

## Supplementary Material

### Diradical Character of Benzo- and Naphtho-Annulated Thiophene-Pyrrole Mixed Oligomer Dications

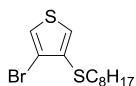
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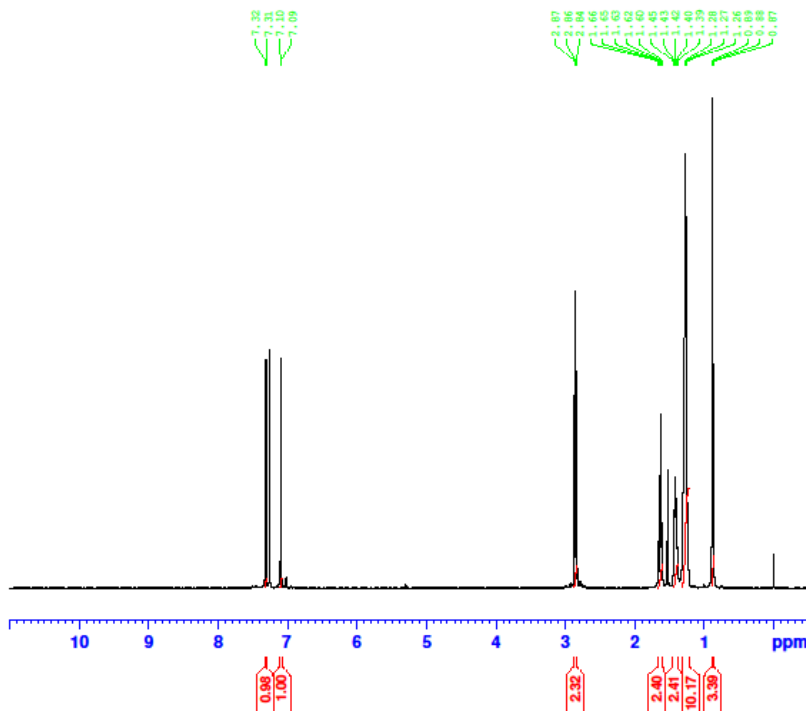
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<sup>1</sup>H NMR spectrum of **1**



<sup>1</sup>H



Current Data Parameters  
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PROCNO 1

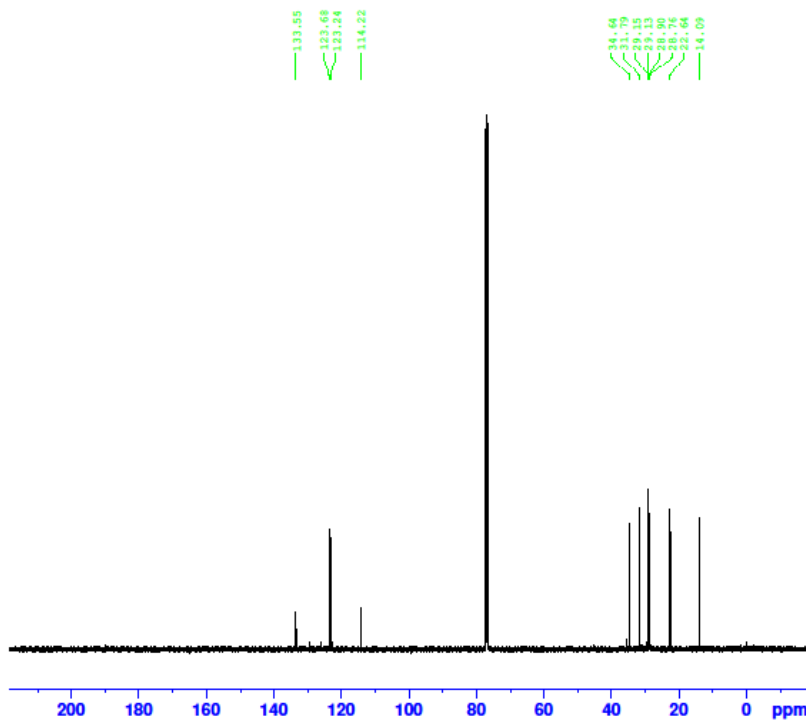
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SOLVENT CDCl3  
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DS 2  
SWH 10330.578 Hz  
FIDRES 0.157632 Hz  
AQ 3.1719425 sec  
RG 128  
EW 48.400 usec  
DE 6.50 usec  
TE 298.2 K  
D1 1.00000000 sec  
TDO 1

CHANNEL f1  
NUC1 1H  
P1 12.10 usec  
PL1 1.00 dB  
PL1W 15.90224075 W  
SF01 500.1330885 MHz

F2 - Processing parameters  
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SF 500.1330130 MHz  
WDW EM  
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LB 0.30 Hz  
GB 0  
PC 1.00

<sup>13</sup>C NMR spectrum of **1**

<sup>13</sup>C



Current Data Parameters  
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EXNO 3  
PROCNO 1

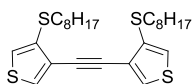
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FIDRES 0.454131 Hz  
AQ 1.1010048 sec  
RG 203  
EW 16.800 usec  
DE 6.50 usec  
TE 298.2 K  
D1 2.00000000 sec  
D11 0.03000000 sec  
TDO 1

CHANNEL f1  
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PL1 -2.50 dB  
PL1W 142.49206543 W  
SF01 125.7703643 MHz

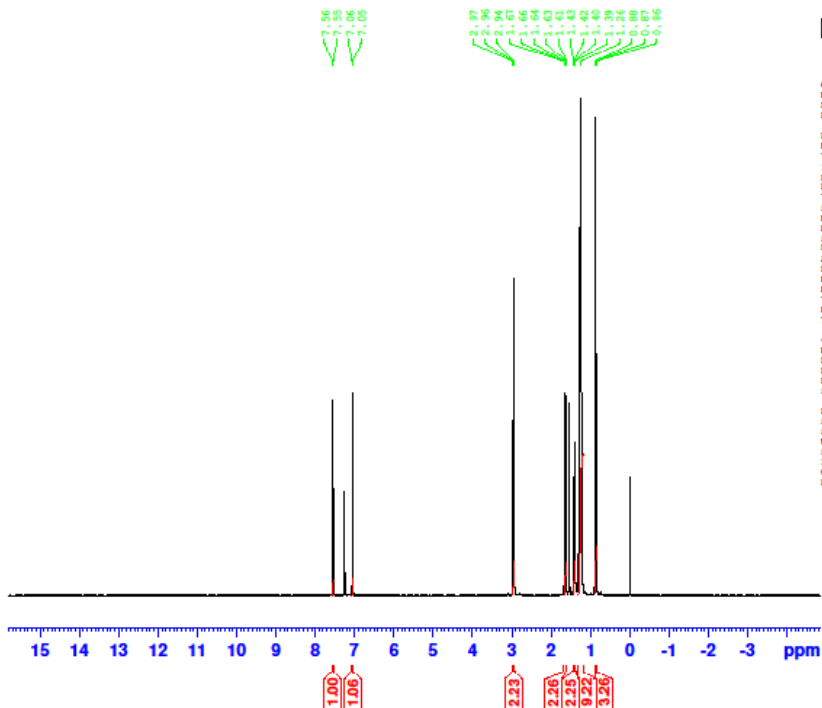
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PL12W 0.35600683 W  
PL13W 0.35600683 W  
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F2 - Processing parameters  
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<sup>1</sup>H NMR spectrum of 2



<sup>1</sup>H



```

Current Data Parameters
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EXPNO 1
PROCNO 1

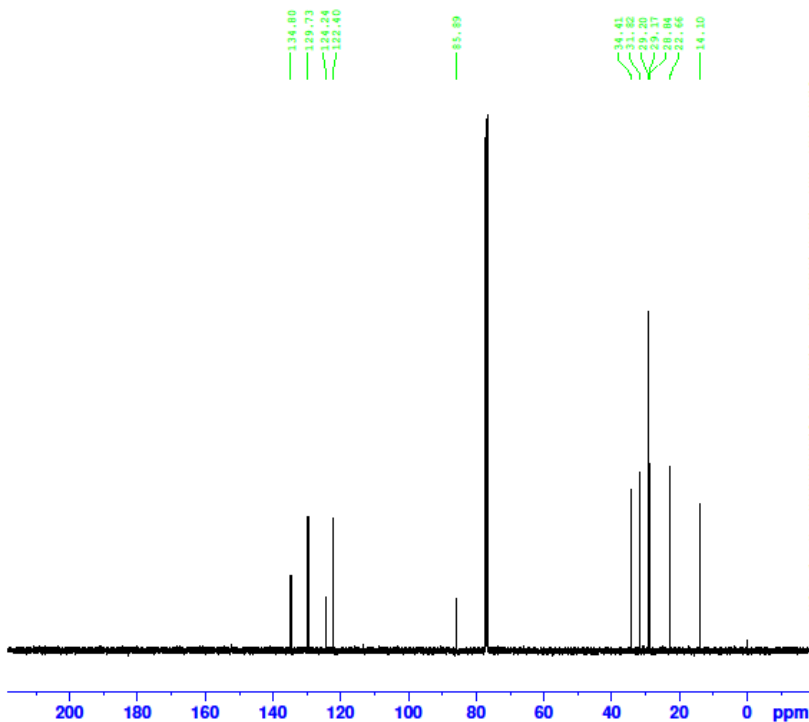
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FIDRES 0.157632 Hz
AQ 3.173425 sec
RG 114
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DE 6.50 umsec
TE 298.2 K
D1 1.0000000 sec
TD0 1

----- CHANNEL f1 -----
NUC1 1H
P1 12.00 umsec
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F2 - Processing parameters
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SF 500.1300139 MHz
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SSB 0
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<sup>13</sup>C NMR spectrum of 2

<sup>13</sup>C



```

Current Data Parameters
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EXPNO 2
PROCNO 1

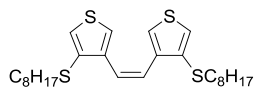
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FIDRES 0.454131 Hz
AQ 1.1010048 sec
RG 203
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DE 6.50 umsec
TE 298.2 K
D1 2.0000000 sec
D11 0.0300000 sec
TD0 1

----- CHANNEL f1 -----
NUC1 13C
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SFO1 125.7783443 MHz

