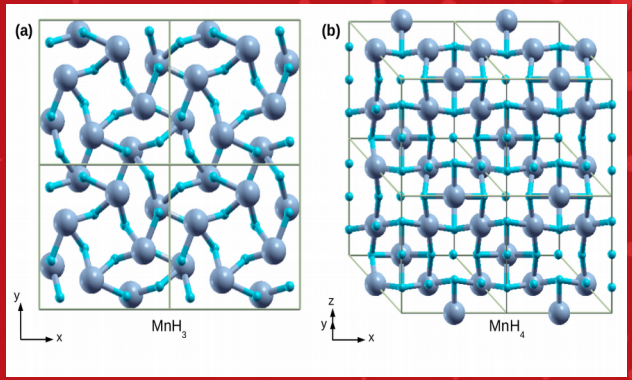
AB-INITIO AND MACHINE LEARNING DRIVEN RESEARCH OF SUPERHYDRIDES

DE LA RECHERCHE À L'INDUSTRIE

INTERNATIONAL ABINIT WORKSHOP JUNE-2021



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LMCE, Université Paris Saclay

OUTLINE

- Challenges with Superhydrides
- Studied Systems
- Machine Learning Enhanced Structure Searching

CHALLENGES WITH SUPERHYDRIDES

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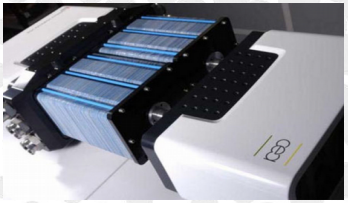
CHALLENGES WITH SUPERHYDRIDES

Hydrides

- Hydrogen enriched alloys.
- Applications for fuel cells (stationary applications).



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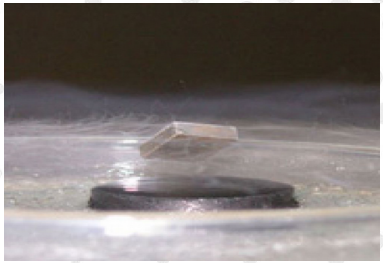
Superhydrides

High hydrogen concentration (H/M ≈ 5)
Promising materials for the Energy sector:

- High density hydrogen storage system.
- High temperature superconductor.



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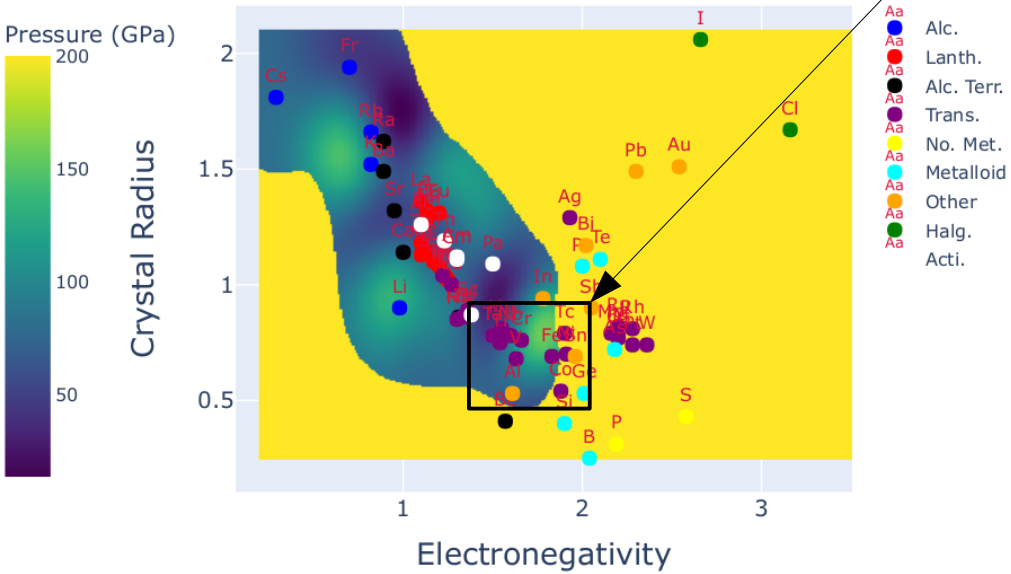
Aim

Pressure synthesis remains too high for industrial applications (range 100 GPa).

