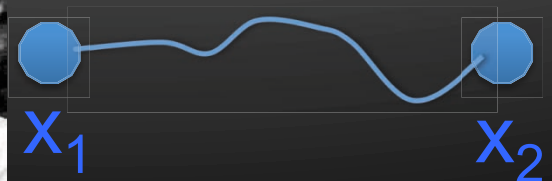
2-Q

$$\hat{H} = \sum_{ij\sigma} t_{ij}\hat{c}^{+}_{i\sigma}\hat{c}_{j\sigma} + \sum_{i} U\hat{n}_{\uparrow}\hat{n}_{\downarrow}$$

3-PI

$$Z = Sp(e^{-\beta\hat{H}}) = \int D[c^*,c]e^{-\int_0^\beta d\tau\left[c^*_\tau\partial_\tau c_\tau + H(c^*_\tau,c_\tau)\right]}$$





Path Integral: quick reference

http://www.physnet.uni-hamburg.de/hp/group_magno/pim_10.php

Literature:

1. J.W. Negele and H. Orland "Quantum Many-Particle Systems" (Addison-Wesley, 1988)
2. N. Nagaosa "Quantum Field Theory in Condensed Matter Physics" (Springer, 1999)
3. A. Atland and B. Simons "Condensed Matter Field Theory" (Cambridge University Press 2006)

http://www.tcm.phy.cam.ac.uk/~bds10/

4. E. Fradkin "Field Theory of Condensed Matter Systems" (Addison Wesley, 1991)
5. P. Coleman "Many Body Physics"

http://www.physics.rutgers.edu/~coleman/pdf/bk.pdf

Summary: $\zeta = \begin{cases} +1 & \text{Boson} \\ -1 & \text{Fermion} \end{cases}$ $\quad [\hat{c}_i, \hat{c}_j^\dagger] = \delta_{ij}^{\zeta}$

$$\hat{c}_i |c\rangle = c_i |c\rangle \quad \text{with} \quad |c\rangle = e^{\zeta \sum_i c_i \hat{c}_i^\dagger} |0\rangle$$

$$\langle c| \hat{c}_i^\dagger = \langle c| c_i^* \quad \text{with} \quad \langle c| = \langle 0| e^{\zeta \sum_i \hat{c}_i c_i^*}$$

$$\hat{c}_i^\dagger |c\rangle = \zeta \partial_{c_i} |c\rangle \quad \text{and} \quad \langle c| \hat{c}_i = \partial_{c_i^*} \langle c|$$

Overlap: $\langle \bar{c} | c \rangle = e^{\sum_i \bar{c}_i^* c_i}$

Completeness: $\int dc^* dc \, e^{-\sum_i c_i^* c_i} |c\rangle\langle c| = \hat{\mathbb{1}}_{\mathcal{F}}$

Trace $\forall \hat{H}$: $\mathrm{Tr}\,\hat{H} = \sum_n \langle n| \hat{H} |n\rangle = \int dc^* dc \, e^{-\sum_i c_i^* c_i} \langle \zeta c| \hat{H} |c\rangle$

P.I. measure: $\mathcal{D}[c^*, c] \equiv \lim_{N\to\infty} \prod_i \frac{dc_i^* dc_i}{(2\pi i)^{(1+\zeta)/2}}$

3.2 Fermion coherent state: $\{\hat{c}_i, \hat{c}_j^+\} = \delta_{ij}$

$$\hat{c}_i |c\rangle = c_i |c\rangle$$

Anticommutivity of $\{\hat{c}_i, \hat{c}_j\} = 0$ implies that eigenvalues c_i anticommute $c_i c_j + c_j c_i = 0$

$\rightarrow$ $c_i c_j = -c_j c_i$ is not ordinary number!

Grassmann Algebra: $\{c_i\}$

1) Anticommuting number: $c_i c_j + c_j c_i = 0$
2) $c_i^2 = 0$
3) Anticommute with Fermi-operators: $c\hat{c}^+ + \hat{c}^+ c = 0$
4) Can be add to and multiply by complex numbers. (f)

   Functions: $F(c_i) = c_i + \sum_j f_j c_j$

5) Differentiation! $\partial_{c_i} c_j = \delta_{ij}$

   N.B. ordering! $\partial_{c_i} c_j c_i = -c_j$

6) Integration:

$$\left.\begin{aligned} \int dc\, 1 &= 0 \\ \int dc\, c &= 1 \end{aligned}\right\} \quad \int \equiv \partial$$

General Path-Integral representation:

$$Z = \int_{c(\beta)=\zeta c(0)} \mathcal{D}[c^*, c]\, e^{-\int_0^\beta d\tau \left[c^* \frac{\partial}{\partial \tau} c + H(c^*, c) - \mu N(c^*, c) \right]}$$

analogy: Feynman: $\int dp\, dq\, e^{i \int_1^2 dt (p \partial_t q - H(p,q))}$

Coherent state: $\int dc^*\, dc\, e^{i \int_1^2 dt (c^* \partial_t c - H(c^*, c))}$

1 ⇝ 2

$\Rightarrow$ c^* ← conjugate → c

with boundary condition!

Boson

$$c(\beta) = c(0)$$
$$c^*(\beta) = c^*(0)$$

Fermion

$$c(\beta) = -c(0)$$
$$c^*(\beta) = -c^*(0)$$

For our Hamiltonian: $Z = \int \mathcal{D}[c^*, c]\, e^{-S[c^*, c]}$

with Action:

$$S[c^*, c] = \int_0^\beta d\tau \left\{ \sum_{ij} c_i^*(\tau) \left[(\partial_\tau - \mu)\delta_{ij} + h_{ij} \right] c_j(\tau) + \frac{1}{2} \sum_{ijkl} V_{ijkl}\, c_i^*(\tau) c_j^*(\tau) c_l(\tau) c_k(\tau) \right\}$$

Hubbard-Stratonovich Transformation

Reminder! Gauß: $\int_{-\infty}^{\infty} dx\, e^{-ax^2+bx} = \sqrt{\frac{\pi}{a}}\, e^{\frac{b^2}{4a}}$

$$\int \mathcal{D}[c^*, c] \exp\left(-\sum_{i,j} c_i^* M_{ij} c_j + \sum_i (J_i^* c_i + c_i^* J_i)\right) = (\det M)^{-\zeta} \exp\left(\sum_{i,j} J_i^* (M^{-1})_{ij} J_j\right)$$

1) One-bosonic field - ϕ ($U n^2$ - interaction)

$$e^{-n_m U_{ml} n_l} = \int \mathcal{D}[\phi]\, e^{-\frac{1}{4}\phi_m U^{-1}_{ml} \phi_l - i\phi_m n_m}$$

2) Fermionic transformation: $(c^*, c) \longrightarrow (f^*, f)$

Grassmann — Grassmann

$$e^{c_i^* G_{ij} c_j} = \det(G) \int \mathcal{D}[f^*, f]\, e^{-f_i^* G^{-1}_{ij} f_j + f_i^* c_i + c_i^* f_i}$$

Path Integral for Everything

Euclidean action

$$
\begin{aligned}
Z &= \int \mathcal{D}[c^*, c] e^{-S} \\
S &= \sum_{12} c_1^* \left(\partial_\tau + t_{12}\right) c_2 + \frac{1}{4} \sum_{1234} c_1^* c_2^* U_{1234} c_4 c_3
\end{aligned}
$$

One- and two-electron matrix elements:

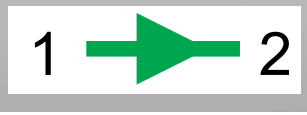
$$
\begin{aligned}
t_{12} &= \int d\mathbf{r}\, \phi_1^*(\mathbf{r}) \left(-\frac{1}{2} \nabla^2 + V(\mathbf{r}) - \mu \right) \phi_2(\mathbf{r}) \\
U_{1234} &= \int d\mathbf{r} \int d\mathbf{r}'\, \phi_1^*(\mathbf{r}) \phi_2^*(\mathbf{r}')\, U(\mathbf{r} - \mathbf{r}')\, \phi_3(\mathbf{r}) \phi_4(\mathbf{r}')
\end{aligned}
$$

Shot notation:

$$
\sum_1 \ldots \equiv \sum_{im} \int d\tau \ldots
$$

One- and Two-particle Green Functions

One-particle Green function



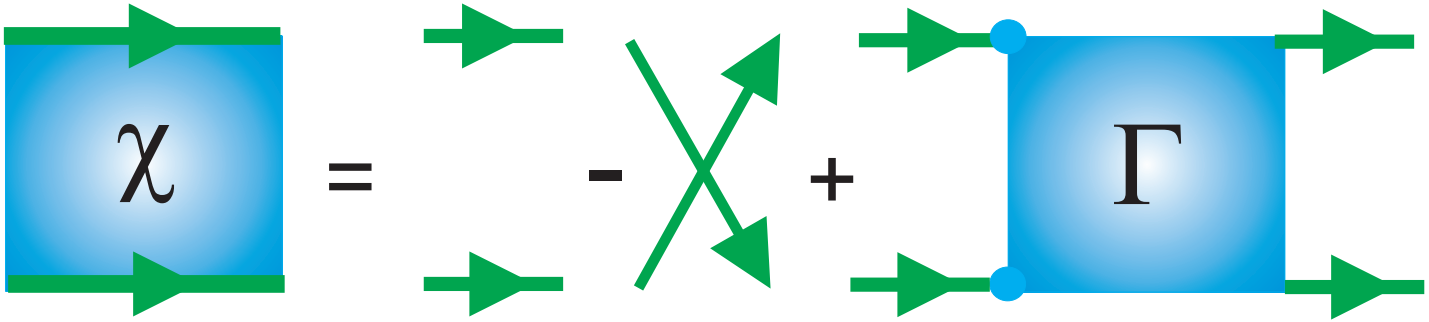
$$G_{12} = -\langle c_1 c_2^* \rangle_S = -\frac{1}{Z} \int \mathcal{D}[c^*, c]\, c_1 c_2^*\, e^{-S}$$

Two-particle Green function (generalized susceptibilities)

$$\chi_{1234} = \langle c_1 c_2 c_3^* c_4^* \rangle_S = \frac{1}{Z} \int \mathcal{D}[c^*, c]\, c_1 c_2 c_3^* c_4^*\, e^{-S}$$

Vertex function:

$$X_{1234} = G_{14} G_{23} - G_{13} G_{24} + \sum_{1'2'3'4'} G_{11'} G_{22'} \Gamma_{1'2'3'4'} G_{3'3} G_{4'4}$$



Baym-Kadanoff functional

Source term $$S[J] = S + \sum_{ij} c_i^* J_{ij} c_j$$

Partition function and Free-energy:

$$Z[J] = e^{-F[J]} = \int \mathcal{D}[c^*, c]\, e^{-S[J]}$$

Legendre transforming from J to G:

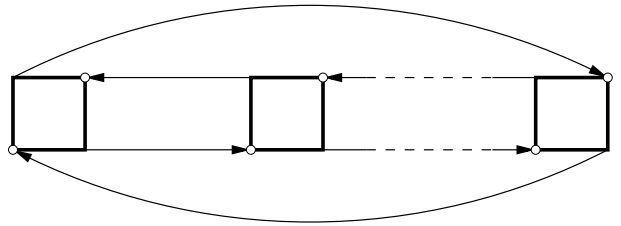
$$F[G] = F[J] - \mathrm{Tr}(JG)$$

$$G_{12} = \frac{1}{Z[J]} \frac{\delta Z[J]}{\delta J_{12}}\bigg|_{J=0} = \frac{\delta F[J]}{\delta J_{12}}\bigg|_{J=0}$$

Decomposition into the single particle part and correlated part

$$F[G] = \mathrm{Tr} \ln G - \mathrm{Tr}\,(\Sigma G) + \Phi[G]$$

$\Phi[G]$ = $\sum_i$



Baym-Kadanoff Functional

$$F[G] = -Tr\ln[-(G_0^{-1} - \Sigma[G])] - Tr(\Sigma[G]G) + \Phi[G]$$

Exact representation of Φ: $V^{\alpha}_{ee} = \alpha V_{ee}$

$$\Phi = \frac{1}{2}\int_0^1 d\alpha Tr[V_{ee}^{\alpha} < \psi^{+}\psi^{+}\psi\psi >]$$

Different Functionals:

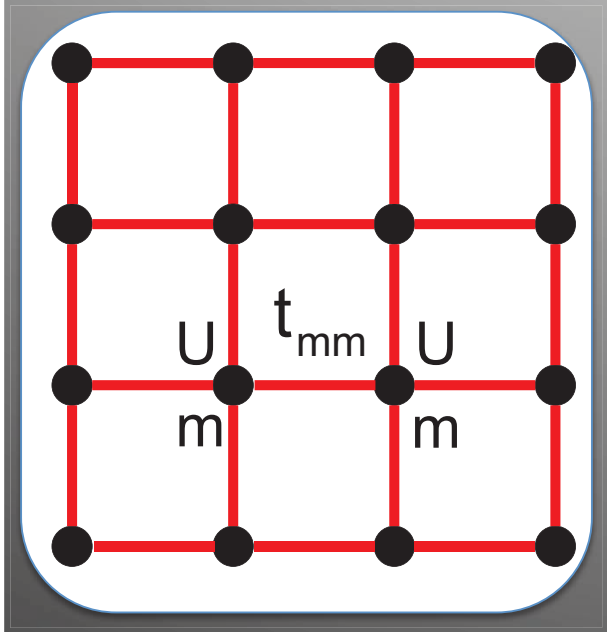
DFT:	$G=\rho$	$J=V=V_h+V_{xc}$
DMFT:	$G=G(i\omega)$	$J=\Sigma_{loc}(i\omega)$
BKF:	$G=G(k,i\omega)$	$J=\Sigma(k,i\omega)$

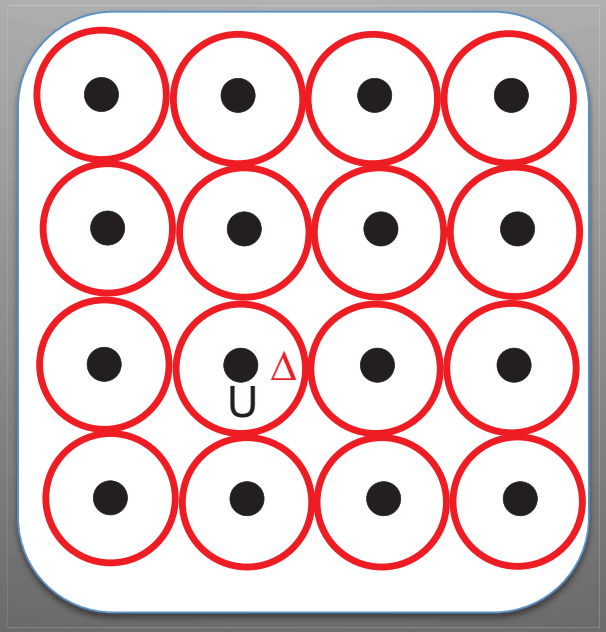
Functionals: MFT- DFT- DMFT-BK

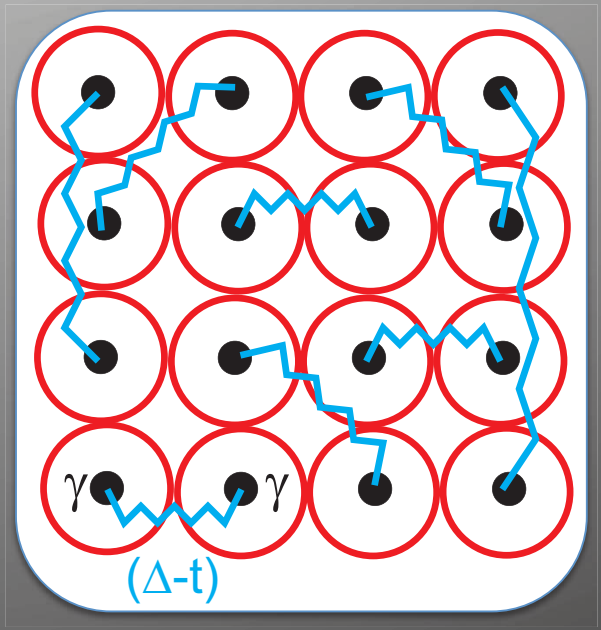
G. Kotliar et. al. RMP (2006), A. Georges (2004)

- Weiss Mean-Field Theory (MFT)
  of classical magnets
- Kohn Density Functional Theory (DFT)
  of inhomogeneous electron gas in solids
- Dynamical Mean-Field Theory (DMFT)
  of strongly correlated electron systems
- Baym-Kadanoff Functional

Hwo to find DMFT-functional?







