Differentiable sampling of molecular geometries with uncertainty-based adversarial attacks

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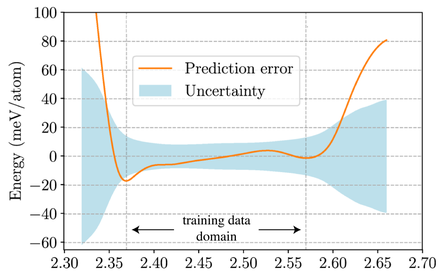
Uncertainty quantification to evaluate robustness of neural network potential

Neural network potentials (NNP) are unreliable outside of training data domain.

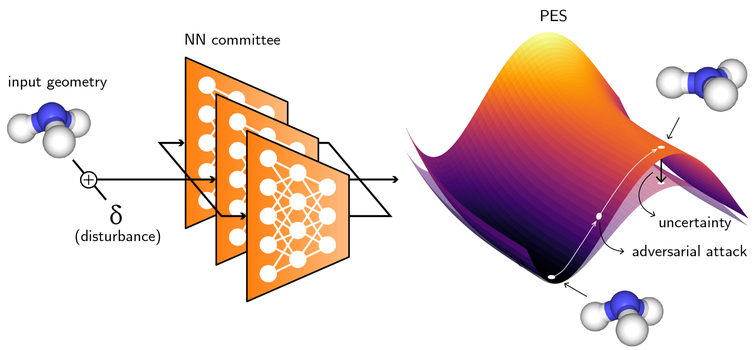
Increasing breadth of data is beneficial to improve NNP performance, but exhaustive exploration is very expensive.



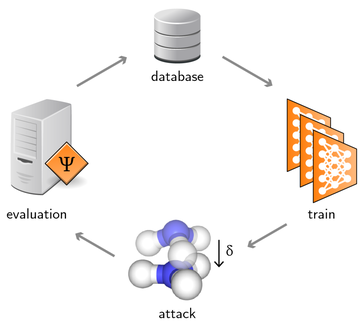
Therefore, assessing trustworthiness of NNP in configurational domains is essential. Quantifying uncertainty of models helps to distinguish unseen domains from training domains. [1,2]

Expanding training set with adversarial attacks on uncertainty of models

Finding local maxima of differentiable uncertainty metric



Adding new adversarial geometries in an active learning loop



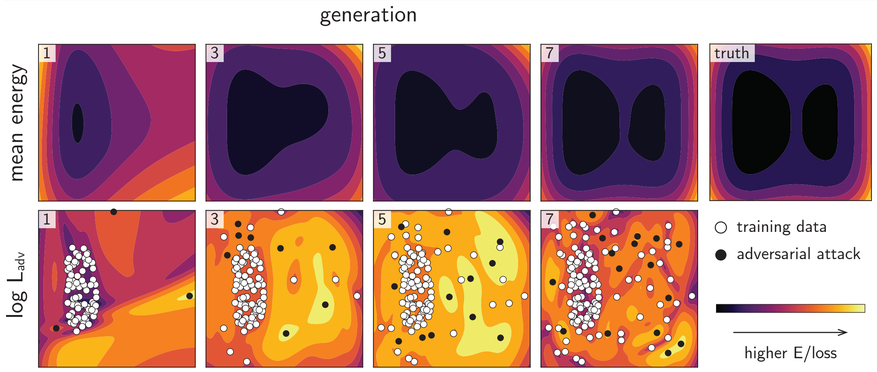
By backpropagating the adversarial loss through the NN committee, the displacement δ is updated using gradient ascent techniques.

$$\min_{\theta} \mathop{\mathbb{E}}_{\mathscr{D}} \left[\max_{\delta \in \Delta} \mathscr{L}_{adv} \left(\mathscr{D}_{\delta}; \theta \right) \right]$$

Convergence to local maxima of adversarial loss corresponds to geometries with high uncertainty and low energy. [3,4]

