Radboud Universiteit

Theory of magnetic interactions in real materials

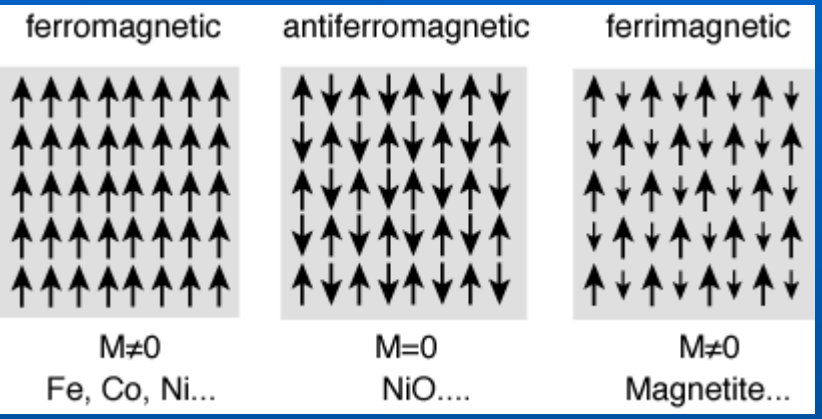
Mikhail Katsnelson

IMM

Institute for Molecules and Materials

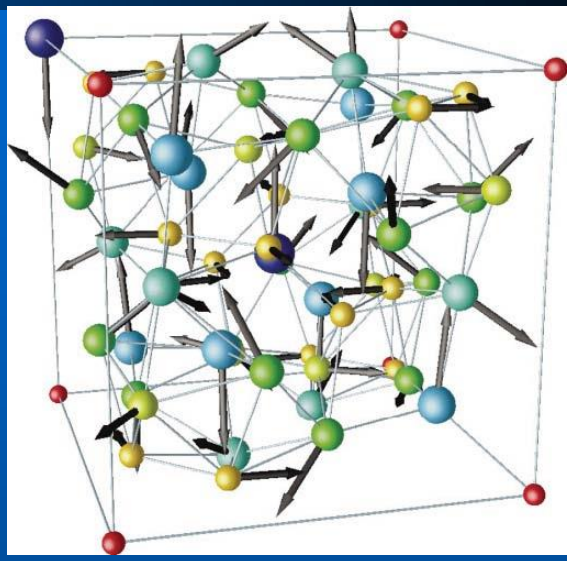
Types of magnetic ordering

Textbook wisdom

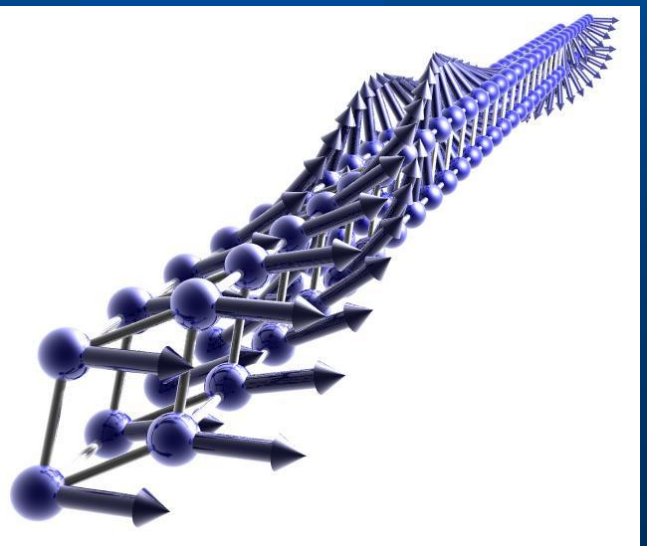


Sometimes very complicated

α-Mn

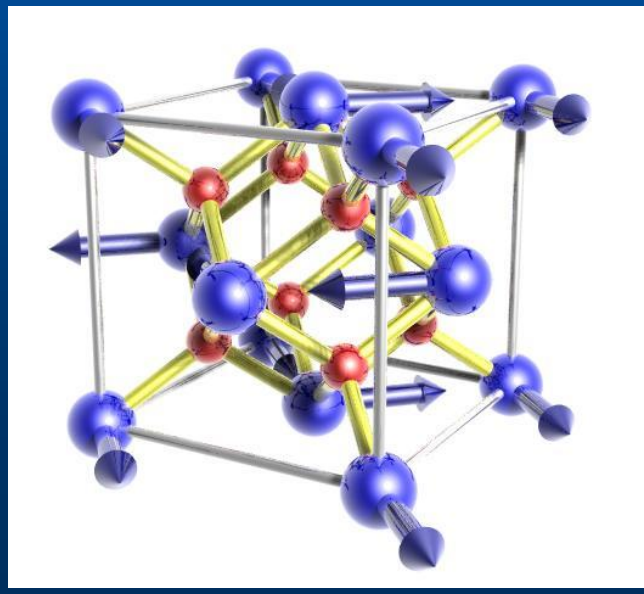


Spin spirals



γ-Fe

UO_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.

Density Functional Theory

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

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

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

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

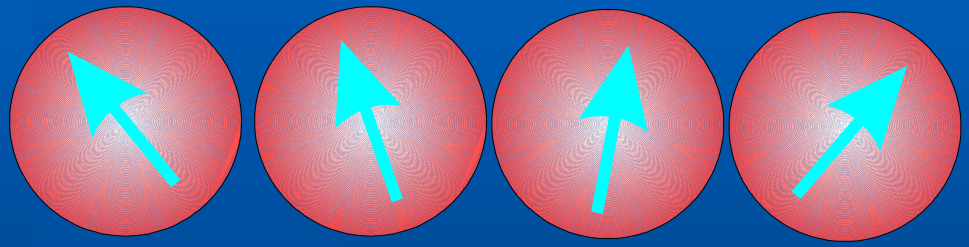
B is self-consistent magnetic field

The question: how to map density functional on classical spin Hamiltonian?

