Supporting Information

Os(II) Metal Phosphors Bearing Tridentate 2,6-Di(pyrazol-3-yl)pyridine Chelate; Synthetic Design, Characterization and Application in OLED Fabrications

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Table S1. The wavelengths, transition probabilities and charge transfer character of the optical transitions for complex **1** in dichloromethane.

State	λ_{cal} (nm)	f	Assignments	MLCT
T_1	453.6	0	HOMO→LUMO(90%) HOMO-1→LUMO(5%)	8.00%
T_2	421	0	HOMO→LUMO+1(56%) HOMO-2→LUMO(19%) HOMO-1→LUMO+1(17%)	10.85%
S_1	410.4	0.0059	HOMO→LUMO(96%)	8.23%
T_3	396	0	HOMO-1→LUMO(86%) HOMO→LUMO(6%)	5.51%
S_2	336.7	0.2564	HOMO-1→LUMO(95%)	5.52%
S_3	309.6	0.0032	HOMO-3→LUMO(95%)	53.31%

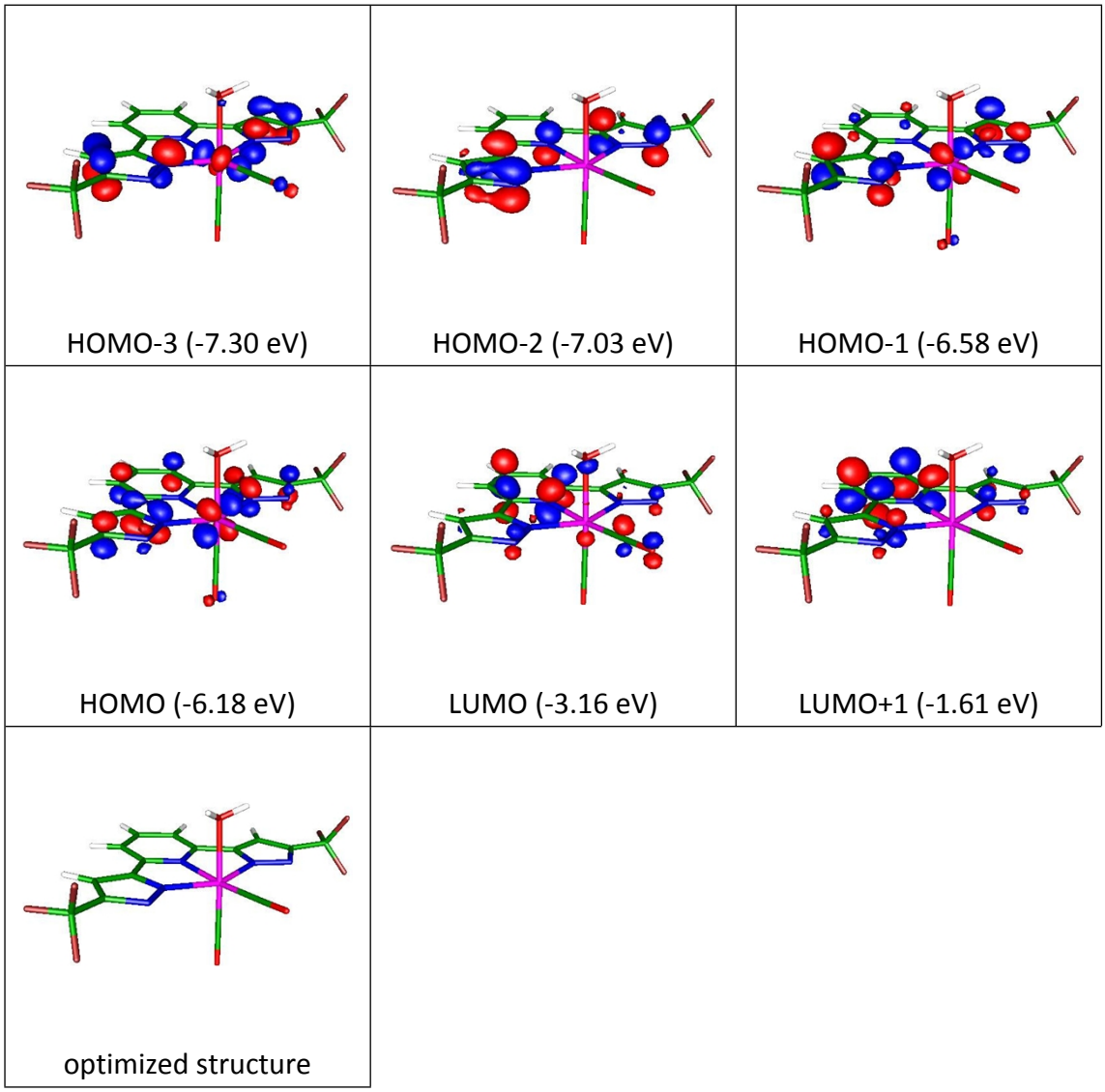