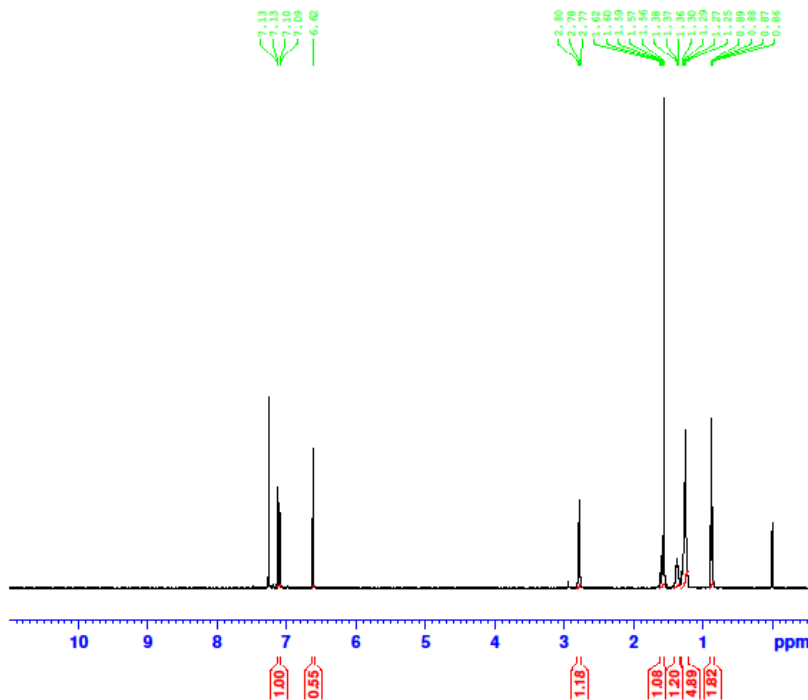
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PL13 17.50 db
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PL1W 0.35600683 W
PL1W 0.35600683 W
SFO2 500.1320005 MHz

F2 - Processing parameters
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SF 125.7577890 MHz
WDW EM
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LB 1.00 Hz
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PC 1.40
    
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<sup>1</sup>H NMR spectrum of 3



<sup>1</sup>H



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EXPNO    1
PROCNO   1

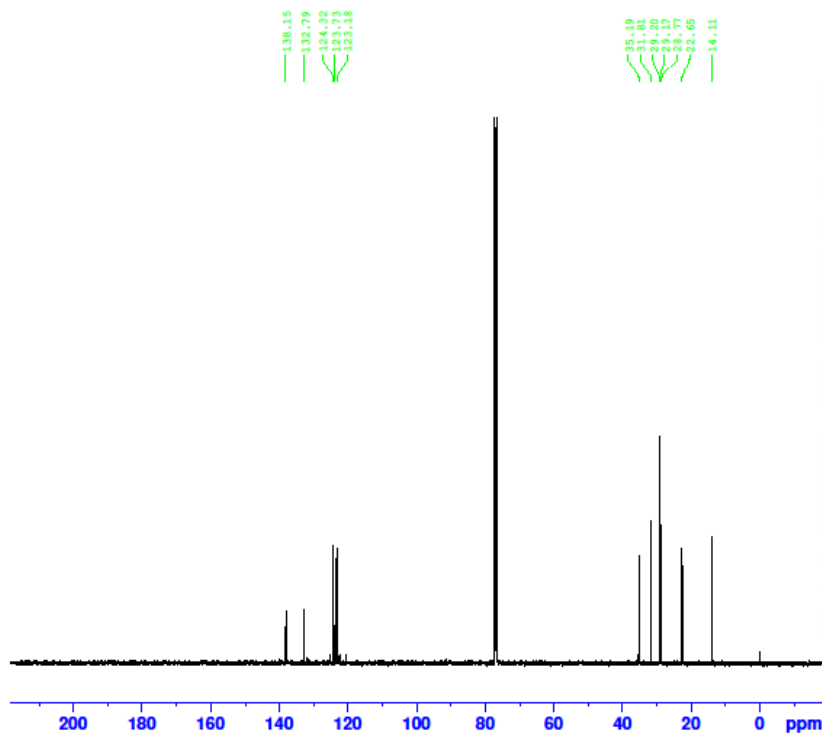
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FIDRES   0.157632 Hz
AQ       3.1719425 sec
RG       181
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DE       6.50 usec
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D1       1.00000000 sec
TD0      1

----- CHANNEL f1 -----
NUC1     1H
P1       12.10 usec
PL1      1.00 dB
PL1W     15.90224075 W
SFO1     500.1330885 MHz

F2 - Processing parameters
SI       32768
SF       500.1330138 MHz
WDW      EM
SSB      0
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PC       1.00
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<sup>13</sup>C NMR spectrum of 3

