Electronic Supplementary Material (ESI) for Photochemical & Photobiological Sciences.

Supporting Information

Optimal Arrangements of 1,3-Diphenylisobenzofuran Molecule Pairs for Fast Singlet Fission

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1. Symmetrized Rotamer Geometries

Table S1. *XYZ* Coordinates for **1**(C_2) in units of Ångstrøm.

35

O	0.0000000	-0.6883432	0.0000000
C	-3.7022031	-2.5379216	0.5387490
C	3.7022031	-2.5379216	-0.5387490
C	-4.8406069	-1.9068328	0.0953780
C	0.7100903	3.8076831	-0.0000001
C	-4.7672583	-0.6295796	-0.4042319
C	-0.7100903	3.8076831	0.0000001
C	4.7672583	-0.6295796	0.4042319
C	-2.4848470	-1.8865396	0.5039176
C	4.8406069	-1.9068328	-0.0953780
C	-1.4240502	2.6580631	0.0000002
C	2.4848470	-1.8865396	-0.5039176
C	1.4240502	2.6580631	-0.0000002
C	0.7188995	1.4214726	-0.0000001
C	-3.5518787	0.0253856	-0.4579342
C	-2.3927734	-0.5930411	0.0000004
C	3.5518787	0.0253856	0.4579342
C	-1.1155413	0.1026812	0.0000002
C	2.3927734	-0.5930411	-0.0000004
C	-0.7188995	1.4214726	0.0000001
C	1.1155413	0.1026812	-0.0000002
H	-3.7565185	-3.5463635	0.9279490
H	3.7565185	-3.5463635	-0.9279490
H	-5.7935982	-2.4199217	0.1331728
H	1.2264639	4.7591286	-0.0000002
H	-5.6579643	-0.1352807	-0.7706559
H	-1.2264639	4.7591286	0.0000002
H	5.6579643	-0.1352807	0.7706559
H	-1.5942178	-2.3790651	0.8674921
H	5.7935982	-2.4199217	-0.1331728
H	-2.5036057	2.6774204	0.0000004
H	1.5942178	-2.3790651	-0.8674921
H	2.5036057	2.6774204	-0.0000004
H	-3.4997328	1.0128702	-0.8924179
H	3.4997328	1.0128702	0.8924179

Table S2. *XYZ* Coordinates for **1**(C_{2v}) in units of Ångstrøm.

35

O	0.0000000	-0.6883431	0.0000000
C	-3.6971879	-2.5379211	0.0000000
C	3.6971879	-2.5379211	0.0000000
C	-4.8395476	-1.9068321	0.0000000
C	0.7100611	3.8076829	0.0000000
C	-4.7707134	-0.6295791	0.0000000
C	-0.7100611	3.8076829	0.0000000
C	4.7707134	-0.6295791	0.0000000
C	-2.4801950	-1.8865391	0.0000000
C	4.8395476	-1.9068321	0.0000000
C	-1.4239920	2.6580629	0.0000000
C	2.4801950	-1.8865391	0.0000000
C	1.4239920	2.6580629	0.0000000
C	0.7188697	1.4214719	0.0000000
C	-3.5558678	0.0253849	0.0000000
C	-2.3926755	-0.5930411	0.0000000
C	3.5558678	0.0253849	0.0000000
C	-1.1154955	0.1026809	0.0000000
C	2.3926755	-0.5930411	0.0000000
C	-0.7188697	1.4214719	0.0000000
C	1.1154955	0.1026809	0.0000000
H	-3.7479867	-3.5463631	0.0000000
H	3.7479867	-3.5463631	0.0000000
H	-5.7921595	-2.4199211	0.0000000
H	1.2264130	4.7591279	0.0000000
H	-5.6646914	-0.1352801	0.0000000
H	-1.2264130	4.7591279	0.0000000
H	5.6646914	-0.1352801	0.0000000
H	-1.5863197	-2.3790651	0.0000000
H	5.7921595	-2.4199211	0.0000000
H	-2.5035030	2.6774199	0.0000000
H	1.5863197	-2.3790651	0.0000000
H	2.5035030	2.6774199	0.0000000
H	-3.5076467	1.0128699	0.0000000
H	3.5076467	1.0128699	0.0000000

Table S3. *XYZ* Coordinates for **1**(C_s) in units of Ångstrøm.

35

O	0.0000000	-0.6883430	0.0000000
C	-3.7022030	-2.5379210	0.5387490
C	3.7022030	-2.5379210	0.5387490
C	-4.8406060	-1.9068320	0.0953770
C	0.7100900	3.8076830	0.0000000
C	-4.7672580	-0.6295790	-0.4042310
C	-0.7100900	3.8076830	0.0000000
C	4.7672580	-0.6295790	-0.4042310
C	-2.4848460	-1.8865390	0.5039170
C	4.8406060	-1.9068320	0.0953770
C	-1.4240500	2.6580630	0.0000000
C	2.4848460	-1.8865390	0.5039170
C	1.4240500	2.6580630	0.0000000
C	0.7188990	1.4214720	0.0000000
C	-3.5518780	0.0253850	-0.4579340
C	-2.3927730	-0.5930410	0.0000000
C	3.5518780	0.0253850	-0.4579340
C	-1.1155410	0.1026810	0.0000000
C	2.3927730	-0.5930410	0.0000000
C	-0.7188990	1.4214720	0.0000000
C	1.1155410	0.1026810	0.0000000
H	-3.7565180	-3.5463630	0.9279490
H	3.7565180	-3.5463630	0.9279490
H	-5.7935980	-2.4199210	0.1331720
H	1.2264630	4.7591280	0.0000000
H	-5.6579640	-0.1352800	-0.7706550
H	-1.2264630	4.7591280	0.0000000
H	5.6579640	-0.1352800	-0.7706550
H	-1.5942170	-2.3790650	0.8674920
H	5.7935980	-2.4199210	0.1331720
H	-2.5036050	2.6774200	0.0000000
H	1.5942170	-2.3790650	0.8674920
H	2.5036050	2.6774200	0.0000000
H	-3.4997320	1.0128700	-0.8924170
H	3.4997320	1.0128700	-0.8924170

Table S4. *XYZ* Coordinates for **1**(C_1) in units of Ångstrøm.

