Replacing Chemical Intuition: Design of Experiments and Reinforcement Learning For the Construction of Training Sets

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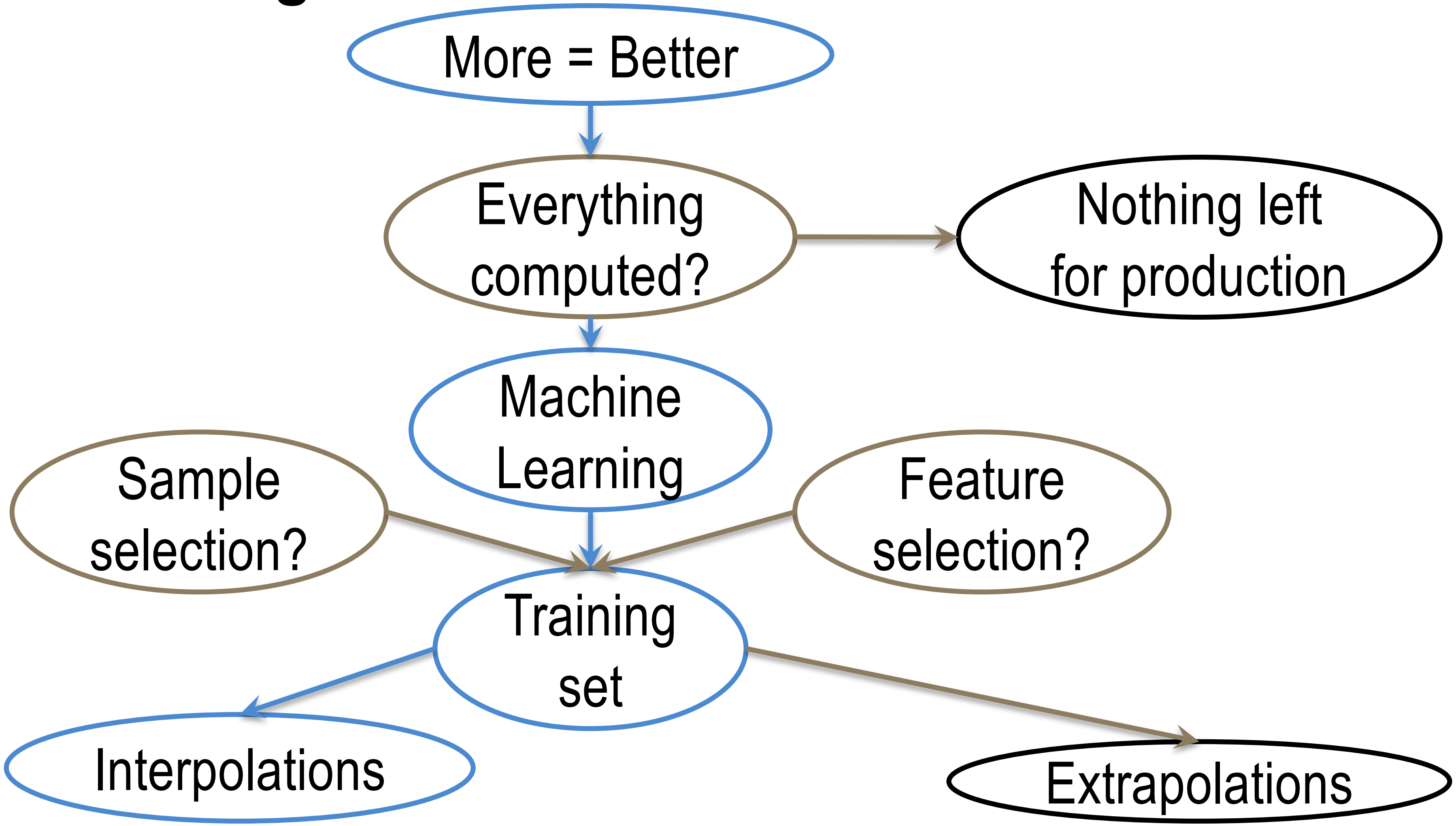
Machine Learning is Data Intensive



Training Set Determination: Two Possibilities

Case 1: **Maximum data set is available**

- Maximize interpolation capabilities for given n.

→ Create most representative subsample

Case 2: **Data set has to be created**

- Data generation is expensive → Compress as much as possible
- Maximize interpolations → Focus on most relevant features

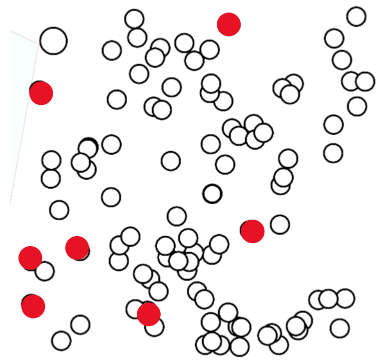
→ Create **smallest** representative sample

Case 1: Maximum Data Set Available

Random Sampling

Advantages

- Easy
- Unbiased
- Sample size can be converged easily



Disadvantages

- No link with interpolation power

→ Converges slowly (large sample sizes needed)

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Case 1: Maximum Data Set Available

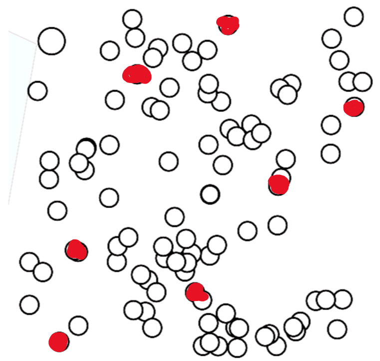
Tang, *J. Am. Stat. Assoc.*, **1993**, *88*, 1392.
Loyola, *Neural Netw*, **2016**, *78*, 75.

Orthogonal Latin Hypercube Sampling

Advantages

- Increased diversity

  → Small samples can be representative



Disadvantages

- Sample size cannot be increased continously
- Feature space has to be defined, but no knowledge of important regions

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Design of Experiments (DoE)

Advantages

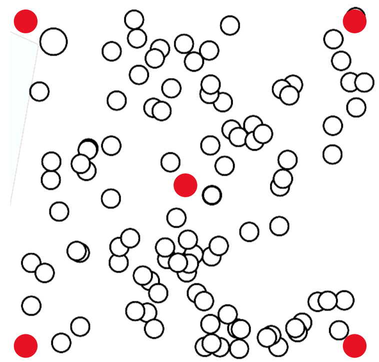
- Increased diversity compared to random; particularly well suited for interpolations

  → Small samples can still be representative

- Systematic construction

Disadvantages

- Feature space has to be defined
- No knowledge of important regions
- “Optimal” points might be unphysical



Case 1: Maximum Data Set Available

Cersonsky et al *Mach. Learn.: Sci. Technol.* **2021**, 2, 035038.

