Symmetry Matters: Machine Learning of Scalar and Tensorial Atomic-Scale Properties

Michele Ceriotti
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SIAM, Portland
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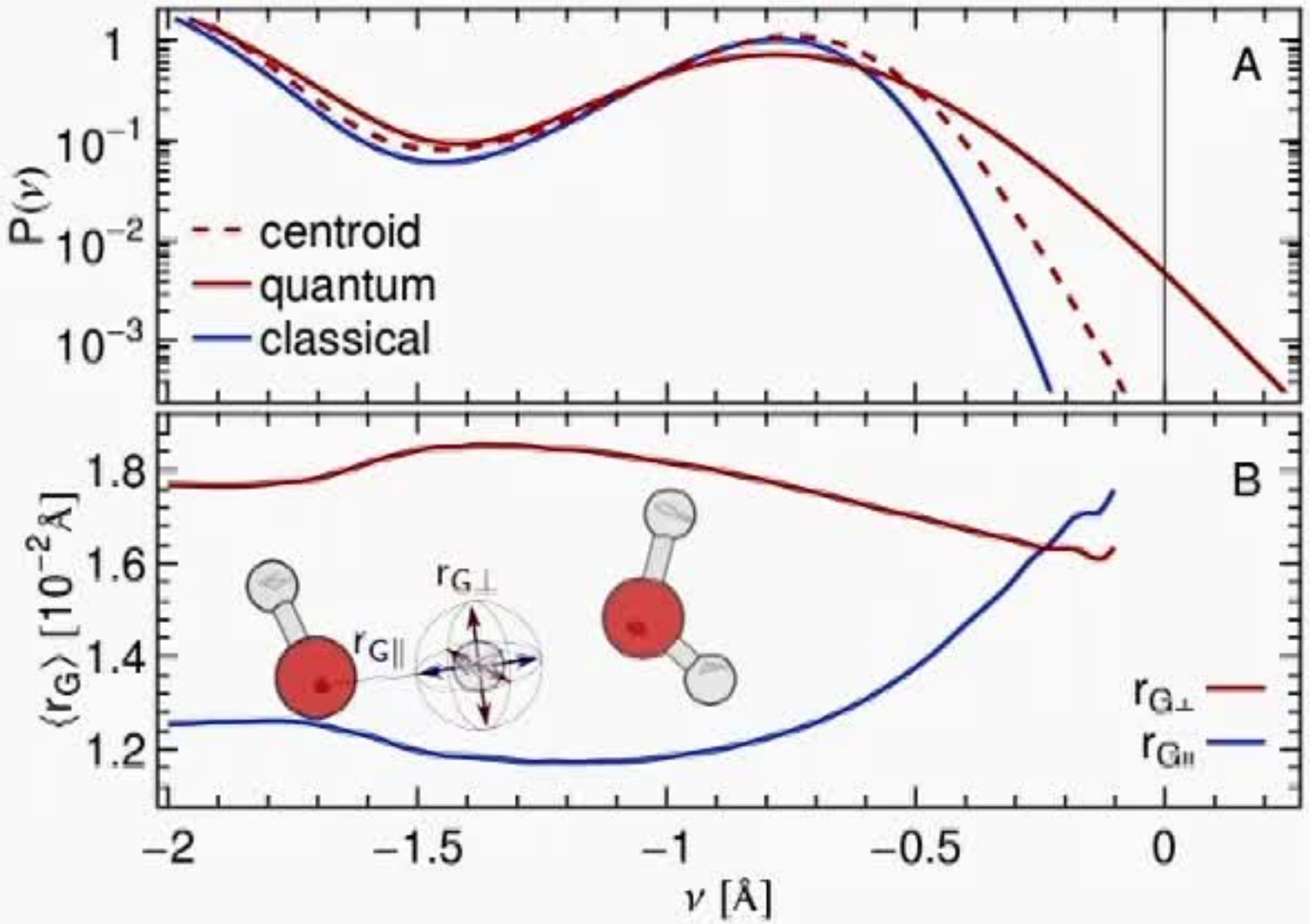
FNSNF

S.De, F.Musil, M.Willatt
A.Grisafi, D.Wilkins

G.Csányi, A.Bartók, C.Poelking,
J.Kermode, N.Bernstein
F.Paruzzo, A.Hofstetter, L.Emsley
C.Corminboeuf, J.Behler, A.Paxton

Bringing Fluctuations into Materials Modelling

- Quantum nuclei with path integral methods
- Anharmonic free energies in solids
- Activated events and phase transitions



Li, Walker, Michaelides, PNAS (2011); **MC** et al., PNAS (2013); **MC** et al., Chem. Rev. (2016)

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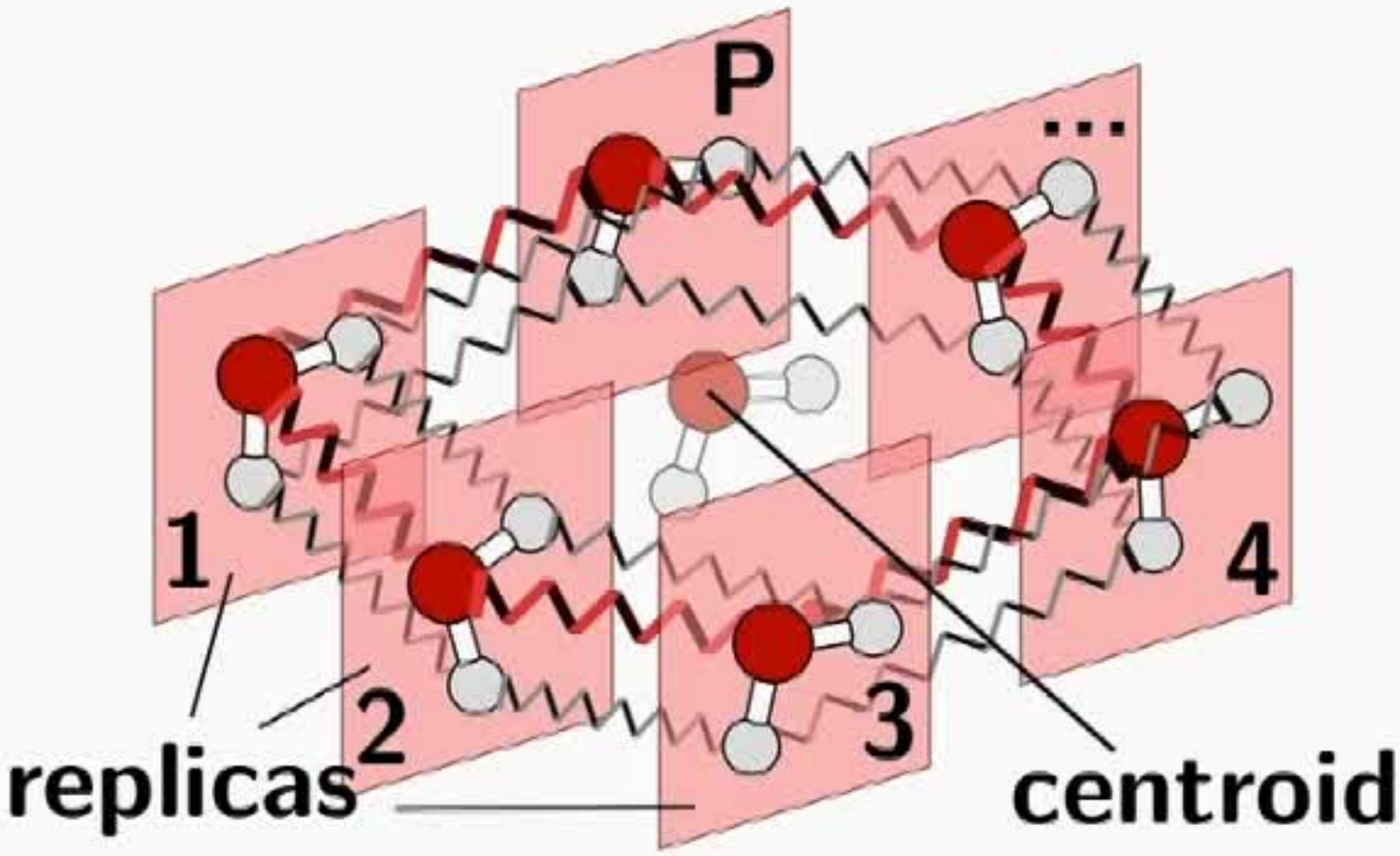

Markland & **MC**, Nat. Rev. Chem. (2018); http://ipi-code.org

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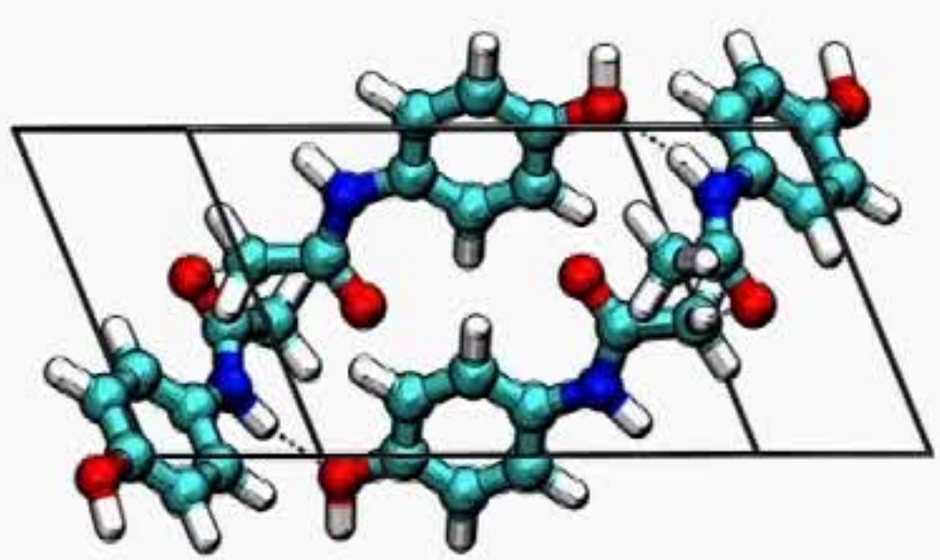

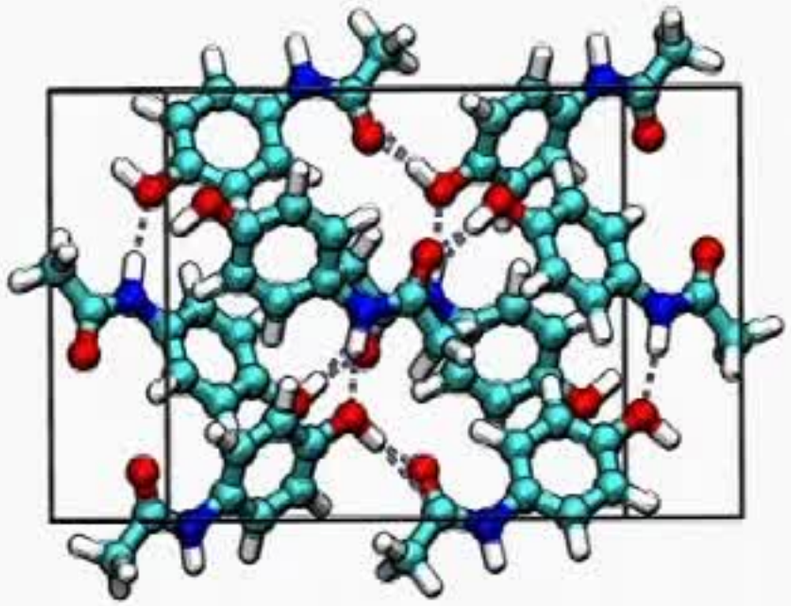

Binding Energy (meV/mol)	fI	fII	ΔΔ
Lattice Energy (PBE+D3)	-1492	-1489	-3
Lattice Energy (PBE0+D3)	-1271	-1271	0
Classical Harmonic	-1500	-1487	-13
Quantum Anharmonic	**-1152**	**-1107**	**-45**

Rossi, Gasparotto & **MC**, Phys. Rev. Lett. (2016)

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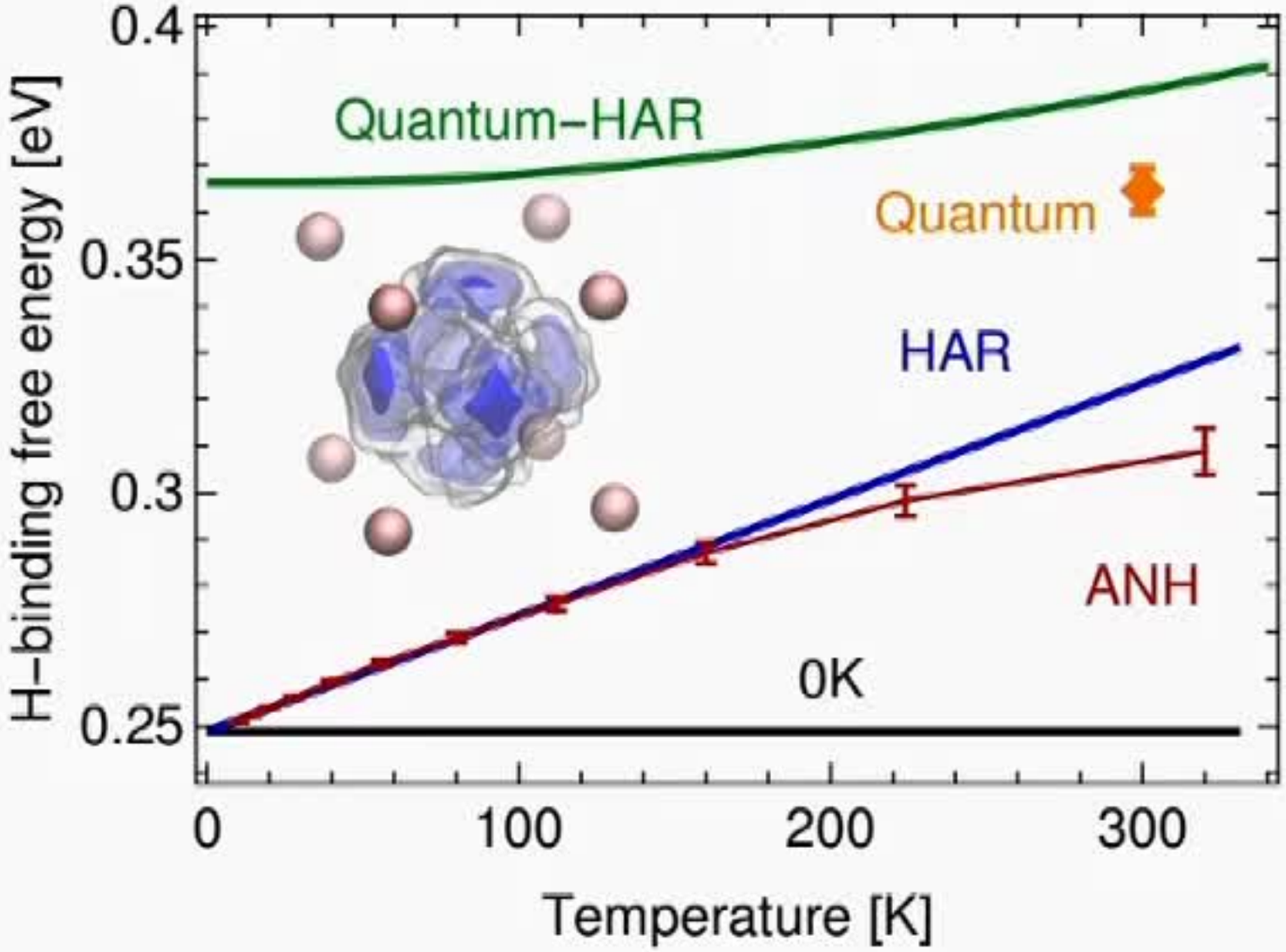

Cheng, Paxton, **MC**, Phys. Rev. Lett. (2018)

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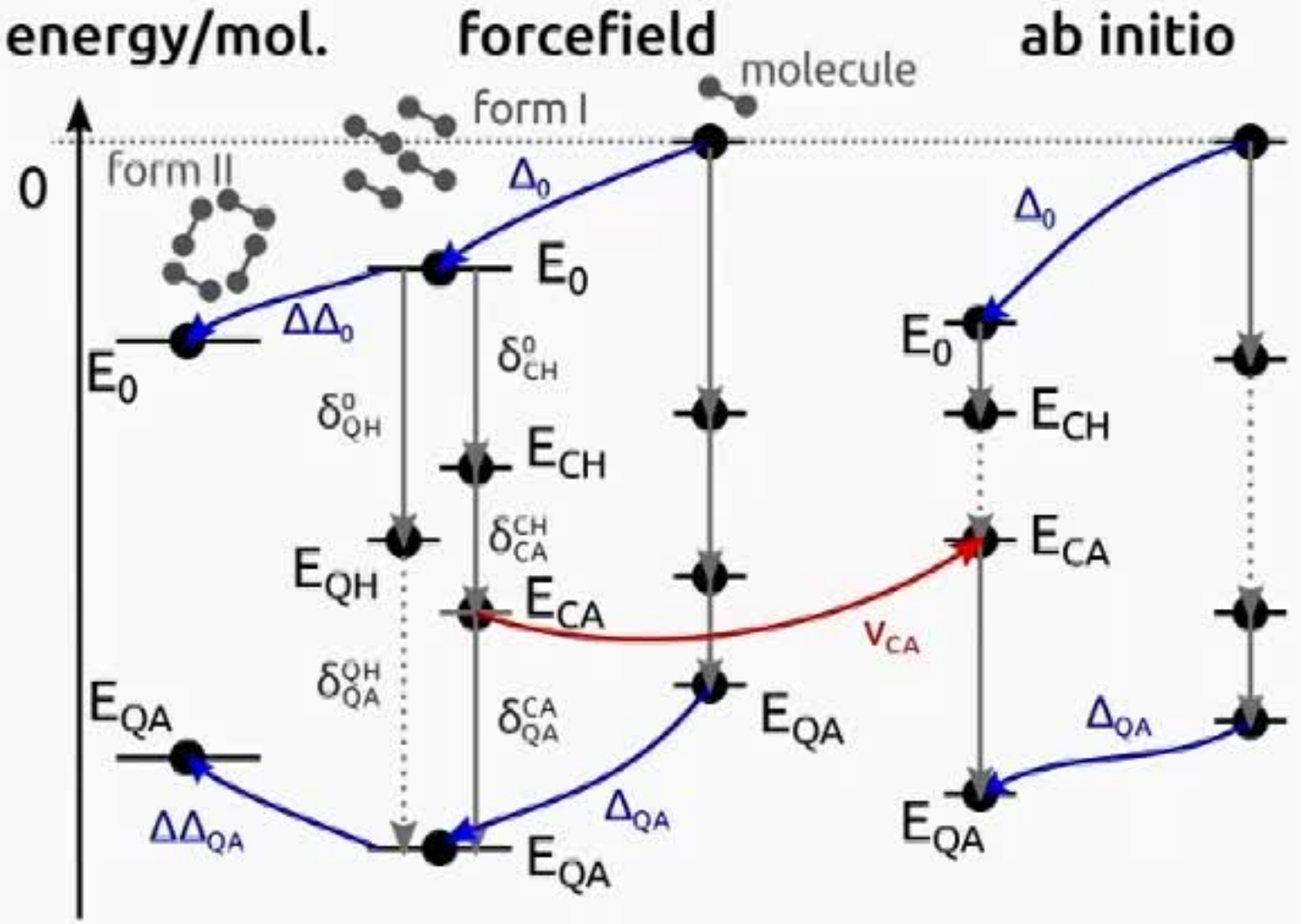

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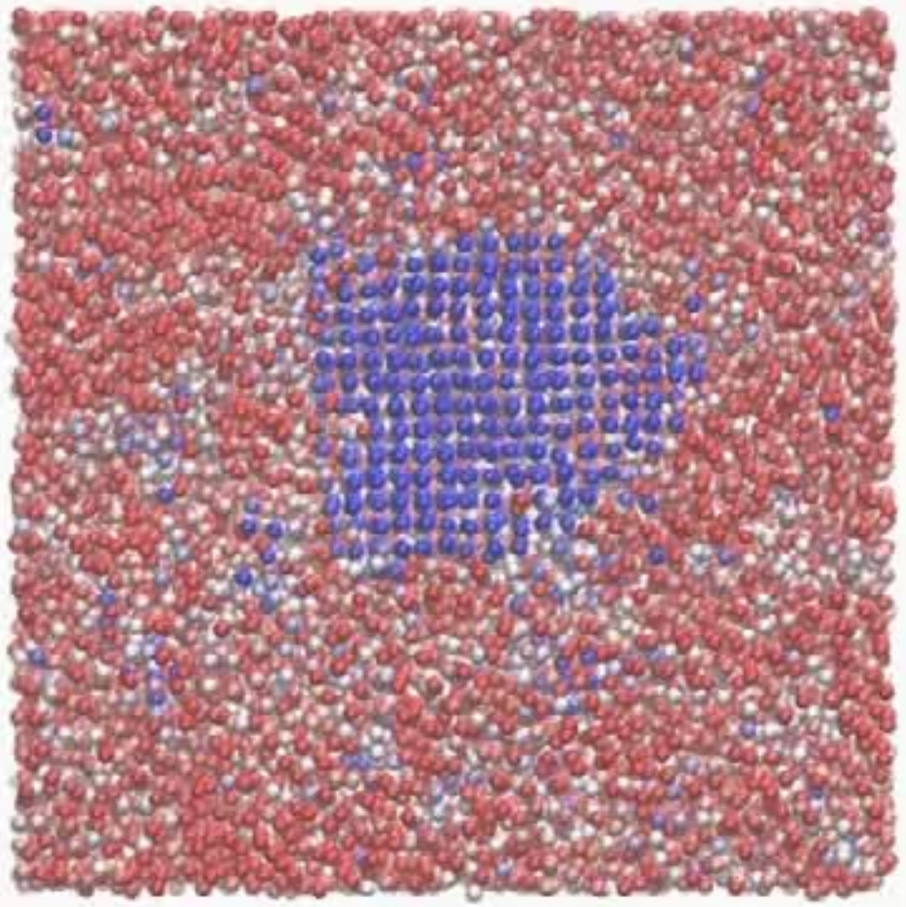
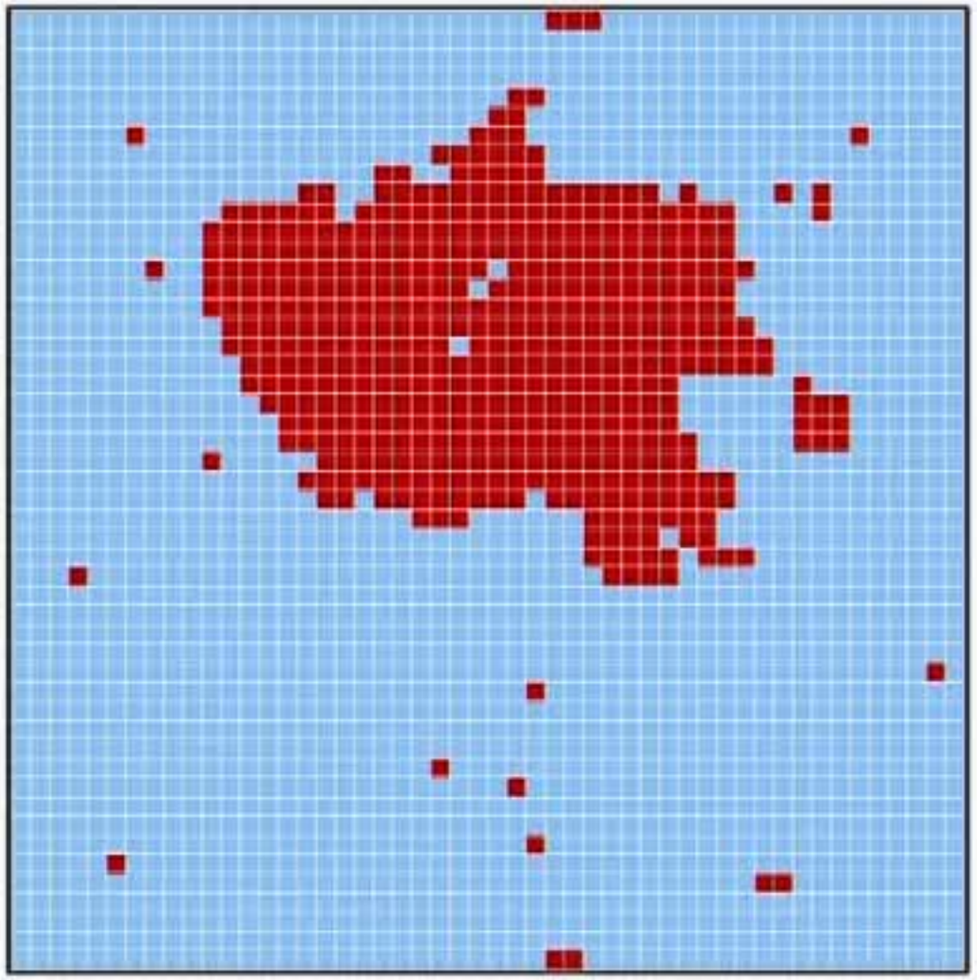
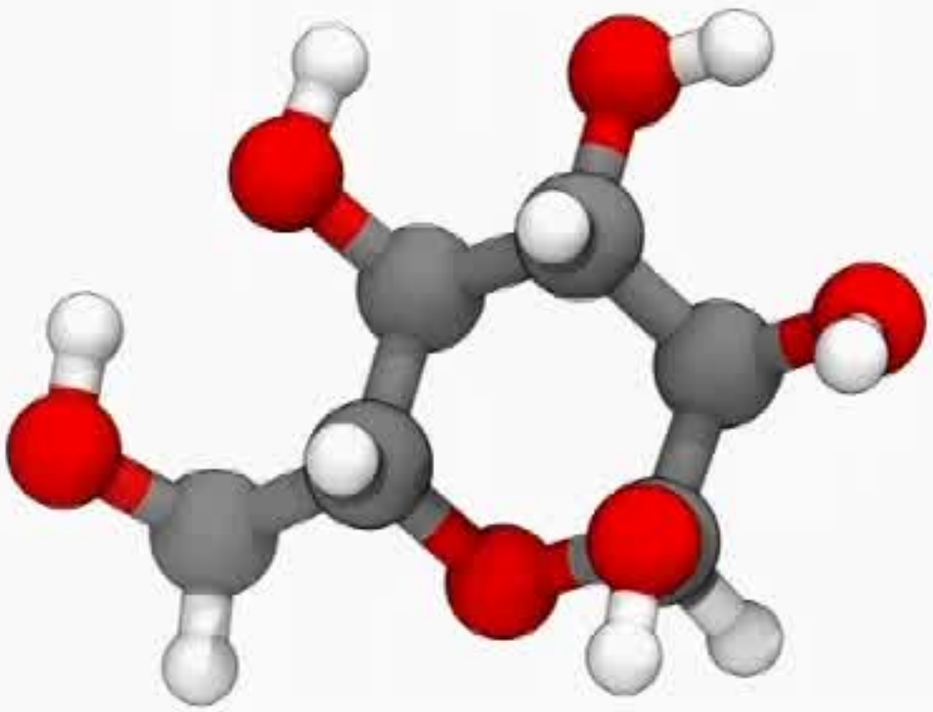


