SUPPLEMENTARY INFORMATION

General Procedures of Preparations

Chemicals of the highest commercial grade available, purchased from Wako, TCI and Kanto were used as received without further purification. (2*S*, 3*S*)-3-Aminoaspartic acid was prepared according to the literature procedures (Ibarra AG *et al.* J Org Chem 1997; 62: 2478).

Table S1: Crystal Data and Structure Refinement for 2, 4 and 6

	2	**4**	**6**
Empirical formula	$C_{19}H_{13}Cl_4CuN_2O_7$	$C_{35}H_{49}CuN_2O_7$	$C_{19}H_{17}CuN_2O_9$
Formula weight	586.67	673.32	480.89
Crystal system	Monoclinic	Triclinic	Monoclinic
Space group	$P2_1/n$ （#14）	*P*1 (#1)	$P2_1/n$ （#14）
a (Å)	10.880 (5)	13.661 (10)	14.34 (4)
b (Å)	10.559 (5)	17.014 (14)	19.24 (4)
c (Å)	28.702 (13)	11.644 (9)	13.77 (4)
α (°)	100.429 (16)	112.63 (3)	91.3 (2)
β (°)		106.69 (6)	
γ (°)		75.46 (6)	
V (Å^3)	3243 (3)	2362 (3)	3798 (17)
Z	4	1	4
Rwp (%)	19.24	26.83	17.87

Table S2: Selected Bond Lengths and Angles for 2

Cu 1-O 1	1.9326(6)Å	O 1-Cu 1-O 2	92.862(17)°
Cu 1-O 2	1.8985(8)Å	O 1-Cu 1-N 1	91.25(2)°
Cu 1-O 3	2.2532(7)Å	O 1-Cu 1-N 2	170.671(4)°
Cu 1-N 1	1.9740(7)Å	O 2-Cu 1-O 3	91.01(2)°
Cu 1-N 2	2.0033(7)Å	O 2-Cu 1-N 1	171.695(4)°
		O 2-Cu 1-N 2	92.705(17)°
		N 1-Cu 1-N 2	84.20(2)°

Table S3: Selected Bond Lengths and Angles for 4

Cu 1-O 1	1.9108(13)Å	O 1-Cu 1-O 2	92.62(7)°
Cu 1-O 2	1.9128(15)Å	O 1-Cu 1-N 1	91.90(7)°
Cu 1-O 3	2.0546(16)Å	O 1-Cu 1-N 2	171.203(9)°
Cu 1-N 1	1.9720(16)Å	O 2-Cu 1-O 3	96.09(6)°
Cu 1-N 2	1.9694(13)Å	O 2-Cu 1-N 1	171.208(10)°
		O 2-Cu 1-N 2	91.90(7)°
		N 1-Cu 1-N 2	84.65(7)°

Table S4: Selected Bond Lengths and Angles for 6

Cu 1-O 1	1.922(4)Å	O 1-Cu 1-O 2	93.9(2)˚
Cu 1-O 2	1.914(4)Å	O 1-Cu 1-N 1	171.26(3)˚
Cu 1-O 3	2.514(5)Å	O 1-Cu 1-N 2	91.6(2)˚
Cu 1-N 1	1.984(4)Å	O 2-Cu 1-O 3	84.38(17)˚
Cu 1-N 2	1.985(4)Å	O 2-Cu 1-N 1	92.14(19)˚
		O 2-Cu 1-N 2	171.678(4)˚
		N 1-Cu 1-N 2	83.12(19)˚

Table S5: Electron Transition of Chiral Salen Cu(II) Complexes 1-9

	π-π*	**d-d**
1	HOMO -2 → LUMO	HOMO -4 → LUMO
2	HOMO → LUMO +1	HOMO -5 → LUMO
3	HOMO -1 → LUMO +1	HOMO -5 → LUMO
4	HOMO → LUMO +1	HOMO → LUMO +2
5	HOMO -2 → LUMO +3	HOMO -5 → LUMO
6	HOMO → LUMO +1	HOMO -4 → LUMO
7	HOMO -1 → LUMO +2	HOMO -3 → LUMO
8	HOMO → LUMO +2	HOMO -3 → LUMO
9	HOMO → LUMO +2	HOMO → LUMO +1

