Interpretability in atomic-scale machine learning
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Machine learning in atomic-scale simulations
Predictive accuracy for real materials

- First-principles calculations promise quantitatively accurate simulations that make no use of experimental data
- Emergent physics from first principles: still a tremendous challenge
- Machine learning to the rescue

\[
\hat{H}(\mathbf{q}) |\Psi\rangle = V(\mathbf{q}) |\Psi\rangle
\]

\[
\langle A \rangle = \int d\mathbf{q} e^{-\beta V(\mathbf{q})} A(\mathbf{q})
\]

Grabowski et al., PRB (2009); Kapil, Engel, Rossi, MC, JCTC (2019)
Predictive accuracy for real materials

- First-principles calculations promise quantitatively accurate simulations that make no use of experimental data.
- Emergent physics from first principles: still a tremendous challenge.
- Machine learning to the rescue.

\[ A \equiv \{ a_i, r_i \} \]

**FIRST-PRINCIPLES QUANTUM MODELS**

\[ \hat{H} |\Psi\rangle = V |\Psi\rangle \]

**DATA-DRIVEN SURROGATE MODELS**

\[ \tilde{V}(|A\rangle) \]

\[ V(\{a_i, r_i\}) \]

Musil et al., Chem. Rev. (2021)
Predictive accuracy for real materials

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\[ A \equiv \{ a_i, r_i \} \]

\[ \hat{H} |\Psi\rangle = V |\Psi\rangle \]

\[ \langle \Psi | \hat{Y} |\Psi\rangle \]

\[ V(\{a_i, r_i\}) \]

\[ \tilde{V}(|A\rangle) \]

\[ \tilde{y}_\lambda^\mu(|A\rangle) \]

\[ H \]

\[ \rho \]

Musil et al., Chem. Rev. (2021)
No regression without representation

- Key step in any atomistic ML task: mapping an atomic structure to a suitable mathematical representation
- Features, distances, kernels, can largely be used interchangeably

\[ \text{train set} \quad \text{inference} \quad \text{classification} \quad \text{dimensionality reduction} \]

\[ \{ |x_i\rangle, y_i \} \]

\[ \Theta \]

\[ \mathcal{A}_1, \mathcal{A}_2, \mathcal{A}_3 \]

\[ y_1, y_2, y_3 \]

\[ \mathcal{X}_1, \mathcal{X}_2, \mathcal{X}_3, \mathcal{X}_4 \]

MC, JCP (2019)
No regression without representation

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MC, JCP (2019)
Interpretable structural representations
What do we want from a representation?

1. Be complete (injective)
2. Reflect basic physical symmetries
3. Be smooth, regular
4. Exploit additivity

Musil et al., Chem. Rev. (2021)
Additivity, and locality

- Additive ansatz for extensive properties $\leftrightarrow$ Additive models / features
- Locality and nearsightedness $\rightarrow$ divide et impera, transferability

$$V(A) = \sum_{i \in A} V(A_i)$$

$$|A\rangle = \sum_i |A_i\rangle$$

$$K(A, B) = \sum_{i,j} k(A_i, B_j)$$
Additivity, and locality

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Additivity, and locality

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A phylogenetic tree of ML representations

- Non-linear functions
- Cartesian coordinates
- Atom density fields
- Internal coordinates
- Atom centred distributions
- Atomic symmetry functions
- Permutation invariant polynomials
- Distance histograms
- Permutations (average)
- Equivalent
- Sorted distances
- Permutations (histogram)
- Molecular matrices
- Permutations (sorting)
- Molecular graphs
- Z matrix
- 3D Voxel
- Internal coordinates
- Cartesian coordinates

- Symmetry
- Other relation
- Family of features
- Named features (body order)
  - 2,3,4: radial, angular, dihedrals
  - n: n-body
  - n*: complete n-body linear basis

