

Abstract Submitted
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Theory of excited state absorptions in pentacene crystals and films¹ SUMITENDRA MAZUMDAR, SOURATOSH KHAN, University of Arizona
— There exists a disconnect between the experimental technique to detect singlet fission in organic molecular systems and current theoretical approaches to this process. Experimentally, singlet fission is detected by time-resolved ultrafast spectroscopy; existing theoretical approaches until now have not allowed calculations of excited state absorptions, and comparisons between experiment and theory are indirect. In this work we present a complete theory of ultrafast excited state absorptions in pentacene herringbone crystals and films. We first calculate ground state absorptions within the Parier-Parr-Pople Hamiltonian and show that our calculations give very good fit to the wavelength dependent absorption spectrum. We then calculate singlet and triplet photoinduced absorptions, and find that, (a) photoinduced molecular rotations leading to creation of nearest neighbor dimers is an essential first step to singlet fission, (b) dimer formation leads to both an excimer and a triplet-triplet state, and (c) while the earliest excited state absorption from the product triplets of singlet fission are intramolecular excitations, later triplet photoinduced absorptions are to an intermolecular charge-transfer state.

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