Supplementary Information

Fluorescent solvatochromism and nonfluorescence processes of charge-transfer-type molecules with a 4-nitrophenyl moiety

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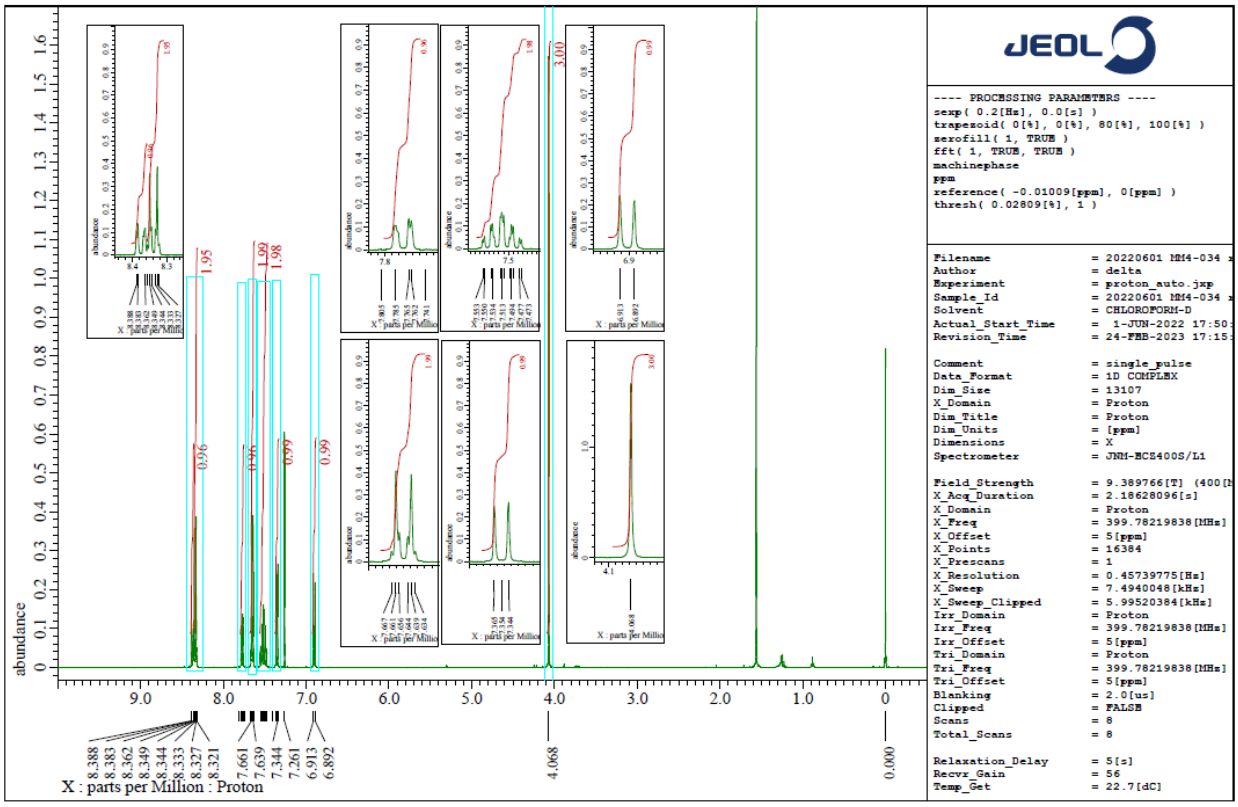

Fig. S1 ^{1}H-NMR spectrum of **1** in $CDCl_3$.

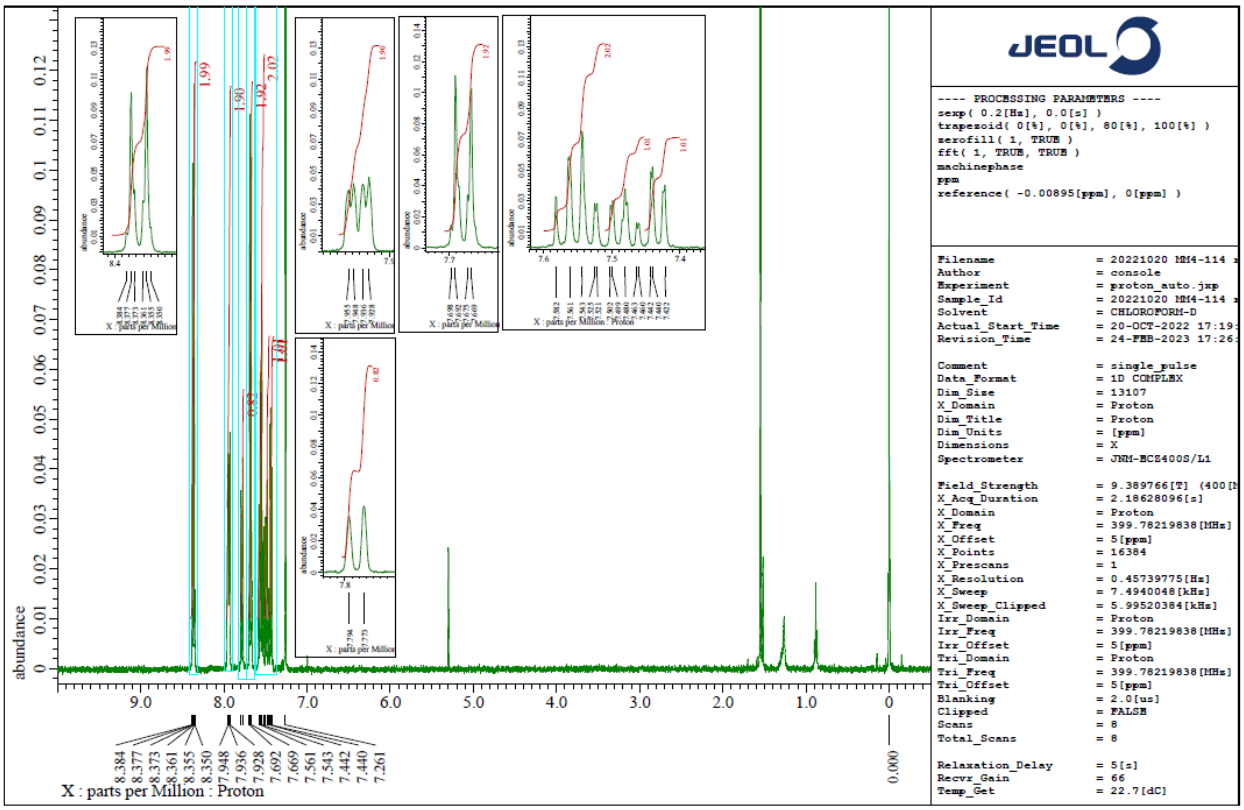

Fig. S2 ^{1}H-NMR spectrum of **2** in $CDCl_3$.

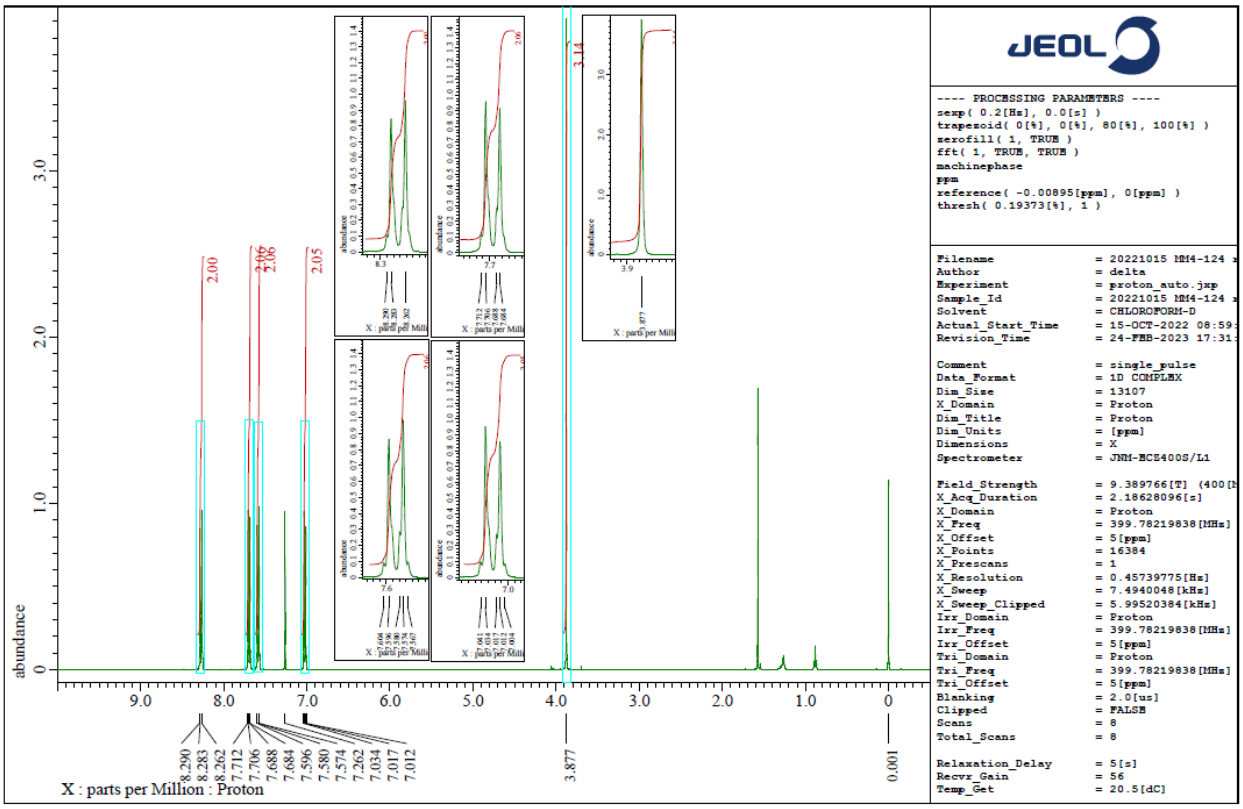

Fig. S3 ^{1}H-NMR spectrum of **3** in $CDCl_3$.

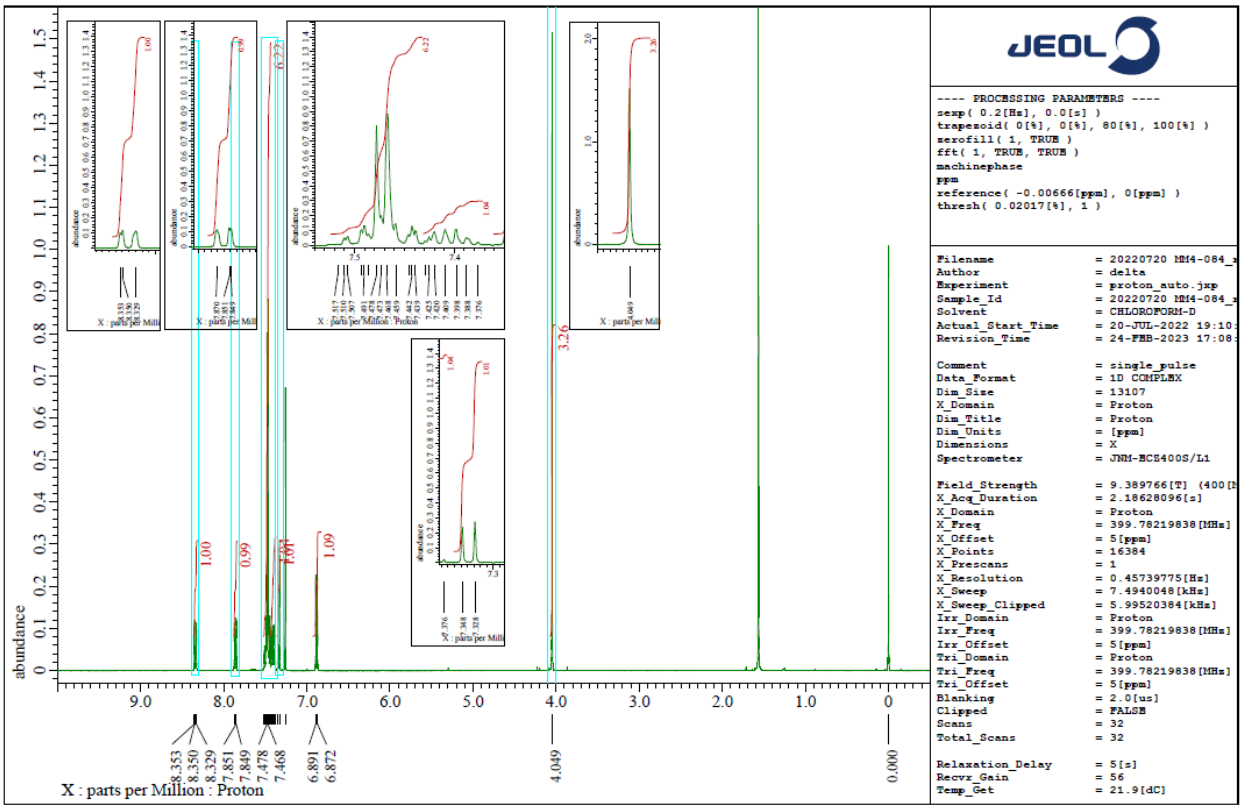

Fig. S4 ^{1}H-NMR spectrum of **4** in $CDCl_3$.

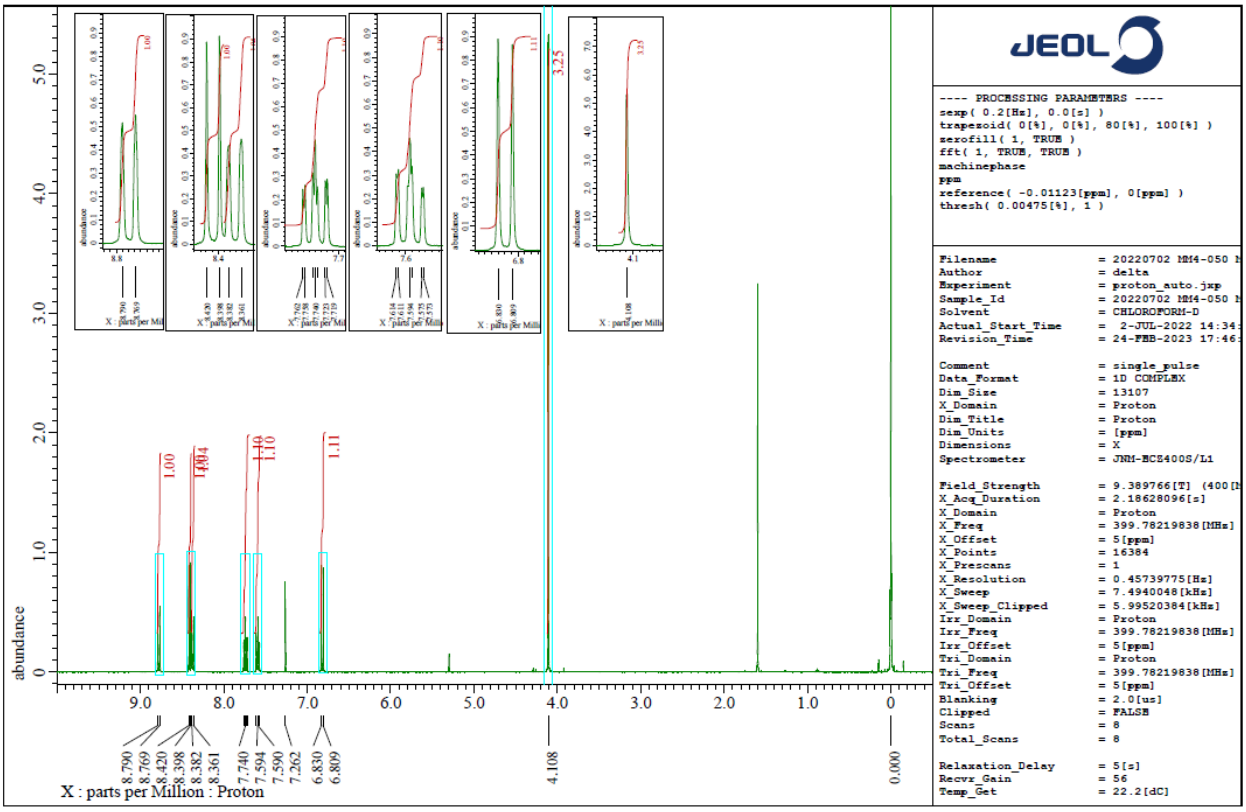

Fig. S5 ^{1}H-NMR spectrum of **5** in $CDCl_3$.

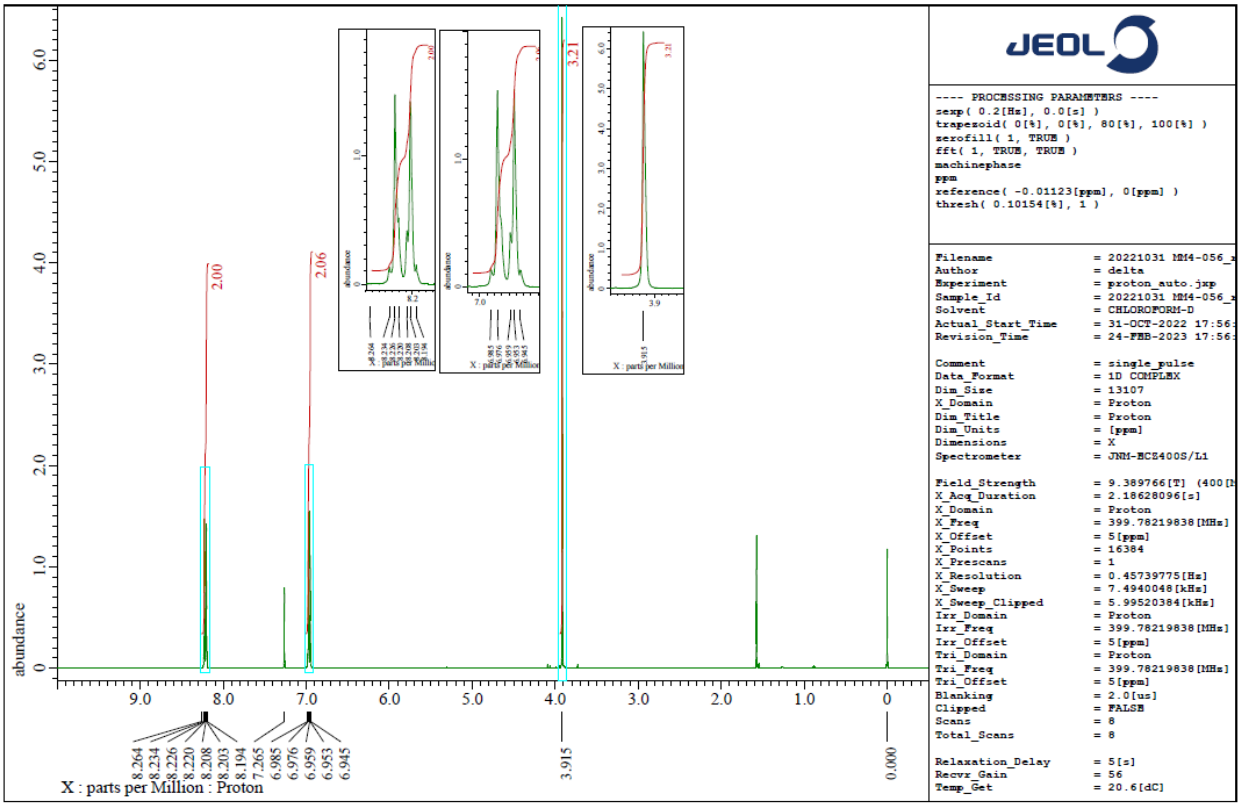

Fig. S6 ^{1}H-NMR spectrum of **6** in $CDCl_3$.

