Machine Learning Potentials for DFT I

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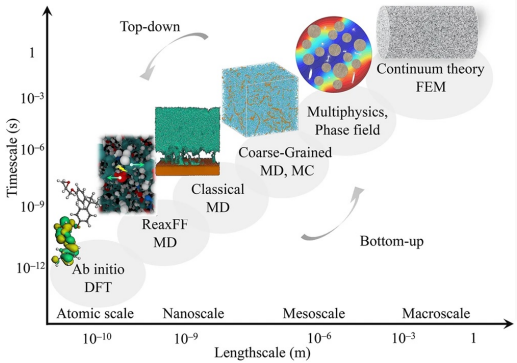
Overview

Computational Materials Science

Simulation studies offer deep insights into the properties and behavior of materials at various scales.



K. Lin and Z. Wang, Commun. Mater., 4 (2023), 1–16.

Computational Methods for Materials Simulation

- Scalable across time and length scales.
- Applied to simulate material properties and behaviors.
- Supports the design of new materials with specific properties.

DFT calculations become computationally expensive as the number of sites increases.

Atomistic Simulation Methods

- In molecular dynamics (MD), the total forces on all the atoms are calculated and Newton's equations of motion are solved to determine how the atoms move in response to those forces.

$$\boldsymbol{F}_i = m_i \boldsymbol{a}_i = m_i \frac{d\boldsymbol{v}_i}{dt} = m_i \frac{d^2 \boldsymbol{r}_i}{dt^2}$$

- The force on an atom is derived from the gradient of the potential energy of the system, U

$$\begin{aligned}
\boldsymbol{F}_i &= -\nabla_i U\left(\boldsymbol{r}_1, \boldsymbol{r}_2, \ldots, \boldsymbol{r}_N\right) \\
&= -\nabla_i U\left(\boldsymbol{r}^N\right) \\
&= -\left(\frac{\partial U\left(\boldsymbol{r}^N\right)}{\partial x_i}\hat{x} + \frac{\partial U\left(\boldsymbol{r}^N\right)}{\partial y_i}\hat{y} + \frac{\partial U\left(\boldsymbol{r}^N\right)}{\partial z_i}\hat{z} \right)
\end{aligned}$$

B.J.Alder,T.E.Wainwrigh, J. Chem. Phys., 27 (1957) 1208–1209.

- Verlet algorithm uses a set of positions at t and $t - \Delta t$, to obtain the positions of atoms at time $t + \Delta t$ with an error as low as $\mathcal{O}(\Delta t^4)$.

$$\mathbf{r}_i(t + \Delta t) = 2\mathbf{r}_i(t) - \mathbf{r}_i(t - \Delta t) + \frac{\Delta t^2}{m_i}\mathbf{F}_i(t) + \mathcal{O}(\Delta t^4)$$

- Using a pairwise interatomic potential, the potential energy of an N-atom system can be calculated as:

$$U(\mathbf{r}^N) = \frac{1}{2}\sum_{i=1}^{N}\sum_{j\neq i}^{N}\varphi_{ij}(r_{ij}),$$

- Since the accuracy of interatomic potentials directly impacts the reliability of MD simulation results, their careful selection and validation are essential for any MD study.

L. Verlet, Phys. Rev., 159 (1967) 98–103.

Atomistic Simulation Methods

Monte Carlo (MC)

- The Monte Carlo (MC) method, originally developed in the 1940s, is a powerful computational tool to model and predict the behavior of materials at the atomic and molecular levels.
- Using the Metropolis algorithm, possible configurations of a system are randomly sampled for evaluation:

$$\Delta E = E(\mathbf{r}'^N) - E(\mathbf{r}^N)$$

- The new configuration is accepted based on a probability calculated using:

$$P_{\text{accept}} = \min\left(1, \exp\left(-\frac{\Delta E}{k_B T}\right)\right)$$

- Once the system reaches equilibrium after sufficient MC steps, thermodynamic properties can be computed as ensemble averages:

$$\langle A \rangle = \frac{1}{M} \sum_{i=1}^{M} A(\mathbf{r}_i^N)$$

N.Metropolis, S.Ulam, JASA, 44 (1949) 335–341.

