

Radboud Universiteit



Theory of magnetic interactions in real materials

Mikhail Katsnelson



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, CrO₂
5. Dzyaloshinskii-Moriya interactions
6. Applications: molecular magnets, adatoms on Si(111) and SiC(0001) surfaces...
7. Manipulation of magnetic interactions by high-frequency laser fields

Epigraphs

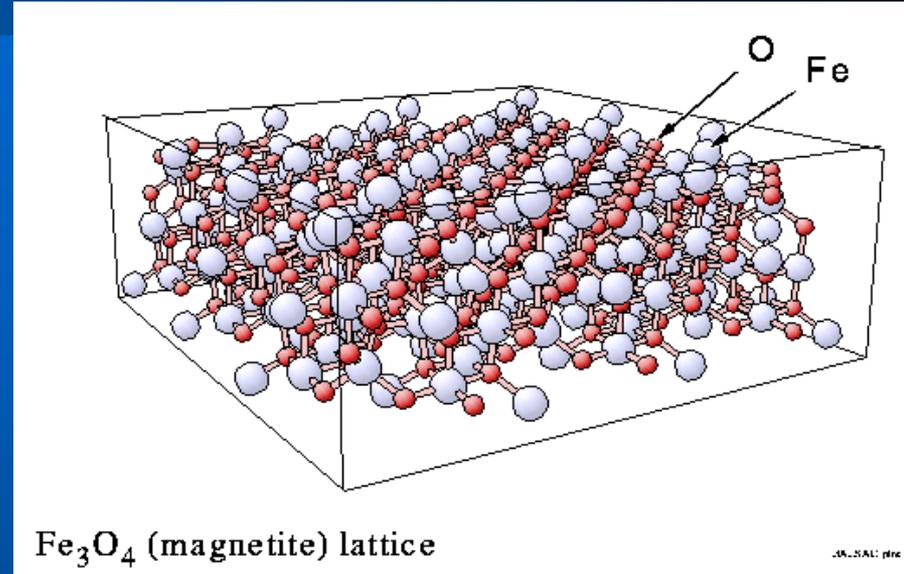
To the theoretical physicists, ferromagnetism presents a number of very interesting, unsolved and beautiful challenges

(Feynman Lectures on Physics)

Make things as simple as possible but not simpler

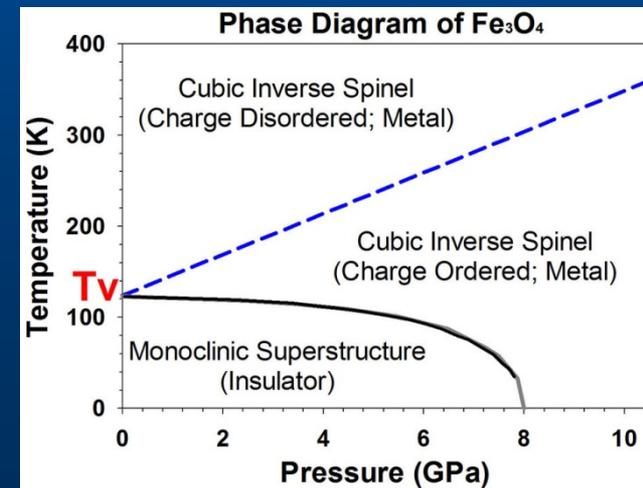
(A. Einstein)

Magnetite – first known magnet



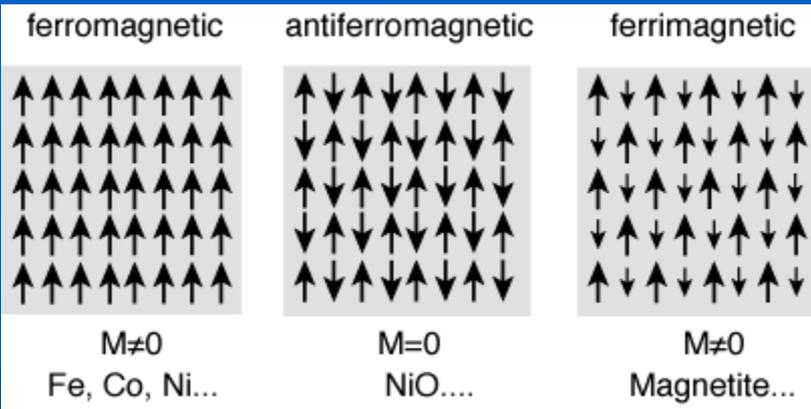
Very complicated structure, still a lot of open questions

