

Modeling of half-metallic double perovskites within the Dynamical Mean-field Theory (DMFT)

Nils Blümer

TP6, WA Prof. van Dongen (KOMET337)

Outline

Motivation: Physics of double perovskites

Introduction: DMFT and earlier DMFT-QMC results

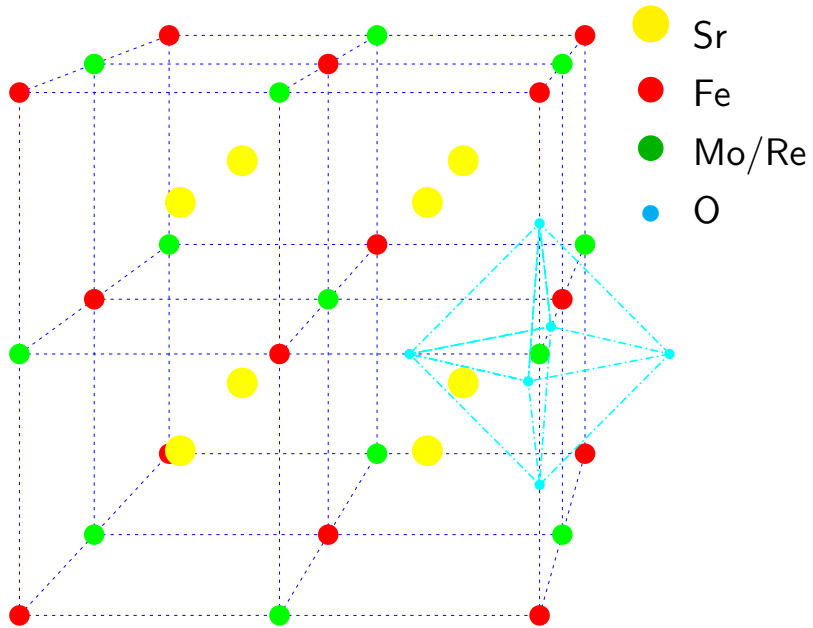
Modeling of $\text{Sr}_2\text{FeMoO}_6$, $\text{Sr}_2\text{FeReO}_6$

DMFT for double perovskites

Summary and Outlook

Motivation: Physics of double perovskites

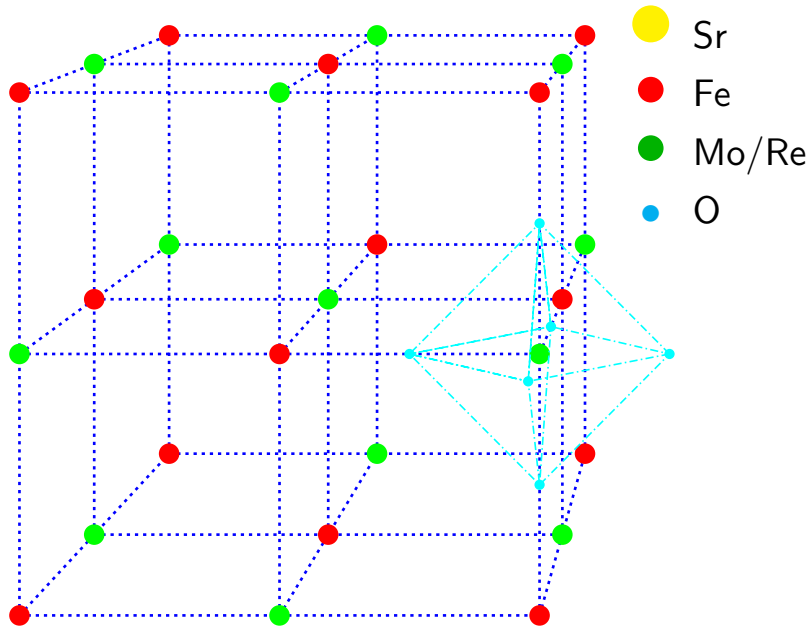
$\text{Sr}_2\text{FeMoO}_6$ and $\text{Sr}_2\text{FeReO}_6$



Valences: Sr^{2+} [Kr]
 Fe^{3+} [Ar] $3d^5$
 Mo^{5+} [Kr] $4d^1$
 O^{2-} [Ne]

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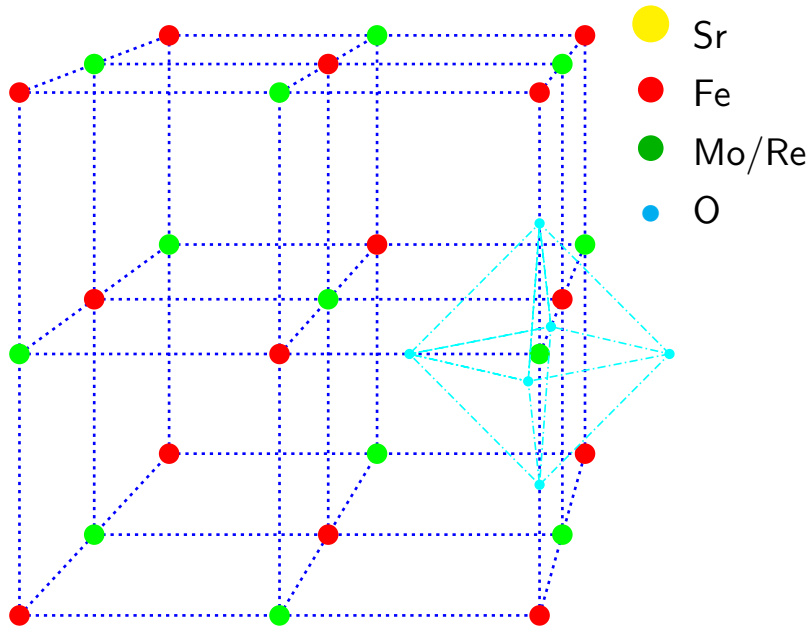
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Cubic symmetry \rightsquigarrow 3 degenerate t_{2g} bands,
2 degenerate e_g bands

Hybridization Fe - O - Mo/Re

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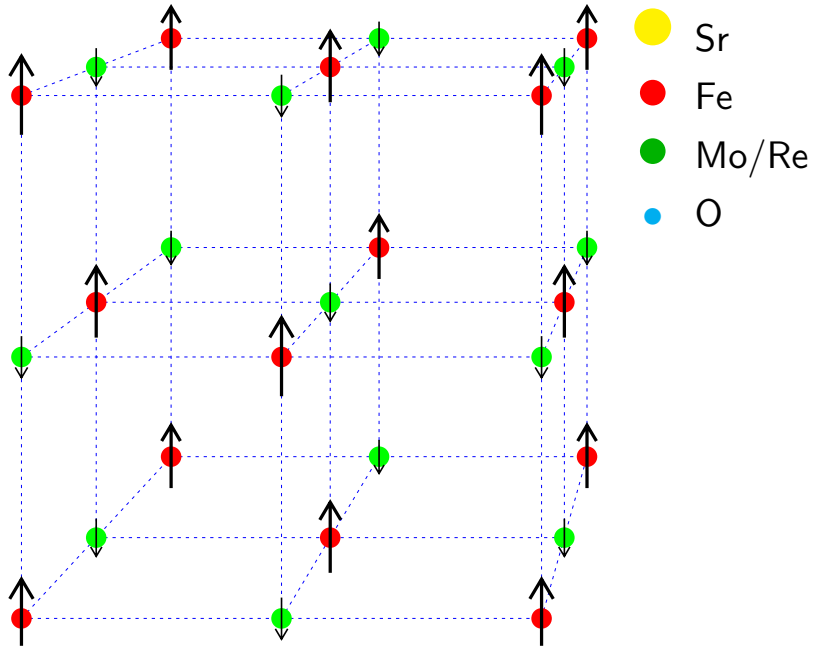
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Net **ferromagnetic** spin polarization, $T_c \approx 400$ K

