

Application-specific Machine-Learned Interatomic Potentials – Exploring the Trade-off Between Precision and Computational Cost

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Abstract

Machine-learned interatomic potentials (MLIPs) are revolutionizing computational materials science and chemistry by offering an efficient alternative to *ab initio* molecular dynamics (MD) simulations. However, fitting high-quality MLIPs remains a challenging, time-consuming, and computationally intensive task where numerous trade-offs have to be considered, e.g. How much and what kind of atomic configurations should be included in the training set? Which level of *ab initio* convergence should be used to generate the training set? Which loss function should be used for fitting the MLIP? Which machine learning architecture should be used to train the MLIP? The answers to these questions significantly impact both the computational cost of MLIP training and the accuracy and computational cost of subsequent MLIP MD simulations. In this study, we highlight that simultaneously considering training set selection strategies, energy versus force weighting, precision of the *ab initio* reference simulations, as well

as model complexity and computational cost of MLIPs can lead to a significant reduction in the overall computational cost associated with training and evaluating MLIPs.

This opens the door to computationally efficient generation of high-quality MLIPs for a range of applications which demand different accuracy versus training and evaluation cost trade-offs.

1 Introduction

Understanding atomic motion is fundamental for determining the physical and chemical properties of materials. Molecular dynamics (MD) simulations have been pivotal to address this challenge with applications ranging from drug design to nanotechnology.^{1,2} Traditionally, MD simulation fell into one of two categories, either relying on empirical force fields to describe interatomic interactions that allow for long/large qualitative simulations with a cost that scales linearly with the number of atoms, or using *ab initio* quantum mechanical methods that enable small/short but very accurate simulations typically scaling cubically with the number of electrons. In the last decade, machine-learned interatomic potentials (MLIPs) have emerged as an alternative that promises near-quantum mechanical accuracy while scaling linearly with the number of atoms.^{3,4}

The most recent developments in the field have prioritized improving the accuracy of MLIPs by incorporating complex atomistic descriptors and sophisticated machine learning models. While such models can now achieve remarkable accuracy, their training requires substantial amounts of high-fidelity *ab initio* training data and their evaluation can be thousands of times more expensive than traditional force fields, leading to significant computational costs at both training and evaluation times.^{5–13} In contrast, other efforts prioritize applications such as high-throughput materials discovery, simulations of large atomic systems, or long timescale simulations, where minimizing the model’s training and evaluation costs is paramount, even at the expense of a decrease in accuracy.^{14–17} Finally, the development of “foundation” or “universal” models – highly complex MLIPs, often graph neural

networks trained across vast chemical spaces^{18–20} – raises questions about the continued need for optimizing application-specific potentials. However, these “universal” models often require fine-tuning for specific material systems to achieve high accuracy.^{21–23} Crucially, fine-tuning preserves the high computational cost associated with the complex architectures of these “universal” models, which can be orders of magnitude greater than simpler alternative MLIPs like the linear Atomic Cluster Expansion (ACE).²¹ Thus, for applications demanding both robustness and speed, tailoring less complex, optimized MLIPs remains crucial. This requires a systematic optimization of the application-specific cost / accuracy trade-off, considering the quality of the training set, the complexity of the model, and the training procedures, which is the central theme of this paper.

This paper explores the critical trade-off between accuracy and computational cost inherent in fitting and evaluating MLIPs. Fig. 1 conceptually maps the key factors that we investigate to navigate this trade-off. Starting with the choice of MLIP complexity dictated by application’s needs in terms of number of simulation, simulation size and timescale with respect to the available computational budget. The optimization involves balancing the MLIP’s predictive error (e.g., energy and force RMSE) against the computational costs for constructing the training set and evaluating the MLIP. The computational cost for generating the Density Functional Theory (DFT) training set itself depends on its precision, specifically it is limited by the choice of convergence parameters such as plane wave energy cut-off and k-point mesh sampling as well as the total number of atomic configurations in the training set. It is important to distinguish this numerical precision from the intrinsic accuracy in comparison to experiment dictated by the choice of exchange-correlation functional, which is not considered here. Instead, the MLIP is benchmarked against the fully-converged DFT results with a given functional. While requiring high numerical precision with stringent DFT settings is common,^{24–26} it incurs substantial computational cost. We demonstrate that utilizing reduced-precision DFT training sets can be sufficient provided the energy and force contributions are appropriately weighted during training. Furthermore, systematic sub-

sampling techniques can identify the most informative configurations, drastically reducing the required training set size. By considering these aspects alongside the choice of MLIP complexity (which governs the computational cost of evaluation), we perform a joint Pareto analysis, conceptually illustrated by the optimal surface in Fig. 1. Our findings reveal that it is possible to achieve near-optimal MLIP accuracy with small, lower-precision DFT training sets. This is especially true when using computationally efficient, reduced-complexity MLIPs. This underscores the substantial benefits of jointly optimizing model complexity, training set precision, and training set to generate application-specific MLIPs with superior accuracy/cost characteristics.

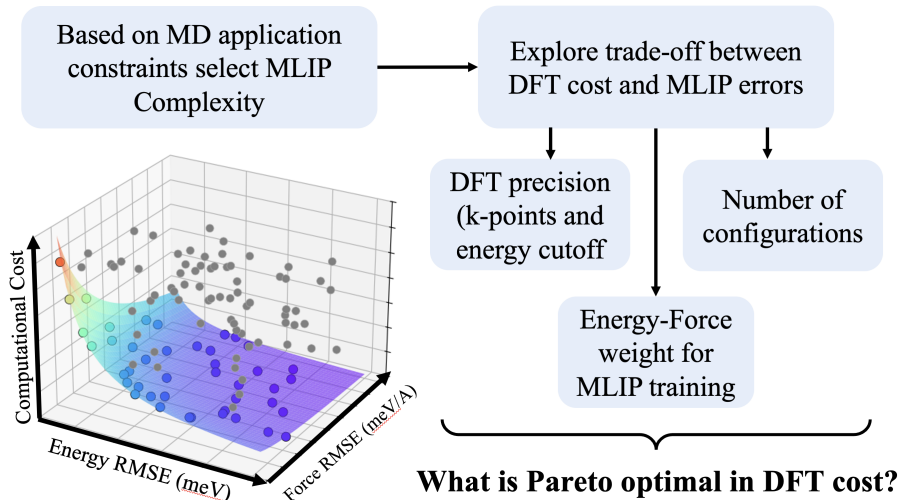


Figure 1: Application-specific MLIPs are constructed starting from desired application of the MLIP which restricts the computational costs per evaluation and consequently the **complexity** of the MLIP. This restricted MLIP **complexity** additionally impacts the benefit of increasing the training set size and *ab initio* precision of the training set. Finally, we identify the energy versus force weight as a central parameter to position the potential on the Pareto Front of computational cost, energy RMSE and Force RMSE.

The paper is organized as follows: Sec. 2 details the computational methodologies employed in this study. This includes the preparation of the DFT training set across six distinct precision levels (Sec. 2.1), the description of the Spectral Neighbor Analysis Potential (SNAP) formalism and its quadratic extension (qSNAP), detailing variations in model complexity and the training procedure involving energy and force weighting (Sec. 2.2), and

the leverage score technique utilized for efficient data sub-sampling (Sec. 2.3). Sec. 3 presents and discusses our findings. We first quantify the nature and magnitude of errors introduced by varying DFT precision levels (Sec. 3.1). Followed by analyzing how these DFT errors propagate into the trained MLIPs (Sec. 3.2). Next, we explore the effects of energy-force weighting (Sec. 3.3) and training set size (Sec. 3.4) on MLIP errors. Subsequently, we perform a multi-objective optimization, integrating DFT precision, training set size (informed by leverage sampling), energy-force weighting and MLIP complexity to map out the Pareto front of accuracy versus computational cost (Sec. 3.5). Sec. 4 discusses the implications of these findings and quantifies the reduction in computational cost. Finally, Sec. 5 summarizes the key conclusions drawn from this work and discusses their implications for the fitting of application-specific MLIPs by inverting the parameter selection to tailor to specific application requirements for accuracy and computational cost.

2 Methods

2.1 DFT Training Set

We generated a training and testing set of atomic Beryllium configurations using the information entropy maximization approach introduced in Refs. 27–29. This technique autonomously generates diverse datasets by creating atomic configurations that maximize the information entropy of the dataset in a feature space while bypassing the need for manual dataset curation by human experts. The MLIPs fitted to such diverse datasets were shown to be extremely robust and transferable.^{28,29} We rely on a subset of 20,000 configurations selected from the dataset introduced in Ref. 28, which was uniformly rescaled from the equilibrium lattice constant of Tungsten to the equilibrium lattice constant of Beryllium. This approach leverages the fact that both systems are unary, allowing the reuse of diverse geometric configurations from the entropy maximization algorithm without repeating the generation step for Beryllium. Each configuration contains on average 50 atoms.

When training MLIPs, the dataset is split evenly into a training and a testing set of 10,000 configuration each.

Table 1: Six Levels of DFT Precision are introduced, referenced as precision level 1 to 6, with different k-point mesh samplings and plane-wave energy cut-offs, resulting in different averaged evaluation run times per configuration.

Precision Level	k-point spacing, \AA^{-1}	Energy cut-off, eV	Average run time per configuration, sec
1	Gamma Point only	300	8.33
2	1.00	300	10.02
3	0.75	400	14.80
4	0.50	500	19.18
5	0.25	700	91.99
6	0.10	900	996.14

Reference energies and forces are calculated using pyiron workflow manager³⁰ and Vienna Ab initio Simulation Package (VASP)^{31,32} at six levels of DFT precision, as shown in Table 1. The calculations employed the Projector Augmented-Wave (PAW) method in conjunction with the Perdew-Burke-Ernzerhof (PBE) exchange-correlation functional. To model electronic smearing, we utilized the Methfessel-Paxton scheme (ISMEAR=1) with a smearing parameter of SIGMA=0.2 eV. The table also reports the average simulation time for single point evaluations using a single NVIDIA A100 GPU; the computational effort required to generate the reference data is seen to vary by a factor of around 100 between low (level 1) and high (level 6) precision simulations.

The per-atom energy distribution (Fig. 2a) spans from -3.75 to -2 eV/atom. Although this dataset is challenging to fit due to its diversity, it has been shown to produce highly transferable and robust potentials. The force component distribution (Fig. 2b) shows a standard deviation of 0.44 eV/ \AA and ranges from -10 to 10 eV/ \AA , with most values concentrated between -2 and 2 eV/ \AA .

