FAENet: Frame Averaging Equivariant GNN for Materials Modeling

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Abstract

Applications of machine learning techniques for materials modeling typically involve functions known to be equivariant or invariant to specific symmetries. While graph neural networks (GNNs) have proven successful in such tasks, they enforce symmetries via the model architecture, which often reduces their expressivity, scalability and comprehensibility. In this paper, we introduce (1) a flexible framework relying on stochastic frame-averaging (SFA) to make any model E(3)-equivariant or invariant through data transformations. (2) FAENet: a simple, fast and expressive GNN, optimized for SFA, that processes geometric information without any symmetry-preserving design constraints. We prove the validity of our method theoretically and empirically demonstrate its superior accuracy and computational scalability in materials modeling on the OC20 dataset (S2EF, IS2RE) as well as common molecular modeling tasks (QM9, QM7-X). A package implementation is available at https://faenet.readthedocs.io.

1. Introduction

Machine Learning (ML) methods have the ability to model complex physical and chemical interactions. It thus holds great potential for accelerating material design, which is essential to various applications such as low-carbon energy, sustainable agriculture or drug discovery. One particularly promising use case of ML is modeling the properties of complex materials systems at lower computational cost compared to expensive quantum mechanical simulation techniques like Density Functional Theory (DFT). The heavy reliance on DFT for materials property prediction continues to impose a significant computational barrier to evaluating large number of material candidates (Chen & Ong, 2022). Graph Neural Networks (GNNs) based on geometric deep learning principles have shown promise in their ability to predict a wide range of molecular properties (Han et al., 2022). A key factor of the success of GNNs is their ability to leverage 3D geometric information via the representation of a collection of atoms in 3D space (Atz et al., 2021), which is updated based on spatial atomic interactions by passing messages between them. Another important aspect is the incorporation of geometric priors that exploit the symmetry of the data, rendering model predictions invariant or equivariant[1] to Euclidean transformations[2], as well as key physics principles such as the conservation of energy (Smidt, 2021).

Symmetries and physical constraints are typically enforced directly into the model architecture, which greatly restricts the flexibility of GNNs to process geometric information (Gasteiger et al., 2021; Fuchs et al., 2020; Satorras et al., 2021). As a result, these models either lack expressivity or present significantly more complex and computationally expensive architectures, as detailed in Section 2. While state-of-the-art GNNs remain orders of magnitude faster than DFT, their inference time still limits the use of ML for downstream practically-relevant applications, which require large-scale evaluations (Agrawal & Choudhary, 2016). Indeed, whether we are trying to discover new drugs, new catalysts or undiscovered material systems, we need to explore exponentially vast search spaces of potential candidates (Bohacek et al., 1996). The above ambitions to accelerate automated material discoveries therefore require designing expressive, robust and computationally scalable models.

To that end, we propose a novel view of 3D molecular and solid-state materials modeling, where symmetries are preserved via data projections instead of architectural constraints. Concretely, we make the following contributions:

- *Symmetry-Preserving Data Augmentation via Stochas-*

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[1]In this work, unless specified otherwise, we consider invariance to be a special case of equivariance and will include invariance in claims regarding equivariance.

[2]Rotations, reflections, and translations, which in 3D space define the group E(3).

tic Frame-Averaging: We propose a flexible framework to develop equivariant GNNs for materials modeling without any architectural requirement. Building upon Frame-Averaging (FA) (Puny et al., 2022), we propose to project data points into a canonical representation, allowing any model to be theoretically (*Full* FA) or empirically (*Stochastic* FA) E(3)-equivariant without losing expressiveness.

- *FAENet:* We introduce the Frame Averaging Equivariant Network (FAENet), a lightweight yet effective GNN whose design is not constrained by symmetry-preserving requirements. FAENet can therefore process geometric information through atom relative positions with full flexibility while rigorously preserving symmetries through the data, as leveraged by FA.

- *FAENet Analysis:* We verify the theoretical properties of the proposed approach, investigate its expressive power, and demonstrate its superior accuracy vs scalability trade-off compared to prior methods on four well known datasets in ML for materials science: OC20 IS2RE, S2EF (2M) for solid-state crystal structure modeling, QM7-X and QM9 for molecular modeling.

2. Related Work

Recent works have expanded the application of ML techniques to a broad set of materials modeling tasks ranging from solid-state (Zitnick et al., 2020; Miret et al., 2022) to molecular (Hoja et al., 2021; Ramakrishnan et al., 2014) structures. Most existing GNN architectures have applied physics-informed 3D symmetries directly in the model architecture, making model predictions explicitly invariant or equivariant to the desired transformations[3].

Various GNNs are constructed to be **E(3)-invariant** by extracting invariant geometric features from atomic positions (Unke & Meuwly, 2019; Klicpera et al., 2020b; Liu et al., 2021; Shuaibi et al., 2021; Ying et al., 2021; Adams et al., 2021). SchNet (Schütt et al., 2017), leverages atom distances via a continuous convolution filter to learn a potential energy surface, making it fast but unable to distinguish between certain types of molecules. ComENet (Wang et al., 2022) and GemNet (Gasteiger et al., 2021) extract additional information via bond angles and torsion angles(between quadruplets of nodes), thereby enabling the architecture to distinguish a larger set of atomic systems. Nevertheless, these methods are computationally expensive, as they use 3-hop neighbourhoods to compute torsion information for

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[3]A function f is invariant w.r.t. transformation t if, for any input x, $f(t(x)) = f(x)$. On the other hand, an equivariant function is such that $f(t(x)) = t(f(x))$. Typical examples include energy and forces, respectively

each update step at both training and inference time. Furthermore, they are usually less expressive than equivariant representations (Wang & Zhang, 2022; Joshi et al., 2022).

Equivariant methods (Thomas et al., 2018; Anderson et al., 2019; Fuchs et al., 2020; Brandstetter et al., 2021; Batatia et al., 2022; Frank et al., 2022) focus on enforcing equivariance by using irreducible representations of the SO(3) group. These works usually combine node features (learned based on atom relative positions) with a continuous equivariant filter (constructed based on spherical harmonics and learnable radial function) via a Clebsh-Gordan tensor product to guarantee E(3)- or SE(3)-equivariant predictions. While these methods are expressive and generalize well, they can be hard to implement and very computationally expensive for training and inference.

Additionally, Schütt et al. (2021); Batzner et al. (2022); Satorras et al. (2021); Thölke & De Fabritiis (2022) modeled equivariant interactions in Cartesian space using both scalar and vector representations. These GNNs achieve good performance and are relatively fast by avoiding expensive operations. However, the authors manually design two separate sets of functions to deal with each type of representation and often use complex operations to mix their information, which renders the global architecture hard to understand. Overall, such models lack formal guarantees.

Data augmentation (DA) is an alternative way of incorporating the desired data symmetries without constraining the model architecture. It has been found extremely effective in computer vision. In the graph domain, Hu et al. (2021) augmented molecular datasets with rotated and reflected input graphs. Despite showing promising results, the accuracy vs. scalability gains are not significant enough to constitute a true Pareto optimal improvement. Potential explanations include the suboptimal utilisation of geometric information given the absence of design constraints, which translates for instance into an overly complex model architecture, as well as soft and partial symmetry-preservation. In the 3D image domain explored by Gerken et al. (2022), equivariant DA also resulted in a mitigated performance-scalability trade-off. Gerken et al. (2022) showed as well that DA methods matched invariant networks in accuracy for invariant tasks on 3D images, with much smaller computational cost. Despite promising results, this area remains underexplored for 3D materials property prediction tasks.

3. Symmetry-Preserving Data Augmentation via Stochastic Frame Averaging

The first part of our modeling framework builds upon the idea of frame averaging, where we map the input data to a canonical plane using Principal Component Analysis (PCA). This mapping offers a unique representation of all Euclidean

transformations of the data and enables to rigorously preserve symmetries without constraining the GNN design. Frame-averaging also preserves the expressive power of the backbone architecture, ultimately leading to expressive architectures at lower inference cost.

3.1. Background

Frame averaging (FA) is a framework introduced by Puny et al. (2022) by which a function (such as a neural network) $\Phi : V \to W$ between normed vector spaces can be transformed into one that is equivariant (or invariant) with respect to a class of symmetries G. Suppose that ρ_1 and ρ_2 are representations of the group G over V and W, respectively. That is, for each element $g \in G$, $\rho_1(g)$ and $\rho_2(g)$ are the transformations that g induces in V and W.

A *frame* is defined as a function $\mathcal{F}(X) : V \to 2^G$, where the following properties are relevant:

- $\mathcal{F}$ is *G-equivariant* if $\forall X \in V, g \in G, \mathcal{F}(\rho_1(g)X) = g\mathcal{F}(X)$, where $g\mathcal{F}(X) = \{gh|h \in \mathcal{F}(X)\}$.

- $\mathcal{F}$ is *bounded* over a domain $K \subset V$ if $\exists c > 0$ such that $\forall g \in \mathcal{F}(X), X \in K, \|\rho_2(g)\|_{op} \leq c$, where $\|\cdot\|_{op}$ is the operator norm over W.

Puny et al. (2022) prove that if a frame is G-equivariant, then any arbitrary map Φ can be made equivariant (or invariant) by averaging predictions over that frame, noted $\langle\Phi\rangle_{\mathcal{F}}$:

$$\langle\Phi\rangle_{\mathcal{F}}(X) = \frac{1}{|\mathcal{F}(X)|} \sum_{g \in \mathcal{F}(X)} \rho_2(g)\Phi(\rho_1(g)^{-1}X)). \quad (1)$$

The authors also show that if $\mathcal{F}$ is bounded, FA-based GNNs $\Phi_{\mathcal{F}}$ are G-equivariant models which preserve the expressive power of the backbone architecture, leading to maximally expressive equivariant models for learning on graphs.

Note that these results extend those of the group averaging operator (Chen et al., 2021; Yarotsky, 2022), which takes the sum in Eq. (1) over all group elements. FA thus offers an efficient way to achieve the same theoretical properties.

In this work, we address the generic problem of materials property prediction where said properties are invariant/equivariant to Euclidean motions. As such, we can use FA to make our GNN Φ symmetry-preserving with respect to the group $G = \text{E}(3)$. Note that we formalise this task as a graph/node regression problem, where we predict properties (e.g., forces $\mathbf{y} \in \mathbb{R}^{n \times 3}$ or energy $y \in \mathbb{R}$) from initial atomic configurations (X, Z), where $X \in \mathbb{R}^{n \times 3}$ is the matrix of 3D atom positions and $Z \in \mathbb{N}^n$ their atomic numbers.

3.2. Frame Construction

The goal of frame construction is to find a frame uniquely determined by the configuration of atoms, such that the projected coordinates remain identical for arbitrary E(3) transformations. Additionally, the frame should be robust to slight distortions, such that similar objects are mapped to similar frames and representations. In order to define such a frame for our materials modeling case, we apply Principal Component Analysis (PCA) and set the principal components of the 3D atomic structure X as frame axes.

Concretely, we define $\mathcal{F}(X)$ using our PCA procedure as follows: First, we compute the centroid $\mathbf{t} = \frac{1}{n}X^\top\mathbf{1} \in \mathbb{R}^3$ and the covariance matrix $\Sigma = (X - \mathbf{1t}^\top)^\top(X - \mathbf{1t}^\top)$ of atom positions X. Then, we solve $\Sigma\mathbf{u} = \lambda\mathbf{u}$ to find the eigenvectors $\mathbf{u}$ of Σ. Under the assumption of distinct eigenvalues $\lambda_1 > \lambda_2 > \lambda_3$, we use the 3×3 orthogonal matrices $U = [\pm\mathbf{u}_1, \pm\mathbf{u}_2, \pm\mathbf{u}_3]$ to define the frame

$$\mathcal{F}(X) = \{(U, \mathbf{t})|U = [\pm\mathbf{u}_1, \pm\mathbf{u}_2, \pm\mathbf{u}_3]\} \subset \text{E}(3). \quad (2)$$

$|\mathcal{F}(X)| = 2^3 = 8$ for 3D atomic systems. We apply the symmetries $g = (U, \mathbf{t}) \in \mathcal{F}(X)$ to the data by defining the following group representations:

$$\rho_1(g)X = XU^\top + \mathbf{1t}^\top \quad (3)$$

$$\rho_2(g)X = \begin{cases} XU^\top & \text{for equivariant predictions} \\ X & \text{for invariant predictions} \end{cases}$$

To compute model inferences, we use the frame $\mathcal{F}(X)$ defined in Eq. (2) and group representations of Eq. (3) to average predictions over the frame, as in Eq. (1).

