Singlet Fission and Magnetic Fields

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Singlet fission is a molecular process wherein a photoexcited singlet (S_1) splits into two triplets with (anti-) correlated spins. Since two excitons are generated from a single absorption event it has been proposed that singlet fission can be used to augment the current of solar cell technologies.[1] Indeed, if fully exploited singlet fission raises the detailed balance limiting energy conversion efficiency of solar cells from 33.7% to 45.9%. However, several obstacles must be overcome before this is achieved. Upon fission, the singlet state produces a triplet-triplet pair state which much subsequently decouple to form 'free triplets' which are useful for current generation

$$S_1 \rightarrow (T_1T_1) \rightarrow T_1 + T_1$$

In many systems the intermediate (T_1T_1) state is stabilized, and an energetic barrier must be overcome in order for fission to occur. Using electron spin resonance (ESR) spectroscopy, we have recently also shown that, while the initially prepared (T_1T_1) state must have overall singlet character, a quintet state, $^5(T_1T_1)$, is also observed some nanoseconds after photoexcitation.[2] This alters the canonical description of the occupation of states in the triplet-triplet pair manifold that was put forth at the outset of singlet fission research.[3]

In this presentation the properties intermediate state in a number of different inter- and intramolecular singlet fission systems will be discussed. Using a range of ultrafast optical spectroscopy and magnetic resonance spectroscopies, the nature of the intermediate state will be elucidated.

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References

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