Hybrid machine-learning and first-principles design for transition metal complexes

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Heather Kulik [1]

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Foundational & Applied Data Science for Molecular and Material Science & Engineering

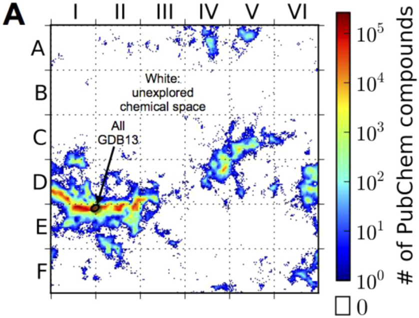
05.22.19

Motivation: chemical discovery

How can we design new materials using computers?

The space of possible chemistries is incredibly vast, with $\mathcal{O}(10^{60})$ small organic molecules.

All potentially undiscovered medicines, catalysts and materials are somewhere, out in this huge space.



Virshup *et al.*, *J. Am. Chem. Soc.*, 135(19): 7296–7303, 2013.

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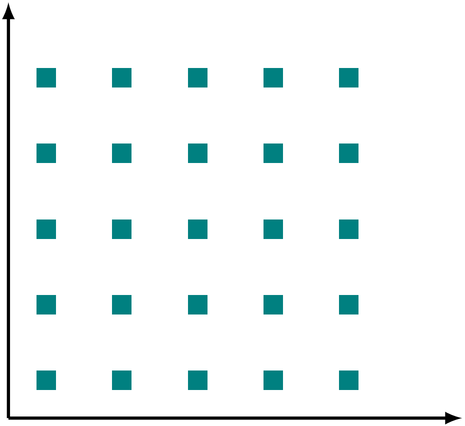

Chemical Design Space C_f

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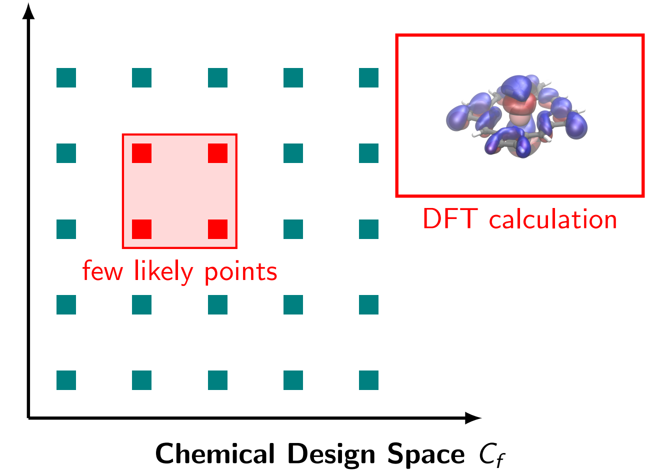

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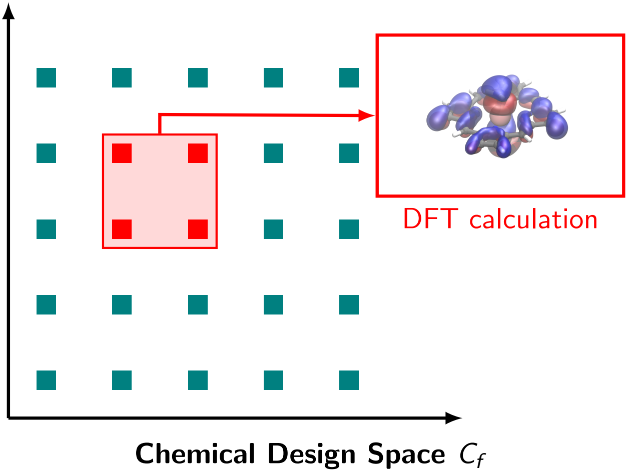

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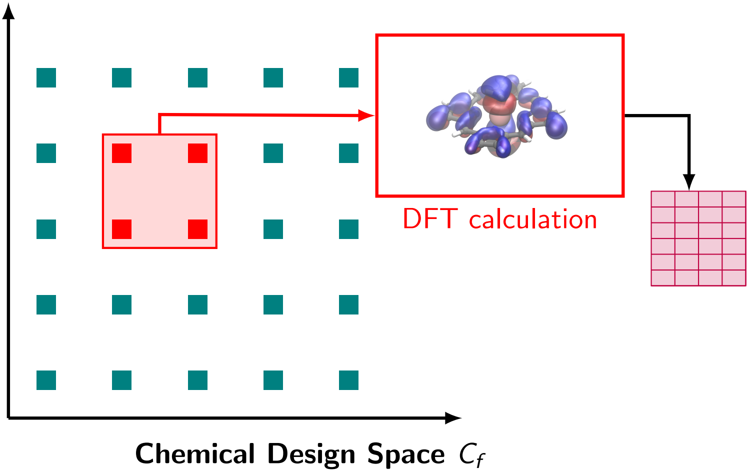

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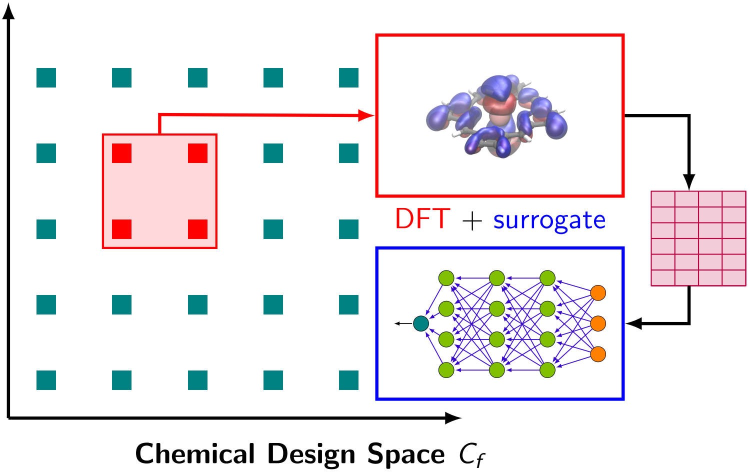

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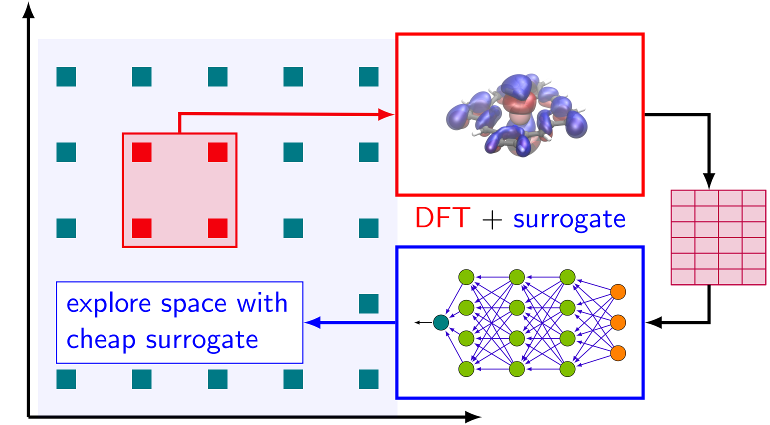

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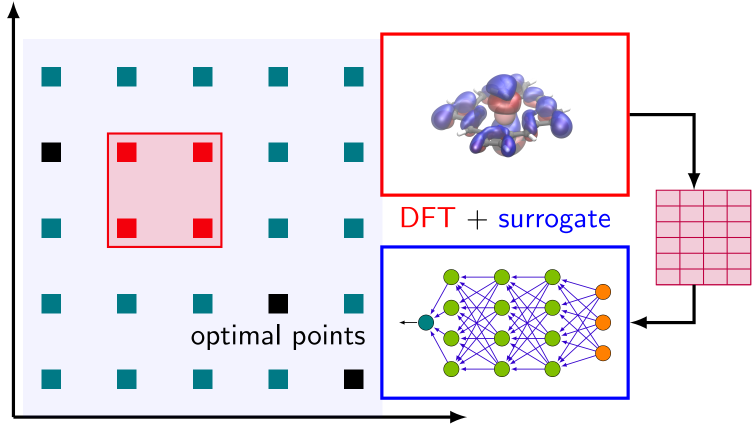

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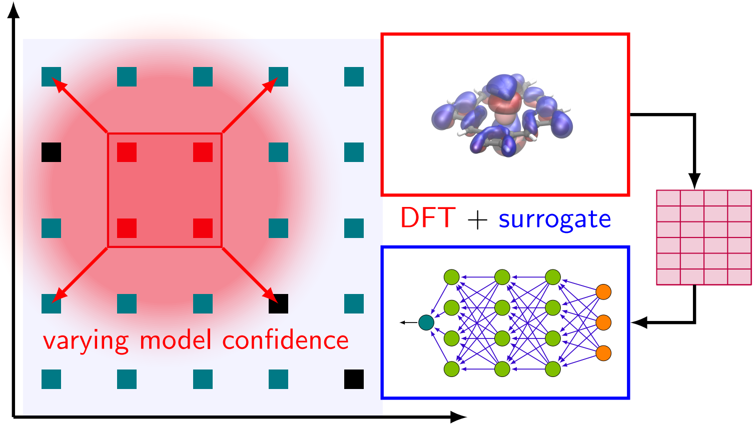

Chemical Design Space C_f

Transition metal complexes

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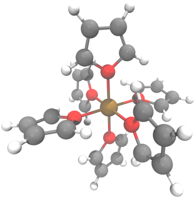

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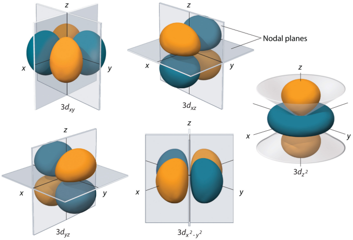

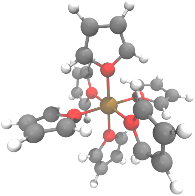

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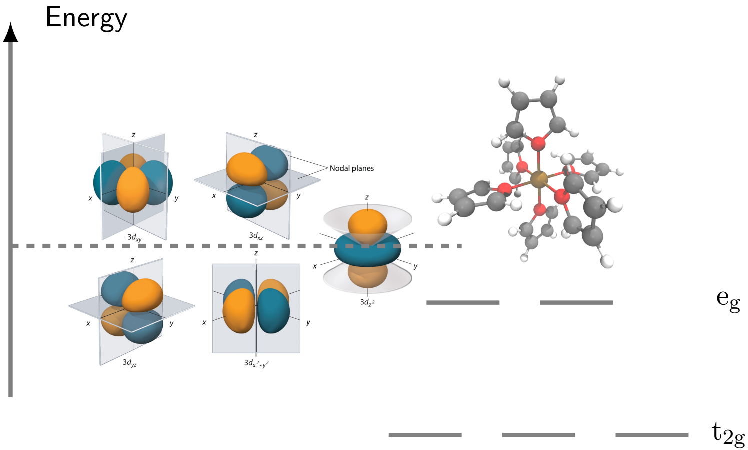

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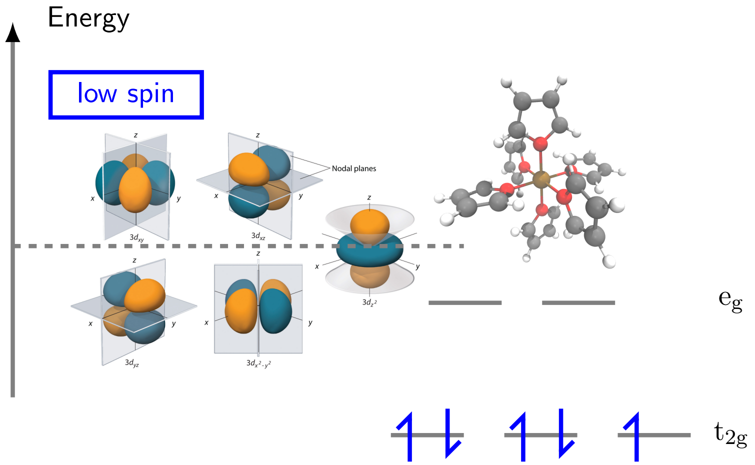

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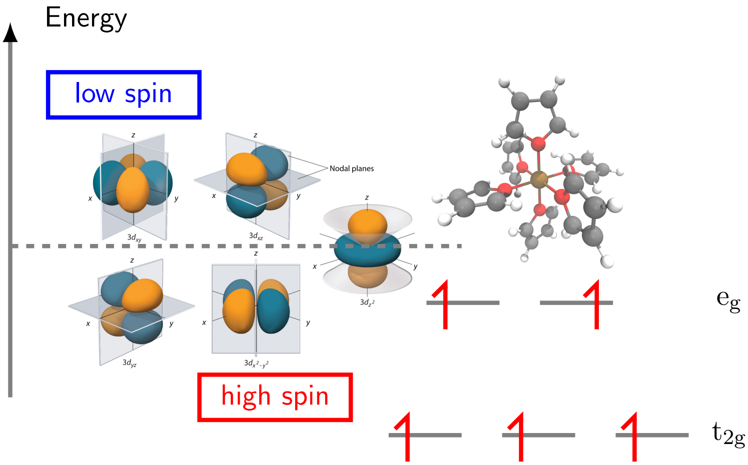

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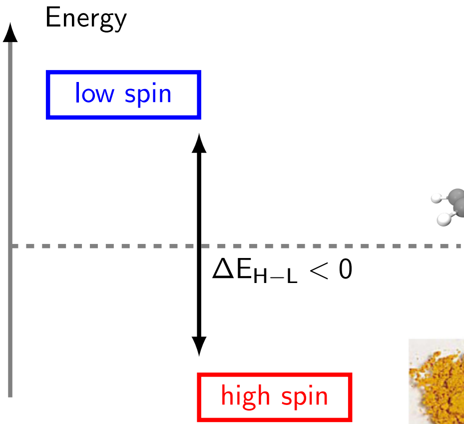

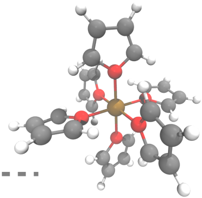

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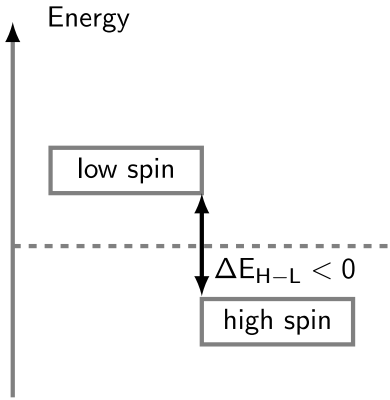

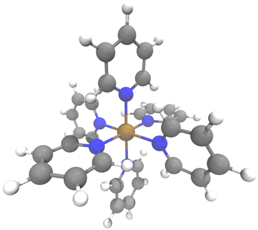

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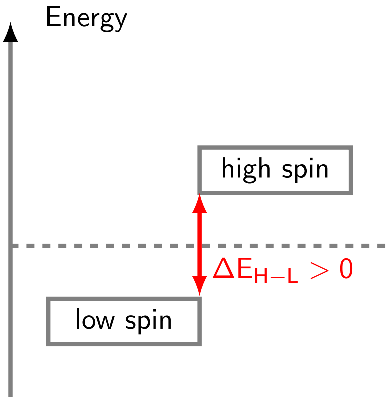

