Radboud Universiteit Nijmegen

From first principles to magnetic Hamiltonians and spin dynamics

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Outline

1. Introduction
2. Time-dependent DFT and magnetic susceptibility
3. Exchange interactions from first principles
4. Beyond DFT: correlated systems and LDA+DMFT
5. Applications: Fe, Ni, Gd, NiO, CrO_2…
6. Dzyaloshinskii-Moriya interactions
7. Application: Molecular magnets
8. Outlook

Epigraphs

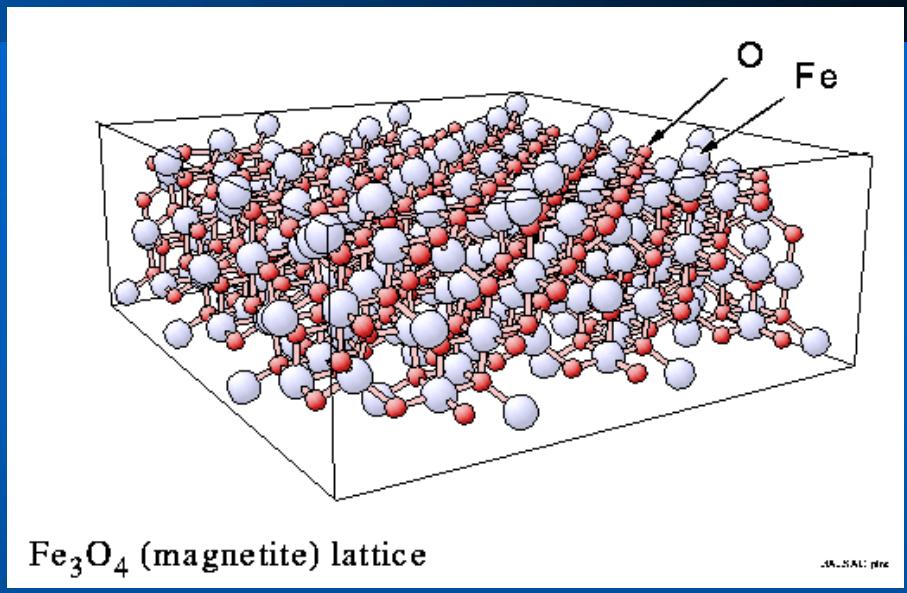
To the theoretical physicists, ferromagnetism presents a number of very interesting, unsolved and beautiful challenges. Our challenge is to understand why it exists at all.

(Feynman Lectures on Physics)

Make things as simple as possible but not simpler

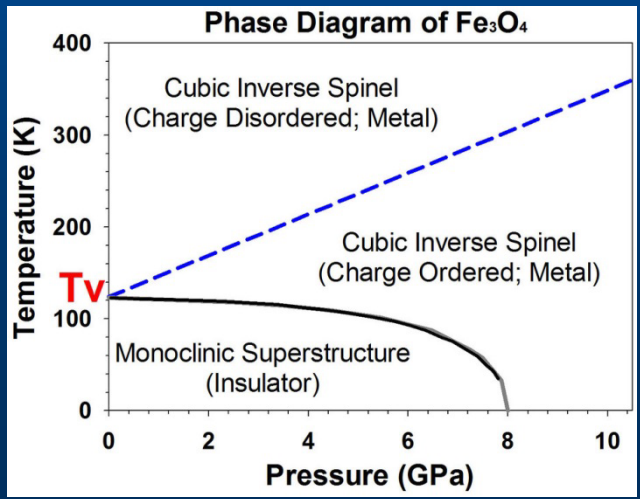
(A. Einstein)

Magnetite – first known magnet



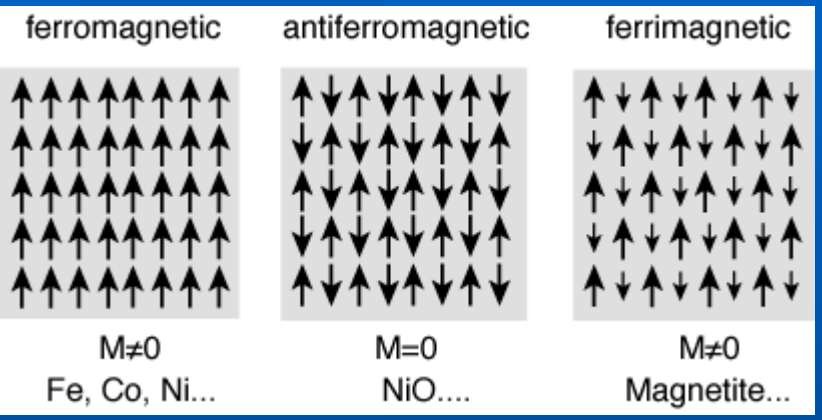
Very complicated structure, still a lot of open questions

Two types of Fe sites (tetra and octa);
Metal-insulator transition;
Charge ordering;
Role of orbital degrees of freedom;
Half-metallicity…



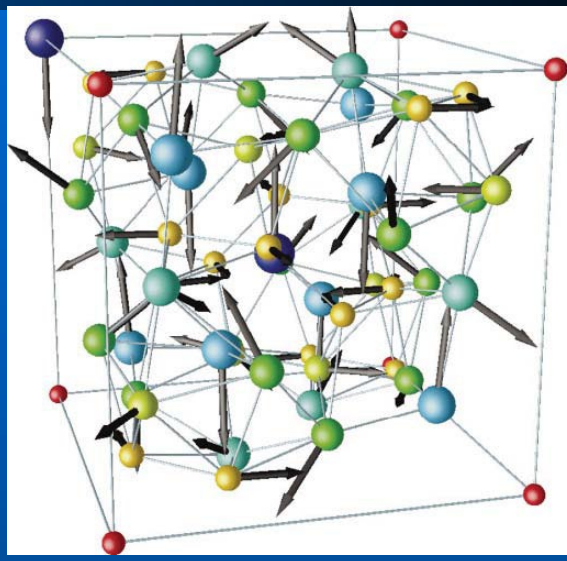
Types of magnetic ordering

Textbook wisdom

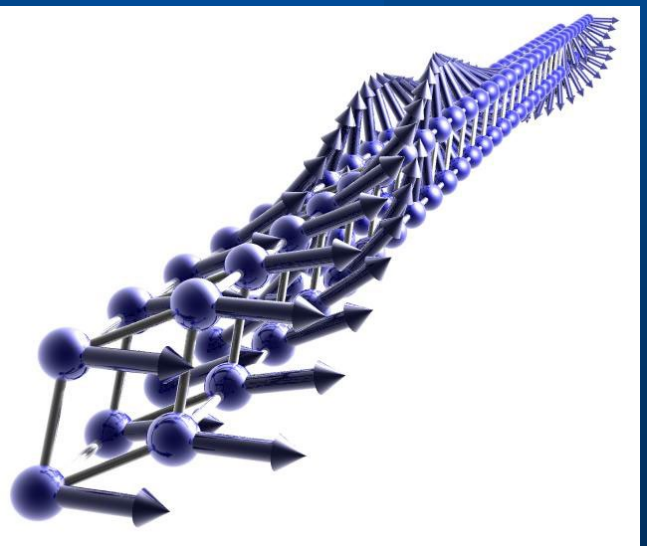


Sometimes very complicated

α-Mn

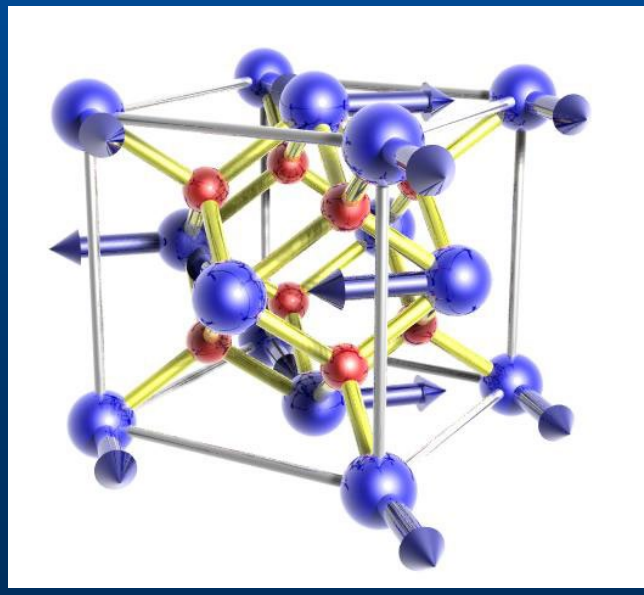


Spin spirals

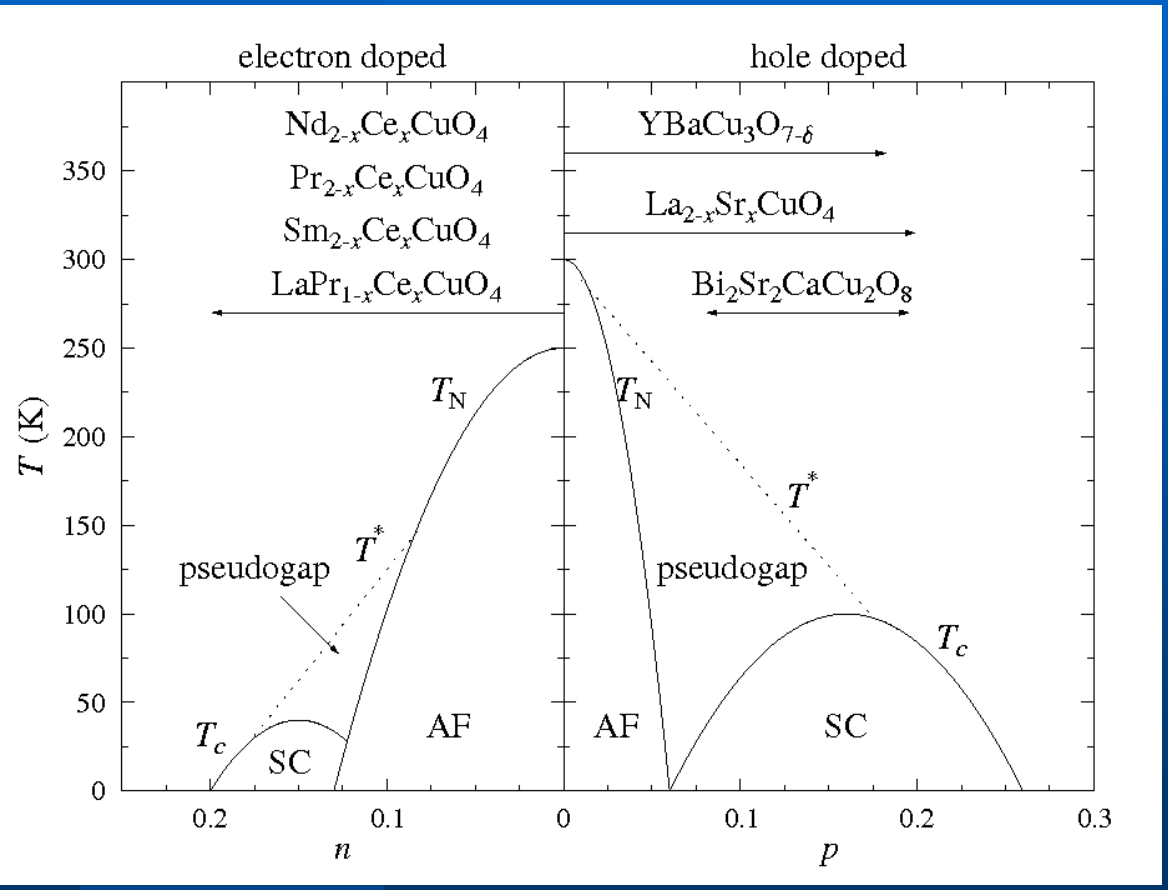


γ-Fe

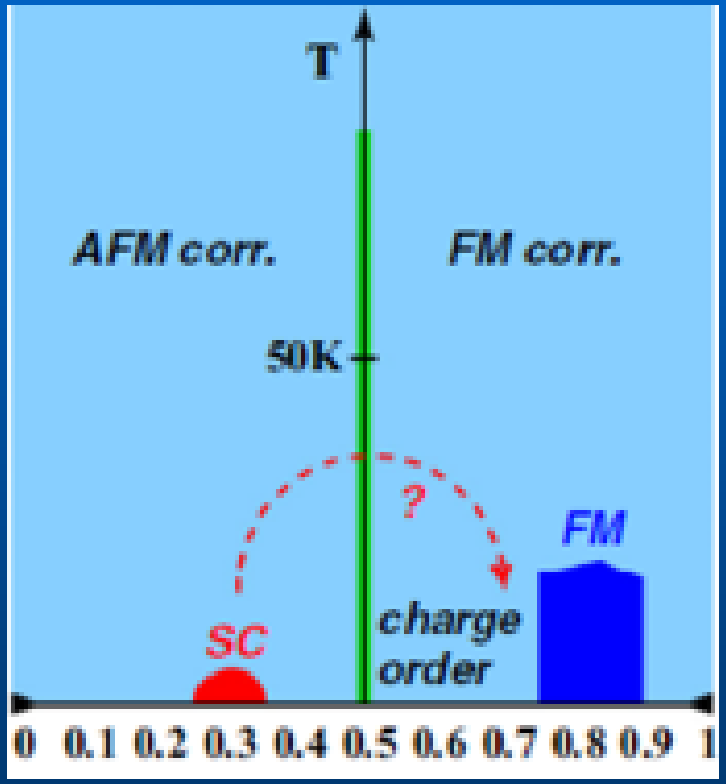
UO_2



Relation to superconductivity and other phenomena



Simplified phase diagram of Cu-O high-Tc superconductors



Layered cobaltates Na_xCoO_2

Types of magnetic interactions

$$\hat{H} = \sum_{ij} J_{ij} \hat{\vec{S}}_i \hat{\vec{S}}_j + \sum_{i\mu\nu} \hat{S}_i^{\mu} A_i^{\mu\nu} \hat{S}_i^{\nu} + \sum_{ij} \vec{D}_{ij} [\hat{\vec{S}}_i \times \hat{\vec{S}}_j]$$

The first term: exchange interactions (Heisenberg model)
Quantum, nonrelativistic (Coulomb interaction plus Pauli principle). Determine the type of magnetic ordering (mostly)

The second term: magnetic anisotropy
Quantum, relativistic (due to spin-orbit interaction). At least, second-order in SOC. Determine "practical" magnetism (hard and soft magnetic materials, hysteresis loop, etc.)

The third term: Dzyaloshinskii-Moriya interactions
Quantum, relativistic (due to spin-orbit interaction). First-order in SOC but require broken inversion symmetry. Responcible for weak FM, skyrmiones etc.

General formulation

System of interacting electrons (many-body problem) + crystal potential

External strong time-dependent laser field (nonequilibrium problem)

Temperature effects (thermal bath, open system, basic statistical mechanics)

Collect all difficulties of modern theoretical physics

Levels of description

- Macroscopic (LLG equations + temperature balance, etc.)

