# Mapping transition metal chemical space with continuous descriptors – feature selection and implications for machine learning models

**Jon Paul Janet** [1] Terry Z.H. Gani [1] Heather Kulik [1]

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254th American Chemical Society National Meeting
Washington, DC

08.23.17

# Data-driven molecular design

**Machine learning is transforming how we design new materials**

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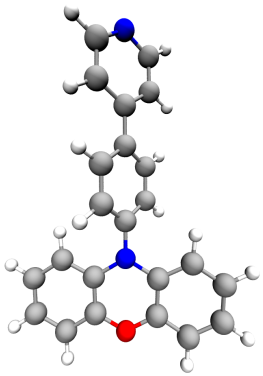
Gomez-Bombarelli, R. *et al.*. *Nat. Mater.*, 15(10):1120-1127, 2016.



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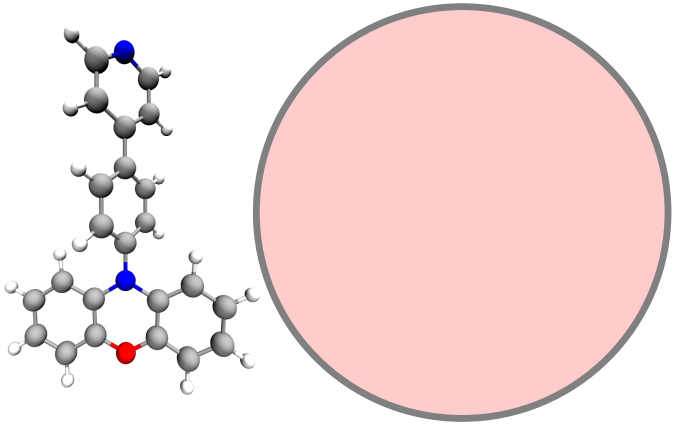

OLED chemical space

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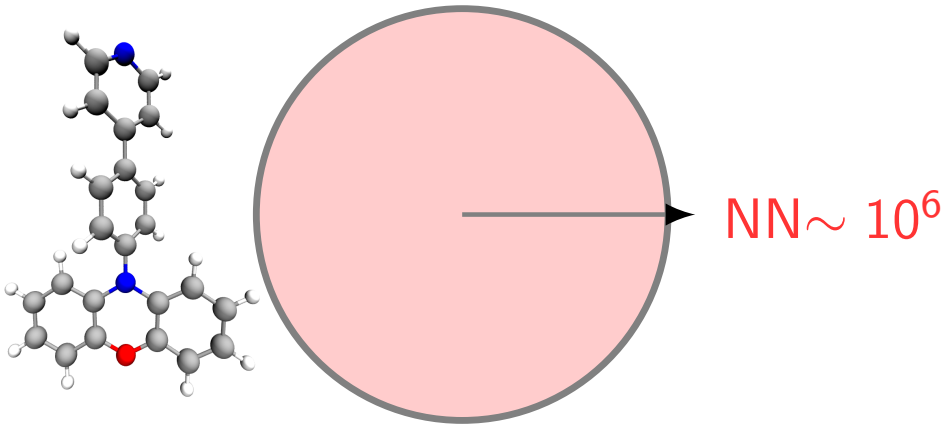


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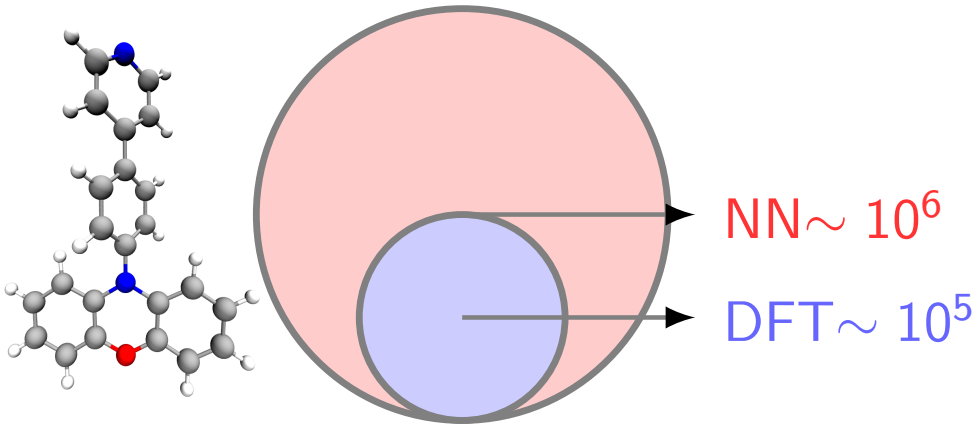


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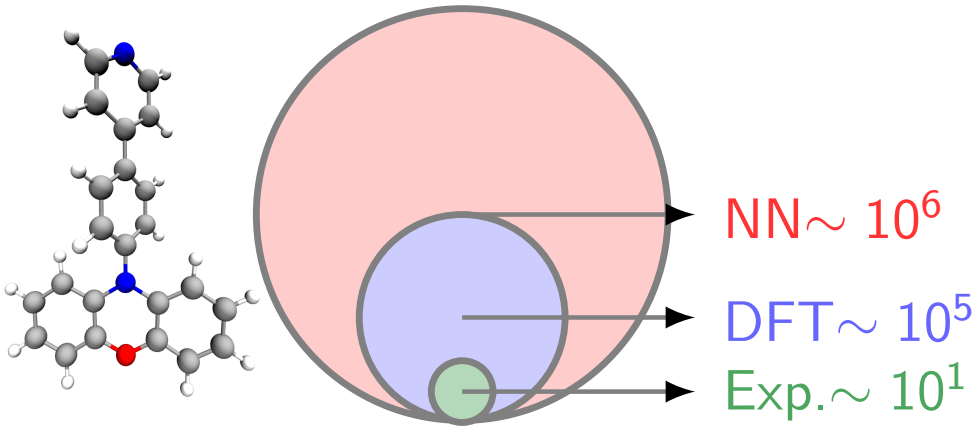


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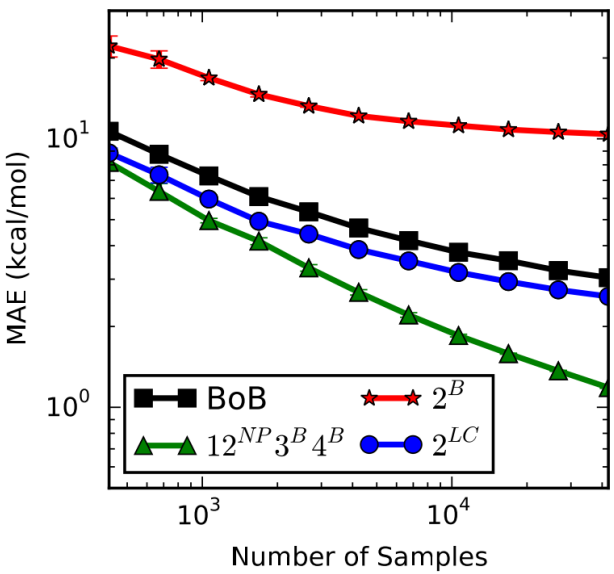


Collins *et al.* *arXiv*, 1701.06649, 2017.

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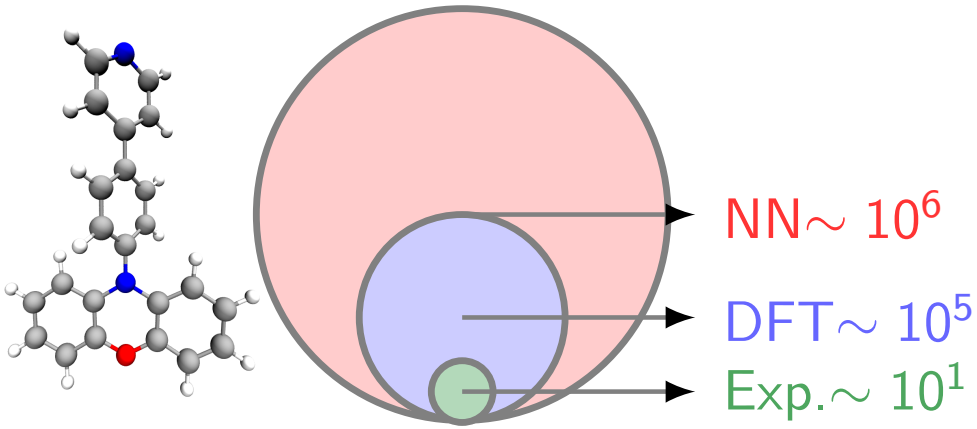


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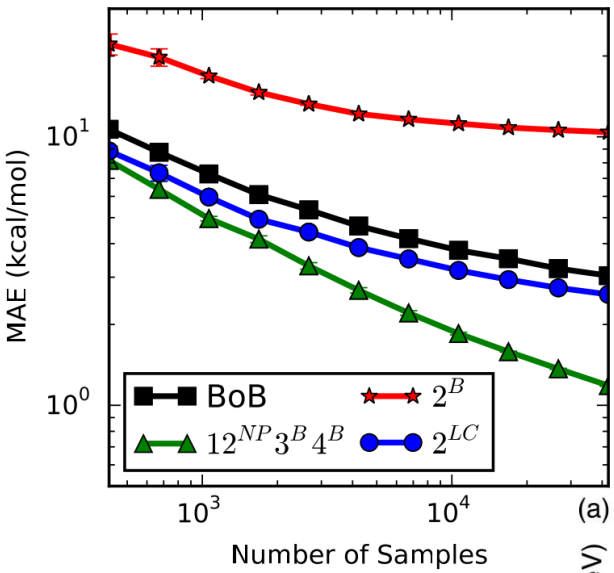


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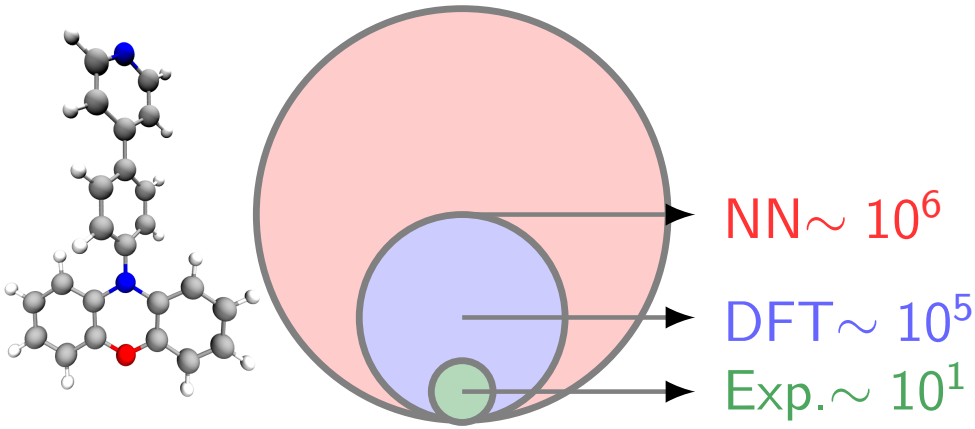


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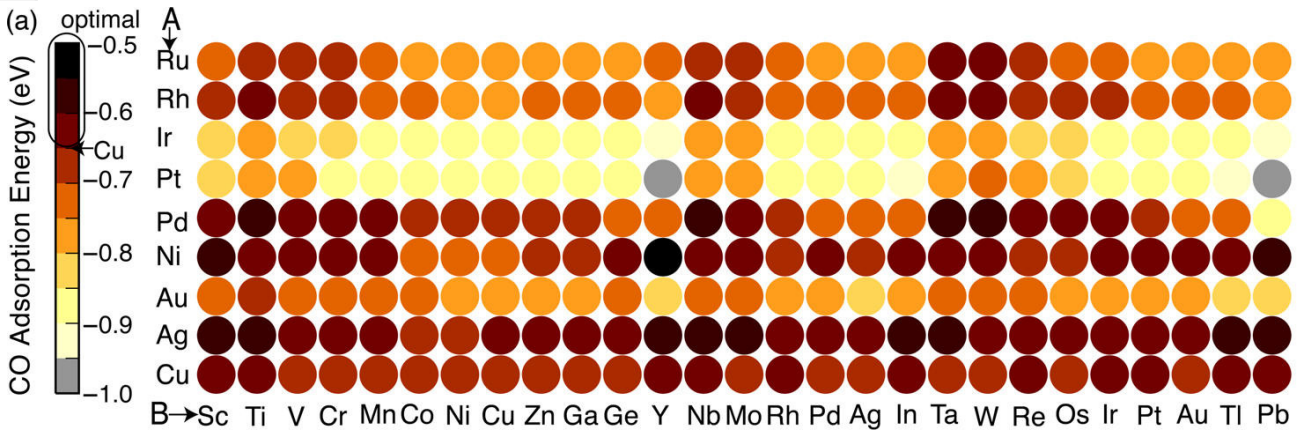
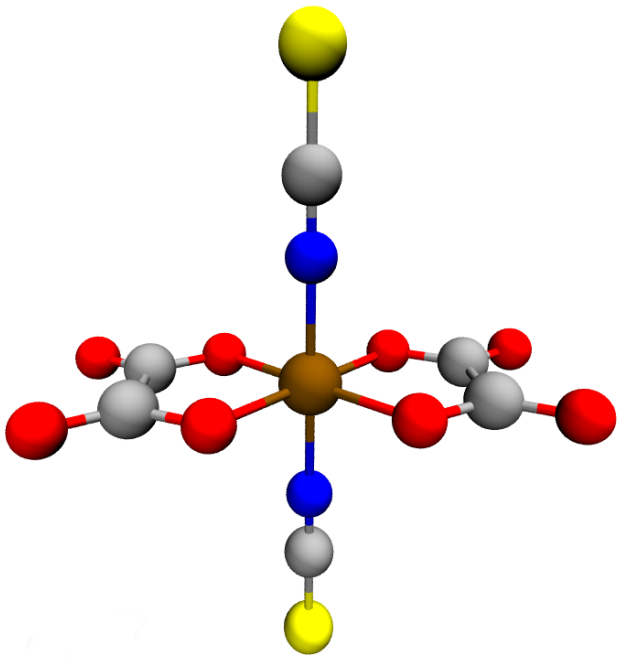
Ma, X. *et al.* J. Phys. Chem. Lett., (18):3528-3533, 2015.



# Transition metal complexes

**We study transition metal complexes**:



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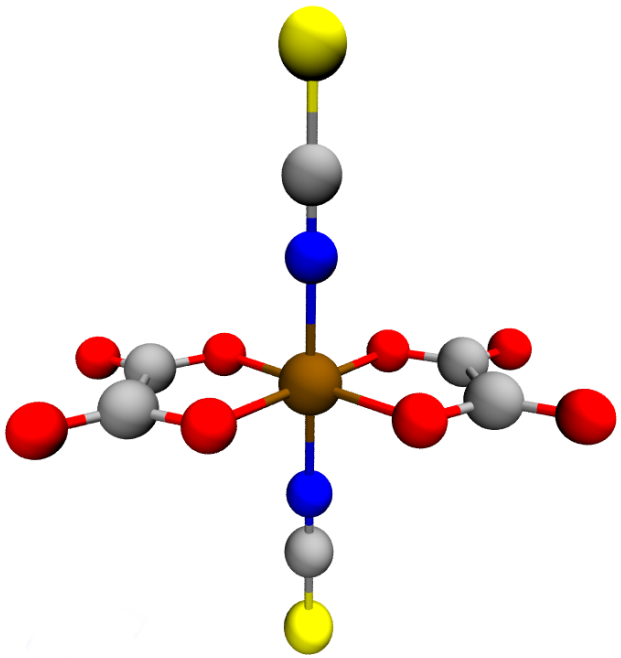
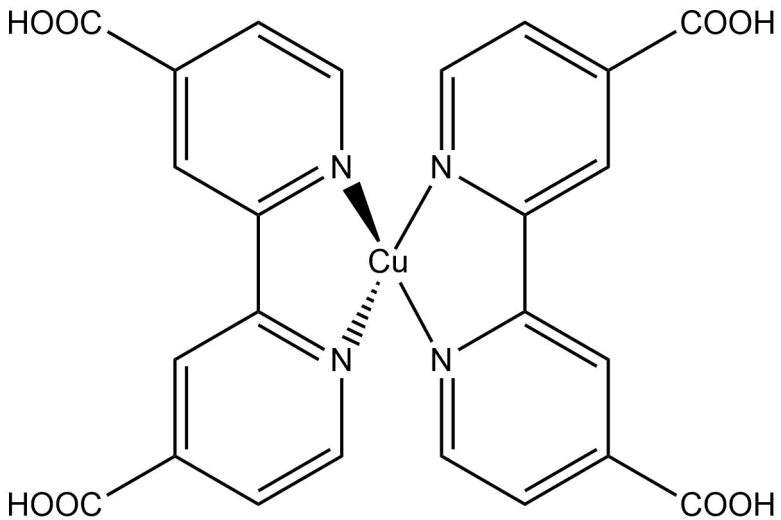


Dye sensitizers:




Bignozzi, C. *et al. Coord. Chem. Rev.*, 257(9): 1472–1492, 2013.

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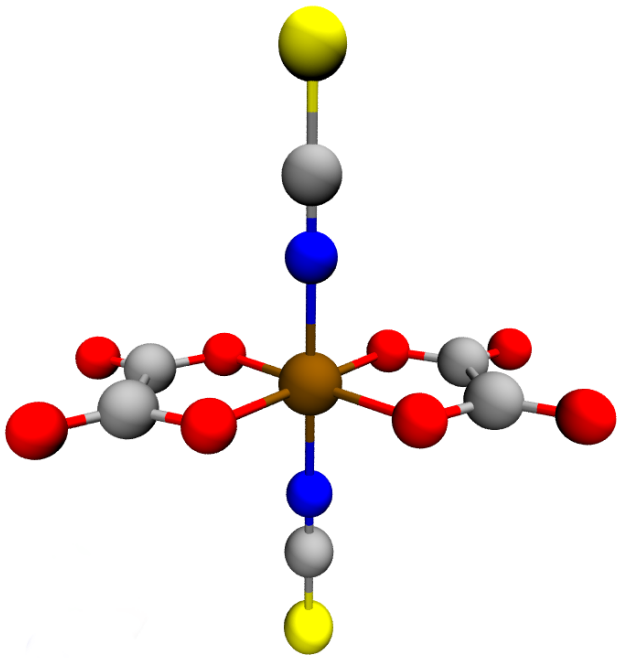
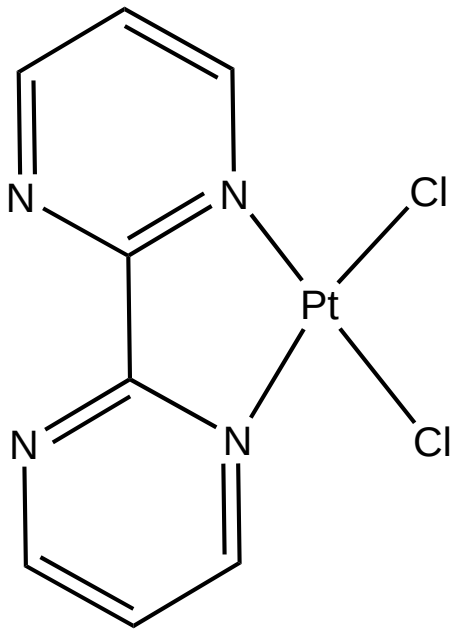


Catalysts:




Periana, R. A. *et al.* *Science*, 280(5363):560–564, 1998.

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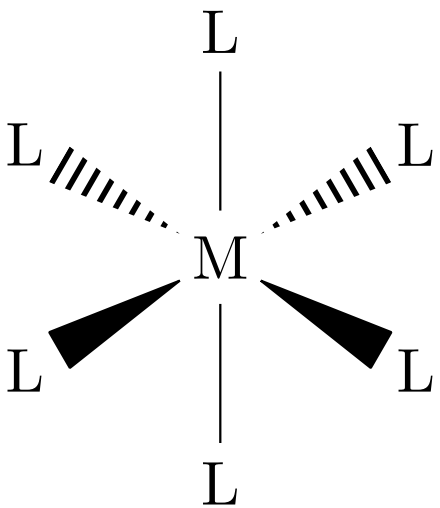
✓ tunable properties



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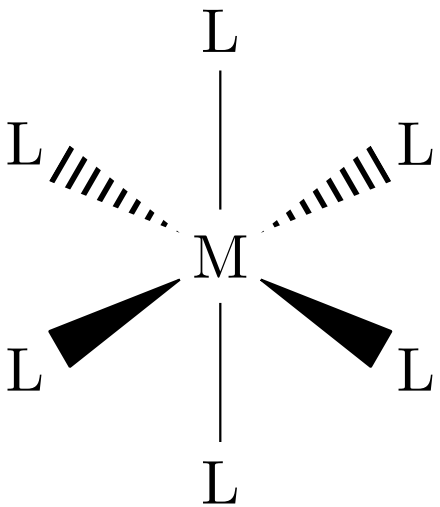


- ✓ tunable properties
- ✓ great potential for molecular devices/catalysis

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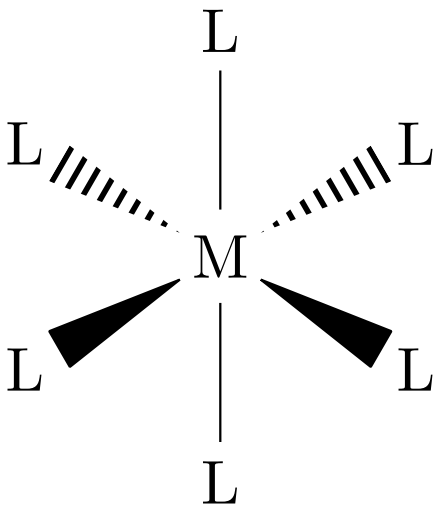


✗ multiple spin states –
**ground state unknown**

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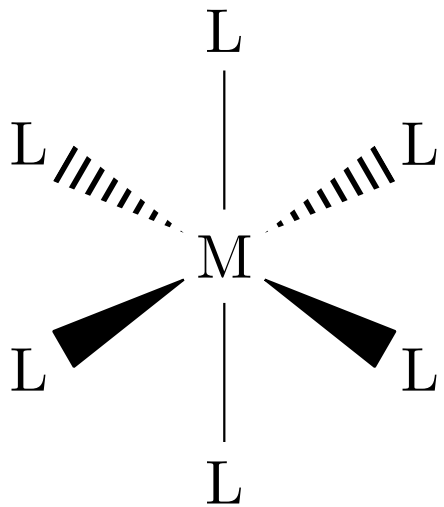


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✗ DFT calculations expensive – **need ML methods**

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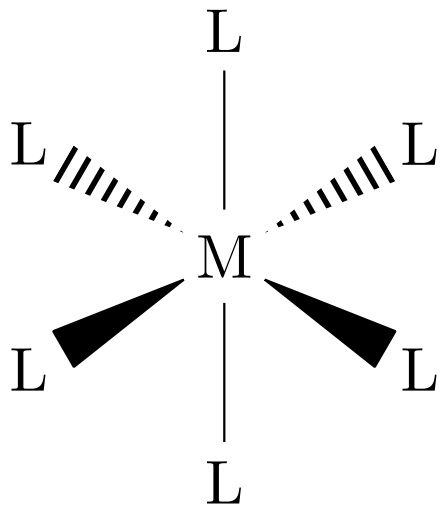


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- ✗ and not always reliable – **uncertainty in functional choice**

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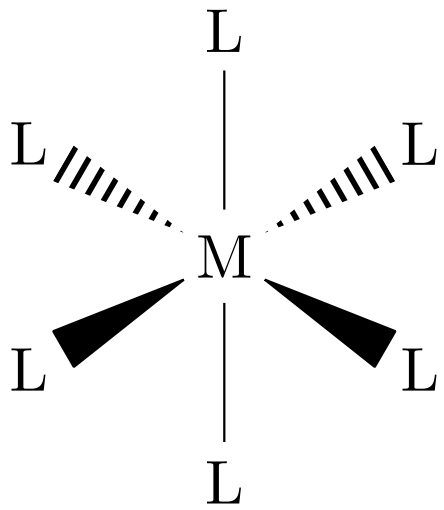
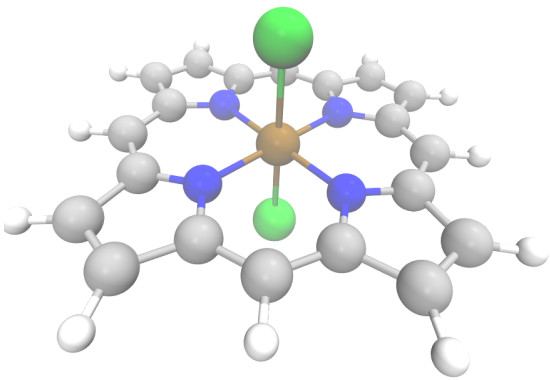



- ✗ multiple spin states – **ground state unknown**
- ✗ DFT calculations expensive – **need ML methods**
- ✗ and not always reliable – **uncertainty in functional choice**
- ✗ difficult to get geometries a priori – **starting structures difficult**

# ML for TM complexes

How can ML help?



property

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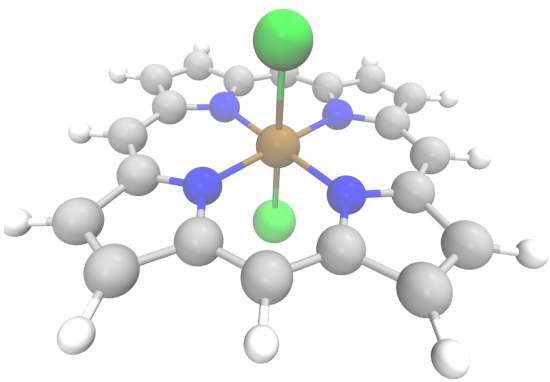

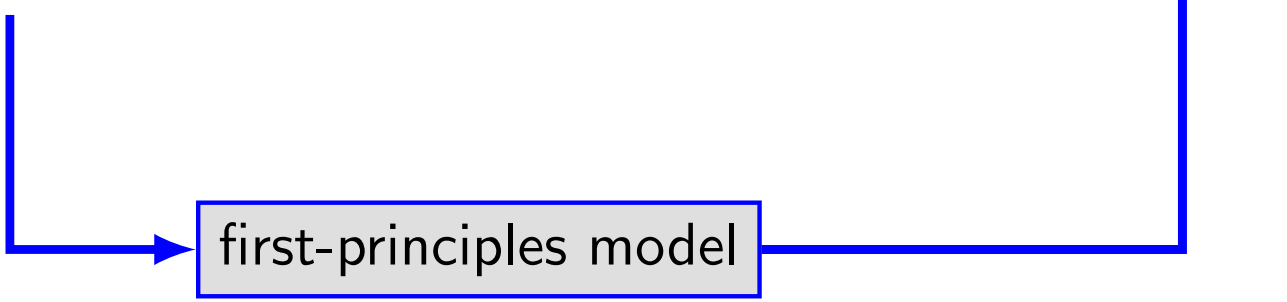

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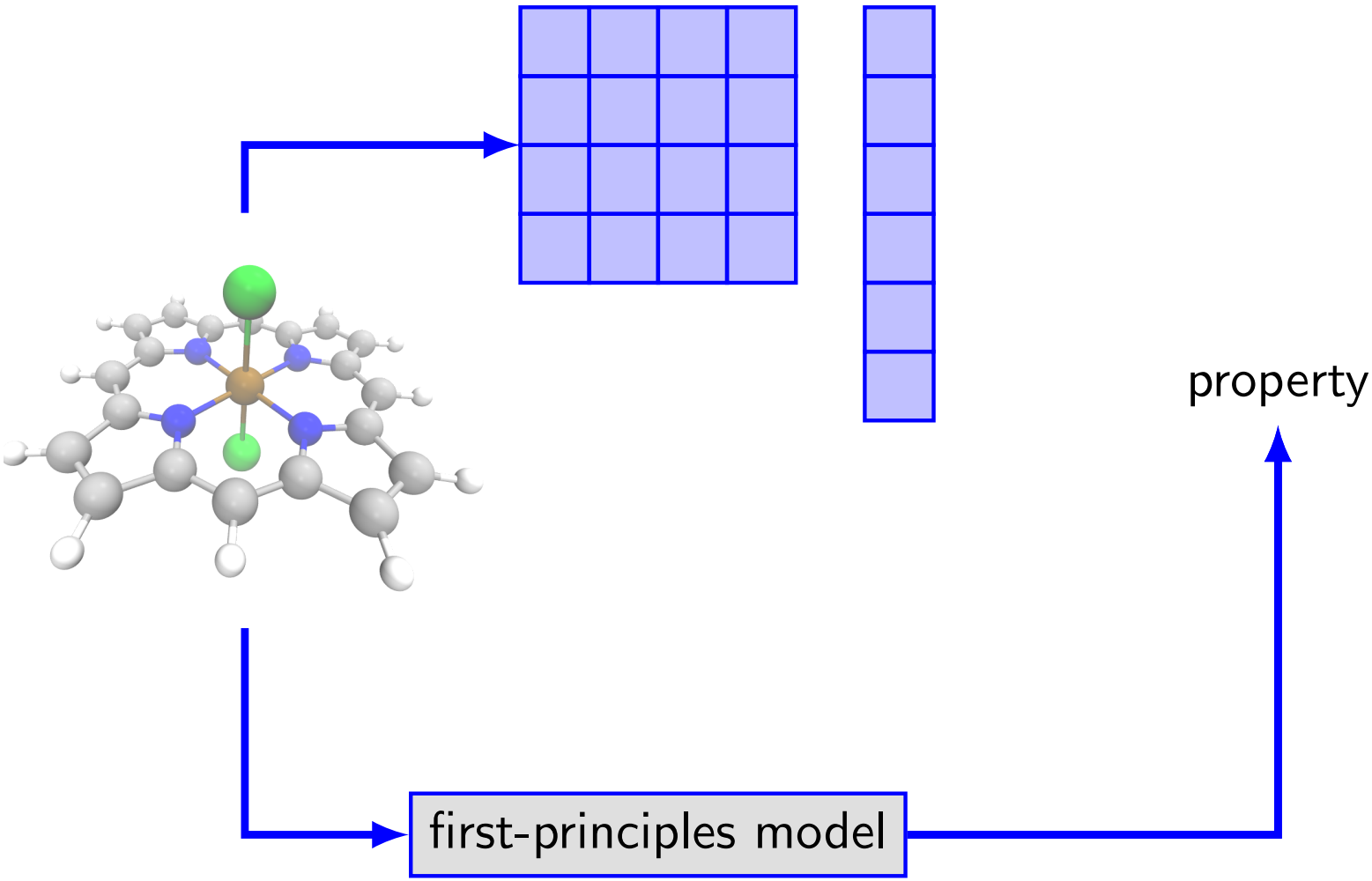

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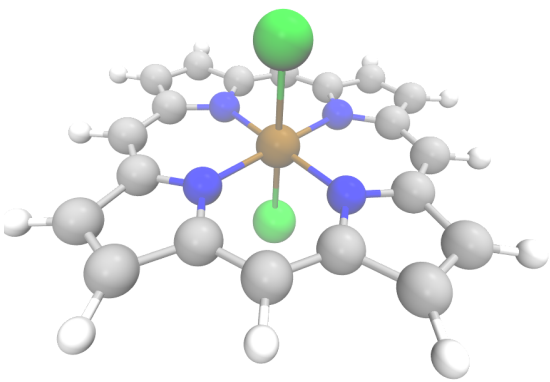

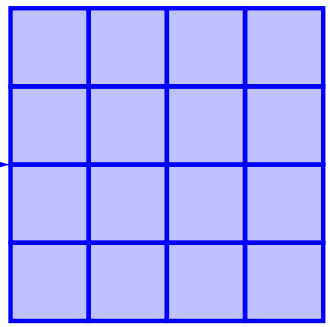

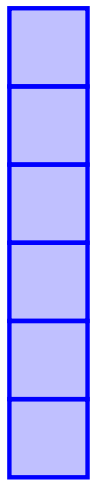

dimension? type?