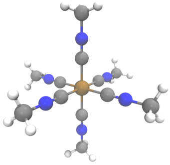

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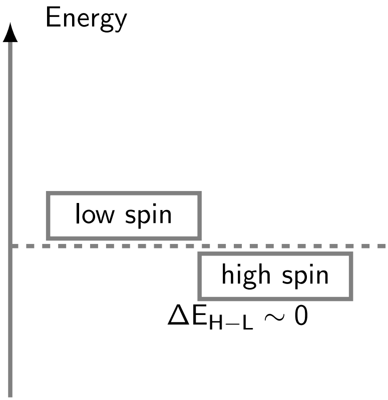

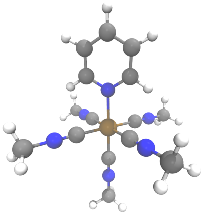

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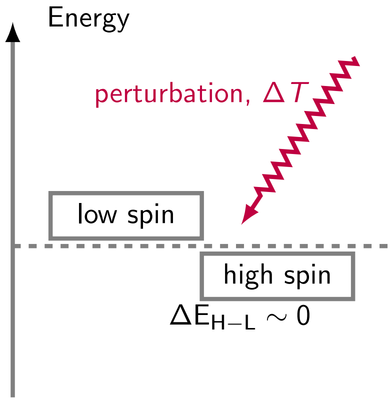

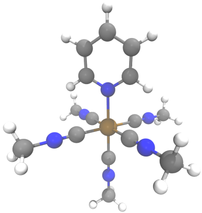

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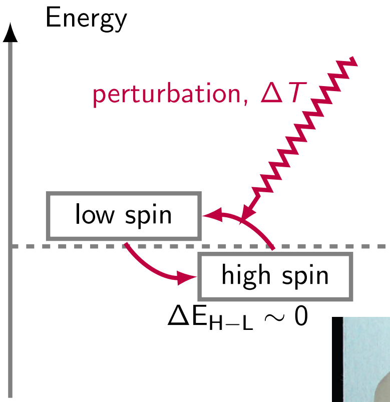
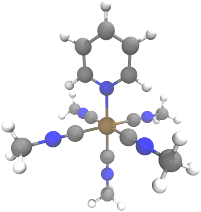
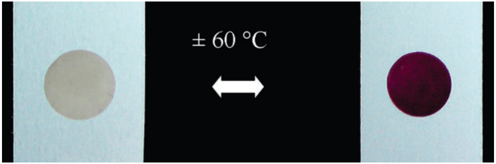






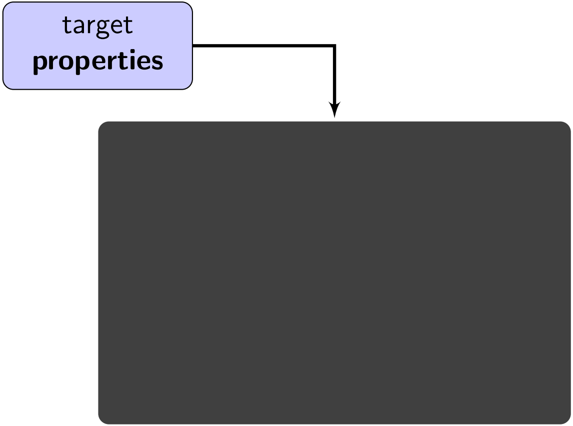
Seredyuk, M *et al.*, *Chem. Mater.*, 18(10):2513–2519, 2006.

The dream of automated design

The dream of automated design

target
properties

The dream of automated design



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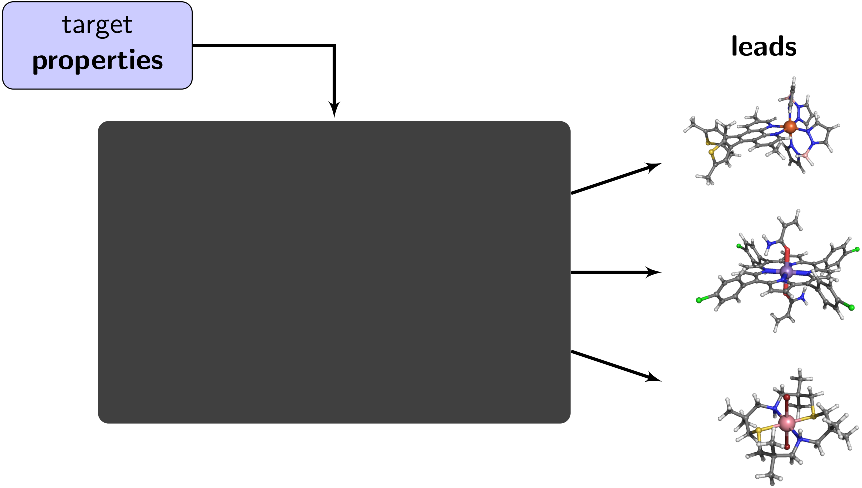

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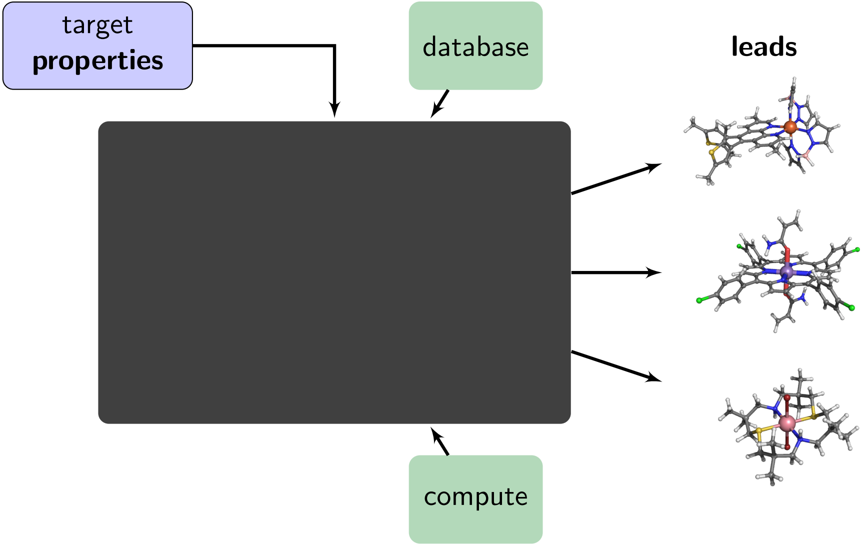

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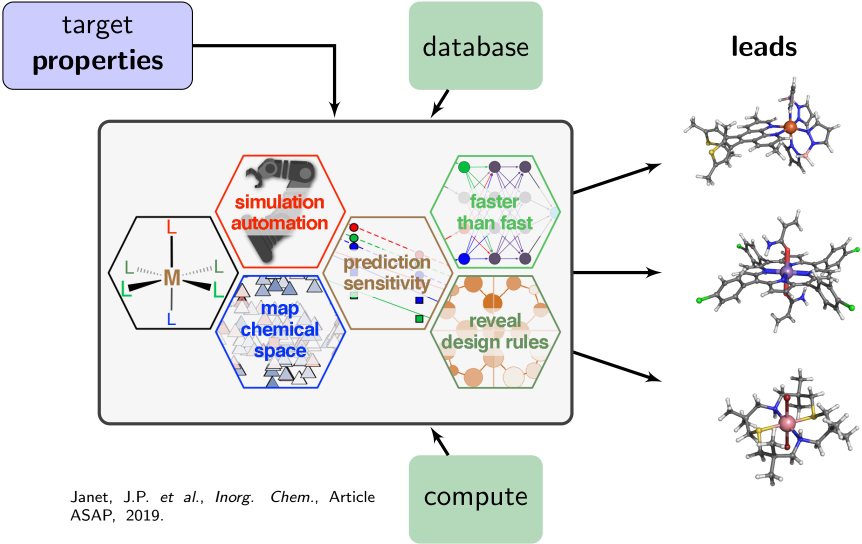

Janet, J.P. *et al.*, *Inorg. Chem.*, Article ASAP, 2019.

Quantum simulation of TM complexes

train on $\sim$ 100–2000 DFT calculations:

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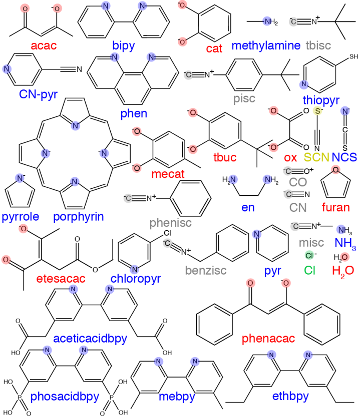

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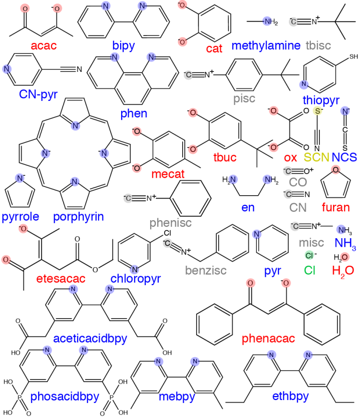

Cr	Mn	Fe	Co

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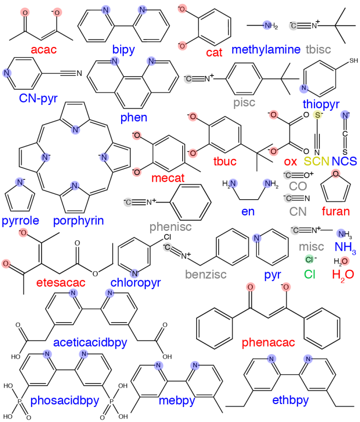

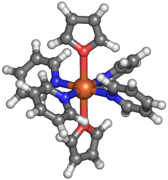

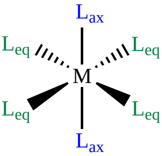

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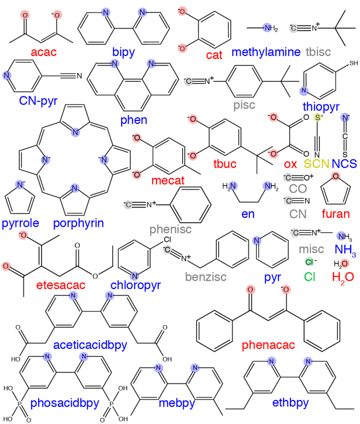

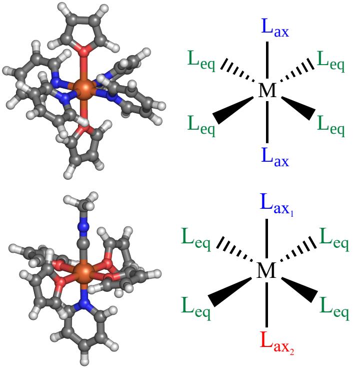

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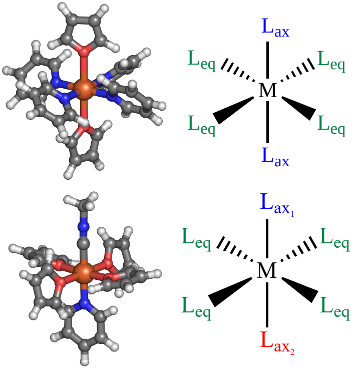
Details:
B3LYP-like DFT
gas phase optimizaton
LANL2DZ/6-31G*
high- and low-spin
M(II)/(III)



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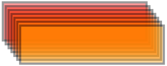
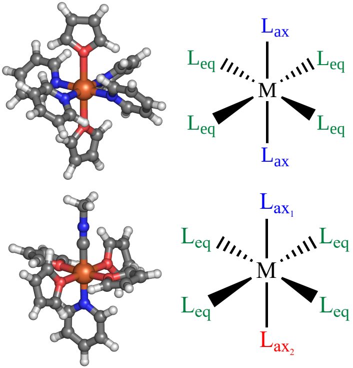
HF exchange varied 0–30%





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Modeling of TM complexes with heuristic representations

First attempt using simple features inspired by inorganic chem:

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metal properties

identity

Fe(II)

oxidation state

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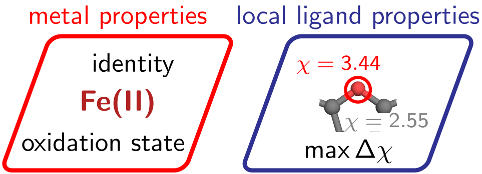

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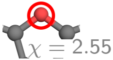
oxidation state

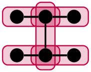
local ligand properties

$\chi = 3.44$



max $\Delta\chi$

global properties



Kier index

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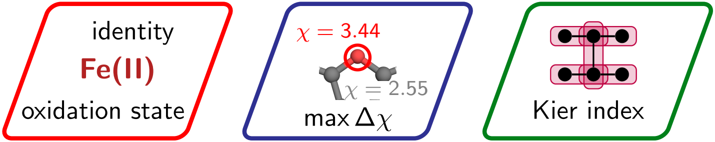

mixed continous discrete ligand-centered: **MCDL-25**

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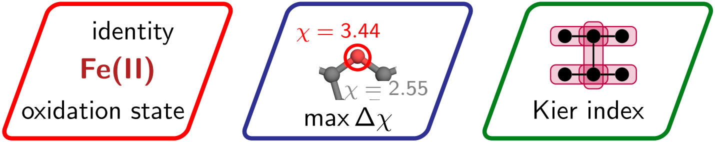

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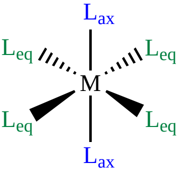

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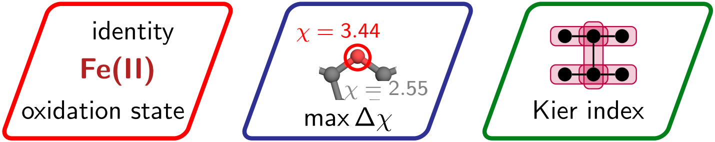
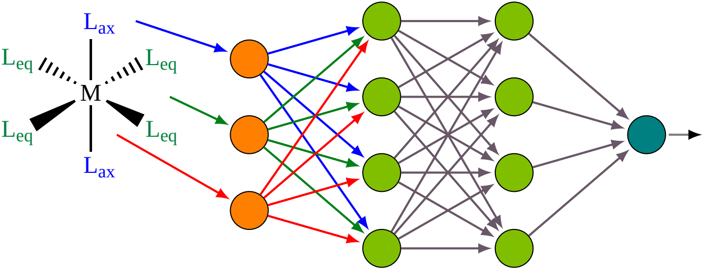
metal properties — local ligand properties — global properties



fully-connected 2-layer ANN, dropout regularization



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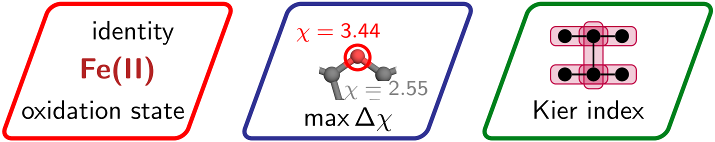
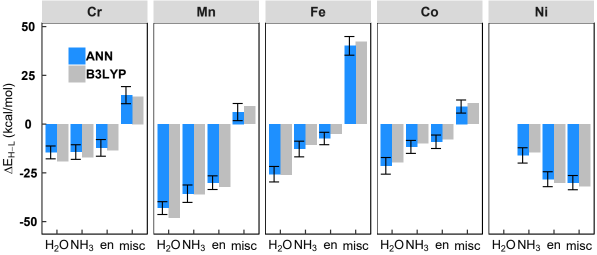
metal properties | local ligand properties | global properties





Janet, J.P. and Kulik, H.J., *Chem. Sci.*, 8:5137–5152, 2017.

