Electronic Supplementary Material (ESI) for Dalton Transactions

Design, synthesis, magnetic properties of a π-radical ligand with photo-excited high-spin state and its Fe(II) complex. The first stage of a new strategy for LIESST materials.

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Supplementary Information Data

NMR spectra of (i), (ii), (iii), **L**^{1} and **L**^{2} and the results of the DFT calculations are presented as the supplementary informations.

NMR spectra of (i), (ii), (iii), L^1 and L^2

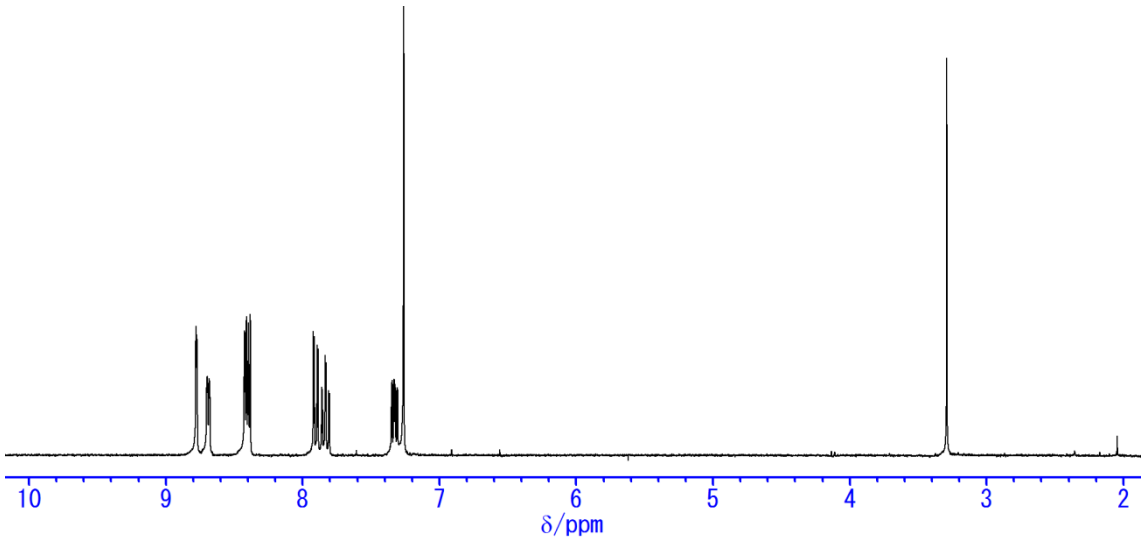


Figure S1. ^{1}H NMR spectrum (300 MHz, $CDCl_3$) of **(i)**.

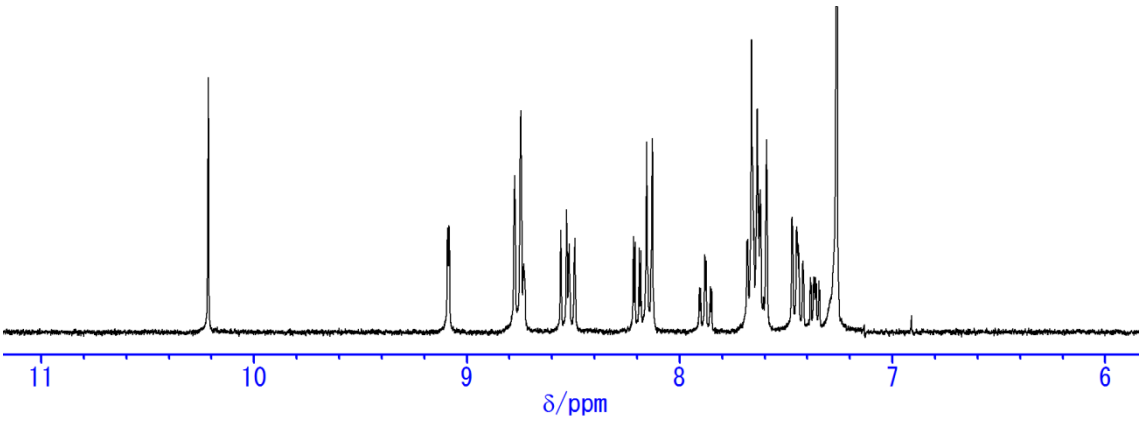


Figure S2. ^{1}H NMR spectrum (300 MHz, $CDCl_3$) of **(ii)**.

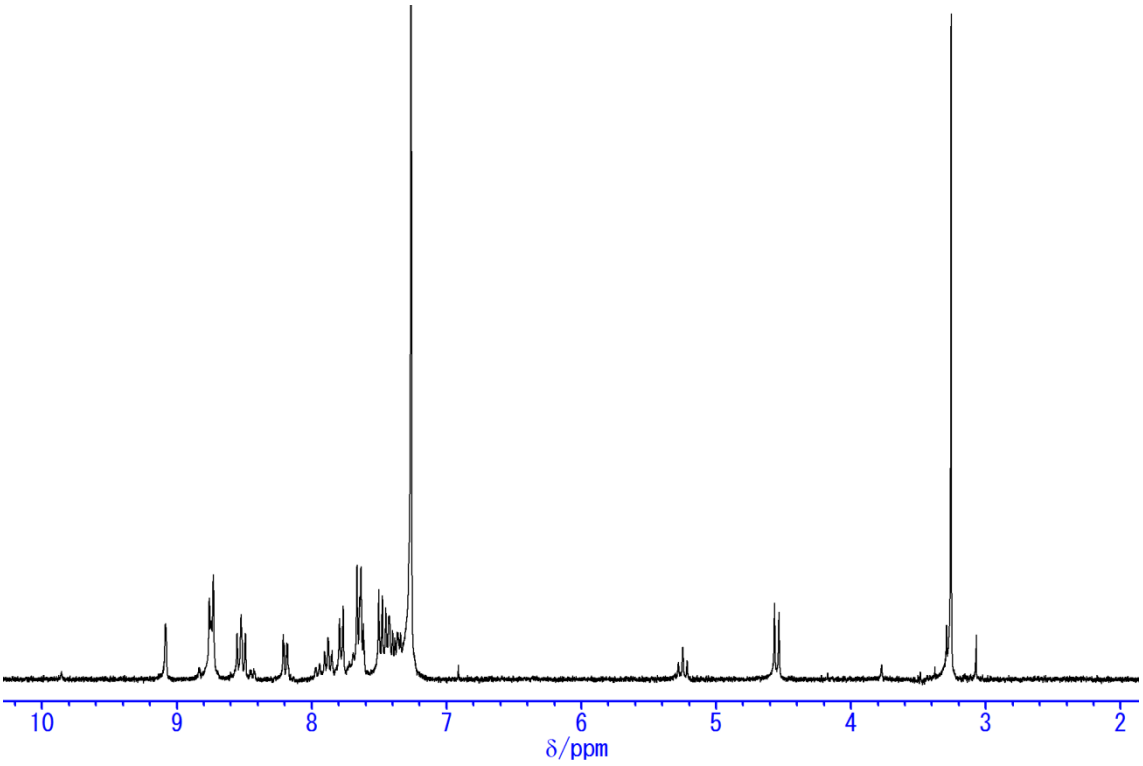


Figure S3. ^{1}H NMR spectrum (300 MHz, $CDCl_3$) of **(iii)**.