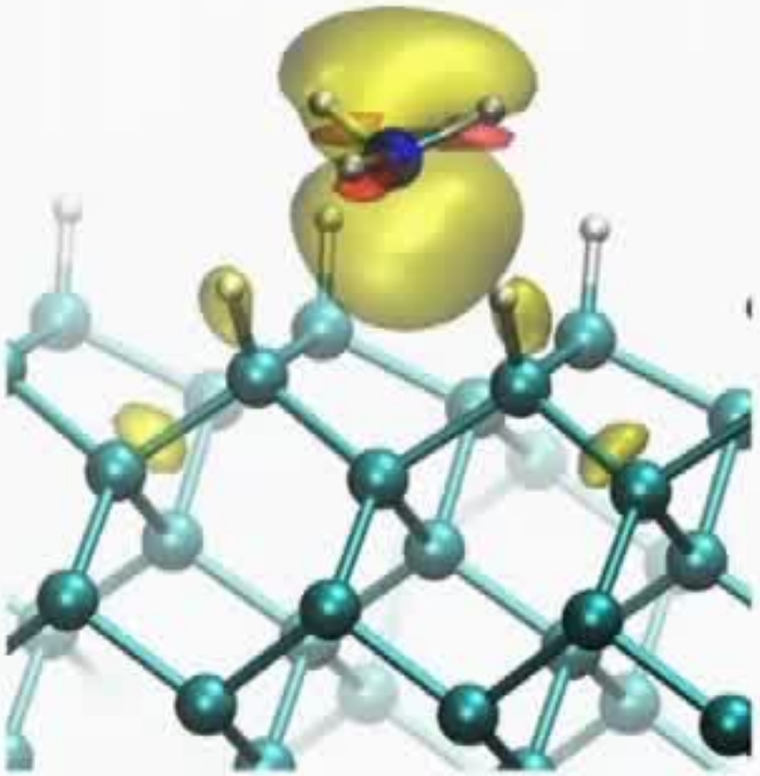


Cheng & **MC**, J. Chem. Phys. (2017)

Machine Learning for Atomic-Scale Properties

- An alternative to quantum mechanics and empirical models
- Predict atomic-scale properties based on coordinates and nature of the atoms, following training on reference configurations
- Well-principled: incorporates structure and symmetries of physical laws
- Not only a fancy interpolator: use ML to gain insights and understanding





$$E(\mathbf{q}) = ML(\mathbf{q}|\{\mathbf{q}_i, V_i\})$$

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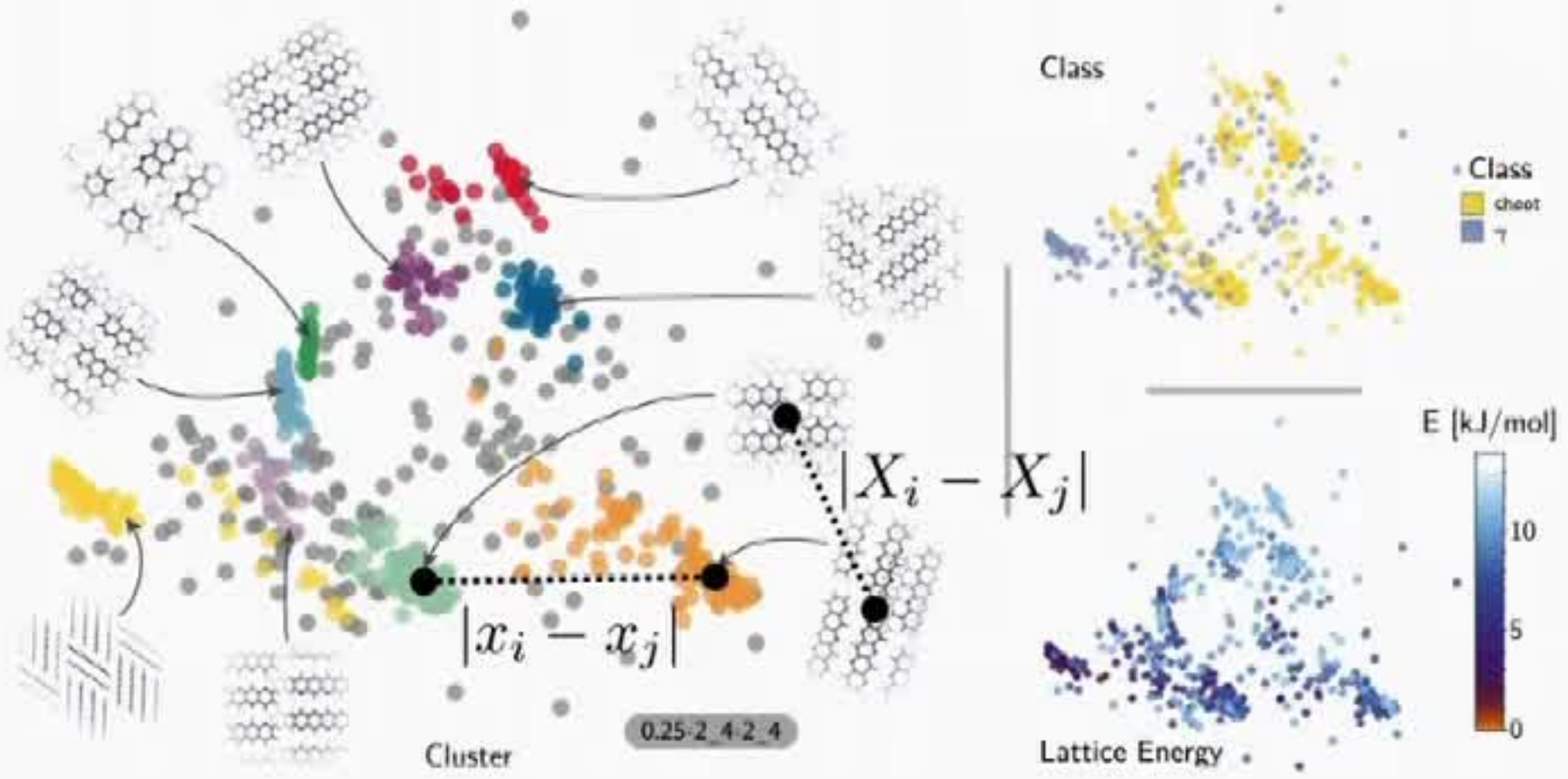
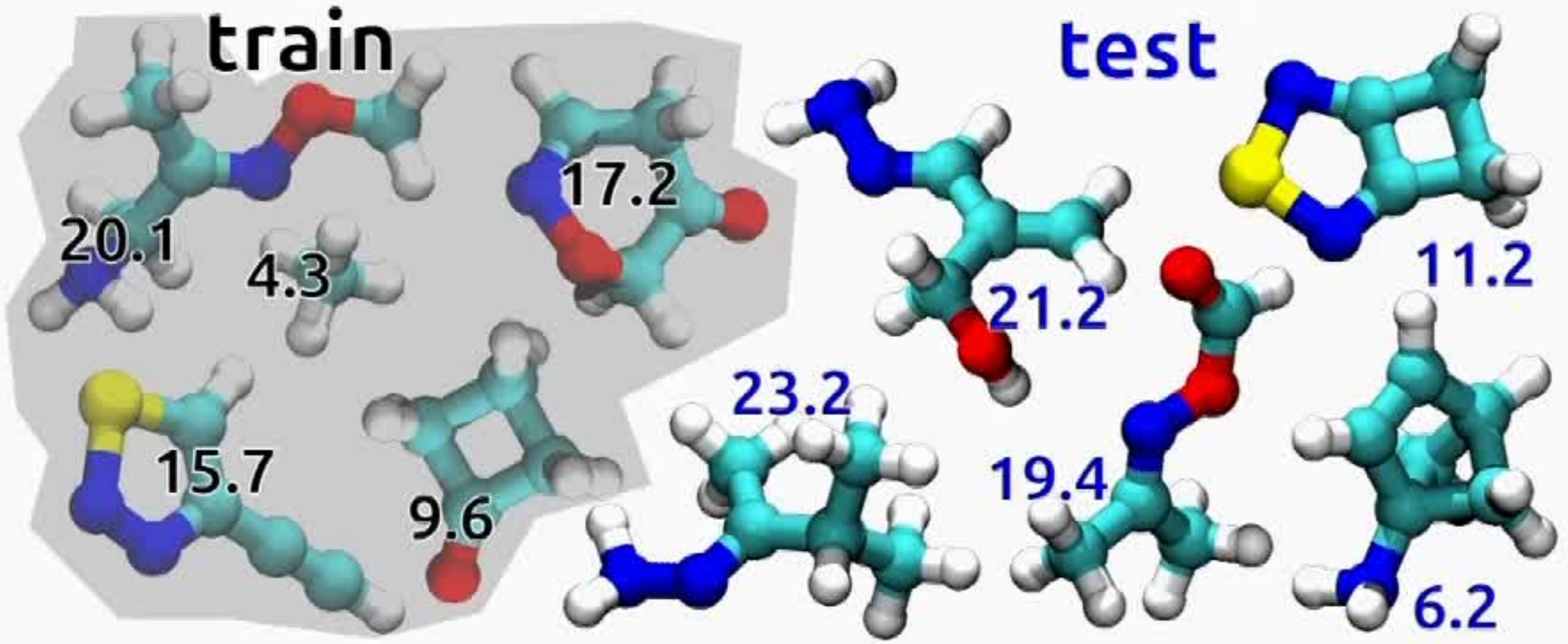


$$\{X_i\} \Rightarrow \{x_i\} \qquad \{x_i\} = \text{argmin} \sum_{ij} \left[s\left(|X_i - X_j|\right) - s\left(|x_i - x_j|\right) \right]^2$$

Musil, De, Yang, Campbell, Day, **MC**, Chemical Science (2018)

Machine-Learning as a Universal Interpolator

- Machine-learning can be regarded as a sophisticated interpolation between a few known values of the properties
- Can it be made as accurate and general as the Schrödinger equation?
- Kernels are the main ingredient. Think of them as scalar products between vectors that represent structures, $K(\mathcal{A},\mathcal{B}) \sim \langle \mathcal{A}|\mathcal{B}\rangle$.



$$E(\mathcal{A}) = \sum_i w_i K(\mathcal{A}, \mathcal{A}_i)$$

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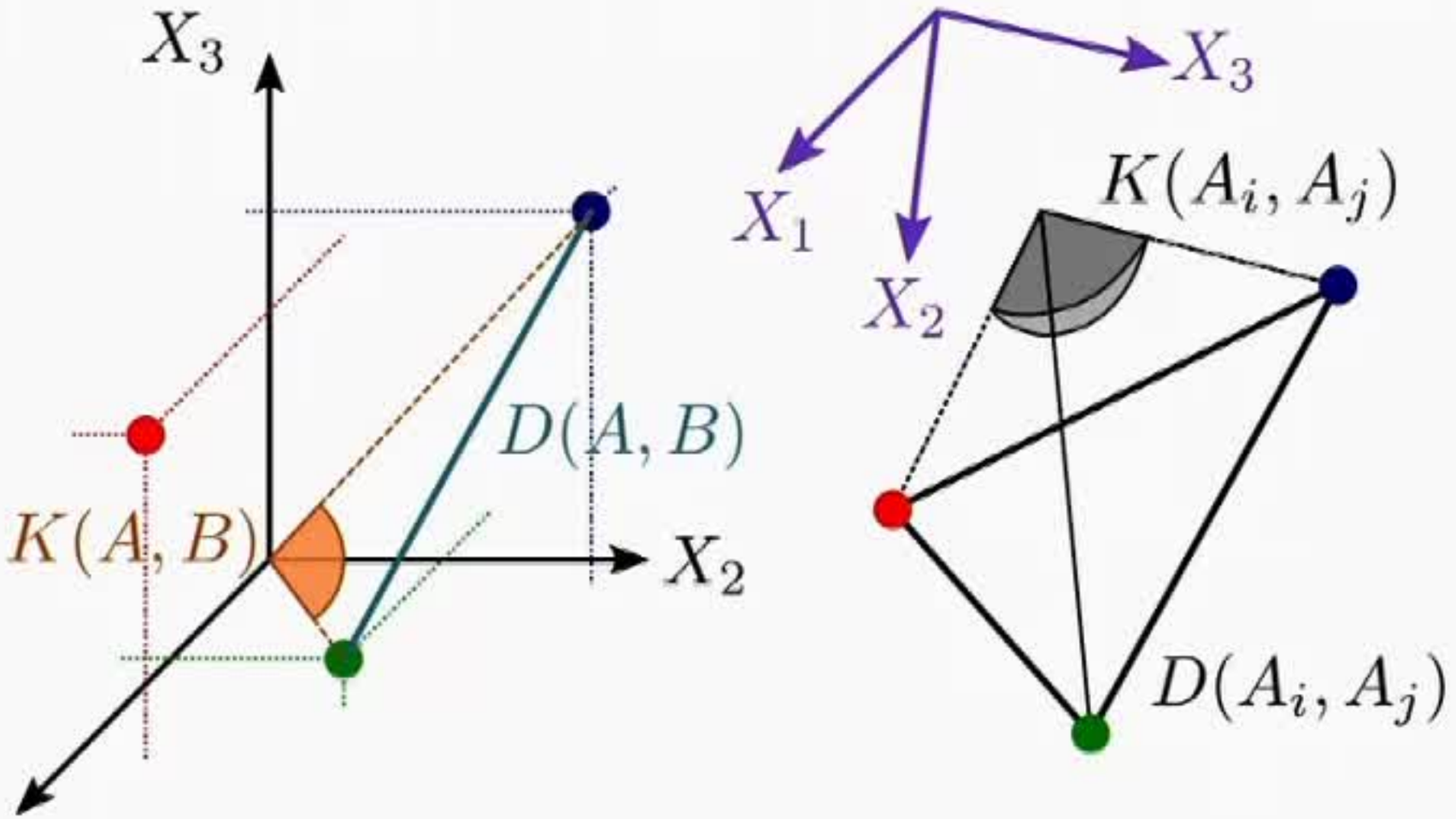
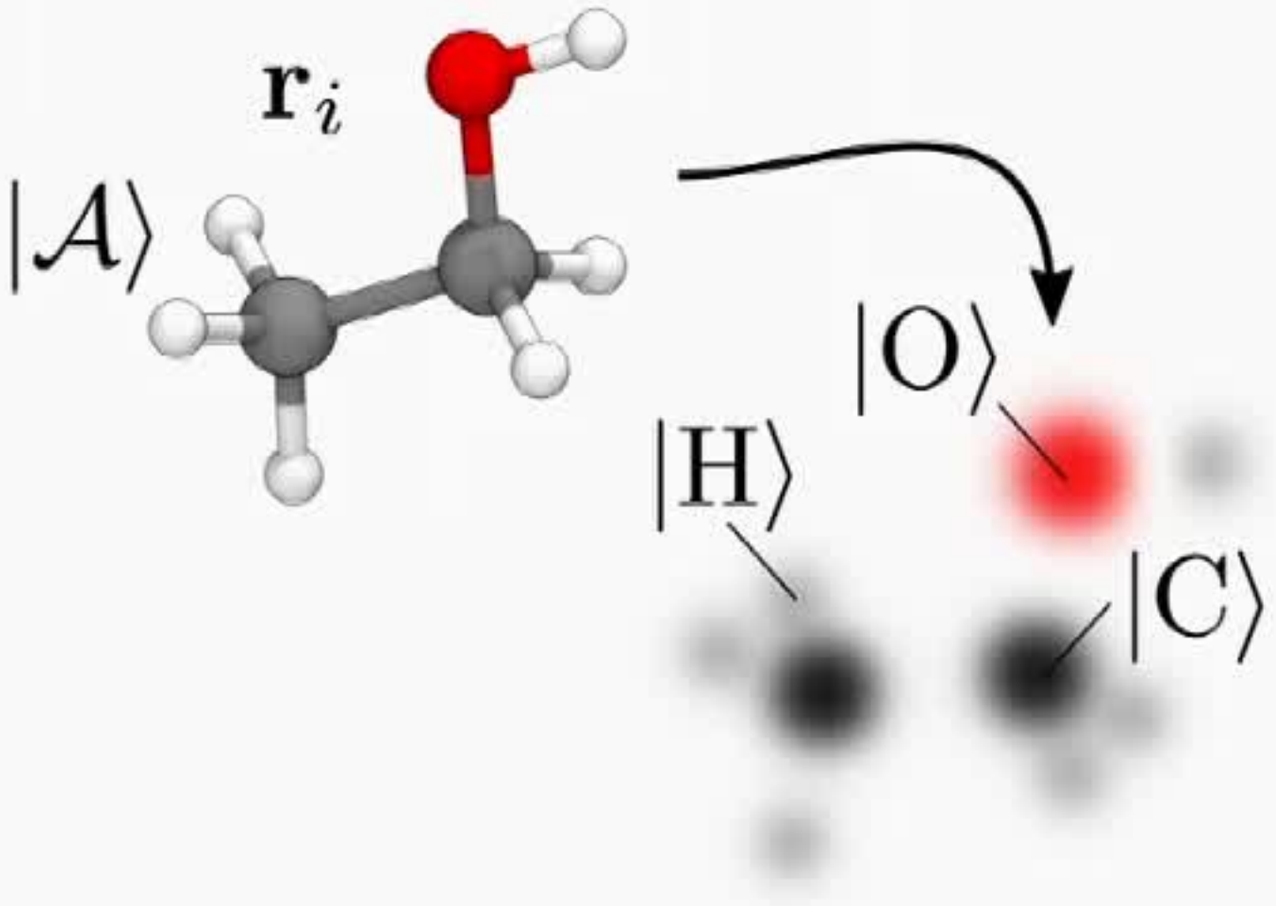


A Symmetry-Adapted Atom-Density Representation

- Structural representation based on a decorated atom-density vector $|\mathcal{A}\rangle$
- Physical symmetries are recovered by integration over group
- Use tensor products to reduce information loss
- $|\mathcal{A}^{(\nu)}\rangle_{\bar{T}}$ leads naturally to atom-centered decomposition
- Rotational average yields $(\nu+1)$-body correlation functions $|\mathcal{X}^{(\nu)}\rangle_{\bar{R}}$



$$\langle \mathbf{r}|\mathcal{A}\rangle = \sum_i g(\mathbf{r}-\mathbf{r}_i)\,|\alpha_i\rangle$$

Willatt, Musil, **MC**, https://arxiv.org/pdf/1807.00408

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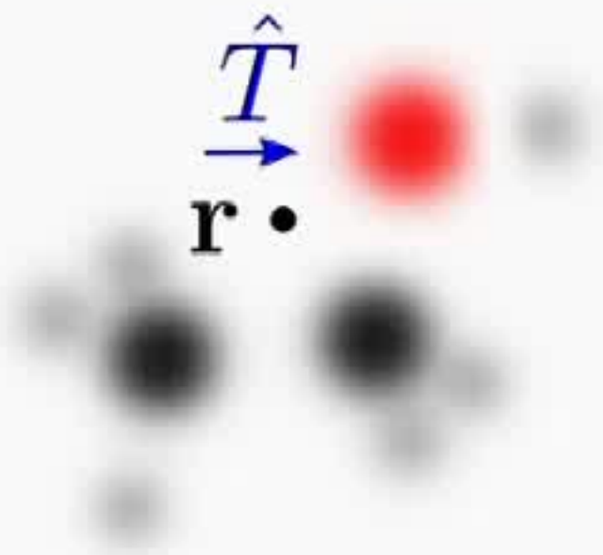

$$\int \mathrm{d}\hat{T} \left\langle \mathbf{r} \middle| \hat{T} \middle| \mathcal{A} \right\rangle = \sum_{\alpha} N_{\alpha} \, |\alpha\rangle$$

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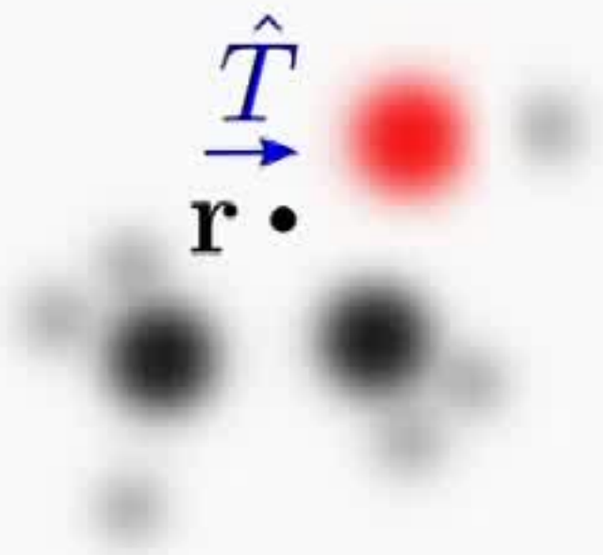

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$$\int \mathrm{d}\hat{T}\,\hat{T}\,|\mathcal{A}\rangle \otimes \hat{T}\,|\mathcal{A}\rangle = \int \mathrm{d}\mathbf{r}'\,\langle \mathbf{r}'|\mathcal{A}\rangle\,\langle \mathbf{r}'+\mathbf{r}|\mathcal{A}\rangle$$

Willatt, Musil, **MC**, https://arxiv.org/pdf/1807.00408

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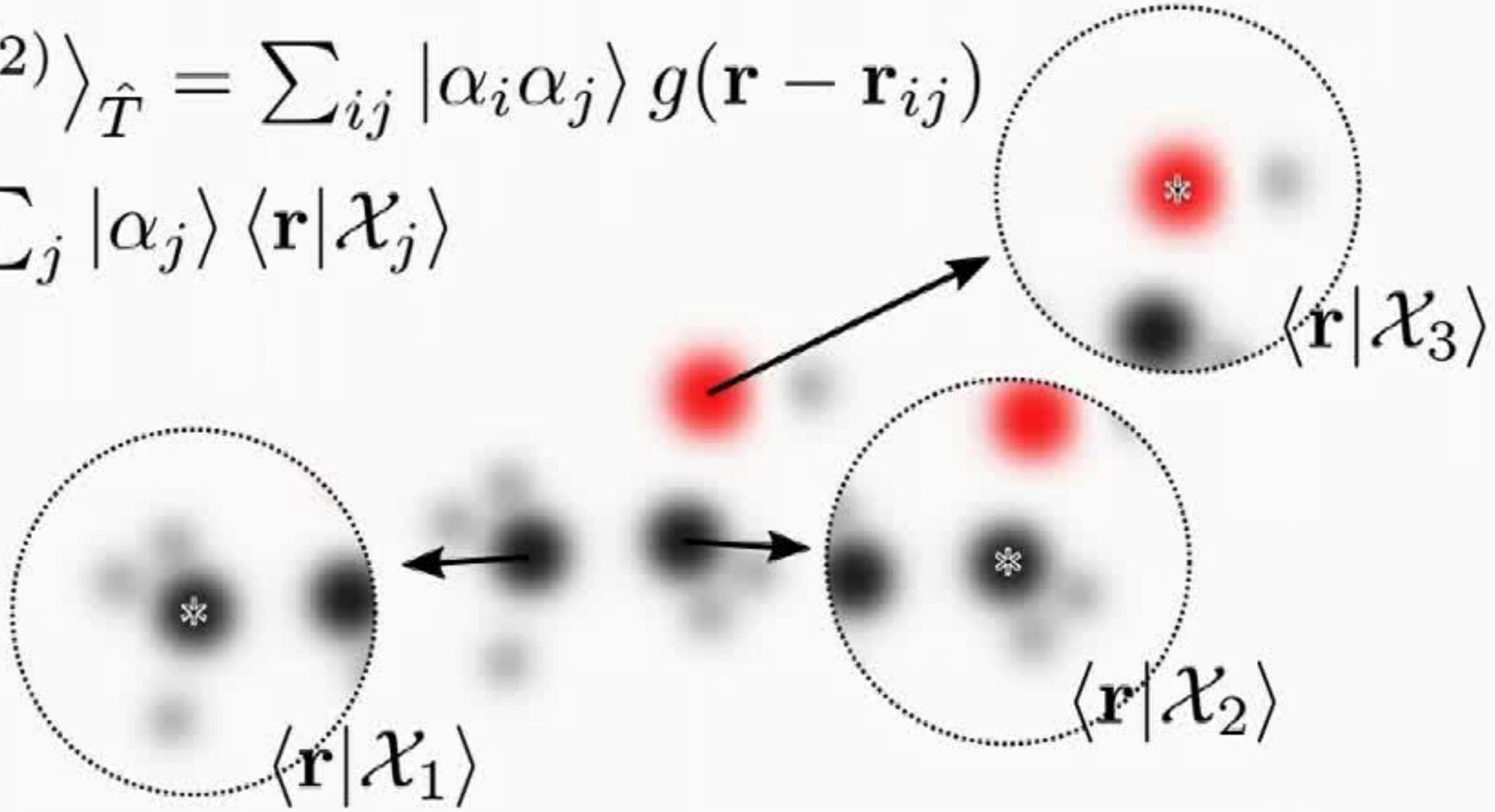
$$\left\langle \mathbf{r}\middle|\mathcal{A}^{(2)}\right\rangle_{\hat{T}} = \sum_{ij} |\alpha_i\alpha_j\rangle\, g(\mathbf{r}-\mathbf{r}_{ij})$$

$$= \sum_j |\alpha_j\rangle \langle \mathbf{r}|\mathcal{X}_j\rangle$$



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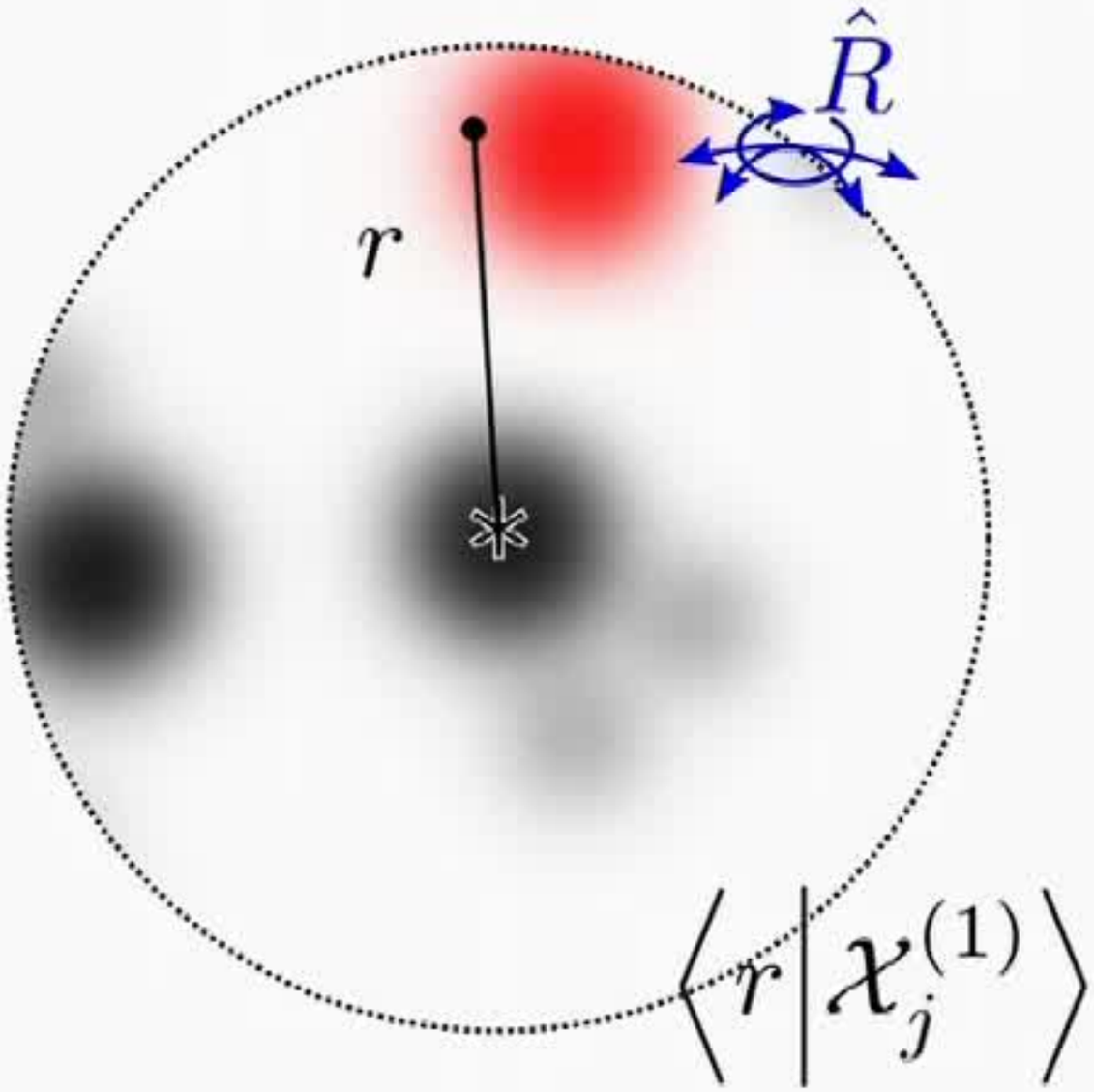