- Microscopic, classical Heisenberg model

- Microscopic, quantum itinerant-electron model

-Ab initio, time-dependent density functional

Multiscale problem

Time-dependent DFT

SE for many-body wave function in configurational space is replaced by single-particle nonlinear self-consistent equation

Spinor $\Psi = \begin{pmatrix} \Psi_+ \\ \Psi_- \end{pmatrix}$

$$i\frac{\partial \Psi}{\partial t} = [H_L - \hat{\boldsymbol{\sigma}} \cdot \mathbf{B}(\mathbf{r},t)]\Psi$$

B is self-consistent magnetic field

$$H_L = -\nabla_{\mathbf{r}}^2 + \sum_{\mathbf{R}} V_{\mathbf{rR}} + 2\int d\mathbf{r}' \frac{n(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} + V_{\mathrm{xc}}$$

Simplifications

Adiabatic approx.: V_{xc} and B_{xc} are the same as in the equilibrium + local (spin) density approx.

$$\mathrm{i}\frac{\partial \psi}{\partial t} = H\psi$$

$$H = -\nabla^2 + V(\mathbf{r}) - \tfrac{1}{2}(\mathbf{B}_{\mathrm{xc}}(\mathbf{r}) + \mathbf{B}_{\mathrm{ext}}(\mathbf{r}))\sigma$$

$$V(\mathbf{r}) = V_{\mathrm{ext}}(\mathbf{r}) + \int \mathrm{d}\mathbf{r}' \frac{n(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} + \frac{\partial}{\partial n}[n\varepsilon_{\mathrm{xc}}]$$

$$\mathbf{B}_{\mathrm{xc}} = -2\frac{\mathbf{m}}{m}\frac{\partial}{\partial m}[n\varepsilon_{\mathrm{xc}}]$$

n,m are charge and spin densities

Linear response: magnetic susceptibility

MIK & Lichtenstein, JPCM 16, 7439 (2004)

$$\mathbf{B}_{\mathrm{ext}}(\mathbf{r}) \rightarrow 0$$

$$\delta B^{\alpha}_{\mathrm{tot}} = \delta B^{\alpha}_{\mathrm{ext}} + \frac{\delta B^{\alpha}_{\mathrm{xc}}}{\delta m^{\beta}} \delta m^{\beta}$$

$$\delta m^{\alpha} = \hat{\chi}^{\alpha\beta} \delta B^{\beta}_{\mathrm{ext}}$$

$$(\hat{\chi}\varphi)(\mathbf{r}) = \int \mathrm{d}\mathbf{r}'\, \chi(\mathbf{r}, \mathbf{r}')\varphi(\mathbf{r}')$$

At the same time (Runge-Gross theorem, 1984) in TD-DFT

$$\delta m^{\alpha} = \hat{\chi}_0^{\alpha\beta} \delta B^{\beta}_{\mathrm{tot}}$$

A response of effective system of noninteracting Kohn-Sham particles (Liu & Vosko 1989 for magnetic case)

Linear response: magnetic susceptibility II

Rigorous exprression

$$\hat{\chi}^{\alpha\beta} = \hat{\chi}_0^{\alpha\beta} + \hat{\chi}_0^{\alpha\gamma} \frac{\delta B_{\mathrm{xc}}^{\gamma}}{\delta m^{\delta}} \hat{\chi}^{\delta\beta}$$

Adiabatic approximation plus LSDA:

$$\frac{\delta B_{\mathrm{xc}}^{\gamma}}{\delta m^{\delta}} = \frac{B_{\mathrm{xc}}}{m} \left(\delta_{\gamma\delta} - \frac{m^{\gamma} m^{\delta}}{m^2} \right) + \frac{m^{\gamma} m^{\delta}}{m^2} \frac{\partial B_{\mathrm{xc}}}{\partial m}$$

Transverse susceptibility is separated from (longitudinal spin + charge) susceptibilities

Transverse susceptibility

$$\chi^{+-}(\mathbf{r},\mathbf{r}',\omega) = \chi_0^{+-}(\mathbf{r},\mathbf{r}',\omega) + \int d\mathbf{r}''\, \chi_0^{+-}(\mathbf{r},\mathbf{r}'',\omega) I_{\rm xc}(\mathbf{r}'') \chi^{+-}(\mathbf{r}'',\mathbf{r}',\omega)$$

$$I_{\rm xc} = \frac{B_{\rm xc}}{m}$$

Local Stoner parameter

$$m = \sum_{\mu\sigma} \sigma f_{\mu\sigma} |\psi_{\mu\sigma}(\mathbf{r})|^2$$

$$n = \sum_{\mu\sigma} f_{\mu\sigma} |\psi_{\mu\sigma}(\mathbf{r})|^2.$$

$$\chi_0^{+-}(\mathbf{r},\mathbf{r}',\omega) = \sum_{\mu\nu} \frac{f_{\mu\uparrow} - f_{\nu\downarrow}}{\omega - \varepsilon_{\mu\uparrow} + \varepsilon_{\nu\downarrow}} \psi^*_{\mu\uparrow}(\mathbf{r}) \psi_{\nu\downarrow}(\mathbf{r}) \psi^*_{\nu\downarrow}(\mathbf{r}') \psi_{\mu\uparrow}(\mathbf{r}')$$

Kohn-Sham states

$$(H_0 - \tfrac{1}{2}\sigma B_{\rm xc})\psi_{\mu\sigma} = \varepsilon_{\mu\sigma}\psi_{\mu\sigma}$$

$$H_0 = -\nabla^2 + V(\mathbf{r})$$

Longitudinal susceptibility

$$\chi^{zz} = \frac{1}{4}\left(K^{\uparrow\uparrow} + K^{\downarrow\downarrow} - K^{\uparrow\downarrow} - K^{\downarrow\uparrow}\right)$$

$$\begin{aligned}
K^{\uparrow\uparrow} &= X_{\uparrow} + X_{\uparrow}U_{\uparrow\uparrow}K^{\uparrow\uparrow} + X_{\uparrow}U_{\uparrow\downarrow}K^{\downarrow\uparrow} \\
K^{\downarrow\downarrow} &= X_{\downarrow} + X_{\downarrow}U_{\downarrow\downarrow}K^{\downarrow\downarrow} + X_{\downarrow}U_{\downarrow\uparrow}K^{\uparrow\downarrow} \\
K^{\uparrow\downarrow} &= X_{\uparrow}U_{\uparrow\downarrow}K^{\downarrow\downarrow} + X_{\uparrow}U_{\uparrow\uparrow}K^{\uparrow\downarrow} \\
K^{\downarrow\uparrow} &= X_{\downarrow}U_{\downarrow\uparrow}K^{\uparrow\uparrow} + X_{\downarrow}U_{\downarrow\downarrow}K^{\downarrow\uparrow}.
\end{aligned}$$

$$X_{\sigma}\left(\mathbf{r},\mathbf{r}'\right) = \sum_{\mu\nu}\frac{f_{\mu\sigma} - f_{\nu\sigma}}{\omega - \varepsilon_{\mu\sigma} + \varepsilon_{\nu\sigma}}\psi^{*}_{\mu\sigma}(\mathbf{r})\psi_{\nu\sigma}(\mathbf{r})\psi_{\mu\sigma}(\mathbf{r}')\psi^{*}_{\nu\sigma}(\mathbf{r}')$$

$$U_{\sigma\sigma'} = \frac{\partial^2\left(n\varepsilon_{\mathrm{xc}}\right)}{\partial n_{\sigma}\partial n_{\sigma'}}$$

$$n_{\sigma} = \frac{1}{2}\left(n + \sigma m\right)$$

Separation of magnon poles

After rigorous manipulations

$$\hat{\chi}^{+-} = (m + \hat{\Lambda})(\omega - I_{\mathrm{xc}}\hat{\Lambda})^{-1}$$

$$\Lambda(\mathbf{r}, \mathbf{r}', \omega) = \sum_{\mu\nu} \frac{f_{\mu\uparrow} - f_{\nu\downarrow}}{\omega - \varepsilon_{\mu\uparrow} + \varepsilon_{\nu\downarrow}} \psi^*_{\mu\uparrow}(\mathbf{r})\psi_{\nu\downarrow}(\mathbf{r})\nabla[\psi_{\mu\uparrow}(\mathbf{r}')\nabla\psi^*_{\nu\downarrow}(\mathbf{r}') - \psi^*_{\nu\downarrow}(\mathbf{r}')\nabla\psi_{\mu\uparrow}(\mathbf{r}')]$$

Magnon pole

$$\omega(\mathbf{q}) = \frac{4}{M}\left[J(0) - J(\mathbf{q})\right]$$

$$J(\mathbf{r}, \mathbf{r}', \omega) = \frac{1}{4}\sum_{\mu\nu} \frac{f_{\mu\uparrow} - f_{\nu\downarrow}}{\omega - \varepsilon_{\mu\uparrow} + \varepsilon_{\nu\downarrow}} \psi^*_{\mu\uparrow}(\mathbf{r})B_{\mathrm{xc}}(\mathbf{r})\psi_{\nu\downarrow}(\mathbf{r})\psi^*_{\nu\downarrow}(\mathbf{r}')B_{\mathrm{xc}}(\mathbf{r}')\psi_{\mu\uparrow}(\mathbf{r}')$$

Im part corresponds to Stoner damping

Alternative definition of exchanges

Static susceptibility

$$\hat{\chi}^{+-}(0) = m(\hat{\Omega}^{-1} - B_{\mathrm{xc}}^{-1})$$

$$\hat{\tilde{\Omega}} = \hat{\Omega}(1 - B_{\mathrm{xc}}^{-1}\hat{\Omega})^{-1}$$

The first way (poles of susceptibility) corresponds Liechtenstein, MIK & Gubanov, J. Phys. F 1984, the second way (static suscept.) Bruno, PRL 2003. The expressions for stiffness constant coincide and are rigorous within the adiabatic approximation + LSDA

Magnetic force theorem

(Lichtenstein,MIK, Gubanov, J. Phys. F 1984; Sol. St. Comm. 1985)

Total energy in DFT

$$E = E_{sp} - E_{dc}$$

$$E_{sp} = \sum_{\nu}^{occ} \varepsilon_{\nu}$$

$$E_{dc} = E_{Hartree} + \int dr Tr\left[\rho \frac{\delta E_{xc}}{\delta \rho}\right] - E_{xc}$$

Variation

$$\delta E = \delta^* E_{sp} + \delta_1 E_{sp} - \delta E_{dc} = \delta^* E_{sp} = \delta^* \int_{-\infty}^{\varepsilon_F} d\varepsilon \left[\frac{1}{\pi} Tr\, \mathrm{Im}\, \hat{G}(\varepsilon)\right]$$

δ^* at fixed potential δ_1 due to change of potential

Magnetic force theorem II

- Torque can be written in terms of variation of the density of states
- Decomposition of the torque in pair terms gives exchange integrals
- These exchange parameters are local (near given magnetic configuration)

Journal of Magnetism and Magnetic Materials 67 (1987) 65–74
North-Holland, Amsterdam

LOCAL SPIN DENSITY FUNCTIONAL APPROACH TO THE THEORY OF EXCHANGE INTERACTIONS IN FERROMAGNETIC METALS AND ALLOYS

A.I. LIECHTENSTEIN, M.I. KATSNELSON [+], V.P. ANTROPOV [+] and V.A. GUBANOV

Table 1
Values of exchange interaction parameters calculated by the cluster Green's function method

Metal	J_0 (meV)	T_c (K)	T_c^{expt} (K)	J_1 (meV)	J_2 (meV)	D (meV Å^2)	D^{expt} (meV Å^2)
Fe	155.7	1200	1040 [a]	20.5	−3.4	294	314 [b]
Ni	49.1	380	630 [a]	1.9	0.23	386	395 [c]

Non-Heisenberg character of exchange interactions in Fe and Ni

S.A. Turzhevskii, A.I. Lichtenstein, and M.I. Katsnelson, Fiz. Tverd. Tela **32**, 1952 (1990) [Sov. Phys. Solid State **32**, 1138 (1990)].

Rotation of a central spin: magnetic moment is not constant, energy change is not cosine

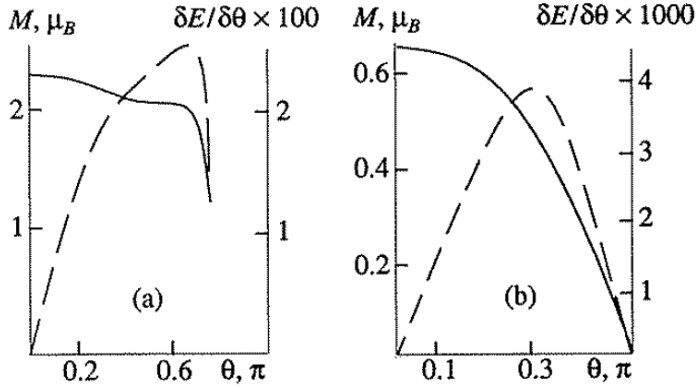


Fig. 4.4. *Magnetic moment in Bohr magnetons (the full curve) and the first derivative of energy with respect to angle of rotation in Ry (the dashed curve) according to calculations in [168]: (a) Fe, (b) Ni.*

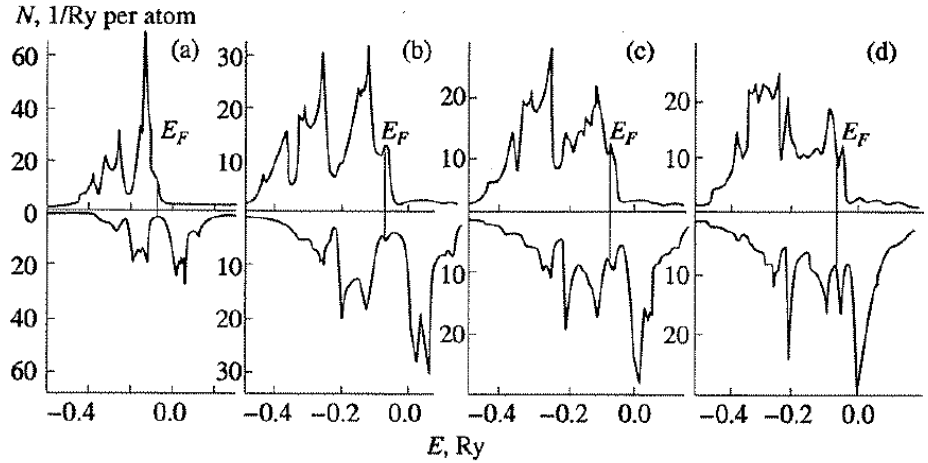


Fig. 4.5. *Electronic density of states for an Fe atom in a metal with the magnetic moment turned through θ = 0 (a), θ = 0.2π (b), θ = 0.35π (c), and θ = 0.5π (d).*

Electronic structure is angle-dependent

Nonlocal corrections to magnon stiffness

MIK & Antropov, PRB 67, 140406 (2003)

Exchange and correlation in spiral state of homogeneous electron gas

Angular gradient corrections

$$E_{xc} = \int d\mathbf{r}\{n\varepsilon_{xc}(n_\uparrow, n_\downarrow) + \lambda(n_\uparrow, n_\downarrow)D\}$$

$$D = (\nabla_\alpha e_\beta)(\nabla_\alpha e_\beta) = (\nabla\theta)^2 + \sin^2\theta(\nabla\varphi)^2$$

$$\lambda(n_\uparrow, n_\downarrow) = -\frac{e^2}{16\pi^2}\left(\frac{1}{F} - \frac{4}{3}\right)(V_{xc}^{\uparrow} p_{F\uparrow} + V_{xc}^{\downarrow} p_{F\downarrow})$$

$$-\frac{e^2}{96\pi^2 F^2}(V_{xc}^{\uparrow} + V_{xc}^{\downarrow})(p_{F\uparrow} + p_{F\downarrow}).$$

