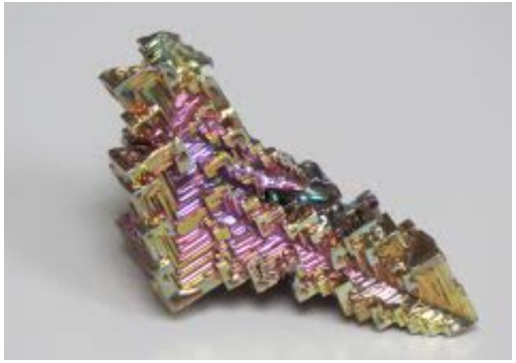
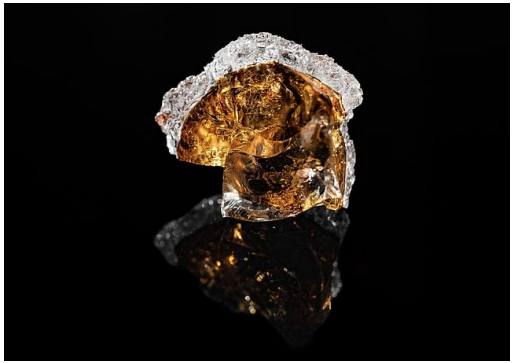
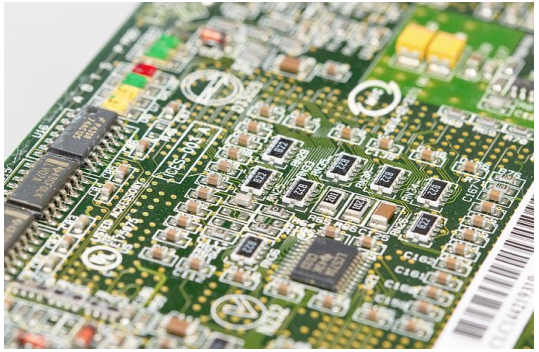
Crystal structure search accelerated by neural networks

Stefaan Hessmann, Niklas W. A. Gebauer, Kristof T. Schütt, Michael Gastegger, Klaus-Robert Müller, Tamio Oguchi, Tomoki Yamashita

Motivation: search for stable structures







Overview

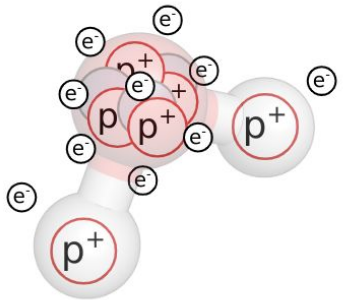
1. Quantum chemistry computations
2. Message passing neural networks
3. Active learning for crystal structure prediction
4. Results

Quantum chemistry computations

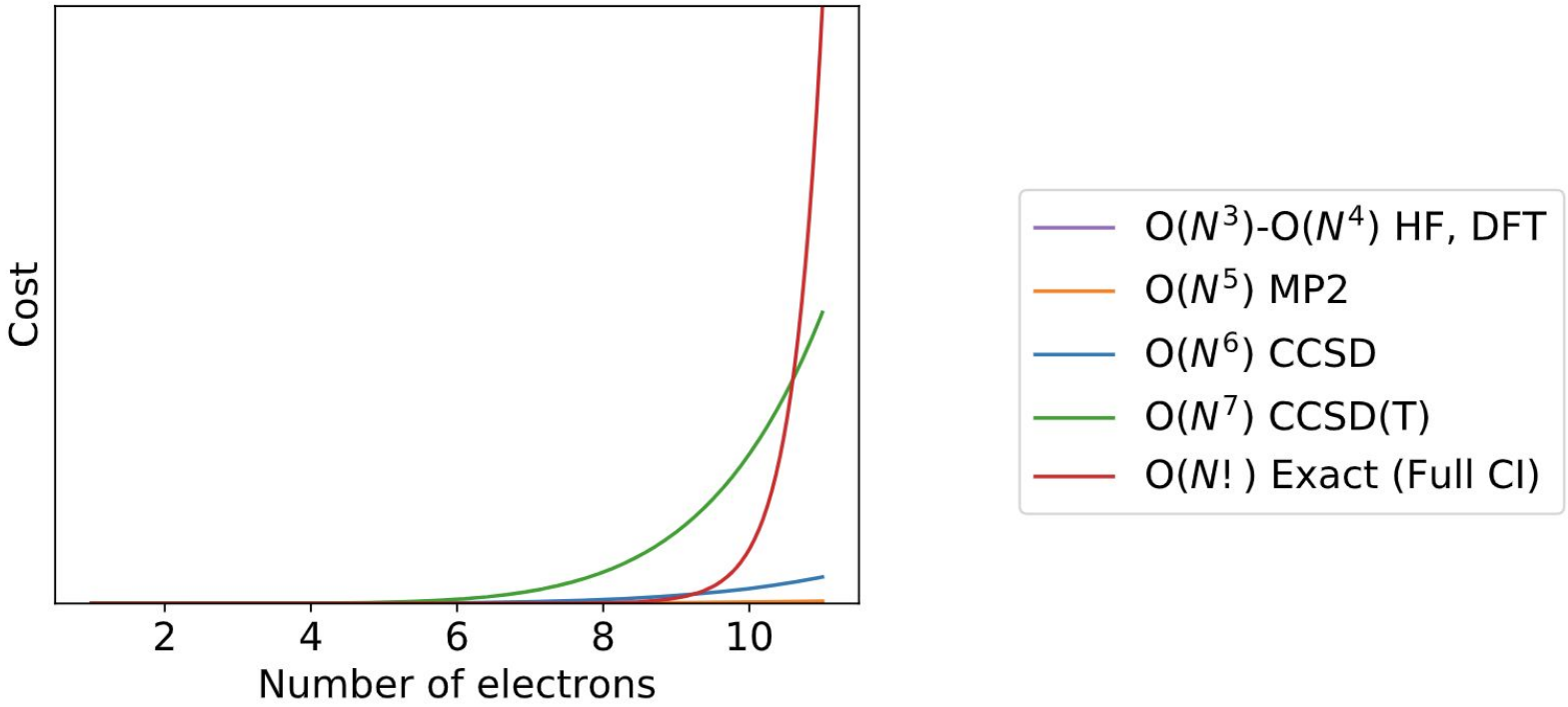
Quantum chemistry computations

$$\hat{H}_{\text{el}}(\overline{\mathbf{R}}, \mathbf{r}, \mathbf{Z})\Psi_{\text{el}}(\overline{\mathbf{R}}, \mathbf{r}) = E_{\text{el}}\Psi_{\text{el}}(\overline{\mathbf{R}}, \mathbf{r})$$

$$\hat{H}_{\text{el}}(\overline{\mathbf{R}}, \mathbf{r}, \mathbf{Z}) = \underbrace{-\sum_{i}^{N} \frac{1}{2}\nabla_i^2}_{\text{electrons}} \underbrace{-\sum_{i}^{N}\sum_{A}^{M} \frac{Z_A}{|r_i - \overline{R}_A|}}_{\text{electrons-nuclei}} + \underbrace{\sum_{i}^{N}\sum_{j>i}^{N} \frac{1}{|r_i - r_j|}}_{\text{electrons-electrons}}$$



