ForceNet: A Graph Neural Network for Large-Scale Quantum Calculations

Weihua Hu 1  Muhammed Shuaibi 2  Abhishek Das 3  Siddharth Goyal 3  Anuroop Sriram 3  Jure Leskovec 1  Devi Parikh 3 4  C. Lawrence Zitnick 3

Abstract
With massive amounts of atomic simulation data available, there is a huge opportunity to develop fast and accurate machine learning models to approximate expensive physics-based calculations. The key quantity to estimate is atomic forces, where the state-of-the-art Graph Neural Networks (GNNs) explicitly enforce basic physical constraints such as rotation-covariance. However, to strictly satisfy the physical constraints, existing models have to make tradeoffs between computational efficiency and model expressiveness. Here we explore an alternative approach. By not imposing explicit physical constraints, we can flexibly design expressive models while maintaining their computational efficiency. Physical constraints are implicitly imposed by training the models using physics-based data augmentation. To evaluate the approach, we carefully design a scalable and expressive GNN model, ForceNet, and apply it to OC20 (Chanussot et al., 2020), an unprecedentedly-large dataset of quantum physics calculations. Our proposed ForceNet is able to predict atomic forces more accurately than state-of-the-art physics-based GNNs while being faster both in training and inference. Overall, our promising and counter-intuitive results open up an exciting avenue for future research.

1. Introduction
Recently, massive physics-based data has been generated by ever-increasing scientific compute (Chanussot et al., 2020; Nakata et al., 2019). This provides a huge opportunity for Machine Learning (ML) approaches to efficiently and accurately model complex physical systems (Bapst et al., 2020; Battaglia et al., 2016; Gilmer et al., 2017; Kipf et al., 2018; Klicpera et al., 2020a;b; Sanchez-Gonzalez et al., 2020; Schütt et al., 2017a). An accurate ML model trained on large data can be used to perform inference orders-of-magnitude faster than the original physics-based calculations.

Of particular practical interest is approximating atomic forces of quantum mechanical systems. This is because the underlying quantum calculations are expensive (several hours per system) (Parr, 1980), and the resulting atomic forces can be used for diverse chemistry applications, such as structure relaxations, molecular dynamics, structural analyses, as well as transition state calculations. (Behler, 2016; del Río et al., 2019; Frederiksen et al., 2007; Henkelman & Jónsson, 2000; Henkelman et al., 2000)

The state-of-the-art approach to predicting atomic forces is physics-based message-passing Graph Neural Networks (GNNs) (Gilmer et al., 2017), with the representative models being SchNet (Schütt et al., 2017a) and DimeNet (Klicpera et al., 2020a;b). These GNNs first predict the energy of the entire system in a rotation-invariant manner, and then predict the per-atom forces by taking the derivative of the energy with respect to the atomic positions. By the architecture’s design, these GNNs produce forces that obey the basic physical rules of rotation-covariance and energy-conservation.

However, designing effective GNNs, while satisfying these physical rules is highly non-trivial. For instance, SchNet (Schütt et al., 2017b) is computationally efficient, but the model only uses atomic distances in its message passing in order to ensure rotation-invariance of its energy prediction. Consequently, SchNet fails to capture the 3D structure explicitly, resulting in sub-optimal generalization performance. The recent DimeNet and DimeNet++ (Klicpera et al., 2020a;b) additionally capture bond angle information in its message passing, but this comes with the cost of expensive message computations involving atom triplets to ensure the rotation-invariance. As a result, DimeNet necessitates tremendous compute to scale to a massive dataset (Figure 1 (left))—1600 GPU days to train DimeNet++-large. Moreover, even DimeNet is unable to model an important physical feature of torsion angles (Leach, 2001), failing to capture the full 3D information in its message passing.

Here we explore an alternative approach, building on the recent framework of Graph Network-based Simulators...
Specifically, by not explicitly imposing physical constraints in the model architecture, we can flexibly design expressive GNN models and use the full 3D atomic positions in a scalable manner. In exchange, the predicted forces are translation-invariant but no longer rotation-covariant. As we demonstrate empirically, this issue can be alleviated by training models on a massive dataset with rotation data augmentation. In other words, we impose physical constraints to the model implicitly through physics-based data rather than explicitly through architectural constraints. Our model also does not explicitly enforce energy conservation. However, this removes the memory-intensive calculations in physics-based GNNs, i.e., compute forces through energy gradients that require second-order derivatives to optimize.

To realize our approach, we carefully design a GNN model that accurately captures 3D atomic structure in a scalable and flexible manner. The resulting model is ForceNet that uses an expressive message passing architecture with carefully-chosen basis and non-linear activation functions.

We evaluate ForceNet on OC20 (Chanussot et al., 2020), a recently-introduced large-scale dataset of quantum physics calculations with 200+ million large atomic structures (20–200 atoms) useful for discovering new catalysts for energy applications (Jouny et al., 2018; Seh et al., 2017; Zitnick et al., 2020) (Figure 2). The dataset was constructed with an unprecedented 70 million CPU hours of compute performing Density Functional Theory (DFT)-based quantum calculations (Parr, 1980)—more than 20 times the compute as compared to the conventional quantum physics datasets of QM9 (Ramakrishnan et al., 2014) and Alchemy (Chen et al., 2019), making it ideal for scalable deep learning approaches. Moreover, unlike many existing quantum physics datasets, OC20 provides non-equilibrium structures of molecules, i.e., 3D structure with non-zero atomic forces, making it a good testbed of our model.

Even without any explicit physical constraints, ForceNet is able to achieve higher accuracy than physics-based GNNs when trained with comparable computational resources (Figure 1 (left)). Moreover, ForceNet (resp. ForceNet-large) achieves prediction errors that are comparable to the state-of-the-art DimeNet++ (resp. DimeNet++-large) with 6 (resp. 8) times less inference time (Figure 1 (right)). Finally, compared to DimeNet++, ForceNet-large achieves more accurate force prediction, while being faster both in training and inference (Figure 1 (left) and (right)).

To understand the ForceNet’s design choices, we perform extensive ablation studies on each architectural component of ForceNet. We find that the expressive edge-level computation for accurately modeling 3D atomic interactions contributes most to the model performance. Overall, we demonstrate that even without explicit physical constraints, a scalable expressive GNN provides promising performance in modeling complex physical systems, opening up an exciting avenue for future research.

2. Related Work

ML for approximating quantum physics calculations has been extensively studied in the literature (Chen et al., 2019; Chmiela et al., 2017, 2018; Christensen & von Lilienfeld, 2020; Gilmer et al., 2017; Khosrabi & Peterson, 2016; Klipper et al., 2020a,b; Ramakrishnan et al., 2014; Schütt et al., 2017a). These studies have been either on relatively small-scale datasets (in the order of 100K structures), small
we demonstrate empirically, the off-the-shelf GNN model (hence, energy-centric). Rotationally-invariant GNNs are used to predict the energy. Atomic forces are then predicted implicitly through negative gradients of energy with respect to the atomic positions, which can be directly regressed to the ground-truth forces using the second-order derivatives. The architecture guarantees that the force-field obeys the basic physical rules of rotation-covariance and energy-conservation.