Corrections to stiffness constant

$$F = (p_{F\uparrow} + p_{F\downarrow})I(n_\uparrow, n_\downarrow)/2\pi^2$$

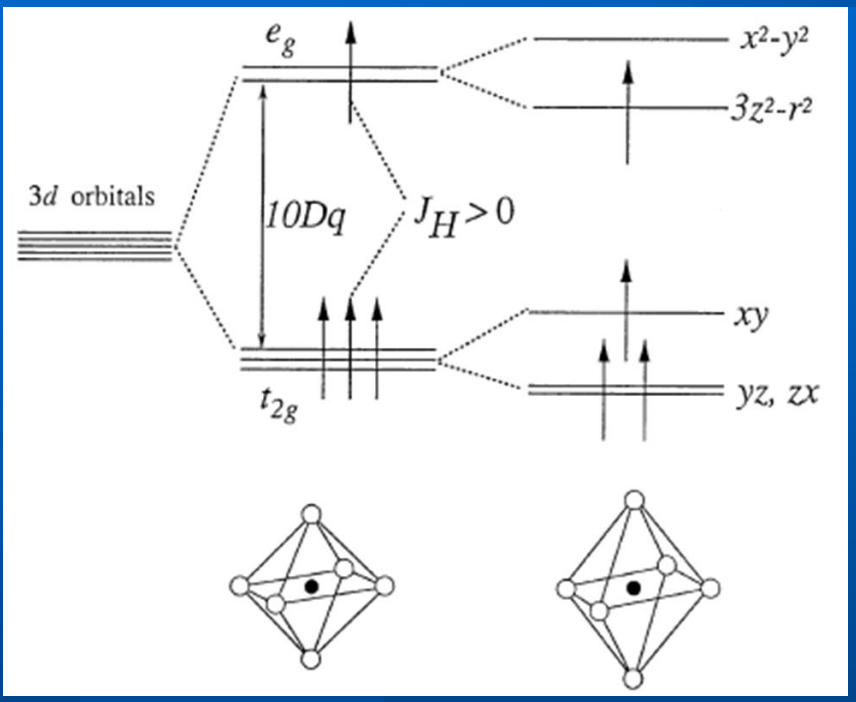
Stiffness constants for Fe and Ni

(in meV/Å^2)

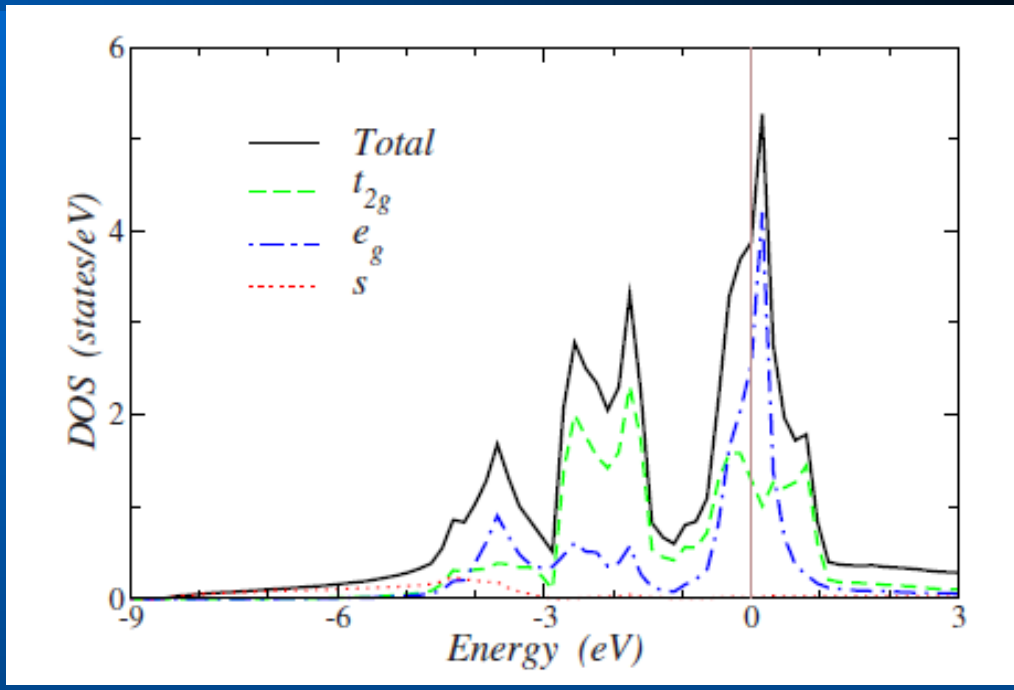
Fe: LSDA	*239*
with gradient corrections	*251*
experiment	*280 - 310*
Ni: LSDA	*692*
with gradient corrections	*735*
experiment	*550-630*

Corrections are quite small

Iron: some details



Crystal field splitting



DOS for nonmagnetic bcc Fe

Stoner criterion is fulfilled due to e_g states only; they should play a special role in magnetism of Fe (Irkhin, Katsnelson, Trefilov, JPCM 5, 8763 (1993))

Iron: detailed analysis

PRL 116, 217202 (2016) PHYSICAL REVIEW LETTERS week ending 27 MAY 2016

Microscopic Origin of Heisenberg and Non-Heisenberg Exchange Interactions in Ferromagnetic bcc Fe

Y. O. Kvashnin,[1] R. Cardias,[2] A. Szilva,[1] I. Di Marco,[1] M. I. Katsnelson,[3,4] A. I. Lichtenstein,[4,5] L. Nordström,[1] A. B. Klautau,[2] and O. Eriksson[1]

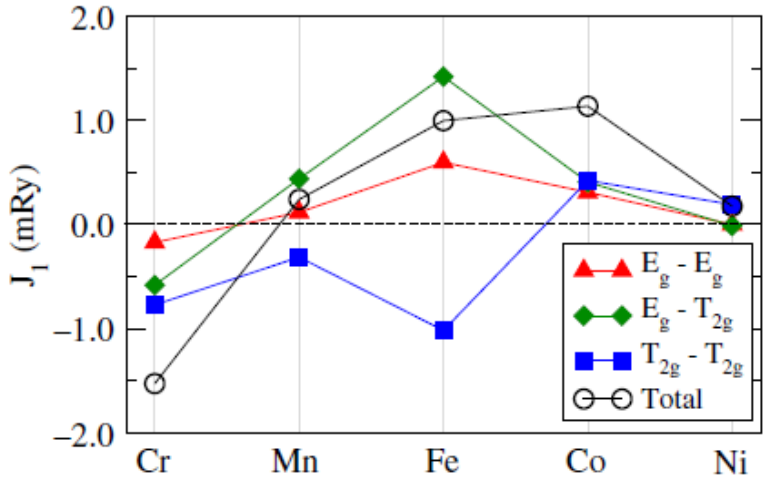
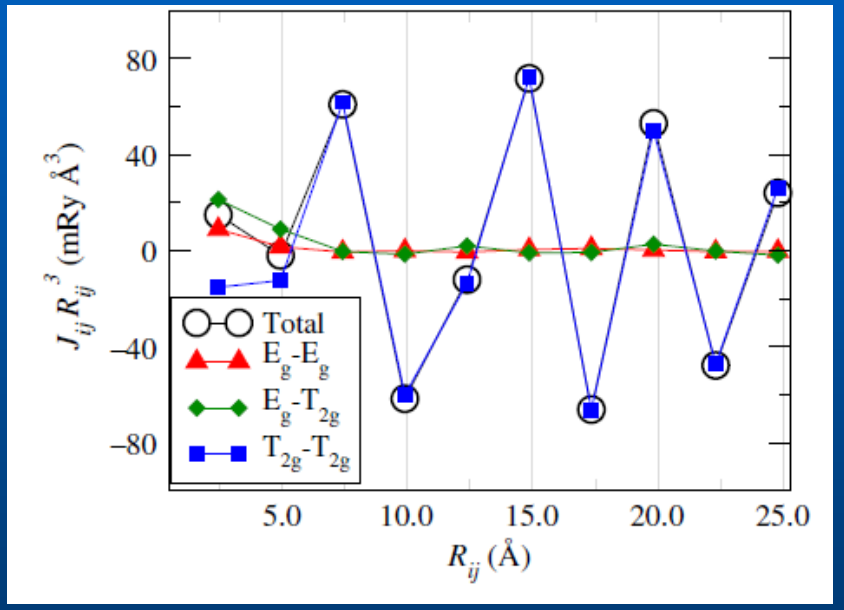
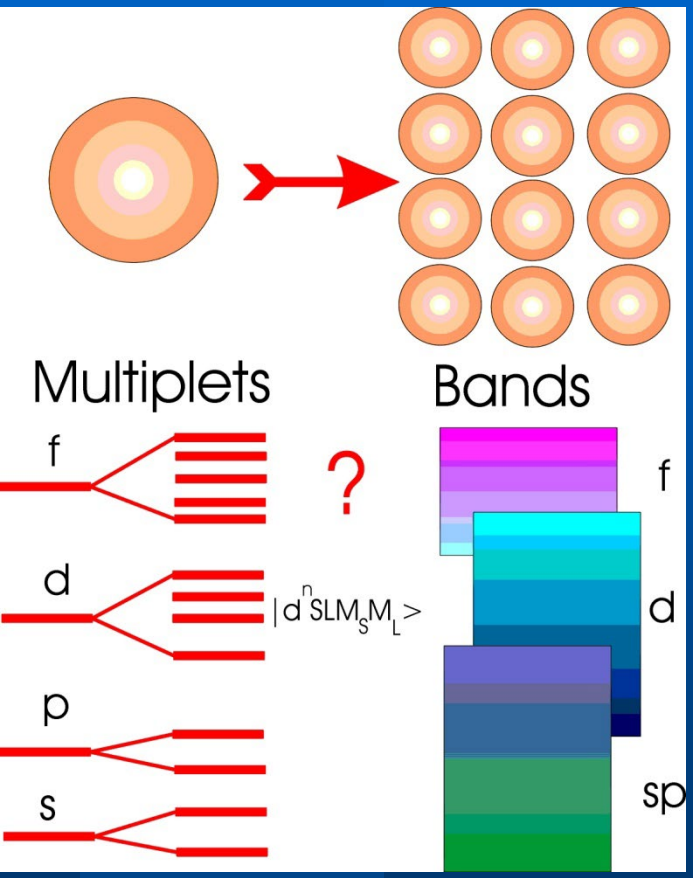


FIG. 1. Orbitally decomposed NN exchange interaction in elemental 3*d* metals in the bcc structure.



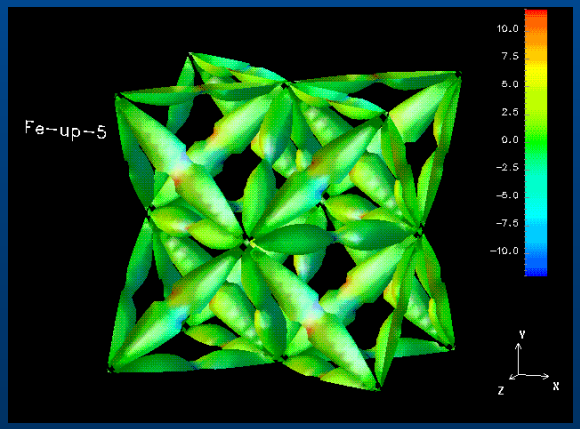
t_{2g} are itinerant electrons providing (Heisenberg-like) RKKY exchange with Friedel oscillations; e_g are more correlated providing (non-Heisenberg) "double exchange" typical for narrom-band systems

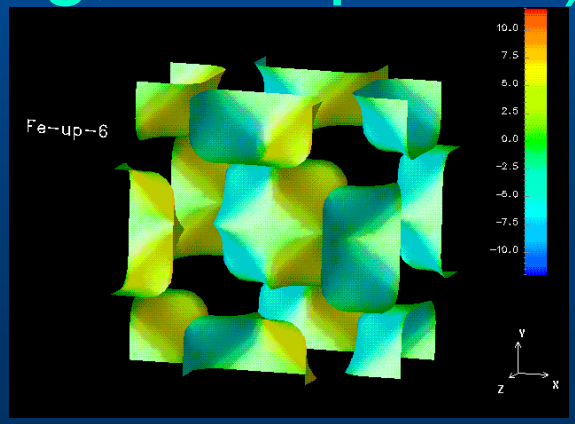
Problem with DFT: coexistence of localized and itinerant behavior



Local magnetic moments do exist above T_C (Curie-Weiss law, spectroscopy, neutrons…)

d electrons are itinerant (FS, chemical bonding, transport…)





Iron, majority spin FS

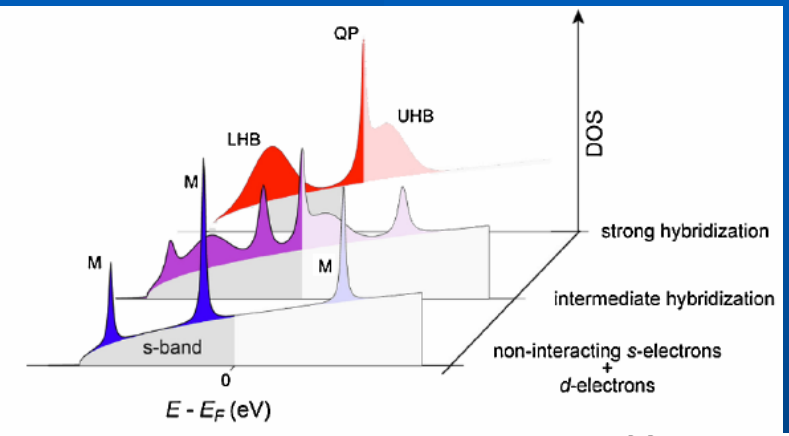
4f electrons are normally pure localized but not 3d

From atomic state to itinerant

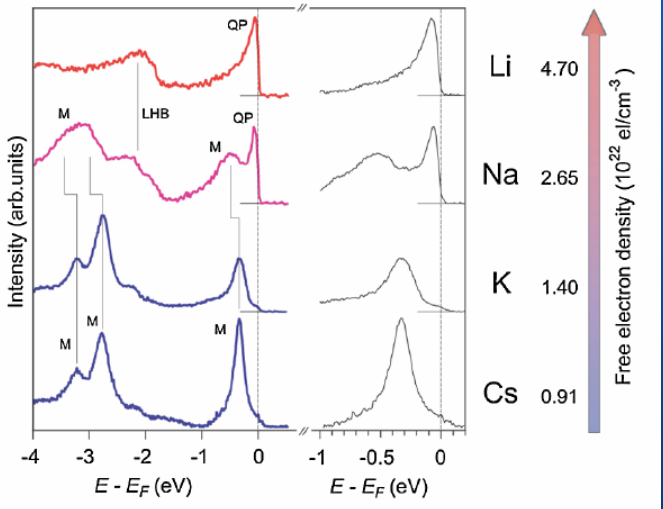
PRL **104,** 117601 (2010) PHYSICAL REVIEW LETTERS week ending 19 MARCH 2010

