Structure prediction of iron hydrides across pressure range with transferable machine-learned interatomic potential

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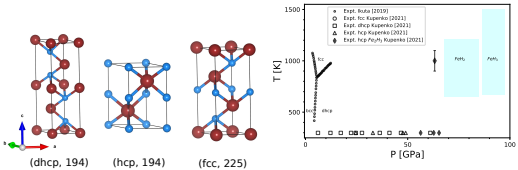
Contents

Background and 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.
- 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.



Goal of this work

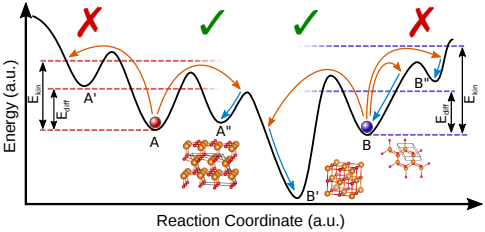
- Machine-learned interatomic potentials (ML-IAPs) enable modeling the potential energy surfaces (PESs) of large systems.
- New (meta)stable structures and phenomena can be discovered on larger length and time scales.
- We conducted an extensive structure search of bulk FeH systems by globally sampling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

Contents

Methods

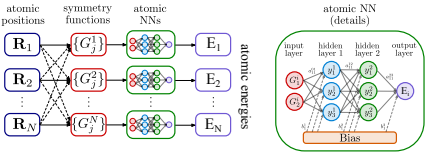
Global optimization:

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



High-Dimensional Neural Network (HDNN) Potential:

- Atom-centered (Behler) method



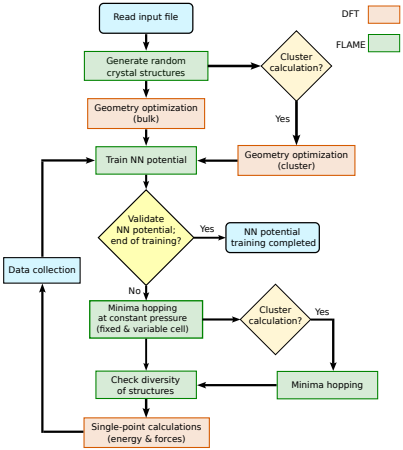
$$E = \sum_{i=1}^{N} E_i$$

Implementation: https://gitlab.com/flame-code/FLAME

- DFT calculations: VASP with the PBE functional
- Transport properties: RT-TDDFT, the ELk code
- Phonon dispersions: PHONOPY

Methods

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

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 (CPU time: $\approx 5 \times 10^5$ core-hours)
- 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
- RMSEs of energy and atomic forces: 30 meV/atom, 0.308 eV/Å

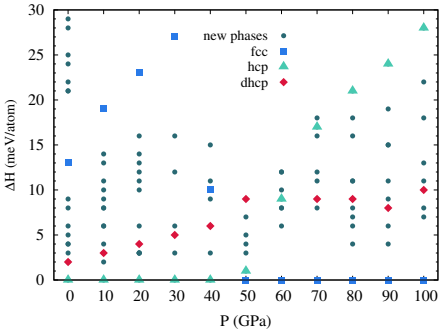
Validation:

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

Contents

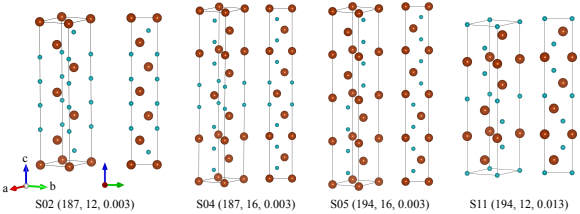
Phase diagram

- 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)
- Minima hopping runs to search on the PESs: 32 runs starting from random structures ($\approx 10^5$ core-hours)
- Refining the results at the level of DFT (energy and space group, phonon) ($\approx 10^6$ core-hours)
- All known structures in databases are found. We also found a dense spectrum of low-enthalpy polymorphs ($<$30 meV/atom) for stoichiometric FeH.
- The relative enthalpy shows the distance from the convex hull as a function of *P*.



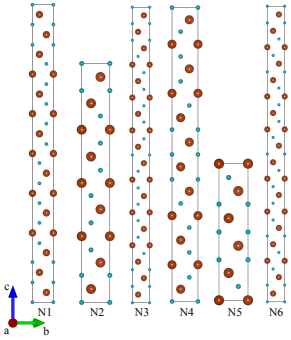
H. Tahmasbi *et al.*, Phys. Rev. Materials (in press) arXiv:2311.06010

New structures



- Some of the newly low-enthalpy modifications of FeH that crystallize in the hexagonal crystal system.
- Motifs same as other binary materials: FeH_6 octahedra and trigonal prisms

The presence of coexisting regions containing known FeH structures! (in agreement with earlier speculations in prior experimental research)

