Exciton Fission, Quantum Coherence, and Solar Energy Conversion Beyond the Limit

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The maximum solar-to-electric power conversion efficiency of a conventional solar cell is determined by the Shockley-Queisser limit of ~31%. One viable approach to exceed this limit is to create two or more electron-hole pairs from the absorption of one photon in a process called signlet fission (SF) or multiple exciton generation. SF has attracted renewed interest because of the great potential of designing molecules for optimal fission yields. However, realizing this potential is difficult because we do not yet know how to obtain high fission yields or to ensure the most efficient extraction of multiple carriers. Here, we show in organic semiconductors direct evidence for a key intermediate known previously only in theory for singlet fission as the multiexciton (ME) state. Although singlet fission can be endoergic, this process is not thermally activated. Instead, a coherent quantum process is responsible for overcoming the energetic barrier on an ultrafast time scale. Surprisingly, we find that multiple electron transfer from the multiexciton state in the organic semiconductor to the C_{60} electron acceptor occurs on a sub-picosecond timescale, one order of magnitude faster than that from the triplet exciton state. Direct charge transfer from the ME state represents the most efficient means of harvesting multiple excitons and may also increase the exciton fission yield in return. These discoveries take us one step closer to designing molecular solar cells with power conversion efficiency potentially exceeding the Shockley-Queisser limit.

W.-L. Chan, M. Ligges, A. Jailaubekov, L. Kaake, L. Miaja-Avila, X.-Y. Zhu, Science 334 (2011) 1541-1545.