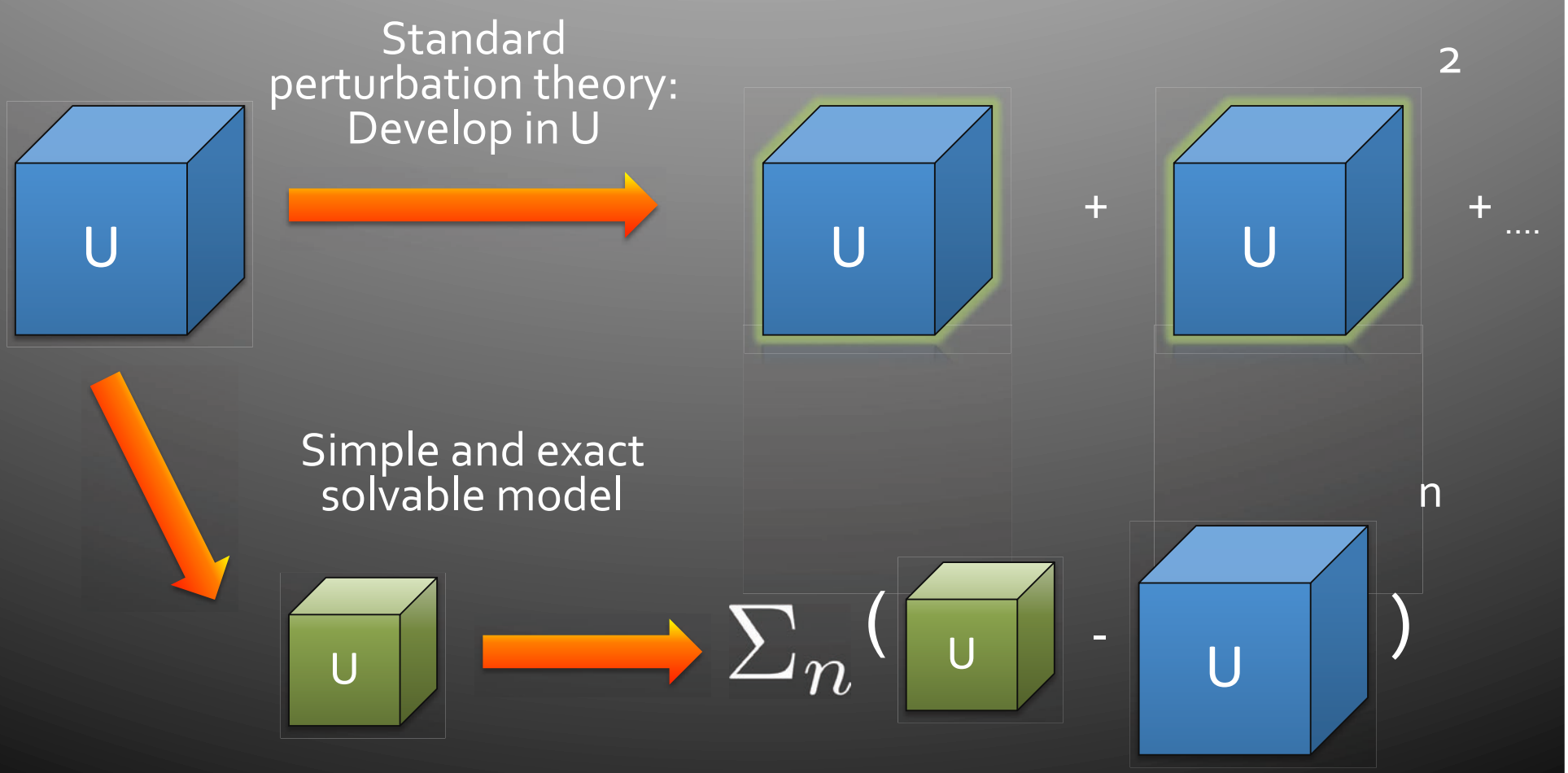
Dual Fermions: Basic

Start from
Correlated Lattice

Find the optimal
Reference System
Bath hybridization

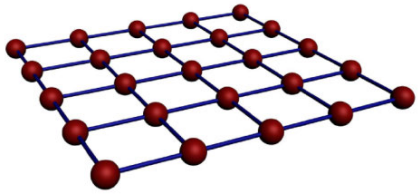
Expand around
DMFT solution

Superperturbation



Dual Fermion scheme

General Lattice Action $H = h + U$

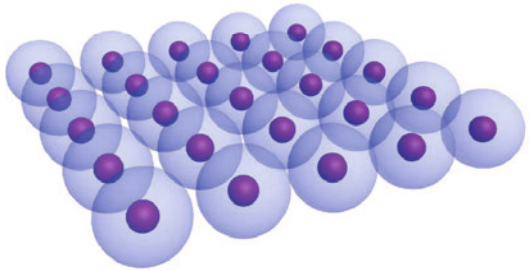


$$S[c^*,c] = \sum_{\omega k m m' \sigma} \left[h_k^{mm'} - (i\omega+\mu)1\right] c^*_{\omega k m\sigma} c_{\omega k m'\sigma} + \frac{1}{4} \sum_{i\{m,\sigma\}} \int_0^\beta U_{1234} c_1^* c_2^* c_3 c_4 d\tau$$

Reference system: Local Action with hybridization Δ_ω

$$S_{loc} = \sum_{\omega m m' \sigma} \left[\Delta_\omega^{mm'} - (i\omega+\mu)1\right] c^*_{\omega m\sigma} c_{\omega m'\sigma} + \frac{1}{4} \sum_{i\{m,\sigma\}} \int_0^\beta U_{1234} c_1^* c_2^* c_3 c_4 d\tau$$

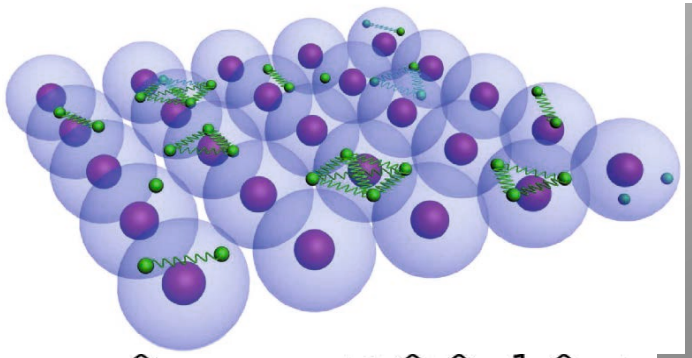
Lattice-Impurity connection:



$$S[c^*,c] = \sum_i S_{loc}[c_i^*, c_i] + \sum_{\omega k m m' \sigma} \left(h_k^{mm'} - \Delta_\omega^{mm'}\right) c^*_{\omega k m\sigma} c_{\omega k m'\sigma}$$

A. Rubtsov, et al, PRB **77**, 033101 (2008)

Dual Transformation



Gaussian path-integral

$$\int D[\vec{f}^*,\vec{f}]\exp(-\vec{f}^*\hat{A}\vec{f}+\vec{f}^*\hat{B}\vec{c}+\vec{c}^*\hat{B}\vec{f}) = \det(\hat{A})\exp(\vec{c}^*\hat{B}\hat{A}^{-1}\hat{B}\vec{c})$$

With

$$\begin{aligned} A &= g_\omega^{-1}(\Delta_\omega - h_k)g_\omega^{-1} \\ B &= g_\omega^{-1} \end{aligned}$$

new Action:

$$S_d[f^*,f] = -\sum_{k\omega}\tilde{\mathcal{G}}^{-1}_{k\omega}f^*_{k\omega}f_{k\omega}+\frac{1}{4}\sum_{1234}\gamma^{(4)}_{1234}f^*_1f^*_2f_4f_3+\dots$$

Diagrammatic:

$$\longrightarrow \quad \mathcal{G}_{k\omega} = \tilde{G}^{DMFT}_{k\omega} - g_\omega$$

$$\square \quad \gamma^{(4)}_{1234} = g^{-1}_{11'}g^{-1}_{22'}\left(\chi_{1'2'3'4'}-\chi^0_{1'2'3'4'}\right)g^{-1}_{3'3}g^{-1}_{4'4}$$

g_ω and $\chi_{\nu,\nu',\omega}$ from DMFT impurity solver

Dual Fermion Action: Details

Lattice - dual action

$$S[c^*, c, f^*, f] = \sum_i S_{\text{site},i} + \sum_{\omega \mathbf{k}\, \alpha\beta} f^*_{\omega \mathbf{k}\alpha} [g_\omega^{-1} \left(\Delta_\omega - t_{\mathbf{k}}\right)^{-1} g_\omega^{-1}]_{\alpha\beta} f_{\omega \mathbf{k}\beta}$$

$$S_{\text{site},i}[c_i^*, c_i, f_i^*, f_i] = S_{\text{loc}}[c_i^*, c_i] + \sum_{\alpha\beta} f^*_{\omega i\alpha}\, g^{-1}_{\omega\,\alpha\beta} c_{\omega i\beta} + c^*_{\omega i\alpha}\, g^{-1}_{\omega\,\alpha\beta} f_{\omega i\beta}$$

For each site I integrate-out c-Fermions:

$$\int \mathcal{D}[c^*, c] \exp\left(-S_{\text{site}}[c_i^*, c_i, f_i^*, f_i]\right) =$$

$$\mathcal{Z}_{\text{loc}} \exp\left(- \sum_{\omega\, \alpha\beta} f^*_{\omega i\alpha}\, g^{-1}_{\omega\,\alpha\beta} f_{\omega i\beta} - V_i[f_i^*, f_i]\right)$$

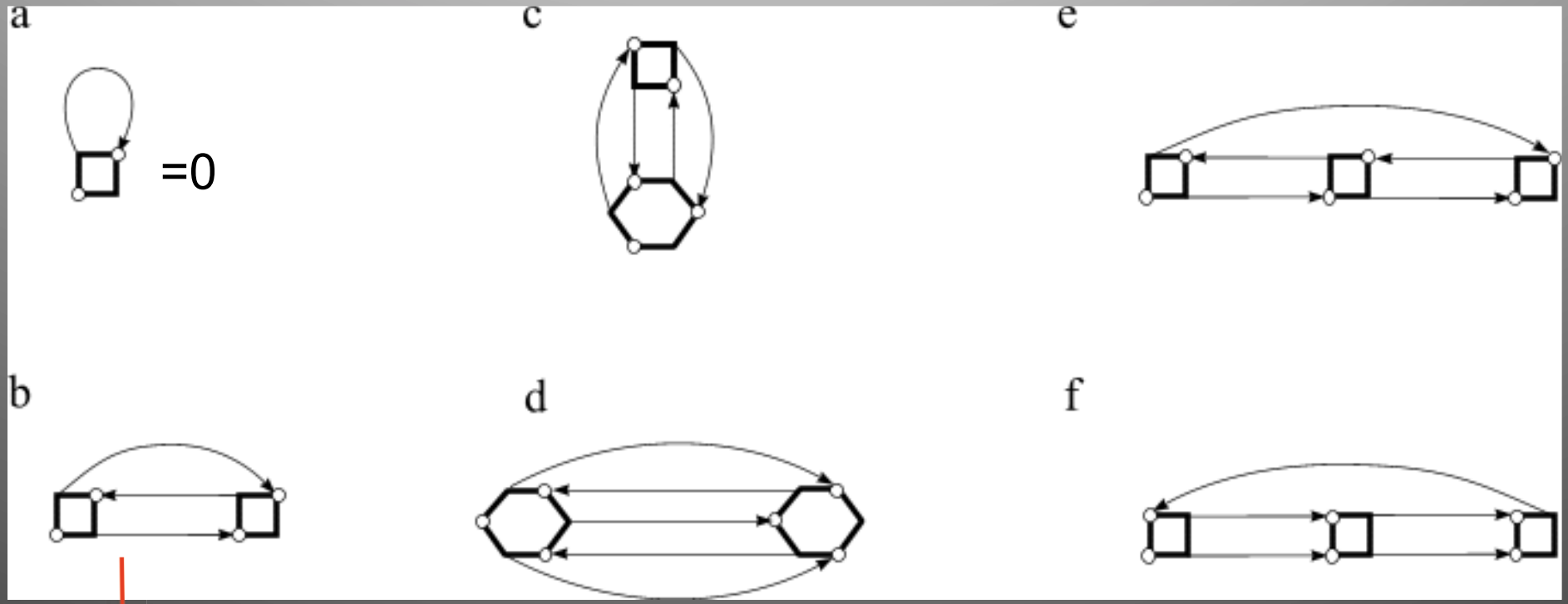
Dual potential:

$$V[f^*, f] = \frac{1}{4} \gamma_{1234} f_1^* f_2^* f_4 f_3 + \ldots$$

$$\gamma_{1234} = g^{-1}_{11'} g^{-1}_{22'} \left[\chi_{1'2'3'4'} - \chi^0_{1'2'3'4'}\right] g^{-1}_{3'3} g^{-1}_{4'4} \qquad \chi^0_{1234} = g_{14} g_{23} - g_{13} g_{24}$$

$$\chi^{1234} = \langle c_1 c_2 c_3^* c_4^* \rangle_{\text{loc}} = \frac{1}{\mathcal{Z}_{\text{loc}}} \int \mathcal{D}[c^*, c] c_1 c_2 c_3^* c_4^* \exp\left(- S_{\text{loc}}[c^*, c]\right)$$

Basic diagrams for dual self-energy



Lines - dual Green's function.

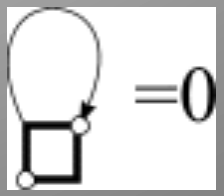
$$\omega \quad \mathbf{k} \qquad \left[\quad \bar{\omega} \qquad \Delta_{\omega} - \quad \mathbf{k} \right]^{-} - \quad \omega$$

$$\Sigma \quad \mathbf{k} \qquad -- \left(- \right) \sum_{\mathbf{k}_1 \mathbf{k}_2} \sum \gamma \qquad \mathbf{k} \qquad \mathbf{k} \qquad \mathbf{k} \quad \mathbf{k} - \mathbf{k} \quad \gamma$$

Condition for Δ and relation with DMFT

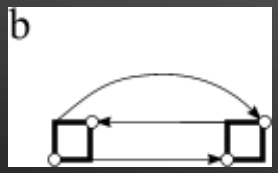
$G^d = G^{DMFT} - g$

To determine Δ, we require
that Hartree correction in dual variables vanishes.
If no higher diagrams are taken into account, one obtains DMFT:



$$\frac{1}{N}\sum_{\mathbf{k}} \tilde{G}^0_\omega(\mathbf{k}) = 0 \quad \Longleftrightarrow \quad \frac{1}{N}\sum_{\mathbf{k}} G^{\mathrm{DMFT}}_\omega(\mathbf{k}) = g_\omega$$

Higher-order diagrams give corrections to the DMFT self-energy,
and already the leading-order correction is nonlocal.



