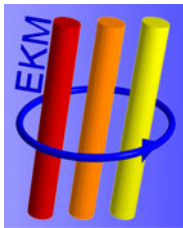


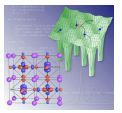
Center for
Electronic Correlations and Magnetism
University of Augsburg

Dynamical Mean-Field Approach for Strongly Correlated Materials

Dieter Vollhardt

Autumn School 2011: Hands-on LDA+DMFT
Forschungszentrum Jülich
October 4, 2011

Supported by DFG FOR 1346



Outline:

- Electronic correlations
- Dynamical Mean-Field Theory (DMFT)
- Applications of DMFT to models and materials

Correlations

Correlation [lat.]: *con* + *relatio* (*"with relation"*)

Correlations in mathematics, natural sciences:

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

e.g., densities:

$$\langle \rho(\mathbf{r}) \rho(\mathbf{r}') \rangle \neq \langle \rho(\mathbf{r}) \rangle \langle \rho(\mathbf{r}') \rangle$$

Correlations (I):
Effects beyond factorization approximations (e.g., Hartree-Fock)

Temporal/spatial correlations in everyday life

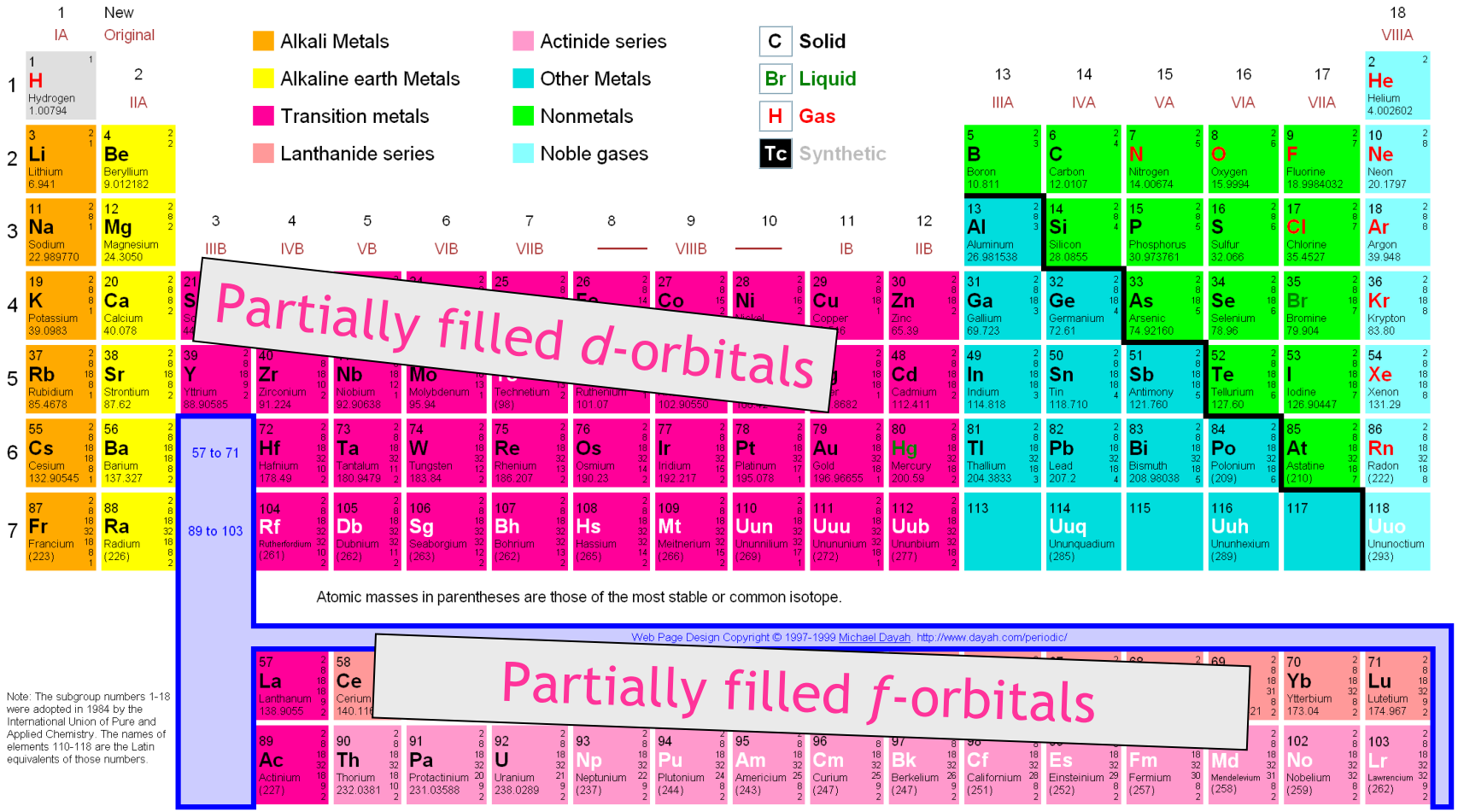
Beware: External periodic potential → long-range order enforced
→ trivial correlations

Temporal/spatial correlations in everyday life

Time/space average inappropriate

Electronic Correlations in the Periodic Table

Periodic Table of the Elements

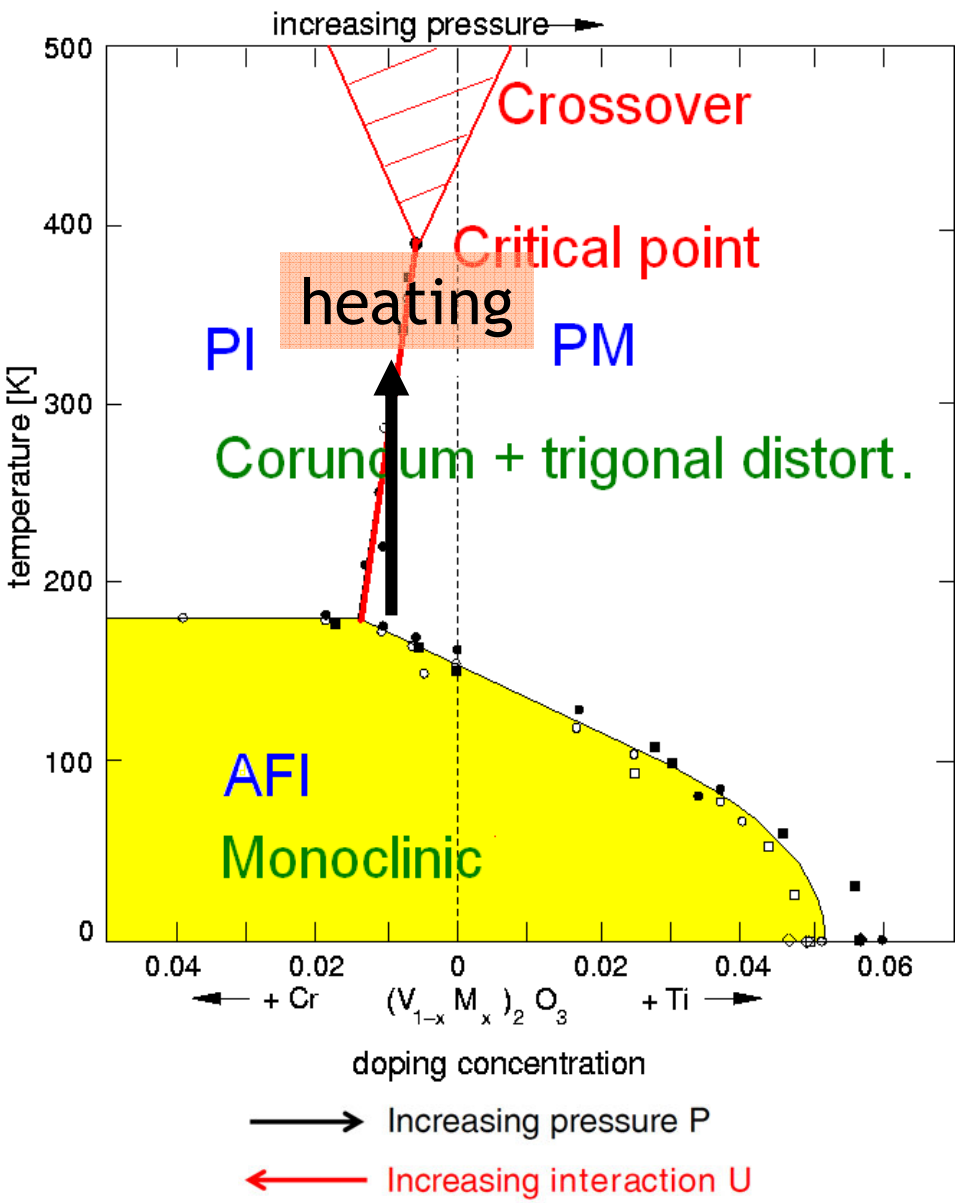


Narrow *d*,*f*-orbitals → strong electronic correlations

Electronic Correlations in Solids

1.

Mott metal-insulator transition in V_2O_3



Rice, McWhan (1970); McWhan, Menth, Remeika, Brinkman, Rice (1973)

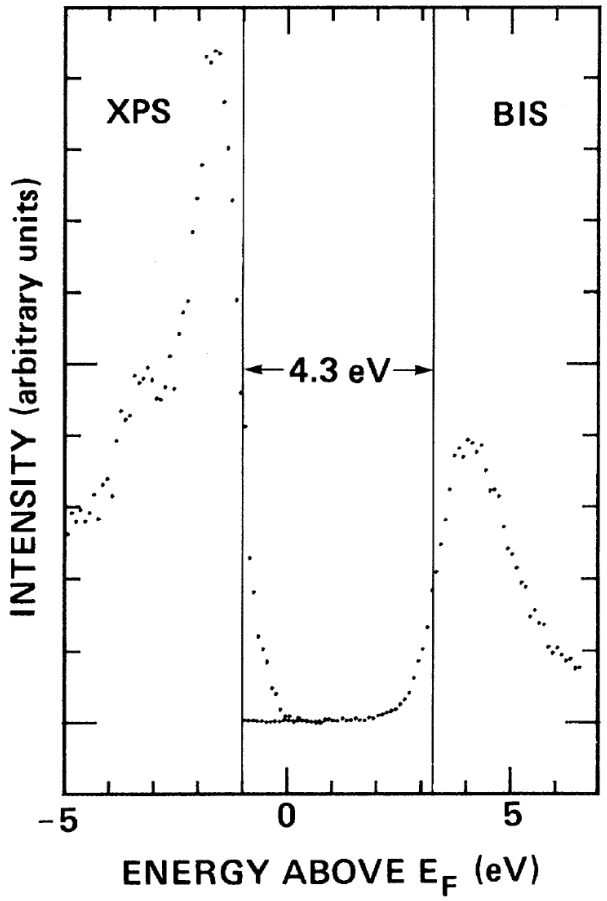
- PI ⟷ PM: 1. order transition without lattice symmetry change
- Anomalous slope of P(T)

  → *Pomeranchuk effect* in ^{3}He

Microscopic explanation?

2.

Photoemission spectra of NiO

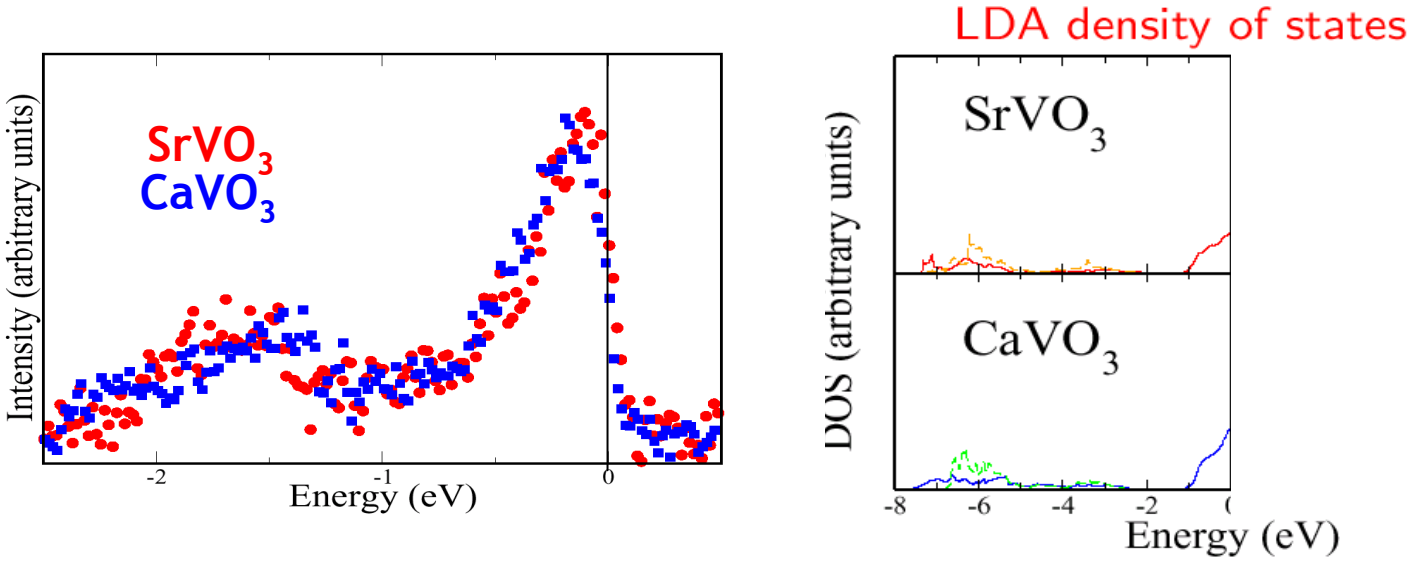


Sawatzky, Allen (1984)

Origin of gap (antiferromagnetism)?