Many advanced GNN architectures have been proposed under the energy-centric framework, such as SchNet (Schütt et al., 2017a), DimeNet (Klicpera et al., 2020b), and its recent improvement, DimeNet++ (Klicpera et al., 2020a). As we discuss in the introduction, these models are either computationally expensive (DimeNet involves message passing over triplets of atoms) or unable to explicitly capture angular information among a set of atoms (SchNet’s message passing only depends on atomic distances). Although DimeNet captures angular information, it is still restricted to bond angles, and torsion angles are not captured explicitly (Leach, 2001). Capturing the full angular information in a rotationally-invariant manner would require even more computation, e.g., message passing over atom quadruplets.

Our scalable expressive message passing architecture is built from SchNet’s continuous filter convolution architecture, where we make an important extension to resolve a number of critical issues when adopting it to the force-centric GNS framework (see Section 3.1.1 for details).

3. ForceNet

Here we introduce ForceNet by describing its model architecture as well as the effective data augmentation strategy to encourage rotation covariance of ForceNet’s predictions. The input to ForceNet is an atomic structure, i.e., a set of atoms and their 3D spatial positions (Figure 2). The output is a 3D vector for each node, representing the predicted $(x, y, z)$ atomic force.

3.1. Model architecture

ForceNet represents atoms as nodes in a GNN and the atomic interactions as edges. The node input features specify the atom’s atomic number and other properties (9-dimensional vector adapted from Xie & Grossman (2018)). Edges in the GNN are constructed from a radius graph of neighboring atoms (Sanchez-Gonzalez et al., 2020; Schütt et al., 2017a). Let $N_t(c)$ denote a set of neighboring atoms that are within the cutoff-distance $c$ away from the target atom $t$. On average an atom has 35 neighbors. A directed edge from source atom $s$ to target atom $t$ is drawn for $s \in N_t(c)$. Let $d_{st} \in \mathbb{R}^3$ be their relative displacement, i.e., a vector pointing from atom $s$ to atom $t$.

ForceNet follows the encoder-decoder architecture of the GNS framework (Battaglia et al., 2016; Kipf et al., 2018; Sanchez-Gonzalez et al., 2020). The encoder uses scalable iterative message passing to compute node embeddings $h_t$
that capture the 3D structure surrounding each atom, and the decoder uses an Multi-Layer Perceptron (MLP) to directly predict per-atom forces from these embeddings. The encoder updates \( h_t \) as:

\[
  h_t^{(k+1)} = F_n \left( m_t + \sum_{s \in N_t} m_{st} \right) + h_t^{(k)},
\]

where the messages \( m_{st} \) and \( m_t \) are summed and passed through the function \( F_n : \mathbb{R}^D \rightarrow \mathbb{R}^D \) that is a 1-hidden-layer MLP with batch normalization (Ioffe & Szegedy, 2015). The dimensionality of the node and hidden layer features is \( D \). Equation (1) follows standard GNN embedding update formulations (Gilmer et al., 2017) with the addition of a residual connection, \( h_t^{(K)} \) (He et al., 2016). We define the pairwise messages \( m_{st} \) and self message \( m_t \) in Section 3.1.1.

The decoder is computed using the last layer \( K \)'s node embeddings \( h_t^{(K)} \), \( f_t = F_f(h_t^{(K)}) \) where \( f_t \) is the 3D force of atom \( t \), and \( F_f \) is a 1-hidden-layer MLP with batch normalization.

The critical aspect of ForceNet is its encoder and specifically the scalable message computation that effectively captures the non-linear and complex 3D atomic interactions to predict the atomic forces. In the following, we present three key architectural components in our message computation.

### 3.1.1 Conditional Filter Convolution

We first present conditional filter convolution, a simple yet effective extension of SchNet's scalable continuous filter convolution (Schütt et al., 2017a). The original continuous filter convolution uses the distance information between neighboring atoms to compute the filter, which is then applied to the embeddings of source atoms. However, this approach has a series of limitations, especially when transitioning from an energy-centric to a force-centric model.

First, to ensure rotation-invariant energy prediction, SchNet’s continuous filter only uses the atom distance as the input edge feature. Hence, the angular information is lost in the message passing. More crucially, the resulting node embeddings are rotation-invariant, but forces need to rotate together with a molecular system. Furthermore, the filter does not depend on the source and target atoms. However, changes in their atom types can result in significant differences in forces between the atoms even if they are placed at a similar distance, because of their varying electronic properties (Gibbs et al., 1998). Hence, the filter may not be expressive enough to model complex non-linear atomic interactions.

We resolve these issues by using our \( E \)-dimensional edge feature \( e_{st} \) (described below) that encodes rotation-covariant directional information, and conditioning on both the source \( h_s^{(k)} \) and target \( h_t^{(k)} \) node information:

\[
  m_{st} = F_c(h_s^{(k)}, e_{st}, h_t^{(k)}) \odot F_d(h_s^{(k)}),
\]

where \( F_c : \mathbb{R}^E \times \mathbb{R}^E \times \mathbb{R}^D \rightarrow \mathbb{R}^D \) is the conditional filter, and \( F_d : \mathbb{R}^D \rightarrow \mathbb{R}^D \) is a learnable linear function that transforms the source node embeddings before the filter is applied. The edge feature is defined as \( e_{st} = \text{Concat}(n_{st}, p_{st}/c) \in \mathbb{R}^7 \), where \( n_{st} \equiv d_{st}/\|d_{st}\| \) is a normalized directional vector and \( p_{st} \in \mathbb{R}^4 \) is a list of four atomic distances \( \|d_{st}\|, \|d_{st}\| - \alpha_s, \|d_{st}\| - \alpha_t, \|d_{st}\| - \alpha_s - \alpha_t \) that take into account the atomic radii (Slater, 1964) \( \alpha_s \) and \( \alpha_t \) of atoms \( s \) and \( t \), respectively.

The conditional filter \( F_c \) combines the raw edge features \( e_{st} \) with the node embeddings to encode the interactions between atoms \( s \) and \( t \) (Figure 3) and is defined as:

\[
  F_c(h_s^{(k)}, e_{st}, h_t^{(k)}) = \alpha(\|d_{st}\|) \cdot F_c(h_s^{(k)}, B(e_{st})),
\]

where \( \alpha(x) = \cos(\pi x/2c) \) is a scalar that decays to zero as \( \|d_{st}\| \) approaches the distance cutoff \( c \). \( F_c \) is a 2-hidden-layer MLP with the hidden size of \( D \) and the three input vectors are concatenated as input. \( B : \mathbb{R}^E \rightarrow \mathbb{R}^D \) is the basis function we discuss in the next section, and \( F_d : \mathbb{R}^D \rightarrow \mathbb{R}^D \) is a learnable linear function that maps the \( B \) dimensional vector to a \( D \) dimensional vector for input to \( F_c \). The parameters for \( F_b \) are shared across layers, while no other parameters are shared across layers. Finally, the self message \( m_t \) is defined by applying an element-wise product between learnable filter \( v \in \mathbb{R}^D \) and \( F_d, \) i.e., \( m_t = v \odot F_d(h_t^{(k)}) \).