N. Metropolis, A. W. Rosenbluth, M. N. Rosenbluth, A. H. Teller, E. Teller, J. Chem. Phys., 21 (1953) 1087–1092.

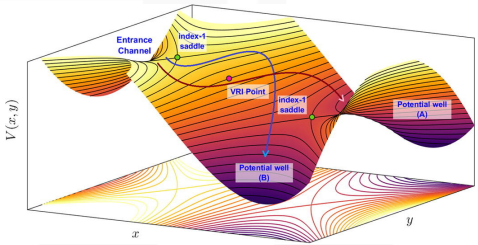
Potential Energy Surface (PES)

Foundation for Atomistic Simulations

- Energy as a function of atomic positions.
- Defines equilibrium structures and reaction pathways.
- Multidimensional landscape of molecular systems.

Importance in Simulations

- **MD:** Atoms move across the PES using Newton's laws.
- **MC:** Samples configurations from the PES for thermodynamic properties.



Potential Energy Surface (PES)

García-Garrido, V.J., M. Katsanikas, M. Agaoglou, and S. Wiggins, 754, Chem. Phys. Lett., (2020): 137714.

Traditional Interatomic Potentials

Traditional Interatomic Potentials

Lennard-Jones Potential

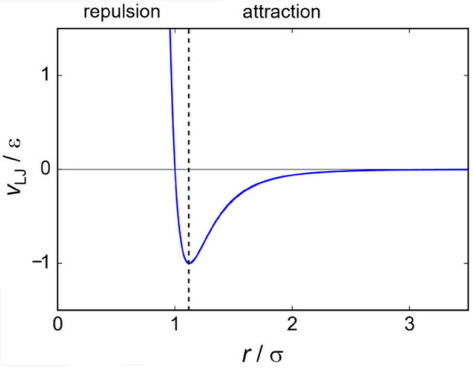
- Simple potential used to describe interactions between atoms or molecules.
- Depends on two parameters: ϵ and σ.

short-range repulsive interactions

$$\varphi_{\mathrm{LJ}}(r) = 4\epsilon \left[\left(\frac{\sigma}{r}\right)^{12} - \left(\frac{\sigma}{r}\right)^{6} \right]$$

long-range attractive interactions

- Parameters are determined using empirical or semi-empirical methods.



Lennard-Jones (12,6) Potential

J. Lenhard, S. Stephan, H. Hasse, Annalen der Physik 536 (2024) 2400115.

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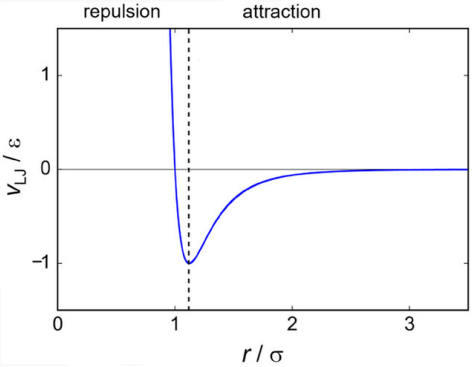

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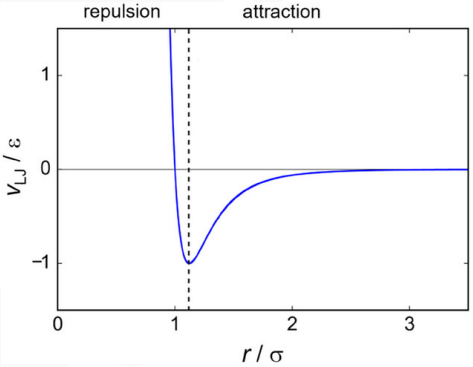

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Embedded Atom Method (EAM)

- Simple pair potentials cannot describe the delocalized valence electrons of metals.
- Within EAM, energy of an atom in a metal is influenced by both pairwise interactions and local electron density.

$$\varphi_{\mathrm{EAM}} = \sum_i F_i(\bar{\rho}_i) + \frac{1}{2}\sum_{i \neq j} \phi_{ij}(r_{ij})$$

embedding functional

pairwise interactions

- The local electron density can be calculated as the sum of contributions of neighboring particles into the electron density.

$$\bar{\rho}_i = \sum_{j \neq i} \rho_j \left(\mathbf{r}_{ij}\right)$$

local electron density

M. S. Daw and M. I. Baskes, Phys. Rev. B, 29 (1984), 6443–6453.

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Traditional Interatomic Potentials

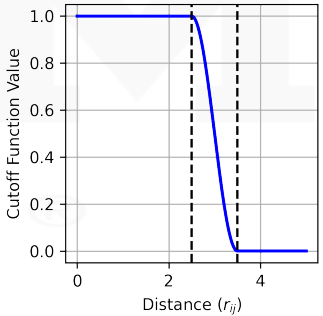
Tersoff Potential

- Developed to describe covalent bonds, such as in carbon and silicon.
- Incorporates angular dependence to capture the directionality of bonds.

