TOWARDS TRAINING BILLION PARAMETER GRAPH NEURAL NETWORKS FOR ATOMIC SIMULATIONS

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ABSTRACT

Recent progress in Graph Neural Networks (GNNs) for modeling atomic simulations has the potential to revolutionize catalyst discovery, which is a key step in making progress towards the energy breakthroughs needed to combat climate change. However, the GNNs that have proven most effective for this task are memory intensive as they model higher-order interactions in the graphs such as those between triplets or quadruplets of atoms, making it challenging to scale these models. In this paper, we introduce *Graph Parallelism*, a method to distribute input graphs across multiple GPUs, enabling us to train very large GNNs with hundreds of millions or billions of parameters. We empirically evaluate our method by scaling up the recently proposed DimeNet++ and GemNet models by over an order of magnitude in the number of parameters. On the large-scale Open Catalyst 2020 (OC20) dataset, these graph-parallelized models lead to relative improvements of 1) 15% on the force MAE metric on the S2EF task and 2) 21% on the AFbT metric on the IS2RS task, establishing new state-of-the-art results.

1 INTRODUCTION

Graph Neural Networks (GNNs) (Gori et al., 2005; Zhou et al., 2020) have emerged as the standard architecture of choice for modeling atomic systems, with a wide range of applications from protein structure prediction to catalyst discovery and drug design (Schütt et al., 2017b; Gilmer et al., 2017; Jørgensen et al., 2018; Schütt et al., 2017a; 2018; Xie & Grossman, 2018; Qiao et al., 2020; Klicpera et al., 2020b). These models operate on graph-structured inputs, where nodes of the graph represent atoms, and edges represent bonds or atomic neighbors within a cutoff distance. Despite their widespread success and the availability of large molecular datasets, training massive GNNs (with up to billions of parameters) is an important but under-explored area. Success of similarly large models in computer vision, natural language processing, and speech recognition (Shoeybi et al., 2020; Huang et al., 2019; Brown et al., 2020; Zhai et al., 2021) suggests that scaling up GNNs could yield large performance gains.

Most previous approaches to scaling GNNs has focused on scaling small models (with up to a few million parameters) to massive graphs. These methods generally assume that we are working with a large, fixed graph, leading to the development of methods like neighborhood sampling (Jangda et al., 2021; Zheng et al., 2021) or efficient partitioning ahead of time (Jia et al., 2020; Ma et al., 2019; Tripathy et al., 2020). These methods do not apply to atomic simulation datasets which contain millions of smaller graphs where it is necessary to consider the entire graph for prediction. In this paper, we focus on this complementary problem of scaling very large models to a dataset of many moderately-sized graphs ($\sim 1k$ nodes, $500k$ edges).

Another limitation of existing methods is that they focus on scaling simple GNN architectures such as Graph Convolutional Networks (GCNs) that only represent lower-order interactions, such as representations for nodes and edges, that are then updated by passing messages between neighboring nodes. In practice, the most successful GNNs used for atomic systems also model higher-order interactions between atoms, such as the interactions between triplets or quadruplets of atoms (Klicpera et al., 2020a; 2021; Liu et al., 2021). These interactions are necessary to capture the geometry of the underlying system, which is critical in making accurate predictions. Scaling up such GNN architectures is challenging because even moderately-sized graphs can contain a large number of higher-order interactions. For example, a single graph with 1000 nodes could contain several mil-

lion combinations of triplets of atoms. In this paper, we introduce *Graph Parallelism*, an approach to scale up training of such GNNs with billions of parameters, by splitting up the graph across multiple GPUs.

We benchmark our approach by scaling up two recent GNN architectures – DimeNet++ (Klicpera et al., 2020a) and GemNet-T (Klicpera et al., 2021) – on the Open Catalyst (OC20) dataset (Chanussot et al., 2021). The OC20 dataset, aimed at discovering new catalyst materials for renewable energy storage, consists of $134M$ training examples spanning a wide range of adsorbates and catalyst materials. A GNN that can accurately predict per-atom forces and system energies on OC20 has the potential to replace the computationally expensive quantum chemistry calculations based on Density Functional Theory (DFT) that are currently the bottleneck in computational catalysis. Our large-scale, graph-parallelized models lead to relative improvements of 1) 15% for predicting forces on the force MAE metric (S2EF task), and 2) 21% on the AFbT metric for predicting relaxed structures (IS2RS task), establishing new state-of-the-art results on this dataset.

2 Graph Parallelism

2.1 Extended Graph Nets

Battaglia et al. (2018) introduced a framework called Graph Network (GN) that provides a general abstraction for many popular Graph Neural Networks (GNNs) operating on edge and node representations of graphs. We build on their work and define the Extended Graph Network (EGN) framework to include GNNs that also operate on higher order terms like triplets or quadruplets of nodes.