**Comparisons with the existing GNS model.** Notice that the filter \( F_c \) itself already contains the source node information \( h_s^{(k)} \), and may be directly aggregated, as done in the off-the-shelf GNS models (Bapst et al., 2020; Sanchez-Gonzalez et al., 2020). Nonetheless, we empirically find that explicitly applying \( F_c \) on \( F_d \) through the element-wise dot
product in Equation (2) significantly improves the performance. There are also other subtle but important differences between ForceNet and the existing GNS model, such as the use of basis functions (Section 3.1.2) and the choice of non-linear activations (Section 3.1.3). We empirically show that these careful architecture design choices in ForceNet contribute to the significant performance improvement over the existing GNS model.

**Enforcing rotation-covariance.** Note also that the use of \( n_{st} \) in \( e_{st} \) results in the model being not necessarily rotation invariant nor covariant. In Section 3.2, we propose to encourage the physical constraint by training models with rotation data augmentation. In Section 4.4, we empirically demonstrate the effectiveness of this strategy in encouraging the rotation-covariance of ForceNet’s predictions.

### 3.1.2. Basis Functions

An important aspect of \( F_e \) is the choice of basis function \( B : \mathbb{R}^E \to \mathbb{R}^B \) that transforms the raw distance features \( e_{st} \) into ones that are more discriminative. Several choices of basis functions have been proposed, such as a Gaussian over 1D distances (Schütt et al., 2017a) and a spherical Bessel function over the joint 2D space of the edge distance and angle (Klicpera et al., 2020b). We extend these ideas to capture the full 3D positional differences between atoms, and systematically study the effectiveness of different basis functions in the context of a force-centric model.

Each of our basis functions \( B \) maps the raw edge features \( e_{st} \) presented in Section 3.1.1 into a \( B \) dimensional vector, where \( B \) varies based on the basis function used. \( B \) is typically much larger than \( E = 7 \) to aid in capturing subtle differences in atom positions.

**Identity:** \( B_{id}(x) = x \). The baseline is to use the edge features \( e_{st} \) directly.

**Linear + Act:** \( B_{linear}(x) = g(Wx + b) \), where \( g(\cdot) \) is the non-linear activation function, and \( W \) and \( b \) are the learnable parameters. When followed by the linear layer \( F_b \), this is equivalent to applying an 1-hidden-layer MLP over the edge features \( e_{st} \).

**Gaussian:** \( B_{gauss}(x) = [b_1, \ldots, b_J] \), where \( b_j \) is the output of the \( j \)-th basis function \( b_j(x) = \exp((x - \mu_j)^2/(2\sigma^2)) \). The Gaussian means are evenly distributed on the interval between 0 and 1, i.e., \( \mu_j = j/(J-1) \) and the standard deviation \( \sigma = 1/(J-1) \). All values of \( x \) are normalized to lie between 0 and 1. \( B_{gauss} \) is applied to each dimension of \( e_{st} \), resulting in a \( B = J \times E \) vector.

**Sine:** \( B_{sin}(x) = [b_1, \ldots, b_J] \), where \( b_j \) is the output of the \( j \)-th basis function \( b_j(x) = \sin(1.1^j x) \). The design is based on function approximation using the Fourier series. In our experiments, we find that using only the sinusoidal component of the Fourier series is sufficient. \( B_{sin} \) is applied to each dimension of \( e_{st} \), resulting in an \( B = J \times E \) vector.

### 3.1.3. Expressive Non-Linearity in MLPs

Our final key component is simple but crucial: the choice of non-linear activation function plays a central role in modeling complex non-linearities of atomic interactions. The ReLU activation (Glorot et al., 2011) is widely used in many deep learning models, including the existing GNS model (Bapst et al., 2020; Sanchez-Gonzalez et al., 2020). However, ReLU may not be ideal in modeling atomic forces, since it results in outputs being modeled as piece-wise linear hyper-planes with sharp boundaries. Ideally, we desire a smooth and expressive non-linear activation function.

We explored a wide array of choices for activation functions, such as Tanh, Leaky-ReLU, SoftPlus (Dugas et al., 2001), Shifted SoftPlus (Schütt et al., 2017a), and Swish (Ramachandran et al., 2017). In our experiments, we find Swish, i.e., \( act(x) = x \cdot \text{sigmoid}(x) \), to perform particularly well. As illustrated in Figure 4, Swish provides a smoother output landscape and has non-zero activation for negative inputs. As we demonstrate in Section 4.5.1 and Figure 4, the replacement of ReLU with Swish consistently and significantly improves the predictive accuracy while maintaining scalability across all choices of basis functions.

### 3.2. Rotation Data Augmentation

We apply random rotation data augmentation to encourage the rotation-covariance of the model’s predictions. Specifically, we randomly rotate the entire system and per-atom forces by the same degree, and let ForceNet predict the rotated forces based on the rotated system.

Rotation augmentation is particularly effective when the physical systems of interest have a canonical axis. Such systems are prevalent in many real applications. For instance, most of the material-type molecular systems have the canonical axis that is vertical to the material surface (Figure 2). At the macroscopic level, most of the physical systems have the natural canonical axis pointing towards the direction of gravity of the earth. Given such a canonical axis, rotation
4 Experiments

In this section, we evaluate ForceNet’s performance in predicting atomic forces. We do so by applying the model to OC20 (Chanussot et al., 2020), a massive dataset on quantum physics calculations on non-equilibrium atomic structures relevant to catalysis discovery.

Throughout this section, we normalize for computational time when comparing models’ predictive performance, i.e., we compare models with similar computational cost in training and inference. This is crucial because simply using more computational resources to train larger models is shown to lead better results in OC20 tasks (Chanussot et al., 2020). However, training time of most existing models is already more than 100 GPU days\(^1\) and even goes up to 1600 GPU days, making it harder to further scale up without improving the models’ computational efficiency. Moreover, fast model inference is crucial for the application of catalyst material discovery, where an ML model needs to make predictions over an enormous number of potential candidates (Zitnick et al., 2020).

4.1 Task Descriptions and Evaluation Metrics

OC20 dataset (Chanussot et al., 2020) contains 200M+ non-equilibrium 3D atomic structures from 1M+ atomic relaxation trajectories. Each structure is associated with the per-structure energy and per-atom forces. Figure 2 shows an illustration of 3D structures.

OC20 provides a variety of prediction tasks relevant to catalyst discovery for renewable energy applications. Our main focus is on the atomic force prediction task, called S2F (Structure to Forces). Following the baseline setting in S2F (Chanussot et al., 2020), we train our models on the 130M training structures that are on relaxation simulation trajectories. In this work, we focus on the S2F task. In Appendix E, we also provide preliminary results on a simulation task by directly applying our S2F models.

We evaluate models on four validation datasets that test different levels of model generalization: In Domain (ID), Out of Domain Adsorbate (OOD Adsorbate), OOD Catalyst, and OOD Both (both the adsorbate and catalyst’s material are not seen in training). Each split contains 1M examples.

Following Chanussot et al. (2020), the Mean Absolute Error (MAE) of forces on free atoms is evaluated for each validation set. Here the free atoms represent atoms that are close to the material surface and are free to move during atomic relaxation simulation (Figure 5 in Appendix A). We use the “average force MAE” to represent the MAE of forces averaged over the four validation sets.

4.2 Model Settings and Computational Time

Below, we describe hyper-parameter settings of ForceNet and baseline models, along with the computational time (in GPU days) to train these models. Table 1 shows the summary of training time, inference time, and sizes of different models. All the training is run under Tesla V100 Volta. The inference time is measured on the validation ID set under GeForce RTX 2080, where the largest possible batch size is used for each model.

