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Molecular exciton formation and energy transfer in a network of closely spaced dyes on a DNA scaffold PAUL CUNNINGHAM, SUSAN BUCKHOUT-WHITE, ELLEN GOLDMAN, IGOR MEDINTZ, JOSEPH MELINGER, US Naval Research Laboratory — Natural light harvesting systems exploit both strong and weak electronic coupling between chromophores to efficiently funnel energy to a reaction center. Here we use the DNA duplex as a scaffold to organize dyes with 4 Å precision and examine how tuning inter-dye electronic coupling from weakly to strongly coupled regimes effects resonance energy transfer (RET). We characterize an energy cascade of cyanine dyes rigidly linked to DNA with double attachment chemistry using ultrafast spectroscopy. When all inter-dye spacings are within 10 Å, molecular excitons form that possess character from all dyes, and the excited state dynamics show only weak wavelength dependence, consistent with strong electronic coupling. Reducing the spectral overlap by varying the dyes in the cascade reduces the electronic coupling strength and restores RET-like interactions where energy sequentially moves down the cascade. Placing a terminal acceptor at a remote distance from the coupled dyes shows efficient energy transfer from the molecular exciton. This study may provide insight into methods of enhancing the energy transfer efficiencies in synthetic light harvesting networks by incorporating both strong and weak electronic couplings.

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