To extend this framework to periodic crystal structures, we also consider the case where the structure is not defined by just X but the extended set $D = (X, Z, C, O)$, where $C \in \mathbb{R}^{3\times3}$ refers to the unit cell Cartesian coordinates (with directional vectors as columns) and $O \in \{-1, 0, 1\}^{n \times n \times 3}$ to the cell offsets between any pair of atoms given periodic boundary conditions. With this new formulation, we keep the same frame $(\mathcal{F}(D) = \mathcal{F}(X))$ but we additionally need to apply symmetries on C, similarly to X, while O and Z remain unchanged by any E(3) transformation. For a more detailed justification, see Appendix A. This yields

$$\rho_1(g)^{-1}(D) = \left((X - \mathbf{1t}^\top)U, Z, (C - \mathbf{1t}^\top)U, O\right)$$
$$\rho_2(g)(D) = \left(XU^\top, Z, CU^\top, O\right), \quad (4)$$

for the equivariant case, while $\rho_2(g)$ remains the identity function for invariant predictions. We insert these functions into Eq. (1) to derive final model predictions. Note that ρ_1^{-1} is computed directly since it is the quantity of interest. Since U is an orthogonal matrix, we have $U^{-1} = U^\top$.

Proposition 3.1. *The frame $\mathcal{F}$ defined in Eq. (2) along with transformations ρ_1, ρ_2 defined in Eq. (4) is bounded and G-equivariant. Hence, any arbitrary map Φ becomes E(3)-invariant/equivariant using Eq. (1) while preserving its expressive power.*

Proof. See Appendix A. □

3.3. Stochastic Frame Averaging

The above frame averaging method, extended to crystals periodic structures and denoted *Full FA* in the rest of the paper, involves the average of GNN predictions $\langle\Phi\rangle(D)$ over $|\mathcal{F}(X)| = 8$ frames. Given our goal to enable scalable GNN training/inference, we propose an efficient approximation to *Full FA* called *Stochastic Frame Averaging (SFA)*.

In SFA, we randomly sample a single symmetry $g_* = (U_*, \mathbf{t})$ uniformly from $\mathcal{F}$ for each data point at every forward pass. Final model predictions thus become:

$$\Phi_{\mathcal{F}}^{invar}(D) = \Phi\left(((X - \mathbf{1}\mathbf{t}^\top)U_*, Z, (C - \mathbf{1}\mathbf{t}^\top)U_*, O)\right)$$
$$\Phi_{\mathcal{F}}^{equiv}(D) = \Phi_{\mathcal{F}}^{invar}(D) \cdot U_*^\top, \tag{5}$$

for the invariant $\Phi_{\mathcal{F}}^{invar}(D)$ and equivariant cases $\Phi_{\mathcal{F}}^{equiv}(D)$.

While sampling only a single frame component does not theoretically guarantee exact invariance or equivariance, we empirically demonstrate in Appendix D that our stochastic approximation is almost perfectly invariant and equivariant with up to $8\times$ faster compute time compared to *Full Frame Averaging*. Our approach can be viewed as a variant of data augmentation where we let the model learn symmetries by stochastically sampling among a few possible projections as opposed to all possible rotations and reflections of the data. This leads to a better preservation of data symmetries in addition to a higher performance with similar compute time when compared to conventional DA (see Appendix D). We hence bridge the gap between data augmentation approaches, which present unleveraged benefits (e.g. no model design constraints), and hard-engineered equivariant methods (with design constraints).

Additionally, this setup imbues our framework with valuable flexibility. SFA can (1) be incorporated into the training pipeline of any GNN; (2) enforce data symmetries approximately or exactly by using all 8 frames; (3) consider SE(3) (not equivariant to reflections) instead of E(3) by restricting $\mathcal{F}(X)$ to orthonormal positive orientation matrices, generally with $|\mathcal{F}(X)| = 4$ elements; (4) be adapted to any dataset. For instance, the z-axis is fixed in OC20 (Chanussot et al., 2021) (i.e. the catalyst is always below the adsorbate). Hence, we could consider only 2D rotation/reflection around the z-axis, which yields a total of 2 frames for SE(3) equivariance, where each has dimension 2×2.

4. FAENet

In this section, we describe our proposed GNN architecture, FAENet, detailed in Fig. 1, which is specifically designed to take advantage of the frame averaging framework introduced in Section 3.3. FAENet leverages 3D geometric information directly through atom relative positions without imposing any architectural constraints. FAENet inherits the theoretical guarantees of FA and satisfies relevant data symmetries. Overall, this yields greater modeling flexibility and the computational scalability needed to perform large-scale material property evaluations.

Graph Creation: Using $\mathcal{F}(D)$ from Eq. (2), we first apply the transformation ρ_1 defined in Eq. (4) to our input data D in order to map it onto a canonical representation $\mathfrak{C} = \{\rho_1(g)^{-1}(D) | g \in \mathcal{F}(D)\}$ with cardinality greater than one. This data projection is computed as a pre-processing step, and has a negligible impact on training time given that PCA is calculated on a 3×3 covariance matrix. During model training, following the SFA approach, we select one canonical element at random and create the graph (i.e. the adjacency matrix) using the cutoff distance c based on projected positions:

$$A_{ij} = \begin{cases} 1 \text{ if } d_{ij} < c \\ 0 \text{ otherwise} \end{cases} \tag{6}$$

where $d_{ij} = ||(\mathbf{x}_i - \mathbf{x}_j) + O_{ij*} \cdot C||$.

Embedding Block: Building upon the work of Duval et al. (2022), we leverage additional domain information to initialize atom representations, denoted $\mathbf{h}_i^{(0)} \in \mathbb{R}^h$ for atom i. We define $\mathbf{h}_i^{(0)}$ as a concatenation of several lookup embeddings based on atomic number, group, and period, as well as a fixed set of physical properties (see Appendix B.1) that were found to be relevant in previous works (Takigawa et al., 2016; Ward et al., 2017). We pass it to two-layer MLP.

To capture the 3D topology of the atomic system, we also learn an edge embedding $\mathbf{e}_{ij}$ between each pair of connected atoms (i, j). Since our model architecture is free of symmetry preserving constraints, we can process geometric information in a very simple and efficient manner using a 2-layer MLP on atom relative positions $\vec{r}_{ij}$. We concatenate radial basis functions (RBF) of distance information d_{ij} to $\vec{r}_{ij}$ as this quantity is very relevant and shown useful in previous works:

$$\mathbf{e}_{ij} = \sigma\left(\text{MLP}(\vec{r}_{ij} || \text{RBF}(d_{ij}))\right). \tag{7}$$

Note that FAENet takes as input both atom relative positions and atomic number, which suffices to uniquely identify any 3D material graph[4]. Moreover, by processing this information using universal approximators (Hornik et al., 1989), FAENet's embedding block has maximal expressive power.

Message Passing: In each interaction block, we propagate messages from neighbouring atoms $j \in \mathcal{N}_i$ to the center

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[4]This stands in contrast to various models (Schütt et al., 2017; Klicpera et al., 2020b; Liu et al., 2021) – which are provably unable to distinguish between certain molecules (e.g. enantiomers, CH4).
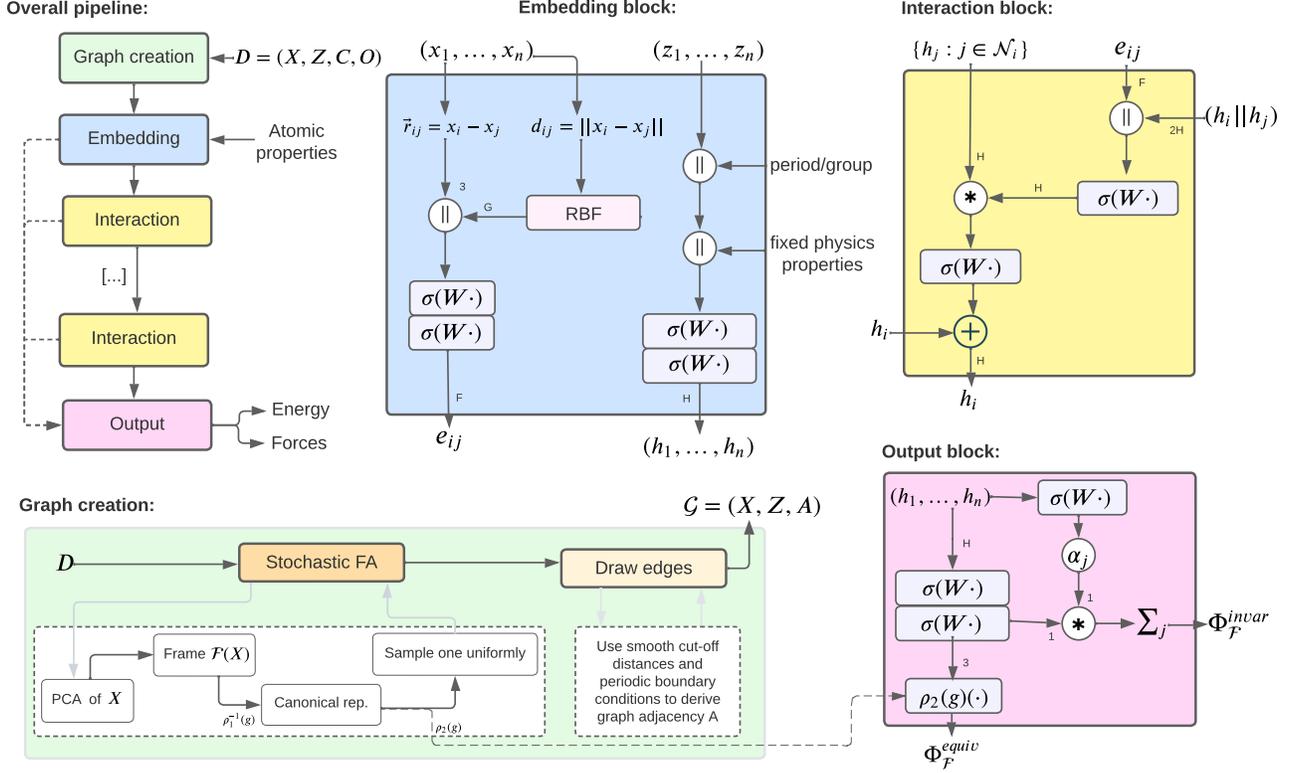
Figure 1. Overview of FAENet architecture making use of the Stochastic Frame Averaging framework. FAENet takes an input sample D and makes an invariant $\Phi_{\mathcal{F}}^{invar}$ (e.g. energy) and/or equivariant prediction $\Phi_{\mathcal{F}}^{equiv}$ (e.g. atomic forces) by passing sequentially through a graph creation step, an embedding block, several interaction layers and an output block. $*$ denotes the continuous convolution operation described in Eq. (8), $\|$ denotes concatenation, figures/letters the embedding dimension and $\sigma(W\cdot)$ a 1-layer MLP with the swish activation function. Note that while the graph creation step is performed only once, we still need to sample a different canonical representation from $\mathfrak{C}$ at each epoch. For Full FA, we apply FAENet on each element of $\mathfrak{C}$ and average after the output block (not shown).

atom i using a simple continuous convolution of $\mathbf{h}_j^{(l)}$ (the atom embeddings at layer l) with 3D geometric information $\mathbf{e}_{ij}$. Specifically, we learn a more precise graph convolution filter $\mathbf{f}_{ij}^{(l)} = \sigma(\mathrm{MLP}(\mathbf{e}_{ij}\|\mathbf{h}_i^{(l)}\|\mathbf{h}_j^{(l)}))$, with an activation function σ, to weigh the propagated message using both geometric information and atom endpoints:

$$\mathbf{h}_i^{(l+1)} = \mathbf{h}_i^{(l)} + \mathrm{MLP}\left(\sum_{j \in \mathcal{N}_i} \mathbf{h}_j^{(l)} \odot \mathbf{f}_{ij}^{(l)}\right). \quad (8)$$

In addition to being efficient, our message passing scheme is accessible and easy-to-understand compared to many approaches that enforce symmetries architecturally. Our GNN simply weighs each message received from neighbouring nodes using a convolution filter learned via an MLP of unique 3D geometric information (i.e. relative atom positions, edge endpoints' representations). This novel functioning grants FAENet with great discriminative capabilities, as demonstrated empirically in Appendix B.2, where we con-

duct an analysis of GNNs' expressive power as proposed by (Joshi et al., 2022). In particular, we show that FAENet is able to distinguish between the proposed complex molecular graphs examples with perfect accuracy, better than any other method. This highlights FAENet's high expressivity and showcases the advantage of leveraging geometric information without any design restrictions. We also corroborate that SFA preserves the expressive power of the backbone architecture and does not hamper FAENet's representation capability. Please refer to Appendix B.2 to see the expressivity analysis in full.

