NORDITA

Spin wave excitations of magnetic metalorganic materials

Johan Hellsvik

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Stockholm, 27 August 2019

Collaborations

The Swedish QuEST for BIFROST and quantum materials

Experiment
Martin Månsson (KTH)
Yasmine Sassa (Chalmers)
Rasmus Toft Pedersen (DTU, ESS)

Theory
Alexander Balatsky (Nordita)
Johan Hellsvik (Nordita)
Olle Eriksson (Uppsala University)

Funding
The Swedish Research Council

Vetenskapsrådet

The organic materials database (OMDB)

R. Matthias Geilhufe (Nordita)
Stanislav Borysov (Nordita, DTU)
Bart Olsthoorn (Nordita)
Roberto Díaz Pérez (Nordita)
Johan Hellsvik (Nordita)
David Carvalho (Nordita)
Alexander Balatsky (Nordita)

https://omdb.mathub.io

SwedNess graduate school on neutron scattering

The Swedish Neutron Week 2019

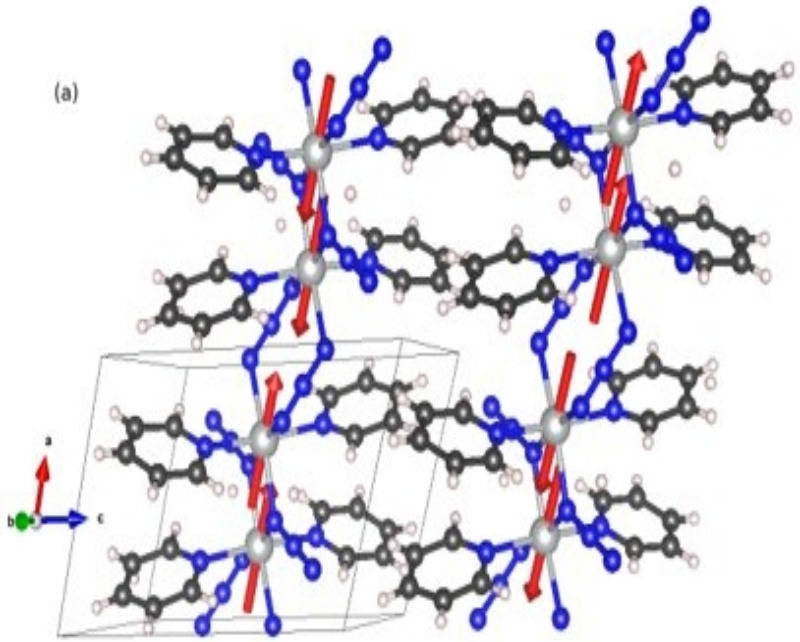
Yasuragi, Stockholm, 6–10 May

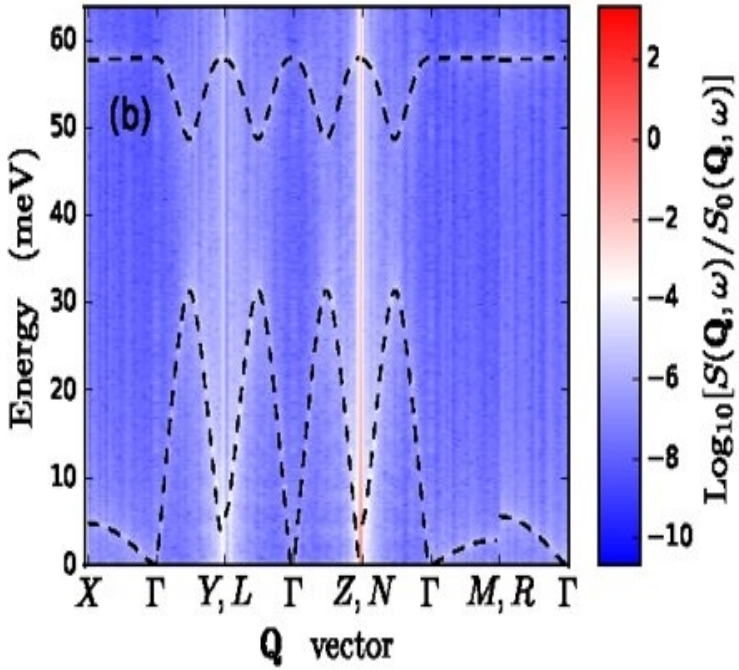
Magnetic excitations: experiment and theory

March 2019, Beamtime at the ISIS neutron and muon source, Rutherford Appleton Laboratory

Nami Matsubara
Ola Kenji Forslund
Martin Månsson
Johan Hellsvik
Elisabetta Nocerino

Magnetic Excitations of organic magnets



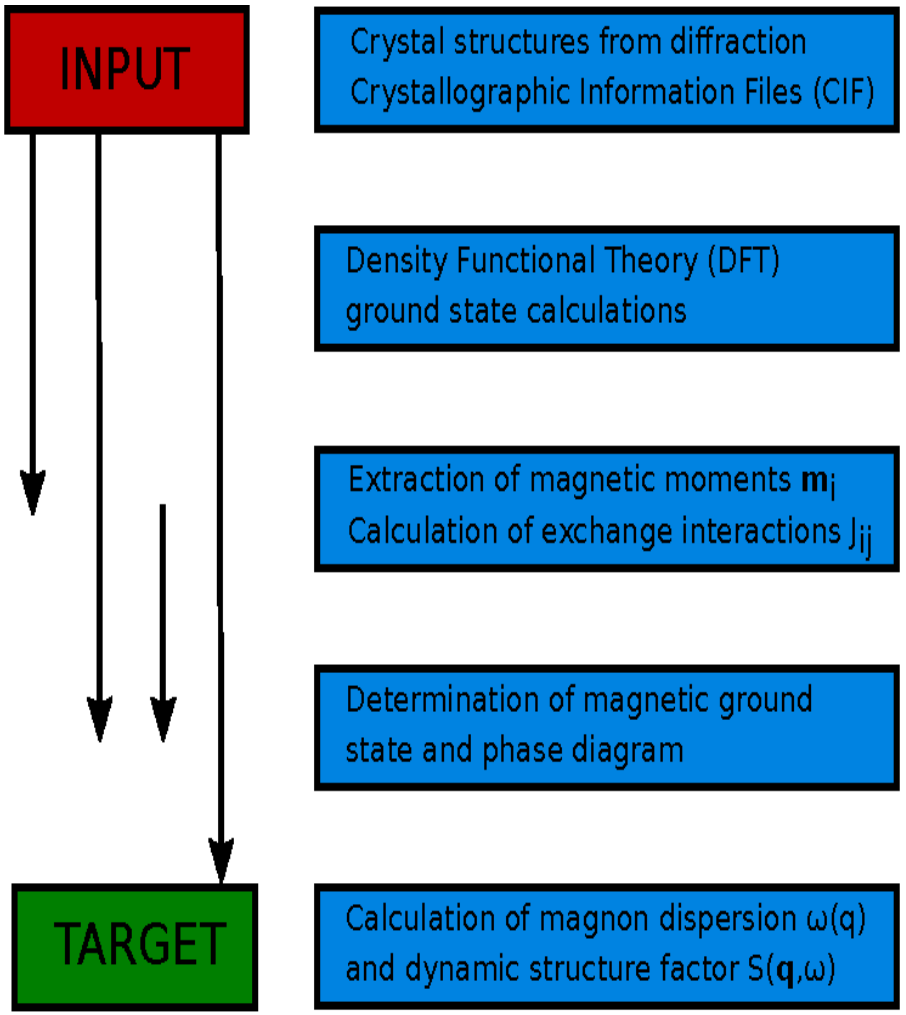


Organic magnetic material with exchange between magnetic metal ions (silver) mediated over organic ligands

Excitation spectra from multiscale ab initio modeling

Hellsvik, Díaz Pérez, Geilhufe, Månsson, and Balatsky, arXiv 1907.01817

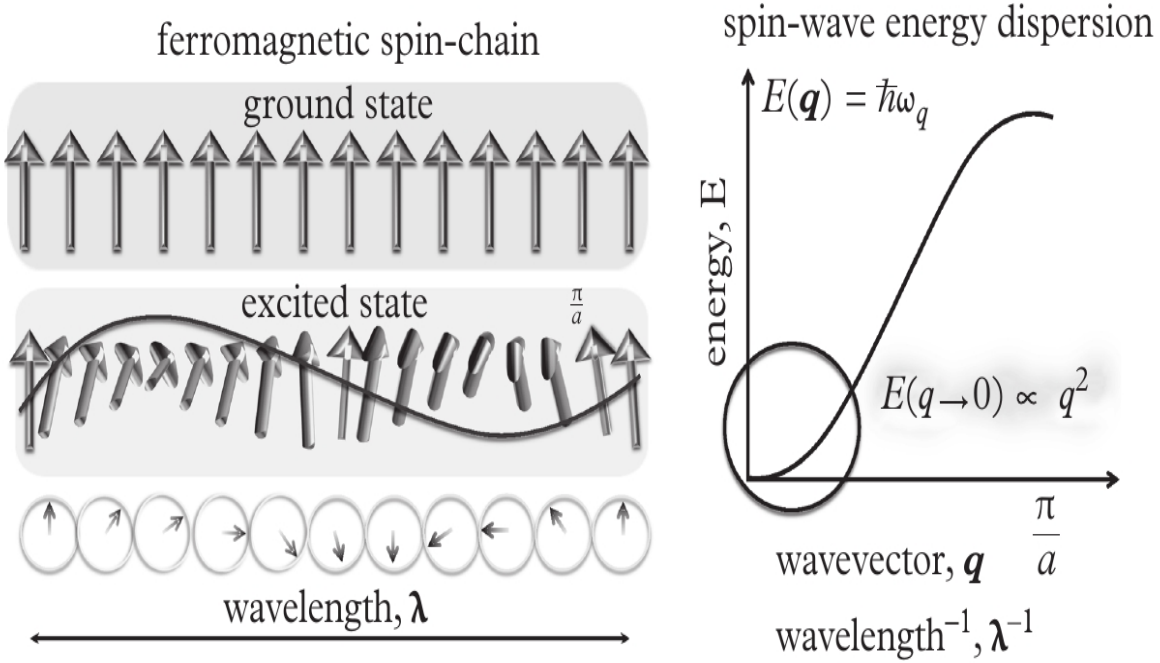
Multiscale ab initio modeling & machine learning



Outline

- Scope: Materials informatics for magnetic excitations on the Organic Materials Database (OMDB)
- Physical entities: Magnetic ground states, interactions, and excitations spectra for crystalline solids
- Ab initio dataset: High throughput calculations for spin Hamiltonians
  Calculation of magnetic ground states and magnon spectra
- Machine learning: Prediction of local magnetic properties: magnetic moments and Heisenberg interactions