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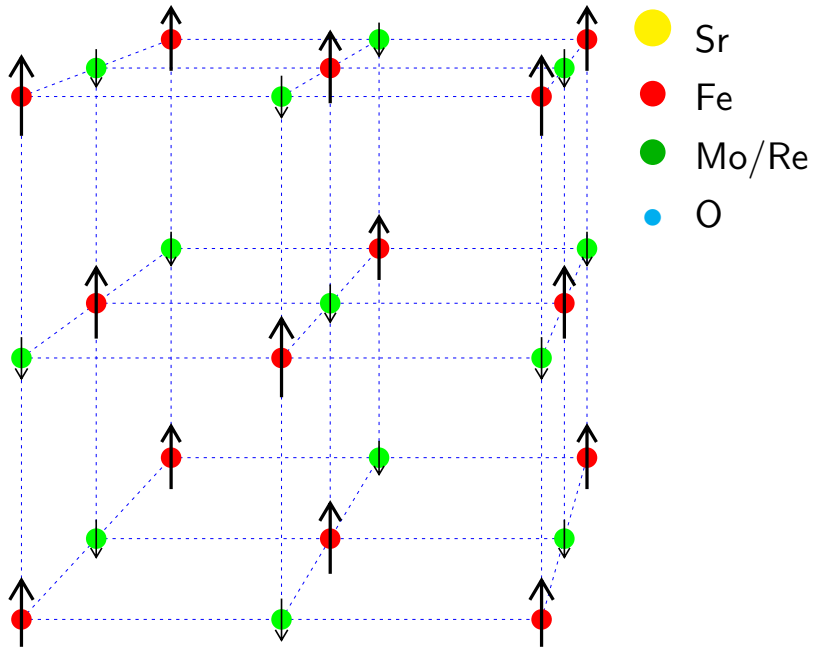
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Calculations: opposite (**AF**) spin polarization of Fe and Mo/Re \rightsquigarrow total system **ferrimagnetic**

Large magnetoresistance for ordered probes

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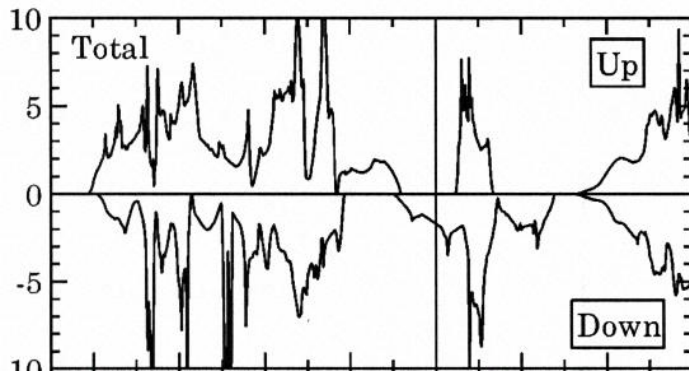
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Calculations/heuristics: systems half-metallic
 (\sim Mott insulator)

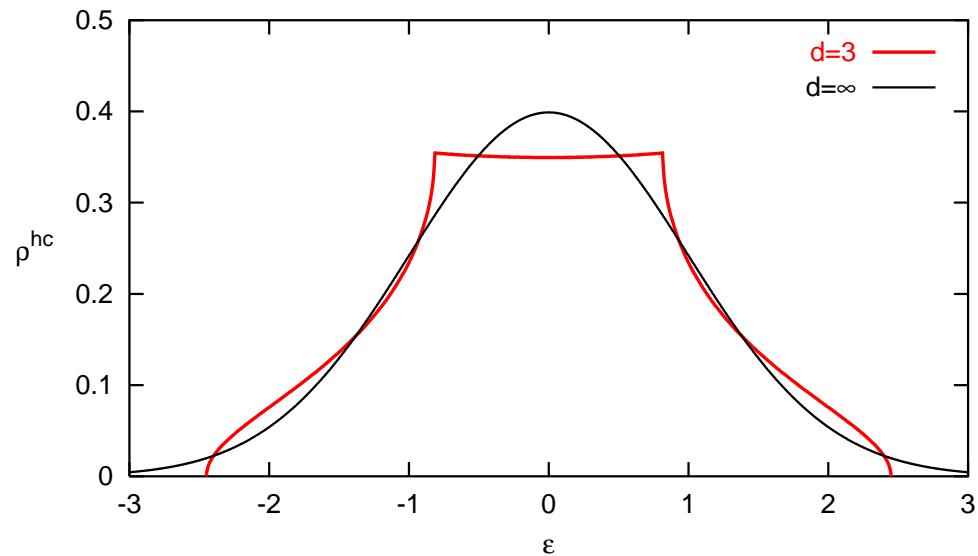


Introduction: Brief History of DMFT

1989 Metzner/Vollhardt: $d \rightarrow \infty$ nontrivial for fermions if $t = t^*/\sqrt{2d}$
Gutzwiller approximation exact

Müller-Hartmann: local self-energy: $\Sigma(\mathbf{q}, \omega) \rightarrow \Sigma(\omega)$

Brandt/Mielisch: exact solution of Falicov-Kimball model for $d = \infty$



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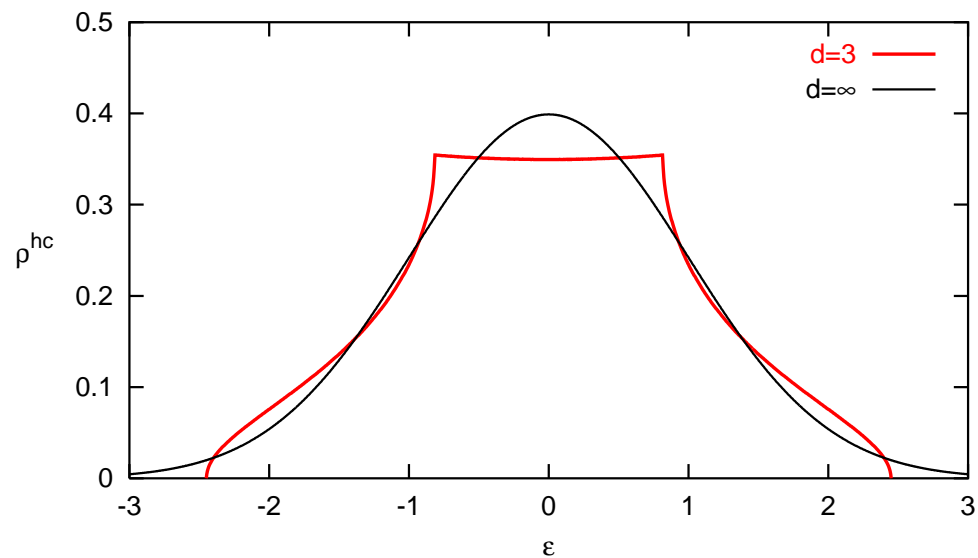
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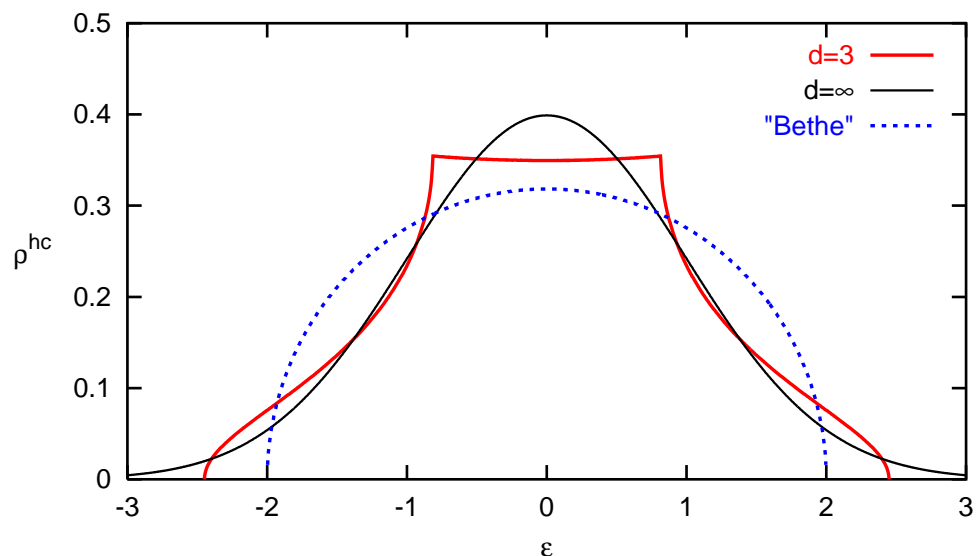
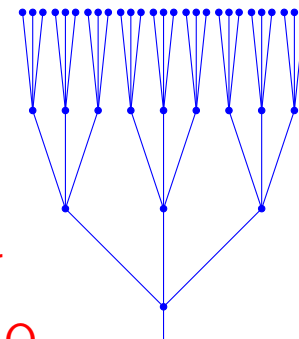
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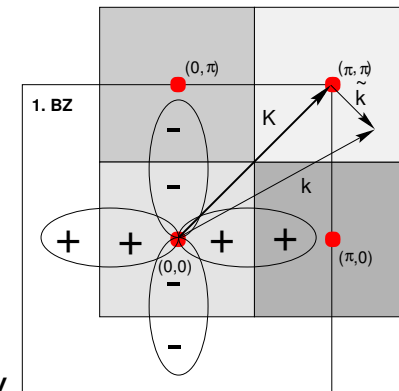
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Vollhardt group: ferromagnetism (NN exchange, asymmetric DOS, multi-band)

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 Jarrell group: dynamical cluster approximation (DCA) \rightsquigarrow d -wave superconductivity

