Physical Chemistry Seminar

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Real-Time Simulations of Ultrafast Electron Relaxation Dynamics

The Kretchmer group develops simulation methods at the intersection of electronic structure theory and quantum dynamics. We apply our methods to examine electron and spin dynamics, non-adiabatic processes, and charge transport in complex molecular and material systems including weakly-bound molecular clusters, chiral and helical molecules, and perovskites. In this talk, I will highlight our recent development of a new methodology to unravel the competing electronic relaxation pathways following inner-shell ionization in weakly bound systems. When an inner-shell electron is removed, another electron relaxes to fill the vacancy, which can transfer energy to a nearby entity, generating a low-energy secondary ionized electron. Critically, the location of the electron that relaxes *and* the secondary ionized electron can originate from *either* the original molecule or a neighboring molecule. The relaxation can also compete with electron, hole, and even proton transfer. Our work combines real-time DFT with a complex absorbing potential to simulate the explicit dynamics of this complex array of processes. We have additionally introduced coupled electron-nuclear dynamics through Ehrenfest dynamics, which enabled an in-depth investigation of the fragmentation pathways in the water dimer following inner-valence ionization. Our simulations identified a previously unknown relaxation channel essential to reproducing the experimentally observed fragmentation products of our collaborators, prompting a reinterpretation of the origin of low-energy electrons in hydrogen-bonded systems such as water.

PHYSICAL SEMINAR

Joshua Kretchmer's Biography

Joshua Kretchmer was born in San Jose, Costa Rica and grew up in Mill Valley, CA. He received his B.S. in Chemistry from UC Berkeley in 2009 and his Ph.D. as an NSF graduate research fellow from Caltech in December 2014 under the mentorship of Prof. Thomas Miller. He left Caltech for Princeton to join the group of Garnet Chan as a post-doctoral scholar. After the Chan group relocated to Caltech, he returned to finish his postdoc there. He joined the faculty in the School of Chemistry and Biochemistry at the Georgia Institute of Technology in 2019. His research group focuses on the development of methods at the interface of quantum dynamics and electronic structure theory for applications in electron dynamics in complex systems. His group is currently supported by single PI grants from the DOE and ACS PRF as well as collaborative grants from the DOE and NSF.