Radboud Universiteit

Theory of magnetic interactions in real materials

Mikhail Katsnelson

IMM

Institute for Molecules and Materials

Outline

1. Introduction
2. Exchange interactions from first principles
3. Beyond DFT: correlated systems and LDA+DMFT
4. Applications: **Fe, Gd, NiO, CrO_2**
5. Dzyaloshinskii-Moriya interactions
6. Applications: **molecular magnets, adatoms on Si(111) surfaces…**
7. Manipulation of magnetic interactions by high-frequency laser fields

Epigraphs

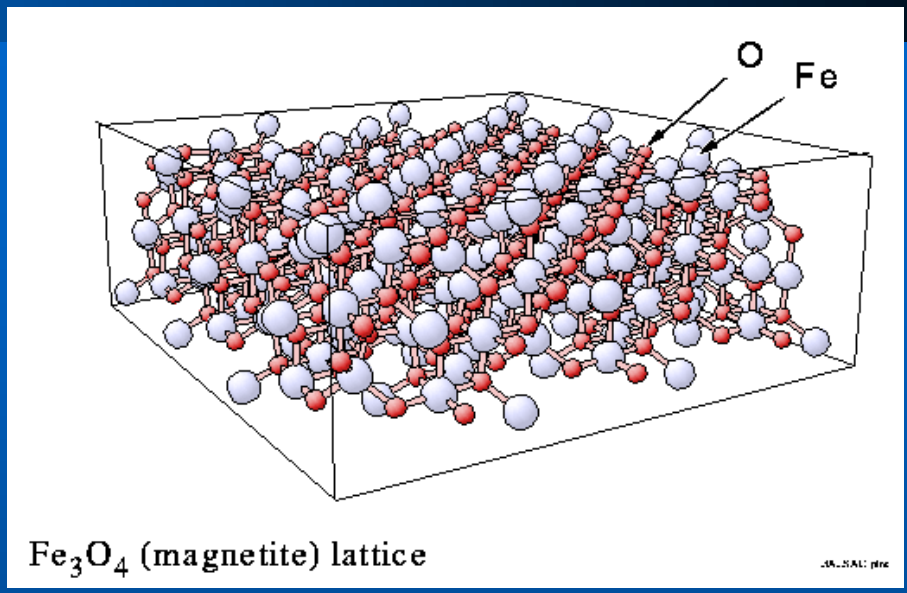
To the theoretical physicists, ferromagnetism presents a number of very interesting, unsolved and beautiful challenges

(Feynman Lectures on Physics)

Make things as simple as possible but not simpler

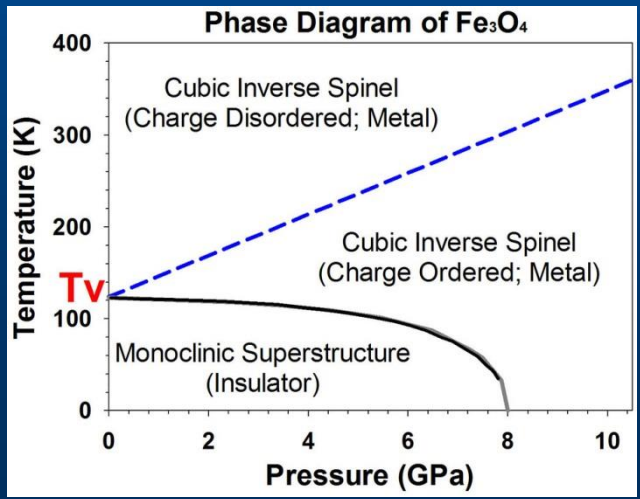
(A. Einstein)

Magnetite – first known magnet



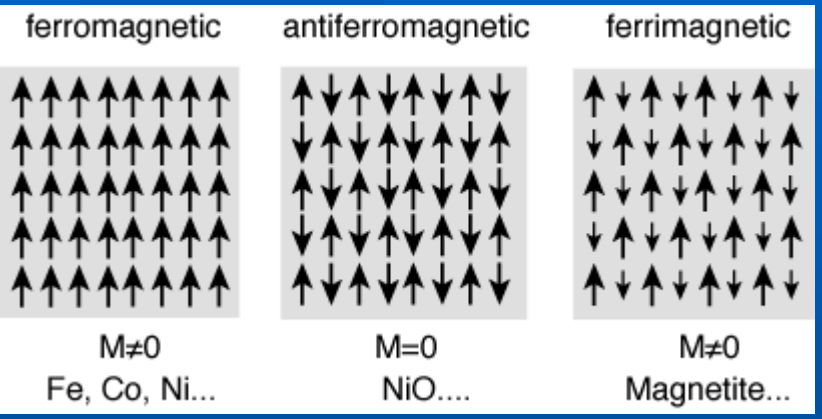
Very complicated structure, still a lot of open questions