Table S6: Electrochemical Measurement of Chiral Salen Cu(II) Complexes 1-9

	Ered [V vs NHE]	**Eox [V vs NHE]**	**HOMO [eV]**	**LUMO [eV]**	**HOMO-LUMO gap [eV]**
1	-0.758	0.682	-4.46	-3.02	1.57
2	-0.798	0.858	-4.64	-2.98	1.66
3	-0.868	0.252	-4.03	-2.91	1.12
4	-0.829	0.388	-4.17	-2.95	1.22
5	-0.938	0.232	-4.01	-2.84	1.17
6	-0.758	0.632	-4.41	-3.02	1.07
7	-0.798	0.672	-4.45	-2.98	1.39
8	-0.838	0.732	-4.51	-2.94	1.61
9	-0.778	0.292	-4.07	-3.00	1.47

Table S7: Absorption Wavelength, Energy Levels and HOMO-LUMO Gap of Chiral Salen Cu(II) Complexes 1-9

	λ/π-π* [nm]	**λ calc./π-π* [nm]**	**E_{red} [V vsNHE]**	**E_{ox} [V vs NHE]**	**HOMO-LUMO gap [eV]**	**HOMO-LUMO gap calc. [eV]**
1	346	381	-0.758	0.682	-3.02	4.216
2	390	398	-0.798	0.858	-2.98	4.266
3	421	410	-0.868	0.252	-2.91	4.059
4	389	394	-0.829	0.388	-2.95	4.014
5	416	408	-0.938	0.232	-2.84	4.153
6	348	405	-0.758	0.632	-3.02	4.182
7	526	389	-0.798	0.672	-2.98	3.890
8	375	392	-0.838	0.732	-2.94	4.053
9	430	429	-0.778	0.292	-3.00	4.270

Table S8: Photovoltaic Performance of DSSC Based on Salen Dyes and N3

	Voc [V]	Jsc [$mAcm^{-2}$]	FF [%]	η [$\times 10^{-2}$%]
1	0.301	0.071	29.0	0.6126
2	0.397	0.400	21.0	3.3486
5	0.208	0.142	24.0	0.7071
9	0.189	0.032	28.0	0.1690
N3	0.405	0.379	20.0	1.3664

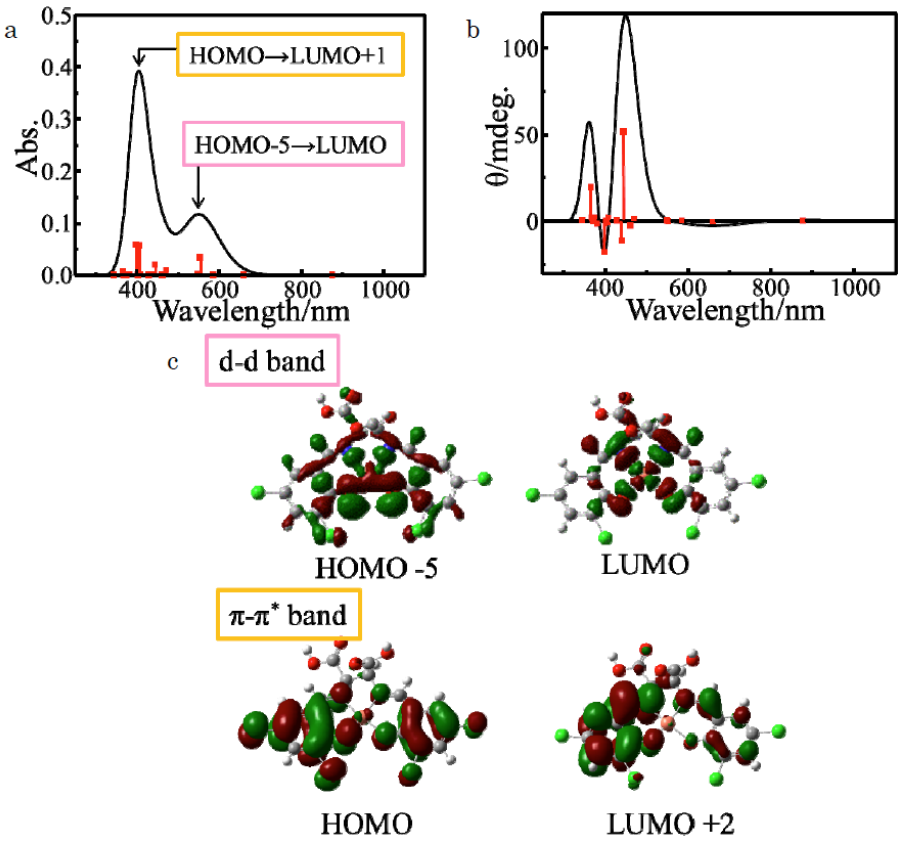

Figure S1: (**a**) Simulated UV-vis spectra, (**b**) simulated CD spectra and (**c**) molecular orbitals of the ground state (HOMO) and excited state (LUMO) for **2**.