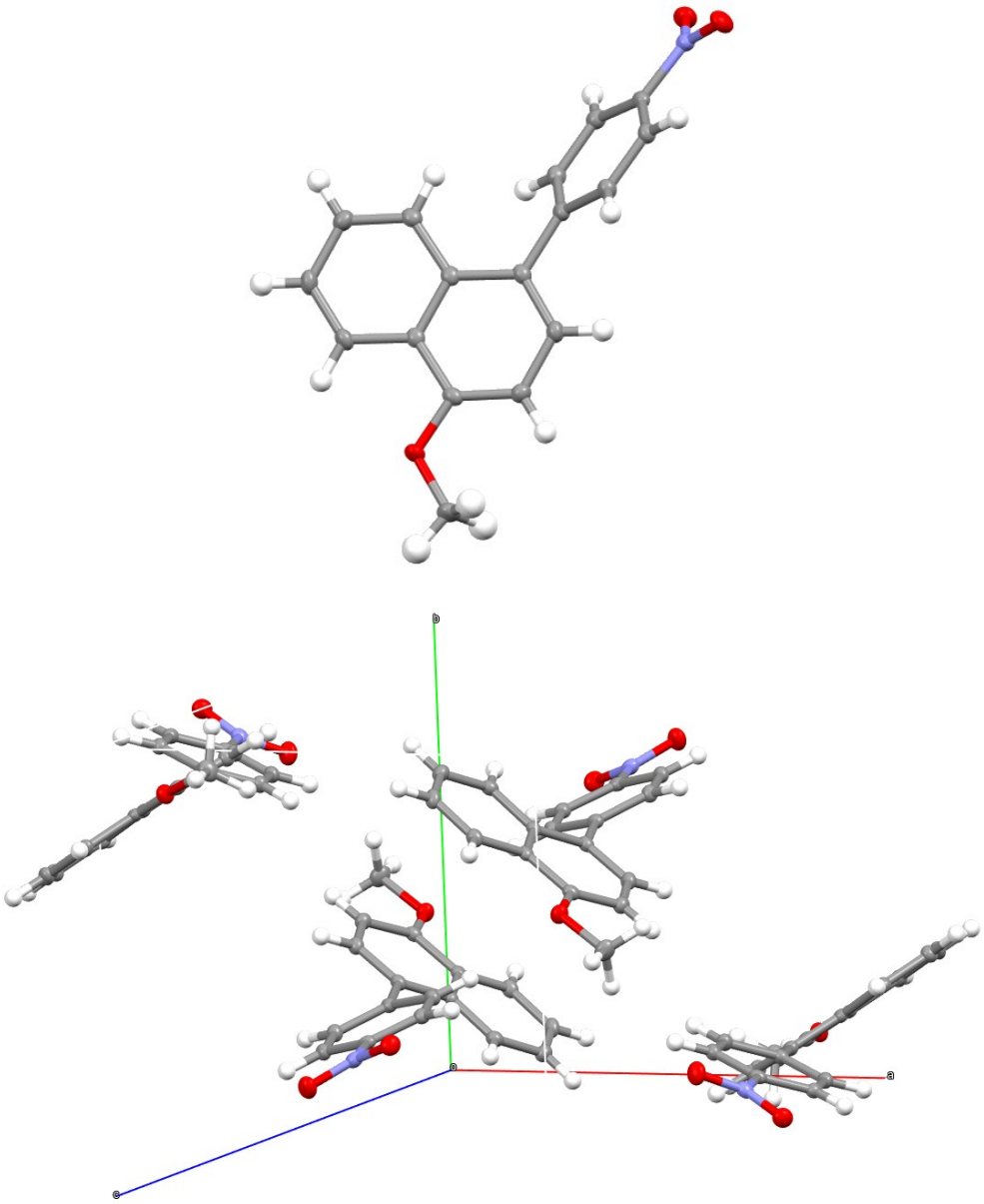

Fig. S7 Crystal structure of **1**.

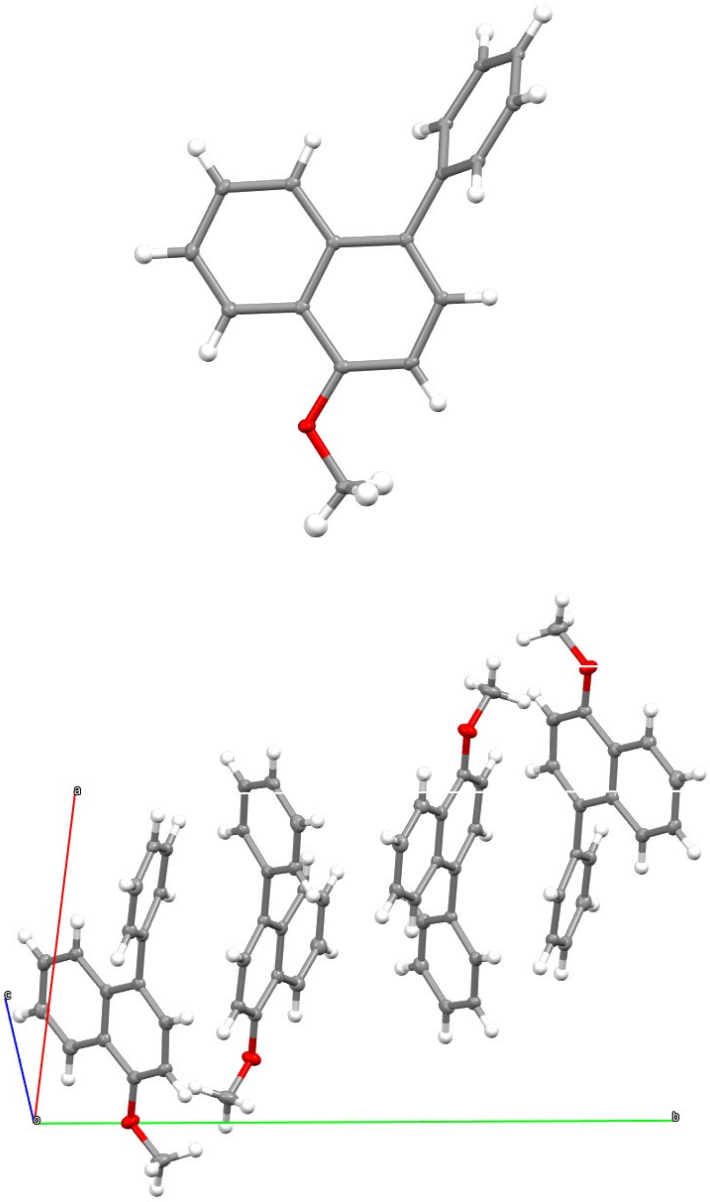

Fig. S8 Crystal structure of **4**.

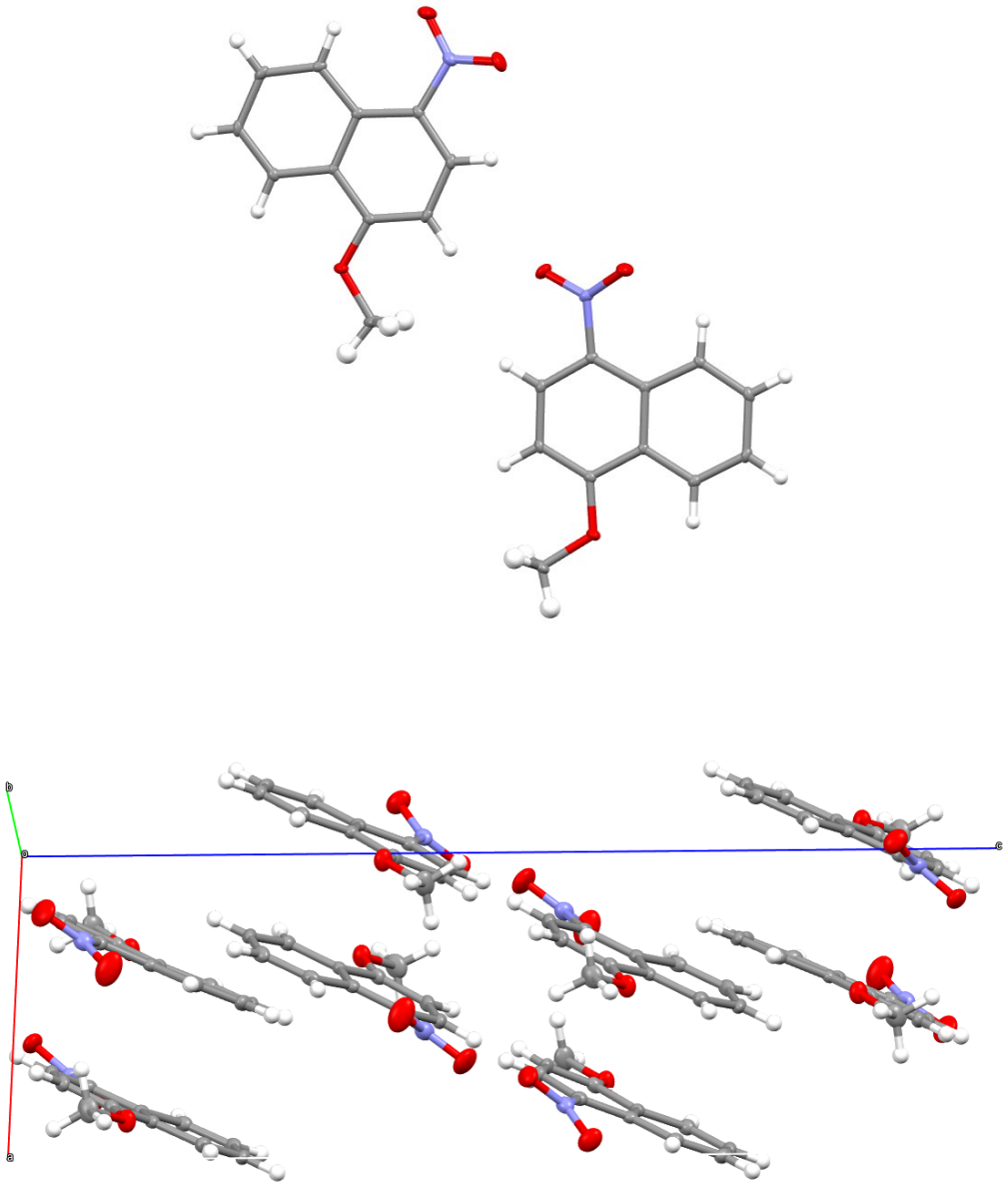

Fig. S9 Crystal structure of **5**.

Table S1 Crystallographic Parameters of **1** and **4**.

	1	**4**
Empirical formula	$C_{17}H_{13}NO_3$	$C_{17}H_{14}O$
Formula weight	279.28	234.28
Temperature / K	93	103
Crystal system	monoclinic	monoclinic
Space group	$P2_1/n$	$P2_1/c$
a / Å	10.0247(4)	11.5001(5)
b / Å	10.2858(3)	17.6022(8)
c / Å	13.4724(5)	5.9565(3)
α / deg	90	90
β / deg	110.611(4)	94.087(5)
γ / deg	90	90
Volume / Å^3	1300.25(9)	1202.69(10)
Z	4	4
ρ_{calc} / g cm^{-3}	1.427	1.294
μ / mm^{-1}	0.099	0.079
F(000)	584	496
Crystal size / mm^3	0.734 × 0.459 × 0.319	0.359 × 0.092 × 0.013
2θ range / deg	4.406–52.742	4.238–52.742
Index ranges	$-11 \le h \le 12$ $-12 \le k \le 12$ $-16 \le l \le 16$	$-12 \le h \le 14$ $-22 \le k \le 18$ $-7 \le l \le 7$
Reflections collected	9971	8417
Independent reflections	2654	2463
Goodness-of-fit on F_2	1.072	1.09
Final R indexes ($I \ge 2\sigma(I)$)	$R_1 = 0.0358$ w$R_2 = 0.0946$	$R_1 = 0.0586$ w$R_2 = 0.0347$

a) $R_1 = \sum\left||F_o| - |F_c|\right| / \sum|F_o|$, $wR_2 = \left\{\sum w\left(F_o^2 - F_c^2\right)^2 / \sum w\left(F_o^2\right)^2\right\}^{1/2}$.

Table S2 Cartesian Coordinates of Optimized Geometries of **1** in CH_2Cl_2.

No.	Label	Ground-State Optimized			S_1-State Optimized		
		X / Å	Y / Å	Z / Å	X / Å	Y / Å	Z / Å
1	C	−0.008	+0.005	+0.006	−0.033	−0.008	+0.003
2	C	+0.000	+0.028	+1.375	−0.039	−0.231	+1.388
3	H	+0.926	+0.038	+1.931	+0.889	−0.323	+1.933
4	C	−1.220	+0.056	+2.081	−1.228	−0.290	+2.071
5	H	−1.184	+0.098	+3.163	−1.195	−0.396	+3.146
6	C	−2.435	+0.065	+1.448	−2.487	−0.108	+1.438
7	C	−2.468	+0.008	+0.016	−2.495	−0.037	−0.008
8	C	−3.677	−0.066	−0.722	−3.675	−0.137	−0.759
9	H	−4.620	−0.086	−0.193	−4.614	−0.318	−0.256
10	C	−3.673	−0.130	−2.089	−3.660	−0.047	−2.141
11	H	−4.611	−0.190	−2.628	−4.589	−0.128	−2.692
12	C	−2.457	−0.125	−2.800	−2.458	+0.131	−2.821
13	H	−2.464	−0.172	−3.882	−2.448	+0.210	−3.900
14	C	−1.270	−0.074	−2.120	−1.271	+0.172	−2.117
15	H	−0.330	−0.085	−2.654	−0.329	+0.269	−2.638
16	C	−1.247	−0.015	−0.707	−1.266	+0.064	−0.717
17	O	+1.102	−0.009	−0.766	+1.070	+0.110	−0.716
18	C	+2.372	+0.000	−0.128	+2.358	+0.018	−0.093
19	H	+2.502	+0.902	+0.474	+2.479	+0.808	+0.648
20	H	+3.108	−0.013	−0.927	+3.076	+0.150	−0.897
21	H	+2.500	−0.884	+0.501	+2.489	−0.962	+0.366
22	C	−3.671	+0.143	+2.268	−3.660	+0.049	+2.247
23	C	−3.906	−0.798	+3.273	−3.778	−0.631	+3.497
24	H	−3.206	−1.610	+3.417	−3.003	−1.321	+3.805
25	C	−5.027	−0.716	+4.081	−4.880	−0.489	+4.292
26	H	−5.214	−1.445	+4.856	−4.975	−1.036	+5.218
27	C	−5.919	+0.323	+3.873	−5.921	+0.382	+3.898
28	C	−5.719	+1.276	+2.887	−5.814	+1.106	+2.689
29	H	−6.429	+2.080	+2.758	−6.595	+1.804	+2.425
30	C	−4.594	+1.179	+2.089	−4.720	+0.934	+1.889
31	H	−4.419	+1.928	+1.328	−4.629	+1.536	+0.996
32	N	−7.109	+0.418	+4.721	−7.043	+0.541	+4.711
33	O	−7.259	−0.415	+5.598	−7.102	−0.080	+5.805
34	O	−7.895	+1.326	+4.511	−7.973	+1.300	+4.330

Table S3 Cartesian Coordinates of Optimized Geometries of **2** in CH_2Cl_2.

No.	Label	Ground-State Optimized			S_1-State Optimized		
		X / Å	Y / Å	Z / Å	X / Å	Y / Å	Z / Å
1	C	+0.000	+0.000	+0.000	−0.037	−0.023	+0.003
2	C	+0.000	−0.001	+1.365	−0.047	−0.312	+1.367
3	H	+0.933	−0.001	+1.914	+0.888	−0.434	+1.898
4	C	−1.220	+0.011	+2.072	−1.237	−0.407	+2.051
5	H	−1.203	+0.032	+3.155	−1.214	−0.566	+3.120
6	C	−2.427	+0.028	+1.417	−2.490	−0.176	+1.413
7	C	−2.454	+0.001	−0.013	−2.487	−0.039	−0.031
8	C	−3.658	−0.056	−0.764	−3.649	−0.123	−0.804
9	H	−4.607	−0.084	−0.247	−4.595	−0.349	−0.333
10	C	−3.635	−0.089	−2.132	−3.611	+0.035	−2.186
11	H	−4.566	−0.135	−2.684	−4.531	−0.029	−2.754
12	C	−2.409	−0.069	−2.830	−2.402	+0.259	−2.839
13	H	−2.406	−0.092	−3.913	−2.378	+0.392	−3.913
14	C	−1.232	−0.033	−2.137	−1.229	+0.278	−2.112
15	H	−0.284	−0.031	−2.663	−0.277	+0.411	−2.612
16	C	−1.220	−0.006	−0.720	−1.240	+0.098	−0.712
17	H	+0.932	−0.002	−0.553	+0.904	+0.072	−0.527
18	C	−3.675	+0.082	+2.223	−3.664	−0.027	+2.210
19	C	−3.941	−0.909	+3.169	−3.794	−0.712	+3.460
20	H	−3.254	−1.739	+3.282	−3.018	−1.395	+3.778
21	C	−5.076	−0.854	+3.960	−4.912	−0.589	+4.232
22	H	−5.289	−1.621	+4.690	−5.018	−1.143	+5.153
23	C	−5.946	+0.210	+3.794	−5.958	+0.268	+3.822
24	C	−5.714	+1.214	+2.867	−5.839	+1.005	+2.622
25	H	−6.409	+2.035	+2.770	−6.622	+1.699	+2.353
26	C	−4.576	+1.141	+2.084	−4.730	+0.853	+1.842
27	H	−4.376	+1.926	+1.366	−4.626	+1.471	+0.961
28	N	−7.151	+0.279	+4.626	−7.100	+0.405	+4.616
29	O	−7.329	−0.598	+5.453	−7.165	−0.215	+5.705
30	O	−7.918	+1.209	+4.451	−8.034	+1.143	+4.217

Table S4 Cartesian Coordinates of Optimized Geometries of **3** in CH_2Cl_2.

No.	Label	Ground-State Optimized			S_1-State Optimized		
		X / Å	Y / Å	Z / Å	X / Å	Y / Å	Z / Å
1	C	+0.000	+0.000	+0.000	+0.008	+0.089	+0.021
2	C	+0.000	+0.000	+1.392	−0.013	−0.197	+1.400
3	H	+0.924	−0.001	+1.951	+0.903	−0.405	+1.935
4	C	−1.208	+0.009	+2.077	−1.204	−0.206	+2.072
5	H	−1.187	+0.028	+3.160	−1.182	−0.411	+3.132
6	C	−2.429	+0.026	+1.408	−2.450	+0.071	+1.425
7	C	−2.406	+0.030	+0.008	−2.384	+0.359	+0.023
8	H	−3.336	+0.022	−0.548	−3.289	+0.560	−0.530
9	C	−1.215	+0.014	−0.688	−1.203	+0.366	−0.653
10	H	−1.202	+0.004	−1.770	−1.163	+0.576	−1.714
11	O	+1.118	−0.011	−0.766	+1.104	+0.121	−0.722
12	C	+2.384	−0.033	−0.120	+2.382	−0.146	−0.132
13	H	+2.526	+0.857	+0.498	+2.606	+0.591	+0.640
14	H	+3.125	−0.043	−0.915	+3.098	−0.062	−0.943
15	H	+2.497	−0.930	+0.494	+2.406	−1.154	+0.283
16	C	−3.709	+0.042	+2.150	−3.685	+0.062	+2.136
17	C	−3.854	−0.672	+3.344	−3.773	−0.405	+3.489
18	H	−3.030	−1.263	+3.722	−2.895	−0.784	+3.990
19	C	−5.045	−0.660	+4.045	−4.952	−0.419	+4.173
20	H	−5.159	−1.217	+4.964	−5.008	−0.785	+5.187
21	C	−6.107	+0.074	+3.540	−6.132	+0.050	+3.547
22	C	−6.004	+0.792	+2.359	−6.086	+0.523	+2.214
23	H	−6.846	+1.361	+1.995	−6.994	+0.884	+1.756
24	C	−4.803	+0.772	+1.673	−4.903	+0.522	+1.537
25	H	−4.710	+1.350	+0.763	−4.899	+0.904	+0.527
26	N	−7.372	+0.091	+4.274	−7.342	+0.046	+4.246
27	O	−7.446	−0.547	+5.311	−7.351	−0.332	+5.448
28	O	−8.295	+0.743	+3.816	−8.389	+0.420	+3.653

Table S5 Cartesian Coordinates of Optimized Geometries of **4** in CH_2Cl_2.

