Supporting Information for "Site-Dependent Fluctuations Optimize Electronic Energy Transfer in the Fenna-Matthews-Olson Protein"

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Table S1. Exciton energies and eigenfunctions at 300 and 77 K.[a]

State	1	2	3	4	5	6	7	8
Energy (cm^{-1})	12174.7 (12281.7)	12315.4 (12421.3)	12326.7 (12443.5)	12422.2 (12515.6)	12460.1 (12570.6)	12648.2 (12768.0)	12670.4 (12785.3)	12712.4 (12794.0)
Site 1	−0.0764 (−0.0680)	0.3505 (0.1606)	0.8123 (0.8562)	−0.1353 (−0.2220)	−0.0348 (−0.0243)	−0.3223 (−0.1614)	−0.2959 (−0.2517)	−0.0225 (−0.3117)
Site 2	−0.1130 (−0.1070)	0.1668 (0.0878)	0.3590 (0.3672)	−0.1398 (−0.1734)	−0.0837 (−0.0533)	0.7876 (0.5396)	0.4229 (0.4050)	0.0695 (0.5983)
Site 3	0.9318 (0.9187)	−0.2214 (−0.3055)	0.1820 (0.1075)	−0.2040 (−0.2039)	0.0619 (0.0711)	0.0537 (0.0362)	0.0221 (0.0035)	0.0274 (0.0554)
Site 4	0.3138 (0.3496)	0.5771 (0.6531)	−0.1305 (0.0196)	0.5376 (0.4265)	−0.4298 (−0.4288)	0.0449 (0.0663)	0.0210 (0.2255)	−0.2742 (−0.1726)
Site 5	0.0530 (0.0610)	0.1697 (0.2405)	−0.1186 (−0.1111)	−0.0605 (−0.1477)	−0.3588 (−0.3386)	−0.0701 (−0.1427)	−0.0608 (−0.6770)	0.9018 (0.5577)
Site 6	0.0287 (0.0296)	−0.1694 (−0.2440)	0.2720 (0.3053)	0.7576 (0.8133)	0.4674 (0.2518)	0.0284 (−0.0536)	0.0826 (−0.1889)	0.3106 (0.2881)
Site 7	0.1044 (0.1136)	0.6399 (0.5738)	−0.2564 (−0.0848)	−0.2271 (−0.0974)	0.6706 (0.7900)	−0.0129 (−0.0550)	0.0591 (−0.0439)	0.0943 (0.1097)
Site 8	0.0087 (0.0085)	−0.0147 (−0.0011)	−0.0858 (−0.0858)	0.0478 (0.0669)	0.0794 (0.0736)	0.5148 (0.8067)	−0.8477 (−0.4753)	0.0086 (−0.3256)

[a] Values in parenthesis are at 77 K.

Table S2. Standard deviations of the absorption spectra and the distributions of site energy of the individual sites, and the ratios between the standard deviations of spectra and distribution at 300 and 77 K. [a]

Site	1	2	3	4	5	6	7	8
Spectra (cm^{-1})	210 (100)	480 (290)	300 (190)	180 (70)	490 (290)	260 (80)	250 (110)	440 (310)
Distribution (cm^{-1})	350 (170)	560 (320)	350 (180)	330 (160)	540 (310)	390 (180)	350 (170)	510 (340)
Ratio	0.59 (0.57)	0.87 (0.89)	0.85 (1.08)	0.55 (0.43)	0.90 (0.94)	0.68 (0.41)	0.72 (0.63)	0.85 (0.91)

[a] Values in parenthesis are at 77 K.

