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Direct transfer of triplet excitons generated by singlet fission to colloidal quantum dots: A route to sensitize silicon solar cells

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Singlet exciton fission is the process in organic semiconductors by which one Coulombically-bound electron-hole pair (exciton) with net singlet spin evolves into a pair of spin-triplet excitons. Although the phenomenon has long been a curiosity in organic crystals[1], we recently demonstrated that singlet fission proceeds rapidly (80fs-400ps)[2-4] and efficiently (approaching quantitative ('200%') yield)[4-5] in thin films made from a range of organic molecules that satisfy the energetic criteria: $E_{sing} \geq 2 \cdot E_{trip}$[4].

Critically, singlet fission is now attracting wider interest[6] because it could boost the power conversion efficiency of photovoltaic devices. Specifically, as first envisioned by Dexter[7], a blue-absorbing singlet fission material could sensitize a red-absorbing solar cell, enabling the generation of additional photocurrent from high-energy photons. However, though we have recently reported fission-based photovoltaic devices where the peak photon-to-electron conversion efficiency is greater than unity [8-9], their overall power conversion performance remains unremarkable, as they rely on immature solar technologies (e.g. organic semiconductors) for the red-absorbing cell.

Here, by detecting a characteristic magnetic field dependence using steady-state and transient spectroscopies, we demonstrate that fission-generated triplet excitons readily undergo exchange-mediated (Dexter) transfer to PbS colloidal quantum dots. Because the spin-states of the quantum dots are mixed at room temperature, this renders the fission-generated excitons emissive. Thus, we consider that longer-range Förster energy transfer may permit the direct sensitization of a conventional back-contacted silicon solar cell with an organic singlet fission material.

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