Predicting electronic screening for fast Koopmans spectral functionals

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Summary

- Koopmans functionals are powerful orbital-density-dependent functionals that predict spectral properties as accurately as state-of-the-art GW[1–4]
- they rely on parameters to capture electronic screening
- we construct a ML framework to predict these parameters
- minimal training data is required to achieve desirable accuracy

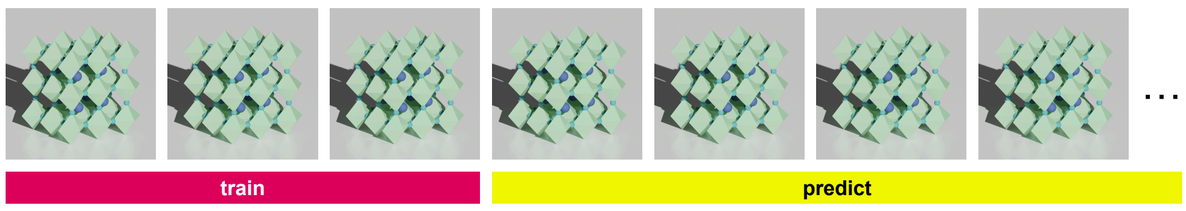
1. What are screening parameters?

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

- can be computed *ab initio*[5–7]
- are the vast majority of Koopmans' computational cost
- must be accurate; if $\psi_i(\boldsymbol{r}) = \sum_j U_{ij}\varphi_j(\boldsymbol{r})$ then

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

2. How can machine learning help?



or train on a small cell and deploy on a larger cell (N.B. not a general-purpose model)

3. Our machine learning framework

$$\rho_i(\boldsymbol{r}) \to p^i_{n_1 n_2 l k_1 k_2} \to \alpha_i$$

Descriptors are power spectrum decompositions[8,9] of orbital densities

$$p^i_{n_1 n_2 l, k_1 k_2} = \pi \sqrt{\frac{8}{2l+1}} \sum_m c^{i*}_{n_1 l m, k_1} c^i_{n_2 l m, k_2}$$

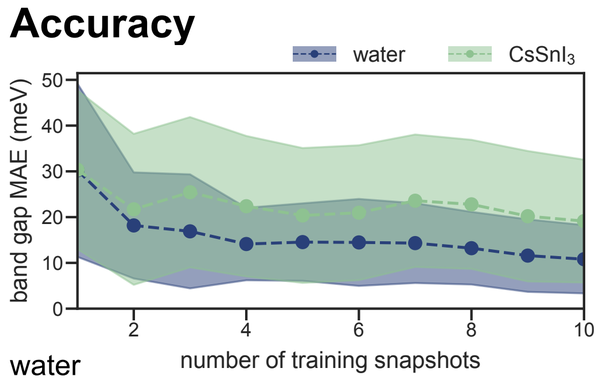
$$c^i_{nlm,k} = \int \mathrm{d}\boldsymbol{r} g_{nl}(r) Y_{lm}(\theta, \varphi) n_i(\boldsymbol{r} - \boldsymbol{R}_i)$$

Network is just ridge regression!