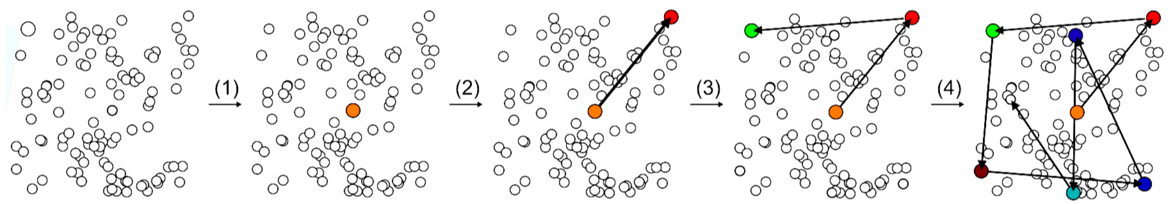
Farthest Point Sampling

Advantages

- Increased diversity compared to random; particularly well suited for interpolations

  → Small samples can still be representative

- Fast convergence, sample size can be increased continuously



Disadvantages

- Feature space has to be defined
- No knowledge of important regions, but tweaks exist

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Compression of Feature matrix (CUR) works at least as well

Choose the fewest representative samples

Enumerate

Explore randomly

Apply methods to generated samples, before expensive computation

Main idea: Learn and Construct on the Fly

- Construction is expensive

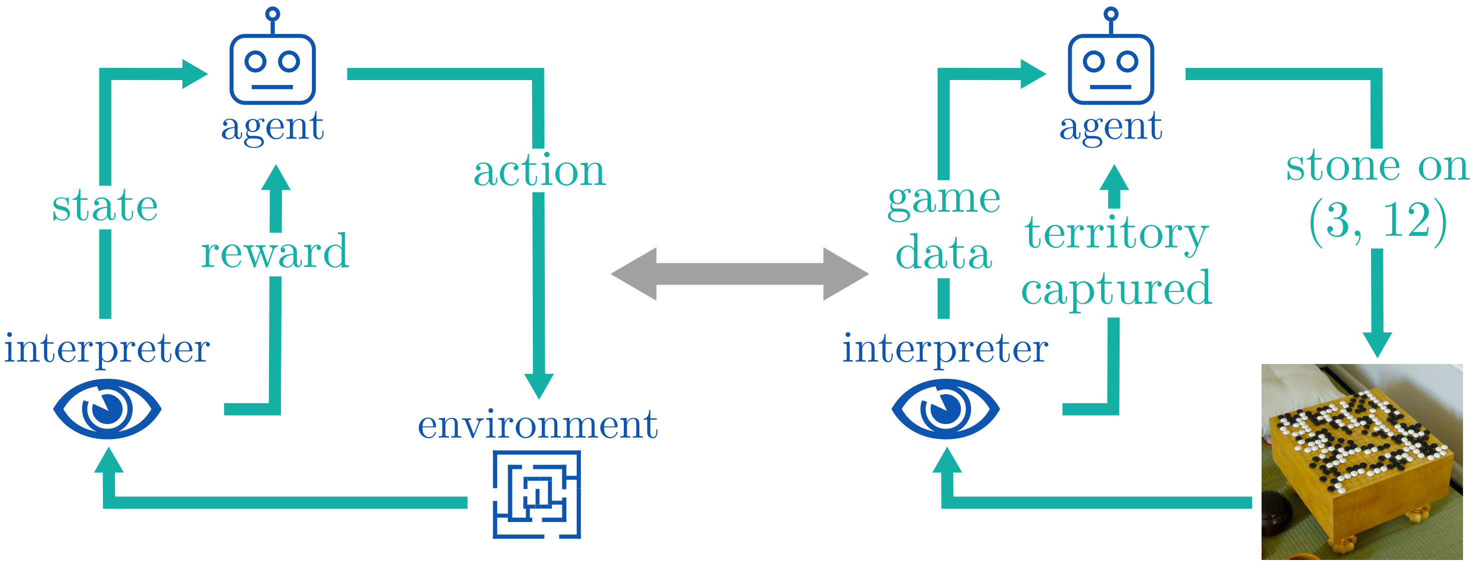
  → Construct as little as possible

- Which datapoints are necessary to improve model robustness?

Extrapolations → Interpolations!

(Active) Reinforcement Learning

- Learn the action patterns that lead to highest reward



- Compromise of exploration (new situations) and exploitation (pursue strategies leading to high rewards)

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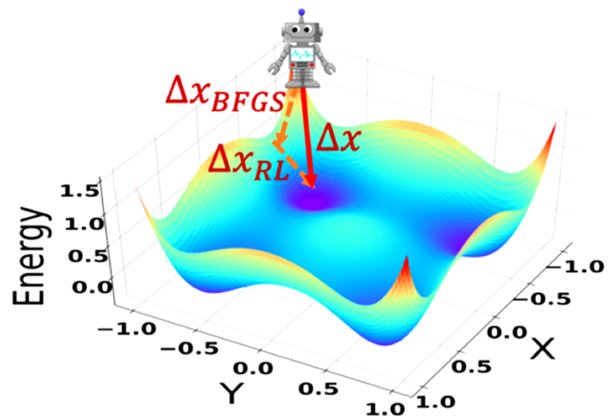
Ahuja et al *J. Chem. Theory Comput.* **2021**, *17*, 818.

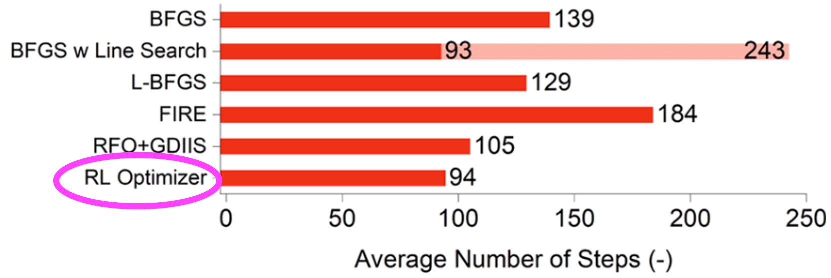
Geometry Optimization

- Input: Gradient and displacement history
- What is the best next step?
- Learn a policy for enhanced optimization step

