VISTA Seminar

Seminar 37

May 11, 2022

10:00 am – 11:30 am EDT / 3:00 – 4:30 BST / 4:00 pm – 5:30 pm Paris

TOC:

Independent-Trajectory Mixed Quantum-Classical Approaches Based on Exact Factorization

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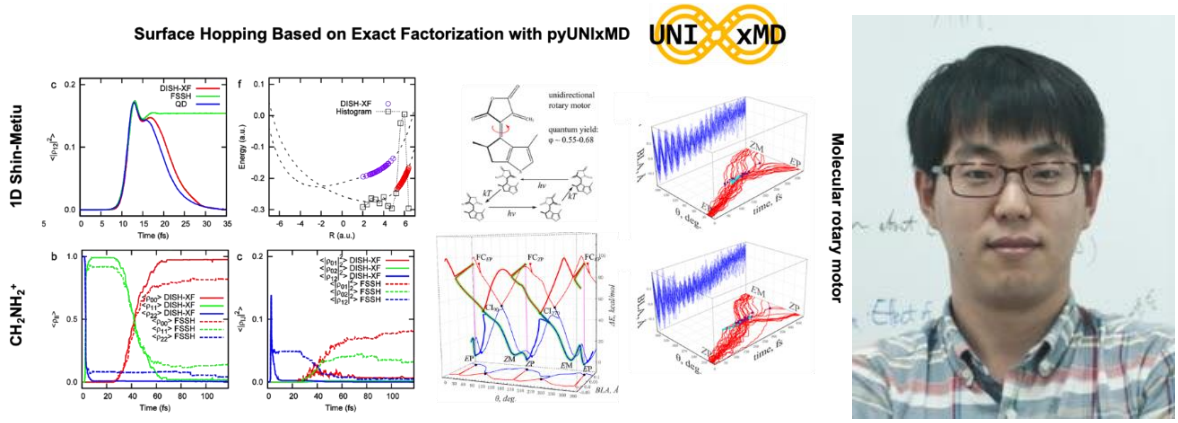

When multiple adiabatic electronic states are coupled to nuclear degrees of freedom, Born-Oppenheimer (BO) approximation breaks down. Especially when a molecular system absorbs a photon or multiple photons, we should treat nuclear motion beyond BO approximation to investigate nuclear wave packet splitting as well as quantum coherence. Statistical analysis based on multiple classical nuclear trajectories shows a nice description of BO populations. However, incorrect account for electron-nuclear correlations gives a wrong description for quantum coherence. Based on recent developments on exact factorization of a molecular wave function, a rigorous derivation of a correct electron-nuclear correlation is available which enables us to deal with proper quantum coherence and nuclear wave packet splitting. This correct electron-nuclear correlation contains not only the conventional nonadiabatic couplings with classical nuclear momenta but also couplings with quantum mechanical features of nuclear density. In this talk, I will present several research works to develop independent-trajectory mixed quantum-classical approaches based on exact factorization which are coupled to surface hopping [1] and Ehrenfest dynamics [2]. In addition, I will introduce a python-based program for excited state molecular dynamics, namely pyUNIxMD [3,4], which was developed by my group and various applications [5-7].

References:

[1] J.-K. Ha, I.S. Lee, S.K. Min, J. Phys. Chem. Lett., **2018**, 9, 1097.
[2] J.-K. Ha, S.K. Min, J. Chem. Phys., 2022 (in press) doi/10.1063/5.0084493.
[3] Lee, I.S.; Ha, J.-K.; Han, D.; Kim, T.I.; Moon, S.W.; Min, S.K., J. Comp. Chem. **2021**, 42, 1755.
[4] Kim, T.I.; Ha, J.-K.; Min, S.K., Top. Cur. Chem., **2022**, 380, 8.
[5] M. Filatov, S.K. Min, K.S. Kim, J. Chem. Theory Comput., **2018**, 14, 4499.
[6] M. Filatov, S.K. Min, K.S. Kim, J. Phys. Chem. Lett., **2018**, 9, 4995.
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Interacting trajectory ensemble in the framework of exact factorization

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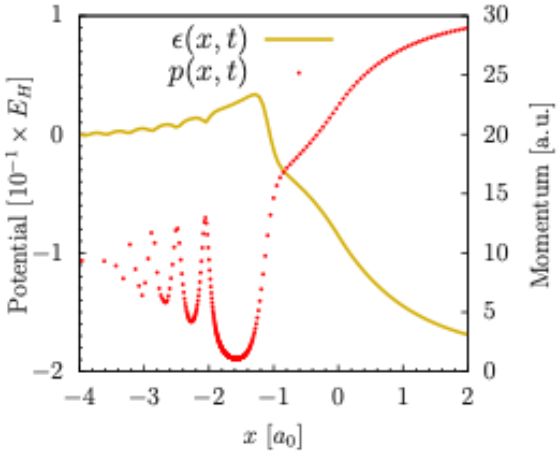

The exact factorization of the electro-nuclear wave function provides an advantageous formalism for the description of non-adiabatic processes[1]. Most notably, its inclusion of the electronic back-reaction on the nuclear subsystem by the means of a single time-dependent potential energy surface (TDPES) together with a vector potential allows to unambiguously define the force driving nuclei in the classical limit. This feat lead to the introduction of a powerful generalization of the Ehrenfest dynamics to the non-adiabatic case: the coupled-trajectories mixed quantum-classical (CT-MQC) method[2,3]. Simulation of nuclear dynamics by the means of classical trajectories offers a much better scaling of the numerical cost with respect to the number of dimensions compared to wave function-based approaches. However, assuming classical behavior for all nuclear degrees of freedom is not always a sensible separation, as it neglects key nuclear quantum effects such as tunneling[4] across classically forbidden regions of the TDPES.
In this talk, I will present the recent efforts undertaken in our team to recover nuclear quantum effects from interacting trajectories[5,6,7] describing rigorously the nuclear motion in the exact factorization ansatz. Dynamics of classical and fully quantum nuclear trajectories will be compared through the study of Tully models[8].

References:

[1] A. Abedi, N. T. Maitra, E. K. U. Gross, Phys. Rev. Lett. (2010)
[2] A. Abedi, , F. Agostini, & E. K. U. Gross, *EPL*, *106*(3), 33001 (2014)
[3] F. Talotta et al., *J. Chem. Theory Comput.* 16, 8, 4833–4848 (2020)
[4] F. Agostini, I. Tavernelli & G. Ciccotti, *Eur. Phys. J. B*, *91*(7), 1-12 (2018)
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[8] J. C. Tully J. Chem. Phys. **93**, 1061 (1990)

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