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Accessing the “Dark” Triplet-Pair State in Singlet Fission Molecules

Singlet fission is a promising strategy to overcome fundamental efficiency limits in optoelectronic and photovoltaic devices. This process is governed by an intermediate correlated triplet-pair state, $^1(TT)$. Despite its central role, direct utilization of the triplet-pair state remains challenging because $^1(TT)$ is optically dark, and its fundamental properties are still poorly understood.

Here, I will present a strategy to directly extract and brighten triplet-pair states by interfacing singlet-fission molecules with monolayer transition-metal dichalcogenides (TMDs). First, we demonstrate direct charge transfer from the $^1(TT)$ state into 2D materials. Unlike singlet exciton transfer, this process is mediated by tunneling, revealing a distinct interfacial transfer mechanism unique to this correlated $^1(TT)$.

We further show that the optically dark $^1(TT)$ state can be effectively brightened through coupling to monolayer WS_2 , dominated by Förster energy transfer. This energy transfer is both ultrafast (≈ 1 ps) and long-range (up to ~ 100 nm). The observed long-range behavior is well described within a quantum electrodynamics (QED) framework. Importantly, brightening of the $^1(TT)$ state enables direct optical access to its underlying spin sublevels.

Together, these results establish a general framework for manipulating correlated multiexciton states at organic–2D interfaces and highlight the potential of hybrid heterostructures for harvesting and controlling dark excitonic species in next-generation optoelectronic and quantum materials.