IMAGING EXCITED STATE DYNAMICS WITH 2D ELECTRONIC SPECTROSCOPY

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Excited states in the condensed phase have extremely high chemical potentials making them highly reactive and difficult to control. Yet in biology, excited state dynamics operate with exquisite precision driving solar light harvesting in photosynthetic complexes though excitonic transport and photochemistry through non-radiative relaxation to photochemical products. Optimized by evolution, these biological systems display manifestly quantum mechanical behaviors including coherent energy transfer, steering wavepacket trajectories through conical intersections and protection of long-lived quantum coherence. To image the underlying excited state dynamics, we have developed a new spectroscopic method allowing us to capture excitonic structure in real time. Through this method and other ultrafast multidimensional spectroscopies, we have captured coherent dynamics within photosynthetic antenna complexes. The data not only reveal how biological systems operate, but these same spectral signatures can be exploited to create new spectroscopic tools to elucidate the underlying Hamiltonian. New data on the role of the protein in photosynthetic systems indicates that the chromophores mix strongly with some bath modes within the system. The implications of this mixing for excitonic transport will be discussed along with prospects for transferring underlying design principles to synthetic systems.