

Correlated electronic structures and spin-dynamics, some recent investigations

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Nordström, Pereiro, Peters, Rodrigues, 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



Electronic structure database

http://gurka.fysik.uu.se/ESP/

http://gurka.fysik.uu.se/ESP/ Google

wiki saob eniro seb SAS News (168) work sj ul stockholm ord torrent LK musik SSH

ESP					
Version	Complete list				
A word of caution	Search	Element	Methods	FAQ	Contact

Element:

Element:

Element:

Element:

Element:

Antal element:

<http://gurka.fysik.uu.se/ESP/>

1 H															2 He		
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
55 Cs	56 Ba	*	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
87 Fr	88 Ra	**	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Uuo	111 Uuu	112 Uub	113 Uut	114 Uuq	115 Uup	116 Uuh	117 Uus	118 Uuo

Computational Materials
Science 44, 1042 (2009)

*	57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
**	89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr



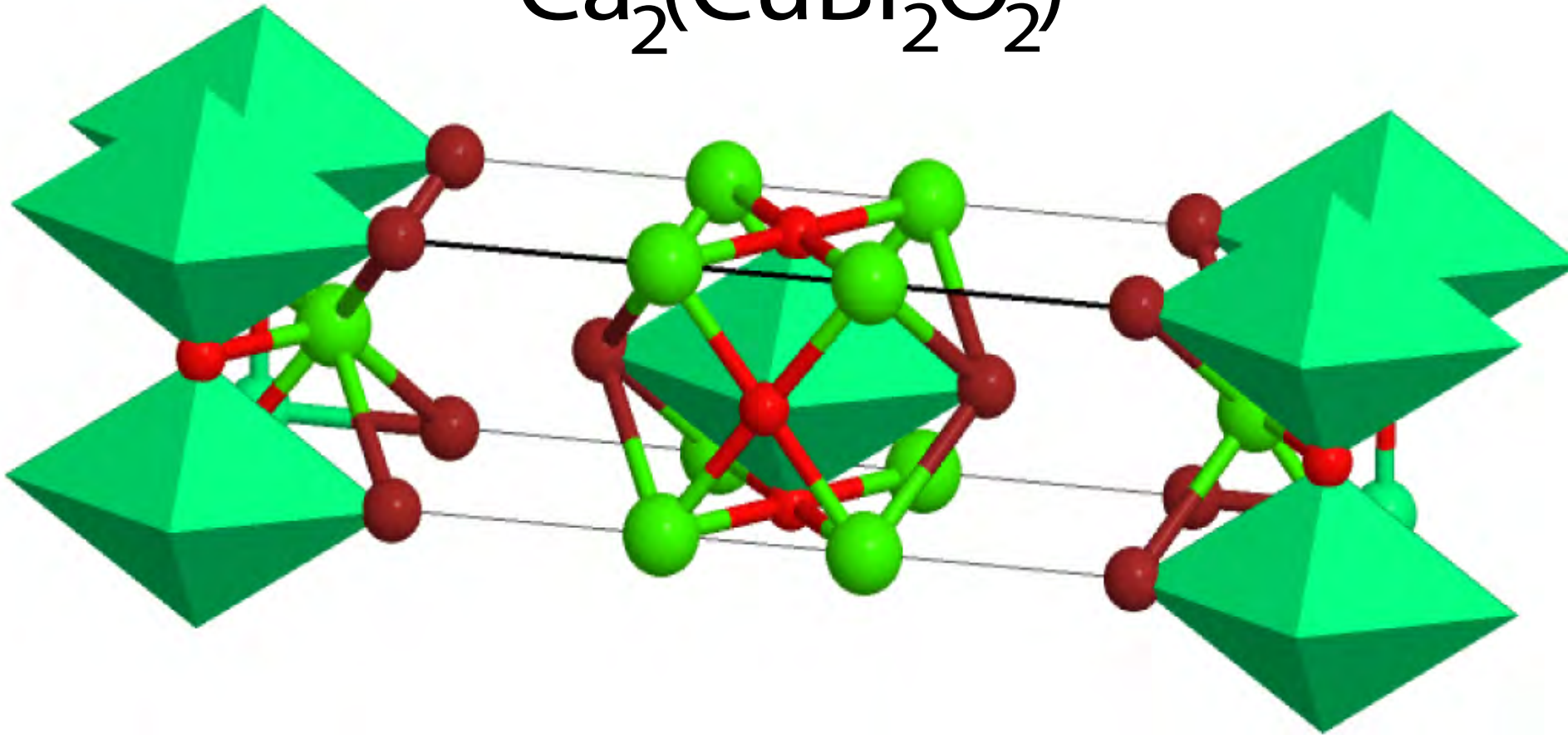
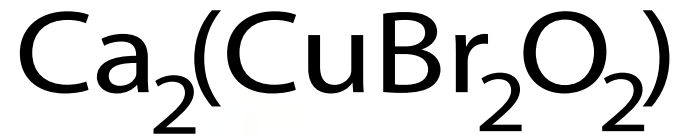
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Possible high temperature superconductors



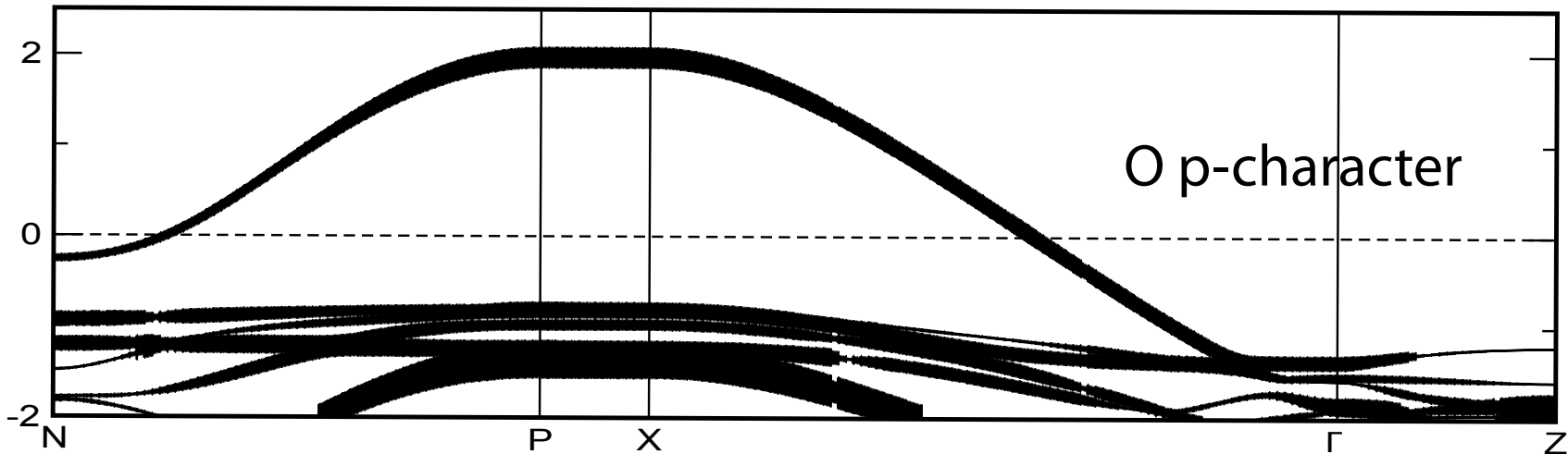
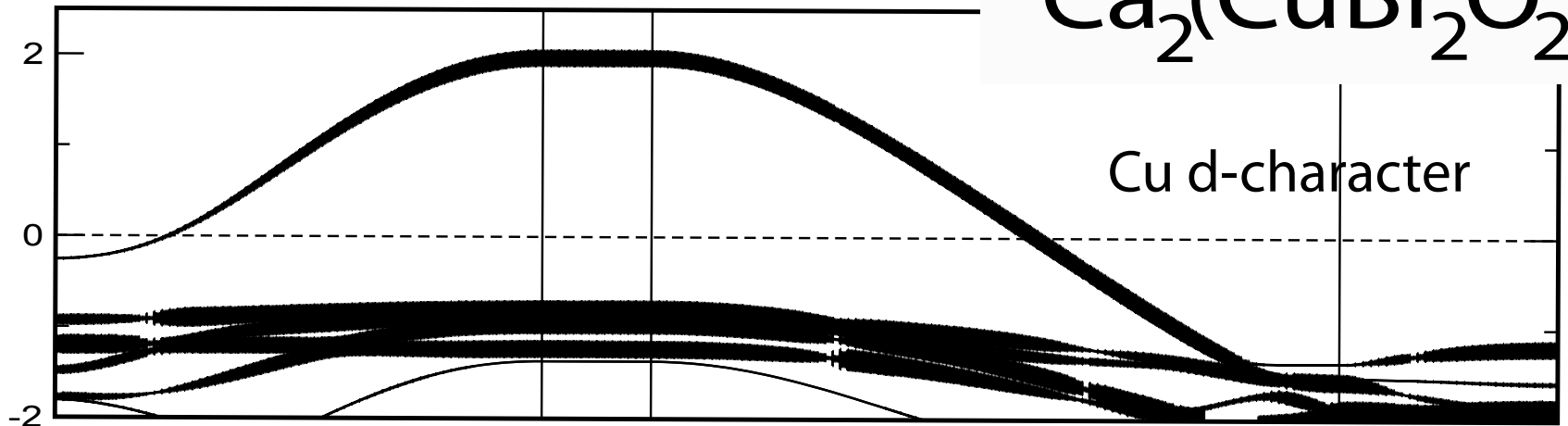


