Dynamical Mean-Field Approach for Strongly Correlated Materials

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Outline:

• Electronic correlations
• Dynamical Mean-Field Theory (DMFT)
• Applications of DMFT to models and materials
Correlations
Correlation [lat.]: \textit{con} + \textit{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 $\rightarrow$ long-range order enforced $\rightarrow$ trivial correlations
Temporal/spatial correlations in everyday life

Time/space average inappropriate
Electronic Correlations in the Periodic Table
### Partially filled d-orbitals

- **d-orbitals**
  - In transition metals and lanthanides

### Partially filled f-orbitals

- **f-orbitals**
  - In actinides and lanthanides

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

**Note:** The subgroup numbers 1–18 were adopted in 1994 by the International Union of Pure and Applied Chemistry. The names of elements 110–118 are the Latin equivalents of those numbers.
Electronic Correlations in Solids
1. **Mott metal-insulator transition in $V_2O_3$**

- PI → PM: 1. order transition without lattice symmetry change

- Anomalous slope of P(T) → *Pomeranchuk effect* in $^3$He

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Microscopic explanation?
2. Photoemission spectra of NiO


Origin of gap (antiferromagnetism)?
Photoemission spectra of (Sr,Ca)VO$_3$

Reason for shift of spectral weight?

Osaka - Augsburg - Ekaterinburg collaboration: Sekiyama et al., 2004
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

with

Technological applications:
- sensors, switches
- magnetic storage
- refrigerators
- functional materials, …
Electronic Correlations: Models

Lecture of F. Lechermann
**Hubbard model**
(tight binding approach)

\[
H = -t \sum_{\langle i,j \rangle, \sigma} c_{i\sigma}^\dagger c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}
\]

\[
\langle n_{i\uparrow} n_{i\downarrow} \rangle \neq \langle n_{i\uparrow} \rangle \langle n_{i\downarrow} \rangle
\]

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

Gutzwiller, 1963
Hubbard, 1963
Kanamori, 1963
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:

\[ \langle S_i S_j \rangle \rightarrow \langle S_i \rangle \langle S_j \rangle \]

\( \rightarrow \) Weiss MFT

mean field
What is a “mean-field theory (MFT)“?

2) Construction by exaggeration

For example:
Spin $S$
Degeneracy $N$
Dimension $d$ /coordination number $Z$

$\{Z \rightarrow \infty\}$

Spin models:
$\text{fcc-lattice (d=3): } Z=12$

exaggeration $\rightarrow$ factorization

static mean field $h_{MF}$
Theory of correlated electrons

\[ H = -t \sum_{\langle i,j \rangle, \sigma} c_i^{\dagger} c_j \sigma + U \sum_i n_{i\uparrow} n_{i\downarrow} \]

Hubbard model

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

Effective dynamical single-site problem
Hubbard model: Simplifications for $d, Z \to \infty$

\[ \left\langle H_{kin} \right\rangle_0 = \frac{-t}{\sqrt{Z}} \sum_{i, \sigma} \sum_{j(\text{NN} \ i)} \left\langle C_{i\sigma}^\dagger C_{j\sigma} \right\rangle_0 \]

Amplitude for hopping $j \to \text{NN} \ i$

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

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

Metzner, DV (1989)
Hubbard model: Simplifications for $d, Z \to \infty$

$$\langle H_{kin} \rangle_0 = -t \sum_{i,\sigma} \sum_{j(\text{NN} \ i)} \left( \frac{1}{Z} \right) \langle C_{i\sigma}^\dagger C_{j\sigma} \rangle_0$$

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:

Metzner, DV (1989)

Hubbard model: Simplifications for $d, Z \to \infty$
Theory of correlated electrons

\[ H = -t \sum_{\langle i,j \rangle, \sigma} c_{i\sigma}^\dagger c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow} \quad \text{Hubbard model} \]


Dynamical single-site mean-field theory
Proper time resolved treatment of local electronic interactions

Useful physical interpretation:

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

Useful physical interpretation:

\[ \Sigma(k, \omega) \]

Kotlier, DV (2004)

Proper time resolved treatment of local electronic interactions

“Dynamical Mean-Field Theory (DMFT)“

Local many-body problem with full dynamics
(i) Effective single impurity problem: “local propagator”

\[ G_{\sigma n} = -\langle \psi_{\sigma n} \psi_{\sigma n}^* \rangle_A. \]

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

(ii) \( k \)-integrated Dyson equation (“lattice Green function“: lattice enters)

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

\[ \rightarrow \text{free electrons in a dynamic potential } \Sigma(\omega) \]

\[ \rightarrow \text{Lecture of M. Kollar} \]
DMFT: Search for the “best” impurity solver

Hubbard I
IPT
NCA

QMC (Hirsch-Fye)
ED
Lanczos
NRG

Recent:
PQMC
DDMRG
CT-QMC

→ Lecture of N. Blümer
→ Lecture of E. Koch
→ Lecture of P. Werner
Application of DMFT:
1. Mott-Hubbard metal-insulator transition
Mott-Hubbard metal-insulator transition

Bethe lattice, $T=0$
Bulla (1999)

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)

Pruschke (2005)
AF phase boundary
DMFT: Metal-insulator transition in the one-band Hubbard model

paramagnetic solution ("frustrated antiferromagnetism")

2002

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)

time-averaged electron density
lattice potential
How to combine?

