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Mapping Density Functional Theorem onto Heisenberg model

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Motivation

Employing Quantum Mechanics in modelling thermal properties of new materials has a significant disadvantage - far from the thermodynamic limit we do not have temperature defined. Hence to get transitional temperatures e.g. **Curie (Néel)** T_C **(**T_N**)** for a proposed **magnetic material**, we are limited to to the model calculations, e.g. the established atomistic spin-dynamics (ASD) simulations [1,2]. This in turn requires us to find the explicit form of the magnetic **Heisenberg Hamiltonian** (cf. *Some Math* for more details)

$$\mathcal{H} \equiv -\frac{1}{2}\sum_{i\neq j} J_{ij}\mathbf{s}_i \cdot \mathbf{s}_j,$$

where J_{ij} are exchange interaction magnitudes of unitary vectors $\{\mathbf{s}_i\}$ on a given lattice.

We are aiming at creating **a generic approach** removing the burden of designing the Hamiltonian from user while **minimising the computational cost** (in contrast to the Korringa-Kohn-Rostoker (KKR) Green function formalism [3] and the frozen-magnon approach [4]).

Some Math & Physics

We assume that there exist a block-diagonal **effective Hamiltonian** $\hat{\mathcal{H}}_{\text{eff}}$ corresponding to the resultant state of our DFT calculations

$$\hat{\mathcal{H}}_{\text{eff}} = \begin{pmatrix} \hat{\mathcal{H}}_{\text{magnetic}} & \\ & \hat{\mathcal{H}}_{\text{remaindered}} \end{pmatrix}.$$

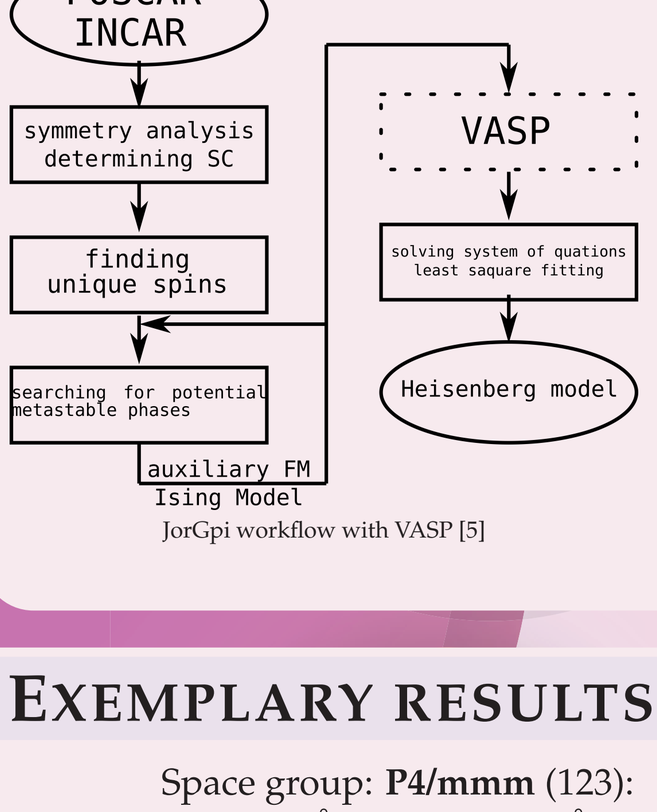
where $\hat{\mathcal{H}}_{\text{magnetic}} = \hat{\mathcal{H}}(\{\hat{\mathbf{M}}_i\})$ depends on the on-site magnetization operators $\{\hat{\mathbf{M}}_i\}$. For the case of colinear-spin system this simplifies to

$$\Delta E_{n\in N} \overset{\Delta M \ll M}{\approx} \sum_{\langle i,j\rangle} -\mathbf{J}_{ij} M_i^0 M_j^n,$$

where we have **a set of** N **metastable magnetic states** with magnetization norm $M_i^n \equiv M_i^0 + \Delta M_i^n$ (i, j are sites with exactly one spin *flipped*).

Mapping software: JorGπ

The aim is to process the **ab-initio model** of magnetic material, generate a number of possibly stable states with rotated spin, and calculate **the effective Heisenberg model** from the output.



JorGpi workflow with VASP [5]

JorGpi requires `Python 3.6` with

numpy	scipy	spglib	matplotlib	setuptools	defusedxml
1.16.0	1.0.0	1.12.0	3.0.0	40.8.0	-

