Quantum Coherence for Light Harvesting

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Two-dimensional electronic spectroscopy has proven to be especially well suited for studying coupled molecular systems, providing new insights into the population and coherence dynamics. Multiple ultra short laser pulses are employed to extract maximum possible information about the system in both time and energy [1]. Amplitude oscillations in the 2D spectra is a direct evidence of the dynamically evolving superposition states. Interestingly enough, they were initially interpreted as a purely electronic superposition with a conclusion that excitation energy can be transferred coherently through the light-harvesting complexes. This finding stimulated an immense scientific effort to understand the mechanisms of such wave-like energy transfer [2]. In the case of photosynthetic reaction center, we identified a new photophysical mechanism of the coherent dynamics; Energy Transfer Induced Coherence Shift (ETICS). In ETICS mechanism, the initial excited state coherence is shifted to the ground state of the donor upon energy transfer step without the loss of vibronically mixed character and phase information. Yet it acquires the picosecond lifetime of the ground state vibrational coherence [3]. Our results suggest that the hypothesis of the wave-like energy transfer has to be revisited.

References

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