Architectural Details: We use GraphNorm (Cai et al., 2021), an efficient batch normalisation method, after each message passing layer to increase our network's robustness and mitigate vanishing/exploding gradient issues. For similar purposes we add skip-connections in the message passing layer, which also help manage the over-smoothing problem (Chen et al., 2020). On top of that, we add jumping connections between each interaction block and the output

block, using concatenation, to further boost discriminative power. Even though node representations get more refined and global as the number of layers increases, using such structural information helps the network generalize better (Xu et al., 2018). Finally, we chose the swish activation function because of its smooth and non-monotonic behaviour. A thorough ablation study of the model architecture is provided in Appendix B.3.

Output Block: We obtain the final atom predictions (y_i or $\vec{y}_i$) by passing the derived atom representations to two dense layers that map the embeddings to the correct dimension. For graph-level predictions, we perform a weighted average of atom-level predictions, where the weight is learned based on final atom embeddings: $\hat{y} = \sum_{i=1,...,n} \alpha(\mathbf{h}_i^{(L)}) \cdot y_i$, where $\alpha(\cdot)$ denote the learnable importance weights.

Note that this is the output $\Phi(D)^{invar}$. For equivariant predictions, like forces, we need to multiply the result by $U^\top$ as indicated in Eq. (5). Since we use an efficient approximation that samples uniformly at random one frame at each epoch among all possible ones, we directly obtain $\Phi_\mathcal{F}$.

For applications where we predict both energy and forces, unlike most previous works (Schütt et al., 2017), we do not compute atomic forces as the energy's gradient with respect to atom positions $F_i = \frac{\partial E}{\partial \mathbf{x}_i}$ (i.e. its definition in physics). The main reason for this is decreased scalability. Kolluru et al. (2022) showed that this process incurs a large computational overhead, increasing memory usage by a factor of 2-4 in addition to decreasing modeling performance in some datasets. Instead, we use two independent output heads: Φ^1 for graph-level energy predictions and Φ^2 for atom-level force predictions. The ability to model energy-conserving forces remains attractive in several applications such as molecular dynamics, where they are important for the stability of the simulation and improve the ability to reach new local minima (Chmiela et al., 2017). As a result, we decide to strengthen energy-conservation by fine-tuning a new loss term: the L_2 norm of atomic force predictions and energy gradient with respect to atom positions:

$$\mathcal{L}_{EC} = \sum_i ||\Phi_\mathcal{F}^2(D) - \nabla \Phi_\mathcal{F}^1(D)||_2. \quad (9)$$

5. Experiments

5.1. Empirical Evaluation of Model Properties

In this section, we empirically verify the correctness of our symmetry preserving framework, including its different theoretical properties[5]. To do so, we consider a set of symmetry preservation metrics (formally defined in Appendix D) and evaluate them on the IS2RE and S2EF taks of

OC20 (see Section 5.2.1; Zitnick et al., 2020) for the following FAENet variants (1) SFA, (2) Full FA, (3) conventional data augmentation (DA) using randomly sampled rotations and reflections of the graph, (4) No symmetry-preservation method (No-FA), (5) No training nor FA/DA (No-Train), (6) $G = SE(3)$ instead of $E(3)$. We also compare with the invariant model SchNet and the data augmentation model ForceNet. Based on the full results provided in Appendix D, we draw the following conclusions:

- *Full FA* as defined in Section 3.3 yields invariant/equivariant predictions, proving the correctness of our framework and implementation.

- *SFA* outperforms *DA* and *No-FA* both in terms of MAE (for comparable compute time) and of learned symmetries. In addition to providing a great approximation of invariance/equivariance, it yields slightly better MAE than *Full FA* while being much faster to run, proving the overall relevance of our approach.

- Not using any kind of symmetry-preserving techniques (No-FA) does not lead to invariant/equivariant predictions even if it clearly learns implicitly to preserve symmetries to some extent (vs No-Train). Besides, No-FA has a "significant" negative effect on performance compared to SFA, suggesting that enforcing symmetries in some way is desirable for OC20; although not critical (possibly due to its 2D nature given fixed z-axis).

Overall, these results support the correctness of our approach and the benefits of using Stochastic FA as a symmetry preserving data augmentation method.

5.2. Model Evaluation

In this section, we evaluate both the performance and scalability of our proposed approach, FAENet with stochastic frame averaging, on four common benchmark datasets: OC20 IS2RE, S2EF, QM9 and QM7-X. Full experimental settings are provided in the Appendix C.

Metrics: Our evaluation metrics focus on understanding the importance of the performance-scalability trade-off of different methods as motivated in Sec. 1. Unless specified otherwise, the main modeling performance metric for each task is the mean absolute error (MAE). Scalability is measured both by the training time for an epoch and the throughput at inference time[6], denoted *Train* and *Infer*.

Baseline Models: We compare our method with a wide variety of GNNs for 3D materials prediction tasks: $E(3)$-invariant models Schnet (Schütt et al., 2017), Dimenet++

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[6]We define throughput as the average number of samples per second that a model can process in its forward pass.

(Klicpera et al., 2020a), GemNet-T (Gasteiger et al., 2021), SphereNet (Liu et al., 2021), ComeNet (Wang et al., 2022), SpinConv (Shuaibi et al., 2021), GemNet-OC (Gasteiger et al., 2022) and Graphformer (Ying et al., 2021); data augmentation based ForceNet (Hu et al., 2021); equivariant models So3krates (Frank et al., 2022), PaiNN (Schütt et al., 2021) and EGNN (Satorras et al., 2021); as well as GNS (Godwin et al., 2021) and SpookyNet (Unke et al., 2021).

5.2.1. OC20

OC20 (Zitnick et al., 2020) is a large dataset for catalysis discovery. It was constructed to train ML models to approximate DFT for structure relaxation and energy prediction, which is fundamental to determine a catalyst's activity and selectivity. *OC20* contains 1,281,040 DFT relaxations of randomly selected catalysts and adsorbates from a set of plausible candidates, where the catalyst surface is defined by a unit cell periodic in all directions. We focus on 2 tasks:

- *Initial Structure to Relaxed Energy* (IS2RE), that is the direct prediction of the relaxed adsorption energy from the initial atomic structure, i.e. a graph regression task requiring E(3)-invariance. It comes with a pre-defined train/val/test split, 450,000 training samples and hidden test labels.

- *Structure to Energy and Forces* (S2EF), that is the prediction of both the overall energy and atom forces, from a set of 2M 3D material structures. According to the dataset creators, the 2M split closely approximates the much more expensive full S2EF dataset, making it suitable for model evaluation (Chanussot et al., 2021).

We evaluated all models on the four ~25K samples splits of the validation set: In Domain (ID), Out of Domain Adsorbates (OOD-ads), Out of Domain catalysts (OOD-cat), and Out of Domain Adsorbates and catalysts (OOD-both). We measure performance on each validation split by the energy MAE (IS2RE, S2EF), atomic forces MAE (S2EF) and the percentage of predicted Energies within a Threshold (EwT) of the ground truth energy (IS2RE). Running times are taken over the ID validation set on similar GPU types[7].

Results IS2RE: Figure 2 and the corresponding full table of results (Table 9) show that FAENet outperforms all existing baseline approaches in terms of Energy MAE and EwT, except for Graphormer (Ying et al., 2021). While we could not obtain comparable running times for Graphormer, we note that it uses an ensemble of 31 models and requires 372 GPU-days to train with A100 GPUs. Similarly, GNS, the runner up of FAENet in terms of MAE, is a 50-layer model which is extremely expensive to run. Lastly, FAENet

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[7]If anything, the A6000 used by ComENet is benchmarked to be faster than the RTX8000 we have used for these models.
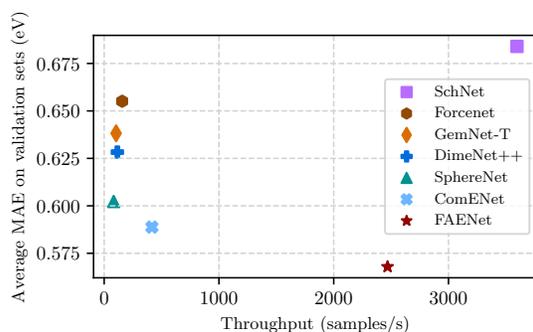

Figure 2. OC-20 Direct IS2RE performance / scalability trade-off. FAENet achieves the best MAE across methods while being much faster than the majority of baselines.

is 21 times faster than DimeNet++ and 6 times faster than ComeNet at inference time (*w.r.t.* throughput). On the other hand, while SchNet is still slightly faster than FAENet, we improve the average validation MAE by 17%. In a word, as illustrated in the Pareto plot in Figure 2, FAENet presents an extremely attractive performance-scalability trade-off.

Results S2EF 2M: Given the substantial cost of training on S2EF compared to IS2RE, results reported in Table 1 include fewer but still enough baselines to showcase the great performance-scalability feature of FAENet. Indeed, FAENet outperforms most baselines in terms of Energy MAE and Force MAE, while presenting the shortest runtime by a sizeable margin (e.g. even faster than SchNet on this dataset for optimal model architectures). Compared to ForceNet, the only other data augmentation approach, FAENet improves energy MAE by 38% while decreasing training time for an epoch by a factor of 20. This illustrates the benefits of our symmetry-preserving DA framework combined with our efficient message passing GNN design. Similarly to Graphormer in IS2RE, GemNet-OC achieves outstanding results on this dataset. Yet, GemNet-OC is significantly larger and more computationally expensive, processing 34 times less samples by GPU seconds than FAENet with respect to inference throughput (Gasteiger et al., 2022). GemNet-OC's functioning is also more complex given the constraints imposed on its functioning. Once again, FAENet offers an extremely desirable performance-scalability compromise.

5.2.2. QM7-X

QM7-X (Hoja et al., 2021) is a dataset containing 7K molecular graphs with up to seven non-hydrogen atoms (C, N, O, S, Cl), drawn from the GDB13 chemical universe (Blum & Reymond, 2009). After sampling and optimizing the structural and constitutional (stereo)isomers of each graph, the authors obtained 42K equilibrium structures. Each of them was then perturbed in order to obtain 99 additional

Table 1. Results on OC20 S2EF 2M, averaged accross all 4 validation splits. Performance is measured in terms of energy and forces MAEs. The best one is bolded, the second best is underlined. Scalability is measured with training time for one epoch (*Train*, in minutes) and inference throughput (*Infer.*, samples per second at inference time). Table 10 shows each val. split score.

Model	Scalability		Average MAE	
	Train ↓	Infer. ↑	Energy ↓	Forces ↑
SchNet	<u>98min</u>	<u>509</u>	1.4917	83.1
DimeNet++	1,157min	52	0.8096	66.3
ForceNet	1,476min	36	0.7548	61.0
GemNet-OC	–	18	**0.2860**	**25.7**
FAENet	**75min**	**623**	<u>0.4642</u>	<u>57.5</u>

non-equilibrium structures, leading to a total of about 4.2M samples. Of the 47 available targets, we focus on energy and forces as per previous work (Unke et al., 2021; Frank et al., 2022) but also because they are the main quantities of interest in real-world applications we care about.

