Research Paper Presentation

EGraFFBench: evaluation of equivariant graph neural network force fields for atomistic simulations

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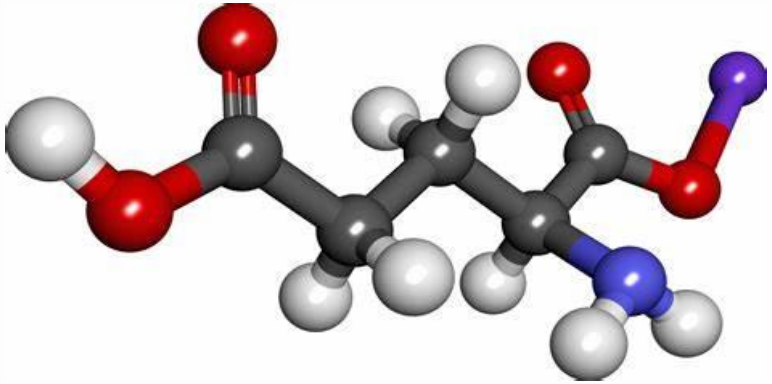
Outline

- Introduction
- Research Problem
- Methods
- Results and Discussions

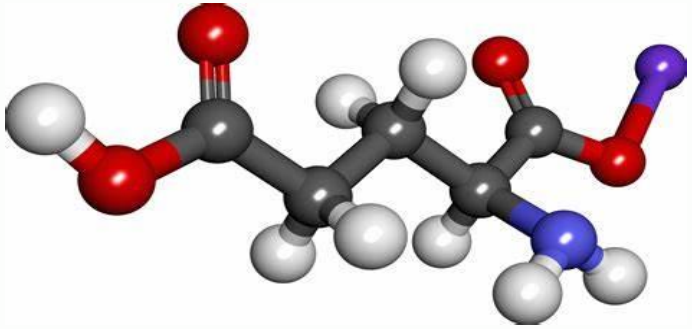
Conclusions

Research Problem



How do these atomic systems evolve with respect to time ?

Solution



We will run atomic simulations on computer

Using quantum data from *ab initio* Density Functional Theory (DFT) simulations

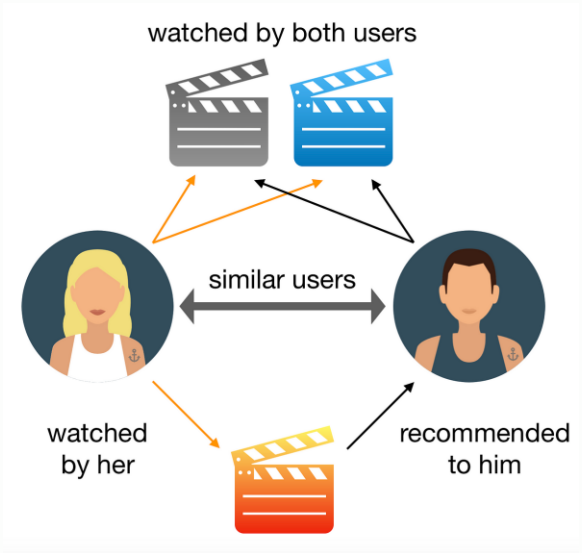
Expensive !!

$$

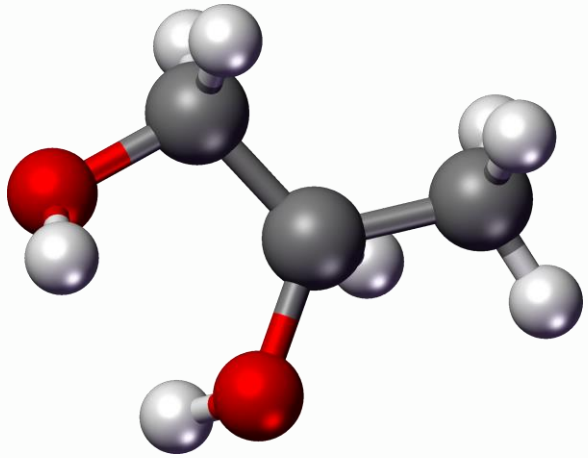
Background of Research

Graph Neural Networks have shown promising potential in areas where data is structured in the forms of graphs.

SOCIAL NETWORKS



RECOMMENDOR SYSTEMS



MOLECULAR CHEMISTRY

This leads to another problem :

LOADS OF DATA (to train deep GNNs)

- Researchers thought of a way to make these models computationally efficient by enforcing additional inductive bias

We used an approach similar to PINNs

EQUIVARIANCE : model outputs transform consistently with input transformations, such as rotations or translations in 3D space.

