

Self-assembling Organic Nanostructures for Solar Energy Conversion

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Self-assembly of small electron donor-acceptor (D-A) molecules into discrete and monodisperse nanostructures provides geometrically defined platforms to emulate the photo-induced electron transfer processes in photosynthesis. Organization of molecules by this thermodynamically driven method can result in architectures with unique inter-chromophore relationships that are otherwise difficult to realize by conventional covalent synthesis. In particular, π -stacked D-A dyads and triads can afford ordered and segregated D/A domains through which photo-generated holes and electrons can be further separated and rapidly transported to electrodes in photovoltaics or to catalysts for solar fuels formation. We have developed assembly strategies based on controlled π -stacking and bio-inspired guanine quadruplexes, which demonstrate that photo-induced charge separation can take place within covalent chromophoric redox partners followed by transport of photo-generated holes and electrons independently through well-ordered, segregated molecular charge conduits (Figure 1).

We are also developing robust organic chromophores that undergo singlet fission (SF), the spontaneous down-conversion of a singlet exciton to two triplet excitons (Figure 2), using guidance from electronic structure calculations to assure the requisite relationships between molecular singlet and triplet exciton energies. We are preparing hierarchical assemblies of these chromophores, starting from covalent dimers, then developing supramolecular assemblies, and engineered crystalline materials to investigate SF in bulk, ordered organic materials. We are using time-resolved optical and EPR spectroscopy to characterize the SF mechanism and the factors that determine its efficiency. We will present results on the role of charge transfer and quintet states in enabling SF in organic materials.

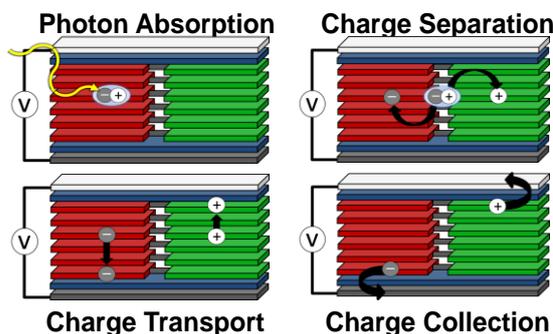


Figure 1. Charge conduit concept.

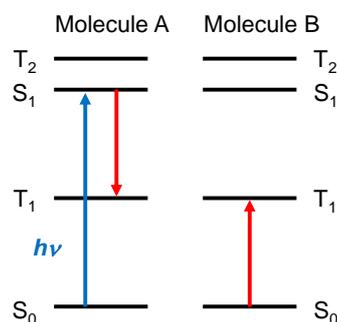


Figure 2. Energy level diagram for SF.