Correlated electronic structures and spin-dynamics, some recent investigations

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Collaborators

Bergman, Bergqvist, Björkman, Blügel, Burlamaqui-Klautau, Di Marco, Delin, Etz, Grechnev, Grånäs,Hellsvik, Johansson, Katsnelson, Keshavarz, Kirilouk, Kvashnin, Lebegue, Lichtenstein, Locht, Nordström, Pereiro, Peters, Rodriques, Skubic, Svane, Szilva, Thunström, Wills

Overview

1. An electronic structure data-base
2. DMFT implementation with examples
3. Calculation of exchange parameters
4. Spin-wave excitations
5. Cohesion and spectra of rare-earths

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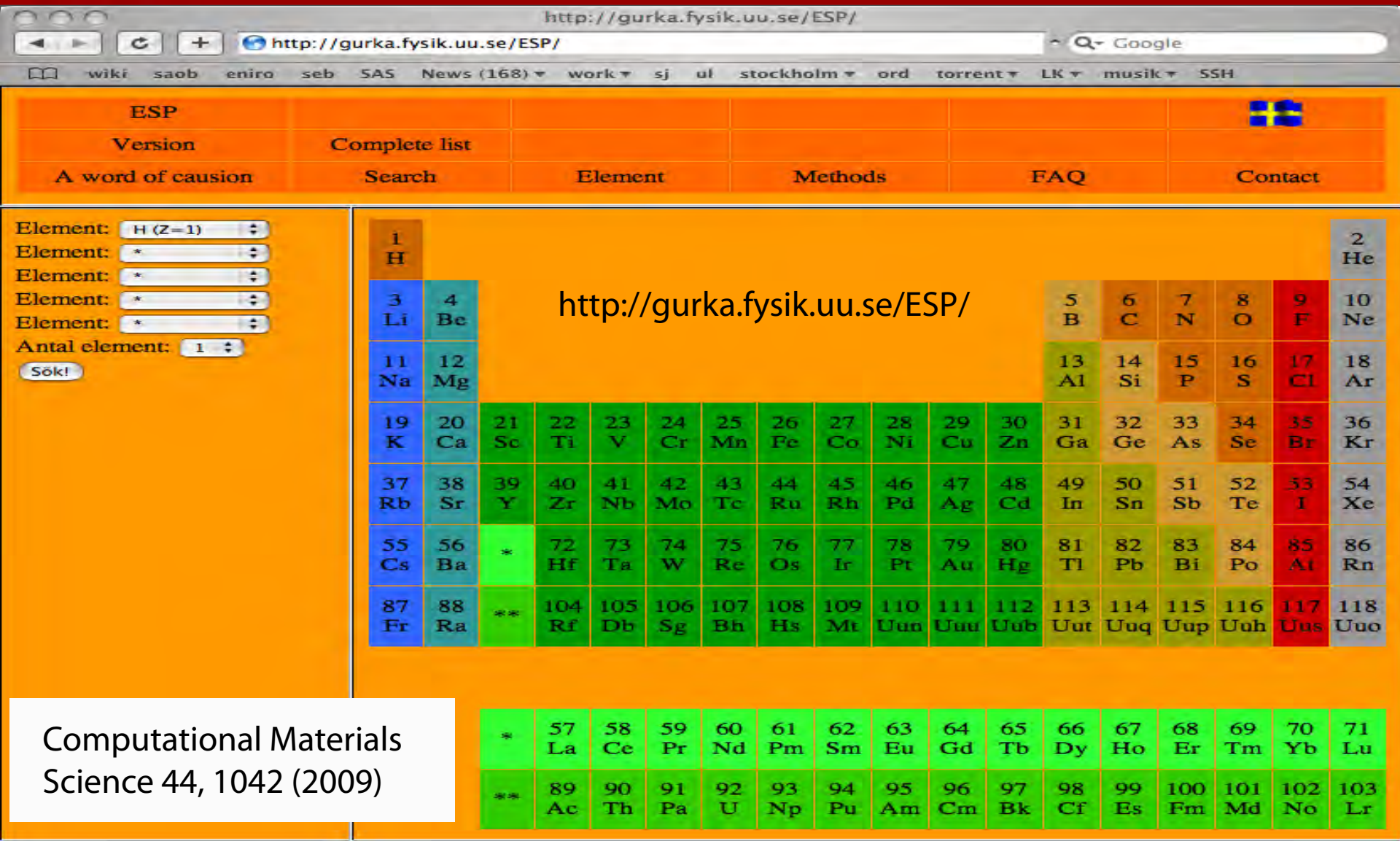
Electronic structure database



Computational Materials Science 44, 1042 (2009)

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Possible high temperature superconductors

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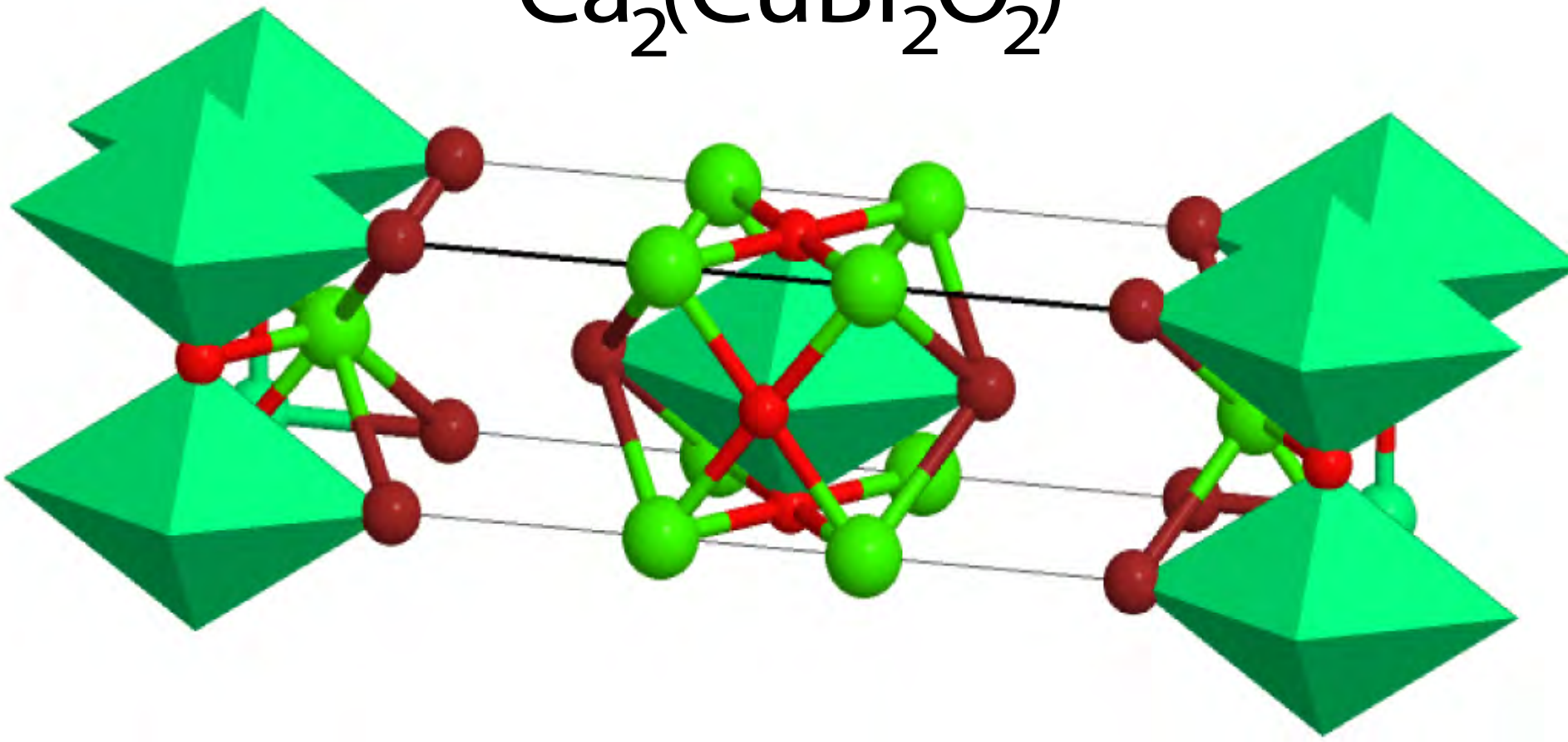
Data-mining criterion-structure

$Ca_2(CuBr_2O_2)$



Computational Materials Science 67 282 (2013)

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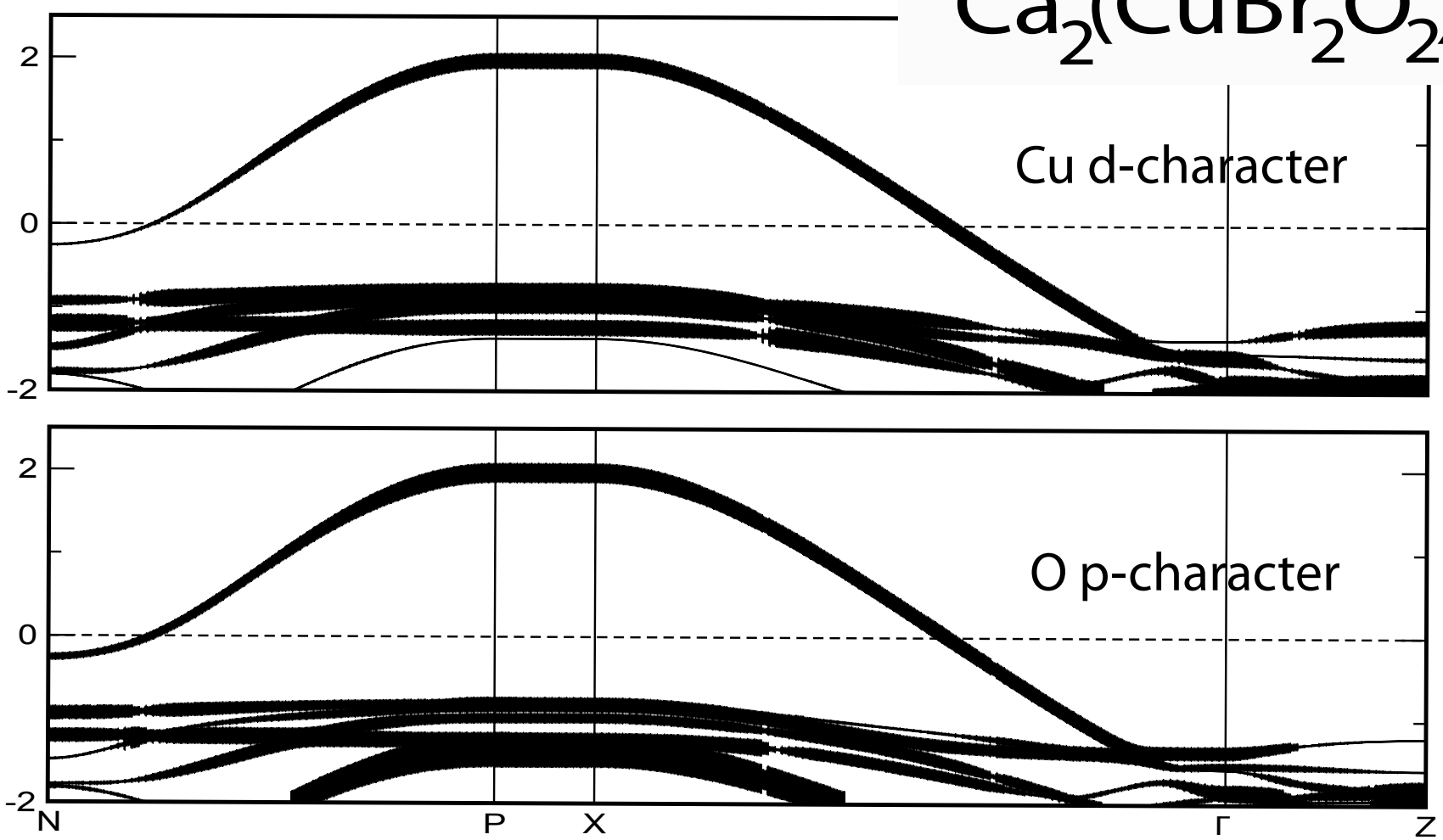
Data-mining criterion-bands/bonds

