Supporting Information

Carbon based Y-type molecules for application in nonlinear optics

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Supporting Information list:

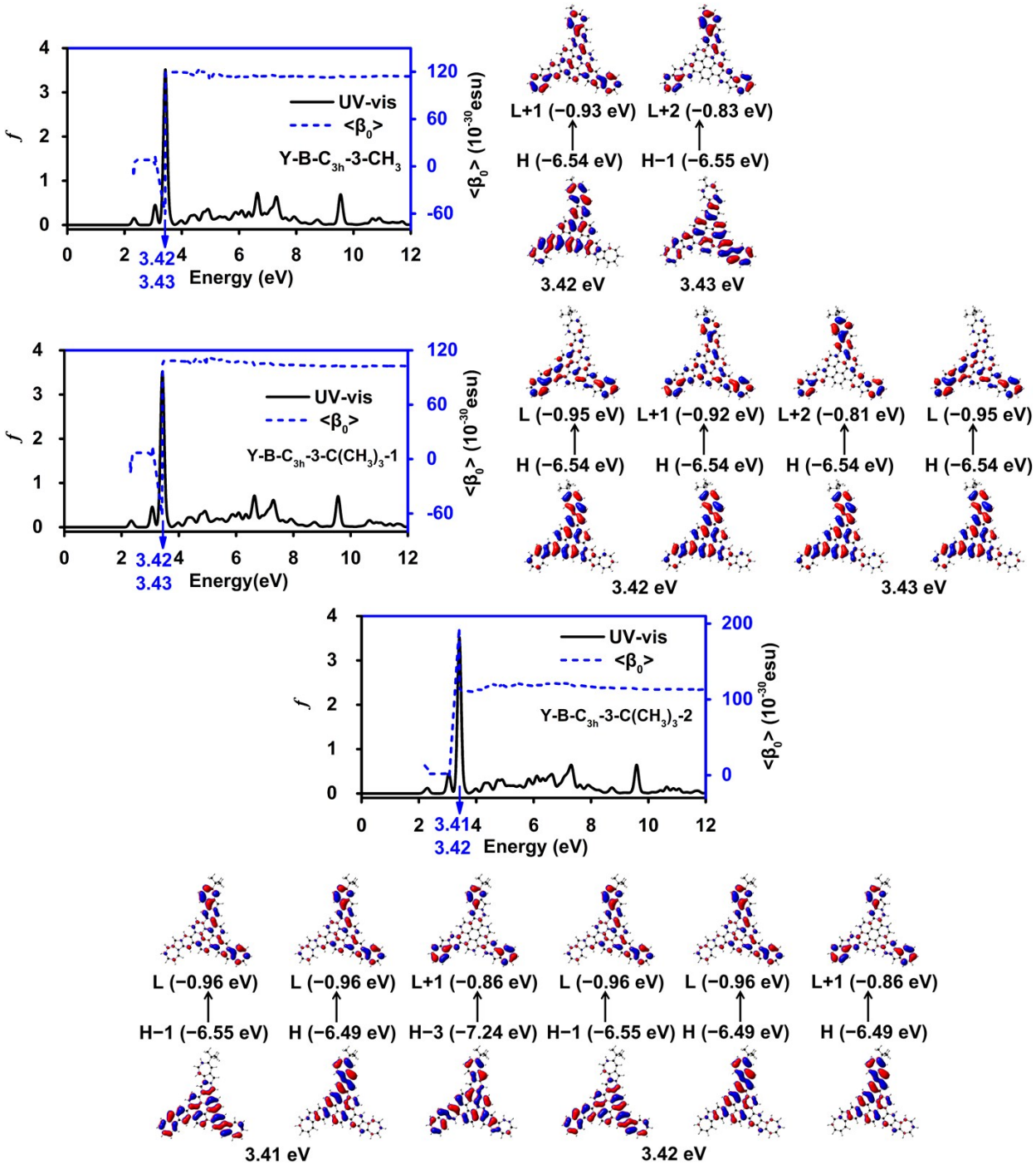


Figure S1. Evolution of the static first hyperpolarizability (<β_0>) with electron excitation in Y-B type molecules with methyl group or isobutyl group at one azulene end in Y-B-C_{3h}-3.

