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Modeling of Singlet Fission Kinetics for a Wide Range of Molecules

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Singlet exciton fission is a process that occurs in organic molecules where one high energy singlet exciton decays into two low energy triplet excitons. Over the years since its first discovery in the 1960s a number of different singlet fission materials have been discovered with a wide range of rates and yields. The mechanism for singlet fission in these materials is still not fully understood, and no method is able to accurately reproduce fission rates over a wide range of timescales. In order to gain a better understanding of the singlet fission mechanism a group of fission materials with vastly different crystal structures and fission rates were modeled. Using a first principles expression the rates were computed with constrained density functional theory with configuration interactions. The computed rates successfully predict the fission rates in the different materials studied. For the slow, weak intermolecular coupling materials singlet fission obeys Marcus theory, but for faster, larger intermolecular coupling materials the rate becomes diabatic in nature. This work alters the guidelines for tailoring molecular properties from a focus on crystal packing and intermolecular coupling to properties like solubility and energy level alignment while maintaining the high fission yield required for photovoltaic applications.