Towards a Feature Space for Magnetic Structures

M.-T. Huebsch, T. Nomoto, M.-T. Suzuki, R. Arita

this poster

Many calculations in solid state physics become dramatically more efficient by using an orthonormal basis adapted to the symmetry of the material. The **cluster multipole (CMP) expansion** for magnetic structures [1] provides an ***orthonormal basis set of magnetic configurations based on the crystallographic point group***. Recent discussions in terms of CMPs established a novel perspective on transverse response properties [2] and spin dynamics in non-collinear antiferromagnets [3]. Our analysis of the experimental data found on MAGNDATA provided by Bilbao Crystallographic Server [4] suggests that only a few terms are finite in the CMP expansion of the most stable configurations in nature. This sparseness can be exploited to **build a larger learning database**, however that is provided that a ***reliable prediction of the most stable configuration can be made–possibly by means of ab-initio calculations***.

Build a database of magnetic structures,

- obeying the 5 V's of Big Data [5]
- using CMP expansion to create candidate magnetic configurations [1]
- determining the most stable configuration by LSDA [6] using VASP [7]

Antiferromagnetic materials are particularly interesting [8], because they

- are robust against perturbation due to magnetic fields,
- produce no stray fields,
- feature ultrafast dynamics, and
- can generate large magnetotransport effects.

Optimized antiferromagnetic materials can open doors for

seamless and low-maintenance energy generation [9]

ultrafast spintronics and robust data retention [3,8]

fundamental understanding of magnetotransport [2,10-12]

Within the data-driven paradigm, **material discovery** [5] can be seen as a learning problem. That necessitates building a database of materials and the desired descriptor, for instance this database might contain crystals and their magnetic configurations.

Big Data 5 V's

1. Volume Size of data

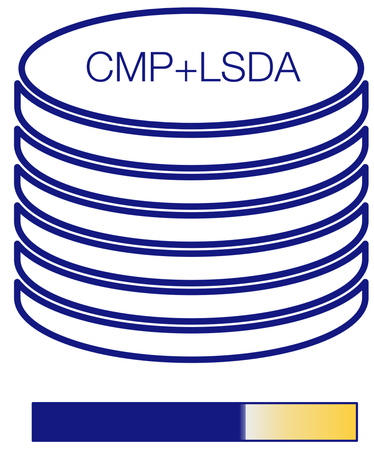
2. Velocity Speed of data generation

3. Variety Different types of data

4. Veracity Accuracy and reliability of data

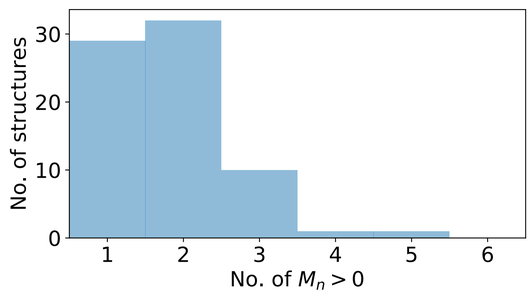
5. Value Extracting useful information from data

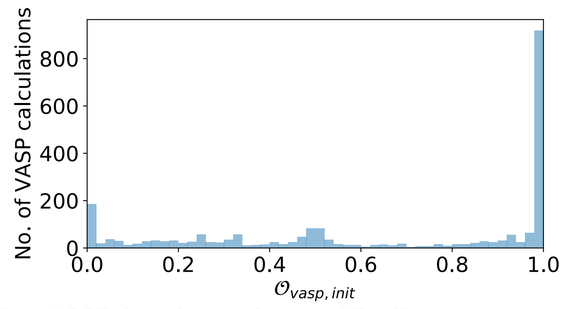
MAGNDATA [4] is a collection of experimentally determined magnetic structures with portable cif-type files. This sets the path for developing a reliable scheme of computational data generation that obeys the 5 V's of Big Data.



Preliminary results

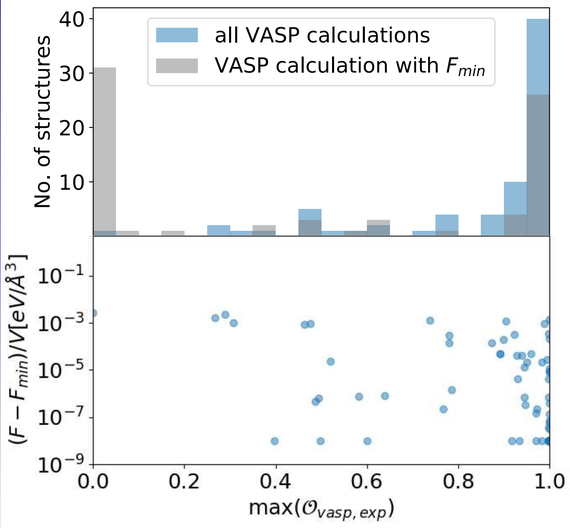
- For 73 (of 171) structures containing 3d transition metals on MAGNDATA with $\mathbf{q} = 0$.





The CMP expansion is a natural expansion for experimental magnetic structures.

- Only about 1-3 CMP coefficients are finite among $3N_{atom}$ magnetic configurations in the CMP basis.



The CMP basis and specific linear combinations with same rank and irreducible representation administer an exhaustive list of candidate magnetic structures.

- Most converged VASP calculations have a large overlap with the initial magnetic configuration; $\mathcal{O}_{vasp,init} \approx 1$.
- Some initial magnetic configurations have almost no overlap with the converged VASP calculation; $\mathcal{O}_{vasp,init} \approx 0$.

