Exchange coupling in selected Eu compounds

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Collaborations

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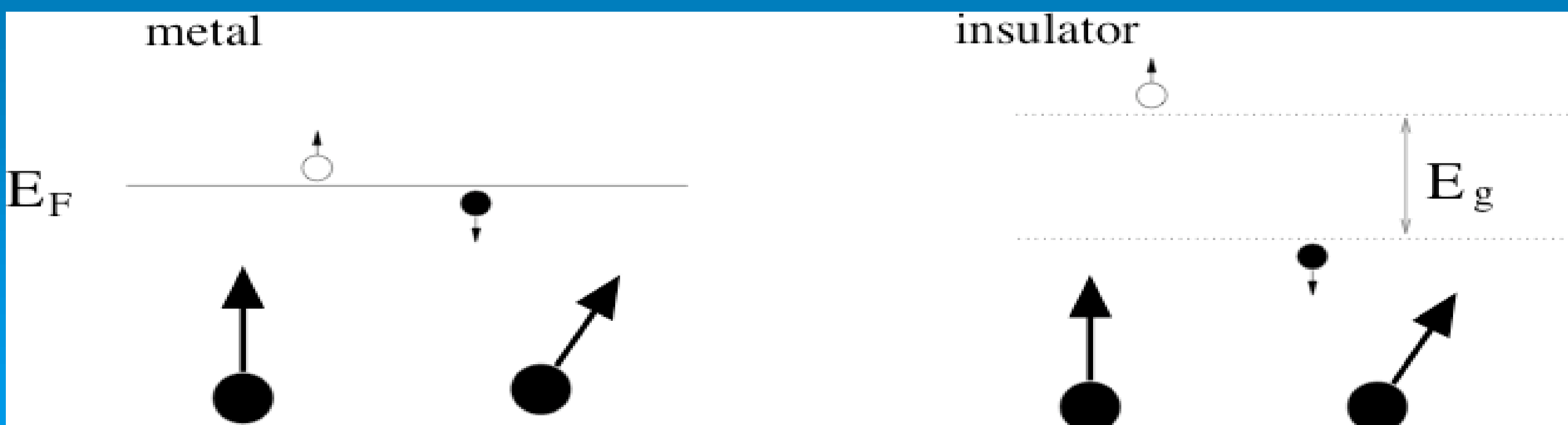
Outline

- Rare earth materials
- Basics of LDA+U
- EuX - ferromagnetic insulators
- Bcc Eu - spin spiral groundstate
- EuB_6 - semimetal?
- Summary

Physics of 4f electrons

- Localized 4f shell - interactions: Coulomb repulsion, spin-orbit coupling, hopping (hybridization)
- Phenomena: localization-delocalization (α - γ Cerium), valence fluctuation, various types of magnetic ordering
- Anderson lattice model



LDA+U method

Motivation:

Treat on-site interactions explicitly because we do not know how to guess appropriate density functional.

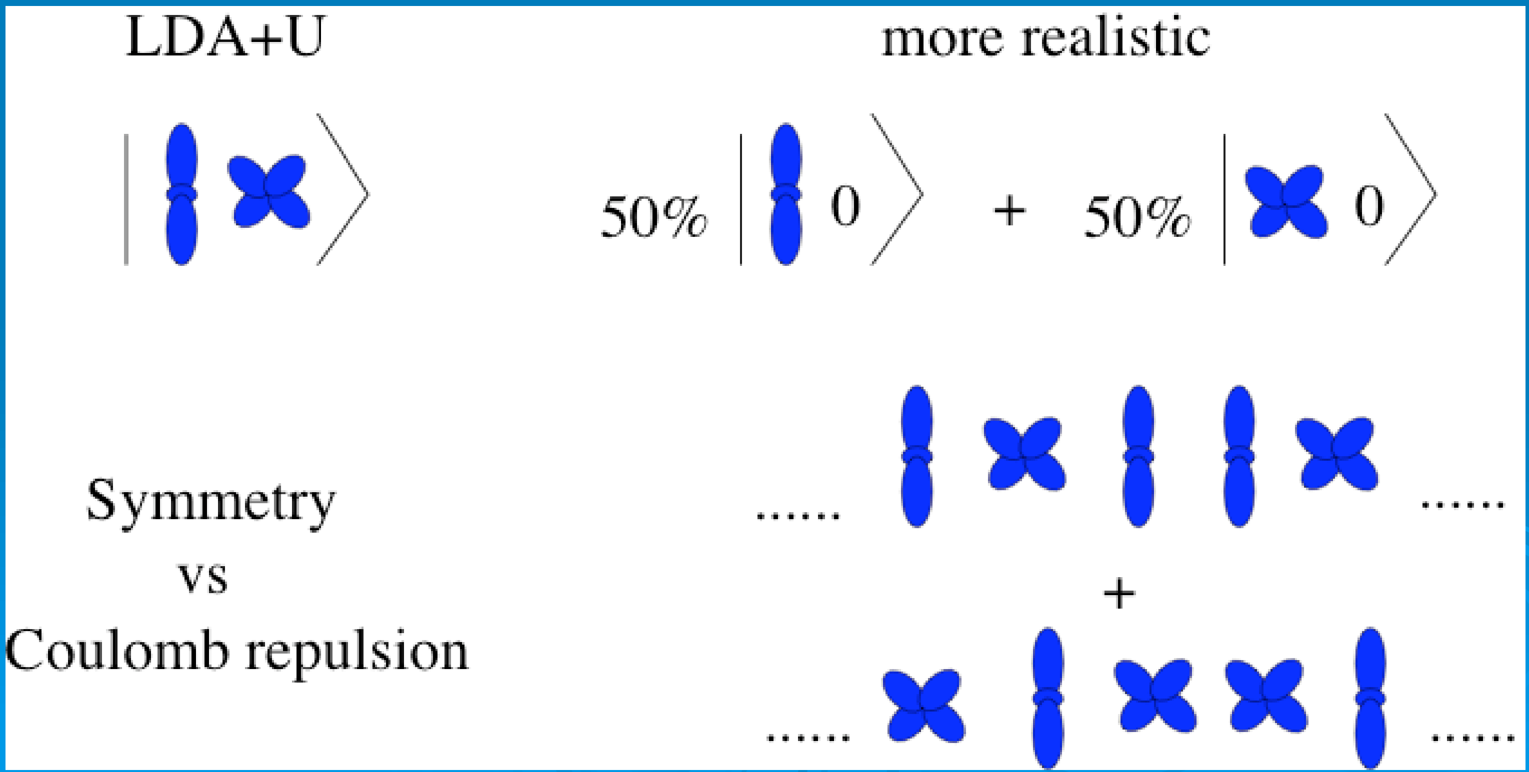
Energy functional: $E[\rho] = E_{LDA}[\rho] + E_U[n_i(\rho)] - E_{DC}[n_i(\rho)]$

Effectvie Hamiltonian: $H = H_0 + H_U(< n_i >)$

H_u has a form of effective crystal field

- LDA+U only reasonable for Mott insulator
- Cannot describe local statistical averages - overestimates ordering

