Higher-Rank Irreducible Cartesian Tensors for Equivariant Message Passing

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Code: https://github.com/nec-research/ictp

38th Conference on Neural Information Processing Systems (NeurIPS 2024)

Machine-Learned Interatomic Potentials

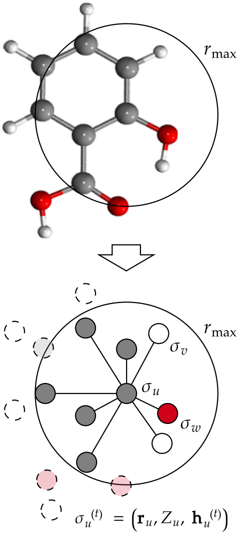
- Learn a function which approximates the potential energy E of an atomic system $S = \{\mathbf{r}_u, Z_u\}_{u=1}^{N_\mathrm{at}}$:

$$f\left(S, \boldsymbol{\theta}\right) : S \mapsto E \in \mathbb{R}$$

- Learn $\boldsymbol{\theta}$ from training data (energy, atomic forces, and stress tensor) at reference level (DFT, CC, ...).
- Consider (semi-)local interactions within r_max:

$$E\left(S, \boldsymbol{\theta}\right) = \sum_{i=u}^{N_\mathrm{at}} \underbrace{E_u\left(\tilde{S}_u, \boldsymbol{\theta}\right)}_{=f\left(\tilde{S}_u, \boldsymbol{\theta}\right)}$$

- Transform $\tilde{S}_u = \{\mathbf{r}_u, Z_u, \{\mathbf{r}_v, Z_v\}_{v \in r_\mathrm{max}}\}$ to incorporate symmetries and many-body terms → message passing.



Many-Body Equivariant Message Passing

- Spherical tensors are conventionally used in equivariant message-passing architectures.
- Their products require complicated numerical coefficients and can be computationally demanding.
- State-of-the-art Cartesian models offer a promising alternative but
  - rely exclusively on convolutions with invariant filters and
  - restrict the construction of many-body features,

  limiting the range of possible architectures and their expressive power.
- We address these limitations by exploring irreducible Cartesian tensors and their irreducible products.

Many-Body Equivariant Message Passing

- Clebsch–Gordan tensor product:

$$\left(Y^{l_1}_{m_1} \otimes Y^{l_2}_{m_2}\right)^{l_3}_{m_3} = \sum_{m_1=-l_1}^{l_1} \sum_{m_2=-l_2}^{l_2} C^{l_3 m_3}_{l_1 m_1, l_2 m_2} Y^{l_1}_{m_1} Y^{l_2}_{m_2},$$

- Convolutions and two-body features:

$$\mathbf{A}^{(t)}_{ukl_3} = \sum_{v \in r_{\mathrm{max}}} \underbrace{\left(R^{(t)}_{kl_1 l_2 l_3}(r_{uv}) \mathbf{Y}_{l_1}(\hat{\mathbf{r}}_{uv})\right)}_{=\text{radial distances} \times \text{unit vectors}} \otimes \underbrace{\left(\sum_{k'} W^{(t)}_{kk'l_2} \mathbf{h}^{(t)}_{vk'l_2}\right)}_{=\text{node embeddings}}$$

- Many-body features:

$$\mathbf{B}^{(t)}_{u\eta_\nu kL} = \underbrace{\left(\mathbf{A}^{(t)}_{ukl_1} \otimes \cdots \otimes \mathbf{A}^{(t)}_{ukl_\nu}\right)}_{\nu\text{-fold}}$$

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- Many-body features:

2-body 3-body 4-body

Irreducible Cartesian Tensors

- Embedding unit vectors ($\mathbf{Y}_l(\hat{\mathbf{r}}) \to \mathbf{T}_l(\hat{\mathbf{r}})$):

$$\mathbf{T}_l(\hat{\mathbf{r}}) = C \sum_{m=0}^{\lfloor l/2 \rfloor} (-1)^m \frac{(2l-2m-1)!!}{(2l-1)!!} \left\{ \hat{\mathbf{r}}^{\otimes(l-2m)} \otimes \mathbf{I}^{\otimes m} \right\}$$

- Irreducible Cartesian tensor product (even, i.e., $l_1 + l_2 - l_3 = 2k$):

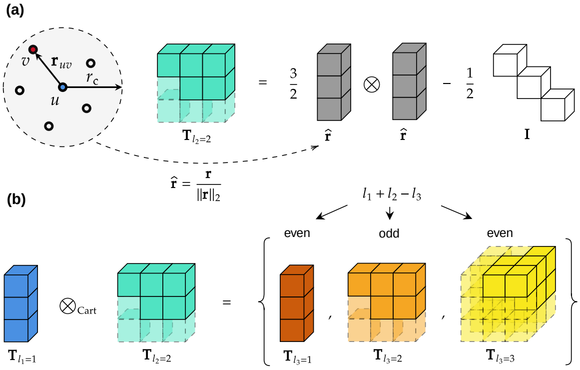
$$(\mathbf{T}_{l_1} \otimes \mathbf{T}_{l_2})_{l_3}$$

$$= C_{l_1 l_2 l_3} \sum_{m=0}^{\min(l_1,l_2)-k} (-1)^m 2^m \frac{(2l_3-2m-1)!!}{(2l_3-1)!!} \left\{ (\mathbf{T}_{l_1} \cdot (k+m) \cdot \mathbf{T}_{l_2}) \otimes \mathbf{I}^{\otimes m} \right\}$$

- Propositions 4.1 & 4.2: The resulting message-passing layers are equivariant to actions of the orthogonal group and preserve the traceless property.