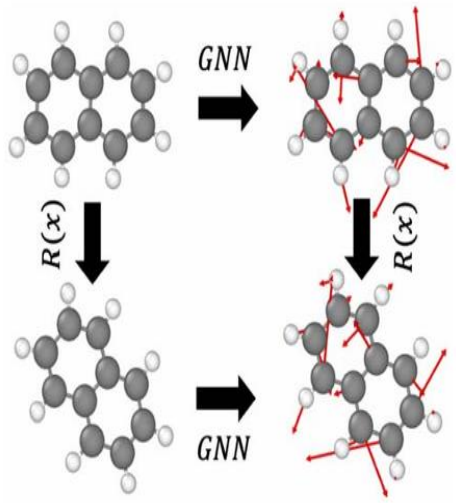
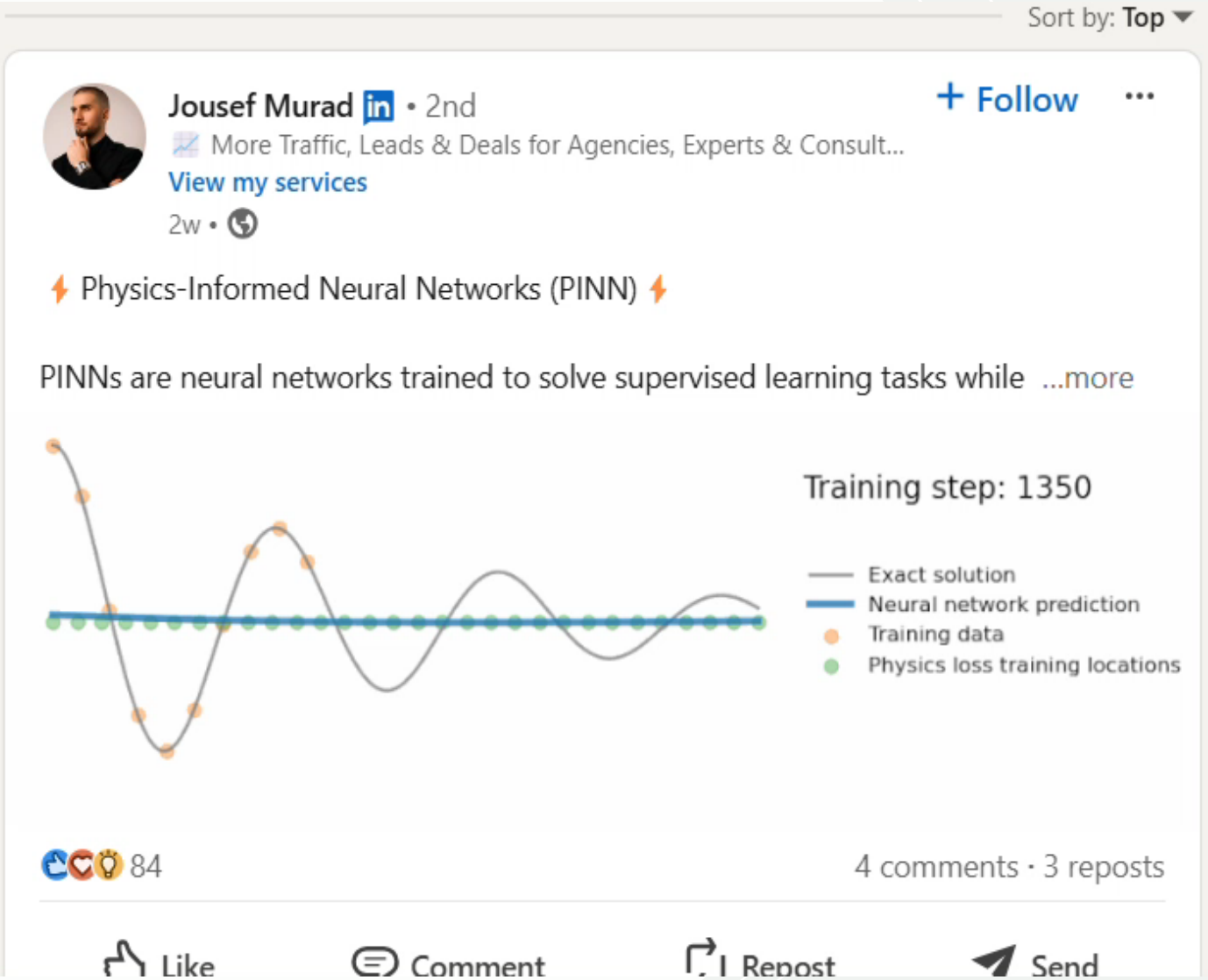


Fig. 1 Equivariant transformation *G* on a molecule under rotation *R*.



Introduction to EGraFF and Atomistic Simulations

Overview of Equivariant Graph Neural Networks for Atomic Simulations

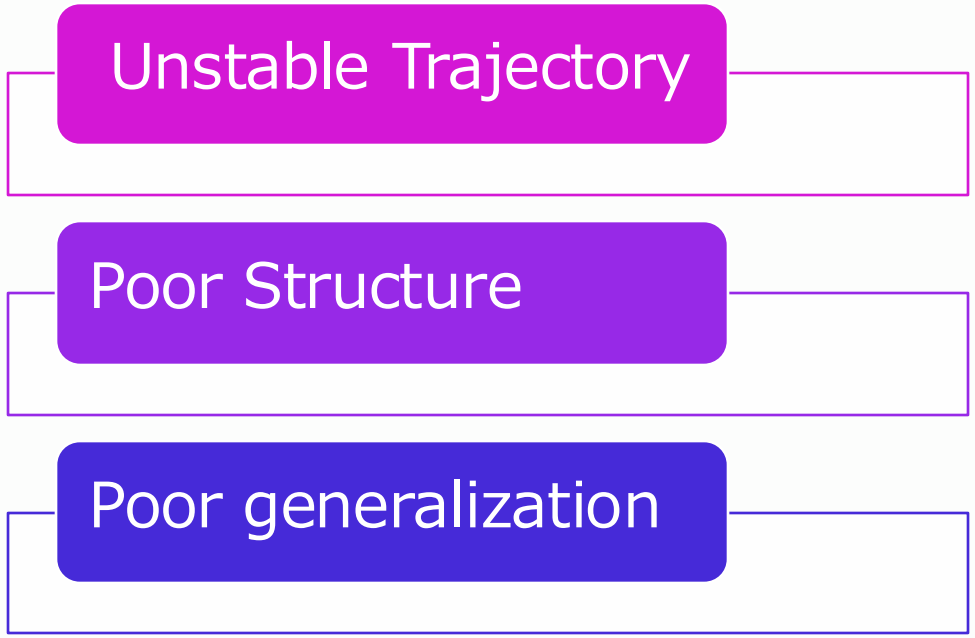
Equivariant Graph Neural Networks (EGraFFs)
EGraFFs leverage graph symmetries to model atomic interactions effectively, enabling detailed atomistic simulations.

Symmetry and Equivariance
These models apply symmetry principles, essential for accurate simulation, allowing predictions invariant to rotation and translation.

Applications in Atomistic Simulations
Used in modeling atomic behaviors in fields like drug discovery, materials science, and nanotechnology.