Magnetic force theorem

(Lichtenstein,MIK, Gubanov, J. Phys. F 1984; Sol. St. Comm. 1985;
Lichtenstein, MIK, Antropov, Gubanov JMMM 1987)

Basic idea: consider the variation of total energy at small rotations of local magnetic moments



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

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

Green-function functionals (GW, DMFT…)

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

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

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

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

Magnetic force theorem

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

Magnetic torque

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

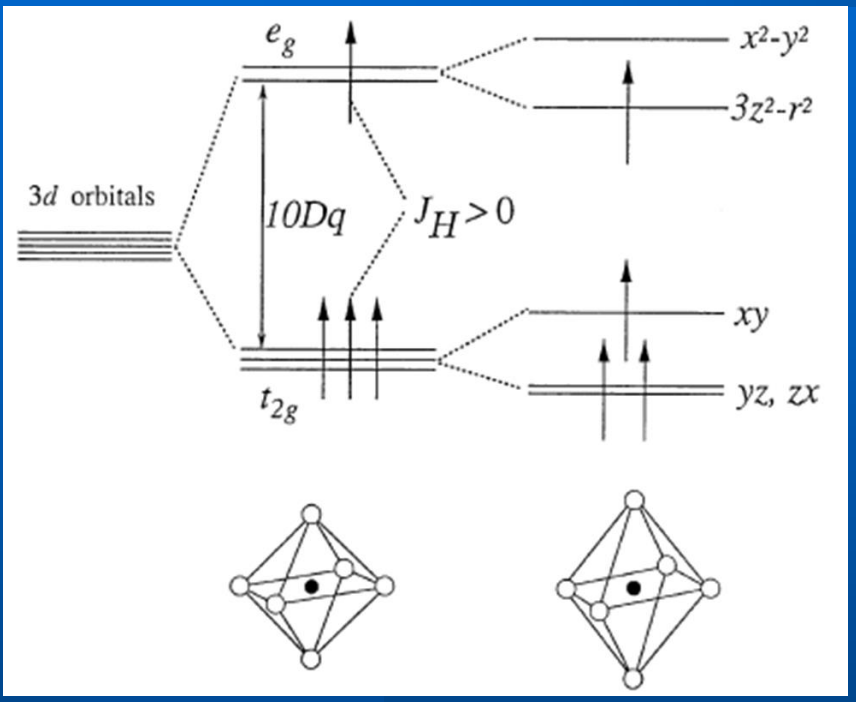
$$\mathbf{V}_i = 2Tr_{\omega L}\left[\mathbf{\Sigma}^s_i \times \mathbf{G}^s_{ii}\right]$$

Exchange interactions with local Sigma

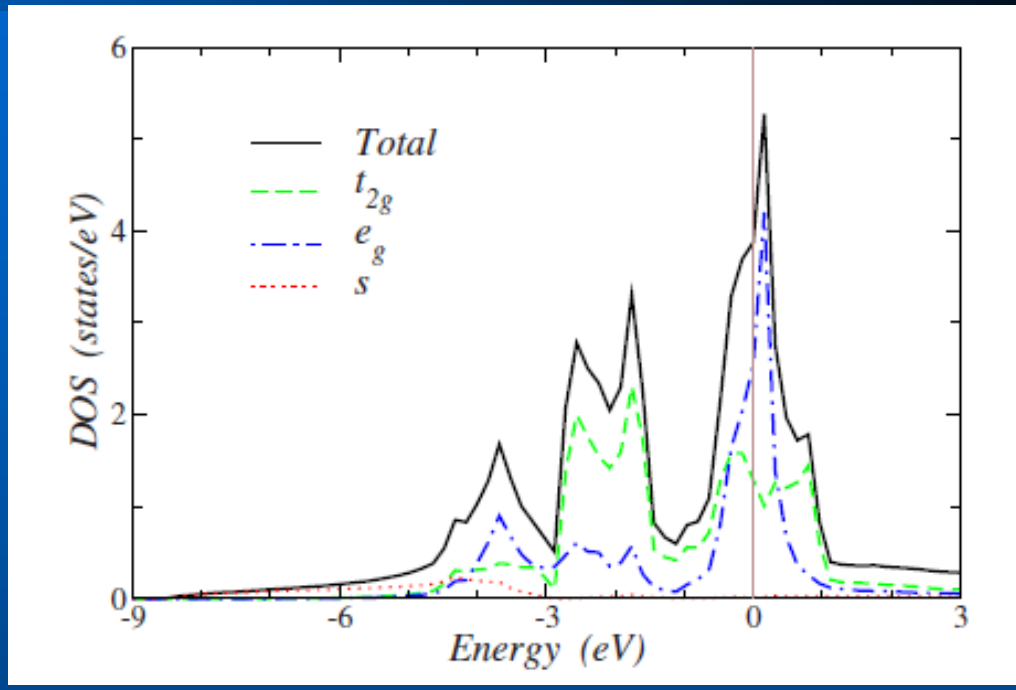
$$J_{ij} = -Tr_{\omega L}\left(\Sigma^s_i G^{\uparrow}_{ij}\Sigma^s_j G^{\downarrow}_{ji}\right)$$

$$\Sigma^s_i = \frac{1}{2}\left(\Sigma^{\uparrow}_i - \Sigma^{\downarrow}_i\right)$$

