Supplementary Information

Molecular Aggregates Associated THz Coherent Vibrations Contribute to Enhancements of Emission Efficiencies

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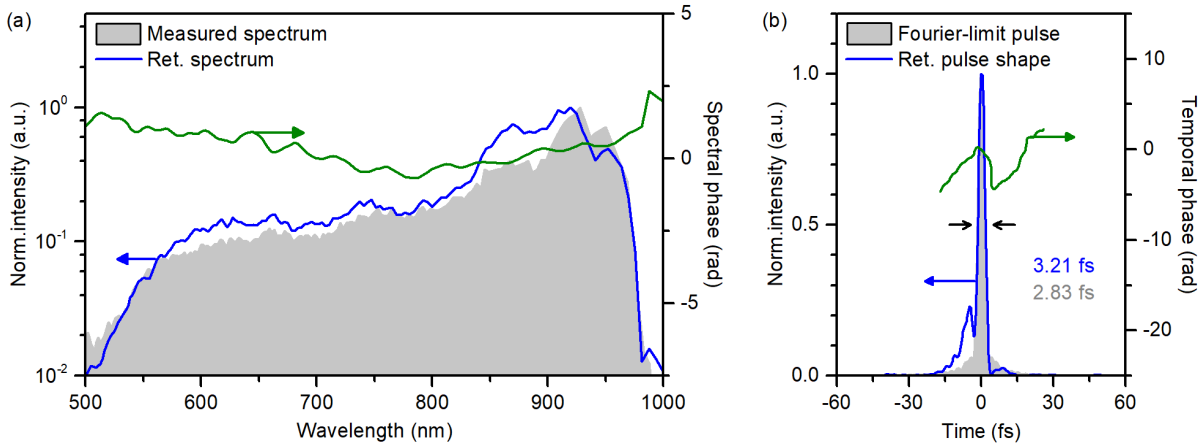

Figure S1 | (a) Spectral information of the pump pulse. Grey area shows the spectrum measured by a commercial spectrometer (HR4000, Ocean optics). The blue and green curves indicate the spectral intensity and phase retrieved by PG-FROG, respectively. (b) Temporal intensity (blue curve) and phase (green curve) measured by PG-FROG. The grey area indicates the Fourier-limit pulse shape.

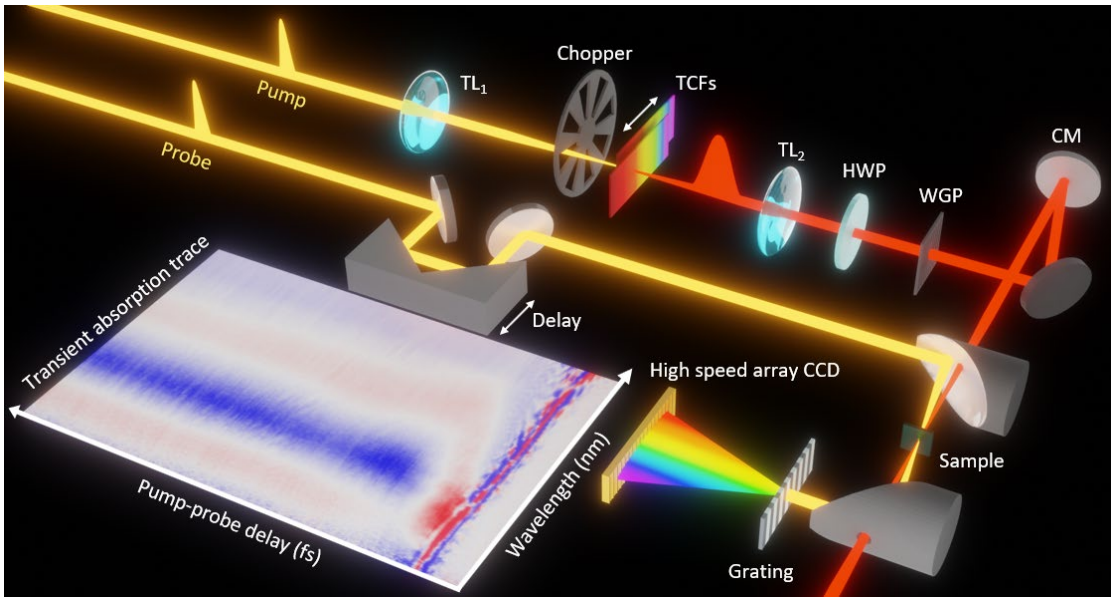

Figure S2 | Experimental setup of ultrafast shot-to-shot transient absorption spectroscopy. TL#: thin lens; TCFs: tunable color filters; HWP: half-wave plate; WGP: wire-grid polarizer. CM: concave mirror.

