Trinity College Dublin
Coláiste na Tríonóide, Baile Átha Cliath
The University of Dublin

Learning Shortcuts in the Chemical Space

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SS TP Capstone Presentation 2024

Objective?

To leverage **reinforcement learning** methods to screen a large number of molecules at low computational cost for promising high temperature **single-molecule magnet** candidates.

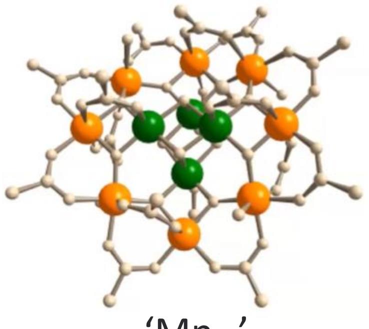
Single-molecule magnets (SMMs)

- Molecules that show a magnetic memory effect, i.e. they retain their magnetisation at low temperatures.

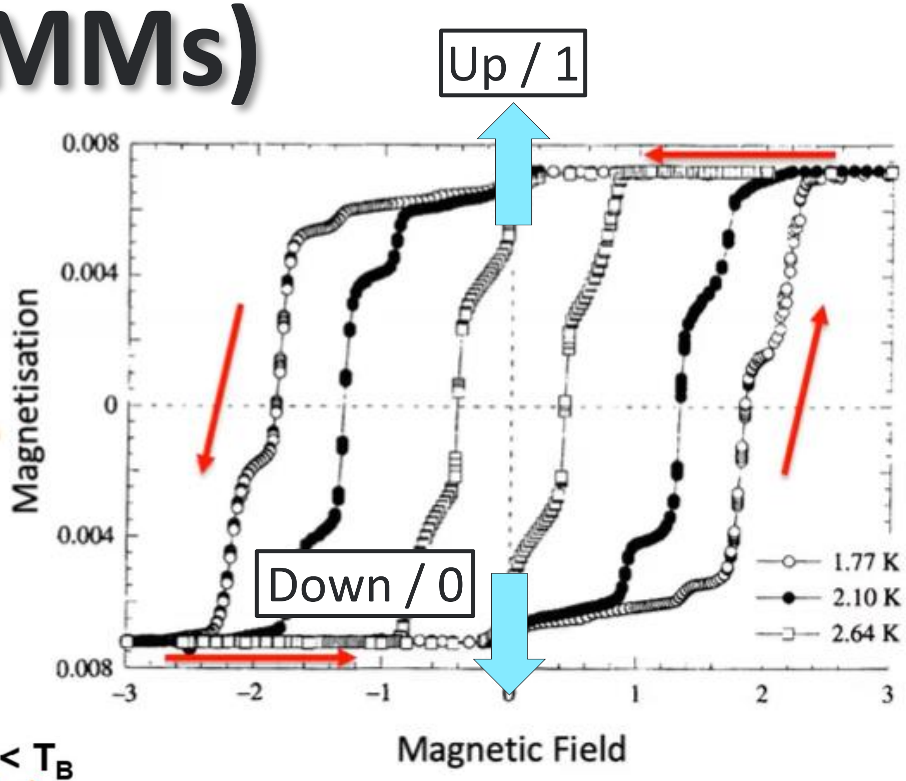
- Why? **Magnetic anisotropy**

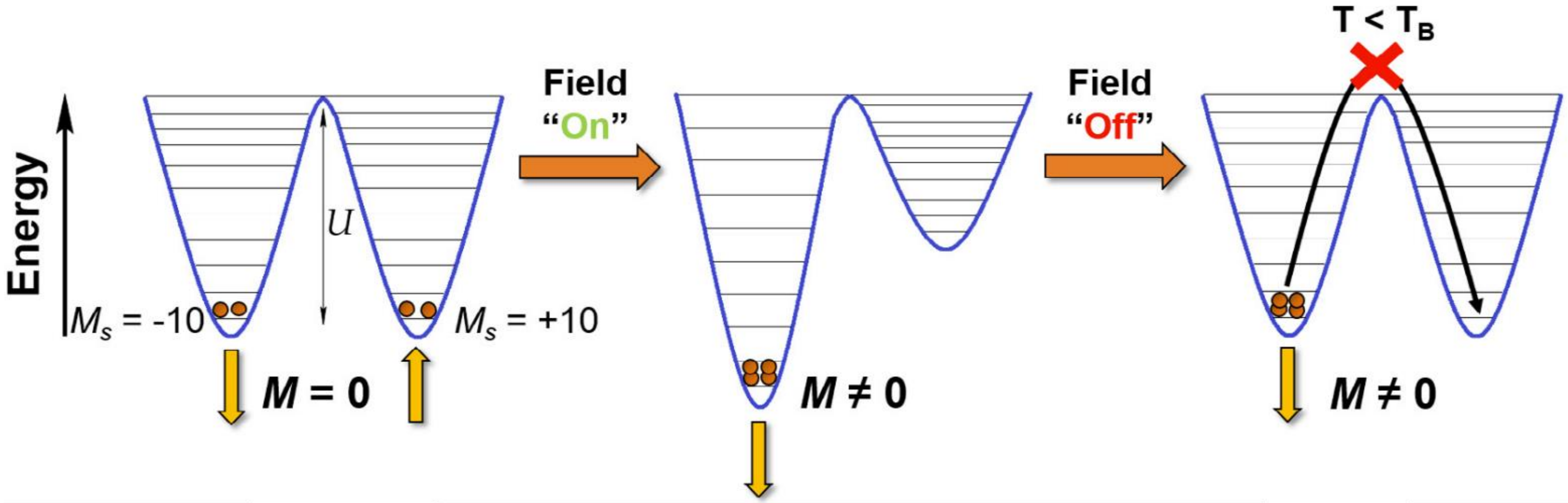
$$E = M_S^2 \cdot D$$

$\Longrightarrow$ Maximal $|M_S|$ in ground state for $D < 0$



‘Mn₁₂’





- Relaxation rate:

$$\tau^{-1} = \tau_0^{-1}\exp[\frac{-U}{k_B T}]$$

$$U \propto |D|$$

We want highly negative D!

