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Prediction of Interatomic Potentials Combining Empirical Potential and Graph Neural Networks

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Abstract

Molecular dynamics (MD) simulation is constrained by the time and spatial scales as well as computational efficiency of traditional methods. While machine learning interatomic potentials (MLIPs) based on graph neural networks (GNNs) improve modeling accuracy, they suffer from insufficient short-range interaction modeling and lack of physical constraints. This paper proposes the DimezblNet model, which explicitly embeds the ZBL empirical potential function into the DimeNet framework. Through physically guided hybrid potential modeling, the description of short-range repulsive interactions is strengthened. On the MD17 dataset, the model reduces the mean absolute error (MAE) of energy and atomic force predictions for molecules like aspirin by 3.7%-5.3% compared to DimeNet. In the QM9 molecular property prediction task, the MAE of polarizability (α) is 0.0452 Å³, outperforming DimeNet (0.0469 Å³, a 3.6% improvement), and the MAE of dipole moment (μ) is 0.0273 D. Experiments show that combining physical priors with data-driven strategies significantly enhances the model's generalization ability and interpretability in complex molecular systems.

Keywords

molecular dynamics simulation, graph neural network, ZBL potential, machine learning interatomic potential

1. Introduction

The origin of molecular dynamics (MD) simulation can be traced back to the 1950s. In 1957, Berni Alder and Tom Wainwright first proposed it through computer simulations of gas-liquid-solid phase transitions in hard-sphere systems (Alder & Wainwright, 1957).

Molecular dynamics simulation is a computer modeling technique that tracks the microscopic trajectories of atomic/molecular systems over time by numerically integrating Newton's Formulas of motion under predefined interaction potentials, thereby predicting the dynamic characteristics and mechanisms of materials or biomolecules at the atomic scale. It is widely applied in multiple scientific and engineering fields, such as biomedicine, materials science and engineering, energy and environment, chemistry and chemical engineering (Huang & Von Lilienfeld, 2021; Keretsu et al., 2020).

Although molecular dynamics simulation has achieved remarkable results in many fields, traditional molecular dynamics simulation still faces key limitations. Constrained by the numerical integration

algorithm of classical Newtonian mechanics, traditional MD can only simulate processes on the nanosecond to microsecond time scale, struggling to cover phenomena with longer time scales like protein folding and material deformation. Moreover, due to the $O(N^3)$ complexity of inter-particle interaction calculations, simulation scales are mostly limited to nanometer systems, making it difficult to fully capture the behavior of macroscopic materials or complex biological systems.

In recent years, with the development of machine learning, machine learning interatomic potentials (MLIPs) have emerged as a new approach to balance accuracy, universality, and efficiency in describing the energy of atomic assemblies for atomic simulations. MLIPs enable machine learning models to accurately predict the properties of chemical systems by leveraging training data generated from ab initio methods. The MLIPs approach combined with graph neural networks (GNNs) is currently a popular method for predicting interatomic potentials. GNNs do not require manual feature engineering but learn molecular representations entirely based on atomic types and coordinates. By aggregating information from adjacent nodes to update node features, GNNs model local interactions, demonstrating good scalability for large-scale molecular systems. Compared with traditional empirical force fields, models based on GNNs for potential energy surface modeling can achieve higher prediction accuracy by learning high-precision quantum mechanical data and exhibit better generalization across different molecules or structures.

However, GNN-based models often lack physical interpretability and constraints, leading to suboptimal prediction accuracy. To address this, this study enhances the short-range interaction modeling capability of the DimeNet (Gasteiger et al., 2020) model by explicitly adding a ZBL (Ziegler & Biersack, 1985) potential calculation module. The ZBL potential is an empirical potential function describing repulsive interactions between atoms at short distances. DimeNet itself is designed to focus on angular information and medium-range interactions, showing insensitivity to strong short-range repulsion. Introducing the ZBL potential explicitly compensates for this short-range interaction modeling, improving the model's prediction accuracy.