$$\varphi_{\text{Tersoff}} = \frac{1}{2} \sum_i \sum_{j \neq i} f_c(r_{ij}) \left[A \exp\left(-\lambda_1 r_{ij}\right) - b_{ij} B \exp\left(-\lambda_2 r_{ij}\right) \right]$$

- The positive term in the Tersoff potential represents short-range Pauli repulsion, while the negative term accounts for long-range bonding attraction.
- The cutoff function smoothly decays from 1 to 0.

$$f_c(r_{ij}) = \begin{cases} 1 & \text{if } r_{ij} < R_1 \\ \frac{1}{2}\left[1 + \cos\left(\frac{\pi(r_{ij} - R_1)}{R_2 - R_1}\right)\right] & \text{if } R_1 \leq r_{ij} \leq R_2 \\ 0 & \text{if } r_{ij} > R_2 \end{cases}$$



J. Tersoff, Phys. Rev. B, 37 (1988), 6991–7000.

Limitations of Traditional Potentials

Key Drawbacks

- While these potentials have been successful in various applications, they face significant limitations:

Limited Accuracy

- Inadequate for many-body systems.
- Poor precision in predicting material properties.

Limited Transferability

- Fitted for specific materials/conditions.
- Poor performance under varying conditions.

Inability to Capture Complex Interactions

- Misses chemical bonding and magnetic effects.
- Fails to account for long-range forces.

Human Dependence

- Relies on human expertise and fixed functional forms.
- Often requires manual tuning for improvements.

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Machine Learning Interatomic Potentials

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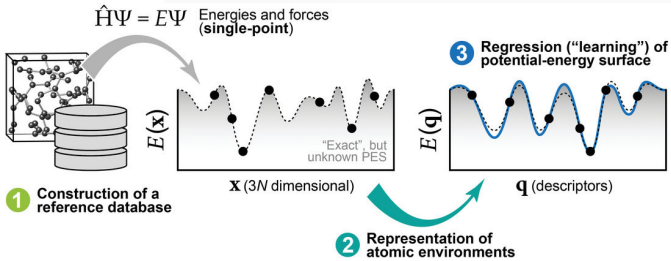
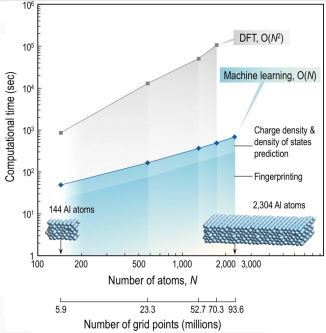
Basics



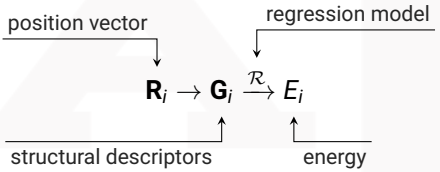


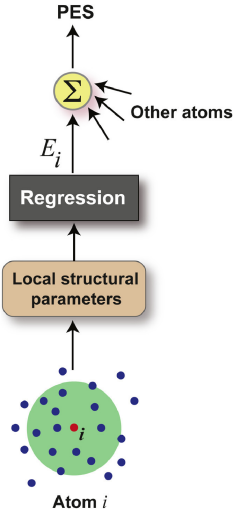
- ML interatomic potentials combine the accuracy of ab initio methods with the efficiency of traditional potentials.
- They are generally trained on large datasets to predict material properties.
- Unlike DFT, ML interatomic potentials scale linearly with system size, $\mathcal{O}(N)$.

V.L.Deringer,M.A.Caro,G.Csanyi, Adv. Mater., 31 (2019) 1902765

N. Fedik, R. Zubatyuk, M. Kulichenko, N. Lubbers, J. S. Smith, B. Nebgen, R. Messerly, Y. W. Li, A. I. Boldyrev, K. Barros, O. Isayev, and S. Tretiak, Nat. Rev. Chem, 6 (2022), 653–672.