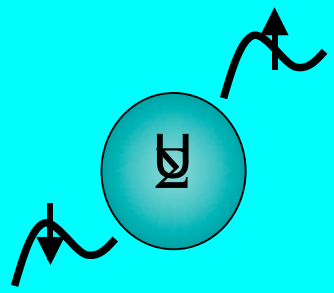
$$\Sigma(k,\omega) = \Sigma_{DMFT}(\omega) + \Sigma_d(k,\omega)/[1+g\Sigma_d(k,\omega)]$$

Dynamical Mean Field Theory

$$\hat{G}(i\omega_n) = \frac{1}{\Omega}\sum_{k}^{BZ} \hat{G}(\vec{k}, i\omega_n)$$

$$\hat{\mathcal{G}}_0^{-1}(i\omega_n) = \hat{G}^{-1}(i\omega_n) + \hat{\Sigma}(i\omega_n)$$

$\mathcal{G}_0(\tau - \tau')$



QMC ED

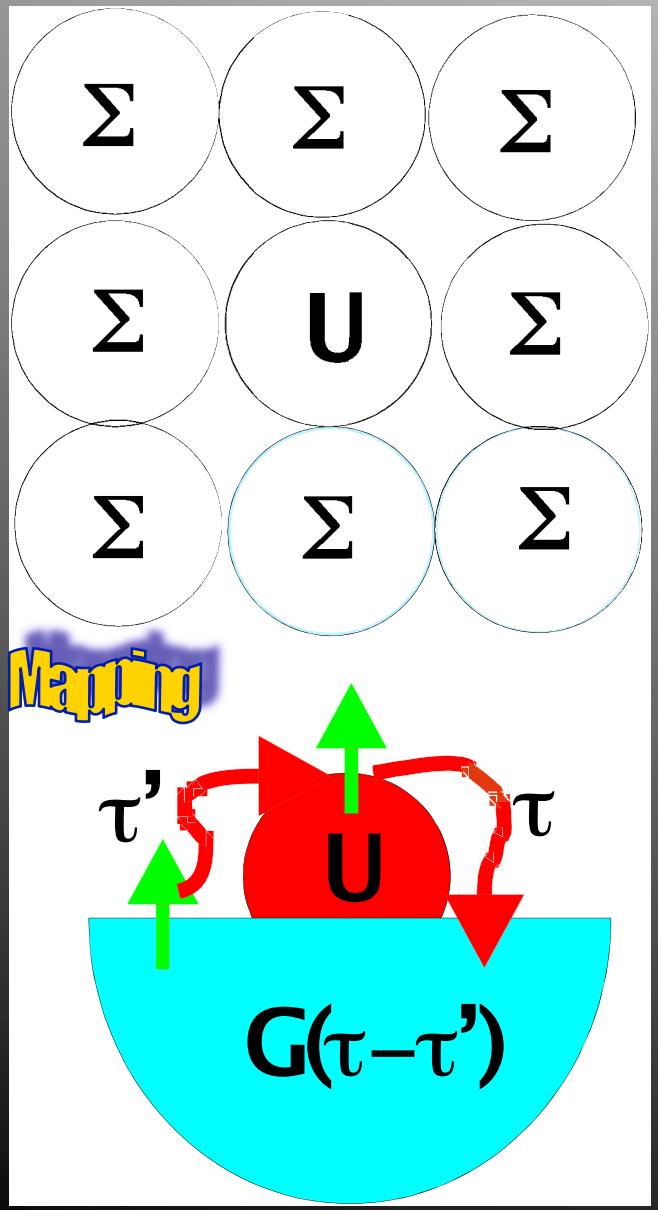
Single Impurity Solver

DMRG FLEX IPT

$$\hat{\Sigma}_{new}(i\omega_n) = \hat{\mathcal{G}}_0^{-1}(i\omega_n) - \hat{G}^{-1}(i\omega_n)$$

V. Metzner and D. Vollhardt (1987)
A. Georges and G. Kotliar (1992)

Dynamical Mean Field Theory



$$\hat{G}(i\omega_n) = \frac{1}{\Omega}\sum_{\vec{k}}^{BZ}\left[\hat{I}(\mu + i\omega_n) - \hat{H}_0(\vec{k}) - \hat{\Sigma}(i\omega_n)\right]^{-1}$$

$$\hat{\mathcal{G}}_0^{-1}(i\omega_n) = \hat{G}^{-1}(i\omega_n) + \hat{\Sigma}(i\omega_n)$$

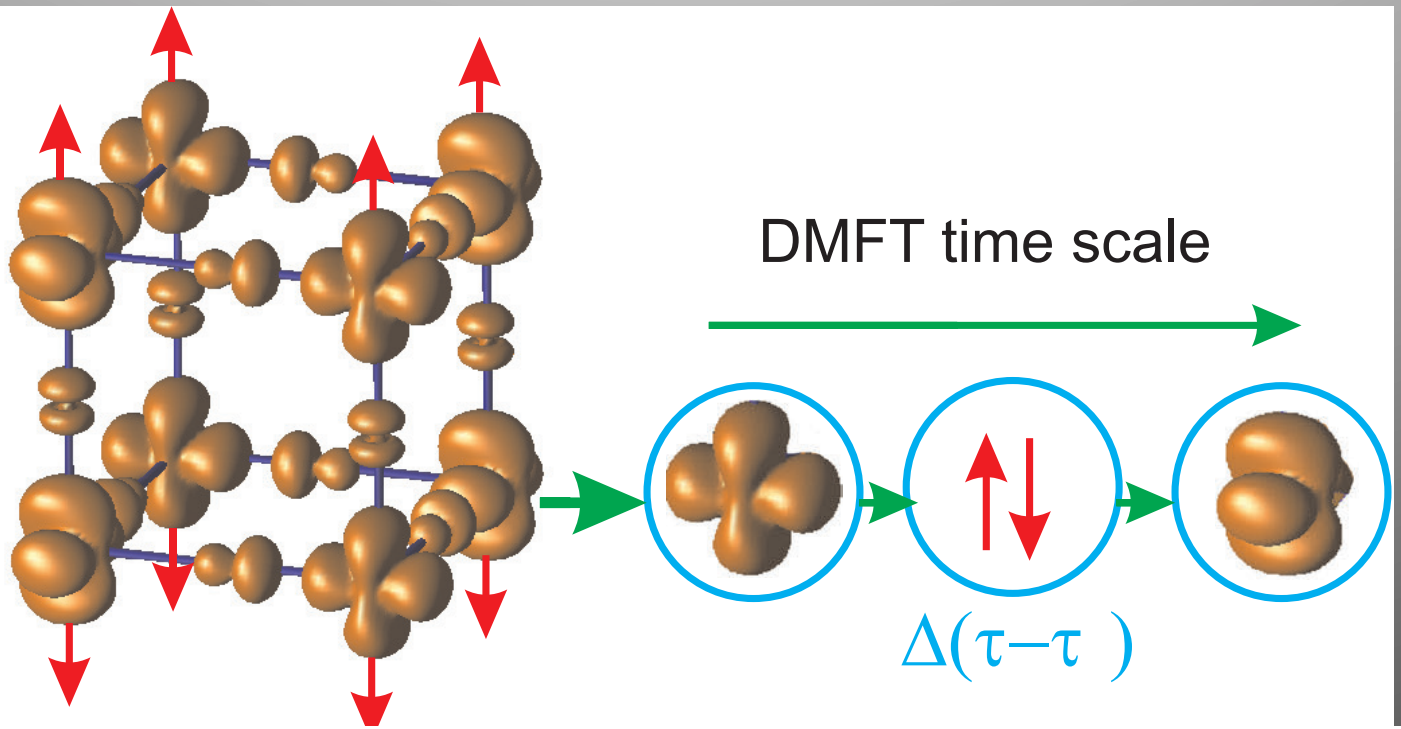
$$S_{eff} = -\int\int d\tau d\tau' c_\sigma^+(\tau)\mathcal{G}_0^{-1}(\tau-\tau')c_\sigma(\tau') + \int d\tau U n^\uparrow(\tau) n^\downarrow(\tau)$$

$$\hat{G}(\tau-\tau') = -\frac{1}{Z}\int D[c,c^+]c(\tau)c^+(\tau')e^{-S_{eff}}$$

$$\hat{\Sigma}_{new}(i\omega_n) = \hat{\mathcal{G}}_0^{-1}(i\omega_n) - \hat{G}^{-1}(i\omega_n)$$

W. Metzner and D. Vollhardt, PRL(1989)
A. Georges et al., RMP 68, 13 (1996)

DMFT: SCF +Fluctuations



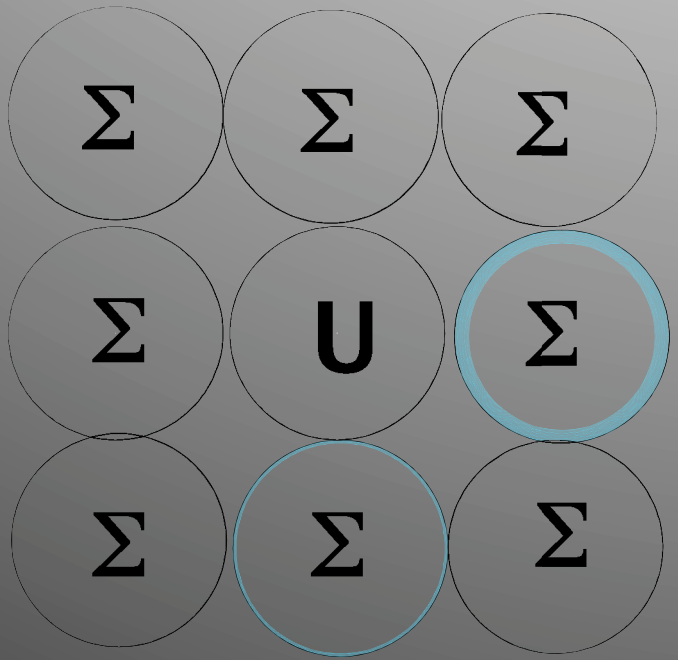
$$S[c^* \;\; c] = -\sum_{\omega \mathbf{k} \sigma \, m m'} c^*_{\omega \mathbf{k} \sigma m} \left[(i\omega + \;\;)\mathbf{1} - t^{m m'}_{\mathbf{k}\sigma} \right] c_{\omega \mathbf{k} \sigma m'} + \sum_i S_{\mathrm{U}}[c^*_i \;\; c_i]$$

$$S_{\mathrm{loc}}[c^* \;\; c] = -\sum_{\omega \, \alpha\beta} c^*_{\omega\alpha} \left[(i\omega + \;\;)\mathbf{1} - \Delta^{\alpha\beta}_{\omega} \right] c_{\omega\beta} + S_{\mathrm{U}}[c^* \;\; c]$$

$$g_{12} = -\langle c_1 c^*_2 \rangle_{\mathrm{loc}}$$

$$\sum_{\mathbf{k}} \left[g^{-1}_{\omega} + \Delta_{\omega} - t_{\mathbf{k}} \right]^{-1} = g_{\omega}$$

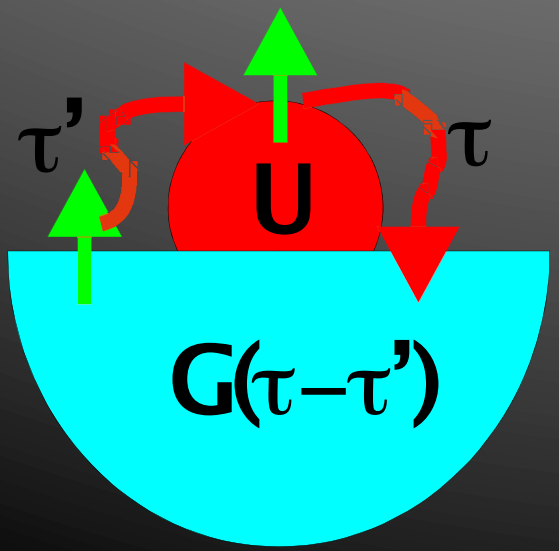
Quantum Impurity Solver



$$Z = \int \mathcal{D}[c^* \;\; c] e^{-S_{simp}}$$

$$S_{simp} = -\sum_{I\;J=0}^{N} \int_0^\beta d\tau \int_0^\beta d\tau' \, c^*_{I\sigma}(\tau) \left[\mathcal{G}^{-1}_\sigma(\tau - \tau')\right]_{IJ} c_{J\sigma}(\tau')$$

$$+ \sum_{I=1}^{N} \int_0^\beta d\tau U n_{I\uparrow}(\tau) n_{I\downarrow}(\tau)$$



What is a best scheme?
Quantum Monte Carlo !

Imputity solver: miracle of CT-QMC

$$S=\sum_{\sigma\sigma'}\int_0^\beta d\tau\int_0^\beta d\tau[-G_0^{-1}(\tau-\tau')c_\sigma^+(\tau)c_\sigma(\tau')+\frac{1}{2}U\delta(\tau-\tau')c_\sigma^+(\tau)c_{\sigma'}^+(\tau)c_{\sigma'}(\tau')c_\sigma(\tau')]$$

$$G_0^{-1}(\tau-\tau')=\delta(\tau-\tau')[\frac{\partial}{\partial\tau}+\mu]-\Delta(\tau-\tau')$$

Interaction expansion CT-INT: A. Rubtsov et al, JETP Lett (2004)

$$Z=Z_0\sum_{k=0}^{\infty}\frac{(-U)^k}{k!}Tr\,\det[G_0(\tau-\tau')]$$

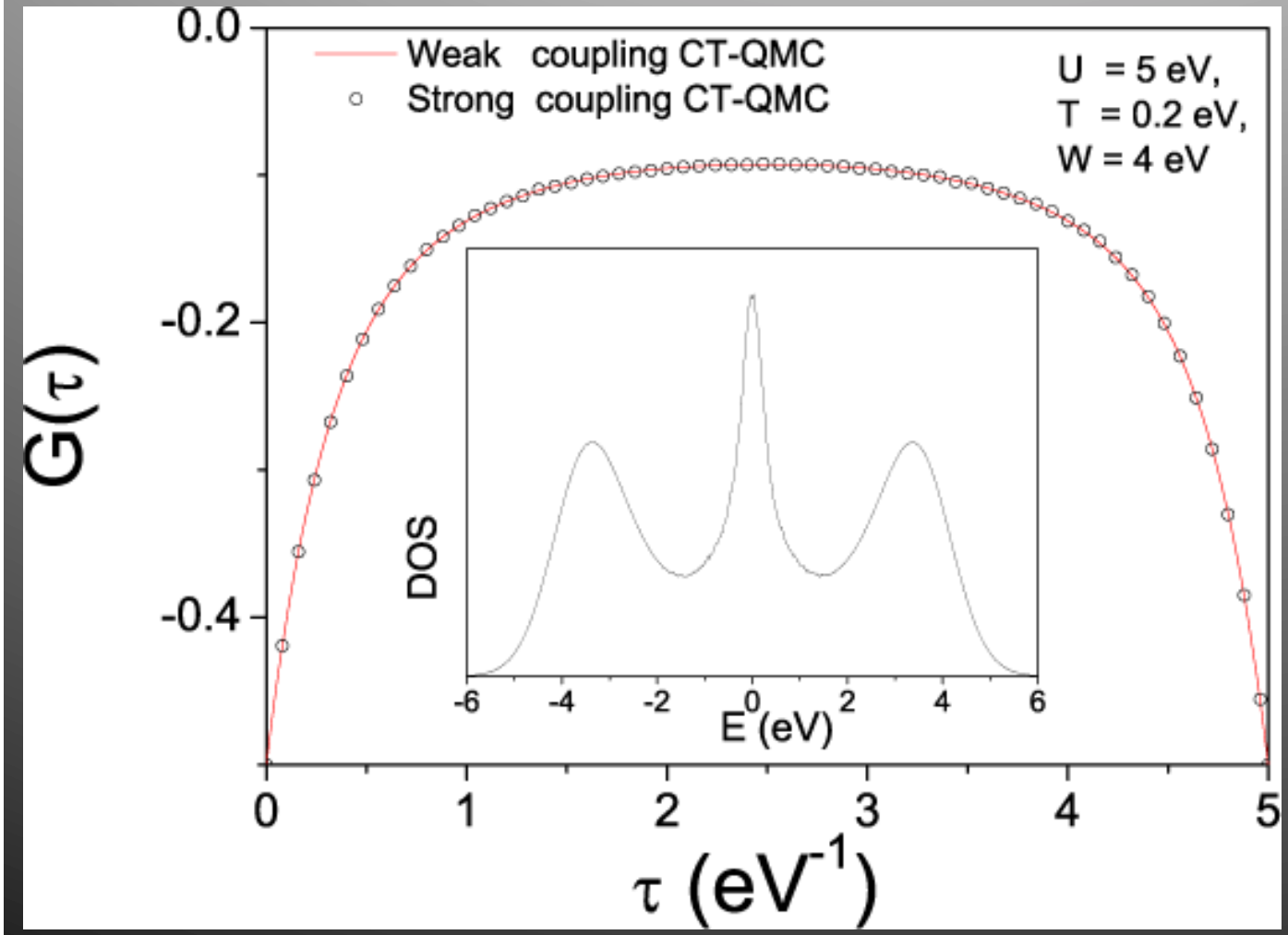
Hybridization expansion CT-HYB: P. Werner et al, PRL (2006)

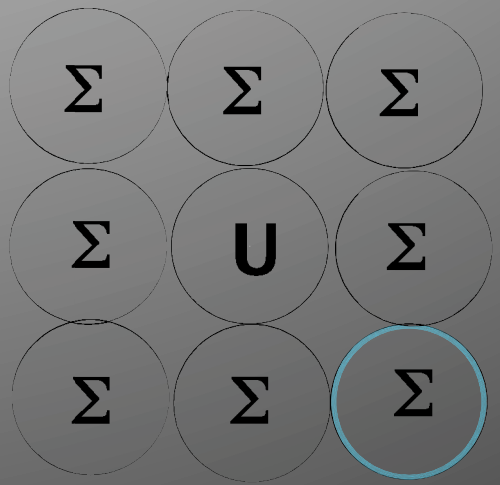
$$Z=Z_0\sum_{k=0}^{\infty}\frac{1}{k!}Tr\left\langle c_\sigma^+(\tau)c_\sigma(\tau')...c_{\sigma'}^+(\tau)c_{\sigma'}(\tau')\right\rangle_0\det[\Delta(\tau-\tau')]$$

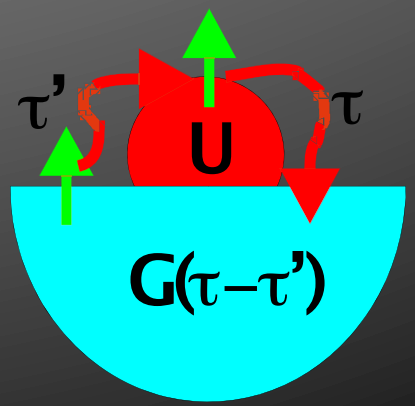
Efficient Krylov scheme: A. Läuchli and P. Werner, PRB (2009)

