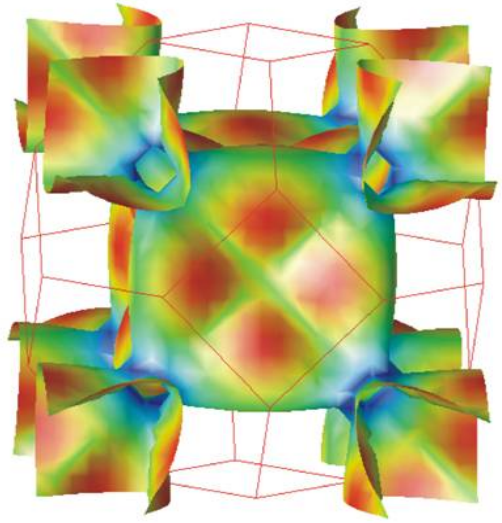
TECHNISCHE UNIVERSITÄT KAISERSLAUTERN

JOHANNES GUTENBERG UNIVERSITÄT MAINZ

Theory of materials with high spin polarization

Nils Blümer

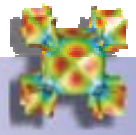


Outline

- Introduction: strong electronic correlations in double perovskites and Heusler compounds
- Theoretical approaches: DFT/LDA and DMFT
- High-precision DMFT-QMC algorithm
- Status/results: Heusler compounds
- Status/results: double perovskites
- Summary and outlook

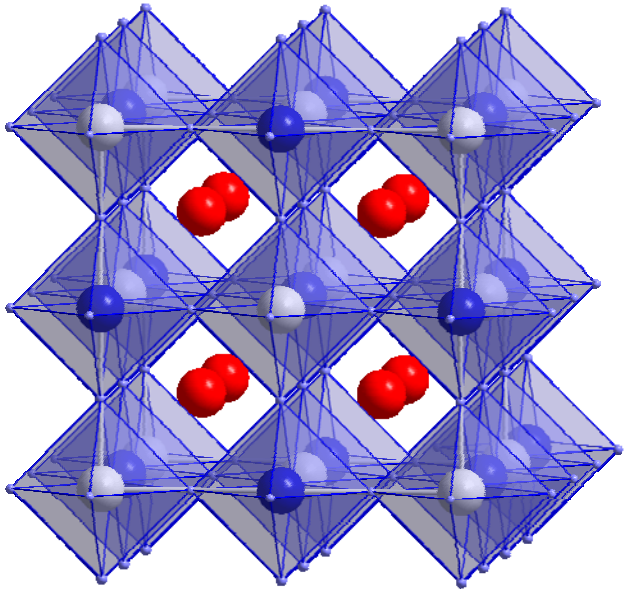
P6 Blümer, van Dongen

P7 Schönhense, Felser



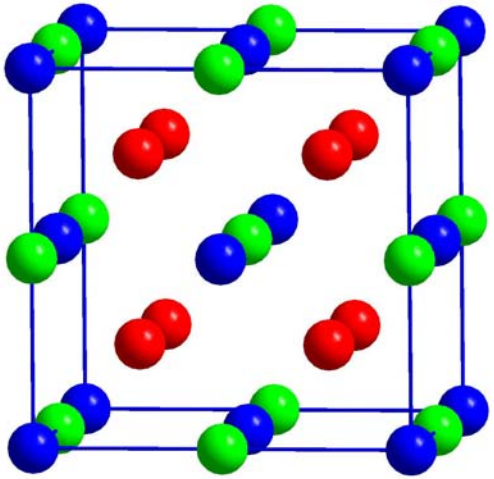
Introduction: strong electronic correlations

double perovskites



A_2XYO_6

Heusler compounds



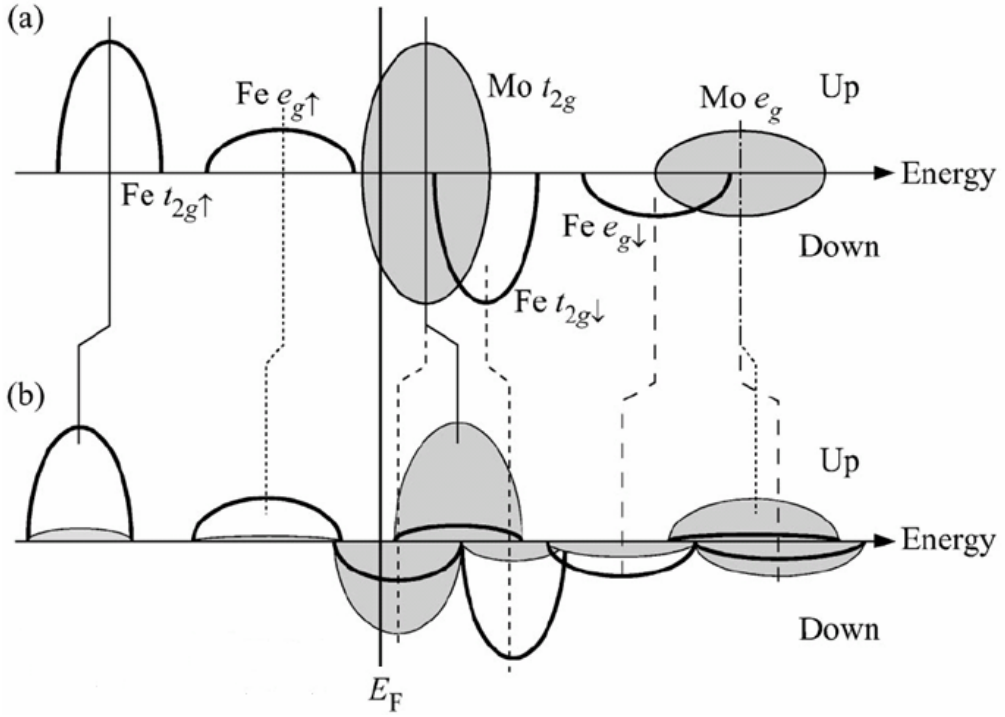
X_2YZ

Strong correlations in double perovskites

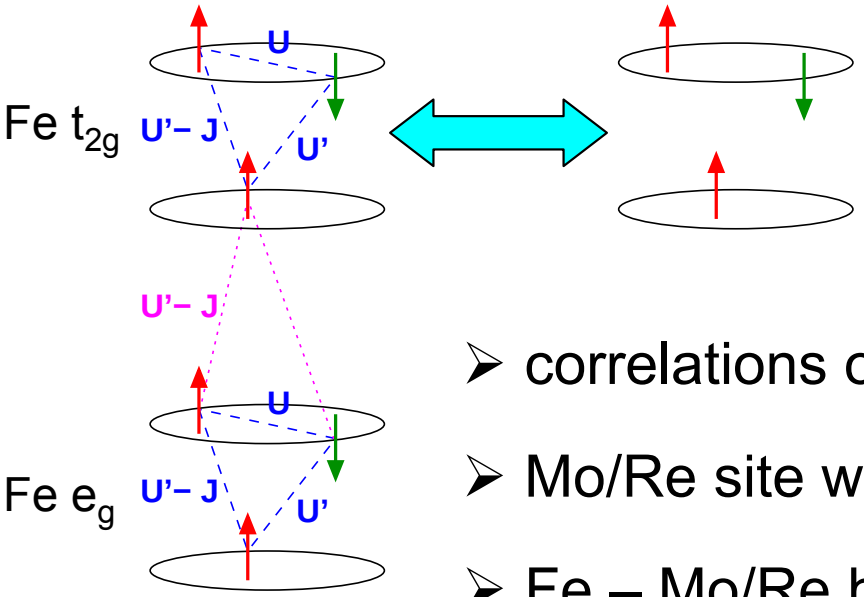
Schematic electronic structure of Sr_2FeMoO_6:

a) hypothetical ionic state,

b) Fe-Mo hybridized state.



