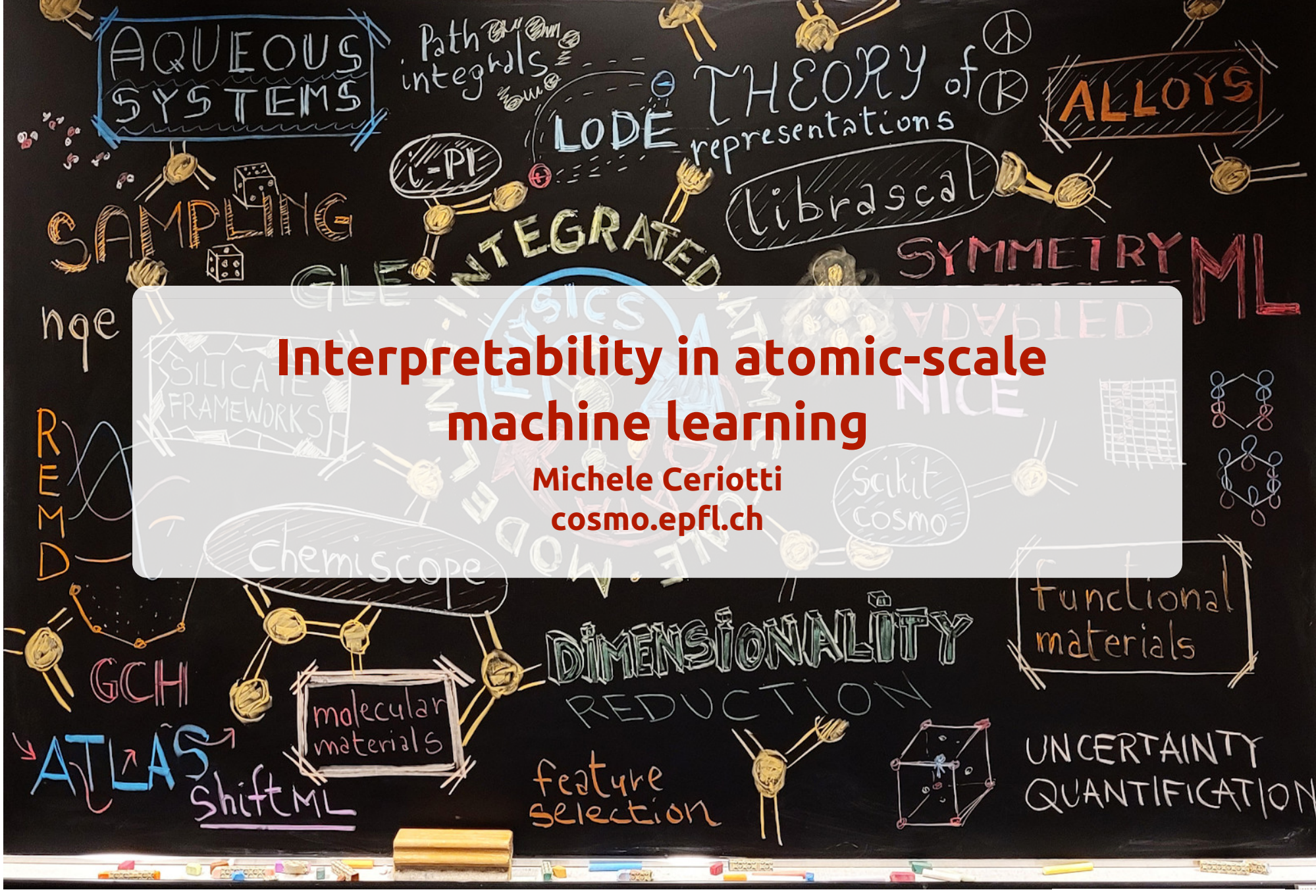

Interpretability in atomic-scale machine learning

Michele Ceriotti
cosmo.epfl.ch

Berkeley, June 2022

Computational science & modeling @ EPFL

cosmo.epfl.ch

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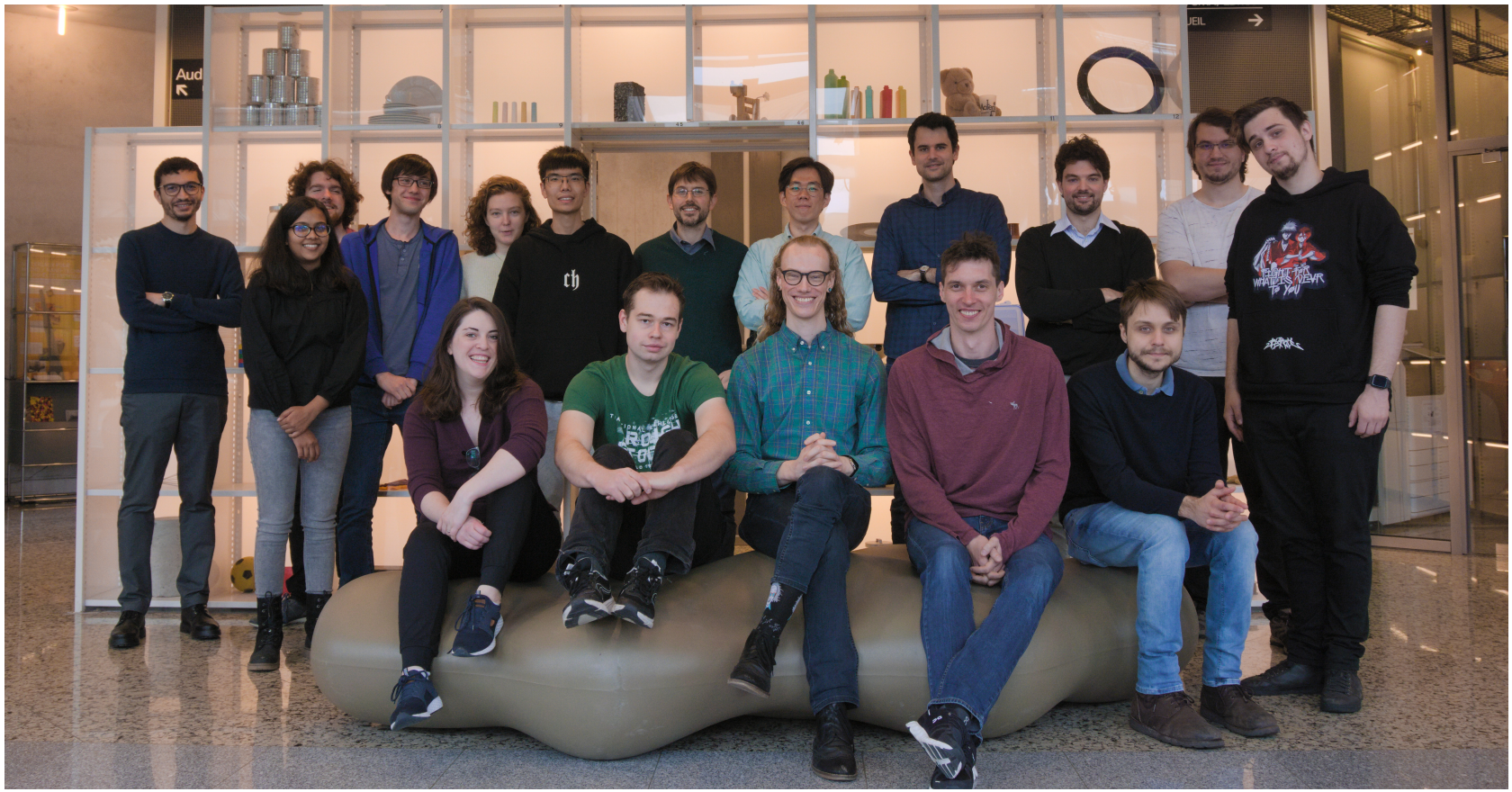


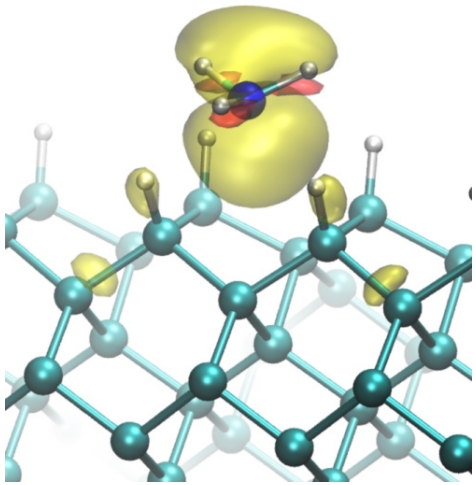
Machine learning in atomic-scale simulations

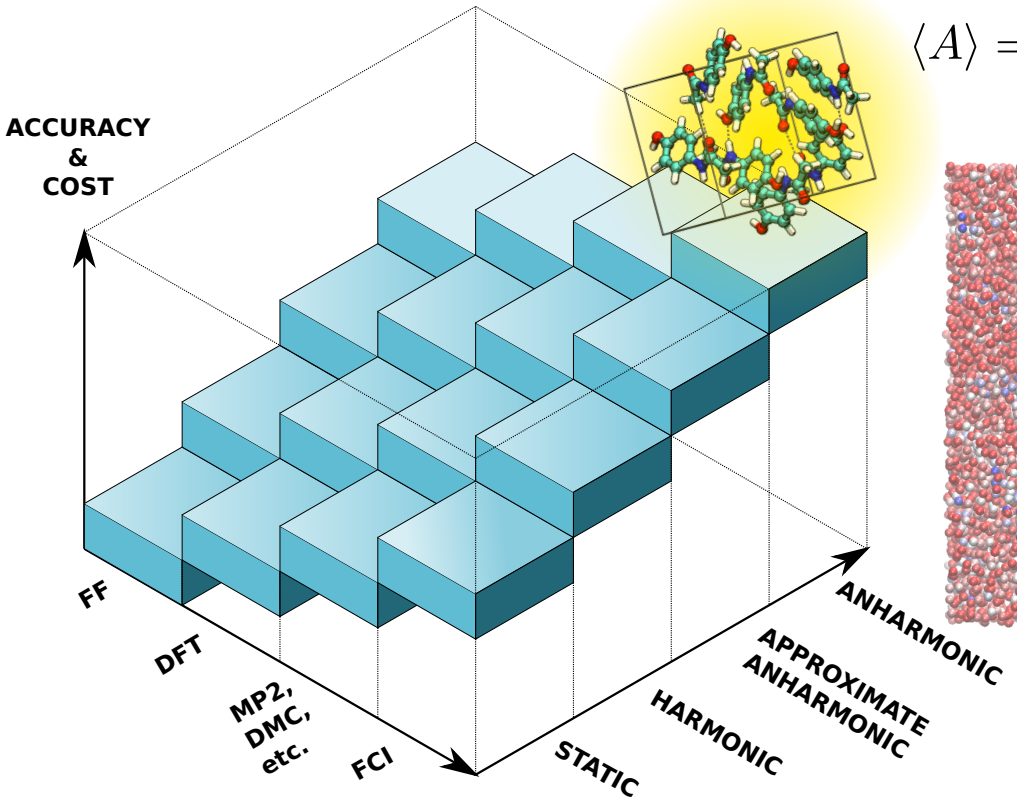
Predictive accuracy for real materials

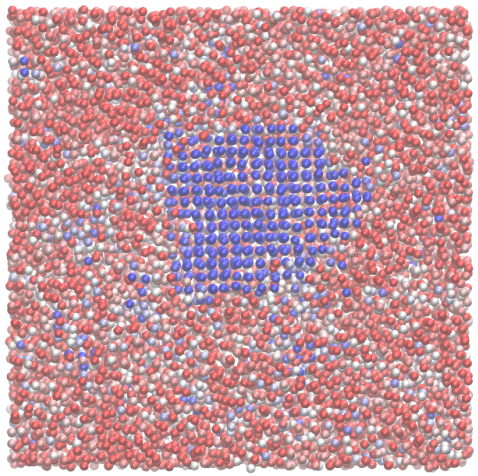
- First-principles calculations promise quantitatively accurate simulations that make no use of experimental data
- Emergent physics from first principles: still a tremendous challenge
- Machine learning to the rescue

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

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







ACCURACY OF ENERGETICS

ACCURACY OF SAMPLING

Grabowski et al., PRB (2009); Kapil, Engel, Rossi, **MC**, JCTC (2019)

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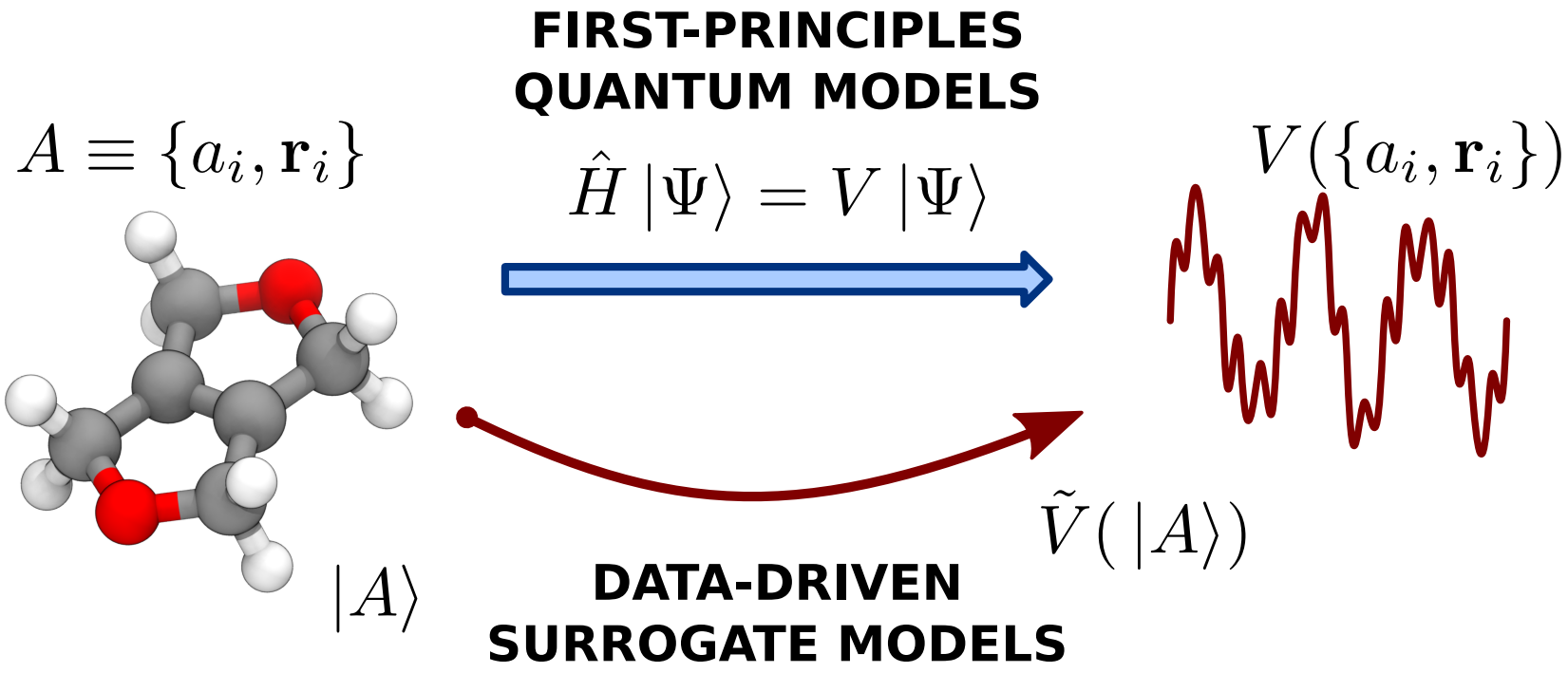

Musil et al., Chem. Rev. (2021)

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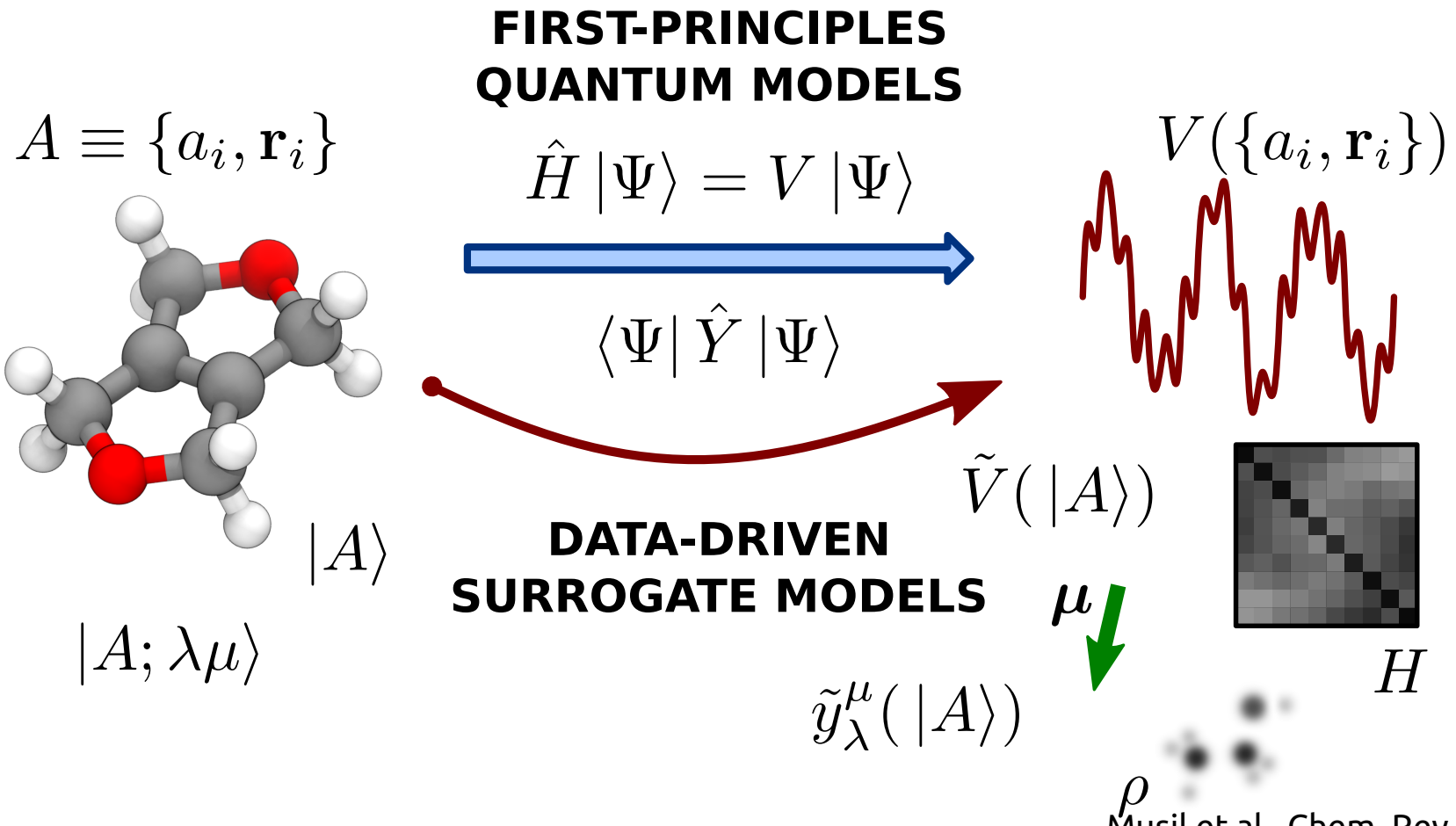
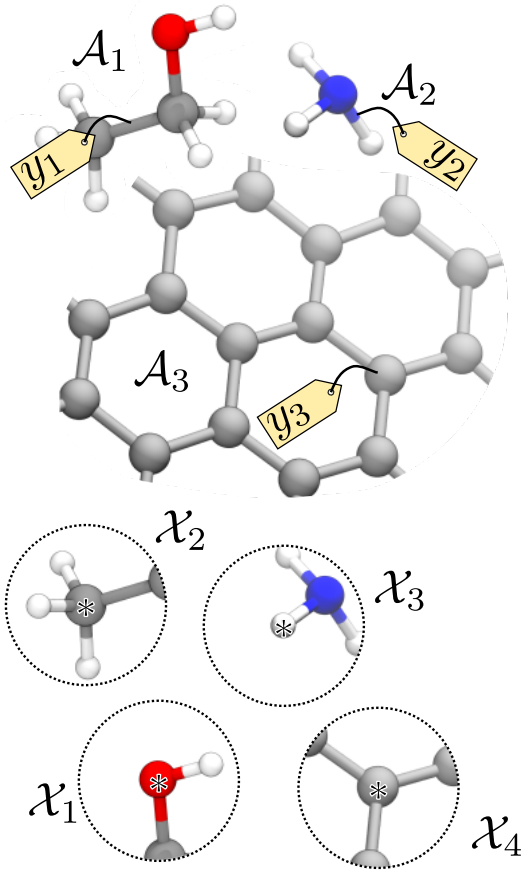
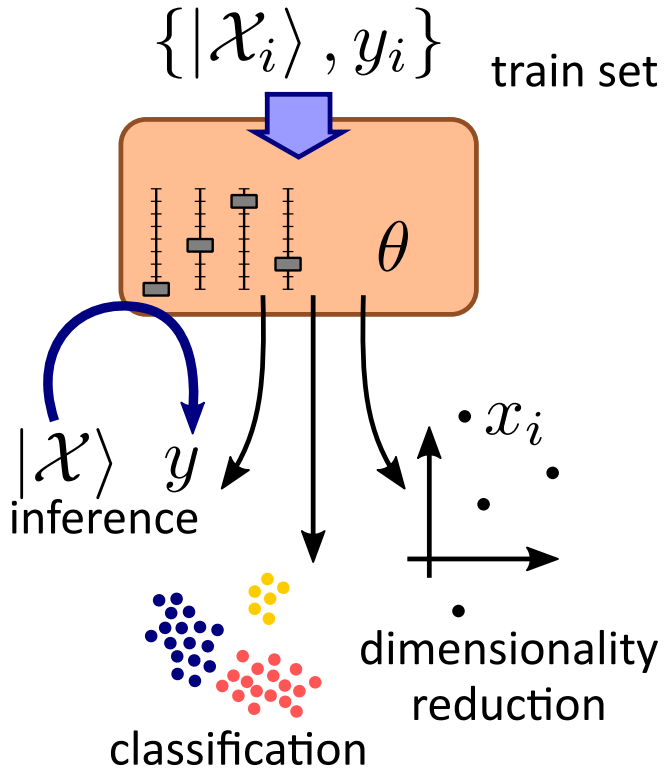


Musil et al., Chem. Rev. (2021)

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





MC, JCP (2019)

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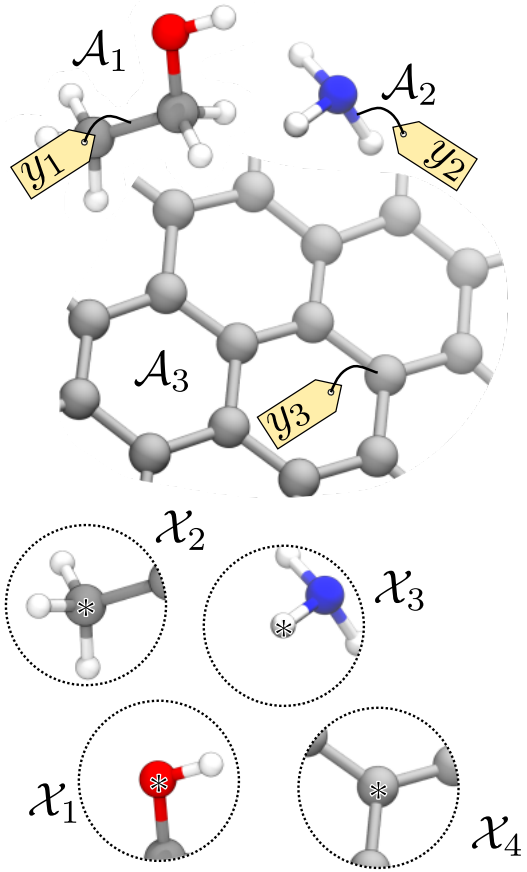
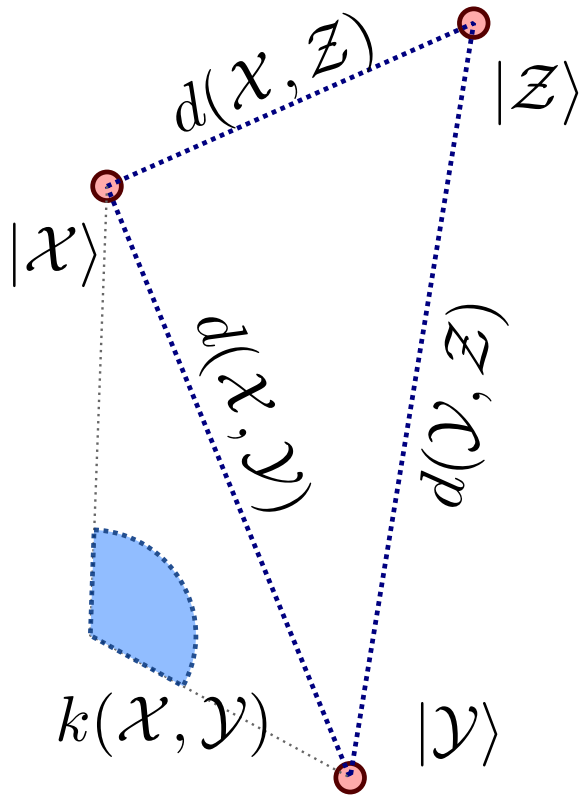
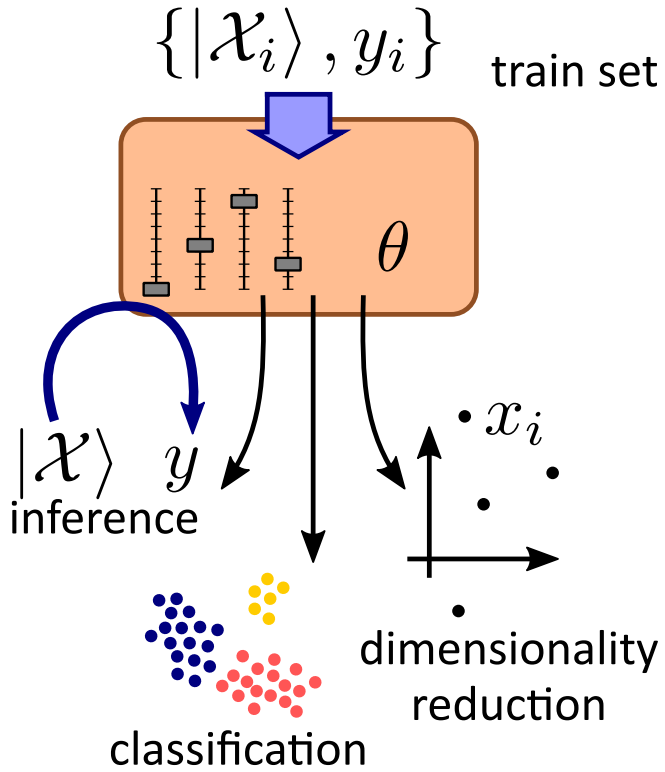




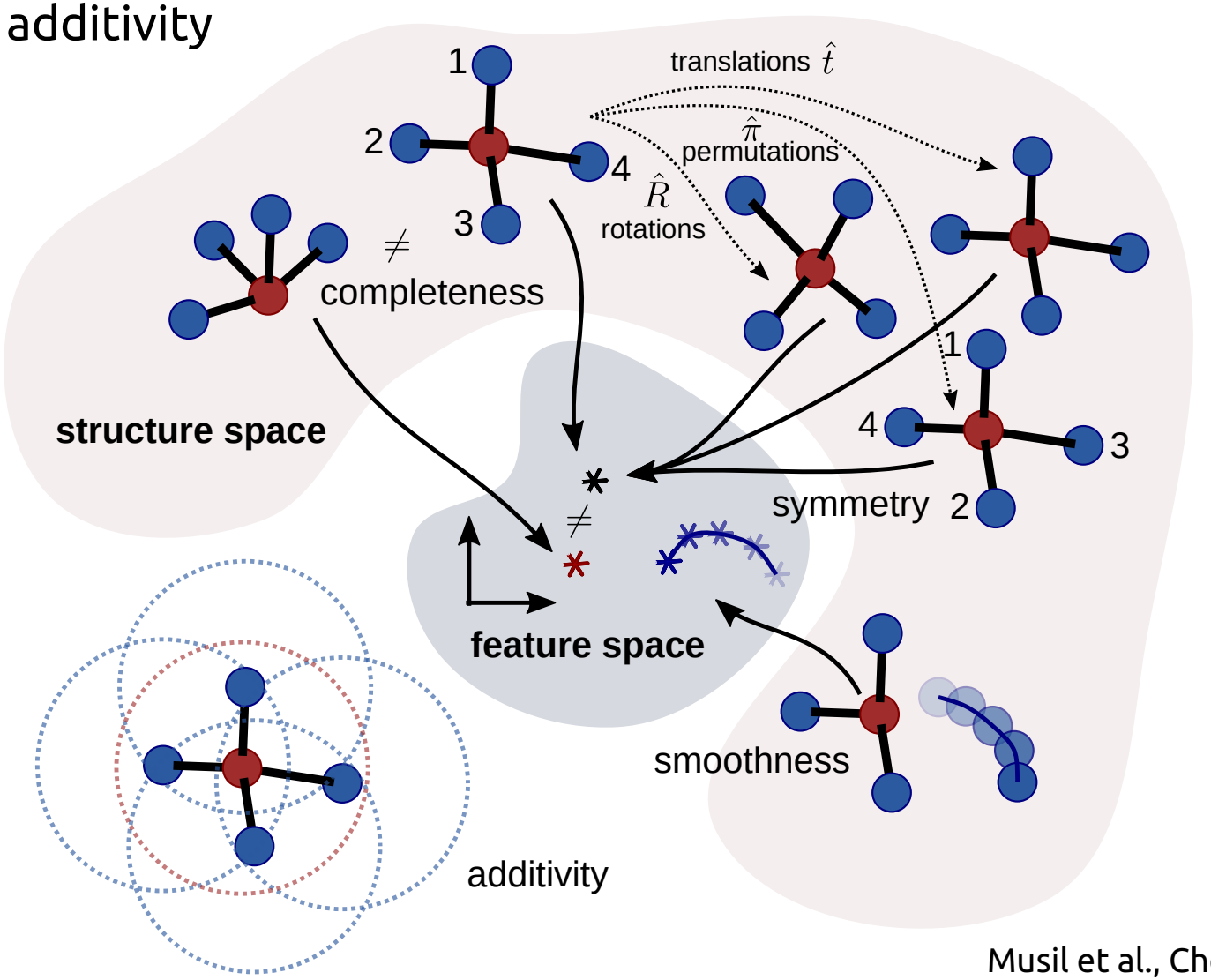


MC, JCP (2019)

Interpretable structural representations

What do we want from a representation?