→ Very successful for organic molecules

No additional computational cost





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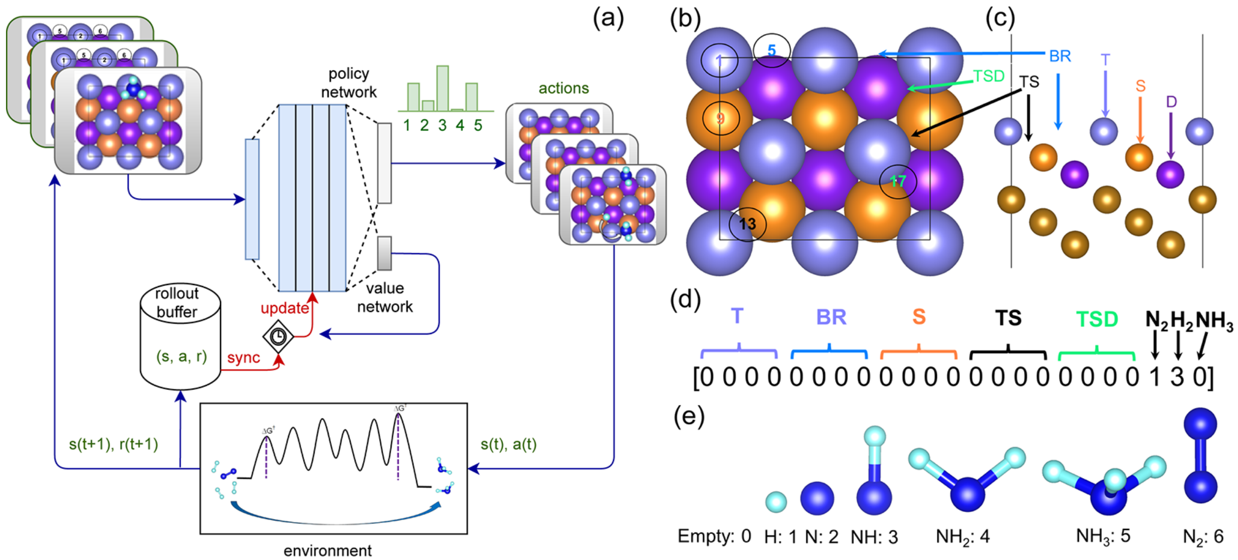
Lan and An *J. Am. Chem. Soc* **2021**, 143, 16804.

Reaction Path Discovery

- Input:
  - (1) Surface state
  - (2) Possible reaction steps
  - (3) Products
- What is the most likely next reaction leading towards products?
- Learn reaction kinetics



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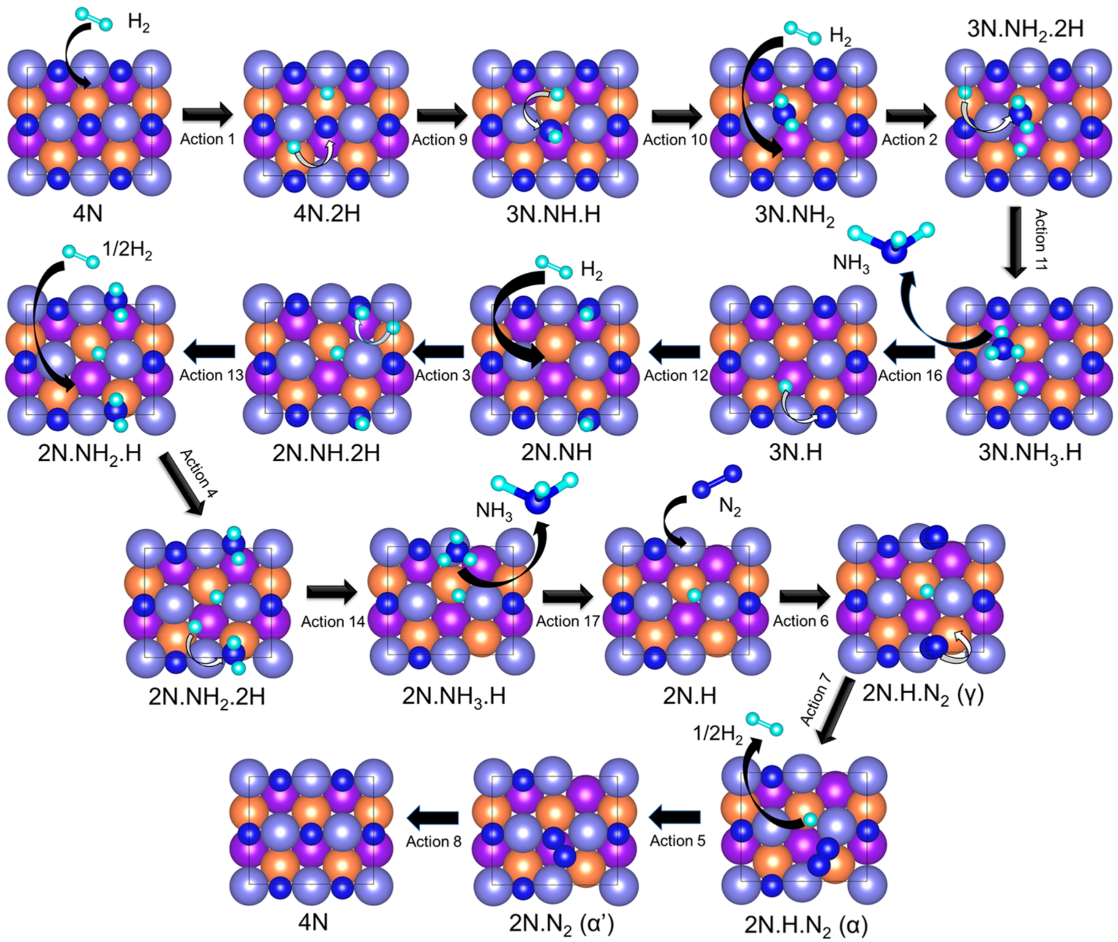
Lan and An *J. Am. Chem. Soc* **2021**, 143, 16804.

Reaction Path Discovery

Identified the most plausible (lowest energy) pathway for Haber-Bosch process on Fe(111):

Order of adsorption/desorption!



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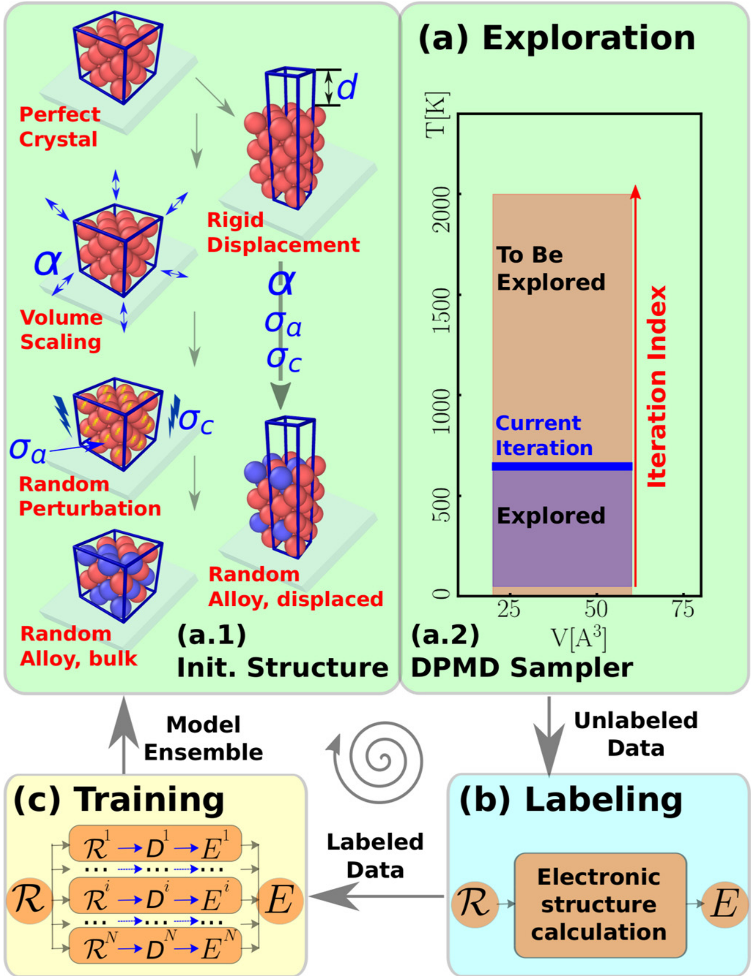
Zhang et al *Phys. Rev. Materials* **2019**, *3*, 023804.