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]

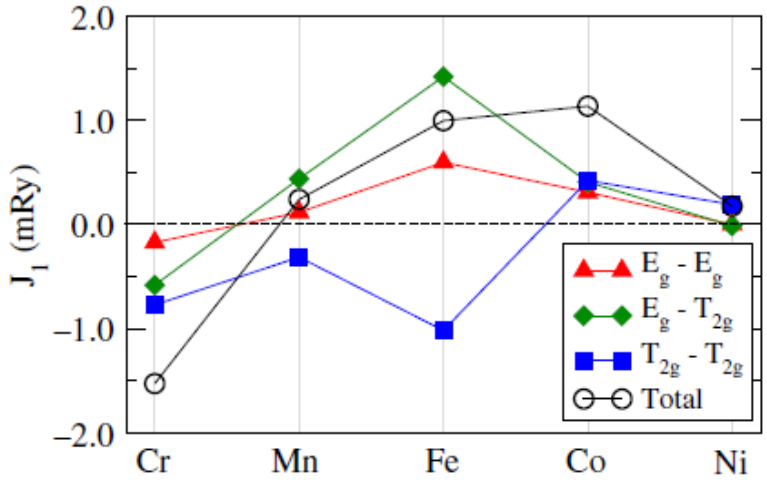
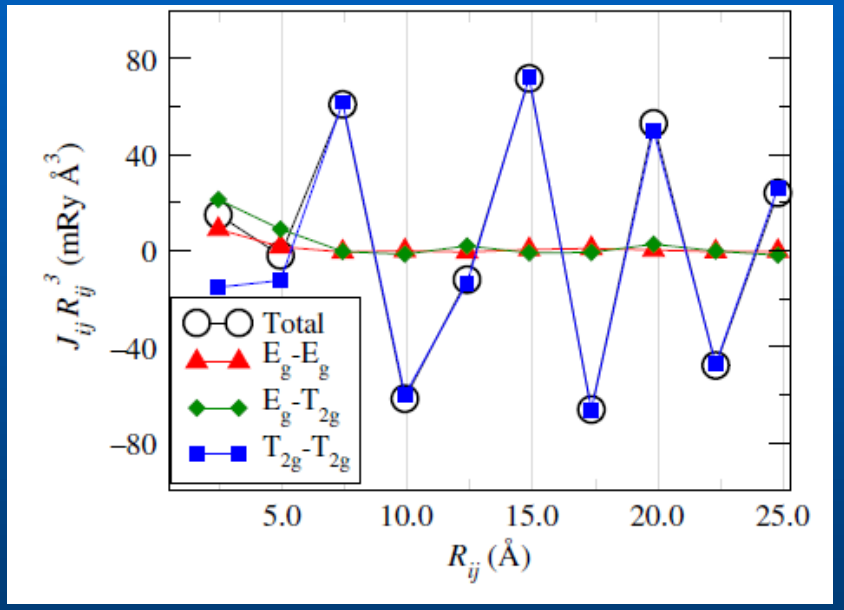


FIG. 1. Orbitally decomposed NN exchange interaction in elemental 3*d* metals in the bcc structure.



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

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)

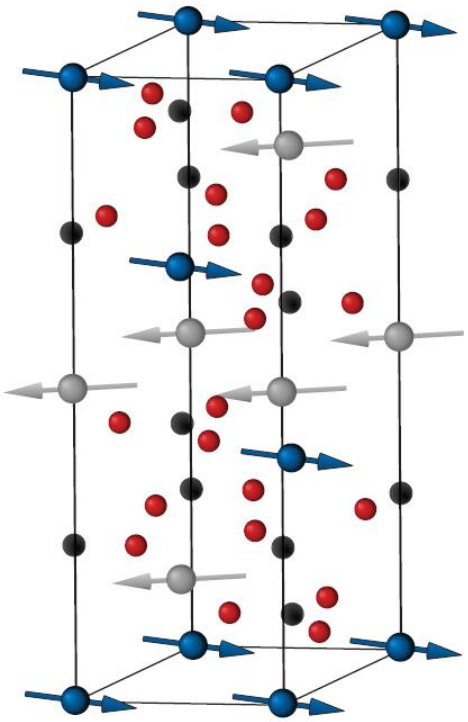


TABLE I. Calculated values of isotropic exchange interactions between magnetic moments in $FeBO_3$ (in meV). The number in parentheses denotes the coordination sphere.