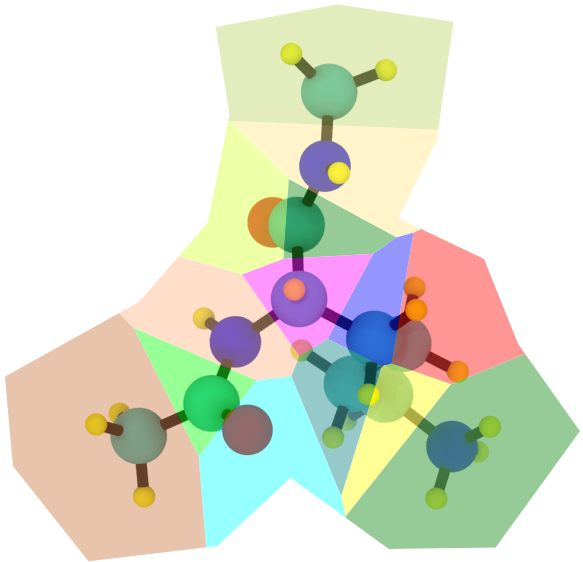
1. Be complete (injective)
2. Reflect basic physical symmetries
3. Be smooth, regular
4. Exploit additivity

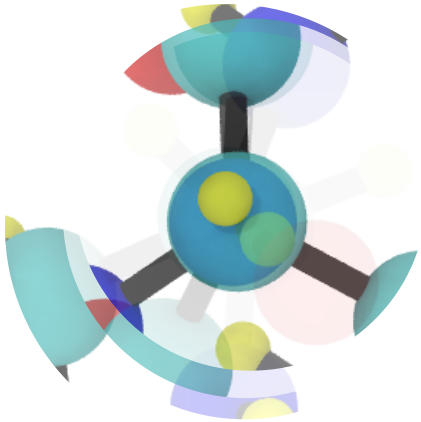


Musil et al., Chem. Rev. (2021)

Additivity, and locality

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





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

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

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

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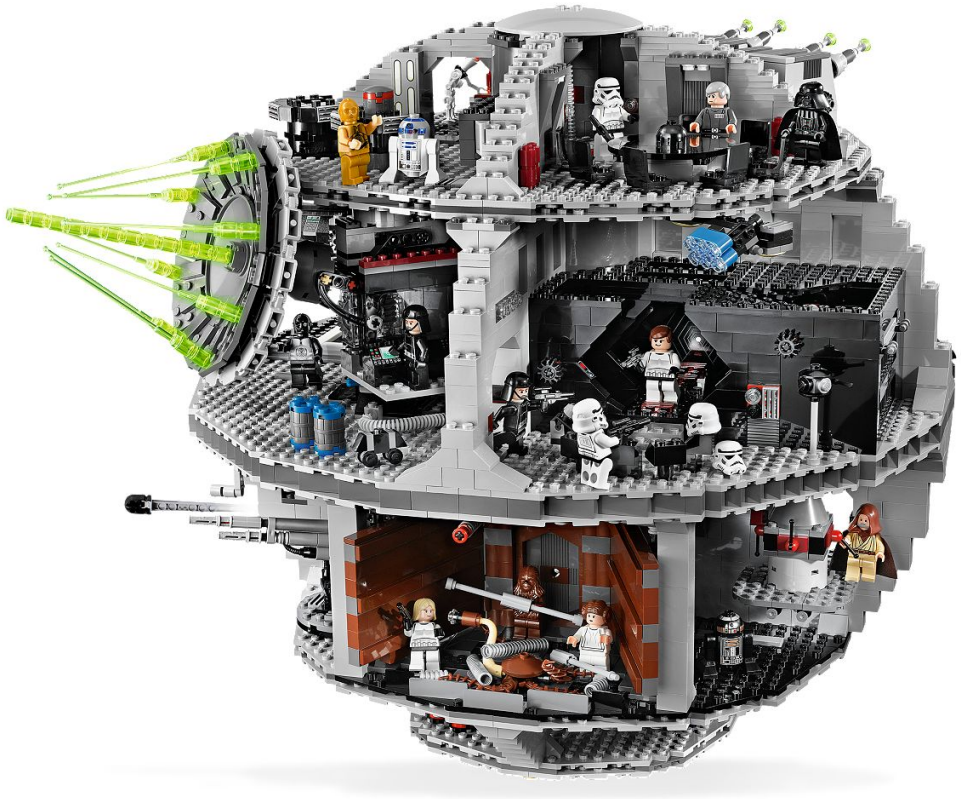
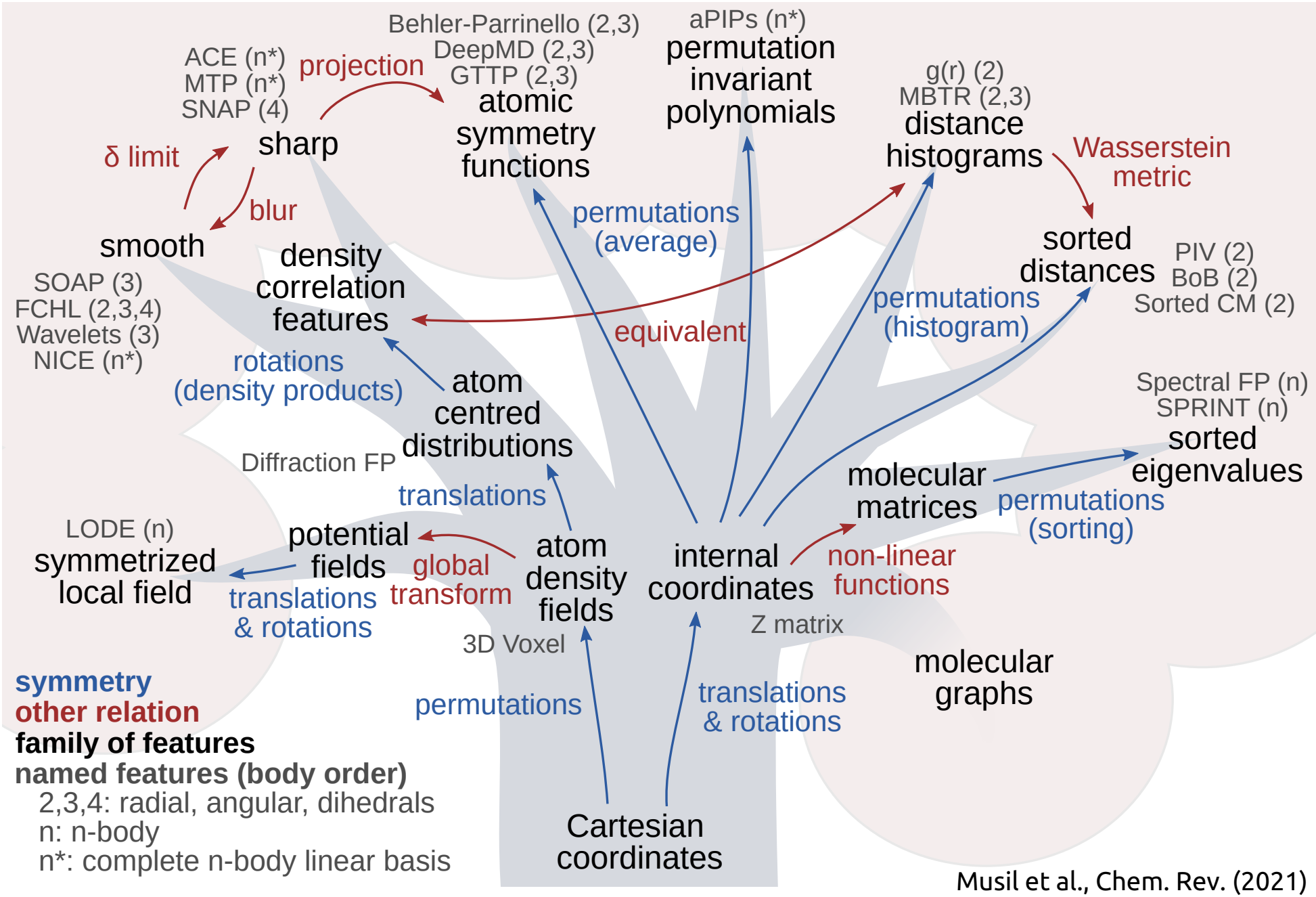


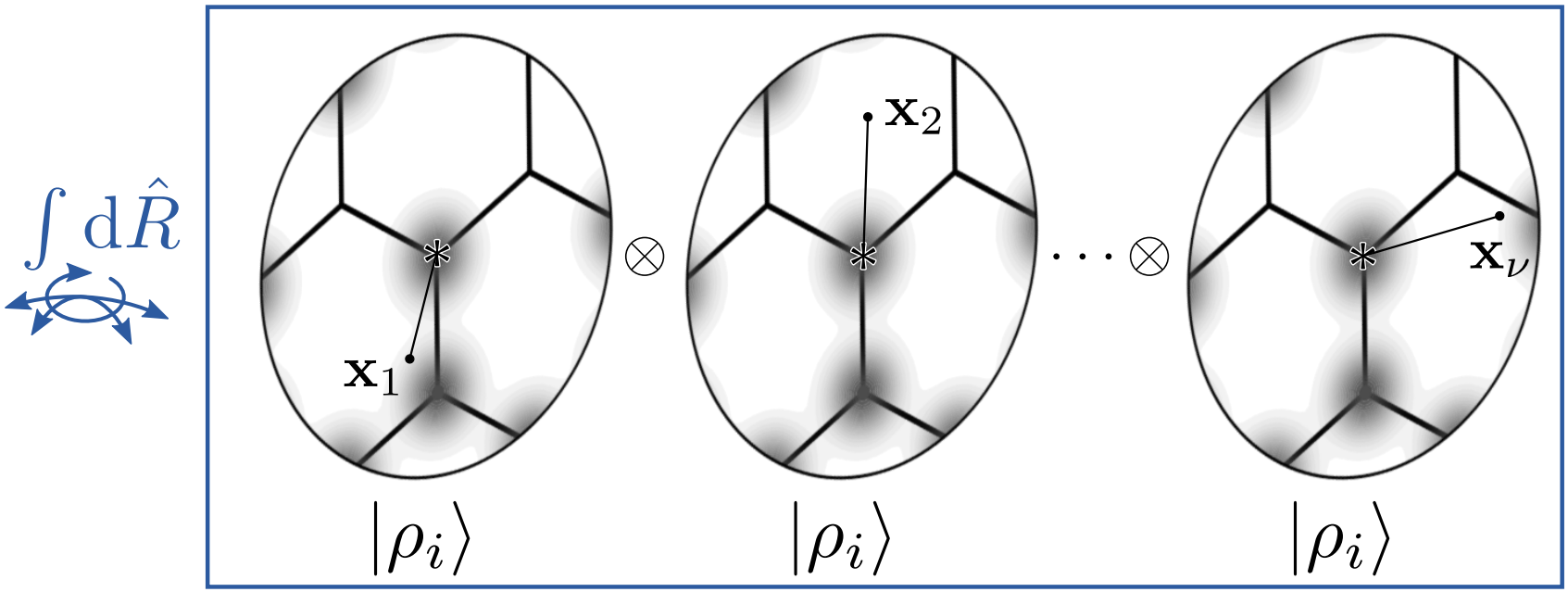
A phylogenetic tree of ML representations



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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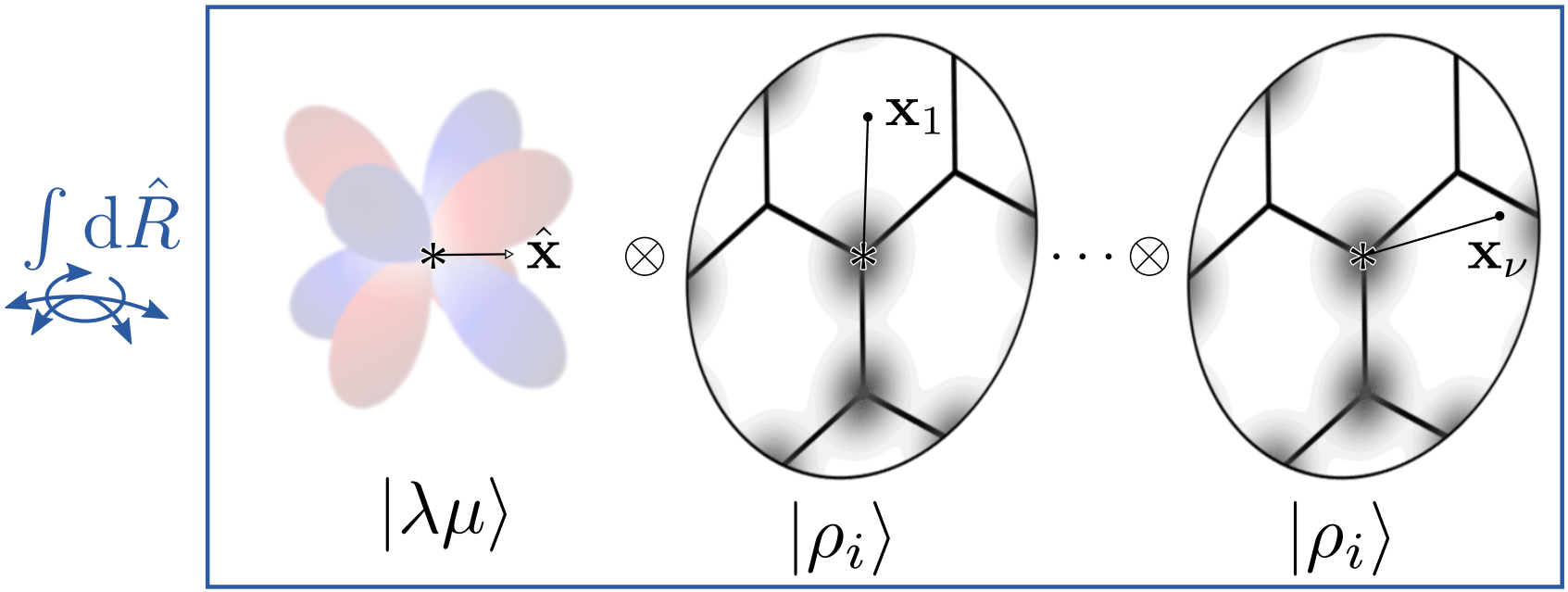

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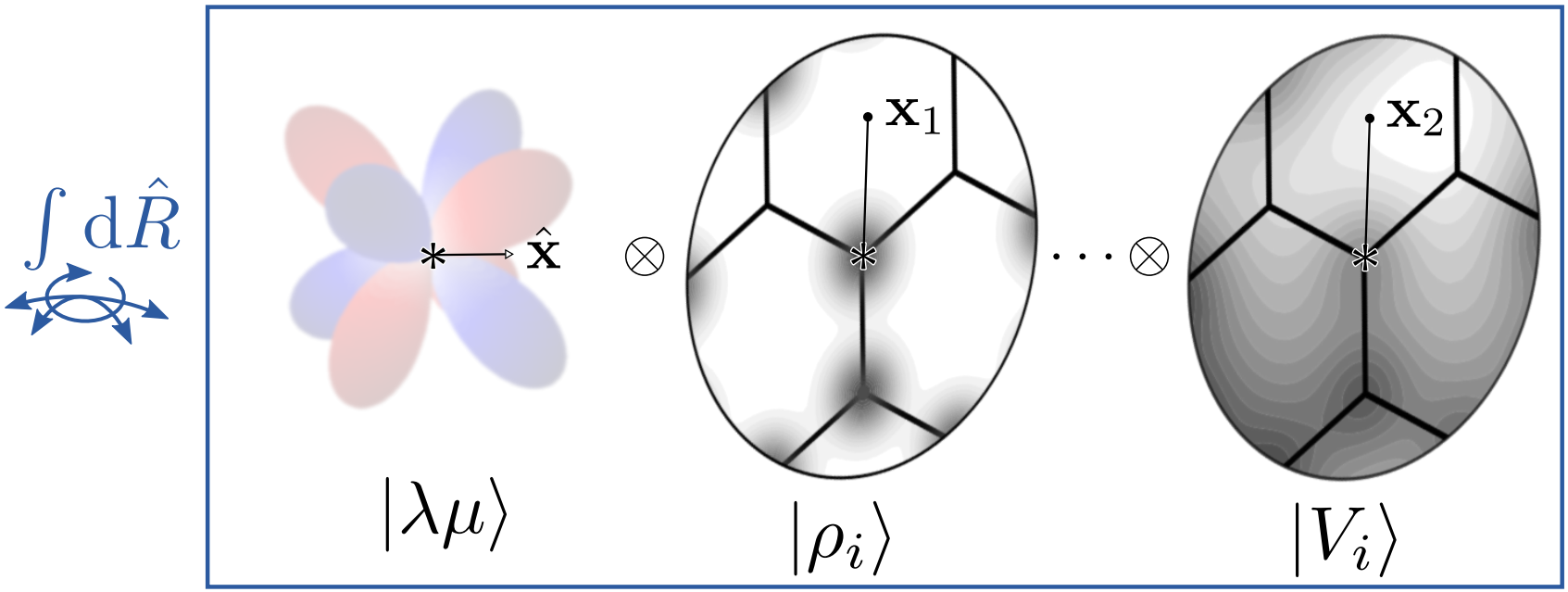

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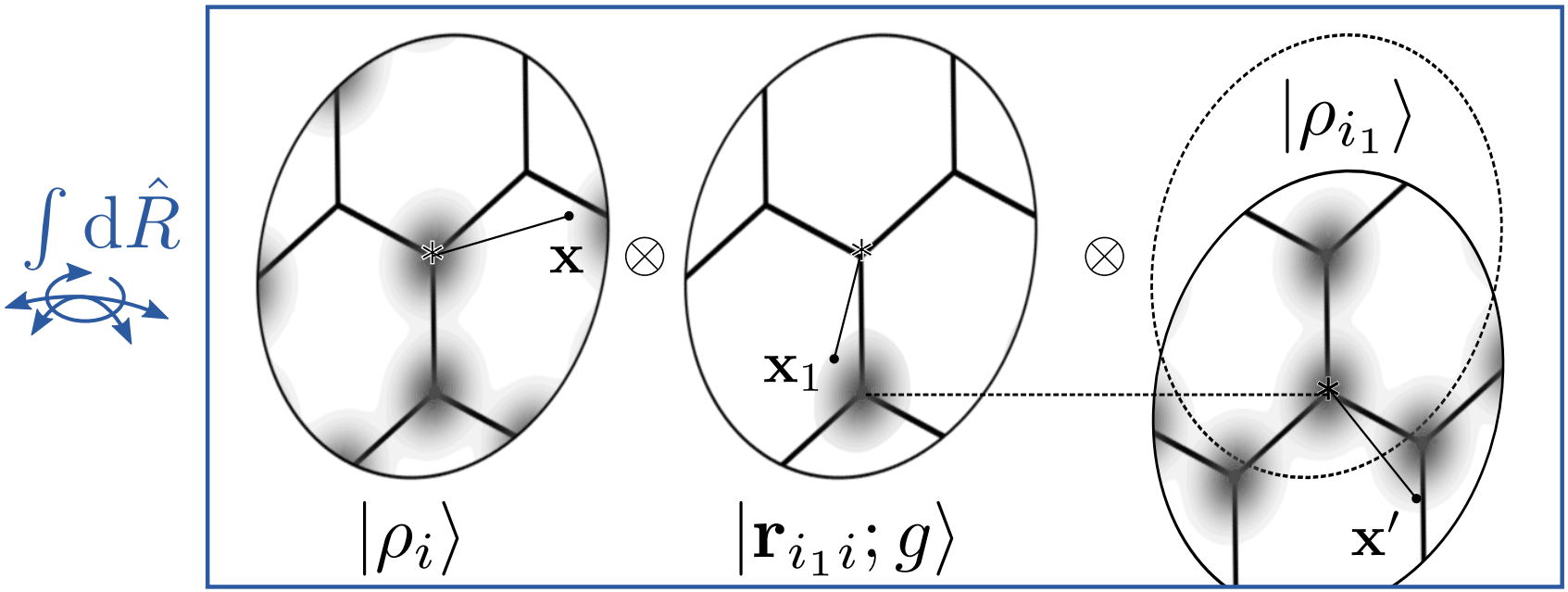


Latest & greatest: Nigam, Pozdnyakov, Fraux, **MC**, JCP (2022)

A hierarchy of equivariant features

- Neighbor density can be expanded in radial functions and $Y_l^m \rightarrow$ equivariant features

$$\langle n_1 | \overline{\rho_i^{\otimes 1}; \lambda\mu} \rangle \equiv \langle n_1 \lambda\, (-\mu) | \rho_i \rangle$$

- Recursive constructiono of N-body features based on sums of angular momenta

$$\langle \ldots; n_\nu l_\nu k_\nu; nlk | \overline{\rho_i^{\otimes(\nu+1)}; \lambda\mu} \rangle = \sum_{qm} \langle n | \overline{\rho_i^{\otimes 1}; lm} \rangle \, \langle \ldots; n_\nu l_\nu k_\nu | \overline{\rho_i^{\otimes \nu}; kq} \rangle \, \langle lm; kq | \lambda\mu \rangle$$

- All equivariant ν-neighbor features transform like angular momenta

$$\langle q | \hat{R} A; \overline{\rho_i^{\otimes \nu}; \lambda\mu} \rangle \sim \sum_{\mu'} D^\lambda_{\mu\mu'}(R) \, \langle q | A; \overline{\rho_i^{\otimes \nu}; \lambda\mu'} \rangle$$

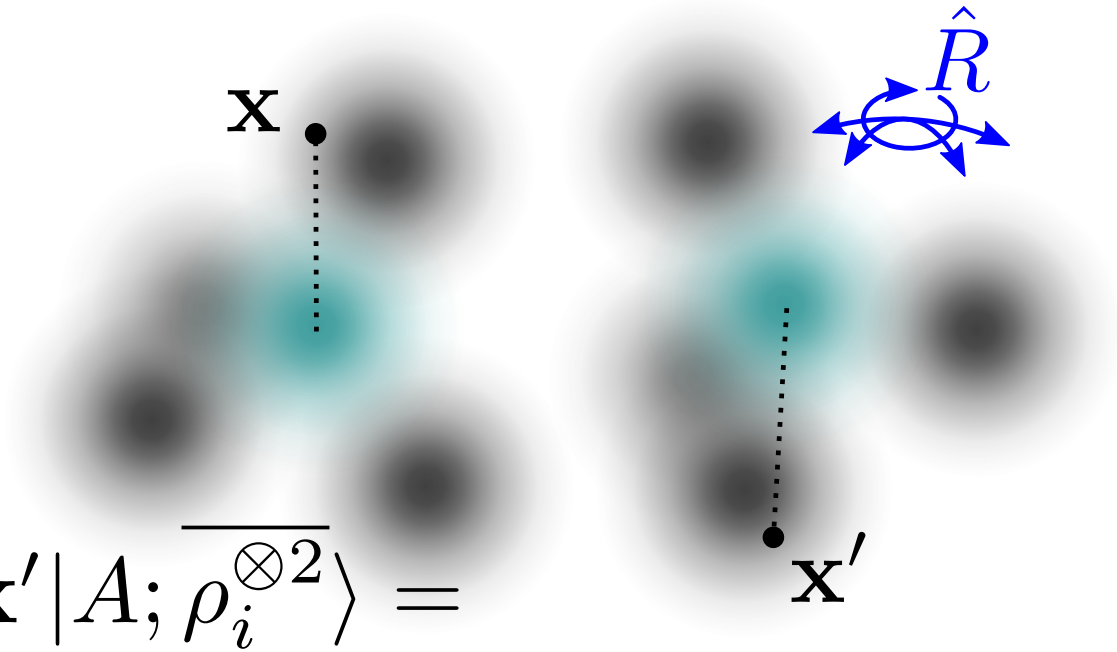
Can be used to compute efficiently *invariant* features $|\overline{\rho_i^{\otimes \nu}; 00}\rangle$

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 $(r_{j_1 i}, r_{j_2 i}, \hat{\mathbf{r}}_{j_1 i} \cdot \hat{\mathbf{r}}_{j_2 i})$
  4. Linear model $\rightarrow$ 3-body potential!



$$\langle \mathbf{x}; \mathbf{x}' | A; \overline{\rho_i^{\otimes 2}} \rangle = \int \mathrm{d}\hat{R} \langle \mathbf{x} | \hat{R}A; \rho_i \rangle \langle \mathbf{x}' | \hat{R}A; \rho_i \rangle$$

Willatt, Musil, **MC**, JCP (2019)

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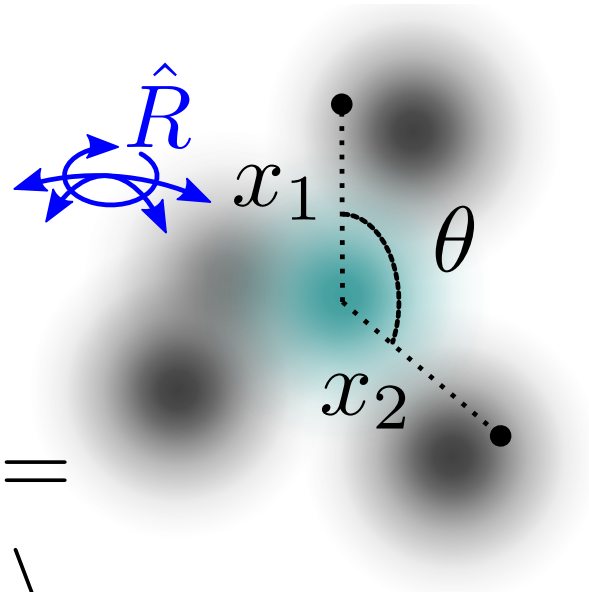

$$\langle x_1; x_2; \theta | A; \overline{\rho_i^{\otimes 2}} \rangle =$$
$$\int \mathrm{d}\hat{R} \langle x_1 \hat{R} \hat{\mathbf{e}}_z | A; \rho_i \rangle$$
$$\langle x_2 \hat{R} (\hat{\mathbf{e}}_z \cos\theta + \hat{\mathbf{e}}_x \sin\theta) | A; \rho_i \rangle$$

Willatt, Musil, **MC**, JCP (2019)

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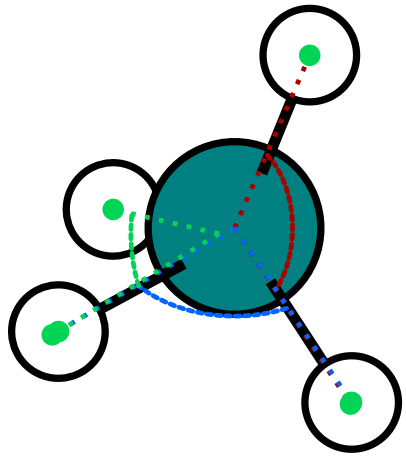
$$\langle x_1; x_2; \theta | A; \overline{\delta_i^{\otimes 2}} \rangle = \sum_{j_1 j_2} \delta(x_1 - r_{j_1 i}) \delta(x_2 - r_{j_2 i}) \delta(\cos\theta - \hat{\mathbf{r}}_{j_1 i} \cdot \hat{\mathbf{r}}_{j_2 i})$$