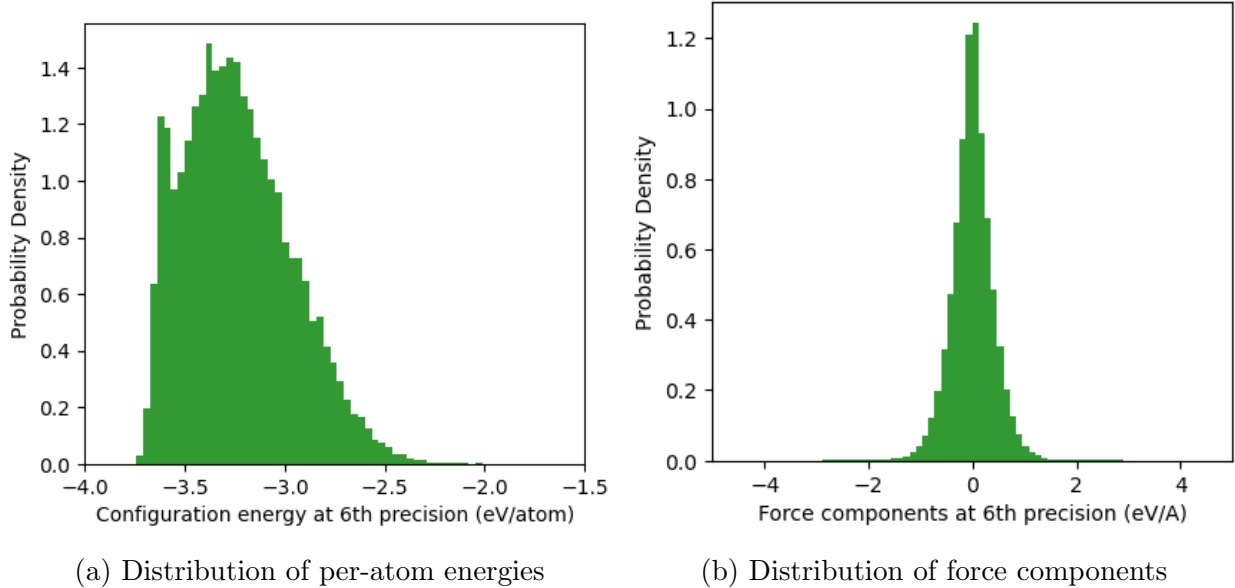


Figure 2: Probability density of configuration energies and forces in the entropy maximized dataset for Beryllium evaluated at precision level 6 (highest).

2.2 ML Potential

The Spectral Neighbor Analysis Potential (SNAP) formalism expands atomic energies and forces using linear combinations of Bispectrum components derived from 4D spherical harmonics, providing rotationally invariant descriptors of local atomic environments.⁶ In the following, we employ its quadratic extension, qSNAP, which incorporates quadratic bispectrum terms, improving the accuracy for complex bonding environments while maintaining computational efficiency.³³ qSNAP descriptors were obtained using FitSNAP software.³⁴ It is important to note that the approaches described below are general and applicable to other functional MLIP forms. In the present context, the use of qSNAP allowed for an extensive exploration of the tradeoffs explored in this study due to the low cost of training each model.

To study the benefits of increased DFT precision for MLIPs with varying levels of complexity, we control the MLIP complexity for the qSNAP MLIP via the $2J_{\max}$ parameter, which corresponds to increasing the angular order of the spherical harmonic expansion. Table 2 reports how increasing $2J_{\max}$ affects the number of descriptors, which corresponds to the number of MLIP coefficients and the computational cost of evaluation for applications,

like calculating MD trajectories with the MLIP. Notably, the evaluation cost varies by nearly an order of magnitude between $2J_{\max} = 4$ and 10, highlighting the direct impact of model complexity on computational cost for the application of MLIP.

Table 2: Number of bispectrum components (descriptors) and computational speed for increasing values of $2J_{\max}$. Taken from Wood and Thompson, 2018.³³

$2J_{\max}$	Number of Descriptors	Atoms-timestep per second
4	105	4×10^5
6	465	1×10^5
8	1540	4×10^4
10	4186	2×10^4

In spite of what its name suggests, training a qSNAP MLIP can be cast as a linear regression task, which greatly facilitates the training of the large number of models which are presented below. In the following, for simplicity we consider only regression to reference energies and forces, although other quantities such as stresses can be added.

The training of the linear model is based on minimizing a weighted least squares loss function:

$$L = \sum_{m=1}^M \left\{ \frac{w_E^2 (\hat{E}_m - E_m)^2}{N_m^2} + \sum_{i=1}^{3N_m} w_F^2 (\hat{F}_{mi} - F_{mi})^2 \right\} \quad (1)$$

where M is the number of configurations, m indexes a particular configuration, N_m represents the number of atoms in configuration m , and i refers to an atomic force component. The reference energies and forces are denoted as E_m and F_{mi} , while their predicted counterparts are \hat{E}_m and \hat{F}_{mi} , respectively. The terms w_E and w_F are weights assigned to energy and force contributions.

The minimizer of this loss function can be expressed as a weighted least squares problem of the form:

$$Wy = WX\beta \quad (2)$$

where $X \in \mathbb{R}^{n \times p}$ is the descriptor matrix with n representing the total number of energy and force components in the dataset, and p being the number of qSNAP descriptor components, $y \in \mathbb{R}^n$ is the vector of reference values (including both energies and forces), and $\beta \in \mathbb{R}^p$

is the vector of MLIP coefficients. The diagonal weight matrix $W \in \mathbb{R}^{n \times n}$ shown in Eq. 1 provides a user-adjustable relative weighting of energy and force terms in the loss function.

$$W = \begin{bmatrix} \frac{w_E}{N_1} & 0 & 0 & 0 & \cdots & 0 \\ 0 & w_F & 0 & 0 & \cdots & 0 \\ 0 & 0 & w_F & 0 & \cdots & 0 \\ 0 & 0 & 0 & \frac{w_E}{N_2} & \cdots & 0 \\ \vdots & \vdots & \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & 0 & 0 & \cdots & w_F \end{bmatrix}. \quad (3)$$

This formulation ensures that minimizing the weighted least squares problem is equivalent to minimizing the loss function defined in Eq. 1. It will be shown below that the choice of weights w_E and w_F plays a critical role in balancing the influence of energy and force errors and depends on both the complexity of the MLIP model and the precision of the training set (Sec. 3.3).

2.3 Training Set Sub-sampling with Leverage Score

A key aspect of the MLIP design challenge is the curation of the training set. Indeed, depending on the complexity of the MLIP, less than 10,000 configurations could be sufficient to obtain a converged MLIP even when using a high-diversity dataset.^{27–29} This begs the question of how to choose a proper subset of configurations to evaluate with DFT. In the following, we use a leverage score based strategy. Leverage quantifies how much a configuration’s features in descriptor space deviate from the average, allowing us to identify configurations with distinctive features. It can also be interpreted in terms of the sensitivity of the i^{th} predicted value \hat{y}_i on the i^{th} dependent value y_i (where the y ’s can be either energies of a particular configurations or force components of a particular atom). High-leverage points therefore have the potential to significantly affect predictions carried out with the trained MLIP. This concept is closely related to the maximum volume approach

or D-optimality criterion used in active learning, which selects data points that expand the coverage in descriptor space.³⁵

The leverage score of data item i is the corresponding diagonal element of the so-called hat matrix $H = X(X^T X)^{-1} X^T$. A numerically stable and efficient procedure to evaluate the leverage involves the singular value decomposition (SVD) of $X = U \Sigma V^T$. From basic properties of the SVD, it can easily be shown that:

$$\begin{aligned} H &= X(X^T X)^{-1} X^T = U \Sigma V^T (V \Sigma U^T U \Sigma V^T)^{-1} V \Sigma U^T = U \Sigma V^T (V \Sigma^2 V^T)^{-1} V \Sigma U^T \\ &= U \Sigma V^T V^T \Sigma^{-2} V^T V \Sigma U^T = U \Sigma V^T V \Sigma^{-2} V^T V \Sigma U^T = U U^T \end{aligned} \quad (4)$$

We explore two strategies for estimation of the configuration’s leverage score: either utilizing the leverage score derived only from one row of data (solely from the energy), a method analogous to CUR decomposition,³⁶ or calculating a total leverage score by summing the leverage scores from all rows in the block of data corresponding to energy and $3N$ force components, an approach analogous to Block CUR decomposition.³⁷

3 Results and Discussion

3.1 Quantifying Errors in Low-Precision DFT Calculations

Understanding the errors introduced by different levels of DFT precision requires recognizing the distinct convergence behaviors of the primary convergence parameters: the plane-wave energy cutoff (ENCUT) exhibits monotonic energy decrease governed by the variational principle, whereas k-point sampling, approximating Brillouin zone integration, shows generally non-monotonic oscillatory convergence. We characterize the statistical properties of these errors observed in our dataset, providing a guide for the development of the fitting strategies introduced below. Fig. 3 compares DFT calculations at the 2nd and 6th precision levels, showing energy relationships (Fig. 3a) and force relationships (Fig. 3b). Both quantities show different statistical behaviors. While the distribution of force errors is centered at zero and

symmetric, suggesting that low-precision forces could potentially be considered as “noisy” versions of exact forces, to a first approximation. Points in Fig. 3 are color-coded by the shortest cell vector length, highlighting that energy errors appear to be significantly affected by the largest k-point spacing along any cell dimension, suggesting that insufficient coverage in the Brillouin zone leads to systematic errors. Force errors don’t show the same bias as energies although they are broader for cells which are shorter in at least one direction, but the effect is much smaller. These trends persist at other precision levels, although the errors rapidly become smaller as precision increases, as shown in Supplementary Figs. ?? and ??. In addition, precision level 1 (lowest) energies show a systematic overestimation relative to level 6 (highest) (Fig. ??). This strong bias is much less pronounced for precision levels 2, 3, 4, and 5, where the energy errors are substantially smaller.

