Supplementary Information

Multiple Quantum-Coherent Energy Transfer Pathways in Photosynthesis: Electronic-Vibrational Mixing within Photosystem II CP43/CP47 Antenna

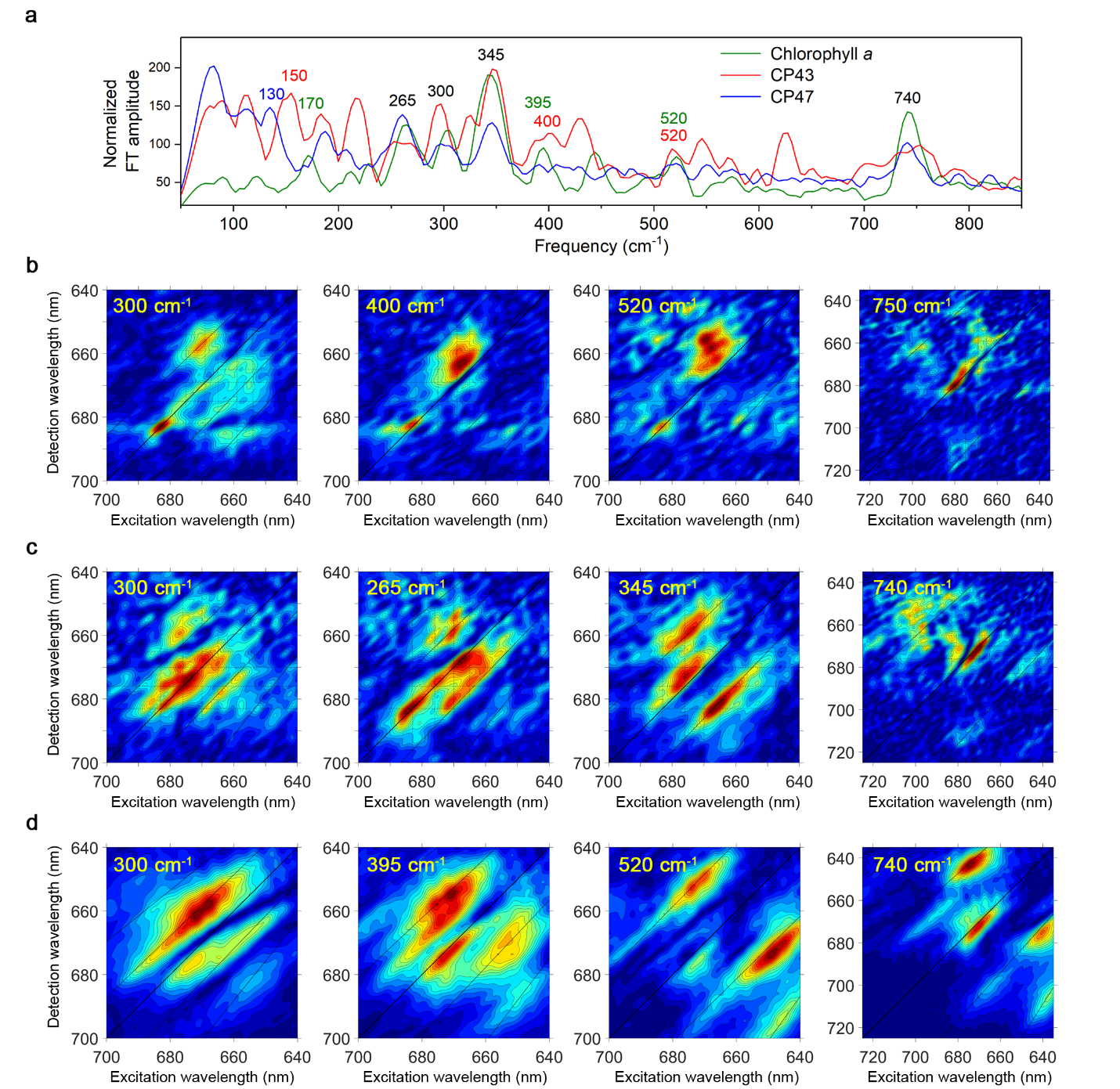
Valentin Maffeis1,3, Patricia Lorente2, Rafael Picorel2, Elisabet Romero1,\*

*1Institute of Chemical Research of Catalonia, Barcelona Institute of Science and Technology, 43007 Tarragona, Spain*

*2Consejo Superior de Investigaciones Científicas, 50059 Zaragoza, Spain*

*3Present address: Université Lyon, ENS Lyon, CNRS, 69364 Lyon, France*

\*eromero@iciq.es



**Fig. S1 Oscillation frequencies and real rephasing 2D frequency maps for CP43, CP47 and monomeric Chl at 77 K.** (**a**) Fourier transform (FT) of the real rephasing 2D spectral traces where the maximum amplitude value of each frequency across the full 2D spectra dataset is displayed. The FT is performed over a population time window 104 – 1504 fs (frequency resolution ± 20 cm-1) with a step of 8 fs (maximum probed frequency ≈ 2100 cm-1). Note that the monomeric Chl data has been divided by a factor of two to facilitate the comparison among the three datatsets. (**b**) Real rephasing 2D frequency maps for CP43 at 300, 400, 520, and 750 cm-1 oscillation frequencies. (**c**) Real rephasing 2D frequency maps for CP47 at 300, 265, 345, and 740 cm-1 oscillation frequencies. (**d**) Real rephasing 2D frequency maps for monomeric Chl at 305, 395, 520, and 740 cm-1 oscillation frequencies.

The 2D frequency maps for several oscillation frequencies are shown in Fig.S1. Note that some major frequencies displayed in Fig.3 are kept in Fig.S1 for comparison.

For CP43, the 300 cm-1 oscillation frequency has significant amplitude in the FT even thought this frequency only appears as a weak band in the FLN spectrum[1](#_ENREF_1). This energy corresponds to the CP669-682 and it has an amplitude distribution similar to the 265 cm-1 shown in Fig.3b, yet with much lower amplitude at the CP position. Therefore, this frequency map can be assigned to a combination of both vibrational[2-7](#_ENREF_2) and vibronic[6](#_ENREF_6),[8](#_ENREF_8),[9](#_ENREF_9) coherence, but with lower vibronic contribution than observed in 265 cm-1 map. The 400 and 520 cm-1 maps display similar amplitude distribution, both show an intense Diagonal682 feature together with minor amplitude around the line below diagonal at the position 682 nm on the detection axis; that is, at (665,682) and (659,682) nm, respectively. These features, which are also present in the 740 cm-1 frequency map around (650,680) nm, indicate that these maps correspond to vibrational coherence.

For CP47, only one frequency map different from the ones displayed in Fig. 3c is shown: the one corresponding to the 300 cm-1 frequency, which similarly to CP43, only appears as a weak band in the FLN spectrum[10](#_ENREF_10). The 300 cm-1 frequency map amplitude distribution is similar to the one found in the 265 cm-1 map, yet with lower CP amplitude. Hence, this map is interpreted as the 265 cm-1 map: it corresponds to vibronic coherence between the 670 and 683 nm states as well as between the 665 and 676 nm states (CP670-683 and CP665-676, respectively).

For monomeric Chl, all the frequency maps correspond to vibrational coherence, the 300 and 395 cm-1 maps represent vibrational coherence generated via de SE, whereas the 520 and 740 cm-1 maps indicate vibrational coherence generated via GSB and SE[6](#_ENREF_6).

**References**

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