3.

Photoemission spectra of $(Sr,Ca)VO_3$



Osaka – Augsburg – Ekaterinburg collaboration: Sekiyama *et al.*, 2004

Reason for shift of spectral weight?

Correlated electron materials

Fascinating topics for fundamental research

- large resistivity changes
- gigantic volume changes
- high-T_c superconductivity
- strong thermoelectric response
- colossal magnetoresistance
- huge multiferroic effects

} large susceptibilities

with

Technological applications:

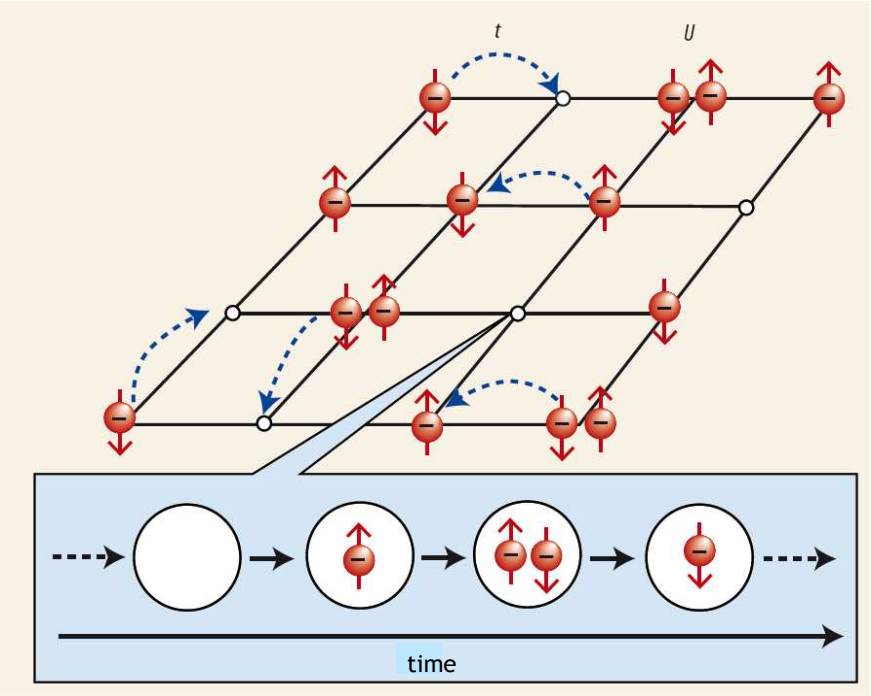
- sensors, switches
- magnetic storage
- refrigerators
- functional materials, ...

Electronic Correlations: Models

→ Lecture of F. Lechermann

Hubbard model

(tight binding approach)



Gutzwiller, 1963
Hubbard, 1963
Kanamori, 1963

$$H = -t \sum_{\langle \mathbf{i},\mathbf{j} \rangle,\sigma} c^{\dagger}_{\mathbf{i}\sigma} c_{\mathbf{j}\sigma} + U \sum_{\mathbf{i}} n_{\mathbf{i}\uparrow} n_{\mathbf{i}\downarrow}$$

$$\langle n_{\mathbf{i}\uparrow} n_{\mathbf{i}\downarrow} \rangle \neq \langle n_{\mathbf{i}\uparrow} \rangle \langle n_{\mathbf{i}\downarrow} \rangle$$

Purely numerical approaches (d=2,3): hopeless

Theoretical challenge:
Construct reliable, comprehensive
non-perturbative approximation scheme

Static (Hartree-Fock-type)
mean-field theories
generally insufficient


WANTED
DEAD OR ALIVE
AND
Reliable
approximation scheme
for
correlated
models and materials
PROFESSORSHIP
REWARD $10,000

Dynamical Mean-Field Theory (DMFT) of Correlated Electrons

What is a “mean-field theory (MFT)“ ?

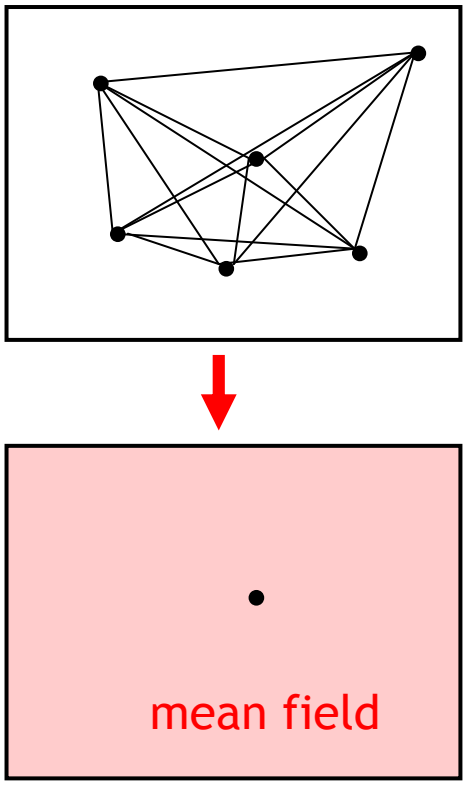
1) Construction by factorization

$$\langle AB \rangle \rightarrow \langle A \rangle \langle B \rangle$$

e.g., spins:

$$\langle S_i S_j \rangle \rightarrow \langle S_i \rangle \langle S_j \rangle$$

→ Weiss MFT



What is a “mean-field theory (MFT)“ ?

2) Construction by exaggeration

For example:

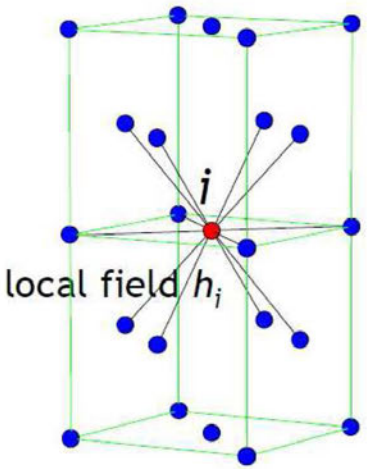
Spin S

Degeneracy N

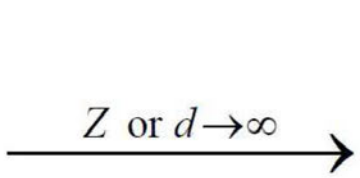
Dimension d /coordination number Z

} → ∞

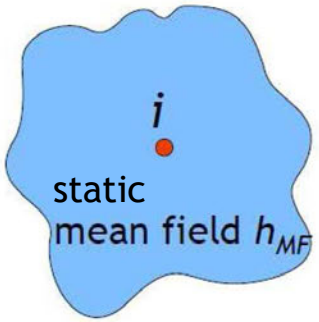
Spin models:



fcc-lattice (d=3): Z=12



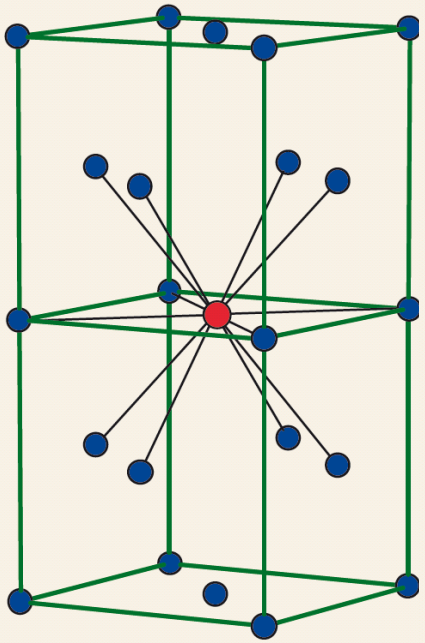
exaggeration → factorization



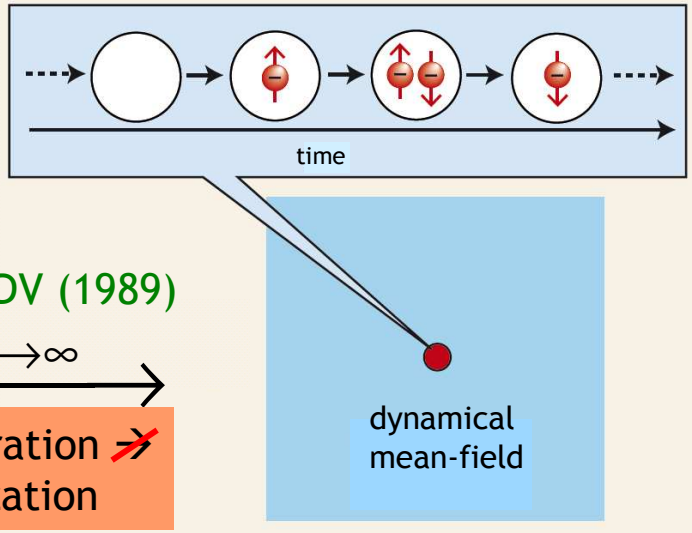
Theory of correlated electrons

$$H = -t \sum_{\langle \mathbf{i},\mathbf{j}\rangle,\sigma} c^{\dagger}_{\mathbf{i}\sigma} c_{\mathbf{j}\sigma} + U \sum_{\mathbf{i}} n_{\mathbf{i}\uparrow} n_{\mathbf{i}\downarrow}$$

Hubbard model



Z=12



Metzner, DV (1989)

$d, Z \to \infty$

exaggeration ~~→~~
factorization

Effective dynamical single-site problem

Müller-Hartmann (1989); Brandt, Mielsch (1989)

Hubbard model: Simplifications for d,Z $\to \infty$

Metzner, DV (1989)

$$\langle H_{kin} \rangle_0 = \underbrace{-t}_{\propto \frac{1}{\sqrt{Z}}} \sum_{\mathbf{i},\sigma} \underbrace{\sum_{\mathbf{j}(NN\ \mathbf{i})}}_{Z} \underbrace{\langle c^{\dagger}_{\mathbf{i}\sigma} c_{\mathbf{j}\sigma} \rangle_0}_{\propto \frac{1}{\sqrt{Z}}}$$

Z or d$\to\infty$

Quantum rescaling $\quad t = \frac{t^*}{\sqrt{Z}}$

Amplitude for hopping j → NN i

$$\left|\text{Amplitude for hopping j} \to \text{NN i}\right|^2 = \text{Probability for hopping j} \to \text{NN i} = \frac{1}{Z}$$

Hubbard model: Simplifications for d,Z → ∞

Metzner, DV (1989)

$$\langle H_{kin} \rangle_0 = \underbrace{-t}_{\propto \frac{1}{\sqrt{Z}}} \sum_{\mathbf{i},\sigma} \underbrace{\sum_{\mathbf{j}(NN\ \mathbf{i})}}_{Z} \underbrace{\langle c^{\dagger}_{\mathbf{i}\sigma} c_{\mathbf{j}\sigma} \rangle_0}_{\propto \frac{1}{\sqrt{Z}}}$$

Z or d→∞

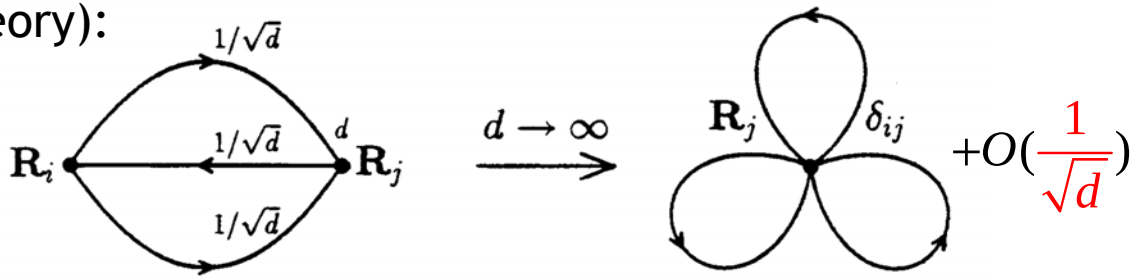
Quantum rescaling $t = \frac{t^*}{\sqrt{Z}}$