Example: 2 equivalent orbitals & 1 electron



Eu chalcogenides

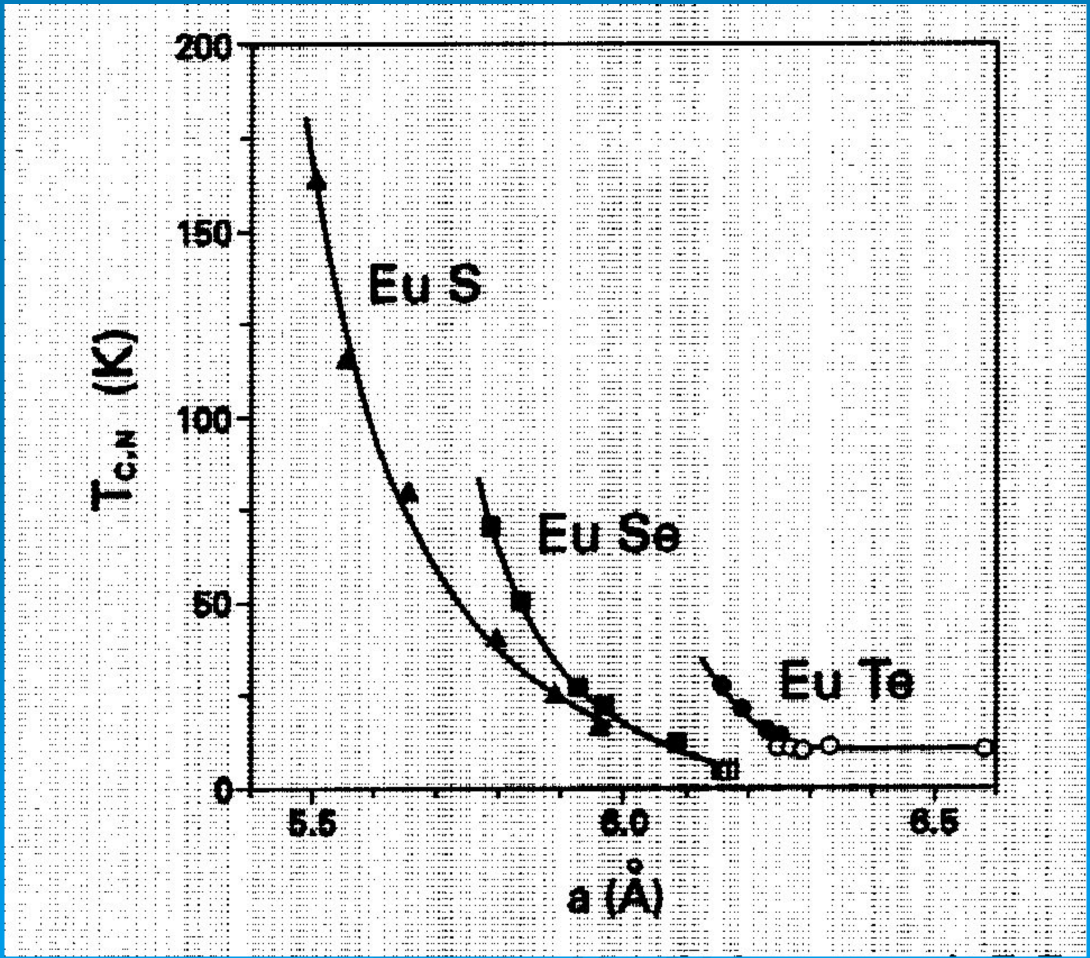
- Eu^{2+} valency = f^7 configuration
- NaCl ----> CsCl (at 12 - 20 GPa)
- EuO and EuS - FM at ambient pressure (Tc=68 K and 16 K)
- EuSe and EuTe - type II AFM at ambient pressure, FM at elevated pressure (~10 GPa)

Quantitative calculation of coupling parameters as a function of pressure.

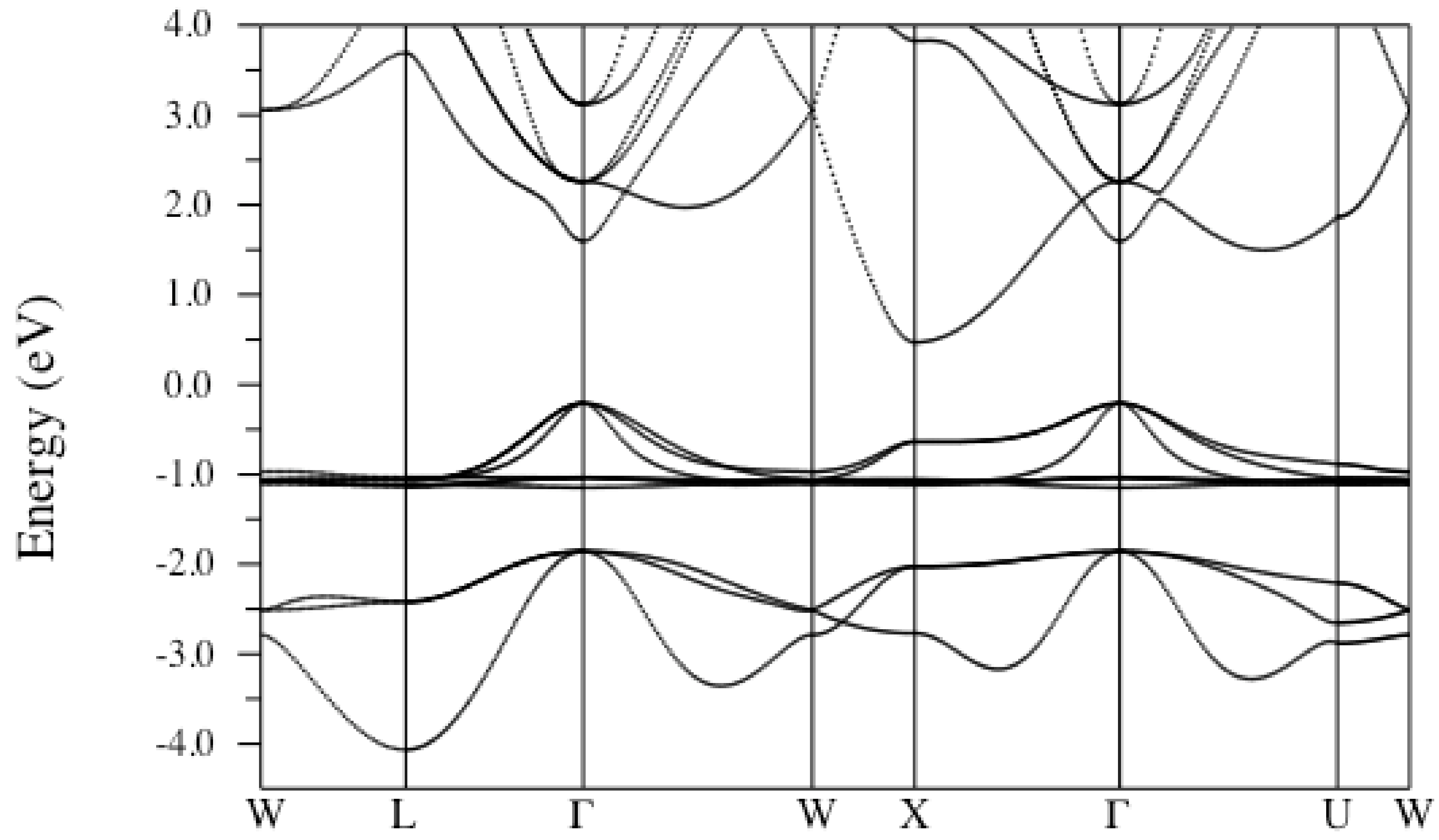
Important coupling processes?

Effect of Pressure

Goncharenko and Mirebeau, PRL **80** 1082 (1998):