- Wasserstein metric
- Delta limit
- Smooth
- Sharp
- Blur
- Density correlation features
- Rotations (density products)
- Translations
- Transform
- Potential fields
- Transform
- Global transform
- Diffraction FP
- LODE (n)
- Symmetrized local field
- SOAP (3)
- FCHL (2,3,4)
- Wavelets (3)
- NICE (n*)
- ACE (n*)
- MTP (n*)
- SNAP (4)
- Behler-Parrinello (2,3)
- DeepMD (2,3)
- GTTP (2,3)
- aPIPs (n*)
- Projection

- Behler-Parrinello (2,3)
- DeepMD (2,3)
- GTTP (2,3)
- aPIPs (n*)
- Projected invariant polynomials
- Sort (histogram)
- Permutations
- Molecular matrices
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Musil et al., Chem. Rev. (2021)
Universal feature construction

- Most frameworks can be expressed in terms of $n$-body correlations of atom positions. Only difference - the choice of basis
- Extension to a fully equivariant framework (NICE)
- ... to features to describe long-range interactions (LODE)
- ... and to message-passing, $N$-center features (MP-ACDC)

Introductory review: Musil et al., Chem. Rev. (2021)
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\[
\int d\hat{R} \quad |\lambda \mu \rangle \quad |\rho_i \rangle \quad |\rho_i \rangle
\]

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\[ \int d\hat{R} \]

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**Latest & greatest:** Nigam, Pozdnyakov, Fraux, **MC**, JCP (2022)
A hierarchy of equivariant features

- Neighbor density can be expanded in radial functions and $Y^m_i \rightarrow$ equivariant features
  
  \[ \langle n_1 | \rho_i^{\otimes 1}; \lambda \mu \rangle \equiv \langle n_1 \lambda (-\mu) | \rho_i \rangle \]

- Recursive construction of $N$-body features based on sums of angular momenta
  
  \[ \langle \ldots; n_\nu l_\nu k_\nu; n l k | \rho_i^{\otimes (\nu+1)}; \lambda \mu \rangle = \sum_{q m} \langle n | \rho_i^{\otimes 1}; l m \rangle \langle \ldots; n_\nu l_\nu k_\nu | \rho_i^{\otimes \nu}; k q \rangle \langle l m; k q | \lambda \mu \rangle \]

- All equivariant $\nu$-neighbor features transform like angular momenta
  
  \[ \langle q | \hat{R} A; \rho_i^{\otimes \nu}; \lambda \mu \rangle \sim \sum_{\mu'} D_{\mu \mu'}^\lambda (R) \langle q | A; \rho_i^{\otimes \nu}; \lambda \mu' \rangle \]

  Can be used to compute efficiently invariant features $| \rho_i^{\otimes \nu}; 00 \rangle$

Nigam, Pozdnyakov, MC, JCP (2020); https://github.com/cosmo-epfl/nice
Features, models and introspection
Symmetrized correlations and potentials

- Symmetrized correlations can be linked to body-ordered expansions
  1. $\nu = 2$ correlations: rotation-averaged tensor product of neighbor densities
  2. This is equivalent to a function of two distances and one angle
  3. In the limit of sharp Gaussians, this is equivalent to a list of 2-neighbors tuples $(r_{ji}, r_{j'i}, \hat{r}_{ji} \cdot \hat{r}_{j'i})$
  4. Linear model $\rightarrow$ 3-body potential!

\[
\langle x; x' | A; \rho_i \otimes 2 \rangle = \int d\hat{R} \langle x | \hat{R}A; \rho_i \rangle \langle x' | \hat{R}A; \rho_i \rangle
\]

Willatt, Musil, **MC**, JCP (2019)
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\[
\langle x_1; x_2; \theta | A; \rho_i^{\otimes 2} \rangle = \int d\hat{R} \langle x_1 \hat{R}\hat{e}_z | A; \rho_i \rangle \\
\langle x_2 \hat{R}(\hat{e}_z \cos \theta + \hat{e}_x \sin \theta) | A; \rho_i \rangle
\]

Willatt, Musil, **MC**, JCP (2019)
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4. Linear model $\rightarrow$ 3-body potential!