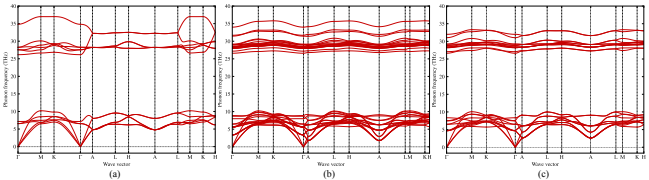


- Low-enthalpy phases N1–N6 are new modifications or stacking faults of FeH that crystallizes in the trigonal crystal system.
- Some of these phases found across all pressures

Phonon dispersions

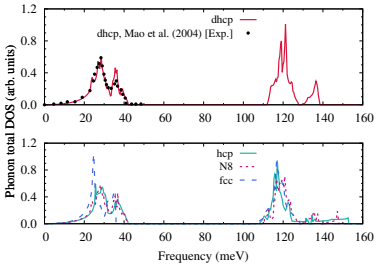
To assess the dynamical stability and compute thermal properties:

- Phonon dispersions of the low energy structures (a) hcp, (b) N8, and (c) dhcp phases at P = 20 GPa



Thermal properties at constant pressure using the quasi-harmonic approximation (QHA):

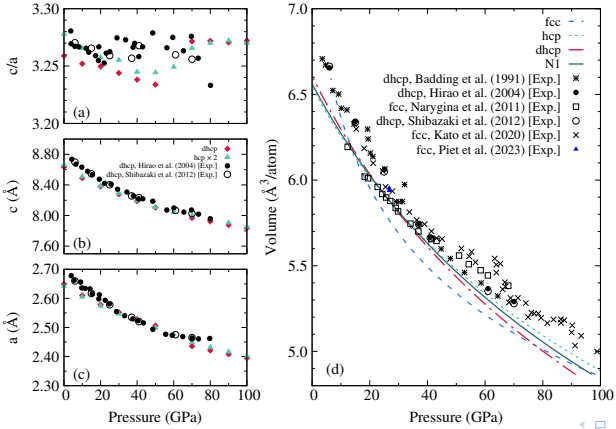
- To minimize the Gibbs free energy and to include the zero-point energy corrections
- The energetic ordering of the structures remain almost unaffected contrasting with the literature



Comparison of the phonon DOS of the four phases of FeH at 20 GPa with the experimental data.

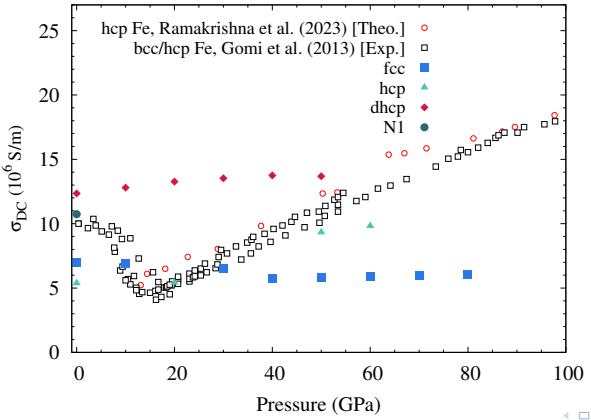
Elastic and Thermodynamic Properties

Lattice parameters and Equation of state:



Electronic Transport Properties

DC conductivity as a function of pressure:



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Summary 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.
- 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.

Machine Learning-Driven Structure Prediction for Iron Hydrides,
H. Tahmasbi, K. Ramakrishna, M. Lokamani, and A. Cangi, Phys. Rev. Materials (in press)
arXiv:2311.06010

References and thanks

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- PyFLAME code: H. Mirhosseini *et al.*, Comput. Mater. Sci. 197, 110567 (2021)
- Atom-based NN approach: J. Behler *et al.*, PRL 98, 146401 (2007)
- Minima hopping method: M. Amsler *et al.*, JCP, 133, 224104 (2010)

- **With thanks to:**
  - Attila Cangi (CASUS/HZDR)
  - Kushal Ramakrishna (CASUS/HZDR)
  - Mani Lokamani (HZDR)

Elastic and Thermodynamic Properties

Bulk modulus:

Comparison of volume and bulk modulus for four FeH phases at P= 10 GPa and $T = 300$ K

Phase	V_0 (Å^3/f.u.)	K_0 (GPa)	K_0'	Reference
hcp	12.857	207.315	5.207	This work
	12.73	246	4.3	Theo. Elssser1998
N1	12.864	207.887	5.295	This work
dhcp	12.896	203.362	5.341	This work
	12.69	248	4.3	Theo. Elssser1998
	13.9±0.125	121±19	5.31±0.9	Exp. Badding1991
	13.875±0.5	147±6	4	Exp. Hirao2004
	12.587	227.2	4.8	Theo. Pepin2014
	13.901	131.1±3	4.83	Exp. Pepin2014
fcc	13.026	195.541	3.957	This work
	12.69	248	4.3	Theo. Elssser1998

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