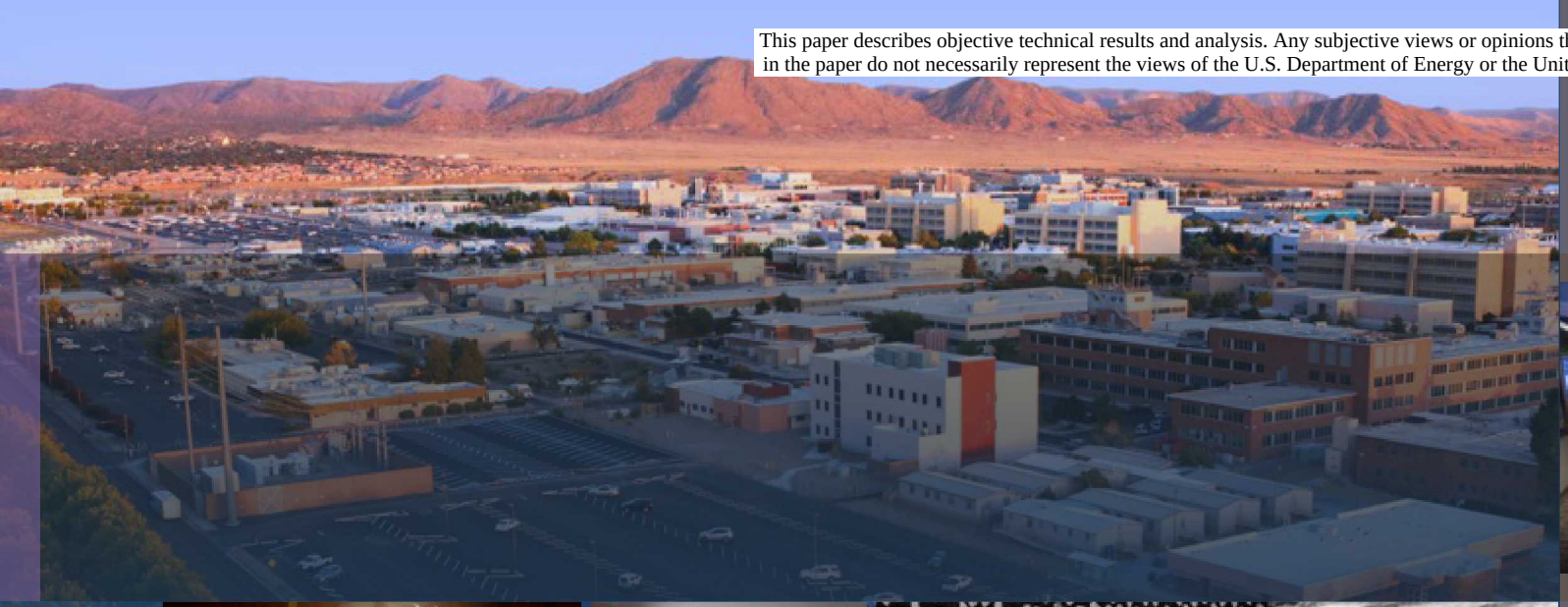

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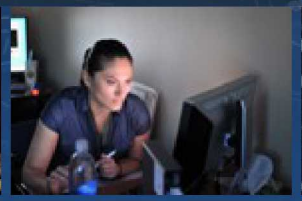

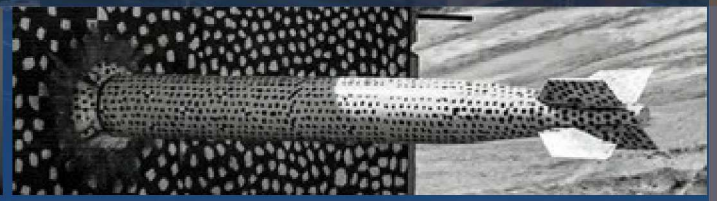

Improving the accuracy of spin-lattice simulations with machine-learning interatomic potentials

PRESENTED BY

J. Tranchida, A.Cangi, M.A. Wood, A.P. Thompson, M. Desjarlais

Contact: jtranch@sandia.gov

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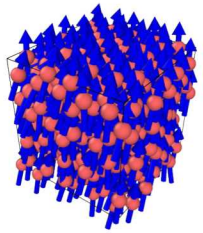
Coupled Spin Dynamics and Molecular Dynamics (SD-MD) methodology