<sup>13</sup>C



```
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EXPNO    2
PROCNO   1

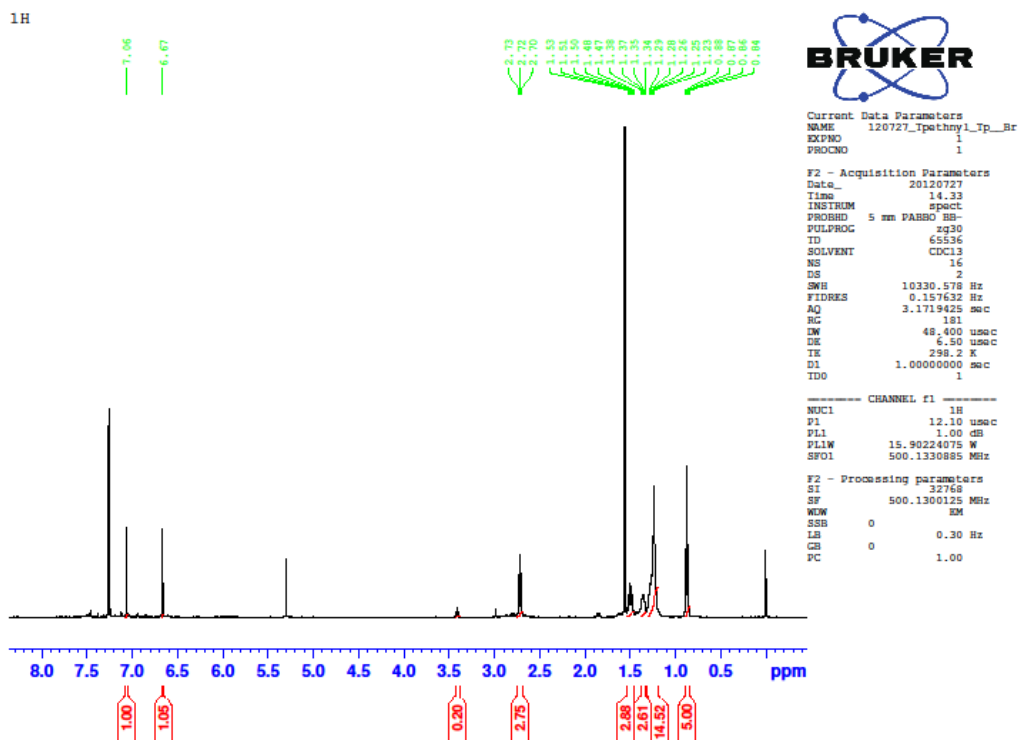
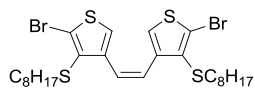
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NS       1024
DS       4
SWH      29761.904 Hz
FIDRES   0.454131 Hz
AQ       1.1010048 sec
RG       323
DM       16.800 usec
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TE       298.2 K
D1       2.00000000 sec
D11      0.03000000 sec
TD0      1

----- CHANNEL f1 -----
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SFO1     125.7703643 MHz

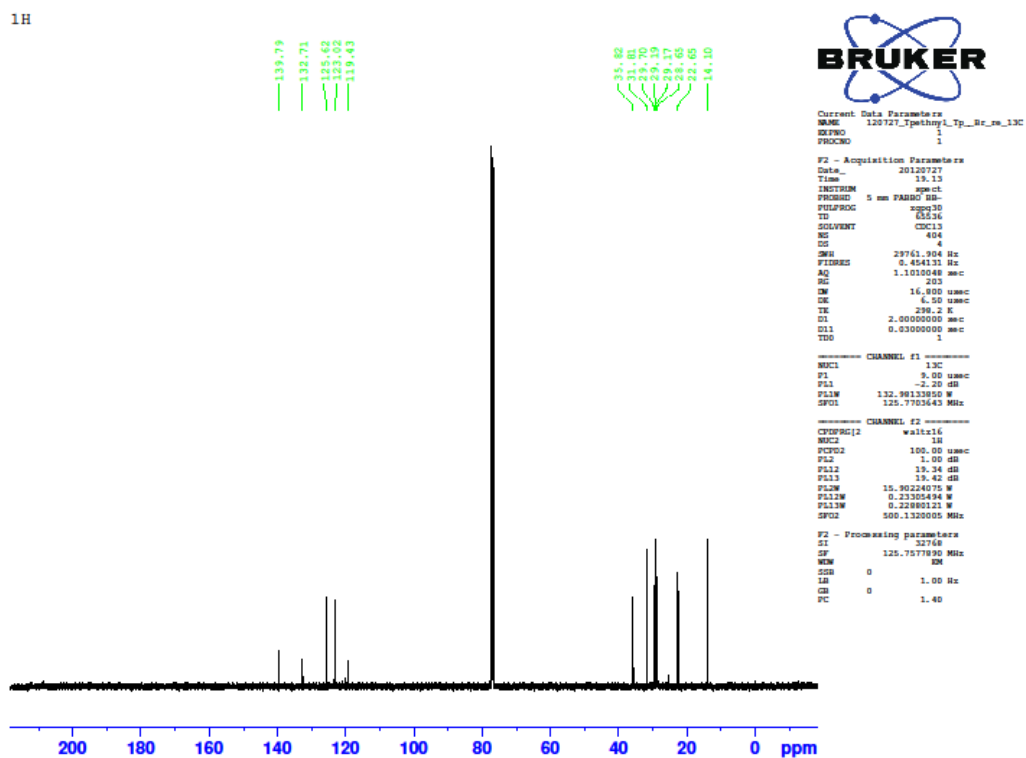
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PL12     17.50 dB
PL13     17.50 dB
PL2W     15.90224075 W
PL12W    0.35000683 W
PL13W    0.35000683 W
SFO2     500.1320005 MHz

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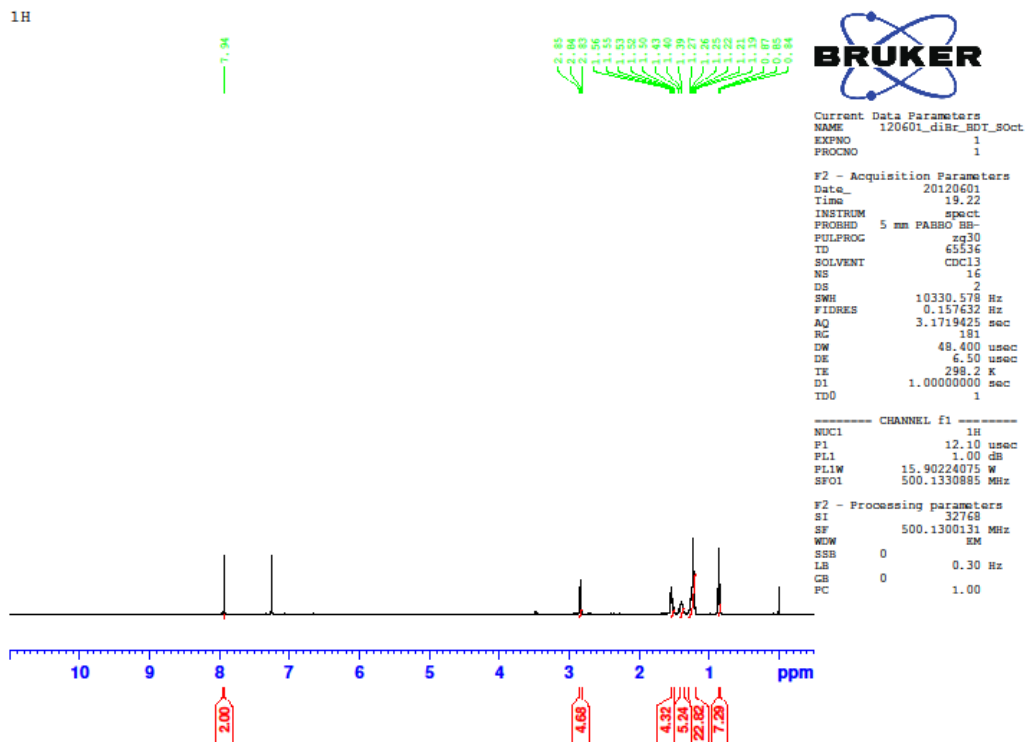
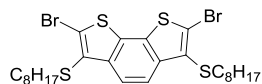
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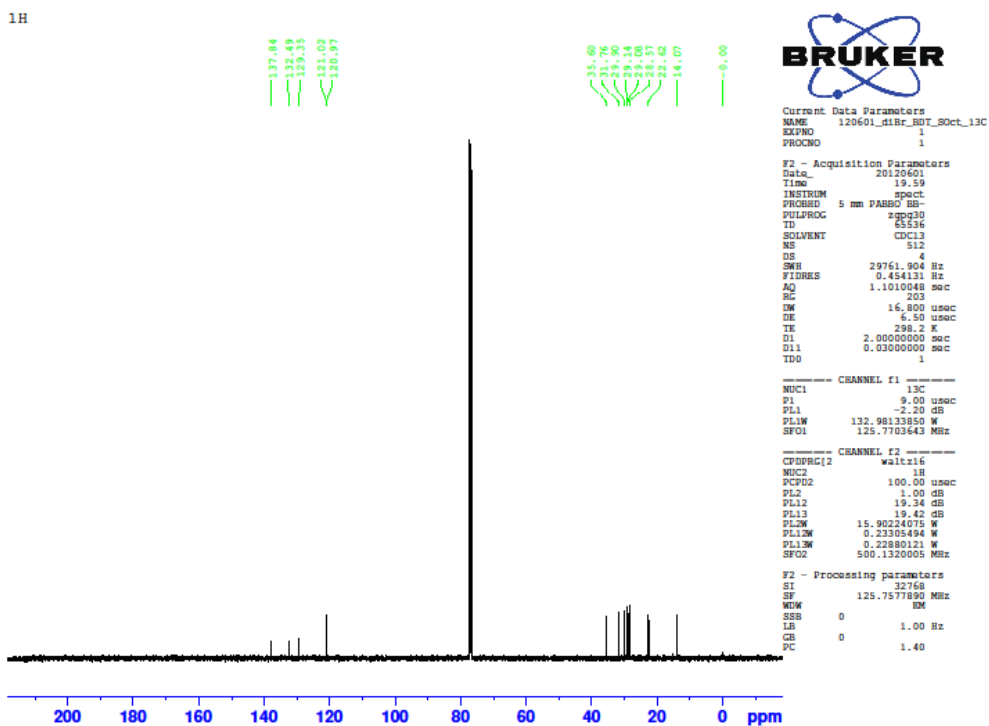
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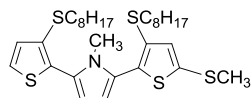
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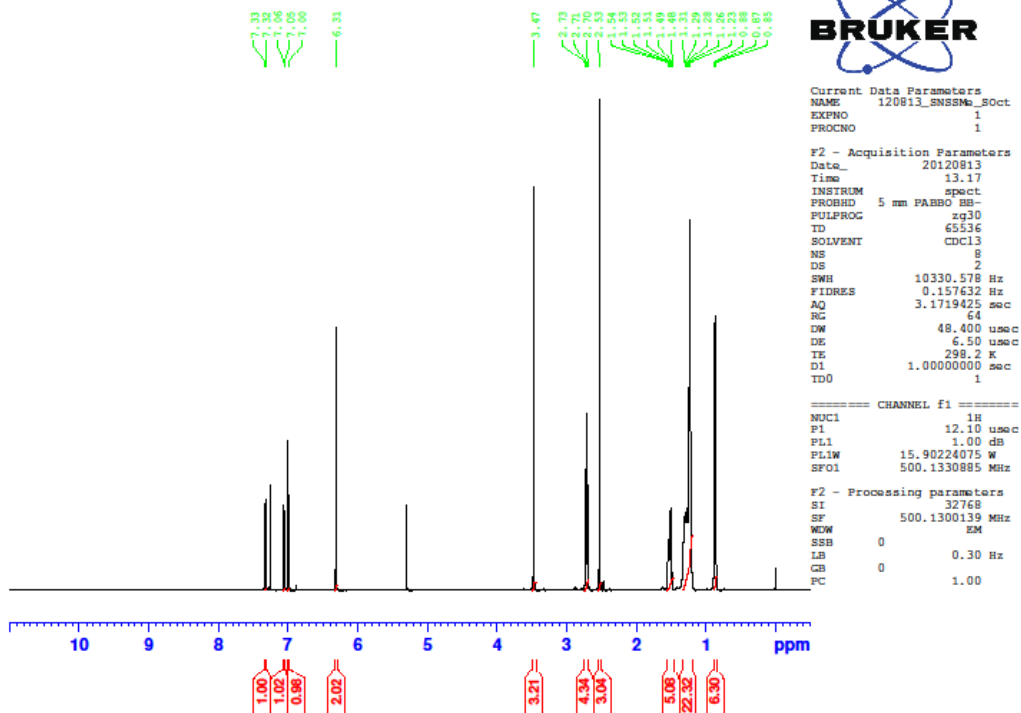
<sup>13</sup>C NMR spectrum of 5



<sup>1</sup>H NMR spectrum of **6**

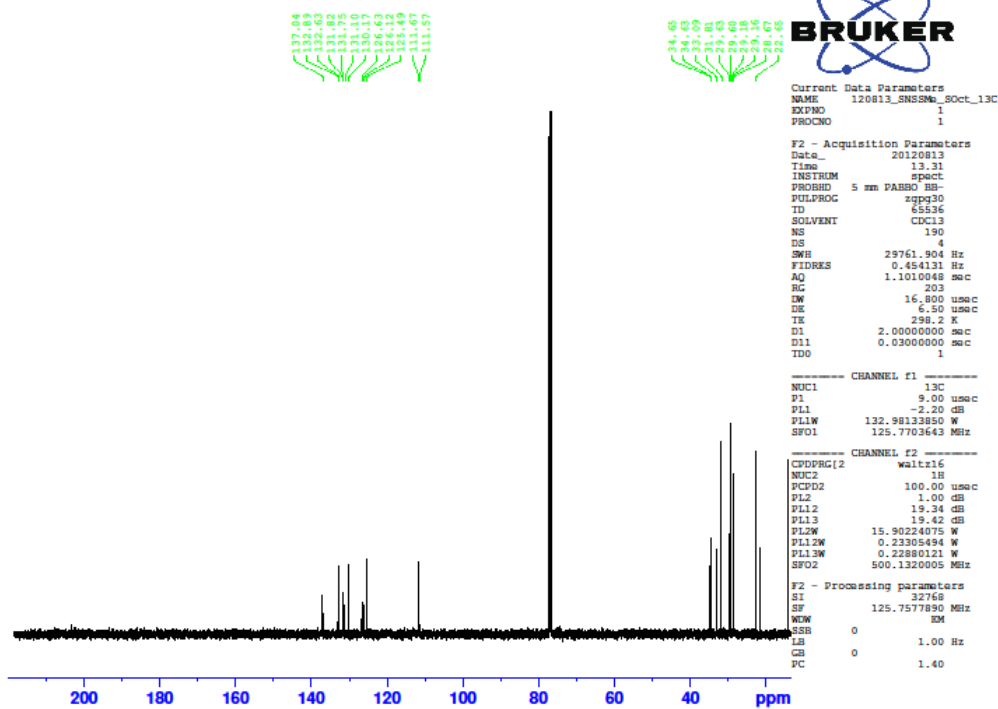


<sup>1</sup>H

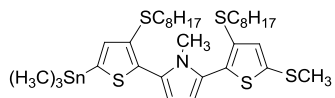


<sup>13</sup>C NMR spectrum of **6**

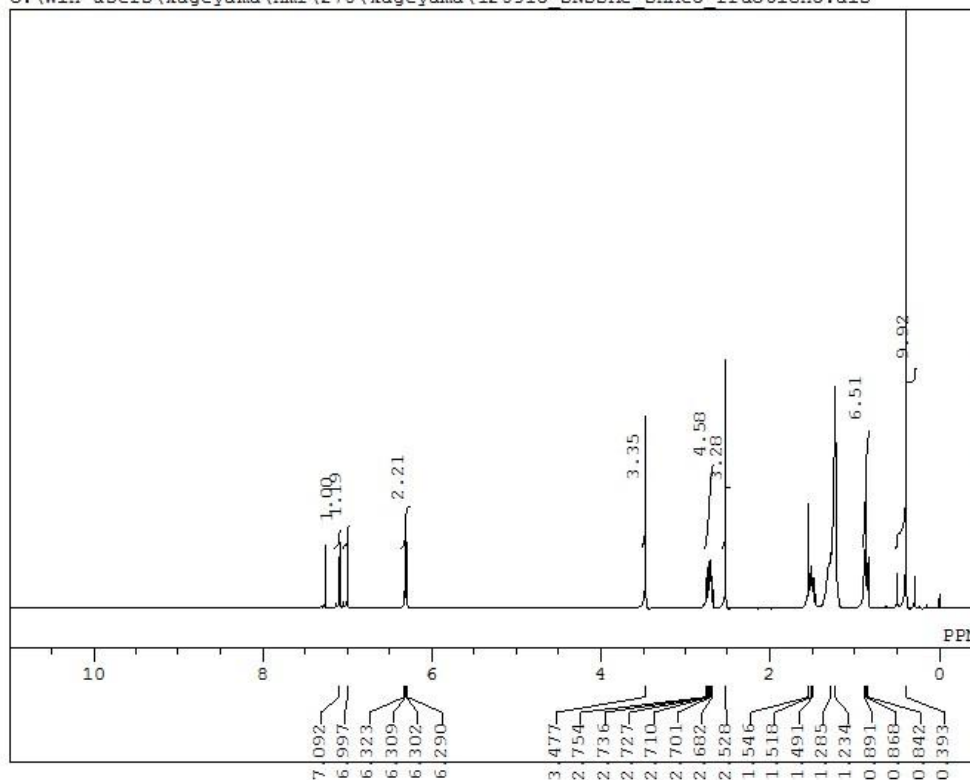
<sup>13</sup>C



<sup>1</sup>H NMR spectrum of 7



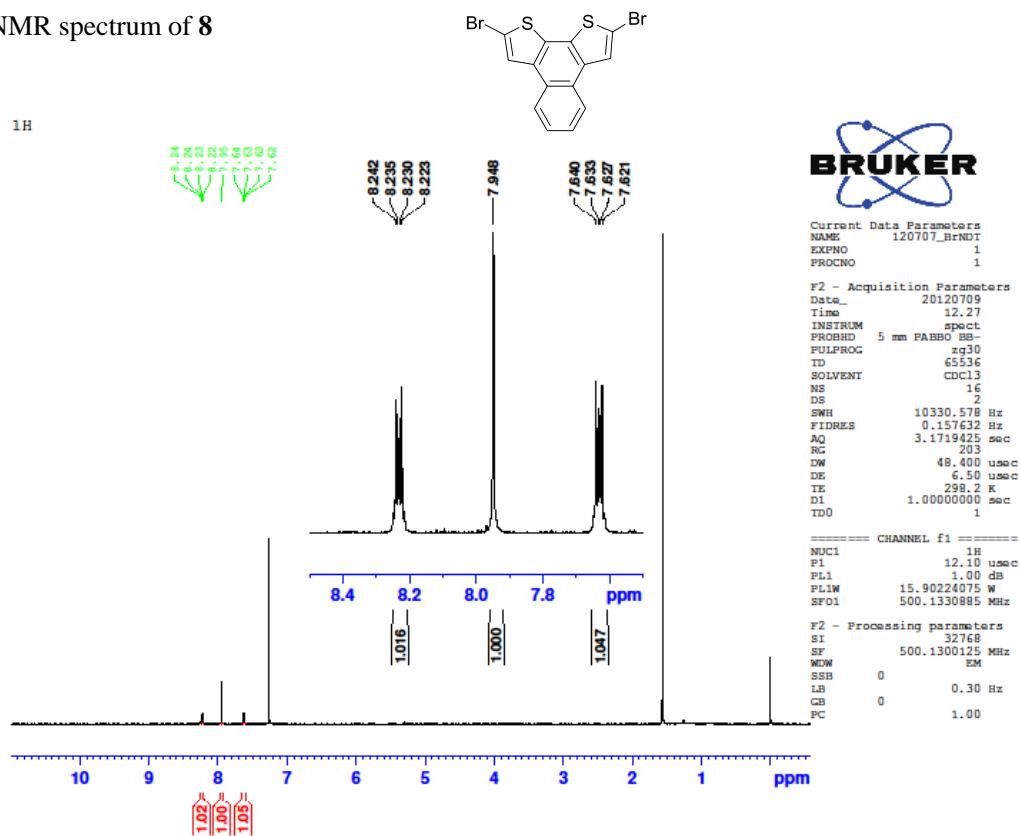
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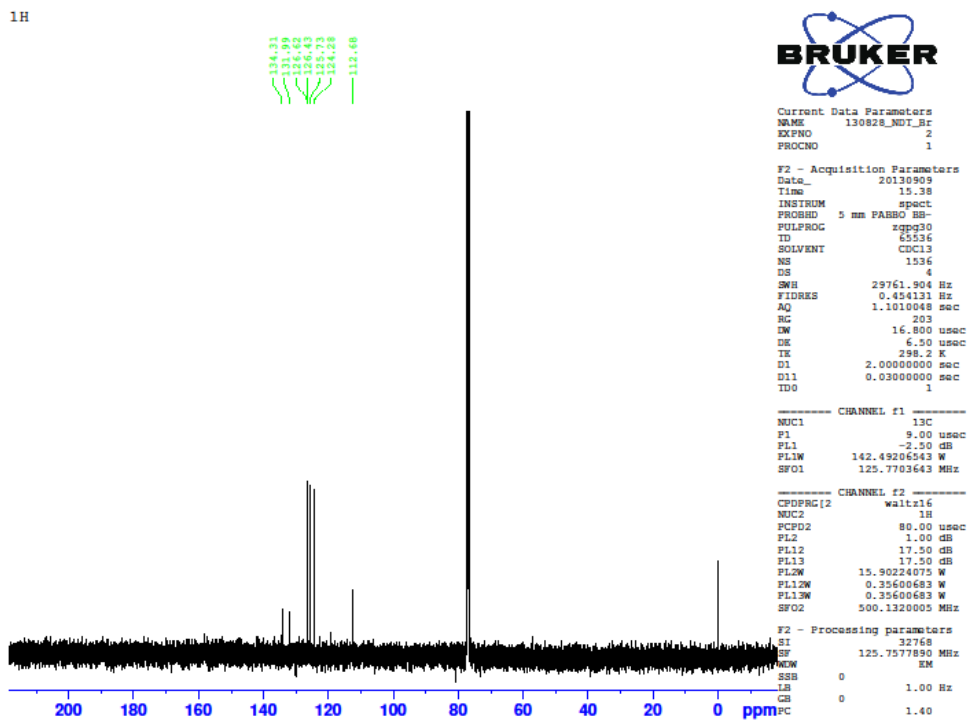
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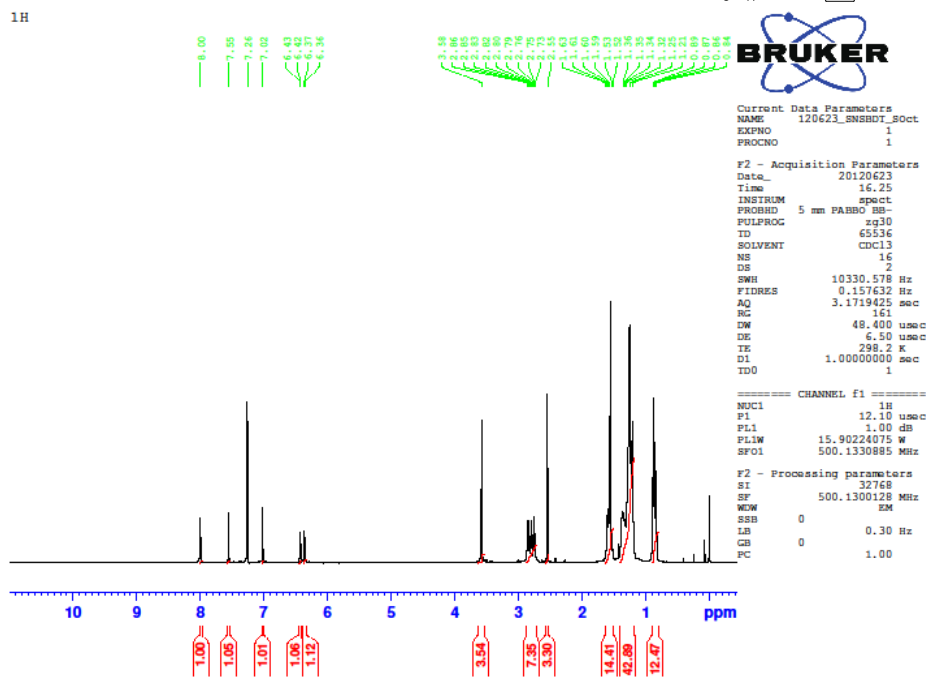
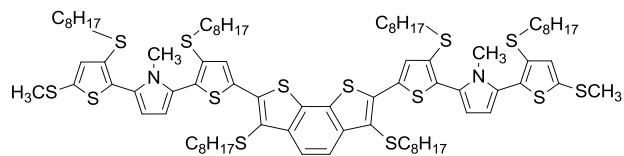
# <sup>1</sup>H NMR spectrum of 8



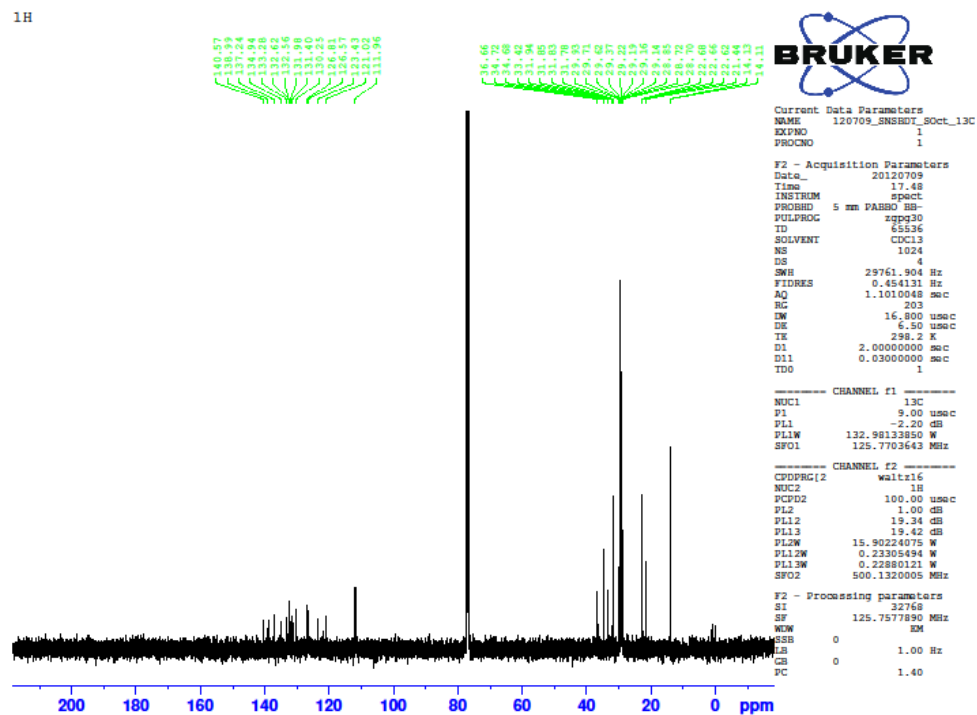
# <sup>13</sup>C NMR spectrum of 8



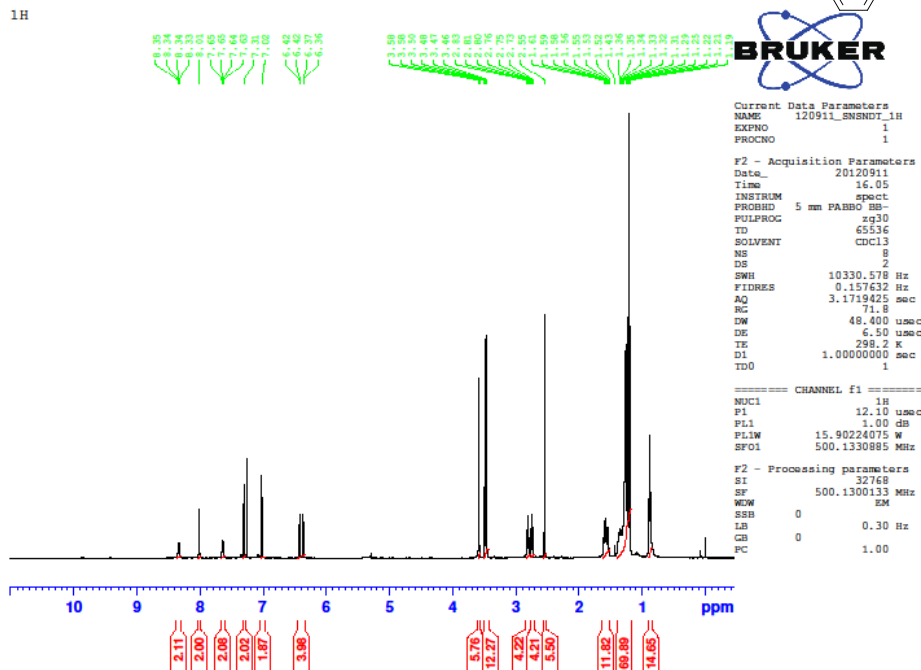
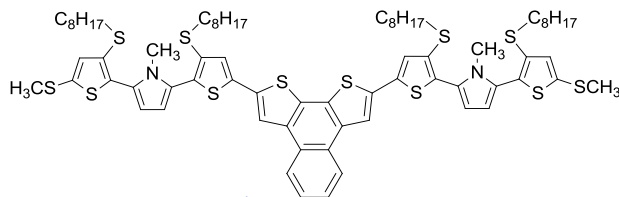
<sup>1</sup>H NMR spectrum of **Bz8TP-C8**



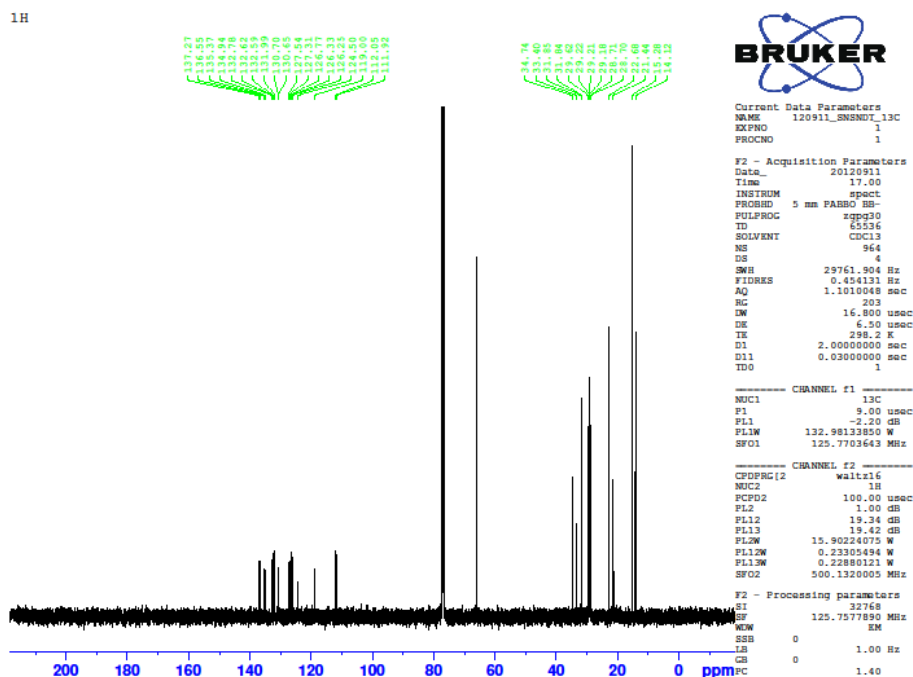
<sup>13</sup>C NMR spectrum of **Bz8TP-C8**



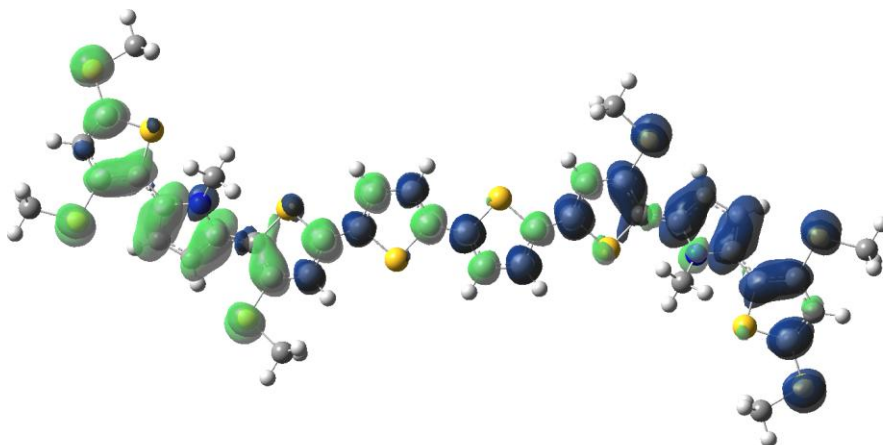
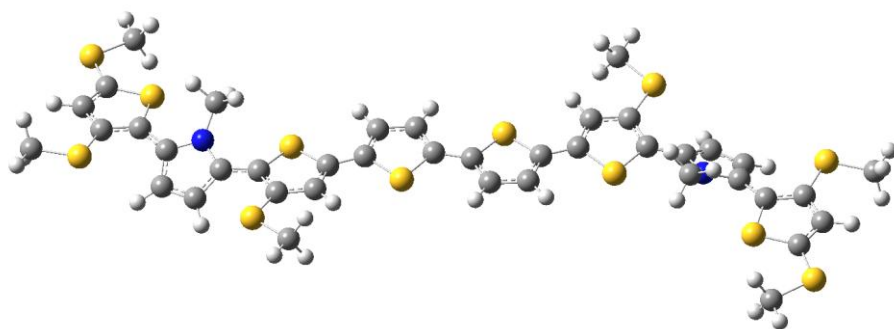
<sup>1</sup>H NMR spectrum of Np8TP-C8



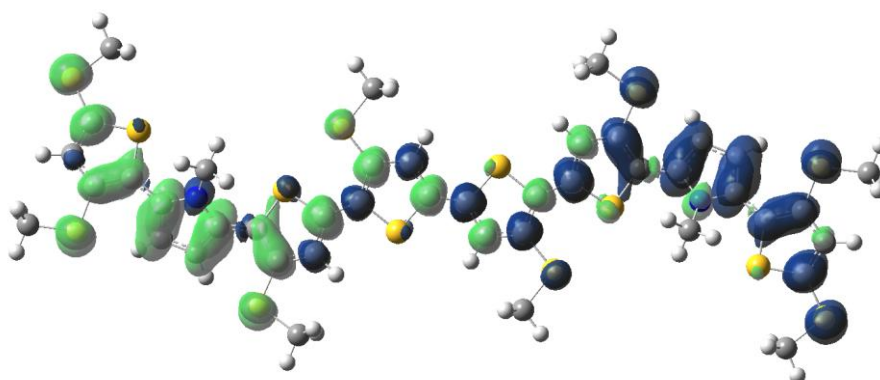
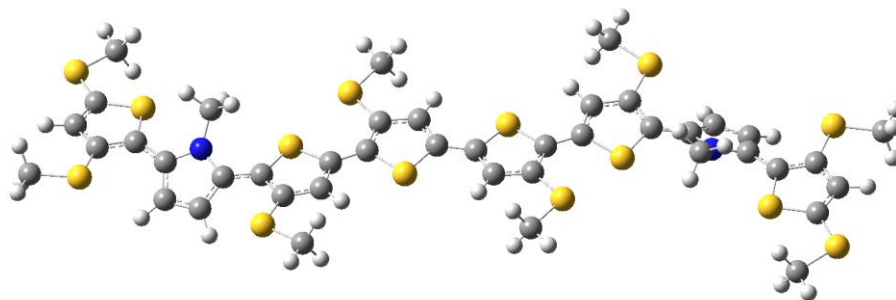
<sup>13</sup>C NMR spectrum of Np8TP-C8



(a)



(b)



**Figure S1.** Optimized structure and spin distribution of (a)  $8\text{TP-C}_1^{2+}$  ( $R^1=\text{SCH}_3, R^2=\text{H}$ ) and (b)  $8\text{TP-C}_1^{2+}$  ( $R^1=\text{SCH}_3, R^2=\text{SCH}_3$ ) at the B3LYP/6-31G(d) level with the broken symmetry method.

**Table S1.** Cartesian coordinate of optimized geometry of singlet **Bz8TP-C<sub>1</sub><sup>2+</sup>** at the UB3LYP/6-31G(d) level with the broken symmetry method.

Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms)		
			X	Y	Z
1	6	0	-0.178159	-0.664840	-2.624507
2	6	0	0.178159	0.664840	-2.624507
3	6	0	-0.366721	-1.355388	-1.399703
4	6	0	-0.182088	-0.674243	-0.174839
5	6	0	-0.742346	-2.729865	-1.204039
6	16	0	-0.446824	-1.724215	1.196791
7	6	0	-0.827180	-3.095545	0.131573
8	6	0	0.366721	1.355388	-1.399703
9	6	0	0.742346	2.729865	-1.204039
10	6	0	0.182088	0.674243	-0.174839
11	6	0	0.827180	3.095545	0.131573
12	16	0	0.446824	1.724215	1.196791
13	6	0	1.168032	4.352165	0.752279
14	6	0	1.294011	4.569904	2.118609
15	16	0	1.473589	5.833430	-0.135136
16	6	0	1.632590	5.889663	2.481471
17	1	0	1.134524	3.782019	2.842906
18	6	0	1.772866	6.733710	1.356882
19	16	0	1.780023	6.477145	4.136992
20	6	0	2.202075	8.093023	1.340993
21	6	0	1.507668	4.974552	5.136562
22	6	0	3.094450	8.724971	2.245828
23	7	0	1.876855	9.024638	0.358376
24	1	0	2.265321	4.214560	4.931785
25	1	0	0.501979	4.573887	4.988053
26	1	0	1.607017	5.303235	6.173725
27	6	0	3.306670	10.015015	1.811609
28	1	0	3.590076	8.232653	3.069034
29	6	0	2.547125	10.215265	0.626621
30	6	0	0.716376	8.912535	-0.533944
31	1	0	4.001927	10.728670	2.227003
32	6	0	2.523205	11.374020	-0.194930
33	1	0	0.997568	8.546617	-1.525300
34	1	0	-0.005756	8.225001	-0.093413
35	1	0	0.244884	9.891207	-0.630125
36	6	0	2.781791	12.718060	0.181796
37	16	0	2.306015	11.276201	-1.951164
38	6	0	2.807703	13.601924	-0.917295
