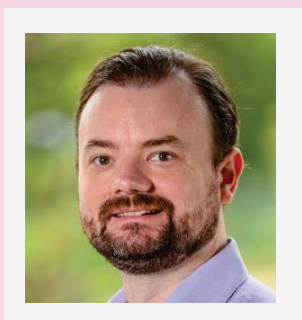


**The Chinese University of Hong Kong  
Department of Chemistry  
Research Seminar Series**

***Pushing boundaries:  
Nonadiabatic dynamics  
simulations of solvent-supported  
electronic states***



Professor William Glover  
Department of Chemistry  
New York University Shanghai

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Contact Person: Prof. Ying-Lung Steve Tse

## Pushing boundaries: Nonadiabatic dynamics simulations of solvent-supported electronic states

William J. Glover<sup>a,b,c</sup>

<sup>a</sup>NYU Shanghai, 1555 Century Avenue, Shanghai, 200122, China

<sup>b</sup>NYU-ECNU Center for Computational Chemistry at NYU Shanghai, Shanghai, 200062, China

<sup>c</sup>Department of Chemistry, New York University, New York, New York, 10003, USA

**Abstract:** A central problem in chemistry is to understand how solvents affect the properties and reactivity of chemical systems. From an electronic perspective, solute-solvent interactions can have a range of effects, from solvatochromic shifts of excitation energies to the emergence of entirely new solvent-supported states. Examples of the latter include charge-transfer-to-solvent states, which lead to the formation of solvated electrons corresponding to an excess electron supported by the solvent but not bound to any single molecule. The aqueous solvated electron  $e^-_{(aq)}$  is of particular interest since low energy electrons induce strand breaks in hydrated DNA. Of relevance is the excited-state dynamics of  $e^-_{(aq)}$  since the ground state is unreactive to DNA. Recent ultrafast time-resolved photoelectron experiments, however, cast doubt on one-electron model simulations which overestimate  $e^-_{(aq)}$ 's excited-state lifetime by an order of magnitude. There is thus considerable room for improvement in the theoretical treatment of solvent-supported states. I will present our recent developments in this area, particularly in using quantum mechanics/molecular mechanics (QM/MM) embedding methods that enable all-electron *ab initio* descriptions of the dynamics of an "active" QM region, with the remainder of the system (solvent) treated with computationally efficient MM forcefields. A challenge arises, however, when applying QM/MM to solvent-supported states, since some of the solvent *must* be included in the QM region. The boundary between QM- and MM-treated solvent molecules should then dynamically adapt over the course of the simulation to avoid their mixing. We have developed a boundary potential approach that maintains QM/MM separation while allowing for density fluctuations, rigorously preserves ensemble averages, and even leaves the dynamics of the QM region unperturbed. Combining our approach with nonadiabatic dynamics simulations, we reconcile theory with experiment and rationalize the ultrafast excited-state lifetime of  $e^-_{(aq)}$  from an electron-transfer perspective. The new technologies thus open the door to studying a wide range of solution-phase chemistry where a QM description of the solvent is necessary.

**Biography:** William Glover is an assistant professor of chemistry at NYU Shanghai and Global Network University assistant professor in the department of Chemistry at NYU. He currently serves as associate director of the NYU-ECNU Center for Computational Chemistry. William received his undergrad degree in Chemistry from the University of Oxford in 2003. He then moved to sunny California where he joined the group of Prof. Ben Schwartz at UCLA and did his PhD research on many-electron mixed quantum/classical descriptions of condensed-phase charge-transfer reactions, graduating in 2009. This was followed by postdoctoral work with Todd Martinez (Stanford) and Ben Schwartz before starting his independent career at NYU Shanghai in 2015. He has established a research program in developing and applying theoretical and computational tools to understand excited-state properties and dynamics of condensed-phase systems, with applications to the photodamage of biological molecules. His work has been supported by the Science and Technology Commission of Shanghai Municipality, the National Natural Science Foundation of China, and the Ministry of Science and Technology National Foreign Experts Program.