

# Fluctuating exchange theory of dynamical electron correlations and magnetism

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Grant Agency of ASCR: project IAA100100616

# INTRODUCTION

DFT inappropriate for ground states of correlated systems such as

- transition metal oxides
- rare earths and their compounds
- actinides and their compounds

DFT is also inappropriate for excited states (spectroscopies)

correlation effects have to be included beyond the DFT

# OUTLINE

- Multiband Hubbard Hamiltonian
- Solution of the many-body problem
- FLEX approximation
- Computational procedure
- Results and discussion
- Conclusions and outlook

# MULTIBAND HUBBARD HAMILTONIAN

$$\begin{aligned}
 H^{\text{Hubb}} &= \sum_{\mathbf{R}\lambda, \mathbf{R}'\lambda'} t_{\mathbf{R}\lambda, \mathbf{R}'\lambda'} a_{\mathbf{R}\lambda}^{\dagger} a_{\mathbf{R}'\lambda'} \\
 &+ \sum_{\mathbf{R}, \lambda, \lambda', \lambda'', \lambda'''} \langle \mathbf{R}\lambda, \mathbf{R}\lambda' | V | \mathbf{R}\lambda'' \mathbf{R}\lambda''' \rangle a_{\mathbf{R}\lambda}^{\dagger} a_{\mathbf{R}\lambda'}^{\dagger} a_{\mathbf{R}\lambda''} a_{\mathbf{R}\lambda'''}
 \end{aligned}$$

$$t_{\lambda, \lambda'}(\mathbf{k}) = \sum_{\mathbf{R}'} e^{-i\mathbf{k}(\mathbf{R}-\mathbf{R}')} t_{\mathbf{R}\lambda, \mathbf{R}'\lambda'} \leftrightarrow E_n(\mathbf{k}) \quad \dots \text{energy bands}$$

$$H^{\text{int}} = \sum_{i, j, k, l} \sum_{\sigma, \sigma'} \langle i\sigma j\sigma' | V | k\sigma l\sigma' \rangle a_{i\sigma}^{\dagger} a_{j\sigma'}^{\dagger} a_{k\sigma} a_{l\sigma'}$$

$$\langle i\sigma j\sigma' | V | k\sigma l\sigma' \rangle \approx \delta_{ik} \delta_{jl} (1 - \delta_{ij} \delta_{\sigma\sigma'}) U + \delta_{il} \delta_{jk} (1 - \delta_{ij}) \delta_{\sigma\sigma'} J$$

$\mathbf{R}$  ... lattice site,  $\lambda = (L\sigma) = (\ell m\sigma)$  ... spinorbital

# GREEN FUNCTIONS

$$G_{ij}^c(t) = -i \langle \Psi_0 | T[a_i(t) a_j^\dagger(0)] | \Psi_0 \rangle$$

$$G_{ij}^r(t) = -i \theta(t) \langle \Psi_0 | [a_i(t), a_j^\dagger(0)]_+ | \Psi_0 \rangle$$

$$G_{ij}^a(t) = i \theta(-t) \langle \Psi_0 | [a_i(t), a_j^\dagger(0)]_+ | \Psi_0 \rangle$$

$$A(t) = \exp(iHt) A(0) \exp(-iHt) \quad \dots \text{Heisenberg picture}$$

$$G_{ijkl}^c(t_3, t_2, t_1, t_0) = -i \langle \Psi_0 | T[a_i(t_3) a_j(t_2) a_k^\dagger(t_1) a_l^\dagger(t_0)] | \Psi_0 \rangle$$

$$G(E) = \int_{-\infty}^{\infty} dt e^{i\omega t} G(t), \quad G(t) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} e^{-i\omega t} G(\omega)$$

# PERTURBATION THEORY AT T=0 K

$$H = H_0 + H_{\text{int}},$$

$$H_{\text{int}}(t) = e^{iH_0 t} H_{\text{int}} e^{-iH_0 t},$$

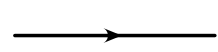
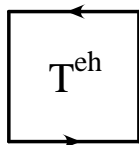
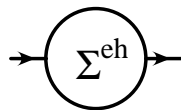
$$G_{\alpha\beta}(t_\alpha, t_\beta) = \frac{-i \langle \Phi_0 | T \{ a_\alpha(t_\alpha) a_\beta^\dagger(t_\beta) S(\infty) \} | \Phi_0 \rangle}{\langle \Phi_0 | S(\infty) | \Phi_0 \rangle},$$

$$S(\infty) = T \exp \left\{ -i \int_{-\infty}^{\infty} dt H_{\text{int}}(t) \right\}.$$

$|\Phi_0\rangle$  ... ground state of the non-interacting system (with Hamiltonian  $H_0$ ).

