Spin-aware quantum-accurate interatomic potentials for heavy elements

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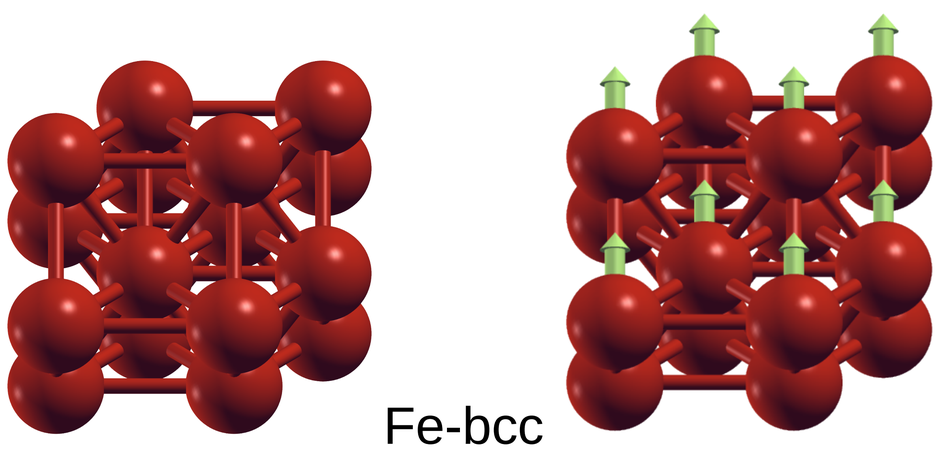
Introduction

Studying matter under extreme conditions using density functional theory (DFT) is computationally expensive, since the degrees of freedom and consequently the configurational space grows rapidly with increasing temperature and pressure. Therefore, the use of DFT for such simulations is limited to fairly small simulation cells and time scales. Machine learning-based interatomic potentials (ML-IAP) provide access to much larger spatial and temporal domains, thus enabling the discovery of new and exotic magnetic materials. A majority of existing descriptors required to construct ML-IAPs neglect the spin degrees of freedom. Here, we present our preliminary ideas/workflows to construct ***"spin-aware" ML-IAP*** using the **SNAP[1]** descriptors and the coupled spin-molecular dynamics framework implemented in LAMMPS [2]. This modeling capability will complement upcoming experiments to magneto-structural properties in shock-compressed or laser-driven samples at elevated temperatures and pressures exposed to strong, pulsed magnetic fields, which are planned at photon sources such as within the HIBEF consortium at the European XFEL.

magneto-elastic ML-IAP training Workflow

DFT Calculations | **Spin configurations**



Spin Hamiltonian

$$H_{mag} = -\sum\sum_{i\neq j}^{N} J(r_{ij})[s_j \cdot s_j - 1]$$
$$-\sum_{i\neq j}^{N} K(r_{ij})[(s_i \cdot s_j)^2 - 1]$$

Heisenberg Hamiltonian

SNAP Potential

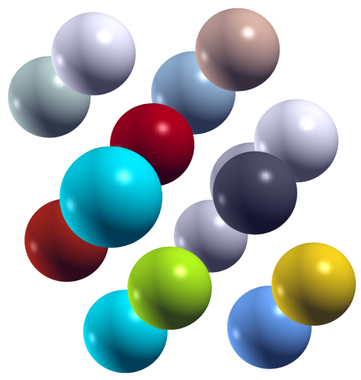
$$\rho(r) = \sum_{j=0,\frac{1}{2},\ldots}^{\infty} \sum_{m=-j}^{j} \sum_{m'=-j}^{j} u_{m,m'}^{j} U_{m,m'}^{j}(\theta_0, \theta, \phi)$$

neighbour density function

$$E_{SNAP}(r^N) = N\beta_0 + \beta \cdot \sum_{i=1}^{N} B^i$$

Optimize linear SNAP coefficients using FitSNAP/DAKOTA

Spin Lattice Dynamics



$$E = \sum_{i=1}^{N} \epsilon(\{r_{ij}, s_i\})$$
$$\vec{F} = \sum_{i=1}^{N} \vec{f}(\{r_{ij}, s_i\})$$
$$\vec{P} = \sum_{i=1}^{N} \vec{p}(\{r_{ij}, s_i\})$$

Incorporating Spin, Lattice & Temperature Fluctuations (SLT)

SNAP descriptors do not incorporate the spin degrees of freedom. A „spin-agnostic“ interatomic potential (IAP) is validated against the magneto-elastic properties in LAMMPS after the spin Hamiltonian contributions are substracted. The joint optimization of the spin Hamiltonian and IAP parameters are performed using DAKOTA and coupled spin-molecular dynamics framework implemented in LAMMPS[2].

- Integrate spin coordinates using spin-augmentation functions into SNAP
- Include momentum distribution, spin-resolved local density of states, average magnetization and radial distribution function during optimization
- Include basis functions to translate SLTs for training novel neural networks
- Investigate coupled spin-lattice properties of materials exposed to strong magnetic fields under high temperatures and pressures

[1] Thompson, Aidan P., et al. "Spectral neighbor analysis method for automated generation of quantum-accurate interatomic potentials." Journal of Computational Physics, 285, 316 (2015)
[2] Tranchida, J., et al. “Massively parallel symplectic algorithm for coupled magnetic spin dynamics and molecular dynamics”, Journal of Computational Physics 372, 406 (2018)
[3] Nikolov, Svetoslov, et al. "Data-driven magneto-elastic predictions with scalable classical spin-lattice dynamics." NPJ Computational Materials, 7, 153 (2021)
[4] Nikolov, Svetoslov, et al. "Dissociating the phononic, magnetic and electronic contributions to thermal conductivity: a computational study in alpha-iron" Journal of Materials Science (2022)
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