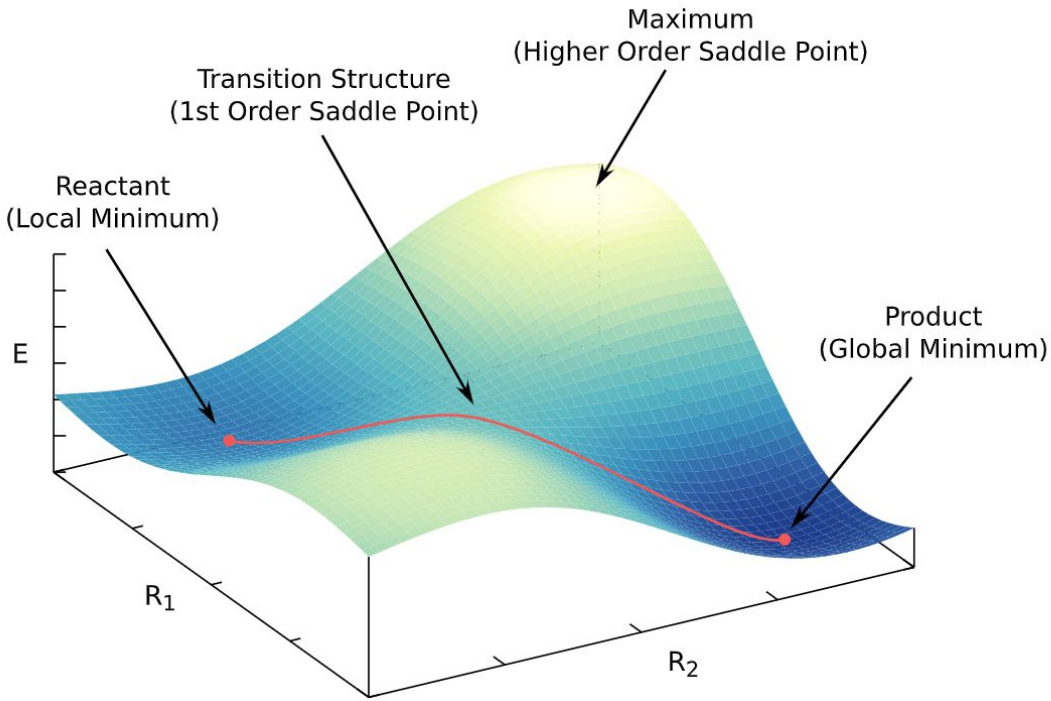
Quantum chemistry computations



Quantum chemistry computations

$$\hat{H}_{\text{el}}(\overline{\mathbf{R}}, \mathbf{r}, \mathbf{Z})\Psi_{\text{el}}(\overline{\mathbf{R}}, \mathbf{r}) = E_{\text{el}}\Psi_{\text{el}}(\overline{\mathbf{R}}, \mathbf{r})$$

$$E_{el} \approx ML(\overline{\mathbf{R}}, \mathbf{Z})$$



Message passing neural networks

Message passing neural networks

Message passing neural network [Gilmer et al 2017]

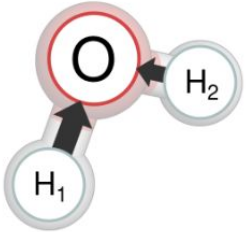
$$\mathbf{m}_i^{t+1} = \sum_{j \in \mathcal{N}(i)} \mathbf{M}_t(\mathbf{s}_i^t, \mathbf{s}_j^t, \|\vec{r}_{ij}\|)$$

$$\mathbf{s}_i^{t+1} = \mathbf{U}_t(\mathbf{s}_i^t, \mathbf{m}_i^{t+1})$$

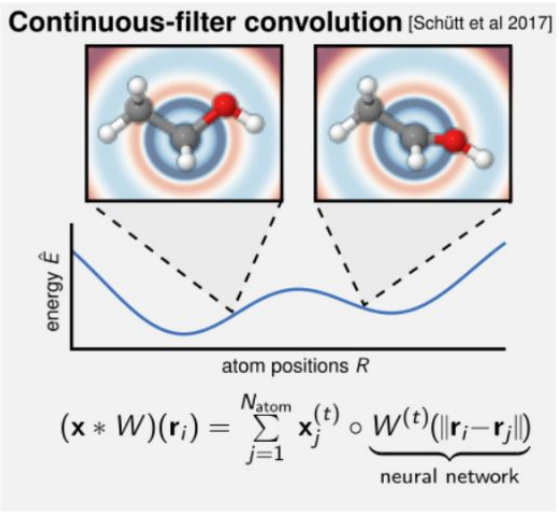
Rotational invariance:

$$\mathbf{M}(\vec{x}) = \mathbf{M}(R\,\vec{x})$$

for any rotation R.



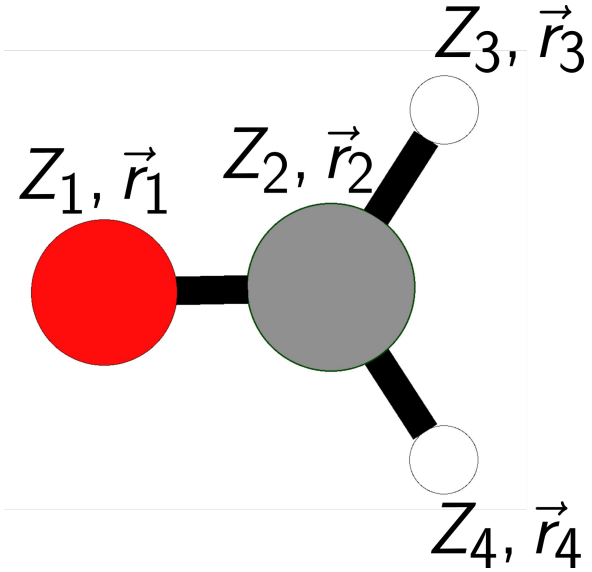
Continuous-filter convolution [Schütt et al 2017]



Schütt et al. (2017) Quantum-chemical insights from deep tensor neural networks. Nat. Commun. 8: 13890
Gilmer et al. (2017) Neural Message Passing for Quantum Chemistry. ICML
Schütt et al. (2017) SchNet: A continuous-filter convolutional neural network for modeling quantum interactions. NeurIPS

Message passing neural networks

atom types Z_i, positions $\vec{r}_i$



Message passing neural networks

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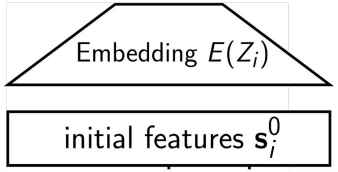
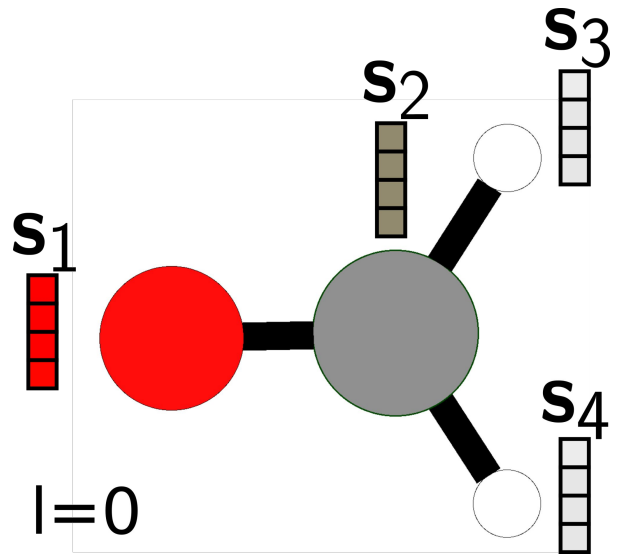
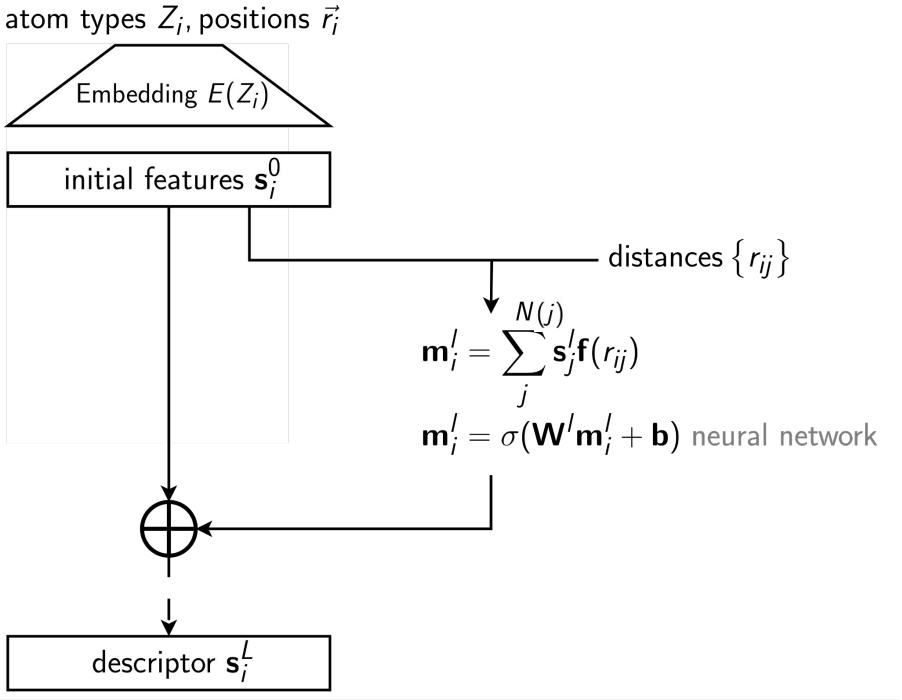
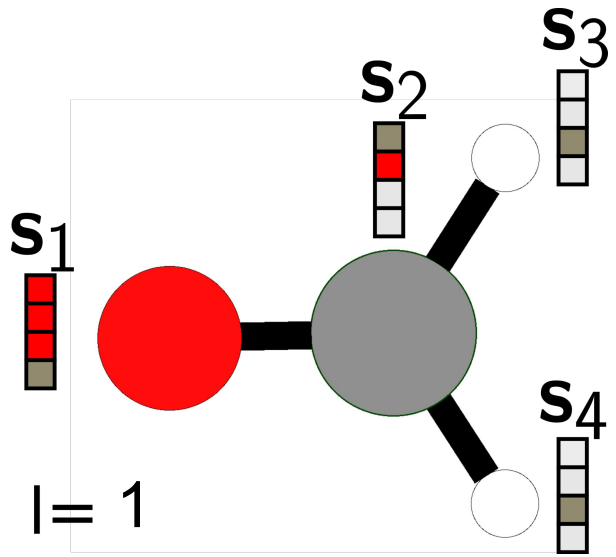