39	16	0	2.951386	13.206613	1.862367
40	6	0	2.573504	12.988298	-2.136724
41	1	0	2.978187	14.667366	-0.836405
42	6	0	3.278383	14.998810	1.744550
43	16	0	2.547125	13.837952	-3.656139
44	1	0	3.408336	15.330202	2.777388
45	1	0	2.429350	15.528940	1.306901
46	1	0	4.196330	15.200952	1.188222
47	6	0	2.185820	12.521627	-4.869707
48	1	0	2.980366	11.771696	-4.886167
49	1	0	2.150403	13.024478	-5.838742
50	1	0	1.215171	12.059700	-4.672558
51	6	0	-1.168032	-4.352165	0.752279
52	6	0	-1.294011	-4.569904	2.118609
53	16	0	-1.473589	-5.833430	-0.135136
54	6	0	-1.632590	-5.889663	2.481471
55	1	0	-1.134524	-3.782019	2.842906
56	6	0	-1.772866	-6.733710	1.356882
57	16	0	-1.780023	-6.477145	4.136992
58	6	0	-2.202075	-8.093023	1.340993
59	6	0	-1.507668	-4.974552	5.136562
60	6	0	-3.094450	-8.724971	2.245828
61	7	0	-1.876855	-9.024638	0.358376

62	1	0	-2.265321	-4.214560	4.931785
63	1	0	-0.501979	-4.573887	4.988053
64	1	0	-1.607017	-5.303235	6.173725
65	6	0	-3.306670	-10.015015	1.811609
66	1	0	-3.590076	-8.232653	3.069034
67	6	0	-2.547125	-10.215265	0.626621
68	6	0	-0.716376	-8.912535	-0.533944
69	1	0	-4.001927	-10.728670	2.227003
70	6	0	-2.523205	-11.374020	-0.194930
71	1	0	-0.997568	-8.546617	-1.525300
72	1	0	0.005756	-8.225001	-0.093413
73	1	0	-0.244884	-9.891207	-0.630125
74	6	0	-2.781791	-12.718060	0.181796
75	16	0	-2.306015	-11.276201	-1.951164
76	6	0	-2.807703	-13.601924	-0.917295
77	16	0	-2.951386	-13.206613	1.862367
78	6	0	-2.573504	-12.988298	-2.136724
79	1	0	-2.978187	-14.667366	-0.836405
80	6	0	-3.278383	-14.998810	1.744550
81	16	0	-2.547125	-13.837952	-3.656139
82	1	0	-3.408336	-15.330202	2.777388
83	1	0	-2.429350	-15.528940	1.306901
84	1	0	-4.196330	-15.200952	1.188222
85	6	0	-2.185820	-12.521627	-4.869707
86	1	0	-2.980366	-11.771696	-4.886167
87	1	0	-2.150403	-13.024478	-5.838742
88	1	0	-1.215171	-12.059700	-4.672558
89	1	0	0.310255	1.202216	-3.557881
90	1	0	-0.310255	-1.202216	-3.557881
91	16	0	1.022620	3.839290	-2.567083
92	16	0	-1.022620	-3.839290	-2.567083
93	6	0	2.760183	3.418128	-3.008838
94	1	0	2.849897	2.357945	-3.254480
95	1	0	3.000207	4.014358	-3.892949
96	1	0	3.442381	3.678916	-2.197158
97	6	0	-2.760183	-3.418128	-3.008838
98	1	0	-2.849897	-2.357945	-3.254480
99	1	0	-3.000207	-4.014358	-3.892949
100	1	0	-3.442381	-3.678916	-2.197158

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HF = -7384.43730628 hartree

**Table S2.** Cartesian coordinate of optimized geometry of singlet **Bz8TP-C<sub>1</sub><sup>2+</sup>** at the RB3LYP/6-31G(d) level.

Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms)		
			X	Y	Z
1	6	0	-0.019285	-0.685834	-2.599768
2	6	0	0.019285	0.685834	-2.599768
3	6	0	-0.041468	-1.405625	-1.370868
4	6	0	-0.018718	-0.694067	-0.141476
5	6	0	-0.087890	-2.817145	-1.181250
6	16	0	-0.034104	-1.777511	1.230891
7	6	0	-0.077760	-3.202040	0.164738
8	6	0	0.041468	1.405625	-1.370868
9	6	0	0.087890	2.817145	-1.181250
10	6	0	0.018718	0.694067	-0.141476
11	6	0	0.077760	3.202040	0.164738
12	16	0	0.034104	1.777511	1.230891
13	6	0	0.098948	4.492872	0.771831
14	6	0	0.105486	4.742045	2.146924
15	16	0	0.118948	6.009412	-0.117581
16	6	0	0.129440	6.094552	2.508482
17	1	0	0.076865	3.938900	2.871490
18	6	0	0.144608	6.955225	1.374699
19	16	0	0.044916	6.705451	4.159708
20	6	0	0.255123	8.368510	1.367632
21	6	0	-0.012196	5.180959	5.160159

22	6	0	0.895920	9.186889	2.335368
23	7	0	-0.160873	9.207014	0.329624
24	1	0	0.894009	4.583568	5.034154
25	1	0	-0.903941	4.590579	4.934989
26	1	0	-0.068116	5.524366	6.195807
27	6	0	0.874384	10.488365	1.884384
28	1	0	1.406175	8.822239	3.214137
29	6	0	0.209909	10.511598	0.629193
30	6	0	-1.187656	8.851913	-0.657749
31	1	0	1.356323	11.338135	2.343539
32	6	0	0.008155	11.637794	-0.217954
33	1	0	-0.748999	8.535117	-1.608060
34	1	0	-1.800986	8.042543	-0.261100
35	1	0	-1.830101	9.716231	-0.828964
36	6	0	-0.087890	13.001362	0.151157
37	16	0	0.009751	11.494150	-1.984505
38	6	0	-0.151600	13.869867	-0.960632
39	16	0	-0.204856	13.516492	1.830127
40	6	0	-0.107867	13.220715	-2.182300
41	1	0	-0.239969	14.945776	-0.886955
42	6	0	-0.280287	15.336116	1.703476
43	16	0	-0.167006	14.043116	-3.717610
44	1	0	-0.328181	15.689021	2.736275
45	1	0	-1.180360	15.661207	1.176506
46	1	0	0.617633	15.740722	1.231247
47	6	0	-0.082904	12.678676	-4.928439
