Electronic correlations in models and materials

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Outline:

- Electronic correlations
- A short history of the limit $d \to \infty$ and dynamical mean-field theory (DMFT)
- Mott-Hubbard metal-insulator transition
- LDA+DMFT for correlated electron materials
- Recent developments

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Correlations

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

• go beyond static mean-field theories (e.g., Hartree-Fock)

Correlations in everyday life

Time/space average insufficient
### Electronic Correlations in Solids

#### Partially filled d-orbitals

#### Partially filled f-orbitals

#### Narrow d,f-orbitals/bands → strong electronic correlations
Correlated electron materials

Fascinating topics for fundamental research

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

Technological applications:
- sensors, switches
- magnetic storage
- refrigerators
- functional materials, ...
Electronic Correlations: Models
The Hubbard model is a quantum mechanical model used in solid-state physics to describe the behavior of electrons in a lattice. It was introduced by John Hubbard in 1963. The Hamiltonian of the Hubbard model is given by:

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

where:
- $H$ is the Hamiltonian of the system.
- $t$ is the hopping parameter, representing the energy cost for an electron to hop from one site to another.
- $c_{i\sigma}^\dagger$ and $c_{i\sigma}$ are the creation and annihilation operators for an electron of spin $\sigma$ at site $i$.
- $U$ is the on-site repulsion energy, describing the energy cost of having two electrons in the same site.
- $n_{i\uparrow}$ and $n_{i\downarrow}$ are the occupation numbers of the up and down spin states at site $i$.

Gutzwiller, 1963
Hubbard, 1963
Kanamori, 1963
$H = -t \sum_{\langle i,j \rangle, \sigma} c_{i\sigma}^\dagger c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}$
Hubbard model

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

Local Hubbard physics:
Local Hubbard physics:

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

Static (Hartree-Fock) mean-field theories generally insufficient.
\[ H = \sum_{i,j,\sigma} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} + U \sum_{i} n_{i\uparrow} n_{i\downarrow} \]

- Gutzwiller variational wave function

\[ \left| \psi_G \right\rangle = e^{-\lambda D} \left| \psi_0 \right\rangle \]

One-particle wave function
(Hartree-Fock, BCS, etc.)

- Gutzwiller approximation

= semi-classical approximation of expectation values

Gutzwiller (1963/65)
Hubbard (1963)
Kanamori (1963)
Gutzwiller-Brinkman-Rice theory

\[
\frac{1}{L} \left\langle \psi_G \middle| H \middle| \psi_G \right\rangle \bigg|_{\langle H \rangle = E} = q(d)\varepsilon_0 + U \frac{d}{d}, \quad \frac{\partial E}{\partial d} = 0
\]

Conditions for ferromagnetism?

Brinkman, Rice (1970)

\[
m^{*} = q^{-1} \xrightarrow{U \to U_c} \infty
\]

describes metal-insulator ("Mott") transition \( \xrightarrow{} V_2O_3 \)

Gutzwiller (1963/65)

\[
d = \frac{1}{4} \left(1 - \frac{U}{U_c}\right)
\]

\[
U_c = 8\varepsilon_0
\]

\[
q = 1 - \left(\frac{U}{U_c}\right)^2
\]

\[
E_g = -L\varepsilon_0 \left(1 - \frac{U}{U_c}\right)^2
\]
Gutzwiller-Brinkman-Rice theory: Application to normal liquid $^3$He

Liquid $^3$He: "almost localized Fermi liquid" (near Mott transition)

Gutzwiller approximation ↔ Landau Fermi liquid theory

$F_0^a = \frac{3}{4} p$, $p \approx 1$

Gutzwiller approximation is remarkably good: Systematic derivation possible?
Gutzwiller wave function

Exact analytic evaluation of $E_G = \frac{\langle \psi_G | H | \psi_G \rangle}{\langle \psi_G | \psi_G \rangle}$ in $d=1$

Metzner, DV (1987/88)

