



Center for Scientific Computing,
Theory and Data

Koopmans spectral functionals

Electronic screening via machine learning

Edward Linscott

MARVEL Pillar IV Meeting, 14 November 2024

Electronic screening via machine learning



$$\alpha_i = \frac{\langle n_i | \varepsilon^{-1} f_{\text{Hxc}} | n_i \rangle}{\langle n_i | f_{\text{Hxc}} | n_i \rangle}$$

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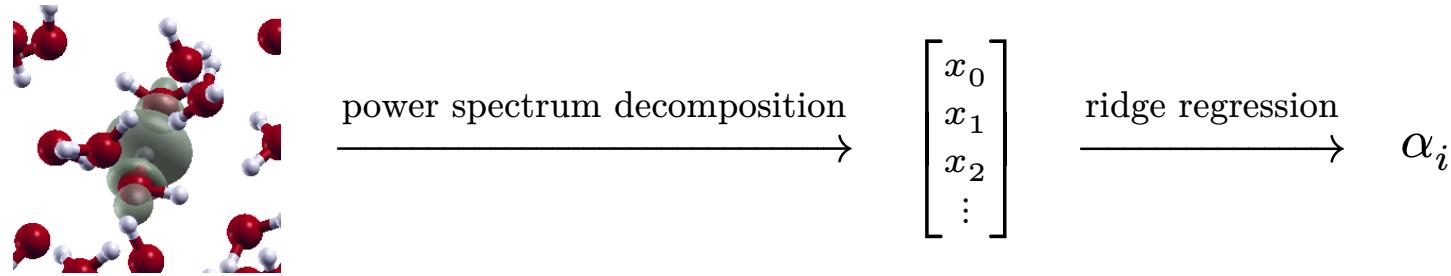
- must be computed *ab initio* via ΔSCF ¹ or DFPT²
- one screening parameter per (non-equivalent) orbital
- corresponds to the vast majority of the computational cost
- critical that they are accurate; if $\psi_i(\mathbf{r}) = \sum_j U_{ij} \varphi_j(\mathbf{r})$ then

$$\Delta\varepsilon_{i \in \text{occ}} = \sum_j \alpha_j U_{ij} U_{ji}^\dagger \left(-E_{\text{Hxc}} [\rho - \rho_j] + E_{\text{Hxc}} [\rho - \rho_j + n_j] - \int d\mathbf{r} v_{\text{Hxc}} [\rho - \rho_j + n_j](\mathbf{r}) n_j(\mathbf{r}) \right)$$

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The machine-learning framework

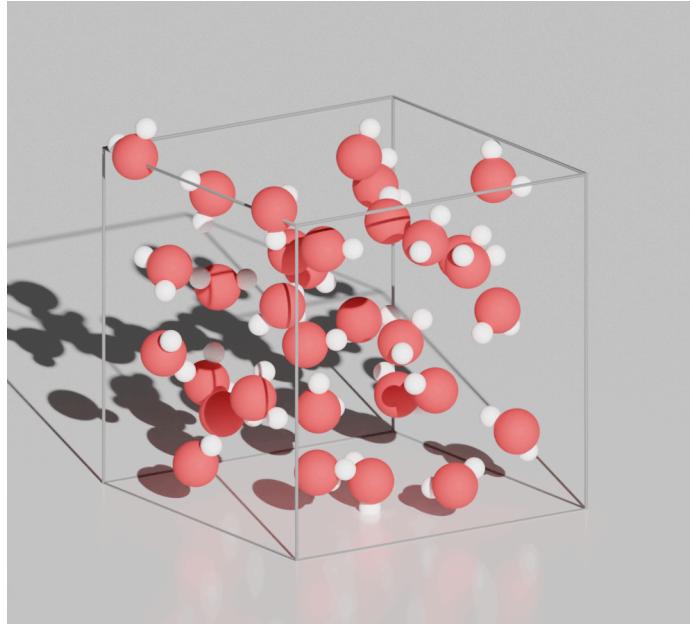


$$c_{nlm,k}^i = \int d\mathbf{r} g_{nl}(r) Y_{lm}(\theta, \varphi) n_i(\mathbf{r} - \mathbf{R}_i)$$

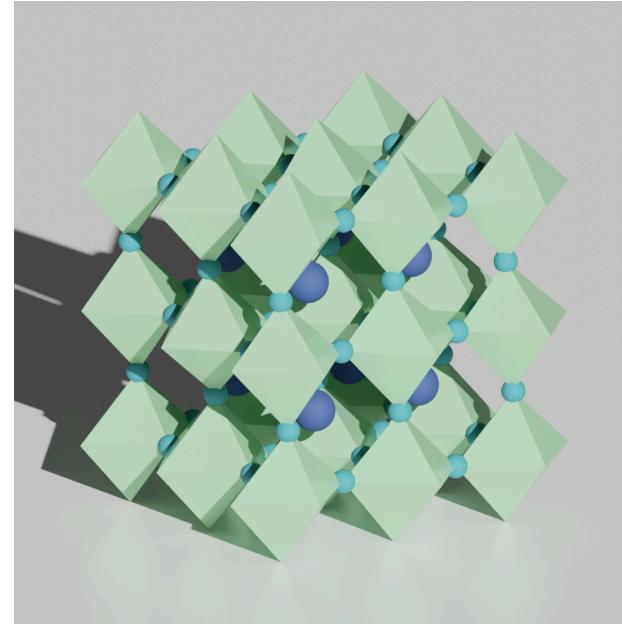
$$p_{n_1 n_2 l, k_1 k_2}^i = \pi \sqrt{\frac{8}{2l+1}} \sum_m c_{n_1 lm, k_1}^{i*} c_{n_2 lm, k_2}^i$$

¹Y. Schubert *et al.* <http://arxiv.org/abs/2406.15205> (2024)