35

O	0.0000000	-0.6883400	0.0000000
C	-3.7450000	-2.4501400	0.4546400
C	3.6971900	-2.5379200	0.0000000
C	-4.8388100	-1.9081900	-0.0070400
C	0.7100600	3.8076800	0.0000000
C	-4.7219400	-0.7191100	-0.4637000
C	-0.7100600	3.8076800	0.0000000
C	4.7707100	-0.6295800	0.0000000
C	-2.5284600	-1.7979400	0.4589000
C	4.8395500	-1.9068300	0.0000000
C	-1.4239900	2.6580600	0.0000000
C	2.4801900	-1.8865400	0.0000000
C	1.4239900	2.6580600	0.0000000
C	0.7188700	1.4214700	0.0000000
C	-3.5073600	-0.0636600	-0.4611900
C	-2.3926800	-0.5930400	0.0000000
C	3.5558700	0.0253800	0.0000000
C	-1.1155000	0.1026800	0.0000000
C	2.3926800	-0.5930400	0.0000000
C	-0.7188700	1.4214700	0.0000000
C	1.1155000	0.1026800	0.0000000
H	-3.8338000	-3.3888300	0.8159000
H	3.7479900	-3.5463600	0.0000000
H	-5.7911900	-2.4216900	-0.0091800
H	1.2264100	4.7591300	0.0000000
H	-5.5779100	-0.2946000	-0.8251500
H	-1.2264100	4.7591300	0.0000000
H	5.6646900	-0.1352800	0.0000000
H	-1.6725300	-2.2208100	0.8196700
H	5.7921600	-2.4199200	0.0000000
H	-2.5035000	2.6774200	0.0000000
H	1.5863200	-2.3790700	0.0000000
H	2.5035000	2.6774200	0.0000000
H	-3.4219000	0.8554600	-0.8152500
H	3.5076500	1.0128700	0.0000000

2. Generation of Pair Coordinates from Structure Parameters

The method of generation of the XYZ coordinates of each pair is described by the following. Partner A is always the same and its coordinates are obtained from Tables S1, S2, S3, or S4 for **1**(C_2), **1**(C_{2v}), **1**(C_s), and **1**(C_1). Partner B's coordinates are the obtained by rotation then translation of the coordinates of partner A according to eqs. 1. The *XYZ* coordinates are stored row-wise as in eq. (1a). The total rotation matrix is the product of the three rotations about the *x*, *y*, and *z* axes ($R_{tot} = R_z \cdot R_y \cdot R_x$) and it is determined by the three parameters R_x, R_y, and R_z as defined in eq. (1b). The translation matrix T is determined by the three parameters T_x, T_y, and T_z and its form is provided in eq. (1c). The *XYZ* coordinates of partner B store row-wise and are obtained through matrix multiplication of A with the Hermitian conjugate total rotation matrix R_{tot} followed by addition of the translation matrix T as shown in eq. (1d). There are as many rows in A, B, and T, as there are atoms in the chromophore, 35 for the case of 1,3-diphenylisobenzofuran.

$$A = \begin{pmatrix} x_1 & y_1 & z_1 \\ x_2 & y_2 & z_2 \\ \vdots & \vdots & \vdots \end{pmatrix}; B = \begin{pmatrix} x_1 & y_1 & z_1 \\ x_2 & y_2 & z_2 \\ \vdots & \vdots & \vdots \end{pmatrix} \tag{1a}$$

$$R_{tot} = \begin{pmatrix} Cos(Ry)Cos(Rz) & -Cos(Rz)Sin(Rx)Sin(Ry) - Cos(Rx)Sin(Rz) & -Cos(Rx)Cos(Rz)Sin(Ry) + Sin(Rx)Sin(Rz) \\ Cos(Ry)Sin(Rz) & Cos(Rx)Cos(Rz) - Sin(Rx)Sin(Ry)Sin(Rz) & -Cos(Rz)Sin(Rx) - Cos(Rx)Sin(Ry)Sin(Rz) \\ Sin(Ry) & Cos(Ry)Sin(Rx) & Cos(Rx)Cos(Ry) \end{pmatrix} \tag{1b}$$

$$T = \begin{pmatrix} Tx & Ty & Tz \\ Tx & Ty & Tz \\ \vdots & \vdots & \vdots \end{pmatrix} \tag{1c}$$

$$B = A \cdot R_{tot}^{\dagger} + T \tag{1d}$$

3. Top 67 Pair Structures for 4 Rotamers of 1,3-Diphenylisobenzofuran

Table S5. Pair Structure Parameters for **1**(C_2) Pairs 1-34.[a]