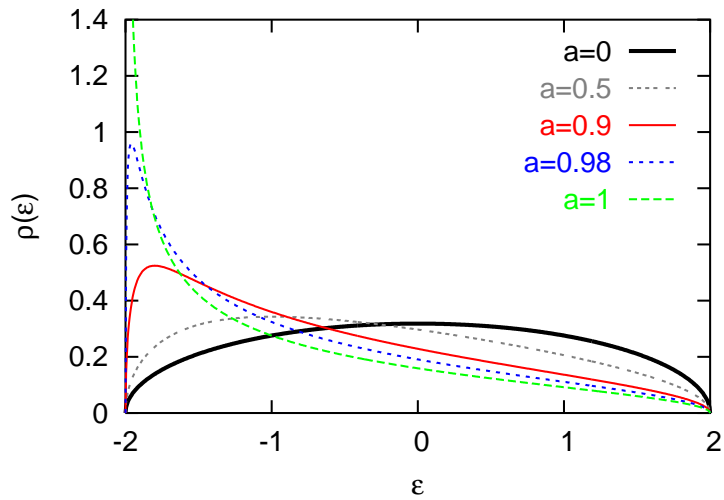


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- 2004 **Physics Today** article; lecture notes; conferences “beyond LDA” etc.
- Future:** complete understanding of abstract models (within DMFT)
realistic calculations (LDA+DMFT)
cluster methods (DCA, CDMFT)

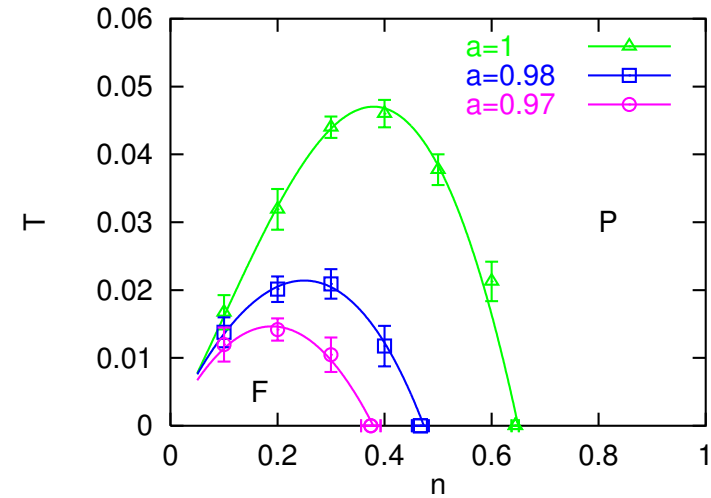
Earlier DMFT-QMC results

Ferromagnetism: asymmetric DOS



Asymmetric model density of states (DOS)

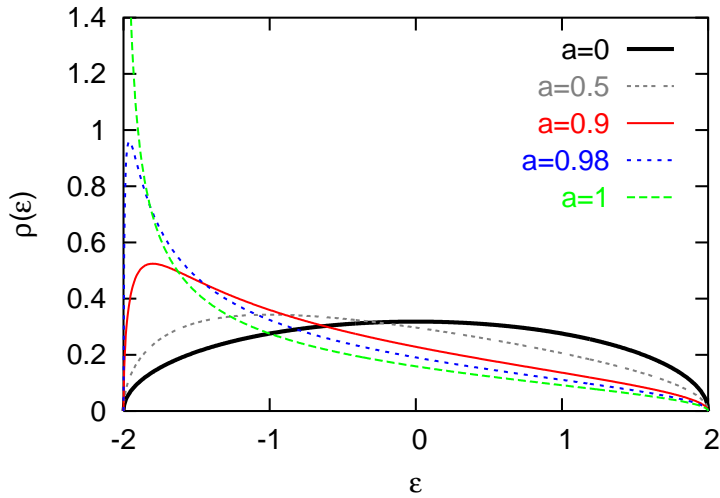
High DOS at E_F good for ferromagnetism!



Curie temperature T_C vs. doping n for $U = W = 4$

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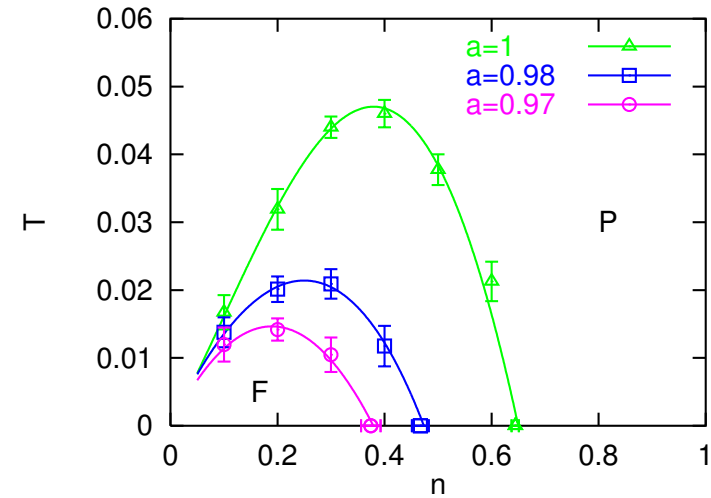


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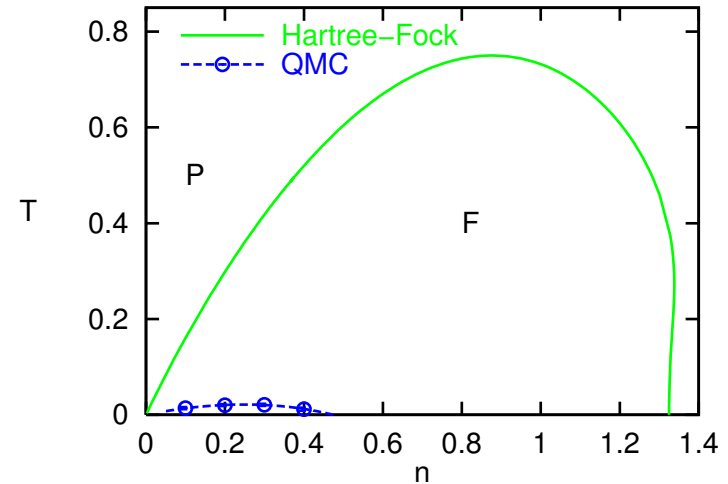
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But: Stoner criterion useless!

[J. Wahle, NB, J. Schlipf, K. Held, D. Vollhardt (1998)]



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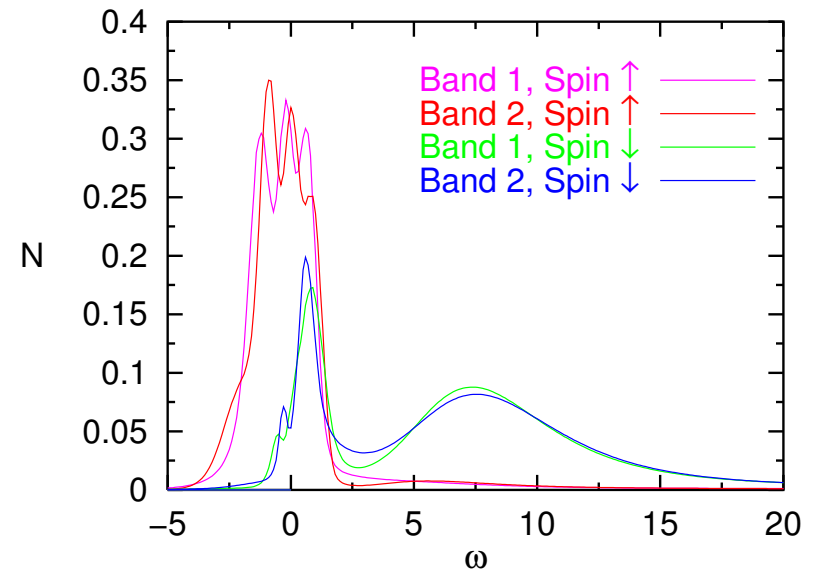
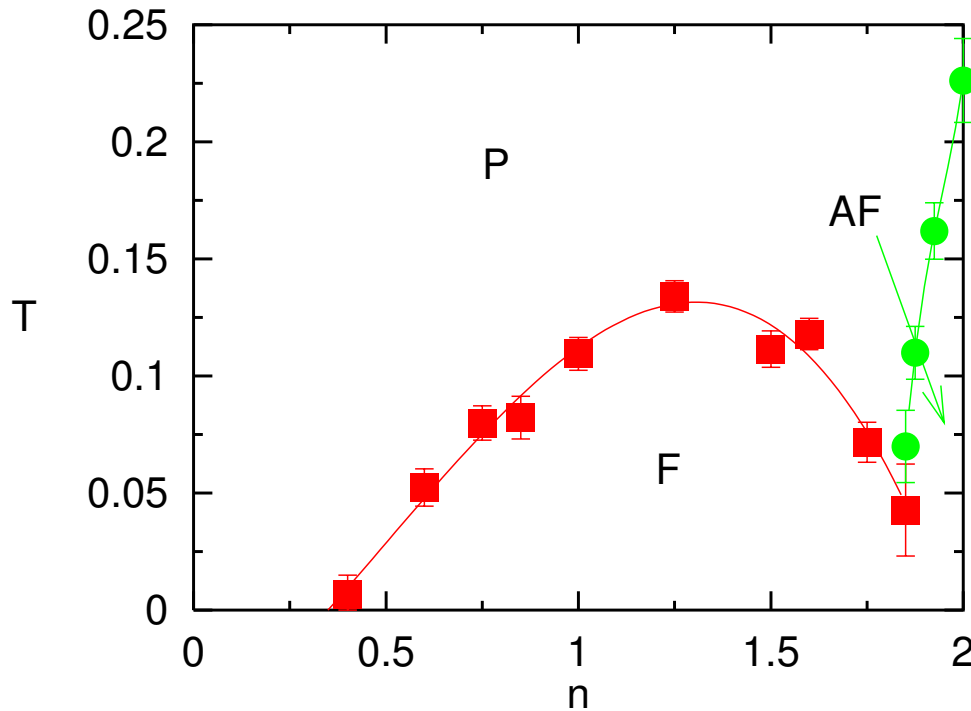
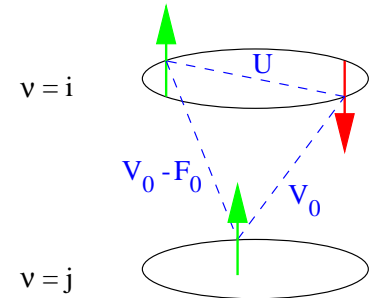