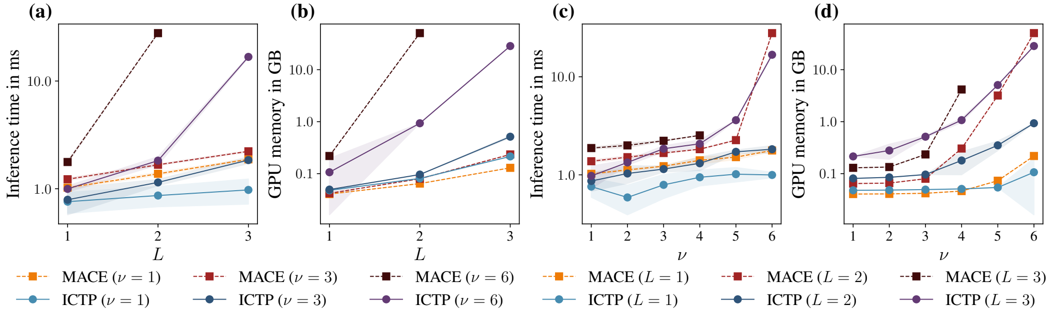
Irreducible Cartesian Tensors

- Embedding unit vectors & irreducible Cartesian tensor products:



- Propositions 4.1 & 4.2: The resulting message-passing layers are equivariant to actions of the orthogonal group and preserve the traceless property.

Scaling and Computational Cost



The required number of calculations:

- Spherical tensors: $(2L+1)^5$ for two-body features and $L^{\frac{1}{2}\nu(\nu+3)}$ (or $\mathcal{K}L^{5(\nu-1)}$) for many-body features.
- Cartesian tensors: $9^L L!/\left(2^{L/2}(L/2)!\right)$ for two-body features and $\mathcal{K}(9^L L!/(2^{L/2}(L/2)!))^{\nu-1}$ for many-body features.

Evaluation on Benchmark Data Sets

- Evaluation on rMD17, MD22, 3BPA, AcAc, and Ta–V–Cr–W data sets.
- Competitive results BUT modifications to architectures are necessary!
- Energy (E, meV) and force (F, meV/Å) RMSEs for the 3BPA data set:

		ICTP$_{\text{full}}$	ICTP$_{\text{sym}}$	ICTP$_{\text{sym+lt}}$	MACE	CACE	MACE	NequIP
300 K	E	**2.70 ± 0.22**	**2.70 ± 0.08**	**2.98 ± 0.34**	**2.81 ± 0.18**	6.3	**3.0 ± 0.2**	3.28 ± 0.10
	F	**9.45 ± 0.29**	**9.39 ± 0.31**	**9.57 ± 0.20**	**9.47 ± 0.42**	21.4	**8.8 ± 0.3**	10.77 ± 0.19
600 K	E	**10.74 ± 0.31**	**10.38 ± 0.80**	**10.29 ± 0.90**	**11.11 ± 1.41**	18.0	**9.7 ± 0.5**	11.16 ± 0.14
	F	**22.99 ± 0.64**	**22.87 ± 0.91**	**23.03 ± 0.76**	**23.27 ± 1.45**	45.2	**21.8 ± 0.6**	26.37 ± 0.09
1200 K	E	**29.80 ± 0.92**	**30.84 ± 1.87**	**31.32 ± 1.80**	**31.15 ± 1.58**	58.0	**29.8 ± 1.0**	38.52 ± 1.63
	F	**62.82 ± 1.23**	**64.54 ± 3.88**	**65.36 ± 3.47**	**65.22 ± 3.52**	113.8	**62.0 ± 0.7**	76.18 ± 1.11
Dihedral slices	E	**9.82 ± 0.79**	10.64 ± 1.07	13.03 ± 3.44	**8.56 ± 1.53**	–	**7.8 ± 0.6**	23.2
	F	**17.52 ± 0.54**	**17.18 ± 0.81**	19.31 ± 0.83	**17.69 ± 1.29**	–	**16.5 ± 1.7**	23.1
Time/structure [ms]		6.45 ± 0.50	5.31 ± 0.02	**3.51 ± 0.22**	4.66 ± 0.05	–	**24.3**	103.5
Memory/batch [GB]		49.66 ± 0.00	42.01 ± 0.11	39.08 ± 0.00	**36.26 ± 0.00**	–	–	–

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