48	1	0	0.860417	12.133638	-4.843621
49	1	0	-0.130318	13.158617	-5.908517
50	1	0	-0.935541	12.003429	-4.820909
51	6	0	-0.098948	-4.492872	0.771831
52	6	0	-0.105486	-4.742045	2.146924
53	16	0	-0.118948	-6.009412	-0.117581
54	6	0	-0.129440	-6.094552	2.508482
55	1	0	-0.076865	-3.938900	2.871490
56	6	0	-0.144608	-6.955225	1.374699
57	16	0	-0.044916	-6.705451	4.159708
58	6	0	-0.255123	-8.368510	1.367632
59	6	0	0.012196	-5.180959	5.160159
60	6	0	-0.895920	-9.186889	2.335368
61	7	0	0.160873	-9.207014	0.329624
62	1	0	-0.894009	-4.583568	5.034154
63	1	0	0.903941	-4.590579	4.934989
64	1	0	0.068116	-5.524366	6.195807
65	6	0	-0.874384	-10.488365	1.884384
66	1	0	-1.406175	-8.822239	3.214137
67	6	0	-0.209909	-10.511598	0.629193
68	6	0	1.187656	-8.851913	-0.657749
69	1	0	-1.356323	-11.338135	2.343539
70	6	0	-0.008155	-11.637794	-0.217954
71	1	0	0.748999	-8.535117	-1.608060
72	1	0	1.800986	-8.042543	-0.261100
73	1	0	1.830101	-9.716231	-0.828964
74	6	0	0.087890	-13.001362	0.151157
75	16	0	-0.009751	-11.494150	-1.984505
76	6	0	0.151600	-13.869867	-0.960632
77	16	0	0.204856	-13.516492	1.830127
78	6	0	0.107867	-13.220715	-2.182300
79	1	0	0.239969	-14.945776	-0.886955
80	6	0	0.280287	-15.336116	1.703476
81	16	0	0.167006	-14.043116	-3.717610
82	1	0	0.328181	-15.689021	2.736275
83	1	0	1.180360	-15.661207	1.176506
84	1	0	-0.617633	-15.740722	1.231247
85	6	0	0.082904	-12.678676	-4.928439
86	1	0	-0.860417	-12.133638	-4.843621
87	1	0	0.130318	-13.158617	-5.908517
88	1	0	0.935541	-12.003429	-4.820909
89	1	0	0.024167	1.240968	-3.531899
90	1	0	-0.024167	-1.240968	-3.531899
91	16	0	0.111975	3.960632	-2.545352

92	16	0	-0.111975	-3.960632	-2.545352
93	6	0	1.910121	3.990217	-2.944144
94	1	0	2.267485	2.988236	-3.190309
95	1	0	2.012925	4.634942	-3.820784
96	1	0	2.486707	4.407268	-2.116020
97	6	0	-1.910121	-3.990217	-2.944144
98	1	0	-2.267485	-2.988236	-3.190309
99	1	0	-2.012925	-4.634942	-3.820784
100	1	0	-2.486707	-4.407268	-2.116020

HF = -7384.42635434 hartree

**Table S3.** Cartesian coordinate of optimized geometry of singlet **Np8TP-C<sub>1</sub><sup>2+</sup>** at the UB3LYP/6-31G(d) level with the broken symmetry method.

Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms)		
			X	Y	Z
1	6	0	-0.001255	0.702904	4.731893
2	6	0	0.001255	-0.702904	4.731893
3	6	0	0.002252	-1.395730	3.535082
4	6	0	0.000808	-0.714332	2.298791
5	6	0	-0.000808	0.714332	2.298791
6	6	0	-0.002252	1.395730	3.535082
7	1	0	-0.002378	1.245593	5.672284
8	1	0	0.002378	-1.245593	5.672284
9	1	0	0.004720	-2.481039	3.549555
10	1	0	-0.004720	2.481039	3.549555
11	6	0	0.001441	-1.418931	1.033902
12	6	0	-0.000350	-0.701724	-0.180649
13	6	0	-0.001441	-2.819815	0.808226
14	16	0	0.001347	-1.757677	-1.571598
15	6	0	-0.006917	-3.181908	-0.526074
16	1	0	-0.013606	-3.556163	1.603622
17	6	0	-0.001441	1.418931	1.033902
18	6	0	0.001441	2.819815	0.808226
19	6	0	0.000350	0.701724	-0.180649
20	6	0	0.006917	3.181908	-0.526074
21	1	0	0.013606	3.556163	1.603622
22	16	0	-0.001347	1.757677	-1.571598
23	6	0	0.014840	4.498756	-1.093415
24	6	0	0.187948	4.856257	-2.422462
25	16	0	-0.212767	5.919692	-0.096942
26	6	0	0.153693	6.245370	-2.664836
27	1	0	0.365964	4.119452	-3.195013
28	6	0	-0.059466	7.000353	-1.485021
29	16	0	0.442050	7.002545	-4.229952
30	6	0	-0.253888	8.406469	-1.376558
31	6	0	0.657659	5.575206	-5.346563
32	6	0	-0.818013	9.272673	-2.348803
33	7	0	0.002252	9.169701	-0.239030
34	1	0	-0.240312	4.953958	-5.380516
35	1	0	1.533761	4.982757	-5.072057
36	1	0	0.822832	6.012611	-6.334011
37	6	0	-0.902021	10.535775	-1.804306
38	1	0	-1.203960	8.958507	-3.306840
39	6	0	-0.390434	10.483048	-0.480214
40	6	0	0.910914	8.766679	0.840216
41	1	0	-1.364924	11.401795	-2.252898
42	6	0	-0.350102	11.539573	0.470433
43	1	0	0.369023	8.368689	1.703007
44	1	0	1.595522	8.005260	0.465468
45	1	0	1.497850	9.630972	1.152875
46	6	0	-0.278087	12.933569	0.221909
47	16	0	-0.559893	11.249939	2.205920
48	6	0	-0.398274	13.708358	1.395348
49	16	0	0.022993	13.586685	-1.383191



50	6	0	-0.559092	12.958995	2.548488
51	1	0	-0.359174	14.789406	1.415976
52	6	0	0.011667	15.391528	-1.107641
53	16	0	-0.730453	13.654584	4.136198
54	1	0	0.183414	15.828583	-2.094015
55	1	0	0.821111	15.695312	-0.439889
56	1	0	-0.955580	15.732098	-0.731573
57	6	0	-0.927666	12.195217	5.216459
58	1	0	-1.829412	11.634152	4.959526