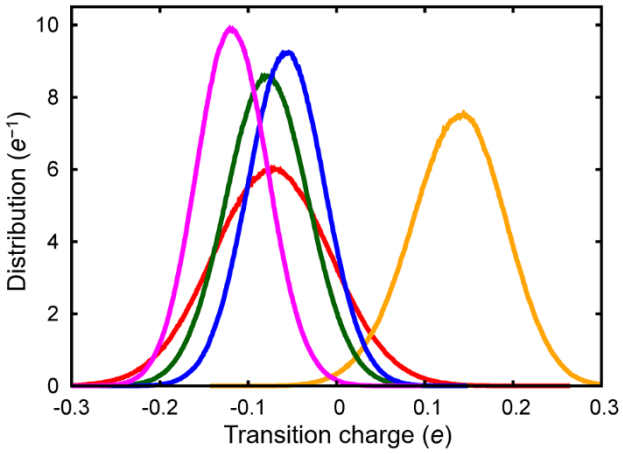

Figure S1. Distributions of the five transition charges with large standard deviations in BChl 8 at 300 K. The distributions of the transition charges of MG, ND, CHB, NB, and C1D (the notations are taken from 3EOJ.pdb file) are shown in red, orange, green, blue, and purple, respectively. The averages (standard deviations) of the transition charges of MG, ND, CHB, NB, and C1D are −0.073 (0.066), 0.137 (0.053), −0.079 (0.047), −0.057 (0.043), and −0.118 (0.040), respectively. The fluctuations of the transition charges for other atoms in BChl 8 are smaller than those of five atoms.

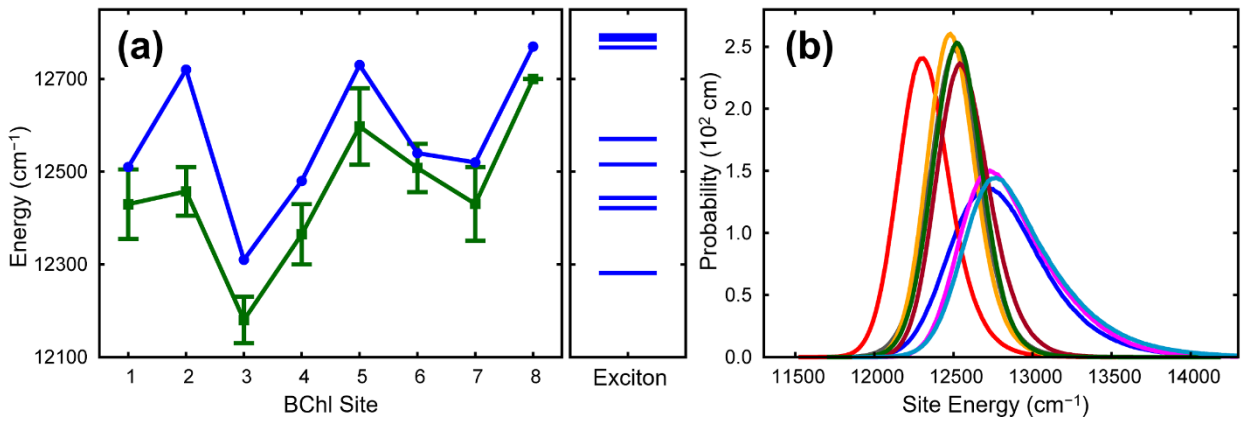

Figure S2. (a) Comparison of the calculated site energies at 77 K (blue) with the result fitted to experimental data (green). Exciton energy levels at 77 K are also plotted. (b) The site-energy distributions obtained from MD simulations at 77 K. The site-energy distributions at sites 1, 2, 3, 4, 5, 6, 7, and 8 are shown by gray, blue, red, orange, magenta, brown, green, and sky-blue lines, respectively.

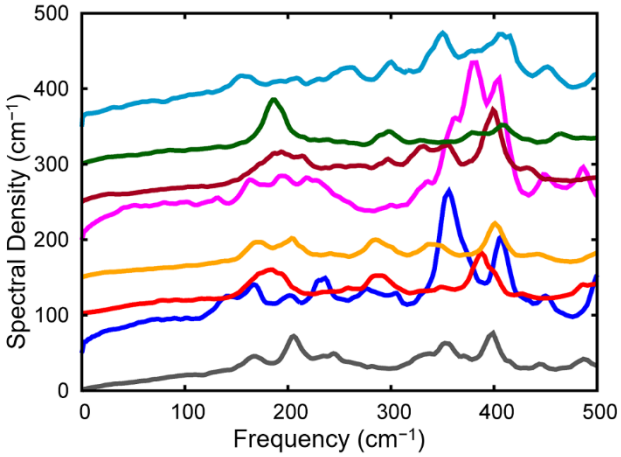

Figure S3. Calculated spectral densities of eight sites at 77 K. Spectral densities are arranged in order from BChl 1 to BChl 8 with an offset of 50 cm^{-1} for clarity. The spectral densities at sites 1, 2, 3, 4, 5, 6, 7, and 8 are shown by gray, blue, red, orange, magenta, brown, green, and sky-blue lines, respectively.

