Supporting Information

Harrop et al. 10.1073/pnas.1402538111

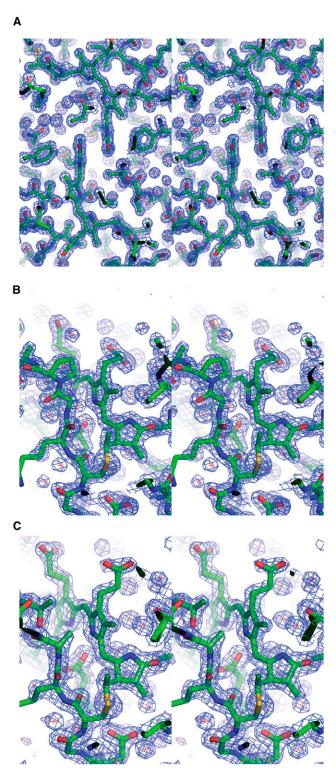


Fig. S1. Electron density. Stereograms showing sample electron density from the three crystal structures. (*A*) Electron density of PC645 centered on the offset-stacked β 50/61 chromophores (contoured at 2 σ). (*B*) Electron density of PC612 centered on the chromophore in the α_1 subunit (contoured at 2 σ). (*C*) Electron density of PE555 centered on the chromophore in the α_2 subunit (contoured at 1.5 σ). The view is similar to that in *B*.

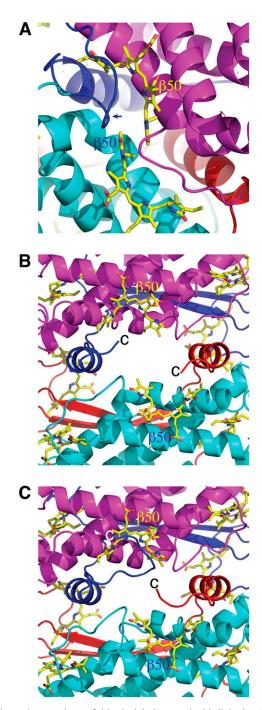


Fig. S2. Central β 50/61 chromophores viewed down the pseudo-twofold axis. (A) The two doubly linked β 50/61 DBV chromophores in the closed-form PC645. The blue arrow indicates the extended C-terminal loop of the α_1 subunit. (B and C) View down the pseudo-twofold axis showing the separation between the doubly linked β 50/61 chromophores in the open-form structures of PC612 (B) and PE555 (C).

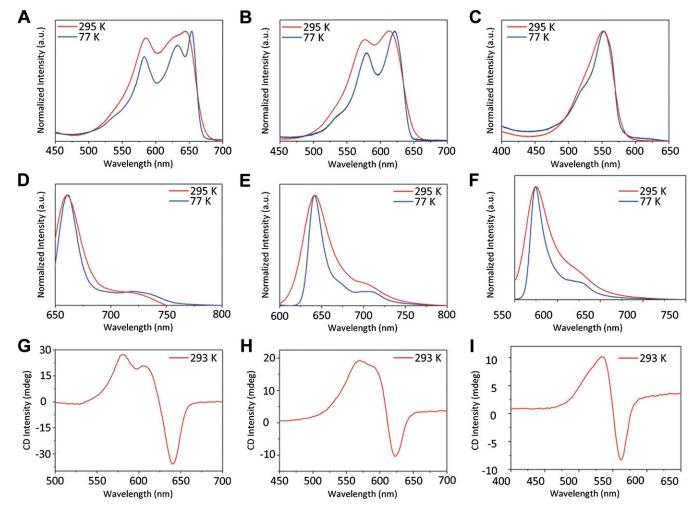


Fig. S3. Spectroscopy of cryptophyte PBPs. The figure shows the absorbance spectra of PC645 (A), PC612 (B), and PE555 (C) at 77 K (blue traces) and 295 K (red traces); the fluorescence spectra of PC645 (D), PC612 (E), and PE555 (F) at 77 K (blue traces) and 295 K (red traces), and the visible circular dichroism spectra of PC645 (G), PC612 (H), and PE555 (I) at 293 K (red traces).