Correlated Electrons Step by Step: Itinerant-to-Localized Transition of Fe Impurities in Free-Electron Metal Hosts

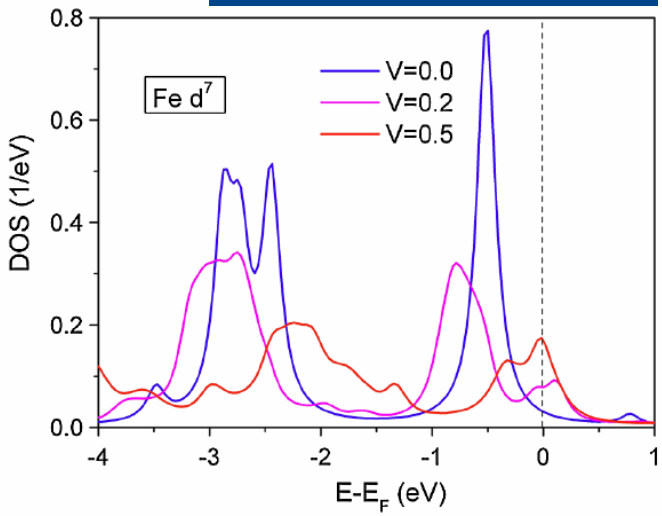
C. Carbone,[1] M. Veronese,[1] P. Moras,[1] S. Gardonio,[1] C. Grazioli,[1] P. H. Zhou,[2] O. Rader,[3] A. Varykhalov,[3] C. Krull,[4] T. Balashov,[4] A. Mugarza,[4] P. Gambardella,[4,5] S. Lebèque,[6] O. Eriksson,[7] M. I. Katsnelson,[8] and A. I. Lichtenstein[9]



Experiment: disappearance of multiplets



Calculations: increase of hybridization



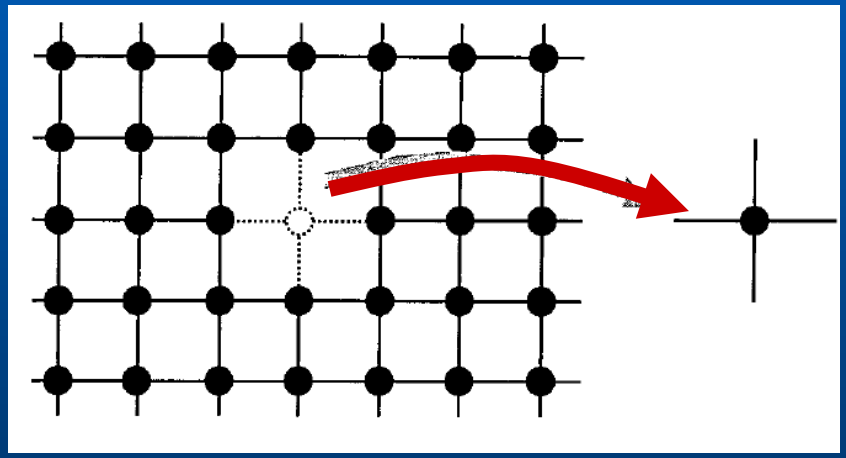
Blue line: exact diagonalization for free atom

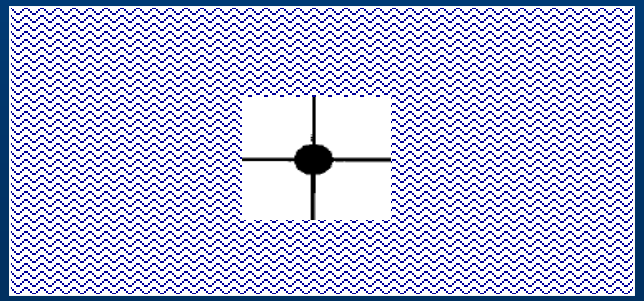
Dynamical Mean Field Theory

A.Georges, G.Kotliar, W.Krauth and M.Rozenberg, Rev. Mod. Phys. '96

A natural generalization of the familiar MFT
to the problem of electrons in a lattice

Key idea: take one site out of a lattice and embed it in a self-consistent bath = mapping to an effective impurity problem





LDA (DFT)+DMFT

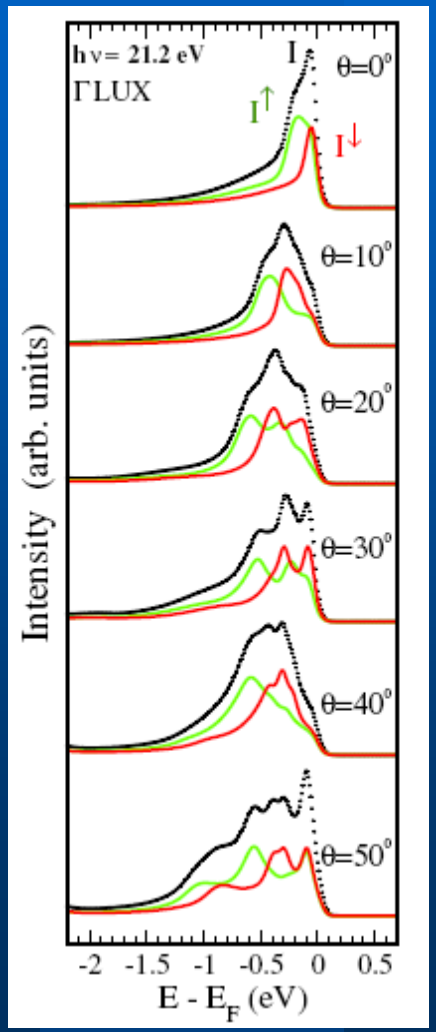
(Lichtenstein & MIK 1997, 1998,1999; Anisimov et al 1997)

LSDA	LDA++
Density functional	Baym-Kadanoff functional
Density $\rho(\mathbf{r})$	Green-Function $G(\mathbf{r},\mathbf{r}',E)$
Potential $V_{xc}(\mathbf{r})$	Self-energy $\Sigma_i(E)$
$E_{tot} = E_{sp} - E_{dc}$	$\Omega = \Omega_{sp} - \Omega_{dc}$
$E_{sp} = \sum_{\lambda<\lambda_F} \varepsilon_\lambda$	$\Omega_{sp} = -Tr\ln[-G^{-1}]$
$E_{dc} = E_H + \int \rho V_{xc} d\mathbf{r} - E_{xc}$	$\Omega_{dc} = Tr\Sigma G - \Phi_{LW}$
Temperature:	Matsubara frequencies: real-T
in the Fermi function	for collective excitations

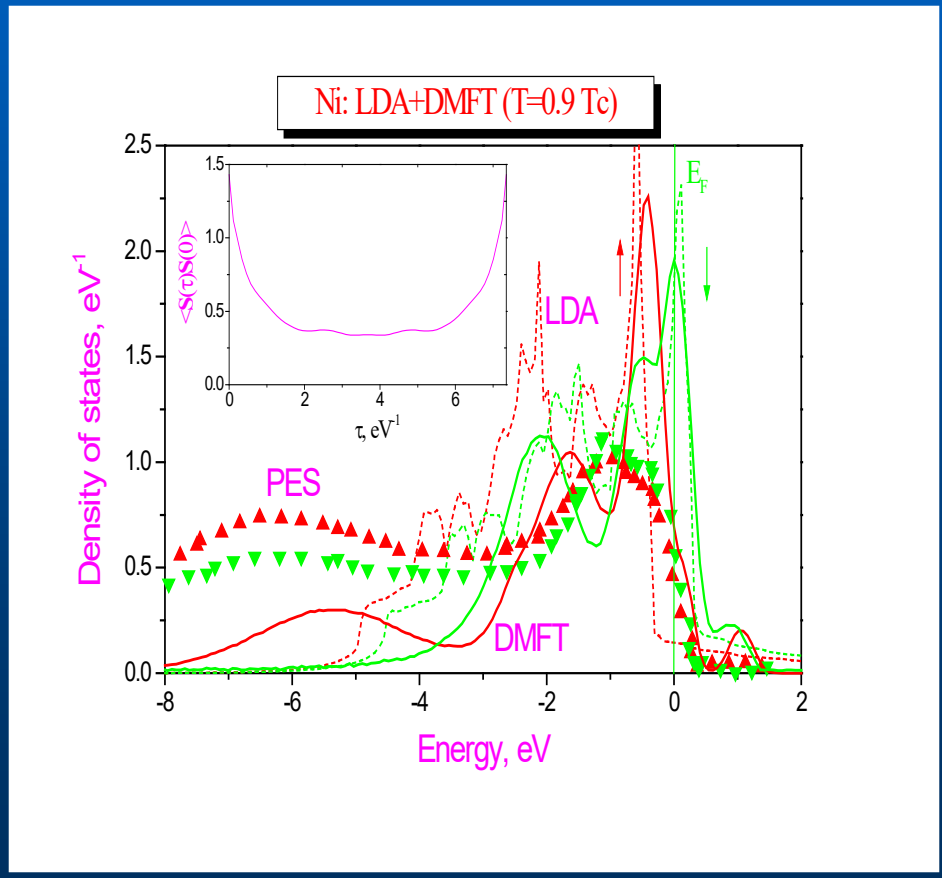
Ferromagnetism of transition metals: LDA+DMFT

Ferromagnetic Ni DMFT vs. LSDA:

- 30% band narrowing
- 50% spin-splitting reduction
- -6 eV sattellite



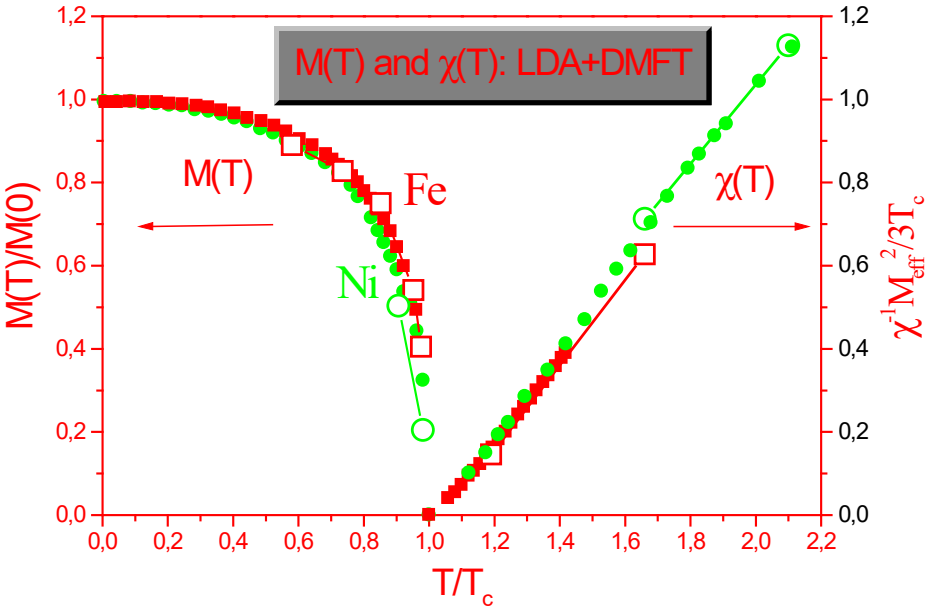
LDA+DMFT with ME
J. Braun *et al*
PRL (2006)



Lichtenstein, MIK, Kotliar, PRL (2001)

DMFT Effective Magnetic Moments: $T>T_c$

W	exp	eff	loc	DLM	Tc	exp
Fe	3.13	3.09	2.8	1.96	1900	1043
Ni	1.62	1.5	1.3	1.21	700	631



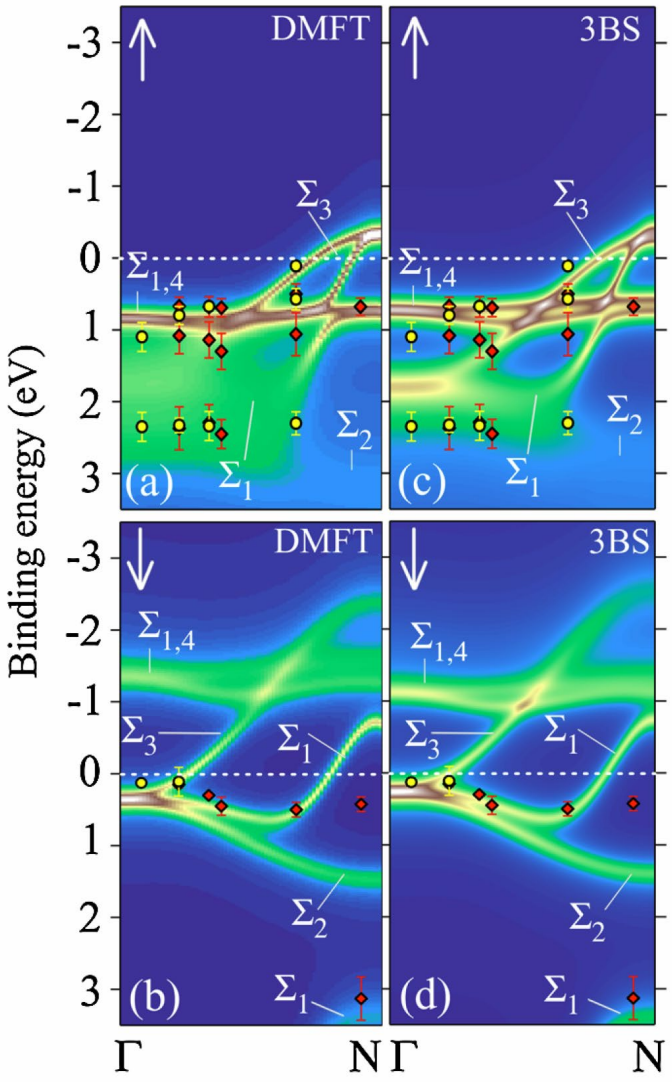
ARPES for iron

PRL 103, 267203 (2009) PHYSICAL REVIEW LETTERS week ending 31 DECEMBER 2009

Strength of Correlation Effects in the Electronic Structure of Iron

J. Sánchez-Barriga,[1] J. Fink,[1,2] V. Boni,[3] I. Di Marco,[4,5] J. Braun,[6] J. Minár,[6] A. Varykhalov,[1] O. Rader,[1] V. Bellini,[3] F. Manghi,[3] H. Ebert,[6] M. I. Katsnelson,[5] A. I. Lichtenstein,[7] O. Eriksson,[4] W. Eberhardt,[1] and H. A. Dürr[1]

Agreement is not bad (much better than LDA/GGA) but essentially worse than in nickel. Correlations in iron are not quite local

