Mesoscale quantum dynamics in molecular aggregates

The principles governing charge and energy transport in novel molecular semiconductors are often obscured by the complex interplay of structural heterogeneities, interfaces, and disorder on the nanometer-to-micron length scale (i.e., the mesoscale). To engineer novel molecular materials that enable next-generation technologies, we need a predictive understanding of how excited-state carriers – such as excitons, electrons, and holes – move through heterogeneous mesoscale systems. However, most computational methods remain intractable for mesoscale molecular materials where the number of molecules is massive, and the material parameters tend to fall into the broad "intermediate regime" where perturbative techniques breakdown. In this talk, I will discuss recent developments in my group that have enabled the first formally-exact mesoscale simulation of exciton dynamics and spectroscopy in molecular materials. These developments offer a glimpse of a new class of quantum dynamics methods that can illuminate photophysical mechanisms of realistic molecular materials.