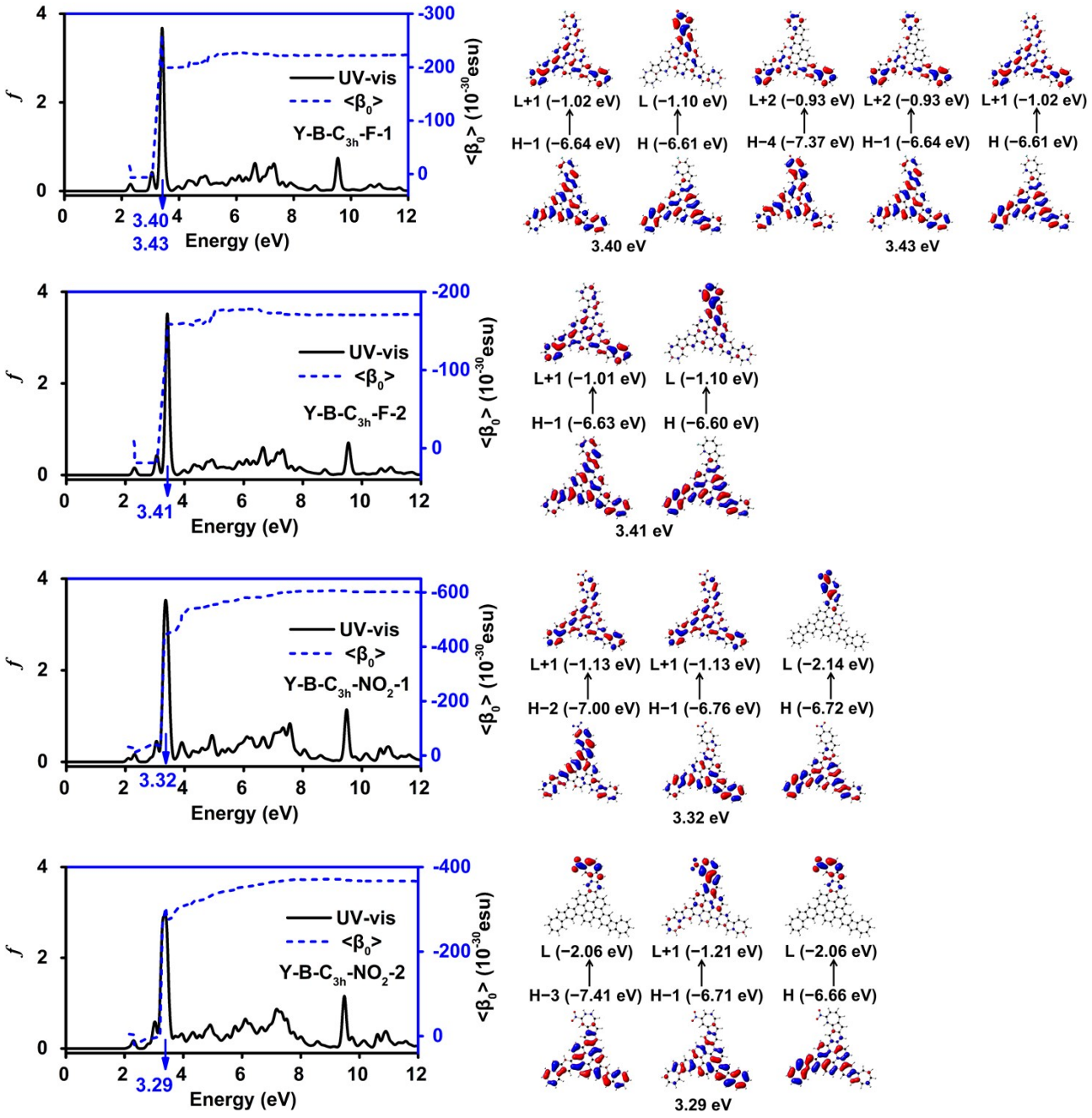


Figure S2. Evolution of the static first hyperpolarizability (<β_0>) with electron excitation in Y-B type molecules with F or NO_2 group at one azulene end in Y-B-C_{3h}-3.