Two test systems



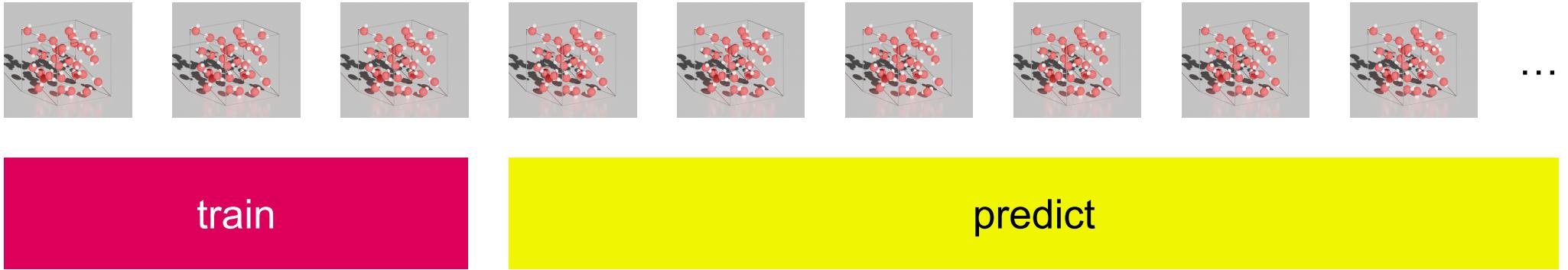
water



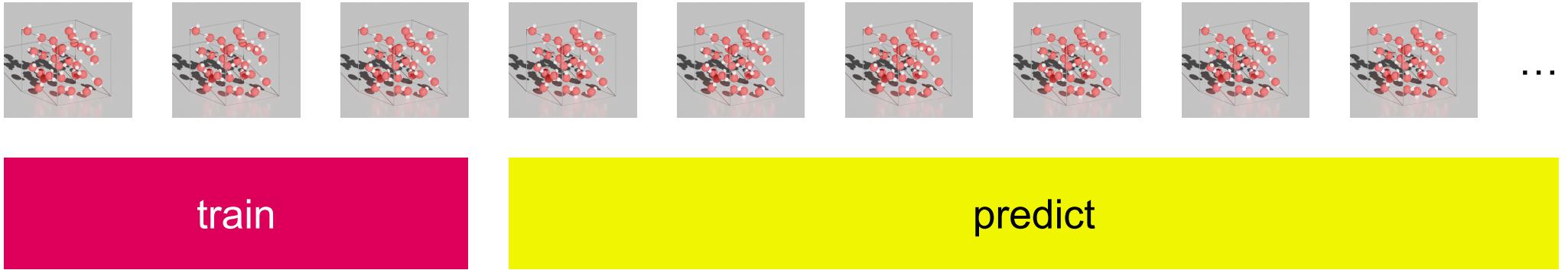
CsSnI₃

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Use case

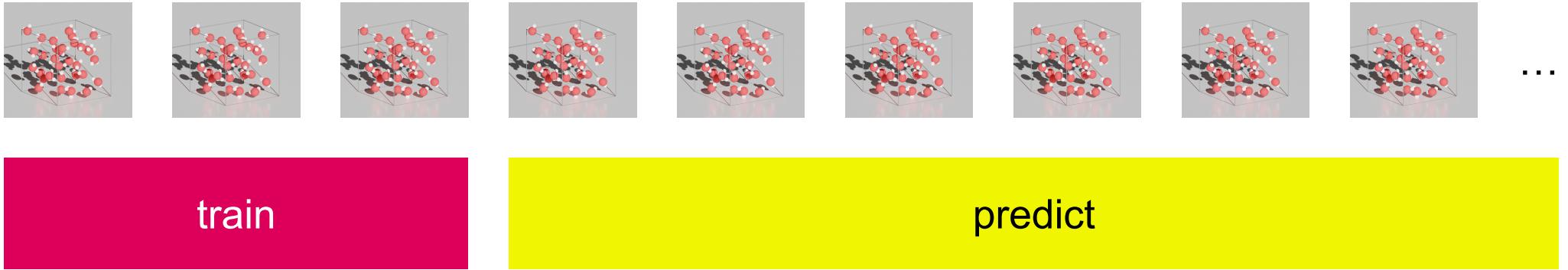


Use case



or train on a small cell and deploy on a larger cell

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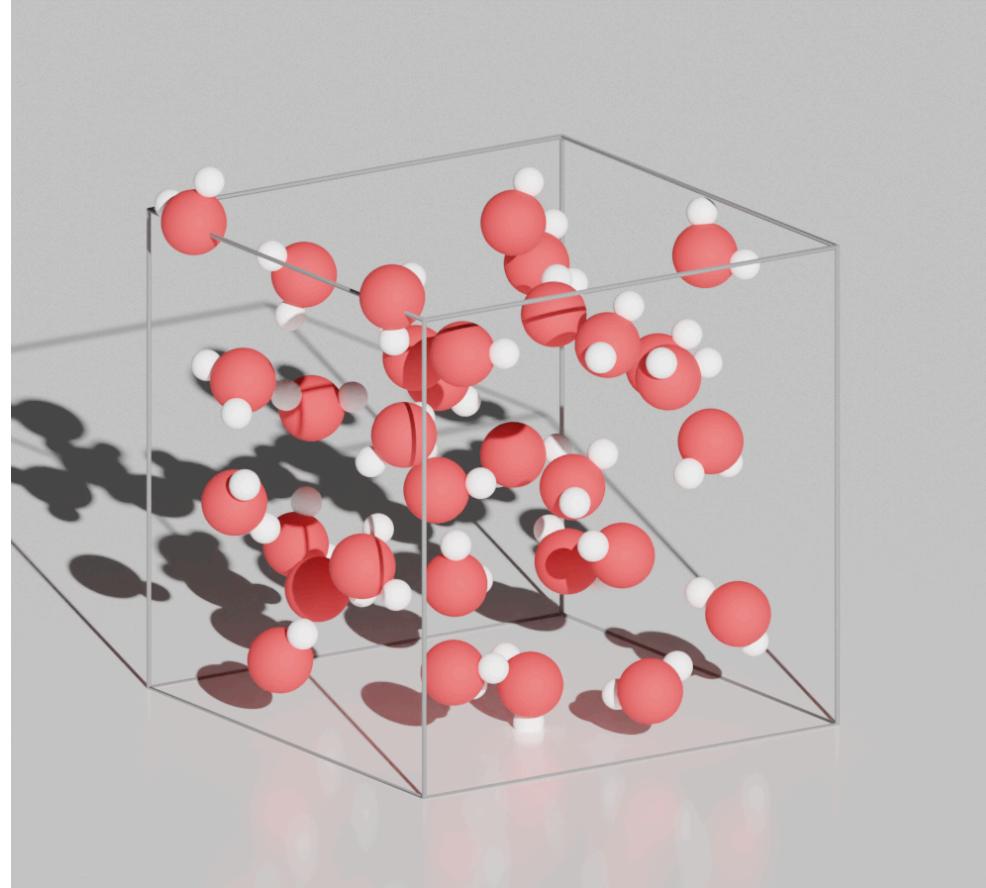


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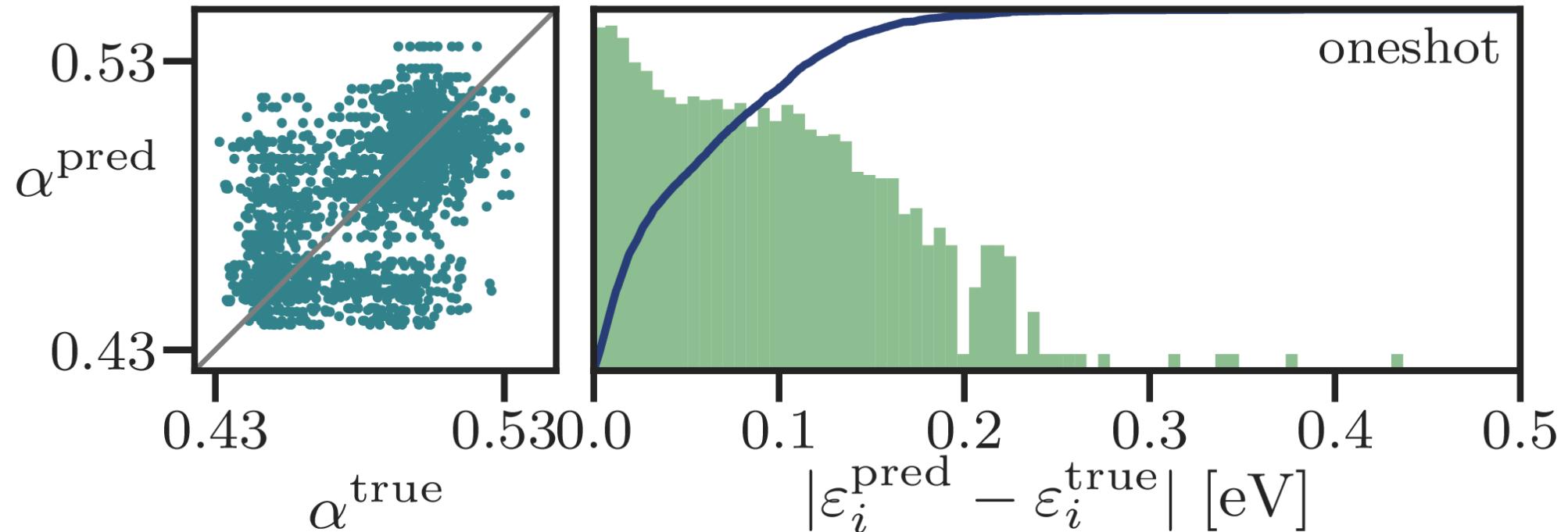
N.B. not a general-purpose model – trained on a case-by-case basis

