1) Motivation

- Understanding the chemical composition of Earth's core is a grand challenge in geosciences and materials science.
- Earth's inner core is believed to be composed of iron-based alloys; however, its density is 2.5% lower than that 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].

2) Methods

- Minima hopping (MH) method to (systematically) explore the PESs [5]
- DFT calculations: VASP [8] with the PBE functional
- Phonon dispersions: PHONOPY [9]
- Training process: six sets with different pressures
- Data generation and potential construction in an iterative process of training NN potential and crystal structure prediction
- Training process: six sets with different pressures
- 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, 0.36eV/Å
- 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, ABD, . . .).

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.

References