In the Graph Network (GN) framework, a graph is defined as a 3-tuple $\mathbf{G} = (\mathbf{u}, V, E)$, where $\mathbf{u}$ represents global attributes about the entire graph; $V = \{\boldsymbol{v}_i\}_{i=1:N^v}$ is the set of all nodes, with $\boldsymbol{v}_i$ representing the attributes of node i; and $E = \{(\mathbf{e}_k, r_k, s_k)\}_{k=1:N^e}$ is the set of edges in the graph where $\mathbf{e}_k$ represents the edge attributes for the edge from node s_k to r_k. A Graph Net is a neural network that contains a series of GN blocks that iteratively operate on the input graph, updating the various representations.

In our *Extended Graph Network (EGN)* framework, a graph is defined as a 4-tuple $\mathbf{G} = (\mathbf{u}, V, E, T)$, where $\mathbf{u}$, V, and E are defined as in the Graph Network, and $T = \{(\mathbf{t}_m, e_{m_1}, e_{m_2}, \ldots)\}$ is the set of higher-order interaction terms involving edges indexed by $m_1, m_2, \ldots$.

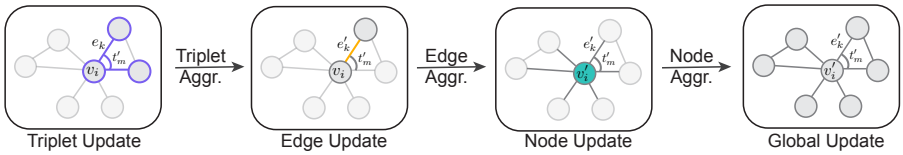
As a concrete example, consider an atomic system represented as a graph in this framework with the nodes representing the atoms and the edges representing atomic neighbors. The node attributes v_i and edge attributes e_k could represent the atom's atomic numbers and distances between atoms respectively. The higher order interactions could represent triplets of atoms, i.e. pairs of neighboring edges with $\mathbf{t}_m$ representing the bond angle, which is the angle between edges that share a common node. Finally, the global attribute $\mathbf{u}$ can represent the energy of the system. For clarity of exposition, we will limit our discussion to triplets in the rest of the paper, but higher order interactions can be handled in a similar manner. We denote these triplets as $(\mathbf{t}_m, e_{m_1}, e_{m_2})$.

In the EGN framework, neural networks contain a series of *EGN blocks* that iteratively update the graph. Each EGN block consists of several *update* and *aggregation* functions that are applied to transform an input graph $(\mathbf{u}, V, E, T)$ into an output graph $(\mathbf{u}', V', E', T')$. Each EGN block starts by updating the highest order interactions, which are then aggregated before updating the next highest order interaction. For instance, the triplet representations are first updated (using $\mathrm{TripletUpdate}$ function) and then aggregated ($\mathrm{TripletAggr}$) to obtain aggregated triplet representations. These aggregated representations are then used to update the edge representation ($\mathrm{EdgeUpdate}$). Next, the edges going into a node are aggregated and used to update the node representations, and so on. This is illustrated in figure 1a.

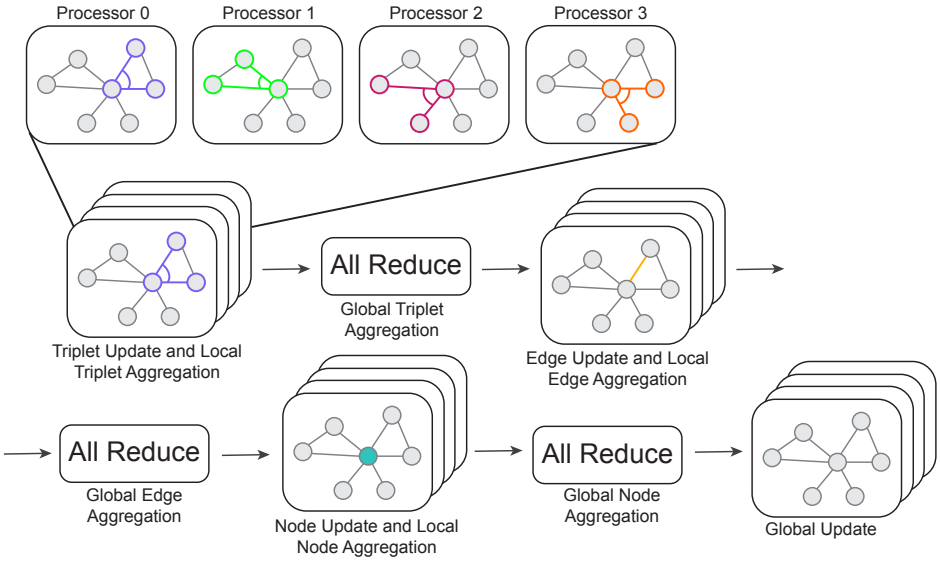
Many popular GNNs can be implemented in this framework by making appropriate choices for the various update and aggregation functions.

