Unraveling quantum coherences in photosynthetic protein complexes at ultralow temperatures

Hong-Guang Duan[2,3], Ajay Jha[2,3,4], Pan-Pan Zhang[3], Vandana Tiwari[2,5], Lipeng Chen[6], Richard J. Cogdell[7], Khuram Ashraf[7], Valentyn I. Prokhorenko[2], Michael Thorwart[1,8], and R.J. Dwayne Miller[9]

[1]*Universität Hamburg, I. Institut für Theoretische Physik, Hamburg, Germany*
[2]*Max Planck Institute for the Structure and Dynamics of Matter, Hamburg, Germany*
[3]*Department of Physics, Ningbo University, Ningbo, P.R. China*
[4]*Rosalind Franklin Institute, Harwell, Oxfordshire OX11 0QX, UK*
[5]*Department of Chemistry, Universität Hamburg, Hamburg, Germany*
[6]*Zhejiang Laboratory, Hangzhou 311100, P.R. China*
[7]*School of Molecular Biosciences, University of Glasgow, Glasgow, UK*
[8]*The Hamburg Center for Ultrafast Imaging, Hamburg, Germany*
[9]*The Departments of Chemistry and Physics, University of Toronto, Toronto, Canada.*

The transfer of excitation energy in biomolecular complexes is inherently mediated by quantum delocalization. It is an interesting question to what extent does quantum coherence play a role in the transfer dynamics. I will discuss under what conditions such natural systems could show dynamic quantum coherent effects beyond the trivial quantum delocalization. In recent joint experimental and theoretical studies [1-3], we have investigated the quantum exciton dynamics in the Fenna-Matthews-Olson (FMO) complex and the Photosystem II Reaction Center (PSIIRC) by two-dimensional electronic spectroscopy in a large range of temperatures down to 20 K. Our experimental results reveal electronic coherence to occur on a time scale as long as 500 fs at 20 Kelvin for the case of FMO [2] and of about 200 fs for PSIIRC at 20 Kelvin [1]. They complete earlier results obtained under ambient conditions where we have found that at room temperature, electronic coherence fades out within 60 fs [3]. Yet, the new low-temperature data allow us to capture evidence of quantum coherence at ultralow temperature and to clearly disentangle electronic and vibrational dynamic coherence. The observed long-lived oscillations are due to Raman vibrational modes on the electronic ground state.

[1] A. Jha, P.-P. Zhang, V. Tiwari, L. Chen, M. Thorwart, R.J.D. Miller, and H.-G. Duan, Unraveling Quantum Coherences Mediating Primary Charge Transfer Processes in Photosystem II Reaction Center, Science Adv. 10 (2024) eadk1312.

[2] Hong-Guang Duan, Ajay Jha, Lipeng Chen, Vandana Tiwari, Richard J. Cogdell, Khuram Ashraf, Valentyn I. Prokhorenko, Michael Thorwart, and R. J. Dwayne Miller, Quantum Coherent Energy Transport in the Fenna-Matthews-Olson Complex at Low Temperature, Proc. Natl. Acad. Sci. 119 (2022) e2212630119.

[3] Hong-Guang Duan, Valentyn I. Prokhorenko, Richard Cogdell, Khuram Ashraf, Amy L. Stevens, Michael Thorwart, and R. J. Dwayne Miller, Nature does not rely on long-lived electronic quantum coherence for photosynthetic energy transfer, Proc. Natl. Acad. Sci. 114 (2017) 8493.