Supplementary Material

Modulating organic photovoltaic properties of non-fullerene acceptors by molecular modification based on Y6: a theoretical study

Cai-Rong Zhang [a*], Hai-Yuan Yu [a], Mei-Ling Zhang [a], Xiao-Meng Liu [a], Yu-Hong Chen [a], Zi-Jiang Liu [b], You-Zhi Wu [c], Hong-Shan Chen [d]

a Department of Applied Physics, Lanzhou University of Technology, Lanzhou, Gansu 730050, China;

b School of Mathematics and Physics, Lanzhou Jiaotong University, Lanzhou 730070, China;

c School of Materials Science and Engineering, Lanzhou University of Technology, Lanzhou, Gansu 730050, China;

d College of Physics and Electronic Engineering, Northwest Normal University, Lanzhou, Gansu 730070, China.

*Corresponding author.

E-mail address: zhcrxy@lut.edu.cn (C.R. Zhang)

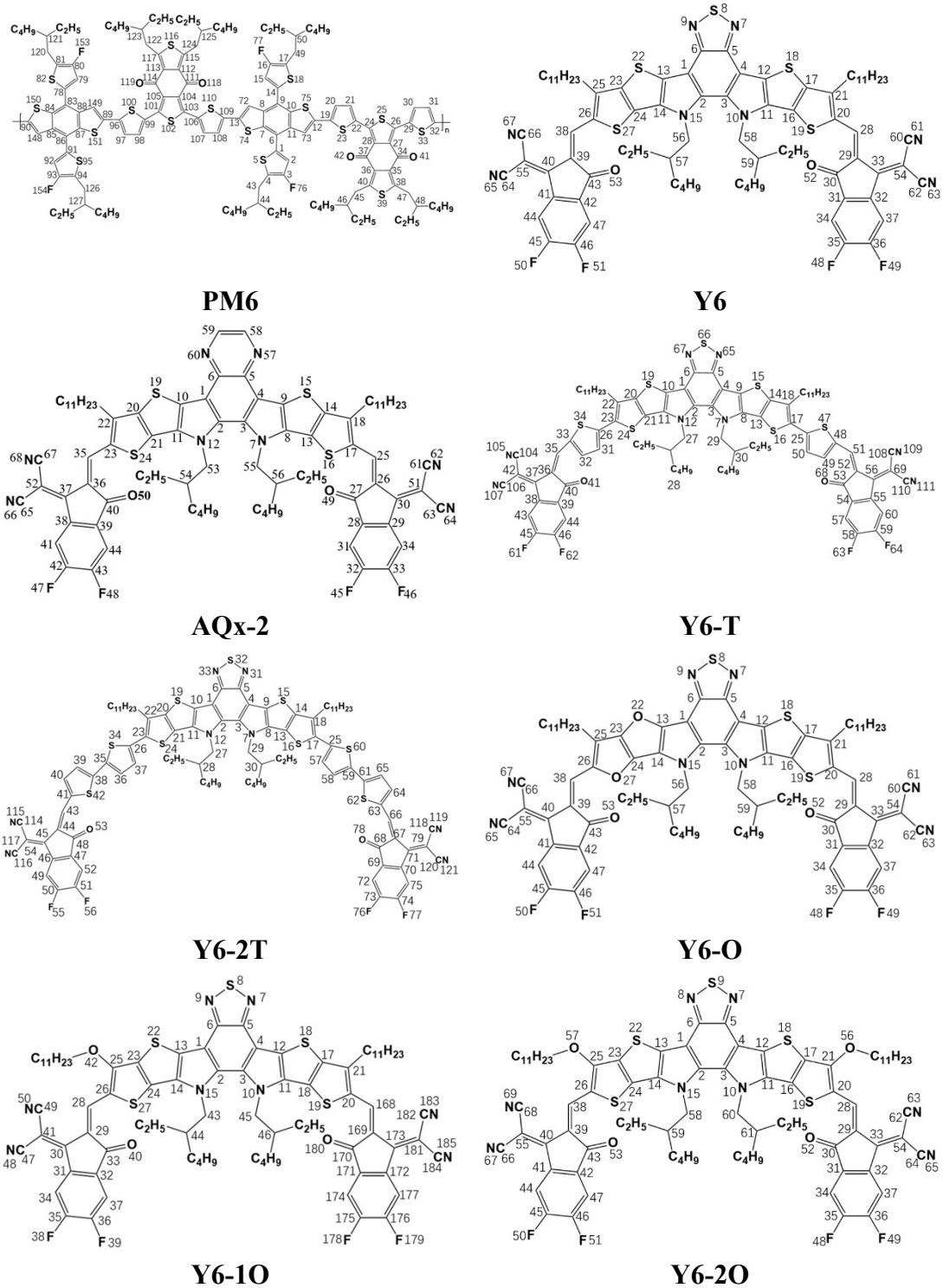


Figure S1. The molecular structures of PM6, Y6, AQx-2, Y6-T, Y6-2T, Y6-O, Y6-1O and Y6-2O. The atomic serial numbers are labeled in order to give geometrical parameters.

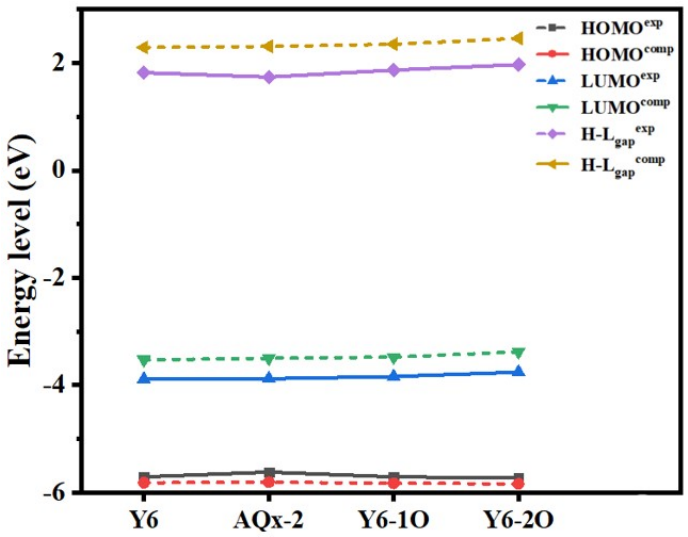
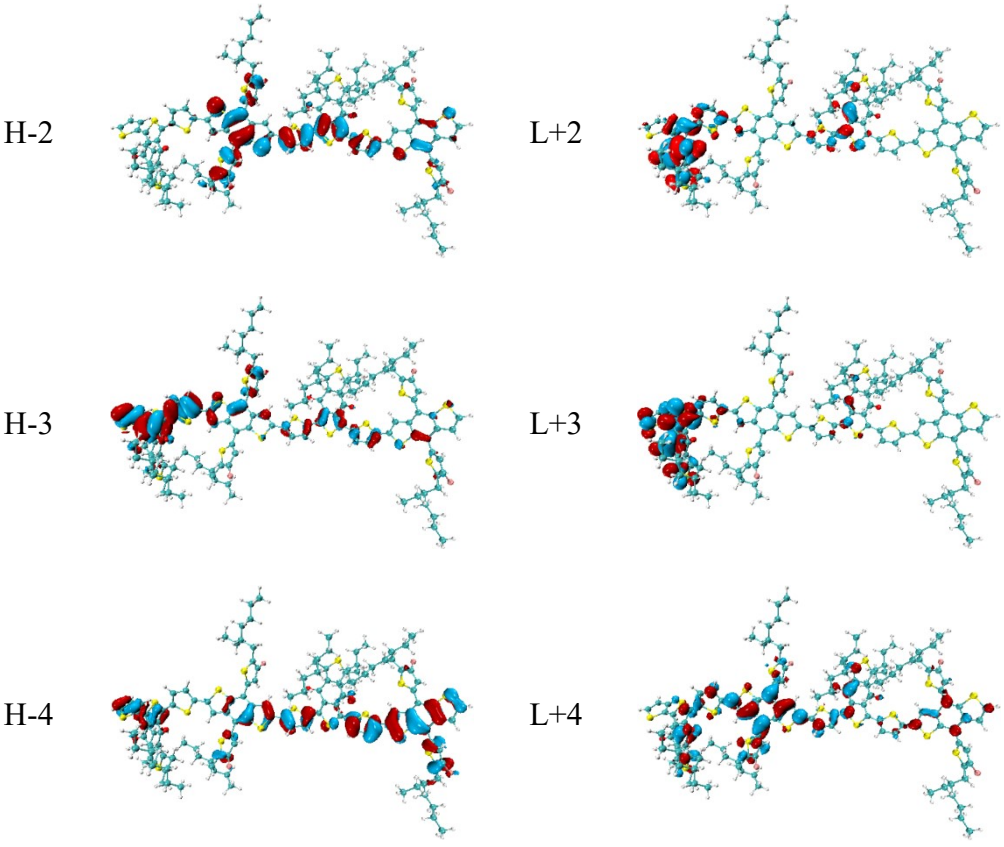
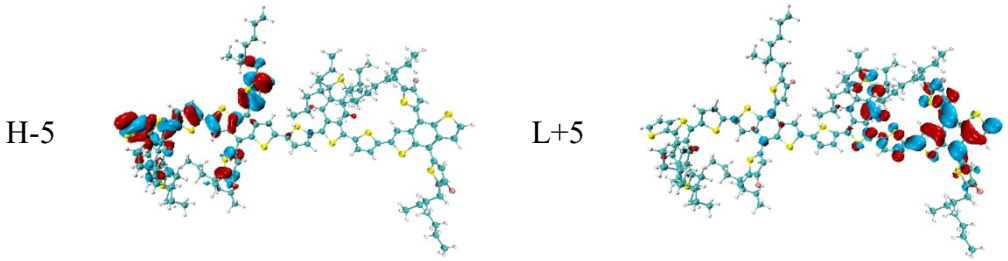


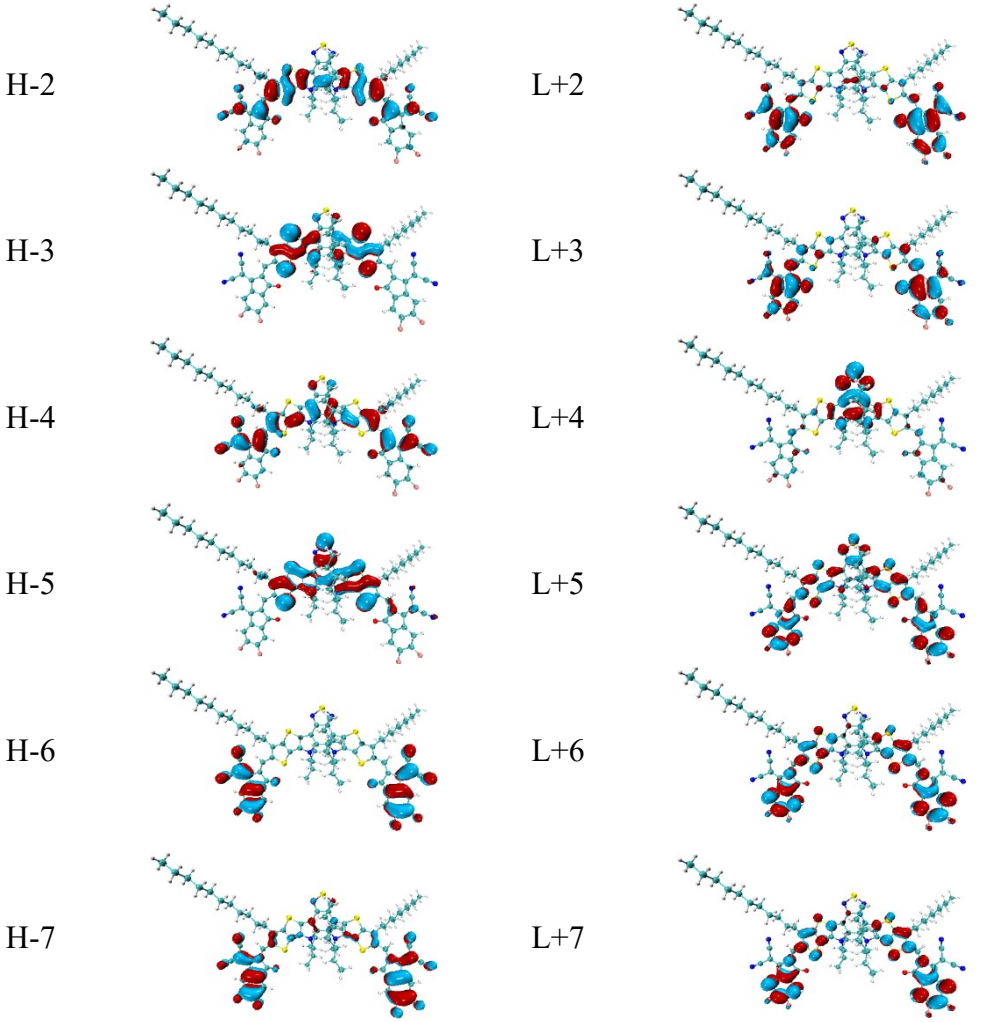
Figure S2. The comparisons of HOMO, LUMO, gap energy of Y6, AQx -2, Y6-1O and Y6-2O between the experiment and calculation.