SD-MD methodology:

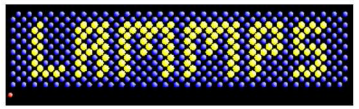
- Adding classical magnetic spins to MD:
  - For every magnetic atom in the system, one has:

$$\begin{cases} \frac{\partial \boldsymbol{r}_i}{\partial t} = \boldsymbol{v}_i \\ \frac{\partial \boldsymbol{v}_i}{\partial t} = \boldsymbol{F}_i\left(\boldsymbol{r}_{ij}, \boldsymbol{s}_{i,j}\right) \\ \frac{\partial \boldsymbol{s}_i}{\partial t} = \boldsymbol{\omega}_i \times \boldsymbol{s}_i \end{cases}$$



$$\boldsymbol{H}_{sl} = \underbrace{\sum_{i=1}^{N} \frac{|\boldsymbol{p}_i|^2}{2m_i} + \sum_{i,j=1}^{N} V\left(r_{ij}\right)}_{\text{MD Hamiltonian}} \underbrace{- \sum_{i,j,i\neq j}^{N} J\left(r_{ij}\right)\, \boldsymbol{s}_i \cdot \boldsymbol{s}_j - \mu_B \mu_0 \sum_{i=0}^{N} g_i \boldsymbol{s}_i \cdot \boldsymbol{H}_{ext}}_{\text{Spin-lattice coupling}}$$

SPIN package in LAMMPS



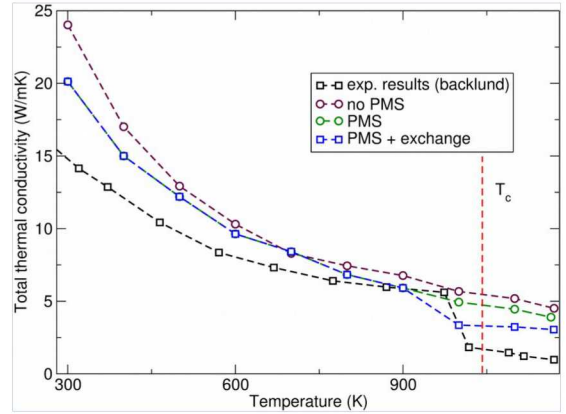
- Scalable symplectic algorithms (massively parallel spin-lattice dynamics).
- Nine types of magnetic interactions (exchange, Zeeman, DMI, ...).
- Resolution of long-range magnetic interactions (Ewald and P3M).
- Three spin minimizers and magnetic GNEB method.

Tranchida, Julien, et al. "Massively parallel symplectic algorithm for coupled magnetic spin dynamics and molecular dynamics." Journal of Computational Physics 372 (2018): 406-425.

Examples of application:

Thermal transport in magnetic materials

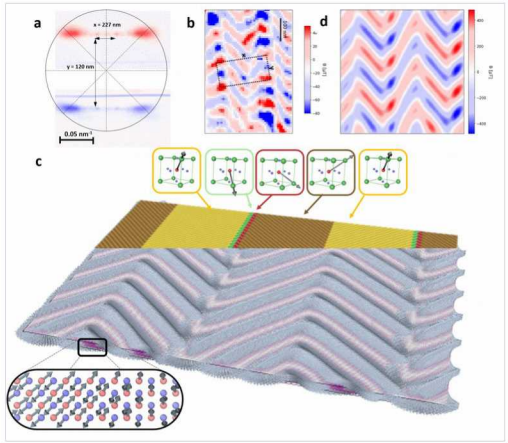
- Abnormal behavior: drop of the lattice thermal conductivity at Tc.
- Green-Kubo equilibrium atomic and spin dynamics approach.



Zhou, Yanguang, et al. "Atomistic Simulation of Phonon and Magnon Thermal Transport across the Ferro-Paramagnetic Transition." arXiv preprint arXiv:1901.00966 (2019).

Magnetic textures in multiferroic materials

- Large scale simulation of spin texttures in Bismuth ferrite.
- Simulation of magnetic configurations at ferroelectric domain walls.