E. Gull, et al, RMP **83**, 349 (2011)

Comparison of different CT-QMC



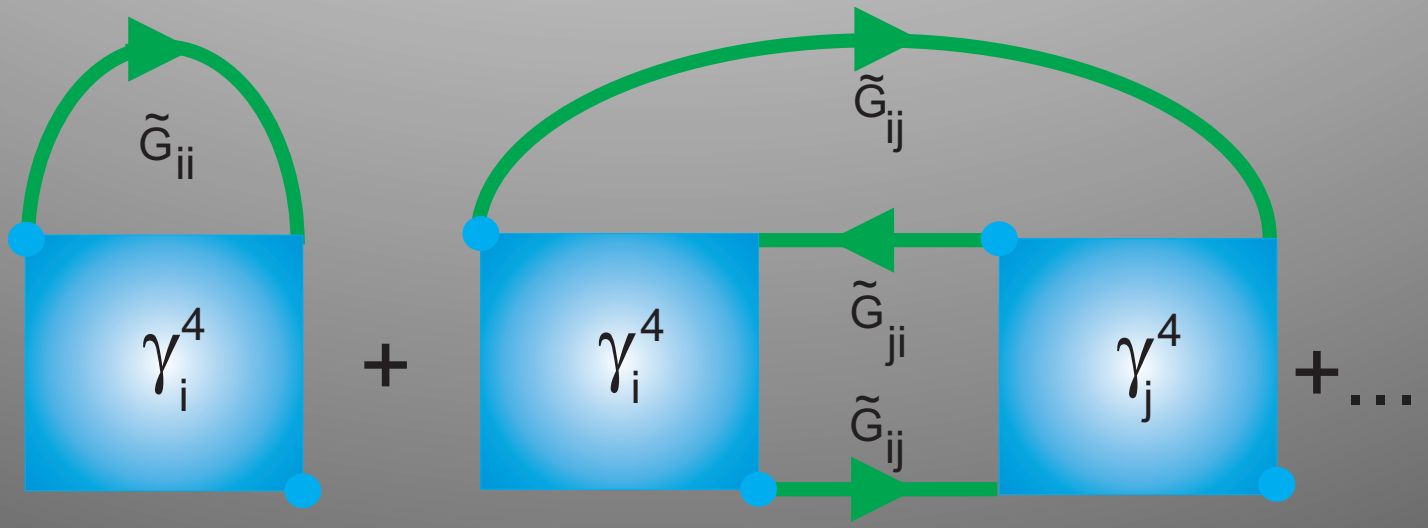




Ch. Jung, unpublished

CT-QMC review:E. Gull et al. RMP (2011)

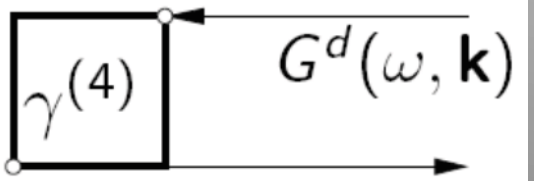
Dual Fermions: Diagrams



$$\tilde{\Sigma}^{(1)}_{12} = -T \sum_{34} \gamma_{1324}\, \tilde{G}^{\mathrm{loc}}_{43}$$

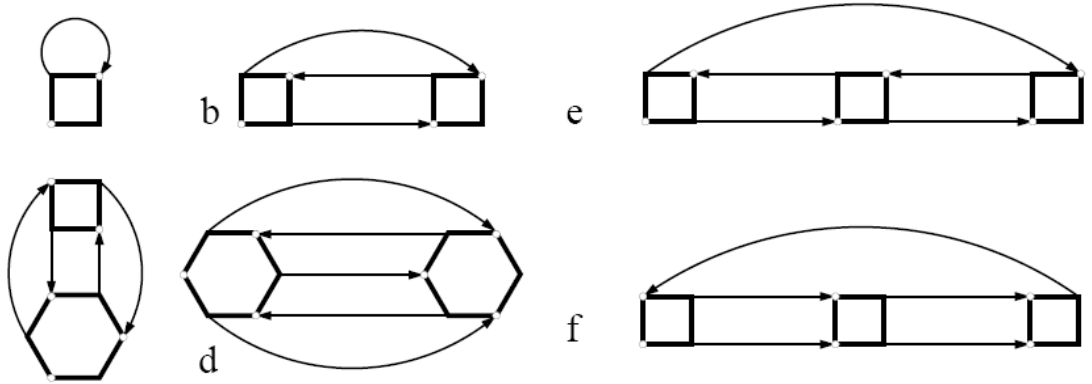
$$\tilde{\Sigma}^{(2)}_{12}(\mathbf{k}) = -\frac{1}{2}\left(\frac{T}{N_k}\right)^2 \sum_{\mathbf{k}_1\mathbf{k}_2} \sum_{345678} \gamma_{1345}\, \tilde{G}_{57}(\mathbf{k}_1)\, \tilde{G}_{83}(\mathbf{k}_2)\, \tilde{G}_{46}(\mathbf{k}+\mathbf{k}_2-\mathbf{k}_1)\, \gamma_{6728}$$

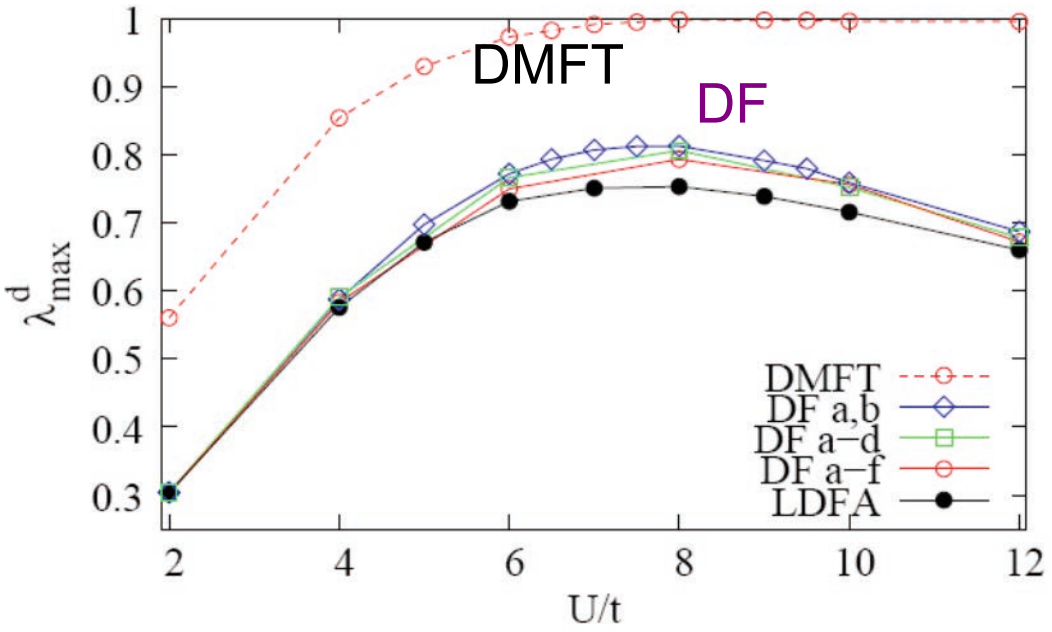
Convergence of Dual Fermions: 2d

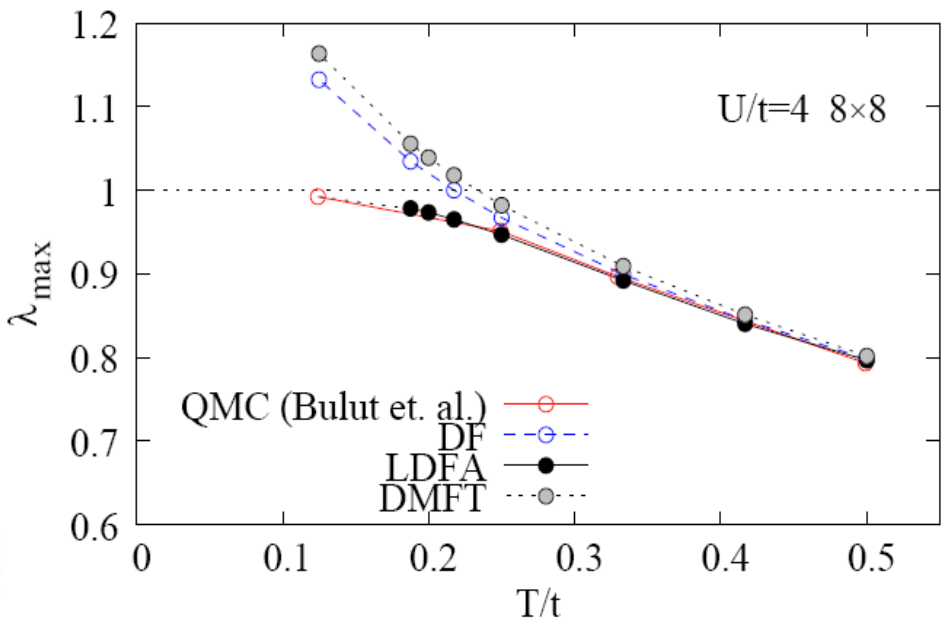


$=1$

$$-\frac{T}{N}\sum_{w'\mathbf{k}'}\Gamma^{d\,\mathrm{irr},s}_{\omega\omega'\Omega}\,G^d_{\omega'}(\mathbf{k}')\,G^d_{\omega'+\Omega}(\mathbf{k}'+\mathbf{q})\phi_{\omega'}=\lambda\phi_{\omega}$$







H. Hafermann, et al. PRL**102**, 206401 (2009)

Dual and Lattice Green's Functions

Two equivalent forms for partition function:

$$F[J^*, J; L^*, L] = \ln \mathcal{Z}_f \int \mathcal{D}[c^*, c; f^*, f] \exp\left(-S[c^*, c; f^*, f] + J_1^* c_1 + c_2^* J_2 + L_1^* f_1 + f_2^* L_2\right)$$

$$F[L^*, L] = \ln \tilde{\mathcal{Z}}_f \int \mathcal{D}[f^*, f] \exp\left(-S_{\mathrm{d}}[f^*, f] + L_1^* f_1 + f_2^* L_2\right)$$

Hubbard-Stratanovich transformation:

$$F[J^*, J; L^*, L] = L_1^*[g(\Delta - h)g]_{12} L_2 + \ln \int \mathcal{D}[c^*, c] \exp\Big(- S[c^*, c] + \\ + J_1^* c_1 + c_2^* J_2 + L_1^*[g(\Delta - t)]_{12} c_2 + c_1^*[(\Delta - t)g]_{12} L_2\Big)$$

Relation between Green functions:

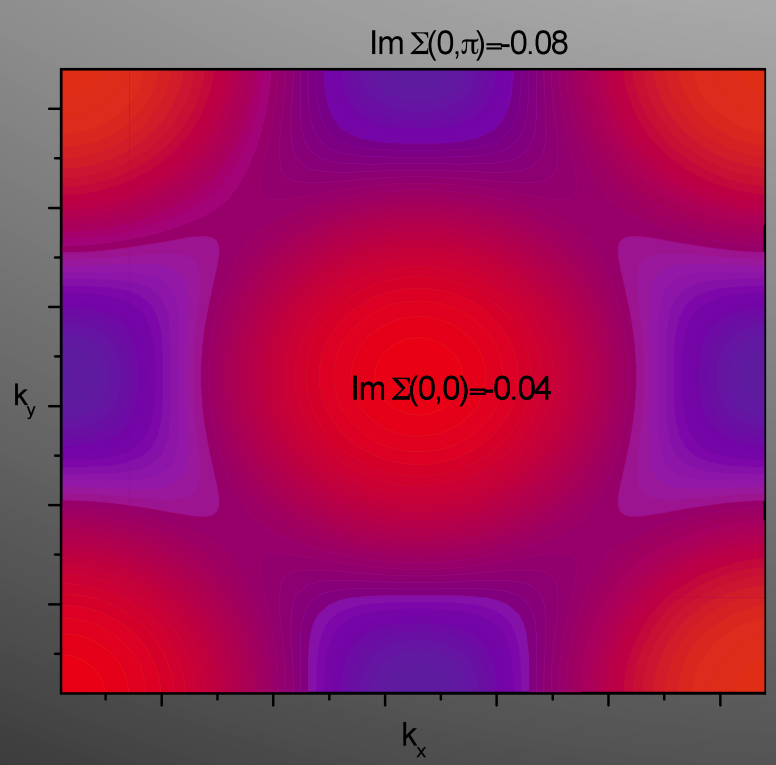
$$\tilde{G}_{12} = -\frac{\delta^2 F}{\delta L_2 \delta L_1^*}\bigg|_{L^*=L=0}$$

$$\tilde{G}_{12} = -[g(\Delta - t)g]_{12} + [g(\Delta - t)]_{11'} G_{1'2'} [(\Delta - t)g]_{2'2}$$

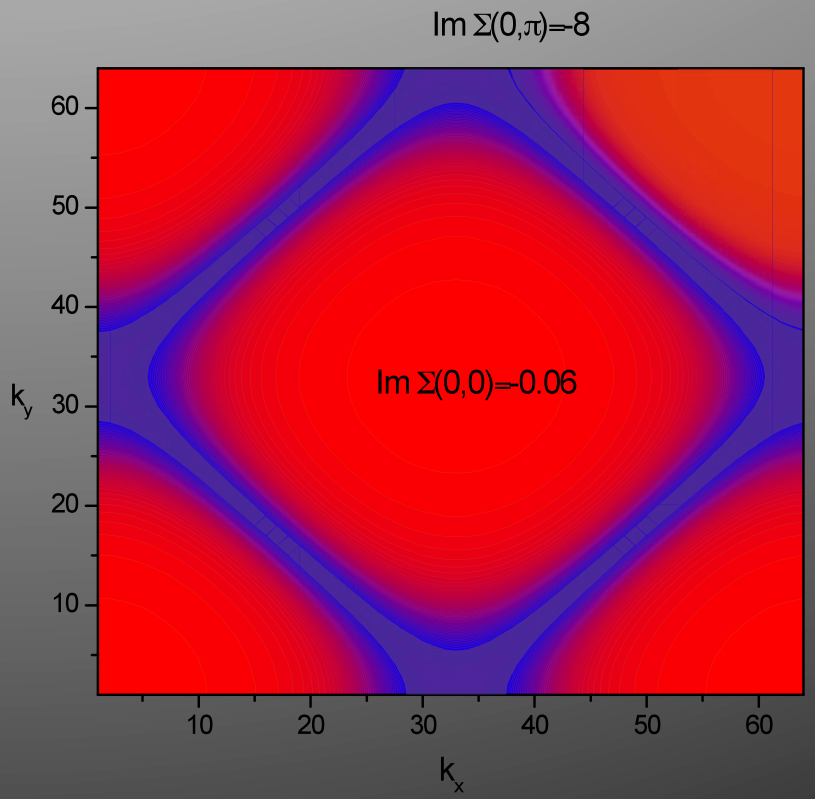
T-matrix like relations via dual self-energy

$$G_\omega(\mathbf{k}) = \left[\left(g_\omega + g_\omega \tilde{\Sigma}_\omega(\mathbf{k}) g_\omega\right)^{-1} + \Delta_\omega - t_k\right]^{-1}$$

ARPES: Im $\Sigma(k, \omega=0)$



U=1



U=2

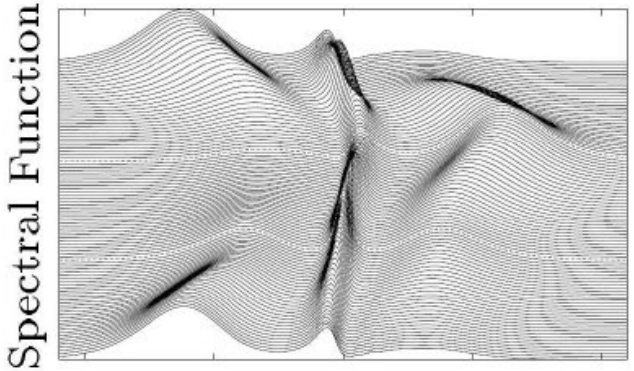
Hubbard model with $8t = 2, \beta = 20$ at half-filling.
Data for Im Σ_k at $\omega = 0$.