T_C vs n for $U = W = 4$ and $a = 0.98$

Ferromagnetism: Hund's rule coupling

Here: 2 bands, semi-elliptic DOS

$$\hat{H} = \left[\hat{H}_{\text{Hub}} \right]_{\nu=1} + \left[\hat{H}_{\text{Hub}} \right]_{\nu=2} + \sum_{i \sigma \sigma'} (V_0 - \delta_{\sigma \sigma'} F_0) \hat{n}_{i, \nu=1, \sigma} \hat{n}_{i, \nu'=2, \sigma'}$$



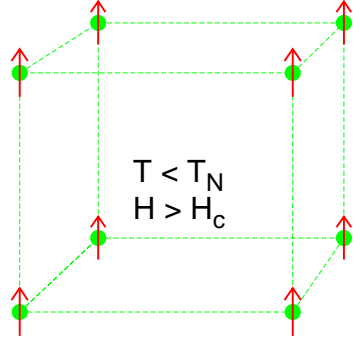
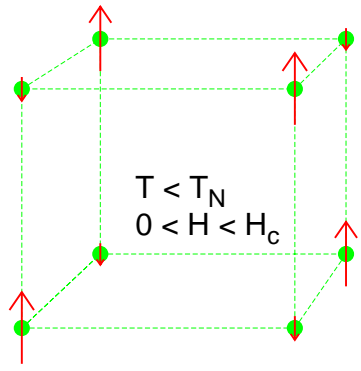
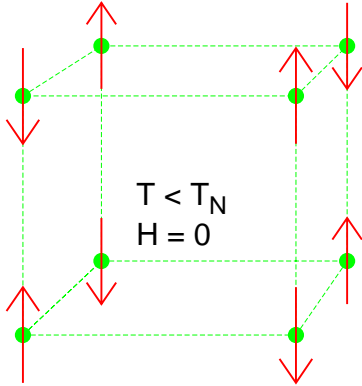
T_C , T_N vs doping n for $U = 9$, $F_0 = 4$, $V_0 = 5$

Not shown: Hartree-Fock completely incorrect

[K. Held, D. Vollhardt (1998)]

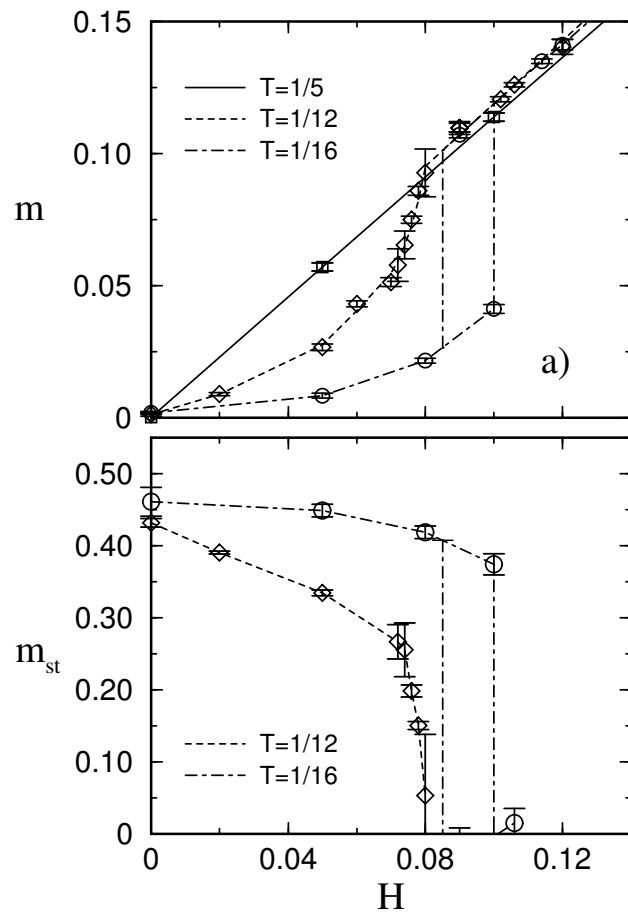
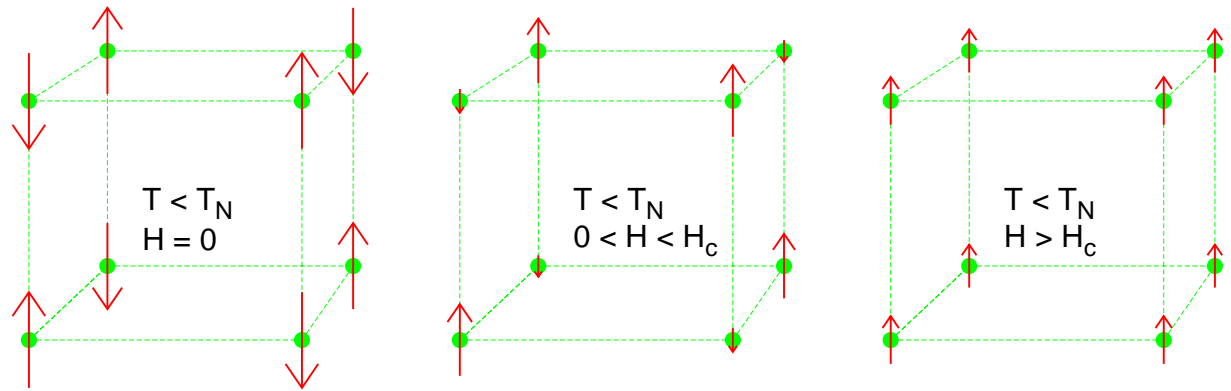
Spectra for $n = 1.25$, $T = 0.09$