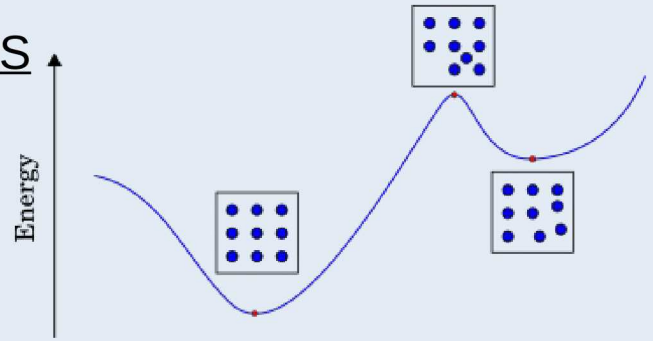


Chauleau, J-Y., et al. "Electric and antiferromagnetic chiral textures at multiferroic domain walls." Nature materials (2019): 1-5.

Spin and Lattice potential energy surfaces (PES), and common approach in S-L dynamics

Molecular dynamics PES

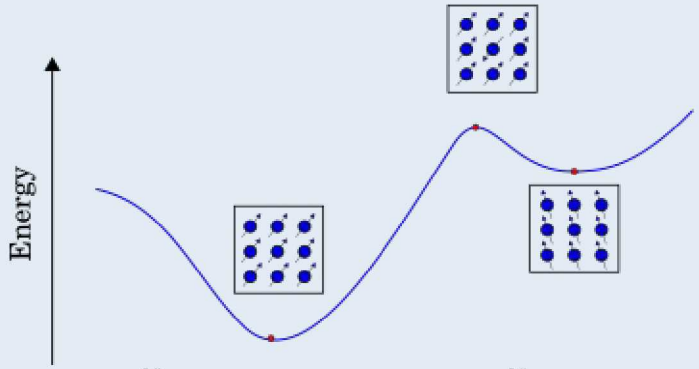
- In metals, usually an EAM-like potential.
- Parametrization can be on ab-initio data, or on experiments (elastic constants, binding-energies, …).



$$\mathcal{H} = \sum_{i=1}^{N} \frac{|\boldsymbol{p}_i|^2}{2m_i} + \sum_{i,j=1}^{N} V\left(r_{ij}\right)$$

Spin dynamics PES

- Exchange interaction, Zeeman, anisotropy, ...
- Parametrized from 1st principles calculations (spin-spirals, ...), or exp. observables (Tc, ...).

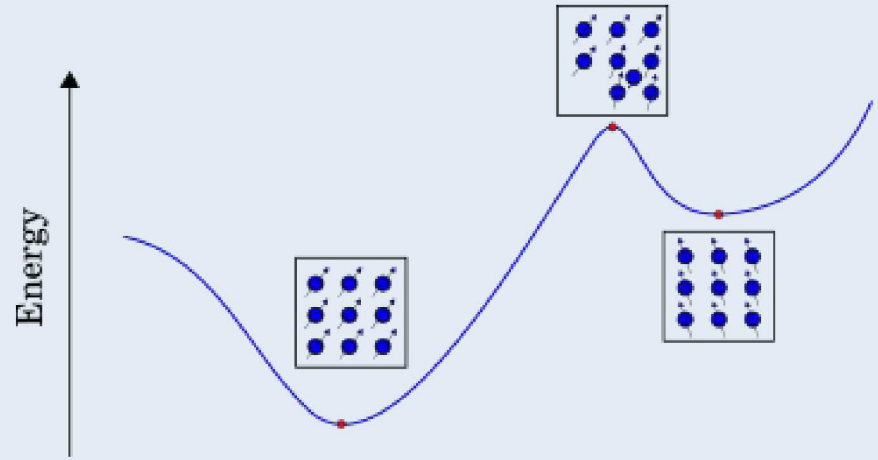


$$\mathcal{H}_s = -\sum_{i,j}^{N} J\left(r_{ij}\right)\ \boldsymbol{s}_i \cdot \boldsymbol{s}_j - \mu_B \mu_0 \sum_{i=1}^{N} g_i \boldsymbol{s}_i \cdot \boldsymbol{H}_{ext}$$

Coupled spin-lattice PES

- Usual approach: overlaying the magnetic and mechanical PES.