Pair	T_Z	T_Y	T_X	R_Z	R_Y	R_X	ΔE_{DS}[b]	ΔE_{BB}[c]	k/k_0[d]
1	3.49	1.45	2.11	68.08	179.60	0.58	51.3	34.7	4306
2	4.00	-0.39	-2.00	248.95	193.73	160.11	42.5	37.6	2944
3	3.93	1.77	-1.89	107.84	174.72	172.76	27.4	24.6	2261
4	4.28	-1.27	-1.42	81.89	165.69	13.89	25.4	30.1	896
5	4.00	1.65	-2.57	127.80	-4.37	188.90	51.4	26.8	892
6	3.98	-0.09	-2.25	79.24	168.15	17.30	4.3	29.0	806
7	3.40	-2.59	4.71	63.41	202.54	0.07	5.4	23.6	717
8	3.80	5.27	0.17	96.43	9.33	197.91	9.3	20.8	546
9	3.55	-2.17	4.46	101.22	199.87	14.64	3.4	20.3	536
10	3.94	3.51	2.33	-2.45	193.37	176.54	143.9	23.9	504
11	3.72	5.46	0.29	102.04	10.42	201.27	27.4	22.6	499
12	3.03	-0.23	-5.84	101.40	-22.06	174.04	18.7	22.3	494
13	3.70	1.35	2.50	50.42	178.00	4.24	90.7	27.2	441
14	3.27	4.94	1.52	78.06	169.03	2.29	60.0	16.0	418
15	3.08	5.82	0.13	88.62	168.62	-2.85	34.5	13.5	354
16	3.40	1.43	-4.86	89.72	-0.80	179.33	125.5	20.2	338
17	3.09	5.79	0.32	263.76	191.48	187.52	32.8	12.9	336
18	3.11	-2.75	3.35	121.96	192.22	201.84	54.1	6.3	316
19	3.39	4.64	-1.98	128.21	-5.17	186.26	3.4	12.0	304
20	3.60	3.22	0.21	95.68	171.33	176.50	22.2	14.4	259
21	3.71	2.98	-0.61	117.31	176.13	10.27	129.0	12.6	250
22	3.92	0.15	-4.49	108.85	-0.07	187.25	55.7	11.5	238
23	3.95	-0.05	3.76	43.51	186.18	7.09	2.2	12.2	239
24	4.21	0.33	-3.92	131.36	1.67	191.97	22.0	9.6	226
25	2.91	3.26	-3.60	146.67	338.63	181.52	0.7	9.8	204
26	4.43	-0.40	-3.57	141.15	192.58	166.69	3.6	9.6	201
27	3.74	1.98	-1.33	103.40	170.46	13.60	29.3	10.0	187
28	3.10	2.53	-3.56	12.82	191.75	193.27	152.4	16.9	182
29	2.96	1.76	-3.65	168.68	165.20	-13.35	134.9	7.8	174
30	4.18	-0.05	4.28	19.72	11.87	192.64	18.9	6.9	168
31	4.31	-0.06	4.23	20.86	15.07	196.68	14.6	5.8	163
32	4.30	-0.06	4.24	20.76	14.65	196.14	15.2	5.9	163
33	3.78	0.06	2.78	269.66	180.29	174.91	4.3	12.6	138
34	4.41	-2.42	-2.24	261.87	195.38	167.53	69.6	10.8	133

[a] Translations in units of Ångstrøm, rotations in units of degree, and energies in units of meV. [b] Davydov splitting $\Delta E_{DS} = E(S^{**}) - E(S^{*})$. [c] Biexciton binding energy. [d] $k_0 = 1.05\times 10^8\ s^{-1}$.

Table S6. Pair Structure Parameters for **1**(C_2) Pairs 35-67.[a]

Pair	T_Z	T_Y	T_X	R_Z	R_Y	R_X	ΔE_{DS}[b]	ΔE_{BB}[c]	k/k_0[d]
35	3.95	-0.98	-2.60	126.74	179.26	172.16	143.7	16.8	132
36	2.95	2.05	-4.56	2.22	14.85	167.49	55.0	9.8	126
37	2.56	-3.66	2.48	108.36	21.51	164.67	6.0	8.4	125
38	3.74	-0.16	2.67	125.01	-0.93	-0.42	118.8	8.3	121
39	3.34	-0.41	-4.62	1.99	8.64	158.98	52.8	8.7	114
40	2.69	-3.49	2.63	110.91	199.42	198.29	8.3	7.2	113
41	4.34	0.86	3.42	-6.25	194.89	166.03	73.9	7.8	110
42	3.60	1.79	2.27	276.56	187.26	179.25	74.3	10.0	109
43	4.06	2.47	0.96	284.59	181.04	167.16	108.5	13.1	104
44	3.97	2.51	1.02	103.85	358.96	-11.17	109.2	13.1	103
45	3.33	-2.36	3.75	167.99	176.37	23.25	90.8	3.9	101
46	3.16	4.72	-2.79	16.97	192.14	182.81	172.9	8.8	97
47	4.24	-0.54	4.23	141.59	5.92	335.42	51.4	7.1	96
48	3.60	5.26	0.55	30.96	188.18	180.40	153.9	11.1	95
49	3.72	-0.79	3.03	58.57	193.24	2.00	53.5	9.1	94
50	4.38	-0.76	-2.08	87.43	-16.82	-17.58	61.8	18.2	92
51	3.61	5.26	0.56	25.20	190.05	180.89	161.2	10.0	93
52	3.26	3.88	-3.88	36.32	185.06	182.56	146.0	11.7	93
53	3.49	3.03	-0.96	64.96	179.99	181.83	229.5	20.0	91
54	4.08	-0.64	-1.68	72.83	-3.20	188.71	82.3	6.4	84
55	4.00	-0.75	-1.98	68.19	178.19	170.26	97.4	4.3	82
56	3.85	0.93	2.57	263.45	180.84	173.98	24.5	4.9	76
57	3.64	2.22	-4.16	150.87	-7.58	189.38	17.5	4.2	76
58	3.46	-0.32	-3.91	201.90	177.26	164.61	103.1	4.5	75
59	4.51	1.01	1.84	84.91	2.71	343.28	11.5	4.5	72
60	4.23	-2.07	-3.70	192.41	174.20	155.49	98.5	7.2	71
61	4.20	2.35	1.34	274.50	191.66	169.21	34.8	3.1	70
62	4.14	2.43	1.18	274.72	189.80	169.08	38.8	3.3	69
63	4.29	6.14	3.04	52.02	199.77	151.00	12.2	4.7	63
64	3.73	-0.67	1.95	108.60	183.77	201.42	98.5	8.7	60
65	3.10	7.34	-2.33	1.89	6.57	176.58	84.2	3.9	59
66	4.58	-0.84	1.17	120.84	-14.53	-6.31	97.6	9.9	58
67	3.50	2.98	1.48	251.52	188.47	184.04	14.1	4.7	57

[a] Translations in units of Ångstrøm, rotations in units of degree, and energies in units of meV. [b] Davydov splitting $\Delta E_{DS} = E(S^{**}) - E(S^{*})$. [c] Biexciton binding energy. [d] $k_0 = 1.05\times 10^8\ s^{-1}$.

