Using single-excitation wavefunctions to compute exciton-binding energies in singlet fission materials

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Due to the possibility of improving in photovoltaic efficiencies, significant effort has been directed towards understanding the singlet fission mechanism. Although accurate quantum chemical calculations would provide a detail-rich view of the singlet fission mechanism, this is complicated by the multiexcitonic nature of one of the key intermediates, the $^1(TT)$ state. Being described as two simultaneous and singlet-coupled triplet excitations on a pair of nearest neighbor monomers, the $^1(TT)$ state is inherently a multielectronic excitation. This fact renders most single-reference quantum chemistry methods incapable of providing accurate results.

In this talk I will discuss our recently developed strategy in which single-spin flip calculations are mapped onto a spin-only Hamiltonian, which is then solved by exact diagonalization to yield the target low-energy biexciton states. Numerical examples are included for a number of different systems, including dimers, trimers, tetramers, and a cluster comprised of seven chromophores.