Diagrams for the kinetic energy

Analytic calculation of all diagrams possible

Numerical check by Monte-Carlo integration

$\frac{v(d)}{v(1)}$

$\frac{3}{4} n$

Great simplifications for $d \to \infty$

Diagrams for the Hubbard interaction
Gutzwiller wave function in $d \to \infty$

$$|\psi_G\rangle = e^{-\lambda H_U} |\psi_0\rangle$$

$$E_G = \frac{\langle \psi_G | H | \psi_G \rangle}{\langle \psi_G | \psi_G \rangle}$$

Gutzwiller approximation becomes **exact** in $d \to \infty$

Metzner, DV (1989)

$d \to \infty$: Evaluation of $E_G$ for arbitrary $|\psi_0\rangle$

Gebhard (1990)

Multi-band generalization:

„Gutzwiller DFT“

Ferromagnetic Ni: Cut of Fermi surface

Bünemann, Gebhard, Ohm, Weiser, Weber (2005)
Correlated lattice fermions in $d \to \infty$
\[ H = J \sum_{\langle i,j \rangle} S_i S_j \quad \xrightarrow{\text{Scaling, } Z \text{ or } d \to \infty} \quad H_{MF} = h_{MF} \sum_i S_i \]

Scaling: \[ J = \frac{J^*}{Z} \]

Local (single-site) mean-field theory

Comprehensive MFT valid for all input parameters
Hubbard model

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

- non-local

\[ \text{Scaling} \quad ? \]

\[ Z \text{ or } d \to \infty \]

Local (single-site) mean-field theory

Hubbard model

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

Amplitude for hopping \( j \rightarrow \text{NN} \ i \)

\[ \left| \text{Amplitude for hopping } j \rightarrow \text{NN} \ i \right|^2 = \text{Probability for hopping } j \rightarrow \text{NN} \ i = \frac{1}{Z} \]
Hubbard model

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

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

Collapse of all connected, irreducible diagrams in position space

⇒ great simplifications

Metzner, DV (1989)
Hubbard model

\[ \langle H_{kin} \rangle_0 = -t \sum_{i,j} \sum_{\sigma} \langle C_i^{\dagger} C_j \rangle_0 \]

\[ Z \text{ or } d \to \infty \]

\[ \frac{1}{\sqrt{Z}} \]

\[ \frac{1}{\sqrt{Z}} \]

Quantum rescaling

\[ t = \frac{t^*}{\sqrt{Z}} \]

Metzner, DV (1989)

Collapse of all connected, irreducible diagrams in position space

\[ \Rightarrow \text{great simplifications} \]

e.g., correlation energy \( E_2 \)

\[ \frac{E_2}{U^2} \propto e_2 \]

\( d = \infty \): excellent approximation for \( d = 3 \)
Correlated Lattice Fermions in $d = \infty$ Dimensions

Walter Metzner and Dieter Vollhardt

Institut für Theoretische Physik C, Technische Hochschule Aachen, Sommerfeldstrasse 26/28, D-5100 Aachen, Federal Republic of Germany

(Received 28 September 1988)


Correlated fermions on a lattice in high dimensions

E. Müller-Hartmann

Institut für Theoretische Physik, Universität zu Köln, Federal Republic of Germany

Received October 12, 1988

• $\Sigma(\mathbf{k}, \omega)$

• Only Hubbard interaction remains dynamical


SECOND ORDER U-PERTURBATION APPROACH TO THE ANDERSON LATTICE MODEL IN HIGH DIMENSIONS

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(Received 4 November 1988 by B. Mühlschlegel)
Falicov-Kimball model in $d \to \infty$

= Hubbard model with immobile  
  e.g., $\downarrow = \text{proton}$, $\uparrow = \text{electron}$

$\Rightarrow$ exact solution

Brandt, Mielsch (1989)

$\Rightarrow$ exact solution

van Dongen, DV (1990)

$\Rightarrow$ exact solution

$d \to \infty$: mean-field theory for fermionic lattice models
$d \to \infty$ mean-field theory: Hubbard model

Generalization of the “Coherent Potential Approximation“
to interacting systems in $d \to \infty$

Janiš (1991); Janiš, DV (1992)

Dynamical (single-site) mean-field theory
$d \to \infty$ mean-field theory: Hubbard model