property

first-principles model

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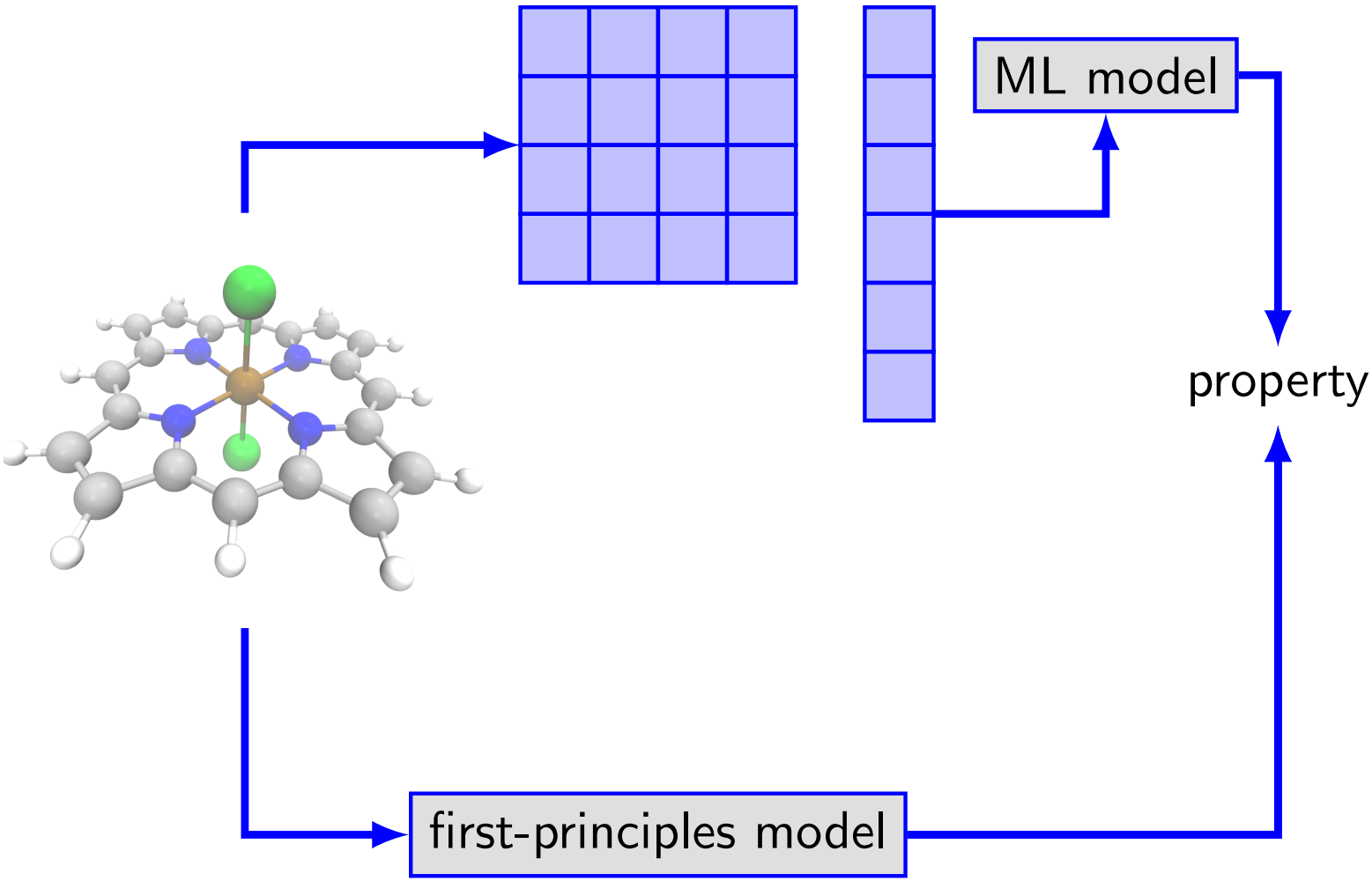

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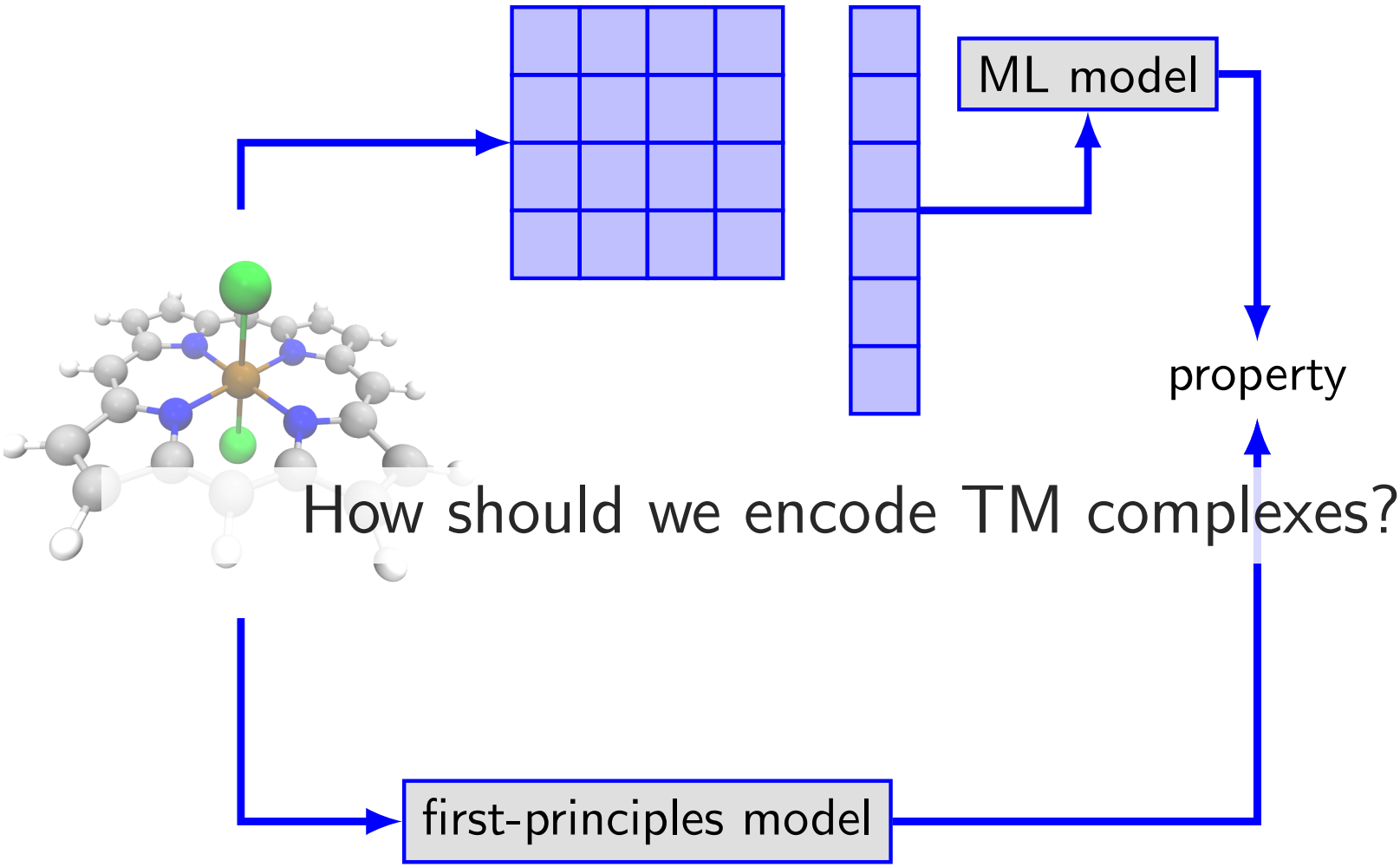
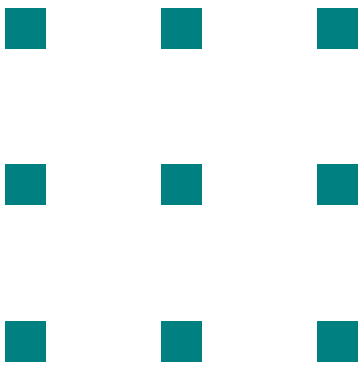


# Descriptors define chemical space



**Chemical Space** $C_f$

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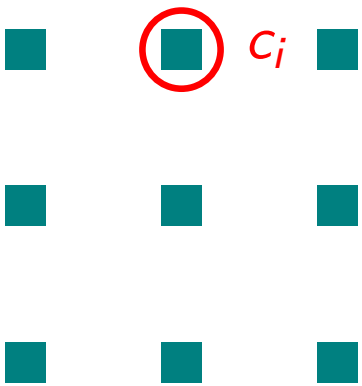


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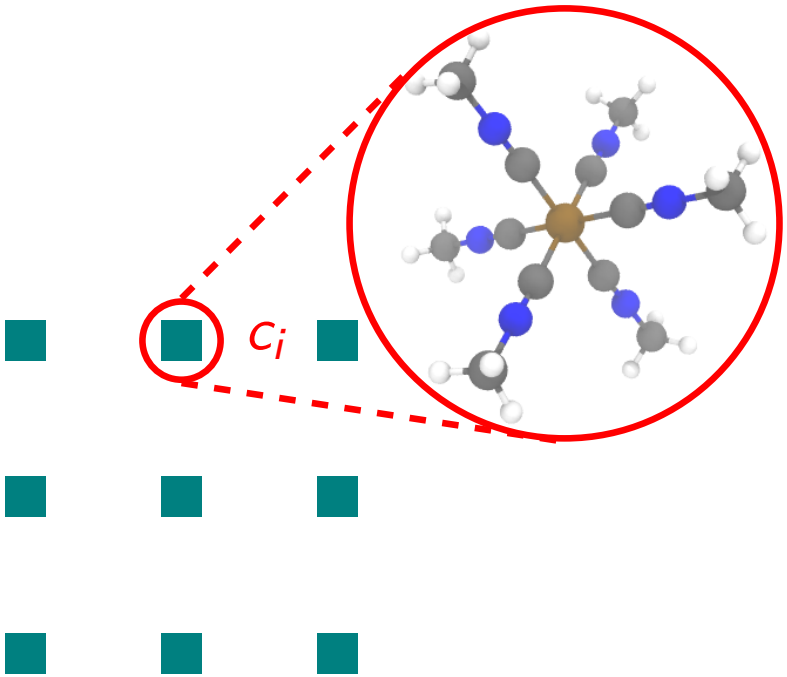


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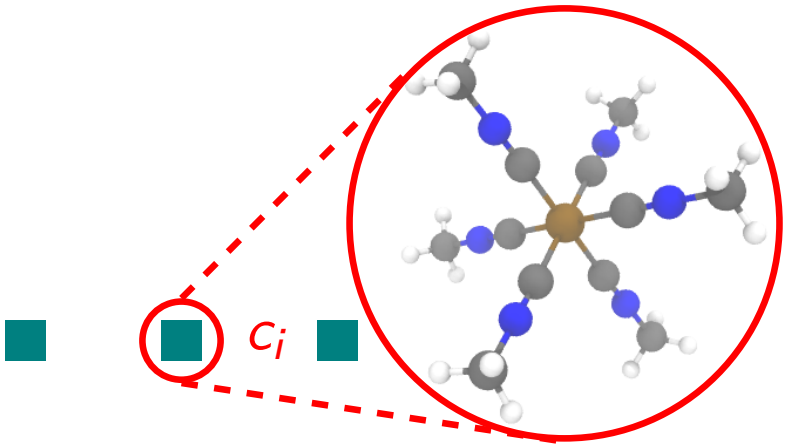


**Chemical Space** $C_f$

**Descriptor Space** $\mathcal{X} \subset \mathbb{R}^d$

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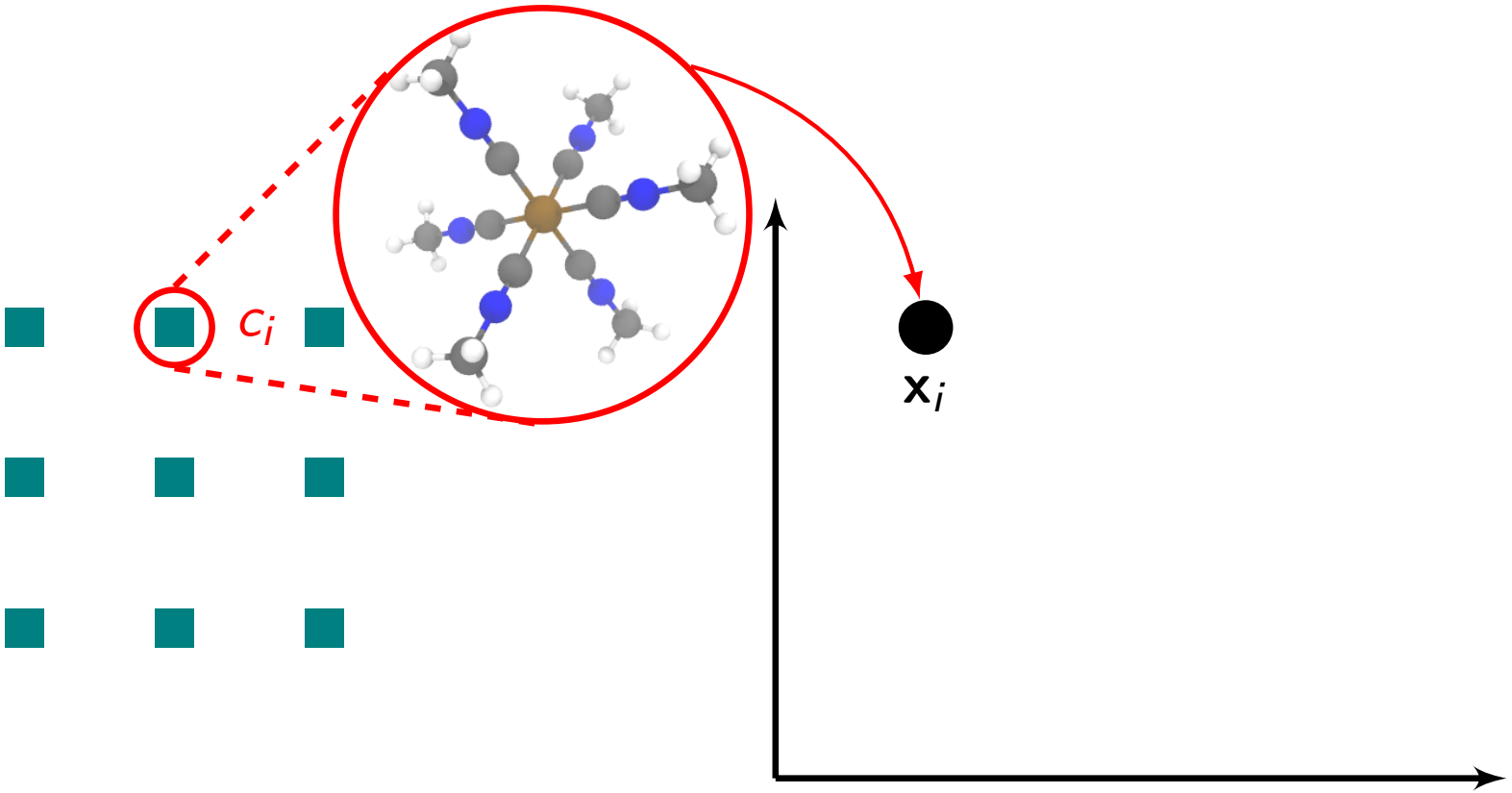


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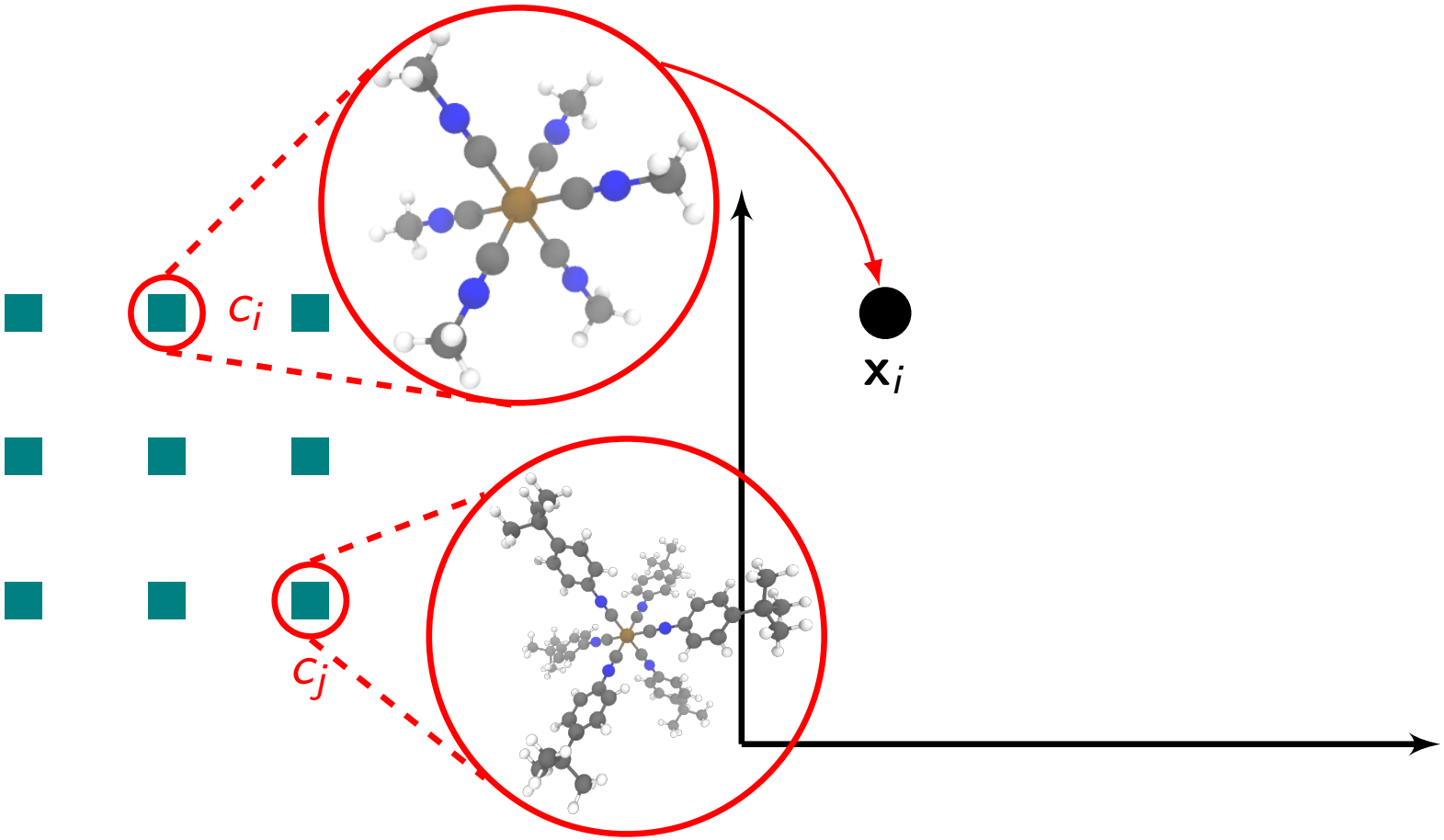


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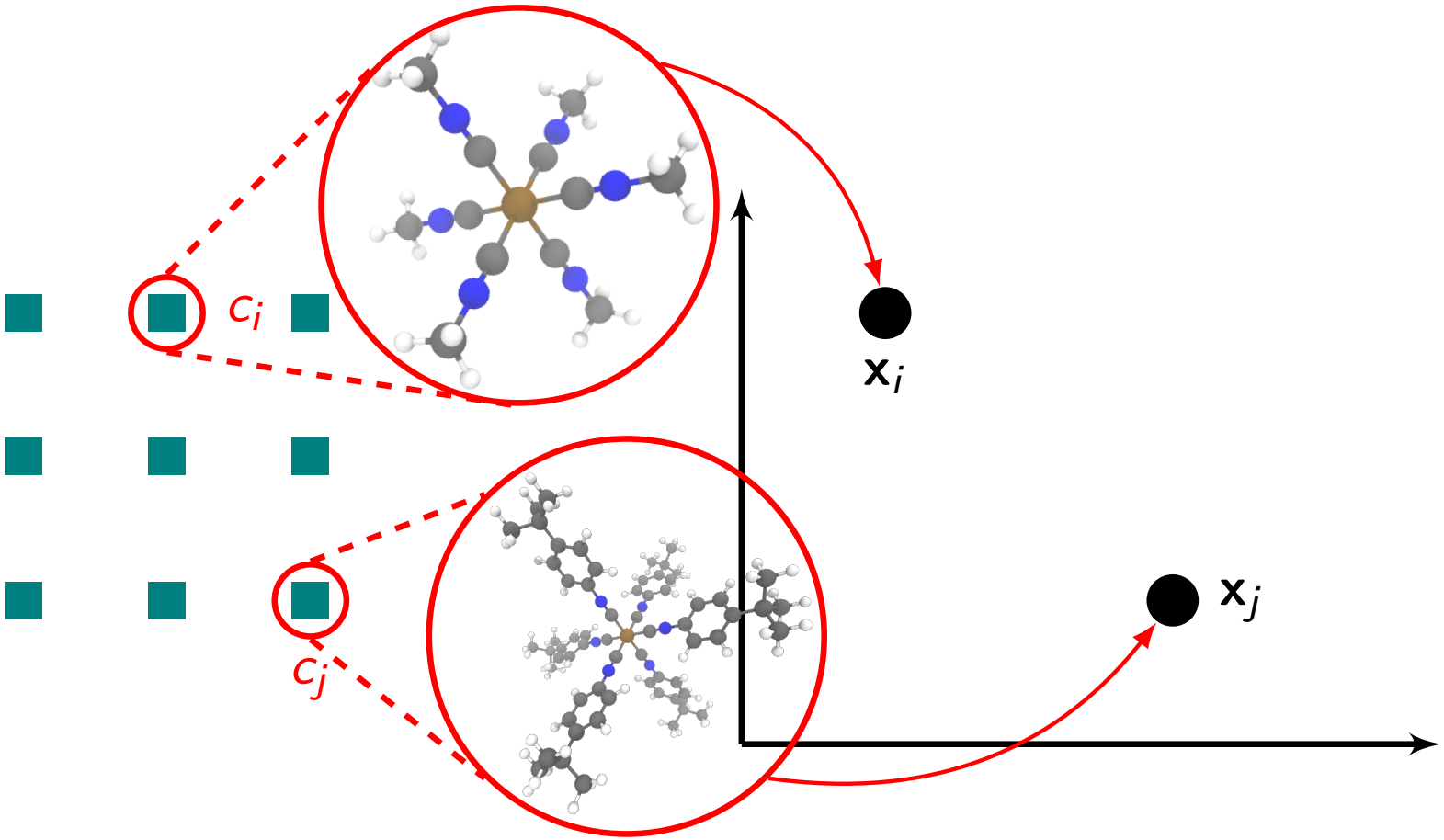


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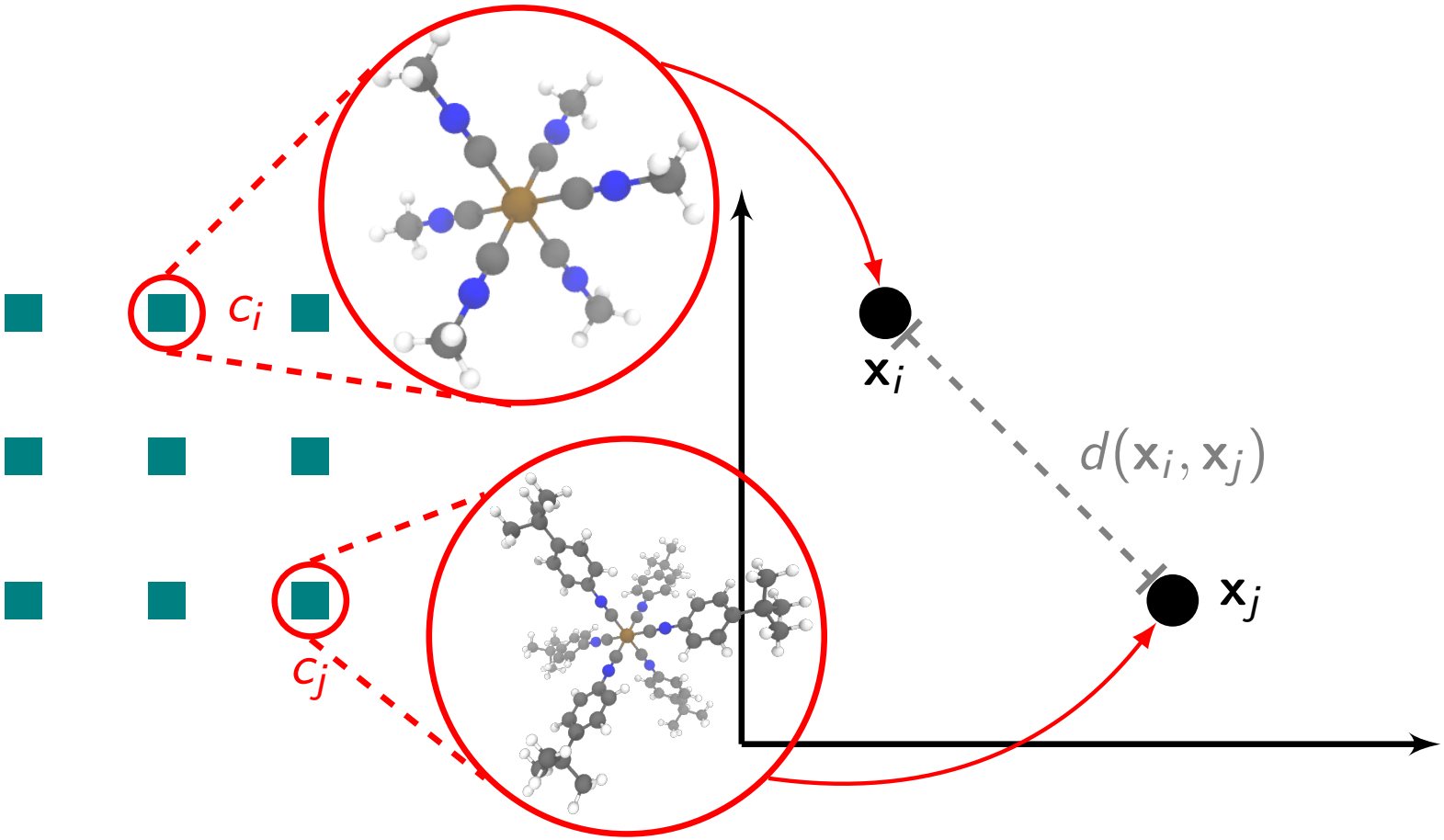


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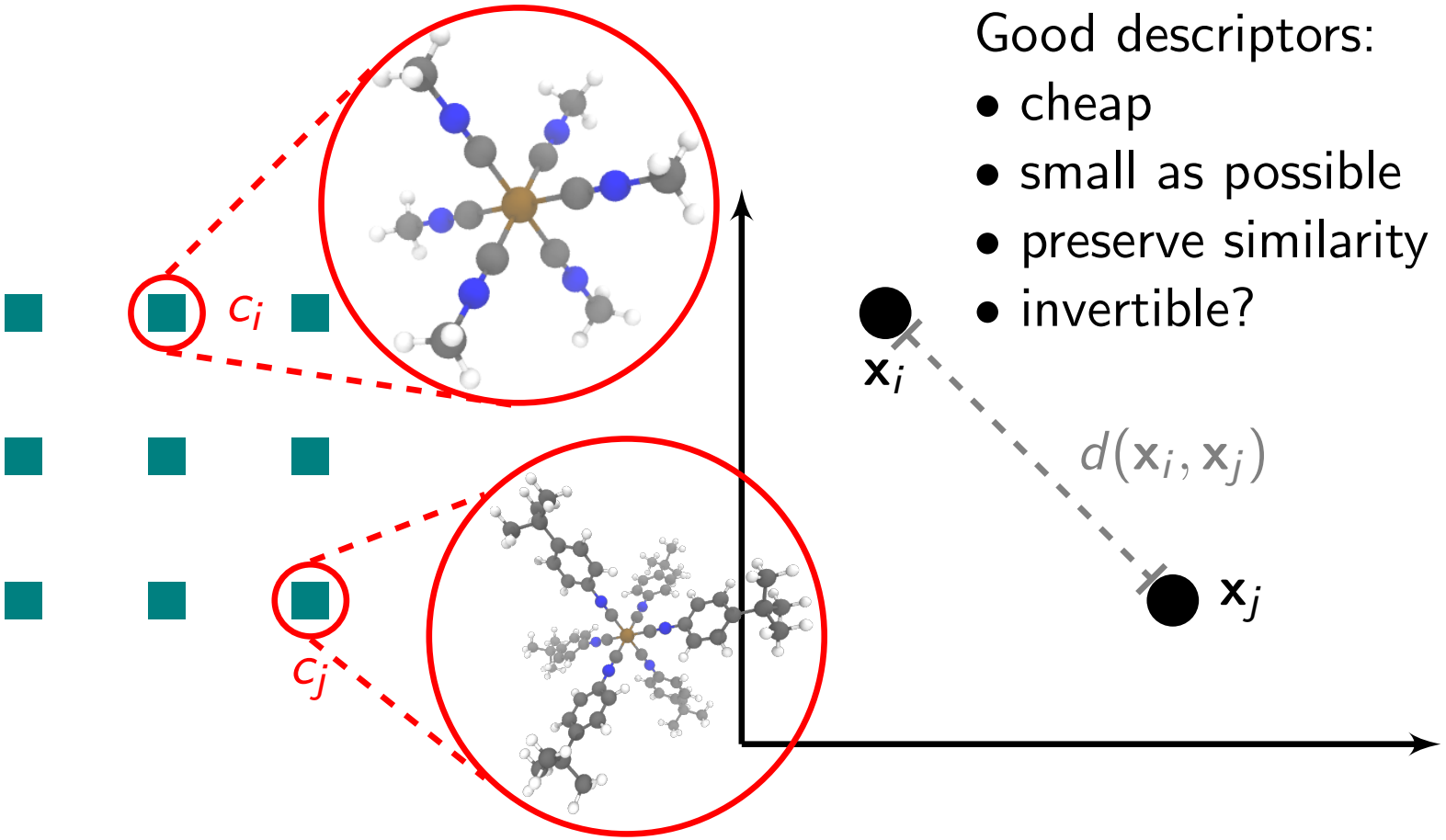



Good descriptors:

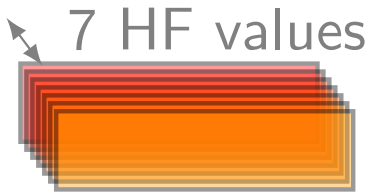
- cheap
- small as possible
- preserve similarity
- invertible?

**Chemical Space** $C_f$

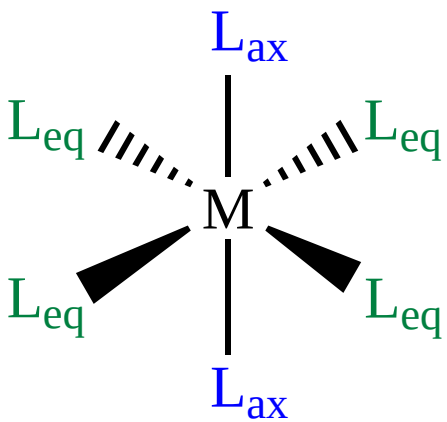
**Descriptor Space** $\mathcal{X} \subset \mathbb{R}^d$

# Predictive modeling

Data for octahedral complexes[1]:




1345 (194) complexes




[1] Janet, J.P., and Kulik, H.J. *Chemical Science*, 2017, 8, 5137-5152.