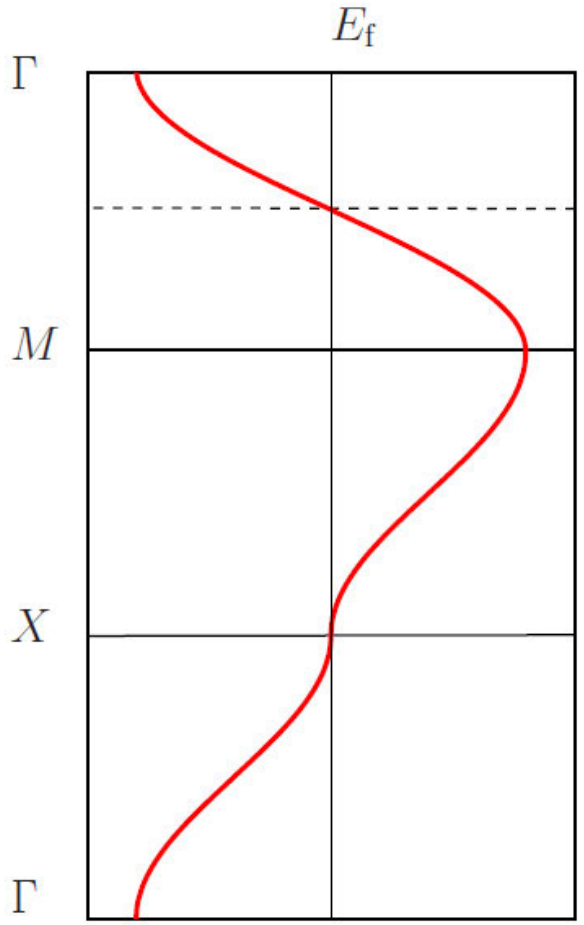
A. Rubtsov, et al, PRB **79**, 045133 (2009)

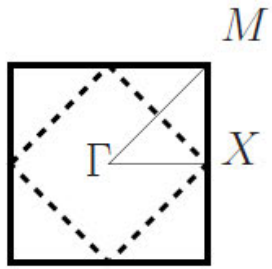
2d-Hubbard: Spectral Function

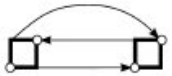
paramagnetic calculation $U/t = 8,\ T/t = 0.235$

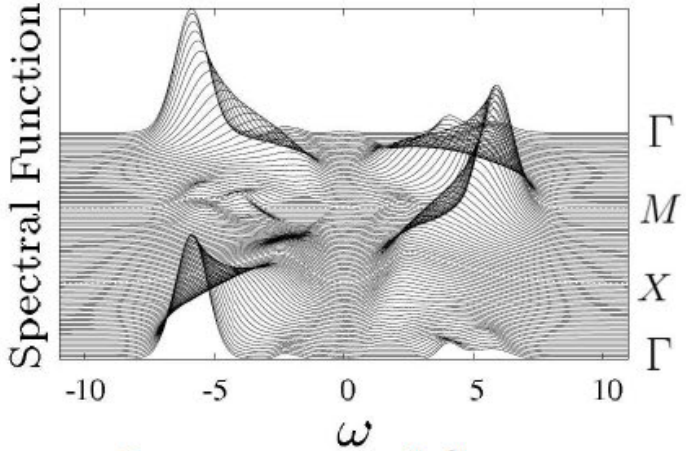
DMFT







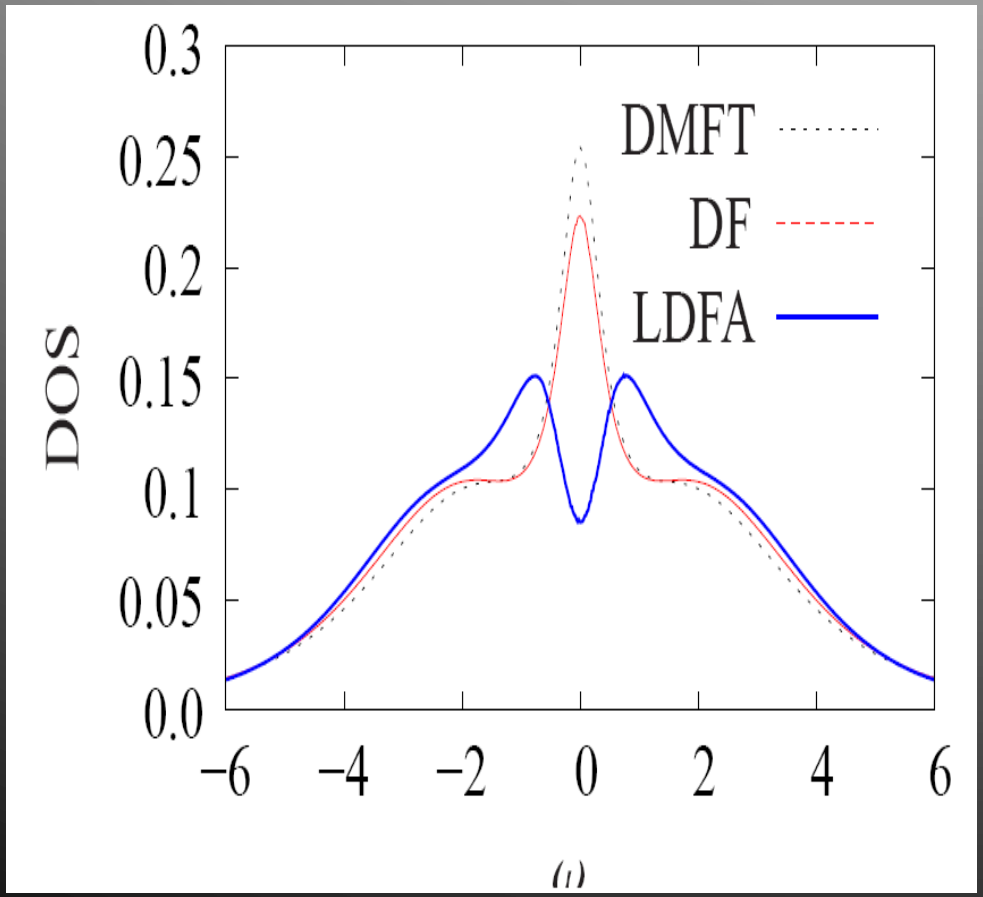
Dual Fermion $\Sigma^d =$ 



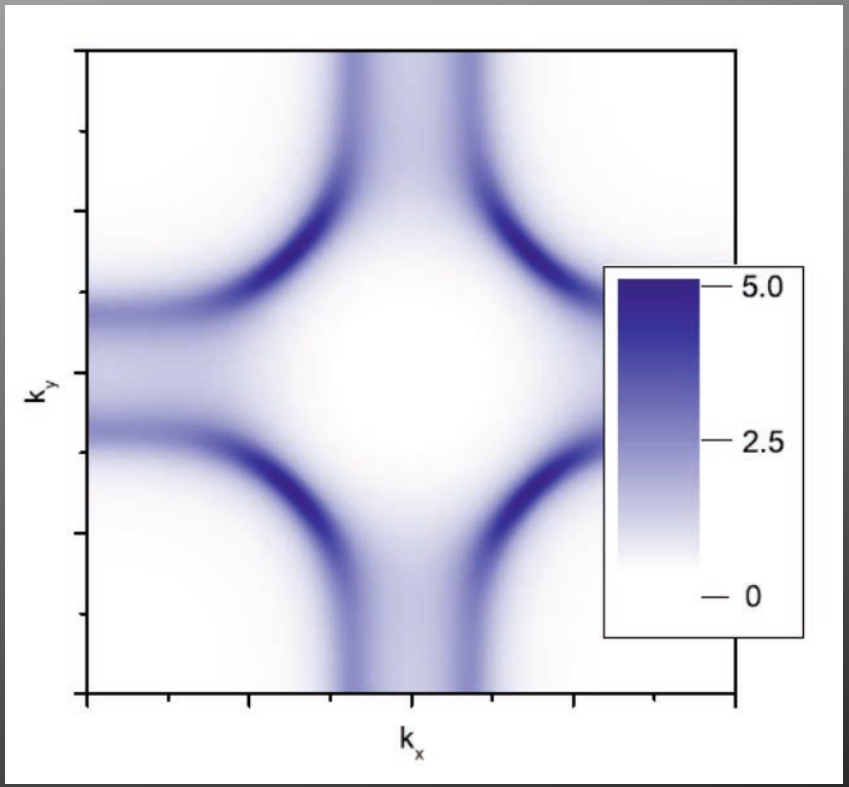
- Strong modifications through AF short-range correlations

Pseudogap in HTSC: dual fermions

$$S[f,f^*] = \sum_{\omega k \sigma} g_\omega^{-2} \left((\Delta_\omega - \epsilon_k)^{-1} + g_\omega \right) f^*_{\omega k \sigma} f_{\omega k \sigma} + \sum_i V_i$$



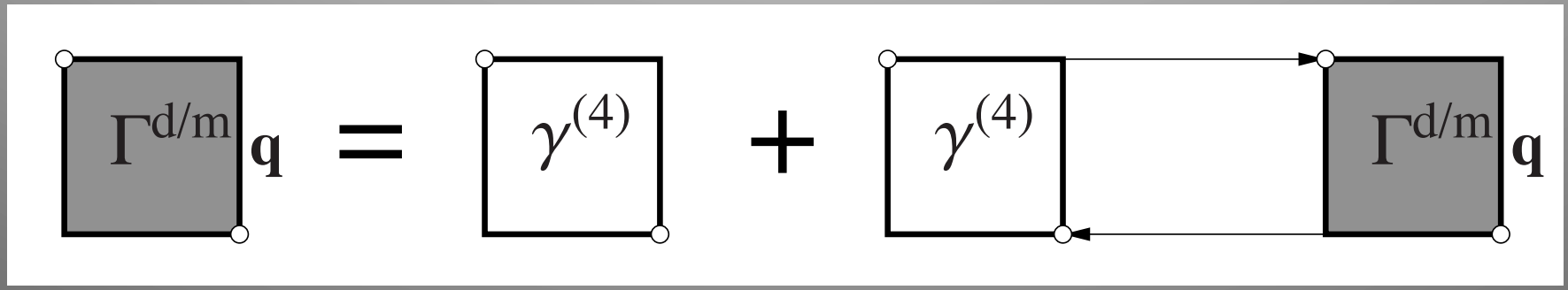
n=1



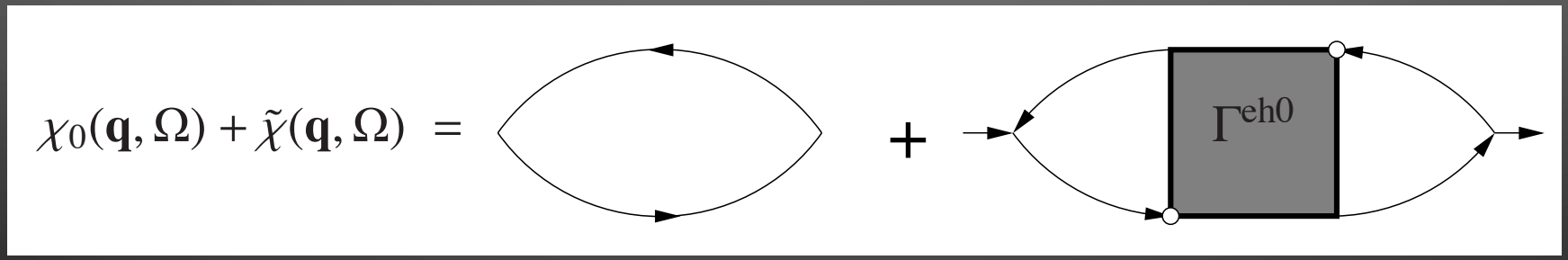
FS, n=0.85

2d: U=W=2

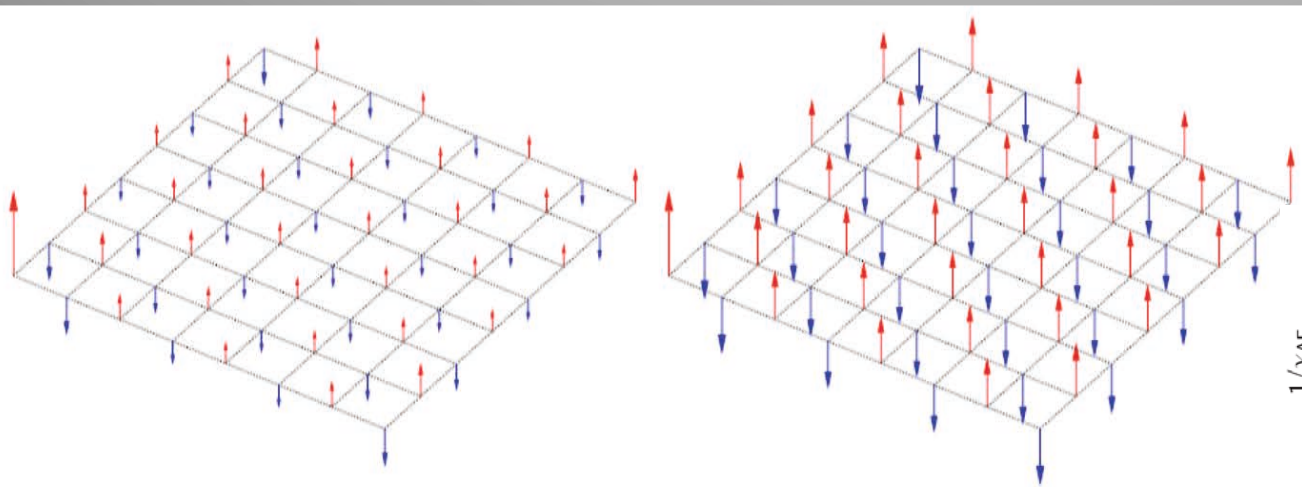
TPGF: Bethe-Salpeter Equations

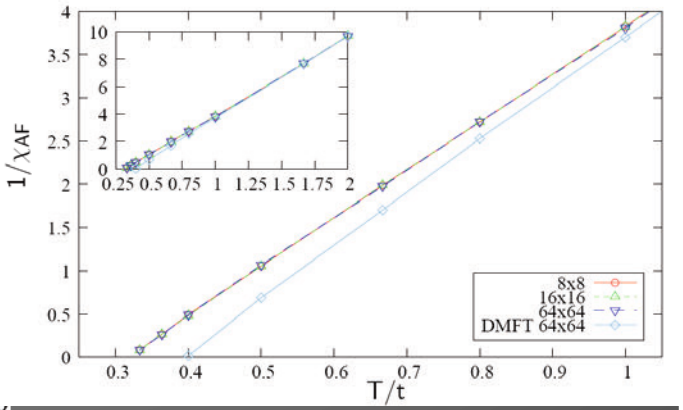


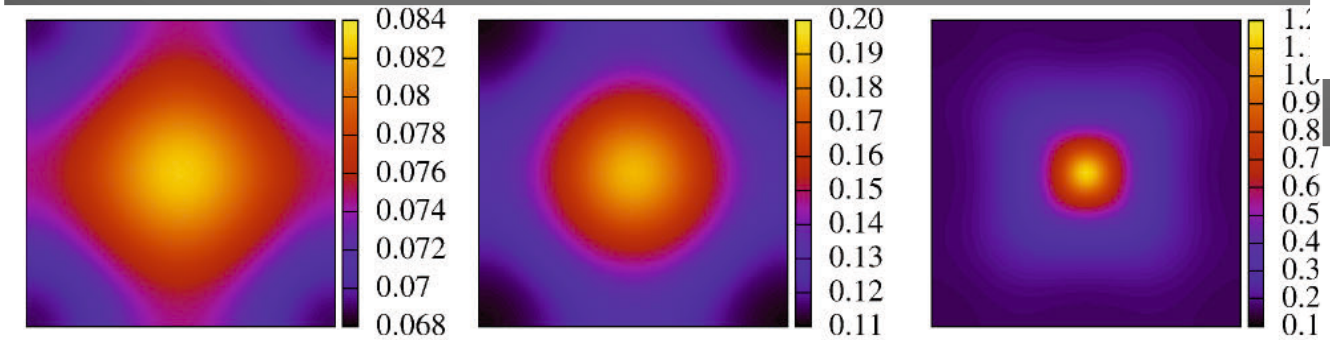
Non-local susceptibility with vertex corrections

