## Supporting Information For:

## Vibronic Wavepackets and Energy Transfer in Cryptophyte Light Harvesting Complexes

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## **DOAS Supplemental**

Figure S1: Selected time traces of PC577 data (grey, orange, cyan in mOD) and fit (black, red, blue). Wavelength step 1.172 nm, every second wavelength (indicated in the ordinate label) is depicted. Zoom from -0.1 ps to 0.4 ps (after the maximum of the IRF).





Figure S2: Selected time traces of PC612 data (grey, orange, cyan in mOD) and fit (black, red, blue). Wavelength step 1.172 nm, every second wavelength (indicated in the ordinate label) is depicted. Zoom from -0.1 ps to 0.4 ps (after the maximum of the IRF).



Figure S3: Selected time traces of PC630 data (grey, orange, cyan in mOD) and fit (black, red, blue). Wavelength step 1.172 nm, every second wavelength (indicated in the ordinate label) is depicted. Zoom from -0.1 ps to 0.4 ps (after the maximum of the IRF).



Figure S4: Selected time traces of PC645 data (grey, orange in mOD) and fit (black, red). Wavelength step 1.172 nm, every second wavelength (indicated in the ordinate label) is depicted. Zoom from -0.1 ps to 0.4 ps (after the maximum of the IRF).

Figure S5: Oscillatory Features. Population dynamics (bottom plot) at 580 nm from the four pump probe spectra as colourlabelled (top 4 plots): PC577 (black), PC612 (red), PC630 (blue), PC645 (green). Experimental sampling frequency is reduced after 2 ps.



Figure S6: Overview of the estimated DOAS and phases of PC577. (A) species populations from Figure 2 depicted until 0.3ps. The maximum of the IRF is at time zero. (B) SADS from Figure 2C. In addition the negative of the steady state absorption (magenta) and emission (cyan) and the pulse spectrum (black) are plotted for reference. (C) superposition of five selected DOAS with frequencies between 332 and 1583/cm, and damping rates between 1.0 and 2.6/ps, which are detailed in panels D-I. Row 2-7: (left column) Cosine oscillations with frequencies  $\tilde{\nu}n$  (in /cm) (where *n* is the DOAS number) and damping rates  $\gamma$  (in 1/ps) written in the legend at the left, using the appropriate color. (middle column) Estimated DOAS (with number indicated in the legend at the far left), normalized (norm) for comparison. (right column) Estimated phase profiles of the DOAS.



Figure S7: Overview of the estimated DOAS and phases of PC612. (A) species populations from Figure 2 depicted until 0.3ps. The maximum of the IRF is at time zero. (B) SADS from Figure 2C. In addition the negative of the steady state absorption (magenta) and emission (cyan) and the pulse spectrum (black) are plotted for reference. (C) superposition of four selected DOAS with frequencies between 663 and 1585/cm, and damping rates between 0.9 and 3.0/ps, which are detailed in panels D-I. Row 2-7: (left column) Cosine oscillations with frequencies  $\tilde{vn}$  (in /cm) (where *n* is the DOAS number) and damping rates  $\gamma$  (in 1/ps) written in the legend at the left, using the appropriate color. (middle column) Estimated DOAS (with number indicated in the legend at the far left), normalized (norm) for comparison. (right column) Estimated phase profiles of the DOAS.



Figure S8: Overview of the estimated DOAS and phases of PC630. (A) species populations from Figure 2 depicted until 0.3ps. The maximum of the IRF is at time zero. (B) SADS from Figure 2C. In addition the negative of the steady state absorption (magenta) and emission (cyan) and the pulse spectrum (black) are plotted for reference. (C) superposition of five selected DOAS with frequencies between 276 and 1583/cm, and damping rates between 0.8 and 4.4/ps, which are detailed in panels D-I. Row 2-7: (left column) Cosine oscillations with frequencies  $\tilde{vn}$  (in /cm) (where *n* is the DOAS number) and damping rates  $\gamma$  (in 1/ps) written in the legend at the left, using the appropriate color. (middle column) Estimated DOAS (with number indicated in the legend at the far left), normalized (norm) for comparison. (right column) Estimated phase profiles of the DOAS.



Figure S9: Overview of the estimated DOAS and phases of PC645. (A) species populations from Figure 2 depicted until 0.3ps. The maximum of the IRF is at time zero. (B) SADS from Figure 2C. In addition the negative of the steady state absorption (magenta) and emission (cyan) and the pulse spectrum (black) are plotted for reference. (C) superposition of five selected DOAS with frequencies between 285 and 1585/cm, and damping rates between 1.0 and 4.8/ps, which are detailed in panels D-I. Row 2-7: (left column) Cosine oscillations with frequencies  $\tilde{vn}$  (in /cm) (where *n* is the DOAS number) and damping rates  $\gamma$  (in 1/ps) written in the legend at the left, using the appropriate color. (middle column) Estimated DOAS (with number indicated in the legend at the far left), normalized (norm) for comparison. (right column) Estimated phase profiles of the DOAS.



Table S1: Identified DOAS and their parameters from global fits: oscillation

frequency  $\omega_n$  (cm<sup>-1</sup>) dephasing rate  $\gamma_n$  (ps<sup>-1</sup>) and the area of the DOAS divided by the dephasing rate  $\gamma_n$ .