2.2 Graph Parallelism for Extended Graph Nets

Training large EGNs can be challenging even on moderately sized graphs because of the large memory footprint required in storing and updating the representation for each triplet, edge, and node. In



(a) Forward computation of an EGN block. First, each triplet representation is updated ($\mathrm{TripletUpdate}$ function), followed by an aggregation of these updated representations ($\mathrm{TripletAggr}$). These aggregated values are then used to update the edge representations ($\mathrm{EdgeUpdate}$), followed by an edge aggregation ($\mathrm{EdgeAggr}$) and so on until the node and global representations are also updated.



(b) Distributed computation of an EGN block. The graph is split up among the different GPUs such that processing unit p contains its subset of triplets $T^{(p)}$ in memory, along with all edges E and nodes V. The triplet attributes are updated in parallel, followed by a triplet aggregation to locally update the edge attributes. Next, an allreduce operation is performed to update all edge attributes globally. We continue this process to update the node and global attributes.

many applications, the number of edges is one or two orders of magnitude larger than the number of nodes, while the number of triplets is one or two orders of magnitude larger still. Thus, storing and updating the triplet representations is often the bottleneck in terms of GPU memory and compute. Many recent methods such as (Klicpera et al., 2020a; 2021) overcome this problem by using a very low-dimensional representation for the triplets. However, we found this to significantly reduce model capacity leading to underfitting for some applications with large training datasets. It is necessary to overcome these memory limitations for better generalization.

One way to get around the memory limits is to distribute the model computation across multiple GPUs. For a graph neural network, a natural way to do this is by splitting the graph across multiple GPUs. The update functions in an EGN are easy to apply in parallel since they are applied independently for each triplet, edge, or node. It is substantially more challenging to apply the aggregations in parallel. To simplify parallel aggregation, we restrict ourselves to aggregation functions that are commutative and associative. This is not limiting in practice since most popular GNN architectures use sum or mean aggregations which can be implemented in this manner.

We will now describe the implementation of the distributed EGN block. Suppose we have access to P processing units that we wish to split the graph computation over. Each unit p is responsible for computing the updates to a subset of triplets, edges and nodes, that we denote by $T^{(p)}, E^{(p)}$, and $V^{(p)}$, respectively. At the beginning of computation, we split the graph so that the processing

unit p contains its subset of triplets $T^{(p)}$ in memory, along with the entire set of edges E and nodes V. During the forward pass, we first update each set of triplets $T^{(p)}$ in parallel to obtain $T'^{(p)}$, followed by a local triplet aggregation. Next, an all-reduce operation is performed on these locally aggregated triplet representations to obtain globally aggregated triplet representations. These globally aggregated representations are then used to update edges in parallel, and the process is repeated for nodes and ultimately for the global graph level representation. This process is illustrated in figure 1b.

In this framework, the highest order interaction attributes are never communicated across processors. Therefore, the communication time is bound by the number of lower order node interactions. In our example, the triplet representations are not communicated, while the edge and node representations are communicated once per EGN block, making the total communication cost equal to $O(N_v D_v + N_e D_e)$, where D_v and D_e are the dimensions of node and edge representations. This makes it possible to work with a large number of triplets and large triplet representations. In section 3, we show concrete instantiations of this framework for two contemporary GNNs.

3 Graph Parallelism for Atomic Simulations

In this section, we present two concrete examples of using GNNs for the problem of predicting the energy and the forces for an atomic system, modeled as a graph whose nodes represent the atoms and whose edges represent the atoms' neighbors. The GNN takes such a graph as input and predicts the energy of the entire system as well as a 3D force vector on each atom. Two paradigms have been proposed in the literature, that we call *energy-centric* and *force-centric* approaches. These approaches differ in how they estimate forces and whether they are energy conserving, which is an important physical property. We begin by describing the components shared by both approaches, followed by one recent model in each paradigm.

3.1 Inputs and Outputs

The inputs to the network are 3D positions $\mathbf{x}_i \in \mathbb{R}^3$ and atomic number z_i for each atom $i \in \{1 \dots n\}$. The outputs are the per-atom forces $\mathbf{f}_i \in \mathbb{R}^3$ and the energy $E \in \mathbb{R}$ of the entire structure. The distance between atoms i and j is denoted as $d_{ij} = ||\mathbf{x}_i - \mathbf{x}_j||$. If edges (i, j) and (j, k) exist, then (i, j, k) defines a *triplet* in the graph and we denote the angle between the edges as $\alpha_{kj,ji}$.

The input graph is constructed with each atom t (*target*) as a node and the edges representing the atom's neighbors $s \in N_t$ where N_t contains all atoms s (*source*) within a distance δ, which is treated as a hyperparameter. Each edge has a corresponding message m_{st} that passes information from source atom s to target atom t.

3.2 Estimating Forces and Energy

As previously stated, there are two paradigms for estimating the energy and forces for an atomic system: *energy-centric models* and *force-centric models*. In energy-centric models, the model first computes the energy E as a function of the inputs by applying a forward pass of the GNN: $E = \mathbf{GNN}(\mathbf{x}, \mathbf{z})$, where $\mathbf{x}$, and $\mathbf{z}$ represent the atomic positions and atomic numbers respectively. The forces are then computed as the gradient of the energy with respect to atomic positions by using backpropagation: $\mathbf{f} = \nabla_{\mathbf{x}} E$.