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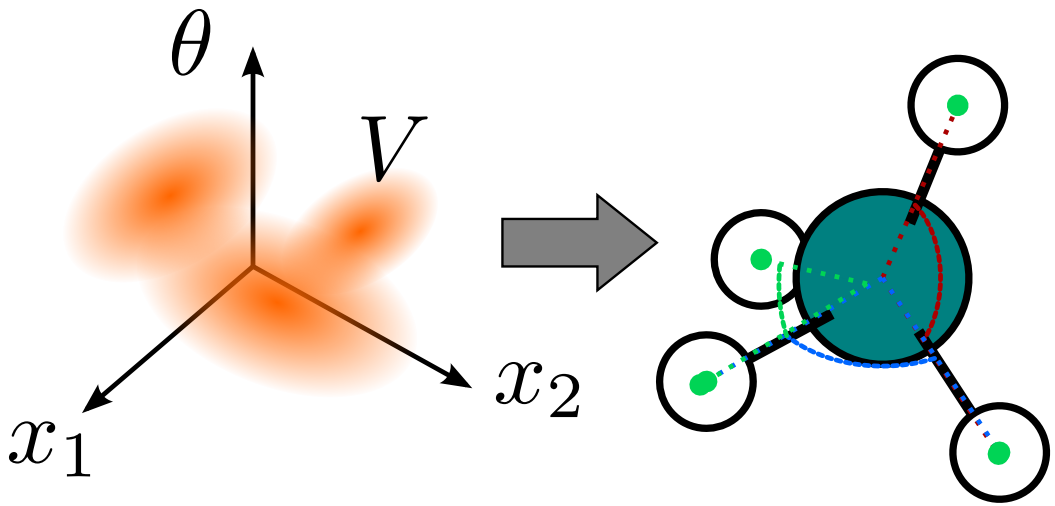
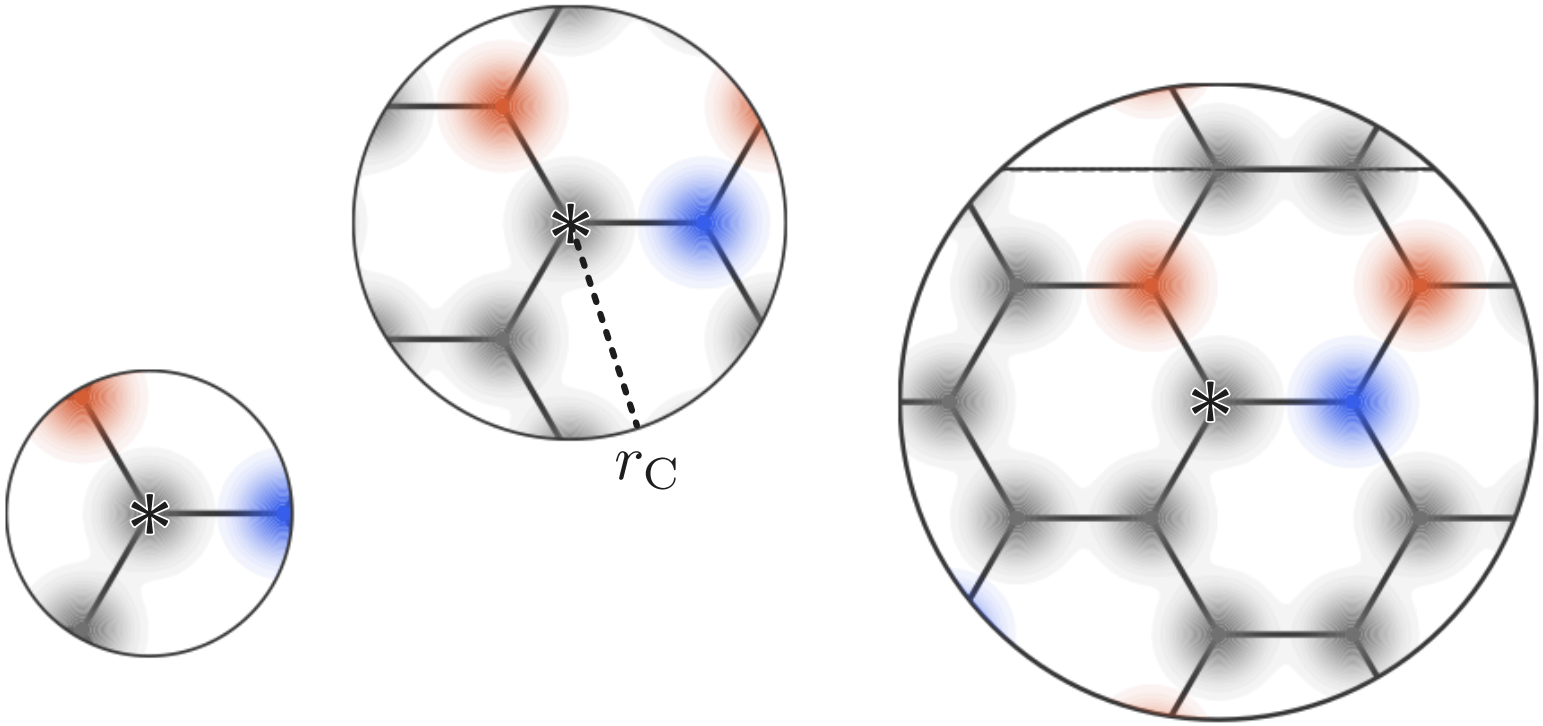


$$\int \langle V | x_1; x_2; \theta \rangle \langle x_1; x_2; \theta | A; \overline{\delta_i^{\otimes 2}} \rangle = \sum_{j_1 j_2} V(r_{j_1 i}, r_{j_2 i}, \hat{\mathbf{r}}_{j_1 i} \cdot \hat{\mathbf{r}}_{j_2 i})$$

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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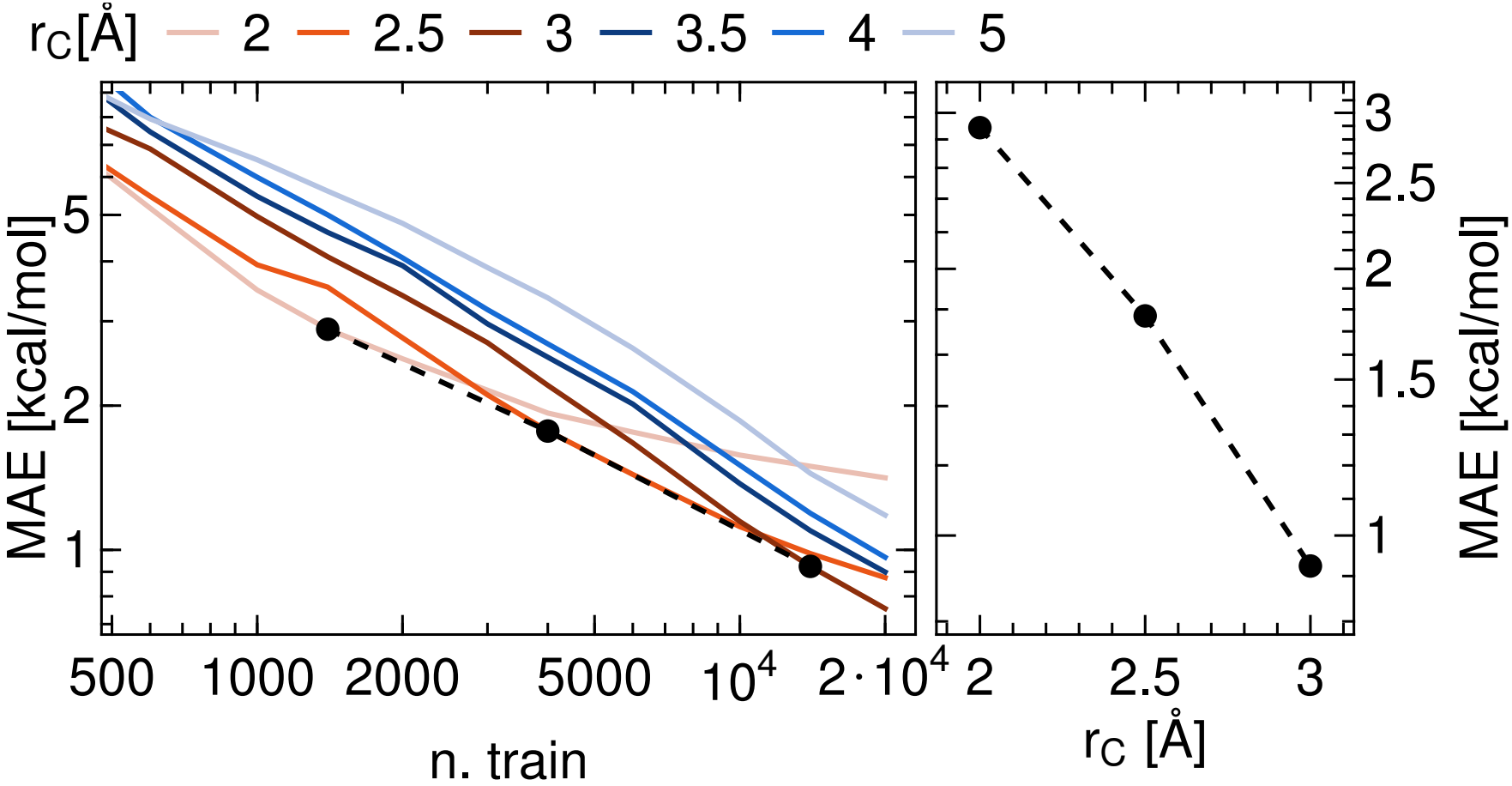

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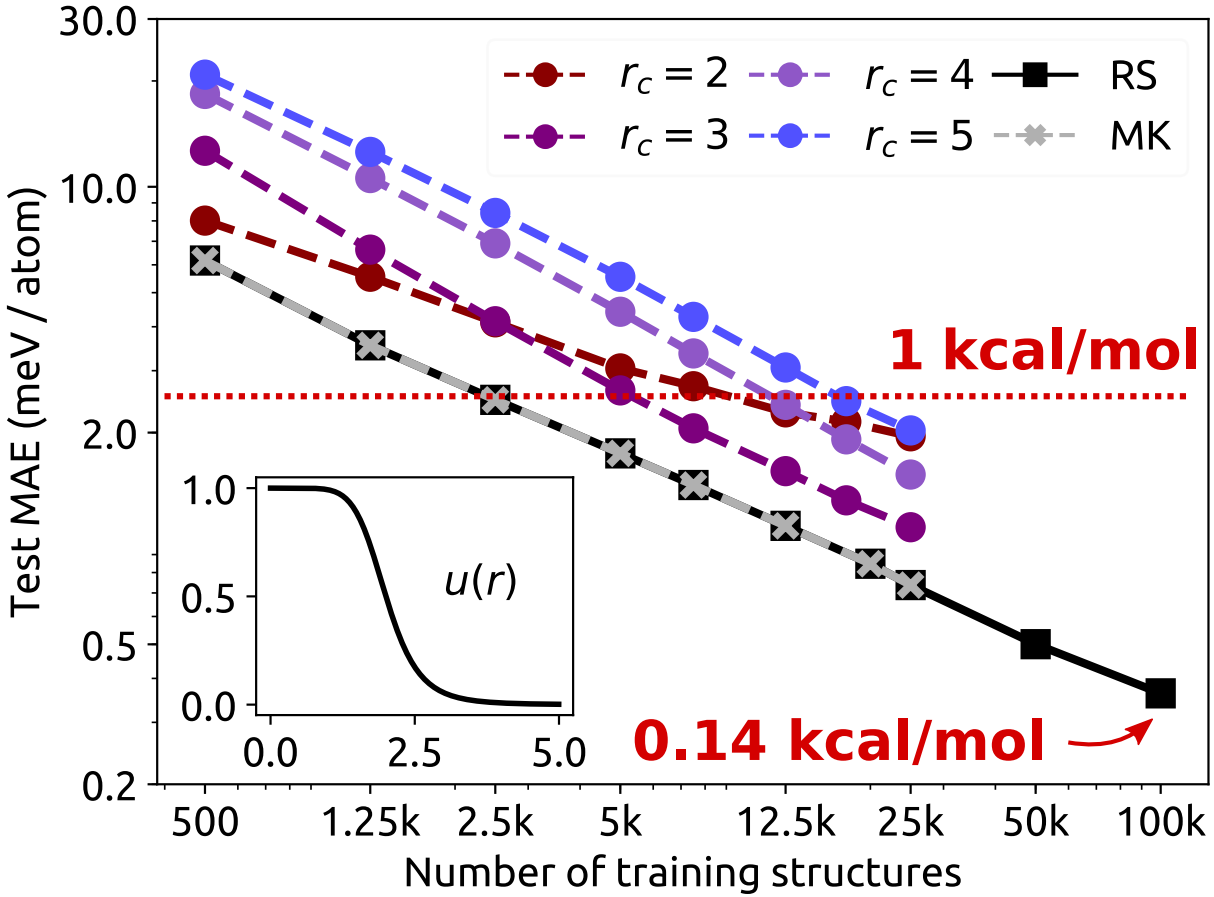
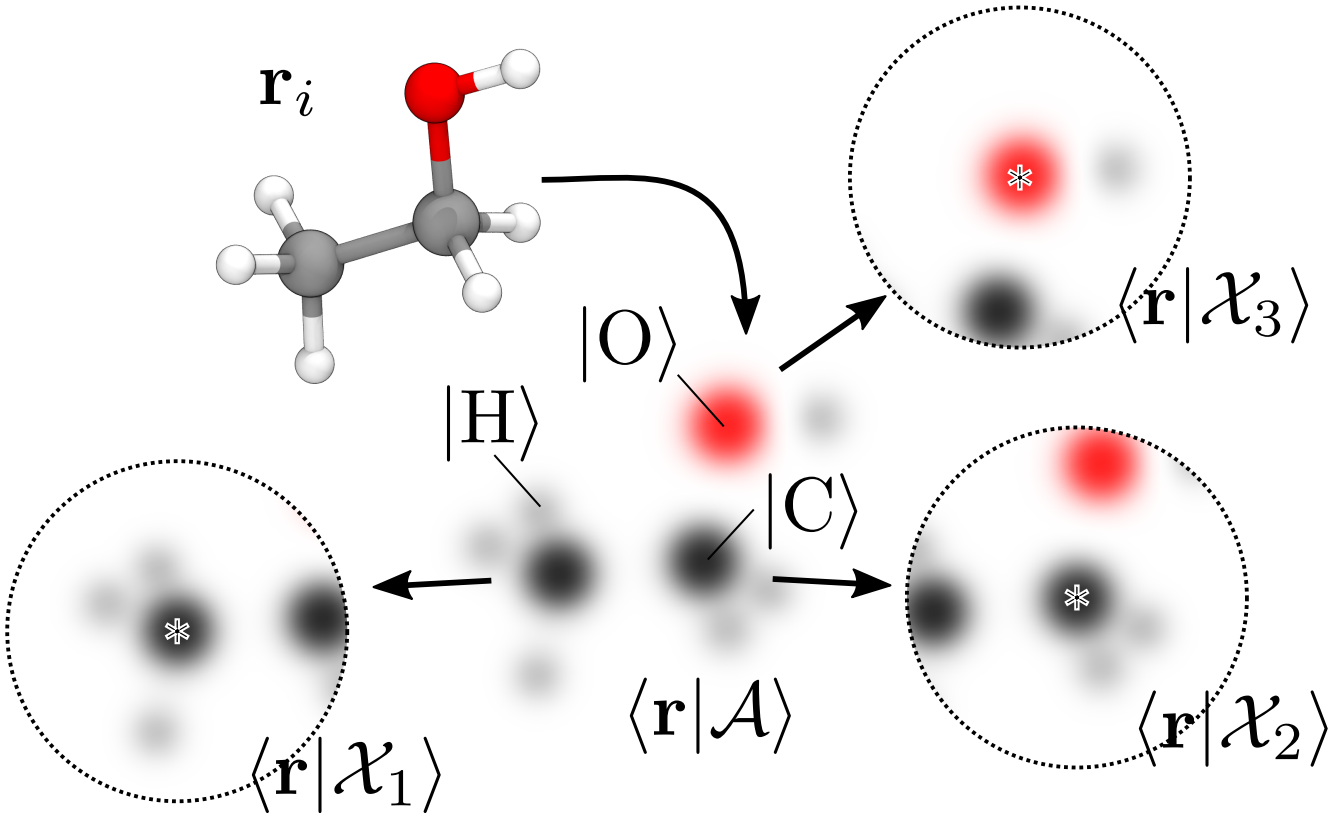


Willatt, Musil, **MC**, PCCP (2018)

Machine-learning the periodic table

- How to learn with multiple species? Decorate atomic Gaussian with elemental kets $|\mathrm{H}\rangle$, $|\mathrm{O}\rangle$, . . .
- 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!



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$$|\mathrm{H}\rangle = 0.5\,|🔥\rangle + 0.1\,|🪨\rangle + 0.2\,|💧\rangle$$
$$|\mathrm{C}\rangle = 0.2\,|🔥\rangle + 0.8\,|🪨\rangle + 0.3\,|💧\rangle$$
$$|\mathrm{O}\rangle = 0.1\,|🔥\rangle + 0.1\,|🪨\rangle + 0.6\,|💧\rangle$$

Empedocles et al. (ca 360BC). Metaphor courtesy of Albert Bartók

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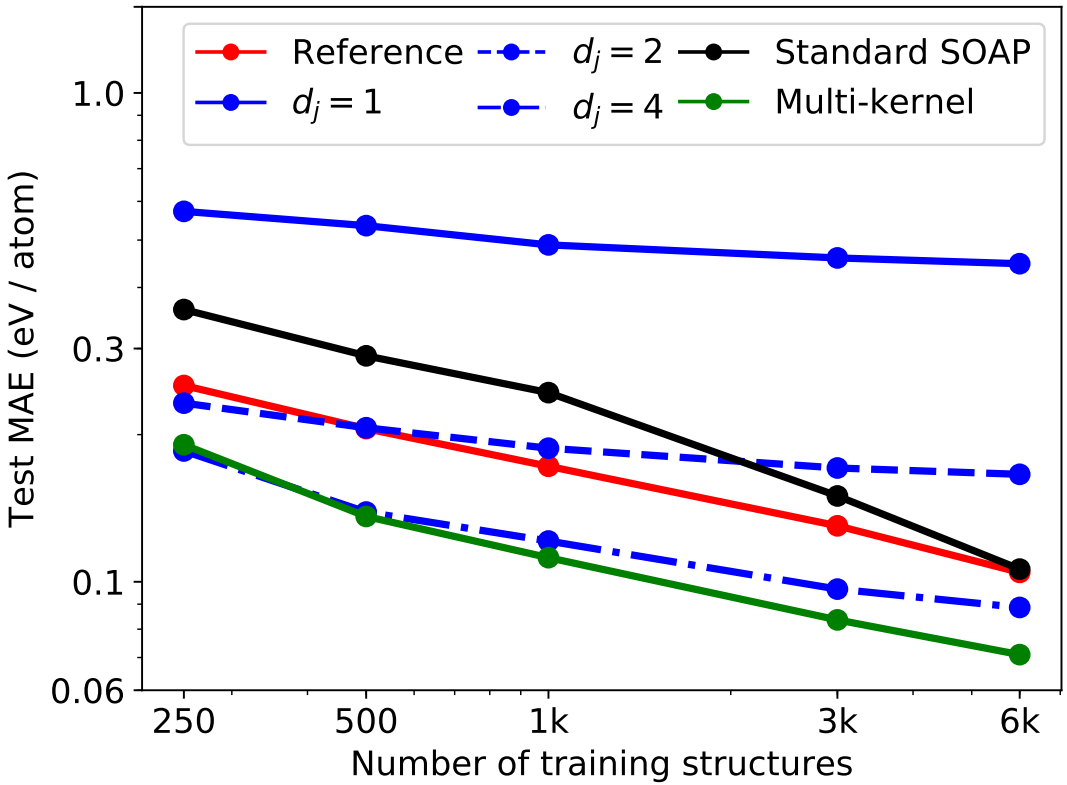

Elpasolite dataset. Reference curve (red) from Faber et al. JCP (2018)

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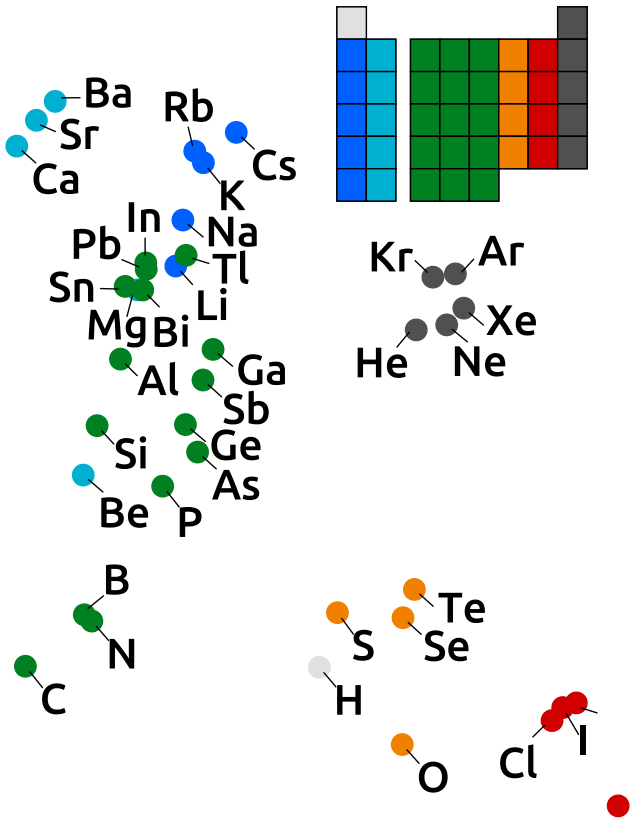
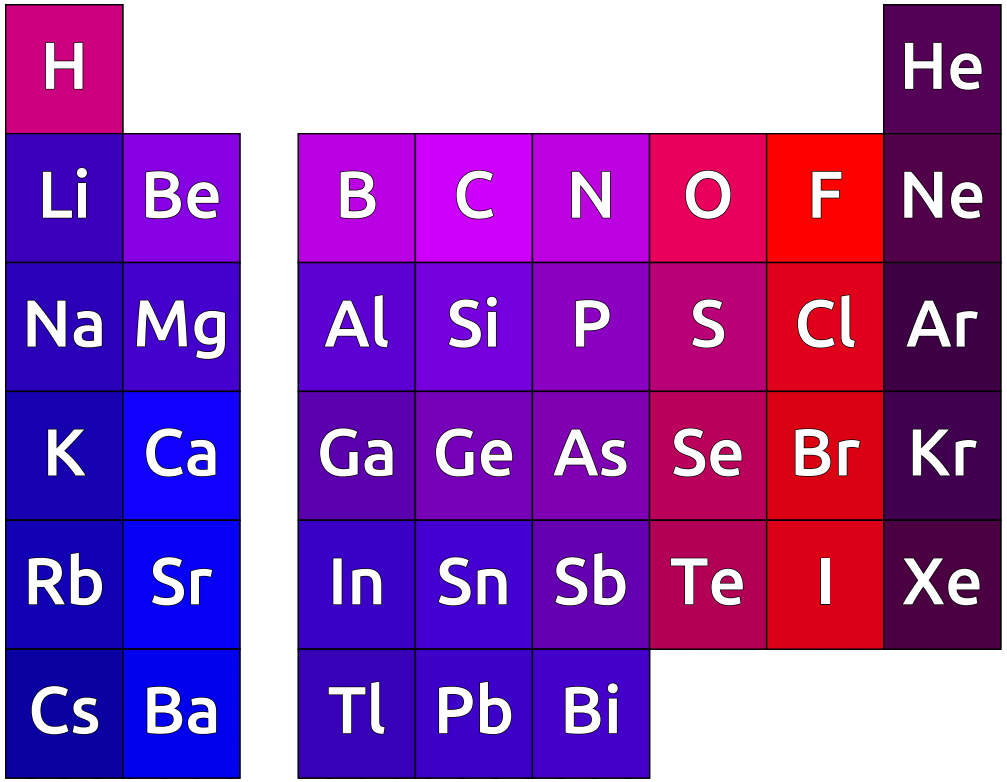


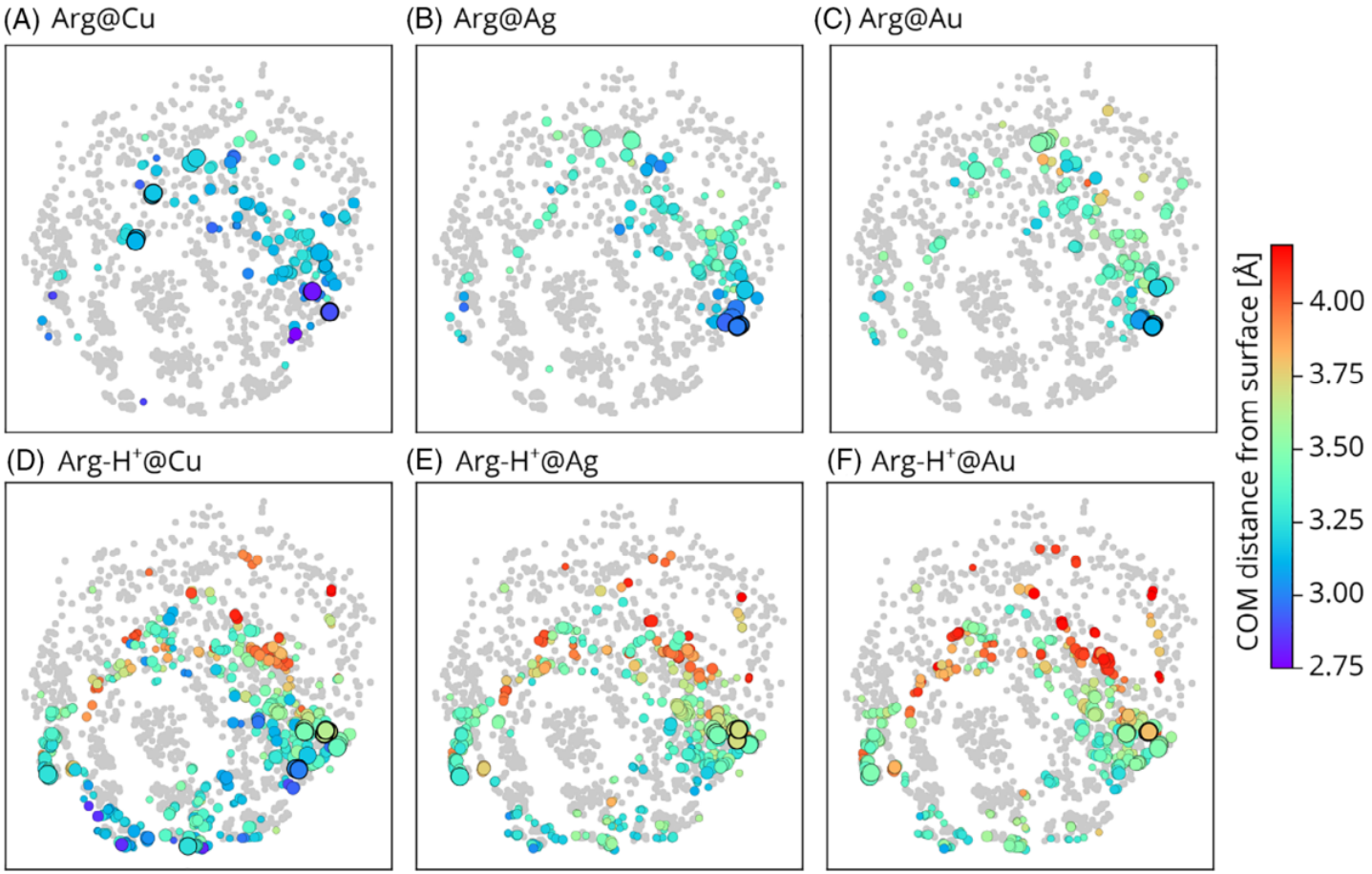


Willatt, Musil, Ceriotti, PCCP (2018)

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)