Single-molecule magnets (SMMs)

Applications of room temperature single-molecule magnets:

- Data storage:

  - Current HDDs ~ 200GB in^{-2}
  - SMMs ~ 20,000GB in^{-2}

  Data centres 100 times smaller/more efficient

- Quantum computing

- Spintronics

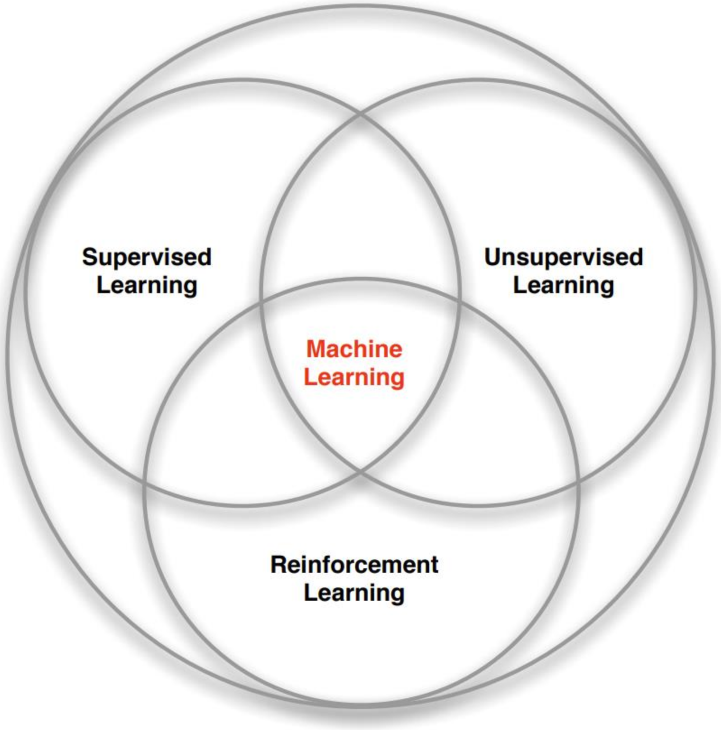
Reinforcement Learning (RL)

- Learn through trial and error by interacting with environment and receiving rewards or penalties based on actions.

- Feedback is delayed.

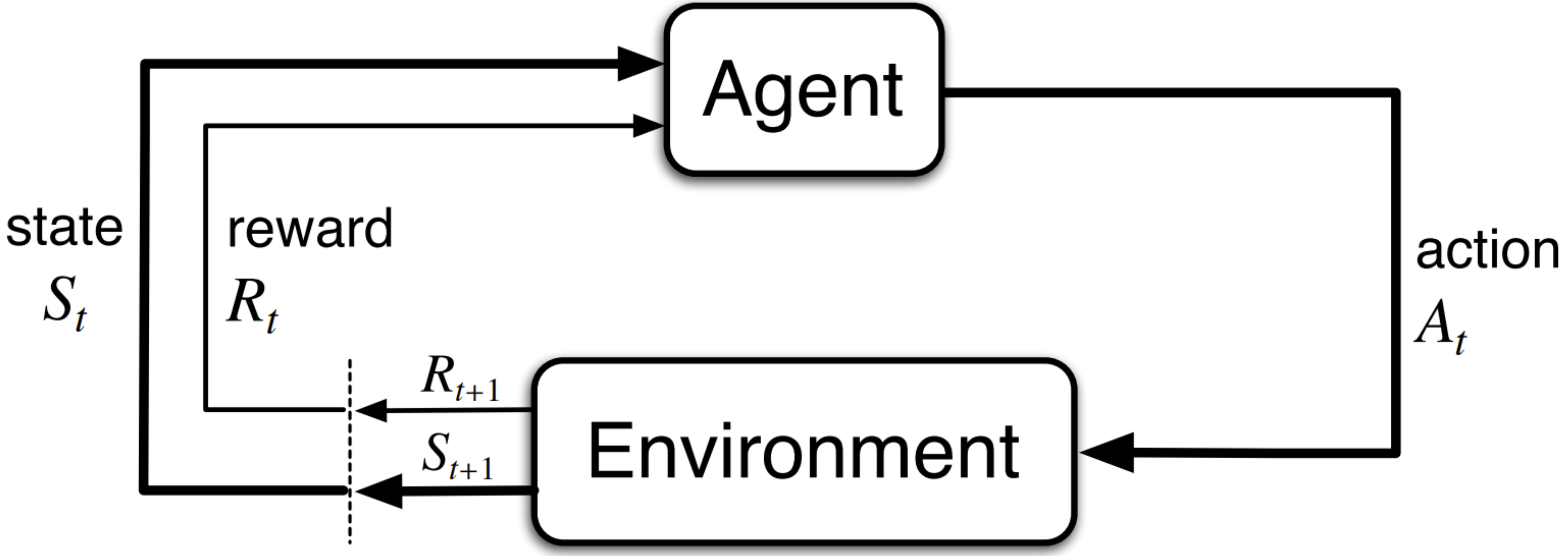
- Importance of balancing exploration vs exploitation.