  Presumably, they are very unstable, because they might correspond to local maxima in the LSDA free energy landscape.

A few initial magnetic configurations converge towards a magnetic configuration that is a linear configuration of itself and another CMP with likely the same irreducible representation.

CMP+LSDA can find the local minimum in the LSDA free energy landscape closely resembling the experimental magnetic structure.

- For most structures the maximum overlap among all VASP calculations w. the experimental configuration is close to 1; (blue).
- The VASP calculation w. lowest found free energy F_{min} does often *not* correspond to the experiment; (blue vs. gray).
- The structures with similar energy gaps, that is btw. the F_{min} configuration and the local minimum closest to the experimental configuration, are currently being analyzed for commonalities.

Cluster multipole (CMP) expansion [1, 12-13]

Vector Poisson equation : $\nabla^2 \mathbf{A}(\mathbf{r}) = -\frac{4\pi}{\mathbf{c}} \mathbf{j}(\mathbf{r})$

current density: $\mathbf{j}(\mathbf{r}) = \mathbf{c}\nabla \times \mathbf{M}(\mathbf{r})$, magnetization density: $\mathbf{M}(\mathbf{r})$

Vector gauge potential: $\mathbf{A}(\mathbf{r}) = \sum_{lm} b_l M_{lm} \frac{\mathbf{Y}^l_{lm}}{r^{l+1}}$,

vector spherical harmonics : $\mathbf{Y}^l_{lm} = \frac{\mathbf{l} Y^l_m(\theta,\phi)}{\mathrm{i}l} \quad (l \geq 1, -l \leq m \leq l)$,

Coulomb gauge : $\nabla \cdot \mathbf{A}(\mathbf{r}) = \mathbf{0}$.

Magnetic cluster multipole (CMP): $M_{lm} = \sqrt{\frac{4\pi}{2l+1}} \sum_{i=1}^{N_{atom}} \mathbf{m}_i \cdot \left[\nabla \left(\mathbf{r}_i Y^*_{lm}(\theta,\phi) \right) \right]$.

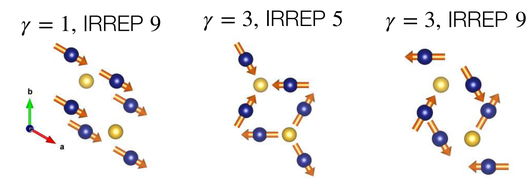
magnetic configuration $|m\rangle = (\mathbf{m_1}, \mathbf{m_2}, \ldots, \mathbf{m_{N_{atom}}})^{\mathbf{T}}$

unit cell of N_{atom} magnetic sites.

Symmetrization according to irreducible representations of the crystallographic point group

$$\mathcal{Y}_{l\gamma} = \sum_m c^\gamma_{lm} Y_{lm}, \quad \gamma = 1,\ldots,2l+1, \quad l\gamma \mapsto n$$

Some example CMP configurations for Mn_3Sn:



Cluster multipole basis set $\left\{ |n\rangle = = (\mathbf{e}^n_1, \mathbf{e}^n_2, \ldots, \mathbf{e}^n_{N_{atom}})^T \right\}$,

complete : $\sum_{n=1}^{3N_{atom}} \frac{|n\rangle\langle n|}{N_{atom}} = \mathbf{1}$, orthonormal : $\langle n|n'\rangle = \delta_{nn'} N_{atom}$

symmetry-adapted CMP: $M_n = \sum_{i=1}^{N_{atom}} \mathbf{m}_i \cdot \mathbf{e}^n_i = \langle m|n\rangle = \langle n|m\rangle$

arbitrary magnetic configuration: $|m\rangle = \frac{1}{N_{atom}} \sum_{n=1}^{N_{atom}} M_n |n\rangle$.

overlap of magnetic configurations: $\mathcal{O}_{mm'} = \left(\frac{\langle m|m'\rangle}{\sqrt{\langle m|m\rangle}\sqrt{\langle m'|m'\rangle}} \right)^2$

[1] M.-T. Suzuki *et al.*, Phys. Rev. B **99**, 174407 (2019).
[2] T. Higo *et al.*, Nat. Photonics **12**, 73–78 (2018).
[3] T. Nomoto and R. Arita, arXiv:1903.02259.
[4] S. V. Gallego *et al.*, J. Appl. Cryst. **49**, 1941–1956 (2016).
[5] G. R. Schleder *et al.*, J. Phys. Mater. **2** 032001 (2019).
[6] D. Hobbs, G. Kresse and J. Hafner, Phys. Rev. B. **62**, 11 556 (2000).
[7] G. Kresse and J. Hafner, Phys. Rev. B **47** , 558 (1993); ibid. **49** , 14 251 (1994).
[8] V. Baltz *et al.*, Rev. Mod. Phys. **90**, 015005 (2018).
[9] M. Ikhlas *et al.*, Nature Physics **13**, 1085–1090 (2017).
[10] T. Jungwirth *et al.*, Nat Nanotechnol., **11**, 231-41 (2016).
[11] M. Seemann *et al.*, Phys. Rev. B **92**,155138 (2015).
[12] M.-T. Suzuki *et al.*, Phys. Rev. B **95**, 094406 (2017).
[13] H. Kusunose, JPSJ **77**, 06 4710 (2008).

RIKEN Since 1917 · Programs for Junior Scientists · 東京大学 THE UNIVERSITY OF TOKYO

www.arita-lab.t.u-tokyo.ac.jp

marie-therese.huebsch@riken.jp