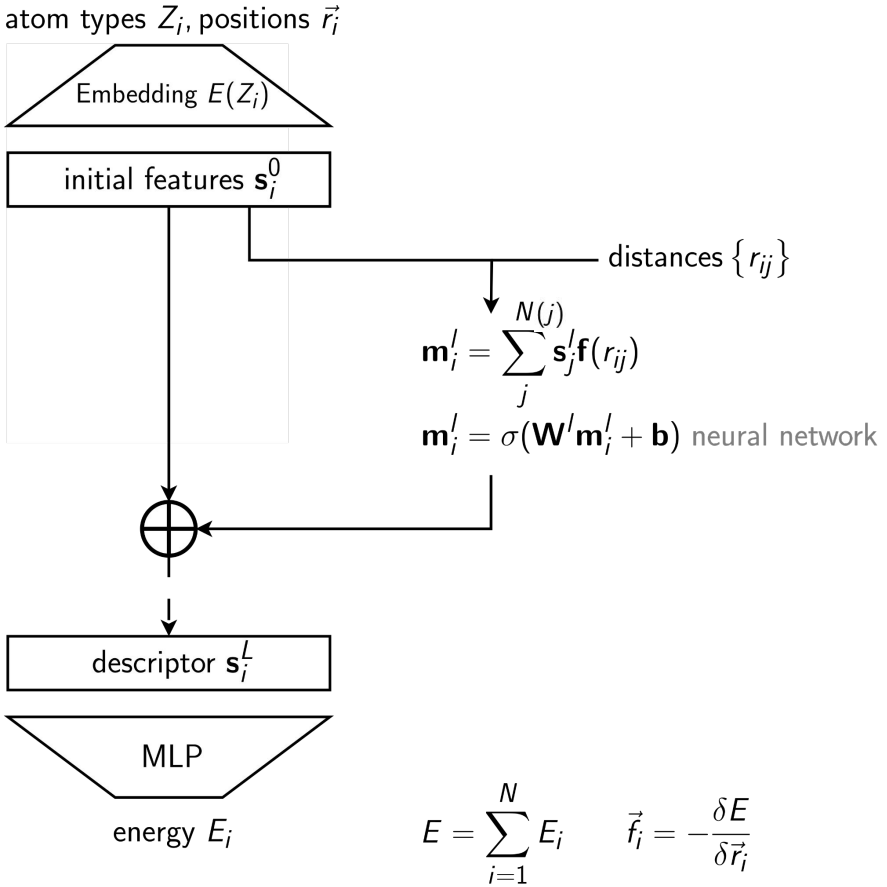


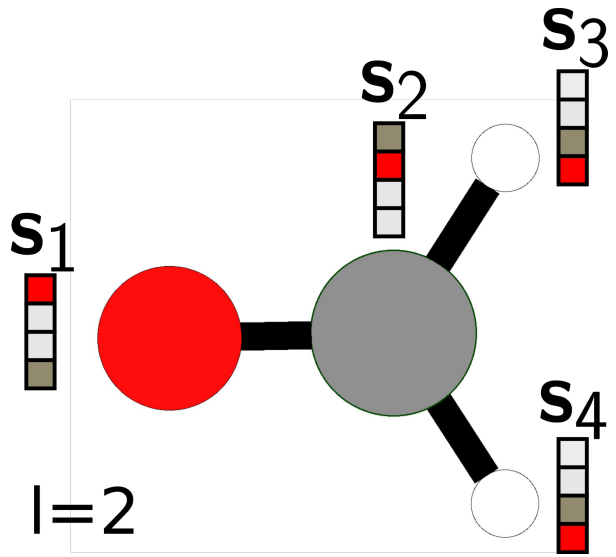
Message passing neural networks





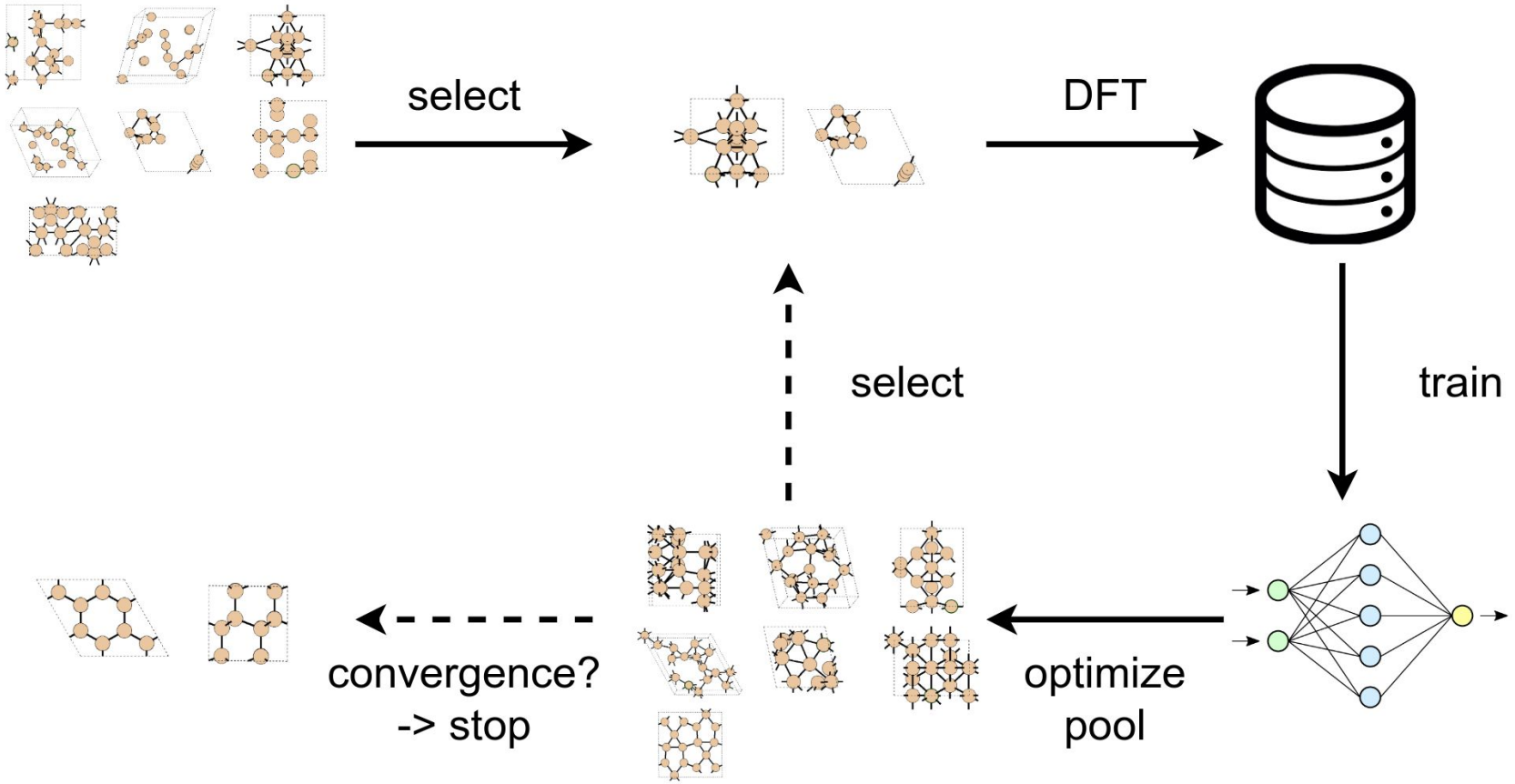
Message passing neural networks



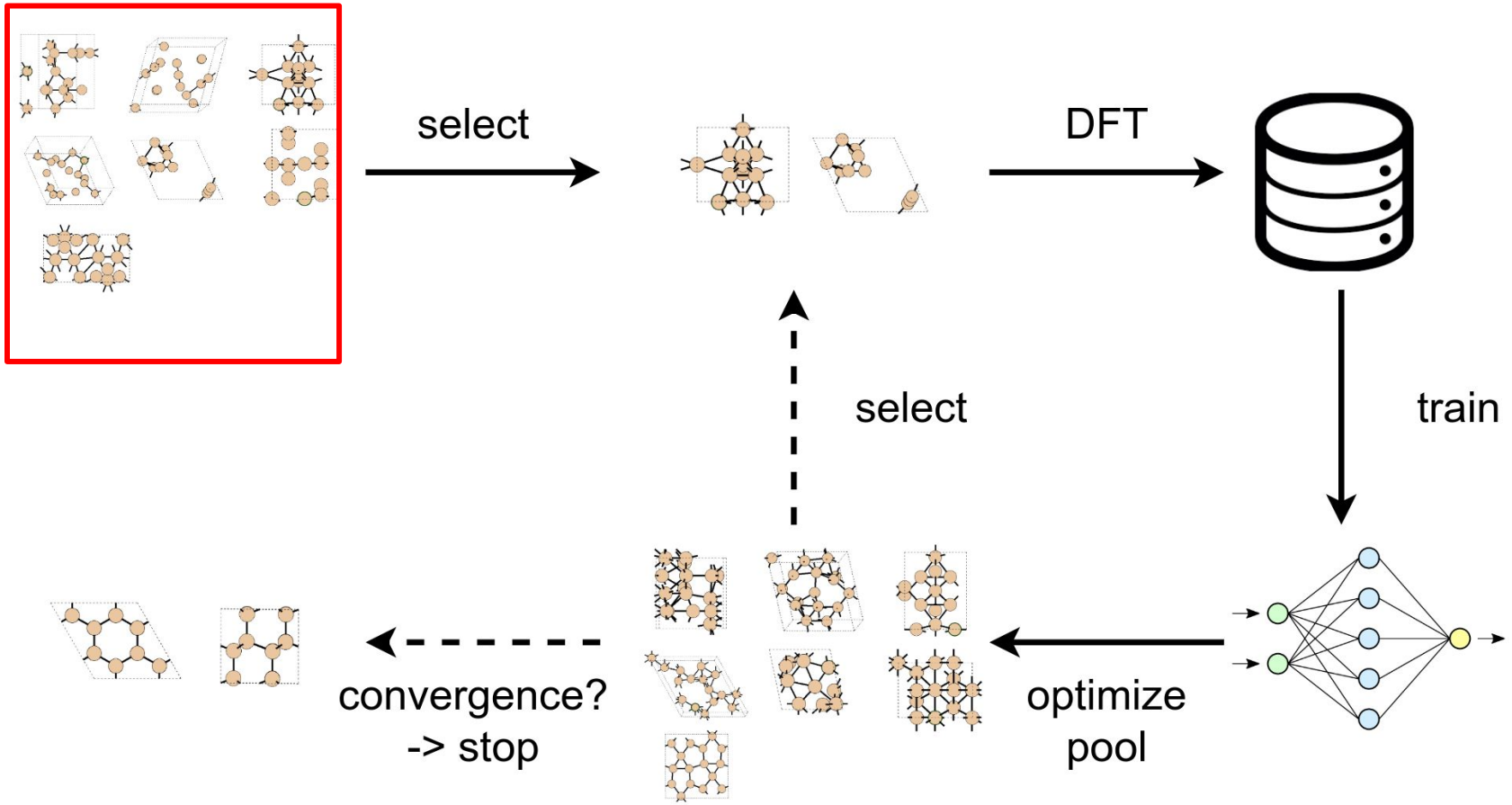


Active learning for crystal structure prediction