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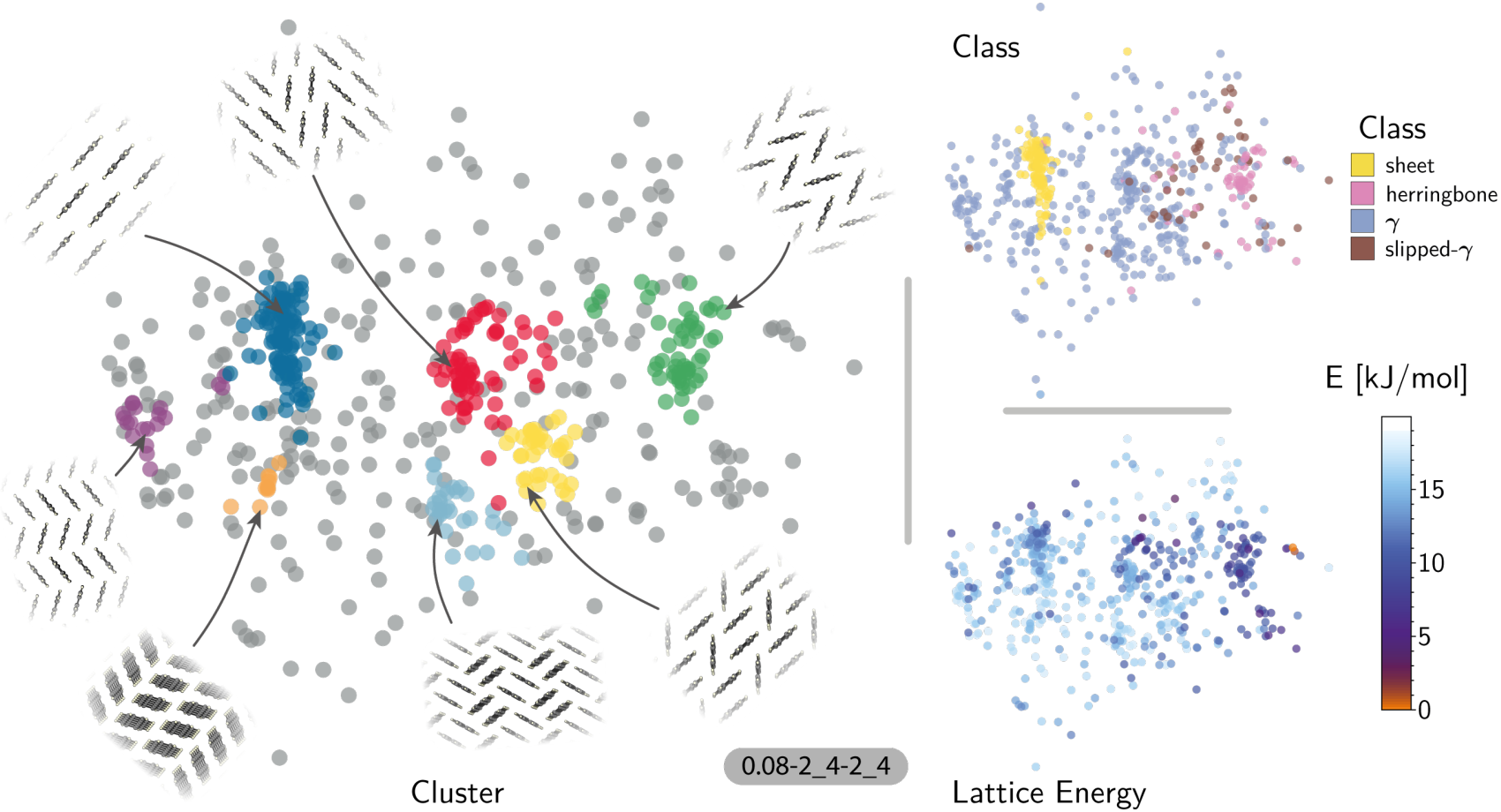
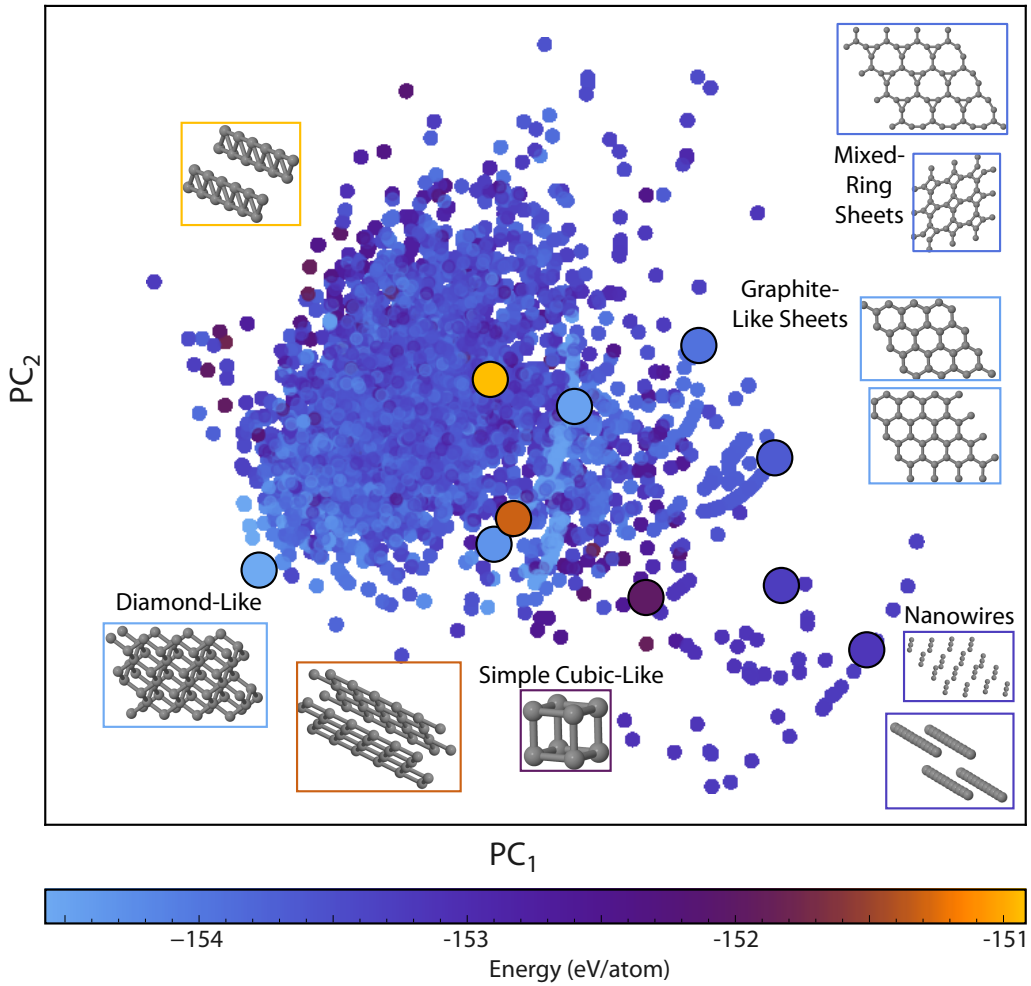


Musil, De, Yang, Campbell, Day, **MC**, Chemical Science (2018);`http://interactive.sketchmap.org`

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

MC, *Unsupervised machine learning in atomistic simulations, between predictions and understanding*, JCP (2019)

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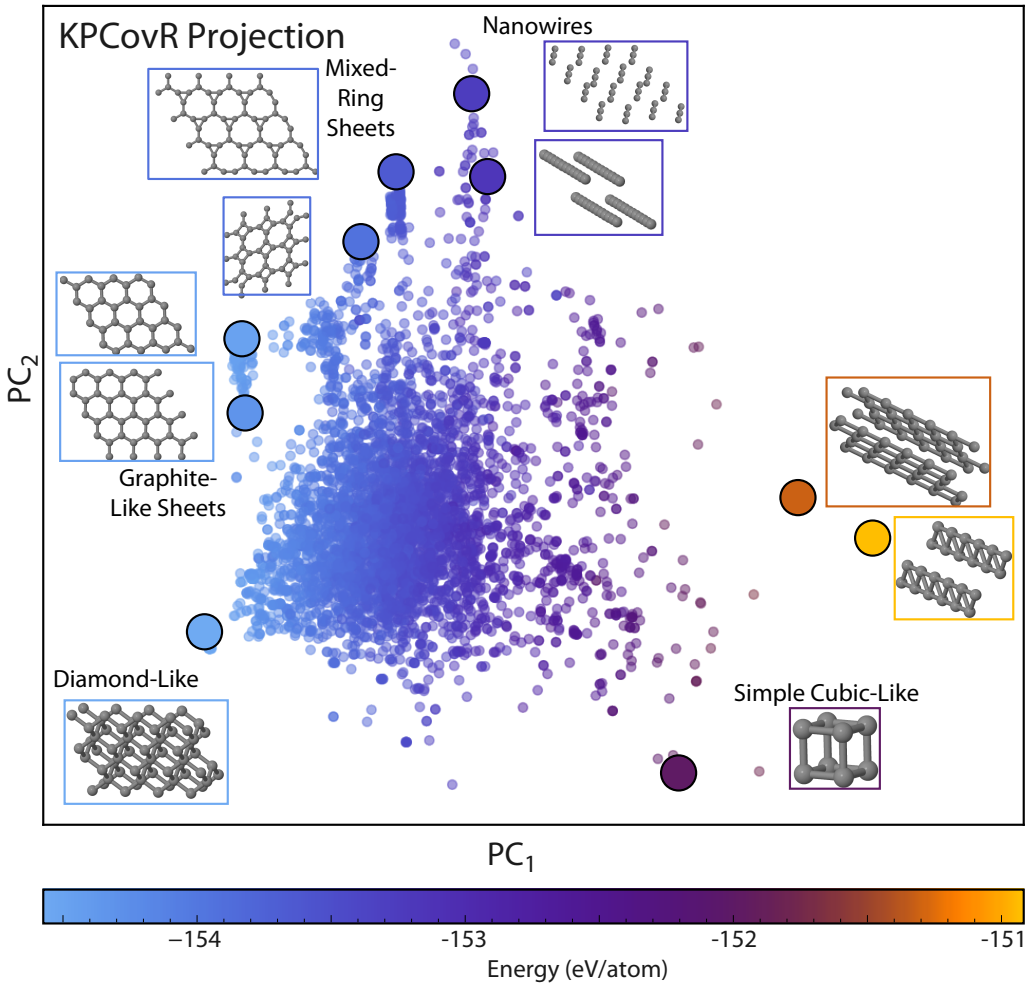
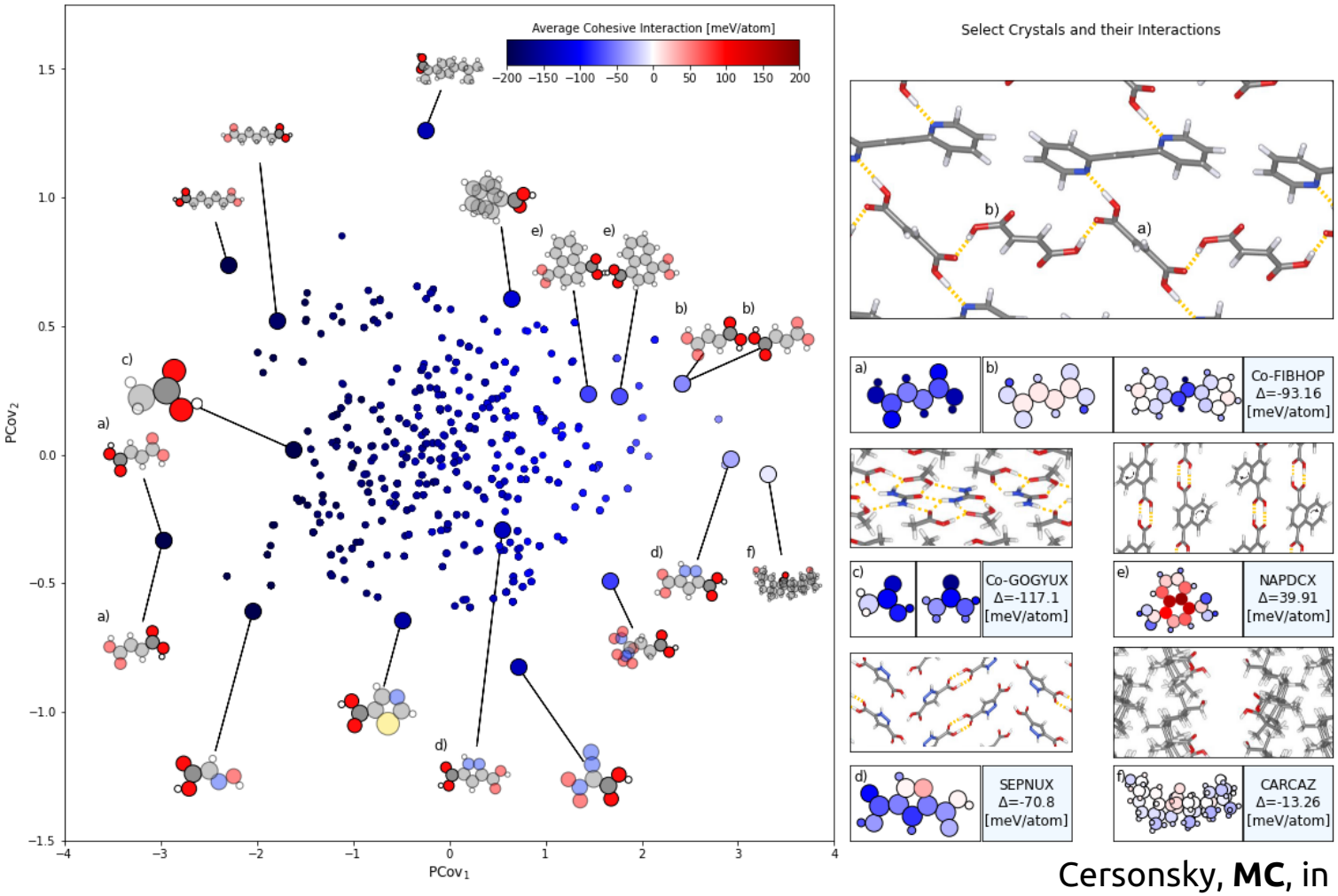


https://www.materialscloud.org/discover/kpcovr/carbons-05

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



Cersonsky, **MC**, in preparation

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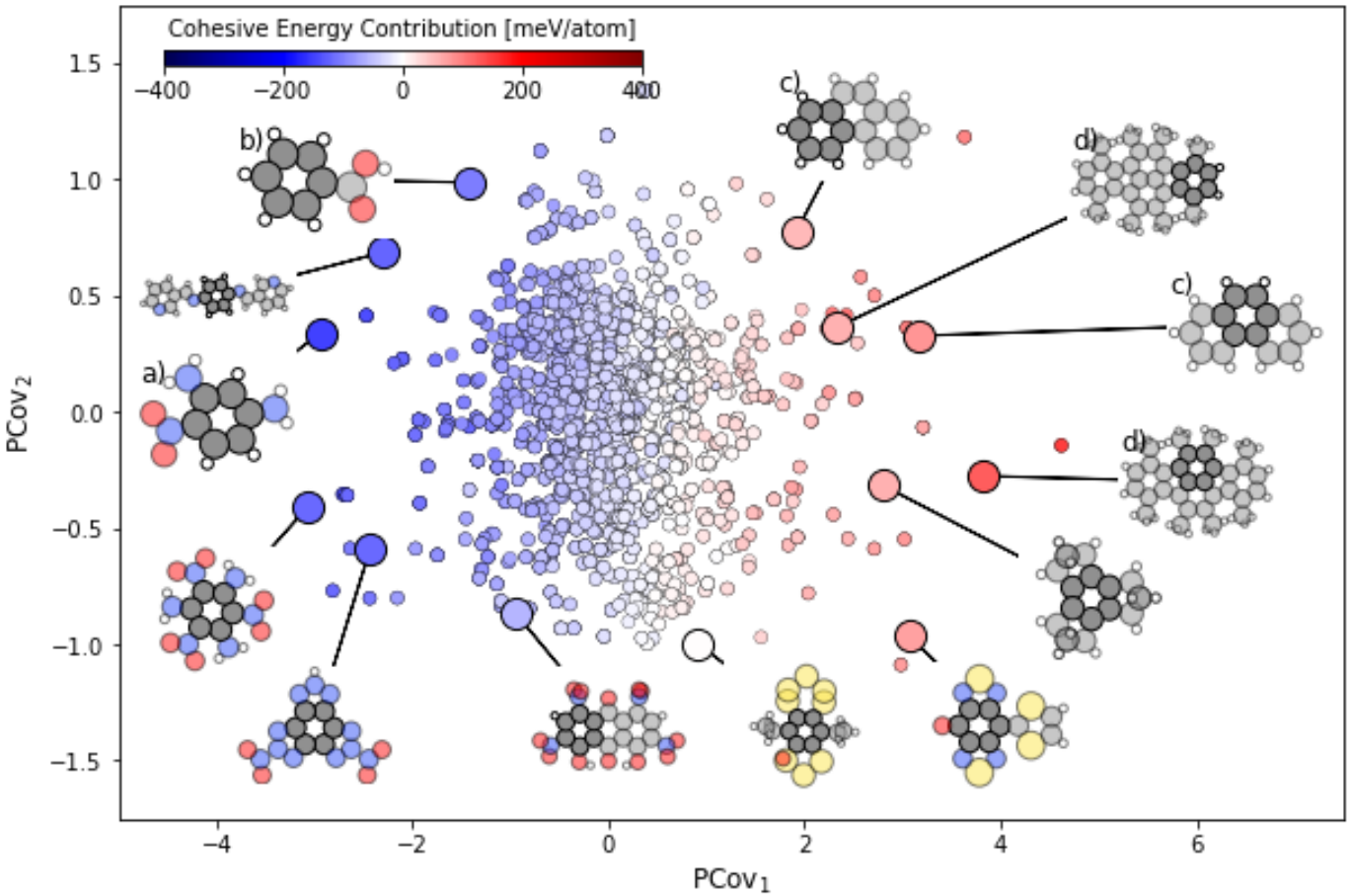
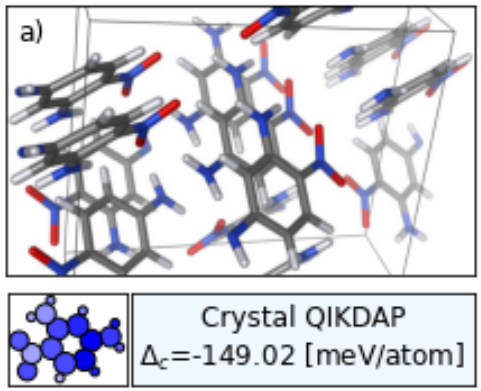
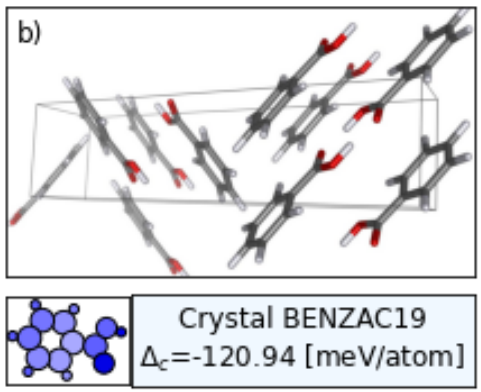


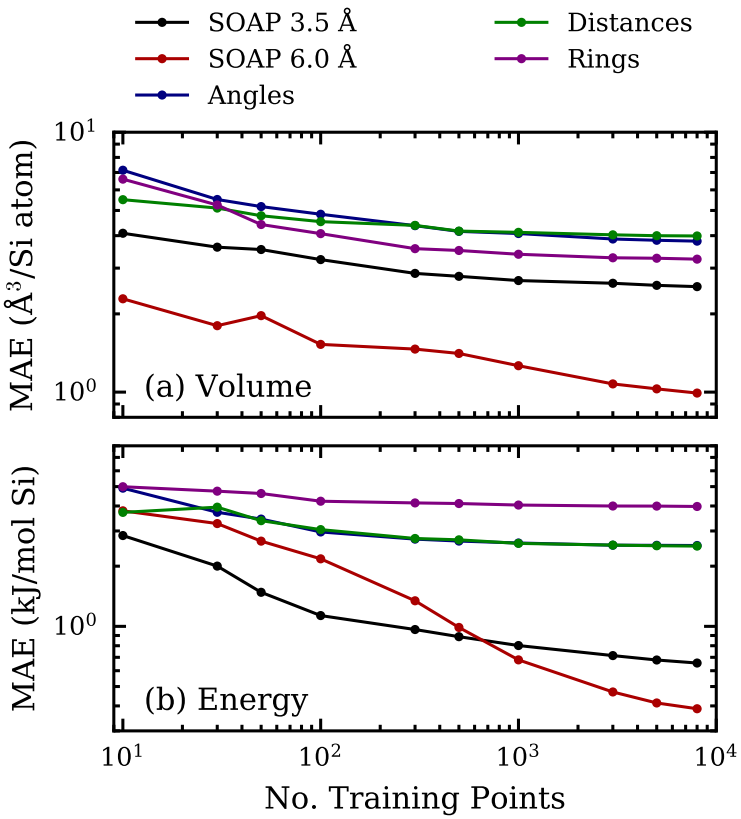


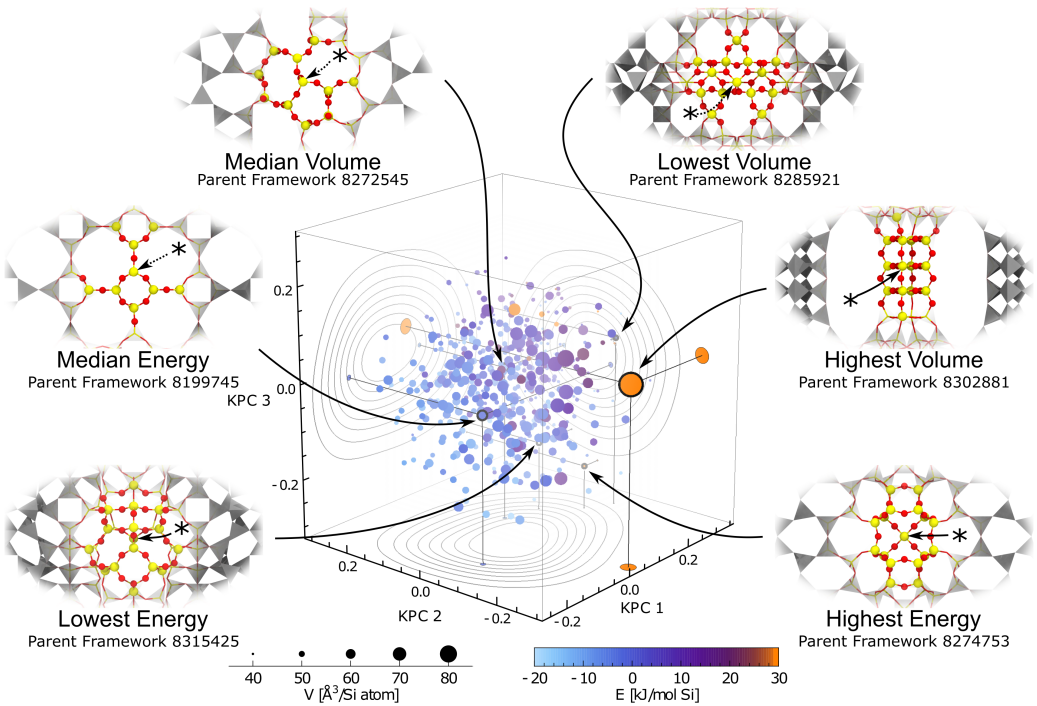


Cersonsky, **MC**, in preparation

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

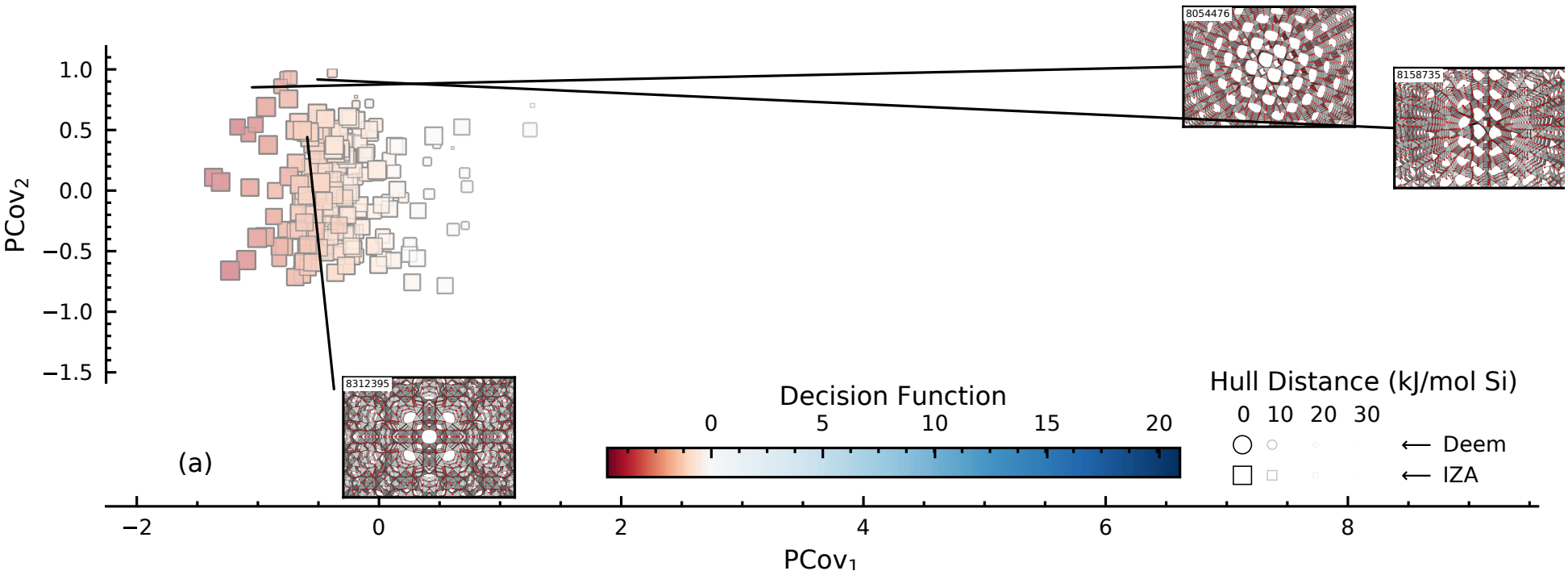




Helfrecht et al., JCP (2019)

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



Helfrecht et al., arxiv:2110.13764

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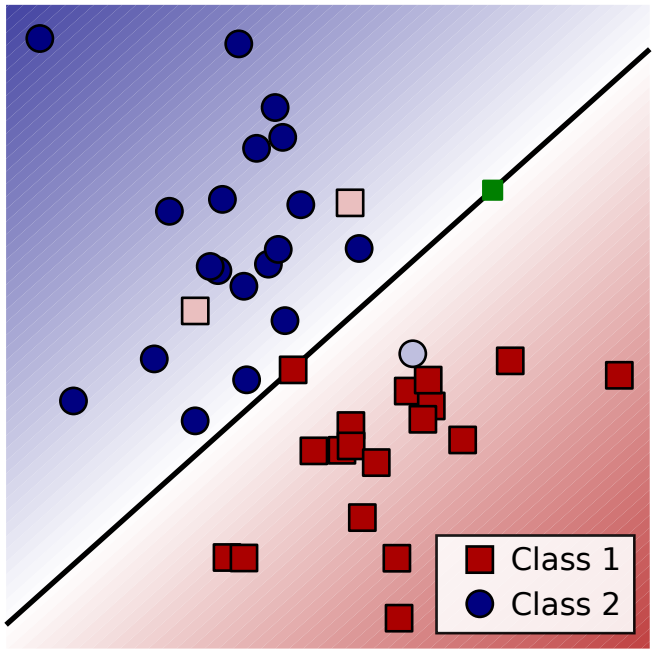
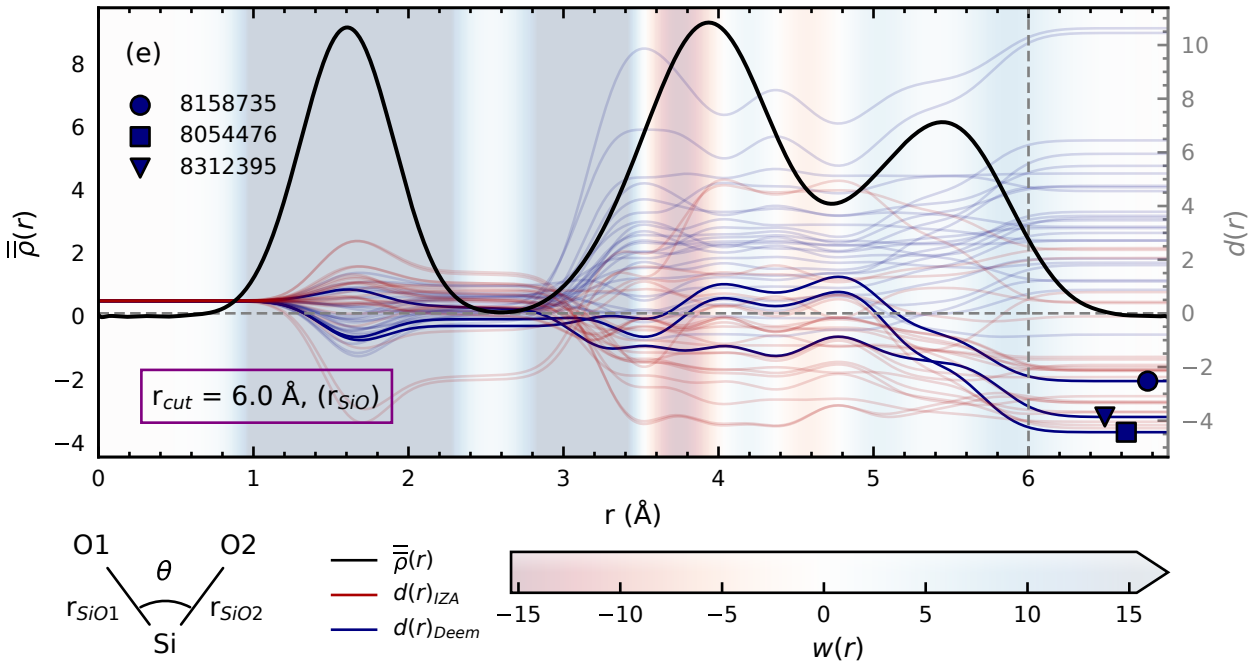
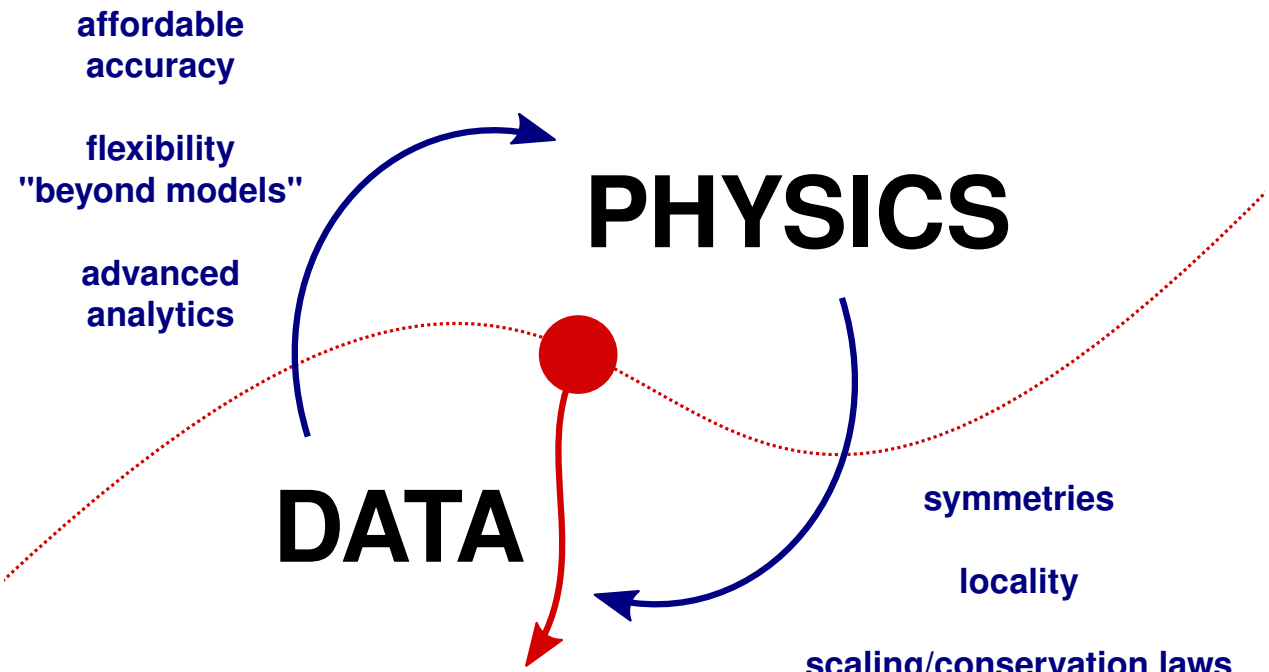




Helfrecht et al., arxiv:2110.13764

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?