Results: We report FAENet's results in Table 2 alongside the performance of SchNet, PaiNN, So3krates and SpookyNet. Since QM7-X is a very recent benchmark, only SpookyNet (which also computes SchNet and PaiNN performance) and So3krates have reported results on it for a comparable task. Unfortunately, the authors of the SpookyNet did not provide their code implementations, making it impractical for us confirm the results. We were able to reproduce the results for SchNet after authors of So3krates shared their codes with us, which included an unreported normalization method to train on shifted energy targets[8]. Nonetheless, we observe that FAENet achieves close-to-SOTA performance in spite of its simple architecture.

Table 2. QM7x results for FAENet and baseline methods based on best-effort replication given limited code availability. FAENet achieves competitive MAE. Best in bold, second best underlined.

Model	*Known molecules*		*Unknown molecules*	
	Energy	Forces	Energy	Forces
SchNet	42.43	56.45	51.05	65.84
PaiNN	15.69	20.30	17.59	24.16
So3krates	15.22	18.44	21.75	23.16
SpookyNet	**10.62**	**14.85**	**13.15**	**17.32**
FAENet	<u>11.42</u>	<u>17.54</u>	<u>15.17</u>	<u>21.23</u>

5.2.3. QM9

QM9 (Ramakrishnan et al., 2014) is a widely used dataset for molecular property prediction. It includes geometric, energetic, electronic, and thermodynamic properties for 134K stable small organic molecules with up to 9 heavy C, O, N,

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and F atoms. The dataset is split into 110K molecules for training, 10K for validation, and remaining 14K for testing.

QM9 Evaluation: We compare FAENet to a variety of methods that have publicly shared implementations and results. We trained independently on each target property a single FAENet model. Following (Liu et al., 2021), we report each model's MAE and training time for a single epoch. We also report its mean error and associated standard deviation across all molecular properties in QM9. To enable comparisons across properties units and ranges, we compute the average *relative improvement*[9] with respect to SchNet.

Results: The full table of results can be found in the Appendix 8. We plot relative improvements against the time we have measured models to take over a training epoch in Fig. 3. In addition to achieving competitive modeling performance, FAENet is the only model faster than SchNet.
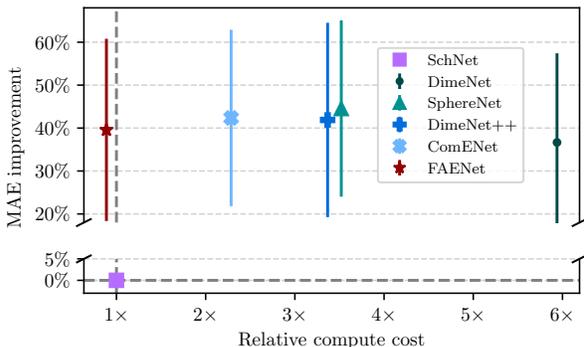

Figure 3. Performance/scalability trade-off on QM9. Large standard deviations and close means on the MAE average relative improvement against SchNet show that in recent years, models have saturated the complexity of QM9. There is however a wide range of (measured) computational costs. FAENet is the fastest.

6. Conclusion

In this paper, we offer an alternative to traditional symmetry-preserving GNN models for 3D materials property prediction. Instead of constraining the GNN functioning, we propose a flexible model-agnostic framework to enforce symmetries through a carefully defined data augmentation method built upon frame averaging. We then use this framework to design FAENet: a simple, fast and expressive GNN model, which directly and unrestrictedly processes atom relative information. While our empirical analysis demonstrates that Stochastic FA provides near-perfect symmetry preservation at up to 8× faster inference time, the rest of our experiments show that FAENet (with SFA) achieves

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[8]https://github.com/thorben-frank/mlff

[9]For a model's MAE m_p on property p, we report $r = \frac{m_p^0 - m_p}{m_p^0}$ where m_p^0 is the MAE of SchNet on p.

competitive modeling performance and superior scalability on various materials modeling tasks.

Future work. While our approach provides clear advantages, future work can improve upon current limitations by studying the conditions when exact equivariance can be imposed with less than the maximum number of frames. This could be particularly important in high-risk applications where guarantees are required to ensure safety. Additionally, both modeling performance and compute scalability require further improvement to effectively enable large-scale computational design of novel materials systems. Once achieved, the combination of fast and robust property prediction GNN with ML-based generative methods like diffusion models, reinforcement learning or GFlowNets, shall enable to meaningfully explore the vast search space in materials discovery.

References

Adams, K., Pattanaik, L., and Coley, C. W. Learning 3d representations of molecular chirality with invariance to bond rotations. *Preprint arXiv:2110.04383*, 2021.

Agrawal, A. and Choudhary, A. Perspective: Materials informatics and big data: Realization of the "fourth paradigm" of science in materials science. *Apl Materials*, 4(5): 053208, 2016.

Anderson, B., Hy, T. S., and Kondor, R. Cormorant: Covariant molecular neural networks. *Advances in neural information processing systems*, 32, 2019.

Atz, K., Grisoni, F., and Schneider, G. Geometric deep learning on molecular representations. *Nature Machine Intelligence*, 3(12):1023–1032, 2021.

Batatia, I., Kovács, D. P., Simm, G. N., Ortner, C., and Csányi, G. Mace: Higher order equivariant message passing neural networks for fast and accurate force fields. *Preprint arXiv:2206.07697*, 2022.

Batzner, S., Musaelian, A., Sun, L., Geiger, M., Mailoa, J. P., Kornbluth, M., Molinari, N., Smidt, T. E., and Kozinsky, B. E(3)-equivariant graph neural networks for data-efficient and accurate interatomic potentials. *Nature communications*, 13(1):1–11, 2022.

Blum, L. C. and Reymond, J.-L. 970 million druglike small molecules for virtual screening in the chemical universe database GDB-13. *Journal of the American Chemical Society*, 131(25):8732–8733, 2009.

Bohacek, R. S., McMartin, C., and Guida, W. C. The art and practice of structure-based drug design: A molecular modeling perspective. *Medicinal Research Reviews*, 16 (1):3–50, 1996.

Brandstetter, J., Hesselink, R., van der Pol, E., Bekkers, E., and Welling, M. Geometric and physical quantities improve E(3) equivariant message passing. *Preprint arXiv:2110.02905*, 2021.

Cai, T., Luo, S., Xu, K., He, D., Liu, T.-y., and Wang, L. Graphnorm: A principled approach to accelerating graph neural network training. In *International Conference on Machine Learning*, pp. 1204–1215. PMLR, 2021.

Chanussot, L., Das, A., Goyal, S., Lavril, T., Shuaibi, M., Riviere, M., Tran, K., Heras-Domingo, J., Ho, C., Hu, W., et al. Open catalyst 2020 (oc20) dataset and community challenges. *ACS Catalysis*, 11(10):6059–6072, 2021.

Chen, C. and Ong, S. P. A universal graph deep learning interatomic potential for the periodic table. *Preprint arXiv:2202.02450*, 2022.

Chen, D., Lin, Y., Li, W., Li, P., Zhou, J., and Sun, X. Measuring and relieving the over-smoothing problem for graph neural networks from the topological view. In *Proceedings of the AAAI Conference on Artificial Intelligence*, volume 34, pp. 3438–3445, 2020.

Chen, H., Liu, S., Chen, W., Li, H., and Hill, R. Equivariant point network for 3D point cloud analysis. In *Proceedings of the IEEE/CVF Conference on Computer Vision and Pattern Recognition*, pp. 14514–14523, 2021.

Chmiela, S., Tkatchenko, A., Sauceda, H. E., Poltavsky, I., Schütt, K. T., and Müller, K.-R. Machine learning of accurate energy-conserving molecular force fields. *Science advances*, 3(5):e1603015, 2017.

Duval, A., Schmidt, V., Miret, S., Bengio, Y., Hernández-García, A., and Rolnick, D. PhAST: Physics-aware, scalable, and task-specific GNNs for accelerated catalyst design. *Preprint arXiv:2211.12020*, 2022.

Fey, M. and Lenssen, J. E. Fast graph representation learning with PyTorch Geometric. In *ICLR Workshop on Representation Learning on Graphs and Manifolds*, 2019.

Frank, T., Unke, O. T., and Muller, K. R. So3krates: Equivariant attention for interactions on arbitrary length-scales in molecular systems. In *Advances in Neural Information Processing Systems*, 2022.

Fuchs, F., Worrall, D., Fischer, V., and Welling, M. SE(3)-transformers: 3D roto-translation equivariant attention networks. *Advances in Neural Information Processing Systems*, 33:1970–1981, 2020.

Gasteiger, J., Becker, F., and Günnemann, S. Gemnet: Universal directional graph neural networks for molecules. *Advances in Neural Information Processing Systems*, 34: 6790–6802, 2021.

Gasteiger, J., Shuaibi, M., Sriram, A., Günnemann, S., Ulissi, Z., Zitnick, C. L., and Das, A. Gemnet-oc: developing graph neural networks for large and diverse molecular simulation datasets. *arXiv preprint arXiv:2204.02782*, 2022.

Gerken, J., Carlsson, O., Linander, H., Ohlsson, F., Petersson, C., and Persson, D. Equivariance versus augmentation for spherical images. In *International Conference on Machine Learning*, pp. 7404–7421. PMLR, 2022.

Godwin, J., Schaarschmidt, M., Gaunt, A., Sanchez-Gonzalez, A., Rubanova, Y., Veličković, P., Kirkpatrick, J., and Battaglia, P. Simple gnn regularisation for 3d molecular property prediction & beyond. *arXiv preprint arXiv:2106.07971*, 2021.

Han, J., Rong, Y., Xu, T., and Huang, W. Geometrically equivariant graph neural networks: A survey. *Preprint arXiv:2202.07230*, 2022.

Harris, C. R., Millman, K. J., van der Walt, S. J., Gommers, R., Virtanen, P., Cournapeau, D., Wieser, E., Taylor, J., Berg, S., Smith, N. J., Kern, R., Picus, M., Hoyer, S., van Kerkwijk, M. H., Brett, M., Haldane, A., del Río, J. F., Wiebe, M., Peterson, P., Gérard-Marchant, P., Sheppard, K., Reddy, T., Weckesser, W., Abbasi, H., Gohlke, C., and Oliphant, T. E. Array programming with NumPy. *Nature*, 585(7825):357–362, September 2020. doi: 10.1038/s41586-020-2649-2.

Hoja, J., Medrano Sandonas, L., Ernst, B. G., Vazquez-Mayagoitia, A., DiStasio Jr, R. A., and Tkatchenko, A. Qm7-x, a comprehensive dataset of quantum-mechanical properties spanning the chemical space of small organic molecules. *Scientific data*, 8(1):1–11, 2021.

Hornik, K., Stinchcombe, M., and White, H. Multilayer feedforward networks are universal approximators. *Neural networks*, 2(5):359–366, 1989.

Hu, W., Shuaibi, M., Das, A., Goyal, S., Sriram, A., Leskovec, J., Parikh, D., and Zitnick, C. L. Forcenet: A graph neural network for large-scale quantum calculations. *Preprint arXiv:2103.01436*, 2021.

Joshi, C. K., Bodnar, C., Mathis, S. V., Cohen, T., and Liò, P. On the expressive power of geometric graph neural networks. In *The First Learning on Graphs Conference*, 2022.

Klicpera, J., Giri, S., Margraf, J. T., and Günnemann, S. Fast and uncertainty-aware directional message passing for non-equilibrium molecules. *Preprint arXiv:2011.14115*, 2020a.

Klicpera, J., Groß, J., and Günnemann, S. Directional message passing for molecular graphs. *Preprint arXiv:2003.03123*, 2020b.

Kolluru, A., Shuaibi, M., Palizhati, A., Shoghi, N., Das, A., Wood, B., Zitnick, C. L., Kitchin, J. R., and Ulissi, Z. W. Open challenges in developing generalizable large scale machine learning models for catalyst discovery. *Preprint arXiv:2206.02005*, 2022.