Z or $d \to \infty$ → Collapse of irreducible diagrams in position space

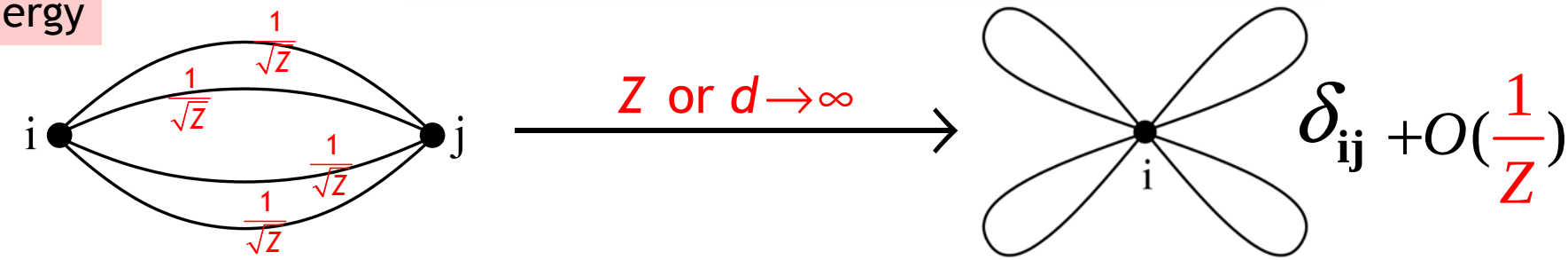
→ great simplifications

Examples (2. order pert. theory):

Self-energy



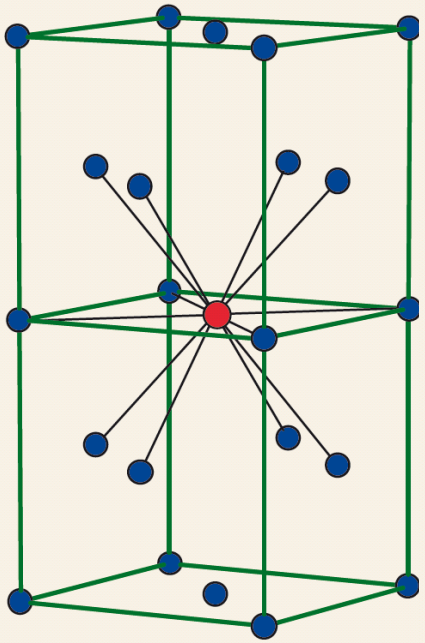
Energy



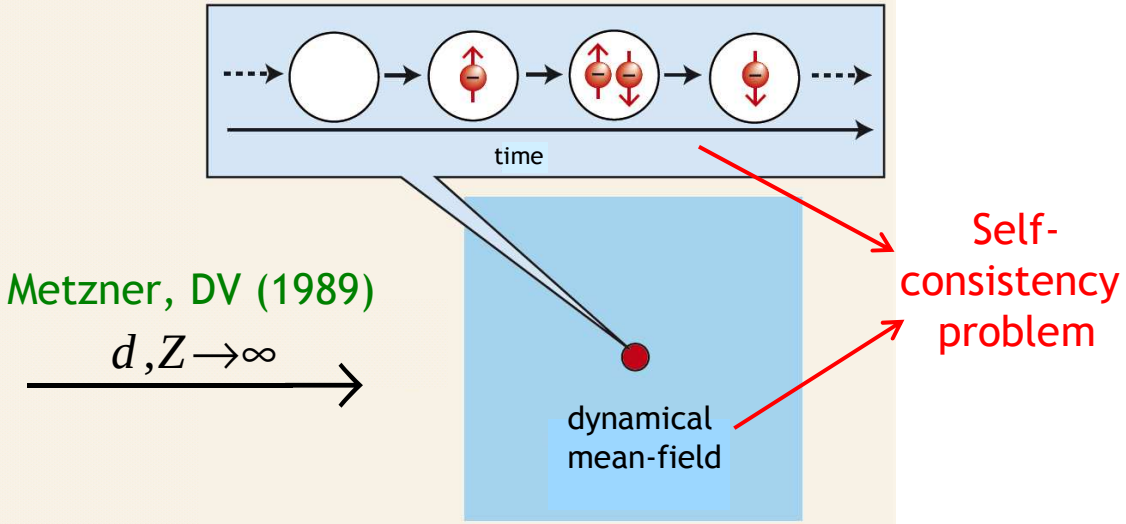
Theory of correlated electrons

$$H = -t \sum_{\langle \mathbf{i},\mathbf{j}\rangle,\sigma} c^{\dagger}_{\mathbf{i}\sigma} c_{\mathbf{j}\sigma} + U \sum_{\mathbf{i}} n_{\mathbf{i}\uparrow} n_{\mathbf{i}\downarrow}$$

Hubbard model



Z=12



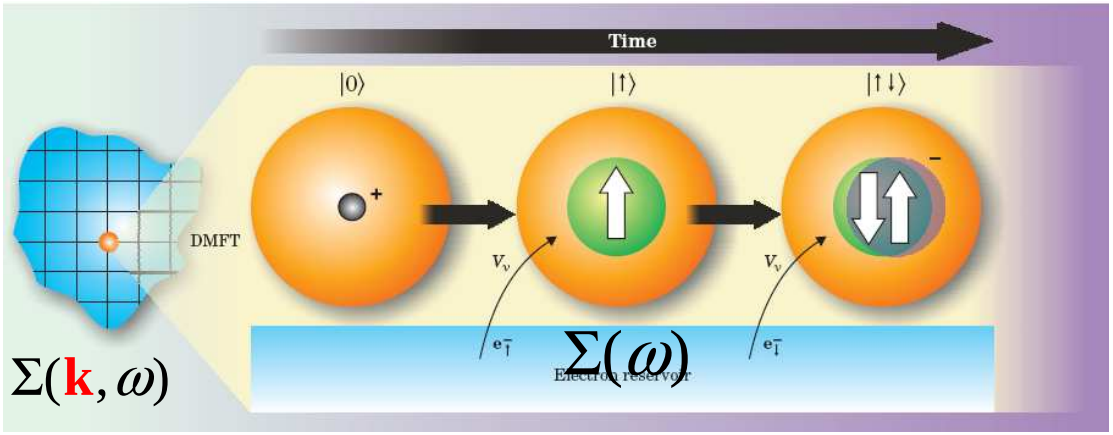
Dynamical single-site mean-field theory

Janiš (1991): Generalization of „coherent potential approx.“

Georges, Kotliar (1992), Jarrell (1992):
Self-consistent single-impurity Anderson model

Useful physical *interpretation*:

Hubbard model $\xrightarrow{d\to\infty}$ single-impurity Anderson model
+ self-consistency

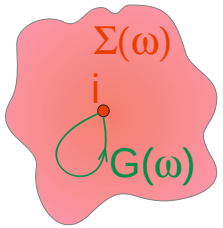


Kotliar, DV (2004)

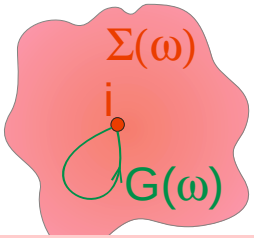
Proper time resolved treatment of local electronic interactions

"Dynamical Mean-Field Theory (DMFT)"

Local many-body problem with full dynamics



DMFT self-consistency equations



Σ(ω): "effective medium"

(i) Effective single impurity problem: "local propagator" $G_{\sigma n} = -\langle \psi_{\sigma n} \psi^*_{\sigma n} \rangle_{\mathcal{A}}$.

$$G = -\frac{1}{\mathcal{Z}} \int \mathcal{D}[\psi,\psi^*] \psi\psi^* e^{\underbrace{\psi^*[G^{-1}+\Sigma]\psi - U\psi^*\psi\psi^*\psi}_{\text{single-site ("impurity") action } A}}$$

(ii) *k*-integrated Dyson equation ("lattice Green function": lattice enters)

$$G(\omega) = \int d\varepsilon \frac{N^0(\varepsilon)}{\omega - \Sigma(\omega) - \varepsilon} = G^0(\omega - \Sigma(\omega))$$

→ free electrons in a dynamic potential Σ(ω)

→ Lecture of M. Kollar

DMFT: Search for the "best" impurity solver

Hubbard I
IPT
NCA

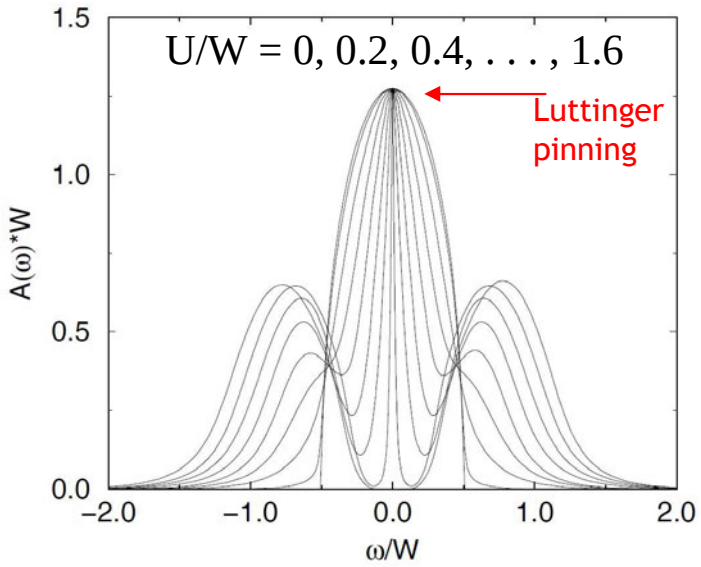
QMC (Hirsch-Fye) → Lecture of N. Blümer
ED
Lanczos → Lecture of E. Koch
NRG

Recent:

PQMC
DDMRG
CT-QMC → Lecture of P. Werner

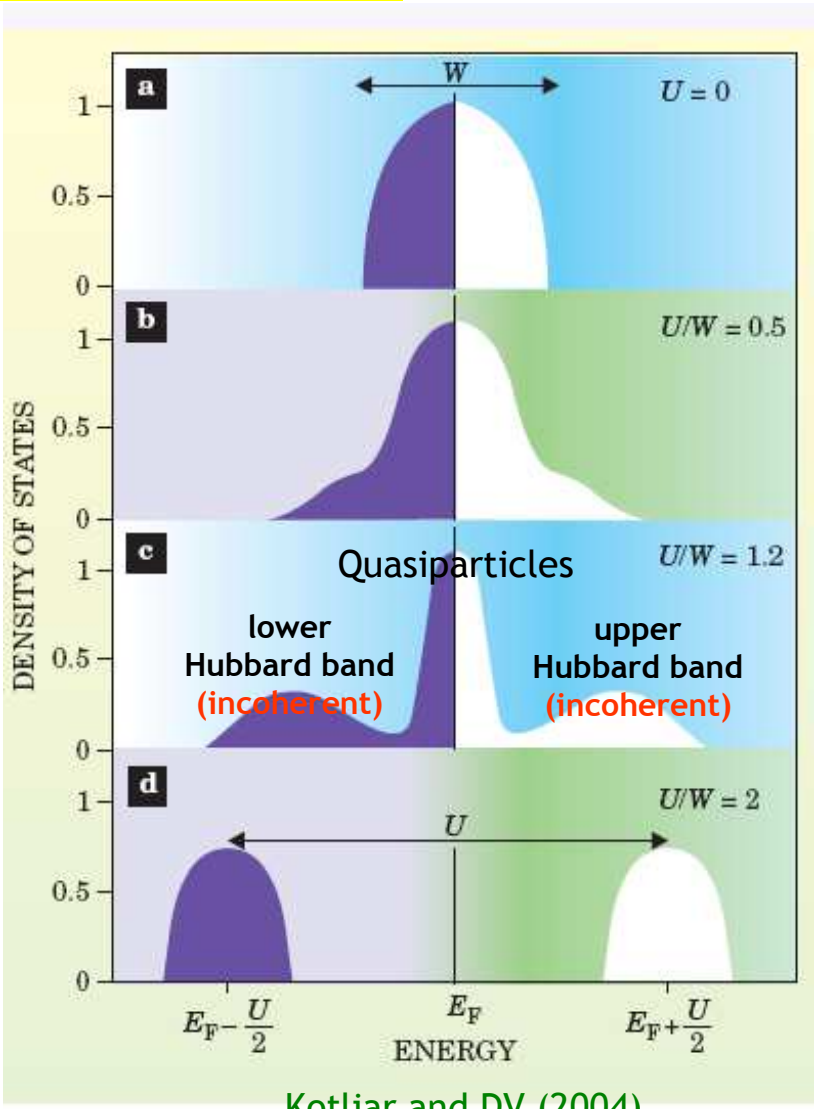
Application of DMFT:
1. Mott-Hubbard metal-insulator transition

Mott-Hubbard metal-insulator transition



Bethe lattice, T=0

Bulla (1999)