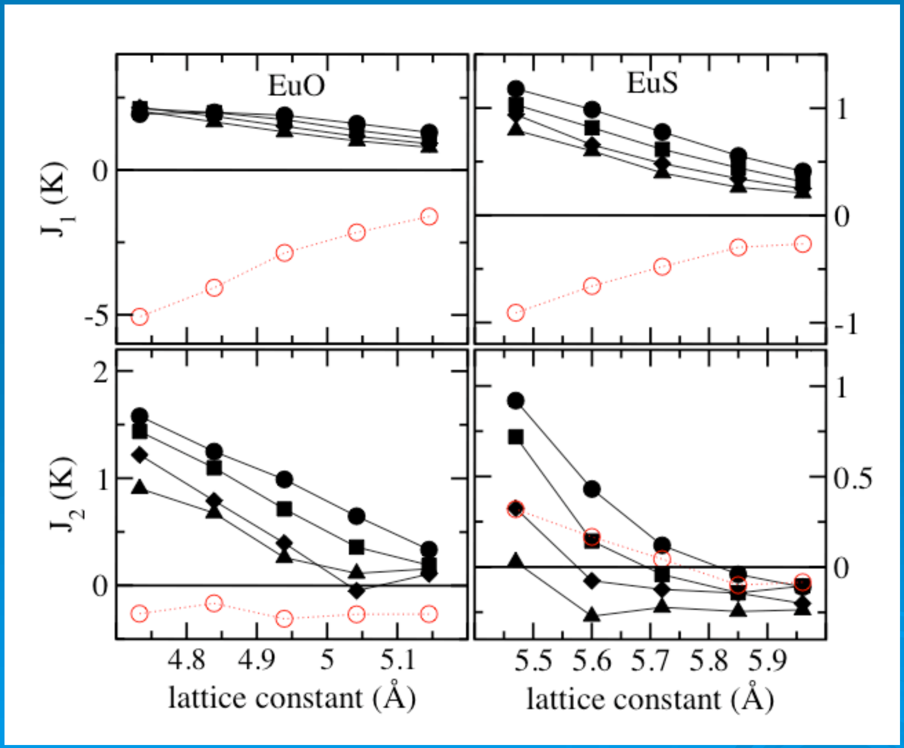


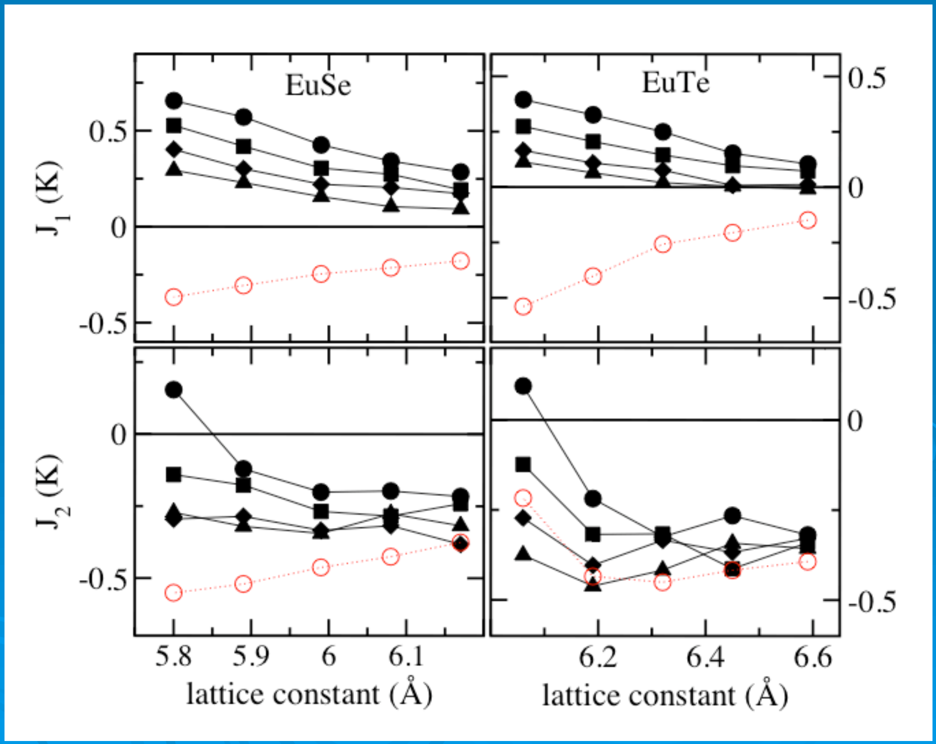
Band structure



Effective exchange J_1-J_2

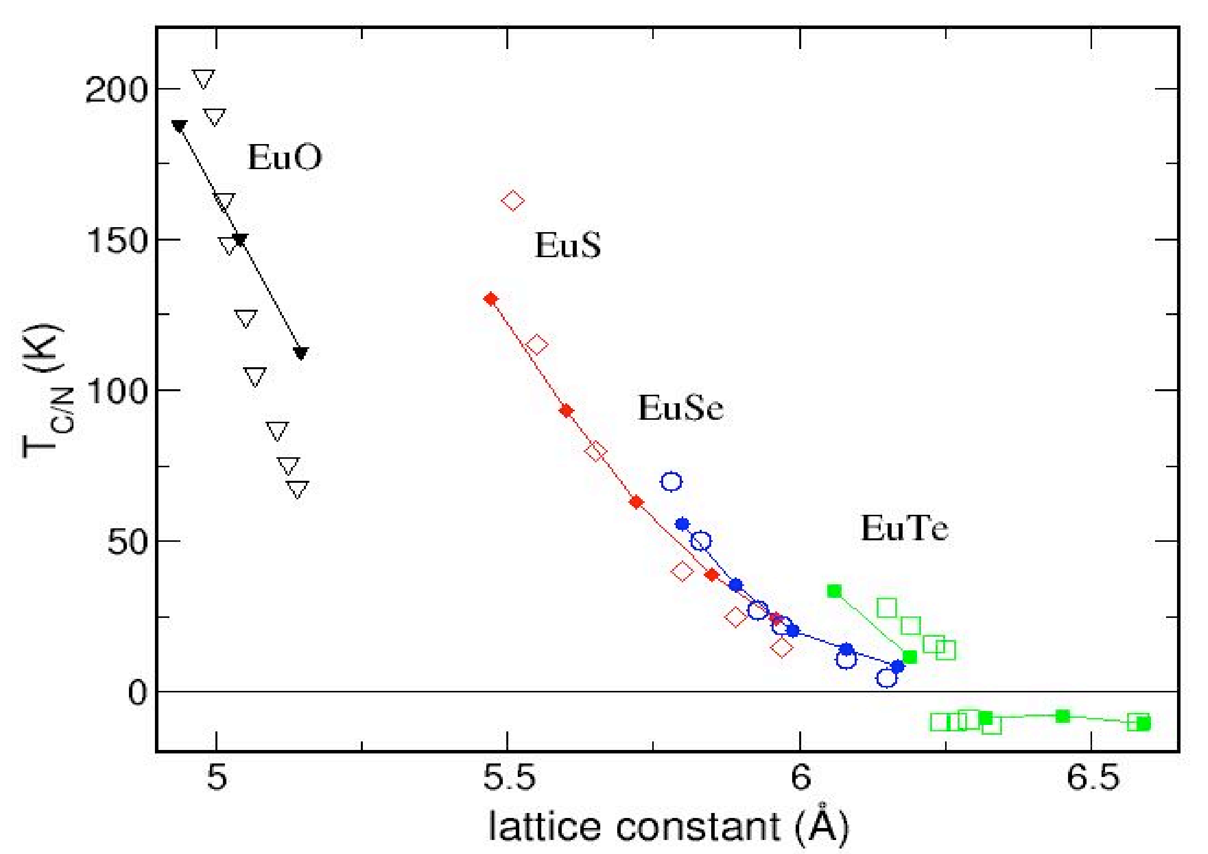
$$H = \frac{1}{2}\sum_{i,j} J_{ij}\mathbf{S}_i \cdot \mathbf{S}_j$$





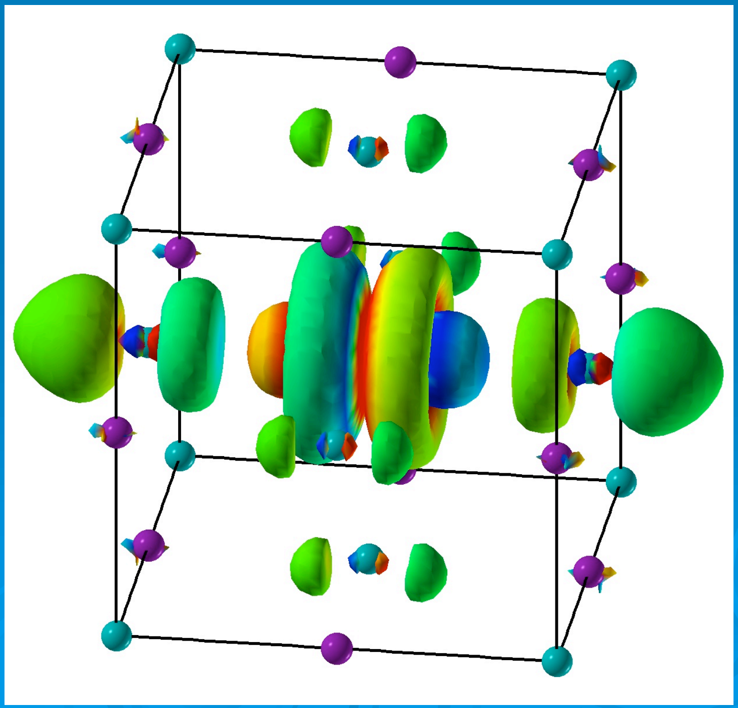
Ordering temperature

$$(k_B T_C)^{-1} = \frac{3}{2S(S+1)} \frac{1}{N} \sum_{\mathbf{q}} [J(\mathbf{0}) - J(\mathbf{q})]^{-1}$$