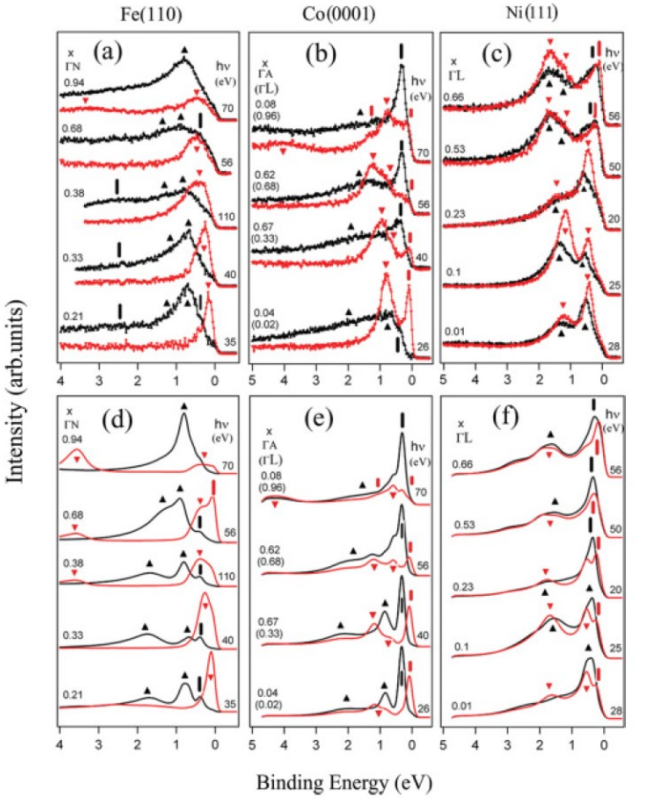


ARPES for 3d metals

PHYSICAL REVIEW B **85**, 205109 (2012)

Effects of spin-dependent quasiparticle renormalization in Fe, Co, and Ni photoemission spectra: An experimental and theoretical study

J. Sánchez-Barriga,[1] J. Braun,[2] J. Minár,[2] I. Di Marco,[3] A. Varykhalov,[1] O. Rader,[1] V. Boni,[4] V. Bellini,[5] F. Manghi,[4] H. Ebert,[2] M. I. Katsnelson,[6] A. I. Lichtenstein,[7] O. Eriksson,[3] W. Eberhardt,[1] H. A. Dürr,[1,8] and J. Fink[1,9]



Variation of U does not help too much for Fe

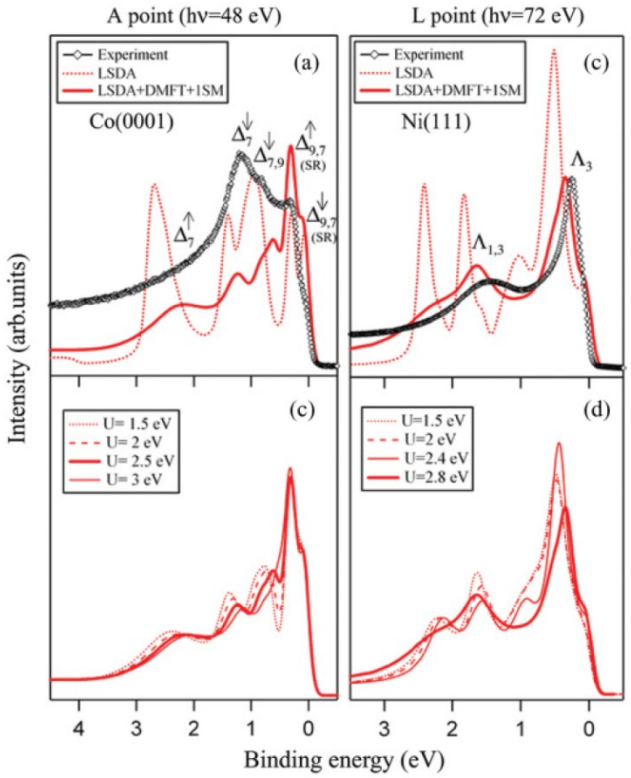


TABLE I. Values of the experimental and theoretical mass enhancement factors m^*/m_0 for majority spin states at high symmetry points of the BBZ of Fe, Co, and Ni, respectively. The theoretical values are derived for $U(\mathrm{Fe}) = 1.5$ eV, $U(\mathrm{Co}) = 2.5$ eV, $U(\mathrm{Ni}) = 2.8$ eV.

Fe			Co			Ni		
	Expt.	Theory		Expt.	Theory		Expt.	Theory
Γ	1.7	1.2	Γ	1.26	1.31	Γ	2.0	1.8
N	1.1	1.2	A	1.29	1.31	Λ	1.9	1.8

Black – spin up, red – spin down
Upper panel – exper,lower - DMFT

Exchange and LW Functional

MIK & Lichtenstein Phys. Rev. B 61, 8906 (2000)

Luttinger-Ward functional

Magnetic force theorem

$$\begin{aligned}\Omega^d &= \Omega^d_{sp} - \Omega^d_{dc} \\ \Omega^d_{sp} &= -Tr\left\{\ln\left[\Sigma - G_0^{-1}\right]\right\} \\ \Omega^d_{dc} &= Tr\Sigma G - \Phi\end{aligned}$$

$$G^{-1} = G_0^{-1} - \Sigma$$

$$\Sigma = \frac{\delta\Phi}{\delta G}$$

$$\delta\Omega = \delta^*\Omega_{sp} + \delta_1\Omega_{sp} - \delta\Omega_{dc}$$

$$\delta_1\Omega_{sp} = \delta\Omega_{dc} = TrG\delta\Sigma$$

$$\delta\Omega = \delta^*\Omega_{sp} = -\delta^* Tr\ln\left[\Sigma - G_0^{-1}\right]$$

Exchange interactions from DMFT

Heisenberg exchange:

$$H = -\sum_{ij} J_{ij} S_i S_j$$

$$\delta\Omega = \delta^* \Omega_{sp} = \mathbf{V}_i \delta\varphi_i$$

Magnetic torque:

$$\delta \mathbf{e}_i = \delta\varphi_i \times \mathbf{e}_i$$

$$\mathbf{V}_i = 2Tr_{\omega L}\left[\Sigma_i^s \times \mathbf{G}_{ii}^s\right]$$

Exchange interactions:

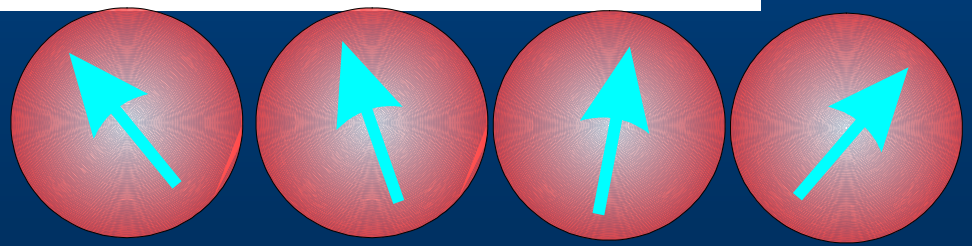
$$J_{ij} = -Tr_{\omega L}\left(\Sigma_i^s G_{ij}^{\uparrow} \Sigma_j^s G_{ji}^{\downarrow}\right)$$

$$\Sigma_i^s = \frac{1}{2}\left(\Sigma_i^{\uparrow} - \Sigma_i^{\downarrow}\right)$$

Spin wave spectrum:

$$\omega_{\mathbf{q}} = \frac{4}{M}\sum_j J_{0j}\left(1 - \cos \mathbf{q}\mathbf{R}_j\right) \equiv \frac{4}{M}[J(0) - J(\mathbf{q})]$$

Non-collinear magnetism :



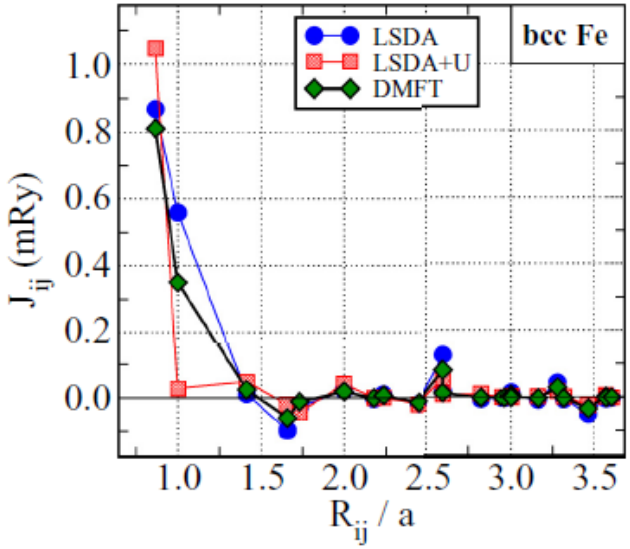
MIK & Lichtenstein Phys. Rev. B 61, 8906 (2000)

Applications

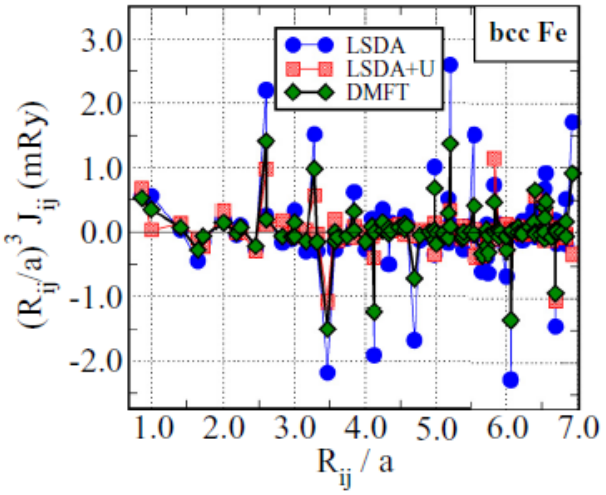
PHYSICAL REVIEW B **91**, 125133 (2015)

Exchange parameters of strongly correlated materials: Extraction from spin-polarized density functional theory plus dynamical mean-field theory

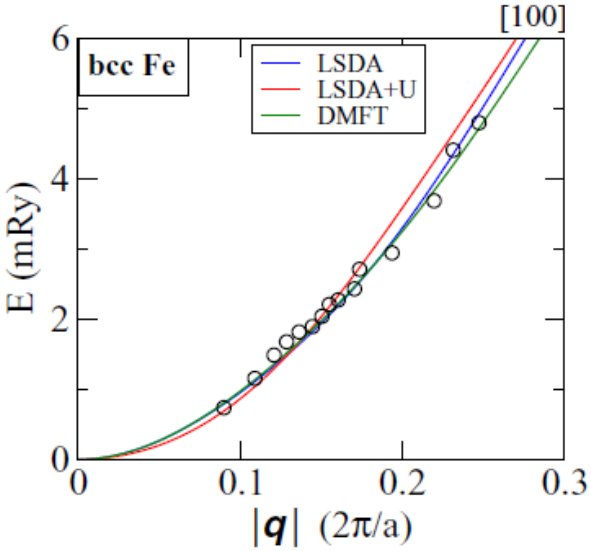
Y. O. Kvashnin,[1] O. Grånäs,[1,2] I. Di Marco,[1] M. I. Katsnelson,[3,4] A. I. Lichtenstein,[4,5] and O. Eriksson[1]



For Fe (and Ni) quite small difference between DFT and DMFT

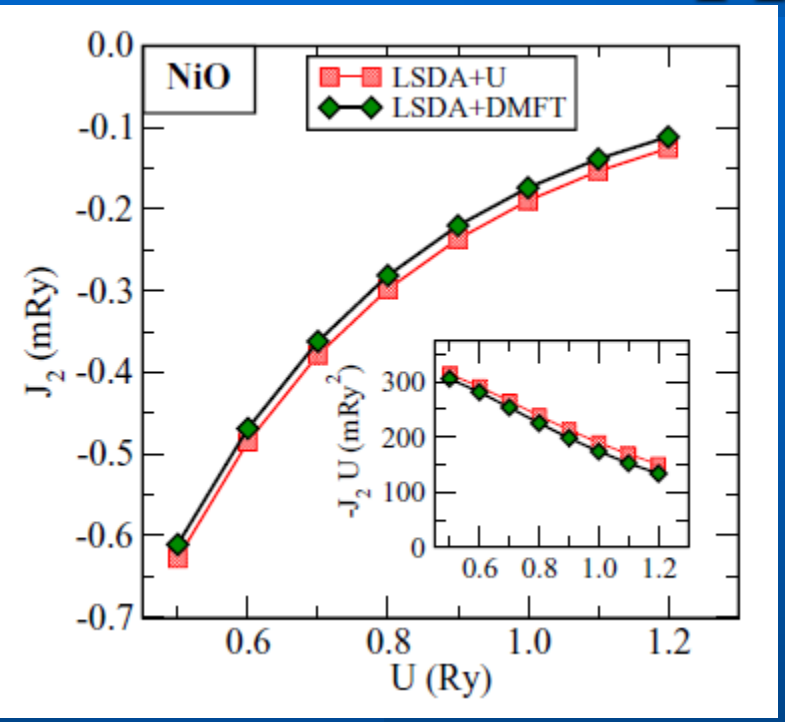


Nontrivial: electronic structure is very different!



Error cancellation?!

Applications II

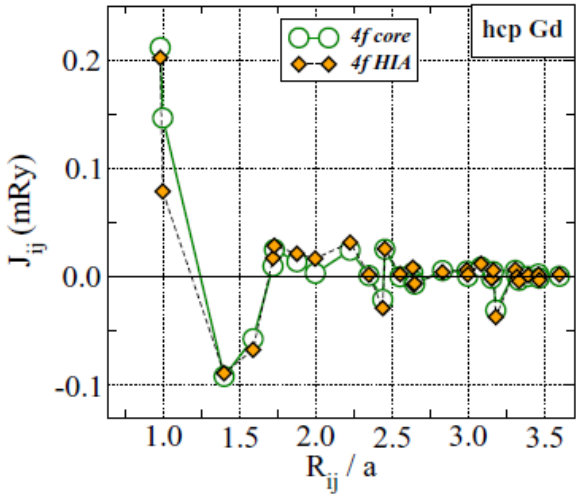


Computational setup	J_1	J_2
LSDA	0.04	−1.58
LSDA + DMFT	−0.003	−0.48
LSDA + U	−0.002	−0.50
LSDA + U (U = 8 eV) (Ref. [42])	0.004/0.0	−0.53
Exp. 1 (Ref. [41])	−0.051	−0.637
Exp. 2 (Ref. [49])	0.051	−0.67

Does not follow a naive formula t^2/U
Difference between Mott and charge transfer insulator

