Electronic Supplementary Material (ESI) for Chemical Science.


# Supporting Information: Thermal site energy fluctuations in photosystem I: New insights from MD/QM/MM calculations

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# Contents

# 1 Details on Structure Preparation

Table S1 lists all amino acids that were missing in the crystal structure of cyanobacterial PSI and were added manually with the *Modeller* interface[S1] to *UCSF Chimera*.[S2] The residue ID refers to the original numbering in the crystal structure. Partially resolved phytyl chains of the chlorophyll residues were replaced with methyl groups and the affected residues renamed from CLA to CLX, to distinguish them later in the MD simulations. Other missing side chains were replaced manually. Figure S1 illustrates all modifications on the chlorophyll residues in the crystal structure.

Table S1: Amino acids added to the crystal structure of 1JB0. Residue IDs are given according to the original numbering of the pdb file.

| **chain** | **res. ID** | **res. name** |
|---|---|---|
| A | 1 to 12 | MET, THR, ILE, SER, PRO, PRO, GLU, ARG, GLU, PRO, LYS, VAL |
| | 263 to 265 | GLY, VAL, ILE |
| B | 740 | GLY |
| E | 70 to 75 | PRO, PRO, LYS, LYS, GLY, LYS |
| F | -22 to 0 | MET, ARG, ARG, PHE, LEU, ALA, LEU, LEU, LEU, VAL, LEU, THR, LEU, TRP, LEU, GLY, PHE, THR, PRO, LEU, ALA, SER, ALA |
| K | 1 to 19 | MET, VAL, LEU, ALA, THR, LEU, PRO, ASP, THR, THR, TRP, THR, PRO, SER, VAL, GLY, LEU, VAL, VAL |
| | 33 | ALA |
| | 44 to 54 | PRO, ILE, ALA, LEU, PRO, ALA, LEU, PHE, GLU, GLY, PHE |
| | 78 to 83 | GLN, TYR, ALA, GLY, ALA, LEU |
| L | 1 to 3 | ALA, GLU, GLU |
| X | 1 to 6 | ALA, THR, LYS, SER, ALA, LYS |

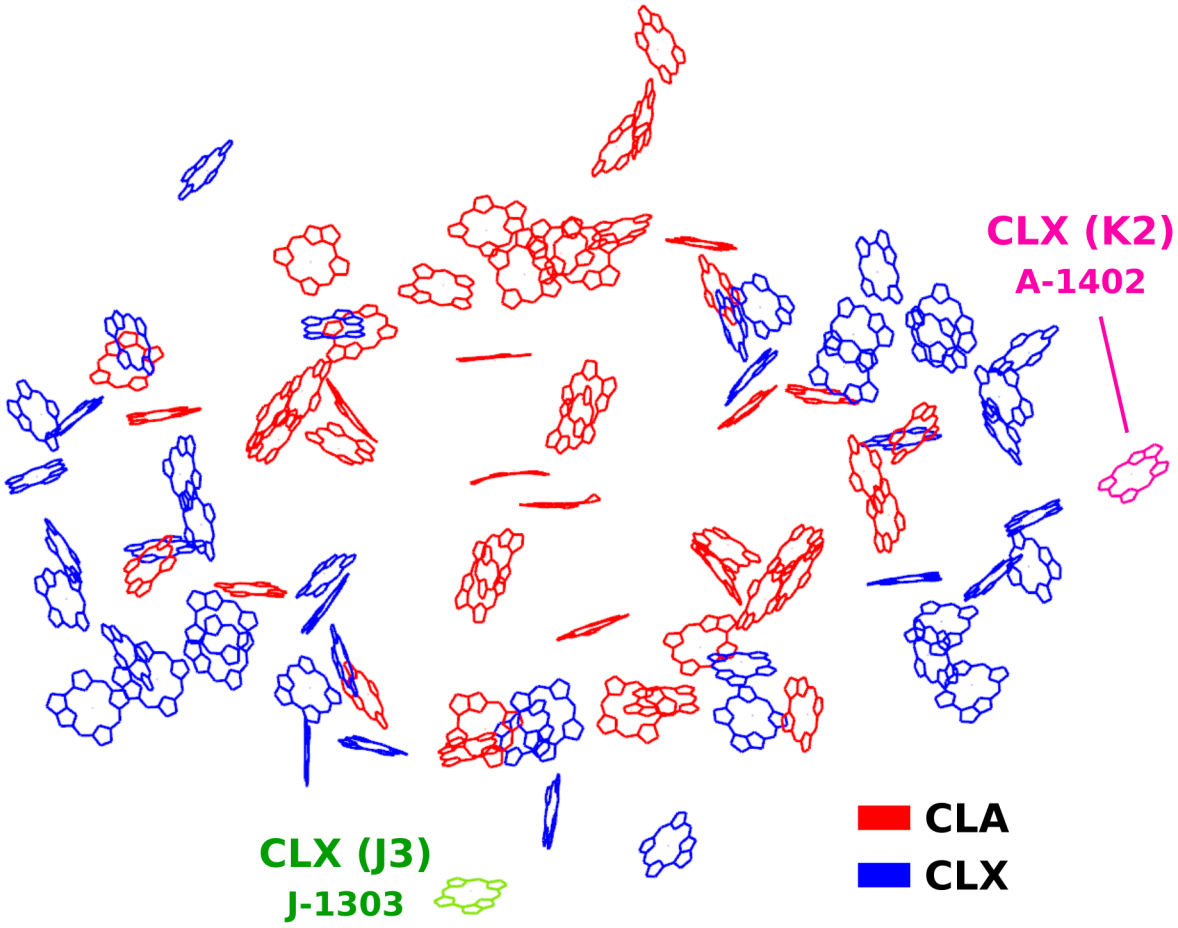


Figure S1: Chlorophyll *a* residue types after modification, as used in the MD simulations. IDs in parentheses refer to the standard chlorophyll numbering, the other notation indicates the chain and residue given in the crystal structure PDB (1JB0).[S3] Red: Intact chlorophylls, blue: phytyl chain replaced with $-CH_3$, magenta: $-COOCH_3$ and $-CH{=}CH_2COOCH_3$ added to residue J1303 and phytyl chain replaced with $-CH_3$, green: $-COOCH_3$ added to residue A1402 and phytyl chain replaced with $-CH_3$.

## 2 Molecular Dynamics Validation

Proper equilibration of the MD simulations was confirmed by checking the convergence of temperature ($T$), total energy ($E$), pressure ($p$), density ($\rho$) and the box vectors (fig. S2). Average values of $T$, $p$, $\rho$ and $E$ are provided in tab. S2.

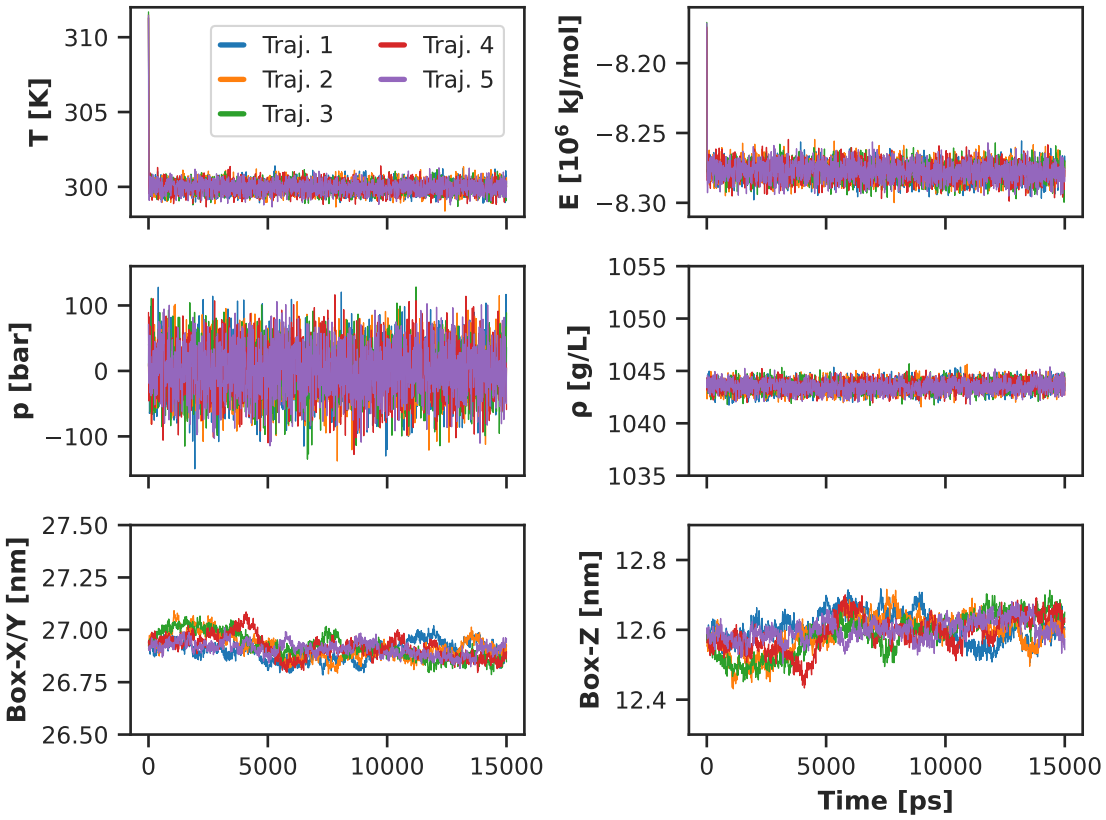


Figure S2: Key thermodynamic parameters during MD trajectories.

Table S2: Thermodynamic averages and standard deviations during the MD simulations.

| Traj. | $\langle T \rangle$ [K] | $\langle p \rangle$ [bar] | $\langle \rho \rangle$ [g L$^{-1}$] | $\langle E \rangle$ [kJ mol$^{-1}$] |
|---|---|---|---|---|
| 1 | 300.0 ± 0.5 | 1 ± 39 | 1043.5 ± 0.6 | (-8.278 ± 0.007)×10$^6$ |
| 2 | 300.0 ± 0.5 | 1 ± 37 | 1043.5 ± 0.6 | (-8.277 ± 0.007)×10$^6$ |
| 3 | 300.0 ± 0.5 | 1 ± 38 | 1043.6 ± 0.6 | (-8.277 ± 0.007)×10$^6$ |
| 4 | 300.0 ± 0.5 | 1 ± 38 | 1043.6 ± 0.6 | (-8.277 ± 0.007)×10$^6$ |
| 5 | 300.0 ± 0.5 | 1 ± 37 | 1043.5 ± 0.6 | (-8.277 ± 0.007)×10$^6$ |

To assess the quality of the trajectories, we evaluated the mass-weighted RMSD of the protein C$\alpha$ backbone, the porphyrin units of the CLA/CLX residues and the BCR and PQN cofactors with respect to the crystal structure. Each frame of the trajectory was aligned to the protein C$\alpha$ atoms of the reference. Hydrogen atoms and all atoms missing in the crystal structure were excluded from the analysis. In all production simulations, the RMSD of the protein backbone as well as that of the cofactors is converged (fig. S3). The time-averaged RMSD (fig. S4) is between 1 Å and 2 Å, which is below the 2.5 Å resolution of the crystal structure, thus validating the sampled geometries.

