Structure prediction of iron hydrides at high pressures with machine-learned interatomic potentials

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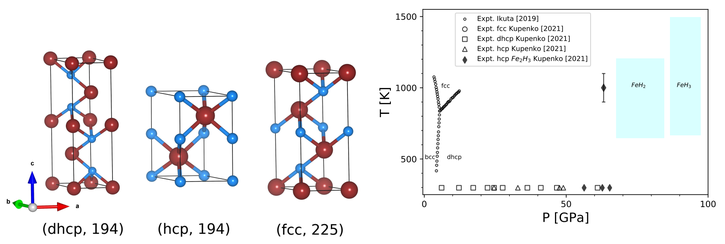
1) Motivation

- Understanding the chemical composition of Earth's core is a grand challenge in geoscience and materials science.
- Earth's inner core is believed to be composed of iron-based alloys; however, its density is 2-5% lower than of pure iron [1].
- Hydrogen is a fundamental element in the Earth's core and the primary contributor to the observed density deficit in the inner core.
- Previous theoretical and experimental studies have shown that the **dhcp** phase of FeH is stable at low pressures (10–40 GPa) and undergoes phase transitions to the **hcp** and **fcc** phases at pressures of up to 80 and 100 GPa, respectively [2, 3].

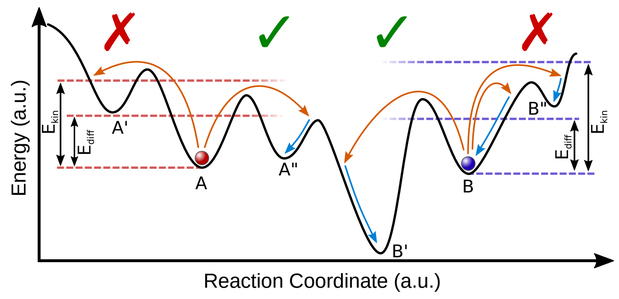


- Machine-learned interatomic potentials (ML-IAPs) enable modeling the potential energy surfaces (PESs) of large systems.
- New (meta)stable structures and phenomena can be discovered on larger length and time scales.
- **Here**: We conducted an extensive structure search of bulk FeH systems by globally sampling the PESs using a highly transferable ML-IAP over a pressure range of $0-100$ GPa.

2) Methods

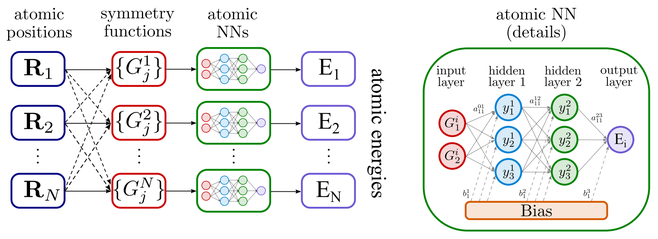
Global optimization:

- Minima hopping (MH) method to (systematically) explore the PESs [5]



High-Dimensional Neural Network (HDNN) Potential:

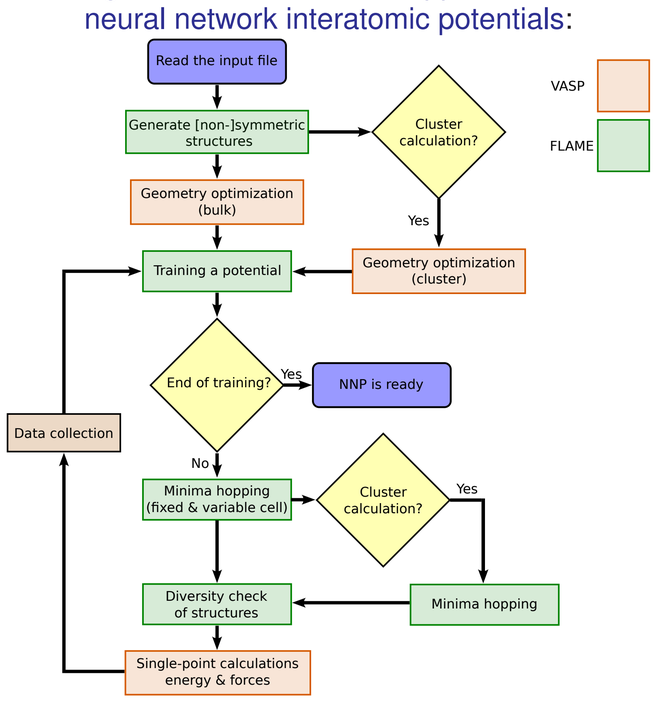
- Atom-centered (Behler) method [6]