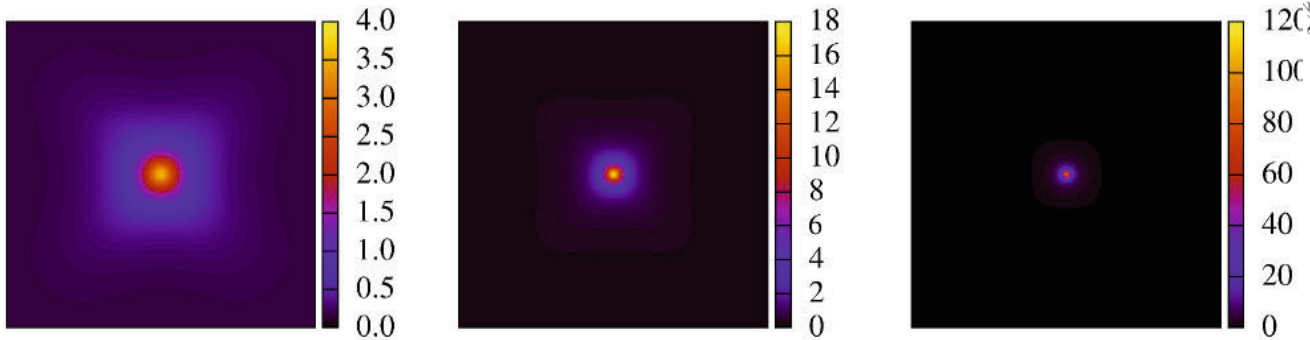


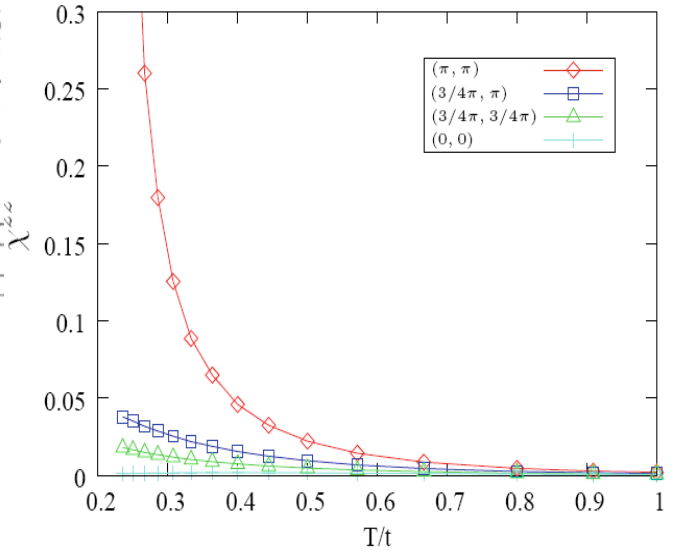
Susceptibility: 2d – Hubbard model











Theory of Correlated Materials

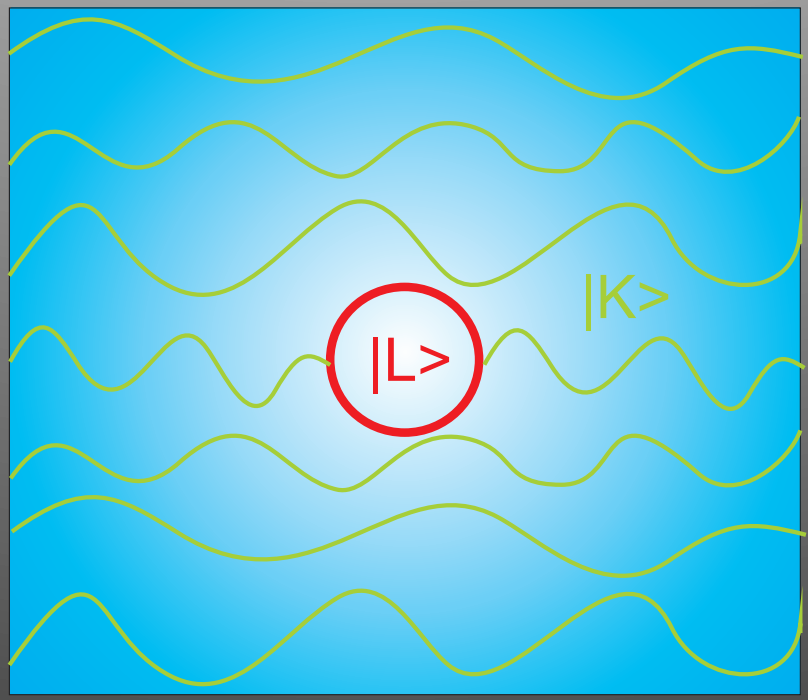
DFT

- Materials-specific
- Fast code packages
- Fails for strong correlations

Models approaches

- Input parameters unknown
- Computationally expensive
- Systematic many-body scheme

Correlated Electrons in Crystals



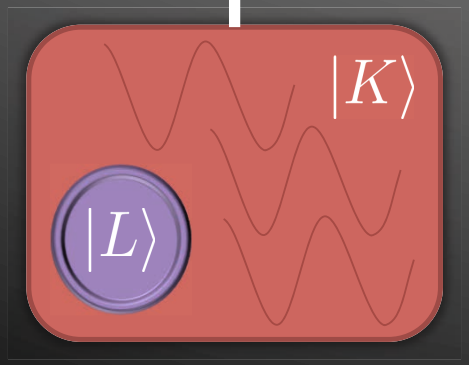
Projector mapping

Idea: Use of projections of DFT wave functions $|K\rangle$ onto localized orbitals $|L\rangle$ to obtain hybridization functions

Local Green function and hybridization function

$$\hat{G}_0^{\mathrm{loc}}(\omega) = \sum_{K,L,L'} |L\rangle \frac{\langle L|K\rangle\langle K|L'\rangle}{\omega + i0^+ - \epsilon_K} \langle L'|$$

$$\hat{\Delta}(\omega) = \omega + i\delta - \epsilon_d - \left[\hat{G}_0^{\mathrm{loc}}(\omega)\right]^{-1}$$



Projector augmented wave basis

$$|K\rangle = |\tilde{K}\rangle + \sum_i (|\phi_i\rangle - |\tilde{\phi}_i\rangle)\langle \tilde{p}_i|\tilde{K}\rangle$$

$$|L\rangle = |\phi_i\rangle$$

$$\langle L|K\rangle = \sum_{\nu'} \langle L|\phi_{\nu'}\rangle\langle \tilde{p}_{\nu'}|\tilde{K}\rangle$$

Implementation with PP, PAW code

F. Lechermann et l, PRB **77**, 205112 (2008), PRB **81**, 085413 (2010),

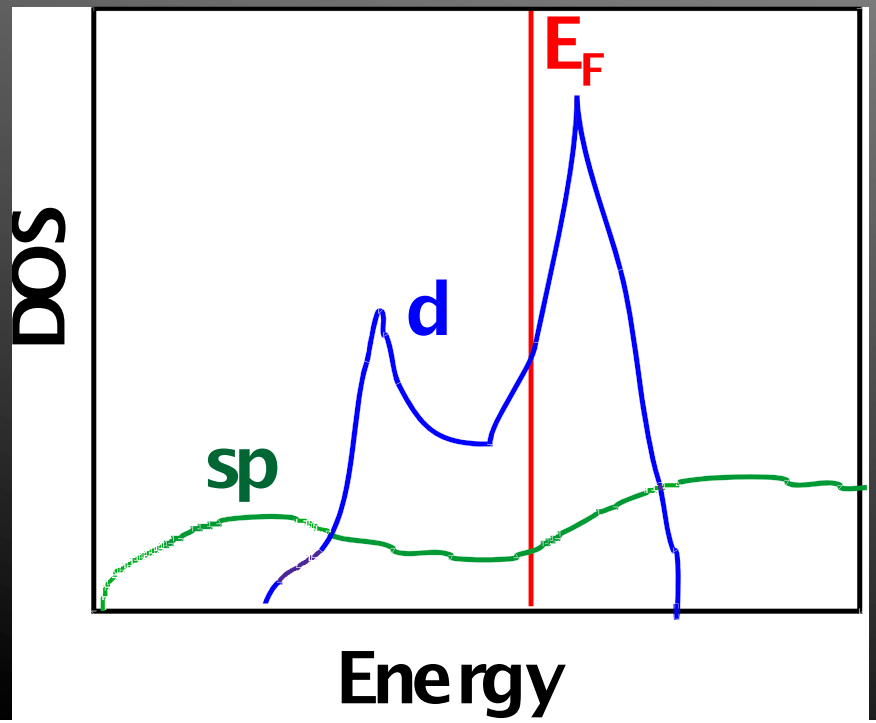
DMFT in local L(N)MTO basis

LDA+DMFT (orthogonal |L> basis):

$$G^{-1}_{LL'}(\vec{k}, i\omega_n) = i\omega_n + \mu - H^{LDA}_{LL'}(\vec{k}) - \Sigma^{DMFT}_{LL'}(i\omega_n)$$

$$G_{LL'}(i\omega_n) = \sum_{\vec{k}\in BZ} G^{-1}_{LL'}(\vec{k}, i\omega_n)$$

$$\hat{G}(i\omega_n) = \sum_{\alpha\in O_h} \hat{U}(\alpha) \sum_{\vec{k}\in IBZ} \hat{G}(\vec{k}, i\omega_n) \hat{U}^{+}(\alpha)$$



Correlated d-states:

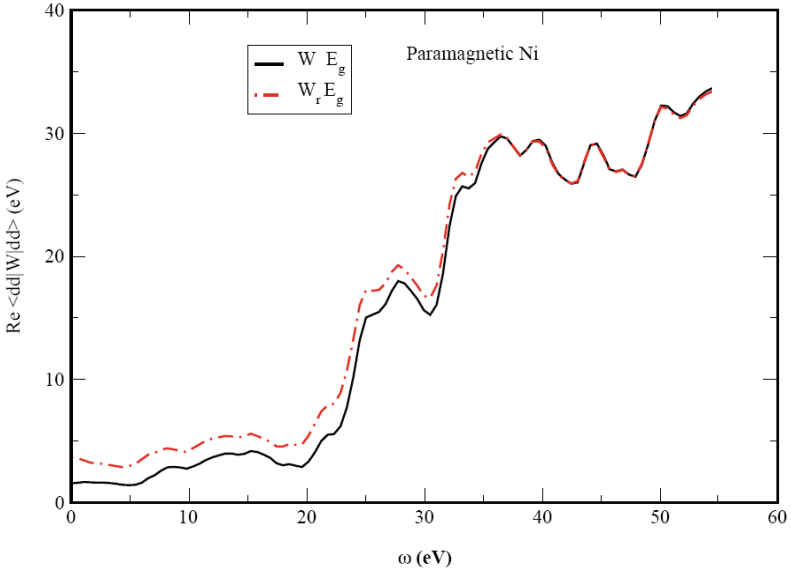
$$\hat{H}(\vec{k}) + \hat{\Sigma}(\omega) = \begin{pmatrix} H_{ss} & H_{ps} & H_{ds} \\ H_{sp} & H_{pp} & H_{dp} \\ H_{sd} & H_{pd} & H_{dd} + \Sigma_{dd} \end{pmatrix}$$

Constrain GW calculations of U

$$P(\mathbf{r},\mathbf{r}';\omega) = \sum_{i}^{occ}\sum_{j}^{unocc} \psi_i(\mathbf{r})\psi_i^*(\mathbf{r}')\psi_j^*(\mathbf{r})\psi_j(\mathbf{r}') \times \left\{ \frac{1}{\omega - \varepsilon_j + \varepsilon_i + i0^+} - \frac{1}{\omega + \varepsilon_j - \varepsilon_i - i0^+} \right\}$$

F. Aryasetiawanan et al
PRB(2004)

$$W_r(\omega) = [1 - vP_r(\omega)]^{-1} v$$



$$W = [1 - vP]^{-1} v$$

$$= [1 - vP_r - vP_d]^{-1} v$$

$$= [(1 - vP_r)\{1 - (1 - vP_r)^{-1} vP_d\}]^{-1} v$$

$$= \{1 - (1 - vP_r)^{-1} vP_d\}^{-1} (1 - vP_r)^{-1} v$$

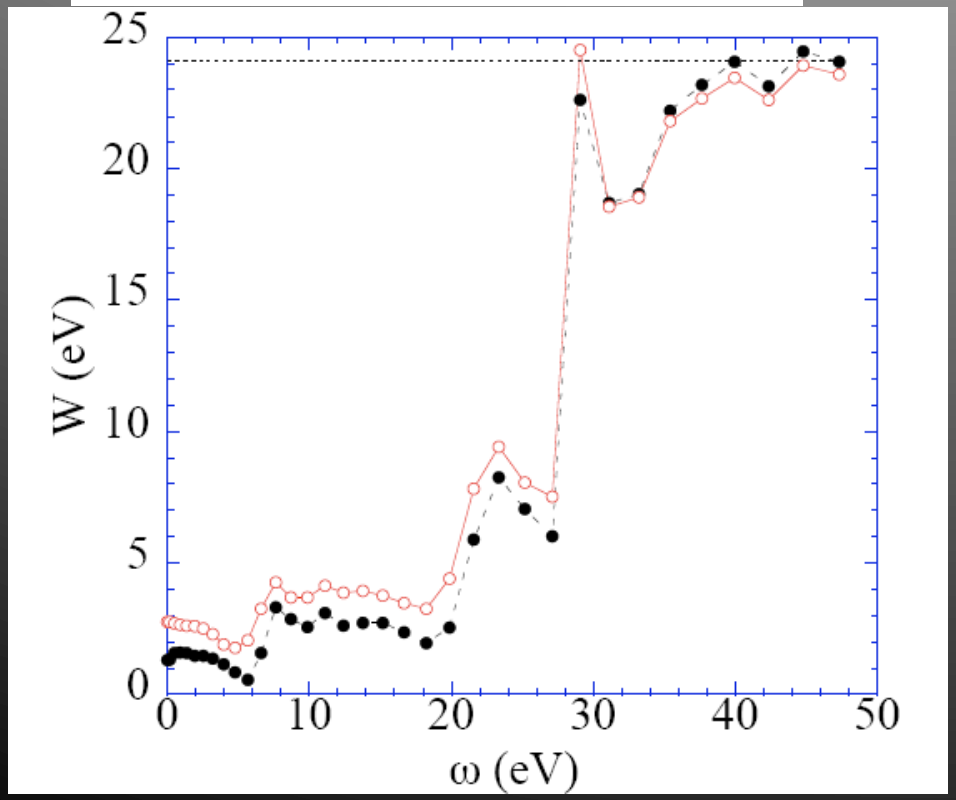
$$= [1 - W_r P_d]^{-1} W_r$$

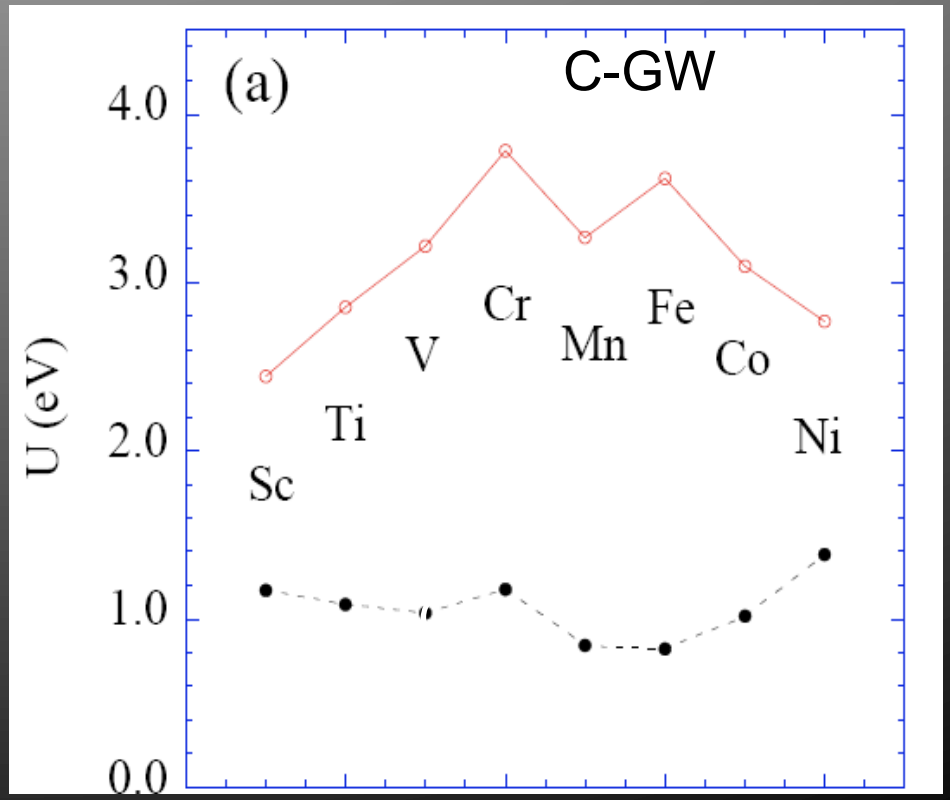
Wannier - GW and effective U(ω)

$$|\varphi_{n\mathbf{R}}\rangle = \frac{V}{(2\pi)^3}\int e^{-i\mathbf{k}\cdot\mathbf{R}}|\psi^{(\mathrm{w})}_{n\mathbf{k}}\rangle d^3k$$

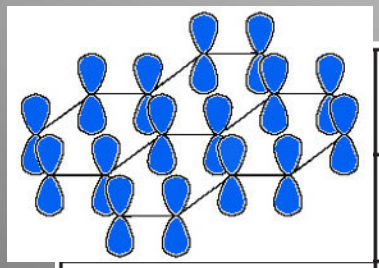
$$|\psi^{(\mathrm{w})}_{n\mathbf{k}}\rangle = \sum_m \mathcal{U}_{mn}(\mathbf{k})|\psi_{m\mathbf{k}}\rangle$$