- Extremely versatile and has had many successes to date:
    - Robotics
    - Autonomous vehicles
    - Finance/trading
    - AlphaGo



Markov Decision Processes (MDPs)

- MDPs provide a mathematical framework for RL problems.



Components of an RL agent

- A policy π maps states to actions: $\pi(a|s) = \mathbb{P}[A_t = a \mid S_t = s]$

- A value function gives a prediction of future reward while following a certain policy.

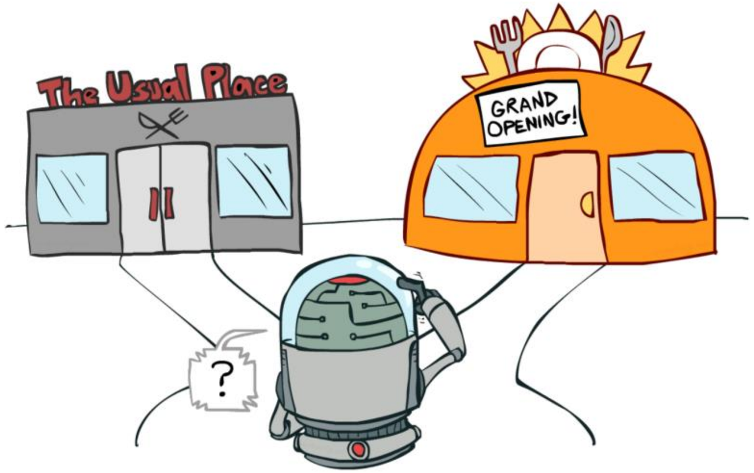
$$q_\pi(s,a) = \mathbb{E}_\pi[R_{t+1} + \gamma R_{t+2} + \gamma^2 R_{t+3} + \cdots \mid S_t = s, A_t = a] \;,\; \gamma \in [0,1]$$

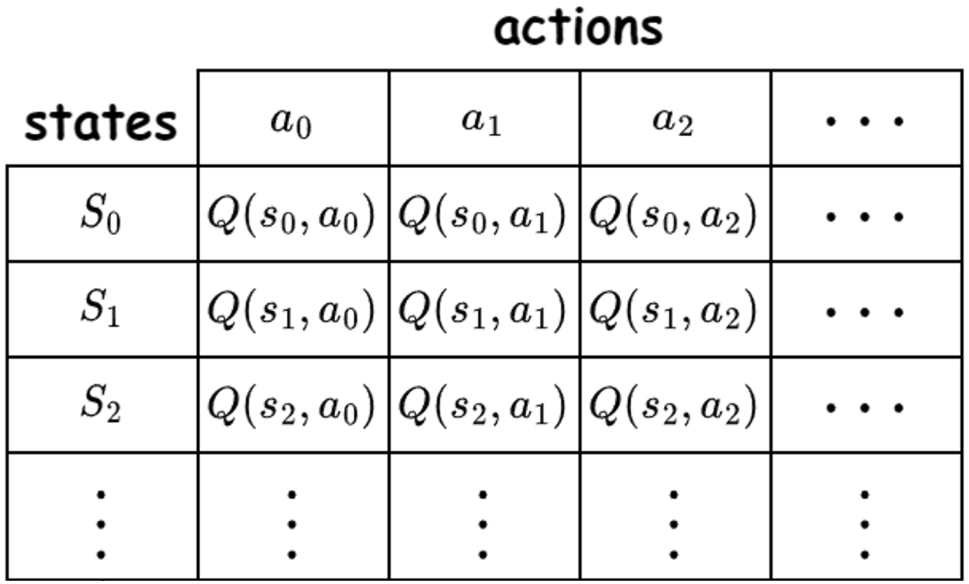
- How do we find the *optimal* policy π_* of our agent?

$$\pi_*(a \mid s) = \begin{cases} 1 \quad if \quad a = \underset{a \in \mathcal{A}}{\operatorname{argmax}}\, q_*(s,a) \\ 0 \quad otherwise \end{cases}$$

Q-Learning

- Tabular Method.
- Epsilon greedy behaviour policy.





actions

states	a_0	a_1	a_2	$\cdots$
s_0	$Q(s_0,a_0)$	$Q(s_0,a_1)$	$Q(s_0,a_2)$	$\cdots$
s_1	$Q(s_1,a_0)$	$Q(s_1,a_1)$	$Q(s_1,a_2)$	$\cdots$
s_2	$Q(s_2,a_0)$	$Q(s_2,a_1)$	$Q(s_2,a_2)$	$\cdots$
$\vdots$	$\vdots$	$\vdots$	$\vdots$	$\vdots$

$$Q(S_t,A_t) \leftarrow Q(S_t,A_t) + \alpha\left(\overbrace{\underbrace{R_{t+1} + \gamma \max_a Q(S_{t+1},a)}_{\text{TD target}} - Q(S_t,A_t)}^{\text{TD error}}\right)$$

