External quantum efficiency exceeding 100% in a singlet-exciton-fission-based solar cell

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Singlet exciton fission can be used to split a molecular excited state in two. In solar cells, it promises to double the photocurrent from high energy photons, thereby breaking the single junction efficiency limit. We demonstrate organic solar cells that exploit singlet exciton fission in pentacene to generate more than one electron per incident photon in the visible spectrum. Using a fullerene acceptor, a poly(3-hexylthiophene) exciton confinement layer, and a conventional optical trapping scheme, the peak external quantum efficiency is $(109\pm1)\%$ at $\lambda = 670$ nm for a 15-nm-thick pentacene film. The corresponding internal quantum efficiency is $(160\pm10)\%$. Independent confirmation of the high internal efficiency is obtained by analysis of the magnetic field effect on photocurrent, which determines that the triplet yield approaches 200\% for pentacene films thicker than 5 nm. To our knowledge, this is the first solar cell to generate quantum efficiencies above 100\% in the visible spectrum. Alternative multiple exciton generation approaches have been demonstrated previously in the ultraviolet, where there is relatively little sunlight. Singlet exciton fission differs from these other mechanisms because spin conservation disallows the usual dominant loss process: a thermal relaxation of the high-energy exciton into a single low-energy exciton. Consequently, pentacene is efficient in the visible spectrum at $\lambda = 670$ nm because only the collapse of the singlet exciton into two triplets is spin-allowed.

$^1$Supported as part of the Center for Excitonics, an Energy Frontier Research Center funded by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences under Award Number DE-SC0001088.