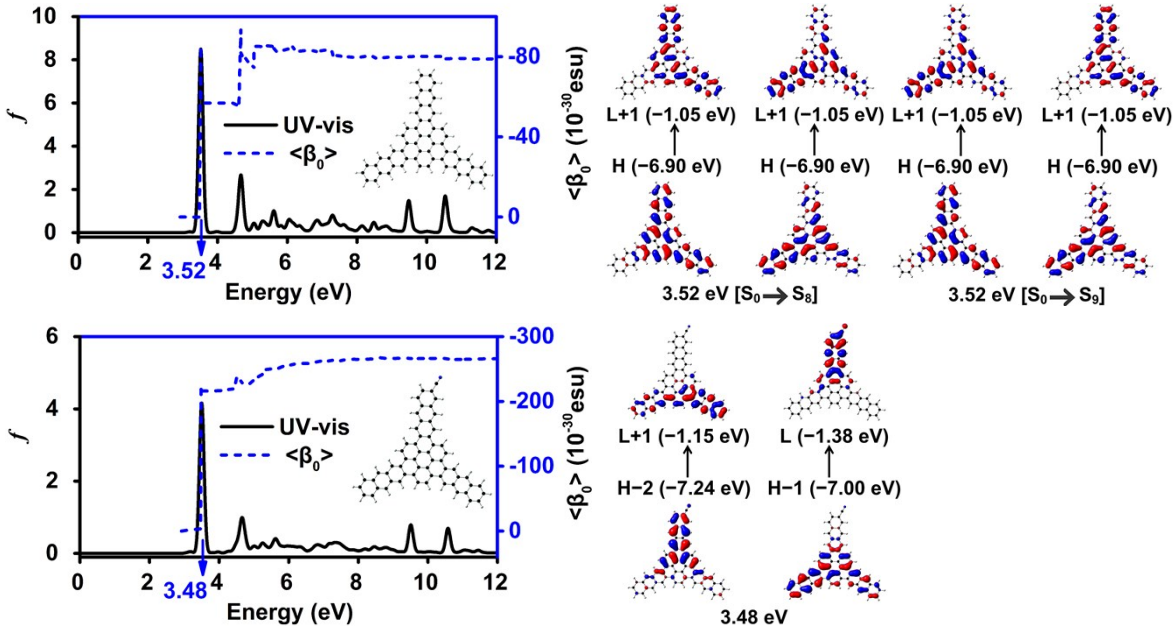


Figure S3. Evolution of the static first hyperpolarizability (<β_0>) with electron excitation in Y-B type molecules with naphthalene ends without (D_{3h}) and with CN group (C_s) at one naphthalene.