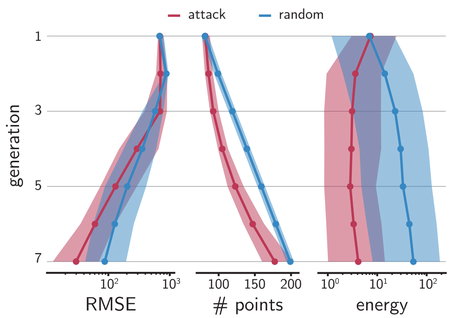
Expanding phase space in a double well potential

Predicting double well potential given only data in a single well



Adversarial sampling vs random sampling

Double well potential



Using the adversarial sampling strategy, median RMSE of final generation is two to three times lower than random sampling strategy, even though number of data points is smaller.

Lower energy range for adversarially sampled points: beneficial for real materials/molecular systems where high-energy configurations will not be visited in production simulations.

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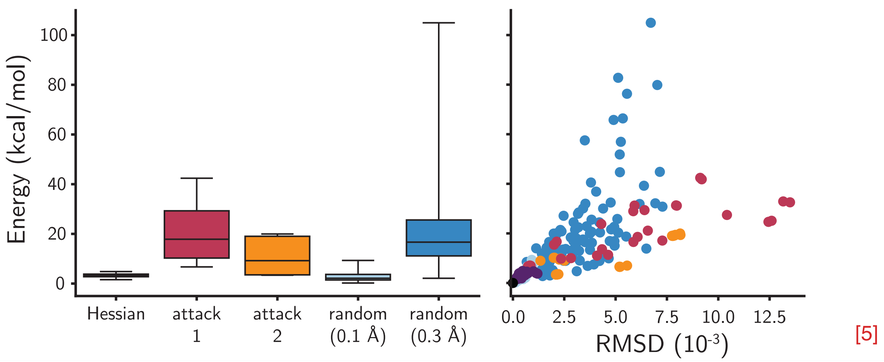
Random sampling: configurations have high energies but low diversity.
Adversarial sampling: configurations within reasonable energy range but high diversity.



[5]

Improvement in energy barrier prediction and stability of molecular dynamics simulation

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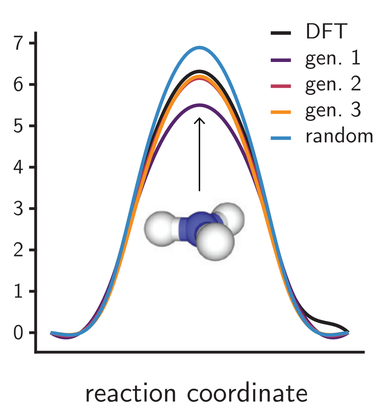

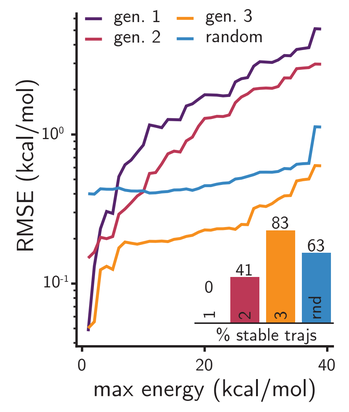

Summary

We propose a new sampling strategy for NNP by combining uncertainty quantification, automatic differentiation, adversarial attacks, and active learning. This strategy allows data-efficient NNP bootstrapping with parallel atomistic simulations to efficiently explore configurational space.

References

1. Xu et al. arXiv:2009.11848. (2020).
2. Venturi et al. J. Chem. Phys. A. 124, 5129-5126. (2020).
3. Szegedy et al. ICLR. (2014).
4. Goodfellow et al. ICLR. (2015).
5. K. Schütt et al. J. Chem. Phys. 148 (24) 241722 (2018).

Acknowledgements

DMSE Department of Materials Science & Engineering
MITei MIT Energy Initiative
AGC
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D. Schwalbe-Koda, A.R. Tan and R. Gómez-Bombarelli. arXiv:2101.11588. (2021).

http://bit.ly/AdversarialSampling