\[
\langle x_1; x_2; \theta | A; \delta_i \otimes^2 \rangle = \\
\sum_{j_1j_2} \delta(x_1 - r_{j_1i}) \delta(x_2 - r_{j_2i}) \delta(\cos \theta - \hat{r}_{j_1i} \cdot \hat{r}_{j_2i})
\]

Willatt, Musil, **MC**, JCP (2019)
Symmetrized correlations can be linked to body-ordered expansions

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3. In the limit of sharp Gaussians, this is equivalent to a list of 2-neighbors tuples \((r_{j_1i}, r_{j_2i}, \hat{r}_{j_1i} \cdot \hat{r}_{j_2i})\)
4. Linear model \(\rightarrow 3\)-body potential!

\[ \int \langle V | x_1; x_2; \theta \rangle \langle x_1; x_2; \theta | A; \delta_{i}^{\otimes 2} \rangle = \sum_{j_1j_2} V(r_{j_1i}, r_{j_2i}, \hat{r}_{j_1i} \cdot \hat{r}_{j_2i}) \]

Willatt, Musil, **MC**, JCP (2019)
Understanding the range of interactions

- Representations are built for different cutoff radii
- Dimensionality/accuracy tradeoff: a measure of the range of interactions
- Multi-scale kernels $K(A, B) = \sum_i w_i K_i(A, B)$ yield the best of all worlds

Bartók, De, Poelking, Kermode, Bernstein, Csányi, **MC**, Science Advances (2017) [*data: QM9, von Lilienfeld&C*]
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![Graph showing the relationship between training data size (n. train) and MAE (MAE [kcal/mol]) for different radii (rc [Å]).](image)

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Willatt, Musil, MC, PCCP (2018)
How to learn with multiple species? Decorate atomic Gaussian with elemental kets |H\rangle, |O\rangle, . . .

Expand each ket in a finite basis, |\alpha\rangle = \sum_J u_{\alpha J} |J\rangle. Optimize coefficients

Dramatic reduction of the descriptor space, more effective learning . . .

. . . and as by-product get a data-driven version of the periodic table!
Machine-learning the periodic table

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- . . . and as by-product get a data-driven version of the periodic table!

$$
|H\rangle = 0.5 |\text{flame}\rangle + 0.1 |\text{rock}\rangle + 0.2 |\text{water}\rangle \\
|C\rangle = 0.2 |\text{flame}\rangle + 0.8 |\text{rock}\rangle + 0.3 |\text{water}\rangle \\
|O\rangle = 0.1 |\text{flame}\rangle + 0.1 |\text{rock}\rangle + 0.6 |\text{water}\rangle
$$

Empedocles et al. (ca 360BC). Metaphor courtesy of Albert Bartók
Machine-learning the periodic table

How to learn with multiple species? Decorate atomic Gaussian with elemental kets $|\text{H}\rangle$, $|\text{O}\rangle$, \ldots 

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Elpasolite dataset. Reference curve (red) from Faber et al. JCP (2018)
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Willatt, Musil, Ceriotti, PCCP (2018)
Maps, predictions and interpretation
Structure-property maps

- Representing databases of conformers, and the effect of perturbations on stability and properties
- Rationalizing structural patterns and motifs that contribute to stability

Maksimov, Baldauf, Rossi, IJQC (2020)
Structure-property maps

- Representing databases of conformers, and the effect of perturbations on stability and properties
- Rationalizing structural patterns and motifs that contribute to stability

Musil, De, Yang, Campbell, Day, **MC**, Chemical Science (2018); http://interactive.sketchmap.org
Beyond unsupervised maps

- Kernel PCA map of a dataset of carbon structures
- KPCovR reveals more clearly structure/stability relations

https://www.materialscloud.org/discover/kpcovr/carbons-10

Beyond unsupervised maps

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https://www.materialscloud.org/discover/kpcovr/carbons-05

Helfrecht, Cersonsky, Fraux, MC, MLST (2020); https://chemiscope.org
Building blocks of molecular materials

- Using data analytics to identify the “synthons” contributing to stability in molecular materials
- Correlate by construction with contributions to cohesive energy
Building blocks of molecular materials

- Using data analytics to identify the “synthons” contributing to stability in molecular materials
- Correlate by construction with contributions to cohesive energy

Cersonsky, MC, in preparation
Physical insights from knock-out ML models

- Limiting accuracy of models built on “traditional” descriptors gives objective criterion to rank their information content.
- Combination of “universal interpolators” and large datasets quantify the significance of heuristic design rules.