ML Potentials

- Deep Neural Networks have several local minima
- Train an ensemble of DNNs together
- Use ensemble to make predictions

Largest deviations?

→ Add training data



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Goings et al *J. Chem. Theory Comput.* **2021**, *17*, 5482.

Approaching Full Configuration Interaction

- Input:

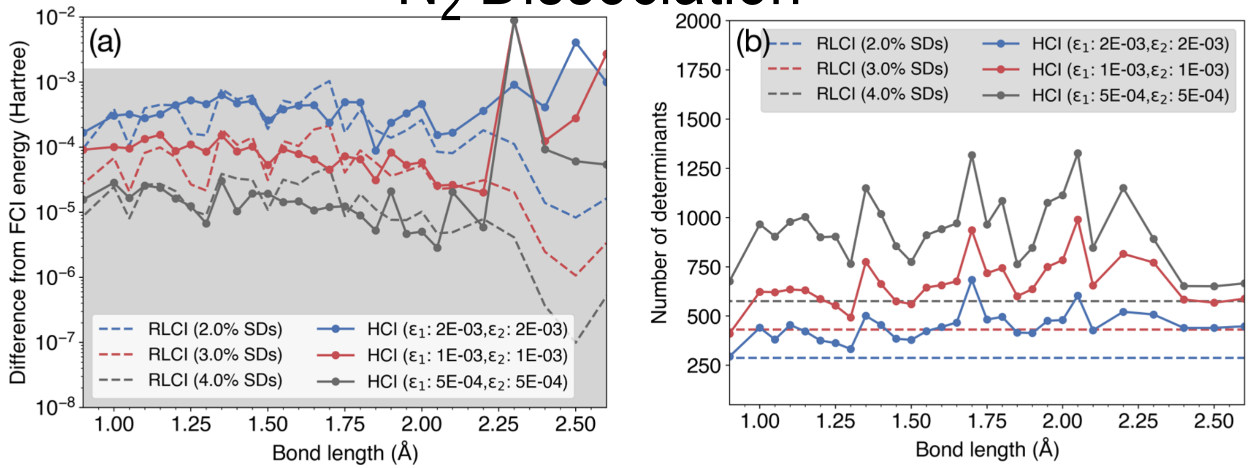
  (1) Current Slater determinants

  (2) Perturbation-based estimates for adding/removing Slater determinants

- What is the ideal combination of Slater Determinants?

- Learn compression of FCI wave function

N_2 Dissociation

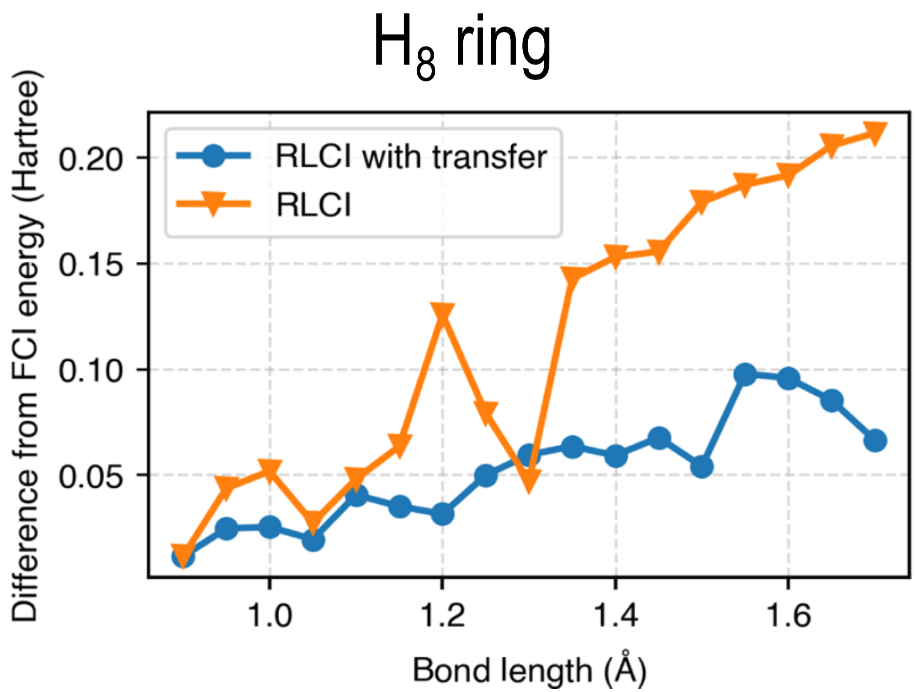


Goings et al *J. Chem. Theory Comput.* **2021**, *17*, 5482.

Approaching Full Configuration Interaction

- Transfer learning is strength of RL
- Learn on one system, adapt knowledge to the next

  → Gain in efficiency and accuracy!