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



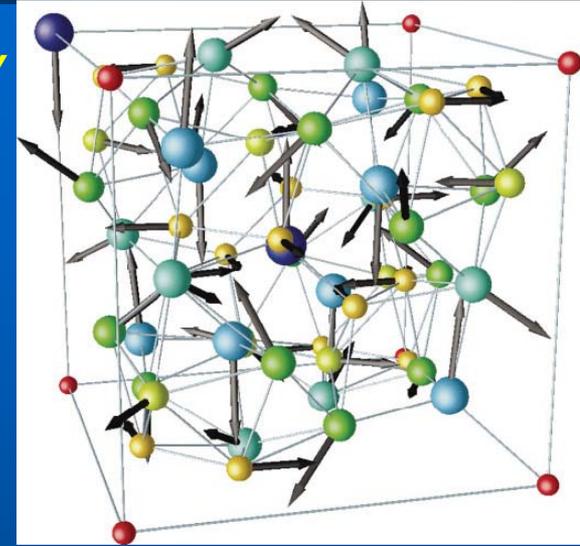
Types of magnetic ordering

Textbook wisdom

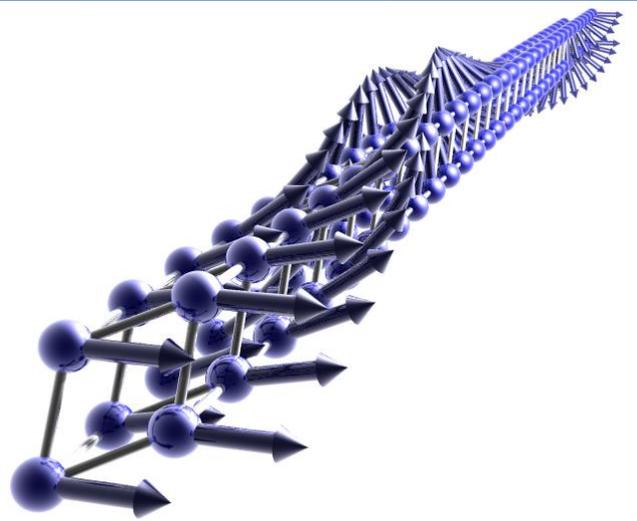


Sometimes very complicated

α -Mn

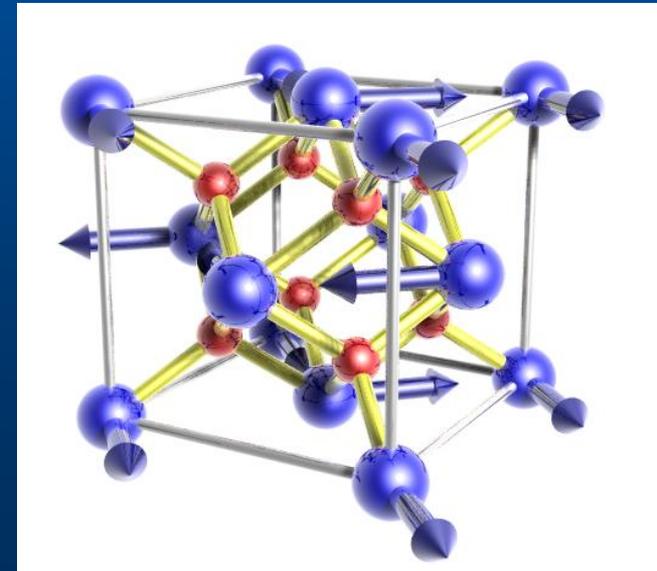


Spin spirals

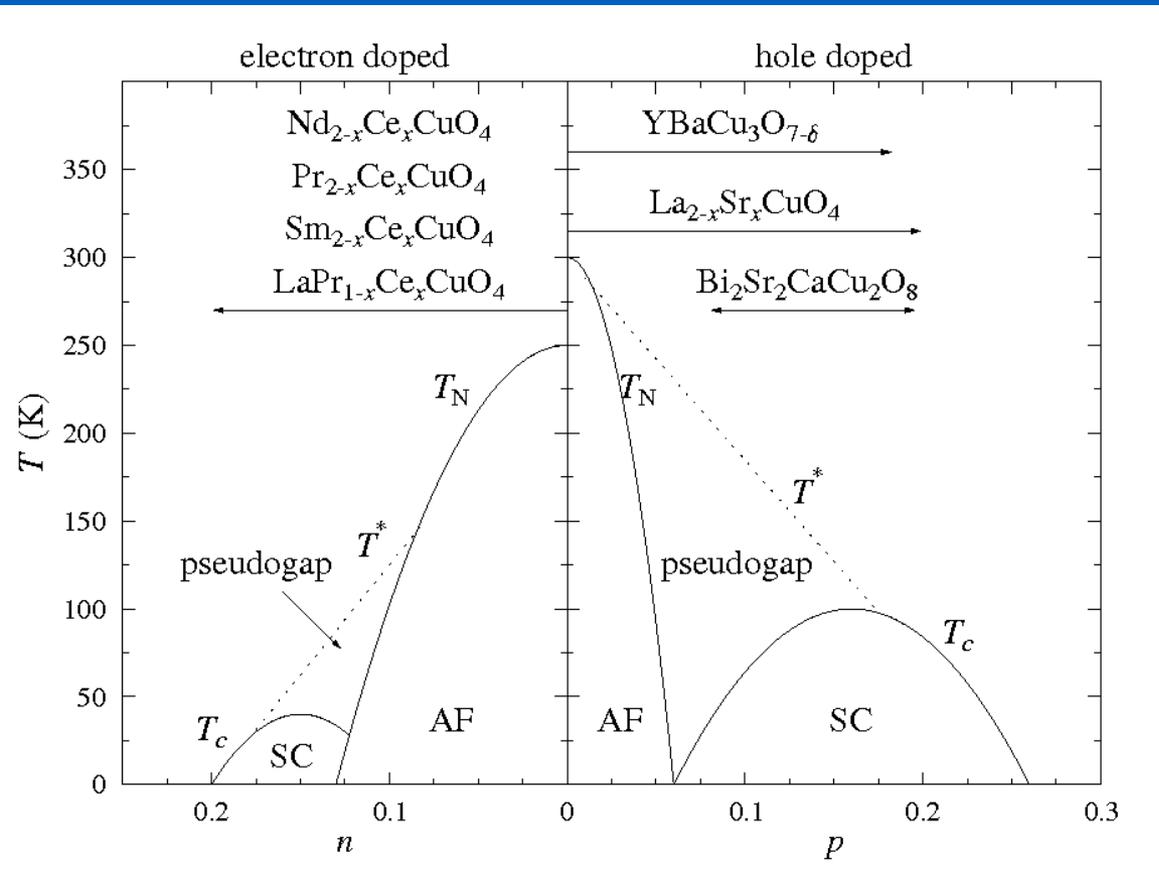


γ -Fe

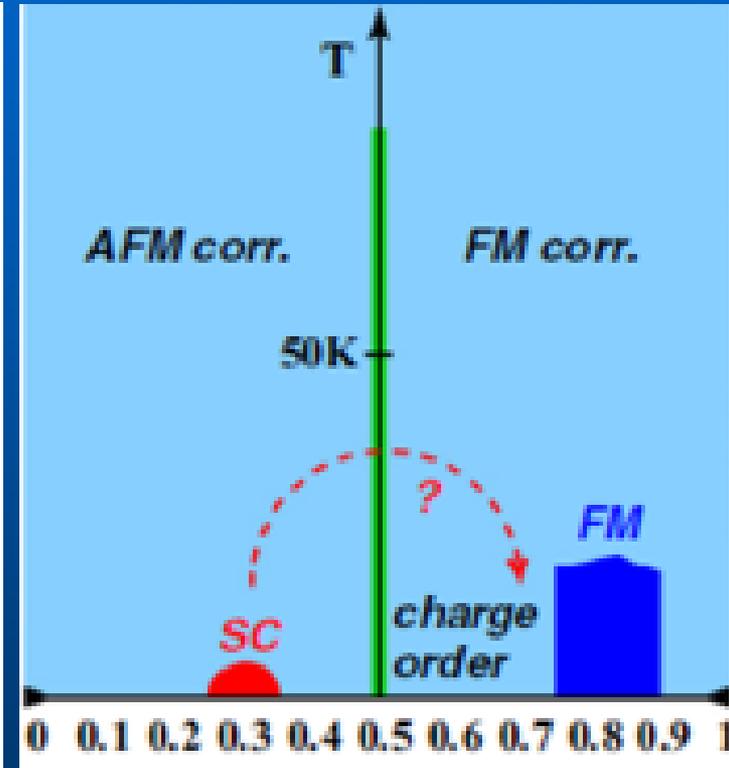
UO_2



Relation to superconductivity and other phenomena



Simplified phase diagram of Cu-O high- T_c superconductors



Layered cobaltates Na_xCoO_2

Ferromagnetism of Fe, Co, Ni

PERIODIC TABLE

PERIODS	Representative Elements s-block		Transition Elements d-block										Representative Elements p-block					Noble gases	
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	
1	H												B	C	N	O	F	He	
2	Li	Be											Al	Si	P	S	Cl	Ne	
3	Na	Mg											Ga	Ge	As	Se	Br	Ar	
4	K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	In	Sn	Sb	Te	I	Kr	
5	Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	Hg	Tl	Pb	Bi	Po	Xe	
6	Cs	Ba		Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn	
7	Fr	Ra		Rf	Db	Sg	Bh	Hs	Mt										
			Inner Transition Elements f-block																
			La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu		
			Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr		



Ferromagnetism of iron is known from ancient times



Iron



Cobalt



Nickel

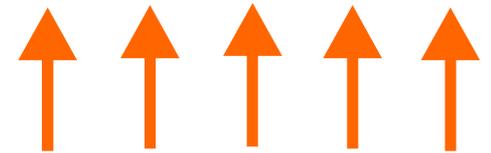
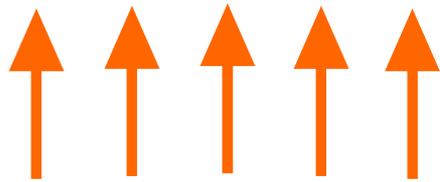
Itinerant-electron ferromagnetism at finite temperatures

Stoner

Heisenberg

Spin-fluctuation

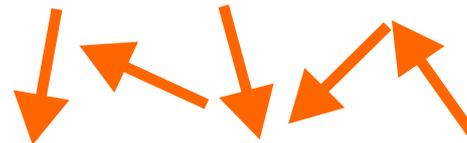
$T=0$



$T < T_c$



$T > T_c$



Stoner criterion

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

$N(E_{\text{F}})$ 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*

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

If Fe would be Stoner magnet it would have $T_{\text{C}} \approx 4000$ K (in reality 1043 K)

*In reality, T_{C} is determined by spin fluctuations,
That is, exchange parameters*

Types of magnetic interactions

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

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. Responsible for weak FM, skyrmions etc.

Density Functional Theory

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

Spinor

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

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

\mathbf{B} is self-consistent magnetic field

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

Simplifications

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

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

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

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

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

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

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

$$E_{sp} = \sum_v^{occ} \epsilon_v$$

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

Variation

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

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

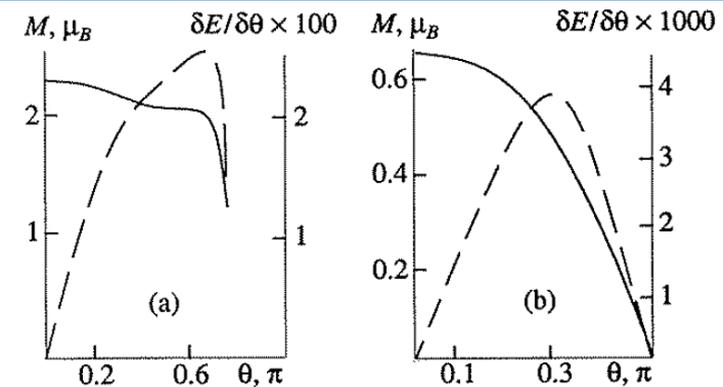
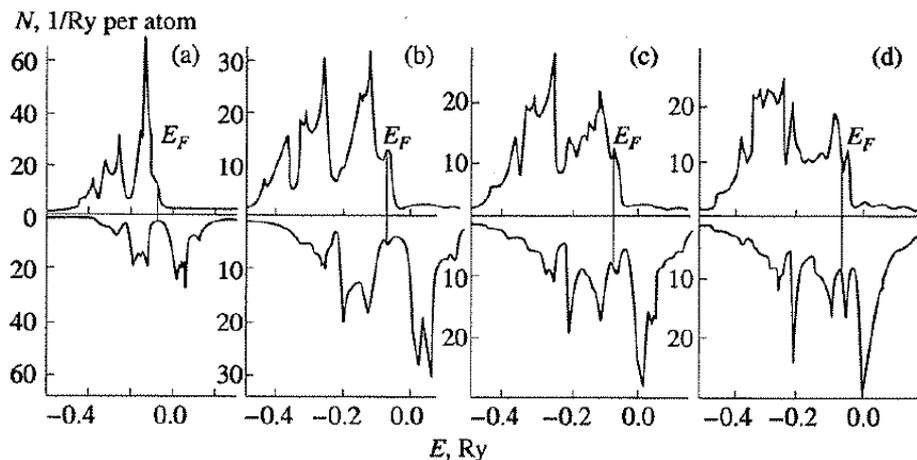


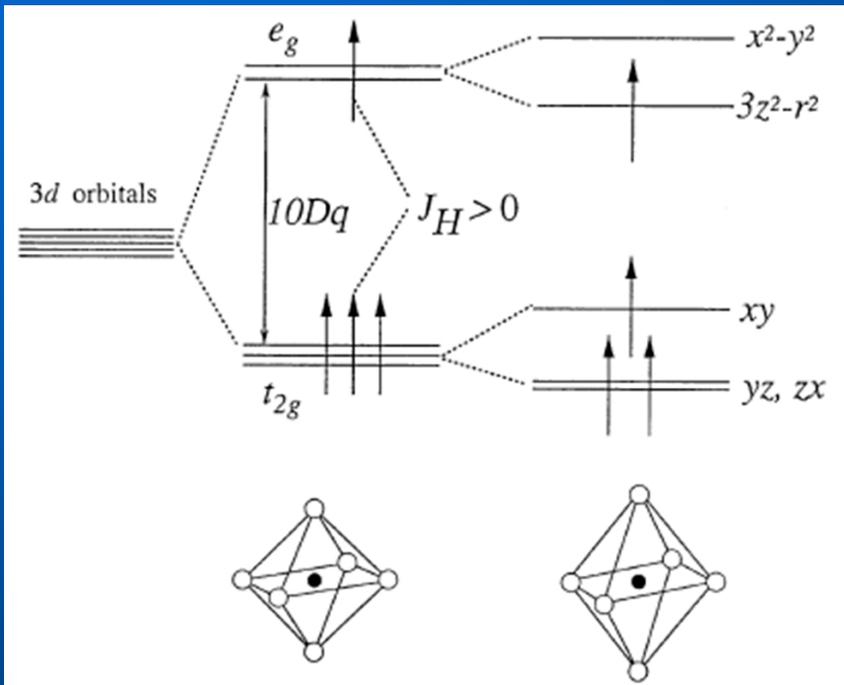
Fig. 4.4. Magnetic moment in Bohr magnetons (the full curve) and the first derivative of energy with respect to angle of rotation in Ry (the dashed curve) according to calculations in [168]: (a) Fe, (b) Ni.