$Ca_2(CuBr_2O_2)$



Computational Materials Science 67 282 (2013)

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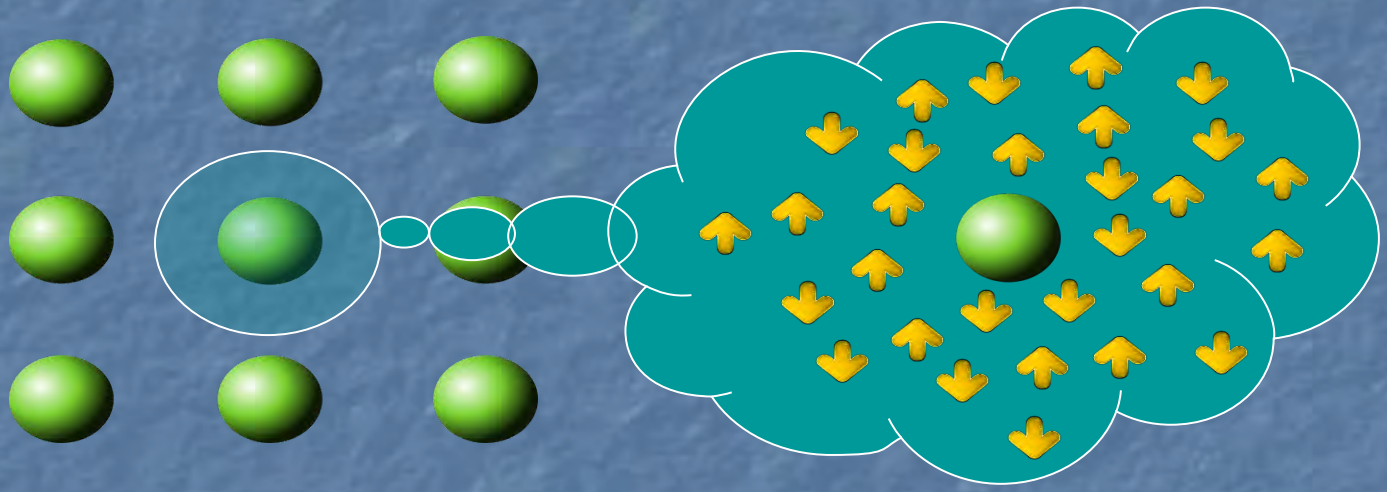
Dynamical mean field theory

$$H = H_{LDA} + \sum_{R} \sum_{\xi_1\xi_2\xi_3\xi_4} U_{\xi_1\xi_2\xi_3\xi_4} c^{\dagger}_{R\xi_1} c^{\dagger}_{R\xi_2} c_{R\xi_3} c_{R\xi_4}$$

The Hubbard model is mapped into an Anderson Impurity Model



FICTITIOUS SYSTEM REPRODUCING THE DYNAMICS

The mapping is made with the condition of preserving the local Green's function and is exact in the limit of infinite nearest neighbors

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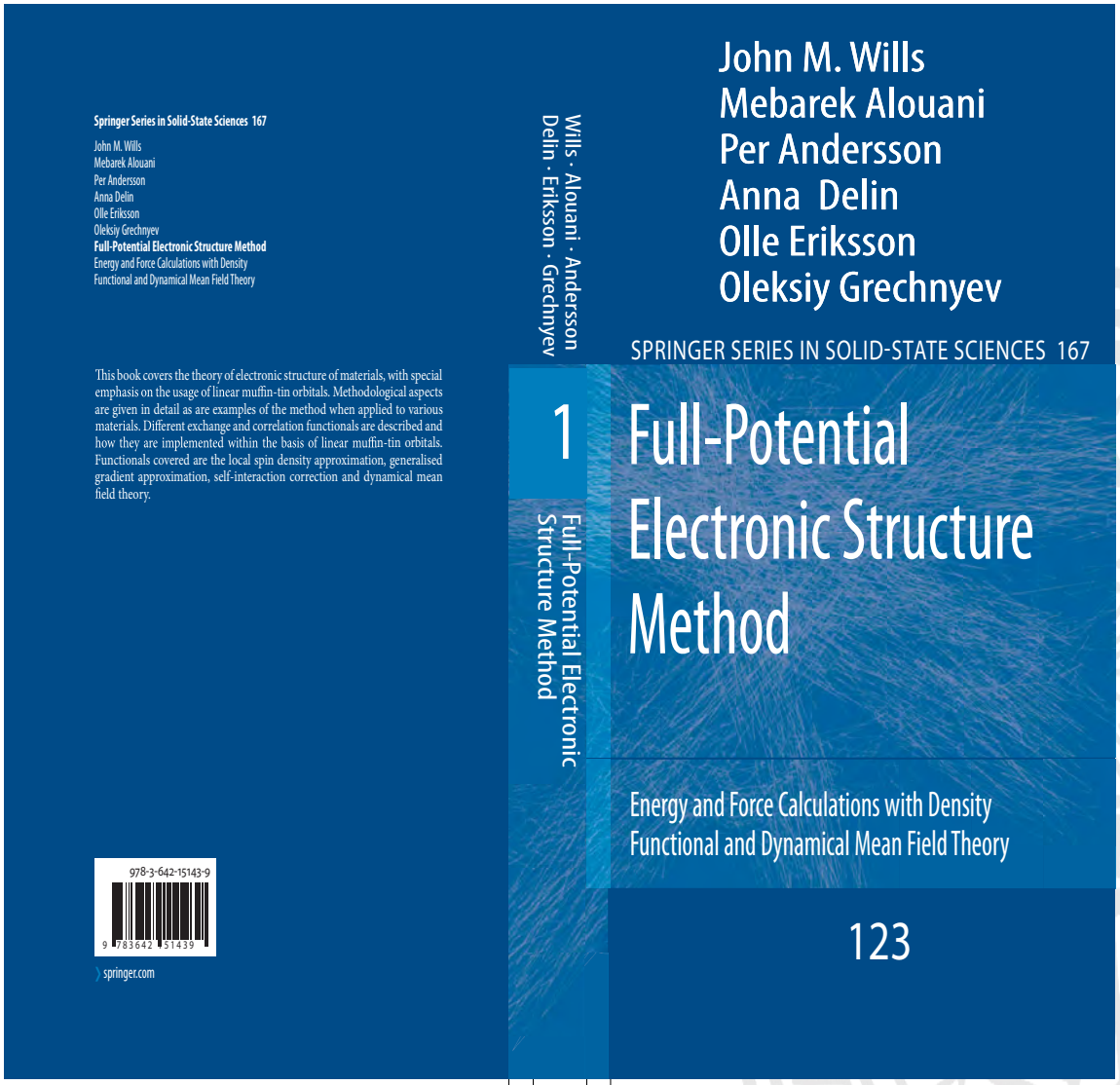
FP-LMTO



DMFT implementation:

1. Grechnev, Di Marco et al.
PRB, 76, 35107 (2007)
2. Di Marco et al
PRB 79 115111 (2009)
3. Thunström et al.
PRB 79, 165104 (2009)
4. Grånäs et al.
Comp. Mat. Sci. 55, 295 (2012)
5. Thunström, Di Marco et al.
PRL 109, 186401 (2012).

Katsnelson+Lichtenstein

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Correlated basis

$$\hat{\boldsymbol{A}}_R \equiv \sum_{\xi,\xi'} |R,\xi\rangle\langle R,\xi| \sum_{\mathbf{k}} \hat{\boldsymbol{A}}_{\mathbf{k}} |R,\xi'\rangle\langle R,\xi'|$$

Two choices of $|R,\xi\rangle$, muffin-tin based and orthogonal:

i) MT

$$\chi_{lm} = i^l Y_{lm}(a\phi_{l\nu} + b\phi'_{l\nu})$$
$$\chi_{lm} = i^l Y_{lm} n_l$$

ii) ORT

$$(\mathbf{H} - \epsilon\mathbf{O})\mathbf{x} = 0$$
$$\mathbf{O} = \mathbf{L}\mathbf{L}^h$$

Cholesky decomposition gives

$$(\mathbf{L}^{-1}\mathbf{H}\mathbf{L}^{-h} - \epsilon\mathbf{1})\mathbf{y} = 0$$
$$\mathbf{y} = \mathbf{L}^h\mathbf{x}$$

Exact Diagonalization Solver

The finite size problem can be solved exactly with a direct construction of all the accessible many-body states.

N=5 electrons in K=10 orbitals:

$$
\begin{aligned}
|\Psi_1^5\rangle &= |1111100000\rangle, \\
|\Psi_2^5\rangle &= |1111010000\rangle, \\
&\vdots \\
|\Psi_M^5\rangle &= |0000011111\rangle.
\end{aligned}
$$

M corresponds to $\binom{K}{N}$

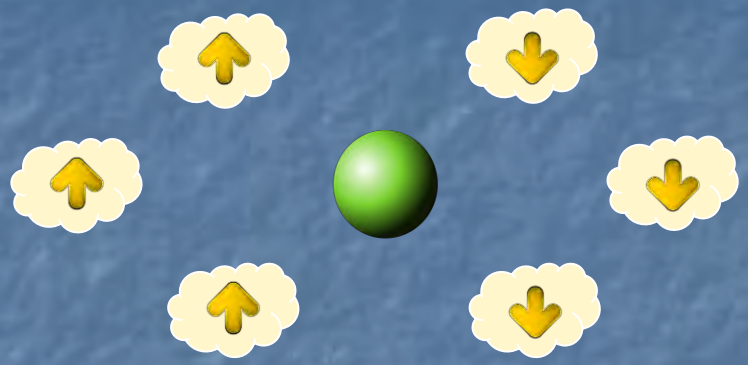
Too large for standard computational resources!

Block diagonalization ⟶ up to 30 bath states!

Exact Diagonalization Solver

The finite size problem can be solved exactly with a direct construction of all the accessible many-body states.

