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Photosynthetic Energy Transfer: Missing in Action (Detected Spectroscopy)?

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Two-dimensional electronic spectroscopy (2DES) has emerged as a powerful tool for studying electronic structure and dynamics in systems ranging from photosynthetic complexes to liquids and solid-state materials. In recent years, motivated by the desire to make spatially resolved measurements and to correlate optical excitation with a wide range of observables, action-detected variants of 2DES have been developed that employ a fully collinear geometry, relying on phase cycling or phase modulation to extract the signals of interest. I will discuss our recent implementations of fluorescence-detected two-dimensional electronic spectroscopy (F-2DES) and our studies of the light-harvesting II (LH2) complex from purple bacteria as a model system to understand F-2DES's ability to monitor energy transfer in multichromophoric systems. We demonstrate that the energy transfer process in LH2 is weak but observable in F-2DES, unlike in coherently detected 2DES where the energy transfer is visible with 100% contrast. We explain the weak signatures using a disordered excitonic model that accounts for experimental conditions. We find that the prominence of excited-state dynamics in action-detected spectroscopy offers a unique probe of excitonic delocalization in multichromophoric systems.

Keyword-1

multidimensional spectroscopy

Keyword-2

ultrafast dynamics

Keyword-3

energy transfer

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