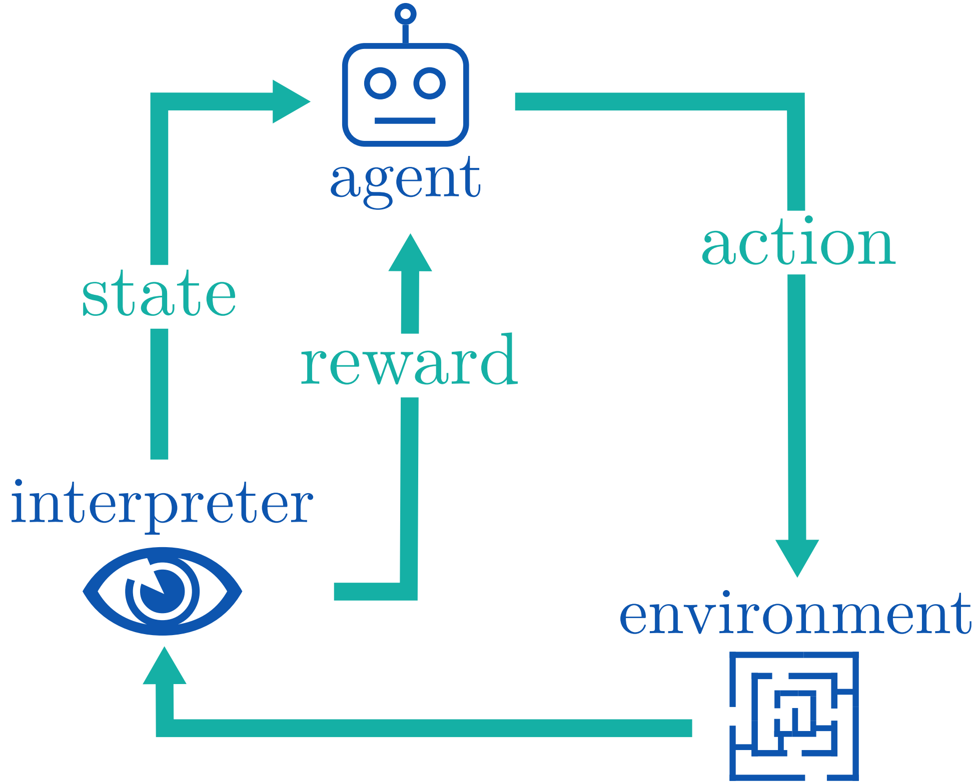
Fully learn at 0.9 Å (30 iterations)
Adapt knowledge for other distances (15 iterations)

Active Reinforcement Learning

- **Action**: Which sample to add to the training set
- **State**: Current training set
- **Reward**:

  (1) Robustness of fitted model

  (2) dominating terms best defined



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Cluster Expansion Model Hamiltonians

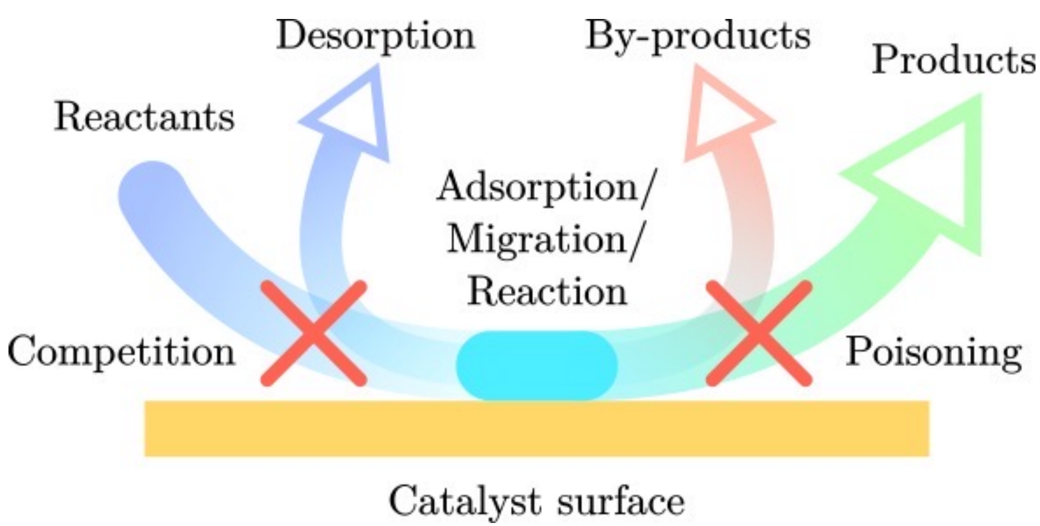
Typical applications

- Description of alloy (bulks, surfaces, NPs)
- Adsorption (and reaction) on surfaces

Typical case for **"one use only" potentials**

→ Minimizing computational cost for its production

→ We knowingly accept inaccuracies

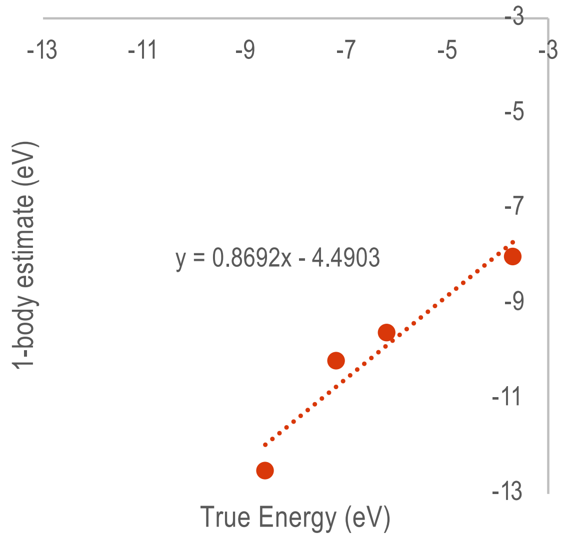


Example: Training of Model Hamiltonian

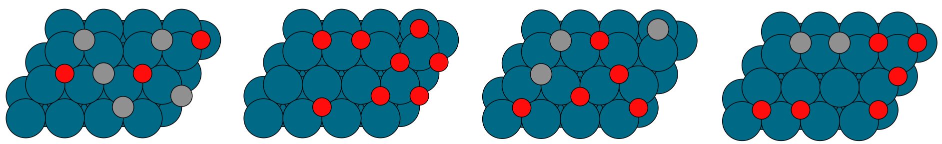
What is a Cluster Expansion?

A linear model, mapping configurations (patterns) and energy contributions

$$E_{ads}(g) = \underbrace{\sum_a \beta_a N_a(g)}_{\text{1-body terms}} + \underbrace{\underbrace{\sum_{a,b} \beta_{a-b} N_{a-b}(g)}_{\text{2-body terms}} + \underbrace{\sum_{a,b,c} \beta_{a-b-c} N_{a-b-c}(g) + \ldots}_{\text{3-body terms}}}_{\text{lateral interactions}}$$



2- and higher body terms matter



1-body estimate (ev):	-12.5	-8	-10.2	-9.6
True energy (eV):	-8.6	-3.7	-7.2	-6.2

What is a ”Good” Training Set?

Dominating strategy

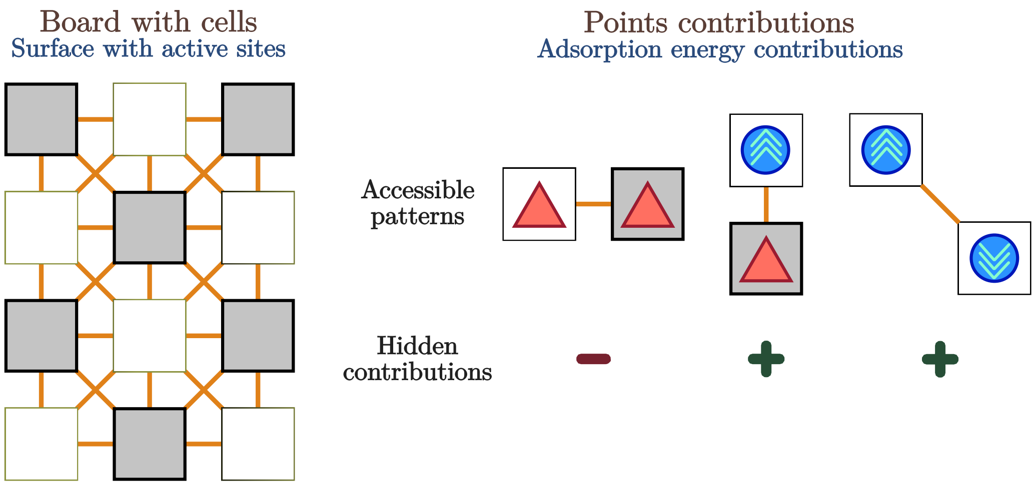
- Construction by hand
- Exploiting chemical intuition
- User time consuming!

