Electronic Correlations in Solids: From Models to Materials

Dieter Vollhardt

Supported by DFG
TRR 80
FOR 1346

Boston University; February 14, 2011
Outline:

- Peculiarities of quantum many-particle systems
- Correlations
- Electronic correlations in solids
- Dynamical Mean-Field Theory: Models vs. materials
- Other Developments & Perspectives
Peculiarities of Interacting Many-Particle Systems
Interacting many-particle systems

Elementary ("bare") particles + fundamental interactions

\[ \text{\# particles } N \rightarrow \infty \]

effective ("quasi") particles + effective interactions
Non-interacting electrons

\[
\text{Spin} = \frac{1}{2} \quad \text{Fermion}
\]

\[N \to \infty \quad \downarrow \quad \text{Pauli exclusion principle of many fermions}\]

Fermi-Dirac statistics

\[\downarrow\]

Ground state: \textbf{Fermi body/surface}
Fermi gas: **Ground state**

Fermi body

Fermi surface
Fermi gas: **Excited state**

- **Fermi surface** ($k_F$) → **Fermi body**
- **k-eigenstates**: infinite life time
- **Switch on repulsive interaction**
Fermi liquid

“Standard model of condensed matter physics”

Well-defined $k$-states ("quasiparticles") with
- finite life time
- effective mass
- effective interaction

1-1 correspondence between one-particle states $(k, \sigma)$

(Quasi-) Particle
(Quasi-) Hole

Landau (1956/58)

Fermi body

Fermi surface $(k_F) \rightarrow$
Simple metals

"Heavy Fermions"

\[ \lim_{T \to 0} \frac{C_V}{T} = \gamma \propto \frac{m^*}{m}, \quad v_F = \frac{\hbar k_F}{m^*} \]

Potassium

Steglich et al. (1979)

\[ m^* \approx 1000 \ m \]

Stewart et al. (1983)

\[ \text{CeCu}_2\text{Si}_2, \text{UBe}_{13}: \]

very heavy quasiparticles

Result of elementary excitations (quasiparticles)
Interacting many-particle systems

Elementary ("bare") particles + fundamental interactions

\[ \text{# particles } N \rightarrow \infty \]

effective ("quasi") particles + effective interactions

Vacuum

\[ \frac{Q}{r} \]

Coulomb potential
Interacting many-particle systems

Elementary ("bare") particles + fundamental interactions

\[ N \rightarrow \infty \]

effective ("quasi") particles + effective interactions

**Electron gas: Screening**
Simplest approximation: Thomas-Fermi

Effective Yukawa potential
Interacting many-particle systems

Elementary ("bare") particles + fundamental interactions

\[ N \to \infty \]

effective ("quasi") particles + effective interactions

Electron gas: Screening
Better approximation: Lindhard

\[ \frac{Q}{r^3} \cos(2k_F r) \]

Friedel oscillations
Electrons in real solids

Interacting many-particle systems

Elementary ("bare") particles + fundamental interactions

\[ \# \text{ particles } N \rightarrow \infty \]

effective ("quasi") particles + effective interactions

"Strong effective interaction" of electrons in localized orbitals
Interacting many-particle systems

\[ N \rightarrow \infty \]

Entirely new phenomena, e.g., phase transitions

Unpredicted behavior “emerges”
Correlations
Correlation [lat.]: con + relatio ("with relation")

Grammar: either ... or

Correlations in mathematics, natural sciences:

\[ \langle AB \rangle \neq \langle A \rangle \langle B \rangle \]

e.g., densities:

\[ \langle \rho(r)\rho(r') \rangle \neq \langle \rho(r) \rangle \langle \rho(r') \rangle \]

Correlations (I):

Effects beyond factorization approximations (e.g., Hartree-Fock)
Temporal/spatial correlations in everyday life

Beware: External periodic potential
Temporal/spatial 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 multiferroic effects

Technological applications:
- sensors, switches
- magnetic storage
- thermoelectrics
- functional materials, ...
Electronic Correlations: Models
$H = -t \sum_{\langle i,j \rangle, \sigma} c_{i\sigma}^\dagger c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}$

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
Dynamical Mean-Field Theory (DMFT) of Correlated Electrons
Theory of correlated electrons

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

Hubbard model

Face-centered cubic lattice (d=3)

Dynamical single-site mean-field theory

Müller-Hartmann (1989); Brandt, Mielsch (1989); Janiš (1991); Janiš, DV (1992)
Theory of correlated electrons

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

Hubbard model

Face-centered cubic lattice (d=3)

Strong simplifications

Self-consistent single-impurity Anderson model

Georges, Kotliar (1992)

Jarrell (1992)
DMFT self-consistency equations for $G$ and $\Sigma$

(i) Effective single impurity problem: local propagator

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

single-site ("impurity") action $A$

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

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

$\Rightarrow$ free electrons in a dynamic potential $\Sigma(\omega)$

Solve with an "impurity solver“, e.g., QMC, NRG, ED,…

$\Sigma(\omega)$: mean field ("dynamical potential“)
Dynamical mean-field theory (DMFT) of correlated electrons

