Generalizing Neural Wave Functions

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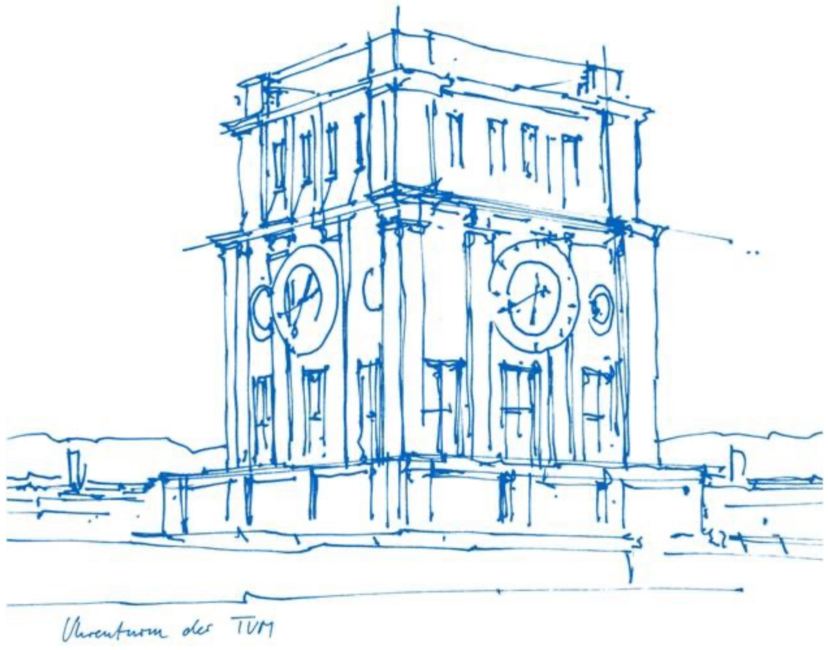