**ForceNet.** We use the spherical function and Swish activation as the default basis and activation functions, since this combination consistently provides the best results (Figure 4). The default model size has 5-layers of message passing and 512-dimensional hidden channels, and the training batch size is set to 256. We also consider a larger variant, ForceNet-large, that uses 7-layer message passing, 768-dimensional hidden channels, and the batch size of 512. Training ForceNet and ForceNet-large takes 31 and 194 GPU days, respectively.

During training, we apply the data-efficient rotation augmentation strategy presented in Section 3.2. We also find it useful to train on both free and fixed atoms, even though the evaluation is only on the free atoms. Specifically, we give a small relative weight of 0.05 to the loss of fixed atoms during training, which is ablated in Table 7 of Appendix D. Further implementation details and hyper-parameters are provided in Appendix C.

**Baseline models.** We compare ForceNet against the following three strong baseline GNN models.

- **SchNet** (Schütt et al., 2017a) is a energy-centric GNN that uses scalable atom-pair-based message passing; hence, a relatively large model size (5-layer message passing with 1024-dimensional hidden channels) can be trained with 194 GPU hours, which is comparable to ForceNet-large.

- **DimeNet++** (Klicpera et al., 2020a) is a recent improvement of DimeNet (Klicpera et al., 2020b) and is also an energy-centric GNN. It uses atom-triplet-based message passing to capture angular information, which makes it computationally expensive. Even training DimeNet++ of a relatively-small model size (3-layer message passing...
Table 1. Comparison of ForceNet to existing GNN models. We mark as bold the best performance and close ones, i.e., within 0.0005 MAE, which according to our preliminary experiments, is a good threshold to meaningfully distinguish model performance. Training time is in GPU days, and inference time is in GPU hours. Median represents the trivial baseline of always predicting the median training force across all the validation atoms.

| Model          | Hidden dim | #Msg layers | #Params | Train time | Inference time | ID MAE | OOD Ads | OOD Cat. | OOD Both | Average |
|----------------|------------|-------------|---------|------------|----------------|--------|---------|----------|----------|---------|
| Median         | –          | –           | –       | –          | 0.0810         | 0.0799 | 0.0799  | 0.0943   | 0.0838   |
| GNS            | 768        | 5           | 12.5M   | 20d        | 1.5h           | 0.0421 | 0.0466  | 0.0430   | 0.0559   | 0.0469  |
| SchNet         | 1024       | 5           | 9.1M    | 194d       | 0.8h           | 0.0443 | 0.0514  | 0.0465   | 0.0618   | 0.0510  |
| DimeNet++      | 192        | 3           | 1.8M    | 587d       | 8.5h           | 0.0332 | 0.0366  | 0.0344   | 0.0436   | 0.0369  |
| DimeNet++-large| 512        | 3           | 10.7M   | 1600d      | 27.0h          | 0.0281 | 0.0318  | 0.0315   | 0.0396   | 0.0328  |
| ForceNet       | 512        | 5           | 11.3M   | 31d        | 1.3h           | 0.0313 | 0.0355  | 0.0334   | 0.0439   | 0.0360  |
| ForceNet-large | 768        | 7           | 34.8M   | 194d       | 3.5h           | 0.0281 | 0.0320  | 0.0327   | 0.0412   | 0.0335  |

Table 2. Analysis of how training data and rotation augmentation affect the stability of ForceNet’s prediction against rotation. 1000 validation ID structures are sampled and randomly rotated 100 times along the vertical axis. For each rotated structure, ForceNet predicts per-atom forces that are then rotated back to compare with the originally-predicted forces. Instability is measured by the average standard deviation of the errors across the 100 rotations for each (free) atom. Smaller instability values indicate the model is closer to being rotation-covariant, where a fully rotation-covariant model would always return a value of 0 regardless of its force MAE.

| Dataset | Rotation aug. | Average instability of per-atom force pred. | Val Force MAE ID Average |
|---------|----------------|---------------------------------------------|--------------------------|
| All (130M) | ✓             | 0.0037                                     | 0.0313                   | 0.0360       |
| All (130M) |               | 0.0069                                     | 0.0314                   | 0.0366       |
| 2M       | ✓             | 0.0041                                     | 0.0332                   | 0.0382       |
| 2M       |                | 0.0093                                     | 0.0346                   | 0.0400       |

All the results of SchNet and DimeNet++ are directly adopted from the OC20 paper (Chanussot et al., 2020). These energy-centric models are trained only on atomic forces, although in principle, they can also be trained on per-system energy. Chanussot et al. (2020) report that training on forces and energy separately achieves better performance on each task compared to joint training. For the GNS model, we reproduce the original model architecture ourselves based on the feedback from the original author of GNS (Sanchez-Gonzalez et al., 2020). Refer to Appendix B for implementation details. On the GNS model, we apply the same training strategies as ForceNet.

4.3. S2F Performance Comparison

We consider the S2F task (atomic force prediction) and compare the performance of ForceNet against the baseline models, while normalizing for the computational time. Main results are plotted in Figure 1, and complete results can be found in Table 1.

From Figure 1 (left), we see that ForceNet gives superior force prediction performance given limited training GPU budgets. Compared with SchNet, ForceNet converges to a better performance with about 6.3 times less compute time. Compared to DimeNet++ (resp. -large), ForceNet (resp. -large) converges to the comparable performance with 18.9 (resp. 8.2) times less training compute. Compared to the GNS model, ForceNet achieves much better performance across all training times although ForceNet takes slightly longer to converge.

In Figure 1 (right), we see that ForceNet achieves prediction performance comparable to DimeNet++, while enabling much faster inference speed. Specifically, compared to DimeNet++ (resp. DimeNet++-large), ForceNet++ (resp. ForceNet-large) is 6.5 (resp. 7.7) times faster, with comparable prediction performance.

Finally, when ForceNet-large is compared against DimeNet++, ForceNet-large reduces the force prediction error by almost 10% on average, while being 3.0 times faster in training and 2.4 times faster in inference.

4.4. Analysis of ForceNet’s Rotation-covariance

We analyze whether ForceNet is able to learn rotation-covariance when predicting atomic forces. We do so by measuring the prediction instability of ForceNet when validation systems are rotated.
We systematically study how choices of different basis and activation functions affect the model performance. We also compare with the “None” baseline, which does not use a basis function and directly concatenates the input raw edge feature into the node embeddings. In Figure 4, we see that the combination of spherical basis and Swish activation performs the best. For comparison, the GNS model uses no basis function (“None”) and ReLU, see Appendix B for full GNS model details.

### 4.5. Ablation on ForceNet’s model designs

Here we perform extensive ablation studies on ForceNet’s key design choices presented in Section 3.

#### 4.5.1. Basis and activation functions

We systematically study how choices of different basis and non-linear activation functions affect the model performance. We also compare with the “None” baseline, which does not use a basis function and directly concatenates the input raw edge feature into the node embeddings. In Figure 4, we see that the combination of spherical basis and Swish activation performs the best. For comparison, the GNS model uses no basis function (“None”) and ReLU, see Appendix B for full GNS model details.