Data-mining criterion-structure





Data-mining criterion-bands/bonds

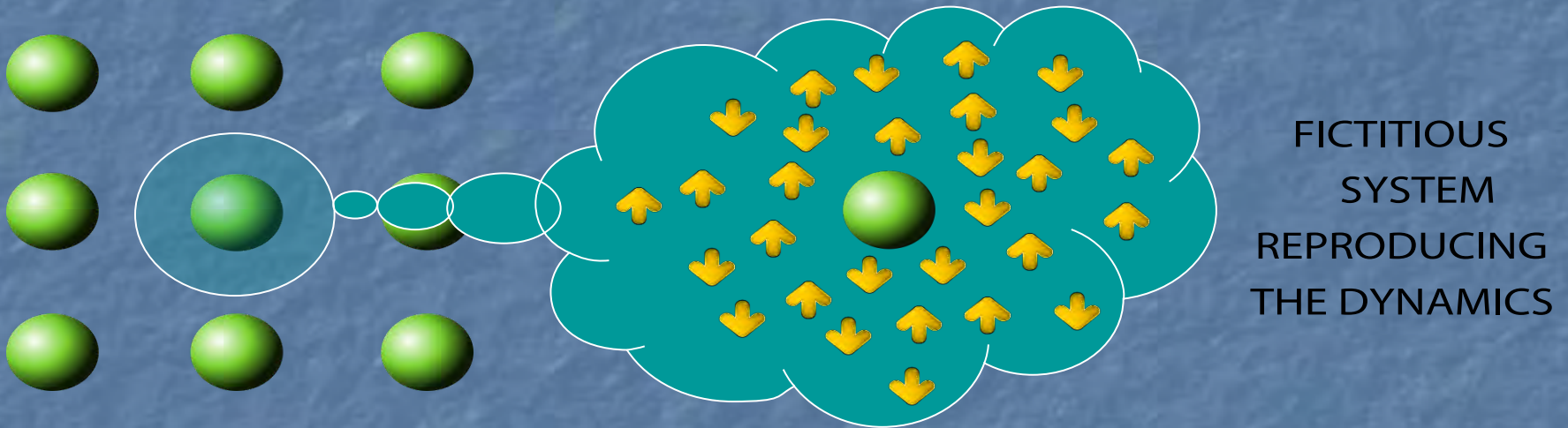




Dynamical mean field theory

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

The Hubbard model is mapped into an Anderson Impurity Model



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



Springer Series in Solid-State Sciences 167

John M. Wills
Mebarek Alouani
Per Andersson
Anna Delin
Olle Eriksson
Oleksiy Grechnev
Full-Potential Electronic Structure Method
Energy and Force Calculations with Density
Functional and Dynamical Mean Field Theory

This book covers the theory of electronic structure of materials, with special emphasis on the usage of linear muffin-tin orbitals. Methodological aspects are given in detail as are examples of the method when applied to various materials. Different exchange and correlation functionals are described and how they are implemented within the basis of linear muffin-tin orbitals. Functionals covered are the local spin density approximation, generalised gradient approximation, self-interaction correction and dynamical mean field theory.



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Wills · Alouani · Andersson
Delin · Eriksson · Grechnev

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1

Full-Potential Electronic
Structure Method

Full-Potential Electronic Structure Method

Energy and Force Calculations with Density
Functional and Dynamical Mean Field Theory

123

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

Correlated basis

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

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:



$$|\Psi_1^5\rangle = |1111100000\rangle,$$

$$|\Psi_2^5\rangle = |1111010000\rangle,$$

⋮

$$|\Psi_M^5\rangle = |0000011111\rangle.$$

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:

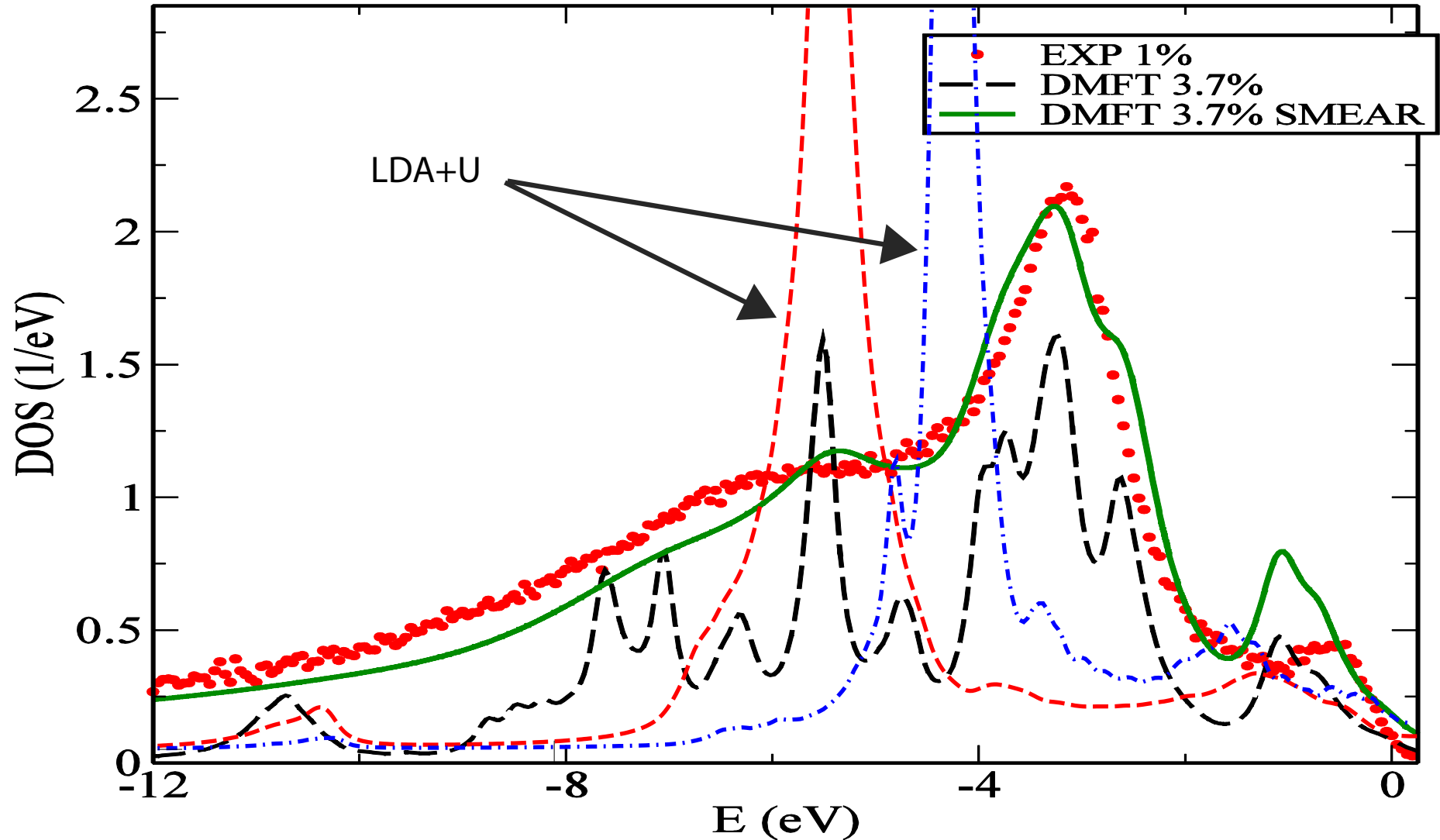


Once the many-body states have been determined, the one-particle Green's function can be obtained through the Lehmann representation