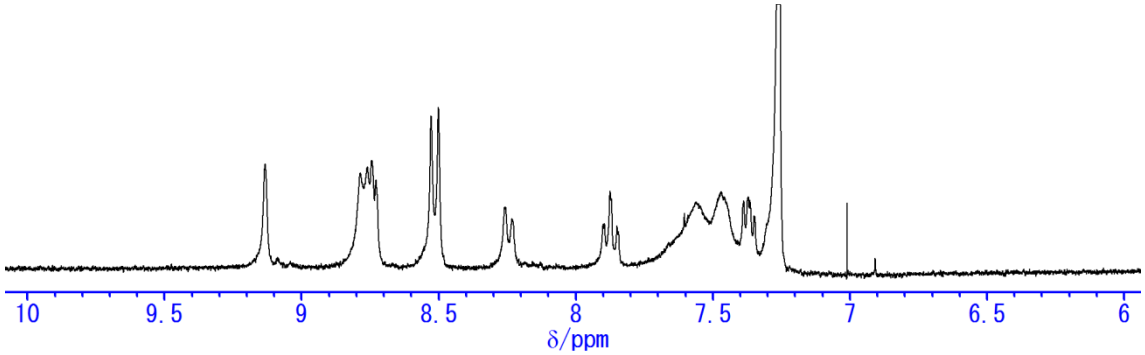


Figure S4. ^{1}H NMR spectrum (300 MHz, $CDCl_3$) of $\mathbf{L^1}$.

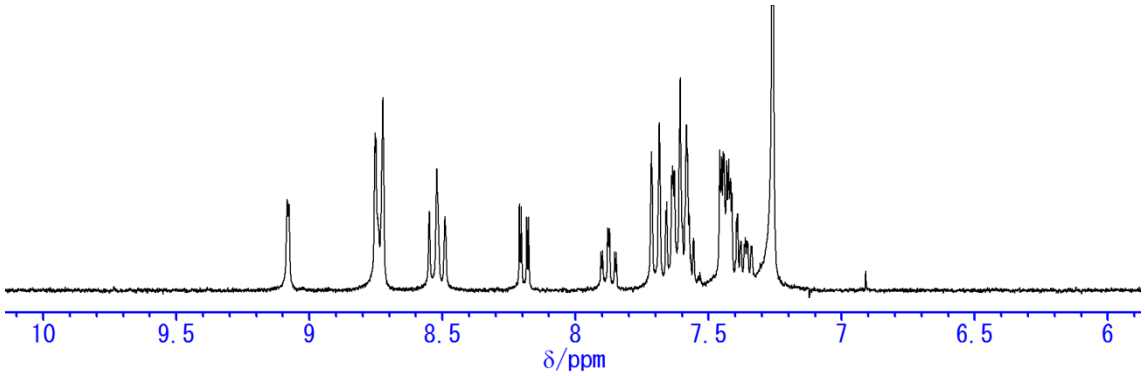


Figure S5. ^{1}H NMR spectrum (300 MHz, $CDCl_3$) of $\mathbf{L^2}$.

DFT calculations of L^1 and complex 1

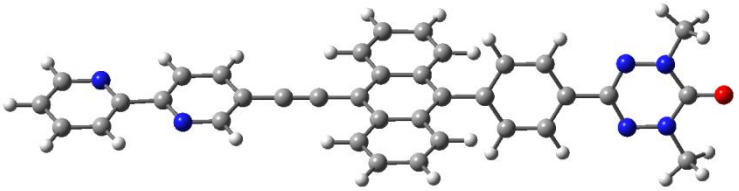


Figure S6. Molecular structure of $\mathbf{L^1}$ (S = 1/2) optimized by the DFT calculation.

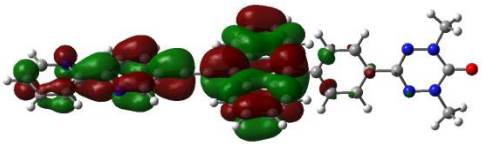
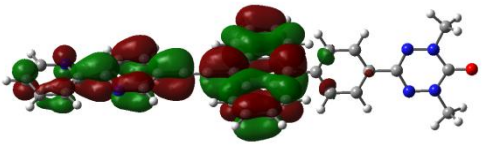
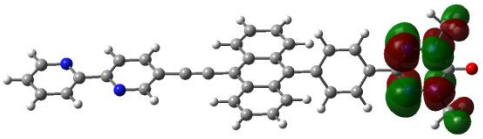
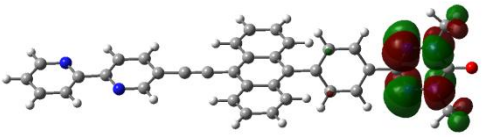
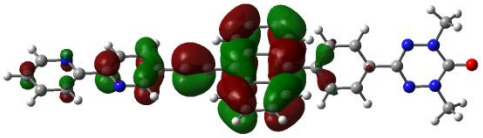
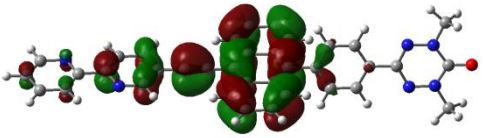
α-LUMO (-0.08222 a.u.) 	β-LUMO+1 (-0.08201 a.u.) 
α-HOMO-1 (SOMO) (-0.19948 a.u.) 	β-LUMO (SOMO) (-0.09671 a.u.) 
α-HOMO (-0.18948 a.u.) 	β-HOMO (-0.18934 a.u.) 

Figure S7. Molecular orbitals of $\mathbf{L^1}$ (S = 1/2) calculated by DFT.