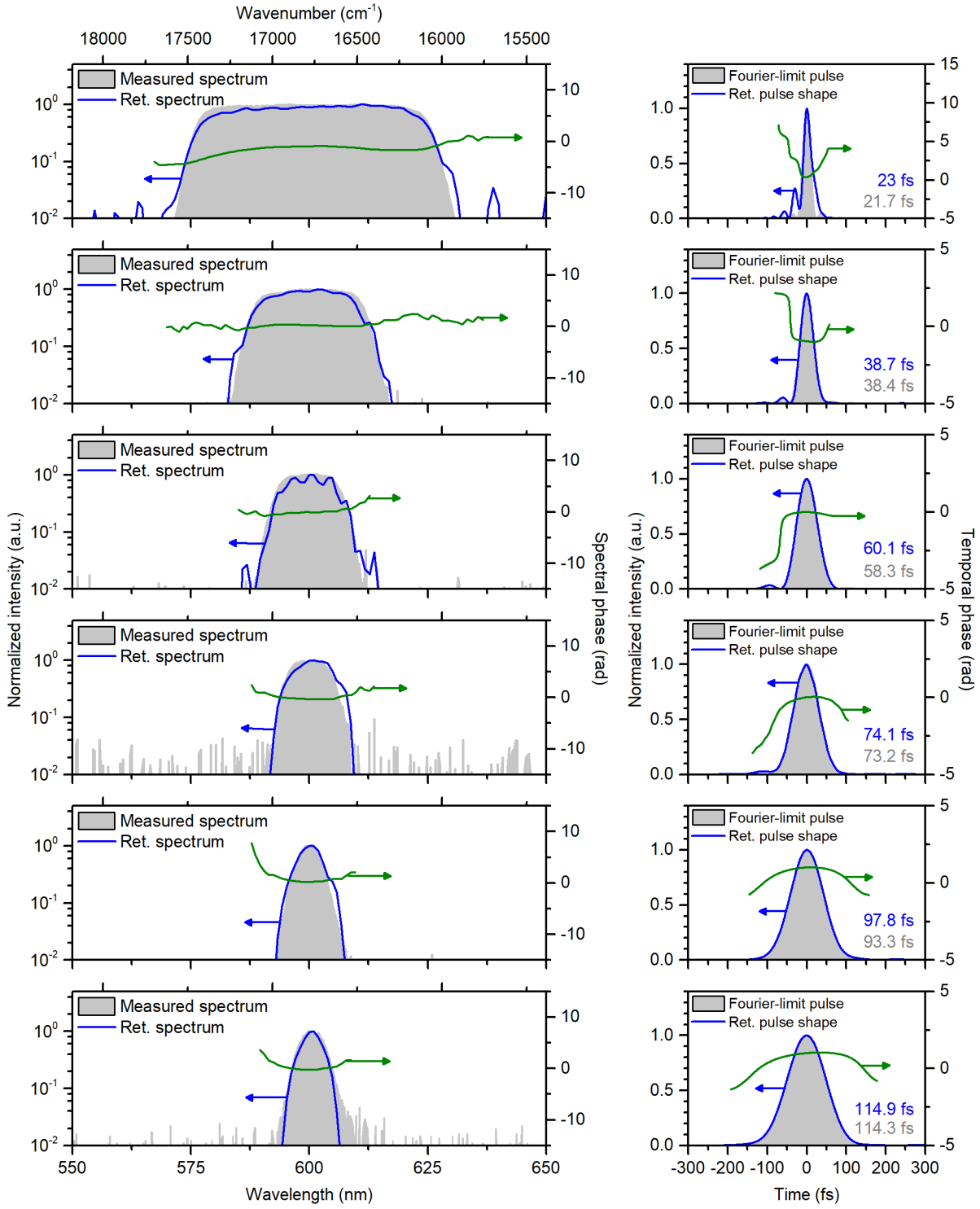

Figure S3 | PG-FROG measurements of pump pulses with different durations. Left panel: measured spectrum (grey area), retrieved spectrum (blue curve) and retrieved spectral phase (green curve). Right panel: Fourier-limit pulse shape obtained from the measured spectrum (grey area), retrieved temporal intensity profile (blue curve) and retrieved temporal phase (green curve). The numbers denote the full-width-at-half-maximum (FWHM) pulse duration of the retrieved pulse (blue) and the case of a Fourier-limit pulse (grey).