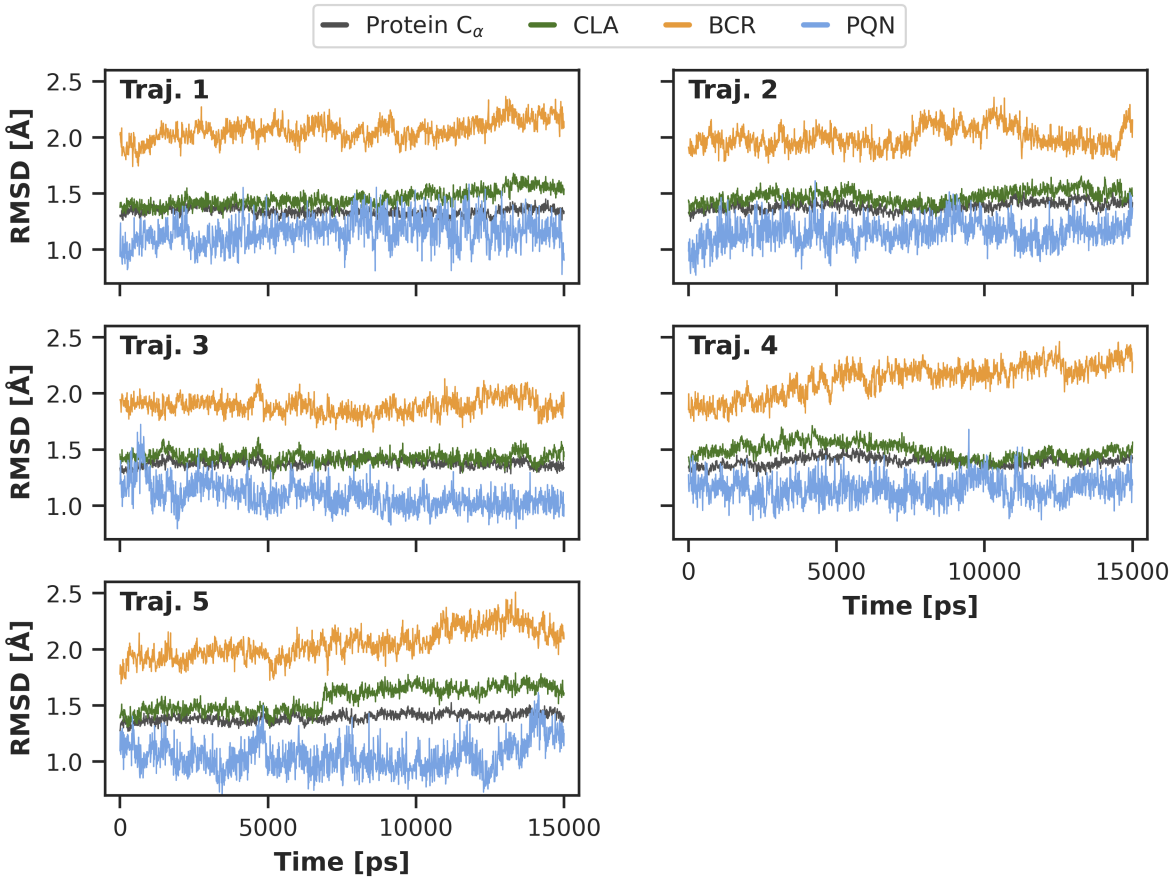


Figure S3: RMSD of different residue groups vs. the crystal structure in each production trajectory over time. CLA: Chlorophyll porphyrin units; BCR: $\beta$-carotene, PQN: phylloquinone.

We also calculated the area per lipid of the POPC membrane (table S3, fig. S5) using the *FATSLiM 0.2.2* toolbox[S4] and applying a cutoff distance of 2.9 nm for leaflet identification.

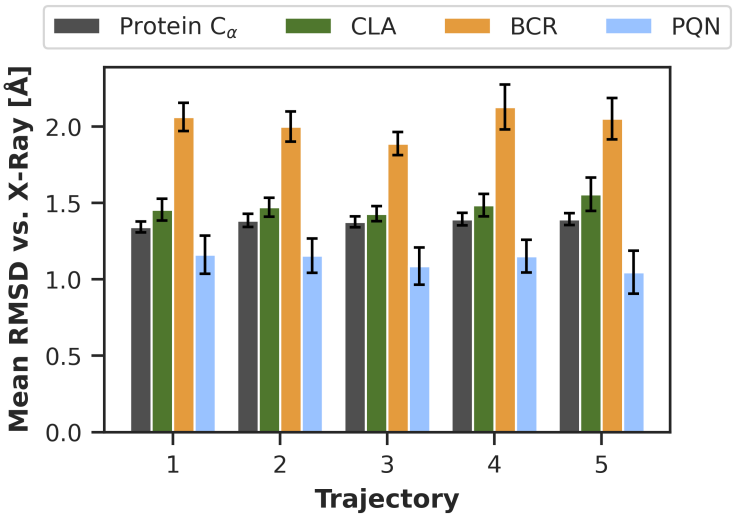


Figure S4: Time-averaged RMSD of different residue groups vs. the crystal structure in each production trajectory. Error bars signify the standard deviation across the trajectory. CLA: Chlorophyll porphyrin units; BCR: $\beta$-carotene, PQN: phylloquinone.

Membrane leaflets were identified by the phosphatidylcholine headgroups, while protein and cofactors were used as interacting groups. The average area per lipid across both membrane leaflets $\langle A \rangle$ is 0.642 nm$^2$, which is in good agreement with previously reported values for POPC bilayers (0.604 nm$^2$,[S5] 0.655 nm$^2$,[S6] 0.631 nm$^2$[S7]). The time-averaged lipid bilayer thickness $\langle D \rangle$ (fig. S6), defined as the distance between the P atom layers in the upper and lower leaflet, converged to 3.67 nm after equilibration, which also corresponds well with previous theoretical (3.725 nm,[S6] 3.75 nm[S7]) and experimental reports (3.7 nm[S8]).

Table S3: Bilayer thickness $\langle D \rangle$ and average area per lipid at 300 K in the full membrane ($\langle A \rangle$), the lower (stromal) leaflet ($\langle A_L \rangle$) and the upper (lumenal) leaflet ($\langle A_U \rangle$).

| Traj. | $\langle A \rangle$ [nm$^2$] | $\langle A_L \rangle$ [nm$^2$] | $\langle A_U \rangle$ [nm$^2$] | $\langle D \rangle$ [nm] |
|---|---|---|---|---|
| 1 | 0.642 ± 0.003 | 0.630 ± 0.005 | 0.655 ± 0.006 | 3.68 ± 0.02 |
| 2 | 0.642 ± 0.005 | 0.634 ± 0.006 | 0.651 ± 0.006 | 3.67 ± 0.02 |
| 3 | 0.642 ± 0.004 | 0.630 ± 0.006 | 0.655 ± 0.014 | 3.67 ± 0.02 |
| 4 | 0.641 ± 0.004 | 0.628 ± 0.006 | 0.654 ± 0.005 | 3.67 ± 0.01 |
| 5 | 0.640 ± 0.004 | 0.630 ± 0.006 | 0.650 ± 0.005 | 3.67 ± 0.01 |

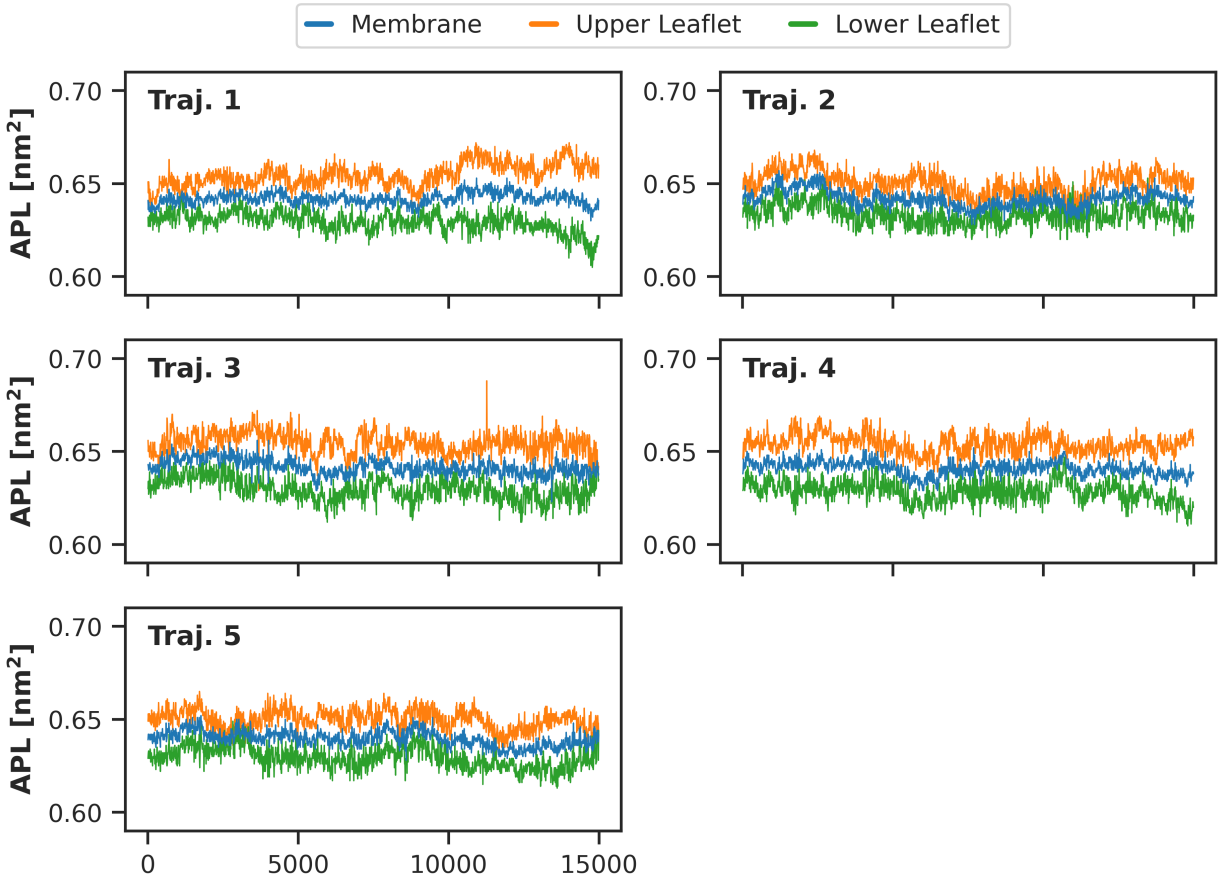


Figure S5: Area per lipid in each production trajectory over time.

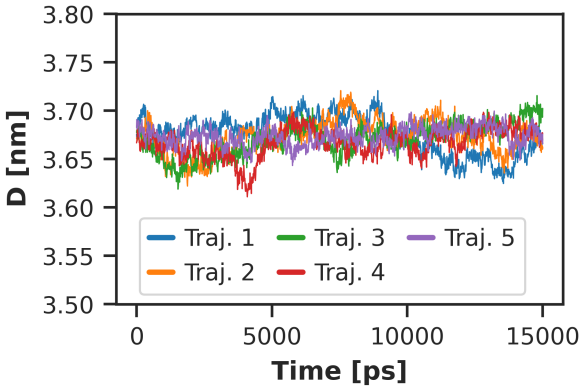


Figure S6: Bilayer thickness in each production trajectory over time.

# 3 QM Method Asessment

## 3.1 Further Chlorophyll Absorption Spectra in Diethyl Ether

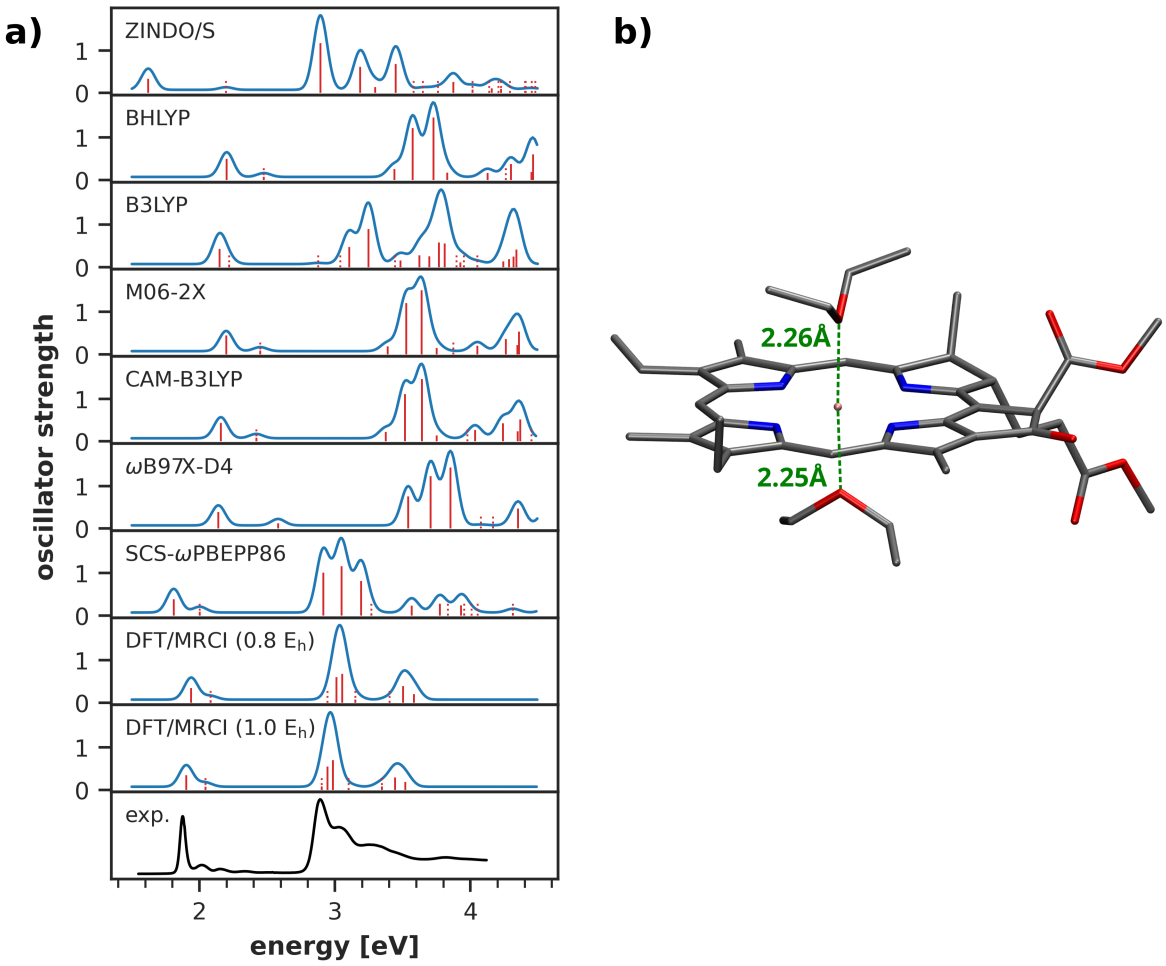


Figure S7: a) Absorption spectra of a single chlorophyll molecule, axially coordinated by two diethyl ether ligands and implicitly solvated in diethyl ether at different levels of theory, compared to an experimental spectrum. Stick spectra were convoluted by Gaussians ($\sigma$ = 0.05 eV). Excited states with an oscillator strength $< 0.1$ are indicated by dotted lines. b) Optimized structure ($r^2$SCAN-3c) used for these benchmark calculations. Hydrogens are omitted for clarity.