NiO: not too big difference between DMFT and LDA + U

Gd: also, DFT works quite good



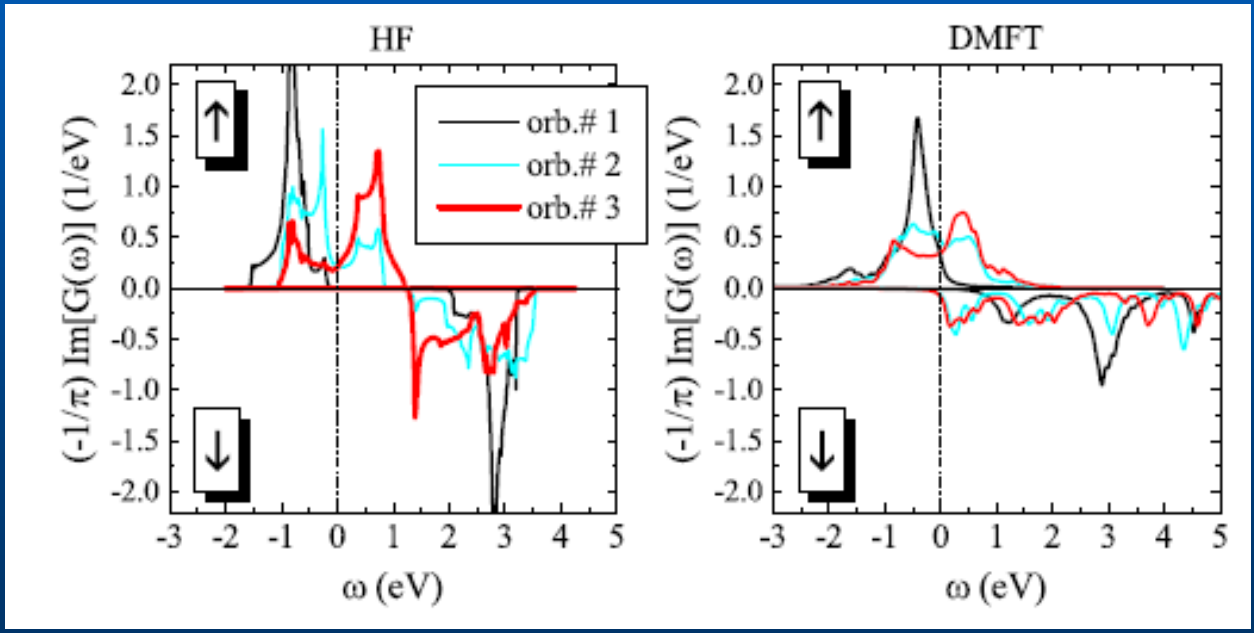
Applications III

PHYSICAL REVIEW B **92**, 144407 (2015)

Mechanisms and origins of half-metallic ferromagnetism in CrO_2

I. V. Solovyev,[1,2,*] I. V. Kashin,[2] and V. V. Mazurenko[2]

Half-metallic FM
DMFT shows
non-quasiparticle
states in the gap
MIK et al, RMP 80, 315 (2008)



Applications IV

Important consequences from DMFT contributions to exchange

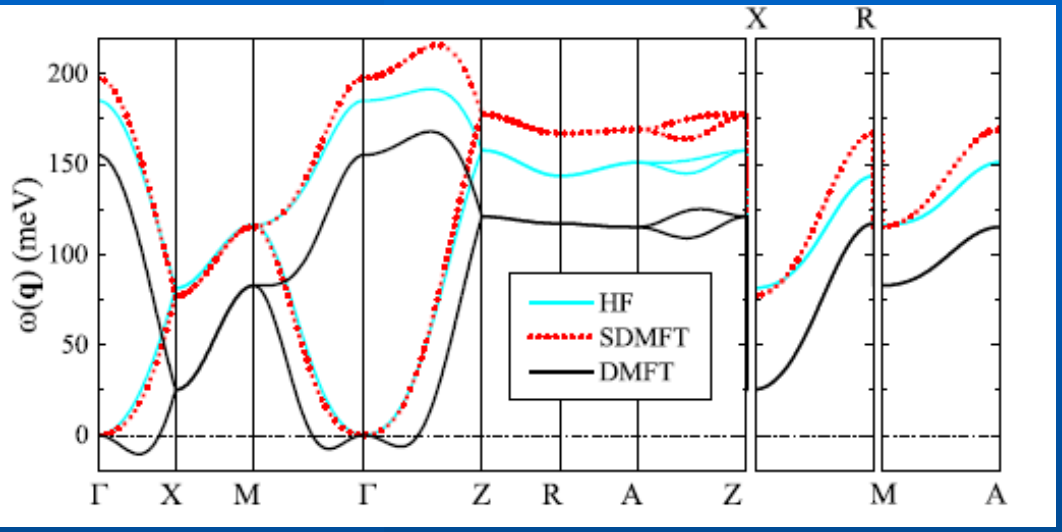
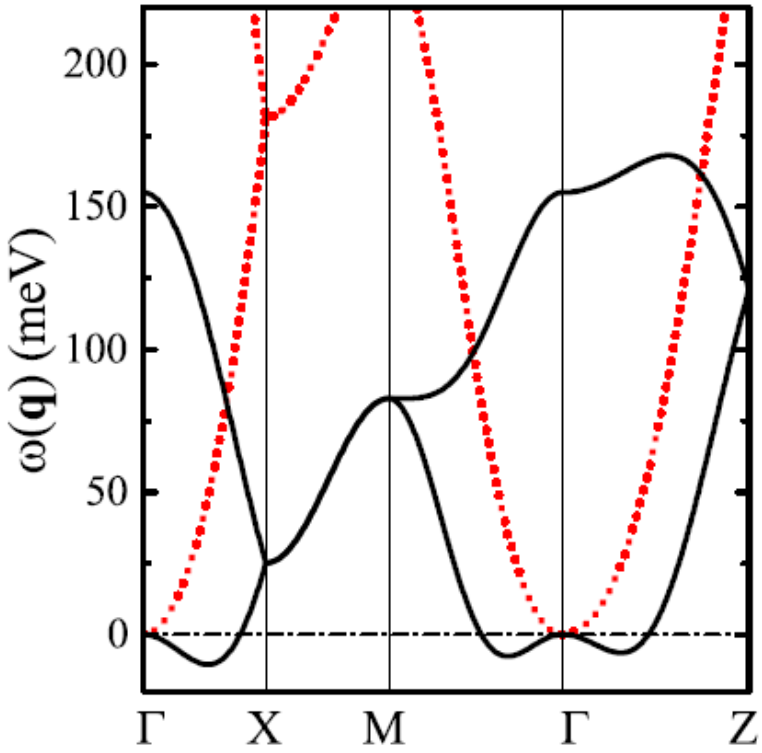


FIG. 9. (Color online) Results of calculations of the spin-wave dispersion with the DMFT parameters obtained for the isolated t_{2g} band (solid line) and after taking into account the additional FM contribution $\Delta J_2 = 17.81$ meV, arising from magnetic polarization of the oxygen band and direct exchange interactions in the t_{2g} band (dotted line). Notations of the high-symmetry points of the BZ are taken from [55].

Direct exchange also plays an important role

Without magnetic polarization of oxygen FM state is unstable within DMFT (but not in simpler approaches)



Dzialoshinskii-Moriya interactions

MIK, Kvashnin, Mazurenko & Lichtenstein, PRB 82, 100403 (2010)

LDA+U

$$\begin{aligned}\hat{H} &= \hat{H}_t + \hat{H}_u \\ &= \sum_{12} c_1^+ t_{12} c_2 + \frac{1}{2}\sum_{1234} c_1^+ c_2^+ U_{1234} c_3 c_4\end{aligned}$$

DM interactions (weak FM, etc.)

$$H_{DM} = \sum_{ij} \vec{D}_{ij}[\vec{e}_i \times \vec{e}_j]$$

Small rotations

$$\hat{R}_i = e^{i\delta\vec{\varphi}_i\vec{J}}$$

$$\hat{\vec{J}} = \hat{\vec{L}} + \hat{\vec{S}}$$

Dzialoshinskii-Moriya interactions II

Starting from collinear configuration

$$\delta\hat{H}_t = \sum_{ij} c_i^+ (\delta\hat{R}_i^+ \hat{t}_{ij} + \hat{t}_{ij}\delta\hat{R}_j) c_j$$

$$= -i \sum_{ij} c_i^+ (\delta\vec{\varphi}_i \hat{\vec{J}}\hat{t}_{ij} - \hat{t}_{ij}\hat{\vec{J}}\delta\vec{\varphi}_j) c_j$$

$$= -\frac{i}{2} \sum_{ij} c_i^+ (\delta\vec{\varphi}_i - \delta\vec{\varphi}_j)(\hat{\vec{J}}\hat{t}_{ij} + \hat{t}_{ij}\hat{\vec{J}}) c_j$$

$$\vec{D}_{ij} = -\frac{i}{2} Tr_{m,\sigma} \langle c_i^+ [\hat{\vec{J}}, \hat{t}_{ij}]_+ c_j \rangle = -\frac{i}{2} Tr_{m,\sigma} N_{ji} [\hat{\vec{J}}, \hat{t}_{ij}]_+$$

$$N_{ji} = \langle c_i^+ c_j \rangle = -\frac{1}{\pi} \int_{-\infty}^{E_f} ImG_{ji}(E) dE$$

$FeBO_3$

LETTERS
PUBLISHED ONLINE: 9 FEBRUARY 2014 | DOI: 10.1038/NPHYS2859
nature physics

Measuring the Dzyaloshinskii–Moriya interaction in a weak ferromagnet

V. E. Dmitrienko[1], E. N. Ovchinnikova[2], S. P. Collins[3*], G. Nisbet[3], G. Beutier[4], Y. O. Kvashnin[5], V. V. Mazurenko[6], A. I. Lichtenstein[7] and M. I. Katsnelson[6,8]

A novel exper. technique to measure DM vector and not only canting angle (resonant X-ray scattering)

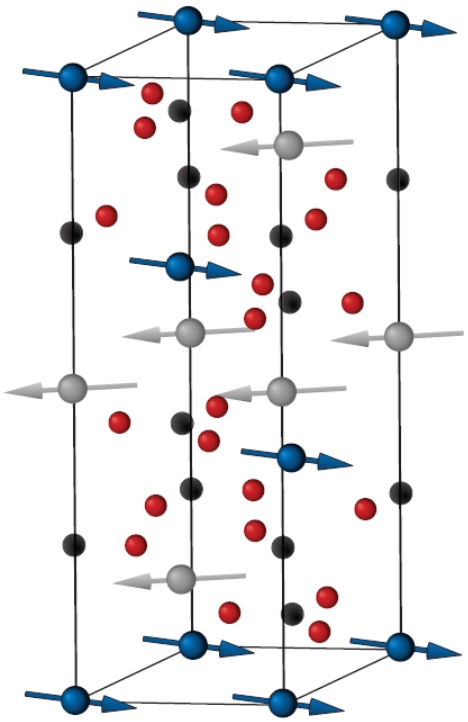


TABLE I. Calculated values of isotropic exchange interactions between magnetic moments in $FeBO_3$ (in meV). The number in parentheses denotes the coordination sphere.

$Fe^{(1)}$	$Fe^{(2)}$	$Fe^{(3)}$	$Fe^{(4)}$	$Fe^{(5)}$	$Fe^{(6)}$	$Fe^{(7)}$
10.28	0.21	0	0.54	-0.08	0	0.02

TABLE III. Parameters of Dzyaloshinskii-Moriya interaction (in meV) calculated by using Eq. (6).

Bond $m-n$	$\mathbf{R}_{mn}$	$\mathbf{D}_{mn}$ (meV)
0-1	(1.0 ; 0.0 ; -0.904)	(-0.25; 0.0; -0.24)
0-2	(-0.5 ; $-\sqrt{3}/2$; -0.904)	(0.12 ; 0.22 ; -0.24)
0-3	(-0.5 ; $\sqrt{3}/2$; -0.904)	(0.12 ; -0.22 ; -0.24)
0-4	(-1.0 ; 0.0 ; 0.904)	(-0.25; 0.0 ; -0.24)
0-5	(0.5 ; $-\sqrt{3}/2$; 0.904)	(0.12 ; -0.22 ; -0.24)
0-6	(0.5 ; $\sqrt{3}/2$; 0.904)	(0.12 ; 0.22 ; -0.24)

Agrees well with exper.

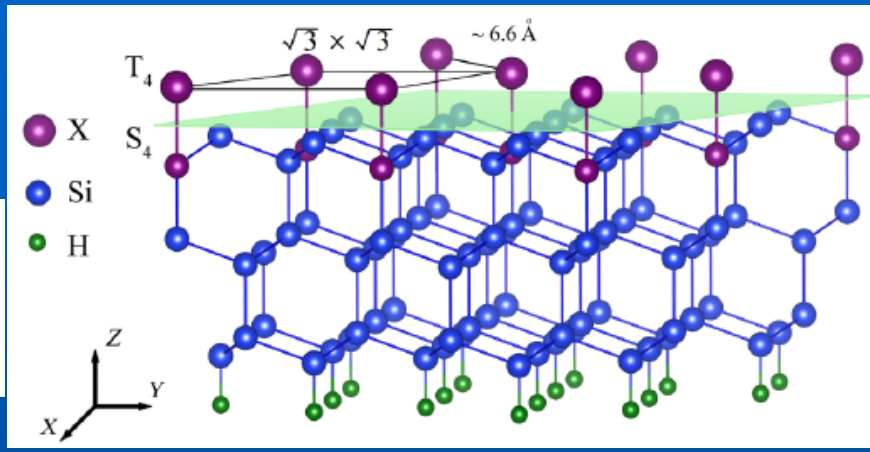
Si(111):X (X=C,Si,Sn,Pb)

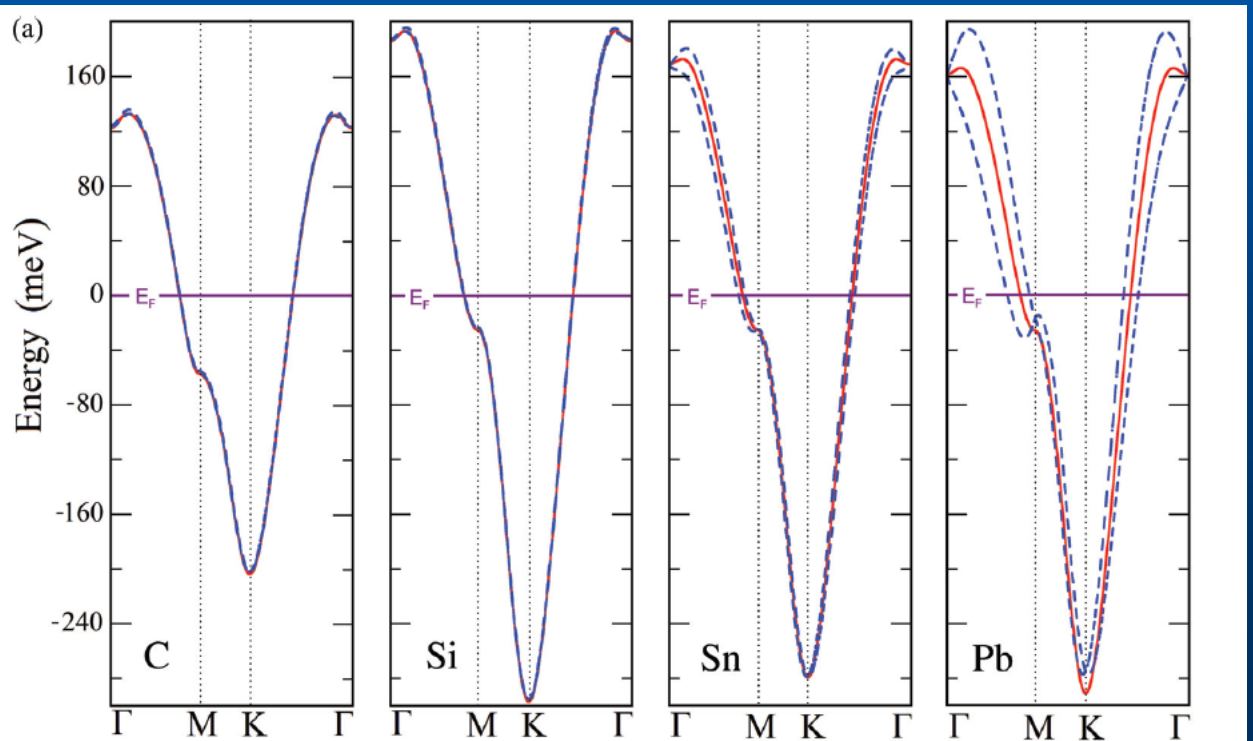
sp-electron magnets

PHYSICAL REVIEW B **94**, 224418 (2016)

