VISTA

VISTA Seminar

Seminar 65

March 20, 2024

10:00 am – 11:30 am EST / 3:00 – 4:30 pm GMT London / 4:00 pm – 5:30 pm CET Paris / 11 pm CST Beijing

TOC:

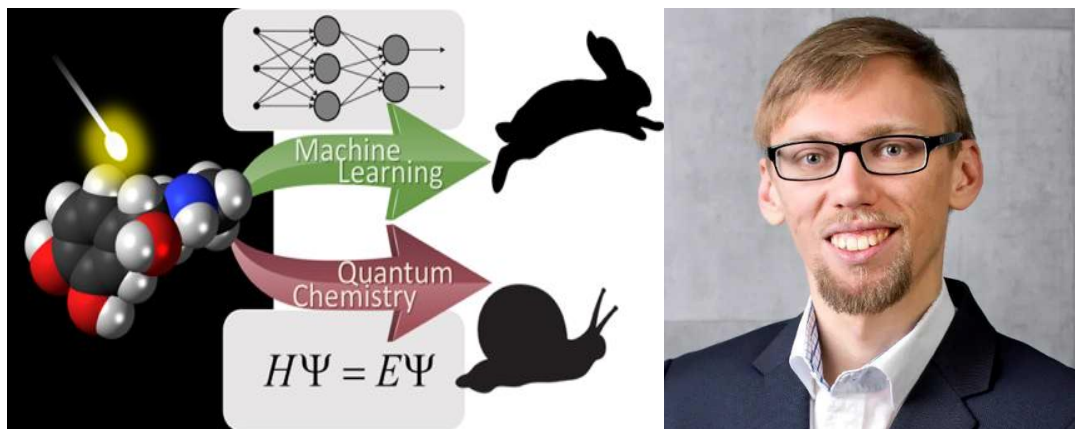# Towards more accessible excited-state simulations with AI

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Machine learning can greatly speed up quantum chemical simulations.[1]

I will present our ongoing journey towards making excited-state simulations more accessible with the help of AI/ML. We started it with the goal of accelerating nonadiabatic excited-state molecular dynamics which are extremely slow and resource-consuming in the on-the-fly surface-hopping framework. Our early works explored the potential gains and possible solutions for ML-accelerated nonadiabatic dynamics[2] and showcased the practical possibility of such simulations[3]. In this context, we developed an efficient approach for substantially reducing the cost of calculating excited-state properties for initial conditions which can be used to obtain precise one-photon UV/vis absorption spectra[4] (later, we developed an efficient universal approach for predicting related two-photon absorption spectra[5]). Interpolating between the initial conditions can be efficiently done with kernel methods in agreement with our systematic study of the optimal choice of ML models for single-molecule PES.[6] However, for dynamics requiring more data, NN approaches turn out to be a good practical choice, both in the context of developing universal Δ-learning improved semi-empirical QM method AIQM1 with a good accuracy and low cost for excited-state simulations[7] and for creating specialized models for nonadiabatic dynamics of specific systems[3,8]. AIQM1 has been successfully applied for excited-state geometry optimizations[7,9] owing to our semi-empirical baseline method[10] and we are currently exploring its prowess for nonadiabatic dynamics. Specialized models can be used for cost-efficient

generation of the nonlinear time-resolved spectra from nonadiabatic dynamics trajectories.[8]

Beyond on-the-fly surface hopping dynamics, we explored ML approaches for accelerating,[11-12] learning trajectories as a function of time,[13] and learning the entire trajectories[14] of the quantum dissipative dynamics.

This research stimulated the development of integrated ML software ecosystems for excited-state simulations based on MLatom,[15] Newton-X,[16] and MLQD[17], and an increasing number of such simulations can be performed online at the https://XACScloud.com. General discussion about the progress in this field is given in our review[1] and book chapters[18-20].

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MLatom 3: A Platform for Machine Learning-Enhanced Computational Chemistry Simulations and Workflows. *J. Chem. Theory Comput.* **2024,** *20*, 1193–1213. See MLatom.com @ XACScloud.com.

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Implementation of energy and gradient for the TDDFT- approximate auxiliary function (aas) method

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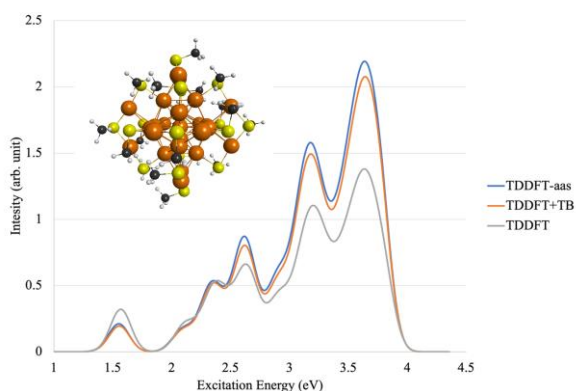
Gold and silver nanoparticles can exhibit unique physical and chemical properties such as plasmonic resonances. These types of plasmonic nanoparticles are usually large, which leads to difficulty in computing their optical properties using the TDDFT method. In this work, we have implemented the TDDFT-aas method, which is an approximate TDDFT method. In this method, instead of calculating the exact two-center electron integrals in the K coupling matrix when solving the Casida equation, we approximate the integrals, thereby reducing the computational cost. Compared with the related TDDFT+TB method, a new type of gamma function is used when calculating the K coupling matrix, which does not depend on the tight binding parameters. The calculated absorption spectra of silver and gold nanoparticles using TDDFT-aas show good agreement with TDDFT+TB results. In addition, we have also implemented the analytical excited-state gradients for the TDDFT-aas method, which makes it possible to calculate the emission energy of the systems.

How to connect

Alexey Akimov is inviting you to a scheduled Zoom meeting.

Topic: VISTA, Seminar 65
Time: Mar 20, 2024 10:00 AM Eastern Time (US and Canada)

Join Zoom Meeting
https://buffalo.zoom.us/j/98974911660?pwd=QlBIYWwzWVFtWFBPMi92OXpCekhIZz09

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