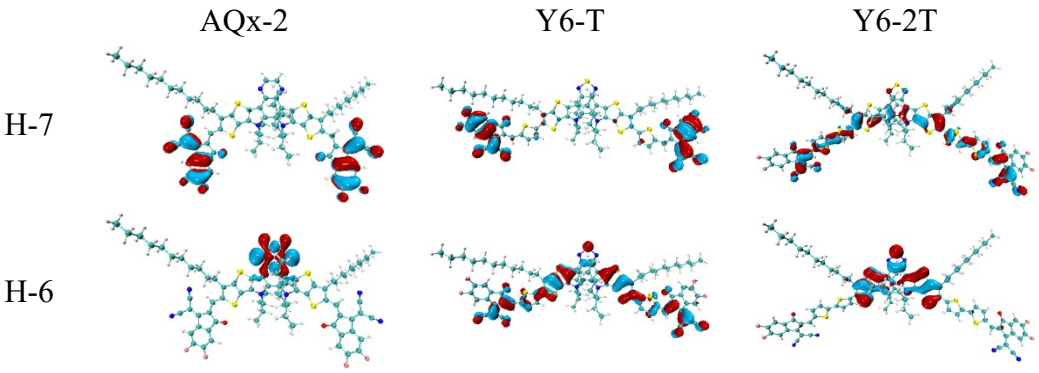
PM6

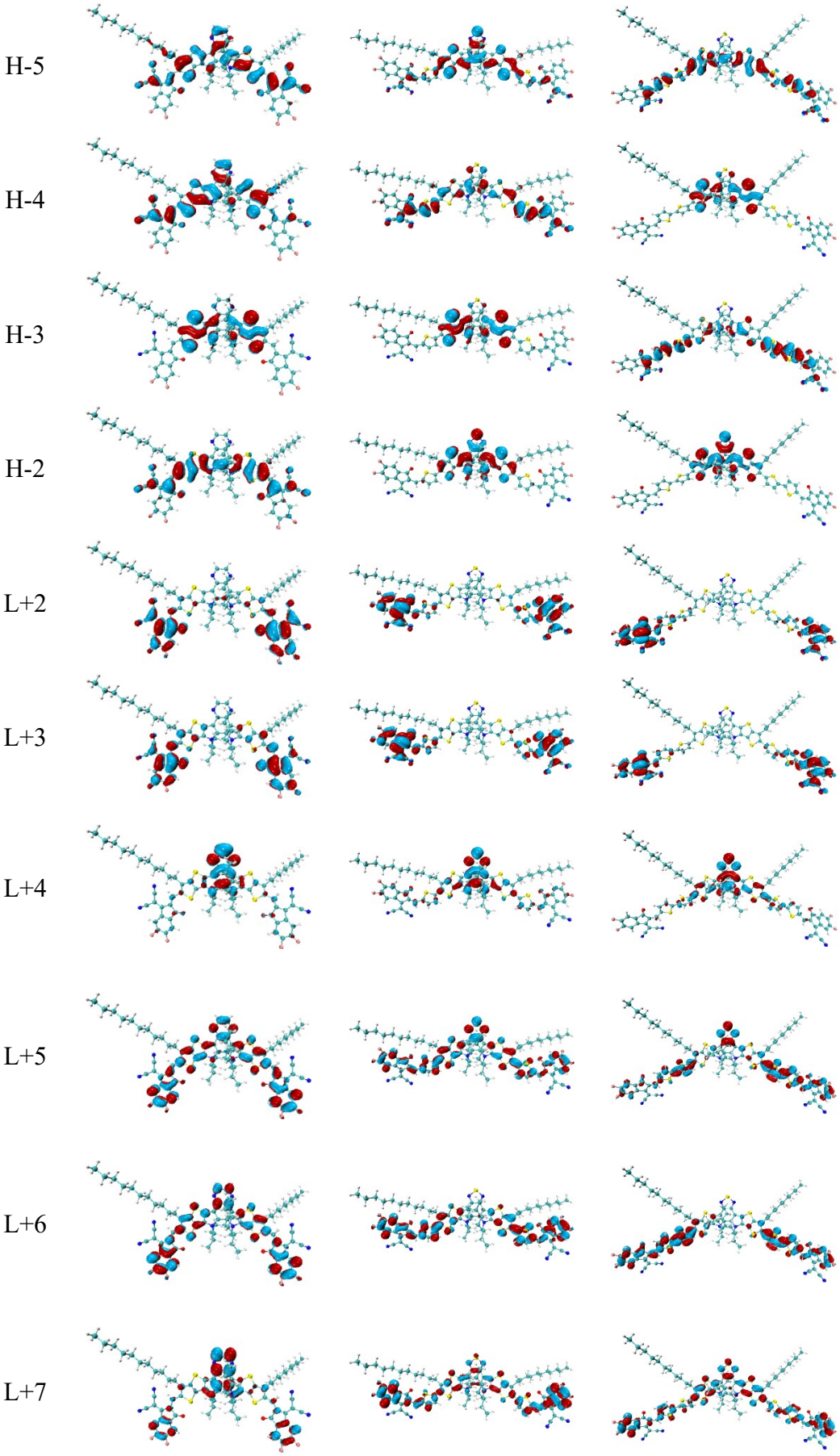


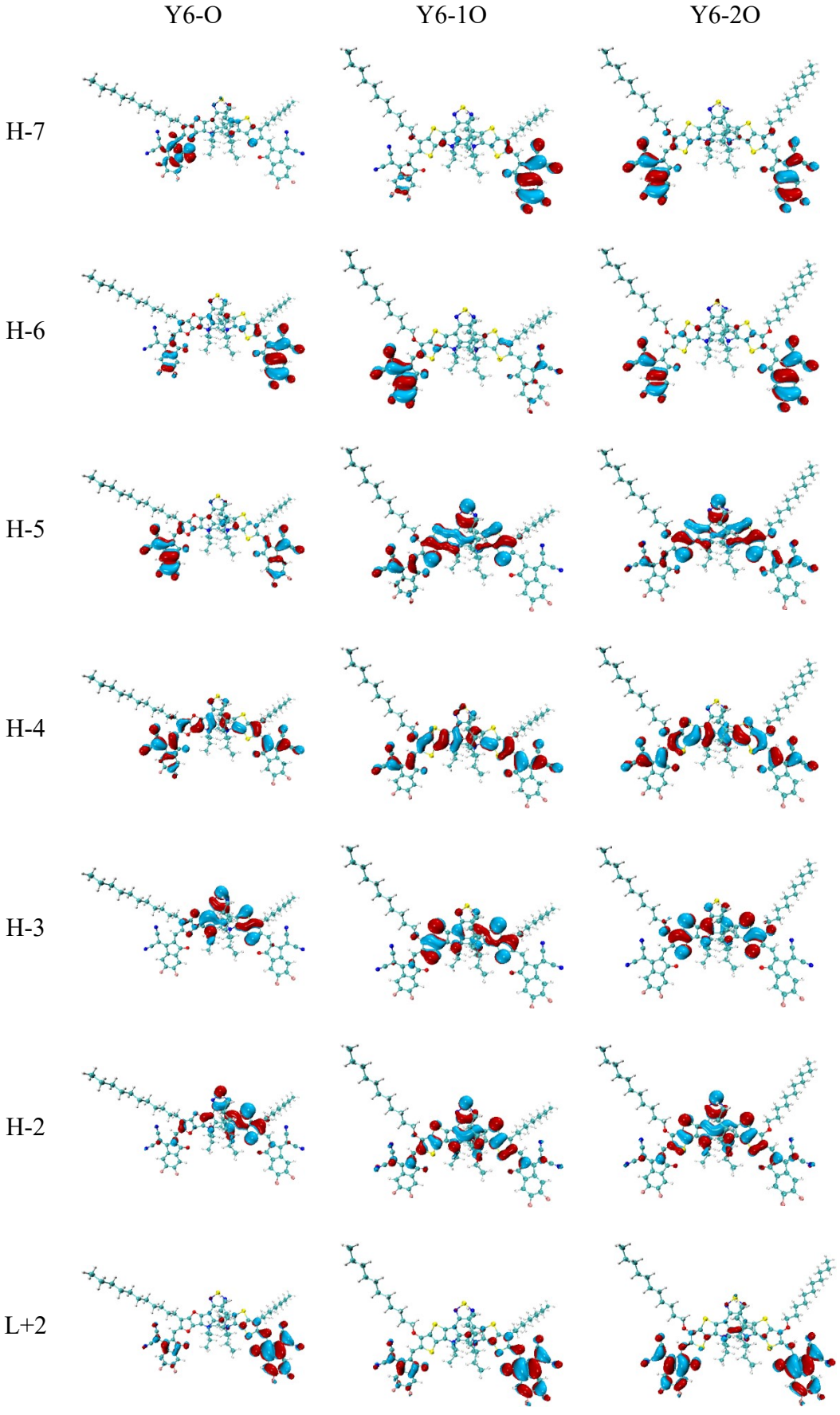


Y6






H-5
H-4
H-3
H-2
L+2
L+3
L+4
L+5
L+6
L+7


Y6-O
Y6-1O
Y6-2O
H-7
H-6
H-5
H-4
H-3
H-2
L+2

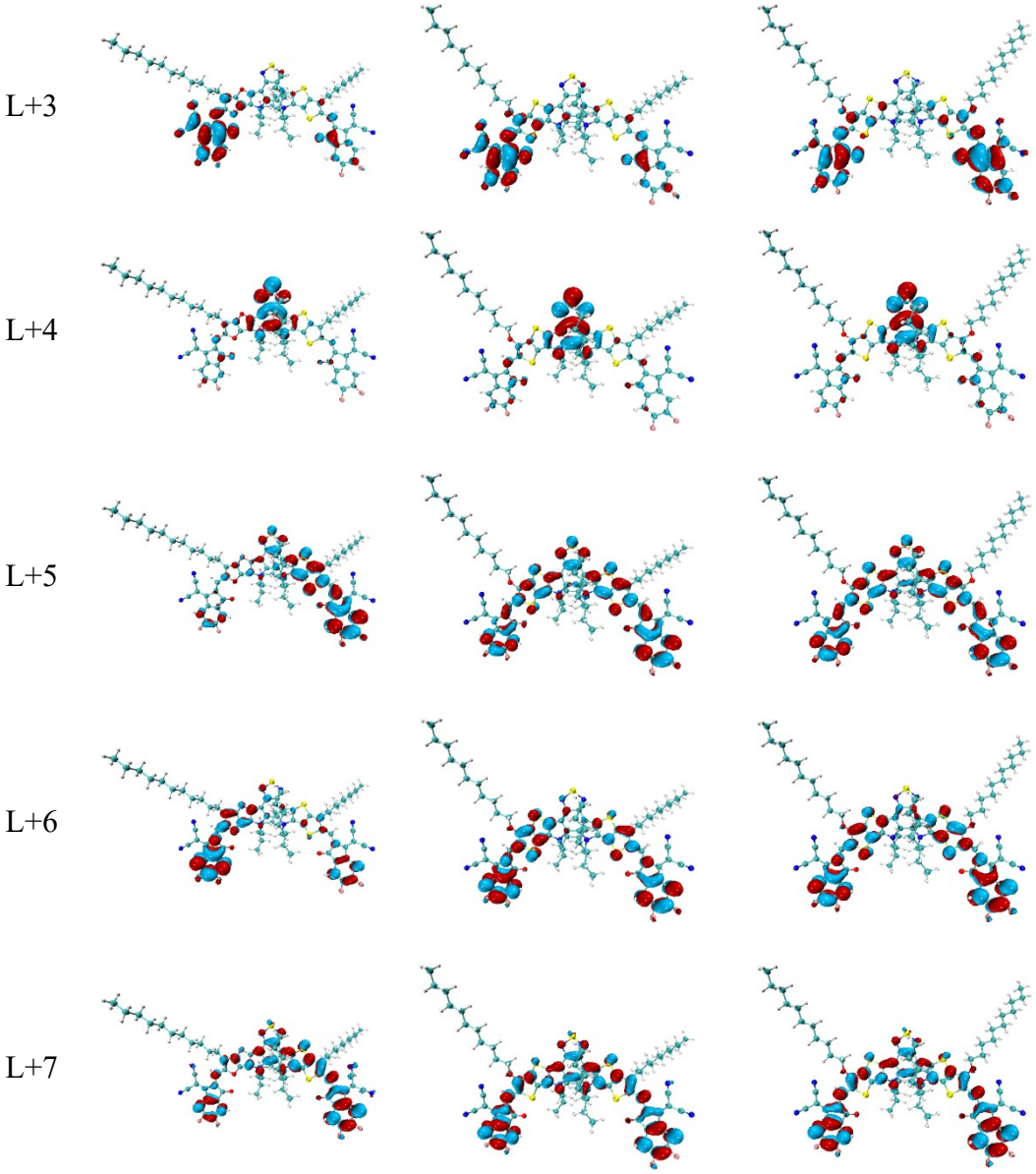
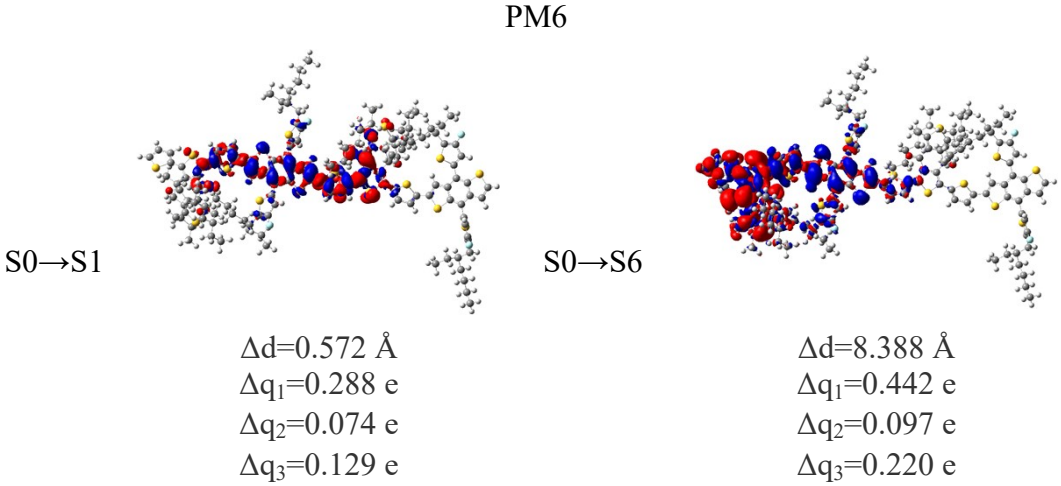
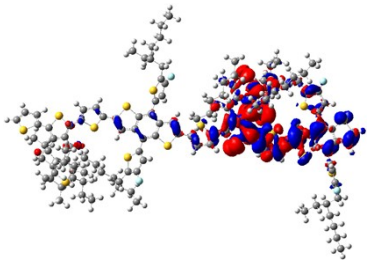


Figure S3. Selected frontier molecular orbitals for the PM6, Y6, AQx-2, Y6-T, Y6-2T, Y6-O, Y6-1O and Y6-2O. (H = HOMO, L = LUMO; LC-PBE/ CEP-121G*).

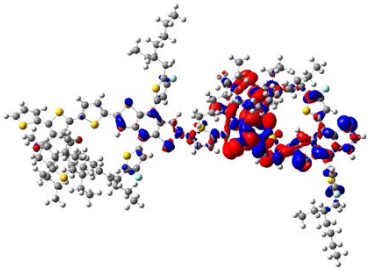


S0→S2



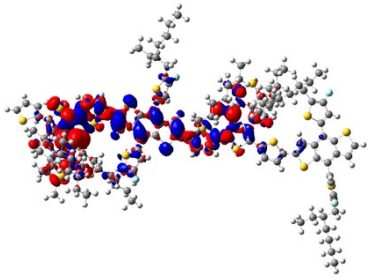
Δd=2.518 Å
Δq_1=0.437 e
Δq_2=0.072 e
Δq_3=0.158 e

S0→S7



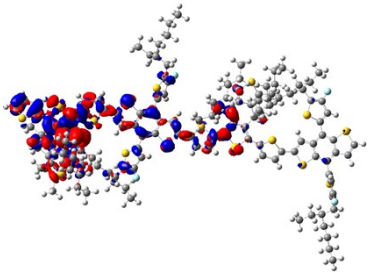
Δd=2.202 Å
Δq_1=0.426 e
Δq_2=0.151 e
Δq_3=0.061 e

S0→S3



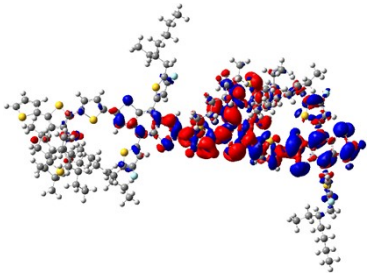
Δd=2.920 Å
Δq_1=0.466 e
Δq_2=0.072 e
Δq_3=0.195 e

S0→S8



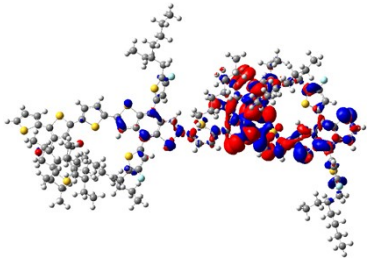
Δd=1.805 Å
Δq_1=0.305 e
Δq_2=0.043 e
Δq_3=0.176 e

S0→S4



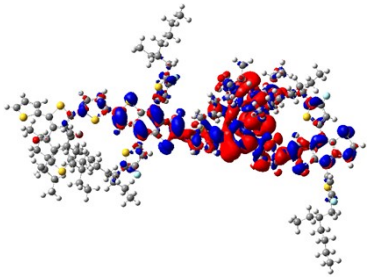
Δd=5.517 Å
Δq_1=0.478 e
Δq_2=0.163 e
Δq_3=0.070 e

S0→S9



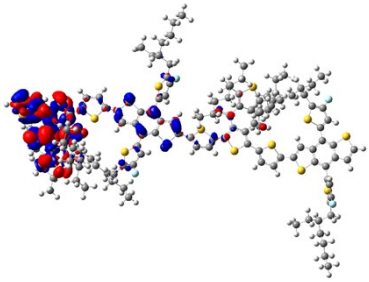
Δd=2.396 Å
Δq_1=0.453 e
Δq_2=0.054 e
Δq_3=0.167 e