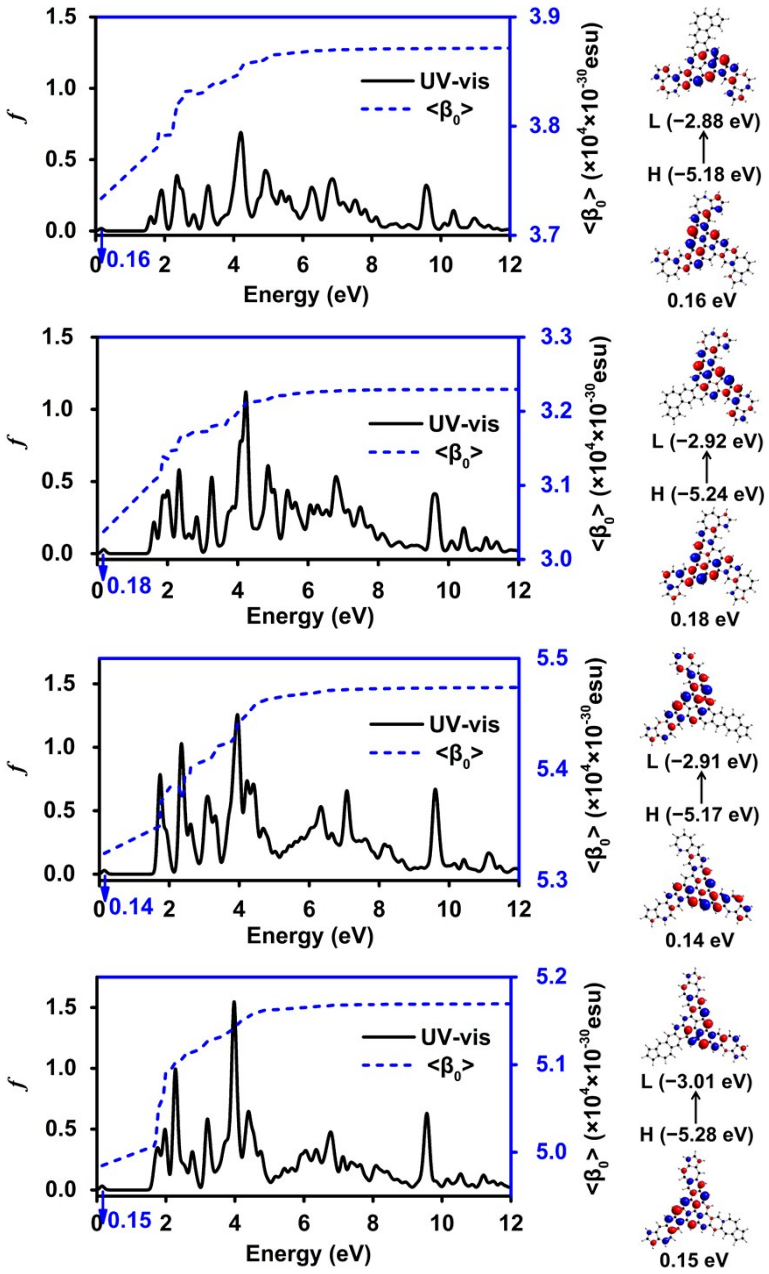


Figure S4. Evolution of the static first hyperpolarizability (<β_0>) with electron excitation in Y-Cs molecules.

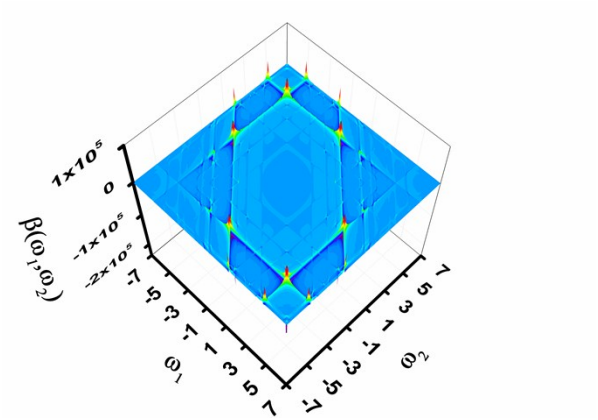
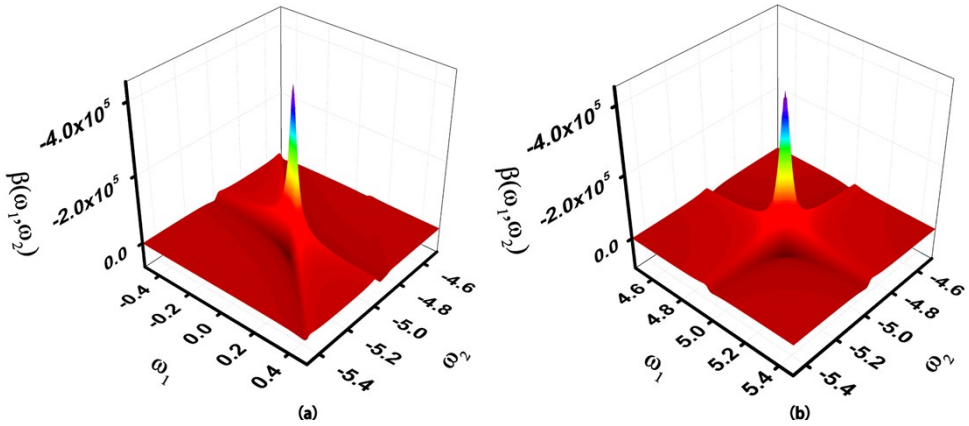