Figure S1. Frontier molecular orbitals pertinent to the optical transitions for complex **1**. For the clarity of viewing, the optimized structure with no involvement of frontier orbitals is shown at the last figure.

Table S2. The wavelengths, transition probabilities and charge transfer character of the optical transitions for complex **2** in dichloromethane.

State	λ_{cal} (nm)	f	Assignments	MLCT
T_1	462.9	0	HOMO→LUMO(96%)	46.63%
S_1	437.8	0.0011	HOMO→LUMO(99%)	48.08%
T_2	435.2	0	HOMO→LUMO+1(63%) HOMO-1→LUMO+1(23%)	32.05%
T_3	384.2	0	HOMO-1→LUMO(82%)	4.12%
S_2	357.9	0.0459	HOMO→LUMO+1(94%)	45.98%
S_3	336.3	0.0042	HOMO→LUMO+2(89%)	36.10%

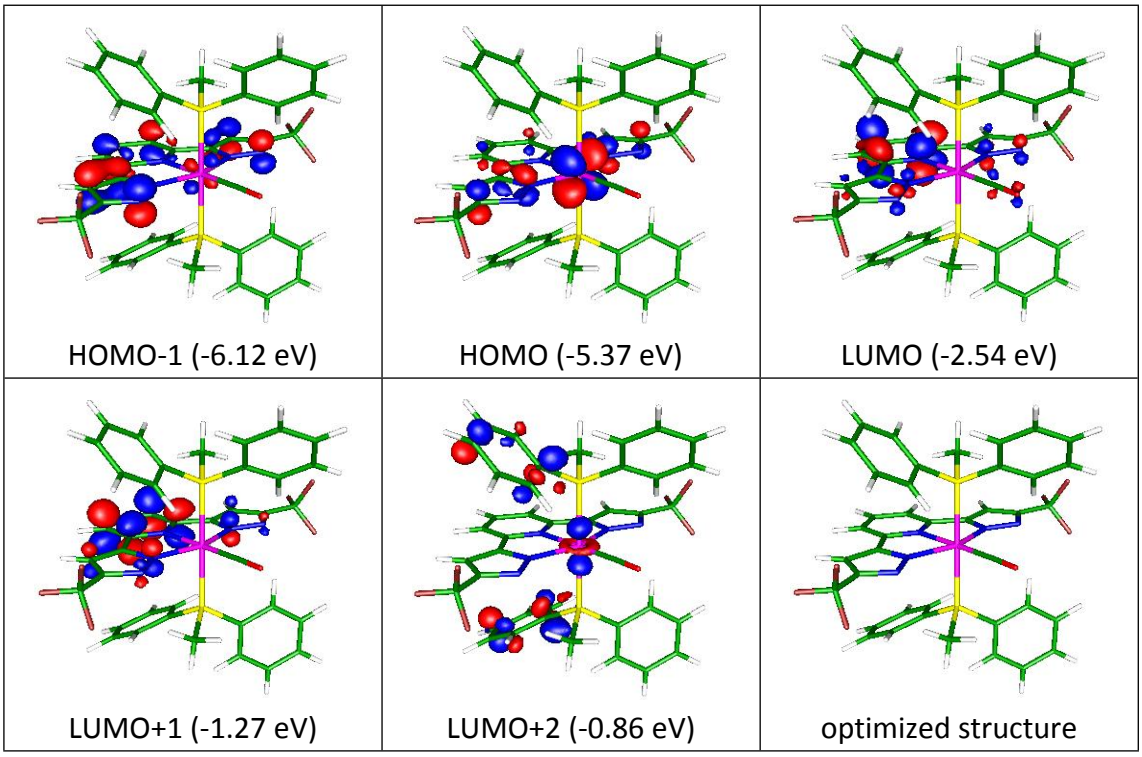

Figure S2. Frontier molecular orbitals pertinent to the optical transitions for complex **2**. For the clarity of viewing, the optimized structure with no involvement of frontier orbitals is shown at the last figure.

