Quantum Beats in the Fenna-Matthews-Olson Complex

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Long- and short-lived quantum beats observed in the ultrafast dynamics of the FMO complex are studied at cryogenic temperature by polarization-controlled 2D electronic spectroscopy. The observed response can be explained with a straight-forward vibronic model that does not require correlated bath interactions.

Over the last 40 years, since the determination of its crystal structure in the mid 70-s, the Fenna-Matthews-Olson complex (FMO) found in certain green sulfur bacteria has been one of the most thoroughly studied photosynthetic complexes [1]. In 2007 FMO reappeared at the forefront of photosynthetic research when long-lived quantum beats were observed in electronic 2D experiments. These were interpreted as originating from excitonic coherences, and were claimed to be evidence for wavelike energy transfer [2]. Subsequently significant experimental and theoretical efforts have been made to substantiate this claim [3,4]. Meanwhile, alternative theoretical explanations to the long-lived quantum beat (QB) contributions not requiring longevity of electronic superpositions have been proposed [5,6,7], however no clear experimental evidence has so far emerged for either view.
Here we use polarization-controlled ultrafast electronic 2D spectroscopy to show that the observed QBs in the FMO spectra are primarily of ground-state vibrational origin, with strongly electronic contributions dephasing in <150 fs.

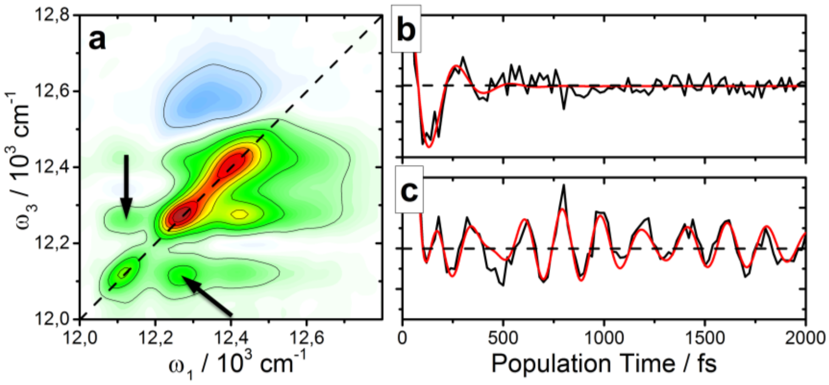

Fig.1 (a) Real 2DES spectrum at 100 fs population time at 77 K, recorded with a <0,0,0,0> pulse-polarization sequence. (b) Rephasing quantum beat kinetics in the above-diagonal cross-peak between exciton 1 and 2. (c) Rephasing quantum beat kinetics in the below-diagonal cross-peak between exciton 1 and 2. Fit to data as solid red line in (b) and (c). Kinetics extracted in points indicated by arrows

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