Table S7. Pair Structure Parameters for **1**(C_{2v}) Pairs 1-34.[a]

Pair	T_Z	T_Y	T_X	R_Z	R_Y	R_X	ΔE_{DS}[b]	ΔE_{BB}[c]	k/k_0[d]
1	3.46	1.28	2.00	64.92	-0.17	-0.26	78.7	75.3	16422
2	3.49	1.19	2.56	48.77	-0.42	-0.93	69.3	54.4	2273
3	3.50	-1.10	1.48	104.39	-0.69	180.89	8.2	34.4	1927
4	3.48	1.35	0.94	82.36	0.09	180.44	16.1	27.8	1625
5	3.46	3.33	2.78	-2.83	-0.33	180.15	147.6	42.7	1539
6	3.38	5.42	0.20	86.56	-0.20	-2.13	35.3	28.0	1469
7	3.36	-0.28	4.12	40.40	-2.50	0.71	10.8	25.0	1419
8	3.44	1.10	4.81	86.41	-1.38	-0.02	96.2	28.3	988
9	3.43	-2.05	0.71	90.83	1.08	180.13	67.1	28.3	772
10	3.45	2.79	0.06	93.81	-0.56	179.63	34.3	23.1	714
11	3.33	4.65	1.88	74.78	-2.40	178.71	34.7	19.9	598
12	3.48	1.35	2.63	123.30	-0.19	0.25	208.2	30.2	494
13	3.47	1.25	3.75	0.00	0.00	180.00	79.8	9.2	461
14	3.38	4.17	2.40	41.27	-1.31	-0.80	20.5	11.8	398
15	3.41	4.30	2.17	50.44	359.31	-0.68	5.4	9.7	380
16	3.44	2.67	3.09	4.88	-0.50	180.10	109.4	19.9	377
17	3.49	-1.17	2.84	54.45	0.39	-0.47	176.8	28.1	332
18	3.44	3.04	1.69	32.35	-0.24	180.14	172.1	19.9	252
19	3.48	-1.66	4.00	158.61	-0.63	-0.64	12.4	10.6	243
20	3.49	2.62	0.89	107.35	0.11	-0.19	181.4	23.7	238
21	3.48	0.11	2.80	152.45	0.09	-0.33	157.8	24.3	236
22	3.47	-1.44	4.08	0.00	0.00	180.00	65.4	6.3	212
23	3.49	1.28	3.02	37.09	0.40	179.96	77.5	13.6	212
24	3.48	-1.51	4.06	137.76	-0.60	181.55	7.8	3.6	201
25	3.46	-0.28	2.88	168.99	0.01	179.93	153.5	22.4	198
26	3.43	1.50	3.43	29.34	-0.53	-0.09	18.1	10.6	193
27	3.63	2.48	2.07	106.00	167.76	-2.43	108.0	14.6	132
28	3.50	-0.59	3.36	154.93	-1.12	185.05	100.1	9.2	117
29	3.51	1.48	2.13	273.30	3.10	175.34	76.1	6.8	113
30	3.48	-2.59	3.34	124.49	0.49	179.40	24.2	6.2	113
31	3.86	3.11	1.30	87.66	158.33	-0.94	11.4	6.5	111
32	3.23	4.63	2.73	-18.19	-9.59	177.66	193.2	9.7	107
33	3.46	-1.54	3.03	85.96	0.52	178.87	114.0	11.9	106
34	4.46	1.82	1.22	97.57	180.20	17.96	56.9	7.4	102

[a] Translations in units of Ångstrøm, rotations in units of degree, and energies in units of meV. [b] Davydov splitting $\Delta E_{DS} = E(S^{**}) - E(S^{*})$. [c] Biexciton binding energy. [d] $k_0 = 1.05\times 10^8\ s^{-1}$.

Table S8. Pair Structure Parameters for **1**(C_{2v}) Pairs 35-67.[a]