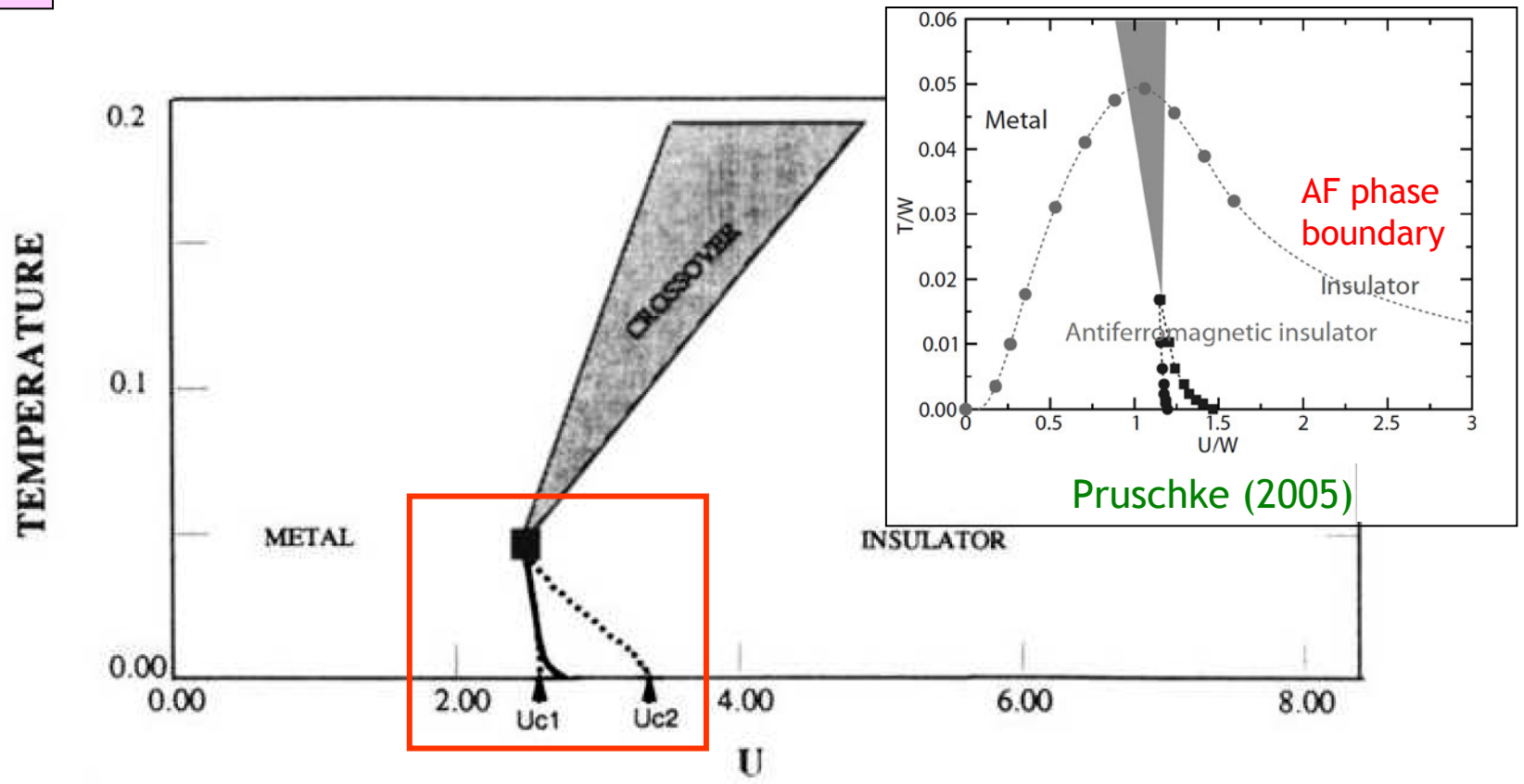
Kotliar and DV (2004)

Correlations (II): lead to transfer of spectral weight

Experimentally detectable ?

DMFT: Metal-insulator transition in the one-band Hubbard model

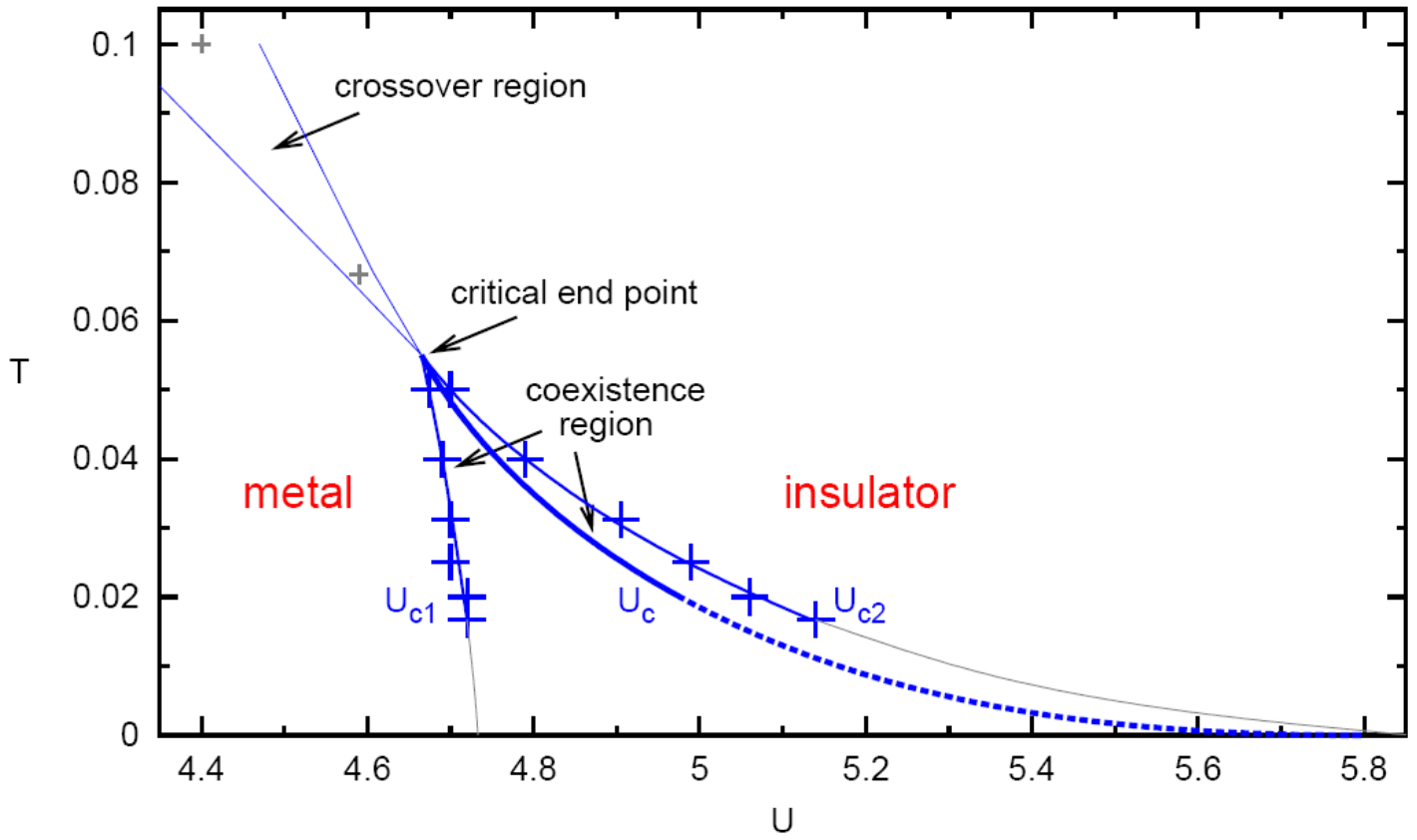
1994 paramagnetic solution ("frustrated antiferromagnetism")



Iterated perturbation theory Rozenberg *et al.* (1994)

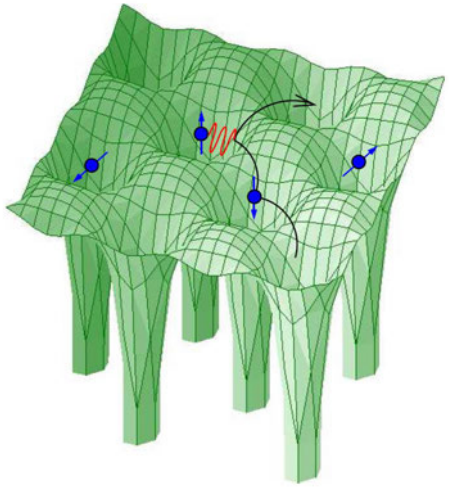
DMFT: Metal-insulator transition in the one-band Hubbard model

2002 paramagnetic solution ("frustrated antiferromagnetism")



Blümer (2002)

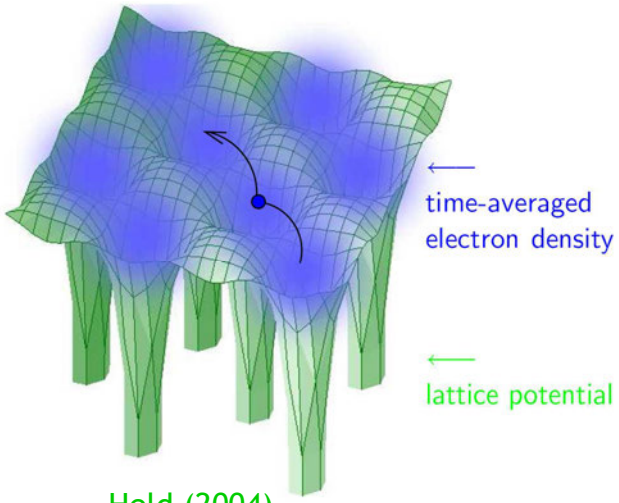
Application of DMFT:
2. Electronically Correlated Materials



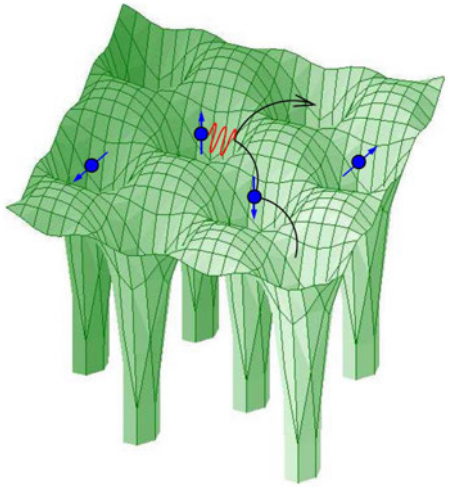
DFT/LDA

+	material specific: "ab initio"
−	fails for strong correlations
+	fast code packages

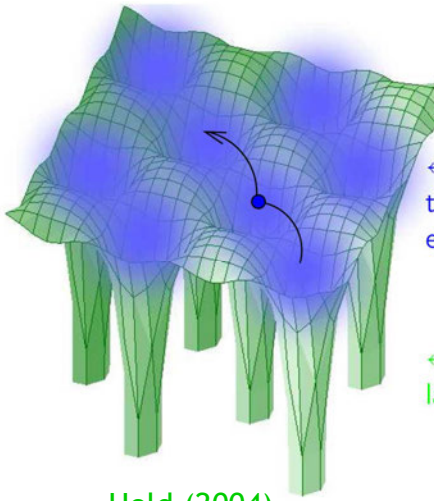
→ Lecture of P. Blöchl



Held (2004)

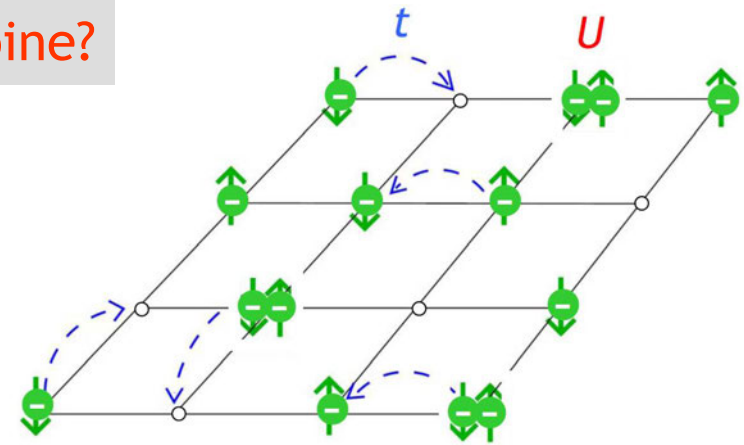


DFT/LDA		Model Hamiltonians	
+	material specific: "ab initio"	−	input parameters unknown
−	fails for strong correlations	+	systematic many-body approach
+	fast code packages	−	computationally expensive



Held (2004)

How to combine?