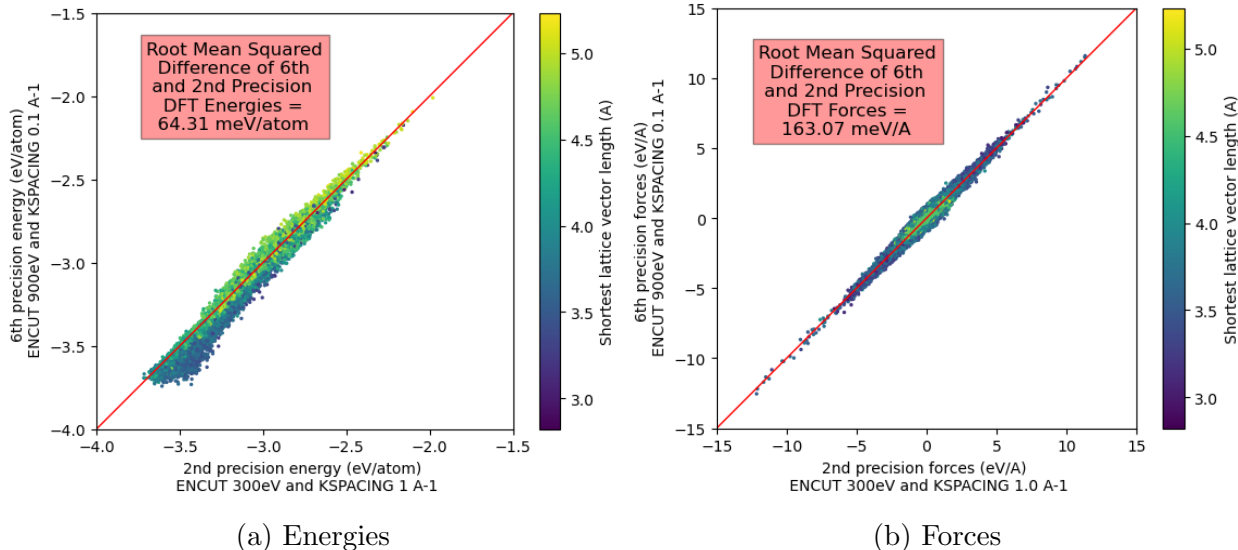


Figure 3: Pairwise relationships between 2nd and 6th precision level DFT data.

Error metrics are summarized in Table 3, which presents the root mean squared differences (RMSD) between the 6th precision level and other precision levels for the energies and forces. The “unshifted” column reports raw errors, while the “bulk shifted” and “mean shifted” columns account for potential systematic energy shifts by first matching the energy of the ground state structure (bulk shifted) or by first shifting all energies by their

mean (mean shifted). Although the energy errors significantly decreases after shifting, reflecting the asymmetric nature of the errors, a simple shift cannot correct for the observed energy errors. The consideration of this shift is motivated by the empirically-observed error cancellation resulting in the faster convergence of energy differences in comparison to the convergence of total energies.³⁸ Finally, we observe that precision level 5 is required to achieve energy errors lower than 1 meV, which is often regarded as a target for accurate MLIPs.

Table 3: Energy and Force Root Mean Squared Differences relative to the 6th precision level.

Precision level	Energy, meV/atom			Forces meV/Å
	unshifted	bulk shifted	mean shifted	
1	497.05	437.96	221.72	417.76
2	64.31	192.14	47.46	163.07
3	15.34	20.21	15.28	77.64
4	5.59	59.34	5.39	51.17
5	0.54	2.42	0.50	10.17

We note that an analysis in terms of pointwise averages omits very important properties of energy and force errors that differentiates them from statistical noise. First, actual errors are correlated in that small changes in the positions of atoms are likely to incur similar errors, leading to smooth local distortions with respect to a fully-converged potential energy surface. Second, errors can also be discontinuous, e.g., when the k-point mesh discretely changes as a simulation cell is smoothly distorted. As will be shown in the next section, the different statistical properties of the energy and force errors can be used to mitigate their impact and reduce error propagation into MLIP models.

3.2 Effect of DFT Precision Errors onto MLIP Training

To investigate the impact of errors introduced by lower-precision DFT simulation on the accuracy of MLIPs, we trained qSNAP potentials to half of the datasets (10,000 configurations) evaluated at the six precision levels and subsequently tested the models on the other half, using both the corresponding precision level or the highest precision level. In addition,

the effect of the relative weight of energies and forces is explored.

Tables 4 and 5 report the root mean squared errors (RMSE) values for energy and force errors for MLIP trained at different precision levels, with both higher energy weight ($w_E : w_F = 150 : 1$) and higher force weight ($w_E : w_F = 12 : 1$). Note that the datasets contain around 150 times more force components than energies as each configuration contains on average 50 atoms (see Sec. 2.1). Each row in the table corresponds to the precision level of the training set. The “testing on self” column reports the RMSE values when tested on the same precision level as the training set, while the “testing on 6th” column reports errors when tested on 6th (highest) precision level data. The ultimate goal is to obtain models with low errors when tested against the highest precision level. Testing errors are evaluated using both an unshifted potential and a shifted potential, where the latter includes a constant energy offset equal to the mean prediction error. Table 5 presents similar data for force RMSE values, with the exception of the absence of a shift correction.

Table 4: Energy root mean squared errors (meV/atom) for MLIPs trained to 10,000 configurations at different DFT precisions and with the highest level of complexity ($2J_{max} = 10$).

Prec. level	Higher energy weight				Higher force weight			
	Training	Testing on self	Testing on 6th		Training	Testing on self	Testing on 6th	
			unshifted	shifted			unshifted	shifted
1	111.74	117.91	473.42	164.02	187.84	188.55	458.17	104.85
2	30.90	33.18	53.76	32.17	39.26	40.06	50.21	25.85
3	11.01	11.67	11.08	10.99	15.12	15.48	9.24	9.14
4	6.20	6.58	5.80	5.62	8.84	9.01	8.16	8.04
5	4.79	5.06	5.05	5.05	7.70	7.84	7.85	7.85
6	4.77	5.05	5.05	5.05	7.70	7.84	7.84	7.84

Table 4 highlights that training errors are larger for MLIPs trained to lower precision data compared to higher precision, indicating that the potential energy surface becomes smoother and hence easier to fit as the precision increases. This can be related to the discussion above (see Sec. 2.1) where the impact of discontinuities and inconsistencies due to incompatible k-point meshes and limited plane wave energy cut-offs is expected to decrease with increasing precision, producing smoother and more internally-consistent potential energy surfaces, as

Table 5: Force root mean squared errors (meV/Å) for MLIPs trained to 10,000 configurations at different DFT precisions and with the highest level of complexity ($2J_{max} = 10$).

Prec. level	Higher energy weight			Higher force weight		
	Training	Testing on self	Testing on 6th	Training	Testing on self	Testing on 6th
1	931.09	944.15	900.29	375.83	379.2	242.75
2	235.72	238.89	186.67	189.33	190.69	116.66
3	143.30	145.03	124.53	130.66	132.06	108.57
4	120.46	121.86	111.50	116.4	117.67	106.89
5	109.41	110.87	110.51	105.71	106.98	106.6
6	109.04	110.52	110.52	105.32	106.6	106.6

commonly reported in the literature.^{38,39} Interestingly, we observe that testing errors are generally lower when models trained to lower precision data are tested on 6th precision data than on their own level of precision. This observation suggests that the inability of MLIPs to capture unphysical behavior such as energy discontinuities/inconsistencies due to discrete changes in k-point mesh sampling can, in fact, be an advantage, since it can be used to partially recover the behavior of smoother high-precision data. Supporting this interpretation, Supplementary Figs. ??, ??, ??, and ?? show strong correlations between energy and force residuals from MLIPs trained to low-precision data and actual DFT error between high- and low-precision DFT energies and forces, indicating that artifacts in the low-precision DFT energy surface are indeed partially corrected by the MLIP. This trend is especially evident when large force weights are used during training, as reflected in Table 4, where the lowest energy RMSE values for the 1st, 2nd, and 3rd precision levels occur at higher force weight. However, this is no longer true for the 4th and 5th precision levels, where DFT errors are small and the MLIP errors become limited by the model’s intrinsic ability to capture the full complexity of the dataset, as indicated by the error saturation when training to higher precisions.

3.3 Energy-Force-Weight Dependence

Fig. 4 illustrates how energy and force RMSEs are affected by energy versus force weights. In these plots, the relative weight of energies versus forces in the regression increases/decreases from the top left to the bottom right. The dotted line represents the RMSE when evaluated at the same precision level as the training and testing set, while the solid line shows errors evaluated on the testing set evaluated at the highest (6th) precision level.

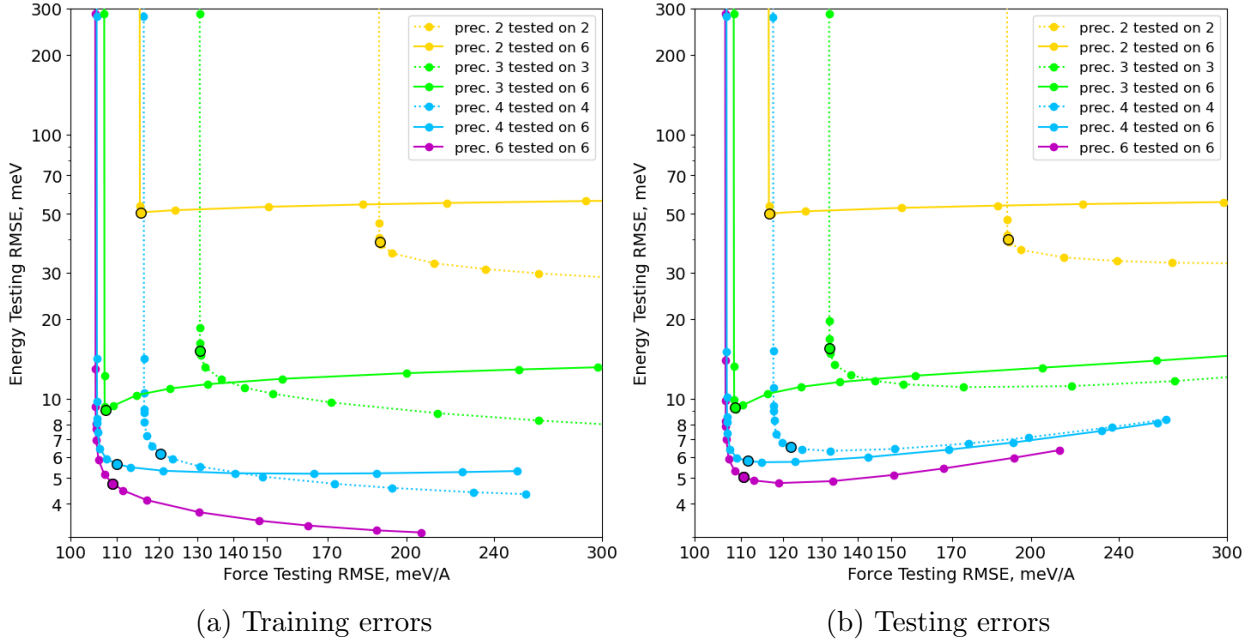


Figure 4: Pareto front of energy and force testing errors for different energy versus force weights in qSNAP fitting ($2J_{\max} = 10$) across various precision levels trained on 10,000 configurations. Precision level 5 is omitted as it visually overlaps with precision level 6. Precision level 1 data falls out of the range of these plots.