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



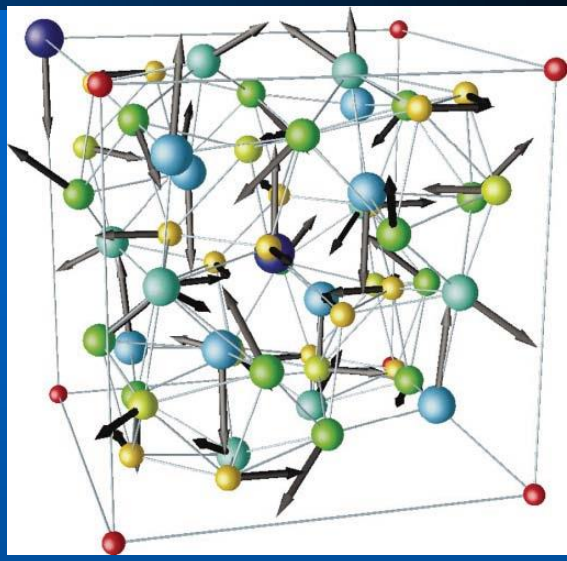
Types of magnetic ordering

Textbook wisdom

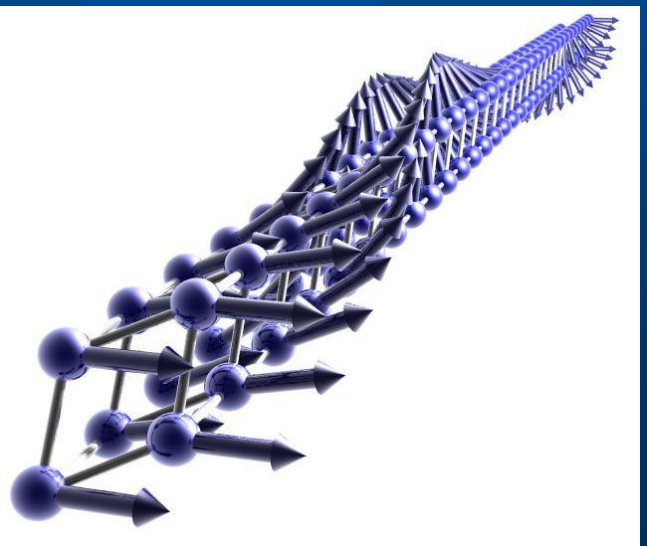


Sometimes very complicated

α-Mn

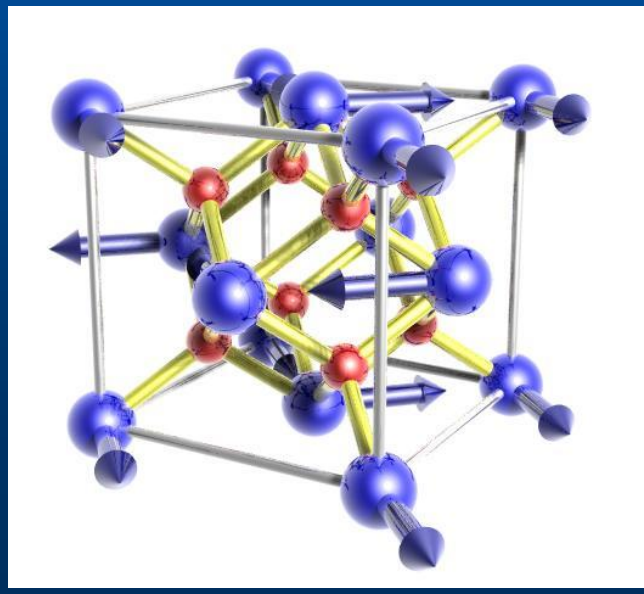


Spin spirals

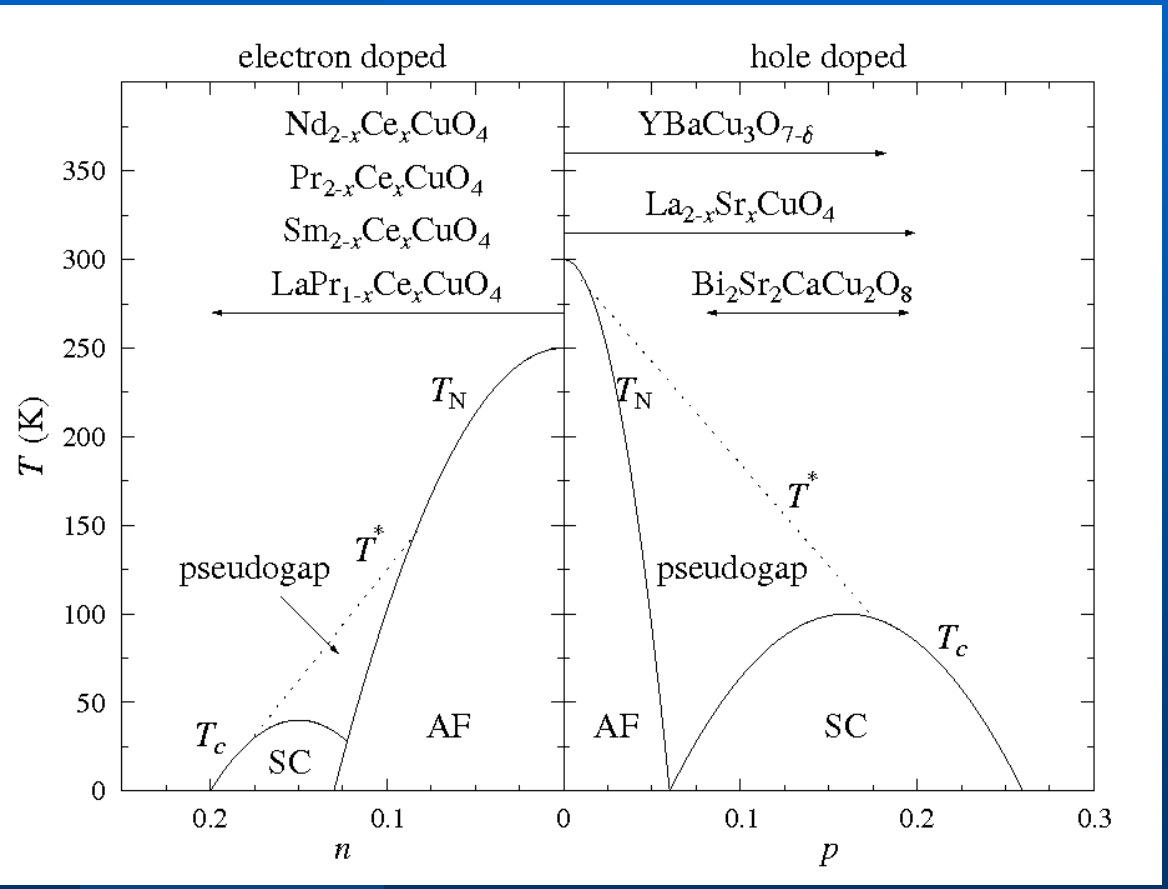


γ-Fe

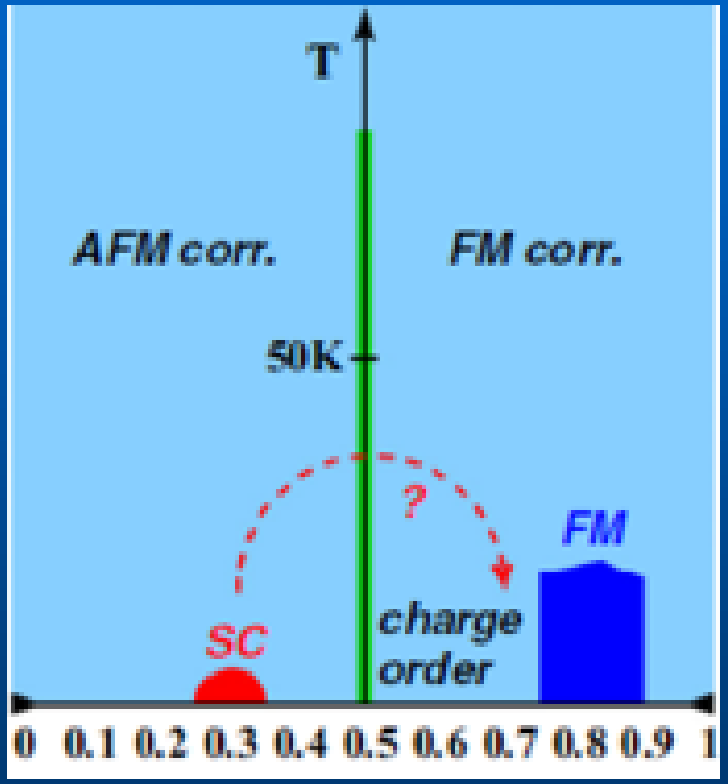
UO_2



Relation to superconductivity and other phenomena

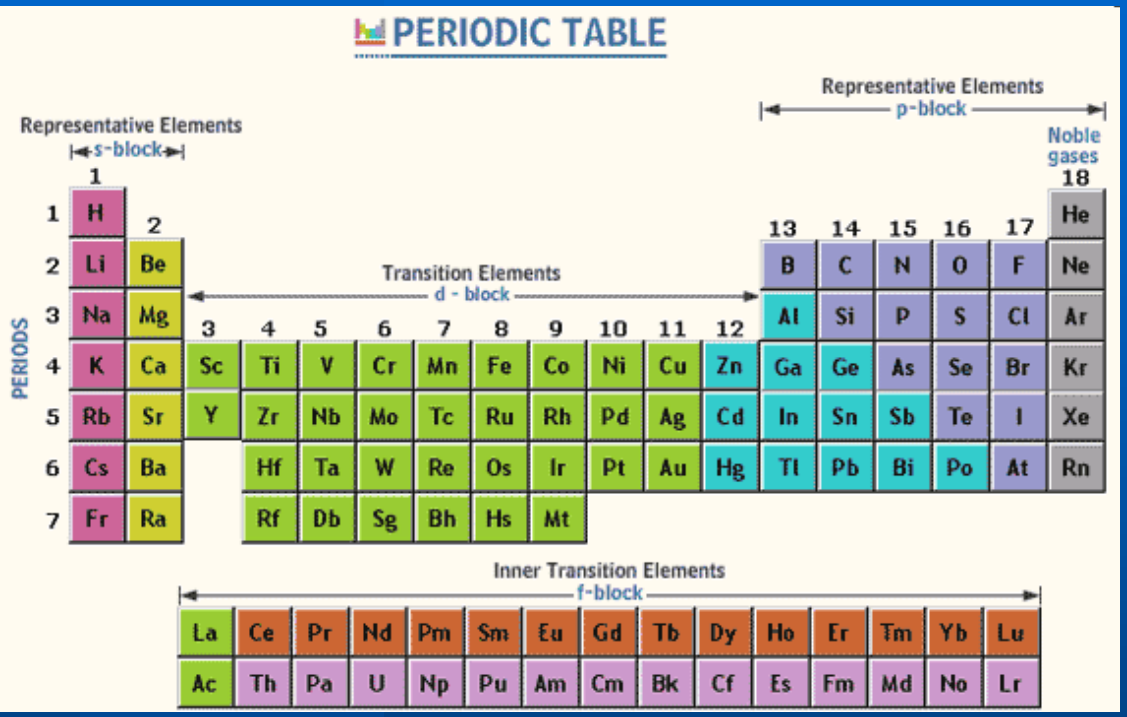


Simplified phase diagram of Cu-O high-Tc superconductors



Layered cobaltates Na_xCoO_2

Ferromagnetism of Fe, Co, Ni



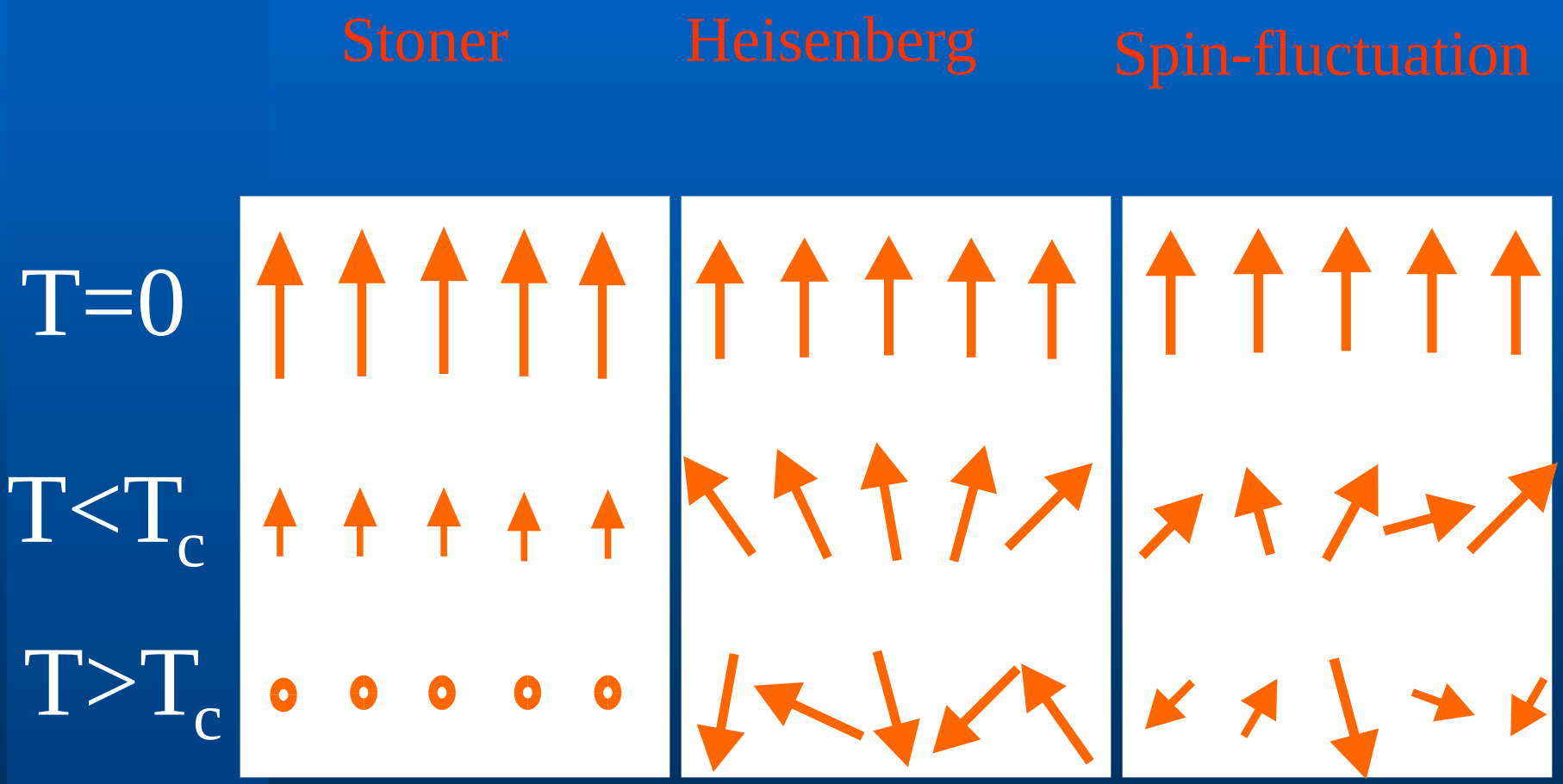
Ferromagnetism of iron is known from ancient times

Iron

Cobalt

Nickel

Itinerant-electron ferromagnetism at finite temperatures



Stoner criterion

$$I_{\mathrm{eff}} N(E_{\mathrm{F}}) > 1$$

$N(E_{\mathrm{F}})$ is the density of one-electron states

I_{eff} is an on-site interaction parameter

Stoner parameter ≈ 0.9 eV for all 3d metals; DOS is crucially important

Equation for the Curie temperature:

f(E) Fermi function

$$I_{\mathrm{eff}} \int \mathrm{d}E \left(-\frac{\partial f}{\partial E} \right) N(E) = 1$$

If Fe would be Stoner magnet it would have T_C ≈ 4000 K (in reality 1043 K)

In reality, T_c is determined by spin fluctuations,
That is, exchange parameters

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.

Density Functional Theory

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

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

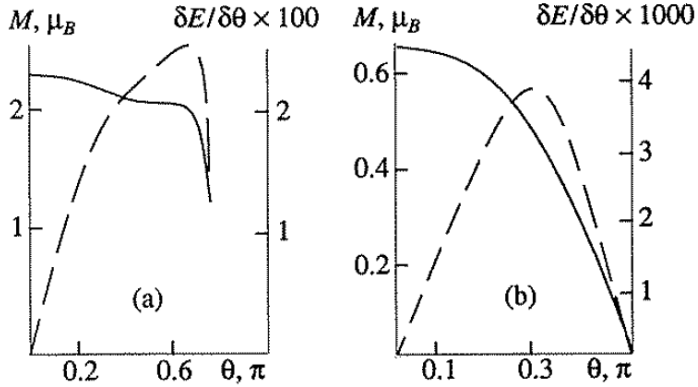


Fig. 4.4. *Magnetic moment in Bohr magnetons (the full curve) and the first derivative of energy with respect to angle of rotation in Ry (the dashed curve) according to calculations in [168]: (a) Fe, (b) Ni.*

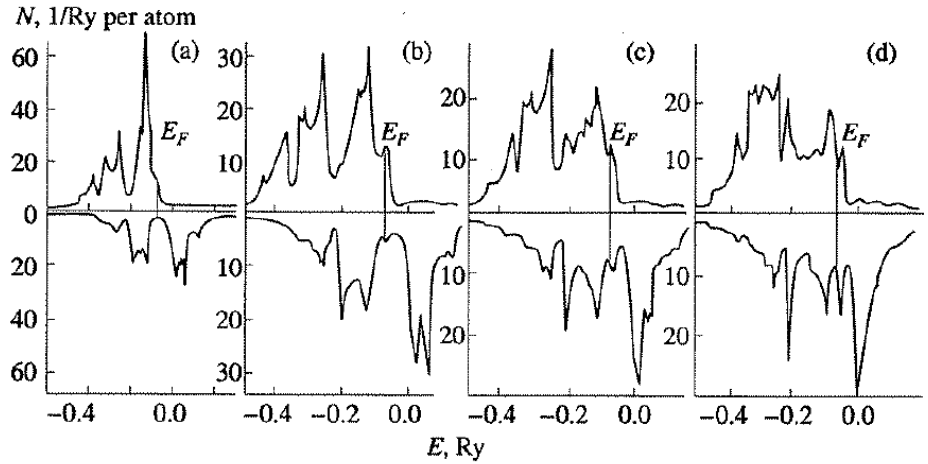


Fig. 4.5. *Electronic density of states for an Fe atom in a metal with the magnetic moment turned through θ = 0 (a), θ = 0.2π (b), θ = 0.35π (c), and θ = 0.5π (d).*

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

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

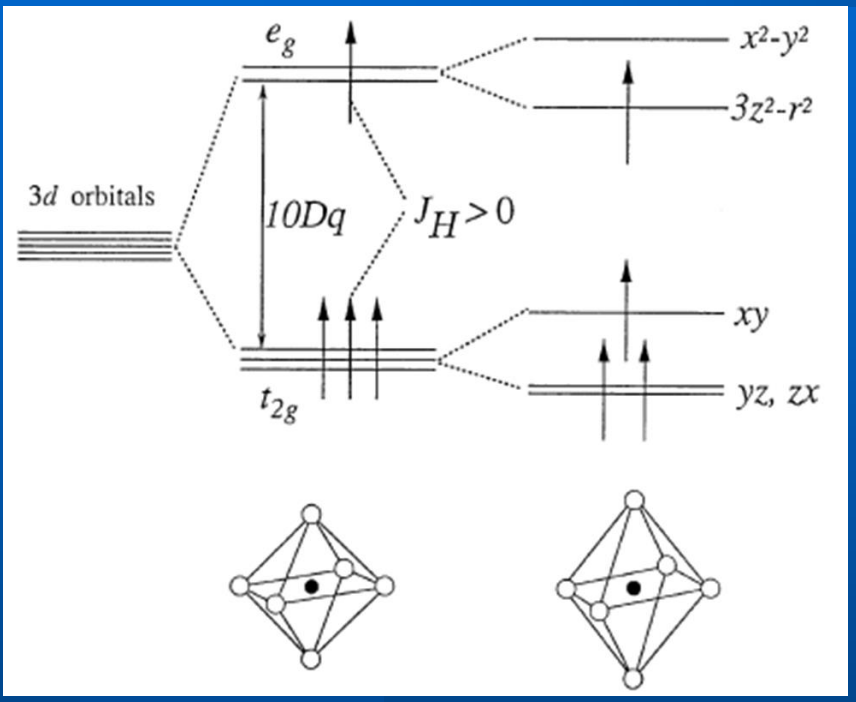
Stiffness constants for Fe and Ni

(in meV/Å^2)

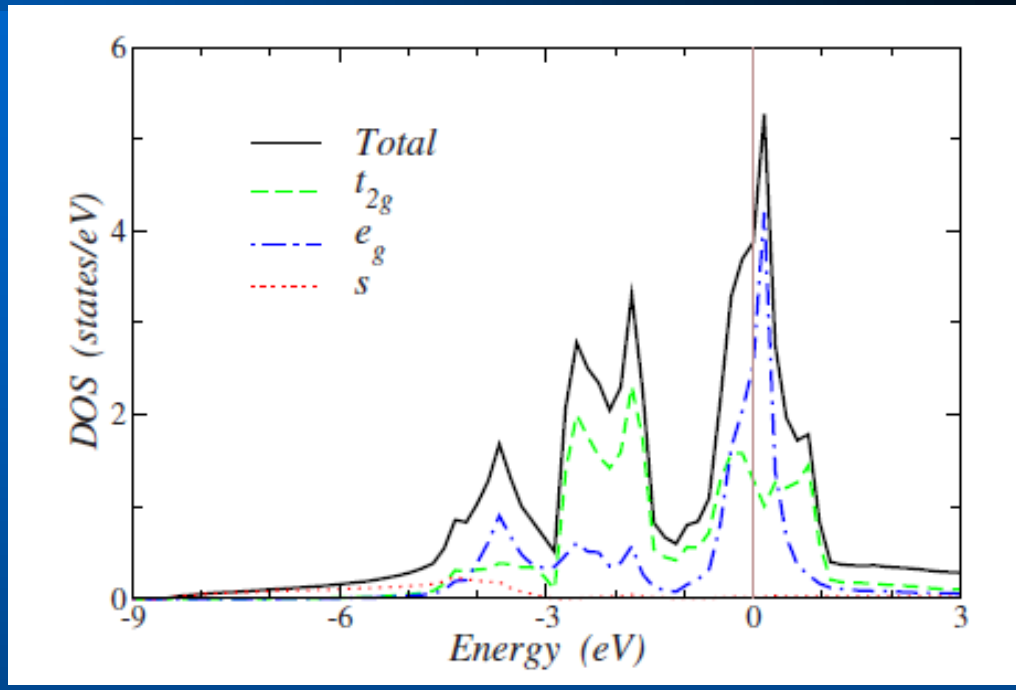
Fe: LSDA	*239*
with gradient corrections	*251*
experiment	*280 - 310*
Ni: LSDA	*692*
with gradient corrections	*735*
experiment	*550-630*

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]