Computational scheme for correlated electron materials:

Material specific electronic structure
(Density functional theory: **LDA, GGA, ...**) or **GW**

+

Local electronic correlations
(Many-body theory: **DMFT**)

→ **LDA+DMFT**

→ Lecture of E. Pavarini

Anisimov, Poteryaev, Korotin, Anokhin, Kotliar (1997)
Lichtenstein, Katsnelson (1998)
Nekrasov, Held, Blümer, Poteryaev, Anisimov, DV (2000)

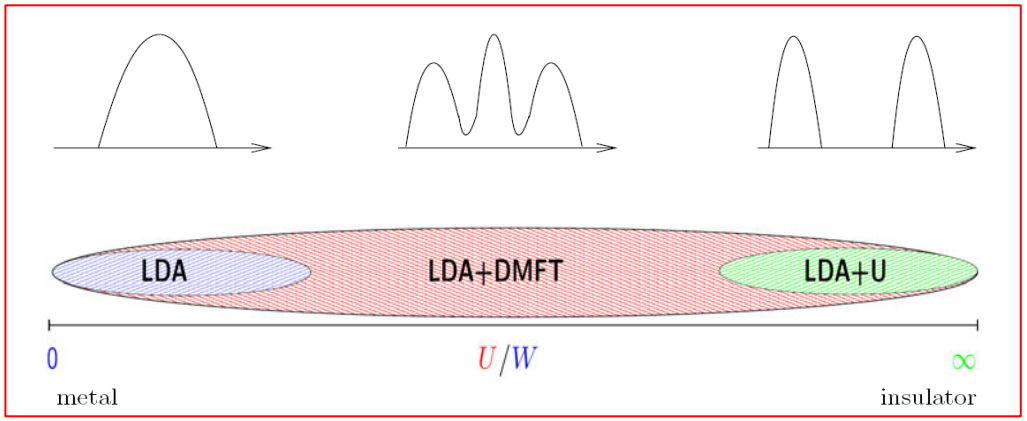
Combination with KKR ("KKR+DMFT") → Lecture of H. Ebert

Computational scheme for correlated electron materials:

Material specific electronic structure
(Density functional theory: **LDA, GGA, ...**) or **GW**

+

Local electronic correlations
(Many-body theory: **DMFT**)



Held *et al.* (Psi-k 2003)

LDA+DMFT (simplest version)

1) Calculate LDA band structure: $\varepsilon_{lml'm'}(k) \rightarrow \hat{H}_{LDA}$

2) Supplement LDA by local Coulomb interaction (only for correlated bands)

$$\hat{\mathcal{H}} = \underbrace{\sum_{\mathbf{k}\, lm\, l'm'\sigma} \epsilon_{lm\, l'm'}(\mathbf{k})\, \hat{c}^{\dagger}_{\mathbf{k}lm\sigma}\, \hat{c}_{\mathbf{k}l'm'\sigma}}_{\text{LDA}}$$

$$+ \underbrace{\sum_{\substack{i=i_d,\\ l=l_d}} {\sum_{m\sigma,m'\sigma'}}' \frac{U^{\sigma\sigma'}_{mm'}}{2} \hat{n}_{ilm\sigma}\hat{n}_{ilm'\sigma'}}_{\text{local Coulomb interaction}} \quad \underbrace{- \sum_{\substack{i=i_d,\\ l=l_d}} {\sum_{m\sigma,m'\sigma'}}' J_{mm'} \hat{c}^{\dagger}_{ilm\sigma}\hat{c}^{\dagger}_{ilm'\sigma'}\hat{c}_{ilm'\sigma}\hat{c}_{ilm\sigma'}}_{\text{Hund's rule coupling}}$$

→ Lecture of F. Aryasetiawan

LDA+DMFT (simplest version)

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$$\hat{\mathcal{H}} = \underbrace{\sum_{\mathbf{k}\, lm\, l'm'\sigma} \epsilon_{lm\, l'm'}(\mathbf{k})\, \hat{c}^{\dagger}_{\mathbf{k}lm\sigma}\, \hat{c}_{\mathbf{k}l'm'\sigma}}_{\text{LDA}} \underbrace{- \sum_{\substack{i=i_d,\\ \ell=\ell_d}} \sum_{m\sigma} \Delta\varepsilon_d \hat{n}_{i\ell m\sigma}}_{\text{double counting correction}}$$

$$+ \underbrace{\sum_{\substack{i=i_d,\\ l=l_d}} {\sum_{m\sigma,m'\sigma'}}' \frac{U^{\sigma\sigma'}_{mm'}}{2} \hat{n}_{ilm\sigma}\hat{n}_{ilm'\sigma'}}_{\text{local Coulomb interaction}} \underbrace{- \sum_{\substack{i=i_d,\\ l=l_d}} {\sum_{m\sigma,m'\sigma'}}' J_{mm'} \hat{c}^{\dagger}_{ilm\sigma}\hat{c}^{\dagger}_{ilm'\sigma'}\hat{c}_{ilm'\sigma}\hat{c}_{ilm\sigma'}}_{\text{Hund's rule coupling}}$$

→ Lecture of F. Aryasetiawan

Contact with experiment, e.g., via

Spectral function ("interacting DOS") in DMFT

k-integrated spectral function
→ PES

$$A(\omega) = -\frac{1}{\pi}\,\mathrm{Im}\,\mathbf{G}(\omega)$$

k-resolved spectral function
→ ARPES

$$\mathbf{G}(\mathrm{k},\omega) = [\omega - \boldsymbol{\Sigma}(\omega) - \mathbf{H}^{0}_{LDA}(\mathrm{k})]^{-1}$$

Matrices in orbital space

$$A(\mathrm{k},\omega) = -\frac{1}{\pi}\,\mathrm{Im}\;Tr\,\mathbf{G}(\mathrm{k},\omega)$$

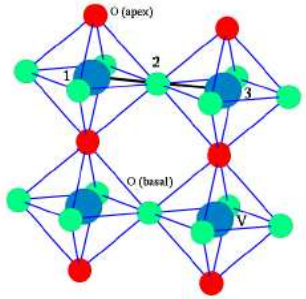
Application of LDA+DMFT

a) $(Sr,Ca)VO_3$: $3d^1$ system

Electronic structure

Crystal structure

$SrVO_3$: $\angle V - O - V = 180°$

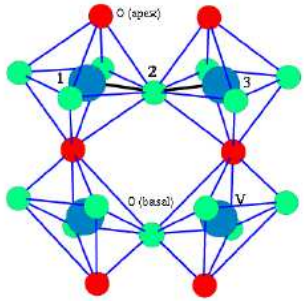


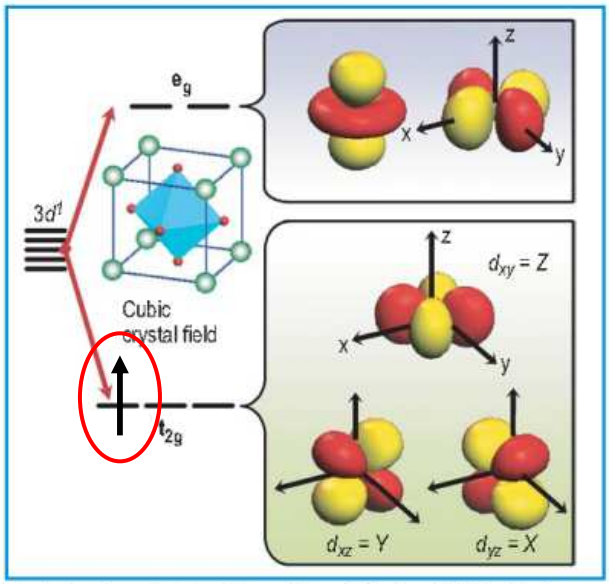
⇓

orthorhombic distortion

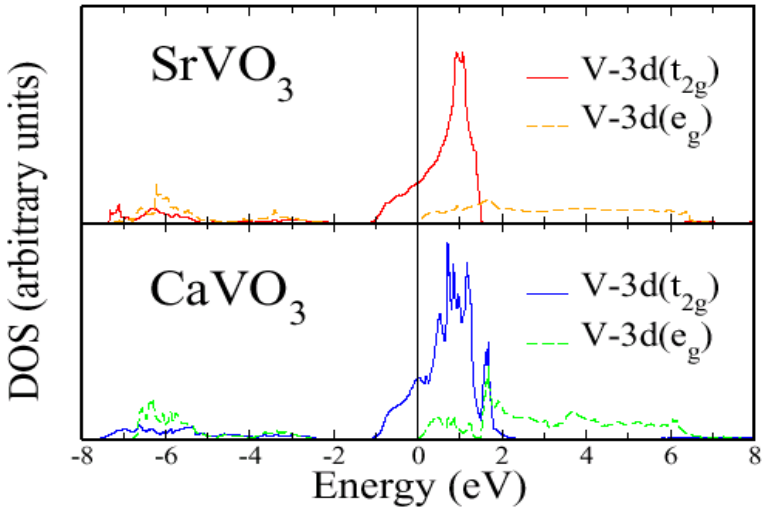
⇓

$CaVO_3$: $\angle V - O - V \approx 162°$





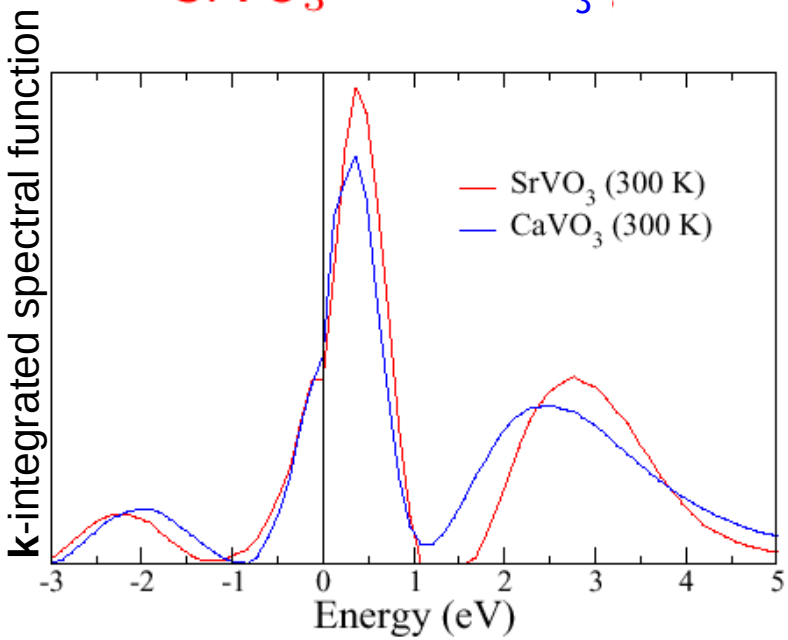
LDA density of states



No correlation effects/spectral transfer

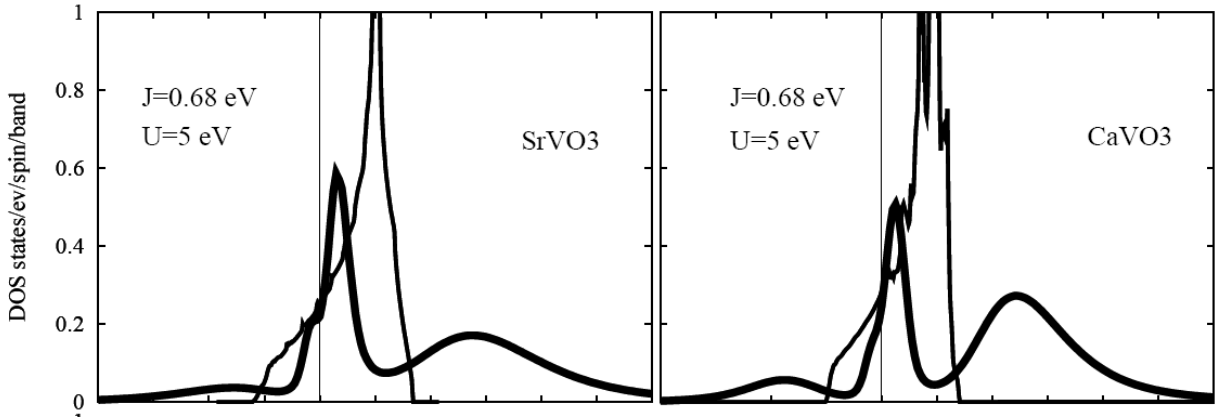
LDA+DMFT results

$SrVO_3$ and $CaVO_3$



Constrained LDA:
U=5.55 eV, J=1.0 eV

Sekiyama *et al.*, (2004)



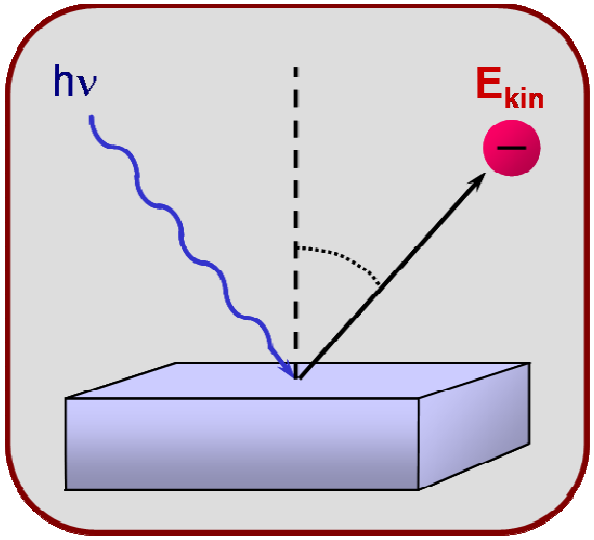
Pavarini *et al.* (2004)

How to measure ?