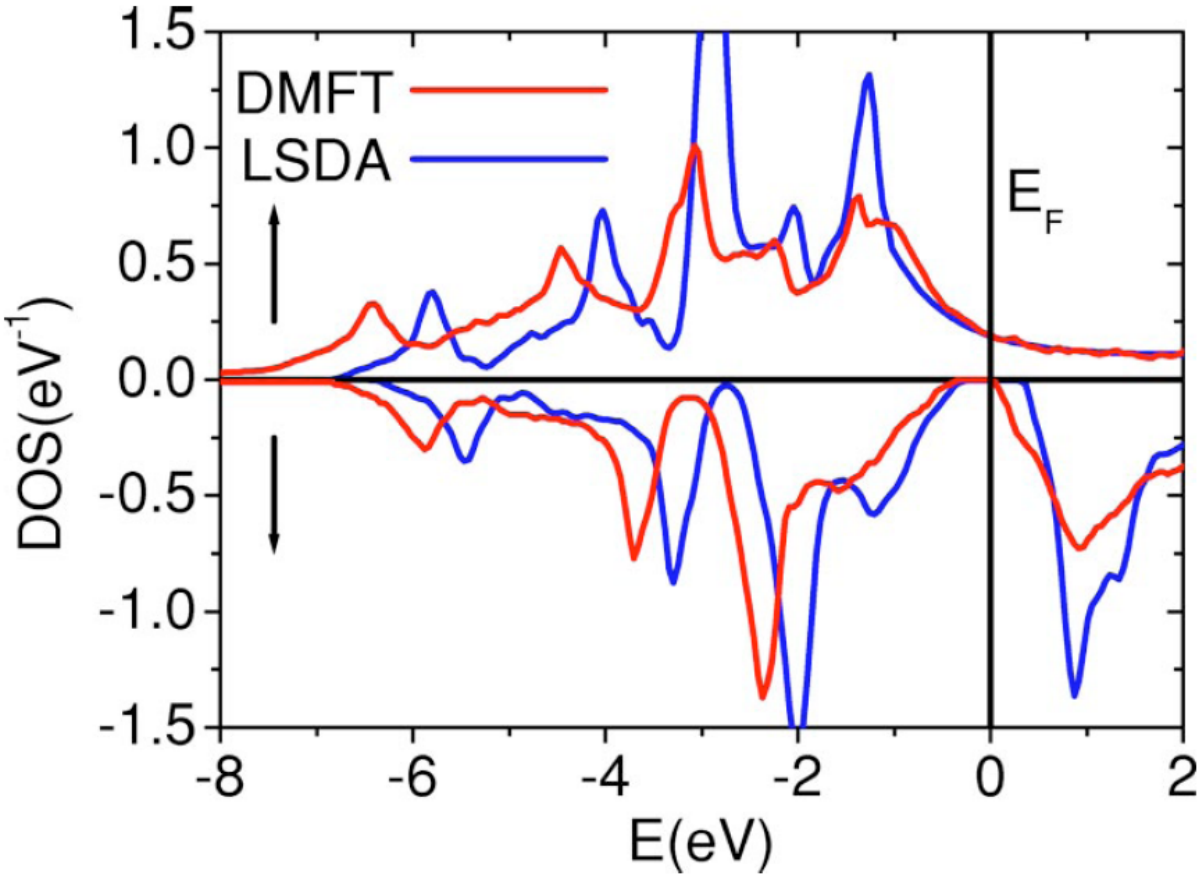
[Saitoh et al., PRB (2002)]



- correlations on Fe site → magnetic, „insulating“ state
- Mo/Re site weakly correlated → no intrinsic magnetism
- Fe – Mo/Re hybridization → ferrimagnetic metal

Strong correlations in Heusler compounds

Early hint: non-quasiparticle states in NiMnSb
[Chioncel, Katsnelson, de Groot, Lichtenstein (2003)]



Correlations impact half-metallic properties stronger than spin-orbit couplings

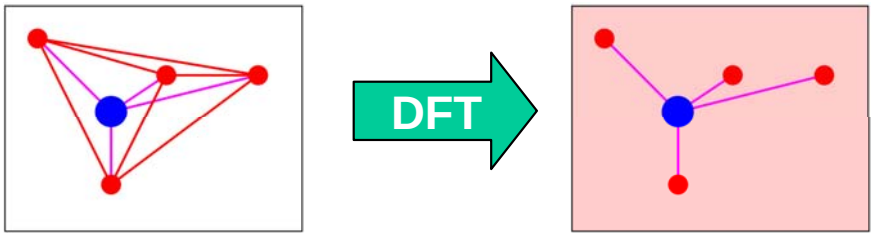
Theoretical approaches I: DFT in LDA

Density functional theory (**DFT**)

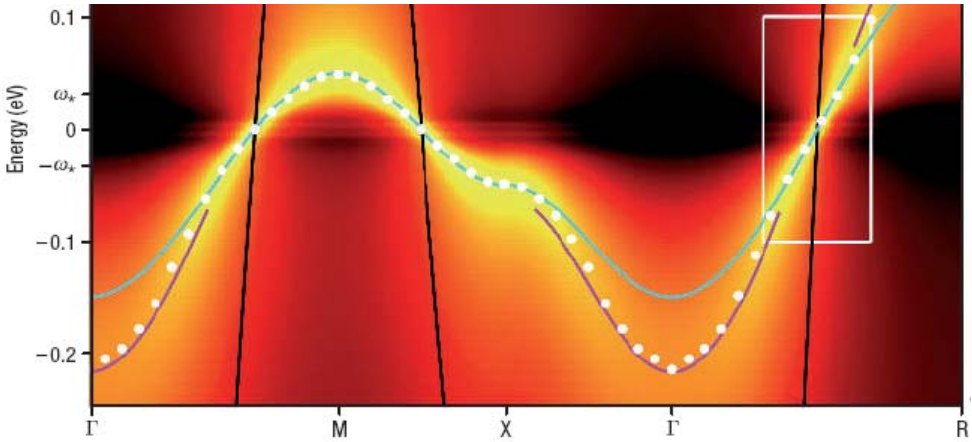
- exact ground state approach
- based on electron density n(**r**)
- Kohn-Sham equations solve effective single-particle problem

- result: ground state energy + n(**r**)
- heuristics: band structure

- problem: exchange-correlation potential unknown



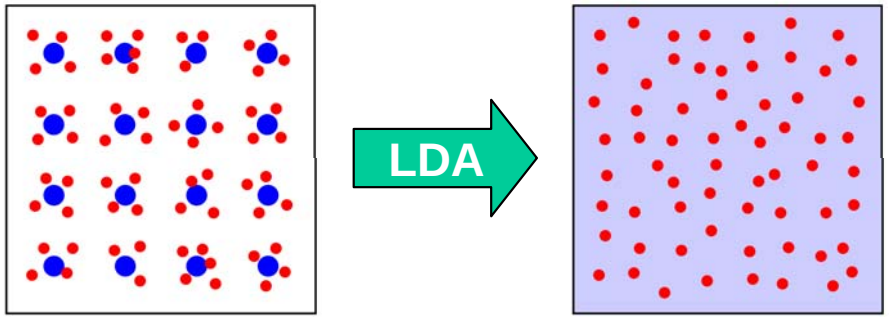
many-body single-particle



[Byzcuk et al., Nature Physics (2006)

Local density approximation (**LDA**)

- exchange-correlation potential taken from jellium model
- not reliable for correlated systems
- often good results, basis for **LDA+U**



e^- in solid jellium model

Theoretical approaches II: DMFT

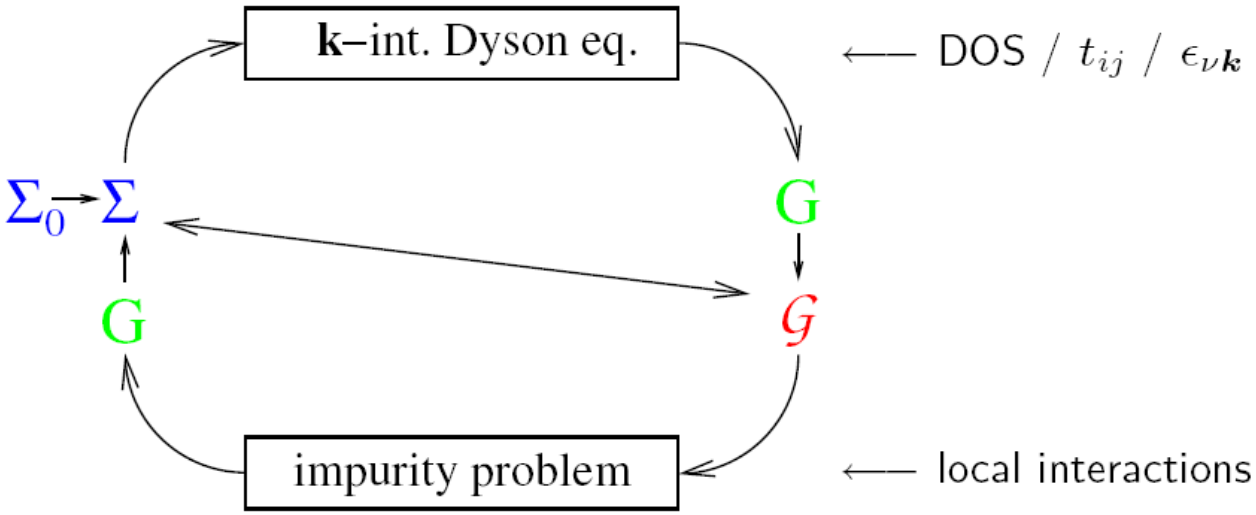
Dynamical mean-field theory (**DMFT**)

