Dynamics of excited states plays a determining role in many materials such as solar cells, photocatalysts, or photo-actuated molecular switches. Electron-hole recombination, interfacial charge transfer, nonradiative excitation energy relaxation, photoinduced nuclear reorganization, and bond breaking - all require accounting for nonadiabatic (NA) effects. Nuclear tunneling and zero-point energies constitute quantum nuclear effects that may non-trivially affect the electron-nuclear dynamics. Understanding details of nonadiabatic and quantum dynamics (NA/QD) at the atomistic level requires advanced tools and methodologies. Often, one also needs to address the computational limitations due to systems’ sizes and processes’ time scales, what makes modeling NA/QD in realistic materials a challenging problem. Over the past few years, my group has been addressing various methodological problems in NA/QD to enable computationally affordable yet reasonably accurate modeling of excited states dynamics in extended atomistic systems. In this presentation, I will overview several approaches we have developed in this regard and will illustrate their utility in modeling charge transfer dynamics in organic and inorganic solar-harvesting systems. The developments in three key areas will be highlighted: a) modeling NA/QD in extended systems via simplified semiclassical and fragmentation approaches as well as via innovative software that leverage linear-scaling electronic structure methods; b) modeling slow NA/QD via the machine-learning-inspired quasi-stochastic Hamiltonian approach and via the enhanced sampling scheme; c) accounting for quantum nuclear effects via the entangled trajectories Hamiltonian dynamics (ETHD) method. Along the way, I will touch on other important methodological questions such as the role of phase correction, decoherence, and the effect of electronic structure choice. Last but not least, I will highlight chemical insights we have gained from the NA/QD modeling in all-inorganic perovskites, quantum dots, and materials’ heterojunctions.