The bottleneck section - **Ising model solver** (cf. *Metastable states*) utilizes `C++17` and GNU Scientific Library.

```
usage: __main__.py [-h] [--input INPUT] [--incar INCAR] [--output OUTPUT]
                   [--cutOff CUTOFF | --neighbor NEIGHBOR]
                   [--Wyckoffs WYCKOFFS] [--reference REFERENCE]
                   [--elements ELEMENTS]
                   [--group {1,2,3,4,5,6,7,8,9,10,11,12,13,14,15,16,17,18} [{1,2,3,4,5,6,7,8,9,10,11,12,1
3,14,15,16,17,18} ...]]
                   [--period {2p,3p,4p,5p,6p,3d,4d,5d,4f,5f} [{2p,3p,4p,5p,6p,3d,4d,5d,4f,5f} ...]]
                   [--block {P,D,F} [{P,D,F} ...]] [--symmetry] [--redundant]
                   [--spin-orbit] [--refined]
                   [--extra-dimentions EXTRA-DIMENTIONS]

Find minimal number of unique spin-flips

optional arguments:
  -h, --help            show this help message and exit
  --input INPUT, -i INPUT
                        input POSCAR file
  --incar INCAR, --INCAR INCAR, -I INCAR
                        input INCAR file
  --output OUTPUT, -o OUTPUT
                        output directory
  --cutOff CUTOFF, -R CUTOFF
                        a cut-off distance (in Å) for calculations
  --neighbor NEIGHBOR, -N NEIGHBOR
                        a rank of the last Neighbor taken into account
  --Wyckoffs WYCKOFFS, -W WYCKOFFS
                        narrows down the atomic selection to the atoms in
                        positions defined by string (eg. 'abc')
  --reference REFERENCE, -r REFERENCE
                        number of reference atom in inputFile
  --elements ELEMENTS, -E ELEMENTS
                        string of all elements taken into account (eg. 'CuO')
  --group {1,2,3,4,5,6,7,8,9,10,11,12,13,14,15,16,17,18} [{1,2,3,4,5,6,7,8,9,10,11,12,13,14,15,16,17,18}
...]
                        group number (eg. 1 <=> 'HLiNaKRbCsFr')
  --period {2p,3p,4p,5p,6p,3d,4d,5d,4f,5f} [{2p,3p,4p,5p,6p,3d,4d,5d,4f,5f} ...]
                        period name (eg. 3d <=>
                        '$Sc$Ti$V$Cr$Mn$Fe$Co$Ni$Cu$Zn$')
  --block {P,D,F} [{P,D,F} ...]
                        block name (eg. P <=> '$B$C$N$O$F$Al$Si$P$S$Cl$Ga$Ge$A
                        s$Se$Br$In$Sn$Sb$Te$I$Tl$Pb$Bi$Po$At$')
  --symmetry, -S        symmetry run only (default False)
  --redundant           creates a redundant system of equations for final
                        calculation of the Heisenberg exchange interaction
                        (default False)
  --spin-orbit, --SOC   (work-in-progress) is sping-orbit coupling enabled
                        (default False)
  --refined             should use refined supercell (default False)
  --extra-dimentions EXTRA-DIMENTIONS, -X EXTRA-DIMENTIONS
                        string "X Y Z" of extra cell copies in each directions
                        (eg. "0 0 1")
```

Metastable states

range	# sites	# possible states
1 NN	2	$2^1 - 1 = 1$
2 NN	8	$2^7 - 1 = 127$
3 NN	16	$2^{15} - 1 = 32\,767$
5 NN	54	$2^{53} - 1 \sim 10^{16}$
8 NN	128	$2^{127} - 1 \sim 10^{38}$
13 NN	250	$2^{249} - 1 \sim 10^{75}$
21 NN	432	$2^{431} - 1 \sim 10^{130}$

Number of possible states (assuming spin-reversal symmetry) of a Bravais bcc lattice versus range of interactions.

> In *the idealized magnetic ground state* the ordering of magnetic moment directions $\mathbf{s}_i^0$ is **unambiguously defined by sign** of exchange interaction magnitudes J_{ij}.

I.e., to find metastable sates we use **a ferromagnetic** (!) 3D Ising model with a solver using Simulated Annealing.

$$H = -\sum_{i\neq j} K_{ij}\sigma_i\sigma_j, \;\&\; K_{ij} \equiv J_{ij}\mathbf{s}_i^0 \cdot \mathbf{s}_j^0 > 0.$$

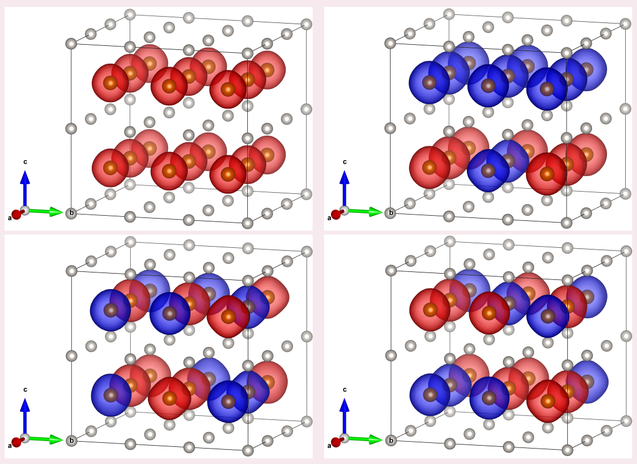
Exemplary results - FePt

Space group: **P4/mmm** (123):

$a = b = 2.66084$ Å; $c = 3.68635$ Å; $\alpha = \beta = \gamma = 90°$

	μ_{Fe} (μ_B)	μ_{Pt} (μ_B)
FLEUR [6]	2.866	0.384
VASP [5]	2.823	0.318
JorGpi	**2.849**	**0.315**

	Distance (Å)	FLEUR [6]	SPR-KKR [7]	JorGpi	Ref. [8]
$J_{[100]}$ (meV)	2.661	25.73	31.52	**35.77**	22.25
$J_{[001]}$ (meV)	3.686	8.82	-3.72	**13.28**	1.15
$J_{[110]}$ (meV)	3.763	19.89	17.56	**19.24**	18.58
$J_{[101]}$ (meV)	4.546	8.20	8.58	**15.67**	16.03
$J_{[111]}$ (meV)	5.268	-15.32	-10.58	**-21.07**	-19.75



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Background vector created by Freepik
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