## 3.2 Truncation of the Phytyl Chain

The effects of truncating the phytyl chain at various lengths in the QM region were tested in one MD snapshot for chlorophyll eC-A1. The rest of the phytyl chain and the protein environment were described by point charges and the QM calculations were performed at the DFT/MRCI level as described in the main article. These test calculations were conducted with the basis set def2-SV(P) as the largest QM region was not computationally feasible otherwise. As the phytyl chain is not involved in the excitation, the site energies are nearly identical for different truncation schemes (table S4) and thus allow a truncation after the first carbon.

Table S4: DFT/MRCI site energies and oscillator strengths in one MD snapshot for chlorophyll eC-A1 with the phytyl chain truncated after the first (C1), sixth (C6) and twelfth (C12) carbon.

| | C1 | | C6 | | C12 | |
|---|---|---|---|---|---|---|
| State | E [eV] | f | E [eV] | f | E [eV] | f |
| 1 | 1.872 | 0.33578 | 1.870 | 0.33873 | 1.869 | 0.34035 |
| 2 | 2.075 | 0.07015 | 2.077 | 0.07000 | 2.074 | 0.06990 |
| 3 | 2.849 | 0.40940 | 2.853 | 0.42848 | 2.850 | 0.41902 |
| 4 | 3.009 | 0.25724 | 3.017 | 0.25709 | 3.013 | 0.24418 |

# 4 Data Set of Averaged Site Energies

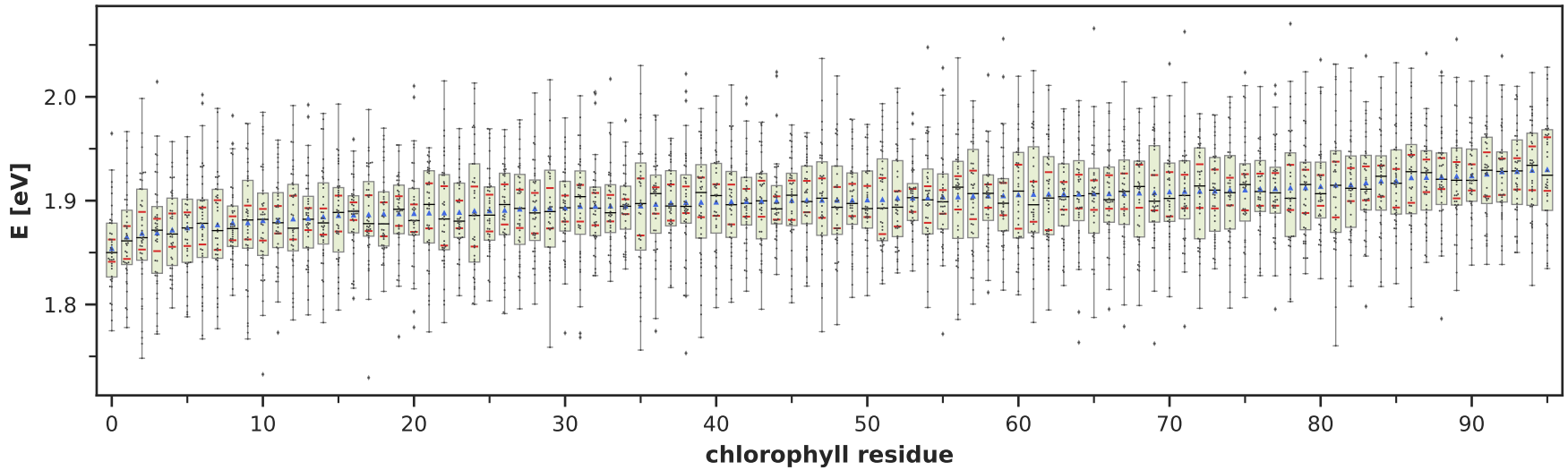


Figure S8: Site energies for each chlorophyll residue, ordered by ascending arithmetic mean (blue triangles) in 40 MD snapshots. Black lines signify the median, red lines its bootstrapped 95 % confidence interval. The green boxes extend from the beginning of the second to the end of the third quartile, whiskers extend to the minimum and maximum values of the data or to 1.5 times the interquartile range in case of outliers (gray diamonds).

Table S5: Arithmetic mean and median of the site energy distributions for each chlorophyll with (subscript *env*) and without (subscript *vac*) point charge environment in 40 MD snapshots. Table rows are sorted in ascending order by the mean of the site energy $E_{\mathrm{env}}^{\mathrm{mean}}$, same as in fig. 8 of the main paper. The column *ID (MD)* refers to the residue ID used in our MD simulations, while *Name* denotes the standard residue numbering scheme in PSI[S3,S9] and *ID (x-ray)* is the residue ID used in the crystal structure PDB (1JB0).[S3]

| index | ID (MD) | Name | ID (x-ray) | $E_{\mathrm{env}}^{\mathrm{mean}}$ [eV] | $E_{\mathrm{vac}}^{\mathrm{mean}}$ [eV] | $\Delta E_{\mathrm{struct}}^{\mathrm{mean}}$ [eV] | $\Delta E_{\mathrm{elec}}^{\mathrm{mean}}$ [eV] | $E_{\mathrm{env}}^{\mathrm{med}}$ [eV] | $E_{\mathrm{vac}}^{\mathrm{med}}$ [eV] | $\Delta E_{\mathrm{struct}}^{\mathrm{med}}$ [eV] | $\Delta E_{\mathrm{elec}}^{\mathrm{med}}$ [eV] |
|---|---|---|---|---|---|---|---|---|---|---|---|
| 0 | 2407 | B22 | 1222 | 1.853455 | 1.879903 | -0.035949 | -0.009847 | 1.85000 | 1.87425 | -0.041602 | -0.00880 |
| 1 | 2417 | B32 | 1232 | 1.864715 | 1.904375 | -0.011477 | -0.023060 | 1.86125 | 1.90520 | -0.010652 | -0.02040 |
| 2 | 2349 | A10 | 1110 | 1.868052 | 1.891107 | -0.024744 | -0.006455 | 1.86435 | 1.89500 | -0.020852 | -0.00785 |
| 3 | 2390 | B5 | 1205 | 1.868535 | 1.921047 | 0.005196 | -0.035912 | 1.87155 | 1.92025 | 0.004398 | -0.03350 |
| 4 | 2386 | B1 | 1201 | 1.871347 | 1.903722 | -0.012129 | -0.015775 | 1.86785 | 1.90600 | -0.009852 | -0.01585 |
| 5 | 2374 | A35 | 1135 | 1.873417 | 1.902643 | -0.013209 | -0.012625 | 1.87375 | 1.90500 | -0.010852 | -0.01185 |
| 6 | 2403 | B18 | 1218 | 1.875532 | 1.911070 | -0.004782 | -0.018937 | 1.87815 | 1.91530 | -0.000552 | -0.01565 |
| 7 | 2392 | B7 | 1207 | 1.876372 | 1.905655 | -0.010197 | -0.012682 | 1.87115 | 1.90300 | -0.012852 | -0.01295 |
| 8 | 2359 | A20 | 1120 | 1.878215 | 1.901123 | -0.014729 | -0.006307 | 1.87310 | 1.90055 | -0.015302 | -0.00680 |
| 9 | 2409 | B24 | 1224 | 1.878295 | 1.899550 | -0.016302 | -0.004655 | 1.88140 | 1.90030 | -0.015552 | -0.00480 |
| 10 | 2415 | B30 | 1230 | 1.880343 | 1.903687 | -0.012164 | -0.006745 | 1.88195 | 1.90460 | -0.011252 | -0.00875 |
| 11 | 2366 | A27 | 1127 | 1.880590 | 1.922427 | 0.006576 | -0.025237 | 1.87870 | 1.92430 | 0.008448 | -0.02605 |
| 12 | 2406 | B21 | 1221 | 1.881983 | 1.918415 | 0.002563 | -0.019832 | 1.87350 | 1.91455 | -0.001302 | -0.01860 |
| 13 | 2382 | PL1 | 1801 | 1.882463 | 1.911225 | -0.004627 | -0.012162 | 1.88210 | 1.91410 | -0.001752 | -0.01370 |
| 14 | 2373 | A34 | 1134 | 1.883987 | 1.901632 | -0.014219 | -0.001045 | 1.87855 | 1.90890 | -0.006952 | 0.00045 |
| 15 | 2342 | A3 | 1103 | 1.885278 | 1.907900 | -0.007952 | -0.006022 | 1.88865 | 1.90915 | -0.006702 | -0.00545 |
| 16 | 2389 | B4 | 1204 | 1.885675 | 1.916680 | 0.000828 | -0.014405 | 1.88980 | 1.91905 | 0.003198 | -0.01395 |
| 17 | 2378 | A39 | 1139 | 1.886075 | 1.916050 | 0.000198 | -0.013375 | 1.87785 | 1.91280 | -0.003052 | -0.01310 |
| 18 | 2354 | A15 | 1115 | 1.886747 | 1.907238 | -0.008614 | -0.003890 | 1.87765 | 1.90600 | -0.009852 | -0.00470 |
| 19 | 2367 | A28 | 1128 | 1.886975 | 1.909623 | -0.006229 | -0.006047 | 1.89170 | 1.91175 | -0.004102 | -0.00395 |
| 20 | 2398 | B13 | 1213 | 1.887160 | 1.900198 | -0.015654 | 0.003563 | 1.88080 | 1.89745 | -0.018402 | 0.00140 |
| 21 | 2351 | A12 | 1112 | 1.887660 | 1.911280 | -0.004572 | -0.007020 | 1.89635 | 1.91160 | -0.004252 | -0.00980 |
| 22 | 2365 | A26 | 1126 | 1.887768 | 1.914357 | -0.001494 | -0.009990 | 1.88235 | 1.91430 | -0.001552 | -0.01250 |
| 23 | 2337 | eC-A1 | 1011 | 1.888330 | 1.901138 | -0.014714 | 0.003793 | 1.88025 | 1.89670 | -0.019152 | 0.00325 |
| 24 | 2425 | J2 | 1302 | 1.889665 | 1.917935 | 0.002083 | -0.011670 | 1.88575 | 1.91325 | -0.002602 | -0.00775 |
| 25 | 2422 | B38 | 1238 | 1.889822 | 1.910750 | -0.005102 | -0.004327 | 1.88605 | 1.91080 | -0.005052 | -0.00615 |
| 26 | 2427 | K1 | 1401 | 1.890318 | 1.904300 | -0.011552 | 0.002618 | 1.89625 | 1.90960 | -0.006252 | 0.00370 |
| 27 | 2418 | B33 | 1233 | 1.891440 | 1.900678 | -0.015174 | 0.007363 | 1.89255 | 1.89590 | -0.019952 | 0.00890 |
| 28 | 2377 | A38 | 1138 | 1.891883 | 1.916625 | 0.000773 | -0.008142 | 1.88820 | 1.90995 | -0.005902 | -0.00785 |
| 29 | 2431 | M1 | 1601 | 1.892145 | 1.914368 | -0.001484 | -0.005622 | 1.88950 | 1.92235 | 0.006498 | -0.00160 |
| 30 | 2353 | A14 | 1114 | 1.892545 | 1.909417 | -0.006434 | -0.000272 | 1.89315 | 1.91350 | -0.002352 | 0.00130 |
| 31 | 2430 | L3 | 1503 | 1.894138 | 1.913080 | -0.002772 | -0.002342 | 1.90385 | 1.91655 | 0.000698 | -0.00525 |

*continued on next page*

Table S5: continued.