$$\mathcal{H}_{sl} = \underbrace{\sum_{i=1}^{N} \frac{|\boldsymbol{p}_i|^2}{2m_i} + \sum_{i,j=1}^{N} V\left(r_{ij}\right)}_{\text{MD Hamiltonian}} \underbrace{- \sum_{i,j,i\neq j}^{N} J\left(r_{ij}\right)\ \boldsymbol{s}_i \cdot \boldsymbol{s}_j - \mu_B \mu_0 \sum_{i=0}^{N} g_i \boldsymbol{s}_i \cdot \boldsymbol{H}_{ext}}_{\text{Spin-lattice coupling}}$$



- Proved sufficient to reproduce qualitative results (magnon-phonon scattering, thermal conductivity, …).

✖ Limitations: not internally consistent, cannot reproduce quantitative DFT results.

A framework for generating magneto-elastic PES for iron, using machine-learning interatomic potentials

Generating DB

Fitting a Spin Hamiltonian

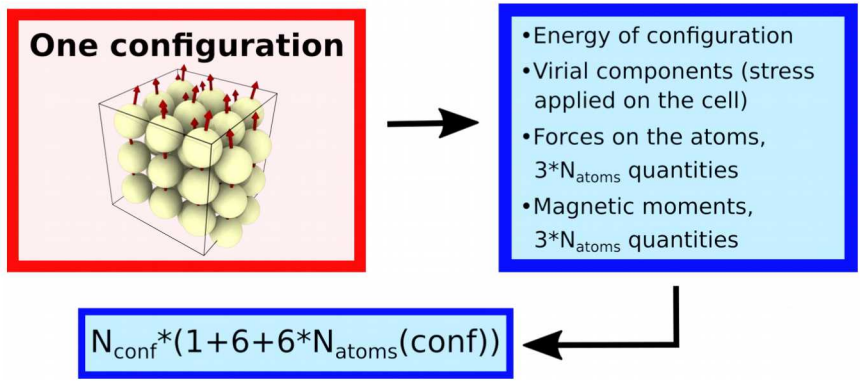
Subtracting magnetic PES

Training a ML-Interatomic potential

Running SD-MD simulations

Content of a database (DB) for machine-learning interatomic potential training

- We focus on **iron**:
  - Weak spin-orbit coupling, but...
  - Interesting spin-lattice coupling at high pressure,
  - Well-known and broadly studied.
- ML-IAP are trained on a DB of atomic configurations:



- ML inter-atomic potential reflects the configurations is in the DB:
  - The physical relevance of the configurations is fundamental.
  - Interatomic potential ‘a la carte’: configurations for a given range of validity.

- Example: if I need a potential simulating phonons and elastic constants only, I will only perform the corresponding ab initio calculations.

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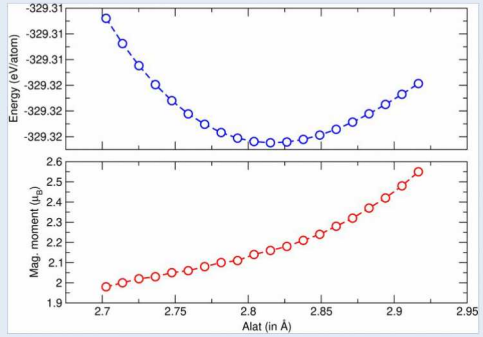
Generating a DB for machine-learning interatomic potential training

- Each configuration is the result of a self-consistent Density Functional Theory (DFT) calculation (performed with Quantum Espresso or VASP).

- Numerical verifications: k-points and PW cutoff convergence, number bands (when high smearing), energy convergence.
- Physics verifications: lattice constant, bulk modulus, Bohr magneton.
- Chosen DFT setup:
  - PBE pseudo-potential
  - Non-collinear and spin-orbit coupling
  - Smearing corresponding to 300K.
- Two sets of configurations generated.

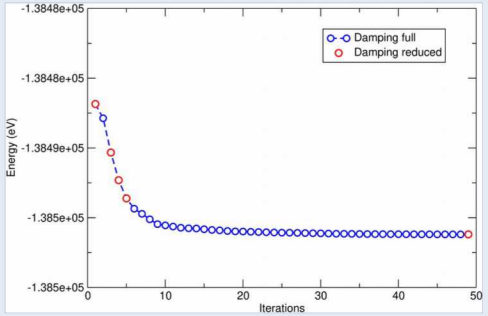
Equation of state configurations

- Lattice compression expansion (~ +/- 4%) on decorrelated 54 atom cells.
- Each run has 20 configurations.
- This group has 200 configurations.