2. Related Works

GNNs are neural networks applied to graph data, leveraging graph structural information to enhance learning task performance. In quantum chemistry applications, graph neural networks typically use nodes to represent atoms and edges to represent connections between central atoms and neighboring atoms within a cutoff radius, forming an atomic graph. The message-passing mechanism of graph neural networks (Gilmer et al., 2017) serves as a general framework for graph neural networks, and such networks are commonly referred to as message-passing neural networks (MPNNs) (Gilmer et al., 2017). They aggregate information from neighboring atoms (including nodes and edges) to update the state of the central atom. After multiple iterations, the influence of more distant atoms on the central atom can also be considered, helping the model better learn the inherent features of the atomic graph.

MPNN is a general framework for graph neural networks, replacing hand-designed distance and angle features with trainable operators dependent only on atomic charges and positions to directly learn representations of heterogeneous chemical environments from training data (Bartók et al., 2010; Gilmer et al., 2017; Zhang et al., 2018). In the MPNN approach, SchNet (Schütt et al., 2017) first modeled 3D atomic systems as continuous-filter convolutional neural networks, encoding atomic distances through radial basis functions to enable efficient learning of quantum mechanical data. DimeNet innovatively introduced a directional message-passing mechanism, explicitly modeling directional interactions between atoms through bond angle embedding. GemNet (Gasteiger et al., 2021) further integrates four-dimensional diagonal tensors to achieve physically accurate modeling of higher-order interactions.

Recently, graph neural network models based on rotational invariance constraints have gradually become the core framework in the deep learning field of interatomic interactions, owing to their symmetry preservation capabilities and high-precision modeling advantages. To satisfy the isotropy principle of physical systems, advanced models embed E(3) group symmetry (translation/rotation/reflection invariance) to maintain coordinate invariance in potential energy predictions, effectively avoiding the low prediction accuracy caused by ignoring symmetry in traditional methods. DimeNet takes atomic distances and bond angles as inputs, encodes angular information through spherical harmonic functions, and converts direction-

sensitive intermediate features into rotation-invariant total energy outputs using invariant pooling (such as atomic summation). Based on DimeNet, GemNet introduces three-dimensional geometric features like dihedral angles, encodes directional information through spherical harmonic-Fourier basis functions, and ensures strictly rotation-invariant final energy predictions by leveraging symmetric aggregation and the inherent invariance of geometric functions. NequIP (Batzner et al., 2022), based on an E(3)-equivariant architecture, preserves direction sensitivity in intermediate layers through tensor features and equivariant convolutions, and finally achieves strictly rotation/translation invariance via scalar head aggregation, enhancing modeling accuracy while ensuring physical symmetry.

This paper constructs a physically guided hybrid potential model by introducing an explicit ZBL potential calculation module into DimeNet. By explicitly constraining short-range strong repulsive interactions with physical potentials, it effectively compensates for extrapolation biases in the model caused by missing data in extreme short-range regions, avoids unphysical energy oscillations when nuclear distances are too close, further enhances the model's physical interpretability, and improves prediction accuracy.

3. Introduction to the DimezblNet Model

The implementation of a general message-passing layer represents each atom i with an atomic embedding $h_i \in R^H$. Atomic embeddings are updated by passing messages along molecular edges in each layer. Messages are typically transformed based on edge embeddings $e_{(ij)} \in R^{H_e}$ and summed over the neighbor atoms j of the central atom i ($j \in N_i$, where N_i is the neighborhood of atoms), as shown in Formula (1) and (2):

$$M_i^{(l+1)} = \sum_{j \in \mathrm{N}_i} f_{\mathrm{int}}\left(h, \mathbf{e}_{(ij)}^{(l)}\right) \tag{1}$$

$$h_i^{(l+1)} = U_{\mathrm{upd}}\left(h_i^{(l)}, M_i^{(l+1)}\right) \tag{2}$$

In Formula (1), $M_i^{(l+1)}$ is a message aggregation function. It collects information from the neighborhood N_i by using a function f_{int}, and this function depends on the current atomic embeddings and the edge feature $e_{(ij)}$. $e_{(ij)}$ usually only depends on the inter - atomic distance, but it can also incorporate additional bond information. In Formula (2), U_{upd} is a learnable message update function used to process the message aggregation in Formula (1).