S0→S5



Δd=5.168 Å
Δq_1=0.587 e
Δq_2=0.079 e
Δq_3=0.129 e

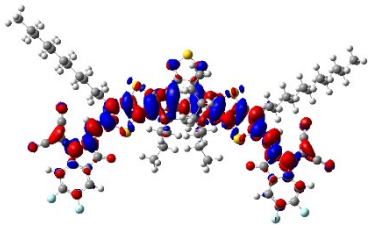
S0→S10



Δd=2.372 Å
Δq_1=0.202 e
Δq_2=0.029 e
Δq_3=0.253 e

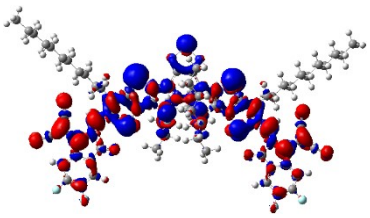
Y6

S0→S1



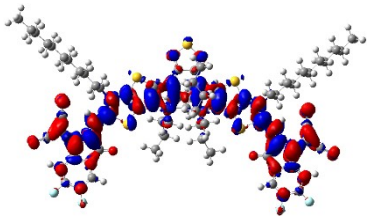
Δd=1.714 Å
Δq=0.421 e

S0→S6



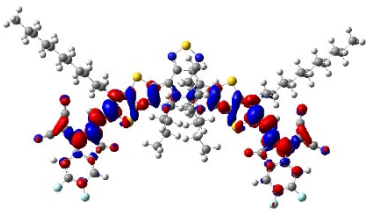
Δd=3.285 Å
Δq=0.697 e

S0→S2



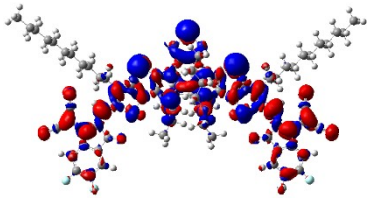
Δd=2.093 Å
Δq=0.516 e

S0→S7



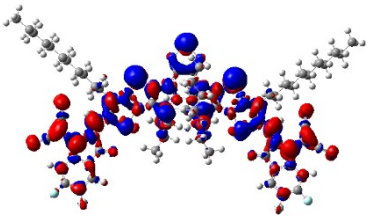
Δd=1.072 Å
Δq=0.302 e

S0→S3



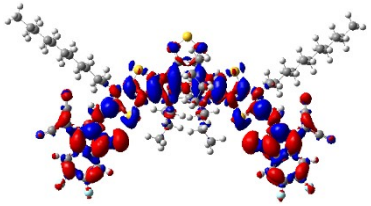
Δd=3.355 Å
Δq=0.648 e

S0→S8



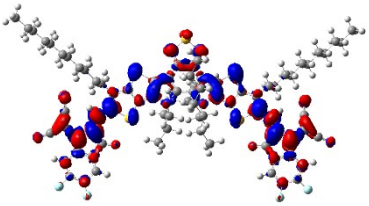
Δd=2.842 Å
Δq=0.638 e

S0→S4



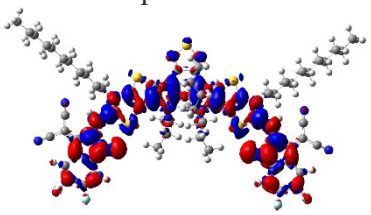
Δd=3.549 Å
Δq=0.656 e

S0→S9



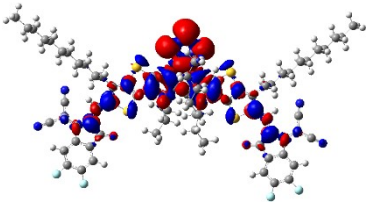
Δd=0.796 Å
Δq=0.331 e

S0→S5

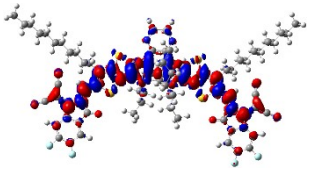
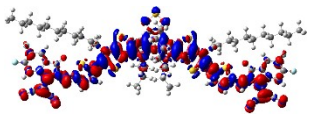
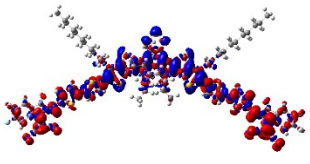


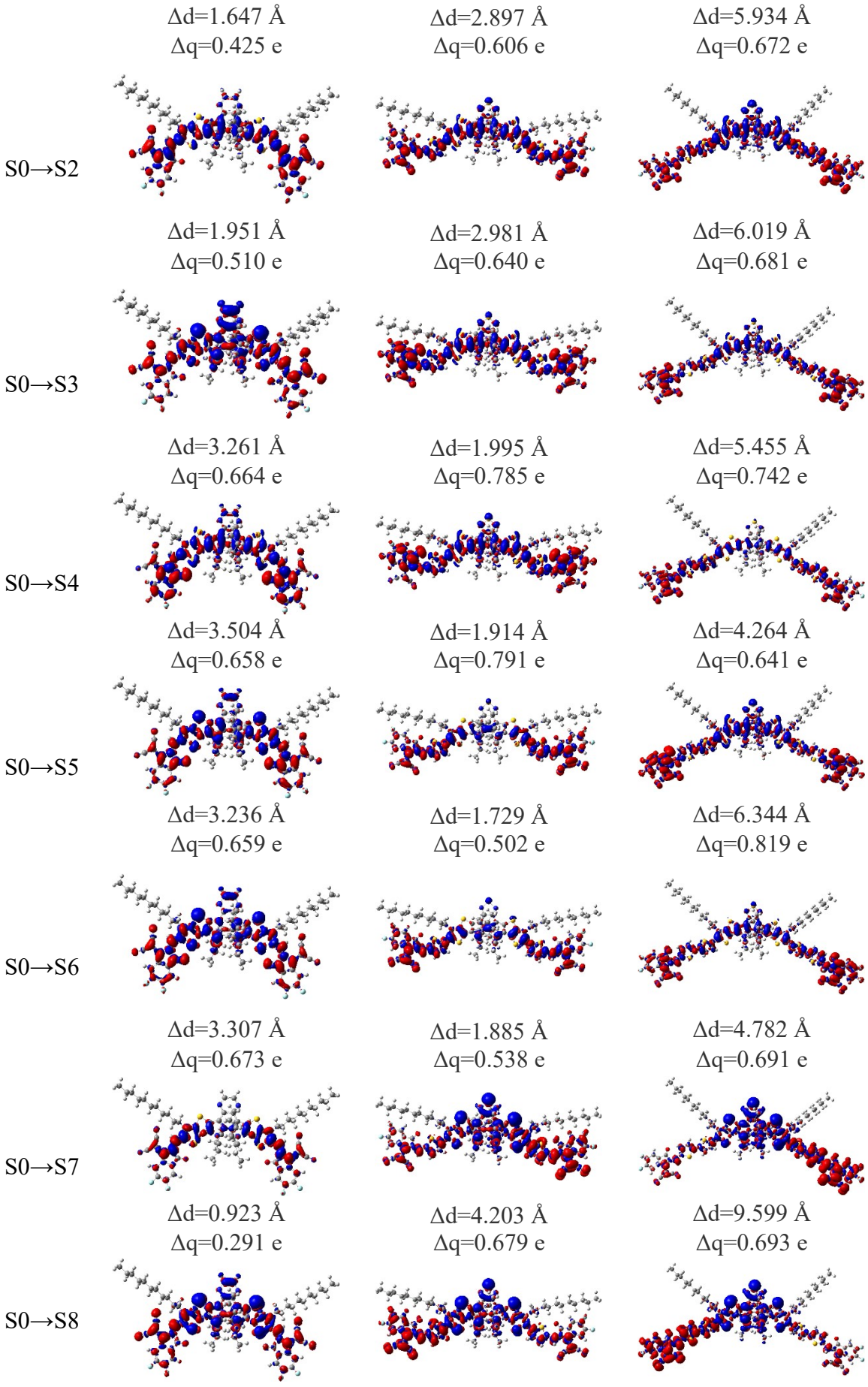
Δd=3.352 Å
Δq=0.622 e

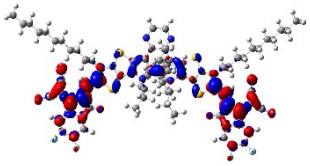
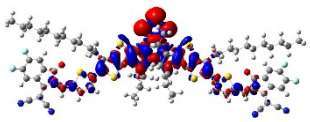
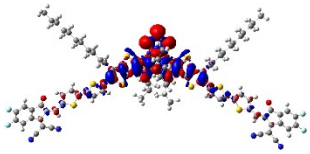
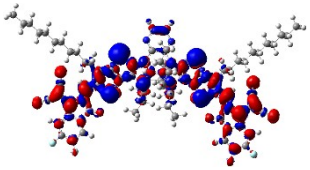
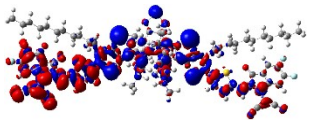
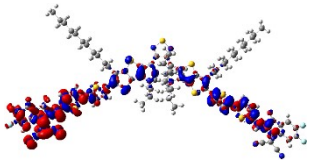
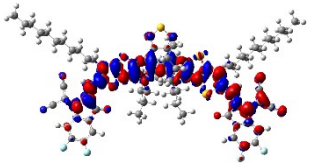
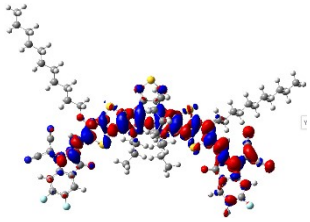
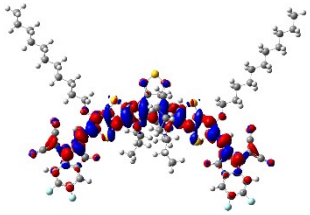
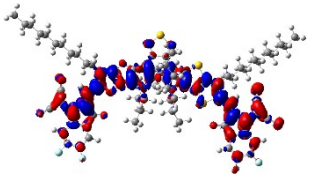
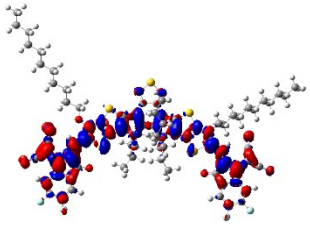
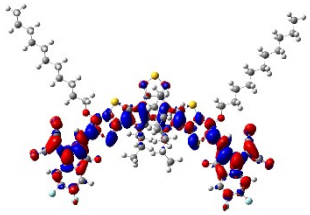
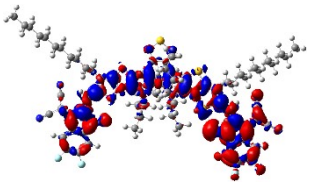
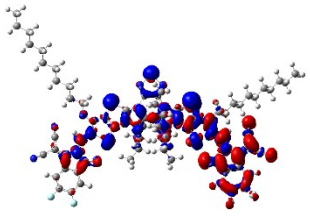
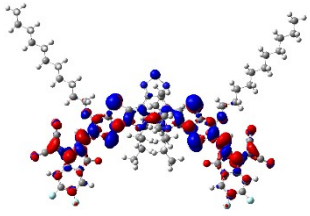
S0→S10

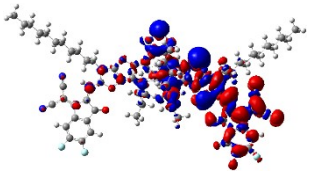
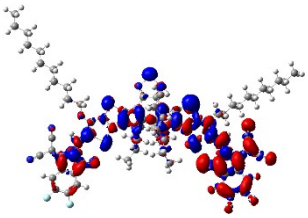
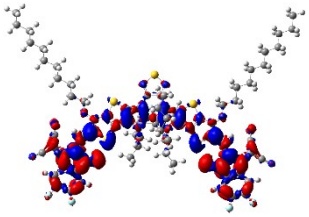
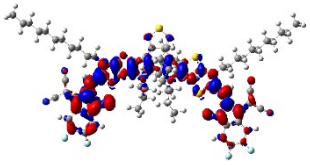
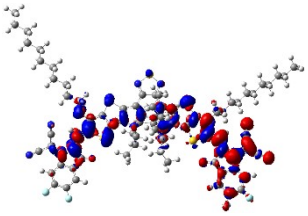
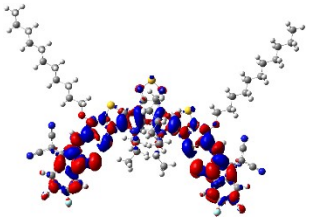
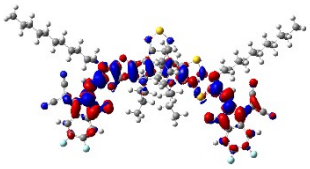
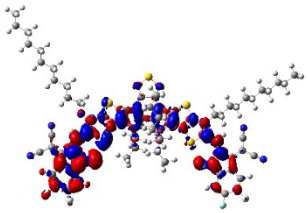
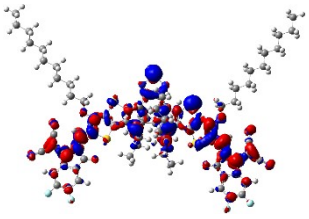
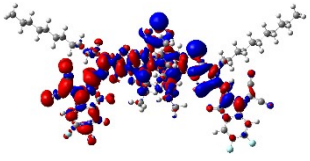
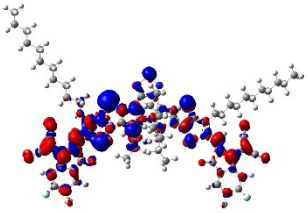
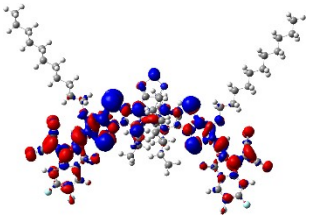
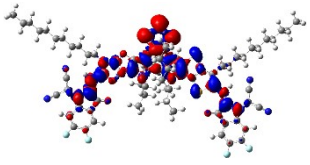
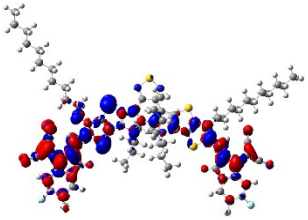
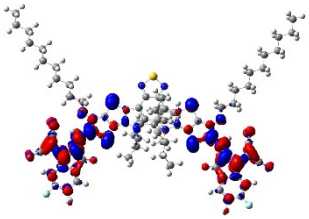


Δd=2.145 Å
Δq=-0.048 e

	AQx-2	Y6-T	Y6-2T
S0→S1			


Δd=1.647 Å
Δq=0.425 e
Δd=2.897 Å
Δq=0.606 e
Δd=5.934 Å
Δq=0.672 e
S0→S2
Δd=1.951 Å
Δq=0.510 e
Δd=2.981 Å
Δq=0.640 e
Δd=6.019 Å
Δq=0.681 e
S0→S3
Δd=3.261 Å
Δq=0.664 e
Δd=1.995 Å
Δq=0.785 e
Δd=5.455 Å
Δq=0.742 e
S0→S4
Δd=3.504 Å
Δq=0.658 e
Δd=1.914 Å
Δq=0.791 e
Δd=4.264 Å
Δq=0.641 e
S0→S5
Δd=3.236 Å
Δq=0.659 e
Δd=1.729 Å
Δq=0.502 e
Δd=6.344 Å
Δq=0.819 e
S0→S6
Δd=3.307 Å
Δq=0.673 e
Δd=1.885 Å
Δq=0.538 e
Δd=4.782 Å
Δq=0.691 e
S0→S7
Δd=0.923 Å
Δq=0.291 e
Δd=4.203 Å
Δq=0.679 e
Δd=9.599 Å
Δq=0.693 e
S0→S8

	Δd=2.931 Å Δq=0.680 e	Δd=3.958 Å Δq=0.675 e	Δd=9.752 Å Δq=0.696 e
S0→S9			
	Δd=1.110 Å Δq=0.359 e	Δd=2.339 Å Δq=-0.007 e	Δd=1.611 Å Δq=0.051 e
S0→S10			
	Δd=2.996 Å Δq=0.722 e	Δd=5.500 Å Δq=0.682 e	Δd=6.707 Å Δq=0.503 e
	Y6-O	Y6-1O	Y6-2O
S0→S1			
	Δd=3.451 Å Δq=0.368 e	Δd=3.253 Å Δq=0.384 e	Δd=1.287 Å Δq=0.340 e
S0→S2			
	Δd=1.763 Å Δq=0.453 e	Δd=2.194 Å Δq=0.496 e	Δd=1.831 Å Δq=0.468 e
S0→S3			
	Δd=4.831 Å Δq=0.612 e	Δd=4.171 Å Δq=0.626 e	Δd=1.929 Å Δq=0.431 e

S0→S4			
	Δd=3.764 Å Δq=0.647 e	Δd=4.823 Å Δq=0.632 e	Δd=3.185 Å Δq=0.609 e
S0→S5			
	Δd=2.167 Å Δq=0.449 e	Δd=2.535 Å Δq=0.385 e	Δd=3.044 Å Δq=0.567 e
S0→S6			
	Δd=1.871 Å Δq=0.383 e	Δd=3.125 Å Δq=0.546 e	Δd=1.912 Å Δq=0.395 e
S0→S7			
	Δd=4.609 Å Δq=0.598 e	Δd=2.959 Å Δq=0.608 e	Δd=3.038 Å Δq=0.639 e
S0→S8			
	Δd=1.266 Å Δq=0.053 e	Δd=1.720 Å Δq=0.446 e	Δd=1.498 Å Δq=0.423 e