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

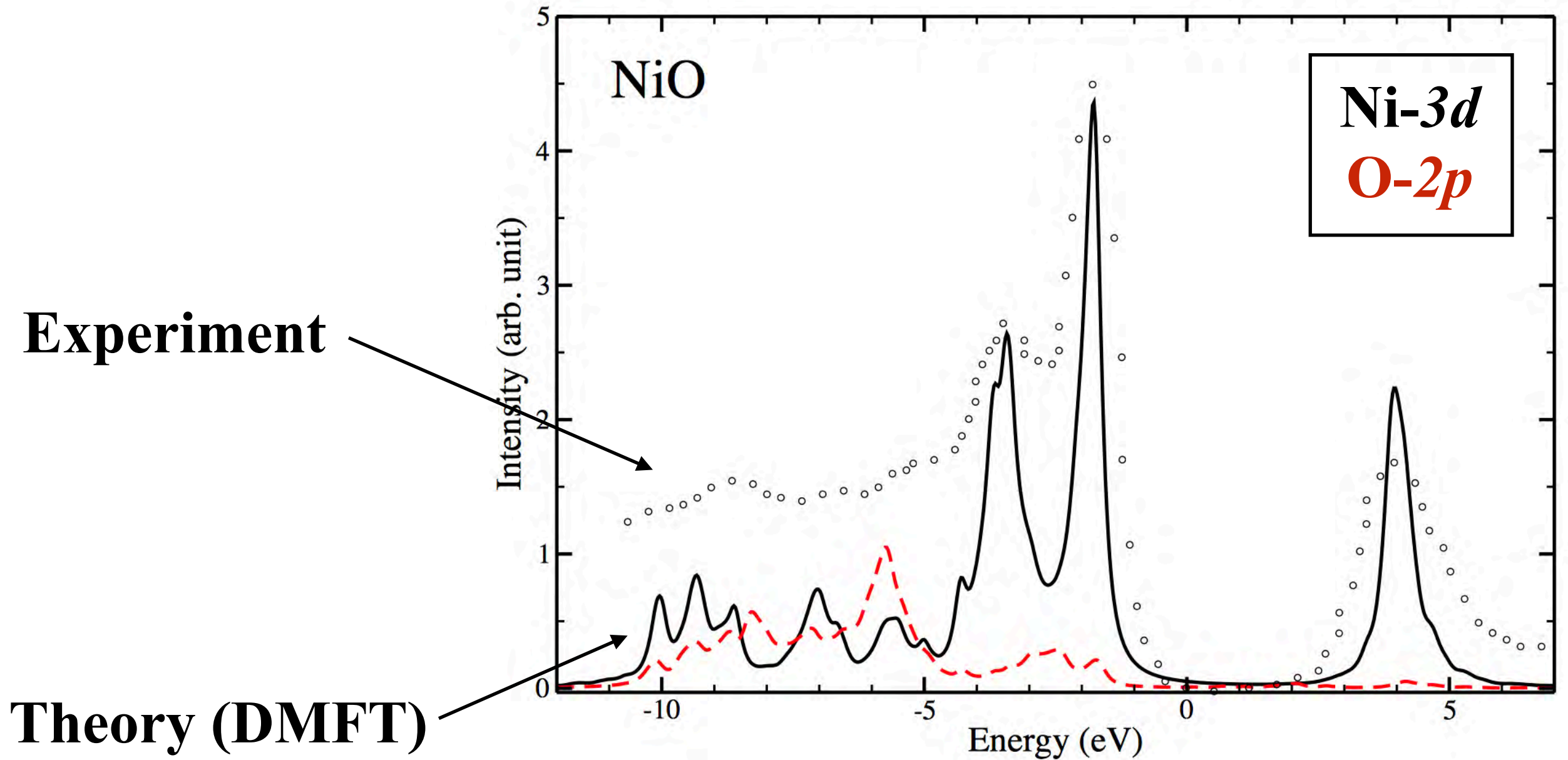


Valence band of Mn-doped GaAs



Paramagnetic NiO

DOS



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

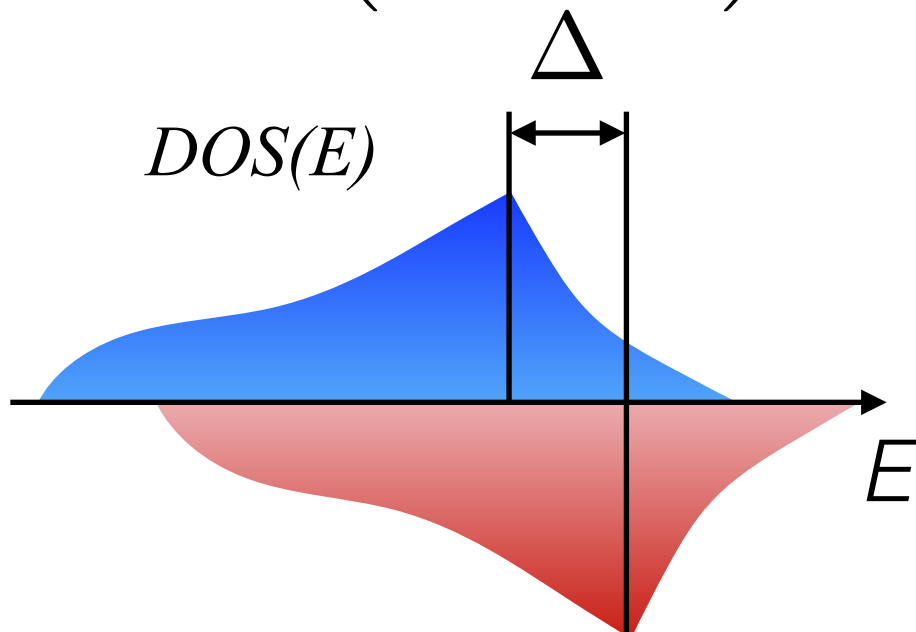
Ab initio exchange parameters from DFT

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

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

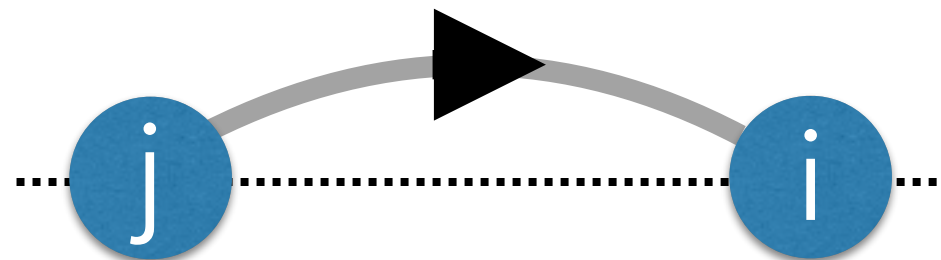
Local exchange splitting

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



Inter-site Green's function

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



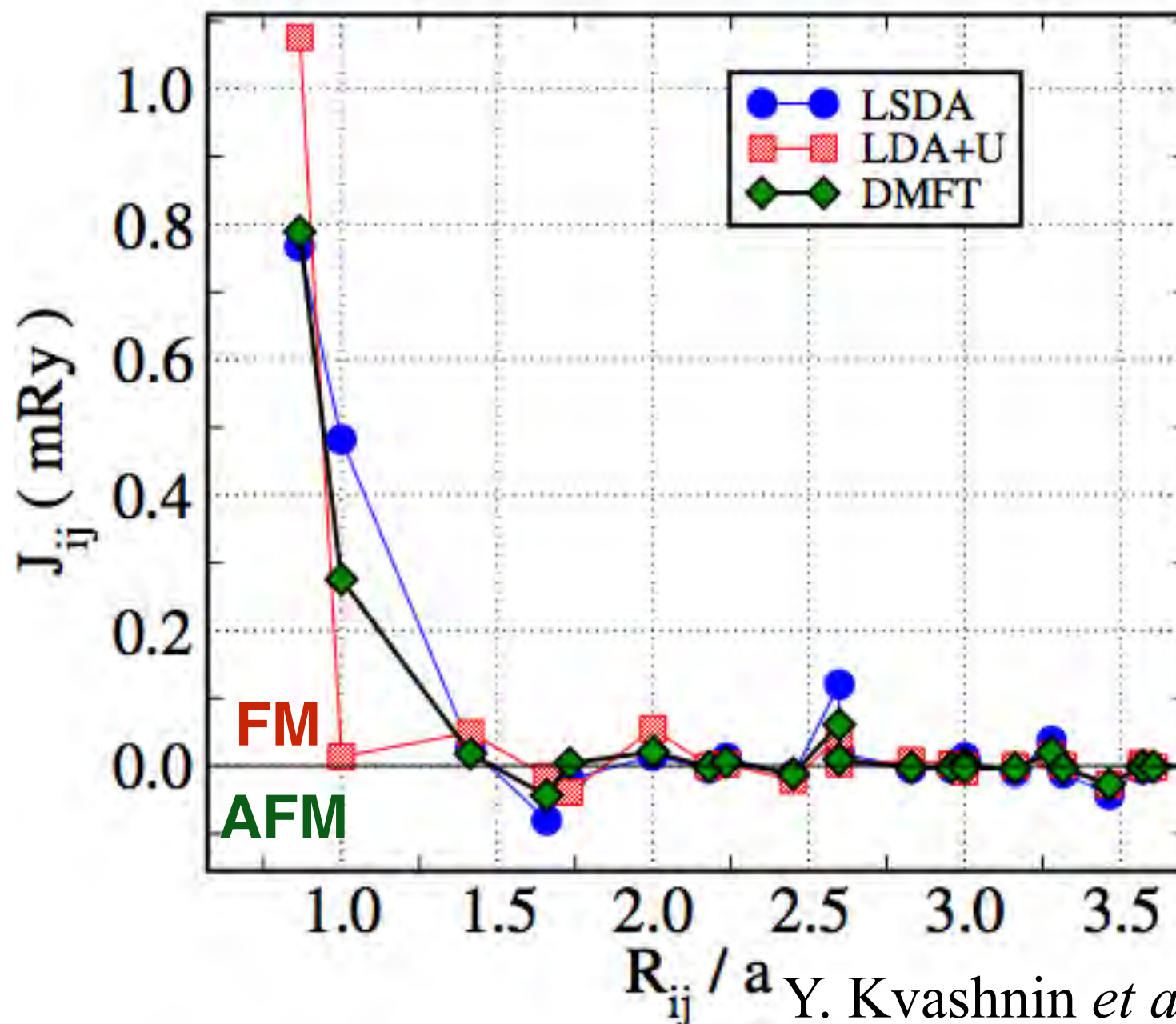
bcc Fe: J_{ij} 's

LDA+ U (Static Mean Field)

$$\hat{\Sigma}(\vec{k}, i\omega_n) \rightarrow \hat{\Sigma}$$

Dynamical Mean Field Theory (DMFT)