Figure S5. Two-dimensional second order NLO spectra of azulene scanned from –7.00 eV to 7.00 eV with step size of 0.05 eV.



$\beta_{Max(0,\ -4.981)} = \beta_{Max(4.980,\ -4.980)} = -429030.26\times10^{-30}$ esu

Figure S6. Two-dimensional second order NLO spectra of azulene with step size of 0.005 eV[(a) ω_1 scanned from –0.5 eV to 0.5 eV and ω_2 scanned from –5.48 eV to –4.48 eV; (b) ω_1 scanned from 4.48 eV to 5.48 eV and ω_2 scanned from –5.48 eV to –4.48 eV].

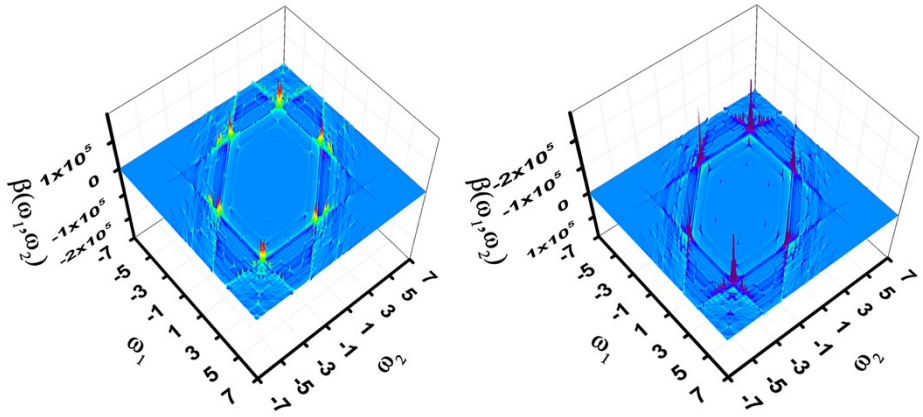


Figure S7. Two-dimensional second order NLO spectra of Y-B-NC-C_2-3 scanned from –7.00 eV to 7.00 eV with step size of 0.05 eV.

Table S1. Major electronic absorption peaks with transition nature in all molecules (f is the oscillator strength in arbitrary unit, E is the transition energy in eV, contribution to $<\beta_0>$ in 10^{-30} esu).