N=5 electrons in K=10 orbitals:



Once the many-body states have been determined, the one-particle Green's function can be obtained through the Lehmann representation

$$G^{\mathrm{ED}}(i\omega)_{\xi_1\xi_2} = \frac{1}{Z}\sum_{\nu\mu}\frac{\langle\mu|\hat{c}_{\xi_1}|\nu\rangle\langle\nu|\hat{c}^{\dagger}_{\xi_1}|\mu\rangle}{i\omega + E_\mu - E_\nu}\left(e^{-\beta E_\mu} + e^{-\beta E_\nu}\right)$$

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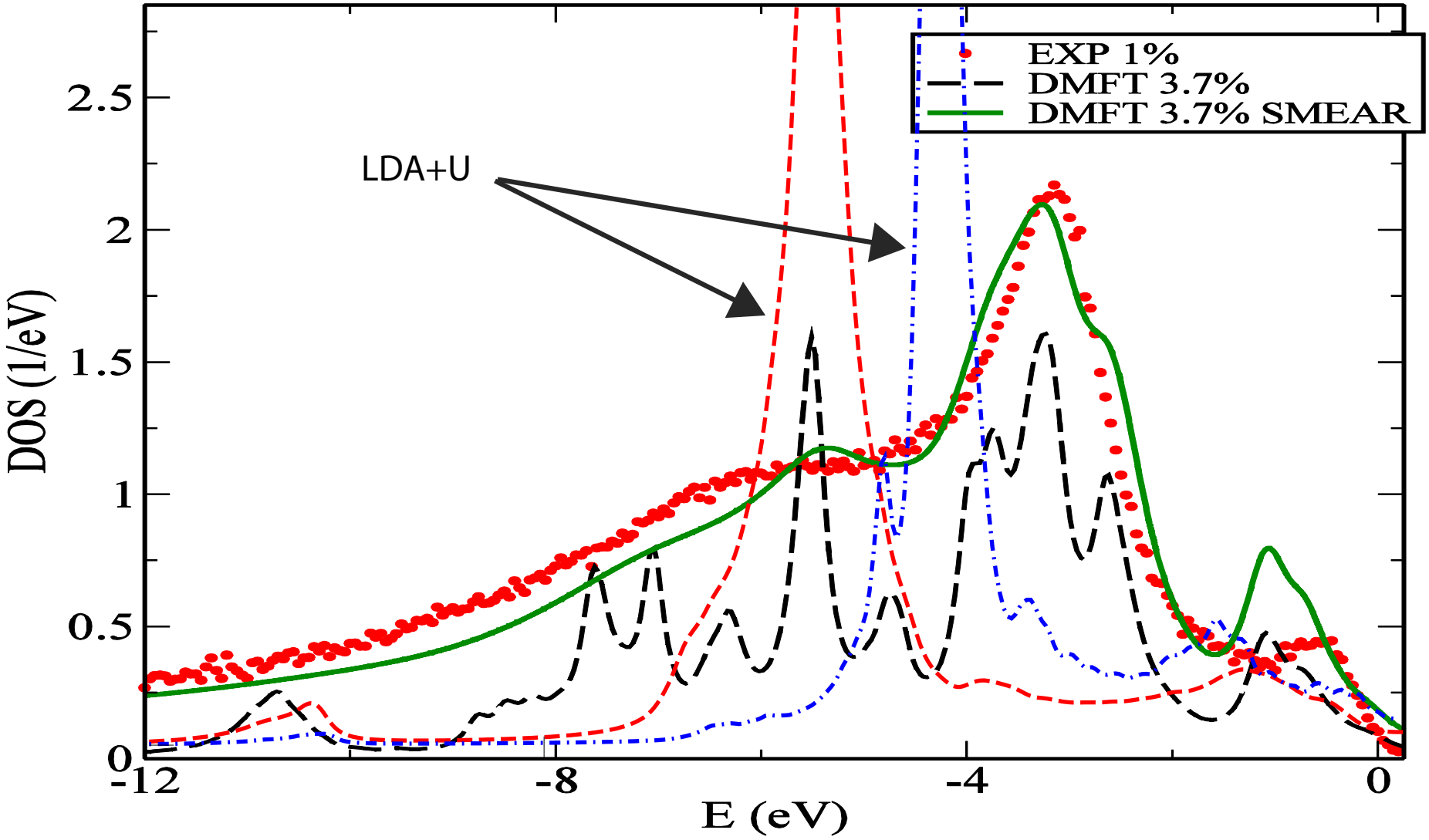
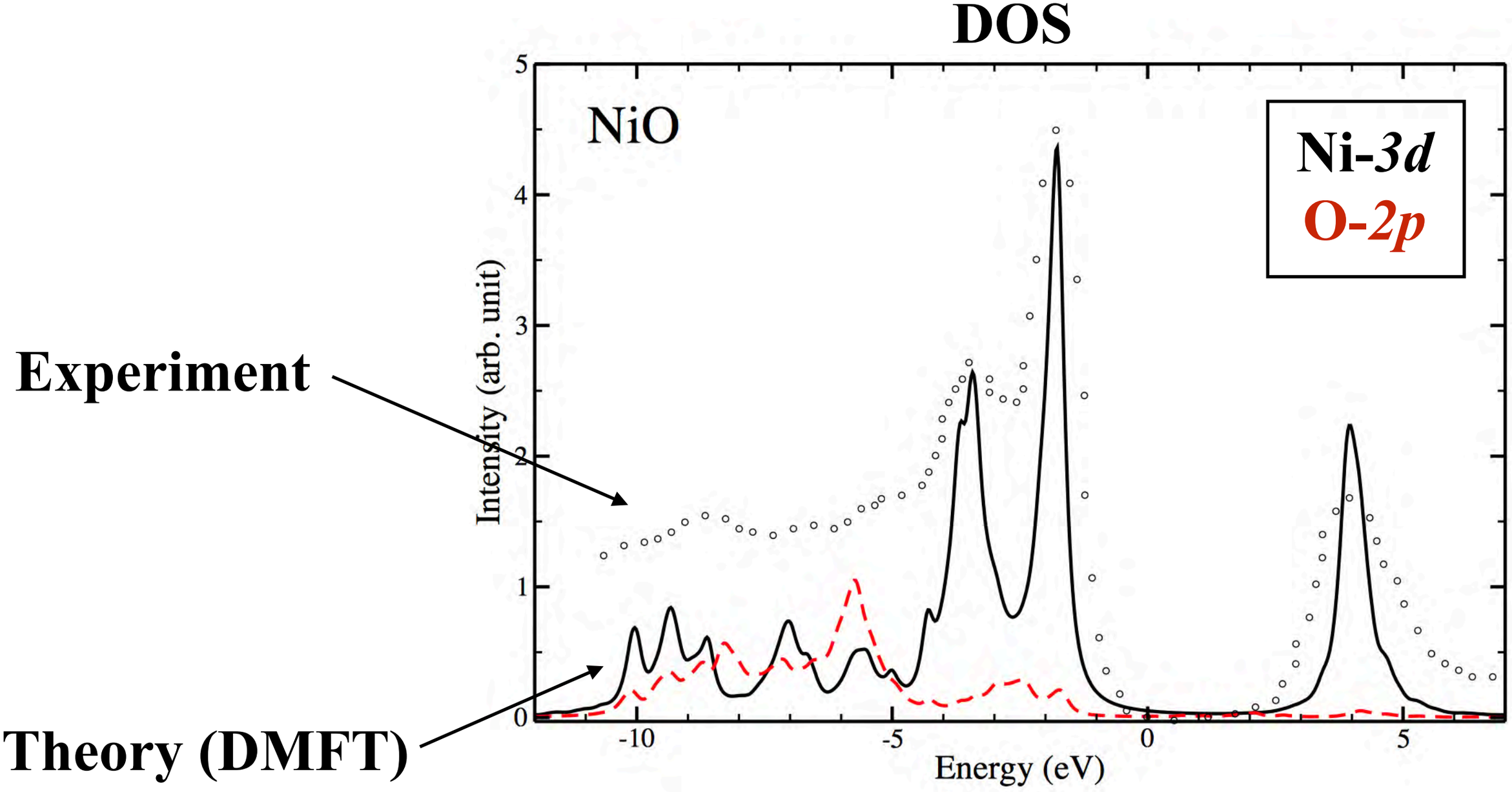
Valence band of Mn-doped GaAs



Paramagnetic NiO



P. Thunström *et al.* PRL **109** 186401 (2012)

A two-step approach to model excitations

1.
DFT calculation and extraction of the exchange parameters

$$\hat{H} = -\sum_{i \neq j} J_{ij} \cdot (\vec{e}_i \cdot \vec{e}_j)$$

2.
Solution of a parameterised Heisenberg Model.

☑ Finite-temperature magnetism.
☑ Simulations for the magnon spectra, Curie Temperature

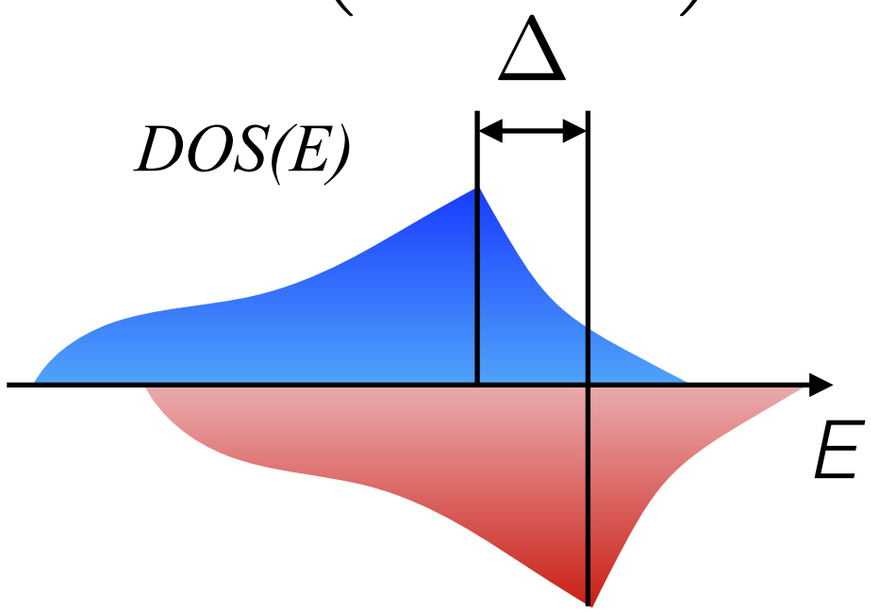
Ab initio exchange parameters from DFT

Lichtenstein *et al* JMMM **67** 65 (1987)

$$J_{ij} = \frac{-1}{4\pi} \int_{-\infty}^{E_f} d\varepsilon \; \mathrm{Tr}_L \left[\Delta_i \cdot G_{ij}^{\uparrow}(\varepsilon) \cdot \Delta_j \cdot G_{ji}^{\downarrow}(\varepsilon) \right]$$

