(*Supporting Information*)

Dependence of Hydration Status of Bacterial Light-Harvesting Complex 2 on Polyol Cosolvents

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S1. Kinetics traces at representative probing wavelengths

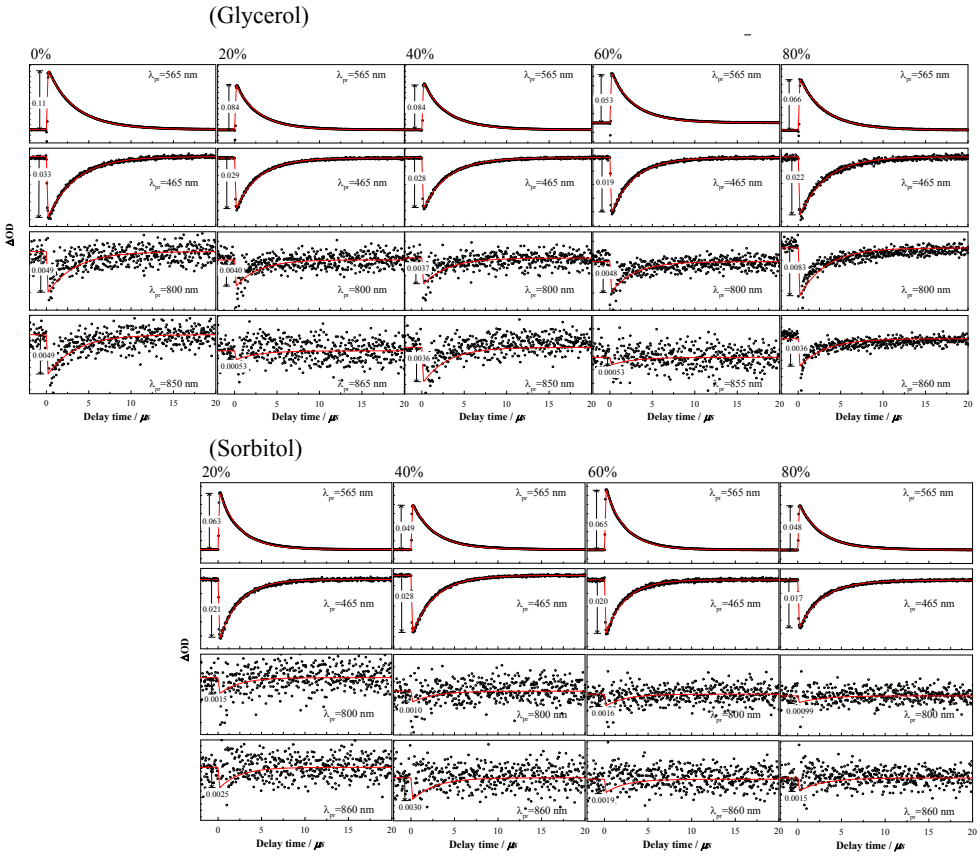


Figure S1. Kinetics traces recorded at indicated probing wavelengths (λ_{pr}) for *Tch. tepidum* LH2 complexes in buffer solution (20 mM Tris·HCl, 0.05% DDM, pH=7.8) with the indicated volume fractions of glycerol and sorbitol. Excitation wavelength was 532 nm (1.4 mJ/pulse). In each case, red solid curves are obtained by global fitting the four kinetics traces to a mono-exponential decay function. In each case, we have also tried to simultaneously fit two kinetics probed at 465 nm and 565 nm to a mono-exponential model function, and achieved satisfactory fitting goodness (fitting curves not shown). As listed below, the decay time constants derived from the 4-curve or the 2-curve global fitting are similar within the error statistics.

Table S1. The triplet excited state lifetime (τ_T) of ^{3}Crt* obtained by global fitting of the two kinetics traces of the BLC and ESA of Crts or four kinetics traces (including the BLC of B800 and B850).

