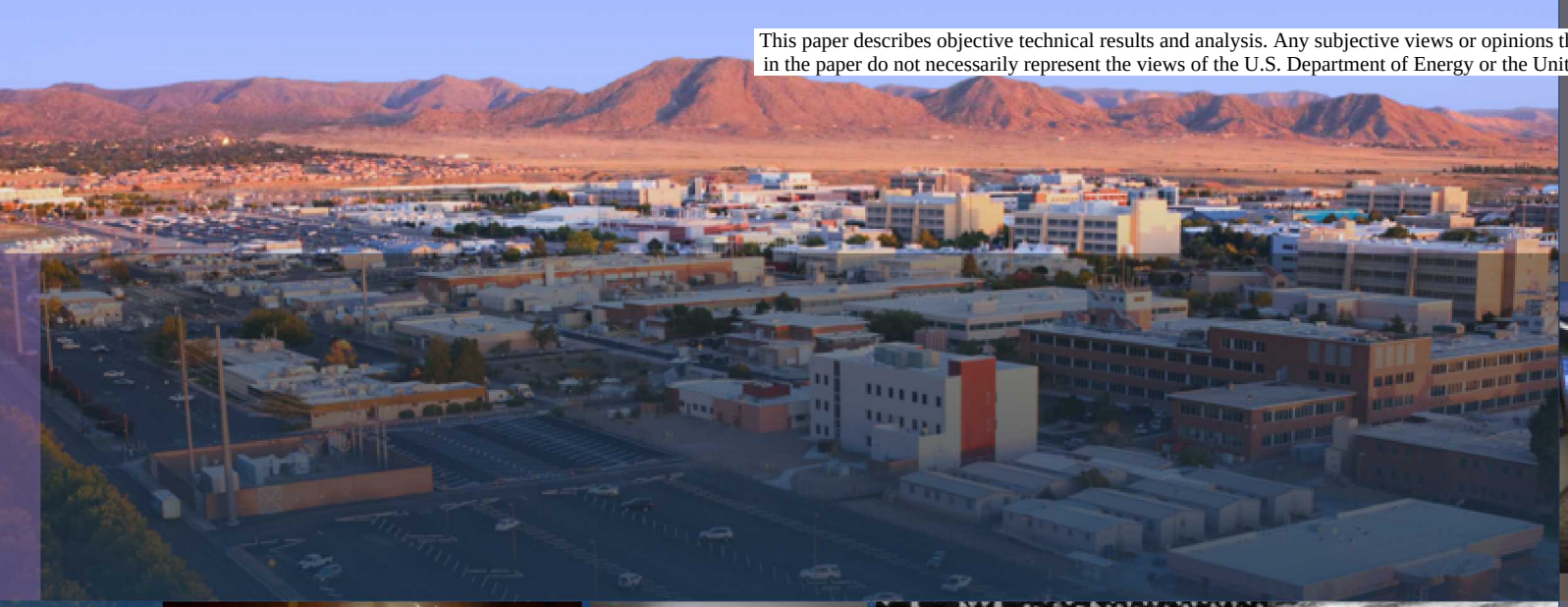

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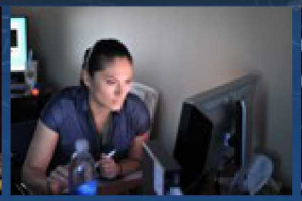

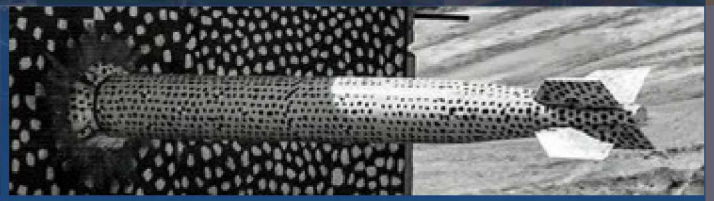

Improving the accuracy of spin-lattice calculations with machine-learned interatomic potentials

PRESENTED BY

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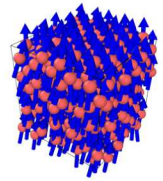
SD-MD methodology, first successes, and some current limitations

Methodology:

Coupled SD - MD

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



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

First successes:

- Simulation of magneto-caloric effects
  - Ma, P. W., & Dudarev, S. L. (2014). Dynamic magnetocaloric effect in bcc iron and hcp gadolinium. Physical Review B, 90(2), 024425.
- Influence of defects on the magnetism of bcc iron
  - Mudrick, M., et al. Combined molecular and spin dynamics simulation of bcc iron with lattice vacancies. In Journal of Physics: Conference Series (Vol. 921, No. 1, p. 012007).
  - Wen, H., Ma, P. W., & Woo, C. H. (2013). Spin-lattice dynamics study of vacancy formation and migration in ferromagnetic BCC iron. Journal of Nuclear Materials, 440(1-3), 428-434.