Liu, Y., Wang, L., Liu, M., Zhang, X., Oztekin, B., and Ji, S. Spherical message passing for 3D graph networks. *Preprint arXiv:2102.05013*, 2021.

Mentel, L. mendeleev – a python resource for properties of chemical elements, ions and isotopes. URL https://github.com/lmmentel/mendeleev.

Miret, S., Lee, K. L. K., Gonzales, C., Nassar, M., and Spellings, M. The open MatSci ML toolkit: A flexible framework for machine learning in materials science. *Preprint arXiv:2210.17484*, 2022.

Morris, C., Ritzert, M., Fey, M., Hamilton, W. L., Lenssen, J. E., Rattan, G., and Grohe, M. Weisfeiler and leman go neural: Higher-order graph neural networks. In *Proceedings of the AAAI conference on artificial intelligence*, volume 33, pp. 4602–4609, 2019.

Paszke, A., Gross, S., Massa, F., Lerer, A., Bradbury, J., Chanan, G., Killeen, T., Lin, Z., Gimelshein, N., Antiga, L., Desmaison, A., Kopf, A., Yang, E., DeVito, Z., Raison, M., Tejani, A., Chilamkurthy, S., Steiner, B., Fang, L., Bai, J., and Chintala, S. Pytorch: An imperative style, high-performance deep learning library. In Wallach, H., Larochelle, H., Beygelzimer, A., d'Alché-Buc, F., Fox, E., and Garnett, R. (eds.), *Advances in Neural Information Processing Systems*, volume 32. Curran Associates, Inc., 2019.

Pozdnyakov, S. N. and Ceriotti, M. Incompleteness of graph convolutional neural networks for points clouds in three dimensions. *arXiv preprint*, 2022.

Puny, O., Atzmon, M., Smith, E. J., Misra, I., Grover, A., Ben-Hamu, H., and Lipman, Y. Frame averaging for invariant and equivariant network design. In *International Conference on Learning Representations*, 2022.

Ramakrishnan, R., Dral, P. O., Rupp, M., and Von Lilienfeld, O. A. Quantum chemistry structures and properties of 134 kilo molecules. *Scientific data*, 1(1):1–7, 2014.

Satorras, V. G., Hoogeboom, E., and Welling, M. E(n) equivariant graph neural networks. In *International conference on machine learning*, pp. 9323–9332. PMLR, 2021.

Schütt, K., Kindermans, P.-J., Sauceda Felix, H. E., Chmiela, S., Tkatchenko, A., and Müller, K.-R. Schnet: A continuous-filter convolutional neural network for modeling quantum interactions. *Advances in neural information processing systems*, 30, 2017.

Schütt, K., Unke, O., and Gastegger, M. Equivariant message passing for the prediction of tensorial properties and molecular spectra. In *International Conference on Machine Learning*, pp. 9377–9388. PMLR, 2021.

Shuaibi, M., Kolluru, A., Das, A., Grover, A., Sriram, A., Ulissi, Z., and Zitnick, C. L. Rotation invariant graph neural networks using spin convolutions. *Preprint arXiv:2106.09575*, 2021.

Smidt, T. E. Euclidean symmetry and equivariance in machine learning. *Trends in Chemistry*, 3(2):82–85, 2021.

Takigawa, I., Shimizu, K.-i., Tsuda, K., and Takakusagi, S. Machine-learning prediction of the d-band center for metals and bimetals. *RSC advances*, 6(58):52587–52595, 2016.

Thölke, P. and De Fabritiis, G. Torchmd-net: Equivariant transformers for neural network based molecular potentials. *Preprint arXiv:2202.02541*, 2022.

Thomas, N., Smidt, T., Kearnes, S., Yang, L., Li, L., Kohlhoff, K., and Riley, P. Tensor field networks: Rotation-and translation-equivariant neural networks for 3d point clouds. *Preprint arXiv:1802.08219*, 2018.

Unke, O. T. and Meuwly, M. PhysNet: A neural network for predicting energies, forces, dipole moments, and partial charges. *Journal of chemical theory and computation*, 15 (6):3678–3693, 2019.

Unke, O. T., Chmiela, S., Gastegger, M., Schütt, K. T., Sauceda, H. E., and Müller, K.-R. Spookynet: Learning force fields with electronic degrees of freedom and nonlocal effects. *Nature communications*, 12(1):1–14, 2021.

Wang, L., Liu, Y., Lin, Y., Liu, H., and Ji, S. ComENet: Towards complete and efficient message passing for 3D molecular graphs. *Preprint arXiv:2206.08515*, 2022.

Wang, X. and Zhang, M. Graph neural network with local frame for molecular potential energy surface. *Preprint arXiv:2208.00716*, 2022.

Wang, Y., Ren, J., Yan, D.-M., Guo, J., Zhang, X., and Wonka, P. Mgcn: Descriptor learning using multiscale gcns. *ACM Transactions on Graphics (TOG)*, 39(4):122–1, 2020.

Ward, L., Liu, R., Krishna, A., Hegde, V. I., Agrawal, A., Choudhary, A., and Wolverton, C. Including crystal structure attributes in machine learning models of formation energies via voronoi tessellations. *Physical Review B*, 96 (2):024104, 2017.

Xu, K., Hu, W., Leskovec, J., and Jegelka, S. How powerful are graph neural networks? *Preprint arXiv:1810.00826*, 2018.

Yarotsky, D. Universal approximations of invariant maps by neural networks. *Constructive Approximation*, 55(1): 407–474, 2022.

Ying, C., Cai, T., Luo, S., Zheng, S., Ke, G., He, D., Shen, Y., and Liu, T.-Y. Do transformers really perform badly for graph representation? *Advances in Neural Information Processing Systems*, 34:28877–28888, 2021.

Zitnick, C. L., Chanussot, L., Das, A., Goyal, S., Heras-Domingo, J., Ho, C., Hu, W., Lavril, T., Palizhati, A., Riviere, M., et al. An introduction to electrocatalyst design using machine learning for renewable energy storage. *Preprint arXiv:2010.09435*, 2020.

A. Frame averaging

Proof. Let $D = (X, Z, C, O) \in V$ be an arbitrary atomic system with X, Z, C, O as given in Sec. 3.2. As our definition of the frame $\mathcal{F}$ in Eq. (2) does not depend on Z, C, O, we propose a simple adaptation of Proposition 1 from (Puny et al., 2022) to prove that $\mathcal{F}$ is G-equivariant and bounded for the space of materials V.

To show that $\mathcal{F}$ is translation equivariant, we need to show that $\mathcal{F}(\rho_1(g)D) = g\mathcal{F}(D)$ for all translations $g = (\mathbf{1}_D, \mathbf{s}) \in T(3)$. On one hand, the group product of these transformations is given by $g\mathcal{F}(D) = (\mathbf{1}, \mathbf{s})(U, \mathbf{t}) = (U, \mathbf{t} + \mathbf{s})$. On the other hand, we compute below the frame of a translated version of D, written $D' = \rho_1(g)(D)$. Let $\mathbf{t}', \Sigma', U'$ denote respectively its centroid, its covariance matrix and the related matrix of eigenvectors.

$$\rho_1(g)(D) = (X + \mathbf{1}\mathbf{s}^\top, Z, C + \mathbf{1}\mathbf{s}^\top, O)$$
$$\mathbf{t}' = \frac{1}{n}(X + \mathbf{1}\mathbf{s}^\top)^\top \mathbf{1} = \frac{1}{n}X^\top \mathbf{1} + \frac{1}{n}\mathbf{s}\mathbf{1}^\top \mathbf{1} = \mathbf{t} + \mathbf{s}$$
$$\Sigma' = \left((X + \mathbf{1}\mathbf{s}^\top) - \mathbf{1}\mathbf{t}'^\top\right)^\top (X + \mathbf{1}\mathbf{s}^\top) - \mathbf{1}\mathbf{t}'^\top))$$
$$= (X - \mathbf{1}t^\top)^\top(X - \mathbf{1}t^\top) = \Sigma$$

Hence, $\mathcal{F}(\rho_1(g)(D)) = \{(U, \mathbf{t} + \mathbf{s}) | U = [\pm\mathbf{u}_1, \pm\mathbf{u}_2, \pm\mathbf{u}_3]\} = g\mathcal{F}(D)$.

To show that $\mathcal{F}$ is rotation equvariant, we repeat the above with $g = (R, \mathbf{0}) \in SO(3)$. The group product of these transformations is $g\mathcal{F}(D) = (RU, R\mathbf{t})$. On the other hand, we compute the frame of a rotated version of D, denoted $D' = \rho_1(g)(D)$.

$$\rho_1(g)(D) = (XR^T, Z, CR^T, O)$$
$$\mathbf{t}' = \frac{1}{n}(XR^\top)^\top \mathbf{1} = \frac{1}{n}RX^T \mathbf{1} = R\mathbf{t}$$
$$\Sigma' = (XR^T - \mathbf{1}\mathbf{t}'^\top)^\top(XR^T - \mathbf{1}\mathbf{t}'^\top)$$
$$= (XR^T - \frac{1}{n}\mathbf{1}(\mathbf{1}^\top XR))^\top(XR^T - \frac{1}{n}\mathbf{1}(\mathbf{1}^\top XR))^\top$$
$$= R(X - \mathbf{t})^\top(X - \mathbf{t})^\top = R\Sigma R^\top$$

The eigendecomposition of Σ' yields $\mathcal{F}(\rho_1(g)(D)) = \{(RU, R\mathbf{t}) | [\pm\mathbf{u}_1, \pm\mathbf{u}_2, \pm\mathbf{u}_3]\} = g\mathcal{F}(D)$. As a result, $\mathcal{F}$ is E(3)-equivariant.

$\mathcal{F}$ is also bounded as for compact $K \in \mathbb{R}^{n \times d}$, the translations are compact and therefore uniformly bounded for $X \in K$. Orthogonal matrices always satisfy $||R||_2 = 1$.

Next, an adaptation of Theorem 4 (Puny et al., 2022) proves that any arbitrary continuous equivariant function $\psi : V \to W$ approximable by a neural network Φ over $K_\mathcal{F}$ is approximable by $\langle\Phi\rangle_\mathcal{F}$. We let $c > 0$ be a constant and consider an arbitrary $X \in K$. $\Psi : V \to W$ is an arbitrary G-equivariant function.

$$||\Psi(D) - \langle\Phi\rangle_\mathcal{F}(D)||_W \leq \frac{1}{|\mathcal{F}(D)|} \sum_{g \in \mathcal{F}(D)} ||\rho_2(g)\Psi(\rho_1^{-1}(D)) - \rho_2(g)\Phi(\rho_1(g)^{-1}(D))||_W$$
$$\leq \max_{g \in \mathcal{F}(D)} ||\rho_2(g)||_{op} ||\Psi(\rho_1^{-1}(D)) - \Phi(\rho_1^{-1}(D))||_W$$
$$\leq c||\Psi - \Phi||_W$$

This completes the proof.

$\square$

Justification for new definition of ρ_1, ρ_2. When E(3) transformations are applied to materials with periodic crystals structures, one also needs to rotate the unit cell on which the structure is defined, in addition to the atom positions. Indeed, in OC20, crystal materials are defined as semi-infinite repeating substructures, and one therefore needs to define a unit cell.

The graph's adjacency matrix is then constructed using periodic boundary conditions (pbc) as follows:

$$A_{ij} = \begin{cases} 1 \text{ if } d_{ij} < c \\ 0 \text{ otherwise} \end{cases}$$

$$\text{with } d_{ij} = ||(\mathbf{x}_i - \mathbf{x}_j) + O_{ij*} \cdot C|| \text{ and cutoff radius } c. \tag{10}$$

So for two different rotations of the same atomic system D, say D_1 and D_2, frame averaging projects D_1 and D_2 to the same canonical position $\hat{X}$. However, their respective unit cell will be different. Since GNN predictions depend both on unit cell and atom positions, this will yield different predictions. In different terms, not applying the transformations ρ_1, ρ_2 on the unit cell too would break rotation equivariance, which was verified empirically. We therefore extended their definition to guarantee E(3)-equivariance for the space of materials with periodic crystal structures.