DFT-MD configurations

- Performing DFT-MD runs on 128 atom cells.
- Decorrelated snapshots
- 40 configurations are stored.



Söderlind, Per, and A. Gonis. "Assessing a solids-biased density-gradient functional for actinide metals." Phys. Rev. B 82.3 (2010): 033102.

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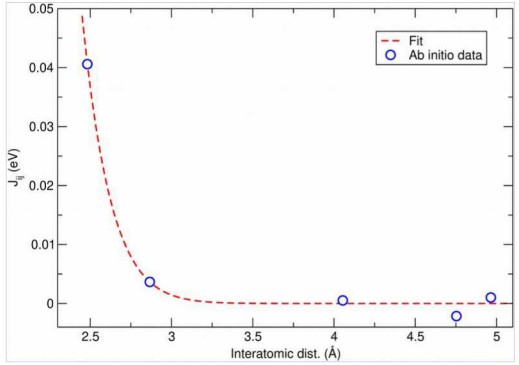
Fitting magnetic interactions on ab-initio results

- Objective: fitting an exchange interaction and a magnetic anisotropy on DFT calculations performed with the same setup as the DB generation.
  - Parametrization of a Heisenberg exchange Hamiltonian:

$$H_{ex} = -\sum_{i,j}^{N} J_{ij}(r_{ij})\, \boldsymbol{s}_i \cdot \boldsymbol{s}_j \quad \text{with} \quad J_{ij}(r_{ij}) = 4\alpha \left(\frac{r_{ij}}{\delta}\right)^2 \cos(\gamma r_{ij}) \exp\left(-\left(\frac{r_{ij}}{\delta}\right)^2\right)$$



  - For now, we are using the parametrization of a Heisenberg Hamiltonian formerly published.

- From this parametrized magnetic interaction, per-atom energy and mechanical forces for each atom of each configurations are computed and stored:

$$E_i = -\sum_{j}^{Neigh} J_{ij}(r_{ij})\, \boldsymbol{s}_i \cdot \boldsymbol{s}_j \qquad \boldsymbol{F}_i = \sum_{j}^{Neigh} \frac{d\, J_{ij}(r_{ij})}{d\, r_{ij}} (\boldsymbol{s}_i \cdot \boldsymbol{s}_j)\, \boldsymbol{e}_{ij}$$

Szilva, A., et al. "Interatomic exchange interactions for finite-temperature magnetism and nonequilibrium spin dynamics." Phys Rev. Lett. 111.12 (2013): 127204.

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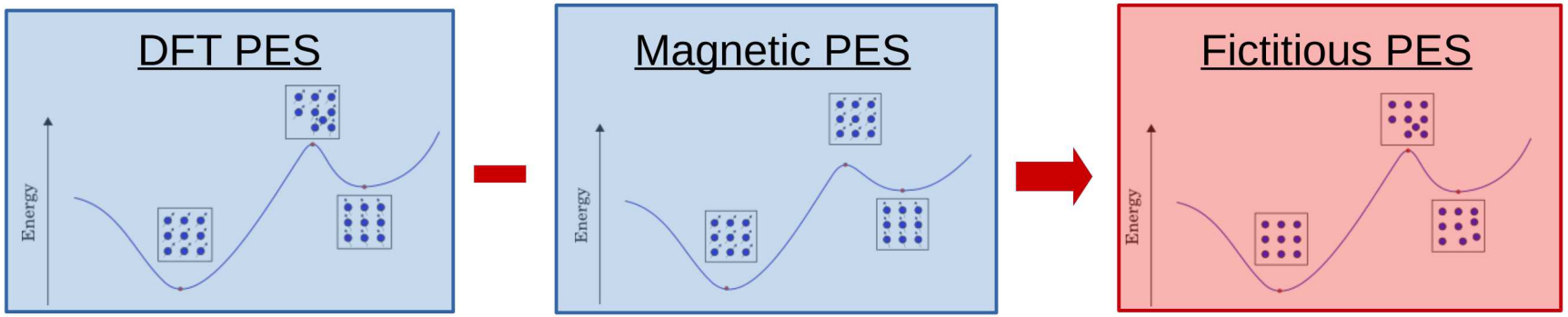
Subtracting the PES corresponding to the magnetic Hamiltonian