Table S3. The wavelengths, transition probabilities and charge transfer character of the optical transitions for complex **3** in dichloromethane.

State	λ_{cal} (nm)	f	Assignments	MLCT
T_1	558.9	0	HOMO-2→LUMO+1(92%)	61.67%
T_2	537	0	HOMO-1→LUMO+3(78%) HOMO-1→LUMO+12(11%) HOMO-1→LUMO+6(7%)	29.38%
T_3	498.8	0	HOMO-2→LUMO+3(70%) HOMO-2→LUMO+12(12%) HOMO-2→LUMO+6(11%)	49.14%
S_1	468.2	0.0001	HOMO→LUMO+3(56%) HOMO-1→LUMO+1(31%)	30.88%
S_2	462.5	0.0082	HOMO→LUMO+1(85%) HOMO-1→LUMO+3(11%)	41.61%
S_3	455.2	0.0002	HOMO-1→LUMO+1(66%) HOMO→LUMO+3(24%)	37.11%

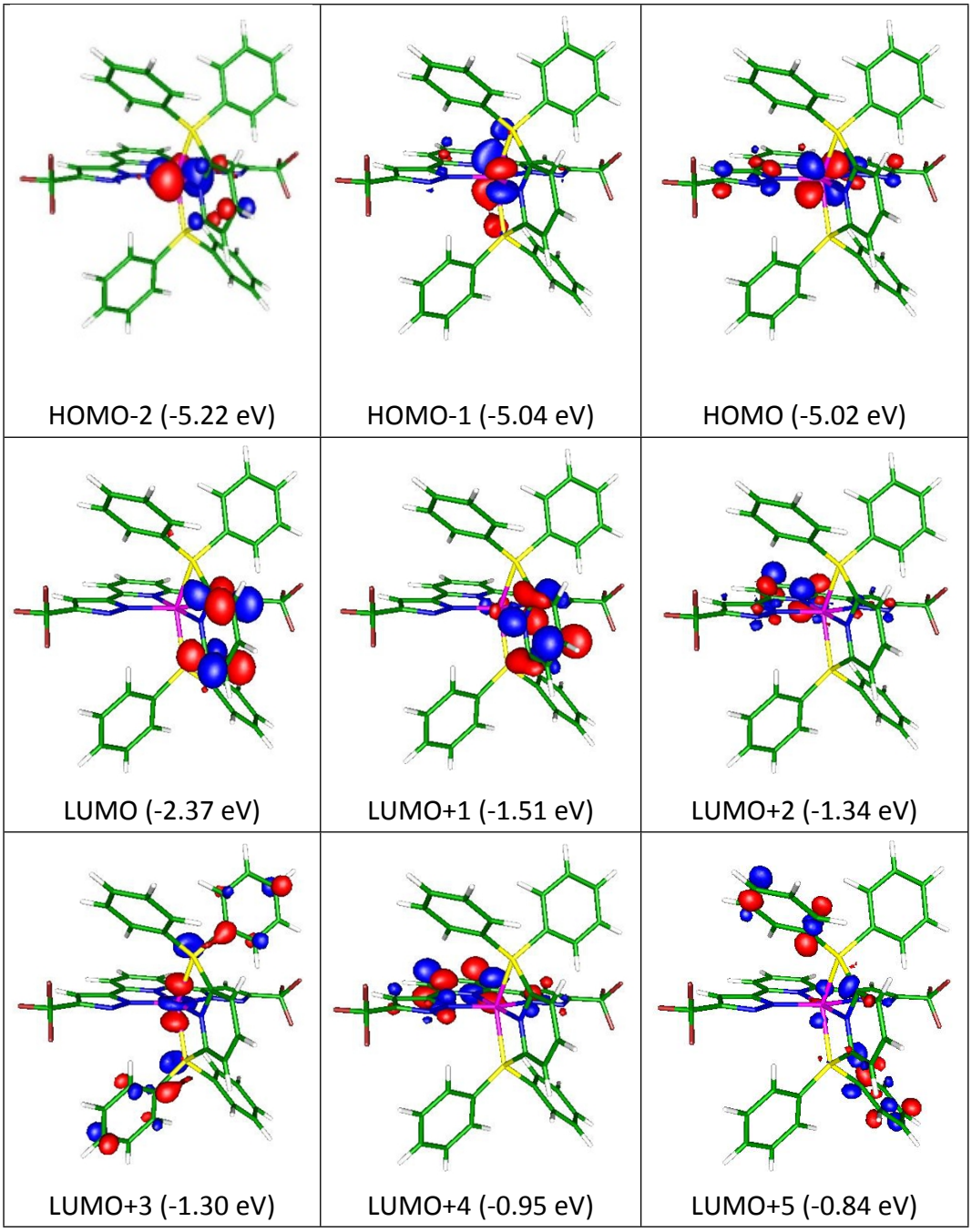

Figure S3-1. Frontier molecular orbitals (from HOMO-2 to LUMO+5) for complex **3**.