Slides → tinyurl.com/ceriotti-2022-berkeley
Review → Musil et al. ChemRev (2021)
Code: → github.com/lab-cosmo

A Dirac notation for ML

features index

$$\langle Q | A \rangle$$

representation target & nature

$$\langle n_1 l_1; \dots n_\nu l_\nu k_\nu | A; \rho_i^{\otimes\nu}; \sigma; \lambda\mu \rangle$$

radial indices

angular channels

structure

correlation order

parity

field center

rot. symmetry

- 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

$$\langle Y|A\rangle = \int \mathrm{d}Q \, \langle Y|Q\rangle \, \langle Q|A\rangle$$

Willatt, Musil, **MC**, JCP (2019); `https://tinyurl.com/dirac-rep`

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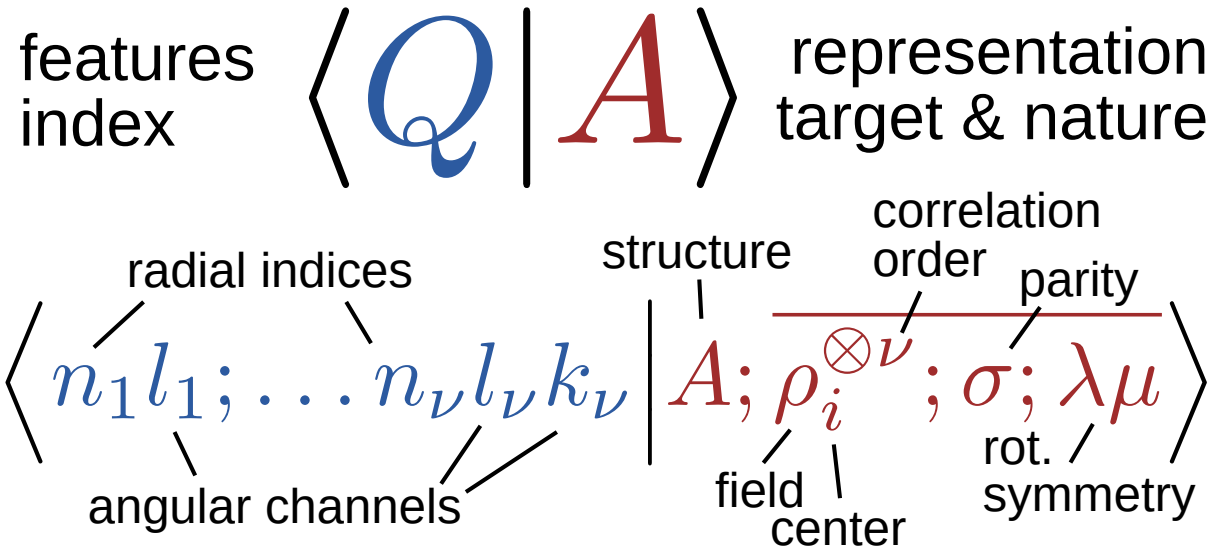

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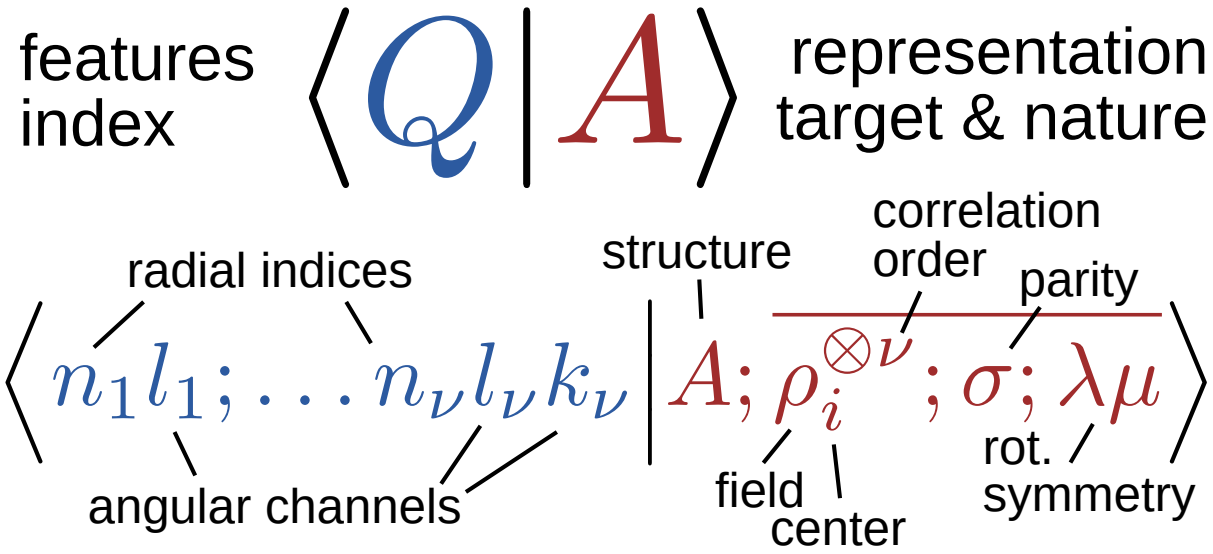
$$k(A,A') = \langle A|A'\rangle \approx \int \mathrm{d}Q\, \langle A|Q\rangle\, \langle Q|A'\rangle$$

Willatt, Musil, **MC**, JCP (2019); `https://tinyurl.com/dirac-rep`

A Dirac notation for ML



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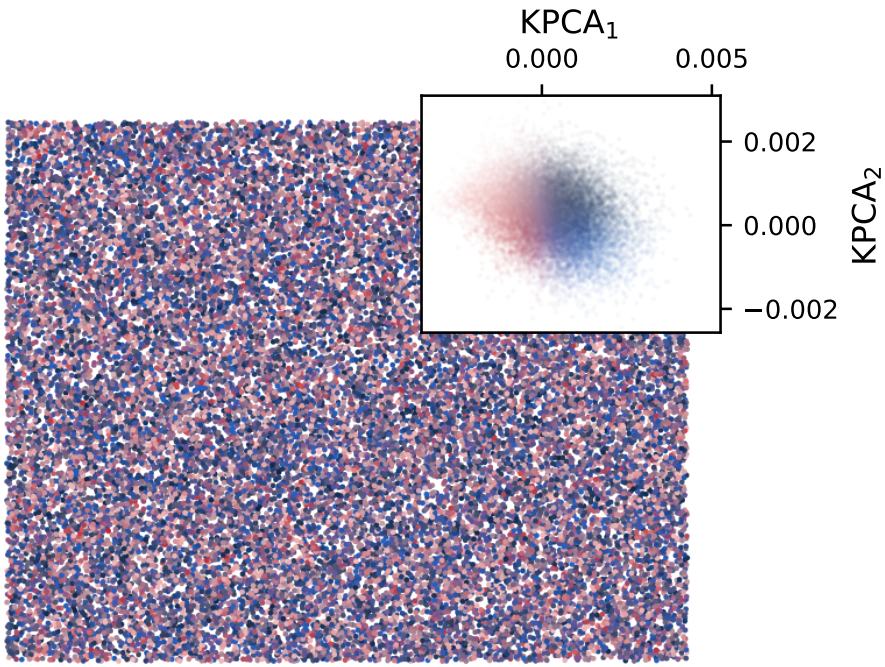
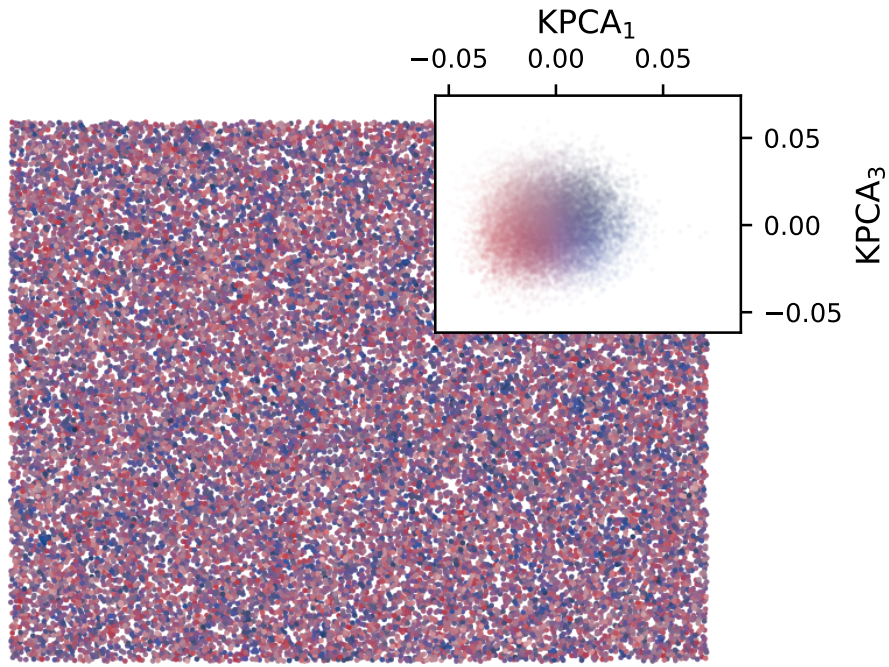
$$E(A) = \langle E|A\rangle \approx \int \mathrm{d}Q \, \langle E|Q\rangle \, \langle Q|A\rangle$$

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





data: Shibuta, Sakane, Takaki, Ohno, Acta Mat. (2016)

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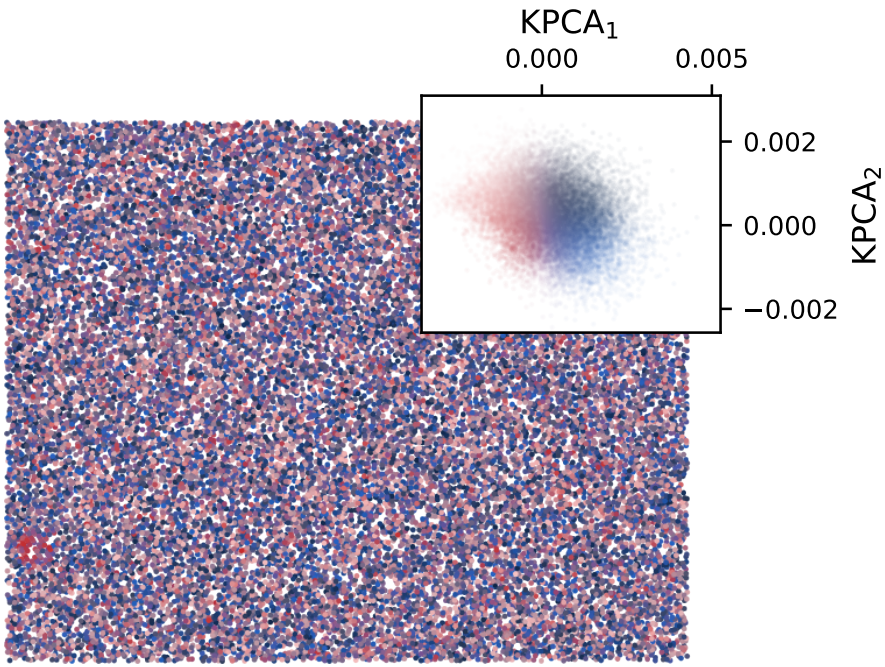

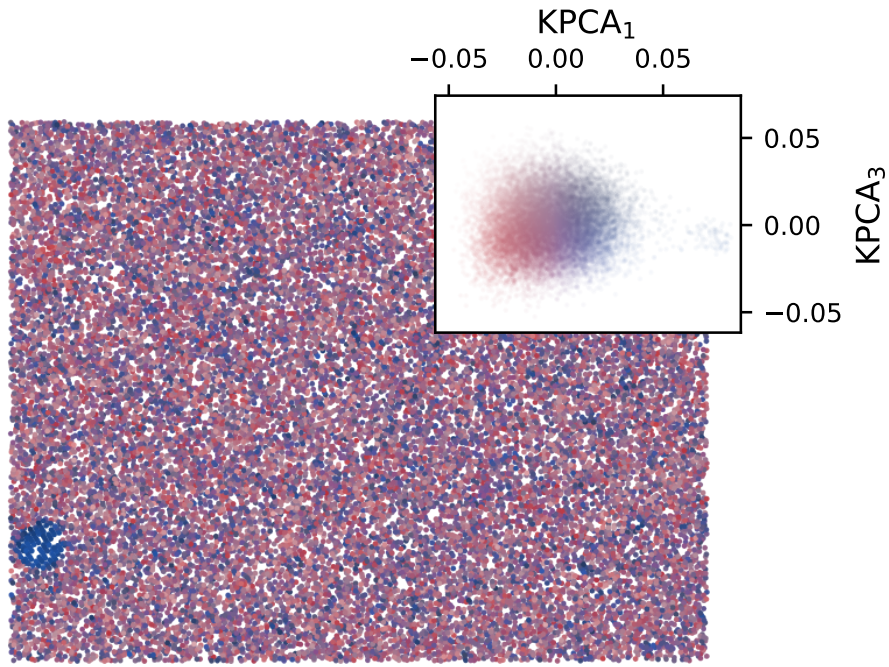

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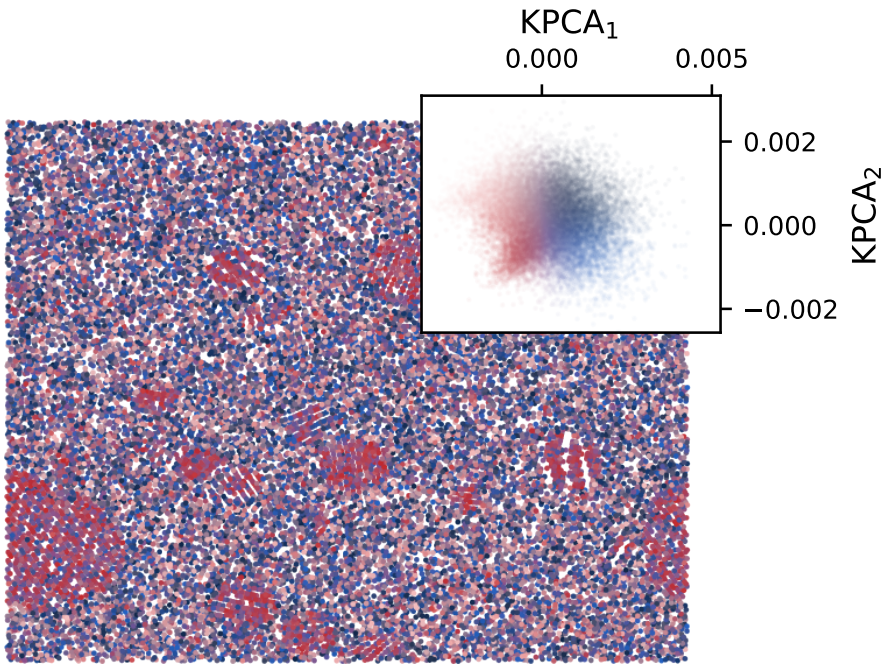

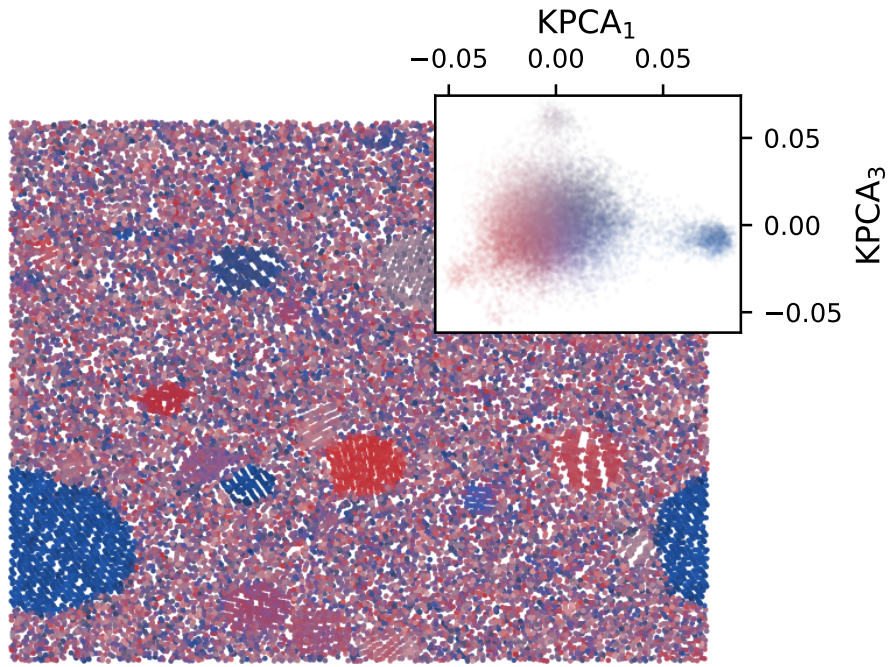

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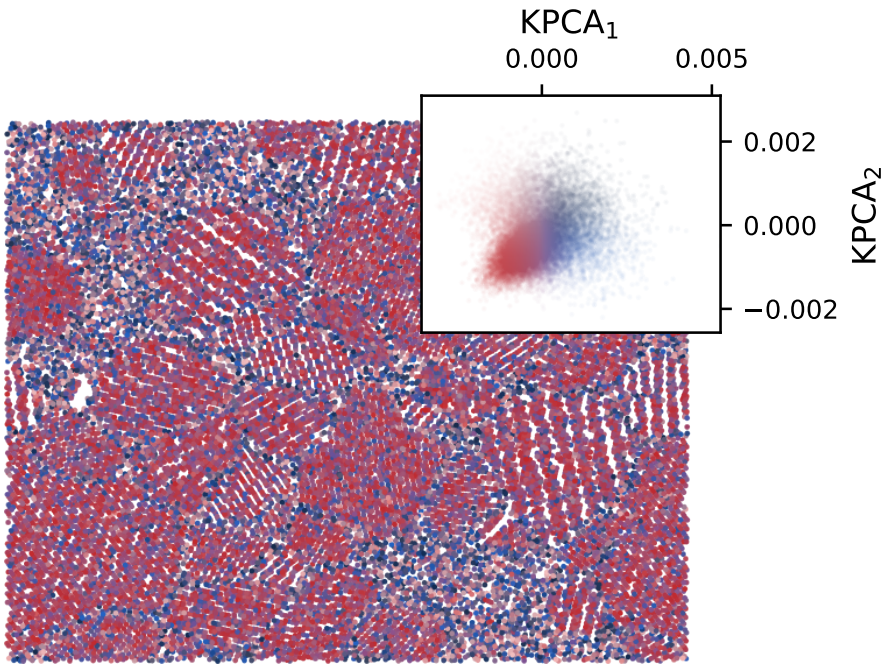

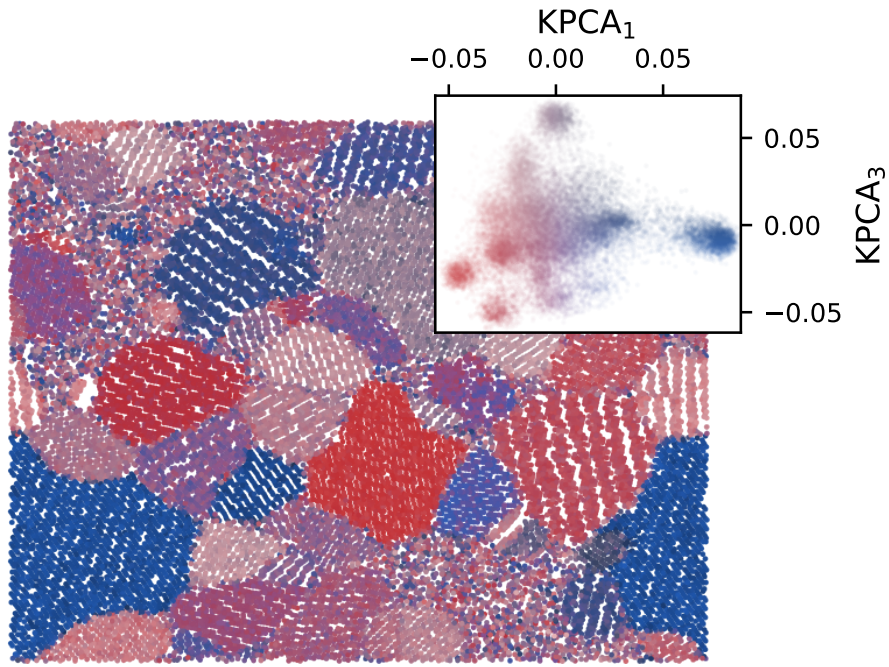

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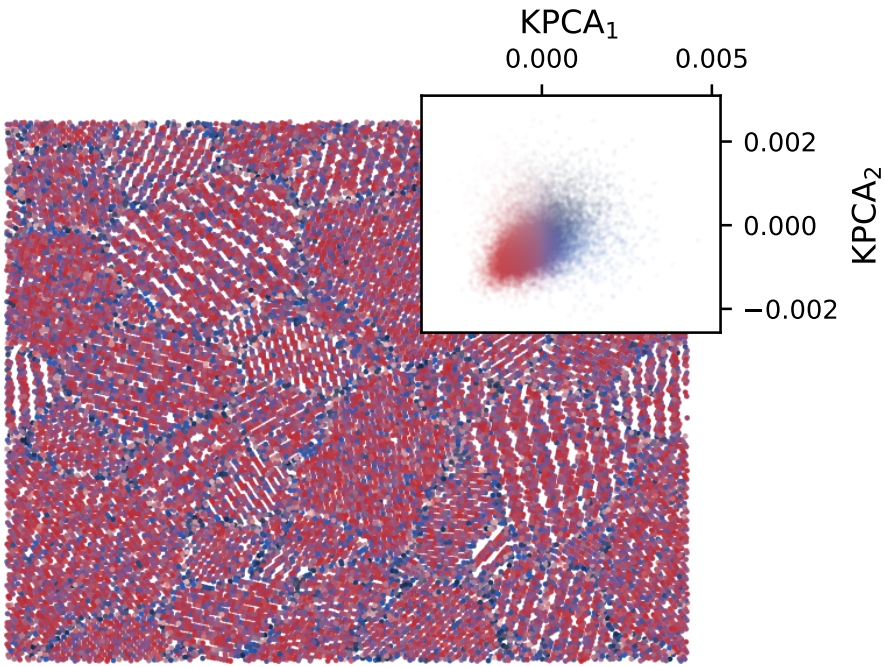

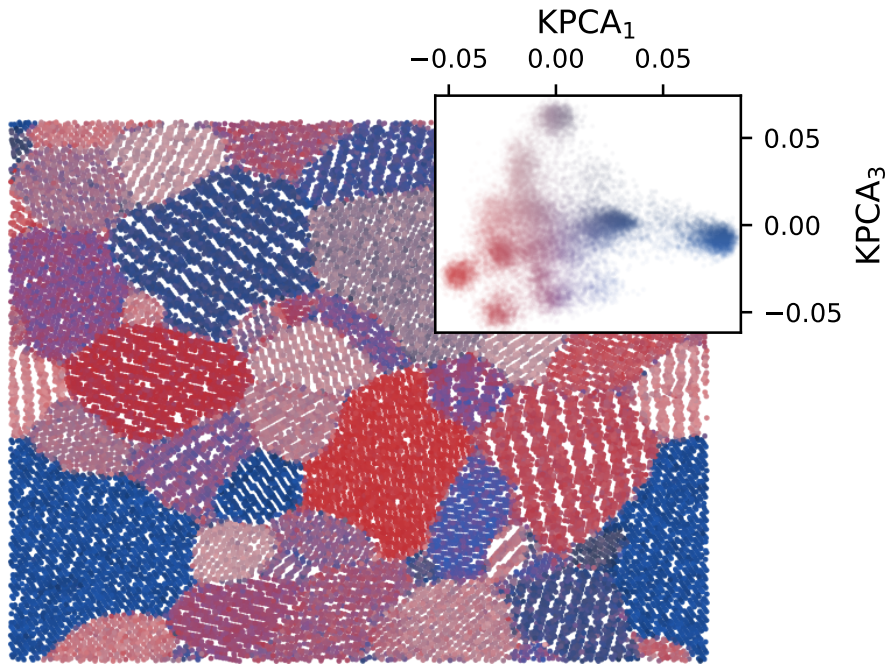

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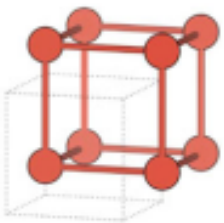
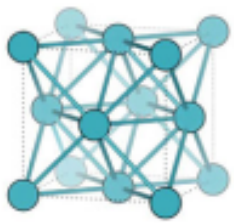
Variations on a theme

- 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 $|\overline{\rho_i^{\otimes 2}}\rangle$ (Ziletti et al. N.Comm 2018)
  - Projection on symmetry functions (Behler-Parrinello, DeepMD)

$$\langle \mathbf{k}|A;\overline{\rho^{\otimes 2}}\rangle = \sum_{ij\in A} e^{i\mathbf{k}\cdot\mathbf{r}_{ij}}$$

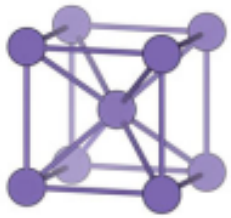


Simple cubic
(sc) structure
spgroup = 221

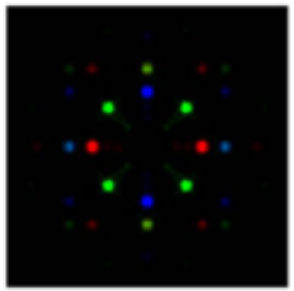


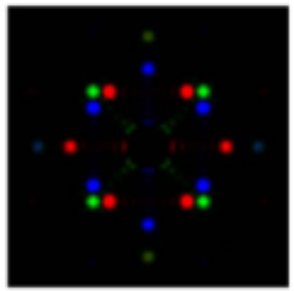
Face-centered-cubic
(fcc) structure
spgroup = 225

