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Cooperative FRET: Comparison between Interacting and Independent FRET Pathways in a Light Harvesting Network PAUL CUNNINGHAM, CHRISTOPHER SPILLMANN, SUSAN BUCKHOUT-WHITE, JOSEPH MELINGER, MARIO ANCONA, ELLEN GOLDMAN, IGOR MEDINTZ, US Naval Research Laboratory — Light harvesting antennae use Förster Resonance Energy Transfer (FRET) between dyes to funnel energy to a reaction center. Inhomogenities in dipole orientation in distributions of static dyes reduce FRET efficiencies, via the large multiplicity of unfavorable orientations. Multiple FRET pathways may compensate for inhomogeneities. We examine multi-step FRET in a series of dye-labeled DNA rail scaffolds where the degree of interaction between two parallel rails was adjusted via their separation. We find that interacting pathways outperform simply redundant independent pathways. For one FRET step, addition of a second donor yields an unexpected increase in FRET efficiency. Monte-Carlo simulations show that suppression of inefficient FRET pathways causes this increase. As the number of donors increases, the FRET efficiency of a static distribution approaches the dynamic limit, where dyes are free to reorient. This suppression is optimal when the rails are close enough to allow fast homo-FRET between them. However, at close separations H-like aggregate formation can lead to energy sinks. These are important considerations when designing light harvesting networks and may aid in the understanding of incoherent hopping transport in other systems.

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