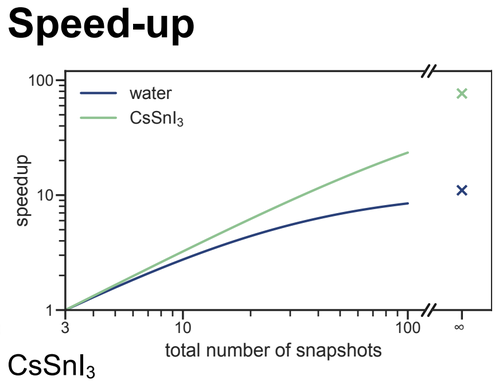
4. Results

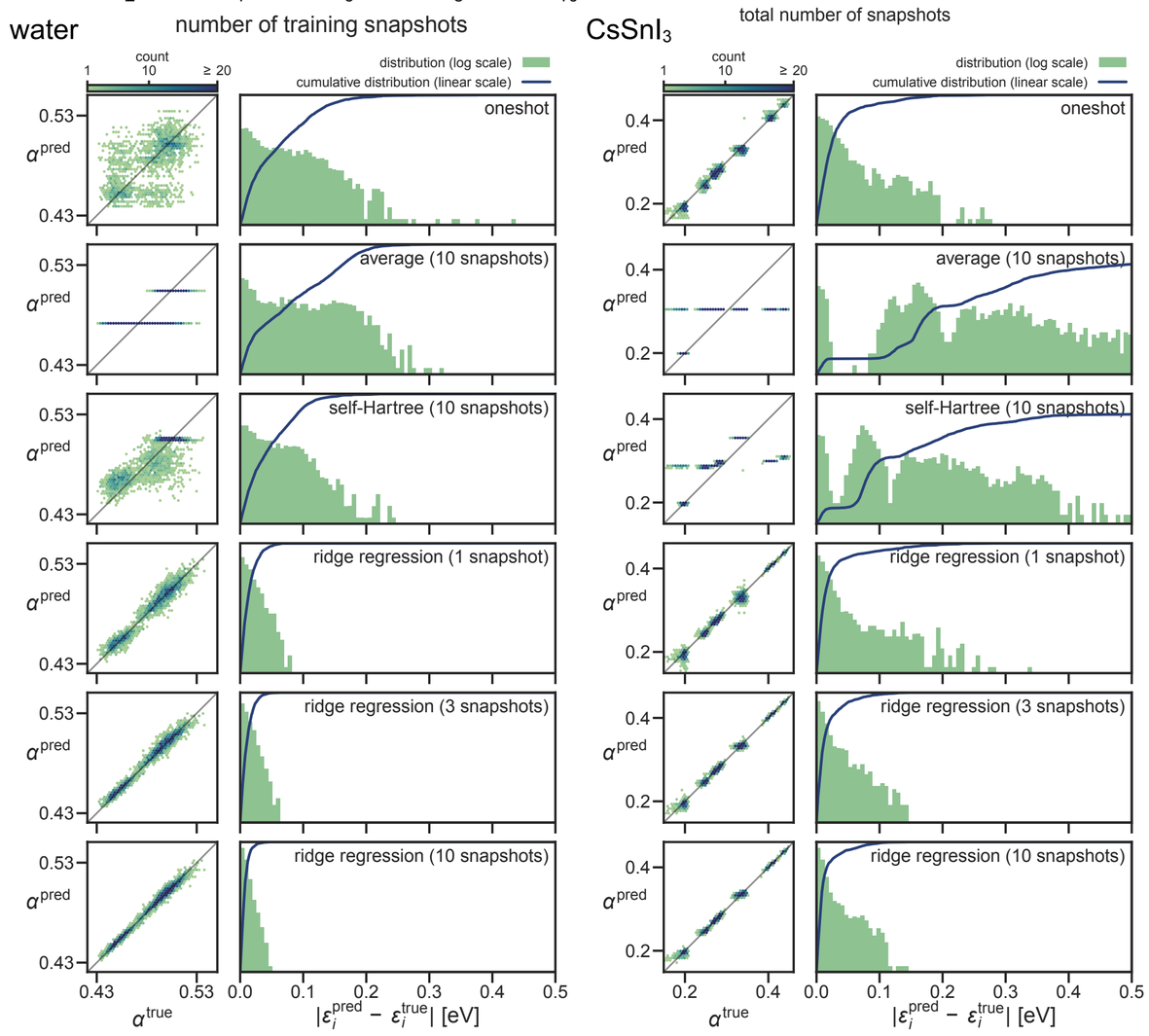
- accurate to $\mathcal{O}$(10 meV) *cf.* typical E_g accuracy of $\mathcal{O}$(100 meV)
- speed-ups from $\mathcal{O}$(10) to $\mathcal{O}$(100) times!
- ridge-regression on one snapshot more accurate than oneshot

Accuracy

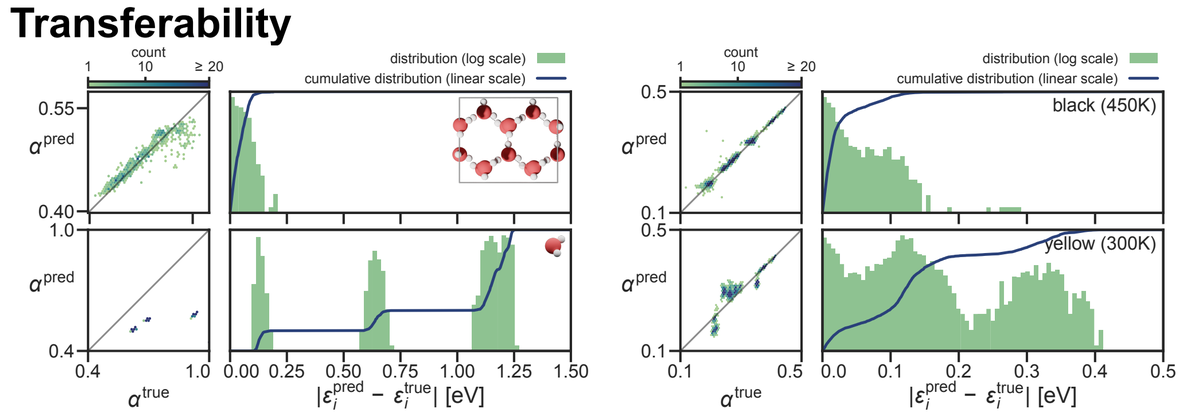


Speed-up





Transferability



5. Takeaways

- lightweight ML can predict Koopmans screening parameters
- more generally, predicting electronic response can be done efficiently with frozen-orbital approximations and ML
- try it now with `koopmans`! (`koopmans-functionals.org`)

References

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For more details, see Schubert *et al.*, npj Computational Materials (2024)

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