ICML 2023

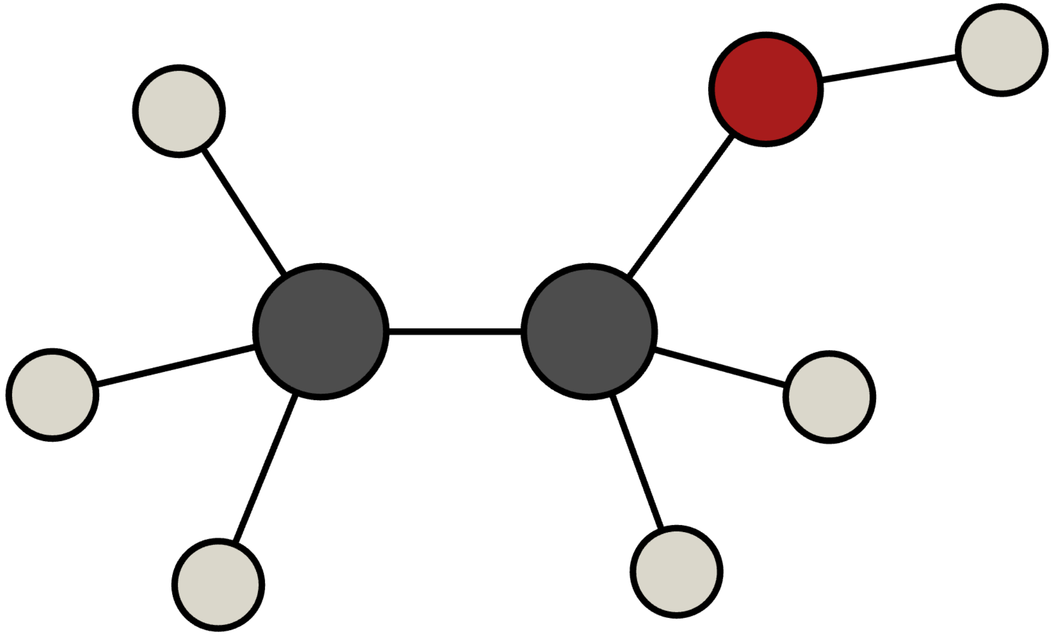
Schrödinger equation

$$\hat{H}\psi(r_1, \dots, r_N) = E\psi(r_1, \dots, r_N)$$

$r \in \mathbb{R}^{N\times 3}$ Electrons

$\psi: \mathbb{R}^{N\times 3} \to \mathbb{R}$ Wave function

$E \in \mathbb{R}$ Energy

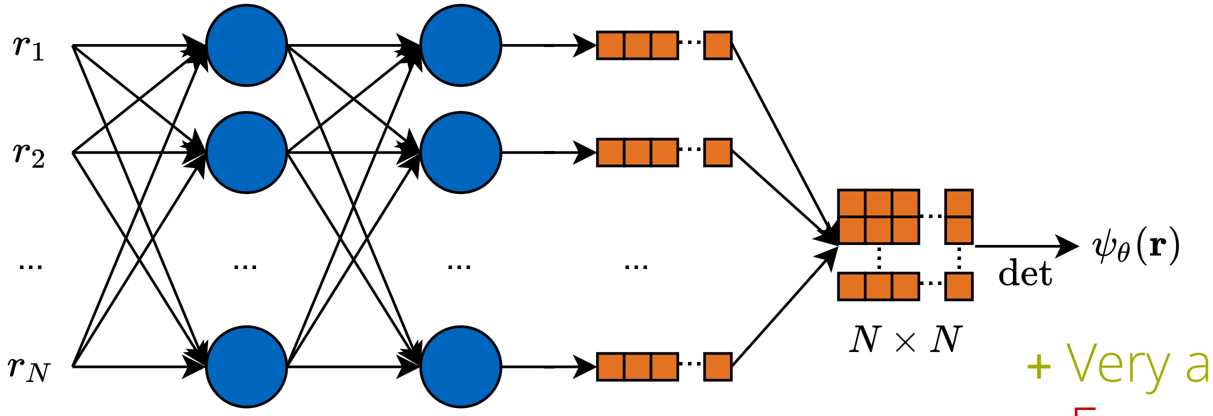


No analytical solutions ☹

$\Rightarrow$ Approximate!