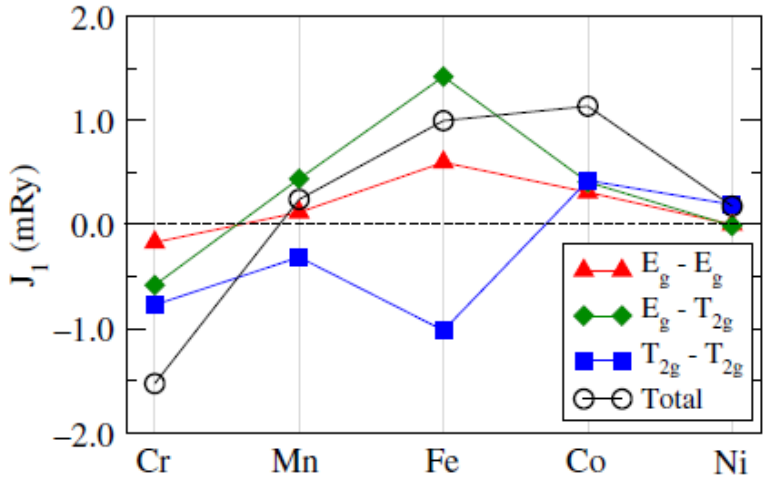
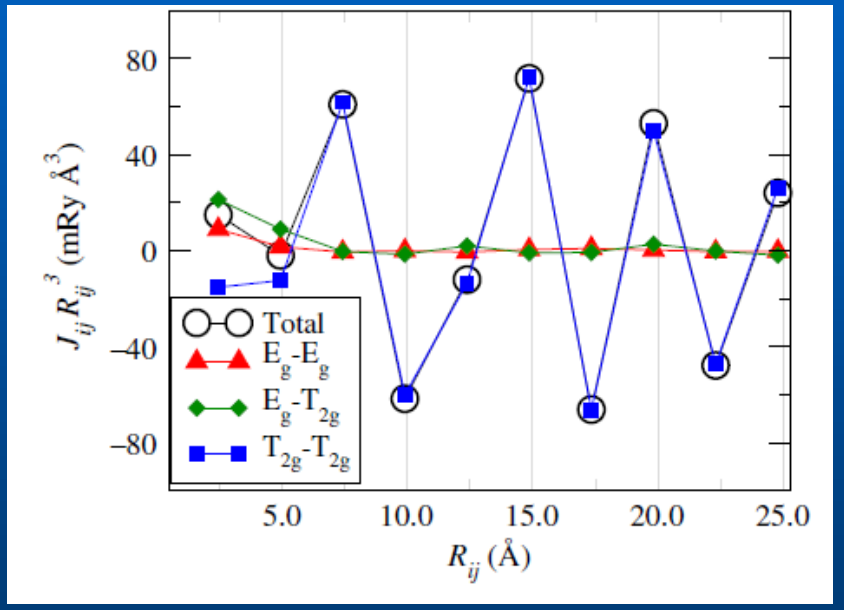
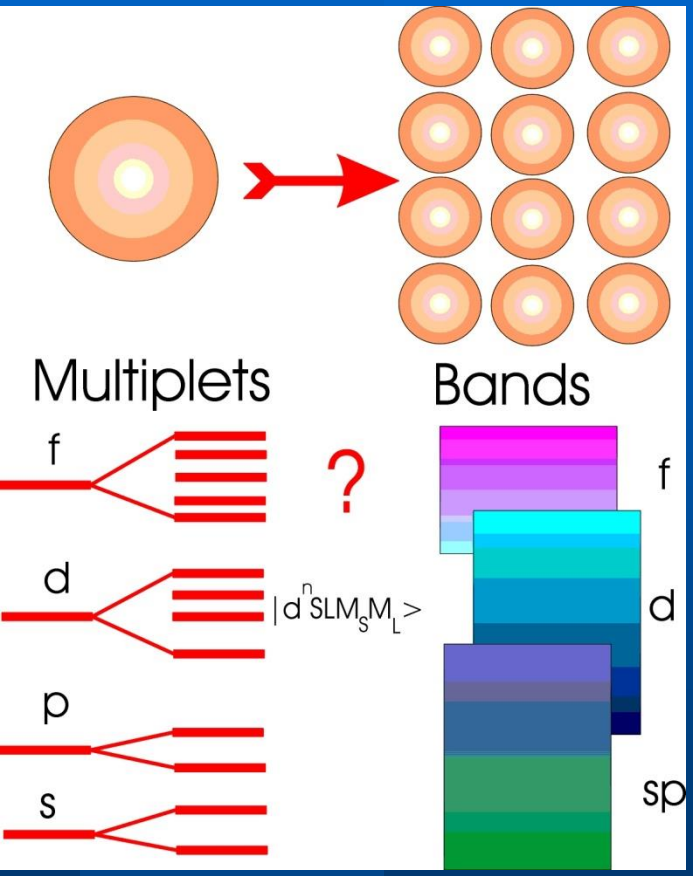


FIG. 1. Orbitally decomposed NN exchange interaction in elemental 3*d* metals in the bcc structure.



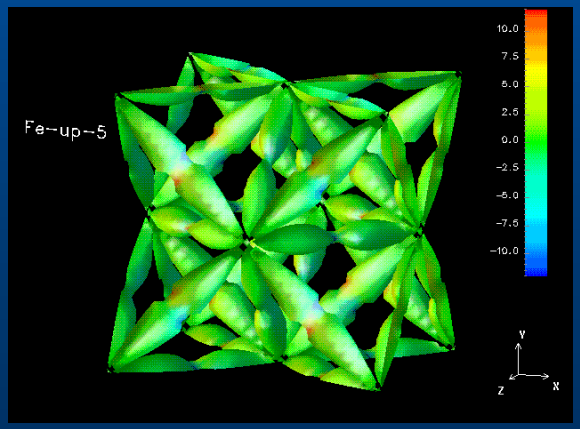
t_{2g} are itinerant electrons providing (Heisenberg-like) RKKY exchange with Friedel oscillations; e_g are more correlated providing (non-Heisenberg) "double exchange" typical for narrom-band systems

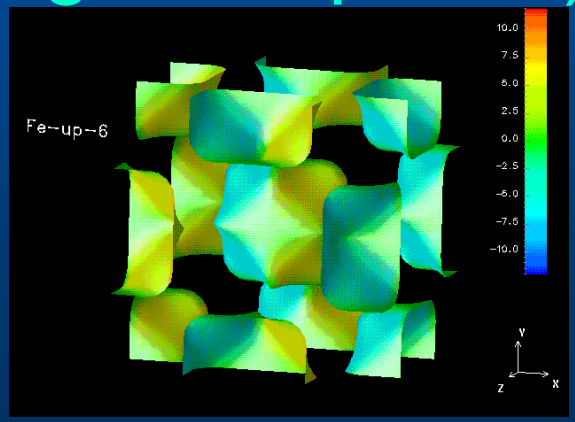
Problem with DFT: coexistence of localized and itinerant behavior



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

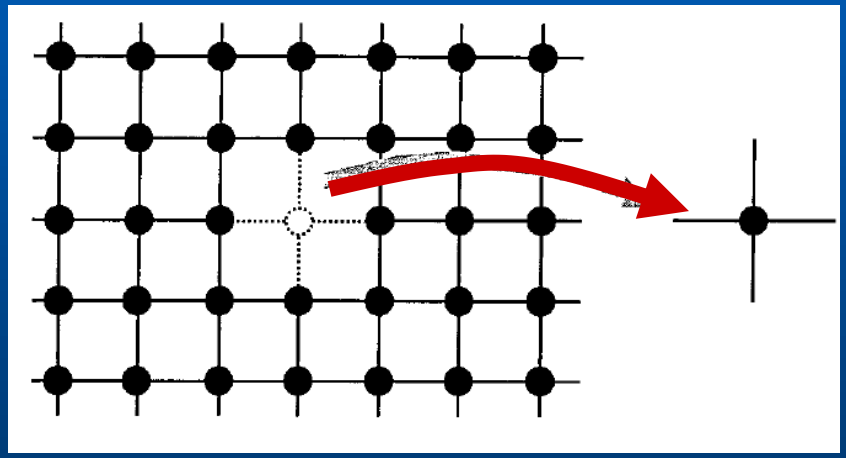
4f electrons are normally pure localized but not 3d

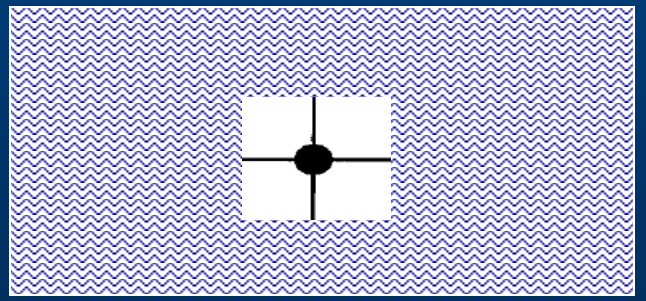
Dynamical Mean Field Theory

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

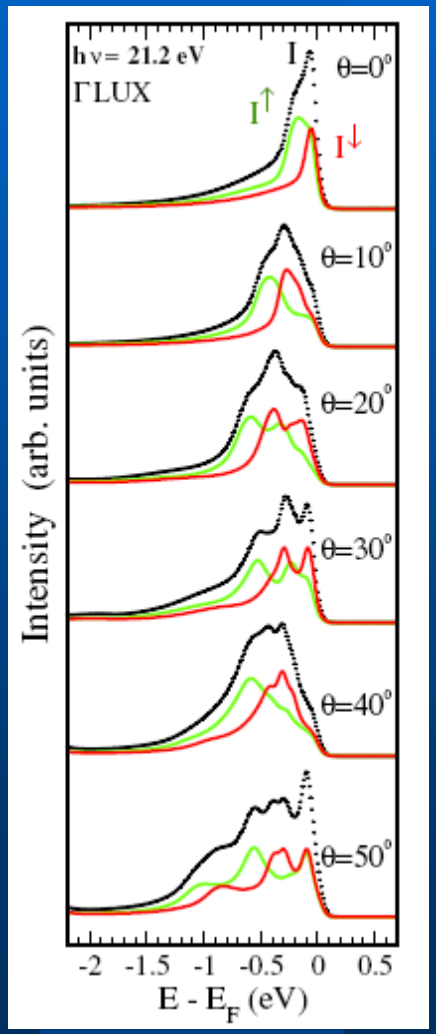




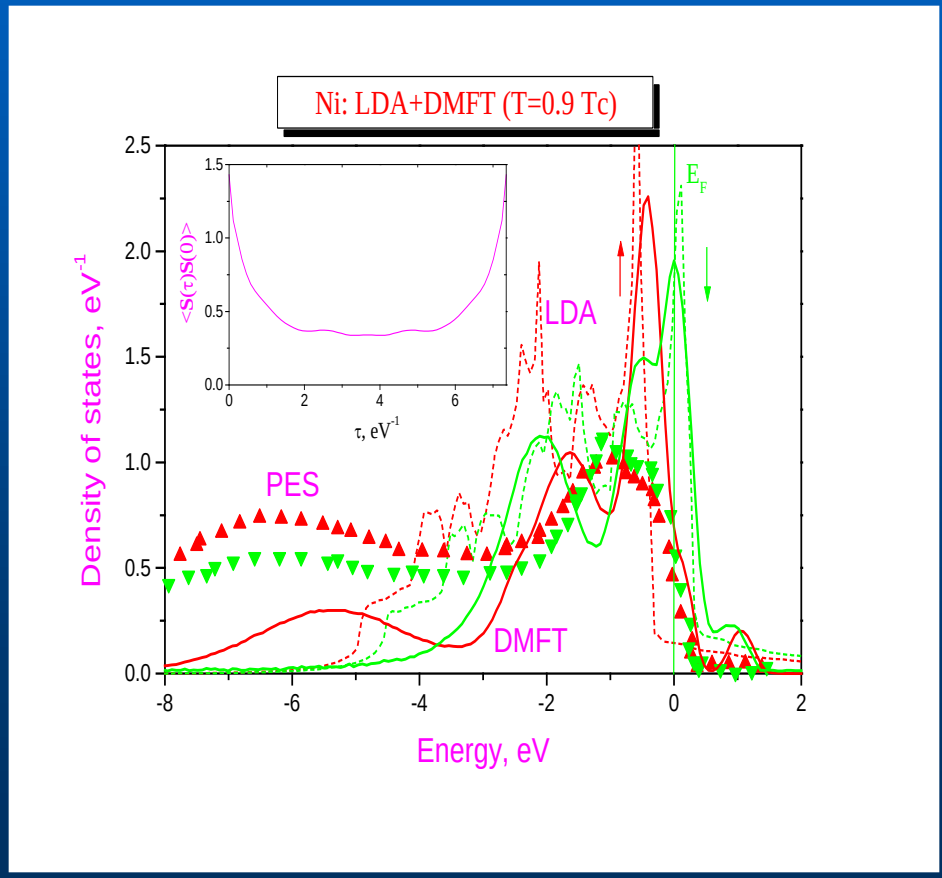
Ferromagnetism of transition metals: LDA+DMFT

Ferromagnetic Ni DMFT vs. LSDA:

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



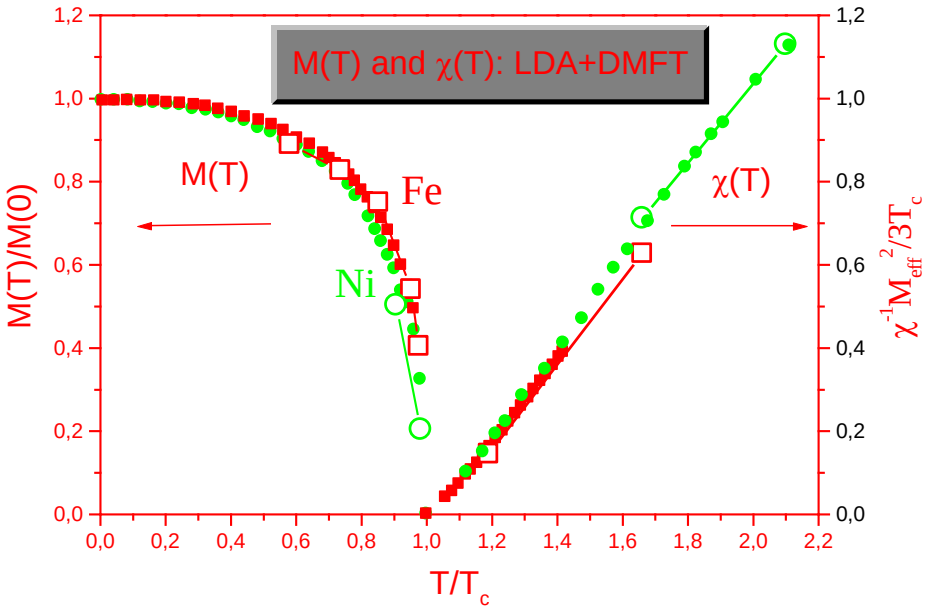
LDA+DMFT with ME
J. Braun *et al*
PRL (2006)



Lichtenstein, MIK, Kotliar, PRL (2001)