No.	Label	Ground-State Optimized			S_1-State Optimized		
		X / Å	Y / Å	Z / Å	X / Å	Y / Å	Z / Å
1	C	+0.000	+0.000	+0.000	−0.056	−0.013	+0.006
2	C	+0.000	−0.001	+1.368	−0.065	−0.412	+1.367
3	H	+0.923	−0.002	+1.930	+0.864	−0.584	+1.891
4	C	−1.225	+0.014	+2.069	−1.248	−0.565	+2.026
5	H	−1.195	+0.034	+3.153	−1.224	−0.828	+3.074
6	C	−2.438	+0.030	+1.434	−2.510	−0.252	+1.394
7	C	−2.460	+0.002	+0.001	−2.512	−0.118	−0.053
8	C	−3.666	−0.051	−0.744	−3.663	−0.220	−0.830
9	H	−4.608	−0.073	−0.214	−4.600	−0.481	−0.359
10	C	−3.657	−0.084	−2.112	−3.642	−0.007	−2.221
11	H	−4.593	−0.129	−2.656	−4.565	−0.071	−2.785
12	C	−2.438	−0.068	−2.818	−2.452	+0.284	−2.852
13	H	−2.441	−0.091	−3.901	−2.426	+0.469	−3.919
14	C	−1.254	−0.034	−2.133	−1.263	+0.320	−2.117
15	H	−0.312	−0.033	−2.664	−0.323	+0.520	−2.613
16	C	−1.235	−0.006	−0.719	−1.265	+0.086	−0.732
17	O	+1.116	−0.001	−0.768	+1.073	+0.178	−0.693
18	C	+2.380	−0.003	−0.121	+2.340	+0.059	−0.050
19	H	+2.507	+0.889	+0.497	+2.424	+0.769	+0.776
20	H	+3.123	−0.003	−0.915	+3.079	+0.293	−0.811
21	H	+2.505	−0.897	+0.495	+2.499	−0.958	+0.314
22	C	−3.686	+0.073	+2.244	−3.664	−0.044	+2.213
23	C	−3.960	−0.933	+3.170	−3.763	−0.633	+3.509
24	H	−3.275	−1.768	+3.265	−2.989	−1.305	+3.856
25	C	−5.099	−0.882	+3.963	−4.863	−0.422	+4.314
26	H	−5.295	−1.676	+4.675	−4.915	−0.911	+5.280
27	C	−5.986	+0.179	+3.842	−5.905	+0.404	+3.899
28	H	−6.876	+0.221	+4.459	−6.763	+0.572	+4.540
29	C	−5.724	+1.189	+2.924	−5.820	+1.025	+2.649
30	H	−6.407	+2.026	+2.827	−6.606	+1.699	+2.327
31	C	−4.586	+1.136	+2.132	−4.739	+0.806	+1.825
32	H	−4.384	+1.934	+1.428	−4.681	+1.335	+0.885

Table S6 Cartesian Coordinates of Optimized Geometries of **5** in CH_2Cl_2.

No.	Label	Ground-State Optimized			S_1-State Optimized		
		X / Å	Y / Å	Z / Å	X / Å	Y / Å	Z / Å
1	C	+0.000	+0.000	+0.000	−0.004	+0.006	−0.003
2	C	+0.000	+0.000	+1.375	−0.029	+0.074	+1.398
3	H	+0.919	+0.000	+1.941	+0.890	+0.104	+1.963
4	C	−1.218	+0.012	+2.061	−1.236	+0.101	+2.053
5	H	−1.223	+0.019	+3.141	−1.274	+0.153	+3.133
6	C	−2.410	+0.041	+1.389	−2.449	+0.060	+1.344
7	C	−2.472	+0.026	−0.040	−2.462	−0.007	−0.079
8	C	−3.661	−0.018	−0.809	−3.658	−0.046	−0.809
9	H	−4.617	−0.006	−0.312	−4.600	−0.024	−0.278
10	C	−3.612	−0.067	−2.177	−3.624	−0.111	−2.188
11	H	−4.537	−0.102	−2.740	−4.551	−0.140	−2.747
12	C	−2.382	−0.071	−2.857	−2.404	−0.137	−2.865
13	H	−2.360	−0.105	−3.939	−2.388	−0.188	−3.946
14	C	−1.215	−0.043	−2.145	−1.214	−0.100	−2.163
15	H	−0.259	−0.058	−2.649	−0.268	−0.120	−2.687
16	C	−1.232	−0.001	−0.732	−1.226	−0.035	−0.766
17	O	+1.110	−0.004	−0.750	+1.101	−0.023	−0.707
18	C	+2.382	−0.011	−0.107	+2.388	+0.014	−0.066
19	H	+2.510	+0.881	+0.508	+2.500	+0.939	+0.499
20	H	+3.116	−0.013	−0.907	+3.108	−0.019	−0.877
21	H	+2.501	−0.908	+0.505	+2.510	−0.854	+0.580
22	N	−3.604	+0.108	+2.219	−3.652	+0.089	+2.065
23	O	−3.551	−0.369	+3.344	−3.931	−1.016	+2.680
24	O	−4.603	+0.655	+1.776	−3.945	+1.248	+2.562

Table S7 Cartesian Coordinates of Optimized Geometries of **6** in CH_2Cl_2.

No.	Label	Ground-State Optimized			S_1-State Optimized		
		X / Å	Y / Å	Z / Å	X / Å	Y / Å	Z / Å
1	C	+0.000	+0.000	+0.000	+0.009	+0.000	−0.004
2	C	+0.000	+0.000	+1.396	+0.007	+0.000	+1.391
3	H	+0.924	+0.000	+1.956	+0.931	+0.000	+1.951
4	C	−1.204	+0.000	+2.080	−1.194	+0.000	+2.080
5	H	−1.221	+0.000	+3.160	−1.205	+0.000	+3.162
6	C	−2.392	+0.000	+1.369	−2.390	+0.000	+1.368
7	C	−2.407	+0.000	−0.023	−2.401	+0.000	−0.032
8	H	−3.348	+0.000	−0.553	−3.340	+0.000	−0.569
9	C	−1.212	+0.000	−0.703	−1.203	+0.000	−0.706
10	H	−1.186	+0.000	−1.784	−1.178	+0.000	−1.788
11	O	+1.112	+0.000	−0.754	+1.125	+0.000	−0.762
12	C	+2.385	+0.000	−0.112	+2.394	+0.000	−0.118
13	H	+3.119	+0.000	−0.913	+3.132	+0.000	−0.915
14	H	+2.512	−0.894	+0.501	+2.521	−0.894	+0.497
15	H	+2.512	+0.894	+0.501	+2.521	+0.894	+0.497
16	N	−3.654	+0.000	+2.094	−3.586	+0.000	+2.053
17	O	−3.622	+0.000	+3.315	−3.747	+0.000	+3.331
18	O	−4.691	+0.000	+1.450	−4.771	+0.000	+1.546

Table S8 Calculated Excited States of **1** in CH_2Cl_2.

Excited State	Transition	Energy (Wavelength)	Oscillator Strength
S_1	MO69 → MO74 (14%) MO73 → MO74 (86%)	3.6858 eV (336.38 nm)	0.4125
S_2	MO68 → MO74 (80%) MO68 → MO79 (20%)	4.0206 eV (308.37 nm)	0.0097
S_3	MO73 → MO74 (15%) MO73 → MO75 (85%)	4.4275 eV (280.03 nm)	0.1019
S_4	MO70 → MO74 (15%) MO72 → MO75 (34%) MO73 → MO75 (11%) MO73 → MO76 (40%)	4.4891 eV (276.19 nm)	0.0233
S_5	MO69 → MO74 (11%) MO69 → MO76 (9%) MO70 → MO74 (48%) MO71 → MO74 (15%) MO73 → MO77 (17%)	4.5880 eV (270.24 nm)	0.0429
S_6	MO63 → MO74 (11%) MO64 → MO74 (62%) MO64 → MO79 (15%) MO67 → MO74 (12%)	4.6131 eV (268.76 nm)	0.0007
S_7	MO69 → MO74 (52%) MO71 → MO74 (48%)	4.8917 eV (253.46 nm)	0.2753
S_8	MO72 → MO74 (58%) MO73 → MO76 (42%)	5.2142 eV (237.78 nm)	0.1666
S_9	MO73 → MO76 (45%) MO73 → MO77 (55%)	5.2766 eV (234.97 nm)	0.0214
S_{10}	MO71 → MO77 (11%) MO72 → MO74 (12%) MO72 → MO75 (50%) MO73 → MO77 (27%)	5.6363 eV (219.98 nm)	0.7787
S_{11}	MO73 → MO78 (64%) MO73 → MO80 (23%) MO73 → MO85 (13%)	5.6701 eV (218.66 nm)	0.0017
S_{12}	MO69 → MO74 (27%) MO71 → MO75 (33%) MO73 → MO79 (40%)	5.7330 eV (216.26 nm)	0.0506
S_{13}	MO69 → MO75 (14%) MO71 → MO74 (29%) MO73 → MO79 (57%)	5.8675 eV (211.31 nm)	0.0237
S_{14}	MO71 → MO74 (28%) MO71 → MO75 (39%) MO72 → MO76 (33%)	6.0796 eV (203.94 nm)	0.1173
S_{15}	MO66 → MO74 (22%) MO73 → MO81 (78%)	6.1703 eV (200.94 nm)	0.0051

S_{16}	MO66 → MO74 (63%) MO70 → MO75 (15%) MO70 → MO79 (22%)	6.2169 eV (199.43 nm)	0.0481
S_{17}	MO73 → MO79 (15%) MO73 → MO80 (53%) MO73 → MO83 (21%) MO73 → MO89 (11%)	6.2538 eV (198.25 nm)	0.0027
S_{18}	MO71 → MO74 (12%) MO71 → MO75 (34%) MO72 → MO77 (29%) MO73 → MO80 (11%) MO73 → MO81 (14%)	6.3018 eV (196.74 nm)	0.3273
S_{19}	MO66 → MO74 (23%) MO69 → MO77 (14%) MO71 → MO76 (29%) MO72 → MO75 (14%) MO72 → MO79 (11%) MO72 → MO88 (9%)	6.3372 eV (195.64 nm)	0.1867
S_{20}	MO69 → MO79 (9%) MO70 → MO76 (24%) MO70 → MO77 (13%) MO71 → MO75 (15%) MO71 → MO77 (8%) MO73 → MO82 (19%) MO73 → MO87 (12%)	6.4003 eV (193.72 nm)	0.3011
S_{21}	MO66 → MO74 (44%) MO69 → MO76 (29%) MO71 → MO77 (27%)	6.4277 eV (192.89 nm)	0.2335
S_{22}	MO71 → MO81 (12%) MO73 → MO78 (15%) MO73 → MO80 (23%) MO73 → MO84 (50%)	6.5173 eV (190.24 nm)	0.0176
S_{23}	MO69 → MO75 (13%) MO69 → MO79 (10%) MO70 → MO76 (29%) MO70 → MO77 (16%) MO73 → MO83 (13%) MO73 → MO85 (19%)	6.5719 eV (188.66 nm)	0.0867
S_{24}	MO69 → MO75 (37%) MO71 → MO75 (9%) MO73 → MO82 (15%) MO73 → MO84 (10%) MO73 → MO85 (9%) MO73 → MO86 (11%) MO73 → MO88 (9%)	6.6691 eV (185.91 nm)	0.0208

S_{25}	MO70 → MO75 (12%) MO70 → MO76 (14%) MO70 → MO77 (8%) MO71 → MO76 (8%) MO72 → MO76 (9%) MO73 → MO82 (11%) MO73 → MO84 (8%) MO73 → MO85 (7%) MO73 → MO86 (7%) MO73 → MO87 (7%) MO73 → MO88 (9%)	6.6987 eV (185.09 nm)	0.0657
S_{26}	MO72 → MO76 (32%) MO72 → MO77 (42%) MO72 → MO84 (14%) MO73 → MO89 (12%)	6.7438 eV (183.85 nm)	0.0140
S_{27}	MO71 → MO78 (11%) MO73 → MO78 (13%) MO73 → MO83 (43%) MO73 → MO84 (10%) MO73 → MO86 (12%) MO73 → MO94 (11%)	6.7844 eV (182.75 nm)	0.0099
S_{28}	MO60 → MO74 (60%) MO61 → MO74 (26%) MO72 → MO77 (14%)	6.8217 eV (181.75 nm)	0.0035
S_{29}	MO70 → MO76 (9%) MO72 → MO76 (16%) MO72 → MO77 (11%) MO72 → MO78 (17%) MO73 → MO86 (30%) MO73 → MO88 (17%)	6.8732 eV (180.39 nm)	0.0443
S_{30}	MO69 → MO75 (19%) MO69 → MO76 (10%) MO70 → MO75 (56%) MO71 → MO75 (15%)	6.8888 eV (179.98 nm)	0.1001

HOMO: MO73, LUMO: MO74

Table S9 Calculated Excited States of **2** in CH_2Cl_2.

Excited State	Transition	Energy (Wavelength)	Oscillator Strength
S_1	MO63 → MO66 (22%) MO65 → MO66 (78%)	3.9484 eV (314.01 nm)	0.2871
S_2	MO60 → MO66 (65%) MO65 → MO66 (35%)	4.0393 eV (306.95 nm)	0.1085
S_3	MO62 → MO66 (30%) MO64 → MO67 (34%) MO65 → MO67 (11%) MO65 → MO69 (25%)	4.5531 eV (272.30 nm)	0.0199
S_4	MO57 → MO66 (42%) MO58 → MO66 (46%) MO63 → MO66 (12%)	4.6065 eV (269.15 nm)	0.0052
S_5	MO57 → MO66 (25%) MO58 → MO66 (26%) MO62 → MO66 (49%)	4.6159 eV (268.60 nm)	0.0200
S_6	MO63 → MO66 (16%) MO65 → MO66 (13%) MO65 → MO67 (71%)	4.6523 eV (266.50 nm)	0.0240
S_7	MO62 → MO66 (30%) MO63 → MO66 (70%)	4.9154 eV (252.24 nm)	0.2923
S_8	MO64 → MO66 (69%) MO64 → MO71 (14%) MO65 → MO69 (17%)	5.2003 eV (238.42 nm)	0.0962
S_9	MO62 → MO66 (12%) MO63 → MO68 (11%) MO64 → MO67 (10%) MO65 → MO68 (42%) MO65 → MO69 (25%)	5.5395 eV (223.82 nm)	0.0948
S_{10}	MO64 → MO67 (77%) MO65 → MO68 (23%)	5.7669 eV (214.99 nm)	1.1268
S_{11}	MO61 → MO66 (13%) MO64 → MO67 (11%) MO65 → MO71 (50%) MO65 → MO73 (11%) MO65 → MO74 (15%)	5.9347 eV (208.91 nm)	0.0761
S_{12}	MO65 → MO70 (67%) MO65 → MO71 (19%) MO65 → MO79 (14%)	6.0127 eV (206.20 nm)	0.0033
S_{13}	MO61 → MO67 (21%) MO63 → MO67 (44%) MO65 → MO71 (20%) MO65 → MO74 (15%)	6.1476 eV (201.68 nm)	0.0168
S_{14}	MO63 → MO68 (19%) MO64 → MO68 (81%)	6.1843 eV (200.48 nm)	0.4537

S_{15}	MO59 → MO66 (47%) MO62 → MO71 (13%) MO65 → MO68 (15%) MO65 → MO69 (13%) MO65 → MO70 (12%)	6.2381 eV (198.75 nm)	0.0268
S_{16}	MO59 → MO66 (21%) MO61 → MO66 (16%) MO62 → MO67 (10%) MO63 → MO67 (14%) MO63 → MO68 (16%) MO64 → MO68 (11%) MO64 → MO71 (12%)	6.3836 eV (194.22 nm)	0.0961
S_{17}	MO61 → MO66 (25%) MO62 → MO71 (11%) MO63 → MO67 (38%) MO65 → MO71 (13%) MO65 → MO75 (13%)	6.3937 eV (193.92 nm)	0.0422
S_{18}	MO61 → MO67 (9%) MO62 → MO68 (29%) MO62 → MO69 (13%) MO63 → MO71 (12%) MO65 → MO71 (14%) MO65 → MO72 (11%) MO65 → MO75 (12%)	6.4321 eV (192.76 nm)	0.1338
S_{19}	MO59 → MO66 (28%) MO61 → MO69 (11%) MO62 → MO67 (17%) MO63 → MO69 (28%) MO64 → MO75 (16%)	6.4508 eV (192.20 nm)	0.0905
S_{20}	MO62 → MO67 (8%) MO62 → MO68 (21%) MO62 → MO69 (9%) MO63 → MO71 (11%) MO65 → MO73 (33%) MO65 → MO75 (8%) MO65 → MO76 (10%)	6.5386 eV (189.62 nm)	0.0362
S_{21}	MO63 → MO70 (10%) MO65 → MO70 (15%) MO65 → MO72 (40%) MO65 → MO73 (24%) MO65 → MO75 (11%)	6.5943 eV (188.02 nm)	0.0093
S_{22}	MO62 → MO67 (49%) MO62 → MO68 (21%) MO65 → MO74 (30%)	6.7031 eV (184.97 nm)	0.1006
S_{23}	MO64 → MO68 (40%) MO64 → MO69 (44%) MO64 → MO72 (16%)	6.7202 eV (184.50 nm)	0.0192

S_{24}	MO62 → MO67 (62%) MO65 → MO71 (18%) MO65 → MO75 (20%)	6.7742 eV (183.02 nm)	0.0957
S_{25}	MO53 → MO66 (9%) MO54 → MO66 (17%) MO57 → MO67 (10%) MO58 → MO66 (12%) MO61 → MO67 (28%) MO62 → MO67 (12%) MO65 → MO77 (12%)	6.8464 eV (181.09 nm)	0.0137
S_{26}	MO65 → MO70 (25%) MO65 → MO77 (75%)	6.8652 eV (180.60 nm)	0.0052
S_{27}	MO54 → MO66 (16%) MO64 → MO68 (18%) MO64 → MO69 (17%) MO64 → MO70 (37%) MO65 → MO75 (12%)	6.8875 eV (180.01 nm)	0.0322
S_{28}	MO53 → MO66 (13%) MO54 → MO66 (57%) MO55 → MO66 (16%) MO64 → MO72 (14%)	6.9054 eV (179.55 nm)	0.0135
S_{29}	MO54 → MO66 (24%) MO55 → MO67 (10%) MO57 → MO66 (23%) MO58 → MO67 (21%) MO61 → MO67 (22%)	6.9274 eV (178.98 nm)	0.0075
S_{30}	MO65 → MO74 (27%) MO65 → MO75 (29%) MO65 → MO76 (18%) MO65 → MO77 (14%) MO65 → MO78 (12%)	7.0359 eV (176.22 nm)	0.0383

HOMO: MO65, LUMO: MO66

Table S10 Calculated Excited States of **3** in CH_2Cl_2.