Table S1. Data collection and refinement statistics

	PC645	PE555	PC612	
Data collection				
Source (λ)	SSRL 9.2 (0.82106 Å)	APS 23ID-D (0.97934 Å)	AS MX1 (0.9537 Å)	
Detector	MarCCD	MarCCD	ADSC Q210	
Space group	P2 ₁ 2 ₁ 2 ₁	P2 ₁	<i>P</i> 6 ₃	
Cell dimensions				
a, b, c, Å	44.96, 93.82,121.91	66.07, 76.74, 142.59	132.08, 132.08, 64.36	
α, β, γ, °	90, 90, 90	90, 92.86, 90	90, 90, 120	
Resolution, Å	1.35 (1.42–1.35)*	1.80 (1.90–1.80)*	1.7 (1.79–1.7)*	
R_{merge}	0.095 (0.408)	0.079 (0.488)	0.074 (0.755)	
l/σ	12.3 (2.5)	11.0 (2.9)	24.1 (3.3)	
Completeness, %	95.2 (74.6)	99.6 (99.9)	99.7 (98.0)	
Redundancy	6.6 (2.8)	2.6 (2.6)	11.3 (11.2)	
B _{overall} , Å ²	15.0	15.2	19.5	
Refinement				
Resolution, Å	1.35 (1.42–1.35)	1.80 (1.90–1.80)	1.7 (1.79–1.7)	
No. reflections (unique)	725,477 (109,224)	339,657 (131,394)	796,218 (70,253)	
R_{work}/R_{free}	0.143/0.181	0.158/0.214	0.167/0.197	
No. atoms				
Protein	3,842	11,813	3,874	
Chromophore	344	1032	344	
Water	764	2195	673	
Rmsd [†]				
Bond lengths, Å	0.010	0.006	0.012	
Bond angles, °	1.33	1.08	1.67	
Ramachandran [†]				
Favored, %	97.8	99.2	98.13	
Outliers, %	0.0	0.0	0.42	

^{*}Values in parentheses are for highest-resolution shell.

Table S2. Transformation between open and closed form

	PE545	PC645	
PE555	73°	74.9°	
PC612	70.5°	73.5°	

Relative rotation between the two $\alpha\beta$ monomers required to map the open-form structures onto the closed-form structures.

Table S3. Rotation of pyrrole A in α chromophores

	α Chain	Dihedral angle, °
PE545	1	+32.2
	2	+31.2
PC645	1	+28.8
	2	+23.2
PE555	1	-42.5
	2	-39.9
PC612	Chain A	-40.1
	Chain C	-38.3

Dihedral angles measured in PyMoI from pyrrole B using atoms: C2B-C1B-CHA-C4A.

 $R_{\text{merge}} = \Sigma_{\text{hkl}} \Sigma_{\text{i}} | I_{\text{i}} < I > | I \Sigma_{\text{hkl}} \Sigma_{\text{i}} I$

R-factor = $_{hkl}||F_{obs}|-|F_{calc}||/\Sigma_{hkl}|F_{obs}|$

[†]Calculated by PHENIX (1).

^{1.} Adams PD, et al. (2002) PHENIX: Building new software for automated crystallographic structure determination. Acta Crystallogr D Biol Crystallogr 58(Pt 11):1948–1954.

Table S4. Matrix of Coulombic gas-phase couplings (cm⁻¹) calculated at the CIS/cc-pvtz level

Protein				Electro	nic coupling	matrix			
PC645		MBV _{α18}	PCB _{β158}	PCB _{β82}	DBV _{β50/61}	MBV _{α18}	PCB _{β158}	PCB _{β82}	DBV _{β50/61}
	$MBV_{\alpha 18}$	_	151	26	19	-7	5	69	-74
	$PCB_{\beta 158}$	151	_	19	22	4	13	-19	37
	$PCB_{\beta 82}$	26	19	_	-64	75	-19	-12	35
	$DBV_{\beta 50/61}$	19	22	-64	_	-76	45	32	647
	$MBV_{\alpha 18}$	-7	4	75	-76	_	160	24	14
	$PCB_{\beta 158}$	5	13	-19	45	160	_	18	32
	$PCB_{\beta 82}$	69	-19	-12	32	24	18	_	-67
	DBV _{β50/61}	-74	37	35	647	14	32	-67	_
PC612									
		$PCB_{\alpha 20}$	$PCB_{\beta 158}$	$PCB_{\beta 82}$	$DBV_{\beta 50/61}$	$PCB_{\alpha 20}$	$PCB_{\beta 158}$	$PCB_{\beta 82}$	$DBV_{\beta50/61}$
	$PCB_{\alpha 20}$	_	146	20	29	-2	-2	40	_ 5
	$PCB_{\beta 158}$	146	_	22	22	-2	8	3	-5
	PCB _{β82}	20	22	_	-67	45	4	2	27
	DBV _{β50/61}	29	22	-67	_	-5	-4	24	29
	$PCB_{\alpha 20}$	-2	-2	45	-5	_	130	19	28
	$PCB_{\beta 158}$	-2	8	4	-4	130	_	19	23
	$PCB_{\beta 82}$	40	3	2	24	19	19	_	-61
	DBV _{β50/61}	-5	-5	27	29	28	23	-61	_
PE555		$\text{PEB}_{\alpha 20}$	$PEB_{\beta 158}$	$PEB_{\beta 82}$	$DBV_{\beta 50/61}$	$PEB_{\alpha20}$	$PEB_{\beta 158}$	$PEB_{\beta 82}$	DBV _{β50/61}
	$PEB_{\alpha 20}$	0	68	17	20	-13	5	26	-8
	$PEB_{\beta 158}$	68	0	12	36	5	5	4	-2
	PEB _{β82}	17	12	0	-60	29	3	4	20
	$DBV_{\beta 50/61}$	20	36	-60	0	-11	1	24	4
	$PEB_{\alpha 20}$	-13	5	29	0	0	63	16	13
	$PEB_{\beta 158}$	5	5	3	1	63	0	12	43
	PEB _{β82}	26	4	4	24	16	12	0	-56
	DBV _{β50/61}	-8	-2	20	4	13	43	-56	0
PE545	•								
		$DBV_{\alpha20}$	$PEB_{\beta 158}$	$PEB_{\beta82}$	PEB _{β50/61}	$DBV_{\alpha20}$	$PEB_{\beta 158}$	$\text{PEB}_{\beta82}$	PEB _{β50/61}
	$DBV_{\alpha 20}$	_	51	17	4	-6	5	62	-64
	$PEB_{\beta 158}$	51	_	11	29	5	9	-11	31
	PEB _{β82}	17	11	_	-56	62	-8	-4	29
	PEB _{β50/61}	4	29	-56	_	-61	25	27	166
	$DBV_{\alpha 20}$	-6	5	62	-61	_	49	18	4
	PEB _{β158}	5	9	-8	25	49	_	12	38
	PEB _{β82}	62	-11	-4	27	18	12	_	-58
	PEB _{β50/61}	-64	31	29	166	4	38	-58	_