DMFT Effective Magnetic Moments: $T>T_c$

⌖	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

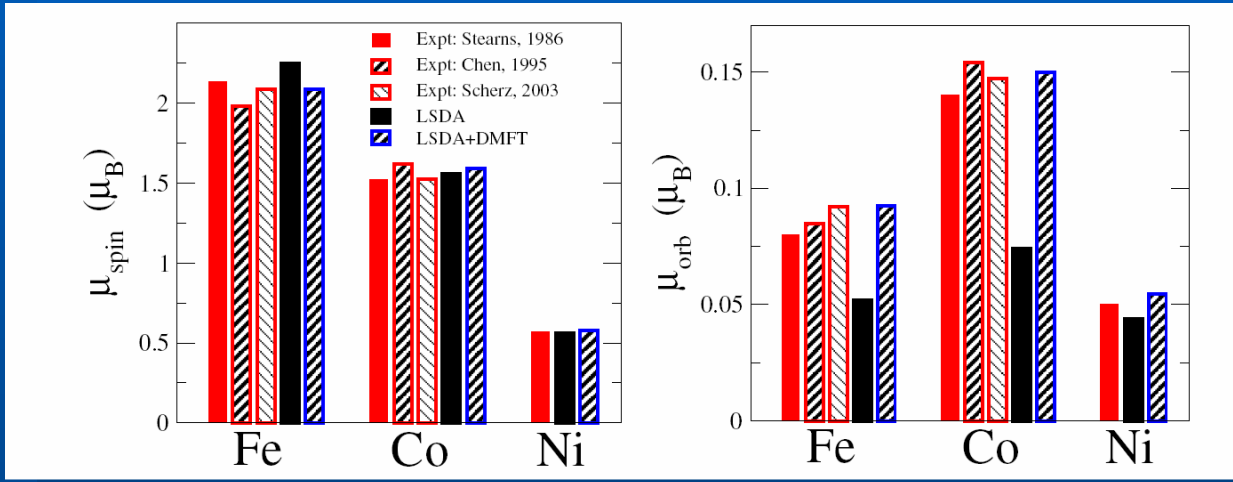


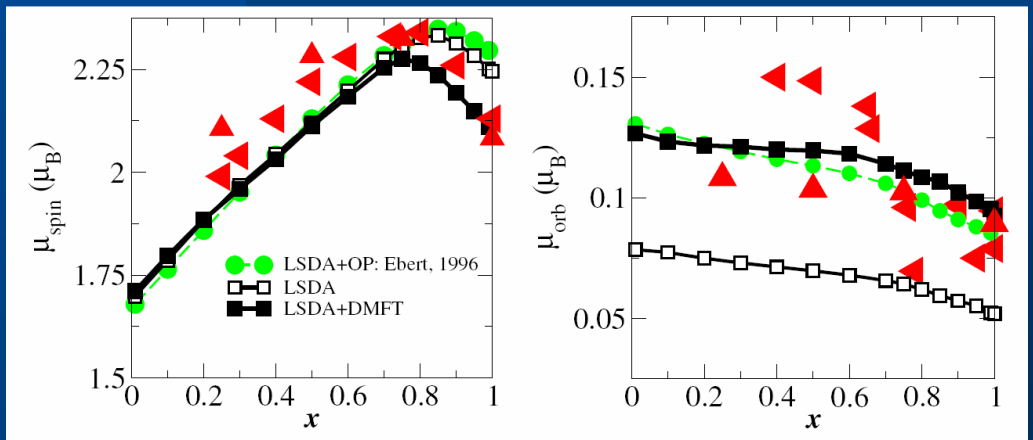
Orbital magnetic moments

Orbital magnetism in transition metal systems: The role of local correlation effects

S. Chadov, J. Minár, M. I. Katsnelson, H. Ebert, D. Ködderitzsch and A. I. Lichtenstein

EPL, **82** (2008) 37001





For Fe_xCo_{1-x} alloys

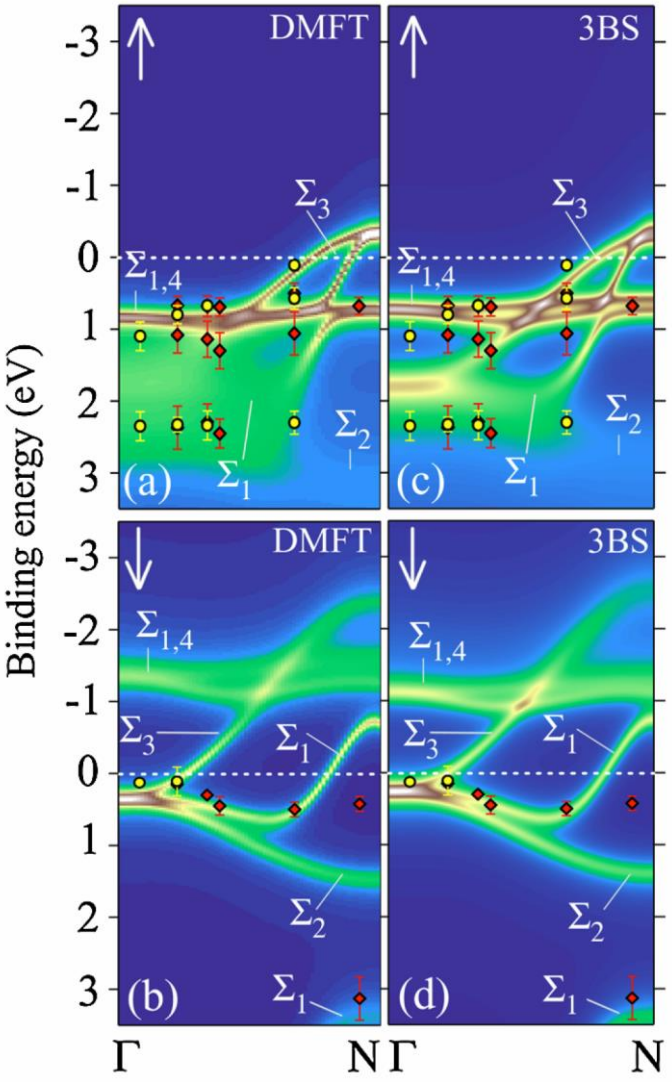
ARPES for iron

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

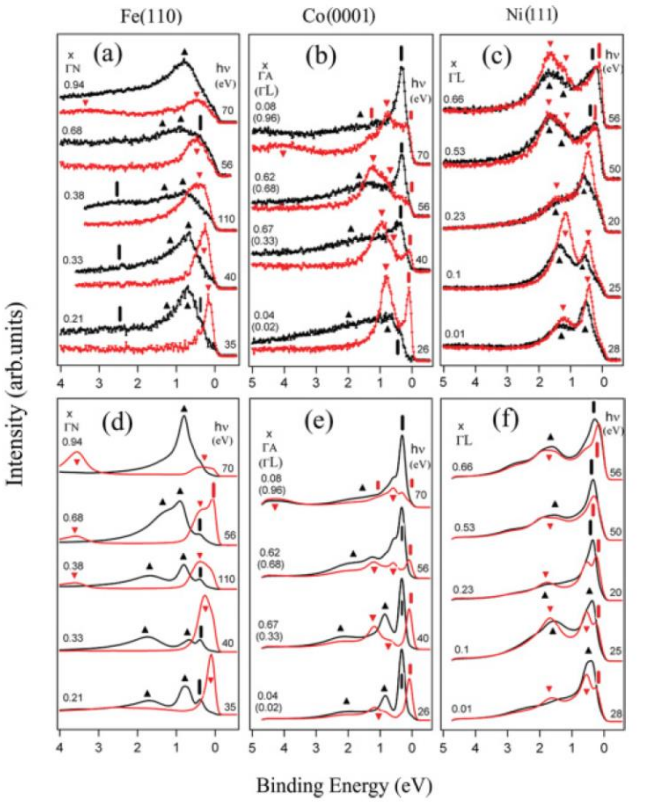


ARPES for 3d metals

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

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

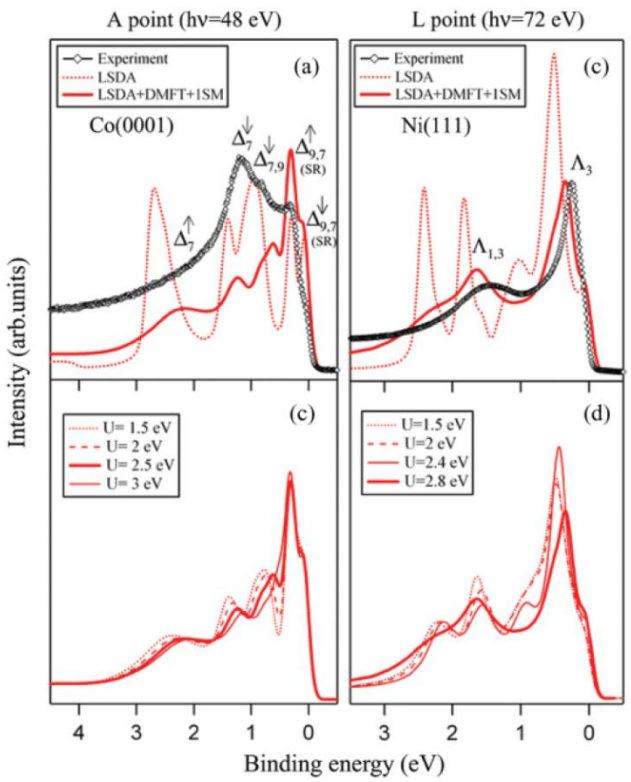
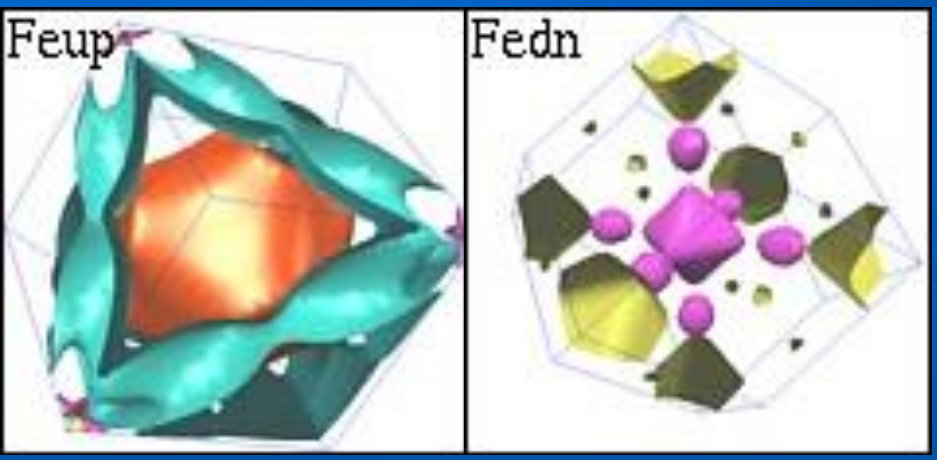
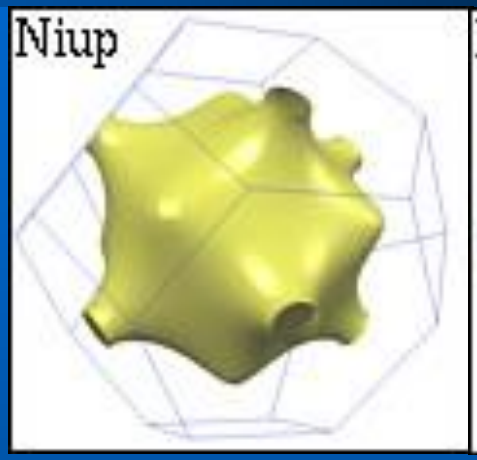


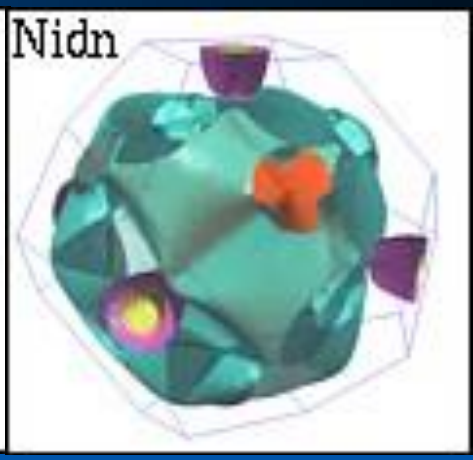
TABLE I. Values of the experimental and theoretical mass enhancement factors m^*/m_0 for majority spin states at high symmetry points of the BBZ of Fe, Co, and Ni, respectively. The theoretical values are derived for $U(\mathrm{Fe}) = 1.5$ eV, $U(\mathrm{Co}) = 2.5$ eV, $U(\mathrm{Ni}) = 2.8$ eV.

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

Why Ni is more local than Fe?







S. Hershfield ***http://www.phys.ufl.edu/fermisurface***

Nickel is almost half-metallic: majority-spin FS almost coincides with the boundaries of the Brillouin band

But the difference for minority spin is even more dramatic

Occupations for majority (minority) electrons
5 means full occupation

Fe: 4.6 (2.34) *Ni: 4.82 (4.15)*

Why Ni is more local than Fe II

Friedel oscillations originating from FS are much weaker in nickel

PHYSICAL REVIEW B, VOLUME 64, 174402

Ab initio calculations of exchange interactions, spin-wave stiffness constants, and Curie temperatures of Fe, Co, and Ni

M. Pajda,[1] J. Kudrnovský,[2,1] I. Turek,[3,4] V. Drchal,[2] and P. Bruno[1]

TABLE I. Effective Heisenberg exchange parameters J_{0j} for ferromagnetic Fe, Co, and Ni for the first 10 shells. Quantities $\mathbf{R}_{0j}$ and N_s denote, respectively, shell coordinates in units of corresponding lattice constants and the number of equivalent sites in the shell.

Fe (bcc)			Co (fcc)			Ni (fcc)		
$\mathbf{R}_{0j}$	N_s	J_{0j} (mRy)	$\mathbf{R}_{0j}$	N_s	J_{0j} (mRy)	$\mathbf{R}_{0j}$	N_s	J_{0j} (mRy)
$(\frac{1}{2}\frac{1}{2}\frac{1}{2})$	8	1.432	$(\frac{1}{2}\frac{1}{2}0)$	12	1.085	$(\frac{1}{2}\frac{1}{2}0)$	12	0.206
(100)	6	0.815	(100)	6	0.110	(100)	6	0.006
(110)	12	−0.016	$(1\frac{1}{2}\frac{1}{2})$	24	0.116	$(1\frac{1}{2}\frac{1}{2})$	24	0.026
$(\frac{3}{2}\frac{1}{2}\frac{1}{2})$	24	−0.126	(110)	12	−0.090	(110)	12	0.012
(111)	8	−0.146	$(\frac{3}{2}\frac{1}{2}0)$	24	0.026	$(\frac{3}{2}\frac{1}{2}0)$	24	0.003
(200)	6	0.062	(111)	8	0.043	(111)	8	−0.003
$(\frac{3}{2}\frac{3}{2}\frac{1}{2})$	24	0.001	$(\frac{3}{2}1\frac{1}{2})$	48	−0.024	$(\frac{3}{2}1\frac{1}{2})$	48	0.007
(210)	24	0.015	(200)	6	0.012	(200)	6	−0.001
(211)	24	−0.032	$(\frac{3}{2}\frac{3}{2}0)$	12	0.026	$(\frac{3}{2}\frac{3}{2}0)$	12	−0.011
$(\frac{3}{2}\frac{3}{2}\frac{3}{2})$	8	0.187	$(2\frac{1}{2}\frac{1}{2})$	24	0.006	$(2\frac{1}{2}\frac{1}{2})$	24	0.001

As a result:

Magnons are much softer in Fe than in Ni (Curie temp. Higher but magnon frequencies lower)

	D_{ex} (meV Å^2)	T_C^{ex} (K)
Fe	280,[a] 330[b]	1044–1045
Co	580,[c, a] 510[b]	1388–1398[c]
Ni	555,[d] 422[a]	624–631