Spin-orbit coupling and magnetic interactions in Si(111):{C,Si,Sn,Pb}

D. I. Badrtdinov,[1] S. A. Nikolaev,[1] M. I. Katsnelson,[1,2] and V. V. Mazurenko[1]



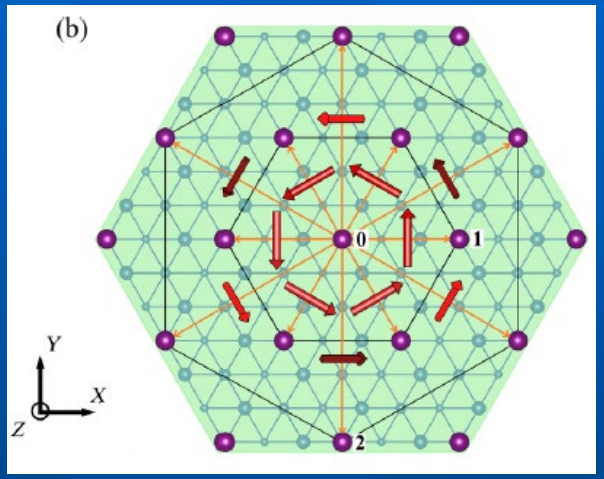


Single narrow band nea the Fermi energy

Red – without SO
Blue – with SO

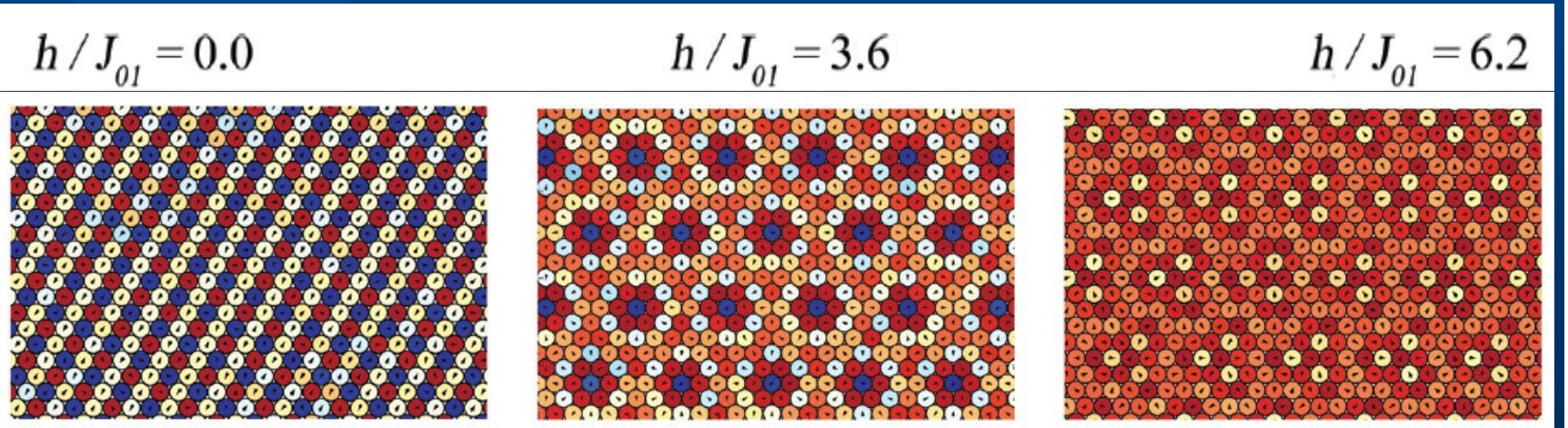
Si(111):X (X=C,Si,Sn,Pb) II

Mott insulator if take into account Hubbard U



Orientation of DMI

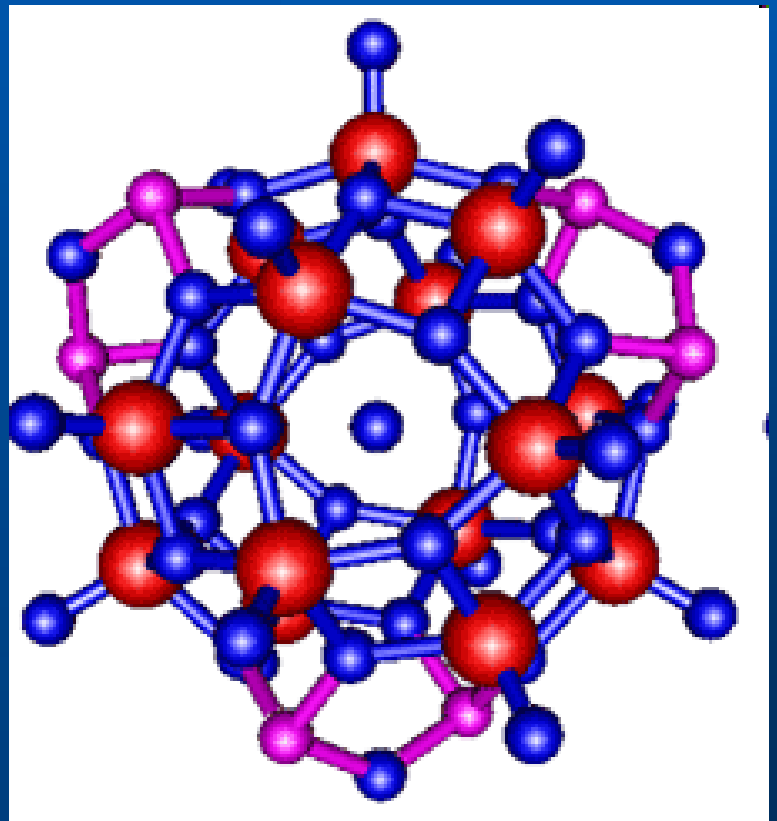
Ground state magnetic configurations for Si(111):Pb in magnetic field (MC simulations)

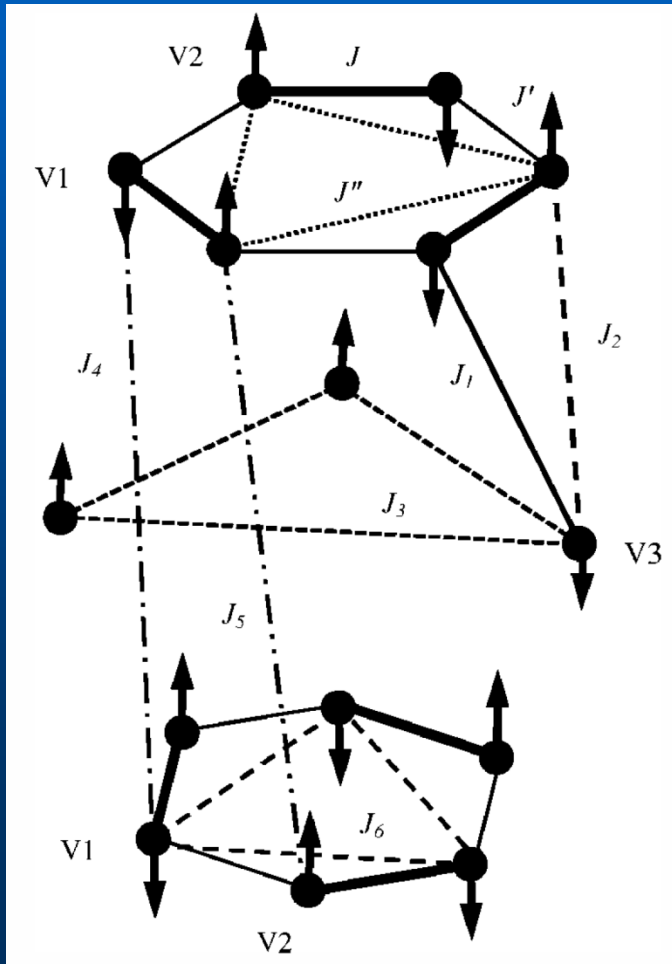


Molecular magnets

Example: V_{15} AFM ground state *S = 1/2*

$$V_{15}(K_6[V_{15}As_6O_{42}(H_2O)]\cdot 8H_2O)$$





LDA+U calculations

PHYSICAL REVIEW B **70**, 054417 (2004)

Electronic structure and exchange interactions in V_{15} magnetic molecules: LDA+U results

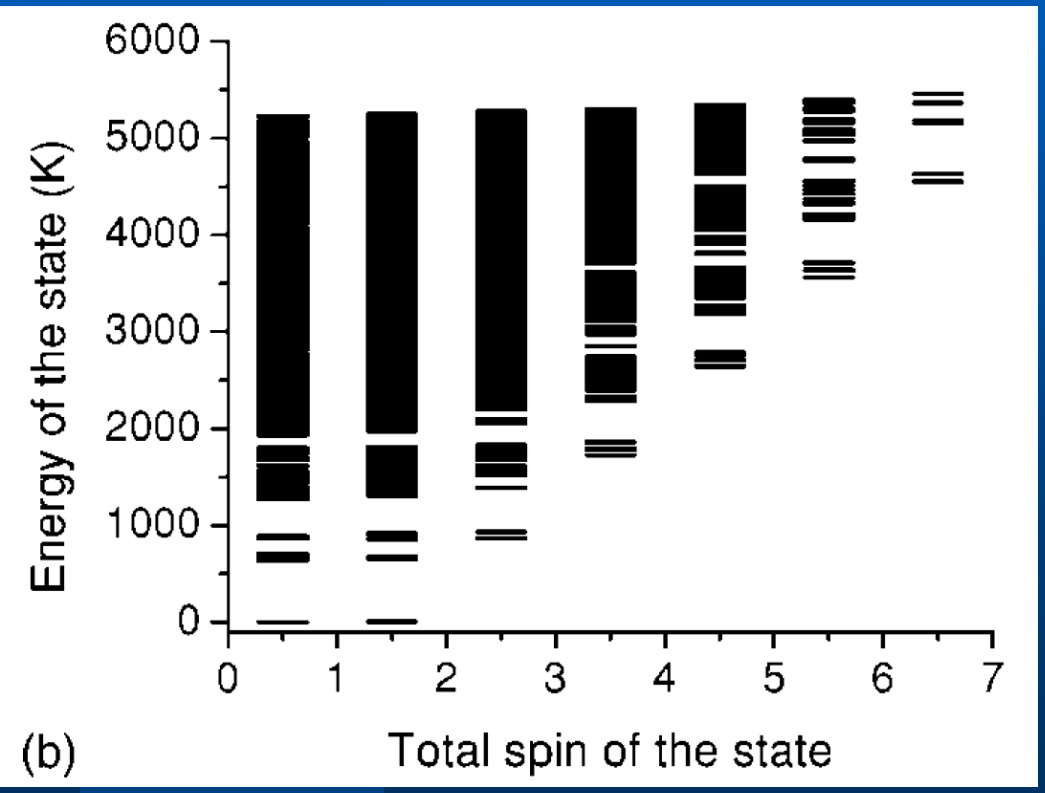
D. W. Boukhvalov,[1,2] V. V. Dobrovitski,[3] M. I. Katsnelson,[2,4] A. I. Lichtenstein,[5] B. N. Harmon,[3] and P. Kögerler[3]

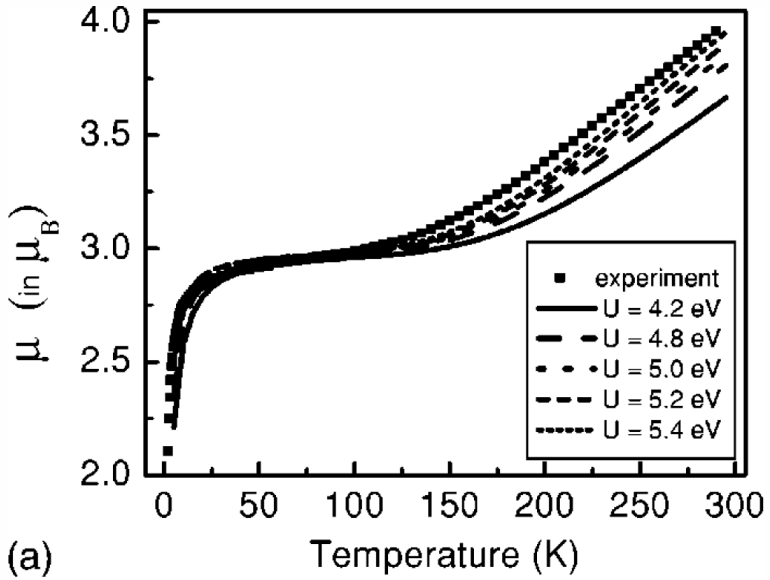
TABLE II. The exchange parameters (in Kelvin), electronic gap, and the magnetic moments of V ions for different magnetic structures of V_{15}. The calculations have been made for U=4 eV, J=0.8 eV.