PC577		area / γ <sub>n</sub>	ω <sub>n</sub>	error	Yn	error
	DOAS1	10	1642.8	0.5	2.35	0.10
	DOAS2	26	1583.4	0.2	2.61	0.04
	DOAS3	7	1454.9	0.8	2.94	0.15
	DOAS4	12	1339.2	0.8	6.21	0.16
	DOAS5	12	1235.0	0.4	2.44	0.07
	DOAS6	11	1017.7	2.0	8.44	0.39
	DOAS7	18	916.2	1.9	8.46	0.36
	DOAS8	12	866.1	0.4	1.42	0.07
	DOAS9	29	803.6	0.7	6.66	0.13
	DOAS10	32	662.2	0.1	1.04	0.03
	DOAS11	22	648.4	1.5	9.31	0.31
	DOAS12	13	521.1	0.4	2.10	0.07
	DOAS13	18	472.9	0.3	2.15	0.05
	DOAS14	16	403.6	1.0	3.85	0.20
	DOAS15	35	357.4	1.8	9.25	0.37
	DOAS16	15	332.1	0.3	1.40	0.06
	DOAS17	29	263.4	0.3	2.64	0.05
	DOAS18	21	223.5	0.4	3.10	0.08

PC612		area / γ <sub>n</sub>	ω <sub>n</sub>	error	Yn	error
	DOAS1	10	1649.0	0.4	2.06	0.08
	DOAS2	33	1585.2	0.2	2.98	0.03
	DOAS3	8	1466.3	0.8	3.97	0.15
	DOAS4	16	1372.9	0.9	3.16	0.16
	DOAS5	21	1348.7	0.6	2.94	0.11
	DOAS6	16	1235.5	0.2	2.05	0.04
	DOAS7	9	1077.6	0.9	3.92	0.18
	DOAS8	15	1047.3	0.3	1.18	0.05
	DOAS9	10	971.5	0.5	2.07	0.09
	DOAS10	60	880.9	4.2	11.97	0.79
	DOAS11	23	863.1	0.4	1.96	0.08
	DOAS12	65	823.5	3.9	10.65	0.78
	DOAS13	29	810.5	0.3	2.34	0.06
	DOAS14	13	697.0	0.8	2.86	0.15
	DOAS15	41	663.0	0.1	0.90	0.02
	DOAS16	32	652.1	1.5	7.40	0.29
	DOAS17	20	515.4	0.3	2.44	0.07
	DOAS18	32	475.6	0.2	2.27	0.05
	DOAS19	28	417.1	1.2	6.49	0.24
	DOAS20	88	325.0	2.7	8.46	0.56
	DOAS21	36	319.2	1.1	3.32	0.22
	DOAS22	68	261.4	0.6	4.45	0.13
	DOAS23	35	227.8	0.7	3.80	0.12
	DOAS24	24	130.1	1.3	7.78	0.28

PC630		area / γ <sub>n</sub>	ω <sub>n</sub>	error	Yn	error
	DOAS1	30	1583.5	0.2	4.40	0.04
	DOAS2	8	1460.2	0.8	3.40	0.16
	DOAS3	15	1331.4	0.9	4.71	0.17
	DOAS4	12	1241.1	0.4	1.70	0.07
	DOAS5	9	1035.3	0.4	1.75	0.08
	DOAS6	30	887.8	2.5	10.74	0.48
	DOAS7	29	830.2	0.4	2.39	0.07
	DOAS8	47	814.2	3.0	9.51	0.57
	DOAS9	21	722.7	3.3	11.44	0.65
	DOAS10	23	662.4	0.2	1.96	0.04
	DOAS11	14	514.5	0.7	3.44	0.14
	DOAS12	16	485.8	0.3	0.82	0.06
	DOAS13	26	472.2	0.7	4.26	0.13
	DOAS14	13	391.1	0.4	1.97	0.08
	DOAS15	62	328.8	0.8	4.44	0.15
	DOAS16	89	327.8	0.0	8.51	0.00
	DOAS17	22	275.7	0.3	1.09	0.05
	DOAS18	80	257.4	3.2	9.25	0.61
	DOAS19	59	220.6	1.7	7.39	0.32

PC645		area / γ <sub>n</sub>	ω <sub>n</sub>	error	Yn	error
	DOAS1	22	1585.4	0.4	4.81	0.07
	DOAS2	15	1321.6	1.2	5.24	0.24
	DOAS3	17	1257.6	2.3	10.89	0.44
	DOAS4	8	1040.2	0.7	2.36	0.13
	DOAS5	26	869.9	0.6	3.10	0.12
	DOAS6	23	849.3	0.5	1.46	0.09
	DOAS7	21	821.1	0.5	2.32	0.10
	DOAS8	16	666.2	0.3	1.43	0.06
	DOAS9	12	520.4	0.9	3.17	0.17
	DOAS10	22	477.6	0.5	3.26	0.10
	DOAS11	15	405.6	1.0	3.72	0.19
	DOAS12	21	330.1	0.5	1.92	0.10
	DOAS13	44	309.0	2.4	8.45	0.48
	DOAS14	15	284.6	0.4	1.01	1.01
	DOAS15	27	212.7	0.9	5.88	0.17