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Dissecting quantum coherence in photosynthesis: lessons from porphyrin dimers AUSTIN SPENCER, Northwestern University, PYOSANG KIM, Argonne National Laboratory, WALEED HELWEH, LIN CHEN, Northwestern University — Many highly efficient photosynthetic protein complexes (PPCs) exhibit strong and persistent coherent spectral signatures that coincide with the early steps of the energy transport process. However, the inherent complexity and scale of proteins conceal the source of these coherences and their role in photosynthesis. To overcome these challenges, we apply broadband two-dimensional electronic spectroscopy (2DES) to porphyrins and their dimers as a bottom-up approach to studying energy transfer in PPCs. Using porphyrin dimers with moderate electronic coupling as a model for PPCs, we test the hypothesis that resonance between vibrational and electronic energy level gaps leads to strong nonadiabatic coupling. In a cofacial zinc porphyrin dimer, 2DES reveals a coherent spectral signature at 350 cm^{-1} that is absent in the monomer and which may be indicative of a Raman-activated vibration that is enhanced by resonance with the excitonic energy gap. Such a conclusion is consistent with the nonadiabatic coherence model developed for PPCs and supports the associated hypothesis that strong electronic–vibrational coupling drives both the observed coherent spectral signatures as well as rapid energy transfer.

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