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

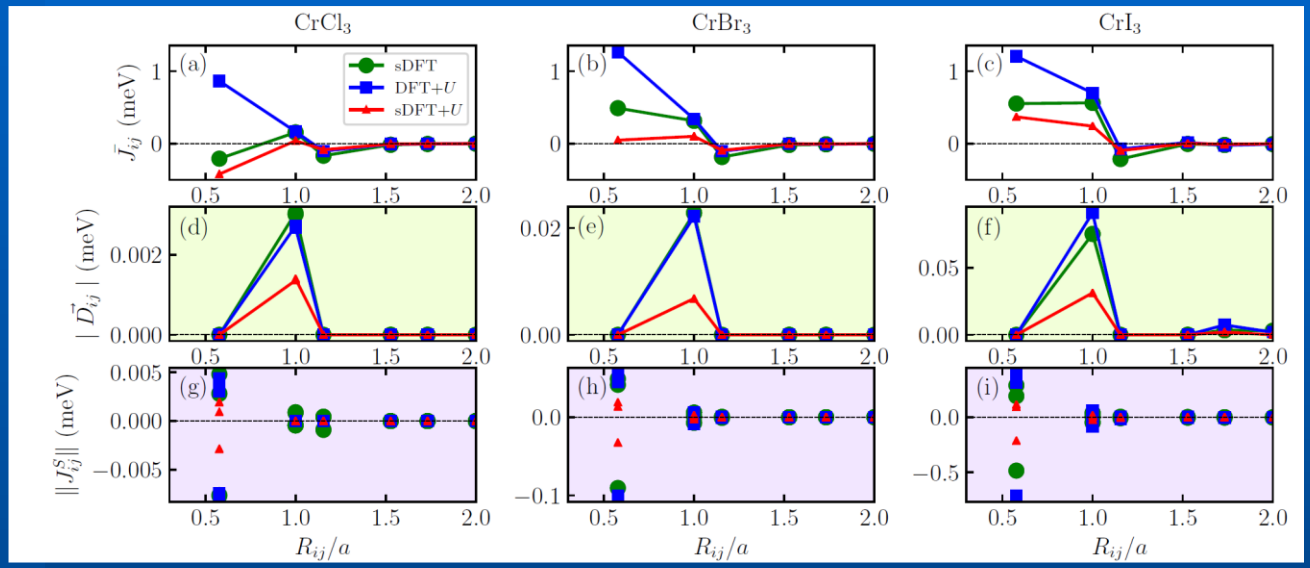
Agrees well with exper.

2D Magnets: CrX_3

Relativistic exchange interactions in CrX_3 (X = Cl, Br, I) monolayers

Y. O. Kvashnin,[1] A. Bergman,[1] A. I. Lichtenstein,[2,3,4] and M. I. Katsnelson[5]

PHYSICAL REVIEW B **102**, 115162 (2020)



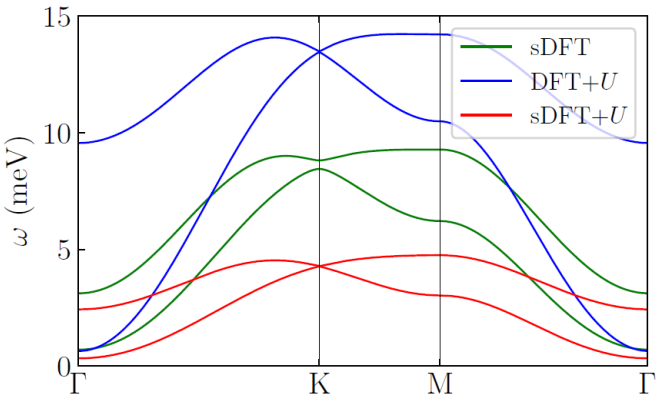


FIG. 6. Simulated adiabatic magnon spectra for monolayered CrI_3 using the parameters, obtained for three computational setups, shown in Fig. 5.

The results are quite sensitive to the method used to calculate electronic structure (GW is also Done, DMFT – work in progress)

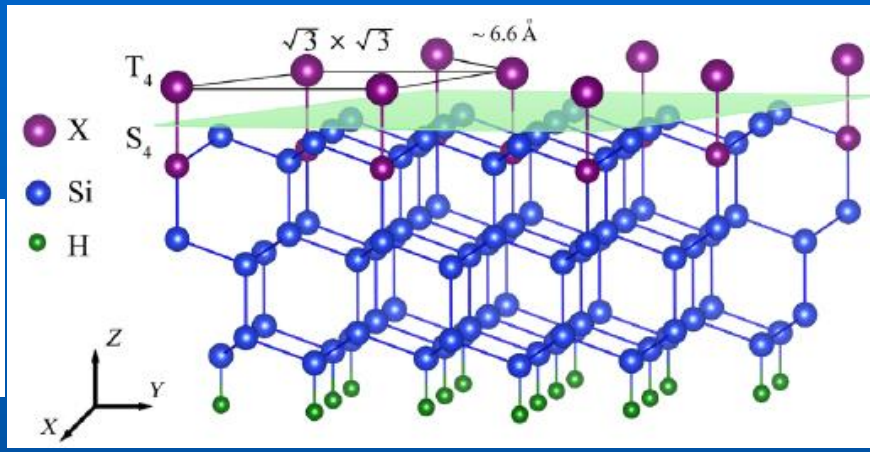
Si(111):X (X=C,Si,Sn,Pb)