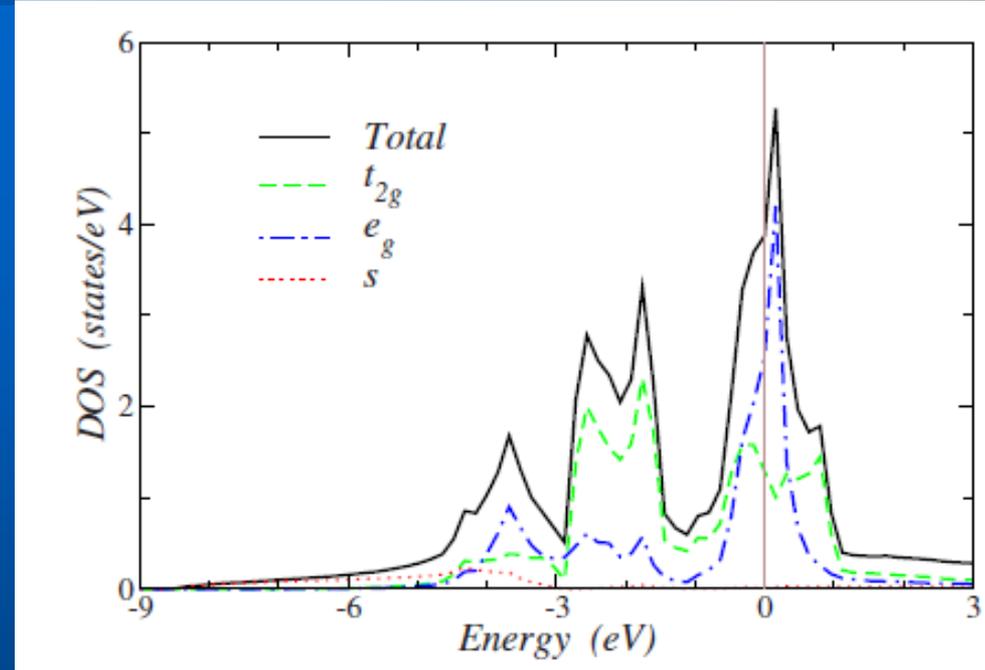
Electronic structure
is angle-dependent

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

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. Eriksson¹

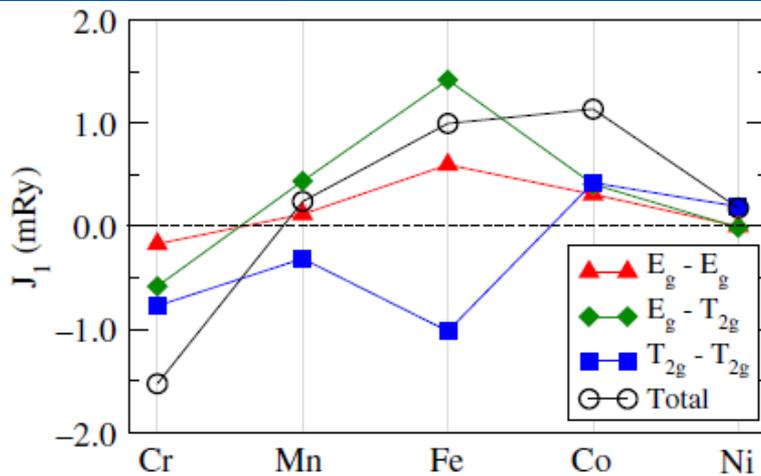
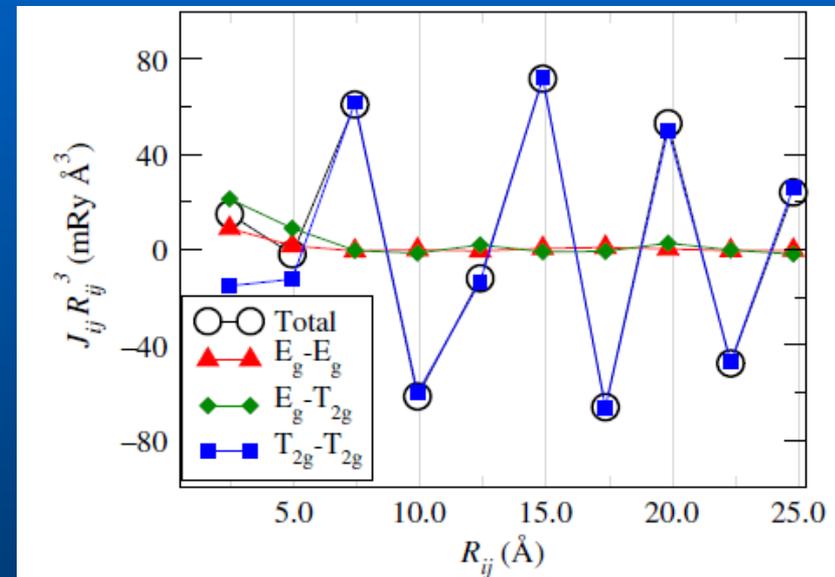
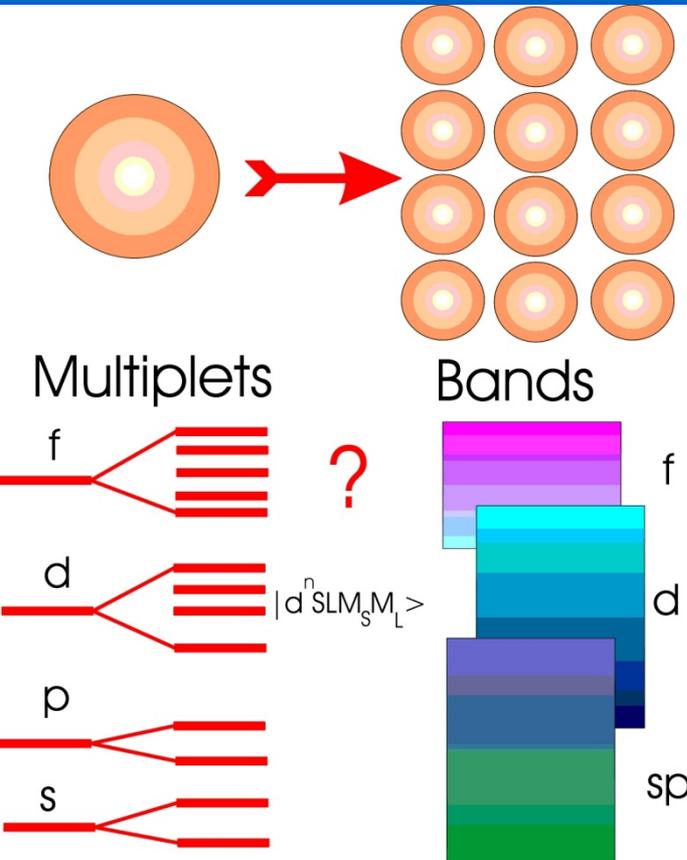


FIG. 1. Orbitaly decomposed NN exchange interaction in elemental 3d metals in the bcc structure.



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 narrow-band systems

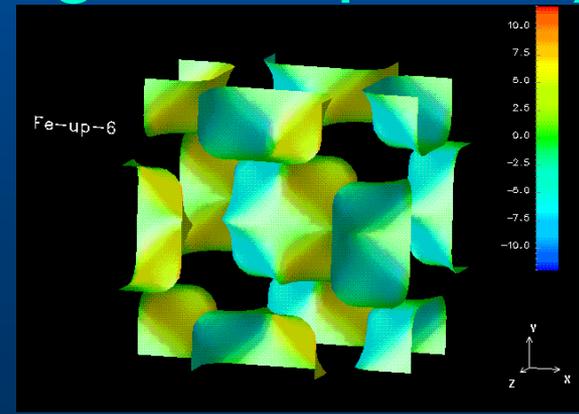
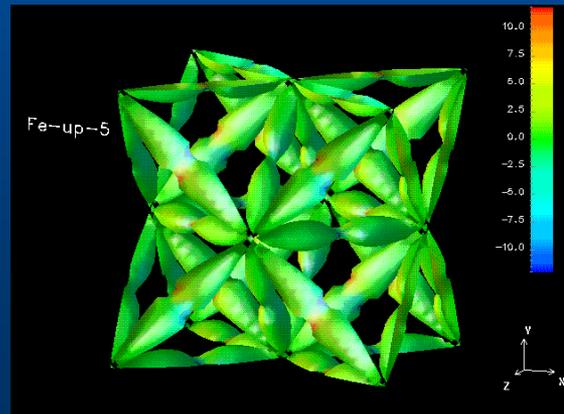
Problem with DFT: coexistence of localized and itinerant behavior



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



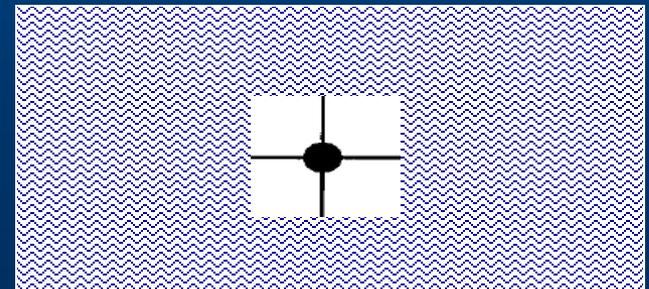
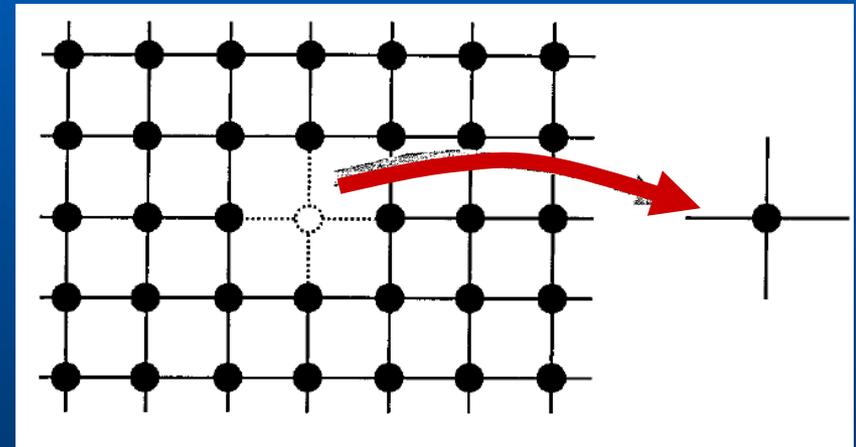
$4f$ electrons are normally pure localized but not $3d$

Dynamical Mean Field Theory I

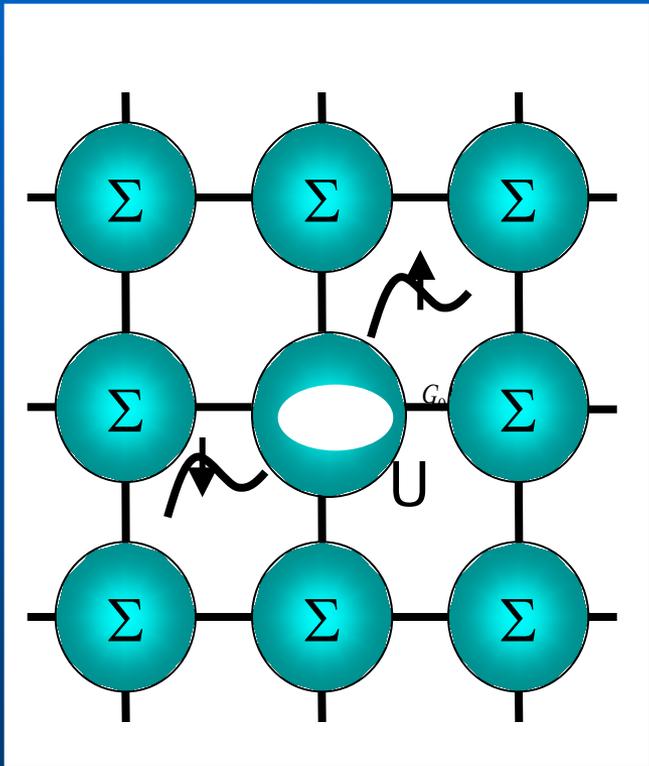
A.Georges, G.Kotliar, W.Krauth and M.Rozenberg, Rev. Mod. Phys. '96

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

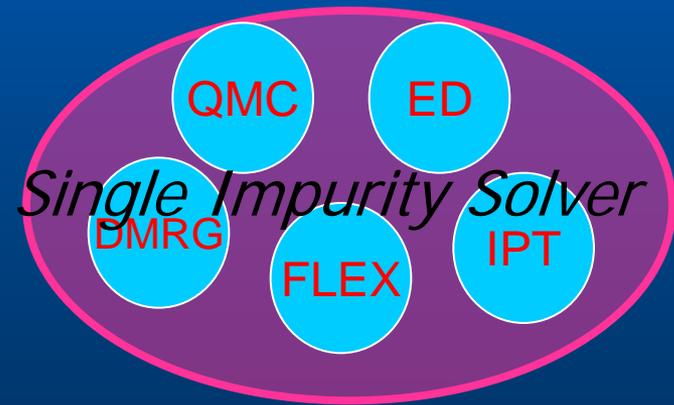


Dynamical Mean Field Theory II



$$\hat{G}(i\omega_n) = \frac{1}{\Omega} \sum_{\vec{k}}^{BZ} \hat{G}(\vec{k}, i\omega_n)$$

$$\hat{G}_0^{-1}(i\omega_n) = \hat{G}^{-1}(i\omega_n) + \hat{\Sigma}(i\omega_n)$$



$$\hat{\Sigma}_{new}(i\omega_n) = \hat{G}_0^{-1}(i\omega_n) - \hat{G}^{-1}(i\omega_n)$$