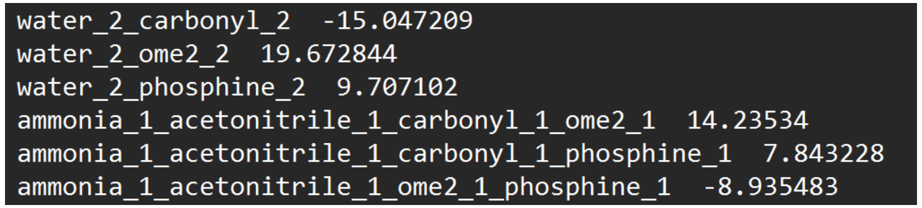
$$Q(s,a) \longrightarrow q_*(s,a)$$

Our Problem

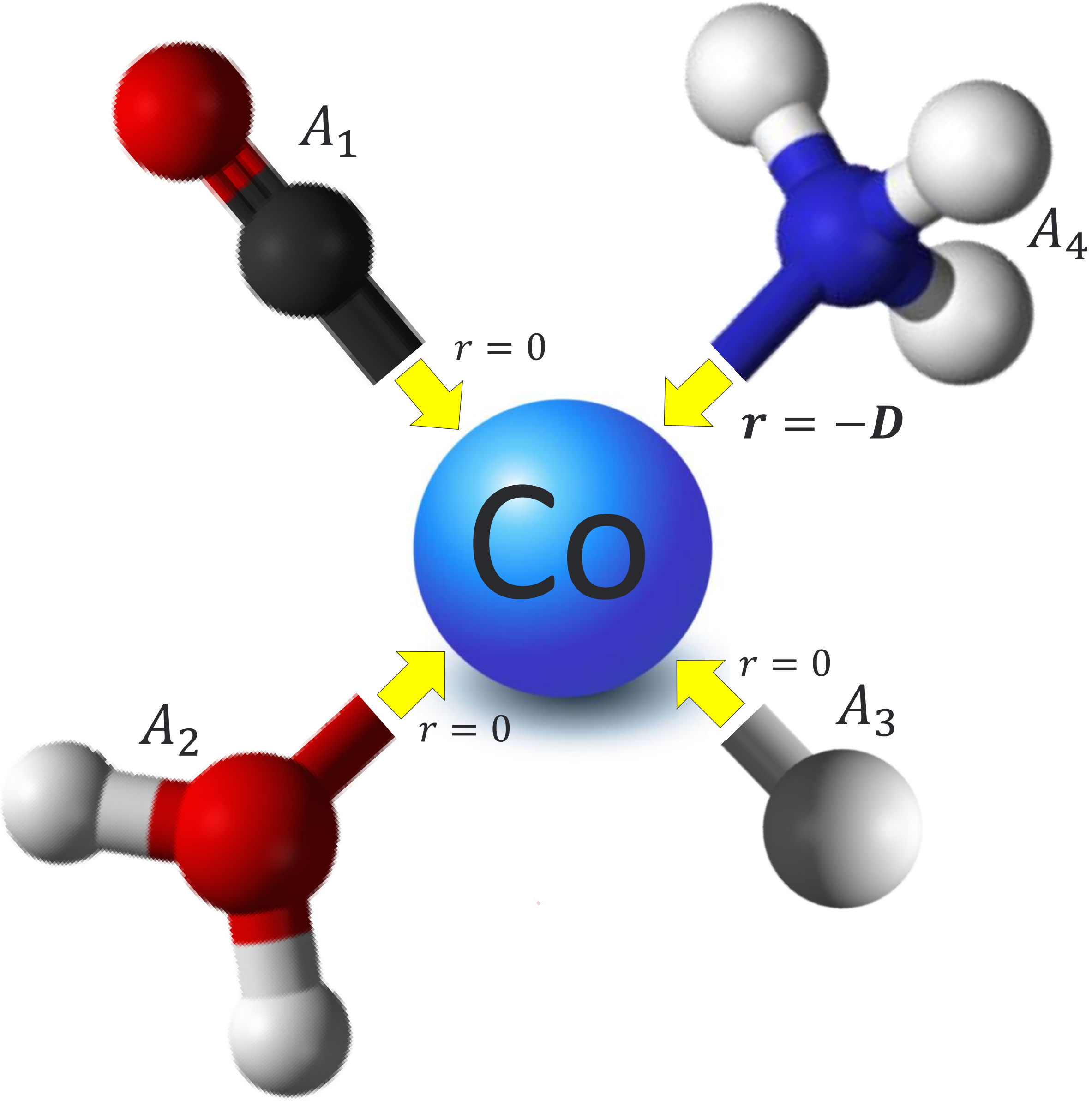
- Construct molecules by adding ligands to a central metal atom.