Pair	T_Z	T_Y	T_X	R_Z	R_Y	R_X	ΔE_{DS}[b]	ΔE_{BB}[c]	k/k_0[d]
35	4.30	2.63	1.29	98.46	165.87	12.20	42.6	4.4	101
36	3.08	7.25	2.67	173.24	8.19	3.28	88.6	5.6	95
37	3.44	7.25	2.01	0.00	0.00	180.00	89.8	5.2	94
38	3.60	7.27	1.68	3.10	4.21	181.52	89.5	5.3	94
39	3.90	0.46	2.63	77.13	180.30	6.51	16.2	5.1	88
40	4.58	6.87	2.85	144.65	-0.32	-17.44	86.9	5.9	66
41	3.54	-2.95	2.05	153.30	-0.88	181.79	173.1	9.4	62
42	2.63	3.99	5.58	85.81	-10.77	8.91	157.4	7.3	60
43	3.36	8.37	-0.75	180.06	180.01	0.05	76.1	4.0	60
44	4.23	1.52	4.21	136.45	176.24	11.84	52.7	6.2	59
45	3.77	5.91	3.63	99.69	1.16	-6.03	131.9	6.9	56
46	4.61	-2.34	0.10	61.60	-19.25	-14.82	151.6	7.0	50
47	3.98	-2.62	0.61	77.21	-8.75	-6.88	124.3	5.7	48
48	3.21	6.53	3.61	0.00	0.00	180.00	66.0	1.8	35
49	2.85	7.02	2.34	152.90	-0.20	8.97	102.6	9.2	32
50	4.27	6.21	3.09	56.37	13.24	189.08	26.0	3.4	32
51	4.29	-2.50	0.80	134.09	168.22	169.22	174.8	5.3	32
52	3.49	0.48	4.46	149.54	0.10	-0.67	0.4	4.2	31
53	2.58	7.11	2.41	98.83	158.78	0.55	82.1	4.6	28
54	3.84	-3.74	1.19	121.40	174.62	174.50	85.8	6.3	27
55	3.99	3.55	4.27	46.86	172.74	15.27	54.1	3.7	25
56	3.47	0.73	3.68	123.49	-0.43	183.17	97.8	2.8	24
57	3.61	3.97	0.54	38.31	-4.45	184.90	193.5	8.8	20
58	3.27	3.04	2.80	144.74	2.99	191.60	226.8	7.4	16
59	3.90	-3.65	1.49	163.01	-5.83	186.27	197.2	6.5	12
60	3.48	-1.26	2.72	40.94	0.02	179.85	218.9	5.5	9
61	4.24	1.75	3.61	54.04	179.84	11.97	20.6	1.6	7
62	3.54	-3.64	2.58	43.28	-1.16	-0.78	225.7	8.6	4
63	3.15	4.89	1.88	59.03	151.61	166.60	18.8	1.0	4
64	3.50	3.44	0.54	0.00	0.00	180.00	296.0	1.9	2
65	4.92	-1.16	0.93	156.54	22.18	202.06	253.2	2.7	1
66	4.67	-0.16	0.41	152.39	17.10	197.31	288.8	9.8	7
67	3.50	-0.39	0.83	16.63	-0.69	177.48	473.3	9.2	1

[a] Translations in units of Ångstrøm, rotations in units of degree, and energies in units of meV. [b] Davydov splitting $\Delta E_{DS} = E(S^{**}) - E(S^{*})$. [c] Biexciton binding energy. [d] $k_0 = 1.05 \times 10^8$ s^{-1}.

Table S9. Pair Structure Parameters for **1**(C_s) Pairs 1-34.[a]

Pair	T_Z	T_Y	T_X	R_Z	R_Y	R_X	ΔE_{DS}[b]	ΔE_{BB}[c]	k/k_0[d]
1	-3.46	1.30	1.93	66.79	180.40	-0.61	70.5	45.9	5998
2	3.53	1.55	1.73	242.59	183.68	179.27	93.6	46.2	3473
3	4.00	-0.39	1.98	290.89	193.66	19.73	44.8	36.8	3126
4	4.02	1.71	1.82	71.41	177.99	9.33	32.2	22.3	1805
5	3.97	-0.11	2.31	279.25	191.79	16.73	15.3	29.5	901
6	3.60	1.45	-2.58	-50.37	3.23	-2.59	63.4	33.2	808
7	-3.38	-2.61	4.69	63.24	157.38	0.08	4.4	23.2	766
8	3.20	5.46	1.03	247.35	190.03	199.86	14.3	23.2	724
9	4.00	1.66	2.58	52.28	175.63	8.89	54.2	25.6	691
10	-3.63	1.31	-2.50	129.39	1.53	176.77	80.7	29.8	614
11	3.44	3.79	2.73	0.00	0.00	180.00	141.5	25.3	580
12	3.22	4.75	1.91	73.79	169.29	202.84	0.2	19.6	542
13	3.06	0.08	5.62	261.46	198.25	186.61	4.8	20.8	533
14	3.60	1.96	1.44	79.09	-4.39	191.52	19.2	11.3	523
15	3.80	5.28	0.15	96.15	9.29	197.67	13.0	20.3	514
16	-3.58	-2.06	4.39	287.04	18.88	163.29	0.2	19.6	513
17	3.53	2.24	1.79	69.62	-3.07	188.99	48.6	11.5	493
18	3.11	3.80	3.04	1.64	166.47	182.99	145.8	23.5	490
19	-3.75	5.35	-0.19	76.47	-10.48	20.36	30.0	22.6	470
20	-3.76	5.33	-0.18	257.01	190.30	199.99	25.9	22.4	469
21	3.71	5.49	-0.28	77.65	190.42	21.51	27.4	22.4	459
22	3.73	5.44	0.27	100.71	370.09	200.66	17.0	22.0	451
23	3.07	5.84	-0.15	91.43	168.66	184.34	39.1	14.7	440
24	3.11	5.78	0.29	266.35	190.91	187.92	43.0	14.8	433
25	-4.17	0.62	4.43	258.10	-0.01	168.81	62.5	16.0	419
26	3.27	4.95	1.53	256.32	191.05	177.42	57.1	15.4	416
27	3.21	3.13	3.20	223.95	190.67	180.21	19.2	15.9	414
28	-3.27	-1.26	1.69	98.35	169.38	1.06	26.0	13.7	370
29	-4.16	-0.24	5.09	268.48	3.31	168.72	35.5	13.5	362
30	-3.46	0.98	-0.07	87.96	174.54	11.11	8.3	17.8	353
31	-4.11	-0.83	5.04	84.18	171.57	-10.83	33.4	12.7	349
32	3.41	1.43	4.86	90.20	179.20	-0.63	124.8	19.6	316
33	-2.93	-2.03	0.90	278.11	199.53	11.77	17.3	16.6	315
34	2.90	0.47	5.38	291.12	15.54	166.12	23.9	16.1	307

[a] Translations in units of Ångstrøm, rotations in units of degree, and energies in units of meV. [b] Davydov splitting $\Delta E_{DS} = E(S^{**}) - E(S^{*})$. [c] Biexciton binding energy. [d] $k_0 = 1.05 \times 10^8$ s^{-1}.

