Choosing the functional for TD-DFT calculations with the computer oracle DELFI

DAVIDE AVAGLIANO[1,2], MARTA SKRETA[1], SEBASTIAN ARELLANO-RUBACH, ALAN ASPURU-GUZIK[1]

[1]*University of Toronto, 80 St. George Street, Toronto, ON, Canada*
[2]*Chimie ParisTech, PSL University, CNRS, Institute of Chemistry for Life and Health Sciences (iCLeHS UMR 8060), F-75005 Paris, France*

davide.avagliano@chimieparistech.psl.eu

ABSTRACT

Choosing the right exchange-correlation functional to be used in density functional theory (DFT) calculations is a non-trivial yet fundamental task in computational chemistry. The choice is particularly hard for excited states calculations when using the time-dependent formulation of DFT (TD- DFT). Machine learning could potentially offer an efficient solution to this problem, however, design an architecture for this task is non-trivial [1]. In this contribution, I will introduce DELFI (Data-driven EvaLuation of Functionals by Inference), a computer oracle to recommend the best functional to use to calculate the excited states of one or a set of molecules [2]. DELFI is a graph attention network trained on quality scores calculated for 38 among the most common functionals of different types and rungs, calculated for a dataset of more than 21.000 organic molecules. This dataset was publicly released and contains almost 4 million points to be used in future applications for machine learning for excited states calculations [3]. The scores of the functionals were assigned following a scoring system we developed to quantitatively define the accuracy of an excited state calculation. This includes at the same time the energy, character, order, and intensity of a manifold of states in comparison to a higher accuracy method. I will explain the scoring system and show the validation of DELFI on a set of organic chromophores and in choosing the best functional on a large dataset of organic photovoltaic molecules. Additionally, I will compare the predictions of DELFI with the choices of expert users, also involving the audience in the poll. Finally, I will show a corresponding web application that allows to easily run DELFI and analyze its results, alleviating the hurdle of choosing the right functional for TD-DFT calculations.

References

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