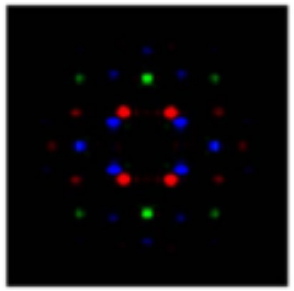
Diamond
(diam) structure
spgroup = 227

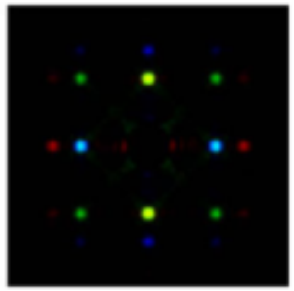


Body-centered-cubic
(bcc) structure
spgroup = 229









Willatt, Musil, **MC**, JCP (2019), https://arxiv.org/pdf/1807.00408

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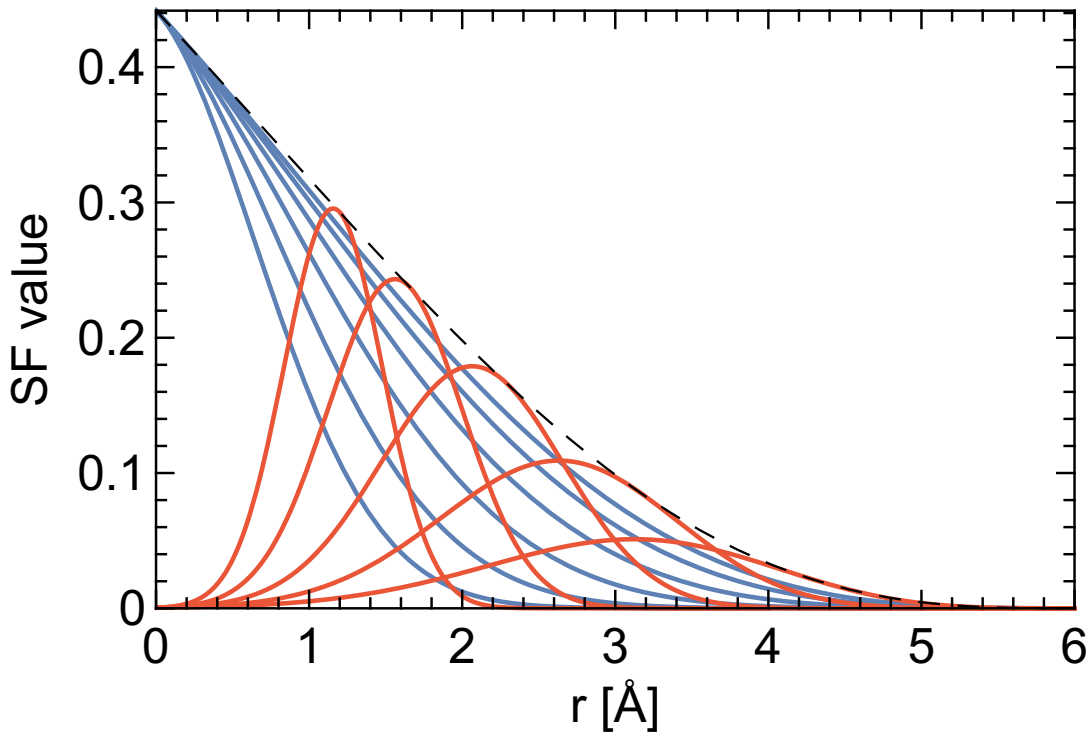
$$\langle abG_2|\overline{\rho_i^{\otimes 1}}\rangle = \delta_{aa_i} \int \mathrm{d}r\, G_2(r)\, \langle br|\overline{\rho_i^{\otimes 1}}; g \to \delta\rangle$$



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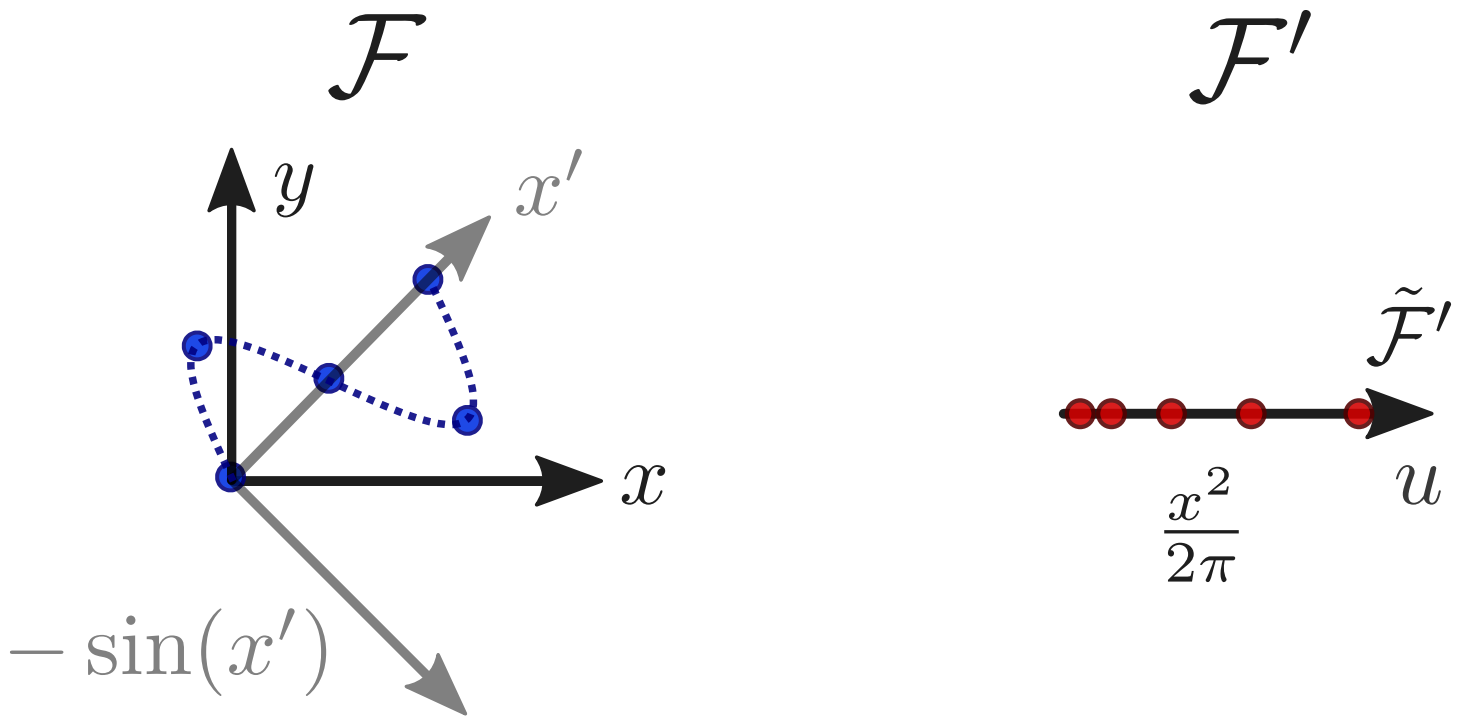
Measuring feature spaces

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

$$\mathrm{GFRE}(\mathcal{F} \to \mathcal{F}') = \min_{\mathbf{P} \in \mathbb{R}^{n_{\mathcal{F}} \times n_{\mathcal{F}'}}} \|\mathbf{X}_{\mathcal{F}'} - \mathbf{X}_{\mathcal{F}}\mathbf{P}\|$$



Goscinski, Fraux, **MC**, MLST (2021)

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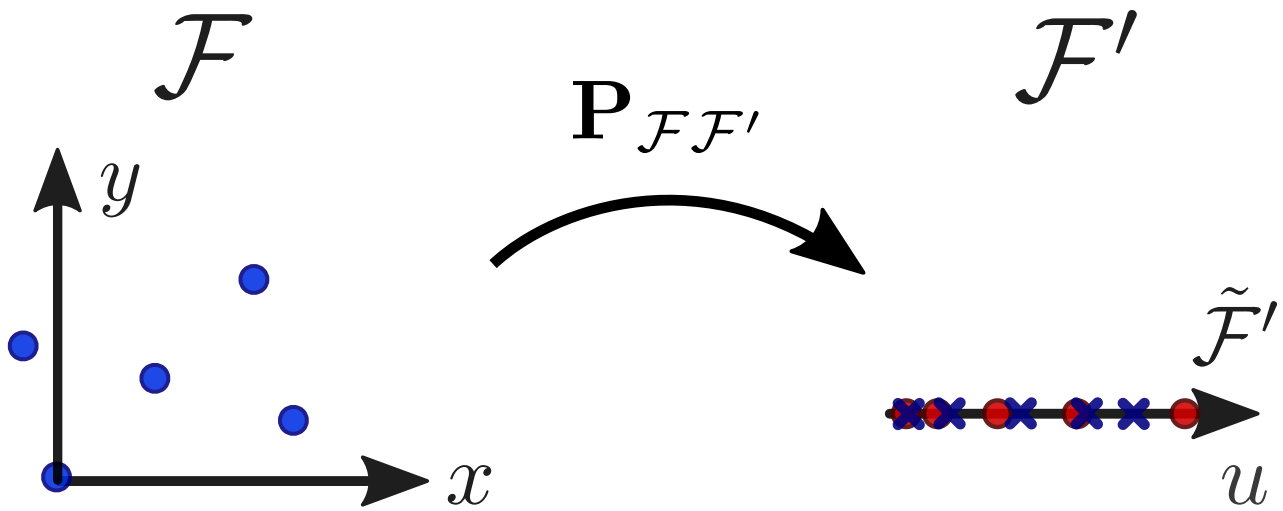

$$\mathrm{GFRE}(\mathcal{F}, \mathcal{F}')$$

Goscinski, Fraux, **MC**, MLST (2021)

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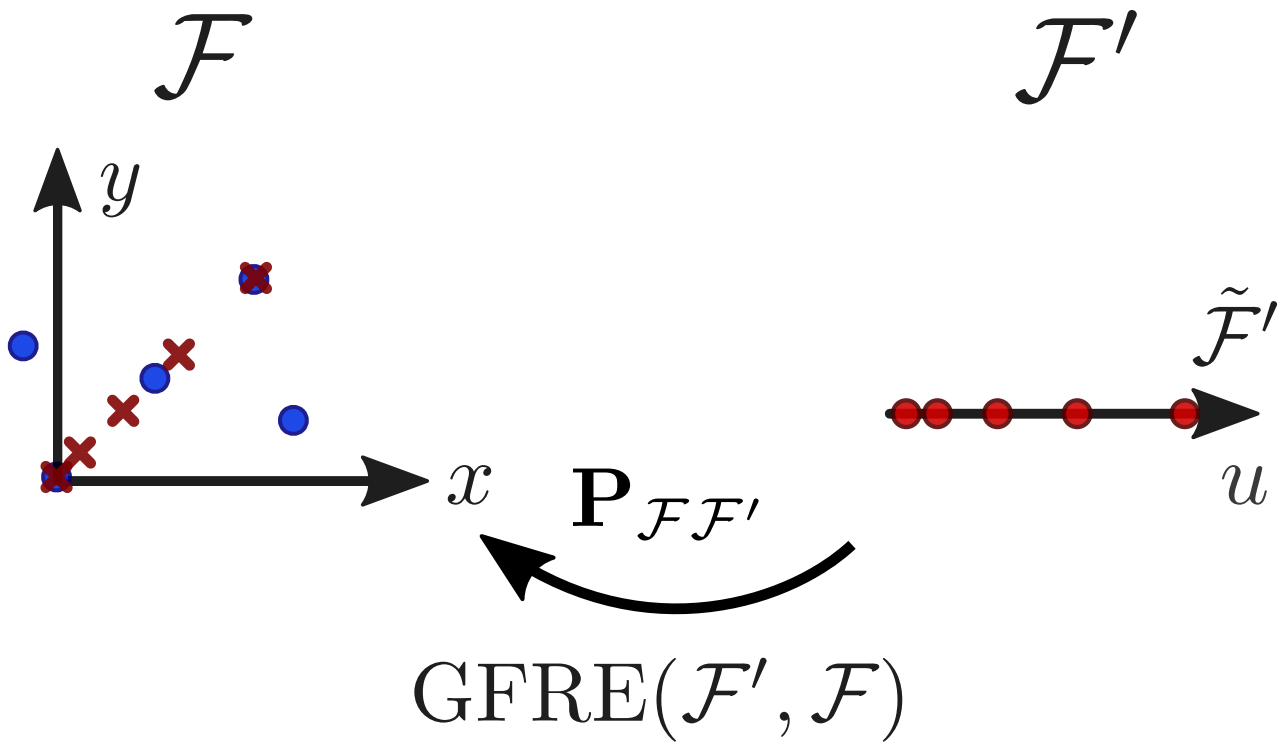

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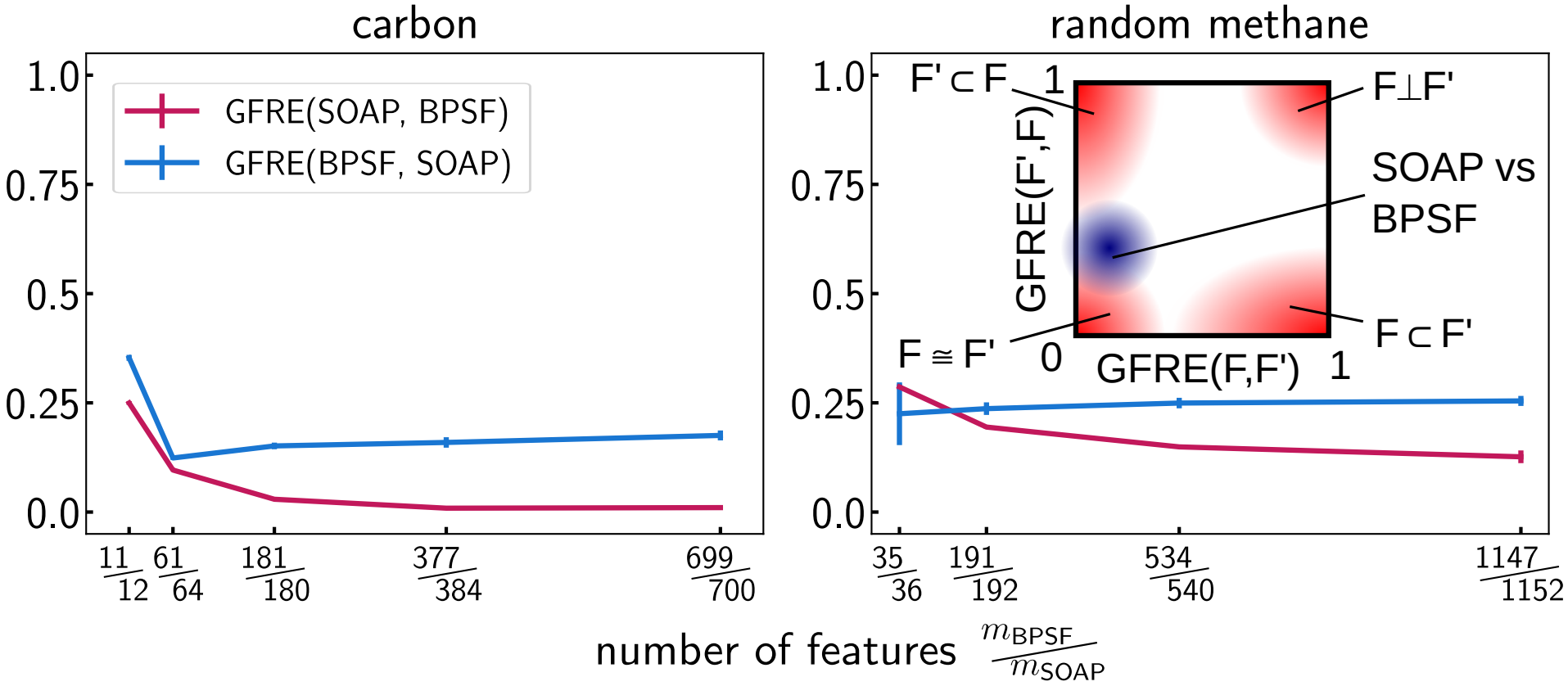
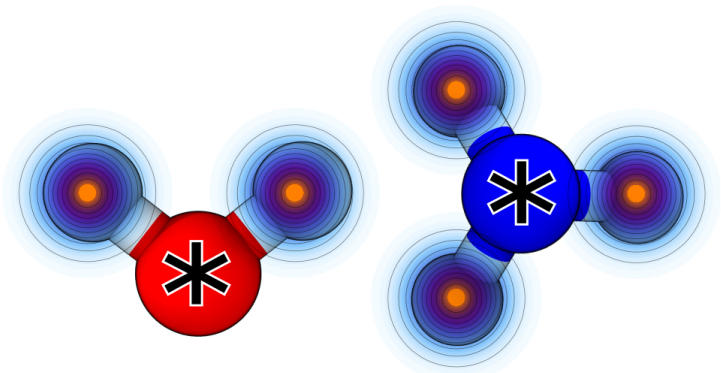
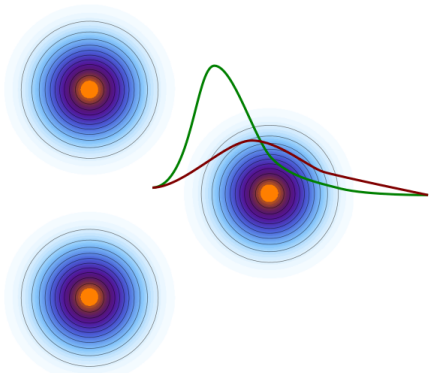


Density expansion and SOAP

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



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



$$\langle nlm|\rho_i\rangle = \int \mathrm{d}\mathbf{r}\, \langle \mathbf{r}|\rho_i\rangle R_n(r) Y_m^l(\hat{\mathbf{r}})$$

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

Density expansion and SOAP

- What if we use radial functions and spherical harmonics?
- Symmetrized tensor product $\rightarrow$ SOAP power spectrum!
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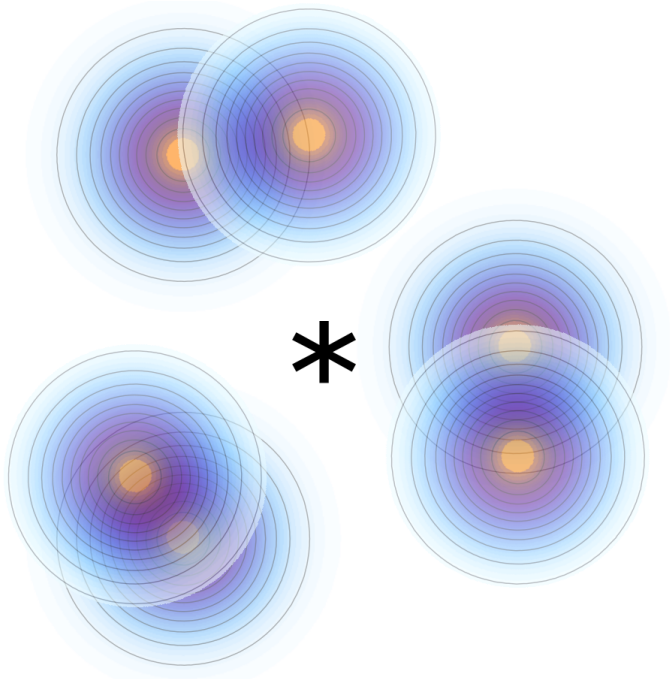

$$\langle nn'l|\overline{\rho_i^{\otimes 2}}\rangle = \sum_m \langle nlm|\rho_i\rangle^\star \langle n'lm|\rho_i\rangle$$
$$p_{nn'l} = \sum_m c^\star_{nlm} c_{n'lm}$$

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

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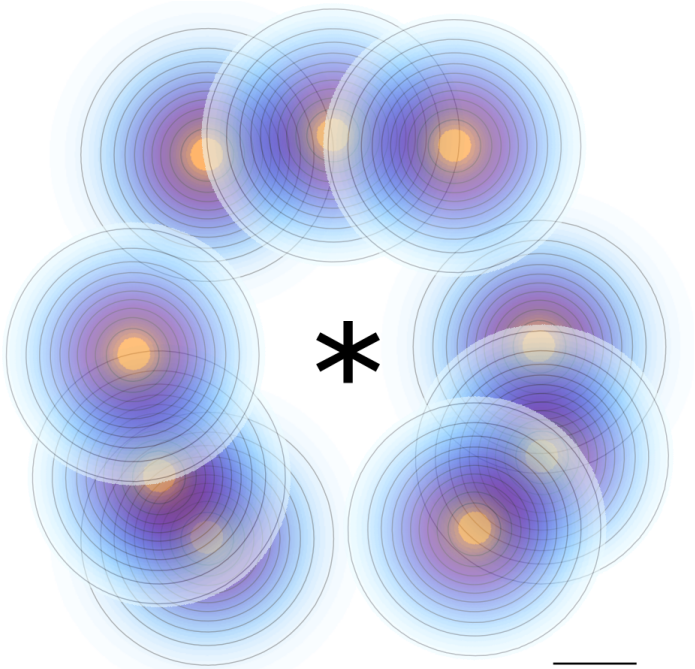
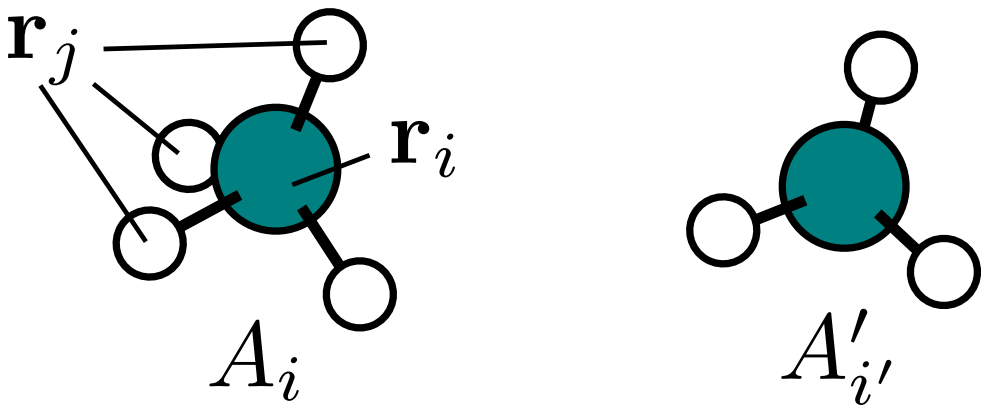


$$\langle n_1 l_1 m_1; n_2 l_2 m_2; \dots n_\nu l_\nu m_\nu | \overline{\rho_i^{\otimes \nu}} \rangle =$$
$$\int \mathrm{d}\hat{R} \langle n_1 l_1 m_1 | \hat{R} | \rho_i \rangle \cdots \langle n_\nu l_\nu m_\nu | \hat{R} | \rho_i \rangle =$$
$$\sum_{m'_1 \dots m'_\nu} \langle n_1 l_1 m'_1 | \rho_i \rangle \cdots \langle n_\nu l_\nu m'_\nu | \rho_i \rangle$$
$$\times \int \mathrm{d}\hat{R}\, D^{l_1}_{m_1 m'_1}(\hat{R}) \cdots D^{l_\nu}_{m_\nu m'_\nu}(\hat{R})$$

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. $g \to \delta$ limit $\Rightarrow$ list of 2-neighbors tuples $(r_{j_1 i}, r_{j_2 i}, \hat{\mathbf{r}}_{j_1 i} \cdot \hat{\mathbf{r}}_{j_2 i})$
  6. Linear model $\Rightarrow$ 3-body potential!



$$\{\mathbf{r}_{ji} = \mathbf{r}_j - \mathbf{r}_i\} \leftrightarrow A_i$$

Bartók, Kondor, Csányi, PRB (2013)

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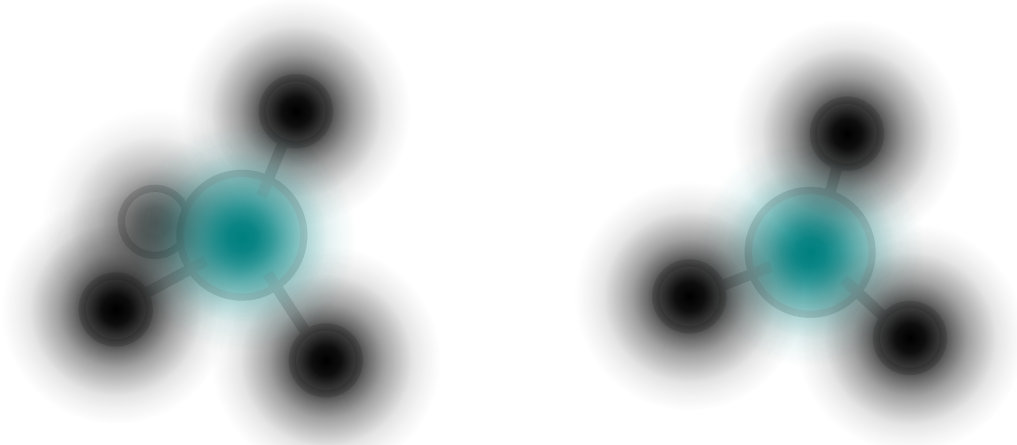

$$\langle a\mathbf{x}|\rho_i\rangle = \sum_{j\in A_i} \delta_{aa_j} \langle \mathbf{x}|\mathbf{r}_{ji}; g\rangle$$
$$\langle \mathbf{x}|\mathbf{r}_{ji}; g\rangle \equiv g(\mathbf{x} - \mathbf{r}_{ji})$$

Bartók, Kondor, Csányi, PRB (2013)

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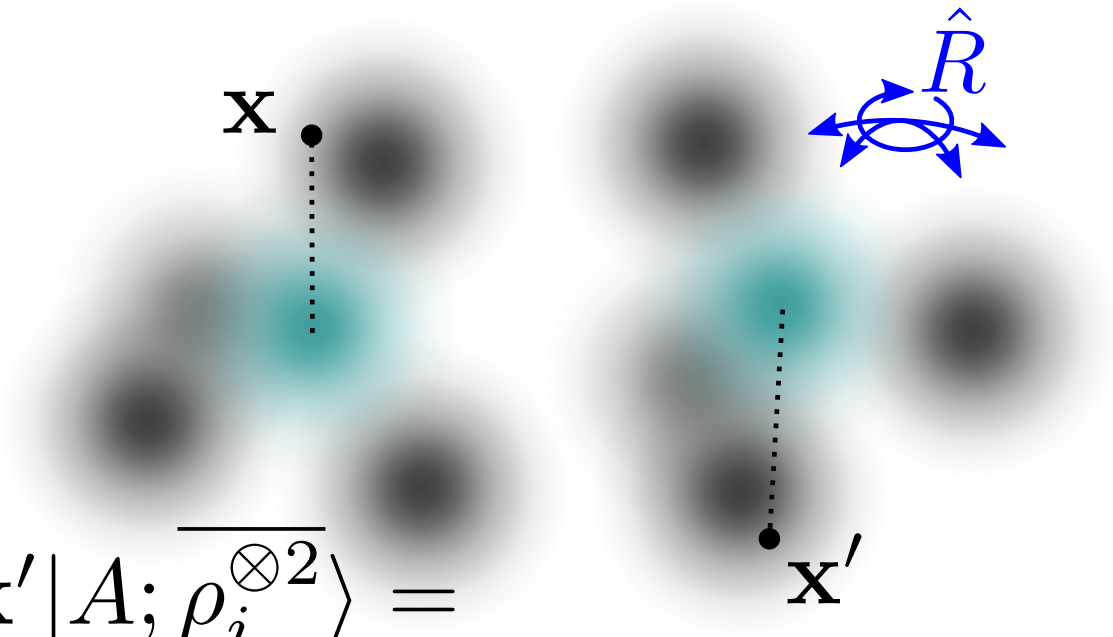

$$\langle \mathbf{x}; \mathbf{x}' | A; \overline{\rho_i^{\otimes 2}} \rangle = \int \mathrm{d}\hat{R} \langle \mathbf{x} | \hat{R}A; \rho_i \rangle \langle \mathbf{x}' | \hat{R}A; \rho_i \rangle$$

Willatt, Musil, **MC**, JCP (2019)

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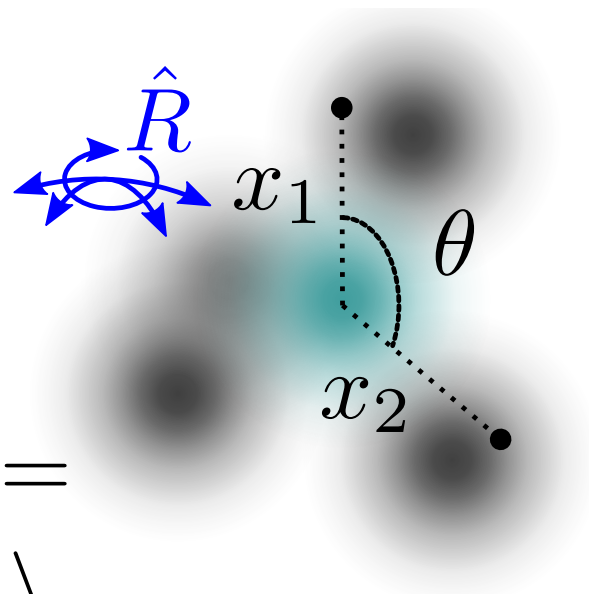

$$\langle x_1; x_2; \theta | A; \overline{\rho_i^{\otimes 2}} \rangle =$$
$$\int \mathrm{d}\hat{R} \langle x_1 \hat{R} \hat{\mathbf{e}}_z | A; \rho_i \rangle$$
$$\langle x_2 \hat{R} (\hat{\mathbf{e}}_z \cos\theta + \hat{\mathbf{e}}_x \sin\theta) | A; \rho_i \rangle$$

Willatt, Musil, **MC**, JCP (2019)

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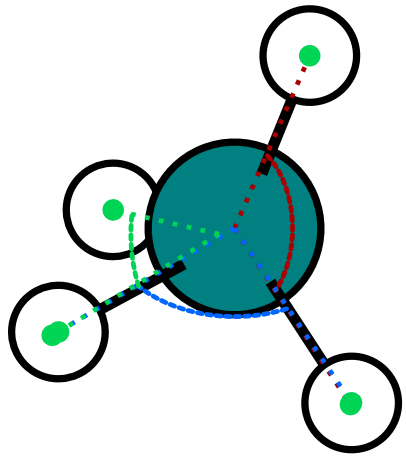
$$\langle x_1; x_2; \theta | A; \overline{\delta_i^{\otimes 2}} \rangle = \sum_{j_1 j_2} \delta(x_1 - r_{j_1 i}) \delta(x_2 - r_{j_2 i}) \delta(\cos\theta - \hat{\mathbf{r}}_{j_1 i} \cdot \hat{\mathbf{r}}_{j_2 i})$$