Georges, Kotliar (1992)

Single-impurity Anderson model + self-consistency

QMC solution: Jarrell (1992)
DMFT self-consistency equations

(i) Effective single-site/impurity problem: “local propagator“

\[ G = -\frac{1}{Z} \int D[\psi, \psi^*] \psi \psi^* e^{\psi^* [G^{-1} + \Sigma] \psi - U \psi^* \psi \psi^* \psi} \]

single-site ("impurity") action \( \mathcal{A} \)

(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 \( \Sigma(\omega) \)

“Impurity solver“

QMC  Hirsch-Fye (1986)
ED   Caffarel, Krauth (1994), Si et al. (1994)
NRG  Bulla (1999)
Dynamical mean-field theory (DMFT) of correlated electrons

**DMFT:** local theory with full many-body dynamics

Kotliar, DV (2004)

Mott-Hubbard metal-insulator transition
Metal-insulator transition in the one-band Hubbard model (DMFT)

Rozenberg, Kotliar, Zhang (1994)

Blümer (2002)
Metal-insulator transition in the one-band Hubbard model (DMFT)

Kotliar, DV (2004)

$S = \gamma T$

$S = k_B \ln 2$

Introduction of spin fluctuations via E-DMFT
Si (1996)

Park, Haule, Kotliar (2008)

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

How to combine?

Held (2004)

Held (2004)
Computational scheme for correlated electron materials:

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

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

\[ \text{LDA+DMFT} \]

Anisimov, Poteryaev, Korotin, Anokhin, Kotliar (1997)
Lichtenstein, Katsnelson (1998)
Nekrasov, Held, Blümer, Poteryaev, Anisimov, DV (2000)
Computational scheme for correlated electron materials:

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

+ 

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

Held, Nekrasov, Keller, Eyert, Blümer, McMahan, Scalettar, Pruschke, Anisimov, DV (Psi-k 2003)
**LDA+DMFT (simplest version)**

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

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

\[
\hat{\mathcal{H}} = \sum_{klm,m'\sigma} \epsilon_{lm'm'}(k) \hat{c}_{klm\sigma}^\dagger \hat{c}_{kl'm'\sigma} - \sum_{i=i_d, m\sigma} \sum_{\ell=l_d} \Delta \epsilon_d \hat{n}_{ilm\sigma} \\
\text{LDA} \\
\text{double counting correction}
\]

\[
+ \sum_{i=i_d, m\sigma} \sum_{\ell=l_d}^\prime \frac{U^\sigma\sigma'}{2} \hat{n}_{ilm\sigma} \hat{n}_{ilm'\sigma'} - \sum_{i=i_d, m\sigma,m'\sigma'} \sum_{\ell=l_d}^\prime J_{mm'} \hat{c}_{ilm\sigma}^\dagger \hat{c}_{ilm'\sigma'}^\dagger \hat{c}_{ilm'\sigma'} \hat{c}_{ilm\sigma'} \\
\text{local Coulomb interaction} \\
\text{Hund’s rule coupling}
\]

3) Solve self-consistently with an impurity solver
Applications of LDA+DMFT
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

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

\[ A(k, \omega) = -\frac{1}{\pi} \text{Im} \, Tr \, G(k, \omega) \]
1. Application: $(\text{Sr,Ca})\text{VO}_3$

**Crystal structure**

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

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

LDA density of states

No correlation effects/spectral transfer
LDA+DMFT results

Constrained LDA:

\[ U=5.55 \text{ eV}, \quad J=1.0 \text{ eV} \]

SrVO$_3$ and CaVO$_3$;

Osaka - Augsburg - Ekaterinburg collaboration:

Comparison with experiment


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

3-peak structure in bulk material confirmed
Kinks in strongly correlated electron systems

Kinks due to electronic interaction in high-$T_c$ cuprates (non-phononic)

Coupling of quasiparticles to spin fluctuations

Manske, Eremin, Bennemann (2001)
Randeria, Paramekanti, Trivedi (2004)
Kordyuk et al. (2004)
Kakehashi, Fulde (2005)

$k$-dependence of self-energy $\Sigma(k, \omega)$ essential

Origin of kinks?