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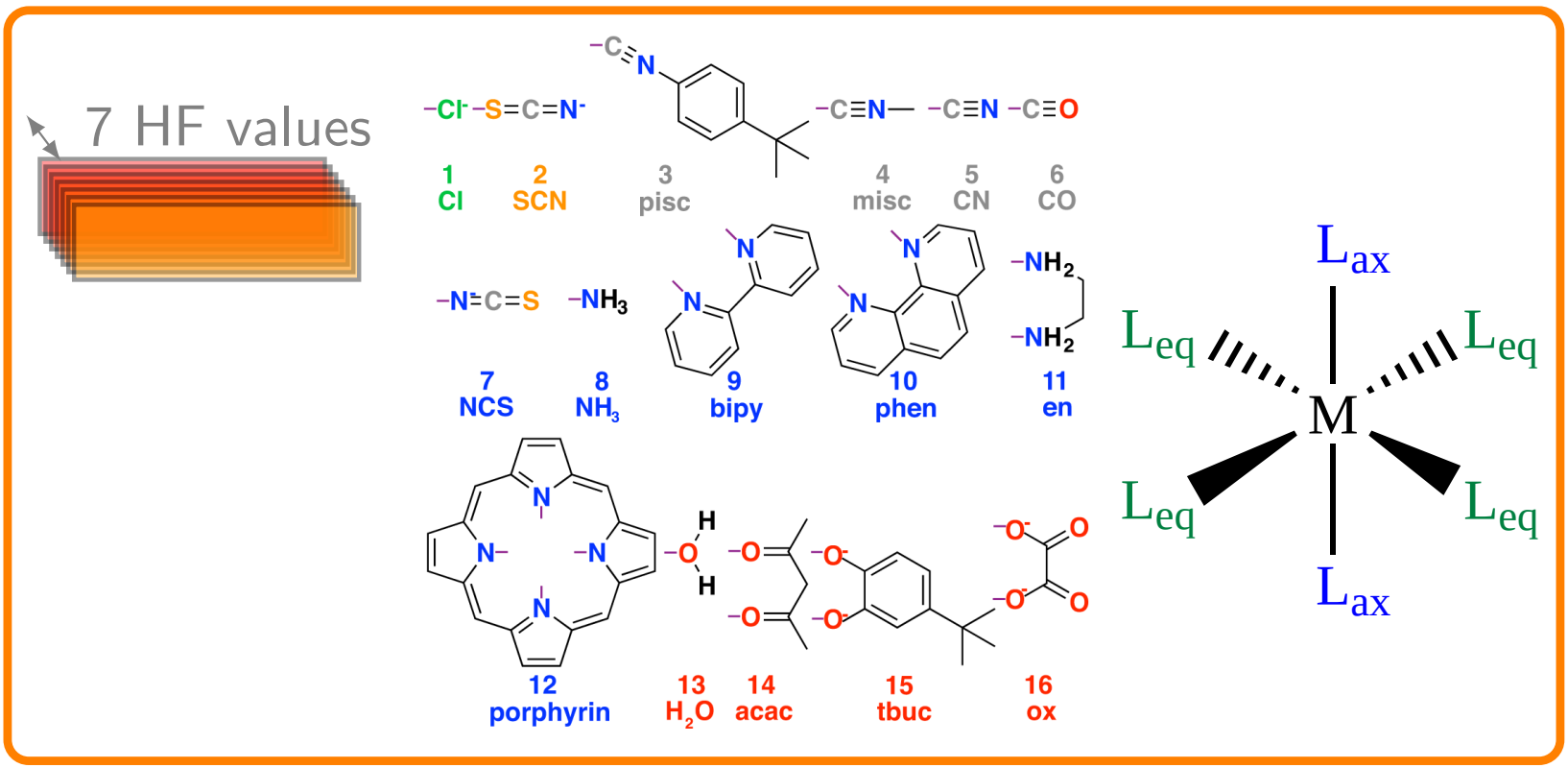


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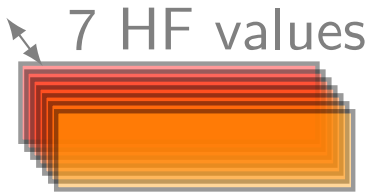


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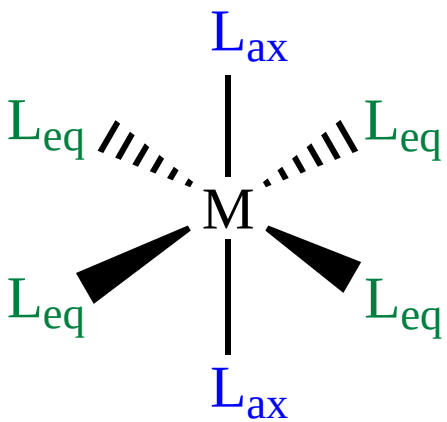


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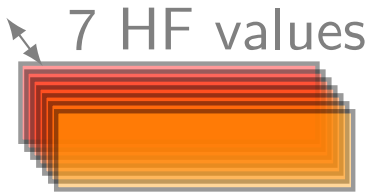
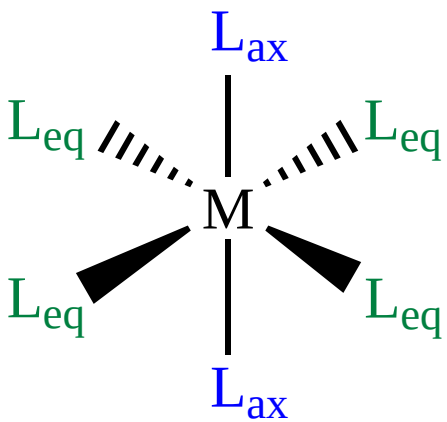



1345 (194) complexes

B3LYP-like DFT
HF exchange in 0-30%
gas phase optimizaton
LANL2DZ/6-31G*
high- and low-spin
M(II)/(III)

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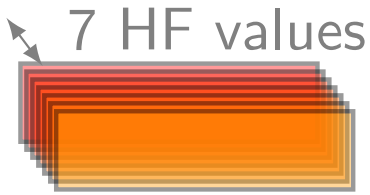


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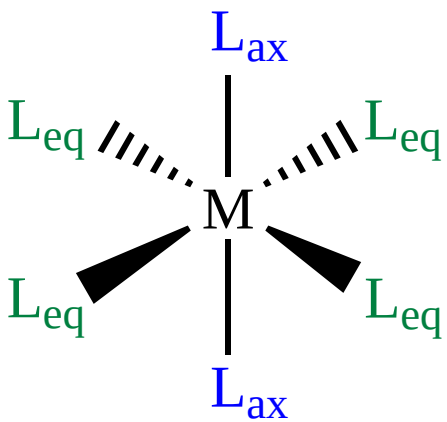
built by molSimplify

`molsimplify.mit.edu`

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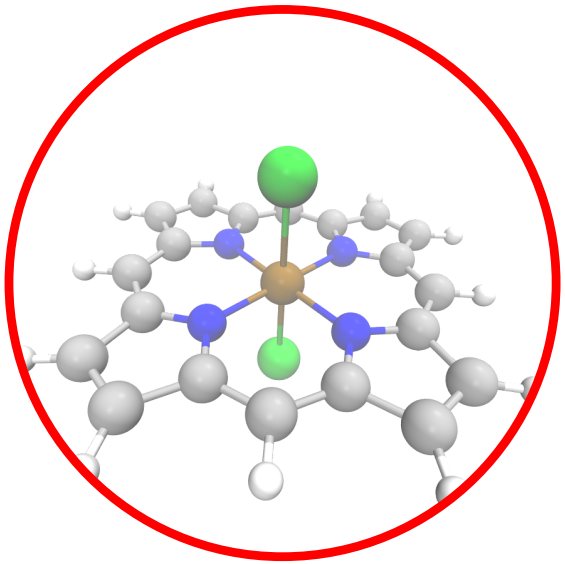

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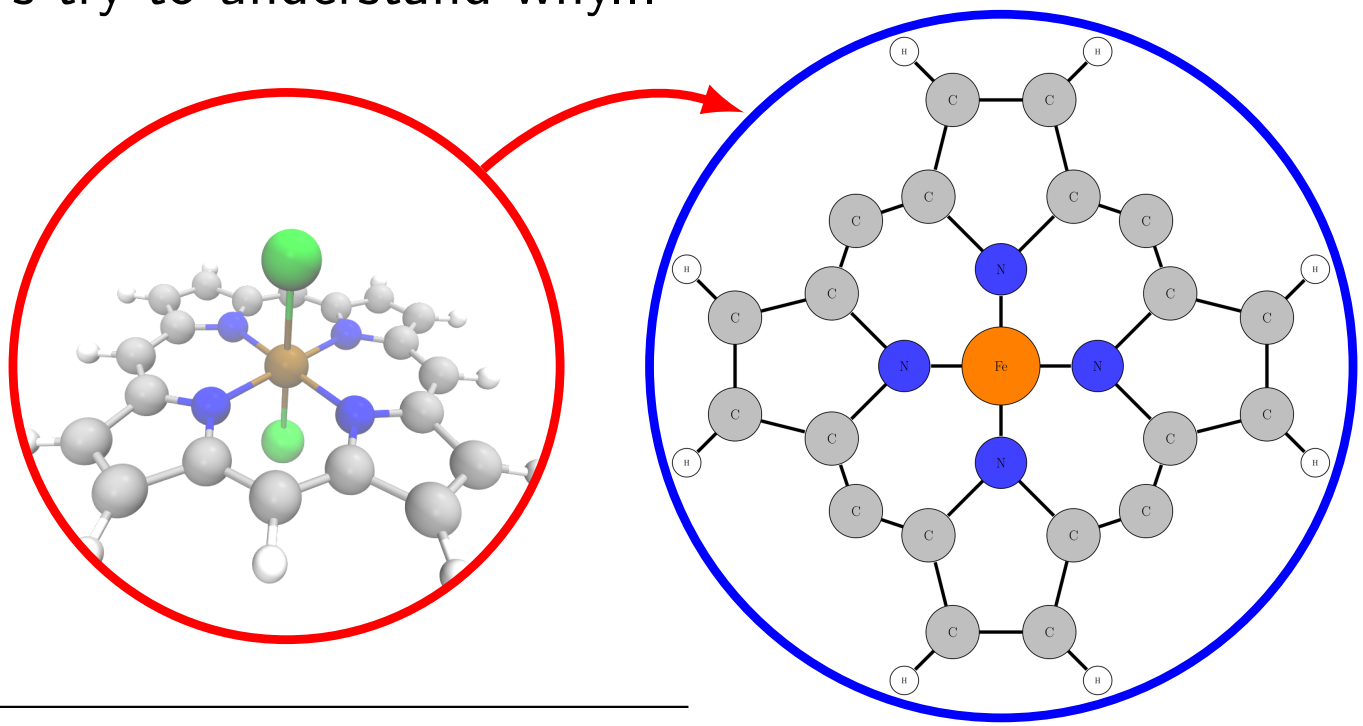


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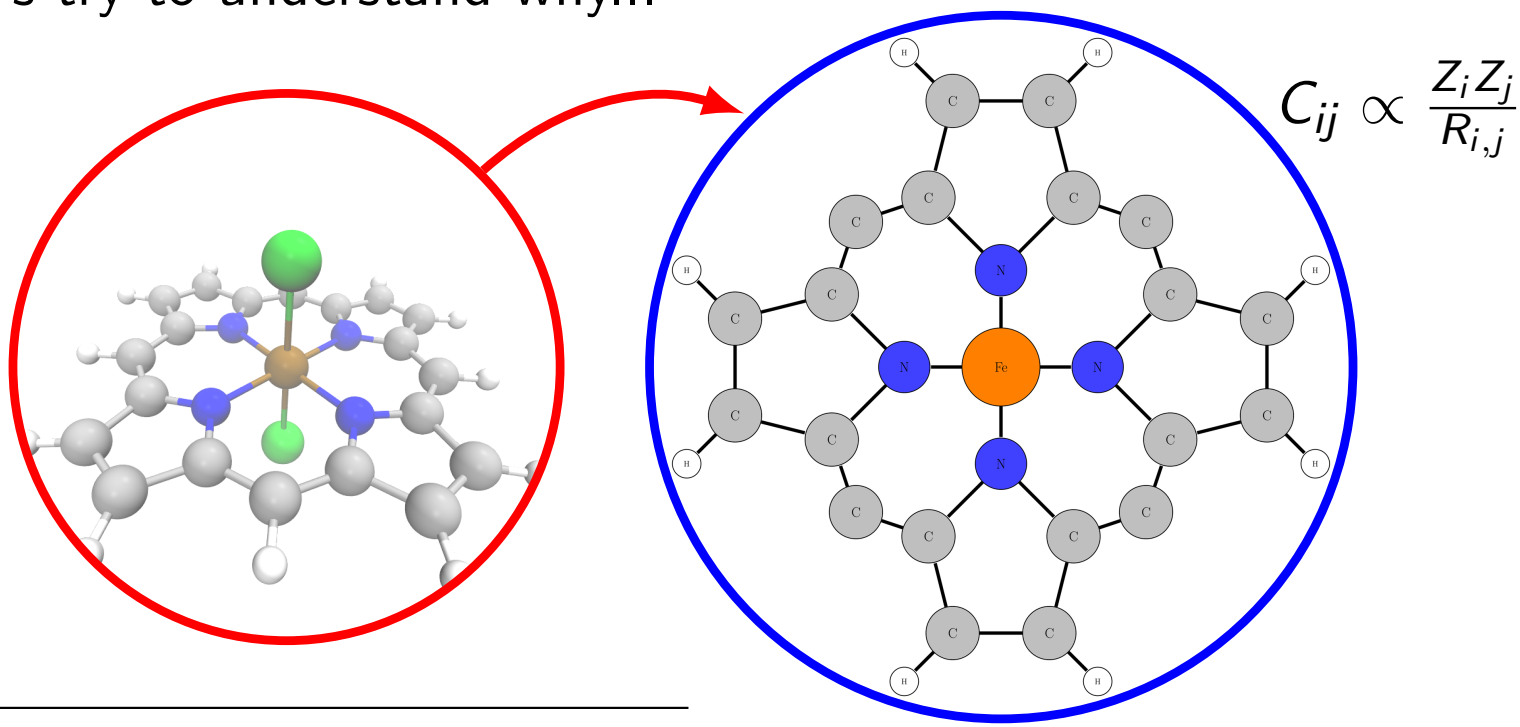


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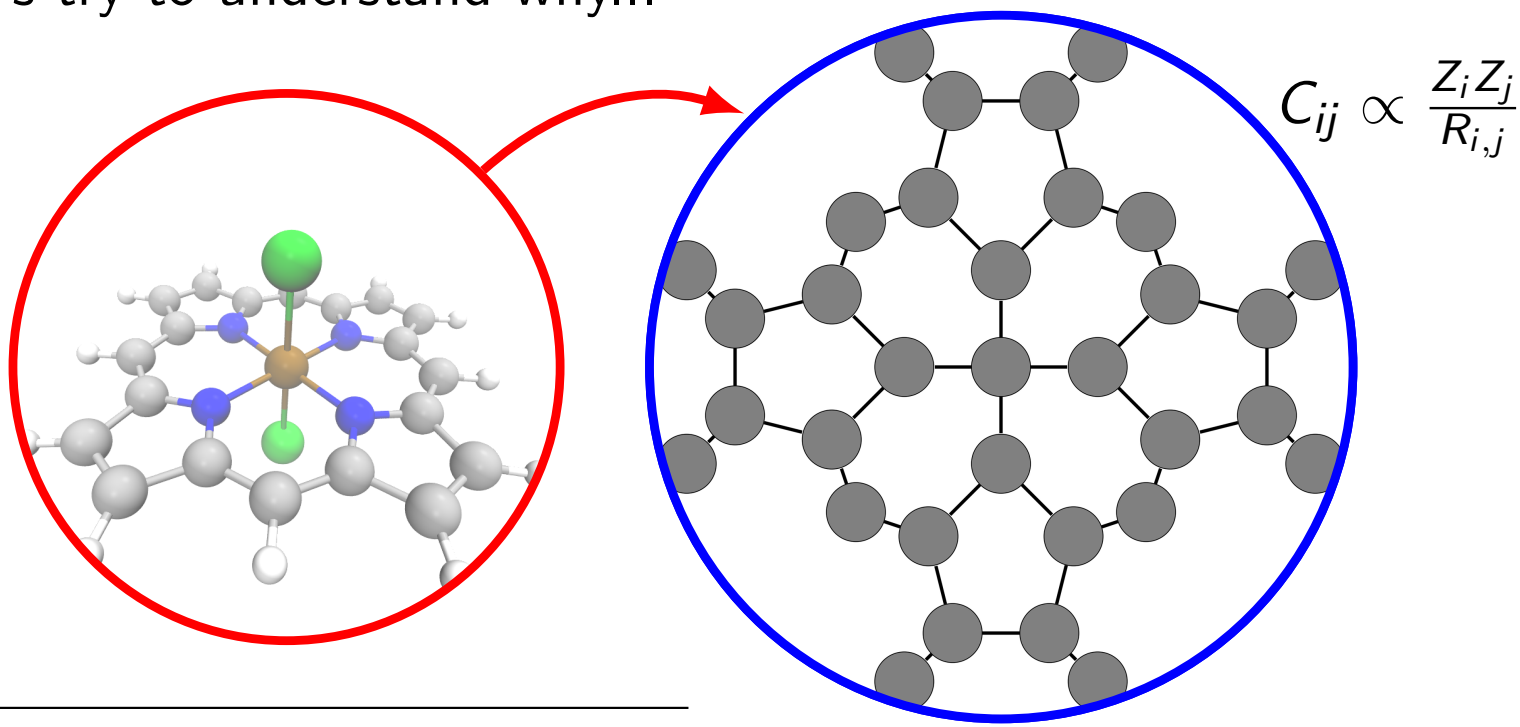


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# A tale of two complexes

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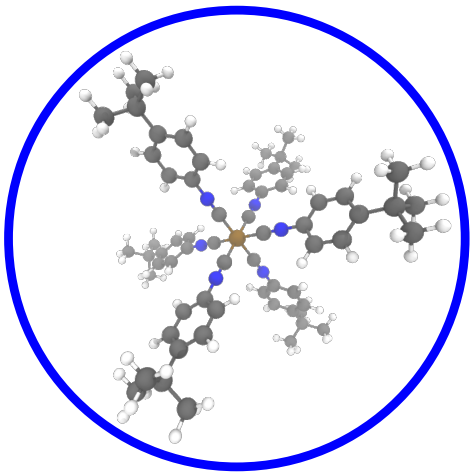

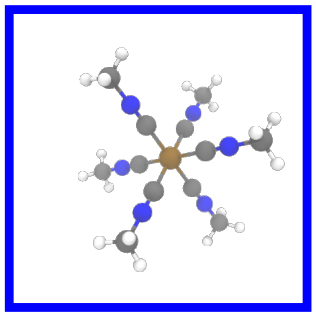

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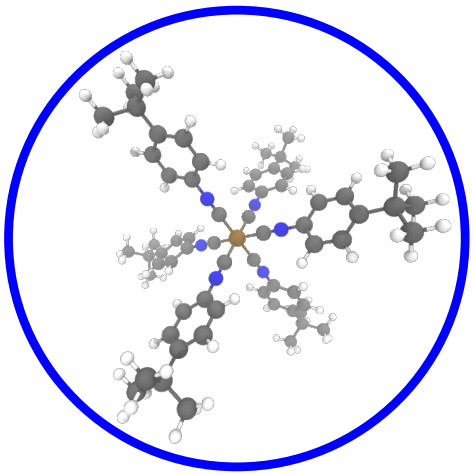
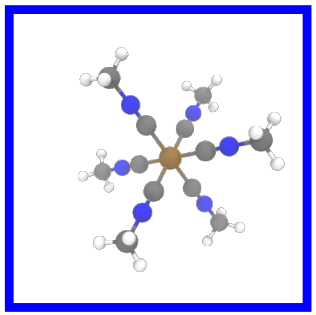
$Fe[pisc]_6^{3+}$   $Fe[misc]_6^{3+}$

$\Delta E_{H\text{-}L} = 37.7$ kcal/mol $\Delta E_{H\text{-}L} = 40.7$ kcal/mol

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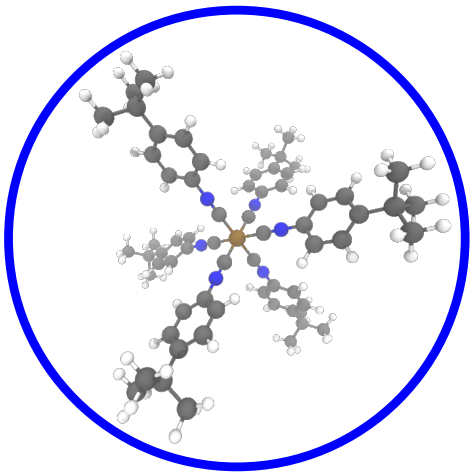

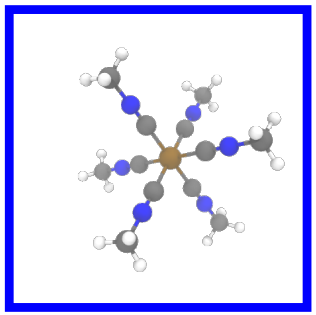

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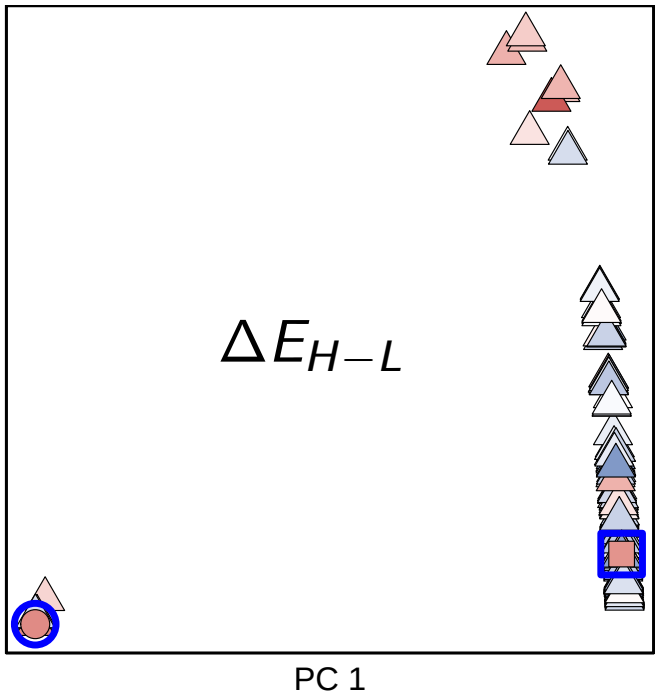


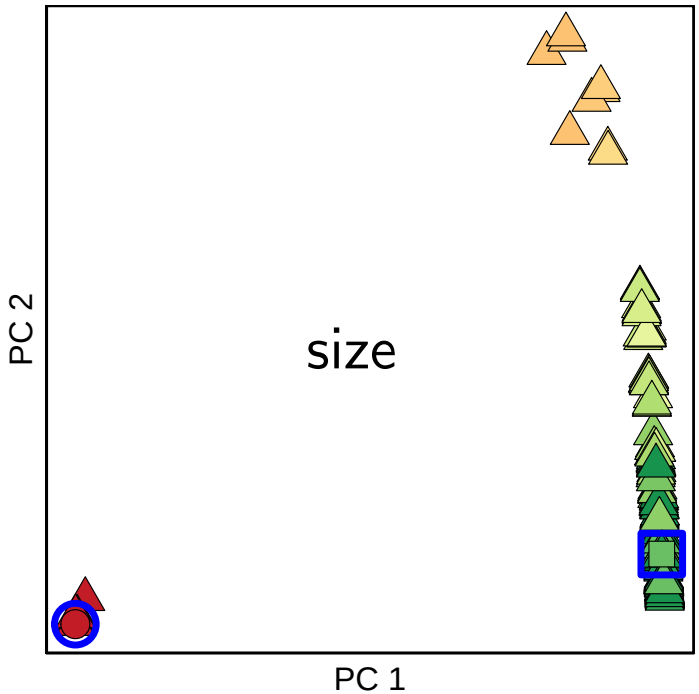

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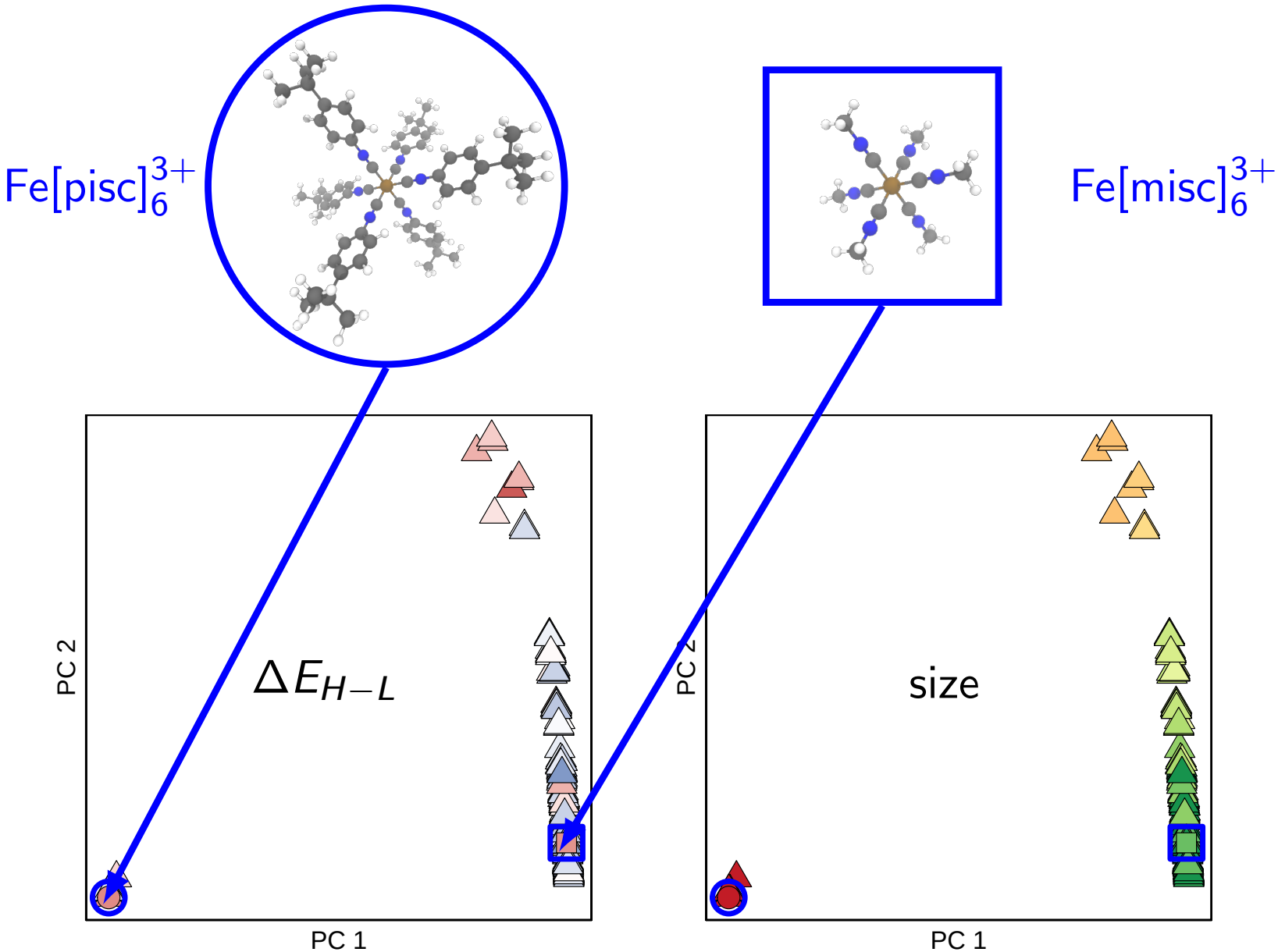

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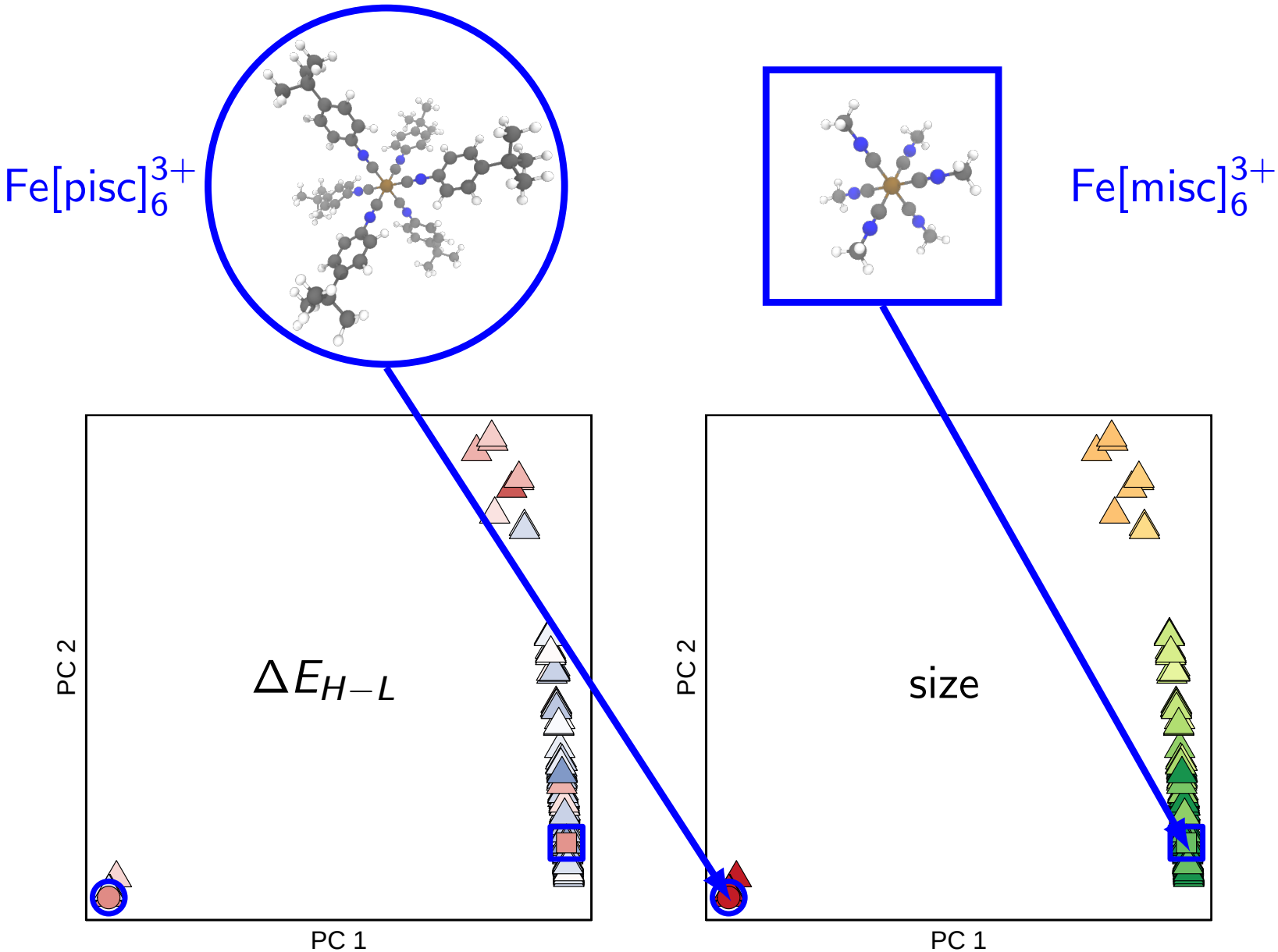

# Describing TM complexes

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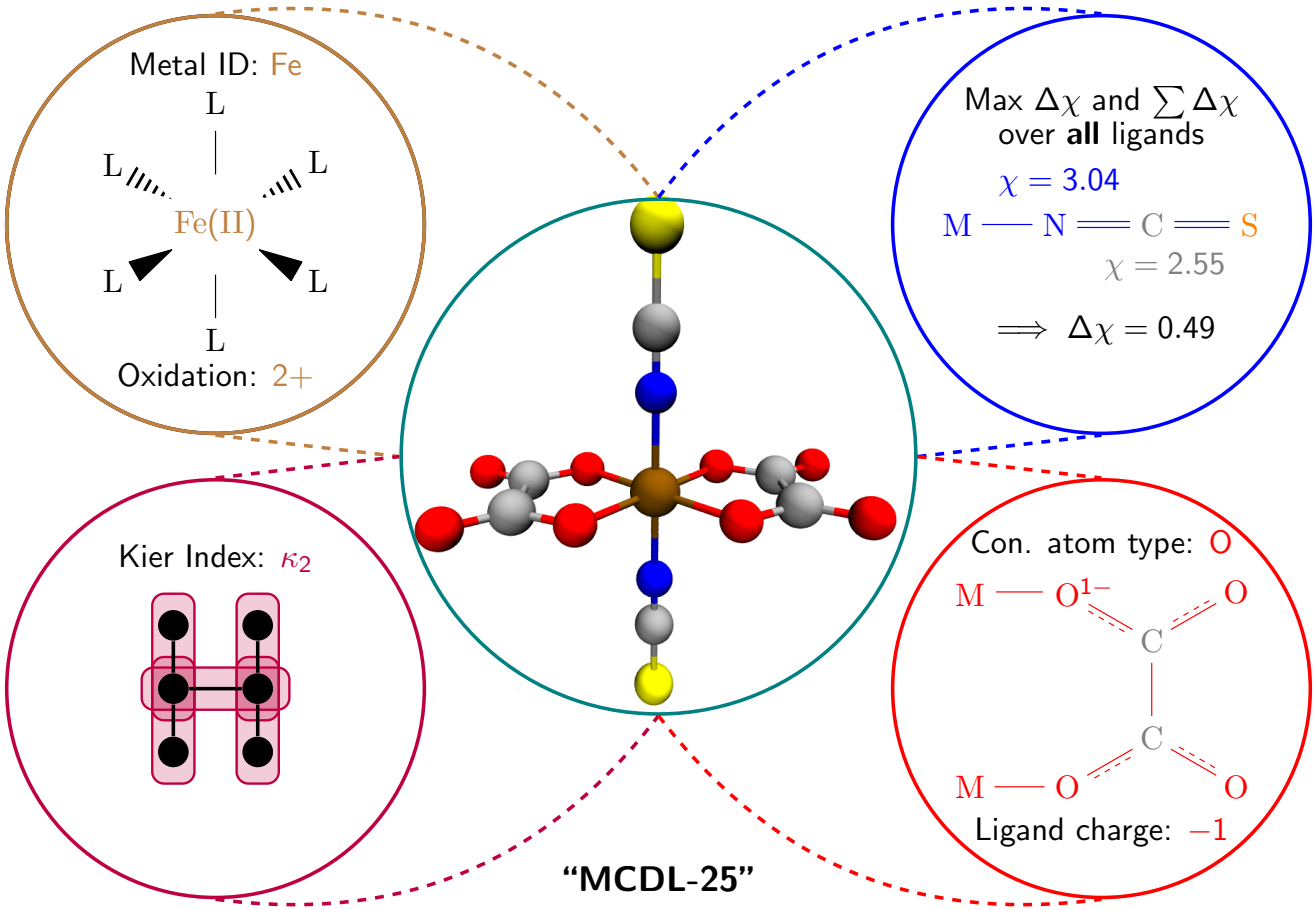


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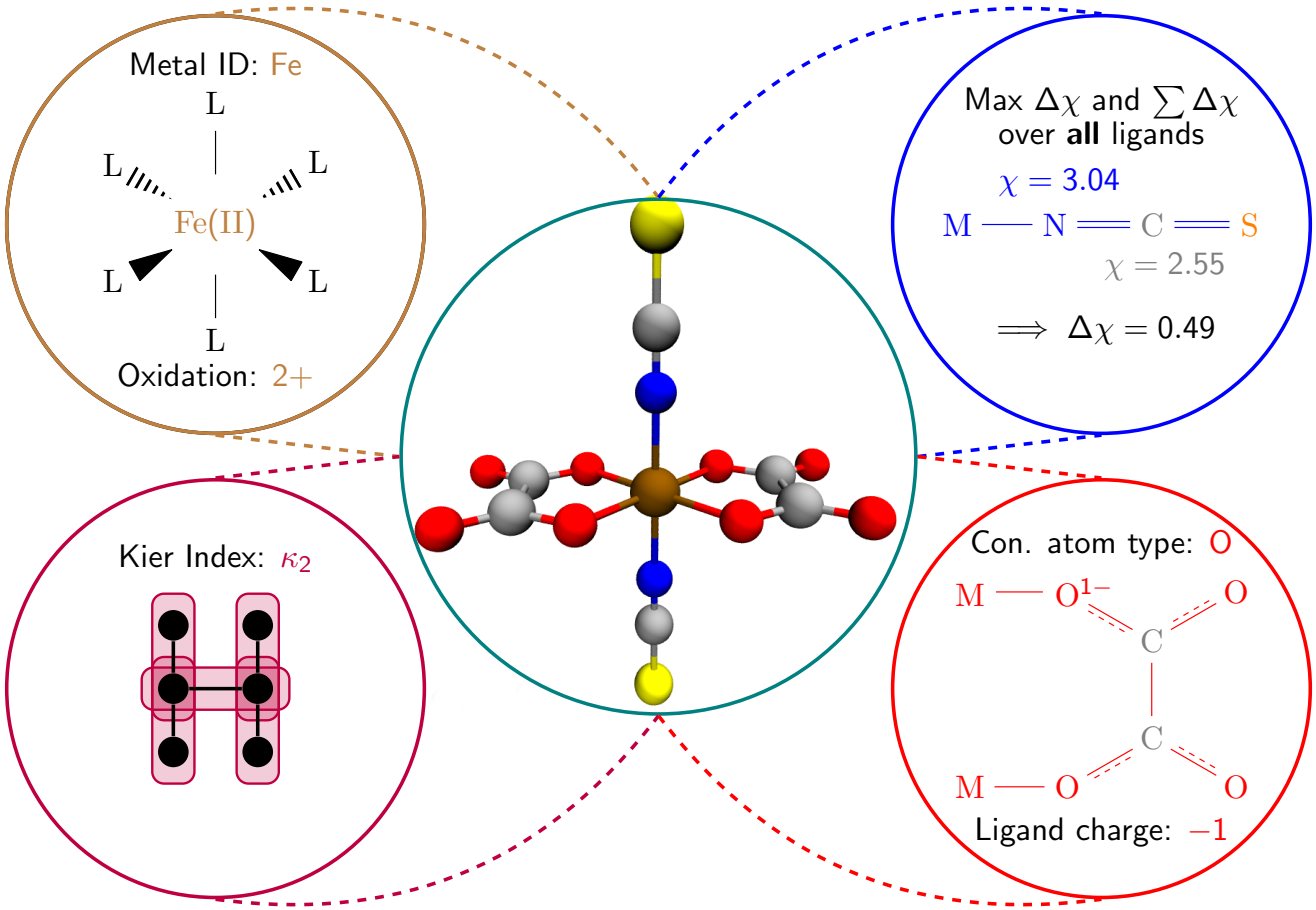
## MCDL: First shell effects are emphasized!