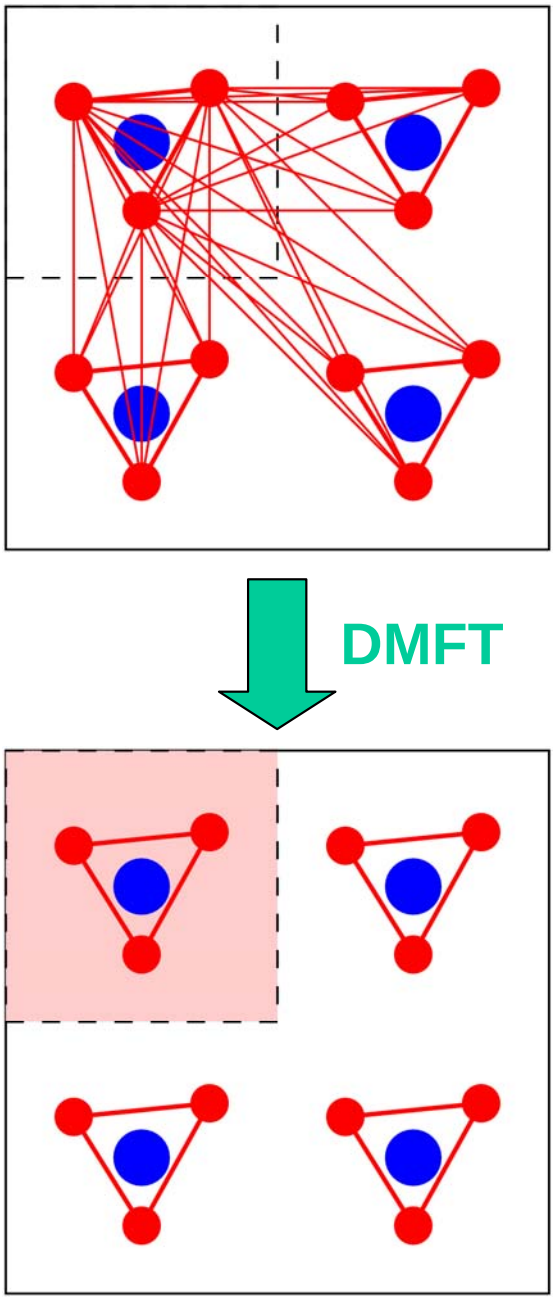
- applicable to Hubbard-type models

$$\hat{H} = \sum_{(i,j),\sigma} t_{ij}(\hat{c}^{\dagger}_{i\sigma}\hat{c}_{j\sigma} + \text{h.c.}) + U \sum_{i} \hat{n}_{i\uparrow}\hat{n}_{i\downarrow}$$

- lattice problem → impurity model, local $\Sigma(\omega)$
- non-perturbative, dynamical on-site correlations

- **LDA+DMFT**: ε_{vk} and interactions from LDA

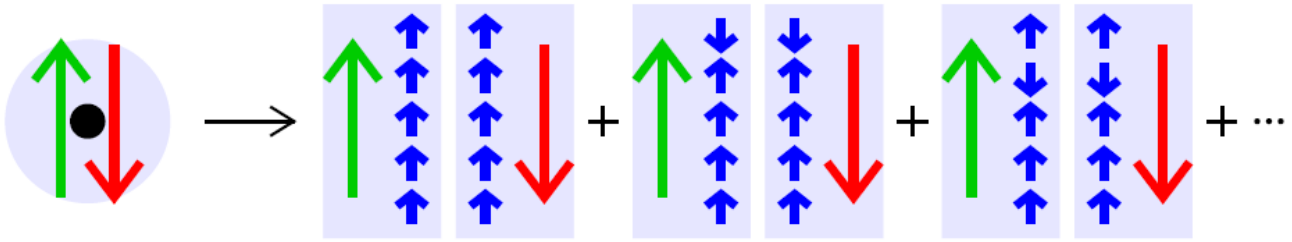




High-precision DMFT-QMC algorithm

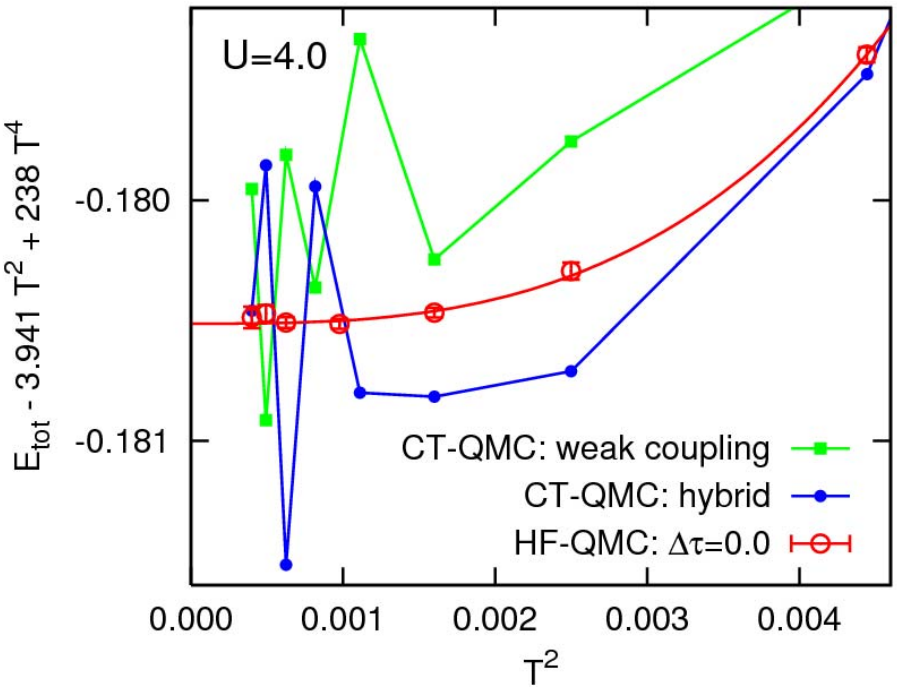
DMFT impurity solver: Hirsch-Fye quantum Monte Carlo (QMC)

- uses Trotter decoupling and Hubbard-Stratonovich transformation:



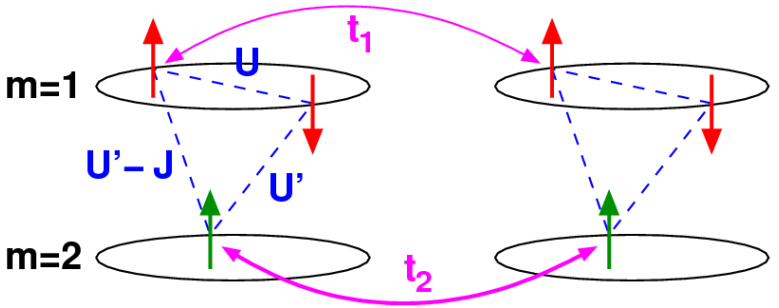
New alternative approaches: continuous-time QMC

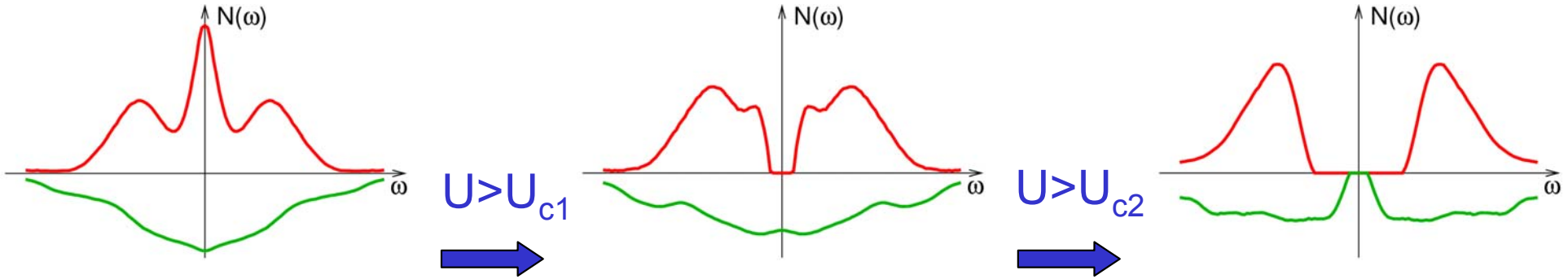
- "*orders of magnitude more efficient than Hirsch-Fye QMC*" [M. Troyer]
- high-precision DMFT-QMC method [Knecht, Blümer] is competitive, at least after extrapolation $\Delta\tau \rightarrow 0$ [cf. Gull et al., cond-mat/0609438]



