

# Supplementary Information

## Transfer learning for chemically accurate interatomic neural network potentials

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### 1 Molecular dynamics trajectories

Here, additional results for the aspirin molecule from the MD17 data set<sup>1–5</sup> are presented. Particularly, we study O–H and C–H distance distributions of the coupled-cluster aspirin data set<sup>4</sup> to explain the observed deviations for C–H and O–H characteristic modes. Fig. 1 and Fig. 2 represent the corresponding results for C–H and O–H distances. From Fig. 1 (left), we see that the respective O–H distances vary between 0.84 and 1.13 Å. However, the respective counts decrease by approaching the boundary values, indicating a somewhat worse sampling of high-energy regions.

From Fig. 1 (right), we observe a steeper potential energy surface for the model fine-tuned on energy values compared to the model trained on energies and forces of 950 configurations from scratch. The model fine-tuned on energy and forces of 128 configurations matches the latter well. Fitting the respective one-dimensional potential energy surfaces by a squared function, we could estimate vibrational frequencies of 3532, 3558, 3589, 3793  $cm^{-1}$  for the pre-trained model, the model trained from scratch and models fine-tuned on energy and force or energy labels, respectively. Note that the calculated values may strongly depend on the fitting procedure and serve only as a rough estimate. The obtained results support our observations in the main manuscript. Moreover, these results support the necessity of a thorough data set generation when fine-tuning with energy values only, e.g., better sampling of high-energy regions or augmenting data with more configurations and respective energy labels.

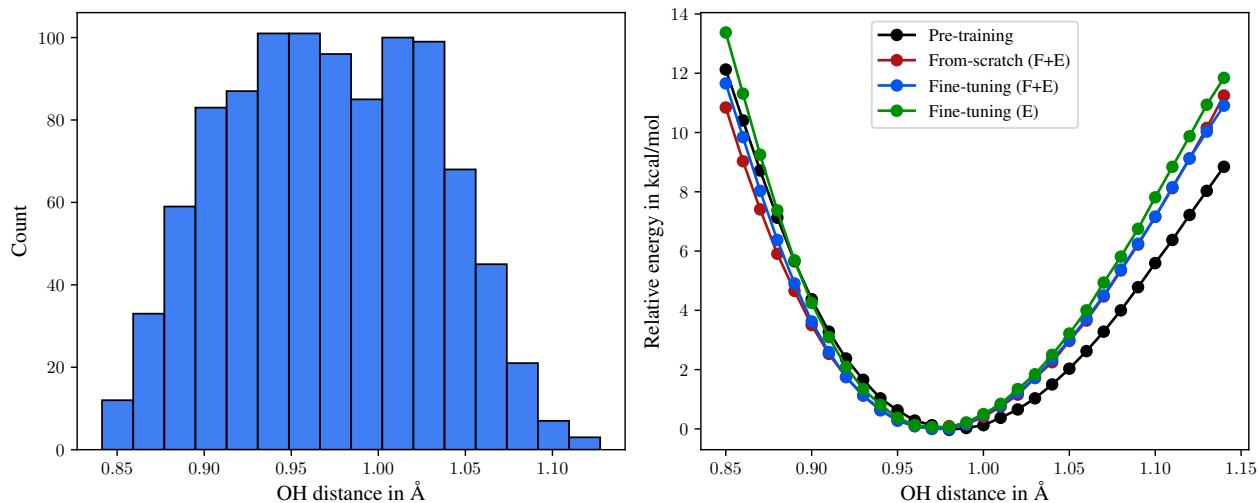
In contrast, from Fig. 2, we observe that the C–H distances have been slightly better sampled than O–H distances. In addition, the aspirin molecule has more C–H bonds than O–H bonds. This fact could explain why only minor deviations of fine-tuned models from those trained from scratch can be seen. The results in Fig. 2 match our observation that the C–H characteristic mode is predicted better than the O–H characteristic mode.

Fig. 3 shows the vibrational power spectrum of the aspirin molecule obtained by computing the Fourier transform of the velocity-velocity auto-correlation function sampled at 100 K. Fig. 4 and Fig. 5 compare power spectra obtained by running simulations with forces from interatomic potentials fine-tuned on 128 and 950 energy and atomic force labels.