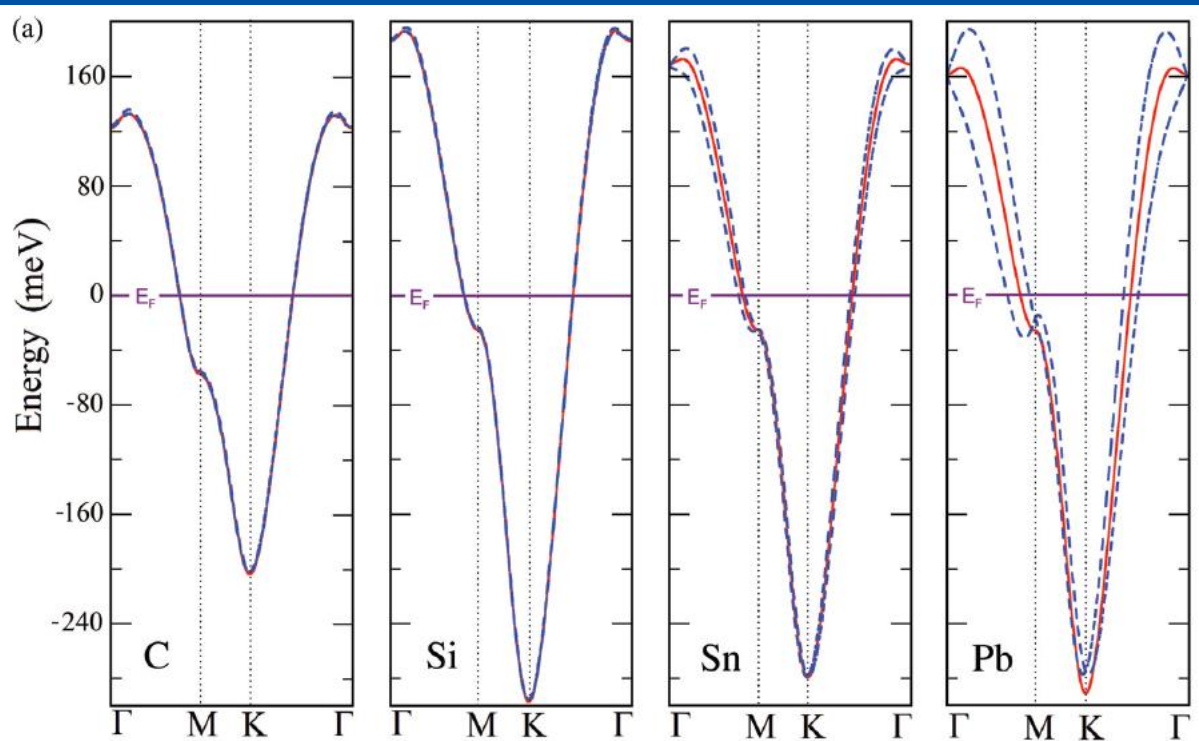
sp-electron magnets

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]