Volume fraction (v/v)	Glycerol		Sorbitol	
	τ_T (μs) by 4 curves	τ_T (μs) by 2 curves	τ_T (μs) by 4 curves	τ_T (μs) by 2 curves
0%	3.34±0.01	3.35±0.01	3.34±0.01	3.35±0.01
20%	2.73±0.01	2.74±0.01	2.35±0.01	2.38±0.00
40%	2.93±0.01	2.95±0.01	2.67±0.02	2.70±0.00
60%	2.75±0.01	2.77±0.01	2.28±0.01	2.29±0.00
80%	3.13±0.01	3.13±0.01	2.72±0.01	2.74±0.00

S2. Spectral dynamics at different polyol volume fractions

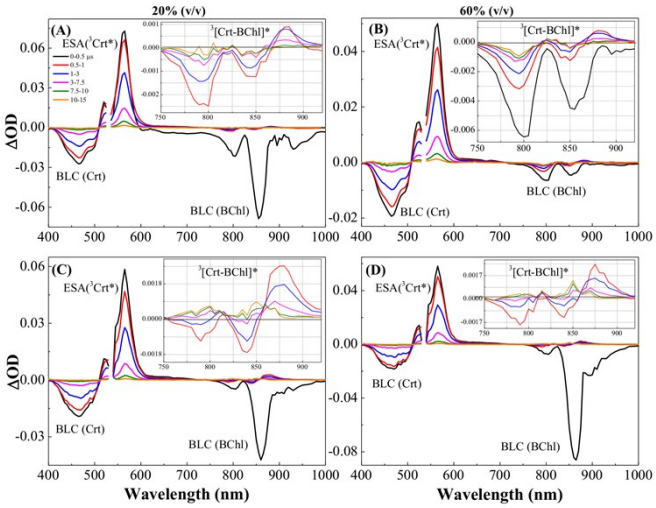
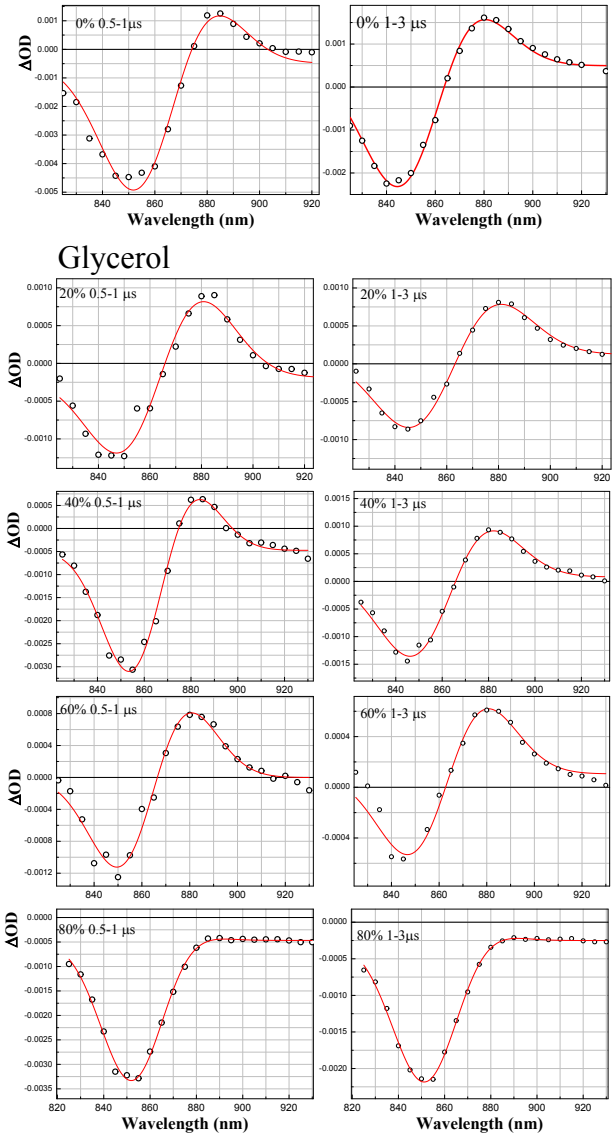