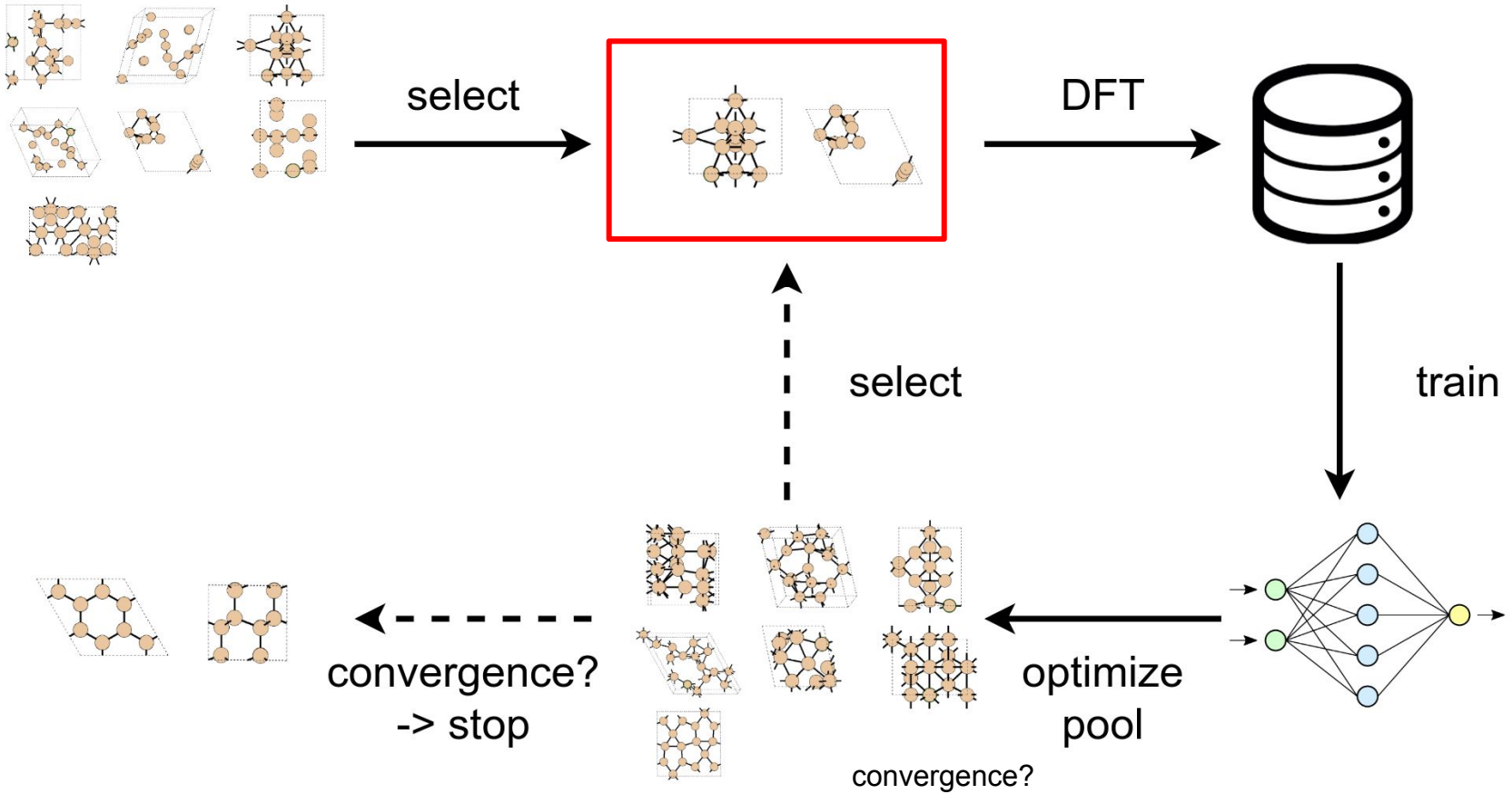
Method overview



Method overview



Method overview



Selecting candidates for DFT evaluation

- scoring function

$$L_{i,\theta} = \overline{E}_{i,\theta} - \omega_F \max(||\overline{F}_{i,\theta}||_2)$$