The softer magnons the stronger nonlocal e-m intercation

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:

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

Magnetic torque:

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

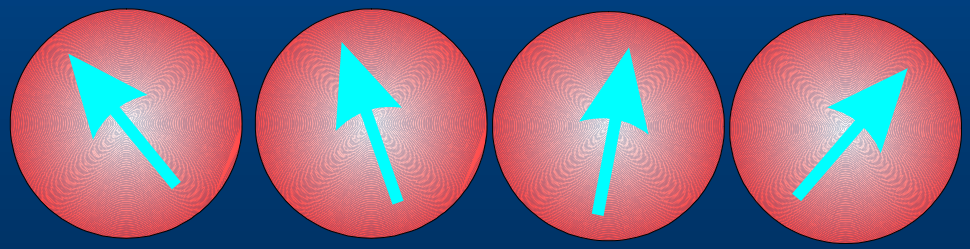
$$\delta\Omega = \delta^* \Omega_{sp} = \mathbf{V}_i \delta\varphi_i$$

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

Exchange interactions:

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

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



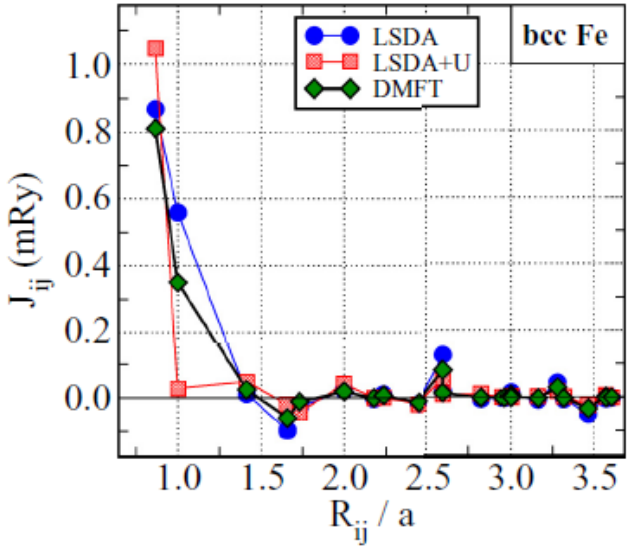
MIK & Lichtenstein Phys. Rev. B 61, 8906 (2000)

Applications

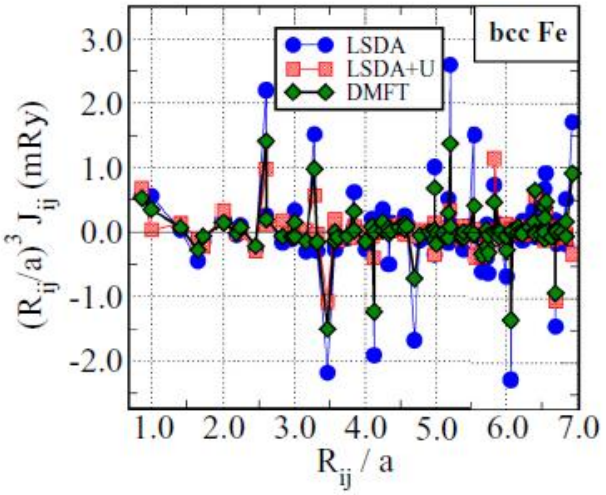
PHYSICAL REVIEW B **91**, 125133 (2015)

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

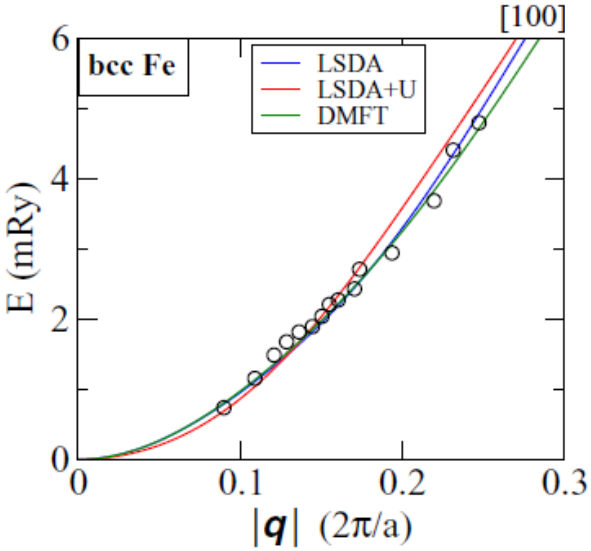
Y. O. Kvashnin,[1] O. Grånäs,[1,2] I. Di Marco,[1] M. I. Katsnelson,[3,4] A. I. Lichtenstein,[4,5] and O. Eriksson[1]



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



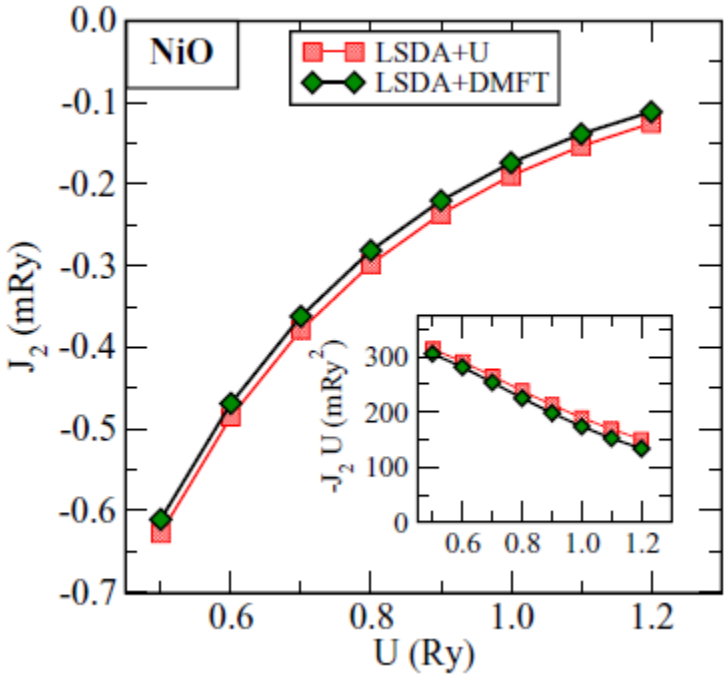
Nontrivial: electronic structure is very different!



←Spin-wave spectrum

Error cancellation?!

Applications II



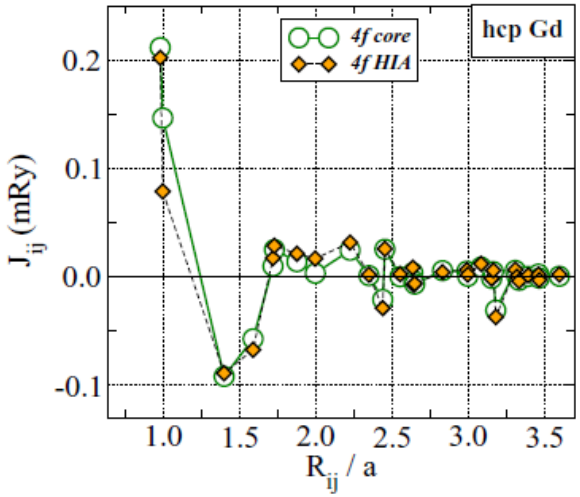
Computational setup	J_1	J_2
LSDA	0.04	−1.58
LSDA + DMFT	−0.003	−0.48
LSDA + U	−0.002	−0.50
LSDA + U (U = 8 eV) (Ref. [42])	0.004/0.0	−0.53
Exp. 1 (Ref. [41])	−0.051	−0.637
Exp. 2 (Ref. [49])	0.051	−0.67

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

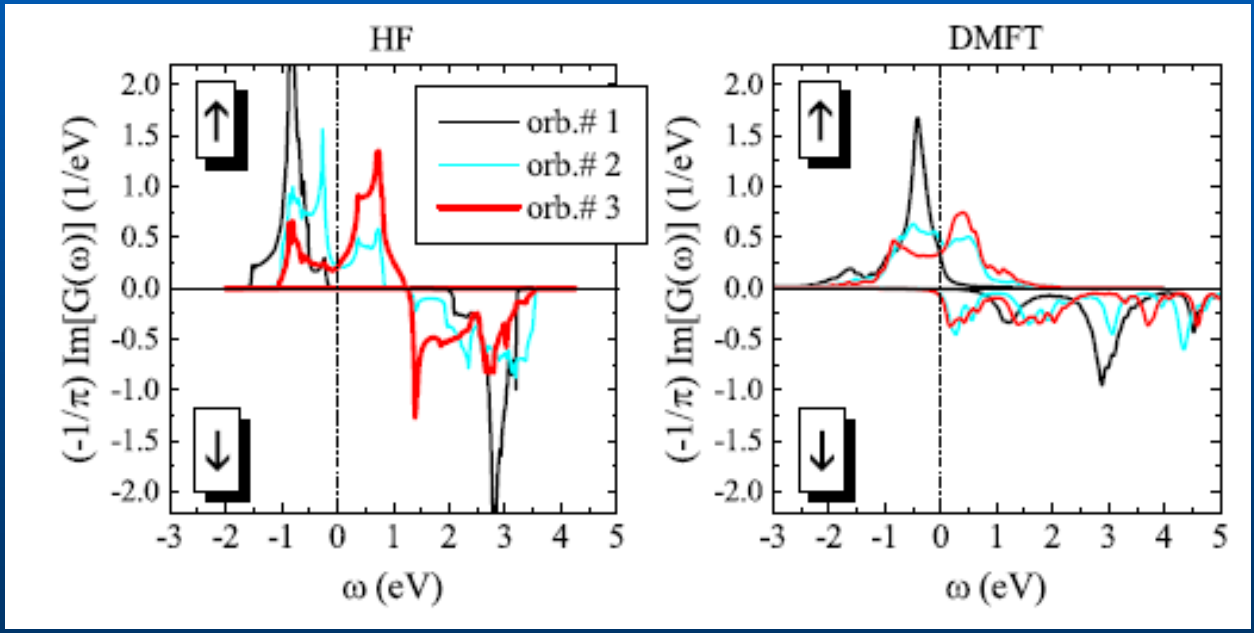
PHYSICAL REVIEW B **92**, 144407 (2015)

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

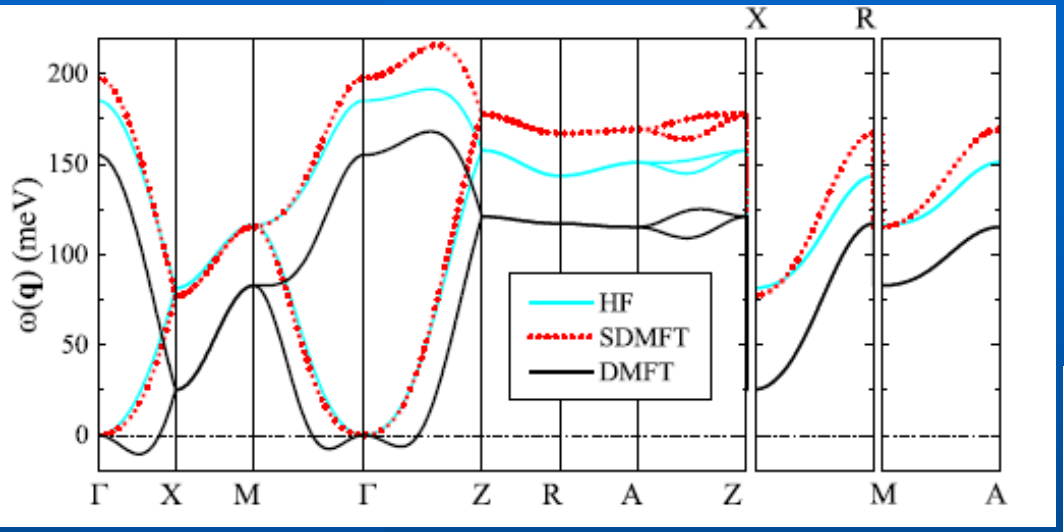
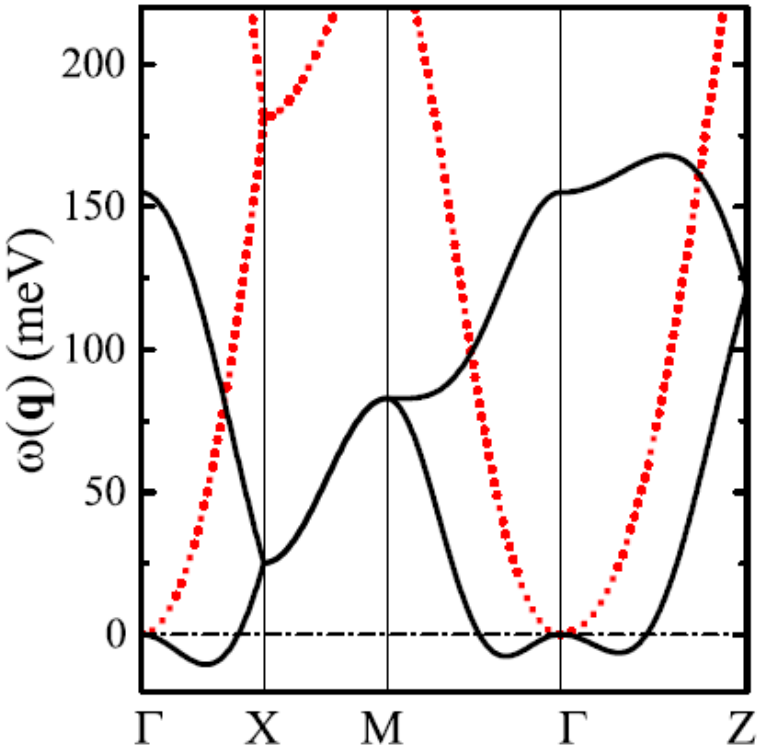


FIG. 9. (Color online) Results of calculations of the spin-wave dispersion with the DMFT parameters obtained for the isolated t_{2g} band (solid line) and after taking into account the additional FM contribution $\Delta J_2 = 17.81$ meV, arising from magnetic polarization of the oxygen band and direct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

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



Direct exchange also plays an important role

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)

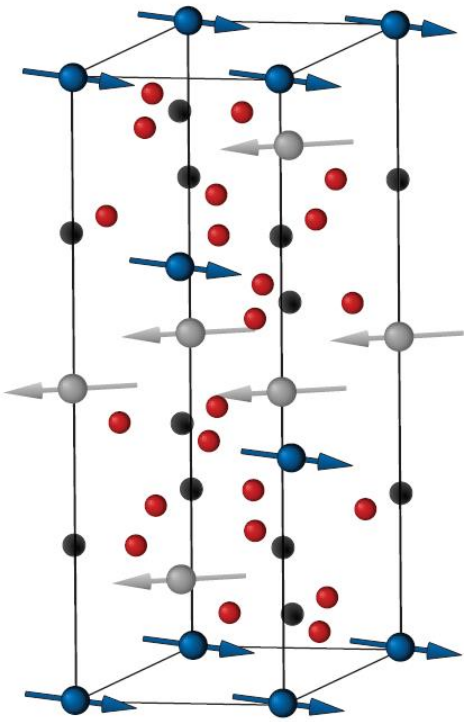


TABLE I. Calculated values of isotropic exchange interactions between magnetic moments in $FeBO_3$ (in meV). The number in parentheses denotes the coordination sphere.