# PERTURBATION THEORY AT T=0 K cont'd

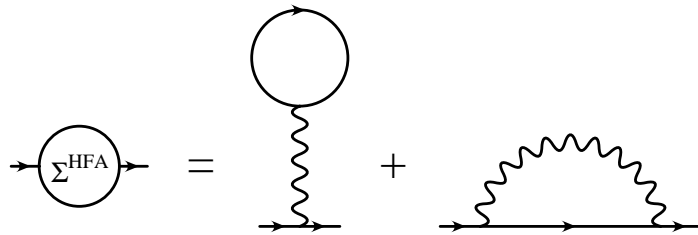
## Vocabulary for diagrams:

 $iG_0$  $G_0$  ... GF of noninteracting system $iG$  $G$  ... GF of interacting system $-iU$  $U$  ... pair interaction $-iv$  $v$  ... antisymmetrized pair interaction $-iT^{eh}$  $T$  ... T-matrix $-i\Sigma^{eh}$  $\Sigma$  ... selfenergy

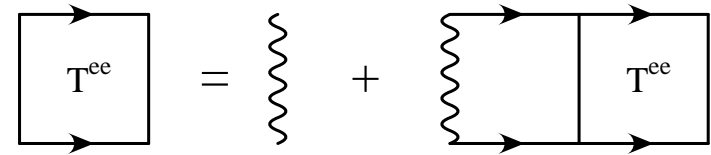
# FLEX

N. Bickers and D. Scalapino, Ann. Phys. (N.Y.) **193**, 206 (1989).

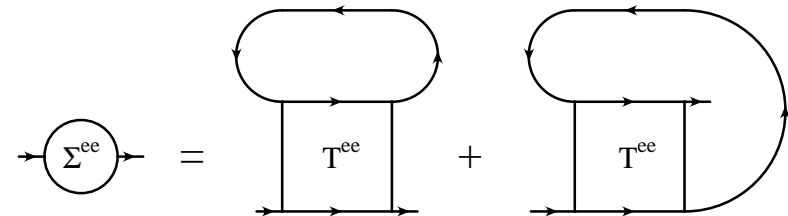
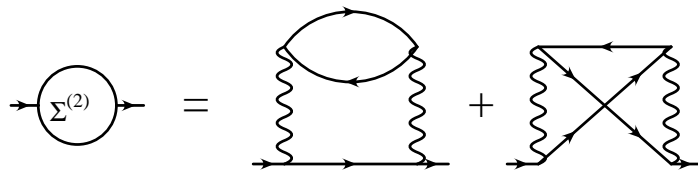
Hartree-Fock



electron-electron channel

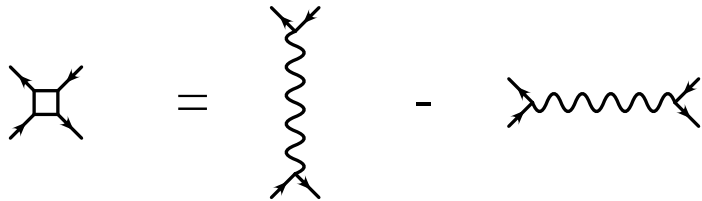


second order



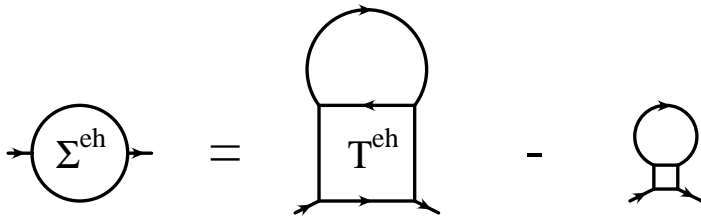
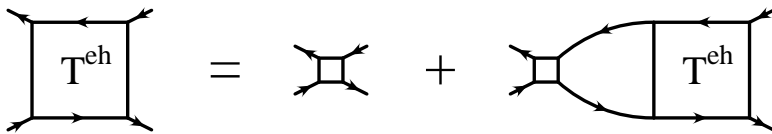
# FLEX: electron-hole channel