Table S5. Transition dipole components d_x , d_y , and d_z and center coordinates R_x , R_y , and R for pigments

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Protein	Pigment	d_{x} , eÅ	d _y , eÅ	d _z , eÅ	R _x , Å	R _y , Å	R _z , Å
PC645							
	$MBV_{\alpha 18}$	0.355	-0.971	2.639	-2.385	-16.224	21.593
	$PCB_{\beta 158}$	2.769	0.397	0.273	14.271	-16.245	13.913
	$PCB_{\beta 82}$	1.370	0.969	2.324	4.985	16.483	19.316
	$DBV_{\beta50/61}$	-1.100	-2.436	0.406	20.417	2.139	27.442
	$MBV_{\alpha 18}$	0.493	0.537	-2.806	17.655	20.716	41.491
	$PCB_{\beta 158}$	1.249	-2.403	-0.690	28.495	6.643	46.518
	$PCB_{\beta 82}$	-0.266	-1.380	-2.463	-3.338	-5.770	45.331
	DBV _{β50/61}	1.215	2.395	-0.056	15.759	-8.186	34.154
PC612							
	$PCB_{\alpha 20}$	-0.751	0.831	2.490	-28.807	22.987	-10.413
	$PCB_{\beta 158}$	-2.123	1.647	-0.375	-39.122	32.954	-21.662
	PCB _{β82}	-2.235	0.677	1.611	-55.940	4.719	-12.822
	DBV _{β50/61}	2.383	1.082	0.605	-58.468	26.870	-9.707
	$PCB_{\alpha 20}$	-1.202	0.954	-2.219	-46.612	2.767	4.613
	$PCB_{\beta 158}$	-1.622	1.963	0.770	-55.849	11.273	17.882
	PCB _{β82}	-0.822	2.177	-1.222	-28.843	30.552	8.051
	DBV _{β50/61}	-1.076	-2.359	-0.701	-51.487	32.116	7.702
PE555							
	$PEB_{\alpha 20}$	-2.076	-1.058	0.290	-14.189	3.580	-22.663
	PEB _{β158}	0.549	-2.213	0.463	-4.787	-10.576	-31.910
	PEB _{β82}	-0.893	-2.006	0.653	6.047	-6.733	1.576
	DBV _{β50/61}	-1.663	1.222	-1.587	-4.487	-21.093	-11.595
	$PEB_{\alpha 20}$	1.927	-1.241	-0.443	-10.132	1.268	5.540
	PEB _{β158}	-0.889	-1.980	-0.773	-21.807	-12.892	12.536
	PEB _{β82}	0.565	-1.973	-0.918	-31.293	-2.617	-19.992
	$DBV_{\beta50/61}$	1.504	0.781	1.727	-24.236	-20.388	-9.142
	$PEB_{\alpha 20}$	-2.076	-1.058	0.290	-14.189	3.580	-22.663
PE545							
	$DBV_{\alpha 20}$	-2.072	-1.349	0.112	5.183	22.303	6.092
	PEB _{β158}	0.268	-0.407	2.292	17.334	12.234	19.256
	PEB _{β82}	-1.393	-0.682	1.763	11.152	45.874	29.776
	PEB _{β50/61}	-0.149	-1.210	-2.010	6.689	25.648	36.445
	$DBV_{\alpha20}$	2.369	0.068	0.785	-7.093	43.092	43.939
	PEB _{β158}	0.645	-2.121	-0.859	-11.291	24.058	48.778
	PEB _{β82}	1.918	-1.221	-0.580	-15.635	29.755	13.951

Table S6. Experimental conditions adopted for the three protein samples

Sample	λ_{exc} , THz	Bandwidth, THz	Pulse duration, fs
PC645	508	61	11
PC612	525	55	14
PE555	545	55	14