

Escaping Excimers in Multichromophoric Aggregates: Role of Complex Assembly Pathways

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One of the greatest appeals of supramolecular assemblies lies with the possibility of realizing newer functionalities and emergent phenomena that are not accessible to its constituent molecules. However, self-assembly can also lead to phenomena that are antagonistic to each other. A case in point is that of aggregates with cofacially aligned dye molecules, the H-aggregates. Symmetry-forbidden, long-lived exciton-coupled states of H-aggregates hold great promise for efficient transport of excitation energy over large distances, but are often scavenged by structurally relaxed excimers that are ubiquitous in these aggregates. The focus of this talk will be on a flexible, covalently-bridged perylene bisimide (PBI) dimer that forms strongly exciton-coupled H-aggregates, in which the dissipative excimer state is fully suppressed. Using time dependent optical absorption and photoluminescence (PL) experiments, we studied the evolution of exciton-coupling and excimer formation through different stages of dimer self-assembly. Mechanistic studies using temperature dependent optical absorption and PL reveal two distinct aggregation pathways. How the competition between these aggregation pathways is influenced by the conformational flexibility of the folda-dimer will also be discussed.

References:

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