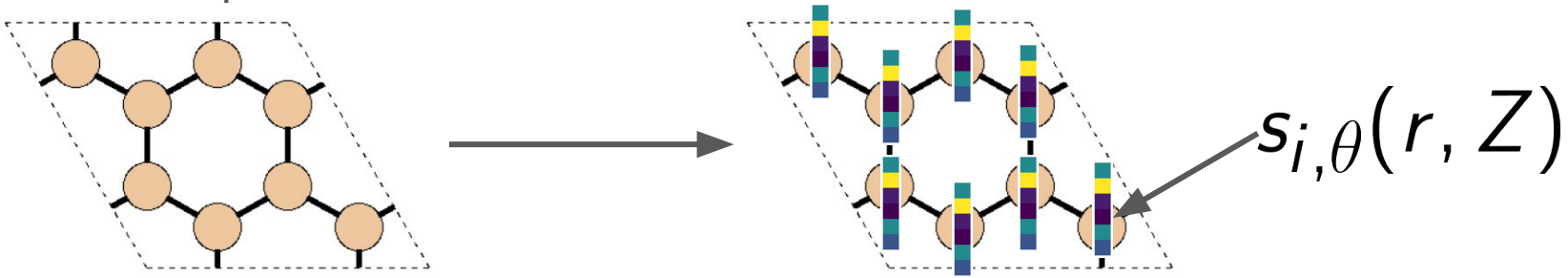
- next candidates

$$\{\arg\min_x(L_\theta(x))\}_n$$

- problem: similar structures yield similar scores
- solution: clustering

Global representation for clustering

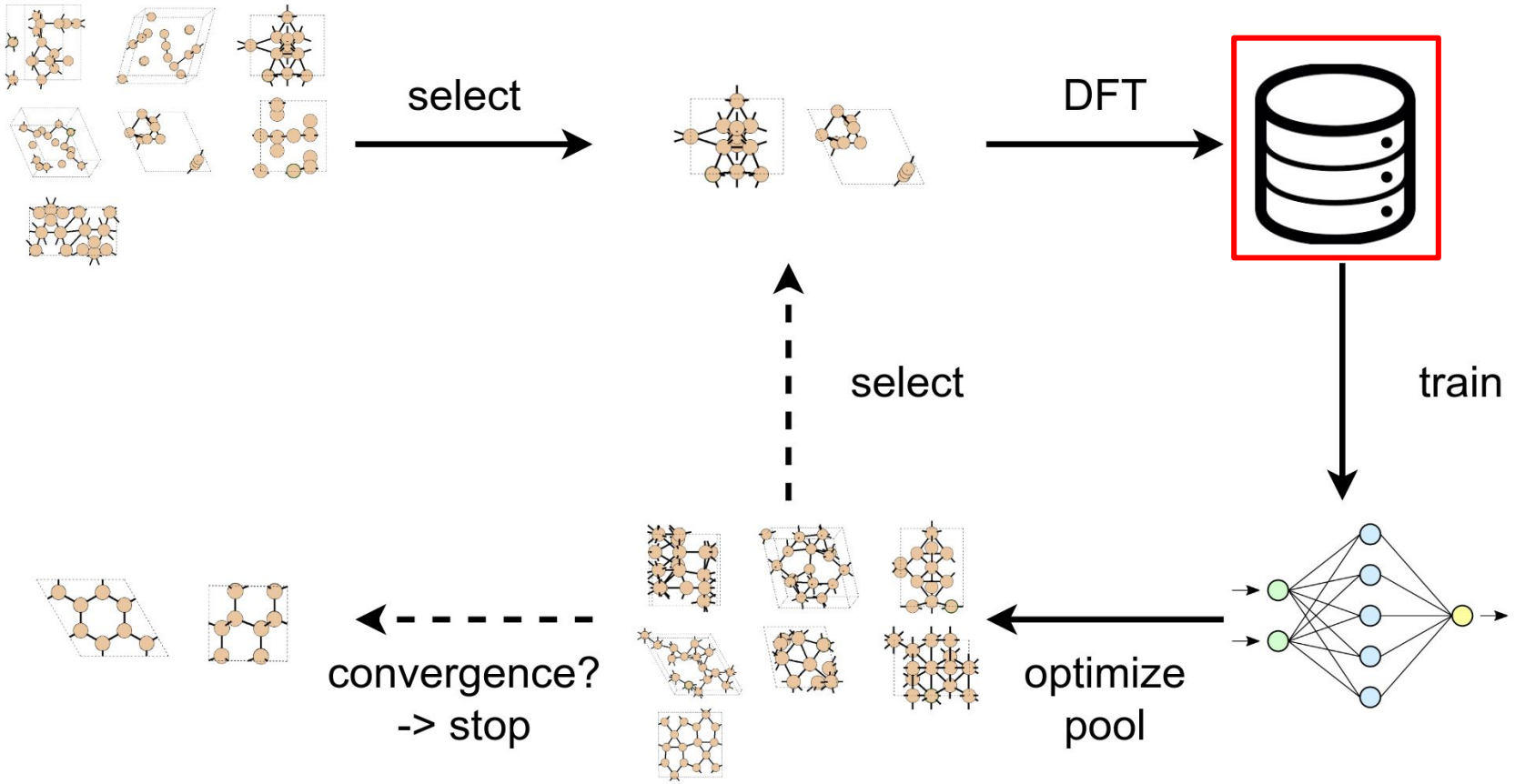
- use local representations



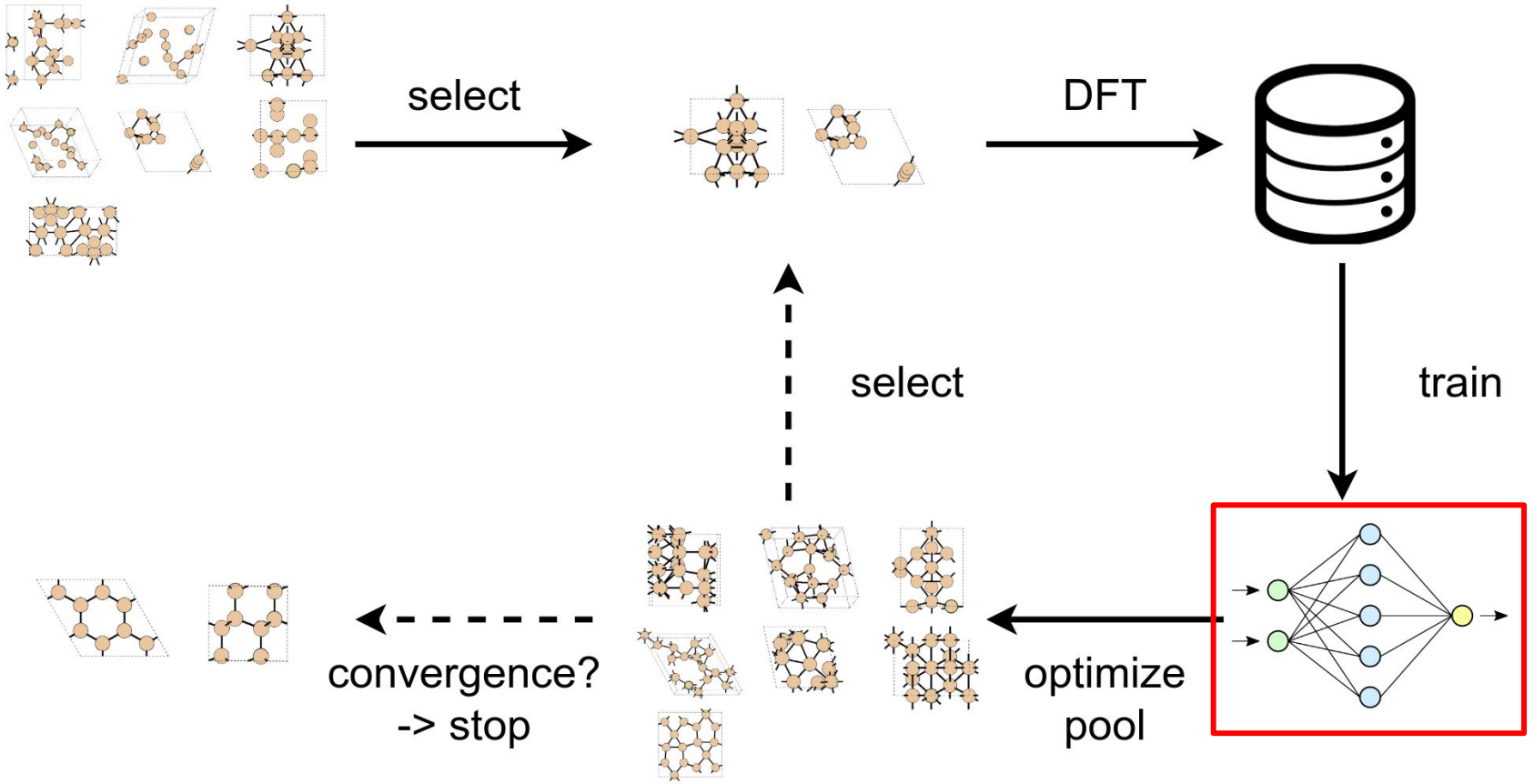
- pooling introduces invariance against permutation

$$\overline{s}_\theta = \sum_{i=0}^{n} s_{i,\theta}(r, Z) \qquad \overline{s}_\theta \epsilon \mathbb{R}^f$$

