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Exciton fission and solar energy conversion beyond the limit

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The absorption of one photon by a semiconductor material usually creates one electron-hole pair, but this general rule breaks down in a few organic semiconductors, such as pentacene and tetracene, where one photon absorption may result in two electron-hole pairs in a process called singlet exciton. Recent measurements in our group by time-resolved two-photon photoemission (TR-2PPE) spectroscopy in crystalline pentacene and tetracene provided the first spectroscopic signatures in singlet fission of a critical intermediate known as the multiexciton state. More importantly, population of the multiexciton state is found to rise concurrently with that of the singlet state on the ultrafast time scale upon photo excitation. This observation provides an experimental foundation for a quantum coherent mechanism in which the electronic coupling creates a quantum superposition of the singlet and the multiexciton state immediately following optical excitation. We demonstrate the feasibility of harvesting the multiexciton state for multiple charge carriers and the implementation of singlet fission for solar energy conversion beyond the Shockley-Queisser limit.