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Title

Estimating the quantum yield of singlet fission with respect to competing processes

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Abstract

There is a great effort in improving the efficiency of solar cells above the Shockley-Queisser limit by carrier multiplication, which can be achieved for example via the singlet fission process. One singlet exciton is converted into two triplet excitons through this mechanism. This process has been recently intensively studied both theoretically and experimentally.

Our aim is to develop an efficient way to estimate the rate constants of competing processes, especially exciton formation or charge transfer state formation in a crystal structure. This knowledge is vital for the calculation of the quantum yield of the singlet fission process, which depends on them. Unfortunately, the dimer-only method already employed to obtain the rates of singlet fission cannot capture the crystal environment needed to represent these systems.

Because of that, we are developing a fresh new approach combining the fluctuation-dissipation theorem with Marcus theory to calculate the rate constants of the charge transfer state. We can extract all the necessary information from the equilibrium time correlation function of energy gaps between the locally excited singlet state and the charge transfer state.

Perylene is chosen as a model molecule for the purposes of development as it can form two different polymorphs which favor competing photoproducts. Excimer formation is the dominant excitation process for the α-modification of perylene, while the singlet fission process is dominant for the β-modification of this crystal. According to our current results, we are able to capture correctly this major difference. On top of that, employing the slow modulation limit reduces the calculation we need to perform to only a simple static calculation.