| index | ID (MD) | Name | ID (x-ray) | $E_{\mathrm{env}}^{\mathrm{mean}}$ [eV] | $E_{\mathrm{vac}}^{\mathrm{mean}}$ [eV] | $\Delta E_{\mathrm{struct}}^{\mathrm{mean}}$ [eV] | $\Delta E_{\mathrm{elec}}^{\mathrm{mean}}$ [eV] | $E_{\mathrm{env}}^{\mathrm{med}}$ [eV] | $E_{\mathrm{vac}}^{\mathrm{med}}$ [eV] | $\Delta E_{\mathrm{struct}}^{\mathrm{med}}$ [eV] | $\Delta E_{\mathrm{elec}}^{\mathrm{med}}$ [eV] |
|---|---|---|---|---|---|---|---|---|---|---|---|
| 32 | 2416 | B31 | 1231 | 1.894322 | 1.912292 | -0.003559 | -0.001370 | 1.89275 | 1.90910 | -0.006752 | -0.00140 |
| 33 | 2405 | B20 | 1220 | 1.894405 | 1.913845 | -0.002007 | -0.002840 | 1.88835 | 1.91235 | -0.003502 | -0.00260 |
| 34 | 2347 | A8 | 1108 | 1.894467 | 1.909172 | -0.006679 | 0.001895 | 1.89460 | 1.91625 | 0.000398 | 0.00280 |
| 35 | 2393 | B8 | 1208 | 1.894478 | 1.923815 | 0.007963 | -0.012737 | 1.89720 | 1.93460 | 0.018748 | -0.01475 |
| 36 | 2404 | B19 | 1219 | 1.896085 | 1.924205 | 0.008353 | -0.011520 | 1.90680 | 1.93340 | 0.017548 | -0.01180 |
| 37 | 2338 | eC-A2 | 1022 | 1.897437 | 1.902822 | -0.013029 | 0.011215 | 1.89485 | 1.89735 | -0.018502 | 0.01175 |
| 38 | 2412 | B27 | 1227 | 1.897847 | 1.924983 | 0.009131 | -0.010535 | 1.89450 | 1.91955 | 0.003698 | -0.01080 |
| 39 | 2411 | B26 | 1226 | 1.898017 | 1.910083 | -0.005769 | 0.004535 | 1.90785 | 1.91685 | 0.000998 | 0.00450 |
| 40 | 2375 | A36 | 1136 | 1.898157 | 1.923840 | 0.007988 | -0.009082 | 1.90490 | 1.93095 | 0.015098 | -0.00455 |
| 41 | 2410 | B25 | 1225 | 1.898637 | 1.930700 | 0.014848 | -0.015462 | 1.89610 | 1.92665 | 0.010798 | -0.01495 |
| 42 | 2345 | A6 | 1106 | 1.898895 | 1.916257 | 0.000406 | -0.000762 | 1.89740 | 1.91895 | 0.003098 | -0.00180 |
| 43 | 2341 | A2 | 1102 | 1.899025 | 1.911435 | -0.004417 | 0.004190 | 1.89980 | 1.91345 | -0.002402 | 0.00290 |
| 44 | 2363 | A24 | 1124 | 1.899347 | 1.910568 | -0.005284 | 0.005380 | 1.89210 | 1.90565 | -0.010202 | 0.00745 |
| 45 | 2364 | A25 | 1125 | 1.899860 | 1.914992 | -0.000859 | 0.001468 | 1.90525 | 1.92215 | 0.006298 | 0.00375 |
| 46 | 2395 | B10 | 1210 | 1.899963 | 1.909255 | -0.006597 | 0.007308 | 1.89910 | 1.90850 | -0.007352 | 0.00400 |
| 47 | 2358 | A19 | 1119 | 1.899990 | 1.923783 | 0.007931 | -0.007192 | 1.90235 | 1.92805 | 0.012198 | -0.00715 |
| 48 | 2371 | A32 | 1132 | 1.900000 | 1.923892 | 0.008041 | -0.007292 | 1.89365 | 1.91495 | -0.000902 | -0.00355 |
| 49 | 2372 | A33 | 1133 | 1.900048 | 1.918360 | 0.002508 | -0.001712 | 1.89730 | 1.91955 | 0.003698 | -0.00260 |
| 50 | 2414 | B29 | 1229 | 1.900293 | 1.900165 | -0.015687 | 0.016728 | 1.89235 | 1.89380 | -0.022052 | 0.01635 |
| 51 | 2381 | K2 | 1402 | 1.900640 | 1.903497 | -0.012354 | 0.013743 | 1.89280 | 1.89495 | -0.020902 | 0.01250 |
| 52 | 2376 | A37 | 1137 | 1.902035 | 1.920567 | 0.004716 | -0.001932 | 1.89370 | 1.91190 | -0.003952 | -0.00140 |
| 53 | 2380 | B37 | 1237 | 1.902203 | 1.937223 | 0.021371 | -0.018420 | 1.90285 | 1.93750 | 0.021648 | -0.02170 |
| 54 | 2420 | B35 | 1235 | 1.902540 | 1.929632 | 0.013781 | -0.010492 | 1.90075 | 1.92495 | 0.009098 | -0.00945 |
| 55 | 2340 | A1 | 1101 | 1.903197 | 1.914115 | -0.001737 | 0.005683 | 1.89890 | 1.91095 | -0.004902 | 0.00565 |
| 56 | 2384 | eC-B2 | 1012 | 1.903217 | 1.908553 | -0.007299 | 0.011265 | 1.91300 | 1.92015 | 0.004298 | 0.01020 |
| 57 | 2394 | B9 | 1209 | 1.904045 | 1.919852 | 0.004001 | 0.000793 | 1.90680 | 1.91695 | 0.001098 | -0.00070 |
| 58 | 2352 | A13 | 1113 | 1.904180 | 1.917625 | 0.001773 | 0.003155 | 1.90705 | 1.91805 | 0.002198 | 0.00345 |
| 59 | 2370 | A31 | 1131 | 1.904322 | 1.907128 | -0.008724 | 0.013795 | 1.89750 | 1.90560 | -0.010252 | 0.01190 |
| 60 | 2355 | A16 | 1116 | 1.905285 | 1.928820 | 0.012968 | -0.006935 | 1.90920 | 1.93720 | 0.021348 | -0.00125 |
| 61 | 2356 | A17 | 1117 | 1.905665 | 1.918992 | 0.003141 | 0.003273 | 1.89605 | 1.90460 | -0.011252 | 0.00580 |
| 62 | 2397 | B12 | 1212 | 1.905957 | 1.926785 | 0.010933 | -0.004227 | 1.90250 | 1.92595 | 0.010098 | -0.00645 |
| 63 | 2426 | J3 | 1303 | 1.905977 | 1.925720 | 0.009868 | -0.003142 | 1.90380 | 1.92390 | 0.008048 | -0.00345 |
| 64 | 2344 | A5 | 1105 | 1.905982 | 1.919150 | 0.003298 | 0.003433 | 1.90475 | 1.91565 | -0.000202 | 0.00365 |
| 65 | 2357 | A18 | 1118 | 1.906210 | 1.919397 | 0.003546 | 0.003413 | 1.90670 | 1.92210 | 0.006248 | 0.00390 |
| 66 | 2387 | B2 | 1202 | 1.906287 | 1.921128 | 0.005276 | 0.001760 | 1.90095 | 1.92060 | 0.004748 | 0.00170 |
| 67 | 2428 | L1 | 1501 | 1.906370 | 1.916643 | 0.000791 | 0.006328 | 1.90890 | 1.91910 | 0.003248 | 0.00750 |
| 68 | 2348 | A9 | 1109 | 1.906925 | 1.918485 | 0.002633 | 0.005040 | 1.91365 | 1.92270 | 0.006848 | 0.00500 |

*continued on next page*

Table S5: continued.

| index | ID (MD) | Name | ID (x-ray) | $E_{\mathrm{env}}^{\mathrm{mean}}$ [eV] | $E_{\mathrm{vac}}^{\mathrm{mean}}$ [eV] | $\Delta E_{\mathrm{struct}}^{\mathrm{mean}}$ [eV] | $\Delta E_{\mathrm{elec}}^{\mathrm{mean}}$ [eV] | $E_{\mathrm{env}}^{\mathrm{med}}$ [eV] | $E_{\mathrm{vac}}^{\mathrm{med}}$ [eV] | $\Delta E_{\mathrm{struct}}^{\mathrm{med}}$ [eV] | $\Delta E_{\mathrm{elec}}^{\mathrm{med}}$ [eV] |
|---|---|---|---|---|---|---|---|---|---|---|---|
| 69 | 2391 | B6 | 1206 | 1.907240 | 1.924843 | 0.008991 | -0.001002 | 1.89950 | 1.92360 | 0.007748 | 0.00190 |
| 70 | 2383 | eC-B1 | 1021 | 1.908070 | 1.917340 | 0.001488 | 0.007330 | 1.90215 | 1.92280 | 0.006948 | 0.00755 |
| 71 | 2362 | A23 | 1123 | 1.908513 | 1.910687 | -0.005164 | 0.014425 | 1.90500 | 1.90355 | -0.012302 | 0.01465 |
| 72 | 2429 | L2 | 1502 | 1.908653 | 1.924425 | 0.008573 | 0.000828 | 1.91415 | 1.92875 | 0.012898 | 0.00230 |
| 73 | 2369 | A30 | 1130 | 1.908765 | 1.920622 | 0.004771 | 0.004743 | 1.91015 | 1.92150 | 0.005648 | 0.00490 |
| 74 | 2343 | A4 | 1104 | 1.909773 | 1.912655 | -0.003197 | 0.013718 | 1.90770 | 1.90795 | -0.007902 | 0.01220 |
| 75 | 2360 | A21 | 1121 | 1.909940 | 1.917733 | 0.001881 | 0.008808 | 1.91550 | 1.91685 | 0.000998 | 0.00660 |
| 76 | 2419 | B34 | 1234 | 1.910905 | 1.917778 | 0.001926 | 0.009728 | 1.90875 | 1.91445 | -0.001402 | 0.01045 |
| 77 | 2421 | B36 | 1236 | 1.911068 | 1.919745 | 0.003893 | 0.007923 | 1.90750 | 1.91250 | -0.003352 | 0.00830 |
| 78 | 2400 | B15 | 1215 | 1.911920 | 1.926585 | 0.010733 | 0.001935 | 1.90230 | 1.91895 | 0.003098 | 0.00295 |
| 79 | 2396 | B11 | 1211 | 1.912047 | 1.924960 | 0.009108 | 0.003688 | 1.91555 | 1.92525 | 0.009398 | 0.00710 |
| 80 | 2399 | B14 | 1214 | 1.913243 | 1.922330 | 0.006478 | 0.007513 | 1.90670 | 1.92305 | 0.007198 | 0.00830 |
| 81 | 2385 | eC-B3 | 1023 | 1.914633 | 1.901227 | -0.014624 | 0.030005 | 1.91480 | 1.89380 | -0.022052 | 0.03080 |
| 82 | 2423 | B39 | 1239 | 1.914892 | 1.926077 | 0.010226 | 0.005415 | 1.91195 | 1.92310 | 0.007248 | 0.00785 |
| 83 | 2401 | B16 | 1216 | 1.916793 | 1.928077 | 0.012226 | 0.005315 | 1.91105 | 1.92530 | 0.009448 | 0.00450 |
| 84 | 2432 | X1 | 1701 | 1.918455 | 1.922808 | 0.006956 | 0.012248 | 1.92350 | 1.92270 | 0.006848 | 0.01230 |
| 85 | 2368 | A29 | 1129 | 1.918708 | 1.926020 | 0.010168 | 0.009288 | 1.91675 | 1.92590 | 0.010048 | 0.01065 |
| 86 | 2346 | A7 | 1107 | 1.921690 | 1.920180 | 0.004328 | 0.018110 | 1.92790 | 1.92585 | 0.009998 | 0.01760 |
| 87 | 2408 | B23 | 1223 | 1.921953 | 1.925617 | 0.009766 | 0.012935 | 1.92700 | 1.92660 | 0.010748 | 0.01320 |
| 88 | 2424 | J1 | 1301 | 1.922465 | 1.923923 | 0.008071 | 0.015143 | 1.92060 | 1.92065 | 0.004798 | 0.01645 |
| 89 | 2361 | A22 | 1122 | 1.923098 | 1.931405 | 0.015553 | 0.008293 | 1.91965 | 1.92830 | 0.012448 | 0.00875 |
| 90 | 2402 | B17 | 1217 | 1.923480 | 1.922458 | 0.006606 | 0.017623 | 1.91955 | 1.91950 | 0.003648 | 0.01715 |
| 91 | 2379 | A40 | 1140 | 1.925195 | 1.924612 | 0.008761 | 0.017183 | 1.92930 | 1.92395 | 0.008098 | 0.01895 |
| 92 | 2413 | B28 | 1228 | 1.927463 | 1.932893 | 0.017041 | 0.011170 | 1.92815 | 1.93645 | 0.020598 | 0.00980 |
| 93 | 2388 | B3 | 1203 | 1.927652 | 1.924952 | 0.009101 | 0.019300 | 1.92865 | 1.92045 | 0.004598 | 0.01755 |
| 94 | 2350 | A11 | 1111 | 1.928800 | 1.939572 | 0.023721 | 0.005828 | 1.93405 | 1.94195 | 0.026098 | 0.00475 |
| 95 | 2339 | eC-A3 | 1013 | 1.929500 | 1.925275 | 0.009423 | 0.020825 | 1.92450 | 1.92815 | 0.012298 | 0.02185 |

