

Supplementary Information

Table S1. The selected bond lengths (in Å), bond angles (in degree) and dihedrals (in degree) of the dye YD2. (B3LYP/6-31g(d,p), gas phase).

Definition	Values	Definition	Values	Definition	Values
19–21	1.42	19–21–25	122.0	19–21–26–28	–71.2
21–25	1.42	25–21–26	118.9	25–21–26–27	–71.1
21–26	1.43	19–21–26	119.1	28–30–36–49	–88.6
30–36	2.04	30–36–31	90.2	27–31–36–42	–92.8
31–36	2.04	30–36–42	89.8	34–35–38–68	–72.3
42–36	2.04	31–36–49	89.8	39–53–52–55	67.0
49–36	2.04	42–36–49	90.2	40–35–38–67	–72.7
46–47	1.42	31–36–42	179.1	50–53–52–54	66.8
47–80	1.22	30–36–49	179.2	43–45–46–47	1.2
80–82	1.42	34–35–38	117.6	48–44–46–47	–1.8
86–87	1.48	39–53–52	117.6	46–47–80–82	–75.5
87–88	1.36	45–46–47	117.5	47–80–82–83	–2.6
87–89	1.22	86–87–89	125.0	47–86–82–81	117.4
35–38	1.50	89–87–88	121.9	84–86–87–89	0.0
53–52	1.50	86–87–88	113.1	85–86–87–88	0.0

Table S2. The selected bond lengths (in Å), bond angles (in degree) and dihedrals (in degree) of the dye YD2-o-C8. (B3LYP/6-31g(d,p), gas phase).

Definition	Values	Definition	Values	Definition	Values
19–21	1.42	19–21–25	122.0	19–21–26–28	71.6
21–25	1.42	19–21–56	119.1	25–21–26–27	71.1
21–26	1.44	25–21–26	118.8	28–30–36–49	89.9
30–36	2.04	30–36–31	90.0	27–31–36–42	92.3
31–36	2.04	30–36–42	90.0	34–35–38–59	–86.5
42–36	2.04	31–36–49	90.0	39–53–52–55	97.7
49–36	2.04	42–36–49	90.0	40–35–38–60	–85.9
46–47	1.42	31–36–42	179.2	50–53–52–54	97.9
47–64	1.22	30–36–49	179.3	43–45–46–47	0.5
64–66	1.42	34–35–38	117.5	48–44–46–47	0.4
70–71	1.48	39–53–52	117.4	46–47–64–66	–12.8
71–72	1.36	45–46–47	117.5	47–64–66–65	114.4
71–73	1.22	70–71–73	125.1	47–64–66–67	–38.6
35–38	1.50	73–71–72	121.8	68–70–71–73	–0.1
53–52	1.50	70–71–72	113.2	69–70–71–72	–0.1

Table S3. The different functional calculated excitation energies (eV), wavelength (nm), oscillator strengths (f) and major transition configurations with coefficients larger than 10% of three stronger absorption bands in UV-vis region for YD2 in THF.