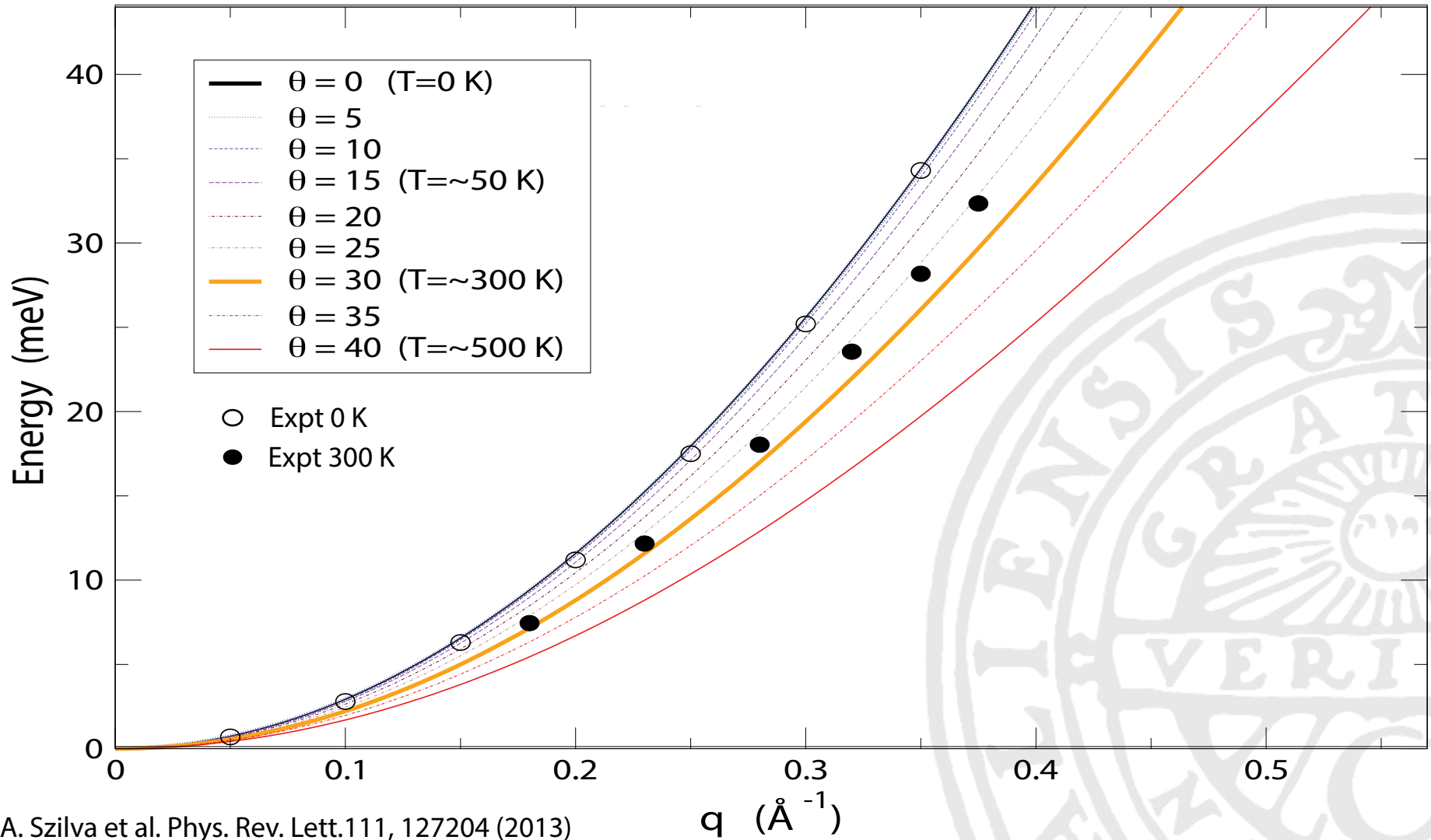
$$\hat{\Sigma}(\vec{k}, i\omega_n) \rightarrow \hat{\Sigma}(i\omega_n)$$



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

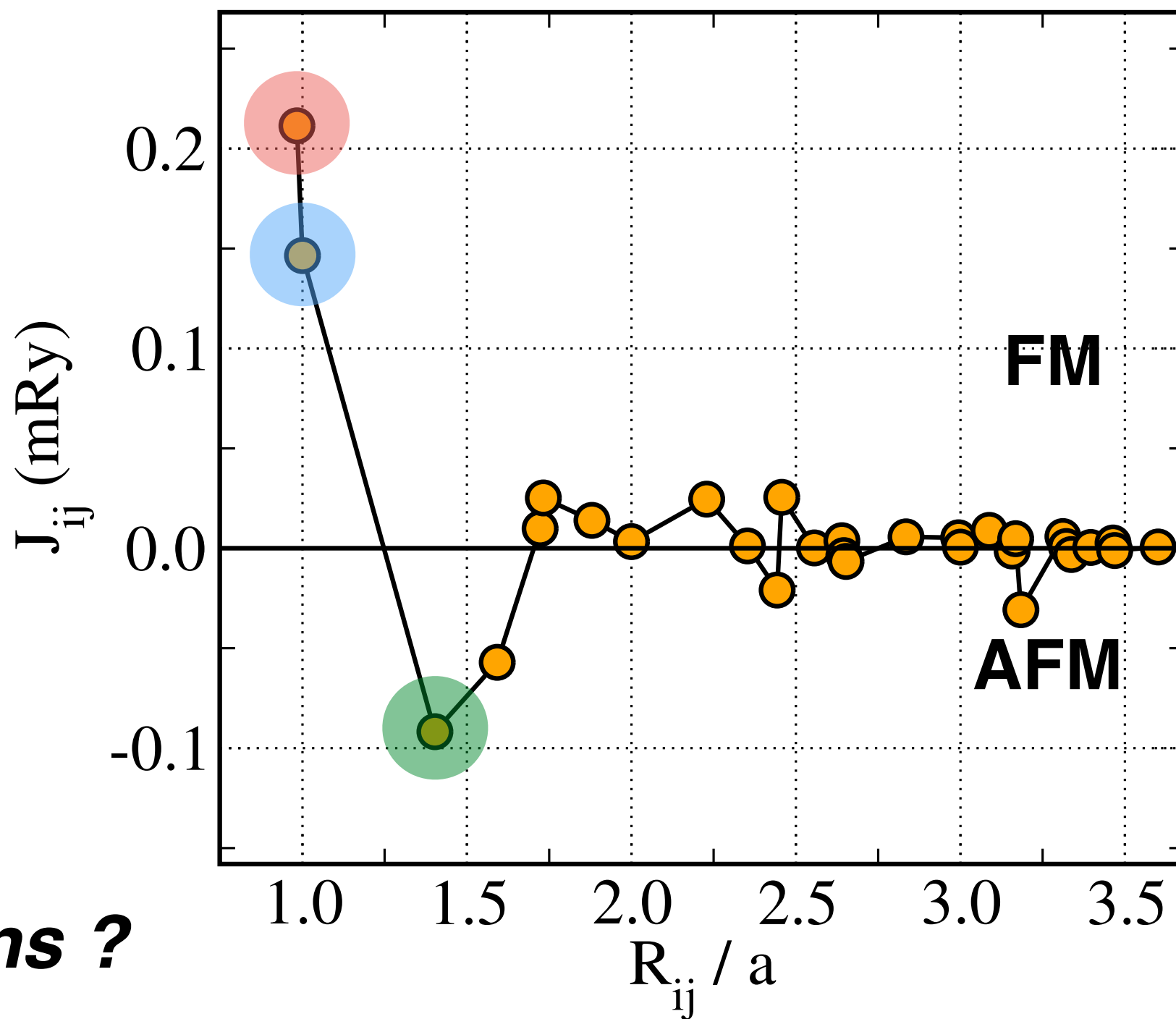
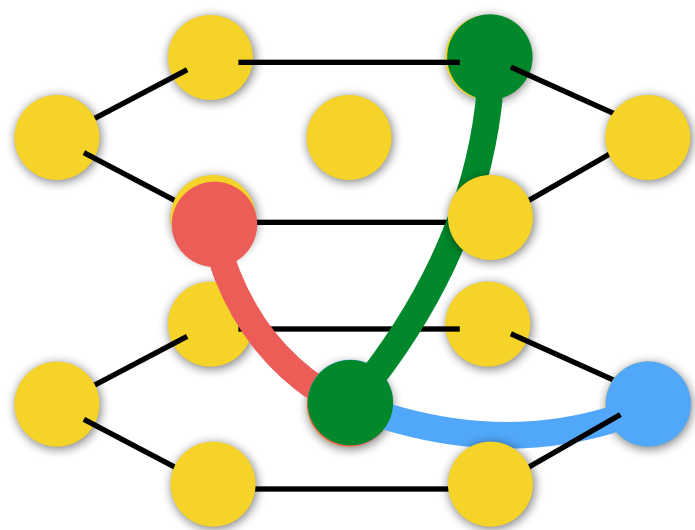


Finite temperature exchange interactions



hcp Gd

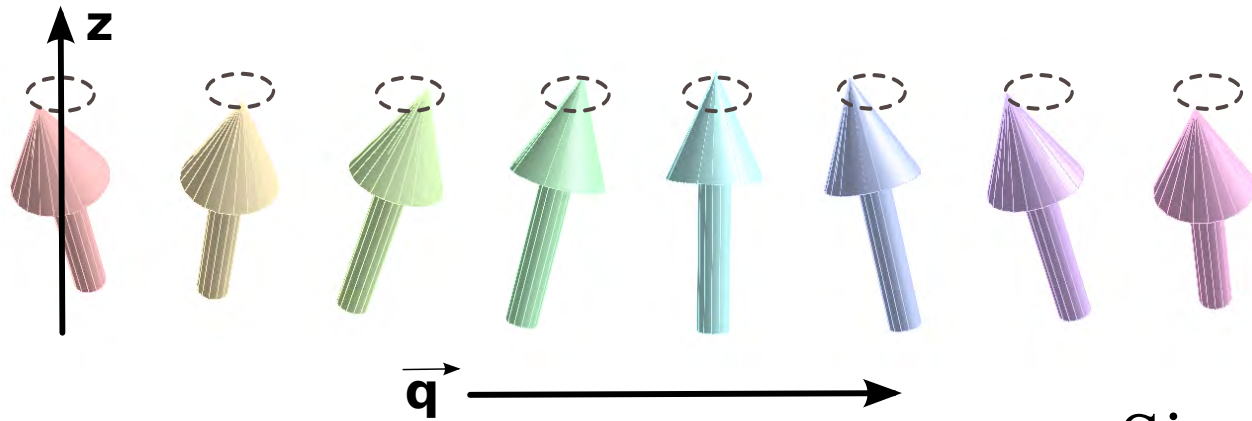
Ferromagnetic metal ($T_c=292$ K)



2nd step : magnons ?