T. Miyake and F. Aryasetiawan
Phys. Rev. B 77, 085122 (2008)



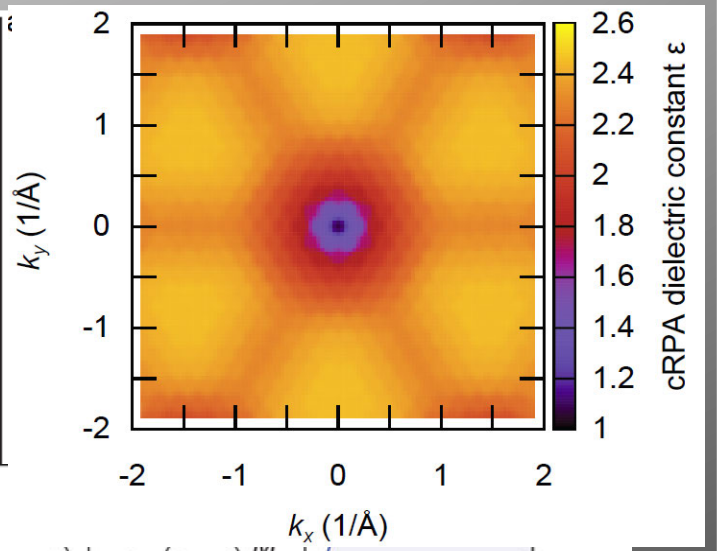


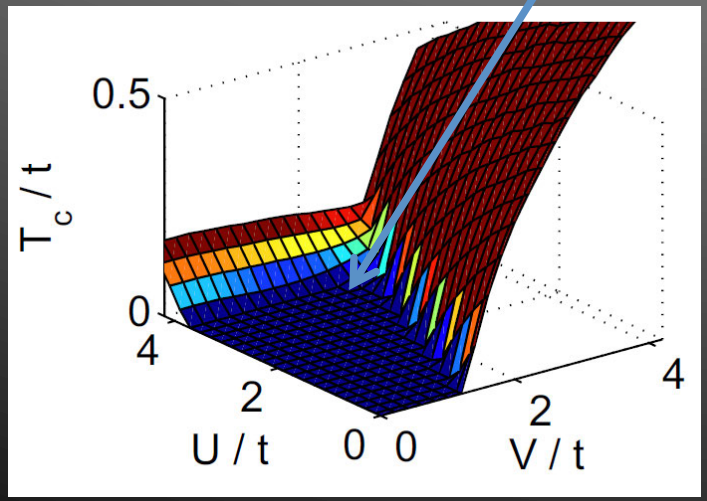
Strength of Coulomb interactions: Graphene



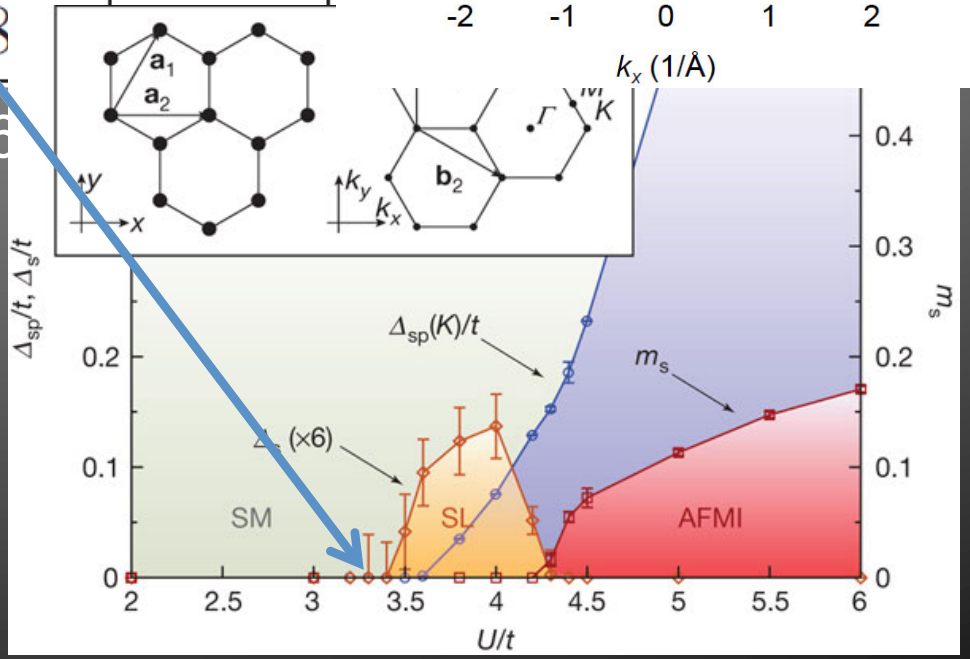
	graphene		graphite	
	bare	cRPA	bare	cRPA
U_{00}^{A} (eV)	17.0	9.3	17.5	8.0
U_{00}^{B} (eV)	17.0	9.3	17.7	8.1
U_{01} (eV)	8.5	5.5	8	

T. Wehling et al., PRL **106**, 236





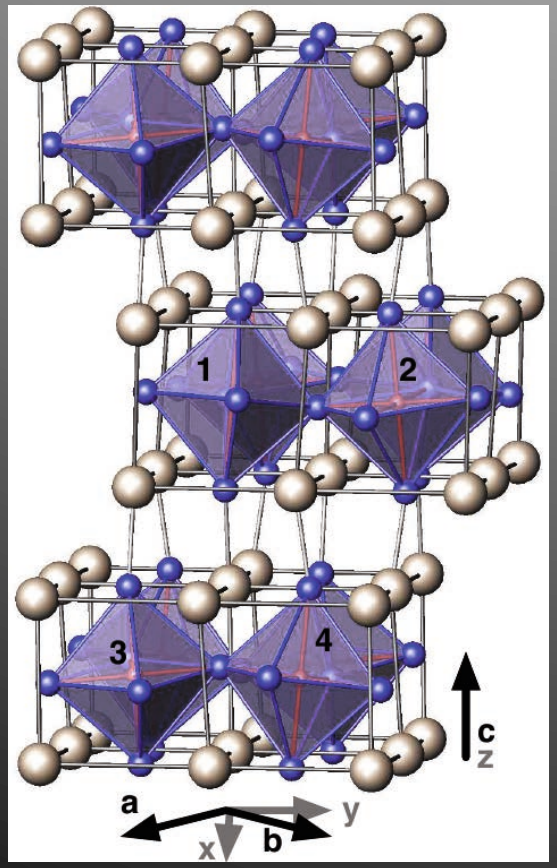
C. Honerkamp, PRL 100, 146404 (2008)

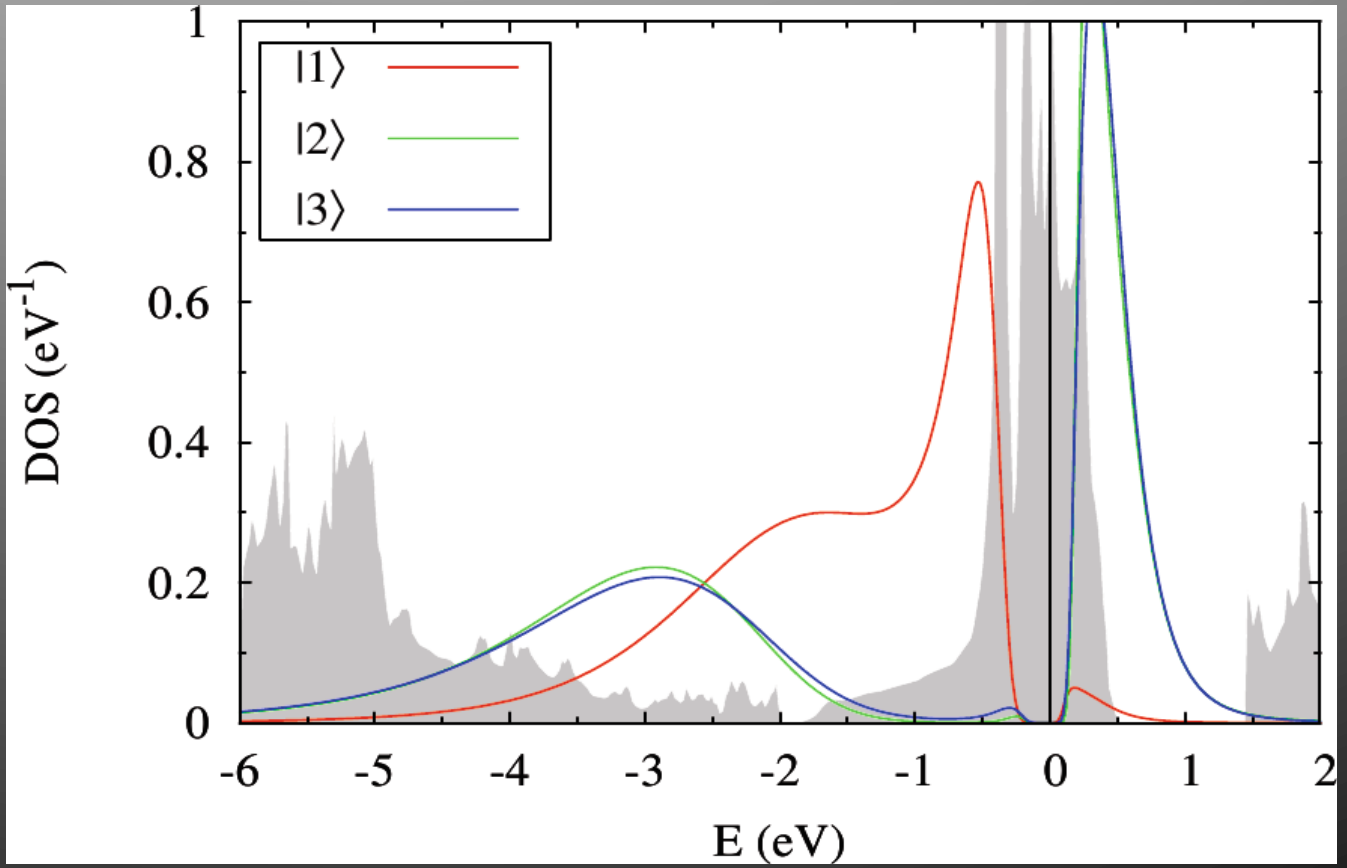


Z. Y. Meng et al., Nature **464**, 847-851 (2010)

Mott Transition in Ca_2RuO_4

- DFT (Grey) vs DFT+DMFT:

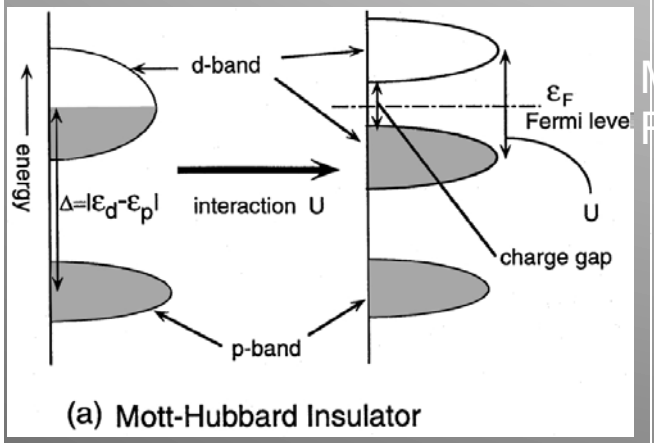




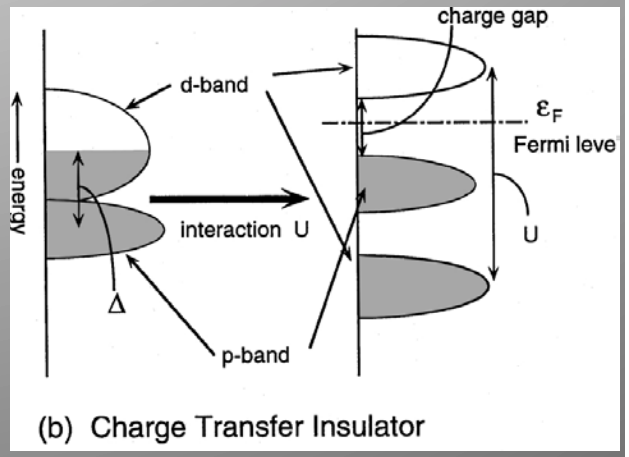
E. Gorelov et al, PRL104, 226401 (2010)

$$|1\rangle \sim d_{xy}, |2,3\rangle \sim d_{xz,yz}$$

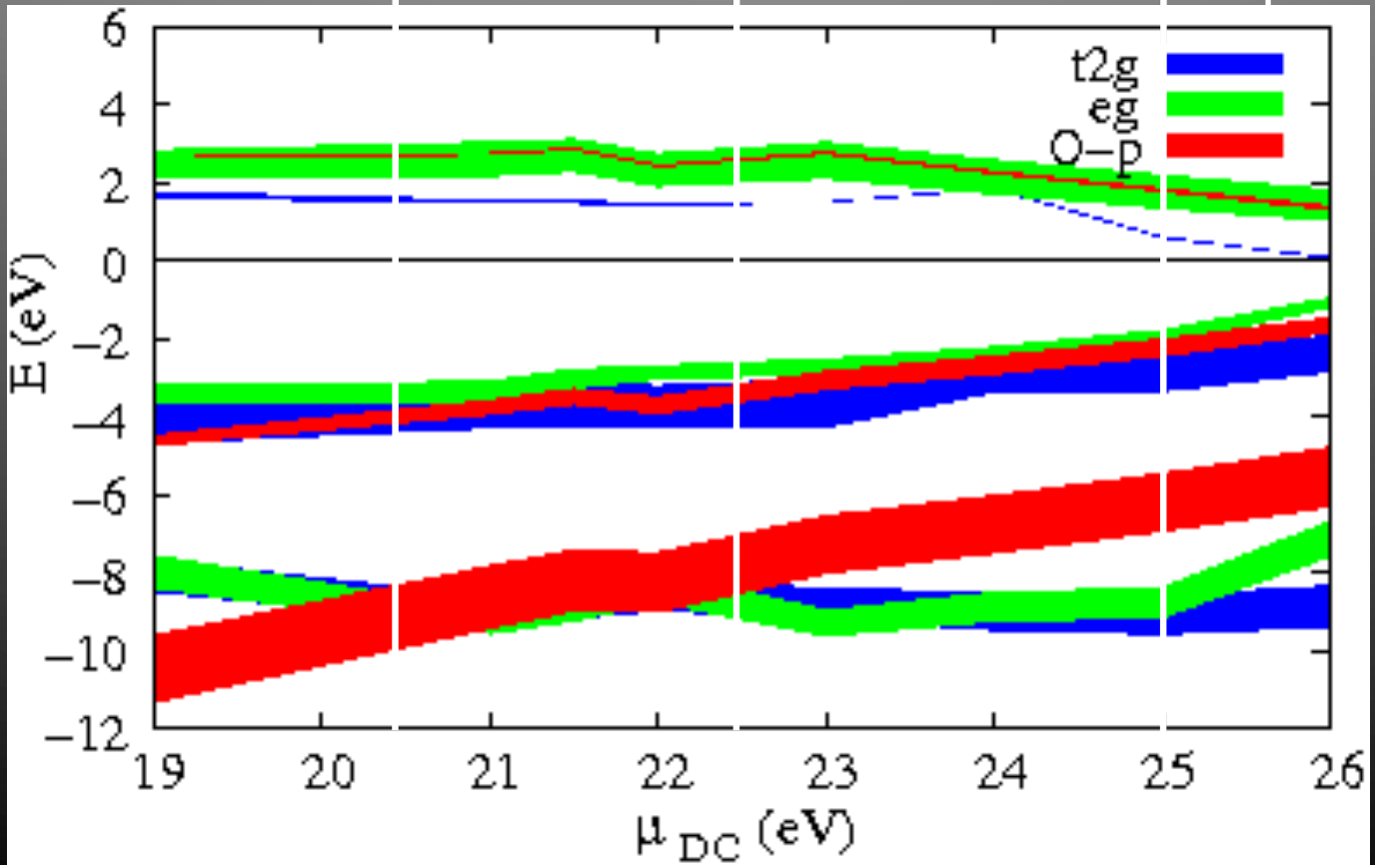
DC- NiO: peak positions and spectral weights



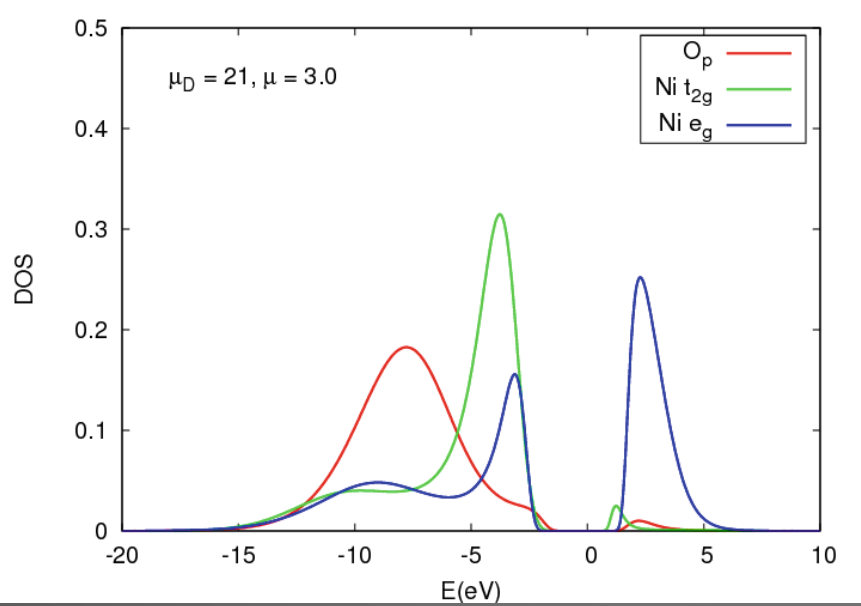
M. Imada, A. Fujimori, and Y. Tokura
Rev. Mod. Phys. 70, 1039 (1998).