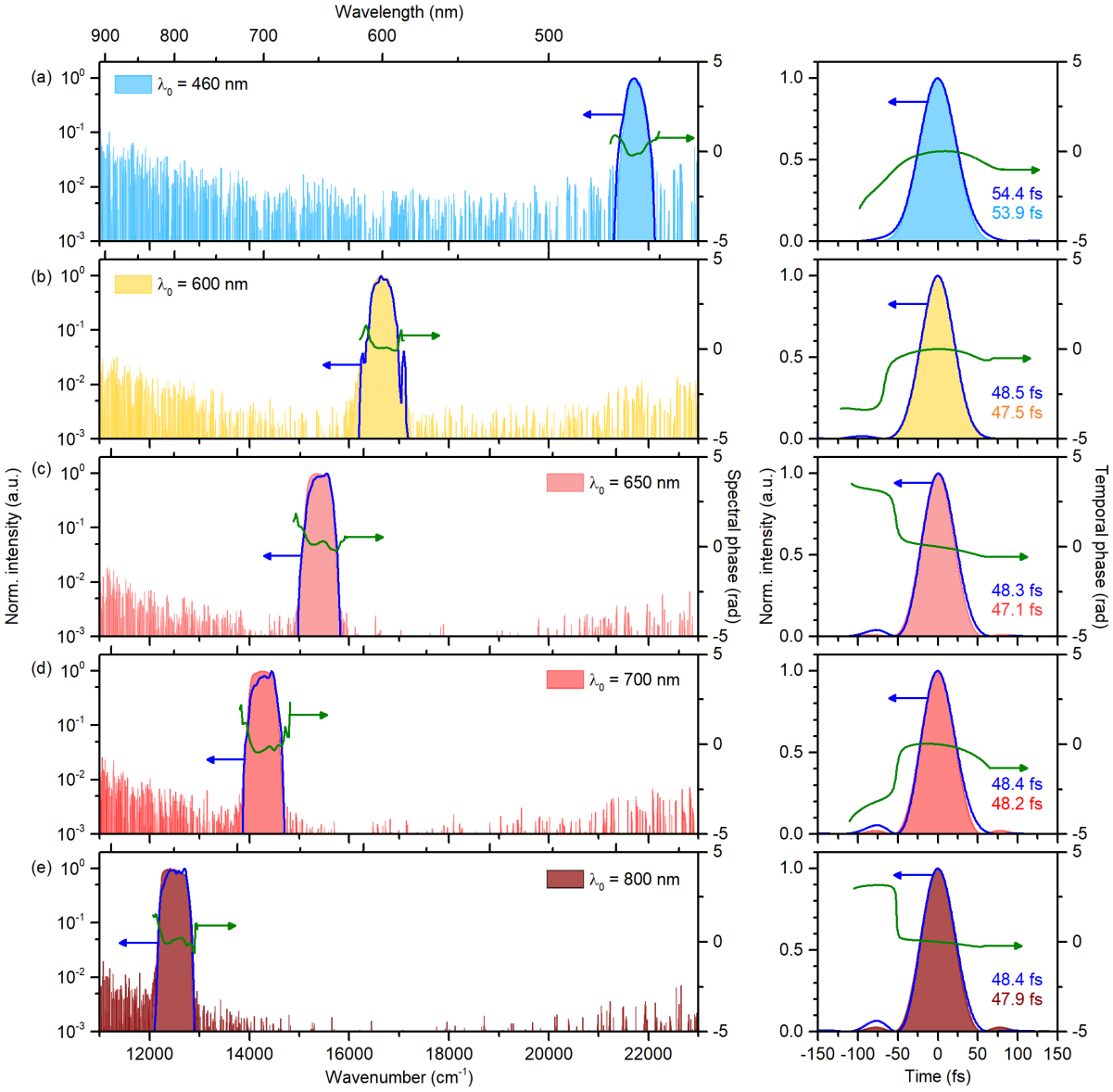

Figure S4 | PG-FROG measurements of pump pulses with different central wavelengths while keeping the same pulse duration. Left panel: measured spectrum (colored area), retrieved spectrum (blue curve) and retrieved spectral phase (green curve). Right panel: Fourier-limit pulse shape obtained from the measured spectrum (colored area), retrieved temporal intensity profile (blue curve) and retrieved temporal phase (green curve). The numbers denote the full-width-at-half-maximum (FWHM) pulse duration of the retrieved pulse (blue) and the case of a Fourier-limit pulse (color-coded).