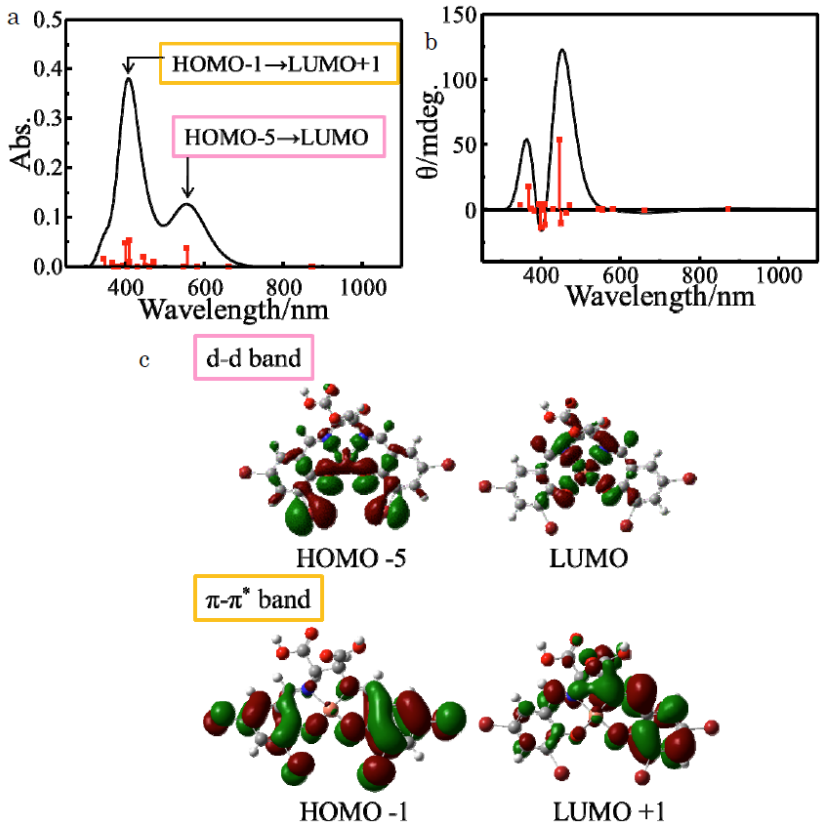

Figure S2: (**a**) Simulated UV-vis spectra, (**b**) simulated CD spectra and (**c**) molecular orbitals of the ground state (HOMO) and excited state (LUMO) for **3**.