Excited State	Transition	Energy (Wavelength)	Oscillator Strength
S_1	MO56 → MO61 (13%) MO57 → MO61 (20%) MO60 → MO61 (67%)	3.8793 eV (319.60 nm)	0.7012
S_2	MO56 → MO61 (77%) MO56 → MO63 (23%)	4.0174 eV (308.62 nm)	0.0300
S_3	MO58 → MO61	4.5699 eV (271.31 nm)	0.0241
S_4	MO54 → MO61 (78%) MO54 → MO63 (22%)	4.6169 eV (268.55 nm)	0.0009
S_5	MO59 → MO63 (25%) MO60 → MO62 (25%) MO60 → MO64 (50%)	4.9254 eV (251.72 nm)	0.0097
S_6	MO57 → MO61 (62%) MO60 → MO63 (38%)	5.2301 eV (237.06 nm)	0.0061
S_7	MO57 → MO61 (8%) MO57 → MO62 (9%) MO58 → MO61 (13%) MO59 → MO61 (24%) MO60 → MO62 (38%) MO60 → MO63 (8%)	5.4451 eV (227.70 nm)	0.0124
S_8	MO57 → MO61 (12%) MO59 → MO61 (47%) MO60 → MO64 (41%)	5.5739 eV (222.44 nm)	0.1167
S_9	MO60 → MO61 (23%) MO60 → MO63 (59%) MO60 → MO64 (18%)	5.6510 eV (219.40 nm)	0.1839
S_{10}	MO60 → MO65 (76%) MO60 → MO66 (24%)	6.0479 eV (205.00 nm)	0.0087
S_{11}	MO55 → MO61 (76%) MO55 → MO63 (12%) MO60 → MO62 (12%)	6.2955 eV (196.94 nm)	0.1579
S_{12}	MO57 → MO63 (17%) MO59 → MO62 (12%) MO59 → MO64 (26%) MO60 → MO63 (9%) MO60 → MO67 (13%) MO60 → MO70 (23%)	6.3969 eV (193.82 nm)	0.0252
S_{13}	MO59 → MO63 (12%) MO60 → MO66 (33%) MO60 → MO67 (23%) MO60 → MO69 (21%) MO60 → MO73 (11%)	6.5371 eV (189.66 nm)	0.3079

S_{14}	MO58 → MO62 (30%) MO59 → MO62 (11%) MO59 → MO64 (19%) MO60 → MO66 (14%) MO60 → MO67 (10%) MO60 → MO69 (7%) MO60 → MO71 (9%)	6.5457 eV (189.41 nm)	0.9240
S_{15}	MO55 → MO61 (11%) MO57 → MO62 (31%) MO58 → MO63 (44%) MO58 → MO70 (14%)	6.6190 eV (187.32 nm)	0.0116
S_{16}	MO60 → MO67 (79%) MO60 → MO69 (21%)	6.6614 eV (186.12 nm)	0.0174
S_{17}	MO59 → MO61 (17%) MO59 → MO63 (50%) MO59 → MO64 (21%) MO60 → MO65 (12%)	6.7340 eV (184.12 nm)	0.2258
S_{18}	MO49 → MO61 (24%) MO50 → MO61 (14%) MO51 → MO61 (38%) MO53 → MO61 (24%)	6.7488 eV (183.71 nm)	0.0035
S_{19}	MO59 → MO62 (23%) MO60 → MO69 (36%) MO60 → MO71 (12%) MO60 → MO73 (29%)	6.9230 eV (179.09 nm)	0.0039
S_{20}	MO59 → MO62 (80%) MO60 → MO67 (20%)	6.9666 eV (177.97 nm)	0.0145
S_{21}	MO60 → MO68 (24%) MO60 → MO70 (26%) MO60 → MO71 (36%) MO60 → MO74 (14%)	6.9926 eV (177.31 nm)	0.0106
S_{22}	MO52 → MO61 (11%) MO59 → MO64 (23%) MO59 → MO65 (15%) MO60 → MO71 (32%) MO60 → MO72 (10%) MO60 → MO74 (9%)	7.0277 eV (176.42 nm)	0.0258
S_{23}	MO57 → MO62 (15%) MO59 → MO64 (15%) MO60 → MO68 (44%) MO60 → MO69 (26%)	7.1403 eV (173.64 nm)	0.0195
S_{24}	MO58 → MO63 (15%) MO59 → MO62 (10%) MO59 → MO65 (20%) MO60 → MO65 (8%) MO60 → MO68 (19%) MO60 → MO69 (18%) MO60 → MO72 (10%)	7.1663 eV (173.01 nm)	0.0976

S_{25}	MO57 → MO62 (38%) MO57 → MO64 (9%) MO59 → MO62 (12%) MO59 → MO63 (9%) MO59 → MO65 (19%) MO60 → MO70 (13%)	7.1914 eV (172.41 nm)	0.2426
S_{26}	MO57 → MO63 (29%) MO58 → MO65 (11%) MO60 → MO69 (18%) MO60 → MO71 (15%) MO60 → MO72 (27%)	7.2534 eV (170.93 nm)	0.0329
S_{27}	MO49 → MO61 (9%) MO53 → MO63 (8%) MO57 → MO63 (29%) MO58 → MO66 (11%) MO59 → MO65 (12%) MO60 → MO68 (12%) MO60 → MO73 (9%) MO60 → MO74 (10%)	7.2836 eV (170.22 nm)	0.0237
S_{28}	MO50 → MO61 (40%) MO53 → MO61 (16%) MO60 → MO72 (29%) MO60 → MO73 (15%)	7.3330 eV (169.08 nm)	0.0205
S_{29}	MO45 → MO61 (12%) MO50 → MO61 (45%) MO57 → MO64 (30%) MO58 → MO66 (13%)	7.3584 eV (168.49 nm)	0.0053
S_{30}	MO50 → MO61 (17%) MO57 → MO63 (9%) MO58 → MO64 (25%) MO58 → MO65 (28%) MO60 → MO68 (8%) MO60 → MO74 (13%)	7.3642 eV (168.36 nm)	0.0030

HOMO: MO60, LUMO: MO61

Table S11 Calculated Excited States of **4** in CH_2Cl_2.

Excited State	Transition	Energy (Wavelength)	Oscillator Strength
S_1	MO62 → MO63	4.3096 eV (287.69 nm)	0.3115
S_2	MO61 → MO63 (31%) MO62 → MO64 (49%) MO62 → MO65 (9%) MO62 → MO66 (11%)	4.4904 eV (276.11 nm)	0.0577
S_3	MO59 → MO65 (12%) MO60 → MO65 (17%) MO62 → MO65 (59%) MO62 → MO66 (12%)	5.2097 eV (237.99 nm)	0.0769
S_4	MO59 → MO63 (18%) MO59 → MO65 (25%) MO61 → MO63 (16%) MO62 → MO66 (41%)	5.2558 eV (235.90 nm)	0.1297
S_5	MO61 → MO63 (21%) MO62 → MO66 (36%) MO62 → MO67 (33%) MO62 → MO73 (10%)	5.5372 eV (223.91 nm)	0.1314
S_6	MO58 → MO64 (18%) MO61 → MO63 (82%)	5.5944 eV (221.62 nm)	0.8041
S_7	MO60 → MO63 (59%) MO61 → MO64 (27%) MO62 → MO67 (14%)	5.8550 eV (211.76 nm)	0.0745
S_8	MO59 → MO63 (21%) MO59 → MO65 (20%) MO60 → MO65 (10%) MO62 → MO64 (9%) MO62 → MO67 (40%)	5.8904 eV (210.49 nm)	0.0218
S_9	MO62 → MO68 (18%) MO62 → MO69 (82%)	6.0824 eV (203.84 nm)	0.0133
S_{10}	MO58 → MO63 (31%) MO59 → MO64 (14%) MO59 → MO67 (15%) MO60 → MO63 (14%) MO60 → MO65 (18%) MO60 → MO67 (8%)	6.1159 eV (202.72 nm)	0.0614
S_{11}	MO62 → MO66 (17%) MO62 → MO67 (17%) MO62 → MO68 (54%) MO62 → MO77 (12%)	6.1475 eV (201.68 nm)	0.0096
S_{12}	MO61 → MO64 (67%) MO61 → MO66 (16%) MO62 → MO68 (17%)	6.2123 eV (199.58 nm)	0.1994
S_{13}	MO58 → MO64 (0%)	6.2642 eV (197.93 nm)	0.2196

S_{14}	MO58 → MO63 (35%) MO59 → MO65 (14%) MO60 → MO63 (10%) MO61 → MO64 (19%) MO62 → MO72 (9%) MO62 → MO74 (13%)	6.3007 eV (196.78 nm)	0.2646
S_{15}	MO59 → MO63 (8%) MO59 → MO64 (6%) MO59 → MO67 (6%) MO60 → MO64 (8%) MO60 → MO65 (9%) MO61 → MO64 (10%) MO62 → MO71 (16%) MO62 → MO72 (12%) MO62 → MO73 (13%) MO62 → MO74 (12%)	6.3845 eV (194.20 nm)	0.0437
S_{16}	MO62 → MO66 (22%) MO62 → MO72 (78%)	6.4250 eV (192.97 nm)	0.0138
S_{17}	MO58 → MO63 (17%) MO59 → MO63 (45%) MO60 → MO63 (17%) MO60 → MO64 (11%) MO60 → MO67 (10%)	6.4720 eV (191.57 nm)	0.0557
S_{18}	MO60 → MO63 (29%) MO62 → MO71 (71%)	6.5566 eV (189.10 nm)	0.0534
S_{19}	MO60 → MO66 (12%) MO60 → MO67 (10%) MO60 → MO68 (11%) MO62 → MO66 (7%) MO62 → MO70 (27%) MO62 → MO71 (12%) MO62 → MO72 (8%) MO62 → MO75 (13%)	6.6002 eV (187.85 nm)	0.0041
S_{20}	MO59 → MO64 (12%) MO61 → MO65 (18%) MO61 → MO66 (21%) MO61 → MO67 (10%) MO62 → MO74 (16%) MO62 → MO75 (12%) MO62 → MO77 (11%)	6.6971 eV (185.13 nm)	0.0210
S_{21}	MO58 → MO64 (11%) MO59 → MO65 (25%) MO60 → MO64 (32%) MO61 → MO66 (32%)	6.7572 eV (183.48 nm)	0.2207

S_{22}	MO59 → MO66 (7%) MO60 → MO65 (12%) MO61 → MO65 (16%) MO61 → MO66 (19%) MO61 → MO67 (8%) MO61 → MO76 (7%) MO62 → MO70 (7%) MO62 → MO72 (11%) MO62 → MO73 (13%)	6.7868 eV (182.68 nm)	0.1650
S_{23}	MO59 → MO66 (25%) MO59 → MO68 (15%) MO60 → MO65 (30%) MO60 → MO67 (9%) MO60 → MO68 (10%) MO62 → MO75 (11%)	6.7875 eV (182.66 nm)	0.2550
S_{24}	MO59 → MO64 (10%) MO59 → MO65 (10%) MO59 → MO66 (12%) MO59 → MO67 (23%) MO59 → MO68 (16%) MO60 → MO66 (9%) MO60 → MO67 (13%) MO60 → MO68 (7%)	6.8858 eV (180.06 nm)	0.2286
S_{25}	MO59 → MO65 (29%) MO60 → MO67 (20%) MO61 → MO65 (28%) MO62 → MO75 (23%)	6.9126 eV (179.36 nm)	0.2681
S_{26}	MO58 → MO64 (18%) MO58 → MO65 (10%) MO61 → MO65 (27%) MO62 → MO70 (9%) MO62 → MO76 (10%) MO62 → MO77 (26%)	6.9674 eV (177.95 nm)	0.0569
S_{27}	MO58 → MO64 (20%) MO59 → MO67 (15%) MO60 → MO64 (15%) MO60 → MO66 (16%) MO61 → MO65 (17%) MO62 → MO75 (9%) MO62 → MO78 (8%)	6.9932 eV (177.29 nm)	0.1587
S_{28}	MO62 → MO74 (52%) MO62 → MO78 (33%) MO62 → MO79 (15%)	7.0206 eV (176.60 nm)	0.0204

S_{29}	MO60 → MO66 (20%) MO60 → MO67 (16%) MO60 → MO68 (15%) MO62 → MO76 (23%) MO62 → MO78 (9%) MO62 → MO80 (8%) MO62 → MO81 (9%)	7.0684 eV (175.41 nm)	0.0125
S_{30}	MO58 → MO64 (9%) MO59 → MO64 (30%) MO59 → MO65 (8%) MO59 → MO66 (16%) MO60 → MO64 (10%) MO60 → MO65 (7%) MO60 → MO66 (9%) MO61 → MO67 (11%)	7.1582 eV (173.21 nm)	0.0672

HOMO: MO62, LUMO: MO63

Table S12 Calculated Excited States of **5** in CH_2Cl_2.

Excited State	Transition	Energy (Wavelength)	Oscillator Strength
S_1	MO53 → MO54	3.5751 eV (346.80 nm)	0.2715
S_2	MO50 → MO54 (45%) MO50 → MO55 (17%) MO51 → MO54 (18%) MO53 → MO54 (20%)	4.0706 eV (304.59 nm)	0.0398
S_3	MO52 → MO54 (60%) MO53 → MO56 (40%)	4.4071 eV (281.33 nm)	0.0085
S_4	MO45 → MO54 (12%) MO48 → MO54 (54%) MO48 → MO55 (20%) MO53 → MO56 (14%)	4.6649 eV (265.78 nm)	0.0090
S_5	MO52 → MO56 (16%) MO53 → MO55 (84%)	4.9162 eV (252.19 nm)	0.0313
S_6	MO53 → MO56	5.1231 eV (242.01 nm)	0.3948
S_7	MO51 → MO54 (79%) MO53 → MO56 (21%)	5.3096 eV (233.51 nm)	0.0692
S_8	MO52 → MO55 (67%) MO53 → MO56 (33%)	5.9109 eV (209.75 nm)	0.4638
S_9	MO53 → MO57 (82%) MO53 → MO64 (18%)	6.1158 eV (202.73 nm)	0.0051
S_{10}	MO48 → MO54 (38%) MO49 → MO54 (62%)	6.2615 eV (198.01 nm)	0.2284
S_{11}	MO49 → MO54 (18%) MO51 → MO55 (24%) MO52 → MO56 (58%)	6.2724 eV (197.67 nm)	0.2020
S_{12}	MO51 → MO56 (45%) MO52 → MO55 (27%) MO52 → MO56 (15%) MO52 → MO61 (13%)	6.5135 eV (190.35 nm)	0.4546
S_{13}	MO52 → MO56 (22%) MO53 → MO60 (19%) MO53 → MO61 (59%)	6.5189 eV (190.19 nm)	0.0994
S_{14}	MO51 → MO57 (22%) MO53 → MO58 (78%)	6.6432 eV (186.63 nm)	0.0450
S_{15}	MO46 → MO54 (17%) MO47 → MO54 (83%)	6.7141 eV (184.66 nm)	0.0053
S_{16}	MO47 → MO54 (19%) MO51 → MO55 (29%) MO51 → MO56 (18%) MO52 → MO55 (9%) MO53 → MO61 (25%)	6.7221 eV (184.44 nm)	0.1619

S_{17}	MO51 → MO57 (11%) MO53 → MO58 (20%) MO53 → MO59 (50%) MO53 → MO60 (10%) MO53 → MO65 (9%)	6.7939 eV (182.49 nm)	0.0117
S_{18}	MO51 → MO58 (14%) MO53 → MO57 (21%) MO53 → MO60 (48%) MO53 → MO62 (17%)	6.9165 eV (179.26 nm)	0.0015
S_{19}	MO45 → MO54 (12%) MO46 → MO54 (46%) MO49 → MO54 (9%) MO50 → MO54 (11%) MO51 → MO56 (9%) MO53 → MO61 (13%)	6.9860 eV (177.48 nm)	0.0043
S_{20}	MO44 → MO54 (12%) MO45 → MO54 (38%) MO48 → MO55 (19%) MO50 → MO55 (31%)	7.0186 eV (176.65 nm)	0.0391
S_{21}	MO52 → MO57 (55%) MO52 → MO58 (21%) MO52 → MO59 (10%) MO52 → MO64 (14%)	7.1390 eV (173.67 nm)	0.0234
S_{22}	MO50 → MO55 (13%) MO53 → MO61 (13%) MO53 → MO63 (74%)	7.2555 eV (170.88 nm)	0.0020
S_{23}	MO52 → MO57 (15%) MO53 → MO61 (39%) MO53 → MO62 (46%)	7.3550 eV (168.57 nm)	0.0256
S_{24}	MO46 → MO54 (28%) MO49 → MO55 (12%) MO50 → MO55 (60%)	7.4274 eV (166.93 nm)	0.0040
S_{25}	MO51 → MO57 (18%) MO53 → MO64 (33%) MO53 → MO65 (30%) MO53 → MO69 (19%)	7.4733 eV (165.90 nm)	0.0048
S_{26}	MO51 → MO60 (16%) MO53 → MO58 (14%) MO53 → MO59 (11%) MO53 → MO60 (13%) MO53 → MO62 (10%) MO53 → MO64 (36%)	7.6111 eV (162.90 nm)	0.0090
S_{27}	MO43 → MO55 (13%) MO45 → MO54 (24%) MO48 → MO56 (19%) MO49 → MO55 (30%) MO50 → MO56 (14%)	7.6136 eV (162.85 nm)	0.0152

S_{28}	MO43 → MO54 (36%) MO45 → MO55 (15%) MO47 → MO54 (11%) MO48 → MO56 (17%) MO50 → MO56 (21%)	7.6437 eV (162.20 nm)	0.0028
S_{29}	MO51 → MO57 (10%) MO52 → MO58 (40%) MO52 → MO60 (19%) MO53 → MO62 (12%) MO53 → MO64 (19%)	7.6829 eV (161.38 nm)	0.0010
S_{30}	MO43 → MO54 (11%) MO46 → MO56 (12%) MO52 → MO60 (26%) MO52 → MO61 (39%) MO52 → MO66 (12%)	7.7234 eV (160.53 nm)	0.0276

HOMO: MO53, LUMO: MO54

Table S13 Calculated Excited States of **6** in CH_2Cl_2.

Excited State	Transition	Energy (Wavelength)	Oscillator Strength
S_1	MO38 → MO41	4.0393 eV (306.94 nm)	0.0000
S_2	MO40 → MO41	4.2588 eV (291.12 nm)	0.4543
S_3	MO36 → MO41	4.6715 eV (265.41 nm)	0.0005
S_4	MO39 → MO41	4.7706 eV (259.89 nm)	0.0081
S_5	MO37 → MO41 (12%) MO39 → MO41 (29%) MO40 → MO42 (59%)	5.6353 eV (220.02 nm)	0.1004
S_6	MO37 → MO41	6.3504 eV (195.24 nm)	0.2403
S_7	MO34 → MO41 (14%) MO39 → MO42 (44%) MO40 → MO45 (42%)	6.4733 eV (191.53 nm)	0.0707
S_8	MO40 → MO43 (61%) MO40 → MO44 (11%) MO40 → MO47 (16%) MO40 → MO50 (12%)	6.6229 eV (187.20 nm)	0.0175
S_9	MO33 → MO41 (28%) MO35 → MO41 (58%) MO35 → MO45 (14%)	6.8362 eV (181.37 nm)	0.0001
S_{10}	MO39 → MO42	6.9556 eV (178.25 nm)	0.5033
S_{11}	MO40 → MO44 (20%) MO40 → MO46 (58%) MO40 → MO47 (22%)	7.1981 eV (172.25 nm)	0.0001
S_{12}	MO34 → MO41 (79%) MO39 → MO42 (21%)	7.3166 eV (169.46 nm)	0.1199
S_{13}	MO40 → MO44	7.3221 eV (169.33 nm)	0.0077
S_{14}	MO39 → MO43 (21%) MO39 → MO44 (11%) MO40 → MO44 (16%) MO40 → MO47 (30%) MO40 → MO48 (13%) MO40 → MO50 (9%)	7.4414 eV (166.61 nm)	0.0032
S_{15}	MO34 → MO42 (13%) MO39 → MO45 (71%) MO40 → MO42 (16%)	7.4949 eV (165.42 nm)	0.3154
S_{16}	MO39 → MO43 (40%) MO39 → MO44 (11%) MO39 → MO47 (10%) MO40 → MO44 (20%) MO40 → MO46 (19%)	7.5496 eV (164.23 nm)	0.0001
S_{17}	MO40 → MO49	7.7215 eV (160.57 nm)	0.0121
S_{18}	MO38 → MO42	7.8129 eV (158.69 nm)	0.0015
S_{19}	MO29 → MO41 (12%) MO33 → MO41 (61%) MO38 → MO42 (27%)	7.9264 eV (156.42 nm)	0.0000

S_{20}	MO39 → MO46 (13%) MO40 → MO46 (15%) MO40 → MO48 (60%) MO40 → MO50 (12%)	7.9886 eV (155.20 nm)	0.0007
S_{21}	MO33 → MO42 (23%) MO35 → MO42 (46%) MO38 → MO42 (31%)	8.0244 eV (154.51 nm)	0.0083
S_{22}	MO35 → MO42 (10%) MO39 → MO44 (36%) MO39 → MO46 (40%) MO39 → MO48 (14%)	8.0863 eV (153.33 nm)	0.0396
S_{23}	MO39 → MO46 (16%) MO39 → MO47 (33%) MO39 → MO48 (10%) MO39 → MO50 (15%) MO40 → MO50 (26%)	8.1960 eV (151.27 nm)	0.0094
S_{24}	MO36 → MO42 (18%) MO39 → MO44 (28%) MO39 → MO46 (12%) MO40 → MO50 (42%)	8.2561 eV (150.17 nm)	0.0005
S_{25}	MO36 → MO42 (75%) MO38 → MO45 (12%) MO39 → MO47 (13%)	8.2687 eV (149.94 nm)	0.0000
S_{26}	MO38 → MO43 (44%) MO38 → MO44 (35%) MO38 → MO48 (21%)	8.4131 eV (147.37 nm)	0.0012
S_{27}	MO39 → MO43 (30%) MO39 → MO46 (70%)	8.4352 eV (146.98 nm)	0.0000
S_{28}	MO37 → MO42	8.4458 eV (146.80 nm)	0.0025
S_{29}	MO31 → MO41 (20%) MO38 → MO41 (14%) MO38 → MO45 (56%) MO38 → MO49 (10%)	8.5198 eV (145.52 nm)	0.0002
S_{30}	MO34 → MO42 (37%) MO38 → MO43 (12%) MO38 → MO44 (9%) MO39 → MO49 (26%) MO40 → MO52 (16%)	8.5276 eV (145.39 nm)	0.1048

HOMO: MO40, LUMO: MO41

Table S14 Molecular-Orbital Populations of **1** in CH_2Cl_2.