P6 High-precision DMFT-QMC algorithm

Test case: 2-band model with orbital-dependent hopping and Ising-type Hund couplings

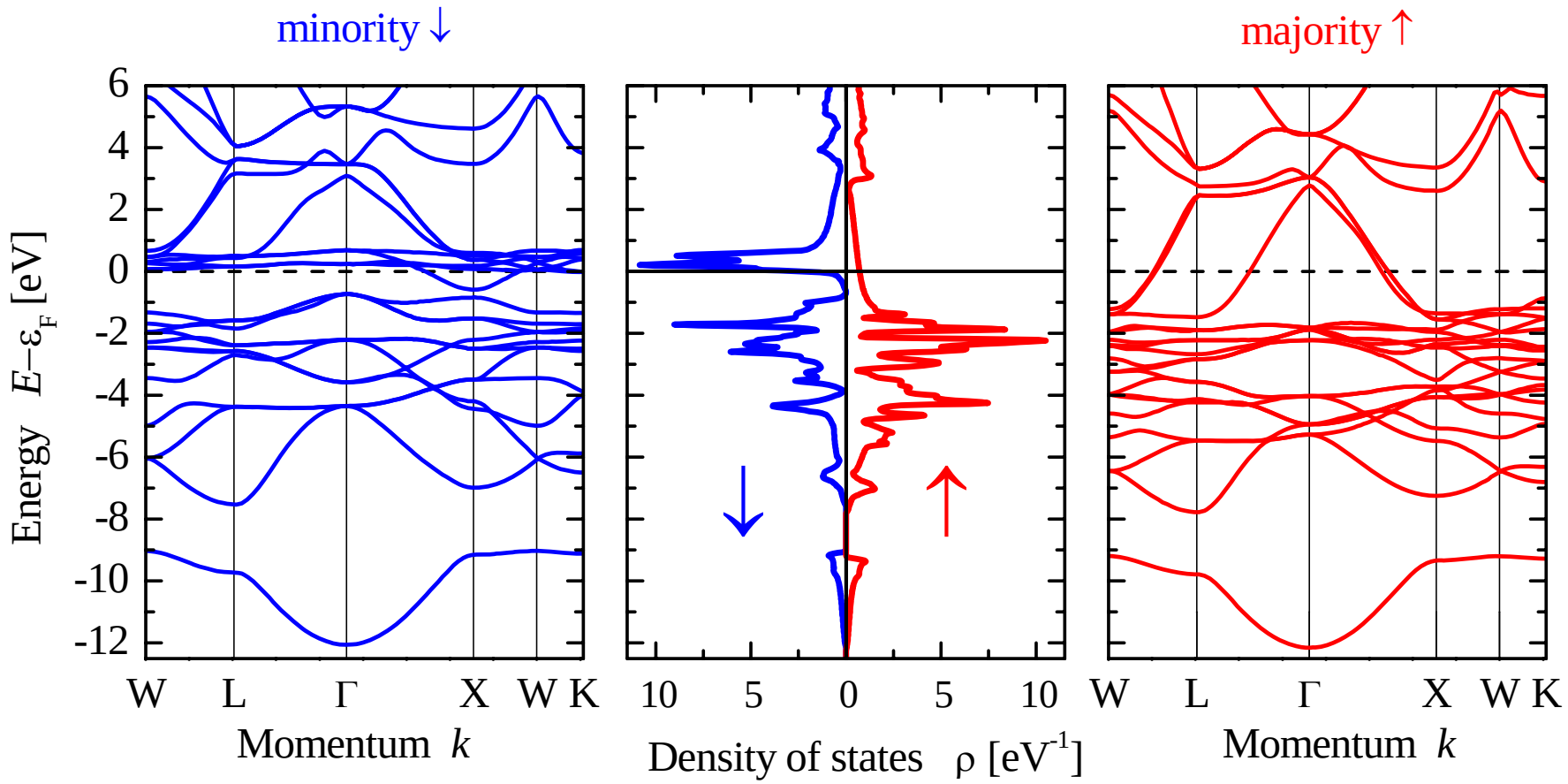




Orbital-selective Mott transitions, intermediate **half-metallic phase**
[Knecht, Blümer, van Dongen, PRB (2005)]

P7 Status/results: Heusler compounds

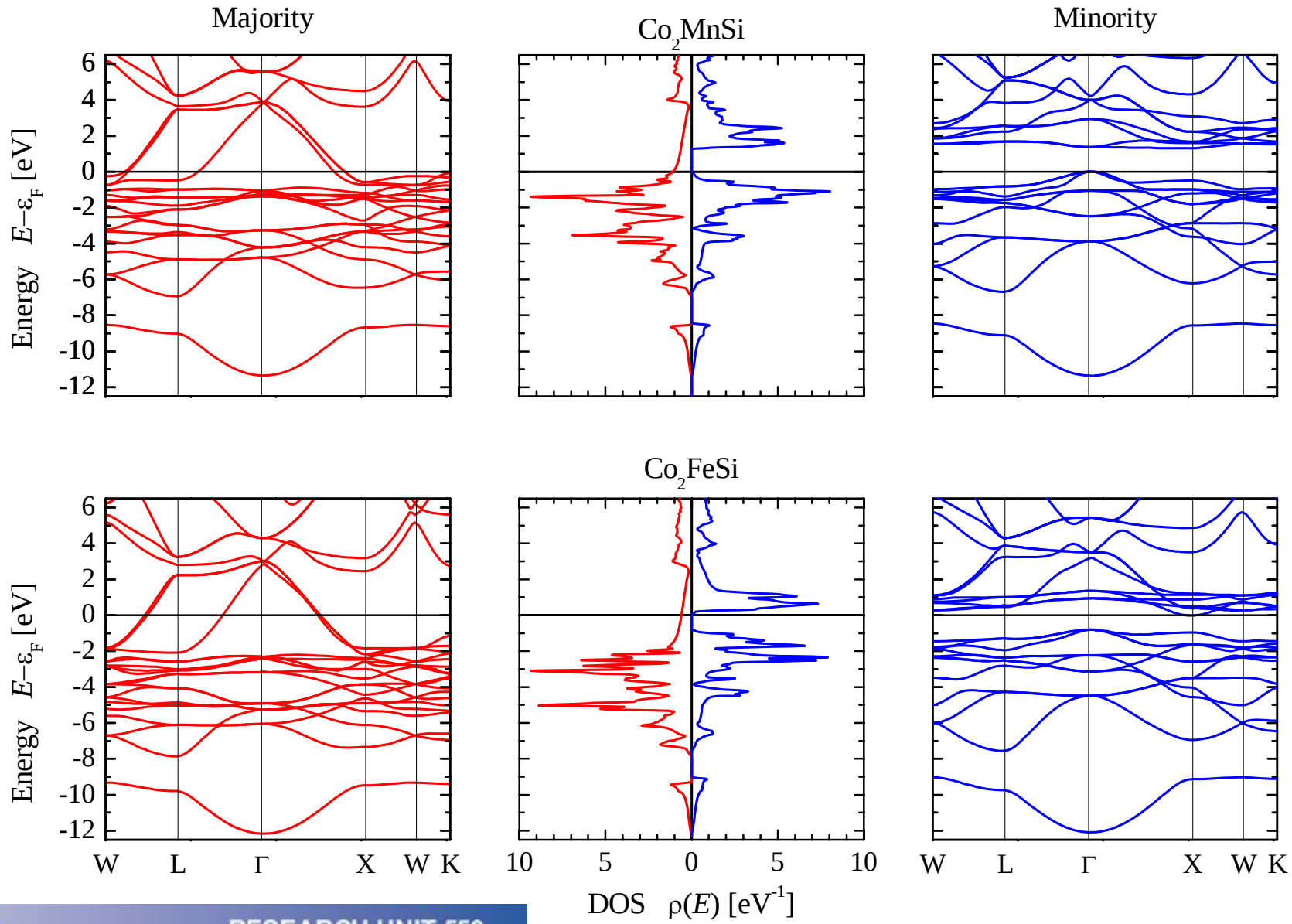
Electronic structure of Co_2FeSi in LSDA



$m_{LSDA} = 5.59\ \mu_B$ < $m_{exp} = 6\ \mu_B$ → LSDA treatment of correlations inadequate