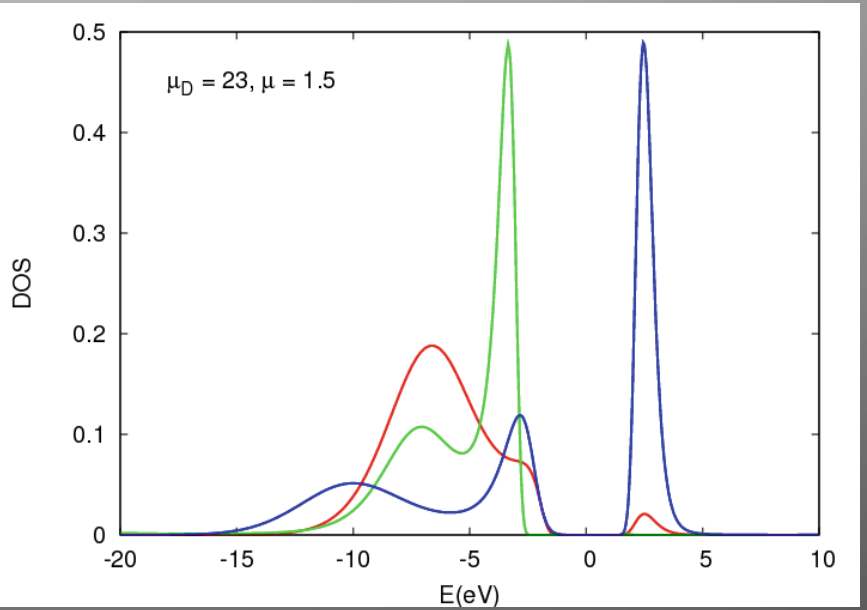


AMF FLL exp



Spectral functions and double counting





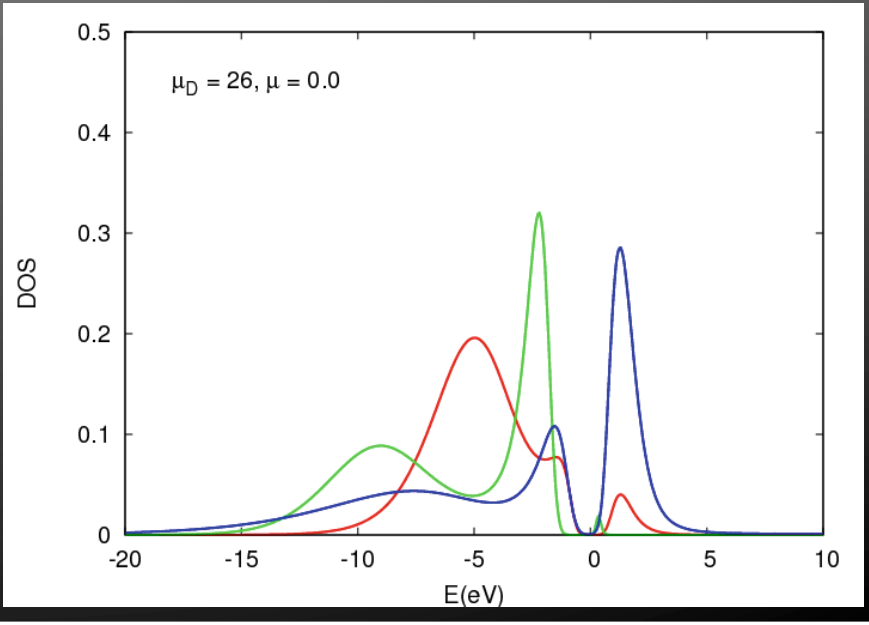
Mott insulator

$\mu_{DC} = 21\text{eV}$

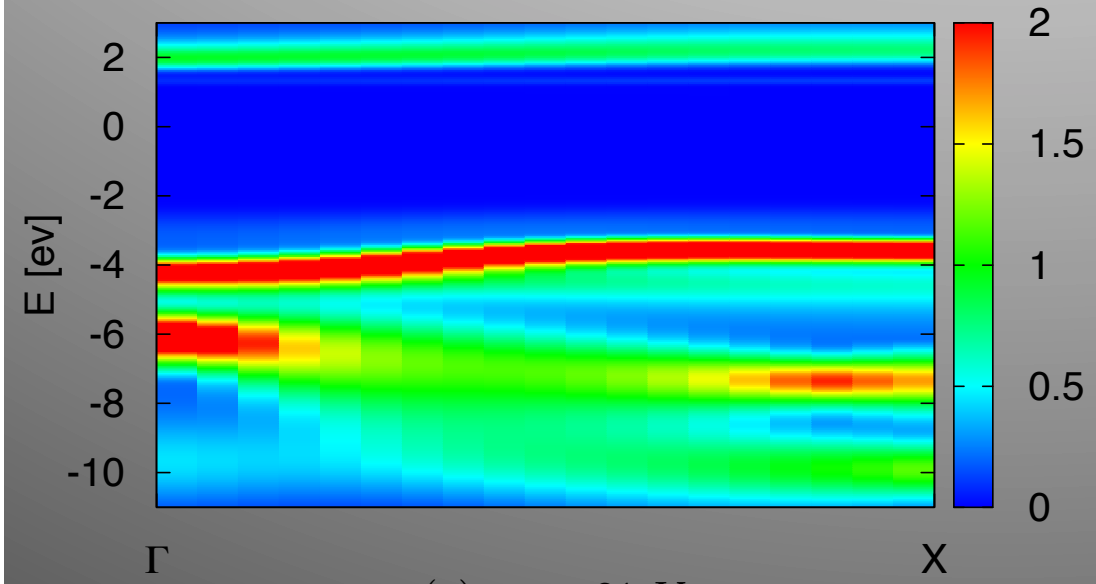
Charge transfer insulator

Almost metallic

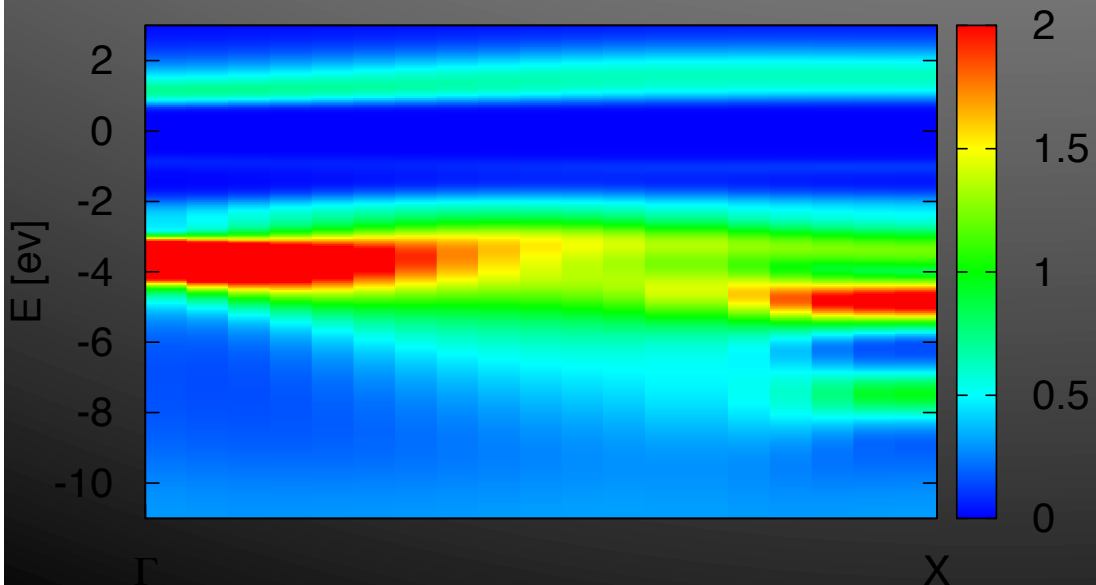
$\mu_{DC} \geq 26\text{eV}$



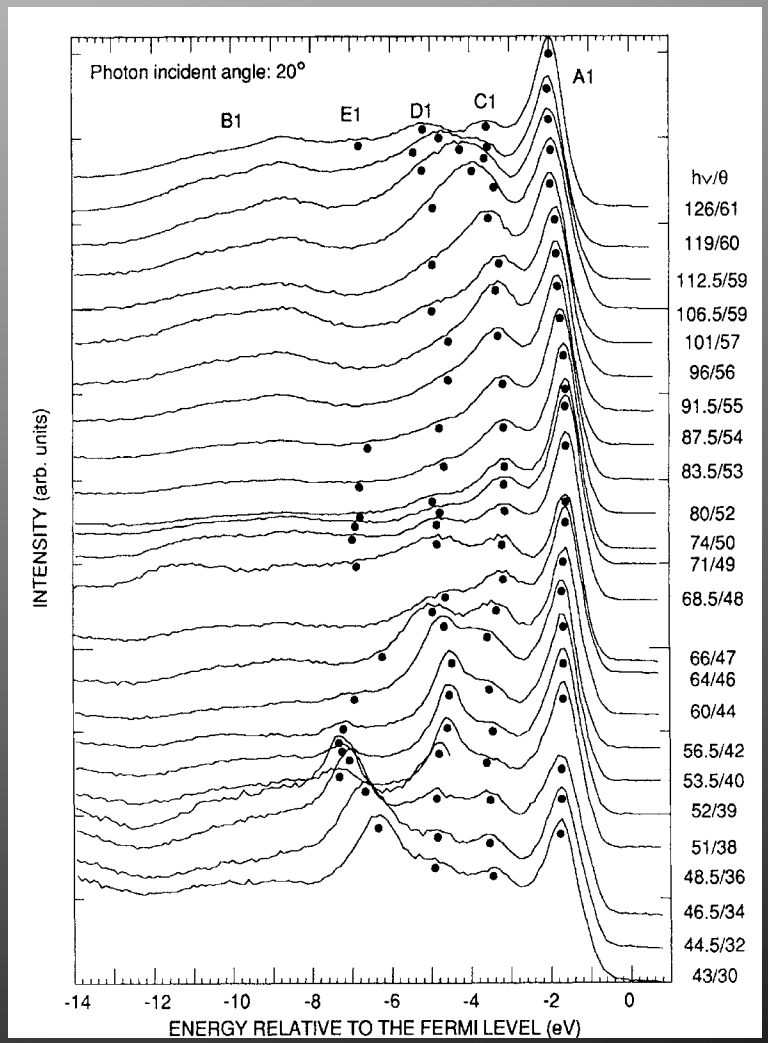
NiO: Spectral Function



$(a)\mu_{dc} = 21\text{eV}$



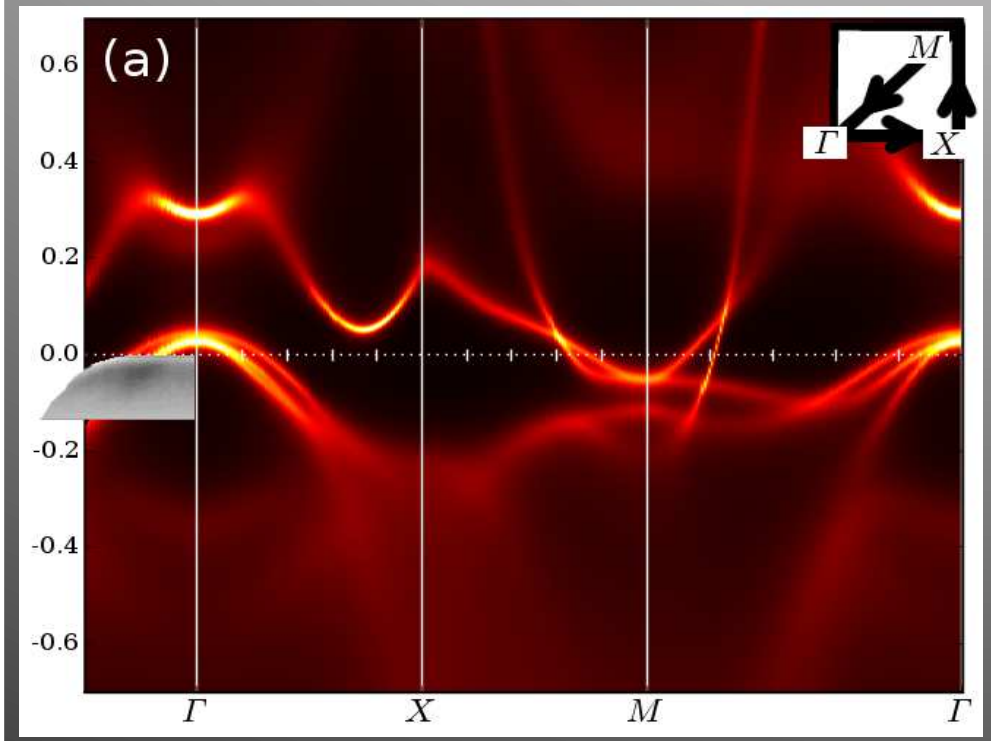
$(b)\mu_{dc} = 25\text{eV}$

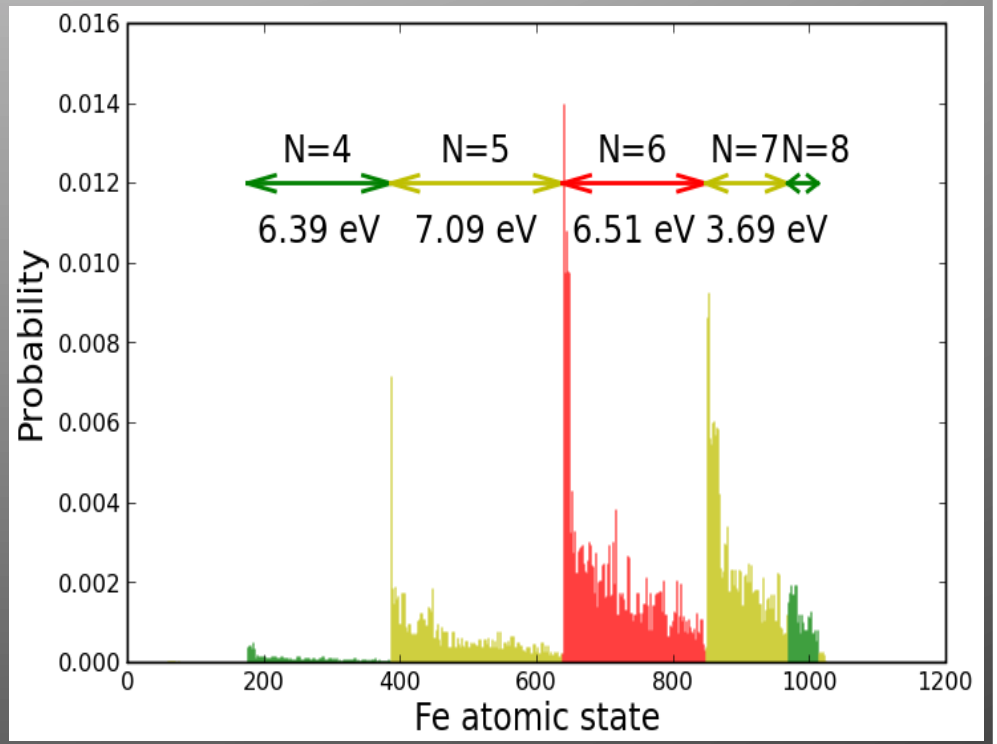


Z.X. Shen,et al PRB (1991)

CT-QMC for Multiorbital system: Fe pnictides

Full rotational-invariant U matrix with 5-orbitals LDA+DMFT calculations





Spectral function vs. ARPES
A. Kutepov et al PRB (2010)

$BaFe_2As_2$ Occupation probabilities

Superconductivity related with non-local spin-fluctuations:
How to calculate non-local correlations effects - EDMFT?
BUT: there is a problem with conservation low:
NEED: more elaborate “non-local” scheme!

$$\Omega^2 \langle \rho\rho \rangle_{\Omega K} = K^2 \langle jj \rangle_{\Omega K}$$

$$\langle \rho\rho \rangle_{\omega, K\to 0} \propto \frac{K^2}{\Omega^2 + \Omega_p^2}$$

Summary

- DFT+DMFT scheme is the simplest way to treat strongly correlated materials
- CT-QMC is numerically exact and very useful Quantum Impurity Solver

  (Lecture of A. Rubtsov)
- DF is an efficient scheme to describe long-range non-local correlation effects in solids