

VISTA Seminar

Seminar 3

9:30 – 11:00 am EDT / 1:30 – 3:00 pm GMT

TOC:

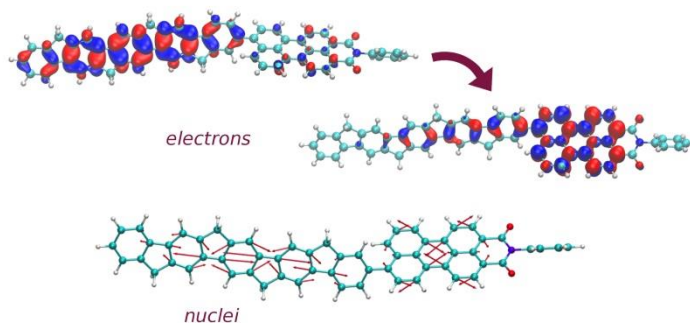
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Non-adiabatic excited state molecular dynamics: Identification, monitoring and freezing normal modes

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Abstract. Photoinduced electronic energy transfer in large multichromophoric molecular systems is naturally accompanied by intramolecular vibrational energy redistributions[1,2]. Ab initio molecular dynamics (AIMD) simulations analysed in terms of vibrational normal modes, is a widely used technique that facilitates understanding of complex structural motions and coupling between electronic and nuclear degrees of freedom[3]. Usually, only a subset of vibrations is directly involved in the process of interest. Herein, different complementary criteria are analyzed to systematically identify the subset of vibrational normal modes that actively participate on the internal conversion process of different multichromophoric conjugated molecules: chlorophylls[4], donor-acceptors[5], and dendrimers[6]. Energy transfer coordinates are analyzed in terms of state-specific normal modes defined according to the different potential energy surfaces (PESs) involved. On one hand, we identify those vibrations that contribute the most to the direction of the main driving force on the nuclei during electronic transitions, represented by the non-adiabatic derivative coupling vector between donor and acceptor electronic states. On the other hand, we monitor normal mode transient accumulations of excess energy and their intramolecular energy redistribution fluxes. We observe that the subset of active modes varies according to the PES on which they belong and these modes experience the most significant rearrangements and mixing. Besides, we apply normal mode constraints[7] in AIMD simulations as implemented in the non-adiabatic excited state molecular dynamics (NEXMD) code[8]. In this way, we directly measure the impact of normal mode constraints on the photoinduced energy transfer. Our results show that the electronic relaxation can be significantly slowed down by freezing a well-selected small subset of active normal modes characterized by their contributions in the direction of energy transfer. The application of these constraints reduces the non-adiabatic coupling between electronic excited states during the entire dynamical simulations. Furthermore, we validate reduced dimensionality models by freezing all the vibrations except a few active modes. Altogether, our analysis can be broadly used to underpin the role vibrational motion in a studied process and to formulate reduced models that describe essential physical phenomena.

References:

- [1] *"Non-adiabatic Excited State Molecular Dynamics: theory and applications for modeling photophysics in extended molecular materials"* T. Nelson, A. White, J. Bjorgaard, A. Sifain, Y. Zhang, B. Nebgen, Benjamin; S. Fernandez-Alberti, D. Mozyrsky, A. Roitberg, Adrian and S. Tretiak, Chem. Rev., 120, 4, 2215-2287 (2020).
- [2] *"Electronic Delocalization, Vibrational Dynamics and Energy Transfer in Organic Chromophores"* T. Nelson, S. Fernandez-Alberti, A. E. Roitberg, and S. Tretiak, J. Phys. Chem. Lett. 8, 3020–3031 (2017).
- [3] *"Coherent Exciton-Vibrational Dynamics and Energy Transfer in Conjugated Organics"* T R. Nelson, D. Ondarse-Alvarez, N. Oldani, B. Rodriguez-Hernandez, L. Alfonso-Hernandez, J. F. Galindo, V. D. Kleiman, S. Fernandez-Alberti*, A. E. Roitberg, Sergei Tretiak*, Nature Comm. Vol. 9, Article number: 2316 (2018).
- [4] *"Internal Conversion and Vibrational Energy Redistribution in Chlorophyll A"*, P. M. Shenai, S. Fernandez-Alberti*, W. P. Bricker, S. Tretiak, and Y. Zhao*, J. Phys. Chem. B 120(1), 49-58 (2016)
- [5] *"Vibrational energy redistribution during donor-acceptor electronic energy transfer: criteria to identify subsets of active normal modes"* L. Alfonso-Hernandez, S. Athanasopoulos, S. Tretiak, B. Miguel, A. Bastida, and S. Fernandez-Alberti, Phys. Chem. Chem. Phys. ,22, 18454-18466 (2020).
- [6] *"Analysis of state-specific vibrations coupled to the unidirectional energy transfer in conjugated dendrimers"*, Miguel A. Soler, Adrian E. Roitberg, Tammie Nelson, Sergei Tretiak, and Sebastian Fernandez-Alberti*. J. Phys. Chem. A, 116, 9802-9810,(2012)
- [7] *"Photoinduced dynamics with constrained vibrational motion: FrozeNM algorithm"* H. Negrin-Yuvero, V. M. Freixas, B. Rodriguez-Hernandez, G. Rojas-Lorenzo, S. Tretiak, A. Bastida and S. Fernandez-Alberti^a submitted (2020)
- [8] *"NEXMD Software Package for Non-adiabatic Excited State Molecular Dynamics Simulations"* W. Malone, B. Nebgen, A. White, Y. Zhang, H. Song, J. A. Bjorgaard, A. E. Sifain, B. Rodriguez-Hernandez, V. Manuel Freixas, S. Fernandez-Alberti, A. E. Roitberg, T. R. Nelson, and S. Tretiak, J. Chem. Theory Comput., J. Chem. Theory Comput. 16, 9, 5771–5783 (2020).

Charge Carrier Dynamics in Hybrid Perovskites: Insights from NAMD Simulations

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An in-depth understanding of the transfer, transport, and recombination processes of light-induced carriers provides us to design and discover functional photoactive materials for optoelectronic and photocatalytic devices. In this talk, I will discuss how nonadiabatic molecular dynamics (NAMD) with time-domain density functional theory methods at room temperature can help us to explore the charge dynamics in structurally complex materials such as halide perovskites. Our systematic study demonstrates that performance-limiting nonradiative carrier recombination processes greatly depends on the electron-phonon interactions induced by structural fluctuations and instantaneous charge localization in these materials. I will focus on a particular set of materials, 2D-halide perovskites to discuss the detailed role of spacer cation dynamics that strongly influences the optoelectronics of these systems. The computational insights gained from these methods allow us to outline a set of robust design principles for halide perovskites to strategically tune their optoelectronic properties.

**References:**

Ghosh *et al.* J. Mater. Chem. A, **2020**, Just Accepted (DOI: 10.1039/D0TA07205B)

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How to connect

Alexey Akimov is inviting you to a scheduled Zoom meeting.

Topic: VISTA, Seminar 3

Time: Oct 8, 2020 09:30 AM Eastern Time (US and Canada)

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103.122.166.55 (Australia)

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