Metamagnetismus



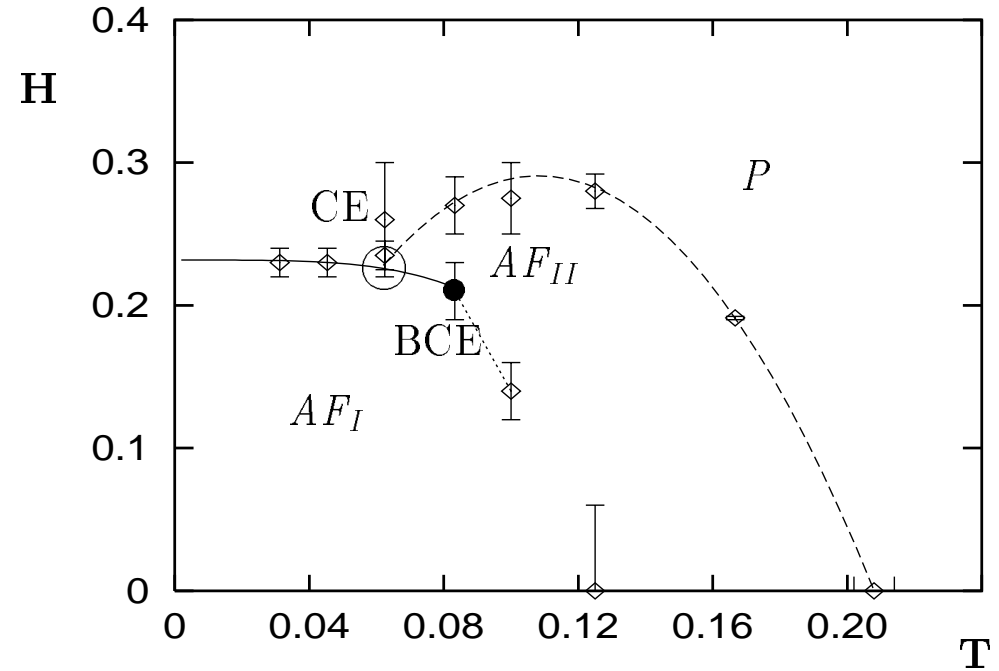
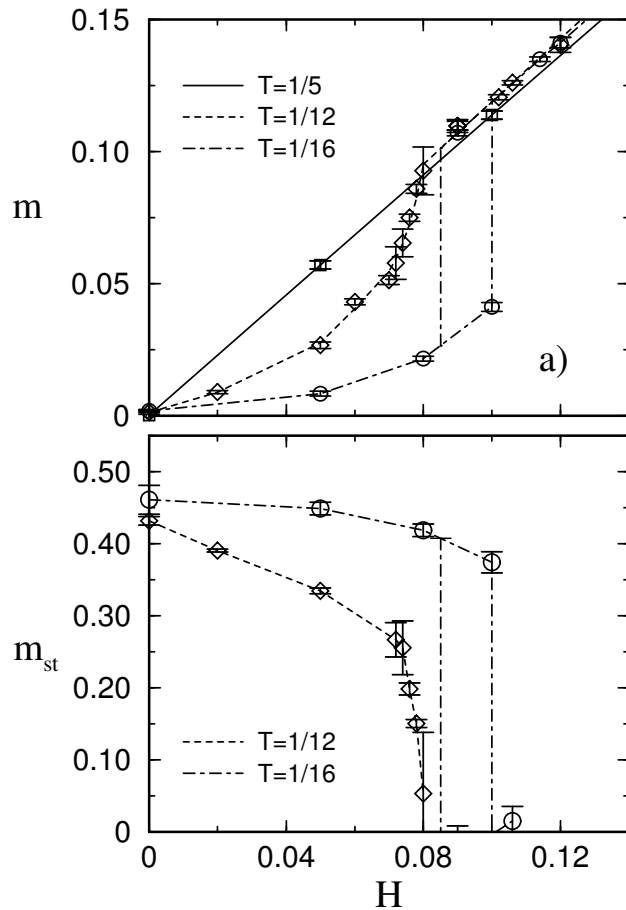
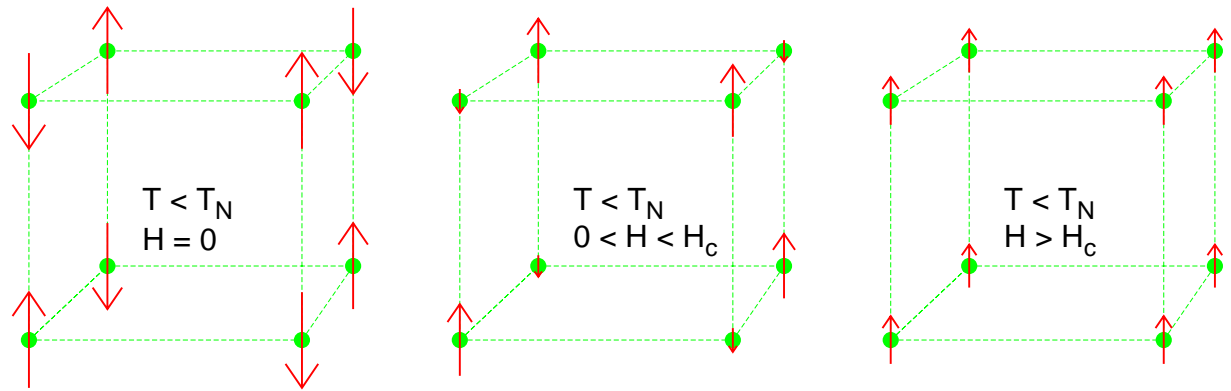
Metamagnetism

Hubbard model with easy axis,
 $n = 1, U = 2$



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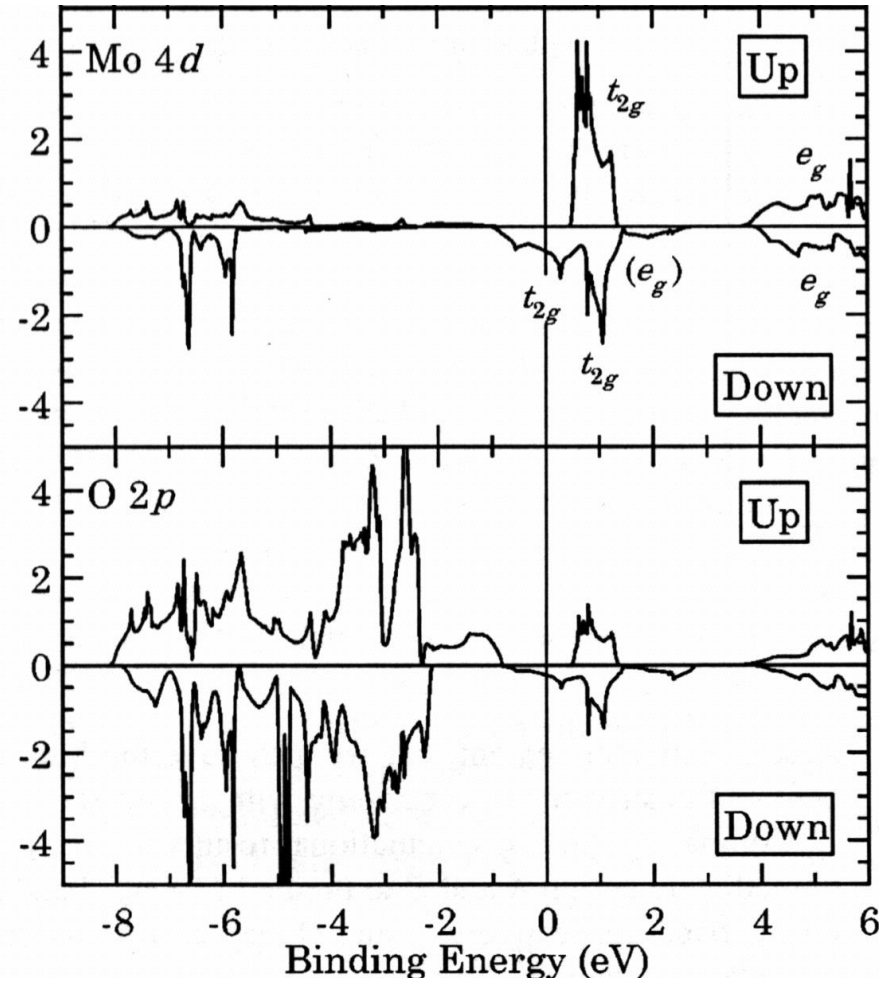
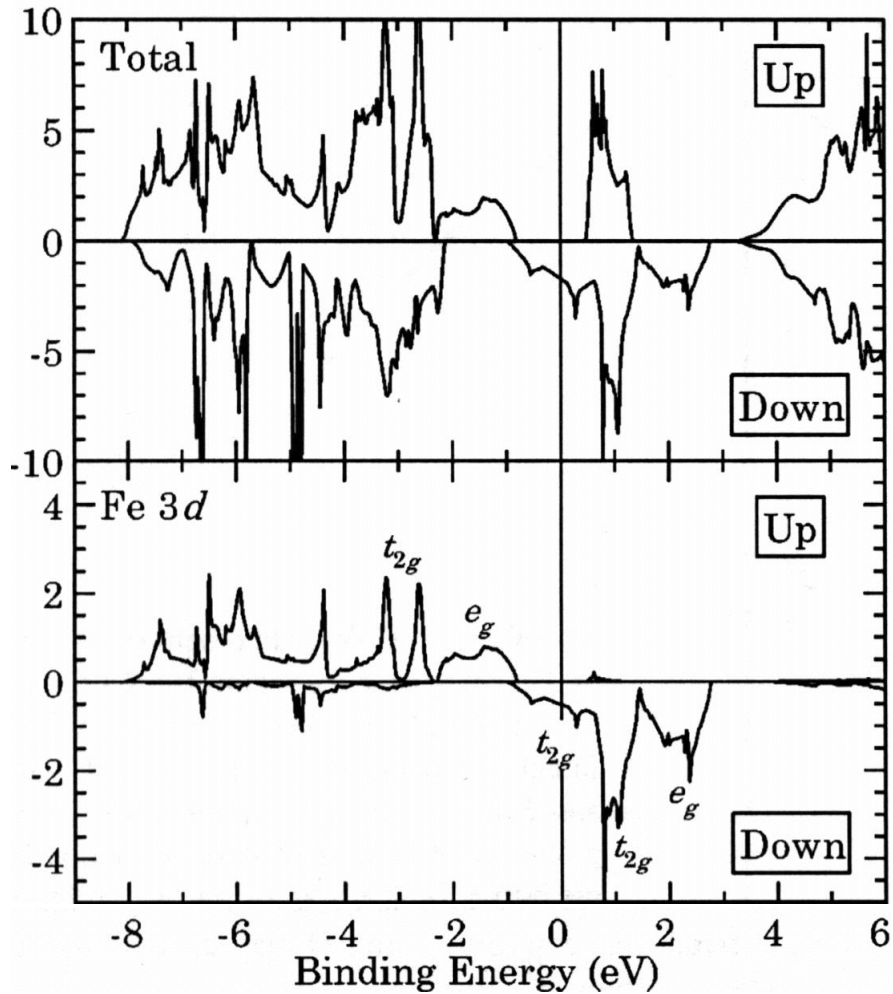