Molecular Orbital	Eigenvalue / Hartrees	MO Population / %			
		MeO-	-Naph-	-ph-	-NO_2
MO78 (LUMO+4)	+0.02568	0.06	71.11	28.70	0.13
MO77 (LUMO+3)	+0.01497	0.06	35.68	64.21	0.05
MO76 (LUMO+2)	+0.00554	0.05	36.14	63.76	0.05
MO75 (LUMO+1)	−0.01102	0.34	36.00	63.29	0.37
MO74 (LUMO)	−0.06378	0.13	15.30	58.31	26.26
MO73 (HOMO)	−0.26626	2.67	58.82	38.38	0.13
MO72 (HOMO−1)	−0.31307	0.17	61.60	38.22	0.01
MO71 (HOMO−2)	−0.33156	2.76	46.69	50.04	0.51
MO70 (HOMO−3)	−0.33921	0.01	65.93	33.62	0.44
MO69 (HOMO−4)	−0.34496	2.94	46.56	49.85	0.65

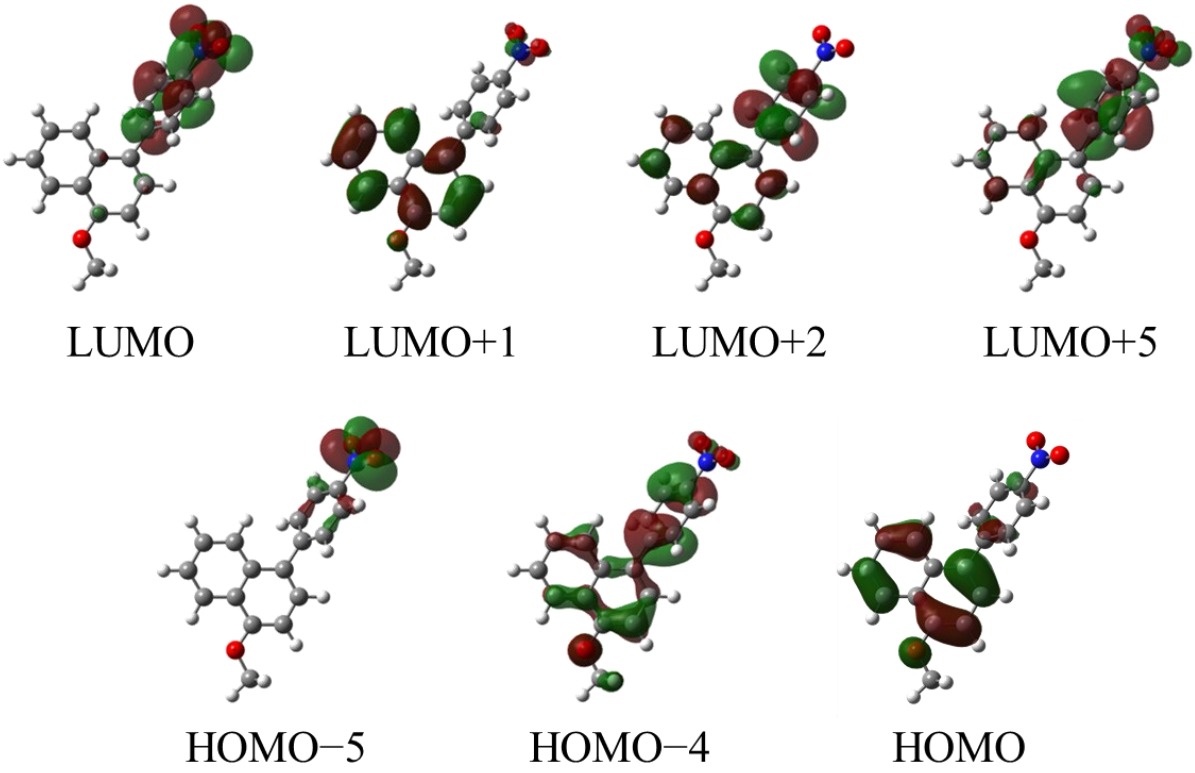

Fig. S10 Selected Kohn–Sham orbitals of **1** for ground-state optimized geometry.

Table S15 Molecular-Orbital Populations of **2** in CH_2Cl_2.

Molecular Orbital	Eigenvalue / Hartrees	MO Population / %			
			Naph-	-ph-	$-NO_2$
MO70 (LUMO+4)	+0.02639		53.33	46.15	0.52
MO69 (LUMO+3)	+0.01513		29.01	70.87	0.12
MO68 (LUMO+2)	+0.00418		23.20	76.70	0.10
MO67 (LUMO+1)	−0.01574		33.32	66.14	0.54
MO66 (LUMO)	−0.06477		8.51	65.43	26.06
MO65 (HOMO)	−0.27932		36.18	63.72	0.10
MO64 (HOMO−1)	−0.31195		65.38	34.61	0.01
MO63 (HOMO−2)	−0.33728		24.39	74.94	0.67
MO62 (HOMO−3)	−0.34057		67.62	31.92	0.46
MO61 (HOMO−4)	−0.36557		42.83	56.17	1.00

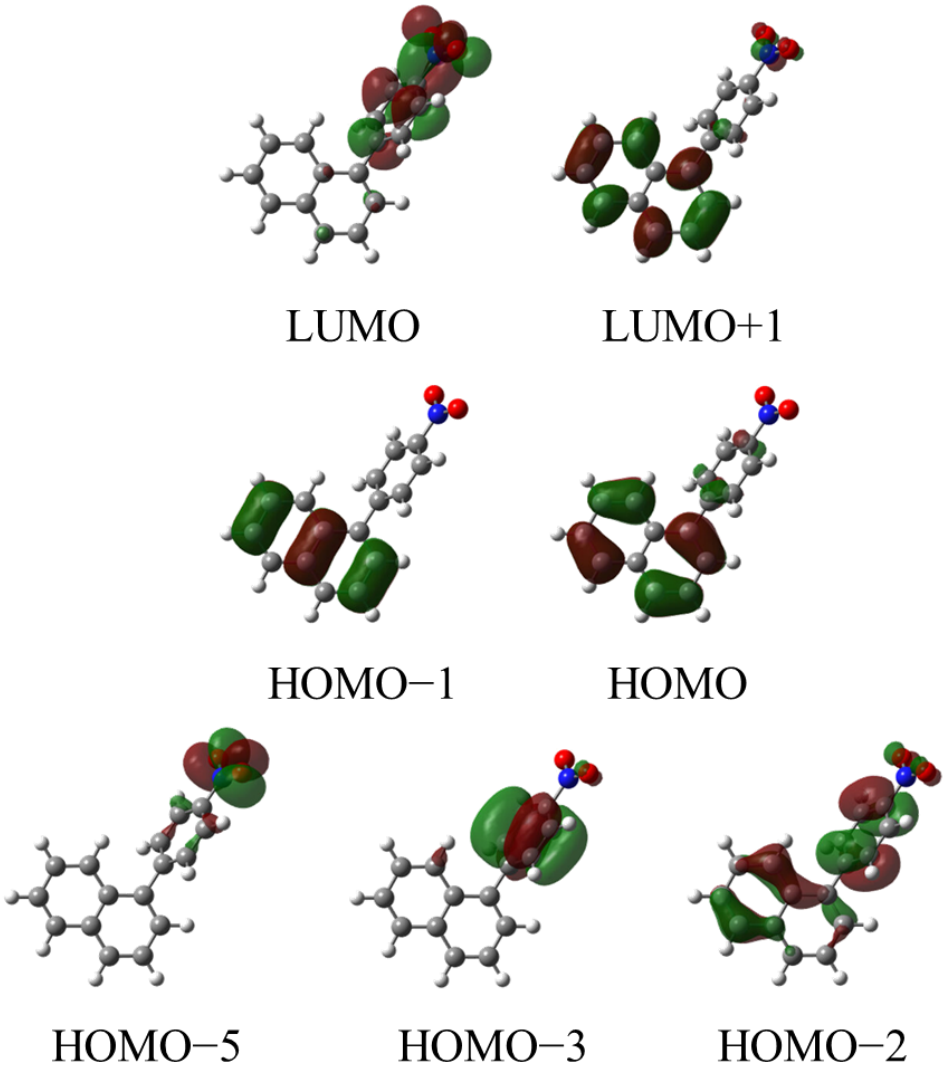

Fig. S11 Selected Kohn–Sham orbitals of **2** for ground-state optimized geometry.

Table S16 Molecular-Orbital Populations of **3** in CH_2Cl_2.

Molecular Orbital	Eigenvalue / Hartrees	MO Population / %			
		MeO-	-ph-	-ph-	$-NO_2$
MO65 (LUMO+4)	+0.02769	0.16	50.45	48.42	0.97
MO64 (LUMO+3)	+0.01836	0.17	34.63	62.50	2.70
MO63 (LUMO+2)	+0.01160	0.48	26.82	70.35	2.35
MO62 (LUMO+1)	+0.00789	0.13	42.74	57.02	0.11
MO61 (LUMO)	−0.06433	0.23	41.52	33.57	24.68
MO60 (HOMO)	−0.28135	2.11	53.27	44.44	0.18
MO59 (HOMO−1)	−0.32727	0.27	80.00	19.65	0.08
MO58 (HOMO−2)	−0.33911	0.02	16.40	82.67	0.91
MO57 (HOMO−3)	−0.34809	2.15	53.75	43.41	0.69
MO56 (HOMO−4)	−0.38268	0.04	5.56	80.37	14.03

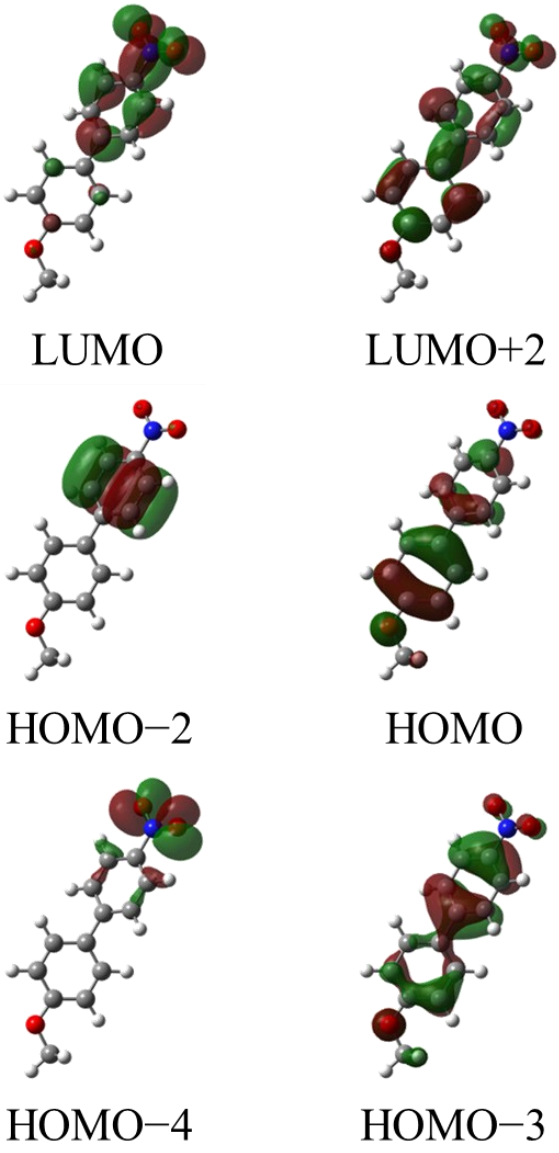

Fig. S12 Selected Kohn–Sham orbitals of **3** for ground-state optimized geometry.

Table S17 Molecular-Orbital Populations of **4** in CH_2Cl_2.

Molecular Orbital	Eigenvalue / Hartrees	MO Population / %			
		MeO-	-Naph-	-ph	
MO67 (LUMO+4)	+0.02948	0.07	78.36	21.57	
MO66 (LUMO+3)	+0.02618	0.06	62.81	37.13	
MO65 (LUMO+2)	+0.02162	0.07	58.09	41.84	
MO64 (LUMO+1)	+0.01312	0.07	46.35	53.58	
MO63 (LUMO)	−0.00832	0.50	52.02	47.48	
MO62 (HOMO)	−0.26070	3.71	68.16	28.13	
MO61 (HOMO−1)	−0.30866	0.17	75.89	23.94	
MO60 (HOMO−2)	−0.31492	0.04	54.23	45.73	
MO59 (HOMO−3)	−0.31794	0.31	52.55	47.14	
MO58 (HOMO−4)	−0.33416	16.43	52.18	31.39	

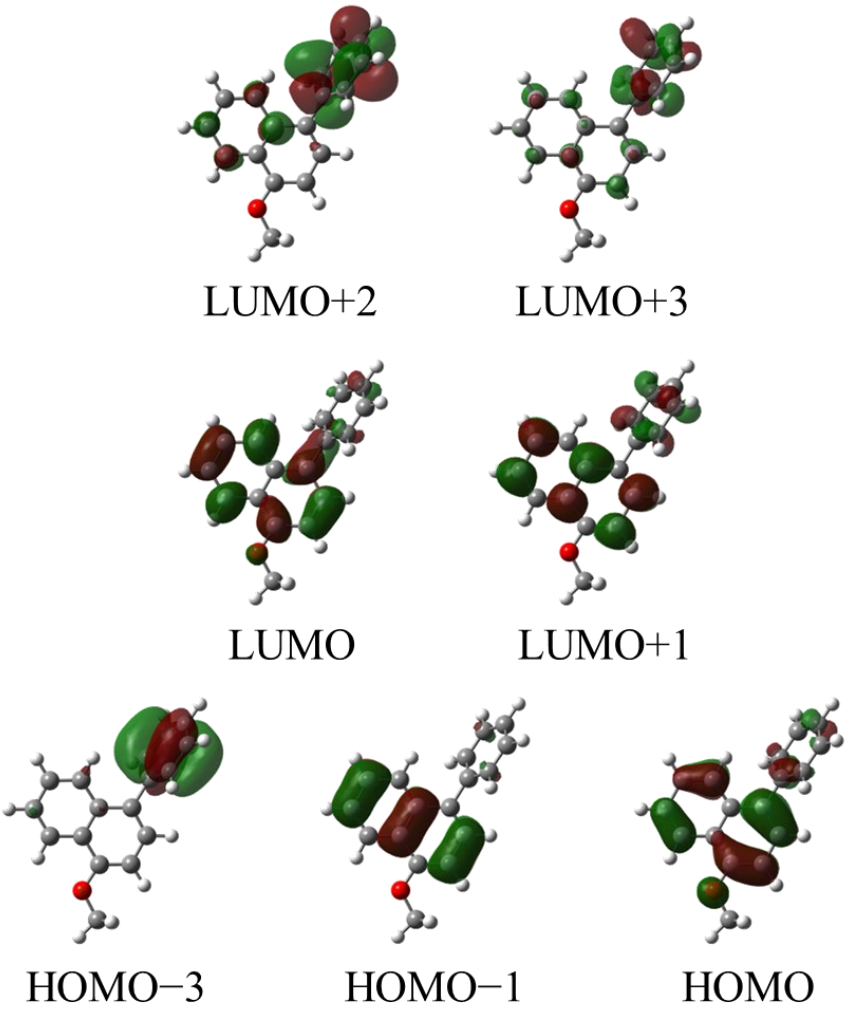

Fig. S13 Selected Kohn–Sham orbitals of **4** for ground-state optimized geometry.

Table S18 Molecular-Orbital Populations of **5** in CH_2Cl_2.

Molecular Orbital	Eigenvalue / Hartrees	MO Population / %			
		MeO-	-Naph-		$-NO_2$
MO58 (LUMO+4)	+0.03934	0.10	99.52		0.38
MO57 (LUMO+3)	+0.02949	0.20	99.18		0.62
MO56 (LUMO+2)	+0.00391	0.15	97.91		1.94
MO55 (LUMO+1)	−0.00559	0.08	99.08		0.84
MO54 (LUMO)	−0.05839	0.15	98.21		1.64
MO53 (HOMO)	−0.28471	3.91	93.44		2.65
MO52 (HOMO−1)	−0.32434	1.15	97.38		1.47
MO51 (HOMO−2)	−0.34314	11.71	84.80		3.49
MO50 (HOMO−3)	−0.37653	2.73	50.90		46.37
MO49 (HOMO−4)	−0.38998	0.11	92.08		7.81

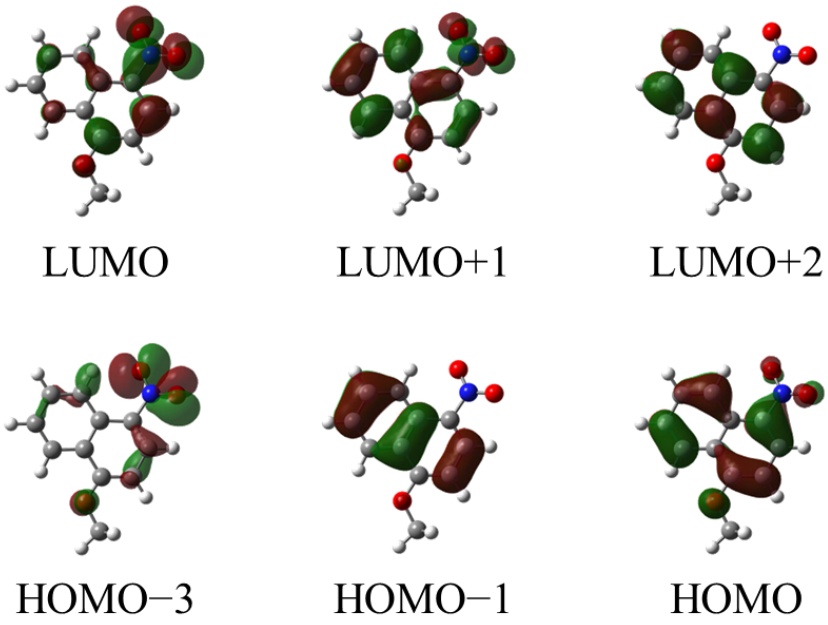

Fig. S14 Selected Kohn–Sham orbitals of **5** for ground-state optimized geometry.

Table S19 Molecular-Orbital Populations of **6** in CH_2Cl_2.

Molecular Orbital	Eigenvalue / Hartrees	MO Population / %			
		MeO-		-ph-	$-NO_2$
MO45 (LUMO+4)	+0.04582	4.16		81.29	14.55
MO44 (LUMO+3)	+0.04469	1.20		97.45	1.35
MO43 (LUMO+2)	+0.02999	1.52		96.60	1.88
MO42 (LUMO+1)	+0.00844	0.78		98.95	0.27
MO41 (LUMO)	−0.05793	3.24		31.24	65.52
MO40 (HOMO)	−0.30894	32.28		61.59	6.13
MO39 (HOMO−1)	−0.34262	0.20		98.27	1.53
MO38 (HOMO−2)	−0.37964	0.08		85.47	14.45
MO37 (HOMO−3)	−0.39461	0.02		0.88	99.10
MO36 (HOMO−4)	−0.40037	0.85		47.46	51.69

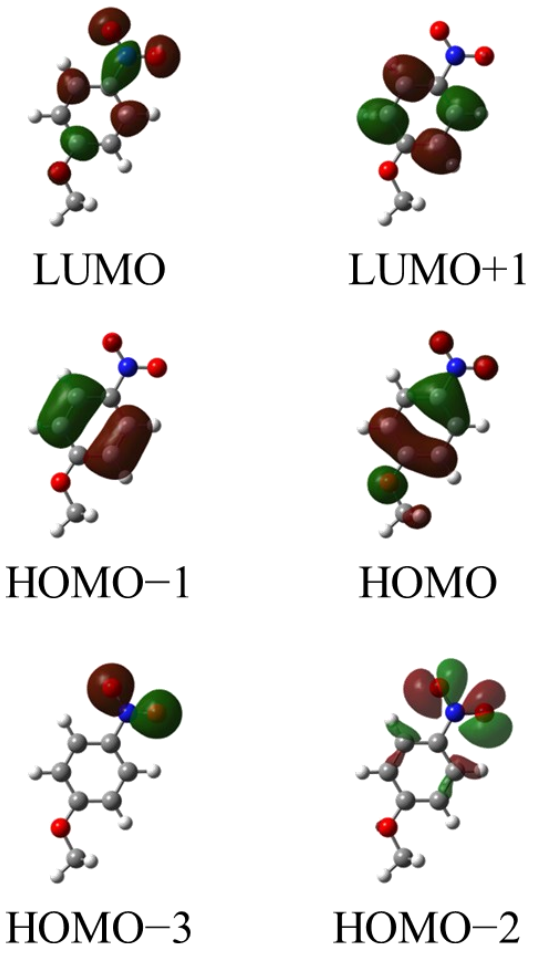

Fig. S15 Selected Kohn–Sham orbitals of **6** for ground-state optimized geometry.