$Fe^{(1)}$	$Fe^{(2)}$	$Fe^{(3)}$	$Fe^{(4)}$	$Fe^{(5)}$	$Fe^{(6)}$	$Fe^{(7)}$
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.

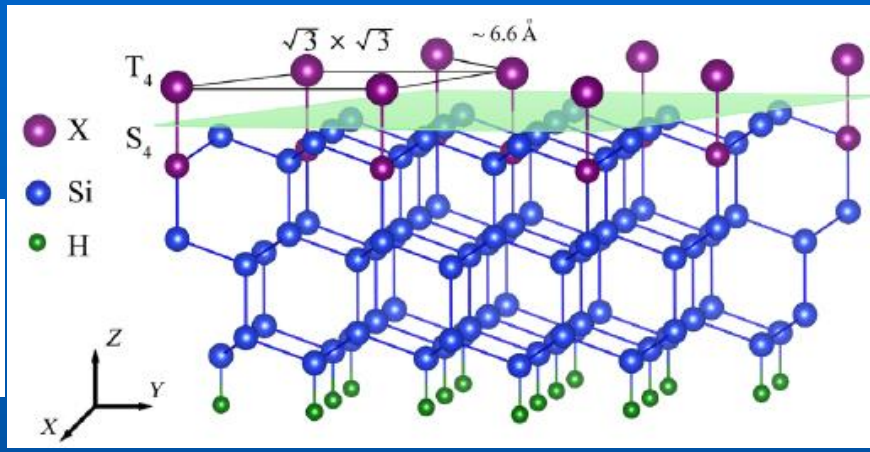
Si(111):X (X=C,Si,Sn,Pb)

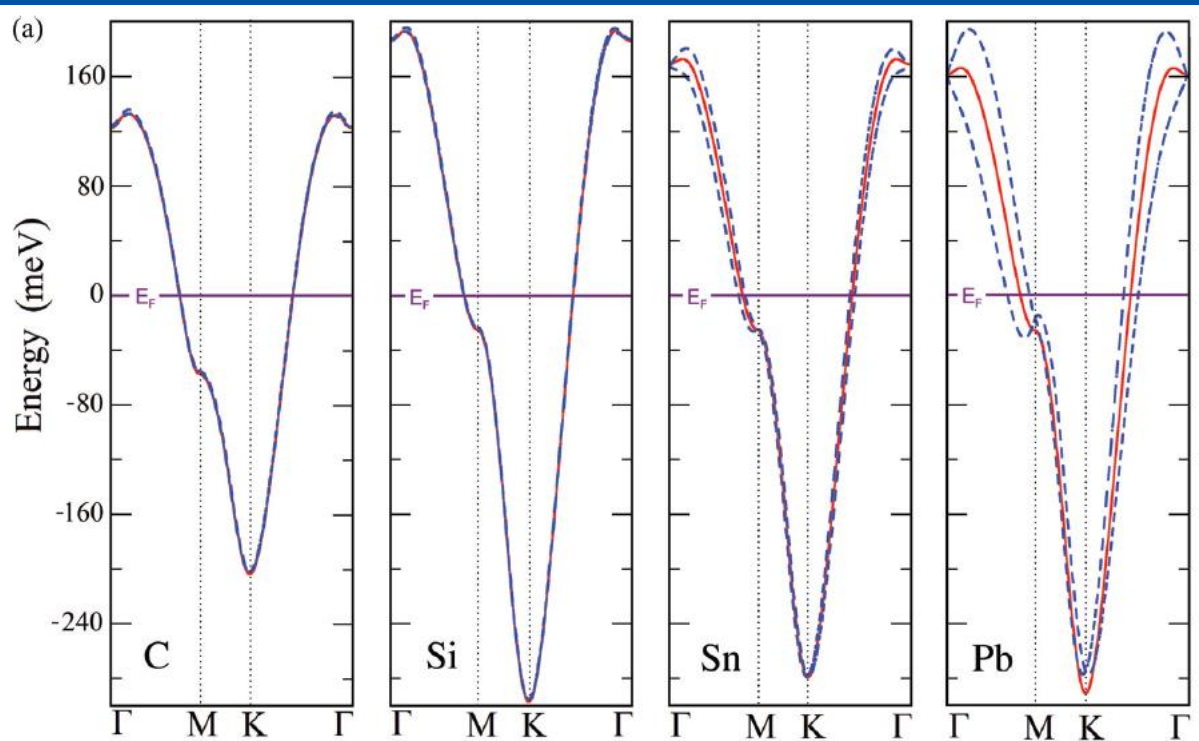
sp-electron magnets

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]



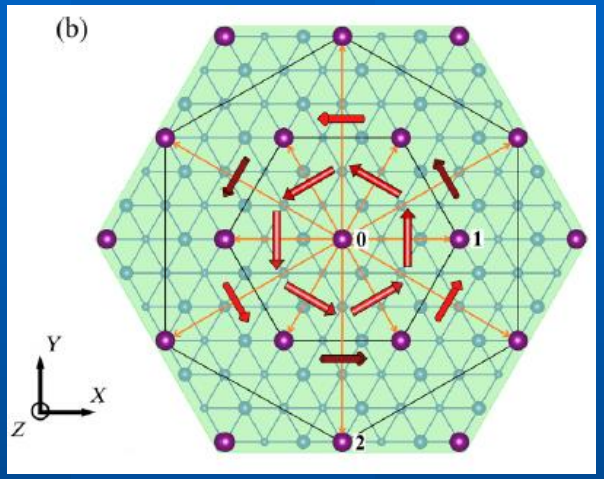


Single narrow band nea the Fermi energy

Red – without SO
Blue – with SO

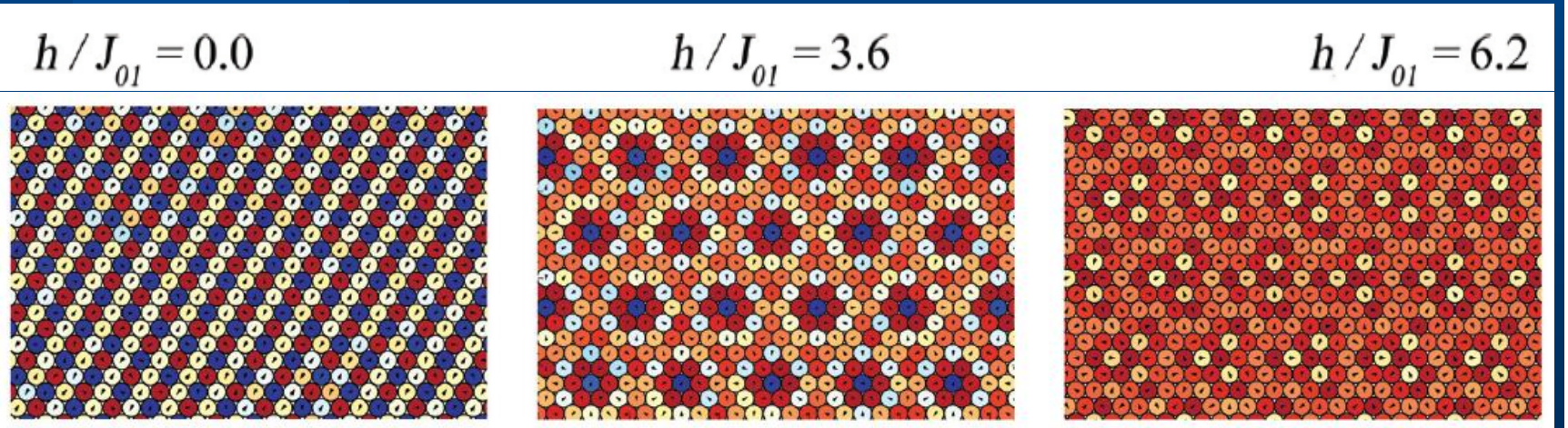
Si(111):X (X=C,Si,Sn,Pb) II

Mott insulator if take into account Hubbard U



Orientation of DMI

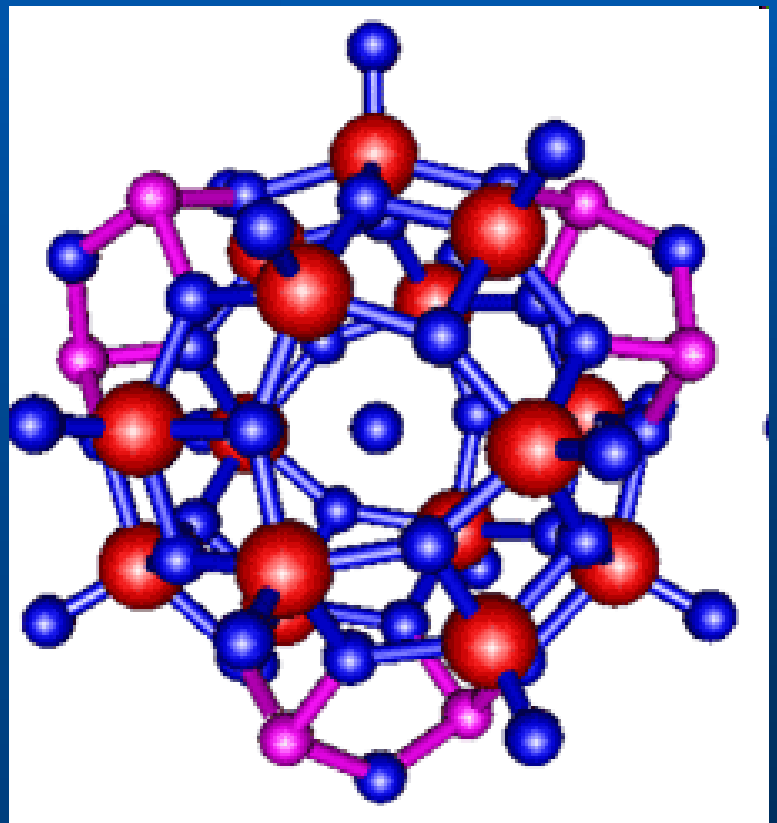
Ground state magnetic configurations for Si(111):Pb in magnetic field (MC simulations)

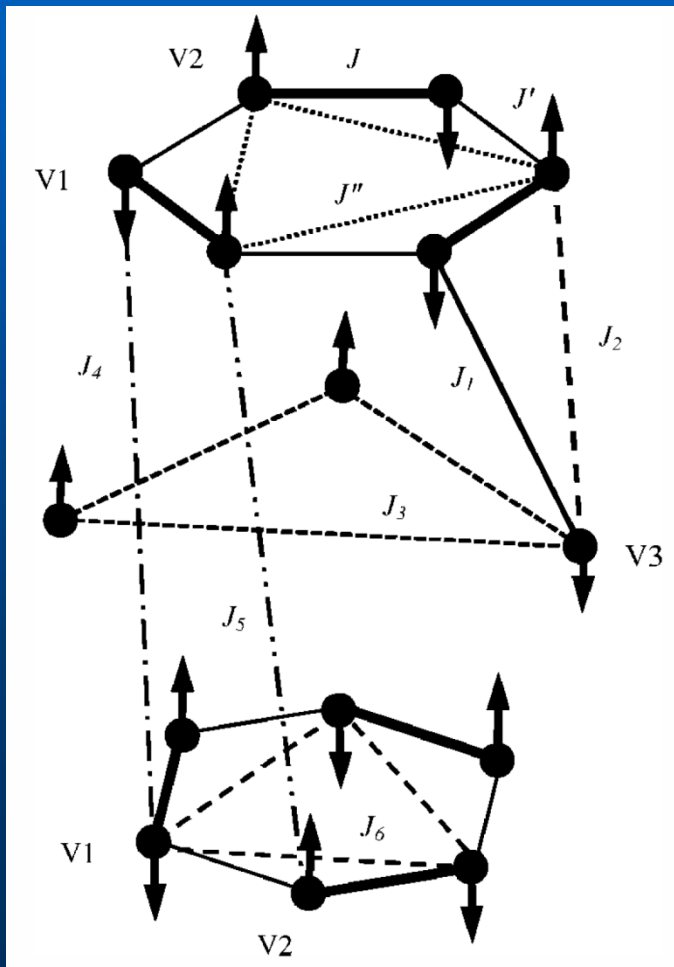


Molecular magnets

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

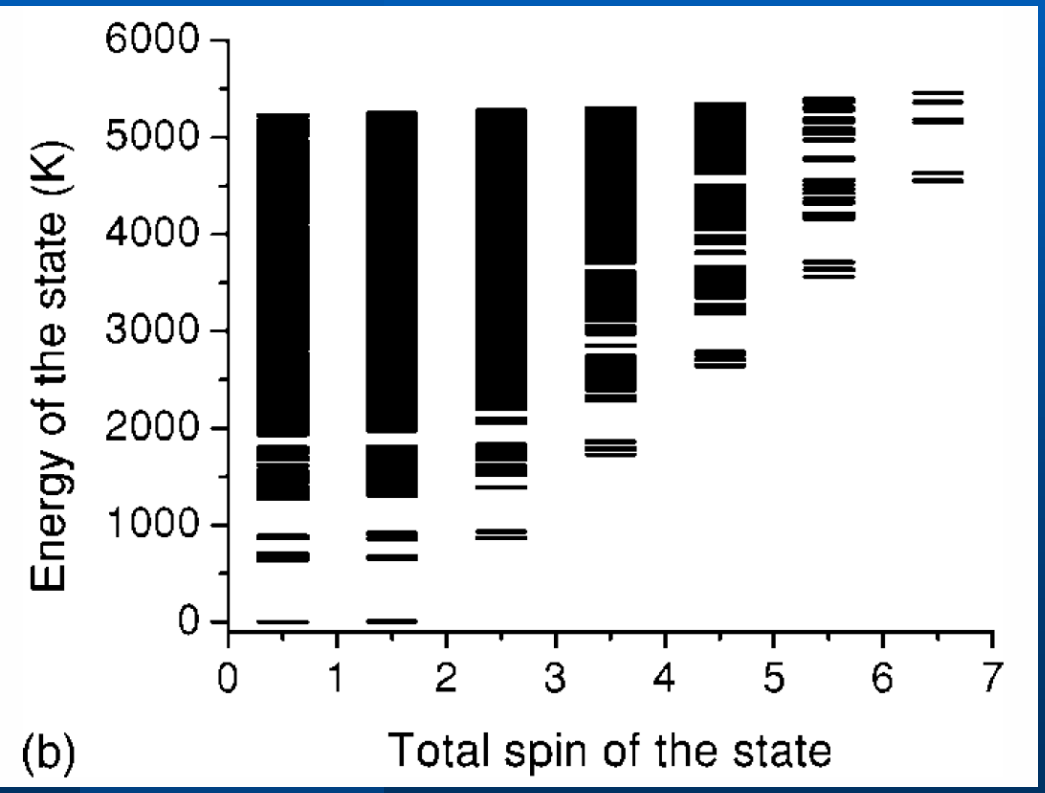
D. W. Boukhvalov,[1,2] V. V. Dobrovitski,[3] M. I. Katsnelson,[2,4] A. I. Lichtenstein,[5] B. N. Harmon,[3] and P. Kögerler[3]