59	1	0	-1.033029	12.596226	6.227052
60	1	0	-0.043308	11.554172	5.178474
61	6	0	-0.014840	-4.498756	-1.093415
62	6	0	-0.187948	-4.856257	-2.422462
63	16	0	0.212767	-5.919692	-0.096942
64	6	0	-0.153693	-6.245370	-2.664836
65	1	0	-0.365964	-4.119452	-3.195013
66	6	0	0.059466	-7.000353	-1.485021
67	16	0	-0.442050	-7.002545	-4.229952
68	6	0	0.253888	-8.406469	-1.376558
69	6	0	-0.657659	-5.575206	-5.346563
70	6	0	0.818013	-9.272673	-2.348803
71	7	0	-0.002252	-9.169701	-0.239030
72	1	0	0.240312	-4.953958	-5.380516
73	1	0	-1.533761	-4.982757	-5.072057
74	1	0	-0.822832	-6.012611	-6.334011
75	6	0	0.902021	-10.535775	-1.804306
76	1	0	1.203960	-8.958507	-3.306840
77	6	0	0.390434	-10.483048	-0.480214
78	6	0	-0.910914	-8.766679	0.840216
79	1	0	1.364924	-11.401795	-2.252898
80	6	0	0.350102	-11.539573	0.470433
81	1	0	-0.369023	-8.368689	1.703007
82	1	0	-1.595522	-8.005260	0.465468
83	1	0	-1.497850	-9.630972	1.152875
84	6	0	0.278087	-12.933569	0.221909
85	16	0	0.559893	-11.249939	2.205920
86	6	0	0.398274	-13.708358	1.395348
87	16	0	-0.022993	-13.586685	-1.383191
88	6	0	0.559092	-12.958995	2.548488
89	1	0	0.359174	-14.789406	1.415976
90	6	0	-0.011667	-15.391528	-1.107641
91	16	0	0.730453	-13.654584	4.136198
92	1	0	-0.183414	-15.828583	-2.094015
93	1	0	-0.821111	-15.695312	-0.439889
94	1	0	0.955580	-15.732098	-0.731573
95	6	0	0.927666	-12.195217	5.216459
96	1	0	1.829412	-11.634152	4.959526
97	1	0	1.033029	-12.596226	6.227052
98	1	0	0.043308	-11.554172	5.178474

HF = -6663.08568693 hartree

**Table S4.** Cartesian coordinate of optimized geometry of singlet **Bz8TP-C<sub>1</sub><sup>2+</sup>** at the RB3LYP/6-31G(d) level.

Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms)		
			X	Y	Z
1	6	0	0.006204	0.701830	4.693858
2	6	0	-0.006204	-0.701830	4.693858
3	6	0	-0.012531	-1.394985	3.494947
4	6	0	-0.006688	-0.713420	2.261171
5	6	0	0.006688	0.713420	2.261171
6	6	0	0.012531	1.394985	3.494947
7	1	0	0.010884	1.245385	5.633612
8	1	0	-0.010884	-1.245385	5.633612
9	1	0	-0.021692	-2.480119	3.509854
10	1	0	0.021692	2.480119	3.509854
11	6	0	-0.013026	-1.421523	0.993841

12	6	0	-0.006884	-0.695095	-0.228640
13	6	0	-0.028253	-2.808243	0.771191
14	16	0	-0.016267	-1.753328	-1.621194
15	6	0	-0.037329	-3.179552	-0.573853
16	1	0	-0.042020	-3.545856	1.565202
17	6	0	0.013026	1.421523	0.993841
18	6	0	0.028253	2.808243	0.771191
19	6	0	0.006884	0.695095	-0.228640
20	6	0	0.037329	3.179552	-0.573853
21	1	0	0.042020	3.545856	1.565202
22	16	0	0.016267	1.753328	-1.621194
23	6	0	0.054464	4.484655	-1.125862
24	6	0	0.163013	4.850134	-2.468811
25	16	0	-0.067297	5.914409	-0.114419
26	6	0	0.164676	6.230038	-2.704945
27	1	0	0.270376	4.110695	-3.252181
28	6	0	0.040247	6.994229	-1.505217
29	16	0	0.399694	6.986822	-4.279907
30	6	0	-0.086900	8.398881	-1.384387
31	6	0	0.539570	5.555759	-5.402422
32	6	0	-0.623819	9.299764	-2.341934
33	7	0	0.195439	9.143168	-0.233562
34	1	0	-0.374903	4.957815	-5.404682
35	1	0	1.409103	4.940519	-5.157811
36	1	0	0.681453	5.988606	-6.395460
37	6	0	-0.671312	10.555363	-1.776674
38	1	0	-1.026547	9.015628	-3.302290
39	6	0	-0.157345	10.466730	-0.456665
40	6	0	1.107570	8.708180	0.830199
41	1	0	-1.107202	11.442458	-2.210872
42	6	0	-0.070425	11.512696	0.507036
43	1	0	0.568568	8.299627	1.689920
44	1	0	1.783171	7.948409	0.435754
45	1	0	1.704856	9.560367	1.155984
46	6	0	0.067297	12.900521	0.275221
47	16	0	-0.309352	11.213504	2.237279
48	6	0	-0.022725	13.668307	1.458030
49	16	0	0.401754	13.559747	-1.323011
50	6	0	-0.228205	12.915370	2.600414
51	1	0	0.067064	14.746042	1.490580
52	6	0	0.475891	15.359328	-1.023948
53	16	0	-0.377960	13.600666	4.196971
54	1	0	0.669839	15.800564	-2.004354
55	1	0	1.297571	15.616317	-0.351467
56	1	0	-0.474525	15.741142	-0.644542
57	6	0	-0.666590	12.140098	5.254213
58	1	0	-1.593128	11.630807	4.978081
59	1	0	-0.764014	12.532286	6.269069
60	1	0	0.182594	11.452877	5.218461
61	6	0	-0.054464	-4.484655	-1.125862
62	6	0	-0.163013	-4.850134	-2.468811
63	16	0	0.067297	-5.914409	-0.114419
64	6	0	-0.164676	-6.230038	-2.704945
65	1	0	-0.270376	-4.110695	-3.252181
66	6	0	-0.040247	-6.994229	-1.505217
67	16	0	-0.399694	-6.986822	-4.279907
68	6	0	0.086900	-8.398881	-1.384387
69	6	0	-0.539570	-5.555759	-5.402422
70	6	0	0.623819	-9.299764	-2.341934
71	7	0	-0.195439	-9.143168	-0.233562