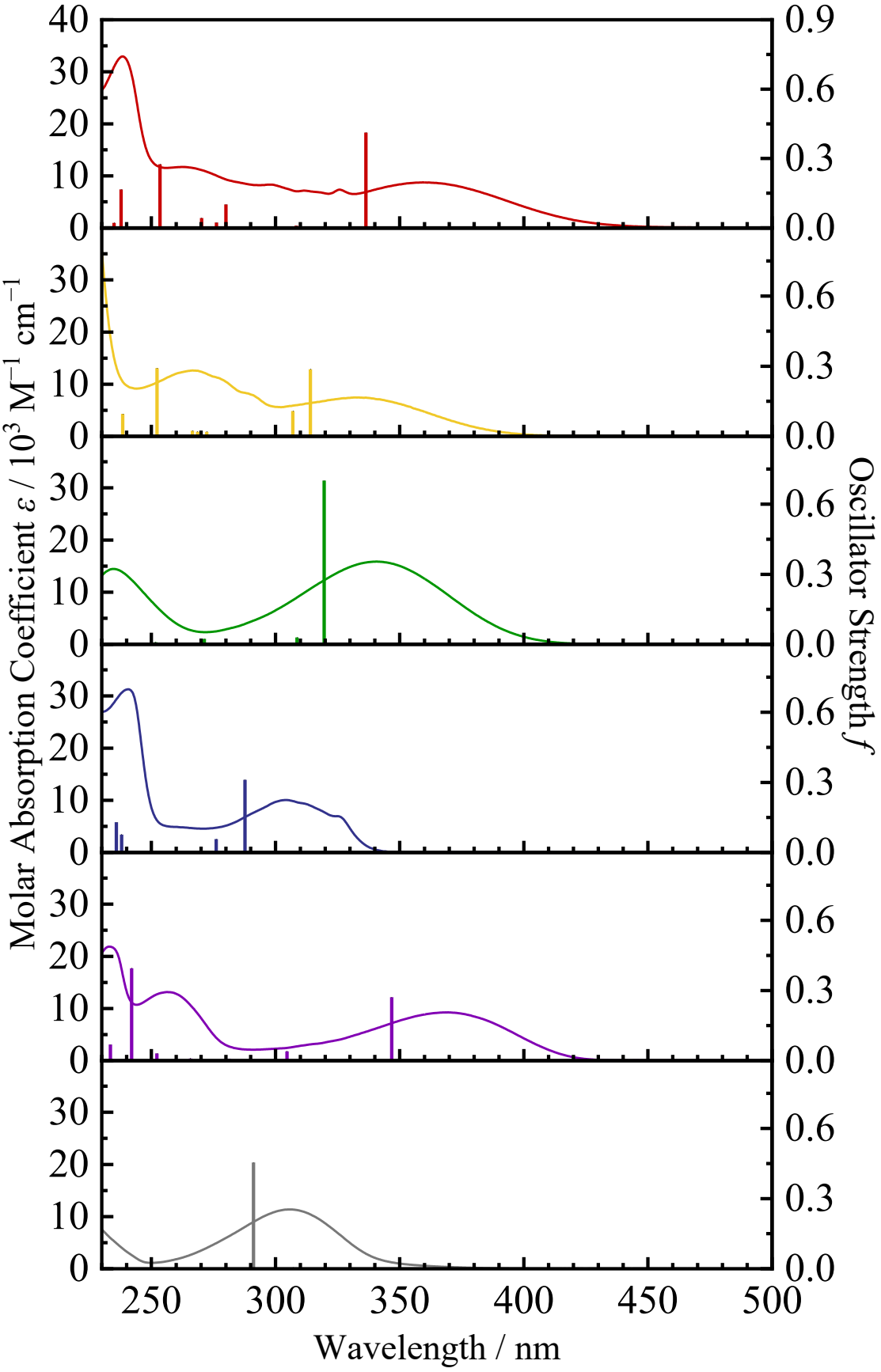

Fig. S16 Comparisons of absorption spectra and oscillator strengths calculated by TD-DFT (perpendicular bars) of **1**–**6** in dichloromethane. The colors correspond to those indicated in Fig. 2.

Table S20 Calculated Excited States of S_1-Optimized Geometry of **1** in CH_2Cl_2.

Excited State	Transition	Energy (Wavelength)	Oscillator Strength
T_1	MO69 → MO74 (14%) MO71 → MO74 (19%) MO73 → MO74 (67%)	1.6932 eV (732.25 nm)	0.0000
T_2	MO67 → MO74 (64%) MO67 → MO75 (20%) MO67 → MO78 (16%)	2.4490 eV (506.26 nm)	0.0000
T_3	MO69 → MO74 (18%) MO71 → MO74 (22%) MO73 → MO74 (17%) MO73 → MO75 (43%)	2.5845 eV (479.73 nm)	0.0000
S_1	MO73 → MO74	2.7957 eV (443.48 nm)	0.8337
S_2	MO68 → MO74 (68%) MO68 → MO75 (19%) MO68 → MO78 (13%)	3.5187 eV (352.36 nm)	0.0026
S_3	MO73 → MO75 (86%) MO73 → MO76 (14%)	4.0276 eV (307.84 nm)	0.0304

HOMO: MO73, LUMO: MO74

Table S21 Calculated Excited States of S_1-Optimized Geometry of **2** in CH_2Cl_2.

Excited State	Transition	Energy (Wavelength)	Oscillator Strength
T_1	MO63 → MO66 (16%) MO65 → MO66 (51%) MO65 → MO67 (33%)	1.6564 eV (748.50 nm)	0.0000
T_2	MO59 → MO66 (82%) MO62 → MO66 (18%)	2.4303 eV (510.15 nm)	0.0000
T_3	MO60 → MO66 (27%) MO65 → MO67 (73%)	2.6298 eV (471.46 nm)	0.0000
S_1	MO63 → MO66 (12%) MO65 → MO66 (75%) MO65 → MO67 (13%)	2.9558 eV (419.47 nm)	0.7398
S_2	MO60 → MO67 (22%) MO61 → MO66 (78%)	3.5322 eV (351.01 nm)	0.0021
S_3	MO65 → MO67 (64%) MO65 → MO68 (36%)	4.1488 eV (298.84 nm)	0.0075

HOMO: MO65, LUMO: MO66

Table S22 Calculated Excited States of S_1-Optimized Geometry of **3** in CH_2Cl_2.

Excited State	Transition	Energy (Wavelength)	Oscillator Strength
T_1	MO60 → MO61	1.8698 eV (663.08 nm)	0.0000
T_2	MO55 → MO61 (71%) MO55 → MO62 (29%)	2.4771 eV (500.51 nm)	0.0000
T_3	MO56 → MO61 (73%) MO56 → MO62 (27%)	3.0396 eV (407.90 nm)	0.0000
S_1	MO60 → MO61	3.0787 eV (402.72 nm)	1.0509
S_2	MO56 → MO61 (74%) MO56 → MO62 (26%)	3.5500 eV (349.25 nm)	0.0049
S_3	MO58 → MO61 (71%) MO60 → MO63 (29%)	4.2556 eV (291.34 nm)	0.0218

HOMO: MO60, LUMO: MO61

Table S23 Calculated Excited States of S_1-Optimized Geometry of **4** in CH_2Cl_2.

Excited State	Transition	Energy (Wavelength)	Oscillator Strength
T_1	MO62 → MO63	1.5033 eV (824.76 nm)	0.0000
T_2	MO59 → MO63 (19%) MO62 → MO65 (81%)	3.0725 eV (403.53 nm)	0.0000
S_1	MO62 → MO63	3.2152 eV (385.62 nm)	0.5058
T_3	MO62 → MO64 (69%) MO62 → MO65 (31%)	3.5198 eV (352.25 nm)	0.0000
S_2	MO61 → MO63 (27%) MO62 → MO64 (57%) MO62 → MO65 (16%)	4.0525 eV (305.94 nm)	0.0635
S_3	MO62 → MO65	4.4379 eV (279.37 nm)	0.1340

HOMO: MO62, LUMO: MO63

Table S24 Calculated Excited States of S_1-Optimized Geometry of **5** in CH_2Cl_2.

Excited State	Transition	Energy (Wavelength)	Oscillator Strength
T_1	MO46 → MO54 (23%) MO49 → MO54 (77%)	−0.4776 eV (−2596.13 nm)	0.0000
T_2	MO49 → MO54 (25%) MO50 → MO54 (28%) MO53 → MO54 (47%)	1.4900 eV (832.13 nm)	0.0000
S_1	MO50 → MO54 (26%) MO53 → MO54 (74%)	1.6543 eV (749.28 nm)	0.0001
T_3	MO51 → MO60 (14%) MO52 → MO56 (16%) MO53 → MO55 (70%)	2.2340 eV (554.99 nm)	0.0000
S_2	MO49 → MO54 (31%) MO50 → MO54 (69%)	3.1406 eV (393.77 nm)	0.0005
S_3	MO45 → MO54 (14%) MO47 → MO54 (30%) MO48 → MO54 (56%)	3.3822 eV (366.58 nm)	0.0016

HOMO: MO53, LUMO: MO54

Table S25 Calculated Excited States of S_1-Optimized Geometry of **6** in CH_2Cl_2.

Excited State	Transition	Energy (Wavelength)	Oscillator Strength
T_1	MO37 → MO41 (76%) MO38 → MO41 (24%)	1.6864 eV (735.21 nm)	0.0000
T_2	MO39 → MO41	1.9297 eV (642.51 nm)	0.0000
S_1	MO39 → MO41	2.4359 eV (508.98 nm)	0.0000
T_3	MO40 → MO41 (85%) MO40 → MO44 (14%)	2.5585 eV (484.59 nm)	0.0000
S_2	MO40 → MO41	3.9518 eV (313.74 nm)	0.6138
S_3	MO38 → MO41	4.3849 eV (282.76 nm)	0.0147

HOMO: MO40, LUMO: MO41

Table S26 Molecular-Orbital Populations of S_1-Optimized Geometry of **1** in CH_2Cl_2.

Molecular Orbital	Eigenvalue / Hartrees	MO Population / %			
		MeO-	-Naph-	-ph-	-NO_2
MO78 (LUMO+4)	+0.02363	0.05	76.19	23.57	0.19
MO77 (LUMO+3)	+0.01515	0.04	57.56	42.24	0.16
MO76 (LUMO+2)	+0.00954	0.05	47.21	52.68	0.06
MO75 (LUMO+1)	−0.02077	0.35	48.33	50.68	0.64
MO74 (LUMO)	−0.08139	0.45	37.91	46.49	15.15
MO73 (HOMO)	−0.25478	2.07	57.58	39.69	0.66
MO72 (HOMO−1)	−0.31880	0.16	63.30	36.52	0.02
MO71 (HOMO−2)	−0.32913	0.71	31.79	66.34	1.16
MO70 (HOMO−3)	−0.34210	0.17	63.46	35.54	0.83
MO69 (HOMO−4)	−0.34790	8.38	27.29	62.84	1.49

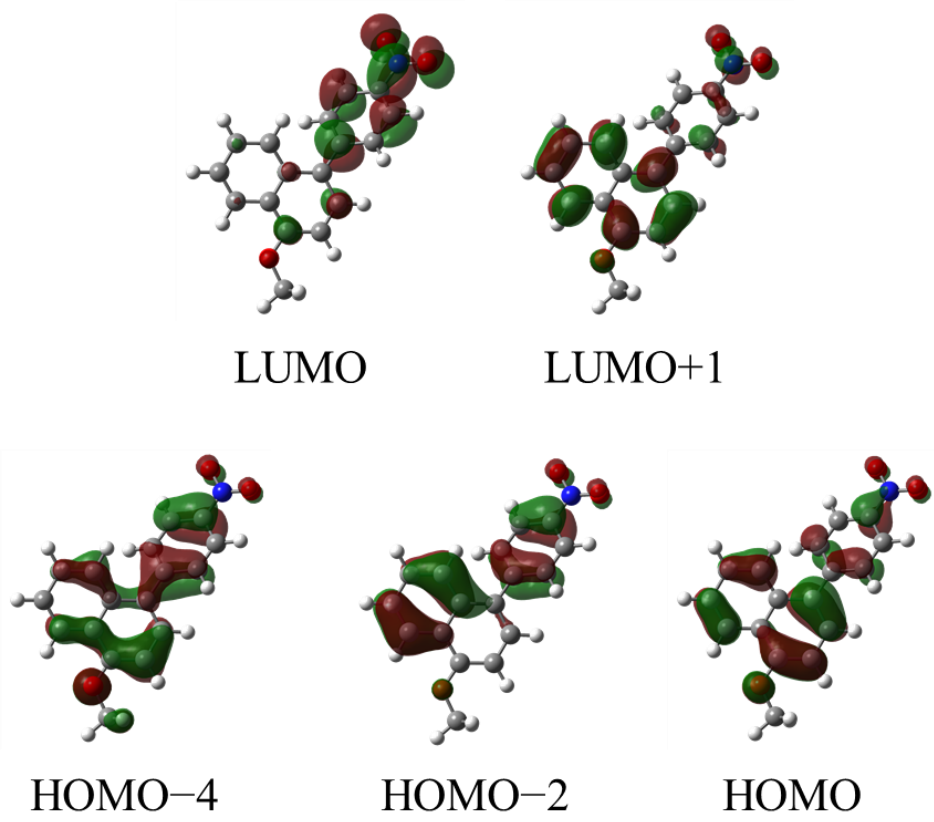

Fig. S17 Selected Kohn–Sham orbitals of **1** for S_1- optimized geometry.

Table S27 Molecular-Orbital Populations of S_1-Optimized Geometry of **2** in CH_2Cl_2.

Molecular Orbital	Eigenvalue / Hartrees	MO Population / %			
			Naph-	-ph-	-NO_2
MO70 (LUMO+4)	+0.02080		64.35	35.36	0.29
MO69 (LUMO+3)	+0.01488		52.16	47.60	0.24
MO68 (LUMO+2)	+0.00731		42.39	57.55	0.06
MO67 (LUMO+1)	−0.02748		19.15	79.77	1.08
MO66 (LUMO)	−0.08521		39.38	45.75	14.87
MO65 (HOMO)	−0.26731		57.80	41.32	0.88
MO64 (HOMO−1)	−0.31803		65.83	34.15	0.02
MO63 (HOMO−2)	−0.33240		45.78	53.24	0.98
MO62 (HOMO−3)	−0.34491		55.04	44.15	0.81
MO61 (HOMO−4)	−0.37387		14.21	79.28	6.51

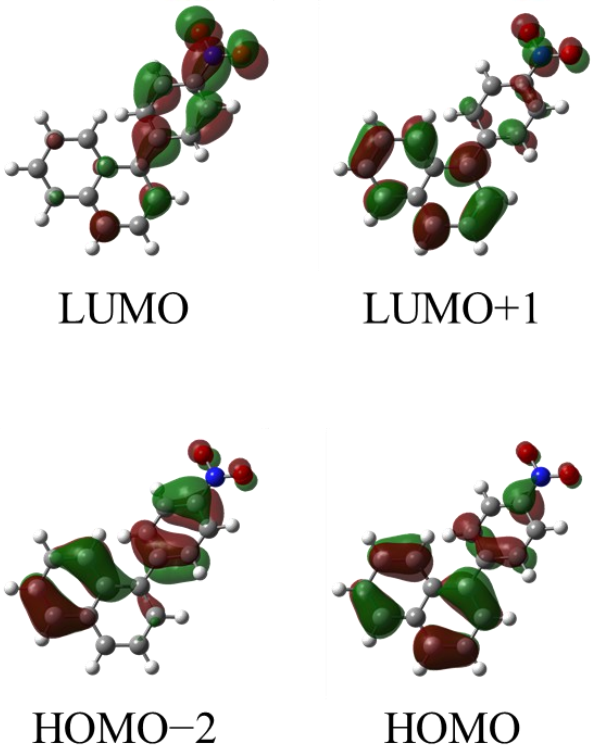

Fig. S18 Selected Kohn–Sham orbitals of **2** for S_1- optimized geometry.

Table S28 Molecular-Orbital Populations of S_1-Optimized Geometry of **3** in CH_2Cl_2.

Molecular Orbital	Eigenvalue / Hartrees	MO Population / %			
		MeO-	-ph-	-ph-	-NO_2
MO65 (LUMO+4)	+0.03120	0.14	41.67	58.11	0.08
MO64 (LUMO+3)	+0.02773	0.24	52.62	45.93	1.21
MO63 (LUMO+2)	+0.00456	0.05	52.99	46.81	0.15
MO62 (LUMO+1)	−0.00280	0.32	28.05	69.75	1.88
MO61 (LUMO)	−0.08136	0.47	25.40	59.09	15.04
MO60 (HOMO)	−0.26964	1.41	52.45	45.68	0.46
MO59 (HOMO−1)	−0.33464	0.62	75.32	23.79	0.27
MO58 (HOMO−2)	−0.34433	0.39	11.96	85.66	1.99
MO57 (HOMO−3)	−0.34949	6.95	20.09	70.58	2.38
MO56 (HOMO−4)	−0.37132	0.00	3.33	88.25	8.42

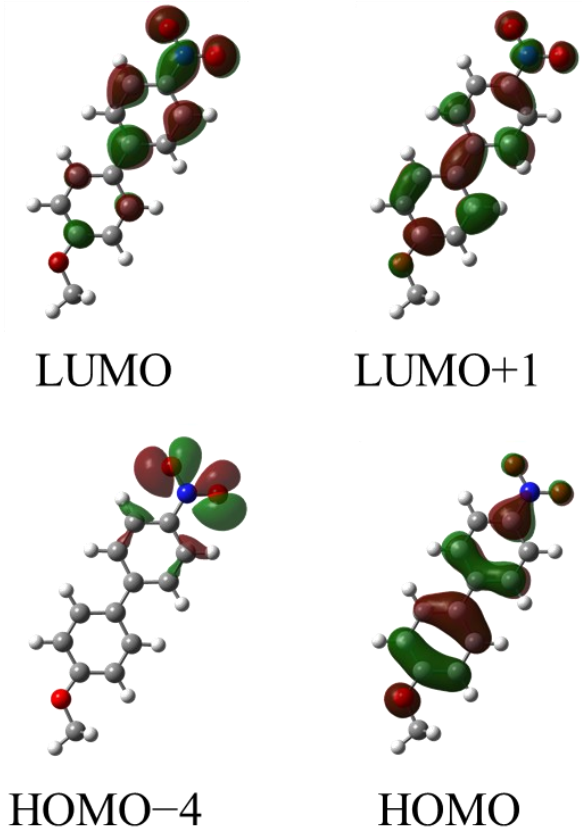

Fig. S19 Selected Kohn–Sham orbitals of **3** for S_1- optimized geometry.

Table S29 Molecular-Orbital Populations of S_1-Optimized Geometry of **4** in CH_2Cl_2.