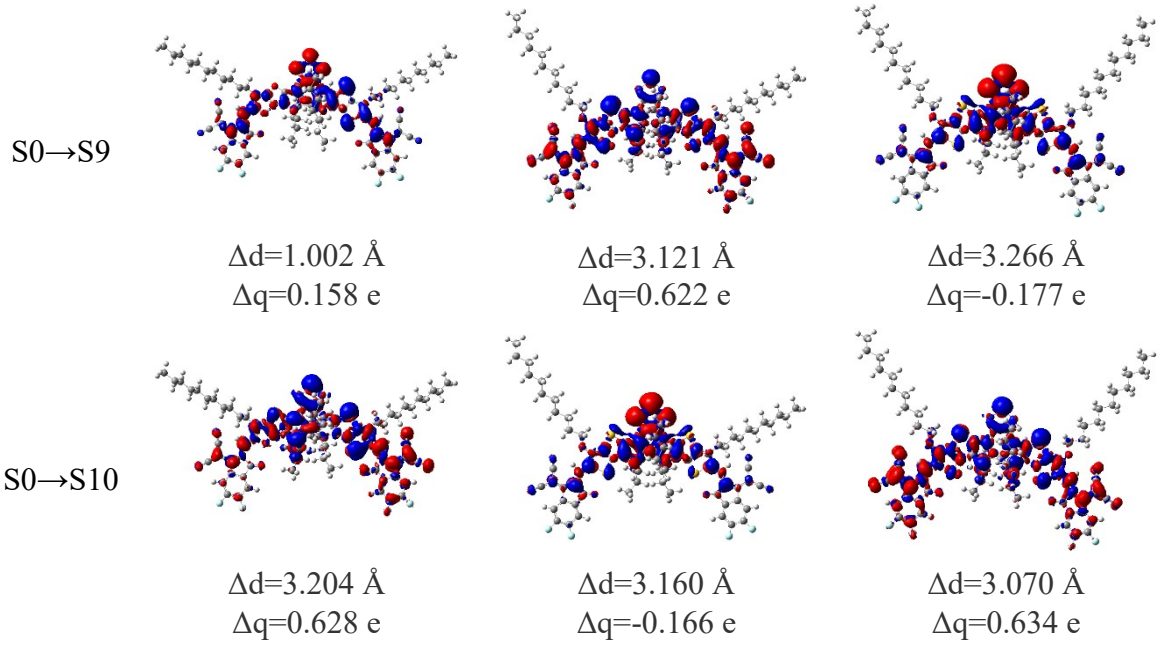
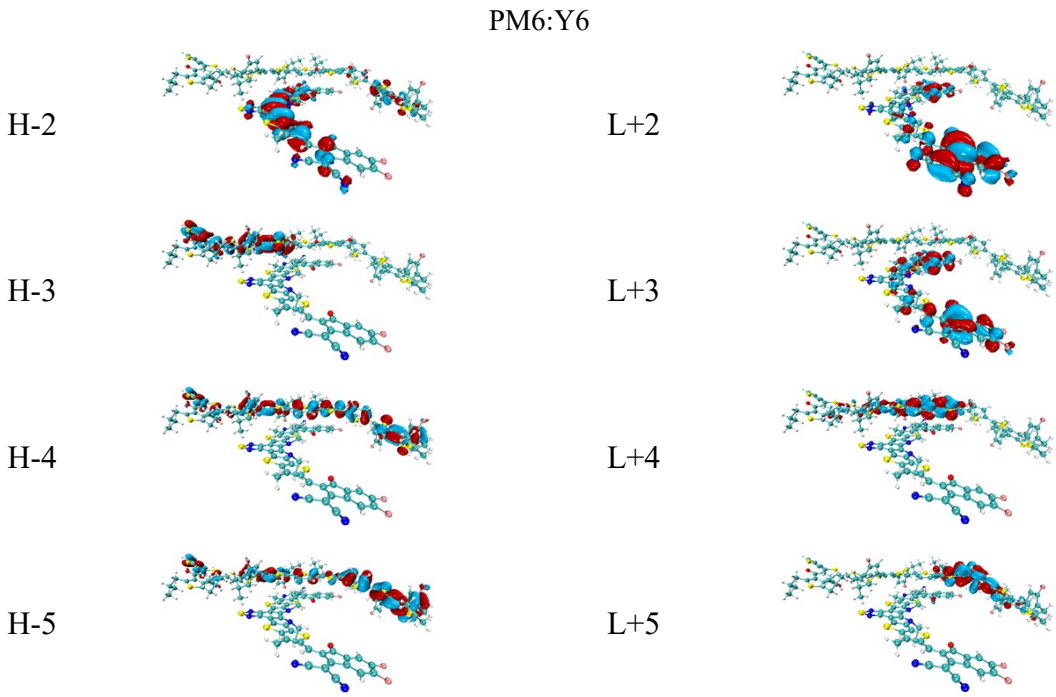
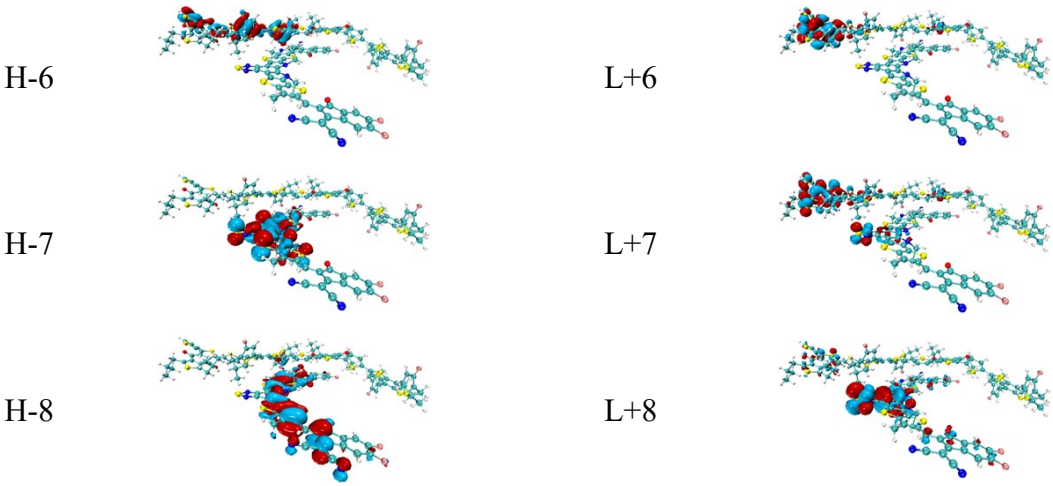
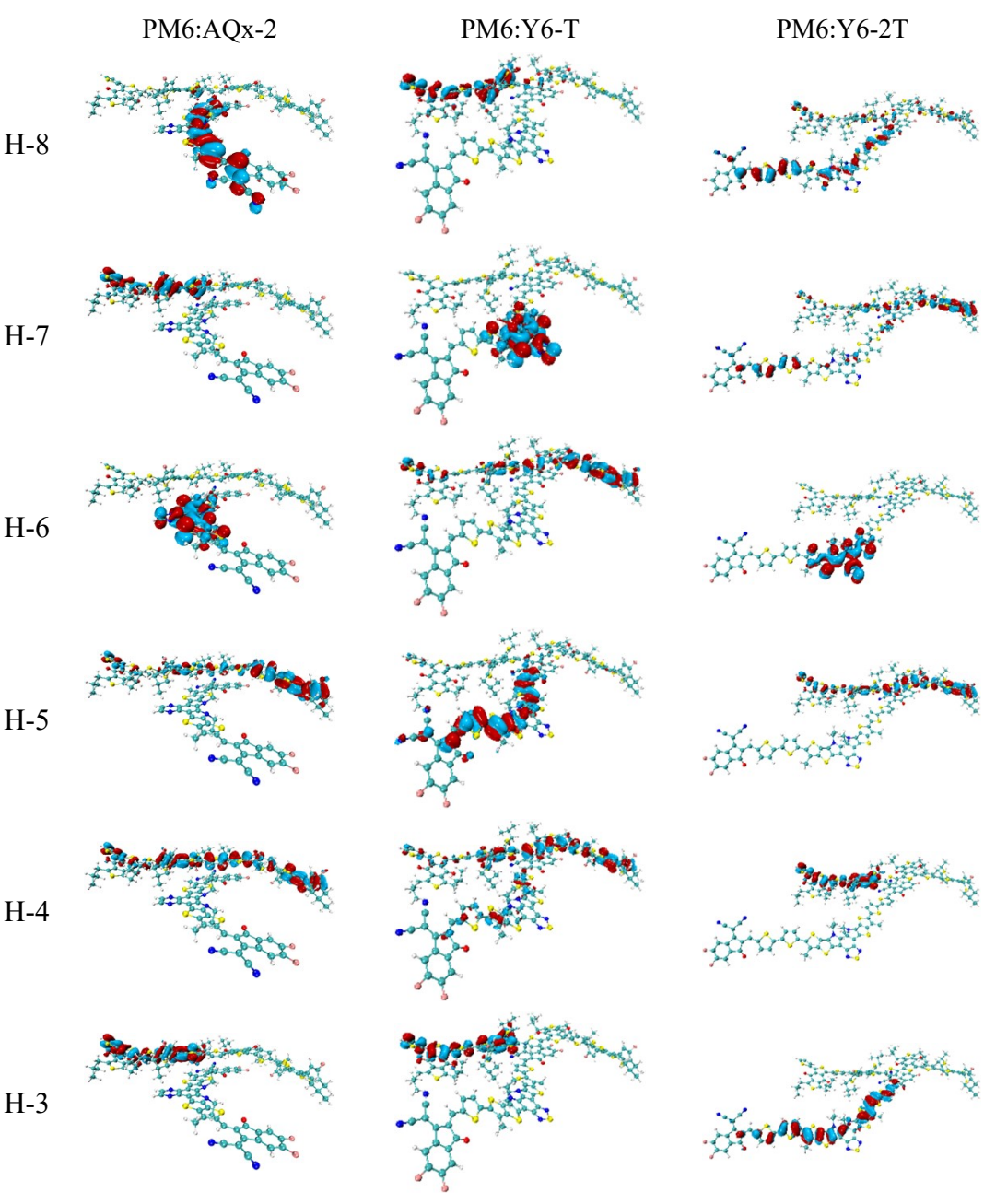
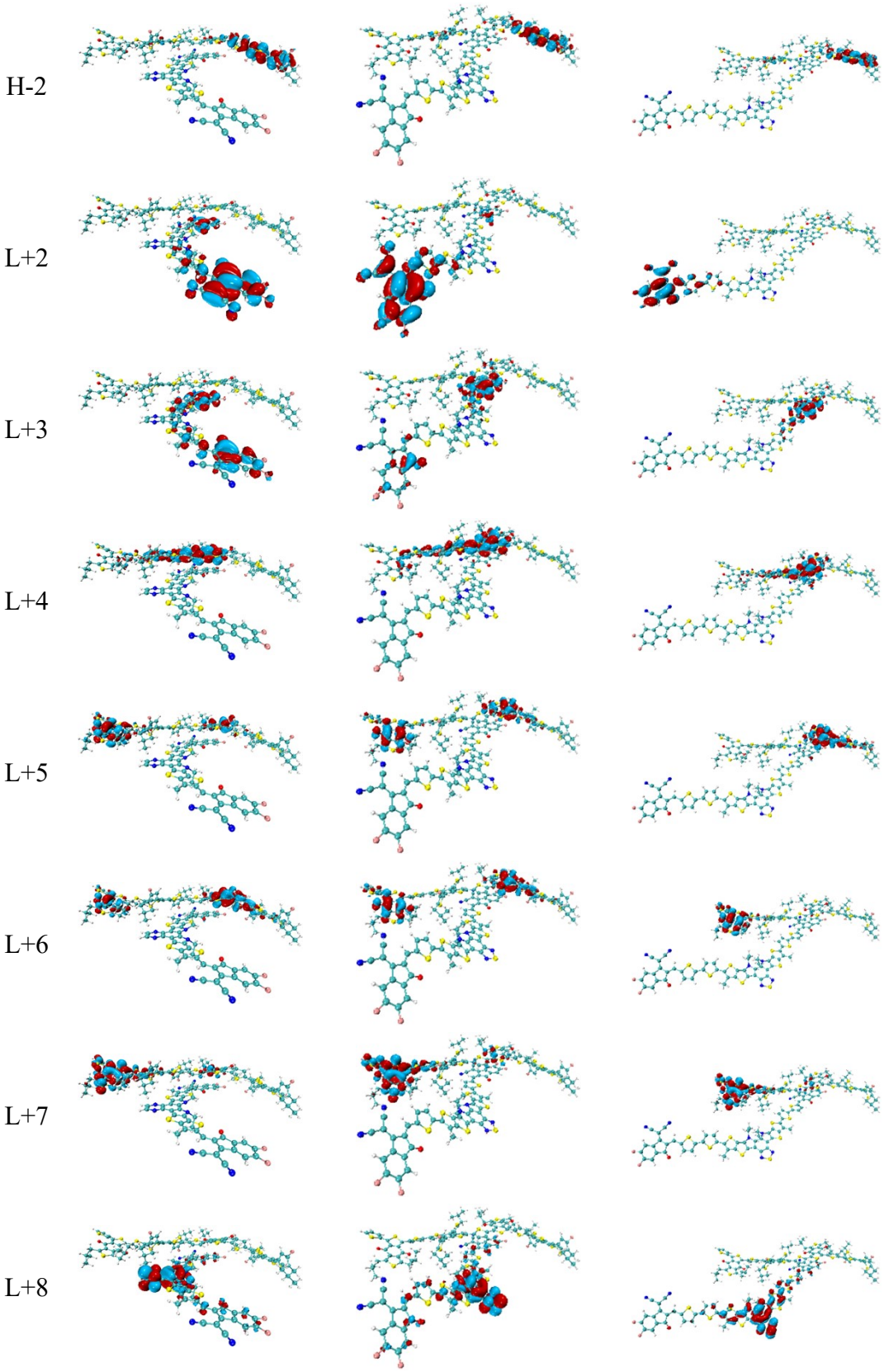


Figure S4. The charge density difference low-lying excited states for PM6 and NFAs Y6, AQx-2, Y6-T, Y6-2T, Y6-O, Y6-1O and Y6-2O. The red and blue areas indicate the increase and decrease of electron density during excitation, respectively. The Δd represent charge transfer distance. The Δq_1, Δq_2 and Δq_3 represent the transferred charges from the donor fragment to the acceptor fragment, from the donor fragment to the thiophene fragment and from the thiophene fragment to the acceptor fragment, respectively. (LC-PBE/CEP-121G*).

