MLIP for Excited State Potential Energy Surfaces

Background

- Molecules exist in the lowest energy state (ground state), using light we can excite these to higher energy states (excited states)
- This is called photochemistry and it underpins processes such as photosynthesis, solar cell conversion and vision

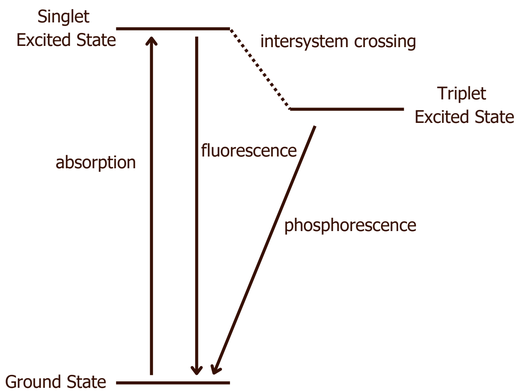
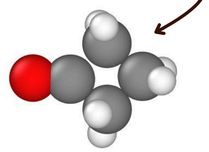


figure 1 - Simple Jablonski showing absorption and emission (flourescence and phosphorescence) of light

- Currently, to simulate these processes we use ab-initio methods (DFT or Post-HF) which are computationally expensive
- Advances in Machine Learning Interatomic Potentials (MLIP) means we can accelerate these simulations for the ground state
- Which leaves the question, what about excited states?

Methodology

- Using a flavour of MLIP called MACE [1], we aim to map the excited state potential energy surfaces (PES) based of its geometry
- Training data 195 configurations of Cyclobutanone labelled with ab-initio calculations (TD-DFT(PBE0)) [2]
- Each configuration consists of geometries, energies and forces for 4 surfaces (1 ground + 3 excited states)



- Using a Multi-head model, for one geometry we can have 4 outputs relating to each surface

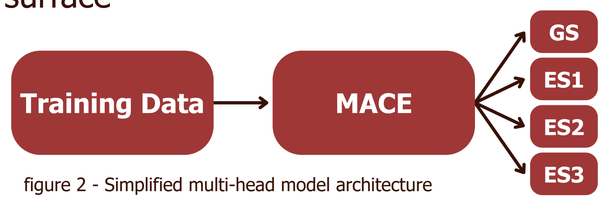


figure 2 - Simplified multi-head model architecture

- Once we have trained a model, it can now be tested on some reaction coordinates of Cyclobutanone

[1] Batatia, I. (2024). A foundation model for atomistic materials chemistry. https://arxiv.org/pdf/2401.00096v2
[2] Eng, J. (2024). The photochemistry of Rydberg-excited cyclobutanone: Photoinduced processes and ground state dynamics. J. Chem. Phys, 160(15). https://doi.org/10.1063/5.0203597

Results

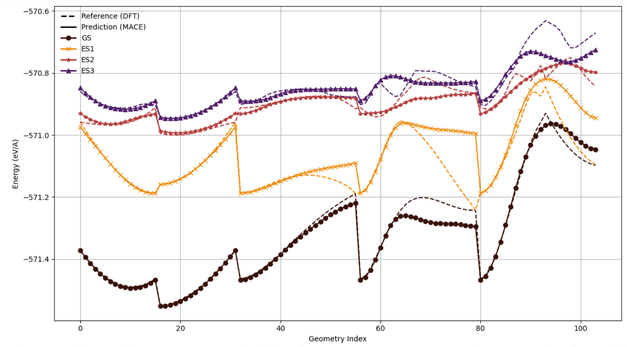


figure 3 - MACE prediction against DFT reference reaction coordinates plot (model test)

- By comparing MACE predictions to ab-initio, we can see that our model can predict the PES
- However, Important challenges in modelling state crossings. highlighted in figures a-d:

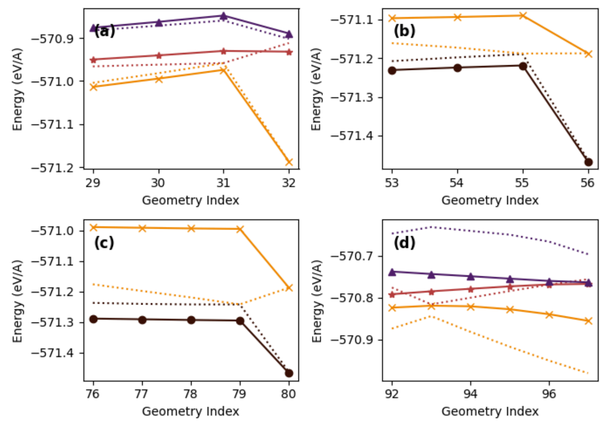


figure 4 - a) Conical Intersection (ES1/ES2) b) CI1(GS/ES1) c) CI2(GS/ES1) d) ES1/ES2/ES3 close together

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