Compounds	f	E	Transition	Transition nature	Contribution to $<\beta_0>$
PNA($C_6H_6N_2O_2$)	0.4960	4.47	$S_0 \rightarrow S_4$	H→L (95.40%)	−31.84
Azulene($C_{10}H_8$)	0.0234	2.21	$S_0 \rightarrow S_1$	H→L (91.51%)	1.94
	0.4145	3.56	$S_0 \rightarrow S_2$	H−1→L (35.00%) H→L+1 (64.60%)	2.97
	0.2331	4.40	$S_0 \rightarrow S_3$	H−1→L (80.28%)	2.64
	2.0474	4.98	$S_0 \rightarrow S_4$	H−1→L (63.28%) H→L+1 (34.05%)	52.86
	0.2752	6.85	$S_0 \rightarrow S_{11}$	H−2→L+1 (34.49%) H−1→L+2 (61.42%)	−0.85
Y-B-NC-C_3-1($C_{36}H_{24}$)	1.2405	4.99	$S_0 \rightarrow S_{25}$	H−2→L (16.56%) H−2→L (16.56%) H−1→L+2 (28.04%)	9.54
Y-B-NC-C_1-2($C_{36}H_{24}$)	0.9562	4.80	$S_0 \rightarrow S_{23}$	H→L+5 (51.36%)	−22.87
Y-B-NC-C_2-3($C_{36}H_{24}$)	3.0325	4.18	$S_0 \rightarrow S_9$	H−4→L+1 (13.15%) H−3→L (18.94%) H−2→L+3 (36.02%)	74.72
Y-B-NC-C_2-4($C_{36}H_{24}$)	3.1090	4.18	$S_0 \rightarrow S_{11}$	H−4→L+2 (16.90%) H→L (13.05%) H−3→L+4 (10.39%) H→L+5 (14.58%)	44.57
Y-B-NC-C_2-5($C_{36}H_{24}$)	2.5249	4.16	$S_0 \rightarrow S_{11}$	H−3→L+3 (11.75%) H−2→L+3 (25.34%)	39.01
	2.5333	4.16	$S_0 \rightarrow S_{12}$	H−2→L+3 (11.28%) H−1→L+4 (16.84%)	−9.89
	0.2723	4.87	$S_0 \rightarrow S_{21}$	H−3→L (68.61%)	5.68
	0.2642	4.87	$S_0 \rightarrow S_{22}$	H−3→L+1 (67.91%) H−3→L+5 (10.04%)	−17.38
Y-B-C_S-1($C_{60}H_{30}$)	4.6117	3.42	$S_0 \rightarrow S_8$	H−3→L (15.93%) H−2→L (14.13%) H−1→L+1 (18.12%)	−58.78
	2.4018	3.45	$S_0 \rightarrow S_9$	no major transition	−50.41
Y-B-C_{3h}-2($C_{60}H_{30}$)	3.9794	3.44	$S_0 \rightarrow S_8$	H−2→L+1 (14.28%) H→L (16.58%) H→L (16.58%)	−95.41
	3.9794	3.44	$S_0 \rightarrow S_9$	H−2→L+1 (14.28%) H→L (16.58%) H→L (16.58%)	72.06
Y-B-C_{3h}-3($C_{60}H_{30}$)	3.5007	3.42	$S_0 \rightarrow S_8$	H→L+1 (11.22%) H→L (14.93%) H→L (14.93%)	28.17
	3.5007	3.42	$S_0 \rightarrow S_9$	H→L+1 (11.22%) H→L (14.93%) H→L (14.93%)	−125.79
Y-B-C_{3h}-2-CN ($C_{61}H_{29}N$)	4.4728	3.40	$S_0 \rightarrow S_8$	H−2→L+1 (23.68%)	−98.97