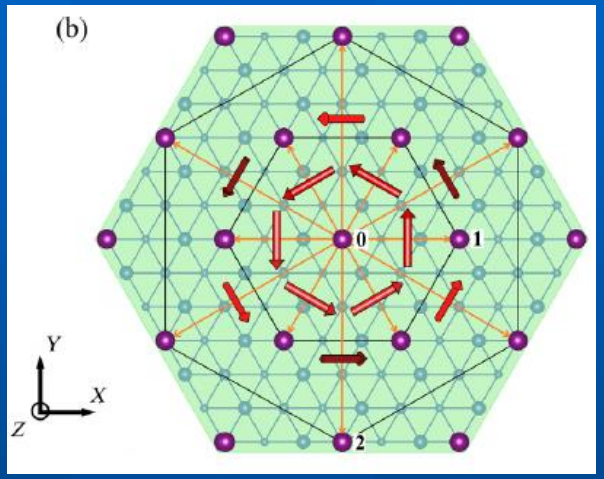


Single narrow band nea the Fermi energy

Red – without SO
Blue – with SO

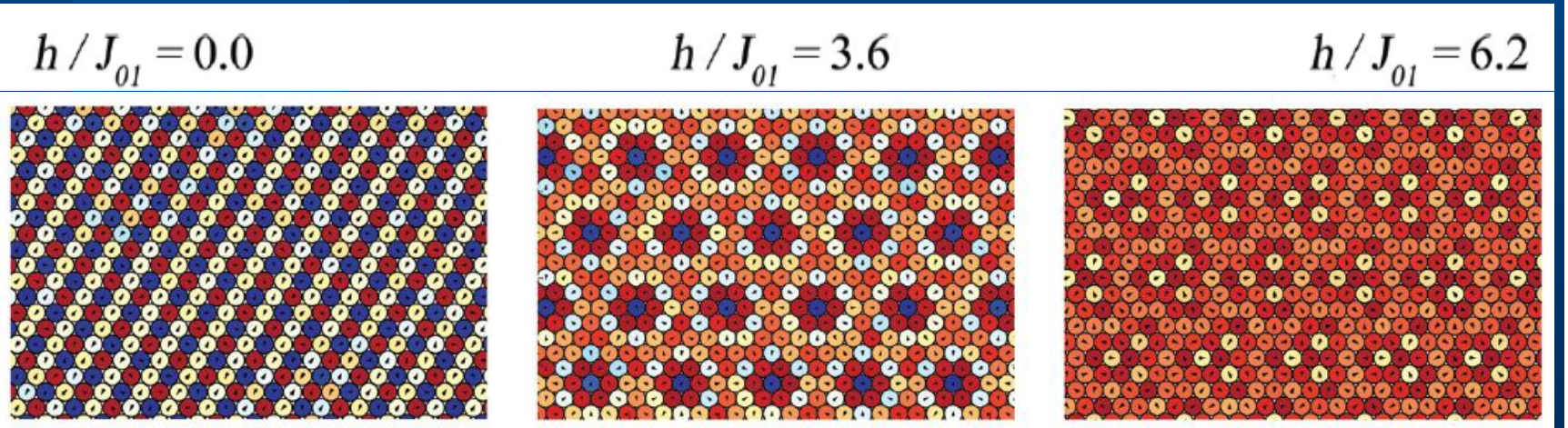
Si(111):X (X=C,Si,Sn,Pb) II

Mott insulator if take into account Hubbard U



Orientation of DMI

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



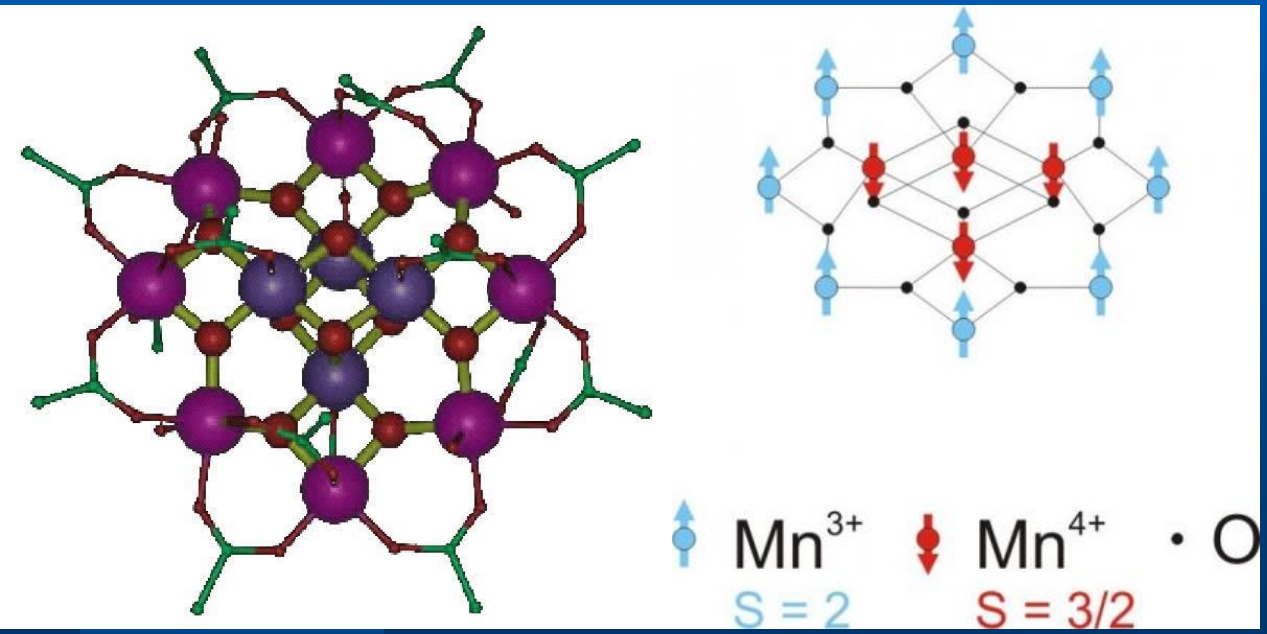
Mn_{12}: full calculations

PHYSICAL REVIEW B **89**, 214422 (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

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

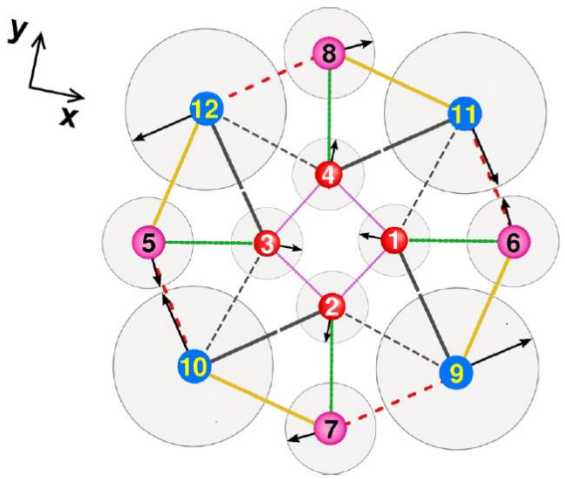


TABLE I. Intramolecular isotropic exchange interaction parameters (in meV) calculated by using the LDA + U approach. Positive sign corresponds to the antiferromagnetic coupling.

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...

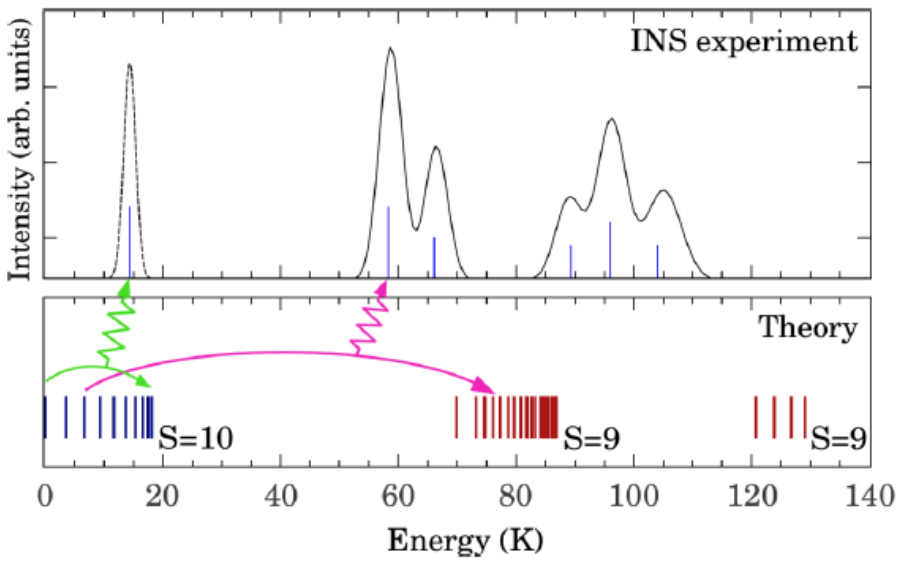


FIG. 2. (Color online) Schematic comparison of the theoretical spectrum obtained by diagonalizing Eq. (1) and INS spectrum taken from Ref. [12] (Figs. 6 and 8 therein). The arrows denote the intra- and interband transitions that correspond to the excitations observed in the INS experiment.

No fitting parameters at all – not so bad!