Check that predictions are E(3)-equivariant. Let D_1 and D_2 be two transformed versions of the same atomic system D such that $\rho_1(g)D_2 = D_1$ for an arbitrary $g = (R, \mathbf{s}) \in E(3)$. $\mathcal{F}(D_1)$ is defined as in Section 3.2, with canonical representation

$$\mathfrak{C}_1 = \{\rho_1(g_1)^{-1}(D_1)|g_1 \in \mathcal{F}(D_1)\}$$
$$\rho_1(g_1)^{-1}(D_1) = ((X - \mathbf{1}\mathbf{t}^\top)U, Z, (C - \mathbf{1}\mathbf{t}^\top)U, O)$$

$\mathcal{F}(D_2)$ is defined using similar computations to the proof above. We obtain centroid $\mathbf{t}' = R\mathbf{t} + \mathbf{s}$, covariance matrix $\Sigma' = R\Sigma R^T$ and eigenvectors $U' = RU$. This yields $\mathcal{F}(D_2) = \{(U', \mathbf{t}')|U' = [\pm\mathbf{u}'_1, \pm\mathbf{u}'_2, \pm\mathbf{u}'_3]\}$ and corresponding canonical representation

$$\mathfrak{C}_2 = \{\rho_1(g_2)^{-1}(D_2)|g_2 \in \mathcal{F}(D_2)\}$$
$$\text{with } \rho_1(g_2)^{-1}(D_2) = ((X' - \mathbf{1}\mathbf{t}'^\top)U', Z, (C - \mathbf{1}\mathbf{t}'^\top)U', O)$$
$$= ((XR^\top + \mathbf{1}\mathbf{s} - \mathbf{1}\mathbf{t}^\top R^\top - \mathbf{1}\mathbf{s})RU, Z, (C - \mathbf{1}\mathbf{t}^\top)RU, O)$$
$$= ((X - \mathbf{1}\mathbf{t}^\top)U, Z, (C - \mathbf{1}\mathbf{t}^\top)U, O)$$
$$= \rho_1(g_1)^{-1}(D_1)$$

Hence, $\mathfrak{C}_1 = \mathfrak{C}_2$. Using Equation (1), GNN predictions are E(3)-equivariant. In the invariant case, $\langle \Phi \rangle_{\mathcal{F}}(D_1) = \langle \Phi \rangle_{\mathcal{F}}(D_2)$.

B. FAENet

B.1. Atom properties

In the embedding block, to construct atom-level embeddings, we use the following properties from the `mendeleev` Python package (Mentel):

1. atomic radius

2. atomic volume

3. atomic density

4. dipole polarizability

5. electron affinity

6. electronegativity (allen)

7. Van-Der-Walls radius

8. metallic radius

9. covalent radius

10. ionization energy (first and second order).

B.2. Expressivity discussion

In this section, we discuss the expressive power of FAENet. Without making any hard claims, we believe our model is expressive, that is, able to distinguish between many different atomic systems up to global group actions. Note that FAENet enforces symmetries via the data by building upon the idea of Frame Averaging, which preserves the expressive power of the backbone architecture. This allows to construct a maximally expressive GNN without any architectural constraints. Next, FAENet processes directly relative atom information in addition to atom characteristic number (and other atomic properties), implying that it leverages enough information to uniquely identify each graph, which is not the case of several GNNs (SchNet, DimeNet, etc.). In the embedding block, since we use two-layer MLPs, which are universal approximators (Hornik et al., 1989), we have the ability to learn injective functions mapping atomic number and geometric information to unique latent representations (provided that the dimensions of output space are large enough).

The expressive power of FANet is thus bound to the expressive power analysis of message passing itself, whose investigation is still an active topic of research. Despite GNNs revolutionising graph representation learning, there is limited understanding about their representation power and properties. Several works recently tried to fill in this gap, especially by comparing GNNs' expressive power to the (Geometric) Weisfeiler-Lehman isomorphism test (Joshi et al., 2022; Xu et al., 2018; Morris et al., 2019). To achieve maximal expressive power, GNNs require an injective aggregation function, an injective update function as well as injective readout function. This would allow to construct unique representations for each graph, enabling to distinguish them. However, it is actually extremely challenging to satisfy these conditions. Also, theory does not correlate (always) with performance for geometric GNNs.

Given the lack of theoretical guarantees about the expressive power of Geometric GNNs, Joshi et al. (2022) propose a series of experiments to assess empirically their ability to distinguish between very similar molecules, up to global E(3) transformations. In particular, the authors propose three experiments, where they train GNNs to map similar (but distinct) graphs to different labels. In particular, they try to:

1. Distinguish *k-chains*, which tests a model's ability to propagate geometric information non-locally as well as its ability to overcome oversquashing with increased model depth; see Table 3.

2. Distinguish *rotationally symmetric structures*, which test a layer's ability to identify neighbourhood orientation; see Table 4.

3. Distinguish counterexamples from (Pozdnyakov & Ceriotti, 2022), which test a layer's ability to create discriminating fingerprints for local neighbourhoods; see Table 5.
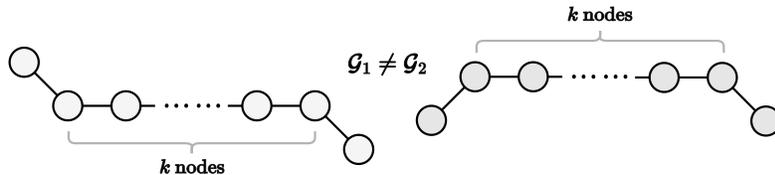
In this paper, we reproduce their experiments to assess the expressivity of FAENet (GNN alone) and FAENet-SFA (when combined with our stochastic frame averaging framework). In the coming subsections, we only refer to FAENet because results are identical both for FAENet et FAENet-SFA, which empirically proves that SFA preserves the expressive power of FAENet (for these use cases). Baseline results were reproduced using the provided codebase [10]. We trained FAENet in the exact similar fashion, using standard hyperparameters.

B.2.1. K-CHAINS

Experiment. Here, we study FAENet's ability to incorporate and propagate geometric information beyond local neighbourhoods. To do so, we consider k-chain geometric graphs, each consisting of $k + 2$ nodes where the k central nodes are arranged in a line and the 2 endpoints differ in their orientation. Because of the latter, every pair of k-chain graphs is different, up to E(3) transformations. In fact, they are $(\lfloor \frac{k}{2} \rfloor + 1)$-hop distinguishable, and $(\lfloor \frac{k}{2} \rfloor + 1)$ Geometric Weisfeiler-Lehman (GWL) iterations are theoretically sufficient to distinguish them (see (Joshi et al., 2022) for more details). Indeed, both right-hand and left-hand parts of the chain are identical with respect to rotations, meaning that one can only distinguish between these graphs by considering these neighbourhoods together. In Table 3, we train FAENet along with G-equivariant and G-invariant GNNs (with an increasing number of layers) to distinguish k-chains.

Results. Thanks to its novel functioning, free of all design constraints, FAENet enables to distinguish between these two k-chain graphs in a single message passing layer, while they are $(\lfloor \frac{k}{2} \rfloor + 1) = 3$-hop distinguishable according to Joshi et al. (2022). FAENet therefore outperforms all baselines with 1 or 2 GNN layers, including GWL algorithm. FAENet-SFA

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[10]https://github.com/chaitjo/geometric-gnn-dojo

Table 3. *k-chain geometric graphs.* We train FAENet and FAENet-SFA with an increasing number of layers to distinguish $k = 4$-chains. We report model accuracy for this binary graph classification task. An accuracy of 50 is equivalent to random predictions. We repeat the experiment 10 times and average results. Anomolous results are marked in red and expected results in green. Best results are in **bold**.

| | ($k = 4$-chains) | | **Number of layers** | | |
	GNN Layer	1	$\lfloor \frac{k}{2} \rfloor = 2$	$\lfloor \frac{k}{2} \rfloor + 1 = \mathbf{3}$	$\lfloor \frac{k}{2} \rfloor + 2$	$\lfloor \frac{k}{2} \rfloor + 3$
Inv.	IGWL	50%	50%	50%	50%	50%
	SchNet	50.0 ± 0.00	50.0 ± 0.00	50.0 ± 0.00	50.0 ± 0.00	50.0 ± 0.00
	DimeNet	50.0 ± 0.00	50.0 ± 0.00	50.0 ± 0.00	50.0 ± 0.00	50.0 ± 0.00
Equiv.	GWL	50%	50%	100%	100%	100%
	E-GNN	50.0 ± 0.00	50.0 ± 0.0	50.0 ± 0.0	50.0 ± 0.0	50.0 ± 0.0
	GVP-GNN	50.0 ± 0.00	50.0 ± 0.0	$\mathbf{100.0 \pm 0.0}$	$\mathbf{100.0 \pm 0.0}$	$\mathbf{100.0 \pm 0.0}$
	TFN	50.0 ± 0.00	50.0 ± 0.0	50.0 ± 0.0	50.0 ± 0.0	80.0 ± 24.5
	MACE	50.0 ± 0.00	50.0 ± 0.0	90.0 ± 20.0	90.0 ± 20.0	95.0 ± 15.0
	FAENet	$\mathbf{100.0 \pm 0.0}$	$\mathbf{100.0 \pm 0.0}$	$\mathbf{100.0 \pm 0.0}$	$\mathbf{100.0 \pm 0.0}$	$\mathbf{100.0 \pm 0.0}$
	FAENet-SFA	$\mathbf{100.0 \pm 0.0}$	$\mathbf{100.0 \pm 0.0}$	$\mathbf{100.0 \pm 0.0}$	$\mathbf{100.0 \pm 0.0}$	$\mathbf{100.0 \pm 0.0}$

demonstrates that Stochastic FA enables preserves the expressive power of FAENet while enforcing symmetries via the data, as discussed deeply in the paper already. Aside from that, G-equivariant GNNs often require more iterations that prescribed by GWL and do not always attain perfect accuracy, pointing to preliminary evidence of oversquashing when geometric information is propagated across multiple layers using fixed dimensional feature spaces. IGWL and G-invariant GNNs are unable to distinguish k-chains for any $k \geq 2$ and $G = O(3)$.

B.2.2. ROTATIONALLY SYMMETRIC STRUCTURES

In this experiment, we try to evaluate FAENet's ability to discriminate the orientation of structures with rotational symmetry. To do so, Joshi et al. (2022) propose to train geometric GNNs to assign two different rotations of some L-fold symmetric structures (attached to an existing system) to different classes.

Results are displayed in Table 4. Although this experiment was designed to illustrate the utility of higher-order tensors in G-equivariant GNNs, FAENet shows outstanding performance for a method using scalar vectors ($L = 0$) only. Methods using cartesian vectors cannot even discriminate two-folds structures; and methods with spherical tensors of order L are unable to identify the orientation of structures with rotation symmetry greater than L-fold, which suggests high computational cost. Here again, FAENet appears as the most desirable method, combining simple functioning, scalability and expressivity.

B.2.3. EDGE CASES

For this experiment, we look at three specific type of graphs proposed by (Pozdnyakov & Ceriotti, 2022) to illustrate the limitations of existing geometric GNNs. Each counterexample consists of a pair of local neighbourhoods that are indistinguishable when comparing their set of k-body scalars[11]. The 3-body and 4-body counterexamples correspond respectively to Fig.1(b) and Fig.2(e) in (Pozdnyakov & Ceriotti, 2022), while the 2-body counterexample corresponds to the running example of (Joshi et al., 2022). The aim of the task is the same, meaning training a single layer GNN to create discriminating fingerprints for local neighbourhoods.