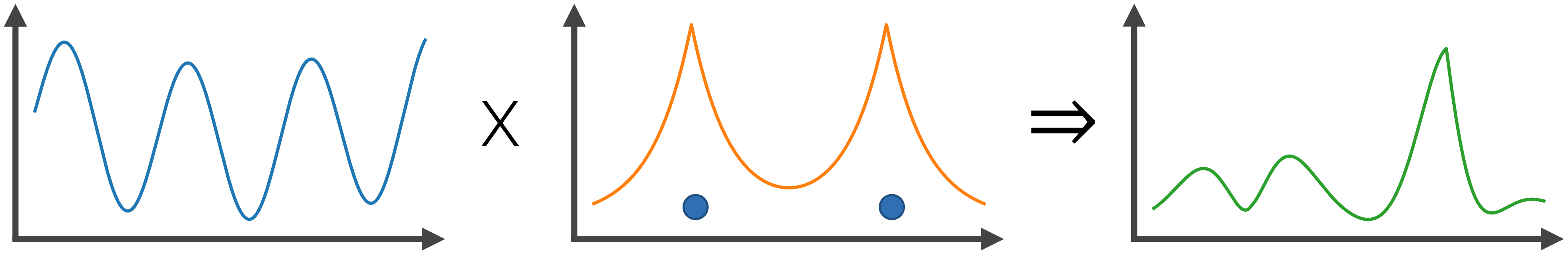
Deep Variational Monte Carlo

1. Must be antisymmetric $\psi_\theta(\dots, r_i, \dots, r_j, \dots) = -\psi_\theta(\dots, r_j, \dots, r_i, \dots)$



+ Very accurate results
– Expensive
O(100k) iterations per structure

2. $\int \psi_\theta(\boldsymbol{r}) d\boldsymbol{r}$ must be finite



Potential Energy Surface Network

MetaGNN

- Encodes geometry
- Parametrizes wave function

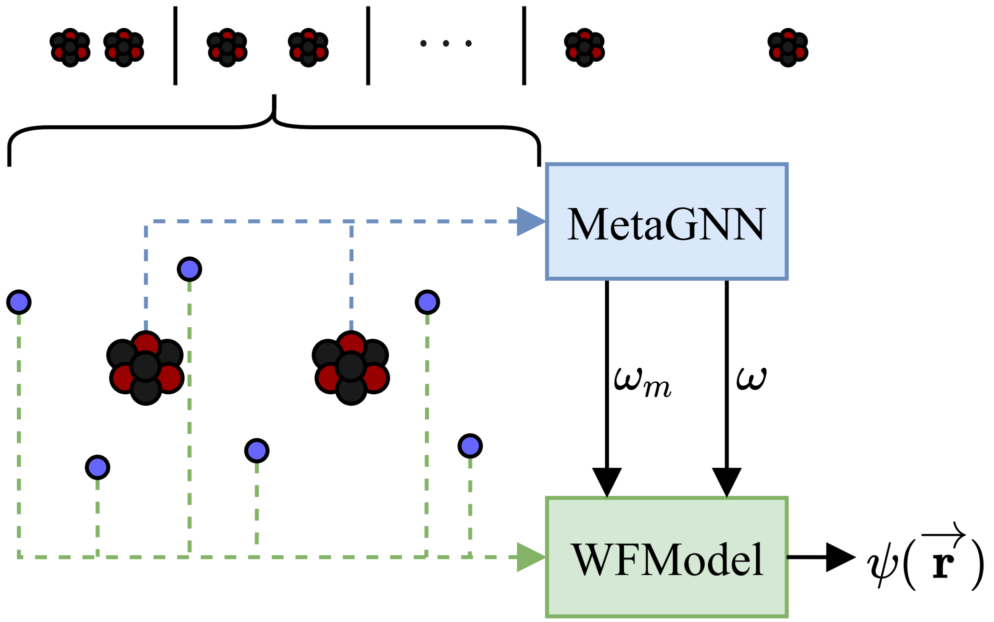
WFModel

- Represents electronic wave function
- Optimized via VMC

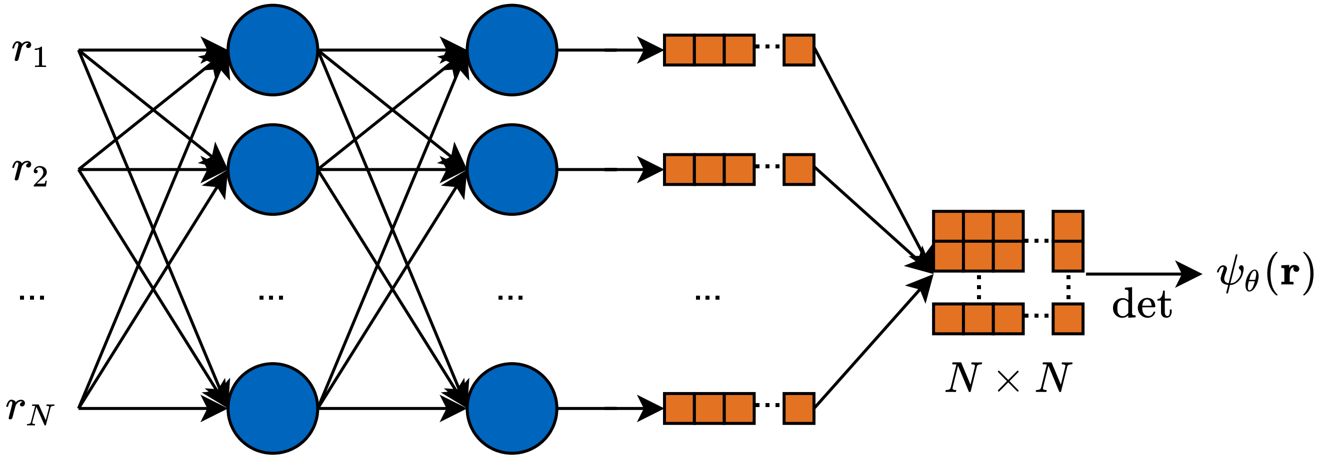
+ end-to-end differentiable

+ single training for PES

– Limited to same set of atoms



Molecular orbitals



