

Triplet Exciton Harvesting Through Singlet Fission

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Singlet fission (SF) is a multiple-exciton generation process which converts one singlet exciton into two triplet excitons and has received considerable attention because of its potential to overcome the Shockley-Queisser limit on the power conversion efficiency (PCE) of solar cells. Recently, intramolecular SF in covalent dimers and oligomers are widely studied because of their well-controlled morphology and electronic coupling. The triplet pair (TT) state of the covalent systems in solution does not dissociate readily, allowing for more thorough studies of the multiexciton state, but is a disadvantage for the triplet harvesting. The covalent system should also undergo efficient SF process in film. However, the SF dynamics will be complicated as both intramolecular and intermolecular SF can occur. Here, we demonstrate a molecular engineering approach to independently optimize the triplet generation and decay process in intramolecular SF films by using terphenyl-bridged TIPS-pentacene tetramers. By controlling the degree of steric hindrance within individual tetramers, we can systematically tune the degree of intermolecular coupling in thin films. Taking advantage of both the intra- and intermolecular SF processes allows us to maintain a rapid triplet pair generation process, even in the case of weak intermolecular coupling. This approach allows us to maintain a picosecond triplet generation process while simultaneously varying the rate constants for triplet-triplet annihilation over three orders of magnitude.