

Quantum Biophysics: Old Roots, New Shoots.

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Essentially, any quantum systems can never be regarded as “isolated systems.” Quantum systems are always in contact with “the outside world,” and hence their quantum natures are sometimes sustained and sometimes destroyed. In condensed phase molecular systems, especially, quantum systems are affected by the huge number of dynamic degrees of freedom such as solvent molecules, amino acid residues in proteins, and so forth. Balance between robustness and fragility of the quantum natures may dramatically alter behaviors of chemical dynamics and spectroscopic signals. In this presentation, I will talk about three topics related to this subject.

The first topic is regarding natural photosynthetic systems. Recently, nuclear vibrational contribution signatures in two-dimensional (2D) electronic spectra of electronic energy transfer in light harvesting proteins and the primary charge separation in the reaction center have attracted considerable interest. As a plausible explanation for this long-lived spectral beating in 2D electronic spectra, quantum-mechanically mixed electronic and vibrational states were proposed and have since been explored. In this talk, we demonstrate that such electronic-vibrational quantum mixtures do not necessarily play a significant role in the electronic energy transfer dynamics and the charge separation, despite contributing to the enhancement of long-lived quantum beating in 2D electronic spectra, contrary to speculations in recent publications.

The second topic is about a model photovoltaic system. In organic photovoltaic materials, the recombination of the once separated electron and hole is a major loss mechanism. Hence, it is the key to elucidate physical mechanisms of how the electron and hole escape from the donor/acceptor interface for understanding the crucial factors determining the energy conversion efficiency of organic solar cells. In this talk, we investigate potential ratchet mechanism that was made possible via the combination of quantum delocalization and its decoherence to obtain insight into the inner working of experimentally observed ultrafast long-range charge separation and protection against the charge recombination at the donor/acceptor interface.

The third topic is quantum light spectroscopy for probing exciton and charge dynamics in complex molecules. Specifically, we address transmission measurement of frequency-entangled broadband photon pairs generated via parametric down-conversion with a monochromatic laser. It is observed that state-to-state dynamics in the system under study are temporally resolved by adjusting the path difference between the entangled twin beams when the entanglement time is sufficiently short. The non-classical photon correlation enables time-resolved spectroscopy with monochromatic pumping instead of a pulsed laser. It is further demonstrated that the signal corresponds to the spectral information along anti-diagonal lines of, for example, two-dimensional Fourier-transformed photon echo spectra. This correspondence inspires us to anticipate that more elaborately engineered photon states would broaden the availability of quantum light spectroscopy.