Manipulation of magnetic interactions by high-frequency laser field

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

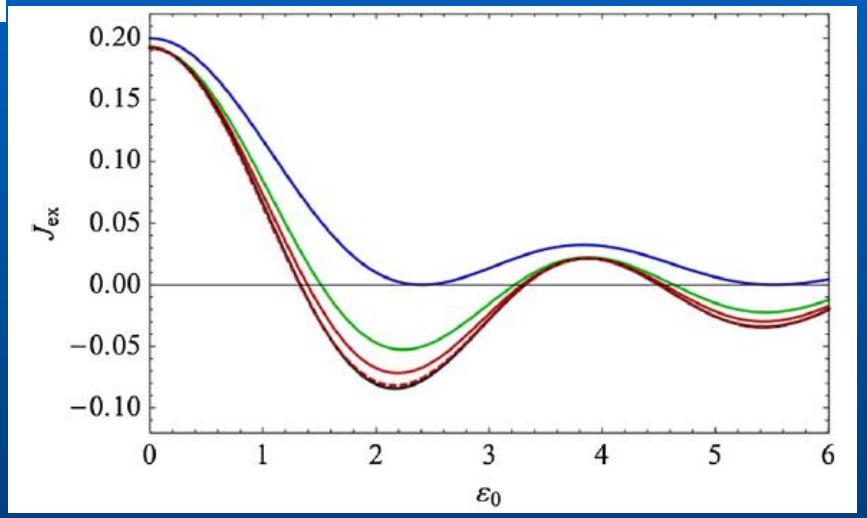
Effective Hamiltonians for Rapidly Driven Many-Body Lattice Systems: Induced Exchange Interactions and Density-Dependent Hoppings

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

One can change a sign of exchange integral

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

1D Hubbard model in strong high-frequency laser field



Color solid curves, from top to bottom:
- Bare exchange interaction
- Second order expansion
- Fourth order expansion
- Exact analytical solution

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

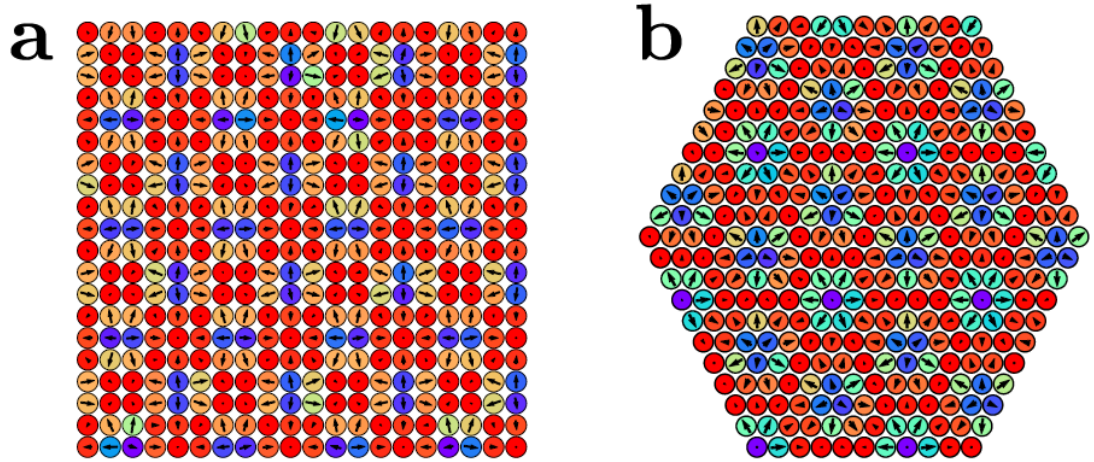
E. A. Stepanov,[1] C. Dutreix,[1,2] and M. I. Katsnelson[1]

Heisenberg-exchange-free nanoskyrmion mosaic

E A Stepanov[1,2,6], S A Nikolaev[2], C Dutreix[3,4,5], M I Katsnelson[1,2] and V V Mazurenko[2]

J. Phys.: Condens. Matter **31** (2019) 17LT01

One can reach a regime when D >> J with unusual nanockyrmione mosaic



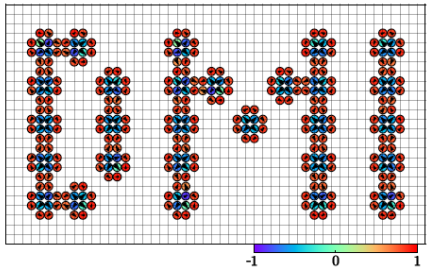


FIG. 1. Nanoskyrmion-designed DMI abbreviation obtained from Monte Carlo simulations of the Heisenberg-exchange-free model on the non-regular square lattice with $B_z = 1.2$. Arrows and colors depict the in-plane and out-of-plane spin projections, respectively.

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 $T = 0.01\,|\mathbf{D}|$.

Putting into a context of standard many-body theory

General way of mapping of interacting fermions onto bosonic fields (collective variables): dual boson approach

Dual boson approach to collective excitations in correlated fermionic systems

A.N. Rubtsov [a], M.I. Katsnelson [b], A.I. Lichtenstein [c,*]

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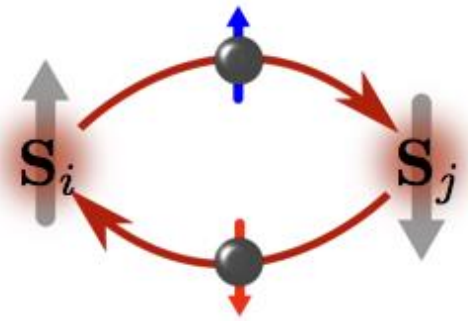
Derivation and generalization of the exchange formula via dual boson approach: “practically exact” for Hubbard model

PHYSICAL REVIEW LETTERS **121,** 037204 (2018)