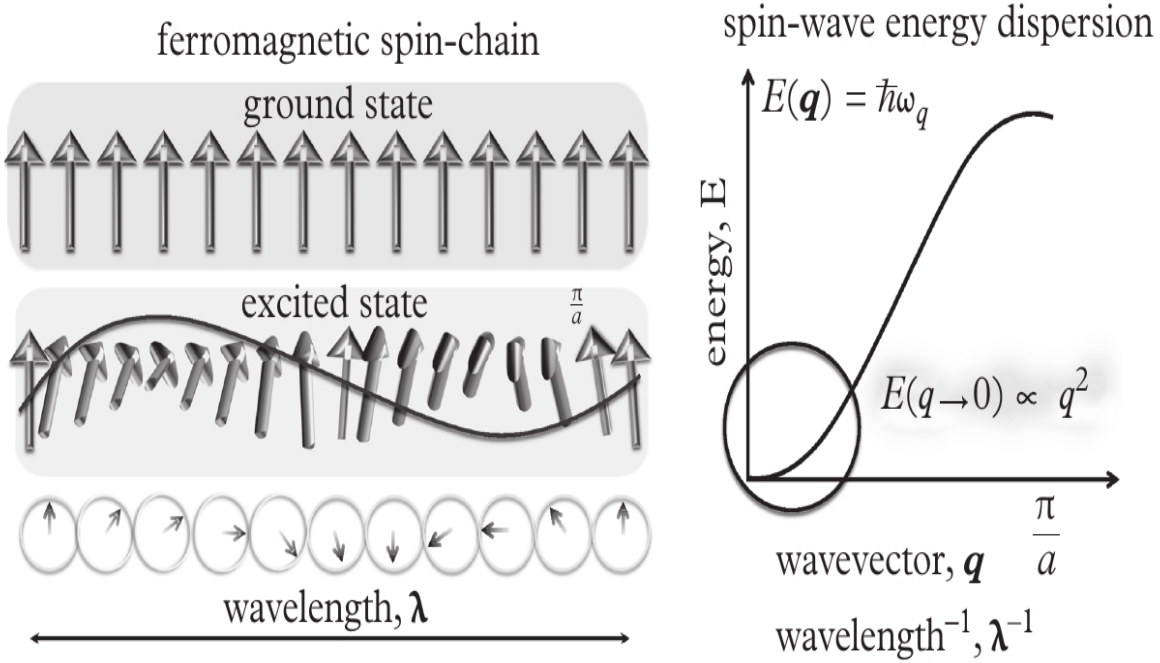
Magnetic Hamiltonians and spin waves



Magnetic Hamiltonians to model low energy magnetic excitations

$$\mathcal{H}_{\text{Heis}} = -\frac{1}{2}\sum_{i \neq j} \mathcal{J}_{ij}\mathbf{m}_i \cdot \mathbf{m}_j,$$

Magnetic Hamiltonians and spin waves



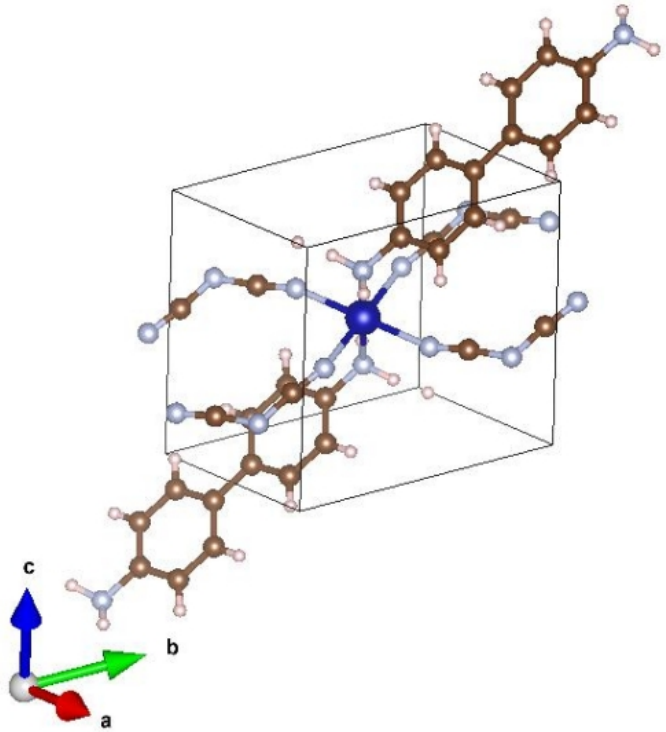
Magnetic Hamiltonians to model low energy magnetic excitations

$$\mathcal{H}_{\text{Heis}} = -\frac{1}{2}\sum_{i\neq j}\mathcal{J}_{ij}\mathbf{m}_i \cdot \mathbf{m}_j,$$

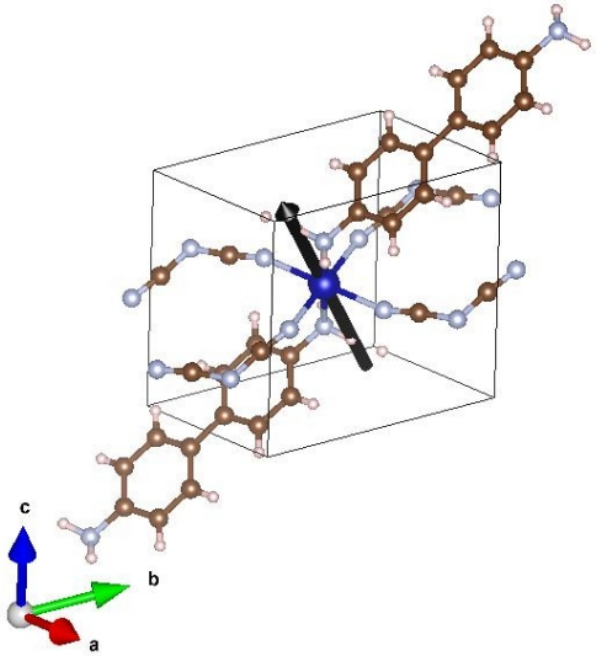
Dispersion relations ω(**q**) with linear spin wave theory

Dynamic structure factor S(**q**,ω) with atomistic spin dynamics (ASD) simulations

Magnetic moments $\mathbf{m}_i$ and Heisenberg interactions J_{ij}

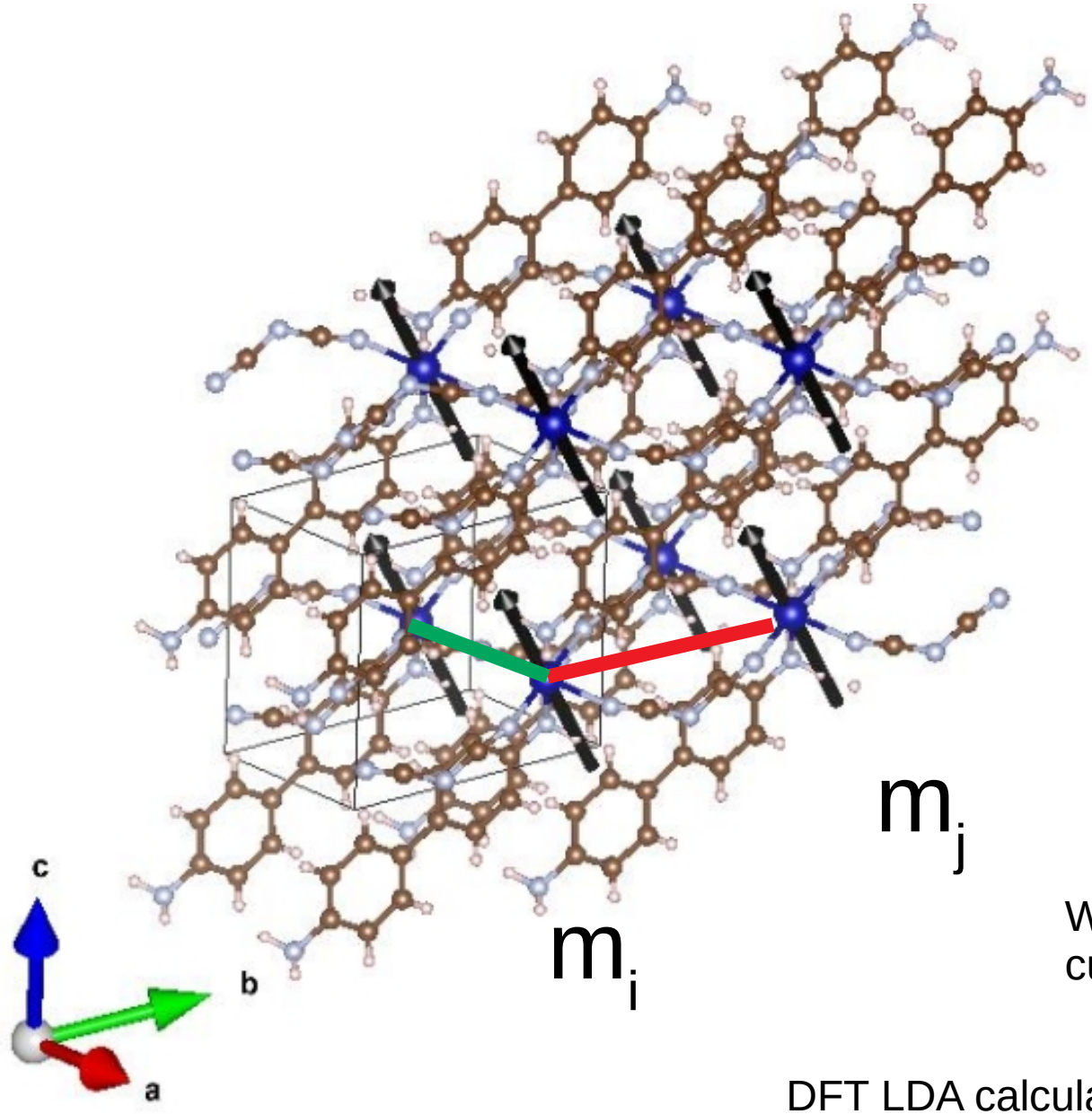


Chemical unit cell



magnetic sites m_i (>0.1 muB)

Magnetic moments $\mathbf{m}_i$ and Heisenberg interactions J_{ij}



$$\mathcal{H}_{\mathrm{Heis}} = -\frac{1}{2}\sum_{i\neq j}\mathcal{J}_{ij}\mathbf{m}_i \cdot \mathbf{m}_j,$$

Nearest neighbour interaction J_{NN}

Next nearest neighbour interaction J_{NNN}

We consider interactions up to a cut-off radius r_c

DFT LDA calculations using the LKAG formula
Liechtenstein *et al.*, J. Mag. Mat **67**, 65 (1987)

$C_{20}H_{10}CoN_{12}$

Formula:
C_{20} H_{10} Co N_{12}
OMDB ID:
11913
COD ID:
2014058

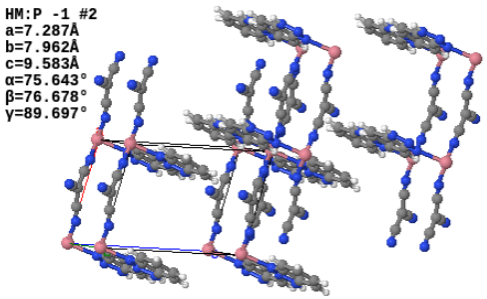
Publication details:
catena-Poly[[bis(1*H*-benzotriazole-\k*N*^3^)cobalt(II)]-di-\m-tricyanomethanido-\k^2^*N*:*N*'] and *catena*-poly[[bis(3,5-dimethyl-1*H*-pyrazole-\k*N*^2^)manganese(II)]-di-\m-tricyanomethanido-\k^2^*N*:*N*']

Publisher:
Acta Crystallographica Section C, 2004, vol: 60, page: m250

Version History:
No changes

Crystallographic Information

COD Data



Explore

load unitcell | load 8 unitcells

Interact

rotate to best view | default zoom

hide symmetry

spin on

a	7.287(2)
b	7.962(3)
c	9.583(3)
α	75.643(4)
β	76.678(4)
γ	89.697(4)

Download cif file | COD link

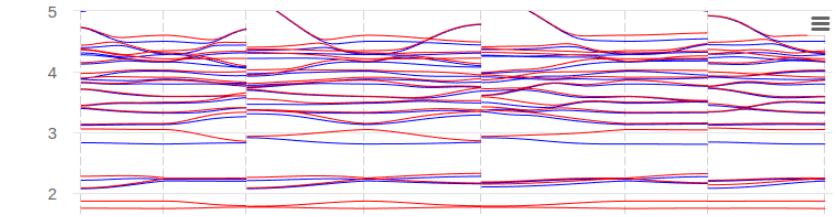
Symmetry properties

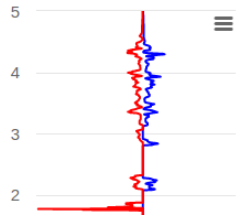
Hermann-Mauguin symmetry space group	P -1
Hall symmetry space group	-P 1
Space group IT number	2

Band structure and density of states

PBE

Select a range with the left mouse button to zoom in





Indirect band gap DFT GGA* (eV)	0.0095
Magnetization density [μ_B/Å^3]	0.0057

* Warning! DFT is not very accurate in estimation of band gaps and tends to underestimate them.