Results

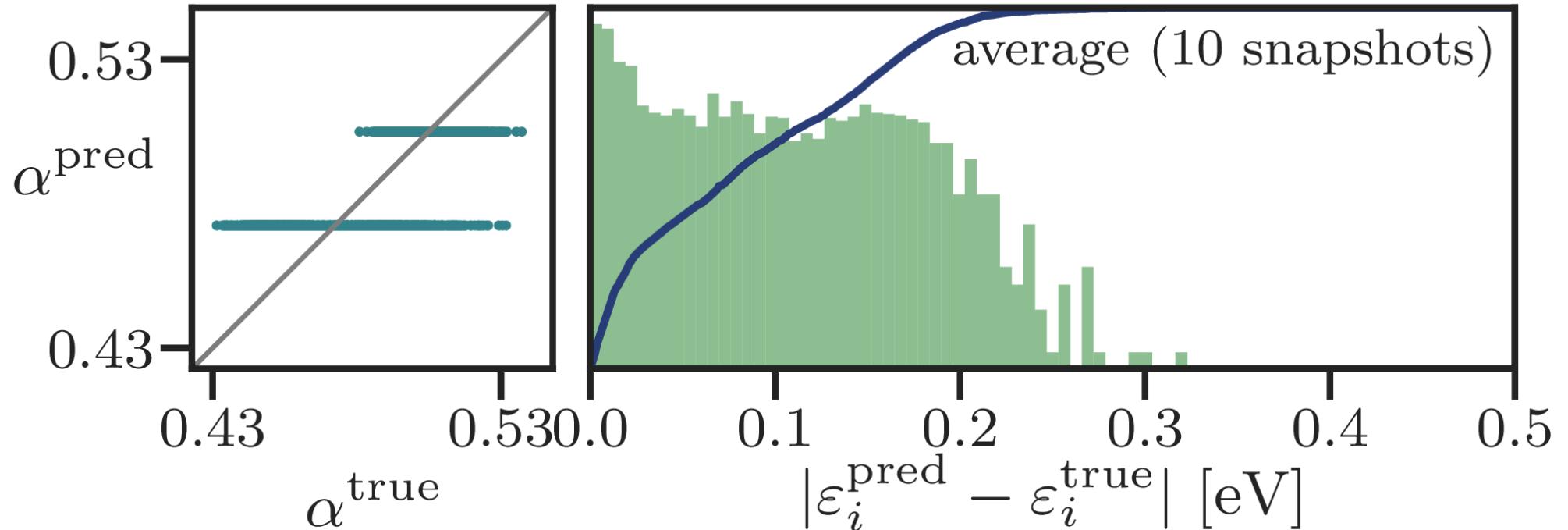
Accuracy



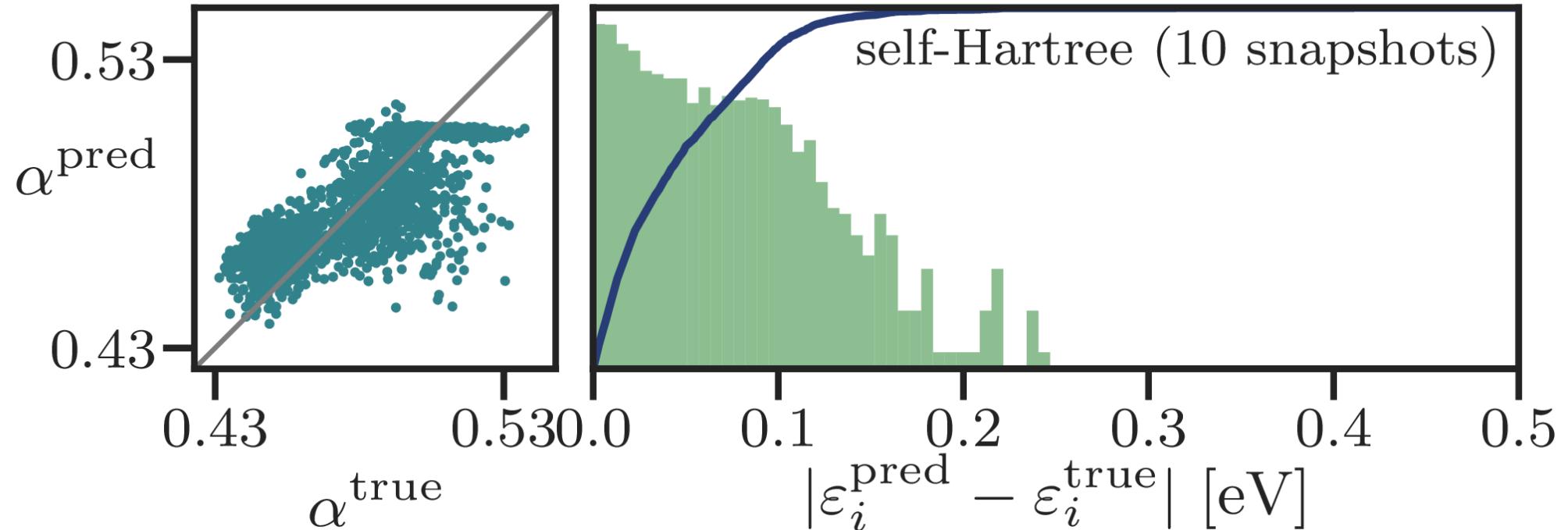
Accuracy



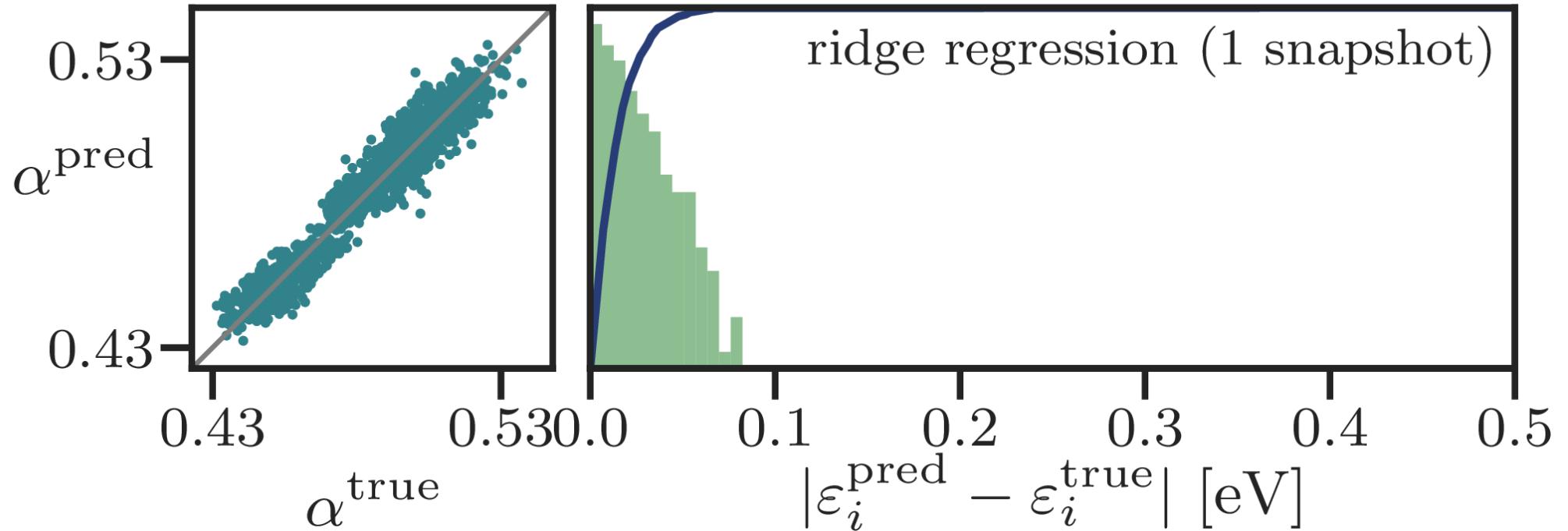
Accuracy



