VISTA

VISTA Seminar

Seminar 53

June 7, 2023

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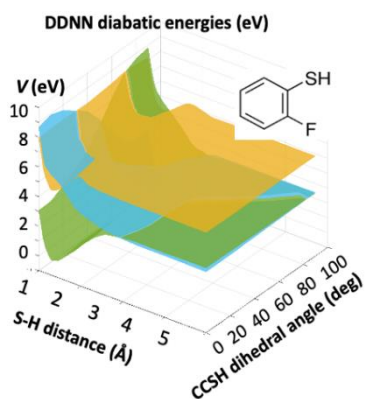
TOC:

Improved methods for semiclassical calculations of electronically nonadiabatic processes

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Electronically nonadiabatic processes are difficult because:
- One must include coherence and decoherence (affordably) in dynamics.
    - trajectory surface hopping is not good enough
- Excited states are closely coupled and strongly correlated.
    - need multiple surfaces and couplings
    - need multi-state ("state interaction"), multireference methods
- Simulations require ensemble averages and long times.
    - need to keep cost down

This seminar addresses four difficulties and four ways to overcome them.

Problem	Solution
Direct dynamics	
Multireference *dynamic correlation* is expensive.	MS-PDFT, L-PDFT [1]
Trajectory surface hopping does not balance *coherence* and *decoherence*.	Coherent switching with decay of mixing [2]
Nonadiabatic couplings in Born-Oppenheimer basis are both *expensive* and *nonphysical*.	Curvature-driven couplings [3]
Dynamics with fitted potentials and couplings	
Allow *more sampling* and *longer-time* simulations, allow quantum dynamics but *hard*	Direct diabatization by neural network [4]

References:
[1] C. Zhou et al. Chem. Sci. 13, 7685 (2022). M.R. Hennefarth et al. DOI:10.1021/acs.jctc.3c00207
[2] Y. Shu et al. JCTC 16, 3464 (2020). Y. Shu & D.G. Truhlar, JCTC **19**, 380 (2022).
[3] Y. Shu et al., JCTC 18, 1320 (2022). L. Zhang et al. JCTC 18, 7073 (2022).
[4] Y. Shu et al., JCTC17, 1106 (2021). Z. Varga et al. Electronic Structure 4, 047002 (2022).
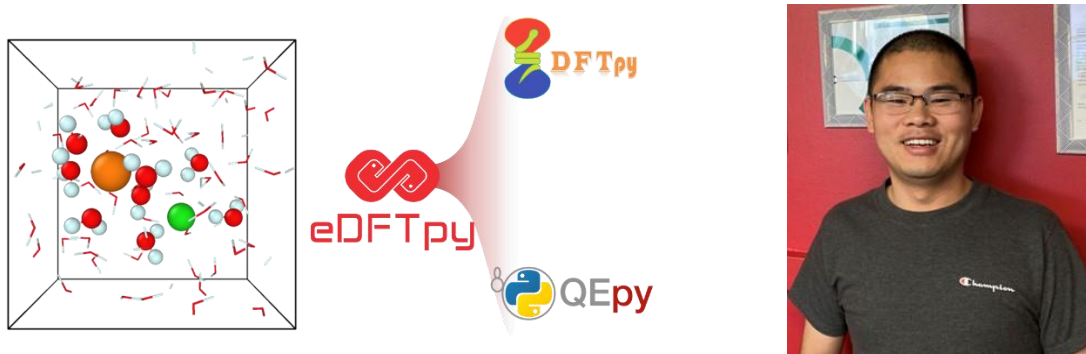
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Ab-initio electronic structure methods for large-scale simulations

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Subsystem Density Functional Theory (DFT) is a divide-and-conquer approach to describe the electronic structure of large systems by partitioning them into smaller, interacting subsystems. The interaction among the subsystems is evaluated by non-additive, orbital-free density functionals which need to be approximated. Two issues have lingered. First, the available approximations constrain the application of subsystem DFT primarily to weakly interacting subsystems. Second, when a single subsystem becomes excessively large, the computational cost is dominated by that subsystem, resulting in minimal advantages compared to the conventional Kohn-Sham DFT simulation. In our work, both issues are tackled head-on. First, we devised an adaptive density embedding method that facilitates the merging and/or splitting of subsystems when they exhibit strong or weak interactions, enabling an efficient redistribution of workload and data. Second, we established a true multi-scale subsystem DFT approach by employing orbital-free DFT to describe large metallic subsystems that would otherwise be too computationally expensive for off-the-shelf DFT solvers. Our object-oriented Python implementations encapsulate these methods, providing a widely applicable, massively parallel, and quasi black-box subsystem DFT approach that is both accurate and efficient.

How to connect

Alexey Akimov is inviting you to a scheduled Zoom meeting.

Topic: VISTA, Seminar 53
Time: Jun 7, 2023 10:00 AM Eastern Time (US and Canada)

Join Zoom Meeting
https://buffalo.zoom.us/j/95688513999?pwd=OTBvczBqbTFjOGRmTWJqcHN2NGdUQT09

Meeting ID: 956 8851 3999
Passcode: 470524