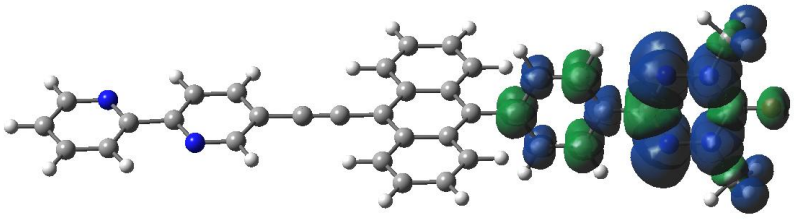


Figure S8. Spin density of $\mathbf{L^1}$ (S = 1/2).

Table S1. Molecular coordinates of $\mathbf{L}^1$ (S = 1/2) optimized by the DFT calculation

	Atomic number	x/Å	y/Å	z/Å
1	6	0.717286	3.700476	-0.416353
2	6	1.397001	2.512830	-0.294116
3	6	0.703181	1.265323	-0.140865
4	6	-0.742249	1.287397	-0.138658
5	6	-1.410335	2.545309	-0.265078
6	6	-0.706308	3.718184	-0.396933
7	6	1.396923	0.035042	-0.012490
8	6	-1.468026	0.067791	-0.019042
9	6	-0.770963	-1.168213	0.102961
10	6	0.674654	-1.179111	0.111972
11	6	1.338955	-2.442227	0.268211
12	1	2.421352	-2.458199	0.295847
13	6	0.631515	-3.613971	0.386621
14	6	-0.792001	-3.599170	0.360222
15	6	-1.468397	-2.410459	0.225646
16	1	1.264352	4.630512	-0.532475
17	1	2.479606	2.503806	-0.316395
18	1	-2.493965	2.552962	-0.257445
19	1	-1.233507	4.661962	-0.492573
20	1	1.156587	-4.556320	0.505051
21	1	-1.341213	-4.530591	0.452704
22	1	-2.551872	-2.393618	0.212745
23	6	-2.888837	0.084477	-0.020057
24	6	-5.531175	0.113810	-0.014880
25	6	-6.272108	1.316893	-0.069270
26	6	-6.270710	-1.091667	0.045615
27	6	-7.661587	1.273087	-0.060585
28	1	-5.749710	2.266001	-0.116928
29	1	-5.752518	-2.044265	0.088408
30	6	-8.310543	0.028409	0.002105
31	1	-8.262565	2.172130	-0.100604
32	6	-4.112059	0.101433	-0.019207
33	6	-9.785098	-0.072682	0.014982
34	6	-10.430355	-1.319481	0.075559
35	6	-11.825494	-1.357008	0.085602

36	1	-9.828361	-2.218050	0.112941
37	6	-11.825474	1.044126	-0.023220
38	6	-12.542846	-0.155563	0.035400
39	1	-12.346308	-2.307868	0.132053
40	1	-12.338160	1.999967	-0.063504
41	1	-13.627085	-0.146034	0.041484
42	6	2.893817	0.018202	-0.007686
43	6	3.617673	0.443741	1.122208
44	6	3.615394	-0.423860	-1.132708
45	6	5.012669	0.427715	1.129997
46	1	3.079260	0.784650	2.000658
47	6	5.010387	-0.440331	-1.130783
48	1	3.075256	-0.752299	-2.014843
49	6	5.726178	-0.014743	0.002137
50	1	5.559011	0.753866	2.006301
51	1	5.555114	-0.779315	-2.003211
52	6	7.204817	-0.033087	0.007641
53	6	9.887780	-0.959152	-2.275393
54	1	10.608351	-0.214037	-2.620540
55	1	10.424229	-1.888799	-2.063891
56	1	9.121793	-1.129357	-3.029401
57	6	9.893549	0.818039	2.313231
58	1	10.553580	0.033674	2.691974
59	1	10.496907	1.703916	2.095953
60	1	9.123435	1.054976	3.044645
61	7	7.817817	-0.474131	-1.112795
62	7	-7.608820	-1.134421	0.053760
63	7	-10.479456	1.093752	-0.033608
64	6	9.963400	-0.070941	0.019156
65	8	11.210327	-0.090521	0.025595
66	7	7.820274	0.392612	1.132691
67	7	9.204845	0.359321	1.104747
68	7	9.202504	-0.477018	-1.074128

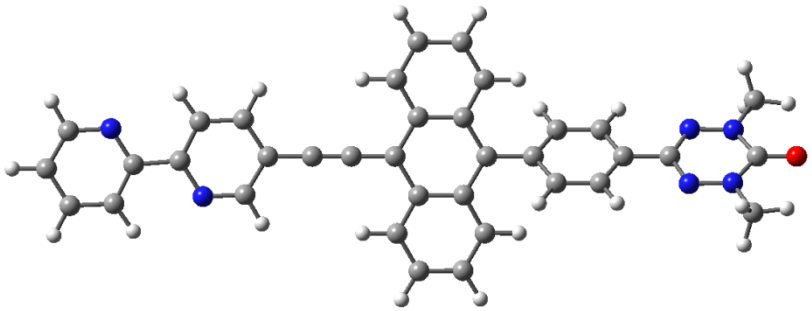
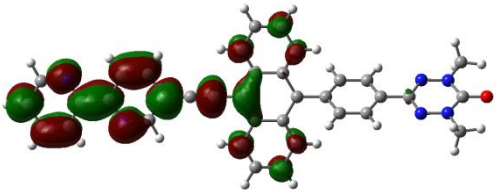
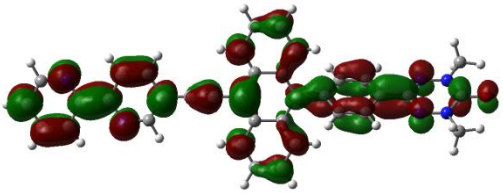
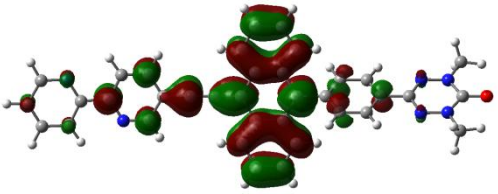
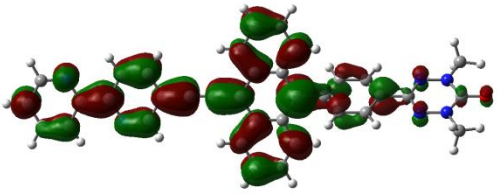
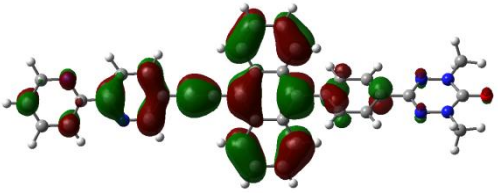
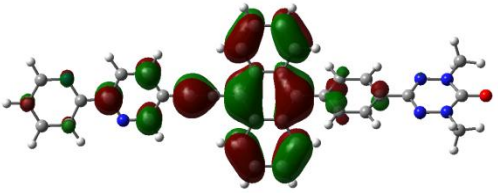
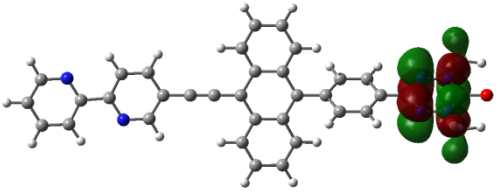
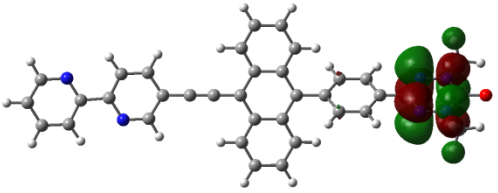