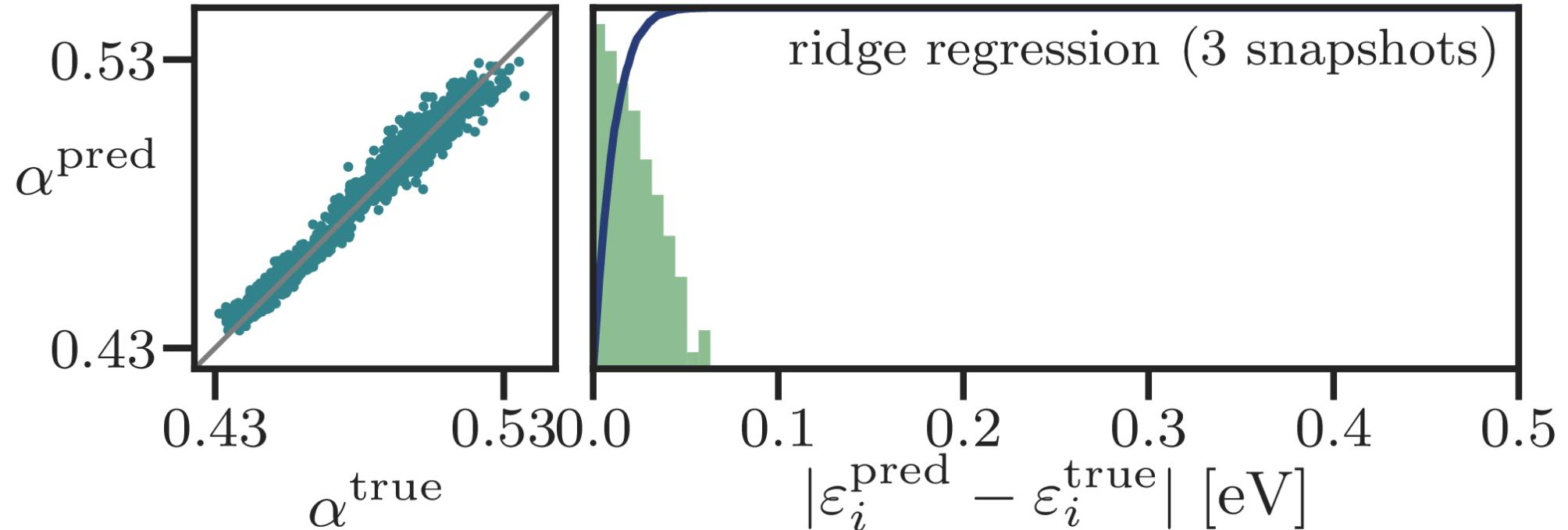
Accuracy



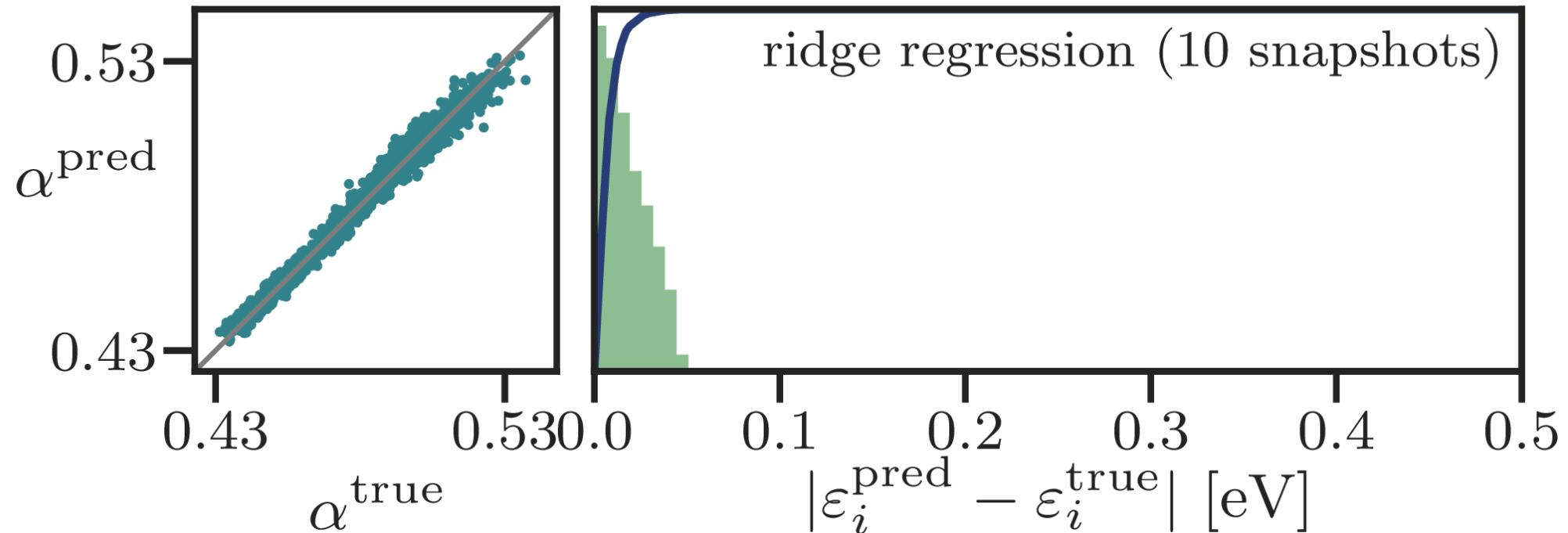
Accuracy



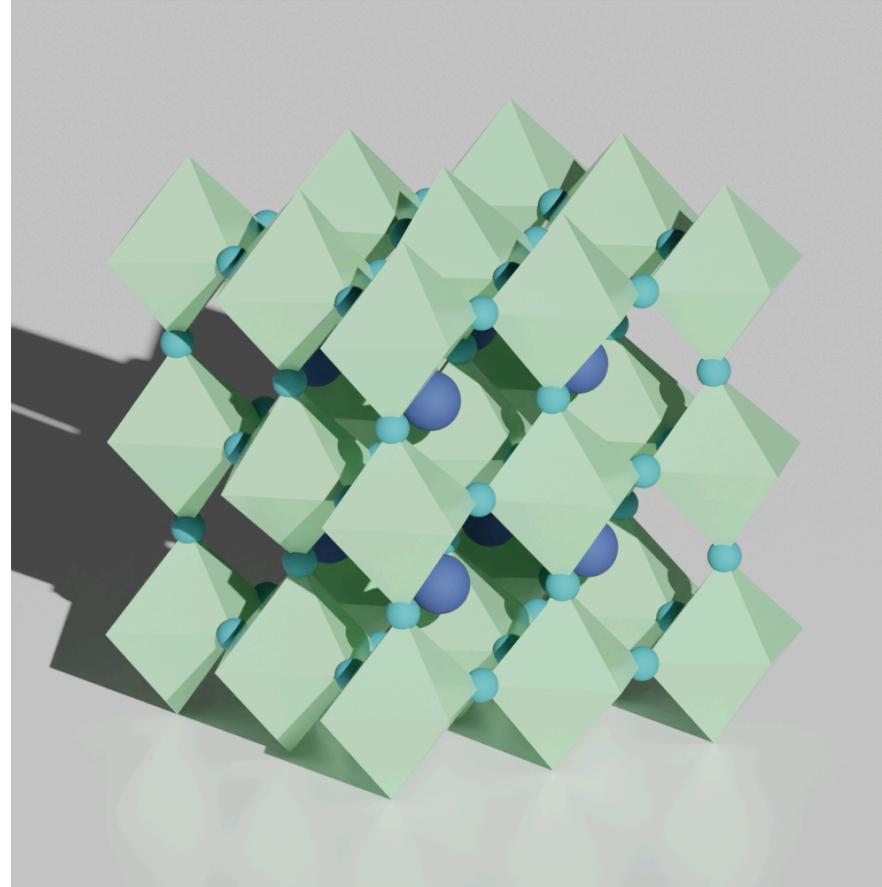
Accuracy



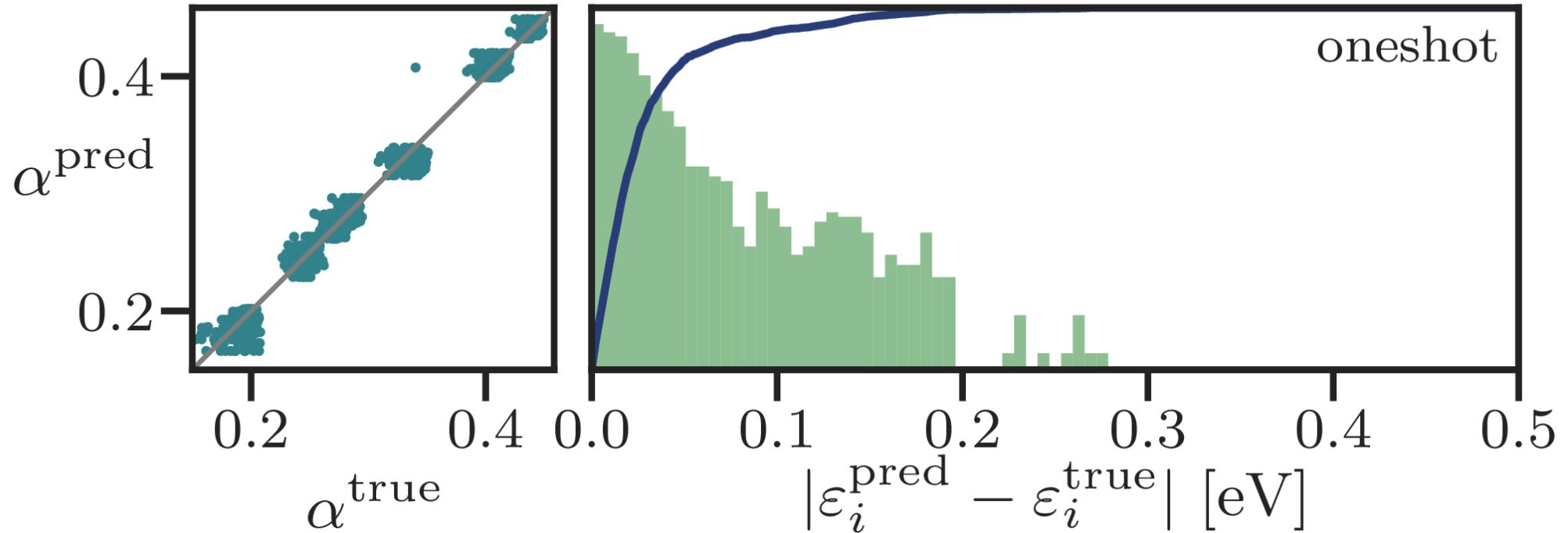