antisymmetrized interaction

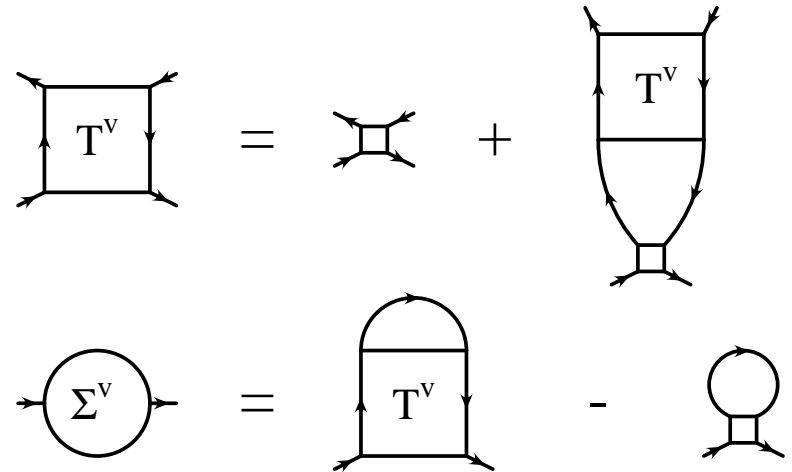


$$v_{ab} = (U - J\delta_{\sigma_a, \sigma_b})(1 - \delta_{ab})$$

RPA



GW (vertical channel)



features

- $\Phi$  – derivable
- conserving approximation
- infinite lifetime at  $E_F$

# SELECTED REFERENCES

## DMFT

A. Georges, G. Kotliar, W. Krauth, and M. Rozenberg, Rev. Mod. Phys. **68**, 13 (1996).

## LDA+DMFT

G. Kotliar, S. Y. Savrasov, K. Haule, V. S. Oudovenko, O. Parcollet, and C. A. Marianetti, Rev. Mod. Phys. **78**, 865 (2006).

## FLEX on real energy axis

Yu. A. Babanov, V. Ye. Naish, O. B. Sokolov, and V. K. Finashkin: Fizika Metallov i Metallovedeniye **32** (1971) 472-484, **35** (1973) 1123-1132, **35** (1973) 1132-1146.

## tests of FLEX for multiband model

V. Drchal et al., J. Phys. Cond. Matter **17** (2005), 61-74.

## FLEX+DMFT for actinide compounds, spin-orbit

L. V. Pourovskii, M. I. Katsnelson, and A. I. Lichtenstein, Phys. Rev. **72**, 115106 (2005).

# OUR APPROACH

- correlation effects for d-states only
- local approximation for selfenergy, i.e.,  $\Sigma_{\mathbf{R}\lambda,\mathbf{R}'\lambda'}(z) = \Sigma_{\mathbf{R}\lambda,\mathbf{R}'\lambda'}(z) \delta_{\mathbf{R},\mathbf{R}'}$ , reasonable for 3-dim systems, while for 2-dim and 1-dim systems is probably not sufficient
- perturbation theory: for simplicity FLEX (which depends only on one energy argument)
- sum up one or more classes of diagrams and derive equations for causal quantities GFs and for selfenergies
- the equations are transformed into equations for retarded quantities which have simpler analytic properties
- subtract contributions of first and second order to avoid double counting
- energy representation: energies along the axis of real energies
- assume  $T = 0$

# EQUATIONS FOR CAUSAL QUANTITIES

for example e-e channel:

$$\Psi(E) = i \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} G(E - \omega)G(\omega) \quad \dots \text{two-particle propagator}$$

$$T(E) = \frac{v}{1 - v\Psi(E)} \quad \dots \text{e-e T-matrix}$$

$$\Sigma(E) = -i \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} T(E + \omega)G(\omega) \quad \dots \text{e-e selfenergy}$$

# EQUATIONS FOR RETARDED QUANTITIES

$$G^c(E) = \text{Re}G^r(E) + i \text{sgn}(E - \mu) \text{Im}G^r(E)$$

$$\Psi_2(E) = - \int_{\mu}^{E-\mu} \frac{d\omega}{\pi} f_2(E - \omega) g_2(\omega)$$

$$T(E) = \frac{v}{1 - v\Psi(E)}$$

$$\Sigma_2(E) = - \int_{\mu}^E \frac{d\omega}{\pi} T_2(E - \omega) G_2(\omega)$$

$$\text{Re}f(E) = \mathcal{P} \int_{-\infty}^{\infty} \frac{d\omega}{\pi} \frac{\text{Im}f(\omega)}{\omega - E}, \quad \text{Im}f(E) = -\mathcal{P} \int_{-\infty}^{\infty} \frac{d\omega}{\pi} \frac{\text{Re}f(\omega)}{\omega - E}$$