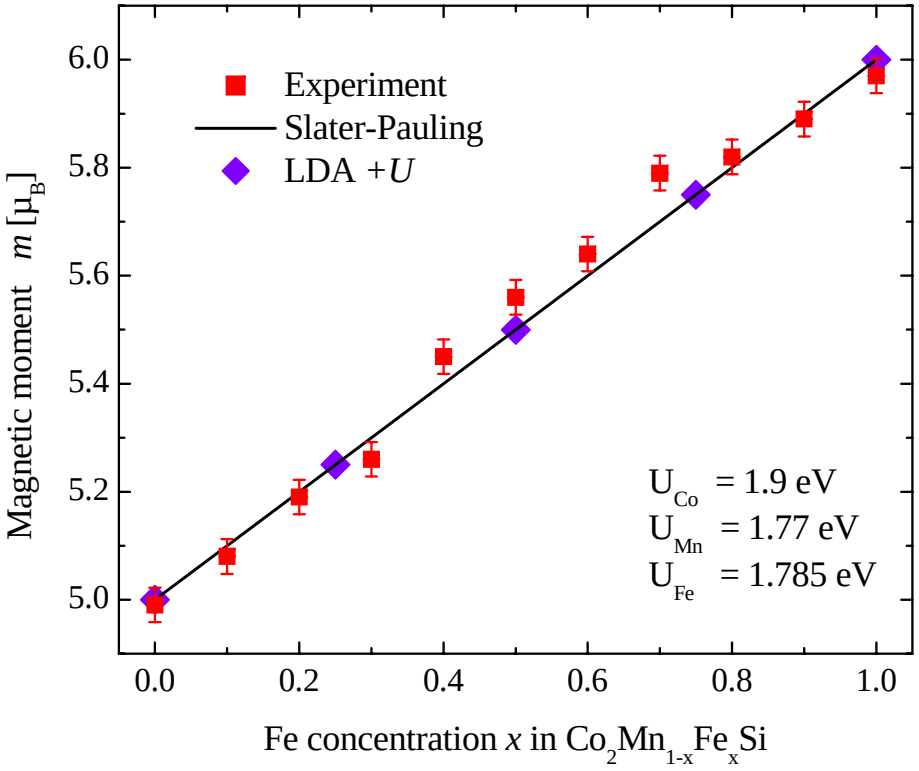
Status/results: Heusler compounds

LDA+*U* for Co_2MnSi and Co_2FeSi

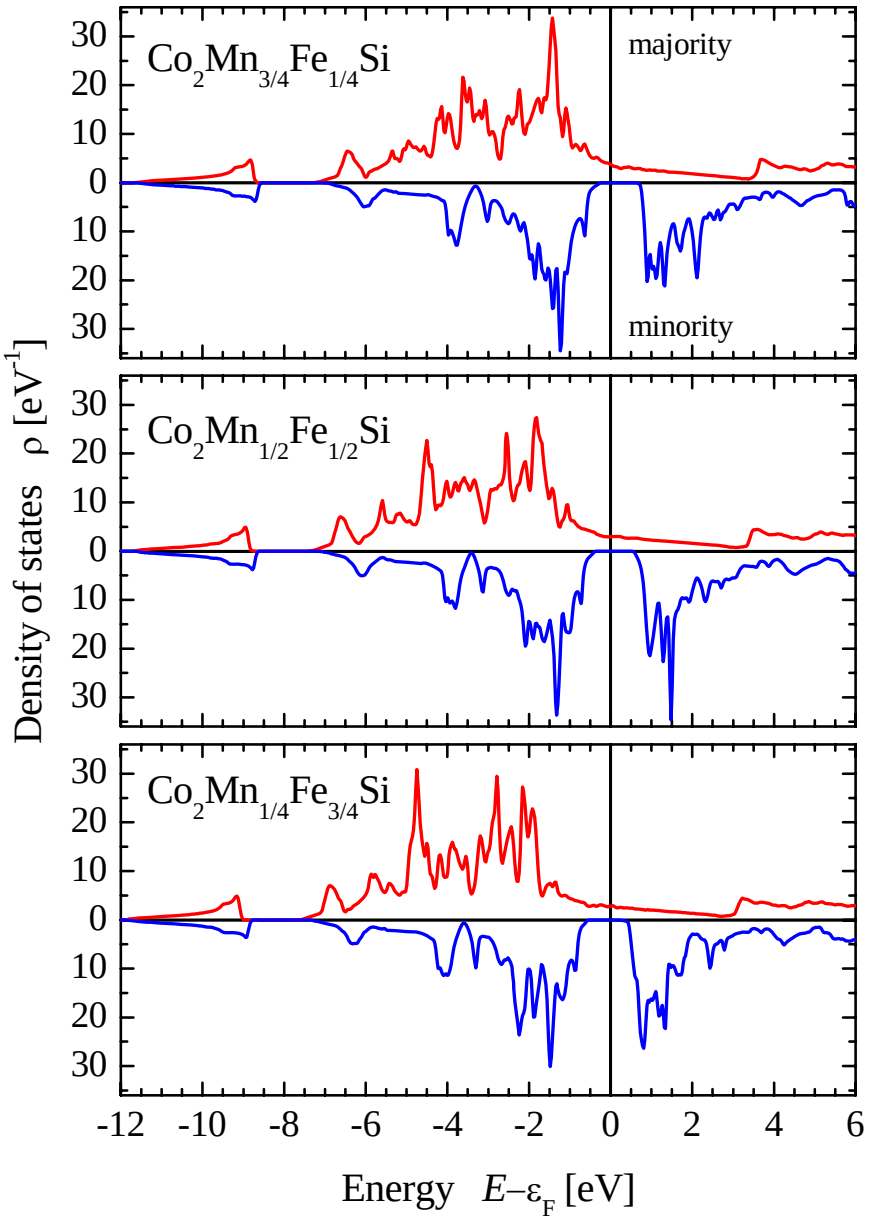


P7 Status/results: Heusler compounds

Magnetic and electronic structure of $Co_2Mn_{1-x}Fe_xSi$

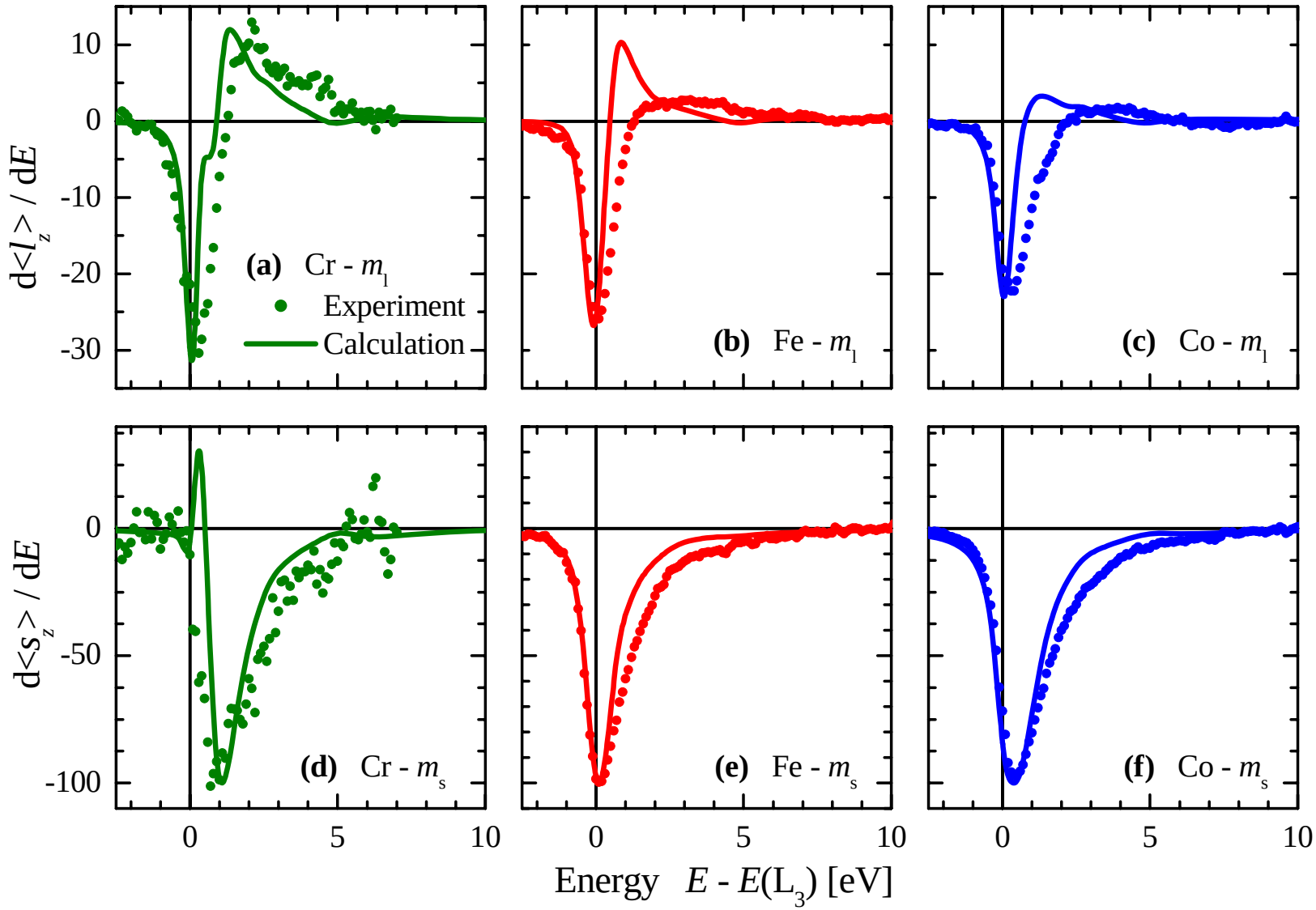


Moment *m* follows Slater-Pauling over the entire range of *x*