#### 4.5.2. Architecture design

Next, we study the architectural building blocks of our conditional-filter-based message passing with the fixed basis and activation functions. We consider seven cases: (1) **Only-dist**: we remove $n_{st}$ from the input edge feature, i.e., $e_{st} = p_{st}$, resulting in the edge features being rotation invariant. (2) **No-atomic-radii**: we set the input edge features to $e_{st} = \text{Concat}(n_{st}, ||d_{st}||)$ (atomic radii information is dropped), (3) **No-node-emb**: filter $F_c$ is a function of only $e_{st}$ (conditioning on source and target node embeddings $n_{s}^{(k)}, h_{t}^{(k)}$ is dropped). (4) **Only-$F_c$**: Filter is directly aggregated, i.e., $m_{st} = F_c$, and self-message $m_t$ is omitted. (5) **Edge-linear-BN**: MLP $F_c$ is replaced with a linear function followed by batch normalization, (6) **Node-linear-BN**: MLP $F_c$ is replaced with a linear function followed by batch normalization. (7) **No-$m_t$**: self-message $m_t$ is removed. Note that in (5) and especially (6), we find it critical to add the batch normalization to facilitate training.

Table 3 shows the results of the seven ablation studies. Most notably, (1) is significantly worse than the rest, because rotation-invariant node embeddings are insufficient for predicting rotation-covariant forces. We also see from (2) and (3) that making the filter less expressive, especially by dropping the dependency on node embeddings, significantly hurts performance. The improvement from element-wise product parameterization $F_c \odot F_d$ is demonstrated in (4). From (5), we see that it is critical to utilize non-linear models for edge features, as atomic forces are highly dependent on their subtle changes, but non-linearities are not essential for node embeddings (6). Finally, from (7), we see that the self-message $m_t$ is not essential in performance.

Overall, our analysis suggests that ForceNet benefits most from its expressive edge-level computation via the conditional filter, which is directly responsible for accurately encoding the 3D neighborhood structure on which the atomic forces depend.

#### 4.5.3. Model scaling

Comparing ForceNet and ForceNet-large in Table 1, we see that a larger model provides significant performance gain, at the cost of 6.3 times more training time and 2.4 times more inference time. Extrapolating through Figure 1, we expect ForceNet to significantly outperform DimeNet++, once comparable computational resources are used. We leave this investigation to future work. More fine-grained ablations on model scaling are shown in Table 4. We see that all the three scaling components help in the current regime of ForceNet.

### 5. Conclusions

In this work, we demonstrate that force-centric GNN models without any explicit physical constraints are able to predict atomic forces more accurately than state-of-the-art energy-centric GNN models, while being faster both in training and inference. We achieve this by carefully designing the message passing architecture, and by training the models on...
massive data and applying physics-based data augmentation.

This work opens up numerous avenues for future research: (1) Apply the same principle to other prediction tasks in OC20 (e.g., predicting per-system energy) and other application domains. (2) Incorporate physics knowledge into ForceNet to increase its generalization performance. (3) Improve computational efficiency of ForceNet, while maintaining its performance, similar to how DimeNet++ has been significantly improved over DimeNet (Klicpera et al., 2020a). (4) Scale up ForceNet with more computational resources.

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References

The atomic simulation environment—a python library for working with atoms. Journal of Physics Condensed Matter, 2017. ISSN 1361648X. doi: 10.1088/1361-648X/a680e.

Ba, J. L., Kiros, J. R., and Hinton, G. E. Layer normalization. arXiv preprint arXiv:1607.06450, 2016.

Bapst, V., Keck, T., Grabska-Barwińska, A., Donner, C., Cubuk, E. D., Schoenholz, S., Obika, A., Nelson, A., Back, T., Hassabis, D., et al. Unveiling the predictive power of static structure in glassy systems. Nature Physics, 16(4):448–454, 2020.

Battaglia, P., Pascanu, R., Lai, M., Rezende, D. J., et al. Interaction networks for learning about objects, relations and physics. In Advances in Neural Information Processing Systems (NeurIPS), pp. 4502–4510, 2016.

Behler, J. Perspective: Machine learning potentials for atomistic simulations. The Journal of chemical physics, 145(17):170901, 2016.

Brockschmidt, M. Gnn-film: Graph neural networks with feature-wise linear modulation. In International Conference on Machine Learning (ICML), pp. 2014–2023, 2020.

Chanussot, L., Das, A., Goyal, S., Lavril, T., Shuaibi, M., Riviere, M., Tran, K., Heras-Domingo, J., Ho, C., Hu, W., Palizhati, A., Sriram, A., Wood, B., Yoon, J., Parikh, D., Zitnick, C. L., and Ulissi, Z. The Open Catalyst 2020 (oc20) dataset and community challenges. arXiv preprint arXiv:2010.09990, 2020.

Chen, G., Chen, P., Hsieh, C.-Y., Lee, C.-K., Liao, B., Liao, R., Liu, W., Qiu, J., Sun, Q., Tang, J., et al. Alchemy: A quantum chemistry dataset for benchmarking ai models. arXiv preprint arXiv:1906.09427, 2019.

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.

Chmiela, S., Sauceda, H. E., Müller, K.-R., and Tkatchenko, A. Towards exact molecular dynamics simulations with machine-learned force fields. Nature communications, 9 (1):1–10, 2018.

Christensen, A. S. and von Lilienfeld, O. A. On the role of gradients for machine learning of molecular energies and forces. Machine Learning: Science and Technology, 1(4): 045018, 2020.

del Río, E. G., Mortensen, J. J., and Jacobsen, K. W. Local bayesian optimizer for atomic structures. Physical Review B, 100(10):104103, 2019.

Dugas, C., Bengio, Y., Bélisle, F., Nadeau, C., and Garcia, R. Incorporating second-order functional knowledge for better option pricing. Advances in neural information processing systems, pp. 472–478, 2001.

Fey, M. and Lenssen, J. E. Fast graph representation learning with pytorch geometric. arXiv preprint arXiv:1903.02428, 2019.

Frederiksen, T., Paulsson, M., Brandbyge, M., and Jauho, A.-P. Inelastic transport theory from first principles: Methodology and application to nanoscale devices. Physical Review B, 75(20):205413, 2007.

Gibbs, G., Hill, F., Boisen, M., and Downs, R. Power law relationships between bond length, bond strength and electron density distributions. Physics and Chemistry of Minerals, 25(8):585–590, 1998.

Gilmer, J., Schoenholz, S. S., Riley, P. F., Vinyals, O., and Dahl, G. E. Neural message passing for quantum chemistry. In International Conference on Machine Learning (ICML), pp. 1273–1272, 2017.
Glorot, X. and Bengio, Y. Understanding the difficulty of training deep feedforward neural networks. In *International Conference on Artificial Intelligence and Statistics (AISTATS)*, pp. 249–256, 2010.

Glorot, X., Bordes, A., and Bengio, Y. Deep sparse rectifier neural networks. In *International Conference on Artificial Intelligence and Statistics (AISTATS)*, pp. 315–323, 2011.

Hamilton, W. L., Ying, R., and Leskovec, J. Inductive representation learning on large graphs. In *Advances in Neural Information Processing Systems (NeurIPS)*, pp. 1025–1035, 2017.

He, K., Zhang, X., Ren, S., and Sun, J. Deep residual learning for image recognition. In *IEEE Conference on Computer Vision and Pattern Recognition (CVPR)*, pp. 770–778, 2016.