Only DMFT-QMC captures 1st and 2nd order!

[K. Held, M. Ulmke, NB, D. Vollhardt (1997)]

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LDSA+U for $\text{Sr}_2\text{FeMoO}_6$



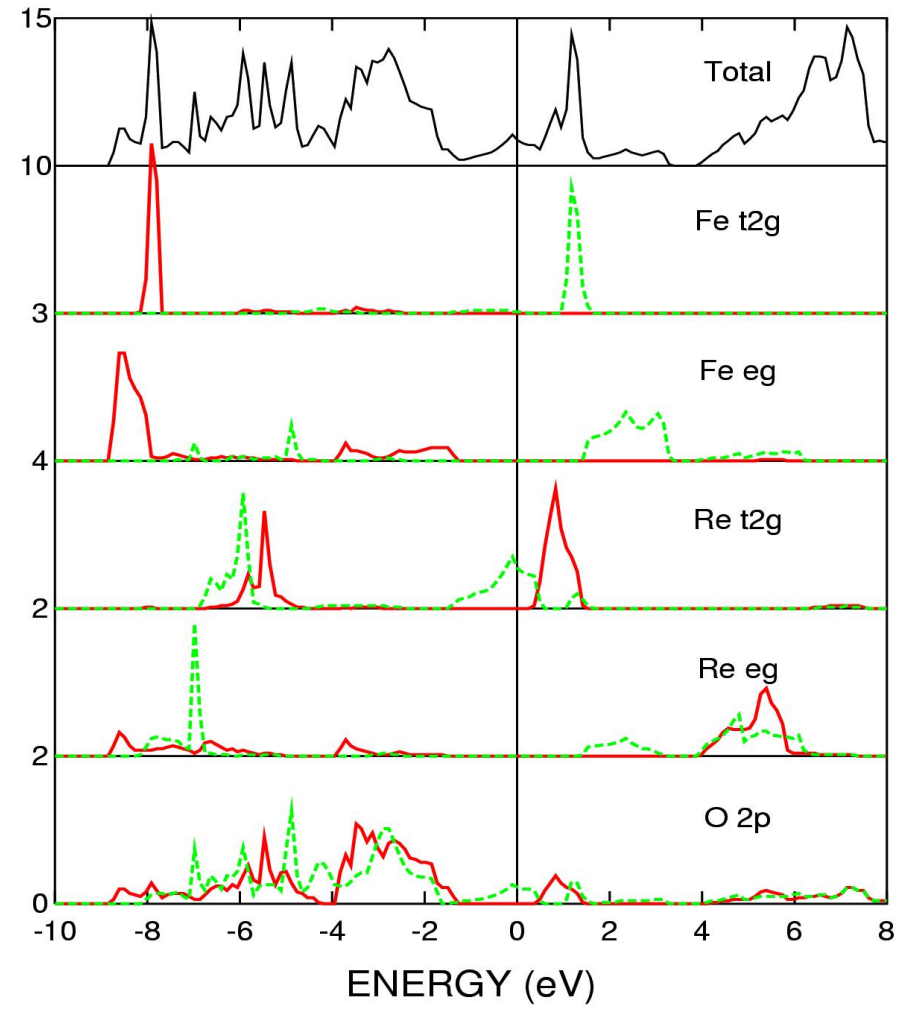
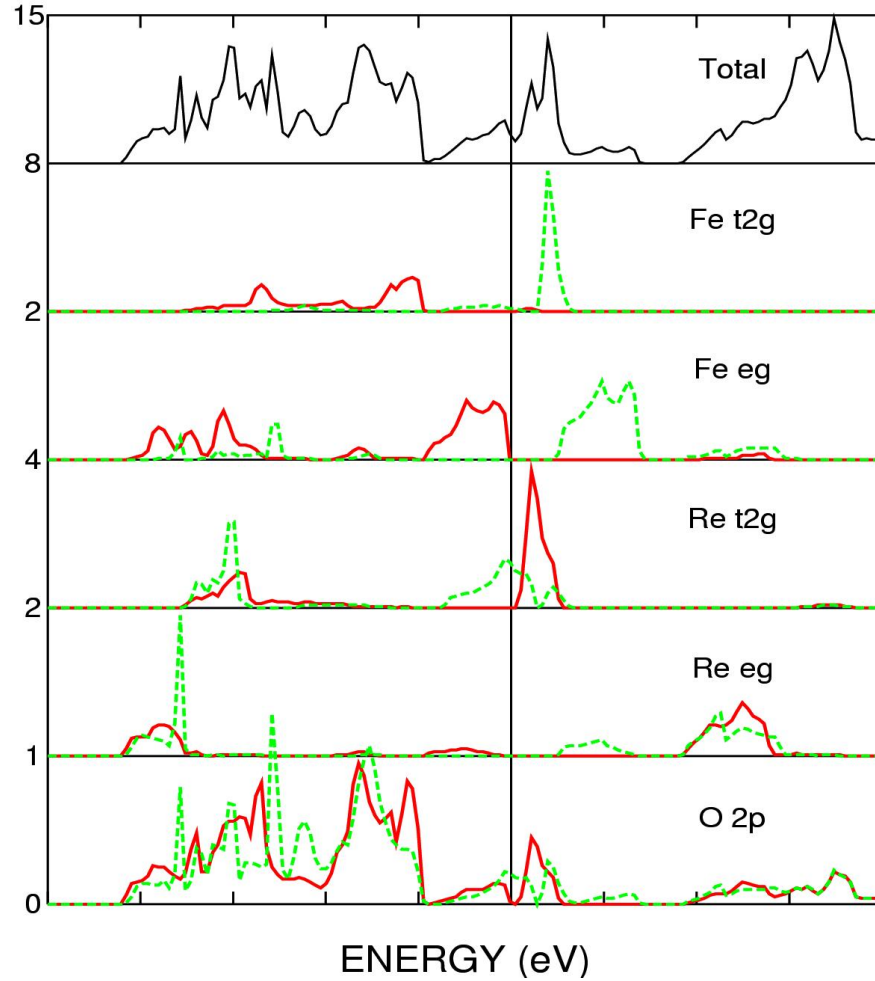
[Saitoh et al. (2002)]

$\text{Sr}_2\text{FeReO}_6$:

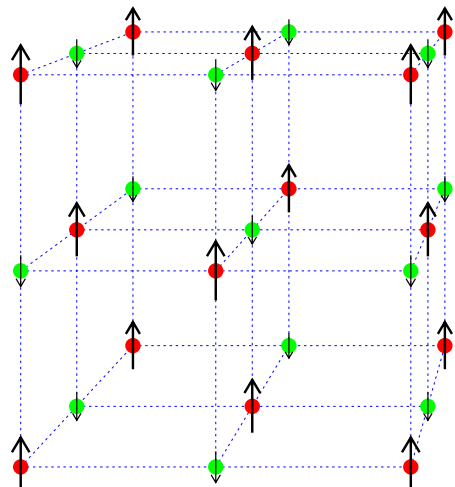
LSDA

vs.

