Machine-learning (ML) models with nearsighted force training (NFT) allow large-scale calculations at *ab initio* quality

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Background & Motivation

- ML models are effective in fitting potential energy surfaces (PES's) created by *ab initio* methods.
- Although ML models enable large-scale simulations, it is impractical to create data at sizes comparable to ML predictions.

➔ The objective is to develop an active-learning framework to create small-size structures right from the target structure.

Theoretical Approach

- Nearsightedness or locality of finite-ranged ML models

$$E = \sum_i E_i \qquad E_i = E\left(\{\vec{R}_{ij}\}\right), \text{ where } |R_{ij}| < R_c$$

$$f_i = -\frac{\partial E}{\partial R_i} = -\sum_j^N \frac{\partial E_j}{\partial R_i} = -\sum_j^{R_{ij}<R_c} \frac{\partial E_j}{\partial R_i} = -\sum_j^{R_{ij}<R_c} \frac{\partial E\left(\{R_{jm}\}\right)}{\partial R_i}$$

- Nearsightedness or locality of *ab initio* methods (DFT)[1]

$$\lim_{R\to\infty} \overline{\Delta n}(r_0, R) = 0$$

- Localizing uncertainties to atoms via ensemble models [2]

atomic uncertainty $\delta_i = 2.58\sigma_f$

structure uncertainty $\delta = \max_i(\{\delta_i\})$

Algorithm

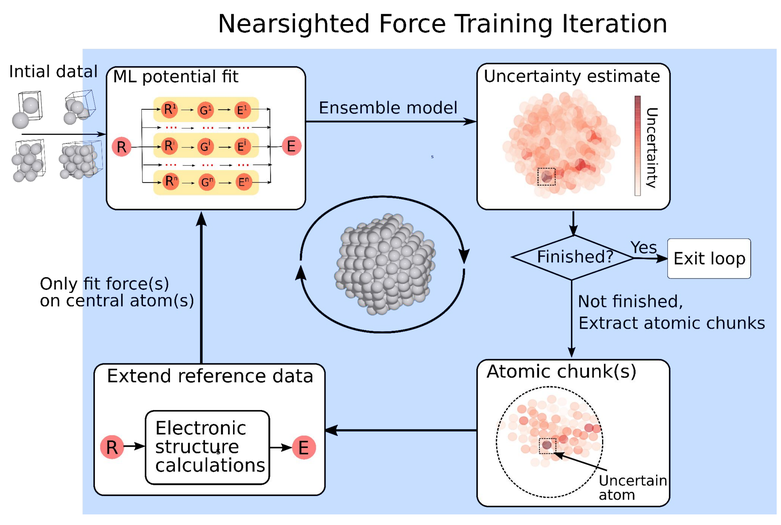


Figure 1: Schematic representation of the NFT framework.

Benchmark: A 260-atom Pt nanoparticle

DFT force locality analysis

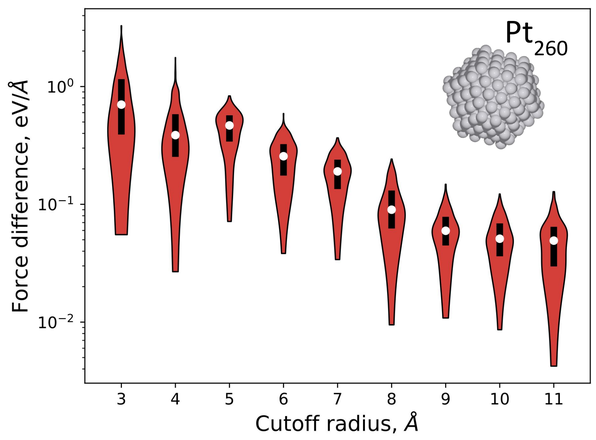


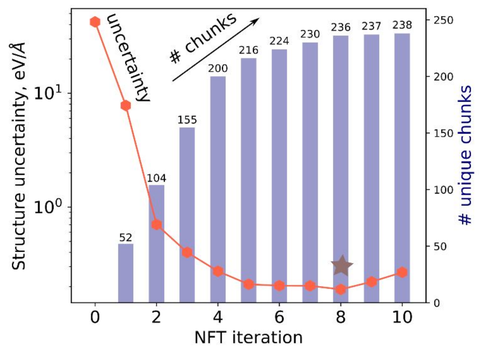
Figure 2: Distribution of force deviations between DFT true and local forces on Pt_{260} at various cutoffs.

- A cutoff of 8 Å gives a good balance between accuracy and efficiency. The maximum and mean deviations are 0.24 and 0.1 eV/Å.

Figure 3: Variations of structure uncertainty and number of chunks at each NFT iteration.

- The uncertainty gradually goes down with the increase of number of atomic chunks. The lowest uncertainty of 0.25 eV/Å is achieved with 8 NFT steps.

Nearsighted force training



Transferability to a larger system

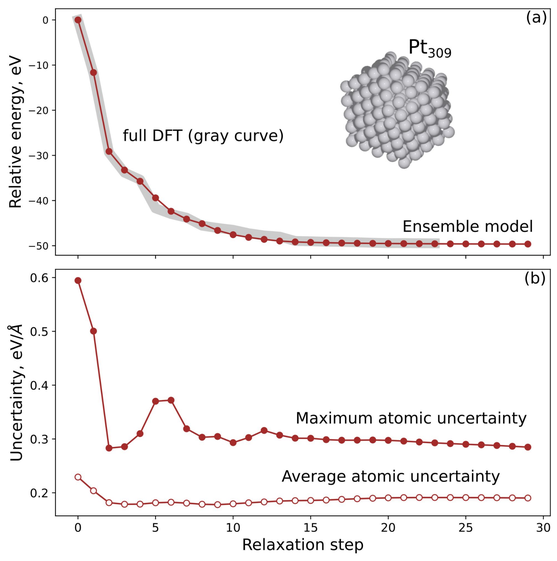


Figure 4: Structure optimization of Pt_{309} with the ensemble model trained on Pt_{260}. (a) energy profile. (b) Uncertainty propagation during relaxation.

- The energy profile by the ML model matches well with the full DFT results, despite not seeing the larger structure.
- After two steps, the structure relaxes to a familiar region, where the ML model can give predictions with high confidence.

Computational Cost

- Full DFT relaxation of Pt_{390} took about 28,680 hours. In contrast, DFT calculations for the ML model took 6207 hours. This represents a fairly dramatic savings (around 80% savings).
- The NFT is 'embarrassingly' parallelizable as jobs for different uncertain atomic chunks can be submitted individually.
- Our approach scales linearly with the system size *versus* the cubic scaling of DFT methods.

Conclusions

- A novel and robust ML framework has been developed, which explores PES's with minimum effort.
- An uncertainty metric is devised to probe the region on the PES's in need of new data.
- We demonstrated the NFT approach on a 309-atom structure on which we performed a structure optimization. The resulting ML models are robust despite not seeing the full-size structure.
- This ML framework is efficient because of its parallelizability and linear scalability.

References

[1] E. Prodan and W. Kohn, "Nearsightedness of electronic matter," Proceedings of the National Academy of Sciences, vol. 102, no. 33, pp. 11635–11638, 2005.

[2] A. A. Peterson, R. Christensen, and A. Khorshidi, "Addressing uncertainty in atomistic machine learning," Physical Chemistry Chemical Physics, vol. 19, pp. 10978–10985, May 2017.

Acknowledgements

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