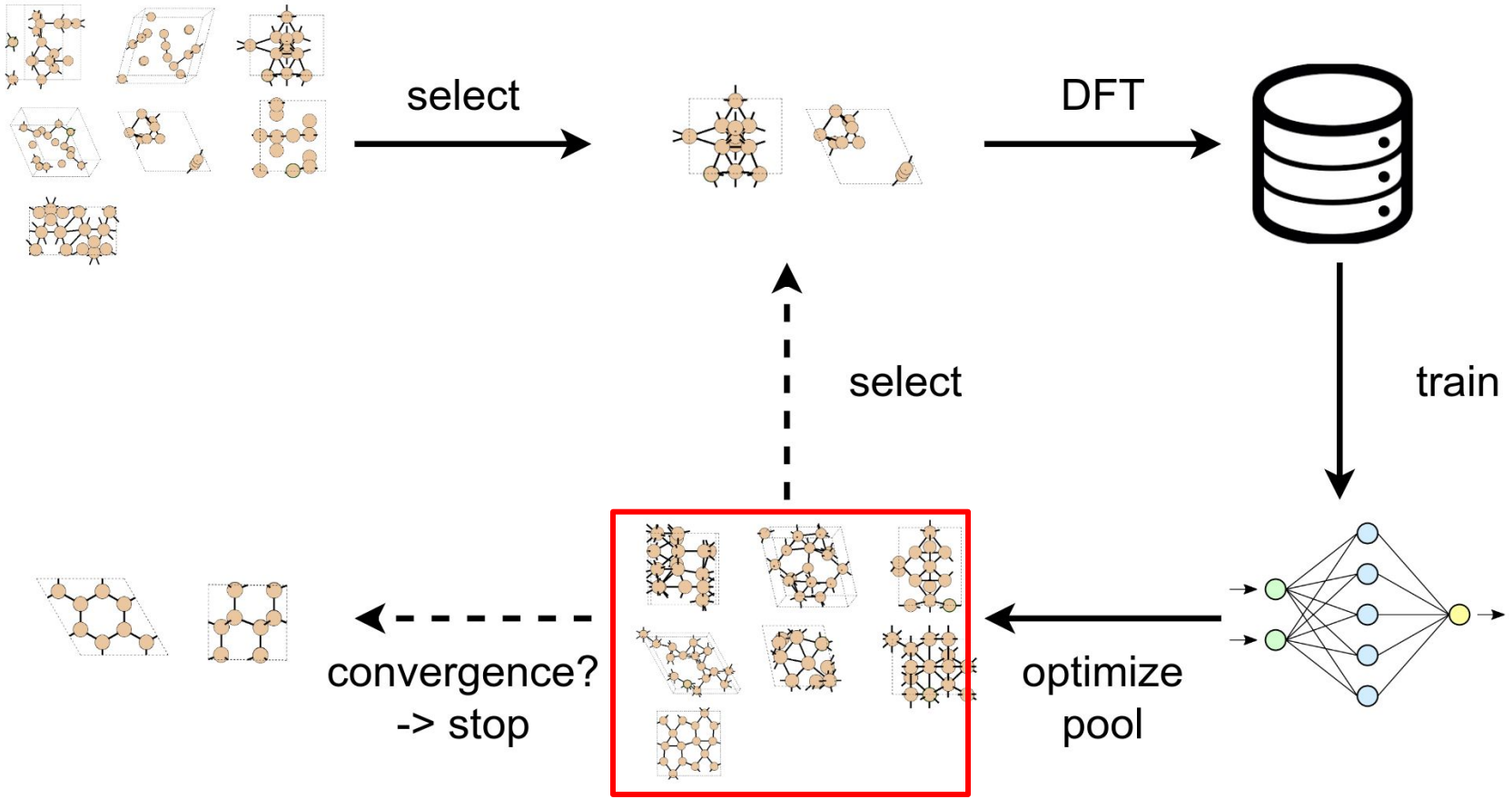
Method overview



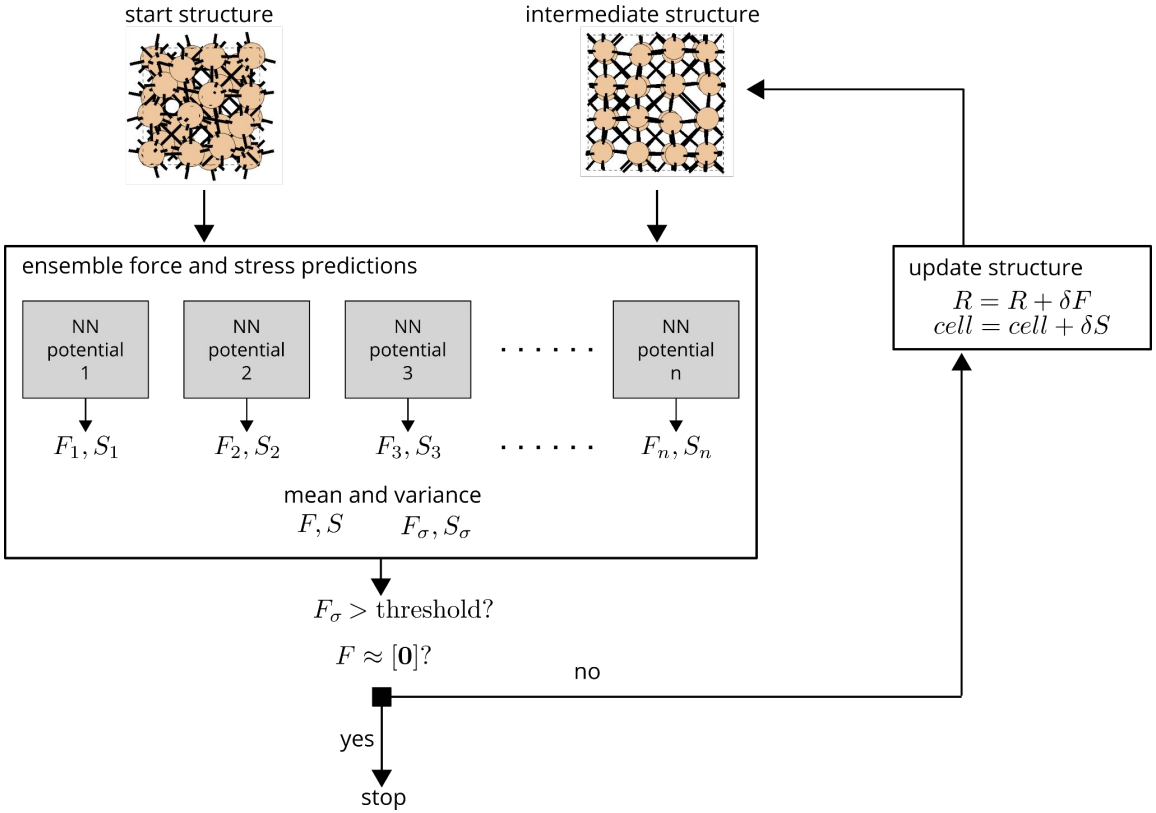
Method overview



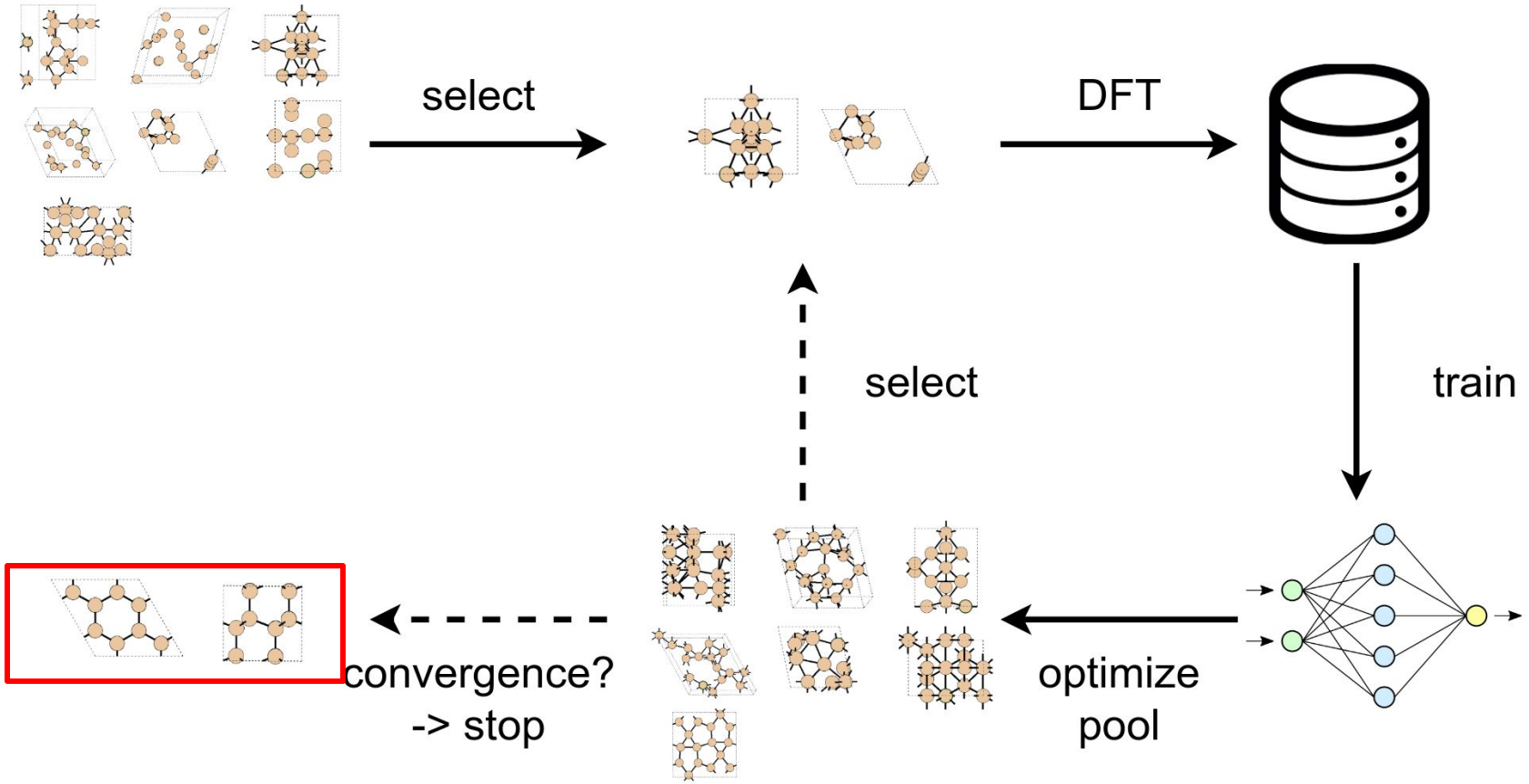
Method overview



Optimization of candidate pool



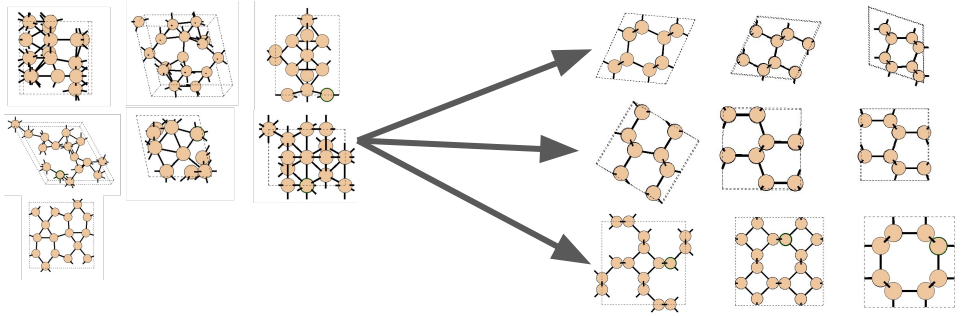
Method overview



Selecting structures for DFT relaxation

current pool

clusters sorted by energy



previous pool