Need for Standardised evaluation in Atomistic Simulations

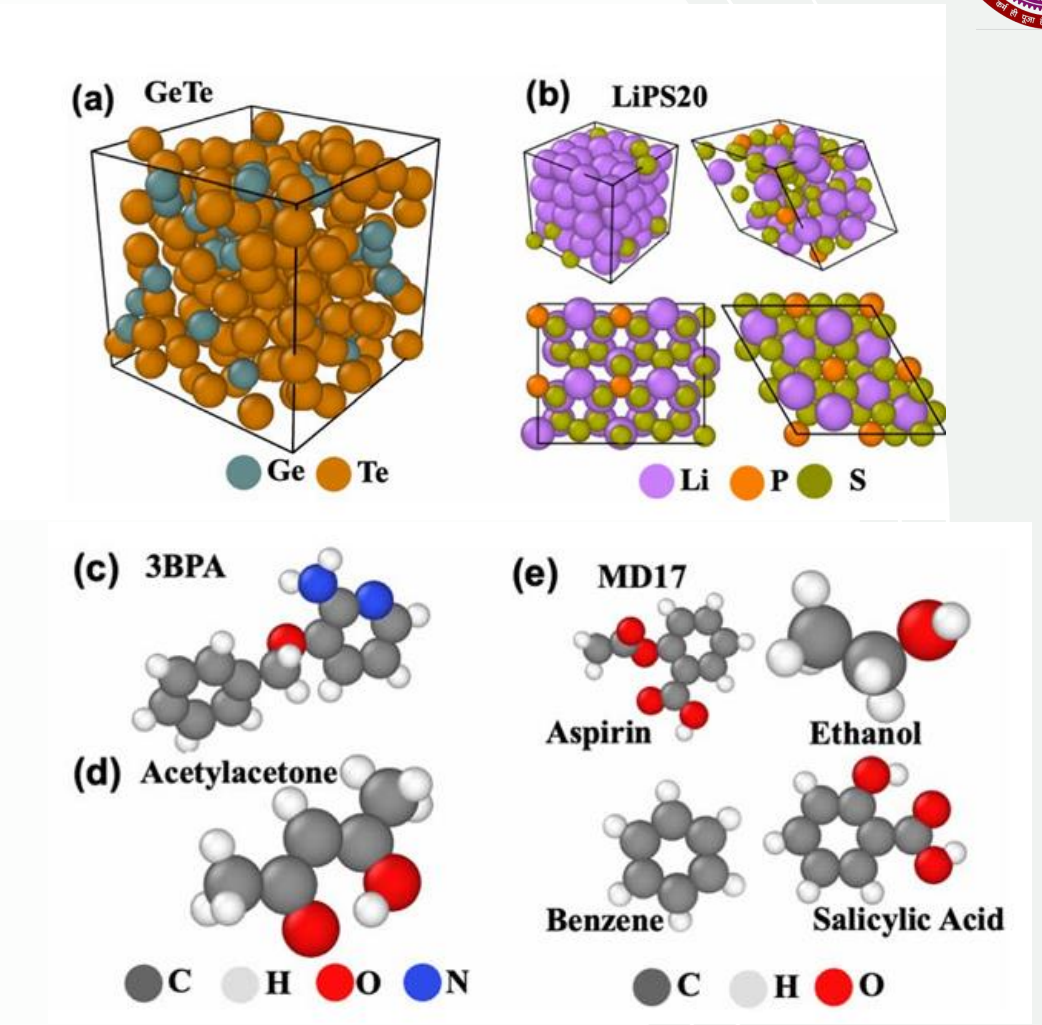
- Low test error does not guarantee the performance of EGraFFs on Atomistic simulations.
- EGraFFs can suffer from major issues like :



What this Paper provides ?

Evaluation of --->

1. • 6 EGraFFs
2. • 10 Datasets (2 new)
3. • New Metrics



MODELS EVALUATED

1) NequIP

Uses spherical harmonic and radial bias functions, ensuring equivariance

Relies on Message Passing

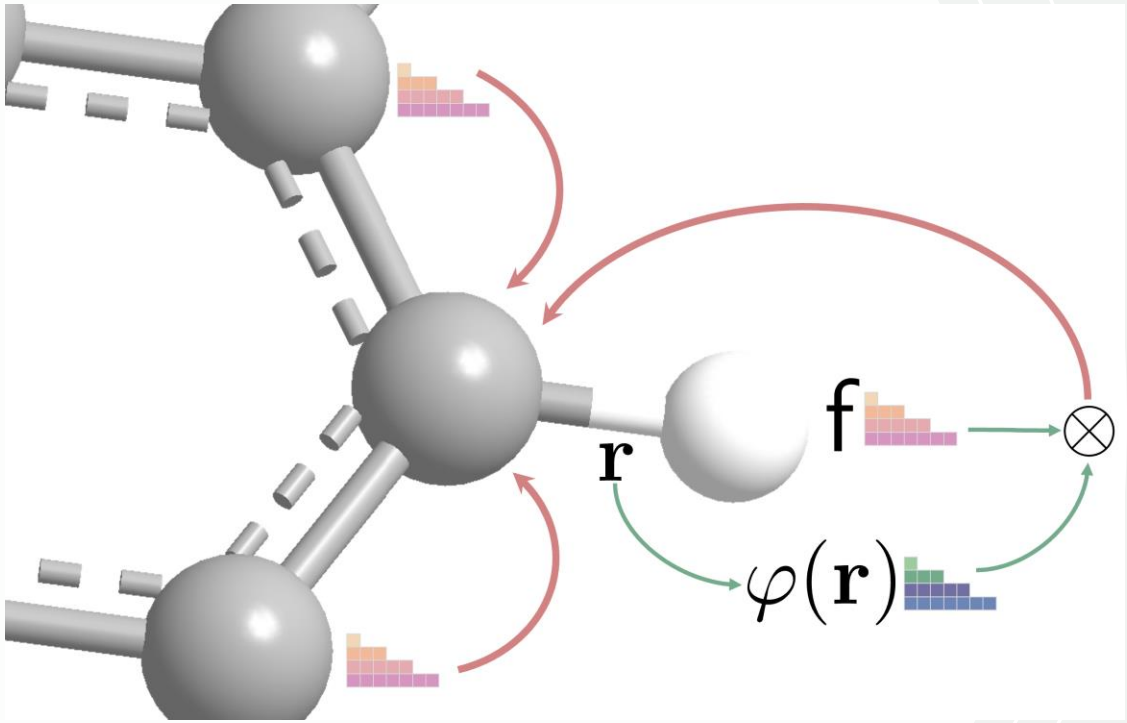
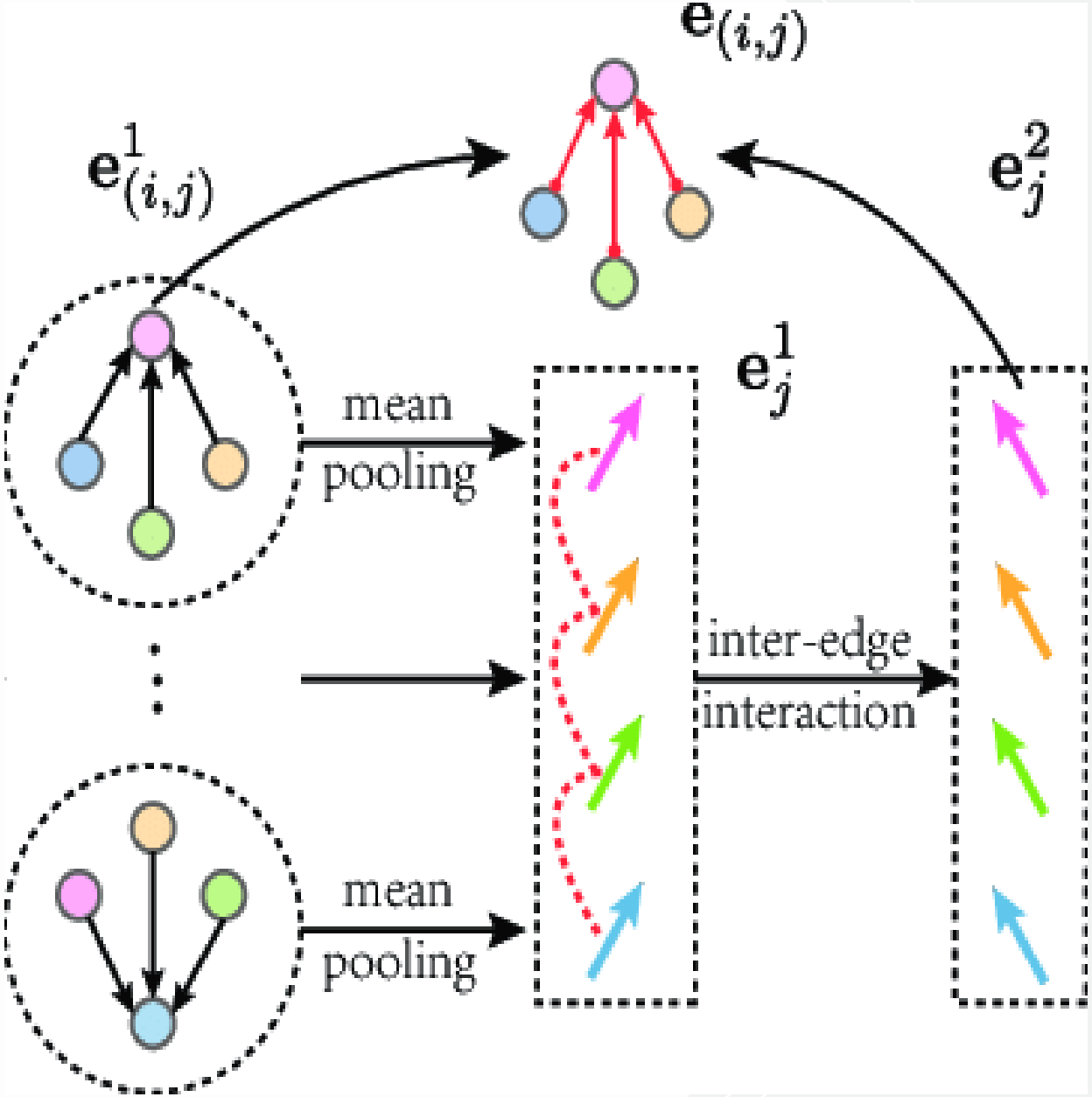