LDSA+U

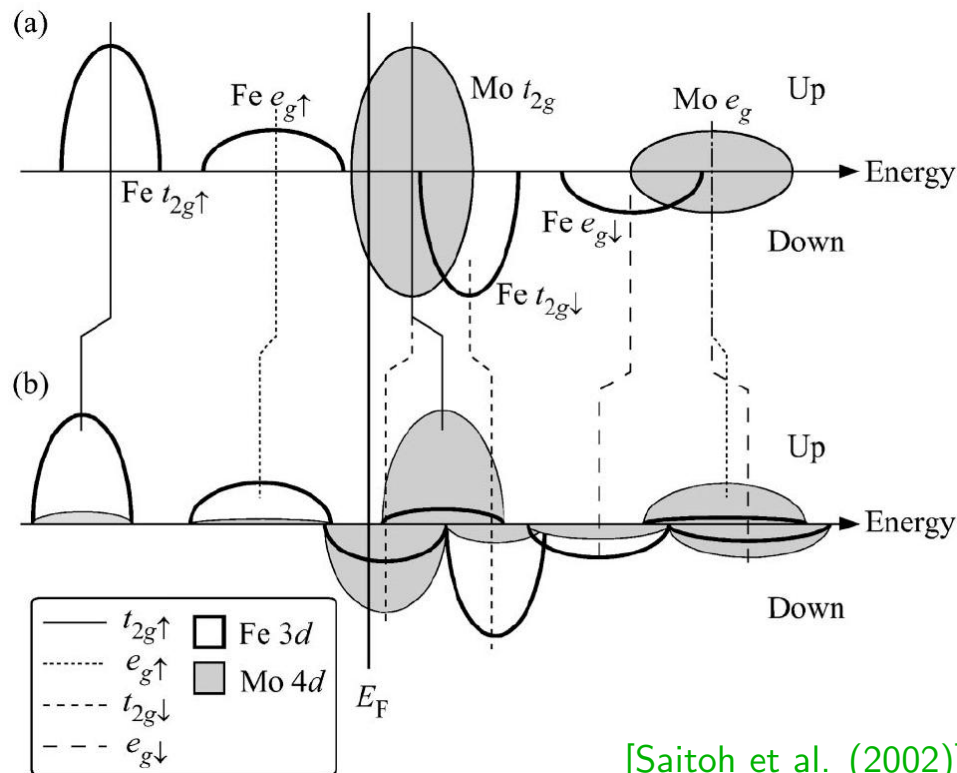


[Wu et al. (2001)]



Microscopic theory (5-band model)

Scheme: hybridization of hypothetical ionic states



[Saitoh et al. (2002)]

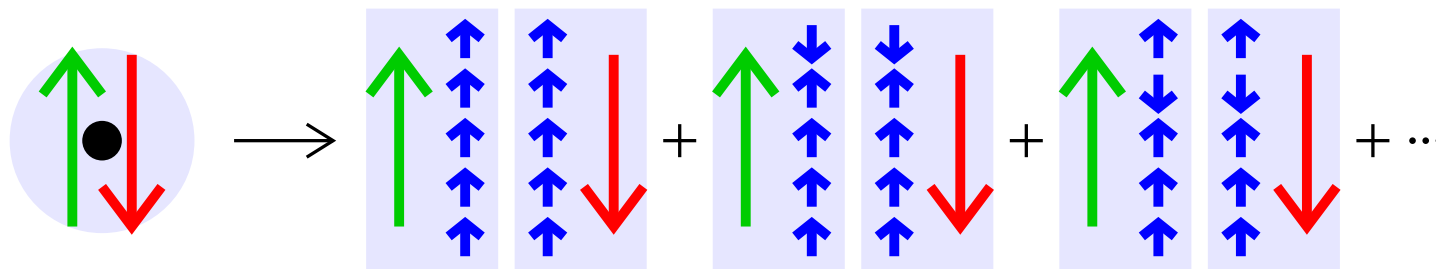
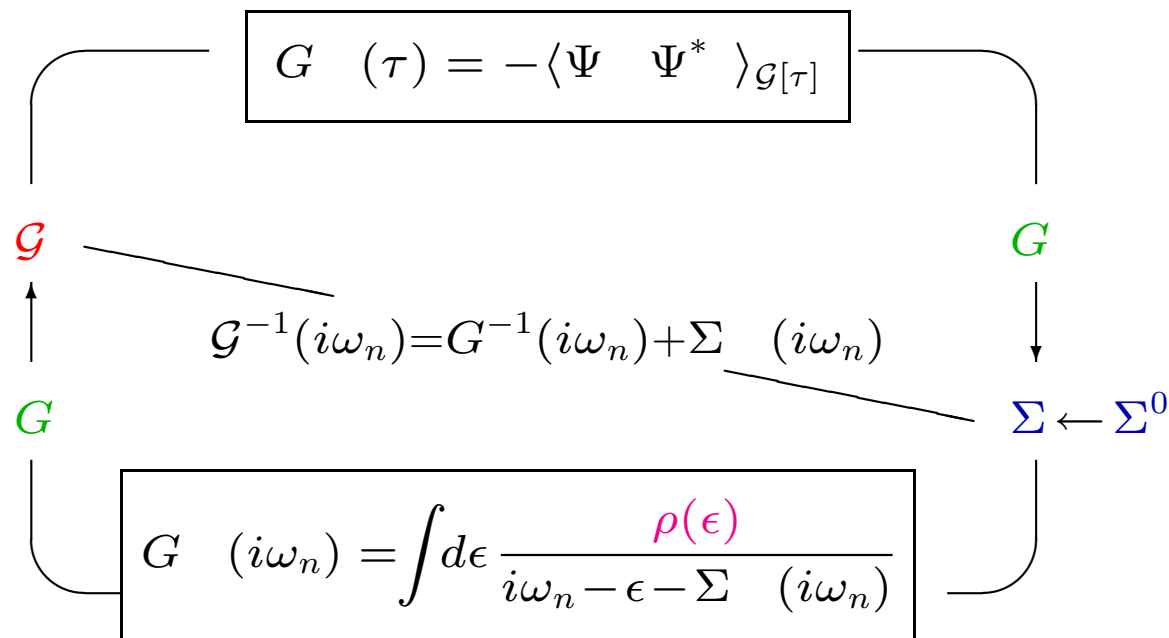
$$\begin{aligned}
 H = & \epsilon_f \sum_{i\nu\sigma} n_{i\nu\sigma}^f + \epsilon_m \sum_{i\nu\sigma} n_{i\nu\sigma}^m + \sum_i U_{\nu\sigma\nu'\sigma'}^f n_{i\nu\sigma}^f n_{i\nu'\sigma'}^f + \sum_i U_{\nu\sigma\nu'\sigma'}^m n_{i\nu\sigma}^m n_{i\nu'\sigma'}^m \\
 & + \sum_{\langle ij \rangle \nu\sigma} t_{fm} \left(f_{i\nu\sigma}^\dagger m_{j\nu\sigma} \right) + \sum_{\langle jj' \rangle \nu\sigma} t_{mm} \left(f_{j\nu\sigma}^\dagger m_{j'\nu\sigma} \right) + \sum_{\langle ii' \rangle \nu\sigma} t_{ff} \left(f_{i\nu\sigma}^\dagger m_{i'\nu\sigma} \right)
 \end{aligned}$$

$$t_{fm} \approx 0.25\text{eV} \quad > \quad t_{mm} \approx 0.15\text{eV} \quad \gg \quad t_{ff} \approx 0.03 \text{ eV} \quad \text{[Phillips et al. (2003)]}$$