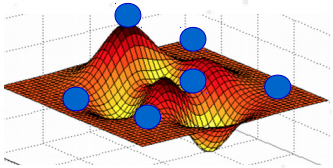
Binary System Studies

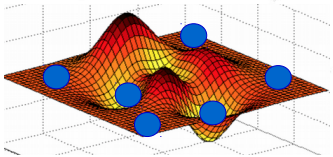
- Not all the elements can be mixed with hydrogen to form superhydrides.
- For binary systems, formation pressure remains high (between 40 and 150 GPa).

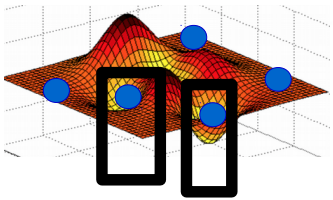
Focus on transition metal type superhydrides



RANDOM SEARCHING METHOD







Sampling Method

Structure Generation

Structural Optimization

- **DFT (AIRSS)**

  Ab Initio Random Structure Searching (AIRSS) [1]

- **Machine Learning**

  Numerical Potentials

Structure Selection

- Enthalpy criteria
- Dynamical stability
- Comparison with experimental X-ray spectrum

[1] C. J. Pickard, R. J. Needs, Journal of Physics-Condensed Matter, **23**, 053201, (2011)

SYSTEMS STUDIED WITH AIRSS

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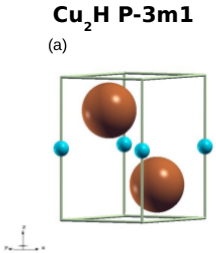
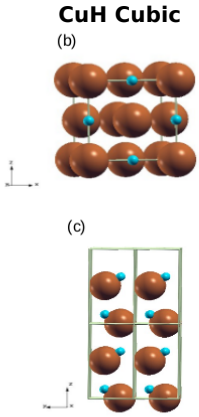
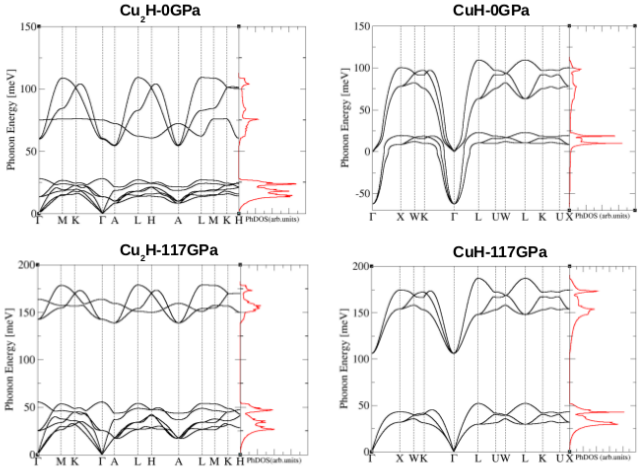
No Superhydride Predicted

- CuH up to 150 GPa.
- CuH cubic stabilized dynamically with pressure.
- Retrieve experimental results [1].





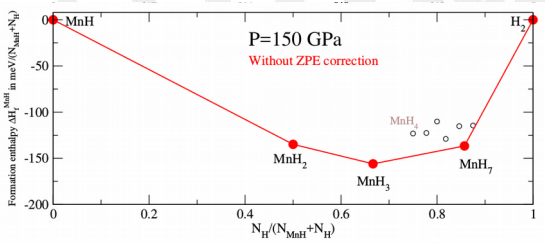


[1] T. Meier, F. Trybel, G. Criniti, D. Laniel, S. Khandarkhaeva, E. Koemets, T. Fedotenko, K. Glazyrin, M. Hanfland, G. Steinle-Neumann, N. Dubrovinskaia, and L. Dubrovinsky, Phys. Rev. B **102**, 165109 (2020)

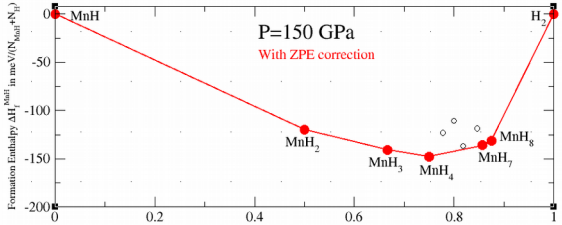
Superhydrides Predicted [1]