hcp Gd: magnons

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

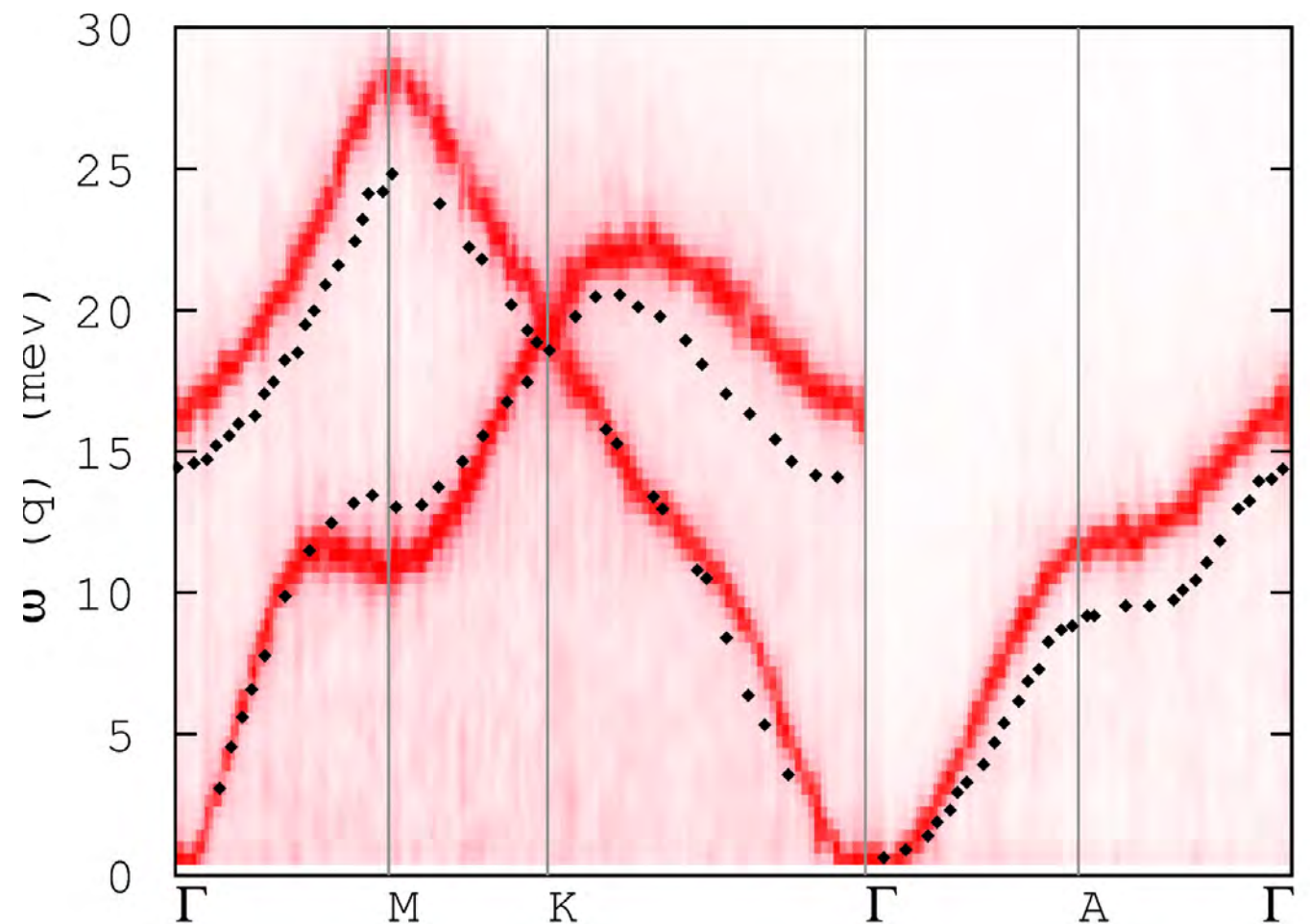


*Solving
the equation of motion:*

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

contains J_{ij} 's

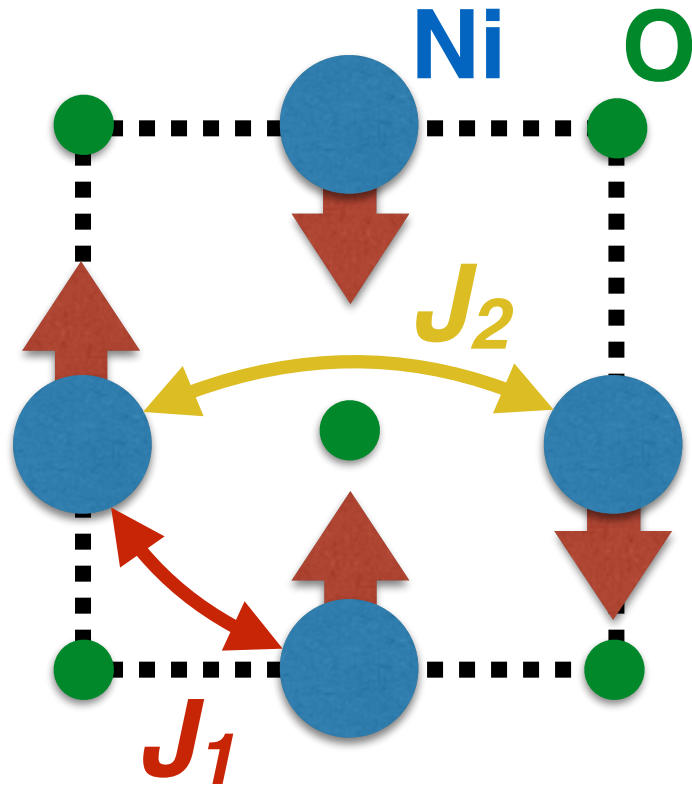
Simulated magnon spectrum at T=78 K



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

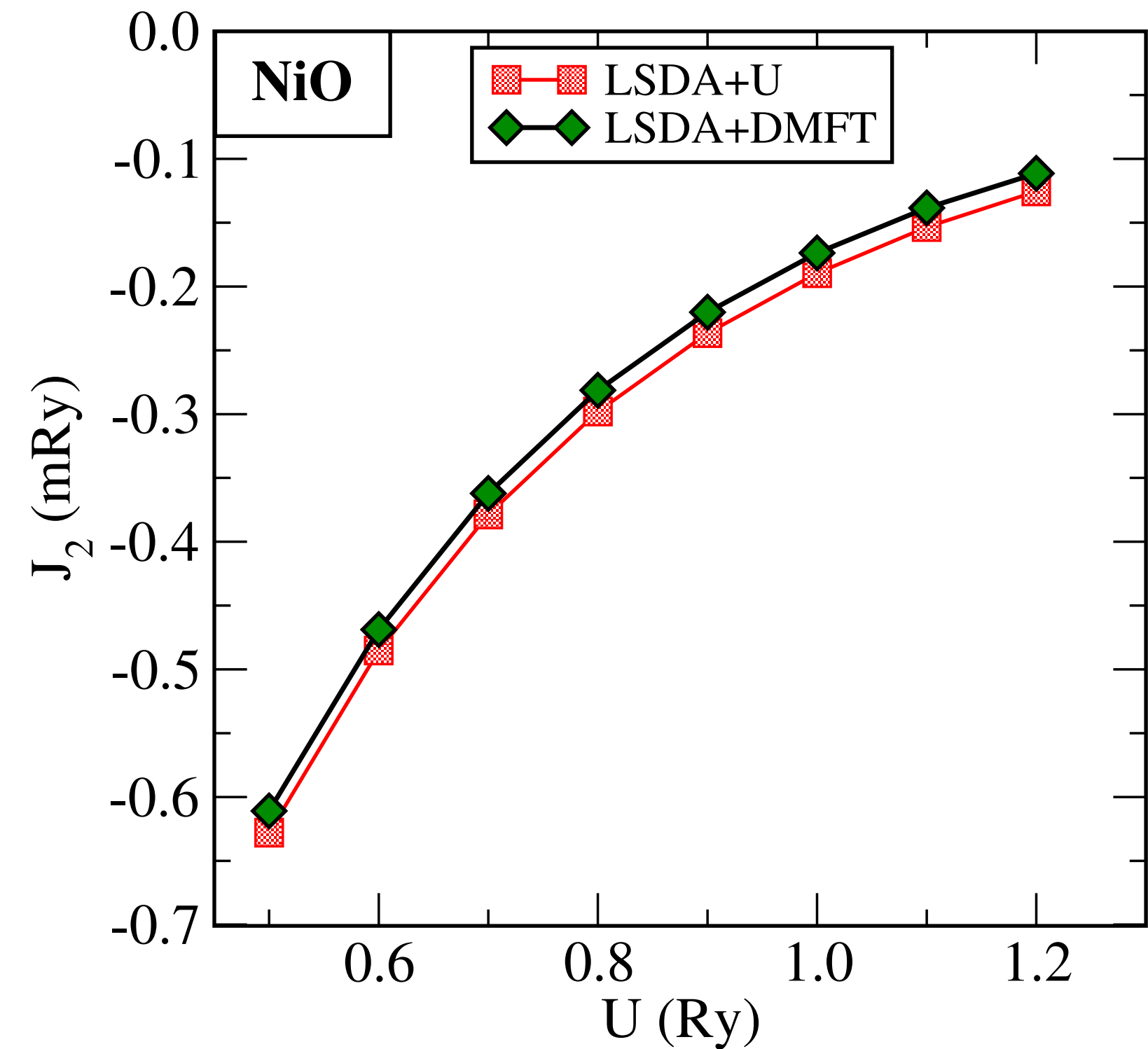
LDA+U already provides a good description of the J_{ij} 's

[1] Jacobsson PRB 88 134427 (2013)

[2] Hutchings PRB 6 3447 (1972)