Henkelman, G. and Jónsson, H. Improved tangent estimate in the nudged elastic band method for finding minimum energy paths and saddle points. *The Journal of chemical physics*, 113(22):9978–9985, 2000.

Henkelman, G., Uberuaga, B. P., and Jónsson, H. A climbing image nudged elastic band method for finding saddle points and minimum energy paths. *The Journal of chemical physics*, 113(22):9901–9904, 2000.

Ioffe, S. and Szegedy, C. Batch normalization: Accelerating deep network training by reducing internal covariate shift. In *International Conference on Machine Learning (ICML)*, pp. 448–456, 2015.

Jouy, M., Luc, W., and Jiao, F. High-rate electroreduction of carbon monoxide to multi-carbon products. *Nature Catalysis*, 2018. ISSN 25201158. doi: 10.1038/s41929-018-0133-2.

Khorshidi, A. and Peterson, A. A. Amp: A modular approach to machine learning in atomistic simulations. *Computer Physics Communications*, 207:310–324, 2016.

Kingma, D. P. and Ba, J. Adam: A method for stochastic optimization. In *International Conference on Learning Representations (ICLR)*, 2015.

Kipf, T., Fetaya, E., Wang, K.-C., Welling, M., and Zemel, R. Neural relational inference for interacting systems. In *International Conference on Machine Learning (ICML)*, 2018.

Kipf, T. N. and Welling, M. Semi-supervised classification with graph convolutional networks. In *International Conference on Learning Representations (ICLR)*, 2017.

Klicpera, J., Giri, S., Margraf, J. T., and Günneumann, S. Fast and uncertainty-aware directional message passing for non-equilibrium molecules. In *NeurIPS-W*, 2020a.

Klicpera, J., Groß, J., and Günneumann, S. Directional message passing for molecular graphs. In *International Conference on Learning Representations (ICLR)*, 2020b.

Kresse, G. and Furthmüller, J. Efficiency of ab-initio total energy calculations for metals and semiconductors using a plane-wave basis set. *Computational Materials Science*, 6(1):15–50, 1996a. ISSN 09270256. doi: 10.1016/0927-0256(96)00008-0.

Kresse, G. and Furthmüller, J. Efficient iterative schemes for ab initio total-energy calculations using a plane-wave basis set. *Physical Review B*, 54(16):11169–11186, 1996b. ISSN 0163-1829. doi: 10.1103/PhysRevB.54.11169.

Kresse, G. and Hafner, J. Ab initio molecular-dynamics simulation of the liquid-metal–amorphous-semiconductor transition in germanium. *Physical Review B*, 49(20):14251–14269, 1994. ISSN 0163-1829. doi: 10.1103/PhysRevB.49.14251.

Leach, A. R. *Molecular modelling: principles and applications*. Pearson education, 2001.

MacRobert, T. M. *Spherical harmonics: an elementary treatise on harmonic functions with applications*. 1947.

Nakata, M., Shimazaki, T., Hashimoto, M., and Maeda, T. Pubchemqc pm6: Data sets of 221 million molecules with optimized molecular geometries and electronic properties. *Journal of Chemical Information and Modeling*, 2019.

Park, C. W., Kornbluth, M., Vandermuse, J., Wolverton, C., Kozinsky, B., and Mailoa, J. P. Accurate and scalable multi-element graph neural network force field and molecular dynamics with direct force architecture. *arXiv preprint arXiv:2007.14444*, 2020.

Parr, R. G. *Density functional theory of atoms and molecules*. In *Horizons of quantum chemistry*, pp. 5–15. Springer, 1980.

Paszke, A., Gross, S., Massa, F., Lerer, A., Bradbury, J., Chanan, G., Killeen, T., Lin, Z., Gimelshein, N., Antiga, L., et al. PyTorch: An imperative style, high-performance deep learning library. In *Advances in Neural Information Processing Systems (NeurIPS)*, pp. 8024–8035, 2019.

Ramachandran, P., Zoph, B., and Le, Q. V. Swish: a self-gated activation function. *arXiv preprint arXiv:1710.05941*, 7, 2017.

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.

Sanchez-Gonzalez, A., Godwin, J., Pfaff, T., Ying, R., Leskovec, J., and Battaglia, P. W. Learning to simulate complex physics with graph networks. In *International Conference on Machine Learning (ICML)*, 2020.
Schütt, K., Kindermans, P.-J., Felix, H. E. S., Chmiela, S., Tkatchenko, A., and Müller, K.-R. Schnet: A continuous-filter convolutional neural network for modeling quantum interactions. In *Advances in Neural Information Processing Systems (NeurIPS)*, pp. 991–1001, 2017a.

Schütt, K. T., Arbabzadah, F., Chmiela, S., Müller, K. R., and Tkatchenko, A. Quantum-chemical insights from deep tensor neural networks. *Nature communications*, 8: 13890, 2017b.

Seh, Z. W., Kibsgaard, J., Dickens, C. F., Chorkendorff, I., Nørskov, J. K., and Jaramillo, T. F. Combining theory and experiment in electrocatalysis: Insights into materials design. *Science*, 355(6321), 2017. ISSN 0036-8075. doi: 10.1126/science.aad4998. URL https://science.sciencemag.org/content/355/6321/eaad4998.

Slater, J. C. Atomic radii in crystals. *The Journal of Chemical Physics*, 41(10):3199–3204, 1964.

Velickovic, P., Cucurull, G., Casanova, A., Romero, A., Lio, P., and Bengio, Y. Graph attention networks. In *International Conference on Learning Representations (ICLR)*, 2018.

Xie, T. and Grossman, J. C. Crystal graph convolutional neural networks for an accurate and interpretable prediction of material properties. *Physical review letters*, 120 (14):145301, 2018.

Xu, K., Hu, W., Leskovec, J., and Jegelka, S. How powerful are graph neural networks? In *International Conference on Learning Representations (ICLR)*, 2019.

Zitnick, C. L., Chanussot, L., Das, A., Goyal, S., Heras-Domingo, J., Ho, C., Hu, W., Lavril, T., Palizhati, A., Riviere, M., Shuaibi, M., Sriram, A., Tran, K., Wood, B., Yoon, J., Parikh, D., and Ulissi, Z. An introduction to electrocatalyst design using machine learning for renewable energy storage. *arXiv preprint arXiv:2010.09435*, 2020.
Supplementary Material

A. Description of OC20 dataset

The OC20 dataset (Chanussot et al., 2020) contains over 130M non-equilibrium structures for training for the S2F task (i.e., atomic force prediction). The structures come from over 650K relaxation trajectories—the movement of the atoms from the initial structure to relaxed structure (equilibrium 3D structures with all-zero atomic forces).

Each structure contains the 3D positions of atoms in an adsorbate and catalyst slab, Figure 5. The adsorbate is a molecule involved in the chemical reaction that interacts with the catalyst’s surface. The adsorbate contains 1 to 11 atoms. The catalyst is represented as a “slab” that repeats infinitely in the $x$ and $y$ directions. The slab structure is repeated in a grid pattern where each repetition is referred to as a “cell”. The center cell has coordinate $(0, 0, 0)$ with the cell to the left right being $(-1, 0, 0)$ and $(1, 0, 0)$ respectively. The slab is not repeated in the $z$ direction. Instead, the atoms at the bottom of the slab are assumed to be fixed and not move during a relaxation, which approximates how they would be held in place by the catalyst’s atoms below the slab. Typically, only the top two layers of the catalyst’s surface are assumed to be free and are moved according to their forces during a relaxation (see Figure 5). Therefore, forces are only evaluated on free catalyst atoms and the adsorbate.