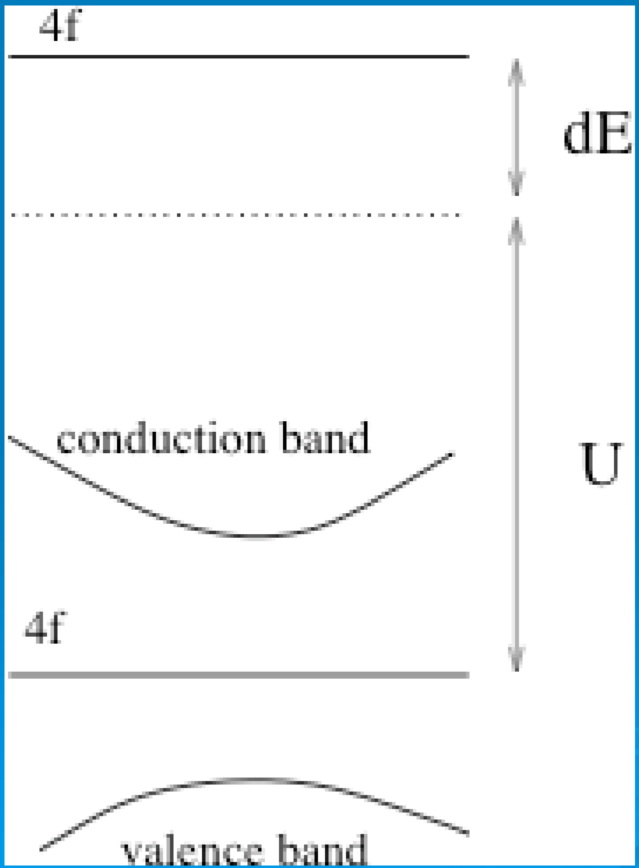
LDA+U vs open core

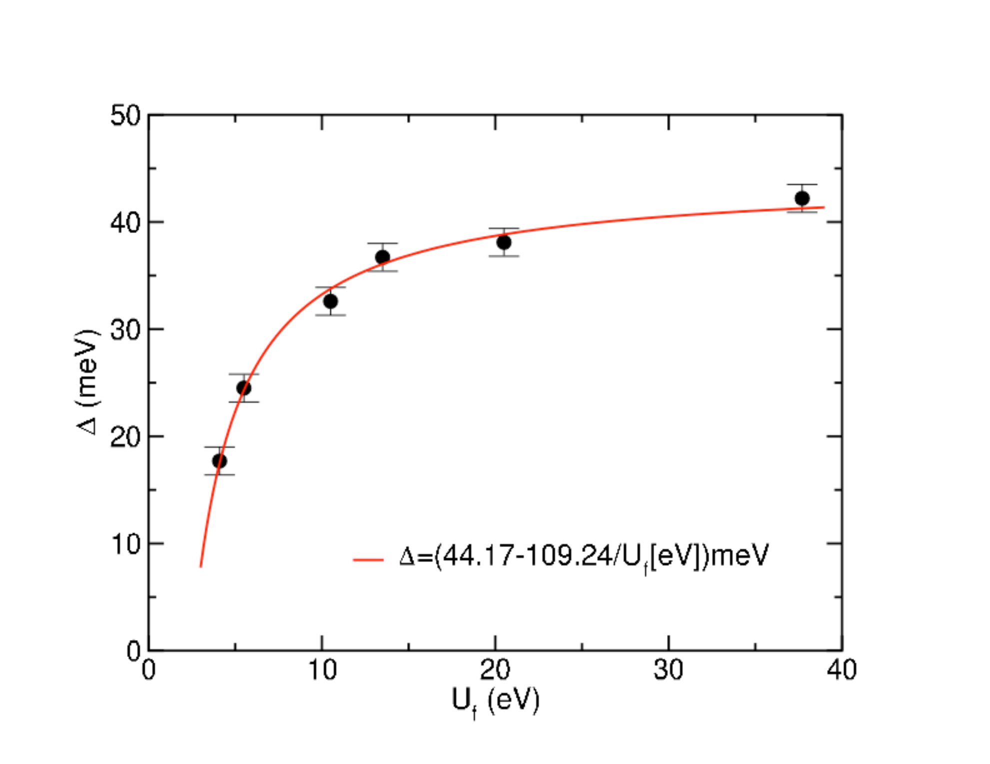
LDA+U	open core
mixing + potential	potential only
all exchange processes	$JS_f.s_d$



f-f super exchange

$J_{se} \sim 1/U_{eff}$



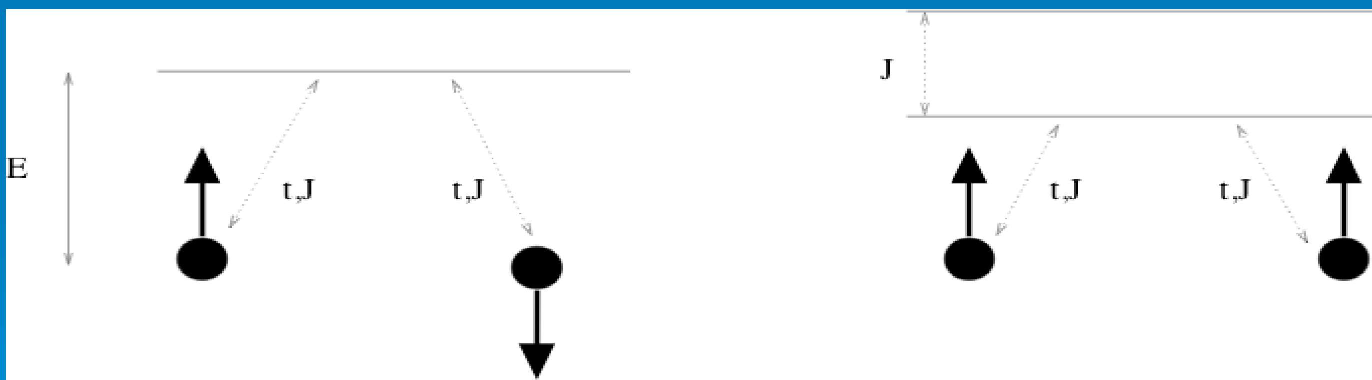


f-d exchange model

Full diagonalization vs mean field

3-site model: U_f- infinite, 2 electrons

$$H = E\sum_{\sigma} d^{\dagger}_{\sigma} d_{\sigma} + t\sum_{\sigma,i}(f^{\dagger}_{i\sigma} d_{\sigma} + h.c.) - J\sum_{i} \mathbf{S}_{fi}\cdot\mathbf{S}_{d}$$



$$E_{\uparrow\uparrow} - E_{\uparrow\downarrow} \approx -8\left(\frac{t}{E}\right)^{2} J$$

EuX - summary

- LDA+U provides reasonable quantitative description of coupling parameters and T_C
- $J_1(p)$ and $J_2(p)$ follow different pressure dependencies
- Hopping from/to f states is crucial for ferromagnetism
- Unoccupied f bands play active role (f-f super exchange)

Bcc Eu

- Local moment metal (f^7)
- Magnetically ordered below T_N=90.5 K
- Spin spiral ground state: $q_0||c$; pitch of 48° per layer

Magnetic interactions (RKKY)?

Quantitative evaluation of T_N and q_0 ?

Origin of the spin-spiral ground state?

Computational approaches:

real space vs reciprocal space (spin spirals)

