Nuclear Quantum Effects in Molecular Dynamics: Theory and Applications

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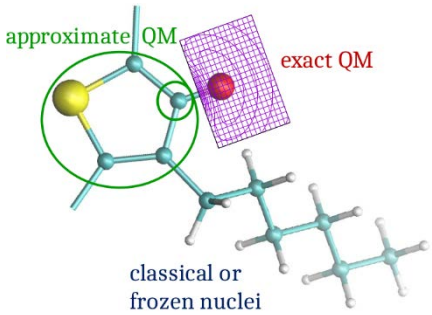
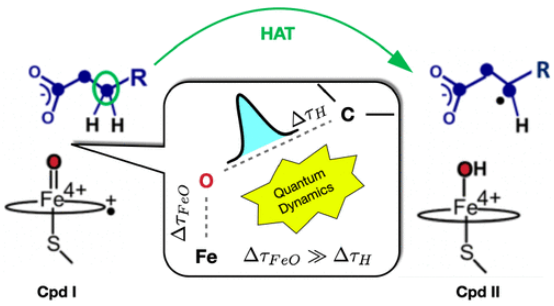
Importance of the quantum-mechanical effects associated with the nuclei is gaining recognition in chemistry and physics, as researchers manipulate matter, light, electric and magnetic fields at the atomistic level for advanced materials applications. For example, the isotope dependence of the proton conductance in low-dimensional boron nitrides, and of the crystallinity of poly(3-hexylthiophene) is attributed, in part, to the nuclear quantum effects. The development of a general dynamics approach, incorporating the nuclear quantum effects and scalable to large molecular systems, remains an outstanding theoretical challenge because of the exponential scaling of computational costs with the system size. We will describe exact [1] and approximate dynamics, inspired by the quantum trajectory formulation of the Schrödinger equation, and optimal moving frames of reference [2]. As an example of the quantum dynamics effects seen in experimental systems, the Kinetic Isotope Effect in Cytochrome P450 Decarboxylase OleT [3], will be discussed.





[1] Matthew Dutra, Sachith Wickramasinghe, and Sophya Garashchuk. Quantum Dynamics with the Quantum Trajectory-Guided Adaptable Gaussian Bases. *Journal of Chemical Theory and Computation* **16**, 18-34 (2020). DOI: 10.1021/acs.jctc.9b00844

[2] Factorized Electron–Nuclear Dynamics with an Effective Complex Potential
Sophya Garashchuk, Julian Stetzler, and Vitaly Rassolov. *Journal of Chemical Theory and Computation* **19** (5), 1393-1408 (2023). DOI: 10.1021/acs.jctc.2c01019

[3] Matthew Dutra, Jose A. Amaya, Shannon McElhenney, Olivia M. Manley, Thomas M. Makris, Vitaly Rassolov, and Sophya Garashchuk. Experimental and Theoretical Examination of the Kinetic Isotope Effect in Cytochrome P450 Decarboxylase OleT. *Journal of Physical Chemistry B* **126** (19), 3493-3504 (2022). DOI: 10.1021/acs.jpcb.1c10280