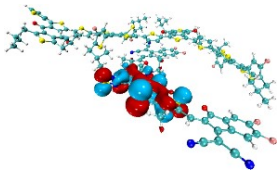


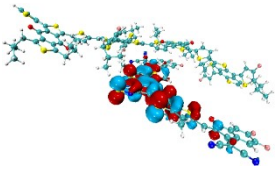

H-6
L+6
H-7
L+7
H-8
L+8

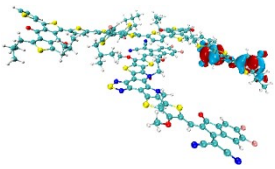

PM6:AQx-2
PM6:Y6-T
PM6:Y6-2T
H-8
H-7
H-6
H-5
H-4
H-3


H-2
L+2
L+3
L+4
L+5
L+6
L+7
L+8
PM6:Y6-O
PM6:Y6-1O
PM6:Y6-2O

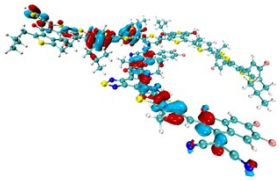
H-8

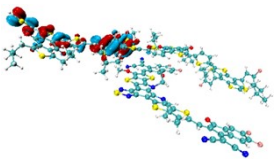


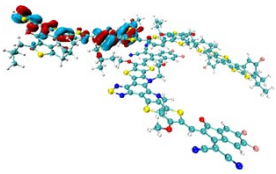




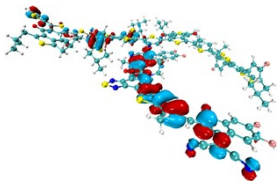
H-7

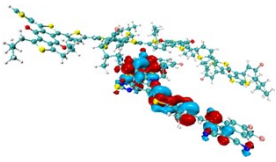


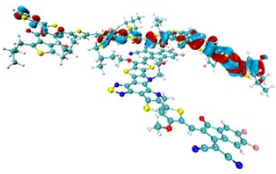




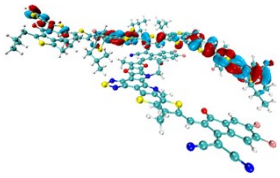
H-6

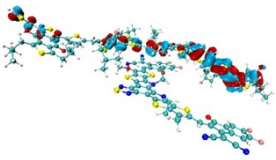


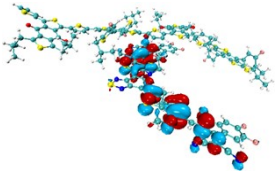




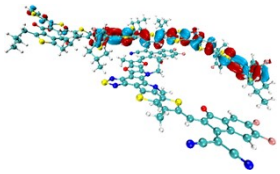
H-5

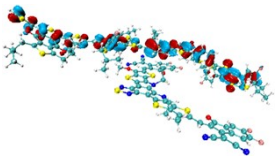


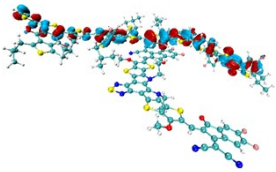




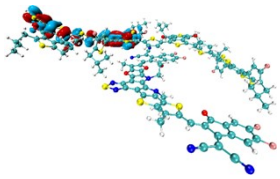
H-4

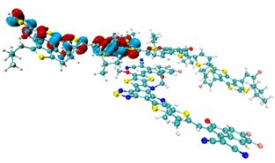


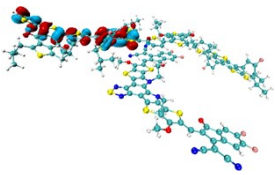




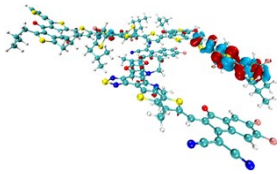
H-3

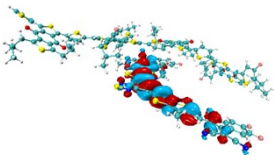


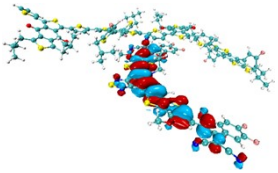




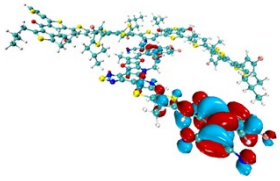
H-2

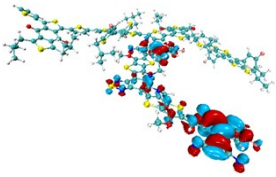




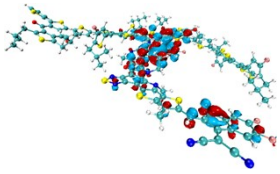


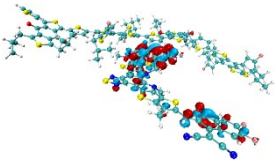
L+2

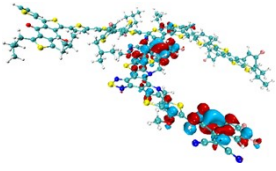




L+3







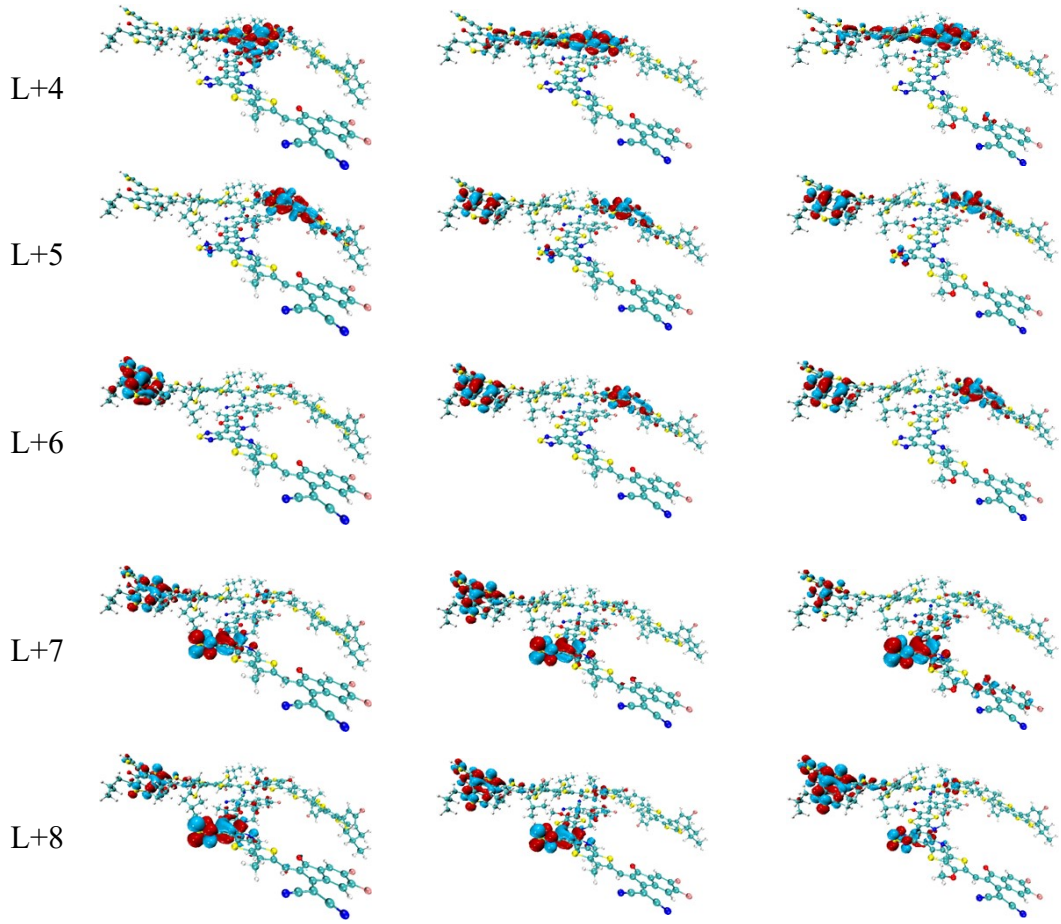
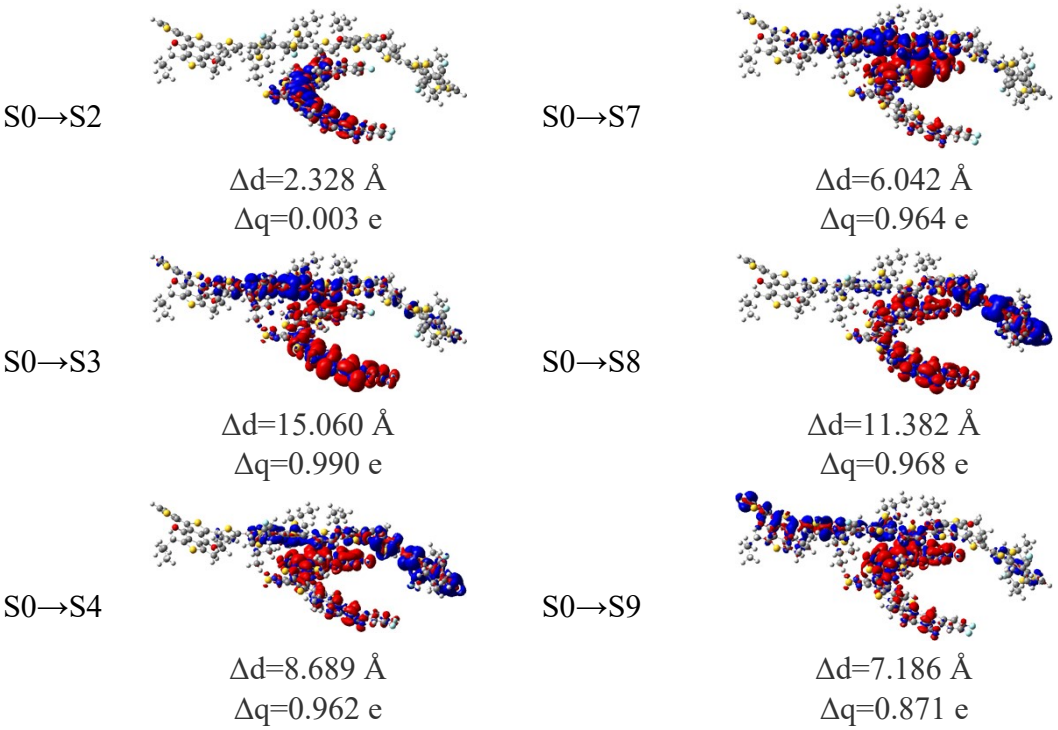
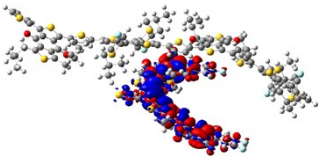


Figure S5. Selected frontier molecular orbitals for seven complex molecules. (H = HOMO, L = LUMO, LC-PBE/ CEP-121G*).

PM6:Y6

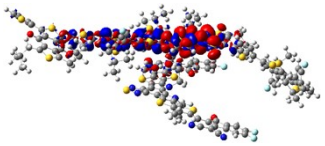


S0→S5

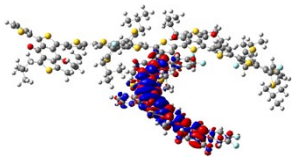
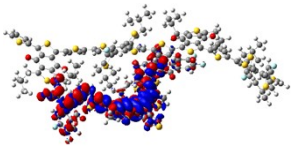
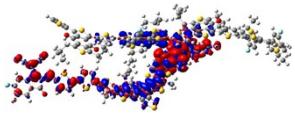
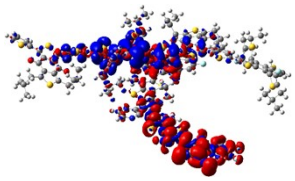
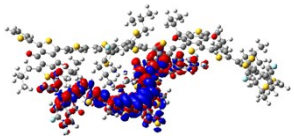
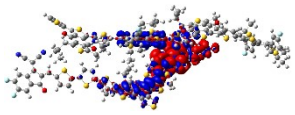
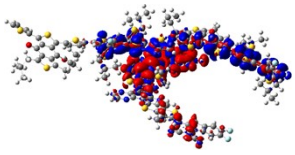
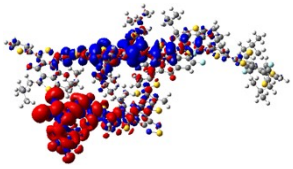
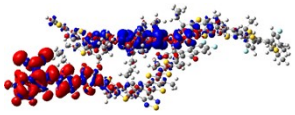
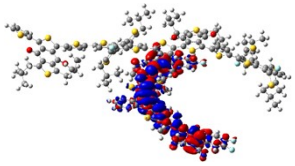
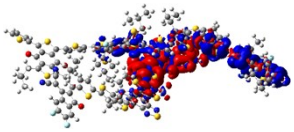
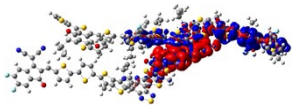
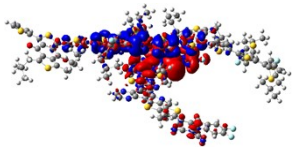
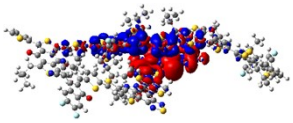
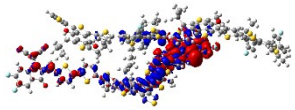
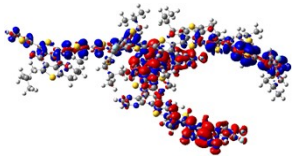
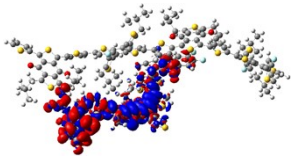
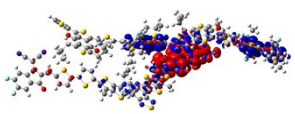


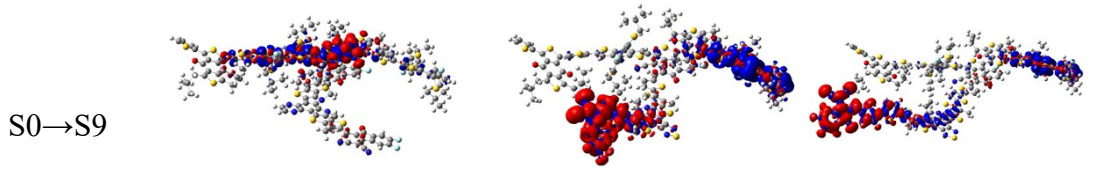
Δd=2.567 Å
Δq=-0.010 e

S0→S10



Δd=3.536 Å
Δq=0.087 e

	PM6:AQx-2	PM6:Y6-T	PM6:Y6-2T
S0→S2	 Δd=2.120 Å Δq=0.024 e	 Δd=4.033 Å Δq=0.039 e	 Δd=2.076 Å Δq=0.444 e
S0→S3	 Δd=16.196 Å Δq=0.988 e	 Δd=3.702 Å Δq=-0.019 e	 Δd=7.903 Å Δq=0.428 e
S0→S4	 Δd=5.935 Å Δq=0.950 e	 Δd=22.699 Å Δq=0.997 e	 Δd=27.183 Å Δq=0.983 e
S0→S5	 Δd=2.225 Å Δq=0.011 e	 Δd=7.818 Å Δq=0.951 e	 Δd=7.062 Å Δq=0.973 e
S0→S7	 Δd=6.139 Å Δq=0.962 e	 Δd=4.460 Å Δq=0.948 e	 Δd=1.623 Å Δq=0.282 e
S0→S8	 Δd=8.604 Å Δq=0.910 e	 Δd=6.795 Å Δq=-0.001 e	 Δd=4.702 Å Δq=0.798 e