- Most of the ML interatomic potentials follow a similar pattern in constructing the PES:



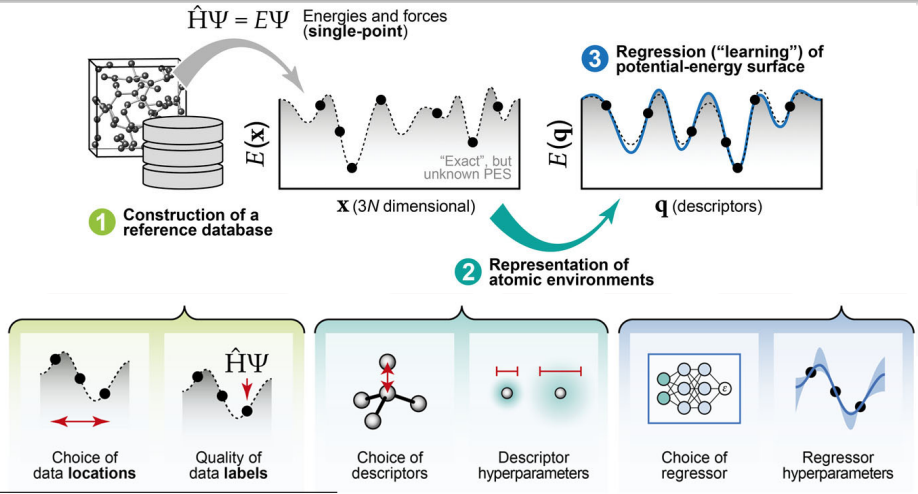


Y. Mishin, Acta Mater., 214 (2021), 116980.

A. Chandrasekaran, D. Kamal, R. Batra, C. Kim, L. Chen, R. Ramprasad, npj Comput. Mater., 5 (2019) 1–7.

Machine Learning Interatomic Potentials

Workflow of Machine Learning Potentials



J. D. Morrow, J. L. A. Gardner, and V. L. Deringer, J. Chem. Phys., 158 (2023), 121501.

V.L.Deringer,M.A.Caro,G.Csanyi, Adv. Mater., 31 (2019) 1902765

Universal Interatomic Potentials

Universal Interatomic Potentials

Definition

What are Universal Interatomic Potentials?

Universal Interatomic Potentials (UIP) are machine learning models designed to predict potential energy and atomic interactions across a wide range of materials and molecular systems.

- Large, open-source structural datasets form the backbone for training machine learning models.
- UIPs are generalizable across different chemical compositions and structural configurations outside the original training dataset.
- UIPs capture complex atomic interactions, making them suitable for diverse applications.

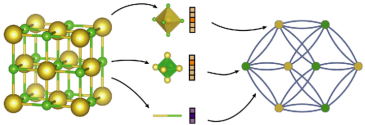
Name	Material Types	No. of Entries
AFLOW	Inorganic	3,530,330
OQMD	Inorganic	1,226,781
COD	General materials	517,261
Materials Project	Inorganic	154,718
NIST-JARVIS	Inorganic	80,000

Universal Interatomic Potentials

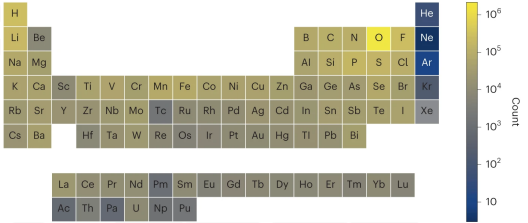
M3GNet

Key Features:

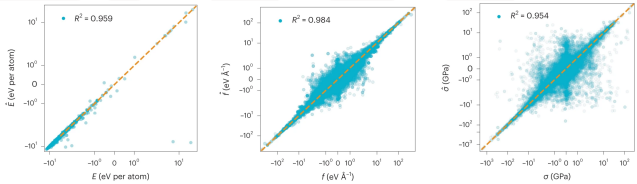
- Graph Neural Network (GNN) with many-body interactions.



- Predicts energies, forces, and stresses.
- Three-body interactions explicitly incorporated.
- Covers 89 elements across the periodic table.
- Trained on `MPF.2021.2.8` dataset.



