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(Supporting Information)

Dependence of Hydration Status of Bacterial

Light-Harvesting Complex 2 on Polyol Cosolvents

 Ying Shi,[†] Jie Yu,[†] Long-Jiang Yu,^{‡,§} Peng Wang,[†] Li-Min Fu,[†] Jian-Ping Zhang,^{†,*} and Zheng-Yu Wang-Otomo^{‡,*}
 [†]Department of Chemistry, Renmin University of China, Beijing 1000872, P. R. China
 [‡]Faculty of Science, Ibaraki University, Mito 310-8512, Japan
 [§]Present address: Department of Biology, Faculty of Science, Okayama University, Okayama, Japan

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 ³Crt* 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).

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Volume	Volume Glycerol		Sorbitol	
fraction (v/v)	$ au_{ m T}$ (µs) by 4 curves	$ au_{\mathrm{T}}$ (µs) by 2 curves	$ au_{ m T}$ (µs) by 4 curves	$ au_{\mathrm{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	$2.74{\pm}0.01$	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 (\mathbf{A} , \mathbf{B}) glycerol and (\mathbf{C} , \mathbf{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 ³Crt*-B8500 interaction bands





Figure S3. The derivative-type ³Crt*-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

$(\alpha$ -apoprotein)	-28	0	+11		
Rsp. molischianum_alpha	MSNPKDDYKIWLVINPSTWLPVIWIVAIAVHAAVLAAPGFNWIALGAAKSAAK				
Tch. tepidum_alpha1	T I EFMGYKPLENDYKFWLVVNPATWL I PTL I AVALTAVLVH I VAFGLEGQGWHAPAAPAAVEAAPAAQ				
Tch. tepidum_alpha2	– SDVAKPRNPEDDWKIWLVVNPATWLMPIFYAVLVLAIAVHAVVFSV-GLGWQ				
Tch. tepidum_alpha3	– SNVAKPRNPEDDWKIWLVVNPATWLMPIFYALLVLAIAVHAVVFSV-GLGWK				
	-30	0			
Rps. acidophilia _10050_alpha	MNQGKIWTVVNPAIGIPALLGSVTVIAILVHLAILSHTTWFPAYWQGGVKKAA				
Rba. sphaeroides 2.4.1_alpha1	MTNGKIWLVVKPTVGVPLFLSAAVIASVVIHAAVLTTTTWLPAYYQGSAAVAAE				
Rba. sphaeroides 2.4.1_alpha2	MNNSKMWLVVNPNLGVPLLLGSVAIASLVVHGAVLTTTPWIANYYQGSEPWPVAAAPAE				
$(\beta$ -apoprotein)	-14	-10 0	+9		
Rsp. molischianum_beta	——MAERSLSGLTEEEAIAVHDQFKTTFSAFIILAAVAHVLVWVWKPWF				
Tch. tepidum_beta1	AETKSLSGLTEQQAKEFHEQFKTTYTAFVGLAALAHLLVIAANPWW				
Tch. tepidum_beta2	AEQSLSGLTEQQAKEFHEQFKTTYTAFVSLAALAHLLVIAANPWW				
Tch. tepidum_beta3	ASLSGLTDQQAKEFHEQFKVTYTAFVGLAALAHLLVIAANPWW				
Rps. acidophilia 10050	ATLTAEQSEELHKYVI	DGTRVFLGLALVAHFLAF	SATPWLH		
Rba. sphaeroides 2.4.1_beta1	MTDDLNKVWPSGLTVAEAEEVHKQLILGTRVFGGMALIAHFLAAAATPWLG				
Rba. sphaeroides 2.4.1_beta2	MTDDPKKVWPSGLT I AEAEEVHKQL I LGTRVFGGMAL I AHFLAAAATPWLG				

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]

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