Molecular Orbital	Eigenvalue / Hartrees	MO Population / %			
		MeO-	-Naph-	-ph	
MO67 (LUMO+4)	+0.03176	0.06	80.41	19.53	
MO66 (LUMO+3)	+0.02703	0.04	61.16	38.80	
MO65 (LUMO+2)	+0.02015	0.06	67.71	32.23	
MO64 (LUMO+1)	+0.01502	0.02	36.67	63.31	
MO63 (LUMO)	−0.03181	0.51	62.47	37.02	
MO62 (HOMO)	−0.23731	2.84	62.40	34.76	
MO61 (HOMO−1)	−0.31310	0.64	83.49	15.87	
MO60 (HOMO−2)	−0.31488	0.04	54.62	45.34	
MO59 (HOMO−3)	−0.31880	0.10	59.05	40.85	
MO58 (HOMO−4)	−0.33736	9.66	74.70	15.64	

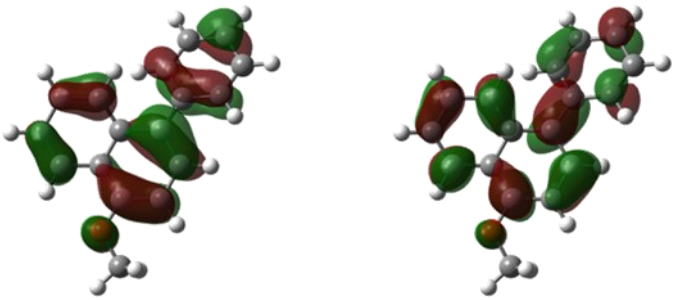

Fig. S20 Selected Kohn–Sham orbitals of **4** for S_1- optimized geometry.

Table S30 Molecular-Orbital Populations of S_1-Optimized Geometry of **5** in CH_2Cl_2.

Molecular Orbital	Eigenvalue / Hartrees	MO Population / %			
		MeO-	-Naph-		$-NO_2$
MO58 (LUMO+4)	+0.03883	0.08	99.79		0.13
MO57 (LUMO+3)	+0.03001	0.22	98.73		1.05
MO56 (LUMO+2)	+0.00710	0.25	93.75		6.00
MO55 (LUMO+1)	−0.02671	4.18	93.46		2.36
MO54 (LUMO)	−0.09588	0.01	97.68		2.31
MO53 (HOMO)	−0.27020	13.60	79.47		6.93
MO52 (HOMO−1)	−0.32572	1.30	98.25		0.45
MO51 (HOMO−2)	−0.34351	19.67	74.67		5.66
MO50 (HOMO−3)	−0.38591	12.55	21.81		65.64
MO49 (HOMO−4)	−0.39539	0.09	6.03		93.88

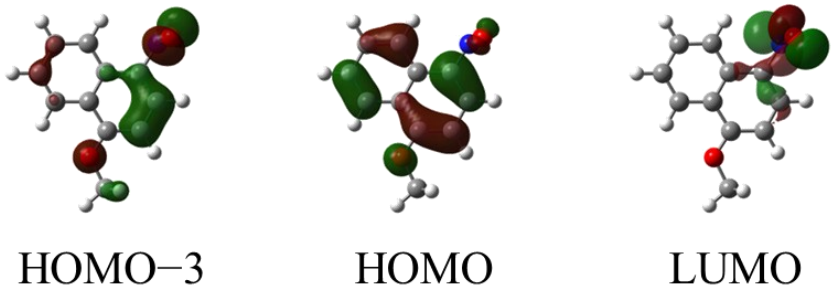

Fig. S21 Selected Kohn–Sham orbitals of **5** for S_1- optimized geometry.

Table S31 Molecular-Orbital Populations of S_1-Optimized Geometry of **6** in CH_2Cl_2.

Molecular Orbital	Eigenvalue / Hartrees	MO Population / %			
		MeO-		-ph-	$-NO_2$
MO45 (LUMO+4)	+0.04394	1.73		96.60	1.67
MO44 (LUMO+3)	+0.04198	3.67		81.68	14.65
MO43 (LUMO+2)	+0.02897	1.43		97.23	1.34
MO42 (LUMO+1)	+0.00167	0.73		99.04	0.23
MO41 (LUMO)	−0.08279	4.37		31.35	64.28
MO40 (HOMO)	−0.31818	31.46		58.00	10.54
MO39 (HOMO−1)	−0.33844	0.02		83.63	16.35
MO38 (HOMO−2)	−0.35054	0.18		91.27	8.55
MO37 (HOMO−3)	−0.36675	0.03		4.86	95.11
MO36 (HOMO−4)	−0.41089	0.47		74.33	25.20

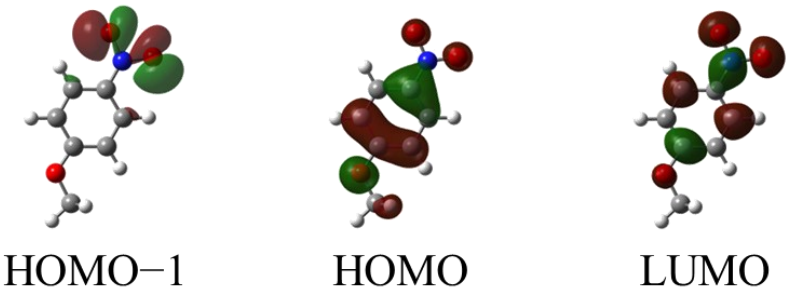

Fig. S22 Selected Kohn–Sham orbitals of **6** for S_1- optimized geometry.

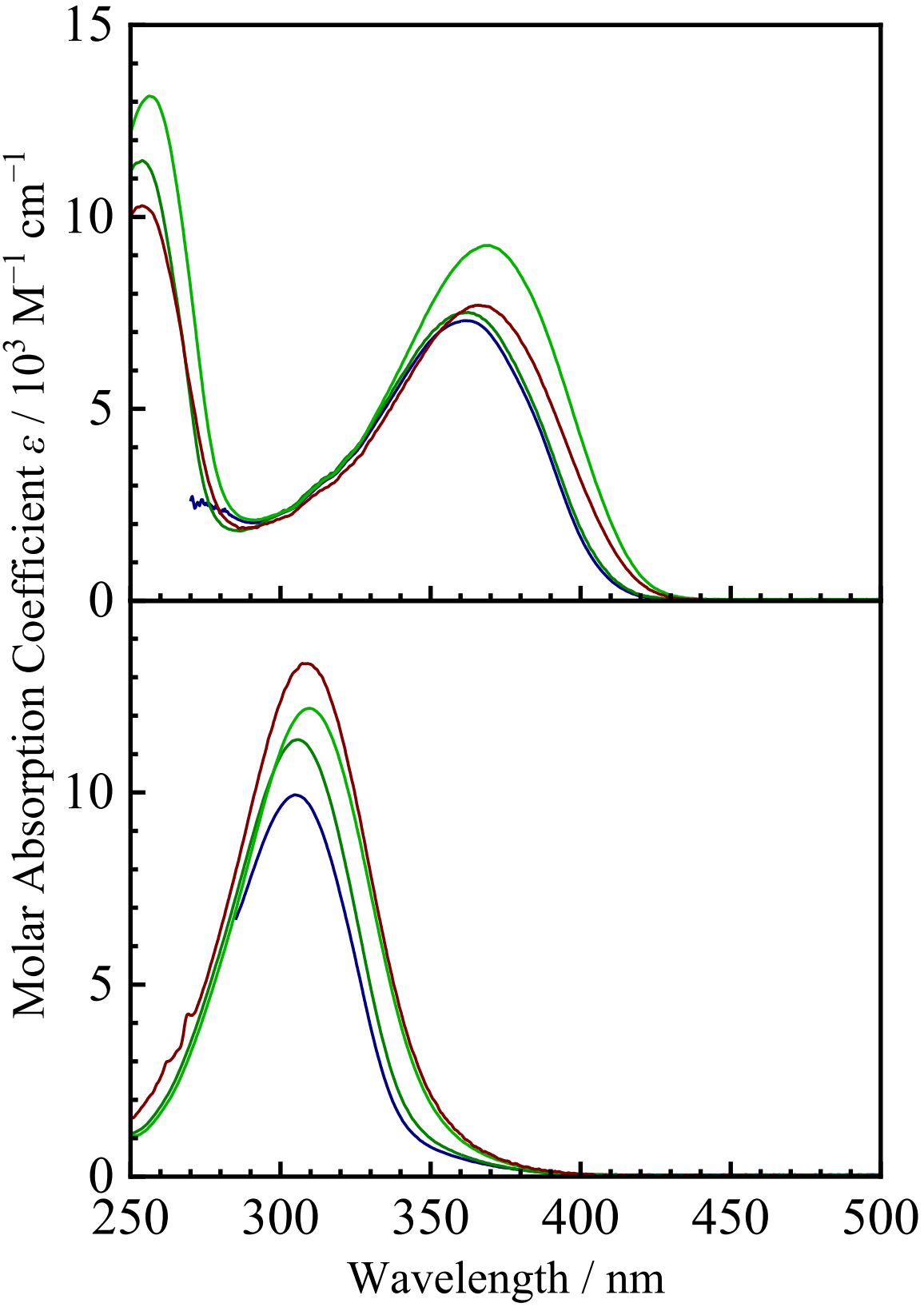

Fig. S23 Absorption spectra of **5** (top) and **6** (bottom) in various solvents. The colors correspond to those indicated in Fig. 3.

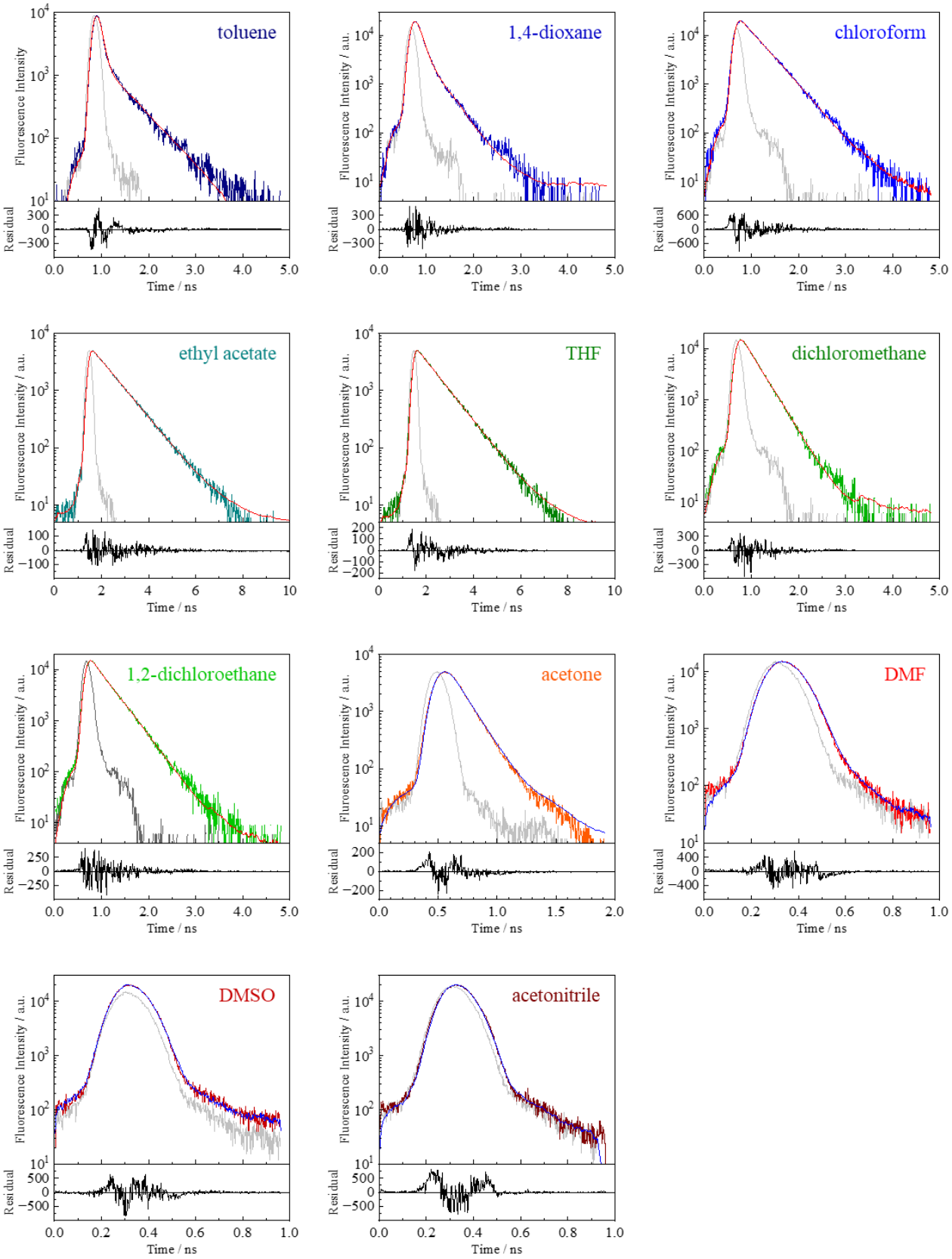

Fig. S24 Fluorescence decay profiles of **1** in various solvents. Grey and red/blue curves represent the instrumental response function and convolution fits by the single- or biexponential decay function, respectively.

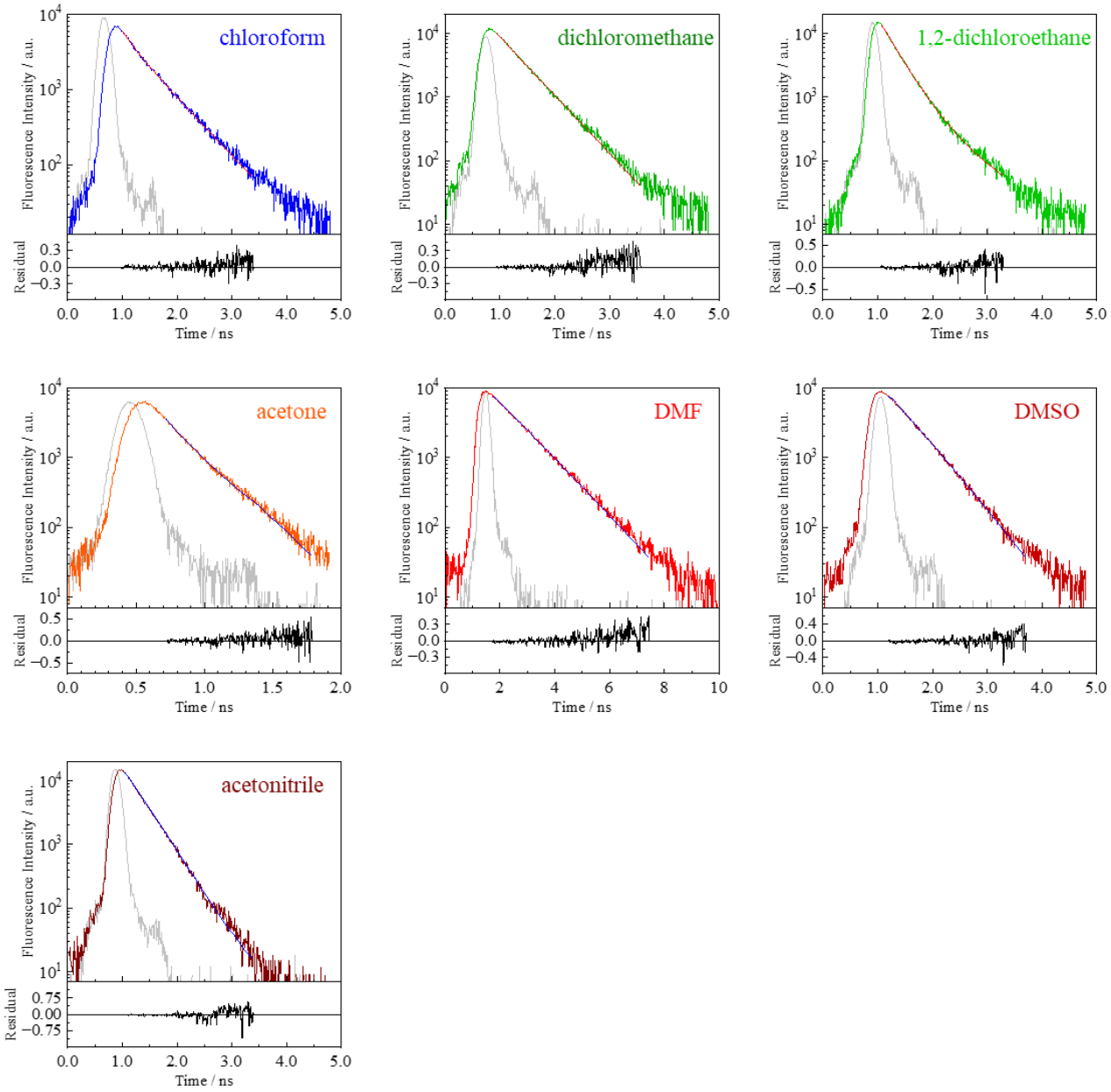

Fig. S25 Fluorescence decay profiles of **2** in various solvents. Grey and red/blue curves represent the instrumental response function and convolution fits by the single- or biexponential decay functions, respectively.

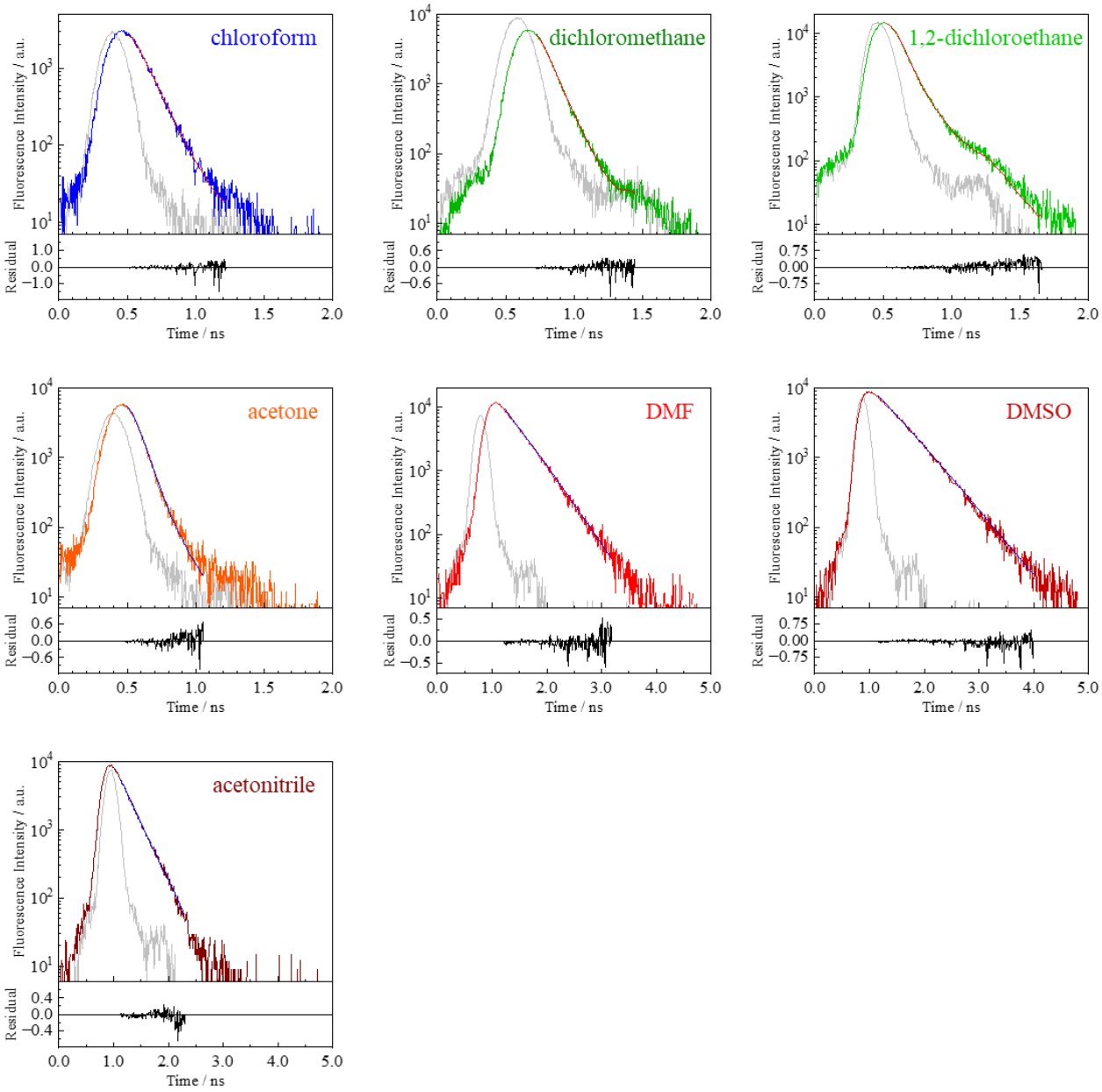

Fig. S26 Fluorescence decay profile of **3** in various solvents. Grey and red/blue curves represent the instrumental response function and convolution fits by the single- or biexponential decay function, respectively.