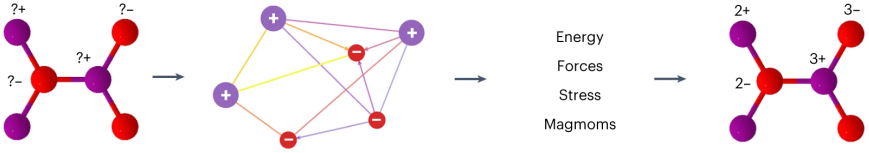
Element counts for all atoms in the `MPF.2021.2.8`

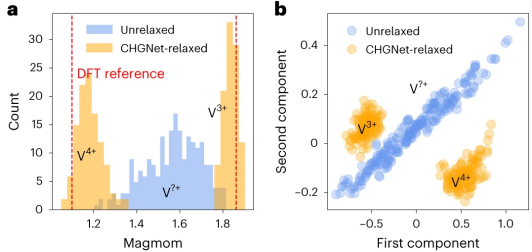


C. Chen, S. P. Ong, Nat. Comput. Sci., 2 (2022) 718–728.

Universal Interatomic Potentials

CHGNet





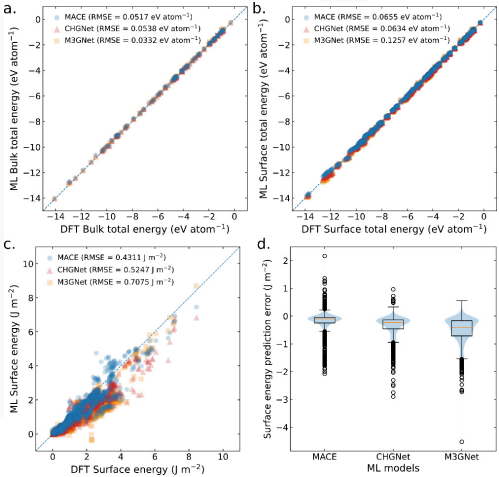
Key Features:

- Incorporates charge and magnetic moment constraints.
- Predicts energy, force, stress, and magnetic moments.
- Charge-informed molecular dynamics (MD).
- Trained on `MPtrj` dataset with 1.58M structures

B. Deng, P. Zhong, K. Jun, J. Riebesell, K. Han, C. J. Bartel, G. Ceder, (2023). arXiv:2302.14231.

Key Features:

- It is a higher-order equivariant message-passing neural network.
- Uses four-body messages to reduce the number of required message-passing iterations .
- Trained on `MPtrj` dataset with 1.58M structures.



I. Batatia, *et al.*, (2024). arXiv:2401.00096.

B. Focassio, L. P. M. Freitas, G. R. Schleder, ACS Appl. Mater. Interfaces (2024).

- It should be noted that the training datasets do not contain specific surface structures.

	RMSE over structures with Ni, Cu, Li, Mo, Si, and Ge			
ML Model	Bulk Total Energy (eV/atom)	Surface Total Energy (eV/atom)	Surface Energy (J/m^2)	Type
M3GNet	**0.0084**	0.0704	0.4246	UIP
CHGNet	0.0791	0.0386	0.5128	UIP
MACE	0.0856	0.0455	0.3512	UIP
$M3GNet_{FT}$	0.0399	0.0590	0.3852	UIP-FT
$MACE_{FT}$	0.0464	**0.0139**	0.2228	UIP-FT
$M3GNet_{S}$	0.0448	0.0859	0.7247	GNN
$MACE_{S}$	0.0372	0.0580	**0.2144**	GNN
MTP_{S}	0.0371	0.0537	0.2820	MTP

- Fine-tuning pre-trained models significantly improves the accuracy.

C. Chen, S. P. Ong, Nat. Comput. Sci. 2 (2022) 718–728.

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MTP_{S}	0.0371	0.0537	0.2820	MTP

- Fine-tuning pre-trained models significantly improves the accuracy.

C. Chen, S. P. Ong, Nat. Comput. Sci. 2 (2022) 718–728.

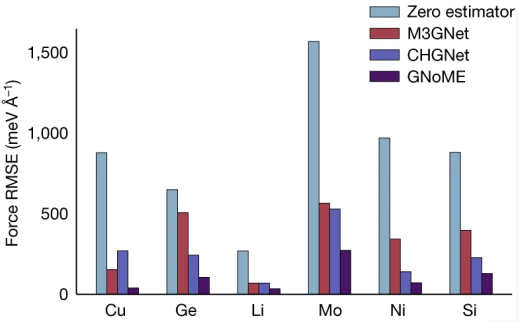
B. Deng, P. Zhong, K. Jun, J. Riebesell, K. Han, C. J. Bartel, G. Ceder, (2023). arXiv:2302.14231.

I. Batatia, *et al.*, (2024). arXiv:2401.00096.

Y.Zuo, C.Chen, X.Li, Z.Deng, Y.Chen, J.Behler, G.Csanyi, A.V. Shapeev, A. P. Thompson, M. A. Wood, S. P. Ong, J. Phys. Chem. A, 124 (2020) 731–745.