Table S10. Pair Structure Parameters for **1**(C_s) Pairs 35-67.[a]

Pair	T_Z	T_Y	T_X	R_Z	R_Y	R_X	ΔE_{DS}[b]	ΔE_{BB}[c]	k/k_0[d]
35	-3.30	-1.15	0.73	275.61	188.26	9.10	8.2	19.1	285
36	-3.12	-2.72	3.45	124.23	-12.07	202.59	49.8	6.4	284
37	3.56	1.49	1.55	98.12	-0.33	193.66	128.8	24.5	273
38	-3.04	-1.50	0.07	89.41	167.50	10.08	6.7	14.3	271
39	3.36	4.65	1.86	57.63	173.78	4.43	12.3	10.8	267
40	3.47	1.22	2.88	114.16	174.33	-2.63	144.2	17.0	259
41	3.92	-0.07	2.71	86.54	172.27	5.95	32.6	11.2	260
42	-3.94	-0.07	3.74	223.22	6.24	172.99	0.7	11.5	240
43	3.50	1.96	2.33	111.63	178.16	0.44	207.6	27.5	231
44	-3.59	3.09	0.06	97.40	187.06	182.56	30.0	11.6	230
45	3.93	0.11	4.48	70.51	180.01	7.40	51.2	11.1	225
46	4.21	0.34	3.95	49.03	181.57	11.91	19.9	9.9	225
47	3.51	2.90	0.03	94.88	175.27	184.43	19.4	16.4	225
48	2.89	3.29	3.63	33.59	158.18	1.40	4.6	10.1	200
49	3.60	3.16	1.00	56.90	173.88	170.84	133.9	10.8	197
50	-4.15	-0.10	4.26	22.52	168.35	192.30	18.0	8.4	197
51	4.45	-0.43	3.57	38.74	193.12	13.42	0.7	9.8	198
52	4.61	1.61	3.56	0.51	15.34	187.17	62.2	7.4	195
53	3.57	3.04	0.44	79.52	-3.23	186.61	58.4	11.4	189
54	2.95	1.76	3.22	17.98	163.39	193.11	168.9	12.6	185
55	3.30	3.24	1.70	60.86	-8.24	9.63	28.3	13.8	184
56	-3.22	-2.63	0.31	80.82	163.21	182.02	74.1	14.4	184
57	2.95	1.77	3.66	11.47	165.00	193.41	135.4	7.9	180
58	4.23	-0.22	2.32	98.76	174.64	162.08	17.0	10.4	173
59	-3.93	1.58	1.25	76.81	181.24	180.50	57.1	7.2	171
60	3.87	-1.26	4.51	219.23	186.83	171.71	47.6	12.4	169
61	3.86	-1.08	1.16	98.56	182.06	201.14	45.2	8.7	162
62	3.11	2.52	3.55	167.54	11.46	13.12	154.5	16.0	162
63	-3.67	-0.29	2.61	267.64	3.17	176.29	4.9	10.6	158
64	3.86	-0.90	1.18	98.04	181.66	195.69	39.8	9.5	157
65	3.76	2.61	1.11	90.81	4.34	198.15	77.5	7.1	156
66	3.98	1.84	4.02	0.00	0.00	180.00	70.6	5.9	155
67	-2.53	-2.76	0.22	83.85	336.55	186.79	67.4	9.4	154

[a] Translations in units of Ångstrøm, rotations in units of degree, and energies in units of meV. [b] Davydov splitting $\Delta E_{DS} = E(S^{**}) - E(S^{*})$. [c] Biexciton binding energy. [d] $k_0 = 1.05 \times 10^8\ s^{-1}$.

Table S11. Pair Structure Parameters for **1**(C_1) Pairs 1-34.[a]

Pair	T_Z	T_Y	T_X	R_Z	R_Y	R_X	ΔE_{DS}[b]	ΔE_{BB}[c]	k/k_0[d]
1	-3.44	1.27	1.98	64.80	180.20	-0.31	76.1	83.0	22252
2	3.45	1.25	2.00	63.74	179.86	0.22	83.0	81.6	20037
3	3.44	1.28	2.04	64.45	-0.08	-0.10	87.1	75.6	16130
4	-3.45	1.28	-2.02	115.52	0.09	179.85	99.5	72.4	13025
5	3.67	1.65	-2.05	109.51	178.55	176.21	49.3	49.4	9267
6	3.94	1.85	-2.01	106.24	-1.80	188.05	42.0	33.1	4286
7	-3.47	1.20	2.55	49.08	180.48	-1.04	69.5	60.7	3148
8	3.47	1.16	2.53	48.00	179.58	0.94	78.3	60.4	2824
9	3.48	-1.10	1.44	102.58	-0.88	181.10	10.9	42.5	2724
10	-3.48	-1.11	1.45	102.66	0.90	178.87	11.7	43.5	2506
11	-3.51	1.20	-2.47	130.70	0.78	178.31	93.1	55.5	2284
12	3.68	1.36	-2.46	130.21	177.46	175.70	85.1	53.1	2196
13	-3.50	1.33	-2.60	-51.65	-0.44	1.57	57.1	51.2	2049
14	-3.39	0.00	5.40	86.64	182.09	-0.30	33.5	31.1	1891
15	3.35	-0.30	4.11	40.27	177.67	-0.62	9.8	28.4	1855
16	-3.45	1.37	0.79	80.62	-0.32	179.51	0.5	32.5	1852
17	3.37	5.42	0.20	86.65	179.82	2.17	34.7	30.7	1818
18	3.36	5.43	-0.18	92.91	179.81	177.60	46.5	31.4	1801
19	3.78	-0.39	-2.11	60.73	168.29	19.85	8.0	26.6	1778
20	-3.52	2.82	2.81	40.83	177.39	-1.19	13.2	26.6	1673
21	3.45	3.43	2.68	1.49	180.17	180.07	161.0	47.5	1658
22	-3.35	5.44	-0.29	98.55	180.99	182.25	33.1	29.1	1496
23	-3.58	1.30	1.18	77.39	174.72	180.94	27.8	24.1	1433
24	3.94	1.66	-2.60	127.14	-4.13	188.29	62.1	34.7	1434
25	-3.42	1.08	4.83	86.39	181.58	0.02	94.0	32.7	1334
26	3.43	1.08	4.83	86.30	178.42	-0.01	93.5	31.9	1265
27	3.38	3.88	-2.58	170.99	179.02	-0.61	128.1	33.8	1291
28	3.53	3.73	2.63	2.05	2.47	181.11	131.0	33.2	1291
29	3.53	3.66	2.74	-0.77	2.29	181.17	131.1	33.1	1287
30	-3.43	1.08	4.86	86.58	1.81	0.01	99.4	31.3	1174
31	-3.41	-2.00	0.75	89.97	-1.12	179.96	55.8	34.7	1066
32	3.47	-0.66	2.12	92.21	-0.48	179.48	36.0	27.8	982
33	3.43	-0.59	3.84	215.46	181.37	179.53	21.2	23.6	968
34	3.88	-0.47	1.69	96.50	166.68	2.33	14.5	22.7	903