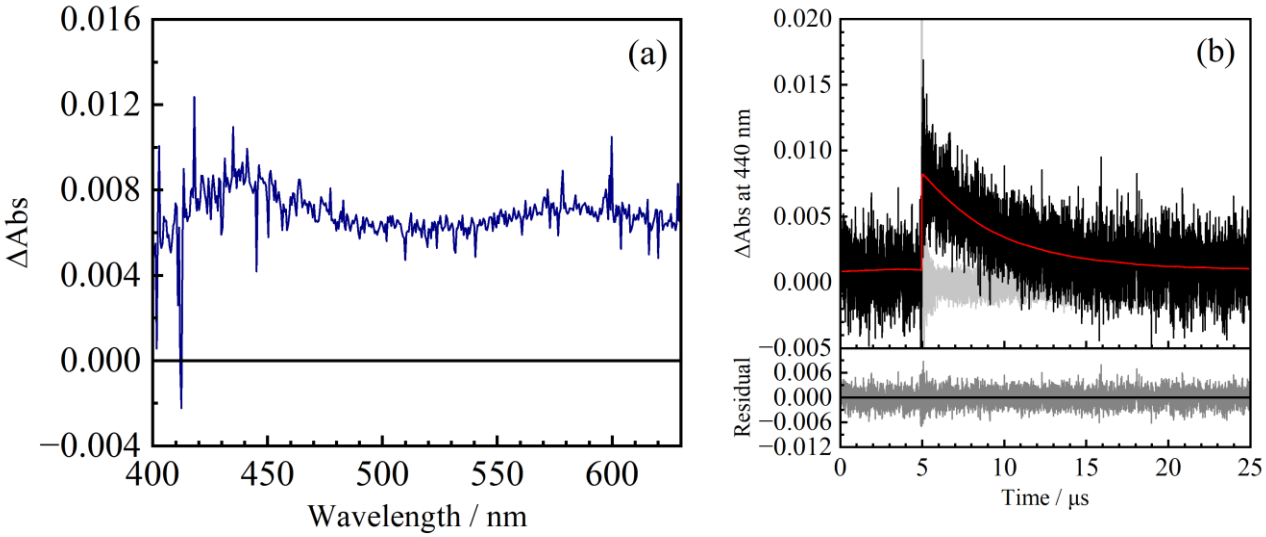

Fig. S27 Transient absorption spectra of **2** in toluene (1×10^{-4} M) at 50 ns after the 355-nm excitation (a) and transient absorption decay profile at 440 nm (b, black). Grey and red curves in (b) represent the instrumental response function and convolution fit by the single exponential decay function, respectively.

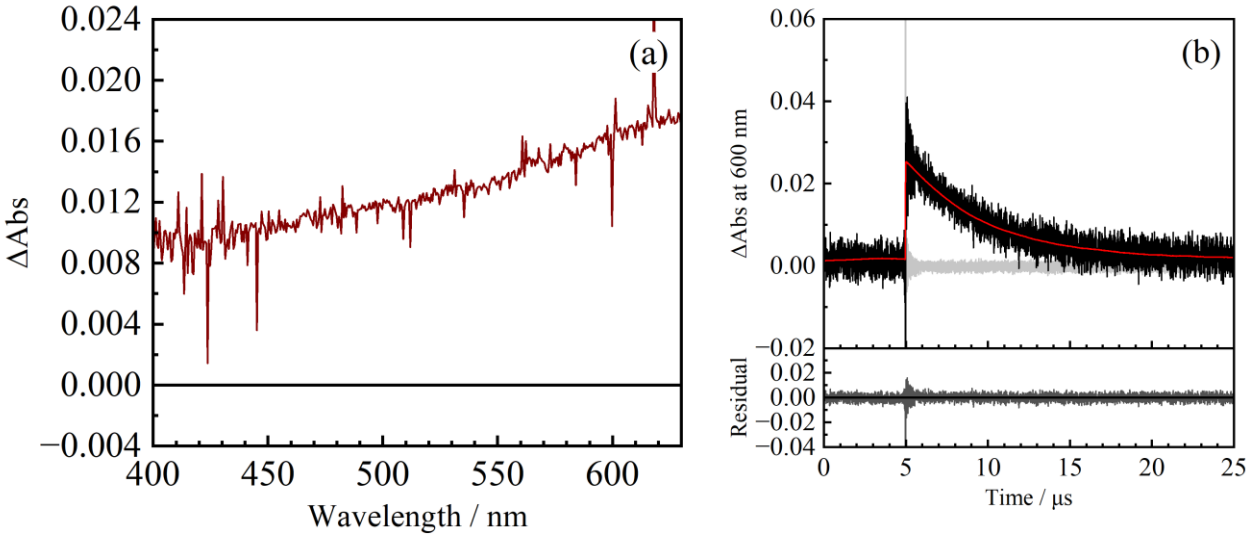

Fig. S28 Transient absorption spectra of **3** in acetonitrile (4×10^{-5} M) at 50 ns after the 355-nm excitation (a) and transient absorption decay profile at 600 nm (b, black). Grey and red curves in (b) represent the instrumental response function and convolution fit by the single exponential decay function, respectively.

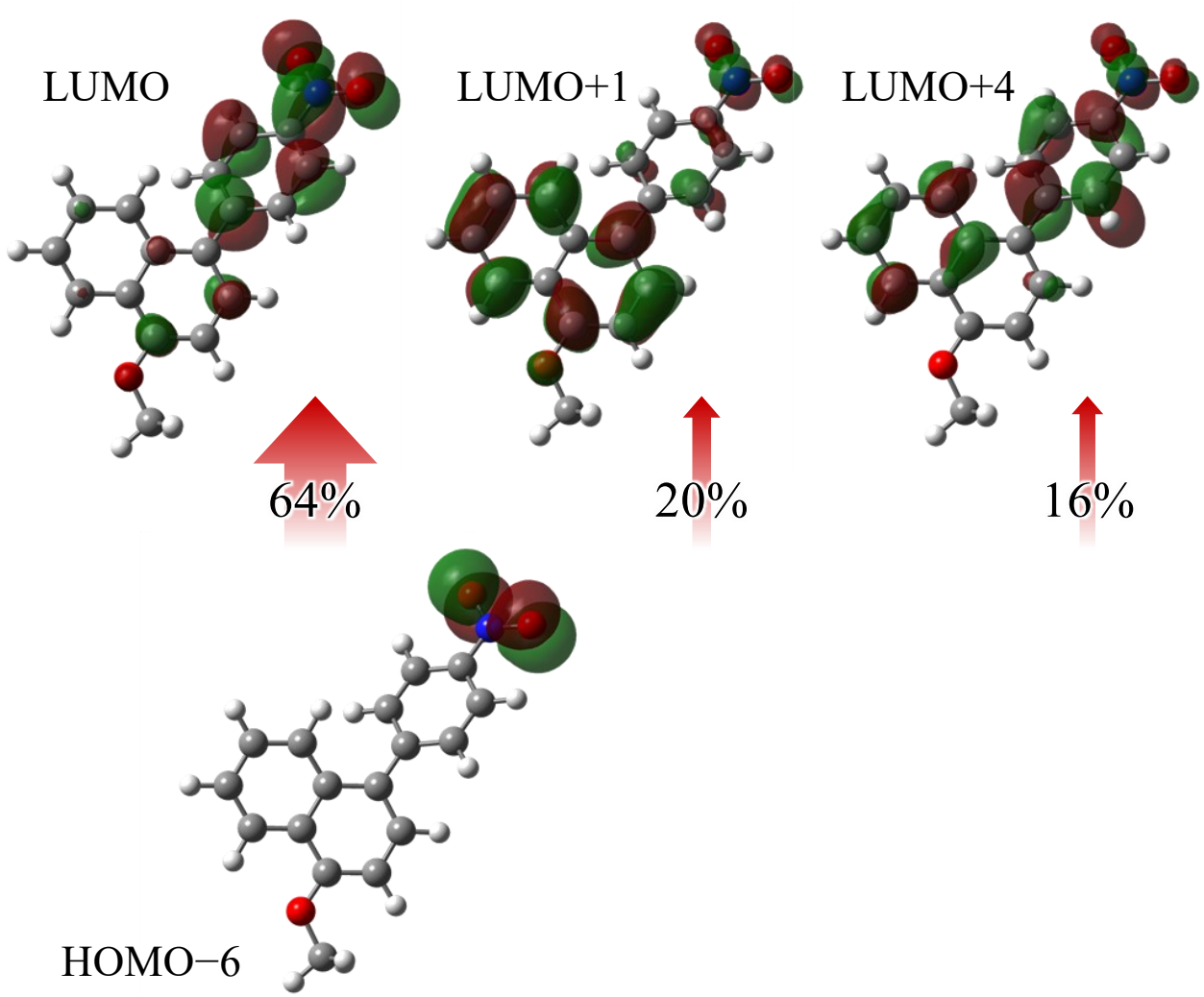

Fig. S29 Molecular-orbital distribution to give T_2 state of **1** in S_1-optimized geometry.

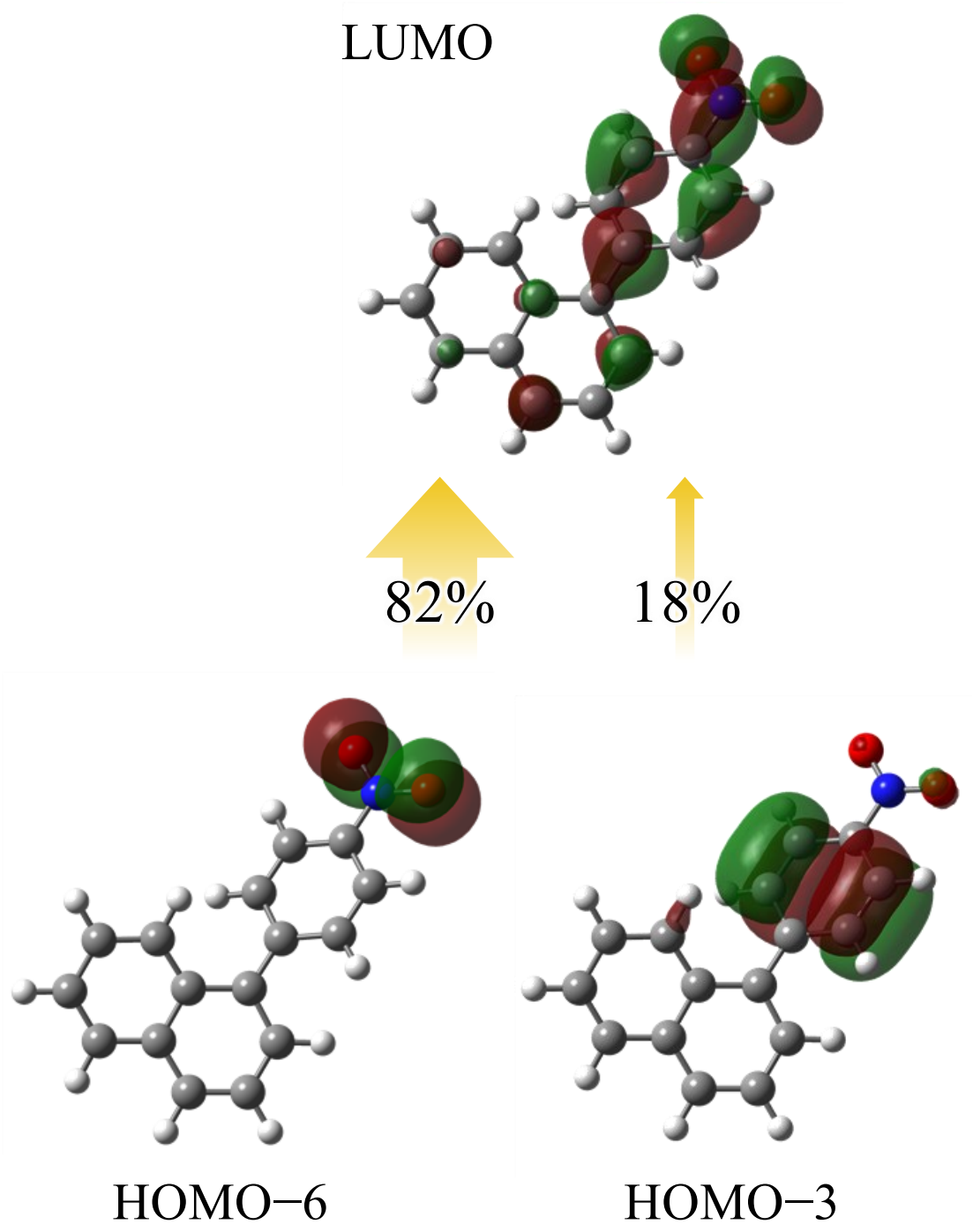

Fig. S30 Molecular-orbital distribution to give T_2 state of **2** in S_1-optimized geometry.

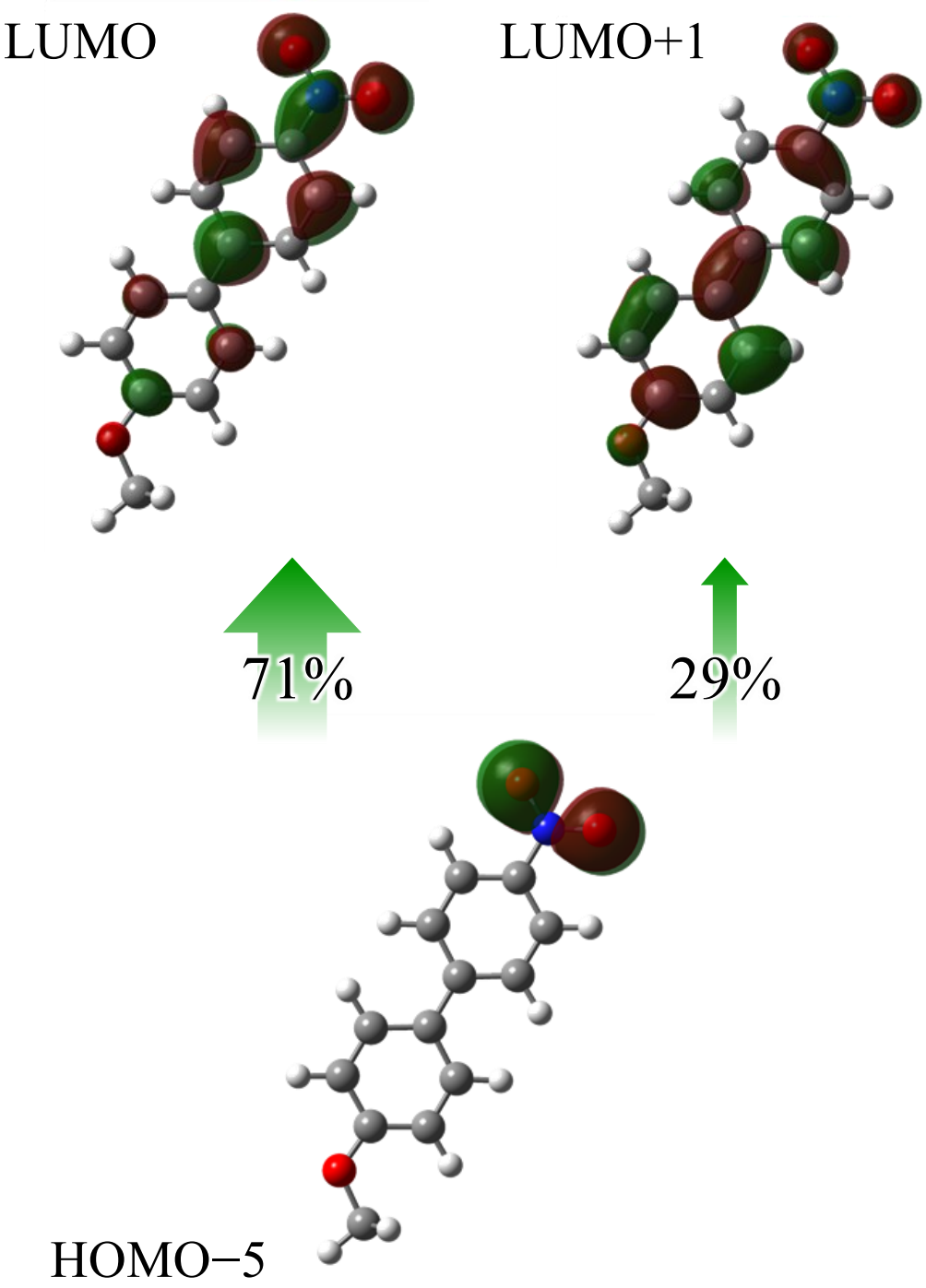

Fig. S31 Molecular-orbital distribution to give T_2 state of **3** in S_1-optimized geometry.

Table S32 Calculated Excited States of S_1-Optimized Geometry of **1** in Toluene.

Excited State	Transition	Energy (Wavelength)	Oscillator Strength
T_1	MO71 → MO74 (13%) MO73 → MO74 (51%) MO73 → MO75 (36%)	1.8120 eV (684.25 nm)	0.0000
T_2	MO67 → MO75 (61%) MO70 → MO74 (39%)	2.3997 eV (516.67 nm)	0.0000
T_3	MO73 → MO75 (80%) MO73 → MO79 (20%)	2.6594 eV (466.20 nm)	0.0000
S_1	MO73 → MO74 (85%) MO73 → MO75 (15%)	2.9589 eV (419.02 nm)	0.8028
S_2	MO68 → MO74 (82%) MO68 → MO79 (18%)	3.5641 eV (347.86 nm)	0.0044
S_3	MO73 → MO75 (86%) MO73 → MO76 (14%)	4.0674 eV (304.82 nm)	0.0284

HOMO: MO73, LUMO: MO74

Table S33 Calculated Excited States of S_1-Optimized Geometry of **1** in THF.

Excited State	Transition	Energy (Wavelength)	Oscillator Strength
T_1	MO69 → MO74 (10%) MO71 → MO74 (13%) MO73 → MO74 (48%) MO73 → MO75 (29%)	1.7052 eV (727.10 nm)	0.0000
T_2	MO67 → MO74 (86%) MO67 → MO79 (14%)	2.4442 eV (507.26 nm)	0.0000
T_3	MO73 → MO75	2.5907 eV (478.57 nm)	0.0000
S_1	MO73 → MO74 (87%) MO73 → MO75 (13%)	2.8148 eV (440.47 nm)	0.8247
S_2	MO68 → MO74	3.5228 eV (351.94 nm)	0.0027
S_3	MO73 → MO75 (86%) MO73 → MO76 (14%)	4.0329 eV (307.43 nm)	0.0302

HOMO: MO73, LUMO: MO74

Table S34 Calculated Excited States of S_1-Optimized Geometry of **1** in Acetone.

Excited State	Transition	Energy (Wavelength)	Oscillator Strength
T_1	MO69 → MO74 (14%) MO71 → MO74 (19%) MO73 → MO74 (67%)	1.6556 eV (748.86 nm)	0.0000
T_2	MO67 → MO74 (64%) MO67 → MO75 (20%) MO67 → MO78 (16%)	2.4642 eV (503.14 nm)	0.0000
T_3	MO69 → MO74 (18%) MO71 → MO74 (21%) MO73 → MO74 (16%) MO73 → MO75 (45%)	2.5663 eV (483.12 nm)	0.0000
S_1	MO73 → MO74	2.7636 eV (448.64 nm)	0.8317
S_2	MO68 → MO74 (68%) MO68 → MO75 (19%) MO68 → MO78 (13%)	3.5069 eV (353.54 nm)	0.0023
S_3	MO73 → MO75 (86%) MO73 → MO76 (14%)	4.0218 eV (308.28 nm)	0.0297

HOMO: MO73, LUMO: MO74