Figure S2. Transient spectra for *Tch. tepidum* LH2 complexes in buffer solution (20 mM Tris·HCl, 0.05% DDM, pH=7.8) with the indicated volume fractions of (**A**, **B**) glycerol and (**C**, **D**) sorbitol. Insets show the blowup spectra in near-infrared spectral region. Excitation wavelength was 532 nm (1.4 mJ/pulse).

S3. Spectral simulation of ^{3}Crt*-B8500 interaction bands



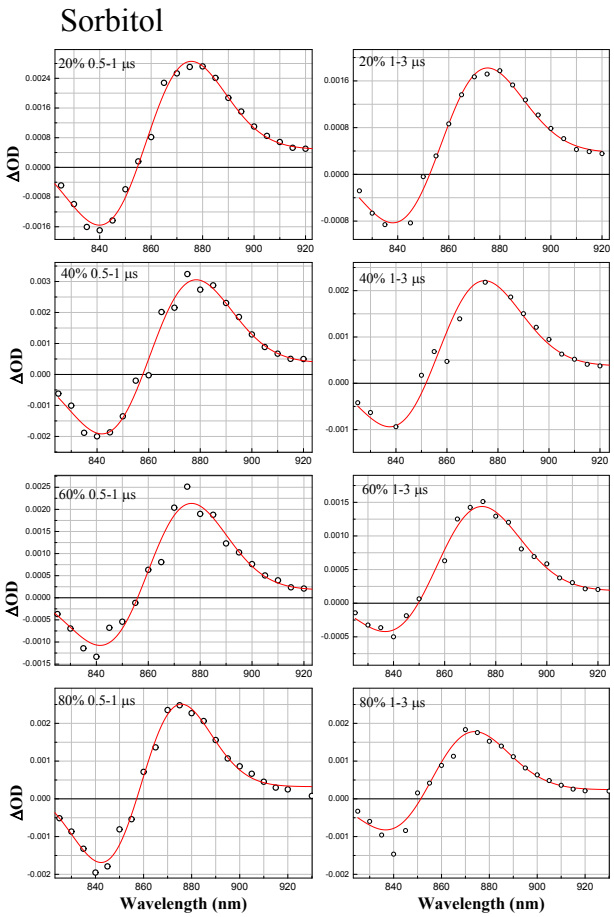


Figure S3. The derivative-type ^{3}Crt*-BChl interaction bands of the transient spectra in 825–925 nm at 0.5–1μs and 1–3 μs of 0% sample (top three), glycerol samples (20-80% v/v) and sorbitol samples (20-80% v/v) were simulated by subtracting the B850 Q_y absorption spectrum $OD_{B850}(\lambda)$ from a red shifted ground-state absorption spectrum $OD_{B850}(\lambda+\delta\lambda)$, i.e. $\Delta OD_{obs}(\lambda) = A_1 \cdot OD_{B850}(\lambda+\delta\lambda) - A_2 \cdot OD_{B850}(\lambda)$, where $\delta\lambda$ represents the amount of spectral shift, and A_1 and A_2 are amplitude factors. The B850 Q_y absorption spectrum $OD_{B850}(\lambda)$ was obtained by spectral decomposition from the steady-state Q_y absorption spectra. The amplitude ratio A_1/A_2 can be used as a metric of the relative contribution of the first term representing the temporal Stark effect induced band shifts of the ground-state B850 Q_y absorption.