72	1	0	0.374903	-4.957815	-5.404682
73	1	0	-1.409103	-4.940519	-5.157811
74	1	0	-0.681453	-5.988606	-6.395460
75	6	0	0.671312	-10.555363	-1.776674
76	1	0	1.026547	-9.015628	-3.302290
77	6	0	0.157345	-10.466730	-0.456665
78	6	0	-1.107570	-8.708180	0.830199
79	1	0	1.107202	-11.442458	-2.210872
80	6	0	0.070425	-11.512696	0.507036
81	1	0	-0.568568	-8.299627	1.689920

82	1	0	-1.783171	-7.948409	0.435754
83	1	0	-1.704856	-9.560367	1.155984
84	6	0	-0.067297	-12.900521	0.275221
85	16	0	0.309352	-11.213504	2.237279
86	6	0	0.022725	-13.668307	1.458030
87	16	0	-0.401754	-13.559747	-1.323011
88	6	0	0.228205	-12.915370	2.600414
89	1	0	-0.067064	-14.746042	1.490580
90	6	0	-0.475891	-15.359328	-1.023948
91	16	0	0.377960	-13.600666	4.196971
92	1	0	-0.669839	-15.800564	-2.004354
93	1	0	-1.297571	-15.616317	-0.351467
94	1	0	0.474525	-15.741142	-0.644542
95	6	0	0.666590	-12.140098	5.254213
96	1	0	1.593128	-11.630807	4.978081
97	1	0	0.764014	-12.532286	6.269069
98	1	0	-0.182594	-11.452877	5.218461

HF = -6663.07779371 hartree

**Table S5.** Cartesian coordinate of optimized geometry of singlet **8TP-C<sub>1</sub><sup>2+</sup>** (R<sup>1</sup>=SCH<sub>3</sub>,R<sup>2</sup>=H) at the UB3LYP/6-31G(d) level with the broken symmetry method.

Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms)		
			X	Y	Z
1	6	0	1.176485	1.519954	0.243567
2	6	0	0.041867	0.715785	0.210558
3	6	0	-0.462422	3.189944	0.190774
4	6	0	0.896266	2.894561	0.231663
5	16	0	-1.405695	1.707971	0.154947
6	6	0	-1.551766	8.360714	-0.143875
7	6	0	-2.754505	6.142848	0.222763
8	6	0	-2.535088	9.176798	-0.761263
9	6	0	-0.741837	10.473619	-0.267424
10	6	0	0.588384	8.832720	1.119171
11	6	0	0.159556	11.570831	-0.200215
12	6	0	0.994529	13.776050	-0.250299
13	6	0	2.184706	13.073453	-0.336027
14	6	0	-1.096416	4.473207	0.168338
15	6	0	-2.042934	10.461384	-0.838459
16	6	0	-2.448479	4.767963	0.279132
17	6	0	-1.602932	6.952976	0.057099
18	6	0	-0.150096	12.954520	-0.171641
19	1	0	1.494594	8.470034	0.625590
20	1	0	0.214630	8.060869	1.792568
21	1	0	-3.192723	3.996258	0.427514
22	1	0	-3.460039	8.816370	-1.185641
23	1	0	0.831203	9.715633	1.711494
24	1	0	-2.507726	11.300825	-1.332996
25	1	0	0.968389	14.857606	-0.233348
26	7	0	-0.461660	9.176979	0.153589
27	16	0	1.914345	11.351896	-0.315095
28	16	0	-0.159556	5.939969	-0.024529
29	16	0	3.748770	13.832116	-0.443716
30	6	0	4.898789	12.416205	-0.535597
31	1	0	4.719774	11.820467	-1.433964
32	1	0	5.896161	12.857852	-0.593723
33	1	0	4.835233	11.798006	0.363425
34	16	0	-4.364527	6.827863	0.433593
35	6	0	-5.420294	5.351308	0.623441
36	1	0	-5.389207	4.719398	-0.267171
37	1	0	-5.152252	4.782567	1.517082
38	1	0	-6.433138	5.742564	0.744566
39	16	0	-1.793738	13.544988	0.034937
40	6	0	-1.596559	15.359174	-0.025808
41	1	0	-2.608955	15.756354	0.077889

42	1	0	-0.987607	15.720448	0.806093
43	1	0	-1.183960	15.683667	-0.983644
44	1	0	2.182952	1.118146	0.285366
45	1	0	1.665286	3.658764	0.266203
46	6	0	-0.041867	-0.715785	0.210558
47	6	0	-1.176485	-1.519954	0.243567
48	16	0	1.405695	-1.707971	0.154947
49	6	0	-0.896266	-2.894561	0.231663
50	1	0	-2.182952	-1.118146	0.285366
51	6	0	0.462422	-3.189944	0.190774
52	1	0	-1.665286	-3.658764	0.266203
53	6	0	1.096416	-4.473207	0.168338
54	6	0	2.448479	-4.767963	0.279132
55	16	0	0.159556	-5.939969	-0.024529
56	6	0	2.754505	-6.142848	0.222763
57	1	0	3.192723	-3.996258	0.427514
58	6	0	1.602932	-6.952976	0.057099
59	16	0	4.364527	-6.827863	0.433593
60	6	0	1.551766	-8.360714	-0.143875
61	6	0	5.420294	-5.351308	0.623441
62	6	0	2.535088	-9.176798	-0.761263
63	7	0	0.461660	-9.176979	0.153589
64	1	0	5.389207	-4.719398	-0.267171
65	1	0	5.152252	-4.782567	1.517082
66	1	0	6.433138	-5.742564	0.744566
67	6	0	2.042934	-10.461384	-0.838459
68	1	0	3.460039	-8.816370	-1.185641
69	6	0	0.741837	-10.473619	-0.267424
70	6	0	-0.588384	-8.832720	1.119171
71	1	0	2.507726	-11.300825	-1.332996
72	6	0	-0.159556	-11.570831	-0.200215
73	1	0	-1.494594	-8.470034	0.625590
74	1	0	-0.214630	-8.060869	1.792568
75	1	0	-0.831203	-9.715633	1.711494
76	6	0	0.150096	-12.954520	-0.171641
77	16	0	-1.914345	-11.351896	-0.315095
78	6	0	-0.994529	-13.776050	-0.250299
79	16	0	1.793738	-13.544988	0.034937
80	6	0	-2.184706	-13.073453	-0.336027
81	1	0	-0.968389	-14.857606	-0.233348