Special points in the Brillouin zone

Formula:
C_{20} H_{10} Co N_{12}
OMDB ID:
11913
COD ID:
2014058

Publication details:
catena-Poly[[bis(1*H*-benzotriazole-\k*N*^3^)cobalt(II)]-di-\m-tricyanomethanido-\k^2^*N*:*N*'] and *catena*-poly[[bis(3,5-dimethyl-1*H*-pyrazole-\k*N*^2^)manganese(II)]-di-\m-tricyanomethanido-\k^2^*N*:*N*']

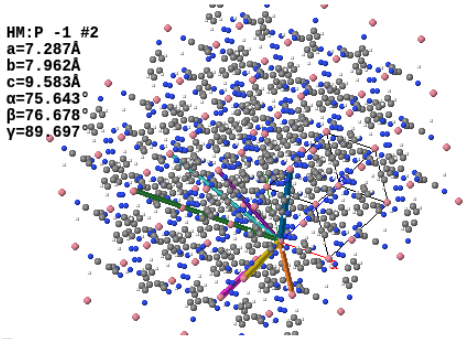
Publisher:
Acta Crystallographica Section C, 2004, vol: 60, page: m250

Version History:
No changes

J_{ij}
Ordered by magnitude

i	j	r [Å]	J_{ij} [meV]	Color
1	1	7.29	-0.28	■
1	1	14.57	0.13	■
1	1	21.86	-0.04	■
1	1	10.82	0.01	■
1	1	10.62	0.01	■
1	1	7.96	-0.01	■
1	1	10.76	0.00	■
1	1	16.57	0.00	■
1	1	9.58	0.00	■

Visualize



☑ Show J_{ij} interactions

Magnetic Moments

i	Species	Magnetic Moment [μ_B]
1	Co	0.6029263

Input files for UppASD calculations

inpsd.dat | jfile_cart | momfile_cart | posfile_cart | qfile_cart

Community Contributions

Add your contribution

There isn't any community contribution for this material.

Services

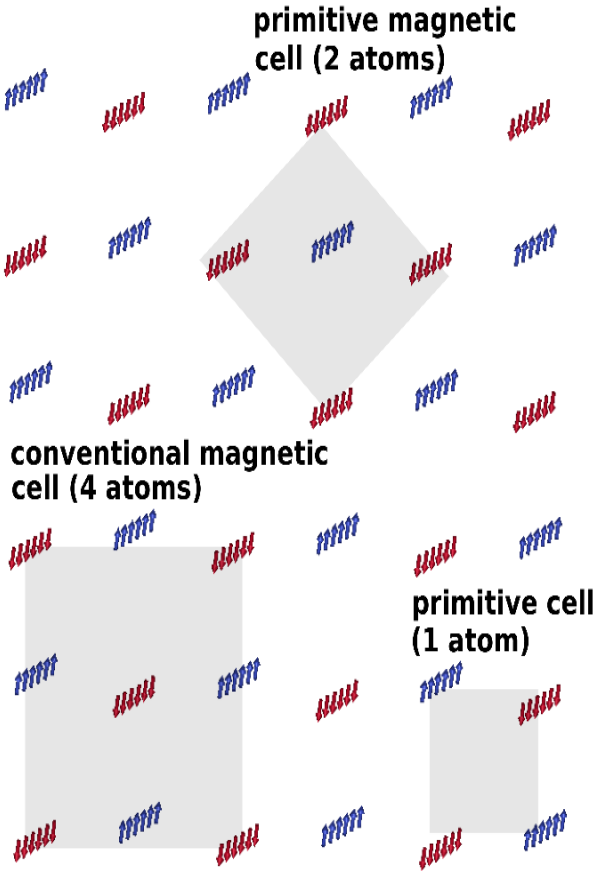
Propose or request a new service

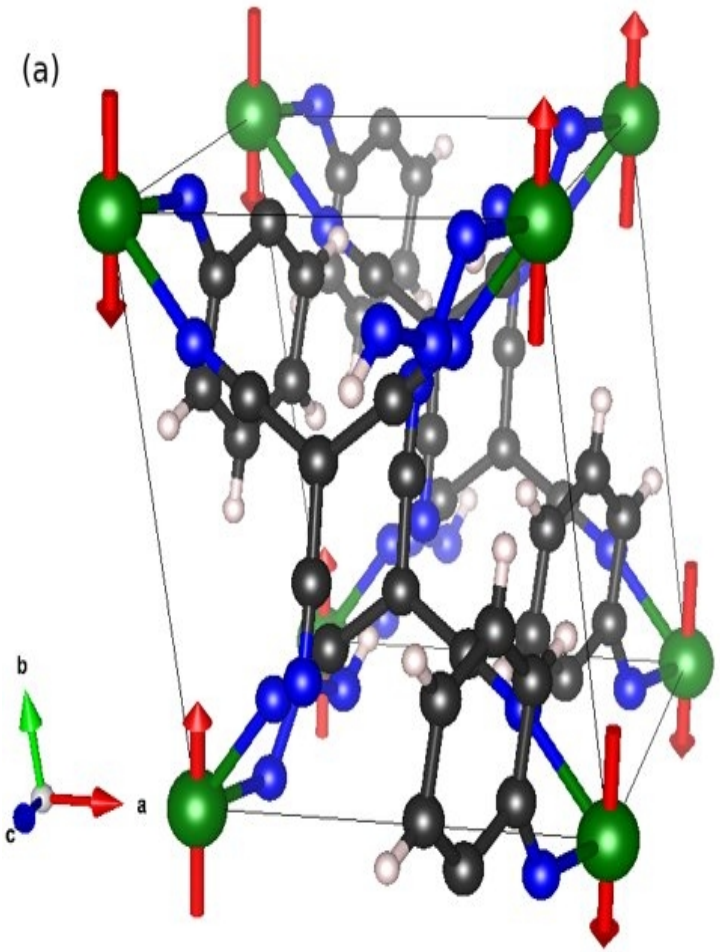
There isn't any service proposal or request for this material.

Similar Materials

Most similar materials with respect to DOS, using cosine distance:

Ground state and primitive magnetic cell





Ground state obtained from atomistic spin dynamics quenching simulation down to T=0 K

Formula:
C_{20} H_{10} Co N_{12}
OMDB ID:
11913
COD ID:
2014058

Publication details:
catena-Poly[[bis(1*H*-benzotriazole-\k*N*^3^)cobalt(II)]-di-\m-tricyanomethanido-\k^2^*N*:*N*'] and *catena*-poly[[bis(3,5-dimethyl-1*H*-pyrazole-\k*N*^2^)manganese(II)]-di-\m-tricyanomethanido-\k^2^*N*:*N*']

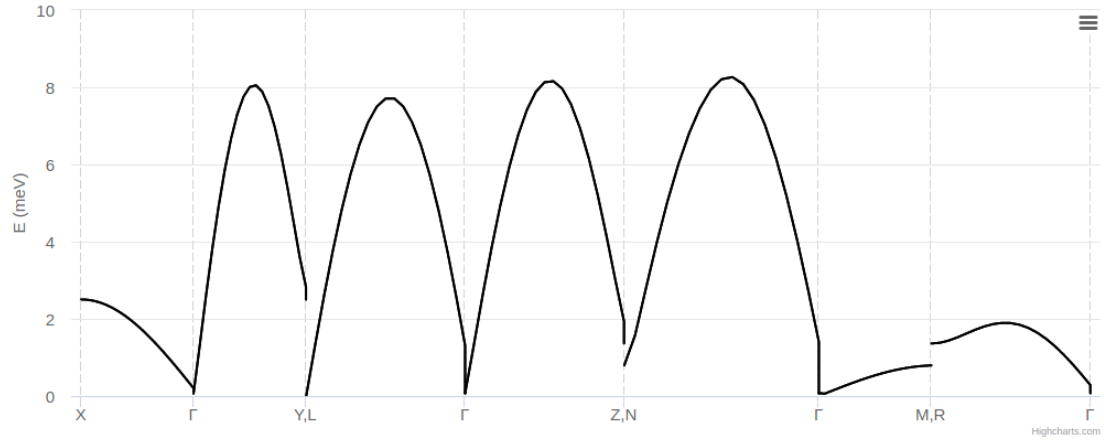
Publisher:
Acta Crystallographica Section C, 2004, vol: 60, page: m250

Version History:
No changes

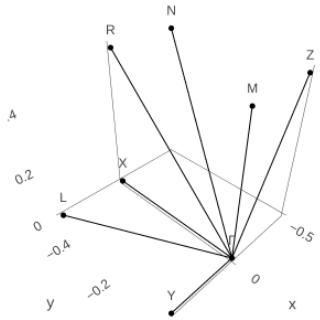
Density of states INCAR.gz POSCAR.gz KPOINTS.gz

Magnetic properties

Spin Wave Dispersion

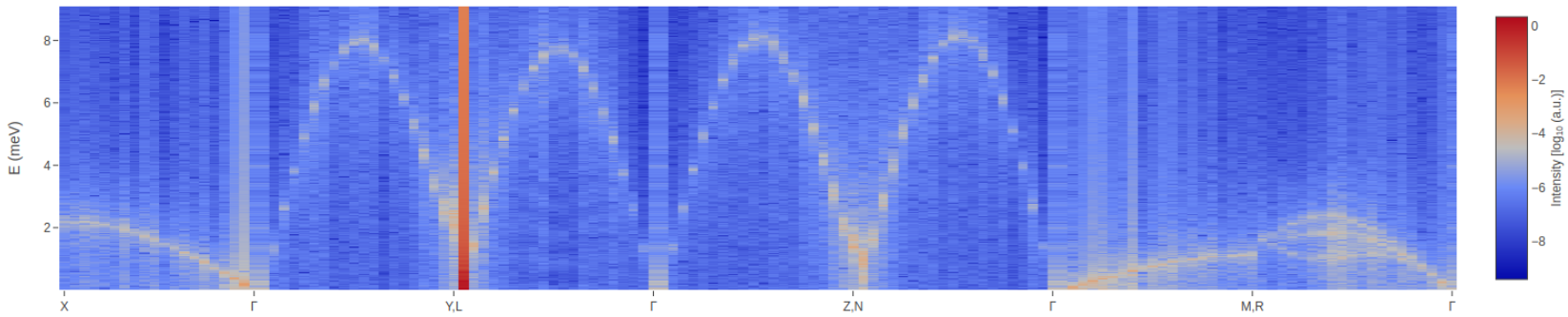


Special points in the Brillouin zone



LDA calculations with RSPt.