Since the DimezblNet model considers rotational invariance in the message-passing layer and achieves directional information transfer, new definitions are given to the aggregation and update functions of the message-passing layer, as shown in Formula (3):

$$h_{ji}^{(l+1)} = U_{\mathrm{upd}}(h_{ji}^{(l)}, \sum_{k \in \mathrm{N}_j ? \{i\}} f_{\mathrm{int}}(h_{kj}^{(l)}, e_{RBF}^{(ji)}, a_{SBF}^{(kj,ji)})) \tag{3}$$

Among them, $e_{RBF}^{(ji)}$ represents the distance between the central atom i and the neighboring atom j. The radial basis representation of d_{ij} is shown in Formula (4):

$$\hat{e}_{\mathrm{RBF},n}(d) = \sqrt{\frac{2}{r_c}} \frac{\sin\left(\frac{n\pi}{r_c} d\right)}{d} \tag{4}$$

Among them, $n \in [1,\ldots,N_{\mathrm{RBF}}]$ represents the range of radial quantum numbers, r_c is the cutoff radius, and $\sin\left(\frac{n\pi}{r_c}d\right)$ is a sine function, representing the oscillatory behavior of the radial basis. $a_{SBF}^{(kj,ji)}$ is a two - dimensional representation based on spherical Bessel functions and spherical harmonics. It is a two - dimensional joint representation of the angular information $\alpha_{(kj,ji)} = \angle \mathbf{x}_k \mathbf{x}_j \mathbf{x}_i$ and the inter - atomic distance d_{ij}, as shown in Formula (5):

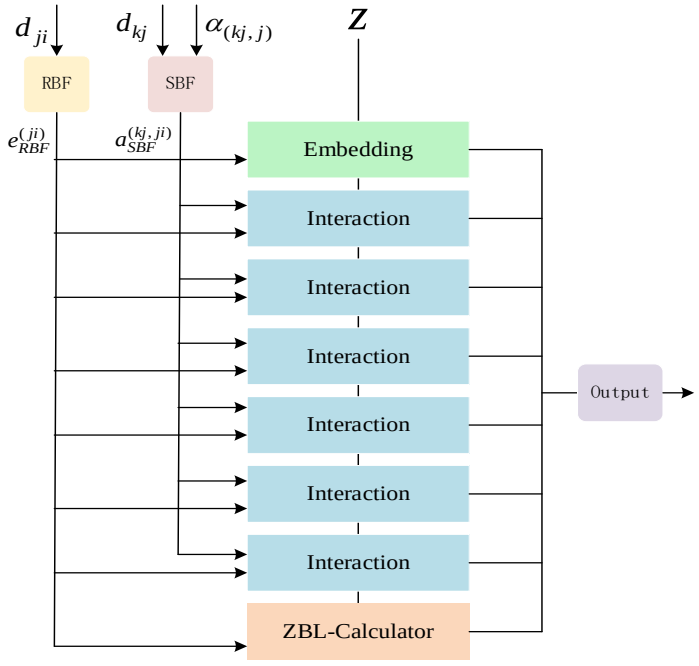
$$\hat{a}_{\mathrm{SRF},ln}(d,\alpha) = \sqrt{\frac{2}{r_c^{\,3} j_{l+1}^2(z_{ln})}} j_l\left(\frac{z_{ln}}{r_c}d\right) Y_l^0(\alpha) \tag{5}$$

Among them, $l \in [1,\ldots,N_{\mathrm{RBF}}]$ represents the range of angular quantum numbers for the j_l type of Bessel function and the Y_l^0 spherical harmonic function. $j_{l+1}(z_{ln})$ is the value of the $l+1$ order Bessel function at z_{ln}. z_{ln} is the n root of the l-th order Bessel function. For $e_{RBF}^{(ji)}$ and $a_{SBF}^{(kj,ji)}$, since the step function is not twice continuously differentiable at the cutoff radius r_c, a polynomial envelope function f_{env} is used as shown in Formula (6). It has a multiple root of 3 at $d = r_c$, so that the first - order and second - order derivatives of $e_{RBF}^{(ji)}$ and $a_{SBF}^{(kj,ji)}$ tend to 0 at the cutoff point.

$$f_{env}(d) = 1 - \frac{(p+1)(p+2)}{2}d^{\,p} + p(p+2)d^{\,p+1} - \frac{p(p+1)}{2}d^{\,p+2} \tag{6}$$

Figure 1 shows the architecture of the DimezblNet model, which includes one RBF layer, one SBF layer, one Embedding layer, six Interaction layers, one ZBL-Calculator layer, and one Output layer.