... Hilbert transform

# SELFCONSISTENCY

- selfenergy  $\Sigma$  is constructed from unperturbed  $G_0$ , selfconsistency only for chemical potential  $\mu$
- selfenergy  $\Sigma$  is constructed from the perturbed GF  $G$
- DMFT selfconsistency: selfenergy  $\Sigma$  is constructed from the bath GF  $\mathcal{G}_0$  defined as

$$\mathcal{G}_0^{-1} = G^{-1} + \Sigma, \quad \Sigma = \Sigma[\mathcal{G}_0]$$

- DMFT+LDA selfconsistency: as the DMFT, but the perturbed GF  $G$  is used to calculate charge density used in the LDA

# MANY-BODY THEORY & TB-LMTO METHOD

$$H_{\mathbf{R}\lambda, \mathbf{R}'\lambda'}^{\text{LMTO}} = C_{\mathbf{R}\lambda} \delta_{\mathbf{R}\mathbf{R}'} \delta_{\lambda\lambda'} + \Delta_{\mathbf{R}\lambda}^{1/2} S_{\mathbf{R}\lambda, \mathbf{R}'\lambda'}^{\gamma} \Delta_{\mathbf{R}'\lambda'}^{1/2}$$

$$C_{\mathbf{R}\lambda} \rightarrow \tilde{C}_{\mathbf{R}\lambda}(z) = C_{\mathbf{R}L} + \Sigma_{\mathbf{R}\lambda}(z)$$

# NUMERICAL IMPLEMENTATION

cubic  $3d$  metals

bcc (Fe) 1632 k-points in the IBZ

fcc (Co, Ni, Cu) 2480 k-points in the IBZ

experimental lattice constant

Ceperley-Alder exchange-correlation energy as parameterized by  
Perdew and Zunger

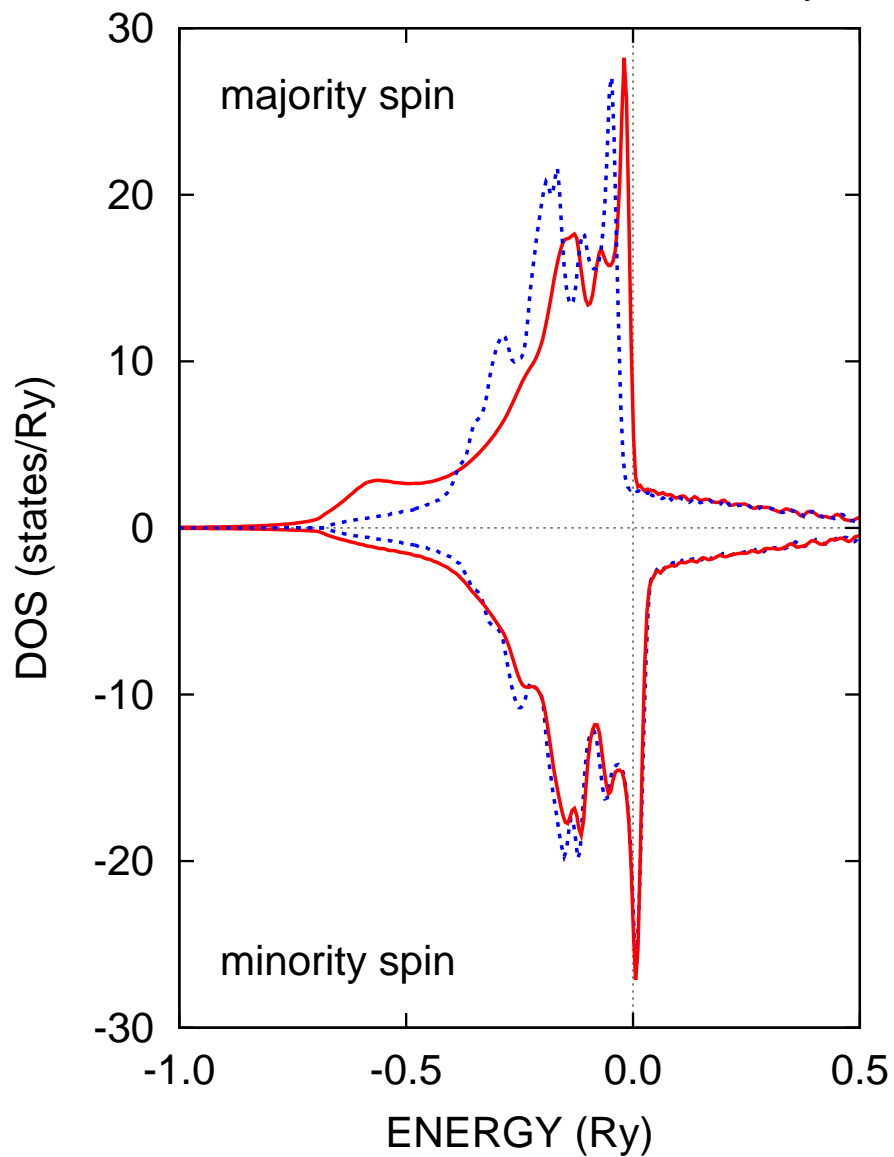
many-body equations solved using Broyden method