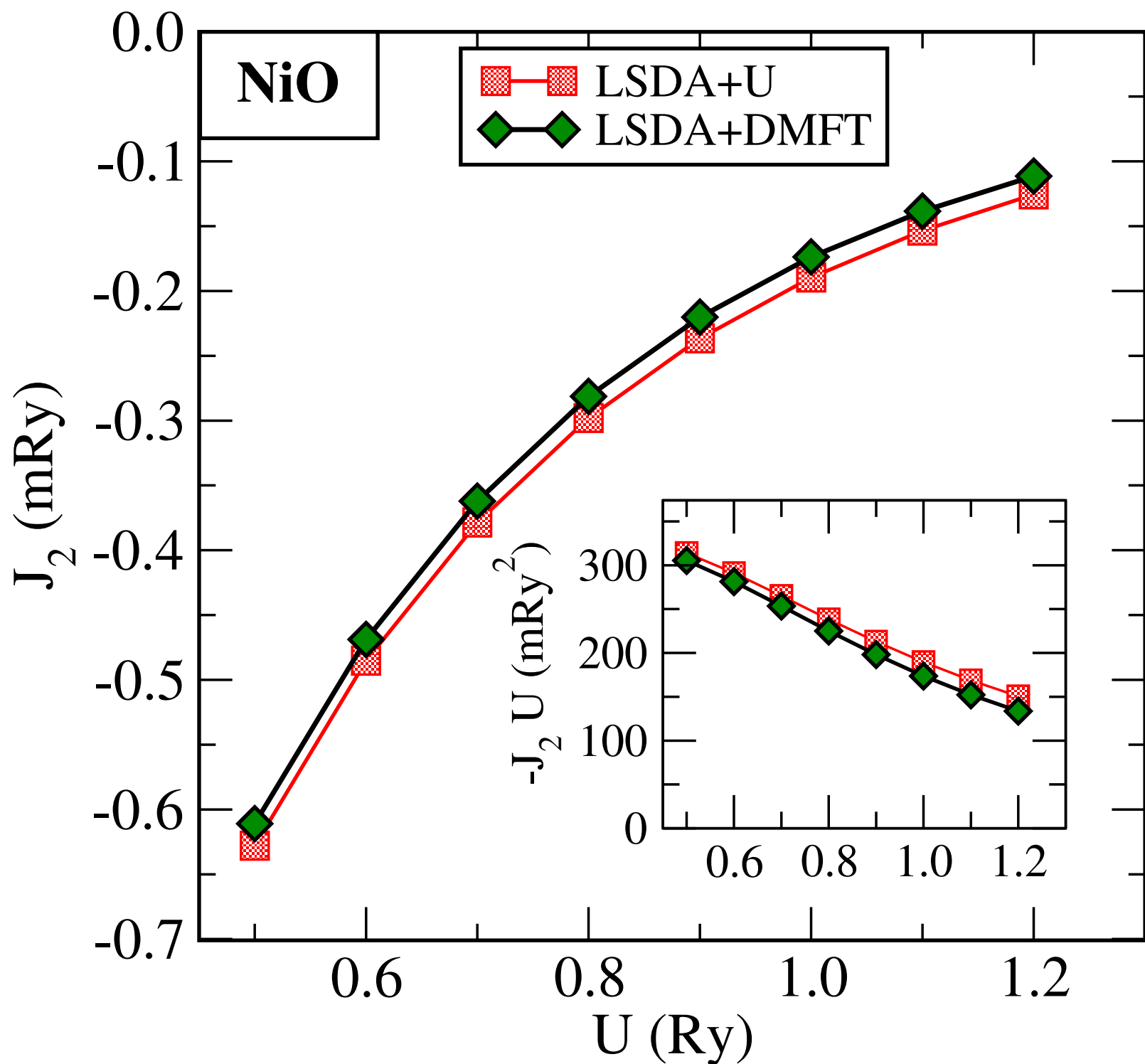
[3] Shanker PRB 7 5000 (1973)

NiO: J_2 as a function of U



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

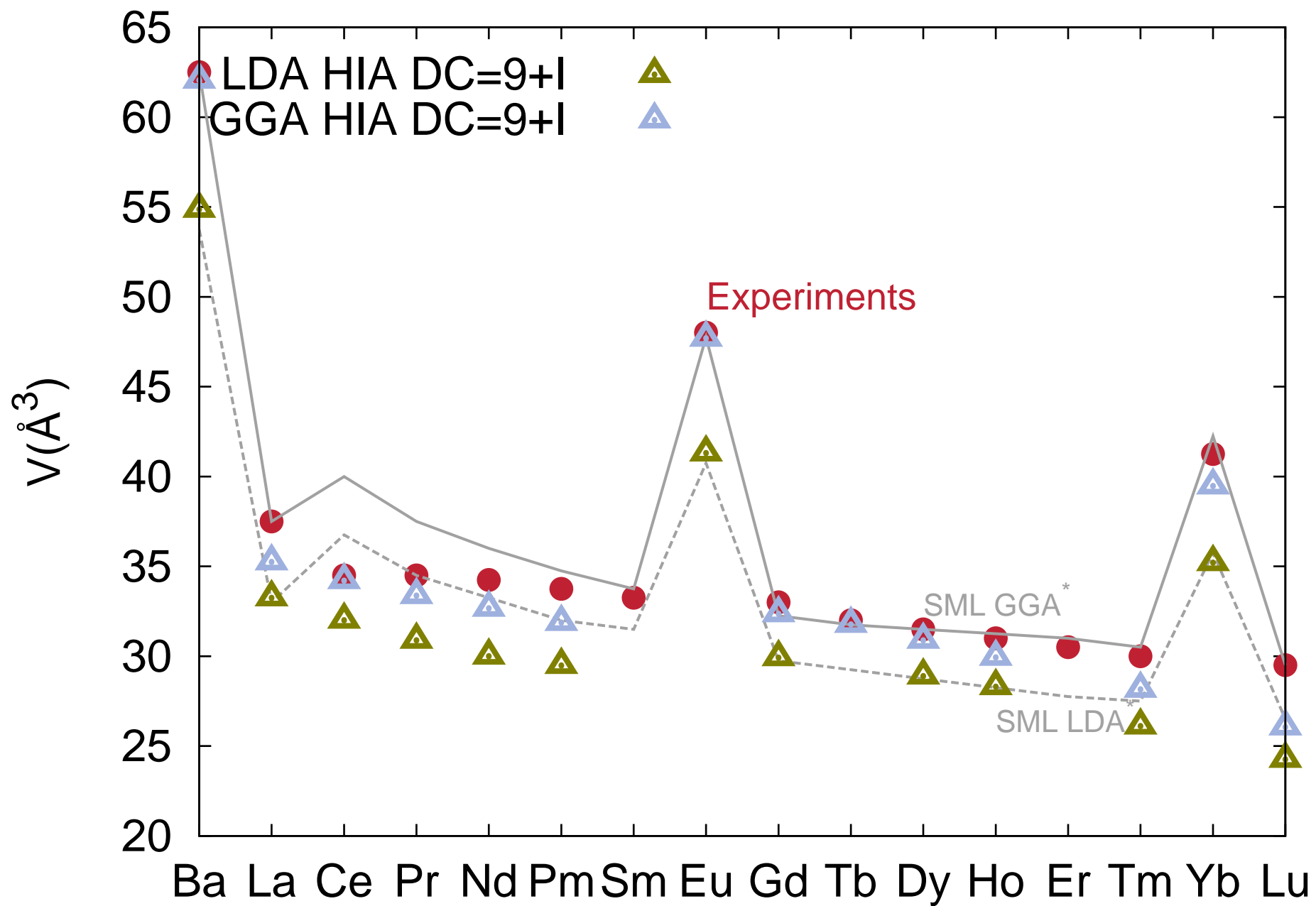
NiO: J_2 as a function of U

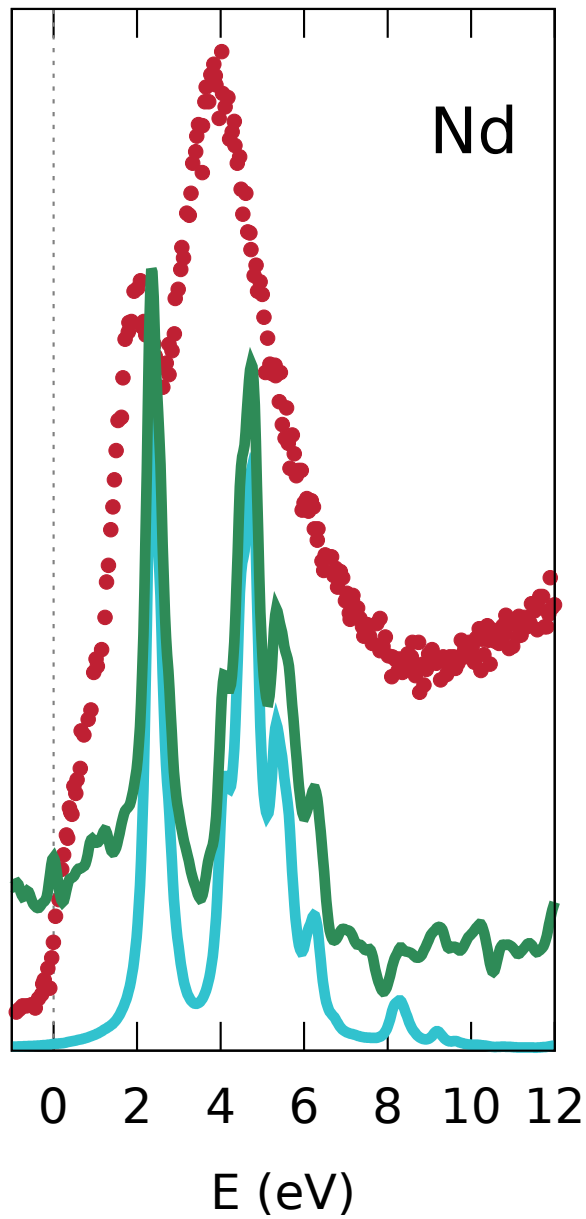
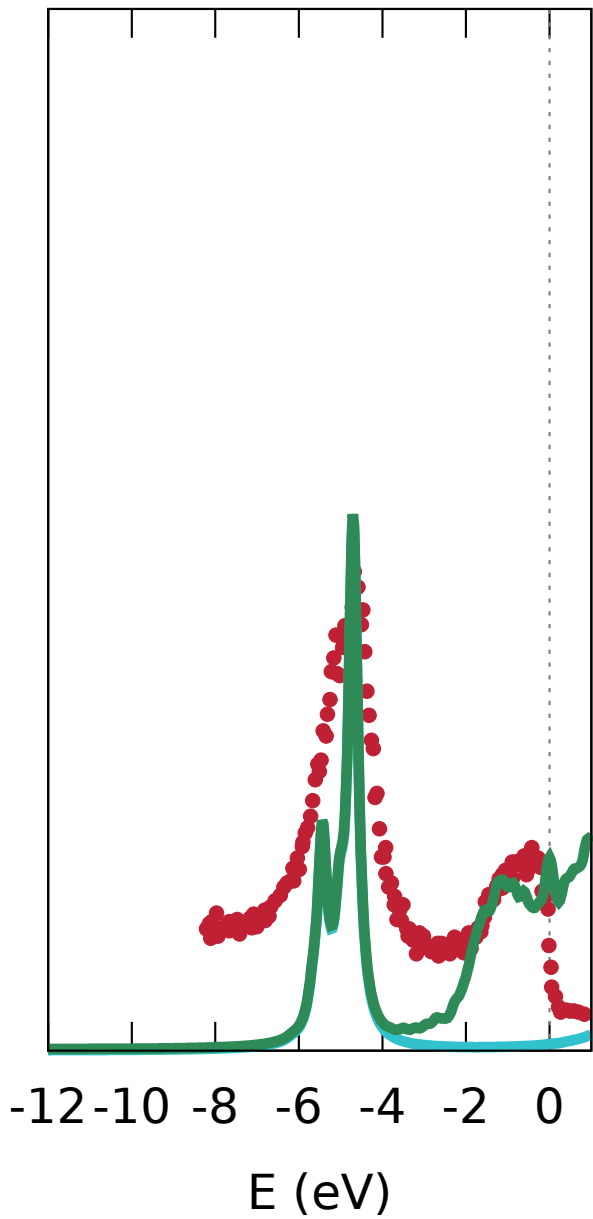