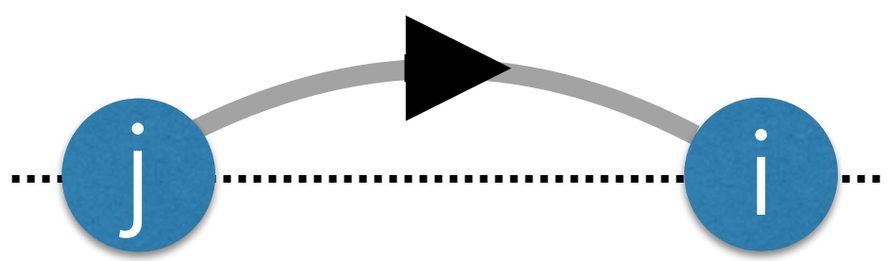
Local exchange splitting

$$\Delta_i = \left(\hat{H}_i^{\uparrow} - \hat{H}_i^{\downarrow} \right)$$



Inter-site Green's function

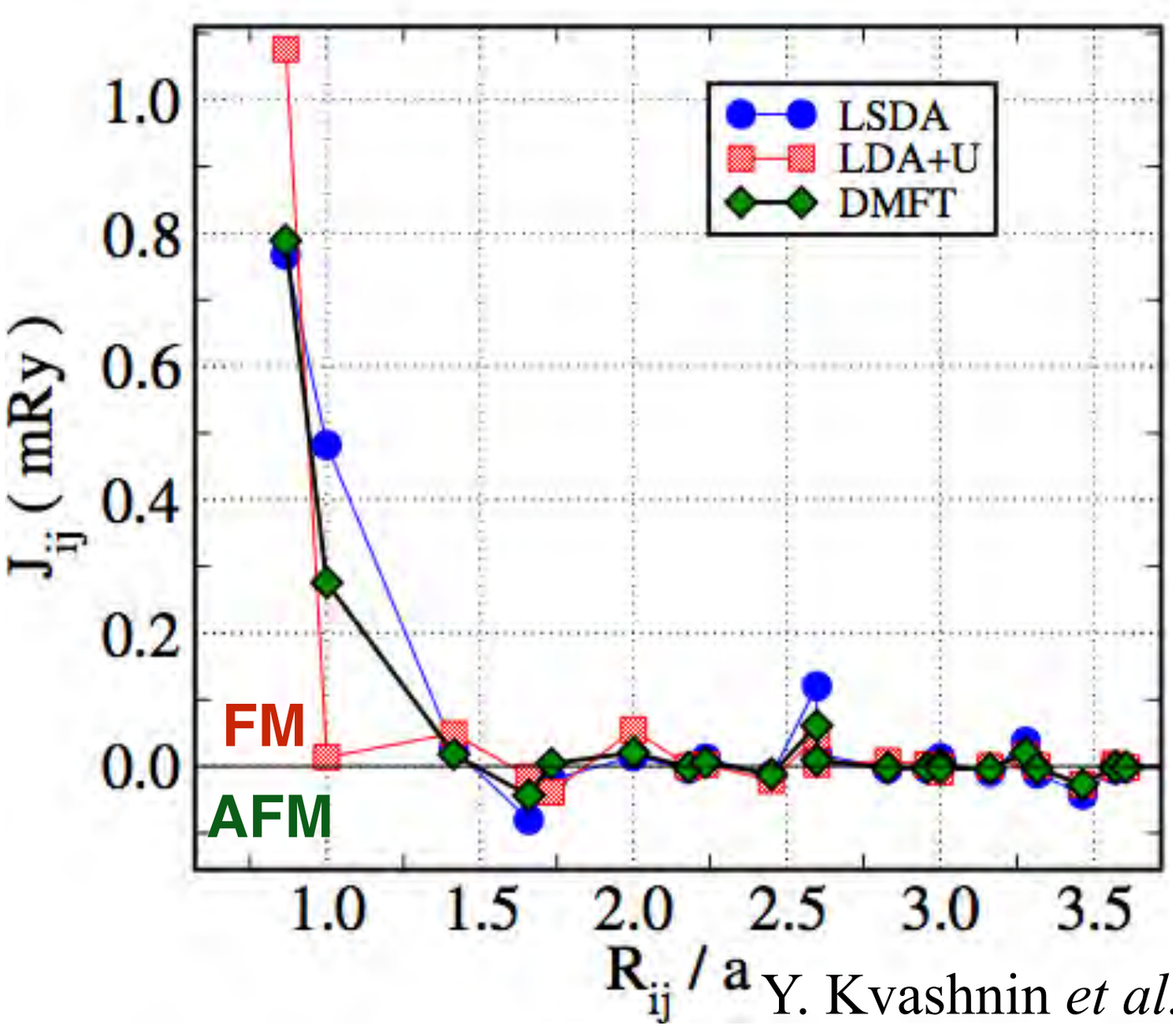
$$G_{ij}^{\sigma}(z) = \left\langle i \middle| \hat{G}(z) \middle| j \right\rangle = \left\langle i \middle| \frac{1}{z - \hat{H}^{\sigma}} \middle| j \right\rangle$$



bcc Fe: J_{ij}'s

LDA+U (Static Mean Field) $\quad \hat{\Sigma}(\vec{k}, i\omega_n) \rightarrow \hat{\Sigma}$

Dynamical Mean Field Theory (DMFT) $\quad \hat{\Sigma}(\vec{k}, i\omega_n) \rightarrow \hat{\Sigma}(i\omega_n)$



Hubbard U:
U=2.3 eV
J=0.9 *eV*

Y. Kvashnin *et al.* PRB 91 125133 (2015)

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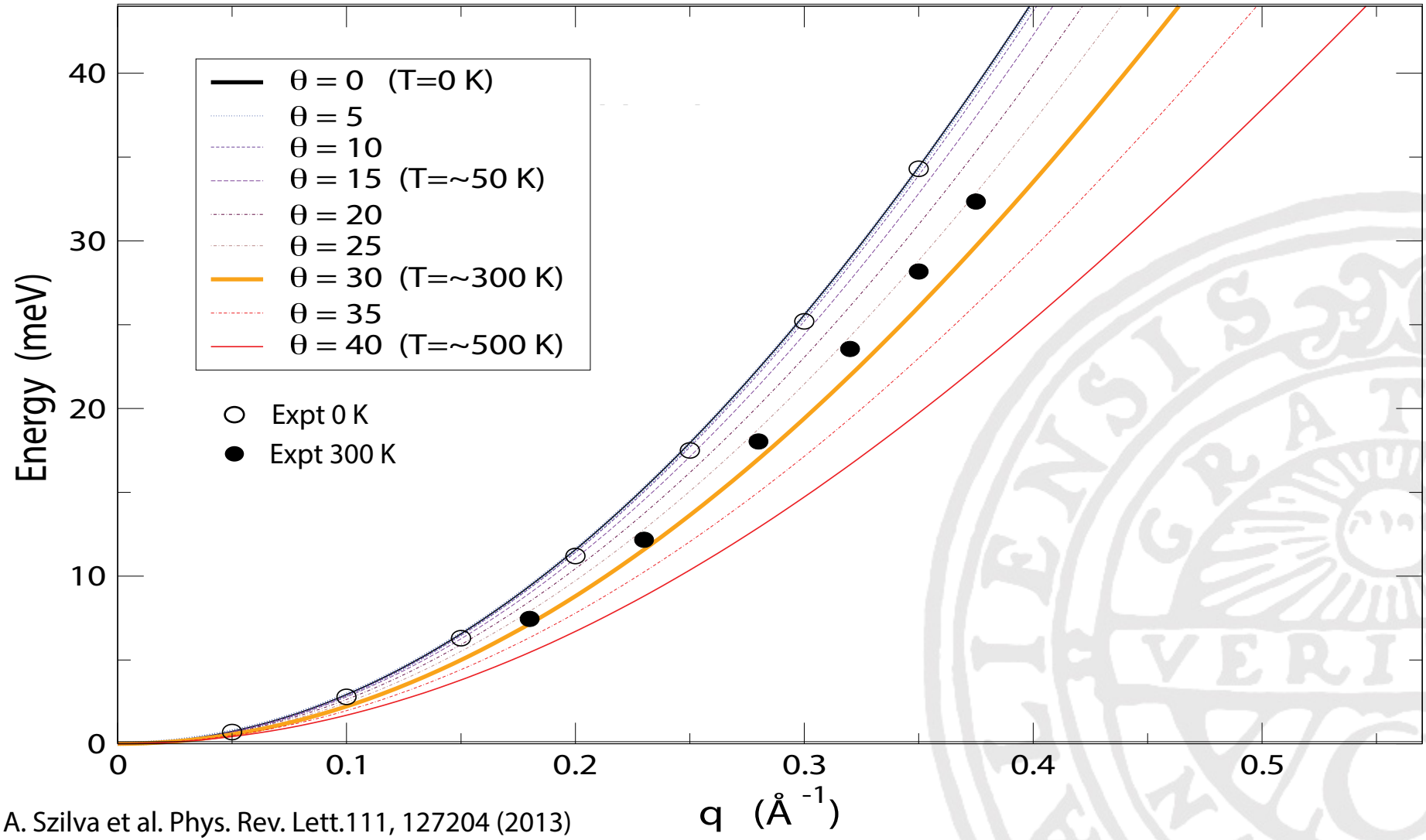
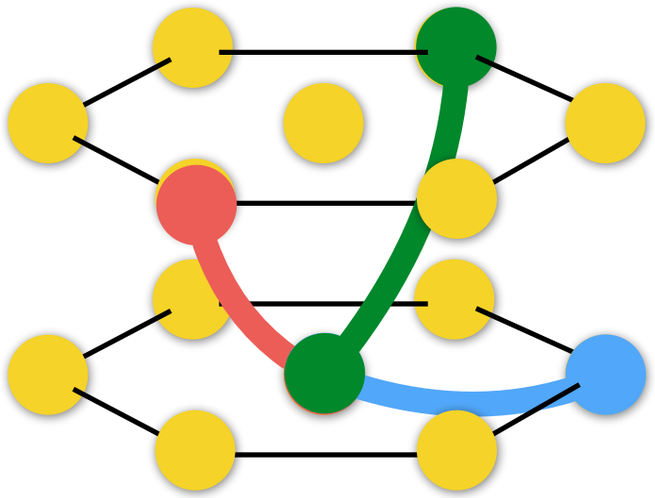
Finite temperature exchange interactions

