Physical Seminar

Monday, February 15, 4 pm

Virtual Event

Alexey Akimov

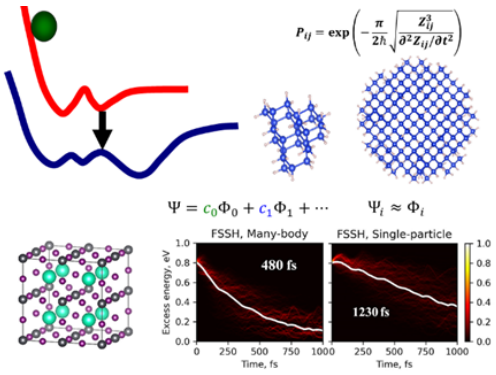
Assistant Professor of Chemistry

SUNY Buffalo

"Modeling Nonadiabatic Dynamics in Nanoscale Solar Energy Materials"

Abstract: The dynamics of excited states is critically important in many materials such as photovoltaic solar energy harvesters, photocatalysts, or photo-actuated molecular switches. Electron-hole recombination, interfacial charge transfer, or nonradiative excitation energy relaxation, all require accounting for nonadiabatic (NA) effects. Computational modeling of NA and quantum dynamics (NA/QD) at the atomistic level can provide valuable insights into these processes and help design new routes for improving materials' efficiencies, but also poses a great challenge to the scientific community due to its taxing computational complexity. The demands for modeling NA/QD in periodic solids and nanoclusters stimulated the development of numerous approximations and the wide adoption of simplified computational schemes. However, the quality of such schemes remains largely unclear, leaving one questioning about their applicability. It is often unclear how one scheme compares with another in a zoo of many ad hoc approaches.

With the idea of facilitating the assessment of various NA/QD schemes, my group developed a modular open-source Libra software and utilized it as a framework for developing new practical tools for NA/QD, assessing a variety of existing approximations, and for applied studies of NA dynamics in various types of nanoscale solar energy materials. In this presentation, I will discuss some of our recent developments for the approximate modeling of NA/QD in nanoscale and condensed-matter systems. First, I will demonstrate how the Landau-Zener-inspired trajectory surface hopping approach can aid in modeling hot carrier relaxation dynamics in Si nanocrystals/quantum dots and help reveal qualitative trends on relaxation rates as a function of the nanoparticles' size and surface termination type. I will then proceed to our recently developed schemes that can account for some many-body effects in modeling NA/QD in nanocrystals and periodic solids. In view of these developments, I will discuss the role of many-body treatment of excited states in Si and CdSe nanoclusters and lead halide perovskites. The qualitative trends in computed NA/QD as the function of the density functional choice and inclusion of relativistic effects will be discussed. Time-allowing, I will present our assessment of several popular decoherence-corrected surface hopping schemes and their ability to yield thermal equilibrium in extended systems, where electron-nuclear back-reaction needs to be neglected.



Zoom Meeting: https://rochester.zoom.us/j/96353495610

Website: https://events.rochester.edu/event/chemistry_physical_seminar_akimov

Host: Prof. Frank Huo • Email: huo@chem.rochester.edu