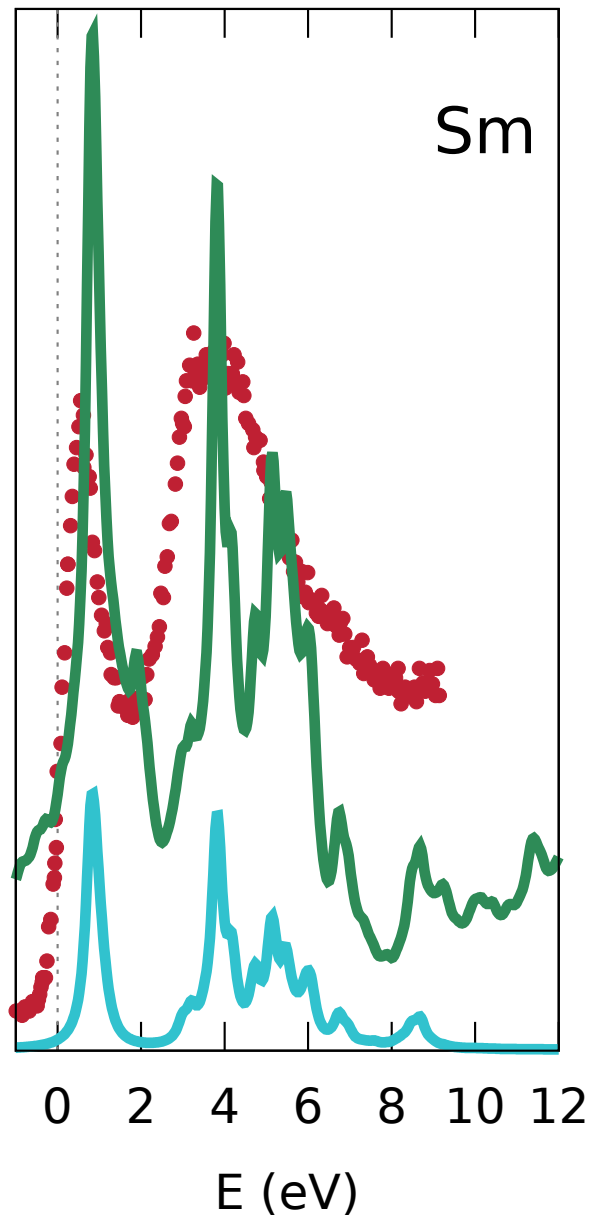
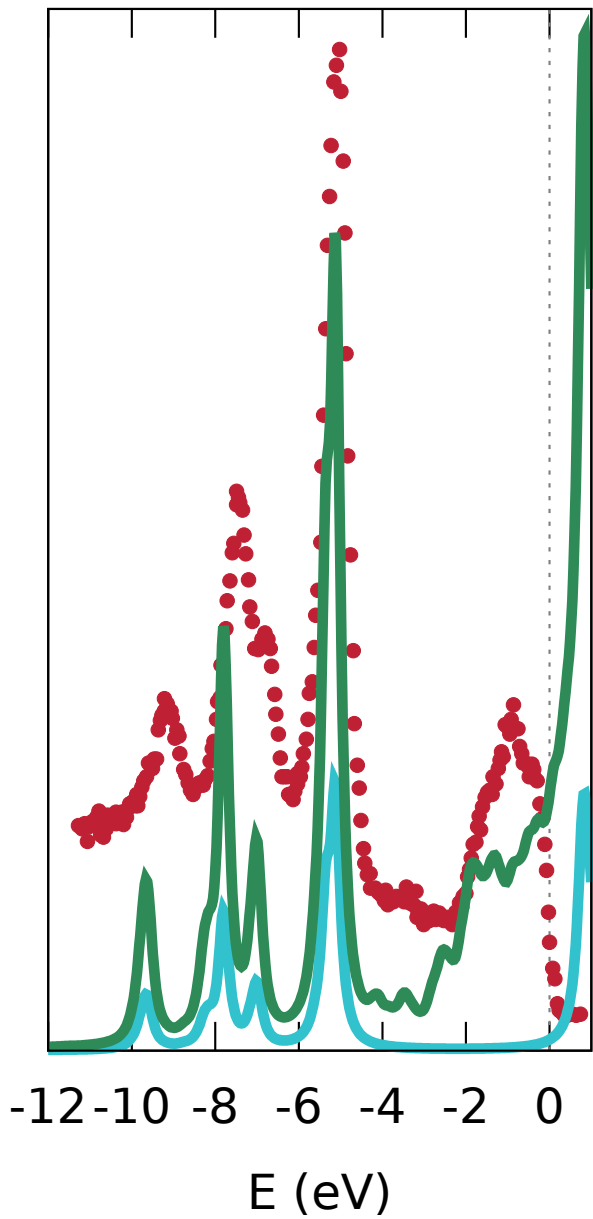
Does $\frac{t^2}{U}$ hold?

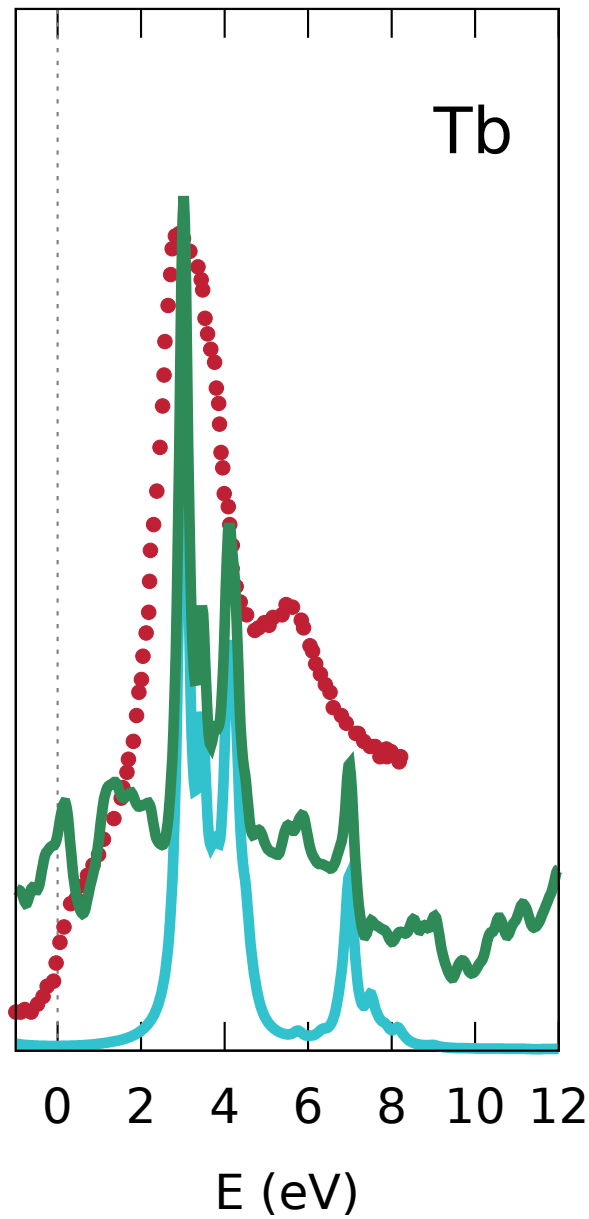
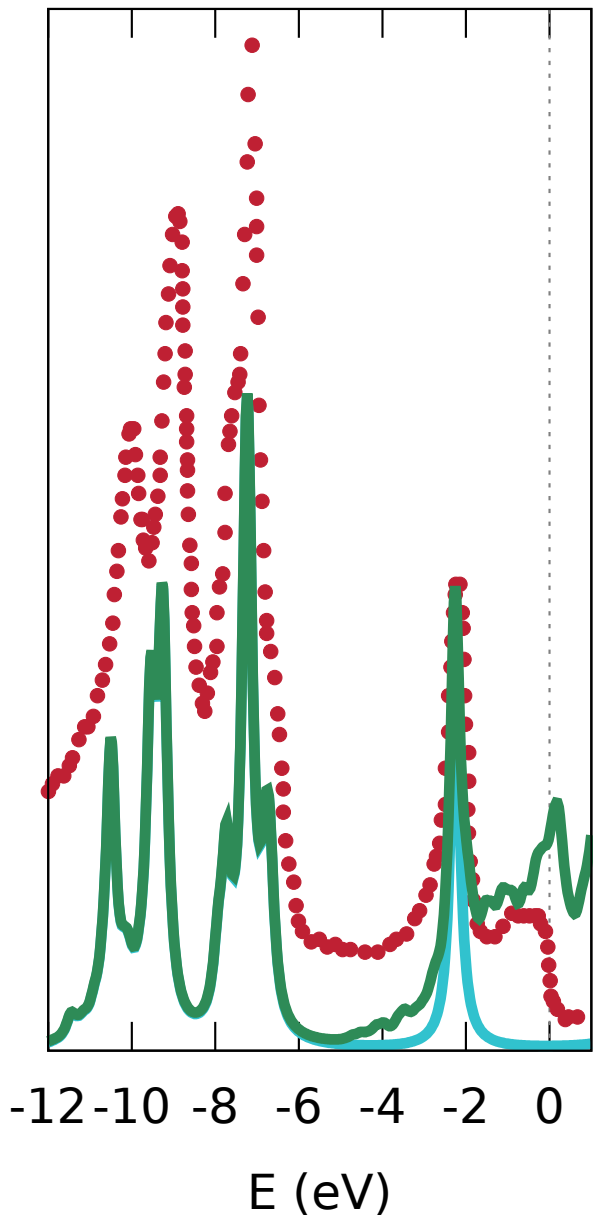
If yes: $J_2 U = t^2$