### (a) Volume

- **SOAP 3.5 Å**
- **SOAP 6.0 Å**
- **Distances**
- **Rings**

### (b) Energy

- **MAE (Å²/Si atom)**
- **MAE (kJ/mol Si)**

**No. Training Points**

Helfrecht et al., JCP (2019)
A look into the zeolite sorting hat

- A SVM classifier of known and hypothetical zeolites based on density correlation features
- Misclassified hypothetical structures have strong potential for synthesis
- Identifying the structural correlations that contribute most to target property by real-space projection of the SVM weights

Helfreucht et al., arxiv:2110.13764
A look into the zeolite sorting hat

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Helfrecht et al., arxiv:2110.13764
Outlook

- Physics vs data-driven modeling: porous divide at the atomic scale
- Physics-based priors: when and up to which point are they useful?
- "Interpretability" is a loose concept: what we gain and what we lose?

PHYSICS

- symmetries
- locality
- scaling/conservation laws
- training targets

DATA

- affordable accuracy
- flexibility "beyond models"
- advanced analytics

integrated ML models
multiparadigm simulations
quantitative description of emergent behavior

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Interpretable atomistic ML

Slides → tinyurl.com/ceriotti-2022-berkeley
Code: → github.com/lab-cosmo
A Dirac notation for ML

A representation maps a structure $A$ (or one environment $A_i$) to a vector discretized by a feature index $Q$.

Bra-ket notation $\langle Q | A; \text{rep.} \rangle$ indicates in an abstract way this mapping, leaving plenty of room to express the details of a representation.

Dirac-like notation reflects naturally a change of basis, the construction of a kernel, or a linear model.

$$\langle Y | A \rangle = \int dQ \langle Y | Q \rangle \langle Q | A \rangle$$

Willatt, Musil, **MC**, JCP (2019); https://tinyurl.com/dirac-rep
A Dirac notation for ML

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Dirac-like notation reflects naturally a change of basis, the construction of a kernel, or a linear model

$$k(A, A') = \langle A | A' \rangle \approx \int dQ \langle A | Q \rangle \langle Q | A' \rangle$$

Willatt, Musil, **MC**, JCP (2019); https://tinyurl.com/dirac-rep
A representation maps a structure $A$ (or one environment $A_i$) to a vector discretized by a feature index $Q$.

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Dirac-like notation reflects naturally a change of basis, the construction of a kernel, or a linear model:

$$E(A) = \langle E|A\rangle \approx \int dQ \langle E|Q\rangle \langle Q|A\rangle$$

Willatt, Musil, MC, JCP (2019); https://tinyurl.com/dirac-rep
What you ask is what you get

- Understanding what goes into a representation is key to achieve meaningful results from automated data analytics
- Example: you don’t *always* want to have rotational invariance

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![Diagram with KPCA visualizations]

Variations on a theme

- Most of the existing density-based representations and kernels emerge as special cases of this framework
  - Basis set choice - e.g. plane waves basis for $|\rho_i^{\otimes 2}\rangle$ (Ziletti et al. N.Comm 2018)
  - Projection on symmetry functions (Behler-Parrinello, DeepMD)

\[
\langle k|A; \rho^{\otimes 2}\rangle = \sum_{ij \in A} e^{ik \cdot r_{ij}}
\]

Variations on a theme

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- Basis set choice - e.g. plane waves basis for $|\rho^{\otimes 2}_i\rangle$ (Ziletti et al. N.Comm 2018)
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$$\langle abG_2 | \bar{\rho}^{\otimes 1}_i \rangle = \delta_{aa} \int dr \ G_2 (r) \langle br | \bar{\rho}^{\otimes 1}_i ; g \rightarrow \delta \rangle$$