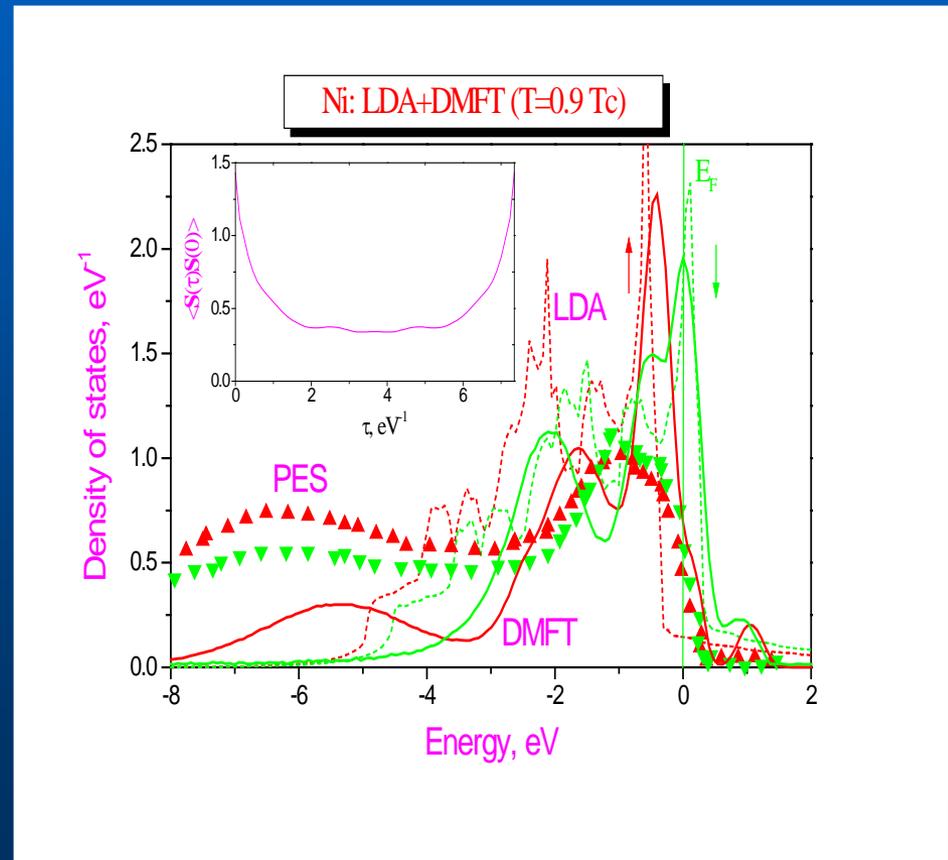
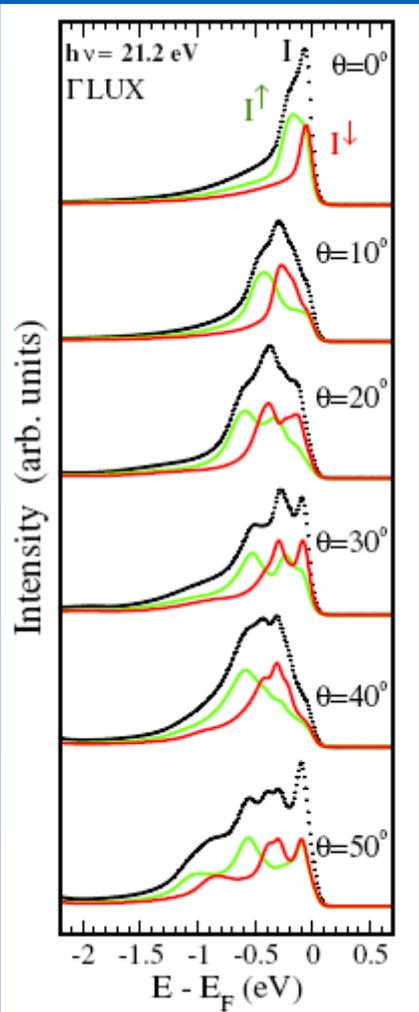
W. Metzner and D. Vollhardt (1987)
A. Georges and G. Kotliar (1992)

Ferromagnetism of transition metals: LDA+DMFT

Ferromagnetic Ni DMFT vs. LSDA:

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

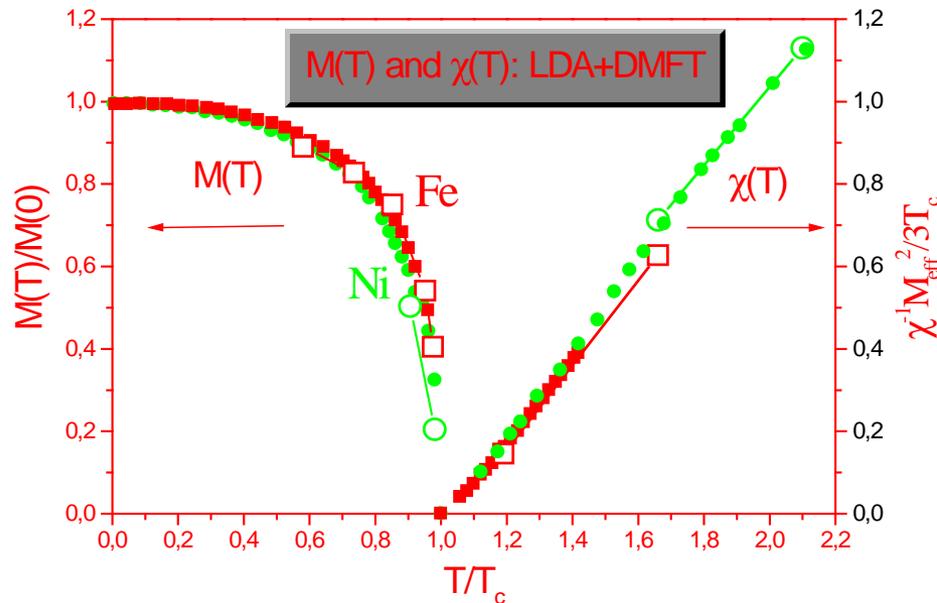
LDA+DMFT with ME
J. Braun *et al*
PRL (2006)



Lichtenstein, MIK, Kotliar, PRL (2001)

DMFT Effective Magnetic Moments: $T > T_c$

VV	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



Exchange and LW Functional

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

Luttinger-Ward functional

Magnetic force
theorem

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

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

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

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

$$\delta_1 \Omega_{sp} = \delta \Omega_{dc} = Tr G \delta \Sigma$$

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

Exchange interactions from DMFT

Heisenberg exchange:

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

Magnetic torque:

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

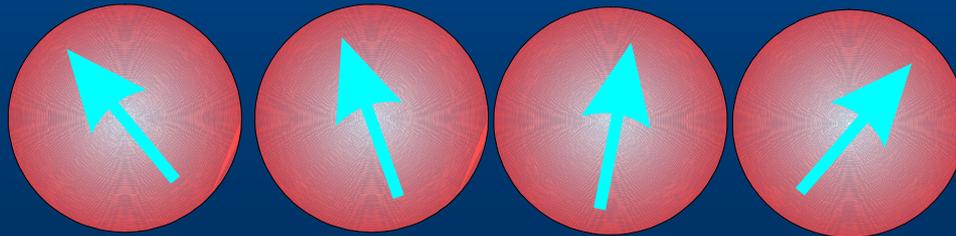
$$\delta \Omega = \delta^* \Omega_{sp} = \mathbf{V}_i \delta \varphi_i$$

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

Exchange interactions:

$$J_{ij} = -\text{Tr}_{\omega L} \left(\Sigma_i^s \mathbf{G}_{ij}^\uparrow \Sigma_j^s \mathbf{G}_{ji}^\downarrow \right)$$

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

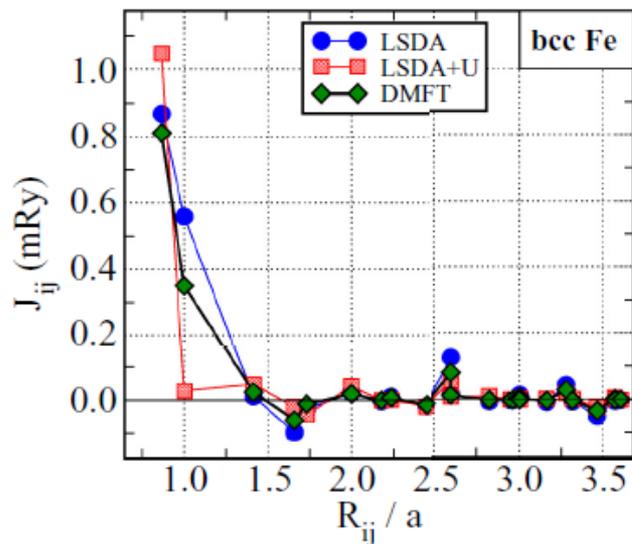


Applications

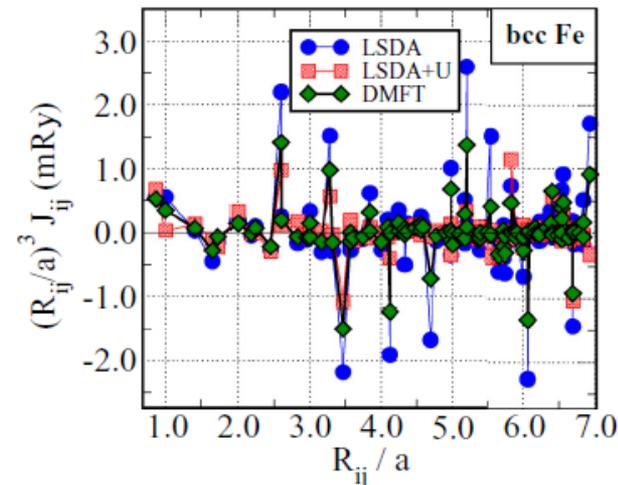
PHYSICAL REVIEW B 91, 125133 (2015)