In force-centric models, the energy and forces are both computed directly during the forward propagation: $E, \mathbf{F} = \mathbf{GNN}(\mathbf{x}, \mathbf{z})$ where $\mathbf{F}$ represents the matrix of all atomic forces. Force-centric models tend to be more efficient in terms of computation time and memory usage compared to the energy-centric models. However, energy-centric models guarantee that the forces are energy conserving which is an important physical property satisfied by atomic systems. In this work, we demonstrate the benefits of scaling up GNNs in both paradigms.

3.3 Energy-Centric model: Dimenet++

Dimenet++ (Klicpera et al., 2020a) is a recently proposed energy-centric model for predicting properties from atomic systems. In this model, the edges are represented by a feature representation

$\mathbf{m}_{ji}$, which are iteratively updated using both directional information (via bond angles) as well as the interatomic distances. The edges are initially represented using a radial basis function (RBF) representation $\mathbf{e}_{RBF}^{(ji)}$ of their lengths d_{ji}. The triplets are represented using a spherical basis function (SBF) expansion $\mathbf{a}_{SBF}^{(kj,ji)}$ of the distances d_{kj} as well as bond angles $\alpha_{(kj,ji)}$. In each block, the messages are updated as:

$$\mathbf{m}'_{ji} = \mathrm{f}_{\mathrm{update}}(\mathbf{m}_{ji}, \sum_{k \in \mathcal{N}_j \setminus \{i\}} \mathrm{f}_{\mathrm{int}}(\mathbf{m}_{kj}, \mathbf{e}_{RBF}^{(ji)}, \mathbf{a}_{SBF}^{(kj,ji)})) \tag{1}$$

where the interaction function f_{int} corresponds to the $\mathrm{TripletUpdate}$ function, the summation corresponds to the $\mathrm{TripletAggr}$ function, and the update function f_{update} to the $\mathrm{EdgeUpdate}$ function respectively. The interaction function consists of a hadamard product of the embeddings, followed by a multi-layer perceptron. To speed up these computations, the embeddings are projected down to a smaller dimension before computing the interactions, and later projected back up to the original dimension.

The updated messages are then input to an output block which are summed up per atom i to obtain a per-atom representation: $\mathbf{h}_i = \sum_j \mathbf{m}'_{ji}$ ($\mathrm{EdgeAggr}$ function). These are then transformed by another MLP to obtain the node representation $\boldsymbol{v}'_i$ ($\mathrm{NodeUpdate}$ function).

Thus, Dimenet++ is a special case of an EGN, and we can closely follow the recipe from section 2.2 to parallelize it.

3.4 FORCE-CENTRIC MODEL: GEMNET

Gemnet (Klicpera et al., 2021) extends Dimenet++ in a number of ways, including the addition of quadruplets as well as a force-centric version. Here, we focus only on the force-centric Gemnet-T model since it was recently shown to obtain state-of-the-art results on the OC20 dataset[1]. Gemnet-T largely follows the structure of the Dimenet++ model, but includes some modifications.

First, Gemnet-T uses a bilinear layer instead of the hadamard product for the interaction function. This is made efficient by optimally choosing the order of operations in the bilinear function to minimize computation. Second, Gemnet-T maintains an explicit embedding for each atom that is first updated by aggregating the directional embeddings involving that atom, similar to Dimenet++. Next, the updated atom embedding is used to update each of the edge embeddings. This creates a second edge update function, $\mathrm{EdgeUpdate}'$, that is run after the node embeddings are updated. Third, Gemnet makes use of symmetric message passing, that is the messages $\mathbf{m}_{ji}$ and $\mathbf{m}_{ij}$ that are on the same edge, but in different directions, are coupled. In a parallel implementation, this step requires an additional all-reduce step since messages $\mathbf{m}_{ji}$ and $\mathbf{m}_{ij}$ could be on different processors.

Thus, Gemnet-T largely follows the EGN framework, with a few minor deviations from the standard formulation. The distributed EGN implementation described in section 2.2 can be used for the Gemnet-T as well, but with additional communication steps to account for the second edge update function and symmetric message passing.

4 EXPERIMENTS

In this section, we present the results of our scaling experiments on the OpenCatalyst 2020 (OC20) dataset (Chanussot et al., 2021). The OC20 dataset contains over 130 million atomic structures used to train models for predicting forces and energies during structure relaxations. We report results for three tasks: 1) Structure to Energy and Forces (S2EF) that involves predicting energy and forces for a given structure; 2) Initial Structure to Relaxed Energy (IS2RE) that involves predicting the relaxed energy for a given initial structure; and 3) Initial Structure to Relaxed Structure (IS2RS) which involves performing a structure relaxation using the predicted forces. Dimenet++ and Gemnet-T are the current state-of-the-art energy-centric and force-centric models respectively.