Figure S9. Molecular structure of $\mathbf{L}^{1}$ (S = 3/2) optimized by the DFT calculation.

α-LUMO (-0.05996 a.u.) 	β-LUMO+3 (-0.04999 a.u.) 
α-HOMO (SOMO) (-0.13743 a.u.) 	β-LUMO+2 (SOMO) (-0.07644 a.u.) 
α-HOMO-1 (SOMO) (-0.19944 a.u.) 	β-LUMO (SOMO) (-0.13552 a.u.) 
α-HOMO-3 (SOMO) (-0.20171 a.u.) 	β-LUMO+1 (SOMO) (-0.09959 a.u.) 

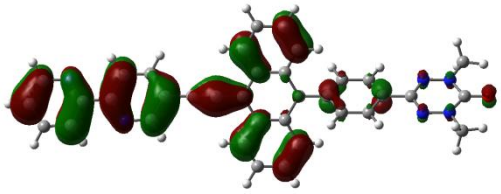
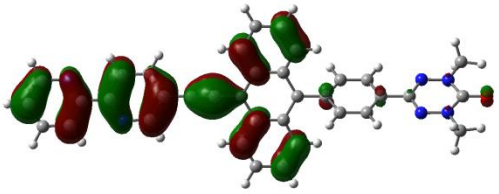
α-HOMO-4 (-0.23662 a.u.)	β-HOMO (-0.22953 a.u.)
	

Figure S10. Molecular orbitals of $\mathbf{L}^{1}$ (S = 3/2) calculated by DFT.

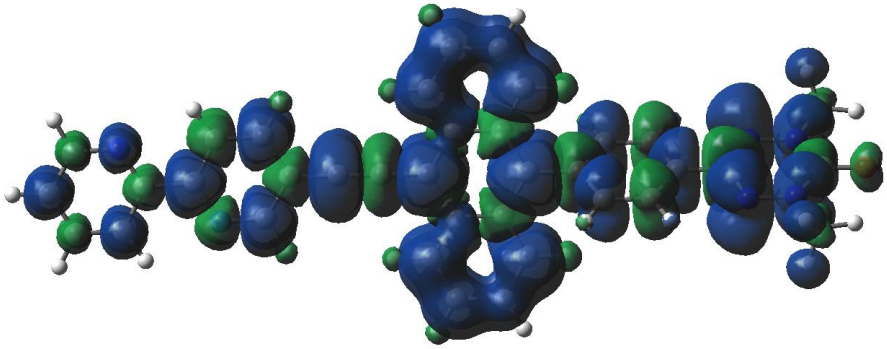


Figure S11. Spin density of $\mathbf{L}^{1}$ (S = 3/2).

Table S2. Molecular coordinates of $\mathbf{L}^{1}$ (S = 3/2) optimized by the DFT calculation

	Atomic number	x/Å	y/Å	z/Å
1	6	-0.693259	3.718828	-0.603551
2	6	-1.378150	2.506570	-0.438225
3	6	-0.697844	1.293081	-0.192355
4	6	0.737517	1.323295	-0.175261
5	6	1.401328	2.547187	-0.341345
6	6	0.698359	3.744911	-0.545050
7	6	-1.408363	0.037243	-0.011688
8	6	1.490487	0.075951	-0.015370
9	6	0.771528	-1.191263	0.144891
10	6	-0.664144	-1.199403	0.165748
11	6	-1.311117	-2.430863	0.412083
12	1	-2.390329	-2.454629	0.488365
13	6	-0.593591	-3.624574	0.574243
14	6	0.798053	-3.613404	0.512110
15	6	1.468267	-2.397120	0.307991
16	1	-1.254493	4.629956	-0.783545
17	1	-2.457844	2.501459	-0.511425
18	1	2.485159	2.558111	-0.319245
19	1	1.241286	4.676001	-0.668286
20	1	-1.129807	-4.550552	0.754716
21	1	1.366116	-4.529672	0.632862
22	1	2.551936	-2.379465	0.283208
23	6	2.875248	0.094693	-0.015689
24	6	5.516749	0.122649	-0.014212
25	6	6.267900	1.322251	-0.139662
26	6	6.259966	-1.084062	0.114629
27	6	7.654008	1.271928	-0.131155
28	1	5.748675	2.268907	-0.240671
29	1	5.737908	-2.030631	0.213425
30	6	8.302026	0.028890	0.001239
31	1	8.259013	2.164452	-0.223653
32	6	4.111755	0.111290	-0.015531
33	6	9.772541	-0.076269	0.015546
34	6	10.415215	-1.320661	0.142685
35	6	11.809824	-1.362249	0.149911