Dynamical structure factor S(Q,ω)



$C_6H_7Cl_3Ti$

Formula:
$C_6 H_7 Cl_3 Ti$
OMDB ID:
11695
COD ID:
2000232

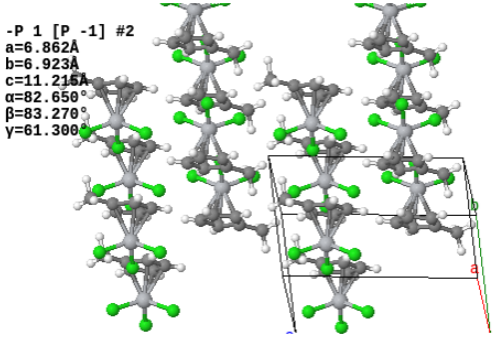
Publication details:
Structure of trichloro(\h^5^-methylcyclopentadienyl)titanium(IV)

Publisher:
Acta Crystallographica Section C, 1991, vol: 47, page: 2216

Version History:
No changes

Crystallographic Information

COD Data



Explore

load unitcell | load 8 unitcells

Interact

rotate to best view | default zoom

hide symmetry

spin on

a	6.8620(10)
b	6.923(2)
c	11.215(2)
α	82.650(10)
β	83.270(10)
γ	61.300(10)

Download cif file | COD link

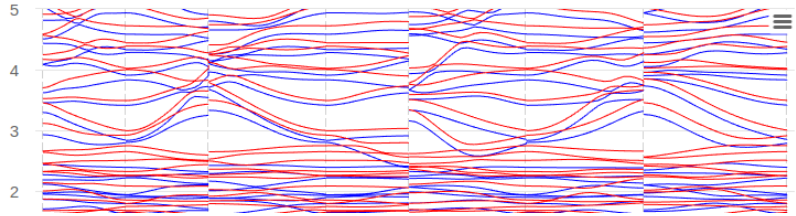
Symmetry properties

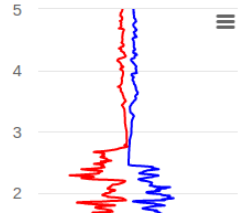
Hermann-Mauguin symmetry space group	P -1
Hall symmetry space group	-P 1
Space group IT number	

Band structure and density of states

PBE

Select a range with the left mouse button to zoom in





Indirect band gap DFT GGA* (eV)	0
Magnetization density [μ_B/Å^3]	0.0086

* Warning! DFT is not very accurate in estimation of band gaps and tends to underestimate them.

Special points in the Brillouin zone

Formula:
$C_6 H_7 Cl_3 Ti$

OMDB ID:
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COD ID:
2000232

Publication details:
Structure of trichloro(\h^5^-methylcyclopentadienyl)titanium(IV)

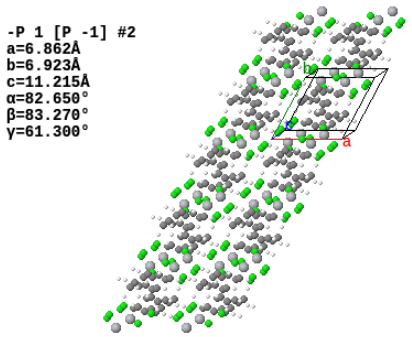
Publisher:
Acta Crystallographica Section C, 1991, vol: 47, page: 2216

Version History:
No changes

J_{ij}
Ordered by magnitude

i	j	r [Å]	J_{ij} [meV]	Color
1	2	8.04	0.26	
2	2	6.92	0.08	
1	1	6.92	0.08	
1	2	7.92	-0.02	
2	2	7.03	-0.01	
1	1	7.03	-0.01	
1	2	5.57	0.01	
1	2	13.93	-0.01	
1	2	6.12	-0.01	
1	2	8.39	-0.01	

Visualize



Show J_{ij} interactions

Magnetic Moments

i	Species	Magnetic Moment [μ_B]
1	Ti	0.2242664
2	Ti	0.2242564

Input files for UppASD calculations

inpsd.dat | jfile_cart | momfile_cart | posfile_cart | qfile_cart

Community Contributions

Add your contribution

There isn't any community contribution for this material.

Services

Propose or request a new service

There isn't any service proposal or request for this material.

Similar Materials

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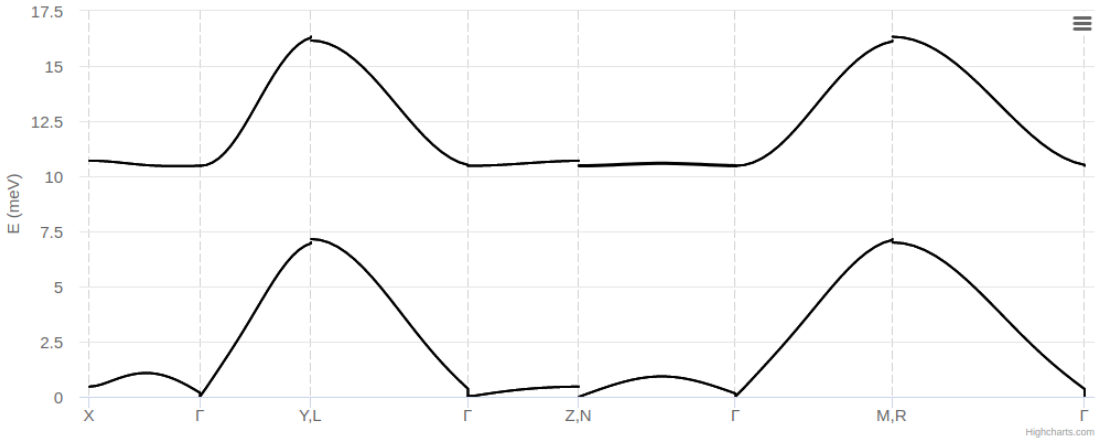
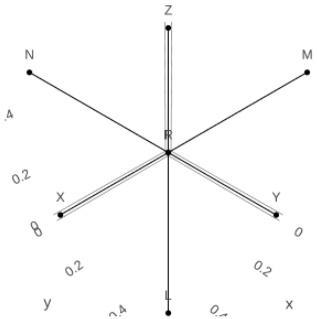
Magnetic properties