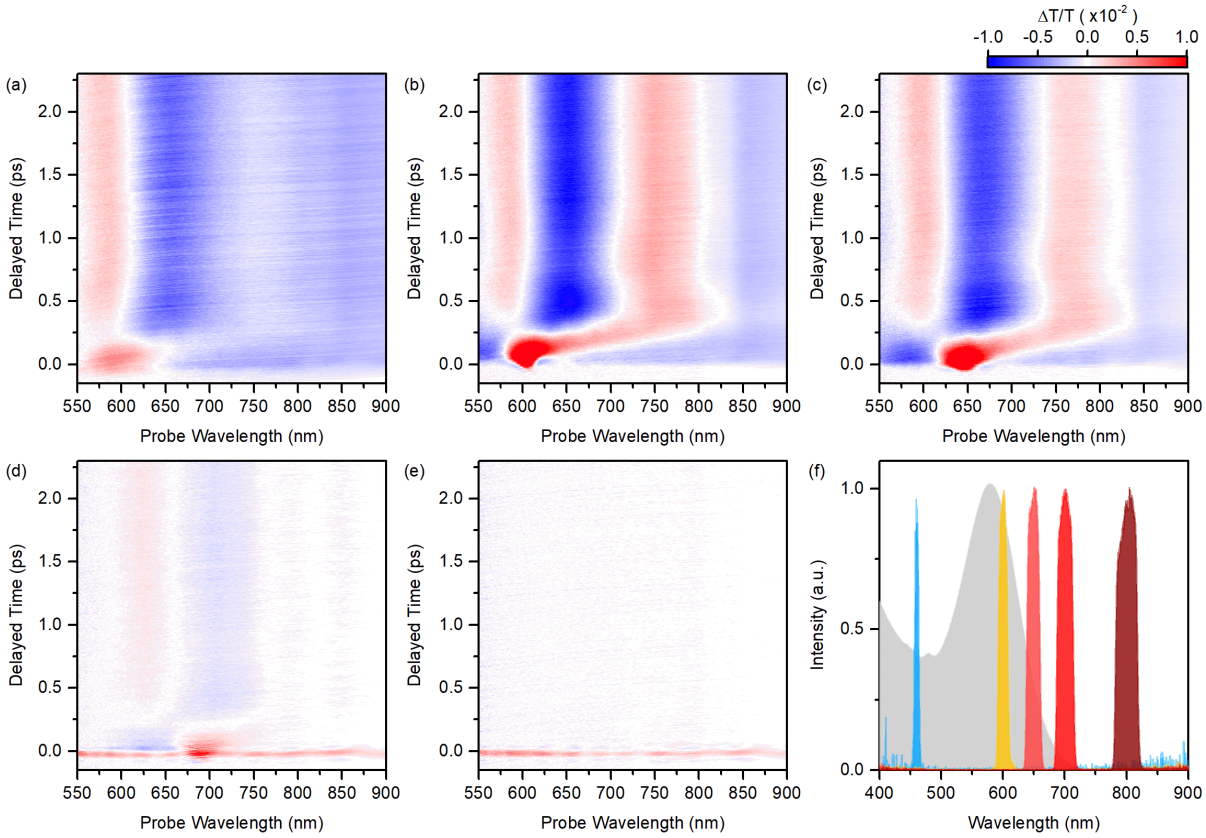

Figure S5 | Central excitation wavelength (λ_0)-dependent TA spectra at room temperature. (a) λ_0 = 460 nm, (b) λ_0 = 600 nm, (c) λ_0 = 650 nm, (d) λ_0 = 700 nm and (e) λ_0 = 800 nm. (f) Comparison of the stead-state absorption spectra (grey shaded area) and the spectra of the pump pulse (blue, yellow, light red, red and brown shaded areas) corresponding to the spectra in Fig. S4a-S4e, respectively.

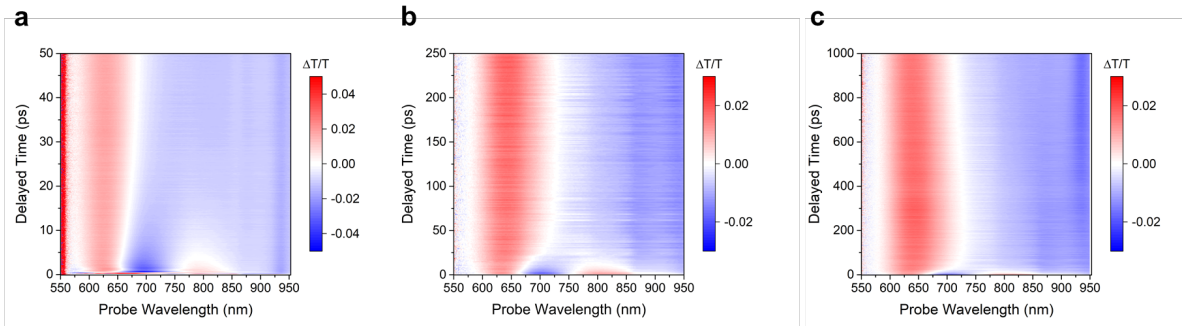

Figure S6 | TA spectra at room temperature displayed over different delay windows. (a) 0 ~ 50 ps, (b) 0 ~ 250 ps and (c) 0 ~ 1 ns. The pump pulse duration and the corresponding spectra are shown in Fig. S2.

Table S1 | Calculated spin orbit coupling of the tetrameric **4H** at the S_0 equilibrium geometry.

	Spin orbit Coupling (cm^{-1})
$\lvert\langle S_1 \rvert H_{SOC} \rvert T_1\rangle\rvert$	11.7
$\lvert\langle S_1 \rvert H_{SOC} \rvert T_2\rangle\rvert$	29.3
$\lvert\langle S_1 \rvert H_{SOC} \rvert T_3\rangle\rvert$	613.1
$\lvert\langle S_1 \rvert H_{SOC} \rvert T_4\rangle\rvert$	300.4
$\lvert\langle S_1 \rvert H_{SOC} \rvert T_5\rangle\rvert$	543.9
$\lvert\langle S_1 \rvert H_{SOC} \rvert T_6\rangle\rvert$	689.2

Table S2 | Calculated spin orbit coupling of the tetrameric **4H** at the S_1 equilibrium geometry.

