

What Drives Intramolecular Singlet Fission?

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Singlet fission (SF) is a promising process which allows two triplet-exciton generation from a singlet exciton through spin-related processes. The mechanisms driving this process can be demonstrated by using external magnetic-dependent spectroscopic techniques including photoluminescence spectroscopy. As proposed by *Merrifield*^[1], the magnetic effects shown in the fluorescence of crystalline tetracene suggest the evolution of spin wavefunction of optical excited excitons from a singlet state to triplet states through a triplet pair state. By expanding the study of *Merrifield*, the dynamics of SF in crystalline tetraene under an external magnetic field has been reported by *Burdett et al.*^[2] Their study shows the presence of quantum beats in the delayed fluorescence under magnetic field confirming the existence of the triplet pair state.

Recently, some of acene derivatives have been reported as efficient intramolecular singlet fission (iSF) materials enabling SF to occur in an individual molecule, independent from their packing configuration.^[3] In this study, novel polyacene derivatives have been investigated using several magnetic-dependent spectroscopic techniques. We probe the fluorescence yield from an excited singlet state (S_1) to a ground state (S_0), which evolves with respect to an external magnetic field strengths. Moreover, the dynamics of iSF in these materials are also investigated in the same manner.

This study provides a further understanding of the iSF process in organic molecules with potential applications in optoelectronic devices. All the dynamics demonstrated suggest a suitable material system for iSF to occur. Moreover, the magnetic-dependent spectroscopy used in this study has been shown to be a powerful tool in probing molecular systems and provides an extra dimension to optical techniques.

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