Accuracy



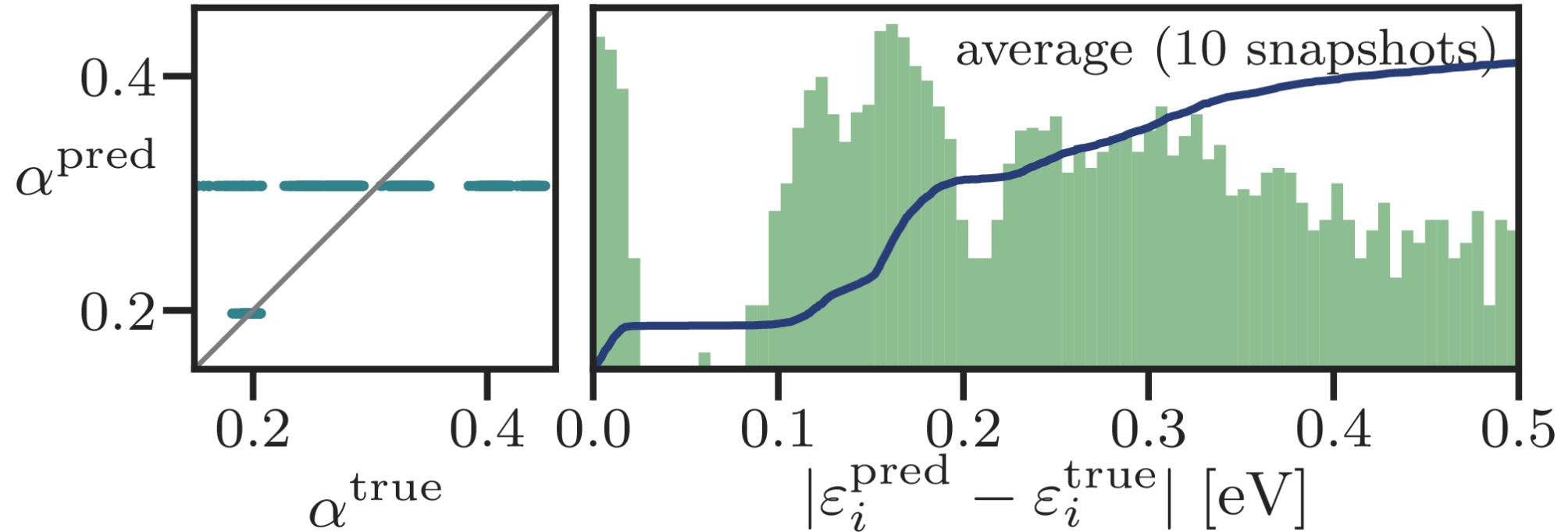
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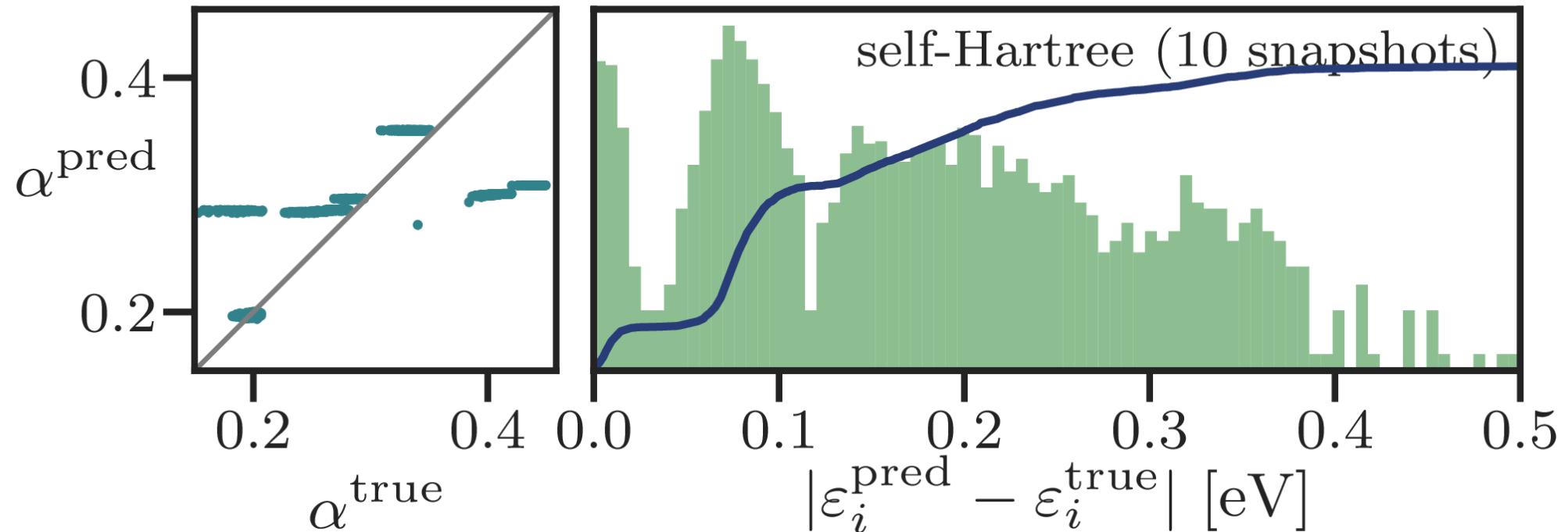
Accuracy



