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

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**Improved methods for semiclassical calculations**

**of electronically nonadiabatic processes**

Donald G. Truhlar

*Department of Chemistry, University of Minnesota, Minneapolis, MN 66455-0431*

*Email: Truhlar#umn.edu (replace # with the “at” sign)*

Chart, surface chart

Description automatically generated A person in a suit and tie

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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](https://doi.org/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).

**Ab-initio electronic structure methods for large-scale simulations**

Xuecheng Shao1, 2 and Michele Pavanello1, 2

*1Department of Physics, Rutgers University, Newark, NJ 07102, USA*

*2Department of Chemistry, Rutgers University, Newark, NJ 07102, USA*

*Email:* [*xuecheng.shao@rutgers.edu*](mailto:xuecheng.shao@rutgers.edu)

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Description automatically generated

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