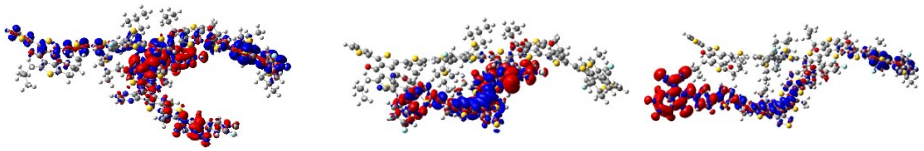


S0→S9

Δd=2.238 Å
Δq=0.110 e

Δd=28.916 Å
Δq=0.996 e

Δd=31.552 Å
Δq=0.724 e



S0→S10

Δd=5.868 Å
Δq=0.929 e

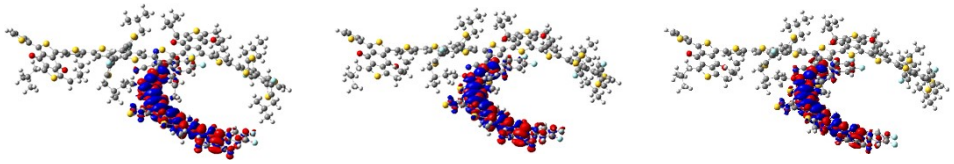
Δd=4.939 Å
Δq=-0.020 e

Δd=22.047 Å
Δq=0.433 e

PM6:Y6-O

PM6:Y6-1O

PM6:Y6-2O

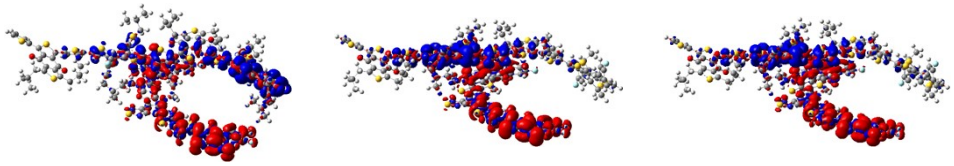


S0→S2

Δd=4.513 Å
Δq=0.006 e

Δd=3.987 Å
Δq=0.003 e

Δd=2.120 Å
Δq=0.001 e

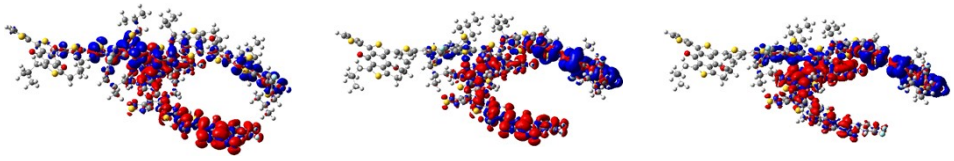


S0→S3

Δd=11.902 Å
Δq=0.961 e

Δd=15.269 Å
Δq=0.985 e

Δd=13.553 Å
Δq=0.983 e

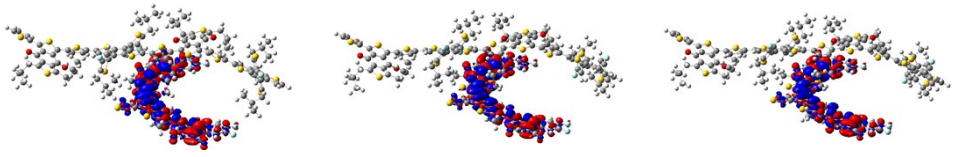


S0→S4

Δd=12.078 Å
Δq=0.988 e

Δd=13.429 Å
Δq=0.984 e

Δd=8.455 Å
Δq=0.969 e

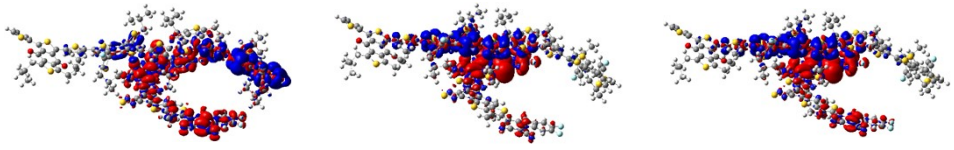


S0→S5

Δd=2.390 Å
Δq=-0.013 e

Δd=2.173 Å
Δq=-0.013 e

Δd=1.924 Å
Δq=-0.013 e



S0→S7

Δd=7.053 Å
Δq=0.966 e

Δd=5.027 Å
Δq=0.948 e

Δd=7.540 Å
Δq=0.947 e

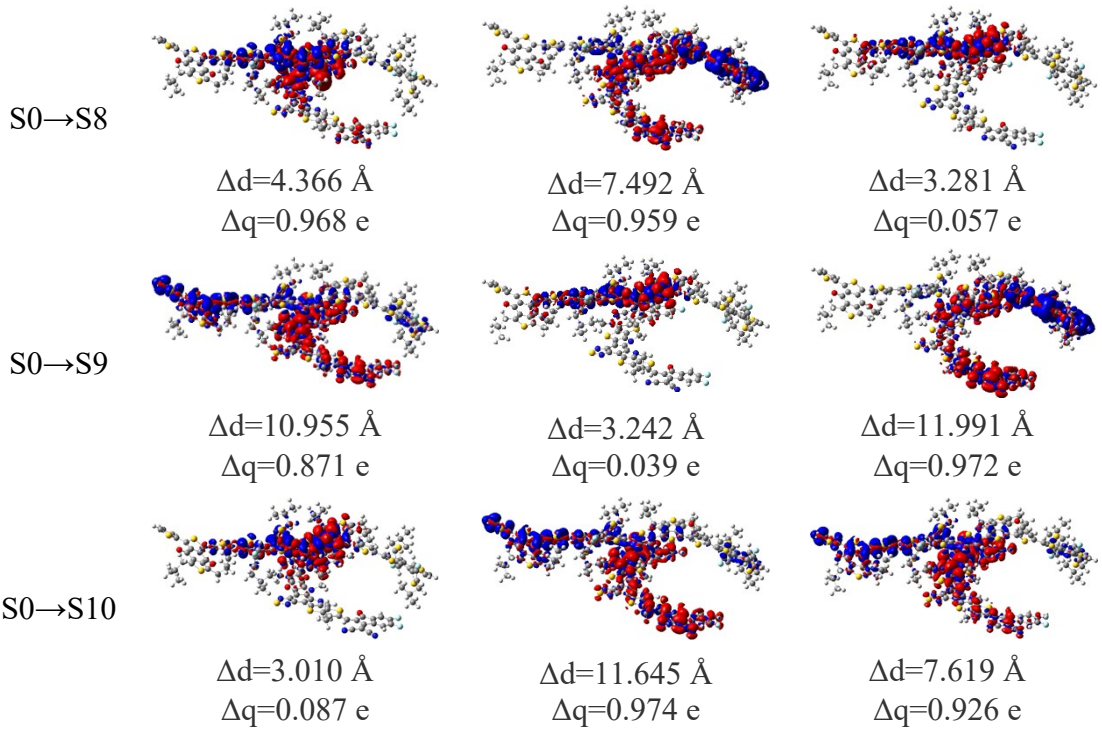


Figure S6. The charge density difference of the excited state of the complex. The red and blue areas indicate the increase and decrease of electron density during the excitation process. Δd represent the charge transfer distance. Δq represent the amount of charge transfer from the donor to acceptor.

Table S1. The selected bond length (in Å), bond angles and dihedral angles (in°) for the optimized eight monomers in the gas phase. (ωB97XD/CEP-121G*).

Definition	Bond length	Definition	Bond angles	Definition	Dihedral angles
			PM6		
38-47	1.512	38-47-48	114.5	27-26-29-30	-149.7
47-48	1.563	39-40-45	120.6	26-27-34-41	-15.0
40-45	1.512	40-45-46	116.8	24-28-37-42	16.3
45-46	1.555	35-34-41	122.9	36-30-45-46	122.4
34-41	1.228	36-37-42	124.0	35-38-48-47	-113.8
37-42	1.227	27-26-29	134.2	21-22-24-28	-138.9
26-29	1.471	22-24-28	130.4	23-19-12-75	143.4
22-24	1.470	12-19-23	117.5	5-1-6-7	-129.3
12-19	1.466	19-12-75	122.4	1-2-3-76	-177.8
1-6	1.483	1-6-11	121.6	5-4-43-44	62.4
3-76	1.355	2-1-6	127.8	8-9-14-15	60.7
4-43	1.511	2-3-76	120.9	10-9-14-18	64.2
43-44	1.549	3-4-43	127.1	14-15-14-77	-178.8
9-14	1.483	4-43-44	117.6	16-17-49-50	107.9
16-77	1.353	8-9-14	120.4	18-17-49-50	-68.7
17-49	1.508	9-14-15	127.5	72-13-109-108	146.5
49-50	1.552	15-16-17	121.2	107-106-103-104	-153.7
13-109	1.465	16-17-49	128.3	103-104-111-118	-16.7
103-106	1.469	17-49-50	113.8	101-105-114-119	16.5
111-118	1.229	74-13-109	120.3	112-115-124-125	90.1
114-119	1.227	110-106-103	125.1	113-117-122-123	121.1
115-124	1.512	104-111-118	122.4	98-99-101-102	52.1
124-125	1.563	105-114-119	121.4	97-96-89-149	143.3
117-122	1.511	115-124-125	114.6	84-83-78-82	-129.4
122-123	1.557	117-122-123	116.5	78-79-80-153	-177.8
99-101	1.470	105-101-99	130.0	82-81-120-121	62.7
89-96	1.467	89-96-97	131.6	86-86-91-92	65.0
86-91	1.484	87-86-91	122.1	85-86-91-95	-113.6
93-154	1.354	86-91-92	127.6	91-92-93-154	-178.7
94-126	1.508	92-93-154	121.2	93-94-126-127	109.1
126-127	1.552	94-126-127	113.6	80-81-120-121	-125.0
78-83	1.483	78-83-84	121.9	120-81-80-153	5.6
80-153	1.355	79-80-153	120.9	79-78-83-88	-129.2
81-120	1.511	81-120-121	117.6		
120-121	1.549				
Definition	Bond length	Definition	Bond angles	Definition	Dihedral angles
			Y6		
35-48	1.341	29-33-54	128.7	48-35-36-49	-0.0

36-49	1.341	29-30-52	128.2	32-33-54-60	179.2
33-54	1.378	20-28-29	134.4	33-29-28-20	-179.7
30-52	1.224	19-20-28	127.3	28-29-30-52	1.3
60-61	1.162	3-10-58	124.7	19-20-28-29	-2.6
62-63	1.161	2-15-56	127.2	17-21-78-77	-90.9
28-29	1.381	99-59-101	112.3	19-16-11-10	0.6
20-28	1.427	93-57-95	111.0	2-3-10-58	-37.4
21-78	1.511	23-89-88	122.5	3-2-15-56	-30.5
10-58	1.483	26-38-39	134.4	15-14-24-27	1.0
15-56	1.480	39-43-53	128.2	22-23-25-89	-3.6
26-38	1.426	39-40-55	128.7	26-38-39-40	179.2
38-39	1.381	41-40-55	124.2	38-39-43-53	0.9
43-53	1.224	44-45-50	119.5	39-40-55-64	179.9
40-55	1.378	47-46-51	120.6	39-43-42-47	179.9
64-65	1.161	64-55-66	112.6	40-41-44-45	-179.9
66-67	1.162	14-24-27	136.3	50-45-46-51	-0.0
45-50	1.341	11-16-19	136.2	9-6-1-13	5.4
46-51	1.341	60-54-62	112.6	7-5-4-12	6.0
55-64	1.441	59-101-100	114.5	3-10-58-59	-87.5
55-66	1.440	57-95-94	115.5	2-15-56-57	-85.6
25-89	1.511	59-99-98	115.7	18-17-16-19	179.9
21-78	1.511	57-93-92	114.4	22-23-24-27	-179.6
54-60	1.440	2-15-56	127.2		
54-62	1.441				

Definition	Bond length	Definition	Bond angles	Definition	Dihedral angles
			AQx-2		
32-45	1.341	32-33-46	118.3	45-32-33-46	-0.0
33-46	1.342	33-32-45	118.8	31-28-27-49	0.0
27-49	1.224	26-30-51	128.8	29-30-51-61	179.5
30-51	1.378	61-51-63	112.5	26-30-51-63	179.7
61-62	1.162	26-27-49	128.2	30-51-61-62	-176.5
63-64	1.161	17-25-26	134.5	27-26-25-17	-0.7
25-26	1.382	14-18-79	122.6	25-17-18-79	2.7
17-25	1.426	3-7-55	124.4	17-18-79-78	82.2
18-79	1.511	7-55-56	113.8	16-13-8-7	0.3
7-55	1.483	2-12-53	127.0	2-3-7-55	-38.4
12-53	1.479	12-53-54	114.7	3-2-12-53	-32.0
22-90	1.511	20-22-90	122.5	35-23-22-90	3.1
23-35	1.426	23-35-36	134.5	23-35-36-37	179.6
35-36	1.382	36-40-50	128.2	35-36-40-50	1.0
40-50	1.224	65-52-67	112.6	36-37-52-67	-0.6
37-52	1.378	36-37-52	128.7	37-52-67-68	-172.9
65-66	1.161	38-37-52	124.1	37-52-65-66	-178.5

67-68	1.162	42-43-48	118.8	41-38-37-52	0.8
42-47	1.341	43-42-47	118.3	47-42-43-48	-0.0
43-48	1.341	22-90-89	111.3	23-22-90-89	82.3
55-56	1.546	100-56-102	112.2	12-53-54-96	-177.2
53-54	1.546	94-54-96	111.1	7-55-56-100	53.9
3-7	1.396	2-3-7	130.7		
2-12	1.392				
Definition	Bond length	Definition	Bond angles	Definition	Dihedral angles
			Y6-T		
45-61	1.341	45-46-62	118.7	61-45-46-62	0.0
46-62	1.341	46-45-61	118.3	44-39-40-41	-0.2
40-41	1.223	39-40-41	125.3	37-42-104-105	178.8
37-42	1.375	36-37-42	128.5	37-42-106-107	178.2
104-105	1.161	104-42-106	112.8	34-33-35-36	-0.0
106-107	1.162	33-35-36	133.7	22-23-26-34	-27.5
35-36	1.376	23-26-34	121.8	19-20-22-91	1.6
33-35	1.433	11-21-24	137.4	7-3-2-12	-12.9
23-26	1.462	20-22-91	123.1	2-12-27-28	-91.1
12-27	1.483	22-91-90	113.4	3-7-29-30	-84.2
27-28	1.545	2-12-27	123.4	15-14-18-80	0.6
7-29	1.478	3-7-29	126.8	18-17-25-47	-28.5
29-30	1.546	14-18-80	123.1	49-48-51-52	179.8
22-91	1.511	18-80-79	113.0	51-52-53-68	0.1
18-80	1.511	17-25-47	121.7	57-54-53-68	-0.2
17-25	1.462	48-51-52	133.7	56-69-108-109	178.0
48-51	1.433	52-53-68	128.1	56-69-110-111	179.2
51-52	1.376	52-56-69	128.1	63-58-59-64	0.0
53-68	1.224	108-69-110	112.8	14-18-80-79	-91.8
56-69	1.375	58-59-64	118.3	20-22-91-90	-92.5
108-109	1.162	59-58-63	118.7	16-17-25-47	151.3
110-111	1.161	95-28-97	112.1	24-23-26-34	152.6
58-63	1.341	101-30-103	110.8		
59-64	1.341				
Definition	Bond length	Definition	Bond angles	Definition	Dihedral angles
			Y6-2T		
73-76	1.340	72-73-76	120.6	76-73-74-77	0.0
74-77	1.341	75-74-77	119.5	72-69-68-78	0.1
68-78	1.223	69-68-78	124.5	71-79-118-119	-177.0
71-79	1.375	70-71-79	124.2	71-79-720-721	-179.8
120-121	1.161	118-79-120	112.4	64-63-66-67	0.2
118-119	1.162	59-61-62	120.8	60-59-61-65	-163.7
66-67	1.376	60-59-61	121.0	16-17-25-60	130.6

63-66	1.437	17-25-60	121.7	25-17-18-90	2.1
59-61	1.461	16-17-25	117.5	15-14-13-16	-179.9
17-25	1.469	14-18-90	123.0	2-3-7-29	-33.0
7-29	1.477	3-7-29	126.5	3-2-12-27	-36.9
29-30	1.546	7-29-30	114.6	19-20-22-101	0.2
12-27	1.481	111-30-113	110.9	24-23-26-34	130.8
27-28	1.547	105-28-107	112.2	34-35-38-42	-165.8
18-90	1.511	2-12-27	124.0	40-41-43-44	-4.5
22-101	1.511	20-22-101	123.1	52-47-48-53	-0.5
23-26	1.469	22-23-26	129.5	45-54-114-115	-178.9
35-38	1.461	37-26-23	127.6	45-54-116-117	-177.9
41-43	1.437	34-35-38	120.9	55-50-51-56	-0.0
43-44	1.376	35-38-42	121.0	23-22-101-100	100.4
48-53	1.223	41-43-44	133.0	17-18-90-89	101.1
45-54	1.375	47-48-53	124.5	7-29-30-113	-177.1
114-115	1.161	46-45-54	124.2	12-27-28-107	-175.2
116-117	1.162	114-54-114	112.3	7-3-2-12	-12.8
50-55	1.341	55-50-51	118.3		
51-56	1.340				
Definition	Bond length	Definition	Bond angles	Definition	Dihedral angles
			Y6-O		
45-50	1.342	44-45-50	119.5	50-45-46-51	0.0
46-51	1.342	47-46-51	120.6	47-42-43-53	2.8
40-55	1.381	39-40-55	128.6	40-55-66-67	-179.0
64-65	1.162	64-55-66	112.5	40-55-64-65	179.0
66-67	1.162	26-38-39	133.8	27-26-38-39	-5.3
43-53	1.220	39-43-53	129.5	38-26-25-89	1.7
38-39	1.384	2-15-56	125.6	23-25-89-88	-94.5
26-38	1.408	15-56-57	113.1	22-23-24-27	-178.2
25-89	1.505	2-3-10	130.1	3-2-15-56	-35.6
15-56	1.484	3-10-58	117.9	2-15-56-57	-93.2
56-57	1.545	10-58-59	114.7	2-3-10-58	-31.0
24-27	1.333	17-21-78	122.6	3-10-58-29	-85.3
13-22	1.372	20-28-29	134.5	10-3-2-15	-12.7
10-58	1.480	29-30-52	128.1	18-17-16-19	-179.4
58-59	1.545	32-33-54	124.2	17-21-78-77	-93.9
12-18	1.751	60-54-62	112.6	21-20-28-29	179.0
16-19	1.731	34-35-48	120.6	28-20-21-78	3.1
21-78	1.511	37-36-49	119.5	28-29-30-52	0.7
28-29	1.381	25-89-88	111.8	33-54-60-61	-173.6
30-52	1.224	21-78-77	111.4	33-54-62-63	-178.4
33-54	1.377	93-57-95	112.4	48-35-36-49	-0.0
60-61	1.162	99-59-101	111.1	61-60-54-62	6.1