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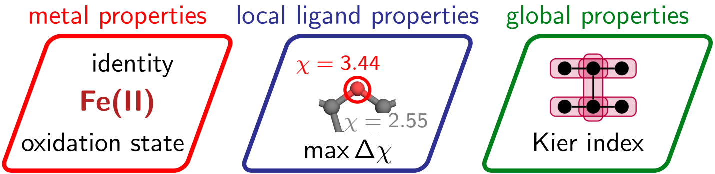

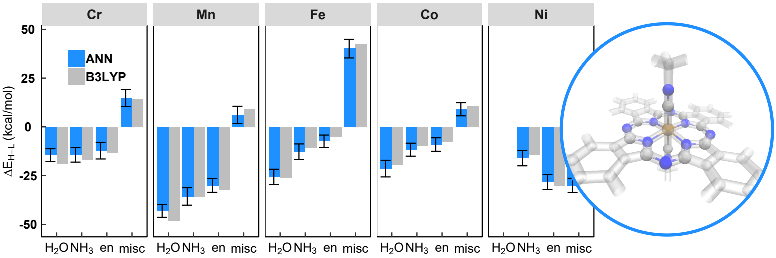

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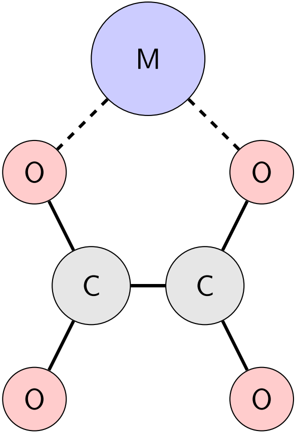
New descriptors – RACs

Proposed a new graph-based set of descriptors for TM complexes[1]



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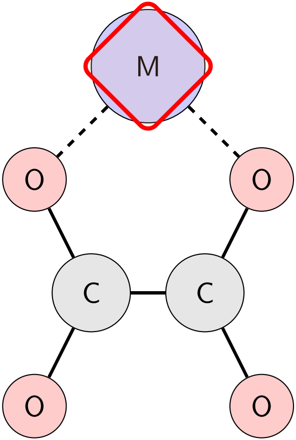

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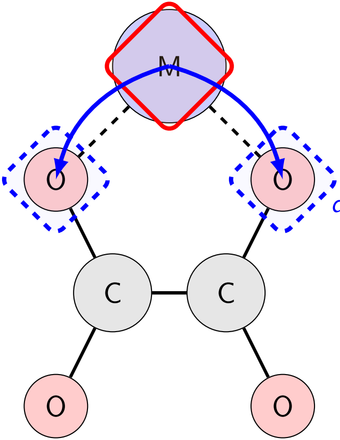

$d_1 : \sum_{M,O} Z_M Z_O$

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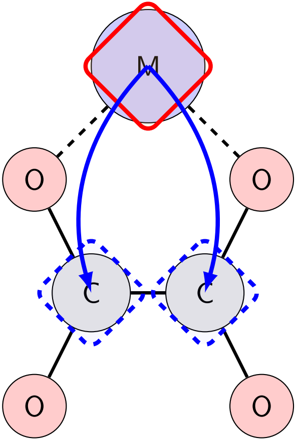

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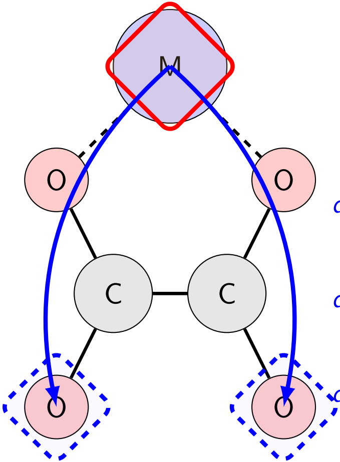

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$$d_3 : \sum_{M,O} Z_M Z_O$$

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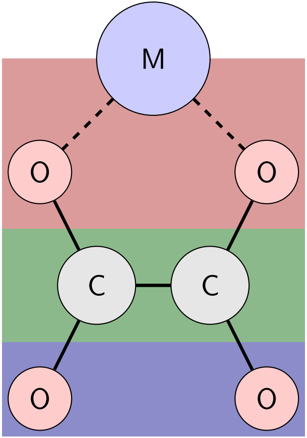

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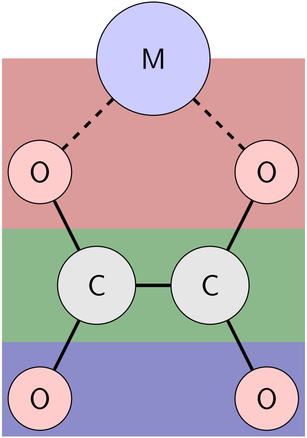
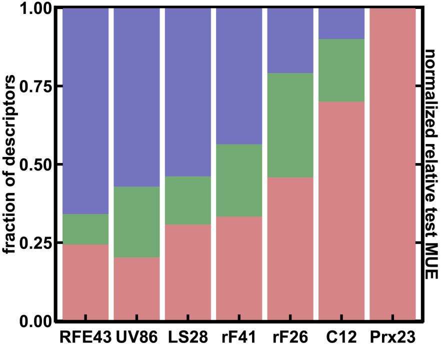


KRR models of spin splitting energies:



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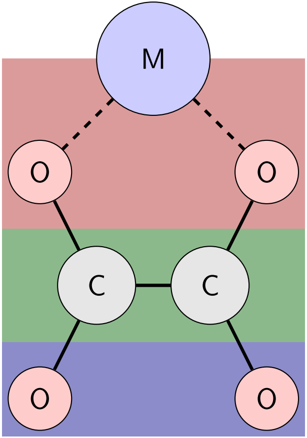
New descriptors – RACs

Proposed a new graph-based set of descriptors for TM complexes[1]



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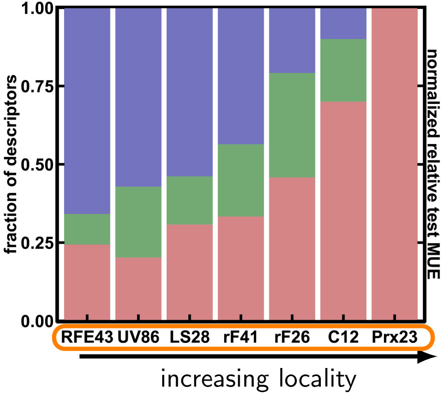

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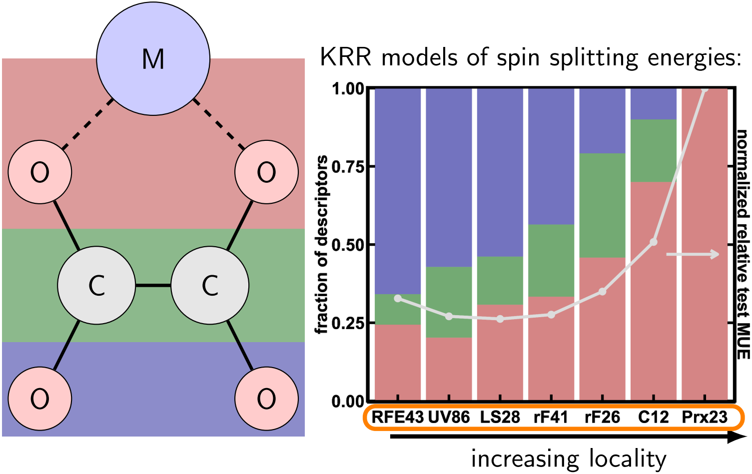

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Extension to catalysis

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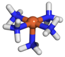

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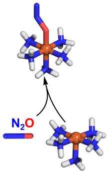

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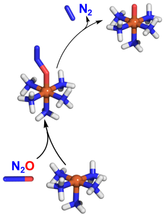

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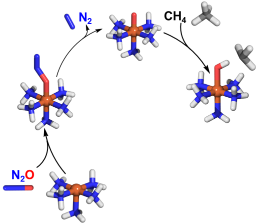

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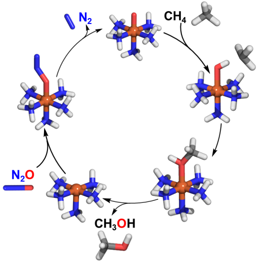

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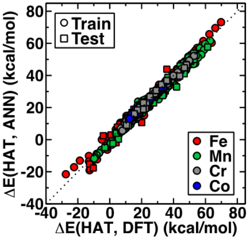

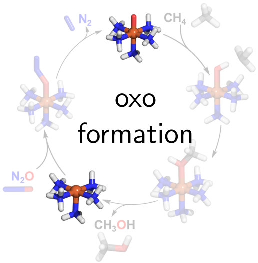

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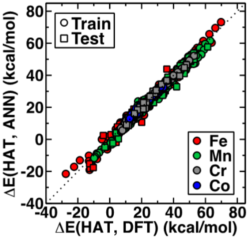

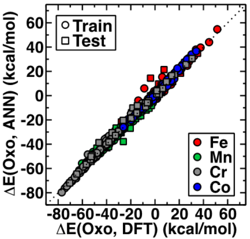

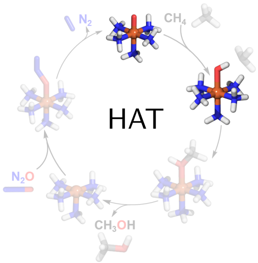

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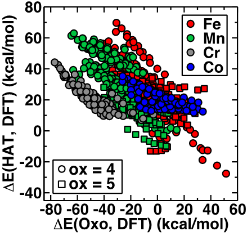

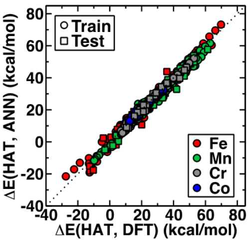

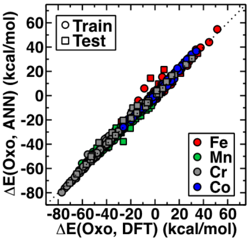

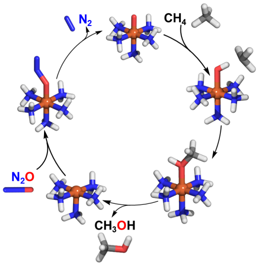

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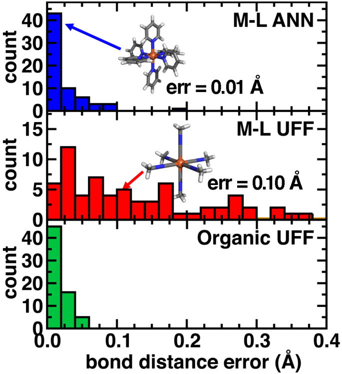

we can predict
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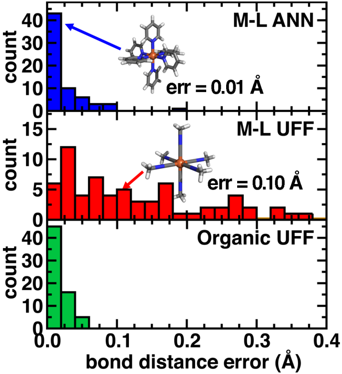
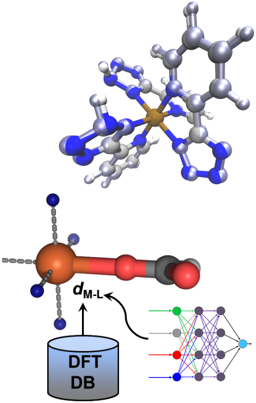


we can predict bond lengths and use this to intialize new calculations



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Beyond prediction: live job management

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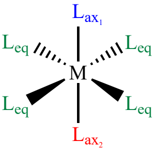

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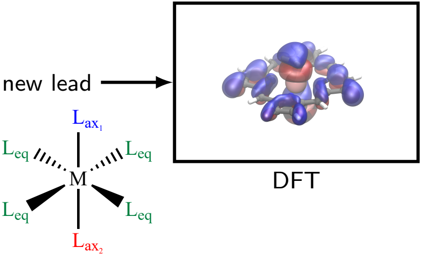

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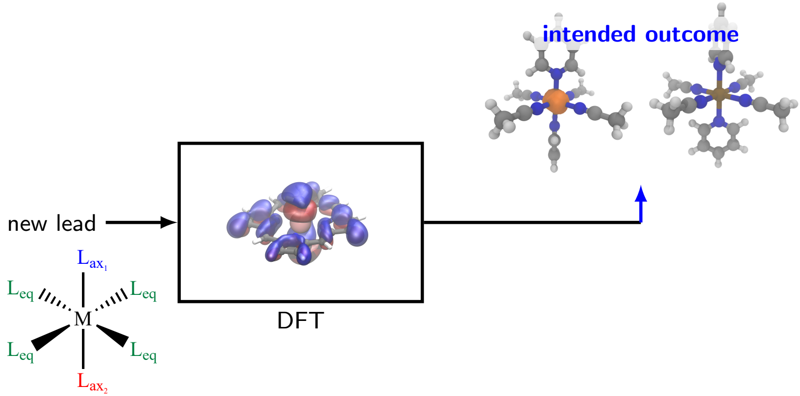

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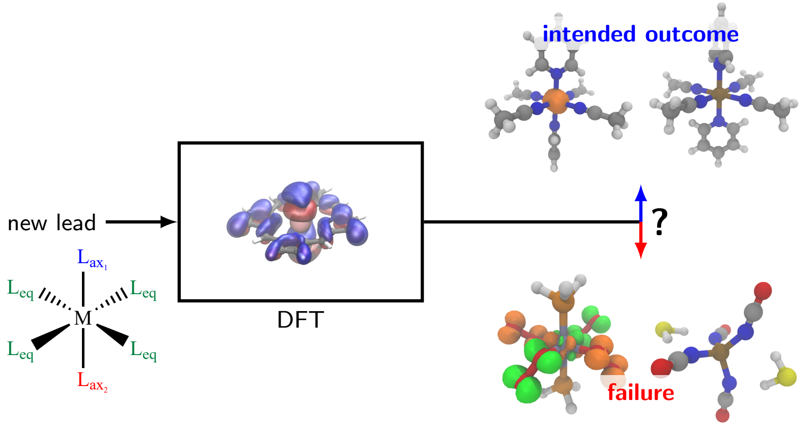

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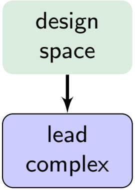

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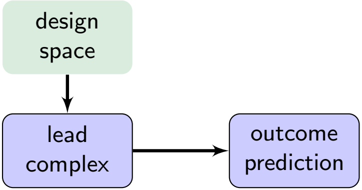

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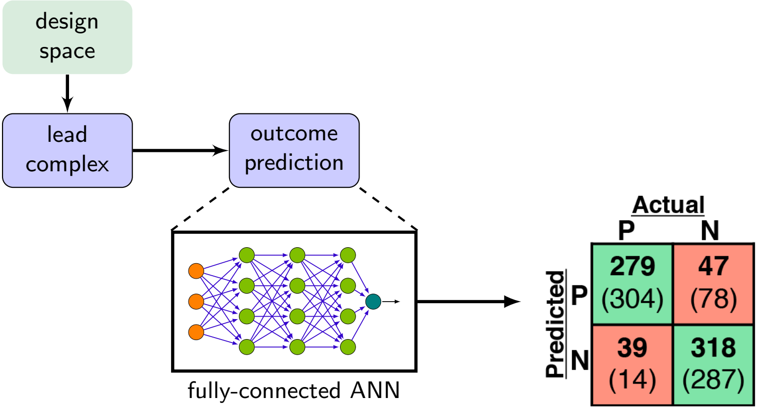