Table S6: Arithmetic mean and median of the site energy distributions for the reaction center chlorophylls including the point charge environment in 200 MD snapshots. The column *ID (MD)* refers to the residue ID used in our MD simulations, while *Name* denotes the standard residue numbering scheme in PSI[S3,S9] and *ID (x-ray)* is the residue ID used in the crystal structure PDB (1JB0).[S3]

| index | ID (MD) | Name | ID (x-ray) | $E_{\mathrm{env}}^{\mathrm{mean}}$ [eV] | $E_{\mathrm{env}}^{\mathrm{med}}$ [eV] |
|---|---|---|---|---|---|
| 0 | 2384 | eC-B2 | 1012 | 1.892429 | 1.89065 |
| 1 | 2338 | eC-A2 | 1022 | 1.893950 | 1.89290 |
| 2 | 2337 | eC-A1 | 1011 | 1.896057 | 1.89335 |
| 3 | 2383 | eC-B1 | 1021 | 1.906744 | 1.90615 |
| 4 | 2385 | eC-B3 | 1023 | 1.915238 | 1.91550 |
| 5 | 2339 | eC-A3 | 1013 | 1.915286 | 1.91185 |

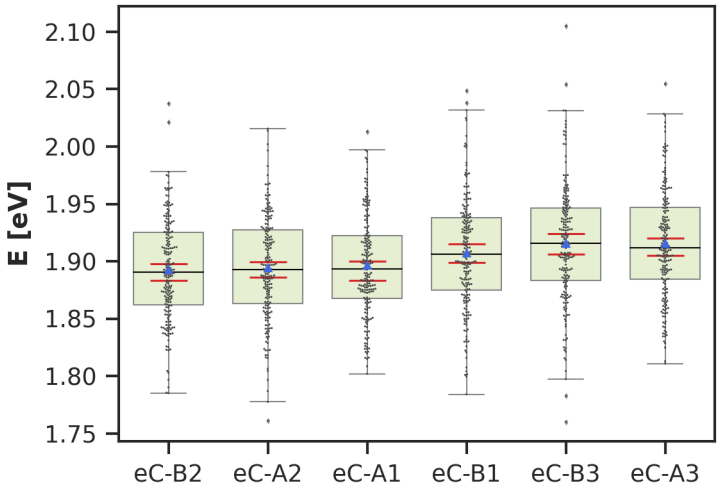


Figure S9: Site energies for the RC chlorophylls, ordered by ascending arithmetic mean (blue triangles) in 200 MD snapshots. Black lines signify the median, red lines its bootstrapped 95 % confidence interval. The green boxes extend from the beginning of the second to the end of the third quartile, whiskers extend to the minimum and maximum values of the data or to 1.5 times the interquartile range in case of outliers (gray diamonds).

# 5 Statistical Considerations

## 5.1 Site Energy Convergence with the Number of Snapshots

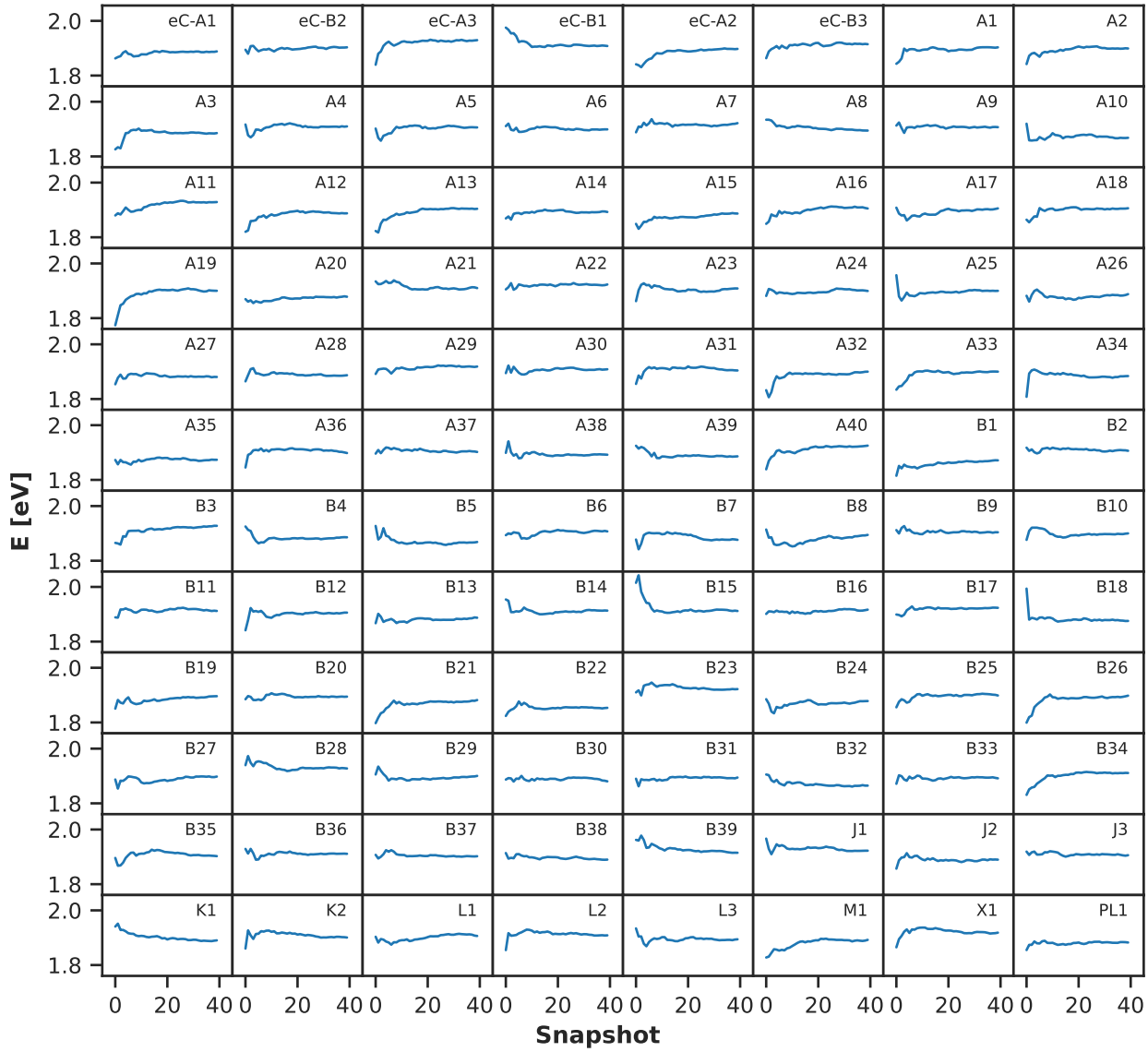


Figure S10: Convergence of the mean site energy for each chlorophyll with the number of snapshots.

## 5.2 ANOVA and HSD Analysis

The raw results of the statistical analysis are provided as csv-files at 10.5281/zenodo.6576313.

A one-way ANOVA was performed to assess the significance of the mean site energies. The analysis showed that there is a statistically significant difference in the site energies between at least two chlorophylls ($F(95, 3744) = 4.175, p = 0.000$). Tukey's Honestly Significant Difference (HSD) test for multiple comparisons found that the mean site energies were significantly different for 165 pairs of chlorophylls, based on $p < 0.05$. They correspond to the extreme cases of red and blue chlorophylls discussed in the main manuscript.

An ANOVA analysis for the site energies of the RC chlorophylls in 200 MD snapshots also revealed significant differences ($F(5, 1194) = 10.148, p = 0.000$). Tukey's HSD test found seven out of 15 pairs of RC chlorophylls with significantly different mean site energies, based on $p < 0.05$.

Concerning the structural site energy shifts, an ANOVA anaylsis shows significant differences in at least two pairs of chlorophylls ($F(95, 3744) = 2.043, p = 0.000$). The same holds for the electrostatic shifts ($F(95, 3744) = 19.413, p = 0.000$). Tukey's HSD test reveals significant differences in $\Delta E_{\text{struct}}$ in 29 out of 4560 possible pairs of chlorophylls. For $\Delta E_{\text{elec}}$, 1479 pairs of chlorophylls with statistically significant differences are found.

# 6 Comparison with Literature Data

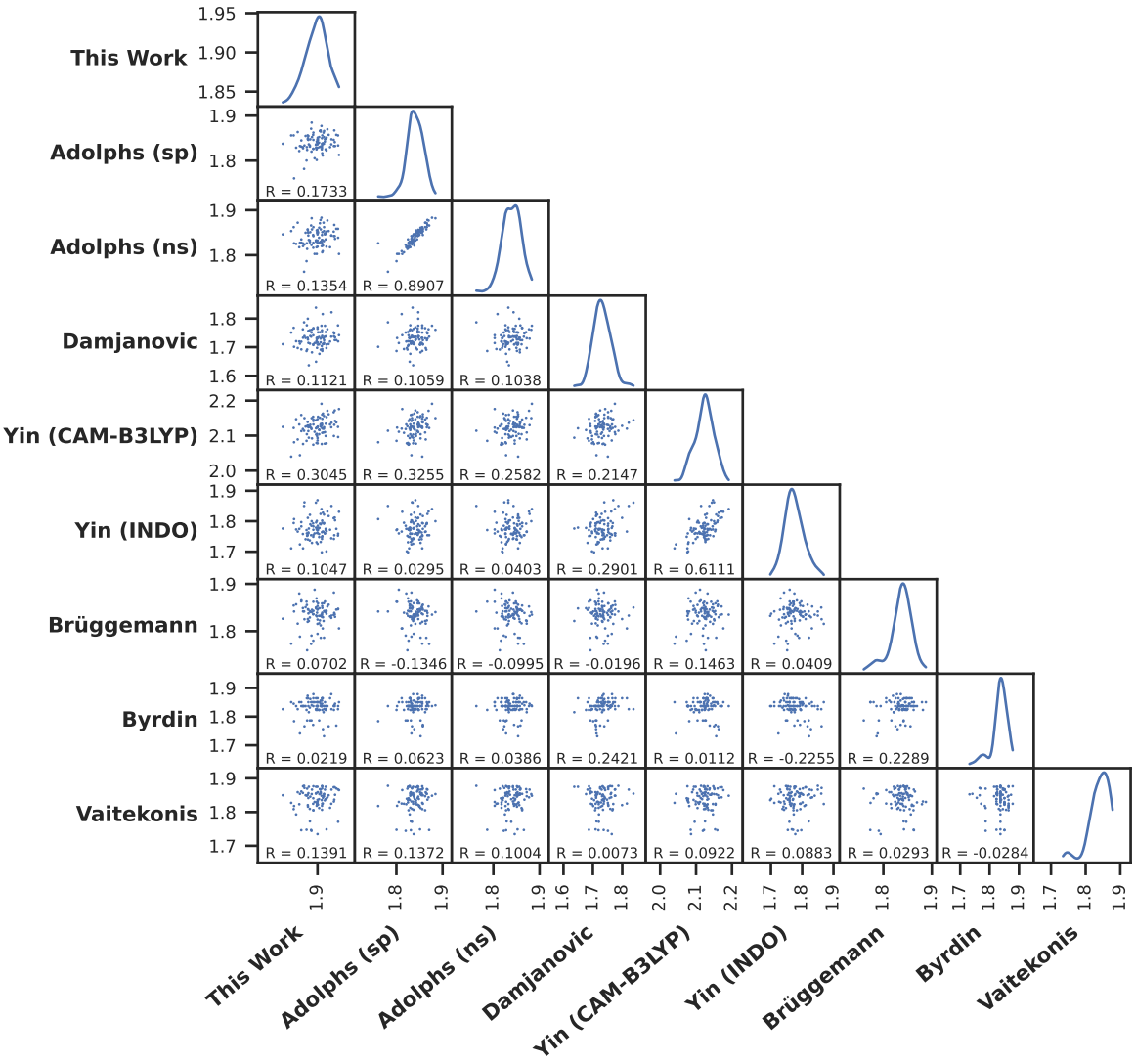


Figure S11: Correlation of previously published site energies with the present work. Adolphs *et al.*[S10] employed the charge density coupling (CDC) method for the protein in standard (st) and non-standard (ns) protonation states. Damjanovic *et al.*[S11] and Yin *et al.*[S12] performed semi-empirical and first quantum chemical calculations. Brüggemann *et al.*,[S13] Byrdin *et al.*[S9] and Vaitekonis *et al.*[S14] performed fits of optical spectra. Energies are given in eV. The diagonal plots illustrate the kernel density estimate (KDE) of each data set.