The forces on the same atom in different cells are identical, since their atom neighbors are identical, resulting in their GNN’s node embeddings to be also identical, e.g., the node embedding of atoms marked $s$ and $s'$ in Figure 5 are the same. When computing the neighborhood of an atom, atoms in neighboring cells need to be also taken into consideration (atom $t$ in Figure 5). Notice that the message from $s$ to $t$ and the message from $s'$ to $t$ are different since the the relative placements of the two atoms are different, resulting in different edge features $e_{st}$ and $e'_{st}$. Computing the edge features between atoms from different cells can be done using the supplied information in the OC20 dataset for periodic boundary conditions.

B. Details of GNS Model

For the GNS results in this paper, we reimplemented the original GNS model (Sanchez-Gonzalez et al., 2020). Since the public code for the original GNS model (Sanchez-Gonzalez et al., 2020) was not available at the time of our experiments, we communicated with one of the authors to confirm the implementation details.

The message in the GNS model is defined as

$$m(h_t^{(l)}, e_{st}, h_s^{(l)}) = \text{MLP} \left( \text{Concat} \left( h_t^{(l)}, e_{st}, h_s^{(l)} \right) \right),$$

where $\text{MLP}(\cdot)$ is a 1-hidden-layer MLP with ReLU activation and layer normalization (Ba et al., 2016) applied before the activation. For aggregating the message, the GNS model used either mean or sum, so we tried both in our experiments. We found sum aggregation to perform better, and report results of sum aggregation in this paper. After the messages are aggregated, GNS uses a learnable linear function to transform the node embeddings. Similar to ForceNet, we additionally apply a batch normalization on the node embeddings, which alleviates training instability and significantly improves performance. The GNS model uses a residual connection, where the computed node embeddings are added into the node embeddings from the previous layer. For the decoder, the GNS model uses a 1-hidden-layer MLP with ReLU activation. All the node embeddings and hidden units in the MLPs have the same dimensionality.

C. Hyper-parameters

For training, we use the Adam optimizer (Kingma & Ba, 2015), with an initial learning rate of 0.0005. We train ForceNet and the GNS model for 500K iterations with the batch size of 256, which is equivalent to 1 epoch for the entire dataset. For ForceNet-large, we use the batch size of 512. All the parameters of the force-centric models are initialized with Xavier uniform initialization (Glorot & Bengio, 2010). The learning rate is kept constant for the first 250K iterations, after which it is halved every 50K iterations. We use the checkpoint with the best validation ID performance, and evaluate the saved model over all four validation sets. MAE over forces is used as the training loss. We will evaluate our models on the hidden test sets once the test server is ready.

For Gaussian and sine basis functions, we use $J = 50$, which gives an output dimensionality of $B = 350$. For Linear+Act, we set $B = 350$. For spherical basis, we use $L = 3$ and $S = 4$, which results in $B = 36 (= 3^2 \cdot 4)$, and we set $J = 50$ for the internally-used sine basis function. For encoding the input atomic node features, we first normalize each dimension to lie between 0 and 1, and adopt the same basis function as used for encoding the edge features. The exception is spherical basis that is specialized for 3D spaces, in which case, the sine basis is used to encode the input atomic node features. We find that increasing $J$ and $L$ beyond the above values does not improve the performance, while significantly decreasing them worsens the performance.
Figure 5. 2D Illustration of a slab that represents a catalyst’s surface and an adsorbate. The slab is tiled in the $x$ and $y$ directions to create the surface (neighboring cells shown as atoms with dashed outlines). Only the cells to the left ($[-1, 0, 0]$) and right ($[1, 0, 0]$) are shown. The adsorbate is also assumed to be tiled with the slab (white, red, and grey atoms). Only the top 2 layers of the slab are allowed to move during a relaxation (dark blue), and the others are fixed (light blue). Neighboring atoms (black arrows) can be from the same cell or neighboring cells ($t$ and $s'$). All atoms within a radius (dotted circle) are assumed to be neighbors.

Table 5. Ablations on basis and activation functions in the ForceNet architecture.

| Basis  | Act.  | ID         | OOD Ads. | OOD Cat. | OOD Both | Average |
|--------|-------|------------|----------|----------|----------|---------|
| Spherical | ReLU  | 0.0324     | 0.0367   | 0.0346   | 0.0454   | 0.0373  |
| Spherical | Swish | 0.0313     | 0.0355   | 0.0334   | 0.0439   | 0.0360  |
| Sine   | ReLU  | 0.0324     | 0.0367   | 0.0346   | 0.0456   | 0.0374  |
| Sine   | Swish | 0.0317     | 0.0360   | 0.0342   | 0.0448   | 0.0367  |
| Gauss  | ReLU  | 0.0335     | 0.0384   | 0.0359   | 0.0476   | 0.0389  |
| Gauss  | Swish | 0.0318     | 0.0364   | 0.0346   | 0.0456   | 0.0371  |
| Linear+Act | ReLU | 0.0340     | 0.0379   | 0.0356   | 0.0464   | 0.0385  |
| Linear+Act | Swish | 0.0321     | 0.0359   | 0.0342   | 0.0445   | 0.0367  |
| Identity | ReLU  | 0.0335     | 0.0377   | 0.0353   | 0.0464   | 0.0382  |
| Identity | Swish | 0.0322     | 0.0364   | 0.0358   | 0.0445   | 0.0368  |
| None   | ReLU  | 0.0379     | 0.0430   | 0.0391   | 0.0519   | 0.0430  |
| None   | Swish | 0.0330     | 0.0383   | 0.0347   | 0.0466   | 0.0382  |

Table 6. Ablations on the message passing architecture of ForceNet.

| Ablation       | ID         | OOD Ads. | OOD Cat. | OOD Both | Average |
|----------------|------------|----------|----------|----------|---------|
| ForceNet       | 0.0313     | 0.0355   | 0.0334   | 0.0439   | 0.0360  |
| (1) Only-dist  | 0.0659     | 0.0673   | 0.0660   | 0.0805   | 0.0699  |
| (2) No-atomic-radii | 0.0321 | 0.0362   | 0.0342   | 0.0447   | 0.0368  |
| (3) No-node-emb | 0.0361     | 0.0409   | 0.0374   | 0.0495   | 0.0410  |
| (4) Only-$F$  | 0.0333     | 0.0372   | 0.0350   | 0.0455   | 0.0378  |
| (5) Edge-linear-BN | 0.0371 | 0.0430   | 0.0388   | 0.0520   | 0.0427  |
| (6) Node-linear-BN | 0.0317 | 0.0356   | 0.0339   | 0.0442   | 0.0364  |
| (7) No-$m_t$  | 0.0314     | 0.0360   | 0.0336   | 0.0444   | 0.0364  |

D. Full Ablation Results

Here we provide full S2F results of our ablation studies, reporting the force MAE on each of the four validation sets.

Full Results on ForceNet Designs. First, we provide the full ablation results on ForceNet designs. Table 5 shows the ablations on basis and activation functions, while Table 6 shows the ablations on the message passing architectures.