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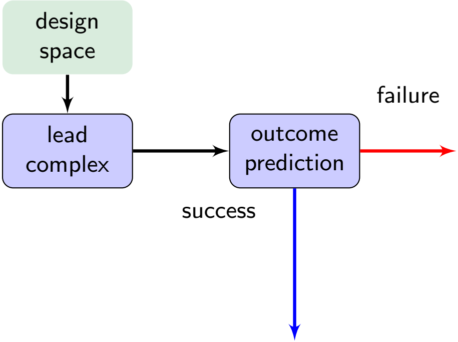

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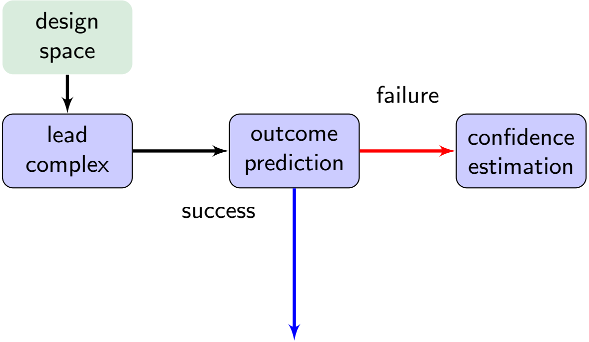

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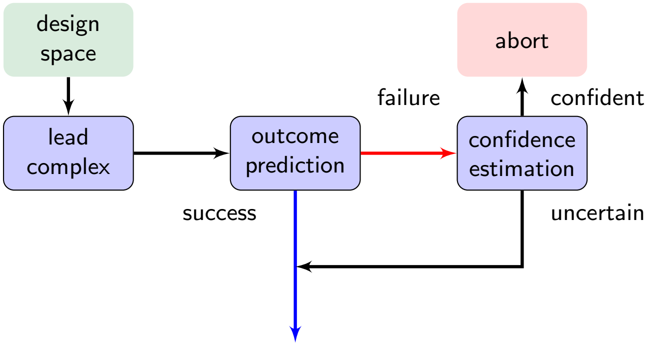

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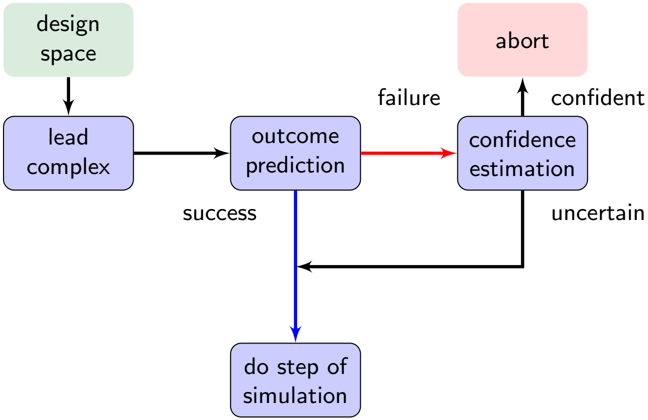

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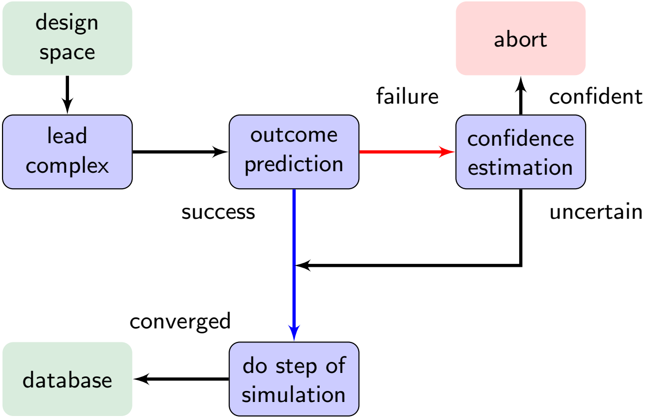

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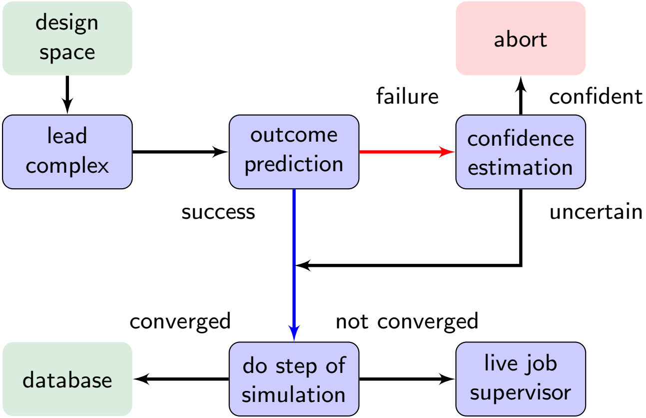

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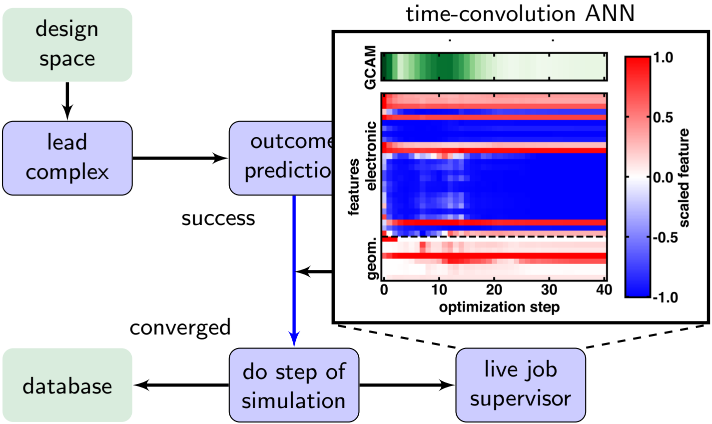

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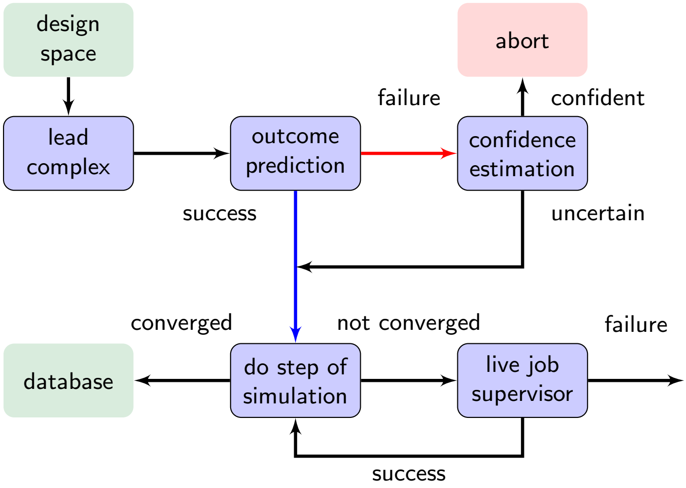

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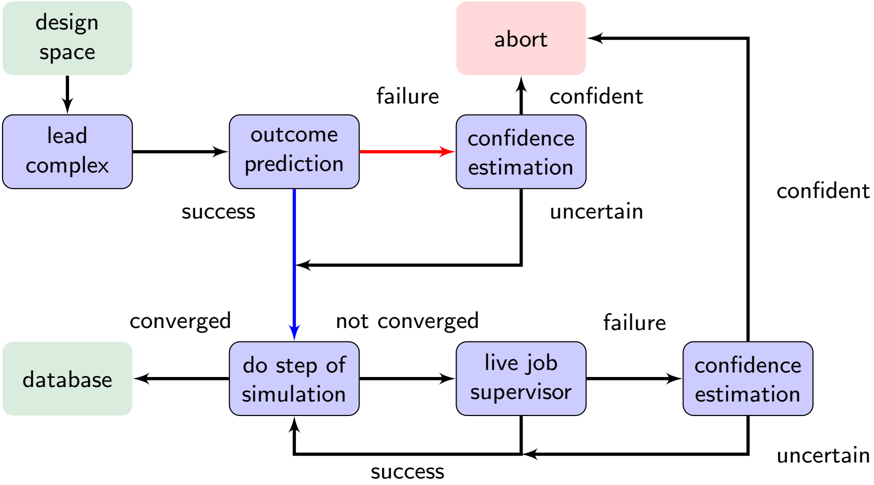

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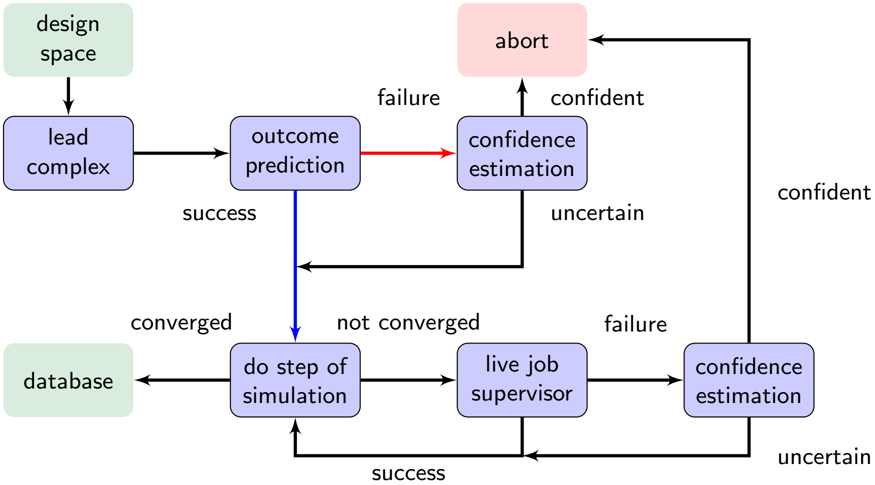

This leads to about **40% time savings** and can abort almost all failures.

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Model transferability

Test-set performance is not necessarily a good metric for general transferability[1]:

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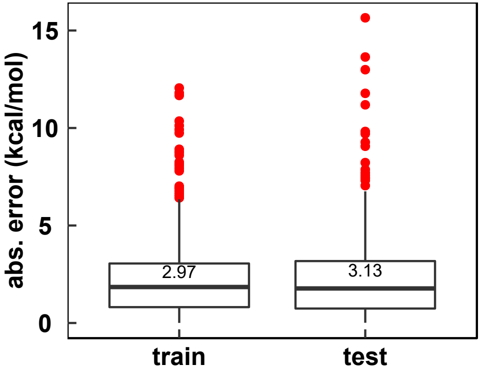

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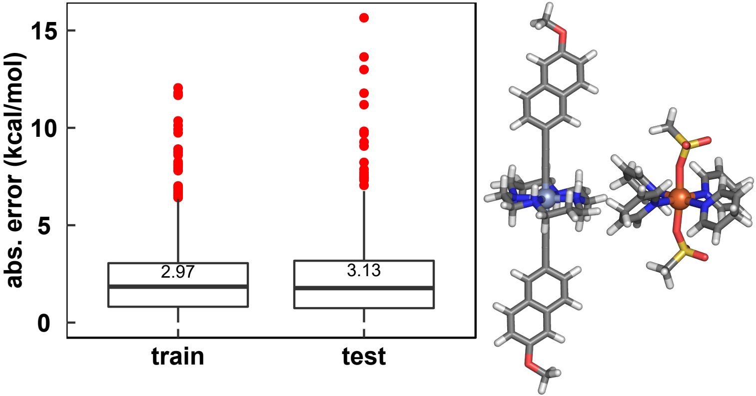

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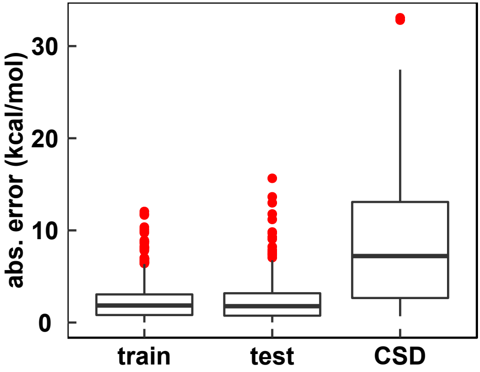

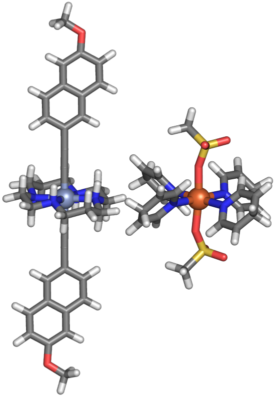

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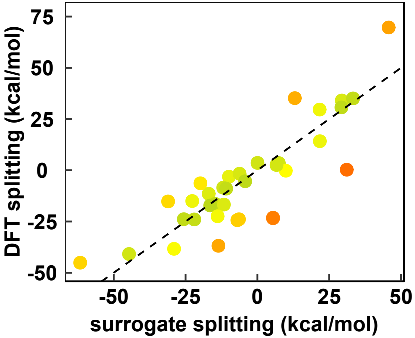

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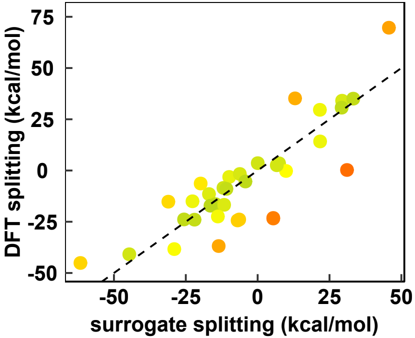

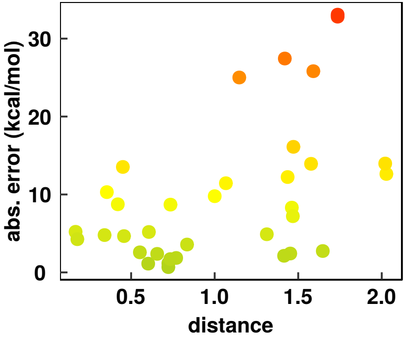

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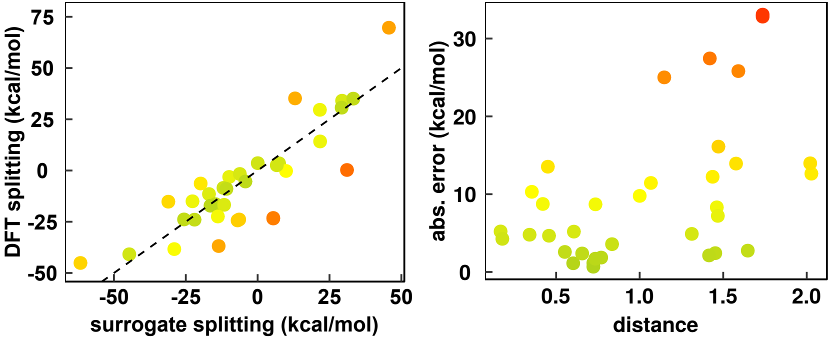

using simple distance worked pretty well!

