Excited State Machine Learning for Chromophores in Complex Environments

Author: Carlo Maino, Supervisor: Dr. N.D.M. Hine

Centre for Doctoral Training in Modelling of Heterogeneous Systems, University of Warwick, CV4 7AL

email: carlo.maino@warwick.ac.uk

Overview

A major challenge in modern materials modelling techniques is the prediction of the optical properties of chromophores in complex environments. This problem is inherently multiscale. Consider a system such as a dye in solvent: the optical properties dependant on photoexcited processes act on a ps-ns timescale, the dye needs to be sampled on a long ns-μs timescale, coupled with the long-ranged solvent interactions on a scale of 10s of nm.

Progress on the prediction of optical properties has been made using linear-scaling time-dependant DFT[8,11]. However, the need for sampling over large time-scales combined with large length-scales means the computational demand remains very high. This project will use Machine Learning (ML) to attempt to accelerate the current state-of-the-art techniques used in optical properties prediction.

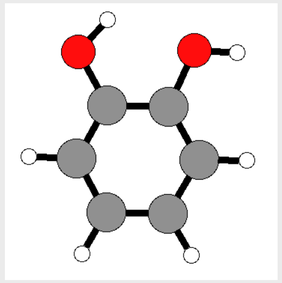

Fig. 1: Catechol

Goals

As an initial target for this project a protocol for generating excited state potential energy surfaces (PES) of organic molecules will be produced. An exemplar system that will be treated as a case study for this project is photodissociation of the molecule catechol[10,4].

Investigating ML Toolkits

1. AMP[5]
2. SchNetPack[7]: in particular SchNOrb[6] and SchNarc[9]
3. MLatom[3]
4. GAP[1]: with particular interest in recent work on organic molecules[2]

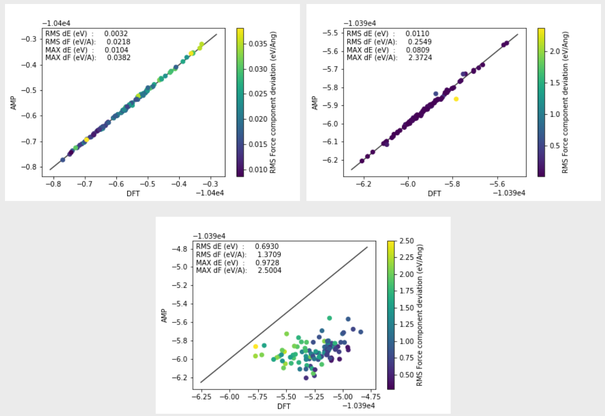

Fig. 2: AMP Calculators trained on AIMD dataset thermostated at 300K, tested on an unseen data. Clockwise from top left: Ground State, First Excited State, Second Excited State

Excited State PES

- Training Neural Network potentials off DFT data for Ground and Excited States of catechol
- Some preliminary results shown in Fig. 2

Binding Curve

- Using a Machine Learned PES, the binding curve for the H^+-dissociation of catechol following absorption from UV-light will be investigated.
- Enhanced sampling will be necessary to retrain around the dissociation pathway to improve the binding curve and PES.

Fig. 3: Exploring the Dissociation Pathway. As we extend the O-H bond length of catechol, we move along the red arrow, however we must also explore the sides of this potential (blue arrows).

Future...

- Further investigations into solvents
- Prediction of excited state lifetimes using MLMD

[1] Bartók, A. P., et al. 2010, 'Gaussian approximationpotentials: the accuracy of quantum mechanics, without the electrons.',Physical reviewletters104 13, 136403. [2] Cole, D., et al. 2020, 'A machine learning based intramolecular potential for a flexible organic molecule', Faraday Discuss.pp. –,URL:http://dx.doi.org/10.1039/D0FD00028K [3] Dral, P. O. 2019, 'Mlatom: A program package for quantum chemical research assisted by machine learning',Journal of computational chemistry. [4] Gerhards, M., et al. 1996, Structure and vibrations of catechol and catechol h2o(d2o) in the s0 and s1 state. [5] Khorshidi, A. and Peterson, A. A. 2016, 'Amp: A modular approach to machine learningin atomistic simulations',Comput. Phys. Commun.207, 310–324. [6] Schütt, K. T., et al. 2019,'Unifying machine learning and quantum chemistry with a deep neural network for molecular wavefunctions',Nature Communications10. [7] Schütt, K. T., et al. 2019, 'Schnetpack: A deep learning toolbox for atomistic systems.',Journal of chemical theory and computation15 1, 448–455. [8] Turner, M. A., et al. 2019, 'Determination of secondary species in solution through pump-selective transient absorption spectroscopy and explicit-solvent tddft.',The journal of physical chemistry. A123 4, 873–880. [9] Westermayr, J., et al. 2020, 'Combining schnet and sharc:The schnarc machine learning approach for excited-state dynamics',The journal of physical chemistry letters. [10] Young, J. D., et al. 2015, 'Torsional motion of the chromophore catechol following the absorption of ultraviolet light.',Physical reviewletters114 23, 233001. [11] Zachlsdorff, T. J. and Isborn, C. M. 2019, 'Modeling absorption spectra of molecules insolution',International Journal of Quantum Chemistry119