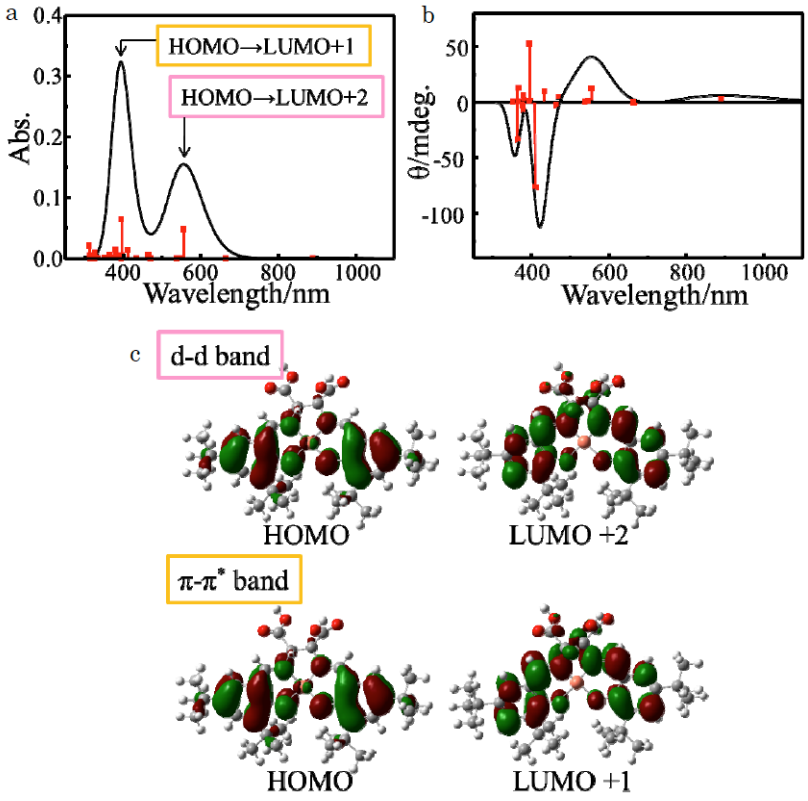

Figure S3: (**a**) Simulated UV-vis spectra, (**b**) simulated CD spectra and (**c**) molecular orbitals of the ground state (HOMO) and excited state (LUMO) for **4**.

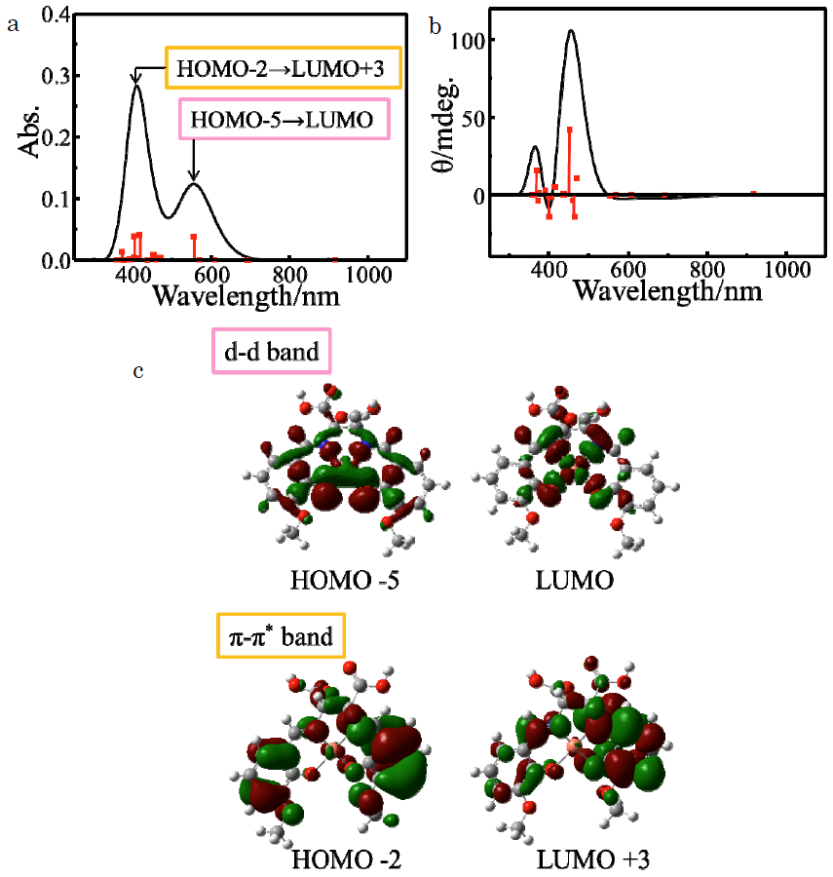

Figure S4: (**a**) Simulated UV-vis spectra, (**b**) simulated CD spectra and (**c**) molecular orbitals of the ground state (HOMO) and excited state (LUMO) for **5**.