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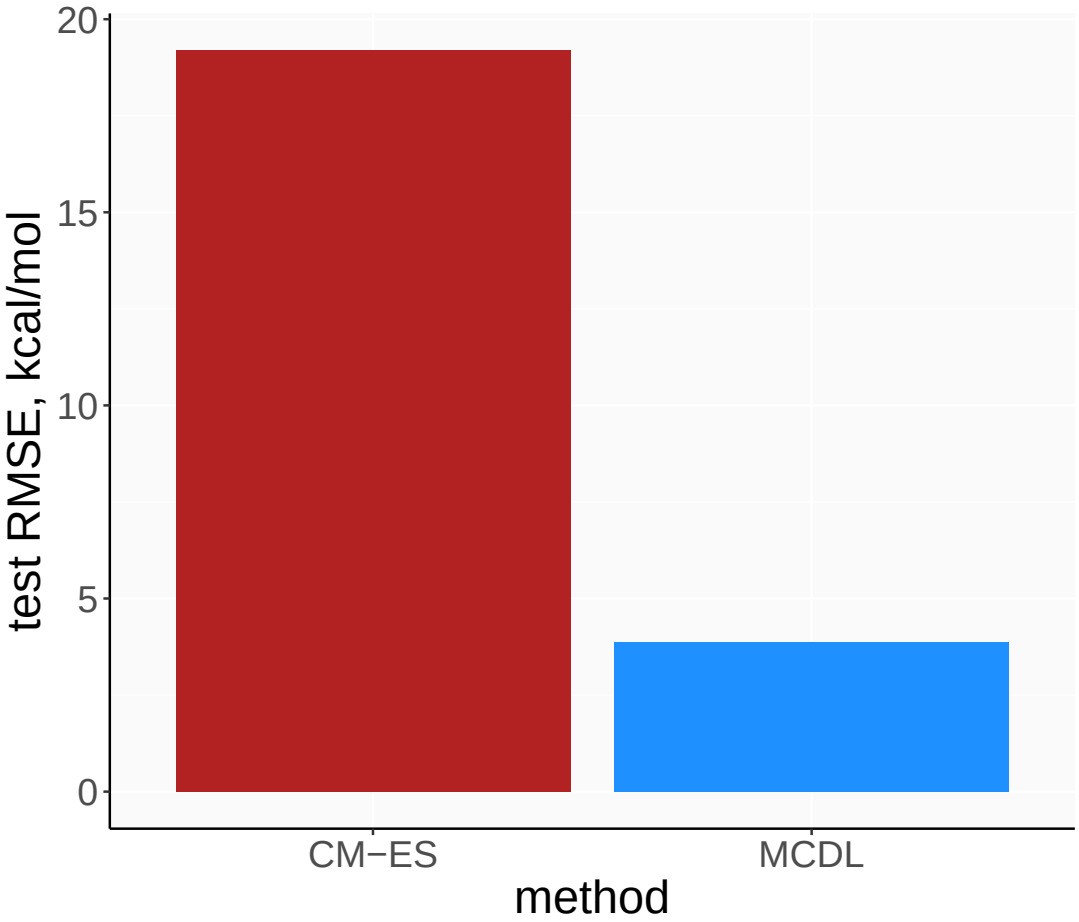
Greatly improves KRR performance:



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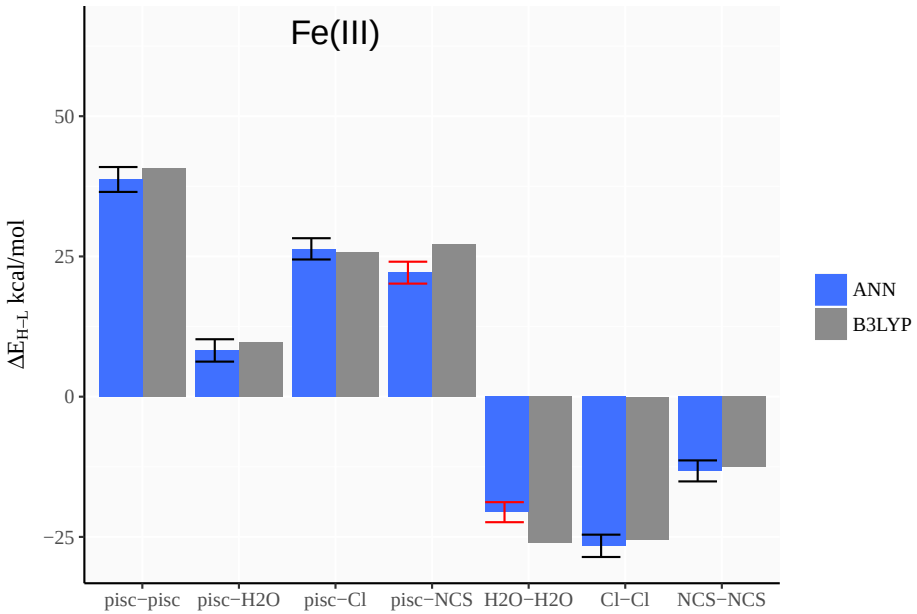


heteroleptic fields

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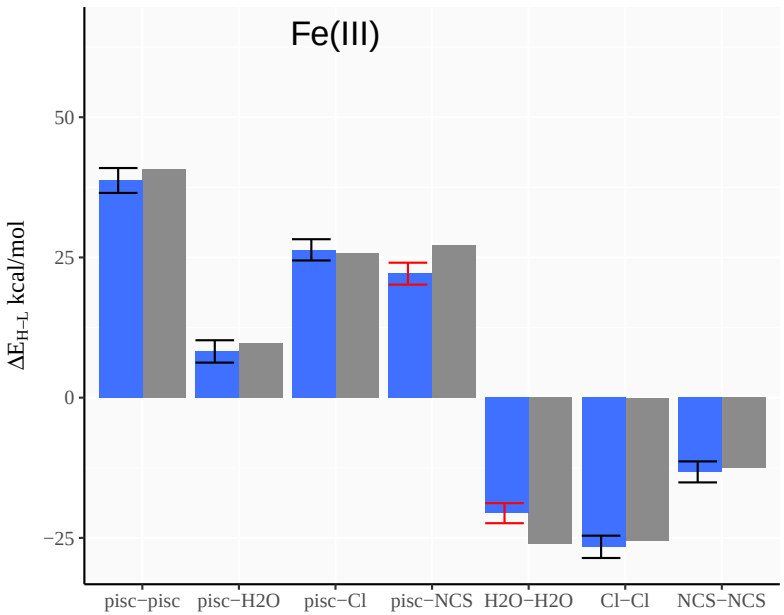


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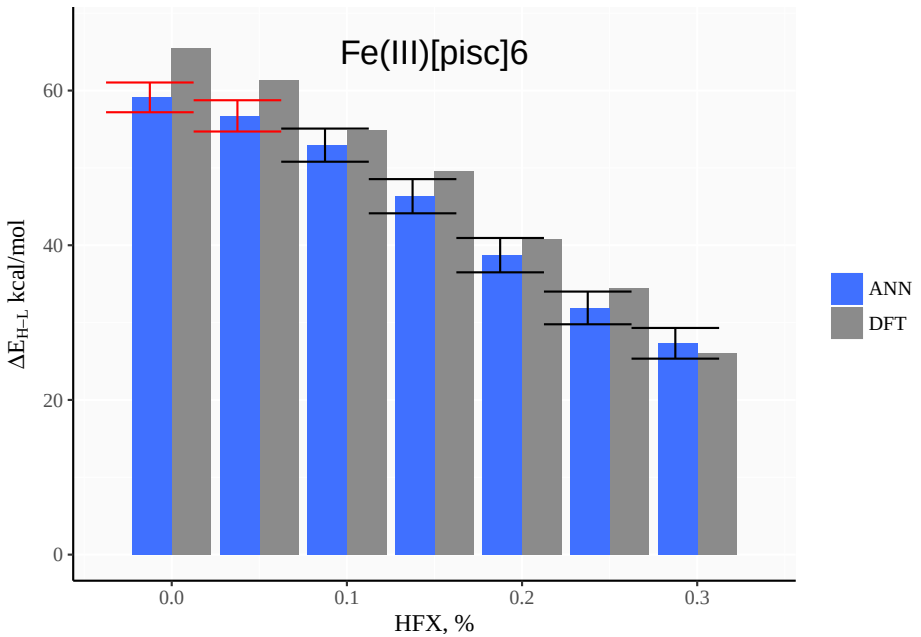


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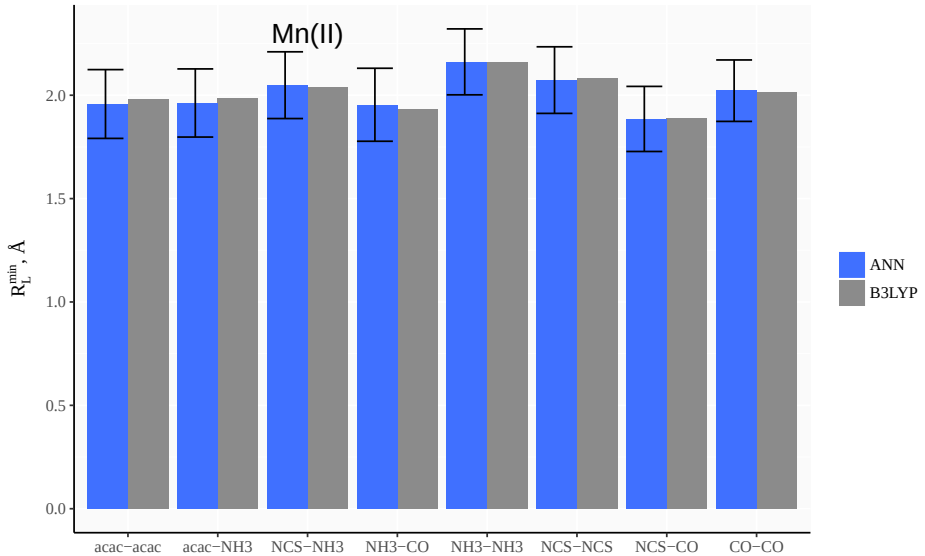


bond lenghts in a **spin** and **oxidation state** dependent manner

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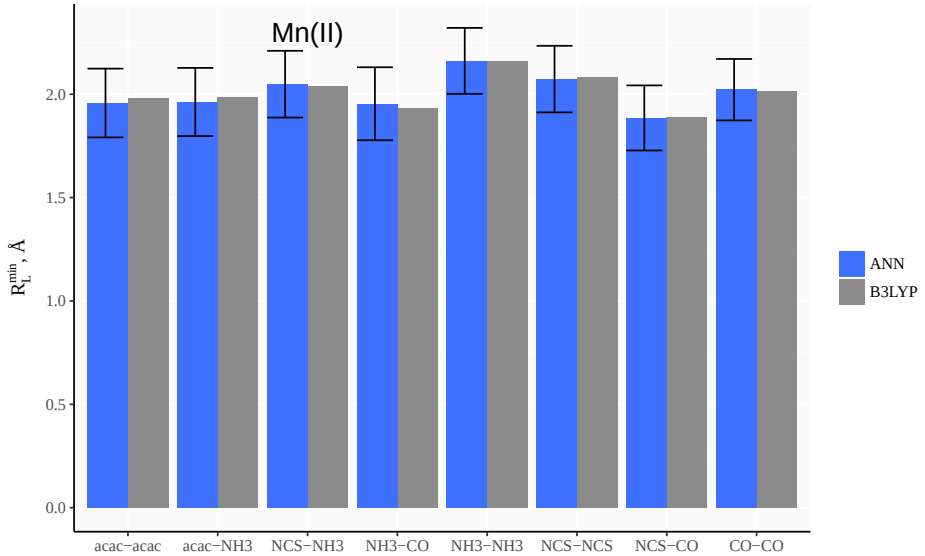


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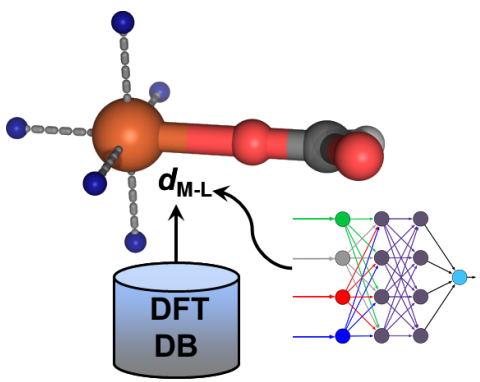


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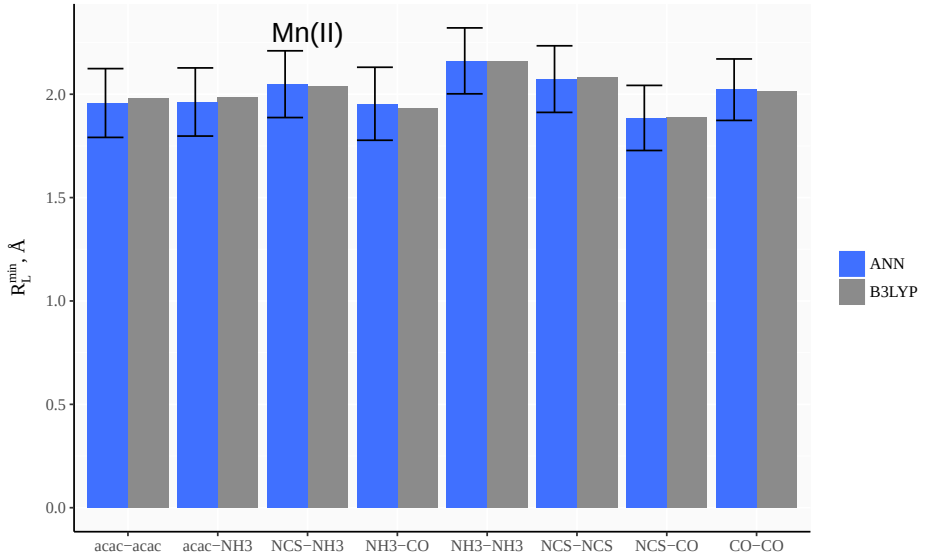


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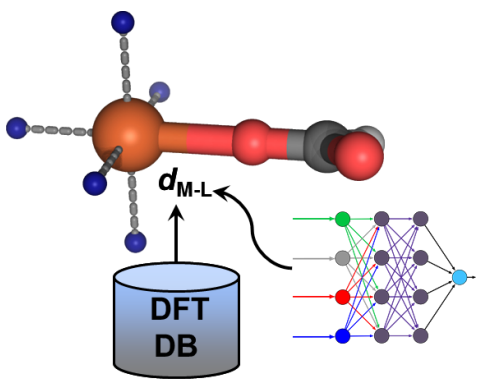


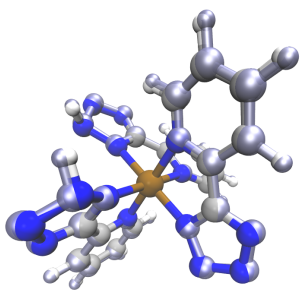

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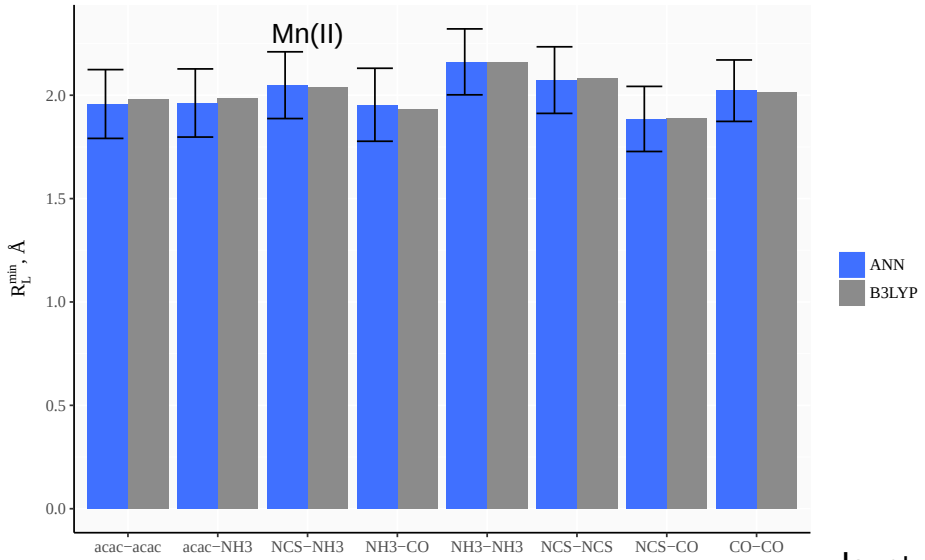


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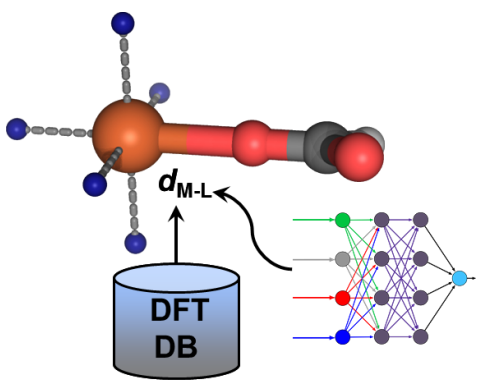


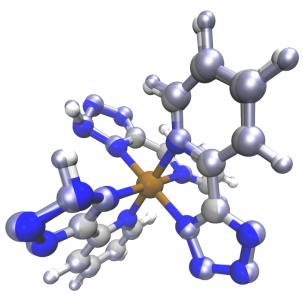

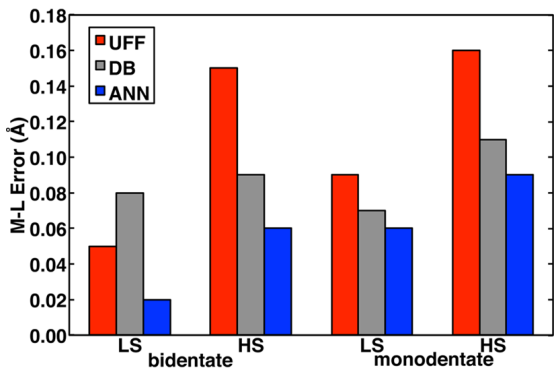


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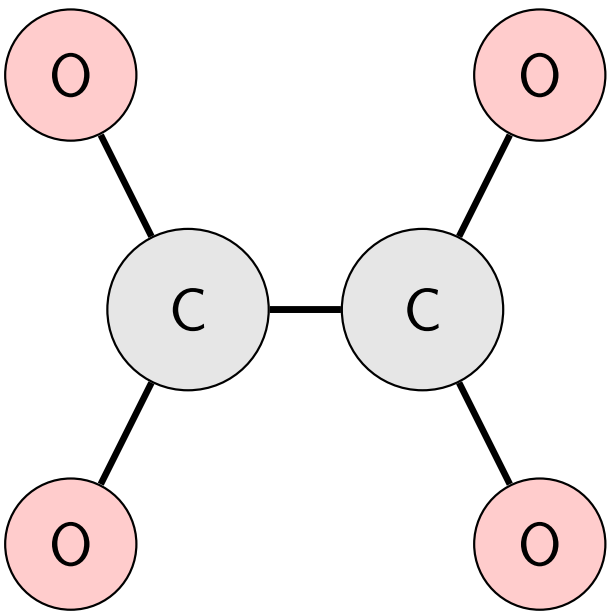


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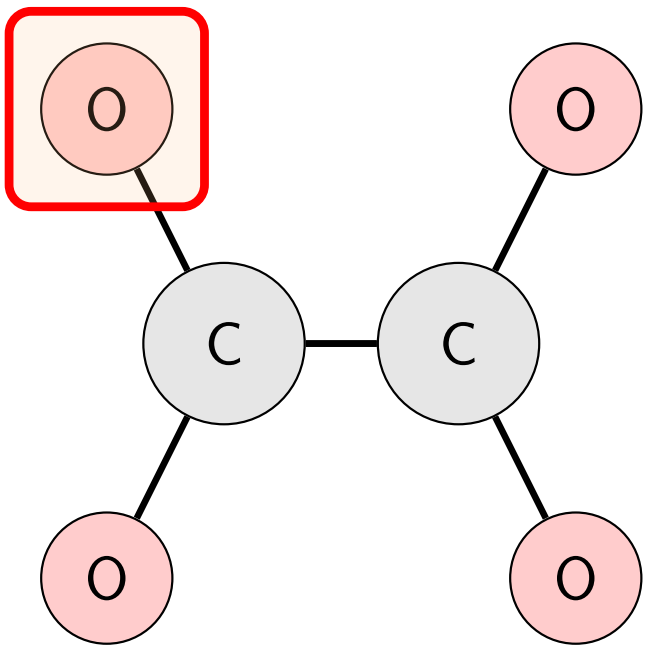


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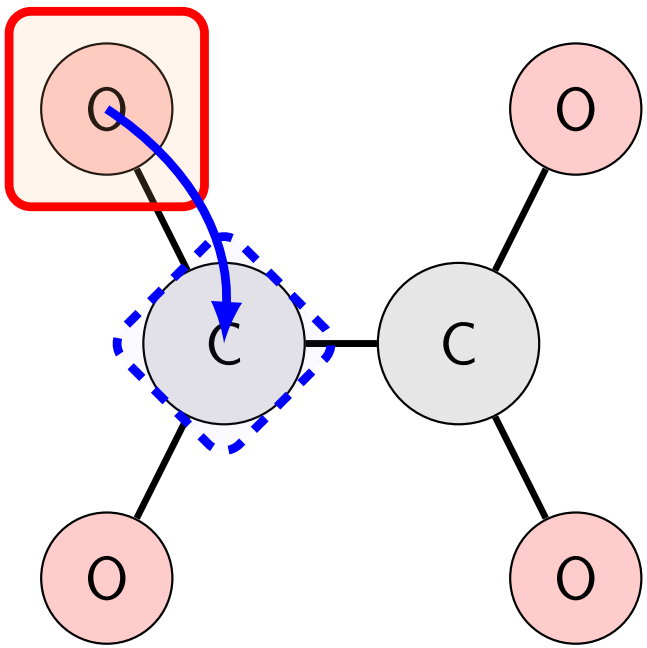


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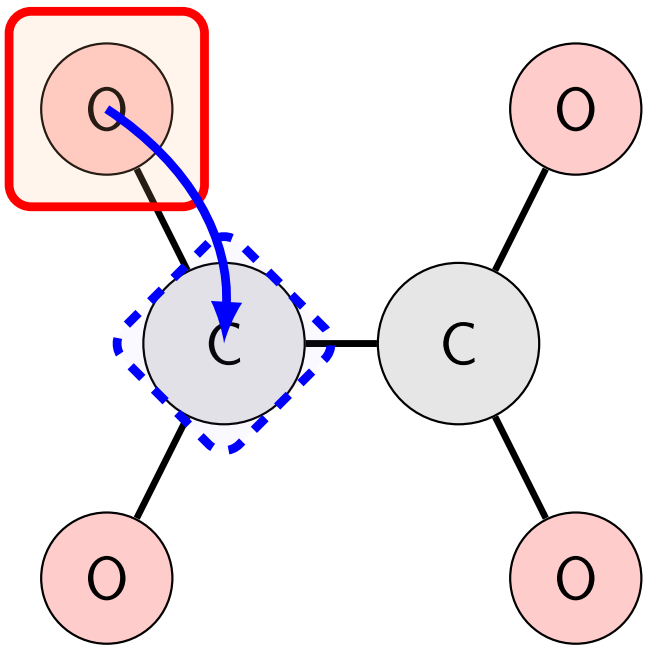


$$d_1 : \sum_{O,C} Z_O Z_C = 48$$

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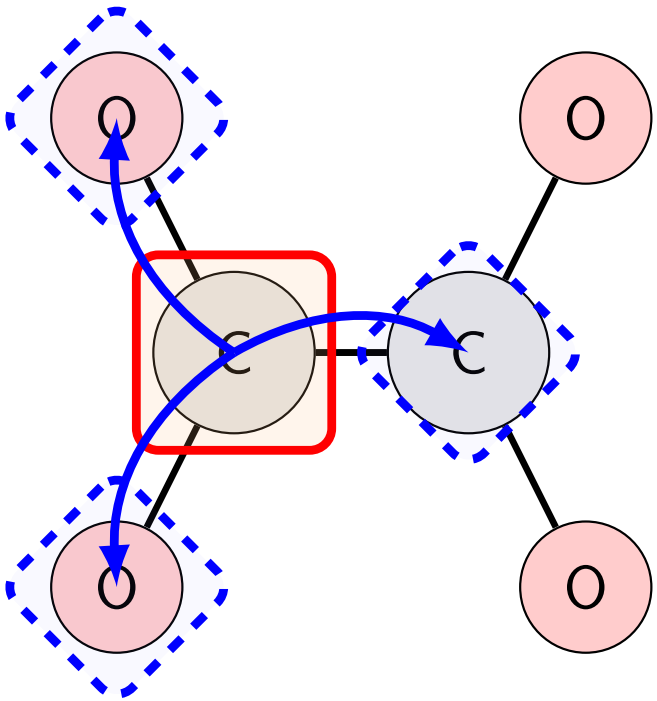


$$d_1 : 48 + \sum_{C,O} Z_O Z_C = 144 + 48$$

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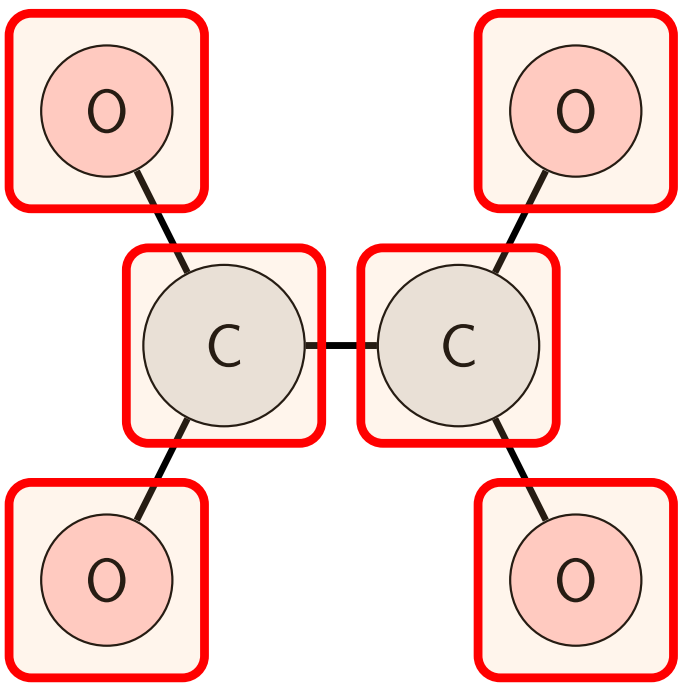


$$d_1 : \sum_i \sum_j Z_i Z_j \delta(d_{i,j}, 1)$$

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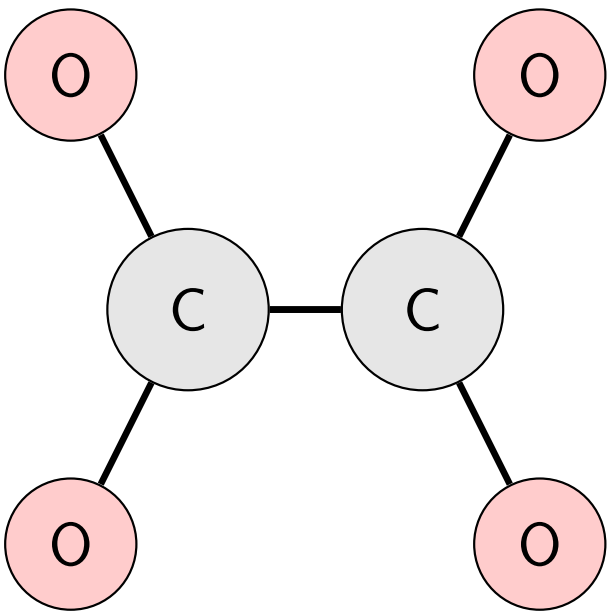
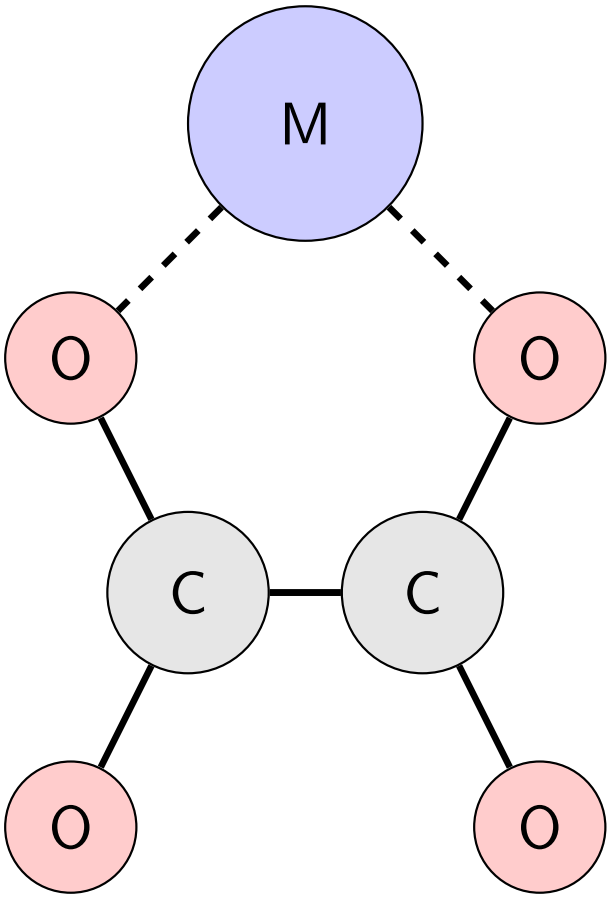



$$d_x : \sum_i \sum_j Z_i Z_j \delta(d_{ij}, x)$$

[4] Broto, P., Moreau, G. and Vandycke, C. *Eur. J. Med. Chem.*, 19(1):71-78, 1984.

# New descriptors

New descriptors based on autocorrelations[4]




How to adapt to TM complexes?

[4] Janet, J.P., and Kulik, H.J., *arXiv*, 1708.06017, 2017.

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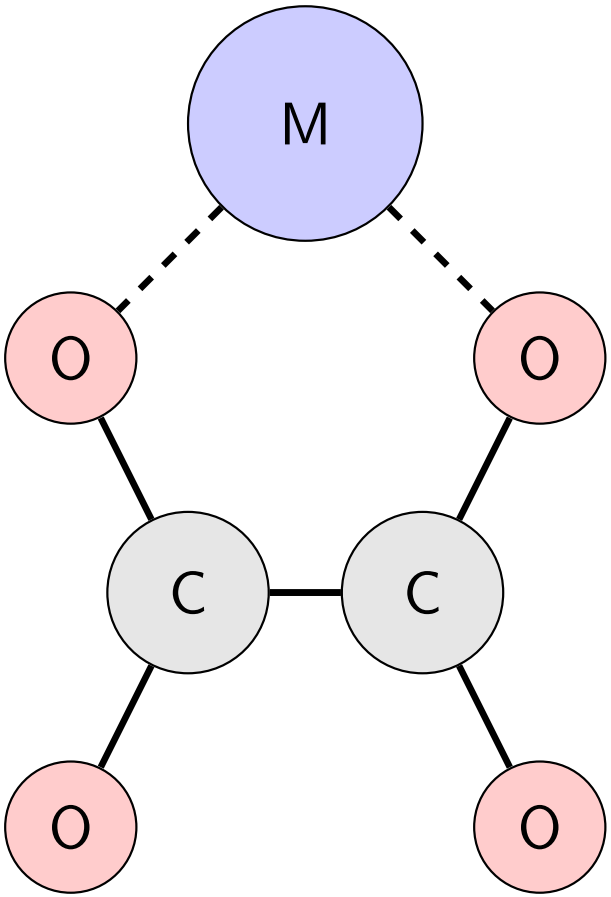


How to adapt to TM complexes?
restrict the scope to focus on
*near-metal atoms*

[4] Janet, J.P., and Kulik, H.J., *arXiv*, 1708.06017, 2017.

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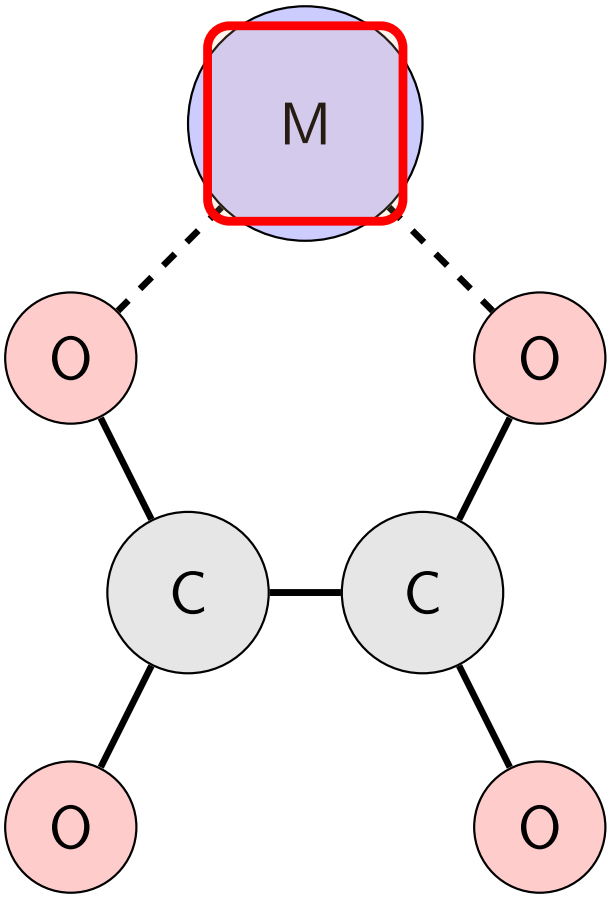


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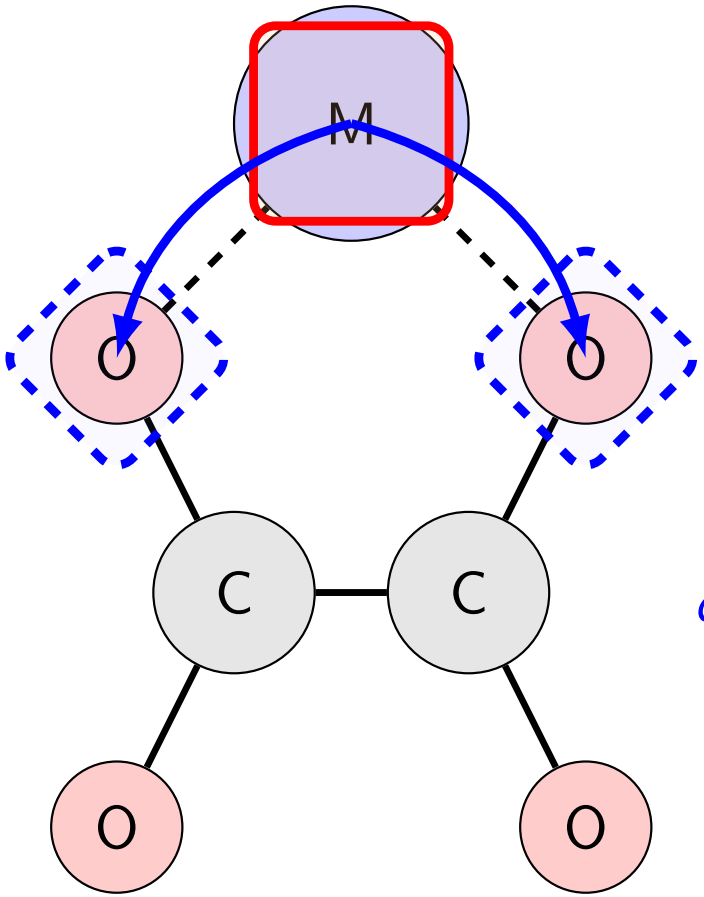


How to adapt to TM complexes?

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$$d_1 : \sum_{M,O} Z_M Z_O$$

[4] Janet, J.P., and Kulik, H.J., *arXiv*, 1708.06017, 2017.

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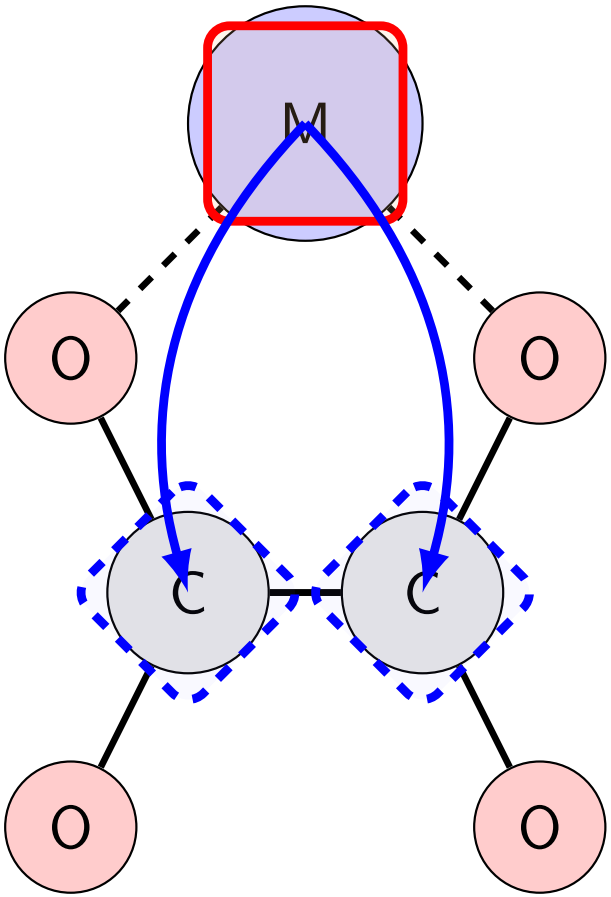
New descriptors based on autocorrelations[4]




How to adapt to TM complexes?

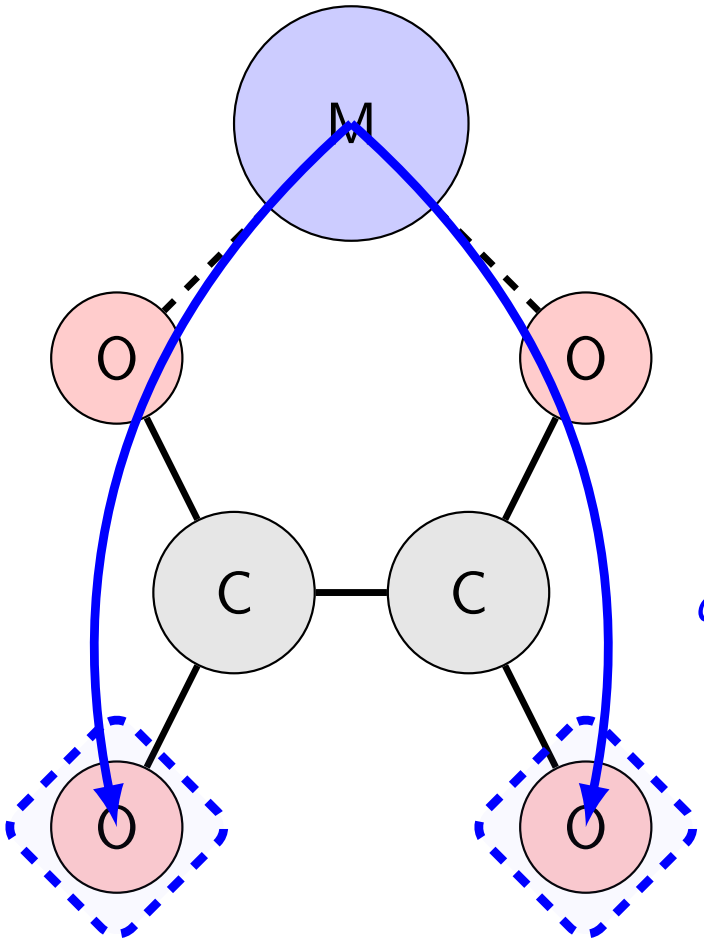
restrict the scope to focus on *near-metal atoms*

$$d_2 : \sum_{M,C} Z_M Z_C$$

[4] Janet, J.P., and Kulik, H.J., *arXiv*, 1708.06017, 2017.

# New descriptors

New descriptors based on autocorrelations[4]




How to adapt to TM complexes?

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$$d_3 : \sum_{M,O} Z_M Z_O$$

[4] Janet, J.P., and Kulik, H.J., *arXiv*, 1708.06017, 2017.

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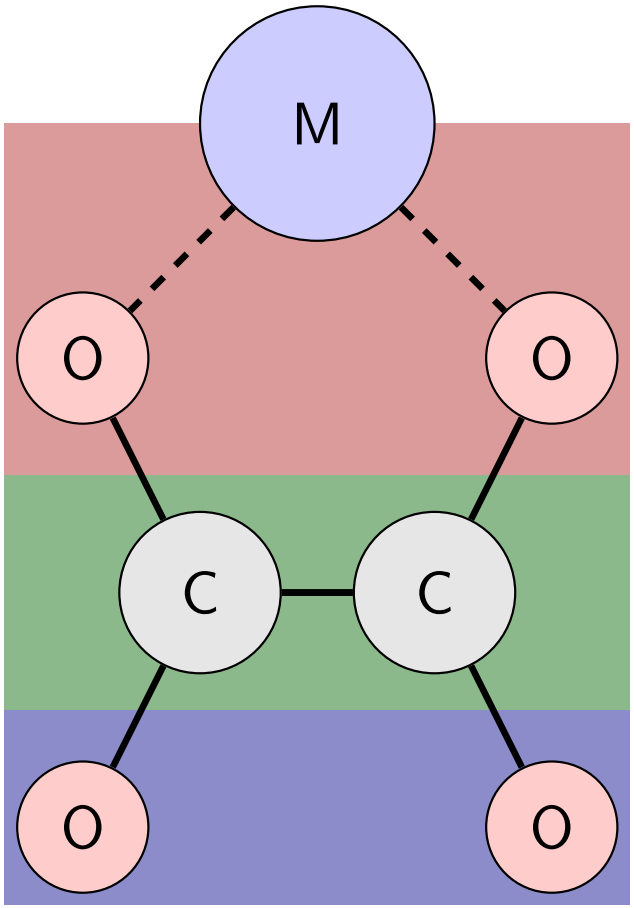
New descriptors based on autocorrelations[4]

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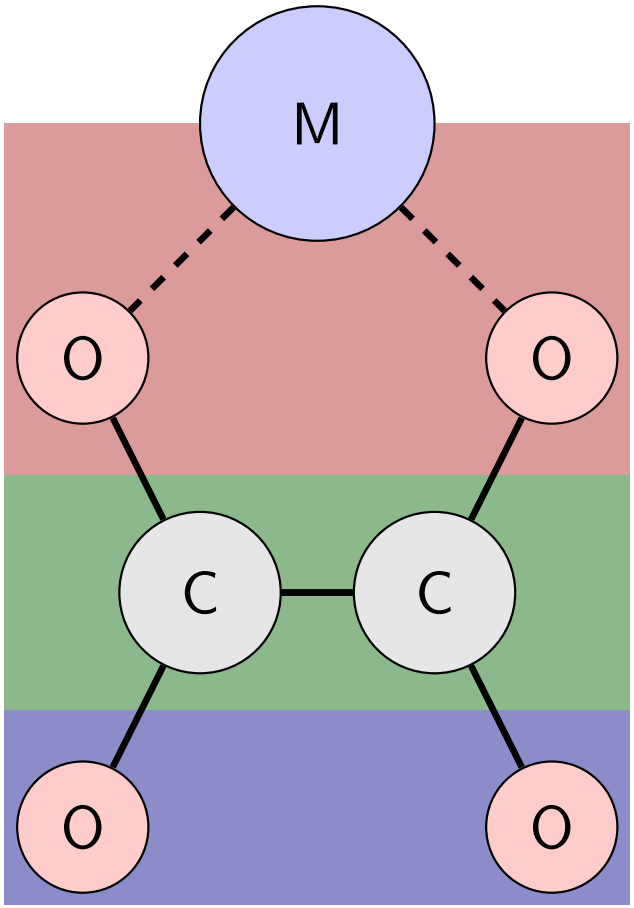


How to adapt to TM complexes?

restrict the scope to focus on *near-metal atoms*

$$d_3 : \sum_{M,O} Z_M Z_O \, (Z_i - Z_j)$$

[4] Janet, J.P., and Kulik, H.J., *arXiv*, 1708.06017, 2017.

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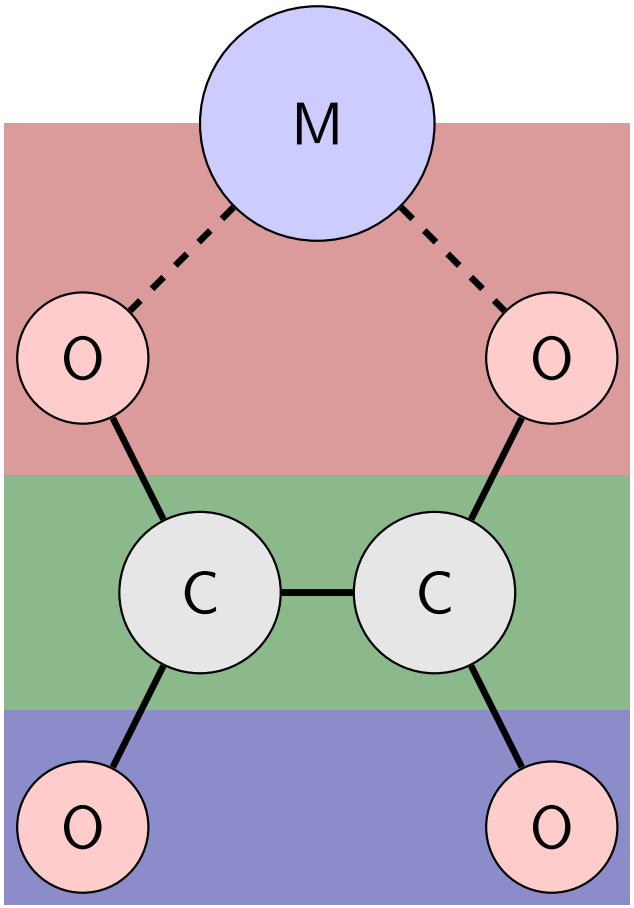


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$$d_3 : \sum_{M,O} Z_M Z_O \, (Z_i - Z_j)$$

properties: $T, \chi, Z, I, S$

[4] Janet, J.P., and Kulik, H.J., *arXiv*, 1708.06017, 2017.

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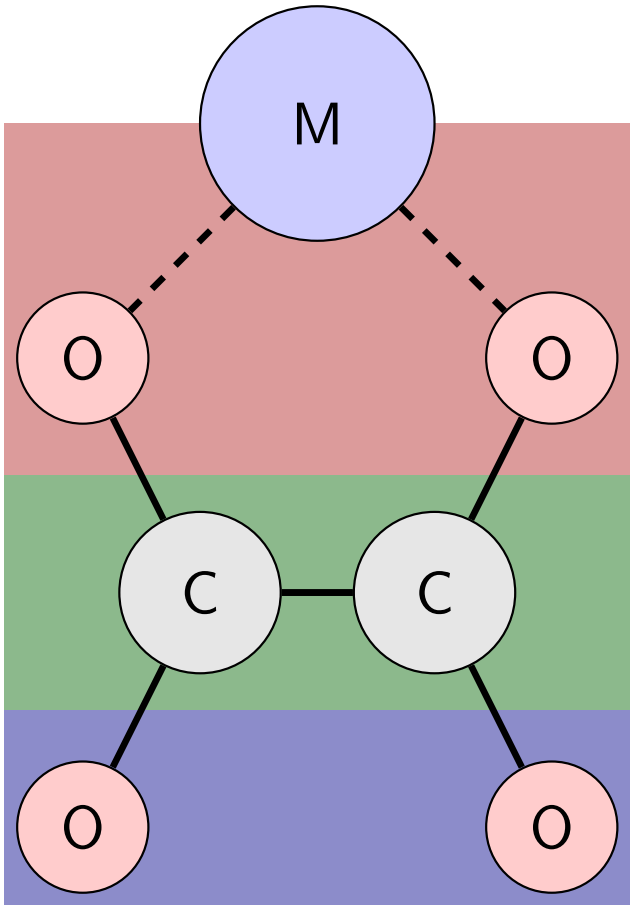



How to adapt to TM complexes?

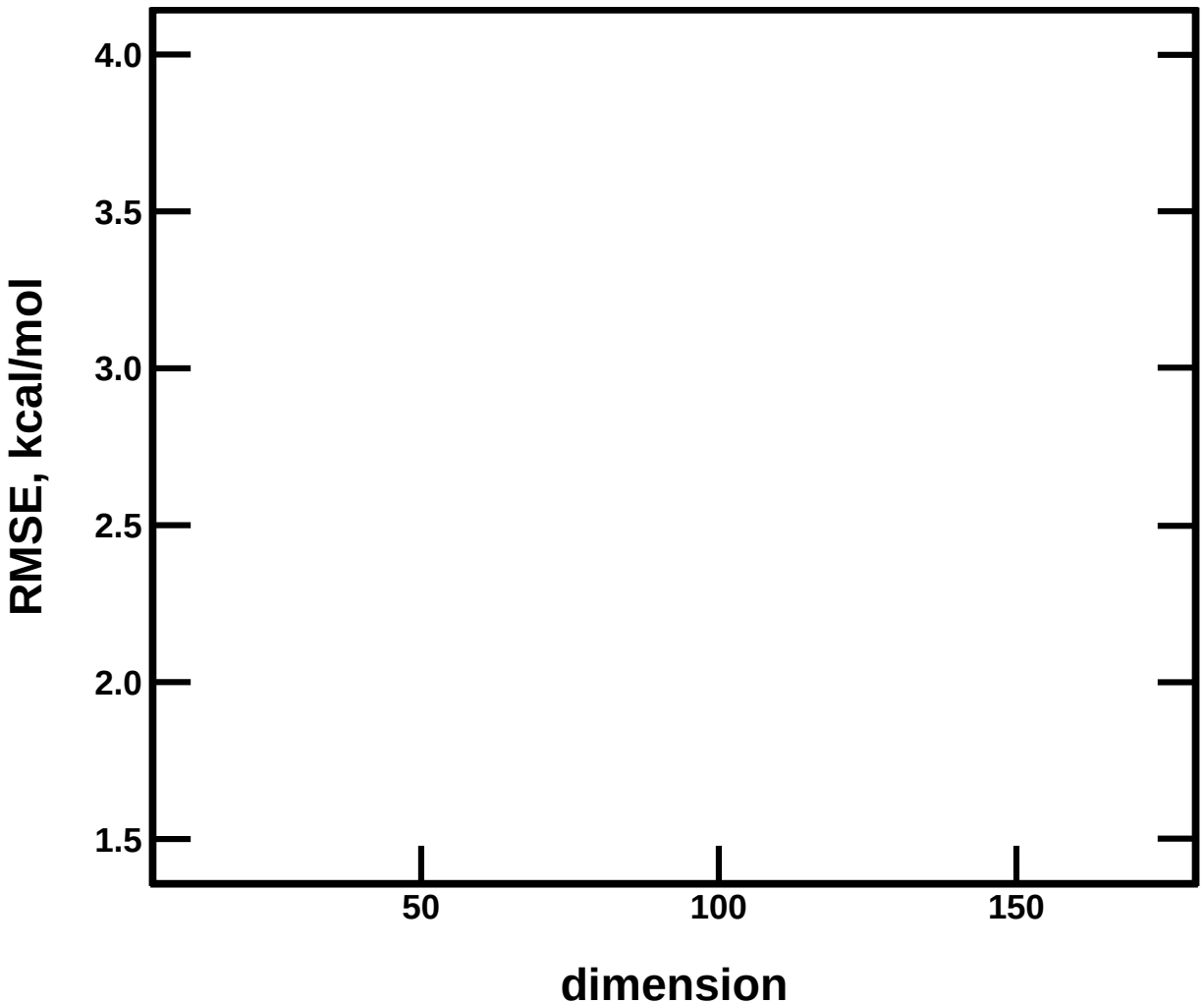
restrict the scope to focus on *near-metal atoms*

$$d_3 : \sum_{M,O} Z_M Z_O \, (Z_i - Z_j)$$

$\sim$ 160 features in total

[4] Janet, J.P., and Kulik, H.J., *arXiv*, 1708.06017, 2017.

# Feature selection

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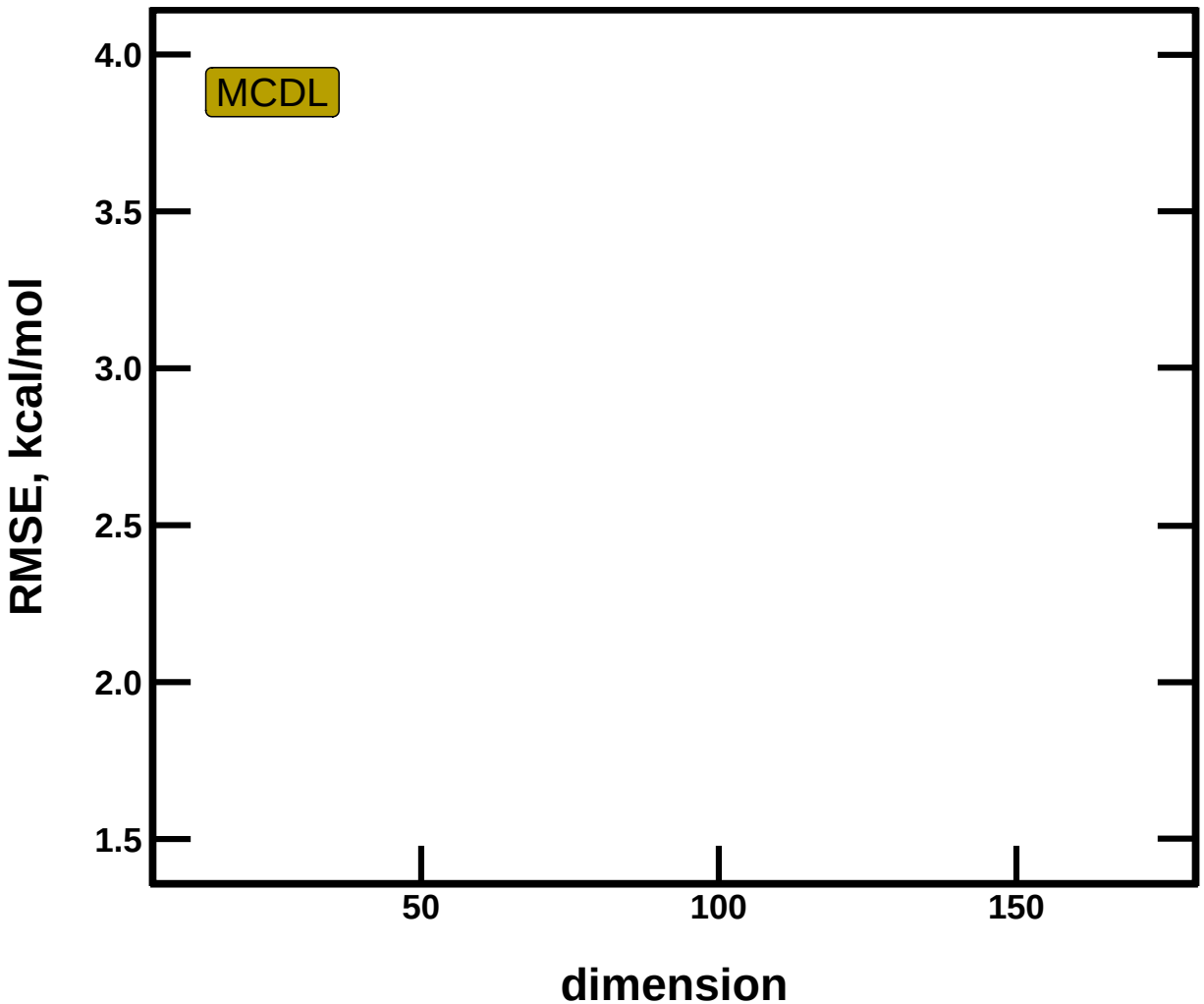


Janet, J.P., and Kulik, H.J., *arXiv*, 1708.06017, 2017.

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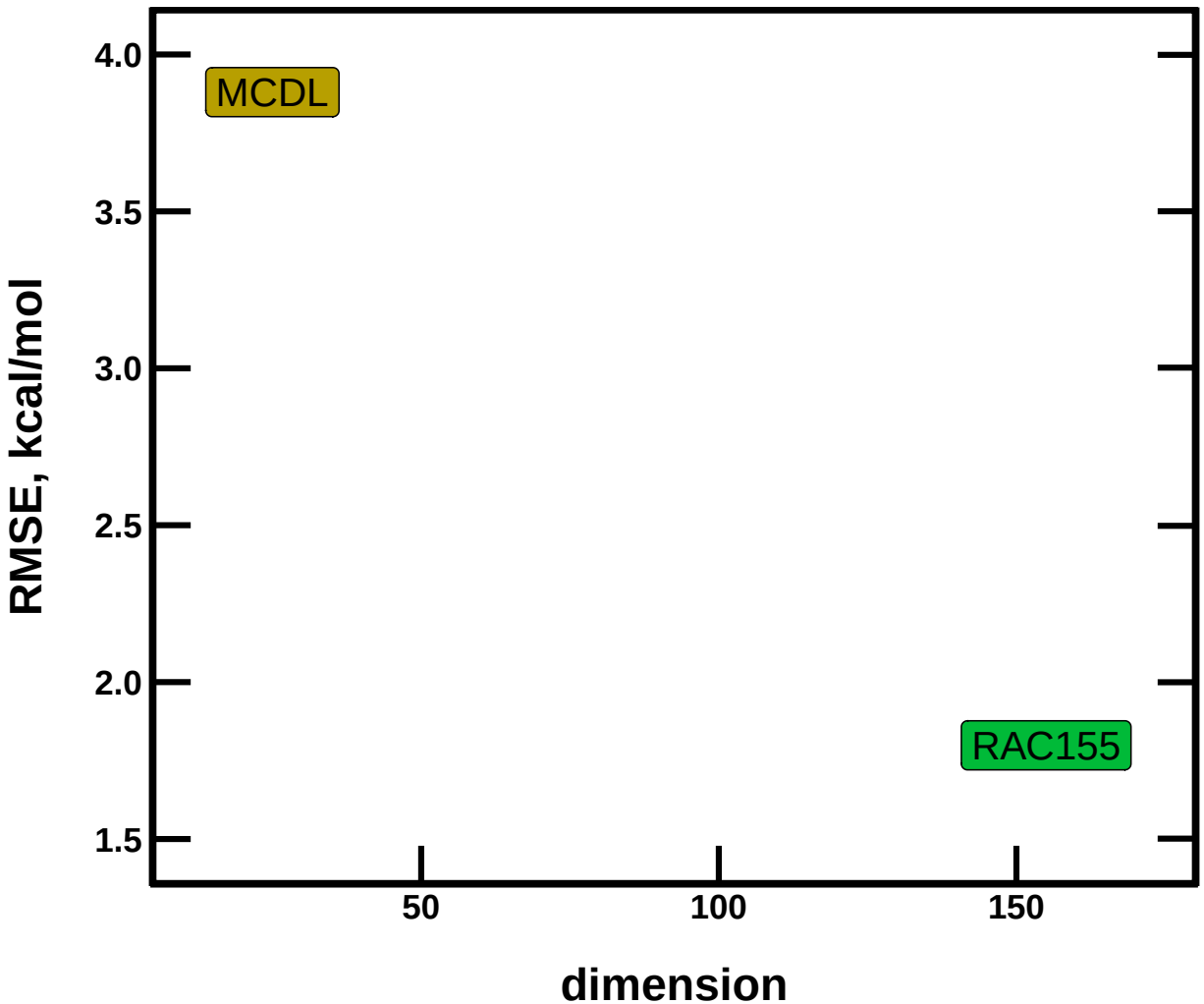


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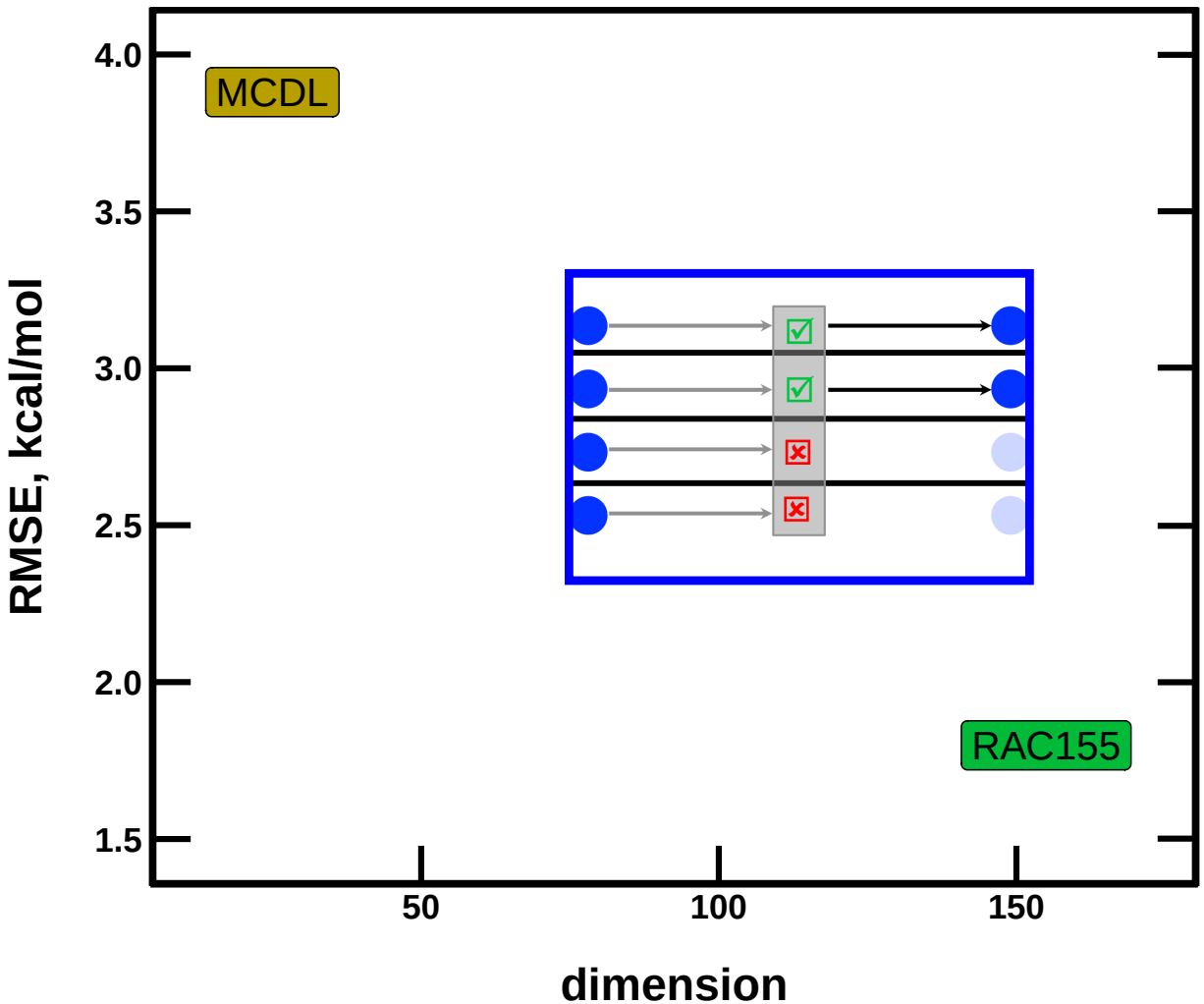


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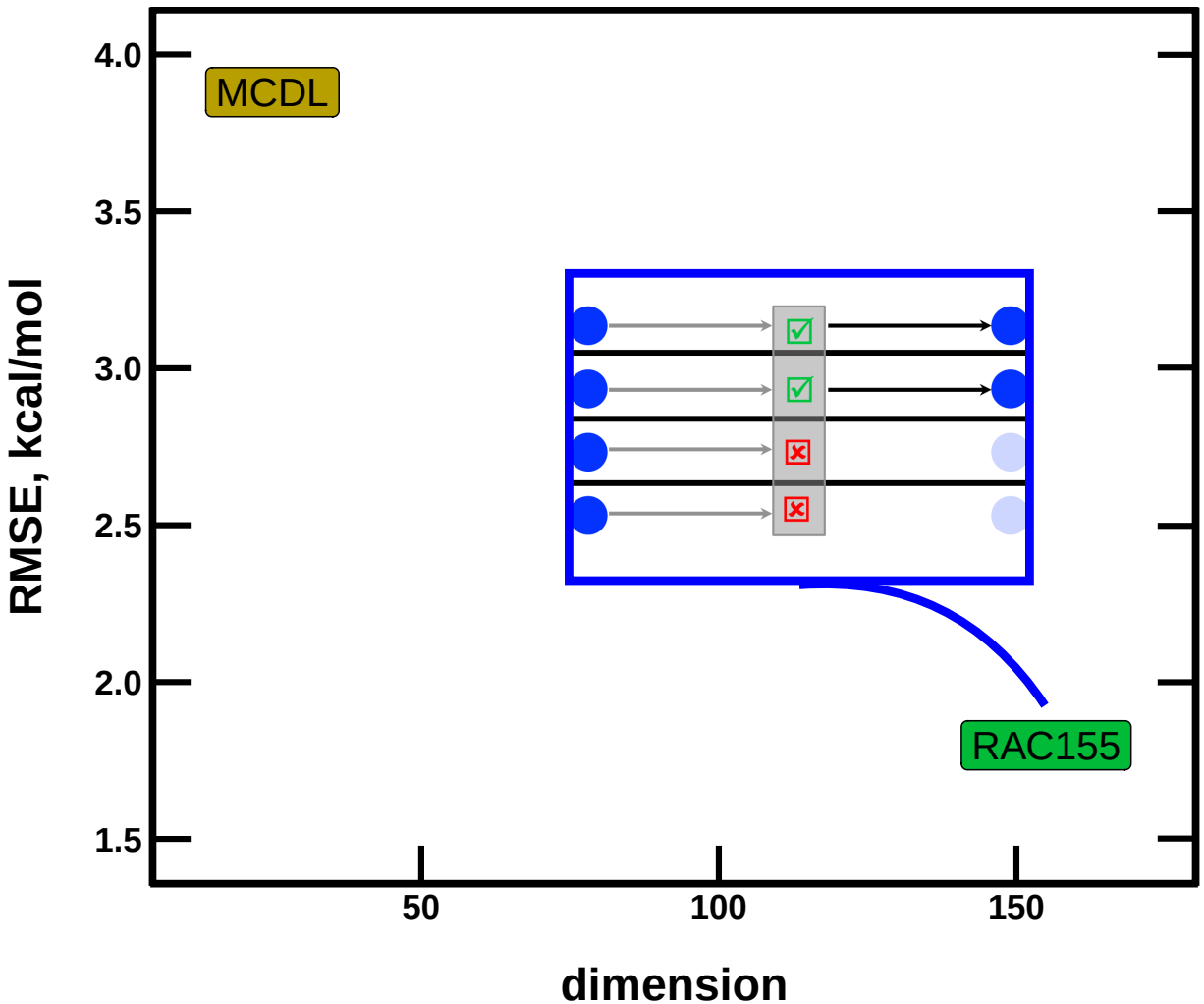


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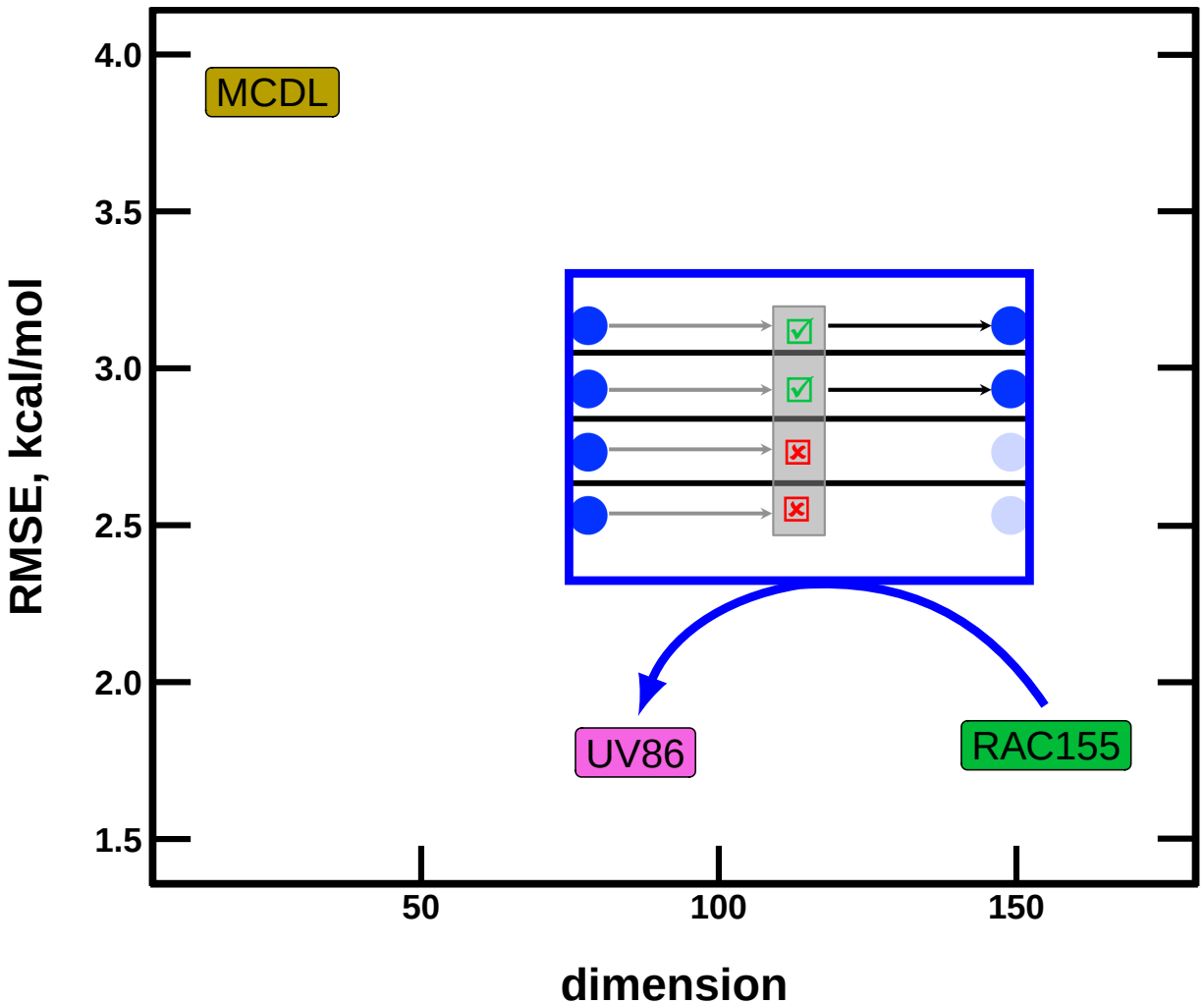


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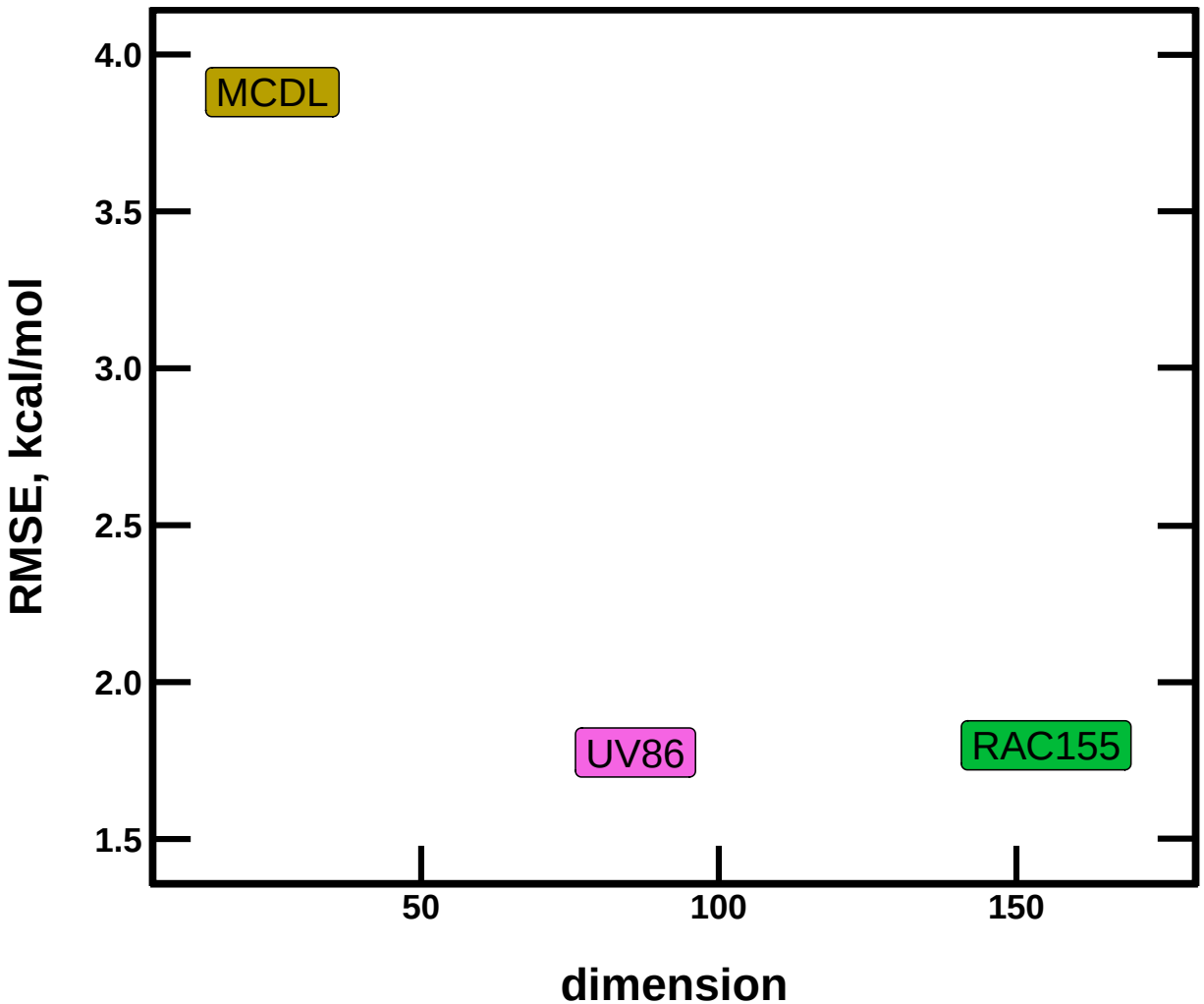


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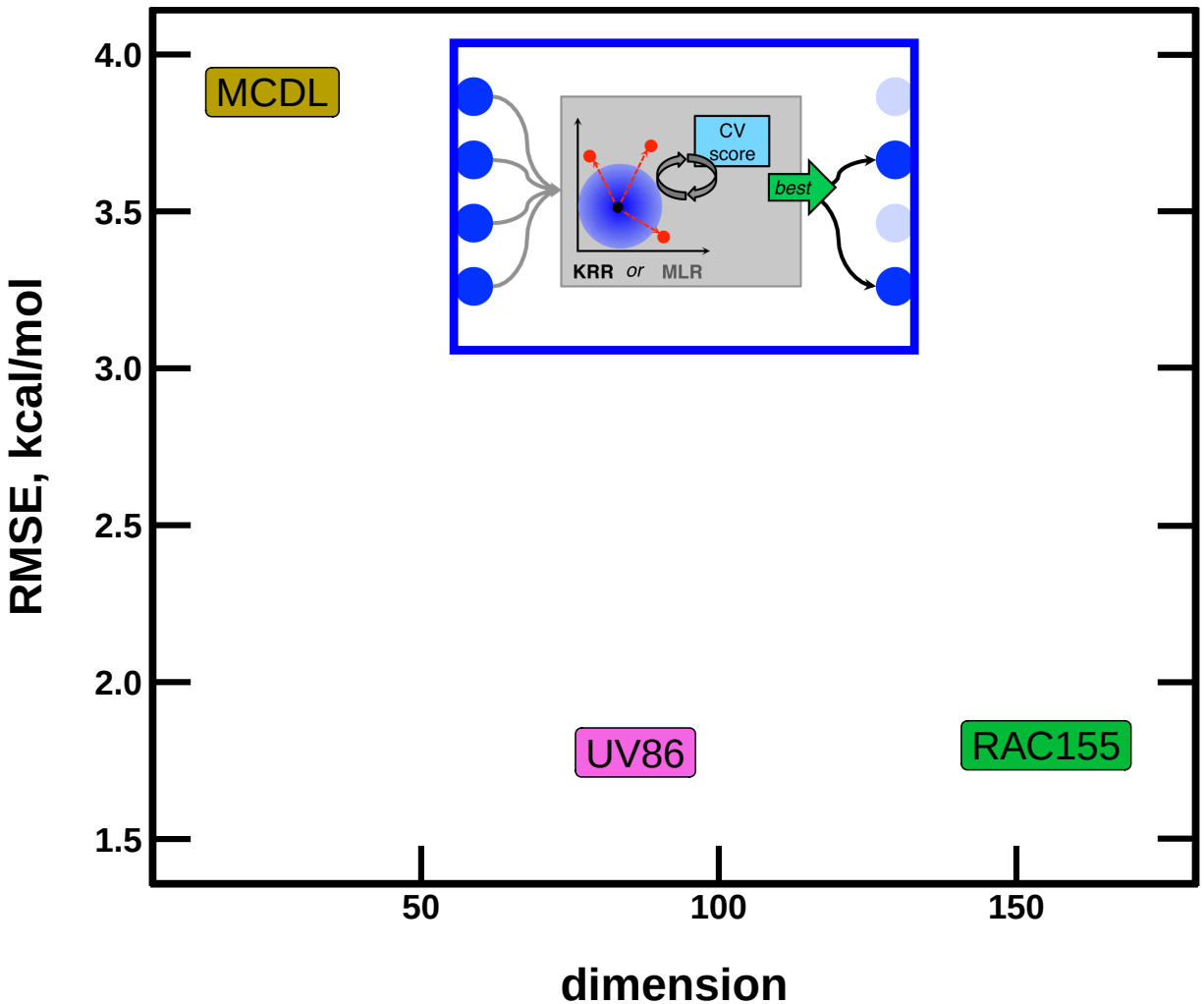


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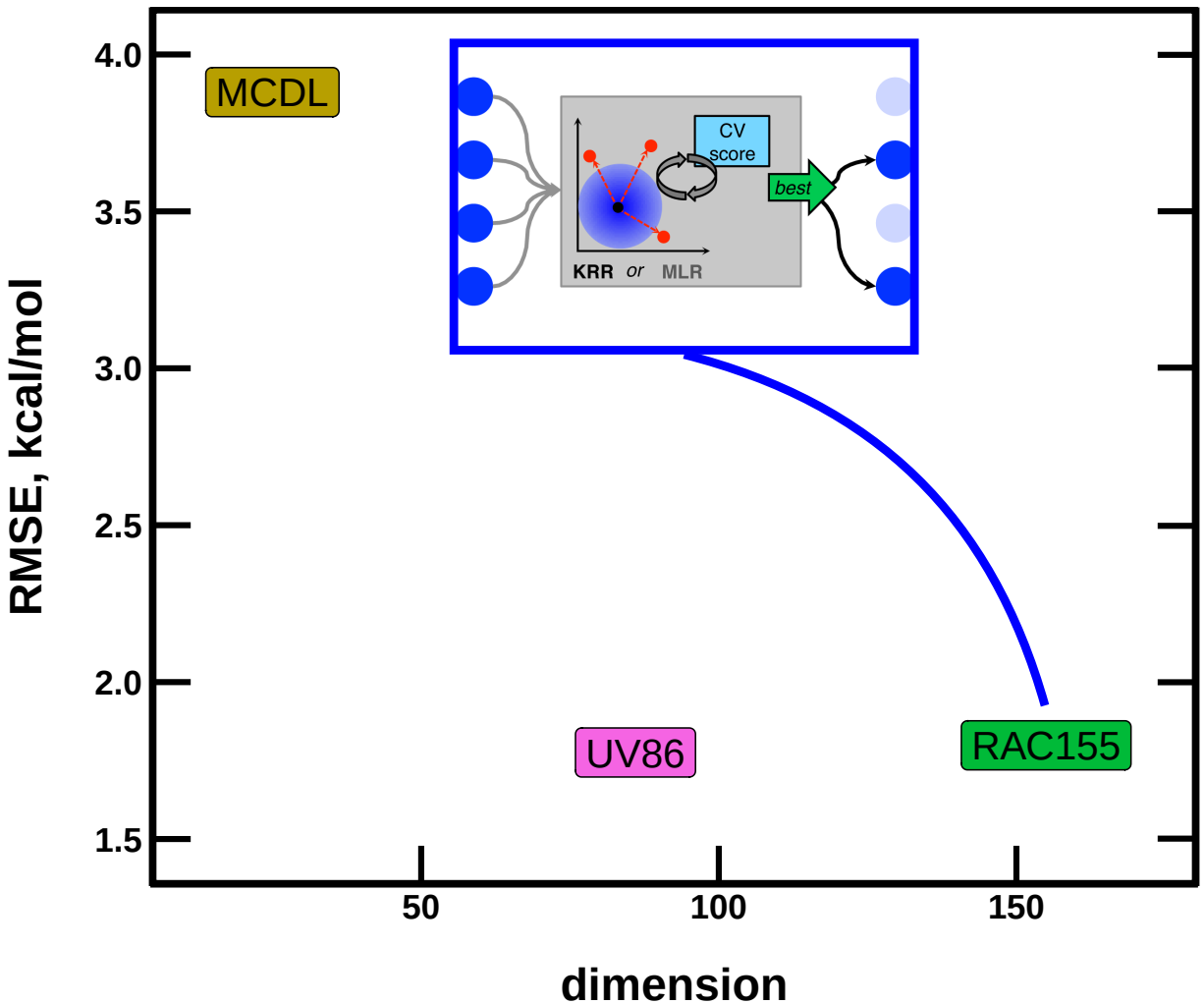


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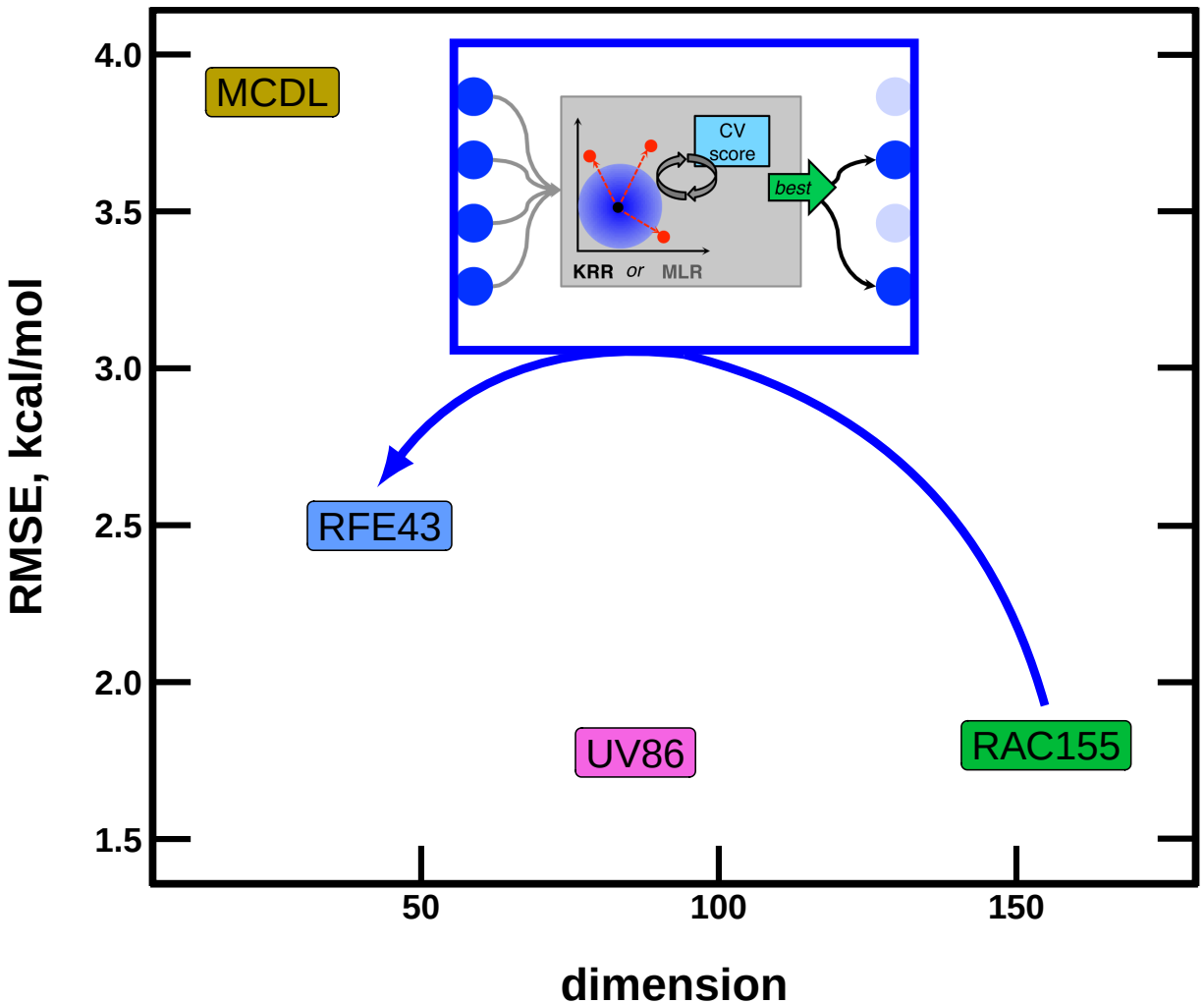


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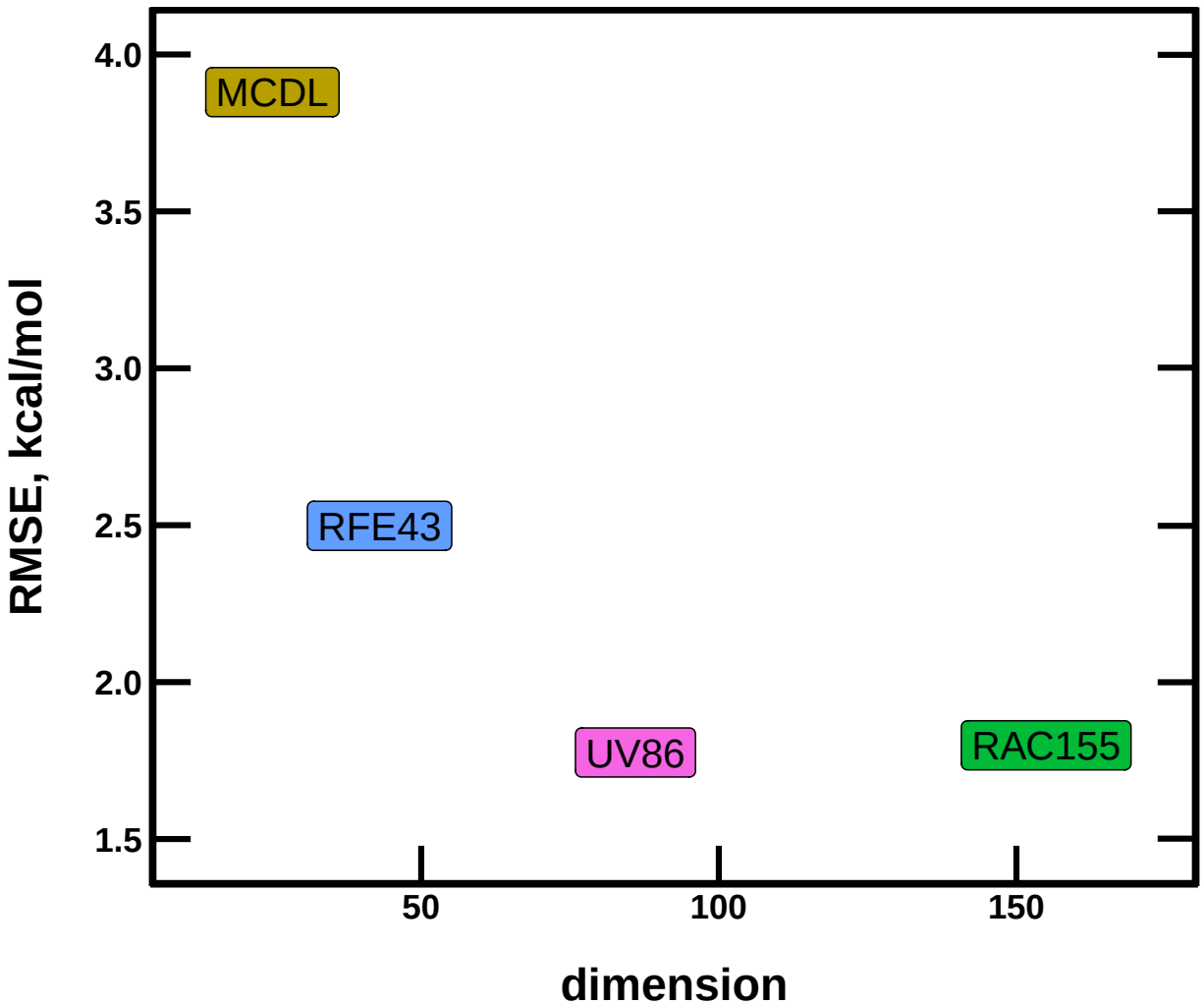


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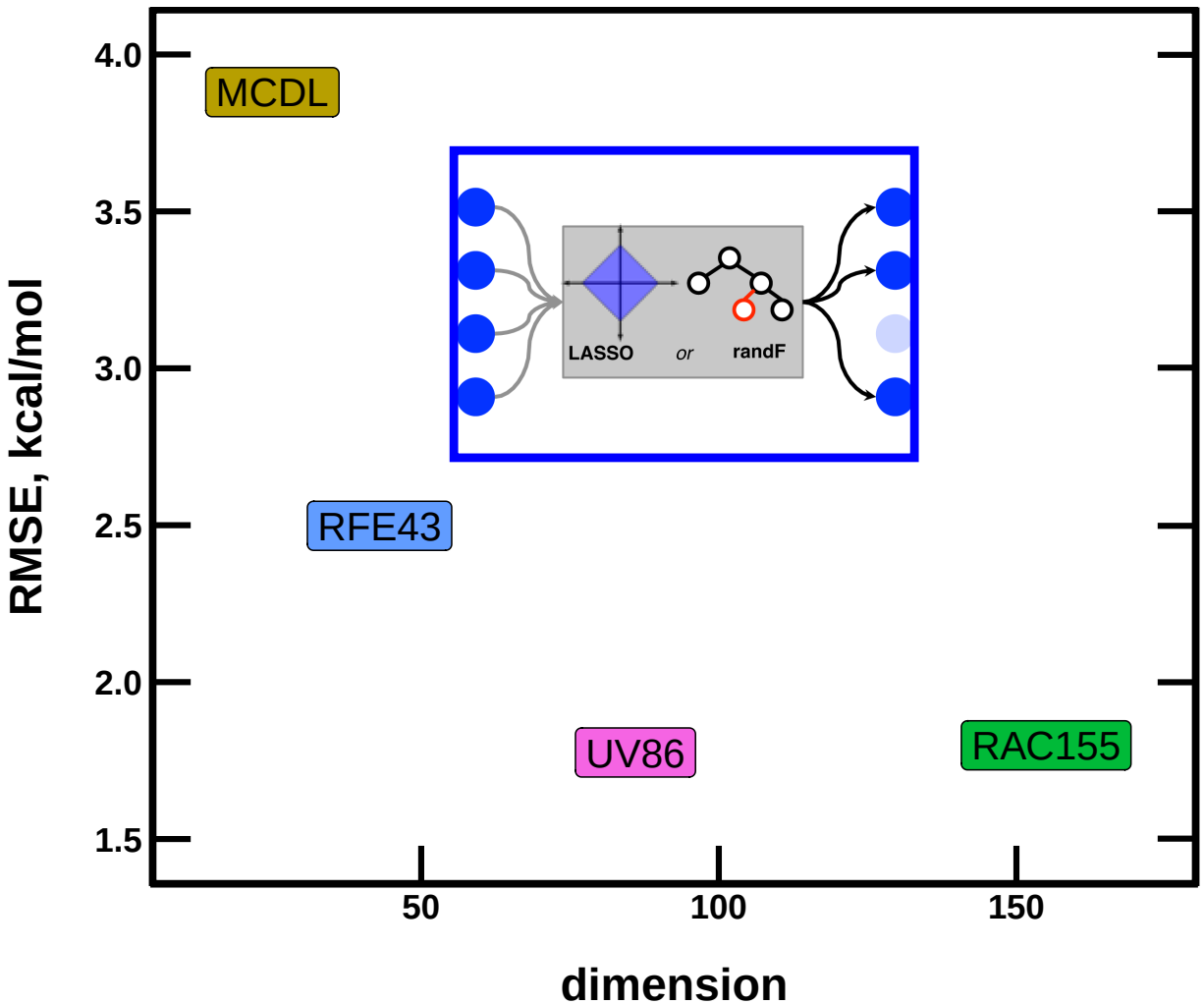


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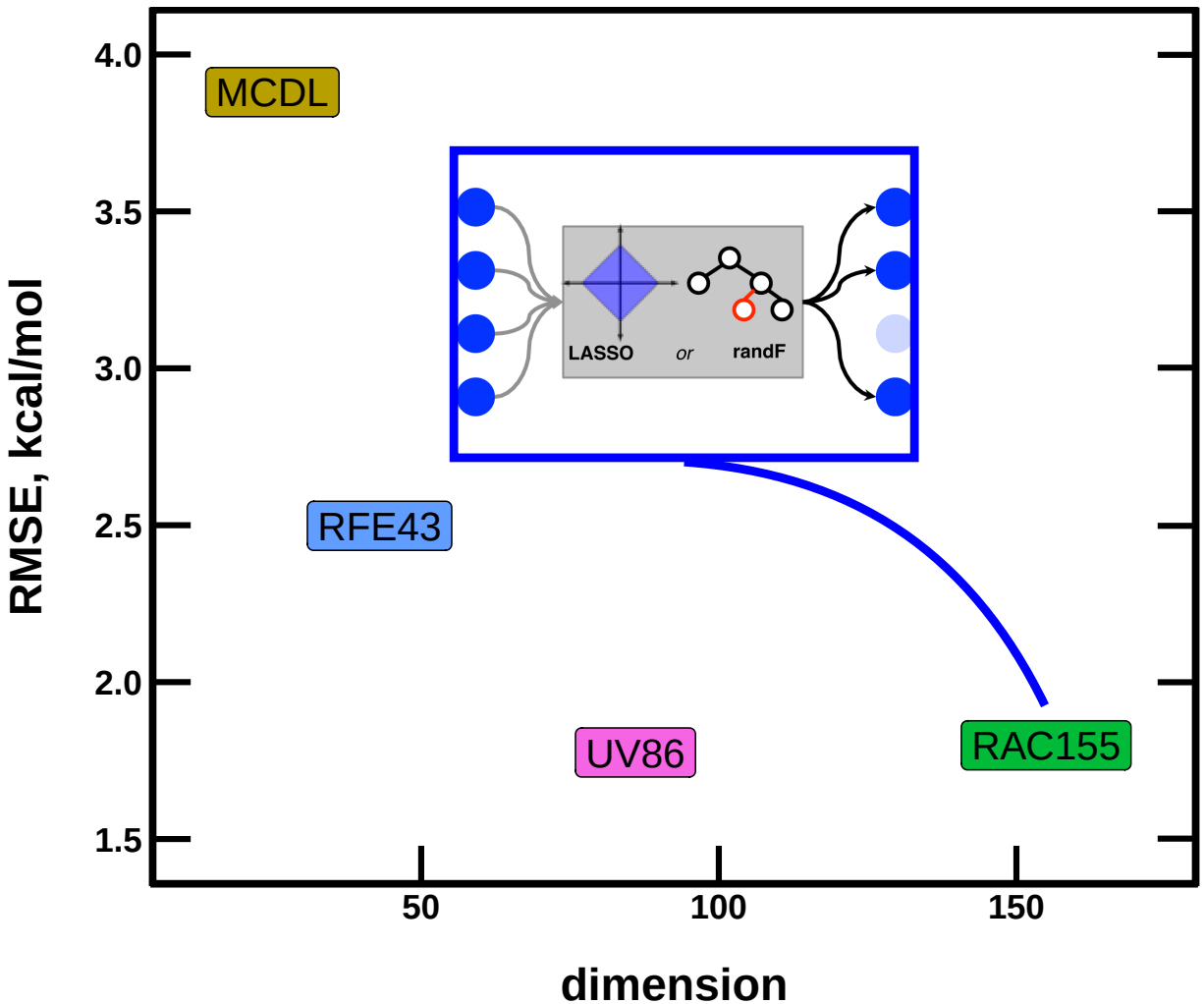


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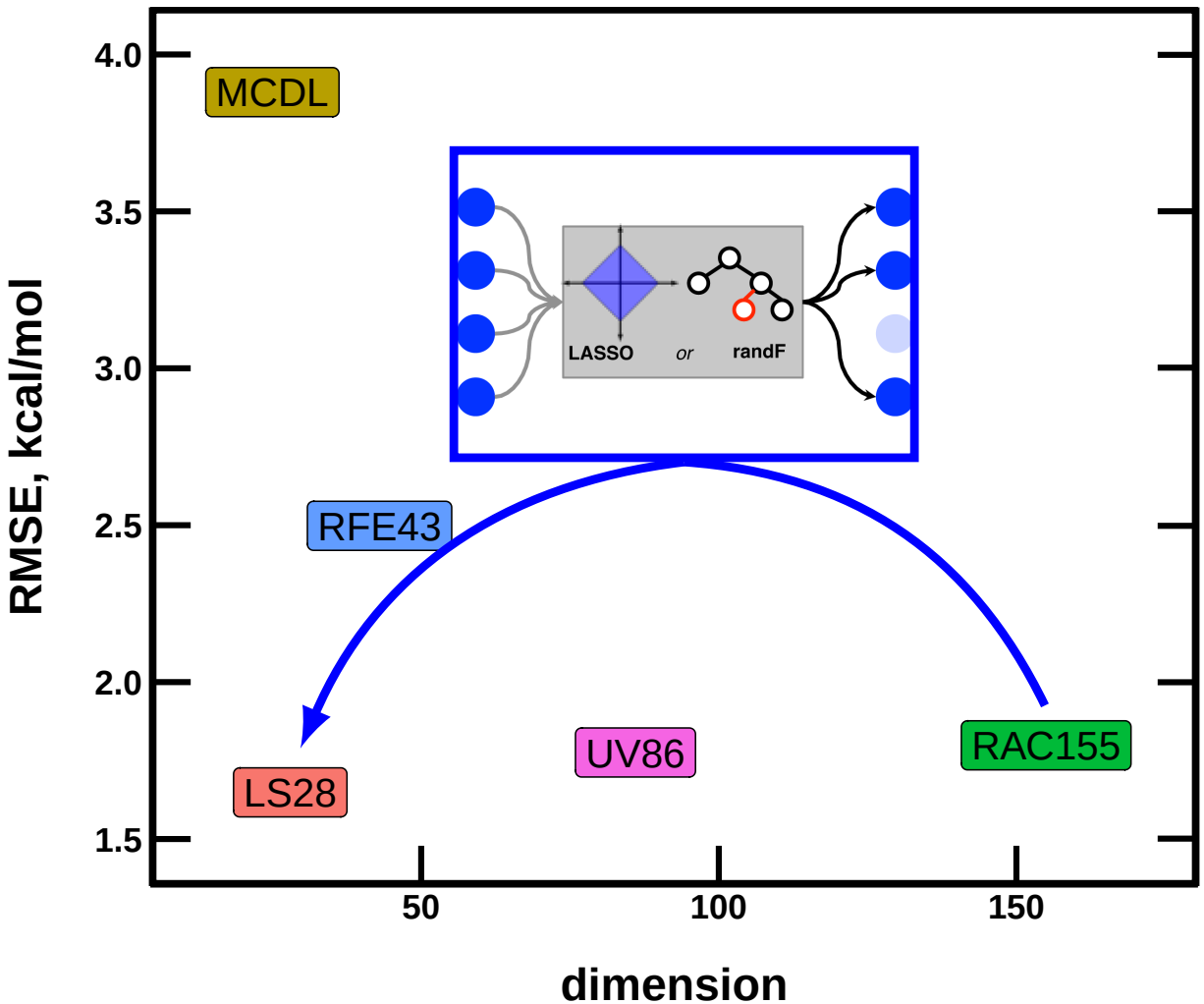


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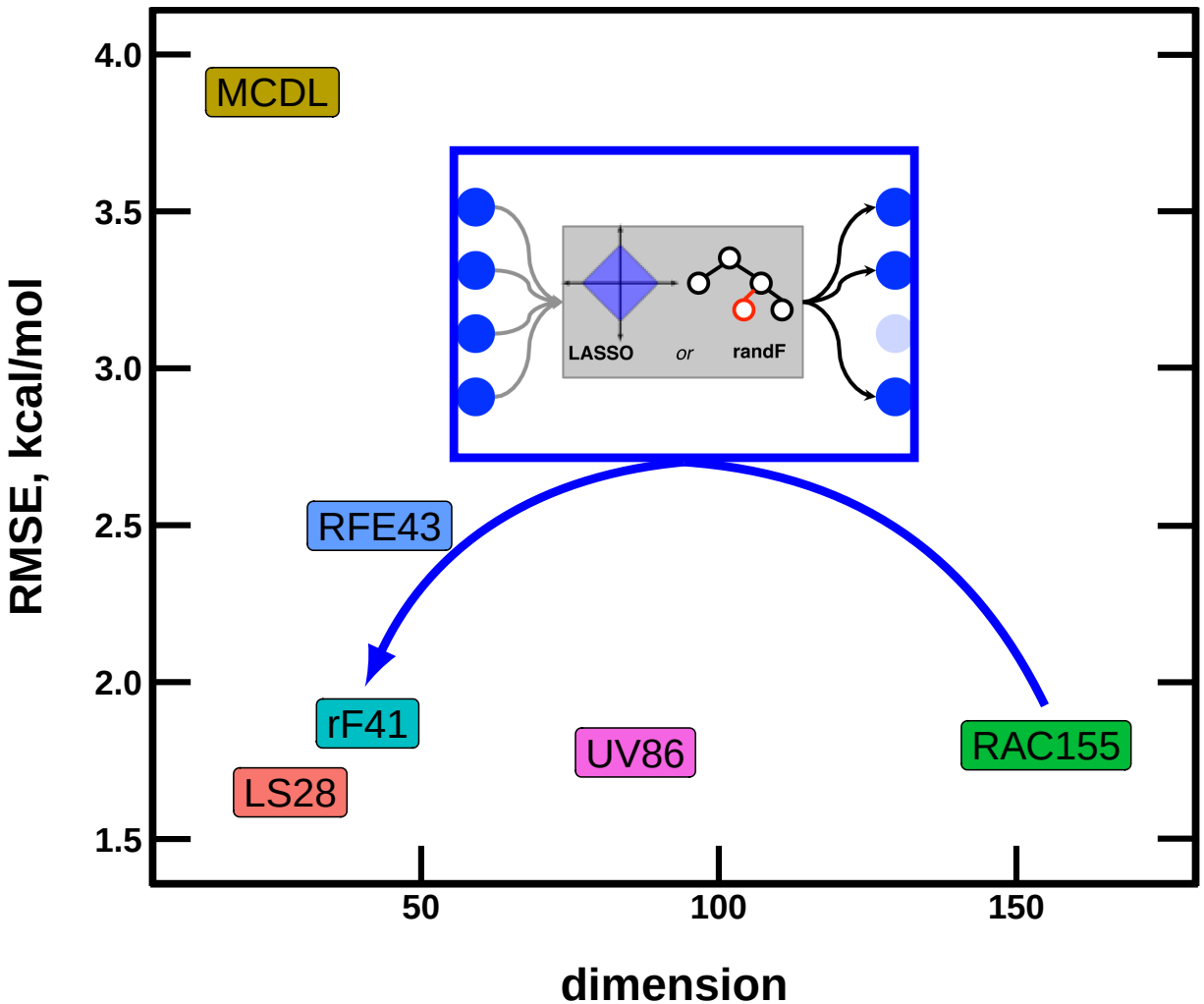


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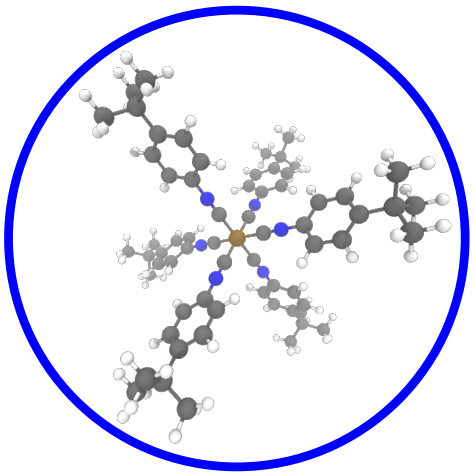
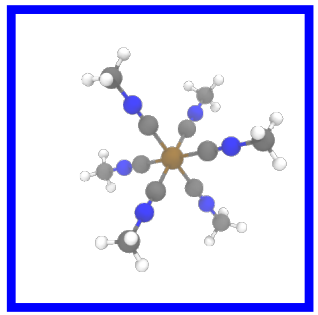
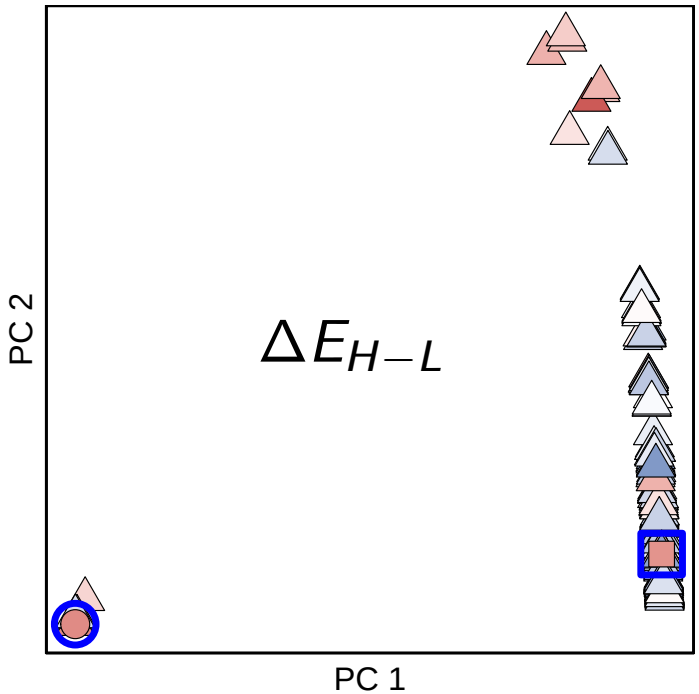
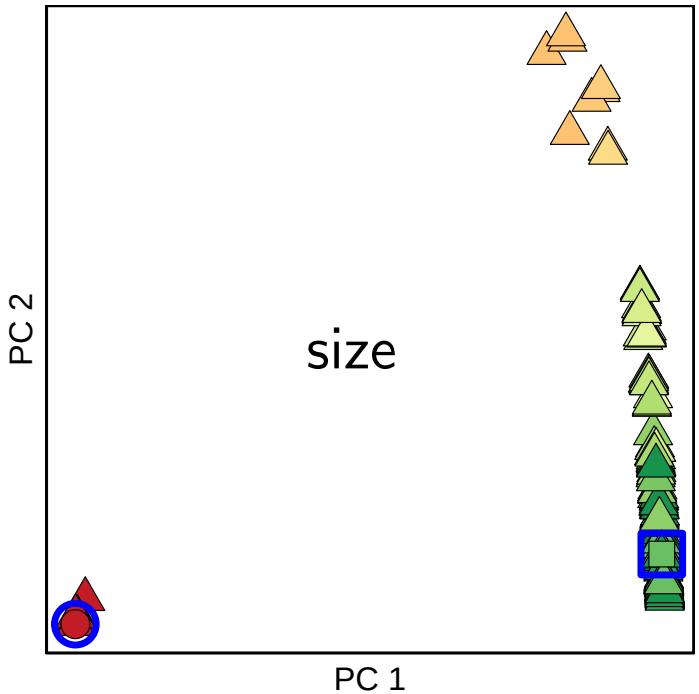
# A tale of two complexes, II







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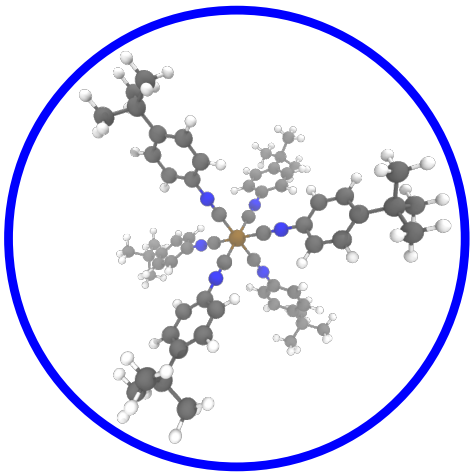

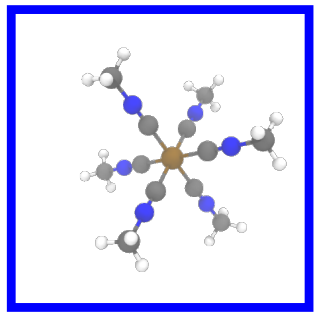

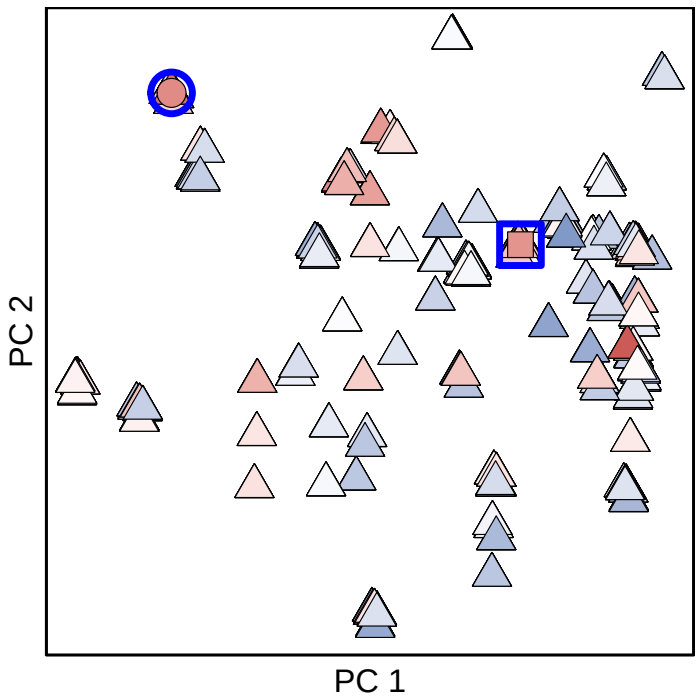


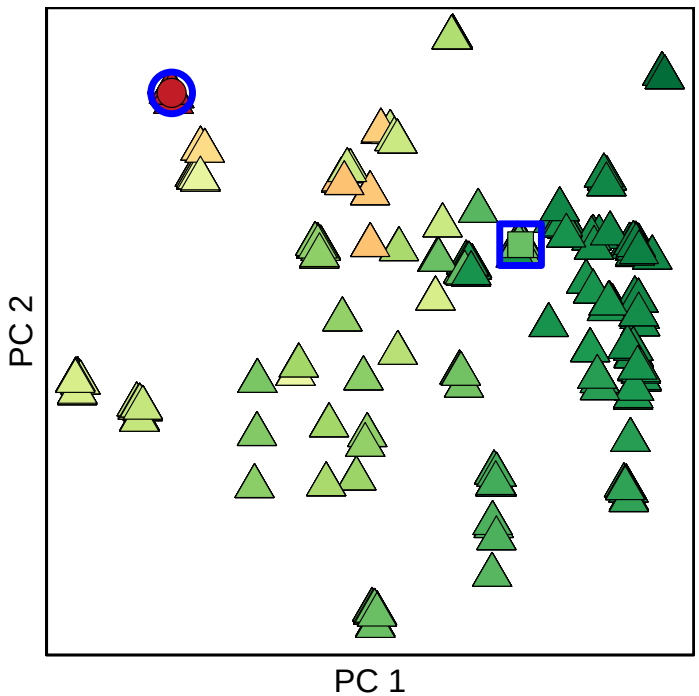

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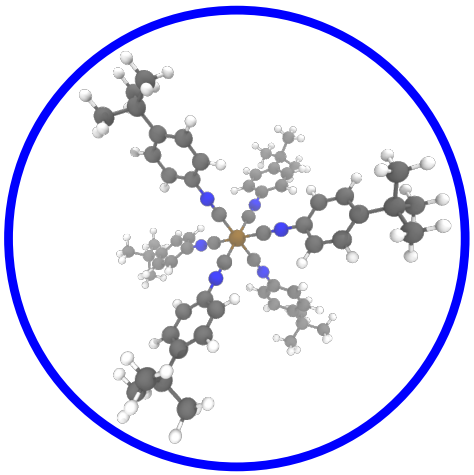
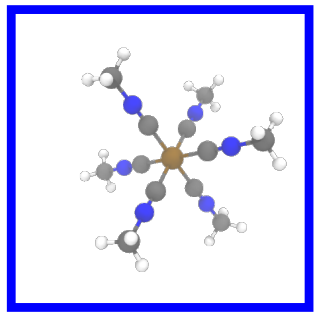
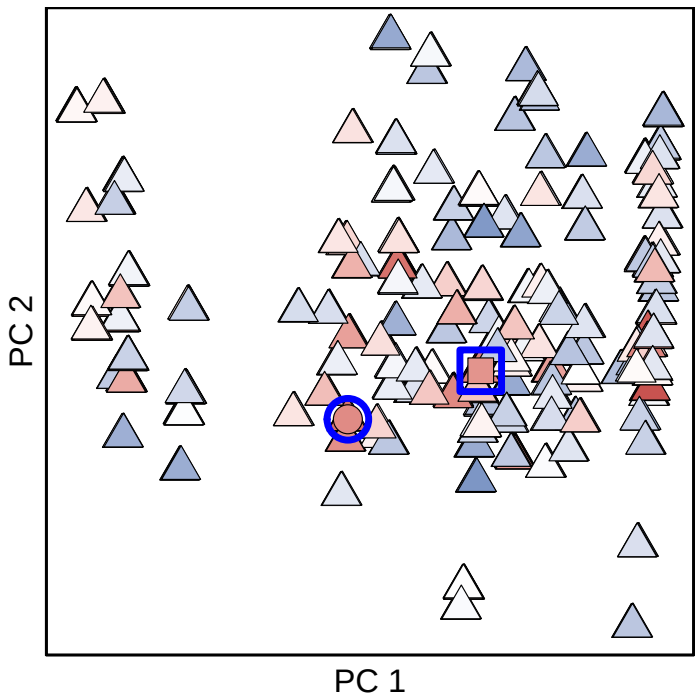
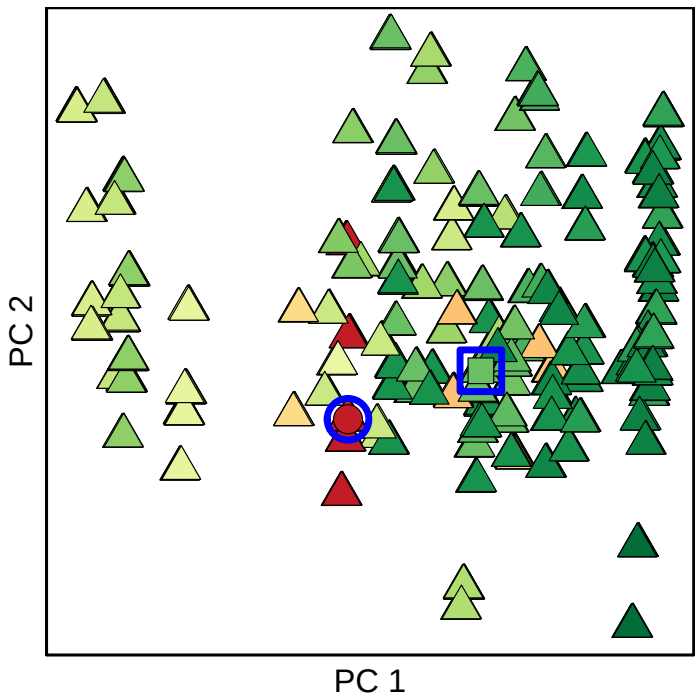
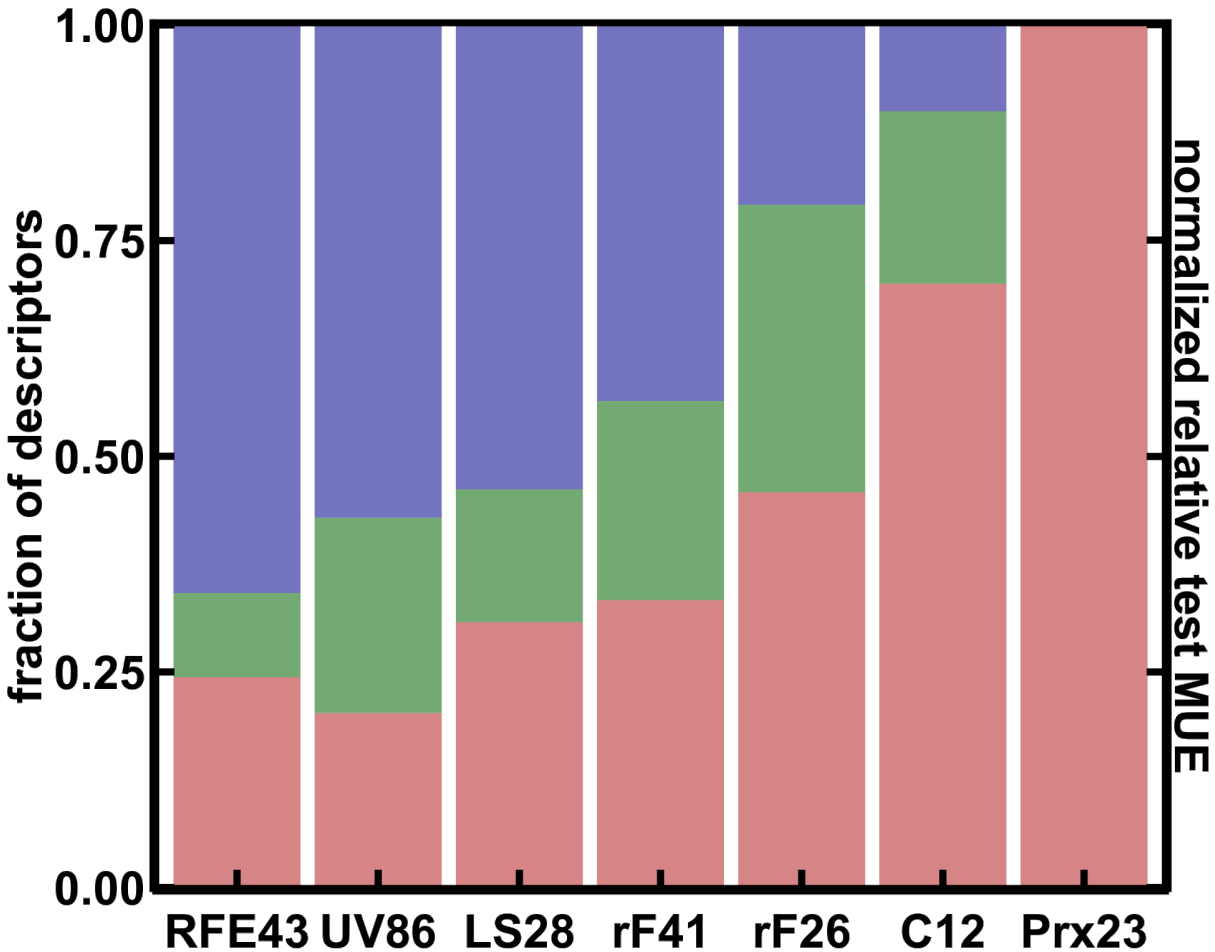






# How local is too local?




Janet, J.P., and Kulik, H.J., *arXiv*, 1708.06017, 2017.

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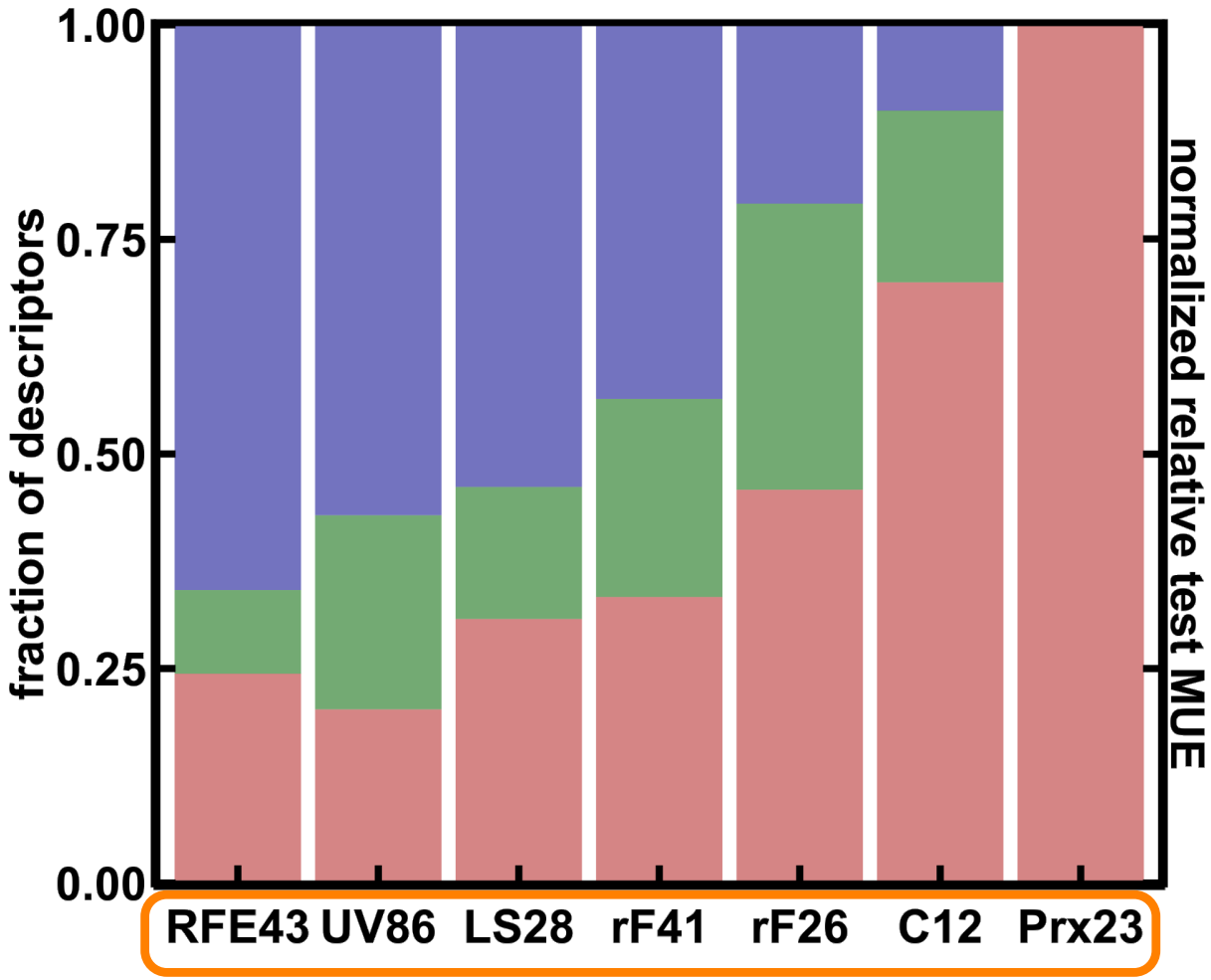


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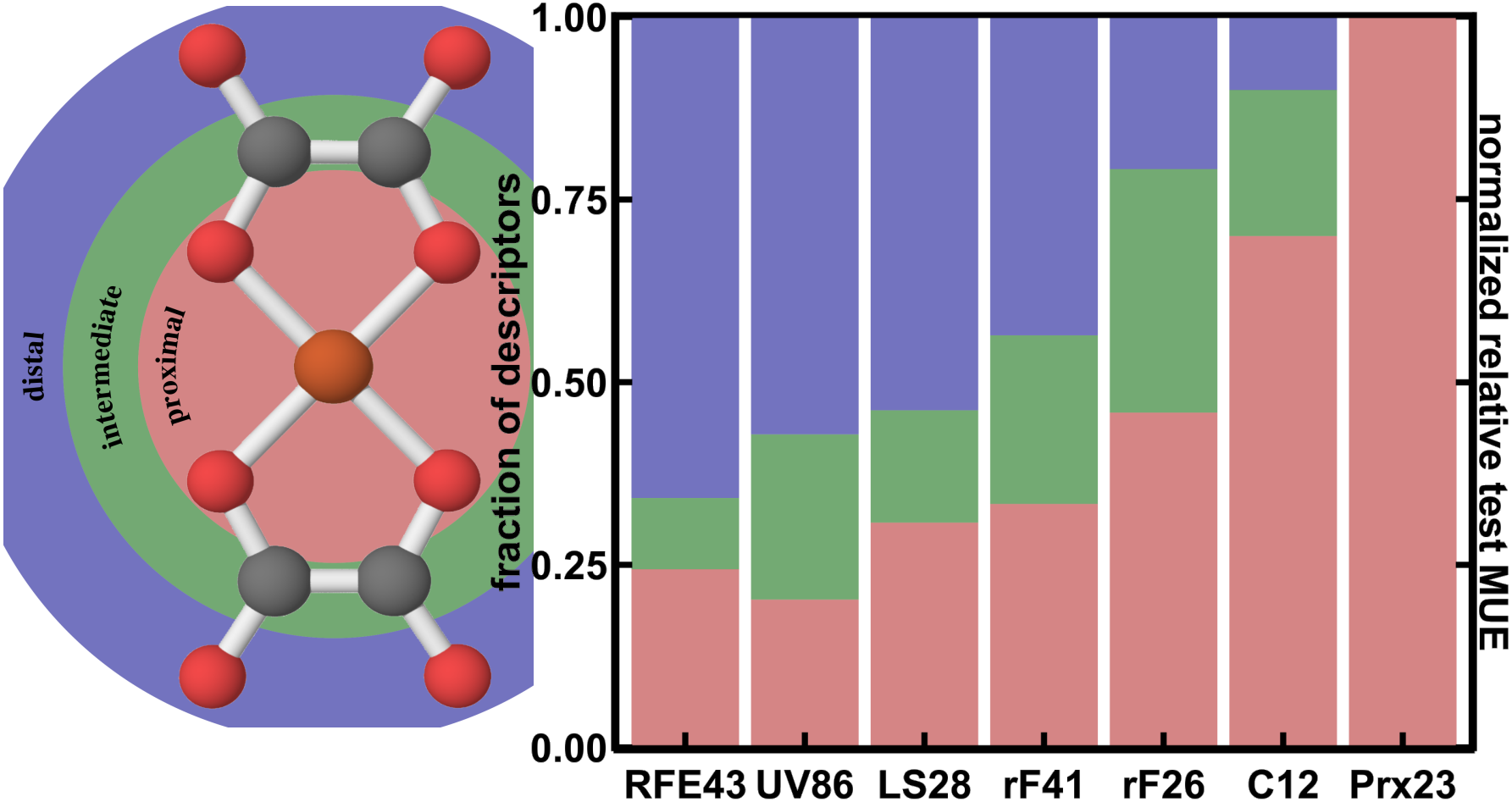


