VISTA Seminar

Seminar 73

July 10, 2024

10:00 am – 11:30 am EDT / 3:00 – 4:30 pm BST London / 4:00 pm – 5:30 pm CEST Paris / 10 pm – 11:30 pm CST Beijing

TOC:

Towards Efficient Simulations of Excited States in Macromolecules and Computational Design of Photoenzymes

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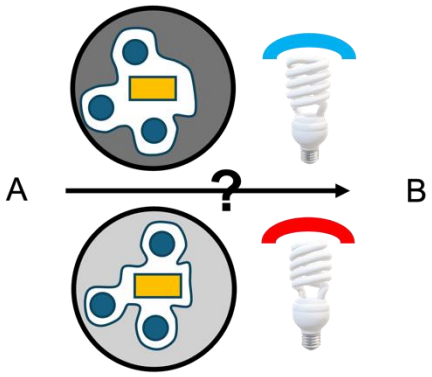

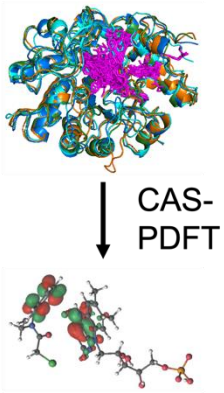

Accurate first-principles simulations of the electronic excited states of molecules and materials are usually computationally expensive, especially those that involve strong correlation. These simulations are nevertheless critical in the understanding and design of photophysical and photochemical processes in macromolecules, such as those in emerging protein-based photoredox catalysts. In this talk, I will discuss our recently developed strategies 1) to automate the active space selection of multireference methods for the high-throughput simulation of molecules and complexes with strong correlation,[1] and 2) to generate starting orbitals that can be useful for nonadiabatic dynamics with multireference methods. I will also discuss our recent results to use a combination of computational methods including molecular dynamics simulations, excited-state calculations using multireference methods, and machine learning to elucidate the spectral tuning mechanism of a new class of protein-based photoredox catalysts and to provide insights on their design.[2]

References:

[1] Kaufold, B. W.; Chintala, N.; Pandeya, P.; Dong, S. S. Automated Active Space Selection with Dipole Moments. *J. Chem. Theory Comput.* **2023**, *19* (9), 2469–2483.
[2] Carceller, J.; Jayee, B.; Page, C.; Oblinsky, D.; Chintala, N.; Mondragón-Solórzano, G.; Cao, J.; Alassad, Z.; Zhang, Z.; White, N.; Scholes, G.; Dong, S.; Hyster, T. Engineering a Photoenzyme to Use Red Light. ChemRxiv March 18, 2024. https://doi.org/10.26434/chemrxiv-2024-cjs5j.

Photoexcitation Dynamics in Perylene Diimides

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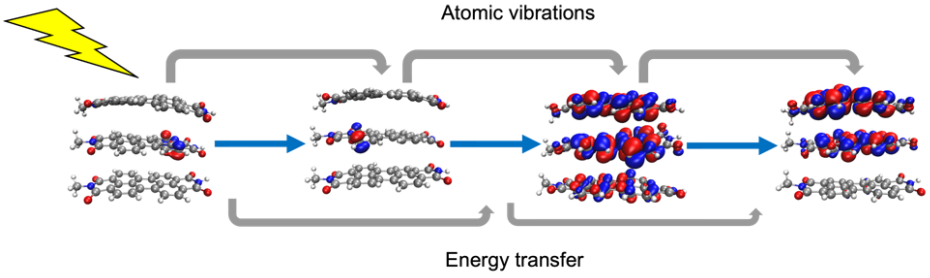

Perylene diimides (PDI) are promising materials for optoelectronics with excellent electrochemical, electrical and photophysical characteristics. In this work, we study the photo-induced excited-state dynamics of a PDI derivative using the Nonadiabatic EXcited state Molecular Dynamics (NEXMD) software that goes beyond the Born-Oppenheimer description of electron-nuclear interactions. By comparing the internal conversion processes of one to three stacked PDI molecules, we determine the role of inter-molecular interactions on the time-scales associated with energy transfer and exciton localization dynamics. We predict that stacking leads to enhanced energy decay because of decreased energy spacing between states. Additionally, studying the dynamics of the transition density, it is demonstrated that stacking impacts the localization of the exciton. For the dimer, the exciton quickly localizes and oscillates between two monomers, while the trimer can host long-time delocalization of the exciton. Lastly, vibrational normal mode analysis during the dynamics allows us to identify the intra- and inter-molecular modes that assist the electronic relaxation.

How to connect

Alexey Akimov is inviting you to a scheduled Zoom meeting.

Topic: VISTA, Seminar 73
Time: Jul 10, 2024 10:00 AM Eastern Time (US and Canada)

Join Zoom Meeting
https://buffalo.zoom.us/j/97473455930?pwd=PUoNP07g3WbG7XOpCYjy9rbnnbsrCm.1

Meeting ID: 974 7345 5930
Passcode: 521005