DMFT for double perovskites

Self-consistency equations

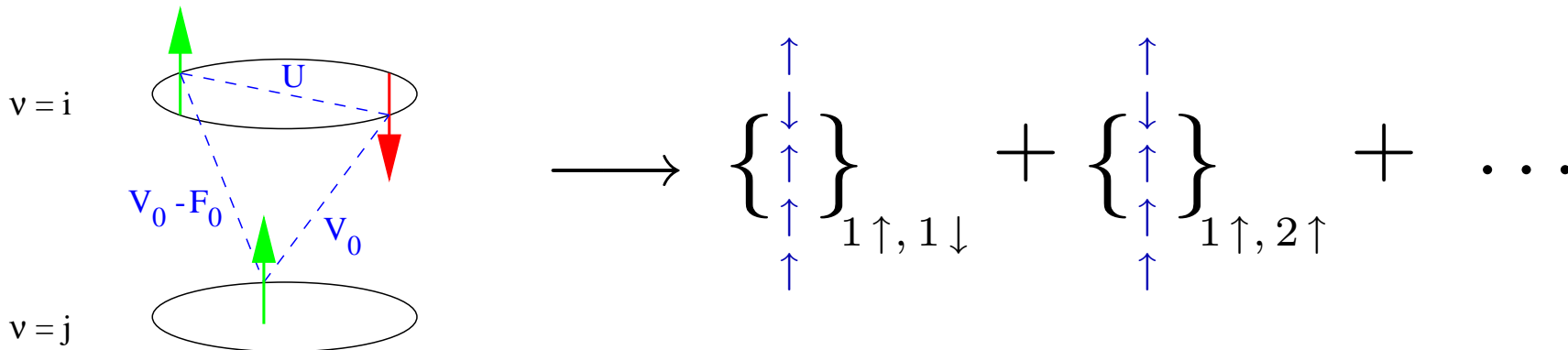
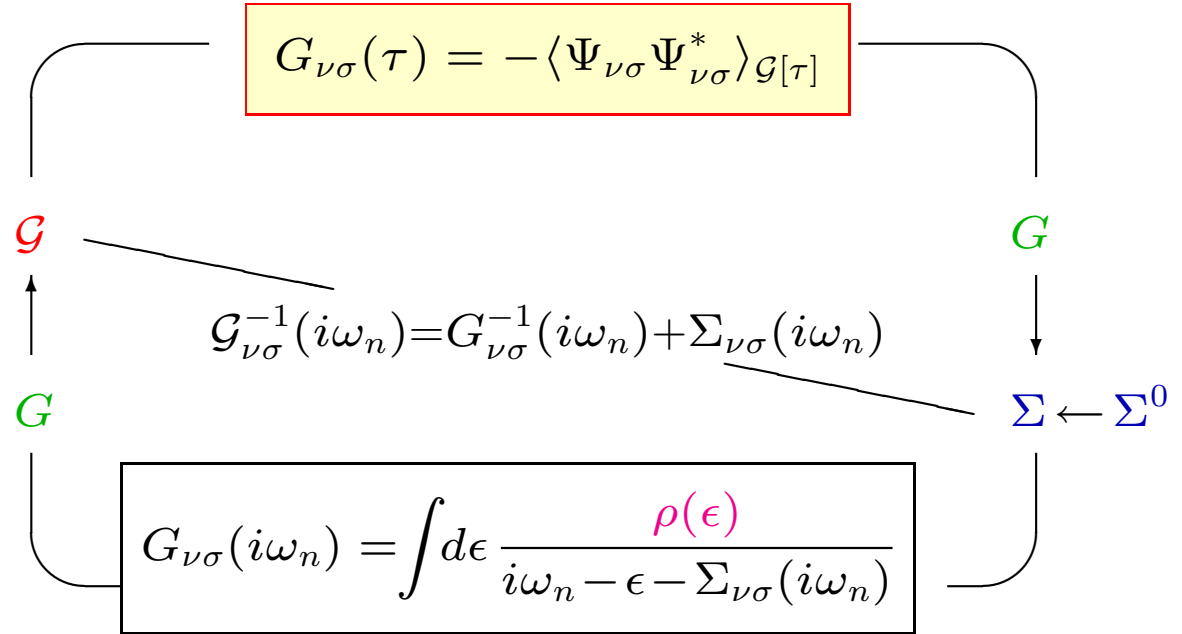
1. Single band, homogeneous



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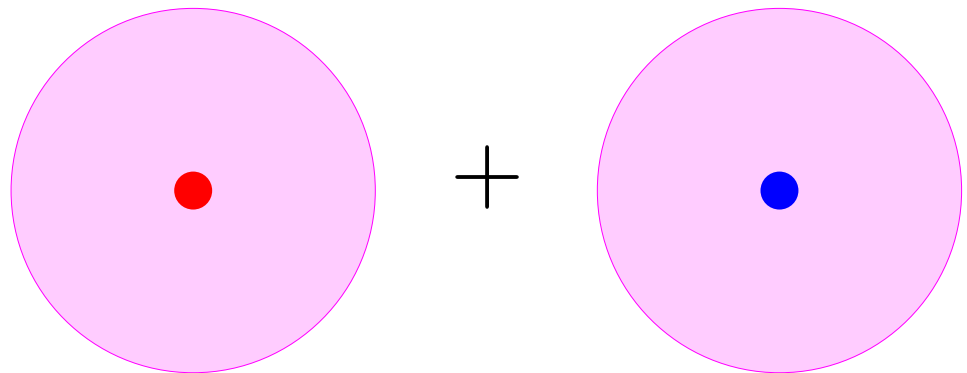
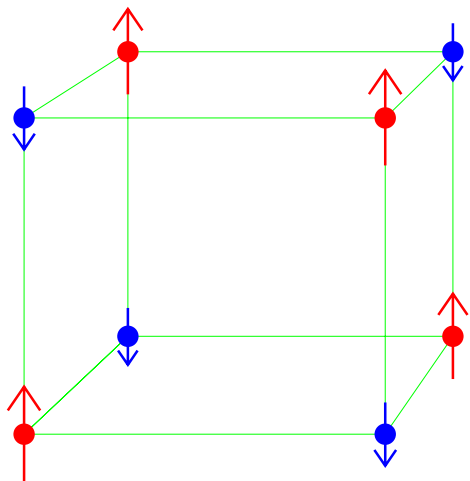
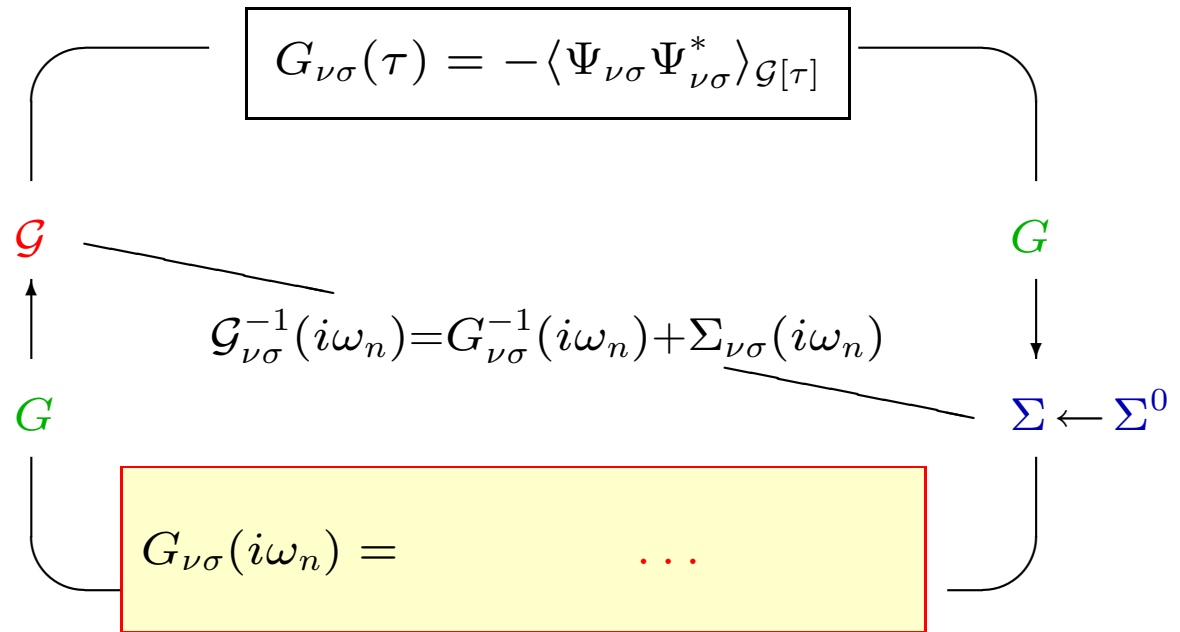
1. Single band, homogeneous
2. Multi-band, homogeneous



DMFT for double perovskites

Self-consistency equations

1. Single band, homogeneous
2. Multi-band, homogeneous
3. Multi-band, $n_i \neq n_j$, $n_\uparrow \neq n_\downarrow$



Generalized Dyson equation

Bravais lattice, Hubbard model, homogeneous phase:

$$G(i\omega_n) = \int d\epsilon \frac{\rho(\epsilon)}{i\omega_n - \epsilon - \Sigma(i\omega_n)}$$

Bravais lattice, Hubbard model, AF or ferrimagnetic phase:

$$G_\alpha(i\omega_n) = \int d\epsilon \frac{\rho(\epsilon)}{i\omega_n - \epsilon^2 / (i\omega_n - \Sigma_{\bar{\alpha}}) - \Sigma_\alpha(i\omega_n)}$$

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Bravais lattice, periodic Anderson model (PAM), on-site hybridization:

$$G_f(i\omega_n) = \int d\epsilon \frac{\rho(\epsilon)}{i\omega_n - t_{df}^2 / (\omega - \epsilon) - \Sigma_f(i\omega_n)}$$

Bravais lattice, periodic Anderson model (PAM), NN hybridization:

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Strategy: Treat correlations for Fe only (\rightsquigarrow multi-band “PAM”)

First: calculations for fixed e_g spin (\rightsquigarrow 3-band model + internal magnetic field)

Then: 5-band model