- The magnetic Hamiltonian is used as a reference potential.
- From each configuration, we subtract the energy, the mechanical forces (and, in principle, the virial components) generated by the parametrized spin Hamiltonian:



- Generates a fictitious DB, on which we will train a ML-interatomic potential.

$$E_i^{Fic} = E_i^{DFT} - E_i^{Mag}$$

$$\boldsymbol{F}_i^{Fic} = \boldsymbol{F}_i^{DFT} - \boldsymbol{F}_i^{Mag}$$

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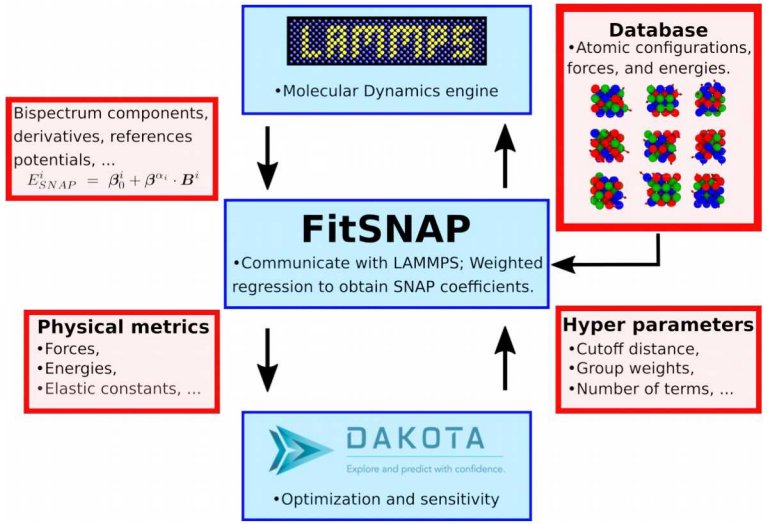
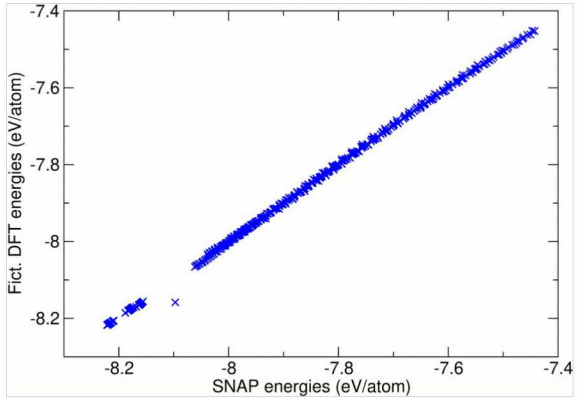
Training a machine-learning interatomic potential on the fictitious PES

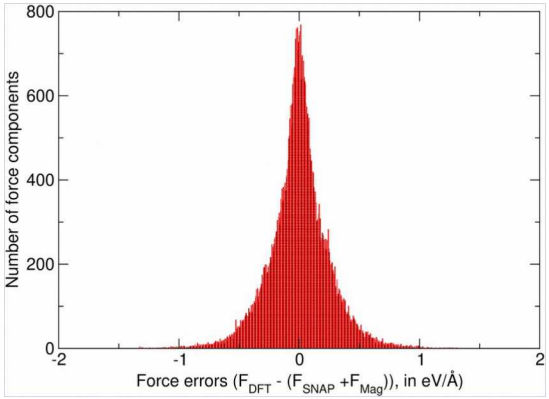
- Using the SNAP approach (developed at Sandia, implemented in LAMMPS).
- Energy and forces are expressed in terms of bispectrum components:

$$\begin{cases} E^i_{SNAP} = \beta_0 + \boldsymbol{\beta} \cdot \boldsymbol{B}^i \\ \boldsymbol{F}^j_{SNAP} = -\boldsymbol{\beta} \cdot \sum_{i=1}^{N} \frac{\partial \boldsymbol{B}_i}{\partial \boldsymbol{r}_j} \end{cases}$$



- Training results, energy and force errors:





Thompson, Aidan P., et al. "Spectral neighbor analysis method for automated generation of quantum-accurate interatomic potentials." Journal of Computational Physics 285 (2015): 316-330.