	Spin orbit Coupling (cm^{-1})
$\lvert\langle S_1 \rvert H_{SOC} \rvert T_1\rangle\rvert$	1.2
$\lvert\langle S_1 \rvert H_{SOC} \rvert T_2\rangle\rvert$	10.6
$\lvert\langle S_1 \rvert H_{SOC} \rvert T_3\rangle\rvert$	585.4
$\lvert\langle S_1 \rvert H_{SOC} \rvert T_4\rangle\rvert$	615.6
$\lvert\langle S_1 \rvert H_{SOC} \rvert T_5\rangle\rvert$	16.7
$\lvert\langle S_1 \rvert H_{SOC} \rvert T_6\rangle\rvert$	185.0

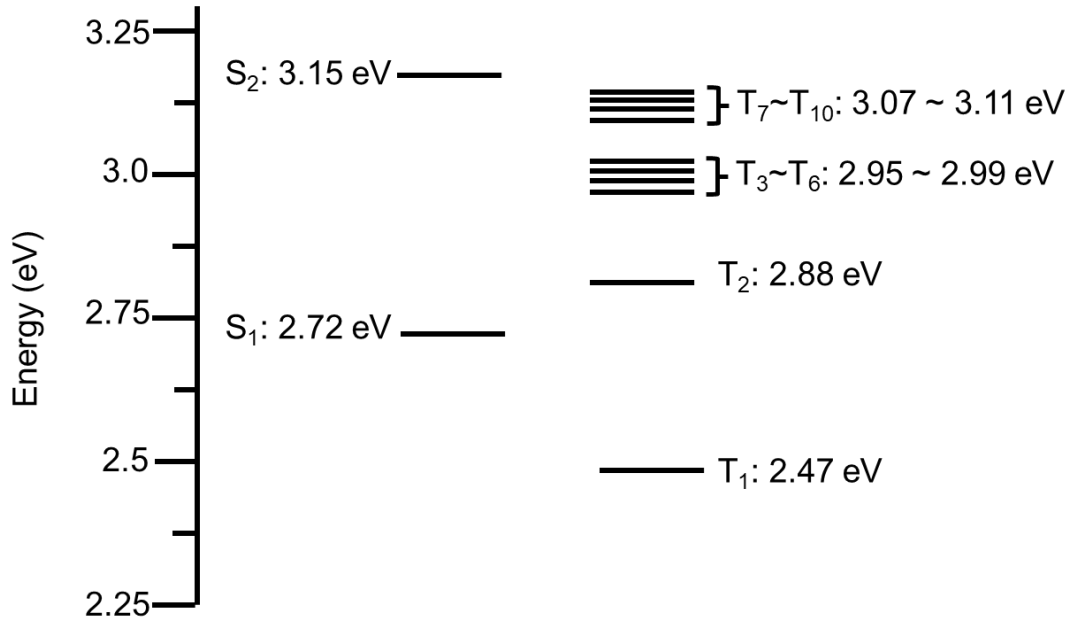

Figure S7 | Schematic of calculated state energy alignment of the tetrameric **4H** at the S_0 equilibrium geometry.

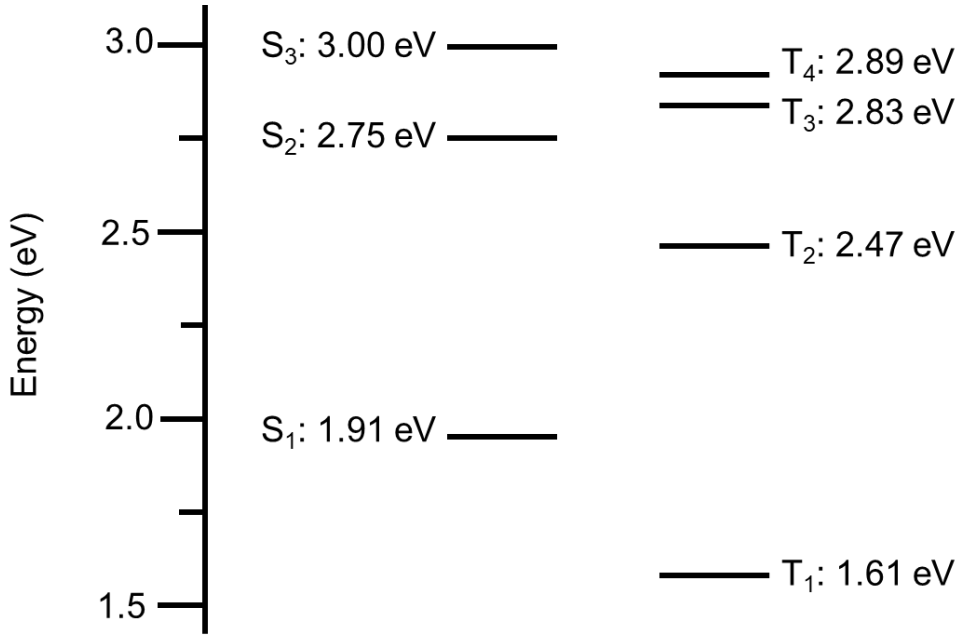

Figure S8 | Schematic of calculated state energy alignment of the tetrameric **4H** at the S_1 equilibrium geometry.