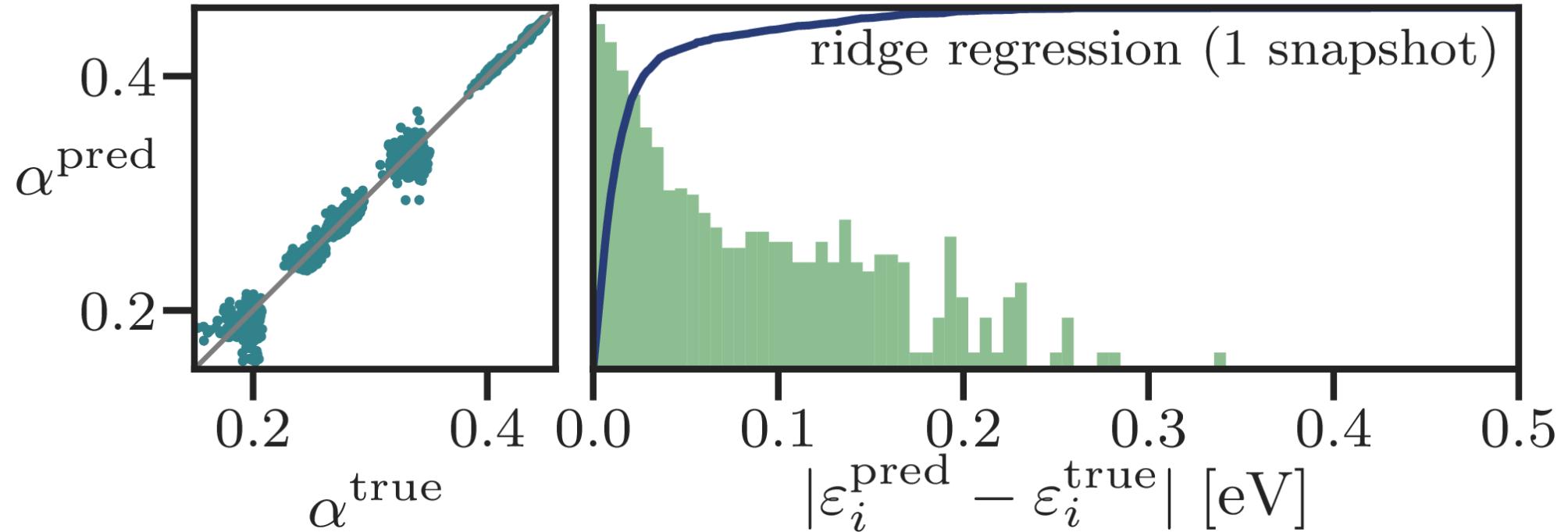
Accuracy



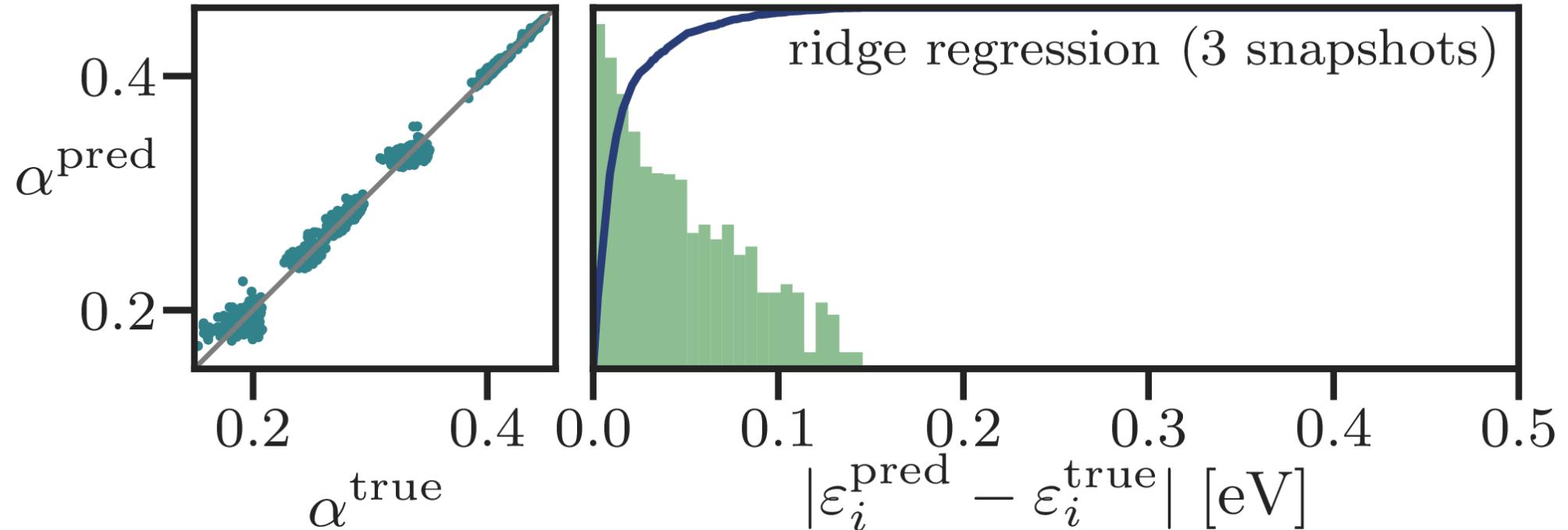
Accuracy



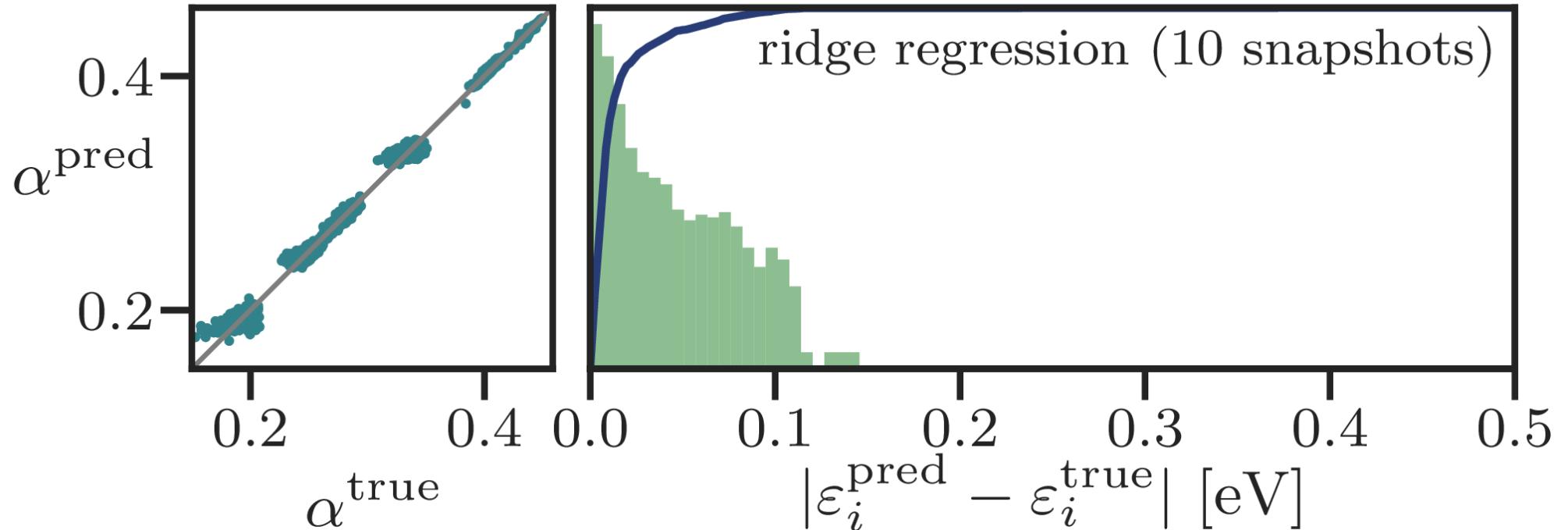
Accuracy



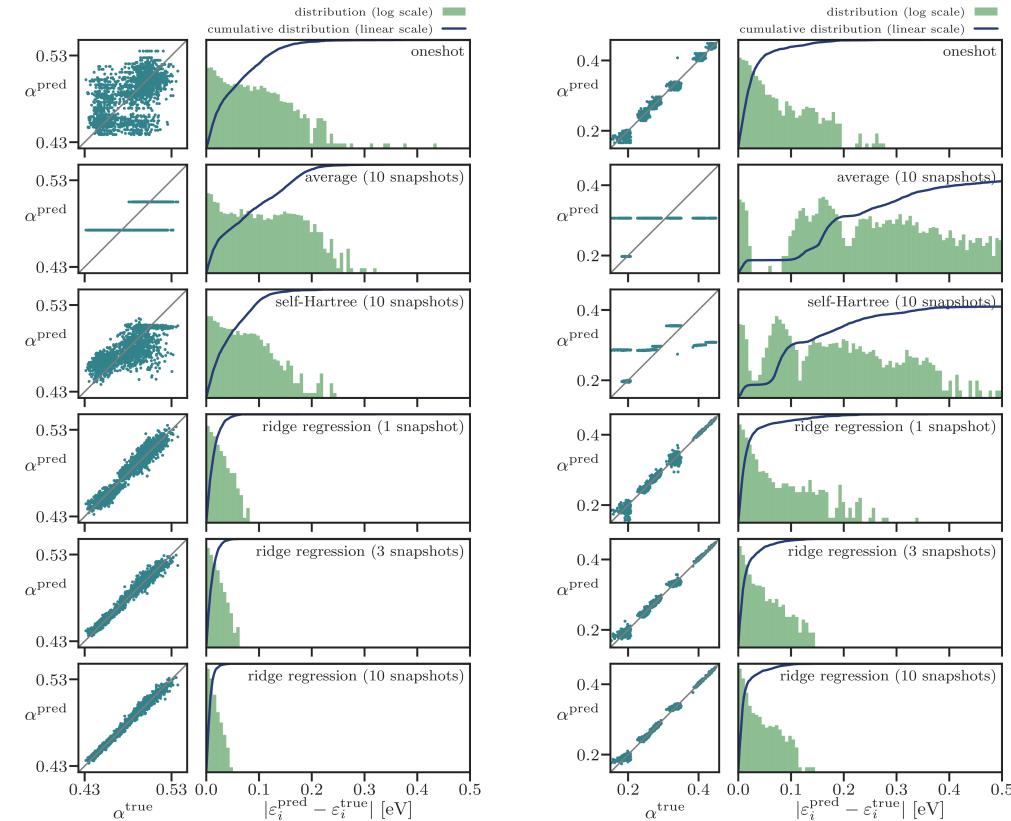
Accuracy



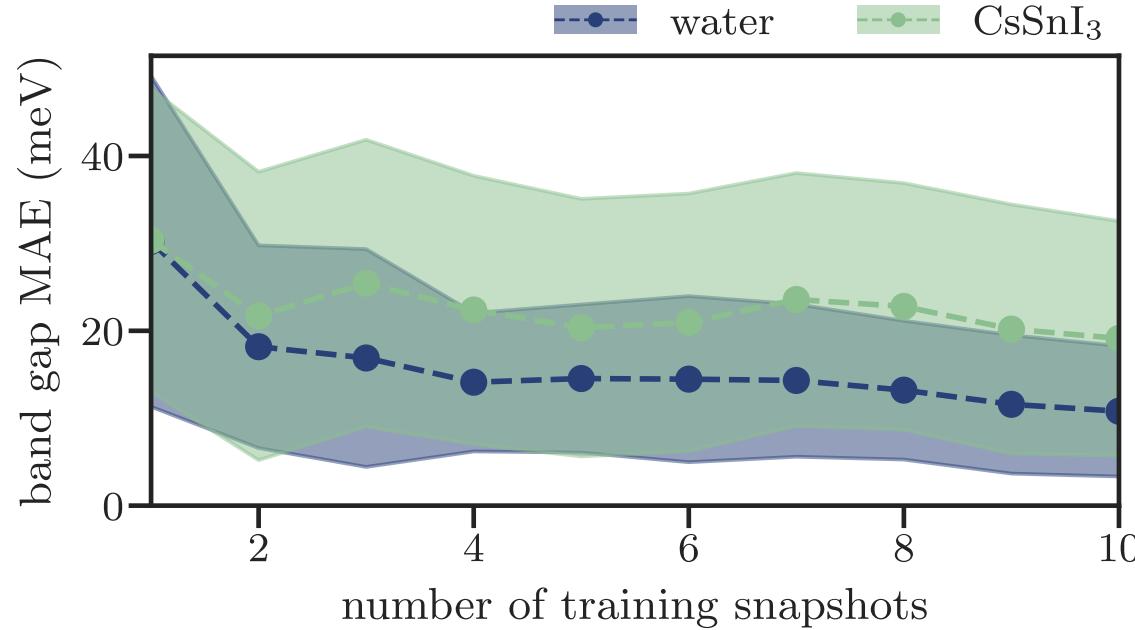
Accuracy



