MAR15-2014-020304

Abstract for an Invited Paper
for the MAR15 Meeting of
the American Physical Society

Anti-correlated vibrations drive fast non-adiabatic light harvesting[1]

DAVID JONAS, University of Colorado

We have recently shown that intramolecular vibrations shared across pigments can drive electronic energy transfer beyond the Born-Oppenheimer framework developed by Forster. The key features of this mechanism are a small change in vibrational equilibrium (less than the zero point amplitude) upon electronic excitation of the pigments and vibrational resonance with the adiabatic electronic energy gap. For identical pigments, delocalized, anti-correlated vibrations increase the speed of energy transfer. The same anti-correlated vibrations are excited by an electronically enhanced Raman process on the ground electronic state of photosynthetic antennas, and these vibrational wavepackets generate all of the reported signatures of photosynthetic energy transfer in femtosecond two-dimensional Fourier transform spectra. The talk will discuss how these results are generalized for differences between donor and acceptor and for multiple vibrations.

[1]This material is based upon work supported by the Air Force Office of Scientific Research under AFOSR Award No. FA9550-14-1-0258