- Very challenging MDP!
- ➢ Very few no. steps
- ➢ No new info until end of episode

- Data



```
water_2_carbonyl_2  -15.047209
water_2_ome2_2  19.672844
water_2_phosphine_2  9.707102
ammonia_1_acetonitrile_1_carbonyl_1_ome2_1  14.23534
ammonia_1_acetonitrile_1_carbonyl_1_phosphine_1  7.843228
ammonia_1_acetonitrile_1_ome2_1_phosphine_1  -8.935483
```



Synthetic Data

$$n_{states}(N,M) = 1 + \sum_{m=1,\,\dots\,,\,M} \binom{m+N-1}{N-1}$$

$\boldsymbol{N}$ = no. ligands to choose from

$\boldsymbol{M}$ = no. ligands in molecule

M \ N	10	20	30
4	1,001	10,626	46,376
6	8,008	230,230	1,947,792
8	43,758	3,108,105	48,903,492

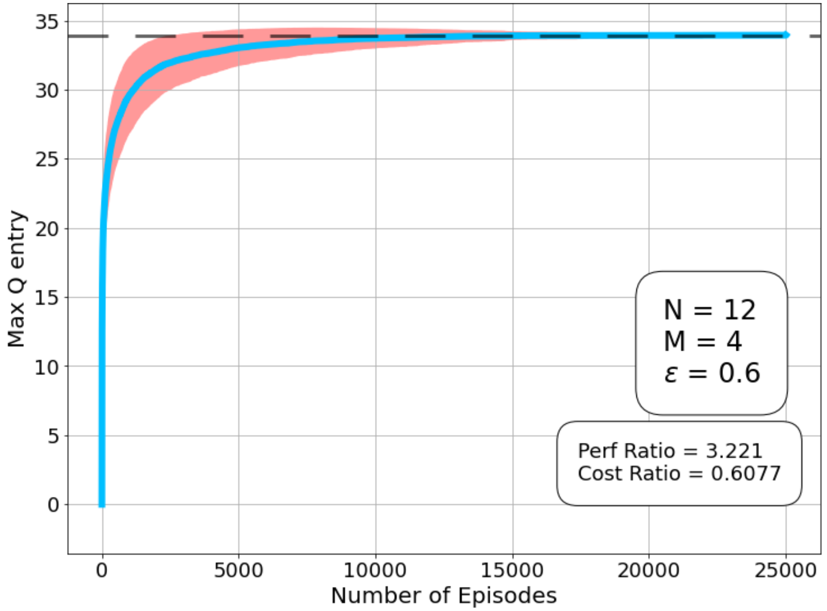
$$L_i \in [-10, 10]$$

$$R_{final} = \sum_{i=1,\dots,M} L_i \quad \Longrightarrow \quad \underbrace{[A, A, \dots, A]}_{M\ times}\ \textbf{best}$$

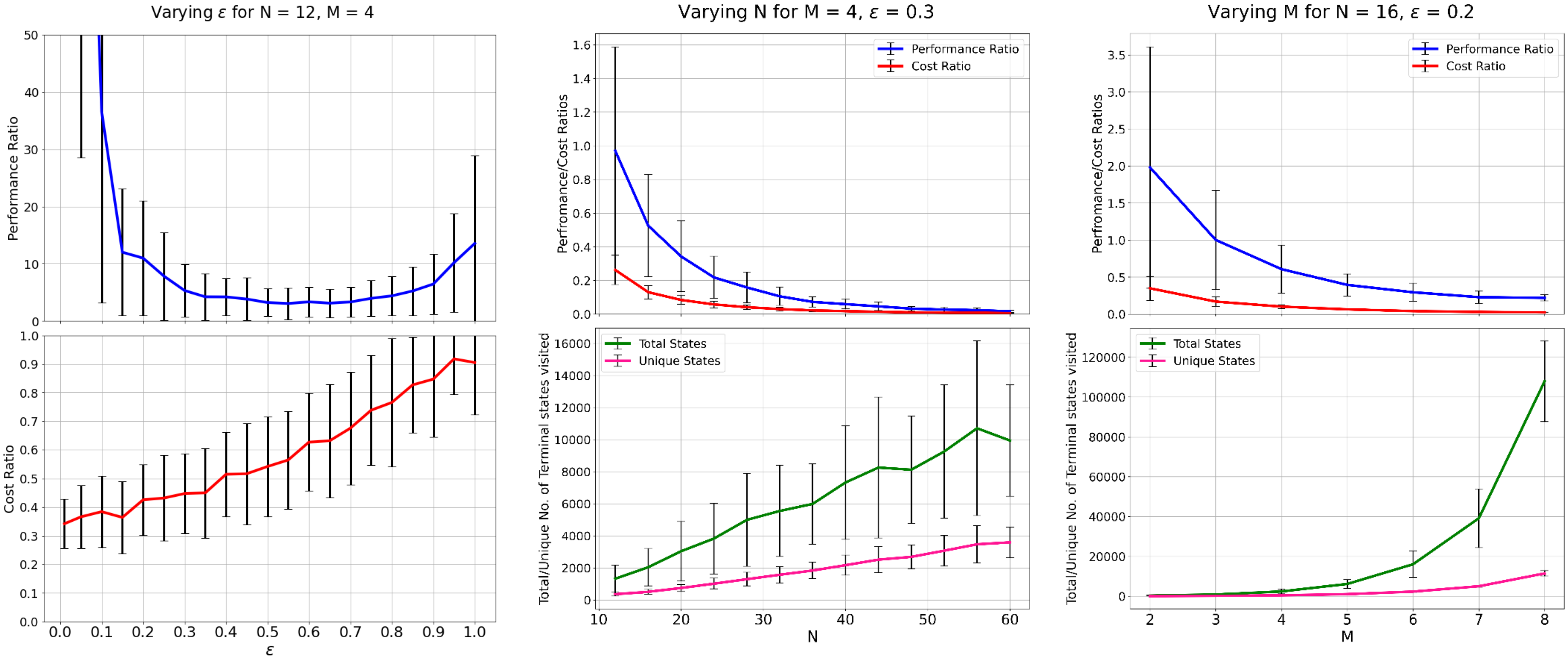
$$\text{Performance Ratio} = \frac{\text{Total no. Terminal States Reached}}{\text{Terminal State Space Size}}$$

$$\text{Cost Ratio} = \frac{\text{Unique no. Terminal States Reached}}{\text{Terminal State Space Size}}$$