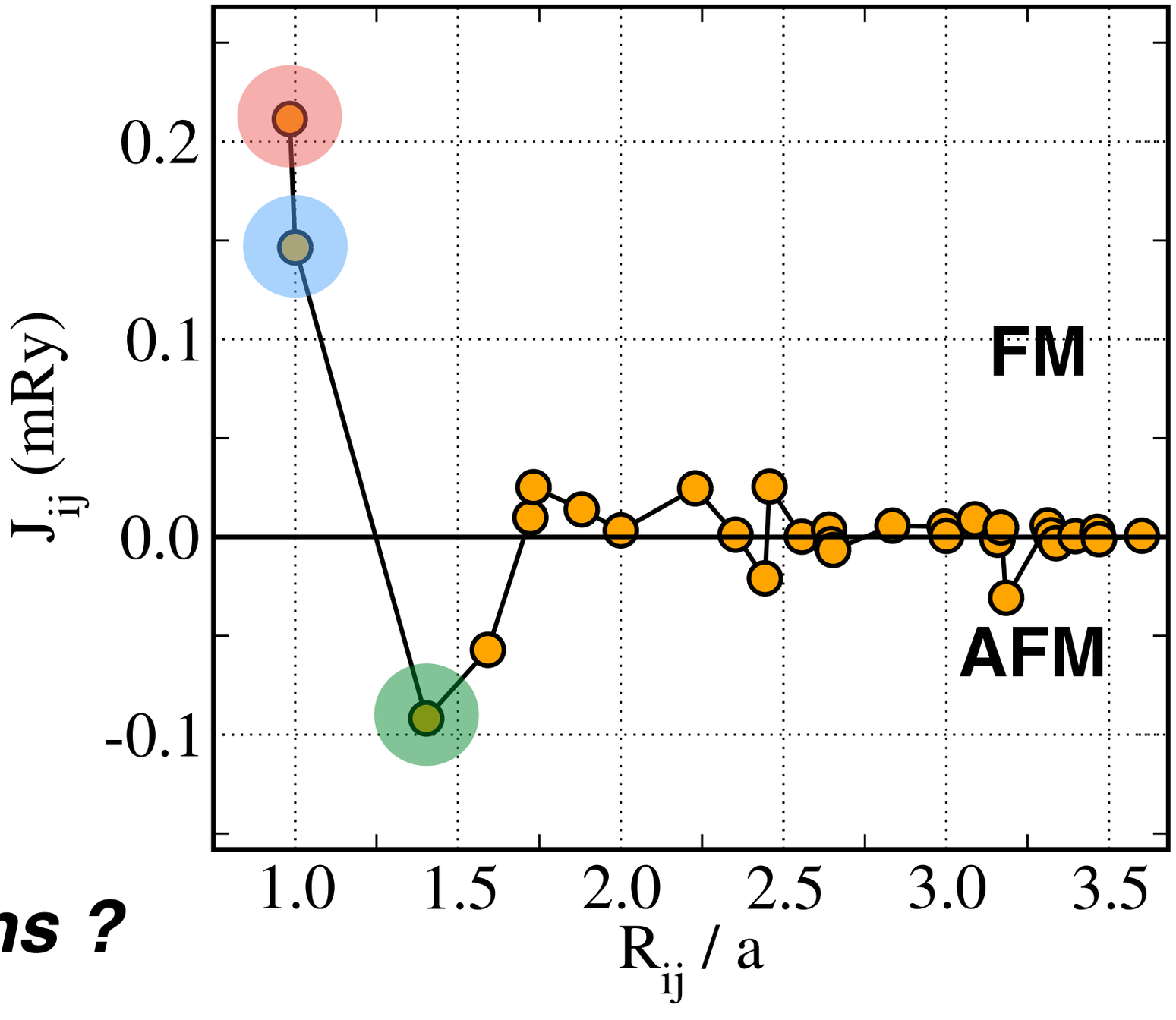


A. Szilva et al. Phys. Rev. Lett.111, 127204 (2013)

hcp Gd

Ferromagnetic metal (T_c=292 K)

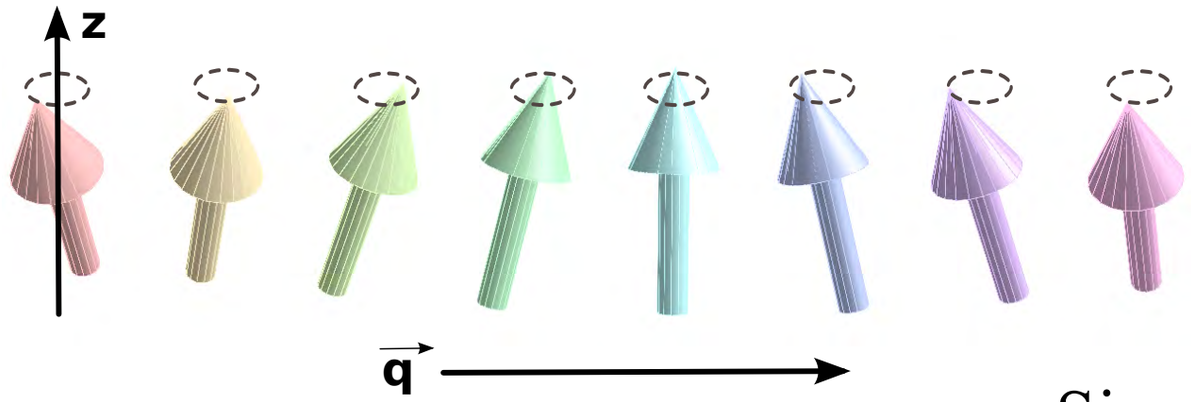




2nd step : magnons ?

hcp Gd: magnons

Results: Atomistic spin dynamics (**UppASD** software)

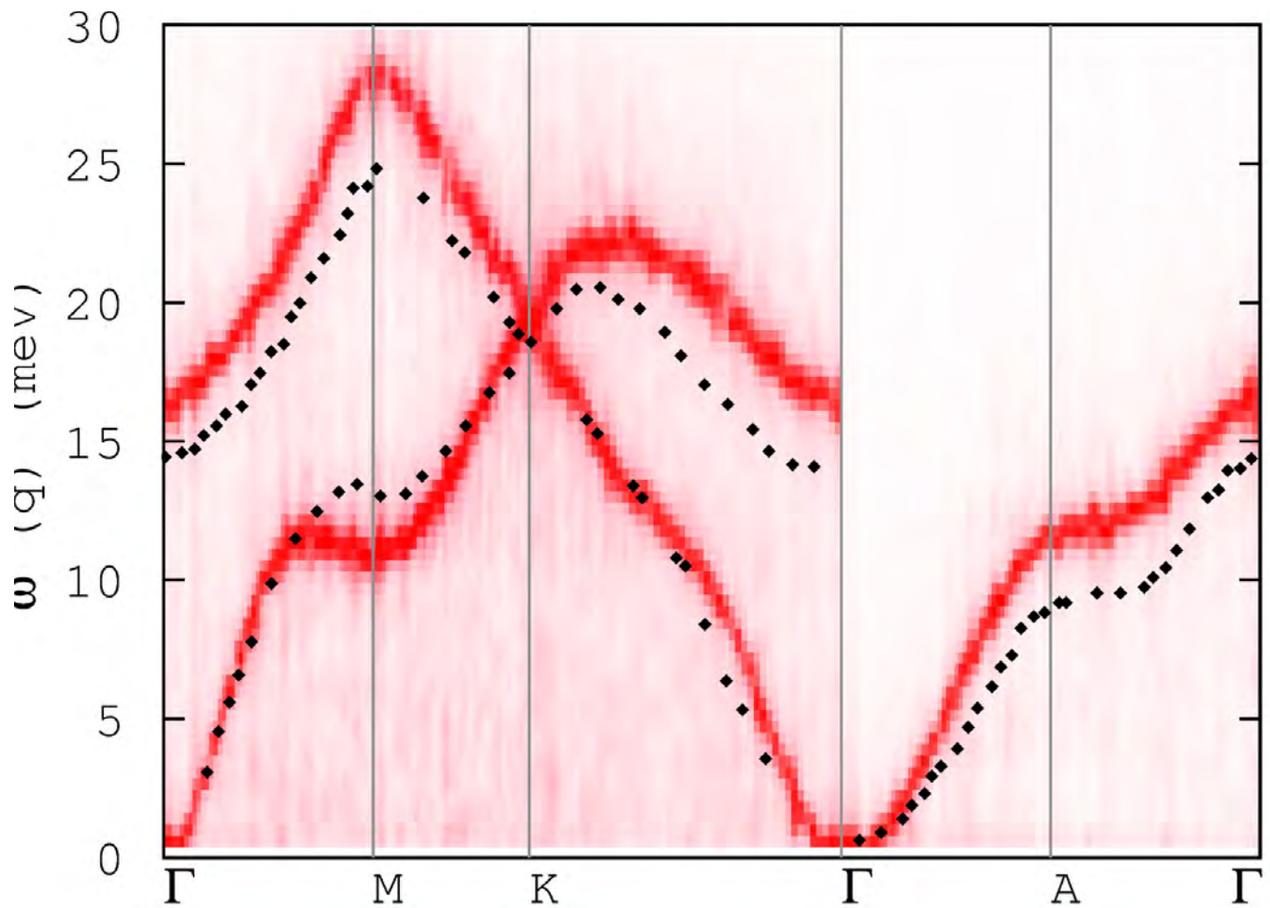


Solving the equation of motion:

$$\frac{d\vec{S}_j}{dt} = \frac{1}{\hbar}\left[\frac{\partial \hat{H}}{\partial \vec{S}_j} \times \vec{S}_j\right]$$

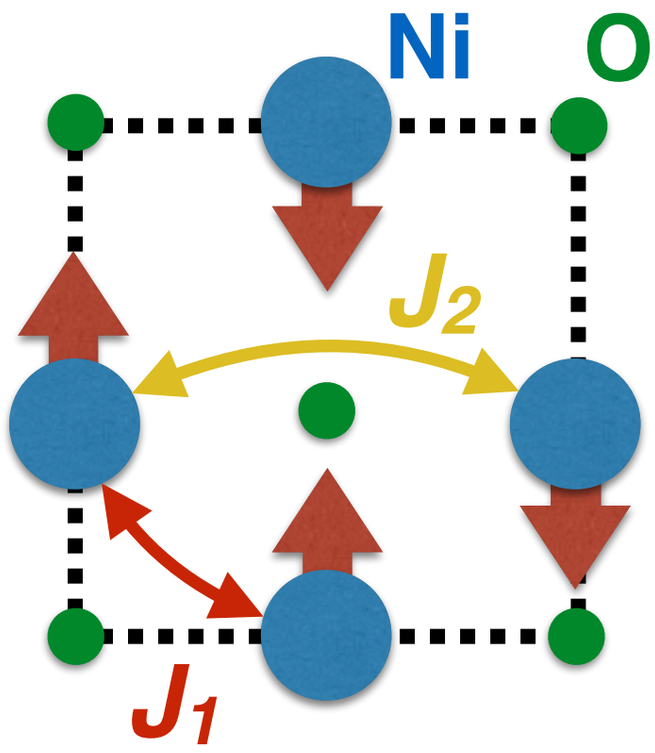
contains J_{ij}'s

Simulated magnon spectrum at T=78 K



Theory: Y.K. & D. Rodriguez
Exp: Koehler *et al.* PRL **24** 16 (1970)