Proper time resolved treatment of local electronic interactions

“Spectral transfer”

Experimentally detectable?
Application of DMFT: Mott-Hubbard metal-insulator transition
1. Squeezable nanocrystal film switching between metal and insulator

Collier, Saykally, Shiang, Henrichs, Heath (1997)

Uncompressed film: insulator

Compressed film: metal
(metallic sheen)

Discontinuous transition
2. Mott metal-insulator transition in $\text{V}_2\text{O}_3$

McWhan et al. (1971)
Spectral weight

Hubbard model, n=1

Correlations (II): Interaction effects leading to transfer of spectral weight
DMFT: Metal-insulator transition in the one-band Hubbard model

1994

Rozenberg, Kotliar, Zhang (1994)

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

Blümer, Dissertation 2002
Universality due to Fermi statistics

Mott-Hubbard metal-insulator transition

Hubbard model (n=1)

Strongly correlated electron materials: \( V_2O_3 \), \( NiSe_{2-x}S_x \), \( \kappa \)-organics, ...

Kotliar, DV (2004)

Universality due to Fermi statistics
**Mott-Hubbard metal-insulator transition**

Kotliar, DV (2004)

**Hubbard model (n=1)**

Strongly correlated electron materials:
- $V_2O_3$
- NiSe$_{2-x}$S$_x$
- $\kappa$-organics, ...

4-site cluster-DMFT

Park, Haule, Kotliar (2008)
Correlated Electron: Materials
### How to combine?

**DFT/LDA**

| + | material specific: “ab initio” |
| - | fails for strong correlations |
| + | fast code packages |

**Model Hamiltonians**

| - | input parameters unknown |
| + | systematic many-body approach |
| - | computationally expensive |

---

**Held (2004)**

- Time-averaged electron density
- Lattice potential

---

**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

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, ...) or GW

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

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

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

Crystal structure

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

$\downarrow$

orthorhombic distortion

$\downarrow$

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

LDA density of states

SrVO$_3$

- V-3d($t_{2g}$)
- V-3d($e_g$)

CaVO$_3$

- V-3d($t_{2g}$)
- V-3d($e_g$)

No correlation effects/spectral transfer
LDA+DMFT results

SrVO$_3$ / CaVO$_3$

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

Mott-Hubbard system

Osaka - Augsburg - Ekaterinburg collaboration:

Measure by electron spectroscopy
Comparison with experiment


Bulk sensitive photoemission spectroscopy
→ occupied states: $A(E) \cdot f(E,T)$

X-ray absorption spectroscopy
→ unoccupied states: $A(E) \cdot [1 - f(E,T)]$

3-peak structure detected
State-of-the-art LDA+DMFT: Electronic correlations & structural transformations

Electron correlations can induce structural transformations

Electronic correlations at the $\alpha-\gamma$ structural phase transition in paramagnetic iron

I. Leonov,¹ A. I. Poteryaev,² V. I. Anisimov,² and D. Vollhardt¹ (2011)
Incorporating Strong Correlations into the Design of Transport Properties of Nuclear Fuel Materials

Quan Yin, Andrey Kutepov, Kristjan Haule, and Gabriel Kotliar
Department of Physics and Astronomy, Rutgers University, Piscataway, NJ 08854

Sergey Y. Savrasov and Warren E. Pickett
Department of Physics, University of California, Davis, CA 95616
(Dated: December 14, 2010)

arXiv:1012.2412
Perspective 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
1, 2, ... multi-electron transfer in metalloprotein complexes

→ Photosynthesis
Goal: Dynamical mean-field approach with predictive power for strongly correlated materials
Other Developments & Perspectives
1. Quantum phase transitions

Driven by quantum fluctuations

Field induced quantum phase transition

Driven by quantum fluctuations

- Non-Fermi liquid behavior
- Emergence of novel degrees of freedom
Driven by quantum fluctuations

- Non-Fermi liquid behavior
- Emergence of novel degrees of freedom

\[ [\rho(T) - \rho_0] \propto T^\epsilon \]


Si, Steglich (2010)
2. 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

Perfetti et al. (2006)
2. Correlated electrons in non-equilibrium

Non-equilibrium DMFT

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

Momentum distribution ($U=3.3$)

Time after quench

Thermalization

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

Eckstein, Kollar (2008)
Freericks, Krishnamurthy, Pruschke (2008)
3. 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 (\(^{40}\)K atoms) Köhl, Esslinger (2006)
3. Correlated cold atoms in optical lattices

Hubbard model with ultracold atoms
Jaksch, Bruder, Cirac, Gardiner, Zoller (1998)

Atomic total angular momentum $L^{\text{tot}} = F \rightarrow N = 2F + 1$ hyperfine states

$\rightarrow$ SU(N) Hubbard models

$N=3$, e.g. $^6\text{Li}$, $U<0$: Color superconductivity, “baryon formation (QCD)”
Correlated many-particle systems: More fascinating than ever