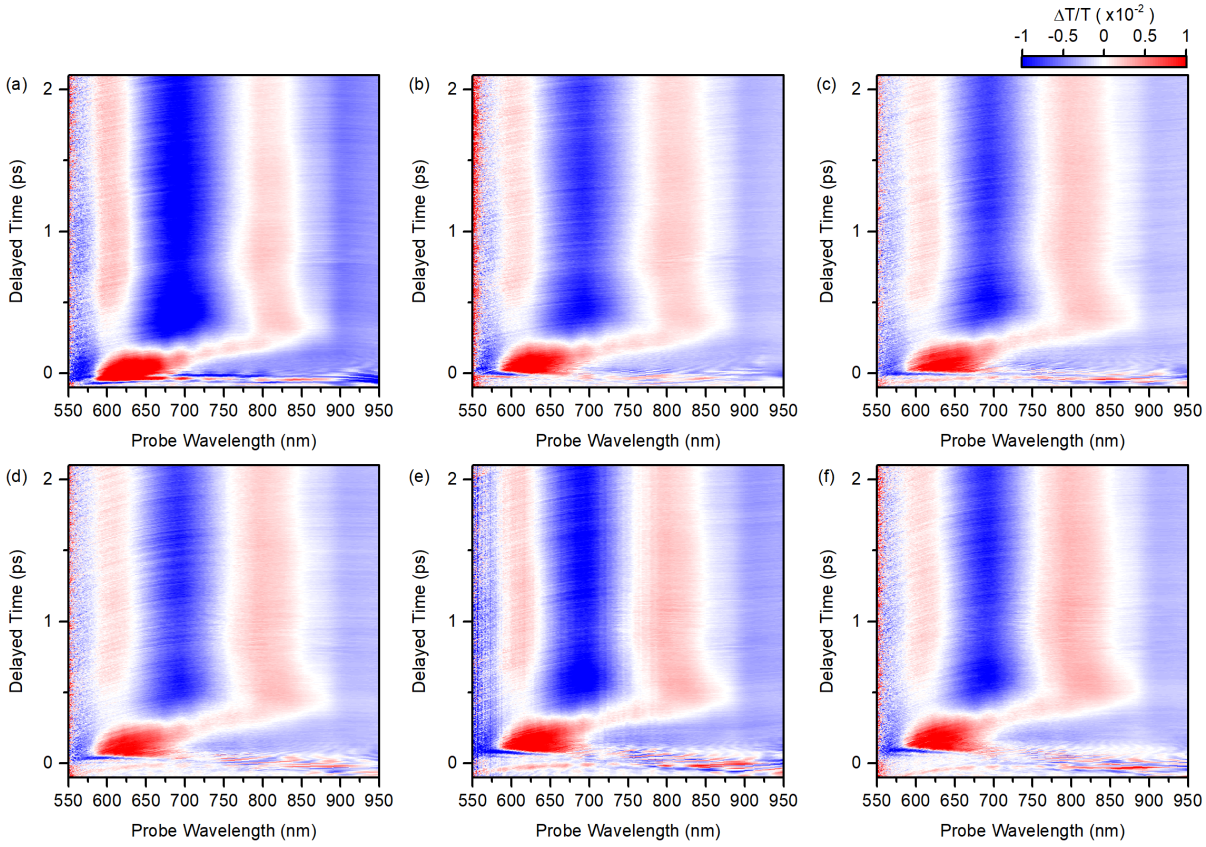

Figure S9 | Pump pulse duration-dependent TA spectra at room temperature. Pulse duration: (a) 20 fs, (b) 40 fs, (c) 60 fs, (d) 80 fs, (e) 100 fs and (f) 115 fs. The spectra and the temporal phases of the pumping pulses are shown in Fig. S3.