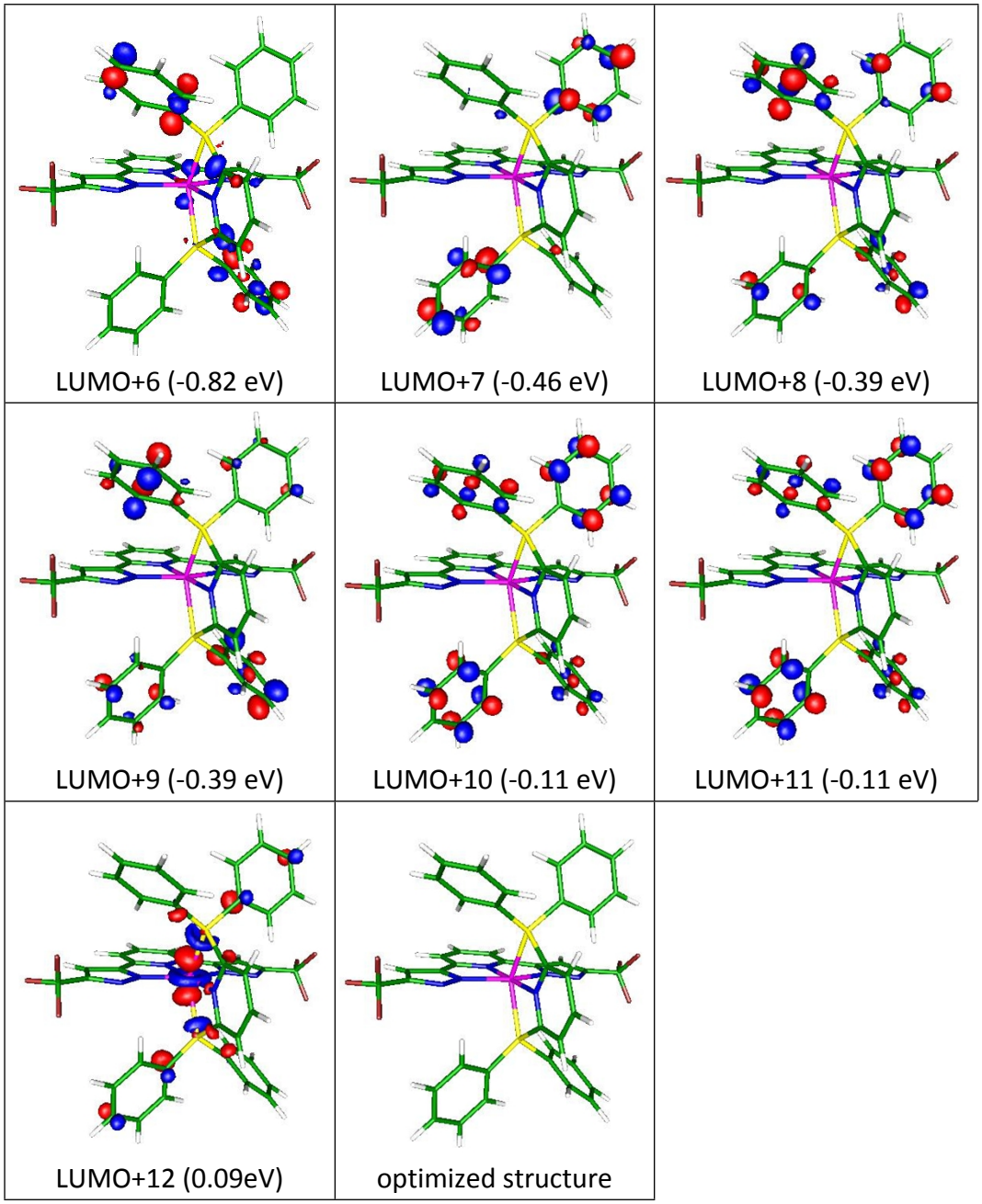

Figure S3-2. Frontier molecular orbitals (from LUMO+6 to LUMO+12) for complex **3**. For the clarity of viewing, the optimized structure with no involvement of frontier orbitals is shown at the last figure.

Table S4. The wavelengths, transition probabilities and charge transfer character of the optical transitions for complex **4** in dichloromethane.

State	λ_{cal} (nm)	f	Assignments	MLCT
T_1	434.9	0	HOMO→LUMO(87%) HOMO-1→LUMO(8%)	15.72%
T_2	418.8	0	HOMO→LUMO+2(54%) HOMO-1→LUMO+2(19%) HOMO-2→LUMO(15%)	13.91%
S_1	393.3	0.0105	HOMO→LUMO(98%)	16.87%
T_3	378	0	HOMO-1→LUMO(83%) HOMO→LUMO(9%)	9.28%
S_2	336.9	0.0048	HOMO→LUMO+1(96%)	18.65%
S_3	323.9	0.2373	HOMO-1→LUMO(96%)	8.94%