Excursion: Spectroscopy

→ Lecture of H. Tjeng

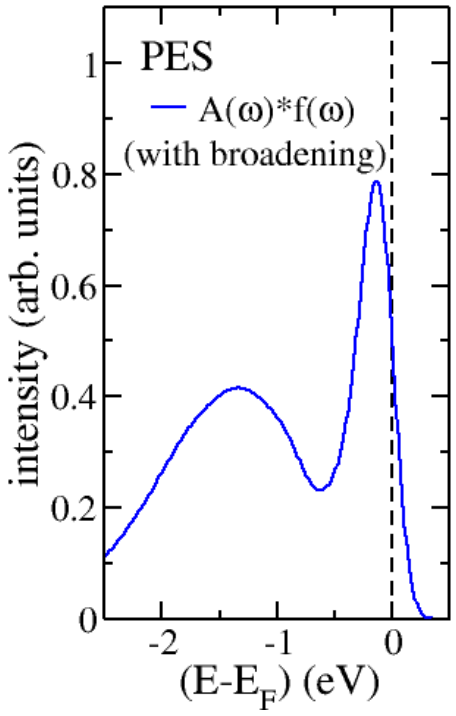
Photoemission Spectroscopy (PES)



Angular Resolved PES = ARPES

Measures occupied states of electronic spectral function

PES



Occupied states (experiment)

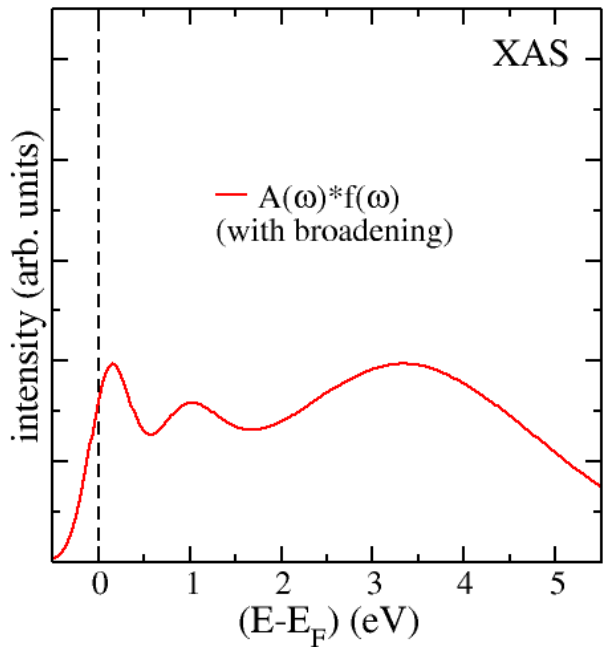
Inverse Photoemission Spectroscopy (IPES)

Measures unoccupied states of electronic spectral function

Information also available by:

X-ray absorption spectroscopy (XAS)

IPES/XAS

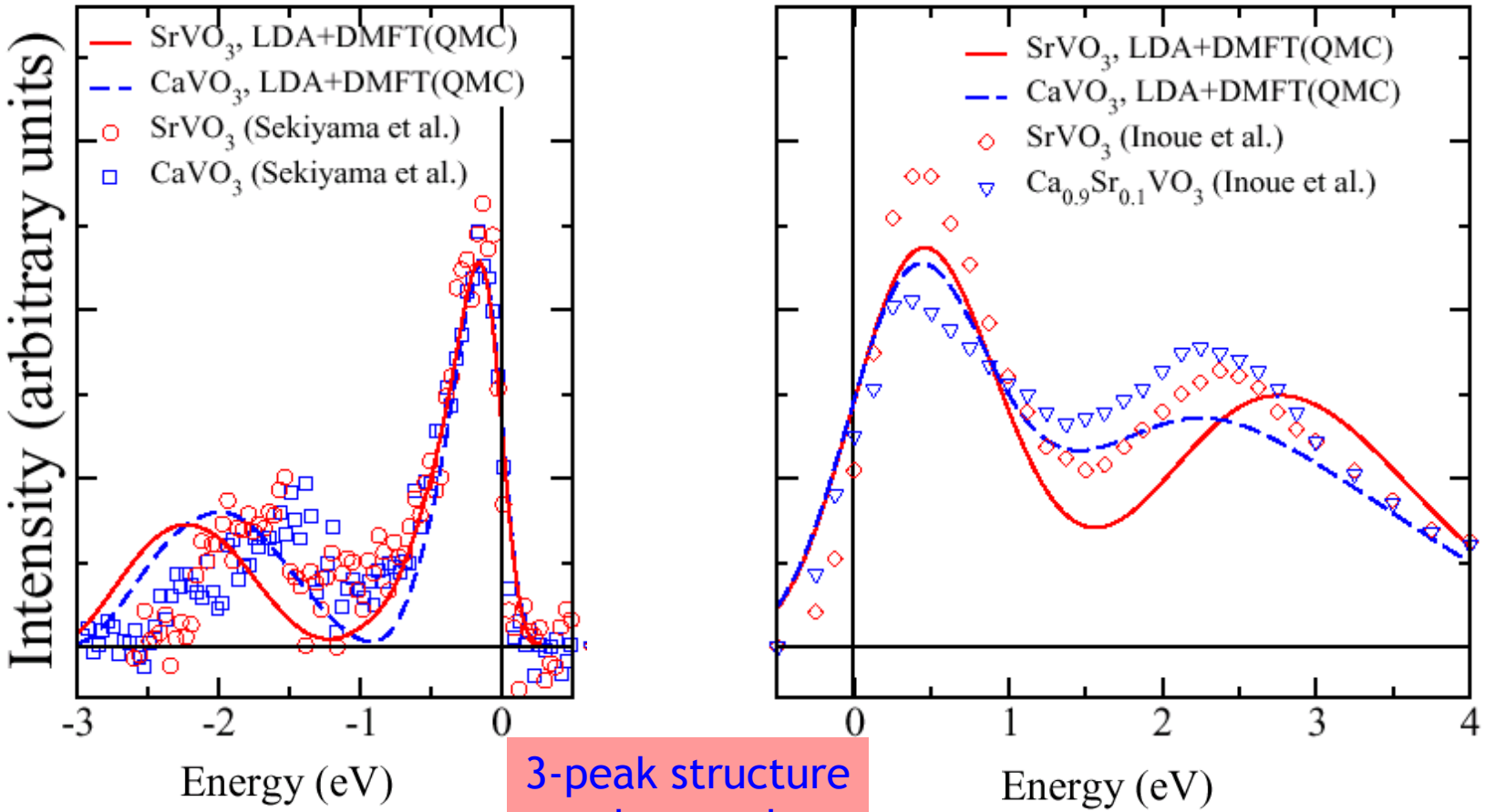


Unoccupied states (measured)

Comparison with experiment

Osaka – Augsburg – Ekaterinburg collaboration, (2004, 2005)

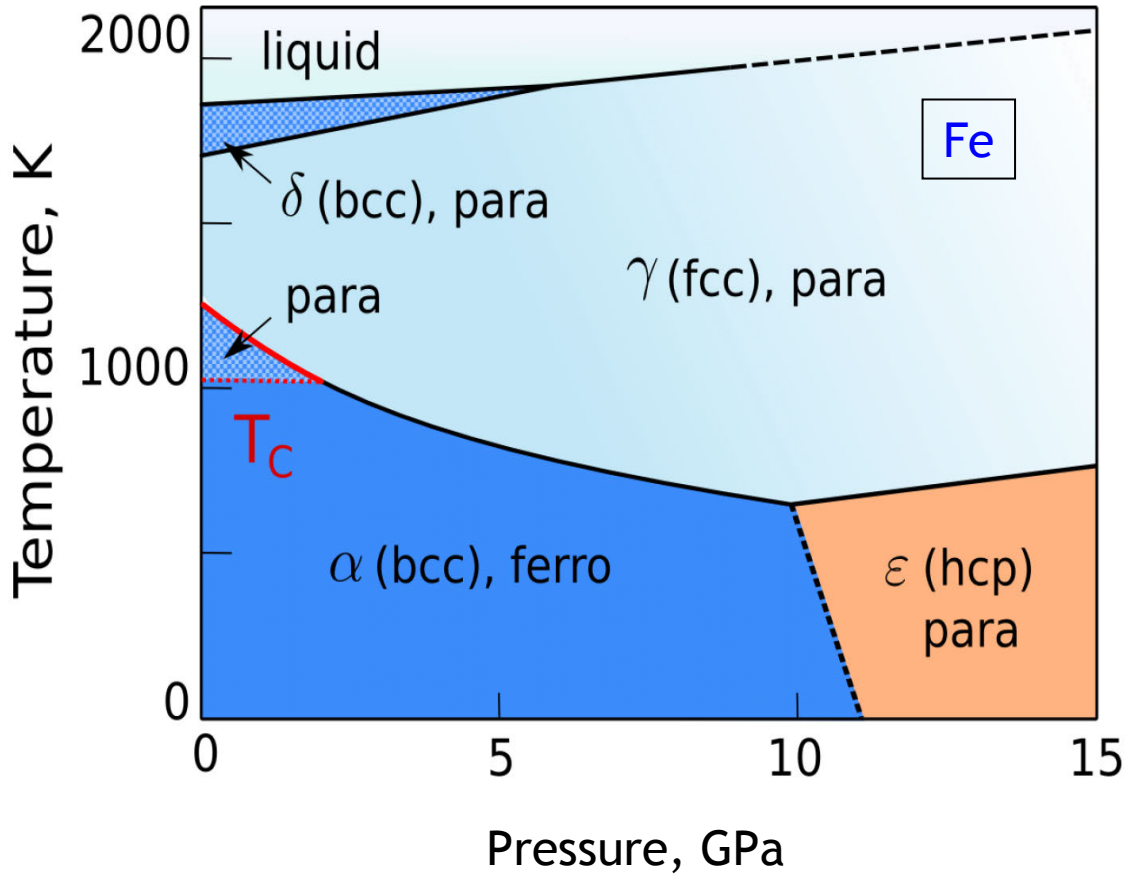
- (i) bulk-sensitive high-resolution photoemission spectra (PES) → occupied states
- (ii) $1s$ x-ray absorption spectra (XAS) → unoccupied states



3-peak structure detected

New developments in LDA+DMFT: Electronic correlations & structural transformations

Electron correlations can induce structural transformations



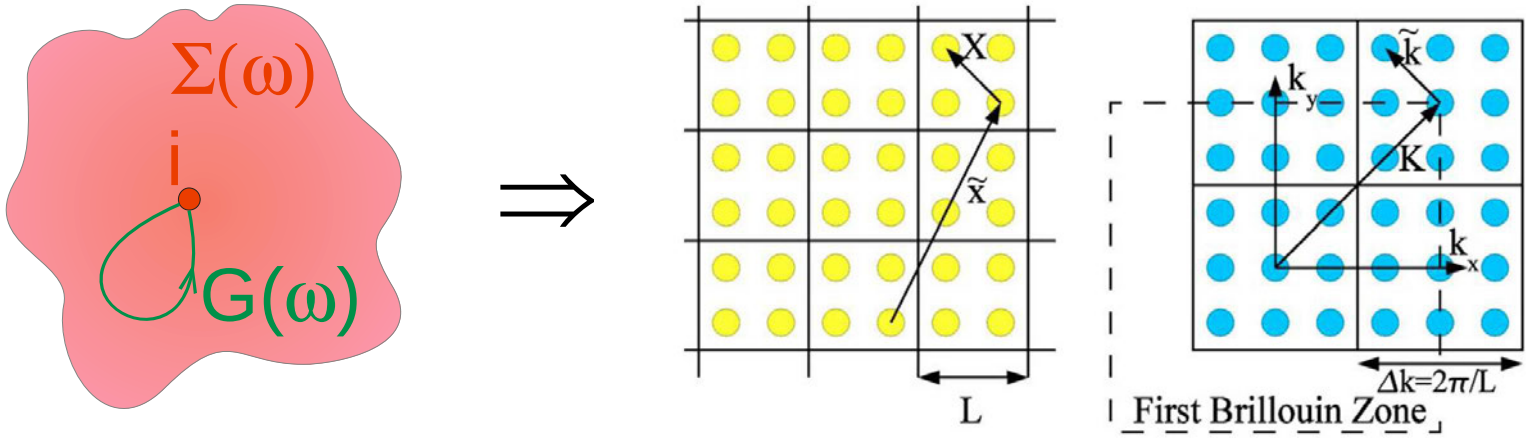
Leonov *et al.* (2011)