Measuring feature spaces

- Quantitative comparison of relative information content of different features, metrics & kernels
- Feature space Reconstruction Error (FRE): linearly-embeddable mutual information

\[
\text{GFRE}(\mathcal{F} \to \mathcal{F}') = \min_{P \in \mathbb{R}^{n_F \times n_{F'}}} \| X_{\mathcal{F}'} - X_{\mathcal{F}} P \|_2
\]

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Interpretable atomistic ML

Gosciniski, Fraux, MC, MLST (2021)
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- Feature space Reconstruction Error (FRE): linearly-embeddable mutual information

\[ \text{GFRE}(F \rightarrow F') = \min_{P \in \mathbb{R}^{n_F \times n_{F'}}} \| X_{F'} - X_F P \| \]

Goscinski, Fraux, MC, MLST (2021)
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\]

\[ \text{GFRE}(\mathcal{F}',\mathcal{F}) \]

\[ 0 \quad 1 \]

\[ \mathcal{F} \cong \mathcal{F}' \quad \mathcal{F} \perp \mathcal{F}' \quad \mathcal{F} \subset \mathcal{F}' \quad \mathcal{F}' \subset \mathcal{F} \]

### Graphs

**Carbon**
- GFRE(SOAP, BPSF)
- GFRE(BPSF, SOAP)

**Random Methane**
- GFRE($\mathcal{F}',\mathcal{F}$)
- SOAP vs BPSF
- $\mathcal{F} \subseteq \mathcal{F}'$
- $\mathcal{F} \cong \mathcal{F}'$
- $\mathcal{F} \perp \mathcal{F}'$
- $\mathcal{F} \subset \mathcal{F}'$
- $\mathcal{F}' \subset \mathcal{F}$
Density expansion and SOAP

- What if we use radial functions and spherical harmonics?
- Symmetrized tensor product $\rightarrow$ SOAP power spectrum!
- Easily generalized to higher body order.
  $\delta$-distribution limit $\rightarrow$ atomic cluster expansion

\[ \langle \mathbf{r} | \rho_i \rangle = \sum_i g(\mathbf{r} - \mathbf{r}_{ij}) \]

\[ \langle nlm | \rho_i \rangle = \int d\mathbf{r} \langle \mathbf{r} | \rho_i \rangle R_n(r)Y^l_m(\hat{r}) \]

Bartók, Kondor, Csányi, PRB (2013); Willatt, Musil, MC, JCP (2019); Drautz, PRB (2019)
Density expansion and SOAP

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$\delta$-distribution limit $\rightarrow$ atomic cluster expansion

$$\langle nn'l | \rho_i^{\otimes 2} \rangle = \sum_m \langle nlm | \rho_i \rangle^* \langle n'l'm | \rho_i \rangle$$

$$P_{nn'l} = \sum_m c_{n'l'm}^* c_{n'l'm}$$

Bartók, Kondor, Csányi, PRB (2013); Willatt, Musil, MC, JCP (2019); Drautz, PRB (2019)
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$$
\langle n_1 l_1 m_1; n_2 l_2 m_2; \ldots n_{\nu} l_{\nu} m_{\nu} | \rho_i^{\otimes \nu} \rangle = \\
\int d\hat{R} \langle n_1 l_1 m_1 | \hat{R} | \rho_i \rangle \cdots \langle n_{\nu} l_{\nu} m_{\nu} | \hat{R} | \rho_i \rangle = \\
\sum_{m'_1 \ldots m'_{\nu}} \langle n_1 l_1 m'_1 | \rho_i \rangle \cdots \langle n_{\nu} l_{\nu} m'_\nu | \rho_i \rangle \\
\times \int d\hat{R} D^{l_1}_{m_1 m'_1} (\hat{R}) \cdots D^{l_{\nu}}_{m_{\nu} m'_{\nu}} (\hat{R})
$$

Bartók, Kondor, Csányi, PRB (2013); Willatt, Musil, **MC**, JCP (2019); Drautz, PRB (2019)
Two-neighbors descriptors