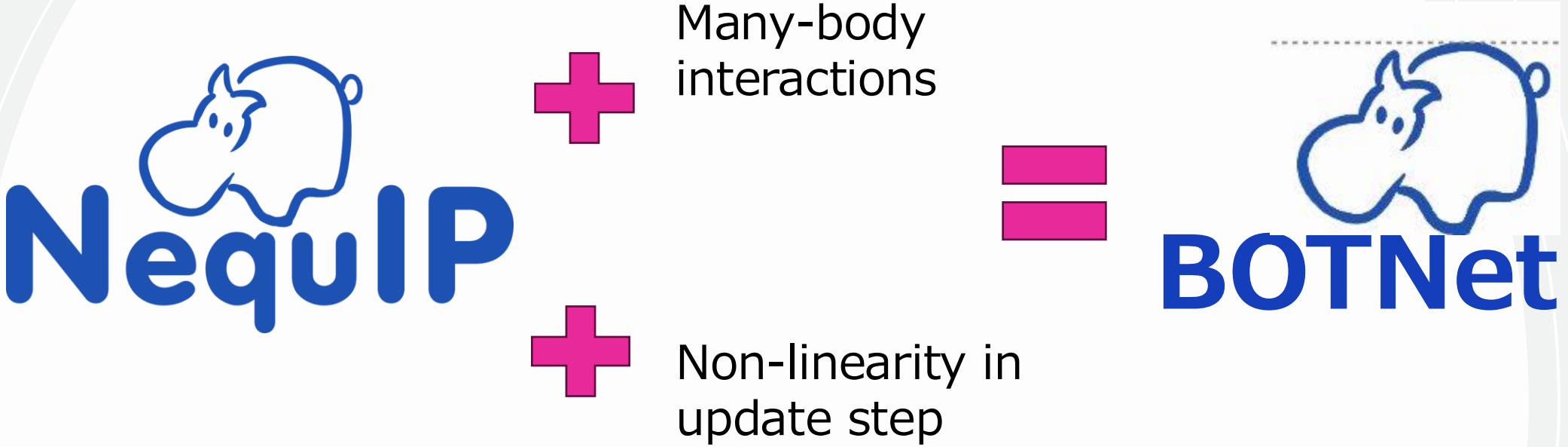
Fig.1 A pictorial demonstration of the message passing scheme in the Tensor Field Network (similar to what used in nequip)

2) Allegro

- NO Message passing , strict locality
- Prediction as a function of final Edge embeddings, unlike others
- Hence, Very scalable