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

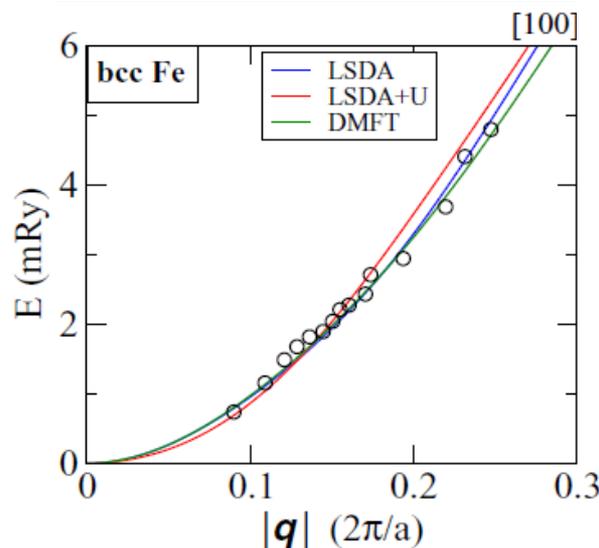
Y. O. Kvashnin,¹ O. Grånäs,^{1,2} I. Di Marco,¹ M. I. Katsnelson,^{3,4} A. I. Lichtenstein,^{4,5} and O. Eriksson¹



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



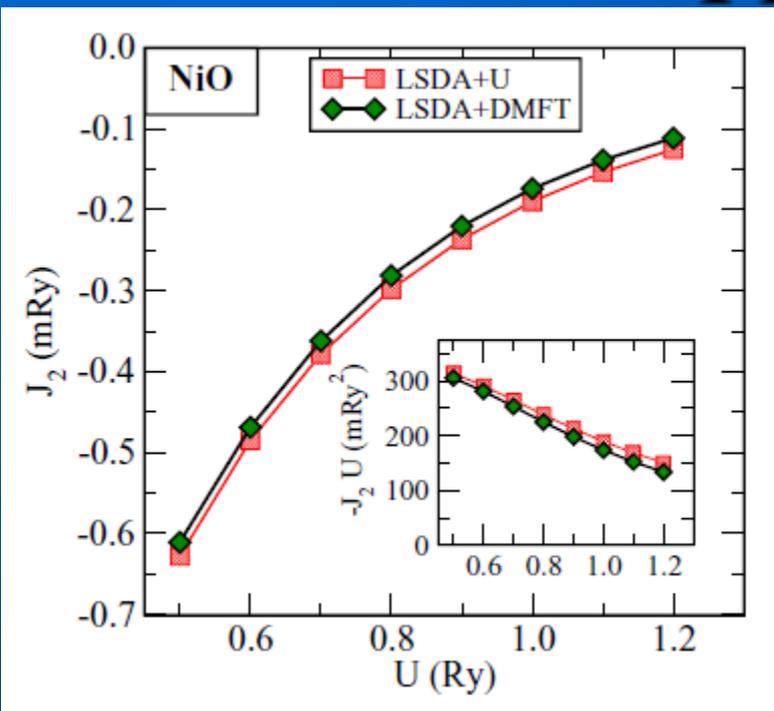
*Nontrivial: electronic
structure is very
different!*



← Spin-wave spectrum

Error cancellation?!

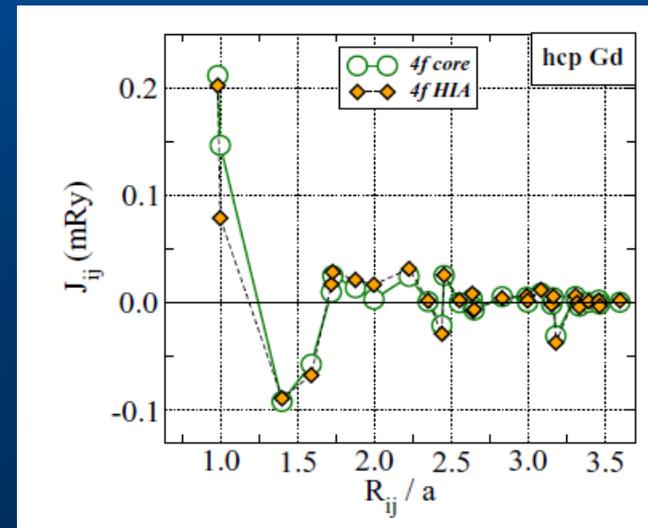
Applications II



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

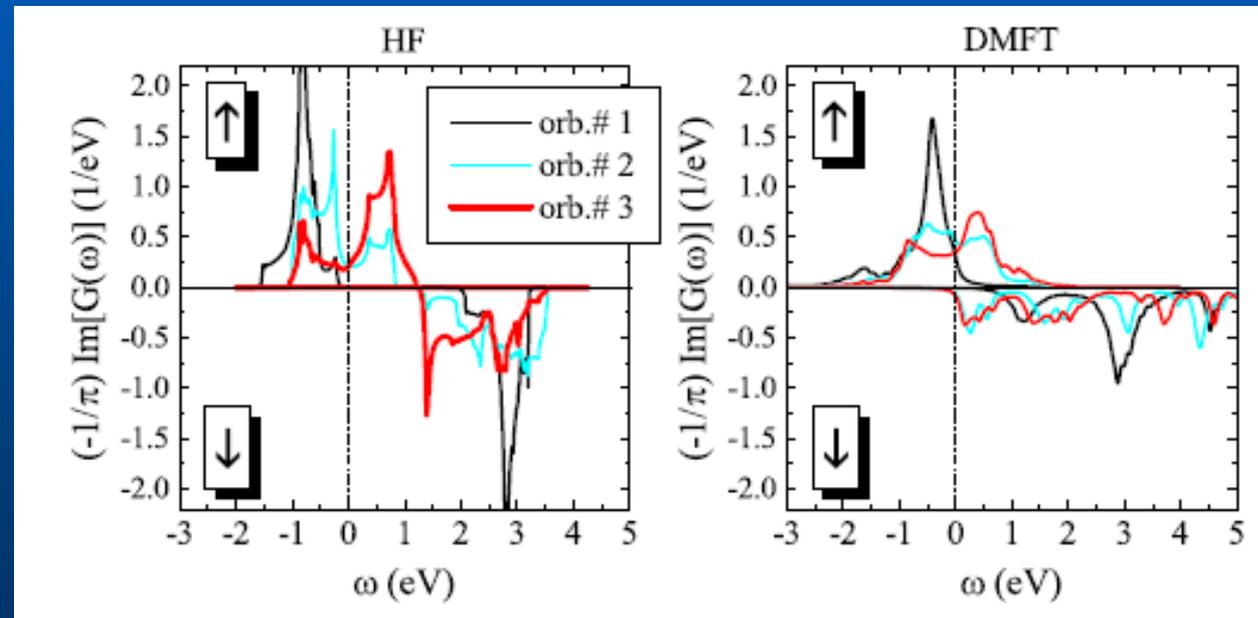
Applications III

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. Mazurenko²

*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

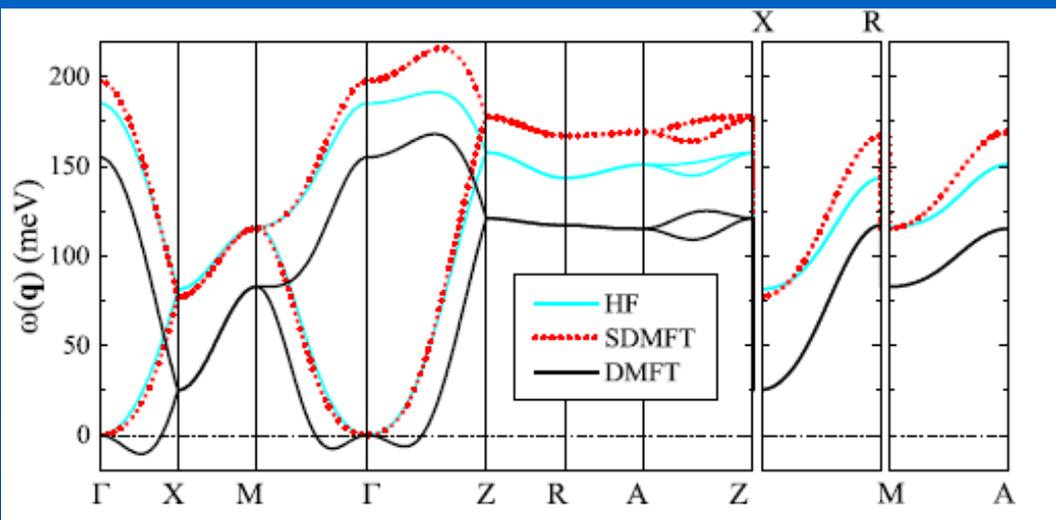
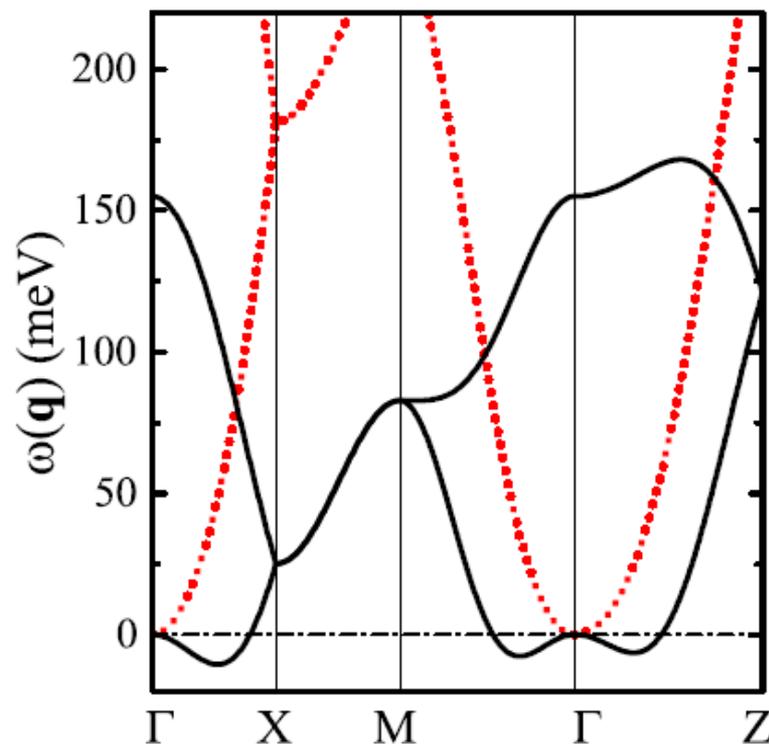


FIG. 9. (Color online) Results of calculations of the spin-wave dispersion with the DMFT parameters obtained for the isolated t_{2g} band (solid line) and after taking into account the additional FM contribution $\Delta J_2 = 17.81$ meV, arising from magnetic polarization of the oxygen band and direct 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

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

*DM interactions
(weak FM, etc.)*

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

Small rotations

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

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

Dzyaloshinskii-Moriya interactions II

Starting from collinear configuration

$$\begin{aligned}\delta\hat{H}_t &= \sum_{ij} c_i^+ (\delta\hat{R}_i^+ \hat{t}_{ij} + \hat{t}_{ij} \delta\hat{R}_j) c_j \\ &= -i \sum_{ij} c_i^+ (\delta\vec{\varphi}_i \hat{J} \hat{t}_{ij} - \hat{t}_{ij} \hat{J} \delta\vec{\varphi}_j) c_j \\ &= -\frac{i}{2} \sum_{ij} c_i^+ (\delta\vec{\varphi}_i - \delta\vec{\varphi}_j) (\hat{J} \hat{t}_{ij} + \hat{t}_{ij} \hat{J}) c_j\end{aligned}$$