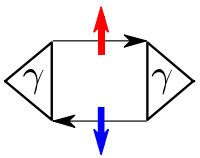
Effective Heisenberg Model and Exchange Interaction for Strongly Correlated Systems

E. A. Stepanov,[1,2] S. Brener,[3] F. Krien,[3] M. Harland,[3] A. I. Lichtenstein,[3,2] and M. I. Katsnelson[1,2]
[1]*Radboud University, Institute for Molecules and Materials, 6525AJ Nijmegen, Netherlands*
[2]*Theoretical Physics and Applied Mathematics Department, Ural Federal University, Mira Street 19, 620002 Ekaterinburg, Russia*
[3]*Institute of Theoretical Physics, University of Hamburg, 20355 Hamburg, Germany*

$$J_{ij} = J_{ij}^{\mathrm{d}} - \mathbf{S}_i \quad \mathbf{S}_j$$



$$J_{\mathbf{q}} = J_{\mathbf{q}}^{\mathrm{d}} - \sum_{\mathbf{k},\nu} \gamma^{-}_{\nu,\omega=0}\, \tilde{G}_{\mathbf{k}+\mathbf{q},\nu\uparrow} \tilde{G}_{\mathbf{k}\nu\downarrow}\, \gamma^{+}_{\nu,\omega=0}$$



Applications to nonmagnetic systems

Charge-ordered systems

PHYSICAL REVIEW B **99**, 115124 (2019)

Effective Ising model for correlated systems with charge ordering

E. A. Stepanov,[1,2] A. Huber,[3] A. I. Lichtenstein,[2,3] and M. I. Katsnelson[1,2]
[1]*Radboud University, Institute for Molecules and Materials, 6525AJ Nijmegen, The Netherlands*
[2]*Theoretical Physics and Applied Mathematics Department, Ural Federal University, Mira Street 19, 620002 Ekaterinburg, Russia*
[3]*Institute of Theoretical Physics, University of Hamburg, 20355 Hamburg, Germany*

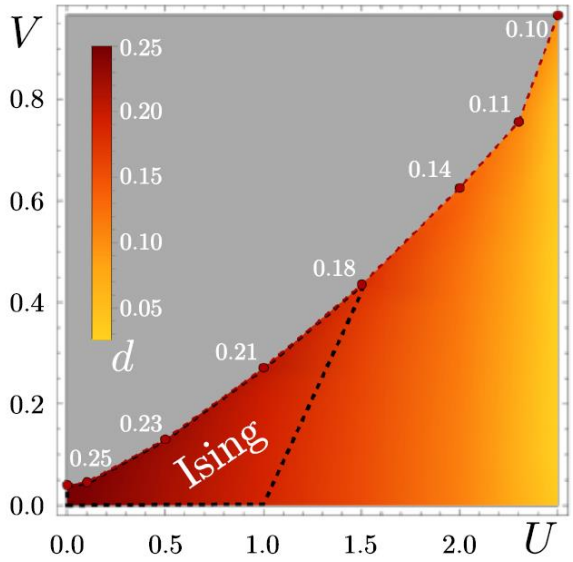


FIG. 1. Double occupancy of the extended Hubbard model shown on the U-V phase diagram. Calculations are performed in the normal phase where the value of the double occupancy d is depicted by color. The gray part corresponds to the charge ordered phase.

Superconductors (e.g. cuprates)

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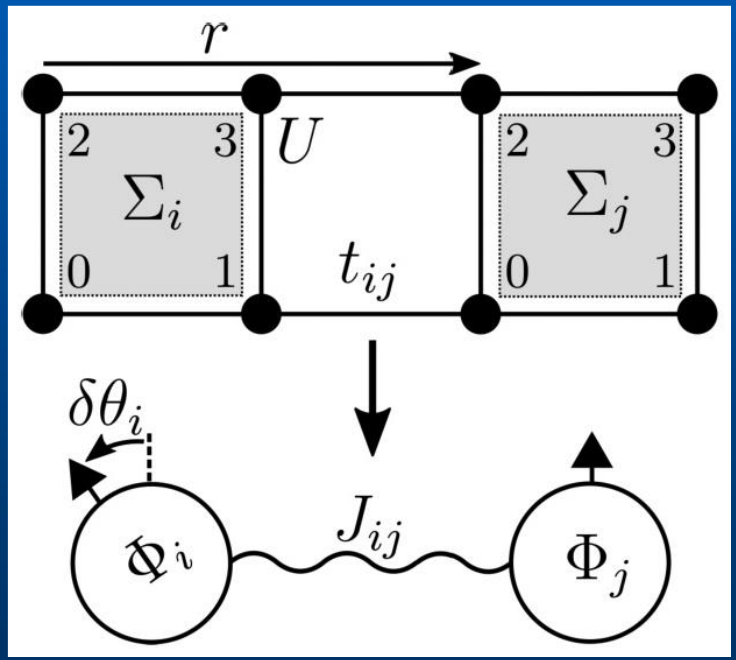
Josephson lattice model for phase fluctuations of local pairs in copper oxide superconductors

Malte Harland,[1] Sergey Brener,[1] Alexander I. Lichtenstein,[1] and Mikhail I. Katsnelson[2]
[1]*Institute of Theoretical Physics, University of Hamburg, Jungiusstraße 9, 20355 Hamburg, Germany*
[2]*Institute for Molecules and Materials, Radboud University, 6525AJ, Nijmegen, The Netherlands*



$$J_{ij} = T\mathrm{Tr}_{\omega\alpha}\left(-G^{p\uparrow}_{ij}S_jG^{h\downarrow}_{ji}S_i + F_{ij}S_jF_{ji}S_i\right)$$

Collaboration

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

and many other people

Thank you for your attention