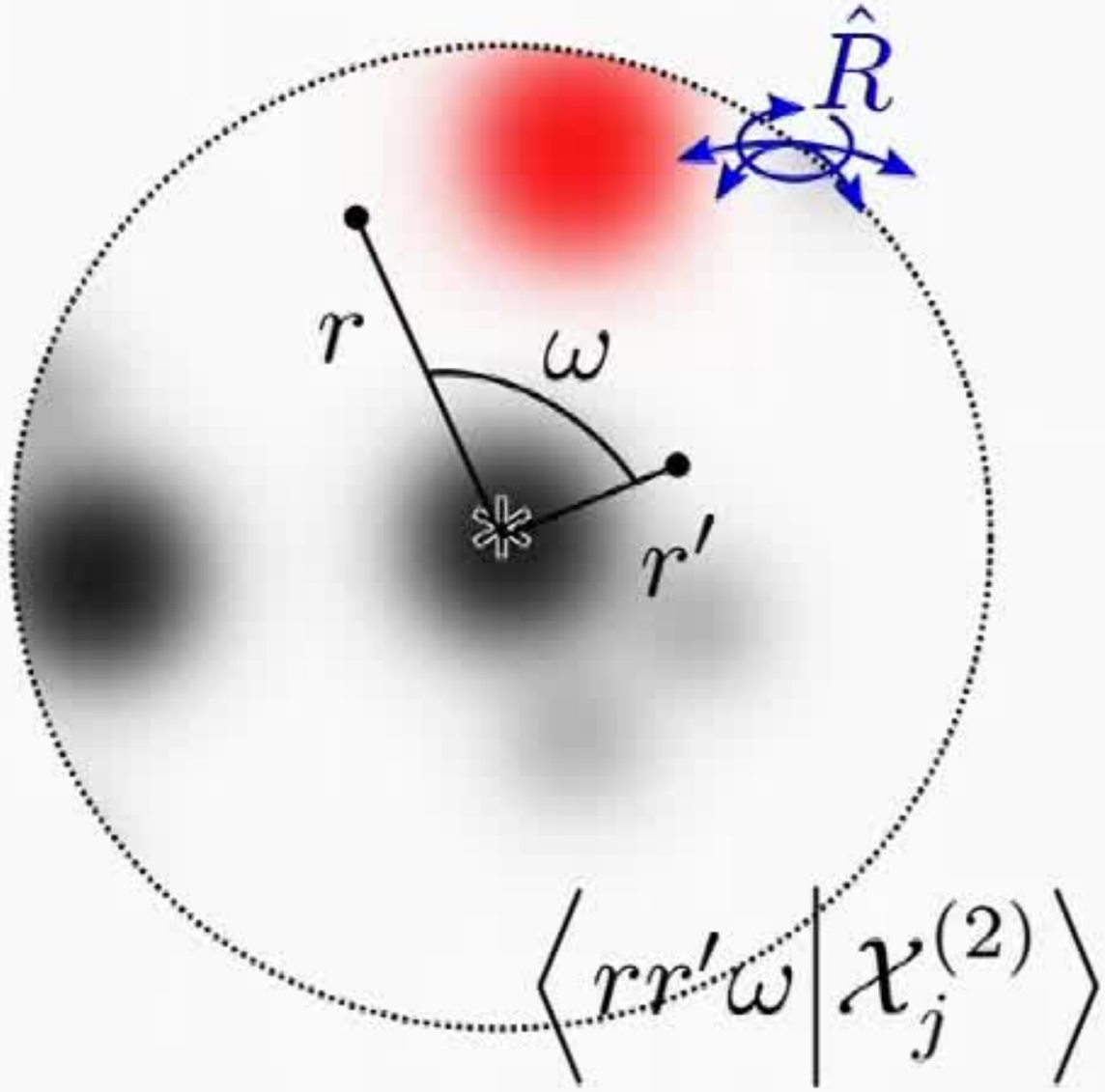

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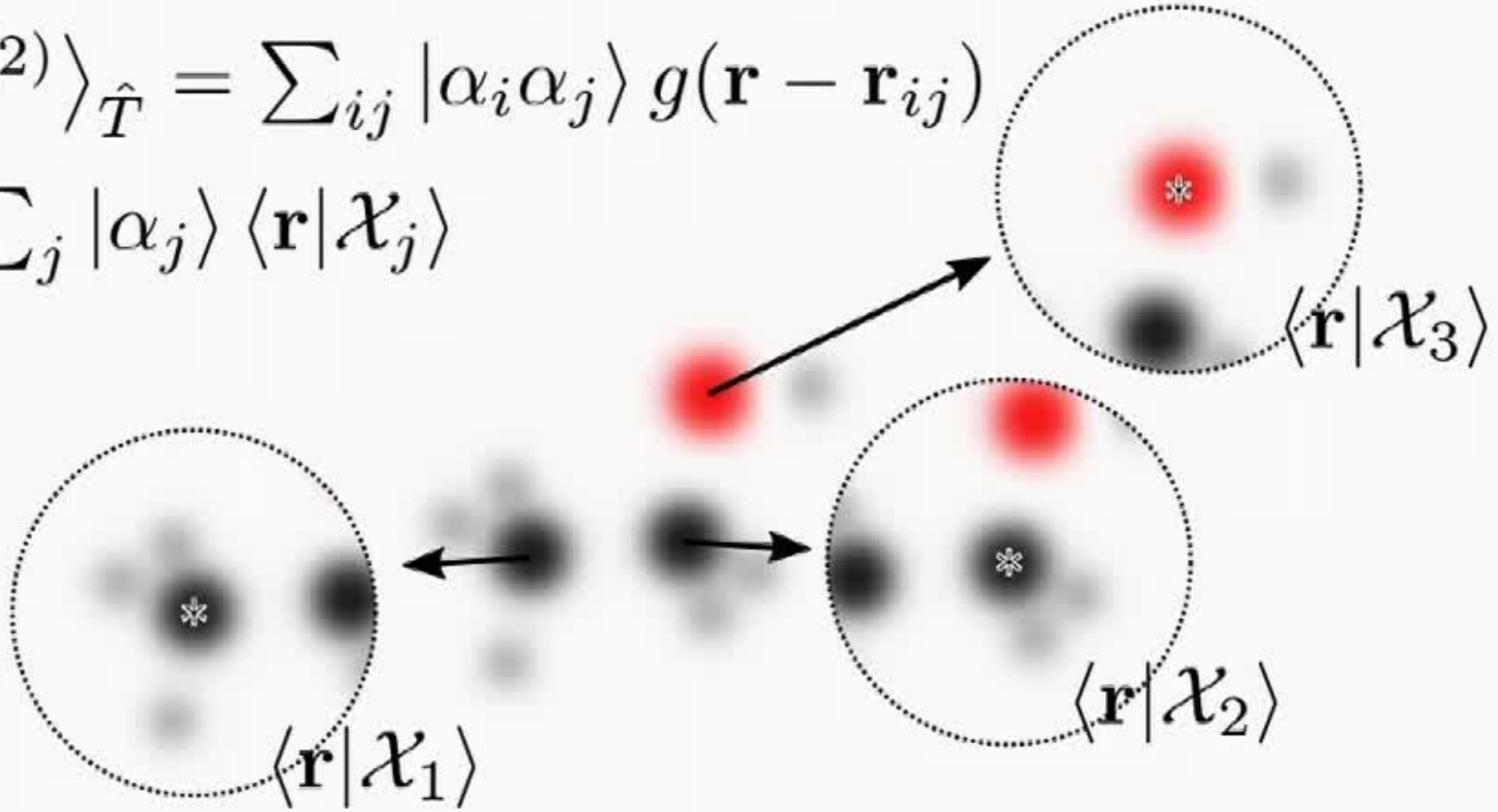

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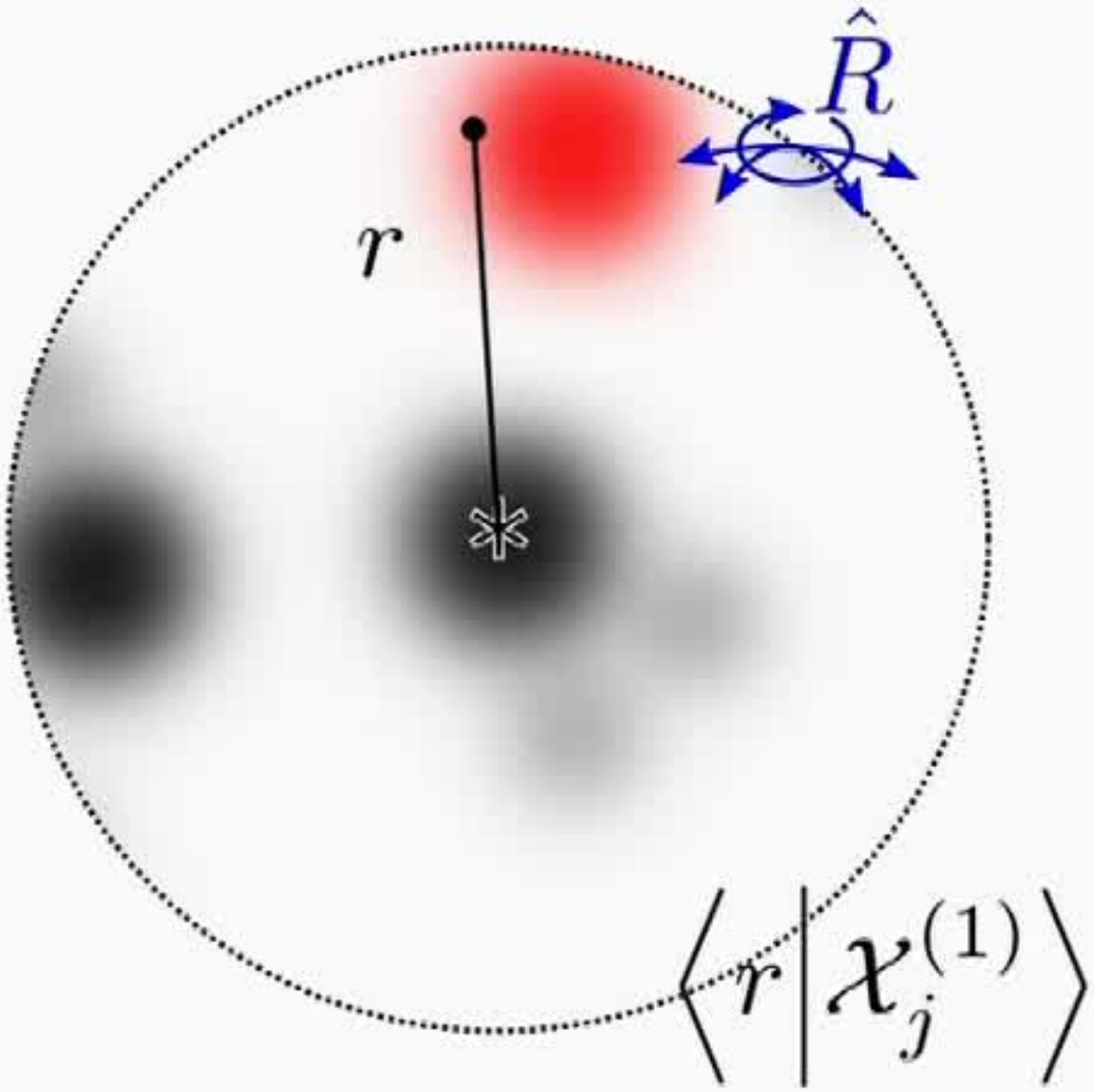
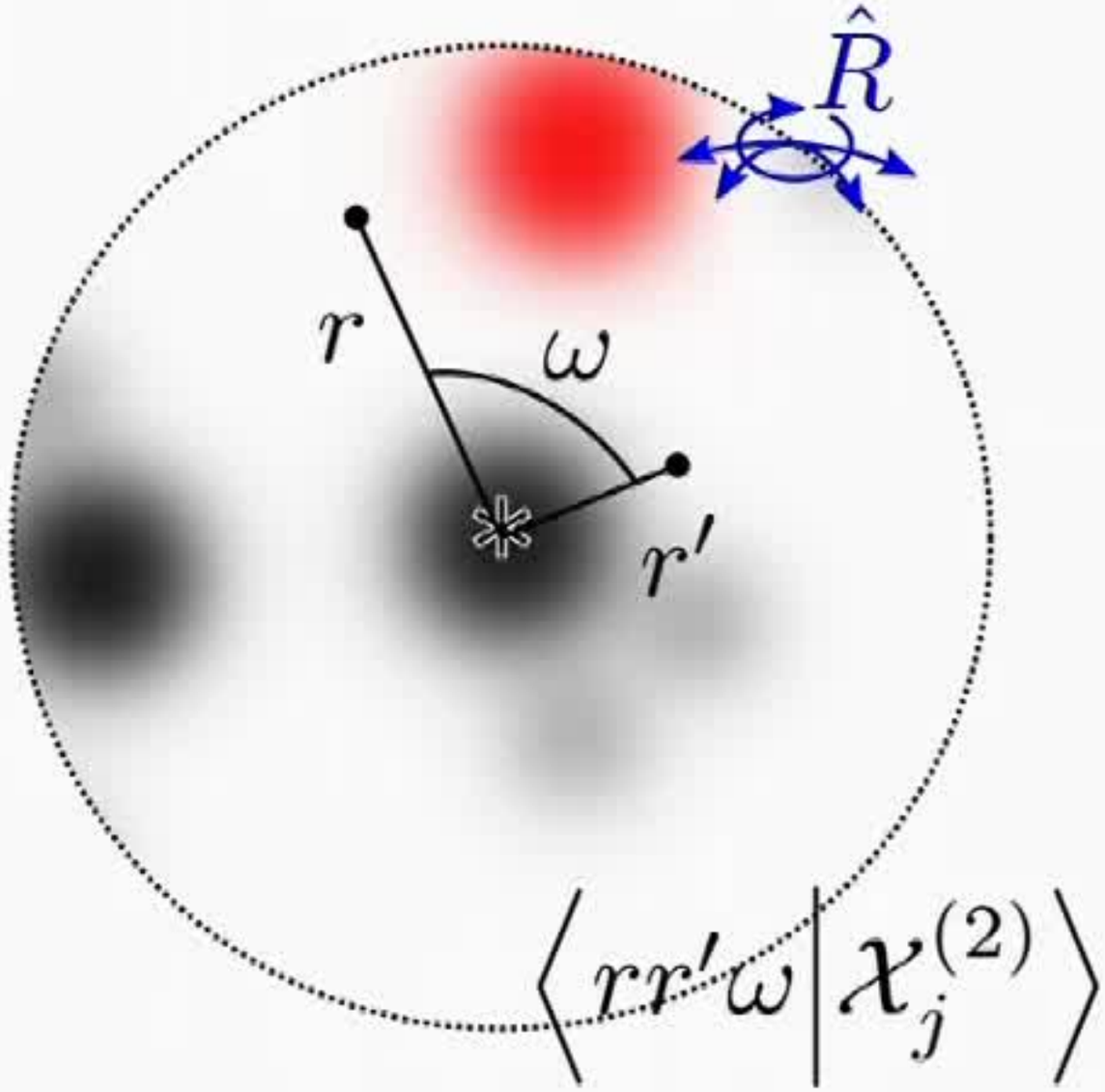
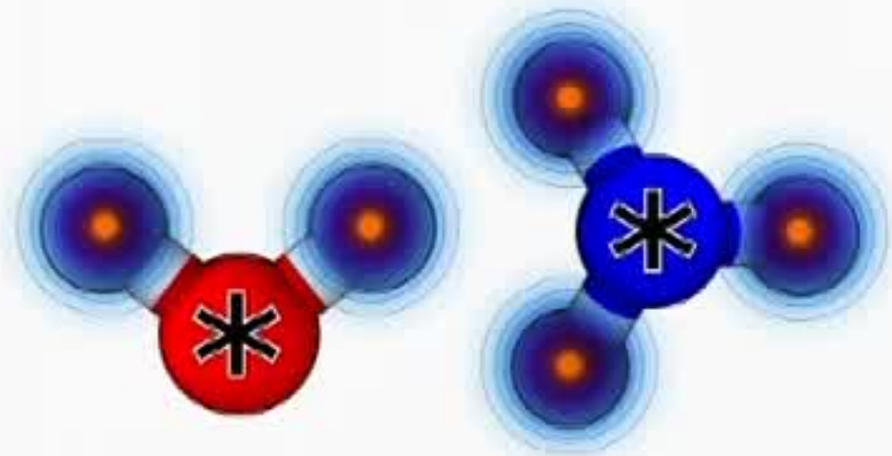


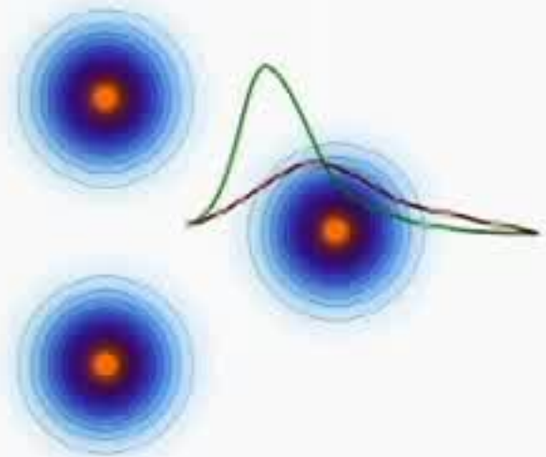


Symmetry Adapted Representations & SOAP Kernel

- Most of the existing density-based representations and kernels emerge as special cases of this framework
- Not necessary to use position basis. Radial functions and spherical harmonics $\rightarrow$ SOAP power spectrum and kernel
- Other strategies to combine local kernels (entropy-regularized match)



$$\langle \mathbf{r}|\mathcal{X}_j\rangle = \psi(\mathbf{r}) = \sum_i g(\mathbf{r}-\mathbf{r}_{ij})$$



$$\langle nlm|\mathcal{X}_j\rangle = \int \mathrm{d}\mathbf{x}\, \psi(\mathbf{r}) R_n(r) Y_m^l(\hat{\mathbf{r}})$$

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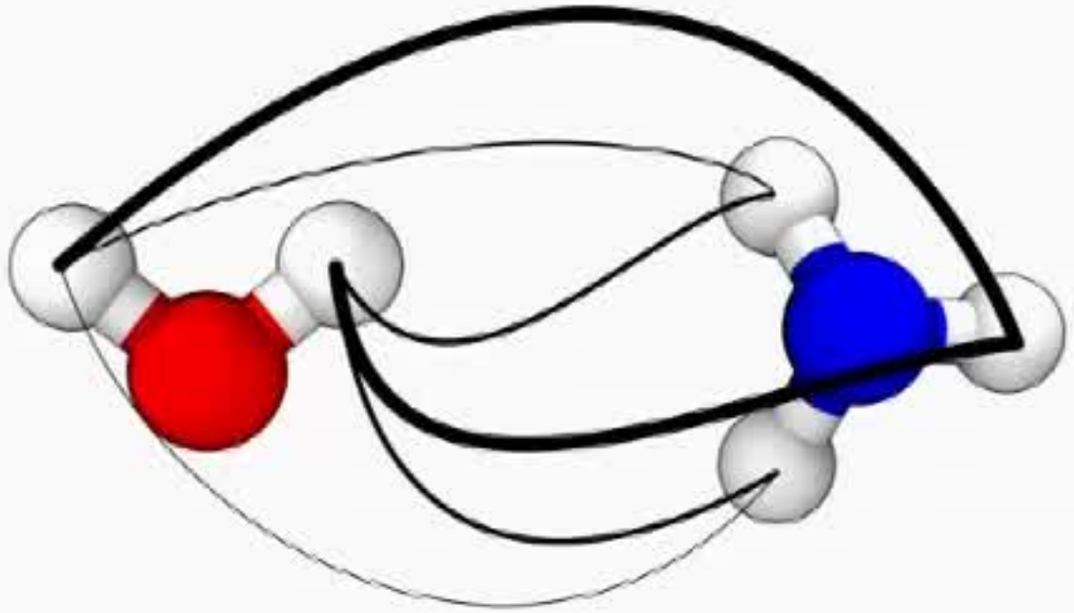
$$K(A,B) = \sum_{i\in A, j\in B} P_{ij} k(\mathcal{X}_i, \mathcal{X}_j)$$

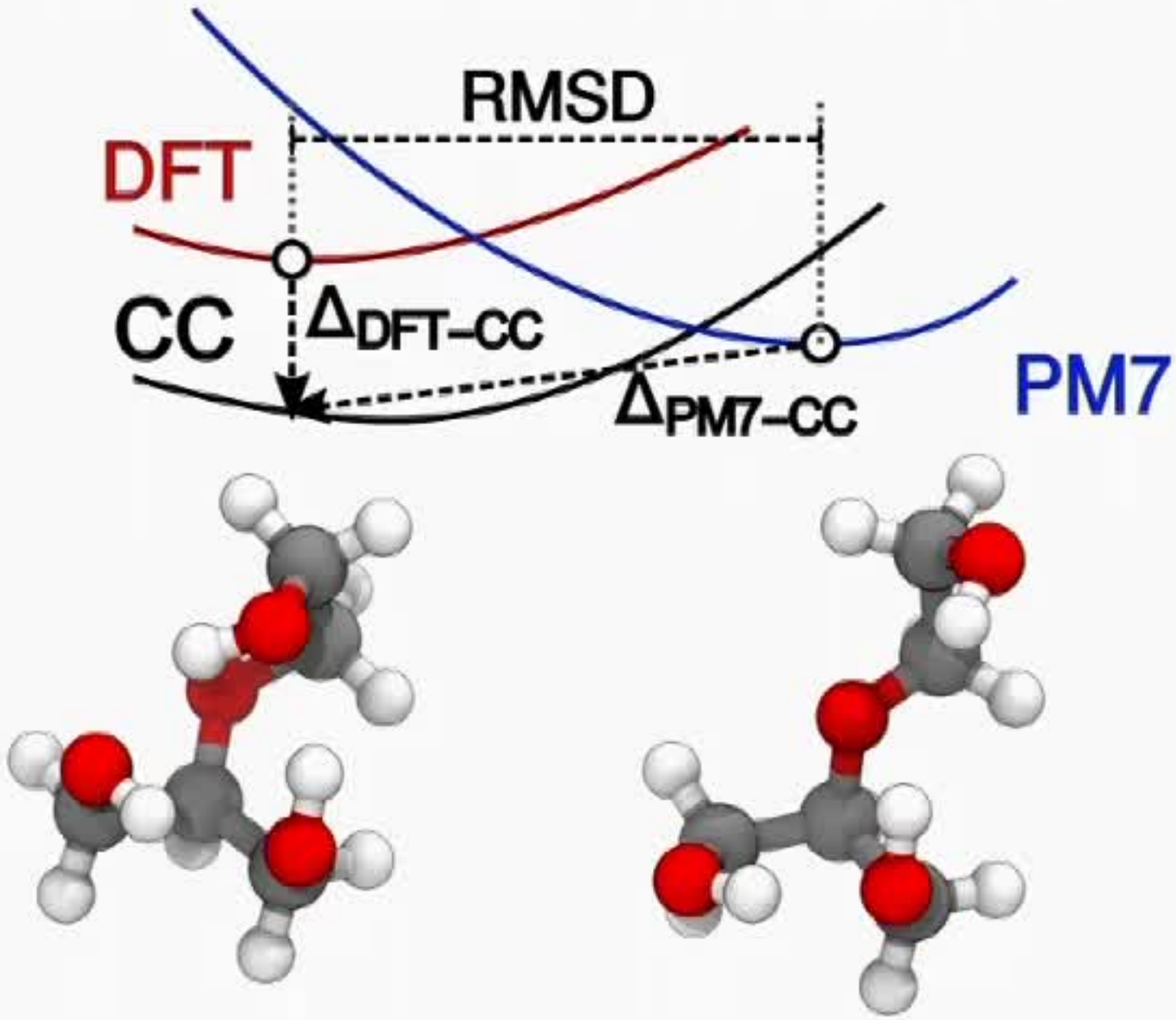


$$\hat{K}^{\gamma}(A,B) \propto$$
$$\max_{\mathbf{P}\in\mathcal{U}} \sum_{ij} P_{ji}(C_{ij}^{A,B} - \gamma \ln P_{ji})$$

De, Bartók, Csányi, **MC**, PCCP (2016); M. Cuturi, NIPS (2013);

100k Molecules with Coupled-Clusters

- CCSD(T) Energetics on the QM9 - 114k useful predictions based on 20k training calculations
- 1kcal/mol error for predicting CCSD(T) based on PM7 geometries; 0.18kcal/mol error for predicting CCSD(T) based on DFT geometries!