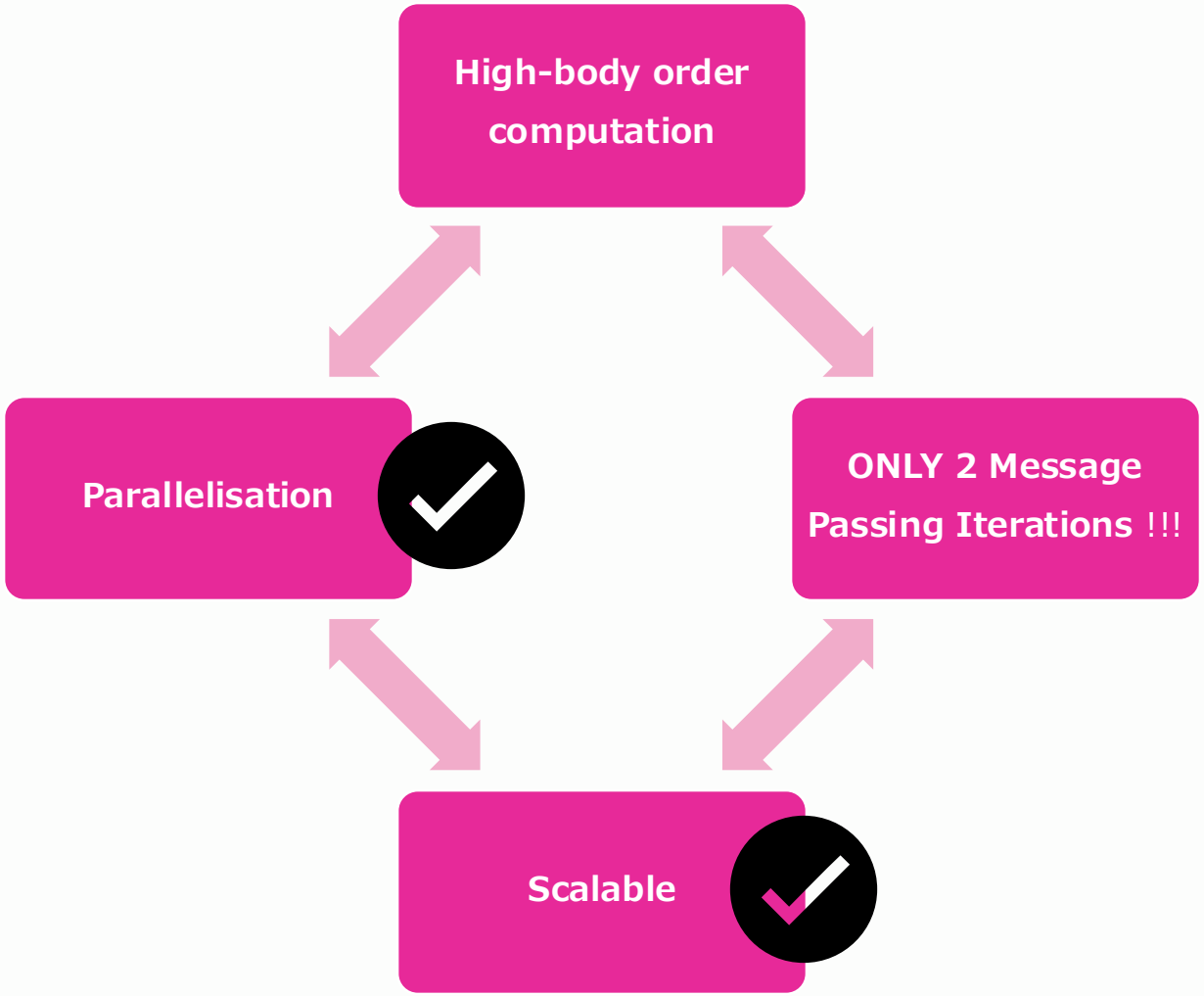


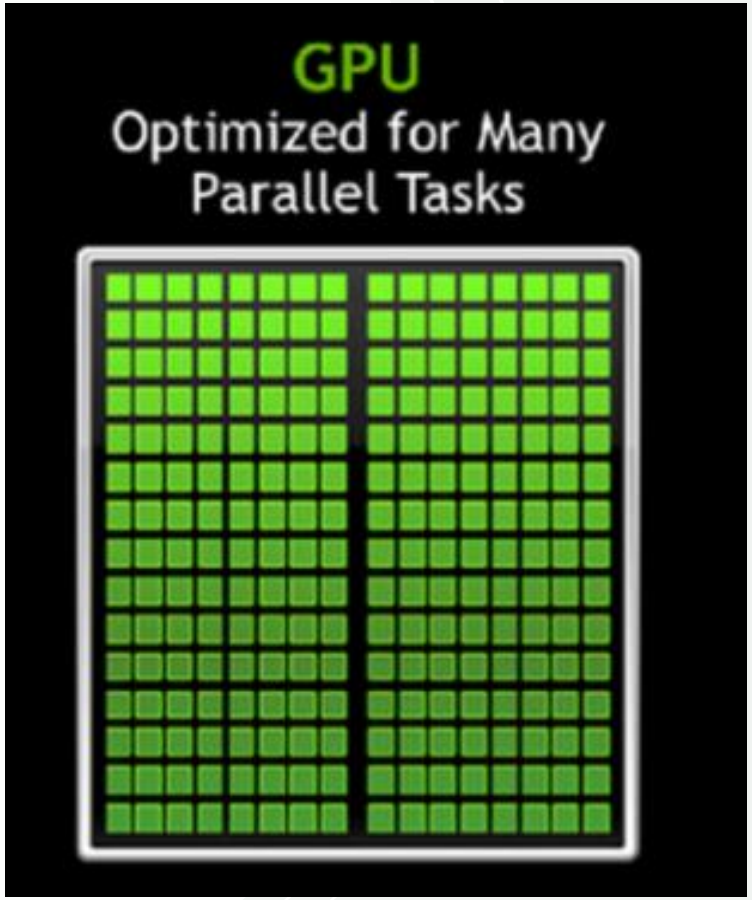
3) BOTNet



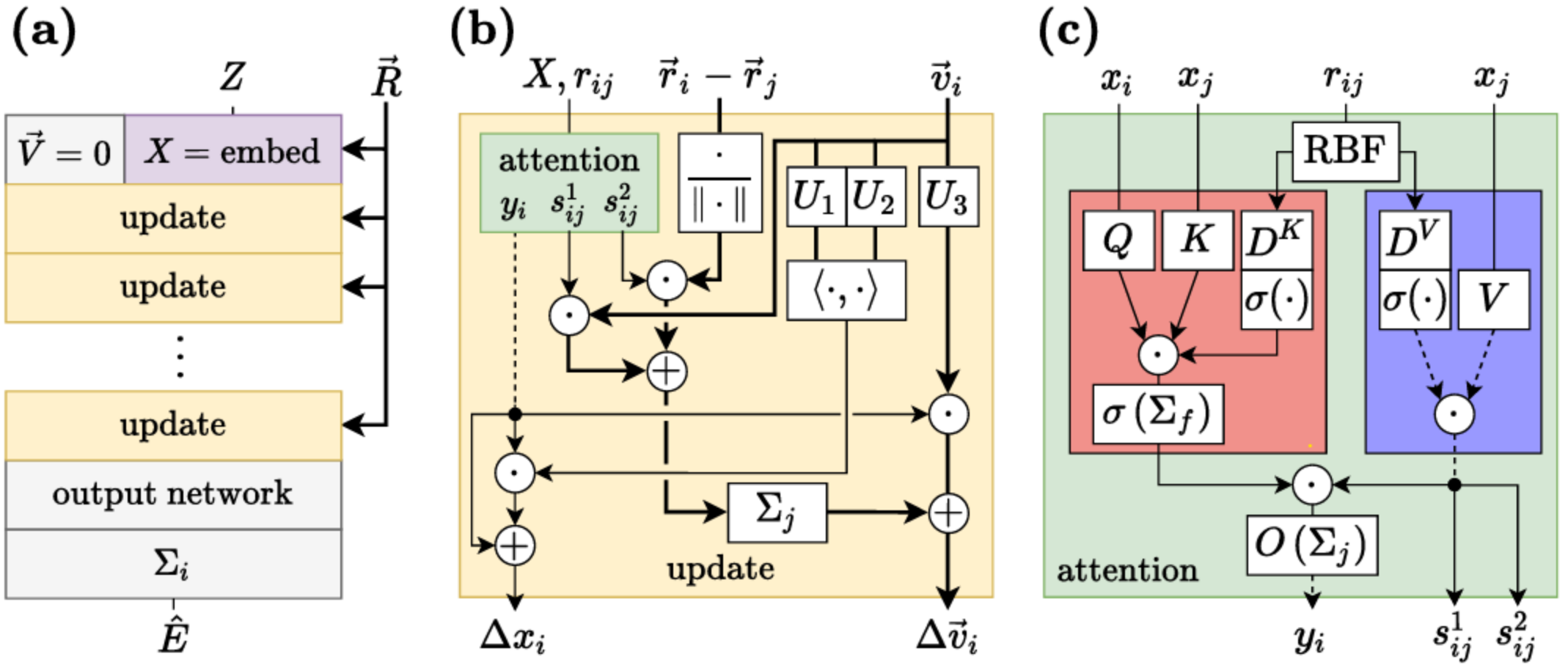
BOTNet15 is a refined body-ordered adaptation of NequIP.

