Injecting Domain Knowledge from Empirical Interatomic Potentials to Neural Networks for Predicting Material Properties

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Abstract

For decades, atomistic modeling has played a crucial role in predicting the behavior of materials in numerous fields ranging from microprocessor design to drug discovery. The most accurate methods in this domain are rooted in first-principles quantum mechanical calculations such as density functional theory (DFT). Because these methods have remained computationally prohibitive, practitioners have traditionally focused on defining physically motivated closed-form expressions known as empirical interatomic potentials (EIPs) that approximately model the interactions between atoms in materials. In recent years, neural network (NN)-based potentials trained on quantum mechanical (DFT-labeled) data have emerged as a more accurate alternative to conventional EIPs. However, the generalizability of these models relies heavily on the amount of labeled training data, which is often still insufficient to generate models suitable for general-purpose applications. In this paper, we propose two generic strategies that take advantage of unlabeled training instances to inject domain knowledge from conventional EIPs to NNs in order to bolster their generalizability. The first strategy, based on weakly supervised learning, trains an auxiliary classifier on EIPs and selects the best-performing EIP to generate energies to supplement the ground-truth DFT energies in training the NN. The second strategy, based on transfer learning, first pretrains the NN on a large set of easily obtainable EIP energies, and then fine-tunes it on ground-truth DFT energies. Experimental results on two benchmark datasets demonstrate that the first strategy improves baseline NN performance by 18% to 51% while the second improves baseline performance by 18% to 26%. Combining them further boosts performance by 22% to 53%.

1 Introduction

Predictive modeling of materials is a field with manifold applications that has been the subject of several cross-disciplinary studies. While modeling is conducted at different length scales, all material behavior ultimately has its origins at the nano scale, where the interactions between individual atoms must be understood. The most accurate methods at this scale are based on quantum mechanics theory, requiring explicit consideration of the electronic degrees of freedom described by the Schrödinger equation. However, these methods are presently limited to systems containing at most several thousand atoms, precluding their use in investigation of important microstructural phenomena such as crack propagation. To overcome this limitation, practitioners have long relied upon heuristic models known as *empirical interatomic potentials* (EIPs), which consist of physically motivated analytical functional forms that strive to model the complex electronic interactions between atoms using only the nuclear coordinates of the atoms and their elemental species as input [36]. In the past several years,

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there has been a surge of interest in the development of machine learning EIPs, particularly those based on neural networks (NNs), as a more accurate alternative to traditional EIPs [5]. In contrast to traditional EIPs, NN potentials contain little inductive bias and, accordingly, require large volumes of training data labeled using first-principles quantum mechanical methods. The first-principles method most commonly used is density functional theory (DFT), which scales as $\mathcal{O}(n_{\mathrm{e}}^3)$ with the number of valence electrons n_{e} in a given configuration of atoms [24]. As a result of this high computational cost, it is expensive to acquire a sufficient number of labeled training instances to create an NN potential that performs accurately over a wide range of applications.

One potential solution to this problem is to seek additional supervision signals. Traditional EIPs are attractive sources of such additional supervision for two reasons. First, their functional forms incorporate prior physical information that allows them to correlate with DFT, and, in regions of relevance to common applications, are often quite accurate. That is, they contain domain knowledge that could benefit the training of NN potentials. Second, EIPs scale linearly with the number of atoms and are thus orders of magnitude faster than DFT, permitting the labeling of massive datasets. Despite the advantages, no research has focused on using EIP supervision signals in training NN potentials.

In this paper, we leverage physically motivated EIPs and unlabeled configurations to tackle the label scarcity challenge for training NN potentials.We propose two generic strategies, weakly supervised learning and transfer learning, for exploiting this additional source of information. In the first strategy, we expand the DFT-labeled training set with unlabeled configurations and their EIP energies. To achieve this goal, we train an auxiliary classifier on the original DFT-labeled training configurations that predicts which one of a selected set of EIPs is likely to produce the most accurate estimate of the DFT energy for each of a large set of unlabeled configurations. The unlabeled configurations are then labeled by their corresponding predicted best-performing EIPs and appended to the training set. We train NN potentials on the expanded training set by optimizing a robust regression loss to mitigate the influence of noise and outliers introduced by the EIP energies. In the second strategy, we adopt a transfer learning approach by way of multi-task pretraining. We first pretrain the representation module of an NN potential to reproduce the energies predicted by the EIPs. During the subsequent fine-tuning stage, the representation module of the NN is paired with a prediction head and trained on the DFT-labeled configurations. The two strategies could be flexibly used and coupled to train any NN potentials.

The contributions of this work are three-fold: 1) We demonstrate that EIPs could provide high quality supervision signals for training NN potentials. This opens a new direction for future development of NN potentials; 2) We propose two effective and generic strategies that take advantage of EIPs and unlabeled configurations to tackle the label scarcity challenge for training NN potentials; 3) We conduct comprehensive experiments on two benchmark datasets and two representative NN potentials that cover most of the NN potential forms currently in use. Numerical experimentation results show that the proposed strategies successfully inject domain knowledge in EIPs into NN potentials and improve the performance of the NN potentials by 18% to 53% on the two datasets.

2 Preliminaries

2.1 Atomic Configurations

The fundamental input in atomistic modeling is an *atomic configuration*. An atomic configuration is a spatial arrangement of atoms $C = \{(Z_i, \mathbf{r}_i)\}_{i=1}^N$ where Z_i and $\mathbf{r}_i$ are the atomic number and the three-dimensional Euclidean coordinates of atom i, respectively. In the simplest scenario, an atomic configuration corresponds to an isolated cluster of atoms that comprise a molecule. However, it may more generally describe a bulk system such as a crystal, which contains an infinite number of atoms distributed over space. These systems are modeled using a small collection of atoms in a finite simulation cell that is effectively repeated across all of space with the aid of periodic boundary conditions (PBCs) [36] (see Appendix A.1).

2.2 Physics-based Empirical Interatomic Potentials

Mathematically, an EIP is a function, $E = \mathcal{V}(C; \theta)$, that takes an atomic configuration C as input and returns its total potential energy E; θ denotes a set of fitting parameters to be determined. The functional form $\mathcal{V}$ of a physics-based EIP is made up of carefully designed analytic expressions that

strive to capture the underlying physics in the material it models [45]. Because the functional form itself is intended to capture most of the relevant physics, such models need relatively few parameters (typically on the order of ten) and are usually fitted to a set of material properties deemed most relevant to real-world applications. It can generally be expected that physics-based EIPs will approximate the first-principles energy surface well in the vicinity of atomic configurations corresponding to the material properties to which they were fit. However, their generalizability can be inconsistent, as shown in Fig. 1.

2.3 Machine Learning Potentials

In contrast, machine learning EIPs employ general-purpose regression algorithms as the functional form $\mathcal{V}$, which do not encode any knowledge of the material it models. Therefore, machine learning EIPs are almost exclusively trained on large sets of DFT data so as to include as much physical knowledge as possible.

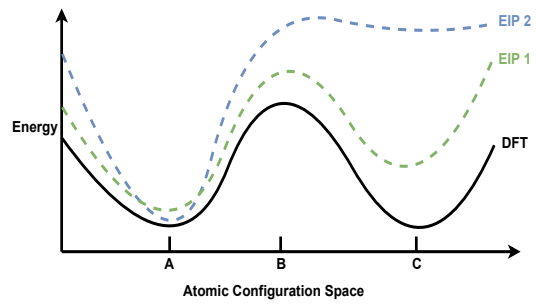


Figure 1: Schematic illustration of the energy landscape defined over atomic configuration space by two physics-based EIPs and by DFT. Both EIPs are fitted to reproduce properties of the DFT energy landscape near atomic configuration A. Away from configuration A, the relative accuracy of the EIPs compared to DFT varies: at point B, EIP 1 is a fair approximation while EIP 2 is less accurate; at point C, neither EIP is accurate.

One important class of machine learning EIPs are neural network (NN)-based potentials. In this paper, we define an NN potential $\mathcal{M} : \mathcal{C} \mapsto \mathcal{E}$ as

$$\mathcal{M}(C) = f_{\text{pred}} \circ f_{\text{rep}}(C),$$

where $f_{\text{rep}} : \mathcal{C} \mapsto \mathcal{R}^{n \times d}$ is the representation learning module that maps each of the n atoms of C to a feature vector of length d based on its local environment, and $f_{\text{pred}} : \mathcal{R}^{n \times d} \mapsto \mathcal{E}$ is the prediction module that maps the feature vectors of the atoms to the total potential energy.

A concrete instance of this type of model are those that use a graph neural network (GNN) [26, 16, 42, 47] as the representation learning module and a multilayer perceptron (MLP) as the prediction module. Before being passed to the representation learning module f_{rep}, an atomic configuration C is first converted to a graph $\mathcal{G} = (V, E)$ with nodes V and edges E. One node is defined for each atom in the simulation cell, as well as for additional padding atoms representing the PBCs. An edge is created between any two nodes with a distance smaller than a prescribed cutoff radius. Next, for each atom i, its atomic number Z_i is one-hot encoded into an initial feature vector $h_i^{(0)}$ of the node corresponding to the atom. The GNN representation learning module f_{rep} then updates the feature vectors using a message passing paradigm [15], i.e., the nodes iteratively aggregate information from their neighbors. Formally, the feature vector of node i at the $l+1$-th layer $\mathbf{h}_i^{(l+1)}$ is updated as a function of the feature vectors of its neighbors $\mathcal{N}(i)$ and itself at the previous layer,

$$\mathbf{m}_i^{(l)} = \text{Agg}^{(l)}\left(\left\{f^{(l)}\left(\mathbf{h}_i^{(l)}, \mathbf{h}_j^{(l)}, \mathbf{e}_{ij}^{(l)}\right) \mid j \in \mathcal{N}(i)\right\}\right), \tag{1}$$

$$\mathbf{h}_i^{(l+1)} = \text{Update}^{(l)}\left(\mathbf{h}_i^{(l)}, \mathbf{m}_i^{(l)}\right), \tag{2}$$

where $f^{(l)}$, $\text{Update}^{(l)}$ are learnable functions, and $\text{Agg}^{(l)}$ is a permutation-invariant function that operates on sets of feature vectors, $\mathbf{e}_{ij}^{(l)}$ denotes the feature of edge connecting node i and node j at the l-th layer. Finally, the prediction module f_{pred} maps the feature vector of each atom to a corresponding energy contribution and sums them to arrive at the total energy of the configuration, i.e., $E = \sum_i \text{MLP}(\mathbf{h}_i)$.

3 Related Works

3.1 Neural Network Potentials

The first modern NN potential was proposed by Behler and Parrinello [4]. In their formulation, an atomic descriptor (i.e., basis functions that transform an atomic configuration into a fixed-length fingerprint vector) based on the bond lengths and bond angles is passed to an MLP. On top of this formulation, a Monte Carlo dropout technique is applied to the MLP to equip the potential with the ability to quantify its predictive uncertainty [44]. The DeePMD method is a similar approach to that of Behler and Parrinello, except that a novel atomic environment descriptor is used [50]. More recently, GNN potentials based on a message passing paradigm such as SchNet have been developed [33]. Similar to SchNet, NequIP [3] is another GNN potential, but in NequIP, equivariant messages rather than invariant ones are passed based on the formulation of tensor field networks [39]. All of these models are trained with supervised learning, without exploring the possibilities of leveraging weakly supervised learning or transfer learning to take advantage of unlabeled data.

3.2 Weakly Supervised Learning

Weakly supervised learning refers to techniques that try to train machine learning models from incomplete (partial training instances are labeled), or inaccurate (noisy labels) supervision signals [52]. Solutions for incomplete supervision usually fall into the category of semi-supervised learning, which assumes that nearby instances have similar labels [27, 35, 32, 22]. For inaccurate supervision, a model either learns directly from noisy labels with noise-robust algorithms [13, 51, 14] or resorts to a small portion of clean labeled data to reduce the noise [41, 46].

3.3 Transfer Learning

Transfer learning [53] refers to a machine learning paradigm that transfers the knowledge a model learned from one or more relevant tasks to benefit a target task. Transfer learning has enjoyed great success, especially in the low-data regime, as demonstrated by the rise of pretrained neural networks. This recent trend began with natural language processing when BERT [10] and successive large pretrained language models [29, 31, 8] were released and quickly gained popularity in other domains such as computer vision [17, 9, 11] and graph learning [48, 20, 49]. These methods pretrain large neural networks on self-supervised tasks in order to encode common contextual knowledge in the structured input. Another approach imparts domain-specific knowledge by pretraining models on tasks related to the target task but for which abundant labeled data is available [19]. The pretrained model is then fine-tuned on the limited training data of the target task.

4 Methods

4.1 Problem Definition

Let $\mathcal{P}$ be a set of physics-based EIPs, $\mathcal{C}$ be the space of all possible atomic configurations, $\mathcal{C}_{\text{DFT}} = \{C_i, E_i^{\text{DFT}}, \{E_i^p\}_{p\in\mathcal{P}}\}_{i=1}^m$ be a set of configurations with corresponding DFT and EIP energies, and $\mathcal{C}_{\text{EIP}} = \{C_i, \{E_i^p\}_{p\in\mathcal{P}}\}_{i=m+1}^{m+n}$ be a set of configurations with only EIP energies. Here, E_i and E_i^p denote the energies predicted for configuration C_i by DFT and the p-th EIP, respectively. In practice, EIPs are much less expensive than DFT, and so the size of $\mathcal{C}_{\text{EIP}}$ is much larger than $\mathcal{C}_{\text{DFT}}$, i.e., $n \gg m$. Our goal is to use this data to train an NN-based regression potential $\mathcal{M}$ to closely approximate the DFT energy surface over the space of all atomic configurations, i.e.,

$$\mathcal{M}(C) \approx E^{\text{DFT}}, \forall C \in \mathcal{C}.$$

4.2 Label Augmentation

Because physics-based EIPs are developed as approximations to DFT, it is intuitive to use their predictions as surrogate labels for configurations without DFT energies in order to resolve the label scarcity issue of training neural networks. However, there are two fundamental challenges. First, as discussed in Sec. 2.2, while physics-based EIPs are designed to be accurate in specific regions of configuration space and generalize better than those based on machine learning, they may still

be very inaccurate in other regions. Given an arbitrary configuration for which no DFT energy is available, it is unknown which EIP from a given set will yield the most accurate prediction and how great the magnitude of its error will be. Second, surrogating DFT labels with EIP labels inevitably introduces noise, and potentially outliers, into the training set that may have pathological effects on the training process. To overcome these difficulties, we propose a label augmentation strategy that can reliably increase the size of the training set while mitigating the influence of noise and outliers.

EIP prediction using an auxiliary classification model. In order to augment training sets with unlabeled configurations and their corresponding EIP-approximated energies, we use an auxiliary classification model to predict the best-performing EIP for a given configuration. For a configuration C, the classification model predicts a discrete distribution over the EIP set $\mathcal{P}' = \mathcal{P} \bigcup \{p_\emptyset\}$ that indicates their probability of being the most accurate EIP for C, i.e., $P(\mathcal{P}' \mid C)$. We introduce a dummy EIP, $p_\emptyset$, to represent the case where none of the EIPs in $\mathcal{P}$ is predicted to approximate DFT to an accuracy level c, i.e., $\frac{1}{N_i}||E_i^p - E_i^{\text{DFT}}||_1 > c, \forall p \in \mathcal{P}$, where N_i is the number of atoms in configuration i, c is set to 0.1. By excluding configurations that are labeled with the dummy class from the training set, c acts as a confidence threshold to control the noise and outliers introduced by using the surrogate EIP energies.

The classification model consists of a representation learning module that converts an atomic configuration to fixed-length feature vectors (one for each atom in the configuration), a permutation-invariant readout function that aggregates them to form a feature vector describing the entire configuration, and a prediction module that maps the configuration representation to a set of probabilities. We train the classification model on $\mathcal{C}_{\text{DFT}}$ by optimizing a cross-entropy loss and apply it to $\mathcal{C}_{\text{EIP}}$. Configurations with a predicted EIP other than $p_\emptyset$ are assigned the corresponding EIP energy and are merged with the configurations that have DFT energies to arrive at the final training set. We denote the set of EIP-labeled configurations as $\hat{\mathcal{C}}_{\text{EIP}} = \{C_i, \{E_i^p\}_{p \in \mathcal{P}}, E_i^{\hat{p}_i}\}_{i=m+1}^{m+s}$ where $1 \leq s \leq n$ is the number of selected unlabeled configurations, and $\hat{p}_i$ and $E_i^{\hat{p}_i}$ are the predicted best-performing EIP and its prediction on C_i.

Regression with robust loss functions. We train the NN potential using configurations with ground-truth DFT energies and configurations with EIP energies selected by the classification model. In regression problems, models are usually trained by optimizing the mean square error (MSE) loss. The MSE loss is sensitive to outliers, as the magnitude of its gradient is linearly proportional to the difference between the predicted value and the ground truth value. To lessen the impact of outliers, we optimize the MSE loss on DFT-labeled configurations, while on EIP labeled-configurations, we optimize the Tukey biweight (bisquare) loss [7, 12, 6], i.e.,

$$\mathcal{L} = \frac{1}{m} \sum_{i=1}^{m} (E_i^{\text{DFT}} - \hat{E}_i)^2 + \frac{\alpha}{s} \sum_{i=m+1}^{m+s} l_{\text{Tukey}}(E_i^{\hat{p}_i} - \hat{E}_i) \tag{3}$$

where $\hat{E}_i$ is model prediction, and $\alpha > 0$ is a hyper-parameter that controls the contribution of EIP-labeled configurations to the loss and its gradient.

The Tukey biweight loss falls under the M-estimation method [21]. The main idea of the Tukey loss is to screen outliers using robust statistics of the regression residuals such as median absolute residuals (MAR) and suppress their influence on the gradient. The Tukey loss function is computed as

$$l_{\text{Tukey}}(r_i) = \begin{cases} \frac{k^2}{6}\left[1 - \left(1 - \left(\frac{r_i}{k}\right)^2\right)^3\right], & |r_i| \leq k \\ \frac{k^2}{6}, & |r_i| > k \end{cases} \tag{4}$$

where $r_i = E_i^{\hat{p}_i} - \hat{E}_i$ is the residual, k is a tuning constant that is commonly set to 4.685σ to produce 95% efficiency when the errors are normally distributed with standard deviation σ. To set the value of k, we estimate the standard deviation as $\hat{\sigma} = \text{MAR}/0.6745$. From Eq. 4, residuals with absolute values greater than k are considered outliers and are rejected for gradient computation. We empirically show that the Tukey loss can precisely reject outliers and reduce noise introduced by EIP-labeled configurations as training proceeds.

In each gradient step, we sample from both DFT- and EIP-labeled configurations to form a batch for gradient back-propagation. The MAR is estimated on residuals of both DFT and EIP labeled

configurations in the batch. Since the MAR and $\hat{\sigma}$ are dynamically estimated during training, the Tukey loss does not introduce any hyperparameters.

4.3 Multi-task Pretraining with Empirical Potential Predictions

We propose a multi-task pretraining strategy to encode the domain knowledge in physics-based EIPs to the parameters of the representation learning module by jointly predicting the set of EIP calculations for configurations with only EIP predictions, $\mathcal{C}_{\text{EIP}}$, and optimizing the following multi-task regression loss:

$$\mathcal{L} = \frac{1}{n|\mathcal{P}|} \sum_{p \in \mathcal{P}} \sum_{i=m+1}^{m+n} (\hat{E}_i^p - E_i^p)^2.$$

During pretraining, we couple the representation learning module with $|\mathcal{P}|$ prediction modules (MLPs) to generate predictions corresponding to different EIPs, i.e., $\hat{E}_i^p = f_{\text{pred}}^p \circ f_{\text{rep}}(C_i)$. The pretrained representation module is then fine-tuned with a randomly initialized prediction module for the downstream DFT prediction task. Note that the representation module could either be naively fine-tuned on configurations with DFT energies by optimizing an MSE loss, or on the training set generated by our proposed label augmentation method by optimizing Eq. 3.

Although transfer learning has been successful in various application domains, it could easily hinder model performance on the target task if the pretraining tasks are unrelated to the target task (negative transfer). We argue that predicting the output of EIPs is relevant and beneficial to the target task of predicting DFT energies. Although EIPs are not perfectly accurate across the space of all possible spatial arrangements of atoms, their functional forms incorporate prior physical information that allow them to correlate with DFT over this space. In Sec. 5, we empirically demonstrate that the multi-task pretraining strategy successfully encodes domain knowledge into configuration representations and creates a smoother DFT energy surface.

5 Experiments

5.1 Datasets

To test our methodology, we experimented with two datasets: the ANI-Al dataset and the KIM-Si dataset. The ANI-Al dataset [34] consists of DFT energies of 6,352 configurations of aluminum in liquid, solid, and liquid-solid coexistence phases. Each configuration contains up to 250 atoms. The KIM-Si dataset is a new dataset that we generated for silicon comprising a total of 16,110 configurations. It builds upon the training set first used in [23] and contains a total of 14,510 perturbed bulk structures, 1,525 randomly generated atomic clusters ranging in size from two atoms to ten atoms, 3 ideal surface defects, and 72 nanostructures constructed by composing two-dimensional structures of silicon in the graphene and silicene geometries. In total, for the KIM-Si dataset, DFT energies are available for 510 of the bulk configurations and the 1600 non-bulk configurations.

We randomly remove the DFT energies of 5,081 configurations in the ANI-Al dataset and treat them as unlabeled configurations. In some of our experiments, we use the removed DFT energies only for evaluation purposes. However, we do not use these DFT energies in our strategies in any form, including but not limited to training, validation, and model selection. For each dataset, we generate three splits by randomly assigning 20% of the DFT-labeled configurations as test sets and the other 80% of the DFT-labeled configurations are training sets. During training, we use 20% of the training set as a validation set for model selection. All of the reported experimental results are averaged over the three different splits to avoid over-fitting to a specific split.

5.2 Experimental Setting

5.2.1 Neural Network-based Potentials

We evaluate our proposed strategies on two classes of neural network potentials that reflect the majority of machine learning EIPs currently in use. The first represents atomic environments using pre-computed descriptors and learns non-linear transformations (MLPs) to map the descriptors to atomic embeddings, while the second uses GNNs to learn atomic representations from configuration

Table 1: Performance of the two proposed strategies on DFT energy prediction tasks. We report the configuration-level and atom-level mean absolute error (MAE, lower the better) in eV and eV/atom, respectively. We denote the label augmentation strategy by LA and the multi-task pretraining strategy by MP. Best performance is shown in bold.

		KIM-Si			ANI-Al		
		Per config	Per atom	Improv.	Per config	Per atom	Improv.
SOAPNet	Baseline	0.7706	0.0975	-	0.2153	0.0017	-
	+LA	0.5595	0.0704	27.58%	0.1786	0.0014	18.14%
	+MP	0.5717	0.0733	25.28%	0.1744	0.0014	19.12%
	+MP+LA	**0.5307**	**0.0657**	**31.88%**	**0.1697**	**0.0013**	**22.13%**
SchNet	Baseline	0.4805	0.0718	-	0.1693	0.0014	-
	+LA	0.4015	0.0549	19.99%	0.0845	0.0007	51.24%
	+MP	0.4034	0.0569	18.40%	0.1296	0.0010	26.00%
	+MP+LA	**0.3719**	**0.0490**	**27.17%**	**0.0816**	**0.0006**	**53.27%**

graphs. We select one representative potential from each class. For our MLP-based potential, we use the Smoother Overlap of Atomic Positions (SOAP) [2] atomic environment descriptor together with a representation module and prediction module consisting of MLPs; we term this potential SOAPNet in later discussions. For our GNN-based potential, we select SchNet [33]. In our label augmentation experiments, we set the representation module of the auxiliary classification model to be the same kind as the corresponding NN potential, e.g., the classification model used for training the SchNet potential has a SchNet GNN as its representation module. We choose sum pooling to be the readout function and an MLP to be the prediction module. Details about the model and training hyper-parameters can be found in Appendix A.2.

5.2.2 Selection of Physics-based EIPs

The EIPs used in our experiments were selected to encompass differing levels of functional complexity. Because EIPs are designed for specific elemental species (in this case, aluminum and silicon), a different set of EIPs had to be chosen for each dataset. A total of ten EIPs were used for aluminum and eight for silicon (see Tabs. 6 and 7). They were taken mainly from the Open Knowledgebase of Interatomic Models (OpenKIM) repository [1, 37, 38]. Most of the silicon EIPs were trained using atomic configurations similar to the bulk diamond configurations in our dataset. This raises the concern of information leakage, where the NN potentials indirectly learn information about the test set during training through supervision signals provided by the EIPs, since they were potentially trained on part of the test set. However, in the first two folds of our cross-validation, the perturbed diamond configurations constitute only 2.4% of the test set, and in the third fold constitute only 3.6%. In the case of aluminum, none of the EIPs used in our experiments were fitted to any configurations similar to those in the ANI-Al dataset. Altogether, this leads us to conclude that the impact of this effect is minimal.

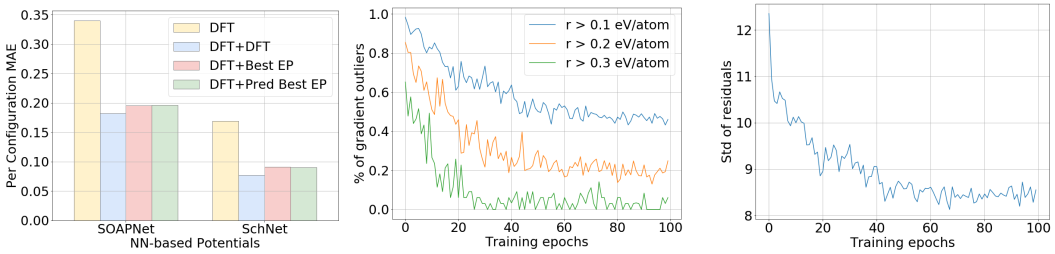
5.3 Experimental Results

5.3.1 Performance of Label Augmentation and Multi-task Pretraining

We test our proposed strategies with SOAPNet and SchNet. As shown in Tab. 1, our proposed strategies improve the performance of the two baseline NNs on both benchmark datasets. In particular, the label augmentation strategy improves the baseline NNs by 18% to 51%, while the multi-task pretraining strategy improves the baselines by 18% to 26%. Combining the two strategies gives further improvement by 22% to 53%.

5.3.2 EIP Energies as High-Quality Supervision Signals for Training NN Potentials

Recall that in the label augmentation strategy, an auxiliary classification model selects unlabeled configurations and predicts their corresponding best-performing EIPs. These configurations are then labeled by the predicted best-performing EIPs for NN potential training. We investigate the quality of EIP labels and the auxiliary classification model by training NN-potentials on three augmented



(a) (b) (c)

Figure 2: (a) Performance of NN potentials trained on the original training sets (yellow bar, DFT-labeled configurations only) and three augmented training sets whose unlabeled configurations are labeled by DFT energies (blue bar), ground-truth best-performing EIP energies (red bar), and predicted best-performing EIP energies (green bar). (b) Percentage of outliers used for computing gradient during training. (c) Standard deviation of residuals during training.

Table 2: Configuration-level MAE with and without the Tukey loss.

	KIM-Si		ANI-Al	
	SOAPNet	SchNet	SOAPNet	SchNet
w/o Tukey	0.5556	0.4374	0.1906	0.1031
w/ Tukey	0.5595	0.4015	0.1786	0.0845

training sets in which the selected unlabeled configurations are labeled by three different sources: DFT-based energies, ground-truth best-performing-EIP energies, and predicted best-performing EIP energies. The DFT-labeled configurations (0.8K) are the same for the three training sets. We only conduct this experiment on the ANI-Al dataset, as all of its configurations have DFT energies available. The number of selected unlabeled configurations is shown in Tab. 5.

Fig. 2a shows the performance of the NN potentials trained on the three augmented training sets and the original training set (where only DFT-labeled configurations are used). As shown in the figure, expanding the training set with ground truth DFT calculations (blue) greatly improves the baseline MAE (yellow bar, model trained on the original training set). Labeling configurations with the ground-truth best-performing EIPs (red bar) performs slightly worse than with DFT energies (blue bar) but still much better than the baseline (yellow bar). This demonstrates that EIPs are valuable sources of supervision signals for training NN-based potentials. Using the predicted best-performing EIPs for labeling (green bar) performs on par with the ground-truth best-performing EIP labeling (red), revealing the utility of the auxiliary classification model.

5.3.3 Importance of the Robust Tukey Loss

We next conduct experiments to investigate the importance of the Tukey loss and its ability to reject outliers during training. As before, we only conduct this experiment on the ANI-Al dataset, using DFT energies for unlabeled configurations to determine noise and outliers introduced by the predicted best-performing EIPs. We define an unlabeled configuration with a predicted best-performing EIP to be an outlier if the absolute difference between its EIP energy and its DFT energy is larger than a threshold, i.e., $|r_i| = \frac{1}{N_i}|E_i^{\text{DFT}} - E_i^{\hat{p}_i}| > c$ where N_i is the number of atoms in configuration i. We categorize the outliers as mild, normal, and severe by setting c to $\{0.1, 0.2, 0.3\}$. The initial number of outliers introduced by the unlabeled configurations can be found in Tab. 5. Figs. 2b and 2c show that the number of outliers of all kinds included for computing gradients decreases as the training proceeds, demonstrating that as the NN-potential gets progressively more accurate, the Tukey loss can effectively eliminate outliers from the training set.

We also conduct an ablation study by replacing the Tukey loss in Eq. 3 with the MSE loss. The results in Tab. 2 show that the models' performance degrades without the Tukey loss. The only exception is when SOAPNet is trained on the KIM-Si dataset, which we postulate is because the classification model does not introduce any influential outliers in this case.

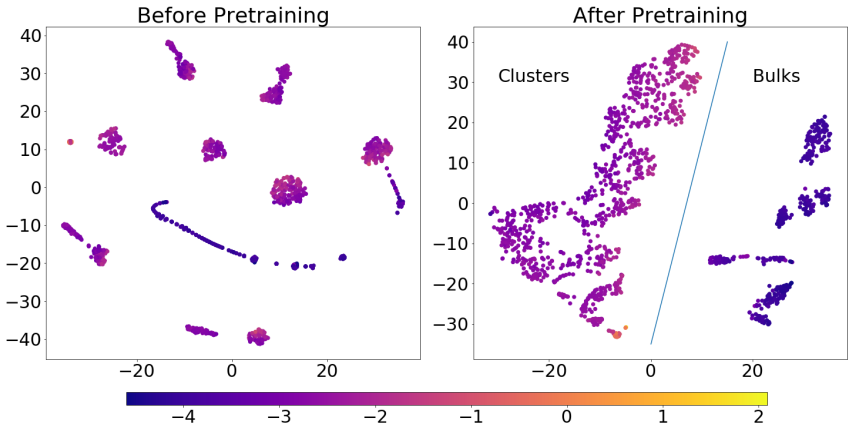


Figure 3: T-SNE plots of silicon configuration representations generated by a randomly initialized SchNet (left) and a SchNet pretrained with our proposed multi-task pretraining strategy (right).

5.3.4 Visualization of Pretrained Configuration Representations

Fig. 3 plots the t-SNE [40] 2D projections of the training silicon configuration representations colored by their per-atom DFT energies. The left-hand figure plots representations generated by a SchNet with random weights and the right-hand figure plots representations generated by a SchNet pretrained by the multi-task strategy. The representations generated by the random SchNet do not exhibit any clear patterns and the energy surface is rough. In the right-hand figure, representations of the atomic cluster configurations (i.e., isolated groups of atoms) and the bulk configurations (crystals) are clearly separated and form clusters in the t-SNE 2D space. The per-atom DFT energy surface of the right-hand figure is much smoother, i.e., configurations with similar energies are close to one another, after pretraining. This verifies our previous statement that, although EIPs lack complete generalizability, they nonetheless correlate reasonably well with DFT over atomic configuration space, and demonstrates that our proposed multi-task pretraining strategy successfully encodes domain knowledge into the NN-based potentials. T-SNE plots for SOAPNet on both datasets and SchNet on the ANI-Al dataset show similar patterns. These figures may be found in the Appendix A.3.

6 Conclusion

In this paper, we show that physics-based EIPs can be used to incorporate domain knowledge into machine learning EIPs by providing additional supervision signals on a large body of unlabeled training instances. Two generic strategies are formulated. The first, label augmentation, is based on weakly supervised learning and uses a classifier to select unlabeled training instances to supplement the original labeled training set. The second, multi-task pretraining, is based on transfer learning and uses the predictions of a set of physics-based EIPs on unlabeled training instances to pretrain a machine learning EIP that is subsequently fine-tuned on the original training set. Our methodology is proven using several experiments, including a cross-validation study that demonstrates significant performance gains for either strategy individually and further improvement when they are combined, a label augmentation study that shows that using EIP energies as surrogate labels provides nearly as much robustness as using additional DFT-labeled training instances, and a statistical experiment that illustrates the utility of the Tukey loss in diminishing noise and excluding outliers. Finally, we present reduced-dimension visualizations that indicate the multi-task pretraining strategy yields training set feature vectors over which the energy varies smoothly.

In this work, we only tested our approaches on systems with a single atomic species (i.e., Si or Al). Many real materials of interest consist of multiple species; for example, silicon is typically doped with other species in application. Therefore, an important direction for future work is to apply our approach to systems with multiple species. Another direction is to replace the heuristic EIPs by more accurate but still computationally cheap methods, such as the semi-empirical tight binding method. These methods better approximate DFT and thus can lead to further improvement when combined with our proposed approaches.

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Checklist

1. For all authors...
   (a) Do the main claims made in the abstract and introduction accurately reflect the paper's contributions and scope? [Yes]
   (b) Did you describe the limitations of your work? [Yes] See Section 6.
   (c) Did you discuss any potential negative societal impacts of your work? [N/A]
   (d) Have you read the ethics review guidelines and ensured that your paper conforms to them? [Yes]
2. If you are including theoretical results...
   (a) Did you state the full set of assumptions of all theoretical results? [N/A]
   (b) Did you include complete proofs of all theoretical results? [N/A]
3. If you ran experiments...
   (a) Did you include the code, data, and instructions needed to reproduce the main experimental results (either in the supplemental material or as a URL)? [Yes] We included in detail experimental settings in Section 5.2 and Appendix A.2 which is sufficient for reproducing the main experimental results. We will release our code if the paper gets accepted.
   (b) Did you specify all the training details (e.g., data splits, hyperparameters, how they were chosen)? [Yes] See Section 5.2 and Appendix A.2.
   (c) Did you report error bars (e.g., with respect to the random seed after running experiments multiple times)? [Yes] See Appendix A.3.
   (d) Did you include the total amount of compute and the type of resources used (e.g., type of GPUs, internal cluster, or cloud provider)? [Yes] See Section A.2.
4. If you are using existing assets (e.g., code, data, models) or curating/releasing new assets...
   (a) If your work uses existing assets, did you cite the creators? [Yes] We appropriately cited the source of datasets that we used in the paper, open source software we used in the experiments.
   (b) Did you mention the license of the assets? [N/A]
   (c) Did you include any new assets either in the supplemental material or as a URL? [No]
   (d) Did you discuss whether and how consent was obtained from people whose data you're using/curating? [N/A] One of the datasets we used was open sourced while the other one was curated by ourselves.
   (e) Did you discuss whether the data you are using/curating contains personally identifiable information or offensive content? [N/A]
5. If you used crowdsourcing or conducted research with human subjects...
   (a) Did you include the full text of instructions given to participants and screenshots, if applicable? [N/A]
   (b) Did you describe any potential participant risks, with links to Institutional Review Board (IRB) approvals, if applicable? [N/A]
   (c) Did you include the estimated hourly wage paid to participants and the total amount spent on participant compensation? [N/A]

A Appendix

A.1 Periodic Boundary Conditions

Under periodic boundary conditions (PBCs), the positions of atoms outside the simulation cell are obtained by generating periodic images of those within the cell through translations commensurate with its periodicity. This methodology is capable of modeling infinite systems because the interactions between atoms separated by more than a modest cutoff distance are very small and thus ignored when defining empirical models. This limited range of interaction gives rise to the concept of an *atomic environment*. The environment of a given atom consists of itself and all other atoms, including periodic images, that fall within a prescribed cutoff distance of it. The consequence of this locality is that an infinite system can be modeled exactly using a finite periodic cell so long as a sufficient number of periodic images surrounding it are explicitly accounted for. An example of PBCs for a two-dimensional square cell and a local atomic environment is illustrated in Fig. 4.

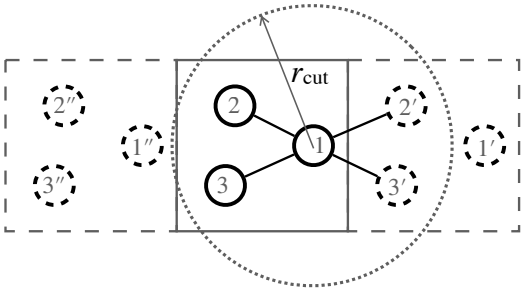

Figure 4: Illustration of periodic boundary conditions for a two-dimensional simulation cell (solid square) containing three atoms. For simplicity, only periodic images in the horizontal direction are shown. The local environment of atom 1 (dashed circle) contains all atoms and their periodic images that fall within a prescribed cutoff distance r_{cut} of it.

A.2 Experimental Settings

We set the number of hidden dimensions of all NNs to 128, the number of stacked NN layers in representation modules (GNNs and MLPs) to 5, and use the shifted softplus activation function in all nodes. All of our models are optimized using the Adam algorithm [25] with a learning rate of 1e-3 and a batch size of 32. For the multi-task pretraining and the training of the auxiliary classification model, we use a slated triangular scheduler [18] for the initial warm up of the weights, and subsequently decrease the learning rate linearly. For training the NNs with DFT energies, we decrease the learning rate linearly from 1e-3 to 1e-5. All NNs are trained for 100 epochs. Results are reported on the checkpoint with the best validation performance. We use validation performance to select the hyperparameter α that controls the contribution of EIP-labeled configurations (cf. Section 4.2) from [0.01, 0.05, 0.1, 0.5, 1, 5, 10].

Our code for the (G)NN potentials and experiments is implemented using PyTorch [30], DGL [43], and DGL-LifeSci [28]. All experiments are conducted on a machine with an Intel(R) Core(TM) i9-10900F CPU and an Nvidia RTX-3090 GPU. Pretraining SchNet on the ANI-Al and the KIM-Si dataset takes 1.5hr and 0.5hr, respectively, while retraining SOAPNet on both datasets takes a few minutes. The time cost of label augmentation depends largely on the number of unlabeled configurations added to the training set. In our case, experiments in all settings take less than 2hrs to finish.

A.3 Additional Experimental Results

A.3.1 Performance of the Two Strategies with Standard Deviation

We report performance of the two strategies on an average over three runs on three random splits and report the mean/std in Tab 3

Table 3: Performance of the two proposed strategies on DFT energy prediction tasks. We report the configuration-level and atom-level mean absolute error (MAE, lower the better) in eV and eV/atom, respectively. We denote the label augmentation strategy by LA and the multi-task pretraining strategy by MP. Standard deviation shown in the brackets.

		KIM-Si		ANI-Al	
		Per config	Per atom	Per config	Per atom
SOAPNet	Baseline	0.7706 (0.0658)	0.0975 (0.0012)	0.2153 (0.0228)	0.0017 (0.0002)
	+LA	0.5595 (0.0336)	0.0704 (0.0026)	0.1786 (0.0239)	0.0014 (0.0002)
	+MP	0.5717 (0.0232)	0.0733 (0.0038)	0.1744 (0.0249)	0.0014 (0.0002)
	+MP+LA	0.5307 (0.0375)	0.0657 (0.0033)	0.1697 (0.0193)	0.0013 (0.0002)
SchNet	Baseline	0.4805 (0.0345)	0.0718 (0.0096)	0.1693 (0.0106)	0.0014 (0.0001)
	+LA	0.4015 (0.0687)	0.0549 (0.0042)	0.0845 (0.0095)	0.0007 (0.0001)
	+MP	0.4034 (0.0723)	0.0569 (0.0115)	0.1296 (0.0164)	0.0010 (0.0001)
	+MP+LA	0.3719 (0.0706)	0.0490 (0.0059)	0.0816 (0.0064)	0.0006 (0.0001)

Table 4: MAE on training sets expanded by configurations with different confidence level.

		KIM-Si		ANI-Al	
	Confidence	Per config	Per atom	Per config	Per atom
SOAPNet	Low	0.5793	0.0732	0.2174	0.0017
	Medium	0.5728	0.0725	0.2133	0.0017
	High	0.6196	0.0778	0.2361	0.0019
SchNet	Low	0.4817	0.0657	0.1140	0.0009
	Medium	0.4916	0.0668	0.1130	0.0009
	High	0.5004	0.0740	0.1242	0.0010

A.3.2 Effect of Expanding Training Set with Different Confidence Level

We expand the training set with DFT energies with EIP-labeled configurations with different confidence level computed by the auxiliary classification model, i.e., $P(\hat{p} \mid C)$ (see Section 4.2). We first sort the configurations by their confidence scores and assign them to different confidence groups. Configurations with a confidence score higher than the 0.66 quantile are assigned to high confidence group, configurations with a confidence score lower than the 0.33 quantile are assigned to low confidence group, other configurations are assigned to the medium confidence group. We expand the DFT-labeled configurations to three different training sets by adding configurations belong to different groups. Results are shown in Tab. 4. The results suggest that the unlabeled configurations with medium confidence contribute the most to the training while configurations with high confidence contribute the least. This is because that the high confidence configurations may be very similar to the configurations to the training set and do not provide extra information. Low and medium confidence configurations are more different than those in the original training set and can provide more information.

A.3.3 Number of Outliers and Configurations Selected By the Classification Model

We show the number of configurations and outliers introduced by the auxiliary classification model in Tab. 5. Results in the table show that the auxiliary classification model reaches an accuracy of 75% to 78% for screening configurations and selecting reasonable EIPs for labeling them. This demonstrates the utility of the classification model.

Table 5: Average number of configurations and outliers selected by the classification models on ANI-Al dataset.

	#Mild	#Normal	#Severe	#Selected	#Unlabeled
SOAPNet	226	42	5	1211	5081
SchNet	271	50	9	1300	5081

A.3.4 Visualization of Pretrained Configuration Representations

We show the t-SNE 2D projection plots for all datasets and NN potentials in Figs. 5, 6, and 7. These plots show that the multi-task pretraining successfully inject domain knowledge into the representation module of NN potentials and create smoother DFT energy surface.

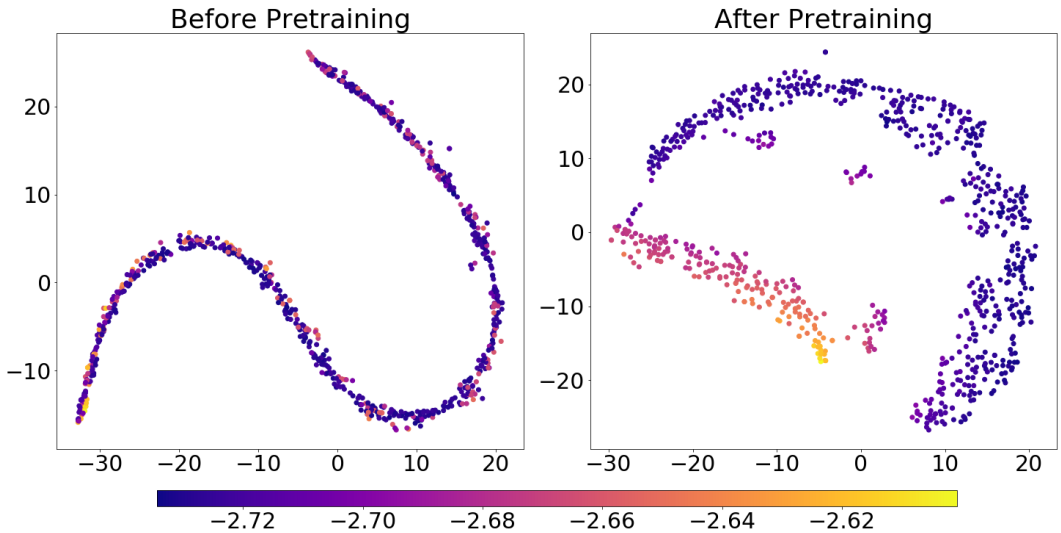

Figure 5: T-SNE plots of aluminum configuration representations generated by a randomly initialized SchNet (left) and a SchNet pretrained with our proposed multi-task pretraining strategy (right).

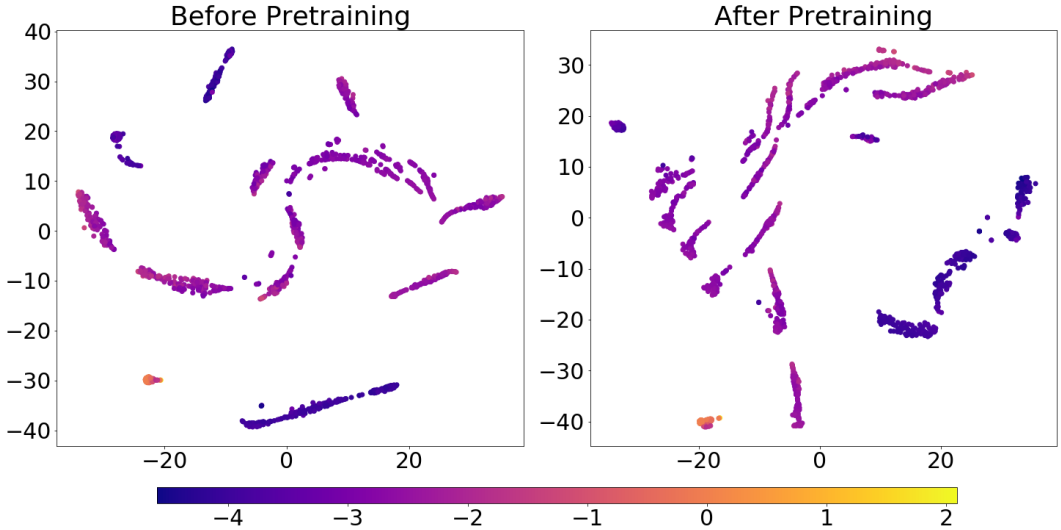

Figure 6: T-SNE plots of silicon configuration representations generated by a randomly initialized SOAPNet (left) and a SOAPNet pretrained with our proposed multi-task pretraining strategy (right).

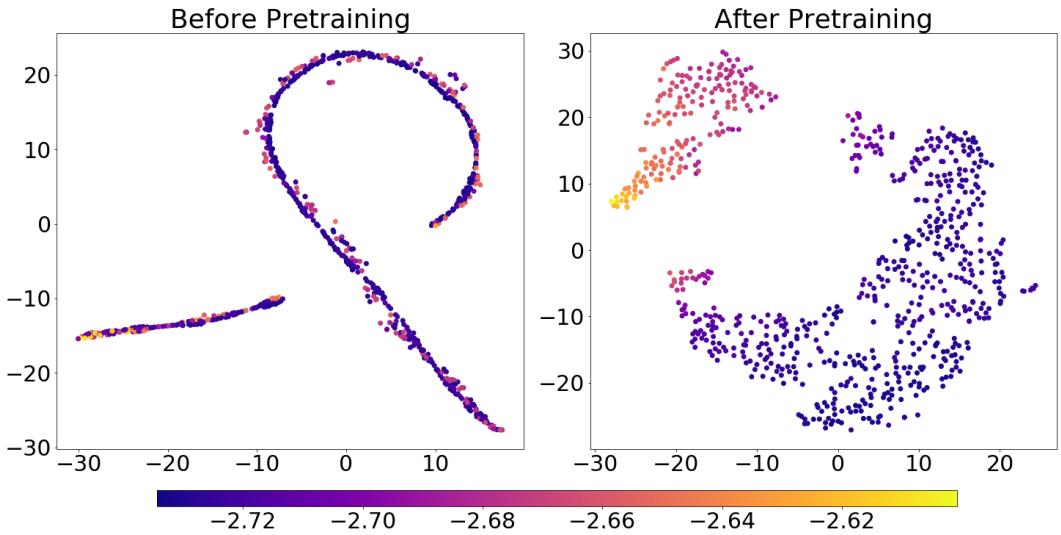

Figure 7: T-SNE plots of silicon configuration representations generated by a randomly initialized SOAPNet (left) and a SOAPNet pretrained with our proposed multi-task pretraining strategy (right).

A.4 EIPs used in Experiments

Table 6: EIPs employed for the KIM-Si dataset

EIP name	Link
EDIP	https://doi.org/10.25950/545ca247
MEAM	https://doi.org/10.25950/b8dc8b23
SW (BalamaneHaliciogluTiller)	https://doi.org/10.25950/3dc2cb7f
SW (ZhangXieHu)	https://doi.org/10.25950/32a4bf2c
Tersoff (T2)	https://doi.org/10.25950/cadc4e78
Tersoff (T3)	https://doi.org/10.25950/d6e8a23e
Tersoff (ErhartAlbe)	https://doi.org/10.25950/6aa22835
Tersoff (TMOD)	https://doi.org/10.1016/j.commatsci.2006.07.013

Table 7: EIPs employed for the ANI-Al dataset

EIP name	Link
EMT	https://doi.org/10.25950/bdbaee6a
Morse (LowCutoff)	https://doi.org/10.25950/977dc2ac
Morse (MedCutoff)	https://doi.org/10.25950/474ccb33
Morse (HighCutoff)	https://doi.org/10.25950/45d9848f
EAM (ErcolessiAdams)	https://doi.org/10.25950/bc2d2486
EAM (SturgeonLaird)	https://doi.org/10.25950/d62edb43
EAM (WineyKubotaGupta)	https://doi.org/10.25950/23542694
EAM (ZopeMishin)	https://doi.org/10.25950/26dbac6e
EAM (Zhakhovsky)	https://doi.org/10.25950/c3a79c52
EAM (ZhouJohnsonWadley)	https://doi.org/10.25950/c775fc98