Perhaps counterintuitively, increasing the energy weight of precision levels 2 and 3 leads to *higher* training and testing energy errors when measured at the 6th precision level (solid line). In these cases, the Pareto front almost collapses to a single point which simultaneously provides the lowest energy and force errors. This outcome stems from the nature of errors in low-precision DFT calculations, caused by insufficient sampling of the k-point mesh and sharp features in the potential energy surface based on the restricted number of plane waves, effecting the total energy of the supercell. The forces are less sensitive and

exhibit faster convergence compared to energies with respect to the DFT convergence parameters like plane wave cut-off and k-point density.³⁹ This is also illustrated by the force error in Fig. 3b which is more symmetric and Gaussian-like, suggesting force-related errors less effected by a reduced plane wave energy cut-off or a reduced k-point mesh sampling, resulting in a statistically more well-behaved training set for learning compared to the errors in energy. Consequently, focusing training excessively on low-precision energies by increasing energy weight can lead the MLIP to partially learn the incorrect low precision potential energy surface, resulting in these systematic errors that distort the potential energy surface away from the high-precision reference. In contrast, by focusing more on forces, the MLIP can most efficiently "average-out" errors, effectively learning a smoother representation of the low-precision energy landscape which is closer to the fully converged high-precision potential energy surface. This suggests that leveraging the larger, statistically more robust force dataset via increased weighting can mitigate the impact of low-precision energy noise. Therefore, carefully adjusting the relative weighting of energies and forces is crucial when training MLIPs on lower-precision data, as increasing force weights can, perhaps paradoxically, produce MLIPs yielding better energies in comparison to the high-precision potential energy surface as well as better force convergence. Note, the increase in test energy errors observed specifically at precision levels 4 and 6 (right panel of Fig. 4) can be attributed to overfitting to the comparatively smaller number of energies, since the same behavior is not observed in training errors on the same level of precision.

In most applications that require simulating material properties that are not accessible with direct DFT simulations, the complexity of the MLIP is limited by the available computational resources e.g., the complexity of the MLIP is chosen based on the goal to achieve a fixed target in terms of the number MD simulation time steps required to investigate a given physical phenomenon of interest. The results presented in the previous section suggest that the impact of DFT errors varies based on the complexity of the MLIP, which provides the opportunity to reduce the DFT precision and increase the computational efficiency dur-

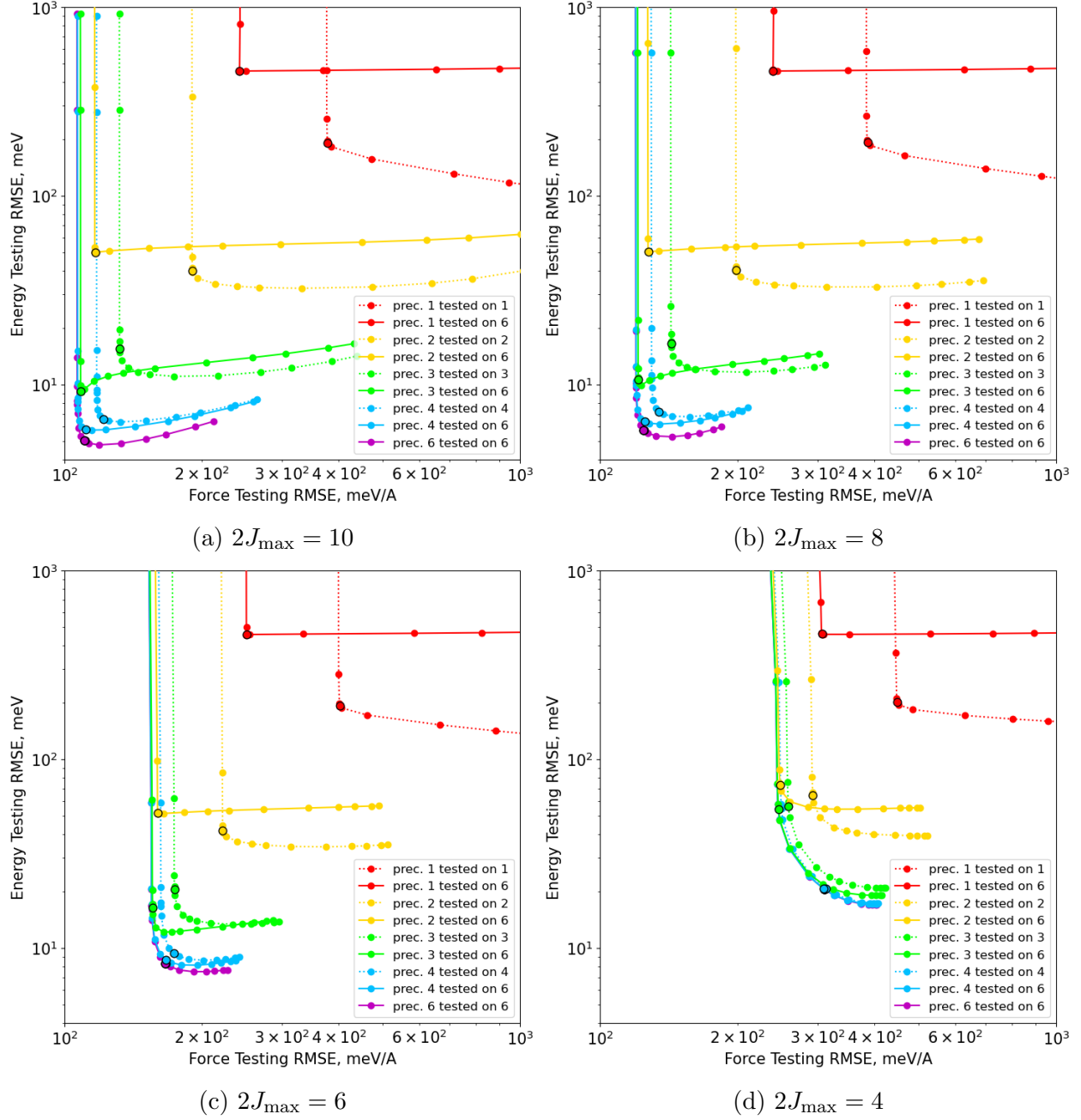


Figure 5: Energy/force Pareto front of testing errors with different energy versus force weights in the fitting and at different precision levels for various $2J_{\max}$ values fitted to a training set with 10,000 configurations.

ing the dataset curation. Indeed, highly complex MLIPs are expected to be more prone to learning the DFT errors compared to simpler models, and so will intrinsically require higher precision DFT training sets, while simpler models benefit from lower-precision training sets, reducing the computational cost to construct the DFT training set. To explore this, the previous analysis is repeated for different model complexities. For qSNAP potentials, this is achieved by varying the angular order of the bispectrum expansion, which is commonly referred as the $2J_{\max}$ parameter⁶ (See Sec. 2.2). As shown in Table 2, increasing $2J_{\max}$ corresponds to a rapid increase in the number of coefficients in the MLIP, and hence to an increase in complexity.

Fig. 5 shows that simple MLIPs are indeed less sensitive to the errors in low-precision DFT training sets. For example, for $2J_{\max} = 4$, the potentials trained to precision levels 3, 4, and 5 yield similar errors when tested on a precision level 6 testing set. In contrast more complex MLIPs with $2J_{\max} = 10$ show a more pronounced dependence on the training set precision level. This suggests that lower complexity MLIPs can leverage low-precision DFT training sets compared to higher complexity MLIPs, as they are less able to learn spurious features of the low-precision DFT potential energy surface.

3.4 Data Sub-sampling via Leverage score

A critical factor in efficiently curating training sets for MLIPs is the trade-off between number of configurations and the precision of the underlying DFT simulations to evaluate the configurations. Here, we explore the effect of data sub-sampling strategies on the accuracy of the obtained MLIP. In this respect, Fig. 6 highlights that leverage sampling significantly outperforms random sampling, in terms of decay rate of both the energy and force errors and of their variance with increasing number of configurations. Significant savings of computational cost are achieved, with only a few hundred configurations being required to consistently reach within 1 meV/atom of the result obtained when using all 10,000 configurations where the error is dominated by the finite expressivity of the MLIP. This results in a reduction of

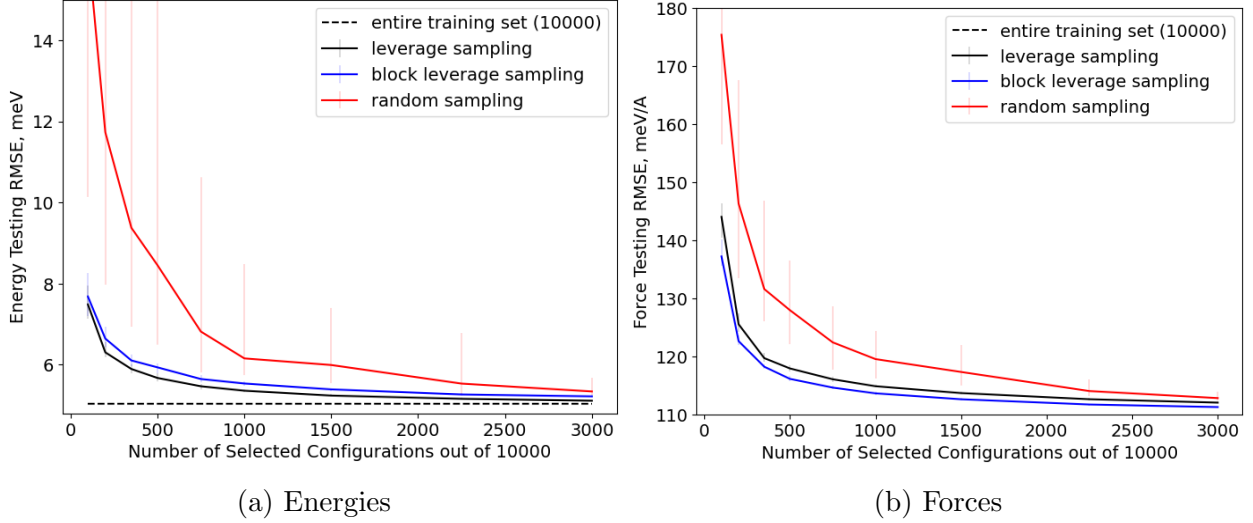


Figure 6: Comparison of regular and block leverage sampling with random sampling ($2J_{max} = 10$) for the highest level of DFT precision and a fixed energy versus forces weight ($w_E/w_F = 150$).

computational cost by a factor of 10 compared to random sampling. Similarly, MLIPs can approach the limiting force errors by about 10 meV/Å using 3 to 4 times less configuration than required by random selection. Finally, block leverage sampling is observed to yield lower force errors while regular leverage sampling leads to lower energy errors, although the differences between the two approaches are modest. It is important to note that the computational cost of leverage sampling is minimal, as it can be obtained at a computational cost comparable to that of a single linear regression solution on the whole dataset of 20,000 configurations. Importantly, configurations can be prioritized using leverage sampling without the need to carry out the related DFT simulation first, as only the features of each atomic configuration are required to compute the leverage score, but the energies and forces computed by DFT are not required.

