CORMORANT: COvaRiant MOleculaR Artificial Neural neTworks

N-Atom interactions

\[ V(\mathbf{r}_1, \ldots, \mathbf{r}_N) = \sum_i u_1(\mathbf{r}_i) + \sum_{i<j} u_2(\mathbf{r}_i, \mathbf{r}_j) + \sum_{i<j<k} u_3(\mathbf{r}_i, \mathbf{r}_j, \mathbf{r}_k) + \ldots \]

- one-body
- two-body
- three-body

Goal: Build general architecture for learning physics

- Exploits symmetries of problem
- Exploits hierarchy of scales
- Based upon intuition of N-Atom interactions

[Stillinger, Sakai, and Torquato, JCP (2002)]
Inspiration: Multipoles expansion

\[ V = \frac{1}{4\pi \varepsilon_0} \sum_{i,j} \frac{q_i q_j}{|\mathbf{r}_i - \mathbf{r}_j|} \]

Multiple moments "one-body":

\[ Q_a^\ell = \sum_{i \in N(a)} q_i |\mathbf{r}_{ia}|^\ell Y^\ell(\hat{\mathbf{r}}_{ia}) \]

Under rotations:

\[ Q^\ell \rightarrow D^\ell(\mathbf{R})Q^\ell \]

Multiple energy "two-body" interaction

\[ V = \frac{1}{4\pi \varepsilon_0} \sum_{a,b} \sum_{\ell_1, \ell_2} Y^{\ell_1+\ell_2}(\mathbf{r}_{ab}) C_{\ell_1, \ell_2, \ell_1+\ell_2} Q_{a}^{\ell_1} \otimes Q_{b}^{\ell_2} |\mathbf{r}_{ab}|^{\ell_1+\ell_2+1} \]

\[ V(\mathbf{R}) \sim Q^0 \cdot Y^0(\mathbf{R}) + Q^1 \cdot Y^1(\mathbf{R}) + Q^2 \cdot Y^2(\mathbf{R}) + \ldots \]
SO(3) Vectors: Generalized multipoles

• Irreducible representation “irrep”:

\[ Q_i^\ell \rightarrow F_i^\ell \in \mathbb{C}^{2\ell+1, \tau_\ell} \]

Multiplicity (number of copies of irrep): \( \tau_\ell \)

Transforms covariantly as: \( F^\ell \rightarrow D^\ell(R)F^\ell \)

Generalizes multipoles, e.g., O-H dipole/quadrupole moment

• Representation (list of irreps):

\[ F_i = \bigoplus_\ell F_i^\ell \]

\[ = (F_i^0, F_i^1, \ldots, F_i^{\ell_{\text{max}}}) \]

• Multiplicity of each SO(3) Vector:

\[ \tau = (\tau_0, \tau_1, \ldots, \tau_{\ell_{\text{max}}}) \]
Covariant operations on SO(3) Vectors

- Clebsch-Gordan decomposition of two representations:

\[
F_1 \otimes_{\text{cg}} F_2 = \bigoplus_c \bigoplus_{\ell = |\ell_1 - \ell_2|} C_{\ell_1, \ell_2, \ell} \cdot \left( F_{1,c}^{\ell_1} \otimes F_{2,c}^{\ell_2} \right)
\]

- Multiplicity is treated as “channels”:

\[
\tau = (c, c, \ldots, c)
\]

- SO(3) vectors can be concatenated:

\[
F_1 \oplus F_2 = \bigoplus_{\ell} (F_1^{\ell} \oplus F_2^{\ell})
\]

- SO(3) vectors can be mixed while preserving covariance

\[
F \leftarrow \bigoplus_{\ell} F^{\ell} W^{\ell} \quad W^{\ell} \in \mathbb{C}^{(\tau_{\text{out}}, \tau_{\text{in}})}
\]
Recipe for a Cormorant

• Goal: Build general architecture for learning covariant physics.

• Activations are SO(3)-Vectors $F_i$
  - Use CG, mixing, concatenation

• Construct from general operations involving n=1, 2, 3, … Clebsch-Gordan operations.
  - We call these “interactions”

\[
F_{i}^{s+1} = \left( \bigoplus_{n=1}^{\Phi_{i}^{(n)}} \left( \left\{ F_{j}^{s} \right\} \right) \right) \cdot W
\]
N-Atom interactions

- We base our Cormorant on $n$-atom interactions.
- Define interactions between an atom $i$ and its neighbors $\{F_j\}$
- Must be permutation invariant for $j_k \neq i$
- Allow for a (learnable) transition amplitude $\Upsilon_{j_k,j_{k+1}}^{(n)}$

$$\Phi_i^{(n)} \left( \{F_j\} \right) = \bigoplus_{j_1, \ldots, j_n = i} \bigoplus_{j_1, \ldots, j_n \neq i} \bigotimes_{k=0}^{n-1} \left( \Upsilon_{j_k,j_{k+1}}^{(n)} \otimes F_{j_{k+1}} \right)$$

\[ j_1, \ldots, j_n \in S_i, j_1, \ldots, j_n \in S_i \]
Cormorant architecture (simplified)