[1] https://opencatalystproject.org/leaderboard.html

Model	#Params	S2EF Test			
		Energy MAE (EV)↓	Force MAE (EV/A)↓	Force Cos↑	EFwT↑
Gemnet	31M	0.2924	0.02422	0.6162	1.2%
SpinConv	8.9M	0.3363	0.02966	0.5391	0.45%
ForceNet-Large	34.8M	2.2628	0.03115	0.5195	0.01%
Dimenet++-Large	10.8M	31.5409	0.03132	0.544	0%
Gemnet-XL	300M	**0.2701**	**0.0204**	**0.6603**	**1.81%**

Table 1: Experimental results on the S2EF task comparing our Gemnet-XL to the top three entries on the OpenCatalyst leaderboard, showing metrics averaged across the 4 test datasets.

4.1 Experimental Setup

Dimenet++-XL. Our Dimenet++ model consists of $B = 4$ interaction blocks, with a hidden dimension of $H = 2048$, an output block dimension of $D = 1536$, and intermediate triplet dimension of $T = 256$. This model has about $240M$ parameters, which is over $20\times$ larger than the Dimenet++-large model used in (Chanussot et al., 2021). We call this model *Dimenet++-XL*. The model was trained with the AdamW optimizer (Kingma & Ba, 2015; Loshchilov & Hutter, 2019) starting with an initial learning rate of 10^{-4}, that was multiplied by 0.8 whenever the validation error plateaus. The model was trained with an effective batch size of 128 on 256 Volta 32GB GPUs with a combination of data parallel and graph parallel training: each graph was split over 4 GPUs with data parallel training across groups of 4 GPUs.

Gemnet-XL. Our Gemnet model consists of $B = 6$ interaction blocks, with an edge embedding size of $E = 1536$, triplet embedding size of $T = 384$ and embedding dimension of the bilinear layer of $B = 192$. We found that it was beneficial to use a small atom embedding size of $A = 128$, much smaller than the previous SOTA Gemnet model. [2]. This model has roughly 300M parameters, which is about $10\times$ larger than the previous SOTA model. However, since we reduced the atom embedding dimension and increased the edge and triplet dimensions, the total amount of compute and memory usage is significantly larger. We call this model *Gemnet-XL*. We followed the same training procedure as with Dimenet++, except for a starting learning rate of 2×10^{-4}.

4.2 Structure to Energy and Forces (S2EF)

The Structure to Energy and Forces task involves taking an atomic structure as input and predicting the energy of the entire structure and the forces on each atom. The S2EF task has four metrics: the energy and force Mean Absolute Error (MAE), the Force Cosine Similarity, and the Energy and Forces within a Threshold (EFwT). The EFwT metric indicates the percentage of energy and force predictions that are below a preset threshold.

Table 1 compares the top 4 models on the Open Catalyst Project leaderboard[3]. with the Gemnet-XL model. Gement-XL obtains a roughly 16% lower force MAE and an 8% lower energy MAE relative to the previous state-of-the-art. Further, Gemnet-XL improves the EFwT metric more than 50% relative to the previous best, although the value is still very small. These results indicate that model scaling is beneficial for the S2EF task.

4.3 Initial Structure to Relaxed Structure (IS2RS)

The Initial Structure to Relaxed Structure (IS2RS) task involves taking an initial atomic structure and predicting the atomic structure that minimizes the energy. This is performed by iteratively predicting the forces on each atom and then using these forces to update the atomic positions. This process is repeated until convergence or 200 iterations. There are three metrics for this task: Average Distance within Threshold (ADwT), that measures the fraction of final atomic positions within a distance threshold of the ground truth; Forces below Threshold (FbT), which measures whether a true energy

[2]https://discuss.opencatalystproject.org/t/new-gemnet-dt-code-results-model-weights/102

[3]https://opencatalystproject.org/leaderboard.html

Model	**#Params**	**Training Dataset**	**IS2RS Test**		
			AFbT↑	ADwT↑	FbT↑
GemNet-T	31M	S2EF-All	27.6%	58.68%	0.7%
DimeNet++-large	10.8M	S2EF-All	21.82%	51.68%	0.4%
DimeNet++	1.8M	S2EF 20M + MD	17.15%	47.72%	0.15%
Spinconv	8.9M	S2EF-All	16.67%	53.62%	0.05%
Dimenet++-XL	240M	S2EF 20M + MD	33.44%	59.21%	1.25%
Gemnet-XL	300M	All	30.82%	62.65%	0.90%

Table 2: Experimental results on the IS2RS task comparing our models to the top four entries on the OpenCatalyst leaderboard, showing metrics averaged across the 4 test datasets. The Dimenet++ and Dimenet++-XL models were trained on the S2EF 20M + MD dataset, that contains additional molecular dynamics data, which has been shown to be helpful for the IS2RS task.

minimum was found (i.e., forces are smaller than a preset threshold); and, the Average Forces below Threshold (AFbt), which averages the FbT over several thresholds.