[a] Translations in units of Ångstrøm, rotations in units of degree, and energies in units of meV. [b] Davydov splitting $\Delta E_{DS} = E(S^{**}) - E(S^{*})$. [c] Biexciton binding energy. [d] $k_0 = 1.05\times 10^8$ s^{-1}.

Table S12. Pair Structure Parameters for **1**(C_1) Pai rs 35-67.[a]

Pair	T_Z	T_Y	T_X	R_Z	R_Y	R_X	ΔE_{DS}[b]	ΔE_{BB}[c]	k/k_0[d]
35	-3.69	-0.32	3.96	220.32	180.31	184.24	6.8	19.7	832
36	-3.38	1.66	1.79	55.79	3.27	180.45	100.2	25.6	832
37	-3.50	-0.60	-2.14	57.52	1.18	177.52	96.0	24.2	826
38	-3.31	4.66	1.84	75.89	182.40	178.82	27.8	21.9	795
39	-4.00	3.42	-2.33	2.47	194.96	184.77	103.5	27.7	823
40	3.31	4.66	-1.87	105.03	177.57	-1.26	34.8	21.7	787
41	-4.00	-0.84	1.54	109.23	190.55	190.80	19.2	20.0	778
42	3.53	1.76	1.39	76.82	177.01	177.22	28.3	17.6	767
43	3.31	4.64	1.91	73.95	177.53	181.37	24.1	21.6	758
44	-3.53	4.75	-1.18	95.80	181.58	184.60	74.7	23.9	753
45	-3.32	4.64	-1.89	105.80	182.43	1.28	34.3	21.9	722
46	3.88	-0.36	-1.35	264.65	187.29	5.38	17.0	22.2	712
47	-3.56	-1.23	-1.05	79.10	-0.85	-6.12	5.4	20.9	646
48	-3.68	-2.07	-4.89	109.03	159.68	184.49	9.0	20.8	647
49	3.44	2.82	-0.07	86.23	179.64	-0.16	22.1	24.7	641
50	3.36	4.11	-2.47	143.23	178.90	178.81	22.1	15.6	637
51	3.27	4.92	1.52	257.98	190.88	177.41	43.1	18.7	625
52	-3.53	-2.50	-4.69	112.75	337.52	176.73	2.7	21.7	621
53	3.58	1.90	-1.51	108.26	182.68	2.24	44.0	12.3	616
54	3.53	-0.67	2.31	106.29	178.18	-3.05	42.4	11.6	610
55	-3.27	4.92	-1.51	102.17	10.86	177.34	50.7	18.7	587
56	-3.53	4.65	-1.23	98.43	6.31	173.07	43.5	17.9	581
57	-4.03	0.88	-4.38	99.74	3.59	171.11	47.8	18.2	578
58	3.45	1.20	-4.78	92.83	-1.29	180.29	85.6	21.4	586
59	-3.98	-0.19	-3.75	138.15	171.41	187.35	12.4	18.9	566
60	-3.53	-0.44	2.69	85.49	177.12	-1.76	20.9	23.8	552
61	-3.44	2.80	0.05	95.72	180.49	179.95	47.1	20.6	549
62	-3.27	5.52	0.96	66.46	8.88	173.20	32.1	15.3	545
63	-3.36	4.11	-2.46	142.70	181.18	181.14	24.0	15.0	547
64	3.27	5.52	-0.92	293.43	188.91	173.56	22.5	14.9	534
65	3.37	0.59	2.94	104.91	177.59	-2.20	94.4	20.3	551
66	-3.47	-1.32	4.54	45.08	179.37	-1.14	10.5	12.4	532
67	-3.46	-1.07	4.63	229.92	0.29	179.17	3.1	11.4	527

[a] Translations in units of Ångstrøm, rotations in units of degree, and energies in units of meV. [b] Davydov splitting $\Delta E_{DS} = E(S^{**}) - E(S^{*})$. [c] Biexciton binding energy. [d] $k_0 = 1.05\times 10^8\ s^{-1}$.

4. Optimized Geometries for Reorganization Energy Calculations

Table S13. *XYZ* Coordinates for Optimized Ground State of **1** in units of Ångstrøm.