3.5 Multi-Objective Optimization of Application-specific MLIPs

To understand the combined influence of DFT precision, training set size, energy-force weighting, and MLIP complexity ($2J_{max}$) on the cost/accuracy trade-off, we performed a systematic joint exploration. We trained numerous qSNAP potentials using a full factorial

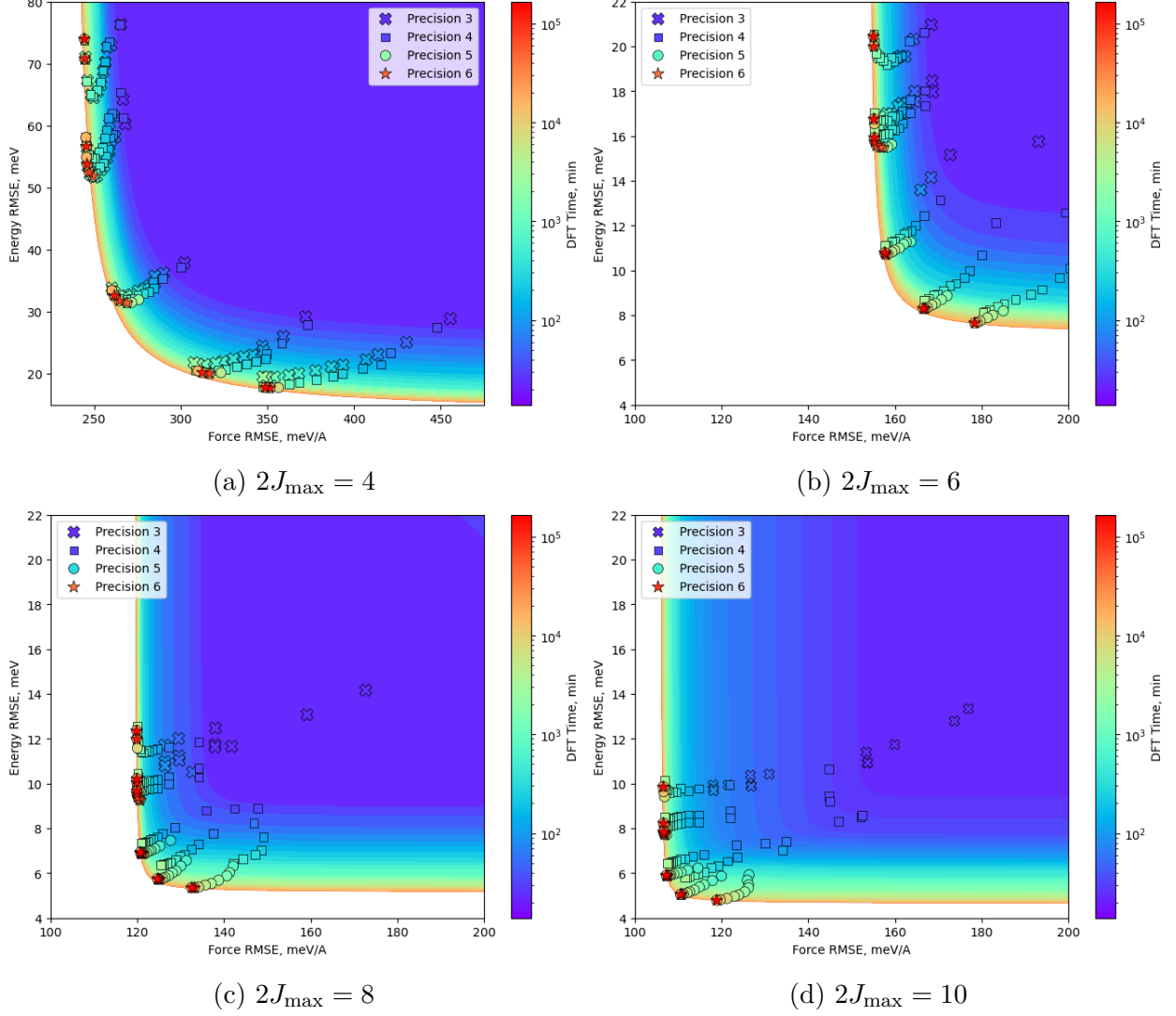


Figure 7: Testing RMSEs (Energy vs. Force) for Pareto-optimal MLIPs of varying complexity ($2J_{\max} \in \{4, 6, 8, 10\}$); all models were tested on level 6th precision DFT testing sets. Marker color and approximate background surface indicates the computational cost of the DFT simulation for all MLIPs. Marker shapes distinguish the level of DFT precision used for the training set. Markers are organized into lines, where each line corresponds to a specific energy-force weight ($w_E/w_F \in \{5, 10, 12.25, 50, 150, 300\}$). Along each line, individual markers denote different training set sizes, ranging from 100 to 10,000 configurations.

design, varying precision levels (1-6), subset sizes selected via leverage sampling, energy-force weights, and four $2J_{max}$ values. This comprehensive analysis allows us to map the Pareto-optimal front of possible MLIPs relating DFT computation time (cost) to energy and force RMSE (accuracy). Fig. 7 reports the MLIPs on this Pareto front, highlighting the broad families of optimal MLIPs in this multi-objective setting. In the figure, marker shapes encode training set precision levels while their colors denote the total DFT computation time required to obtain the training set. The markers align along rough lines that correspond to varying training subset sizes at a specific energy-force weight. Rather than pinpointing a specific MLIP from the force-energy Pareto front, we analyze the front as a whole, offering insights into the trade-offs between DFT precision and MLIP settings depending on the desired accuracy in energy vs. forces, which is a user-specified preference. MLIPs, which are not Pareto optimal, are hidden.

Several key conclusions emerge: neither the 1st nor 2nd precision levels appear on the Pareto front, indicating that smaller higher-precision subsets always outperform larger very-low-precision datasets. We postulate that this reflects an inherent trade-off where larger number of configurations of very noisy data are required to “average out” intrinsic errors, compared to high-precision data where the ultimate accuracy limit—the point at which errors are controlled only by the model’s finite complexity can be expected to occur earlier. This phenomenon can be frequently observed on the Pareto front (especially in panels (a) and (b) of Fig. 7) where different DFT precision levels can lead to similar accuracies and overall DFT computational costs, indicating that the lower-precision training sets were hence larger than their high-precision counterparts. Of course, whether the extra amount of low-precision training set configuration can be obtained at a sufficiently low computational cost compared to a smaller number of configurations at higher DFT precision is application-specific. Similarly, the 6th (highest) level precision training set are mostly absent except at the very edge of the accessible error range, due to marginal increase in precision obtained in spite of the significantly higher computational cost.

Correlating these findings with Table 3 suggests that different levels of DFT precision corresponding to the error much lower than the ultimate accuracy achievable by the MLIP due to finite complexity are unlikely to be optimal, as the amount of extra information gained by high-precision DFT simulation has a limited impact on the accuracy of the MLIP. This explains the scarcity of DFT simulation of level 6 precision on the Pareto front. Similarly, the value of low-precision DFT simulation is limited when the DFT errors significantly exceeds the accuracy achievable by the MLIP due to the need to average out these errors. This is also consistent with the absence of the 1st and 2nd precision level of DFT precision on the Pareto front, as their intrinsic errors exceed the representation capabilities of all MLIPs considered here. These observations are consistent with a rule of thumb where matching the ultimate accuracy of the MLIP and the precision of the DFT simulation is desirable.

Further analysis of Fig. 7, Fig. 8 and Supplementary Fig. ?? reveals that the optimal DFT level precision depends strongly on both the MLIP complexity ($2J_{\max}$) and whether energy or force accuracy is prioritized. For complex MLIPs ($2J_{\max} = 10$), the limiting energy and force errors saturate around 4.5 meV/atom and 105 meV/Å respectively. In this case, it is possible to approach both of these limits simultaneously through a proper choice of the energy/force weights. Approaching limiting force errors (c.f., Fig. 8d) is possible even with lower precision training set, e.g., the level 4 precision, while approaching the ultimate energy error level requires precision level 5 training set (c.f., Fig. ??), which is consistent with the observation that the comparatively unbiased statistical properties of the forces make them more susceptible to being averaged out. Nonetheless, the effect is relatively modest in absolute terms, as even precision level 4 training sets produce MLIPs whose errors are within 1.5 meV/atom and 5 meV/Å of the saturation limit. Perhaps most surprising is the observation that very good MLIPs can be obtained very efficiently. For example, models with energy and force errors within 3 meV/atom and 20 meV/Å of the saturation limit can be obtained with only approximately 2h of total computational time for DFT simulation using precision level 4. In fact, errors show a very fast decrease in the first hour of DFT