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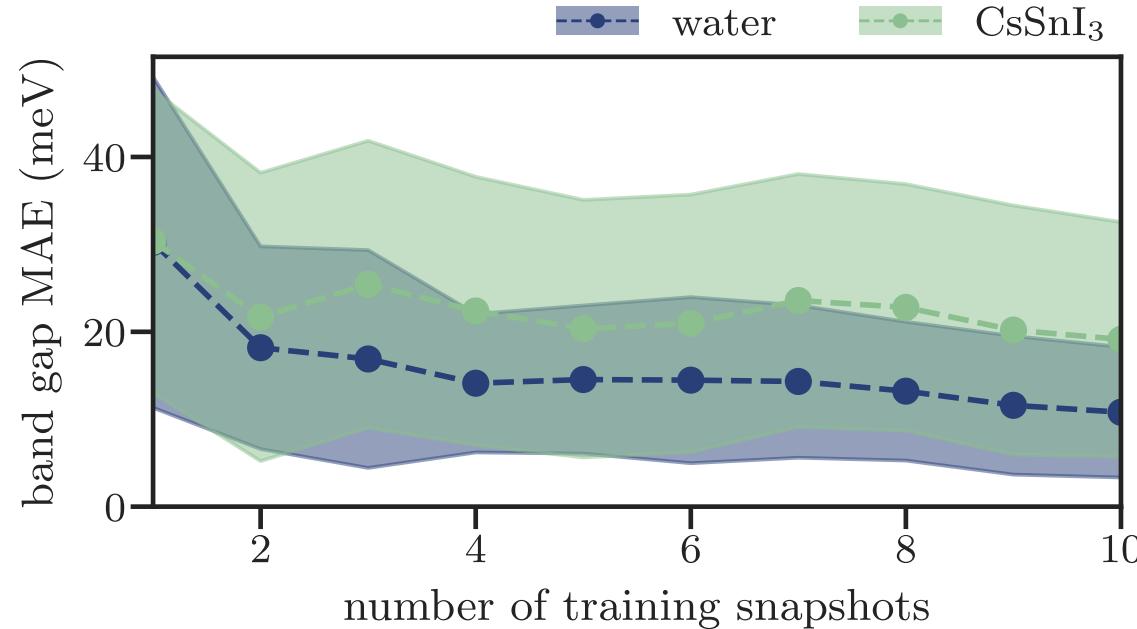


Accuracy



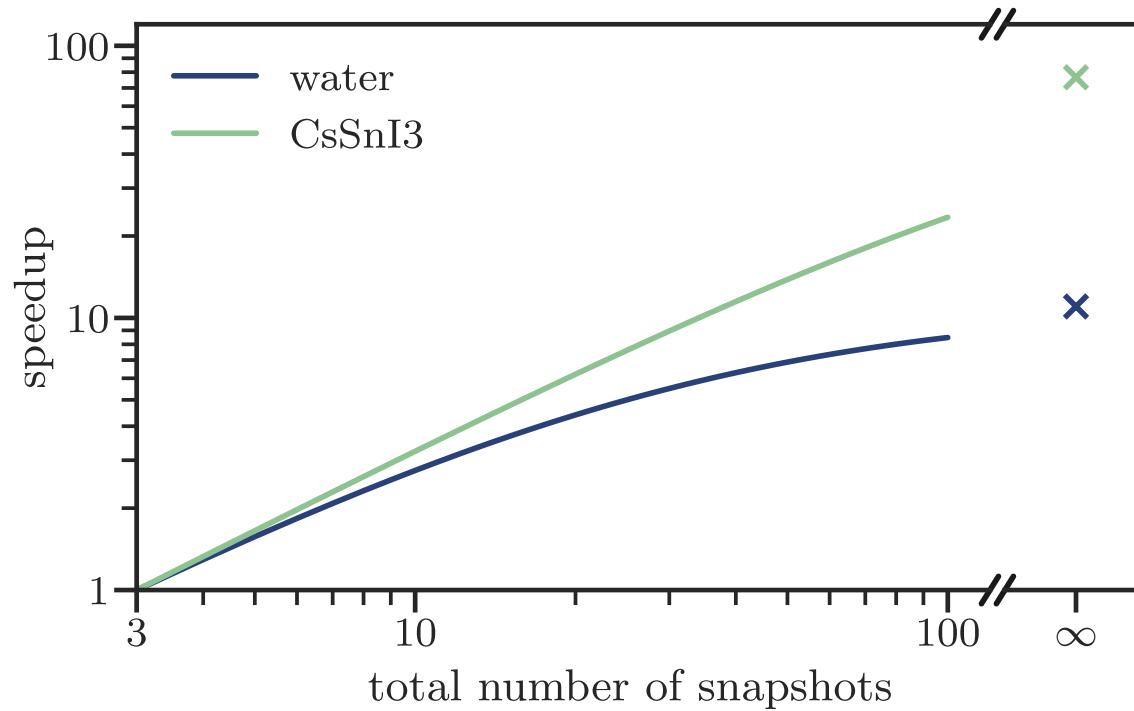
accurate to within $\mathcal{O}(10 \text{ meV})$ cf. typical band gap accuracy of $\mathcal{O}(100 \text{ meV})$;