Magnetic properties of NiO



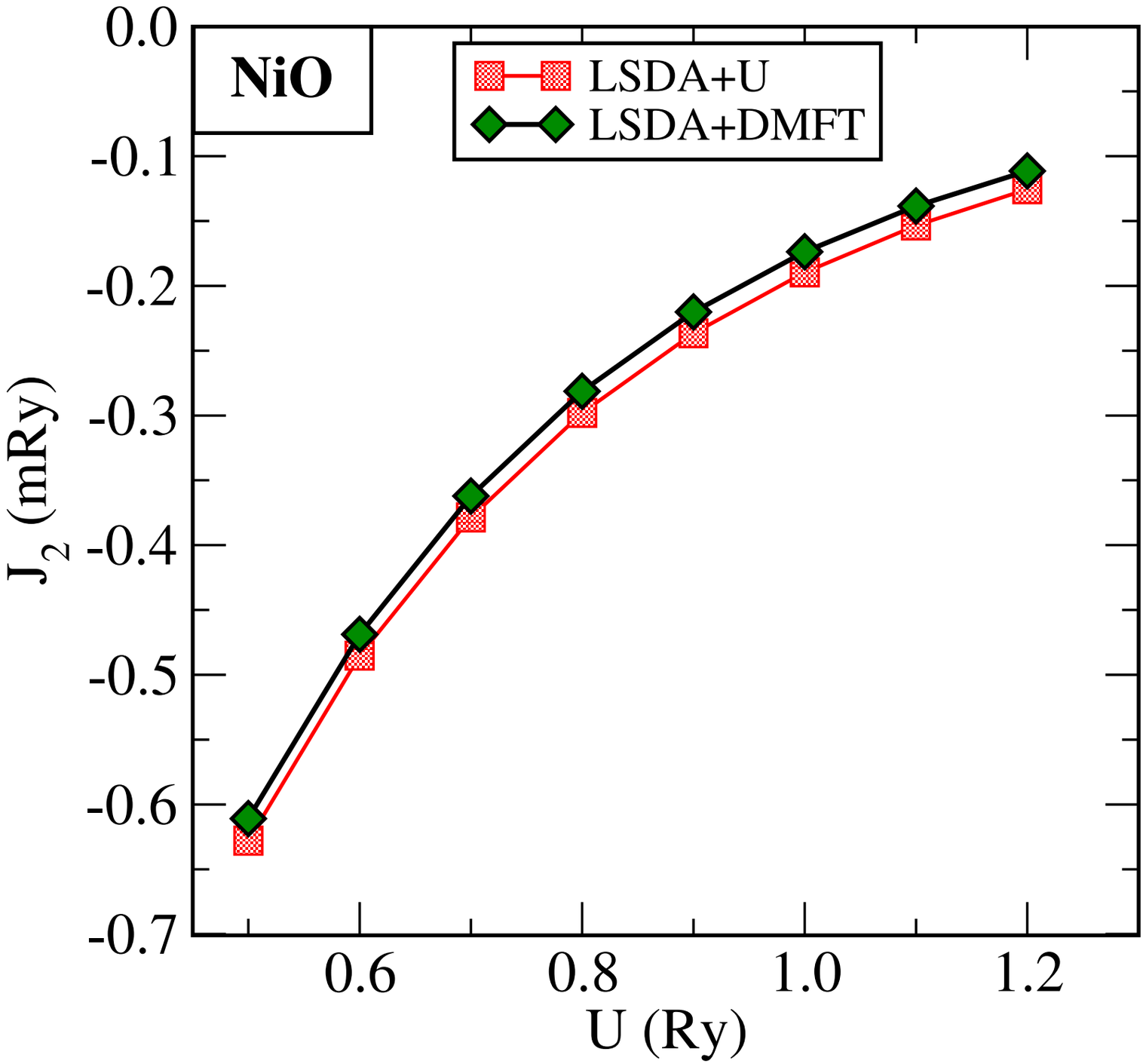
PRB 91 125133 (2015)

Computational setup	J_1	J_2
LSDA	0.04	−1.58
LSDA + DMFT	−0.003	−0.48
LSDA + U	−0.002	−0.50
LSDA + U ($U = 8$ eV) (Ref. [1])	0.004/0.0	−0.53
Exp. 1 (Ref. [2])	−0.051	−0.637
Exp. 2 (Ref. [3])	0.051	−0.67

LDA+U already provides a good description of the J_{ij}'s

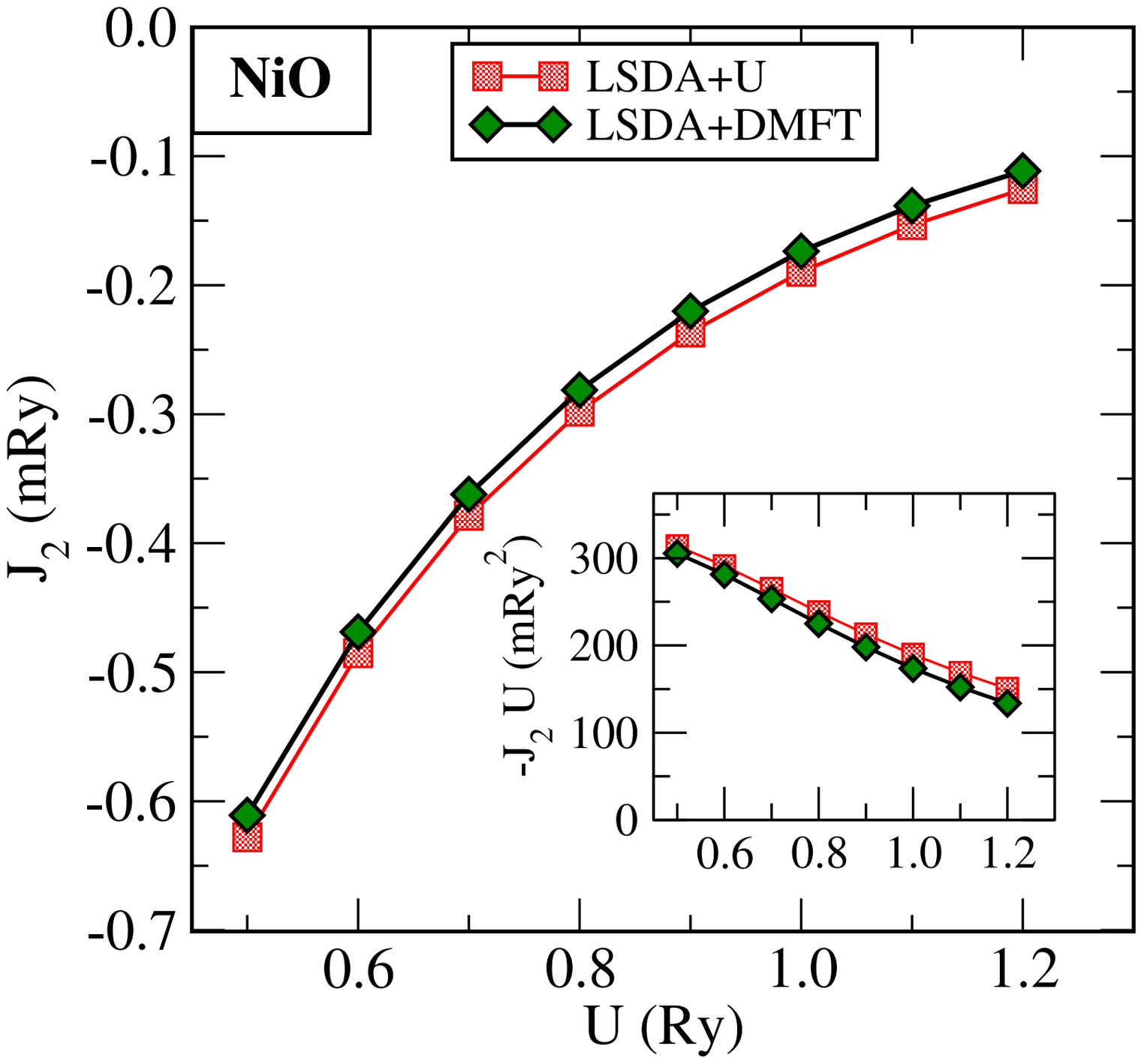
[1] Jacobsson PRB 88 134427 (2013)
[2] Hutchings PRB 6 3447 (1972)
[3] Shanker PRB 7 5000 (1973)

