Multiple Exciton Generation via Intermolecular Singlet Fission within Aqueous Nanoparticles

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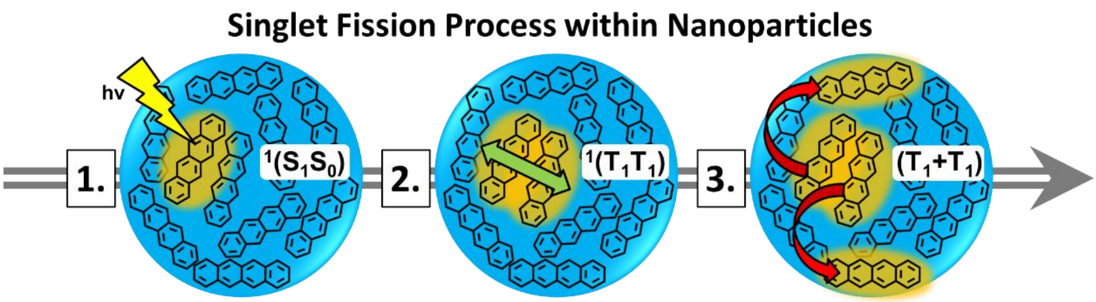
The Shockley-Queisser limit describes the maximum efficiency of solar cells, by accounting for the various intrinsic loss processes and is currently placed at roughly 33% for single junction solar cells. Losses through thermalization, which is the inability to absorb high energy photons and dissipating the excess energy via heat, have the largest impact on the reduced efficiency of solar cells.[1] The majority of the visible solar spectrum is not utilizable, since most of these unusable high energy photons are located within it. Energetically down-converting these photons would unlock this part of the solar spectrum, reduce thermalization losses and subsequently increase the Shockley-Queisser limit.

Singlet fission is such a photophysical process, which through the interaction of chromophores down-converts high energy singlet excited states into two individual triplet excited states of lower energy. The typically spin-forbidden transition from the singlet to the two triplet excited states is mediated by a correlated triplet pair of singlet multiplicity, resulting in an ultrafast and spin-allowed process and generating multiple excitons from the absorption of one photon.[2]

In order for singlet fission to occur, sufficient electronic coupling between interacting chromophores needs to be provided. In this study, we opted for an intermolecular approach via the self-assembly of aqueous nanoparticles to ensure sufficient coupling. Nanoparticles of dicarboxylic acid tetracene together with chiral and achiral amines were prepared and characterized by means of steady-state and time-resolved spectroscopic as well as microscopic techniques.



1) A. Rao, R. H. Friend, *Nat. Rev. Mater*. **2017**, *2*, 17063. 2) R. Casillas, I. Papadopoulos, T. Ullrich, D. Thiel, A. Kunzmann, D. M. Guldi, *Energy Environ. Sci.* **2020***, 13*, 2741.

