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Singlet fission in pentacene through multiple exciton quantum states ZHIYONG ZHANG, PAUL ZIMMERMAN, Stanford University, CHARLES MUSGRAVE, University of Colorado — Multi-exciton generation (MEG) has been reported for several materials and may dramatically increase solar cell efficiency. Singlet fission is the molecular analogue of MEG and has been observed in various systems, including tetracene and pentacene, however, no fundamental mechanism for singlet fission has yet been described, although it may govern MEG processes in a variety of materials. Because photoexcited states have single-exciton character, singlet fission to produce a pair of triplet excitons must involve an intermediate state that: (1) exhibits multi-exciton (ME) character, (2) is accessible from S1 and satisfies the fission energy requirement, and (3) efficiently dissociates into multiple electron-hole pairs. Here, we use sophisticated *ab initio* calculations to show that singlet fission in pentacene proceeds through a dark state (D) of ME character that lies just below S1, satisfies the fission energy requirement ($E_D > 2E_{T0}$), and splits into two triplets (2×T0). In tetracene, D lies just above S1, consistent with the observation that singlet fission is thermally activated in tetracene. Rational design of photovoltaic systems that exploit singlet fission will require *ab initio* analysis of ME states such as D.

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