Max Q entry vs no. episodes, 1000 runs

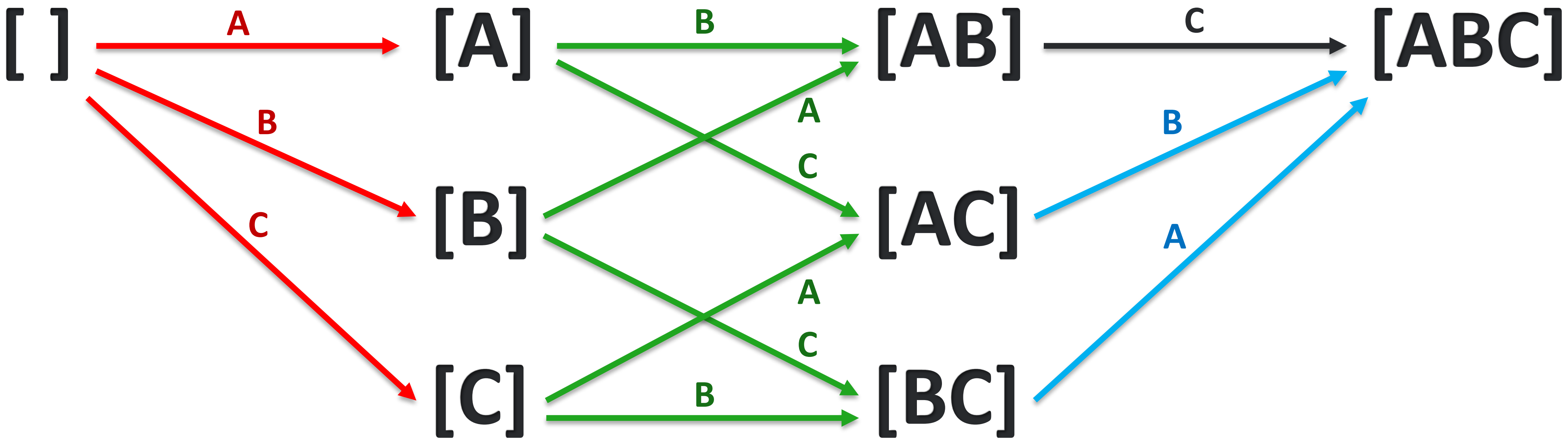


Synthetic Data



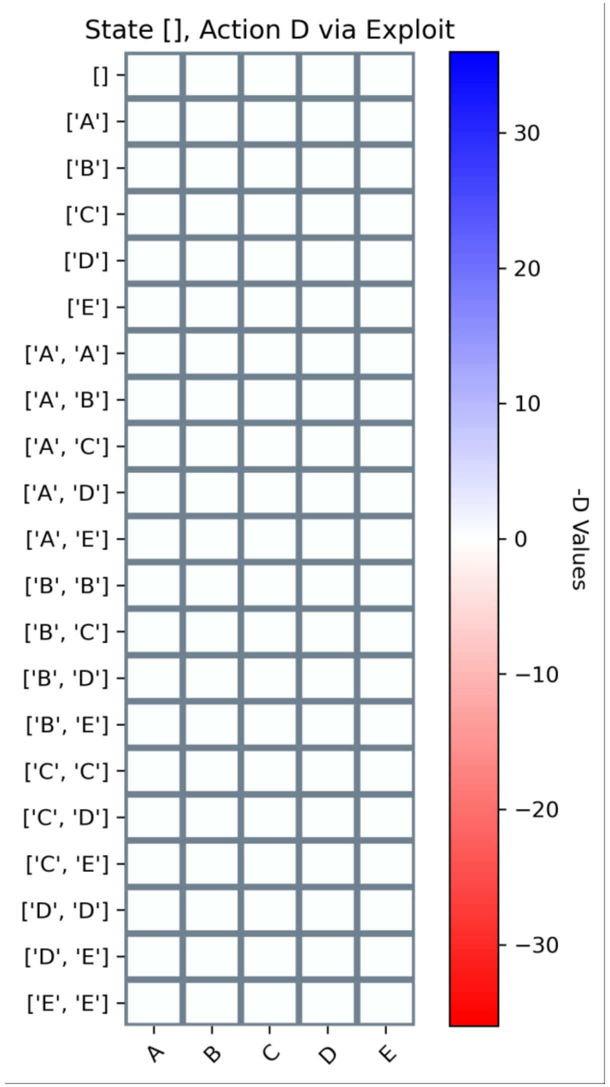
Algorithmic improvements

Order-invariance when adding ligands ⟹ Multiple Q updates per step

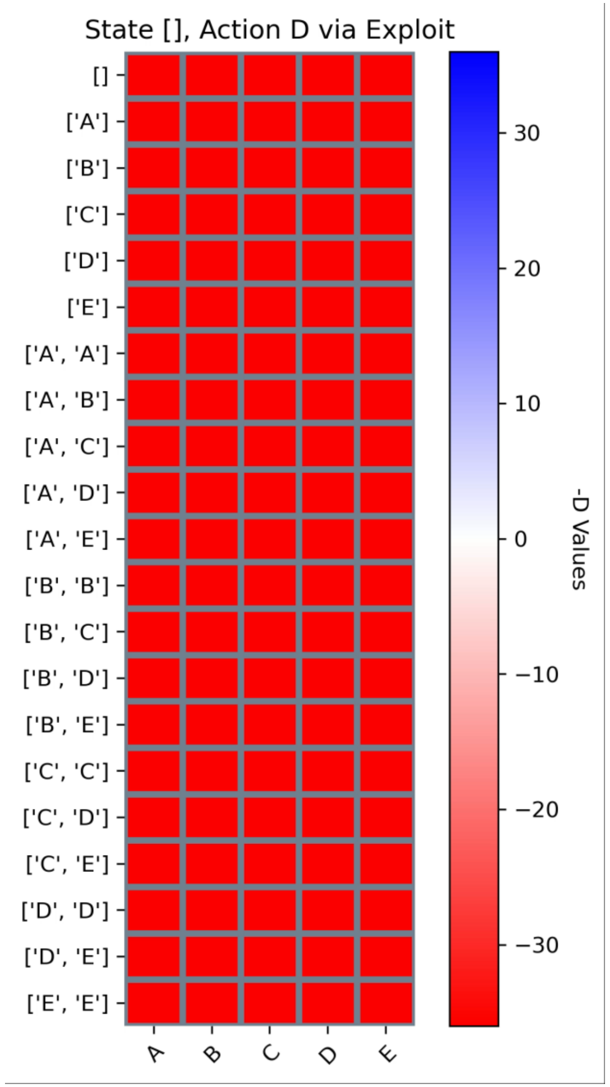


Algorithmic improvements

Vanilla



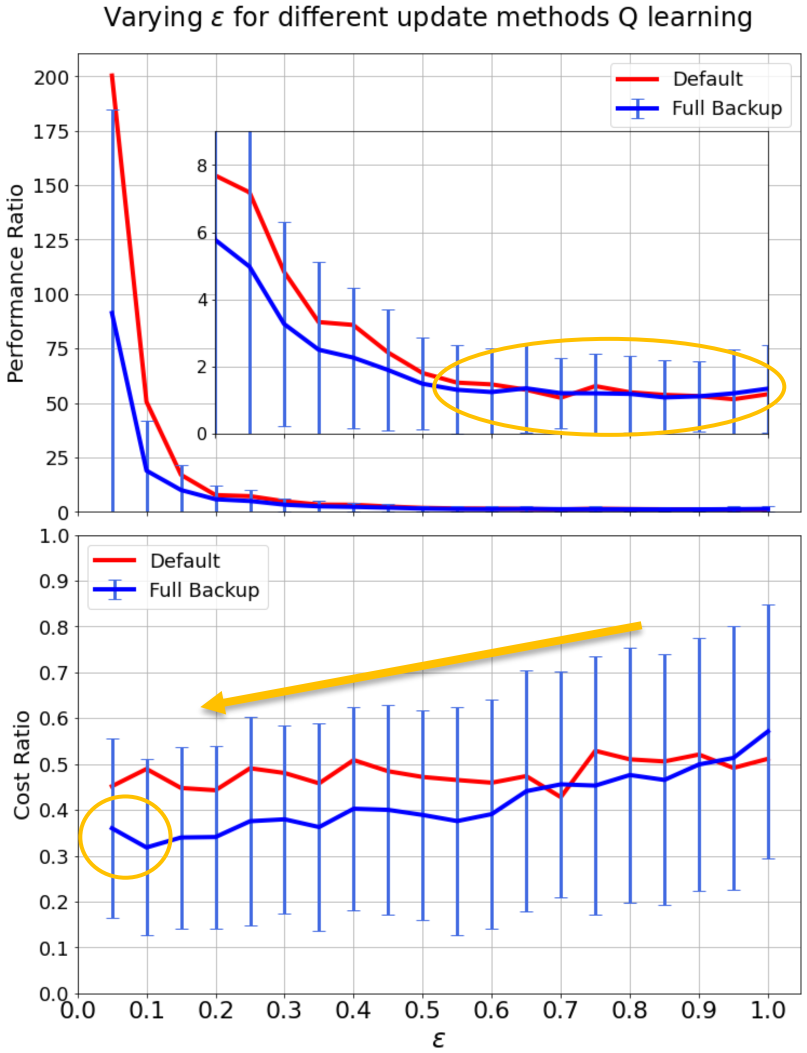