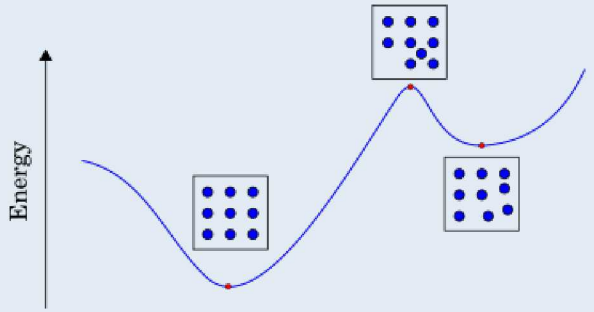
Some current limitations:

- ✖ No release in an open-source MD code
  - preventing from the development of a large user base
- ✖ Insufficient level of parallelization
  - does not allow the simulation of large magnetic devices
- ✖ Simulation of the spin-orbit coupling
  - fundamental importance for the simulation of magnetoelasticity
- ✖ Simulation of the long-range dipolar interaction
  - Unable to stabilize the domain structure of actual magnets

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

Molecular dynamics

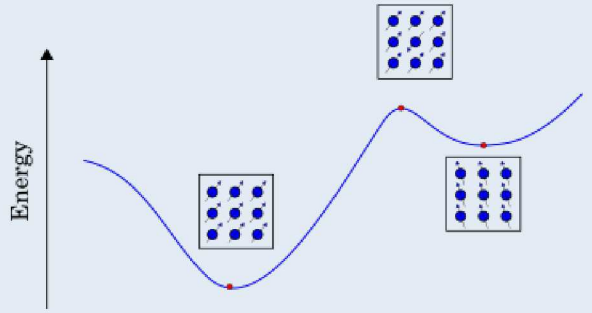
- In metals, usually an EAM-like potential.
- Parametrization can be on ab-initio data, or on observables (elastic constants, binding-energies, …).



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

Spin dynamics

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

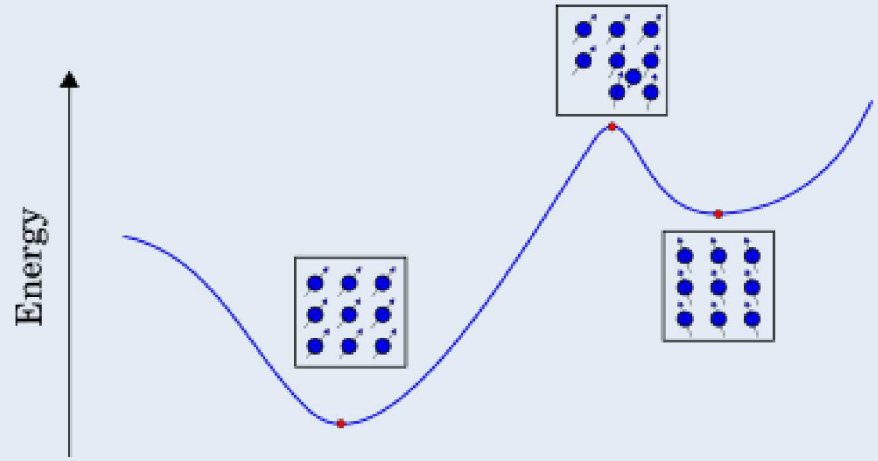


$$\mathcal{H}_s = -\sum_{i,j}^{N} J\left(r_{ij}\right)\, \boldsymbol{s}_i \cdot \boldsymbol{s}_j - \mu_B \mu_0 \sum_{i=1}^{N} g_i \boldsymbol{s}_i \cdot \boldsymbol{H}_{ext}$$

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 for a broad range of effects (magnon-phonon scattering, thermal conductivity, …).
- ✖ Limitations: overlays two PES parametrized from different calculations => in principle incorrect.

A framework for generating magneto-elastic PES using ML interatomic potentials

Generating DB

Fitting a Spin Hamiltonian

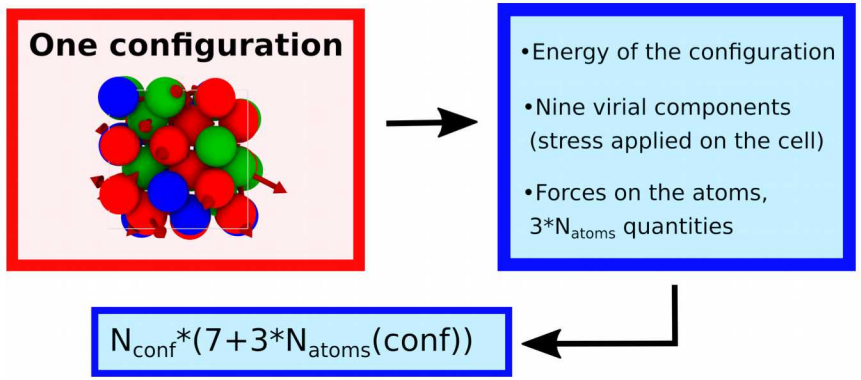
Subtracting magnetic PES

Training a ML-Interatomic potential

Running SD-MD simulations

Database generation for interatomic potential training

- DB for ML-IAP contain atomic configurations.
- Each configuration is the result of a self-consistent Density Functional Theory (DFT) calculation (performed with Quantum Espresso, SeqQuest, or VASP).