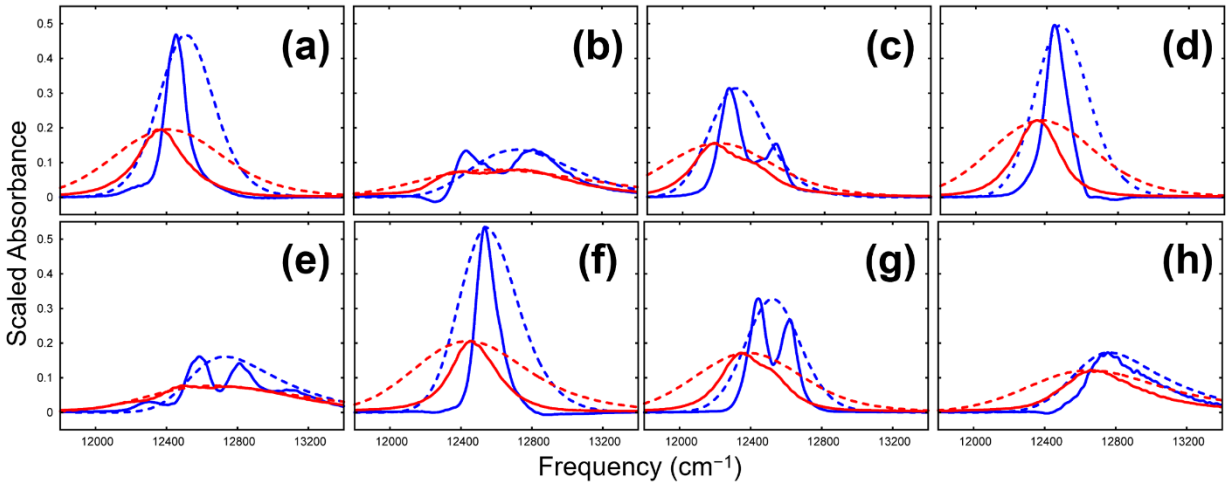

Figure S4. Comparison of the absorption spectrum (solid) with the distribution of site energy of each site (dashed) at 300 K (red) and 77 K (blue): (a) site 1, (b) site 2, (c) site 3, (d) site 4, (e) site 5, (f) site 6, (g) site 7, and (h) site 8.