Automatically created, better than random

- **Relevant**: Better sampling of regions with important lateral interactions
- **Diverse**: Don’t miss patterns
- **As small as possible**: Low redundancy

Staub and Steinmann, submitted.

Cluster Expansion: A board game perspective



- Adsorbing one adsorbate after the other
- Learn positive and negative lateral interactions
- No prior knwoledge
- Long-term strategy

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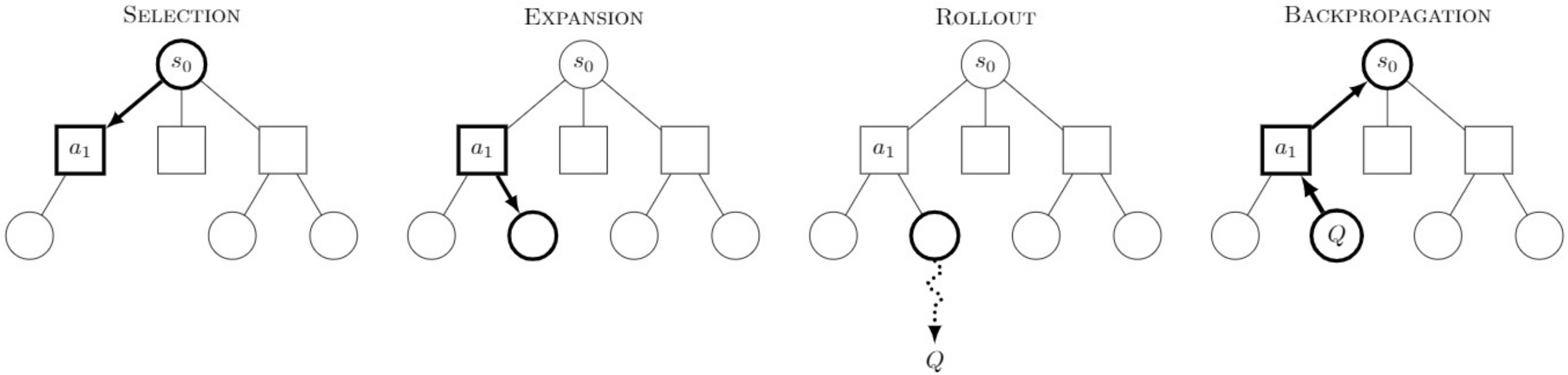
UCT: A Typical Reinforcement Learning Approach

Upper Confidence bounds applied to Trees :

Monte Carlo Tree Search



Based on previous plays | Explore a new possibility | Determine the score | Update information

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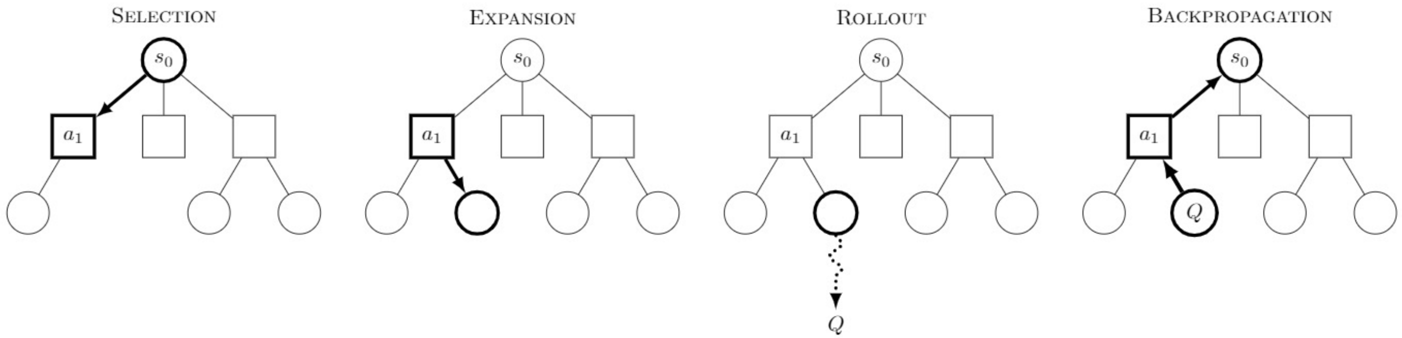
Auer et al *Mach. Learn.* **2002**, *47*, 235.

UCT: A Typical Reinforcement Learning Approach

- Upper Confidence bounds applied to Trees :
  Monte Carlo Tree Search



- Combined with Upper Confidence Bounds
- **Optimal Exploitation/Exploration trade-off**
  Like in multi-armed bandit problem (e.g., UCB1)

$$\frac{w_i}{n_i} + c\sqrt{\frac{\ln N_i}{n_i}}$$

N_i: Total plays passing through parent node
n_i: plays passing through considered node
w_i: Wins going through node i
c: exploration constant; √2 in theory

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Staub and Steinmann, submitted.

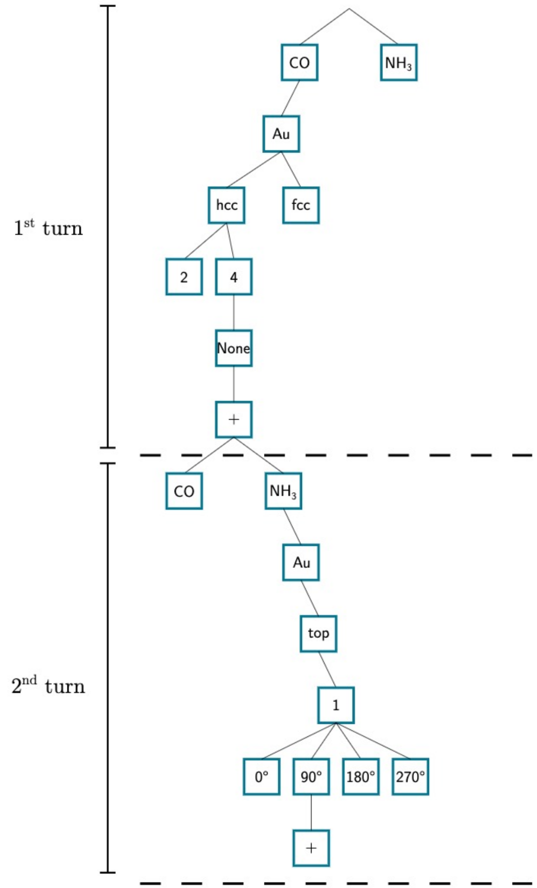
Particularities when Constructing a Training Set

- UCT is trained by simulating games
- Relies on fast evaluations of the true score
- Here: true score = DFT computation (expensive!)