Overall, we see trends that are consistent with the averaged results in Figure 4 and Table 3. Specifically, from Table 5, we see that the combination of spherical basis functions and the Swish activation results in the best performance across the four validation sets. From Table 6, we see that the conditional filter convolution design gives superior performance compared to the more simplified architectures, except for (6) and (7), in which the performance is comparable.

Full Results on Training Strategies. Next, we provide the full ablation results of our training strategies, fixing the model architecture to the default ForceNet.

The results are shown in Table 7. First, we see that rotation augmentation helps, especially for the three out-of-distribution validation sets. Second, we see that providing small reweighted supervision on fixed atoms is also helpful, significantly improving the validation performance (evaluated on free atoms) compared to the two baseline strategies: (1) uniform loss weighting (equally weighting the losses for fixed and free atoms) and (2) zero-loss weighting (ignoring losses on fixed atoms during training).

\(^2^\)We do not observe much gain by training models longer than 1 epoch. This is probably because of the redundancy in data, i.e., out of 130M data points, there are 650k unique atom configurations (ignoring the positional differences).
Table 7. Ablations on training strategies for ForceNet.

| Model       | Rotation avg. | Weight on fixed atoms | ID | OOD Ads. | OOD Adv. | OOD Cat. | OOD Both. | Average |
|-------------|---------------|-----------------------|----|----------|---------|---------|-----------|---------|
| ForceNet    | ✓ 0.05        | 0.0113                | 0.0355 | 0.0334 | 0.0339 | 0.0360 |
| ForceNet    | ✓ 0.05        | 0.0214                | 0.0359 | 0.0341 | 0.0448 | 0.0366 |
| ForceNet    | ✓ 1           | 0.0669                | 0.0411 | 0.0390 | 0.0506 | 0.0419 |
| ForceNet    | ✓ 0           | 0.0333                | 0.0385 | 0.0348 | 0.0465 | 0.0383 |

E. Structure Relaxation Simulation Results

Here we apply ForceNet to the IS2RS (Initial Structure to Relaxed Structure) task. The goal is to predict the relaxed structure, i.e., 3D structure with zero-forces on all free atoms, from the initial structure. This can be achieved by simulating a relaxation trajectory: iteratively updating the atomic positions of free atoms according to their predicted forces until convergence, i.e., the predicted forces are below a pre-specified threshold.

Specifically, the structure relaxations are performed using a PyTorch implementation of the Atomic Simulation Environment’s (ASE) (Hjo, 2017) L-BFGS optimizer. Relaxations were terminated when a max-absolute per-atom force of 0.01 eV/Å or 200 simulation steps, whichever comes first. All DFT calculations were performed in the Vienna Ab Initio Simulation Package (VASP) (Kresse & Furthmüller, 1996a;b; Kresse & Hafner, 1994). Both ASE and VASP are popular packages within the computational chemistry and catalysis communities.

The performance on the IS2RS task is evaluated by the two standard metrics (Chanussot et al., 2020): (1) Average Force below Threshold (AFbT), measuring whether the predicted relaxed structure actually has small forces calculated by ground-truth DFT, and (2) Average Distance within Threshold (ADwT), measuring the geometrical closeness between the predicted relaxed structure and ground-truth relaxed structure. For both metrics, the higher, the better.

Figure 6 compares the performance of different models, while taking the inference efficiency into account. The full results for all the validation sets are provided in Table 8. Here the inference time is measured on a Tesla V100 Volta GPU, where we use the largest possible batch size for each model and perform 100K relaxations. All the models are the same as Figure 1 and Table 1, originally trained for the S2F task.

We see from Figure 6 (left) that in terms of AFbT, both ForceNet models outperform GNS and SchNet, while the inference time of ForceNet, GNS and SchNet is comparable to each other. Compared to DimeNet++, both ForceNet and ForceNet-large have lower AFbT. However, the inference of both ForceNet models is much faster than that of DimeNet++ (5.4 times faster for ForceNet, and 2.2 times faster for ForceNet-large). Moreover, we see that there is still a room for ForceNet-large to be further scaled up to give AFbT comparable to DimeNet++. Regarding ADwT, from Figure 6 (right), we see that ForceNet-large outperforms DimeNet++, while being 2.2 times faster in inference. ForceNet-large is slightly worse than DimeNet++, but is 5.4 times faster in inference.

Overall, the above results are encouraging given the faster inference time of ForceNet compared to DimeNet++. However, the results also suggest a potential limitation of ForceNet’s force-centric approach: the superior performance of ForceNet-large over DimeNet++ in the S2F task (i.e., estimate the forces of 3D structures along the simulation trajectory) does not directly translate into its superior performance on the IS2RS simulation task. We deduce this is due to the compounding error problem of the force-centric approach pointed out by the GNS work (Sanchez-Gonzalez et al., 2020), i.e., model’s prediction errors accumulate along the simulation trajectory, which forces the model to make increasingly erroneous prediction over structures that are far away from the simulation trajectory. The energy-centric models may suffer less from the problem since their built-in physical constraints allow them to make more well-behaved force prediction over the off-trajectory structures, which eventually leads to better simulation results despite the worse force prediction results.

Fortunately, OC20 additionally provides 94M off-trajectory structures obtained by either perturbing the on-trajectory structures or performing molecular dynamics from relaxed structures (Chanussot et al., 2020). We believe that these structures can be used to mitigate the compounding error problem of the force-centric approach by robustifying its off-trajectory force prediction. In fact, the original GNS work (Sanchez-Gonzalez et al., 2020) has demonstrated that training their force-centric models on perturbed off-trajectory structures significantly reduces the compounding error, thereby improving their simulation results. We leave this investigation to future work.
Figure 6. Comparison of IS2RS performance in terms of AFbT and ADwT averaged over the four validation sets. The $x$-axis is the IS2RS inference time in GPU hours measured over 100K relaxations.

Table 8. Full IS2RS results. Inference time is in GPU hours and measured over 100K relaxations.

| Model     | Inference time | ID  | OOD Ads. | OOD Cat. | OOD Both | Average | ID  | OOD Ads. | OOD Cat. | OOD Both | Average |
|-----------|----------------|-----|----------|----------|----------|---------|-----|----------|----------|----------|---------|
| GNS       | 74.3h          | 2.22| 0.66     | 1.44     | 0.62     | 1.24    | 30.60| 23.13    | 30.92    | 31.15    | 28.95   |
| SchNet    | 54.1h          | 4.90| 2.66     | 2.75     | 2.90     | 3.30    | 35.54| 29.80    | 26.86    | 28.39    | 30.15   |
| DimeNet++ | 407.6h         | 17.41| 14.41    | 14.19    | 14.55    | 15.14   | 48.75| 45.19    | 48.59    | 53.14    | 48.92   |
| DimeNet++-large | 814.6h | 24.22| 20.40    | 20.13    | 20.31    | 21.27   | 52.45| 48.47    | 50.98    | 54.82    | 51.68   |
| ForceNet  | 75.1h          | 10.75| 7.74     | 7.54     | 7.78     | 8.45    | 46.83| 41.26    | 46.45    | 49.60    | 46.04   |
| ForceNet-large | 186.9h | 14.77| 12.23    | 12.16    | 11.46    | 12.66   | 50.59| 45.16    | 49.80    | 52.94    | 49.62   |