Results are reported in Table 5. While Joshi et al. (2022) concluded that Geometric GNN layers with body order k cannot distinguish the corresponding k-body counterexample, FAENet does enable to distinguish the 4-body counterexample while

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[11] Body-order of scalarisation: number of nodes involved in computing local invariant scalars.
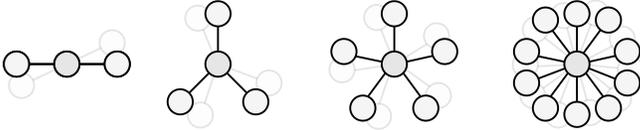
	GNN Layer	**Rotational symmetry**			
		2 fold	3 fold	5 fold	7 fold
Scalar	FAENet$_{L=0}$	**100.0 ± 0.0**	**100.0 ± 0.0**	**100.0 ± 0.0**	**100.0 ± 0.0**
	FAENet-SFA$_{L=0}$	**100.0 ± 0.0**	**100.0 ± 0.0**	**100.0 ± 0.0**	**100.0 ± 0.0**
Cart.	E-GNN$_{L=1}$	50.0 ± 0.0	50.0 ± 0.0	50.0 ± 0.0	50.0 ± 0.0
	GVP-GNN$_{L=1}$	50.0 ± 0.0	50.0 ± 0.0	50.0 ± 0.0	50.0 ± 0.0
Spherical	TFN/MACE$_{L=1}$	50.0 ± 0.0	50.0 ± 0.0	50.0 ± 0.0	50.0 ± 0.0
	TFN/MACE$_{L=2}$	**100.0 ± 0.0**	50.0 ± 0.0	50.0 ± 0.0	50.0 ± 0.0
	TFN/MACE$_{L=3}$	**100.0 ± 0.0**	**100.0 ± 0.0**	50.0 ± 0.0	50.0 ± 0.0
	TFN/MACE$_{L=5}$	**100.0 ± 0.0**	**100.0 ± 0.0**	**100.0 ± 0.0**	50.0 ± 0.0
	TFN/MACE$_{L=7}$	**100.0 ± 0.0**	**100.0 ± 0.0**	**100.0 ± 0.0**	**100.0 ± 0.0**

Table 4. Rotationally symmetric structures. We train single layer G-equivariant GNN models (with order L tensors) to distinguish two *distinct* rotated versions of L-fold symmetric structures. We report model accuracy of this binary graph classification task, averaged over 10 runs. Anomolous results are marked in red and expected results in green. Best in **bold**.

	GNN Layer	**Edge cases**		
		2-body	3-body	4-body
Inv.	SchNet$_{2\text{-body}}$	50.0 ± 0.0	50.0 ± 0.0	50.0 ± 0.0
	DimeNet$_{3\text{-body}}$	**100.0 ± 0.0**	50.0 ± 0.0	50.0 ± 0.0
$O(3)$-Equiv.	E-GNN$_{2\text{-body}}$	50.0 ± 0.0	50.0 ± 0.0	50.0 ± 0.0
	GVP-GNN$_{3\text{-body}}$	**100.0 ± 0.0**	50.0 ± 0.0	50.0 ± 0.0
	TFN$_{2\text{-body}}$	50.0 ± 0.0	50.0 ± 0.0	50.0 ± 0.0
	MACE$_{3\text{-body}}$	**100.0 ± 0.0**	50.0 ± 0.0	50.0 ± 0.0
	FAENet$_{2\text{-body}}$	**100.0 ± 0.0**	**100.0 ± 0.0**	**100.0 ± 0.0**
	FAENet-SFA$_{2\text{-body}}$	**100.0 ± 0.0**	**100.0 ± 0.0**	**100.0 ± 0.0**

Table 5. Counterexamples from (Pozdnyakov & Ceriotti, 2022). We train geometric GNNs at distinguishing edge cases structures, which are (supposedly) indistinguishable using k-body scalarisation. We repeat the experiment 10 times and report the accuracy. FAENet manages to distinguish them all while presenting 2-body order only.

being only a 2-body method[12]. This demonstrates its expressivity and the strength of the novel paradigm proposed (i.e. unconstrained GNN functioning; symmetries enforced via the data without loosing expressive power).

B.3. Ablation Study on IS2RE

In this section, we perform a thorough ablation study of FAENet on the OC20 IS2RE dataset. Even if we motivate a simple and efficient GNN architecture, we propose in this paper a new paradigm for incorporating geometric information into atom representations (since we construct a model free of all symmetry-preserving constraints). We therefore compared various relevant design choices with respect to the embedding block, the message passing scheme and the output block. The results displayed in Table 6 empirically validate FAENet's current architecture. In particular, we tried the following ideas and drew the corresponding conclusions:

- *Only $\vec{r}_{ij}$* involves using $\vec{r}_{ij}$ alone in the embedding block to derive each edge representation, instead of $\vec{r}_{ij}||RBF(d_{ij})$.

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[12]Actually, since FAENet utilises non-linearities in its message passing, the concept of k-body order looses relevance. FAENet could be considered as a many-body method, demonstrating the advantage of its frame based paradigm.

Table 6. Ablation study of FAENet – we study the impact of various components on the model performance-scalability trade-off. Scalability is measured via the throughput (the number of processed samples per seconds) while performance is measured by the Energy MAE, averaged across all four validation splits.

Component	Throughput (samples/s)	MAE (average)
FAENet (baseline)	**2129**	**568**
- *Only $\vec{r}_{ij}$*	2390	603
- *Only Spherical Harmonics*	1441	626
- *All*	1560	585
- *2nd layer in MLP*	1828	588
- *Simple MP*	2706	606
- *Basic MP*	3065	629
- *Updownscale MP*	1505	592
- *Attention mecha*	1929	712
- *No complex MP*	2265	586
- *Simple Energy Head*	2224	589
- *No Jumping Connections*	2038	582

Please refer to Equation (7).
$\rightarrow$ Using a radial basis function (RBF) of distance information in addition to r_{ij} improves performance significantly. Running time is a bit higher, obviously, but not enough to outweigh the performance gains.

- *Only Spherical harmonics*: we replace $\left(\vec{r}_{ij}||\text{RBF}(d_{ij})\right)$ by spherical harmonics (of order up to 3) on $\frac{\vec{r}_{ij}}{||\vec{r}_{ij}||}$ in Equation (7). Spherical harmonics form a basis for irreducible representations in SO(3) and were shown to be very informative quantities by previous works (Frank et al., 2022; Thomas et al., 2018; Batatia et al., 2022).
  $\rightarrow$ We show that using directly a 2-layer MLP of relative information is more attractive than (higher-order) spherical harmonics.

- *All*. We concatenate spherical harmonics to directional information and distance information ($\vec{r}_{ij}$, d_{ij} and SH).
  $\rightarrow$ We show that adding spherical harmonics in addition to directional information does not provide additional benefits for this task.

- *2-layer-MLP*: we replace the two-layer-MLP of the Embedding block by a one-layer-MLP, see Equation (7).
  $\rightarrow$ No clear conclusion here. Reducing MLP to 1-layer hurts performance but improves slightly compute time. We favoured performance here.

- *Basic MP*: we do not transform the edge embedding inside the interaction block. $\mathbf{f}_{ij}^{(l)} = \mathbf{e}_{ij}$
  $\rightarrow$ This message passing form is much more scalable (obviously) but significantly less efficient. This proves the relevance of refining the filters using edge information and atom information at each layer.

- *Simple MP*: we do not compute the filter using node features $\mathbf{h}_i$ and $\mathbf{h}_j$. In mathematical form, we use $\mathbf{f}_{ij}^{(l)} = \sigma(\text{MLP}(\mathbf{e}_{ij}))$ instead of $\mathbf{f}_{ij}^{(l)} = \sigma(\text{MLP}(\mathbf{e}_{ij}||\mathbf{h}_i^{(l)}||\mathbf{h}_j^{(l)}))$.
  $\rightarrow$ This Message Passing form is shown to be more scalable but not as efficient. Using an edge's endpoints helps learn more relevant filters.

- *Updownscale MP*: we additionally transform node embeddings inside each interaction block, before the continuous convolution.
  $\rightarrow$ This does not show increased performance, validating current design.

- *Attention mecha*: we weight each message by an attention coefficient, learned using neighbour's feature vector, node's feature vector and edge embedding. This is expressed as $h_i^{(l+1)} = h_i^{(l)} + \sum_{j \in N_i} \alpha_{ij} W h_j^{(l)}$ with $\alpha_{ij} = Att(h_i, h_j, \vec{r}_{ij}) = h_i^T[e_{ij} \odot h_j]/\sqrt{F}$.
  $\rightarrow$ It does not yield good results in practice. Current filter construction is more efficient.

- *No complex MP*: we use two-layers MLP instead of one-layer MLP in each Interaction block of the model (see Fig. 1).
  → This was not found beneficial, both in terms of performance and running time.

- *Simple Energy Head*: involves replacing the current weighted average of atom's final representation to compute graph-level prediction (in the Output Block) by the standard global pooling approach.
  → The weighted average is found (slightly) beneficial.

- *No jumping connections*: We remove the jumping connections from each Interaction Block to the Output block.
  → Jumping connections are found (slightly) beneficial.

C. Extended Results

C.1. Experimental setup – Software and Hardware

We would like to acknowledge and thank authors of the following Python libraries that we have used to realise this work, in particular Pytorch v.1.13 (Paszke et al., 2019), PyTorch Geometric v2.2.0 (Fey & Lenssen, 2019) NumPy v1.23.5 (Harris et al., 2020) and authors of the OCP Github repository which we used as a starting point.

Experiments were run on 1 NVIDIA RTX8000 GPUs. We would like to also thank our lab's IT staff for putting together and maintaining our computing infrastructure as well as for supporting researchers.

C.2. Hyper Parameters

We detail FAENet's list of hyperparameters for all four datasets IS2RE, S2EF, QM7-X and QM9 in Table 7.

Table 7. Model and optimization hyper parameters for FAENet used in experiments reported.

	OC20-IS2RE	OC20-S2EF	QM7X	QM9
Activation function	swish	swish	swish	swish
Cutoff distance	6	6	5	6
Force head hidden channels	-	256	256	-
Hidden channels	384	256	500	400
Max number of neighbors	40	30	40	30
Number of filters	480	480	400	480
Number of gaussians	104	136	50	100
Interaction blocks	5	7	5	5
Period & Group channels	64	64	32	32
Tag channels	64	32	-	-
Optimizer	AdamW	AdamW	AdamW	AdamW
Scheduler	Cosine Annealing	Cosine Annealing	Reduce On Plateau	Reduce On Plateau
Warmup steps	6,000	30,000	3,000	3,000
Learning Rate	0.002	0.00025	0.0002	0.001
Batch size	256	192	100	64
Energy loss coeff.	-	1	1	-
Force loss coeff.	-	100	100	-
Energy grad. loss coeff.	-	5	15	-
Steps	22K	85K	1.4M	1.5M

C.3. QM9 Dataset

We provide the detailed list of per-model property MAE performance in Table 8. The results reported in are comptued as followed. For each property p we compute the *relative improvement* with respect to SchNet as:

$$R_p^{\text{Model}} = \frac{\text{MAE}_p^{\text{SchNet}} - \text{MAE}_p^{\text{Model}}}{\text{MAE}_p^{\text{SchNet}}}. \tag{11}$$

Property	Unit	SchNet	PhysNet	MGCN	DimeNet	DimeNet++	SE(3)-T	SphereNet	ComENet	FAENet
μ	D	0.033	0.0529	0.0560	0.0286	0.0297	0.051	0.0245	0.0245	0.0289
α	a_0^3	0.235	0.0615	0.0300	0.0469	0.0435	0.142	0.0449	0.0452	0.0527
ϵ_{HOMO}	meV	41	32.9	42.1	27.8	24.6	35.0	22.8	23.1	24.5
ϵ_{LUMO}	meV	34	24.7	57.4	19.7	19.5	33.0	18.9	19.8	20.0
$\Delta\epsilon$	meV	63	42.5	64.2	34.8	32.6	53.0	31.1	32.4	40.2
$\langle R^2\rangle$	a_0^2	0.073	0.765	0.110	0.331	0.331	-	0.268	0.259	0.498
ZPVE	meV	1.7	1.39	1.12	1.29	1.21	-	1.12	1.20	1.26
U_0	meV	14	8.15	12.9	8.02	6.32	-	6.26	6.59	6.79
U	meV	19	8.34	14.4	7.89	6.28	-	6.36	6.82	6.80
H	meV	14	8.42	14.6	8.11	6.53	-	6.33	6.86	6.74
$0G$	meV	14	9.4	16.2	8.98	7.56	-	7.78	7.98	7.91
c_v	$\frac{cal}{mol\,K}$	0.033	0.028	0.038	0.025	0.023	0.052	0.022	0.024	0.023
$R_{p=1..11}$	mean	-	0.247	-0.021	0.366	0.419	-	0.445	0.423	0.395
	std	-	0.331	0.421	0.208	0.226	-	0.205	0.205	0.212
Time	s	98	-	-	582	330	-	345	224	87

Table 8. Comparisons between FAENet and baseline models on QM9 dataset in terms of (1) MAE, (2) *relative improvement* $R_{p=1..11}$ with respect to SchNet, and (3) Time, which we measured for 1 epoch of training using the same hardware and software.