62-63	1.161	3-2-15	129.1	64-55-66-67	1.0
35-48	1.341	7-8-9	99.7		
36-49	1.341				
Definition	Bond length	Definition	Bond angles	Definition	Dihedral angles
			Y6-1O		
35-38	1.342	34-35-38	119.5	38-35-36-39	-0.0
36-39	1.342	37-36-39	120.5	30-41-49-50	-167.2
33-40	1.226	31-30-41	124.4	30-41-47-48	-177.4
30-41	1.378	47-41-49	112.7	37-32-33-40	-0.4
47-48	1.162	29-33-40	128.2	26-28-29-33	-6.6
49-50	1.162	26-28-29	133.4	24-23-25-42	-177.7
28-29	1.383	23-25-42	128.4	22-23-24-27	-177.6
26-28	1.418	25-42-84	119.3	3-2-15-43	-36.0
25-42	1.339	2-15-43	124.7	2-3-10-45	-30.4
15-43	1.483	15-43-44	113.6	18-17-16-19	-179.4
43-44	1.546	2-3-10	130.3	20-21-61-60	81.6
10-45	1.479	3-10-45	127.0	168-169-170-180	-0.1
45-46	1.546	10-45-46	114.6	21-20-168-169	179.5
21-61	1.511	21-61-60	111.6	173-181-182-183	-173.6
20-168	1.427	20-168-169	134.6	173-181-184-185	-177.8
168-169	1.380	171-170-180	125.3	171-174-175-178	-179.9
170-180	1.223	172-173-181	124.2	172-177-176-179	-180.0
182-183	1.162	182-181-184	112.5	178-175-176-179	-0.1
184-185	1.161	174-175-178	120.6	10-3-2-15	-13.9
175-178	1.341	177-176-179	119.5		
176-179	1.341				
Definition	Bond length	Definition	Bond angles	Definition	Dihedral angles
			Y6-2O		
35-48	1.342	34-35-48	120.6	48-35-36-49	-0.1
36-49	1.342	37-36-49	119.5	34-31-30-52	-0.1
30-52	1.226	32-33-54	124.3	33-54-62-63	-168.7
33-54	1.378	62-54-64	112.7	33-54-64-65	-176.6
62-63	1.162	31-30-52	125.3	19-20-28-29	-6.5
64-65	1.162	20-28-29	133.3	16-17-21-56	-177.1
28-29	1.383	17-21-56	128.5	17-21-56-103	-21.5
20-28	1.418	21-56-103	119.8	18-17-16-19	-178.4
21-56	1.338	11-10-60	119.7	10-3-2-15	-13.3
10-60	1.484	10-60-61	113.7	2-3-10-60	-36.9
60-61	1.546	79-61-81	112.3	3-2-15-58	-29.9
15-58	1.480	2-15-58	127.0	22-23-24-27	-177.9
58-59	1.546	73-59-75	111.1	23-25-57-92	-177.9
25-57	1.337	23-25-57	128.5	25-26-38-39	177.8

26-38	1.418	25-57-92	119.7	47-42-43-53	-0.2
38-39	1.384	26-38-39	133.4	40-55-66-67	-177.0
43-53	1.226	42-43-53	125.2	40-55-68-69	-170.9
40-55	1.378	40-41-44	131.8	50-45-46-51	-0.1
66-67	1.162	66-55-68	112.7	3-10-60-61	-87.5
68-69	1.162	44-45-50	119.5	2-15-58-59	-86.2
45-50	1.342	47-46-51	120.6		
46-51	1.3420				

Table S2. The absorption peak wavelength and energy of donor PM6 and acceptor Y6, AQx-2, Y6-1O, Y6-2O.

	Experiment (nm/eV)	Theoretical (nm/eV)	Difference (eV)
PM6	616.81/2.010	500.61/2.477	0.467
Y6	816.00/1.519	689.91/1.7971	0.278
AQx-2	830.00/1.494	684.43/1.8115	0.318
Y6-1O	802.08/1.546	670.95/1.8479	0.302
Y6-2O	811.43/1.528	647.64/1.9144	0.386

Table S3. Electronic transition energies (eV), excitation wavelengths (nm), corresponding oscillator strengths (*f*), and main transition configurations for eight monomer molecules. CT represent the charge transfer from the donor segment to the acceptor segment. DLE and ALE represent the local excitation that occurs on the donor segment and the acceptor segment. (In the solid phase, LC-PBE/CEP-121G*).

States	Main transition configurations	ESC	E (eV/nm)	*f*
PM6 (ω=0.118, ε_s=3.1, ε_d=3.0)				
S2	H-1→L+1 (33%); H→L+1 (42%)	CT	2.6692/464.5	0.1717
S3	H→L+2 (56%); H→L+3 (11%)	CT	2.7174/456.27	0.0218
S4	H-1→L (62%); H-1→L+2 (11%)	CT	2.8403/436.52	0.1326
S5	H-1→L+1 (57%); H→L+1 (28%)	CT	2.8805/430.42	0.0364
S6	H-3→L+3 (10%); H→L+2 (10%)	CT&ALE	2.8936/428.48	0.1365
S8	H-3→L (36%); H-3→L+2 (19%); H-2→L+2 (15%) H→L+2 (10%)	CT&LE	3.0747/403.24	0.0789
S9	H-4→L+1 (17%); H-3→L+1 (12%)	CT	3.1237/396.92	0.1437
S10	H-3→L+3 (23%)	CT&LE	3.1377/395.14	0.1073
S12	H-1→L+2 (28%); H→L+4 (20%)	CT&LE	3.215/385.65	0.1286
S13	H-2→L+1 (17%); H-1→L+5 (24%)	CT&DLE	3.236/383.14	0.0727
S14	H→L+4 (11%)	LE	3.2437/382.23	0.0516
Y6 (0.118, ε_s=3.7, ε_d=3.5)				
S3	H-1→L (92%)	CT&DLE	2.4228/511.74	0.0101
S4	H→L+2 (94%)	CT	2.4296/510.31	0.0865
S5	H→L+3 (88%)	CT	2.5196/492.08	0.1197

S6	H-1→L+1 (52%); H-3→L (44%)	CT&DLE	2.5577/484.75	0.1023
S8	H-3→L (46%); H-1→L+1 (42%)	CT&DLE	2.7737/447.00	0.0222
S10	H→L+4 (77%); H-2→L+1 (20%)	DLE	2.8802/430.47	0.0120
S11	H-3→L+1 (85%)	CT	2.9113/425.87	0.0134
S12	H-1→L+2 (81%); H-3→L+3 (10%)	CT	3.1043/399.40	0.0048
S13	H-1→L+3 (73%); H-3→L+2 (18%)	CT	3.1409/394.74	0.0484
S14	H-2→L+2 (83%)	CT	3.1840/389.40	0.0524
S15	H-2→L+3 (79%)	CT	3.2625/380.03	0.0120
	AQx-2 (0.118, ε_s=3.6, ε_d=3.5)			
S3	H-1→L (94%)	CT&DLE	2.3738/522.30	0.0101
S4	H→L+2 (93%)	CT	2.4398/508.17	0.0948
S5	H→L+3 (45%); H-1→L+1 (32%); H-3→L (15%)	CT&DLE	2.5214/491.73	0.1251
S6	H→L+3 (44%); H-1→L+1 (33%); H-3→L (16%)	CT	2.5239/491.24	0.1419
S8	H-3→L (64%); H-1→L+1 (31%)	CT	2.7632/448.70	0.0021
S10	H-3→L+1 (85%)	CT	2.9029/427.10	0.0133
S11	H→L+4 (94%)	CT	2.9933/414.21	0.0024
S12	H-1→L+2 (84%)	CT	3.0610/405.04	0.0037
S13	H-1→L+3 (78%); H-3→L+2 (14%)	CT	3.0976/400.26	0.0515
S14	H-2→L+2 (83%)	CT	3.1816/389.69	0.0591
S15	H-2→L+3 (79%)	CT	3.2543/380.99	0.0170
	Y6-T (ω=0.118, ε_s=3.7, ε_d=3.5)			
S2	H→L+1 (98%)	CT	1.7683/701.15	0.1657
S4	H→L+3 (94%)	CT	2.2633/547.80	0.0090
S5	H-1→L (93%)	CT&DLE	2.3320/531.66	0.1190
S7	H-2→L (90%)	CT	2.4763/500.68	0.0343
S8	H-2→L+1 (79%); H-3→L (11%)	CT	2.5316/489.75	0.0143
S9	H→L+4 (88%)	DLE&CT	2.6224/472.79	0.0636
S10	H-3→L (77%); H-2→L+1 (13%)	CT	2.7494/450.95	0.0536
S11	H-3→L+1 (84%)	CT	2.8016/442.55	0.0063
S12	H-1→L+2 (81%)	CT	2.8710/431.85	0.0018
S13	H-1→L+3 (74%)	CT	2.8987/427.72	0.0542
S15	H-4→L+1 (79%)	CT&DLE	3.0035/412.80	0.0224
	Y6-2T (0.118, ε_s=3.8, ε_d=3.5)			
S2	H→L+1 (97%)	CT	1.6259/762.56	0.2180
S5	H→L+3 (81%); H-1→L (12%)	CT	2.2171/559.22	0.0214
S6	H-1→L+1 (48%); H→L+2 (40%)	DLE&CT	2.2214/558.14	0.2683
S7	H-2→L (88%)	CT	2.4391/508.32	0.0206
S8	H-2→L+1 (85%)	CT	2.4499/506.08	0.0282
S10	H-3→L (70%); H-1→L+1 (11%)	CT&DLE	2.5735/481.77	0.1381
S11	H-3→L+1 (73%); H-1→L (13%)	DLE&CT	2.5844/479.74	0.1240
S12	H-4→L (78%); H-4→L+1 (13%)	CT	2.7102/457.47	0.0013
S13	H-4→L+1 (68%); H-4→L (12%)	CT	2.7246/455.05	0.0018
S14	H-1→L+2 (60%); H-3→L+3 (13%); H-4→L+1 (13%)	CT&DLE	2.7343/453.44	0.0756
S15	H-1→L+3 (67%); H-3→L+2 (16%)	CT	2.7401/452.48	0.3334

Y6-O (0.118, ε_s=3.7, ε_d=3.5)				
S3	H→L+2 (86%)	CT	2.4389/508.36	0.0632
S4	H-2→L (65%); H-1→L (27%)	CT&DLE	2.4841/499.11	0.0374
S5	H→L+3 (51%); H-1→L (26%); H-2→L (15%)	CT&DLE	2.5289/490.27	0.0226
S8	H→L+4 (55%); H-2→L+1 (23%); H-1→L+1 (15%)	CT&DLE	2.8307/438.00	0.0274
S9	H→L+4 (42%); H-2→L+1 (35%); H-1→L+1 (17%)	CT&DLE	2.8770/430.95	0.0630
S10	H-3→L (86%)	CT&DLE	3.0419/407.59	0.0274
S11	H-1→L+2 (76%)	CT	3.1263/396.58	0.0494
S12	H-2→L+2 (80%)	CT	3.1768/390.28	0.0141
S13	H-7→L (42%); H-7→L+1 (21%); H-7→L+3 (10%)	CT&ALE	3.2129/385.89	0.0052
S14	H-3→L+1 (64%); H-4→L (11%)	CT&DLE	3.2731/378.80	0.0103
S15	H-1→L+3 (42%); H-4→L (20%)	CT&DLE	3.2879/377.09	0.0238
Y6-1O (0.118, ε_s=3.6, ε_d=3.5)				
S3	H-1→L (35%); H→L+2 (30%); H-2→L (22%)	CT&DLE	2.4591/504.19	0.0927
S4	H→L+2 (55%); H-1→L (20%); H-2→L (14%)	CT&DLE	2.4629/503.41	0.0447
S7	H-3→L (40%); H-1→L+1 (27%); H-2→L+1 (14%)	CT	2.6701/464.34	0.0508
S8	H-1→L+1 (57%); H-2→L+1 (19%); H-3→L (14%)	CT&DLE	2.8008/442.67	0.1200
S9	H-2→L+1 (56%); H-3→L (33%)	CT&DLE	2.8532/434.54	0.0339
S10	H→L+4 (93%)	CT&DLE	2.8789/430.67	0.0036
S11	H-3→L+1 (77%)	CT	3.0141/411.35	0.0456
S12	H-1→L+2 (66%)	CT	3.1109/398.55	0.0504
S13	H-2→L+2 (70%)	CT	3.1361/395.35	0.0144
S14	H-1→L+3 (73%)	CT	3.1913/388.51	0.0045
S15	H-4→L (84%)	DLE&CT	3.2423/382.40	0.0280
Y6-2O (0.118, ε_s=3.6, ε_d=3.5)				
S3	H-1→L (83%)	DLE&CT	2.4845/499.03	0.1684
S4	H→L+2 (85%)	CT	2.4949/496.95	0.0585
S6	H-2→L (84%)	DLE&CT	2.6555/466.90	0.1097
S7	H-3→L (48%); H-1→L+1 (29%); H-2→L+1 (12%)	CT&DLE	2.6741/463.65	0.0431
S9	H→L+4 (95%)	CT&DLE	2.8818/430.23	0.0018
S10	H-2→L+1 (68%); H-3→L (25%)	CT&DLE	2.9318/422.89	0.0169
S11	H-3→L+1 (71%); H-1→L+2 (14%)	CT&DLE	3.0263/409.69	0.0266
S12	H-1→L+2 (68%); H-3→L+1 (11%)	CT	3.0799/402.56	0.0125
S13	H-1→L+3 (78%)	CT	3.1567/392.77	0.0042
S14	H-2→L+2 (75%); H-3→L+3 (10%)	CT	3.2507/381.41	0.0022
S15	H-4→L (86%)	CT&DLE	3.2630/379.97	0.0038

Table S4. The lowest singlet excitation energy E_{S1}(eV), the lowest triplet excitation energy E_{T1}(eV), and singlet–triplet energy gap ΔE_{ST} ($\Delta E_{ST} = E_{S1} - E_{T1}$) for PM6 and NFAs calculated at the given ω value (in Bohr^{-1}), static and dynamic dielectric constants. (In the solid, LC-PBE/CEP-121G*).

molecule	E_{S1}	E_{T1}	ΔE_{ST}	ω	ε_s	ε_d
PM6	2.4767	1.7650	0.7117	0.118	3.1	3.0
Y6	1.7971	1.3425	0.4546	0.118	3.7	3.5
AQx-2	1.8115	1.3557	0.4558	0.118	3.6	3.5
Y6-T	1.6302	1.3090	0.3212	0.118	3.7	3.5
Y6-2T	1.5865	1.3563	0.2302	0.118	3.8	3.5
Y6-O	1.7783	1.2816	0.4967	0.118	3.7	3.5
Y6-1O	1.8479	1.3812	0.4667	0.118	3.6	3.5
Y6-2O	1.9144	1.4247	0.4897	0.118	3.6	3.5

Table S5. Electronic transition energies (eV), excitation wavelengths (nm), corresponding oscillator strengths (*f*), and main transition configurations for eight monomer molecules. CT represent the charge transfer from the donor segment to the acceptor segment. DLE and ALE represent the local excitation that occurs on the donor segment and the acceptor segment. (In the solid phase, LC-PBE/CEP-121G*).

States	Main transition configurations	ESC	E (eV/nm)
PM6 (ω=0.118, ε_s=3.1, ε_d=3.0)			
T1	H→L (57%); H→L+4 (11%)	LE	1.7650/702.45
T2	H-1→L+1 (20%); H-1→L+5 (21%)	CT&DLE	1.9665/630.47
T3	H-3→L (13%); H→L+5 (12%)	CT&ALE	2.0562/602.98
Y6 (ω=0.118, ε_s=3.7, ε_d=3.5)			
T1	H→L (81%); H-2→L+1 (11%)	DLE&CT	1.3425/923.53
T2	H→L+1 (58%); H-2→L (31%)	DLE&CT	1.5480/800.93
T3	H→L+2 (41%); H-2→L+1 (15%); H-4→L (15%)	CT&DLE	2.0513/604.42
AQx-2 (ω=0.118, ε_s=3.6, ε_d=3.5)			
T1	H→L (81%); H-2→L+1 (12%)	DLE&CT	1.3557/914.54
T2	H→L+1 (58%); H-2→L (32%)	CT&DLE	1.5430/803.53
T3	H→L+2 (41%); H-2→L+1 (14%)	CT&DLE	2.0660/600.12
Y6-T (ω=0.118, ε_s=3.7, ε_d=3.5)			
T1	H→L (68%); H-1→L+1 (14%)	CT&DLE	1.3090/947.17
T2	H→L+1 (60%); H-1→L (23%)	CT&DLE	1.3842/895.71
T3	H-1→L+1 (22%); H-4→L (20%); H→L+4 (20%)	CT&DLE	1.8768/660.61
Y6-2T (ω=0.118, ε_s=3.8, ε_d=3.5)			
T1	H→L (45%); H-1→L+1 (19%); H-3→L (13%)	CT&DLE	1.3563/914.14
T2	H→L+1 (41%); H-1→L (22%); H-3→L+1 (14%)	CT&DLE	1.3736/902.62
T3	H→L (39%); H-3→L (19%); H-1→L+1 (14%)	CT&DLE	1.7116/724.38
Y6-O (ω=0.118, ε_s=3.7, ε_d=3.5)			
T1	H→L (76%)	DLE&CT	1.2816/967.42

T2	H→L+1 (50%); H-1→L (28%)	DLE&CT	1.5248/813.12
T3	H→L+2 (28%); H→L+3 (19%); H-1→L+1 (13%); H-4→L (13%)	CT&DLE	2.0406/607.59
	Y6-1O (ω=0.118, ε_s=3.6, ε_d=3.5)		
T1	H→L (76%)	DLE&CT	1.3812/897.66
T2	H→L+1 (51%); H-1→L (22%)	CT&DLE	1.5721/788.65
T3	H→L+2 (29%); H-4→L (13%); H→L+3 (11%); H-1→L+1 (10%)	CT&DLE	2.0454/606.16
	Y6-2O (ω=0.118, ε_s=3.6, ε_d=3.5)		
T1	H→L (77%); H-1→L+1 (13%)	DLE&CT	1.4247/870.25
T2	H→L+1 (46%); H-1→L (38%)	CT&DLE	1.5825/783.47
T3	H→L+2 (43%); H-1→L+1 (15%); H-4→L (11%)	CT&DLE	2.0412/607.41

Table S6. Electronic transition energies(eV), corresponding oscillator strengths (f), excitation wavelengths(nm), excited states characters (ESC) and main transition configurations for complex molecules. (LC-PBE/CEP-121G*).