Kinks at $|\omega_c| \approx 0.24$ eV

Nekrasov, Held, Keller, Kondakov, Pruschke, Kollar, Andersen, Anisimov, DV (2006)
SrVO$_3$ and CaVO$_3$

Byczuk, Kollar, Held, Yang, Nekrasov, Pruschke, DV (2007)

Kinks in the quasiparticle dispersion $E_k$ at $\pm \omega_*$

- Generic features of strongly correlated electrons
- No coupling to other excitations required

local spin/charge fluctuations $\rightarrow$ Raas, Grete, Uhrig (2009)
Kinks in the quasiparticle dispersion $E_k$ at $\pm \omega_*$

$\omega_* = Z_{FL} D_{LDA}$

Kinks in high-resolution ARPES of Ni(110)

2. Application: Mott-Hubbard vs. charge-transfer insulators

- Charge-transfer gap clearly seen
- Valence band: p-d character
- Conduction band: d-character

E.g., NiO:
include correlated Ni-3d + O-2p states ("p-d hybridization")

Kuneš, Anisimov, Lukoyanov, DV (2007)

3. Application: Correlation induced structural transformations

KCuF₃: Prototypical Jahn-Teller system

Kugel, Khomskii (1982)

T > Tₑ ≈ 38 K: Correlated paramagnetic insulator with strong cooperative JT distortion

GGA+DMFT implementation with plane-wave pseudo-potentials

Leonov, Binggeli, Korotin, Anisimov, Stojić, DV (2008)

- paramagnetic insulator
- δ_{JT}^{opt} = 4.1%

→ Structural transformation caused by electronic correlations

Orbital order

Pavarini, Koch, Lichtenstein (2008)
Other recent developments
1. Inhomogeneous systems: Layers, surfaces, interfaces

Mott-insulator-band-insulator heterostructure: Charge density profile

Okamoto, Millis (2004)

Potthoff, Nolting (1999)

1. Inhomogeneous systems: Optical lattices and traps

- Fermionic atoms in a $3d$ potential trap  
  Helmes, Costi, Rosch (2008)

- $2d$ square lattice with harmonic confinement
  Snoek, Titvinidze, Töke, Byczuk, Hofstetter (2008)

R-DMFT: Real-space magnetization (AF) profiles
2. Non-equilibrium DMFT

Hamiltonian $H(t) \rightarrow$ Green function $G(t,t')$

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

- Discontinuity at Fermi surface
  - Möckel, Kehrein (2008)
  - Uhrig (2009)

- Momentum distribution $(U=3.3)$
  - Möckel, Kehrein (2008)
  - Uhrig (2009)

- Prethermalization
  - Möckel, Kehrein (2008)

- Thermalization
  - Möckel, Kehrein (2008)

Schmidt, Monien (2002)
Freericks, Turkowski (2006)
Eckstein, Kollar (2008)
**Application: Pump-probe spectroscopy**

1\textsuperscript{st} light pulse: Pumps into non-equilibrium state
2\textsuperscript{nd} light pulse: Probes to study relaxation

**Time-resolved photoemission spectroscopy**

Mott insulator (FK model, $U=10$)
photo-excited into metallic state ($U=1$):

- Kin. energy of emitted electrons
- Time after pump [$\hbar/W \sim 10\text{fs}$]

- Initial metallic state
- UHB
- LHB

Pulse duration $\delta$

$\leftrightarrow$

Energy resolution

$\Delta E \approx \hbar/\delta$

Freericks, Krishnamurthy, Pruschke (2008)

Eckstein, Kollar (2008)
Beyond single-site DMFT

Cluster Extensions

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

Dynamical vertex approximation (DΓA) Toschi, Katanin, Held (2006)

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

Dual fermion approach Rubtsov, Katsnelson, Lichtenstein (2008)
Conclusions and outlook

- DMFT is the canonical mean-field theory for correlated electrons
- Provides insights into their characteristic properties
- **Goal:** Develop LDA+DMFT into a comprehensive computational tool with **predictive power** for complex correlated materials
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