In order to develop a working chemical intuition about organic electronic materials, and organic photovoltaic materials, in particular, it appears to be imperative to understand the relationship between molecular-level structure and the excited state relaxation phenomena of exciton migration and charge separation within conjugated polymers and at organic donor/acceptor interfaces. In this talk, I will describe recent progress in using a mixed quantum/classical non-adiabatic molecular dynamics simulation approach that employs an all-atom description of the intermolecular interactions coupled to a semi-empirical (PPP) electronic Hamiltonian. Results exploring several systems at ambient temperature will be discussed. The systems will include phenylene-vinylene and thiophene oligomers, as well as cyanine and fullerene components. The roles of molecular structural fluctuations and intermolecular electronic couplings, as well as the role of external fields, will be explored.