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

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

FeBO₃

LETTERS

PUBLISHED ONLINE: 9 FEBRUARY 2014 | DOI: 10.1038/NPHYS2859

nature
physics

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

Measuring the Dzyaloshinskii–Moriya interaction in a weak ferromagnet

V. E. Dmitrienko¹, E. N. Ovchinnikova², S. P. Collins^{3*}, G. Nisbet³, G. Beutier⁴, Y. O. Kvashnin⁵,
V. V. Mazurenko⁶, A. I. Lichtenstein⁷ and M. I. Katsnelson^{6,8}

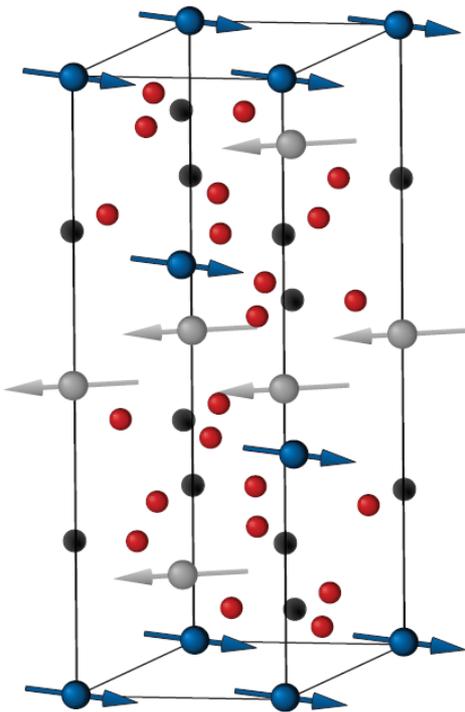


TABLE I. Calculated values of isotropic exchange interactions between magnetic moments in FeBO₃ (in meV). The number in parentheses denotes the coordination sphere.

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.*

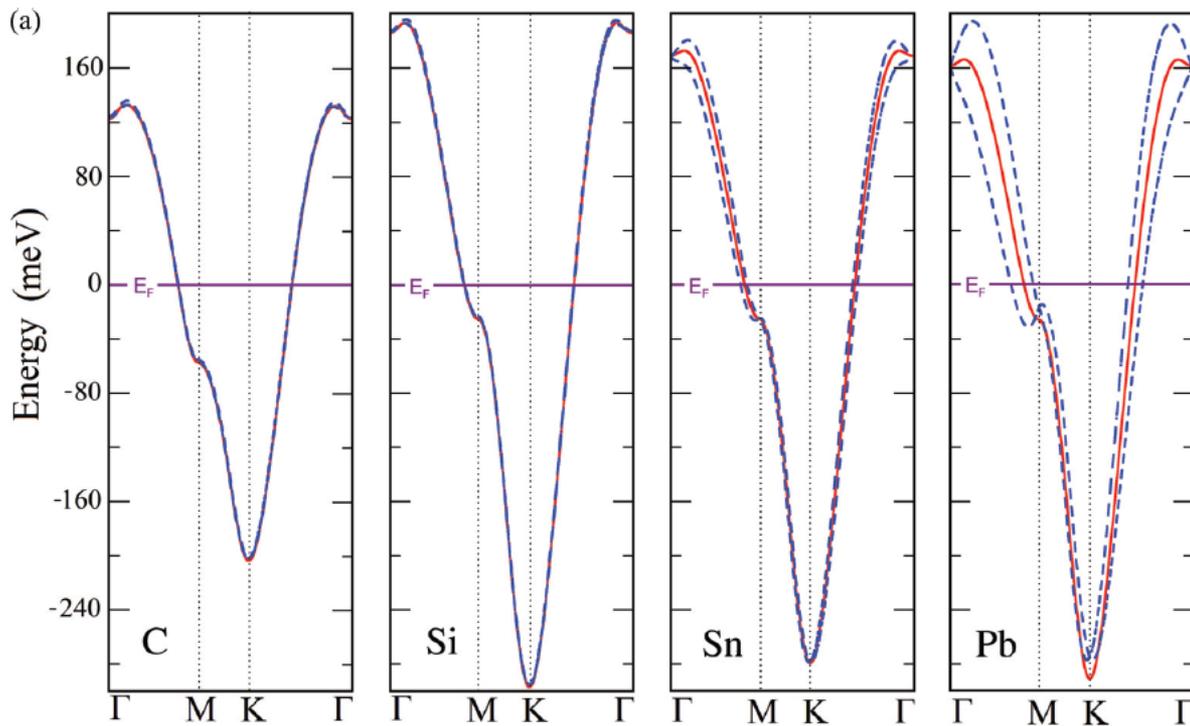
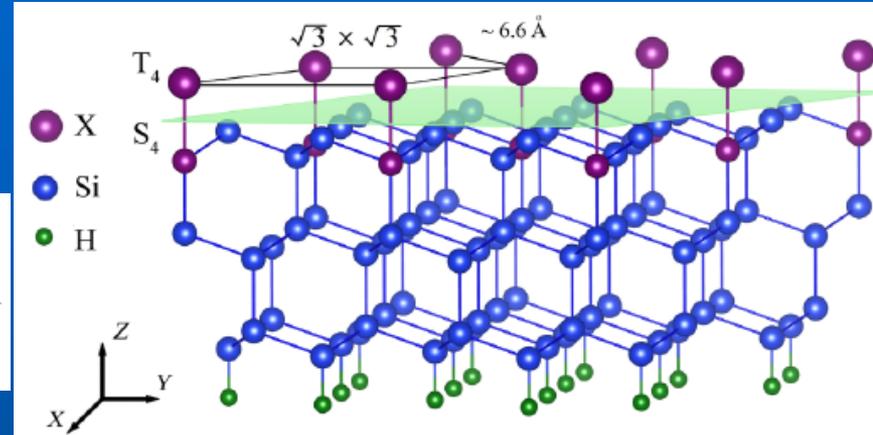
Si(111):X (X=C, Si, Sn, Pb)

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. Mazurenko¹



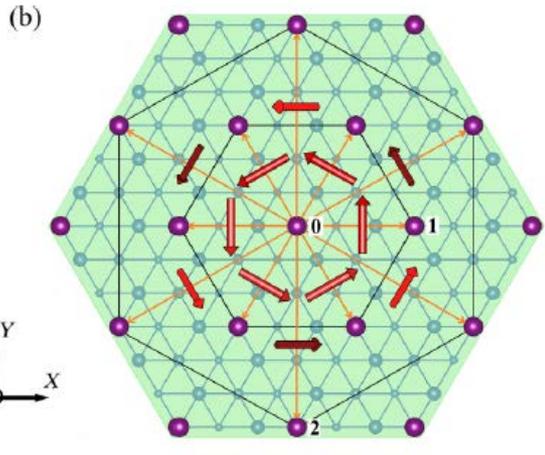
Single narrow band near the Fermi energy

*Red – without SO
Blue – with SO*

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

Mott insulator if take into account Hubbard U

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

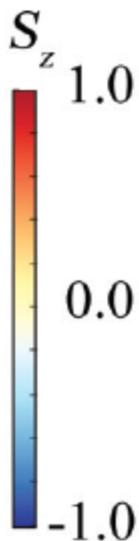
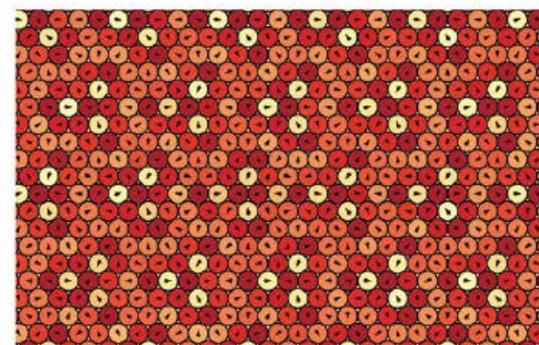
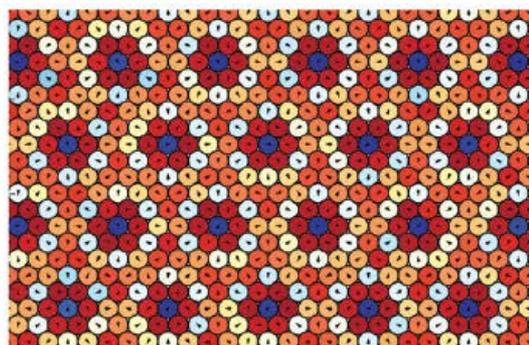
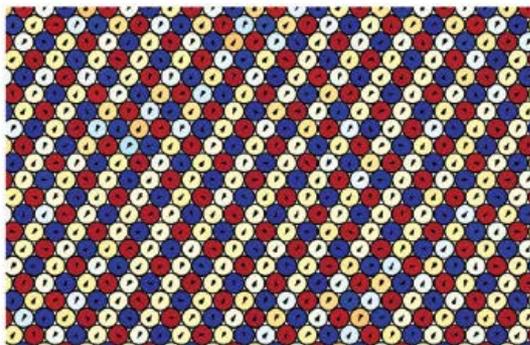


