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Ultrafast Nonlinear Frequency Generation in Excitonic Systems and the Dynamics of Novel Photosynthetic Pigment Analogs.

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Efficient energy transfer in many light harvesting complexes is mediated by excitonic coupling and delocalization. By directly exploiting exciton-exciton interactions using frequency generation ultrafast electronic spectroscopy, we can connect the spatial, temporal and dynamic landscapes of these complex systems. These measurements reveal the relationship between delocalized excitations even in spectrally congested aggregates, providing a novel and generalizable means to understand relaxation in strongly coupled systems. In addition, we will discuss ultrafast nonlinear spectroscopy measurements of new synthetic analogs of highly conserved natural light harvesting pigments. These molecular systems are tunable and redox active, providing new pathways to controllable and efficient energy and charge transfer in artificial light harvesting systems.