Universal Interatomic Potentials

GNoME



GNoME achieves a lower RMSE compared to CHGNet and M3GNet, even on unseen materials.

Key Features:

- Developed by Google DeepMind.
- It is based on the NequIP architecture.
- Operates within an active learning framework to generate candidate structures.
- Trained on custom `GNoME` dataset with 89M structures.
- Discovered 381 000 new stable crystals.

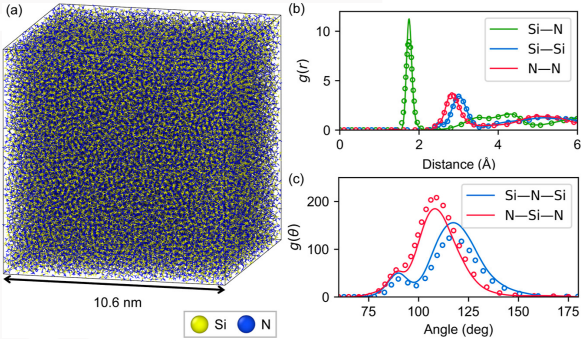
A. Merchant, S. Batzner, S. S. Schoenholz, M. Aykol, G. Cheon, E. D. Cubuk, Nature 624 (2023) 80–85.

Universal Interatomic Potentials

SevenNet

Key Features:

- It is based on the NequIP architecture.
- Efficient parallelization using spatial decomposition algorithms.
- Achieves over 80% parallel efficiency in weak-scaling tests and exhibits near-ideal strong-scaling performance.
- Trained on `MPtrj` dataset with 1.58M structures.

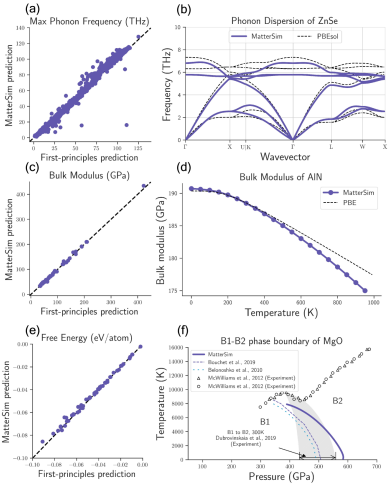


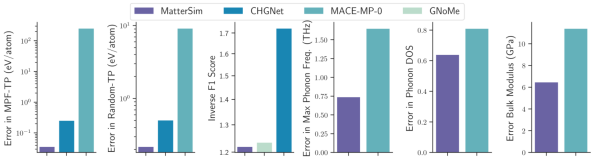
Suitable for generating amorphous structures.

Y. Park, J. Kim, S. Hwang, S. Han, J. Chem. Theory Comput. 20 (2024) 4857–4868.

Universal Interatomic Potentials

MatterSim





Key Features:

- Developed by Microsoft Research AI.
- Integrated with M3GNet and transformer-based architectures (Graphormer).
- Covers a vast materials space across the periodic table (0-5000 K and 0-1000 GPa).
- Trained on custom `MatterSim` dataset with 17M structures.

H. Yang, C. Hu, Y. Zhou, X. Liu, Y. Shi, J. Li, G. Li, Z. Chen, S. Chen, C. Zeni, M. Horton, R. Pinsler, A. Fowler, D. Zugner, T. Xie, J. Smith, L. Sun, Q. Wang, L. Kong, C. Liu, H. Hao, Z. Lu, (2024). arXiv:2405.04967.

Universal Interatomic Potentials

Comparison of Model Accuracy

Model	Dataset	Model Parameters	Structures	RMSE			
				Energies (eV/atom)	Forces (eV/Å)	Stresses (GPa)	Magnetic Moments (μ_B)
M3GNet	MPF.2021.2.8	227.5K	188k	0.035	0.072	0.41	-
CHGNet	MPtrj	412.5K	1.58M	0.030	0.077	0.348	0.032
MACE	MPtrj	4.69M	1.58M	0.013	0.040	0.020	-
SevenNet	MPtrj	842.4K	1.58M	0.024	0.067		-
GNoME	Custom	16.2M	89M	0.011	-	-	-
MatterSim	Custom	182.0M	17M	0.012	0.077	0.164	-

- As the number of training structures increases, the accuracy improves.
- As the number of model parameters increases, the accuracy improves.
- Equivariant model offers similar accuracy to the invariant model with fewer parameters.