States	Main transition configurations	ESC	E(eV/nm)	f
	PM6:Y6 (ω=0.118, ε_s=3.4, ε_d=3.3)			
S3	H→L+1 (56%); H→L (30%)	CT	2.0059/618.10	0.0082
S4	H-1→L (54%); H-2→L (17%)	CT	2.0193/614.00	0.0290
S5	H-2→L+1 (74%); H-1→L+1 (22%)	ALE&CT	2.1361/580.42	0.3159
S6	H-3→L (32%); H-4→L (20%); H-1→L (14%); H-3→L+1 (13%)	CT	2.1588/574.32	0.0026
S7	H→L+3 (54%); H→L+2 (32%)	CT	2.2286/556.33	0.0173
S8	H-1→L+1 (59%); H-2→L+1 (21%); H-3→L (10%)	CT	2.2718/545.75	0.0010
S9	H-4→L (40%); H-3→L (25%)	CT	2.3233/533.66	0.1085
S11	H-2→L+2 (72%); H-1→L+2 (20%)	CT	2.4398/508.17	0.0843
S12	H-3→L+1 (36%); H-3→L (20%); H-4→L+1 (13%); H-4→L (12%)	CT	2.5056/494.83	0.0004
S13	H-7→L (87%)	CT	2.5122/493.53	0.0266
S14	H-2→L+3 (53%); H-1→L+3 (22%)	CT	2.5372/488.67	0.1288
S15	H-5→L (33%); H-3→L+1 (13%); H-2→L+3 (12%)	CT	2.5579/484.71	0.1021
	PM6:AQx-2 (ω=0.118, ε_s=3.4, ε_d=3.3)			
S3	H→L+1 (59%); H→L (34%)	CT	2.0308/610.52	0.0167
S4	H-2→L (57%); H-2→L+1 (10%); H-4→L (10%)	CT	2.0811/595.76	0.0280
S5	H-1→L+1 (93%)	ALE	2.1413/579.01	0.3046
S6	H-2→L (33%); H-3→L (30%); H-3→L+1 (13%); H-4→L (12%)	CT	2.2096/561.12	0.0025
S7	H→L+3 (56%); H→L+2 (28%)	CT	2.2435/552.64	0.0283
S8	H-2→L+1 (43%); H-3→L (21%); H-4→L (11%)	CT	2.3412/529.58	0.0754
S10	H-4→L (34%); H-2→L+1 (34%)	CT	2.3743/522.19	0.0391
S11	H-1→L+2 (90%)	ALE	2.4556/504.90	0.0955
S12	H-6→L (90%)	CT	2.4791/500.12	0.0263

S13	H-3→L+1 (45%); H-3→L (23%)	CT	2.5391/488.30	0.0006
S14	H-1→L+3 (82%)	ALE	2.5480/486.59	0.1915
S15	H→L+2 (56%); H→L+3 (30%)	CT	2.5954/477.71	0.0324
	PM6:Y6-T (ω=0.118, ε_s=3.4, ε_d=3.3)			
S3	H→L+1 (91%)	ALE	1.8356/675.44	0.2153
S4	H-1→L (74%); H-1→L+1 (25%)	CT	2.0495/604.95	0.0009
S5	H-2→L+1 (53%); H-2→L (27%)	CT	2.0812/595.73	0.0051
S6	H-3→L+1 (31%); H-4→L+1 (16%); H-2→L (13%), H-3→L (12%)	CT	2.2224/557.88	0.0087
S7	H-1→L+3 (77%); H-1→L+2 (10%)	CT	2.2352/554.69	0.0090
S8	H→L+2 (85%)	ALE	2.2726/545.56	0.3507
S9	H-2→L (57%); H-2→L+1 (39%)	CT	2.2825/543.19	0.0016
S10	H→L+3 (76%)	ALE	2.3375/530.41	0.1562
S12	H-5→L (45%); H-4→L (24%)	CT	2.3668/523.85	0.0522
S13	H-5→L (27%); H-4→L+1 (23%); H-3→L+1 (21%); H-3→L (10%)	CT	2.3940/517.90	0.0410
S14	H-5→L+1 (67%); H-4→L+1 (11%)	ALE	2.4585/504.31	0.4771
S15	H→L+4 (90%)	CT	2.5208/491.84	0.0029
	PM6:Y6-2T (ω=0.118, ε_s=3.5, ε_d=3.3)			
S2	H-1→L+1 (38%); H→L+1 (30%); H→L (27%)	CT&ALE	1.6410/755.54	0.1573
S3	H-1→L+1 (50%); H→L+1 (42%)	ALE&CT	1.7027/728.16	0.3361
S5	H-2→L+1 (79%)	CT	2.0532/603.86	0.0169
S6	H-3→L (44%); H→L+3 (17%); H→L+2 (12%)	ALE	2.1570/574.80	0.9336
S7	H→L+3 (35%); H-3→L (25%); H-1→L+3 (24%)	ALE	2.1782/569.20	0.3291
S8	H-3→L+1 (27%); H-4→L+1 (24%); H-5→L+1 (13%); H-2→L+1 (12%); H-2→L (10%)	CT&ALE	2.2132/560.20	0.0854
S9	H-2→L (63%); H→L+2 (25%)	CT&ALE	2.2232/557.68	0.0681
S10	H→L+2 (45%); H-2→L (26%)	CT&ALE	2.2271/556.71	0.0222
S11	H-1→L+3 (33%); H→L+3 (21%); H-3→L+1 (13%); H→L+2 (10%); H-4→L+1 (10%)	ALE	2.2526/550.40	0.3996
S12	H-3→L+1 (36%); H-1→L+3 (17%); H-4→L+1 (16%); H→L+3 (14%)	CT	2.2783/544.20	0.1010
S14	H-5→L+1 (58%); H-4→L+1 (26%)	CT	2.3958/517.51	0.0344
S15	H→L+4 (75%); H-1→L+4 (19%)	CT	2.3993/516.75	0.1055
	PM6:Y6-O (ω=0.118, ε_s=3.4, ε_d=3.3)			
S3	H-2→L (45%); H→L+1 (26%); H→L (22%)	CT	2.0066/617.88	0.0056
S4	H-2→L (42%); H→L+1 (33%); H→L (14%)	CT	2.0426/606.99	0.0344
S5	H-1→L+1 (91%)	ALE	2.1373/580.10	0.3003
S6	H-4→L (34%); H-4→L+1 (16%); H-2→L+1 (15%); H-3→L (14%)	CT	2.1988/563.87	0.0030
S7	H-2→L+1 (72%); H-3→L (11%)	CT	2.2903/541.34	0.0059
S8	H→L+3 (71%); H→L+2 (15%)	CT	2.3106/536.59	0.3147
S9	H-3→L (47%); H-4→L (29%)	CT	2.3537/526.76	0.0074
S11	H-1→L+2 (81%)	CT	2.4257/511.13	0.0763

S12	H-4→L+1 (26%); H-5→L (26%); H-4→L (22%)	CT	2.5170/492.59	0.0046
S13	H-1→L+3 (56%)	CT&ALE	2.5419/487.76	0.1042
S14	H-8→L (83%)	CT	2.5637/483.61	0.0301
S15	H→L+2 (64%); H→L+3 (14%)	CT	2.5665/483.09	0.0034
	PM6:Y6-1O (ω=0.118, ε_s=3.4, ε_d=3.3)			
S3	H→L+1 (51%); H→L (42%)	CT	2.0133/615.83	0.0198
S4	H-1→L (81%)	CT	2.0876/593.91	0.0134
S5	H-2→L+1 (82%)	ALE	2.1957/564.67	0.3666
S6	H-3→L (28%); H-3→L+1 (19%); H-1→L+1 (15%); H-4→L (13%); H-4→L+1 (10%)	CT	2.2400/553.50	0.0101
S7	H→L+3 (68); H→L+2 (16%)	CT	2.3091/536.94	0.0432
S8	H-1→L+1 (62%; H-3→L (17%)	CT	2.3453/528.65	0.0202
S10	H-4→L (56%; H-3→L (22%)	CT	2.4194/512.46	0.0241
S11	H-2→L+2 (76%)	CT	2.4640/503.18	0.0832
S12	H-6→L (74%)	ALE	2.4957/496.79	0.2154
S13	H-3→L+1 (37%); H-3→L (24%); H-4→L (13%); H-4→L+1 (12%)	CT	2.5227/491.47	0.0088
S14	H→L+2 (75%); H→L+3 (15%)	CT	2.5557/485.13	0.0064
S15	H-8→L (50%); H-2→L+3 (22%)	ALE&CT	2.5765/481.21	0.1189
	PM6:Y6-2O (ω=0.118, ε_s=3.4, ε_d=3.3)			
S3	H→L+1 (56%); H→L (29%)	CT	2.1275/582.77	0.0213
S4	H-1→L (71%)	CT	2.1402/579.31	0.0307
S5	H-2→L+1 (82%)	ALE	2.2405/553.38	0.3932
S6	H-3→L (36%); H-1→L (17%); H-4→L (16%); H-3→L+1 (14%)	CT	2.2824/543.22	0.0155
S7	H→L+3 (43%); H→L+2 (34%)	CT	2.3270/532.81	0.0325
S9	H-1→L+1 (78%)	CT	2.4014/516.30	0.0019
S10	H-4→L (50%); H-3→L (22%)	CT	2.4591/504.19	0.0166
S11	H-2→L+2 (77%)	CT	2.5003/495.88	0.0584
S12	H-5→L (79%)	ALE	2.5076/494.43	0.2069
S13	H-2→L+3 (69%)	ALE	2.6207/473.10	0.3852
S14	H-3→L+1 (28%); H-3→L (16%); H-4→L+1 (11%)	CT	2.6283/471.73	0.1736
S15	H→L+6 (29%); H→L+8 (16%); H→L+5 (15%)	CT	2.6375/470.08	0.0132

Table S7. Calculated ionization potentials of the donors IP_D, electron affinities of the acceptor EA_A, fundamental gaps defined as the difference between the ionization energy of donor and the electron affinities of acceptors ($IP_{D-}EA_A$), first adiabatic excitation energy of acceptor E_{s1A}, quality center distances D_{D-A} (Å) between donor and acceptor of the complexes. (LC-PBE/CEP-121G*).

Complex	IP_D	EA_A	$IP_{D-}EA_A$	E_{s1A}	D_{D-A}
PM6:Y6	5.53	3.64	1.89	1.70	10.40
PM6:AQx-2	5.53	3.60	1.93	1.72	9.26

PM6:Y6-T	5.53	3.55	1.98	1.54	12.45
PM6:Y6-2T	5.53	3.55	1.98	1.43	16.16
PM6:Y6-O	5.53	3.66	1.87	1.68	9.23
PM6:Y6-1O	5.53	3.60	1.93	1.75	8.98
PM6:Y6-2O	5.53	3.49	2.04	1.82	9.09

Table S8. Calculated electronic coupling V (in eV) in CR process of PM6:Y6, PM6:AQx-2, PM6:Y6-T, PM6:Y6-2T, PM6:Y6-O, PM6:Y6-1O and PM6:Y6-2O complexes. u corresponds the transition dipole moment (in a.u); μ_g and μ_e represent the dipole moment at the ground state and excited state (in Debye), respectively. $\Delta\mu_{ge}$ indicates the change in dipole moment between the ground state and the lowest singlet excited state (in Debye); ΔE is the energy difference between the ground state and the excited state (in eV). (LC-PBE/CEP-121G*).

		PM6:Y6	PM6:AQx-2	PM6:Y6-T	PM6:Y6-2T
u	X	-0.8985	0.8869	0.9108	5.5281
	Y	-0.1743	0.1925	1.5055	0.6243
	Z	0.0255	-0.0296	-0.2514	-0.4791
U_g	X_1	0.9169	1.9348	1.9922	0.0079
	Y_1	-0.9119	-0.7852	-2.1505	-6.8412
	Z_1	-1.7875	-1.9649	-3.4203	-0.9021
U_e	X_2	18.4787	18.5420	-1.7888	29.6311
	Y_2	-15.1217	-15.3331	10.6779	-18.1627
	Z_2	6.0141	5.9835	4.4665	-7.0744
u		24.91	24.71	48.37	151.94
$\Delta\mu_{ge}$		255.84	251.19	166.20	345.85
ΔE		1.62	1.63	1.65	1.60
V_{CR}		0.15	0.16	0.42	0.53

		PM6:Y6-O	PM6:Y6-1O	PM6:Y6-2O
u	X	0.8744	-0.9060	-0.9638
	Y	-0.2730	-0.1462	-0.1805
	Z	-0.0420	0.0159	0.0217
U_g	X_1	0.1663	3.0632	6.0785
	Y_1	0.7895	1.3983	0.8071
	Z_1	2.4251	-1.7092	-1.4949
U_e	X_2	-19.0055	21.1463	22.1500
	Y_2	-14.0973	-18.7223	-13.6812
	Z_2	-4.8883	6.0561	6.6037
u		24.95	24.98	26.69
$\Delta\mu_{ge}$		271.37	301.28	247.32
ΔE		1.66	1.72	1.74
V_{CR}		0.15	0.14	0.18

$$\Delta\mu_{ge} = \sqrt{(X_2 - X_1)^2 + (Y_2 - Y_1)^2 + (Z_2 - Z_1)^2} \qquad u = \sqrt{X^2 + Y^2 + Z^2}$$

Table S9. H_{if} (meV) is the off-diagonal Hamiltonian of charge-localized state, S_{if} corresponds the overlap matrix element, E_i and E_f are the energy (in eV) of initial and final states, and electronic coupling V (in eV) in CT and ED process of complexes. (LC-PBE/CEP-121G*).

Complex	ETP	H_{if}	S_{if}	E_i	E_f	V
PM6:Y6	CT D(H)→A(L)	71.66214	0.02416	2.65533	6.02695	-0.03324
	ED A(H)→D(H)	2.21414	0.01046	2.65538	5.94924	-0.04279
PM6:AQx-2	CT D(H)→A(L)	-10.41090	-0.00138	2.79212	5.90054	-0.00402
	ED A(H)→D(H)	-4.32000	0.00146	2.79214	5.77554	0.01057
PM6:Y6-T	CT D(H)→A(L)	-3.93131	0.00395	2.65876	5.00972	-0.01908
	ED A(H)→D(H)	-16.57593	-0.00248	2.65868	5.02722	0.00706
PM6:Y6-2T	CT D(H)→A(L)	-4.53693	0.00790	2.64575	4.21039	-0.03161
	ED A(H)→D(H)	-1.31071	-0.00381	2.64556	4.28051	-0.01189
PM6:Y6-O	CT D(H)→A(L)	-58.87923	-0.01787	2.62604	6.09047	0.01900
	ED A(H)→D(H)	59.84709	0.00666	2.62573	6.06151	-0.03093
PM6:Y6-1O	CT D(H)→A(L)	85.03574	0.04032	2.65182	5.86140	-0.08660
	ED A(H)→D(H)	-21.30855	-0.00017	2.64963	5.96874	0.02056
PM6:Y6-2O	CT D(H)→A(L)	-38.40091	-0.02356	2.65048	5.97408	0.06319
	ED A(H)→D(H)	-7.74632	-0.00745	2.64959	5.94901	-0.02426