- Construction of a three-body ($\nu = 2$) invariant atomic descriptor
  1. Define relative position of neighbors (translation-invariant)
  2. Positions are transformed in a neighbor density (permutation invariant)
  3. Symmetrize over rotations a tensor product of the neighbor densities
  4. This is equivalent to a function of two distances and one angle
  5. $g \rightarrow \delta$ limit $\Rightarrow$ list of 2-neighbors tuples ($r_{j1i}, r_{j2i}, \hat{r}_{j1i} \cdot \hat{r}_{j2i}$)
  6. Linear model $\Rightarrow$ 3-body potential!

\[ \{ r_{jii} = r_j - r_i \} \leftrightarrow A_i \]

Bartók, Kondor, Csányi, PRB (2013)
Two-neighbors descriptors

Construction of a three-body ($\nu = 2$) invariant atomic descriptor

1. Define relative position of neighbors (*translation-invariant*).
2. Positions are transformed in a neighbor density (*permutation invariant*).
3. Symmetrize over rotations a tensor product of the neighbor densities.
4. This is equivalent to a function of two distances and one angle.
5. $g \rightarrow \delta$ limit $\Rightarrow$ list of 2-neighbors tuples ($r_{j1i}, r_{j2i}, \hat{r}_{j1i} \cdot \hat{r}_{j2i}$).
6. Linear model $\Rightarrow 3$-body potential!

$$
\langle a\mathbf{x} | \rho_i \rangle = \sum_{j \in A_i} \delta_{aa_j} \langle \mathbf{x} | \mathbf{r}_{ji} ; g \rangle \\
\langle \mathbf{x} | \mathbf{r}_{ji} ; g \rangle \equiv g(\mathbf{x} - \mathbf{r}_{ji})
$$

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\[
\langle x; x' | A; \rho_i \otimes 2 \rangle = \int d\hat{R} \langle x | \hat{R}A; \rho_i \rangle \langle x' | \hat{R}A; \rho_i \rangle
\]

Willatt, Musil, MC, JCP (2019)
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\[ \langle x_1; x_2; \theta | A; \rho_i^\otimes 2 \rangle = \int d\hat{R} \langle x_1 \hat{R}\hat{e}_z | A; \rho_i \rangle \]
\[ \langle x_2 \hat{R}(\hat{e}_z \cos \theta + \hat{e}_x \sin \theta) | A; \rho_i \rangle \]

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$$\langle x_1; x_2; \theta | A; \delta_i \otimes 2 \rangle = \sum_{j_1j_2} \delta(x_1 - r_{j_1i}) \delta(x_2 - r_{j_2i}) \delta(\cos \theta - \hat{r}_{j_1i} \cdot \hat{r}_{j_2i})$$

Willatt, Musil, MC, JCP (2019)
Two-neighbors descriptors

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  1. Define relative position of neighbors (translation-invariant)
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\[
\int \langle V | x_1; x_2; \theta \rangle \langle x_1; x_2; \theta | A; \delta_i^{\otimes 2} \rangle = \sum_{j_1j_2} V(r_{j_1i}, r_{j_2i}, \hat{r}_{j_1i} \cdot \hat{r}_{j_2i})
\]
Density trick in an $\langle nlm \rangle$ basis

- The symmetrized correlations can be computed in closed form using a discrete basis
  - The neighbor density can be expanded on a basis of radial functions $\langle x|n \rangle \equiv R_n(x)$ and spherical harmonics $\langle \hat{x}|lm \rangle \equiv Y_l^m(\hat{x})$
  - Spherical harmonics transform linearly under rotations based on Wigner rotation matrices $D^l(\hat{R})$
  - Orthogonality of Wigner matrices yields the SOAP powerspectrum

$$\langle \hat{x}|lm \rangle$$

$$\langle x|n \rangle$$

$$\langle nlm|A; \rho_i \rangle = \int dx \langle n|x \rangle \langle lm|\hat{x} \rangle \langle x|A; \rho_i \rangle$$
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\[
\hat{R}|lm\rangle = \sum_{m'} D^l_{mm'}(\hat{R})|lm'\rangle
\]