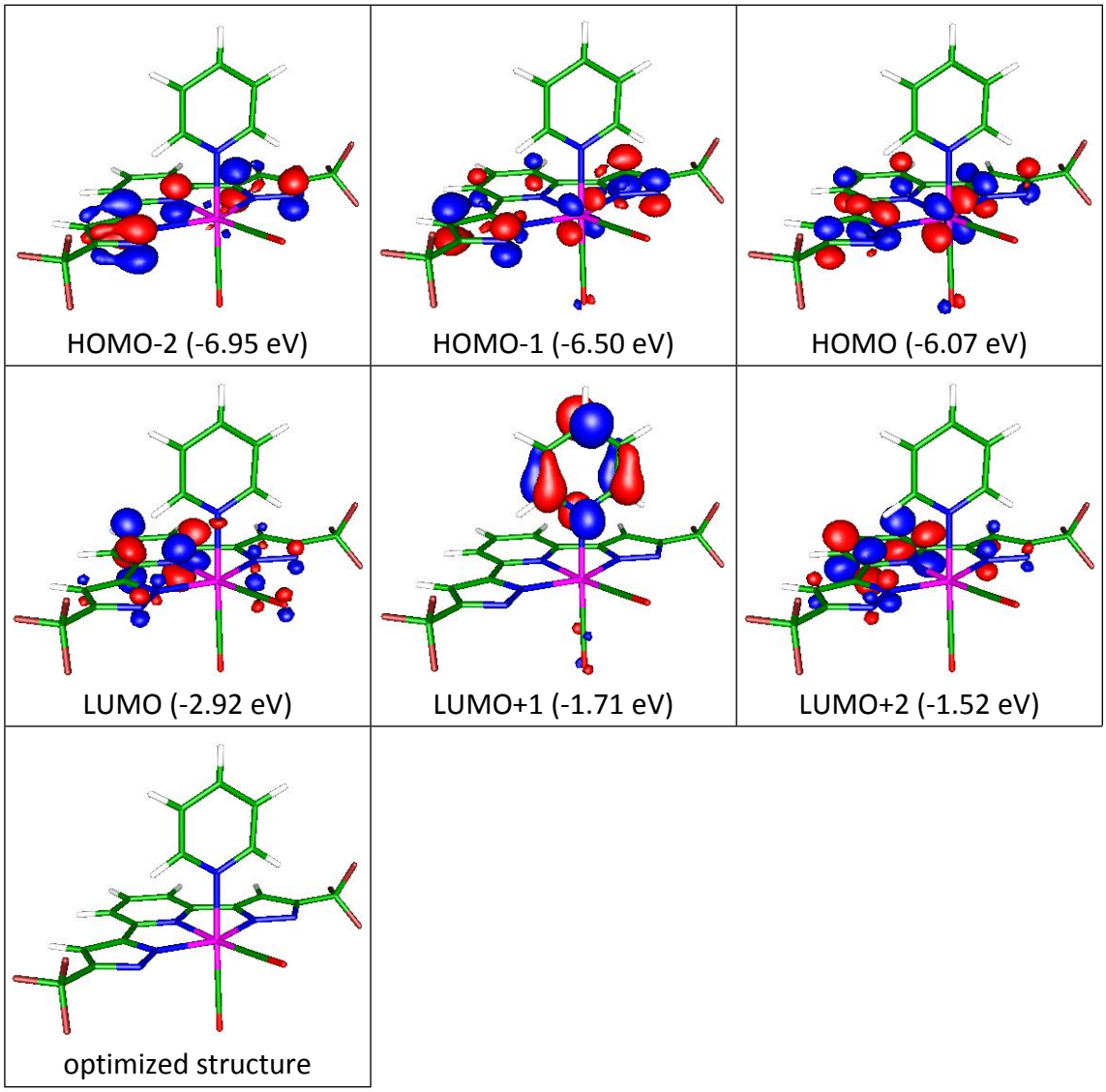

Figure S4. Frontier molecular orbitals pertinent to the optical transitions for complex **4**.

Table S5. The wavelengths, transition probabilities and charge transfer character of the optical transitions for complex **5** in dichloromethane.

State	λ_{cal} (nm)	f	Assignments	MLCT
T_1	555.7	0	HOMO-1→LUMO(89%) HOMO→LUMO(5%)	63.80%
T_2	498	0	HOMO→LUMO(89%)	24.67%
S_1	481.2	0.0011	HOMO→LUMO(77%) HOMO-1→LUMO(21%)	36.07%
S_2	453.8	0.1129	HOMO-1→LUMO(76%) HOMO→LUMO(21%)	59.12%
T_3	439.2	0	HOMO→LUMO+1(91%) HOMO-2→LUMO+1(6%)	26.42%
S_3	407.4	0.0056	HOMO→LUMO+1(97%)	27.83%