- New structures : MnH_2, MnH_3, MnH_4, MnH_7, MnH_8.
- Verify dynamical stability.
- At 150 GPa, Zero Point Energy (ZPE) computation
  => Stabilize MnH_4 and MnH_8

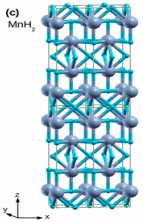
Without ZPE



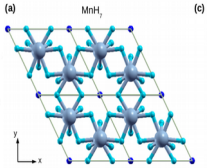
With ZPE



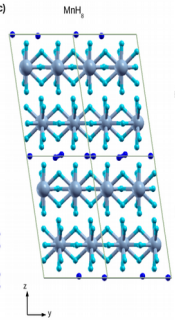
MnH_2 I4/mmm



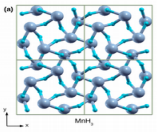
MnH_7 P6/mmm



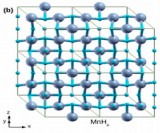
MnH_8 P-1



MnH_3 I4/m



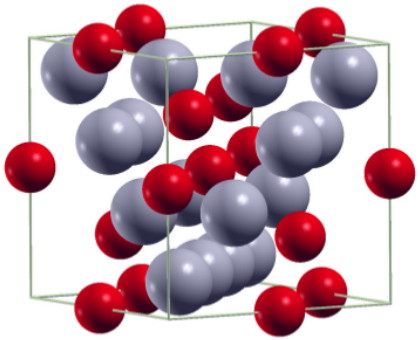
MnH_4 R32



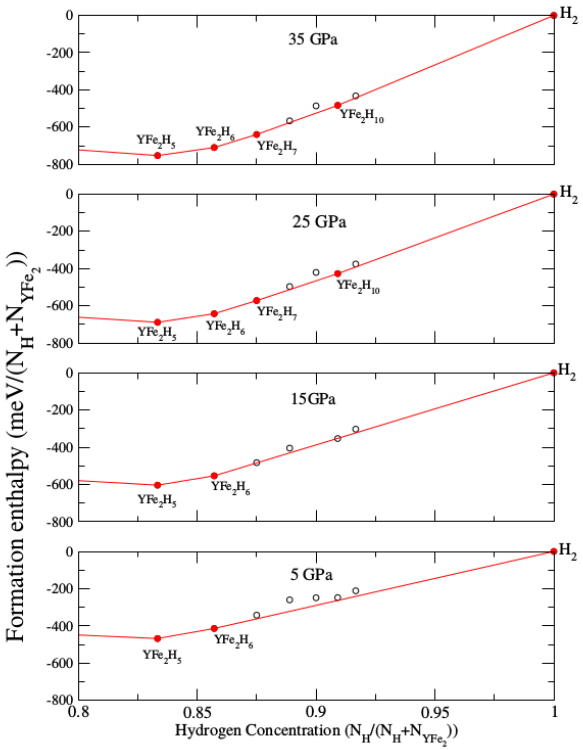
[1] J.-B Charraud, G. Geneste and M. Torrent, Phys. Rev. B **100**, 224102 (2019)

Interstitial Hydrides YFe_2H_x

- Use experimental results at low pressure [1].
- Hydrogens occupy interstitial site of a distorted YFe_2 cubic structure.
- High hydrogen concentration reached at relatively low pressure (YFe_2H_{10}, 25 GPa).



YFe_2 Cubic Sublattice



[1] V. Paul-Boncour and S. Matar, Phys. Rev. B **70**, 184435 (2004)

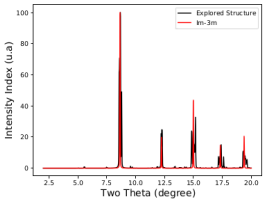
YTTRIUM SUPERHYDRIDES

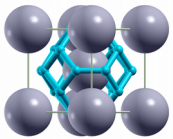
Superhydrides Predicted

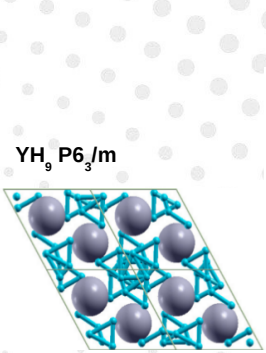
- YH_7 near 100 GPa.
- YH_6 [1], YH_9, YH_{12} near 180 GPa.