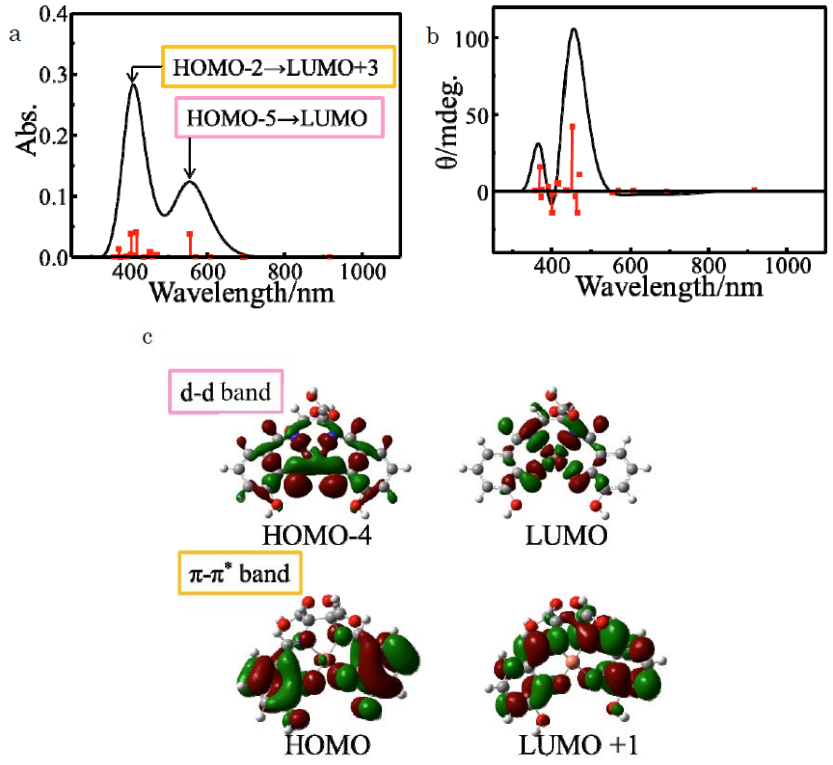

Figure S5: (**a**) Simulated UV-vis spectra, (**b**) simulated CD spectra and (**c**) molecular orbitals of the ground state (HOMO) and excited state (LUMO) for **6**.

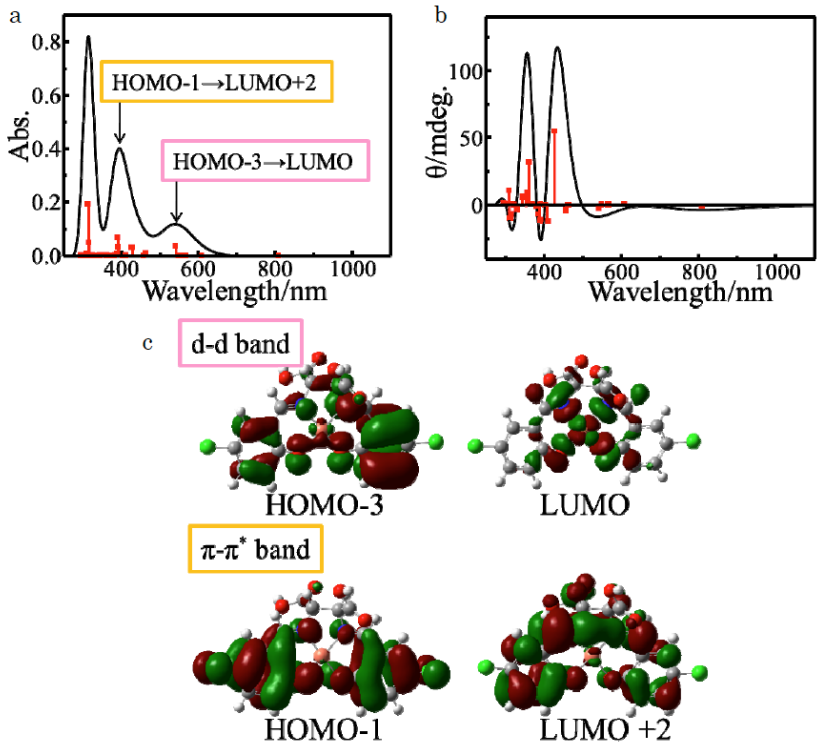

Figure S6: (**a**) Simulated UV-vis spectra, (**b**) simulated CD spectra and (**c**) molecular orbitals of the ground state (HOMO) and excited state (LUMO) for **7**.