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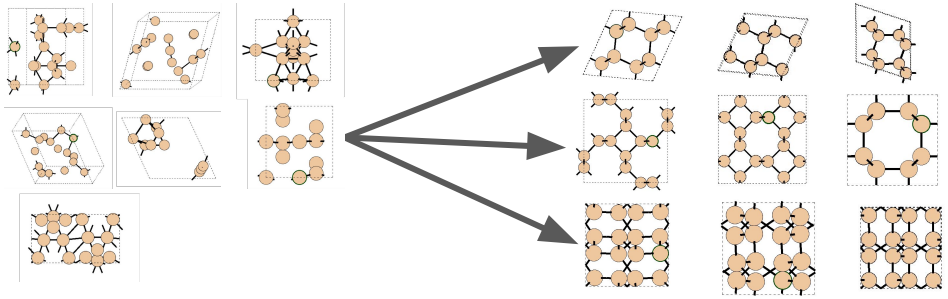
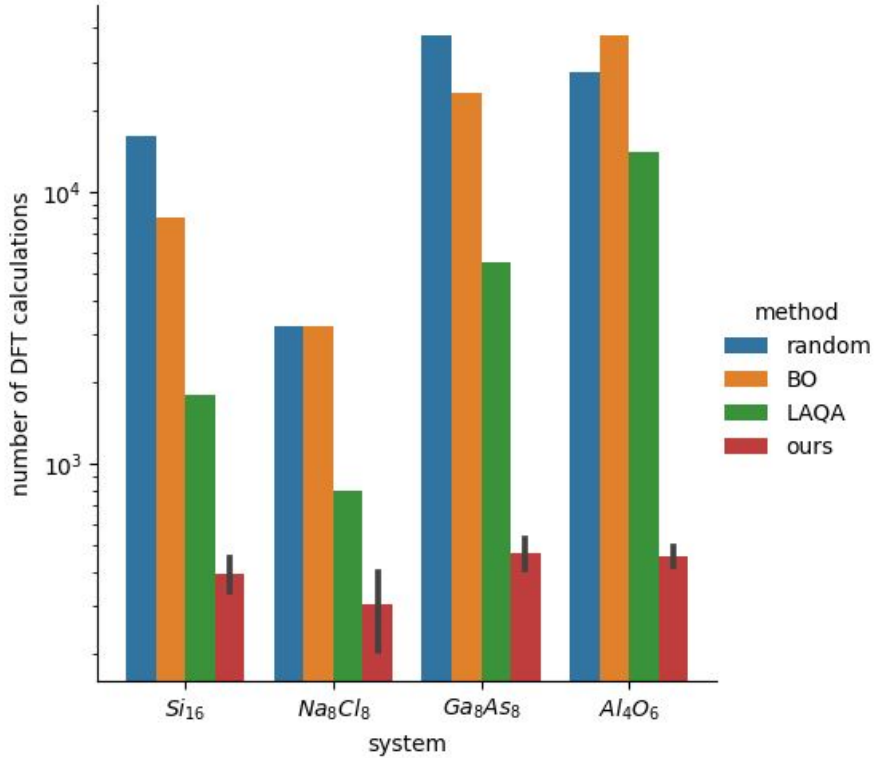


stop if top-k clusters are equal

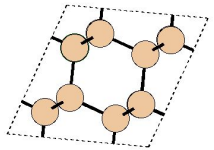
Results

Results

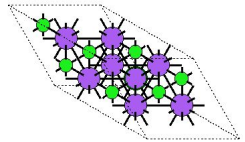


target structures:

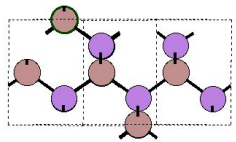
- Si: MP-149



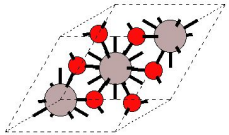
- NaCl: MP-22862



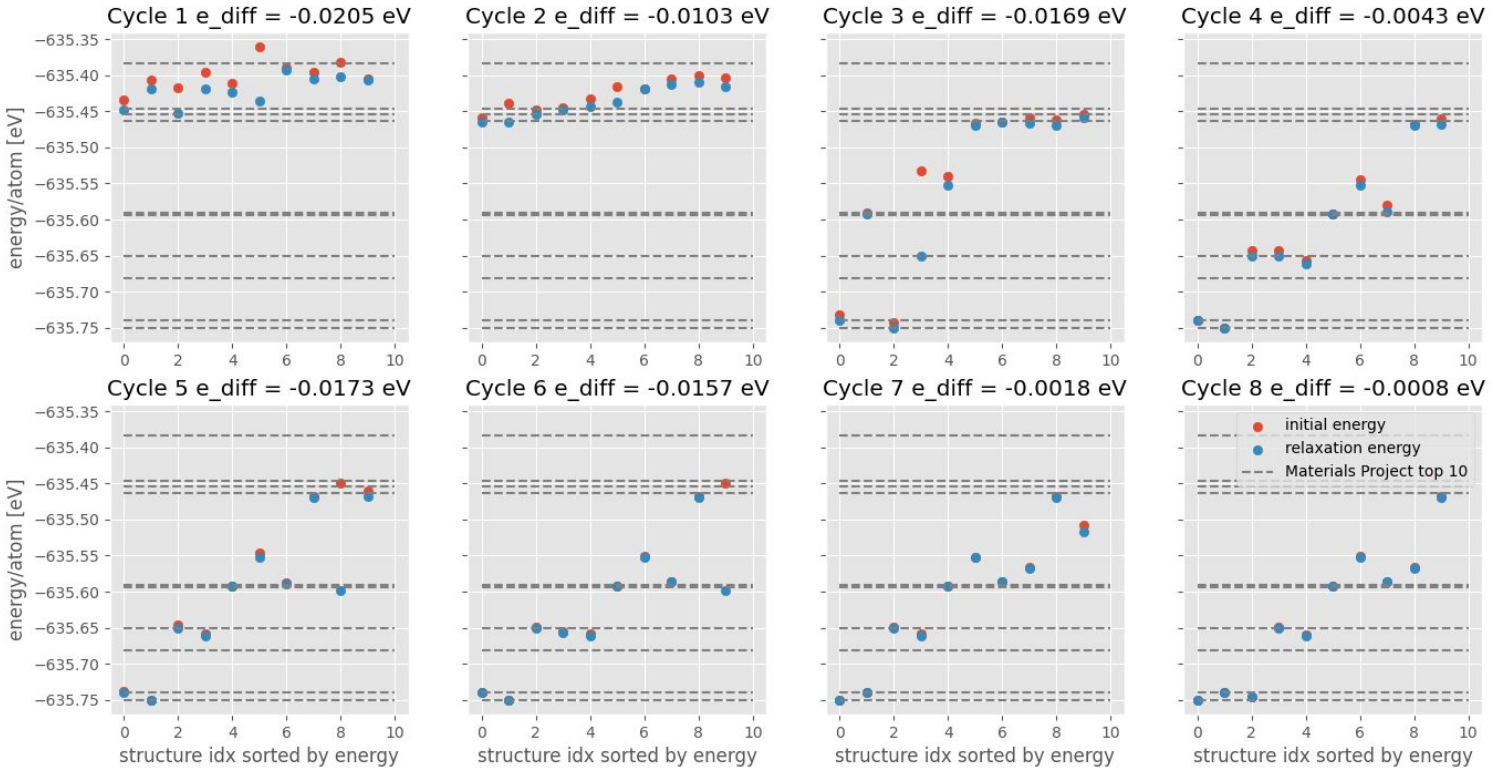