Spin Wave Dispersion

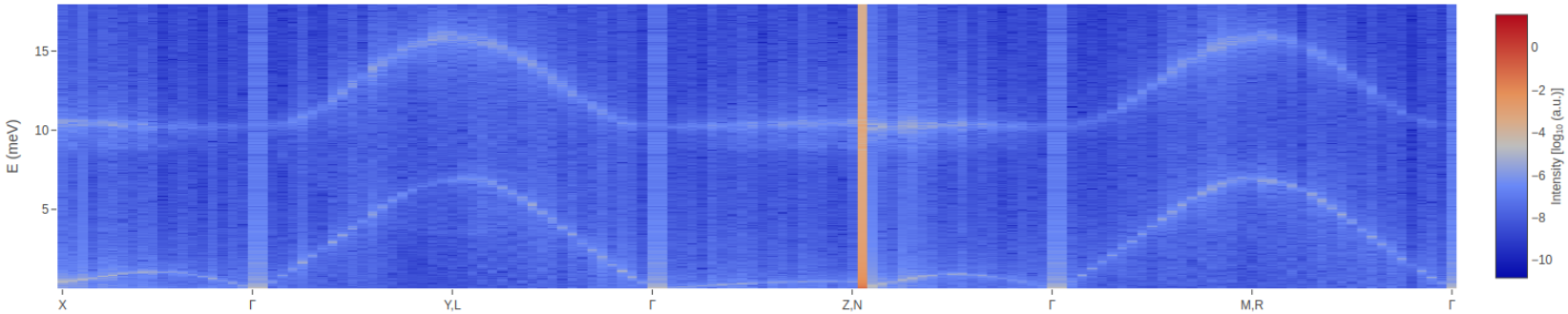


Special points in the Brillouin zone

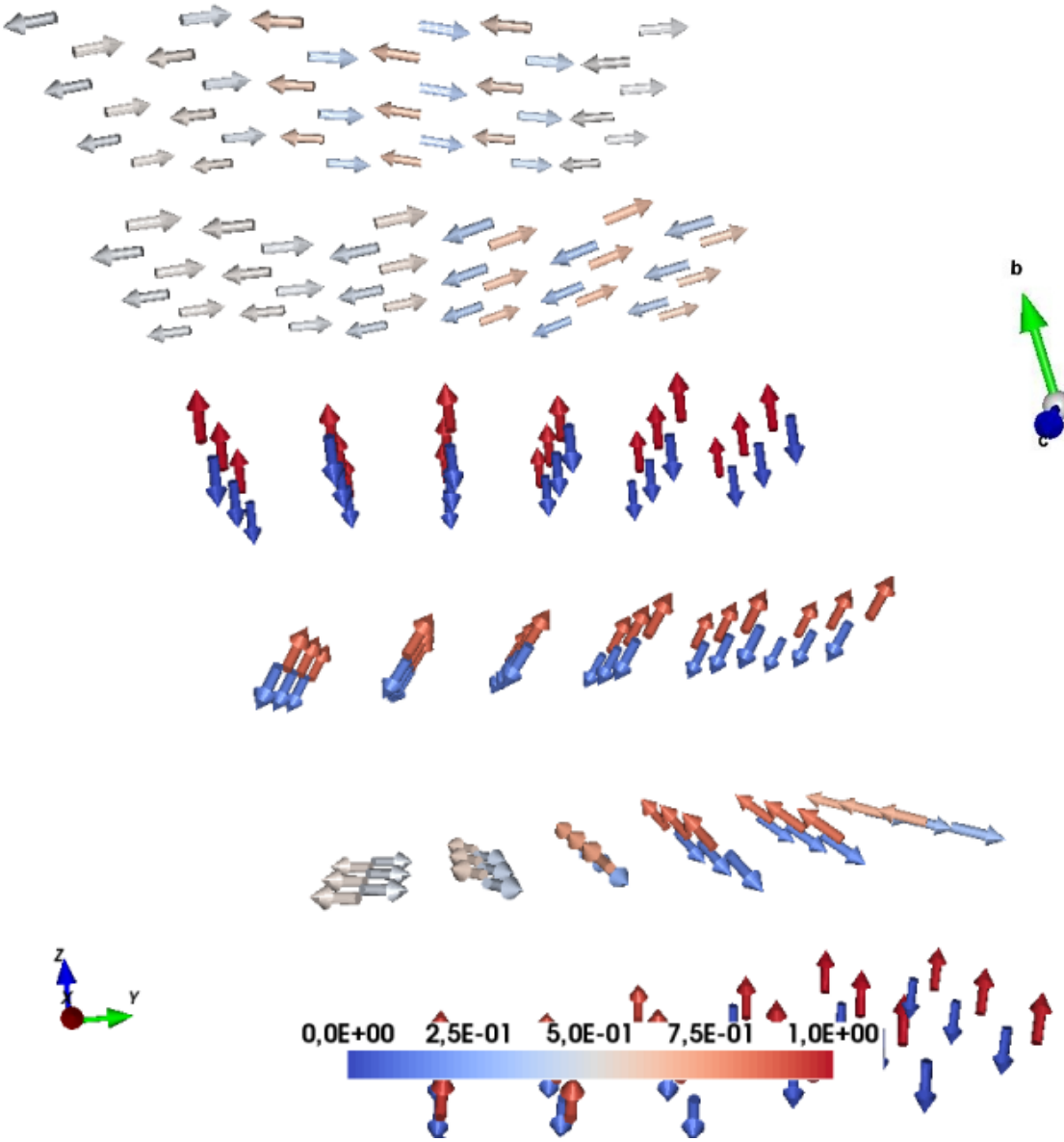


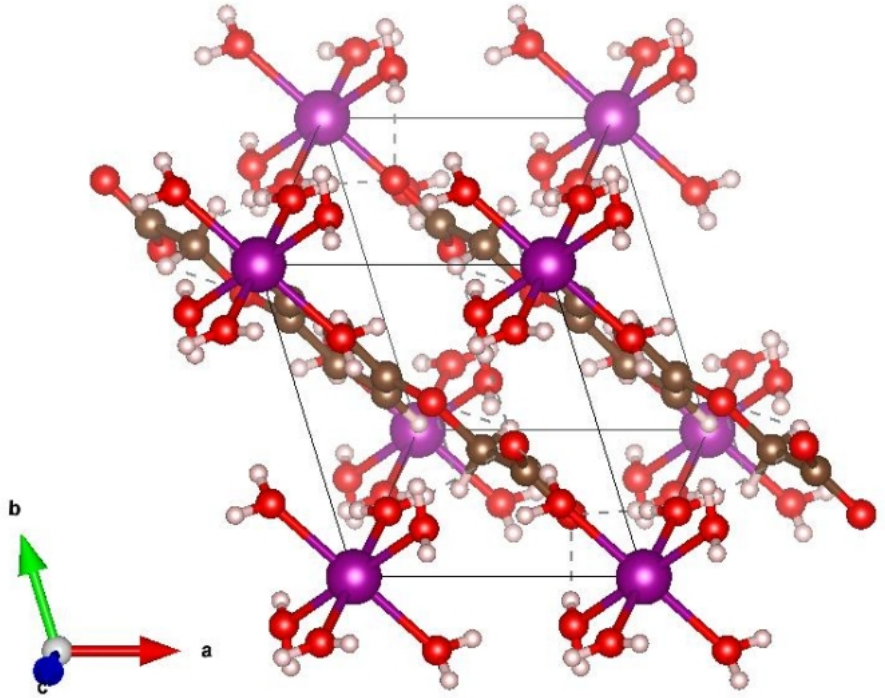
LDA calculations with RSPt.

Dynamical structure factor S(Q,ω)



Material COD 2203562

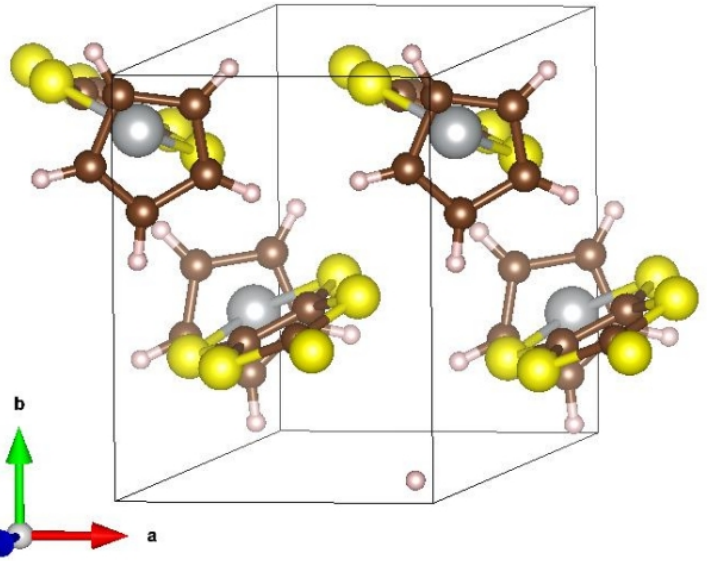


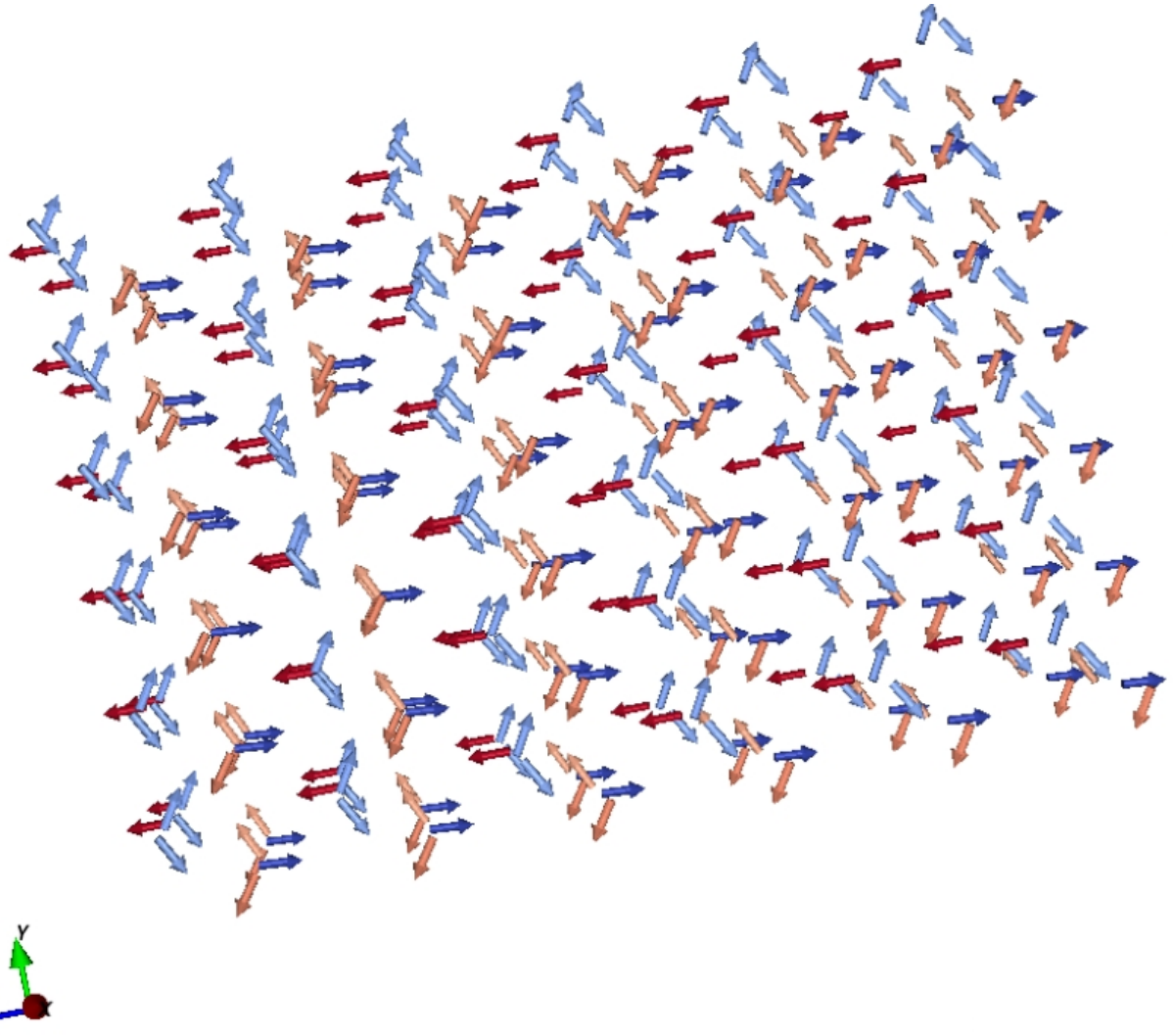


AFM spin spiral ordering

Modulated with incommensurate wave vector

Material COD 7008182

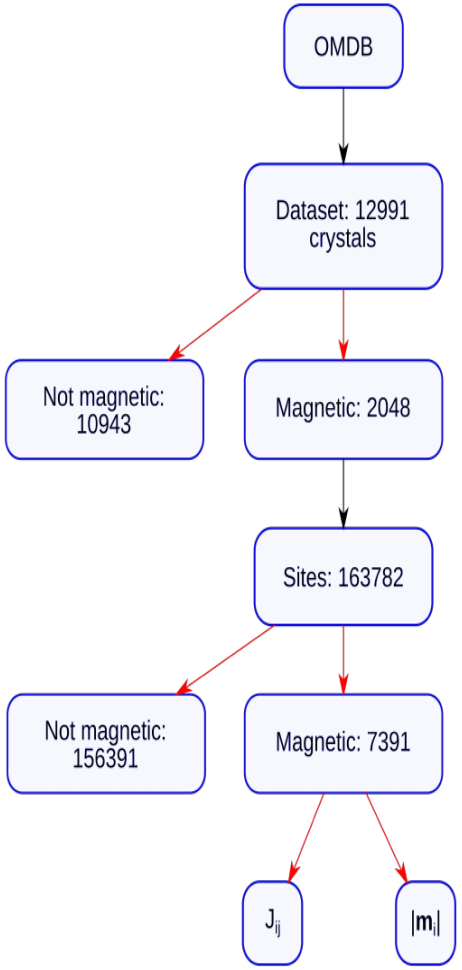




120 degree AFM configuration

Commensurate magnetic ordering?

Machine learning of magnetic properties



Challenge: Predict local properties of complex crystal structures

Target: Magnetic moments and Heisenberg interactions

Strategy:

1. Classification of crystals as magnetic or non-magnetic
2. Classification of atoms as magnetic or non-magnetic
3. Regression for $|\mathbf{m}_i|$ and J_{ij}

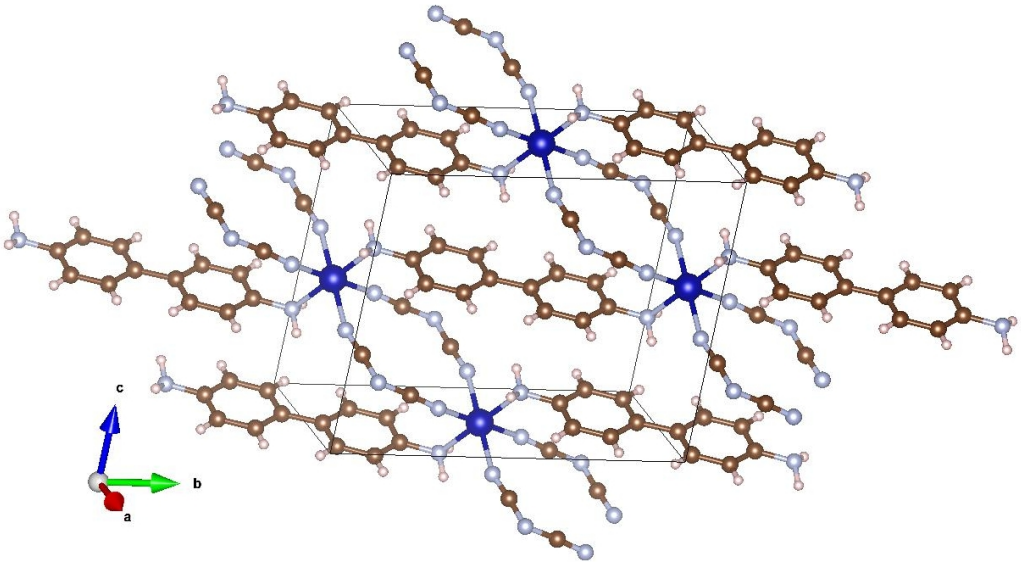
Roberto Díaz Pérez, MSc thesis, Stockholm University and Nordita (2019)
R. Díaz Pérez, R. M. Geilhufe, J. Hellsvik, and A. V. Balatsky, *in preparation* (2019).