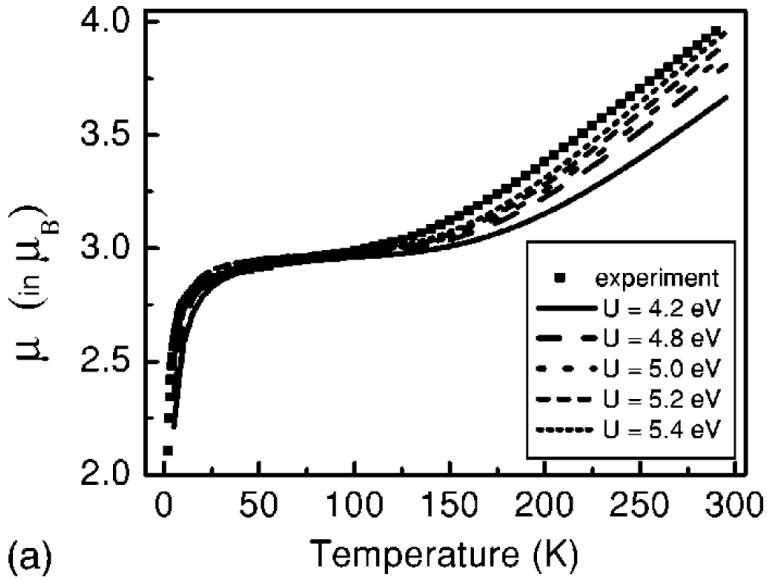
TABLE II. The exchange parameters (in Kelvin), electronic gap, and the magnetic moments of V ions for different magnetic structures of V_{15}. The calculations have been made for U=4 eV, J=0.8 eV.

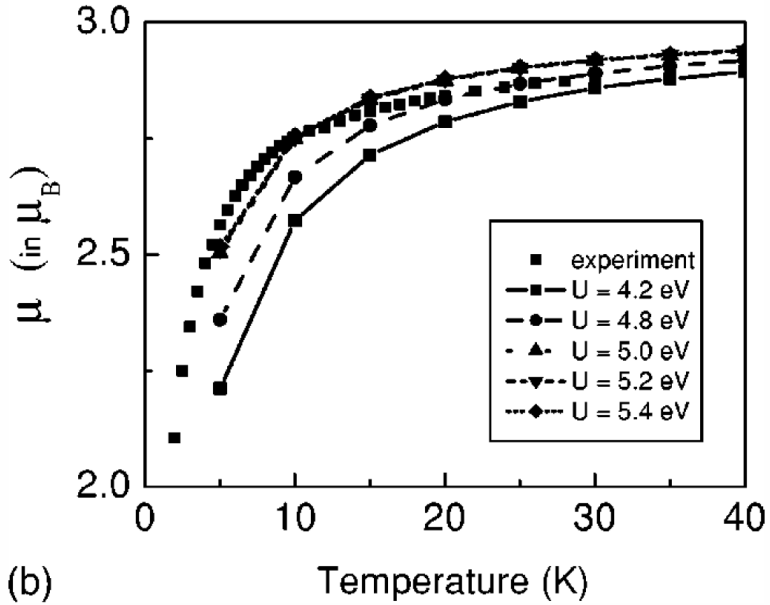
parameter	AFM1	AFM2	FM
J	−910	−905	−942
J'	−45	−46	−53
J''	−136	−139	−156
J_1	−219	−247	−255
J_2	−134	−128	−132
J_3	−5	−5	−6
J_4	−13	−12	−15
J_5	−3	−3	−3
J_6	−3	−3	−3
gap	1.08	1.02	1.16
μ_{V1}	−0.94	−0.93	−0.99
μ_{V2}	+0.91	+0.92	−0.97
μ_{V3}	−1.00	+0.97	−1.00

LDA+U calculations II

Exact diagonalization for Heisenberg model







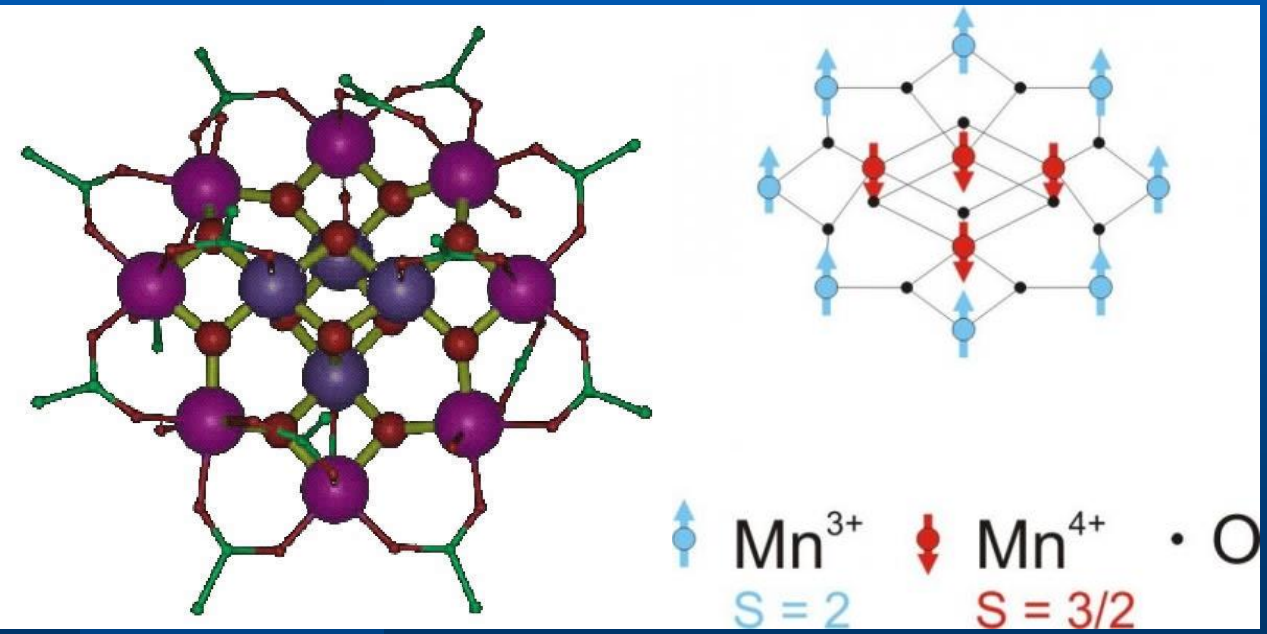
Mn_{12}: full calculations

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

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

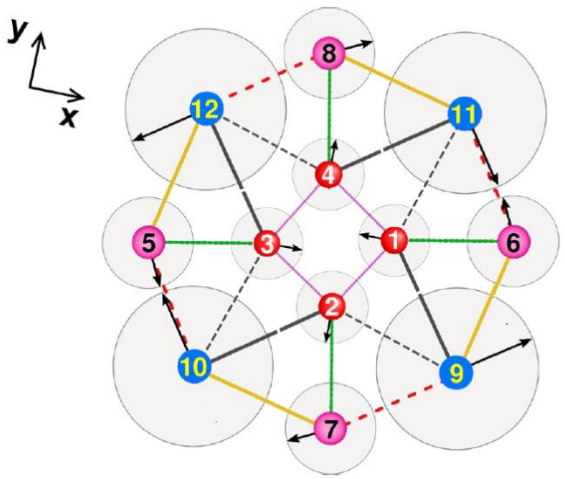


TABLE I. Intramolecular isotropic exchange interaction parameters (in meV) calculated by using the LDA + U approach. Positive sign corresponds to the antiferromagnetic coupling.

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...

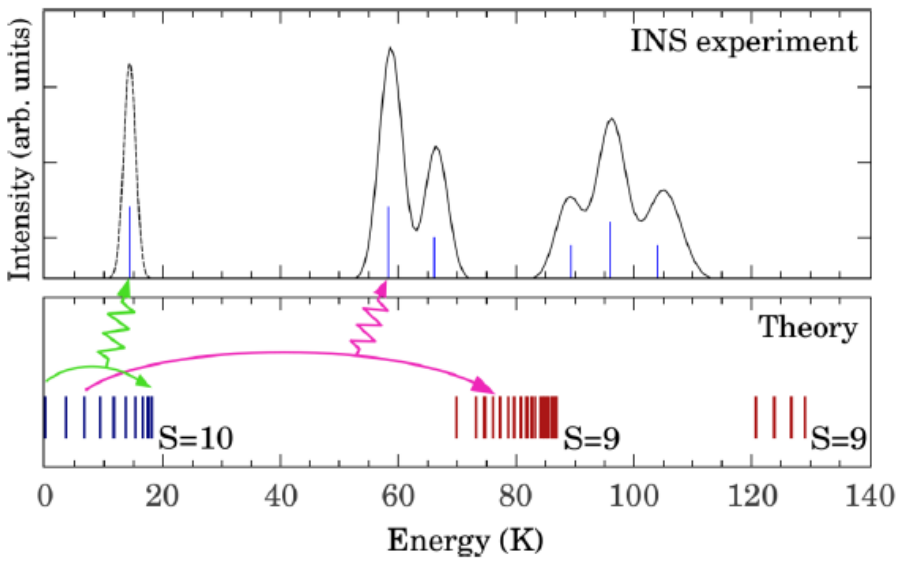
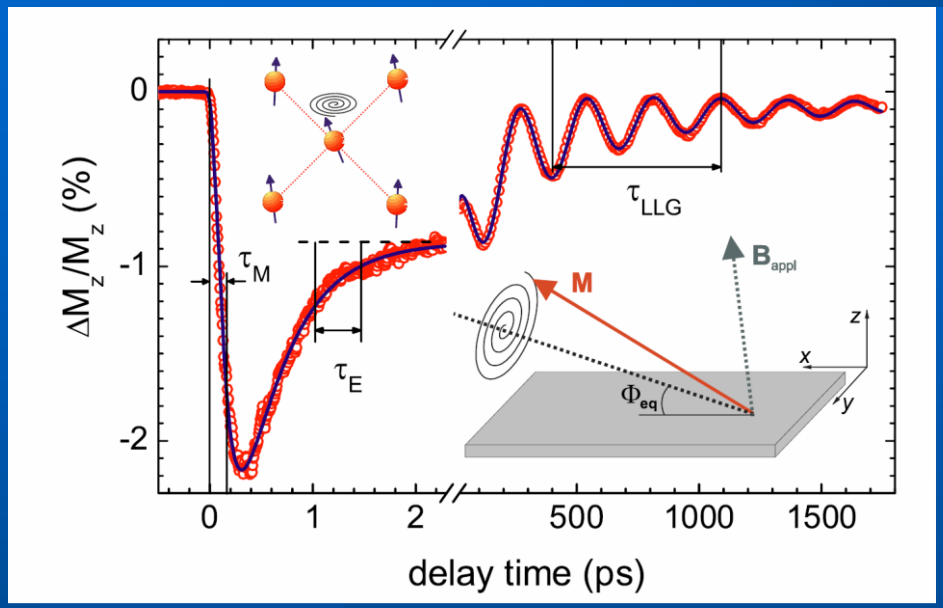


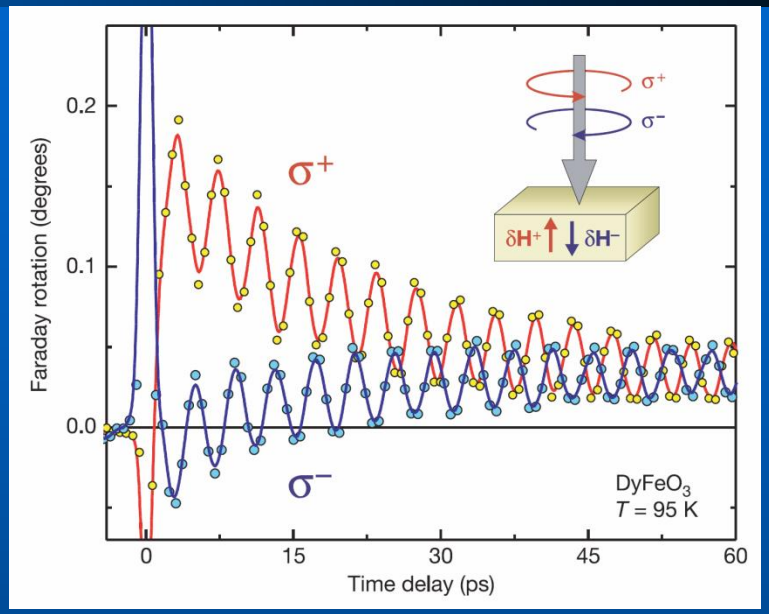
FIG. 2. (Color online) Schematic comparison of the theoretical spectrum obtained by diagonalizing Eq. (1) and INS spectrum taken from Ref. [12] (Figs. 6 and 8 therein). The arrows denote the intra- and interband transitions that correspond to the excitations observed in the INS experiment.

No fitting parameters at all – not so bad!