36	1	9.810765	-2.213921	0.231639
37	6	11.817288	1.029532	-0.091200
38	6	12.531266	-0.167719	0.031113
39	1	12.327697	-2.310944	0.246727
40	1	12.333416	1.979758	-0.185730
41	1	13.615528	-0.161591	0.032992
42	6	-2.889544	0.018089	-0.007791
43	6	-3.615691	-0.760570	-0.937676
44	6	-3.629835	0.777935	0.926550
45	6	-5.008416	-0.776948	-0.937559
46	1	-3.075227	-1.340684	-1.678177
47	6	-5.022432	0.757890	0.935451
48	1	-3.099899	1.372208	1.663418
49	6	-5.732507	-0.018995	0.001326
50	1	-5.548407	-1.369159	-1.666095
51	1	-5.573069	1.335718	1.667547
52	6	-7.207242	-0.038625	0.006603
53	6	-9.875533	1.560490	1.877052
54	1	-10.951126	1.435356	1.768855
55	1	-9.558044	1.269974	2.881791
56	1	-9.594974	2.602068	1.699799
57	6	-9.845899	-1.710459	-1.842130
58	1	-10.923597	-1.604902	-1.734024
59	1	-9.533868	-1.420227	-2.848552
60	1	-9.546302	-2.745741	-1.658664
61	7	-7.832005	0.746315	0.912088
62	7	7.594007	-1.128989	0.121618
63	7	10.471894	1.083974	-0.099968
64	6	-9.973380	-0.076382	0.017784
65	8	-11.221179	-0.093919	0.023225
66	7	-7.817580	-0.840069	-0.894211
67	7	-9.200138	-0.834543	-0.862685
68	7	-9.214085	0.703408	0.891427

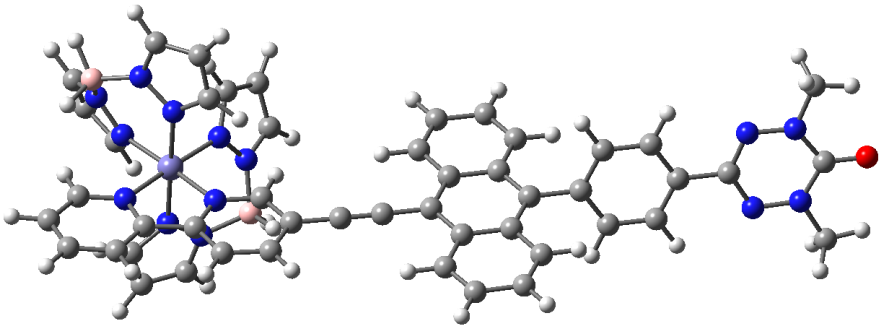
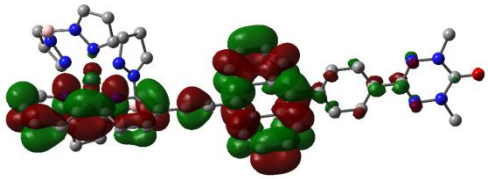
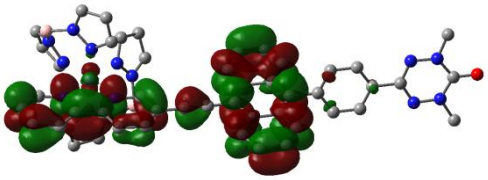
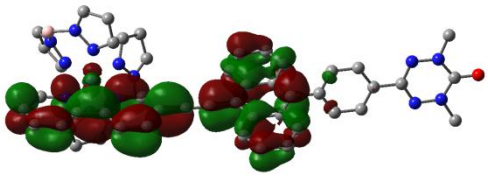
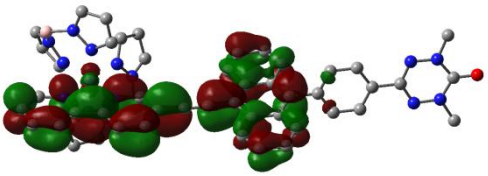
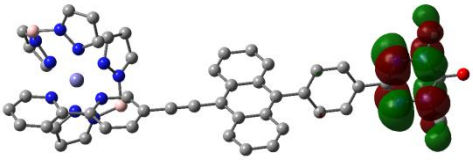
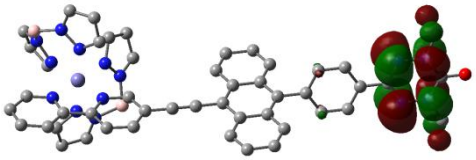
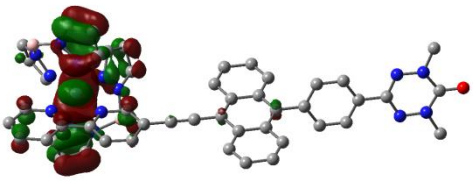
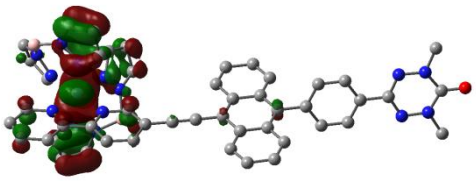


Figure S12. Molecular structure of **1** (S = 1/2) optimized by the DFT calculation.

α-LUMO+1 (-0.07721 a.u.) 	β-LUMO+2 (-0.10159 a.u.) 
α-LUMO (-0.10171 a.u.) 	β-LUMO+1 (-0.10961 a.u.) 
α-HOMO-4 (SOMO) (-0.21054 a.u.) 	β-LUMO (SOMO) (-0.19432 a.u.) 
α-HOMO (-0.19432 a.u.) 	β-HOMO (-0.19520 a.u.) 

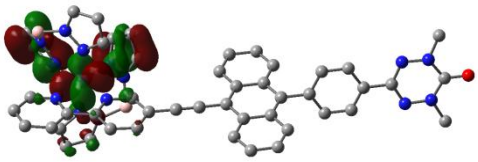
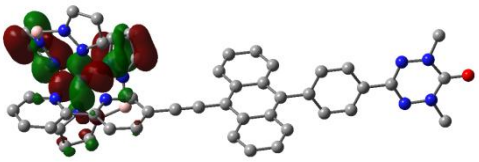
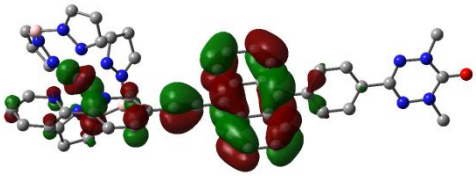
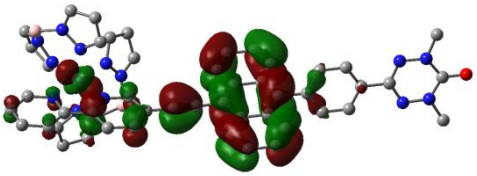
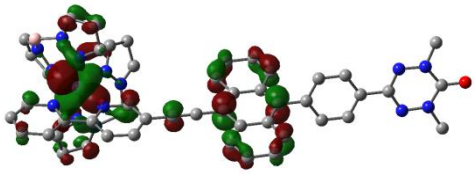
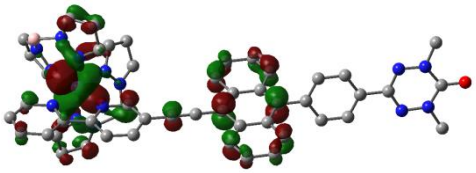
α-HOMO-1 (-0.19520 a.u.) 	β-HOMO-1 (-0.20061 a.u.) 
α-HOMO-2 (-0.20075 a.u.) 	β-HOMO-2 (-0.20385 a.u.) 
α-HOMO-3 (-0.20387 a.u.) 	β-HOMO-3 (-0.22208 a.u.) 

