

Appendix to the paper ‘‘HAGO-Net: Hierarchical Geometric Message Passing for Molecular Representation Learning’’

A1. Proofs of Results

In this section, we present the proofs of the lemmas and theorems in the main paper.

A1.1. Proof of Theorem 4

Theorem 4 *For a chiral molecule \mathcal{A} and its enantiomer \mathcal{A}' , $\text{ROT}(\mathcal{A}) = -\text{ROT}(\mathcal{A}')$. In other words, the ROT is a chiral identity in Definition 3 (in the main paper).*

Proof. Let us consider two enantiomers, denoted as \mathcal{A} and \mathcal{A}' , of a chiral molecule. As \mathcal{A} and \mathcal{A}' are mirror images of each other, we can establish a 3D rectangular coordinate system (x, y, z) . Without loss of generality, we can assume that \mathcal{A} and \mathcal{A}' are symmetric with respect to the xy -plane. In this way, for one atom a_i belonging to \mathcal{A} with coordinates (x_i, y_i, z_i) , the coordinates of its corresponding mirror atom a'_i in \mathcal{A}' would be $(x_i, y_i, -z_i)$.

Then, we calculate the vector \vec{e}_i from the chiral atom a_{chiral} with coordinates $(x_{\text{chiral}}, y_{\text{chiral}}, z_{\text{chiral}})$ to any atom a_i in \mathcal{A} , and the corresponding vector \vec{e}'_i in the mirroring molecule \mathcal{A}' ,

$$\begin{aligned}\vec{e}_i &= (x_i - x_{\text{chiral}}, y_i - y_{\text{chiral}}, z_i - z_{\text{chiral}}) \\ &= (\tilde{x}_i, \tilde{y}_i, \tilde{z}_i) \\ \vec{e}'_i &= (x_i - x_{\text{chiral}}, y_i - y_{\text{chiral}}, -z_i - (-z_{\text{chiral}})) \\ &= (\tilde{x}_i, \tilde{y}_i, -\tilde{z}_i)\end{aligned}\quad (1)$$

With Eq. 1, the normal vector \vec{n} of the plane spanned by vectors \vec{e}_j, \vec{e}_k in \mathcal{A} , and the mirroring one \vec{n}' in \mathcal{A}' , can be written as follows,

$$\begin{aligned}\vec{n} &= \vec{e}_j \times \vec{e}_k \\ &= (\tilde{y}_j \tilde{z}_k - \tilde{y}_k \tilde{z}_j, \tilde{x}_k \tilde{z}_j - \tilde{x}_j \tilde{z}_k, \tilde{x}_j \tilde{y}_k - \tilde{x}_k \tilde{y}_j) \\ \vec{n}' &= \vec{e}'_j \times \vec{e}'_k \\ &= (\tilde{y}_k \tilde{z}_j - \tilde{y}_j \tilde{z}_k, \tilde{x}_j \tilde{z}_k - \tilde{x}_k \tilde{z}_j, \tilde{x}_j \tilde{y}_k - \tilde{x}_k \tilde{y}_j)\end{aligned}\quad (2)$$

We also have $\vec{e}_l = (\tilde{x}_l, \tilde{y}_l, \tilde{z}_l)$ and $\vec{e}'_l = (\tilde{x}_l, \tilde{y}_l, -\tilde{z}_l)$. As $\text{ROT}(\mathcal{A}) = \text{sign}(\langle \vec{n}, \vec{e}_l \rangle) = \text{sign}(\langle \vec{e}_j \times \vec{e}_k, \vec{e}_l \rangle)$ and $\text{ROT}(\mathcal{A}') = \text{sign}(\langle \vec{n}', \vec{e}'_l \rangle) = \text{sign}(\langle \vec{e}'_j \times \vec{e}'_k, \vec{e}'_l \rangle)$. It is evident that $\text{ROT}(\mathcal{A}) = -\text{ROT}(\mathcal{A}')$ by combining with Eq. (2). This leads us to our final conclusion. \square

A1.2. Proof of Lemma 6

Lemma 6 *If an invariant function f takes positions as input, its derivative w.r.t. position is rotation equivariant and translation invariant.*

Proof. Since f is invariant to translations and rotations, we can express this as follows:

$$f(p + \tau) = f(p), \quad (3)$$

$$f(Ap) = f(p), \quad (4)$$

where p signifies a position, τ represents a translation, and A is a rotation matrix, i.e., a transformation matrix that is used to perform a rotation in Euclidean space.