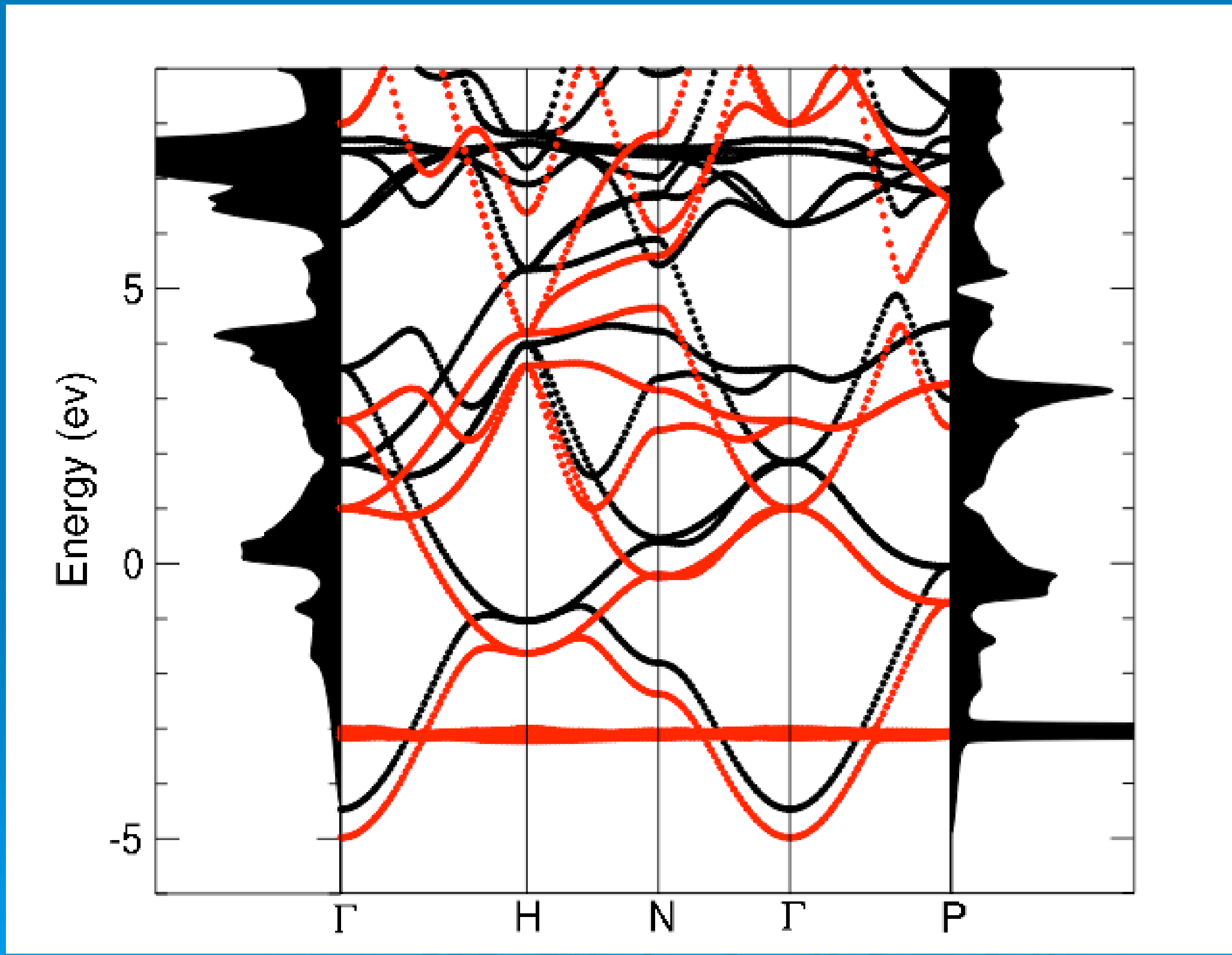
Spin spirals

$$\mathbf{M}(\mathbf{r}) = (\cos(\mathbf{R}\cdot\mathbf{q})\sin\Theta, \sin(\mathbf{R}\cdot\mathbf{q})\sin\Theta, \cos\Theta)$$

- Spin-orbit coupling neglected
- Symmetry operations: lattice translations coupled to spin rotations
- Generalized Bloch theorem:

$$\psi_{\mathbf{k},\mathbf{q}}(\mathbf{r}) = \begin{pmatrix} e^{i(\mathbf{k}+\mathbf{q}/2)\cdot\mathbf{r}} f(\mathbf{r}) \\ e^{i(\mathbf{k}-\mathbf{q}/2)\cdot\mathbf{r}} g(\mathbf{r}) \end{pmatrix}$$

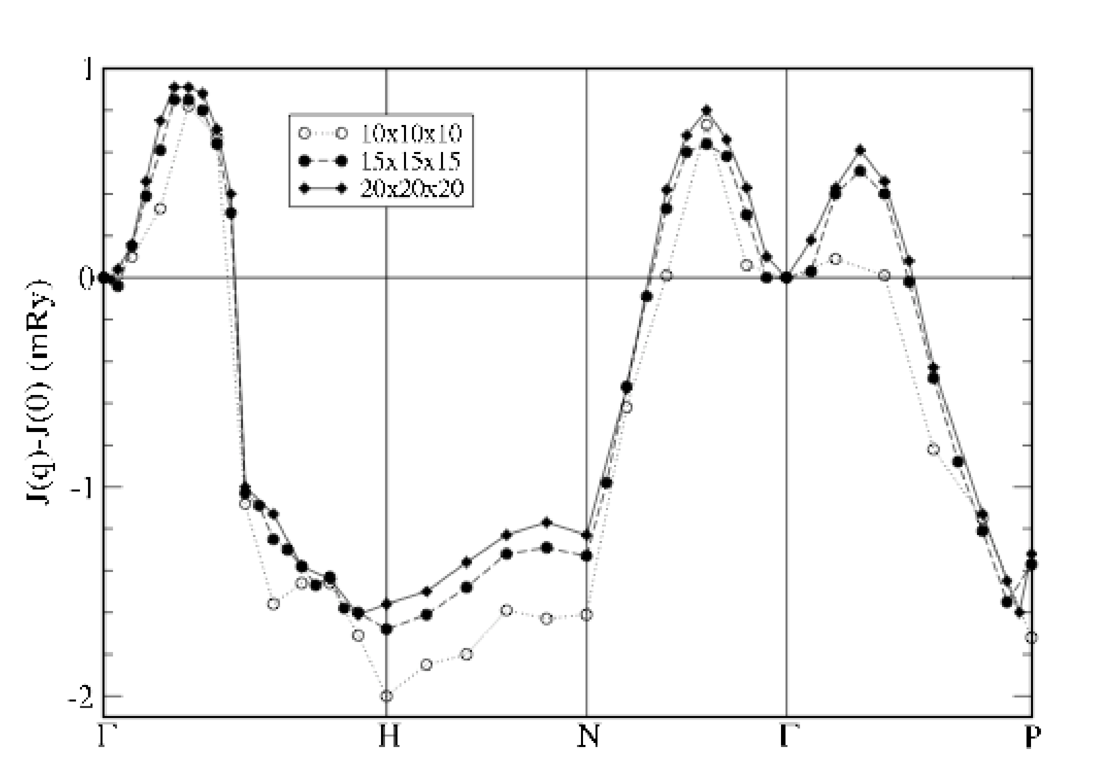
Band structure



Spin spirals - J(q)

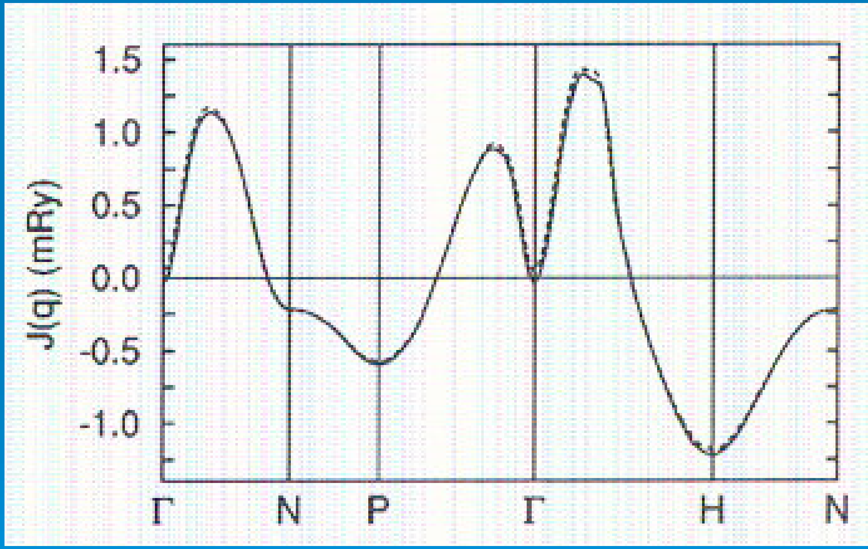
$$H = \frac{1}{2}\sum_{i,j} J_{ij}\mathbf{S}_i \cdot \mathbf{S}_j$$