P7 Status/results: Heusler compounds

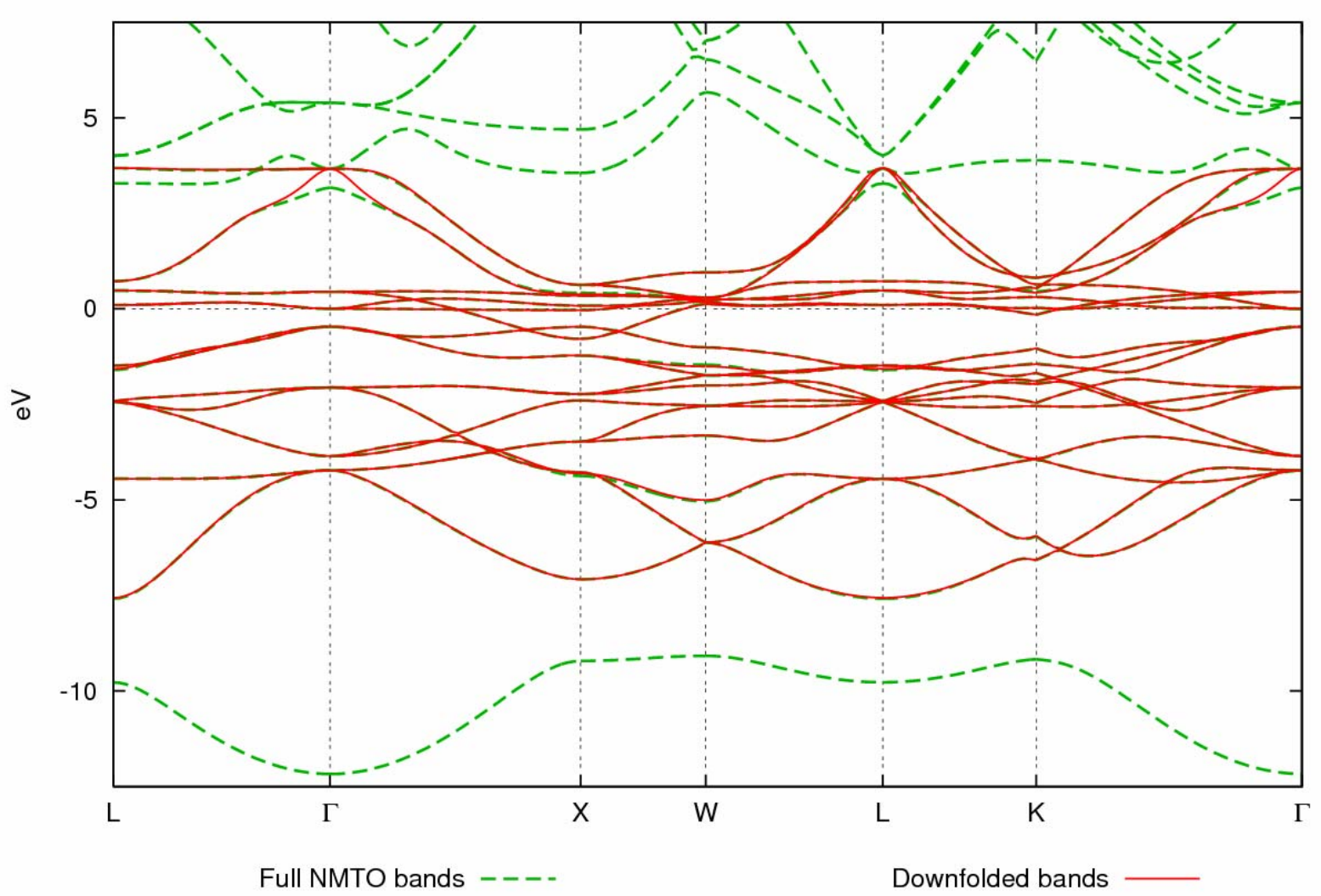
XMCD sum rules for $Co_2Cr_{0.6}Fe_{0.4}Al$ (SPR-KKR with CPA)



[Wurmehl et al., J. Phys. D (2007)]

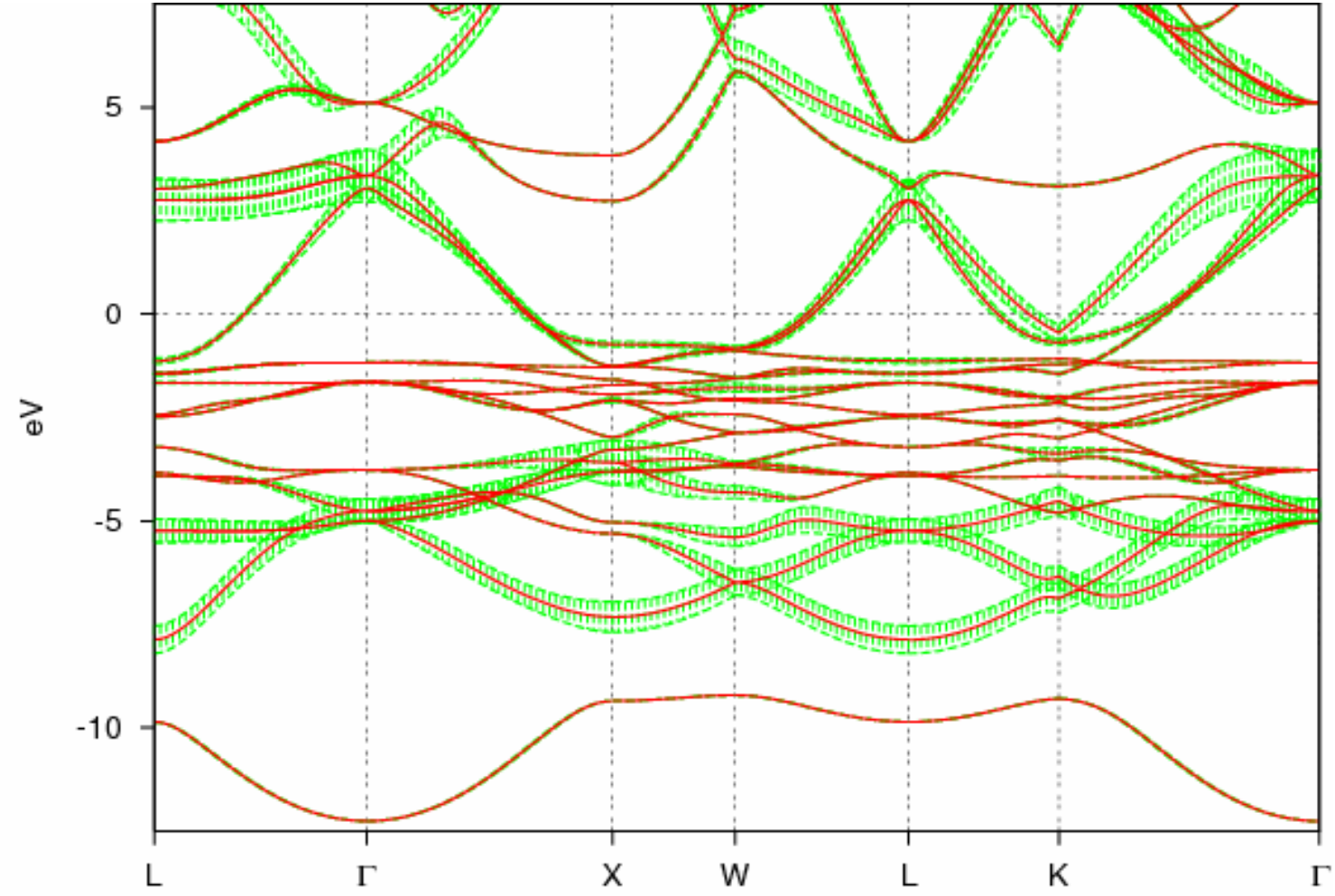
P6 Status/results: Heusler compounds

1st step in LDA+DMFT: low-energy Hamiltonian from LMTO + NMTO with downfolding for Co_2FeSi → 18 bands (2x5 Co d + 5 Fe d + 3 Si p)