Orientation of DMI

$$h / J_{01} = 0.0$$

$$h / J_{01} = 3.6$$

$$h / J_{01} = 6.2$$



SiC(0001):Sn

Bartdinov, Nikolaev, Rudenko, Katsnelson, Mazurenko, 2018

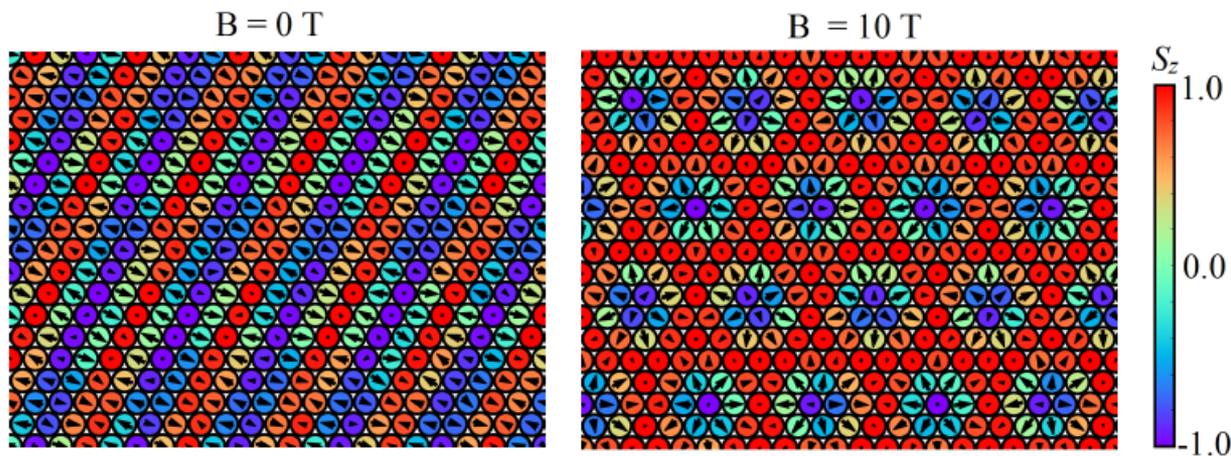
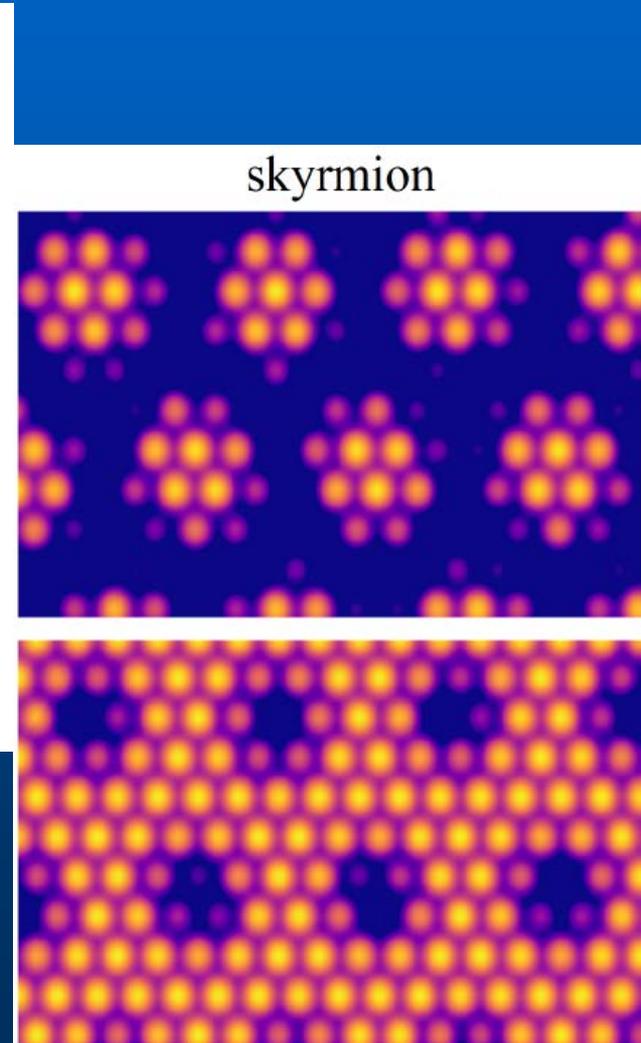


FIG. 3. Snapshots of the Sn/SiC(0001) spin texture obtained within Monte Carlo approach for different values of external magnetic field. The simulation temperature is 36 mK.

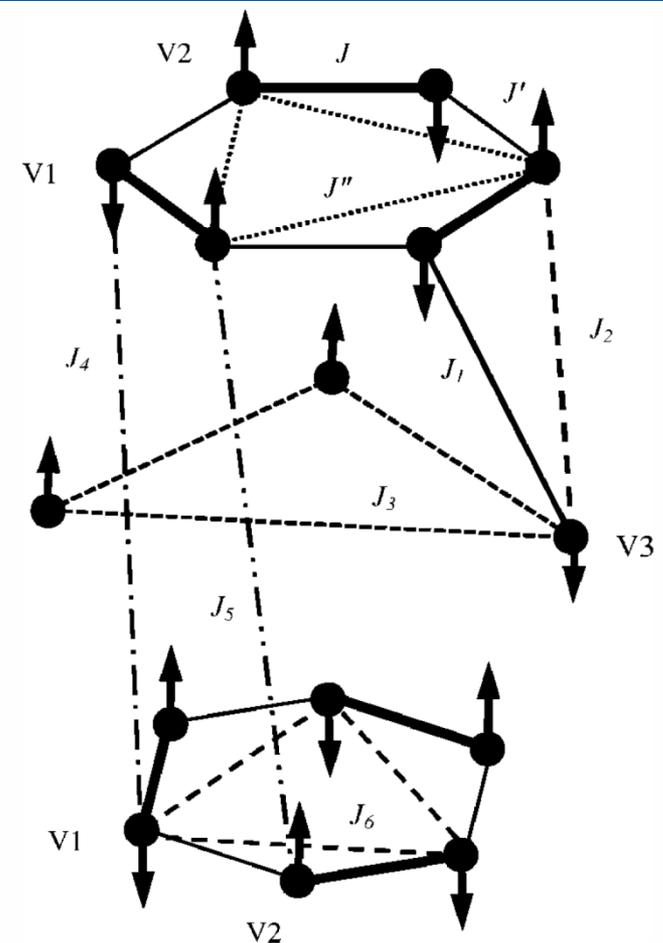
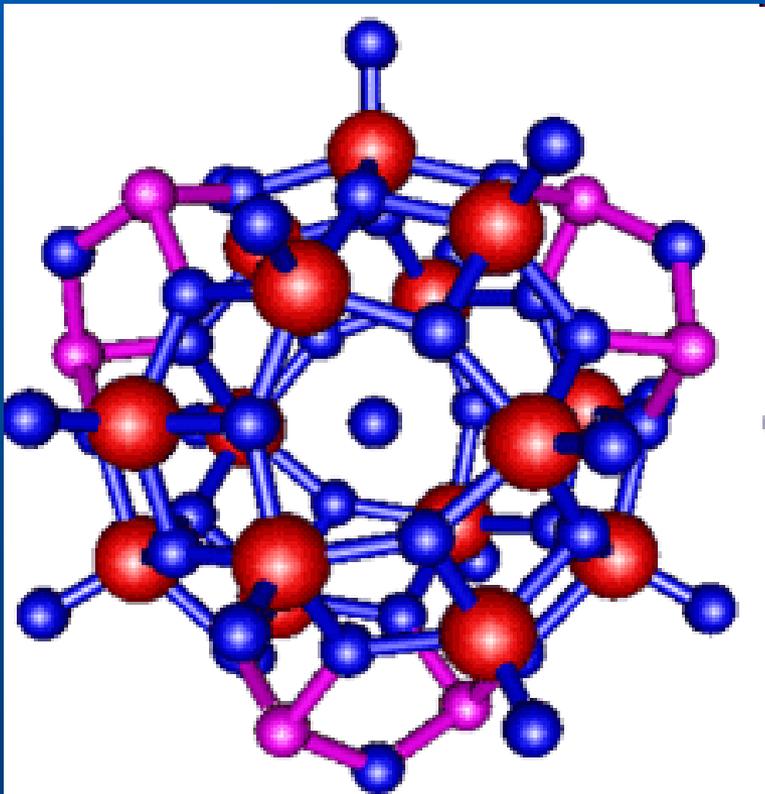


Simulated spin-polarized STM images for different tip polarization

Molecular magnets

Example: V_{15}

AFM ground state $S = 1/2$



LDA+U calculations

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

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

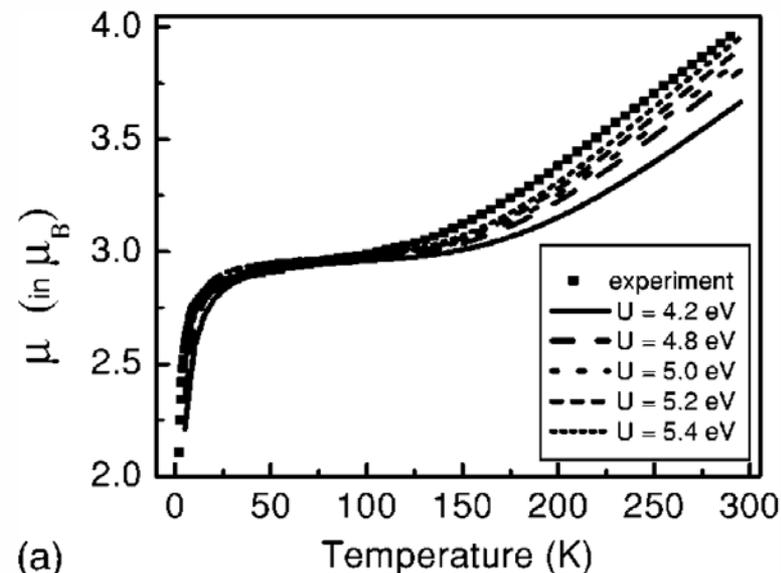
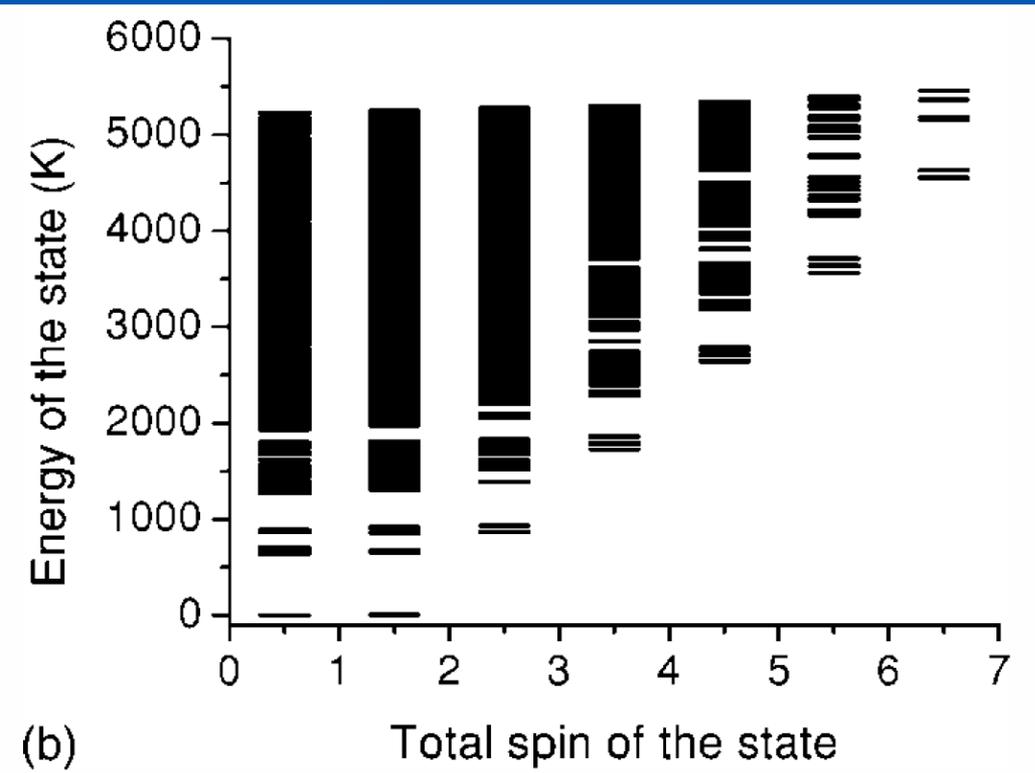
D. W. Boukhvalov,^{1,2} V. V. Dobrovitski,³ M. I. Katsnelson,^{2,4} A. I. Lichtenstein,⁵ B. N. Harmon,³ and P. Kögerler³

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.