Accuracy



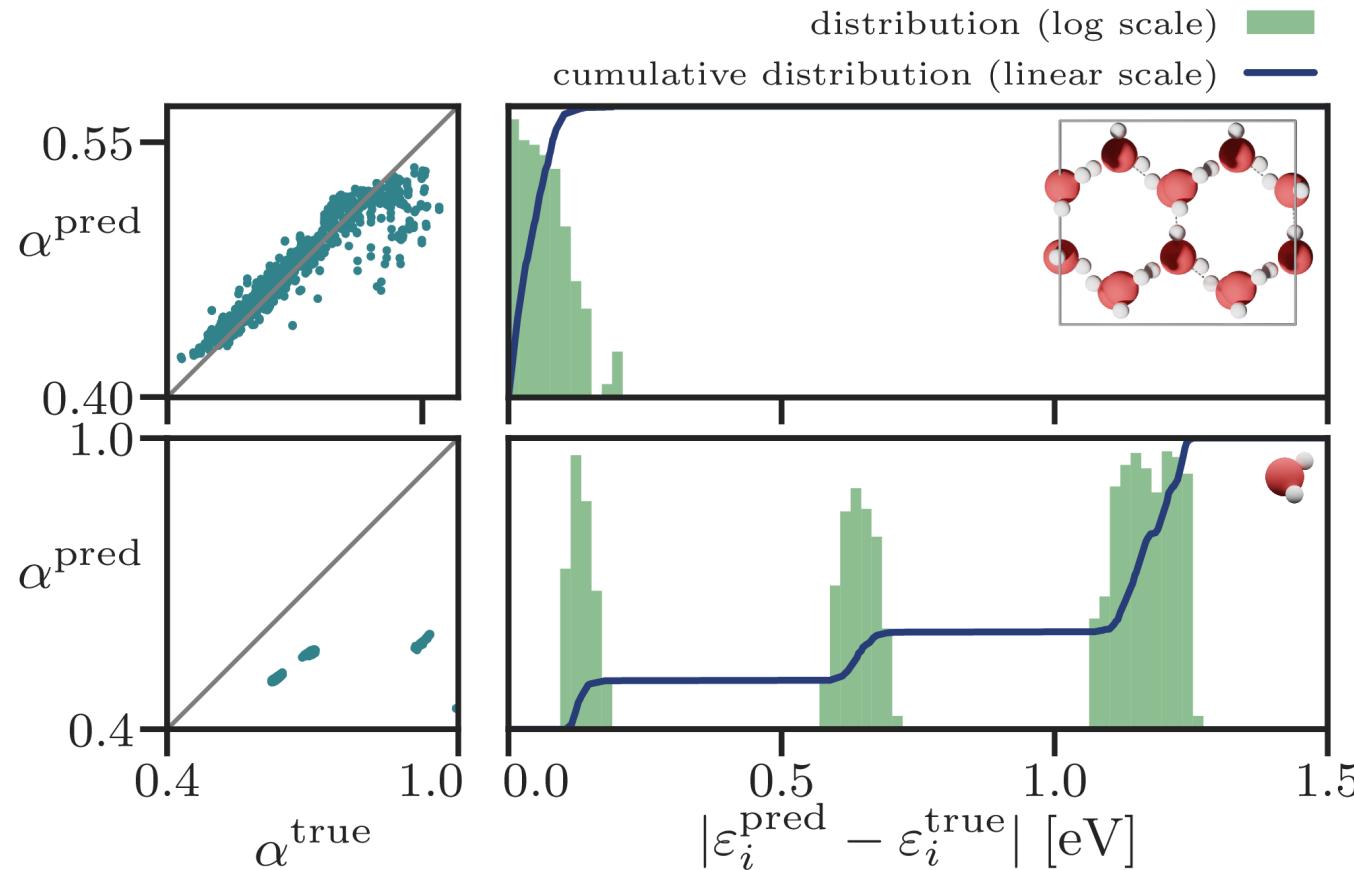
accurate to within $\mathcal{O}(10 \text{ meV})$ cf. typical band gap accuracy of $\mathcal{O}(100 \text{ meV})$;
 ridge-regression on one snapshot more accurate than oneshot

Speedup

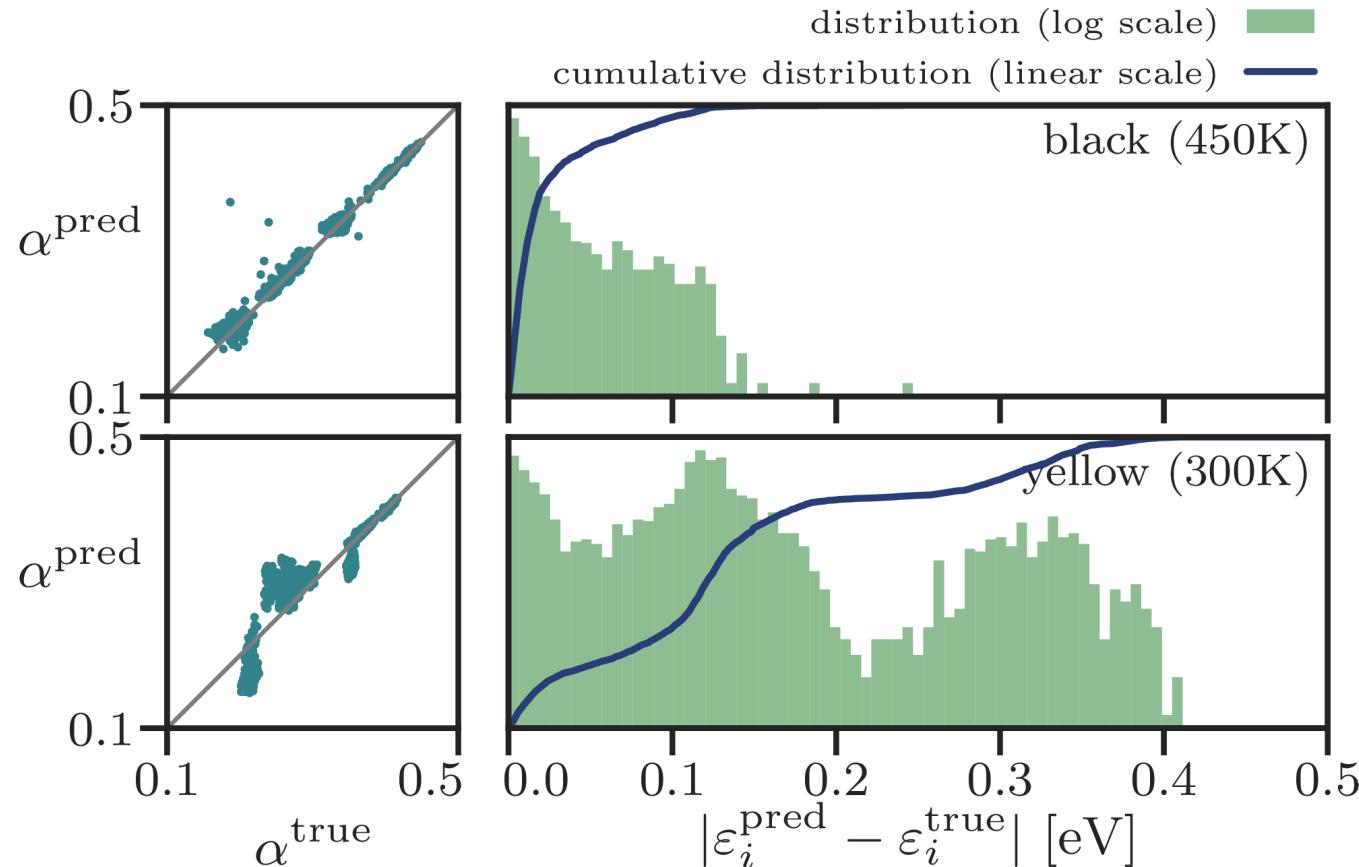


speedup of $\mathcal{O}(10)$ to $\mathcal{O}(100)$

Transferability (or lack thereof)



Transferability (or lack thereof)



Integrated in koopmans



- new block in the input file
- simple control of training/testing/predicting
- generates the ML model as a .pkl file for use in subsequent calculations

```
"ml": {  
    "predict": true,  
    "model_file": "cssni3_10_snapshot_model.pkl",  
    "n_max": 6,  
    "l_max": 6,  
    "r_min": 0.5,  
    "r_max": 4.0,  
    "estimator": "ridge_regression",  
    "descriptor": "orbital_density",  
    "occ_and_emp_together": true  
},
```

Conclusions

- lightweight machine-learning models can predict Koopmans screening parameters with high accuracy
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¹Y. Schubert *et al.* <http://arxiv.org/abs/2406.15205> (2024)

Conclusions

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- for more details see our arXiv preprint¹

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References



- N. L. Nguyen, N. Colonna, A. Ferretti & N. Marzari. Koopmans-Compliant Spectral Functionals for Extended Systems. *Physical Review X* **8**, 21051 (2018)
- R. De Gennaro, N. Colonna, E. Linscott & N. Marzari. Bloch's Theorem in Orbital-Density-Dependent Functionals: Band Structures from Koopmans Spectral Functionals. *Physical Review B* **106**, 35106 (2022)
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- N. Colonna, R. De Gennaro, E. Linscott & N. Marzari. Koopmans Spectral Functionals in Periodic Boundary Conditions. *Journal of Chemical Theory and Computation* **18**, 5435–5448 (2022)
- Y. Schubert, S. Luber, N. Marzari & E. Linscott. Predicting Electronic Screening for Fast Koopmans Spectral Functional Calculations. <http://arxiv.org/abs/2406.15205> (2024)