\[
\langle nlm; n'l'm'|A; \rho^\otimes 2 \rangle = \int d\hat{R} \langle nlm|\hat{RA}; \rho_i \rangle \langle n'l'm'|\hat{RA}; \rho_i \rangle
\]
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\[
\int d\hat{R} \sum_{kk'} D^l_m(\hat{R}) D^{l'}_{m'}(\hat{R}) \propto \delta_{ll'} \delta_{mm'} \delta_{kk'}
\]

\[
\langle nn'l|A; \rho_i \otimes 2 \rangle = \sum_m \langle nlm|A; \rho_i \rangle \langle n'lm|A; \rho_i \rangle
\]
A hierarchy of equivariant features

- A generalization of the definition yields $N$-body features that transform like angular momenta
  \[ \langle X|\rho_i^\otimes\nu;\sigma;\lambda\mu \rangle \]

- Recursive construction based on sums of angular momenta and an expansion of the atom density
  \[ \langle n_1 l_1 k_1|\rho_i^\otimes1;\lambda\mu \rangle \equiv \langle n_1 \lambda (-\mu)|\rho_i \rangle \delta_{l_1 \lambda} \delta_{k_1 \lambda} \delta_{\sigma 1} \equiv \langle n_1|\rho_i^\otimes1;\lambda\mu \rangle \]

  \[ \langle \ldots; n_\nu l_\nu k_\nu; n lk|\rho_i^\otimes(\nu+1);\sigma;\lambda\mu \rangle = \delta_{\sigma((-1)^{l+k+\lambda s})} c_{k\lambda} \times \]

  \[ \sum_{qm} \langle lm; kq|\lambda\mu \rangle \langle n|\rho_i^\otimes1; lm \rangle \langle \ldots; n_\nu l_\nu k_\nu|\rho_i^\otimes\nu; s; kq \rangle \]

- Can be used to compute efficiently invariant features \[ |\rho_i^\otimes\nu; 0; 00 \rangle \]

Nigam, Pozdnyakov, MC, JCP (2020)
NICE features for ML

- Problem: number of features grows exponentially with $\nu$
- Solution: an $N$-body iterative contraction of equivariants (NICE) framework
  - After each body order increase, the most relevant features are selected and used for the next iteration
  - Systematic convergence with $\nu$ and contraction truncation

\[ \langle n | \rho_i^{\otimes^1 lm} \rangle \]

\[ \langle N_\nu | \rho_i^{\otimes^\nu kq} \rangle \]

\[ \langle nlk | \lambda \mu \sigma \rangle \]

\[ \langle N_\nu | \rangle \]

\[ \langle N_\nu; nlk | \rho_i^{\otimes^\nu \lambda \mu} \rangle \]

\[ \langle N_\nu + 1 | \rho_i^{\otimes^\nu + 1 \lambda \mu} \rangle \]

Nigam, Pozdnyakov, MC, JCP (2020)
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![Graph showing NICE performance with different $\nu$ values and contraction orders.](image-url)

Nigam, Pozdnyakov, MC, JCP (2020)
Machine-learning the periodic table

- How to learn with multiple species? Decorate atomic Gaussian with elemental kets $|H\rangle$, $|O\rangle$, $|\ldots\rangle$
- Expand each ket in a finite basis, $|\alpha\rangle = \sum_J u_{\alpha J} |J\rangle$. Optimize coefficients
- Dramatic reduction of the descriptor space, more effective learning $\ldots$
- $\ldots$ and as by-product get a data-driven version of the periodic table!
How to learn with multiple species? Decorate atomic Gaussian with elemental kets $|H\rangle$, $|O\rangle$, . . .

Expand each ket in a finite basis, $|\alpha\rangle = \sum_J u_{\alpha J} |J\rangle$. Optimize coefficients

Dramatic reduction of the descriptor space, more effective learning . . .