				H−1→L+1 (12.74%) H→L (12.59%)	
	3.9006	3.45	$S_0 \rightarrow S_9$	H−4→L+2 (16.54%) H−1→L+2 (14.83%) H→L+1 (14.83%)	45.96
Y-B-C_{3h}-2-NC ($C_{61}H_{29}N$)	4.5311	3.41	$S_0 \rightarrow S_8$	H−2→L+1 (18.23%) H−1→L+1 (15.03%) H→L (12.59%)	−33.77
	3.9141	3.45	$S_0 \rightarrow S_9$	H−4→L+2 (18.84%) H−1→L+2 (13.20%) H→L+1 (15.25%)	76.08
Y-B-C_{3h}-3-CN1($C_{61}H_{29}N$)	0.1208	2.19	$S_0 \rightarrow S_1$	H−4→L (19.92%) H−2→L (54.21%) H−1→L (13.01%)	−31.38
	3.8662	3.35	$S_0 \rightarrow S_8$	H−2→L (14.29%) H−1→L (13.09%) H→L (18.34%)	−341.73
	3.2017	3.43	$S_0 \rightarrow S_9$	H−4→L+2 (11.80%) H−1→L+2 (22.38%) H→L+1 (14.46%)	24.96
Y-B-C_{3h}-3-CN2($C_{61}H_{29}N$)	0.2494	2.86	$S_0 \rightarrow S_4$	H−2→L+1 (22.79%) H−2→L+1 (15.74%)	−32.97
	0.1939	3.38	$S_0 \rightarrow S_8$	H−3→L (10.29%) H→L (28.07%)	−451.66
Y-B-C_{3h}-3-CH_3($C_{61}H_{32}$)	3.5159	3.42	$S_0 \rightarrow S_8$	H→L+1 (14.18%)	−72.54
	3.4927	3.43	$S_0 \rightarrow S_9$	H−1→L+2 (14.99%)	179.86
Y-B-C_{3h}-3-$C(CH_3)_3$-1 ($C_{64}H_{38}$)	3.5591	3.42	$S_0 \rightarrow S_8$	H→L (12.49%) H→L+1 (19.20%)	−76.83
	3.5108	3.43	$S_0 \rightarrow S_9$	H→L+2 (19.04%) H→L (12.49%)	170.41
Y-B-C_{3h}-3-$C(CH_3)_3$-2 ($C_{64}H_{38}$)	3.4659	3.41	$S_0 \rightarrow S_8$	H−1→L (16.82%) H→L (15.90%)	195.35
	3.5228	3.42	$S_0 \rightarrow S_9$	H−3→L+1 (10.38%) H−1→L (15.50%) H→L (10.36%) H→L+1 (11.35%)	−82.45
Y-B-C_{3h}-3-F-1($C_{60}H_{29}F$)	3.6772	3.40	$S_0 \rightarrow S_8$	H−1→L+1 (15.04%) H→L (21.10%)	−262.56
	3.4096	3.43	$S_0 \rightarrow S_9$	H−4→L+2 (12.04%) H−1→L+2 (19.62%) H→L+1 (15.16%)	62.88
Y-B-C_{3h}-3-F-2($C_{60}H_{29}F$)	3.5192	3.41	$S_0 \rightarrow S_8$	H−1→L+1 (13.17%) H→L (23.04%)	−159.21
Y-B-C_{3h}-3-NO_2-1 ($C_{60}H_{29}NO_2$)	3.5323	3.32	$S_0 \rightarrow S_9$	H−2→L+1 (21.42%) H−1→L+1 (11.94%) H→L (17.23%)	−407.36
Y-B-C_{3h}-3-NO_2-2 ($C_{60}H_{29}NO_2$)	2.9487	3.29	$S_0 \rightarrow S_9$	H−3→L (10.73%) H−1→L+1 (12.80%)	−268.18

				H→L (23.75%)	
Y-B-D_{3h}($C_{60}H_{30}$)	4.2492	3.52	S_0→S_8	H→L+1 (29.17%) H→L+1 (29.17%)	−85.34
	4.2492	3.52	S_0→S_9	H→L+1 (29.17%) H→L+1 (29.17%)	28.45
Y-B-D_{3h}-CN($C_{61}H_{29}N$)	4.1297	3.48	S_0→S_8	H−2→L+1 (29.17%) H−1→L (29.17%)	−220.72
Y-C-C_{3h}-1($C_{58}H_{30}$)	0.0478	0.16	S_0→S_1	H→L (91.30%)	37336.50
Y-C-C_S-2($C_{58}H_{30}$)	0.0511	0.18	S_0→S_1	H→L (91.91%)	30375.50
Y-C-C_S-3($C_{58}H_{30}$)	0.0397	0.14	S_0→S_1	H→L (89.26%)	53243.80
Y-C-C_{3h}-4($C_{58}H_{30}$)	0.0522	0.15	S_0→S_1	H→L (91.04%)	49846.40
Y-C-C_S-5($C_{58}H_{30}$)	0.1023	0.29	S_0→S_1	H→L (91.85%)	14705.50
Y-C-C_{3h}-6($C_{58}H_{30}$)	0.0628	0.17	S_0→S_1	H→L (90.86%)	42145.20
Y-Ref ($C_{50}H_{21}N_7O_2S_2$)	3.0853	2.58	S_0→S_1	H−1→L+1 (35.28%) H→L (39.36%)	−115.47
	0.9635	2.70	S_0→S_2	H−1→L (40.16%) H→L+1 (35.41%)	−96.27