$$J(\mathbf{q}) = \sum_i e^{i\mathbf{q}\cdot\mathbf{R}_i} J_{0i}$$

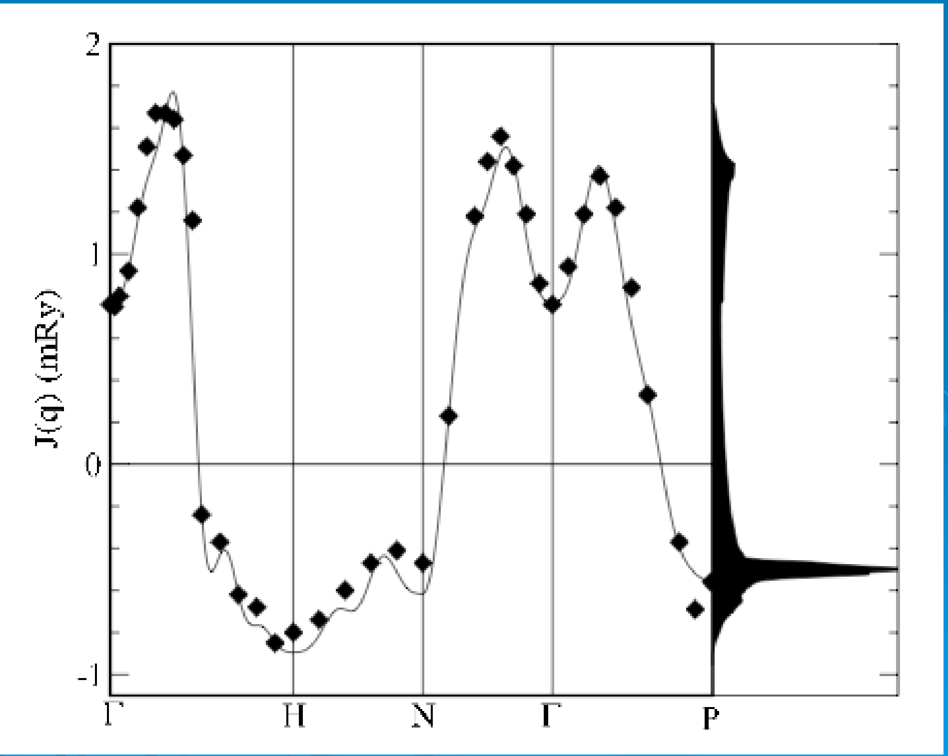


Normalized J(q)

Goldstone sum rule: $$\int_{BZ} d\mathbf{q} J(\mathbf{q}) = 0$$

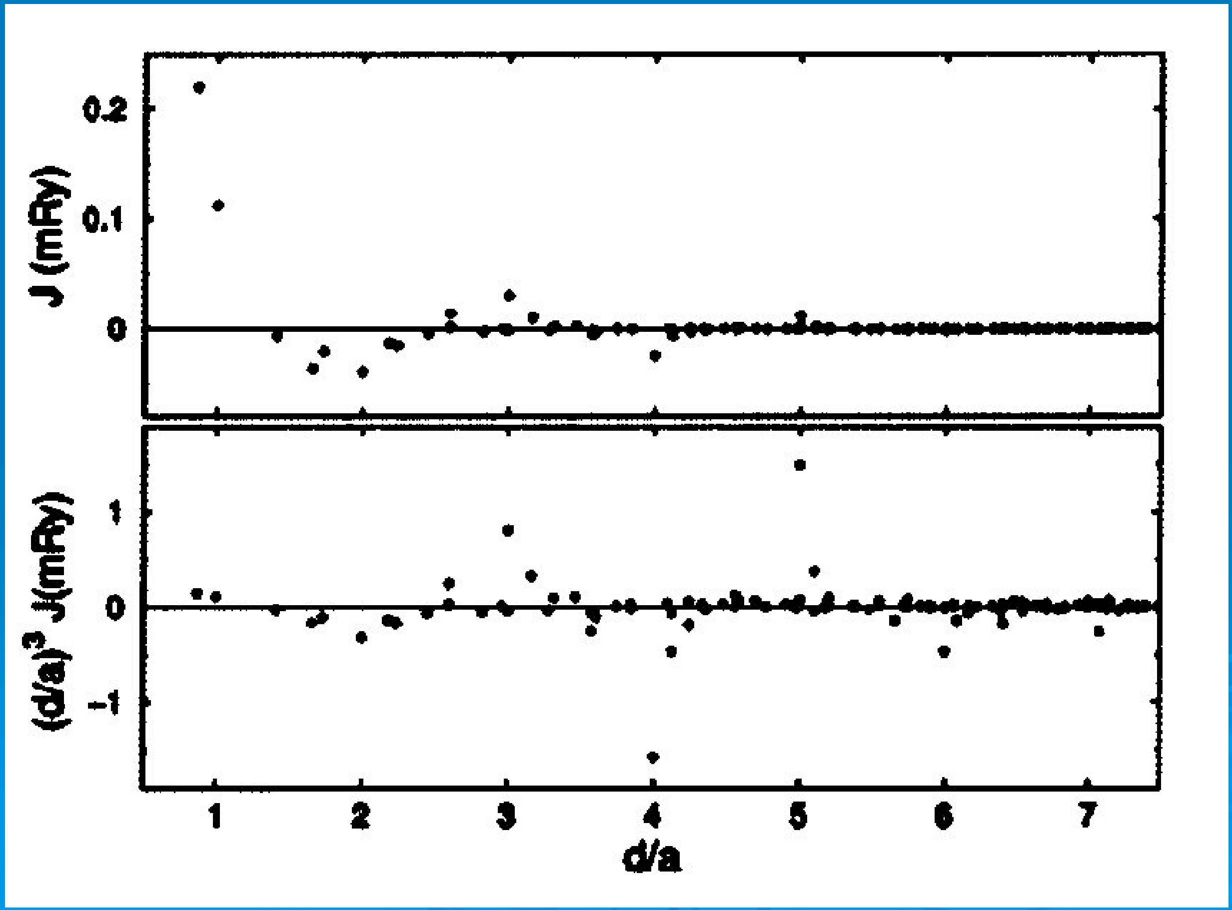


Turek *et al*. PRB **68** 224431 (2003)
real space LMTO calculation



Real space exchange parameter J_{0R}

Long range J_{ij} ; RKKY oscillations:



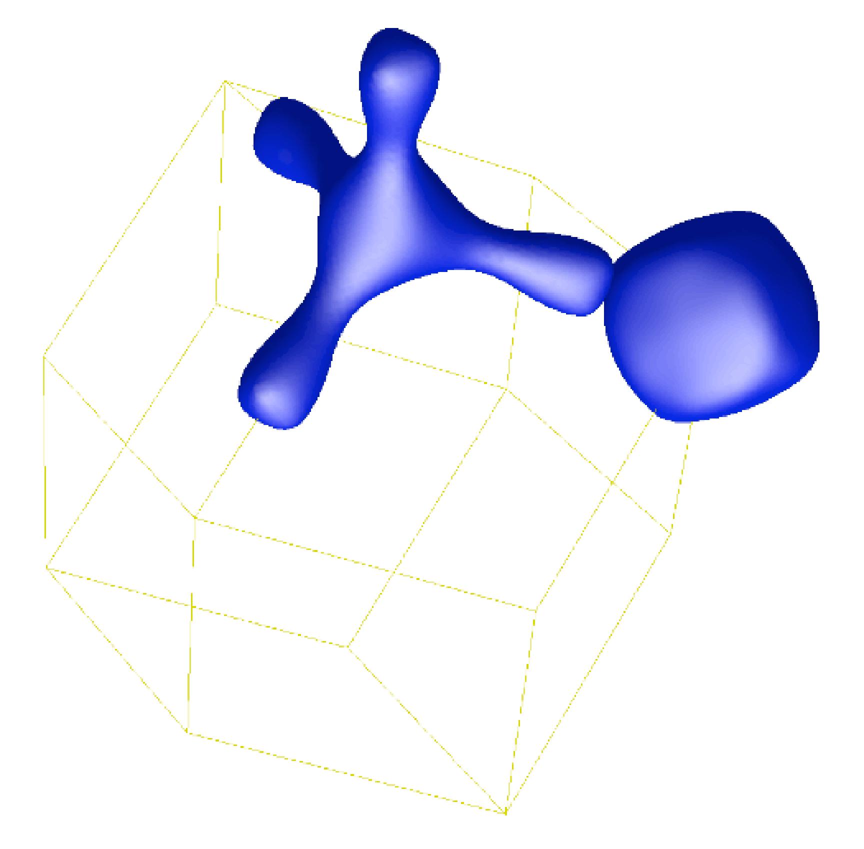
Fermi surface

Generalized susceptibility

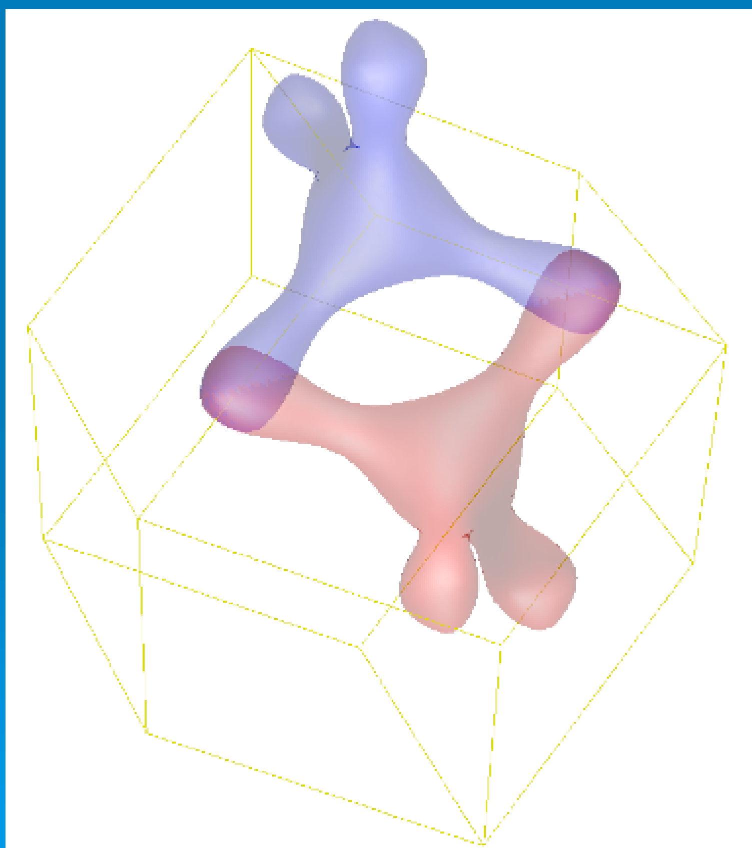
$$\chi(\omega, \mathbf{q}) = \sum_{\mathbf{k}} \frac{f(\epsilon_{\mathbf{k}+\mathbf{q}}) - f(\epsilon_{\mathbf{k}})}{\omega + \epsilon_{\mathbf{k}+\mathbf{q}} - \epsilon_{\mathbf{k}}}$$

Two sheets of FS:

Lobed tetrahedron - P
Rounded cube - H



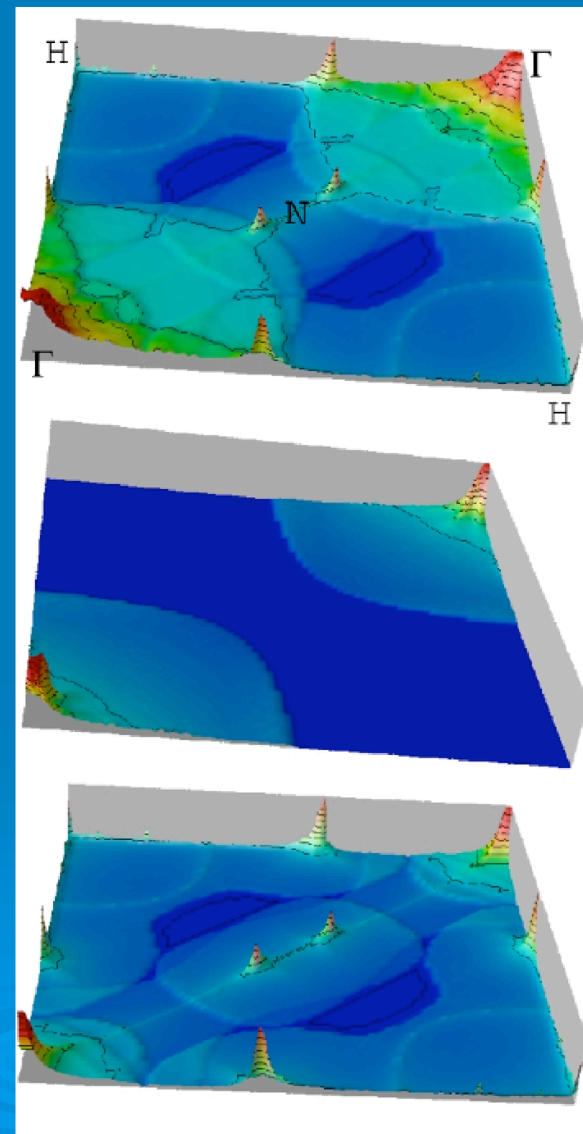
Generalized susceptibility



Total:

Rounded
 cube:

Lobed
 tetrahedron:



bcc Eu - summary

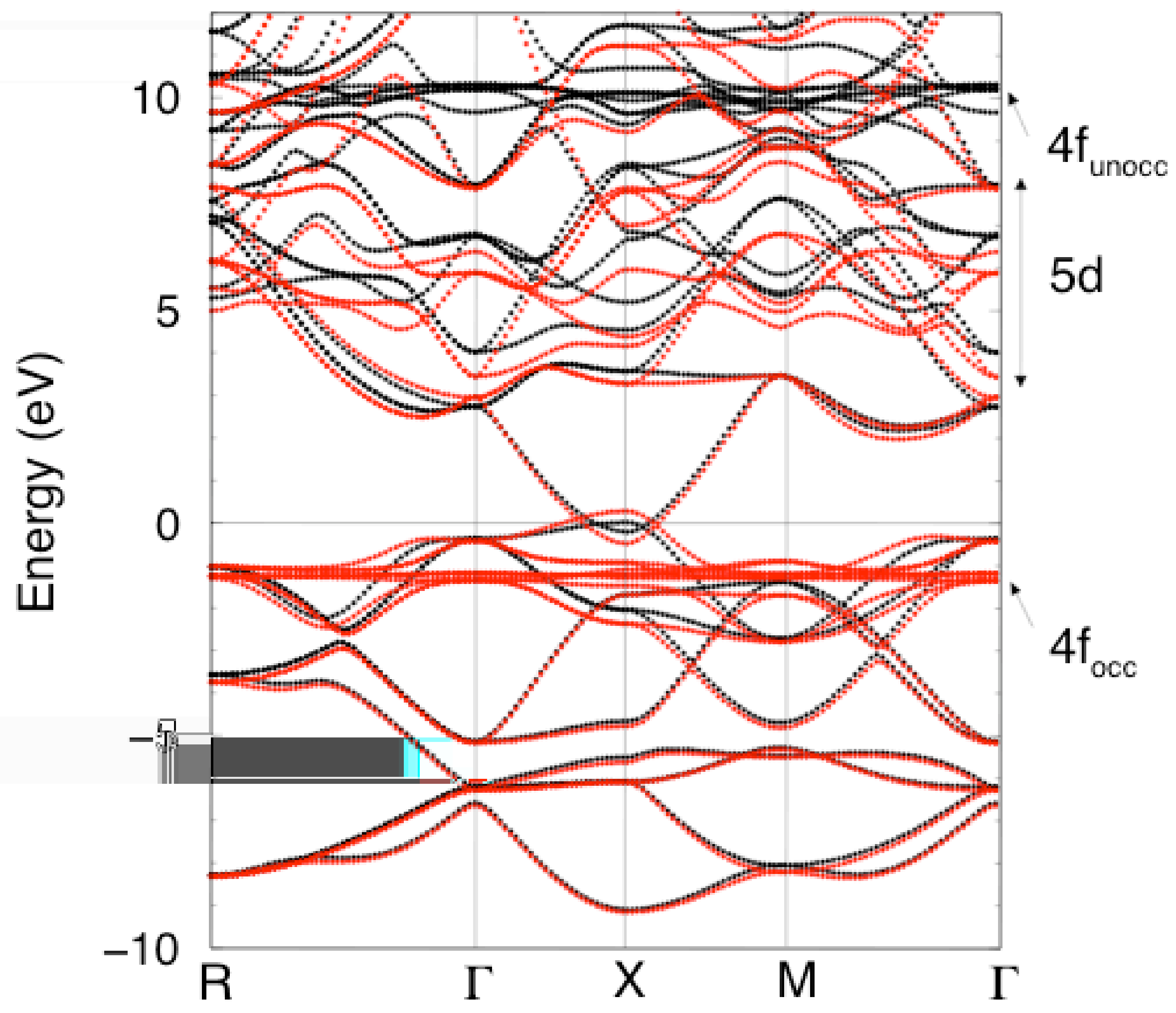
- Ordering temperature T=112 K (exp.91 K)
- Spiral propagation vector q||cubic axis associated with a nesting feature of the paramagnetic FS
- $JS_f.s_d$ coupling of local moments
- RKKY type of inter-site coupling

EuB_6

- Simple cubic structure
- Eu^{2+} valency
- Ground state unclear (semimetal vs narrow gap insulator
- Two magnetic transitions (15.1K & 12.7 K)
- Collosal magneto-resistence

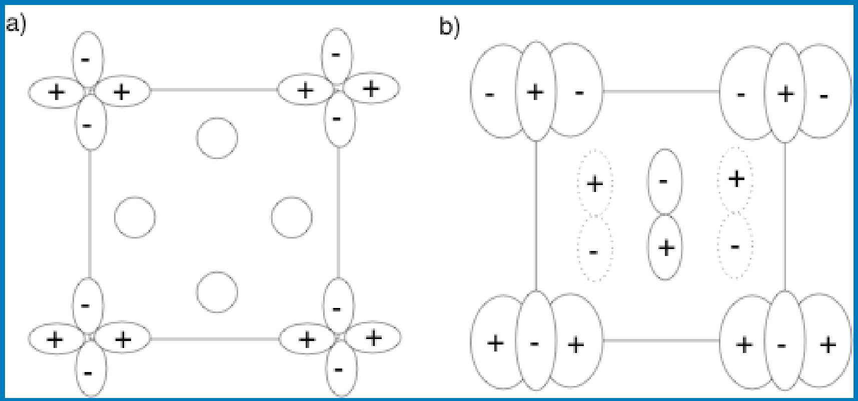
Nature of ground state? Magnetic properties?