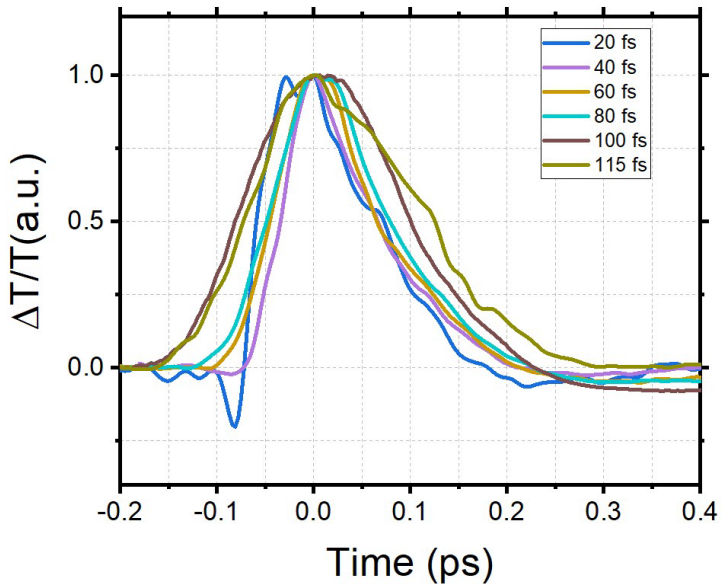

Figure S10 | Time traces obtained by sampling the pulse duration-dependent TA spectra at 615 nm.

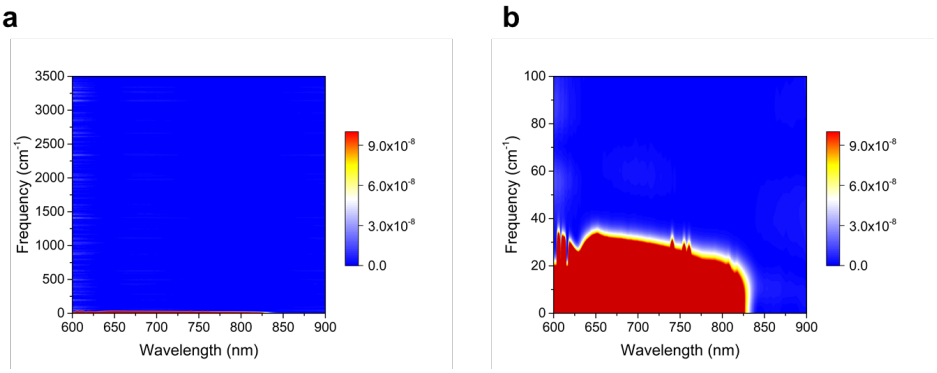

Figure S11 | Wavelength-resolved fast Fourier transform power spectra. The time window is set as 0.1~4.5 ps. (a) Frequency ranges from 10 ~ 3500 cm^{-1}. (b) Frequency ranges from 10 ~ 100 cm^{-1}.