  → We are actually exploiting the learning period!

  → Accelerate Learning

  → Increasing incentive for exploration (curiosity)



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Example: Training of Model Hamiltonian

Garivier et al *Procs Mach. Learn. Res.* **2011**, *19*, 359.
Staub and Steinmann, submitted.

Tweaks of UCT to the Construction of Training Sets

- When comparing unseen nodes

  → minimize the variance (A-optimal DoE)

  → Accelerate exploration (curiosity)

- Use KL-UCB as faster converging compared to UCB1

  → Tighter bounds, make exploitation as efficient as possible

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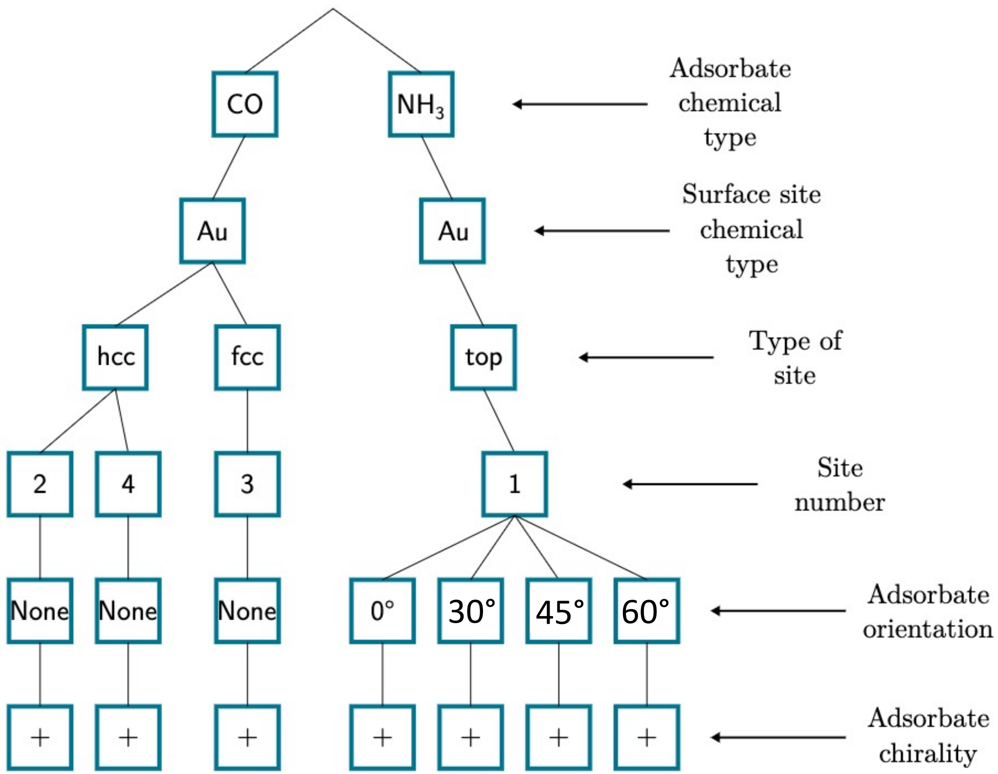
Thiede et al. *arXiv*, **2020**, 2012.11293.

Staub and Steinmann, submitted.

Proof of Principle: Tree



Completely general, all kind of surfaces and adsorbates

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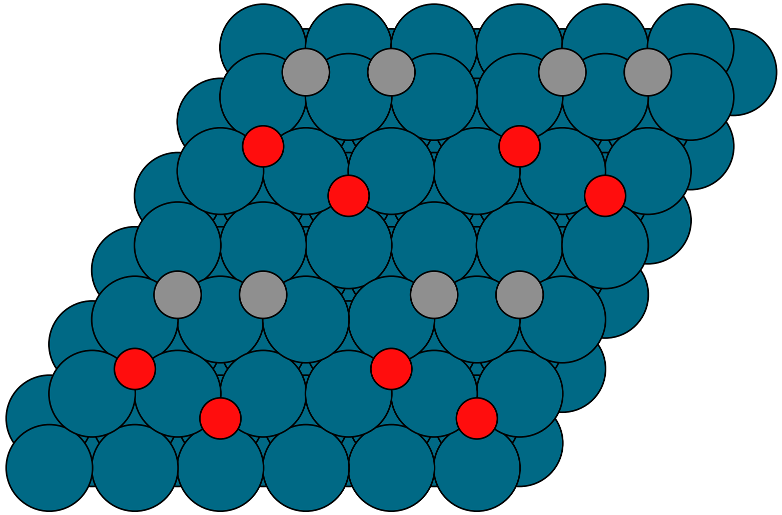
Proof of Principle: System

- CO oxidation on Pd(111)
- O, CO as adsorbates
- Ternary (fcc and hcp) adsorption sites



Piccinin et al *ACS Catal*. **2014**, *4*, 2143.

Proof of Principle: System

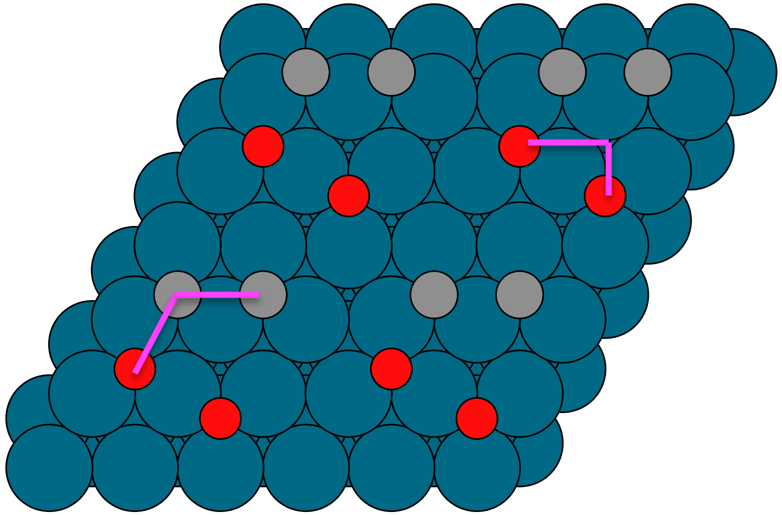
- CO oxidation on Pd(111)
- O, CO as adsorbates
- Ternary (fcc and hcp) adsorption sites
- Model Hamiltonian

