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Exciton transport and dissociation at organic interfaces

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This paper focuses on modeling studies of exciton transport and dissociation at organic interfaces and includes three parts: 1) Experiments have shown that the values of exciton diffusion length L_D in conjugated polymers (CPs) are rather low, in the range of 5-10 nm, apparently regardless of their chemical structure and solid-state packing. In contrast, larger L_D values have been reported in molecular materials that are chemically more well-defined than CPs. Here we demonstrate that energetic disorder alone reduces the exciton diffusion length more than one order of magnitude, from values typically encountered in molecules (>50nm) to values actually measured in CPs (<10nm). 2) A number of organic crystals show anisotropic excitonic couplings, with weak interlayer interactions between molecules that are more strongly coupled within the layers. The resulting energy carriers are intra-layer 2D excitons that diffuse along the interlayer direction. We model this analytically for infinite layers and using quantum-chemical calculations of the electronic couplings for anthracene clusters. We show that the exciton hopping rates and diffusion lengths depend in a subtle manner on the size and shape of the interacting aggregates, temperature and the presence of energetic disorder. 3) The electronic structure at organic/organic interfaces plays a key role, among others, in defining the quantum efficiency of organic-based photovoltaic cells. Here, we perform quantum-chemical and microelectrostatic calculations on molecular aggregates of various sizes and shapes to characterize the interfacial dipole moment at pentacene/C60 heterojunctions. The results show that the interfacial dipole mostly originates in polarization effects due to the asymmetry in the multipolar expansion of the electronic density distribution between the interacting molecules. We will discuss how the quadrupoles on the pentacene molecules produce direct electrostatic interactions with charge carriers and how these interactions in turn affect the energy landscape around the interface and therefore also the energy barrier for exciton dissociation into free carriers.