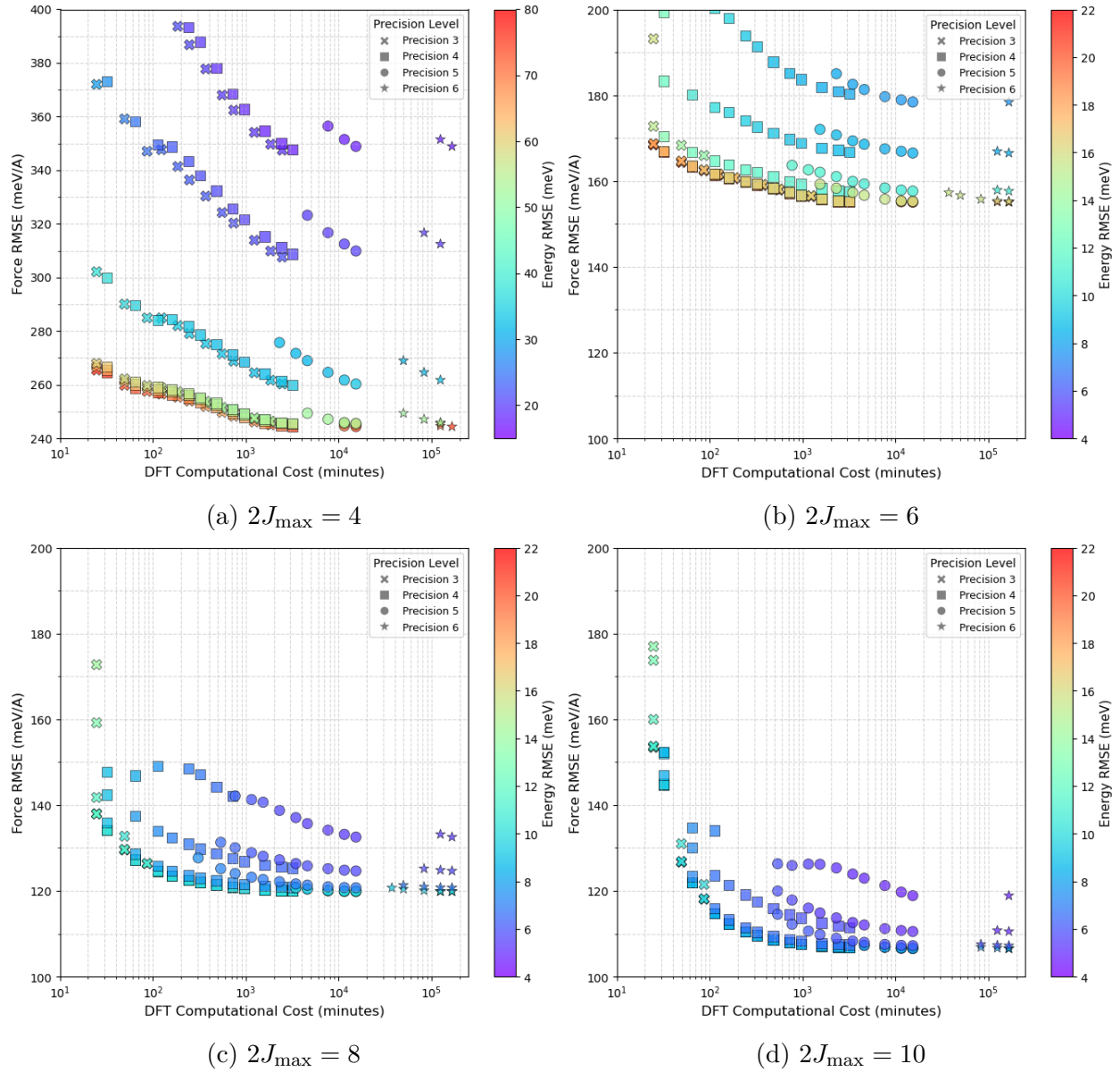


Figure 8: Testing RMSEs (Force vs. DFT time) for Pareto-optimal MLIP models of varying complexity ($2J_{\max} \in \{4, 6, 8, 10\}$); all models were tested on the level 6 precision DFT testing set. Marker color indicates the energy testing RMSE. Marker shapes distinguish the DFT precision levels used in training. Markers are organized into lines, where each line corresponds to a specific energy-force weight ($w_E/w_F \in \{5, 10, 12.25, 50, 150, 300\}$). Along each line, individual markers denote different training set sizes, ranging from 100 to 10,000 configurations.

simulations, followed by a significant slowing down where further improvements come at a high computational cost. This shows that it is possible to obtain accurate models at an extremely low computational cost. Interestingly, the results also show that the optimal precision level in fact depends on the total computational resources available for creating the DFT training set, as a small amount of computational resources (10-20 minutes) favors precision level 3 training sets, intermediate budgets (20-1000 minutes) tend to favor precision level 4 training sets, and large budgets (>3000 minutes) precision level 5 or rarely even level 6 training sets. Intuitively, this suggests that capturing the broad features of the energy landscape is better achieved with a larger number of low-precision configuration in the training set than with a small number of high-precision configuration in the training set, but that refining the detailed features of the landscape gradually calls for higher-precision configuration in the training sets.

With low-complexity MLIPs ($2J_{\max} = 4$ and $2J_{\max} = 6$), limiting energy and force errors worsen significantly. Simultaneously approaching both limits using a simple energy-force weighting is no longer feasible due to a strong energy-force error trade-off. In contrast to high-complexity MLIPs, the decrease in errors is also significantly more gradual as more DFT simulation are added to the training set. For example, focusing on “intermediate” models that attempt to strike the balance between energy and force errors (e.g., energy errors between 30 and 40 meV/atom and force errors between 260 and 300 meV/Å for $2J_{\max} = 4$), one observes that increasing the computational resources for DFT simulation from 10^2 to 10^3 minutes leads to a substantial decrease in energy and force errors (in absolute terms) by about 7 meV/atom and 20 meV/Å, respectively, which is significantly larger than the corresponding decrease for $2J_{\max} = 10$. This is perhaps counterintuitive, as simpler models should require less data to constrain, and could therefore be expected to converge faster with increasing training set size. However, this intuition does not extend to misspecified models where no combination of free parameters can perfectly reproduce the training data. In this case, adding more configurations to an already large training set (i.e., much larger than the number

of adjustable parameters) can still significantly affect the MLIPs accuracy, even leading to worsening test errors in some cases,¹⁷ a phenomenon that can also be observed here for energy errors at large force weights. Although the leverage sampling strategy should partially mitigate this trend by introducing the most influential points first, the slow convergence of energy and force errors with respect to the training set size is consistent with the behavior of highly misspecified models.

4 Discussion

The above results explore the trade-offs between computational cost and accuracy when fitting application-specific MLIPs. They provide guidance for selecting the level of DFT precision and MLIP complexity based on the specific application of the MLIP and the acceptable computational cost to evaluate the MLIP for MD simulation. Our work shows that significant computational cost can be saved by considering multiple factors that can influence the quality of the MLIP.

First, we find that very tightly converging DFT calculations can be wasteful when coupled with MLIPs of limited expressivity. Tables 4 and 5, along with Fig. 7, show that MLIPs trained on precision level 5 training sets provide the same level of accuracy as those trained on precision level 6 at a 10-fold reduction in cost. This is consistent with the rule of thumb that the accuracy of the model and of the data benefits from being roughly matched.

Second, further savings are possible by considering how DFT errors translate into MLIP errors, which is itself dependent on the expressivity of the model. Our Pareto analysis (Fig. 7) shows that if applications allow for more expressive MLIPs (e.g., $2J_{\max} \geq 6$), training to medium precision data (level 4 in our case) selected by leverage sampling can produce MLIPs nearly as accurate as those trained on precision level 5 training sets. This approach can reduce the DFT computational cost by about 10 times compared to using the full precision level 5 training set, and up to about 100 times compared to using the highest

precision level. In optimal conditions, convergence with respect to training set size can occur extremely quickly, using only a handful of GPU hours.

Third, if the application requires very computationally efficient MLIPs (e.g., $2J_{\max} = 4$), these simpler MLIPs are less sensitive to DFT errors. Fig. 7d shows that in these cases, lower-precision data can be used. This further reduces the computational cost of the DFT simulation while maintaining the same accuracy similar to higher-precision training sets for the same simple MLIP. Using precision level 3 saves about 50 times the computational cost for DFT simulation compared to precision level 5, and over 500 times compared to precision level 6 to characterize the whole training set. However, we also observed that very simple potentials can counterintuitively require a larger amount of data to achieve convergence with respect to training set size due misspecification effects.

Of course, when assembling datasets for applications that require accuracy at the expense of inference cost, or when curating databases of reference results, e.g., to train universal models, erring on the side of caution and employing tight DFT convergence settings remains a safe strategy. However, even in this setting, quickly generating lower-precision data to fine-tune universal models might prove to be the superior option.

5 Conclusion

Developing cost-effective, accurate machine-learned interatomic potentials (MLIP) remains essential, particularly for specific applications requiring computationally efficient MLIPs for long/large Molecular Dynamics (MD) simulation or in cases where multiple different materials must be considered on a limited computational budget. In order to delineate the importance of the different factors that contribute to the cost/accuracy tradeoff of MLIPs, we considered the role of DFT training set precision, MLIP model complexity, training set sub-sampling, and relative energy/force reweighting. Our study demonstrates the benefits of leveraging lower-precision DFT training sets, which are computationally much more efficient

to obtain than the typically high-precision DFT precision advocated in the literature. However, efficiently leveraging lower-precision training sets is shown to require the consideration of the relative weighting of energies and forces in the overall loss function, as DFT errors in the forces converge faster than errors in the energy.³⁹ Coupled with a training set sub-selection strategies based on leverage sampling, we mapped out the Pareto front of locally optimal MLIPs with respect to energy and force errors, and computational cost to construct the DFT training set. The results confirm that the use of lower precision DFT training sets can be beneficial when the precision of the DFT training set can be roughly matched to the intrinsic levels of errors due to finite complexity of the MLIP. This is especially true for constrained computational budgets where lower precision DFT training sets dominate the Pareto front. Through a careful optimization of the DFT precision, energy/force weights, and training set size, we observed that MLIPs for unary beryllium that approach the intrinsic error saturation limit by a few meV/atom and meV/Å respectively can be obtained with as little as 2 hours of aggregate computational resources for DFT simulation using medium precision training sets, but that fully converging the MLIPs can require orders of magnitude more computational resources. Counterintuitively, we also observed that converging low-complexity MLIPs can be even more costly in terms of computational resources for DFT simulation, due to slow convergence with respect to training set size, which we attribute to the effect of model misspecification. Our study suggests that extremely large efficiency gains can be achieved through a joint consideration of the multiple factors that control the cost/accuracy trade-off of MLIPs, highlighting the advantages of application-specific MLIPs.

6 Author contributions

Ilgar Baghishov: Methodology, Software, Data Curation, Writing - Original Draft. **Jan Janssen:** Conceptualization, Methodology, Data Curation, Writing - Review & Editing, Supervision, Funding acquisition. **Graeme Henkelman:** Methodology, Writing - Review &

Editing, Supervision, Funding acquisition. **Danny Perez:** Conceptualization, Methodology, Writing - Review & Editing, Supervision, Funding acquisition.

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Application-specific Machine Learned Interatomic Potentials – Exploring the Trade-off Between Precision and Computational Cost

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Supporting Information Available

Table 1: Table of Training and Testing Energy RMSE (meV). The rows correspond to MLIPs trained at 6 different precision levels, with the first column showing training errors and the remaining columns showing testing errors across 6 precision levels. The bold diagonal elements represent testing RMSE at the same precision level as training.