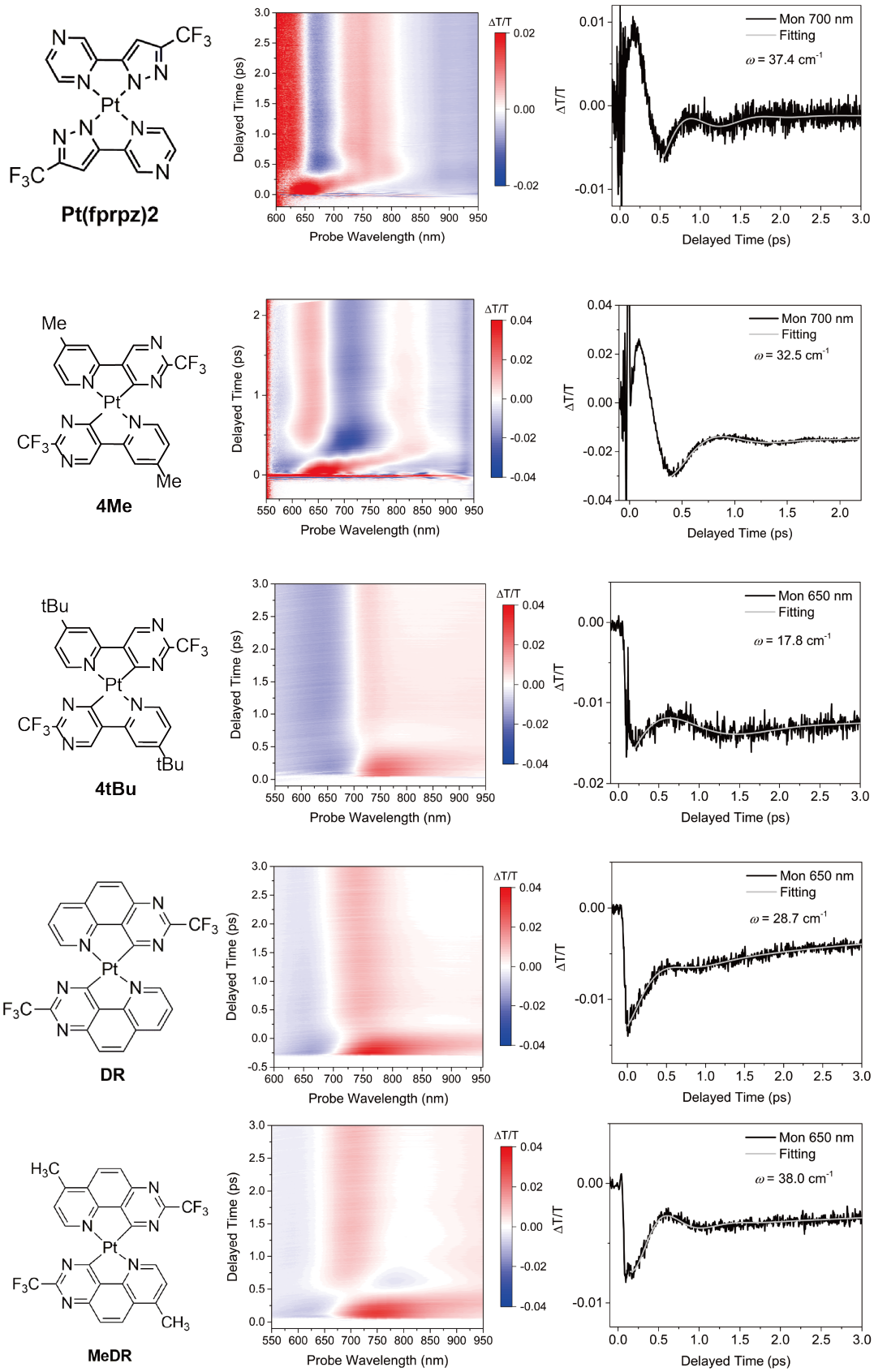

Figure S12 | TA spectra of different aggregate Pt complexes in thin film with 3.2 fs pumping pulse at room temperature.

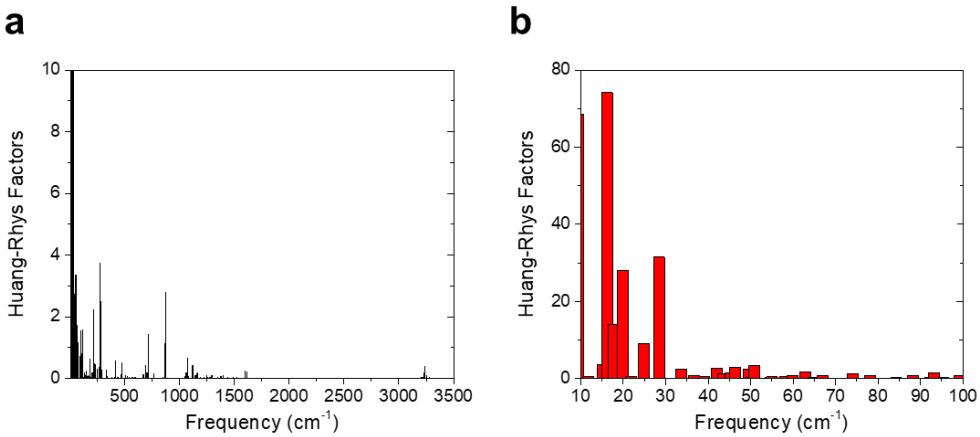

Figure S13 | Calculated Huang-Rhys factors between S_0 and S_1 of the tetrameric **4H**. (a) Frequency ranges from 10 ~ 3500 cm^{-1}. (b) Frequency ranges from 10 ~ 100 cm^{-1}.

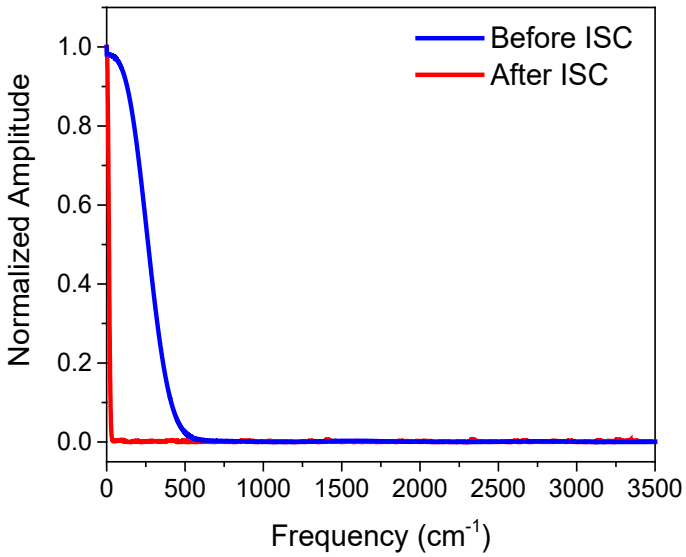

Figure S14 | Fast Fourier transform spectra under different time windows. The time windows of "Before ISC" and "After ISC" corresponds to [0 ps, 0.25 ps] and [0.2 ps, 4.5 ps], respectively. The integrated spectral range is 610 nm to 940 nm.