Aggregation (one-body):

\[ F^{(ag)}_i \leftarrow \sum_j \Upsilon_{ij} \otimes_{cg} F_j \]

Identity (one-body):

\[ F^{(id)}_i \leftarrow F_i \]

Non-linearity (two-body):

\[ F^{(cg)}_i \leftarrow F_i \otimes_{cg} F_i \]

Concatenation and mixing:

\[ F^{(out)}_i \leftarrow (F^{(ag)}_i \oplus F^{(id)}_i \oplus F^{(cg)}_i) \cdot W \]

Network: Iterate this several times
Experimental results: MD-17 and QM-9

Table 1: Mean absolute error of various prediction targets on QM-9 (left) and conformational energies (in units of kcal/mol) on MD-17 (right). The best results within a standard deviation of three Cormorant training runs (in parenthesis) are indicated in bold.

<table>
<thead>
<tr>
<th>Target Type</th>
<th>Cormorant</th>
<th>SchNet</th>
<th>NMP</th>
<th>WaveScatt</th>
</tr>
</thead>
<tbody>
<tr>
<td>$a$ (bohr$^{-1}$)</td>
<td>0.085</td>
<td>0.235</td>
<td>0.092</td>
<td>0.160</td>
</tr>
<tr>
<td>$\Delta e$ (eV)</td>
<td>0.061</td>
<td>0.063</td>
<td>0.069</td>
<td>0.118</td>
</tr>
<tr>
<td>$\epsilon$ (eV)</td>
<td>0.034</td>
<td>0.041</td>
<td>0.043</td>
<td>0.085</td>
</tr>
<tr>
<td>$\epsilon$ LUMO (eV)</td>
<td>0.038</td>
<td>0.034</td>
<td>0.038</td>
<td>0.076</td>
</tr>
<tr>
<td>$\mu$ (D)</td>
<td>0.038</td>
<td>0.033</td>
<td>0.030</td>
<td>0.340</td>
</tr>
<tr>
<td>$C'_v$ (cal/mol K)</td>
<td>0.026</td>
<td>0.033</td>
<td>0.040</td>
<td>0.049</td>
</tr>
<tr>
<td>$G$ (eV)</td>
<td>0.020</td>
<td>0.014</td>
<td>0.019</td>
<td>0.022</td>
</tr>
<tr>
<td>$H$ (eV)</td>
<td>0.021</td>
<td>0.014</td>
<td>0.017</td>
<td>0.022</td>
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<tr>
<td>$R^2$ (bohr$^2$)</td>
<td>0.961</td>
<td>0.073</td>
<td>0.180</td>
<td>0.410</td>
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<tr>
<td>$U$ (eV)</td>
<td>0.022</td>
<td>0.019</td>
<td>0.020</td>
<td>0.022</td>
</tr>
<tr>
<td>$U_0$ (eV)</td>
<td>0.022</td>
<td>0.014</td>
<td>0.020</td>
<td>0.022</td>
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<tr>
<td>ZPVE (meV)</td>
<td>2.027</td>
<td>1.700</td>
<td>1.500</td>
<td>2.000</td>
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<th>DTNN</th>
<th>SchNet</th>
<th>GDML</th>
<th>sGDML</th>
</tr>
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<tbody>
<tr>
<td>Aspirin</td>
<td>0.098</td>
<td>0.201</td>
<td>–</td>
<td>0.120</td>
<td>0.270</td>
<td>0.190</td>
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<tr>
<td>Benzene</td>
<td>0.023</td>
<td>0.065</td>
<td>0.040</td>
<td>0.070</td>
<td>0.070</td>
<td>0.100</td>
</tr>
<tr>
<td>Ethanol</td>
<td>0.027</td>
<td>0.055</td>
<td>–</td>
<td>0.050</td>
<td>0.150</td>
<td>0.070</td>
</tr>
<tr>
<td>Malonaldehyde</td>
<td>0.041</td>
<td>0.092</td>
<td>0.190</td>
<td>0.080</td>
<td>0.160</td>
<td>0.100</td>
</tr>
<tr>
<td>Naphthalene</td>
<td>0.029</td>
<td>0.095</td>
<td>–</td>
<td>0.110</td>
<td>0.120</td>
<td>0.120</td>
</tr>
<tr>
<td>Salicylic Acid</td>
<td>0.066</td>
<td>0.106</td>
<td>0.410</td>
<td>0.100</td>
<td>0.120</td>
<td>0.120</td>
</tr>
<tr>
<td>Toluene</td>
<td>0.034</td>
<td>0.085</td>
<td>0.180</td>
<td>0.090</td>
<td>0.120</td>
<td>0.100</td>
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SchNet: [Schütt, et al, 2017]  
WaveScatt: [Eickenberg, et al, 2018]  
DeepMD: [Zhang, et al, 2017]  
DTNN: [Schütt, et al, 2017]  
GDML: [Chmiela, et al, 2017]  
sGDML: [Chmiela, et al, 2018]