$$E = \sum_{i=1}^{N} E_i$$

- Implementation:https://gitlab.com/flame-code/FLAME [7]
  - DFT calculations: VASP [8] with the PBE functional
  - Phonon dispersions: PHONOPY [9]

Workflow diagram of the automated approach for developing neural network interatomic potentials:

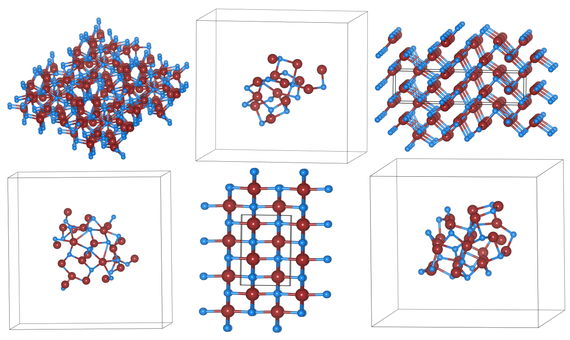


- Data generation and potential construction in an iterative process of training NN potential and crystal structure prediction
- Training process: six sets with different pressures $P = 0, 10, 20, 40, 50, 80$ GPa
- High transferability to systems with different boundary conditions and at a range of pressures $0-100$ GPa
- Implementation:https://gitlab.com/flame-code/PyFLAME [10]

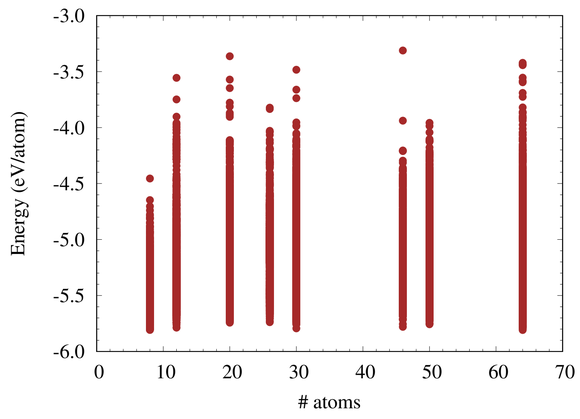
3) Neural network potential

Training:

- Train a HDNN interatomic potential with PyFLAME using a diverse dataset of atomic configurations



- Training data set: 33,338 clusters and crystalline structures with different sizes and symmetries
- Validation data set: 20% unseen structures from the training data set



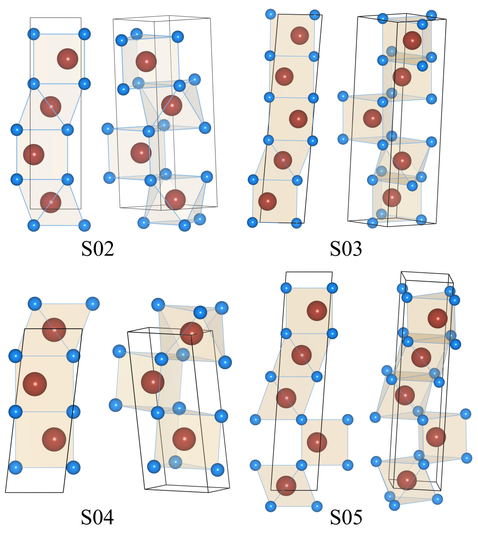
- NN architecture: $70-20-20-1$, 1861 trainable parameters
- Descriptors: 16 radial and 54 angular symmetry functions [4]
- RMSEs of energy and atomic forces: 30 meV/atom, 0.308 eV/Å