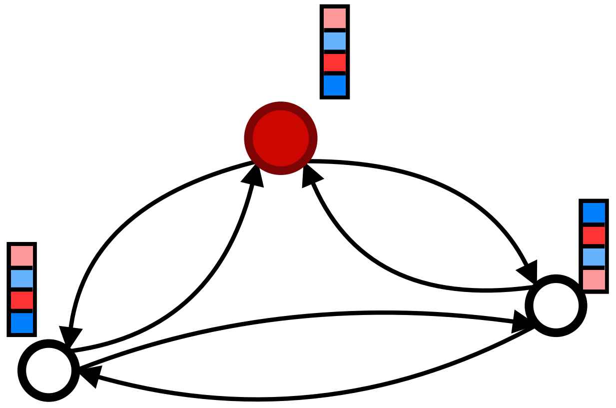
$$\psi(\boldsymbol{r}) = \begin{bmatrix} \phi_1(r_1) & \cdots & \phi_N(r_1) \\ \vdots & \ddots & \vdots \\ \phi_1(r_N) & \cdots & \phi_N(r_N) \end{bmatrix}$$

Challenges:

- Varying number of orbitals
- Similarities between orbitals
- Structure induced
- Learned functions

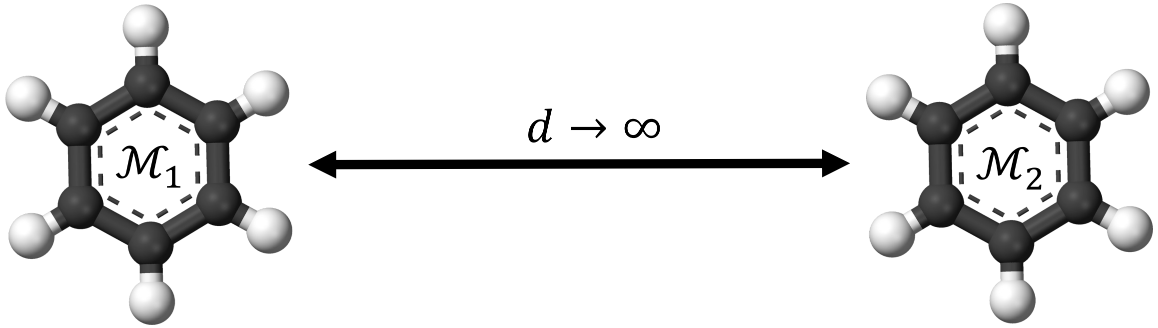
Graph-learned orbital embeddings

1. Learn nuclei embeddings



- PESNet reparametrization

Size consistency



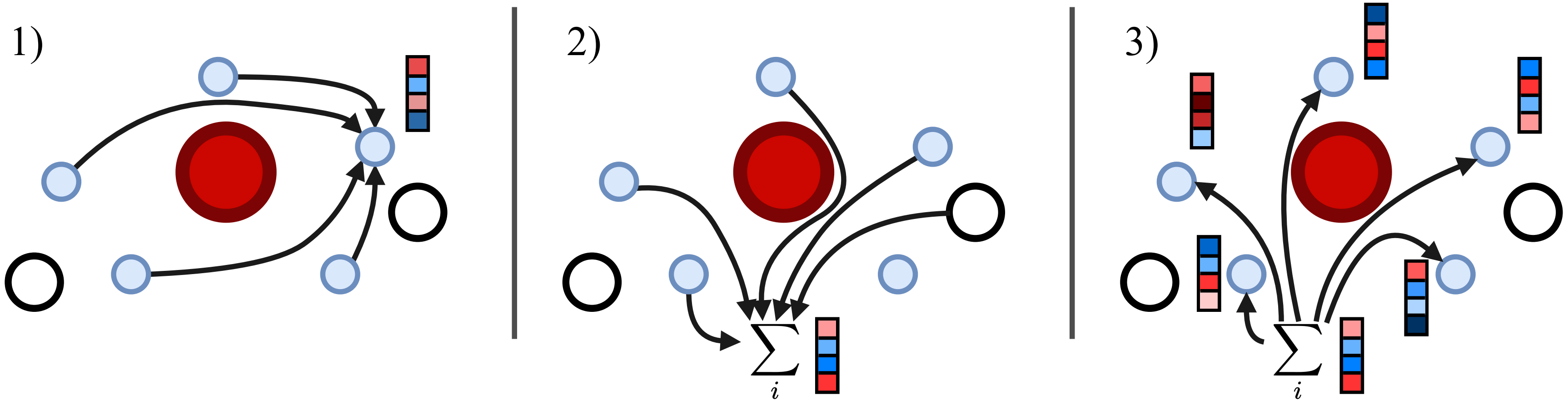
$$E_{total} = E(\mathcal{M}_1) + E(\mathcal{M}_2)$$

$$\Rightarrow \psi_{total} = \psi_1\psi_2 = \begin{vmatrix} \Phi_1(r_{\mathcal{M}_1}) & 0 \\ 0 & \Phi_2(r_{\mathcal{M}_2}) \end{vmatrix}$$