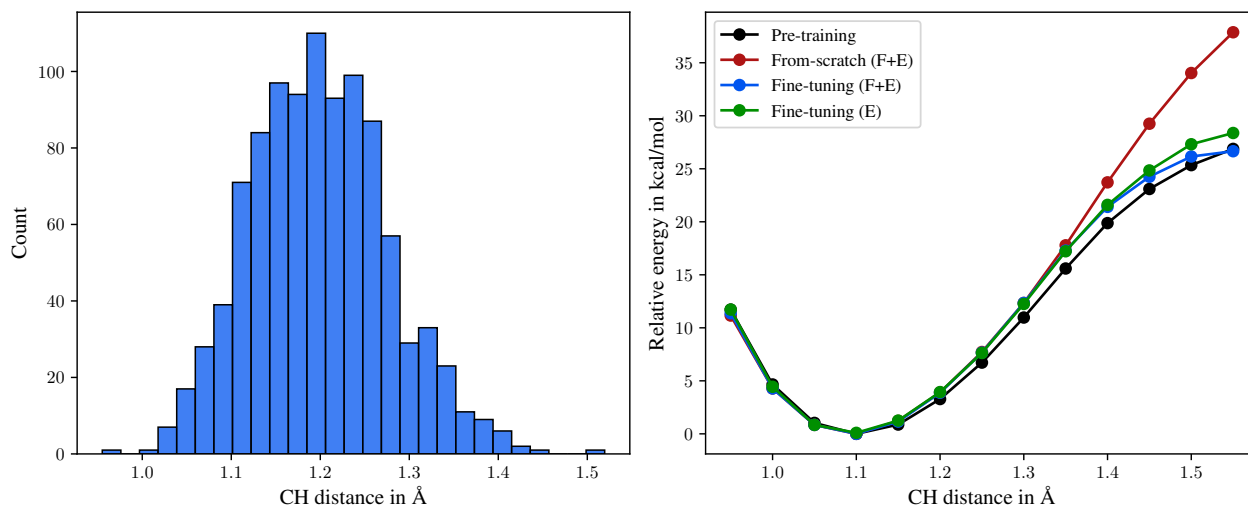
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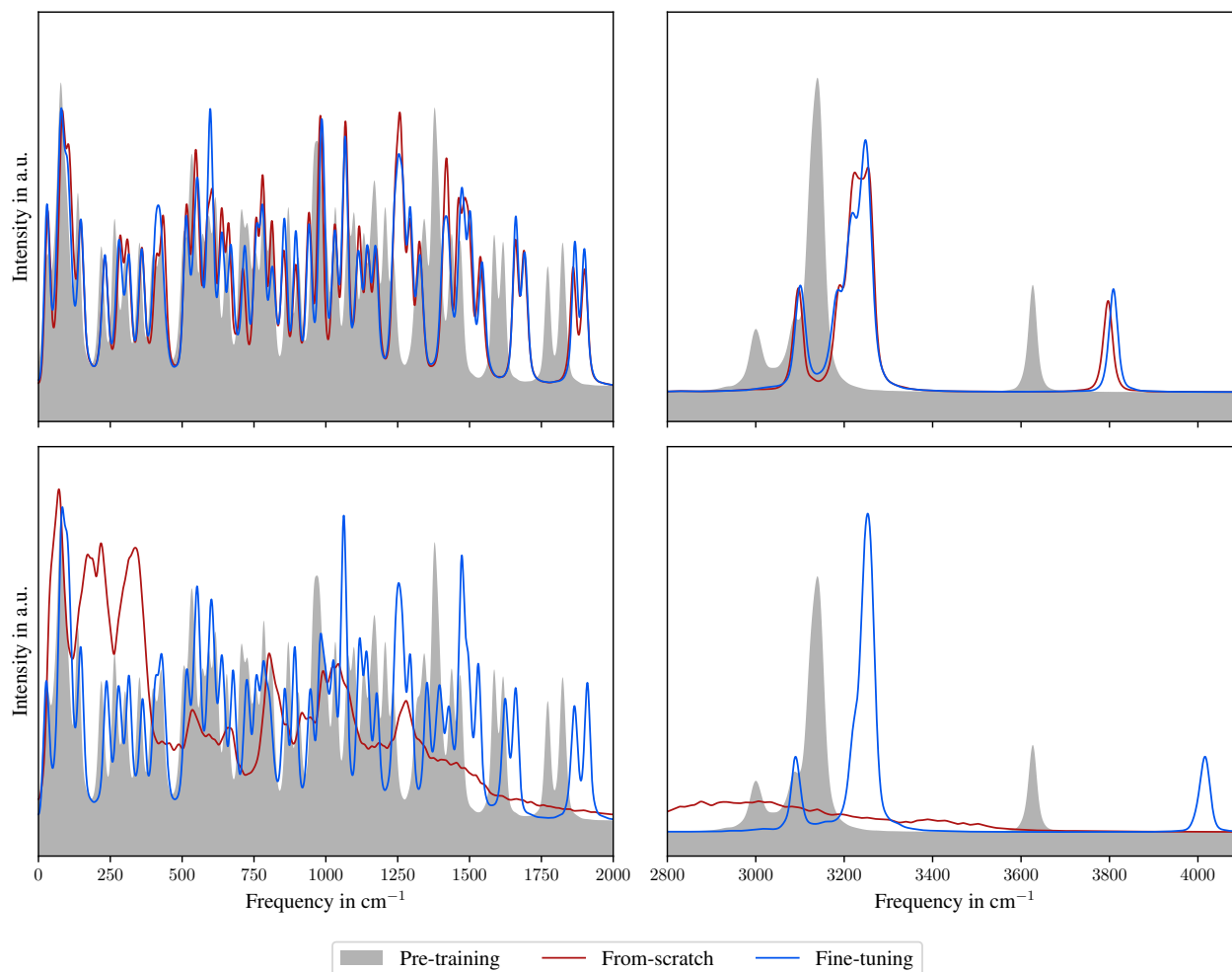
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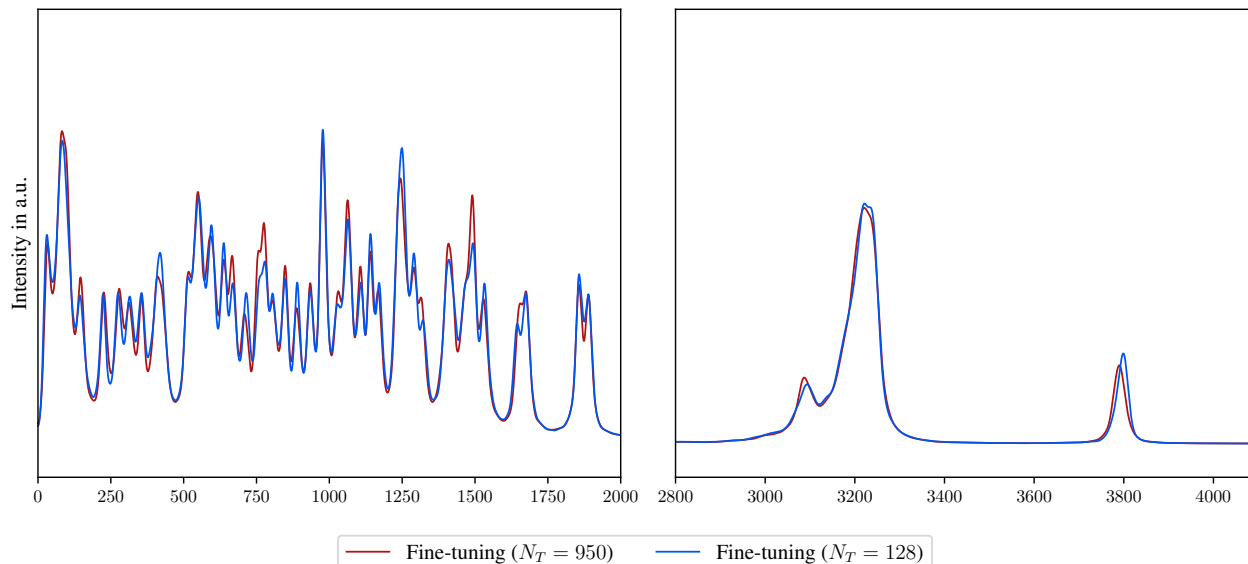
**Figure 1** (Left) Distribution of O–H distances in the coupled-cluster aspirin data set. (Right) Relative energy dependence on the O–H distance for four interatomic potentials.



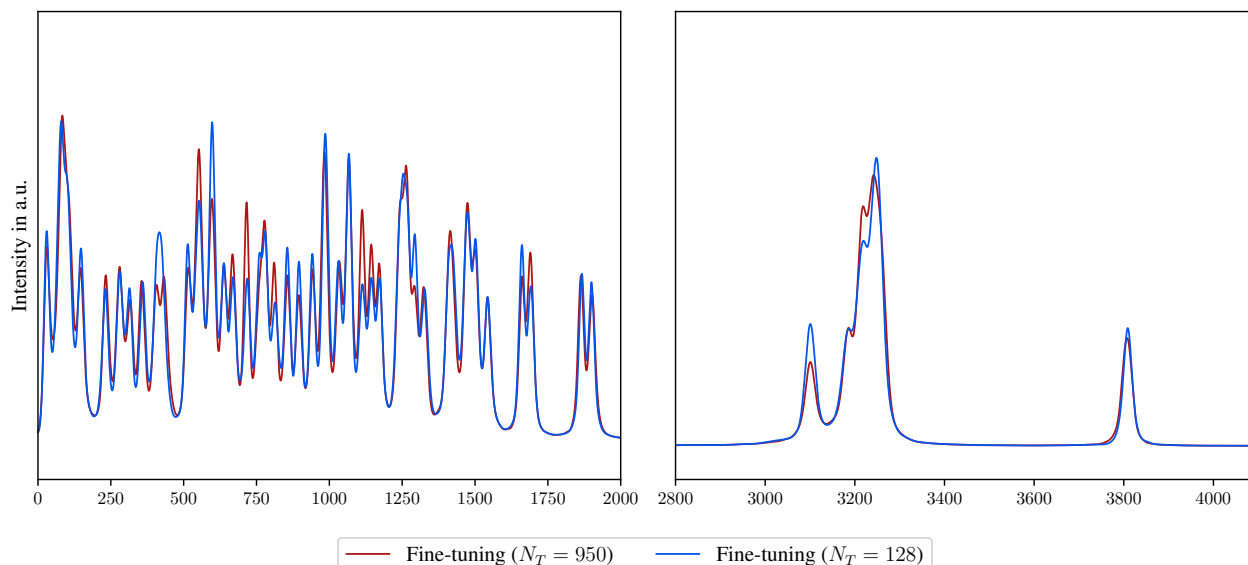
**Figure 2** (Left) Distribution of C–H distances in the coupled-cluster aspirin data set. (Right) Relative energy dependence on the C–H distance for four interatomic potentials.