Table 1 shows the results on the IS2RS task, comparing our models with the top few models on the OpenCatalyst leaderboard. Both the Dimenet++-XL and Gemnet-XL models outperform all existing models. The Dimenet++-XL model obtains a relative improvement of 53% on the AFbT metric, and more than triples the FbT metric compared to the Dimenet-large model. The Gemnet-XL model obtains similar improvements compared to the smaller Gemnet-T model. These results underscore the importance of model scaling for this task.

4.4 Initial Structure to Relaxed Energy (IS2RE)

The Initial Structure to Relaxed Energy (IS2RE) task takes an initial atomic structure and attempts to predict the energy of the structure after it has been relaxed. Two approaches can be taken to address this problem, the direct and the relaxation approaches (Chanussot et al., 2021). In the direct approach, we treat this task as a regression problem, and train a model to directly estimate the relaxed energy for a given atomic structure. Relaxation approaches typically outperform the direct approaches, though they are generally two orders of magnitude slower during inference time.

Table 3 compares our Gemnet-XL model to the top three models from the Open Catalyst Project leaderboard. There are two metrics for the IS2RE task: energy Mean Absolute Error (MAE) and the Energy within Threshold (EwT) which measures the percentage of time the predicted energy is within a threshold of the true energy. Table 3 shows that the Gemnet-XL model obtains a roughly 8% lower energy MAE and a 12% higher EwT compared to the previous best, which is the smaller Gemnet-T model, demonstrating the benefits of scaling up IS2RE models.

Since direct IS2RE models are very fast at inference time, there are applications where they are more useful than relaxation based approaches. It is possible to convert a trained S2EF model into a direct IS2RE model by fine-tuning it on the IS2RE training data. We finetune our Gemnet-XL model in this manner for 5 epochs, starting with an initial learning rate of 3×10^{-5} that is exponentially decayed by multiplying with 0.95 at the end of each epoch. We call the resuting model *Gemnet-XL-FT*, which obtains a roughly 9.5% better energy MAE and 24% better EwT compared to the current state-of-the-art direct approach (table 3).

4.5 Scaling Analysis

In this section, we analyze the scaling behavior of our models in terms of weak scaling efficiency. Weak scaling studies the effect on the run time when computation is scaled proportional to the number of processors. We study weak scaling efficiency in terms of scaling batch size as well as scaling model size. For these experiments, we use 4 different Gemnet-T models, ranging from 125.4 million parameters to 1.12 billion parameters. The hyper-parameters of these models are specified in table 4.

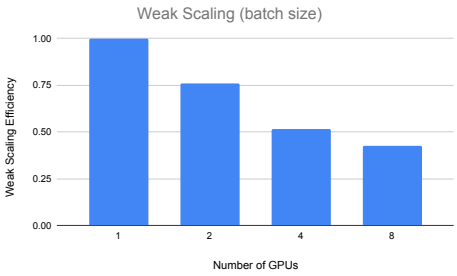
Model	Approach	S2EF Test	
		Energy MAE (EV)↓	EwT↑
Gemnet	Relaxation	0.3997	9.86%
Spinconv	Relaxation	0.4343	7.90%
Noisy Nodes	Direct	0.5099	4.51%
Gemnet-XL	Relaxation	**0.3712**	**11.13%**
Gemnet-XL-FT	Direct	0.4623	5.60%

Table 3: Experimental results on the IS2RE task comparing our Gemnet-XL to the top thr entries on the OpenCatalyst leaderboard, showing metrics averaged across the 4 test datasets.

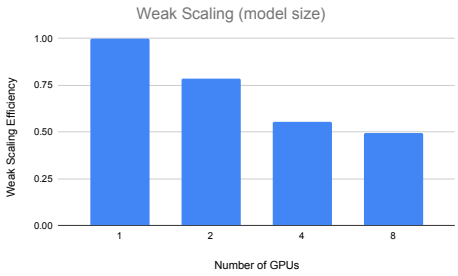
Edge Dim	Atom Dim	Triplet Dim	Bilinear Dim	Num Params	Num GPUs
1024	1024	128	64	125.4M	1
1504	1504	160	96	269.9M	2
2048	2048	256	128	500.5M	4
3072	3072	384	192	1.12B	8

Table 4: Parameters used for scaling analysis. Number of interaction blocks is kept fixed at 3. To measure weak scaling efficiency by batch size, we only use the first model and increasing the batch size proportional to number of GPus. To measure weak scaling efficiency by model size, we train different models on different numbers of GPUs (last column).

To study the effect of scaling the batch size, we train the smallest Gemnet-T model from table 4 on different numbers of GPUs by scaling the batch size with the number of GPUs. Figure 2a shows that we obtain roughly 46% scaling efficiency with 8 GPUs. Next, we study the effect of scaling the model size, by training increasingly larger models on multiple GPUs, that could not be trained on a single GPU. Our largest model contains, which we train on 8 GPUs, contains over 1B parameters. Figure 2b shows that with this model, we are able to obtain a scaling efficiency of roughly 50% with 8 GPUs.



(a) Weak scaling efficiency by batch size. The model size is fixed and batch size is set proportional to the number of GPUs.



(b) Weak scaling efficiency by model size. The model size is scaled to keep the computational cost roughly proportional to the number of GPUs.

Figure 2: Weak scaling efficiency for the Gemnet-T model as a function of the number of GPUs.

5 Related Work

GNNs for simluating atomic systems Many GNN based approaches have been proposed for the task of estimating atomic properties such as (Schütt et al., 2017b; Gilmer et al., 2017; Jørgensen et al., 2018; Schütt et al., 2017a; 2018; Xie & Grossman, 2018; Qiao et al., 2020; Klicpera et al., 2020b), where the atoms are represented by nodes and neighboring atoms are connected by edges. An early approach for force estimation was the SchNet model Schütt et al. (2017a), which computed forces using only the distance between atoms without the use of angular information. SchNet pro-

posed the use of differentiable edge filters which enabled constructing energy-conserving models by estimating forces as the gradient of the energy. Subsequent work (Klicpera et al., 2020a;b; 2021; Liu et al., 2021) has extended on the SchNet model by adding bond angles and dihedral angles, which has resulted in improved performance. These models make use of higher order interactions among nodes which make them highly compute and memory intensive. An alternate approach for estimating forces is to directly regress the forces as an output of the network. While this approach does not enforce energy conservation or rotational equivariance, recent work (Hu et al., 2021) has shown that such models can still accurately predict forces.

Distributed GNN Training Recent research on distributed GNN training has focused on the setting of training a small, simple GNN on a single, large graph. One line of work has focused on sampling local neighborhoods around nodes to create mini-batches (Jangda et al., 2021; Zhang et al., 2020; Zhu et al., 2019). While these approaches can scale to very large graphs, they are not suitable for the task of modeling atomic systems where it is important to consider the entire graph for predicting the energy and forces.

An alternate line of work, that is more similar to ours, keeps the entire graph in memory by efficiently partitioning the graph among multiple nodes (Jia et al., 2020; Ma et al., 2019; Tripathy et al., 2020). These methods still operate in a single graph regime that is partitioned ahead of time. Thus the focus of that work is on finding efficient partitions of the graph, which is not applicable to our problem since we operate on millions of graphs. Further, these works do not consider the problem of training very large GNNs, or GNNs that operate on higher order interaction terms like triplets, that are important for our task.

Model Parallelism Several model parallel methods have been developed recently for training large models that do not fit into memory by splitting the model among multiple GPUs. GPipe (Huang et al., 2019) splits up deep networks depth-wise into several partitions of consecutive layers that are placed on different GPUs. During training, mini-batches are split up into micro-batches and pipelining is used to overlap communication with computation. Another approach, adopted by (Shoeybi et al., 2020) splits up the model breadth-wise, where each layer is partitioned among multiple GPUs using custom algorithms to distribute computation them. We see model parallelism as a complementary approach to graph parallelism that can be profitably combined to train even larger models. We leave this for future work.

6 Discussion

In this paper, we have presented a new method for training large GNNs for modeling atomic simulations. We showed that training larger versions of Dimenet++ and Gemnet-T models can yield significant improvements on each task of the OC20 dataset. It is possible to combine graph parallelism with model parallel methods such as pipeline parallelism (Huang et al., 2019) to train even larger models, which could yield further improvements.

We believe that scaling to large models is essential to effectively model atomic simulations from a wide variety of adsorbates and catalysts. However, scaling up current models may not be sufficient by itself. Novel GNN architectures that better capture the 3D geometry, while respecting physical priors, combined with large scale are likely needed to make substantial progress on this important task.

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A APPENDIX

A.1 ADDITIONAL RESULTS

Tables 5, 6 and 7 show results from each of the four test sets for the S2EF, IS2RE and IS2RS tasks respectively. Dimenet++-XL and Gemnet-XL achieve the best results for each test set along each metric.

Model	**S2EF Test**			
	Energy MAE (EV)↓	Force Cos↑	Force MAE (EV/A)↓	EFwT↑
		ID		
GemNet-T	0.2257	0.637	0.02099	2.4%
Spinconv	0.2612	0.5479	0.02689	0.82%
ForceNet-Large	2.0674	0.533	0.02782	0.02%
Dimenet++-large	29.3333	0.5633	0.02807	0%
Gemnet-XL	**0.2120**	**0.6759**	**0.0181**	**3.30%**
		OOD Ads		
GemNet-T	0.2099	0.6242	0.02186	1.15%
Spinconv	0.2752	0.5345	0.02769	0.38%
ForceNet-Large	2.4188	0.5212	0.02834	0.01%
Dimenet++-large	30.0338	0.5503	0.02896	0%
Gemnet-XL	**0.1980**	**0.6642**	**0.0186**	**1.62%**
		OOD Cat		
GemNet-T	0.3403	0.5813	0.02445	0.93%
Spinconv	0.3501	0.5187	0.02849	0.46%
ForceNet-Large	2.0203	0.4936	0.03089	0.01%
Dimenet++-Large	30.0437	0.5109	0.0312	0%
Gemnet-XL	**0.3083**	**0.6306**	**0.0206**	**1.72%**
		OOD Both		
GemNet-T	0.3937	0.6221	0.02956	0.3%
Spinconv	0.4585	0.5554	0.03556	0.14%
Forcenet-Large	2.5447	0.5302	0.03754	0%
Dimenet++-Large	36.7529	0.5517	0.03705	0%
Gemnet-XL	**0.362**	**0.6704**	**0.0245**	**0.61%**