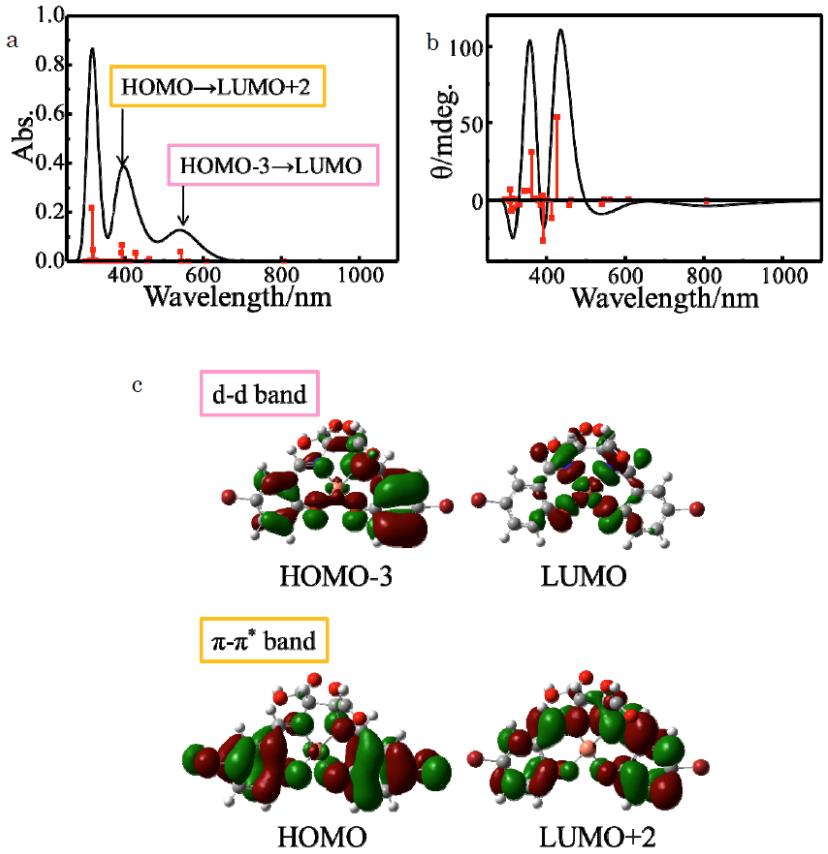

Figure S7: (**a**) Simulated UV-vis spectra, (**b**) simulated CD spectra and (**c**) molecular orbitals of the ground state (HOMO) and excited state (LUMO) for **8**.

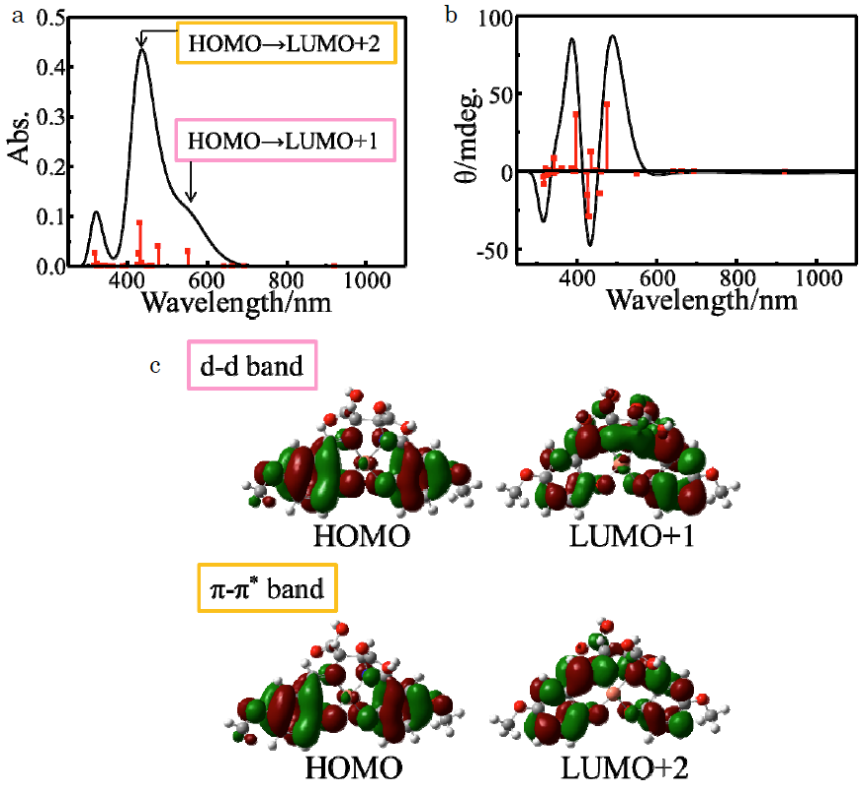

Figure S8: (**a**) Simulated UV-vis spectra, (**b**) simulated CD spectra and (**c**) molecular orbitals of the ground state (HOMO) and excited state (LUMO) for **9**.