Excited State Dynamics in Hybrid Nanoscale Materials: Time-Domain Ab Initio Studies

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Excited state dynamics play key roles in numerous novel molecular and nanoscale materials designed for photovoltaics, photocatalysis, electronics, spintronics and many other applications. Controlling these far-from-equilibrium processes and steering them in desired directions require understanding of material's dynamical response on the nanometer scale and with fine time resolution. We couple real-time time-dependent density functional theory for the evolution of electrons with non-adiabatic molecular dynamics for atomic motions to model such non-equilibrium response in the time-domain and at the atomistic level. The talk will describe the basics of the simulation methodology and will discuss several exciting applications among the broad variety of systems and processes studied in our group, including semiconducting and metallic quantum dots, hybrid organic/inorganic perovskites, transition metal dichalcogenides, metallic and semiconducting films, graphene, carbon nanotubes, molecular crystals and assemblies used in singlet fission, organic polymers, etc. Photo-induced charge and energy transfer, Auger-type processes, energy losses and charge recombination create many challenges due to large differences between molecular and periodic, and organic and inorganic matter. Our simulations provide a unifying description of quantum dynamics on the nanoscale, characterize the timescales and branching ratios of competing processes, resolve debated issues, and generate theoretical guidelines for development of novel systems.