Figure S13. Molecular orbitals of **1** (S = 1/2) calculated by DFT.

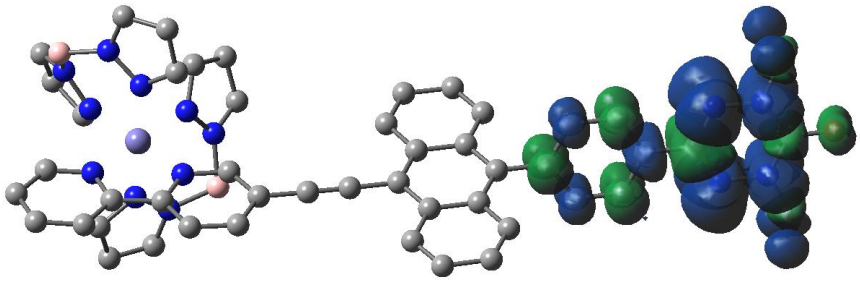


Figure S14. Spin density of **1** (S = 1/2).

Table S3. Molecular coordinates of **1** (S = 1/2) optimized by the DFT calculation

	Atomic number	x/Å	y/Å	z/Å
1	26	-5.581488	-0.300887	0.042355
2	5	-3.373503	-1.037988	2.618752
3	7	-4.206876	1.137669	-0.103404
4	7	-5.699095	-0.019404	2.060951
5	7	-4.255113	-1.813328	0.302892
6	7	-4.774974	-0.472914	2.990129
7	7	-3.519184	-2.059353	1.455799
8	6	-2.868966	0.961631	-0.072443
9	6	-1.947688	2.034577	-0.177945
10	6	-2.479957	3.341998	-0.319834
11	6	-3.865126	3.518115	-0.368068
12	6	-4.716628	2.400912	-0.265791
13	6	-6.731040	0.512905	2.773940
14	6	-6.482150	0.413601	4.160104
15	6	-5.233867	-0.220068	4.251470
16	6	-4.096865	-2.900114	-0.500756
17	6	-3.249414	-3.848643	0.114164
18	6	-2.910814	-3.279059	1.351153
19	5	-7.863897	-1.520671	-2.281599
20	7	-6.809017	1.249161	-0.205221
21	7	-5.457556	-0.580320	-1.976659
22	7	-7.048191	-1.694766	0.178664
23	7	-6.420532	-1.194570	-2.763188
24	7	-7.806197	-2.185205	-0.878132
25	6	-8.165107	1.202889	-0.225984
26	6	-8.953598	2.350817	-0.394202
27	6	-8.319245	3.597755	-0.543821
28	6	-6.918184	3.652524	-0.507684
29	6	-6.184126	2.463167	-0.332179
30	6	-4.391182	-0.343205	-2.791781
31	6	-4.656184	-0.793946	-4.103061
32	6	-5.950971	-1.329266	-4.038792
33	6	-7.307549	-2.495877	1.248353
34	6	-8.241867	-3.496464	0.900706
35	6	-8.526765	-3.266460	-0.453443

36	1	-8.609158	0.226438	-0.100049
37	1	-10.034458	2.258490	-0.412281
38	1	-8.901227	4.504623	-0.680849
39	1	-6.407199	4.603118	-0.612230
40	1	-4.279333	4.512585	-0.488061
41	1	-1.810302	4.192716	-0.397715
42	1	-2.520477	-0.055272	0.032099
43	1	-4.603951	-2.955887	-1.450062
44	1	-2.945622	-4.808313	-0.274718
45	1	-2.293174	-3.653120	2.153369
46	1	-4.647655	-0.499452	5.113335
47	1	-7.115909	0.742185	4.969190
48	1	-7.599780	0.913046	2.279243
49	1	-3.489479	0.107788	-2.413619
50	1	-4.003006	-0.747002	-4.960601
51	1	-6.562827	-1.785064	-4.801906
52	1	-9.180653	-3.790489	-1.133638
53	1	-8.637109	-4.275827	1.533727
54	1	-6.808049	-2.332029	2.188991
55	1	-8.513272	-0.498094	-2.246305
56	1	-8.357856	-2.305418	-3.056370
57	1	-2.923813	-1.613692	3.580838
58	1	-2.647429	-0.123448	2.296636
59	6	-0.545970	1.786530	-0.142421
60	6	0.660640	1.554260	-0.112900
61	6	2.056955	1.265187	-0.086290
62	6	3.008784	2.329578	-0.119314
63	6	2.499231	-0.092341	-0.032165
64	6	2.596167	3.704343	-0.170106
65	6	4.426588	2.026884	-0.108671
66	6	3.919543	-0.385204	-0.014363
67	6	1.567133	-1.183977	0.012921
68	6	3.521520	4.730520	-0.220278
69	1	1.533248	3.924842	-0.168926
70	6	5.358044	3.125111	-0.175827
71	6	4.870314	0.674359	-0.055669
72	6	4.329711	-1.764750	0.067206

73	1	0.504799	-0.962992	0.001942
74	6	2.001169	-2.494959	0.079077
75	6	4.923142	4.435455	-0.229152
76	1	3.190234	5.765343	-0.257965
77	1	6.420645	2.908877	-0.190442
78	6	3.402766	-2.788510	0.110641
79	1	5.389178	-1.993783	0.102714
80	1	1.278832	-3.306280	0.114648
81	1	5.642904	5.248395	-0.281385
82	1	3.733277	-3.822103	0.175009
83	6	6.339171	0.368355	-0.043623
84	6	7.117017	0.596494	1.114337
85	6	6.980683	-0.150891	-1.191140
86	6	8.491630	0.315375	1.128176
87	1	6.639204	0.988488	2.008751
88	6	8.356337	-0.427268	-1.186517
89	1	6.399301	-0.328960	-2.092372
90	6	9.127176	-0.197034	-0.024998
91	1	9.080650	0.489292	2.022496
92	1	8.842661	-0.819326	-2.073555
93	6	10.583432	-0.486632	-0.017165
94	6	13.313143	-1.023043	-0.004529
95	8	14.542625	-1.263411	0.000784
96	7	12.624885	-0.501709	1.101819
97	7	12.488430	-1.248366	-1.117418
98	6	13.050677	-1.793470	-2.360942
99	1	14.119417	-1.953913	-2.217946
100	1	12.556205	-2.740464	-2.601013
101	1	12.882097	-1.085613	-3.179153
102	6	13.340609	-0.206758	2.350941
103	1	14.391708	-0.463623	2.218173
104	1	13.239869	0.857711	2.587074
105	1	12.908889	-0.795513	3.166940
106	7	11.263177	-0.221139	1.130293
107	7	11.122150	-0.992477	-1.158709