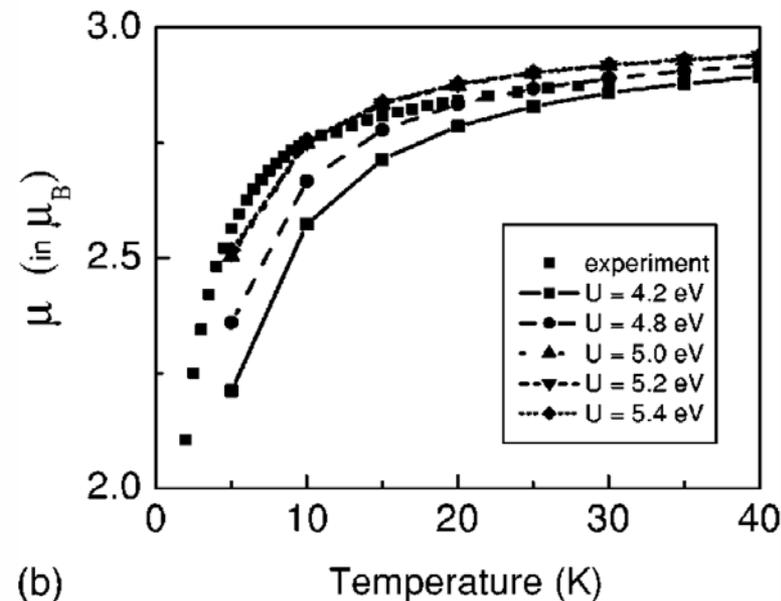
parameter	AFM1	AFM2	FM
J	-910	-905	-942
J'	-45	-46	-53
J''	-136	-139	-156
J_1	-219	-247	-255
J_2	-134	-128	-132
J_3	-5	-5	-6
J_4	-13	-12	-15
J_5	-3	-3	-3
J_6	-3	-3	-3
gap	1.08	1.02	1.16
μ_{V1}	-0.94	-0.93	-0.99
μ_{V2}	+0.91	+0.92	-0.97
μ_{V3}	-1.00	+0.97	-1.00

LDA+U calculations II

Exact diagonalization
for Heisenberg model



(a)



(b)

Mn₁₂: full calculations

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

First-principles modeling of magnetic excitations in Mn₁₂

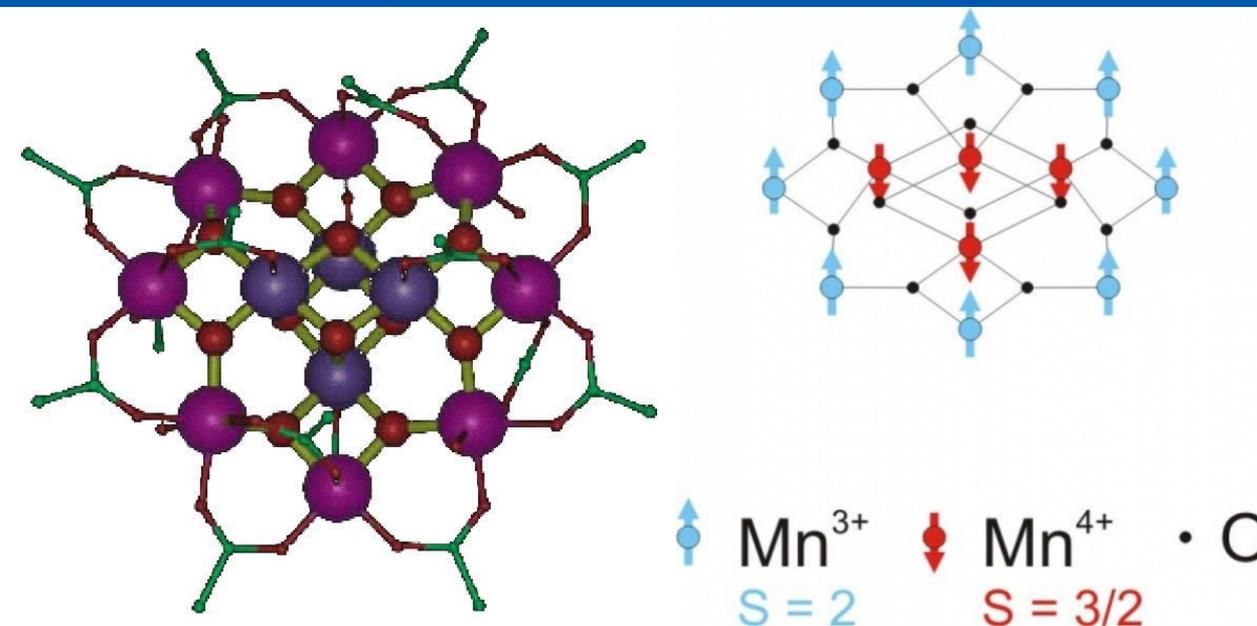
V. V. Mazurenko,¹ Y. O. Kvashnin,^{2,3} Fengping Jin,⁴ H. A. De Raedt,⁵ A. I. Lichtenstein,⁶ and M. I. Katsnelson^{1,7}

Motivation

The prototype molecular magnet

Dimension of Hilbert space:
 $(2 \times 2 + 1)^8 (2 \times 3/2 + 1)^4 = 10^8$

A real challenge!



Mn₁₂: full calculations II

Inelastic neutron 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 10⁴

Cannot be justified quantitatively!

Full LDA+U calculations plus Lanczos ED

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

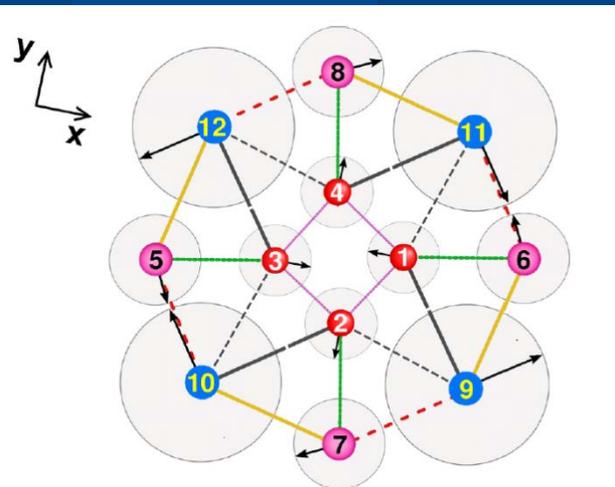


TABLE I. Intramolecular isotropic exchange interaction parameters (in meV) calculated by using the LDA + U approach. Positive sign corresponds to the antiferromagnetic coupling.

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	

Mn₁₂: 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 i th and j th 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...

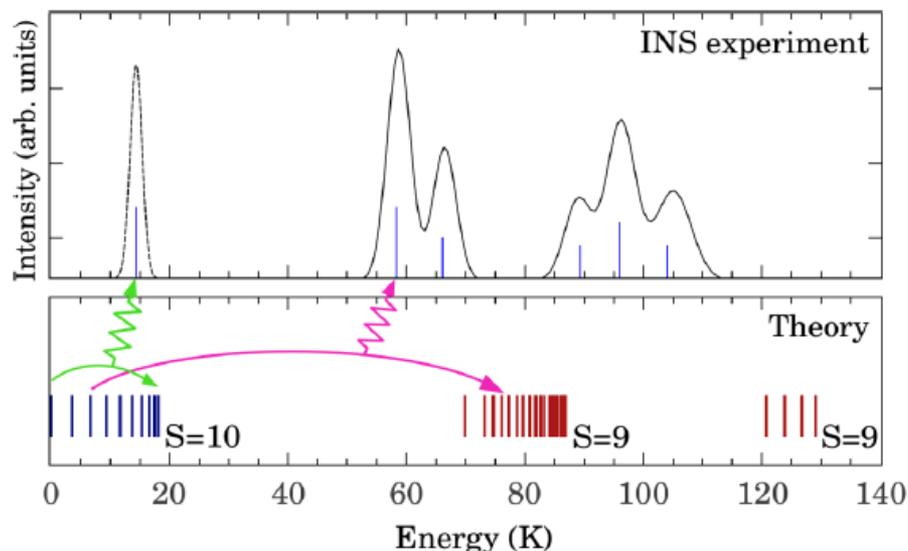


FIG. 2. (Color online) Schematic comparison of the theoretical spectrum obtained by diagonalizing Eq. (1) and INS spectrum taken from Ref. [12] (Figs. 6 and 8 therein). The arrows denote the intra- and interband transitions that correspond to the excitations observed in the INS experiment.

No fitting parameters at all – not so bad!

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*



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. Katsnelson¹

One can reach a regime when $D \gg J$ with unusual nanoskyrmion mosaic (Stepanov, Nikolaev, Dutreix, MIK, Mazurenko, 2018)

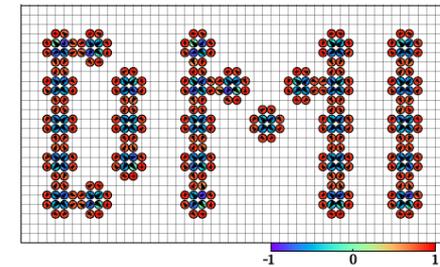
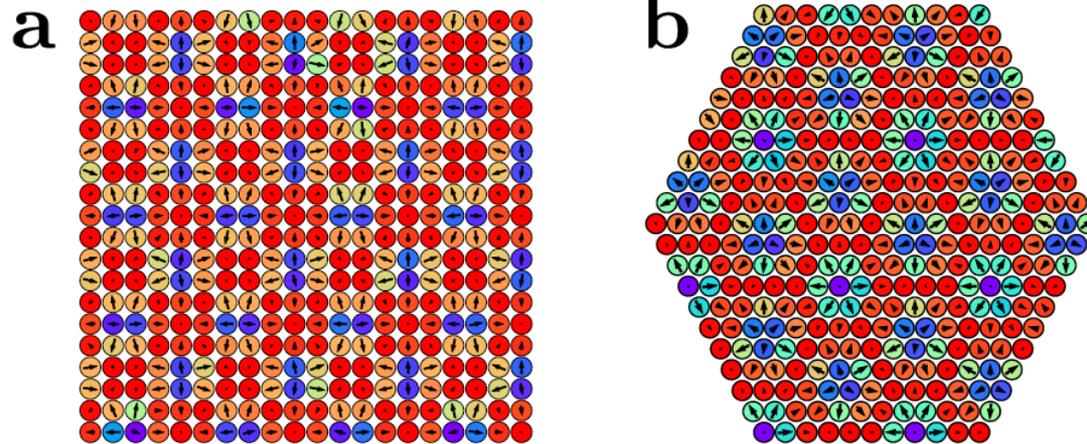


FIG. 1. Nanoskyrmion-designed DMI abbreviation obtained from Monte Carlo simulations of the Heisenberg-exchange-free model on the non-regular square lattice with $B_z = 1.2$. Arrows and colors depict the in-plane and out-of-plane spin projections, respectively.

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 $T = 0.01 |D|$.

Beyond the talk

Finite-temperature effects

Ab initio spin dynamics for real systems

Intermediate level: TB spin dynamics

Spin and orbital magnetism

Nonequilibrium 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