# 7 Dimer Excited States

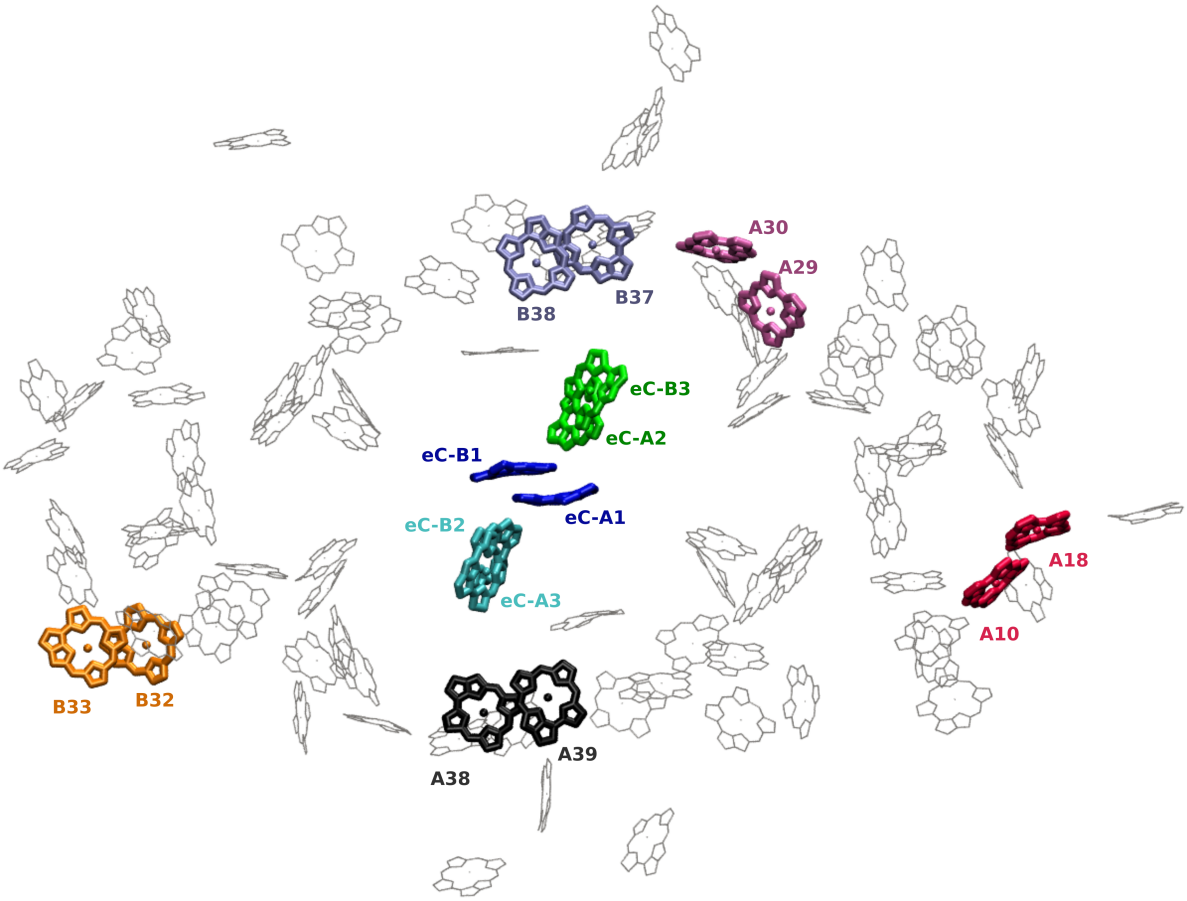


Figure S12: Selected chlorophyll pairs to compute excited states at the QM($\omega$B97X-D4/def2-TZVP)/MM level of theory.

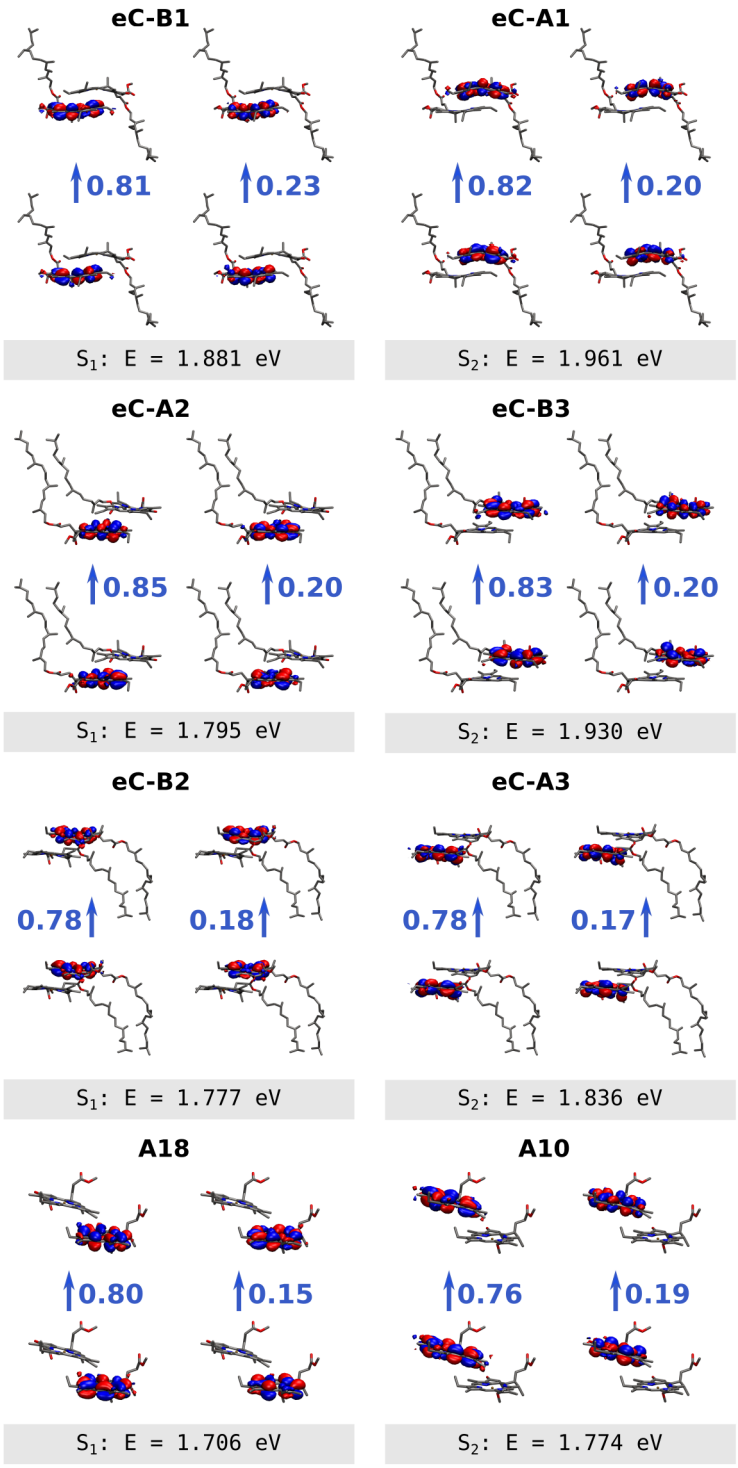


Figure S13: Natural transition orbtials and excitation energies for the first and second excited state of selected chlorophyll pairs in PSI at the QM($\omega$B97X-D4/def2-TZVP)/MM level (isovalue: 0.02). The name above the figures refers to the chlorophyll, on which the excitation is localized. Numbers next to the arrows denote the singular value of the transition.

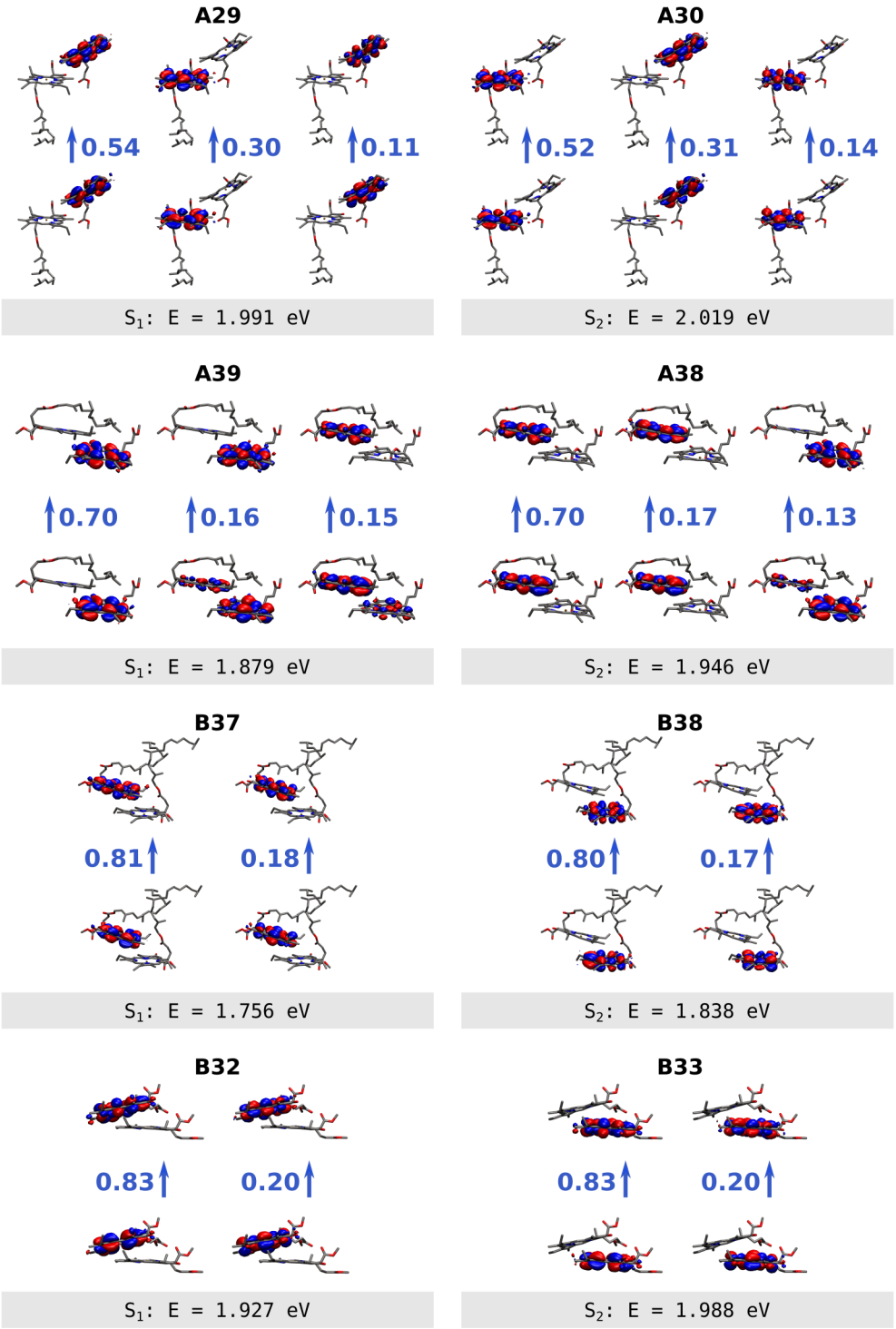


Figure S13 (cont.): Natural transition orbtials and excitation energies for the first and second excited state of selected chlorophyll pairs in PSI at the QM($\omega$B97X-D4/def2-TZVP)/MM level (isovalue: 0.02). The name above the figures refers to the chlorophyll, on which the excitation is localized. Numbers next to the arrows denote the singular value of the transition.

