



Quantum Efficiency Seminar and Colloquium David Coker

Theoretical Chemistry, University Boston

Modeling coherent excitation energy transfer in photosynthetic light harvesting

Recent 2D photon-echo experimental evidence suggests that the excitation energy transfer in light harvesting systems occurs coherently rather than via an incoherent hopping mechanism proposed in many earlier models of the process. More surprisingly, Scholes and co-workers have found evidence for coherent transfer even at ambient temperature in photosynthetic marine algae [E. Collini et. al, Nature 2010, 463, 644-647]. In this talk we use an iterative linearized density matrix (ILDM) propagation approach to study the coherent exciton transfer dynamics in phycocyanin PC645 from Chroomonas CCMP270 under ambient conditions (T=294K) with a multi-state system-bath dissipative model hamiltonian. The numerical results indicate that the oscillatory population beating lasts more than 400 fs and shows strong coherence between the DBV dimer and DBV-MBV bilin chromophores, an observation that agrees well with the experimental findings. Moreover, the quantum beating survives for nearly ten periods, and this long lived coherent superposition is likely to be responsible for providing a mechanism for the system to avoid excitation trapping and localization, providing sufficient time for the excitation to explore the entire complex and reach the acceptor, and thus has the potential to enhance the harvesting efficiency. Our calculations explore the influence of high and low frequency structures in the model environmental spectral density on the persistence of quantum coherence in these systems.

Date:Tuesday, February 1st, 20114:15 pmLocation:FRIAS Seminar Room, Albertstr. 19, Freiburg

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