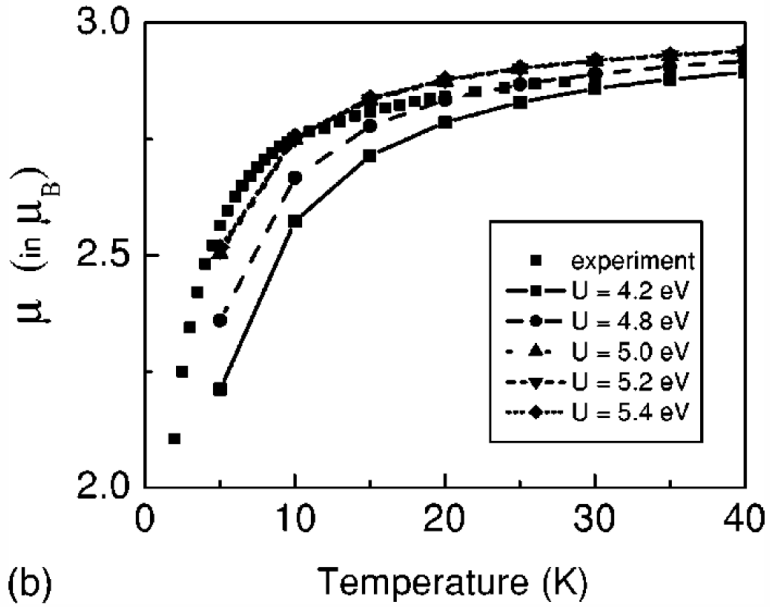
parameter	AFM1	AFM2	FM
J	−910	−905	−942
J'	−45	−46	−53
J''	−136	−139	−156
J_1	−219	−247	−255
J_2	−134	−128	−132
J_3	−5	−5	−6
J_4	−13	−12	−15
J_5	−3	−3	−3
J_6	−3	−3	−3
gap	1.08	1.02	1.16
μ_{V1}	−0.94	−0.93	−0.99
μ_{V2}	+0.91	+0.92	−0.97
μ_{V3}	−1.00	+0.97	−1.00

LDA+U calculations II

Exact diagonalization for Heisenberg model







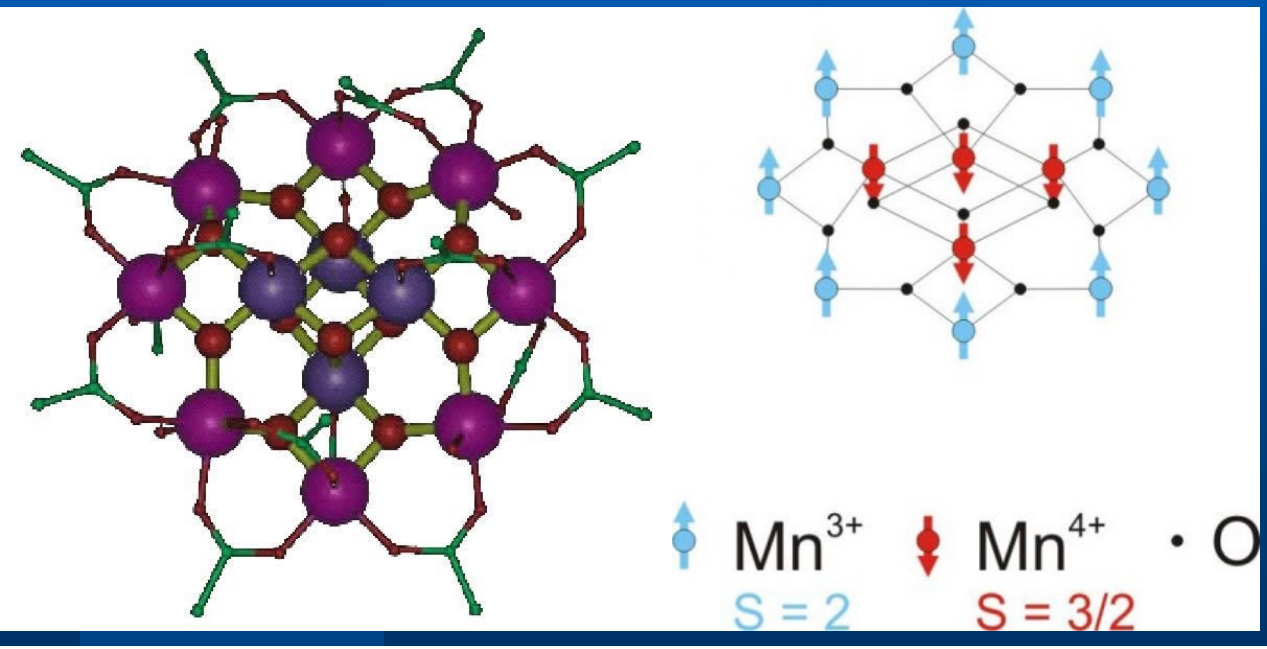
Mn_{12}: full calculations

PHYSICAL REVIEW B **00**, 004400 (2014)

First-principles modeling of magnetic excitations in Mn_{12}

V. V. Mazurenko,[1] Y. O. Kvashnin,[2,3] Fengping Jin,[4] H. A. De Raedt,[5] A. I. Lichtenstein,[6] and M. I. Katsnelson[1,7]

Motivation



The prototype molecular magnet

Dimension of Hilbert space:
$(2^x2+1)^8(2^x3/2+1)^4=10^8$

A real challenge!

$$[Mn_{12}O_{12}(CH_3COO)_{16}(H_2O)_4]\cdot 2CH_3COOH\cdot 4H_2O$$

Mn_{12}: full calculations II

Inelastic netron scattering data: cannot be explained without strong DM interactions (MIK, Dobrovistki & Harmon, PRB 1999)

Eight-spin model: S = ½ dimers from S=2 and S=3/2
Dimensionality of Hilbert space decreases to 10^4
Cannot be justified quantitatively!

Full LDA+U calculations plus Lanczos ED

$$\hat{H} = \sum_{ij} J_{ij} \hat{S}_i \hat{S}_j + \sum_{i\mu\nu} \hat{S}_i^{\mu} A_i^{\mu\nu} \hat{S}_i^{\nu} + \sum_{ij} \vec{D}_{ij} [\hat{S}_i \times \hat{S}_j]$$

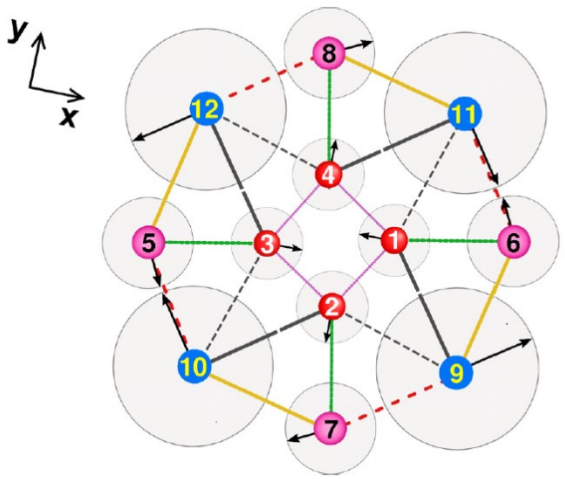


TABLE I. Intramolecular isotropic exchange interaction parameters (in meV) calculated by using the LDA + U approach. Positive sign corresponds to the antiferromagnetic coupling.

Bond (i, j)	1–6	1–11	1–9	6–9	7–9	1–4	1–3
J_{ij} (this work)	4.6	1.0	1.7	−0.45	−0.37	−1.55	−0.5
J_{ij} (Ref. [4])	4.8	1.37	1.37	−0.5	−0.5	−1.6	−0.7
J_{ij} (Ref. [26])	7.4	1.72	1.72			−1.98	

Mn_{12}: full calculations III

TABLE II. Intramolecular anisotropic exchange interaction parameters calculated by using the LDA + U approach. $\vec{R}_{ij}$ is a radius vector connecting ith and jth atoms (in units of $a = 17.31$ Å).

Bond (i, j)	$\vec{R}_{ij}$	$\vec{D}_{ij}$ (meV)
2–7	(0.03; −0.16; 0.0)	(−0.008; −0.013; −0.002)
4–8	(−0.03; 0.16; 0.0)	(0.008; 0.013; −0.002)
1–6	(0.16; 0.03; 0.0)	(−0.013; 0.008; −0.002)
3–5	(−0.16; −0.03; 0.0)	(0.013; −0.008; −0.002)
1–11	(0.06; 0.18; 0.07)	(−0.020; 0.03; −0.055)
3–10	(−0.06; −0.18; 0.07)	(0.020; −0.03; −0.055)
2–9	(0.18; −0.06; −0.07)	(−0.03; −0.020; −0.055)
4–12	(−0.18; 0.06; −0.07)	(0.03; 0.020; −0.055)
1–9	(0.11; −0.16; 0.04)	(0.020; 0.014; 0.03)
3–12	(−0.11; 0.16; 0.04)	(−0.020; −0.014; 0.03)
2–10	(−0.16; −0.11; −0.04)	(−0.014; 0.020; 0.03)
4–11	(0.16; 0.11; −0.04)	(0.014; −0.020; 0.03)
6–9	(−0.04; −0.18; 0.04)	(−0.006; −0.004; −0.012)
5–12	(0.04; 0.18; 0.04)	(0.006; 0.004; −0.012)
7–10	(−0.18; 0.04; −0.04)	(0.004; −0.006; −0.012)
8–11	(0.18; −0.04; −0.04)	(−0.004; 0.006; −0.012)
7–9	(0.15; 0.1; −0.07)	(0.020; −0.004; 0.012)
8–12	(−0.15; −0.1; −0.07)	(−0.020; 0.004; 0.012)
6–11	(−0.1; 0.15; 0.07)	(−0.004; −0.020; 0.012)
5–10	(0.1; −0.15; 0.07)	(0.004; 0.020; 0.012)
4–1	(−0.10; 0.06; 0.11)	(−0.014; 0.005; −0.013)
1–2	(−0.06; −0.10; 0.11)	(−0.005; −0.014; −0.013)
3–4	(0.07; 0.1; 0.11)	(0.005; 0.014; −0.013)
2–3	(−0.10; 0.07; −0.11)	(0.014; −0.005; −0.013)
1–3	(−0.16; −0.03; 0.0)	(−0.006; 0.030; 0)
2–4	(−0.04; 0.17; 0.0)	(−0.030; −0.006; 0)

Plus anisotropy tensors...

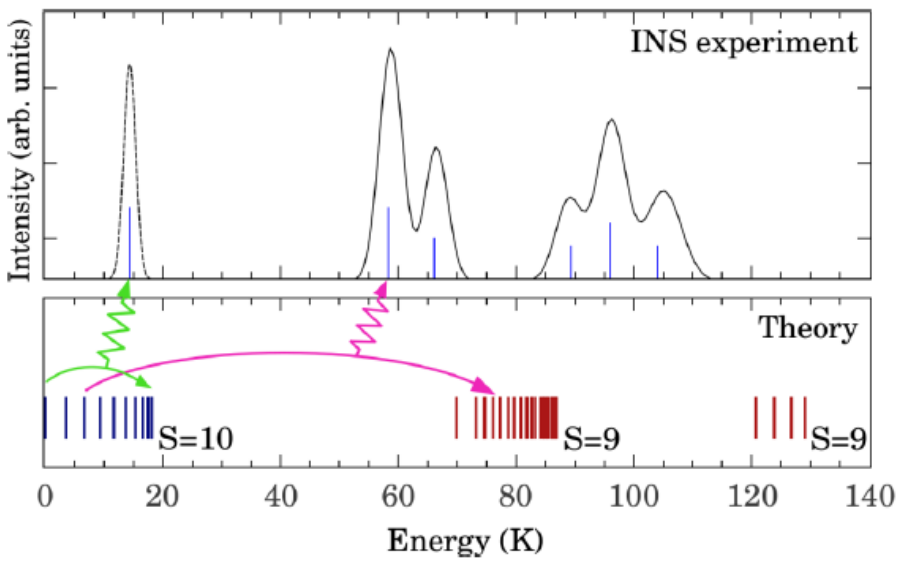


FIG. 2. (Color online) Schematic comparison of the theoretical spectrum obtained by diagonalizing Eq. (1) and INS spectrum taken from Ref. [12] (Figs. 6 and 8 therein). The arrows denote the intra- and interband transitions that correspond to the excitations observed in the INS experiment.

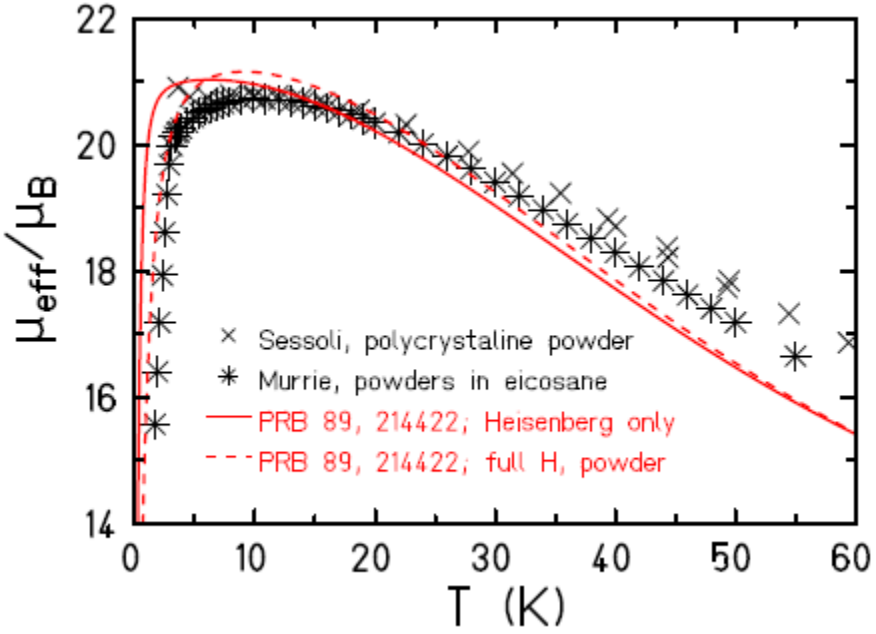
No fitting parameters at all – not so bad!

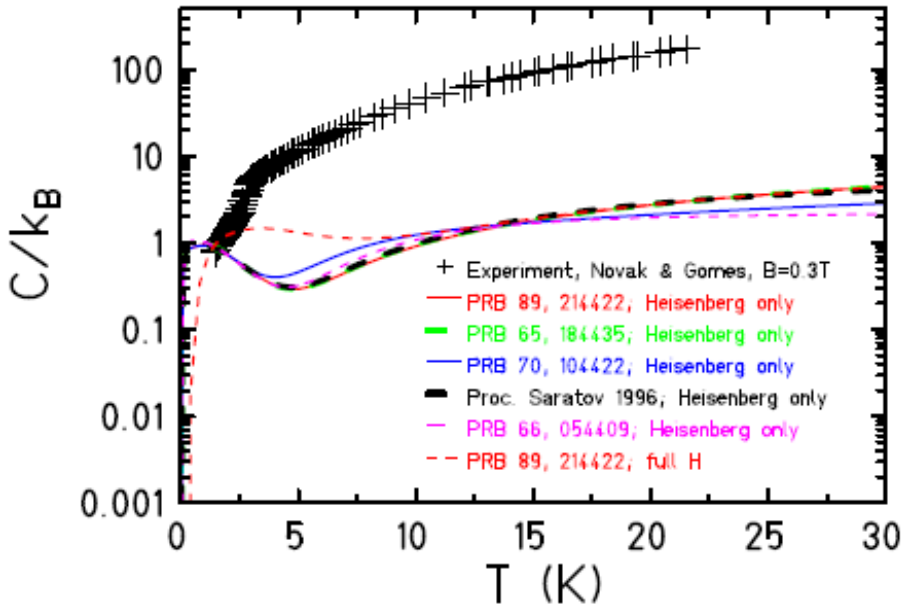
Mn_{12}: full calculations IV

PHYSICAL REVIEW B **92**, 064424 (2015)

Thermodynamic observables of Mn_{12}-acetate calculated for the full spin Hamiltonian

Oliver Hanebaum and Jürgen Schnack*





Also, thermodynamic quantities can be calculated

Beyond the talk

Finite-temperature effects

Ab initio spin dynamics for real systems

Intermediate level: TB spin dynamics

And many, many specific applications to real materials

Collaboration

Recent:

A. Lichtenstein and S. Brener (Hamburg)
A. Secchi and A. Rudenko (Nijmegen)
V. Mazurenko (Ekaterinburg)
Ya. Kvashnin and O. Eriksson (Uppsala)

and many other people involved in development of the formalism and calculations for specific materials in 1987-2013, esp. V. Antropov (Ames) and D. Boukhvalov (Seoul)

Thank you for your attention