More complex representations

Results are worse for more complex representations[1]:

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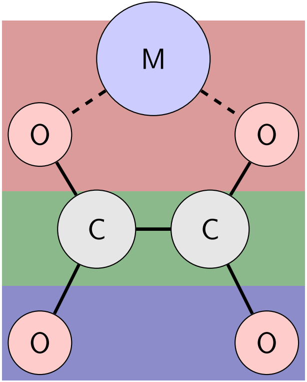

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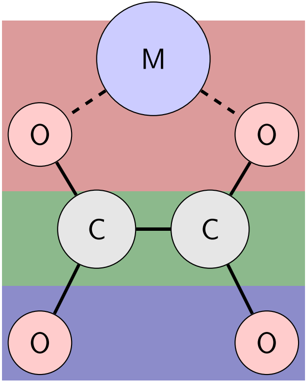

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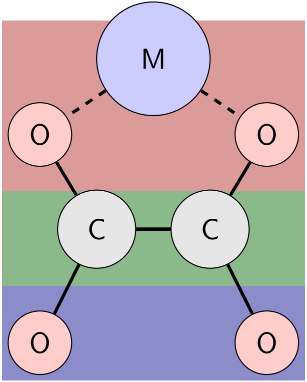

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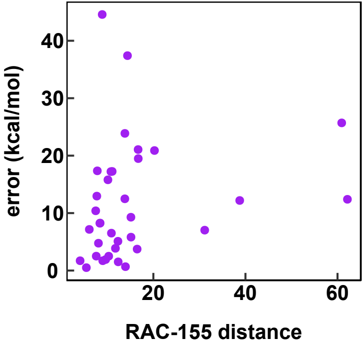

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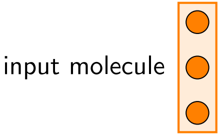

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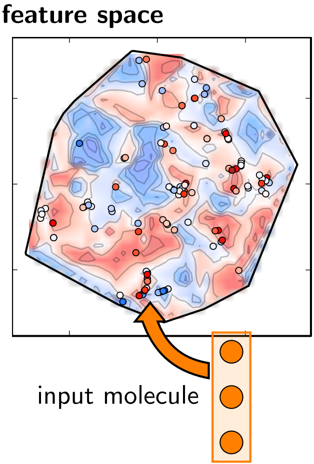

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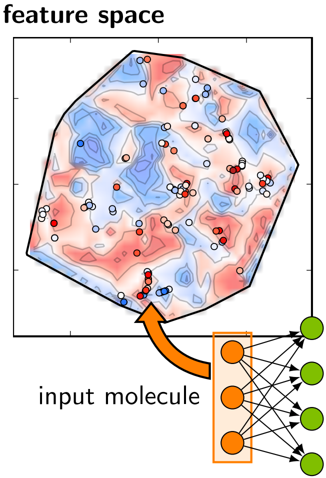

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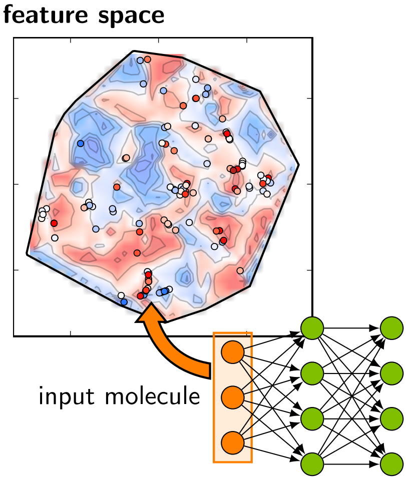

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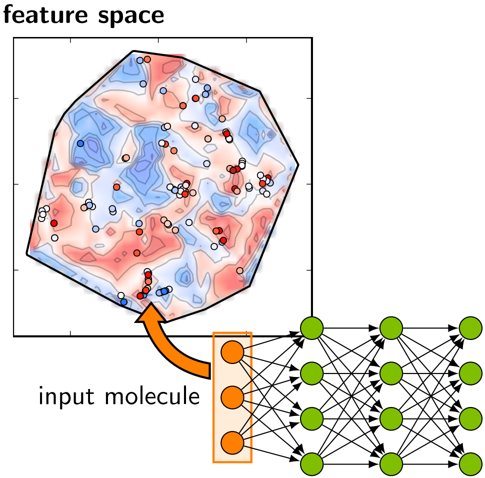

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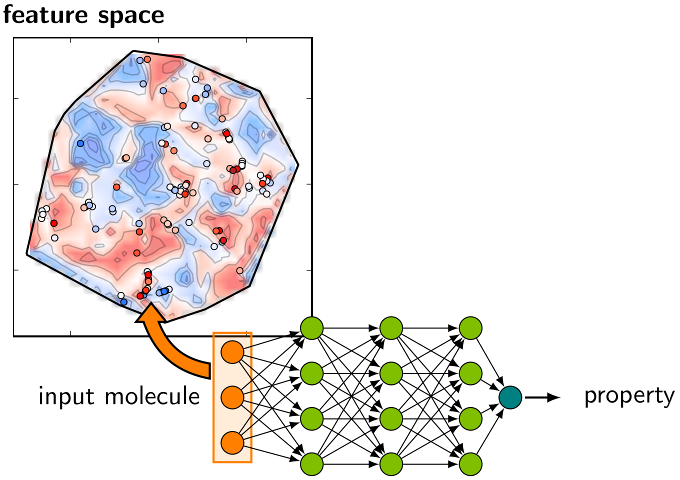

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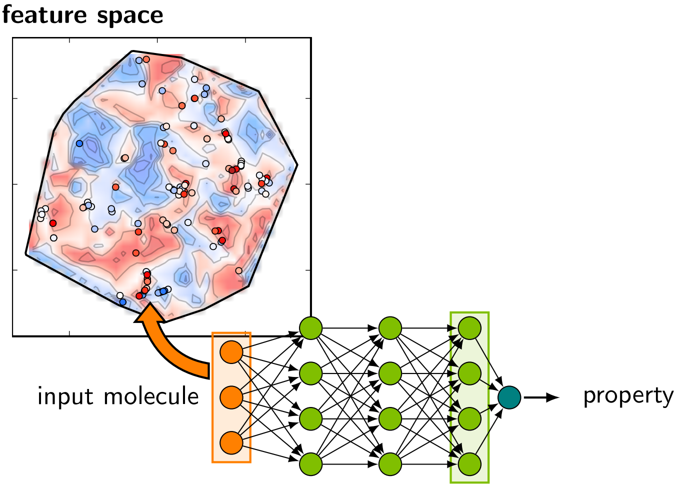

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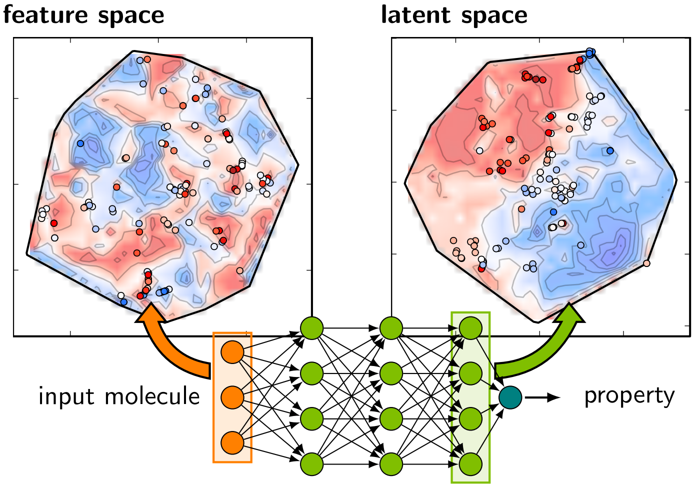

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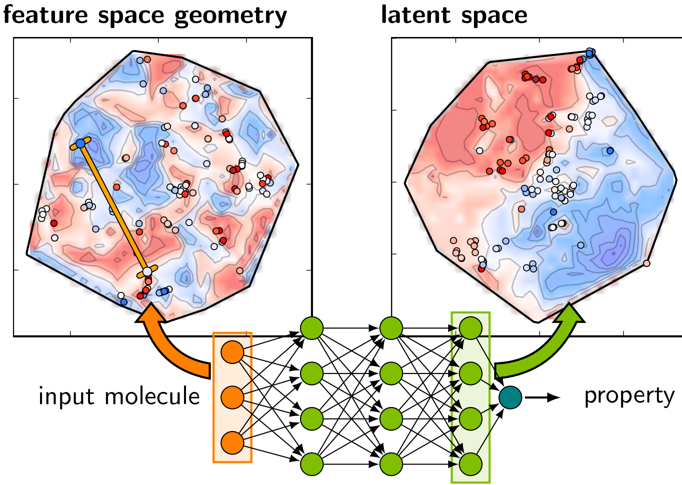

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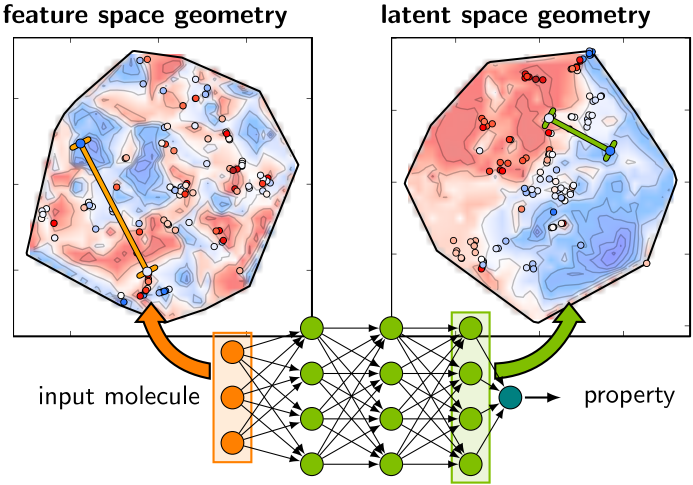

Other UQ metrics

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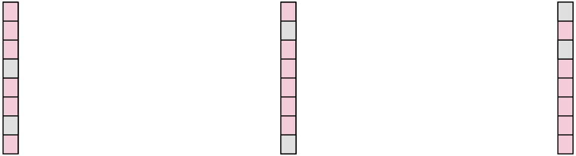

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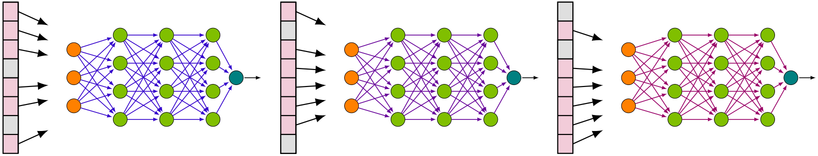

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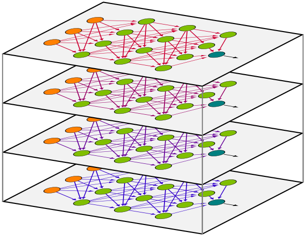

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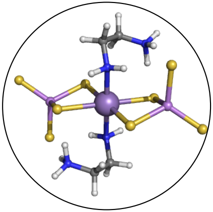

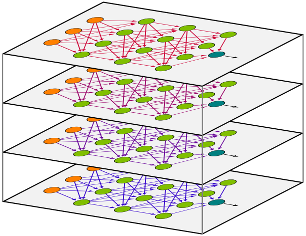

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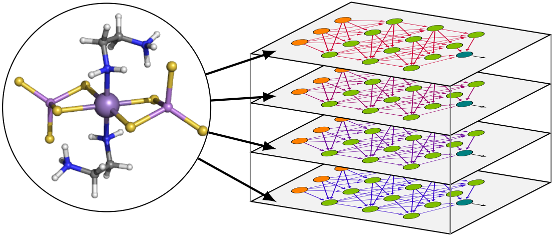

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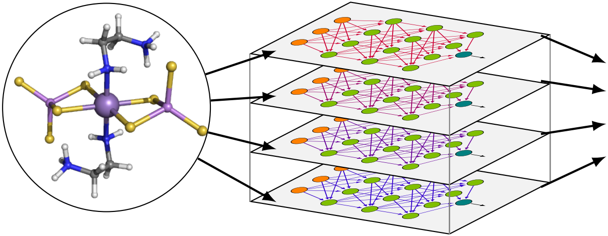

$\bar{y}(x^*), \sigma^2(x^*)$

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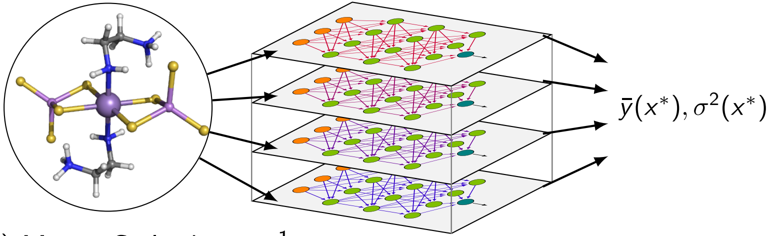

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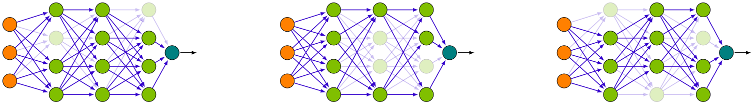

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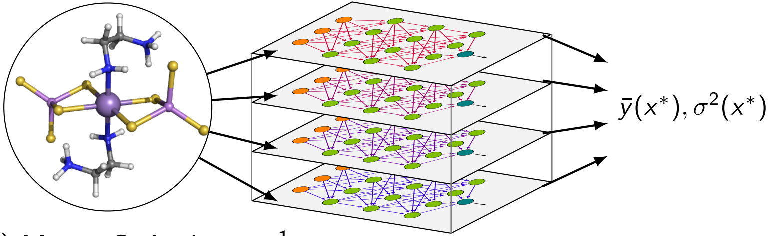

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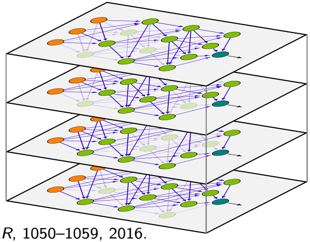

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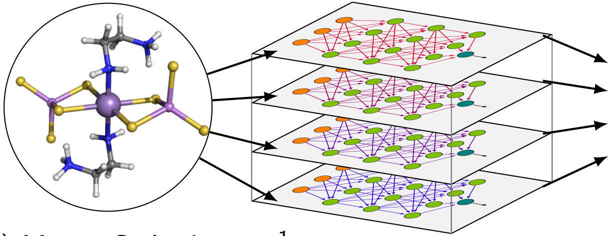

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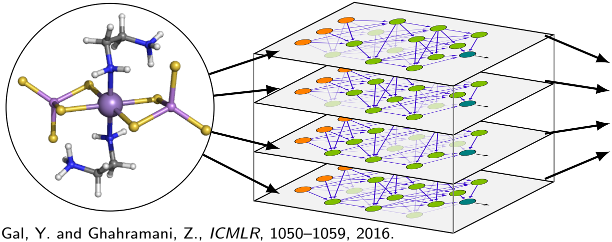

$\bar{y}(x^*), \sigma^2(x^*) + \tau$

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A challenging test case: CSD II

'Out-of-distribution' test: spin-splitting energies of 116 structures from the CSD, from training-like to very different.