	$U$ [Ry]	$J$ [Ry]
bcc-Fe	0.12	0.08
fcc-Ni	0.18	0.12

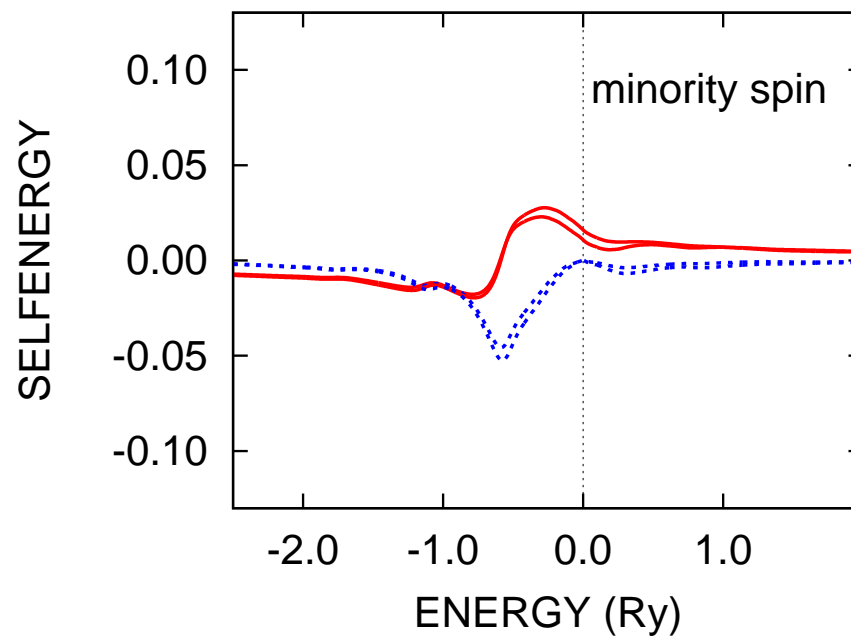
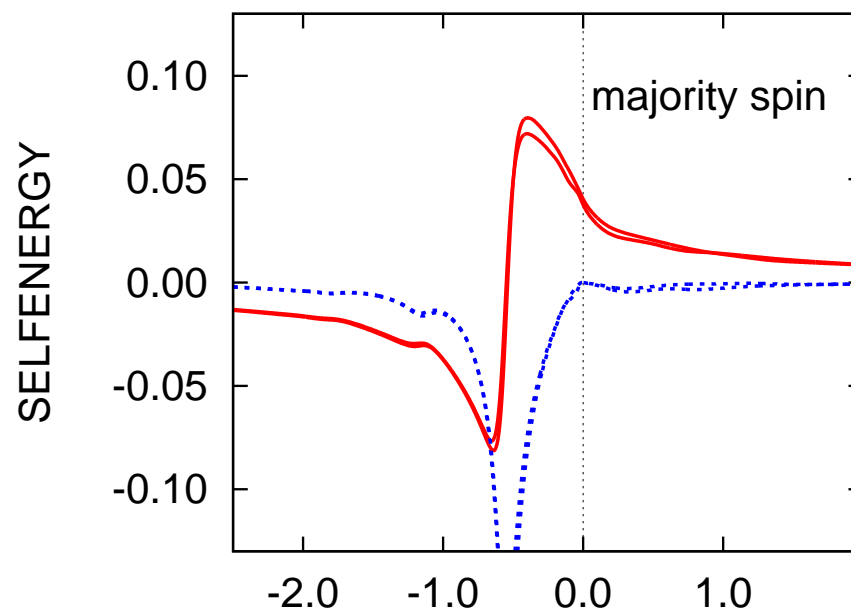
# fcc NICKEL