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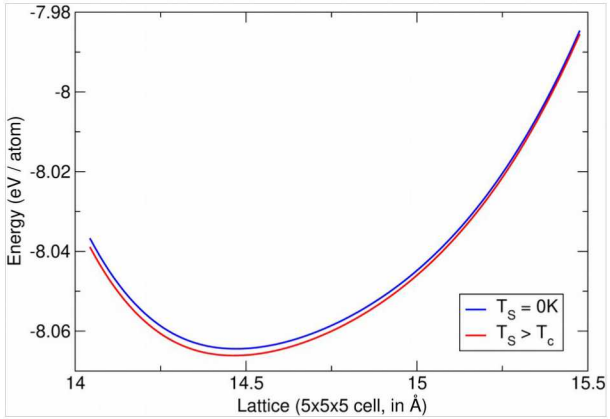
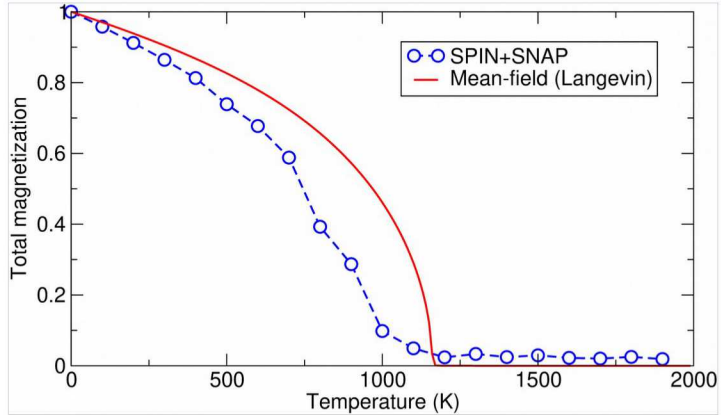
Running SD-MD simulations

- Sum-up the contributions of the SNAP and Spin potentials:

$$\mathcal{H}_{sl} = \sum_{i}^{N} \frac{|\boldsymbol{p}_i|^2}{2m_i} + \sum_{i,j}^{N} V^{SNAP}(\boldsymbol{r}_{ij}) - \sum_{i,j}^{N} J_{ij}(r_{ij})\, \boldsymbol{s}_i \cdot \boldsymbol{s}_j$$

- Tested minimization, NVE and NVT runs.
- Two calculation examples: EOS for two different spin temperatures, and Curie curve.





- Next test: influence of magnon-phonon scattering on the phonon dispersion.

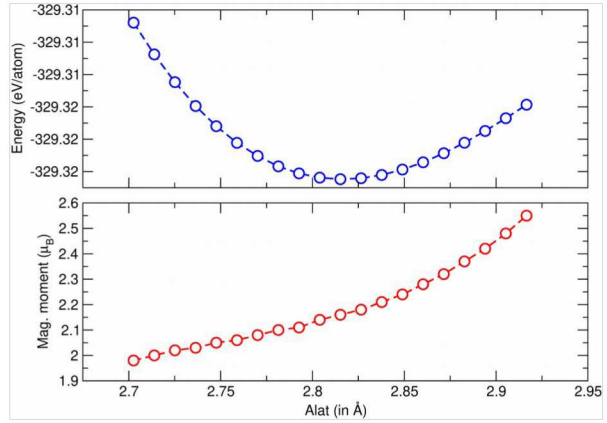
Conclusions

Results:

- A new framework aiming at improving the accuracy of coupled spin and lattice simulations was developed.
- An initial DB of configurations was produced by DFT calculations (should work for phonons and elastic constants).
- We tested the framework on iron, and for simple SD-MD calculations.

Perspectives:

- Improving the potential's range of validity by adding more diverse configuration sets.
- Improving the spin model:
  - Accounting for the effects of the spin-orbit coupling, should lead a stronger S-L coupling.
  - Accounting for longitudinal spin fluctuations (very important for nickel, or for iron at higher pressure).



- Any suggestion welcome, feel free to contact us (jtranch@sandia.gov).