Ramakrishnan et al., Scientific Data (2014); Ramakrishnan et al., JCTC (2015)

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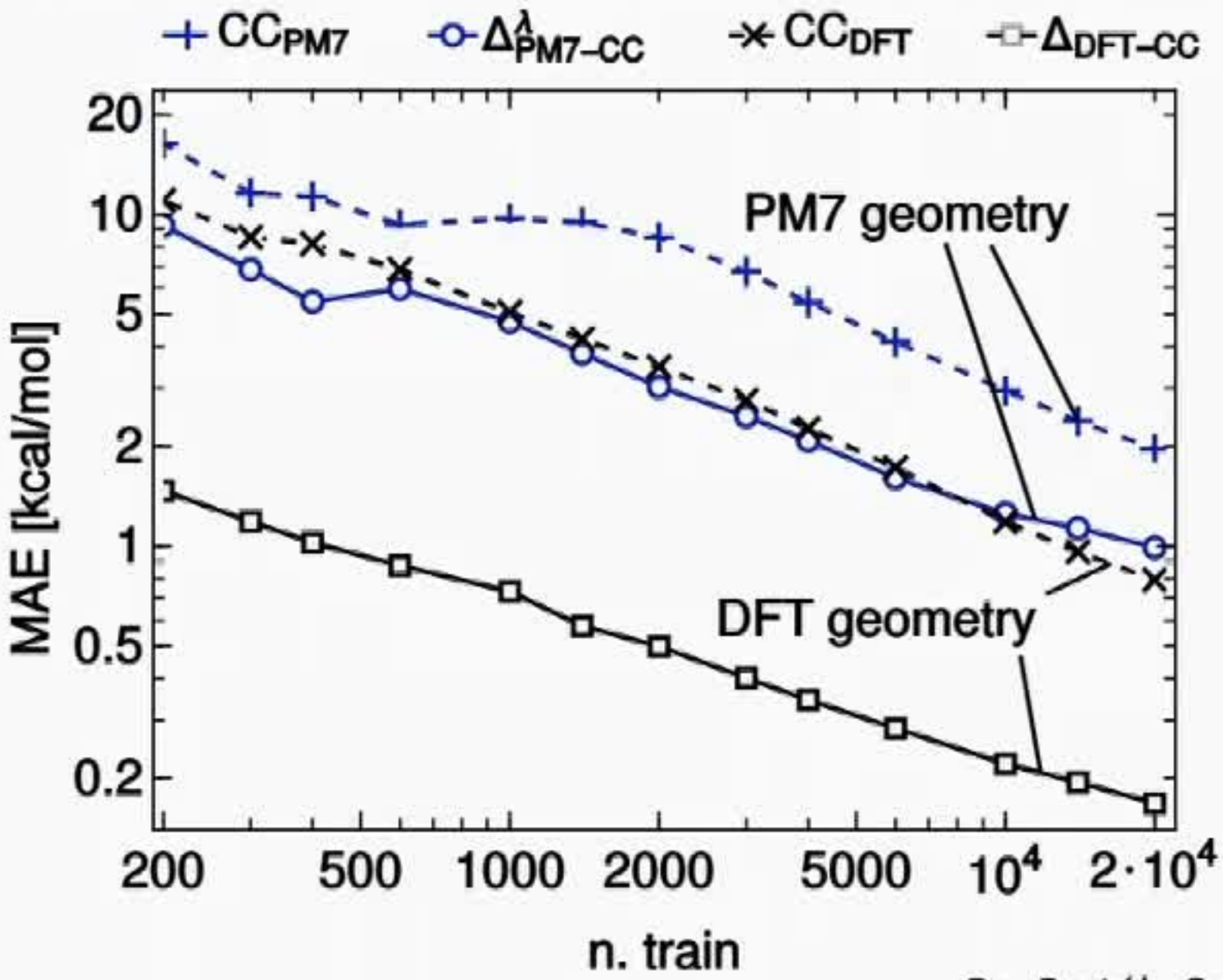
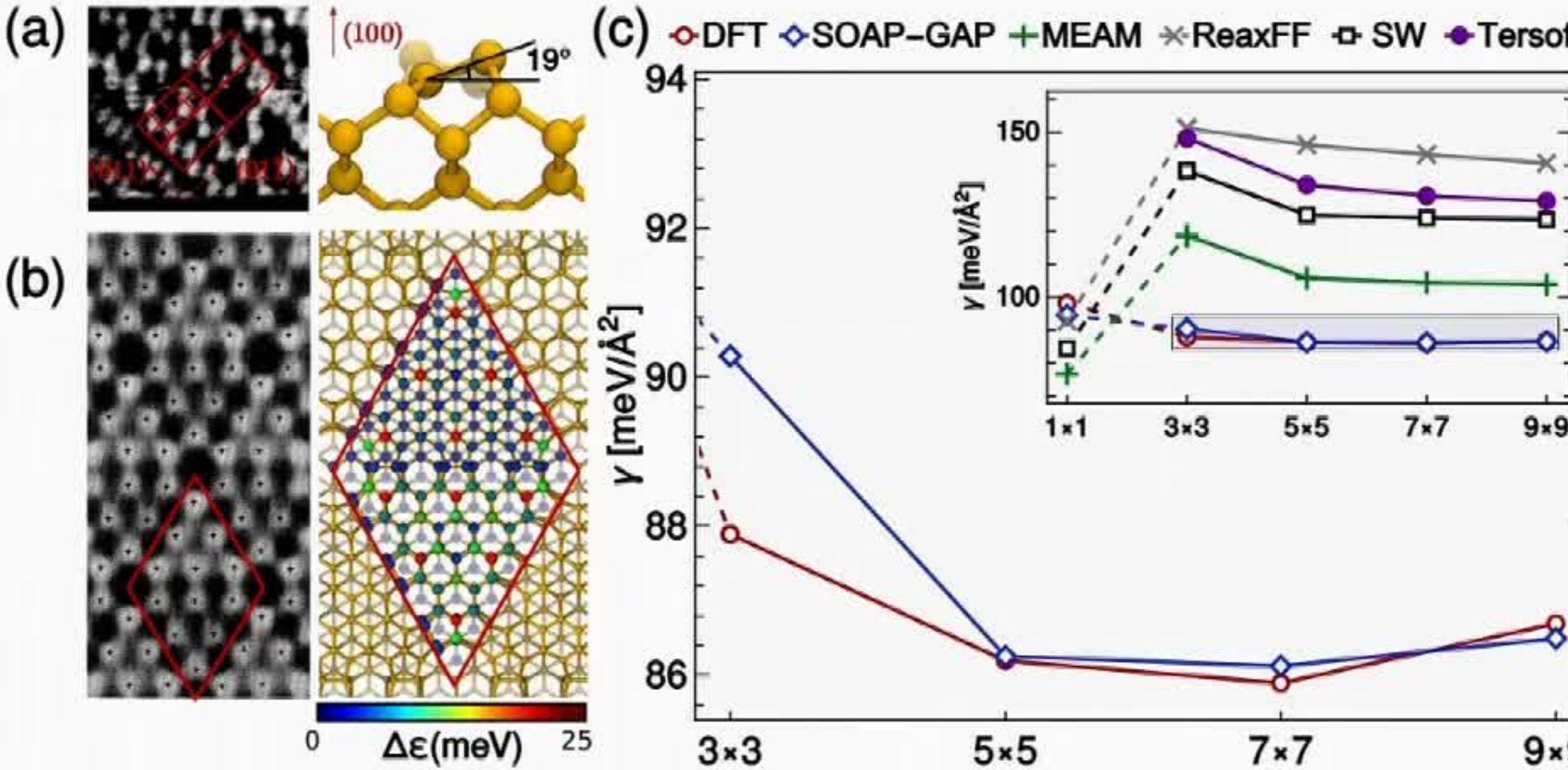


De, Bartók, Csányi, **MC**, PCCP (2016);
Bartok, De, Kermode, Bernstein, Csanyi, **MC**, Science Advances (2017)

Silicon Surfaces - Complexity in a Simple Material

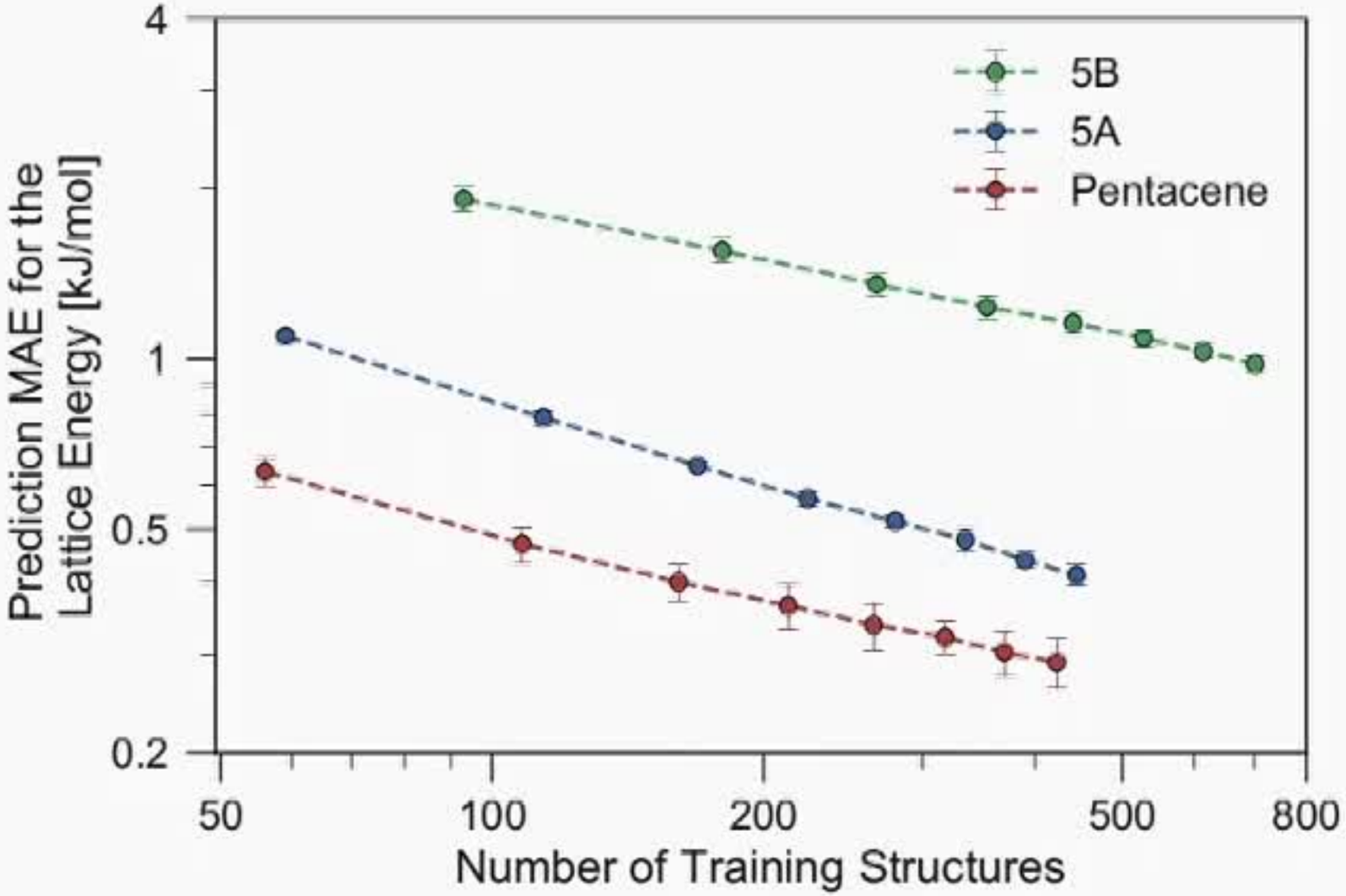
- More than just molecules: a SOAP-GAP model for Si can capture the dimer tilt in Si(100)-2x1, and the delicate energy balance that determines the stability of the Si(111) 7x7 DAS reconstruction

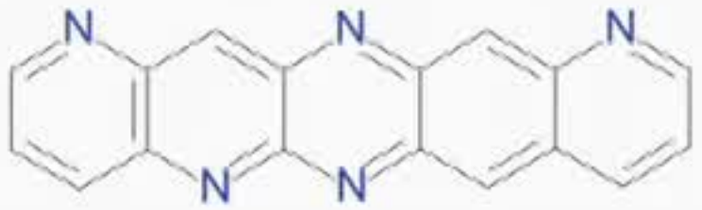


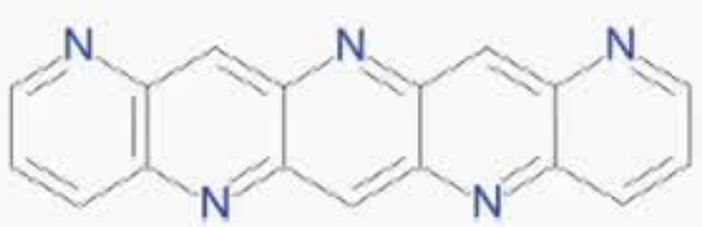
Bartok, De, Kermode, Bernstein, Csanyi, **MC**, Science Advances (2017)

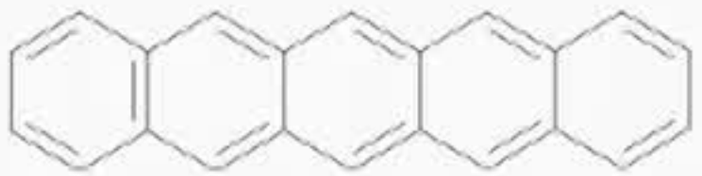
Accurate Predictions for Molecular Crystals

- Substituted pentacenes - model systems for molecular electronics
- Easily achieve sub-kcal/mol accuracy, with REMatch-SOAP kernels





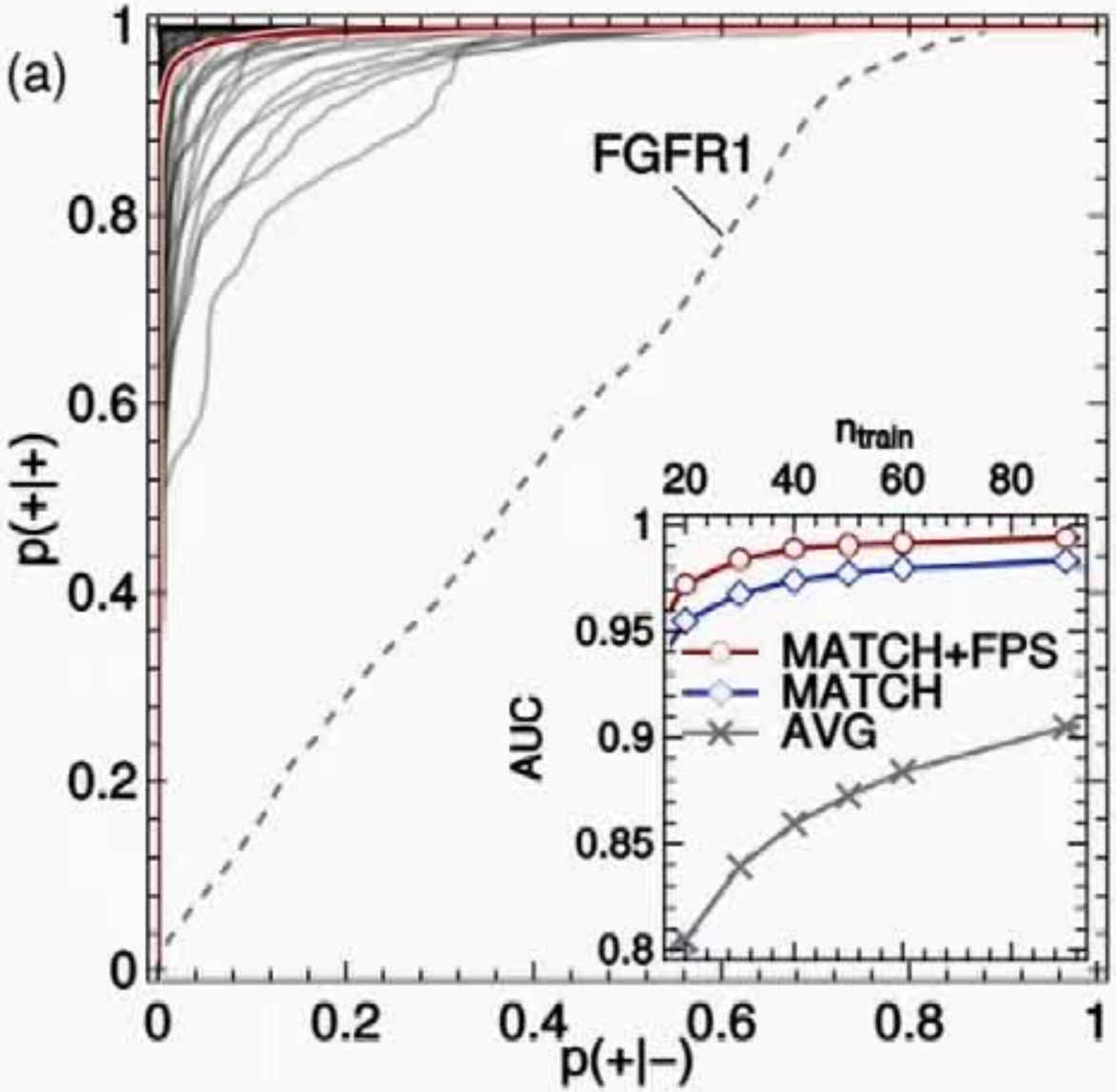


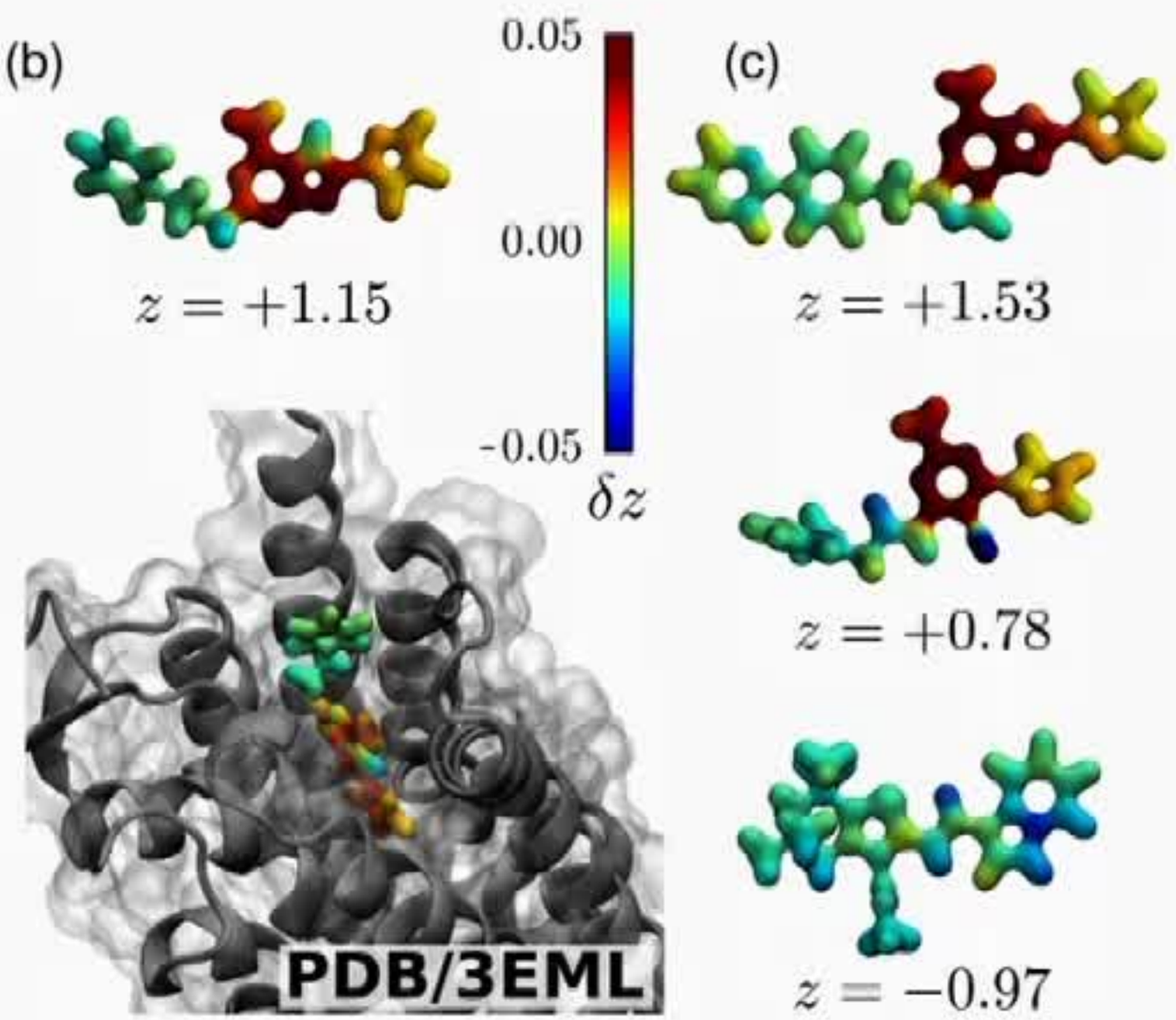


Musil, De, Yang, Campbell, Day, **MC**, Chemical Science (2018)

Recognizing Active Ligands for Receptor Proteins

- 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

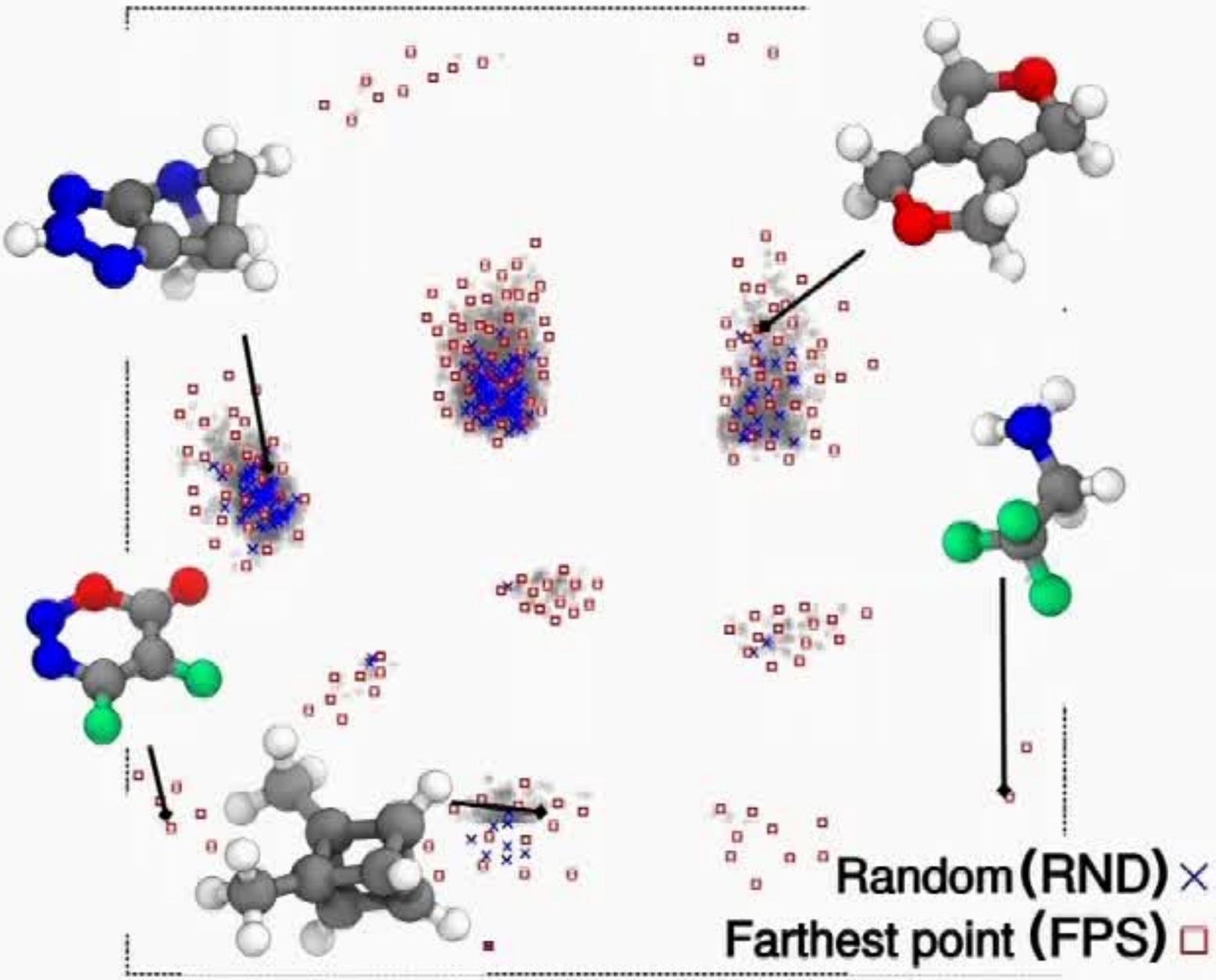




Bartok, De, Kermode, Bernstein, Csanyi, **MC**, Science Advances (2017)

Thorough Sampling of Compound Space

- The train set should cover uniformly the relevant space
  - FPS is a simple, constructive strategy to optimize the training set, opening doors to active learning



Bartok, De, Kermode, Bernstein, Csanyi, **MC**, Science Advances (2017)

MC, Tribello, Parrinello, PNAS (2011); http://sketchmap.org

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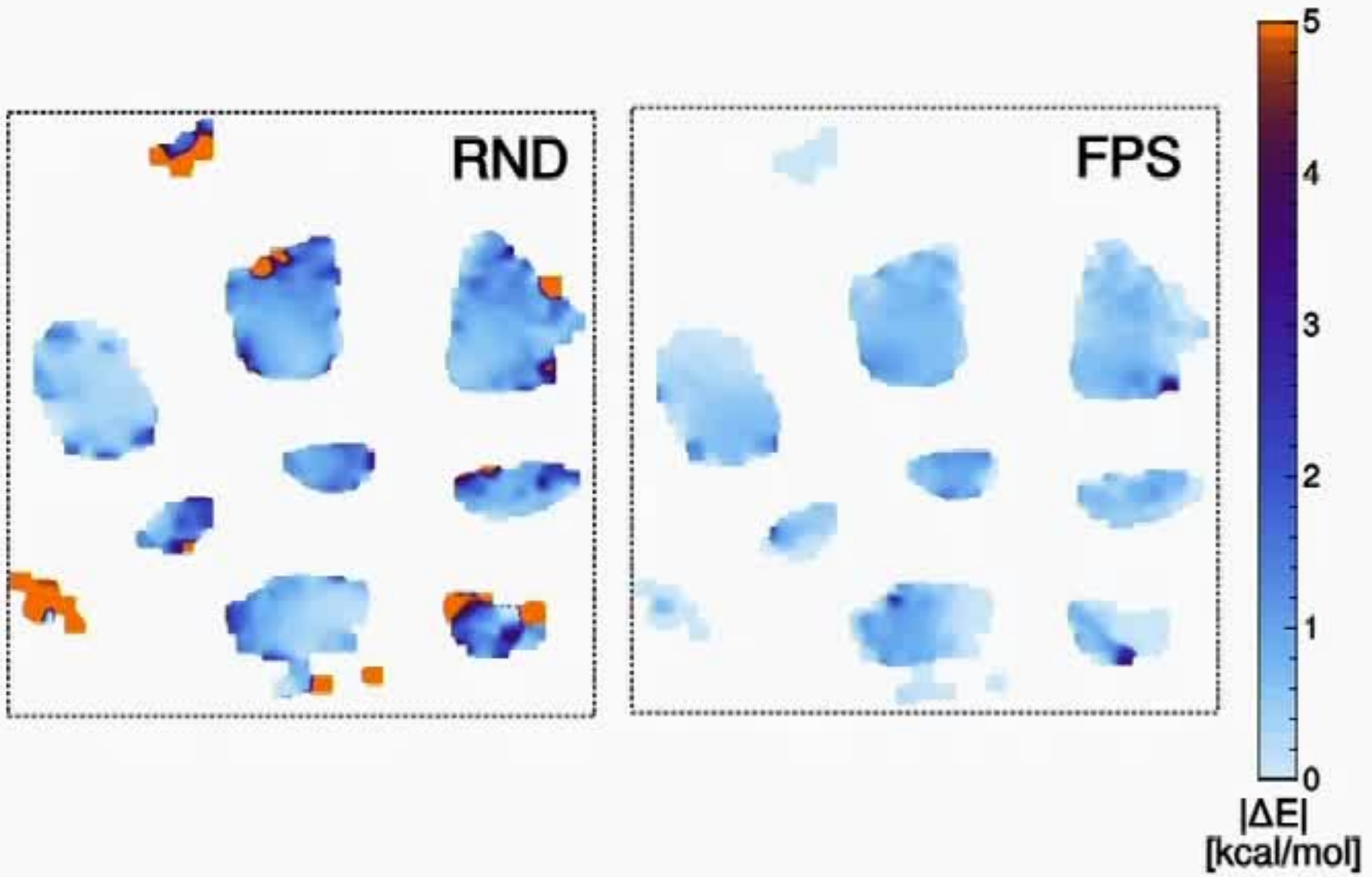

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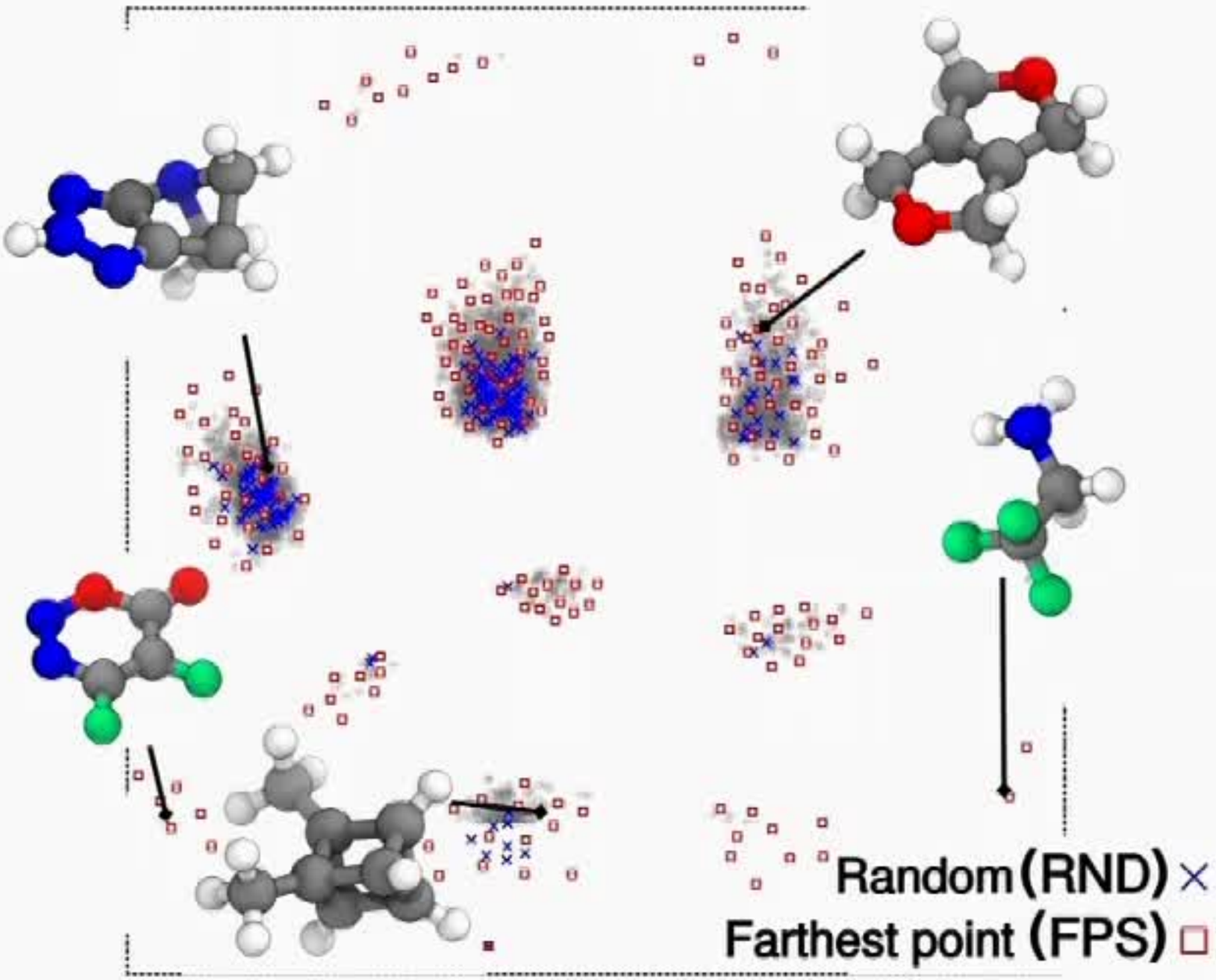

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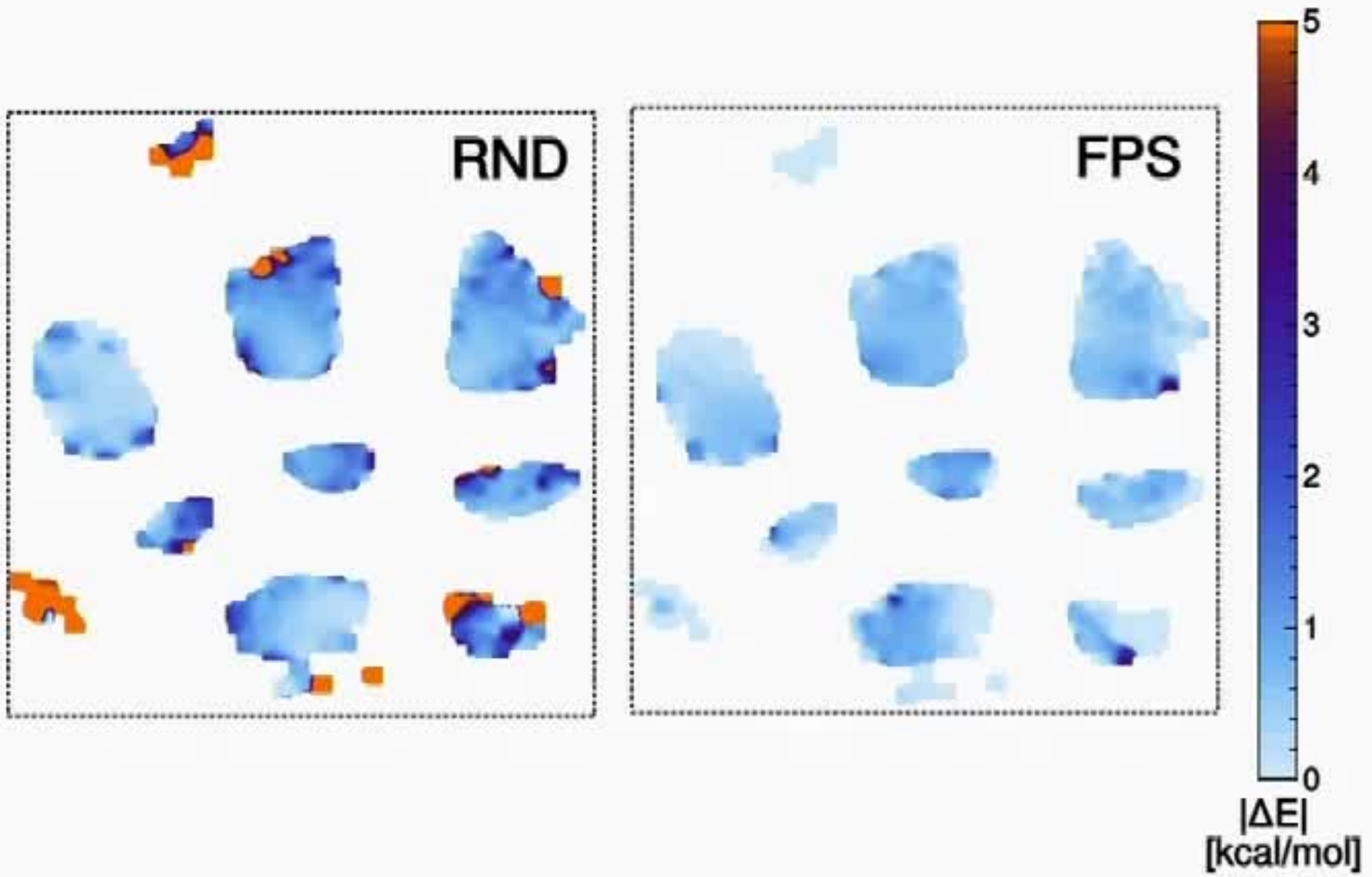

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More than Interatomic Potentials

- Solid-state NMR relies on GIPAW-DFT to determine crystal structure of molecular materials
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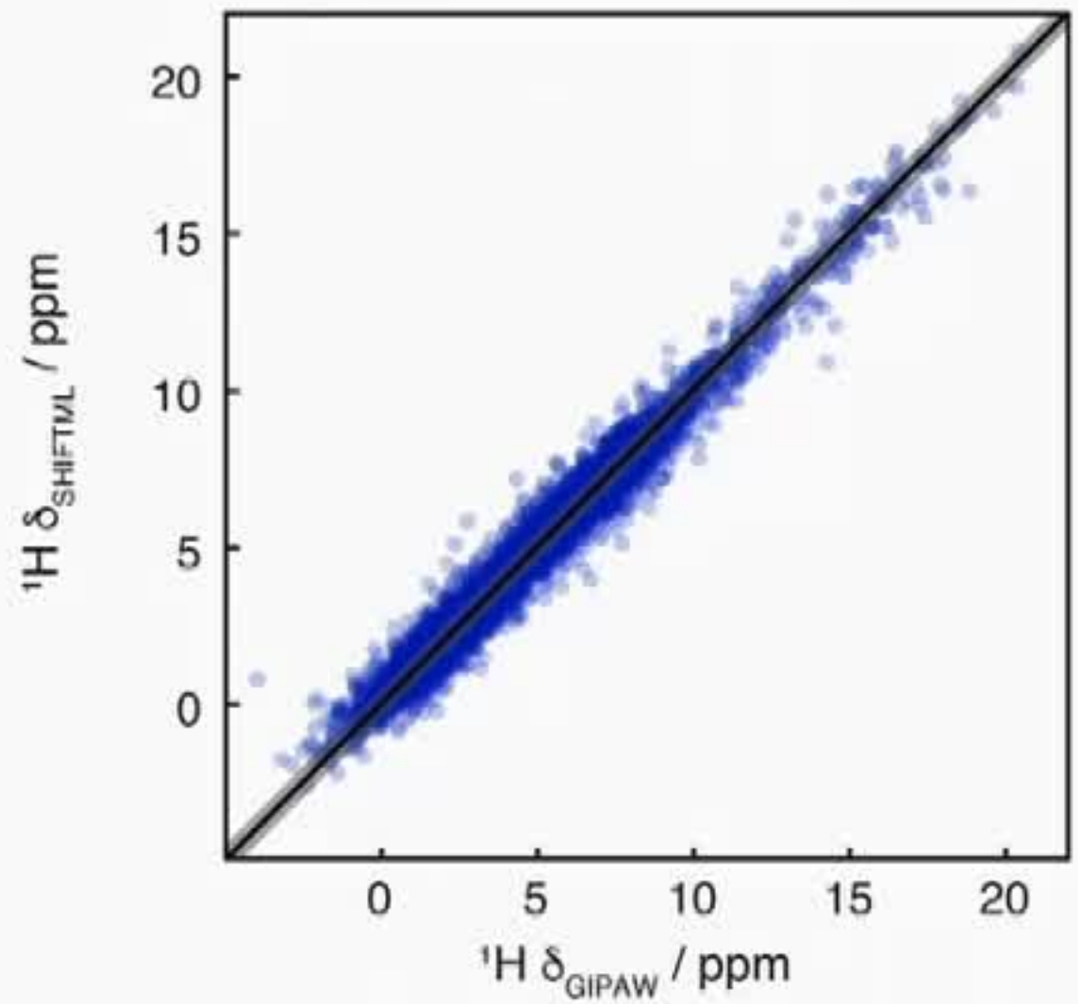

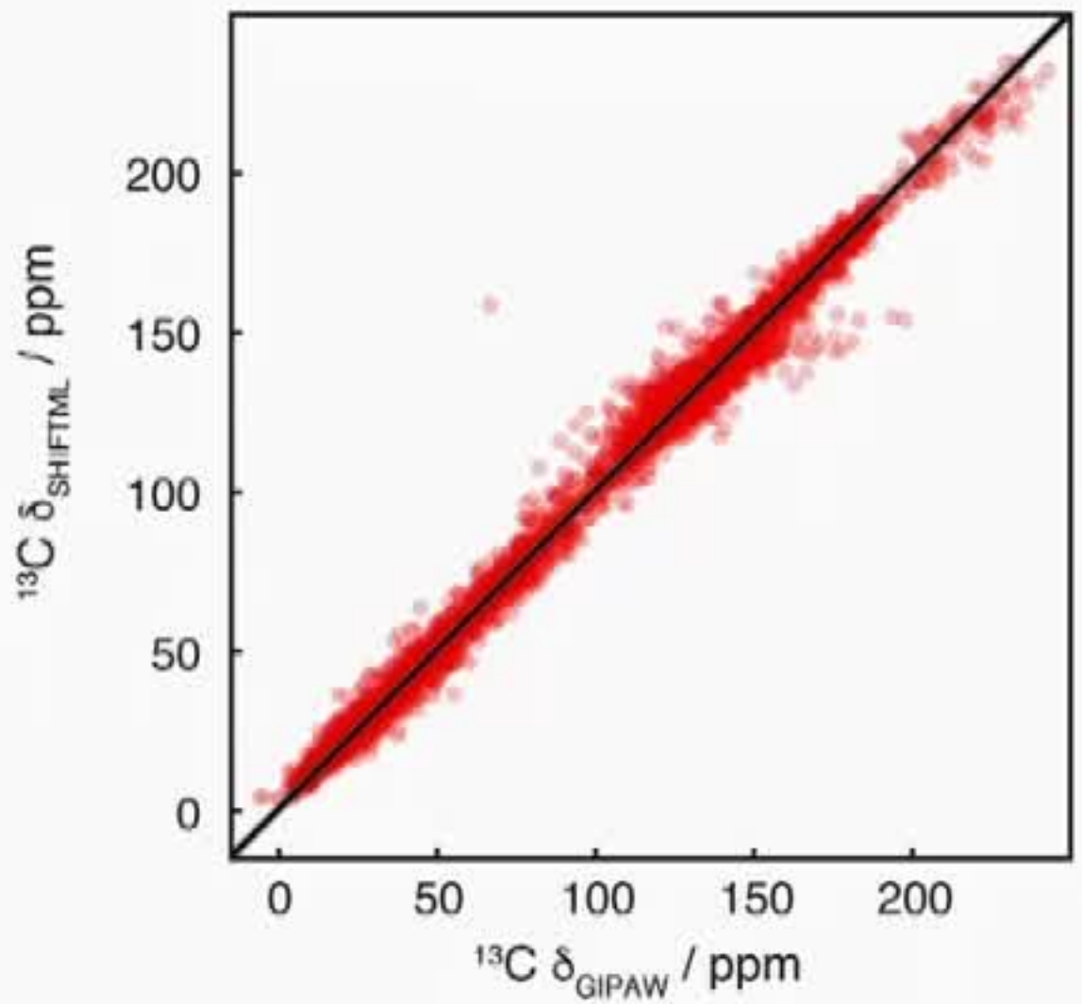

w/Emsley, Paruzzo, Hofstetter, http://shiftml.org

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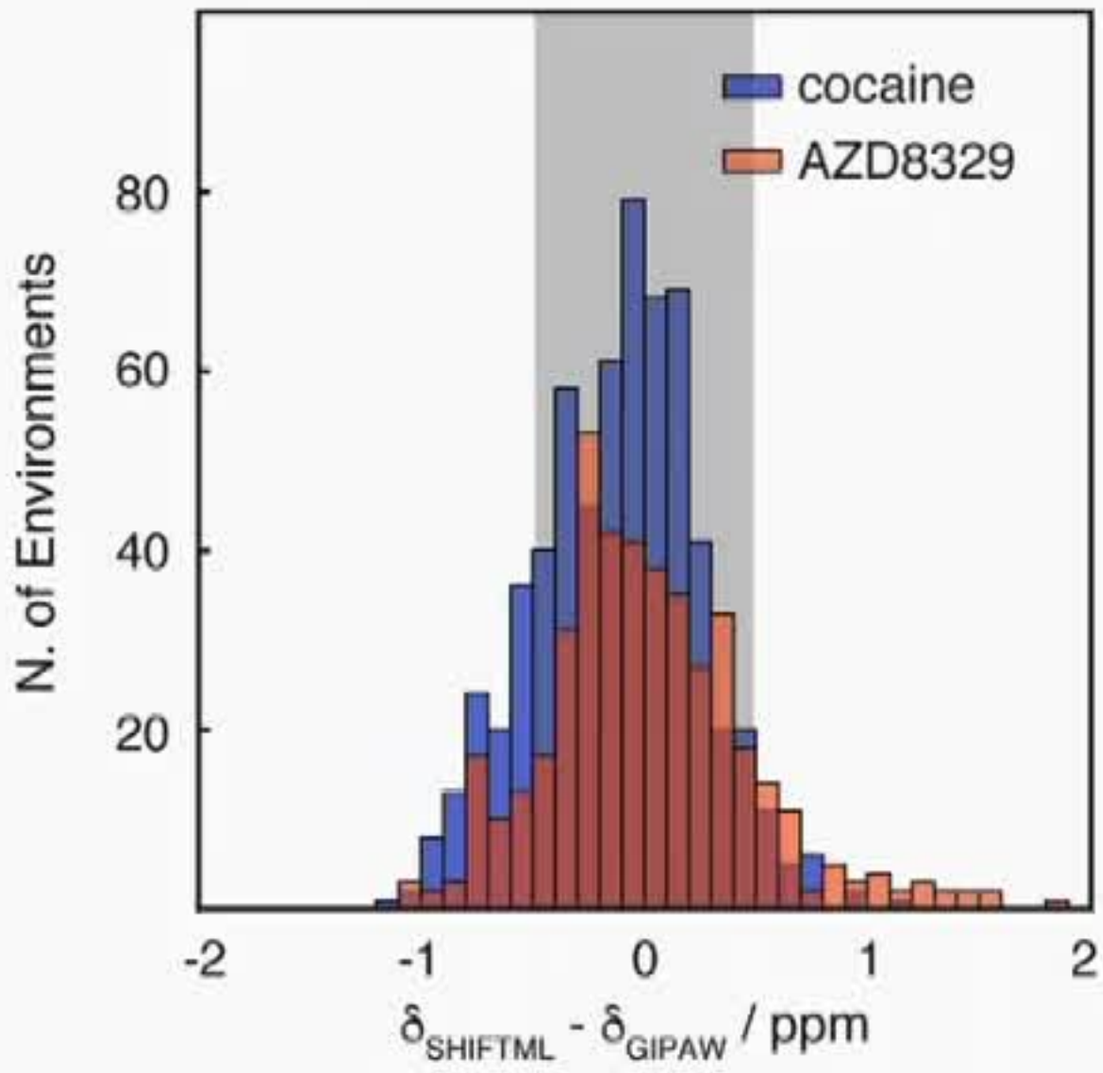

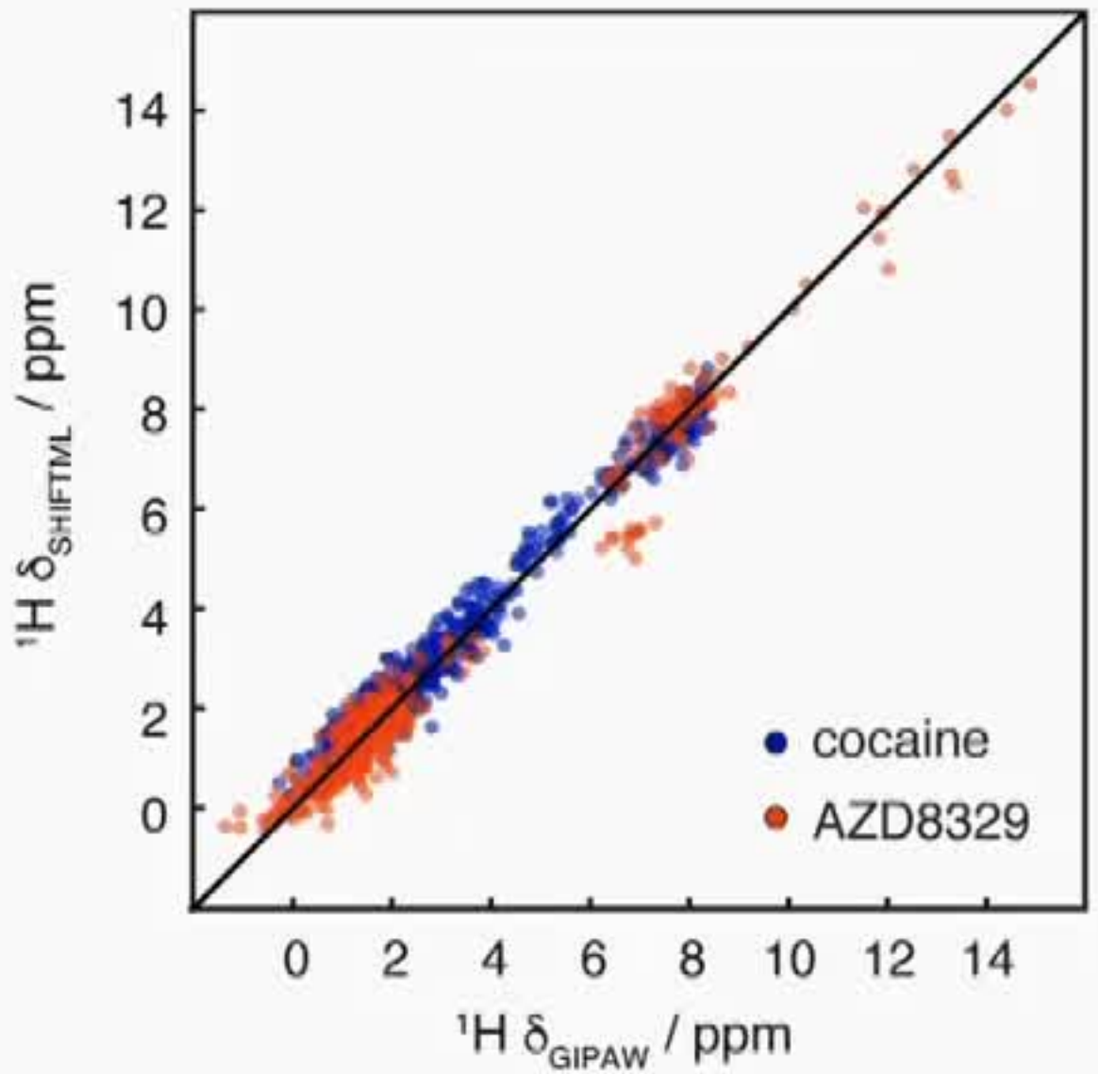

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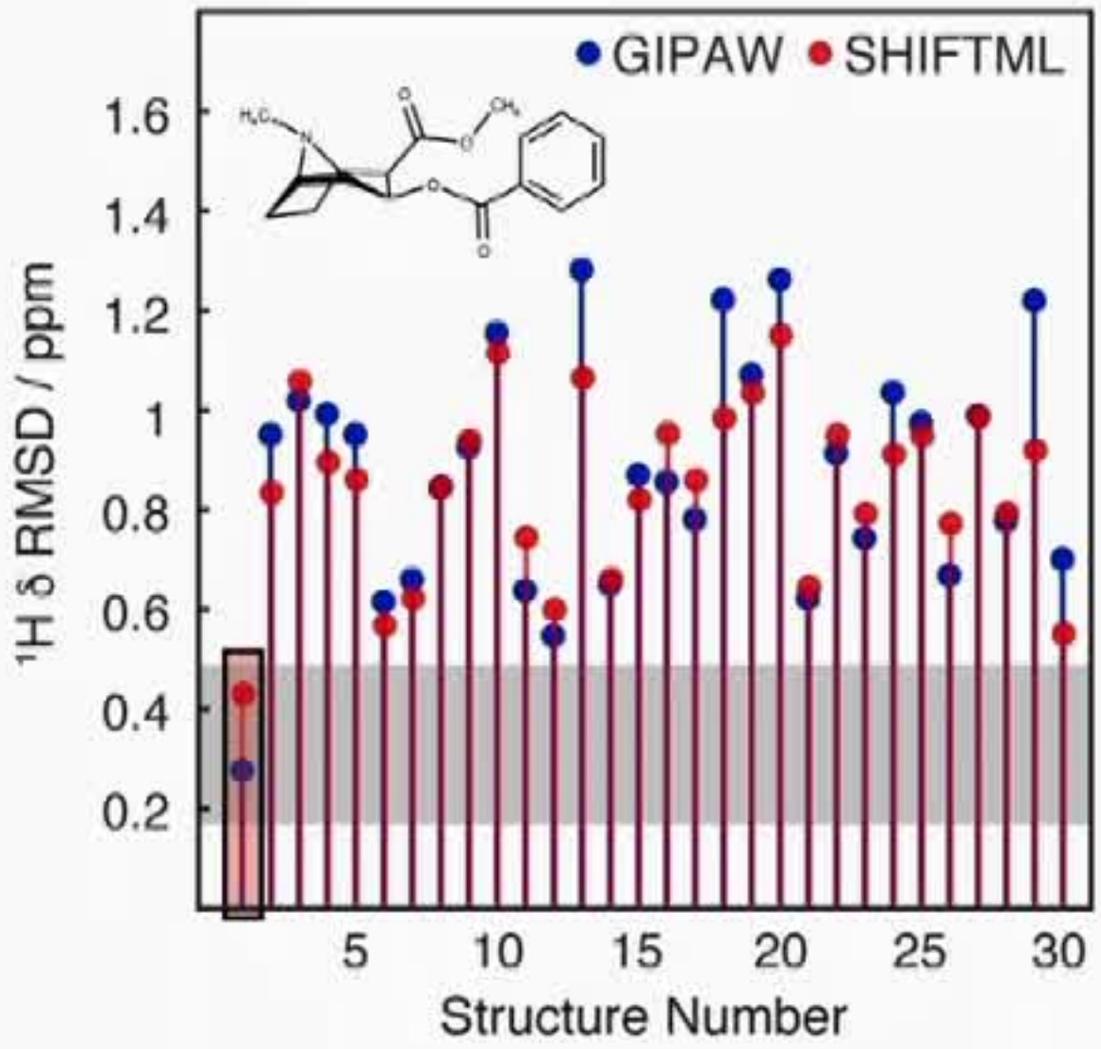
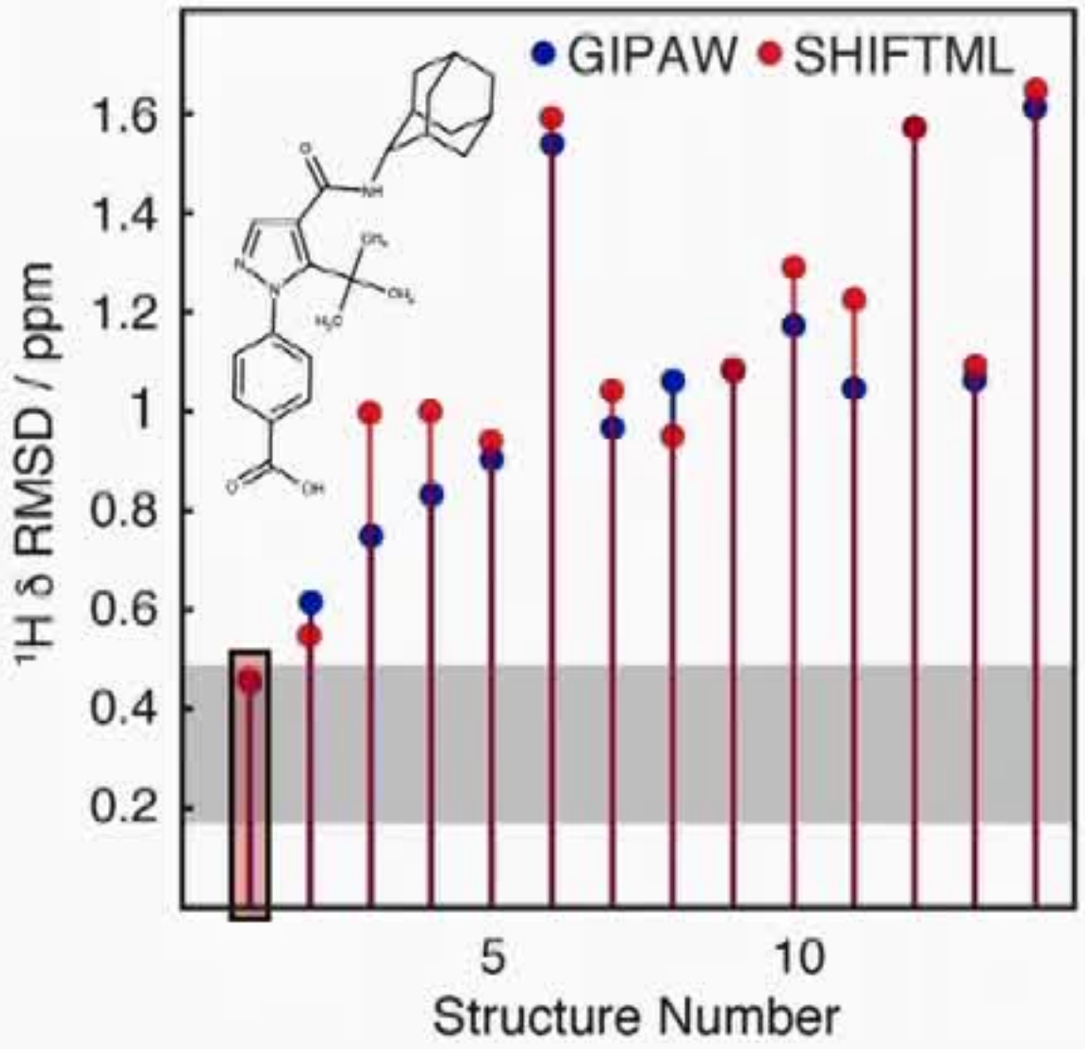
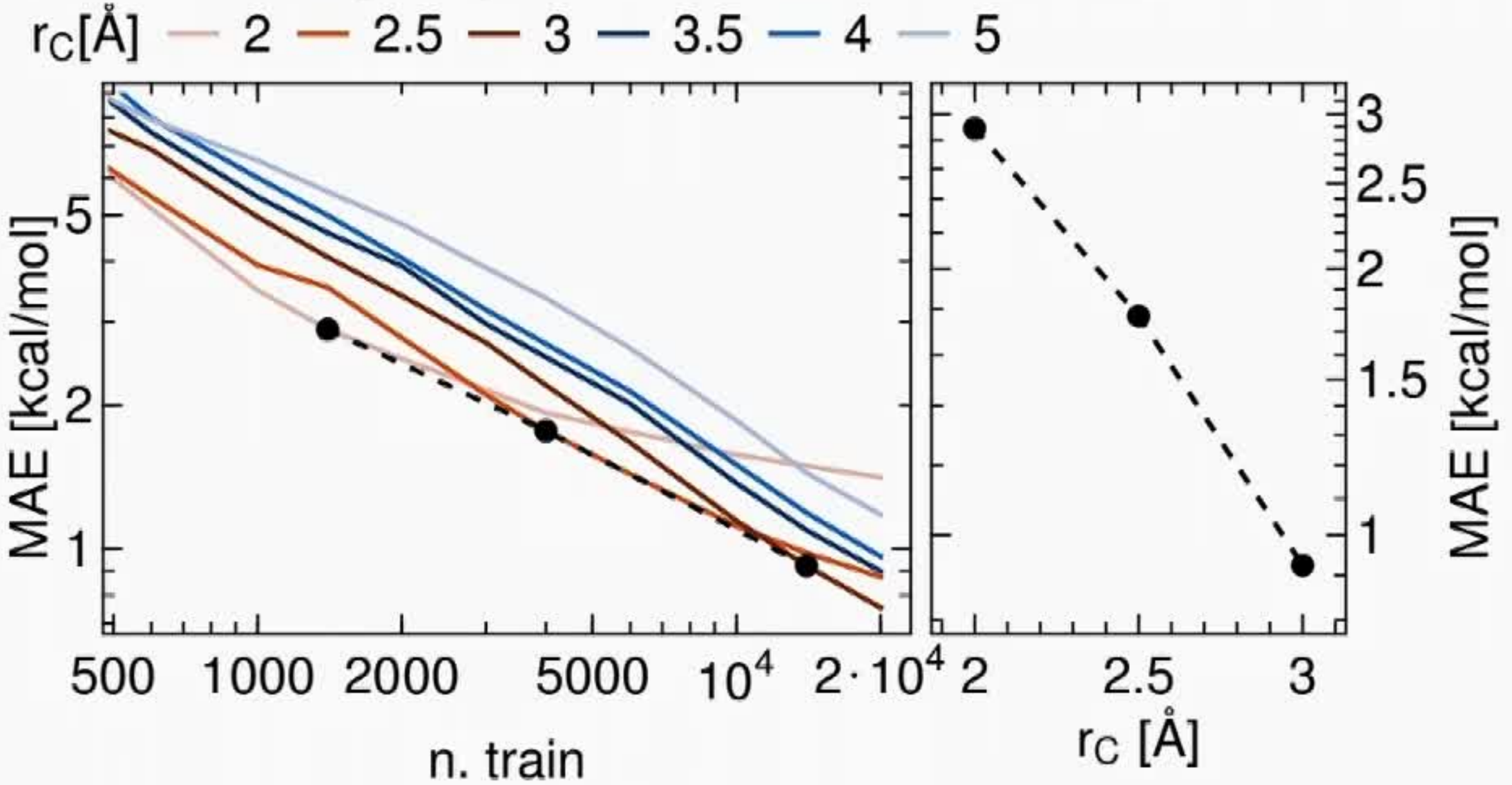




w/Emsley, Paruzzo, Hofstetter, http://shiftml.org

Understanding the Range of Interactions

- Environment kernels can be built for different cutoff radii
- Dimensionality/accuracy tradeoff, a measure of the range of interactions
- A multi-scale kernel $K(A,B) = \sum_i w_i K_i(A,B)$ yields the best of all worlds - chemical accuracy on QM9 with $\sim$ 5000 train structures



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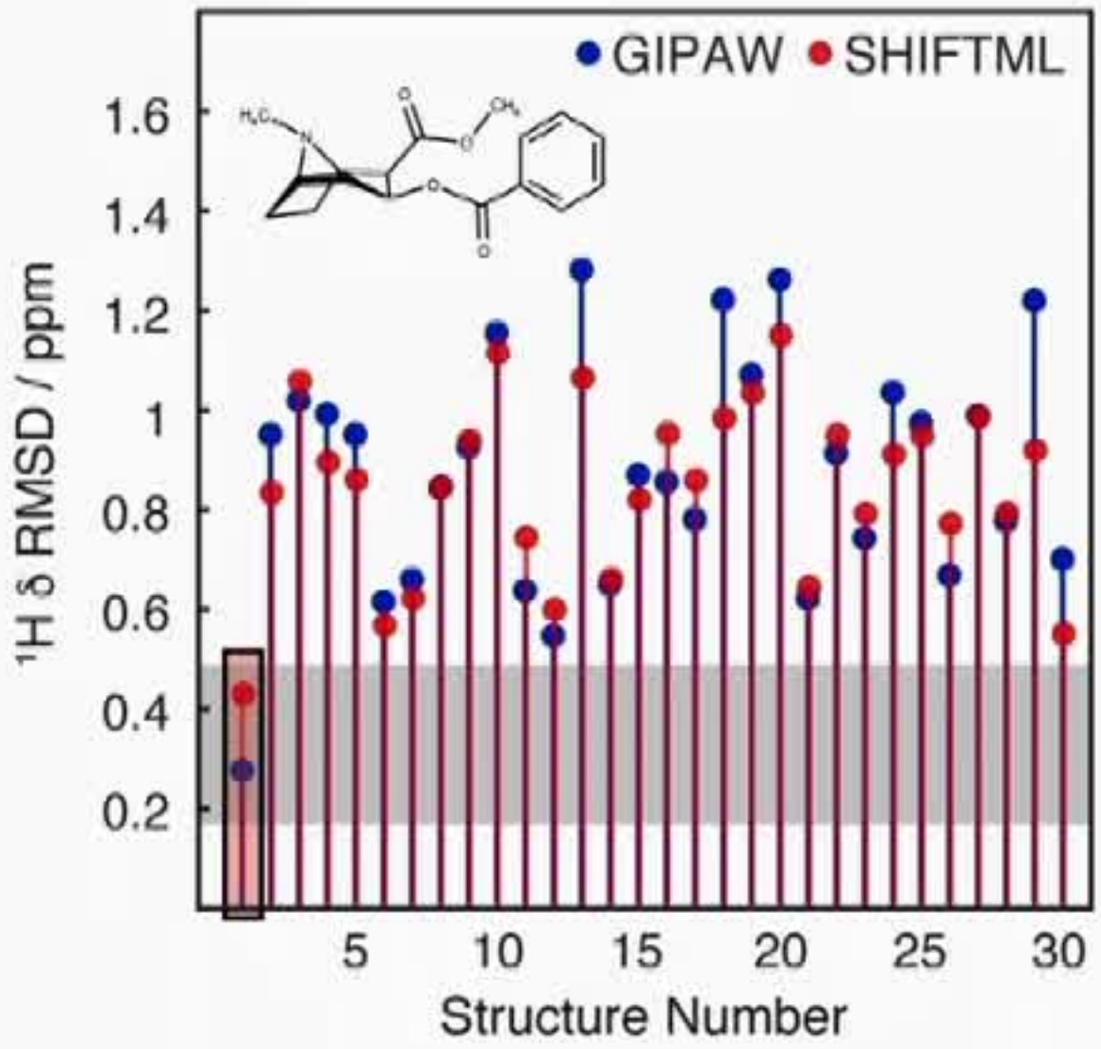

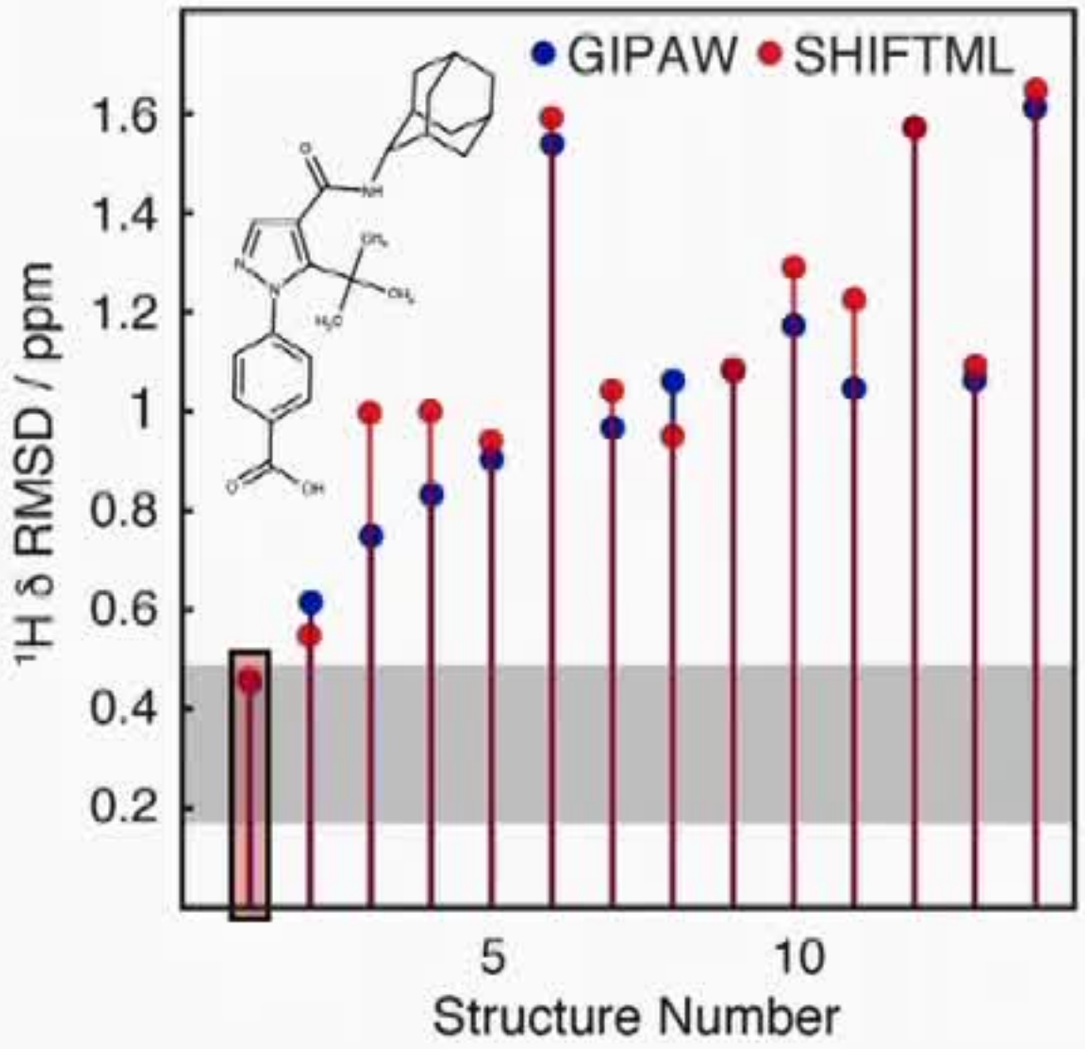

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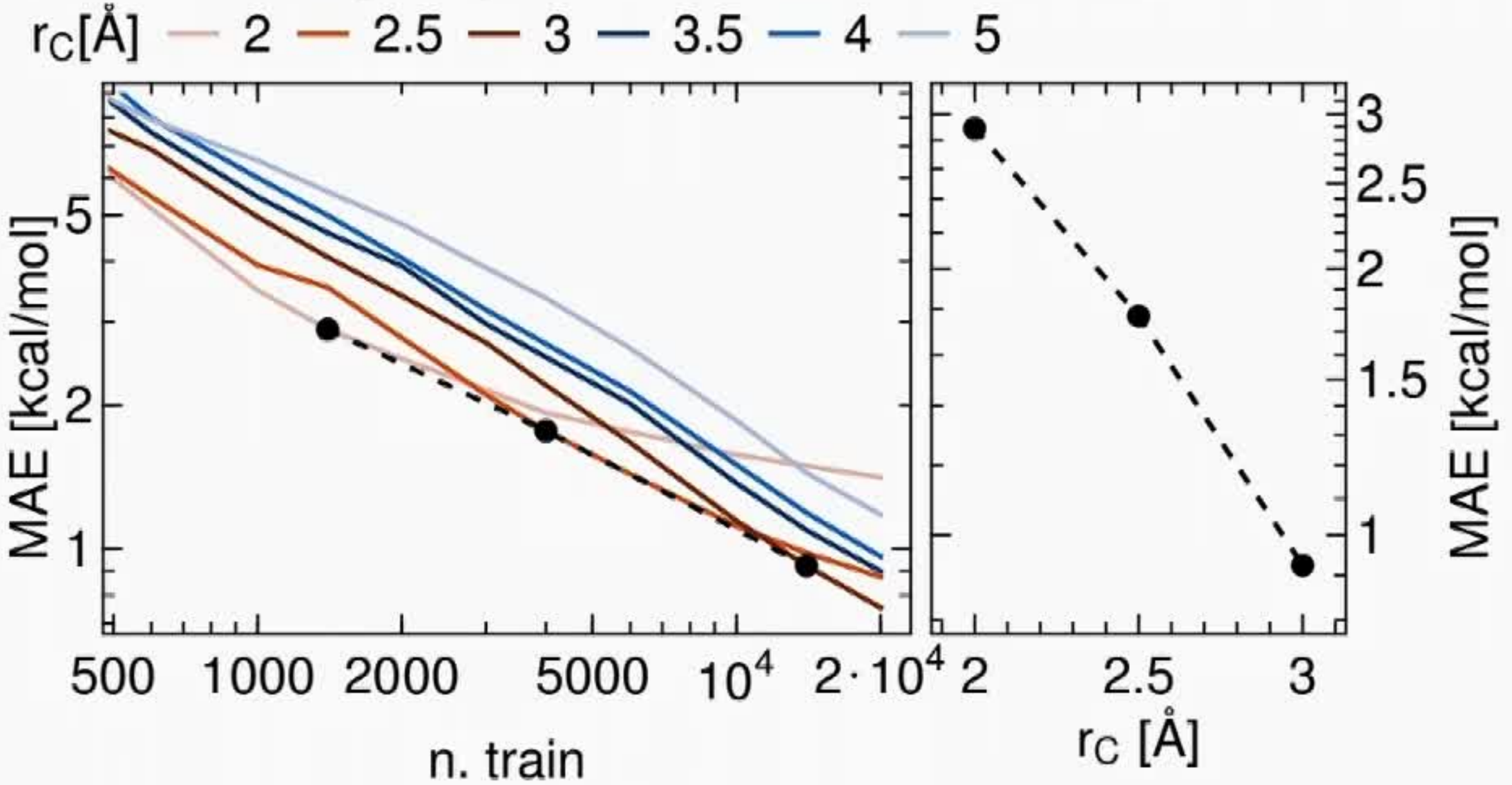

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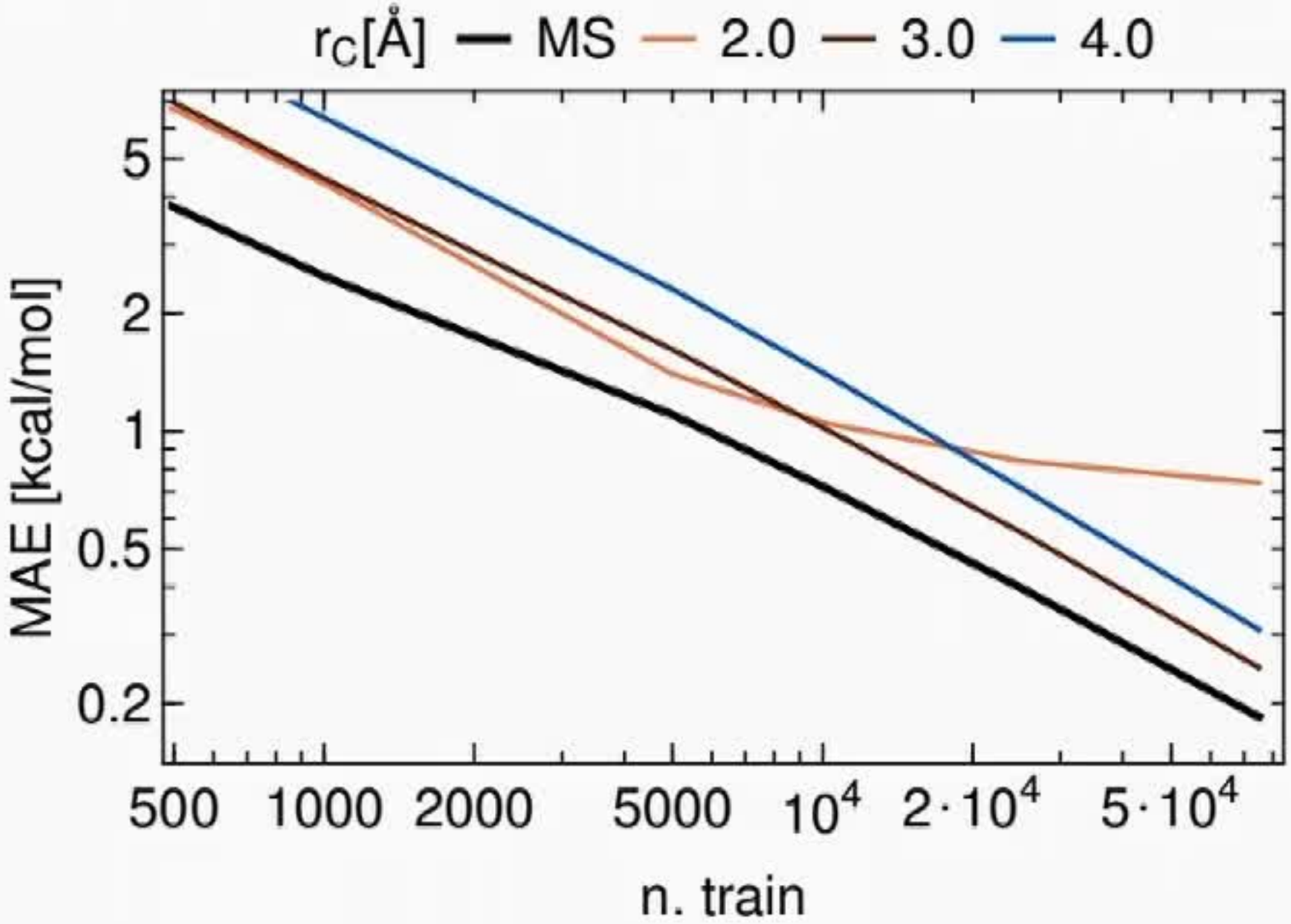
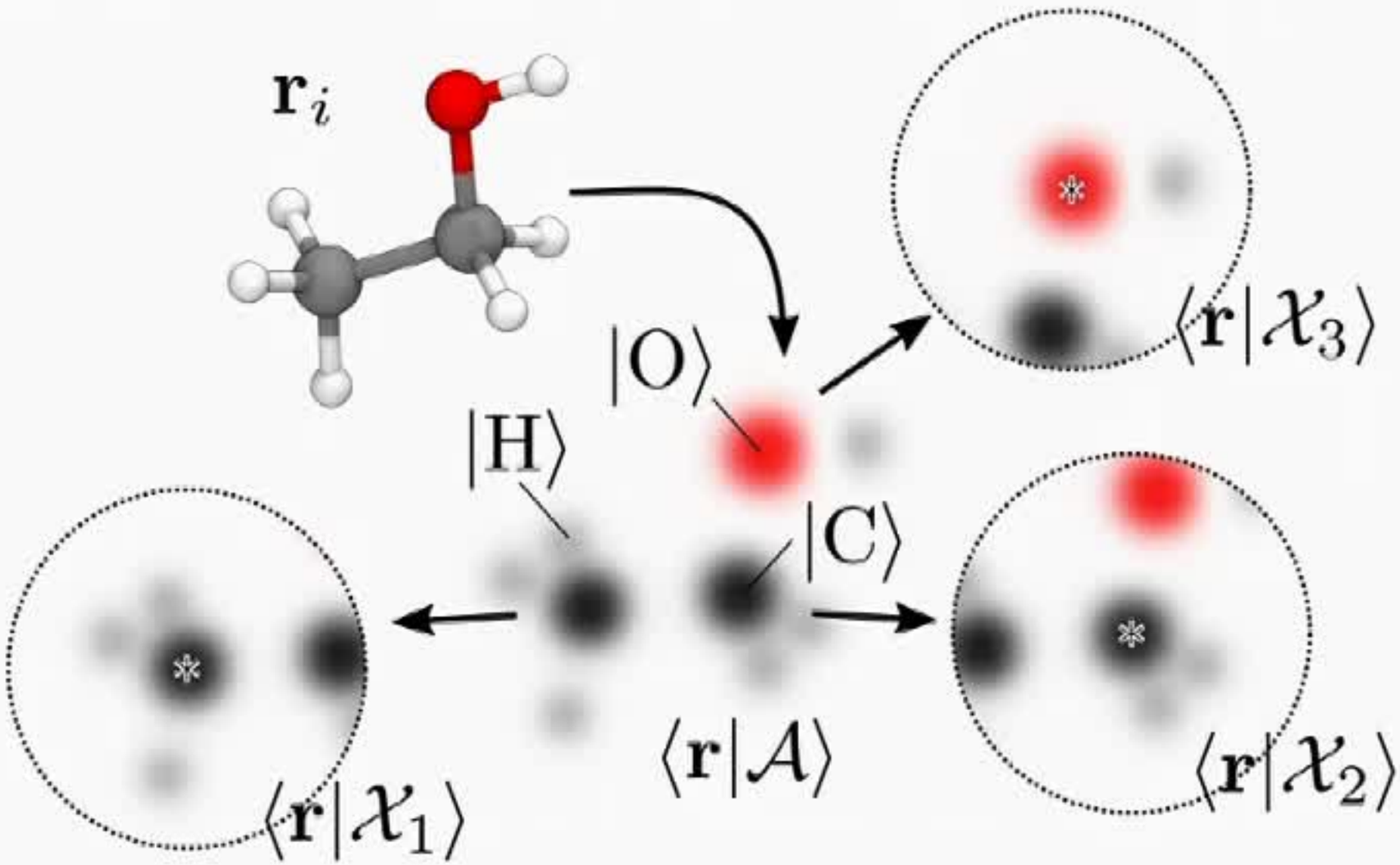


Bartók, De, Kermode, Bernstein, Csányi, **MC**, Science Advances (2017)

Machine-learning the periodic table

- SOAP normally considers orthogonal 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 feature space, more effective learning . . .
- . . . and as by-product get a data-driven version of the periodic table!



Willatt, Musil, **MC**, https://arxiv.org/pdf/1807.00236.pdf

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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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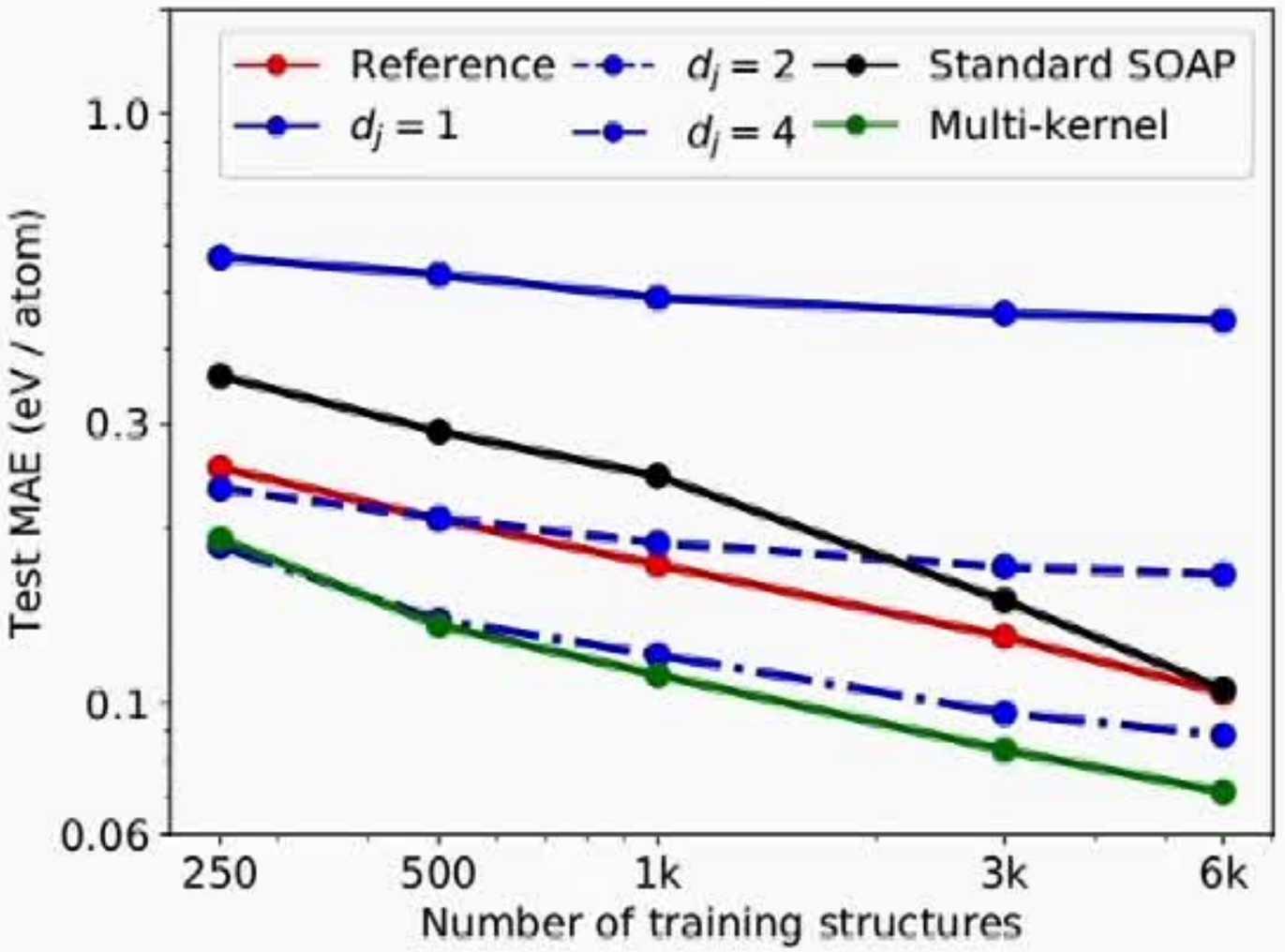

Elpasolite dataset from Faber et al PRL (2016). Reference curve (red) from Faber et al. JCP (2018)

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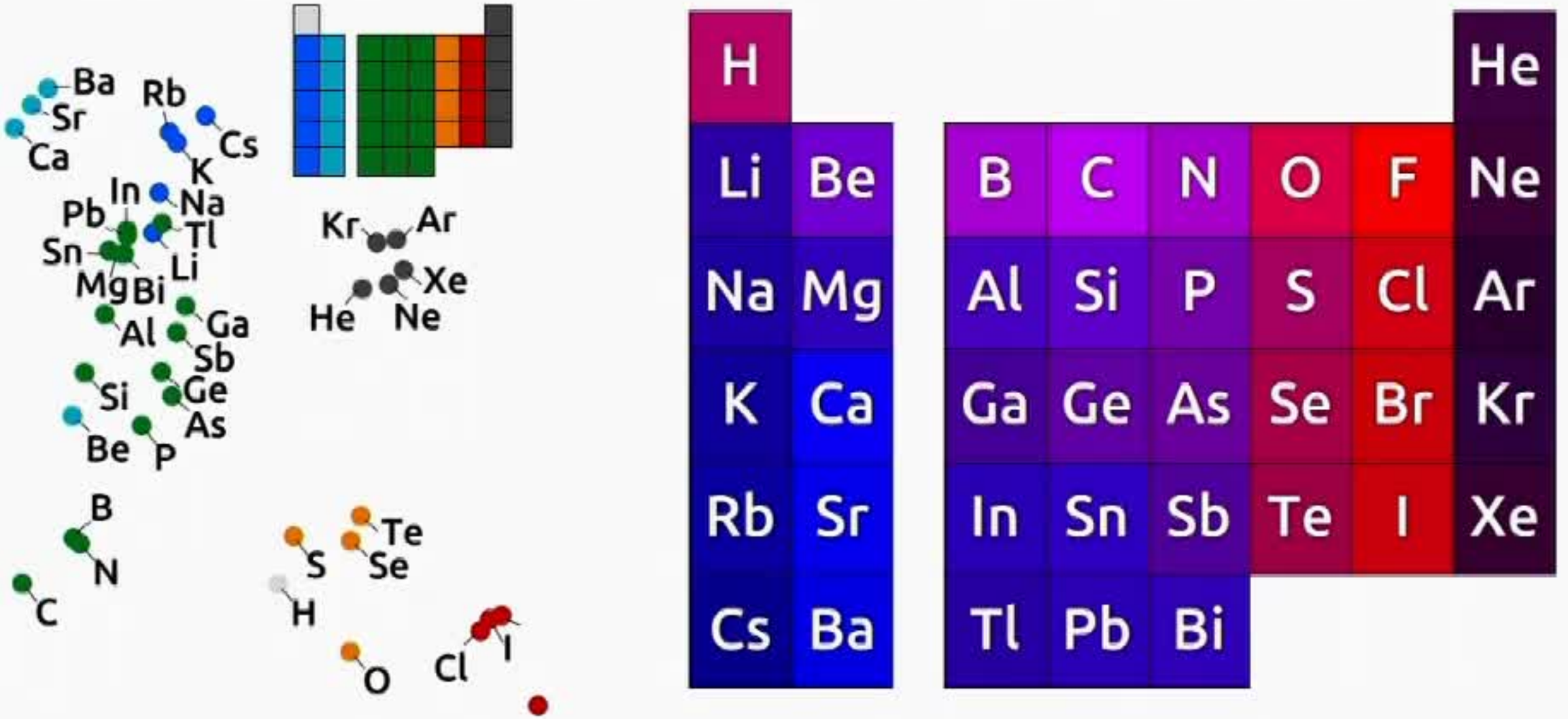


Symmetries in Machine-Learning

- In a Gaussian Process framework, the kernel represents correlations between properties. This must be reflected in how it transforms under symmetry operations applied to the inputs

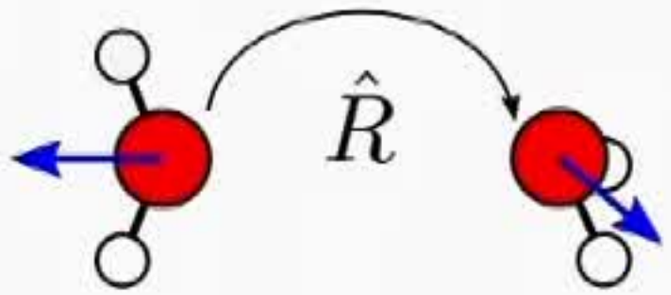
$$k(\mathcal{X},\mathcal{X}') \leftrightarrow \langle y(\mathcal{X}); y(\mathcal{X}')\rangle \text{, so } k\left(\hat{S}\mathcal{X},\hat{S}'\mathcal{X}'\right) \leftrightarrow \left\langle y\left(\hat{S}\mathcal{X}\right); y\left(\hat{S}'\mathcal{X}'\right)\right\rangle$$

- Properties that are *invariant* under $\hat{S}$ must be learned with a kernel that should be insensitive to the operation

$$k\left(\hat{S}\mathcal{X},\hat{S}'\mathcal{X}'\right) = k(\mathcal{X},\mathcal{X}')$$

- How about machine-learning tensorial properties **T**? The kernel should be *covariant* to rigid rotations - need a symmetry-adapted framework

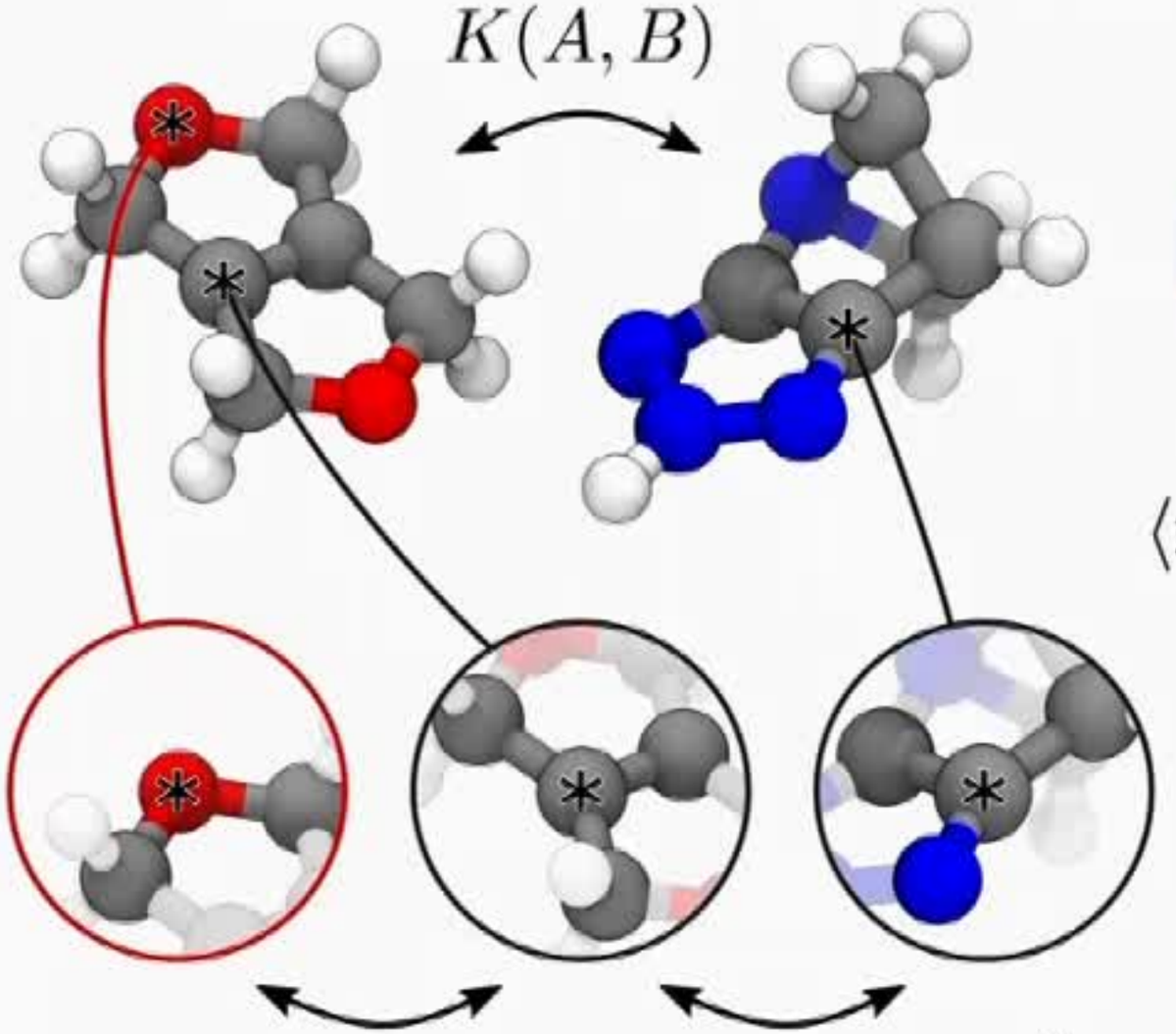
$$k_{\mu\nu}(\mathcal{X},\mathcal{X}') \leftrightarrow \langle T_\mu(\mathcal{X}); T_\nu(\mathcal{X}')\rangle \rightarrow k_{\mu\nu}\left(\hat{R}\mathcal{X},\hat{R}'\mathcal{X}'\right) = R_{\mu\mu'}k_{\mu'\nu'}(\mathcal{X},\mathcal{X}')R'_{\nu\nu'}$$

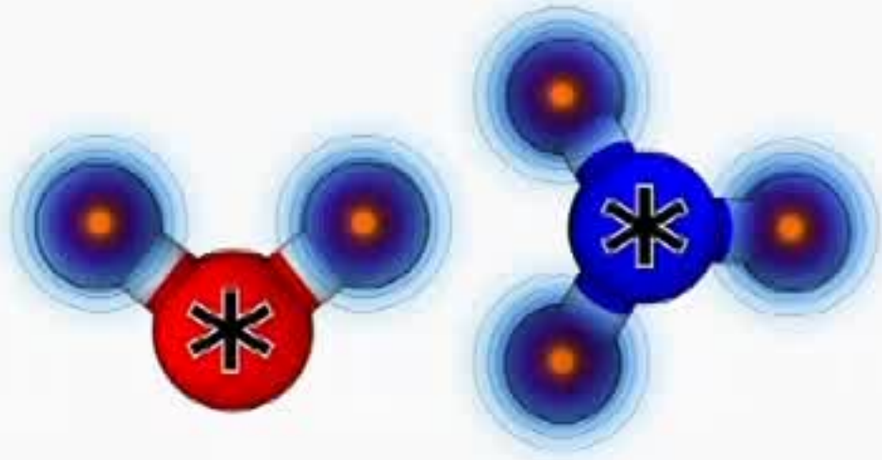


Glielmo, Sollich, & De Vita, PRB (2017); Grisafi, Wilkins, Csányi, & **MC**, PRL (2018)

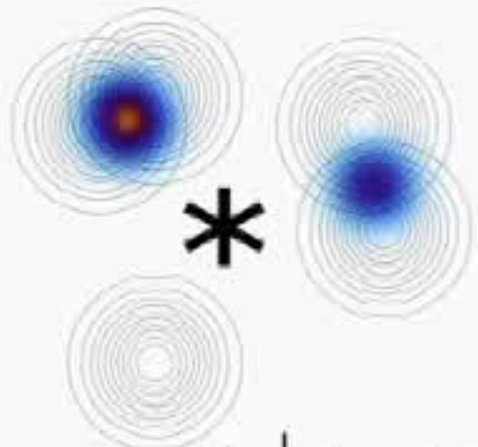
λ−SOAP: A $SO(3)$ Compliant Kernel for Tensors

- Recall the definition of SOAP, based on the atom-density overlap
- Each tensor can be decomposed into irreducible spherical components $\mathbf{T}^\lambda$, corresponding to the representations of $SO(3)$
- A hierarchy of λ-SOAP kernels can be defined to learn tensorial quantities





$$\langle \mathbf{r}|\mathcal{X}_j\rangle = \psi(\mathbf{r}) = \sum_i g(\mathbf{r}-\mathbf{r}_{ij})$$



$$\left\langle \mathcal{X}^{(\nu)} \middle| \mathcal{X}'^{(\nu)} \right\rangle_{\hat{R}} \sim \int \mathrm{d}\hat{R} \left| \int \psi(\mathbf{r})\psi'(\hat{R}\mathbf{r})\mathrm{d}\mathbf{r} \right|^\nu$$

Grisafi, Wilkins, Csányi, & **MC**, PRL (2018)

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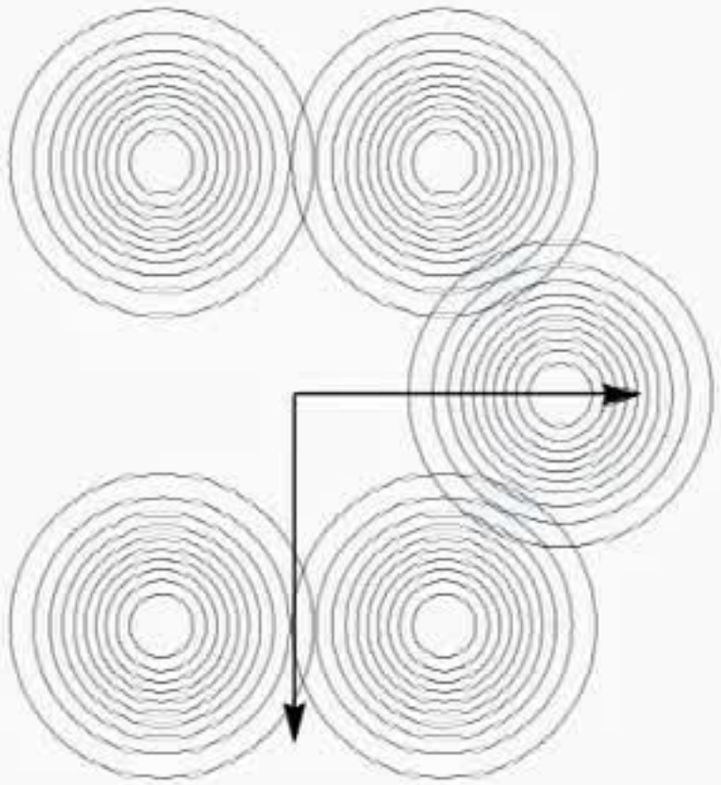
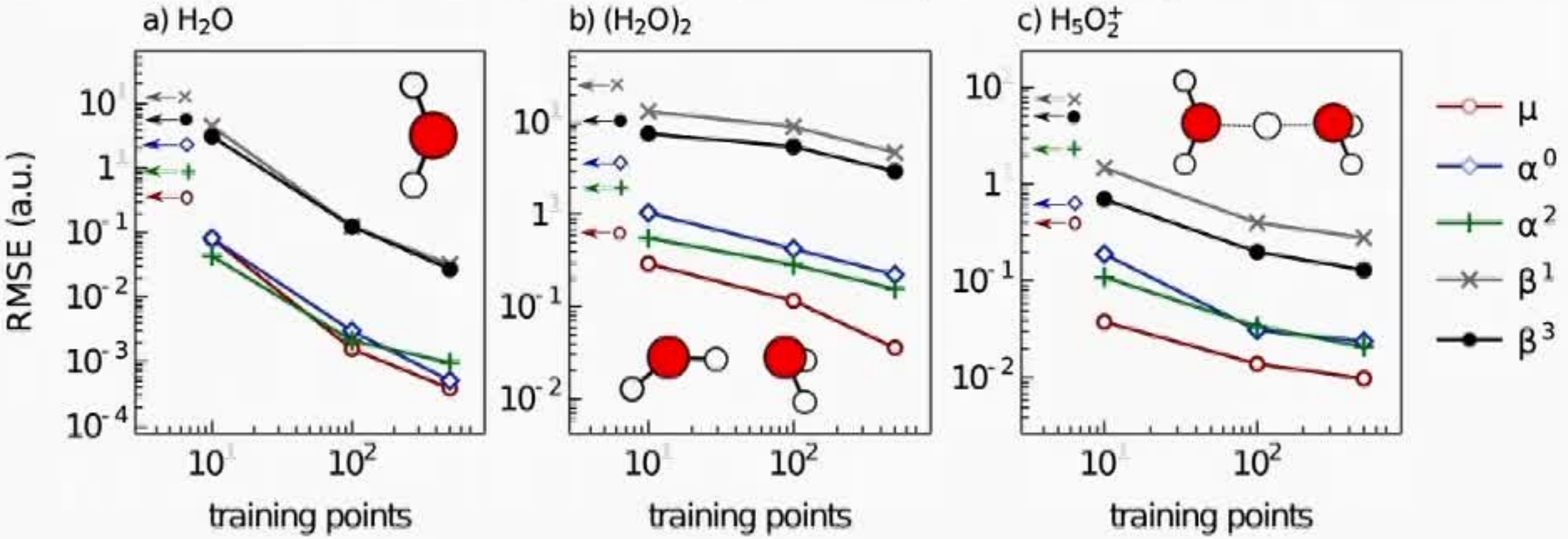


$$k^{\lambda}_{\mu\nu}\left(\mathcal{X},\mathcal{X}'\right)=\int \mathrm{d}\hat{R} D^{\lambda}_{\mu\nu}\left(\hat{R}\right)\left|\psi\left(\mathbf{r}\right)\psi'\left(\hat{R}\mathbf{r}\right)\right|^{\nu} \qquad \left\langle \mathbf{r}\lambda\mu\middle|\mathcal{X}_j\right\rangle=\left|\lambda\mu\right\rangle\psi\left(\mathbf{r}\right)$$

Grisafi, Wilkins, Csányi, & **MC**, PRL (2018)

Machine-Learning the Dielectric Response of Water

- A demonstration of the SA-GPR framework, and the λ-SOAP kernel - learning the dielectric response of water oligomers
- The kernels for multi-atomic systems can be built with an additive ansatz - and that gives meaningful partitioning in molecular contributions
- Works great for bulk systems (liquid & ice) after fixing non-additive terms



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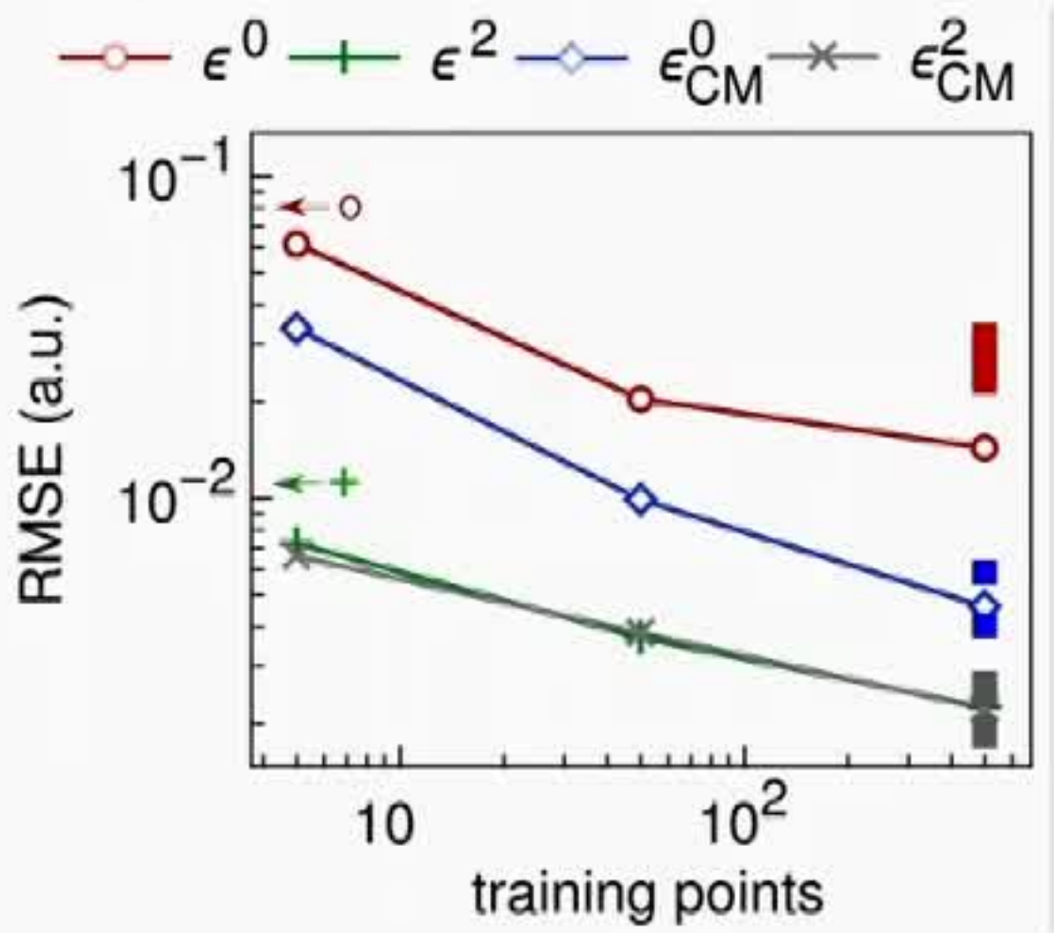

Clausius-Mossotti: $\alpha = (\epsilon - 1)(\epsilon + 2)^{-1} V$

Grisafi, Wilkins, Csányi, & **MC**, PRL (2018)

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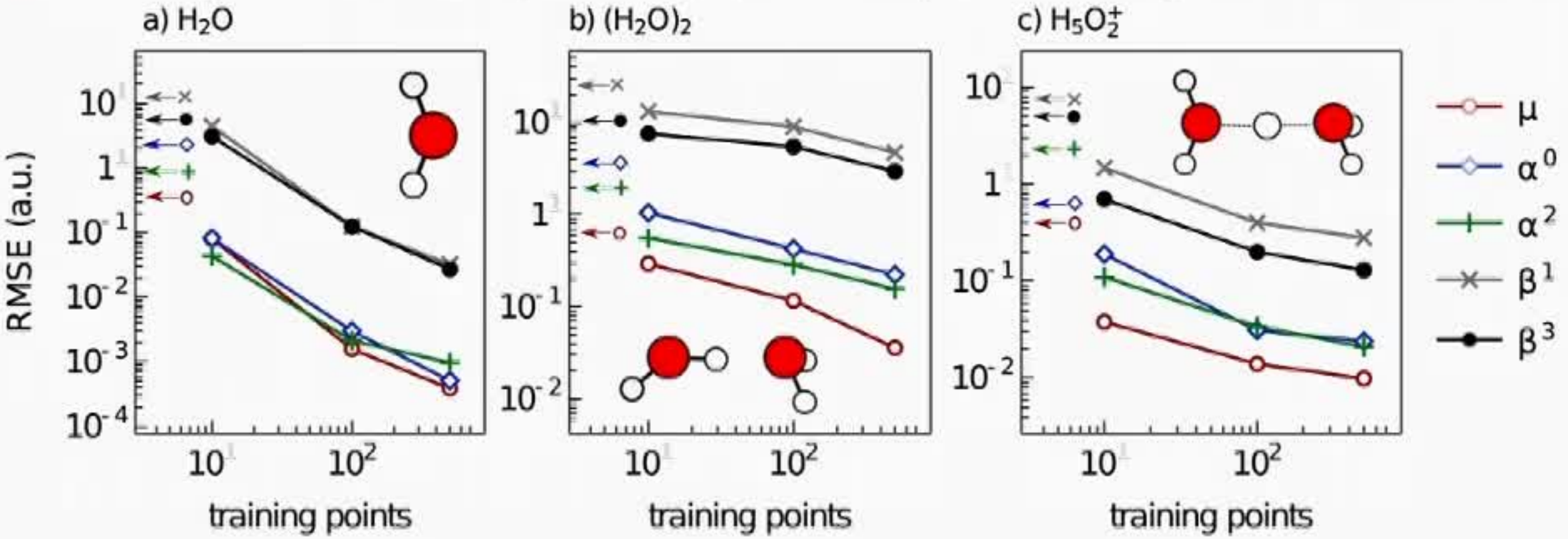