**Figure 3** Vibrational power spectrum of the aspirin molecule obtained by computing the Fourier transform of the velocity-velocity auto-correlation function sampled at 100 K. (Top) Comparison of models trained from scratch on 950 and fine-tuned on 128 energy and atomic force labels. (Bottom) Comparison of models trained from scratch and fine-tuned on 950 energy labels only. The characteristic C-H and O-H peaks can be seen around  $3200\text{ cm}^{-1}$  and  $3800\text{ cm}^{-1}$ , respectively.



**Figure 4** Vibrational power spectrum of the aspirin molecule obtained by computing the Fourier transform of the velocity-velocity auto-correlation function sampled at 300 K. Comparison of models fine-tuned on 128 and 950 energy and atomic force labels. The characteristic C-H and O-H peaks can be seen around  $3200\text{ cm}^{-1}$  and  $3800\text{ cm}^{-1}$ , respectively.



**Figure 5** Vibrational power spectrum of the aspirin molecule obtained by computing the Fourier transform of the velocity-velocity auto-correlation function sampled at 100 K. Comparison of models fine-tuned on 128 and 950 energy and atomic force labels. The characteristic C-H and O-H peaks can be seen around  $3200\text{ cm}^{-1}$  and  $3800\text{ cm}^{-1}$ , respectively.

## Notes and references

- [1] S. Chmiela, A. Tkatchenko, H. E. Sauceda, I. Poltavsky, K. T. Schütt and K.-R. Müller, *Sci. Adv.*, 2017, **3**, e1603015.
- [2] K. T. Schütt, F. Arbabzadah, S. Chmiela, K. R. Müller and A. Tkatchenko, *Nat. Commun.*, 2017, **8**, 13890.
- [3] S. Chmiela, H. E. Sauceda, K.-R. Müller and A. Tkatchenko, *Nat. Commun.*, 2018, **9**, 3887.
- [4] H. E. Sauceda, S. Chmiela, I. Poltavsky, K.-R. Müller and A. Tkatchenko, *J. Chem. Phys.*, 2019, **150**, 114102.
- [5] A. S. Christensen and O. A. von Lilienfeld, *Mach. Learn.: Sci. Technol.*, 2020, **1**, 045018.