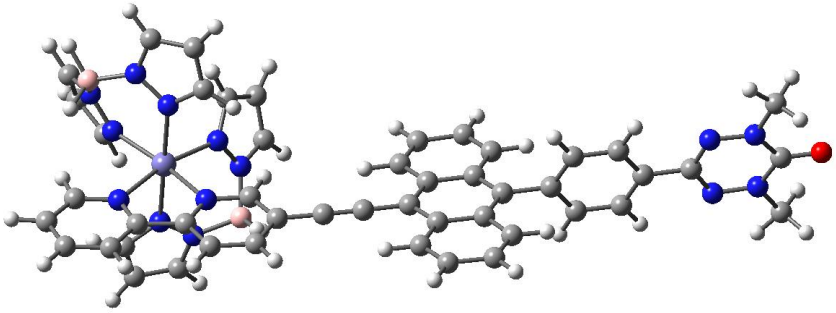


Figure S15. Molecular structure of **1** (S = 5/2) optimized by the DFT calculation.

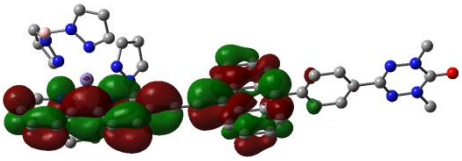
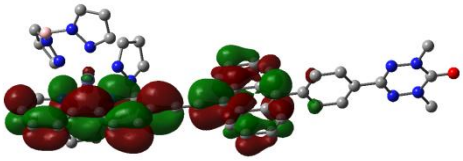
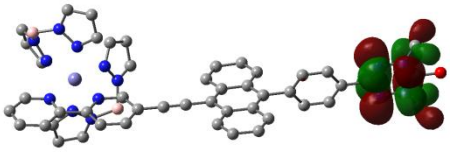
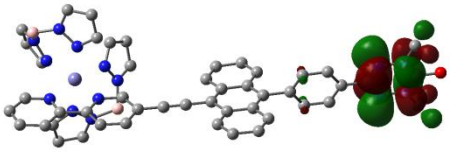
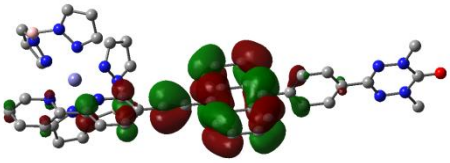
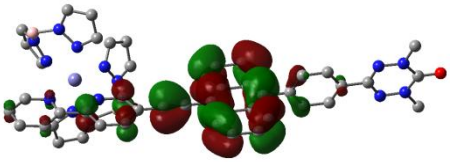
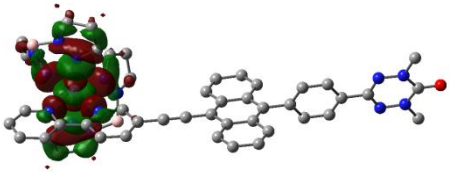
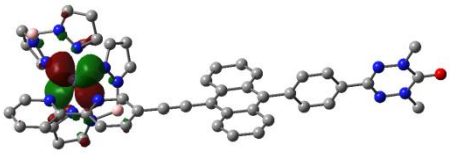
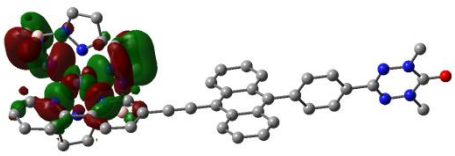
α-LUMO (-0.10265 a.u.) 	β-LUMO+1 (-0.10315 a.u.) 
α- HOMO-1 (SOMO) (-0.20792 a.u.) 	β-LUMO (SOMO) (-0.10590 a.u.) 
α-HOMO (-0.20167 a.u.) 	β-HOMO-1 (-0.20153 a.u.) 
α- HOMO-2 (-0.21078 a.u.) 	β-HOMO (-0.17933 a.u.) 
α- HOMO-3 (-0.22342 a.u.) 	

Figure S16. Molecular orbitals of **1** (S = 5/2) calculated by DFT.

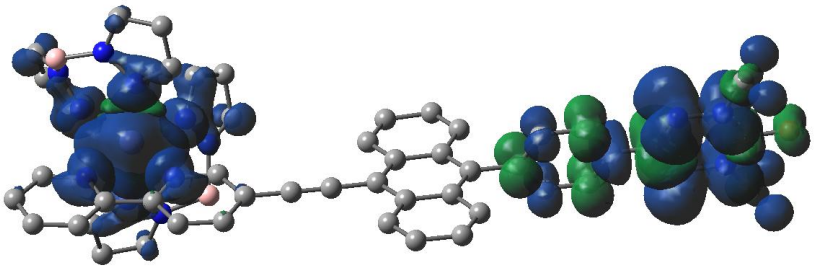


Figure S17. Spin density of **1** (S = 5/2).

Table S4. Molecular coordinates of **1** (S = 5/2) optimized by the DFT calculation

	Atomic number	x/Å	y/Å	z/Å
1	26	5.610579	-0.408219	0.037802
2	5	3.448811	-1.590708	-2.511092
3	7	4.156010	1.299590	-0.172937
4	7	5.838433	-0.662046	-2.154203
5	7	4.109998	-1.981079	-0.034773
6	7	4.897991	-1.261775	-2.981500
7	7	3.490275	-2.430842	-1.196709
8	6	2.824661	1.111261	-0.162580
9	6	1.894859	2.180418	-0.271168
10	6	2.431153	3.486684	-0.401183
11	6	3.817713	3.672123	-0.411352
12	6	4.673014	2.556417	-0.291413
13	6	6.941537	-0.443815	-2.924102
14	6	6.724158	-0.889726	-4.247279
15	6	5.416666	-1.405121	-4.235878
16	6	3.875328	-2.923346	0.922218
17	6	3.100239	-3.979669	0.395126
18	6	2.882142	-3.627343	-0.948533
19	5	7.862104	-0.771214	2.749662
20	7	6.824930	1.475134	-0.132211
21	7	5.380335	-0.240337	2.234520
22	7	7.297131	-1.727117	0.406538
23	7	6.374751	-0.540586	3.156425
24	7	7.950939	-1.852782	1.628839
25	6	8.177067	1.467403	-0.112516
26	6	8.935433	2.643815	-0.237432
27	6	8.256078	3.866190	-0.384905
28	6	6.851995	3.872819	-0.405963
29	6	6.154232	2.653078	-0.277776
30	6	4.246452	-0.011484	2.955576
31	6	4.496376	-0.153456	4.338872
32	6	5.857776	-0.493651	4.418689
33	6	7.669546	-2.801099	-0.346570
34	6	8.567824	-3.620193	0.371835
35	6	8.715464	-2.983298	1.616414