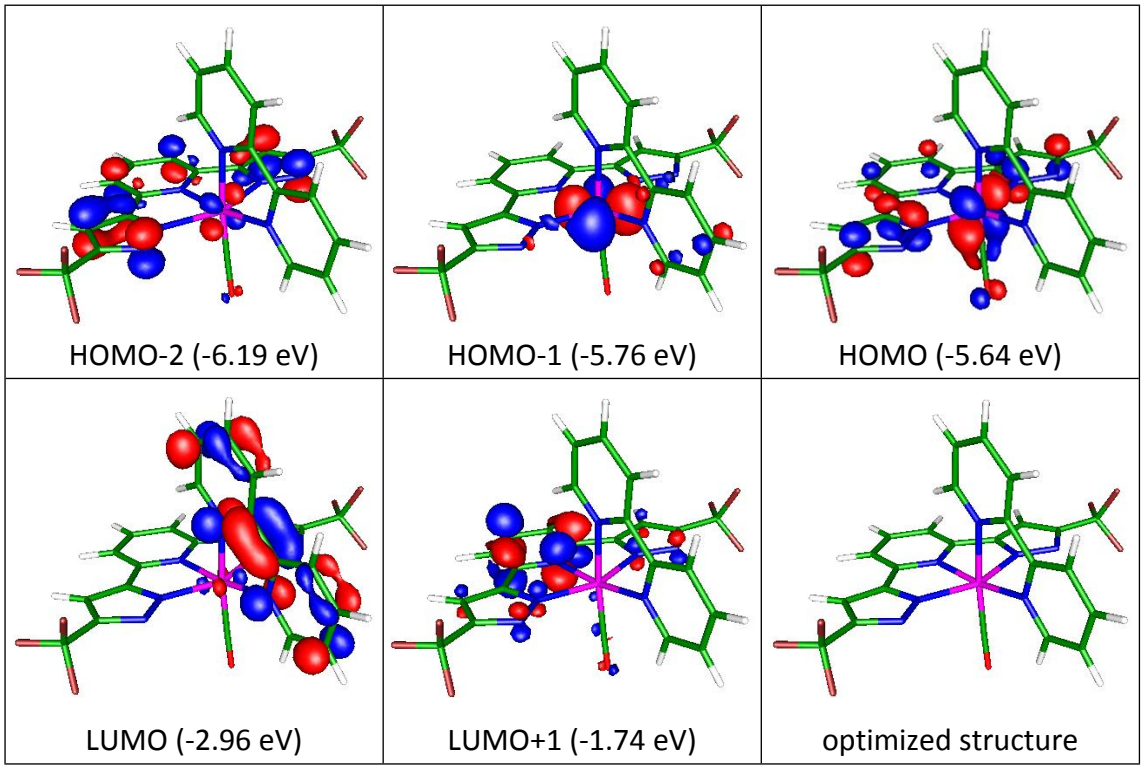

Figure S5. Frontier molecular orbitals pertinent to the optical transitions for complex **5**. For the clarity of viewing, the optimized structure with no involvement of frontier orbitals is shown at the last figure.

Table S6. The wavelengths, transition probabilities and charge transfer character of the optical transitions for complex **6** in dichloromethane.

State	λ_{cal} (nm)	f	Assignments	MLCT
T_1	566.8	0	HOMO-1→LUMO(76%) HOMO→LUMO(16%)	55.68%
T_2	506.6	0	HOMO→LUMO(78%) HOMO-1→LUMO(17%)	39.49%
S_1	501	0.007	HOMO→LUMO(89%) HOMO-1→LUMO(9%)	38.24%
S_2	466.3	0.0672	HOMO-1→LUMO(88%) HOMO→LUMO(9%)	61.01%
T_3	444.5	0	HOMO→LUMO+1(85%)	31.08%
S_3	415.8	0.0019	HOMO→LUMO+1(90%) HOMO-1→LUMO+1(5%)	36.21%