. . . and as by-product get a data-driven version of the periodic table!

$$
\begin{align*}
|H\rangle &= 0.5 |\text{\textcolor{red}{\H}}\rangle + 0.1 |\text{\textcolor{black}{\O}}\rangle + 0.2 |\text{\textcolor{blue}{\O}}\rangle \\
|C\rangle &= 0.2 |\text{\textcolor{red}{\H}}\rangle + 0.8 |\text{\textcolor{black}{\O}}\rangle + 0.3 |\text{\textcolor{blue}{\O}}\rangle \\
|O\rangle &= 0.1 |\text{\textcolor{red}{\H}}\rangle + 0.1 |\text{\textcolor{black}{\O}}\rangle + 0.6 |\text{\textcolor{blue}{\O}}\rangle
\end{align*}
$$

Empedocles et al. (ca 360BC). Metaphor courtesy of Albert Bartók
Machine-learning the periodic table

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<table>
<thead>
<tr>
<th>Number of training structures</th>
<th>Test MAE (eV / atom)</th>
</tr>
</thead>
<tbody>
<tr>
<td>250</td>
<td>Reference</td>
</tr>
<tr>
<td>500</td>
<td>$d_j = 2$</td>
</tr>
<tr>
<td>1k</td>
<td>$d_j = 1$</td>
</tr>
<tr>
<td>3k</td>
<td>$d_j = 4$</td>
</tr>
<tr>
<td>6k</td>
<td>Multi-kernel</td>
</tr>
</tbody>
</table>

Elpasolite dataset. Reference curve (red) from Faber et al. JCP (2018)
How to learn with multiple species? Decorate atomic Gaussian with elemental kets $|\text{H}\rangle$, $|\text{O}\rangle$, $\ldots$

Expand each ket in a finite basis, $|\alpha\rangle = \sum_J u_{\alpha J} |J\rangle$. Optimize coefficients.

Dramatic reduction of the descriptor space, more effective learning $\ldots$

$\ldots$ and as by-product get a data-driven version of the periodic table!

Willatt, Musil, Ceriotti, PCCP (2018)
Recognizing active protein ligands

- A SOAP-REMatch-based KSVM classifies active and inactive ligands with 99% accuracy; non-additive model is crucial!
- Sensitivity analysis helps identify the active “warhead” and could guide drug design and optimization

Structure-property landscapes

- Clustering/sketch-maps based on REMatch-SOAP correlate well with qualitative classification of packing motifs, and with properties (ex.: azapentacene structure-energy-property landscape maps)

Musil, De, Yang, Campbell, Day, MC, Chemical Science (2018); http://interactive.sketchmap.org
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Musil, De, Yang, Campbell, Day, MC, Chemical Science (2018); http://interactive.sketchmap.org
Principal Covariates Regression

- Very simple idea to combine PCA and latent-space LR to find a dimensionality reduction that preserves variance and predicts well

\[
\ell = \alpha \|X - XP_{XT}P_{TX}\|^2 + (1 - \alpha) \|Y - XP_{XT}P_{TY}\|^2
\]

- Solution can be found working in sample space (looking for the eigenvectors of a modified Gram matrix)

\[
\tilde{K} = \alpha XX^T + (1 - \alpha) XP_{XY}P_{XY}^T X^T
\]

- ... or in feature space by diagonalizing a modified covariance

\[
\tilde{C} = \alpha X^TX + (1 - \alpha) (X^TX)^{-1/2} X^TYY^TX (X^TX)^{-1/2}
\]

S. de Jong and HAL Kiers, Scandinavian Symposium on Chemometrics (1992)
Kernel PCovR

Kernel versions of PCovR can be obtained with a modified kernel
\[
\tilde{K} = \alpha K + (1 - \alpha) \hat{Y}\hat{Y}^T,
\]
diagonalizing it and finding the projector
\[
P_{K_T} = \left( \alpha I + (1 - \alpha) (K + \lambda I)^{-1} \hat{Y}\hat{Y} \right) U_{\tilde{K}} \Lambda_{\tilde{K}}^{1/2}
\]
Where unsupervised meets supervised

- Using KPCovR to reveal structure-property relations in databases of materials structures

Helfrecht, Cersonsky, Fraux, **MC**, MLST (2020)
A Generalized Convex Hull Construction