36	1	8.643630	0.498062	0.013164
37	1	10.018924	2.595263	-0.211459
38	1	8.807423	4.797391	-0.481812
39	1	6.318486	4.809280	-0.521598
40	1	4.222807	4.672632	-0.508060
41	1	1.761826	4.336745	-0.492073
42	1	2.484830	0.087231	-0.068861
43	1	4.277136	-2.798541	1.916127
44	1	2.760418	-4.867537	0.905910
45	1	2.342760	-4.141878	-1.729362
46	1	4.833425	-1.853630	-5.025601
47	1	7.409262	-0.852447	-5.080342
48	1	7.827481	0.000177	-2.497024
49	1	3.318620	0.224239	2.457717
50	1	3.798381	-0.036134	5.153571
51	1	6.479261	-0.698966	5.276974
52	1	9.307886	-3.256241	2.476517
53	1	9.032439	-4.537289	0.043730
54	1	7.270920	-2.930229	-1.341267
55	1	8.329242	0.271239	2.348996
56	1	8.464240	-1.176430	3.715987
57	1	2.908303	-2.251198	-3.366482
58	1	2.851135	-0.556902	-2.314203
59	6	0.494180	1.928354	-0.247978
60	6	-0.710006	1.682517	-0.224675
61	6	-2.101922	1.374346	-0.191141
62	6	-3.068198	2.425881	-0.170340
63	6	-2.524559	0.009446	-0.174092
64	6	-2.675559	3.807192	-0.192701
65	6	-4.481056	2.103092	-0.132079
66	6	-3.939737	-0.303599	-0.122542
67	6	-1.577574	-1.069915	-0.199678
68	6	-3.615763	4.821087	-0.191538
69	1	-1.616097	4.042931	-0.214506
70	6	-5.428674	3.189295	-0.144821
71	6	-4.905140	0.743482	-0.103107
72	6	-4.328925	-1.690583	-0.069181

73	1	-0.519659	-0.834601	-0.253384
74	6	-1.992158	-2.388261	-0.160166
75	6	-5.013053	4.506682	-0.173109
76	1	-3.299519	5.861119	-0.210524
77	1	-6.488231	2.958199	-0.138727
78	6	-3.387746	-2.702060	-0.087227
79	1	-5.383507	-1.935682	-0.006603
80	1	-1.258641	-3.189969	-0.179978
81	1	-5.744766	5.310450	-0.185433
82	1	-3.702449	-3.741638	-0.043488
83	6	-6.368381	0.416299	-0.049246
84	6	-7.042262	-0.087753	-1.184825
85	6	-7.108259	0.606337	1.140075
86	6	-8.412204	-0.387859	-1.137465
87	1	-6.490877	-0.236060	-2.109957
88	6	-8.476493	0.301008	1.196370
89	1	-6.605540	0.986879	2.025759
90	6	-9.144732	-0.197725	0.055712
91	1	-8.923034	-0.768683	-2.015486
92	1	-9.035424	0.444899	2.114889
93	6	-10.594249	-0.515893	0.110442
94	6	-13.303183	-1.108137	0.213343
95	8	-14.526322	-1.375685	0.259916
96	7	-12.523619	-1.294111	-0.934191
97	7	-12.591401	-0.595606	1.304085
98	6	-13.300835	-0.356734	2.569326
99	1	-14.117462	0.357240	2.420014
100	1	-13.728328	-1.290375	2.949632
101	1	-12.571866	0.042028	3.275298
102	6	-13.158005	-1.830055	-2.147238
103	1	-13.575695	-2.823470	-1.952834
104	1	-13.972305	-1.174663	-2.473173
105	1	-12.385679	-1.886914	-2.914742
106	7	-11.163359	-1.009781	-1.023404
107	7	-11.233859	-0.286443	1.290162

Table S5. Calculated results for L^1 and **1**[a]

L^1 (UB3LYP/6-31G)	GS (S = 1/2) [Hartree/Particle]	ES (S = 3/2) [Hartree/Particle]	Δ(ES-GS) [kJ mol^{-1}]
E_{el}(UB+HF-LYP)	-1790.070912	-1790.017834	139.4
E_{el} + zero-point energy	-1789.538689	-1789.487712	133.8
E_{el} + thermal energy	-1789.505819	-1789.453168	138.2
E_{el} + thermal enthalpy	-1789.504875	-1789.452224	138.2
E_{el} + thermal free energy	-1789.609691	-1789.561377	126.8
Complex **1** (UB3LYP/LANL2DZ)	LS (S = 1/2) [Hartree/Particle]	HS (S = 5/2) [Hartree/Particle]	Δ(HS-LS) [kJ mol^{-1}]
E_{el}(UB+HF-LYP)	-2868.324909	-2868.321553	8.811
E_{el} + zero-point energy	-2867.492105	-2867.493648	-4.051
E_{el} + thermal energy	-2867.437942	-2867.438939	-2.618
E_{el} + thermal enthalpy	-2867.436998	-2867.437995	-2.618
E_{el} + thermal free energy	-2867.587757	-2867.592155	-11.55

[a] Zero-point energy and enthalpic and entropic thermal contributions to thermodynamic properties at 298.15 K and 1 atm were calculated from harmonic vibrational frequencies obtained at the UB3LYP/6-31G level for L^1 and the UB3LYP/LANL2DZ level for **1** without scaling factors.

Table S6. Bond distances (Å) for optimized structures of **1**

	S = 1/2	S = 5/2
Fe–N1	1.995	2.253
Fe–N2	1.993	2.247
Fe–N3	2.028	2.173
Fe–N4	2.042	2.215
Fe–N5	2.028	2.175
Fe–N6	2.042	2.218