

Revealing quantum coherence in photosynthetic complexes using ultrafast spectroscopy: Simulation studies

Qiang Shi*, Shuming Bai, Kai Song

Institute of Chemistry, Chinese Academy of Sciences, No.2 North 1st Street,
Zhongguancun, Beijing, 100190, China, *qshi@iccas.ac.cn

Simulations are performed to study quantum coherence in photosynthetic complexes using ultrafast spectroscopy. It is found that: (1) Different types of quantum coherence can be distinguished in pump-probe polarization anisotropy measurements. (2) Specific pulse shaping schemes are important in observing coherent energy transfer in the single molecule two-color double-pump experiment.

We will present simulation studies of the quantum coherence in two different types of ultrafast spectroscopic measurements: (1) Effects of three types of quantum coherence: electronic, vibrational, and vibronic coherences, on the ultrafast pump-probe polarization anisotropy are studied. Simulations of model dimer systems show that they have rather different spectral characteristics, which can be used to determine the type of coherence that exists in specific systems. We then simulate the pump-probe spectra of the Fenna-Matthews-Olson (FMO) complex at low temperature, and show that a model with only quantum coherence agrees with previous experiments. Finally, the condition in which long time vibronic coherence can be observed in systems like the FMO complex is investigated. (2) Effects of pulse shaping on observing coherent energy transfer in the single bacterial light harvesting 2 (LH2) complex are studied, as in the recent single molecule two-color double-pump experiment (*Science* **340**, 1448 (2013)). It is found that, quantum coherent energy transfer can be observed when the time delay and phase difference between the two laser pulses are controlled independently. However, the pulse shaping schemes have great impact on observing quantum coherent energy transfer.

References:

- [1] S. Savikhin, D. R. Buck, and W. S. Struve, *Chem. Phys.* **223**, 303 (1997).
- [2] S.-M. Bai, K. Song, and Q. Shi, *J. Phys. Chem. Lett.* **6**, 1954 (2015).
- [3] K. Song, S.-M. Bai, and Q. Shi, *in preparation*.