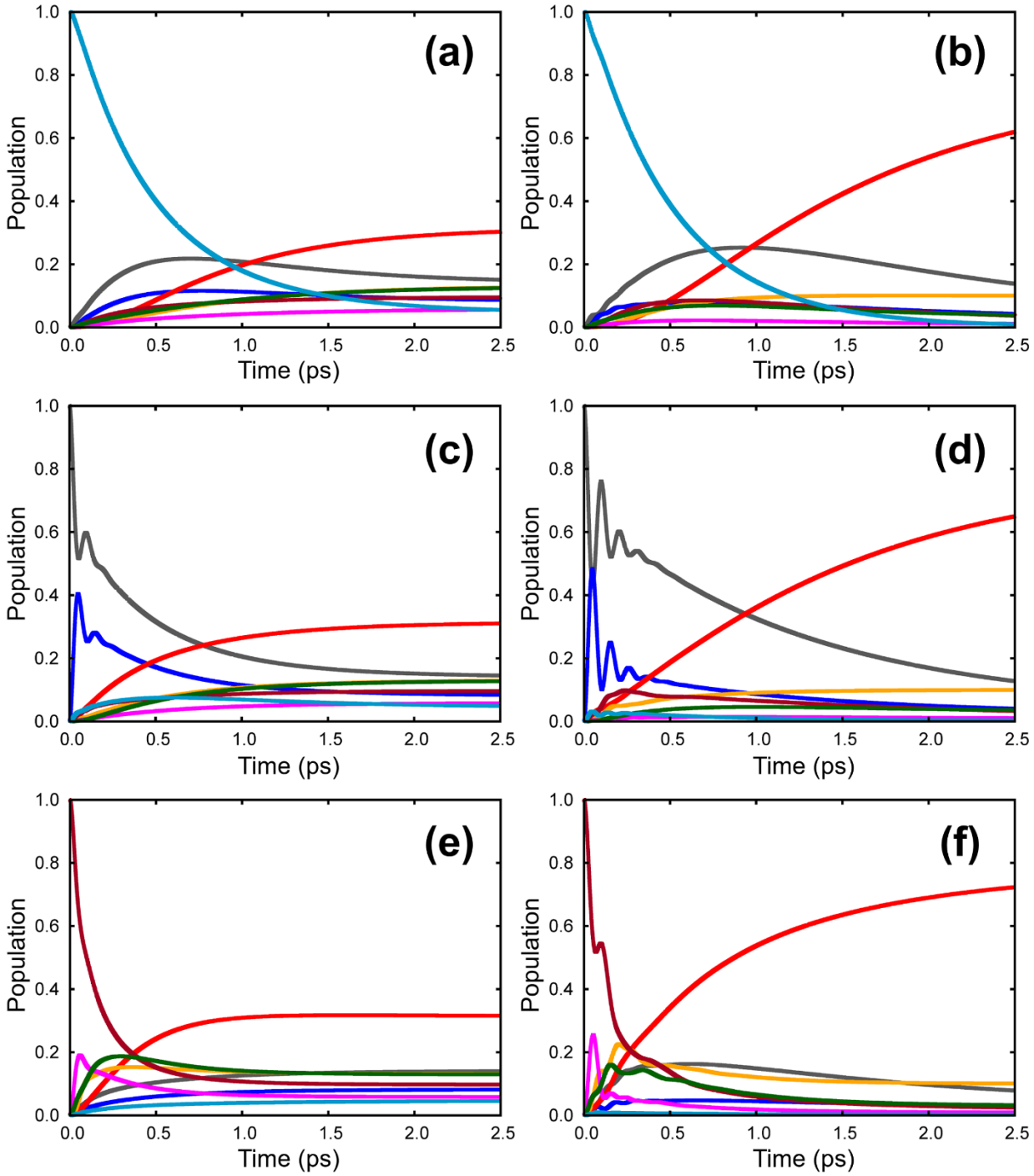

Figure S5. Population dynamics at (a) 300 K (the same as Figure 4a) and (b) 77 K after the initial excitation of site 8. Population dynamics at (c) 300 K and (d) 77 K after the initial excitation of site 1. Population dynamics at (e) 300 K and (f) 77 K after the initial excitation of site 6. The populations of sites 1, 2, 3, 4, 5, 6, 7, and 8 are shown by gray, blue, red, orange, magenta, brown, green, and sky-blue lines, respectively.