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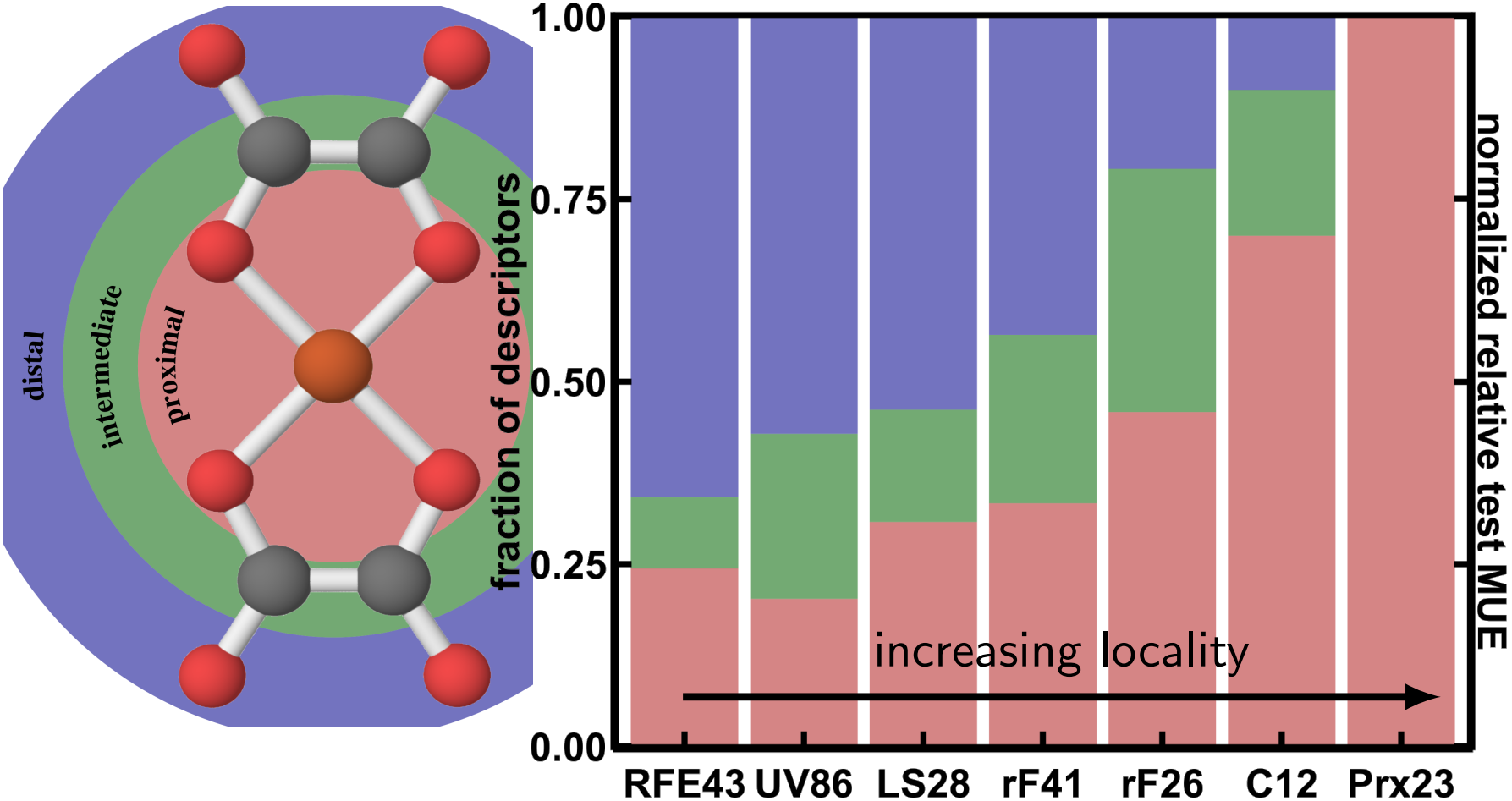


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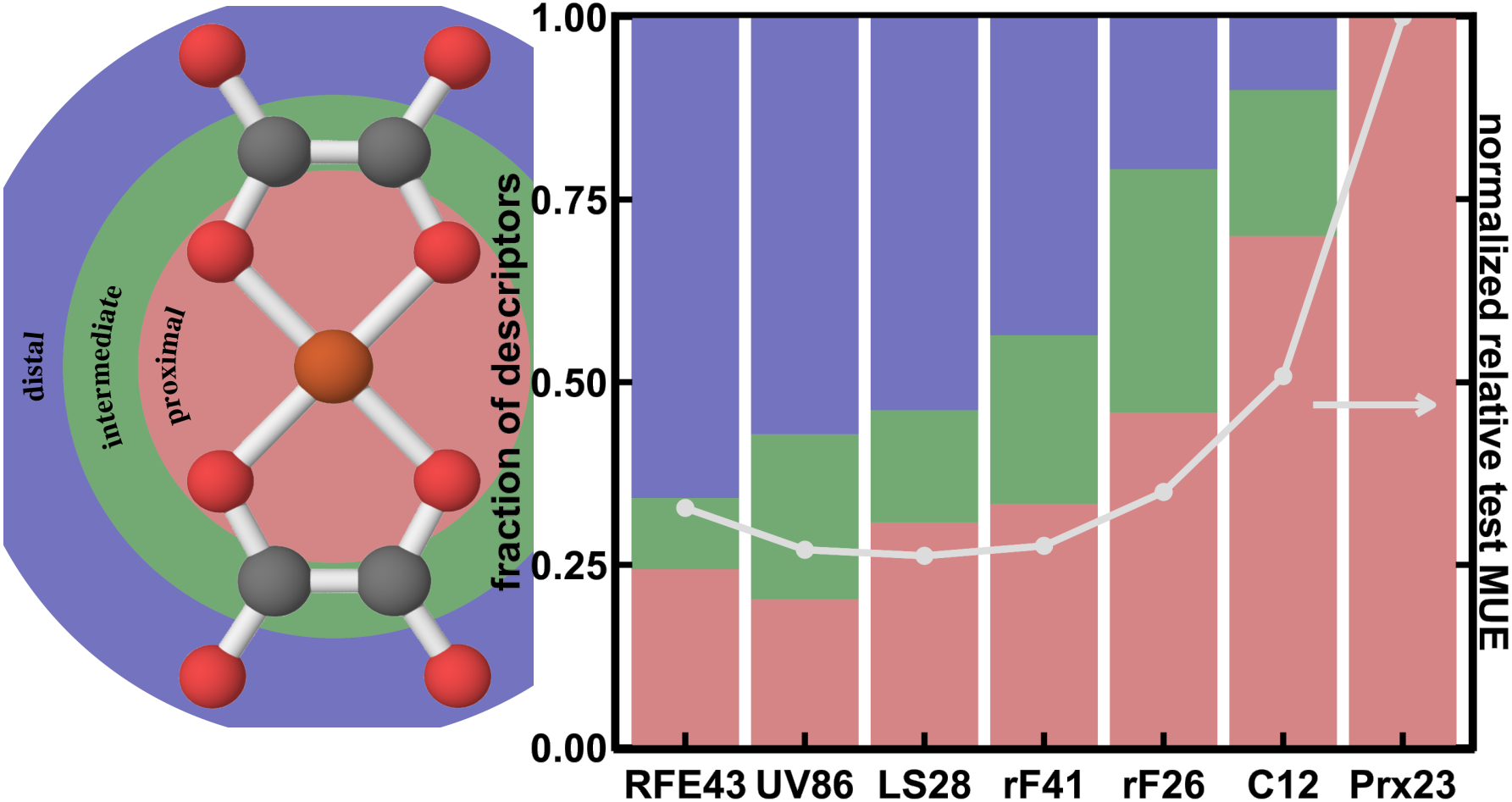
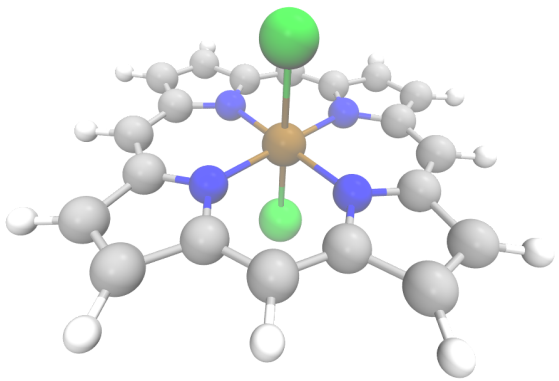
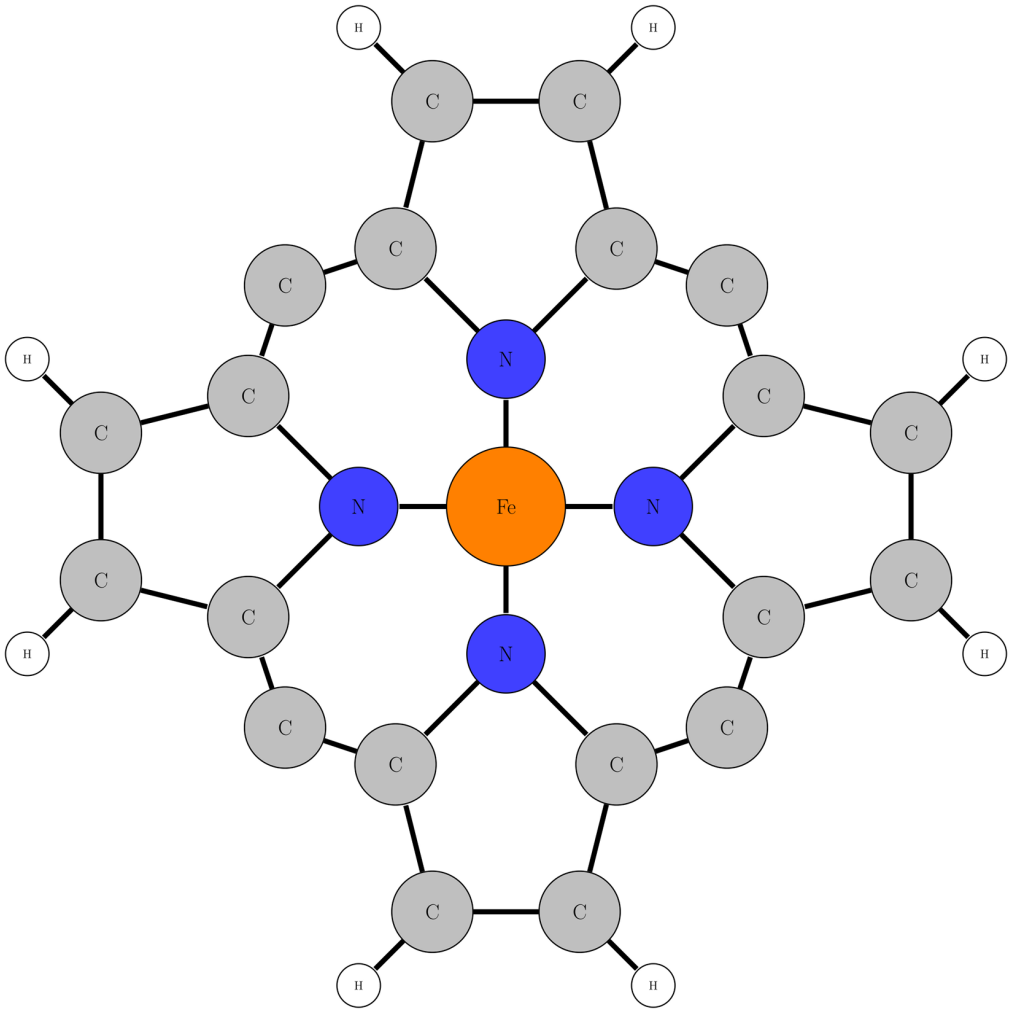



Janet, J.P., and Kulik, H.J., *arXiv*, 1708.06017, 2017.

# How does the information localize?



# How does the information localize?



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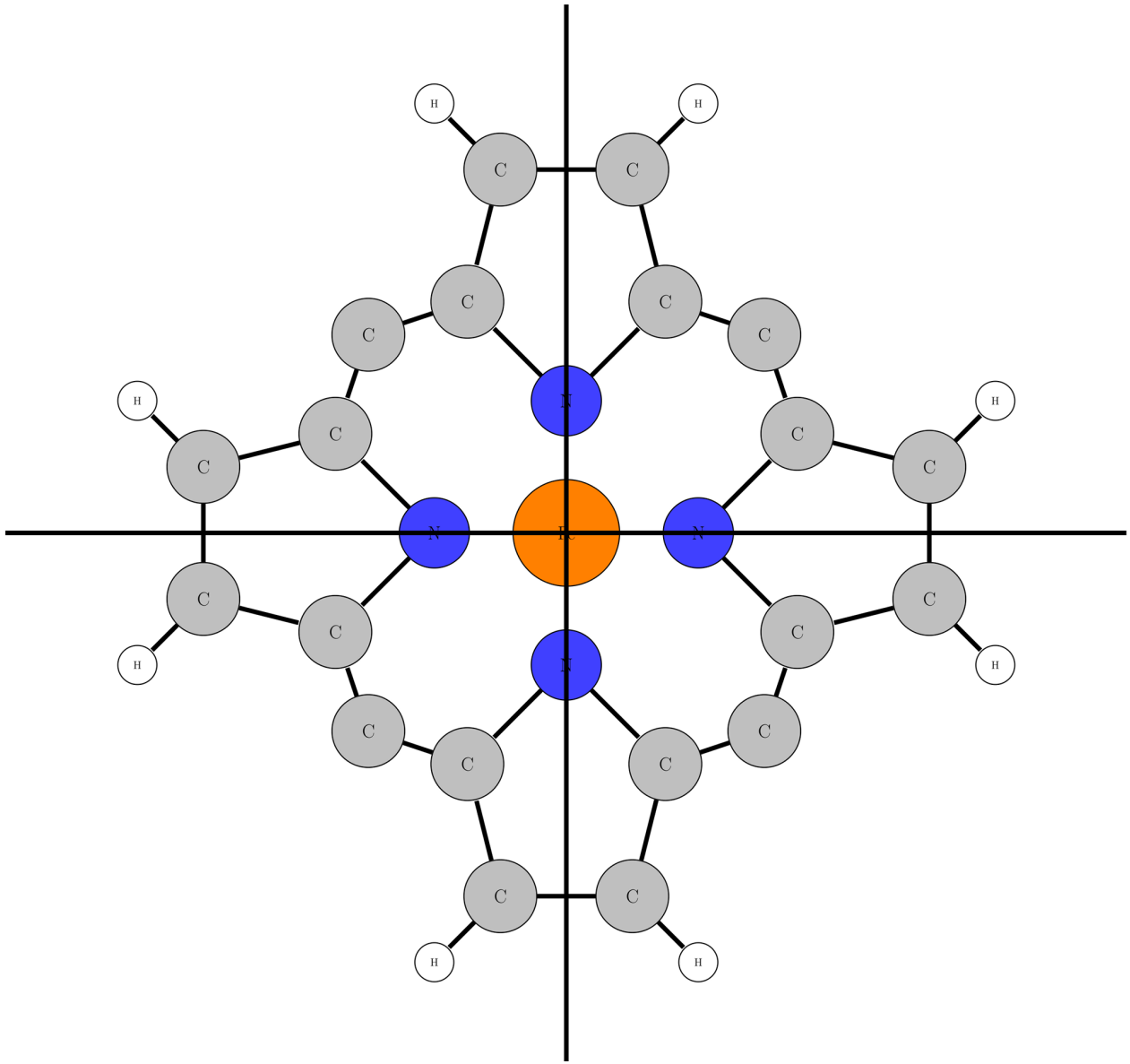
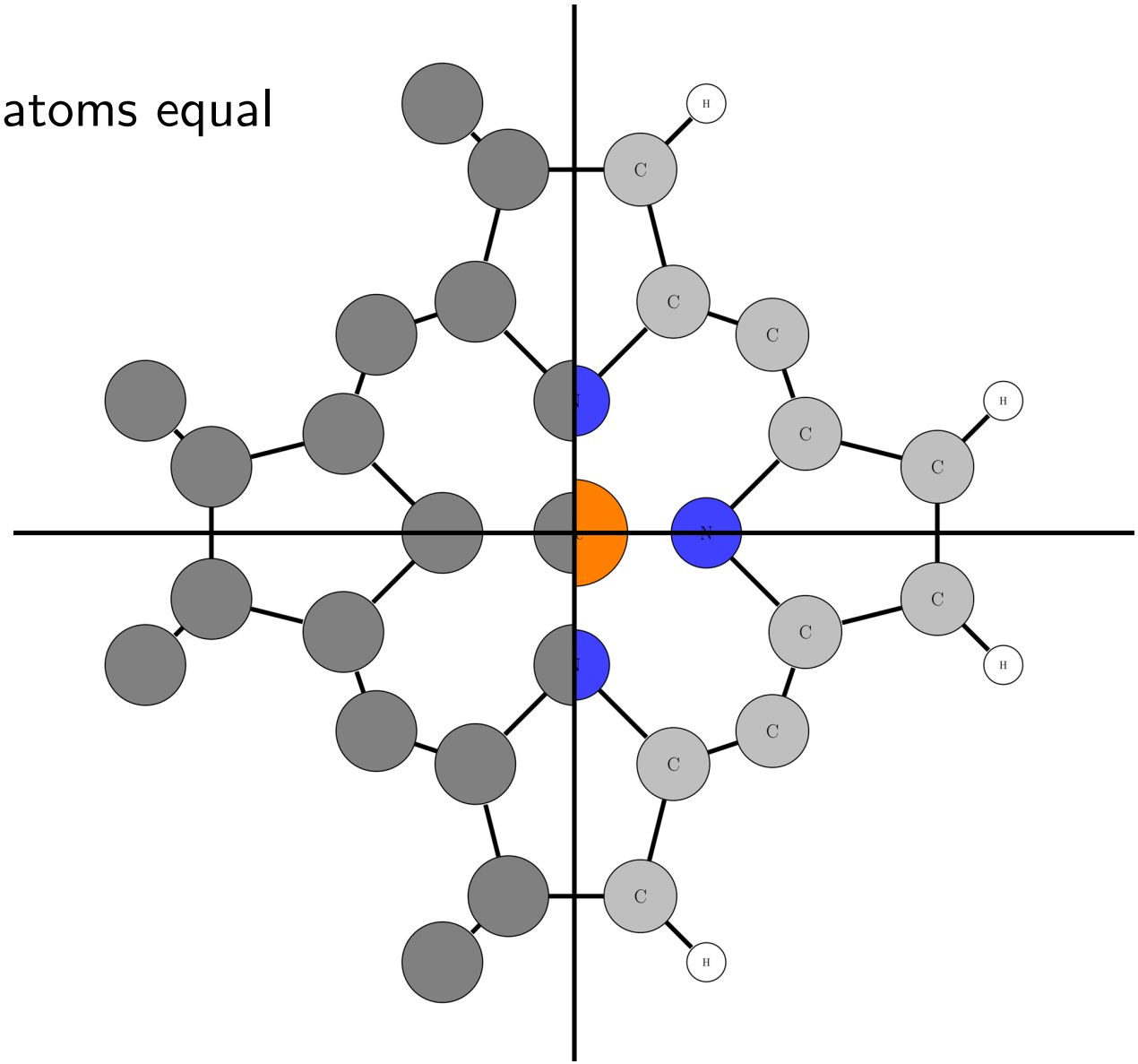


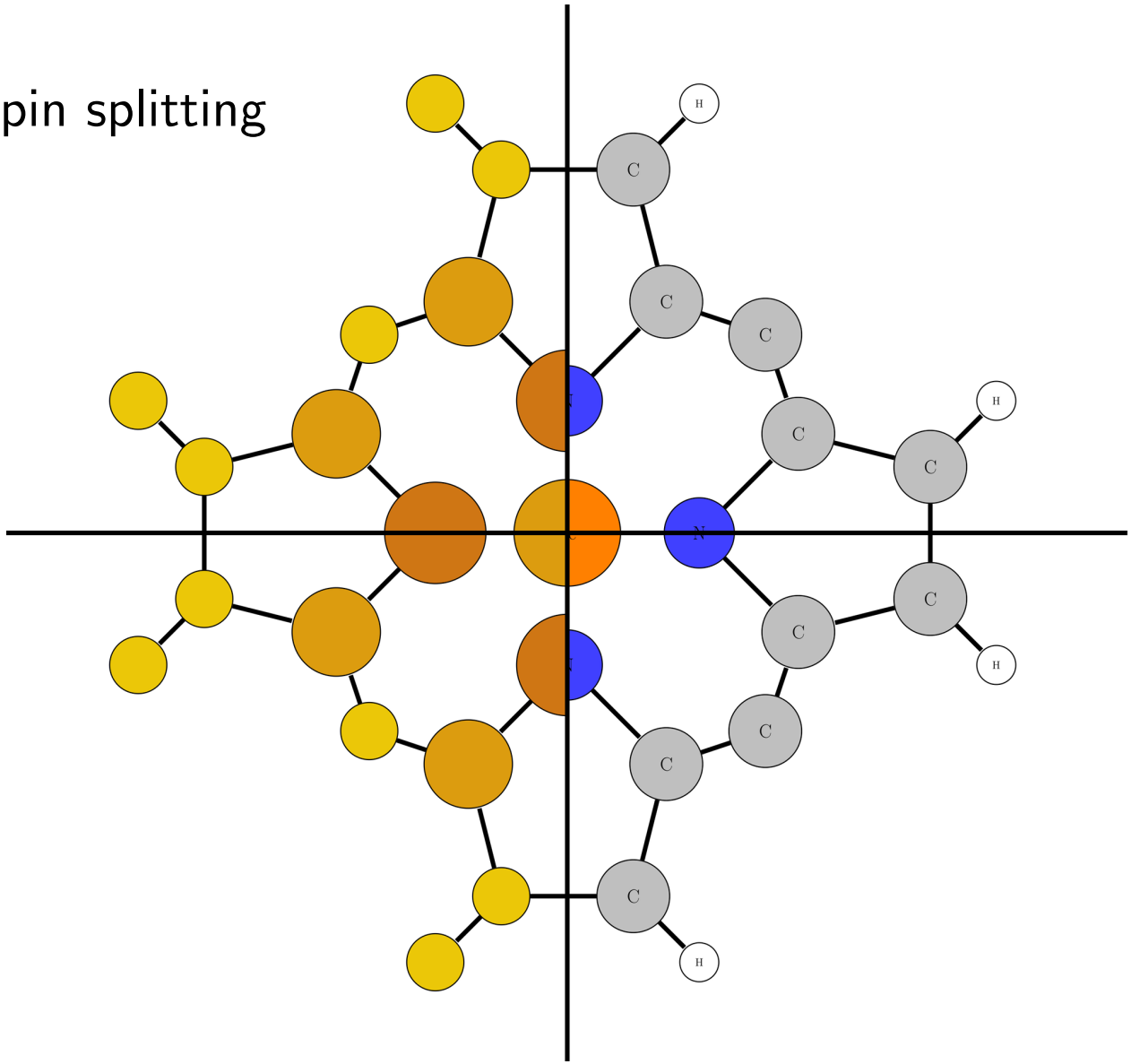
# How does the information localize?

All atoms equal



# How does the information localize?

rF41 - spin splitting



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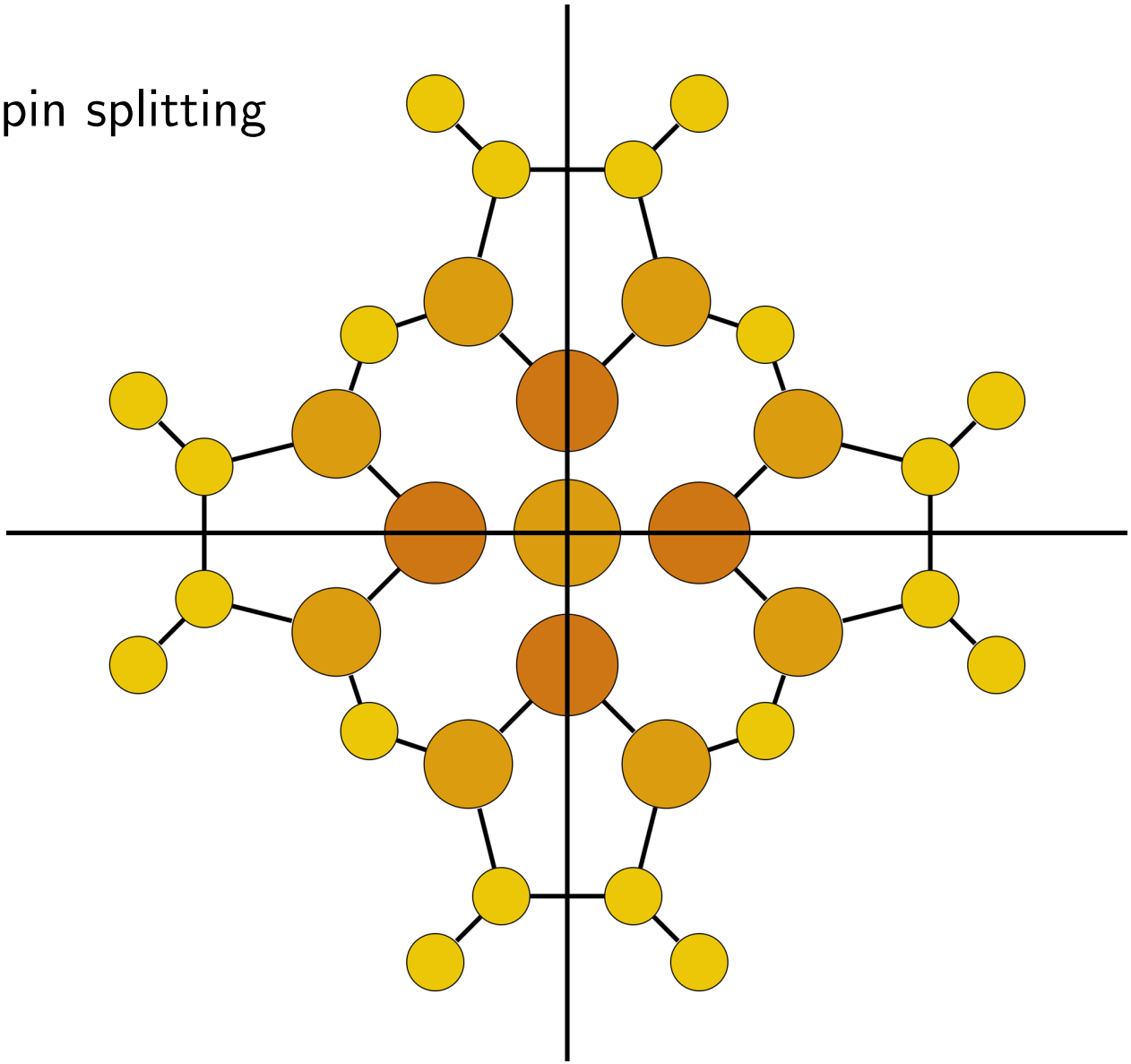
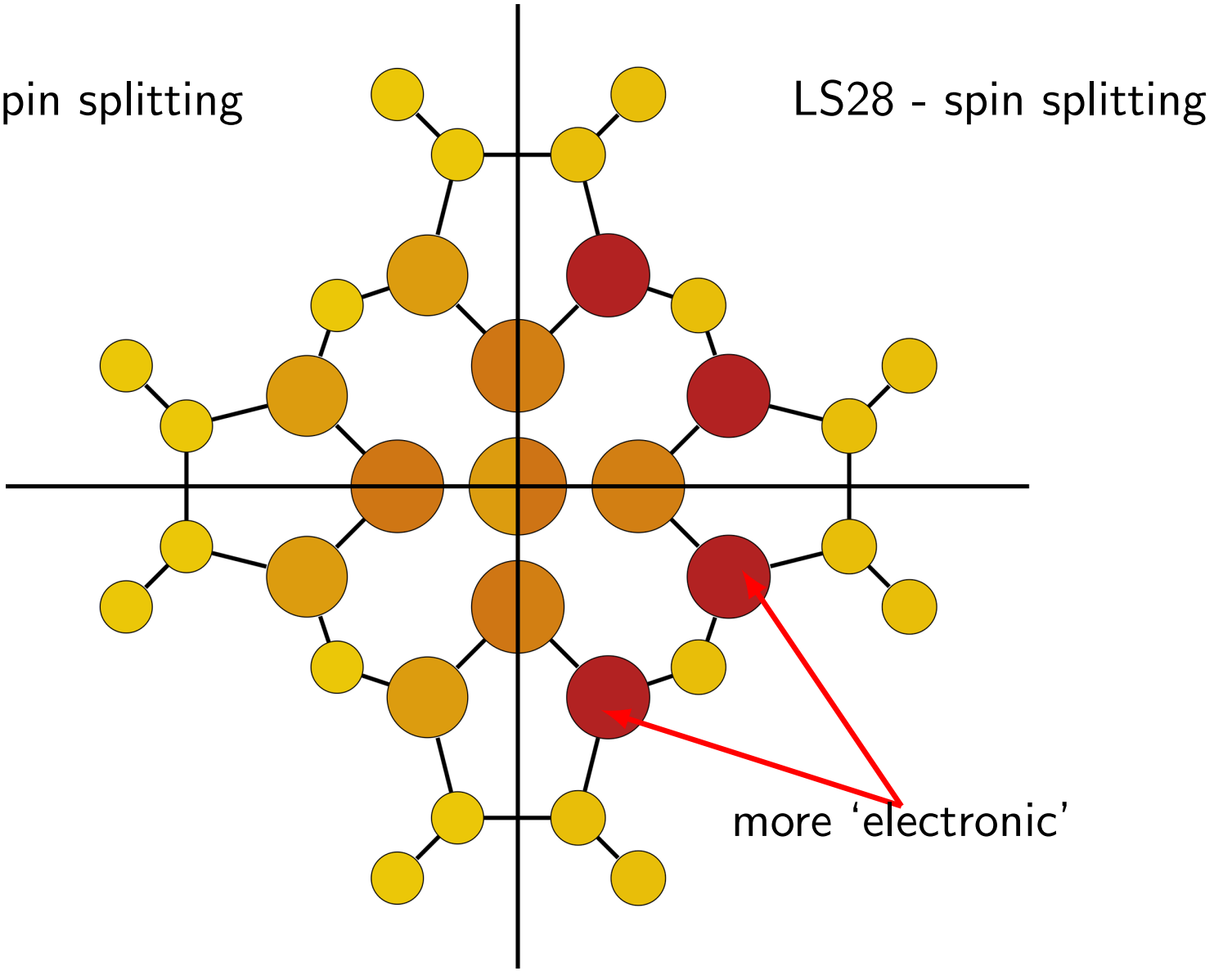
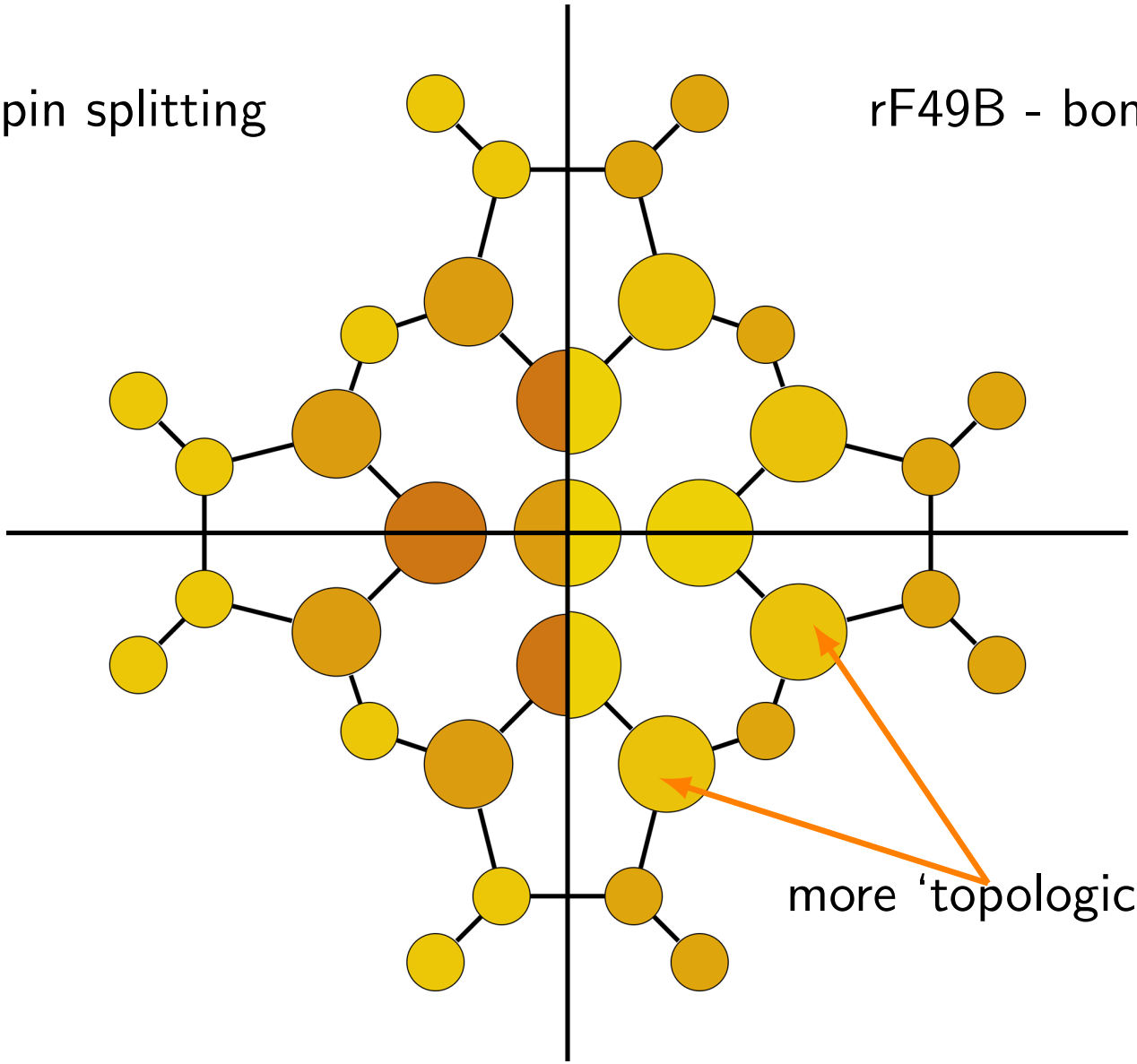
rF41 - spin splitting



# How does the information localize?



# How does the information localize?



# Outlook

Conclusions:

- machine learning TM complexes faces unique challenges but has enormous potential
- descriptors from organic ML are poorly suited to inorganic systems with moderate data
- ACs are a promising starting point for low-cost descriptors
- imbuing 'chemical intuition' to descriptor construction can drastically improve learning

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- ACs are a promising starting point for low-cost descriptors
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Future work:

- use ACs to drive exploration/rational design in TM complex space

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