Singlet fission and triplet-triplet annihilation: symmetry and dimeric designs.

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Singlet fission (SF) is a process where a singlet exciton is split into a pair of triplet excitons. The increase in the excitonic generation can be exploited to enhance the efficiency of solar cells. Molecules with conjugated π bonds are commonly developed for optoelectronic applications including SF, due to their low energy gaps. The electronic coupling for SF in such well-stacked π-conjugated molecule pairs can be rather limited due to the orthogonal π and π* orbital overlaps that are involved in the coupling elements, leading to a large cancellation in the coupling. In the present work, we show that such limits can be removed by involving triplet states of different origins, such as those with nonbonding n orbitals. We demonstrate such an effect for formaldehyde and methylenimine dimers, with a low-lying n–π* triplet state (T_1) in addition to the π–π* triplet (T_2). We show that the coupling can be enhanced by 40 times or more for the formaldehyde dimer, and 15 times or more for the methylenimine dimer, with the T_1–T_2 state as the end product of SF. Our results provide a new molecular design concept for better SF (and triplet–triplet annihilation, TTA) materials that allows future development. The commonly seen influence of charge-transfer component in SF or TTA will also be reported in the presentation.