Representations of crystals

Crystals: Defined by
Lattice vectors ($\mathbf{a}_1$, $\mathbf{a}_2$, $\mathbf{a}_3$), atomic sites ($\{\mathbf{r}_i\}$), atomic numbers, ($\{Z_i\}$).

The description $\mathbf{a}_1$, $\mathbf{a}_2$, $\mathbf{a}_3$, $\{\mathbf{r}_i, Z_i\}$ is not unique

For a low symmetric compound, consider e.g. a permutation of the lattice vectors



A good descriptor for a crystal should be:

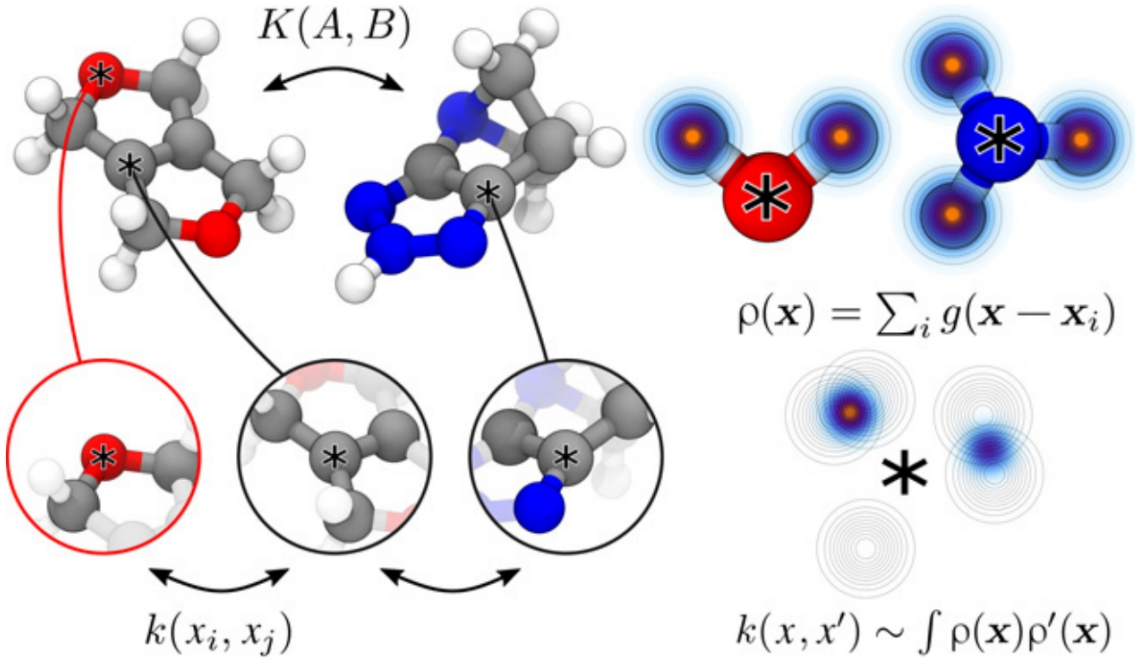
1. Translationally invariant

2. Invariant with respect to the size of the cell

Multihot encoding – presence/absence of elements

Formula	C	H	Fe	Br	...	$\boldsymbol{x}$
$C_{28}H_{18}Fe$	1	1	1	0	...	[1 1 1 0 ...]
$C_{18}H_{13}Br$	1	1	0	1	...	[1 1 0 1 ...]

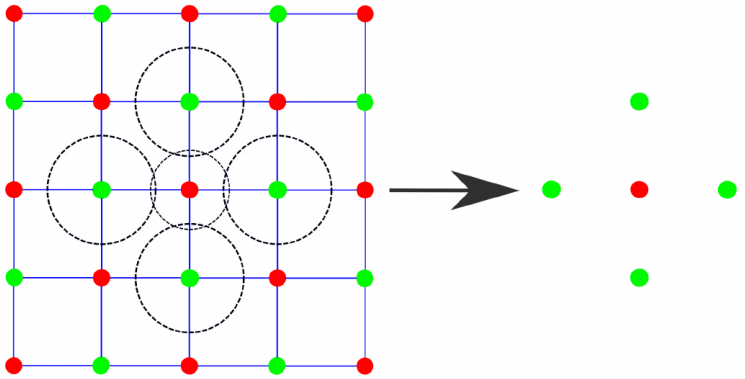
Smooth overlap of atomic positions (SOAP)



A. P. Bartók, S. De, C. Poelking, N. Bernstein, J. R. Kermode, G. Csányi, and M. Ceriotti, Sci. Adv. 3(12):e1701816 (2017)

Local environment representations

$$V_i^{\text{One-Hot}} = \delta_{ik},$$



Central site	C	H	O	$V_{\text{One-Hot}}$
C	1	0	0	[1 0 0]
H	0	1	0	[0 1 0]
O	0	0	1	[0 0 1]

$$V_i^{\text{distance}} = \sum_l \frac{1}{|\boldsymbol{r}_l - \boldsymbol{r}_{\text{central}}|} \delta_{is_l}$$

Radial Multihot: Concatenate to a vector $\boldsymbol{V} = [\boldsymbol{V}_{\text{One-Hot}} \boldsymbol{V}_{\text{distance}}]$

Examples: Carbon dioxide CO_2 and methane CH_4

For central atom carbon, both have $\boldsymbol{V}_{\text{C}}^{\text{One-Hot}} = [1,\ 0,\ 0]$

$$\boldsymbol{V}_{\text{CH}_4,\text{C}}^{\text{distance}} = [0,\ 3.67,\ 0]$$

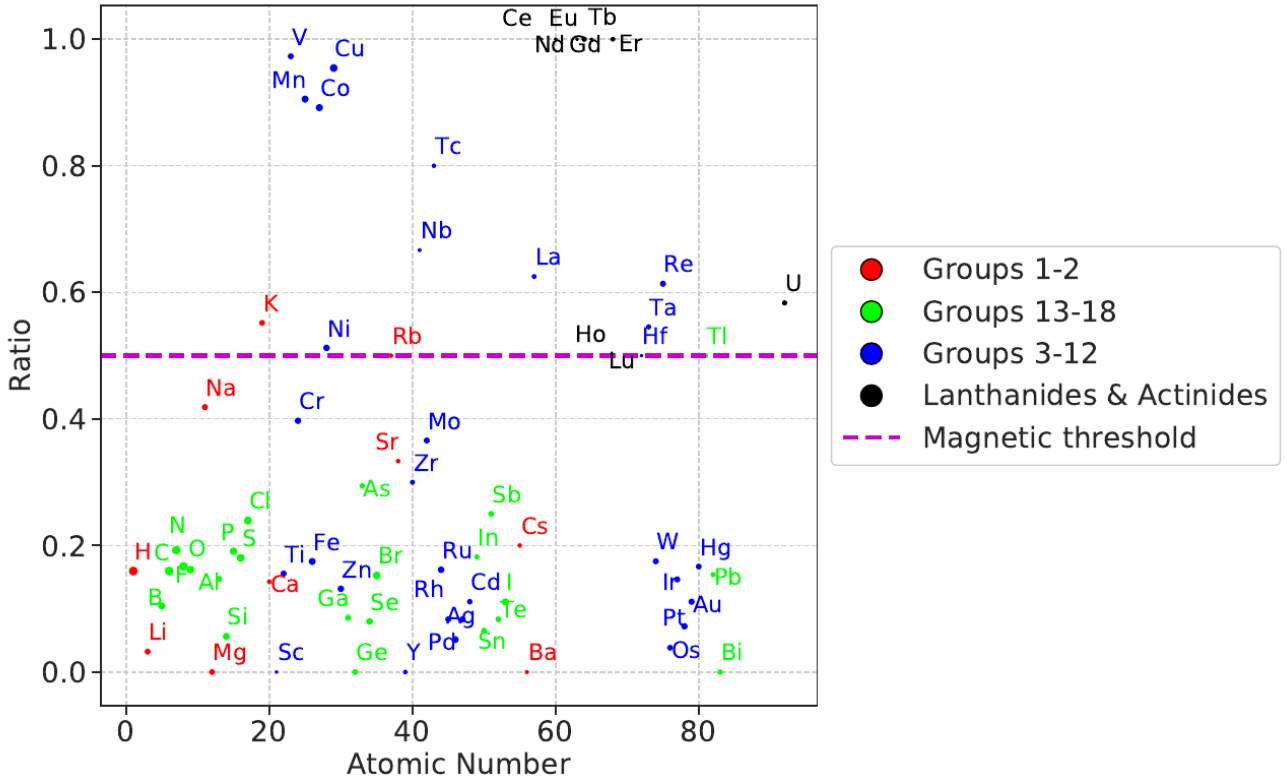
$$\boldsymbol{V}_{\text{CO}_2,\text{C}}^{\text{distance}} = [0,\ 0,\ 1.72]$$

$$\boldsymbol{V}_{\text{CH}_4,\text{C}} = [1,\ 0,\ 0,\ 0,\ 3.67,\ 0]$$

$$\boldsymbol{V}_{\text{CO}_2,\text{C}} = [1,\ 0,\ 0,\ 0,\ 0,\ 1.72]$$

Classification of magnetic crystals – baseline model

Ratios of magnetic materials for training set



$$\text{Accuracy} := \frac{\text{Correct predictions}}{\text{Total predictions}}$$

$$\text{F1} := \frac{2 \times \text{recall} \times \text{precision}}{\text{recall} + \text{precision}}$$