4) MACE


High-body order computation
Parallelisation
ONLY 2 Message Passing Iterations !!!
Scalable


GPU
Optimized for Many Parallel Tasks

Equivariant Graph Transformers



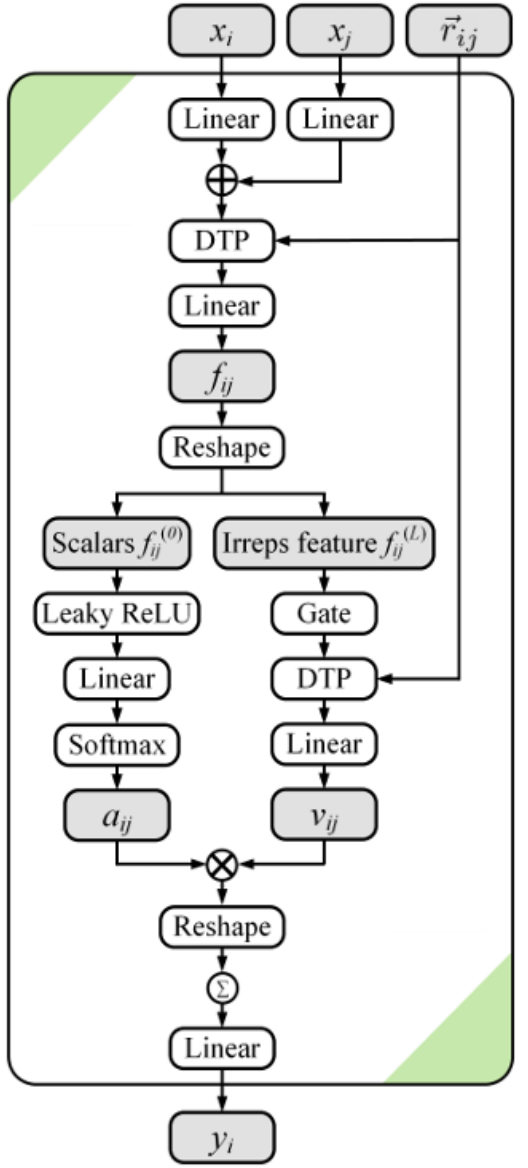
Transformer Models

TorchMDNet

- Modified multi-head attention mechanism
- Integrates edge data in the dot product

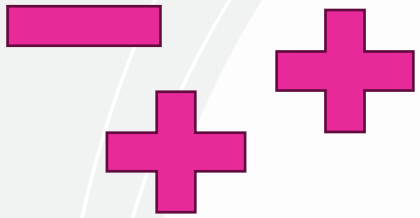
Equiformer

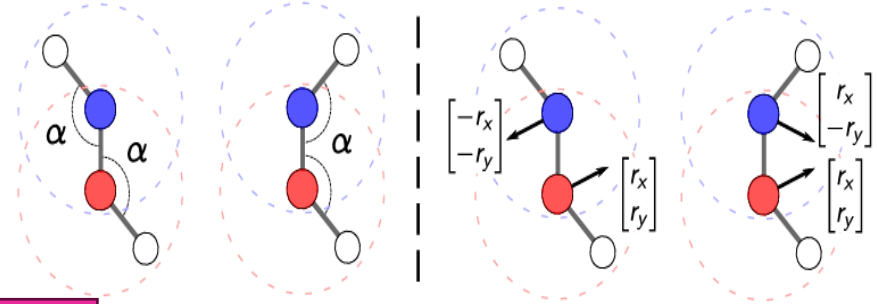
- Introduces a new attention mechanism – Graph attention
- Attention + Equivariance = Graph attention

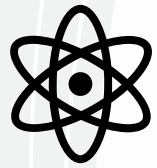


(b) Equivariant Graph Attention

PaiNN

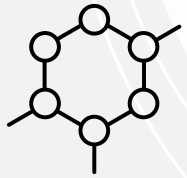




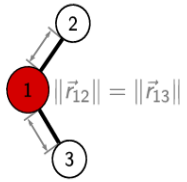
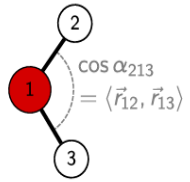
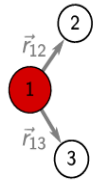


Considers the inter-atomic polarization

Message passing



Varying polarizability in different chemical environments.

Features	Distances	Angles	Directions
H_2O			
Message M at atom i	$\sum_{j\in\mathcal{N}_i} \|\vec{r}_{ij}\|$	$\sum_{j\in\mathcal{N}_i}\sum_{k\in\mathcal{N}_i} \alpha_{jik}$	$\sum_{j\in\mathcal{N}_i} \frac{\vec{r}_{ij}}{\|\vec{r}_{ij}\|}$
Scaling with neighbors	$\mathcal{O}(\|\mathcal{N}\|)$	$\mathcal{O}(\|\mathcal{N}\|^2)$	$\mathcal{O}(\|\mathcal{N}\|)$
Resolve change of $\|\vec{r}_{1j}\|$	yes	no	no
Resolve change of α_{213}	no	yes	yes

DimeNeT++

(best for angular information)