Table S2. Temporal Stark shifts ($\delta\lambda$) derived by least-square spectral fitting the interaction bands at 0.5–1 μs and 1–3 μs regimes (Figure S3) to equation $\Delta OD_{obs}(\lambda) = A_1 \cdot OD_{B850}(\lambda+\delta\lambda) - A_2 \cdot OD_{B850}(\lambda)$. The amplitude ratio A_1/A_2 indicates the relative contribution of the temporal Stark shift to the observed interaction bands.

Samples	**0.5-1 μs**		**1-3 μs**	
	A_1/A_2	**δλ (nm)**	A_1/A_2	**δλ (nm)**
0%	0.57	18.71	0.83	7.90
		Glycerol		
20%	1.00	15.90	0.89	11.22
40%	0.53	21.39	0.92	11.63
60%	0.90	16.32	0.88	13.90
80%	0.59	3.67	0.64	3.54
		Sorbitol		
20%	1.02	4.82	1.03	3.05
40%	1.03	9.68	1.03	3.10
60%	1.16	9.78	1.24	10.65
80%	1.03	6.77	1.01	2.14

S4. Comparison of amino acid sequences of LH2 α,β-apoproteins

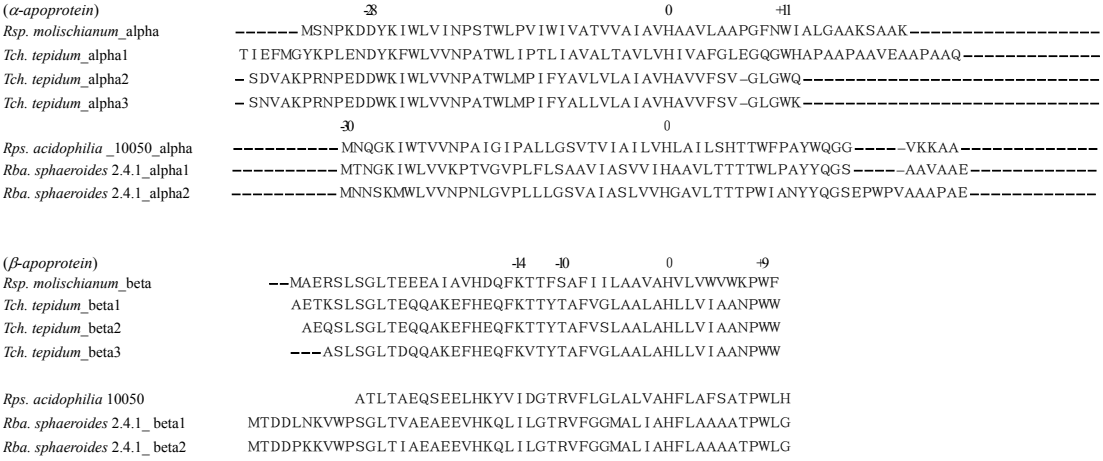


Figure S4. Comparison of the amino acid sequences of LH2 α,β-apoproteins of *Rs. molischianum*, *Rps. acidophila*, *Tch. tepidum* and *Rba. sphaeroides* 2.4.1.[S1,S2,S3]

References

[S1] Sekine, F.; Horiguchi, K.; Kashino, Y.; Shimizu, Y.; Yu, L. J.; Kobayashi, M.; Wang, Z. Y., Gene sequencing and characterization of the light-harvesting complex 2 from thermophilic purple sulfur bacterium *Thermochromatium tepidum*. *Photosynth. Res.* **2012**, 111 (1-2), 9-18.

[S2] Koepke, J.; Hu, X.; Muenke, C.; Schulten, K.; Michel, H., The crystal structure of the light-harvesting complex II (B800-850) from *Rhodospirillum molischianum*. *Structure* **1996**, 4 (5), 581-597.

[S3] Zeng, X.; Choudhary, M.; Kaplan, S., A second and unusual pucBA operon of *Rhodobacter sphaeroides* 2.4.1: genetics and function of the encoded polypeptides. *J. Bacteriol.* **2003**, 185 (20), 6171-6184.