- The SNAP potential is trained on those configurations.

The physical relevance of all configurations in the DB is of fundamental importance!

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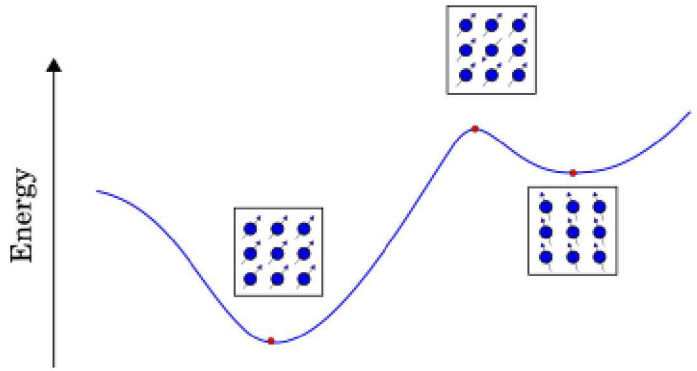
Fitting magnetic interactions on ab-initio results

- Our objective is to fit 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:

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

- Later on, adding the effects of the Spin-orbit coupling (magnetocrystalline anisotropy here).

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

Pajda, Marek, et al. "Ab initio calculations of exchange interactions, spin-wave stiffness constants, and Curie temperatures of Fe, Co, and Ni." Physical Review B 64.17 (2001): 174402.

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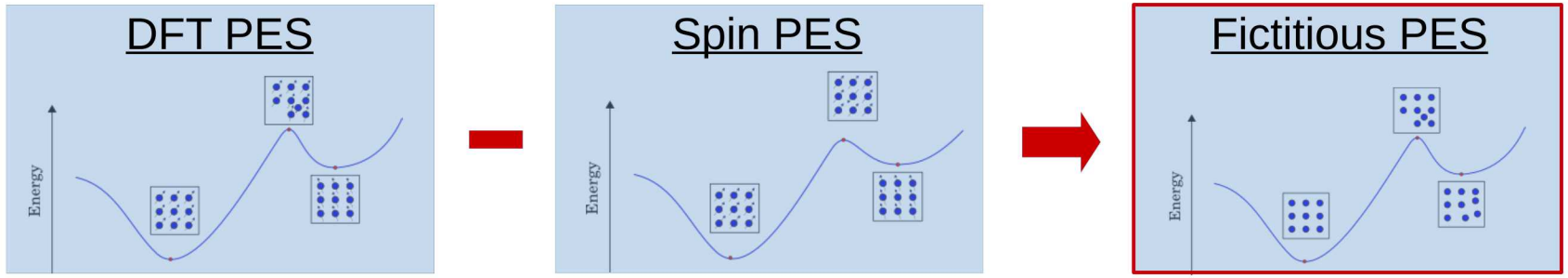
Subtracting the PES corresponding to the magnetic Hamiltonian

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



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

A framework for generating magneto-elastic PES using ML interatomic potentials

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Training a ML-Interatomic potential

Running SD-MD simulations

Training a machine-learning interatomic potential

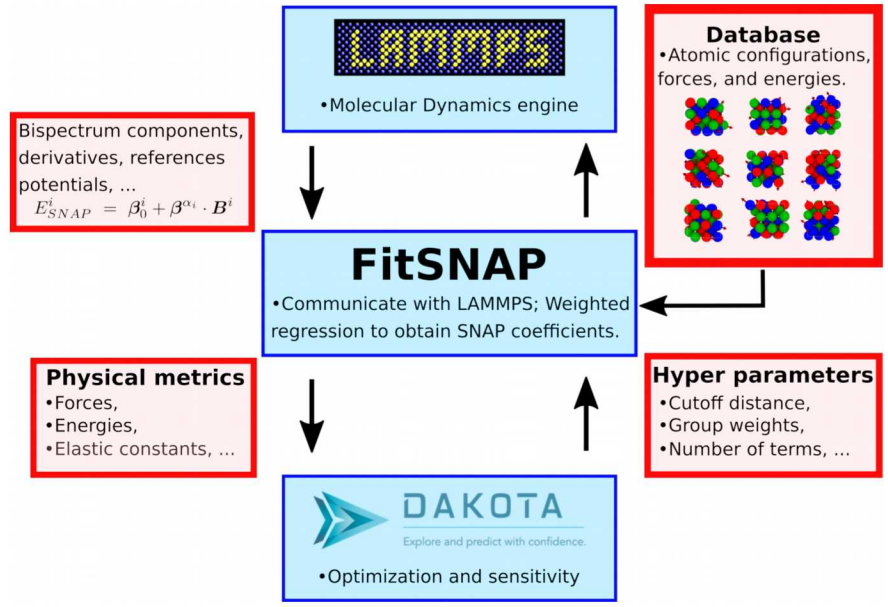
- The starting point of the approach expresses the atomic density.

