Equivariant Message Passing for the Prediction of Tensorial Properties and Molecular Spectra

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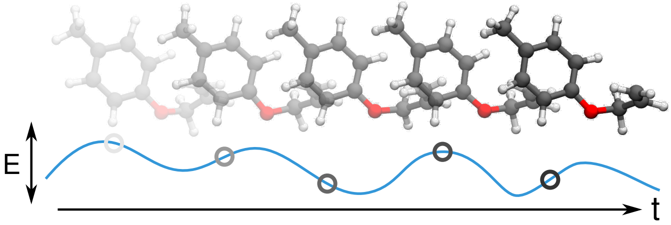
Ab initio molecular dynamics simulations

Computationally costly quantum mechanical calculations

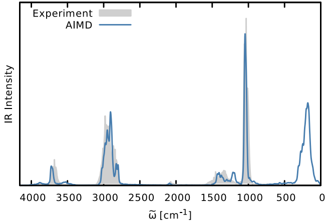
- energy E
- forces $\vec{F} = -\frac{\partial E}{\partial \mathbf{r}}$
- dipole moment $\vec{\mu}$
- polarizability tensor α



$$\rho_{\dot{\mu}}(\tau) = \int_{-\infty}^{+\infty} \dot{\boldsymbol{\mu}}(t)\dot{\boldsymbol{\mu}}(t+\tau)\mathrm{d}t$$

$$\hat{H}\Psi = E\Psi$$

Method	Complexity
Hartree Fock	$O(n^3) - O(n^4)$
Density Functional Theory	$O(n^3) - O(n^4)$
MP2	$O(n^5)$
CCSD	$O(n^6)$
CCSD(T)	$O(n^7)$
Full CI	$O(n!)$

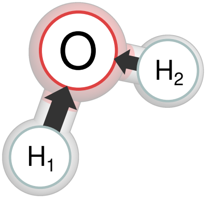


Representing atomic environments

Message passing neural network [Gilmer et al 2017]

$$\mathbf{m}_i^{t+1} = \sum_{j \in \mathcal{N}(i)} \mathbf{M}_t(\mathbf{s}_i^t, \mathbf{s}_j^t, \|\vec{r}_{ij}\|)$$

$$\mathbf{s}_i^{t+1} = \mathbf{U}_t(\mathbf{s}_i^t, \mathbf{m}_i^{t+1})$$

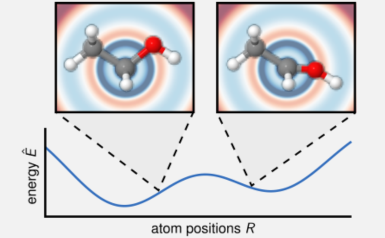


Rotational invariance:

$$\mathbf{M}(\vec{x}) = \mathbf{M}(R\,\vec{x})$$

for any rotation R.

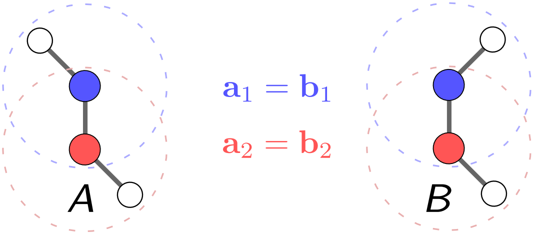
Continuous-filter convolution [Schütt et al 2017]



$$(\mathbf{x} * W)(\mathbf{r}_i) = \sum_{j=1}^{N_{\text{atom}}} \mathbf{x}_j^{(t)} \circ \underbrace{W^{(t)}(\|\mathbf{r}_i - \mathbf{r}_j\|)}_{\text{neural network}}$$

Limits of rotationally invariant message passing

We require **local representations** for $\mathcal{O}$(atoms) scaling. This introduces additional (unwanted) symmetries:



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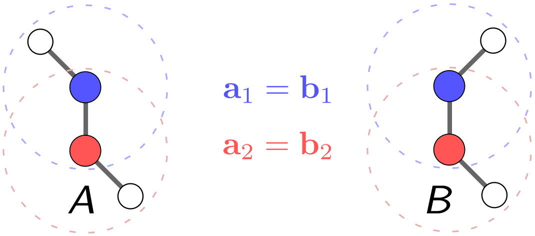


Atom representations need to retain some additional directional information!