Beyond DMFT

Cluster Extensions

→ Lecture of A. Lichtenstein

- Dynamical cluster approx. (DCA) — Hettler *et al.* (1998, 2000)
- Cluster DMFT (CDMFT) — Kotliar *et al.* (2001)
- Self-energy functional theory — Potthoff (2003)



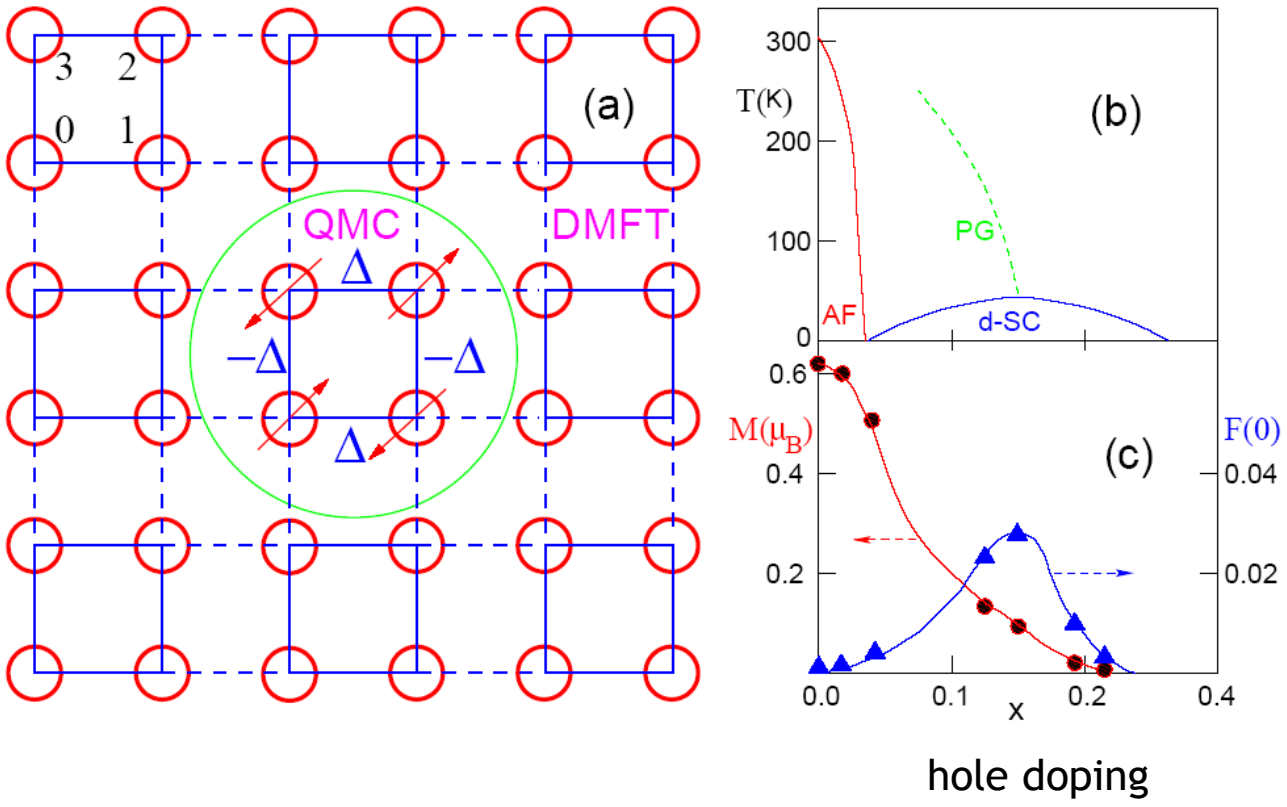
Dynamical vertex approximation (DΓA)

→ Lecture of K. Held

Local + non-local self-energy diagrams from local irreducible vertex

Toschi, Katanin, Held (2006)

Antiferromagnetic *d*-wave
2 × 2 periodically repeated cluster



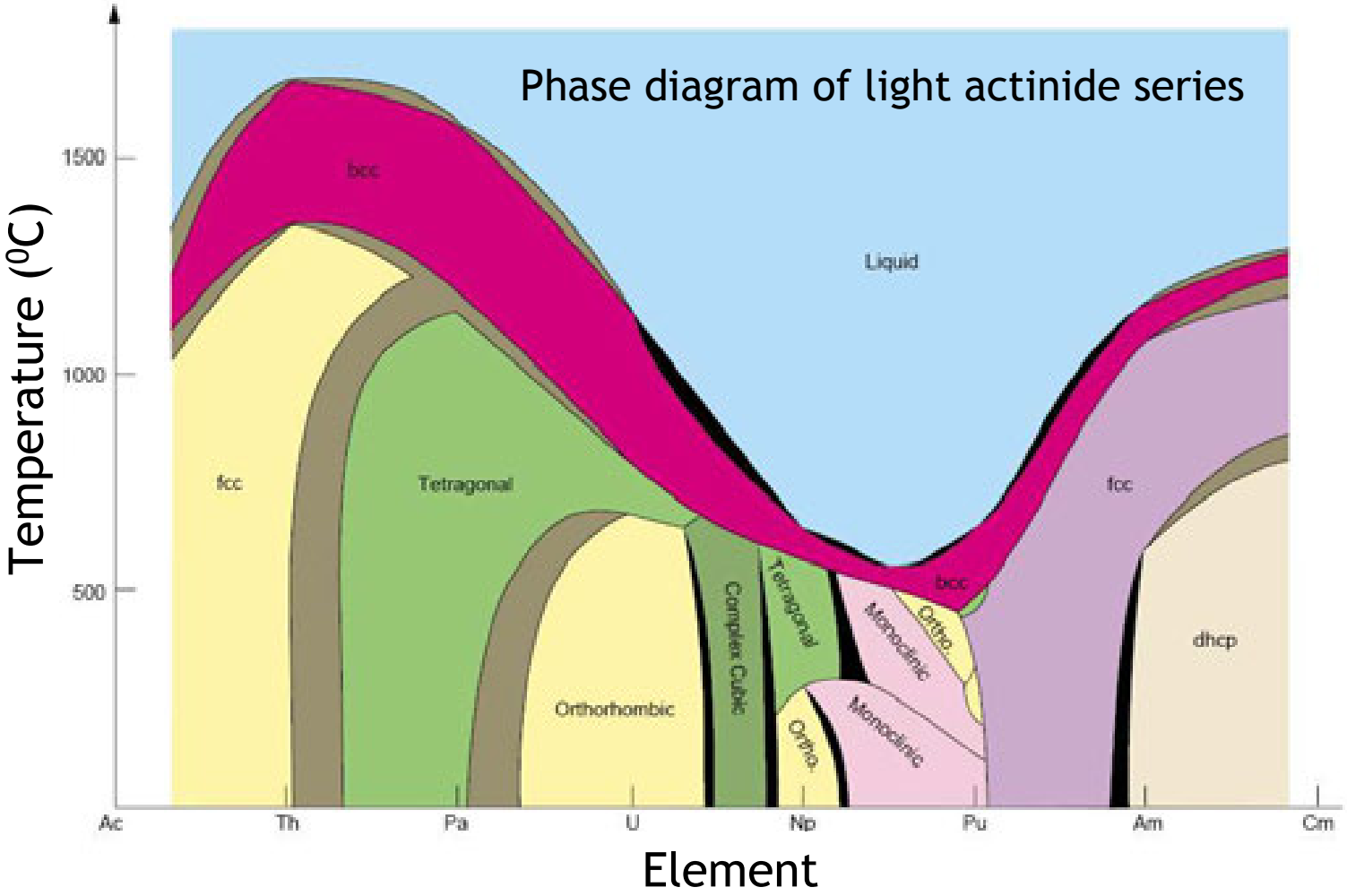
hole doping

Lichtenstein, Katsnelson (2000)

Perspectives of the LDA+DMFT approach

Perspectives of the LDA+DMFT approach

Explain and predict properties of complex correlated materials:

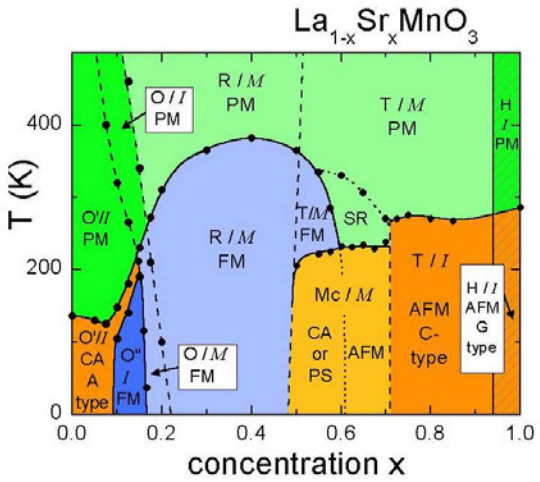


Phase diagram connecting individual binary alloy diagrams
Black: two-phase regions; Brown : details unknown

Boring and Smith (2000)

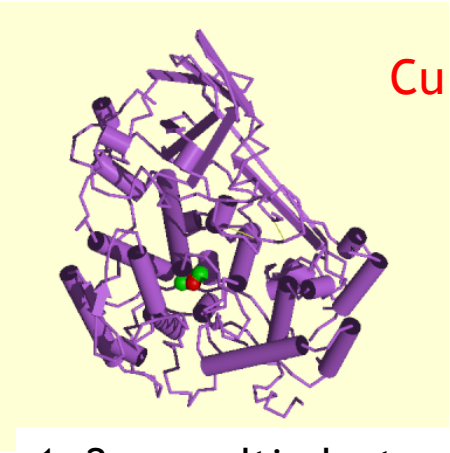
Perspectives of the LDA+DMFT approach

Explain and predict properties of complex correlated materials:



Phase diagram of $La_{1-x}Sr_xMnO_3$

Hemberger *et al.* (2002)



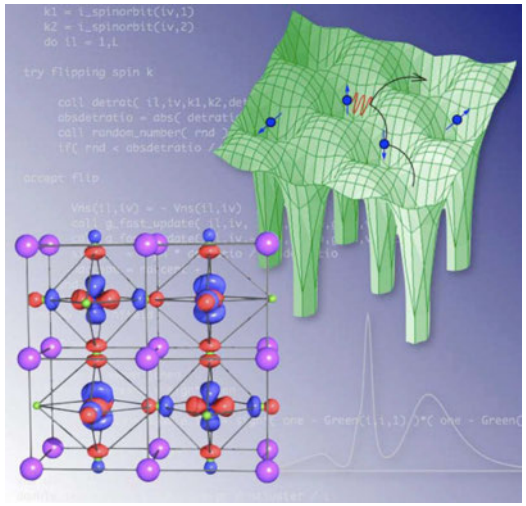
1, 2, … multi-electron transfer in metalloprotein complexes

→ Photosynthesis

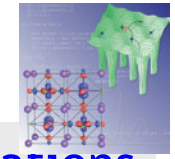
Goal: Dynamical mean-field approach with predictive power for strongly correlated materials

DFG Research Unit FOR 1346

Operates since July 2010



Projects of the DFG Research Unit FOR 1346



P1 Realistic many-body approach to materials with strong nonlocal correlations
(Lechermann, Potthoff, Lichtenstein)

P2 LDA+DMFT approach to multi-band correlation phenomena:
Susceptibilities and structural relaxation
(Kollar, Kuneš, Vollhardt)

P3 A self-consistent, relativistic implementation of the LSDA+DMFT method
(Ebert, Minár)

P4 Massively parallel simulations of strong electronic correlations:
Realistic Coulomb vertex and multiplet effects
(Pavarini, Koch, Blügel)

P5 Doping of prototypical Mott insulators:
Correlations, electronic structure, and electron-lattice effects
(Claessen, Valentí, Jeschke)

P6 Electronic structure of exemplary correlated materials
(Haverkort, Khomskii, Tjeng)

P7 Merging GW and dynamical mean-field theory
(Held, Toschi, Kresse) [Austria]

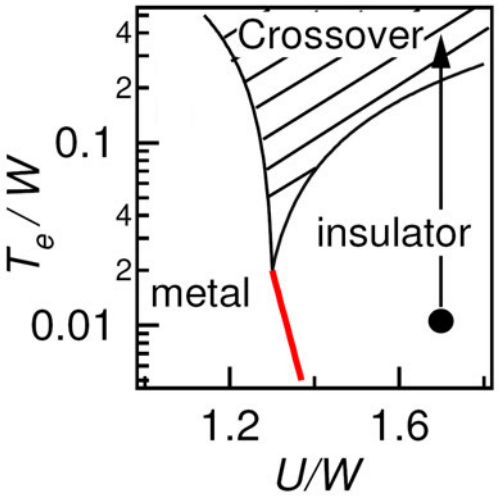
P8 Quantum Monte Carlo impurity solvers for multi-orbital problems
and frequency dependent interactions
(Assaad, Blümer, Werner [Switzerland])