Willatt, Musil, **MC**, JCP (2019)

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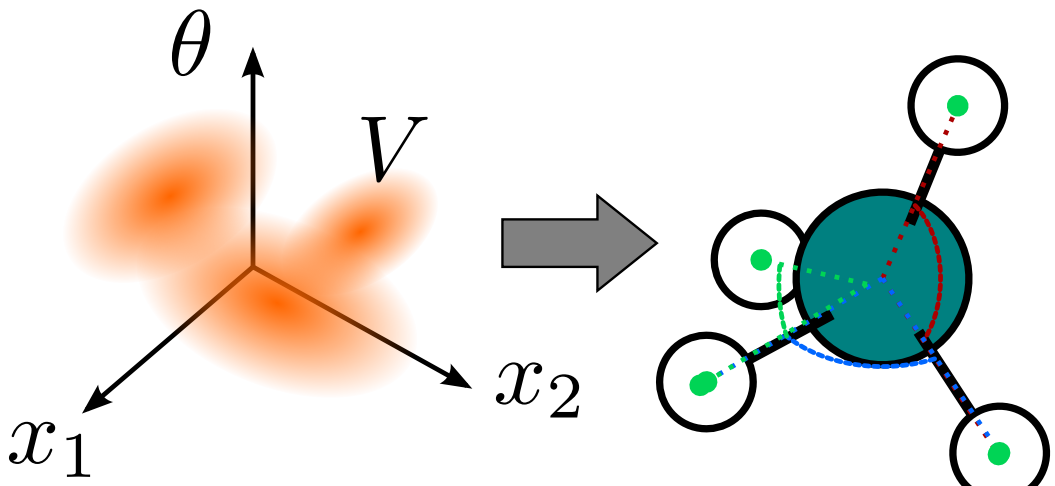
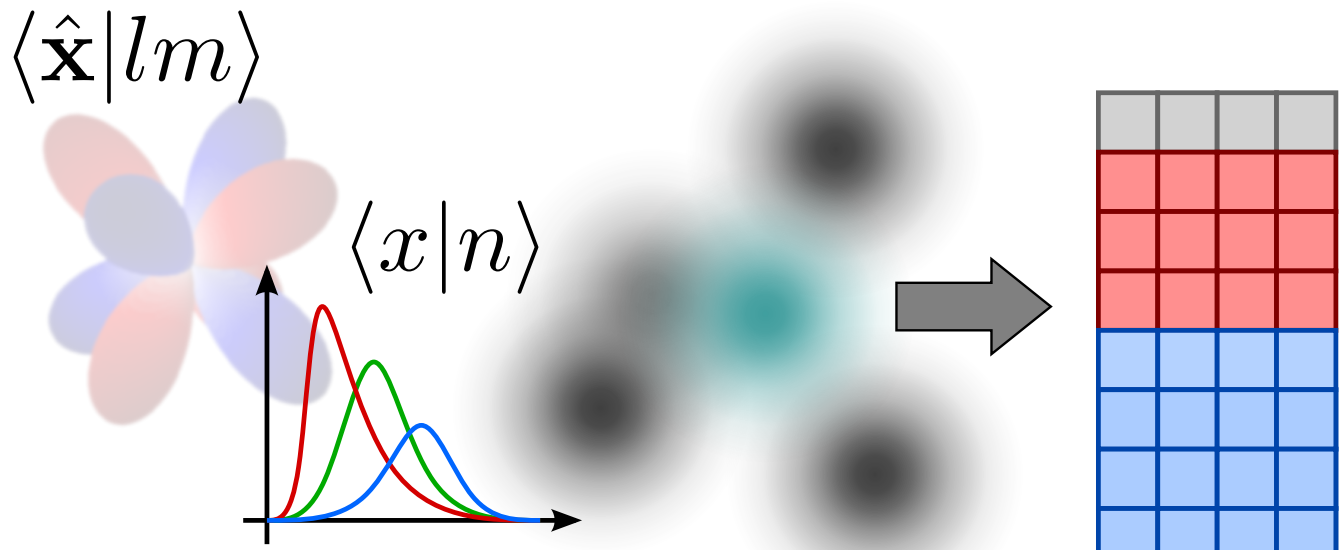


$$\int \langle V | x_1; x_2; \theta \rangle \langle x_1; x_2; \theta | A; \overline{\delta_i^{\otimes 2}} \rangle = \sum_{j_1 j_2} V\left(r_{j_1 i}, r_{j_2 i}, \hat{\mathbf{r}}_{j_1 i} \cdot \hat{\mathbf{r}}_{j_2 i}\right)$$

Willatt, Musil, **MC**, JCP (2019)

Density trick in an $\langle nlm|$ 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 $\langle \hat{\mathbf{x}}|lm\rangle \equiv Y_l^m(\hat{\mathbf{x}})$
  - Spherical harmonics transform linearly under rotations based on Wigner rotation matrices $\mathbf{D}^l\left(\hat{R}\right)$
  - Orthogonality of Wigner matrices yields the SOAP powerspectrum



$$\langle nlm|A;\rho_i\rangle = \int \mathrm{d}\mathbf{x}\langle n|x\rangle\langle lm|\hat{\mathbf{x}}\rangle\langle \mathbf{x}|A;\rho_i\rangle$$

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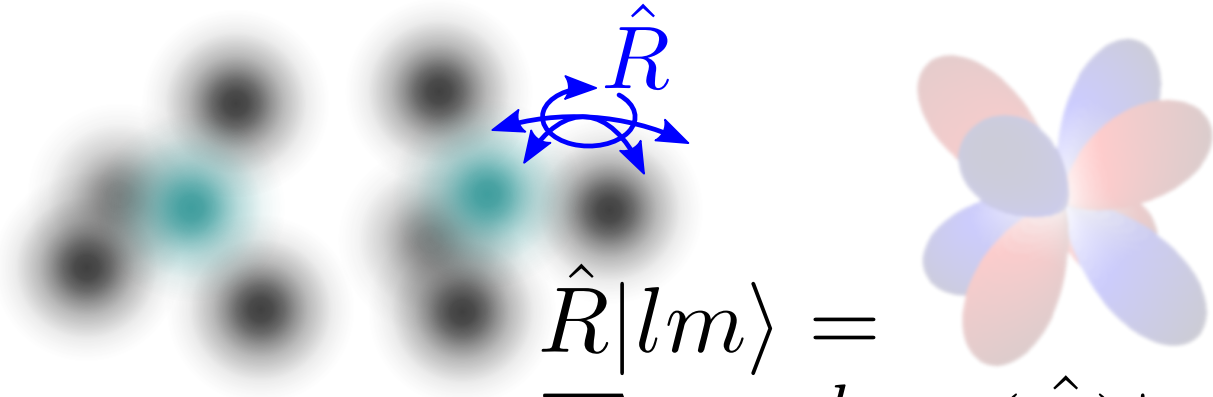

$$\hat{R}|lm\rangle = \sum_{m'} D^l_{mm'}(\hat{R})|lm'\rangle$$

$$\langle nlm; n'l'm'|A; \overline{\rho_i^{\otimes 2}}\rangle = \int \mathrm{d}\hat{R}\langle nlm|\hat{R}A; \rho_i\rangle\langle n'l'm'|\hat{R}A; \rho_i\rangle$$

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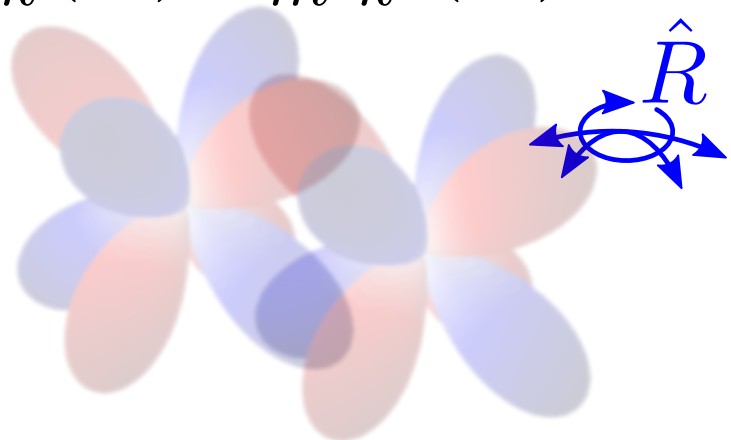
$$\int \mathrm{d}\hat{R} \sum_{kk'} D^l_{mk}(\hat{R}) D^{l'}_{m'k'}(\hat{R}) \propto \delta_{ll'}\delta_{mm'}\delta_{kk'}$$



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A hierarchy of equivariant features

- A generalization of the definition yields *N*-body features that transform like angular momenta

$$\langle X|\overline{\rho_i^{\otimes\nu};\sigma;\lambda\mu}\rangle$$

- Recursive construction based on sums of angular momenta and an expansion of the atom density

$$\langle n_1\, l_1\, k_1|\overline{\rho_i^{\otimes 1};\lambda\mu}\rangle \equiv \langle n_1\lambda\,(-\mu)|\rho_i\rangle\,\delta_{l_1\lambda}\delta_{k_1\lambda}\delta_{\sigma 1} \equiv \langle n_1|\overline{\rho_i^{\otimes 1};\lambda\mu}\rangle$$

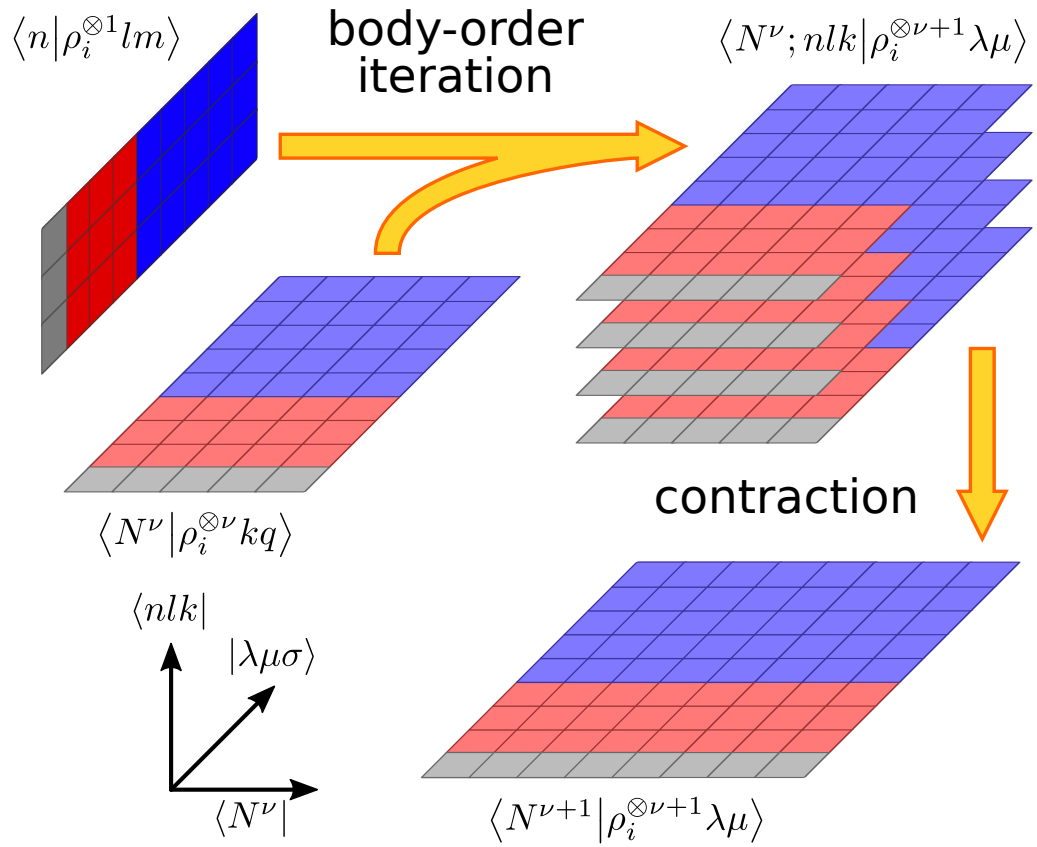
$$\langle \ldots;n_\nu\, l_\nu\, k_\nu;nlk|\overline{\rho_i^{\otimes(\nu+1)};\sigma;\lambda\mu}\rangle = \delta_{\sigma((-1)^{l+k+\lambda}s)}\,c_{k\lambda}\times \sum_{qm}\langle lm;kq|\lambda\mu\rangle\,\langle n|\overline{\rho_i^{\otimes 1};lm}\rangle\,\langle \ldots;n_\nu\, l_\nu\, k_\nu|\overline{\rho_i^{\otimes\nu};s;kq}\rangle$$

- Can be used to compute efficiently *invariant* features $|\overline{\rho_i^{\otimes\nu};0;00}\rangle$

Nigam, Pozdnyakov, **MC**, JCP (2020)

NICE features for ML

- Problem: number of features grows exponentially with ν
- 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 ν and contraction truncation



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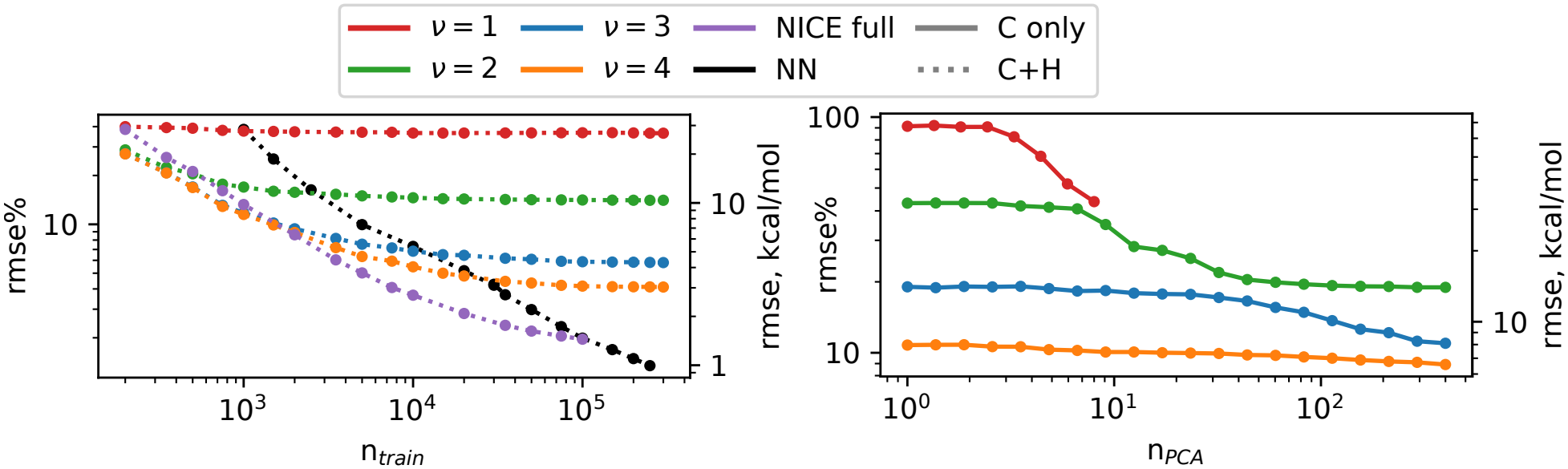
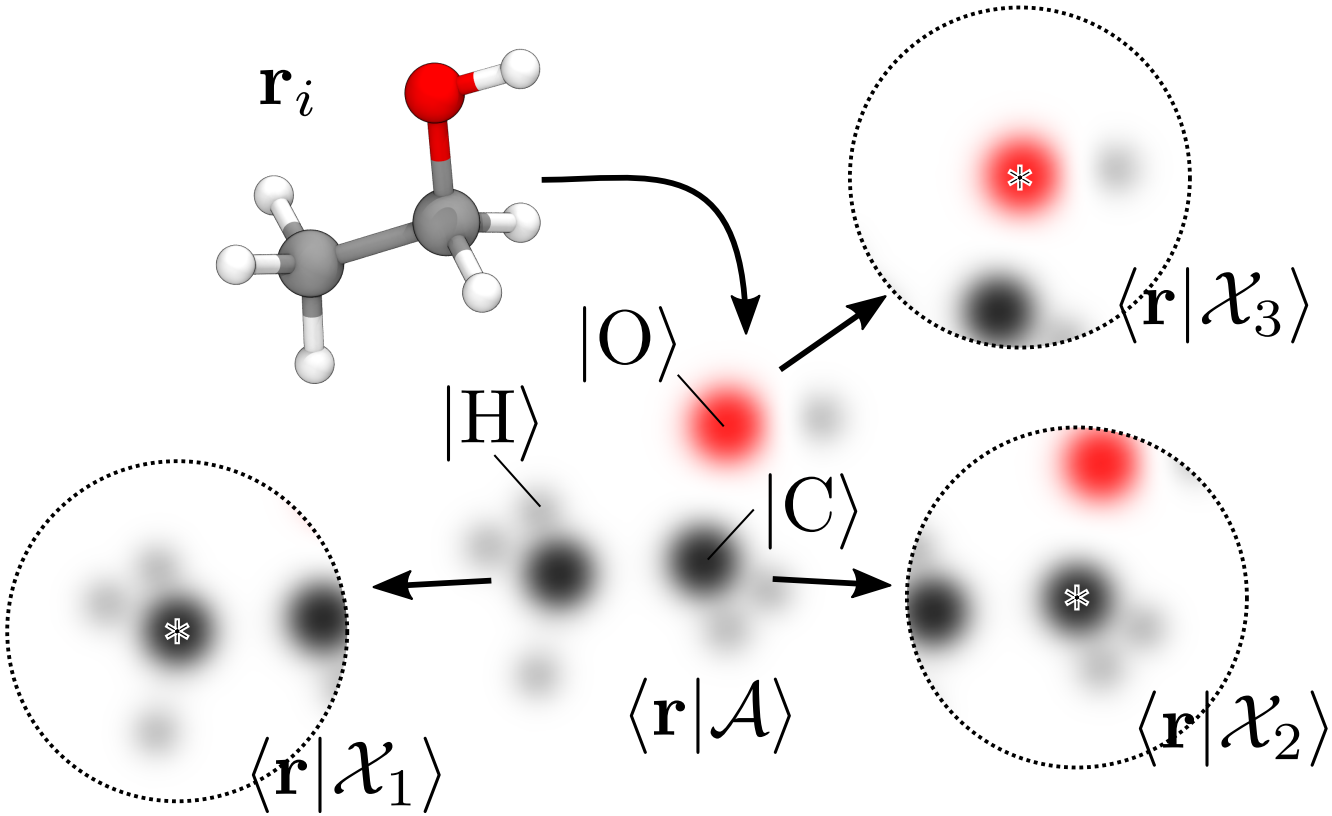


Nigam, Pozdnyakov, **MC**, JCP (2020)

Machine-learning the periodic table

- How to learn with multiple species? Decorate atomic Gaussian with elemental kets $|\mathrm{H}\rangle$, $|\mathrm{O}\rangle$, . . .
- 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!



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$$|\mathrm{H}\rangle = 0.5\,|🔥\rangle + 0.1\,|🪨\rangle + 0.2\,|💧\rangle$$
$$|\mathrm{C}\rangle = 0.2\,|🔥\rangle + 0.8\,|🪨\rangle + 0.3\,|💧\rangle$$
$$|\mathrm{O}\rangle = 0.1\,|🔥\rangle + 0.1\,|🪨\rangle + 0.6\,|💧\rangle$$

Empedocles et al. (ca 360BC). Metaphor courtesy of Albert Bartók

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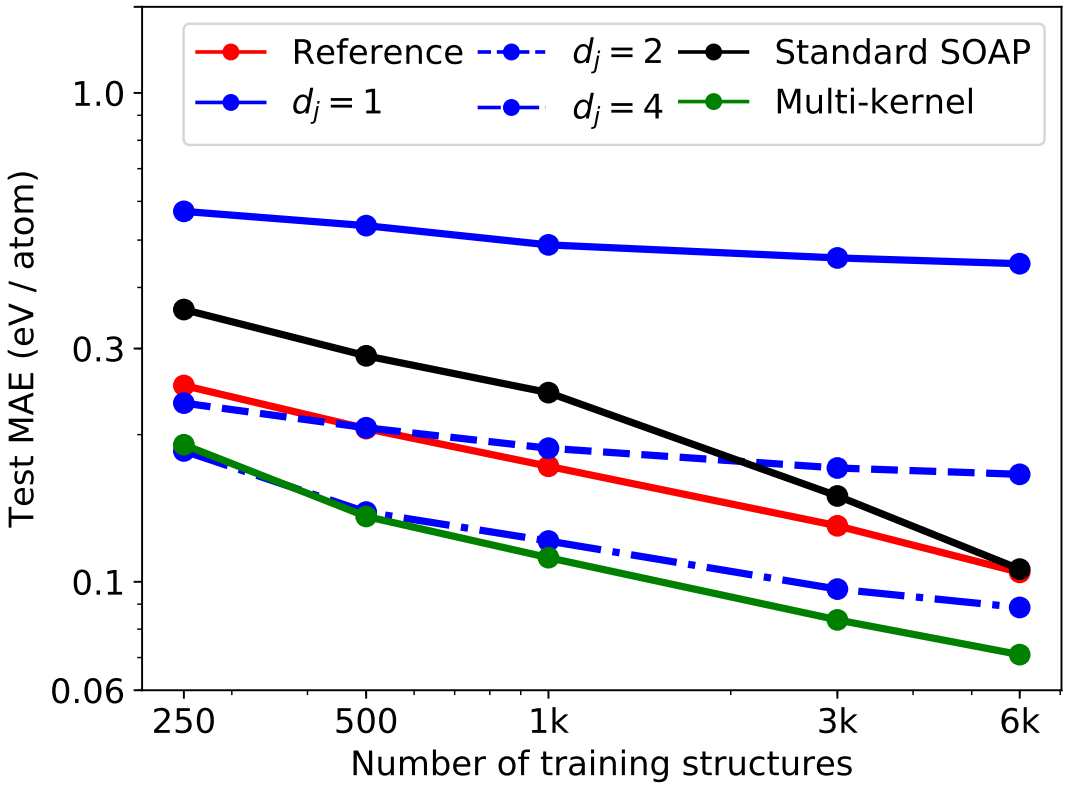

Elpasolite dataset. Reference curve (red) from Faber et al. JCP (2018)

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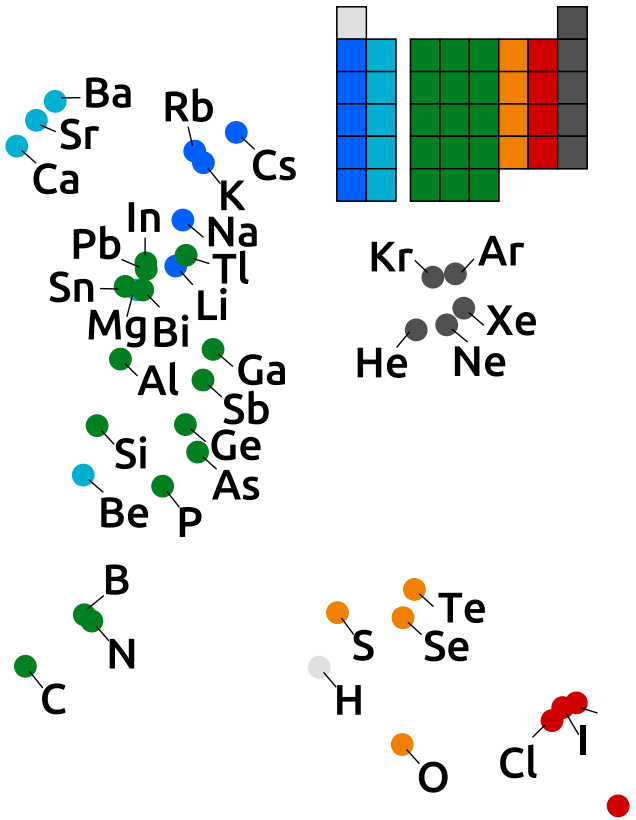
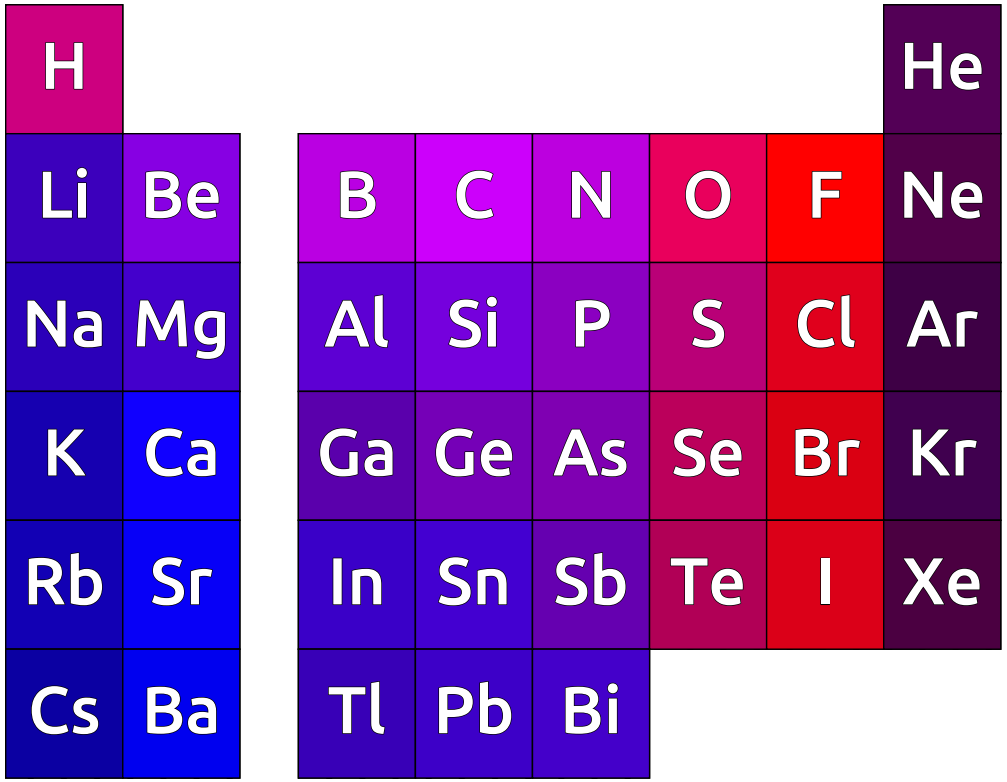
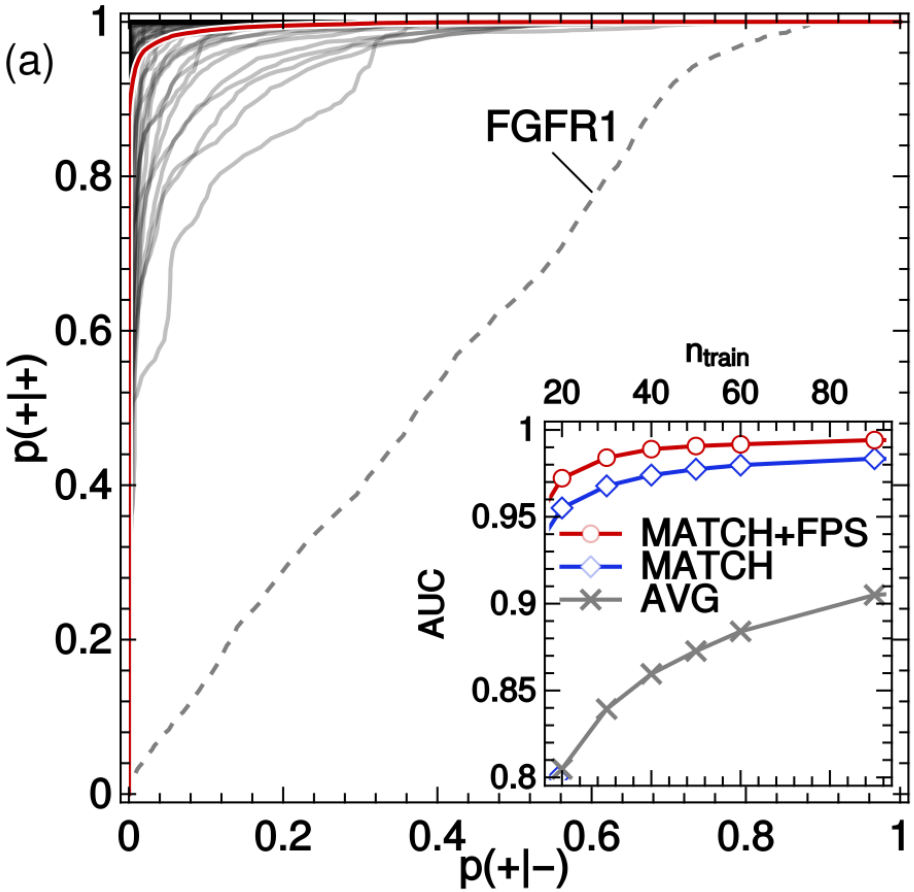