# 8 Excitonic Red-Shift of the Triad B31-B32-B33

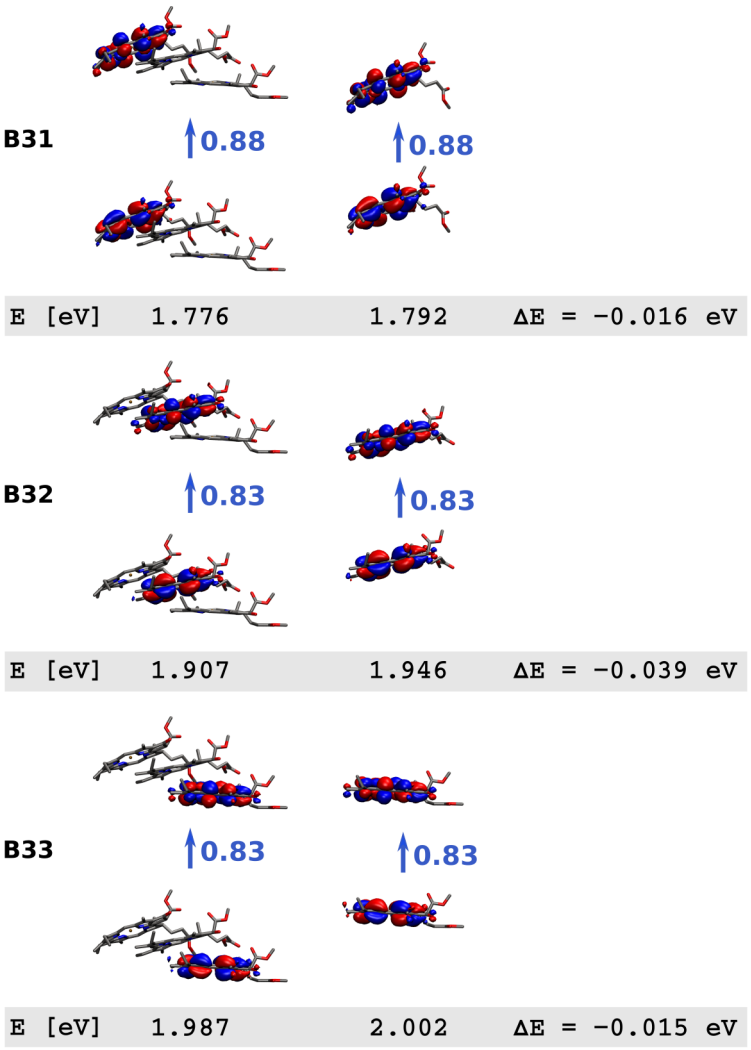


Figure S14: Natural transition orbtials for the $Q_y$ states of the triad B31-B32-B33 in PSI at the QM($\omega$B97X-D4/def2-TZVP)/MM level (isovalue: 0.02). Numbers next to the arrows denote the weight of the transition. In the left column the entire triad was treated quantum mechanically, in the right column only one of the monomers was included in the QM region. The resulting difference in the excitation energies can be attributed to excitonic coupling including both short- and long-range effects. The results show that the excited states remain localized on one of the monomers (cf. also fig. S13) and that excitonic coupling shifts the collective absorption band of this chlorophyll triad further to the red.

# 9 Exciton Energies and Fluctuations

Table S7: Mean exciton energies and chlorophyll contributions to each exciton. Chlorophylls were assigned to an exciton domain if their weight $w_i$ in the excitonic wavefunction was larger than 0.1. Contribution coefficients $c_i$ of each chlorophyll to the respective exciton are also provided. Table rows are sorted in ascending order by the exciton energy $E_{\mathrm{exc}}$. The column *ID (MD)* refers to the residue ID used in our MD simulations, while *Name* denotes the standard residue numbering scheme in PSI[S3,S9] and *ID (x-ray)* is the residue ID used in the crystal structure PDB (1JB0).[S3]

| Exciton | $E_{\mathrm{exc}}$ [eV] | name | ID (MD) | ID (x-ray) | $w_i$ | $c_i$ |
|---|---|---|---|---|---|---|
| 0 | 1.828042 | B31 | 2416 | 1231 | 0.195118 | -0.441721 |
| | | B32 | 2417 | 1232 | 0.649384 | 0.805844 |
| | | B33 | 2418 | 1233 | 0.152557 | -0.390586 |
| 1 | 1.848102 | B22 | 2407 | 1222 | 0.932454 | -0.965636 |
| 2 | 1.855110 | A32 | 2371 | 1132 | 0.300483 | 0.548163 |
| | | B7 | 2392 | 1207 | 0.637277 | -0.798296 |
| 3 | 1.855323 | eC-A1 | 2337 | 1011 | 0.494504 | -0.703210 |
| | | eC-B1 | 2383 | 1021 | 0.378447 | 0.615181 |
| 4 | 1.857625 | A18 | 2357 | 1118 | 0.140398 | -0.374697 |
| | | A10 | 2349 | 1110 | 0.798581 | 0.893634 |
| 5 | 1.858303 | A14 | 2353 | 1114 | 0.442904 | 0.665511 |
| | | A12 | 2351 | 1112 | 0.527440 | -0.726251 |
| 6 | 1.861035 | K1 | 2427 | 1401 | 0.243838 | -0.493799 |
| | | A34 | 2373 | 1134 | 0.503734 | 0.709742 |
| | | A33 | 2372 | 1133 | 0.146774 | -0.383111 |
| 7 | 1.861079 | B4 | 2389 | 1204 | 0.100653 | -0.317259 |
| | | B5 | 2390 | 1205 | 0.716660 | 0.846557 |
| 8 | 1.863934 | A35 | 2374 | 1135 | 0.760822 | 0.872251 |
| 9 | 1.866067 | B19 | 2404 | 1219 | 0.217122 | -0.465964 |
| | | B18 | 2403 | 1218 | 0.698064 | 0.835502 |
| 10 | 1.867530 | A21 | 2360 | 1121 | 0.178706 | 0.422736 |
| | | A20 | 2359 | 1120 | 0.778045 | -0.882069 |
| 11 | 1.868081 | A27 | 2366 | 1127 | 0.613017 | 0.782954 |
| | | A26 | 2365 | 1126 | 0.333531 | -0.577521 |
| 12 | 1.868466 | B1 | 2386 | 1201 | 0.878091 | 0.937065 |
| 13 | 1.870030 | B24 | 2409 | 1224 | 0.661983 | 0.813623 |
| | | B25 | 2410 | 1225 | 0.178490 | -0.422481 |
| 14 | 1.870608 | A38 | 2377 | 1138 | 0.365919 | 0.604912 |
| | | A39 | 2378 | 1139 | 0.581874 | -0.762807 |
| 15 | 1.871598 | B37 | 2380 | 1237 | 0.357380 | -0.597813 |
| | | B38 | 2422 | 1238 | 0.596142 | 0.772102 |
| 16 | 1.874623 | B29 | 2414 | 1229 | 0.152928 | -0.391060 |
| | | B30 | 2415 | 1230 | 0.769399 | 0.877154 |

*continued on next page*

Table S7: continued.

| Exciton | $E_{\mathrm{exc}}$ [eV] | name | ID (MD) | ID (x-ray) | $w_i$ | $c_i$ |
|---|---|---|---|---|---|---|
| 17 | 1.875401 | A15 | 2354 | 1115 | 0.255024 | 0.504999 |
| | | A28 | 2367 | 1128 | 0.182773 | -0.427519 |
| | | A3 | 2342 | 1103 | 0.362707 | 0.602252 |
| 18 | 1.875542 | K1 | 2427 | 1401 | 0.123142 | 0.350916 |
| | | A3 | 2342 | 1103 | 0.244751 | 0.494723 |
| | | A15 | 2354 | 1115 | 0.376818 | -0.613855 |
| | | A28 | 2367 | 1128 | 0.122904 | -0.350577 |
| 19 | 1.876628 | B20 | 2405 | 1220 | 0.161884 | -0.402348 |
| | | B21 | 2406 | 1221 | 0.708178 | 0.841533 |
| 20 | 1.879461 | PL1 | 2382 | 1801 | 0.280999 | 0.530093 |
| | | A37 | 2376 | 1137 | 0.189624 | 0.435459 |
| | | A36 | 2375 | 1136 | 0.165709 | -0.407074 |
| 21 | 1.880295 | eC-A2 | 2338 | 1022 | 0.146223 | -0.382391 |
| | | PL1 | 2382 | 1801 | 0.567957 | 0.753629 |
| 22 | 1.880905 | eC-B3 | 2385 | 1023 | 0.103896 | 0.322330 |
| | | eC-A2 | 2338 | 1022 | 0.300868 | -0.548514 |
| | | A25 | 2364 | 1125 | 0.164430 | 0.405500 |
| | | A16 | 2355 | 1116 | 0.135008 | -0.367435 |
| 23 | 1.882831 | eC-B2 | 2384 | 1012 | 0.540912 | -0.735467 |
| | | eC-A3 | 2339 | 1013 | 0.163749 | 0.404659 |
| 24 | 1.884245 | eC-A2 | 2338 | 1022 | 0.160273 | -0.400342 |
| | | A36 | 2375 | 1136 | 0.165179 | -0.406422 |
| | | A37 | 2376 | 1137 | 0.173057 | 0.416001 |
| | | A16 | 2355 | 1116 | 0.138346 | 0.371949 |
| 25 | 1.884562 | B33 | 2418 | 1233 | 0.355979 | -0.596640 |
| | | B31 | 2416 | 1231 | 0.382522 | 0.618484 |
| | | B23 | 2408 | 1223 | 0.106091 | -0.325716 |
| 26 | 1.885946 | B13 | 2398 | 1213 | 0.852941 | -0.923548 |
| 27 | 1.886643 | A28 | 2367 | 1128 | 0.219895 | 0.468930 |
| | | A2 | 2341 | 1102 | 0.177815 | -0.421681 |
| | | A5 | 2344 | 1105 | 0.109066 | 0.330252 |
| | | A6 | 2345 | 1106 | 0.263407 | -0.513231 |
| | | J2 | 2425 | 1302 | 0.121208 | -0.348149 |
| 28 | 1.887070 | J2 | 2425 | 1302 | 0.132203 | -0.363597 |
| | | A2 | 2341 | 1102 | 0.159052 | 0.398813 |
| | | A6 | 2345 | 1106 | 0.195989 | -0.442706 |
| | | A28 | 2367 | 1128 | 0.286312 | -0.535082 |
| 29 | 1.887657 | B27 | 2412 | 1227 | 0.545232 | -0.738398 |
| 30 | 1.888189 | B4 | 2389 | 1204 | 0.605488 | 0.778131 |
| 31 | 1.888801 | B35 | 2420 | 1235 | 0.364311 | 0.603582 |
| | | B36 | 2421 | 1236 | 0.165751 | -0.407125 |
| 32 | 1.890070 | J2 | 2425 | 1302 | 0.697440 | -0.835129 |
| | | A6 | 2345 | 1106 | 0.216526 | 0.465323 |
| 33 | 1.890081 | B26 | 2411 | 1226 | 0.334362 | -0.578240 |
| | | B2 | 2387 | 1202 | 0.191483 | 0.437587 |

*continued on next page*

Table S7: continued.

| Exciton | $E_{\mathrm{exc}}$ [eV] | name | ID (MD) | ID (x-ray) | $w_i$ | $c_i$ |
|---|---|---|---|---|---|---|
| | | B10 | 2395 | 1210 | 0.132953 | -0.364627 |
| 34 | 1.890788 | B8 | 2393 | 1208 | 0.117774 | -0.343182 |
| | | B9 | 2394 | 1209 | 0.558238 | 0.747153 |
| | | B17 | 2402 | 1217 | 0.209701 | -0.457931 |
| 35 | 1.891450 | A31 | 2370 | 1131 | 0.431046 | -0.656541 |
| | | L3 | 2430 | 1503 | 0.234599 | 0.484354 |
| 36 | 1.891923 | M1 | 2431 | 1601 | 0.682920 | -0.826390 |
| | | B26 | 2411 | 1226 | 0.138329 | 0.371927 |
| 37 | 1.892697 | A17 | 2356 | 1117 | 0.274275 | -0.523713 |
| | | A24 | 2363 | 1124 | 0.114487 | -0.338359 |
| | | A26 | 2365 | 1126 | 0.229358 | 0.478914 |
| 38 | 1.893645 | L3 | 2430 | 1503 | 0.676789 | 0.822672 |
| 39 | 1.893818 | A19 | 2358 | 1119 | 0.427088 | 0.653520 |
| | | A8 | 2347 | 1108 | 0.307000 | -0.554076 |
| 40 | 1.893838 | B8 | 2393 | 1208 | 0.158821 | -0.398523 |
| | | B19 | 2404 | 1219 | 0.128308 | -0.358202 |
| | | B26 | 2411 | 1226 | 0.139354 | -0.373302 |
| | | B20 | 2405 | 1220 | 0.142135 | 0.377008 |
| 41 | 1.893865 | B20 | 2405 | 1220 | 0.111430 | -0.333811 |
| | | B8 | 2393 | 1208 | 0.287124 | -0.535839 |
| | | M1 | 2431 | 1601 | 0.121646 | -0.348778 |
| | | B10 | 2395 | 1210 | 0.131978 | 0.363288 |
| 42 | 1.894098 | A8 | 2347 | 1108 | 0.557365 | 0.746569 |
| | | A19 | 2358 | 1119 | 0.197224 | 0.444099 |
| | | A23 | 2362 | 1123 | 0.103607 | -0.321880 |
| 43 | 1.895421 | B23 | 2408 | 1223 | 0.118736 | 0.344581 |
| | | B14 | 2399 | 1214 | 0.240402 | -0.490308 |
| | | B15 | 2400 | 1215 | 0.173575 | 0.416623 |
| | | B34 | 2419 | 1234 | 0.113378 | -0.336717 |
| 44 | 1.896178 | A4 | 2343 | 1104 | 0.268726 | -0.518388 |
| | | A3 | 2342 | 1103 | 0.129138 | 0.359358 |
| | | A9 | 2348 | 1109 | 0.101682 | -0.318876 |
| | | A2 | 2341 | 1102 | 0.337640 | 0.581068 |
| 45 | 1.897241 | A33 | 2372 | 1133 | 0.190437 | 0.436391 |
| | | A27 | 2366 | 1127 | 0.153116 | 0.391301 |
| | | A26 | 2365 | 1126 | 0.249558 | 0.499558 |
| 46 | 1.898247 | B10 | 2395 | 1210 | 0.100942 | -0.317713 |
| | | A1 | 2340 | 1101 | 0.232190 | 0.481861 |
| | | A38 | 2377 | 1138 | 0.132243 | -0.363653 |
| 47 | 1.898388 | B10 | 2395 | 1210 | 0.136873 | -0.369964 |
| | | A1 | 2340 | 1101 | 0.310317 | -0.557061 |
| | | A38 | 2377 | 1138 | 0.178030 | 0.421936 |
| 48 | 1.898794 | A24 | 2363 | 1124 | 0.118554 | 0.344317 |
| | | B10 | 2395 | 1210 | 0.101067 | 0.317910 |