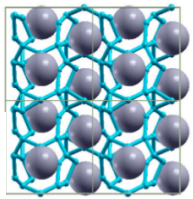
Problem at 150 GPa

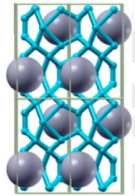
- Experimental X-ray spectrum does not match those of the predicted structures (without superposition).
- Probable more complex structure unreachable with AIRSS.

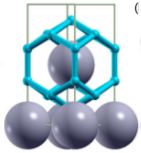


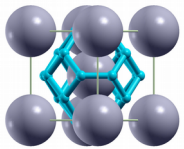












[1] H. Liu, I. I. Naumov, R. Hoffmann, N. W. Ashcroft, and R. J. Hemley, PNAS **114**, 6990-6995 (2017)

[2] F. Peng, Y. Sun, C. J. Pickard, R. J. Needs, Q. Wu, and Yanming Ma, Phys. Rev. Lett. **119**, 107001 (2017)

[3] I. A. Troyan et al., Adv. Mater., **33**, 2006832 (2021)

AIRSS'S LIMITS

High Dimensional Surface

- 3N+6 dimensions for a given atom number N.
- Exponential increase of the local minima number.

With Full DFT

- 99 % of the computational time due to Ab-Initio optimization.
- A few dozen of millions CPU hours to predict a binary material with up to 30 atoms in the primitive cell.
- Method reaches its limits to predict complex binary systems (YH_x) or ternary systems.

Use Machine Learning method to speed up the search an reach more complex system's prediction.

MACHINE LEARNING ENHANCED STRUCTURE SEARCHING

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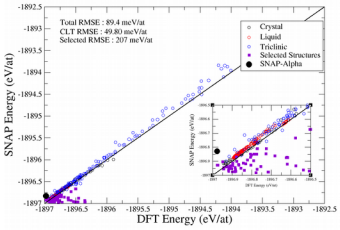
NUMERICAL POTENTIAL

Training Database

- Atomic configurations
- Properties computed by DFT

Descriptor Model

Catch atomic environments' informations
- Bispectrum [1]
- Symmetry functions [2]



Statistical Model

- Spectral Neighbourg Analysis Potential (SNAP) [1]
- Neural Networks [2]

Physical Quantities

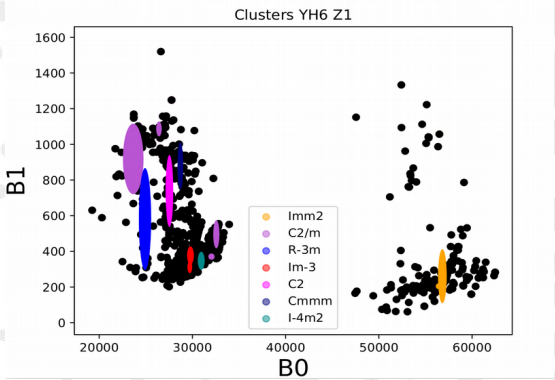
[1] A.P Thompson, JCP **285,** 316-330 (2015)

[2] Jörg Behler, JCP **134,** 074106 (2011)

ANALYSIS TOOLS

Enthalpy Comparisons

- Take the structures having the lowest predicted enthalpies.

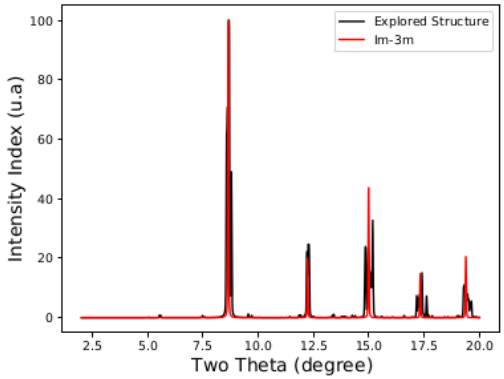


Bispectrum Analysis

- Describe atomic environments with vectors (bispectrum).
- Clustering (Gaussian Mixtures).

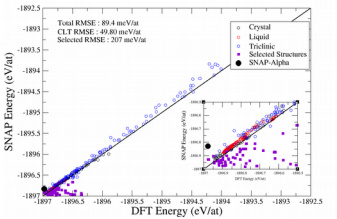
X-Ray Spectrum

- Compute the X-ray spectrum of the predicted structures.
- Compare with a reference one and a metric.