ferromagnetic fcc Ni

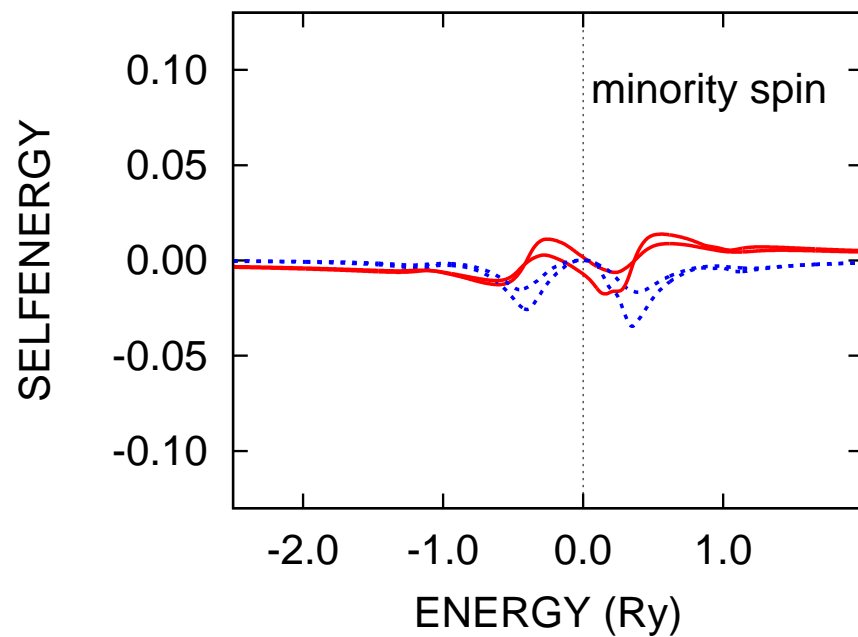
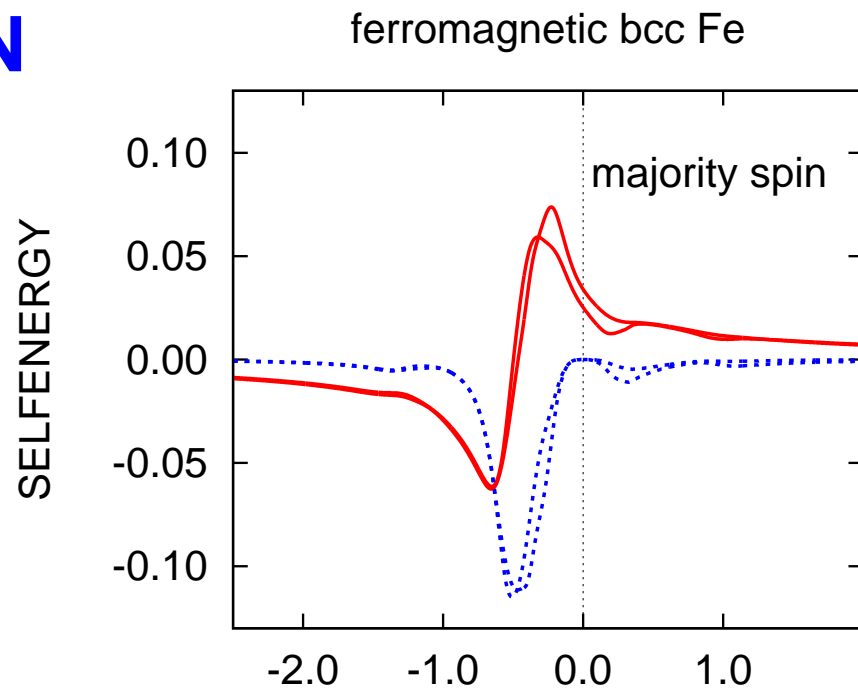
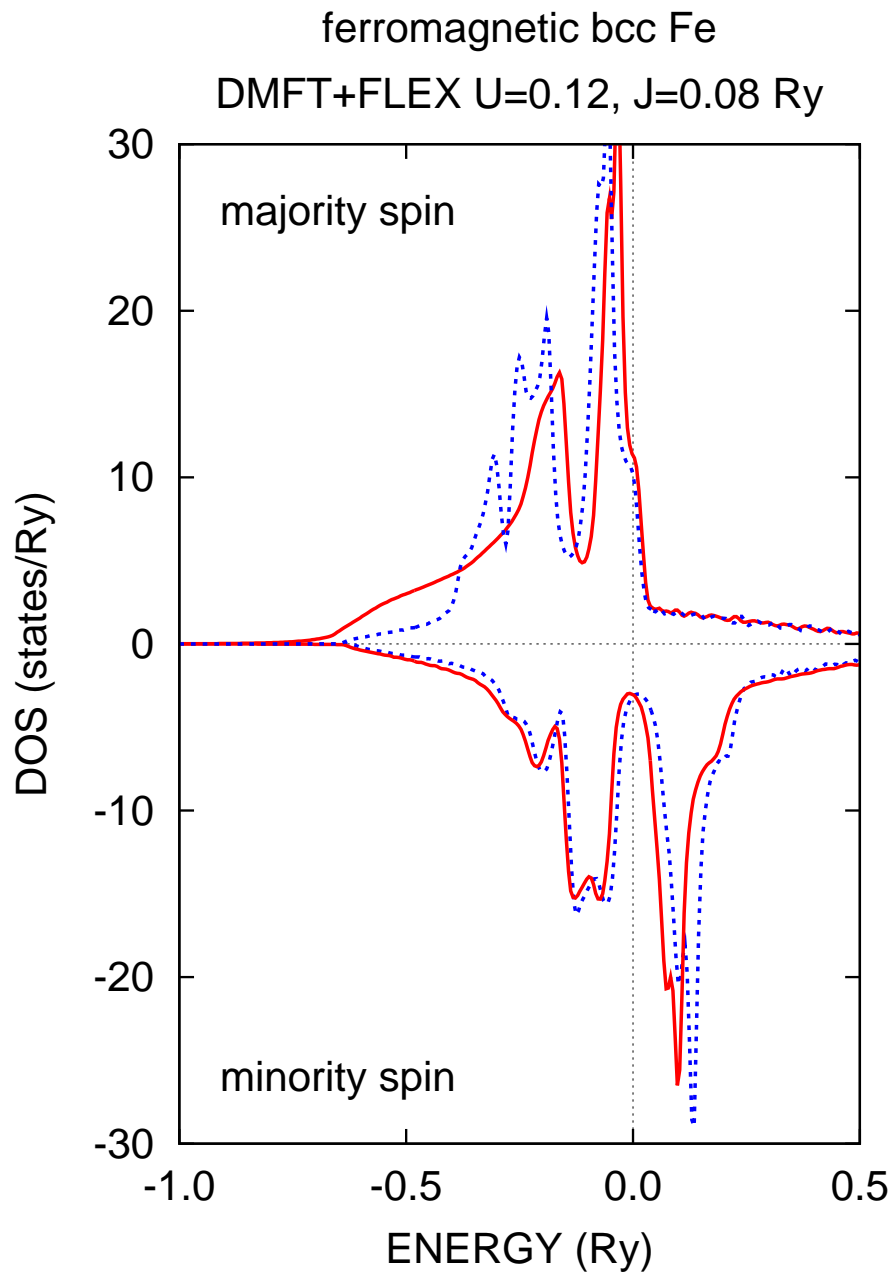
DMFT+FLEX  $U=0.18, J=0.12$  Ry



ferromagnetic fcc Ni



# bcc IRON



# RESULTS

	$m^{\text{LDA}} [\mu_B]$	$m^{\text{FLEX}} [\mu_B]$	$m^{\text{exp}} [\mu_B]$
bcc-Fe	2.237	2.128	2.216
fcc-Ni	0.633	0.601	0.616

Ni: satellite at -6 eV

broadening due to the  $\text{Im}\Sigma$

infinite lifetime of quasiparticles at Fermi energy (Luttinger theorem)

# CONCLUSIONS AND OUTLOOK

- FLEX good for weak interactions
- other systems such as DMS
- actinides: relativistic formulation necessary (other basis:  $|m\sigma\rangle \rightarrow |\kappa\mu\rangle$ ), jj-coupling
- search for other impurity solvers suitable for stronger interactions