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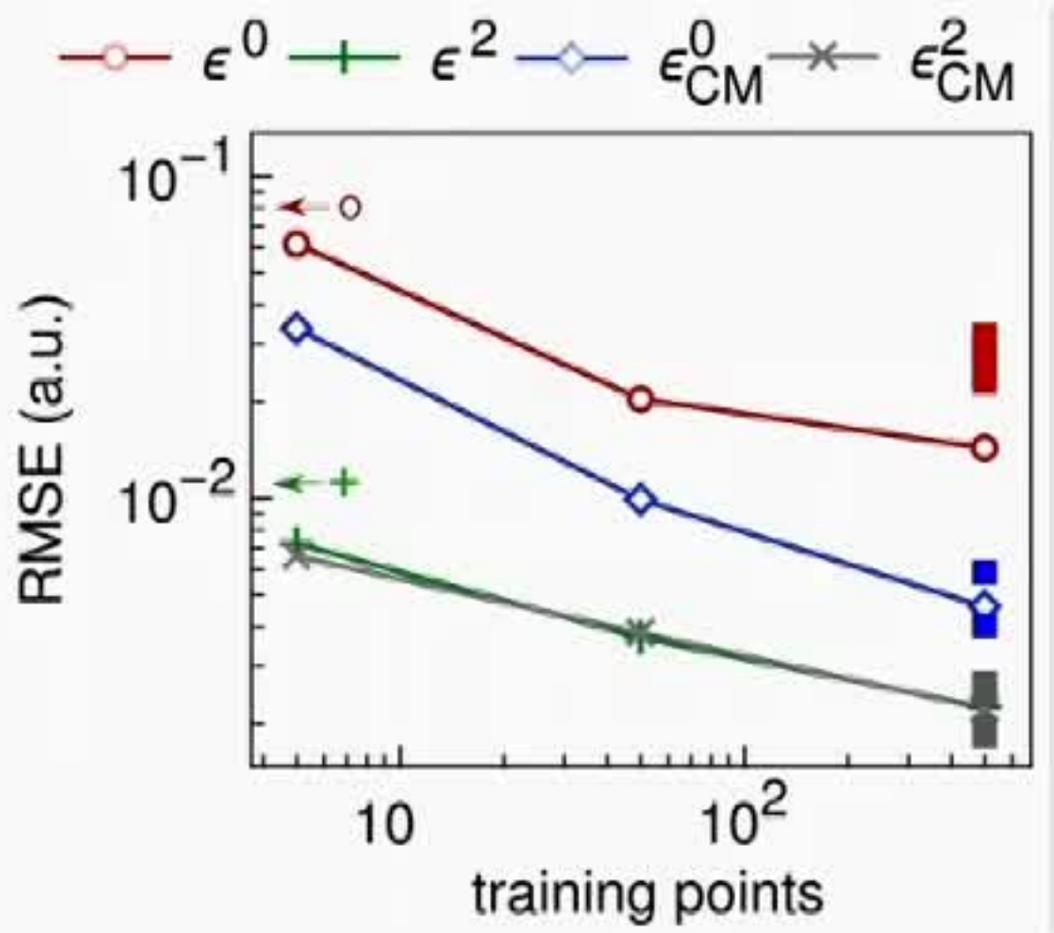

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Predicting the Full Polarizability of Molecules

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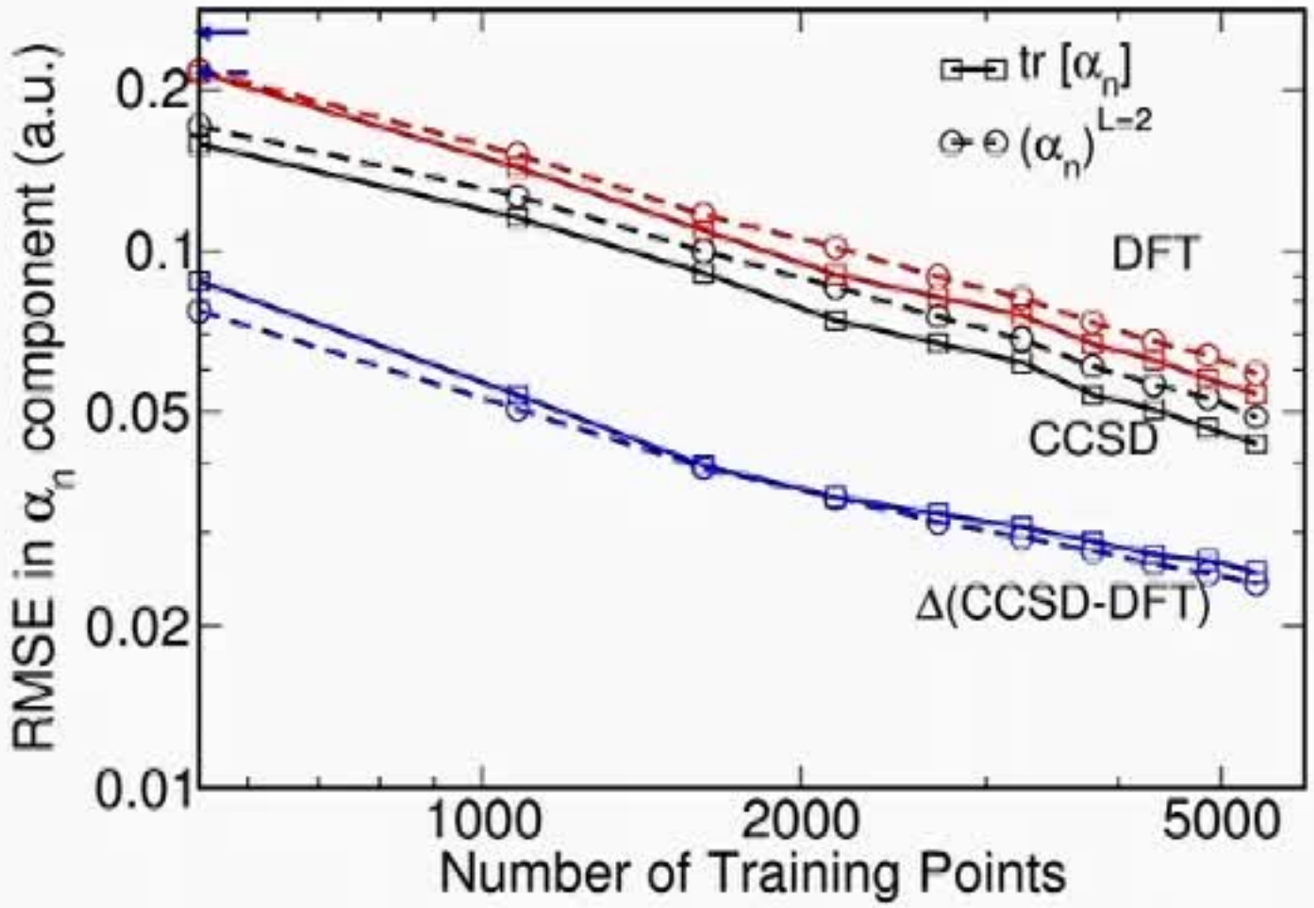
- Better than DFT on 52 challenging showcase molecules.



QM7b dataset (organic molecules, <7 CNOFS, Montavon et al. NJP (2013)); w/ R. DiStasio, Y. Yang

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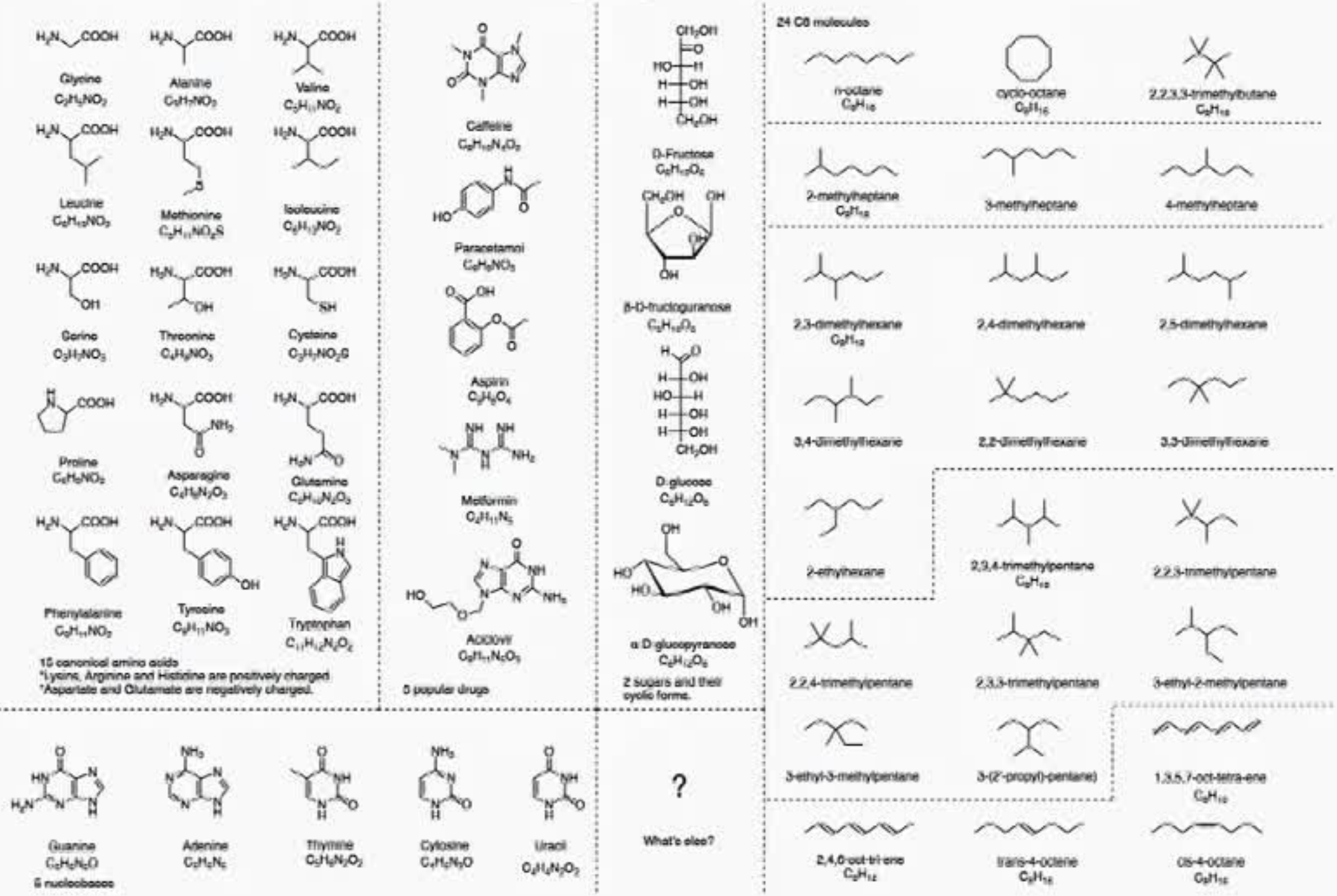

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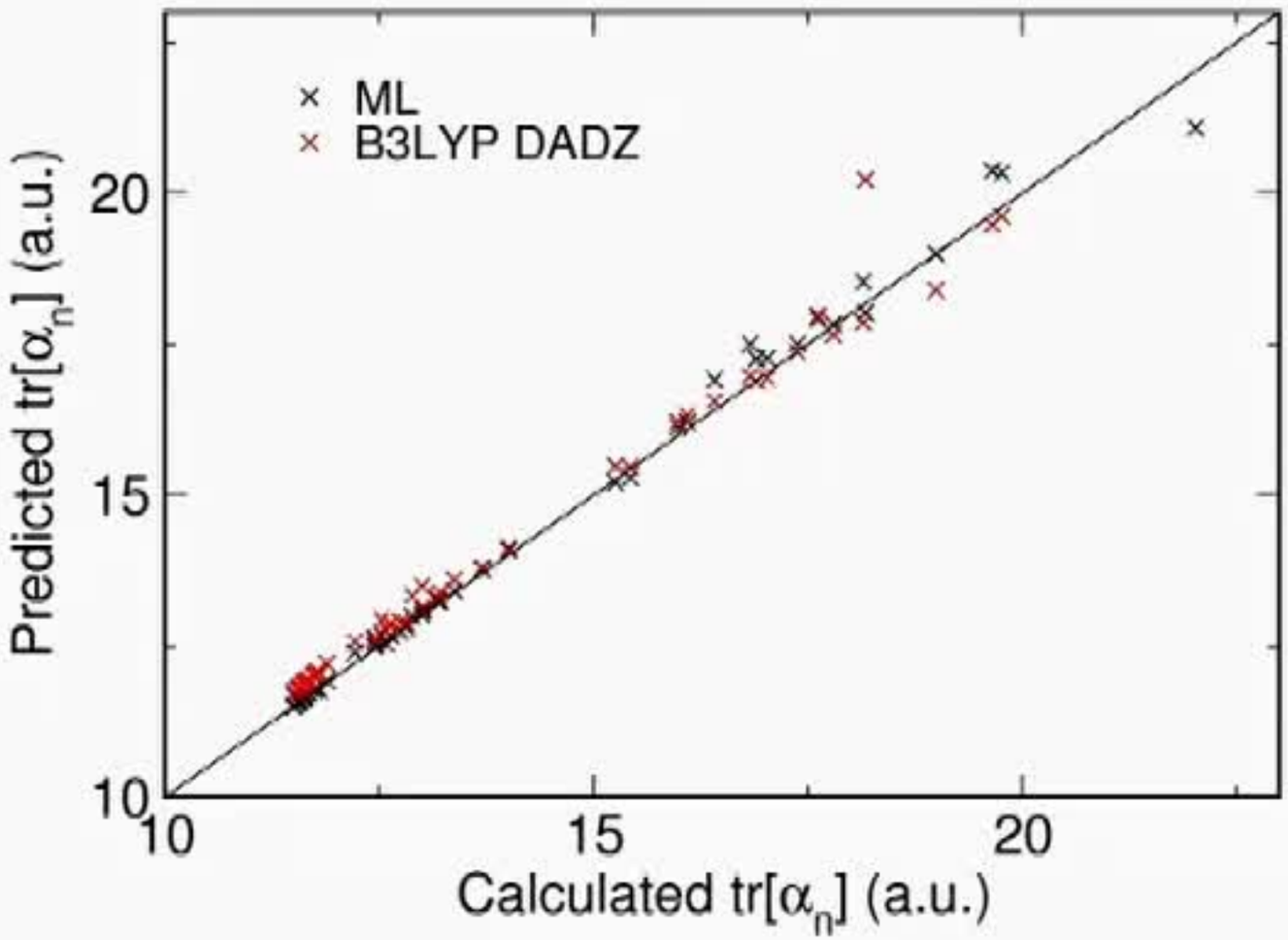

$\sigma(\cdot)$ [a.u.]	$\lambda = 0$	$\lambda = 2$
CCSD vs B3LYP	0.350	0.456
SA-GPR vs CCSD	0.244	0.205
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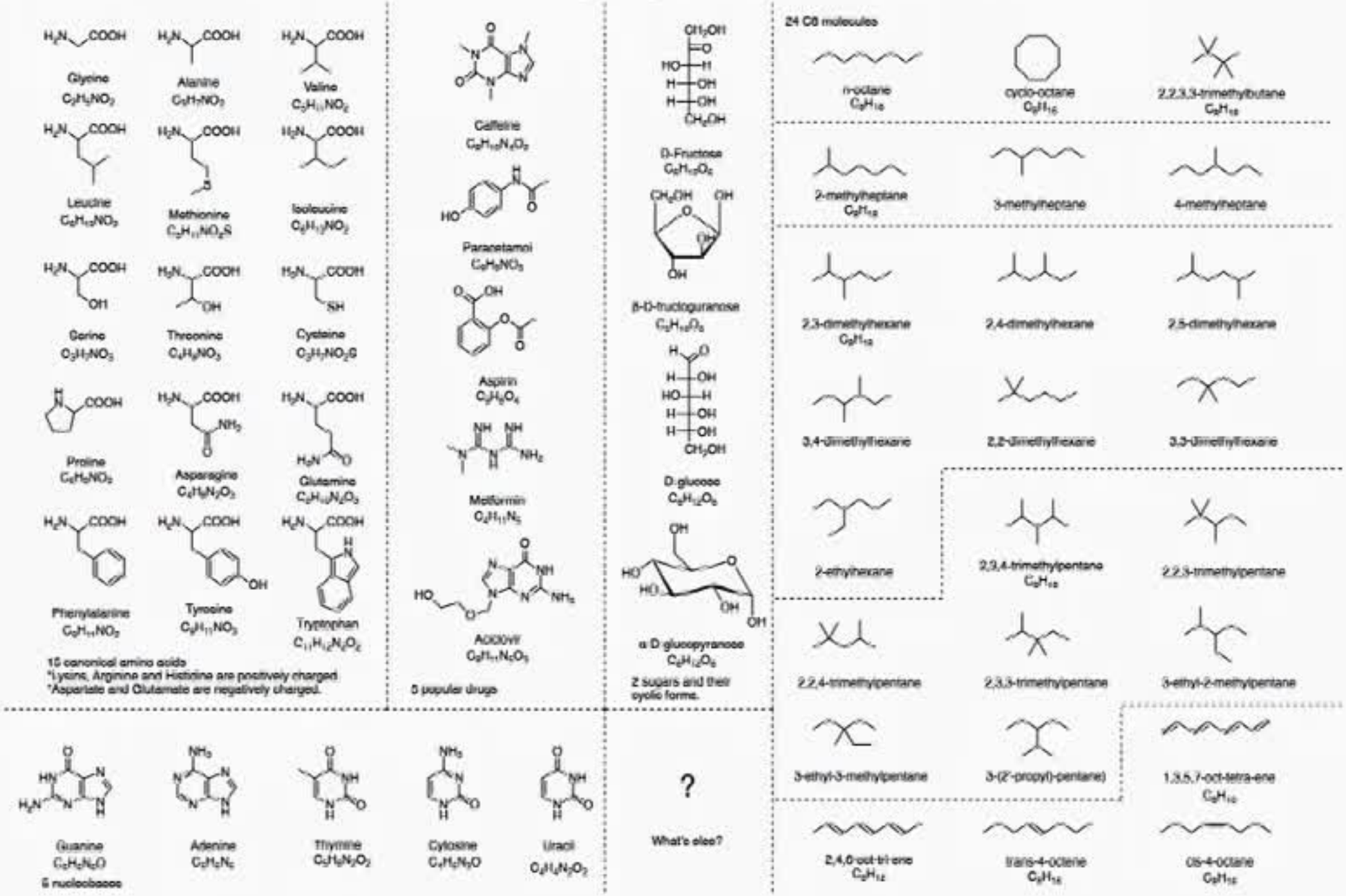

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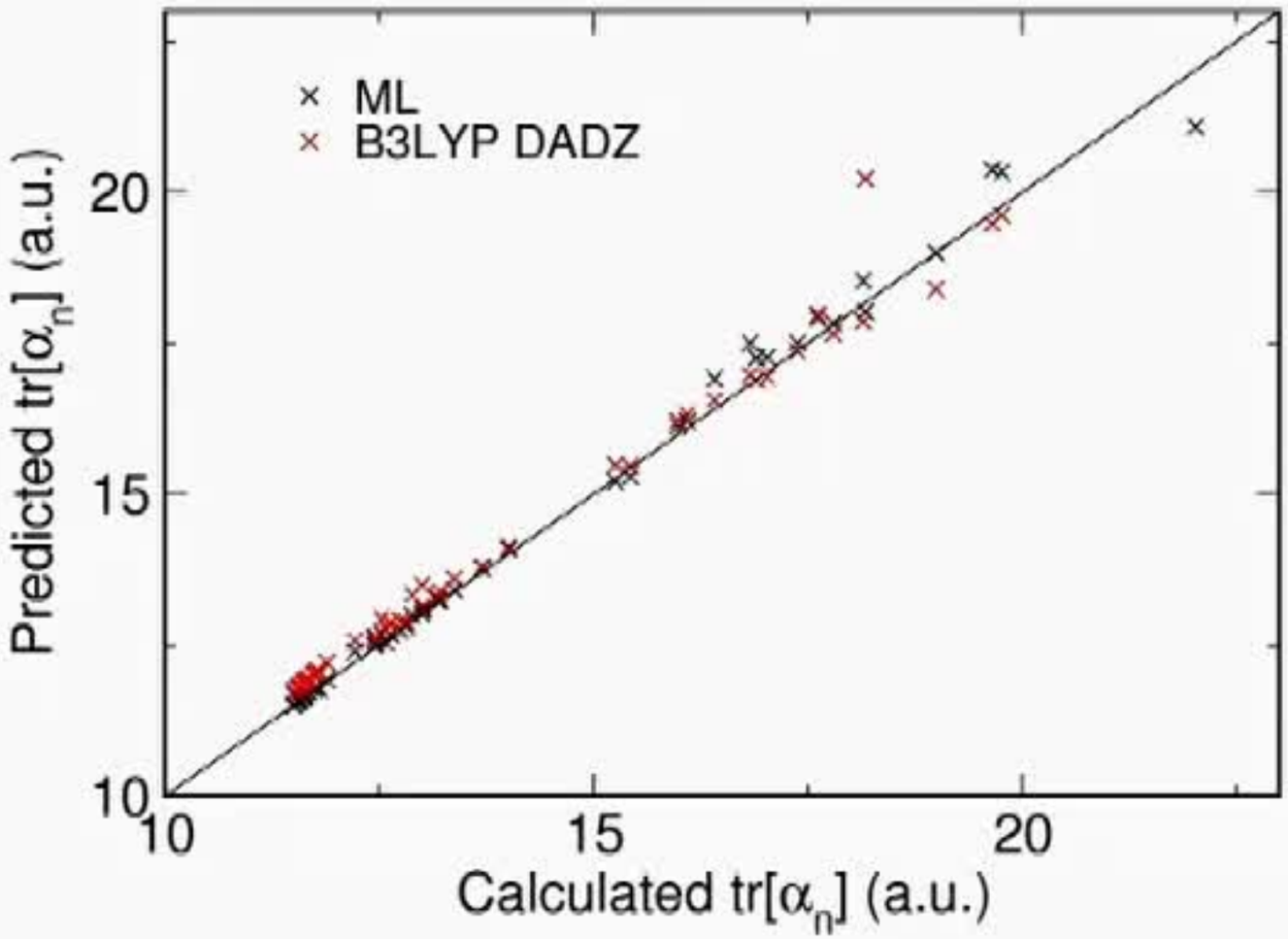

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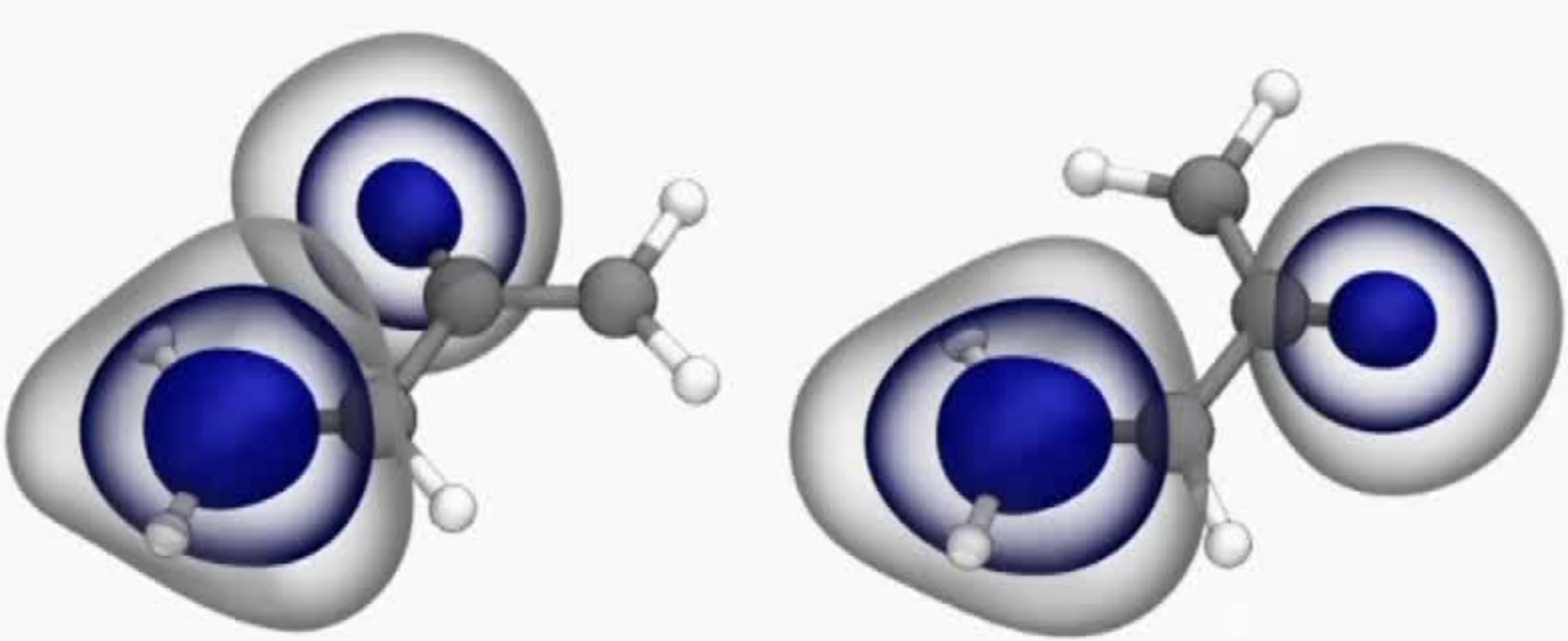
Machine Learning the Electron Density

- Decompose the density in localized ("à la Boys/Wannier") atom-centered contributions using a $\phi_k \equiv R_n Y_m^l$ expansion

$$\mathcal{F}(\rho) = \int d\mathbf{r} \left| \rho(\mathbf{r}) - \sum_{ik} c_{ik} \phi_k (\mathbf{r} - \mathbf{r}_i) \right|^2$$

- Machine-learn the expansion coefficients
- Highly transferable: learn on butadiene, predict on octatetraene



w/ C. Corminboeuf, B. Meyer, A. Fabrizio

Machine Learning the Electron Density

- Decompose the density in localized ("à la Boys/Wannier") atom-centered contributions using a $\phi_k \equiv R_n Y_m^l$ expansion

$$\mathcal{F}(\rho) = \int d\mathbf{r} \left| \rho(\mathbf{r}) - \sum_{ik} c_{ik} \phi_k (\mathbf{r} - \mathbf{r}_i) \right|^2$$

- Machine-learn the expansion coefficients
- Highly transferable: learn on butadiene, predict on octatetraene