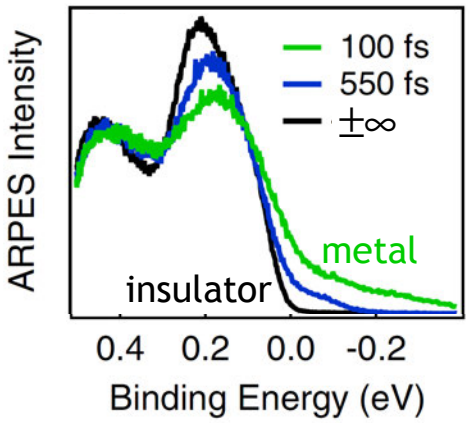
P9 Energies and forces for materials with strong correlations
(Blöchl, Pruschke)

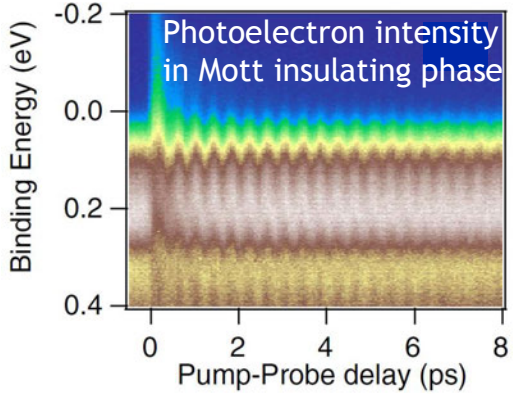
Application of DMFT:
3. Recent Developments

a. Correlated electrons in non-equilibrium

Real-time evolution of correlation phenomena, e.g.,
time-resolved photoemission spectroscopy







Perfetti *et al.* (2006)

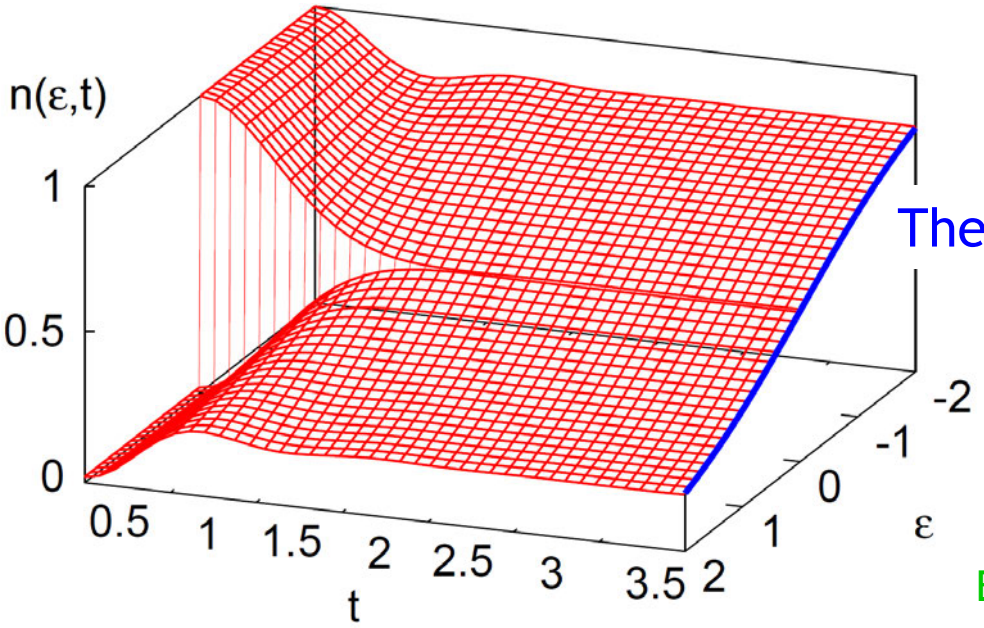
Required: Theory of non-equilibrium beyond linear response in correlated bulk materials

a. Correlated electrons in non-equilibrium

Non-equilibrium DMFT — Freericks, Turkowski (2006)

Quench in Hubbard model from *U=0* to *U>0* — Eckstein, Kollar, Werner (2009)

Momentum distribution *(U=3.3)*



time after quench

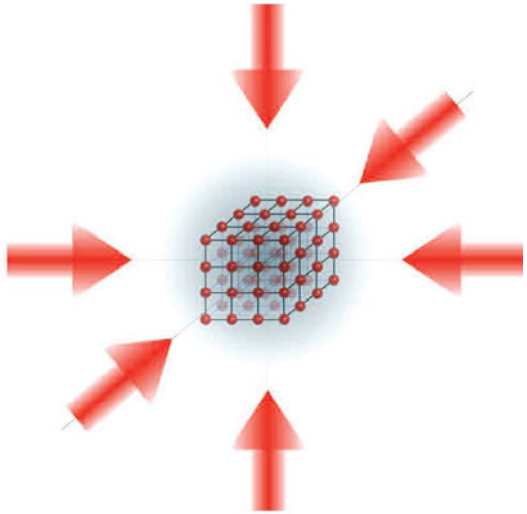
Thermalization

Application, e.g.
- time resolved PES
- pump-probe experiments

Eckstein, Kollar (2008)

Freericks, Krishnamurthy, Pruschke (2008)

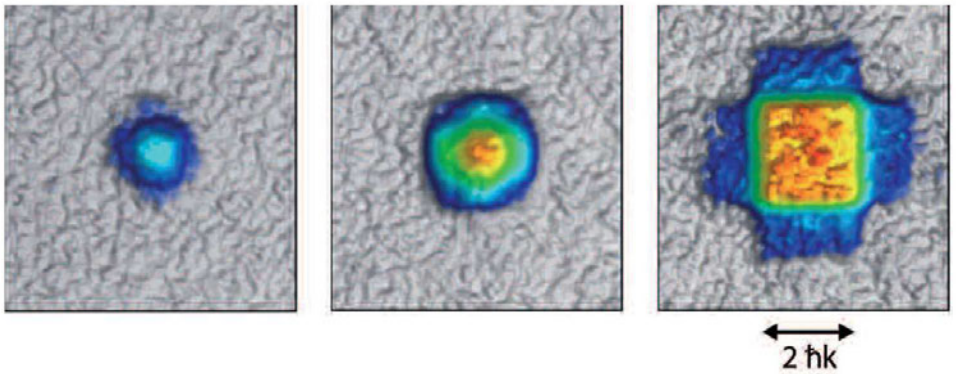
b. Correlated cold atoms in optical lattices



Greiner *et al.* (2002)

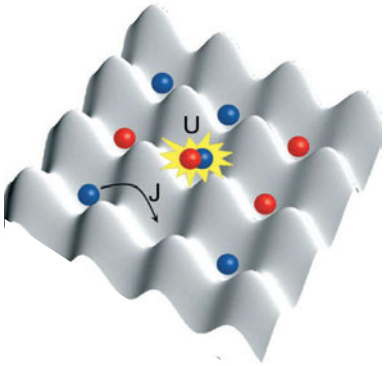
Bosonic/fermionic atoms in optical lattices: Exp. realization of models

High degree of tunability: “quantum simulator”



Observation of Fermi surface (^{40}K atoms) Köhl, Esslinger (2006)

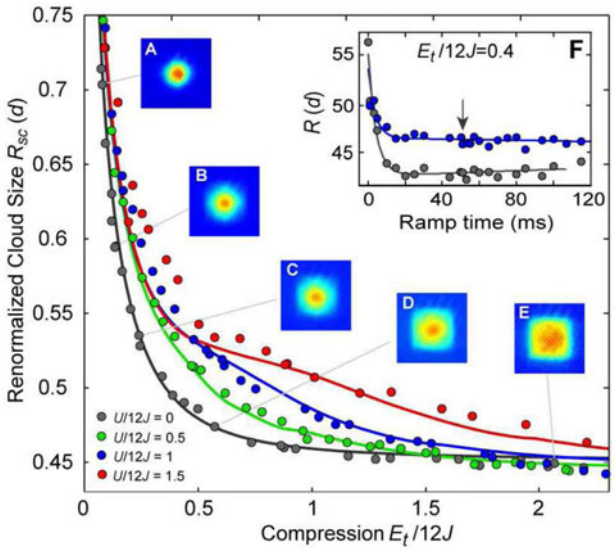
b. Correlated cold atoms in optical lattices



Hubbard model with ultracold atoms Jaksch *et al.*, (1998)

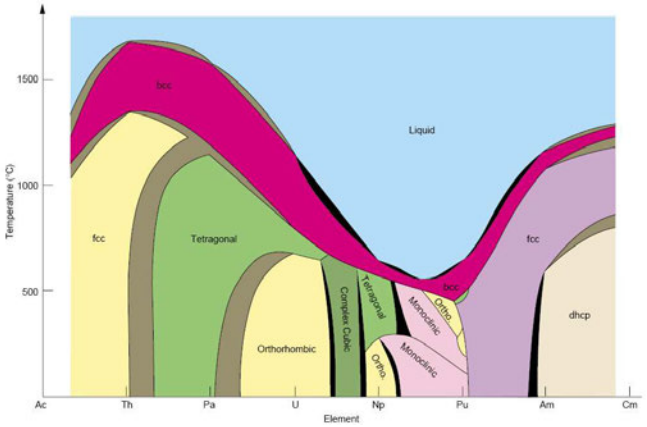
Atomic total angular momentum $L^{tot} = F$ → N=2F+1 hyperfine states
→ SU(N) Hubbard models Honerkamp and Hofstetter (2004)

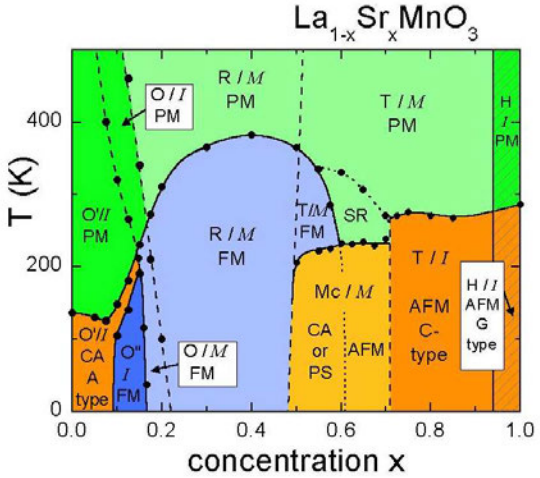
^{40}K atoms

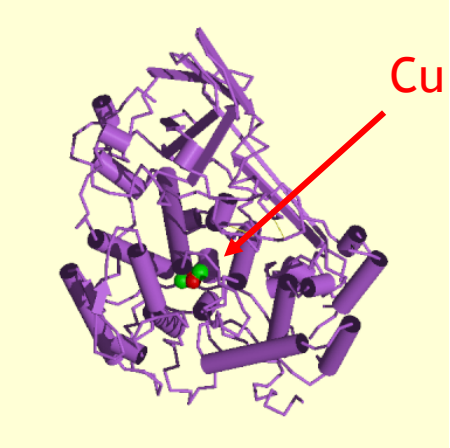


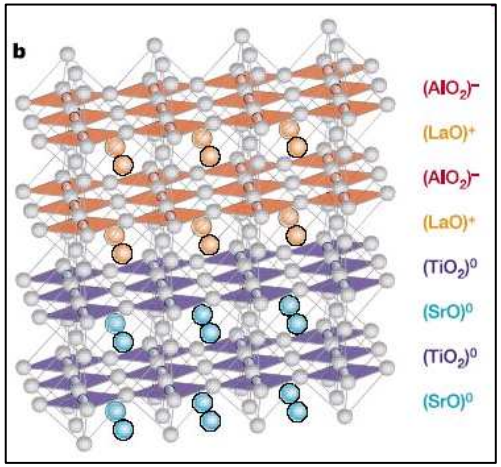
DMFT results experimentally confirmed

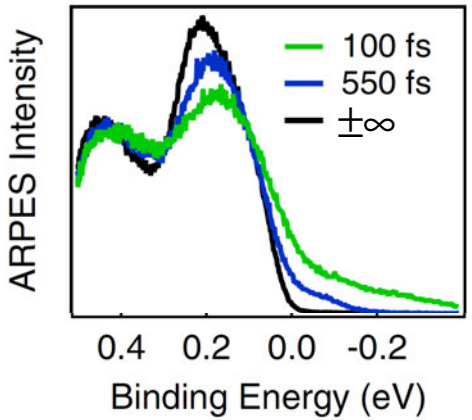
Metallic and Insulating Phases of Repulsively Interacting Fermions in a 3D Optical Lattice Schneider *et al.*, (2008)

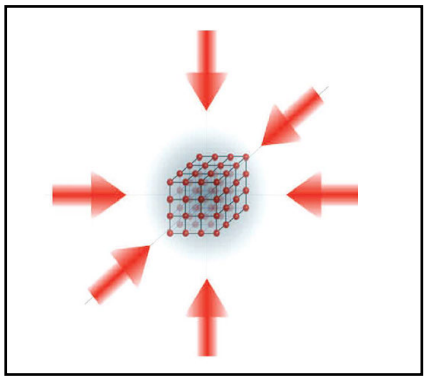












Wide field of applications for DMFT based techniques