Quantum Coherent Energy in Light-harvesting Systems

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Quantum coherence plays a central role in natural and artificial light-harvesting complexes and is explored in the context of transfer efficiency and sunlight excitation.

(1) The transfer rate in between LH2 is dramatically enhanced by the coherence resulting from the 9-fold ring symmetry. [1] We have developed a numerically exact method based on stochastic path integrals to predict emission spectra and generalized Forster rate of molecular aggregates and found that the enhanced rate in LH2 is 9ps without disorder and around 20ps with disorder, in consistent with previous measurements. [2]

(2) We have studied coherent quantum transport in disordered systems and clearly showed an optimal diffusion constant at an intermediate level of noise/temperature. [3,4] Detailed calculations indicate the crucial role of localization length and predict charge mobility in organic semiconductors close to experimental values. To simulate these large quantum systems, we have developed several numerical methods: transfer tensor method (TTM), stochastic path integrals (sPI), and stochastic hierarchy equation of motion (sHEOM). [5,6]

(3) The dynamics excited from the ground state incoherently can exhibit coherence on the picosecond time-scale for parameters relevant for photosynthetic complexes. We have derived a relationship between the properties of the steady-state on the exciton manifold and the transient dynamics induced by initial preparation, thus connecting incoherent sunlight excitation and ultrafast non-linear spectroscopy.

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(5) Jeremy and Cao, J. Chem. Phys., 139, 134106 (2013)
(6) Cerrillo and Cao, PRL 112, 110401 (2014)