Decoding Photophysics in Optoelectronic Materials via Coupled Nuclear-Electron Dynamics

Wei-Tao Peng

Department of Chemistry, Tunghai University

Abstract

The design and optimization of state-of-the-art optoelectronic materials often rely on static energy alignments and/or structure-property relationships. However, static pictures are fundamentally insufficient to capture the complex, non-adiabatic processes that dictate device performance. To truly understand phenomena such as molecular responses under external field, exciton dissociation, and charge separation, a rigorous treatment of coupled nuclear-electron dynamics is required.

In this talk, I will present our recent advancements in theoretical and computational methodologies designed to tackle these dynamic challenges. First, I will introduce our GPU-accelerated real-time Time-Dependent Complete Active Space Configuration Interaction (TD-CASCI) method, an efficient tool for simulating molecular responses under varying laser shaped pulses. Second, I will discuss the eXcitonic state Surface Hopping (X-SH) method, which enables fully atomistic simulations of exciton and charge transport in organic donor-acceptor blends at realistic length scales (> 10 nm). Our dynamic simulations reveal that the transport mechanism is governed by transient delocalization, fundamentally different than traditional hopping and band transport. Moving beyond static pictures, these dynamic approaches offer a predictive theoretical foundation for future material design.