<table>
<thead>
<tr>
<th>DFT/LDA</th>
<th>Model Hamiltonians</th>
</tr>
</thead>
<tbody>
<tr>
<td>+ material specific: “ab initio”</td>
<td>- input parameters unknown</td>
</tr>
<tr>
<td>- fails for strong correlations</td>
<td>+ systematic many-body approach</td>
</tr>
<tr>
<td>+ fast code packages</td>
<td>- computationally expensive</td>
</tr>
</tbody>
</table>

Held (2004)

time-averaged electron density

lattice potential
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

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

Combination with KKR ("KKR+DMFT")

→ Lecture of E. Pavarini

→ 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: \( \varepsilon_{lm'm'}(k) \rightarrow \hat{H}_{LDA} \)

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

\[
\hat{h} = \sum_{k l m l' m' \sigma} \varepsilon_{lm'l'm'}(k) \hat{c}_{klm\sigma}^\dagger \hat{c}_{kl'm'\sigma}
\]

LDA

\[
+ \sum_{i=i_d, m\sigma, m'\sigma'} \sum' \frac{U_{mm'}^{\sigma\sigma'}}{2} \hat{n}_{ilm\sigma} \hat{n}_{ilm'\sigma'}
\]

local Coulomb interaction

\[
- \sum_{i=i_d, m\sigma, m'\sigma'} \sum' J_{mm'}^{\sigma\sigma'} \hat{c}_{ilm\sigma}^\dagger \hat{c}_{ilm'\sigma'}^\dagger \hat{c}_{ilm'\sigma} \hat{c}_{ilm\sigma'}
\]

Hund’s rule coupling

→ Lecture of F. Aryasetiawan
LDA+DMFT (simplest version)

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

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

\[
\hat{h} = \sum_{klm, l'm'} \epsilon_{lml'm'}(k) \hat{c}^\dagger_{klm\sigma} \hat{c}_{kl'm'\sigma} - \sum \sum \Delta \epsilon_d \hat{n}_{ilm\sigma}
\]

LDA

double counting correction

\[
+ \sum_{i=i_d, m\sigma, m'\sigma'} \sum_{l=l_d} \frac{U^{\sigma\sigma'}}{2} \hat{n}_{ilm\sigma} \hat{n}_{ilm'\sigma'} - \sum \sum \sum \frac{J^{\sigma\sigma'}}{2} \hat{c}^\dagger_{ilm\sigma} \hat{c}^\dagger_{ilm'\sigma'} \hat{c}_{ilm'\sigma} \hat{c}_{ilm\sigma'}
\]

local Coulomb interaction

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

\[
A(\omega) = -\frac{1}{\pi} \text{Im} G(\omega)
\]

**k-resolved** spectral function
→ ARPES

\[
G(k, \omega) = \left[ \omega - \Sigma(\omega) - H_{LDA}^0(k) \right]^{-1}
\]

Matrices in orbital space

\[
A(k, \omega) = -\frac{1}{\pi} \text{Im} \, Tr \, G(k, \omega)
\]
Application of LDA+DMFT

a) (Sr,Ca) VO$_3$: 3d$^1$ system
Electronic structure

Crystal structure

SrVO$_3$: $\angle V - O - V = 180^\circ$

CaVO$_3$: $\angle V - O - V \approx 162^\circ$

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


Pavarini *et al.* (2004)

**How to measure?**
Excursion: Spectroscopy

Photoemission Spectroscopy (PES)

Angular Resolved PES = ARPES

Measures occupied states of electronic spectral function
Occupied states (experiment)
Inverse Photoemission Spectroscopy (IPES)

Measures unoccupied states of electronic spectral function

Information also available by:

X-ray absorption spectroscopy (XAS)
**Unoccupied states**
*(measured)*
Comparison with experiment

- (i) bulk-sensitive high-resolution photoemission spectra (PES) → occupied states
- (ii) 1s x-ray absorption spectra (XAS) → unoccupied states
New developments in LDA+DMFT: 
Electronic correlations & structural transformations

Electron correlations can induce structural transformations

Leonov et al. (2011)
Beyond DMFT

Cluster Extensions

• Dynamical cluster approx. (DCA)  Hettler et al. (1998, 2000)
• Cluster DMFT (CDMFT)  Kotliar et al. (2001)

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 $\times$ 2 periodically repeated cluster

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 of light actinide series

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$_x$MnO$_3$

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
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

Required: Theory of non-equilibrium beyond
linear response in correlated bulk materials
**a. Correlated electrons in non-equilibrium**

Non-equilibrium DMFT

Quench in Hubbard model from $U=0$ to $U>0$

Eckstein, Kollar, Werner (2009)

Momentum distribution ($U=3.3$)

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

Freericks, Krishnamurthy, Pruschke (2008)

Eckstein, Kollar (2008)

Freericks, Turkowski (2006)
b. Correlated cold atoms in optical lattices

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

High degree of tunability: “quantum simulator”

Observation of Fermi surface \((^{40}\text{K atoms})\)  Köhl, Esslinger (2006)
b. Correlated cold atoms in optical lattices

Hubbard model with ultracold atoms  
\[ \text{Jaksch et al.}, \ (1998) \]

Atomic total angular momentum \( L^{\text{tot}} = F \rightarrow N = 2F + 1 \) hyperfine states
\[ \rightarrow \text{SU}(N) \text{ Hubbard models} \]

\[ ^{40}\text{K} \text{ atoms} \]

DMFT results experimentally confirmed

Metallic and Insulating Phases of Repulsively Interacting Fermions in a 3D Optical Lattice  
\[ \text{Schneider et al.}, \ (2008) \]
Wide field of applications for DMFT based techniques