NiO: J_2 as a function of U



Does $\frac{t^2}{U}$ ***hold ?***

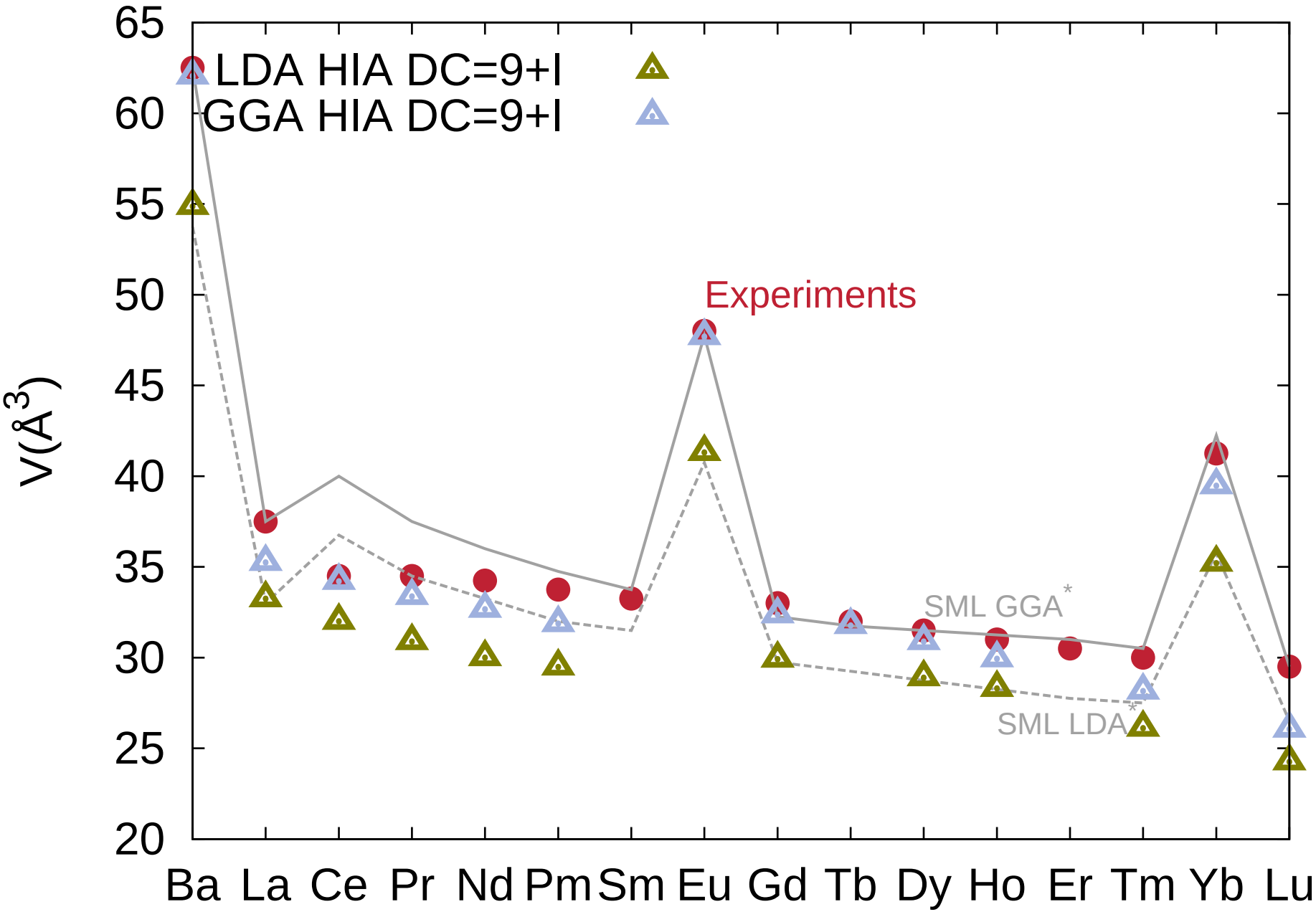
NiO: J_2 as a function of U

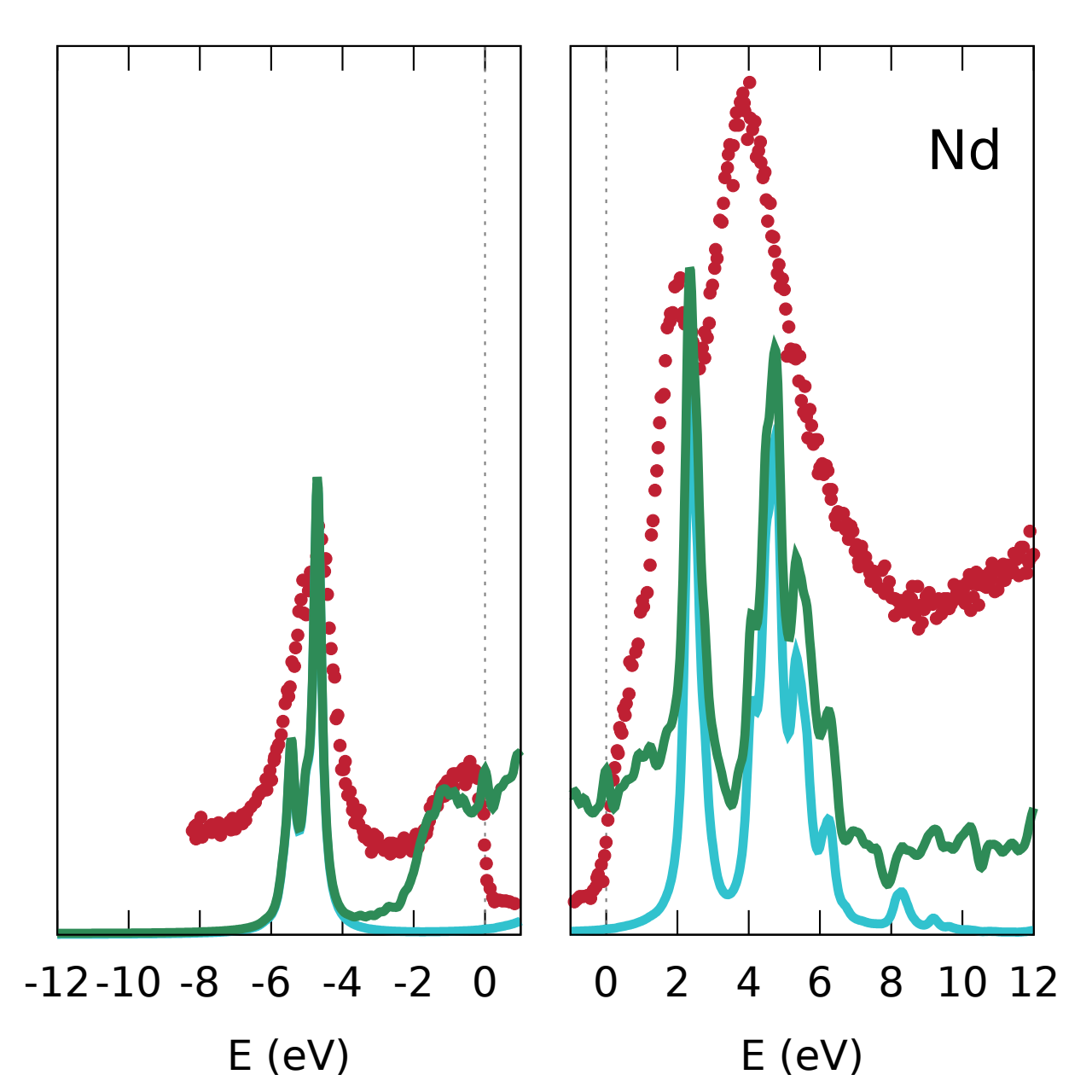


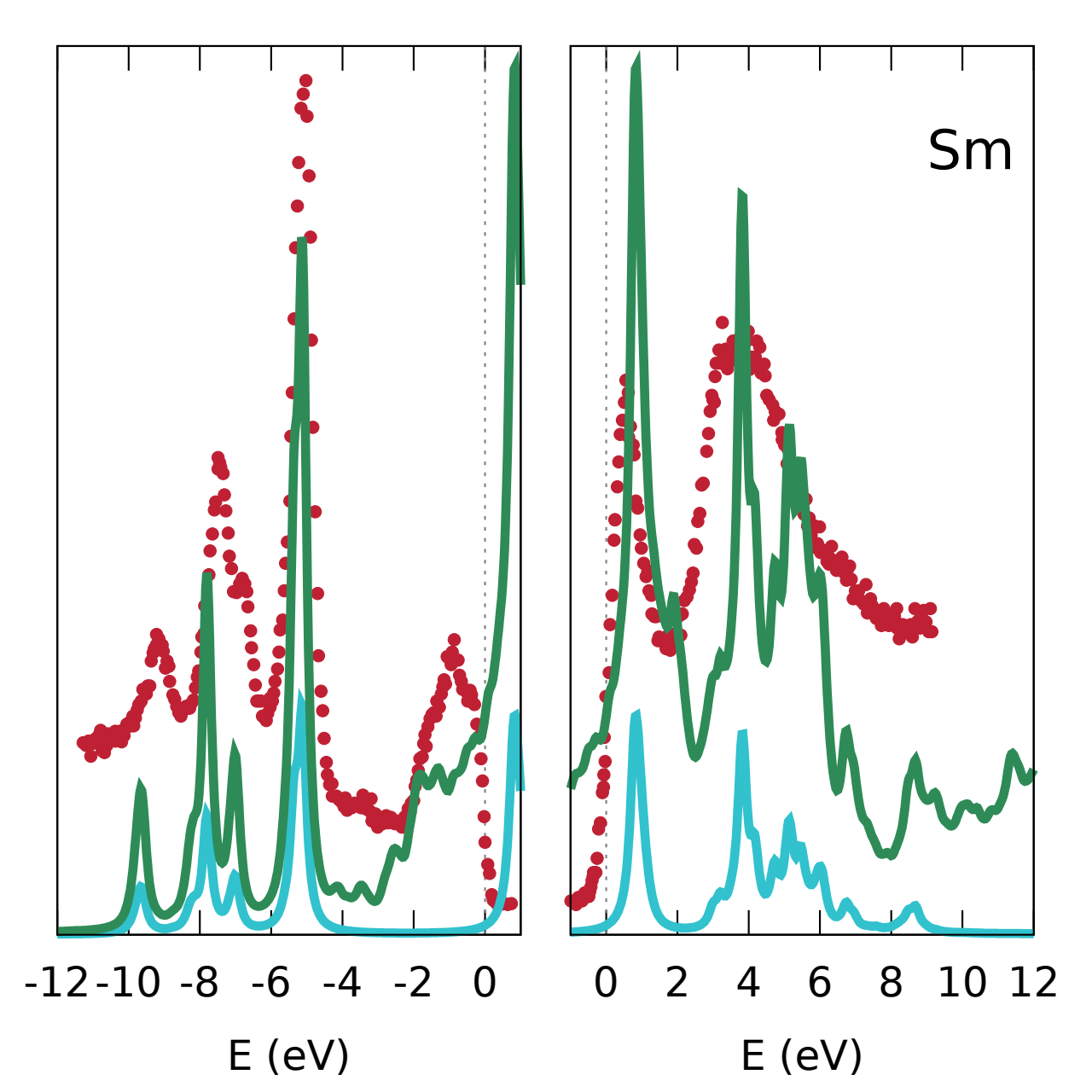
Does $\frac{t^2}{U}$ ***hold ?***

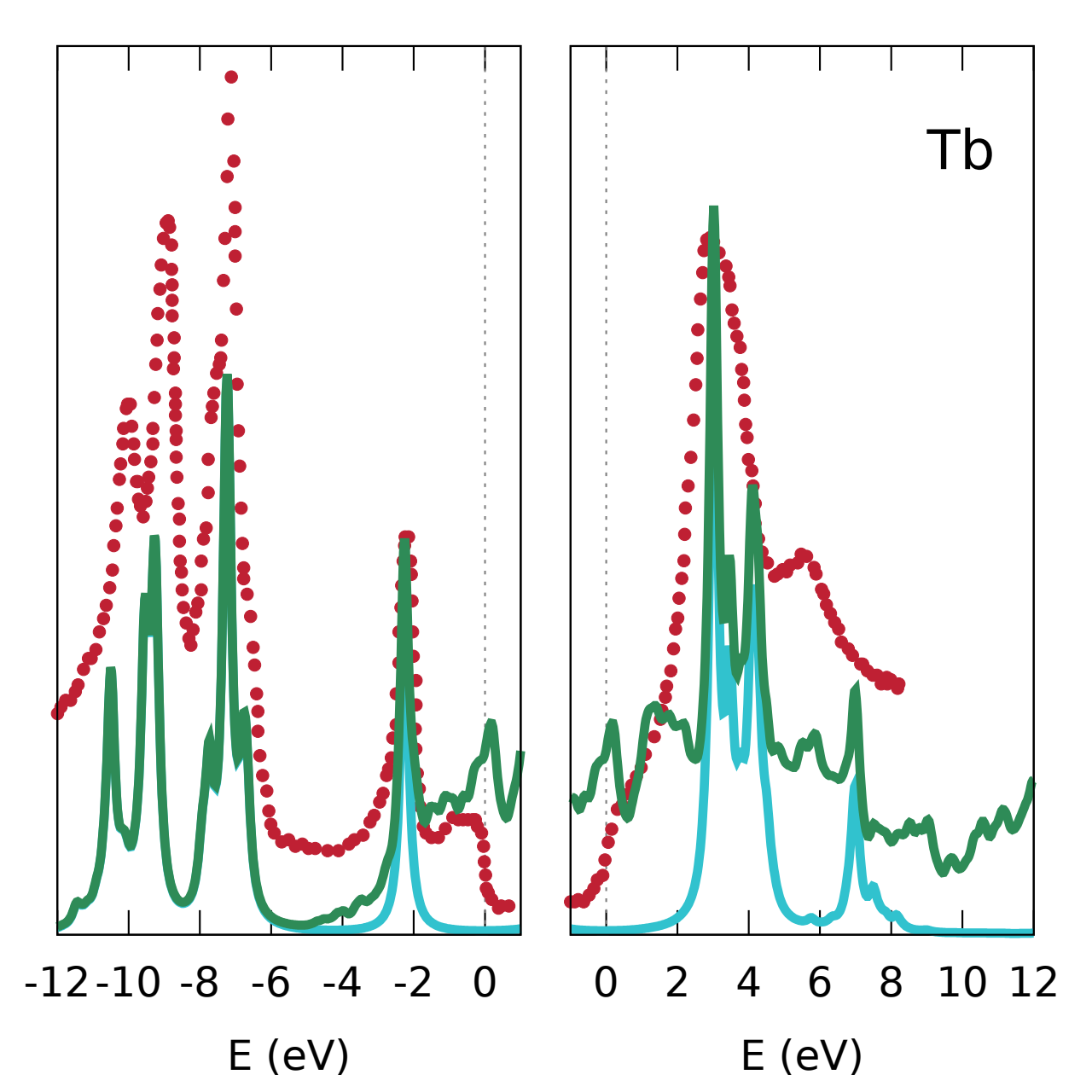
If yes: $J_2 U = t^2$

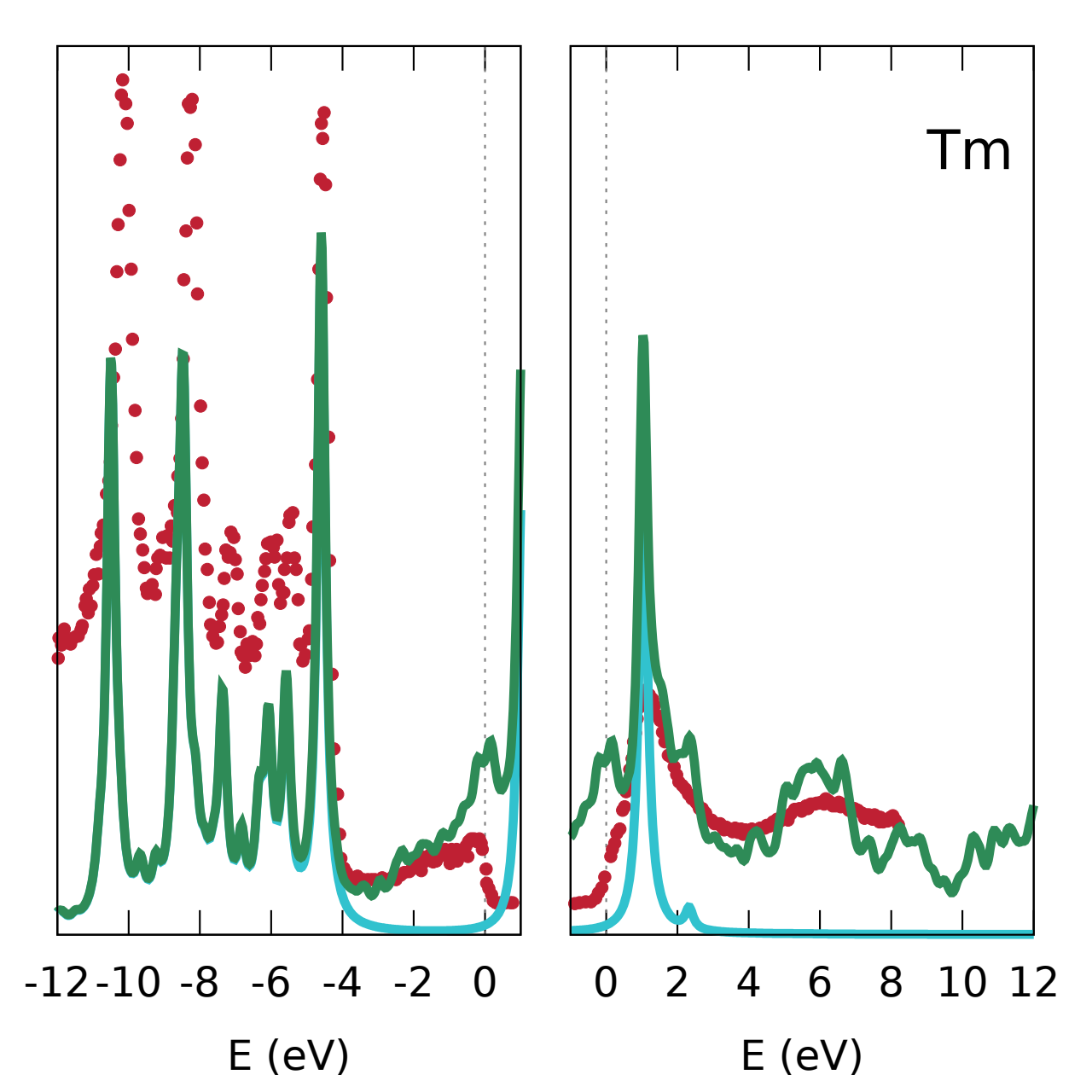
Hopping changes too!

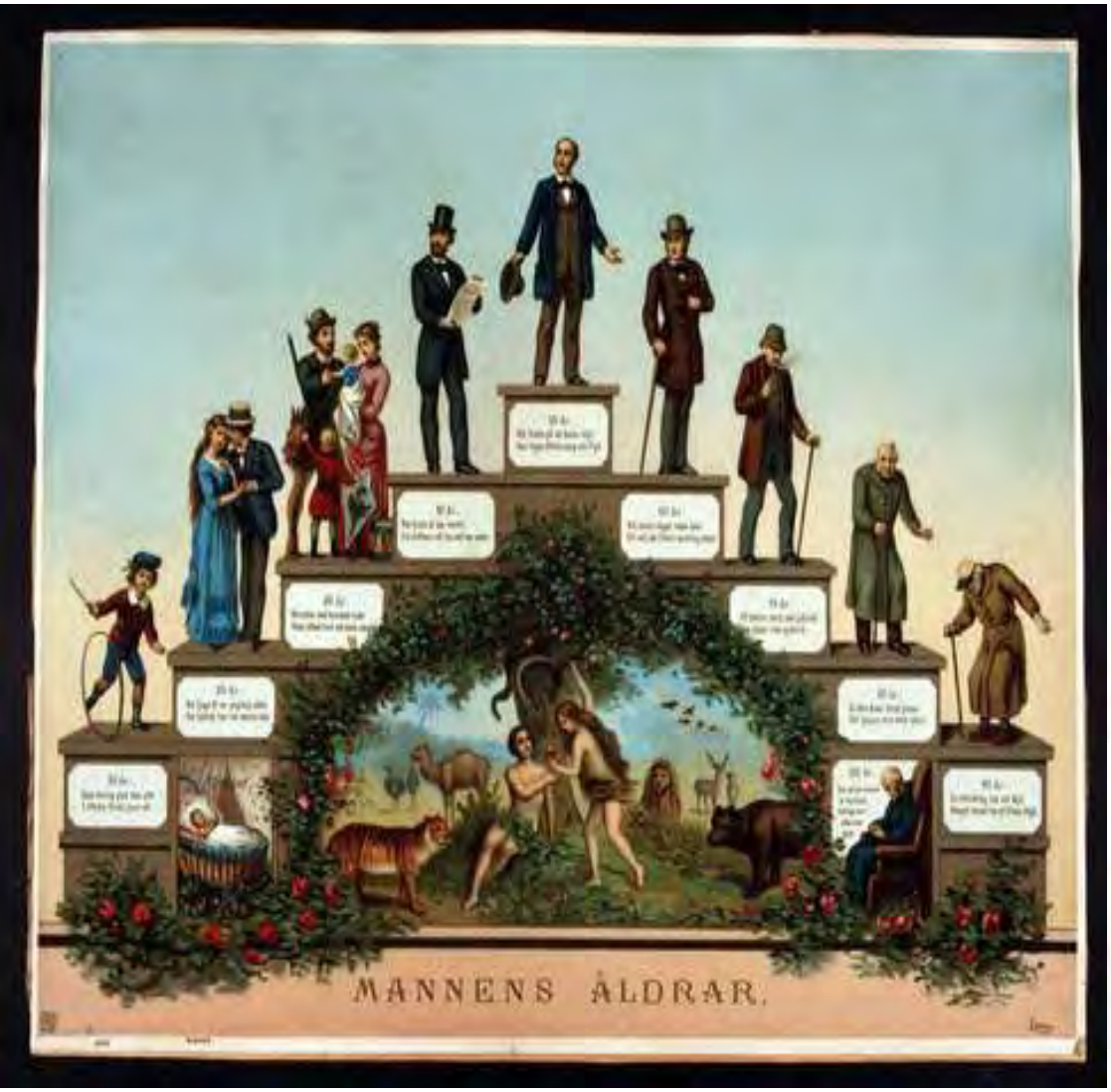

LDA HIA DC=9+I
GGA HIA DC=9+I
Experiments
SML GGA*
SML LDA*
V(Å3)
65
60
55
50
45
40
35
30
25
20
Ba La Ce Pr Nd Pm Sm Eu Gd Tb Dy Ho Er Tm Yb Lu


Nd
-12 -10 -8 -6 -4 -2 0
E (eV)
0 2 4 6 8 10 12
E (eV)


Sm
-12
-10
-8
-6
-4
-2
0
E (eV)
0
2
4
6
8
10
12
E (eV)


Tb
-12
-10
-8
-6
-4
-2
0
0
2
4
6
8
10
12
E (eV)
E (eV)


Tm
-12
-10
-8
-6
-4
-2
0
E (eV)
0
2
4
6
8
10
12
E (eV)



Mannens olika åldrar

MELLAN 15-20 ÅR ÄR MANNEN SOM EN FIAT, LITEN MEN ETTRIG.