*continued on next page*

Table S7: continued.

| Exciton | $E_{\mathrm{exc}}$ [eV] | name | ID (MD) | ID (x-ray) | $w_i$ | $c_i$ |
|---|---|---|---|---|---|---|
| 49 | 1.898831 | B10 | 2395 | 1210 | 0.126285 | -0.355366 |
| 50 | 1.899570 | A30 | 2369 | 1130 | 0.464012 | -0.681184 |
| | | A29 | 2368 | 1129 | 0.232011 | 0.481676 |
| | | K2 | 2381 | 1402 | 0.109151 | 0.330380 |
| 51 | 1.899850 | K2 | 2381 | 1402 | 0.726171 | 0.852157 |
| 52 | 1.900646 | B30 | 2415 | 1230 | 0.109560 | 0.330999 |
| | | J1 | 2424 | 1301 | 0.101269 | -0.318228 |
| | | B29 | 2414 | 1229 | 0.600378 | 0.774840 |
| 53 | 1.903487 | B19 | 2404 | 1219 | 0.260007 | 0.509908 |
| | | B16 | 2401 | 1216 | 0.240773 | -0.490687 |
| | | B20 | 2405 | 1220 | 0.235371 | 0.485151 |
| | | A13 | 2352 | 1113 | 0.128478 | -0.358438 |
| 54 | 1.903513 | A13 | 2352 | 1113 | 0.729275 | -0.853976 |
| 55 | 1.903947 | L1 | 2428 | 1501 | 0.597210 | 0.772793 |
| | | L2 | 2429 | 1502 | 0.337425 | -0.580883 |
| 56 | 1.904647 | A36 | 2375 | 1136 | 0.106929 | -0.327000 |
| | | A24 | 2363 | 1124 | 0.101461 | 0.318529 |
| | | A17 | 2356 | 1117 | 0.214894 | -0.463566 |
| | | A25 | 2364 | 1125 | 0.111551 | 0.333992 |
| 57 | 1.904731 | B25 | 2410 | 1225 | 0.149997 | 0.387294 |
| | | B12 | 2397 | 1212 | 0.413936 | 0.643379 |
| | | B11 | 2396 | 1211 | 0.135412 | -0.367984 |
| 58 | 1.905707 | B25 | 2410 | 1225 | 0.235655 | 0.485443 |
| | | B14 | 2399 | 1214 | 0.105429 | -0.324698 |
| | | B12 | 2397 | 1212 | 0.392341 | -0.626371 |
| 59 | 1.906036 | A22 | 2361 | 1122 | 0.107270 | -0.327520 |
| | | J3 | 2426 | 1303 | 0.116709 | -0.341628 |
| | | A19 | 2358 | 1119 | 0.104776 | 0.323691 |
| | | A23 | 2362 | 1123 | 0.484701 | 0.696204 |
| 60 | 1.906093 | J3 | 2426 | 1303 | 0.823790 | 0.907629 |
| 61 | 1.907203 | A9 | 2348 | 1109 | 0.664706 | 0.815295 |
| | | A4 | 2343 | 1104 | 0.174504 | -0.417737 |
| 62 | 1.908090 | A38 | 2377 | 1138 | 0.222040 | -0.471211 |
| | | A39 | 2378 | 1139 | 0.234728 | -0.484487 |
| | | A1 | 2340 | 1101 | 0.282835 | -0.531822 |
| 63 | 1.908553 | A31 | 2370 | 1131 | 0.119445 | 0.345608 |
| | | B6 | 2391 | 1206 | 0.346391 | -0.588550 |
| | | L1 | 2428 | 1501 | 0.101030 | -0.317852 |
| 64 | 1.909289 | A5 | 2344 | 1105 | 0.459890 | -0.678152 |
| | | A7 | 2346 | 1107 | 0.142323 | 0.377257 |
| | | A9 | 2348 | 1109 | 0.137955 | 0.371422 |
| 65 | 1.909864 | B11 | 2396 | 1211 | 0.119742 | 0.346038 |
| | | X1 | 2432 | 1701 | 0.295189 | 0.543313 |
| | | B35 | 2420 | 1235 | 0.226218 | -0.475624 |

*continued on next page*

Table S7: continued.

| Exciton | $E_{\mathrm{exc}}$ [eV] | name | ID (MD) | ID (x-ray) | $w_i$ | $c_i$ |
|---|---|---|---|---|---|---|
| | | B36 | 2421 | 1236 | 0.101370 | -0.318387 |
| 66 | 1.911168 | L2 | 2429 | 1502 | 0.417685 | -0.646285 |
| | | B39 | 2423 | 1239 | 0.199829 | -0.447022 |
| | | L1 | 2428 | 1501 | 0.204593 | -0.452320 |
| 67 | 1.912181 | B6 | 2391 | 1206 | 0.135390 | 0.367954 |
| | | B39 | 2423 | 1239 | 0.277248 | 0.526544 |
| | | B2 | 2387 | 1202 | 0.121776 | -0.348964 |
| 68 | 1.912666 | B39 | 2423 | 1239 | 0.155128 | 0.393862 |
| | | B11 | 2396 | 1211 | 0.337936 | -0.581323 |
| 69 | 1.914460 | B11 | 2396 | 1211 | 0.130986 | -0.361920 |
| | | B3 | 2388 | 1203 | 0.145256 | 0.381125 |
| | | B10 | 2395 | 1210 | 0.113752 | -0.337271 |
| | | B2 | 2387 | 1202 | 0.243389 | -0.493345 |
| 70 | 1.915101 | A21 | 2360 | 1121 | 0.289830 | 0.538359 |
| | | A18 | 2357 | 1118 | 0.156792 | -0.395970 |
| | | A22 | 2361 | 1122 | 0.214854 | -0.463523 |
| 71 | 1.916105 | A18 | 2357 | 1118 | 0.285880 | 0.534678 |
| 72 | 1.916961 | A18 | 2357 | 1118 | 0.265878 | 0.515634 |
| 73 | 1.918735 | B37 | 2380 | 1237 | 0.349259 | 0.590981 |
| | | B38 | 2422 | 1238 | 0.220158 | 0.469210 |
| 74 | 1.919690 | A4 | 2343 | 1104 | 0.135612 | 0.368256 |
| | | A40 | 2379 | 1140 | 0.289933 | -0.538454 |
| | | eC-A3 | 2339 | 1013 | 0.190786 | 0.436791 |
| 75 | 1.921137 | eC-A3 | 2339 | 1013 | 0.167977 | 0.409850 |
| | | A4 | 2343 | 1104 | 0.116551 | -0.341395 |
| 77 | 1.921671 | B15 | 2400 | 1215 | 0.139387 | 0.373346 |
| | | B16 | 2401 | 1216 | 0.218466 | -0.467404 |
| 78 | 1.923494 | A14 | 2353 | 1114 | 0.290541 | 0.539019 |
| | | A12 | 2351 | 1112 | 0.259554 | 0.509465 |
| 79 | 1.924126 | B16 | 2401 | 1216 | 0.121233 | 0.348186 |
| | | X1 | 2432 | 1701 | 0.202728 | 0.450253 |
| | | B23 | 2408 | 1223 | 0.153157 | -0.391352 |
| | | B34 | 2419 | 1234 | 0.171598 | -0.414244 |
| 80 | 1.924600 | J1 | 2424 | 1301 | 0.540084 | -0.734904 |
| 81 | 1.925327 | B31 | 2416 | 1231 | 0.103011 | -0.320953 |
| | | B15 | 2400 | 1215 | 0.148255 | 0.385039 |
| | | B16 | 2401 | 1216 | 0.162039 | 0.402540 |
| | | B32 | 2417 | 1232 | 0.121666 | -0.348806 |
| | | B33 | 2418 | 1233 | 0.123588 | -0.351551 |
| 82 | 1.926194 | A29 | 2368 | 1129 | 0.180379 | 0.424710 |
| | | B39 | 2423 | 1239 | 0.114209 | -0.337949 |
| | | eC-B3 | 2385 | 1023 | 0.292771 | -0.541083 |
| 83 | 1.928196 | A40 | 2379 | 1140 | 0.212333 | -0.460796 |
| 84 | 1.929014 | A40 | 2379 | 1140 | 0.195152 | -0.441760 |

*continued on next page*

Table S7: continued.

| Exciton | $E_{\mathrm{exc}}$ [eV] | name | ID (MD) | ID (x-ray) | $w_i$ | $c_i$ |
|---|---|---|---|---|---|---|
| | | eC-B3 | 2385 | 1023 | 0.108588 | -0.329527 |
| 85 | 1.932087 | A11 | 2350 | 1111 | 0.236406 | -0.486216 |
| | | B28 | 2413 | 1228 | 0.166345 | -0.407854 |
| 86 | 1.932273 | A11 | 2350 | 1111 | 0.156550 | -0.395664 |
| | | B36 | 2421 | 1236 | 0.133267 | -0.365057 |
| 87 | 1.934053 | A7 | 2346 | 1107 | 0.100599 | 0.317173 |
| | | B3 | 2388 | 1203 | 0.243536 | -0.493494 |
| | | B17 | 2402 | 1217 | 0.103352 | 0.321484 |
| 88 | 1.934916 | B17 | 2402 | 1217 | 0.123412 | 0.351300 |
| | | B3 | 2388 | 1203 | 0.174263 | -0.417448 |
| 89 | 1.937186 | A22 | 2361 | 1122 | 0.124780 | -0.353243 |
| 90 | 1.938129 | B17 | 2402 | 1217 | 0.214266 | 0.462889 |
| 91 | 1.939676 | B17 | 2402 | 1217 | 0.136998 | 0.370132 |
| | | B3 | 2388 | 1203 | 0.131734 | 0.362952 |
| 92 | 1.940546 | B28 | 2413 | 1228 | 0.362478 | -0.602061 |
| 93 | 1.947943 | eC-B1 | 2383 | 1021 | 0.117403 | -0.342641 |
| 94 | 1.954418 | eC-A1 | 2337 | 1011 | 0.206972 | 0.454942 |
| | | eC-B1 | 2383 | 1021 | 0.264167 | 0.513972 |
| 95 | 1.962741 | B23 | 2408 | 1223 | 0.270108 | -0.519719 |
| | | B31 | 2416 | 1231 | 0.131710 | -0.362919 |

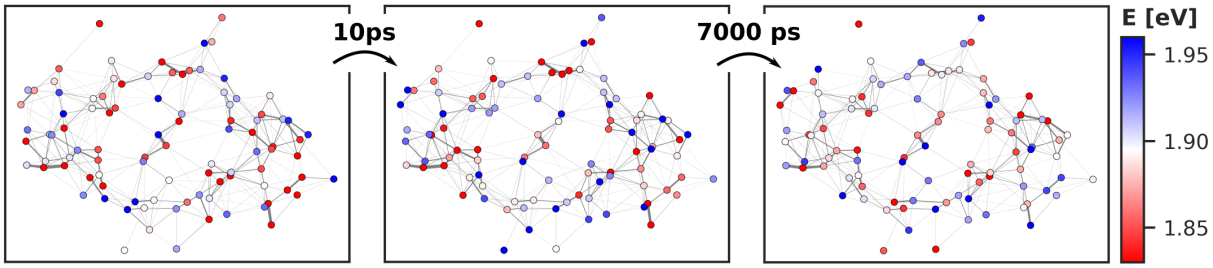


Figure S15: Exciton energies in three selected MD frames, analogous to Fig. 8 of the main article. Each dot signifies a chlorophyll and is colored by the lowest energy exciton domain that this chlorophyll belongs to ($w_i > 0.1$). Connecting lines represent the coupling strength ($V_{ij} > 1\,\mathrm{meV}$) with thicker lines denoting stronger coupling. The energy axis is centered around the mean of all exciton energies.

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