According to the chain rule of the derivative of the composition of two functions, we apply the derivative to both sides of Eq. 3,

$$\frac{df(p + \tau)}{d(p + \tau)} \cdot \frac{d(p + \tau)}{dp} = \frac{df(p)}{dp}. \quad (5)$$

This simplifies to,

$$f'(p + \tau) = f'(p), \quad (6)$$

which shows that the derivative of a translation-invariant function is also translation-invariant.

In the same way, we apply the derivative to both sides of Eq. 4,

$$\begin{aligned}\frac{df(Ap)}{d(Ap)} \cdot \frac{d(Ap)}{dp} &= \frac{df(p)}{dp} \\ \frac{df(Ap)}{d(Ap)} \cdot A &= \frac{df(p)}{dp} \\ f'(Ap) &= f'(p) \cdot A^T.\end{aligned}\quad (7)$$

For rotation matrix, $A^{-1} = A^T$. Pre-multiplying by the rotation matrix Ap results in a rotation equivalent to the rotation produced by post-multiplying by the transpose of the rotation matrix $f'(p)A^T$. Thus, Eq. 7 shows that the derivative of a rotation-invariant function is rotation-equivalent. \square

Table 1: Geometric quantities used in HAGO-Graph

Layer	Geometric quantity	Geometric basis function
1- body layer, <i>i.e.</i> , atoms:		
intra- n -body geometry	atomic number	linear transformation
inter- n -body geometry	distance	radial basis function
2- body layer, <i>i.e.</i> , atomic pairs:		
intra- n -body geometry	length	radial basis function
inter- n -body geometry	plane angle	cubic basis function
3- body layer, <i>i.e.</i> , atomic triplets:		
intra- n -body geometry	triangle area, plane angle	cubic basis function
inter- n -body geometry	dihedral angle	spherical Bessel basis function
4- body layer, <i>i.e.</i> , atomic quintuplets:		
intra- n -body geometry	tetrahedron volume, ROT	linear transformation

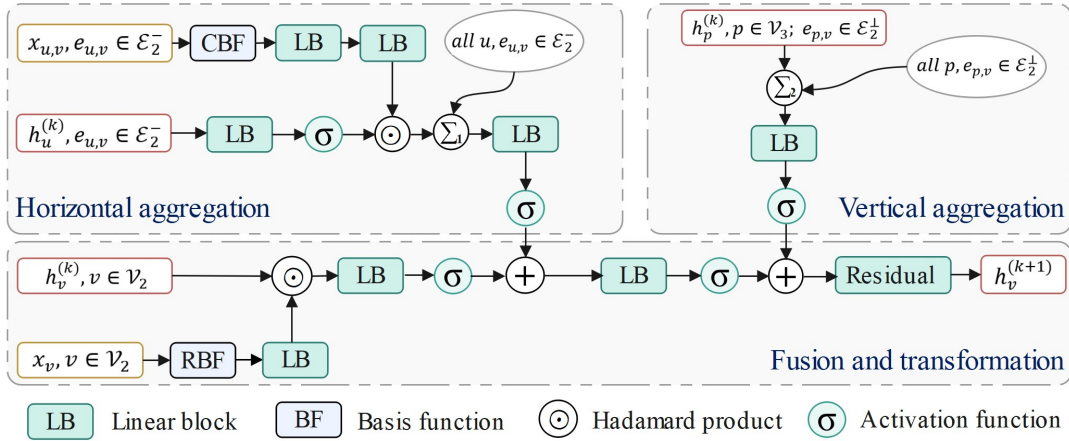


Figure 1: The main architecture of the 2-body layer in HAGO-Net.

A1.3. Proof of Theorem 7

We first present Lemma A1 and Lemma A2, and then we prove Theorem 7 based on the two lemmas.

Lemma A1 *The representation learned by HAGO-Net is invariant to translation and rotation transformations.*

Proof. In this paper, we transform atomic positions \mathcal{P} into translation- and rotation-invariant geometric quantities to construct HAGO-Graph, as shown in Table 1. That is, HAGO-Graph is translation- and rotation-invariant. As the input of HAGO-Net is HAGO-Graph, according to Lemma A1, HAGO-Net is invariant to translation and rotation transformations. \square

Lemma A2 *For a composite function $\phi_1 \circ \phi_2(\mathcal{P})$ taking spatial positions \mathcal{P} as input, if $\phi_2(\mathcal{P})$ is translation- and rotation-invariant, the entire composite function $\phi_1 \circ \phi_2(\mathcal{P})$ is invariant to translation and rotation transformations.*

Proof. Let \mathcal{P}' be an isomorphism of \mathcal{P} after translation and rotation transformations. Given $\phi_2(\mathcal{P})$ is translation- and rotation-invariant, we have $\phi_2(\mathcal{P}) = \phi_2(\mathcal{P}')$. Therefore, the entire composite function $\phi_1 \circ \phi_2(\mathcal{P})$ is translation- and rotation-invariant since $\phi_1 \circ \phi_2(\mathcal{P}) = \phi_1 \circ \phi_2(\mathcal{P}')$. \square

Theorem 7 The representation of DHAGO-Net is rotation equivariant and translation invariant.

Proof. The proof is straightforward by combining with Lemma A1 and Lemma A2. \square

A2. Geometric Quantities in HAGO-Graph

We summarize the geometric quantities used at different layers of the HAGO-Graph in Table 1.

Table 1 also presents the geometric basis functions used to extract information from the geometric quantities. We only use radial basis function (RBF), basis function (CBF), and low-order spherical harmonic functions. There may be more powerful architectures to implement HAGO-MPS with more basis functions, such as irreps features and high-order spherical harmonics.

A3. Architecture of HAGO-Net

We illustrate the architecture of HAGO-Net by taking a typical 2-body layer as an example, as shown in Fig. 1. The architecture consists of three main parts, horizontal aggregation, vertical aggregation, and fusion and transformation. For a node $u \in \mathcal{V}_n$ at the n -th layer of \mathcal{G} , let $\mathbf{h}_u^{(k)}$ denote

the representation vector of node v at k -th iteration. LB denotes to a linear block with one linear layer, and the adopted activation function is SiLU (Sigmoid Linear Unit).

The horizontal aggregation aims at message passing between nodes at 2-body scale, thereby extracting geometric information in 2-bodies. The CBF denotes the cubic basis function. $\mathbf{x}_{u,v}$ denotes the inter-2-body geometric feature between target node u and neighbor node v , which is then encoded by CBF and LBs. We use a Hadamard product to integrate the geometric feature into the representation of each neighbor node v . After aggregating all neighbors of node v at the 2-body scale, the node representation goes through an LB block and an activation function.

The vertical aggregation aims to interact with the upper scale, *i.e.*, 3-body scale in this example. The messages $\mathbf{h}_p^{(k)}$ from the 3-body scale are aggregated and then encoded by an LB block with an activation function.

Fusion and transformation aims to fuse the messages from the neighbors and from the upper scale, and then conduct transformation. The RBF denotes radial basis function, and \mathbf{x}_v is the intra-2-body geometric feature. The geometric message is integrated by using a Hadamard product.

A4. Details in Experiments

A4.1. Hardware and Software Settings

HAGO-Net is implemented using PyTorch-Geometric 2.0.4 and is built upon PyTorch 1.11.0. Our models are trained and evaluated on NVIDIA Tesla V100 GPUs, each with 32GB memory. The used computer is Inspur-NF5468M6, equipped with 475GB of RAM. All programs operate on Ubuntu 18.04.6 within a virtual environment established by Conda 4.12.0. The file describing the detailed environment requirements for HAGO-Net is provided in the code Appendix.

A4.2. Description of Molecular Property

In order to understand the experimental results on the QM9 dataset, we provide a brief description of each molecular property in the QM9 dataset in Table 2. The properties U_0 , U , H , and G are highly related to atoms in a molecule. While, the properties ϵ_{HOMO} , ϵ_{LUMO} , and $\Delta\epsilon$ globally characterize a molecule.

A4.3. Model Configuration

We provide detailed settings of the hyper-parameters utilized for HAGO-Net in our experiments. In every experiment, the size of HAGO-Net’s hidden layer channel is uniformly set to 128. Regarding the learning rate strategy, we adopt StepLR throughout the training phases. StepLR modulates the learning rate by a decay ratio after a predetermined number of epochs. Specific hyper-parameters, inclusive of their values or search spaces, for tasks of property prediction on the QM9 dataset, force prediction on the MD17 dataset, and R/S classification are delineated in Table 4, Table 5, and Table 3, respectively.

Table 2: Description of molecular properties in QM9 dataset

Property	Description
μ , the norm of the dipole moment	The dipole moment approximates the electric field far from a molecule. The magnitude of the dipole moment indicates the degree of charge separation in the molecule, with larger values suggesting greater polarity, the anisotropy in charge distribution.
α , the norm of the static polarizability	The polarizability of a molecule quantifies a molecule’s propensity to induce a dipole moment under an external field. This characteristic correlates with the prominence of Van der Waals interactions.
ϵ_{HOMO} , the highest occupied molecular orbital	ϵ_{HOMO} is the energy of the highest occupied electronic state at zero temperature. The allowed states that electrons can occupy in a molecule are discrete, and no two electrons may occupy the same state. Electrons stack from lowest to highest energy.
ϵ_{LUMO} , the lowest unoccupied molecular orbital	ϵ_{LUMO} is the energy of the lowest occupied electronic state at zero temperature.
$\Delta\epsilon$, the electron energy gap	The energy gap between ϵ_{LUMO} and ϵ_{HOMO} . It represents the minimum energy transition that can take place when an electron is excited from an occupied state to an unoccupied one.
R^2 , the electronic spatial extent	R^2 is the second moment of the charge distribution. It characterizes the spatial distribution of electrons in the molecule.
ZPVE, the zero point vibrational energy,	Vibrational energy at zero temperature.
$U0$, the atomization energy at 0K,	Energy required to break up the molecule into its constituent atoms at absolute zero.
U , the atomization energy at room temperature	Energy required to break up the molecule into its constituent atoms at room temperature.
H , the enthalpy of atomization at room temperature	Enthalpy required to break up the molecule into its constituent atoms at room temperature under fixed pressure.
G , the free energy of atomization	Energy required to break up the molecule into its constituent atoms at fixed temperature and pressure.
C_v , the heat capacity at room temperature	C_v is the amount of heat that must be added to one unit of mass of the molecule in order to cause an increase of one unit in temperature at room temperature.

Table 3: Values/search space for hyper-parameters on R/S classification.

Hyperparameter	Search space
Cutoff	5
Envelope exponent	5
Number of interaction layers	3, 4, 5
Number of RBFs	6
Number of spherical harmonics	7
Out embedding channels	64, 128, 256
Readout MLP Hidden Size	32, 64, 128, 256
Number of readout layers	1, 2, 3, 4
Batch size	8, 16, 32
Initial learning rate	5e-5, 5e-4
Max epochs	100

Table 4: Values/search space for hyper-parameters on QM9

Hyperparameter	Search space
Cutoff	5
Envelope exponent	5
Number of interaction layers	3, 4, 5
Number of RBFs	6
Number of spherical harmonics	2, 3, 5, 7
Batch size	16, 32
Initial learning rate	1e-4, 5e-4, 1e-3
Learning rate strategy	StepLR
Learning rate decay ratio	0.5, 0.6
Learning rate step size	50, 100, 150
Max epochs	500, 750, 1000

Table 5: Values/search space for hyper-parameters on MD17

Hyperparameter	Search space
Cutoff	5
Envelope exponent	5
Number of interaction layers	3, 4, 5
Number of RBFs	6
Number of spherical harmonics	2, 3, 5, 7
Batch size	1, 2, 4, 8
Initial learning rate	1e-4, 5e-4, 1e-3
Learning rate strategy	StepLR
Learning rate decay ratio	0.5, 0.6
Learning rate step size	50, 100, 200
Max epochs	500, 1000, 2000