$$\rho_i(\boldsymbol{r}) = \delta(\boldsymbol{r}) + \sum_{r_{ij}<R_{cut}} f_c(r_{ij})\, w_i\, \delta(\boldsymbol{r}-\boldsymbol{r}_{ij})$$

With:
- Wi weights defining atomic species
- Fc radial switching functions

- This density can be expressed in terms of bispectrum components. Then, those bispectrum components can express the energy and forces on a given atom:

$$\begin{cases} E^i_{SNAP} = \beta^i_0 + \boldsymbol{\beta}^{\alpha_i} \cdot \boldsymbol{B}^i \\ \boldsymbol{F}^j_{SNAP} = -\boldsymbol{\beta} \cdot \sum_{i=1}^{N} \dfrac{\partial \boldsymbol{B}_i}{\partial \boldsymbol{r}_j} \end{cases}$$



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.

A framework for generating magneto-elastic PES using ML interatomic potentials

Generating DB

Fitting a Spin Hamiltonian

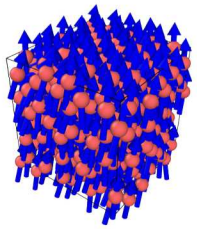
Subtracting magnetic PES

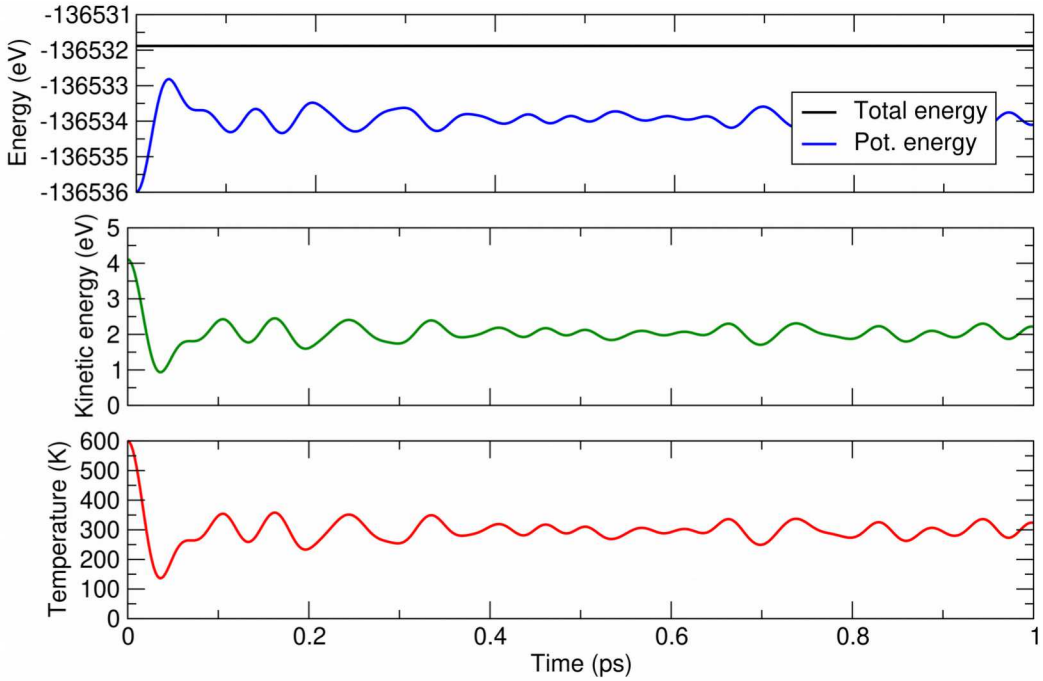
Training a ML-Interatomic potential

Running SD-MD simulations

Running SD-MD simulations

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





Conclusions

- A new framework aiming at improving the accuracy of coupled spin and lattice simulations was developed.
- We tested the framework on iron, and for simple SD-MD calculations
- Our next objective is to improve the accuracy of this potential: by adding more configurations to the DB (defects, free surfaces, …) and improving the accuracy of the spin Hamiltonian (spin-orbit coupling, longitudinal spin fluctuations, ...).
- Open to collaborations, feel free to contact us (jtranch@sandia.gov).