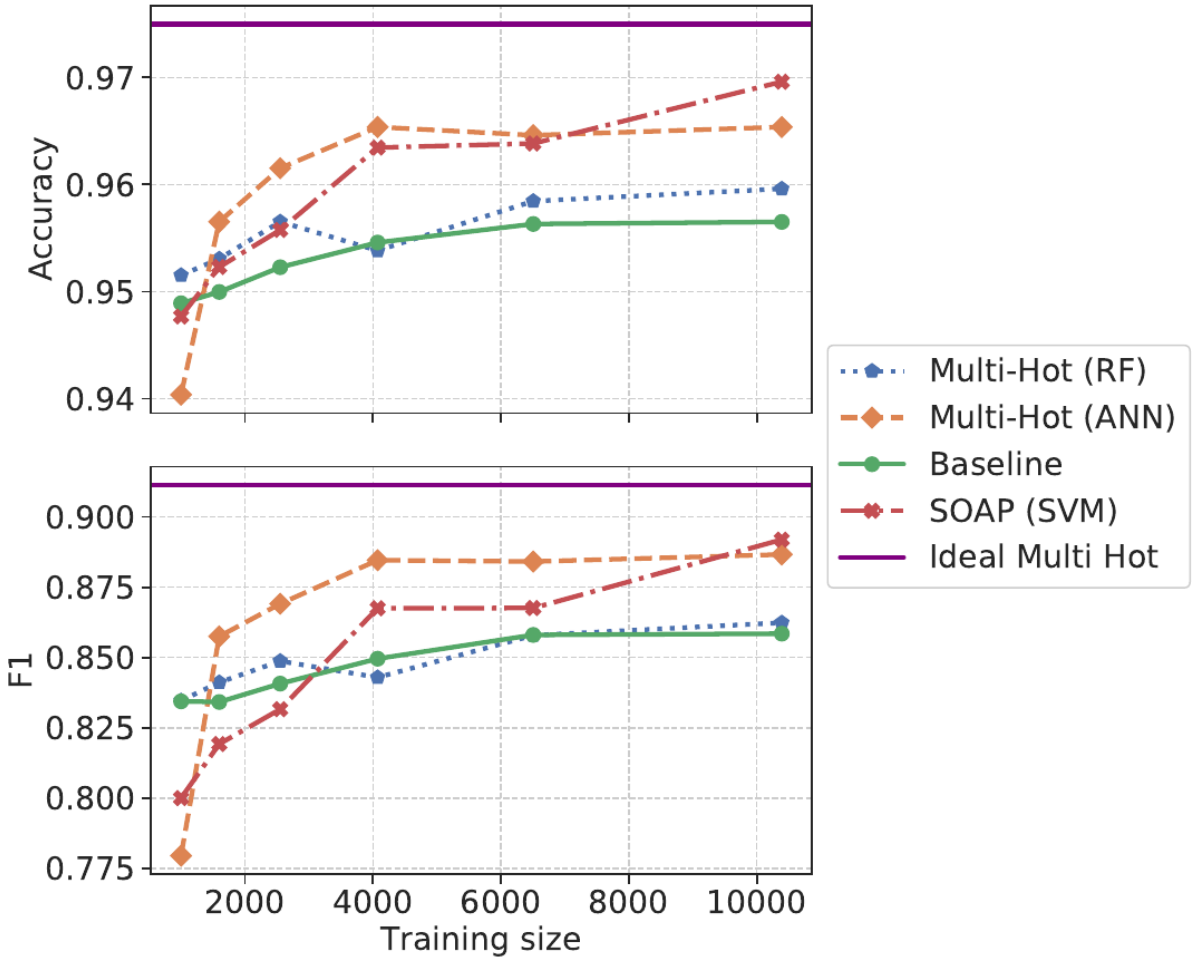
Metrics for test set

$$\text{Accuracy} = 0.951;$$

$$\text{F1} = 0.831;$$

	Predicted: Positive	**Predicted: Negative**
Actual: Positive	313	77
Actual: Negative	50	2159

Classification of magnetic crystals – machine learning



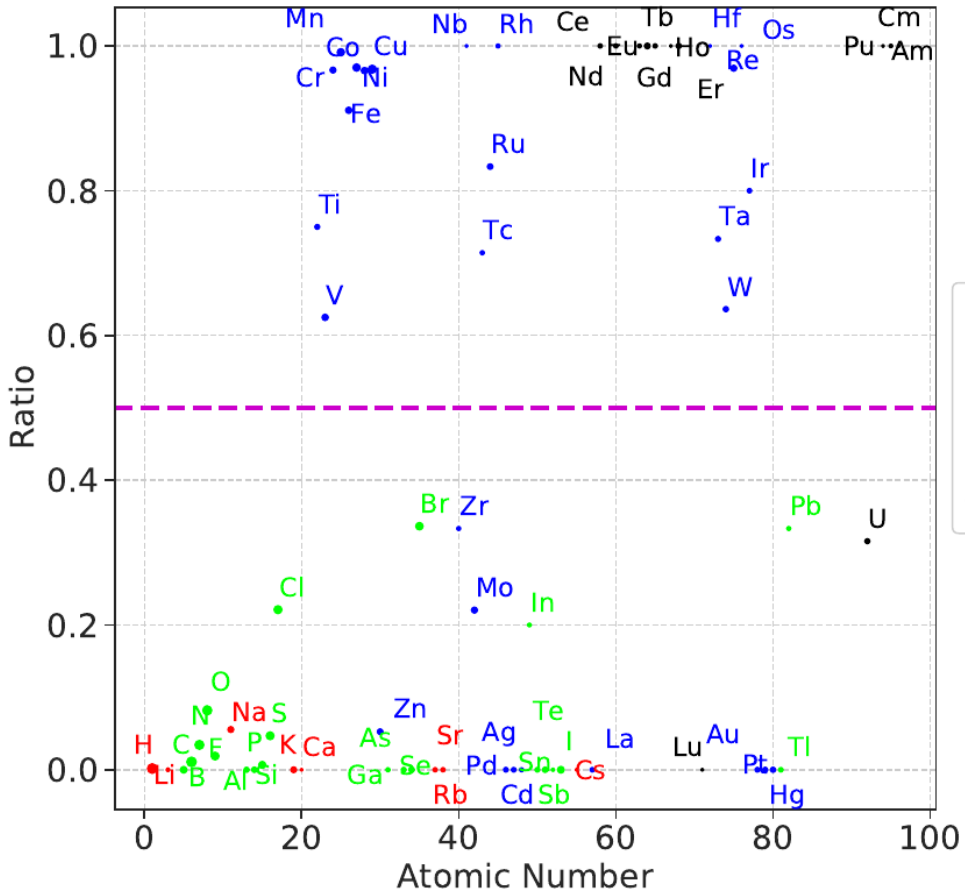
For full training set, SOAP achieves

$$\text{Accuracy} = 0.97;$$

$$\text{F1} = 0.89;$$

Classification of magnetic sites – baseline model

Ratios of magnetic sites for training set



Group in periodic table is a strong indicator

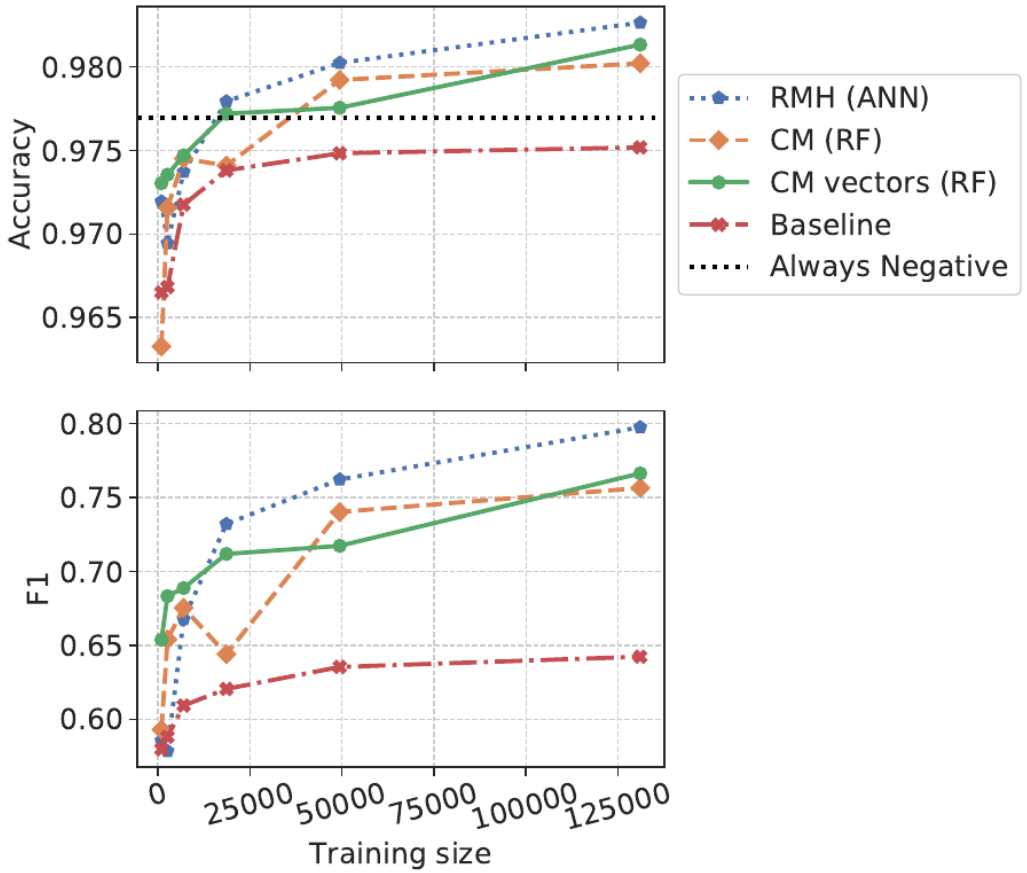
Metrics for test set

$$\text{Accuracy} = 0.975$$

$$\text{F1} = 0.642;$$

	Predicted: Positive	**Predicted: Negative**
Actual: Positive	695	751
Actual: Negative	23	29722

