Conformation Change of Exciton Pair: Spin-Entanglement Transport during Singlet Fission Studies by Time-Resolved EPR

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Singlet fission (SF) is expected to exceed the Shockley–Queisser theoretical limit of efficiency of organic solar cells. Spin-entanglement in the triplet pair state via one singlet exciton is a promising phenomenon for several energy conversion applications including quantum information science. However, direct observation of the electron spin polarization by transports of entangled spin-states has not been demonstrated. Furthermore, vibronic mechanisms of the dissociation of the triplet excitons are poorly understood. In this study, time-resolved electron paramagnetic resonance has been utilized to observe the transportations of the singlet and quintet characters generating spin-correlated correlated triplet pair (SCTP) by probing the electron spin polarization (ESP) generated in thin films of 6,13-bis(triisopropylsilylethynyl)pentacene.[1] We have clearly demonstrated that the ESP detected in resonance field positions of the individual triplet excitons are dependent on morphology and on detection delay time after laser flash to cause SF. The ESPs were clearly explained by quantum superposition[1,2,3] of singlet-triplet-quintet wavefunctions via picosecond triplet-exciton dissociation as the electron spin polarization transfer from strongly exchange-coupled singlet TT states to the weakly-coupled SCTP via spin-spin dipolar couplings with preserving conformations of the excitons. Although the coherent superposition of the spin eigenstates was not directly detected, the present interpretation of the spin correlation of the separated T+T exciton pair may pave new avenues not only for elucidating the vibronic role on the de-coupling[2,3,4] between the two excitons but also for scalable quantum information processing using quick T+T dissociation via one-photon excitation.

References

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