Julia WESTERMAYR

MolTag Thesis Supervisor: Leticia GONZÁLEZ, Institute of Theoretical Chemistry, University of Vienna

MolTag Co-Supervisor: Chris OOSTENBRINK, Institute of Molecular Modeling and Simulation, University of Natural Resources and Life Sciences, Vienna

Associated MolTag student, 3rd Funding Period

Defense: September 24, 2020

PhD Thesis title: "Machine Learning for Excited-State Molecular Dynamics Simulations"

With the rise of machine learning in quantum chemistry, many exciting opportunities have emerged to advance the research field of photochemistry. However, the high complexity and computational efforts associated with a description of coupled electrons and nuclei complicate an application of machine learning techniques, such that this research field remained mainly untouched until the beginning of this work. The main goal of this thesis is to employ machine learning in the simulation of photodynamics. To this aim, machine learning models are developed to describe a manifold of molecular excited-state potentials of different spin multiplicities, their derivatives, and couplings thereof. Central to this thesis is the identification of key obstacles in applying efficient, yet accurate machine learning for photodynamics and overcoming the following hurdles:
(1) Excited-state properties carry an arbitrary sign that prohibits a meaningful fitting. These properties can be rendered learnable with a so-called phase correction algorithm developed in this work. As an alternative, a machine-learning intrinsic solution is implemented in a new approach called SchNarc.
(2) Couplings between electronic states are often missing in quantum chemistry codes. Therefore, approximations based on machine-learned potentials are provided.
(3) Due to the lack of machine learning studies on this topic, many open questions remain on how to best treat excited states. Thus, various models and molecular representations are compared.
(4) A cost-effective training set generation is required that still allows for a desired accuracy.
In order to meet this requirement, an existing efficient scheme for the ground state is adapted for the excited states. The performance of all these novel strategies and methods is investigated using several small molecules. One big achievement of this work is to enable long time scale photodynamics with neural networks for the methylenimmonium cation. The complex photochemistry of the amino acid tyrosine illustrates the challenges machine learning models still need to overcome, but demonstrates the scopes and possibilities of machine learning for excited electronic states of molecules.