	Precision Level	Train RMSE	Test RMSE with precision level					
			1	2	3	4	5	6
Trained on	1	111.74	117.91	423.99	469.50	471.65	473.20	473.42
	2	30.90	448.25	33.18	51.43	51.83	53.56	53.76
	3	11.01	492.74	61.29	11.67	11.17	11.05	11.08
	4	6.20	494.95	62.64	15.79	6.58	5.74	5.80
	5	4.79	496.43	64.22	16.29	7.27	5.06	5.05
	6	4.77	496.64	64.39	16.31	7.33	5.06	5.05

Table 2: Table of Training and Testing force errors (meV/Å) The rows correspond to MLIPs trained at 6 different precision levels, with the first column showing training errors and the remaining columns showing testing errors across 6 precision levels. The bold diagonal elements represent testing RMSE at the same precision level as training.

	Precision Level	Train RMSE	Test RMSE with precision level					
			1	2	3	4	5	6
Trained on	1	931.09	944.15	906.49	902.43	901.73	900.36	900.29
	2	235.72	439.12	238.89	200.43	193.04	186.92	186.67
	3	143.30	435.09	203.90	145.03	133.97	124.86	124.53
	4	120.46	434.8	198.42	136.93	121.86	111.85	111.50
	5	109.41	434.56	198.47	136.81	121.54	110.87	110.51
	6	109.04	434.5	198.44	136.82	121.56	110.88	110.52

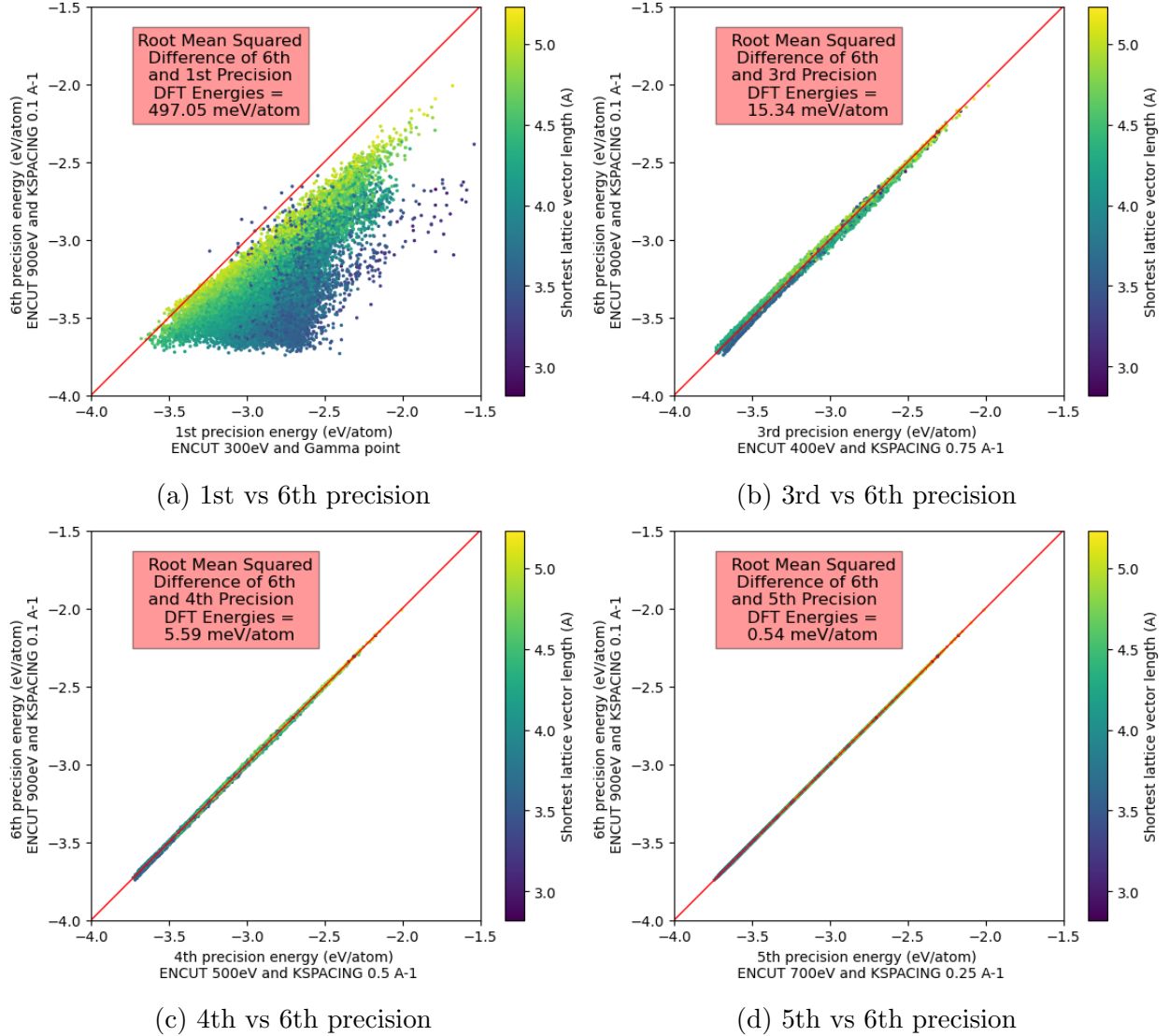
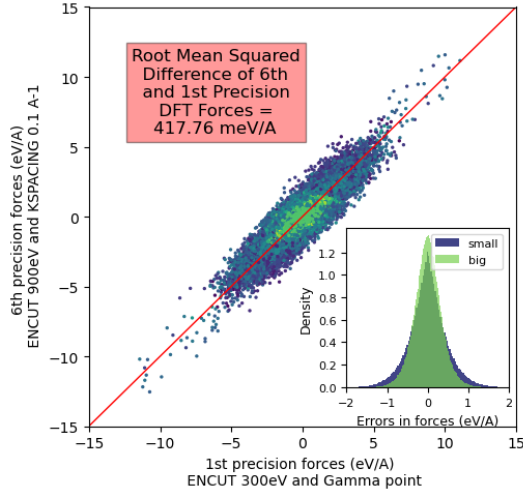
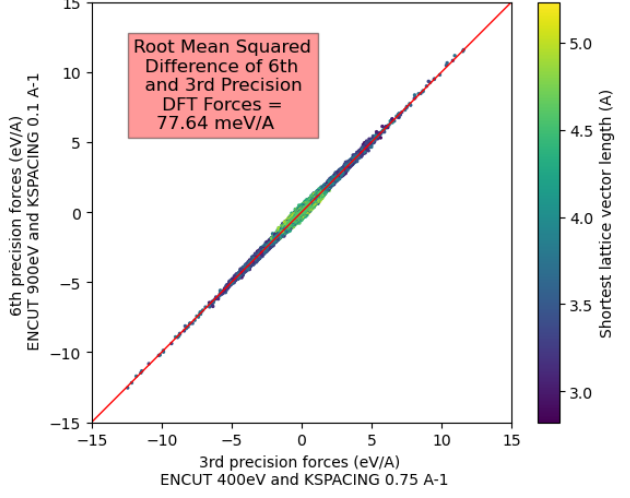


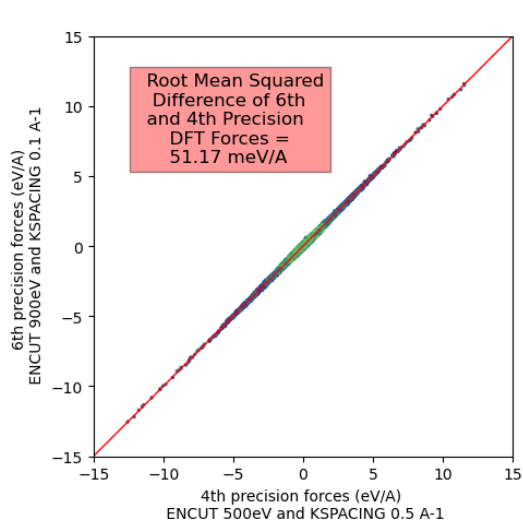
Figure 1: Pairwise relationships between DFT energies of various precision levels.



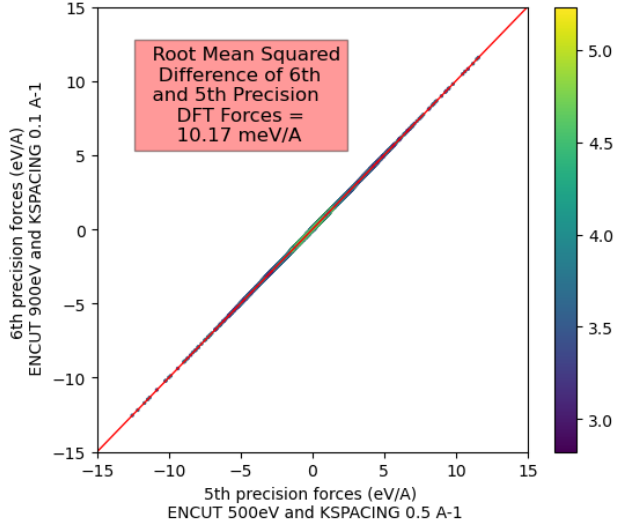
(a) 1st vs 6th precision



(b) 3rd vs 6th precision



(c) 4th vs 6th precision



(d) 5th vs 6th precision

Figure 2: Pairwise relationships between DFT forces of various precision levels.

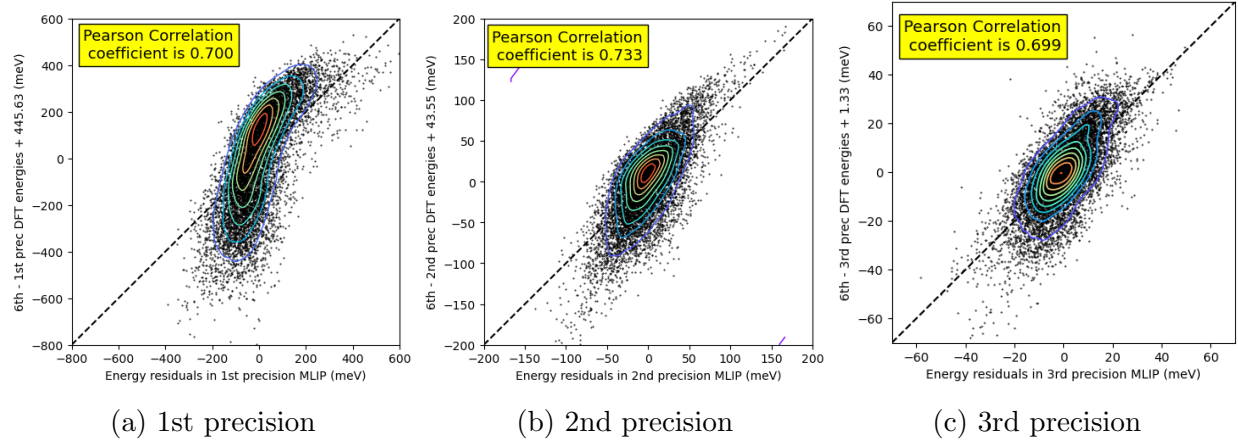


Figure 3: Energy correlation between high energy-weighted MLIP training errors and intrinsic DFT precision errors.