Full backup updates

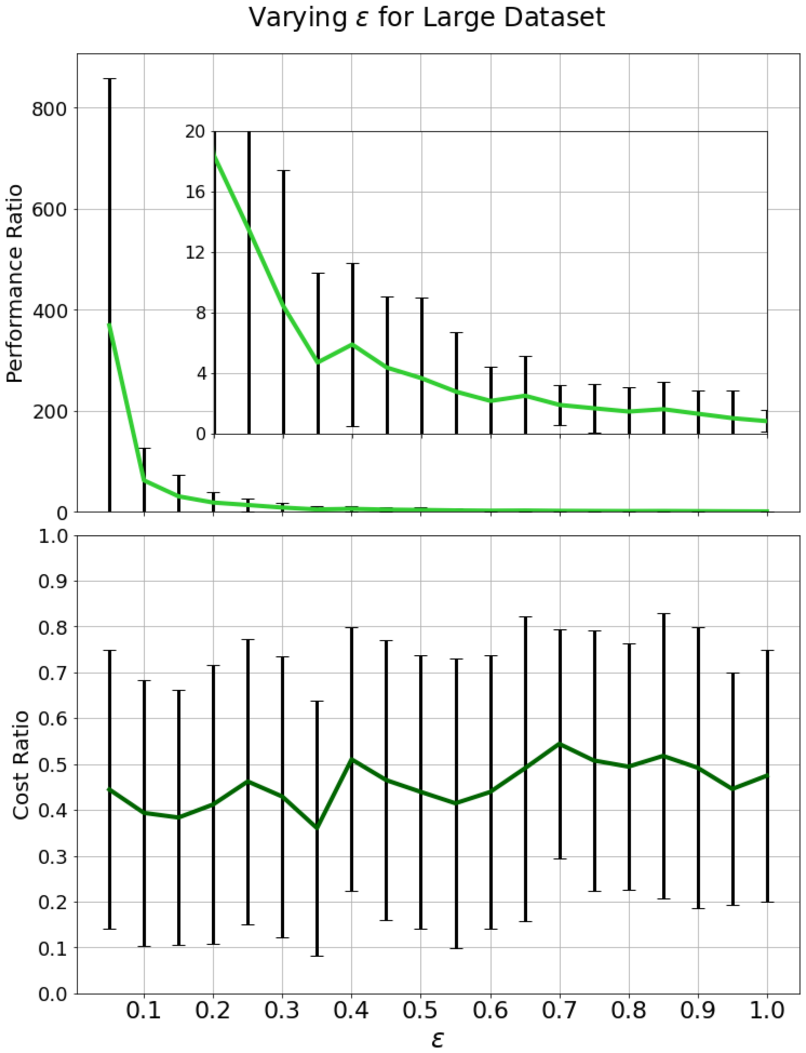


Physical data

Dataset 1:

$$N = 13$$
$$M = 4$$
$$n_{molecules} = 766$$





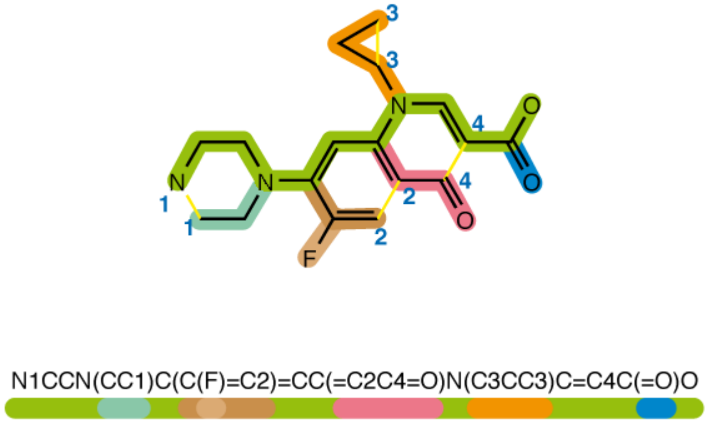
Dataset 2:

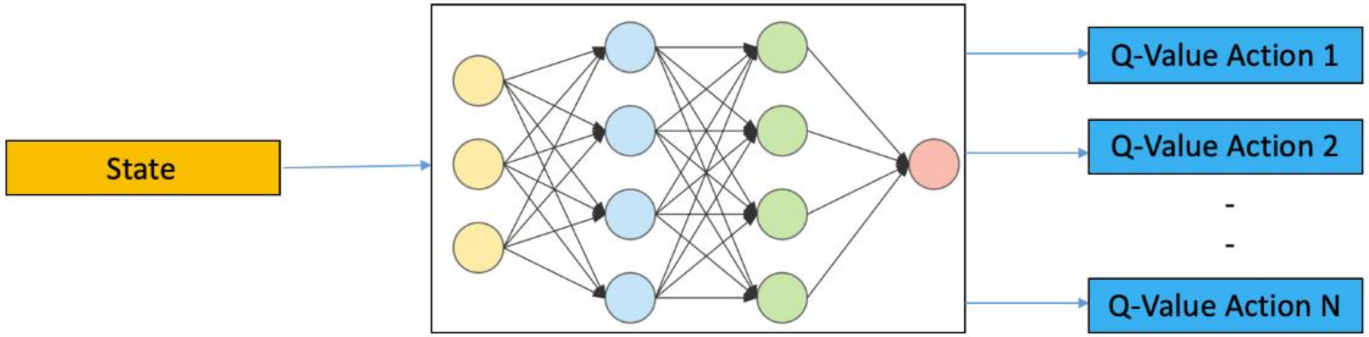
$$N = 206$$
$$M = 2$$
$$n_{molecules} = 21{,}321$$

Next Steps

- Return to synthetic data with more complexity/correlation between ligands in the reward function
  - ➢ Too little of no. steps?
  - ➢ Pattern too complicated?

- Introduce featurisation and use deep Q-Learning:
  - ➢ SMILES
  - ➢ Coordinates
  - ➢ Bispectrum Components





Thank you!

References:

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