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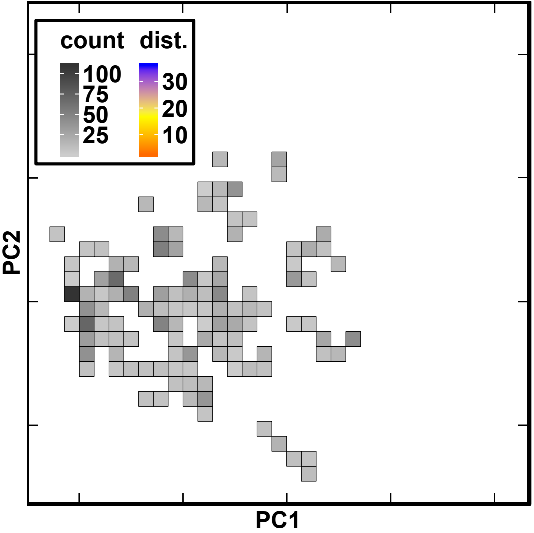

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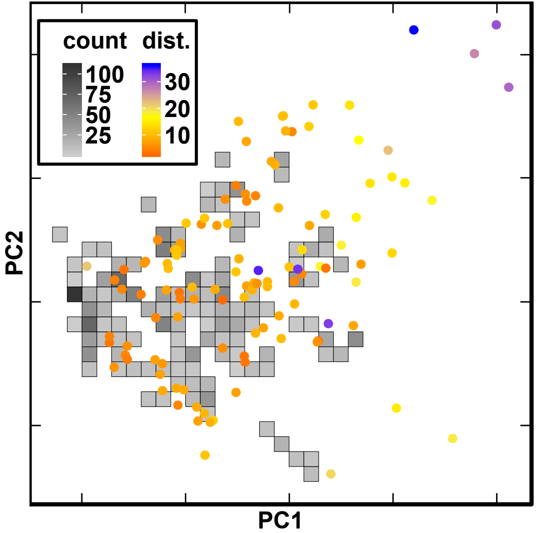

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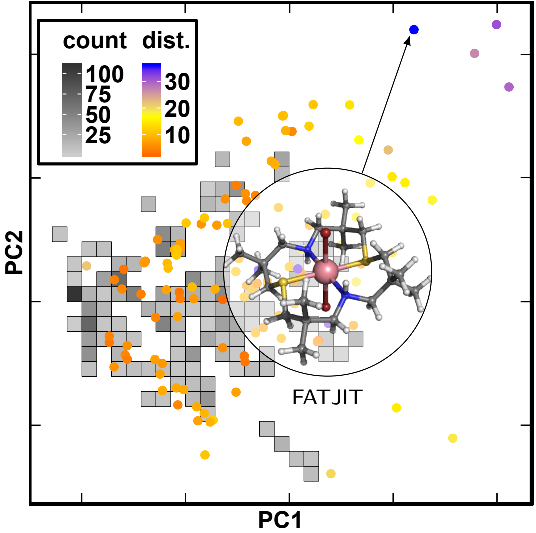

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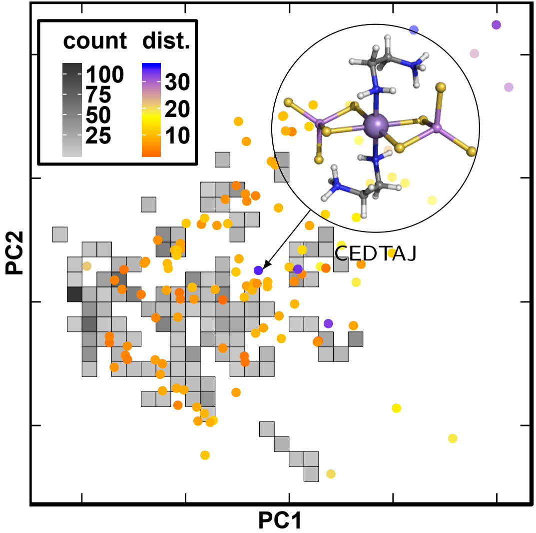

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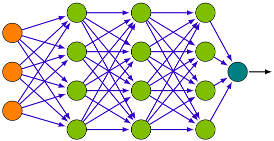

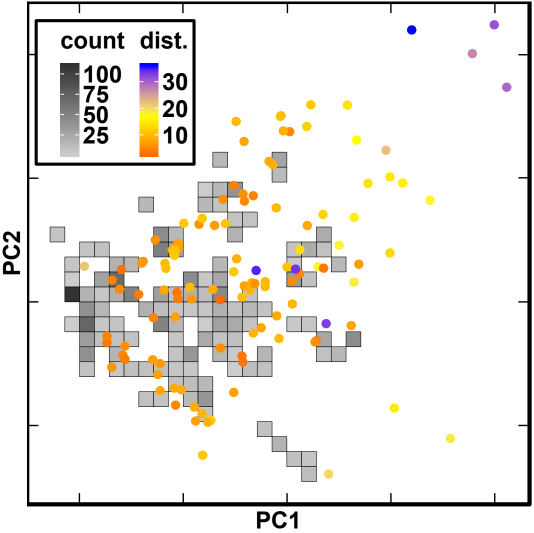

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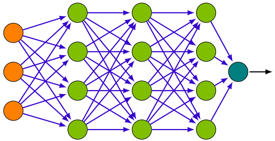

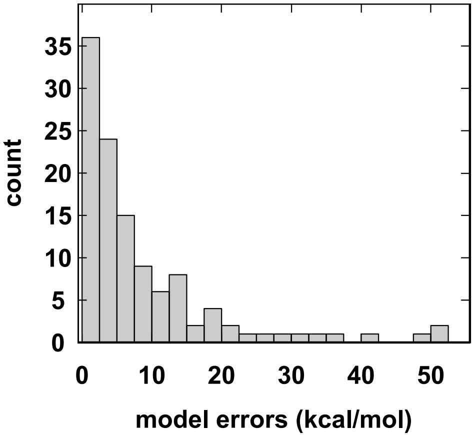

Latent distances give stable error control

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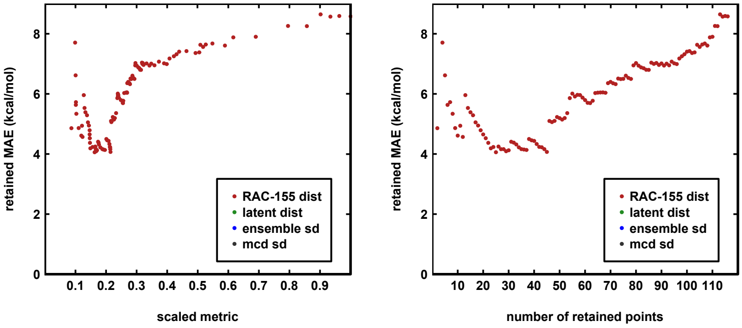

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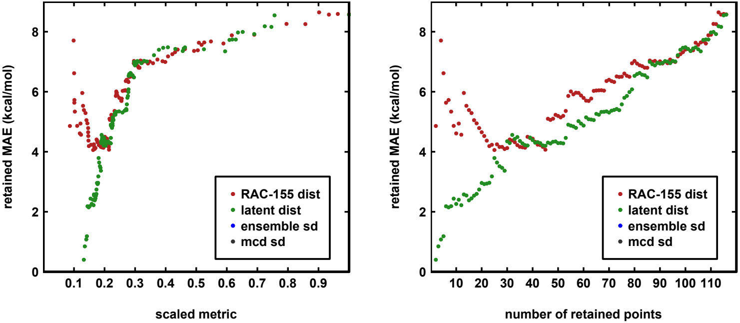

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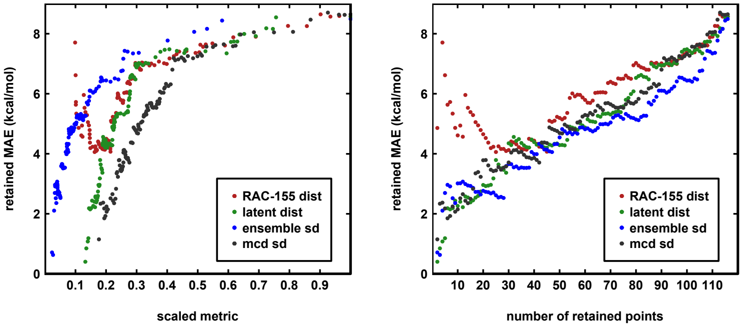

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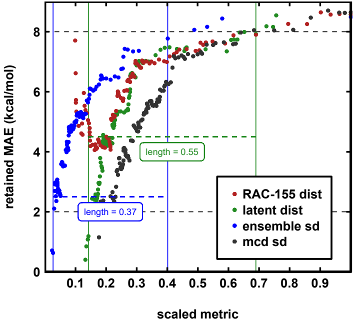

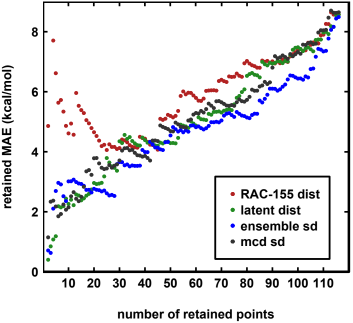

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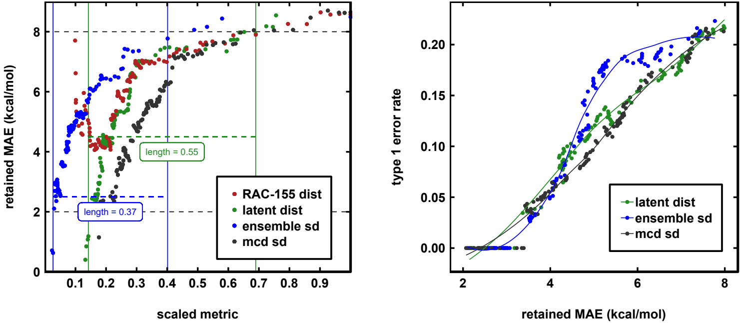

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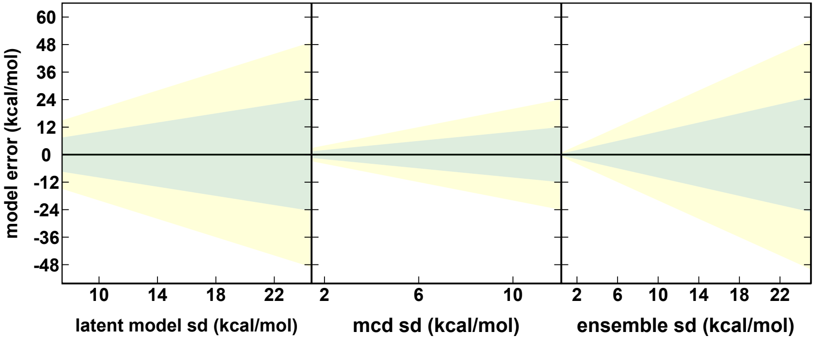

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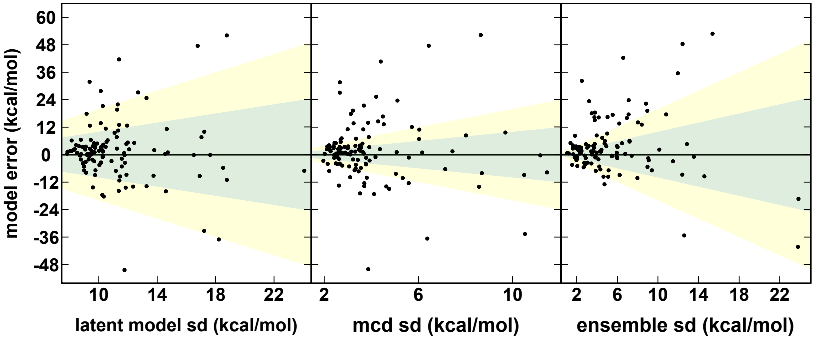

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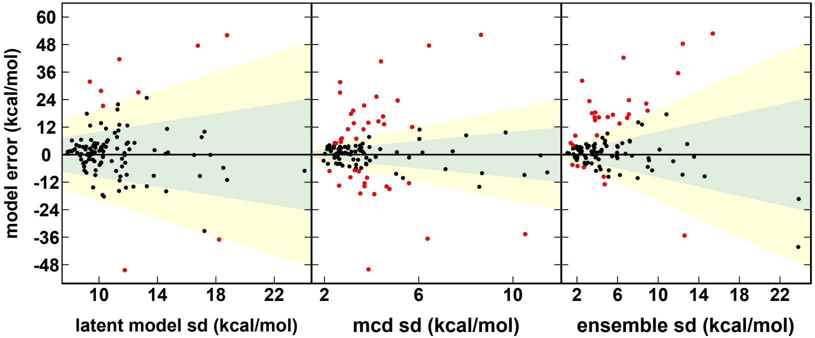

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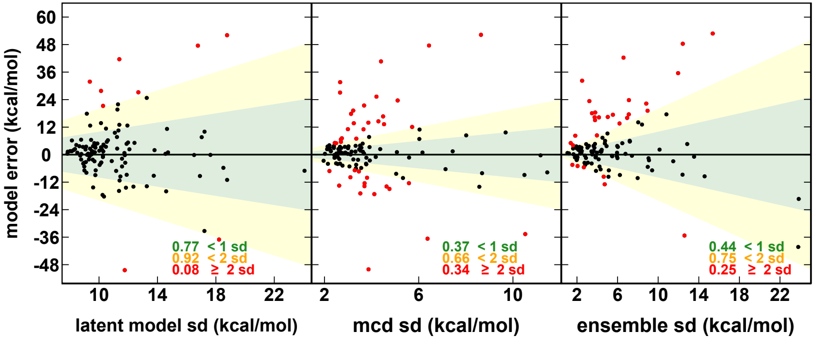

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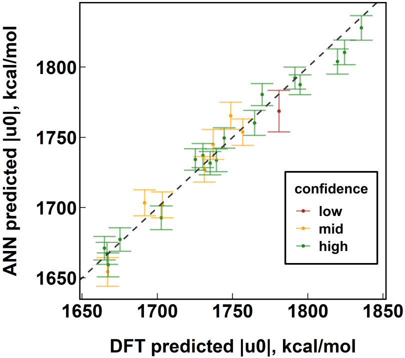

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Algorithmic chemical discovery

design space

candidate materials

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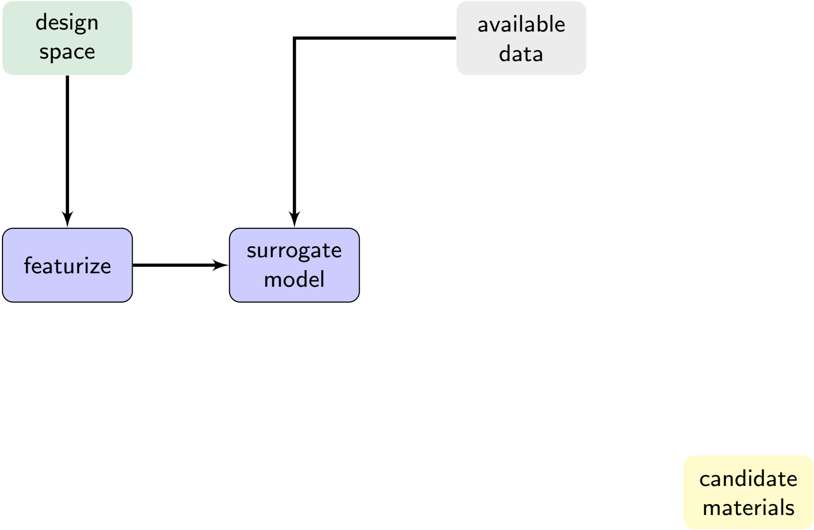