Rotationally equivariant message passing

Equivariant message passing:

$$\vec{\mathbf{m}}_i^{v,t+1} = \sum_{j \in \mathcal{N}(i)} \vec{\mathbf{M}}_t(\mathbf{s}_i^t, \mathbf{s}_j^t, \vec{\mathbf{v}}_i^t, \vec{\mathbf{v}}_j^t, \vec{r}_{ij})$$

with

$$R\,\vec{\mathbf{M}}(\vec{\mathbf{x}}) = \vec{\mathbf{M}}(R\,\vec{\mathbf{x}})$$

for any rotation R.

Equivariant building blocks:

- Any (nonlinear) function of scalars: $\mathbf{f}(\mathbf{s})$
- Scaling of vectors: $\mathbf{s} \circ \vec{\mathbf{v}}$
- Linear combinations of equivariant vectors: $\mathbf{W}\vec{\mathbf{v}}$
- Vector products: $\vec{\mathbf{v}}_1 \times \vec{\mathbf{v}}_2$
- Scalar products: $\mathbf{s} = \|\vec{\mathbf{v}}\|^2$, $\mathbf{s} = \langle \vec{\mathbf{v}}_1, \vec{\mathbf{v}}_2 \rangle$

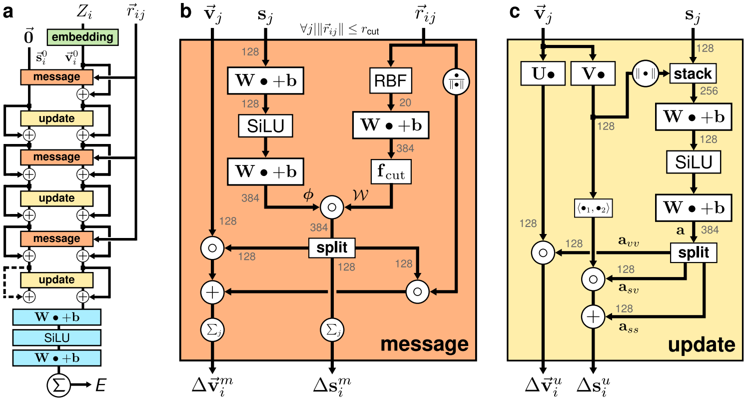
PaiNN - polarizable atom interaction neural network

Scalar features

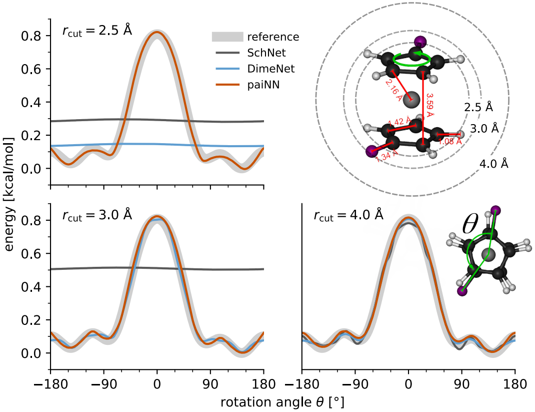
$$\Delta \mathbf{s}_i^m = (\phi_s(\mathbf{s}) * \mathcal{W}_s)_i = \sum_j \phi_s(\mathbf{s}_j) \circ \mathcal{W}_s(\|\vec{r}_{ij}\|),$$

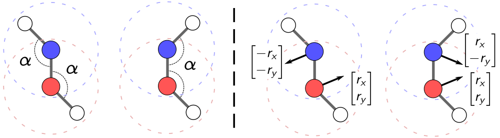
Vectorial features

$$\Delta \vec{\mathbf{v}}_i^m = \sum_j \vec{\mathbf{v}}_j \circ \phi_s(\mathbf{s}_j) \circ \mathcal{W}_{vv}(\|\vec{r}_{ij}\|) + \sum_j \phi_{vs}(\mathbf{s}_j) \circ \mathcal{W}'_{vs}(\|\vec{r}_{ij}\|) \frac{\vec{r}_{ij}}{\|\vec{r}_{ij}\|},$$



Propagation of directional information





Equivariant message passing is able to propagate directional information beyond the radial cutoff

Schütt, Unke, Gastegger (2021). Equivariant message passing for the prediction of tensorial properties and molecular spectra. arXiv:2102.03150