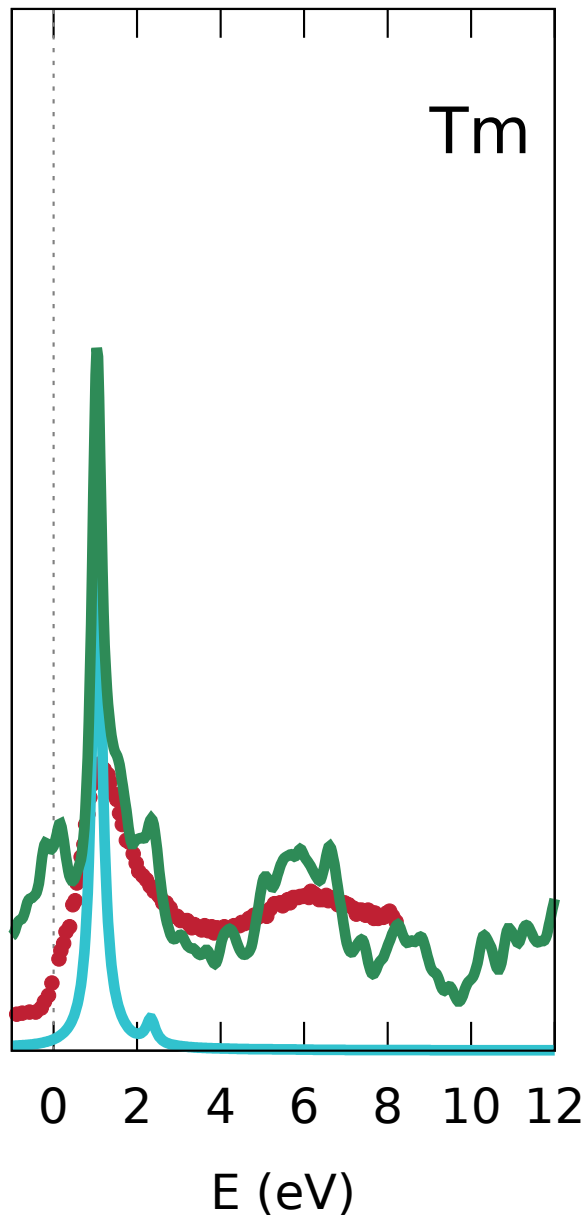
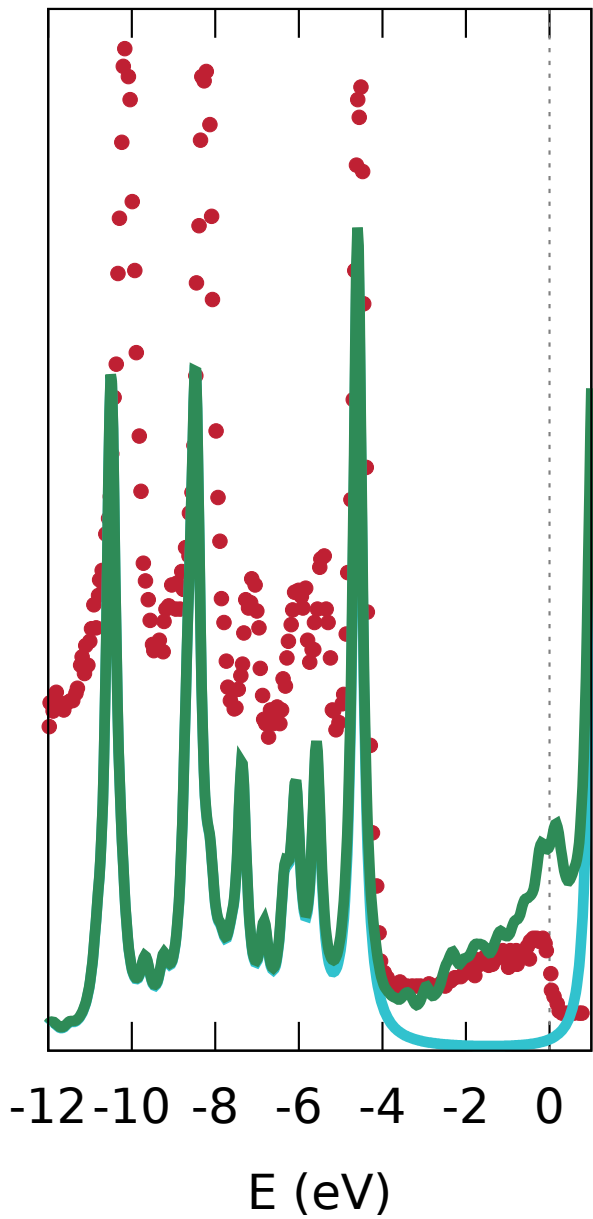
Hopping changes too!











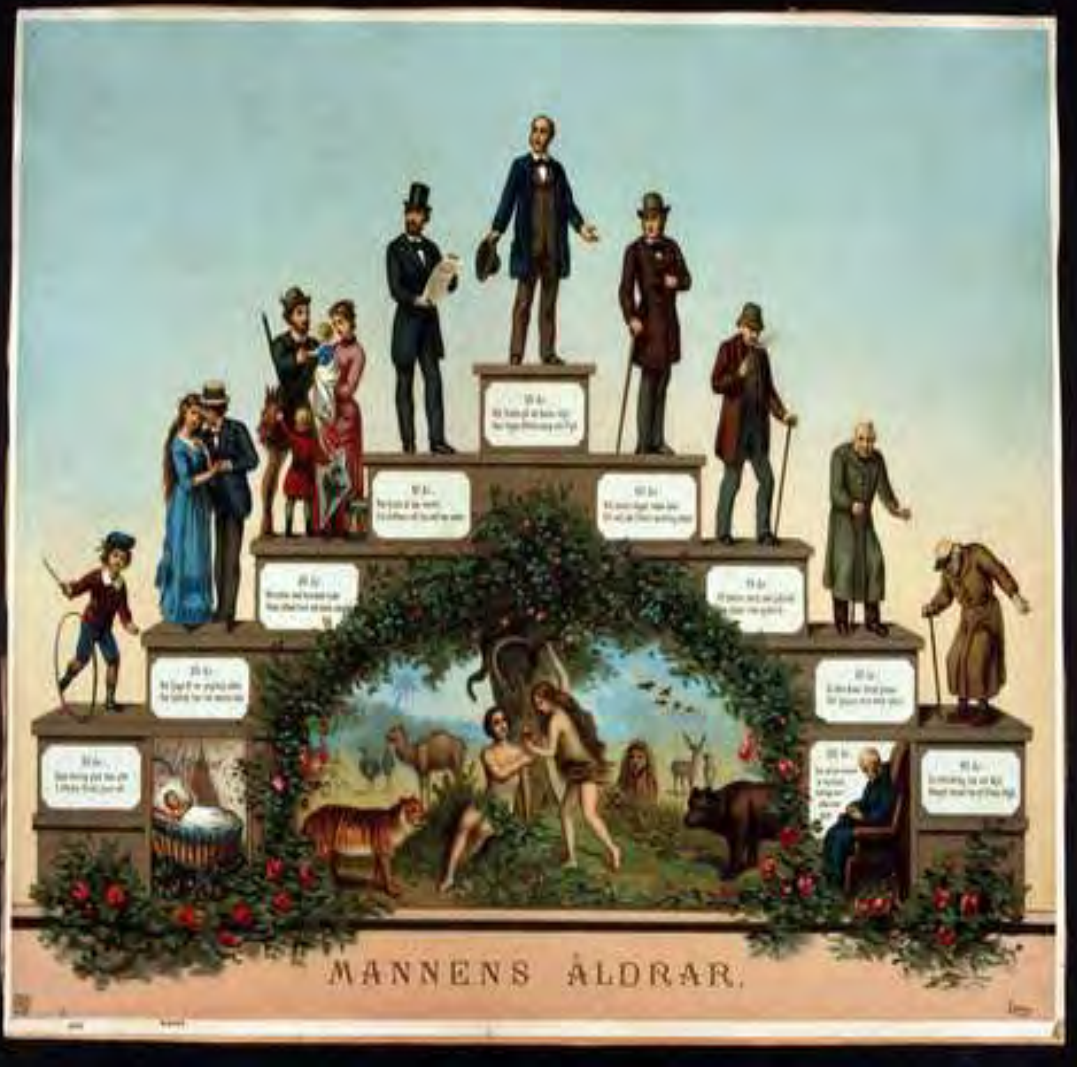
Mannens olika åldrar

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

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 50-60 ÅR ÄR MANNEN SOM EN GAMMAL FORD, MAN
MÅSTE HÅLLA I HONOM LITE SPRIT FÖR ATT FÅ HONOM ATT
STARTA.



Impact of correlations on J 's

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

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

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!

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