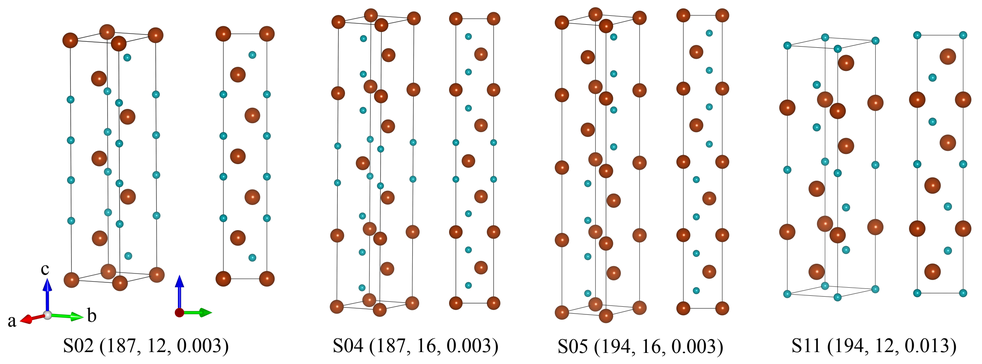
Validation:

- Geometry optimization
- Global optimization using MH to screen the PESs of crystal structures of FeH of various sizes and pressures

4) Structural search

- A systematic search on the PESs of FeH with simulation cells up to 18 f.u. at pressures $0-100$ GPa (in steps of 10 GPa)
- Refining the results at the level of DFT (energy and space group)
- All known structures in databases are found. We also found a dense spectrum of low-enthalpy polymorphs (<30 meV/atom) for stoichiometric FeH.

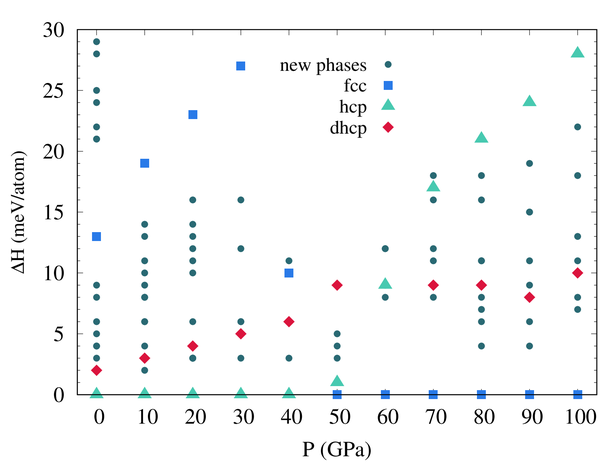




- Some of the new low-enthalpy polymorphs of FeH, new modifications and stacking of the known **dhcp**, **hcp**, and **fcc** structures, at $P = 20$ GPa
- Motifs: MgO_6 octahedra and trigonal prisms
- Connectivity of the octahedra: edge-sharing and face-sharing
- These motifs are stacked on top of each other in various sequences and directions (*AB, ABC, ABCD,* ...).

5) Phase diagram

- Without corrections of zero-point vibrations of H atoms!
- The relative enthalpy shows the distance from the convex hull as a function of P.



Conclusion and outlook

- We constructed a highly transferable HDNN potential for exploring the energy landscape of iron hydride across a range of pressures ($0-100$ GPa).
- We investigate the phase diagram of iron hydride based on large-scale structure prediction using the ML-IAP.
- We find a dense spectrum of novel low enthalpy polymorphs across the considered pressure range.
- Free energy calculations could change the energy order of structures and phase transitions.
- We can now investigate the phase diagram and PESs of iron superhydrides FeH_n ($n \geq 3$) which exhibit special electrical properties such as superconductivity.

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