Figure 1: Architecture of the DimezblNet model



There shows the architecture of the DimezblNet model, which includes one RBF layer, one SBF layer, one Embedding layer, six Interaction layers, one ZBL-Calculator layer, and one Output layer.

3.1 Embedding Layer

The atomic number is represented by learnable, randomly initialized atomic type embeddings $Z_i^{(0)} \in ¡^{F}$, and these embedded atomic sequences are shared across molecules. The message input generated by the atomic number and the inter - atomic distance is shown in Formula (7):

$$h_{ji}^{(1)} = \sigma([Z_j^{(0)} \| Z_i^{(0)}]e_{\mathrm{RBF}}^{(ji)}\mathbf{W} + \mathbf{b}) \tag{7}$$

Where the weight matrix W and bias b are learnable.

3.2 Interaction Layer

The interaction layer here is the implementation of the aggregation function and update function in Formula 3. Among them, the two - dimensional representation $a_{SBF}^{(kj,ji)}$ is transformed into an N_{linear} - dimensional representation through a linear layer. The purpose of this processing is to make the dimension of $a_{SBF}^{(kj,ji)}$ independent of the subsequent bilinear layer. The subsequent bilinear layer uses a larger $N_{\mathrm{linear}} \times F \times F$ - dimensional weight representation. For the representation of the radial basis, the way of using element - wise multiplication $e_{\mathrm{RBF}}^{(ji)}W$ e h_{kj} is superior to the bilinear layer.

3.3 ZBL-Calculator Layer

This layer implements the calculation of ZBL potential, with the formula shown in Formula (8):

$$v_{zbl} = \frac{Z_i Z_j}{d_{ij}} \cdot \psi(x) \cdot \frac{e_i e_j}{4\pi\varepsilon_0} \tag{8}$$

Among them, $\frac{e_i e_j}{4\pi\varepsilon_0}$ is a physical scale factor in the ZBL potential energy calculation formula that can reflect the strength of the charge interaction. $\psi(x)$ is a screening function, and its purpose is to simulate the weakening effect caused by the overlap of electron clouds at short distances for the Coulomb repulsion between two atomic nuclei, as shown in Formulas (9) and (10).

$$\psi(x) = c_1 e^{d_1 x} + c_2 e^{d_2 x} + c_3 e^{d_3 x} + c_4 e^{d_4 x} \tag{9}$$

$$x = \frac{(Z_i^{0.23} + Z_j^{0.23}) \cdot d_{ij}}{a_0} \tag{10}$$

Where c_n, d_n, a_0 are parameters fitted through experiments and first-principles calculations.

3.4 Output Layer

The message embeddings of each layer are passed to the output layer. The output layer uses the radial basis function $e_{\mathrm{RBF}}^{(ji)}$ to transform each message embedding h_{ji}, which ensures continuous differentiability. The incoming messages of each atom i are accumulated to obtain $H_i = \sum_j h_{ji}$, and then multiple dense layers are used for transformation to generate the atomic - level output $v_i^{(l)} + v_{zbl}$. Subsequently, they are accumulated again to obtain the final result $V = \sum_i \sum_l v_i^{(l)}$. The force is obtained from the negative gradient of the total energy relative to the atomic positions, as shown in Formula (11):

$$\vec{F}_i = -\nabla_i V_{\mathrm{pot}} \tag{11}$$

4. Experimental Analysis

4.1 Evaluation Metrics

To evaluate the model's performance, this paper uses the mean absolute error (MAE) for each metric.

