Abstract

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Abstract

Singlet fission (SF) is a photophysical phenomenon described more than half a century ago in which a singlet exciton shares its energy with a ground state molecule forming two triplet excitons. The pursuit of SF chromophores has been resurrected by the community only recently since it might lead to an efficient exciton downconversion. Its harnessing will lead to a jump in the efficiency of solar cells breaking the Shockley-Queisser limit.

The process implies that the interaction of at least two molecules is needed. Therefore, our goal was to deliver a simple yet robust method to obtain the rate of SF within a molecular dimer. This method also provides an insight into improving the known structures via mutual disposition optimization in non-prohibitive amounts of time. The whole theory was deployed by our lab using an in-house Fortran program package called *Simple*. It successfully passed tests against state-of-the-art theoretical methods such as *Non-Orthogonal Configuration Interaction* and *Ab Initio Frenkel-Davydov Exciton Model*. It also correctly predicts the trend in real-life experiments on some organic dyes of practical interest.

Other experiments have shown that the interaction cannot be limited only to two molecules, which is in stark contrast to the usual approach. An example of this is the two crystal polymorphs of diphenylisobenzofuran, only one of which efficiently undergoes SF while the other succumbs to fast charge separation. The "dimer" theories provide similar results for both polymorphs breaking the qualitative agreement. Due to the coupling of excited and charge transfer states on other neighbors, the inclusion of more molecules is thus imperative. We present the future steps and hurdles to achieve a more general description of SF built on the framework of *Simple*.