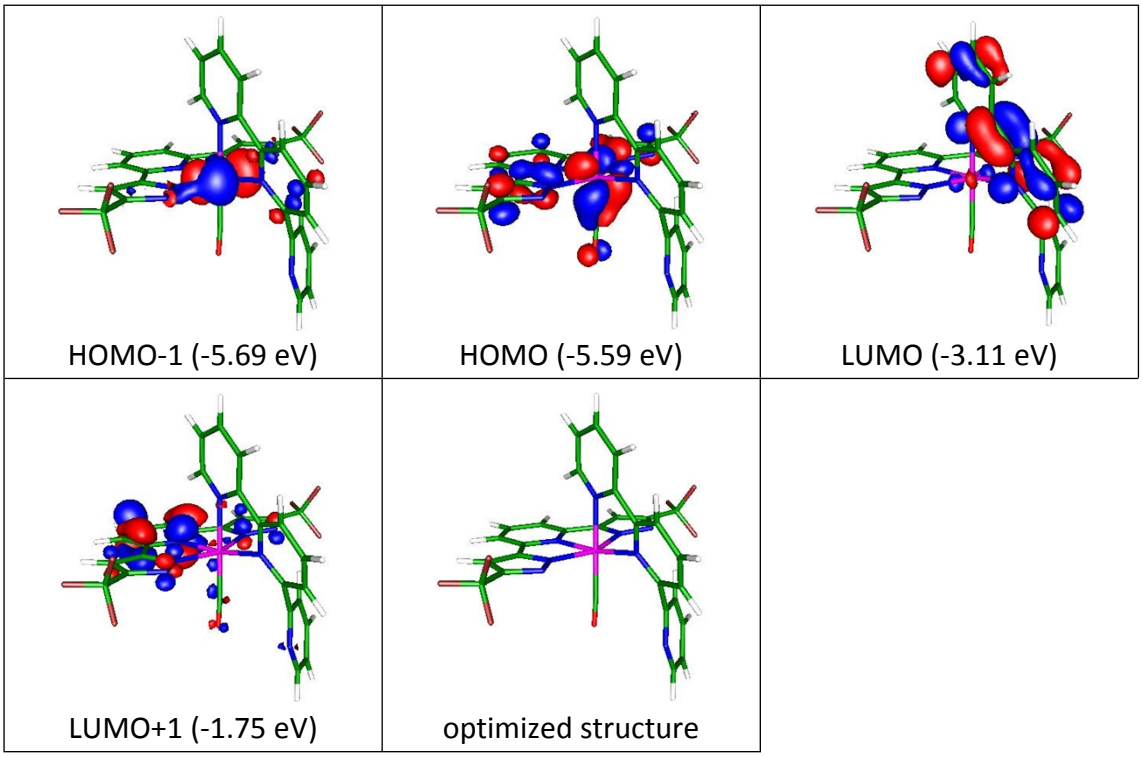

Figure S6. Frontier molecular orbitals pertinent to the optical transitions for complex **6**. For the clarity of viewing, the optimized structure with no involvement of frontier orbitals is shown at the last figure.

Table S7. The electron density distribution (%) in the molecular fragment of frontier molecular orbitals for complex **1** obtained from the Mulliken population analysis (MPA) method.

	Os	pz2py	$(CO)_2$	H_2O
HOMO-3	65.09	19.49	15.40	0.03
HOMO-2	2.91	95.74	1.25	0.10
HOMO-1	14.78	81.94	3.21	0.08
HOMO	17.54	78.20	4.05	0.19
LUMO	8.97	79.74	9.26	2.00
LUMO+1	0.46	98.02	1.45	0.05

Table S8. The electron density distribution (%) in the molecular fragment of frontier molecular orbitals for complex **2** obtained from the Mulliken population analysis (MPA) method.

	Os	pz2py	$(PPh_2Me)_2$	CO
HOMO-1	6.51	91.41	2.08	0.04
HOMO	50.06	46.67	3.28	0.02
LUMO	1.49	90.41	3.09	4.97
LUMO+1	1.15	96.08	2.75	0
LUMO+2	9.5	1.51	88.72	0.27

Table S9. The electron density distribution (%) in the molecular fragment of frontier molecular orbitals for complex **3** obtained from the Mulliken population analysis (MPA) method.

	Os	pz2py	P2N
HOMO-2	73.08	7.17	19.77
HOMO-1	51.35	21.30	27.32
HOMO	51.08	43.51	5.42
LUMO	1.79	1.75	96.45
LUMO+1	6.05	0.80	93.16