4.2 QM9

The QM9 (Ramakrishnan et al., 2014; Ruddigkeit et al., 2012) dataset was used to benchmark the performance of DimezblNet in predicting molecular properties. QM9 consists of approximately 130,000 molecules in equilibrium. In model training, 11,000 training samples, 10,000 validation samples, and 10,831 test samples were used. The results were compared with the reported results of PPGN (Maron et al., 2019), SchNet, PhysNet (Unke & Meuwly, 2019), Cormorant (Anderson et al., 2019), and DimeNet, as shown in Table 1.

Table 1: Mean Absolute Errors (MAE) and Units of Prediction Targets in QM9

Target	Unit	PPGN	SchNet	PhysNet	Cormorant	DimeNet	DimezblNet
μ	D	0.017	0.033	0.0529	0.13	0.0286	0.0273
α	a_0^3	0.131	0.235	0.0615	0.092	0.0469	0.0452
ε_{HOMO}	meV	40.3	41	32.9	36	27.8	26.7
ε_{LUMO}	meV	32.7	34	24.7	36	19.7	18.9
$\Delta\varepsilon$	meV	60	63	42.5	60	34.8	33.1
R^2	a_0^2	0.592	0.703	0.75	0.673	0.231	0.217
ZPVE	meV	3.12	1.7	1.395	1.98	1.29	1.23
U^0	meV	36.8	18	14	13	8.29	8.07
U	meV	36.8	19	14	–	7.89	7.63
H	meV	36.3	14	14	–	8.11	7.92
G	meV	36.4	14	14	–	8.98	8.71
c_v	(K*cal)/mol	0.055	0.033	0.028	0.031	0.0249	0.0237

4.3 MD17

The MD17 (Chmiela et al., 2017) dataset was used to test the performance of DimezblNet in molecular dynamics simulation. This benchmark aims to predict the energy and atomic forces of eight small organic molecules, with given atomic coordinates of thermalized (i.e., non-equilibrium, slightly perturbed) systems. The ground-truth data were calculated through molecular dynamics simulations using density functional theory (DFT). In model training, 1,000 training samples and 10,000 validation/test samples were used. The results were compared with the reported results of sGDML (Chmiela et al., 2019), SchNet, and DimeNet, as shown in Table 2.

Table 2: Mean Absolute Errors (MAE) of Energies (kcal mol^{-1}) and Forces ($\text{kcal mol}^{-1}\text{Å}^{-1}$) for 8 Small Organic Molecules in MD17

Molecule		SGDML	SchNet	DimeNet	DimezblNet
Aspirin	Energy	0.19	0.37	0.204	0.194
	Forces	0.68	1.35	0.499	0.473
Benzene	Energy	0.1	0.08	0.078	0.075
	Forces	0.06	0.31	0.187	0.181
Ethanol	Energy	0.07	0.08	0.064	0.062
	Forces	0.33	0.39	0.23	0.217
Malonaldehyde	Energy	0.10	0.13	0.104	0.098
	Forces	0.41	0.66	0.383	0.367
Naphthalene	Energy	0.12	0.16	0.122	0.117
	Forces	0.11	0.58	0.215	0.208
Salicylic acid	Energy	0.12	0.2	0.134	0.127
	Forces	0.28	0.85	0.374	0.361

Toluene	Energy	0.1	0.12	0.102	0.097
	Forces	0.14	0.57	0.216	0.207
Uracil	Energy	0.11	0.14	0.115	0.107

5. Conclusion

This study constructs the DimezblNet model with enhanced physical constraints by integrating the ZBL empirical potential and graph neural networks, effectively addressing the modeling bias of short-range atomic interactions. On the MD17 dataset, the model's prediction accuracy for energy and atomic forces comprehensively surpasses models like SchNet and DimeNet. In QM9 molecular property prediction, the errors of polarizability (α) and dipole moment (μ) are reduced to 0.0452 $Å^3$ and 0.0273 D, respectively, verifying the advantages of DimezblNet in electron structure-sensitive properties. This method not only improves prediction accuracy but also enhances model interpretability through explicit physical potential constraints, providing an efficient tool for multi-scale modeling in material design and biomolecular simulation.

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Conflicts of Interest

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