  Up to 3-body terms

  Up to next-nearest neighbors

  Maximum 3 sites involved

  → 70 possible terms, but only 48 considered important



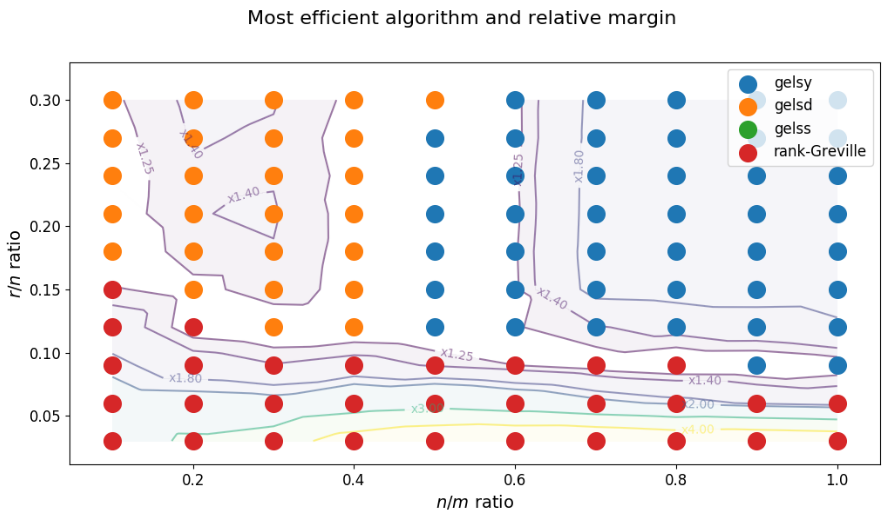
Staub and Steinmann, Appl. Math. Comput. 2021, 399, 125996.

Recursive Least-Squares Solver

- (Active) Reinforcement Learning of a linear problem

  → **Update least squares** solution instead of recomputing it!

- Exploit rank-deficiency and rank-factorization



At low rank *r* to row *n* ratio, rank-Greville is faster (independent on row/column ratio, *n/m*) even for full solution of the least-squares problem!

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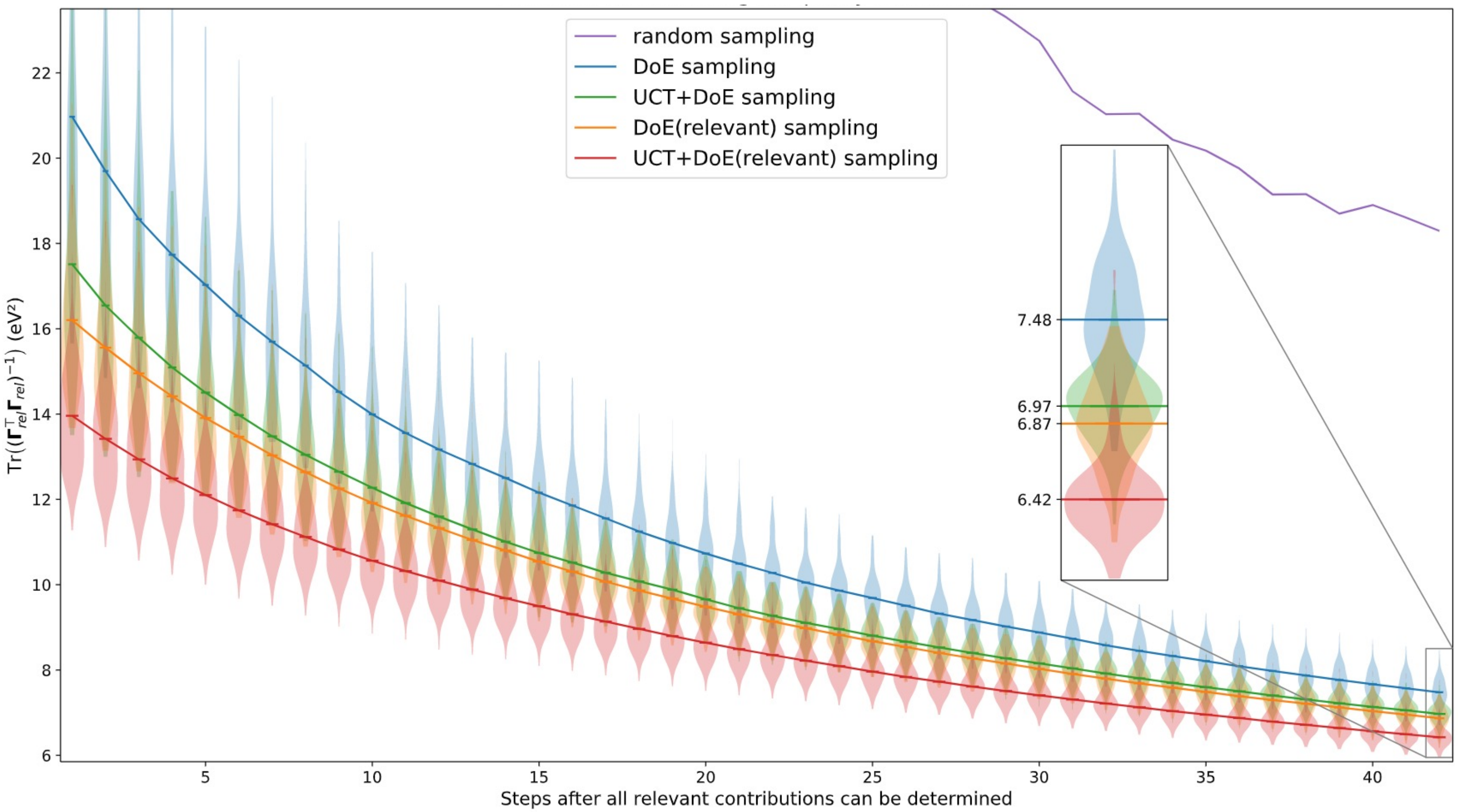
https://gitlab.com/lch_interfaces/rank-greville

Staub and Steinmann, submitted.

Proof of Principle: Results



UCT+DoE allow to automatically focus on most relevant features

Conclusions

- Compact, representative training sets for robust interpolations
- Random sampling is a poor strategy for small training sets
- Farthest-point sampling is convenient and robust

- Reinforcement learning is underused in chemistry

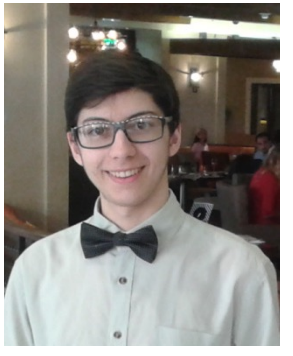
  Optimization problems are frequent: From geometry to FCI

- (Active) Reinforcement learning for automatic optimal training set construction.

Acknowledgements

PhD (and Post-Doc)

Ruben Staub



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