P6 Status/results: Heusler compounds

Si p fat bands for Co_2FeSi: significant weight near Fermi energy



help from

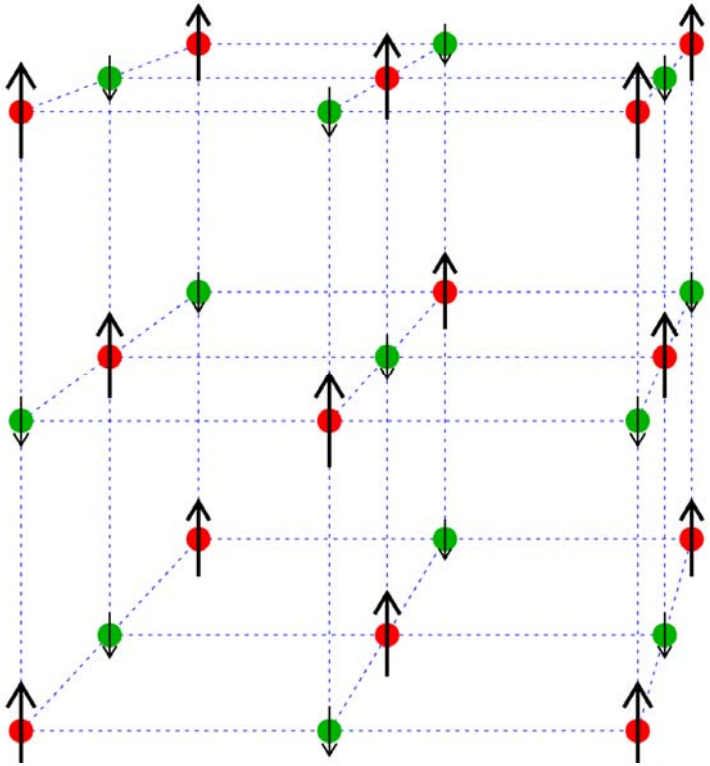
P7

O. Andersen ...

- need 18-band DMFT solver
- or consider (slightly) simpler problems

Status/results: double perovskites

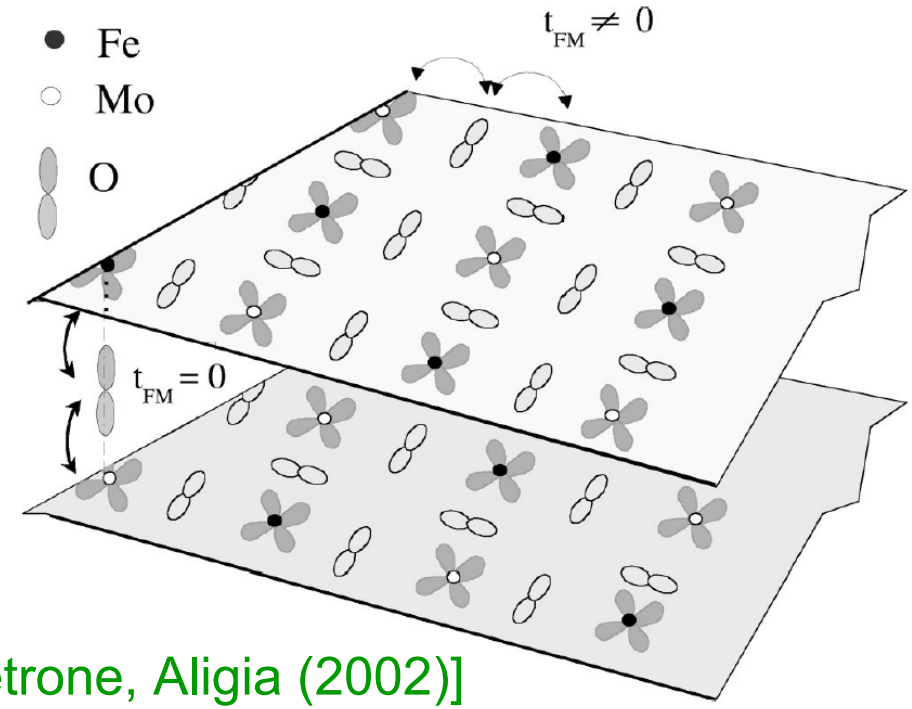
Goal: correlated-electron theory for Sr_2FeMoO_6 and Sr_2FeReO_6



Valences (Sr_2FeMoO_6):

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

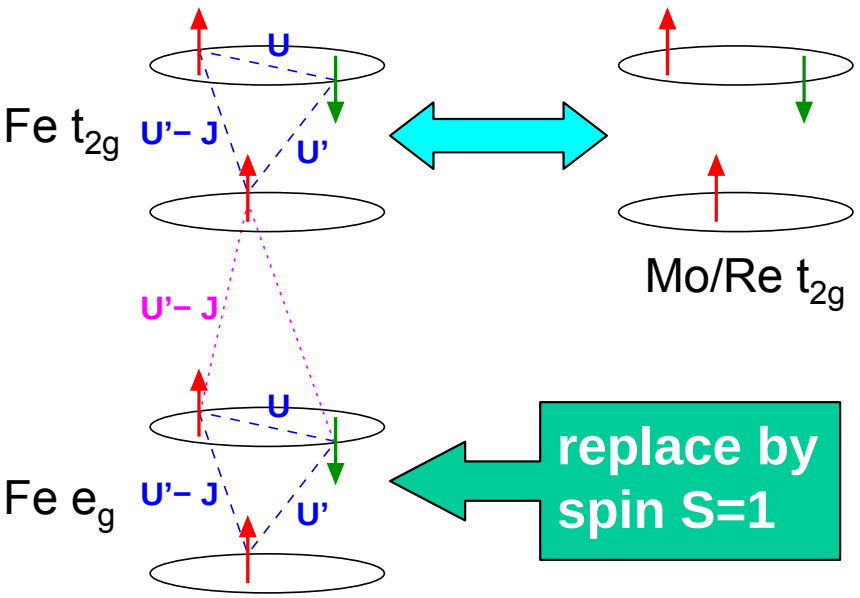
Cubic symmetry → 3 degenerate t_{2g} bands
2 degenerate e_g bands



t_{2g} hybridization
Fe – O – Mo/Re

[Petrone, Aligia (2002)]

P6 Status/results: double perovskites



Full-Hamiltonian LDA+DMFT calculations for 2-site 3-band Hubbard model with

- Fe t_{2g} – t_{2g} interactions
- Fe t_{2g} – e_g interactions
- Fe – Mo/Re hybridization
- controlled DMFT solver: QMC

$$H = \epsilon_F \sum_{i\alpha\sigma} n^f_{i\alpha\sigma} + \epsilon_M \sum_{i\alpha\sigma} n^m_{i\alpha\sigma}$$

$$+ U \sum_{i\alpha} n^f_{i\alpha\uparrow} n^f_{i\alpha\downarrow} + \sum_{i\sigma\sigma'} \sum_{\alpha\neq\alpha'} (U' - J\,\delta_{\sigma\sigma'})\, n^f_{i\alpha\sigma} n^f_{i\alpha'\sigma'} - J \sum_{i\alpha} \boldsymbol{S}_i \cdot \boldsymbol{s}^f_{i\alpha}$$

$$- t_{MF} \sum_{\langle ij\rangle\alpha\sigma} (f^\dagger_{i\alpha\sigma} m_{j\alpha\sigma} + \text{h.c.}) - t_{MM} \sum_{\langle jj'\rangle\alpha\sigma} (m^\dagger_{j\alpha\sigma} m_{j'\alpha\sigma} + \text{h.c.})$$

$t_{MF} \approx 0.25\text{eV}$, $t_{MM} \approx 0.15\text{eV}$, $t_{FF} \approx 0.03\text{eV}$, [Phillips et al. (2003)]

P6 Status/results: double perovskites

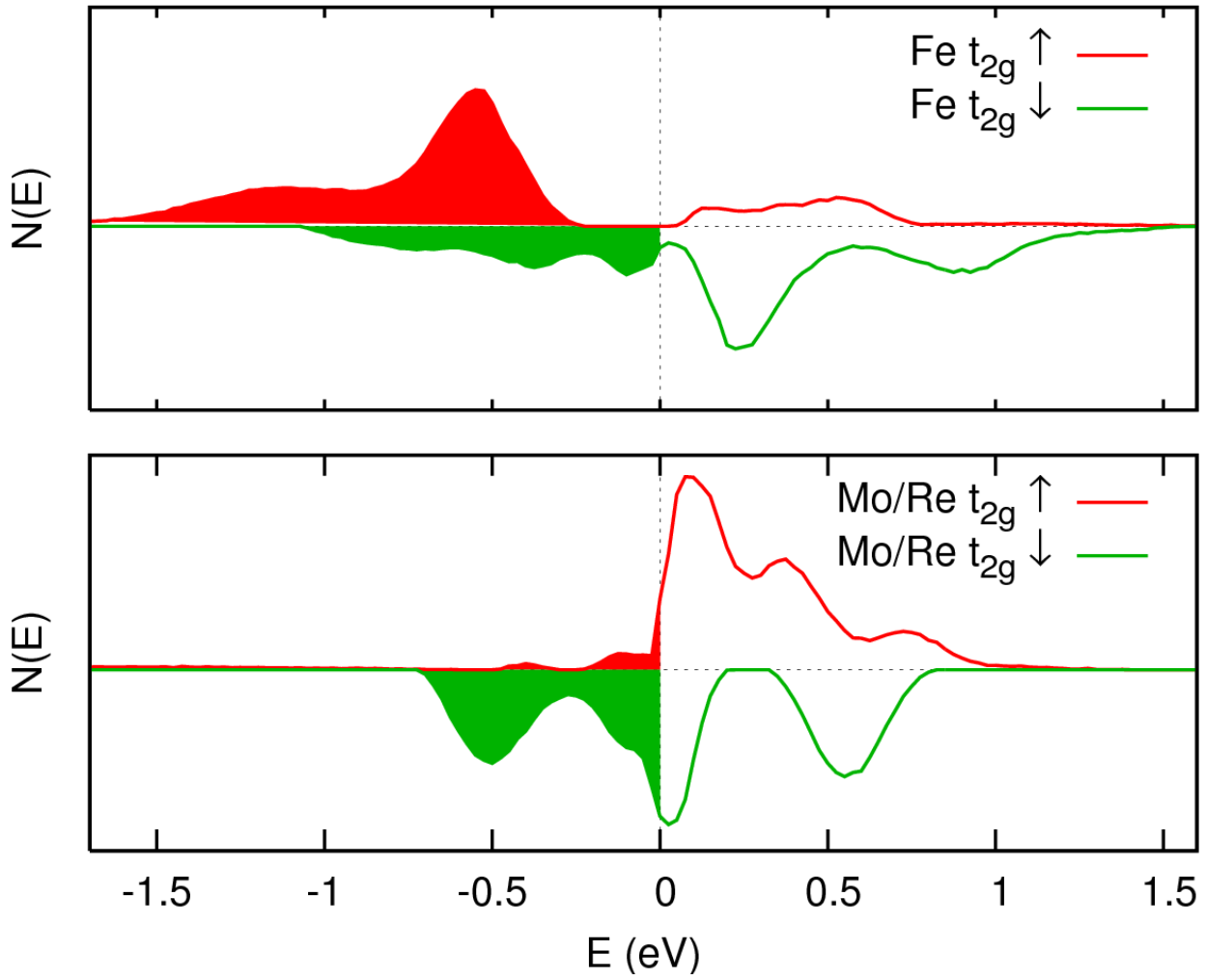
Preliminary DMFT-QMC results (moderate interactions ~ 1eV)

t_{2g} fillings
- n_{Fe} = 3.47
- $n_{Mo/Re}$ = 1.75

t_{2g} moments
- m_{Fe} = 1.5 μ_B
- $m_{Mo/Re}$ = -1.0 μ_B

n = 5.2, m = 2.5 μ_B

todo:
- ➢ adjust fillings
- ➢ stronger interactions
- ➢ check QMC, MEM ...
- ➢ make contact with experiments



Summary and outlook

P6

- Methodological development: multi-band DMFT-QMC
- Test case: 2-band model
- Co_2FeSi low-energy Hamiltonian from NMTO with downfolding
- Initial LDA+DMFT(QMC) results for double perovskites

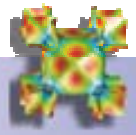
Outlook

- LDA+DMFT for (half-, full-) Heusler alloys
- LDA+DMFT(QMC) for double perovskites
- Photoemission spectra, element-specific moments, Curie temperatures, ...

P5, P7, P10
P11, P1, P4

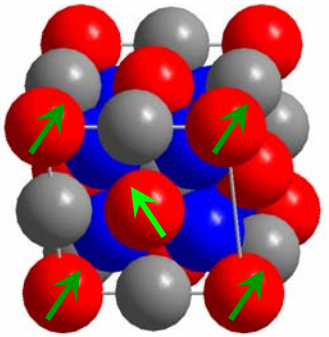
P7

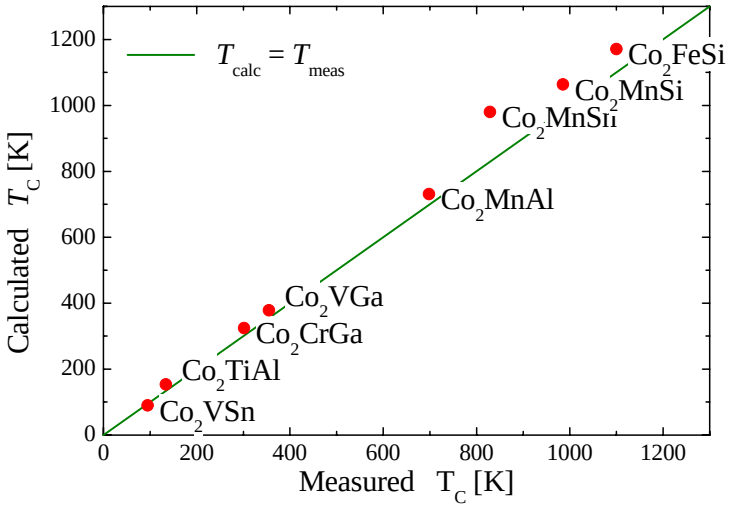
- DFT calculations (within LDA, GGA), LDA+U ...
- Photoemission spectra (including matrix element effects)
- Relativistic calculations, disorder (within CPA)



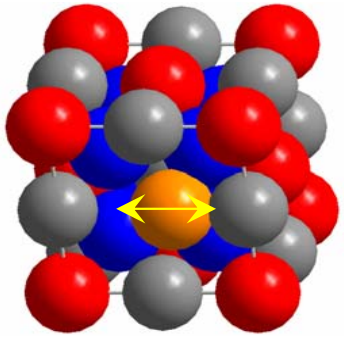
P7 Outlook: extensions of DFT and LDA

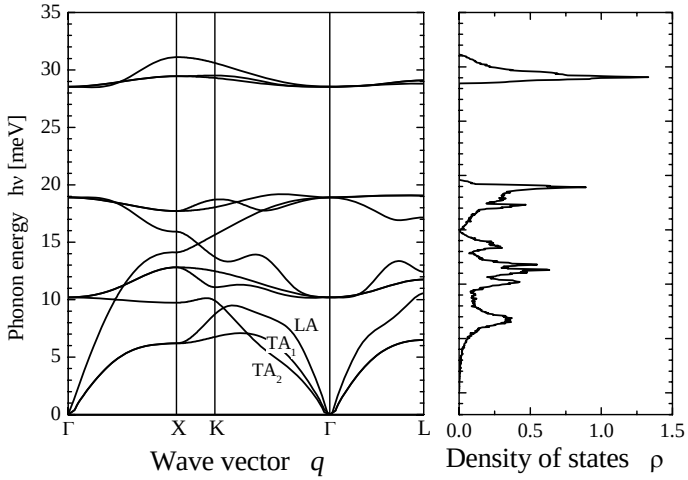
- Non collinear spin
  → T_C, magnons
  (with J. Kübler)





- Supercells
  → phonons





- Exact exchange
  → minority gap

$$E_{xc}(\text{EXX}) = E_{xc}(\text{LDA}) + \{E_x(\text{HF}) - E_x(\text{LDA})\}$$