MELLAN 20-30 ÅR ÄR MANNEN SOM EN PORSCHE. SNABB OCH VÄLSMORD.

MELLAN 30-50 ÅR ÄR MANNEN SOM EN OPEL RECORD LOVAR MER ÄN DEN HÅLLER.

MELLAN 50-80 ÅR ÄR MANNEN SOM EN GAMMAL FORD! MAN MÅSTE HÄLLA I HONOM LITE SPRIT FÖR ATT FÅ HONOM ATT STARTA.

Impact of correlations on J's

Exchange parameters from **Dynamical Mean Field Theory (DMFT)**:

$$J_{ij} = \frac{\mathrm{T}}{4} \sum_n \mathrm{Tr} \left[\hat{\Delta}_i(i\omega_n) \hat{G}^{\uparrow}_{ij}(i\omega_n) \hat{\Delta}_j(i\omega_n) \hat{G}^{\downarrow}_{ji}(i\omega_n) \right]$$

Katsnelson & Lichtenstein PRB **61** 8906 (2000)

G contains **self-energy**:

$$G_{ij}(z) = \left\langle i \left| \frac{1}{z - \hat{H} - \hat{\Sigma}(z)} \right| j \right\rangle$$

Exchange field becomes dynamical!

$$\Delta(i\omega_n) = \left(\hat{H}_i^{\uparrow} - \hat{H}_i^{\downarrow} \right) + \left(\Sigma_i^{\uparrow}(i\omega_n) - \Sigma_i^{\downarrow}(i\omega_n) \right)$$