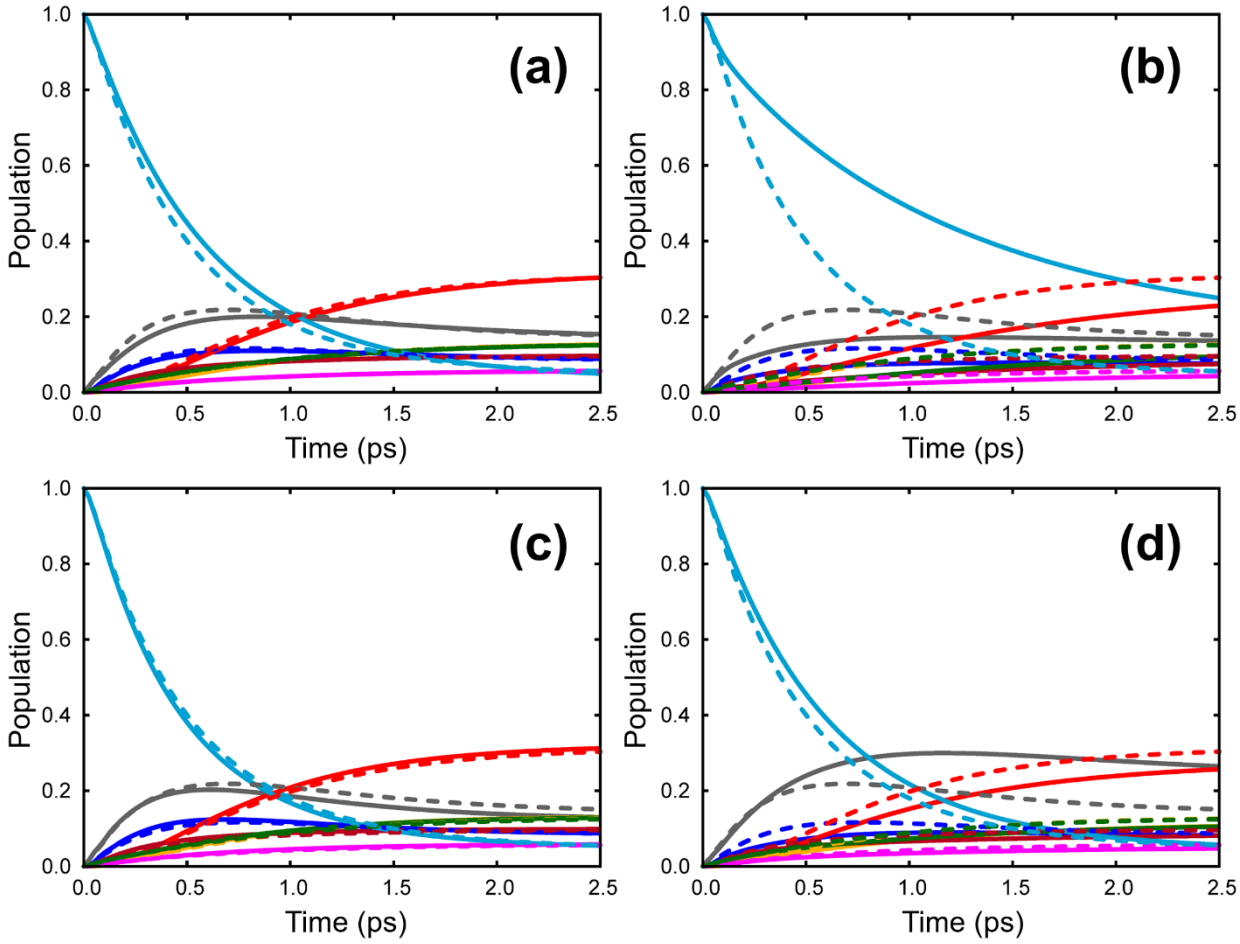

Figure S6. Population dynamics after the initial excitation of site 8 at 300 K calculated with the reorganization energy of (a) 65.5 cm^{-1} and (b) 393 cm^{-1} at site 8. In (a, b), the dynamics calculated with the original reorganization energy (131 cm^{-1}) are shown by the dashed line. Population dynamics after the initial excitation of site 8 at 300 K calculated with the reorganization energy of (c) 35 cm^{-1} and (d) 210 cm^{-1} at site 1. In (c, d), the dynamics calculated with the original reorganization energy (70 cm^{-1}) are shown by the dashed line. The populations of sites 1, 2, 3, 4, 5, 6, 7, and 8 are shown by gray, blue, red, orange, magenta, brown, green, and sky-blue lines, respectively.

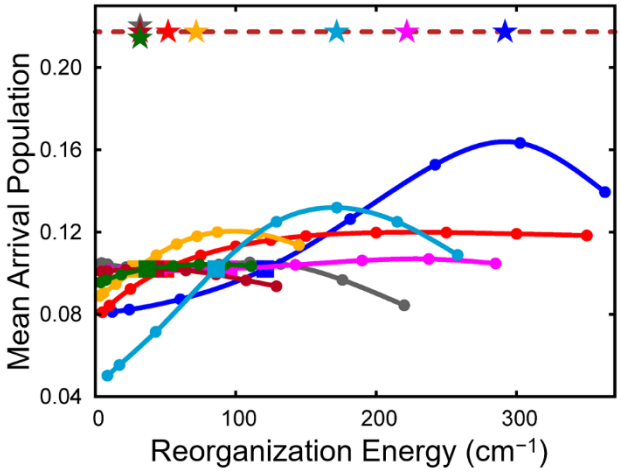

Figure S7. $\bar{\alpha}(t=1\text{ ps})$ with respect to reorganization energy of each site at 77 K. Squares correspond to the original reorganization energy at each site. The dashed is $\bar{\alpha}(t=1\text{ ps})$ (= 0.217) obtained with the optimized set of reorganization energies and the colored stars are the reorganization energy of each site in the optimized set at 77 K. $\bar{\alpha}(t=1\text{ ps})$ of sites 1, 2, 3, 4, 5, 6, 7, and 8 are shown by gray, blue, red, orange, magenta, brown, green, and sky-blue lines, respectively.