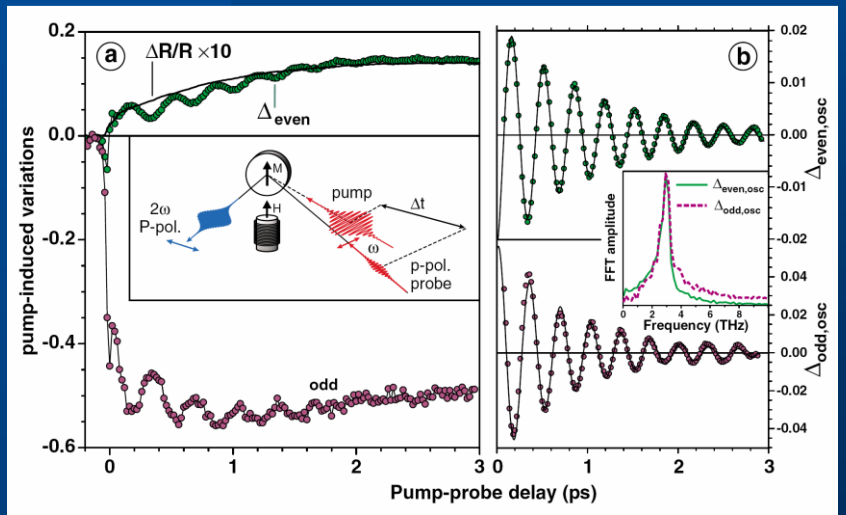
Ultrafast magnetism: Examples



Nickel Koopmans et al PRL 2005



Orthoferrites Kimel et al Nature 2005



Gadolinium Melnikov et al PRL 2003

Ultrafast magnetism: a theory

Non-equilibrium magnetic interactions in strongly correlated systems

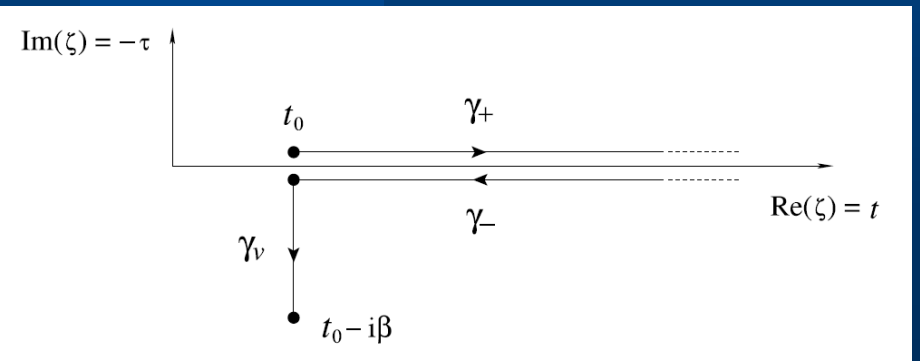
Annals of Physics 333 (2013) 221–271

A. Secchi [a,*], S. Brener [b], A.I. Lichtenstein [b], M.I. Katsnelson [a]

$$\hat{H}(t) \equiv \hat{H}_T(t) + \hat{H}_V \qquad \hat{H}_T(t) \equiv \sum_{i_a\lambda_a} \sum_{i_b\lambda_b} T_{i_a\lambda_a, i_b\lambda_b}(t) \sum_{\sigma} \hat{\phi}^{\dagger}_{i_a\lambda_a\sigma} \hat{\phi}_{i_b\lambda_b\sigma}$$

$$\hat{H}_V \equiv \frac{1}{2} \sum_{i} \sum_{\lambda_1\lambda_2\lambda_3\lambda_4} \sum_{\sigma\sigma'} V_{\lambda_1\lambda_2\lambda_3\lambda_4} \hat{\phi}^{\dagger}_{i\lambda_1\sigma} \hat{\phi}^{\dagger}_{i\lambda_2\sigma'} \hat{\phi}_{i\lambda_3\sigma'} \hat{\phi}_{i\lambda_4\sigma}$$

Consider dynamics of Baym-Kadanoff-Keldysh countour



Path integral over Grassmann variables

$$\mathcal{Z} = \int \mathcal{D}\left[\bar{\phi}, \phi\right] e^{iS\left[\bar{\phi}, \phi\right]}$$

Ultrafast magnetism: a theory II

Introduce rotations

$$\bar{\phi}_{a\pm}(t) = \bar{\psi}_{a\pm}(t) \cdot R^{\dagger}_{a\pm}(t), \qquad \phi_{a\pm}(t) = R_{a\pm}(t) \cdot \psi_{a\pm}(t)$$

$$\bar{\phi}_{av}(\tau) = \bar{\psi}_{av}(\tau) \cdot R^{\dagger}_{av}(\tau), \qquad \phi_{av}(\tau) = R_{av}(\tau) \cdot \psi_{av}(\tau)$$

$$R_i(z) \equiv \begin{pmatrix} \sqrt{1 - |\xi_i(z)|^2} & \xi_i^*(z) \\ -\xi_i(z) & \sqrt{1 - |\xi_i(z)|^2} \end{pmatrix}$$

*Expand effective actions up to the second order in "Holstein-Primakoff" fields ξ, ξ**

$$\xi_i(z) \equiv -\mathrm{e}^{\mathrm{i}\varphi_i(z)} \sin\left[\theta_i(z)/2\right]$$

$$\mathcal{Z} = \int \mathcal{D}\left[\bar{\psi}, \psi\right] \mathrm{e}^{\mathrm{i}S\left[\bar{\psi}, \psi\right]} \int \mathcal{D}\left[\theta, \varphi\right] \mathrm{e}^{\mathrm{i}S'\left[\bar{\psi}, \psi, \xi^*(\theta,\varphi), \xi(\theta,\varphi)\right]}$$

Integrate over Grassman variables neglecting vertex corrections

Ultrafast magnetism: a theory III

General expression of nonlocal in time exchange interactions in terms of Baym-Kadanoff-Keldysh Green's functions. E.g., time-dependent stiffness constant:

$$D_{\alpha\beta}(t) \equiv -\frac{\mathrm{i}}{2M}\sum_{\eta}\eta\sum_{\sigma}\int_{t_0}^{\infty}\mathrm{d}t'\,\mathrm{sign}(t'-t)\,\overline{\Sigma}^{S}(t)\,\overline{\Sigma}^{S}(t')$$

$$\times\frac{1}{n}\sum_{\boldsymbol{k}}\frac{\partial G_{\boldsymbol{k}}^{\eta\sigma}(t',t)}{\partial k_\alpha}\,\frac{\partial G_{\boldsymbol{k}}^{\bar{\eta}\bar{\sigma}}(t,t')}{\partial k_\beta}.$$

Additional terms (twist exchange) of the structure $\propto (\boldsymbol{\sigma}_1 \times \boldsymbol{\sigma}_2)\cdot\boldsymbol{\sigma}_3$

(at equilibrium forbidden by time-reversal symmetry)

The first step is done, a lot of things to do

Manipulation of magnetic interactions by high-frequency laser field

PRL **115,** 075301 (2015) PHYSICAL REVIEW LETTERS week ending 14 AUGUST 2015

Effective Hamiltonians for Rapidly Driven Many-Body Lattice Systems: Induced Exchange Interactions and Density-Dependent Hoppings

A. P. Itin[1,2] and M. I. Katsnelson[1,3]

General idea: average over fast variables (cf. "Kapitza pendulum")

One can change a sign of exchange integral

http://butikov.faculty.ifmo.ru/Russian/ParamPendulumKIO.pdf

General approach to many-body Hamiltonians

$$i\dot{X} = \epsilon\mathcal{H}X$$

where $\epsilon = 1/\omega$ is a small parameter, ω is a frequency of perturbation, $\mathcal{H}(t)$ is the Hamiltonian of the system, X is a column of coefficients of expansion of a quantum state in a certain basis, and fast time was introduced $(t \to t/\epsilon)$,

$$X = C\tilde{X}$$

Unitary transformation to the new wave funstion

The new Hamiltonian:

$$i\dot{\tilde{X}} = [C^{-1}\epsilon\mathcal{H}C - iC^{-1}\dot{C}]\tilde{X}$$

Series expansion:

$$C = \exp[\epsilon K_1 + \epsilon^2 K_2 + \epsilon^3 K_3 + \cdots]$$

K_i are skrew-Hermitian periodic in time matrices

Using the average procedure to find a series for the effective Hamiltonian

General approach to many-body Hamiltonians II

$$i\dot{K}_1 = \mathcal{H}(t) - \langle \mathcal{H}(t) \rangle \equiv \{\mathcal{H}\}, iK_1 = \int \{\mathcal{H}\} dt,$$

$$i\dot{K}_2 = \left\{ \mathcal{H}K_1 - K_1\mathcal{H} - \frac{i}{2}(\dot{K}_1 K_1 - K_1 \dot{K}_1) \right\},$$

$$\epsilon \mathcal{H}_{\text{eff}} = [C^{-1}\epsilon \mathcal{H} C - iC^{-1}\dot{C}] = \epsilon \mathcal{H}_0 + \epsilon^2 \mathcal{H}_1 + \cdots$$

$\{X\} \equiv X - \langle X(t) \rangle$ $\quad$ $\langle X(t) \rangle \equiv (1/2\pi) \int_0^{2\pi} X(t') dt'$

The integration constants are choosen in such a way that $\langle K_i \rangle = 0$

Expansion of the effective Hamiltonian:

$$\mathcal{H}_0 = \langle \mathcal{H} \rangle, \qquad \mathcal{H}_1 = \frac{1}{2} \langle [\{\mathcal{H}\}, K_1] \rangle,$$

$$\mathcal{H}_2 = \frac{1}{2} \langle [\{\mathcal{H}\}, K_2] \rangle + \frac{1}{12} \langle [\{[\{\mathcal{H}\}, K_1]\}, K_1] \rangle, \ldots$$

$[A, B] = AB - BA$

Hubbard model in strong electric field

One-dimensional Hubbard model

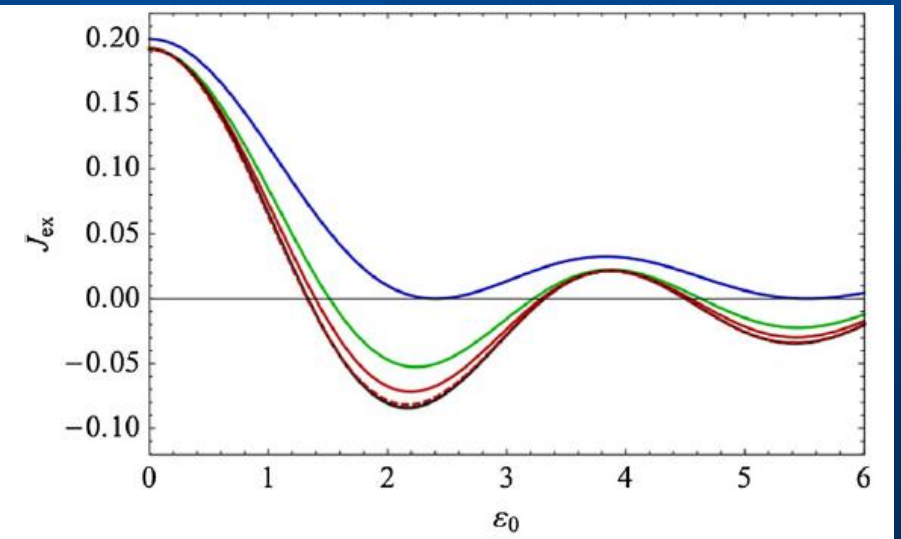
$$H = H_H + H_d(t),$$
$$H_d(t) = \omega\mathcal{E}(\omega t)\sum_j j n_j,$$
$$H_H = J\sum_{i,\sigma}(c^\dagger_{i,\sigma}c_{i+1,\sigma} + c^\dagger_{i+1,\sigma}c_{i,\sigma}) + U\sum_i n_{i,\sigma}n_{i,-\sigma}$$

Effective exchange interaction if frequency is much larger than hopping

$$A = -4J^2U\sum_{m=1}^{\infty} J_m^2(\mathcal{E}_0)/[m^2\omega^2 - U^2]$$ *

Itin & Katsnelson, PRL 2015

Change of sign of exchange interaction!



U = 10, J = 1, ω = 16

Color solid curves, from top to bottom:
- Bare exchange interaction
- Second order expansion;
- Fourth order expansion
- Formula (*)

Dashed line shows numerical results for nonequilibrium exchange (J. Mentink et al)

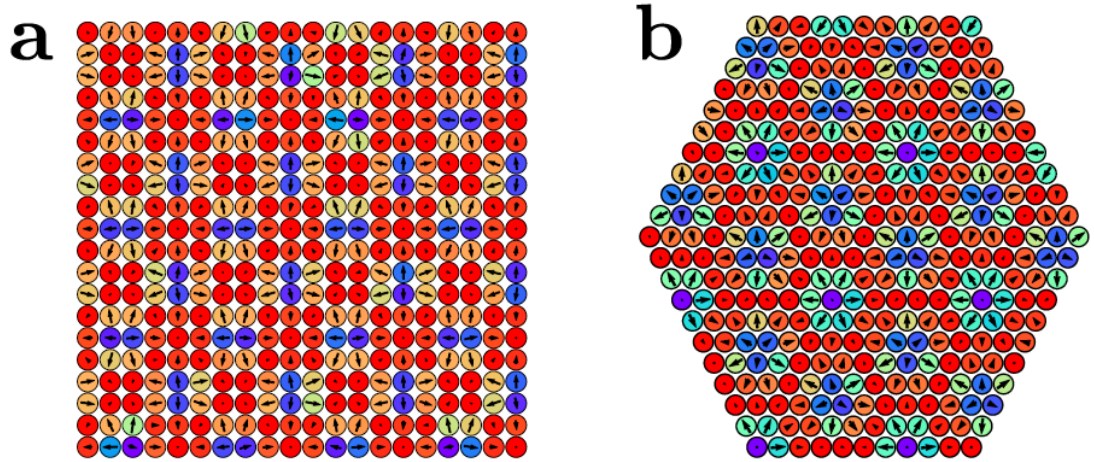
Using laser field to manipulate magnetic structure

PRL 118, 157201 (2017) PHYSICAL REVIEW LETTERS week ending 14 APRIL 2017

Dynamical and Reversible Control of Topological Spin Textures

E. A. Stepanov,[1] C. Dutreix,[1,2] and M. I. Katsnelson[1]

One can reach a regime when D >> J with unusual nanockyrmione mosaic (Stepanov, Nikolaev, Dutreix, MIK, Mazurenko, 2018)



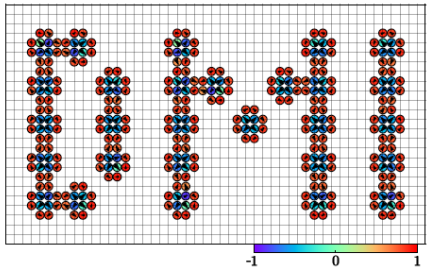


FIG. 1. Nanoskyrmion-designed DMI abbreviation obtained from Monte Carlo simulations of the Heisenberg-exchange-free model on the non-regular square lattice with $B_z = 1.2$. Arrows and colors depict the in-plane and out-of-plane spin projections, respectively.

FIG. 3. Fragments of the spin textures and spin structure factors obtained with the Heisenberg-exchange-free model on the square 20×20 (**a**) and triangular 21×21 (**b**) lattices. The values of magnetic fields were chosen $B_z = 3.0$ and $B_z = 3.2$ for the triangular and square lattices, respectively. The calculated skyrmion numbers for the triangular (blue triangles) and square (red squares) lattices (**c**). An applied magnetic field is in units of DMI. The temperature is equal to $T = 0.01\,|\mathbf{D}|$.

Beyond the talk

Finite-temperature effects

Ab initio spin dynamics for real systems

Intermediate level: TB spin dynamics

Spin and orbital magnetism

And many, many specific applications to real materials

Collaboration

Recent:

A. Lichtenstein and S. Brener (Hamburg)
A. Secchi, E. Stepanov, 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