Table 5: Full set of results on the S2EF task comparing our Gemnet-XL to the top three entries on the OpenCatalyst leaderboard, showing metrics from each test set.

Model	**Approach**	**S2EF Test**	
		Energy MAE (EV)↓	EwT↑
		ID	
GemNet-T	Relaxation	0.3901	12.37%
Spinconv	Relaxation	0.4207	9.4%
Noisy Nodes	Direct	0.4776	5.71%
Gemnet-XL	Relaxation	**0.3764**	**13.25%**
Gemnet-XL-FT	Direct	0.4194	7.52%
		OOD Ads	
GemNet-T	Relaxation	0.3907	9.11%
Spinconv	Relaxation	0.4381	7.47%
Noisy Nodes	Direct	0.5646	3.49%
Gemnet-XL	Relaxation	**0.3677**	**10.00%**
Gemnet-XL-FT	Direct	0.5258	3.95%
		OOD Cat	
GemNet-T	Relaxation	0.4339	10.09%
Spinconv	Relaxation	0.4579	8.16%
Noisy Nodes	Direct	0.4932	5.02%
Gemnet-XL	Relaxation	**0.4022**	**11.61%**
Gemnet-XL-FT	Direct	0.4373	6.76%
		OOD Both	
GemNet-T	Relaxation	0.3843	7.87%
Spinconv	Relaxation	0.4203	6.56%
Noisy Nodes	Direct	0.5042	3.82%
Gemnet-XL	Relaxation	**0.3383**	**9.65%**
Gemnet-XL-FT	Direct	0.4665	4.19%

Table 6: Experimental results on the IS2RE task comparing our Gemnet-XL to the top three entries on the OpenCatalyst leaderboard, showing metrics from each test set.

Model	**Training Dataset**	**IS2RS Test** AFbT↑	ADwT↑	FbT↑
	ID			
Gemnet-T	S2EF-ALL	33.75%	59.18%	2.0%
Dimenet++-large	S2EF-ALL	25.65%	52.45%	1.0%
SpinConv	S2EF-ALL	21.10%	53.68%	0.2%
DimeNet++	S2EF 20M + MD	21.08%	48.6%	0.2%
Dimenet++-XL	S2EF 20M + MD	**40.00%**	59.90%	**2.4%**
Gemnet-XL	S2EF-ALL	34.61	**62.73%**	**2.4%**
	OOD Ads			
Gemnet-T	S2EF-ALL	26.84%	54.59%	0.2%
Dimenet++-large	S2EF-ALL	20.73%	48.47%	0.4%
SpinConv	S2EF-ALL	15.70%	48.87%	0.0%
DimeNet++-	S2EF 20M + MD	17.05%	42.98%	0.0%
Dimenet++-XL	S2EF 20M + MD	**36.01%**	55.68%	**1.6%**
Gemnet-XL	S2EF-ALL	30.32%	**58.57%**	0.6%
	OOD Cat			
Gemnet-T	S2EF-ALL	24.69%	58.71%	0.4%
Dimenet++-large	S2EF-ALL	20.24%	50.99%	0%
SpinConv	S2EF-ALL	15.86%	53.92%	0%
DimeNet++-	S2EF 20M + MD	16.43%	48.19%	0%
Dimenet++-XL	S2EF 20M + MD	**29.62%**	58.43%	**0.6%**
Gemnet-XL	S2EF-ALL	29.33%	**62.60%**	0.2%
	OOD Both			
Gemnet-T	S2EF-ALL	25.11%	62.23%	0.2%
Dimenet++-large	S2EF-ALL	20.67%	54.82%	0.2%
SpinConv	S2EF-ALL	14.01%	58.03%	0%
DimeNet++-	S2EF 20M + MD	14.02%	51.09%	**0.4%**
Dimenet++-XL	S2EF 20M + MD	28.14%	62.85%	**0.4%**
Gemnet-XL	S2EF-ALL	**29.02%**	**66.72%**	**0.4%**

Table 7: Experimental results on the IS2RS task comparing our models to the top four entries on the OpenCatalyst leaderboard, showing metrics for each test dataset. The Dimenet++ and Dimenet++-XL models were trained on the S2EF 20M + MD dataset, that contains additional molecular dynamics data, which has been shown to be helpful for the IS2RS task.