Captures directions during interaction as a message passing network

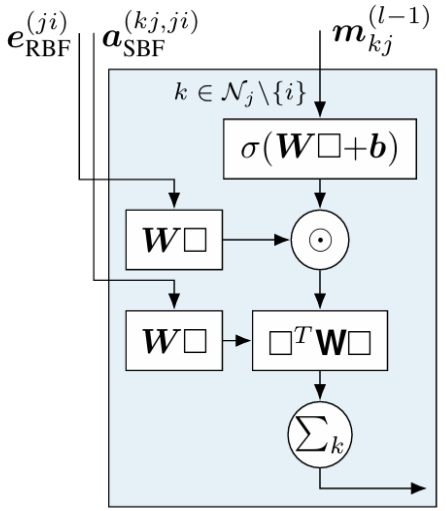
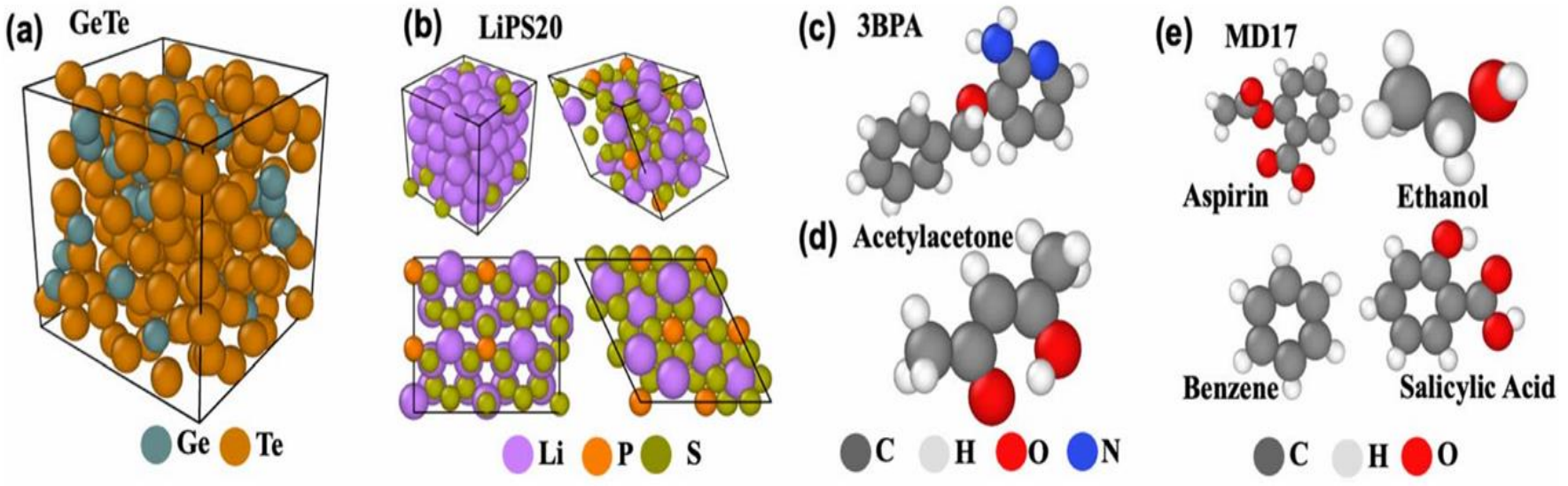


Figure 2: DimeNet's original "directional message passing" block.

DATASETS



Visualisation of datasets. (a) $GeTe_4$, (b) LiPS20, (c) 3BPA, (d) Acetylacetone, (e) MD17.

We need to have challenging datasets to benchmark the GNN models . Authors have contributed 2 new datasets (LiPS20 and GeTe) .

- **MD17**: A dataset containing small organic molecules, used widely for benchmarking machine learning (ML) force fields. **This includes molecules like aspirin, ethanol, naphthalene, and salicylic acid**. This dataset is essential for testing model generalization across different chemical structures.
- **3BPA**: This dataset features a large, flexible organic molecule, **3-(benzyloxy)pyridin-2-amin**e (3BPA), with snapshots from simulations at different temperatures. The complexity in the dihedral energy surface makes it challenging for ML force fields.
- **LiPS**: Consists of **lithium, phosphorus, and sulfur elements** and represents a crystalline material relevant for battery applications. This dataset is used for studying force and energy error benchmarks in stable systems.
- **Acetylacetone (AcAc)**: Generated through simulations at multiple temperatures, this dataset requires models to generalize well to dihedral angles and presents a test of a model's ability to predict potential energy surfaces accurately.
- **GeTe** (New Dataset): This dataset is based on Car-Parrinello molecular dynamics of germanium and tellurium atoms, focusing on phase-change materials like GeTe. The dataset includes data on crystalline-to-disordered phase transitions, which are challenging for traditional force fields due to structural changes.
- LiPS20 : Modification of the LiPS datasets focusssing on different structures and variations of the molecule mainly focussed in LiPS

EVALUATION METRICS

STATIC

- **MAE (Mean Absolute Error)**

$$\mathrm{MAE} = \frac{1}{N}\sum_{i=1}^{N}|y_i - \hat{y}_i|$$

DYNAMIC

- **Wright's Factor (WF)**

$$R_\chi = \left[\frac{\sum_{i=1}^{n}\left(g(r) - g_{\mathrm{ref}}(r)\right)^2}{\sum_{i=1}^{n}\left(g_{\mathrm{ref}}(r)\right)^2}\right]$$

- **Energy Violation Error (EV) and Force Violation Error (FV)**

$$\mathrm{FV}(t) = \frac{\|\hat{\mathcal{F}}(t) - \mathcal{F}(t)\|_2}{\left(\|\hat{\mathcal{F}}(t)\|_2 + \|\mathcal{F}(t)\|_2\right)}$$

$$\mathrm{EV}(t) = \frac{\left(\hat{E}(t) - E(t)\right)^2}{\hat{E}(t)^2 + E(t)^2}$$

- **Jensen-Shannon Divergence (JSD)**

$$\mathrm{JSD}\left(g(r)\|g_{\mathrm{ref}}(r)\right) = \frac{1}{2}\left(\mathrm{KL}(g(r)\|\hat{g}(r)) + \mathrm{KL}\left(g_{\mathrm{ref}}(r)\|\hat{g}(r)\right)\right)$$