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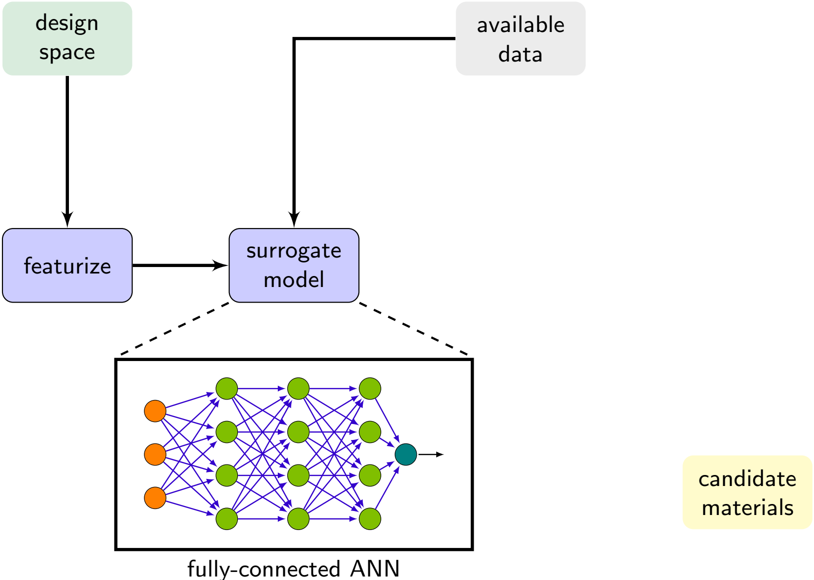

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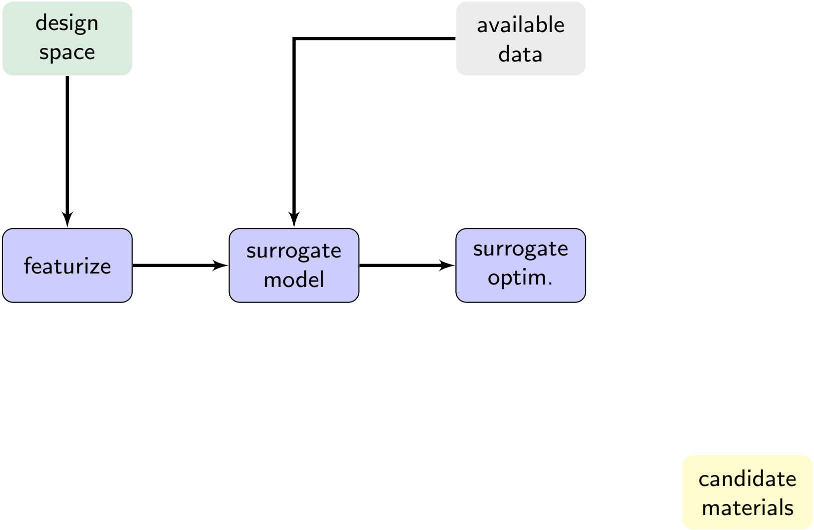

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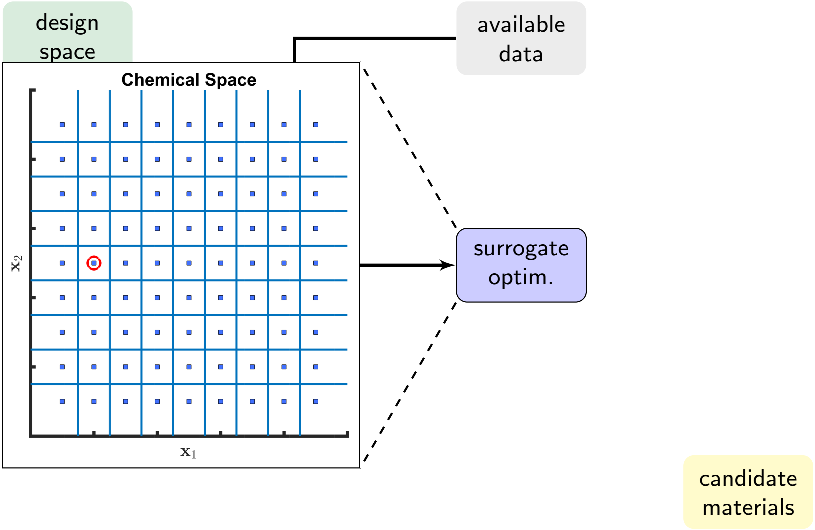

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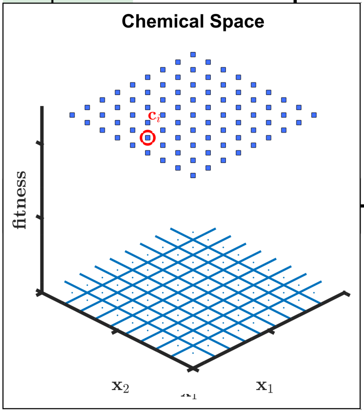
design space

available data



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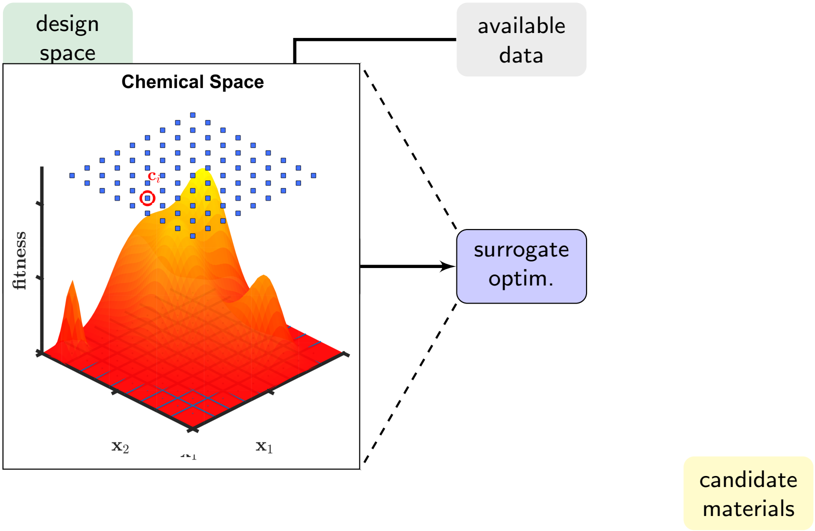

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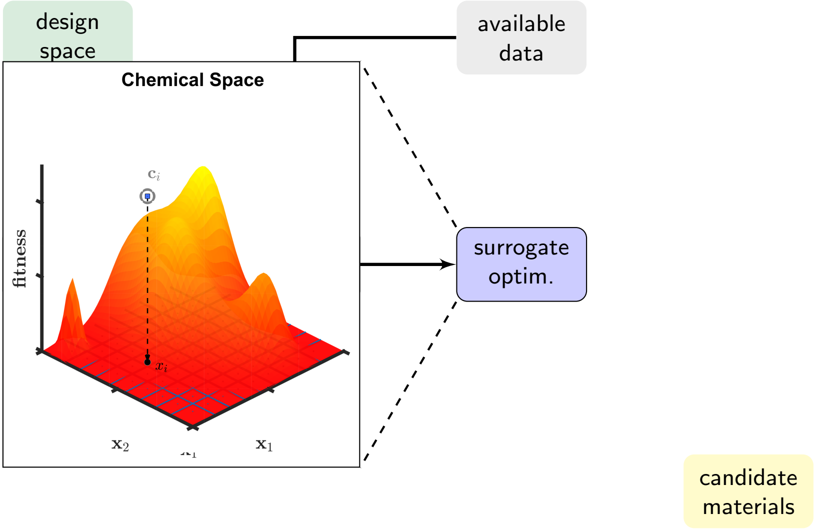

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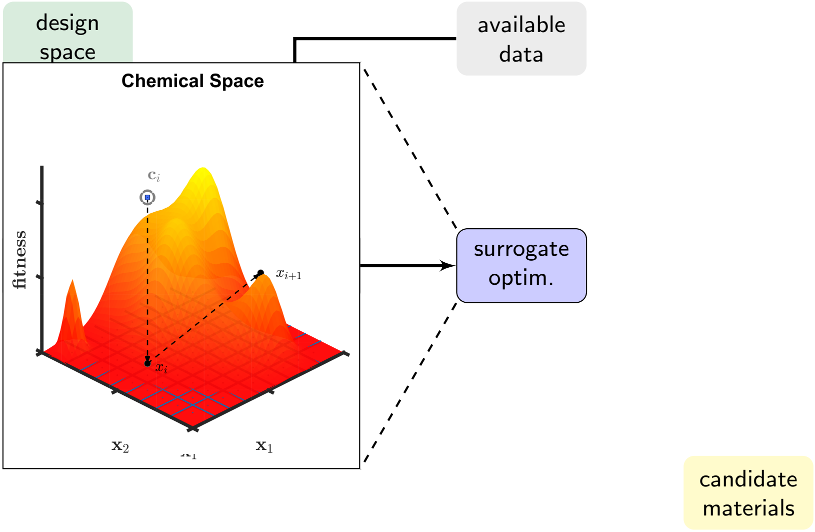

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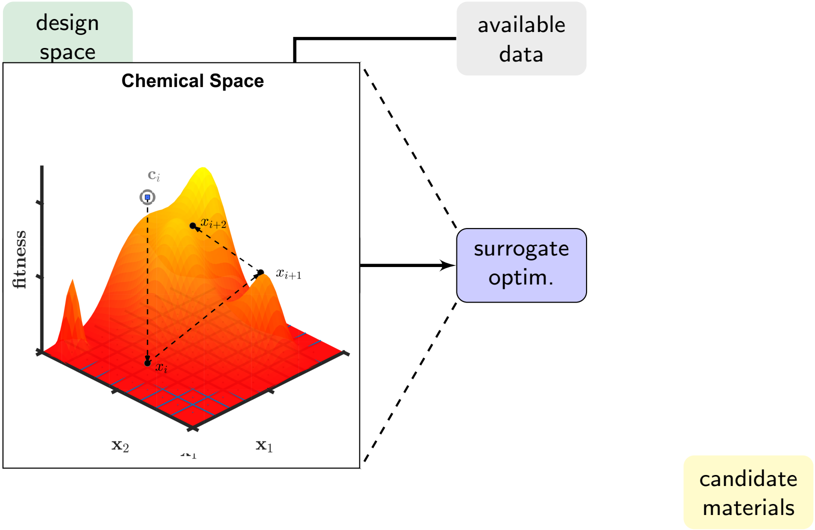

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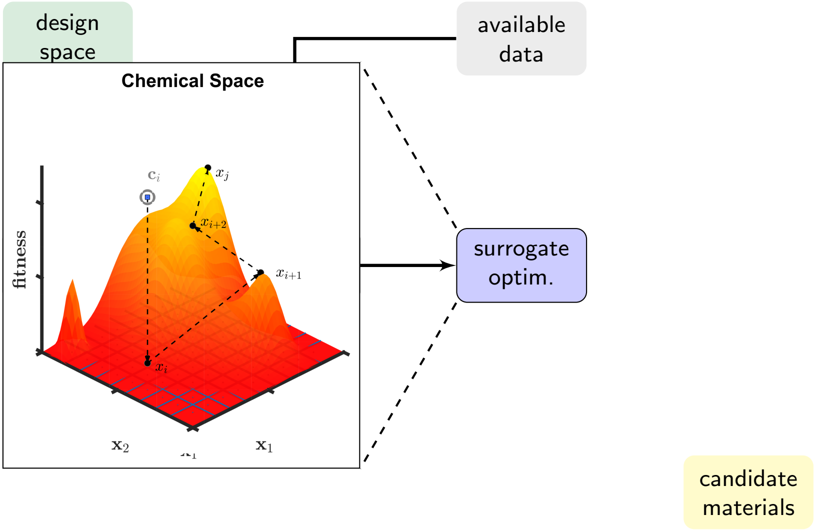

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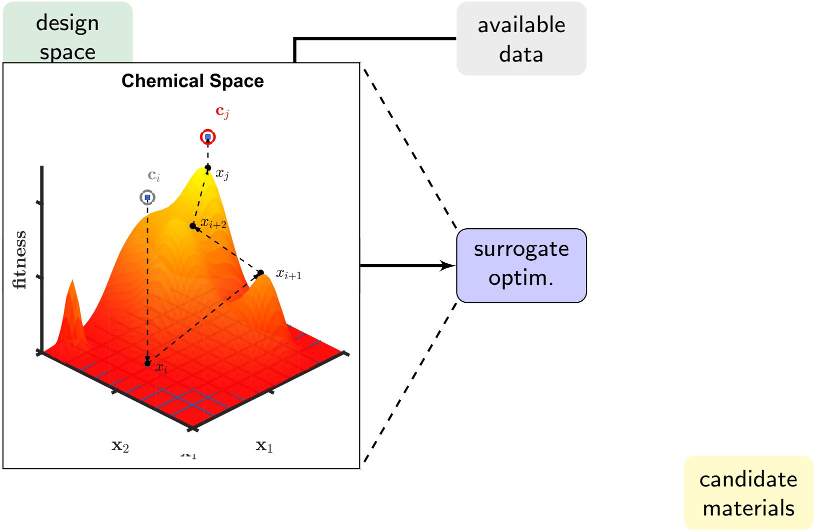

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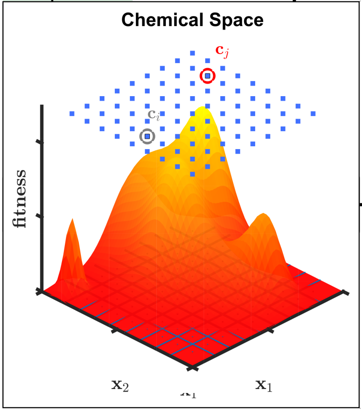

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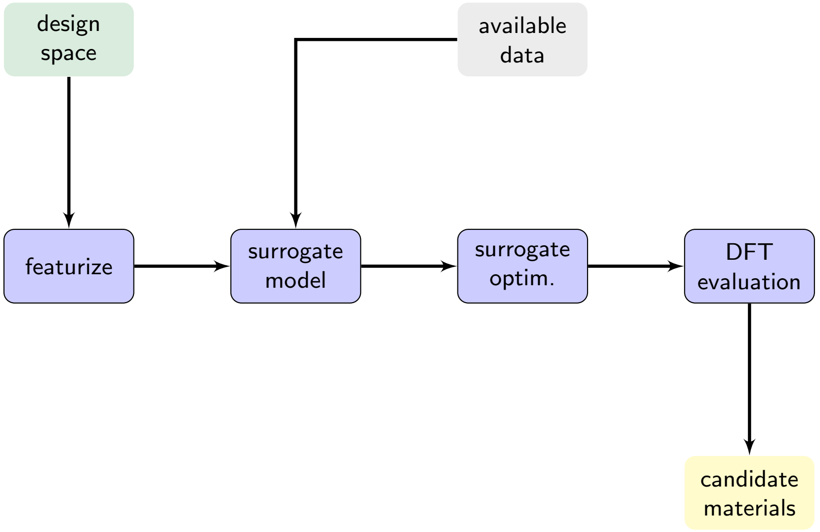