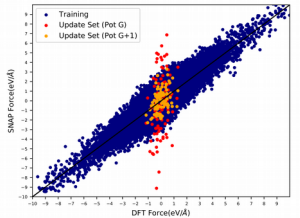
ACTIVE LEARNING PROCESS

Potential's Initialization

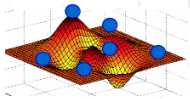


Active Learning

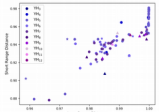
Potential's Update



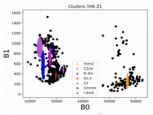
Random Searching



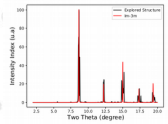
Selection



Bispectrum Clustering



X-Ray Spectrum



Single Point DFT

ABINIT

DFT Optimization

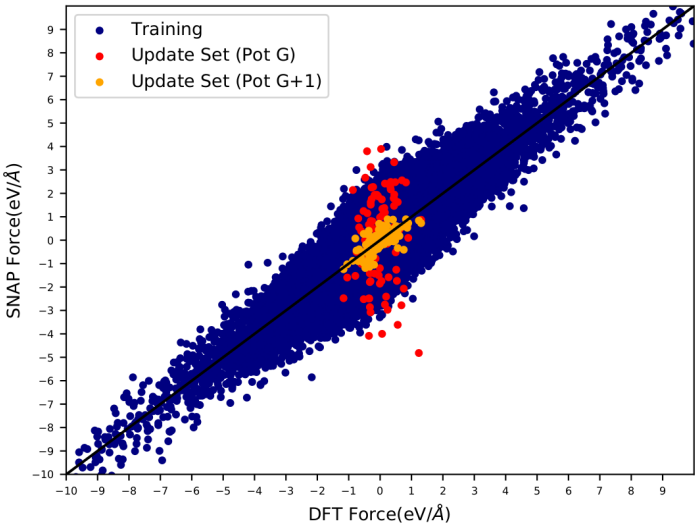
ABINIT

ML POTENTIAL UPDATES

Update Process

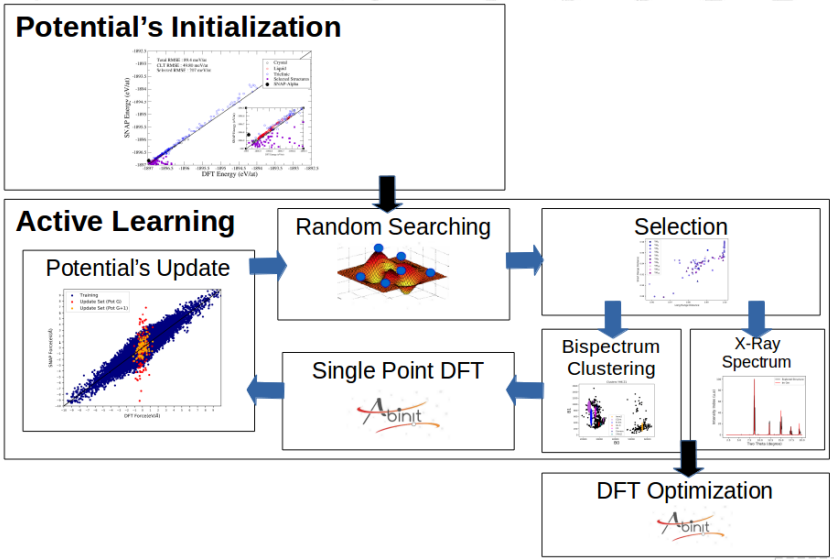
- Pot G trained on an initial potential (blue) and explores structures.
- Pot G evaluated but not trained on selected structures (red).
- Pot G+1 evaluated on the selected structures included in its training database (yellow).