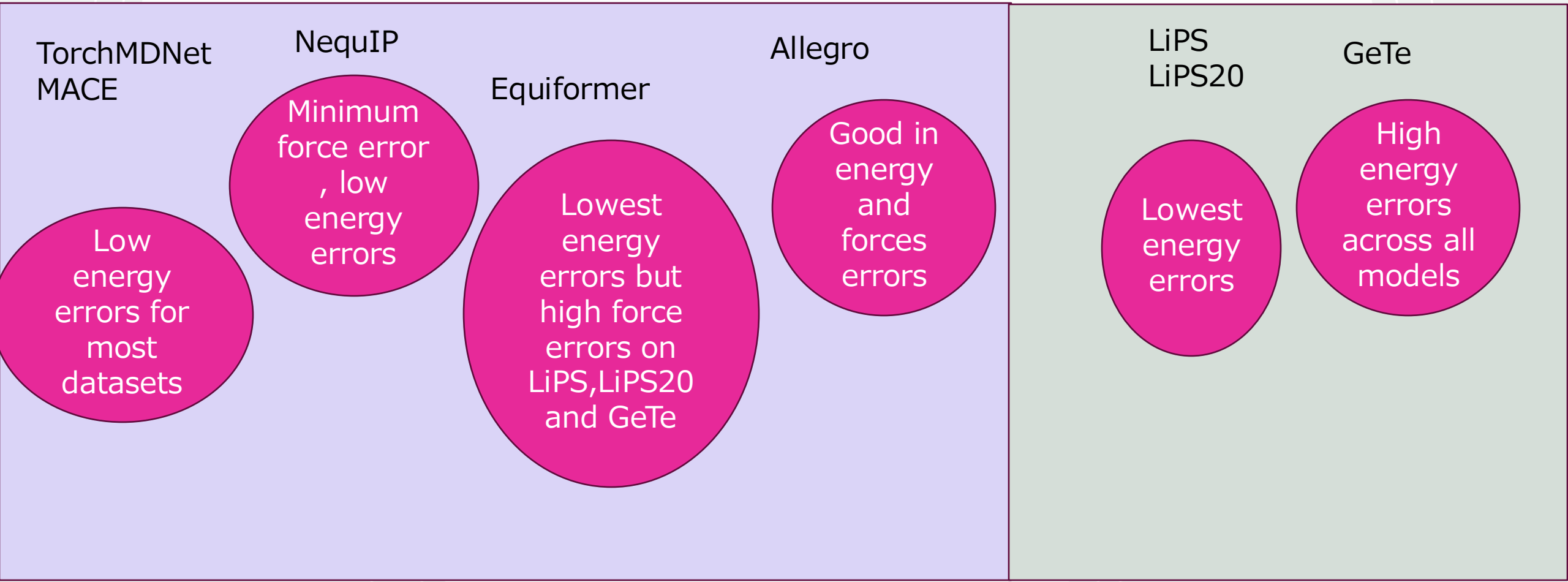
- **Geometric Mean of EV and FV**

$$\mathrm{GM}(EV, FV) = \sqrt{EV \times FV}$$

RESULTS

Energy and Forces

No single model outperforms others for all datasets (dataset specific nature of models



Forward Simulations

NequIP

Performs best in capturing the overall atomic structure , followed by TorchMDNet

- **TorchMDNet , despite having low energy MAE , does not guarantee low EV**
- **NequIP and Allegro show the least FV**
- **MACE and BOTNet perform better in terms of EV.**

Training and Inference Time

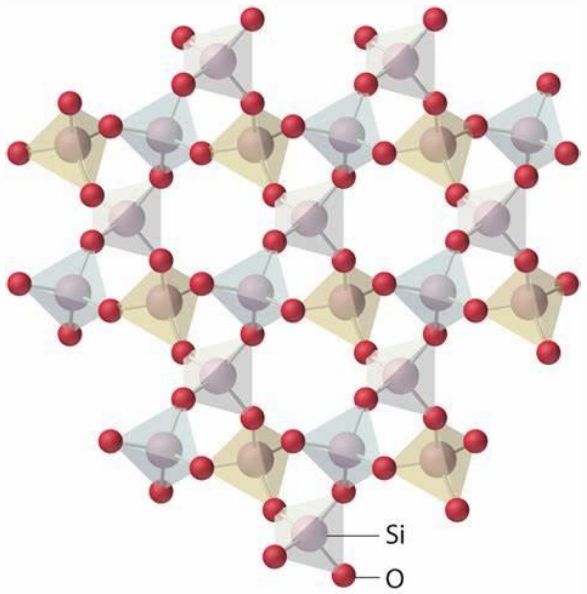
- **Transformer models require longer time**

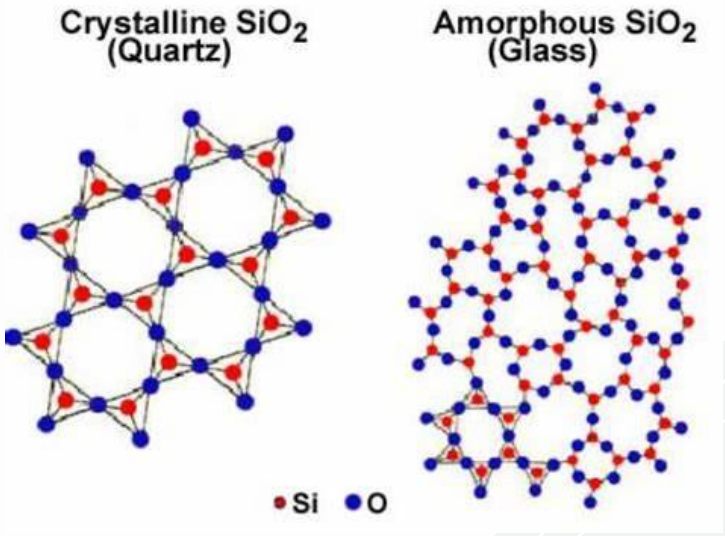
NequIP and Allegro take more time per epoch but converge faster

MACE and TorchMDNet have the lowest per-epoch training time.

Challenging Tasks on EGraFFs

- **Generalizability to higher temperatures**
- **Out of Distribution tasks on the LiPS Datasets**
  - ❖ Unseen Crystalline Structures
  - ❖ Generalizability to unseen compositions.





Conclusion

- **Dataset decides :** The performance of the model is highly dependent on the dataset
- **Structure** : Structural metrics should be evaluated
- **Stability during dynamics :** Energy and Force violations during MD simulations should be noted
- **Challenges :** Out of distribution challenges and High temperature generalization .

THANKS !!

Authors of the Paper:

EGraFFBench: evaluation of equivariant graph neural network force fields for atomistic simulations † Vaibhav Bihani, Santiago Miret, ‡ aSajid Mannan, ‡ aUtkarsh Pratiush, ‡ a Tao Du,b Zhimin Chen,b cMatthieu Micoulaut,d Morten M. Smedskjaer, and N. M. Anoop Krishnan *

For more details , refer to the original research paper here :
EGraFFBench: evaluation of equivariant graph neural network force fields for atomistic simulations - Digital Discovery (RSC Publishing)