Functional	States	Major transition configurations	E(nm/eV)	f
CAM-B3LYP	S ₁	H - 1→L + 1 (25%); H→L (69%)	585/2.12	0.3012
	S ₃	H - 2→L (51%); H - 1→L + 1 (24%); H→L (16%)	443/2.80	0.3372
	S ₅	H - 2→L (35%); H - 1→L + 1 (45%); H→L (10%)	397/3.13	2.0155
M062X	S ₁	H - 1→L + 1 (21%); H→L (75%)	573/2.17	0.3551
	S ₃	H - 2→L (58%); H - 1→L + 1 (23%); H→L (11%)	452/2.75	0.1824
	S ₅	H - 2→L (31%); H - 1→L + 1 (50%); H→L (11%)	397/3.12	2.1369
PBE0	S ₁	H→L (96%)	902/1.37	0.2574
	S ₄	H - 3→L (18%); H - 2→L + 1 (14%); H - 1→L (52%); H→L + 2 (13%)	604/2.05	0.2844
	S ₉	H - 2→L + 1 (41%); H - 1→L + 2 (45%)	481/2.58	0.9151
BMK	S ₁	H - 1→L + 1 (13%); H→L (83%)	582/2.13	0.4536
	S ₄	H - 2→L + 1 (19%); H - 1→L (29%); H→L + 1 (51%)	430/2.88	0.4875
	S ₅	H - 2→L (22%); H - 1→L + 1 (58%)	403/3.08	2.2109
ωB97XD	S ₁	H - 1→L + 1 (27%); H→L (66%)	595/2.08	0.2667
	S ₃	H - 2→L (39%); H - 1→L + 1 (30%); H→L (20%)	428/2.90	0.6154
	S ₅	H - 2→L (42%); H - 1→L + 1 (37%)	391/3.17	1.7495
HSE06	S ₁	H→L (94%)	677/1.83	0.3660
	S ₃	H - 2→L (78%); H - 1→L + 1 (21%)	537/2.31	0.1294
	S ₇	H - 2→L (11%); H - 1→L + 1 (39%); H→L + 2 (21%)	423/2.93	1.1253
LC-ωPBE	S ₁	H - 1→L + 1 (38%); H→L (59%)	628/1.98	0.1397
	S ₃	H - 1→L + 1 (48%); H→L (36%)	399/3.11	1.7768
	S ₁	H→L (96%)	874/1.42	0.2542
OPT-LC-ωPBE	S ₄	H - 3→L (19%); H - 2→L + 1 (15%); H - 1→L (62%)	598/2.07	0.3024
	S ₉	H - 2→L + 1 (50%); H - 1→L + 2 (31%)	475/2.61	1.1893
	S ₁	H - 1→L + 1 (20%); H→L (75%)	575/2.16	0.3731
MPW1K	S ₃	H - 2→L (62%); H - 1→L + 1 (21%); H→L (11%)	461/2.69	0.1178
	S ₅	H - 2→L (29%); H - 1→L + 1 (51%); H→L (10%)	401/3.09	2.2289

Table S4. The different functional calculated excitation energies (eV), wavelength (nm), oscillator strengths (f) and major transition configurations with coefficients larger than 10% of three stronger absorption bands in UV-vis region for YD2-o-C8 in THF.

Functional	States	Major transition configurations	E(nm/eV)	f
CAM-B3LYP	S ₁	H - 1 → L + 1 (23%); H → L (73%)	592/2.10	0.3470
	S ₃	H - 2 → L (53%); H - 1 → L + 1 (25%); H → L (12%)	439/2.82	0.3594
	S ₅	H - 2 → L (33%); H - 1 → L + 1 (45%); H → L (11%)	397/3.12	1.8676
M062X	S ₁	H - 1 → L + 1 (19%); H → L (78%)	579/2.14	0.4027
	S ₃	H - 2 → L (61%); H - 1 → L + 1 (23%)	447/2.78	0.1888
	S ₅	H - 2 → L (28%); H - 1 → L + 1 (52%); H → L (11%)	397/3.12	1.9965
PBE0	S ₁	H → L (94%)	879/1.41	0.2944
	S ₃	H - 3 → L (17%); H - 1 → L (68%)	619/2.00	0.2027
	S ₁₆	H - 7 → L + 1 (15%); H - 2 → L + 1 (25%); H - 1 → L + 2 (35%)	477/2.60	0.7692
HSE06	S ₁	H → L (93%)	671/1.85	0.4237
	S ₃	H - 2 → L (78%); H - 1 → L + 1 (17%)	535/2.32	0.1058
	S ₉	H - 1 → L + 1 (42%); H → L + 2 (25%)	420/2.95	1.0332
LC-ωPBE	S ₁	H - 1 → L + 1 (35%); H → L (63%)	632/1.96	0.1792
	S ₃	H - 2 → L (11%); H - 1 → L + 1 (51%); H → L (32%)	399/3.11	1.7253
OPT-LC-ωPBE	S ₁	H → L (95%)	853/1.45	0.2862
	S ₃	H - 3 → L (25%); H - 1 → L (63%)	614/2.02	0.1651
	S ₁₆	H - 2 → L + 1 (35%); H - 1 → L + 2 (29%)	472/2.63	1.0264

Figure S1. Plot of the error function $\delta_{IP}(\omega)$ used for optimization of ω in the long-range corrected DFT functionals.