LUMO+2	4.73	77.29	17.95
LUMO+3	21.02	1.37	77.60
LUMO+4	1.85	95.23	2.90
LUMO+5	2.75	7.94	89.28
LUMO+6	8.22	2.43	89.35
LUMO+7	2.75	3.79	93.44
LUMO+8	1.52	0.54	97.91
LUMO+9	0.28	0.15	99.54
LUMO+10	0.58	0.13	99.31
LUMO+11	0.21	1.20	98.58
LUMO+12	26.73	1.52	71.73

Table S10. The electron density distribution (%) in the molecular fragment of frontier molecular orbitals for complex **4** obtained from the Mulliken population analysis (MPA) method.

	Os	pz2py	py	CO
HOMO-2	4.67	92.73	0.75	1.86
HOMO-1	13.74	83.31	0.42	2.55
HOMO	21.64	73.59	0.51	4.25
LUMO	4.43	87.47	1.51	6.57
LUMO+1	2.21	3.85	89.15	4.75
LUMO+2	0.58	94.46	3.24	1.71

Table S11. The electron density distribution (%) in the molecular fragment of frontier molecular orbitals for complex **5** obtained from the Mulliken population analysis (MPA) method.

	Os	pz2py	bpy	CO
HOMO-2	9.40	87.85	0.72	2.05
HOMO-1	75.22	9.03	14.36	1.37
HOMO	32.81	57.52	2.63	7.07
LUMO	5.09	0.98	92.88	1.02
LUMO+1	4.12	91.50	1.00	3.35

Table S12. The electron density distribution (%) in the molecular fragment of frontier molecular orbitals for complex **6** obtained from the Mulliken population analysis (MPA) method.

	Os	pz2py	tpy	CO
HOMO-1	70.22	15.57	13.97	0.25
HOMO	40.94	48.15	2.88	8.04
LUMO	4.61	1.39	92.96	1.01
LUMO+1	4.37	84.45	8.41	2.76