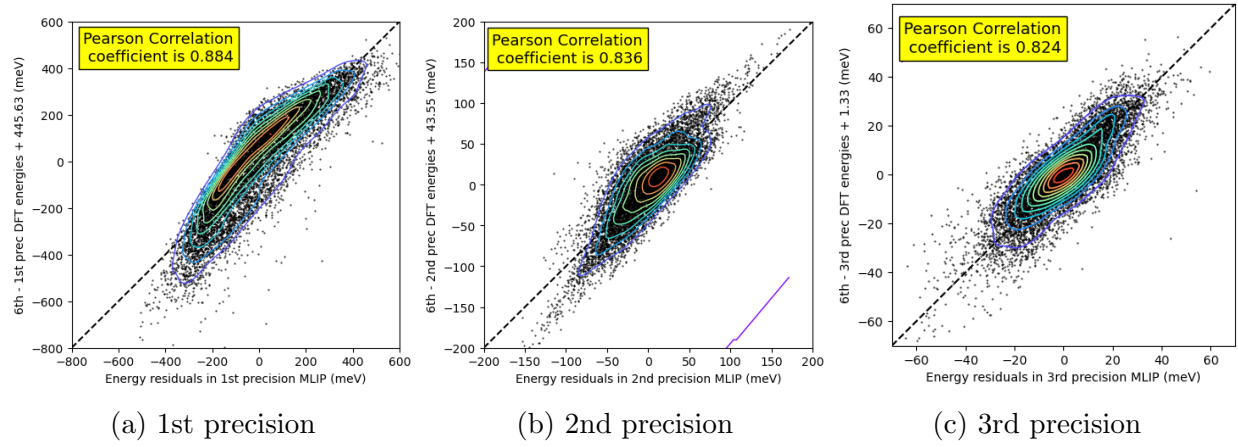


Figure 4: Energy correlation between high force-weighted MLIP training errors and intrinsic DFT precision errors.

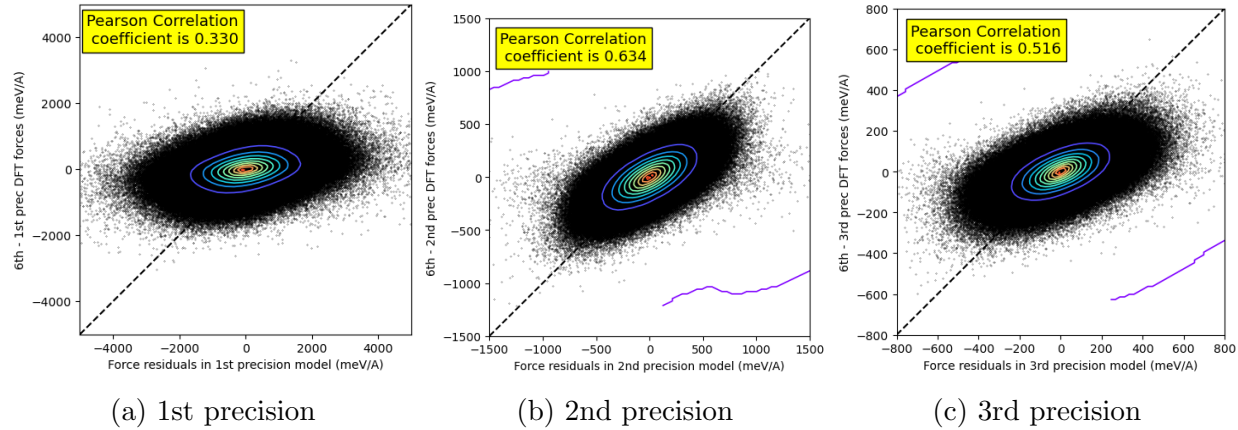


Figure 5: Force correlation between high energy-weighted MLIP training errors and intrinsic DFT precision errors.

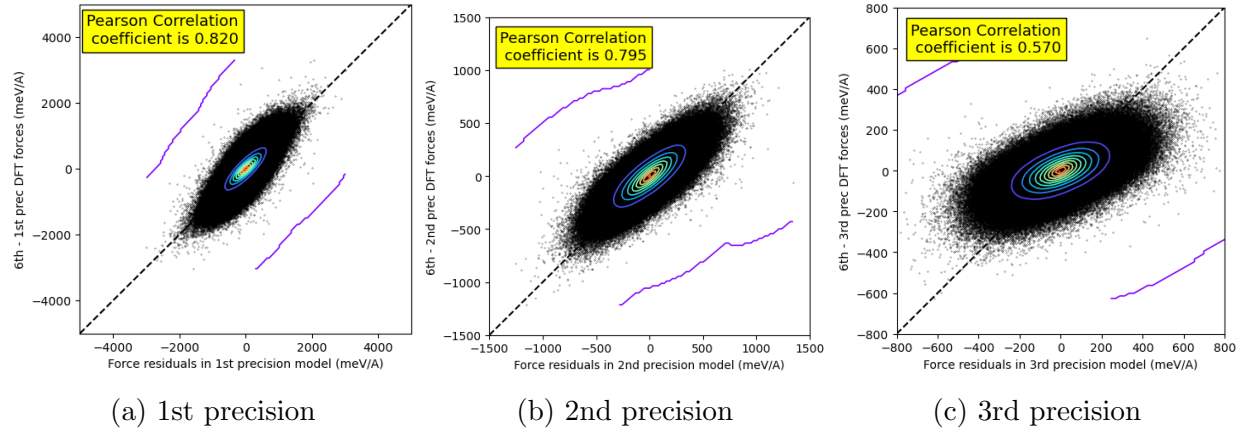


Figure 6: Force correlation between high force-weighted MLIP training errors and intrinsic DFT precision errors.

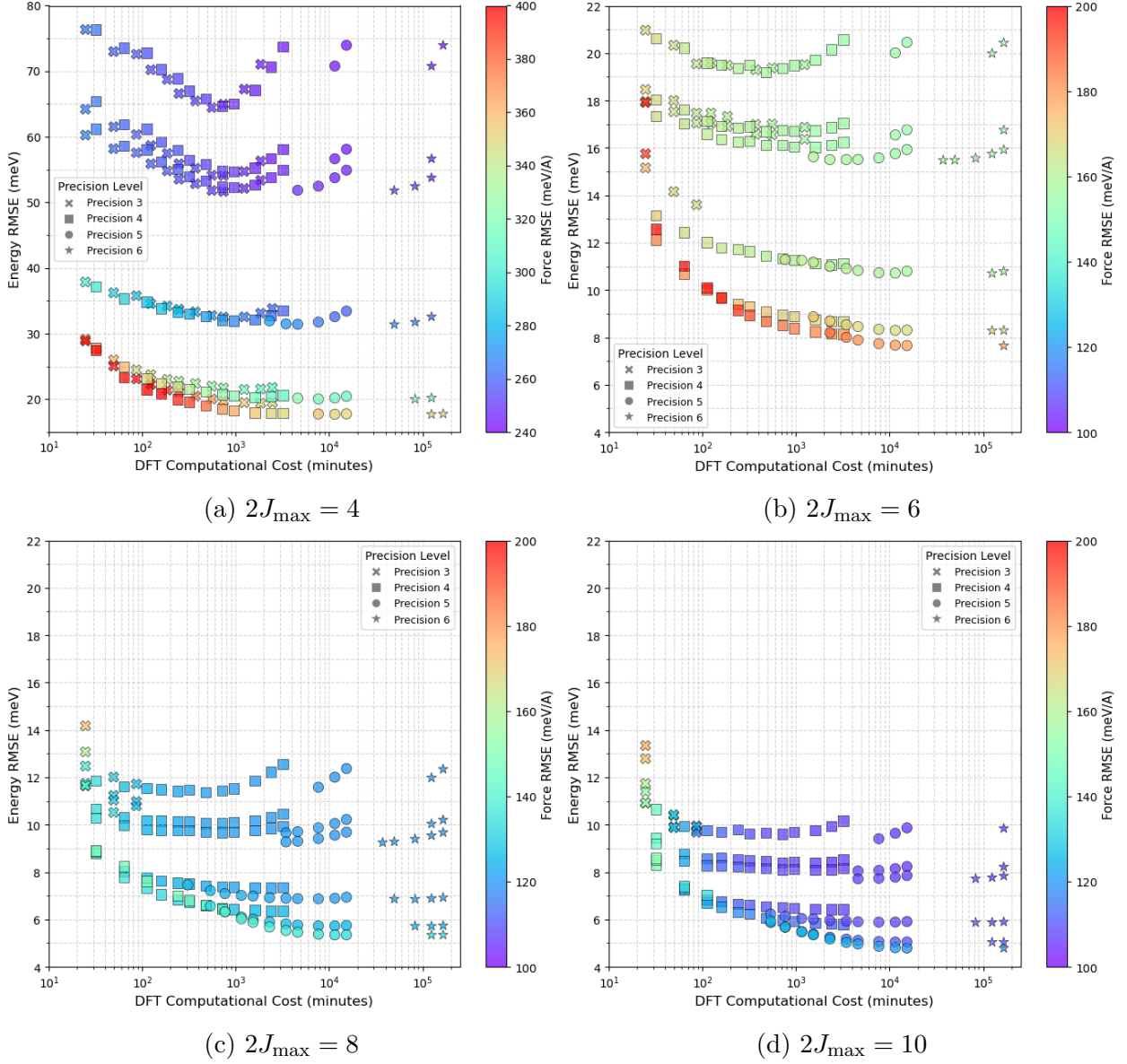


Figure 7: Testing RMSEs (Energy vs. DFT time) for Pareto-optimal MLIP models of varying complexity ($2J_{\max} \in \{4, 6, 8, 10\}$); all models were tested on 6th precision DFT data. Marker color indicates the force testing RMSE. Marker shapes distinguish the DFT precision levels used in training. Markers are organized into lines, where each line corresponds to a specific energy-force weight ($w_E/w_F \in \{5, 10, 12.25, 50, 150, 300\}$). Along each line, individual markers denote different training set sizes, ranging from 100 to 10,000 configurations.