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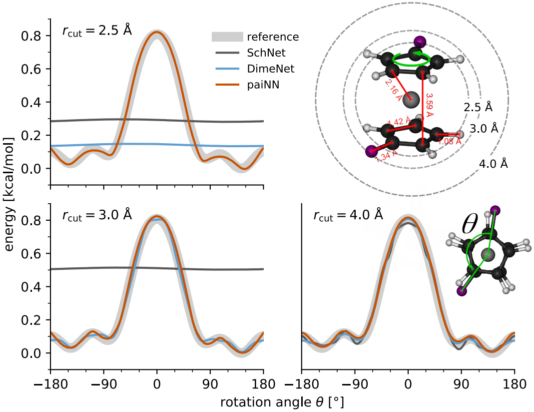

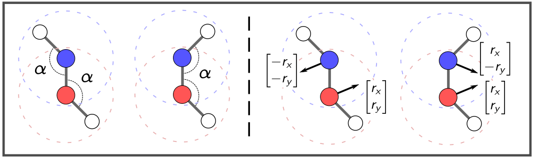

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Predicting tensorial properties

Dipole moment

$$\vec{\mu} = \sum_{i=1}^{N} \vec{\mu}_{\text{atom}}(\vec{\mathbf{v}}_i) + q_{\text{atom}}(\mathbf{s}_i)\vec{r}_i$$

Order-M, rank-r tensors:

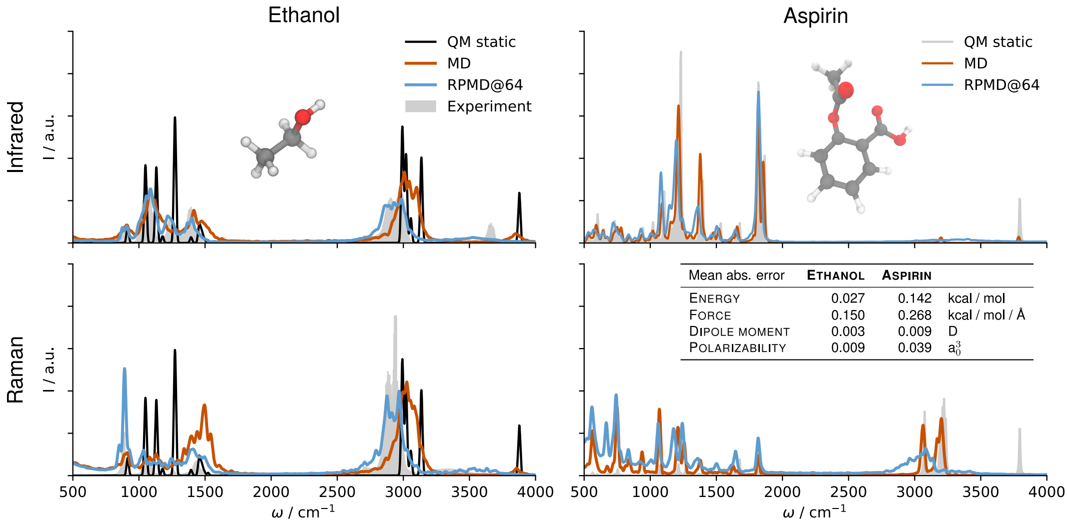
$$T = \sum_{i=1}^{N} \sum_{k=1}^{R} \lambda(\mathbf{s}_i)\vec{\nu}(\vec{\mathbf{v}}_i)_{k,1} \otimes \cdots \otimes \vec{\nu}(\vec{\mathbf{v}}_i)_{k,M}$$

Polarizability tensor:

$$\boldsymbol{\alpha} = \sum_{i=1}^{N} \alpha_0(\mathbf{s}_i)\, I_3 + \vec{\nu}(\vec{\mathbf{v}}_i) \otimes \vec{r}_i + \vec{r}_i \otimes \vec{\nu}(\vec{\mathbf{v}}_i)$$

Molecular spectra

RPMD @ 64 beads of Aspirin: 25 years → 1 h



Mean abs. error	ETHANOL	ASPIRIN	
ENERGY	0.027	0.142	kcal / mol
FORCE	0.150	0.268	kcal / mol / Å
DIPOLE MOMENT	0.003	0.009	D
POLARIZABILITY	0.009	0.039	a_0^3

Conclusion

- Equivariant message passing enables propagation of directional information beyond the cutoff.
- PaiNN enables data-efficient, accurate predictions of tensorial properties.
- Further experiments in the paper:
  - Accurate predictions on QM9 and MD17 benchmark datasets.
  - Ablation studies showing the importance of equivariant features also for scalar properties.