Two-dimensional Electronic Spectroscopy of Fenna-Matthews-Olson Complex at Ambient Temperature

Hong-Guang Duan[1,2,3], Valentyn I. Prokhorenko[1], Richard Cogdell[4], Khuram Ashraf[4], Michael Thorwart[2,3], and R. J. Dwayne Miller[1,3,5]

[1]Max Planck Institute for the Structure and Dynamics of Matter, Luruper Chaussee 149, 22761, Hamburg, Germany
[2]I. Institute for Theoretical Physics, University of Hamburg, Jungiusstrasse 9, 20355 Hamburg, Germany
[3]The Hamburg Center for Ultrafast Imaging, Luruper Chaussee 149, 22761 Hamburg, Germany
[4]Institute of Molecular, Cell and Systems Biology, College of Medical, Veterinary and Life Sciences, University of Glasgow, Glasgow G12 8QQ, UK
[5]Department of Chemistry and Physics, University of Toronto, 80 St. George Street, Toronto Canada M5S 3H6,
dwayne.miller@mpsd.mpg.de

We have performed the 2D electronic spectroscopy of the Fenna-Matthews-Olson (FMO) complex using a broadband laser source with transform-limited 16-fs pulses. No clear oscillations were observed in a series of 2D spectra at ambient temperature.

The FMO complex is found in the low light-adapted green sulfur bacteria. Its task is the energy transport from peripheral chlorosomal antennae to the reaction center. The structure [1] shows three identical subunits, each contains eight bacteriochlorophyll a molecules. By virtue of its relatively small size, it provides a very important model for the study of natural excitation energy transfer (EET) and has been extensively studied experimentally and theoretically. In 1990s, Savikhin et al. [2] observed a quantum beating in the FMO complex at low temperature using time-resolved fluorescence anisotropy. Recently, the 2D spectra of FMO complex were measured at 77K [3] and 277K [4], long-lived oscillations were reported. However, the origin of these long-lived oscillations is still unclear and under debate. In fact, as an alternative explanation, vibrationally-assisted quantum coherence was recently proposed to explain these observations. In order to clarify the nature of this oscillation, we have performed photon-echo electronic 2D spectroscopy of the FMO complex at ambient temperature. The energy transfer pathways and the corresponding time scales were identified using a global fit approach. No obvious long-lived quantum coherence can be identified after the Fourier-transform analysis of the residuals.

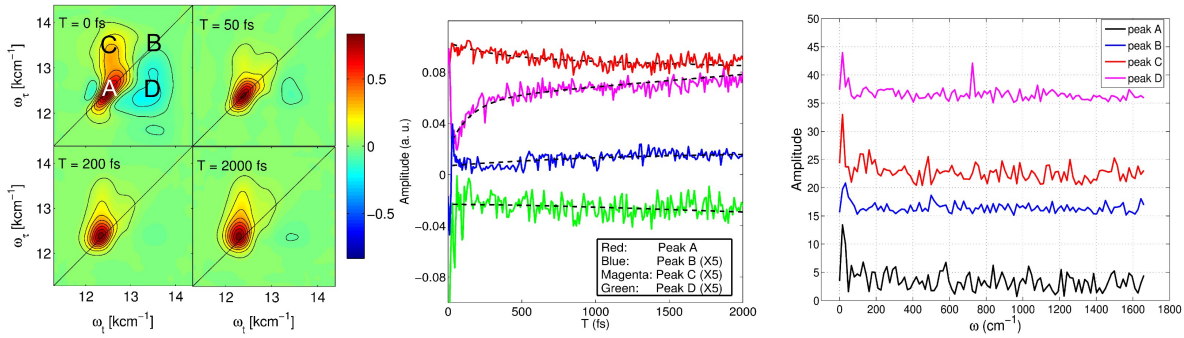

Fig. 1. Left: measured 2D spectra at different waiting times. Middle: decay trace for the spectral position on 2D spectra depicted on the left. Right: Fourier transform of the residuals after substracting the fit of population dynsmics (black dash line in middle figure).

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