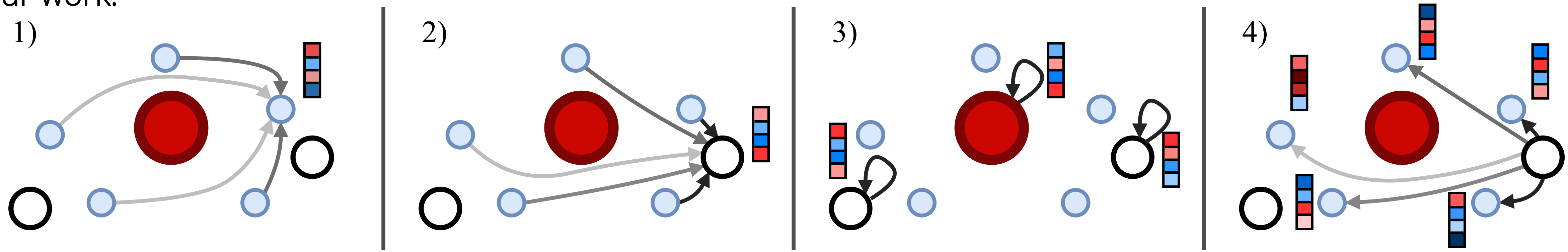
$$\Rightarrow \phi_i(r_j, \boldsymbol{r}) = \begin{cases} \phi_i(r_j | \boldsymbol{r}_{\mathcal{M}_1}) & \text{, if } i, j \in \mathcal{M}_1 \\ \phi_i(r_j | \boldsymbol{r}_{\mathcal{M}_2}) & \text{, if } i, j \in \mathcal{M}_2 \\ 0 & \text{, else} \end{cases}$$

Size-consistent Neural Wave Functions

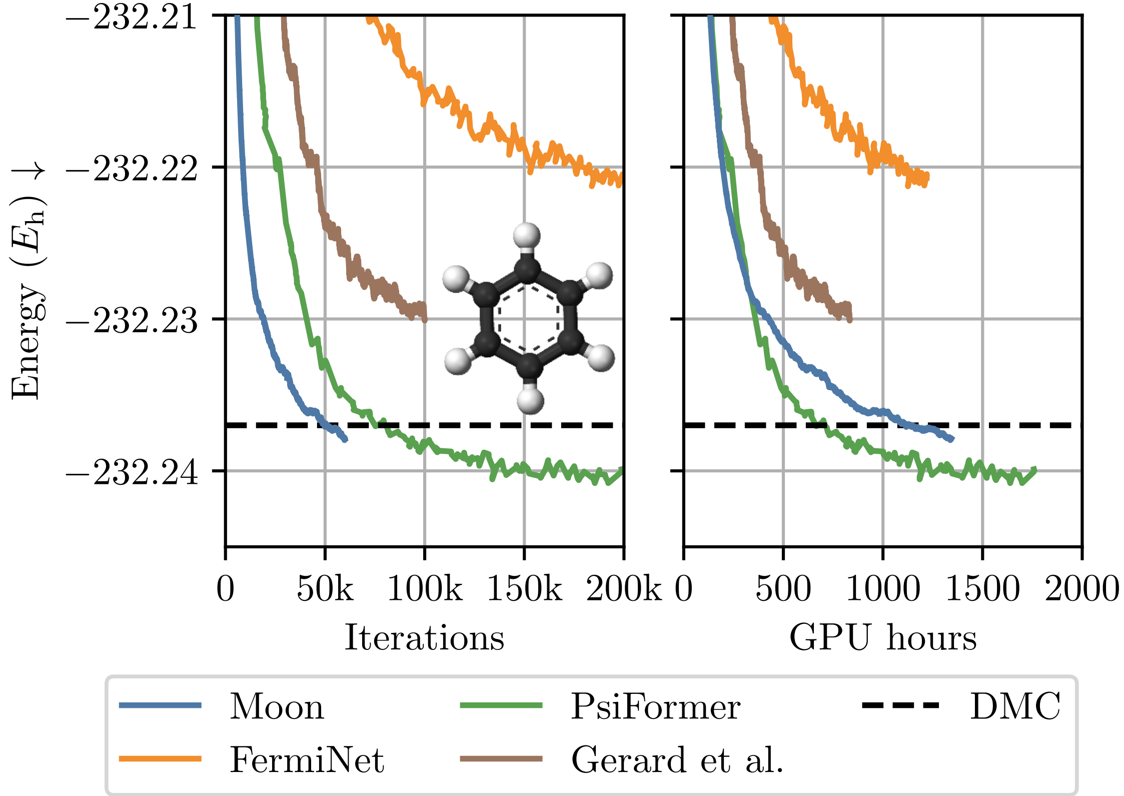
Previous work:



Our work:

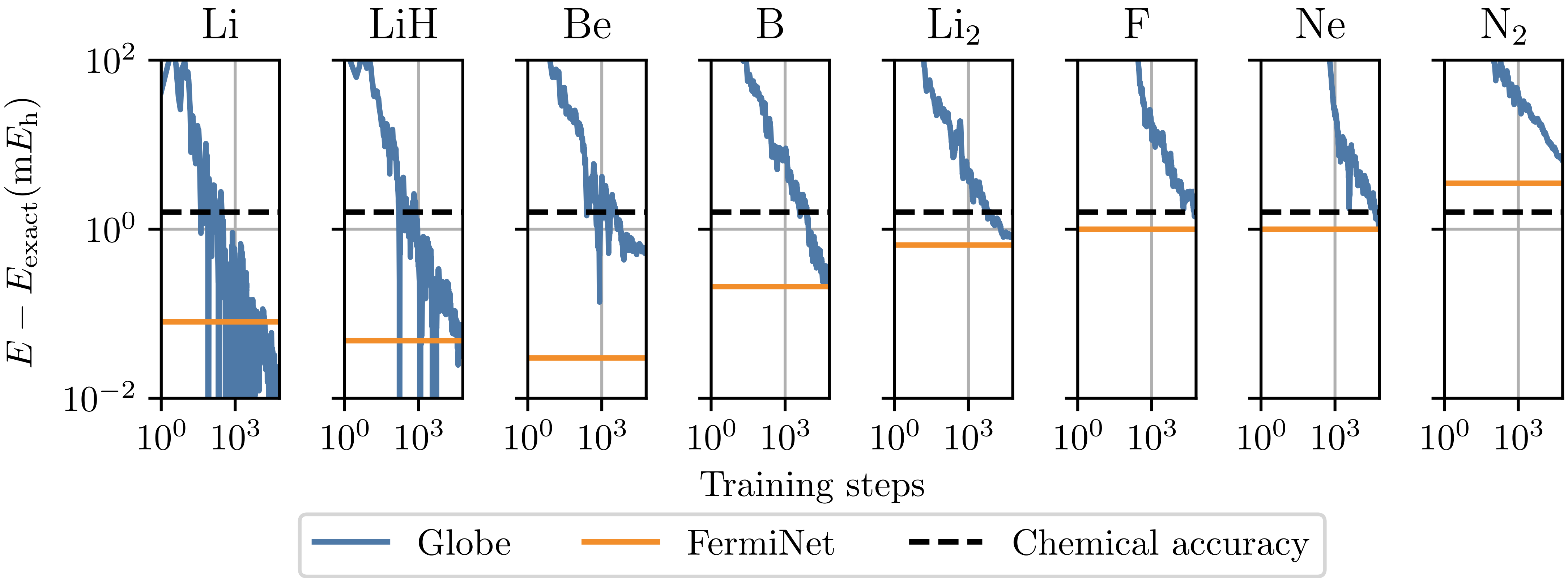


Experiments – Large molecules



Size-consistency is important for large molecules

Experiments – Different molecules



Ground state on many systems simultaneously

Conclusion

Graph-learned orbital embeddings

- Embed orbitals as points in space
- Learn orbital functions via message passing

Size-consistent wave function

- Local interactions within the molecular graph
- Implement correct limit behavior

Experiments

- Ground-state of various systems
- Accelerated and better convergence on large systems

https://www.cs.cit.tum.de/daml/globe/