35

O	0.0297399	0.0100260	-0.0302392
C	-0.3181500	-4.1172971	-0.4161545
C	3.8798596	2.4230274	0.0355755
C	0.7299928	-4.9369186	-0.0197967
C	3.6485269	0.0053403	-0.0246503
C	4.4380495	1.1147193	-0.0093507
C	-0.1745180	-2.7419799	-0.4109166
C	1.9236312	-4.3620133	0.3933202
C	2.5329507	2.6217906	0.0250725
C	2.0721871	-2.9863464	0.4075434
C	1.6762903	1.4932971	-0.0203652
C	1.1705131	-0.7118144	-0.0029452
C	2.2403393	0.1685874	-0.0048747
C	1.0280344	-2.1503645	-0.0037252
C	0.3004609	1.3322136	-0.0450977
C	-1.7881501	4.4174047	-0.4416432
C	-3.0342330	3.9627609	-0.0341553
C	-3.1819333	2.6372899	0.3521195
C	-2.0975424	1.7791392	0.3467747
C	-0.8326002	2.2295993	-0.0515671
C	-0.7004608	3.5622608	-0.4574416
H	-1.2563005	-4.5546342	-0.7385777
H	4.5472663	3.2761317	0.0756605
H	0.6153761	-6.0144550	-0.0274299
H	4.0947725	-0.9794124	-0.0665383
H	5.5163686	1.0064854	-0.0274290
H	-0.9925480	-2.1102825	-0.7337337
H	2.7441159	-4.9904321	0.7207483
H	2.1306696	3.6252684	0.0683283
H	2.9953040	-2.5558549	0.7724576
H	-1.6633557	5.4455330	-0.7620529
H	-3.8851272	4.6337287	-0.0238665
H	-4.1517323	2.2694063	0.6676577
H	-2.2183748	0.7507262	0.6636005
H	0.2535419	3.9234108	-0.8181068

Table S14. *XYZ* Coordinates for Optimized S_1 State of **1** in units of Ångstrøm.

35

O	-0.0027080	0.0002502	-0.0298401
C	-0.4094857	-4.0721449	-0.1027314
C	3.9246067	2.4179098	0.0414391
C	0.6919129	-4.9049039	0.0682186
C	3.6553347	0.0030077	-0.0598584
C	4.4667530	1.1393890	-0.0267214
C	-0.2538151	-2.7025815	-0.1288774
C	1.9581250	-4.3460977	0.2199159
C	2.5436027	2.6255009	0.0559483
C	2.1300296	-2.9785733	0.1943771
C	1.7004850	1.4990133	-0.0006172
C	1.1677660	-0.7050179	-0.0171372
C	2.2590013	0.1808273	-0.0219546
C	1.0264961	-2.1194250	0.0123541
C	0.3052574	1.3316577	-0.0276092
C	-1.7604372	4.4335772	-0.2579100
C	-3.0437661	3.9102698	-0.1248456
C	-3.2120517	2.5381787	0.0323193
C	-2.1194638	1.6979724	0.0611498
C	-0.8088463	2.2143744	-0.0599258
C	-0.6577327	3.6067395	-0.2261037
H	-1.3987869	-4.5002995	-0.2181531
H	4.5872227	3.2744924	0.0834781
H	0.5649327	-5.9810367	0.0880325
H	4.1060927	-0.9744598	-0.1384376
H	5.5435483	1.0198594	-0.0539781
H	-1.1126626	-2.0603172	-0.2731001
H	2.8186242	-4.9886831	0.3670197
H	2.1540678	3.6289204	0.1345385
H	3.1154365	-2.5638704	0.3450799
H	-1.6230709	5.5001492	-0.3942936
H	-3.9056628	4.5669117	-0.1485290
H	-4.2085064	2.1231235	0.1330147
H	-2.2561622	0.6326708	0.1926883
H	0.3268279	4.0289574	-0.3607499

Table S15. *XYZ* Coordinates for Optimized T_1 State of **1** in units of Ångstrøm.

35

O	0.0085100	-0.0076850	-0.2500771
C	-0.4059972	-4.0892663	-0.0291888
C	3.8945546	2.4244312	0.3640092
C	0.7021778	-4.9250794	0.0774295
C	3.5911318	0.0288253	0.4614221
C	4.4109063	1.1601476	0.5559158
C	-0.2538597	-2.7200608	-0.0633736
C	1.9743330	-4.3631311	0.1428855
C	2.5339032	2.6101926	0.0900472
C	2.1433736	-2.9956603	0.1140102
C	1.7060040	1.5011436	0.0054788
C	1.1602668	-0.7247088	-0.0093960
C	2.2455680	0.1941494	0.1706176
C	1.0305187	-2.1304363	0.0166833
C	0.3028927	1.3387381	-0.2403456
C	-1.7428548	4.4424914	-0.5723482
C	-2.9845518	3.9095518	-0.9086134
C	-3.1253341	2.5289456	-1.0172937
C	-2.0521618	1.6932198	-0.7965391
C	-0.7803438	2.2170193	-0.4614428
C	-0.6581379	3.6207015	-0.3536666
H	-1.4011278	-4.5158841	-0.0854063
H	4.5441420	3.2892686	0.4288999
H	0.5774392	-6.0009527	0.1035877
H	4.0214502	-0.9477102	0.6278886
H	5.4635710	1.0346945	0.7800589
H	-1.1220871	-2.0793853	-0.1382142
H	2.8456484	-5.0045184	0.2117453
H	2.1606036	3.6127242	-0.0590382
H	3.1441807	-2.5913011	0.1369811
H	-1.6238508	5.5158026	-0.4762091
H	-3.8319329	4.5624856	-1.0798821
H	-4.0875651	2.1024743	-1.2775155
H	-2.1707004	0.6221906	-0.8893532
H	0.2853168	4.0630330	-0.0708618