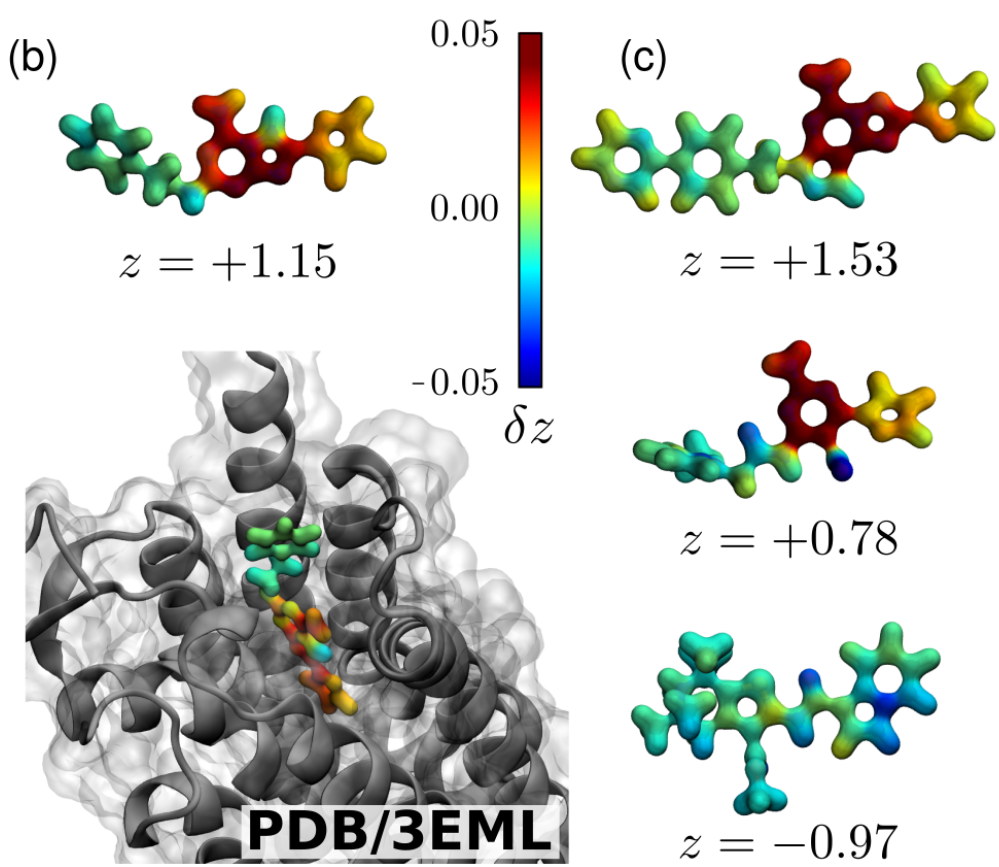


Willatt, Musil, Ceriotti, PCCP (2018)

Recognizing active protein ligands

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

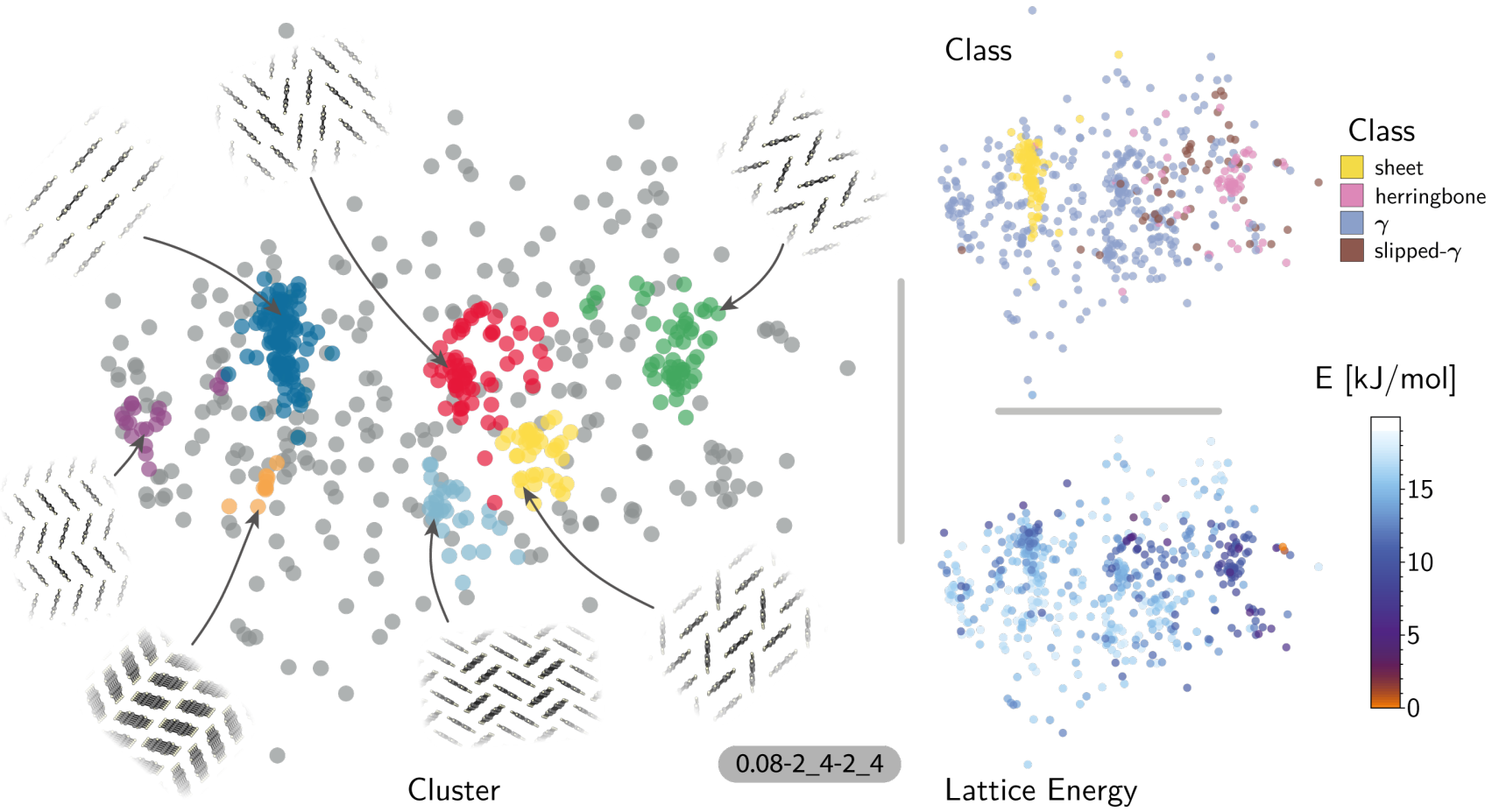




Bartok, De, Poelking, Kermode, Bernstein, Csanyi, **MC**, Science Advances (2017) *[data: DUD-E, Shoichet]*

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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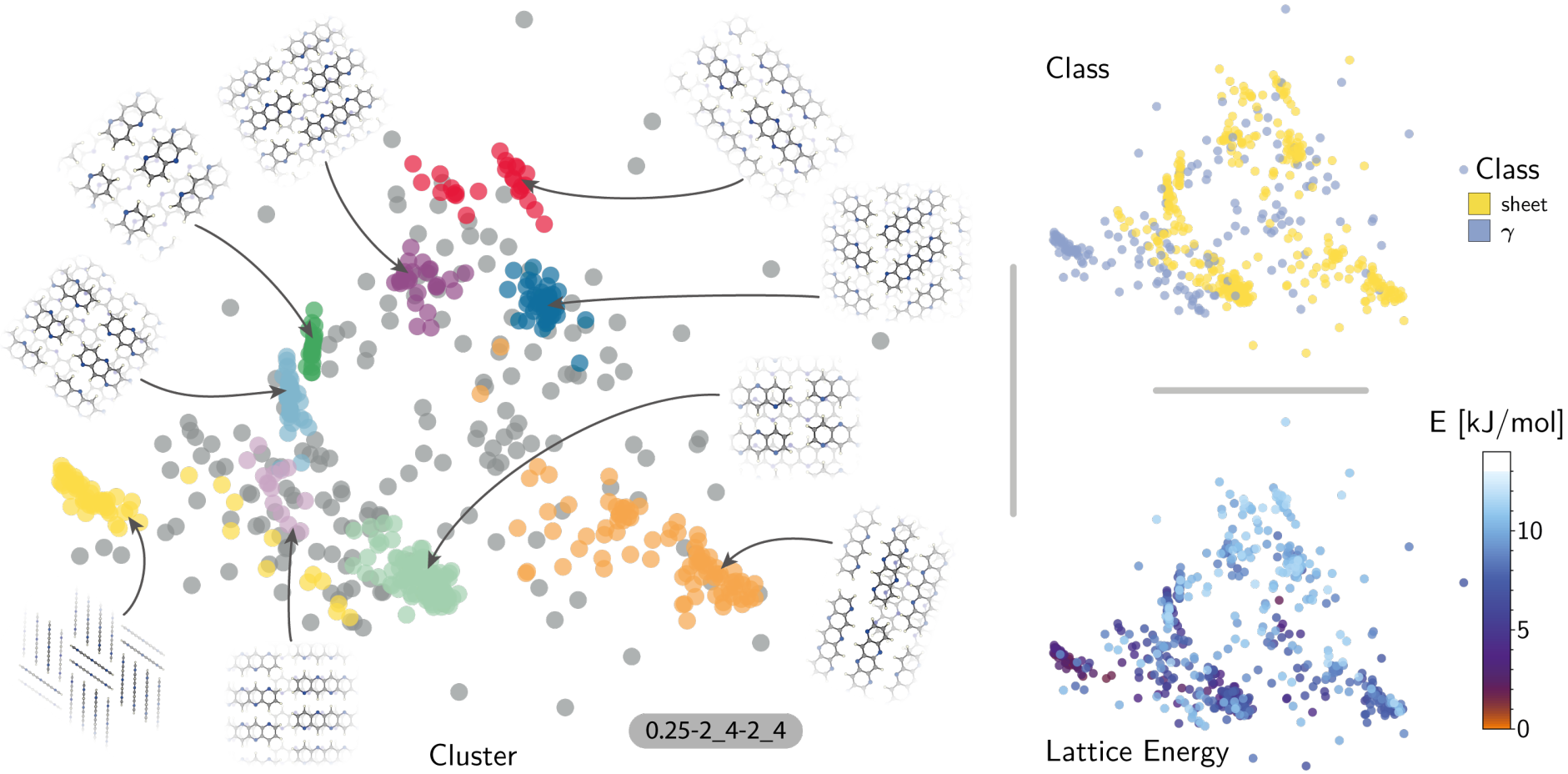

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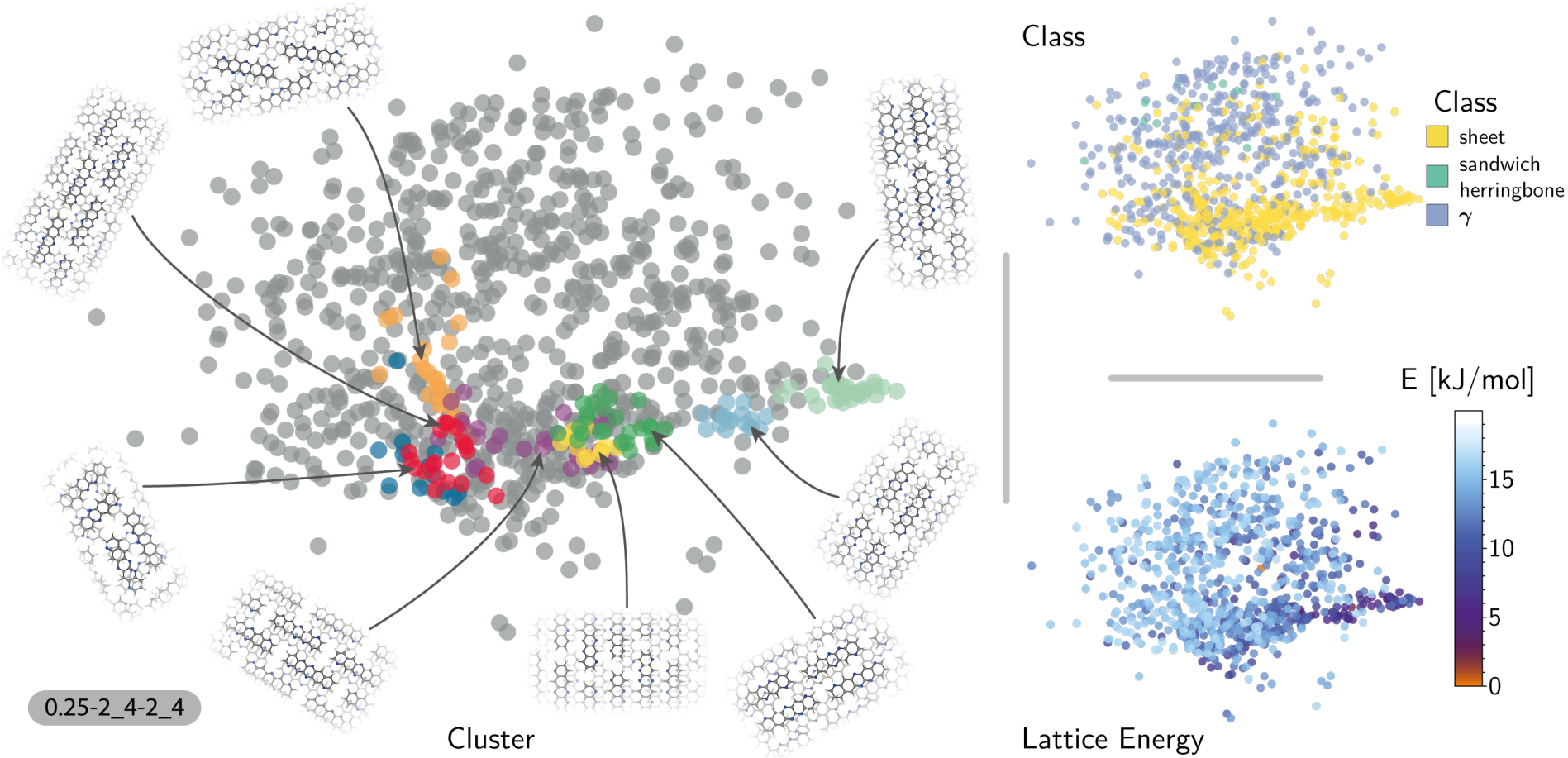


Musil, De, Yang, Campbell, Day, **MC**, Chemical Science (2018);`http://interactive.sketchmap.org`

Principal Covariates Regression

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

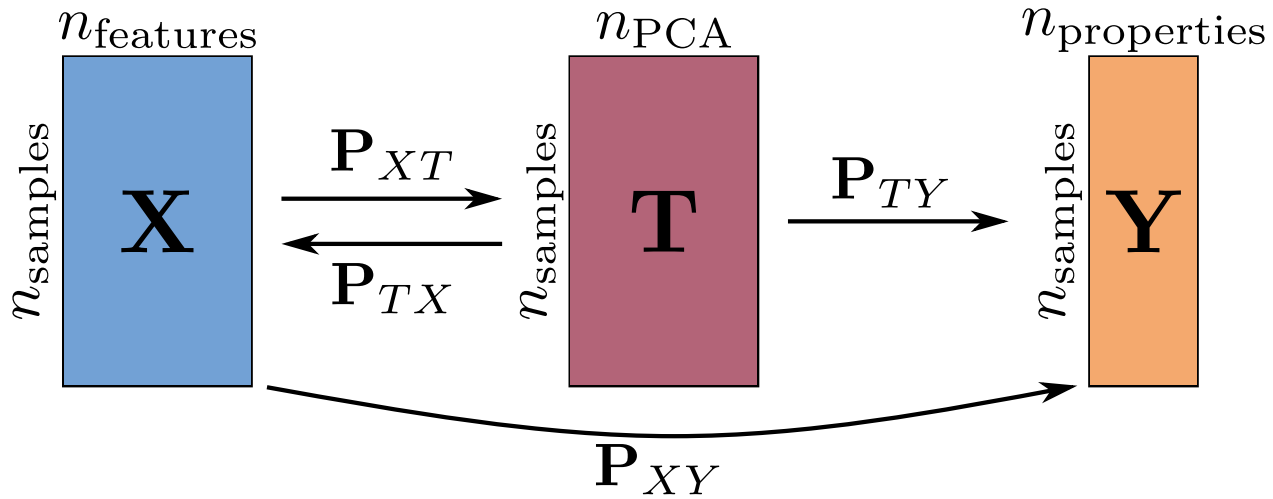
$$\ell = \alpha \left\| \mathbf{X} - \mathbf{X}\mathbf{P}_{XT}\mathbf{P}_{TX} \right\|^2 + (1-\alpha) \left\| \mathbf{Y} - \mathbf{X}\mathbf{P}_{XT}\mathbf{P}_{TY} \right\|^2$$

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

$$\tilde{\mathbf{K}} = \alpha \mathbf{X}\mathbf{X}^T + (1-\alpha)\, \mathbf{X}\mathbf{P}_{XY}\mathbf{P}_{XY}^T\mathbf{X}^T$$

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

$$\tilde{\mathbf{C}} = \alpha \mathbf{X}^T\mathbf{X} + (1-\alpha)\, \left(\mathbf{X}^T\mathbf{X}\right)^{-1/2} \mathbf{X}^T\mathbf{Y}\mathbf{Y}^T\mathbf{X} \left(\mathbf{X}^T\mathbf{X}\right)^{-1/2}$$

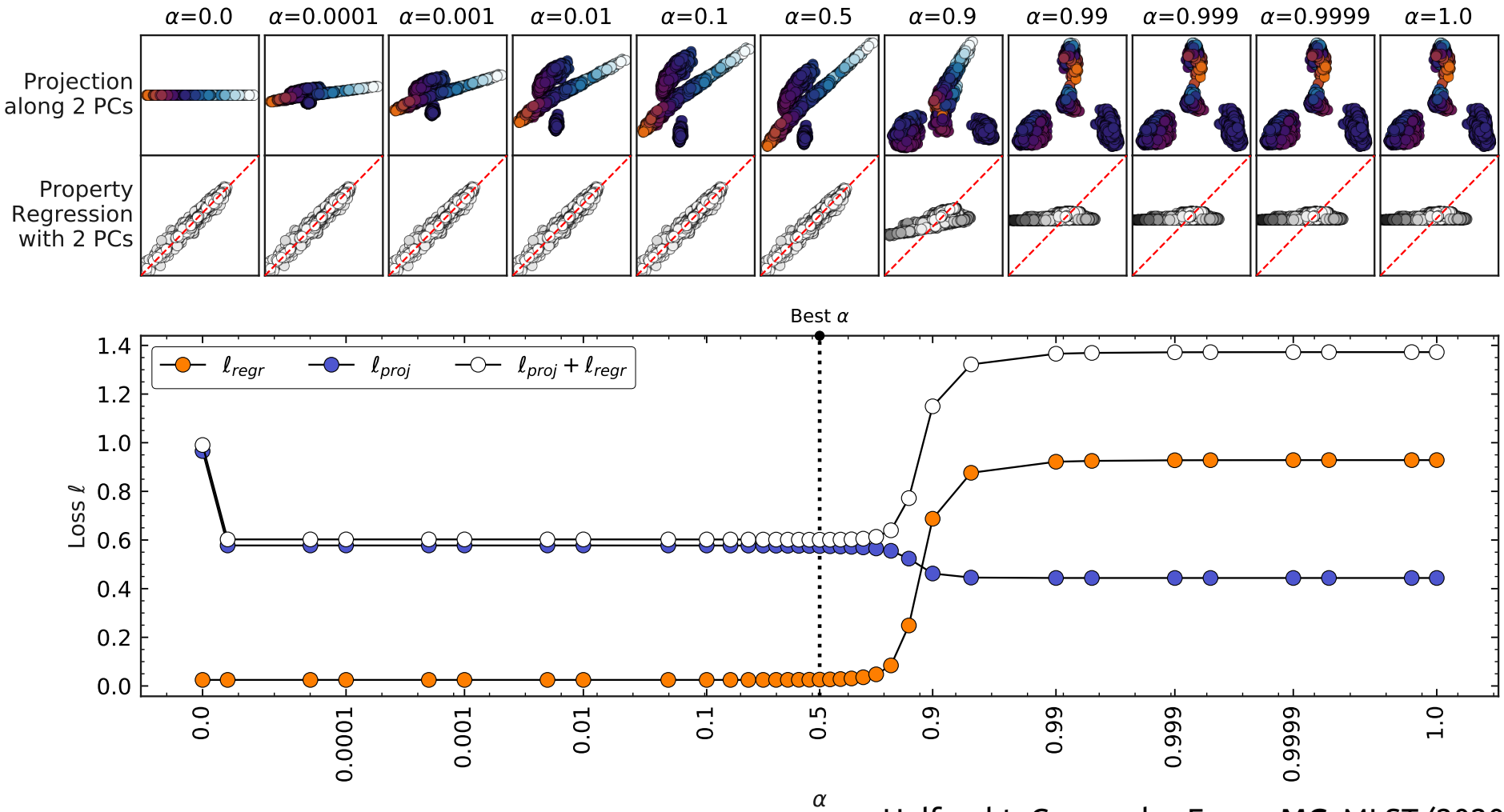


S. de Jong and HAL Kiers, Scandinavian Symposium on Chemometrics (1992)

Kernel PCovR

- Kernel versions of PCovR can be obtained with a modified kernel $\tilde{\mathbf{K}} = \alpha\mathbf{K} + (1-\alpha)\,\hat{\mathbf{Y}}\hat{\mathbf{Y}}^T$, diagonalizing it and finding the projector

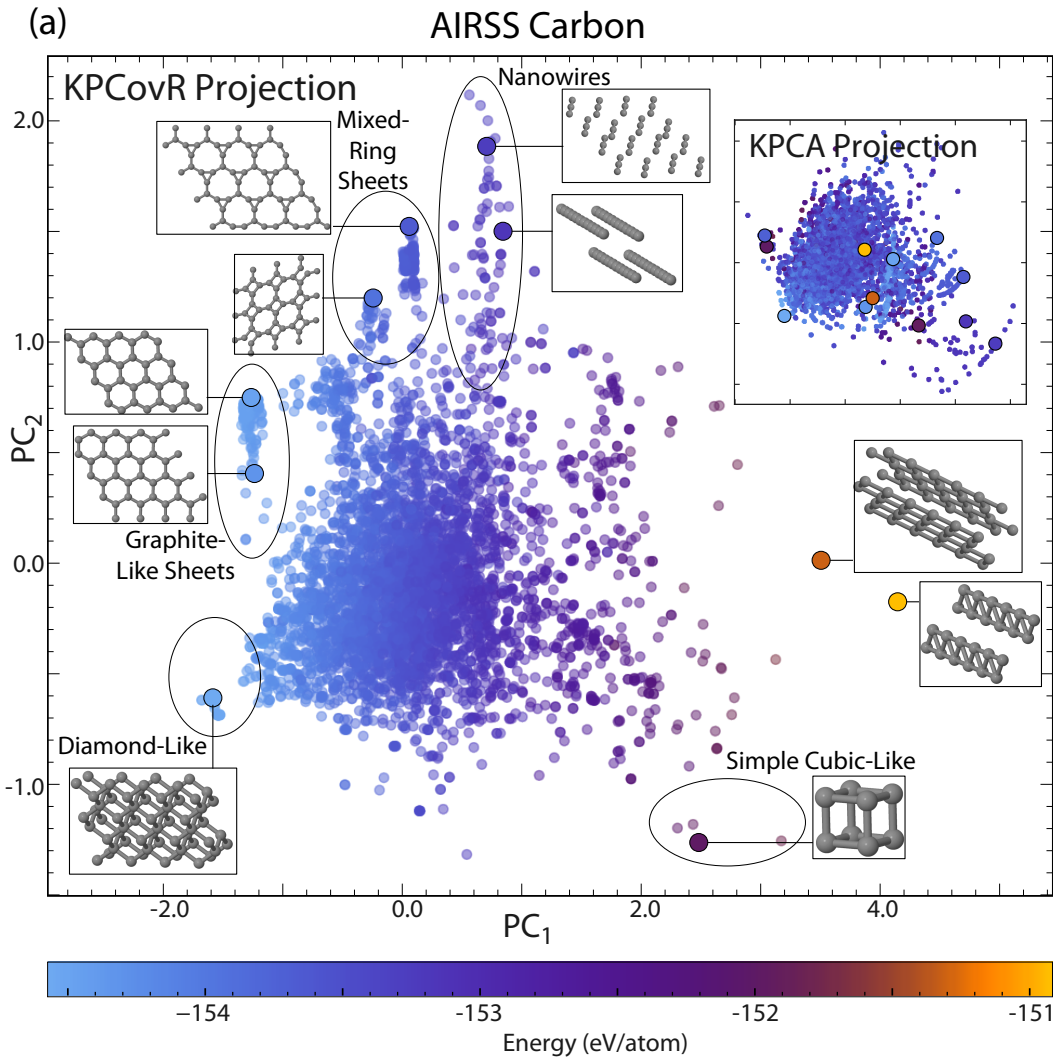
$$\mathbf{P}_{\mathbf{K}T} = \left(\alpha\mathbf{I} + (1-\alpha)\left(\mathbf{K} + \lambda\mathbf{I}\right)^{-1}\mathbf{Y}\hat{\mathbf{Y}}\right)\mathbf{U}_{\tilde{\mathbf{K}}}\Lambda_{\tilde{\mathbf{K}}}^{1/2}$$



Helfrecht, Cersonsky, Fraux, **MC**, MLST (2020)

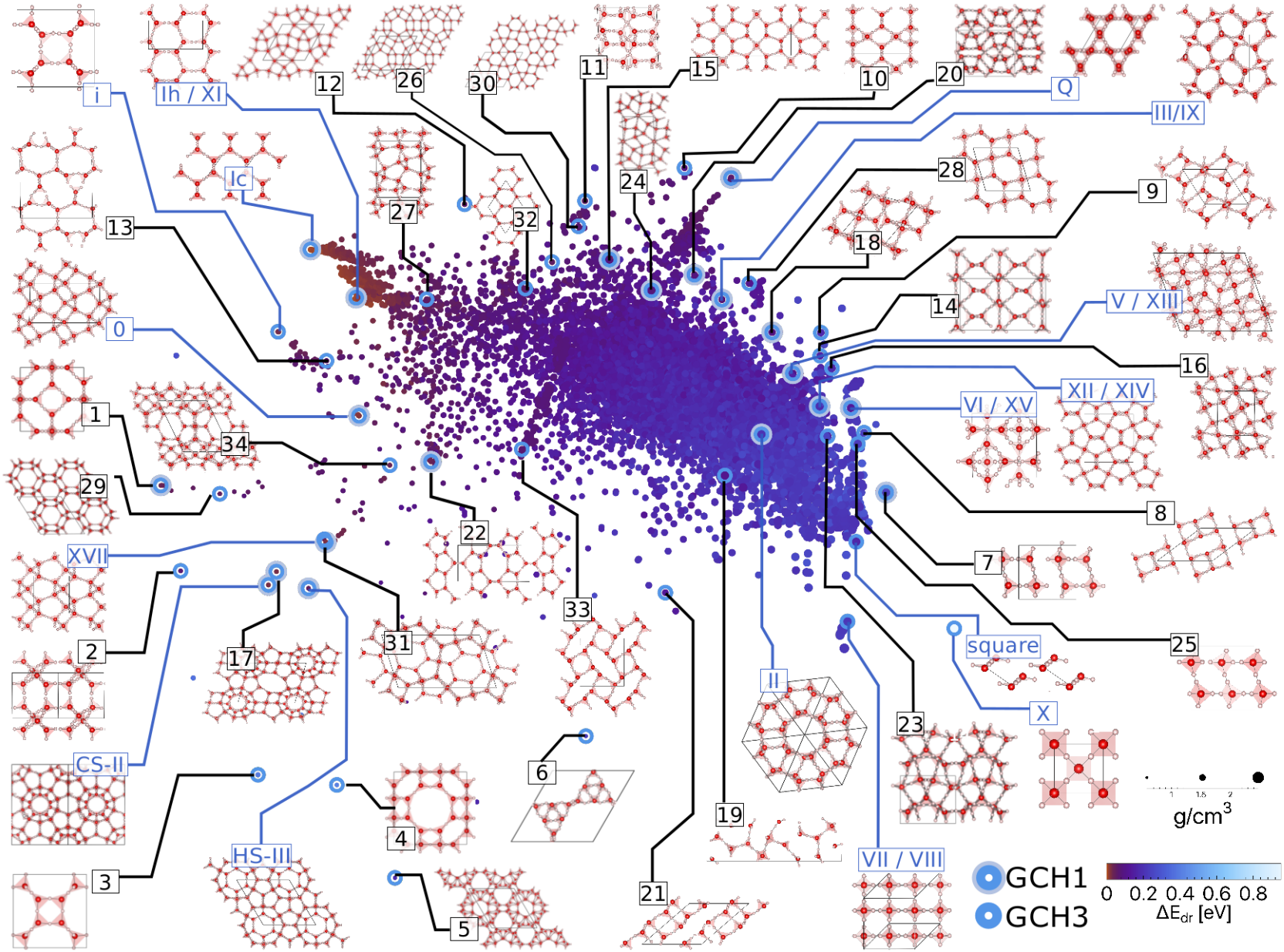
Where unsupervised meets supervised

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



Helfrecht, Cersonsky, Fraux, **MC**, MLST (2020)

A Generalized Convex Hull Construction



Anelli, Engel, Pickard & **MC**, PRM (2019); Engel, Anelli, **MC**, Pickard & Needs, Nature Comm. (2018)