82	6	0	1.596559	-15.359174	-0.025808
83	16	0	-3.748770	-13.832116	-0.443716
84	1	0	2.608955	-15.756354	0.077889
85	1	0	0.987607	-15.720448	0.806093
86	1	0	1.183960	-15.683667	-0.983644
87	6	0	-4.898789	-12.416205	-0.535597
88	1	0	-4.719774	-11.820467	-1.433964
89	1	0	-5.896161	-12.857852	-0.593723
90	1	0	-4.835233	-11.798006	0.363425

HF=-6433.20019687 hartree

**Table S6.** Cartesian coordinate of optimized geometry of singlet **8TP-C<sub>1</sub><sup>2+</sup>** ( $R^1=\text{SCH}_3, R^2=\text{SCH}_3$ ) at the UB3LYP/6-31G(d) level with the broken symmetry method.

Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms)		
			X	Y	Z
1	6	0	1.248272	1.449640	0.242273
2	6	0	0.076496	0.713786	0.192316
3	6	0	-0.296802	3.218304	0.164534
4	6	0	1.061758	2.851929	0.224343
5	16	0	-1.311203	1.774594	0.111007
6	6	0	-1.574999	8.360374	-0.189383
7	6	0	-2.679021	6.088322	0.161090
8	6	0	-2.587667	9.131608	-0.817424
9	6	0	-0.861314	10.508513	-0.303601

10	6	0	0.526838	8.929935	1.097943
11	6	0	-0.012291	11.647035	-0.223703
12	6	0	0.721281	13.888111	-0.257757
13	6	0	1.942926	13.241855	-0.333595
14	6	0	-0.935975	4.496199	0.139069
15	6	0	-2.153470	10.437321	-0.889497
16	6	0	-2.308242	4.736143	0.230143
17	6	0	-1.564024	6.952604	0.010530
18	6	0	-0.385339	13.013556	-0.194255
19	1	0	1.452019	8.603003	0.614863
20	1	0	0.179032	8.145256	1.770405
21	1	0	-3.019797	3.933944	0.374957
22	1	0	-3.490259	8.729615	-1.252453
23	1	0	0.726499	9.824499	1.688849
24	1	0	-2.650520	11.255312	-1.388576
25	1	0	0.644860	14.967260	-0.237427
26	7	0	-0.526838	9.226460	0.120652
27	16	0	1.751981	11.510048	-0.321897
28	16	0	-0.073325	6.012324	-0.042047
29	16	0	3.473157	14.071749	-0.420532
30	6	0	4.684873	12.708629	-0.515570
31	1	0	4.538442	12.113133	-1.420006
32	1	0	5.662395	13.193820	-0.562586
33	1	0	4.642199	12.080100	0.377542
34	16	0	-4.324258	6.698672	0.342644
35	6	0	-5.307459	5.174121	0.538215
36	1	0	-5.234710	4.534670	-0.344700
37	1	0	-5.020329	4.628854	1.440677
38	1	0	-6.340078	5.514686	0.644633
39	16	0	-2.057627	13.528575	-0.006890
40	6	0	-1.941479	15.349593	-0.066298
41	1	0	-2.972216	15.701044	0.020698
42	1	0	-1.362741	15.739054	0.774431
43	1	0	-1.528703	15.691896	-1.017826
44	1	0	2.221163	0.979057	0.302400
45	6	0	-0.076496	-0.713786	0.192316
46	6	0	-1.248272	-1.449640	0.242273
47	16	0	1.311203	-1.774594	0.111007
48	6	0	-1.061758	-2.851929	0.224343
49	1	0	-2.221163	-0.979057	0.302400
50	6	0	0.296802	-3.218304	0.164534
51	6	0	0.935975	-4.496199	0.139069
52	6	0	2.308242	-4.736143	0.230143
53	16	0	0.073325	-6.012324	-0.042047
54	6	0	2.679021	-6.088322	0.161090
55	1	0	3.019797	-3.933944	0.374957
56	6	0	1.564024	-6.952604	0.010530
57	16	0	4.324258	-6.698672	0.342644
58	6	0	1.574999	-8.360374	-0.189383
59	6	0	5.307459	-5.174121	0.538215
60	6	0	2.587667	-9.131608	-0.817424
61	7	0	0.526838	-9.226460	0.120652
62	1	0	5.234710	-4.534670	-0.344700
63	1	0	5.020329	-4.628854	1.440677
64	1	0	6.340078	-5.514686	0.644633
65	6	0	2.153470	-10.437321	-0.889497
66	1	0	3.490259	-8.729615	-1.252453
67	6	0	0.861314	-10.508513	-0.303601
68	6	0	-0.526838	-8.929935	1.097943
69	1	0	2.650520	-11.255312	-1.388576
70	6	0	0.012291	-11.647035	-0.223703
71	1	0	-1.452019	-8.603003	0.614863
72	1	0	-0.179032	-8.145256	1.770405
73	1	0	-0.726499	-9.824499	1.688849
74	6	0	0.385339	-13.013556	-0.194255
75	16	0	-1.751981	-11.510048	-0.321897
76	6	0	-0.721281	-13.888111	-0.257757
77	16	0	2.057627	-13.528575	-0.006890
78	6	0	-1.942926	-13.241855	-0.333595
79	1	0	-0.644860	-14.967260	-0.237427

80	6	0	1.941479	-15.349593	-0.066298
81	16	0	-3.473157	-14.071749	-0.420532
82	1	0	2.972216	-15.701044	0.020698
83	1	0	1.362741	-15.739054	0.774431
84	1	0	1.528703	-15.691896	-1.017826
85	6	0	-4.684873	-12.708629	-0.515570
86	1	0	-4.538442	-12.113133	-1.420006
87	1	0	-5.662395	-13.193820	-0.562586
88	1	0	-4.642199	-12.080100	0.377542
89	16	0	-2.370957	-4.035280	0.292126
90	16	0	2.370957	4.035280	0.292126
91	6	0	-3.872104	-2.996140	0.314250
92	1	0	-3.912788	-2.371936	1.210027
93	1	0	-3.949062	-2.389315	-0.590927
94	1	0	-4.704801	-3.702935	0.338156
95	6	0	3.872104	2.996140	0.314250
96	1	0	3.912788	2.371936	1.210027
97	1	0	3.949062	2.389315	-0.590927
98	1	0	4.704801	3.702935	0.338156

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HF = -7308.19843859 hartree