Classification of magnetic sites – machine learning

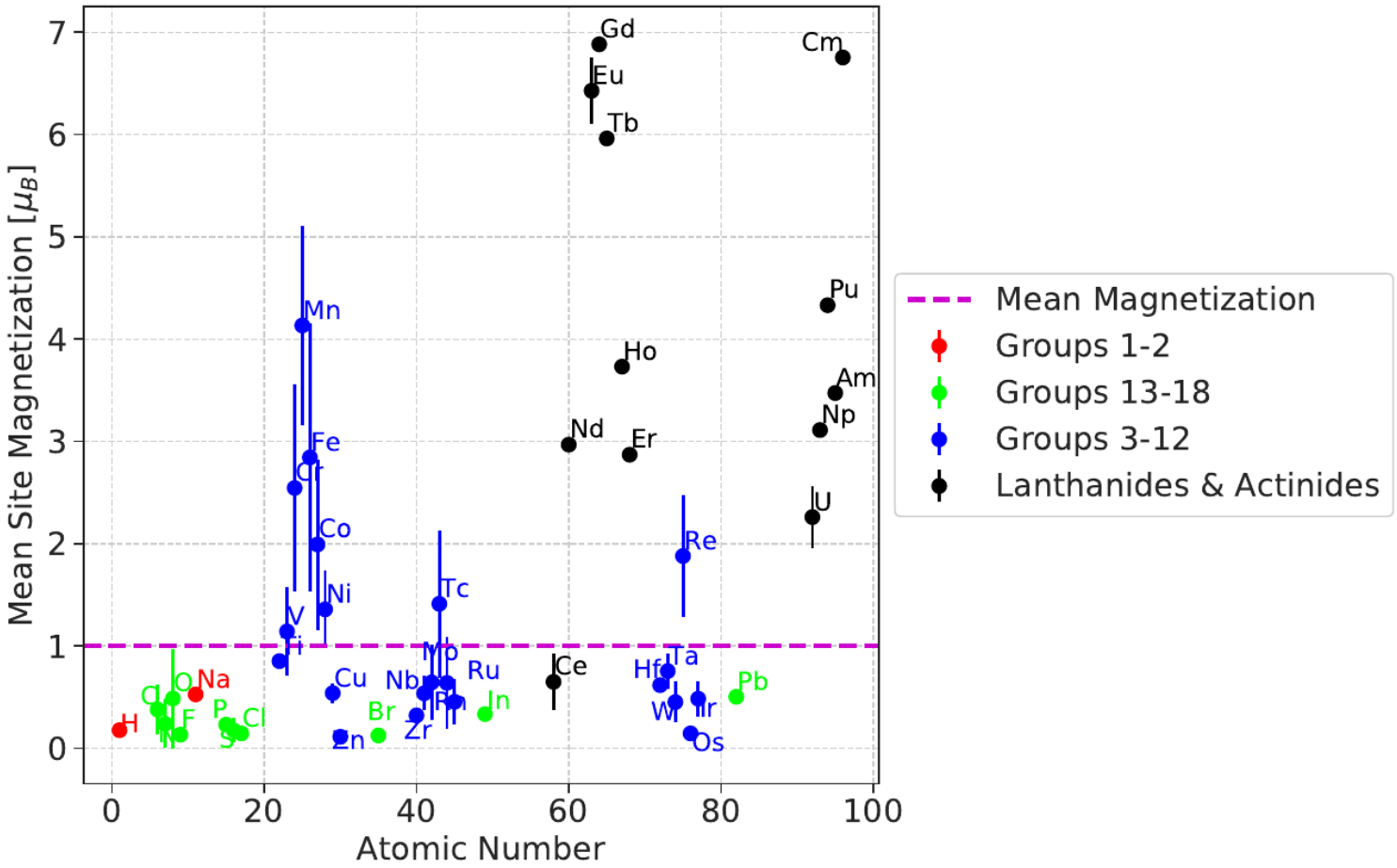


Best performance for Radial Multi-Hot

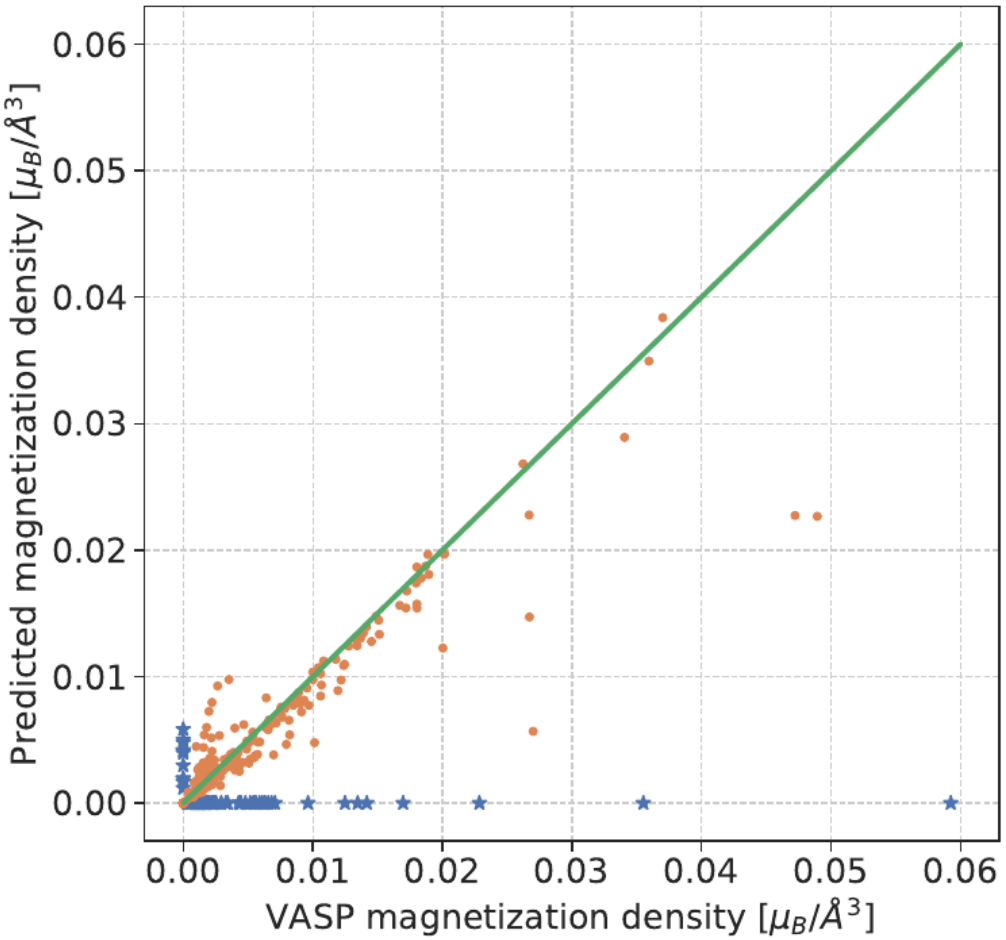
Accuracy=0.99

F1=0.89

Regression for site magnetic moments – statistics



Regression for magnetization density – machine learning



Performance for Radial Multi-Hot

Metric:

$$r^2 = 1 - \frac{\mathrm{MSE}}{\mathrm{VAR}}$$

where

MSE = Mean Square Error

VAR = Variance

For all data: r^2=0.67

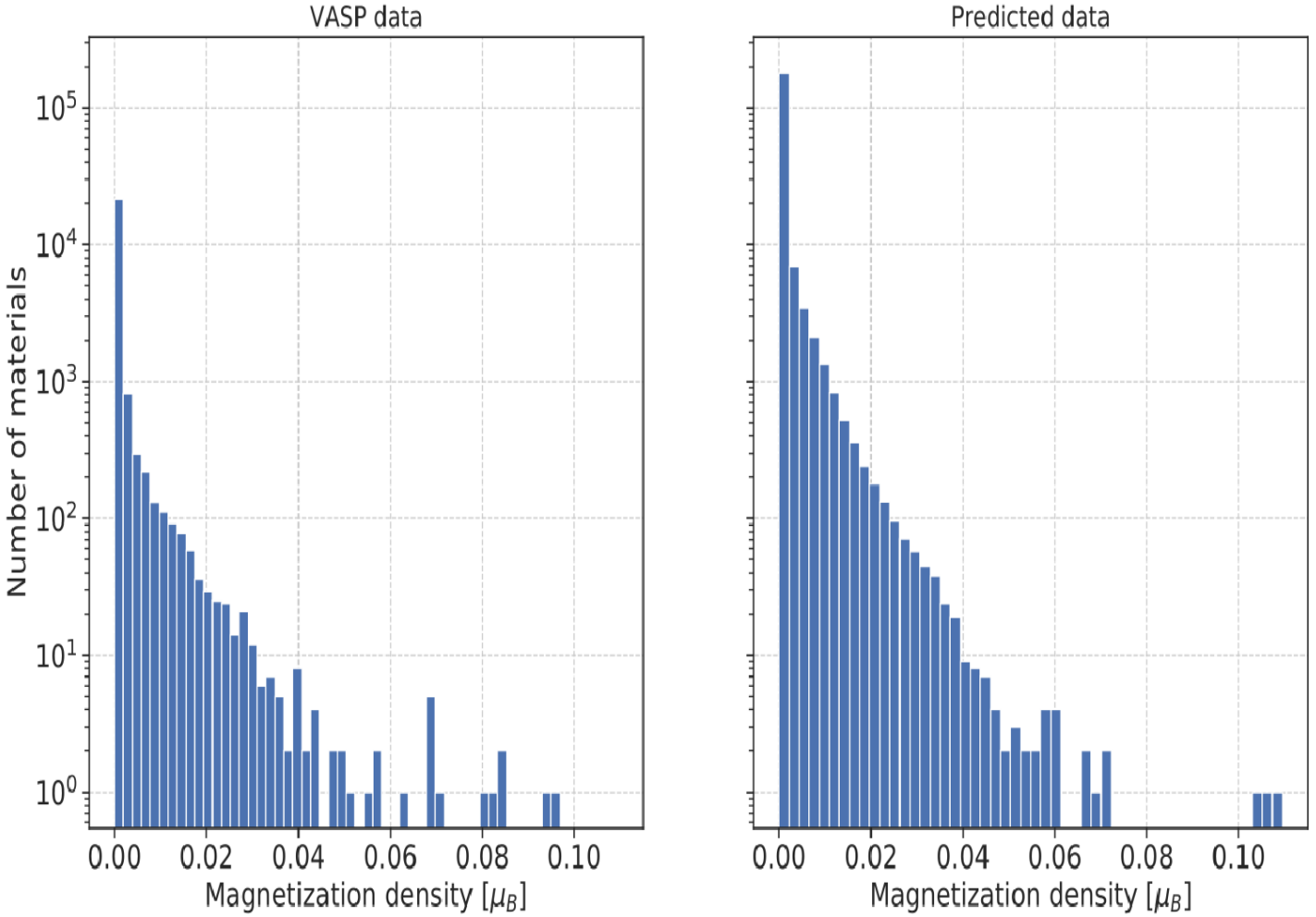
Blue asterisks represent missclassified materials

Removing missclassified data (magnetic or not magnetic)

r^2=0.89

which is a large improvement

Prediction of magnetization density

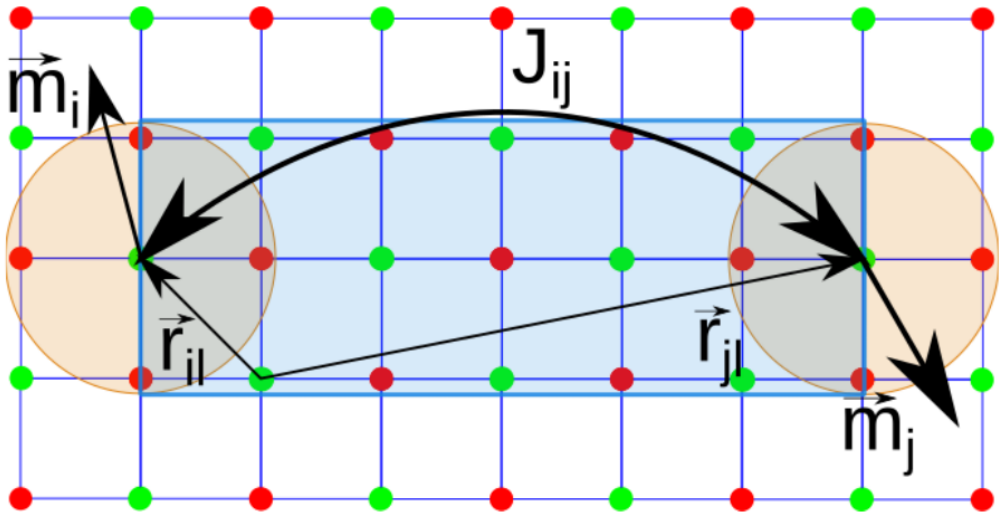


Distribution of DFT calculated magnetization density for 23,486 materials contained in the OMDB.

Distribution of machine learning predicted magnetization density for 196,471 materials.

Prediction of Heisenberg interactions – representation

Idea: The interaction depend on the local chemistry of sites $\mathbf{r}_i$ and $\mathbf{r}_j$, and the linking region.



Conclusions

- Extending the OMDB to magnetic excitations
- High throughput calculation of magnetic Hamiltonians and spin wave spectra
- Machine learning for magnetic properties

Conclusions

- Extending the OMDB to magnetic excitations
- High throughput calculation of magnetic Hamiltonians and spin wave spectra
- Machine learning for site-specific magnetic properties

Thank you for your attention!

Organic Materials Database

https://omdb.mathub.io