We then compute and report in Fig. 3 the mean and standard deviation of the series $R^{\text{Model}}_{p=0..11}$. Note that we treat property $\langle R^2\rangle$ as an outlier and exclude it from the aforementioned statistics because SchNet is oddly the best by a very large margin and we have not been able to reproduce such good results. In addition, in spite of our effort to re-run baselines ourselves, we could not re-implement PhysNet (Unke & Meuwly, 2019), MGCN (Wang et al., 2020) nor SE(3)-Transformers (Fuchs et al., 2020) in our pipeline.

C.4. OC20 full table of results

We report the full results for OC20 IS2RE in Table 9 and for S2EF 2M in Table 10.

Table 9. Exhaustive table of results on *OC20 IS2RE* "All" dataset, for all 4 validation splits (ID, OOD Ads, OOD Cat, OOD Both). We measure performance in terms of energy MAE and the percentage of Energy within Threshold (EwT) of the ground truth energy. We average results across the four val splits (Average). The best performance is shown in bold and the second best is shown with underlines. Scalability is measured with training time for one epoch (train, in minutes) and inference throughput (infer, number of samples processed in a second).

Model	Time		Energy MAE [eV] ↓					EwT ↑				
	Train ↓	Infer. ↑	ID	OOD Ads	OOD Cat	OOD Both	Average	ID	OOD Ads	OOD Cat	OOD Both	Average
SchNet	9min	3597	0.6372	0.7342	0.6611	0.7035	0.6840	2.96%	2.22%	3.03%	2.38%	2.65%
DimeNet++	170min	115	0.5716	0.7224	0.5612	0.6615	0.6283	4.26%	2.06%	4.10%	3.21%	3.40%
SphereNet	290min	83	0.5632	0.6682	0.5590	0.6190	0.6023	4.56%	2.70%	4.59%	2.70%	3.64%
EGNN	-	-	0.5497	0.6851	0.5519	0.6102	0.5992	4.99%	2.50%	4.71%	2.88%	3.77%
PaiNN	-	-	0.5781	0.7037	0.5701	0.6139	0.6164	4.31%	2.60%	4.35%	2.74%	3.50%
Forcenet	120min	157	0.6582	0.7017	0.6323	0.6285	0.6551	3.14%	2.47%	2.58%	2.83%	3.13%
SpinConv	-	-	0.5583	0.7230	0.5687	0.6738	0.6309	4.08%	2.26%	3.82%	2.33%	3.12%
GemNet-T	200min	104	0.5561	0.7342	0.5659	0.6964	0.6382	4.51%	2.24%	4.37%	2.38%	3.38%
ComENet	20min	416	0.5558	0.6602	0.5491	0.5901	0.5888	4.17%	2.71%	4.53%	2.83%	3.56%
GNS	-	-	0.5400	0.6500	0.5500	0.5900	0.5825	-	-	-	- -	
Graphormer	-	-	**0.4000**	**0.5700**	**0.4200**	**0.5000**	**0.4725**	**8.97**%	**3.45**%	**8.18**%	**3.79** %	**6.09** %
FAENet	12min	2469	0.5446	0.6115	0.5707	0.5449	0.5679	4.46%	2.95%	4.67%	3.01%	3.78%

Table 10. Exhaustive table of results on *OC20 S2EF* "2M" dataset, for all 4 validation splits (ID, OOD Ads, OOD Cat, OOD Both). This table expands on Table 1. We measure performance in terms of energy MAE and forces MAE. We average results across the four val splits (Average). The best performance is shown in bold and the second best is shown with underlines. Scalability is measured with training time for one epoch (train, in minutes) and inference throughput (infer, number of samples processed in a second).

	Time		Energy MAE [eV] $\downarrow$					Force MAE [meV] $\downarrow$				
Model	Train $\downarrow$	Infer.$\uparrow$	ID	OOD Ads	OOD Cat	OOD Both	Average	ID	OOD Ads	OOD Cat	OOD Both	Average
SchNet	98min	509	1.4075	1.5228	1.4383	1.5982	1.4917	76.3	86.4	79.7	90.1	83.1
DimeNet++	1157min	52	0.7438	0.8425	0.7788	0.8734	0.8096	57.3	68.6	66.0	73.5	66.3
ForceNet	1476min	36	0.6757	0.7667	0.6650	0.8987	0.7548	55.3	61.6	72.7	54.8	61.0
GemNet-OC	–	18	-	-	-	-	**0.2860**	–	–	–	–	**25.7**
FAENet	75min	623	0.3960	0.4548	0.4295	0.5766	<u>0.4643</u>	51.1	58.9	50.7	69.4	<u>57.5</u>

D. Empirical Evaluation of Model Properties

In this section, we verify the correctness of our symmetry preserving framework, including all different theoretical properties.

D.1. Methods and Metrics

For this experiment, we look at different methods: (1) FAENet combined with full frame averaging (Full-FA); (2) FAENet with Stochastic Frame Averaging (SFA); (3) FAENet with Stochastic Frame Averaging for the SE(3) group instead of E(3), i.e. less frames to sample from; (4) FAENet with data augmentation (DA); (5) FAENet alone (No-FA); (6) untrained FAENet (No-Train); (7) ForceNet alone (8) SchNet alone. For each method, we study some targeted equivariant and invariant properties via the following metrics:

- *Pos*. To check the correctness of our implementation of frame averaging, we let *Pos* be a binary variable which takes the value 1 if, for arbitrary datapoints D_1, D_2 such that $D_2 = \rho_1^{(-1)}(g)(D_1)$ for any $g \in E(3)$, $\mathfrak{C}_1 = \mathfrak{C}_2$, i.e. they lead to the same representation in the projected space; and 0 otherwise. By definition, it shall be the case for *Full FA* and not for the others. For methods which don't use FA, we set $pos = 0$ if D_1 and D_2 are mapped to the same representation.

- *Rot-I* and *Refl-I*. We measure the rotation invariance *Rot-I* and reflection invariance *Refl-I* property of FAENet by computing the difference in GNN prediction between every samples D_1 (of the ID val split) and D_2 defined as a SO(3) transformation of D_1, as above. We also compute the percentage variation in prediction with respect to energy (when rotated), with *%-diff*: $100 \times \frac{Rot\text{-}I}{y_{true}}$.

- *F-Rot-E* and *F-Refl-E*. In a similar fashion, we evaluate force predictions's rotation and reflection equivariance to E(3) transformations, denoted respectively *F-Rot-E* and *F-Refl-E*.

D.2. Results

For the IS2RE and S2EF datasets, we display the obtained results in Table 11 and in Table 12. From them, we draw the following conclusions:

- **Full FA** yields near-perfect invariant/equivariant model predictions, proving the correctness of our implementation. Note that metrics are not exactly 0 for Full FA (and Schnet) due to software precision limitations.

- **Stochastic Frame Averaging (SFA)** produces a great approximation to equivariant/invariant predictions: *Rot-I*, *F-Rot-E*, *Refl-I* and *F-Refl-E* get close to 0 (the scale is in meV, not eV), *%-diff* is below 2%. Invariant metrics on IS2RE are divided by a factor of ≈ 8 for SFA compared to No-FA or ForceNet, showing that our approach learns efficiently to enforce symmetries. In different terms, energy predictions are much closer for two rotated (or reflected) versions of the same graph when using SFA compared to DA or No-FA. Besides, SFA is our best performing model. When comparing to Full FA (or even other equivariant/invariant models Table 9), SFA demonstrates that perfectly enforcing symmetries does not necessarily lead to better performance. Presenting a setting that makes it easy for the model to learn to accomplish the required task seems essential too.

- **SE**(3)**-SFA** approximation yields slightly better metrics because FAENet has to learn data symmetries from less frames (cf SFA), which is easier. By construction, it is less invariant/equivariant to reflections. However, this does not seem to impact performance for this application.

- **Data augmentation (DA)** achieves worse symmetry metrics and MAE score on both datasets, with similar run time (the PCA computation is negligible for FA). Equivariance seems harder to learn than invariance for DA, compared to SFA. For instance, on S2EF, rotation equivariance for DA decreases by 60% compared to SE(3)-SFA while performance drops by 9%. We can expect even bigger differences on other datasets since the z-axis is fixed in OC20: the adsorbate is always on top of the catalyst. This means that we only look at 2D rotations/reflections, making it easier to learn symmetries via data augmentation. SFA could be (even) more useful on "pure" 3D cases. This being said, DA gives very satisfying results on this dataset, confirming that enforcing symmetries implicitly via the data is an attractive (and under-exploited) approach in materials modeling.

- **No-FA**, i.e. not using any kind of symmetry preservation techniques, does not lead to invariant/equivariant predictions (e.g. invariance metrics are roughly 8 times higher than for SFA) although these metrics improve significantly compared to **No-Train** on S2EF. This means that FAENet learn symmetry to some extent, even when not directly encouraged to do so. No-FA showcases a "significant" decrease in performance (569 to 595, 464 to 522), proving that enforcing symmetries to some extent still seem desirable. Again, equivariance seems harder than invariance to learn for non-symmetry preserving methods. Again, we have 2D symmetries and expect this gap to be higher on QM9/QM7-X.

- Repeating these experiments on **ForceNet** for IS2RE confirms the trend observed for FAENet: SFA is more efficient than DA, both in terms of performance and in terms of enforcing equivariance/invariance. It also reveals that the design of FAENet is superior, probably because it leverages more efficiently geometric information.

Table 11. Test of FAENet's theoretical properties on IS2RE depending on the different symmetry-preservation techniques used. We report several metrics (see D.1) measuring perfect symmetry-preservation (Pos), rotation invariance (Rot-I), reflection invariance (Refl-I), percentage variation in prediction when rotated (%-diff) and energy average validation MAE. Everything (but Pos) is given in meV. Note that we ideally want to minimise these metrics.

Model/Metrics	Pos	Rot-I	%-diff	Refl-I	MAE
Full FA	0	0.07	0.26	0.05	578
SFA	1	6.57	2.12	6.54	569
SE(3)-SFA	1	6.35	1.87	8.90	567
DA	1	7.41	2.97	8.22	582
No-Train	1	21.8	6.03	24.9	595
No-FA	1	51.1	18.3	55.3	595
SchNet	0	0.00	0.00	0.00	684
ForceNet	1	61.6	14.8	63.0	655
Forcenet-DA	1	18.9	7.47	20.8	683
Forcenet-SFA	1	7.11	2.59	7.14	638

Table 12. Test FAENet's theoretical properties on S2EF depending on the different symmetry-preservation methods used. We report several metrics (see D.1) measuring implementation correctness (*Pos*), energy rotation invariance (*Rot-I*), energy reflection invariance (*Refl-I*), force reflection equivariance (*F-Refl-E*), force rotation equivariance (*F-Rot-E*) and energy MAE. Everything is in meV.

Model/Metrics	Pos	Rot-I	Refl-I	F-Rot-E	F-Refl-E	MAE
Full FA	0	0.05	0.03	0.07	0.05	489
Stocha. FA	1	6.94	6.96	4.79	4.78	464
SE(3)-SFA	1	6.23	8.86	3.78	5.25	478
DA	1	8.88	9.12	6.12	6.46	504
No-FA	1	26.81	27.61	10.27	10.12	522
No-train	1	86.03	87.09	32.31	31.92	–
SchNet	0	0.00	0.00	0.00	0.00	918