Force Correlations



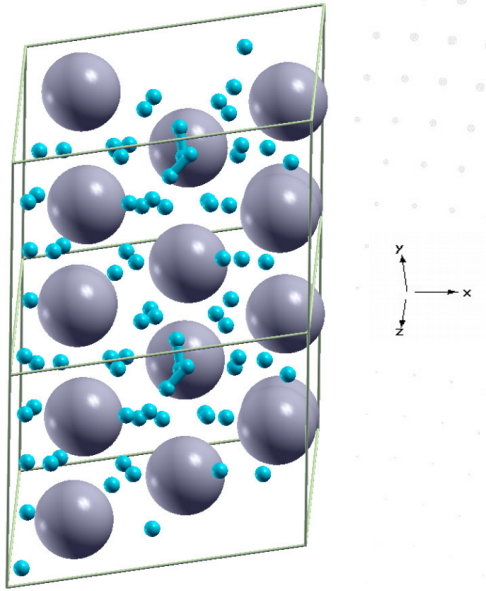
Structure Exploration

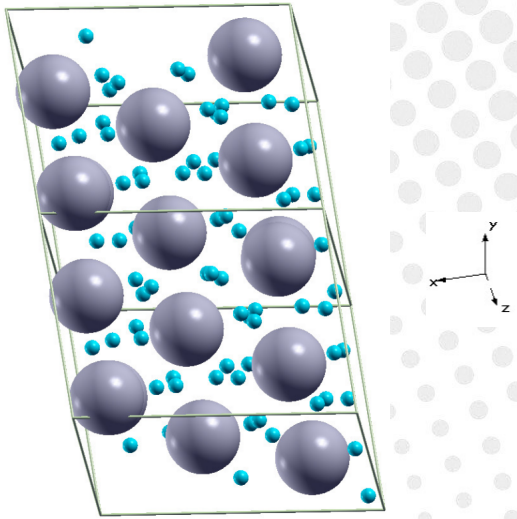
- 1.6 Millions of structures explored (YH_4-YH_7).
- Around 10 000 CPU hours, would require more than 10^9 with AIRSS.
- SNAP and Neural Network potentials.



NUMERICAL POTENTIAL RESULTS

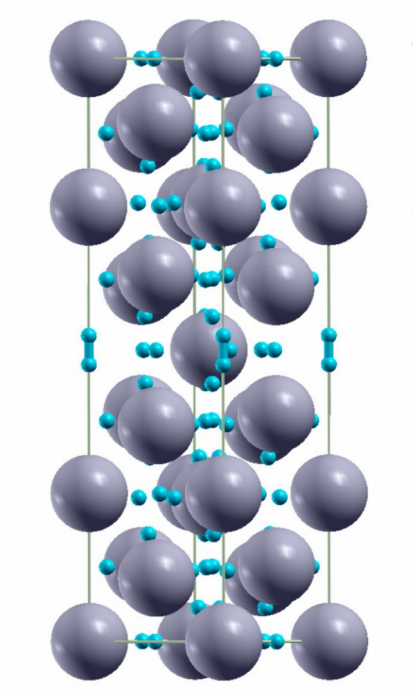
YH_5 P1 (48 atoms, NN Potential)

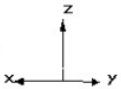


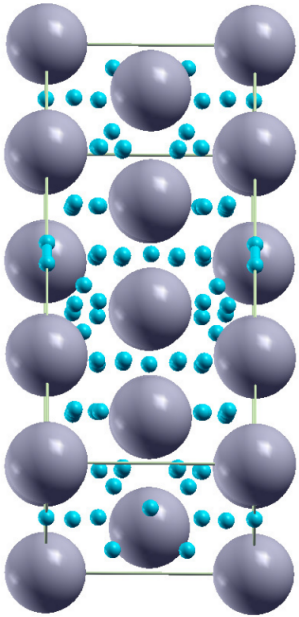


NUMERICAL POTENTIAL RESULTS

YH_5 Immm (42 atoms, SNAP Potential)







CONCLUSION

With AIRSS

- Contribution to the transition metal superhydrides' research.
- Copper, not interesting to form superhydrides.
- New MnH_x superhydrides predicted with relatively low pressure synthesis.
- High potentialities for hydrogen storage with YFe_2H_x.

AIRSS and Machine Learning

- Can now explore several millions of structures against a few thousands with AIRSS.
- YH_x: More complex phases identified with symmetry, unreachable with AIRSS. But don't yet explain the experimental X-ray spectrum.
- Software AI²RSS under preparation for a github repository.

CONCLUSION

ML Approach's Forces

- More complex structure explorations possible.
- Reduce by a factor 10^5 the computational cost.

Ways of Improvements

- Improvements in the structure generation method.
- Use directly X-ray spectrum and probabilistic neural networks (VAE).

Thank you for your attention