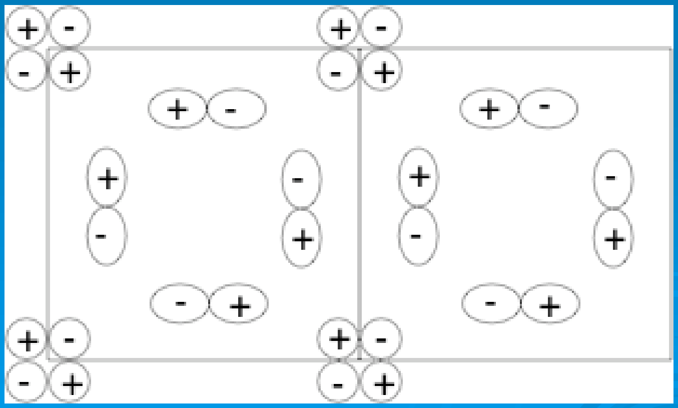
Spin polarized band structure



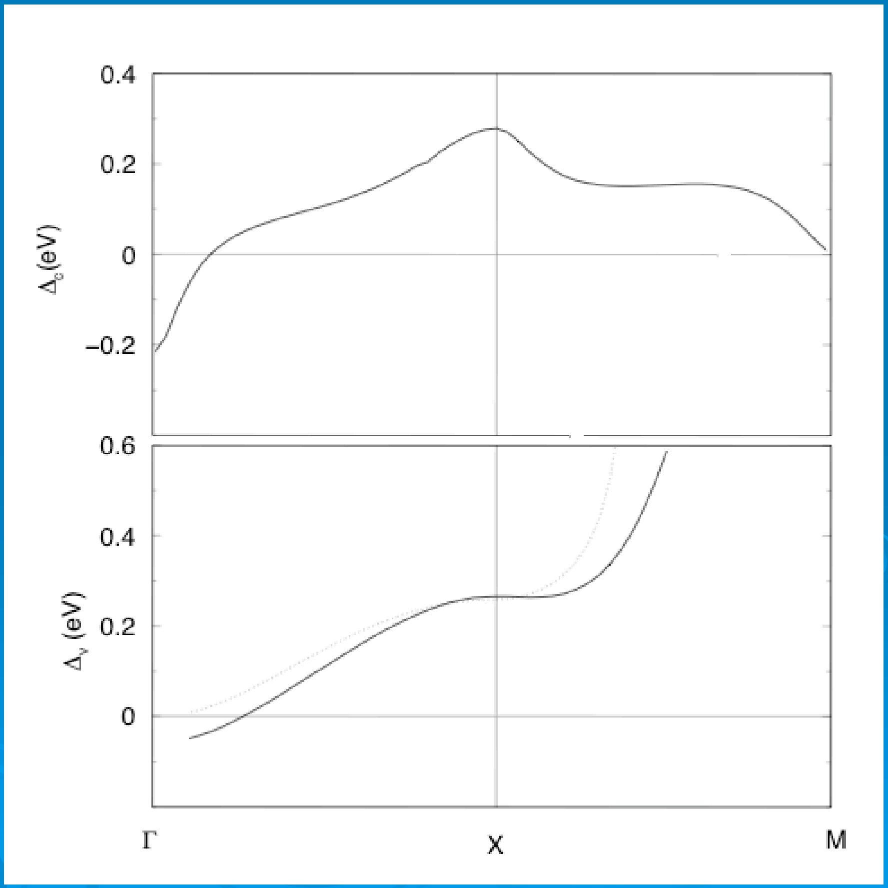
Exchange splitting

Orbitals at X point:





Splitting between spin up and down bands:



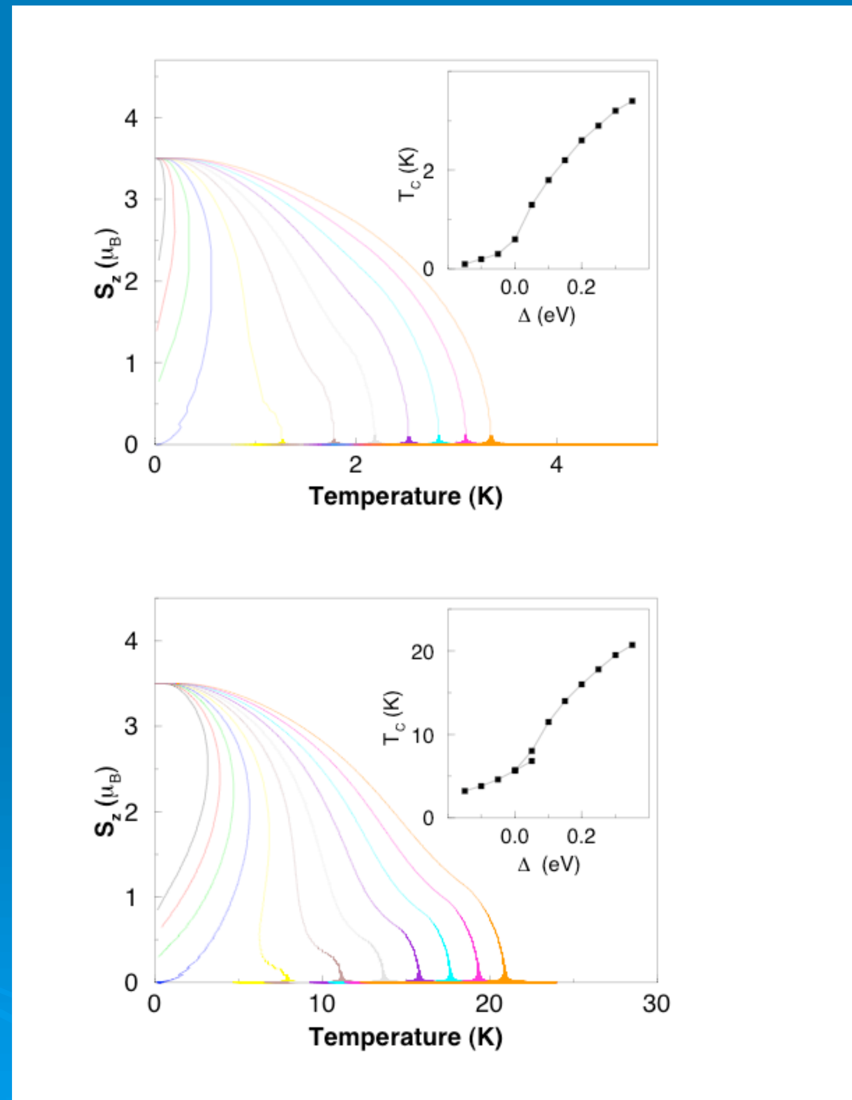
Molecular field model

Magnetization curves as a function of band gap (overlap) and exchange parameter J:

$$H_{MF} = \sum_{\mathbf{k},\sigma}[\epsilon^v_{\mathbf{k}\sigma} v^\dagger_{\mathbf{k}\sigma} v_{\mathbf{k}\sigma} + \epsilon^c_{\mathbf{k}\sigma} c^\dagger_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma}] - \sum_i h S^z_i$$

$$h = \sum_\sigma \sigma(J^c n^\sigma_c(S^z) - J^v n^\sigma_v(S^z))$$

$$S^z = B_{7/2}(\frac{h}{k_B T}),$$



EuB_6 - summary

- The local moments are FM coupled to the conduction band and AFM coupled to the valence band
- AFM coupling can be understood in terms of band mixing (Schrieffer-Wolff transformation), FM coupling is due to intra-atomic f-d exchange
- This type of exchange leads to magnetization dependent carrier concentration