- GaAs: MP-2534



- AlO: MP-1143

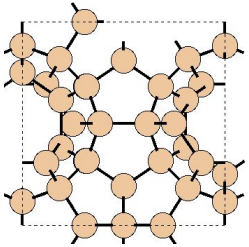


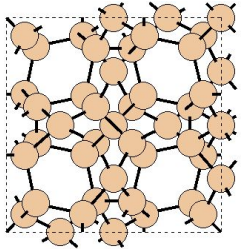
Results: multiple minima

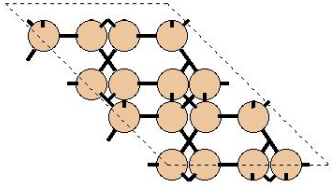


Results: transferability Si16 to Si46

top 3 minima:

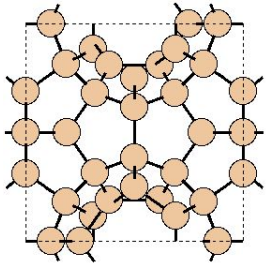




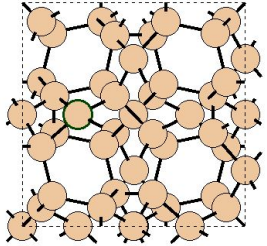


target structures:

- MP-971662



- MP-971661



Thank you!

Questions?