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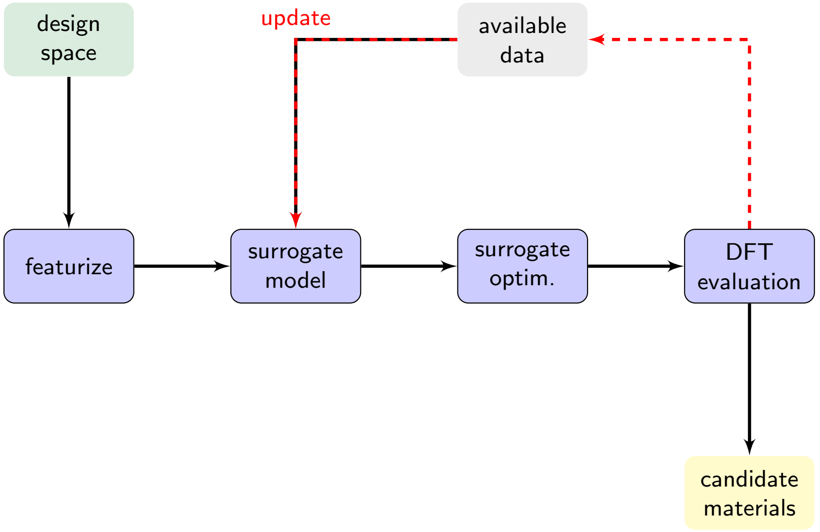

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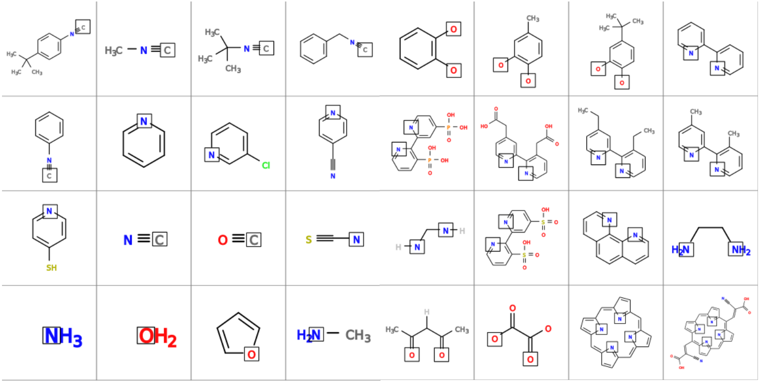

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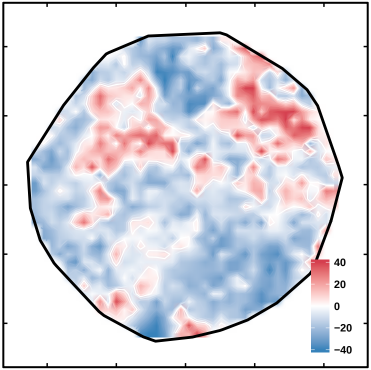

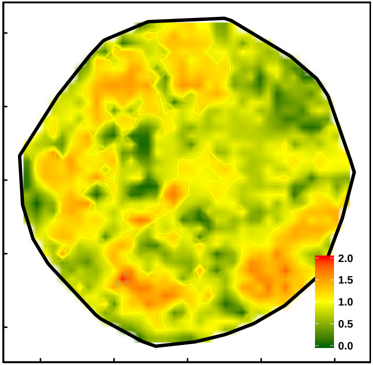

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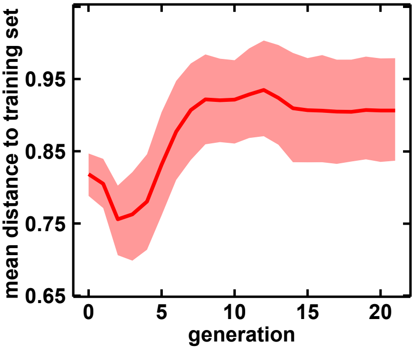

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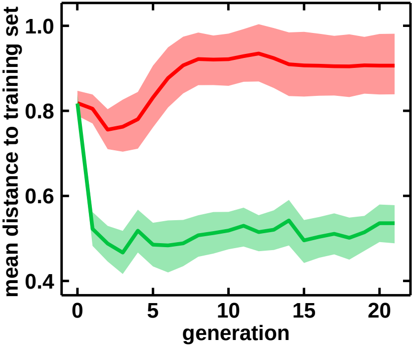

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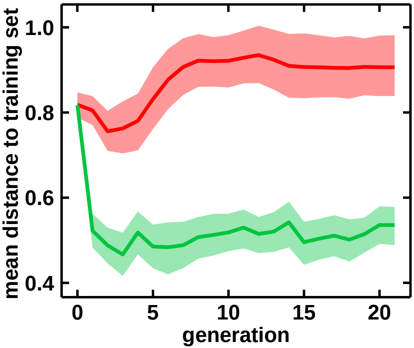
$$F_{s+d}(x) := \exp\left[-\left(\frac{\Delta E_{\text{H-L}}(x)}{p_{\Delta E_{\text{H-L}}}}\right)^2\right] \exp\left[-\left(\frac{d(x)}{p_d}\right)^2\right]$$



Janet, J.P., Chan, L. and Kulik, H.J., *J. Phys. Chem. Lett.*, 9(5):1064–1071, 2018.

Demonstration

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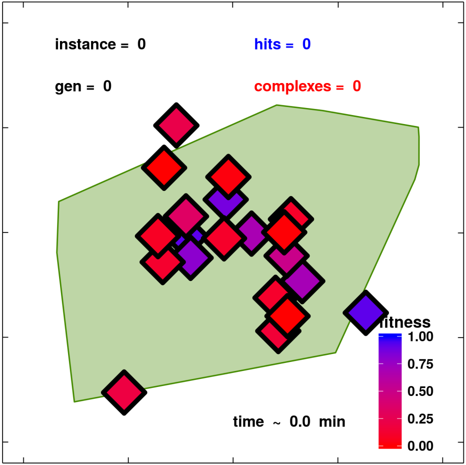

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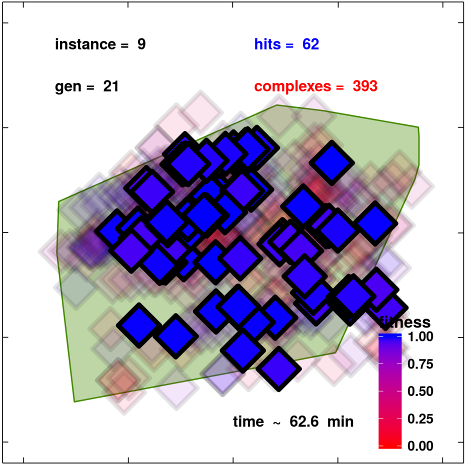
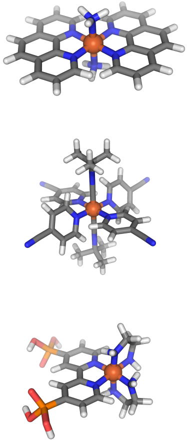
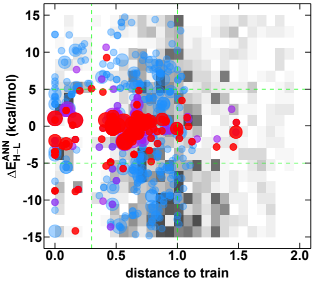




Discovery results

Spin splitting design:

We combine ANN predictions and uncertainties using an evolutionary algorithm.
Error control allows 60% of leads to be validated with DFT.[1]



[1] Janet, J.P., Chan, L. and Kulik, H.J., *J. Phys. Chem. Lett.*, 9(5):1064–1071, 2018.

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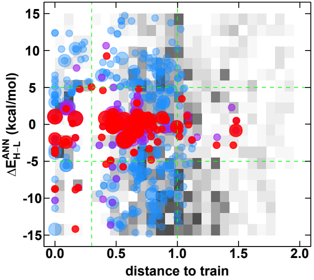
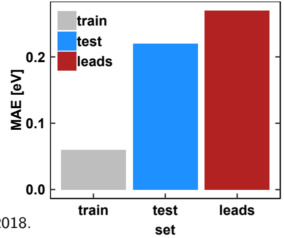


Frontier orbital properties:

This approach also works for frontier orbtial design[2], obtaining average HOMO of 3.98 eV compared to target 4.00 eV.



[1] Janet, J.P., Chan, L. and Kulik, H.J., *J. Phys. Chem. Lett.*, 9(5):1064–1071, 2018.
[2] Nandy, A. *et al.*, *Ind. Eng. Chem. Res.*, 57(42):13973–13986, 2018.

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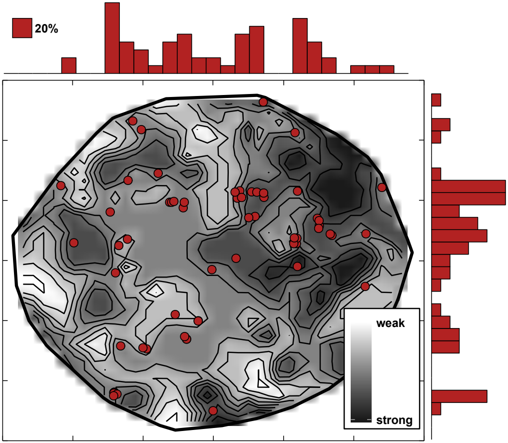

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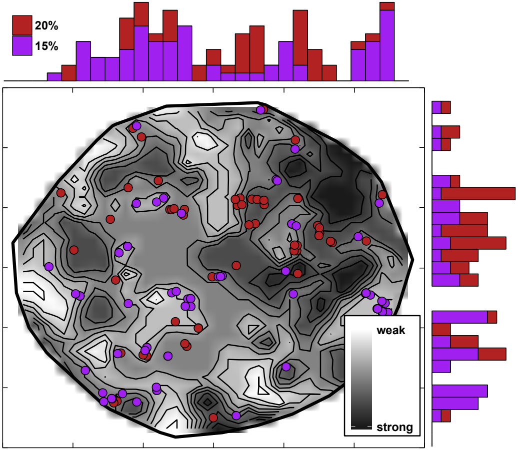

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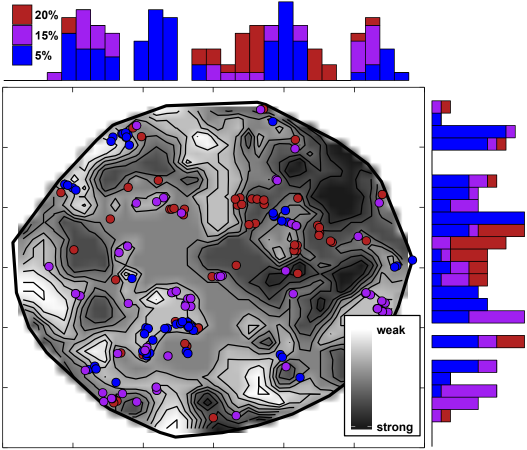
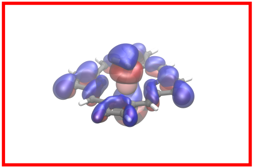


Computational chemistry and machine learning

Awkward roommates or match made in heaven?

physics-driven



slow, accurate (?)

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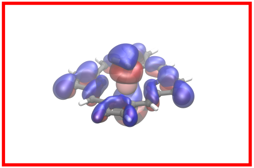

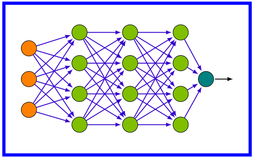

slow, accurate (?)

fast, uncertainty-aware

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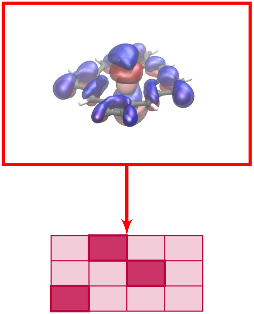

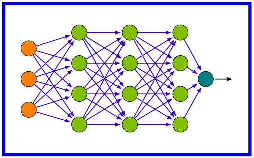

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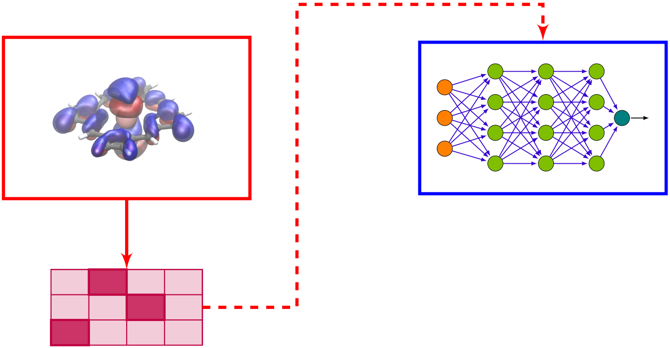

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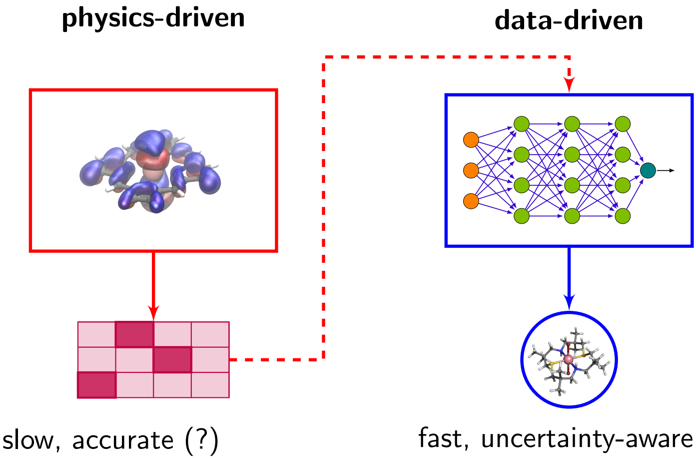

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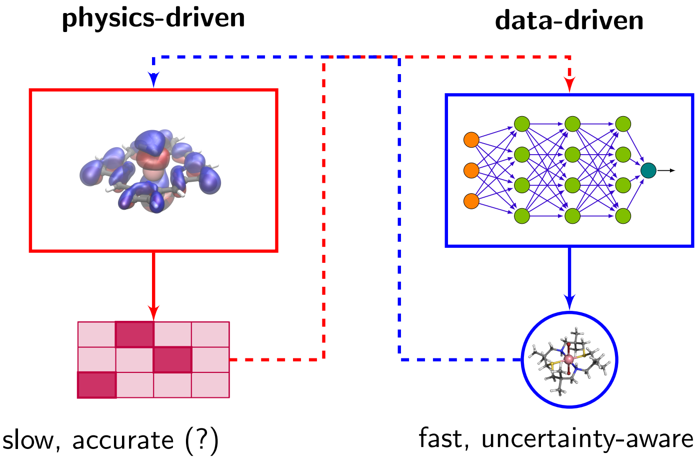

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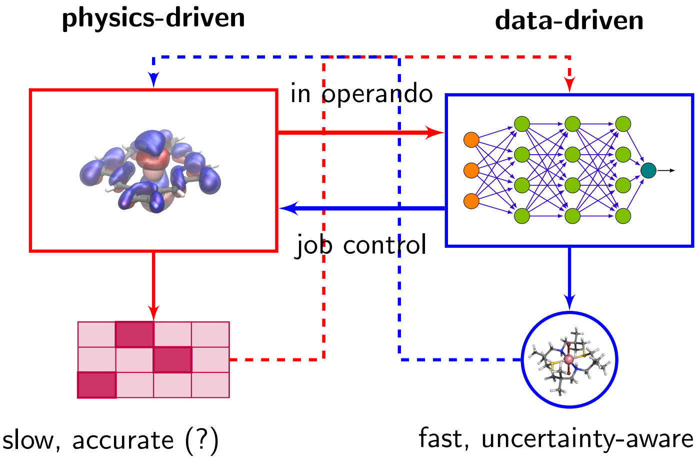

Acknowledgments

Thanks to the Kulik group and funding partners: