Electronic Supplementary Material (ESI) for Dalton Transactions.

Metal-ligand synergy driven functionalisation of alkylene linked bis(aldimine) on diruthenium(II) platform. Cyclisation *versus* oxygenation

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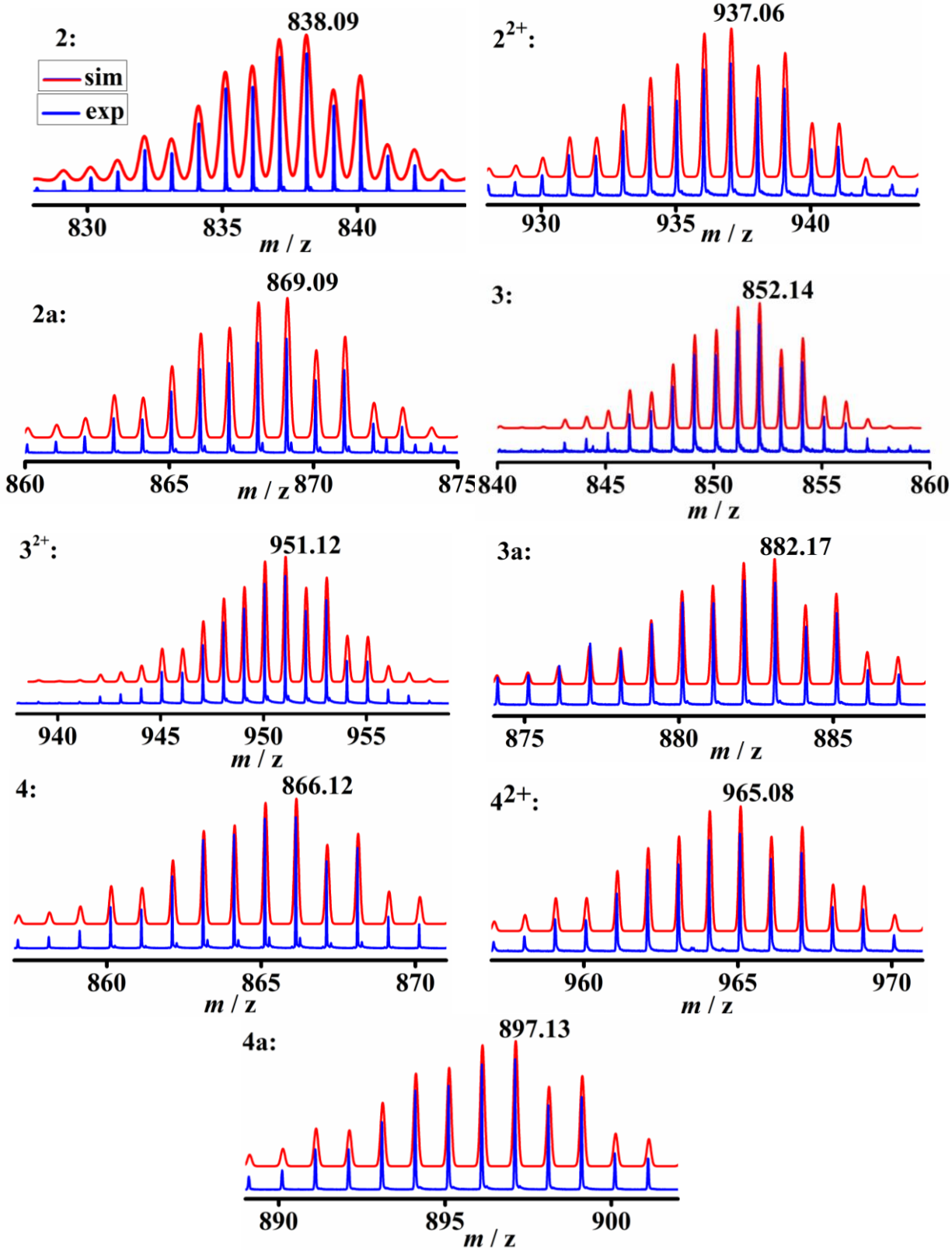


Fig. S1a Experimental and simulated ESI(+) mass spectrometry of **2**, $\{[\mathbf{2}](ClO_4)_2\text{-}ClO_4\}^+$, $\{\mathbf{2a}+H\}^+$, $\{\mathbf{3}+H\}^+$, $\{[\mathbf{3}](ClO_4)_2\text{-}ClO_4\}^+$, **3a**, **4**, $\{[\mathbf{4}](ClO_4)_2\text{-}ClO_4\}^+$and $\{\mathbf{4a}+2H\}^+$ in CH_3CN.

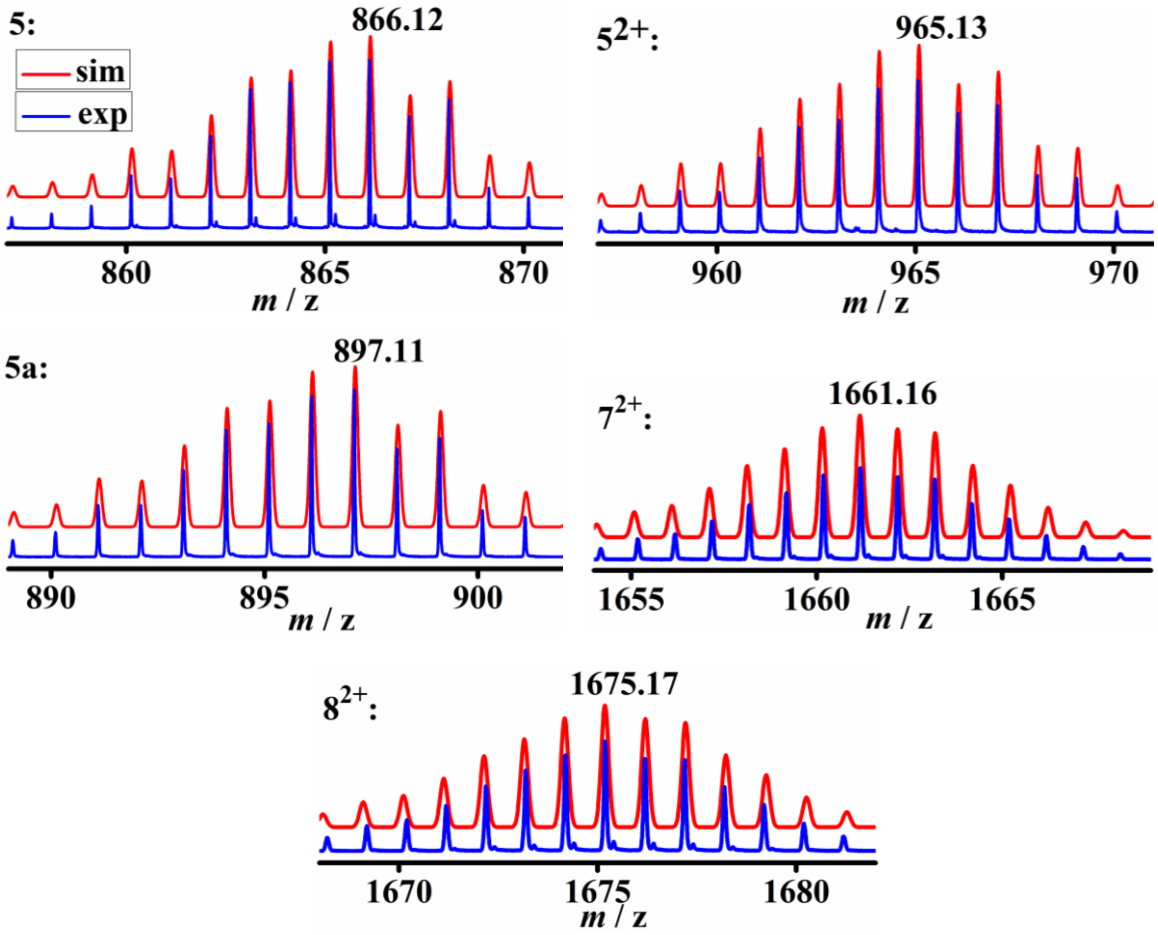
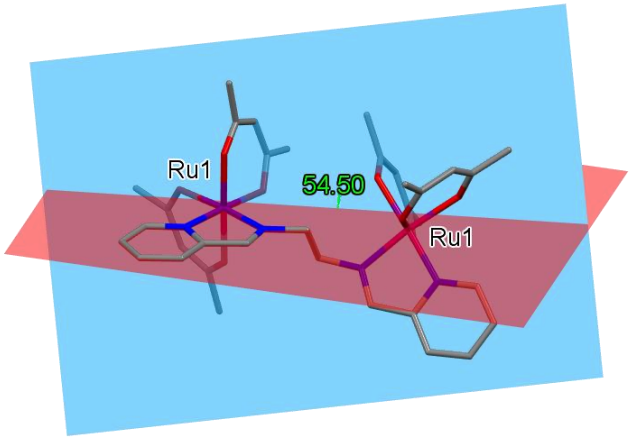
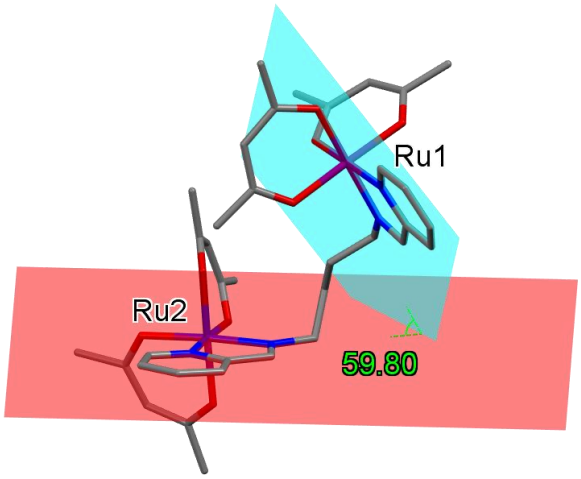


Fig. S1b Experimental and simulated ESI(+) mass spectrometry of **5**, {[**5**]$(ClO_4)_2$-ClO_4}$^+$, {**5a**+2H}$^+$, {[**7**]$(ClO_4)_2$-ClO_4}$^+$ and {[**8**]$(ClO_4)_2$-ClO_4}$^+$ in CH_3CN.

2^{2+}:



4^{2+}:



5^{2+}:

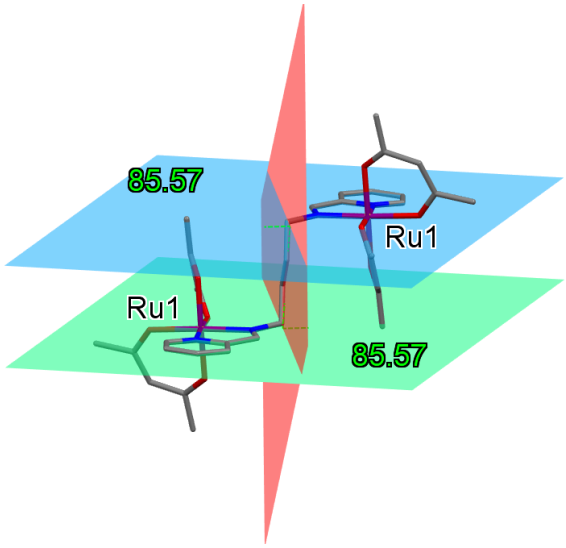
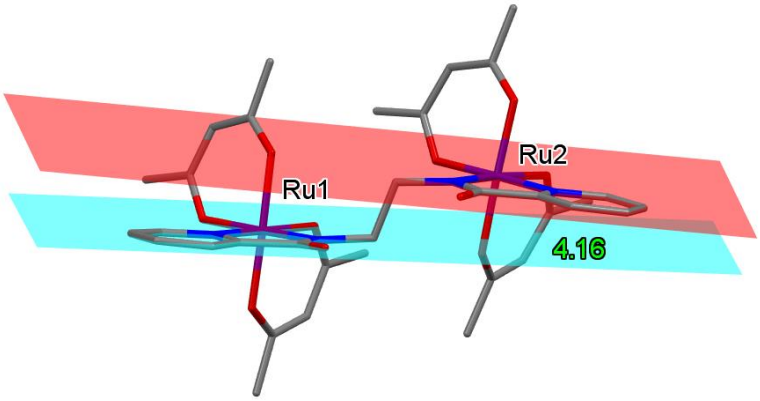


Fig. S2a Torsional angle (deg) between the planes.

2a:



3a:

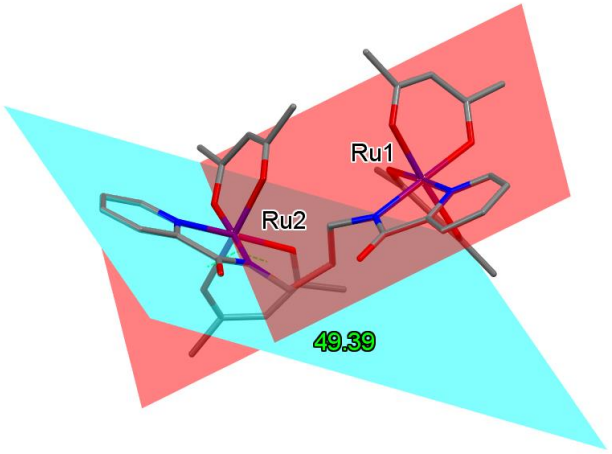
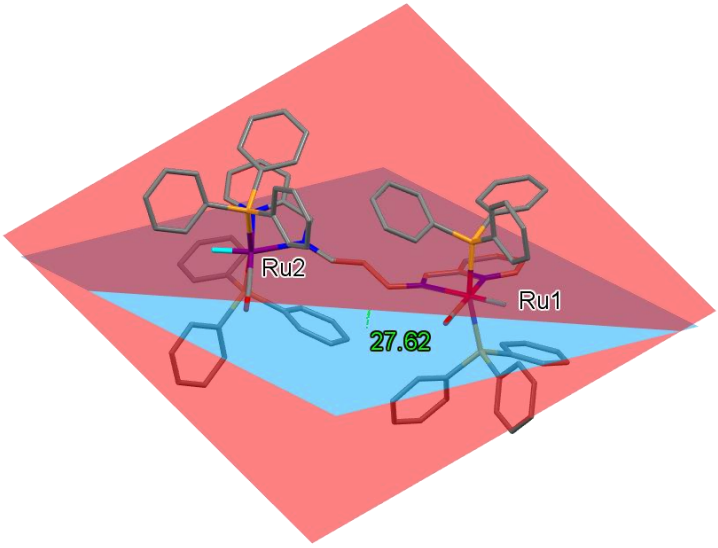


Fig. S2b Torsional angle (deg) between the planes.

7^{2+}:



8^{2+}:

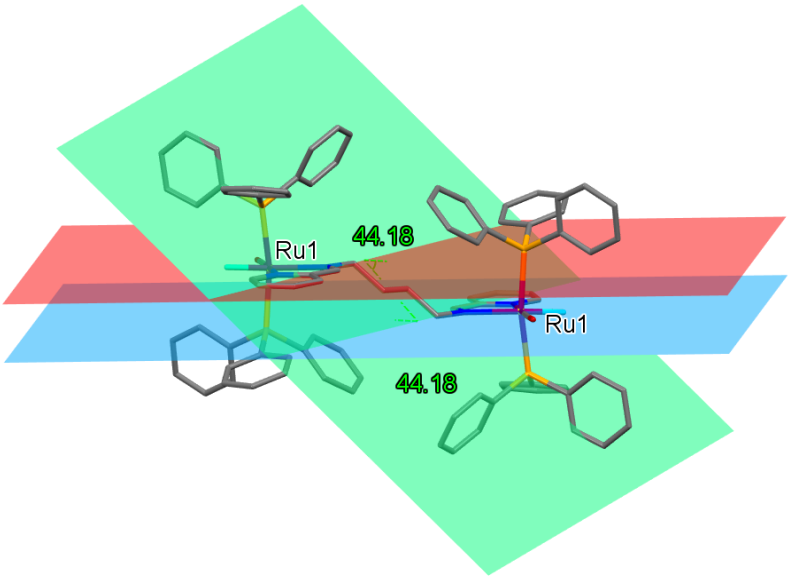


Fig. S3 Torsional angle (deg) between the planes.

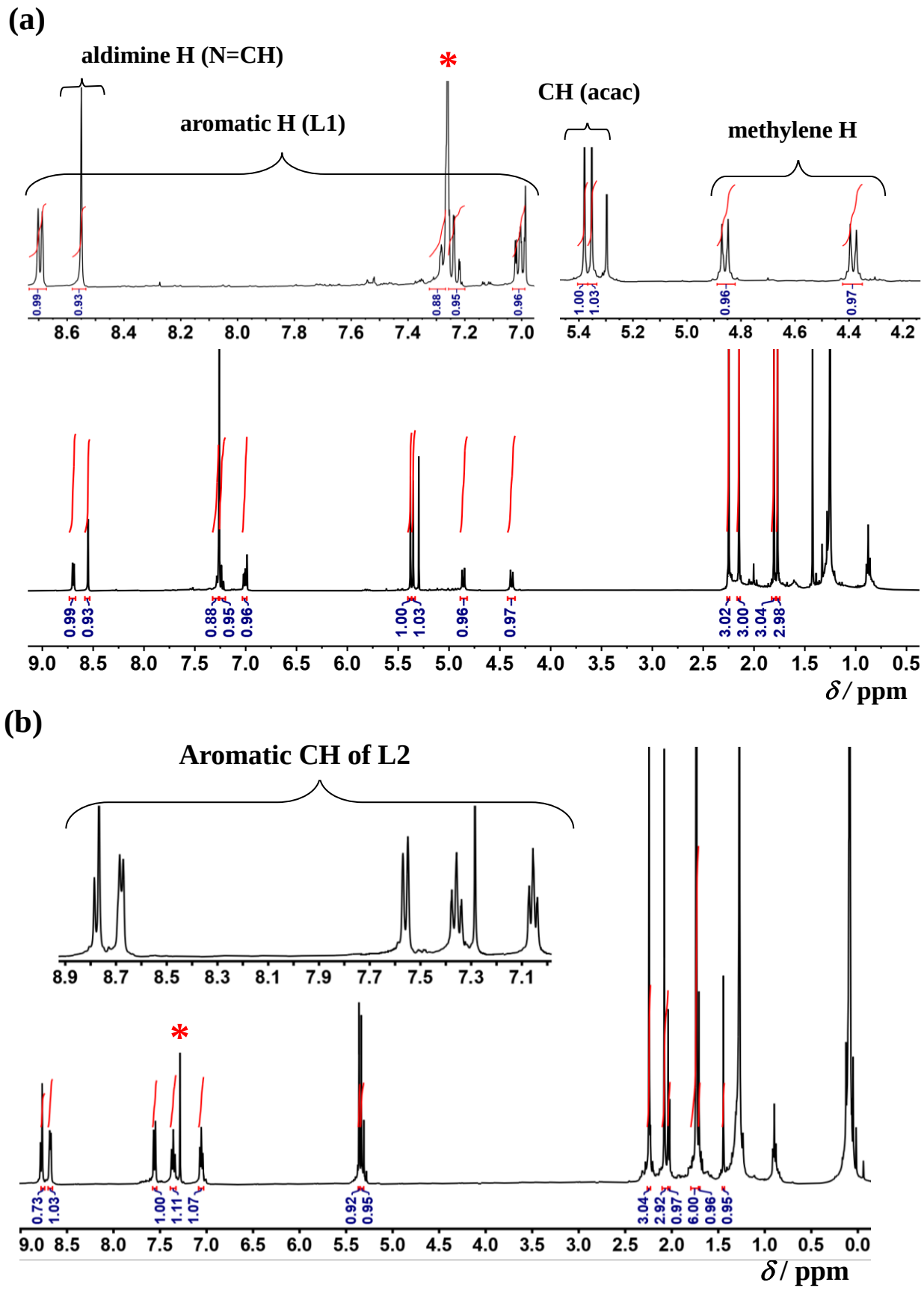


Fig. S4a ^{1}H NMR of (a) **2** and (b) **3** in $CDCl_3$ with TMS (δ = 0 ppm) as internal standard. Inset shows the segmented spectrum. (*) stands for solvent impurity.

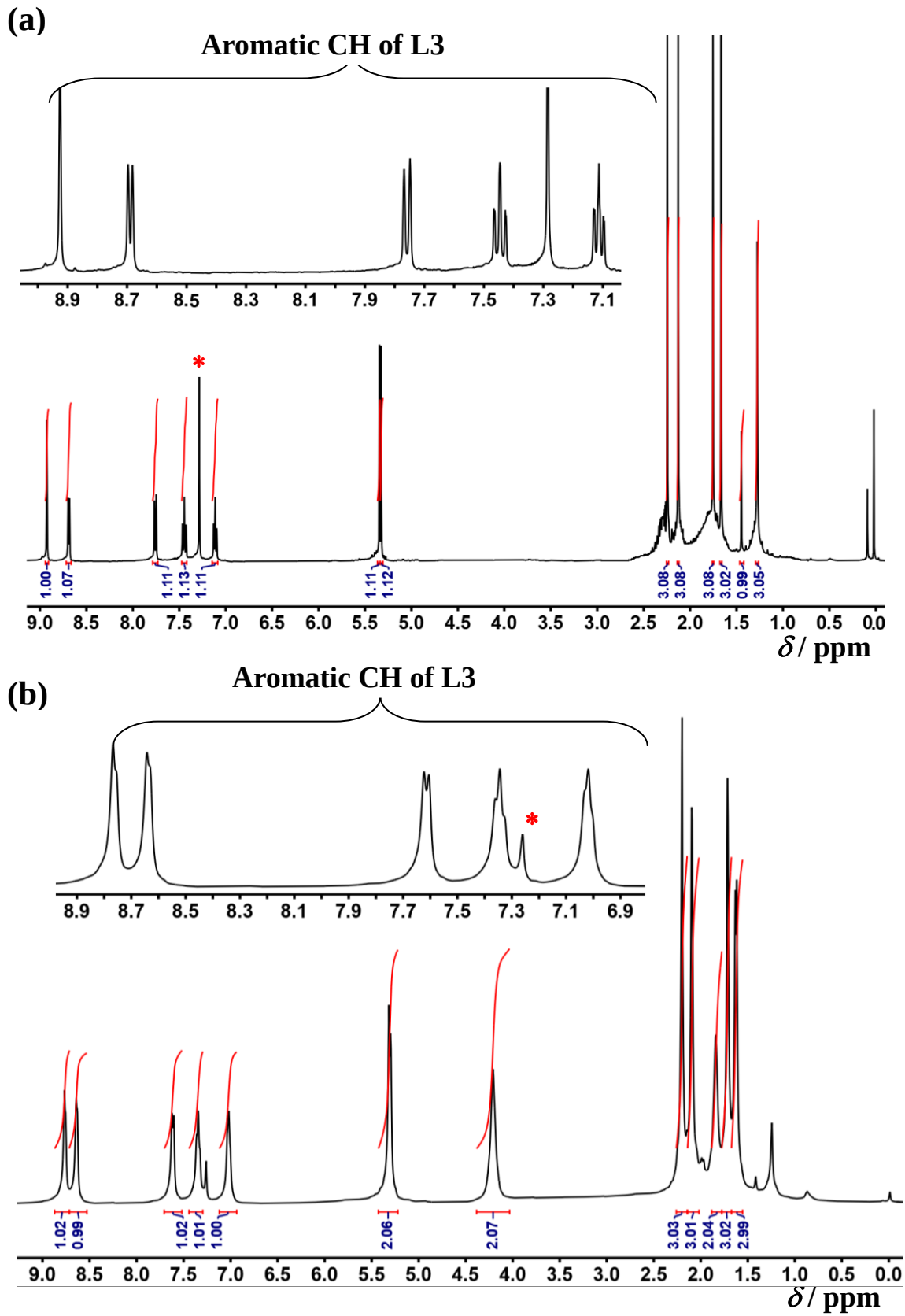


Fig. S4b ^{1}H NMR of (a) **4** and (b) **5** in $CDCl_3$ with TMS (δ = 0 ppm) as internal standard. Inset shows the segmented spectrum. (*) stands for solvent impurity.

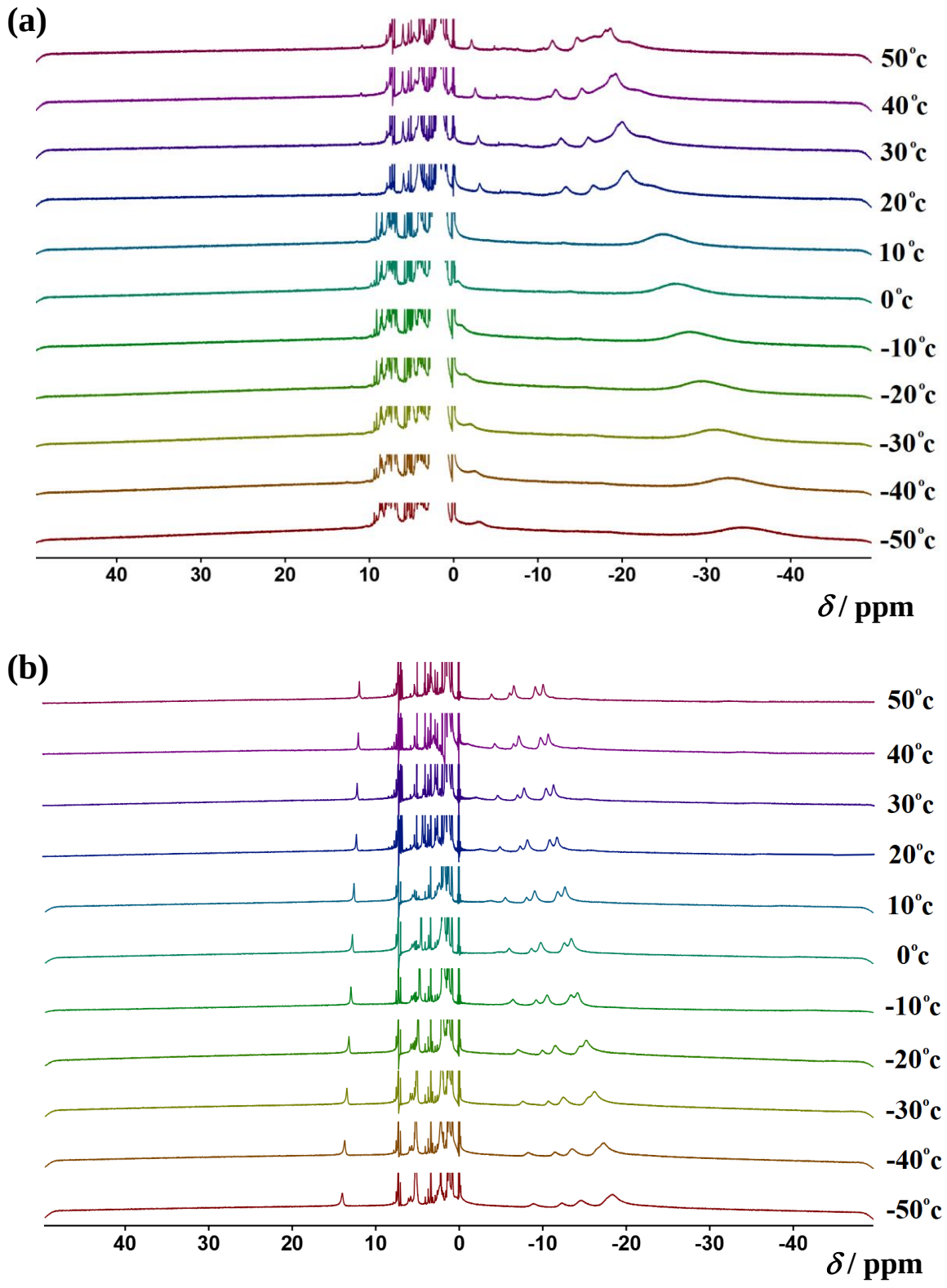


Fig. S5a Variable temperature ^{1}H NMR of (a) [**2**](ClO$_4$)$_2$ and (b) **2a** in CDCl$_3$ with TMS (δ = 0 ppm) as internal standard.

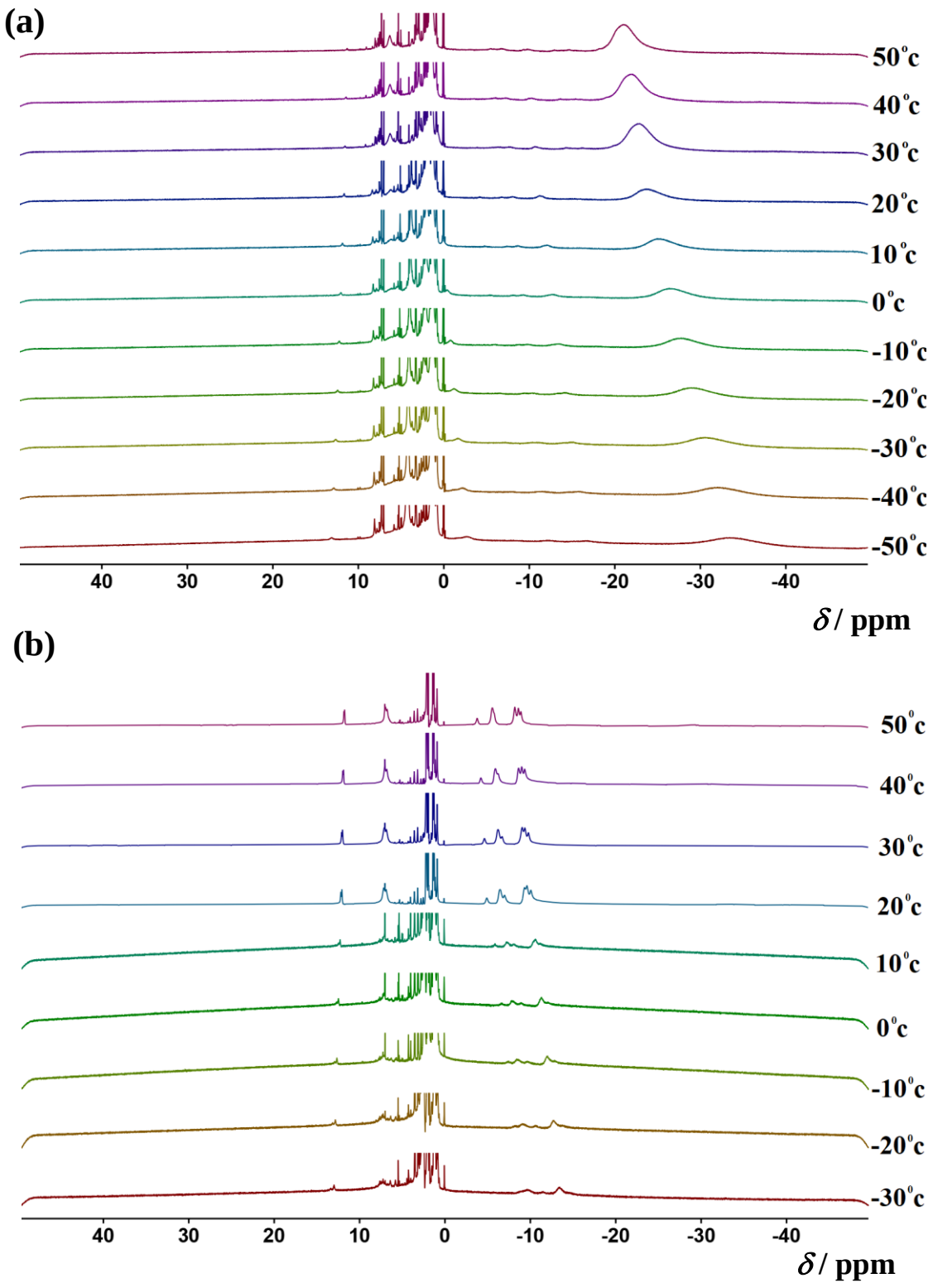


Fig. S5b Variable temperature ^{1}H NMR of (a) [**3**](ClO$_4$)$_2$ and (b) **3a** in CDCl$_3$ with TMS (δ = 0 ppm) as internal standard.

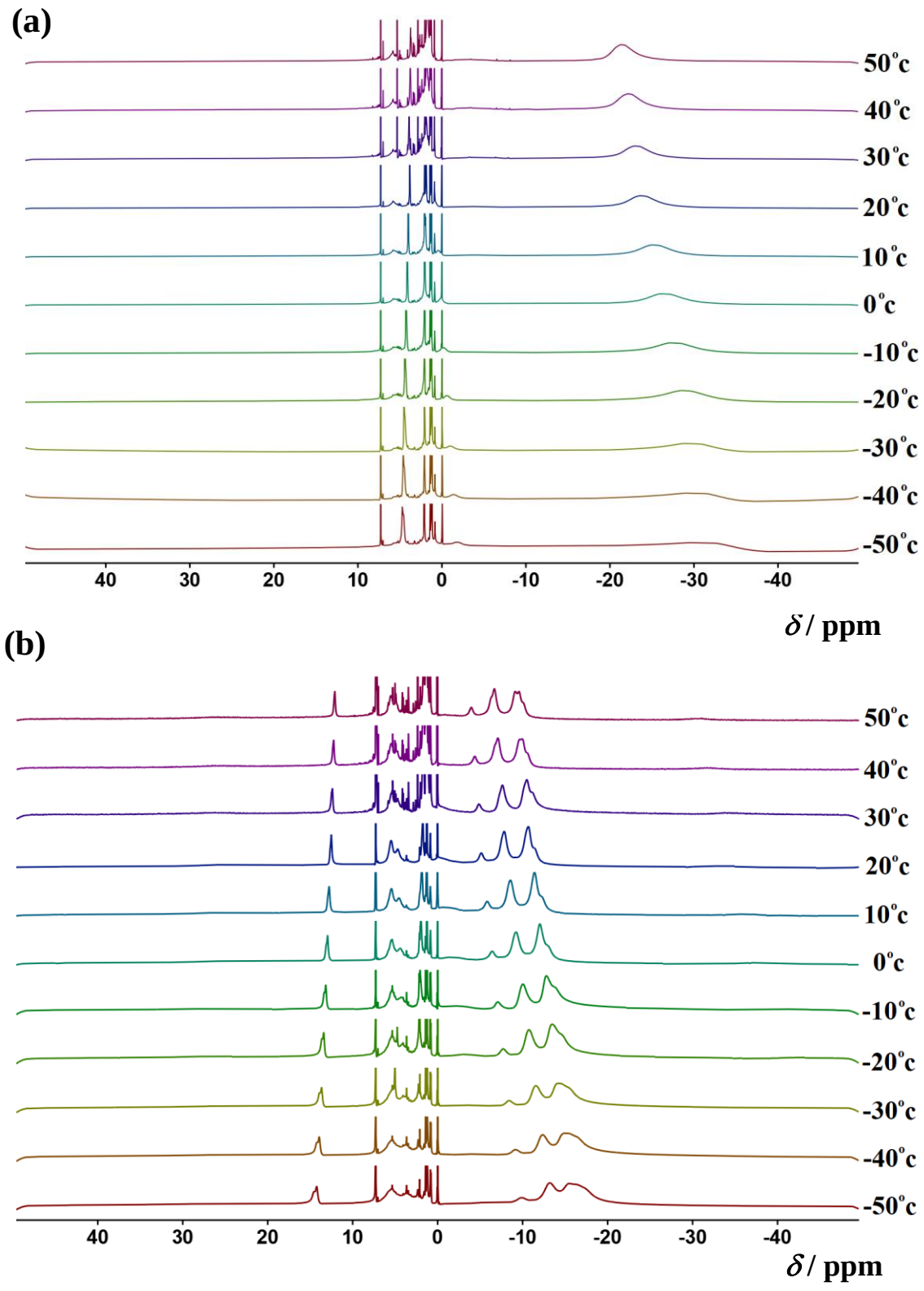


Fig. S5c Variable temperature ^{1}H NMR of (a) [**4**](ClO$_4$)$_2$ and (b) **4a** in CDCl$_3$ with TMS (δ = 0 ppm) as internal standard.

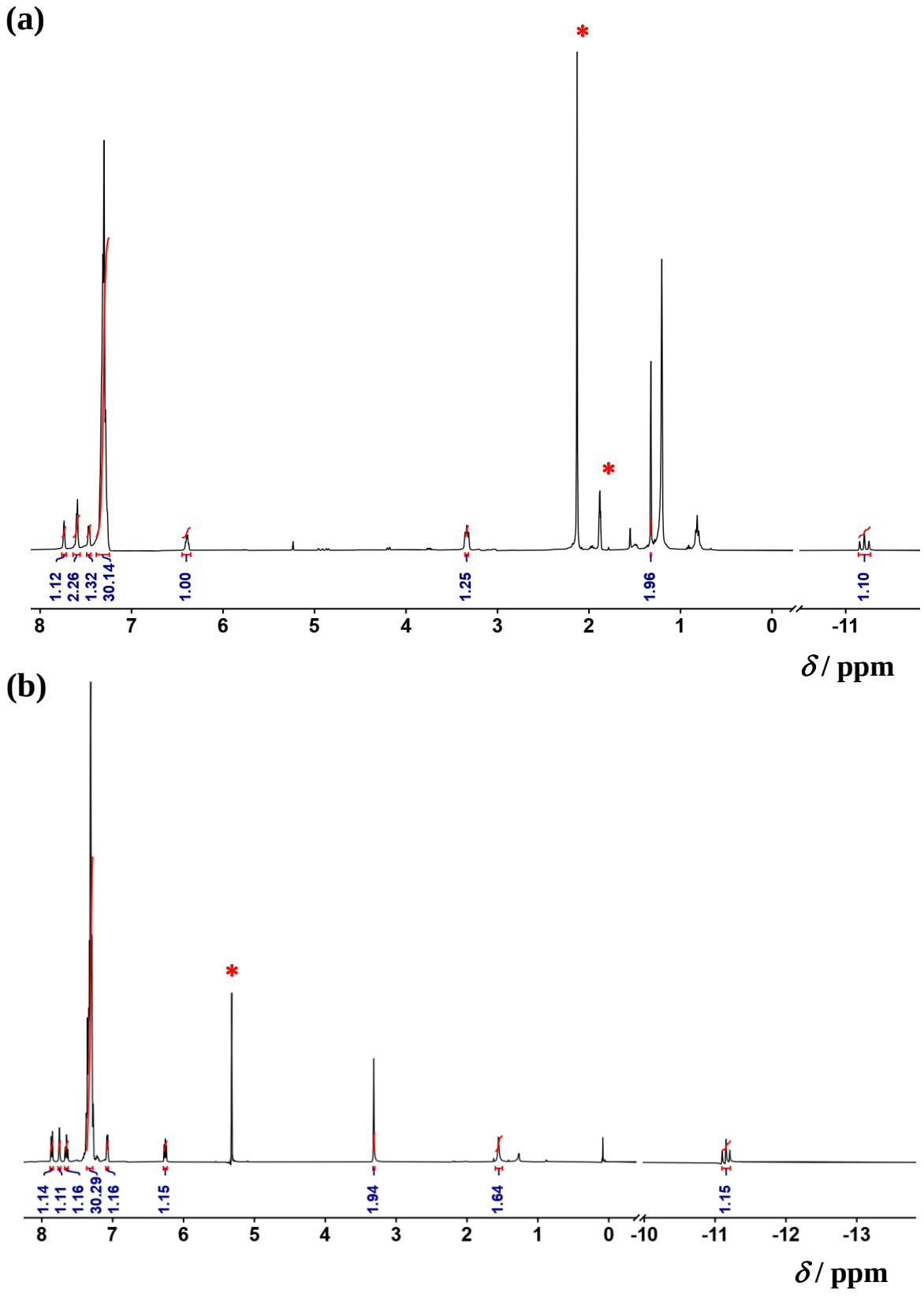


Fig. S6a ^{1}H NMR of (a) [**7**](ClO$_4$)$_2$ and (b) [**8**](ClO$_4$)$_2$ in CD_3CN and CD_2Cl_2, respectively, with TMS (δ = 0 ppm) as internal standard. (*) stands for solvent impurity.

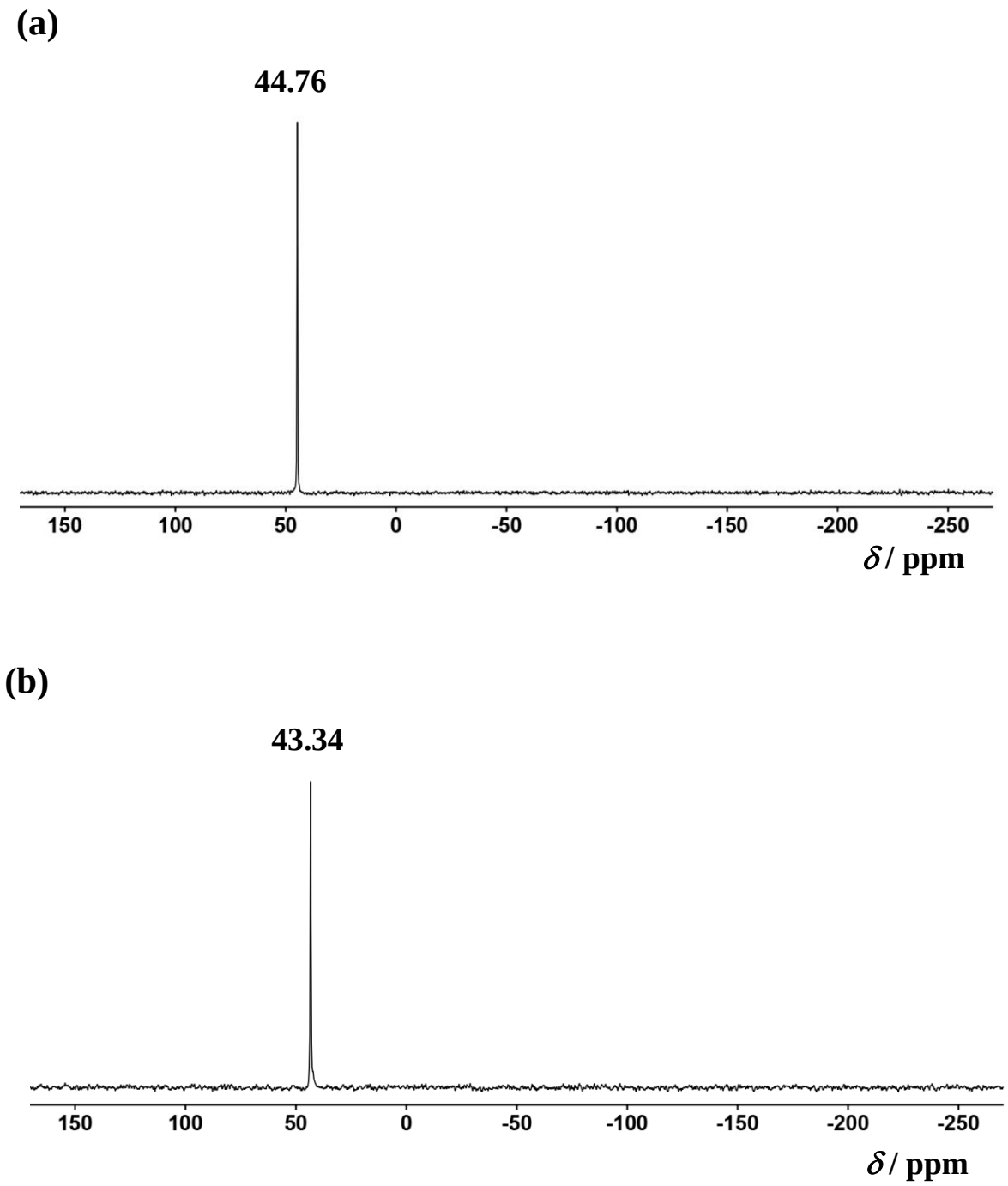


Fig. S6b ^{31}P NMR of (a) [**7**]$(ClO_4)_2$ and (b) [**8**]$(ClO_4)_2$ in CD_3CN and CD_2Cl_2, respectively, with TMS (δ = 0 ppm) as internal standard.

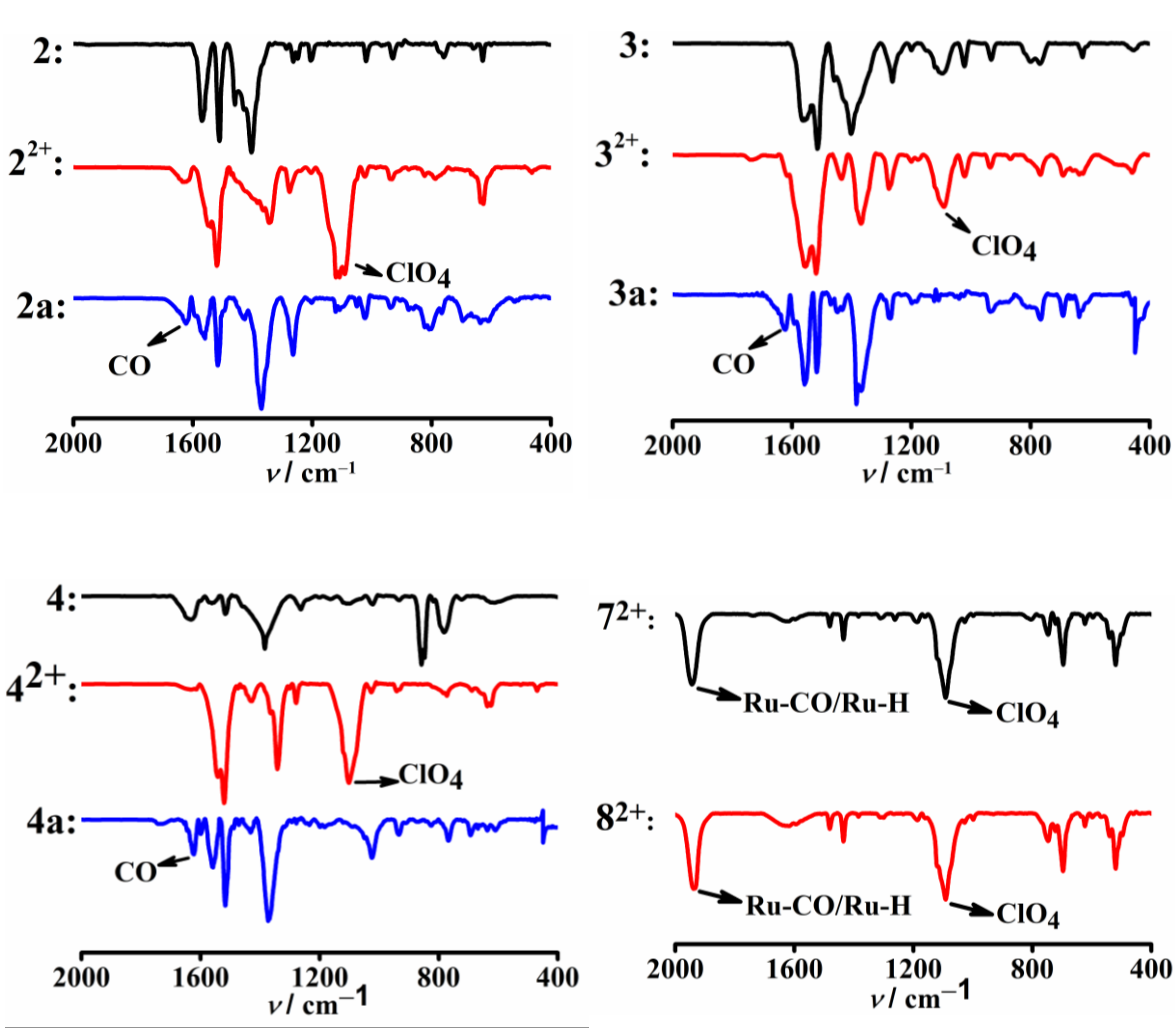


Fig. S7 FT-IR spectra as KBr pellets.

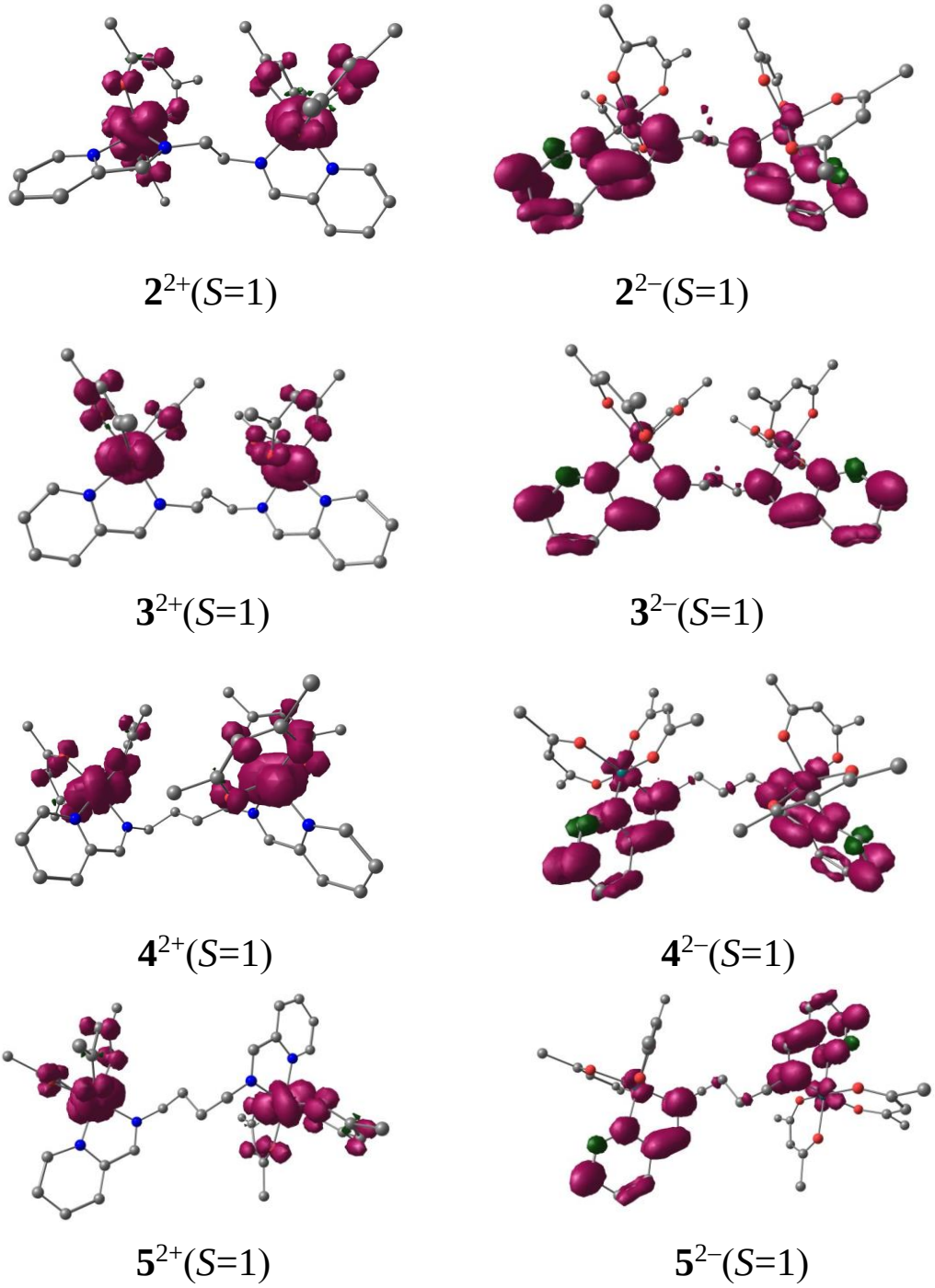


Fig. S8a DFT calculated Mulliken spin density plots for $\mathbf{2}^n$, $\mathbf{3}^n$, $\mathbf{4}^n$ and $\mathbf{5}^n$ (n =+2, –2).

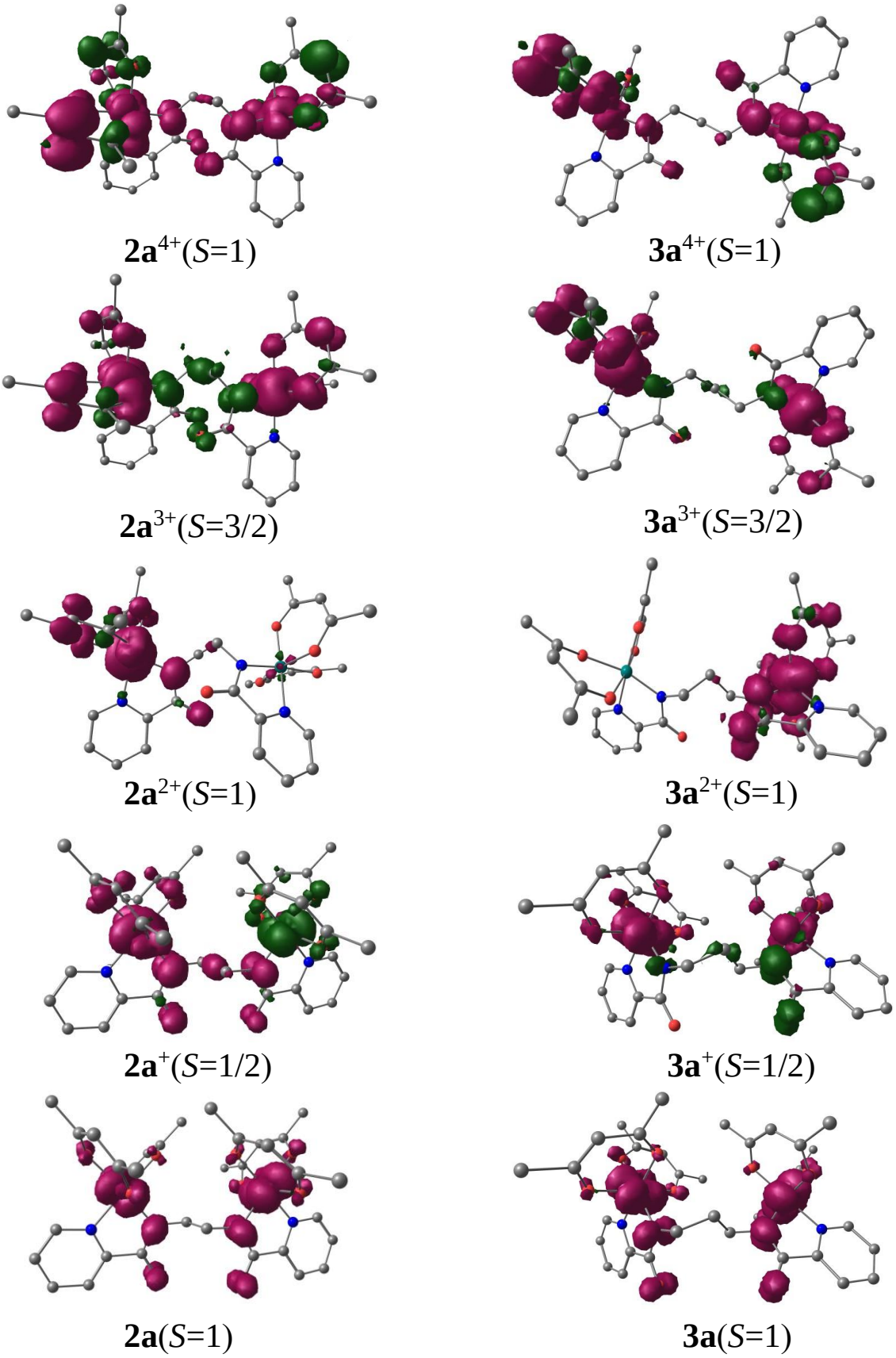
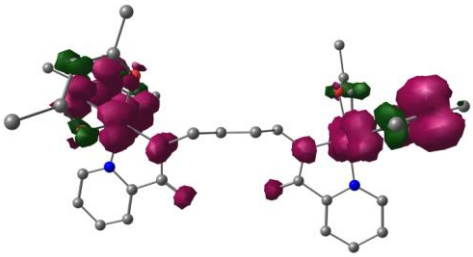
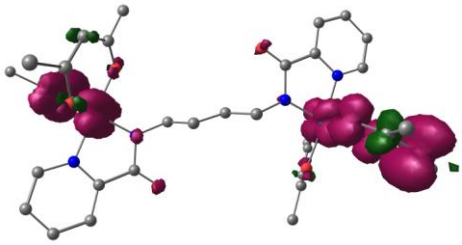


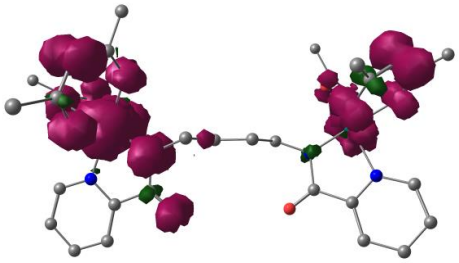
Fig. S8b DFT calculated Mulliken spin density plots for **2a**^{n} and **3a**^{n} (n =+4, +3, +2, +1, 0).



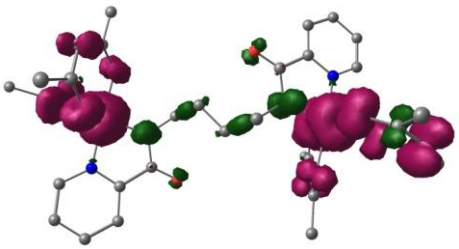
4a^{4+}(S=1)



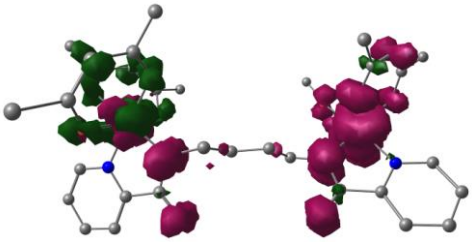
5a^{4+}(S=1)



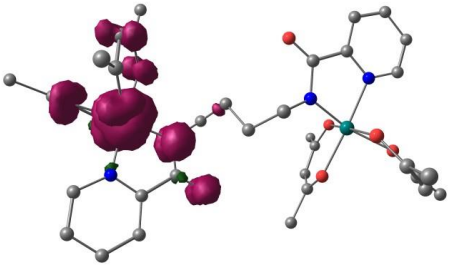
4a^{3+}(S=3/2)



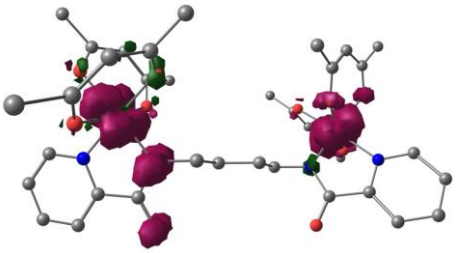
5a^{3+}(S=3/2)



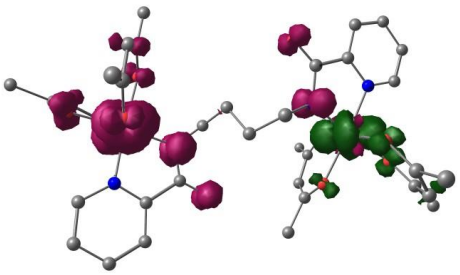
4a^{2+}(S=1)



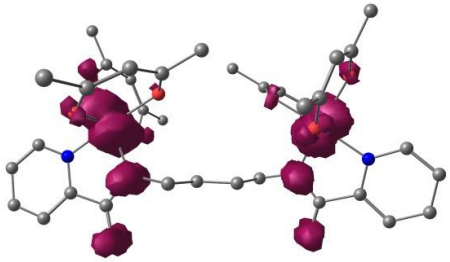
5a^{2+}(S=1)



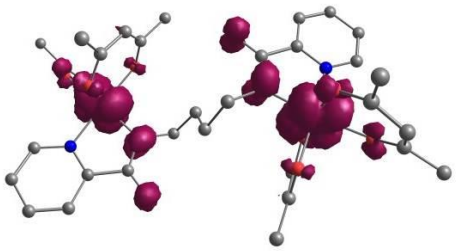
4a^{+}(S=1/2)



5a^{+}(S=1/2)



4a(S=1)



5a(S=1)

Fig. S8c DFT calculated Mulliken spin density plots for **4a**^{n} and **5a**^{n} (n =+4, +3, +2, +1, 0).

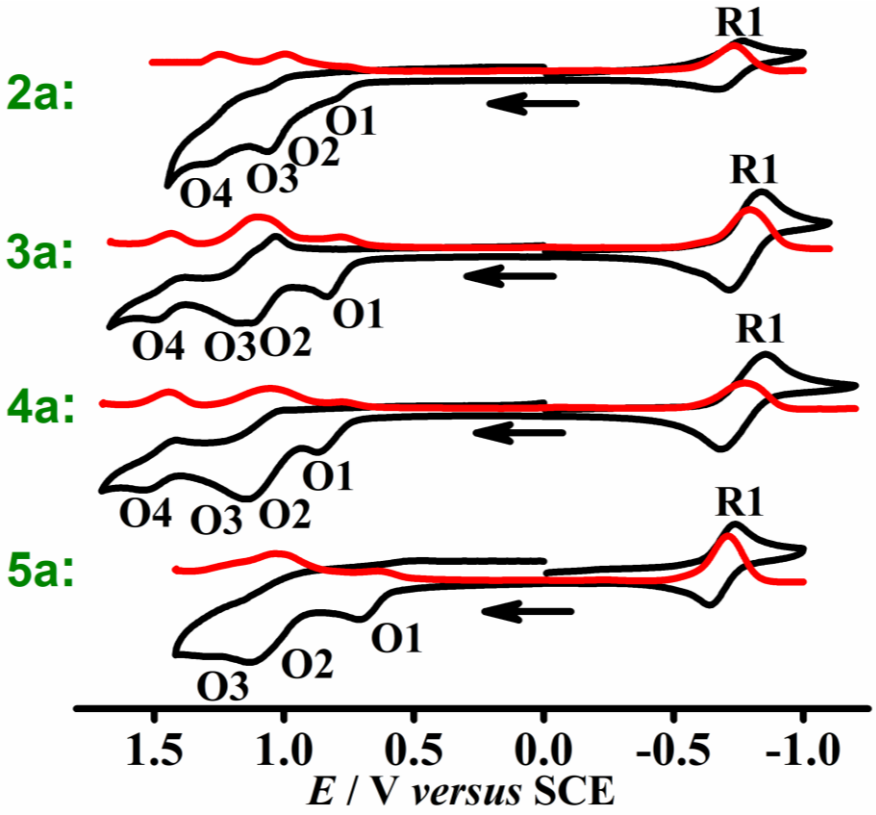


Fig. S9 Cyclic (red) and differential pulse (black) voltammograms of **2a**, **3a**, **4a** and **5a** in CH_3CN.

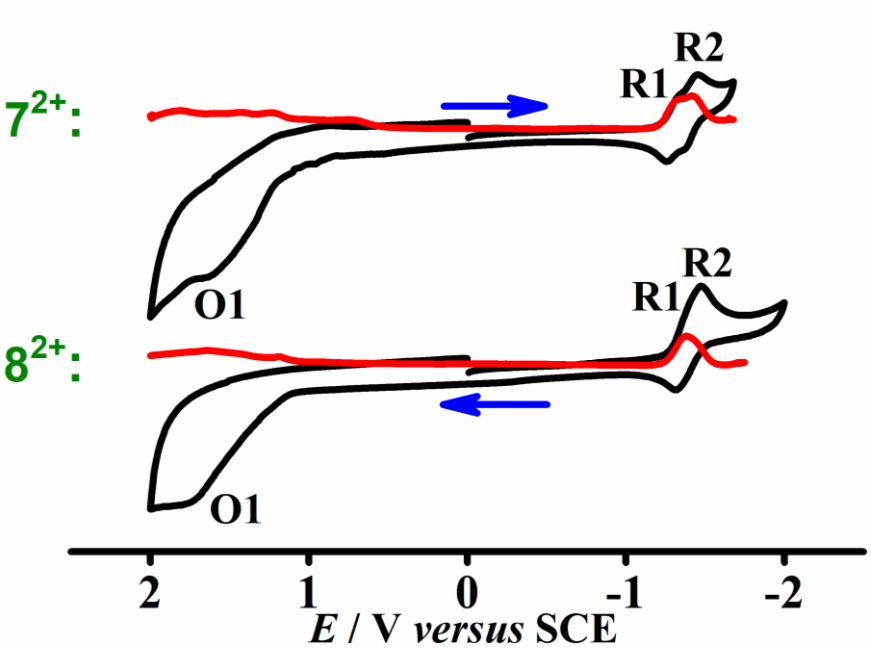


Fig. S10 Cyclic (red) and differential pulse (black) voltammograms of [**7**]$(ClO_4)_2$ and [**8**]$(ClO_4)_2$ in CH_3CN.

Scheme S1 Electronic forms of **2a**^{n}-**5a**^{n} (n= +4, +3, +2, +1, 0, -2)

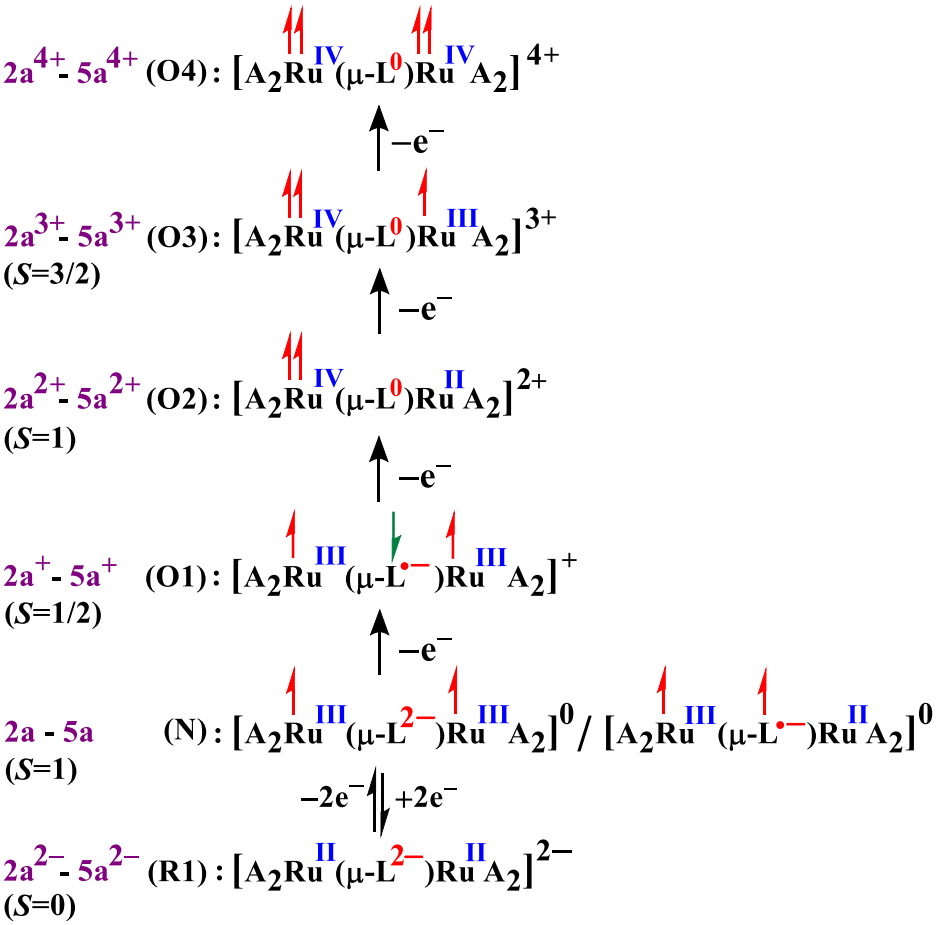


Table S1a Selected crystallographic parameters

Complex	[**2**](ClO$_4$)$_2$•2CH$_2$Cl$_2$	**2a**	**3a**	[**4**](ClO$_4$)$_2$•3CH$_2$Cl$_2$	[**5**](ClO$_4$)$_2$•4CHCl$_3$
empirical formula	$C_{36}H_{46}N_4O_{16}Cl_6Ru_2$	$C_{34}H_{40}N_4O_{10}Ru_2$	$C_{35}H_{42}N_4O_{10}Ru_2$	$C_{39}H_{52}N_4O_{16}Cl_8Ru_2$	$C_{40}H_{50}N_4O_{16}Cl_{14}Ru_2$
formula weight	1205.61	866.84	880.86	1318.58	1541.28
crystal system	Monoclinic	Triclinic	Triclinic	Monoclinic	Monoclinic
space group	*C*2/c	*P*-1	*P*-1	*P*2$_1$/c	*C*2/c
a (Å)	17.9403(5)	12.5742(10)	10.986(2)	12.6627(3)	22.5574(6)
b (Å)	12.6420(2)	12.8511(11)	13.108(3)	36.9819(7)	13.0011(3)
c (Å)	22.9581(7)	13.6116(12)	15.434(3)	12.1433(3)	21.5012(6)
α (deg)	90	72.100(8)	67.924(3)	90	90
β (deg)	114.810	69.913(8)	72.987(4)	103.949(2)	97.806(3)
γ (deg)	90	81.135(7)	77.492(4)	90	90
V (Å3)	4726.3(3)	1962.8(3)	1955.3(7)	5518.9(2)	6247.2(3)
Z	4	2	2	4	4
μ (mm^{-1})	1.049	0.825	0.830	0.999	1.144
T (K)	150.00(10)	150.00(10)	105.0	150.00(10)	150.00(10)
ρ_{cald}(g cm^{-3})	1.694	1.467	1.496	1.587	1.639
F(000)	2432.0	880.0	896.0	2664.0	3088.0
θ range(deg)	1.954 to 33.786	1.656 to 24.999	1.953 to 25.178	1.657 to 24.999	1.812 to 24.999
data/restraints/parameter	8096/ 0/ 293	6910/ 0/ 459	7005/ 0/ 468	9712/ 7/ 630	5509/ 12/ 347
R1, wR2 [I>2σ(I)]	0.0413, 0.0841	0.0677, 0.1683	0.0816, 0.1530	0.0577, 0.1558	0.0697, 0.1819
R1, wR2(all data)	0.0655, 0.0933	0.0906, 0.1878	0.1230, 0.1693	0.0752, 0.1693	0.0836, 0.1912
GOF	1.039	1.057	1.090	1.031	1.092
largest diff. peak/hole [e Å^{-3}]	1.10/ -0.83	2.09/ -0.64	1.56/.–1.15	0.96/ -0.57	1.54/ -0.94

Table S1b Selected crystallographic parameters

Complex	[**7**]$(ClO_4)_2$	[**8**]$(ClO_4)_2$• $2CH_2Cl_2$
empirical formula	$C_{89}H_{78}N_4O_{10}Cl_2P_4Ru_2$	$C_{92}H_{84}N_4O_{10}Cl_6P_4Ru_2$
formula weight	1760.47	1944.35
crystal system	Monoclinic	Triclinic
space group	$P2_1/c$	P-1
a (Å)	14.3963(3)	13.0632(3)
b (Å)	21.3283(5)	313.3460(2)
c (Å)	27.8750(6)	14.6777(2)
α (deg)	90	64.066(2)
β (deg)	91.558(2)	72.589(2)
γ (deg)	90	82.031(2)
V (Å3)	8555.8(3)	2195.72(8)
Z	4	1
μ (mm^{-1})	0.549	0.660
T (K)	150.00(10)	150.00(10)
ρ_{cald}(g cm^{-3})	1.367	1.470
F(000)	3608.0	994.0
θ range(deg)	1.707 to 25.000	1.601 to 25.000
data/restraints/parameters	15054/ 34/ 1006	7746/ 0/ 536
R1, wR2 [I>2σ(I)]	0.0563, 0.1501	0.0473, 0.1263
R1, wR2(all data)	0.0722, 0.1618	0.0549, 0.1334
GOF	1.037	1.051
largest diff. peak/hole [e $Å^{-3}$]	2.10/ -0.88	1.29/ -1.29

Table S2a Selected experimental and DFT-calculated bond lengths (Å)

Bond lengths (Å)	[**2**](ClO4)2		**2a**		**3a**		[**4**](ClO4)2		[**5**](ClO4)2	
	X-ray	DFT	X-ray	DFT	X-ray	DFT	X-ray	DFT	X-ray	DFT
Ru1-N1	2.038(2)	2.090	2.031(6)	2.075	2.029(8)	2.075	2.042(4)	2.090	2.044(4)	2.088
Ru1-N2	2.045(19)	2.109	2.022(5)	1.999	2.006(7)	1.999	2.034(5)	2.099	2.042(5)	2.097
Ru1-O1	2.007(17)	2.044	2.035(5)	2.085	2.037(6)	2.090	2.018(4)	2.042	2.013(4)	2.041
Ru1-O2	1.996(17)	2.030	1.978(5)	2.041	2.016(6)	2.041	1.996(4)	2.041	2.004(4)	2.033
Ru1-O3	1.996(17)	2.033	1.997(4)	2.070	2.017(6)	2.071	1.993(4)	2.025	1.991(4)	2.028
Ru1-O4	2.015(17)	2.038	2.042(5)	2.112	2.030(6)	2.110	2.016(4)	2.038	2.017(5)	2.039
Ru2-N3	-	-	2.001(5)	1.998	2.018(8)	2.000	2.040(5)	2.089	-	-
Ru2-N4	-	-	2.031(5)	2.075	2.020(8)	2.070	2.033(5)	2.092	-	-
Ru2-O5	-	-	2.031(5)	2.111	2.031(6)	2.111	2.011(4)	2.046	-	-
Ru2-O6	-	-	2.007(4)	2.071	2.020(6)	2.063	1.991(4)	2.036	-	-
Ru2-O7	-	-	1.979(5)	2.040	1.996(6)	2.043	1.997(4)	2.028	-	-
Ru2-O8	-	-	2.023(5)	2.085	2.048(6)	2.098	2.012(4)	2.046	-	-
N2-C6	1.290(3)	1.291	1.318(9)	1.369	1.327(11)	1.367	1.282(7)	1.291	1.282(9)	1.291
N3-C10	-	-	-	-	1.340(11)	1.368	-	-	-	-
N3-C9	-	-	1.338(9)	1.369	-	-	-	-	-	-
N3-C11	-	-	-	-	-	-	1.268(8)	1.291	-	-
C6-O9	-	-	1.248(8)	1.232	1.248(10)	1.234	-	-	-	-
C10-O10	-	-	-	-	1.262(10)	1.234	-	-	-	-
C9-O10	-	-	1.248(8)	1.232	-	-	-	-	-	-
Ru1---Ru2	6.541	6.339	6.081	6.106	6.541	6.614	7.361	7.408	7.784	8.397

Table S2b Selected experimental and DFT-calculated bond lengths (Å)

Bond lengths (Å)	[**7**](ClO$_4$)$_2$		[**8**](ClO$_4$)$_2$	
	X-ray	DFT	X-ray	DFT
Ru1-N1	2.122(4)	2.178	2.129(3)	2.242
Ru1-N2	2.185(4)	2.254	2.175(3)	2.323
Ru1-P1	2.3588(13)	2.453	2.3685(9)	2.532
Ru1-P2	2.3443(13)	2.459	2.3636(9)	2.533
Ru1-C16	1.851(5)	1.867	-	-
Ru1-C9	-	-	1.845(4)	1.927
Ru1-H	1.501(15)	1.596	1.540	1.65
Ru2-N3	2.180(4)	2.255	-	-
Ru2-N4	2.137(4)	2.179	-	-
Ru2-P3	2.3536(13)	2.459	-	-
Ru2-P4	2.3837(13)	2.453	-	-
Ru2-C17	1.837(6)	1.867	-	-
Ru2-H	1.501(15)	1.596	-	-
N2-C6	1.284(6)	1.286	1.281(5)	1.325
N3-C10	1.296(6)	1.286	-	-
C16-O1	1.135(6)	1.161	-	-
C17-O2	1.156(6)	1.161	-	-
C9-O1	-	-	1.157(5)	1.196
Ru1---Ru2	8.952	9.058	10.015	10.799

Table S3a Selected experimental and DFT-calculated bond angles (deg.)

	[**2**](ClO4)2		**2a**		**3a**		[**4**](ClO4)2		[**5**](ClO4)2	
Bond angles (degree)	X-ray	DFT	X-ray	DFT	X-ray	DFT	X-ray	DFT	X-ray	DFT
N1-Ru1-N2	79.01(8)	78.16	80.2(2)	80.75	79.9(3)	80.6	79.26(19)	78.10	79.1(2)	78.18
N1-Ru1-O1	177.76(8)	175.71	176.9(2)	177.33	176.2(3)	176.6	176.80(17)	175.5	178.0(2)	175.23
N1-Ru1-O2	89.36(7)	88.12	87.1(2)	86.35	88.1(3)	84.9	88.33(17)	89.39	89.17(18)	88.92
N1-Ru1-O3	91.26(8)	92.39	92.3(2)	92.94	92.2(3)	94.1	91.34(17)	91.60	92.16(19)	91.90
N1-Ru1-O4	94.03(8)	94.87	94.9(2)	94.99	97.6(3)	94.7	94.47(17)	95.41	94.7(2)	95.26
N2-Ru1-O1	98.99(8)	98.38	96.8(2)	97.11	96.5(3)	97.7	97.56(17)	97.60	98.9(2)	97.38
N2-Ru1-O2	91.07(7)	92.81	93.0(2)	93.20	94.9(3)	94.9	90.85(16)	95.72	90.99(19)	95.97
N2-Ru1-O3	88.20(8)	88.05	88.1(2)	90.39	89.0(3)	88.8	87.40(17)	85.53	88.7(2)	85.32
N2-Ru1-O4	173.04(8)	172.73	174.8(2)	175.18	177.3(3)	173.9	173.59(17)	171.98	173.7(2)	171.76
O1-Ru1-O2	91.68(7)	89.52	94.0(19)	92.19	93.4(2)	92.2	91.39(16)	89.67	91.26(18)	89.79
O1-Ru1-O3	87.67(7)	90.01	86.69(19)	88.67	86.5(3)	88.8	88.84(16)	89.42	87.39(19)	89.49
O1-Ru1-O4	87.97(7)	88.67	88.0(2)	87.20	86.0(2)	87.2	88.71(16)	88.98	87.30(19)	89.31
O2-Ru1-O3	178.93(7)	179.07	178.62(19)	176.18	176.1(2)	176.0	178.25(17)	178.54	178.53(18)	178.60
O2-Ru1-O4	88.95(7)	89.00	88.4(2)	88.77	85.9(2)	88.4	90.31(16)	88.87	89.87(19)	88.83
O3-Ru1-O4	91.87(7)	90.20	90.5(2)	87.55	90.2(2)	87.8	91.43(16)	89.98	90.64(19)	89.96
N3-Ru2-N4	-	-	80.5(2)	80.74	80.7(3)	81.0	79.0(2)	78.12	-	-
N3-Ru2-O5	-	-	174.0(2)	175.18	175.0(3)	176.1	174.31(19)	173.49	-	-
N3-Ru2-O6	-	-	87.8(2)	90.37	88.8(3)	90.5	86.22(18)	88.77	-	-
N3-Ru2-O7	-	-	94.6(2)	93.09	93.1(3)	93.6	92.85(18)	91.88	-	-
N3-Ru2-O8	-	-	96.2(2)	97.16	96.5(3)	96.4	97.0(2)	96.80	-	-
N4-Ru2-O5	-	-	94.0(2)	95.02	94.4(3)	95.4	95.82(18)	95.64	-	-

N4-Ru2-O6	-	-	92.88(19)	93.03	92.6(3)	89.3	92.63(18)	94.62	-	-
N4-Ru2-O7	-	-	86.9(2)	86.14	86.7(3)	88.6	88.14(18)	85.68	-	-
N4-Ru2-O8	-	-	176.7(2)	177.32	177.2(3)	177.4	175.92(19)	173.18	-	-
O5-Ru2-O6	-	-	90.2(2)	87.54	90.4(2)	87.9	91.74(17)	89.92	-	-
O5-Ru2-O7	-	-	87.4(2)	88.99	87.6(2)	87.9	89.27(17)	89.45	-	-
O5-Ru2-O8	-	-	89.3(2)	87.14	88.4(2)	87.2	88.24(17)	89.57	-	-
O6-Ru2-O7	-	-	177.56(18)	176.26	177.9(3)	175.1	178.66(17)	179.33	-	-
O6-Ru2-O8	-	-	86.87(19)	88.63	87.1(3)	90.5	87.61(17)	89.77	-	-
O7-Ru2-O8	-	-	93.5(2)	92.35	93.6(2)	91.8	91.54(17)	89.99	-	-
N2-C6-O9	-	-	127.6(7)	126.86	128.3(8)	126.8	-	-	-	-
N3-C10-O10	-	-	-	-	127.1(8)	127.1	-	-	-	-
N3-C9-O10	-	-	127.1(7)	126.96	-	-	-	-	-	-

Table S3b Selected experimental and DFT-calculated bond angles (deg.)

Bond angles (degree)	[**7**](ClO4)2		[**8**](ClO4)2	
	X-ray	DFT	X-ray	DFT
N1-Ru1-N2	76.32(14)	73.73	75.55(11)	75.03
N1-Ru1-P1	90.96(11)	91.49	90.37(8)	92.16
N1-Ru1-P2	90.81(11)	91.90	90.31(8)	91.28
N1-Ru1-C16	177.67(19)	177.40	-	-
N1-Ru1-C9	-	-	179.11(14)	178.57
N1-Ru1-HA	93.53	92.64	91.29	92.29
N2-Ru1-P1	102.87(10)	96.05	95.17(8)	95.62
N2-Ru1-P2	92.82(10)	98.39	94.49(8)	98.07
N2-Ru1-C16	101.90(18)	102.66	-	-
N2-Ru1-C9	-	-	104.56(15)	103.59
N2-Ru1-HA	168.60	167.29	166.41	167.31
P1-Ru1-P2	164.19(5)	165.55	170.18(3)	166.31
P1-Ru1-C16	87.97(16)	88.82	-	-
P1-Ru1-C9	-	-	90.50(12)	88.30
P1-Ru1-HA	71.67	82.65	88.95	84.27
P2-Ru1-C16	90.78(16)	88.41	-	-
P2-Ru1-C9	-	-	88.80(12)	88.58
P2-Ru1-HA	92.53	83.17	81.25	82.35
C16-Ru1-HA	88.10	89.96	-	-
C9-Ru1-HA	-	-	88.67	89.10
N3-Ru2-N4	75.99(15)	74.71	-	-
N3-Ru2-P3	96.40(11)	97.91	-	-
N3-Ru2-P4	92.49(11)	96.15	-	-
N3-Ru2-C17	105.1(2)	102.68	-	-
N3-Ru2-H	161.50	167.26	-	-
N4-Ru2-P3	90.22(12)	91.85	-	-
N4-Ru2-P4	88.96(11)	91.44	-	-
N4-Ru2-C17	177.3(2)	177.39	-	-
N4-Ru2-H	85.62	92.60	-	-
P3-Ru1-P4	170.59(5)	165.92	-	-
P3-Ru2-C17	87.25(16)	88.50	-	-
P3-Ru2-H	81.69	83.42	-	-
P4-Ru2-C17	93.40(16)	88.82	-	-
P4-Ru2-H	88.91	87.76	-	-
C17-Ru2-H	93.17	90.00	-	-

Table S4 Determination of solution magnetic moment (Evans method) of $\mathbf{2}^{2+}$, **2a**, $\mathbf{3}^{2+}$, **3a**, $\mathbf{4}^{2+}$, **4a**, $\mathbf{5}^{2+}$, **5a**

Solution magnetic moment of Ru^{III}-Ru^{III} derived $\mathbf{2}^{2+}$, **2a**, $\mathbf{3}^{2+}$, **3a**, $\mathbf{4}^{2+}$, **4a**, $\mathbf{5}^{2+}$and **5a** were determined by Evans method. Pure $\mathbf{2}^{2+}$, **2a**, $\mathbf{3}^{2+}$, **3a**, $\mathbf{4}^{2+}$, **4a**, $\mathbf{5}^{2+}$and **5a** in $CDCl_3$, $CDCl_3$, $(CD_3)_2SO$, $CDCl_3$, $CDCl_3$, $CDCl_3$, $(CD_3)_2SO$ and $(CD_3)_2SO$ were taken in a 5 mm NMR tube. In another coaxial NMR tube only reference $CDCl_3$, $CDCl_3$, $(CD_3)_2SO$, $CDCl_3$, $CDCl_3$, $CDCl_3$, $(CD_3)_2SO$ and $(CD_3)_2SO$ solvent were taken. Proton NMR of the sample together with the Evans tube was recorded, which showed two peaks corresponding to the solvent residual peak of $CDCl_3$, $CDCl_3$, $(CD_3)_2SO$, $CDCl_3$, $CDCl_3$, $CDCl_3$, $(CD_3)_2SO$ and $(CD_3)_2SO$ in the sample and reference. The shift in the position due to the paramagnetic Ru(III) was noted. Experiment was carried out in 400 MHz NMR spectrometer at 298 K.

$$\chi_g = \frac{-3\Delta f}{4\pi \nu_0 m} + \chi_0 + \frac{\chi_0(d_0 - d_s)}{m} \qquad \text{-------- eq. 1}$$

The mass susceptibility (χ_g) was calculated using eq. 1, where Δf (Hz), ν_0 (Hz), m (g/cm^3), d_0/ d_s and χ_0 corresponded to shift in frequency, operating frequency of NMR spectrometer, concentration of the substance, density of pure solvent and solution, and mass susceptibility of the solvent, respectively. The molar susceptibility (χ_m) was obtained by multiplying the mass susceptibility (χ_g) by the molar mass. This result was used to calculate the effective magnetic moment μ_{eff} in eq. 2.

$$\mu_{eff} = 2.83 \times (\chi_M T)^{1/2} \quad BM \qquad \text{-------- eq. 2}$$

Calculation for $\mathbf{2}^{2+}$:

3.16 mg of $\mathbf{2}^{2+}$ was dissolved in ~0.54 cm^3 of $CDCl_3$. The shift in the position due to the paramagnetic Ru(III) = 0.072 ppm.

$$\chi_M = 0.0030 \text{ cm}^3/\text{mol}$$
$$\mu_{eff} = 2.83 \times (0.0030 \times 298.15)^{1/2} \text{ BM}$$
$$= 2.70 \text{ BM}$$

Calculation for **2a**:

2.8 mg of **2a** was dissolved in ~0.53 cm^3 of $CDCl_3$. The shift in the position due to the paramagnetic Ru(III) = 0.075 ppm.

$$\chi_M = 0.0029 \text{ cm}^3/\text{mol}$$
$$\mu_{eff} = 2.83 \times (0.0029 \times 298.15)^{1/2} \text{ BM}$$
$$= 2.65 \text{ BM}$$

Calculation for $\mathbf{3}^{2+}$:

4.6 mg of $\mathbf{3}^{2+}$ was dissolved in ~0.54 cm^3 of DMSO-d6. The shift in the position due to the paramagnetic Ru(III) = 0.099 ppm.

$$\chi_M = 0.0034 \text{ cm}^3/\text{mol}$$
$$\mu_{eff} = 2.83 \times (0.0034 \times 298.15)^{1/2} \text{ BM}$$
$$= 2.86 \text{ BM}$$

Calculation for **3a**:

3.97 mg of **3a** was dissolved in ~0.52 cm^3 of $CDCl_3$. The shift in the position due to the paramagnetic Ru(III) = 0.097 ppm.

$$\chi_M = 0.0027 \text{ cm}^3/\text{mol}$$
$$\mu_{eff} = 2.83 \times (0.0027 \times 298.15)^{1/2} \text{ BM}$$
$$= 2.55 \text{ BM}$$

Calculation for **4**$^{2+}$:

4.3 mg of **4**$^{2+}$ was dissolved in ~0.53 cm^3 of $CDCl_3$. The shift in the position due to the paramagnetic Ru(III) = 0.099 ppm.

$$\chi_M = 0.0031 \text{ cm}^3/\text{mol}$$
$$\mu_{eff} = 2.83 \times (0.0031 \times 298.15)^{1/2} \text{ BM}$$
$$= 2.73 \text{ BM}$$

Calculation for **4a**:

3.71 mg of **4a** was dissolved in ~0.52 cm^3 of $CDCl_3$. The shift in the position due to the paramagnetic Ru(III) = 0.092 ppm.

$$\chi_M = 0.0034 \text{ cm}^3/\text{mol}$$
$$\mu_{eff} = 2.83 \times (0.0034 \times 298.15)^{1/2} \text{ BM}$$
$$= 2.86 \text{ BM}$$

Calculation for **5**$^{2+}$:

3.96 mg of **5**$^{2+}$ was dissolved in ~0.41 cm^3 of $(CD_3)_2SO$. The shift in the position due to the paramagnetic Ru(III) = 0.113 ppm.

$$\chi_M = 0.0026 \text{ cm}^3/\text{mol}$$
$$\mu_{eff} = 2.83 \times (0.0026 \times 298.15)^{1/2} \text{ BM}$$
$$= 2.50 \text{ BM}$$

Calculation for **5a**:

3.96 mg of **5a** was dissolved in ~0.56 cm^3 of $(CD_3)_2SO$. The shift in the position due to the paramagnetic Ru(III) = 0.096 ppm.

$$\chi_M = 0.0029 \text{ cm}^3/\text{mol}$$
$$\mu_{eff} = 2.83 \times (0.0029 \times 298.15)^{1/2} \text{ BM}$$
$$= 2.64 \text{ BM}$$

Table S5 TD-DFT (UB3LYP/6-31G**/LANL2DZ) calculated electronic transitions for **2** ^{n}, **3** ^{n}, **4** ^{n}, **5** ^{n}, **2a**, **3a**, **4a** and **5a** (n = +2, 0) in CH_3CN

λ /nm expt (DFT)	ε / dm^3 mol^{-1}cm^{-1} (f)	Transitions	Character
		2 (S=0)	
590	11240	HOMO-2→LUMO(0.66)	Ru(dπ)→L(π*)
(518)	(0.070)		
(517)	(0.106)	HOMO-3→LUMO(0.67)	Ru(dπ)→L(π*)
398	10390	HOMO-1→LUMO+2(0.41)	Ru(dπ)/acac(π)→L(π*)
(405)	(0.071)	HOMO→LUMO+3(0.39)	Ru(dπ)/acac(π)→L(π*)
(358)	(0.040)	HOMO-4→LUMO+2(0.36)	Ru(dπ)→L(π*)
		HOMO-3→LUMO+2(0.21)	Ru(dπ)→L(π*)
273	37970	HOMO-4→LUMO+6(0.39)	Ru(dπ)→acac(π*)
(327)	(0.025)	HOMO-2→LUMO+5(0.13)	Ru(dπ)→acac(π*)
		3(S=0)	
583	20210	HOMO-2→LUMO(0.61)	Ru(dπ)/acac(π)→L(π*)
(516)	(0.064)		
399	20820	HOMO→LUMO+3(0.55)	Ru(dπ)/acac(π)→L(π*)
(410)	(0.069)	HOMO→LUMO+5(0.23)	Ru(dπ)/acac(π)→L(π*)
(404)	(0.036)	HOMO-1→LUMO+3(0.52)	Ru(dπ)/acac(π)→L(π*)
		HOMO-1→LUMO+8(0.15)	Ru(dπ)/acac(π)→L(π*)/Ru(dπ)
274	59960	HOMO-5→LUMO+4(0.23)	Ru(dπ)→acac(π*)
(343)	(0.082)	HOMO-4→LUMO+5(0.22)	Ru(dπ)→acac(π*)
		4 (S=0)	
582	20080	HOMO-3→LUMO(0.44)	Ru(dπ)→L(π*)
(509)	(0.036)	HOMO-2→LUMO+1(0.42)	Ru(dπ)→L(π*)
(505)	(0.201)	HOMO-2→LUMO(0.46)	Ru(dπ)→L(π*)
		HOMO-3→LUMO(0.39)	Ru(dπ)→L(π*)
399	18830	HOMO→LUMO+3(0.47)	Ru(dπ)/acac(π)→L(π*)
(408)	(0.055)	HOMO→LUMO+5(0.14)	Ru(dπ)/acac(π)→acac(π*)
(360)	(0.062)	HOMO-4→LUMO+3(0.32)	Ru(dπ)→L(π*)
		HOMO-4→LUMO+7(0.21)	Ru(dπ)→acac(π*)
276	36230	HOMO-1→LUMO+10(0.38)	Ru(dπ)/acac(π)→Ru(dπ)/L(π*)
(268)	(0.061)	HOMO-1→LUMO+9(0.32)	Ru(dπ)/acac(π)→Ru(dπ)/L(π*)
(266)	(0.041)	HOMO-7→LUMO+4(0.41)	acac(π)→acac(π*)
		HOMO-6→LUMO+5(0.29)	acac(π)→acac(π*)
		5 (S=0)	
588	15930	HOMO-3→LUMO(0.41)	Ru(dπ)→L(π*)
(504)	(0.226)	HOMO-2→LUMO(0.31)	Ru(dπ)→L(π*)
392	20400	HOMO-1→LUMO+2(0.51)	Ru(dπ)/acac(π)→L(π*)
(407)	(0.116)	HOMO→LUMO+3(0.29)	Ru(dπ)/acac(π)→L(π*)

(360)	(0.047)	HOMO-4→LUMO+3(0.38) HOMO-4→LUMO+7(0.27)	Ru(dπ)→L(π*) Ru(dπ)→acac(π*)
273 (342)	64310 (0.038)	HOMO-4→LUMO+5(0.37) HOMO-4→LUMO+12(0.25)	Ru(dπ)→acac(π*) Ru(dπ)→Ru(dπ)/L(π*)
2^{2+} (*S*=1)			
584 (593)	7515 (0.062)	HOMO-4(β)→LUMO(β)(0.70)	acac(π)→Ru(dπ)/acac(π*)
479 (537)	13805 (0.026)	HOMO-1(β)→LUMO(β)(0.57) HOMO(β)→LUMO+1(β)(0.55)	Ru(dπ)/acac(π)→Ru(dπ)/acac(π*) Ru(dπ)/acac(π)→Ru(dπ)/acac(π*)
382 (397)	16530 (0.033)	HOMO-3(α)→LUMO(α)(0.56) HOMO-1(β)→LUMO+2(β)(0.29)	Ru(dπ)/acac(π)→L(π*) Ru(dπ)/acac(π)→L(π*)
331 (309)	25275 (0.034)	HOMO-3(β)→LUMO+7(β)(0.44) HOMO-1(α)→LUMO+4(α)(0.20)	Ru(dπ)/acac(π)→acac(π*) acac(π)/Ru(dπ) →acac(π*)
276 (277)	60535 (0.080)	HOMO-6(α)→LUMO+5(α)(0.43) HOMO-7(α)→LUMO+4(α)(0.39)	acac(π)/Ru(dπ) →acac(π*) acac(π)/Ru(dπ) →acac(π*)
3^{2+} (*S*=1)			
505 (542)	6670 (0.032)	HOMO-6(β)→LUMO+1(β)(0.73)	Ru(dπ)/acac(π)→Ru(dπ)/acac(π*)
381 (393)	21180 (0.039)	HOMO-3(α)→LUMO(α)(0.44) HOMO-6(α)→LUMO(α)(0.19)	acac(π)→L(π*) Ru(dπ)/acac(π)→L(π*)
336 (308)	23630 (0.039)	HOMO-2(β)→LUMO+4(β)(0.45) HOMO-3(β)→LUMO+5(β)(0.44)	acac(π)/Ru(dπ)→L(π*) acac(π)/Ru(dπ)→L(π*)
273 (272)	46060 (0.178)	HOMO-8(α)→LUMO(α)(0.46) HOMO-8(α)→LUMO+1(α)(0.29)	acac(π)→L(π*) acac(π)→L(π*)
4^{2+} (*S*=1)			
577 (580) (543)	7640 (0.054) (0.031)	HOMO-5(β)→LUMO+1(β)(0.71) HOMO-6(β)→LUMO(β)(0.80)	acac(π)/Ru(dπ)→Ru(dπ)/acac(π*) acac(π)/Ru(dπ)→Ru(dπ)/acac(π*)
469 (396)	12210 (0.042)	HOMO-2(α)→LUMO(α)(0.57) HOMO-1(β)→LUMO+2(β)(0.27)	acac(π)/Ru(dπ)→L(π*) Ru(dπ)/acac(π)→L(π*)
378 (393)	14420 (0.037)	HOMO-3(α)→LUMO+1(α)(0.49) HOMO-3(α)→LUMO(α)(0.34)	Ru(dπ)/acac(π)→L(π*) Ru(dπ)/acac(π)→L(π*)
274 (278) (271)	58260 (0.093) (0.038)	HOMO-4(α)→LUMO+6(α)(0.35) HOMO-2(α)→LUMO+4(α)(0.33) HOMO-6(α)→LUMO+2(α)(0.58) HOMO-7(α)→LUMO+2(α)(0.49)	acac(π)/Ru(dπ)→acac(π*) Ru(dπ)/acac(π)→L(π*) acac(π)/Ru(dπ)→L(π*) acac(π)/Ru(dπ)→L(π*)
5^{2+} (*S*=1)			
571 (581)	6640 (0.071)	HOMO-4(β)→LUMO+1(β)(0.93)	Ru(dπ)/acac(π)→Ru(dπ)/acac(π*)
473 (535)	12250 (0.067)	HOMO-6(β)→LUMO+1(β)(0.81)	acac(π)/Ru(dπ)→Ru(dπ)/acac(π*)
381 (394)	13040 (0.048)	HOMO-3(α)→LUMO+1(α)(0.46) HOMO-3(α)→LUMO(α)(0.43)	Ru(dπ)/acac(π)→L(π*) Ru(dπ)/acac(π)→L(π*)

328 (309)	20000 (0.031)	HOMO-3(β)→LUMO+7(β)(0.49) HOMO-2(β)→LUMO+7(β)(0.44)	Ru(dπ)/acac(π)→acac(π*) Ru(dπ)/acac(π)→acac(π*)
276 (276)	60080 (0.043)	HOMO-4(β)→LUMO+7(β)(0.54) HOMO-6(β)→LUMO+7(β)(0.54)	Ru(dπ)/acac(π)→acac(π*) acac(π)/Ru(dπ)→ acac(π*)
234 (267)	39550 (0.177)	HOMO-9(α)→LUMO+1(α)(0.39) HOMO-6(α)→LUMO+7(α)(0.27)	acac(π)/Ru(dπ)→L(π*) Ru(dπ)→ acac(π*)
2a (*S*=1)			
508 (527)	14920 (0.046)	HOMO-4(β)→LUMO+1(β)(0.66)	Ru(dπ)/L(π)/acac(π)→Ru(dπ)/acac(π*)/L(π*)
438 (453)	27040 (0.026)	HOMO-8(β)→LUMO(β)(0.55) HOMO-5(β)→LUMO+1(β)(0.41)	acac(π)/ L(π)→Ru(dπ)/acac(π*) L(π)/acac(π)→Ru(dπ)/acac(π*)/L(π*)
393 (370)	39450 (0.031)	HOMO(β)→LUMO+3(β)(0.42) HOMO-3(β)→LUMO+2(β)(0.35)	Ru(dπ)/L(π)/acac(π)→L(π*) Ru(dπ)/acac(π)→L(π*)
333 (347)	49560 (0.052)	HOMO-3(α)→LUMO(α)(0.32) HOMO-10(β)→LUMO(β)(0.27)	Ru(dπ)/acac(π)→L(π*) acac(π)/Ru(dπ)→ Ru(dπ)/acac(π*)
3a (*S*=1)			
508 (528)	13360 (0.042)	HOMO-4(β)→LUMO+1(β)(0.82)	acac(π)/Ru(dπ)→Ru(dπ)/L(π*)
434 (485)	25540 (0.030)	HOMO-6(β)→LUMO+1(β)(0.73)	L(π)→ Ru(dπ)/L(π*)
384 (375)	34650 (0.035)	HOMO-1(β)→LUMO+2(β)(0.42) HOMO-1(α)→LUMO(α)(0.37)	Ru(dπ)/acac(π)/L(π)→L(π*) Ru(dπ)/acac(π)/L(π)→L(π*)
337 (357)	39580 (0.030)	HOMO-3(β)→LUMO+3(β)(0.47) HOMO-2(α)→LUMO+1(α)(0.32)	Ru(dπ)→L(π*) Ru(dπ)/L(π)→L(π*)
4a (*S*=1)			
503 (532)	12740 (0.055)	HOMO-5(β)→LUMO(β)(0.61)	L(π)→ Ru(dπ)/acac(π*)
375 (370)	38490 (0.036)	HOMO(β)→LUMO+3(β)(0.36) HOMO-3(β)→LUMO+2(β)(0.36)	Ru(dπ)/L(π)/acac(π)→acac(π*) Ru(dπ)/L(π)→L(π*)
340 (342)	39500 (0.033)	HOMO-4(α)→LUMO(α)(0.20) HOMO(β)→LUMO+5(β)(0.19)	Ru(dπ)/acac(π)→L(π*) Ru(dπ)/L(π)/acac(π)→L(π*)
273 (296)	73920 (0.038)	HOMO-5(α)→LUMO+2(α)(0.35) HOMO-4(α)→LUMO+4(α)(0.24)	L(π)→L(π*) Ru(dπ)/acac(π)→acac(π*)
5a (*S*=1)			
505 (525) (490)	12710 (0.047) (0.031)	HOMO-9(β)→LUMO(β)(0.86) HOMO-6(β)→LUMO+1(β)(0.88)	L(π)/acac(π)/Ru(dπ)→Ru(dπ)/L(π*) acac(π)/L(π)→Ru(dπ)
374 (365)	41890 (0.032)	HOMO-2(β)→LUMO+3(β)(0.47) HOMO(α)→LUMO+3(α)(0.44)	L(π)/Ru(dπ)→ L(π*)/acac(π*) L(π)/Ru(dπ)→ L(π*)/acac(π*)
339	44690	HOMO-2(β)→LUMO+3(β)(0.49)	L(π)/Ru(dπ)→ L(π*)/acac(π*)

(356)	(0.033)	HOMO-3(α)→LUMO+1(α)(0.18)	Ru(dπ)/acac(π)→L(π*)
274 (292)	87420 (0.031)	HOMO-4(α)→LUMO+5(α)(0.38) HOMO-2(β)→LUMO+9(β)(0.37)	L(π)/Ru(dπ)→acac(π*) L(π)/Ru(dπ)→L(π*)

Table S6 DFT calculated selected MO compositions ((U/R)B3LYP/LanL2DZ/6-31G**)

Complex	Energy (eV)	MO	% Contribution Ru/acac/L
2^{2+} (*S*=1) $\Delta E_{((S=0)-(S=1))}$= 9326 cm^{-1}	-8.090	β-LUMO	65/28/7
2 (*S*=0) $\Delta E_{((S=1)-(S=0))}$= 7895 cm^{-1}	-3.756 -1.665	HOMO LUMO	62/31/7 14/4/81
2^{2-} (*S*=1) $\Delta E_{((S=0)-(S=1))}$= 3279 cm^{-1}	2.988	SOMO 1	12/5/83
3^{2+} (*S*=1) $\Delta E_{((S=0)-(S=1))}$= 9421 cm^{-1}	-8.572	β-LUMO	69/23/8
3 (*S*=0) $\Delta E_{((S=1)-(S=0))}$= 7630 cm^{-1}	-4.034 -1.893	HOMO LUMO	63/30/8 14/3/83
3^{2-} (*S*=1) $\Delta E_{((S=0)-(S=1))}$= 4814 cm^{-1}	2.780	SOMO 1	13/4/83
4^{2+} (*S*=1) $\Delta E_{((S=0)-(S=1))}$= 9555 cm^{-1}	-8.028	β-LUMO	65/27/8
4 (*S*=0) $\Delta E_{((S=1)-(S=0))}$= 7467 cm^{-1}	-3.806 -1.734	HOMO LUMO	62/33/5 13/4/83
4^{2-} (*S*=1) $\Delta E_{((S=0)-(S=1))}$= 5533 cm^{-1}	2.751	SOMO 1	13/5/82
5^{2+} (*S*=1) $\Delta E_{((S=0)-(S=1))}$= 9765 cm^{-1}	-7.951	β-LUMO	62/32/6
5 (*S*=0) $\Delta E_{((S=1)-(S=0))}$= 7980 cm^{-1}	-3.831 -1.751	HOMO LUMO	63/32/5 13/4/83
5^{2-} (*S*=1) $\Delta E_{((S=0)-(S=1))}$= 5448 cm^{-1}	2.469	SOMO 1	12/4/84
Complex	Energy (eV)	MO	% Contribution Ru/acac/L"
2a^{4+} (*S*=1) $\Delta E_{((S=0)-(S=1))}$= 2727 cm^{-1}	-15.650	β-LUMO	45/47/8
2a^{3+} (*S*=3/2) $\Delta E_{((S=1/2)-(S=3/2))}$= 754 cm^{-1}	-13.971 -12.588	SOMO 1 β-LUMO	26/59/15 61/27/12
2a^{2+} (*S*=1) $\Delta E_{((S=0)-(S=1))}$= 982 cm^{-1}	-11.267 -9.733	SOMO 1 β-LUMO	30/52/18 57/24/19
2a^{+} (*S*=1/2)	-8.230 -7.075	SOMO β-LUMO	43/37/19 43/17/40
2a (*S*=1) $\Delta E_{((S=0)-(S=1))}$= 8294 cm^{-1}	-4.738 -2.469	SOMO 1 β-LUMO	30/18/52 60/26/14
2a^{2-} (*S*=0) $\Delta E_{((S=1)-(S=0))}$= 9753 cm^{-1}	1.995	HOMO	52/11/37
3a^{4+} (*S*=1) $\Delta E_{((S=0)-(S=1))}$= 1295 cm^{-1}	-14.496	β-LUMO	45/51/4
3a^{3+} (*S*=3/2)	-13.067	SOMO 1	32/48/20

$\Delta E_{((S=1/2)-(S=3/2))}$= 401 cm^{-1}	-12.428	β-LUMO	54/37/9
3a^{2+} (*S*=1) $\Delta E_{((S=0)-(S=1))}$= 693 cm^{-1}	-11.030 -9.485	SOMO 1 β-LUMO	34/51/15 59/26/15
3a^{+} (*S*=1/2)	-7.788 -6.152	SOMO β-LUMO	46/15/40 67/21/12
3a (*S*=1) $\Delta E_{((S=0)-(S=1))}$= 8661 cm^{-1}	-4.930 -2.537	SOMO 1 β-LUMO	36/32/32 65/17/18
3a^{2-} (*S*=0) $\Delta E_{((S=1)-(S=0))}$= 10160 cm^{-1}	1.911	HOMO	57/12/30
4a^{4+} (*S*=1) $\Delta E_{((S=0)-(S=1))}$= 6558 cm^{-1}	-14.824	β-LUMO	4/42/10
4a^{3+} (*S*=3/2) $\Delta E_{((S=1/2)-(S=3/2))}$=538 cm^{-1}	-13.230 -12.642	SOMO 1 β-LUMO	30/55/15 31/44/25
4a^{2+} (*S*=1) $\Delta E_{((S=0)-(S=1))}$= 932 cm^{-1}	-10.843 -9.096	SOMO 1 β-LUMO	31/44/25 62/25/13
4a^{+} (*S*=1/2)	-7.553 -5.921	SOMO β-LUMO	51/35/14 59/13/28
4a (*S*=1) $\Delta E_{((S=0)-(S=1))}$= 10132 cm^{-1}	-4.817 -2.523	SOMO 1 β-LUMO	28/7/65 60/21/19
4a^{2-} (*S*=0) $\Delta E_{((S=1)-(S=0))}$= 9818 cm^{-1}	1.348	HOMO	60/12/28
5a^{4+} (*S*=1) $\Delta E_{((S=0)-(S=1))}$= 8531 cm^{-1}	-14.332	β-LUMO	45/50/4
5a^{3+} (*S*=3/2) $\Delta E_{((S=1/2)-(S=3/2))}$= 1758 cm^{-1}	-13.144 -12.822	SOMO 1 β-LUMO	33/46/21 31/54/14
5a^{2+} (*S*=1) $\Delta E_{((S=0)-(S=1))}$= 479 cm^{-1}	-10.752 -9.217	SOMO 1 β-LUMO	34/51/15 58/26/16
5a^{+} (*S*=1/2)	-8.056 -7.000	SOMO β-LUMO	35/39/27 48/20/32
5a (*S*=1) $\Delta E_{((S=0)-(S=1))}$= 9652 cm^{-1}	-5.143 -2.795	SOMO 1 β-LUMO	22/6/72 62/13/25
5a^{2-} (*S*=0) $\Delta E_{((S=1)-(S=0))}$= 9528 cm^{-1}	1.781	HOMO	61/14/25

Table S7 Composition and energies of selected molecular orbitals of $\mathbf{2}^{2+}$ (*S*=1)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	-5.921	8	83	9
LUMO+4	-5.941	7	74	17
LUMO+3	-6.028	3	15	82
LUMO+2	-6.040	2	8	90
LUMO+1	-7.219	8	3	90
LUMO	-7.454	7	2	91
SOMO 1	-10.655	29	67	3
SOMO 2	-10.673	31	66	3
HOMO-2	-10.989	48	44	8
HOMO-3	-11.014	51	41	8
HOMO-4	-11.323	71	20	9
HOMO-5	-11.347	73	19	8
		β-spin		
LUMO+5	-5.993	3	16	80
LUMO+4	-6.004	3	11	86
LUMO+3	-7.203	9	3	88
LUMO+2	-7.440	9	3	89
LUMO+1	-8.084	64	28	7
LUMO	-8.090	65	28	7
HOMO	-10.711	57	34	9
HOMO-1	-10.724	58	32	9
HOMO-2	-10.907	60	32	7
HOMO-3	-10.911	62	31	7
HOMO-4	-11.127	19	75	6
HOMO-5	-11.155	16	78	6

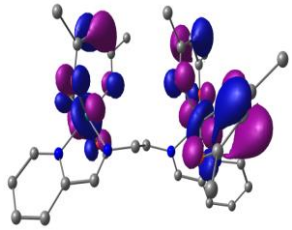
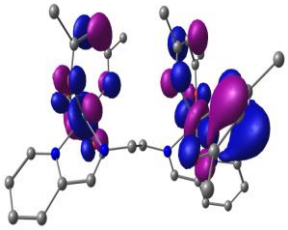
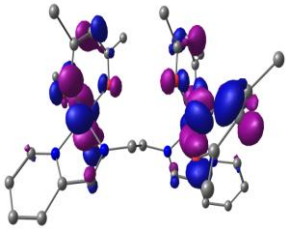
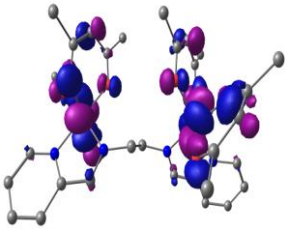
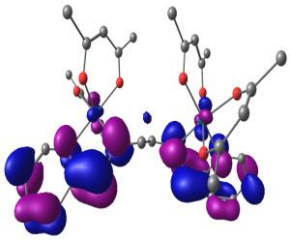
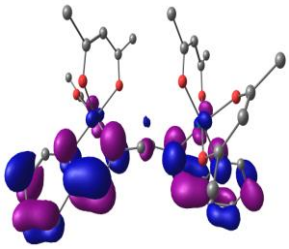
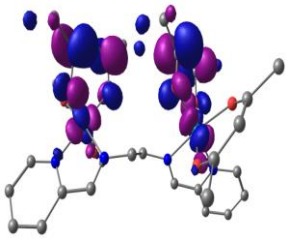
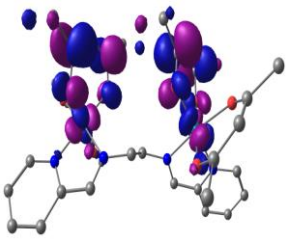
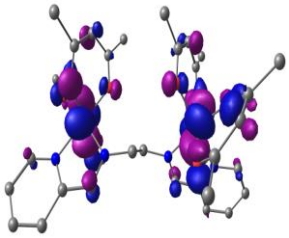
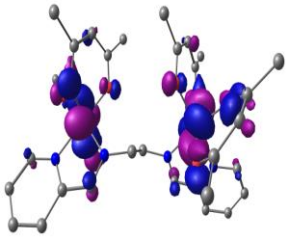
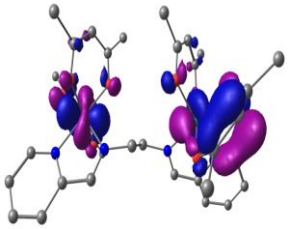
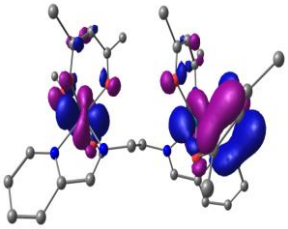
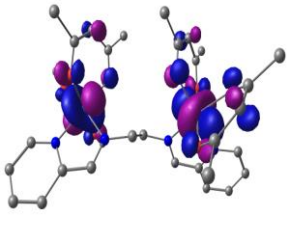
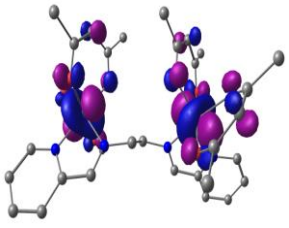
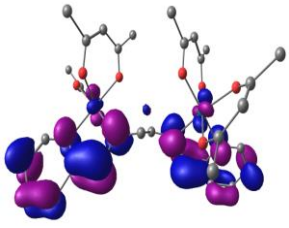
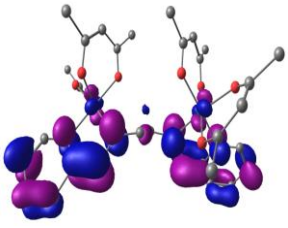
α-spin			
			
SOMO 1	SOMO 2	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S8 Composition and energies of selected molecular orbitals of **2** (*S*=0)

MO	Energy (eV)	% Composition		
		Ru	acac	L
LUMO+5	-0.234	6	88	7
LUMO+4	-0.238	6	87	7
LUMO+3	-0.626	4	4	92
LUMO+2	-0.642	4	5	91
LUMO+1	-1.437	13	5	81
LUMO	-1.665	14	4	81
HOMO	-3.756	62	31	7
HOMO-1	-3.760	63	31	6
HOMO-2	-4.377	64	19	17
HOMO-3	-4.380	65	19	16
HOMO-4	-4.471	74	17	8
HOMO-5	-4.482	75	17	8

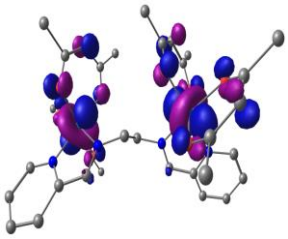
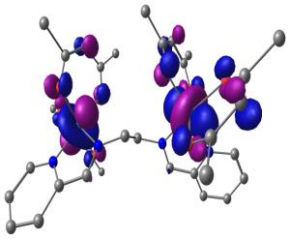
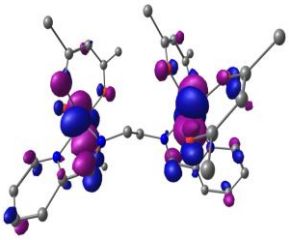
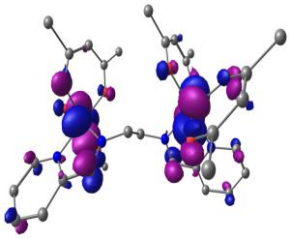
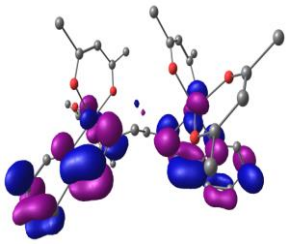
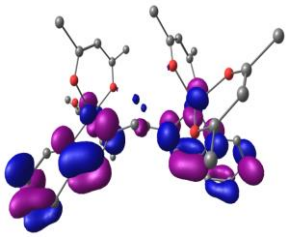
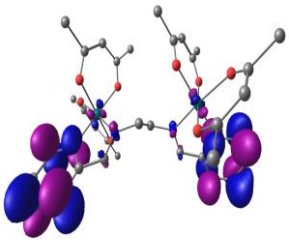
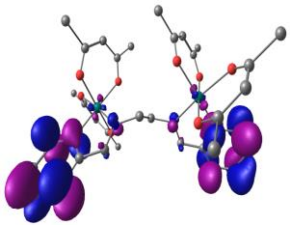
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S9 Composition and energies of selected molecular orbitals of $\mathbf{2}^{2-}$ (*S*=1)

MO	Energy (eV)	% Composition		
		Ru	acac	L
	α-spin			
LUMO+5	5.065	7	2	91
LUMO+4	5.040	8	3	89
LUMO+3	4.667	8	85	5
LUMO+2	4.650	6	88	3
LUMO+1	4.468	5	91	4
LUMO	4.467	5	92	4
SOMO 1	2.988	12	5	83
SOMO 2	2.762	12	5	84
HOMO-2	1.322	74	19	7
HOMO-3	1.314	75	19	6
HOMO-4	0.908	79	15	7
HOMO-5	0.869	78	15	7
	β-spin			
LUMO+5	4.889	13	13	73
LUMO+4	4.786	12	53	34
LUMO+3	4.668	6	87	5
LUMO+2	4.604	8	68	23
LUMO+1	4.447	2	89	8
LUMO	4.401	2	72	25
HOMO	1.373	73	18	9
HOMO-1	1.361	74	18	8
HOMO-2	1.091	74	15	11
HOMO-3	1.090	73	15	12
HOMO-4	0.944	78	15	7
HOMO-5	0.907	77	15	7

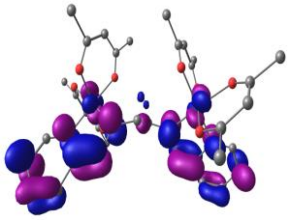
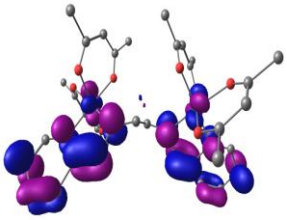
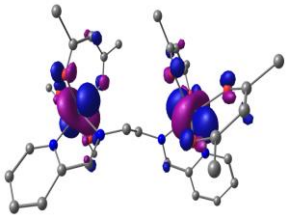
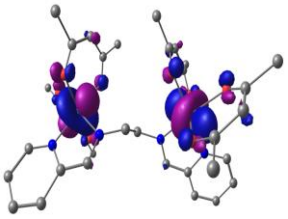
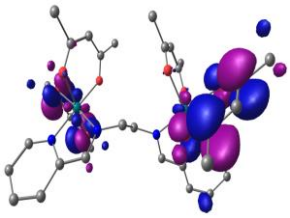
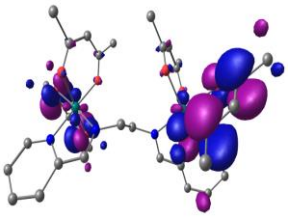
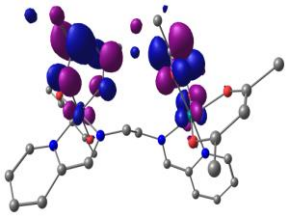
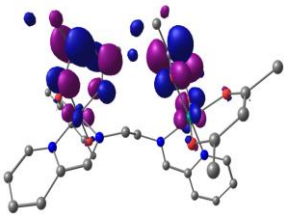
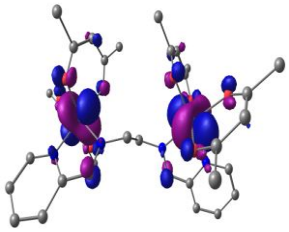
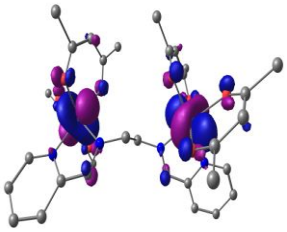
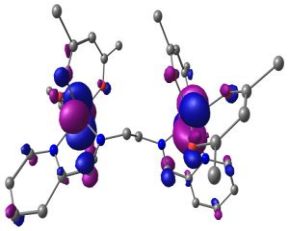
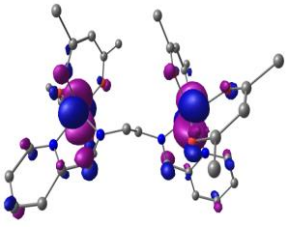
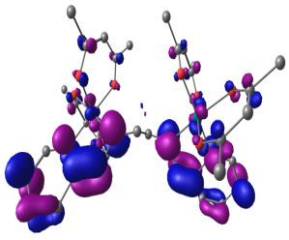
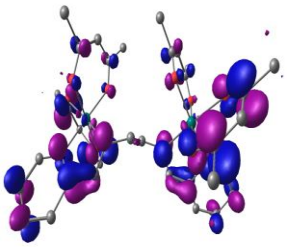
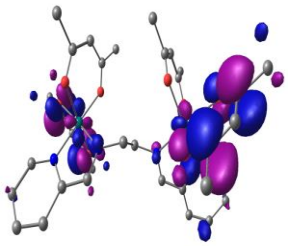
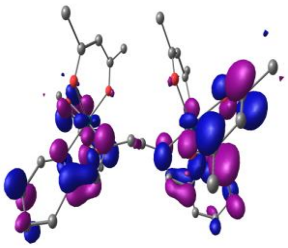
α-spin			
			
SOMO 1	SOMO 2	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S10 Composition and energies of selected molecular orbitals of **2a**$^{4+}$ (*S*=1)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	-12.226	30	58	13
LUMO+4	-12.277	6	90	4
LUMO+3	-12.574	49	43	8
LUMO+2	-12.633	53	38	8
LUMO+1	-15.513	31	6	63
LUMO	-15.861	37	53	10
SOMO 1	-17.303	3	10	86
SOMO 2	-17.356	1	1	98
HOMO-2	-17.441	22	48	30
HOMO-3	-17.551	20	61	19
HOMO-4	-17.670	38	21	41
HOMO-5	-17.729	17	60	23
		β-spin		
LUMO+5	-12.406	43	49	7
LUMO+4	-12.433	24	70	6
LUMO+3	-15.233	43	10	47
LUMO+2	-15.451	42	51	7
LUMO+1	-15.588	53	39	8
LUMO	-15.650	45	47	8
HOMO	-17.103	23	67	10
HOMO-1	-17.240	5	6	89
HOMO-2	-17.324	35	32	33
HOMO-3	-17.412	17	18	66
HOMO-4	-17.487	30	23	46
HOMO-5	-17.650	13	5	82

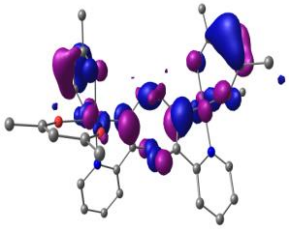
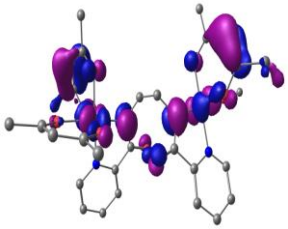
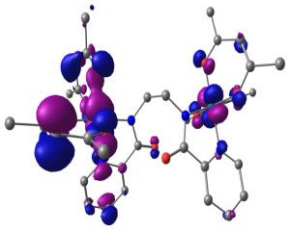
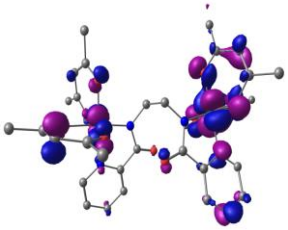
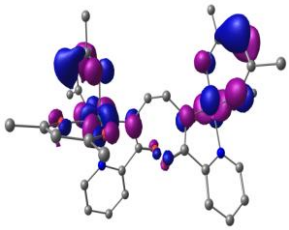
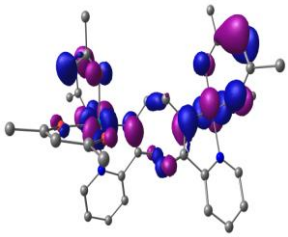
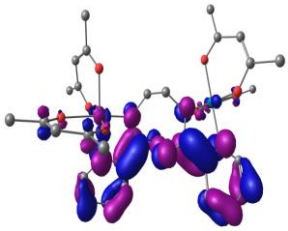
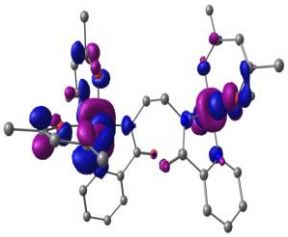
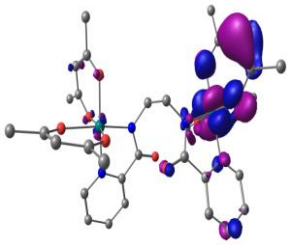
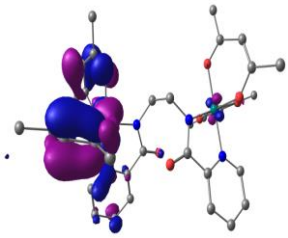
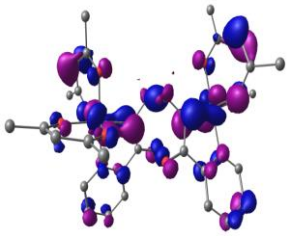
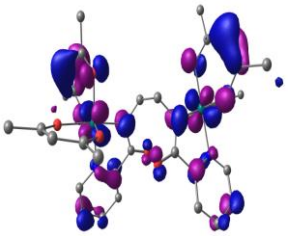
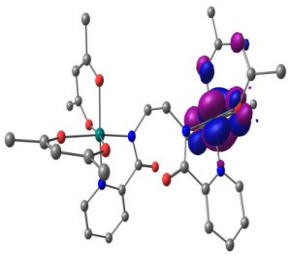
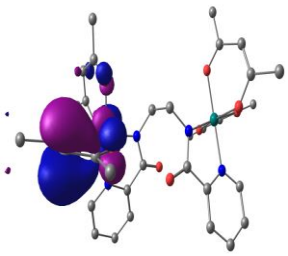
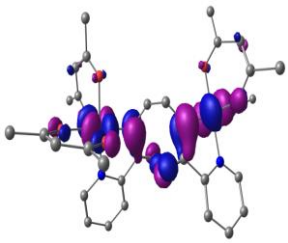
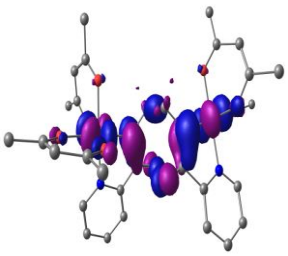
α-spin			
			
SOMO 1	SOMO 2	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S11 Composition and energies of selected molecular orbitals of **2a**$^{3+}$ (*S*=3/2)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	-9.410	33	47	20
LUMO+4	-9.437	9	85	7
LUMO+3	-9.451	12	78	10
LUMO+2	-9.803	55	37	8
LUMO+1	-9.807	54	37	9
LUMO	-13.498	25	68	7
SOMO 1	-13.971	26	59	15
SOMO 2	-14.525	14	38	48
SOMO 3	-14.590	12	69	19
HOMO-3	-14.620	8	83	9
HOMO-4	-14.882	14	17	70
HOMO-5	-14.910	6	2	91
		β-spin		
LUMO+5	-9.511	14	81	5
LUMO+4	-9.521	20	73	7
LUMO+3	-11.930	43	9	48
LUMO+2	-12.140	55	16	29
LUMO+1	-12.518	60	32	8
LUMO	-12.588	61	27	12
HOMO	-14.438	33	51	16
HOMO-1	-14.477	28	61	11
HOMO-2	-14.615	19	57	24
HOMO-3	-14.759	18	7	75
HOMO-4	-14.768	8	16	76
HOMO-5	-14.780	14	17	69

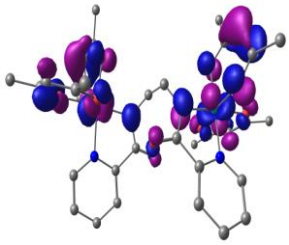
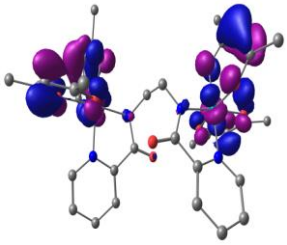
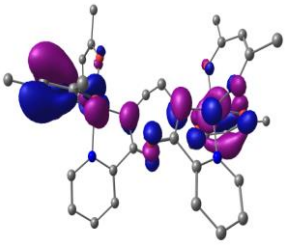
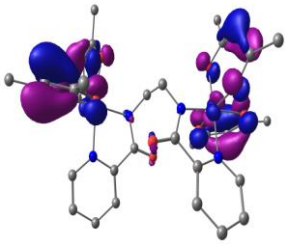
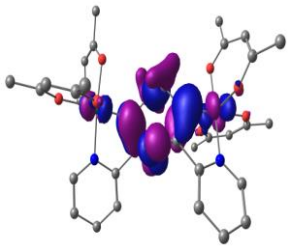
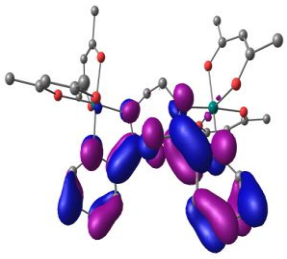
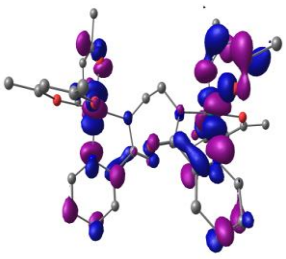
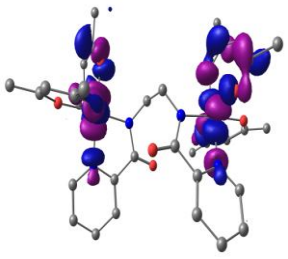
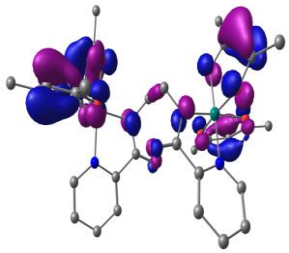
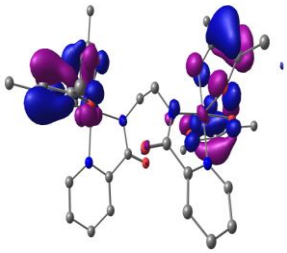
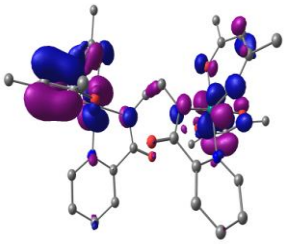
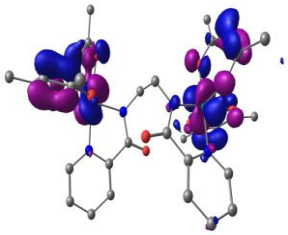
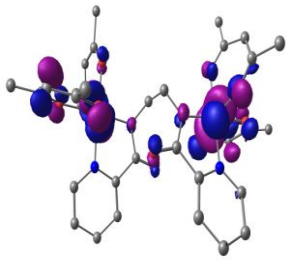
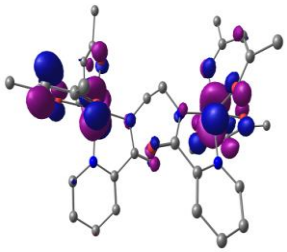
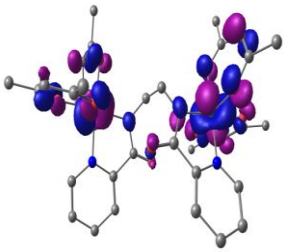
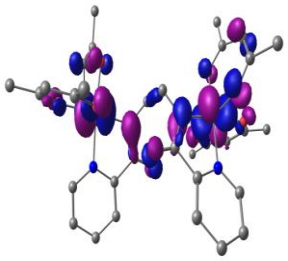
α-spin			
			
SOMO 1	SOMO 2	SOMO 3	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S12 Composition and energies of selected molecular orbitals of **2a**$^{2+}$ (*S*=1)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	-6.570	19	27	54
LUMO+4	-6.653	23	40	37
LUMO+3	-6.684	28	52	20
LUMO+2	-6.826	6	4	90
LUMO+1	-7.037	51	37	11
LUMO	-9.719	52	23	25
SOMO 1	-11.267	30	53	18
SOMO 2	-11.462	22	67	11
HOMO-2	-11.603	9	75	17
HOMO-3	-11.853	10	83	7
HOMO-4	-11.942	27	31	42
HOMO-5	-11.987	54	26	20
		β-spin		
LUMO+5	-6.627	30	45	24
LUMO+4	-6.655	15	27	58
LUMO+3	-6.721	16	35	49
LUMO+2	-9.073	54	15	31
LUMO+1	-9.619	60	25	16
LUMO	-9.733	57	24	19
HOMO	-11.271	31	55	14
HOMO-1	-11.443	10	76	15
HOMO-2	-11.648	12	75	13
HOMO-3	-11.728	25	66	8
HOMO-4	-11.969	56	27	17
HOMO-5	-12.039	30	11	59

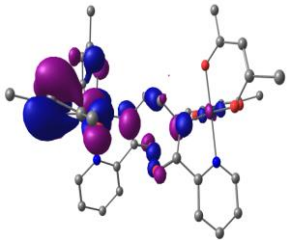
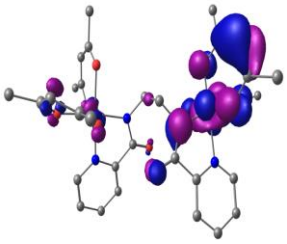
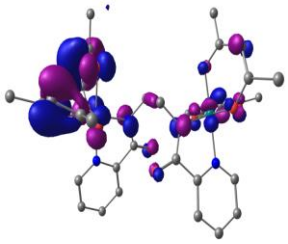
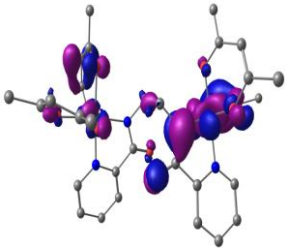
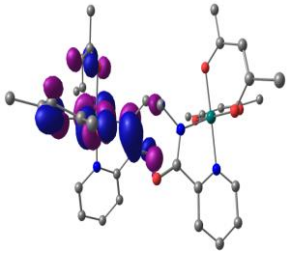
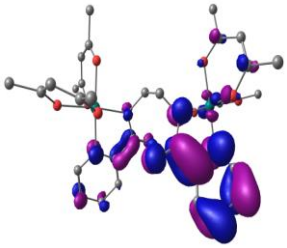
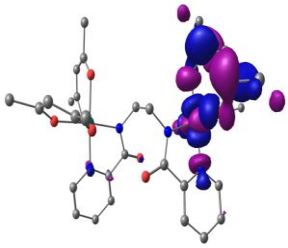
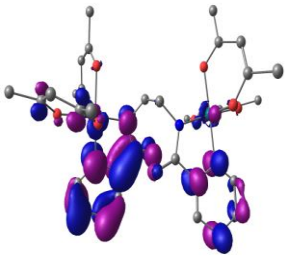
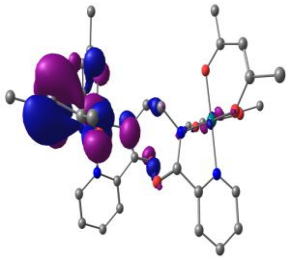
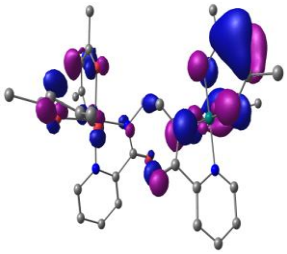
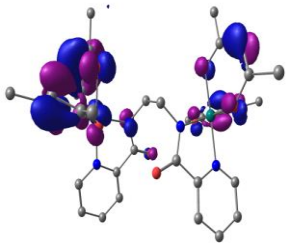
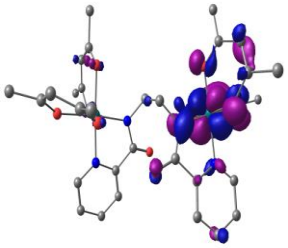
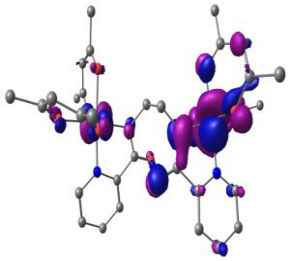
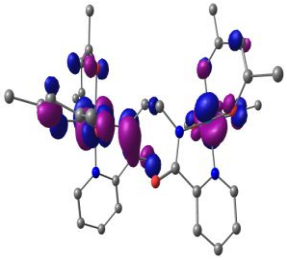
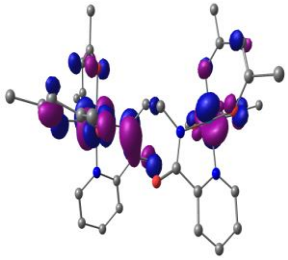
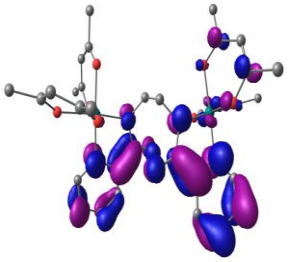
α-spin			
			
SOMO 1	SOMO 2	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S13 Composition and energies of selected molecular orbitals of **2a**$^{+}$ (*S*=1/2)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	-3.576	7	88	5
LUMO+4	-3.654	5	89	6
LUMO+3	-3.799	12	81	7
LUMO+2	-4.059	3	5	92
LUMO+1	-4.154	3	2	95
LUMO	-5.855	67	19	14
SOMO	-8.230	43	37	19
HOMO-1	-8.424	37	40	22
HOMO-2	-8.595	35	57	8
HOMO-3	-8.737	33	57	11
HOMO-4	-8.809	36	56	15
HOMO-5	-8.919	22	50	22
		β-spin		
LUMO+5	-3.649	12	80	9
LUMO+4	-3.729	7	85	8
LUMO+3	-3.993	4	8	88
LUMO+2	-4.056	3	3	94
LUMO+1	-5.737	62	15	22
LUMO	-7.075	43	17	40
HOMO	-7.830	53	22	25
HOMO-1	-8.377	32	64	4
HOMO-2	8.568	26	62	12
HOMO-3	-8.697	26	62	12
HOMO-4	-8.843	44	39	17
HOMO-5	-9.025	54	30	16

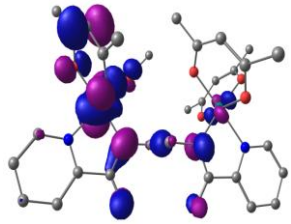
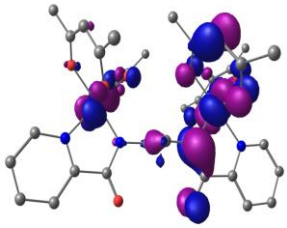
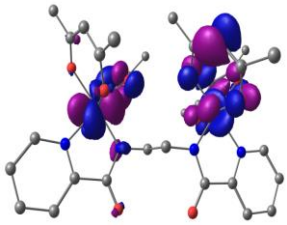
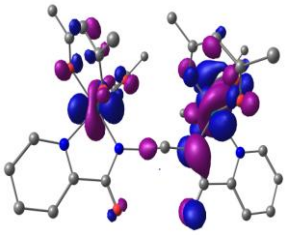
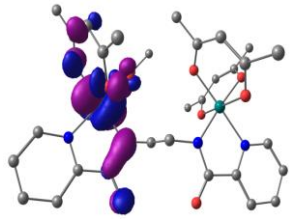
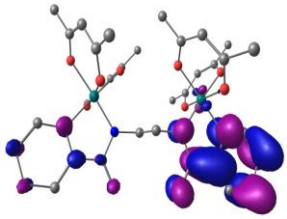
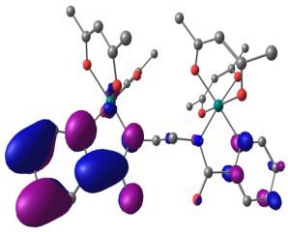
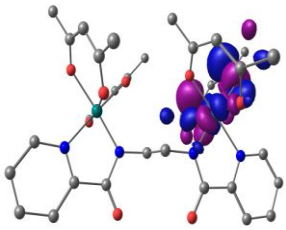
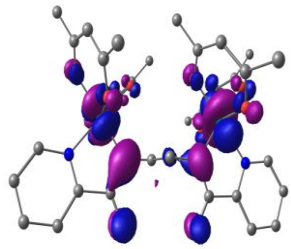
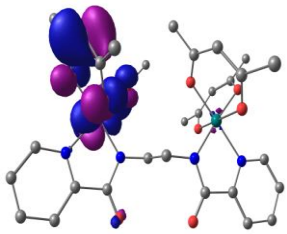
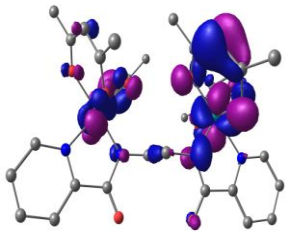
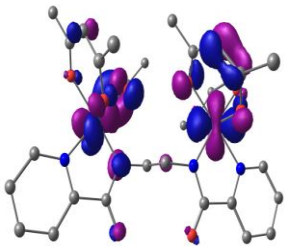
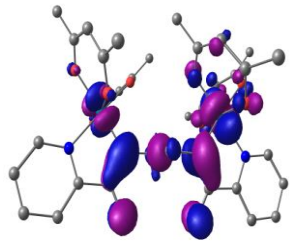
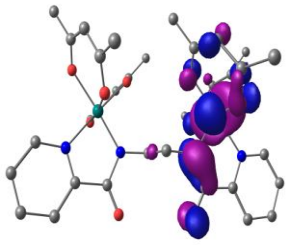
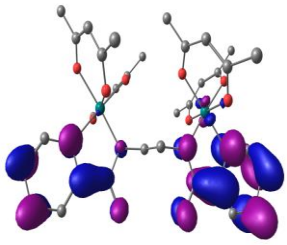
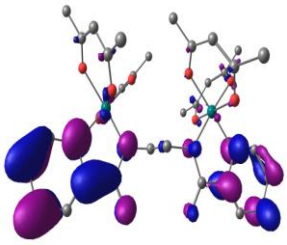
α-spin			
			
SOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S14 Composition and energies of selected molecular orbitals of **2a** (*S*=1)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	-0.622	6	79	15
LUMO+4	-0.693	3	91	6
LUMO+3	-0.753	6	88	6
LUMO+2	-0.817	3	94	3
LUMO+1	-1.143	5	5	90
LUMO	-1.384	5	2	93
SOMO 1	-4.738	30	18	52
SOMO 2	-4.974	34	41	25
HOMO-2	-5.070	54	39	7
HOMO-3	-5.158	49	45	6
HOMO-4	-5.382	44	31	24
HOMO-5	-5.596	34	9	57
		β-spin		
LUMO+5	-0.701	6	86	6
LUMO+4	-0.755	8	91	5
LUMO+3	-1.121	5	8	88
LUMO+2	-1.369	5	3	91
LUMO+1	-2.417	58	21	21
LUMO	-2.469	60	26	14
HOMO	-4.727	46	26	28
HOMO-1	-4.781	49	40	11
HOMO-2	-5.018	43	41	17
HOMO-3	-5.329	62	23	15
HOMO-4	-5.426	44	20	36
HOMO-5	-5.490	16	25	59

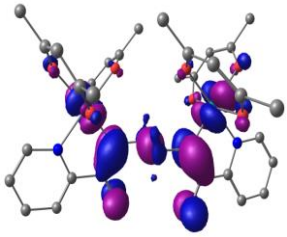
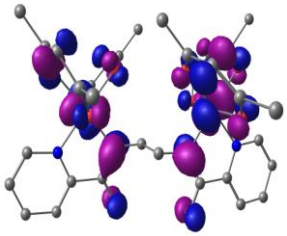
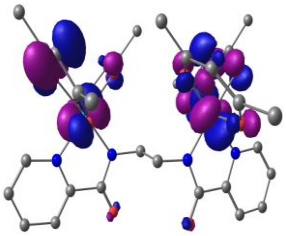
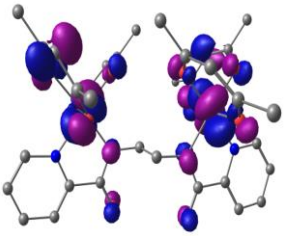
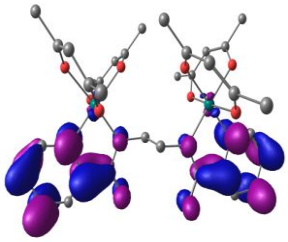
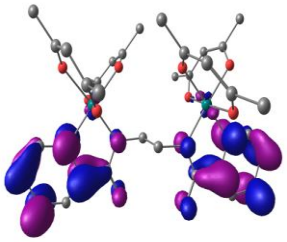
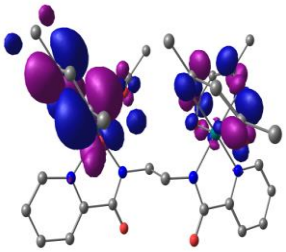
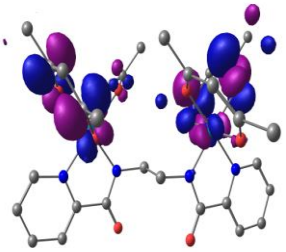
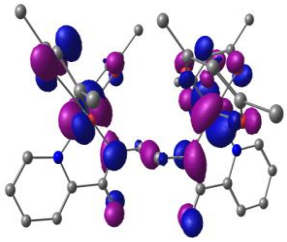
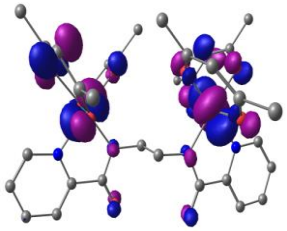
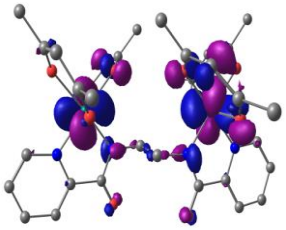
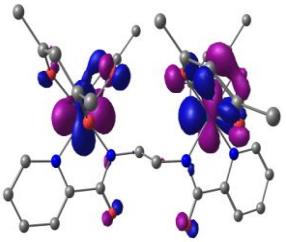
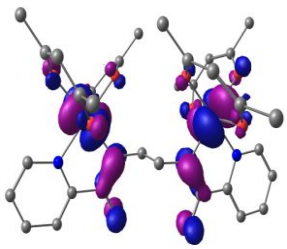
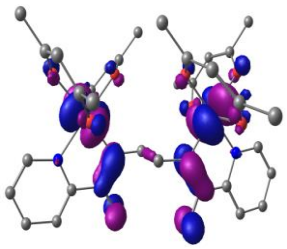
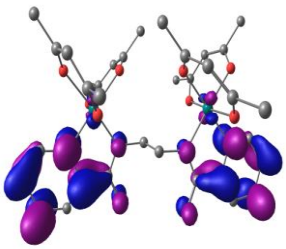
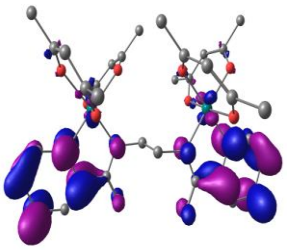
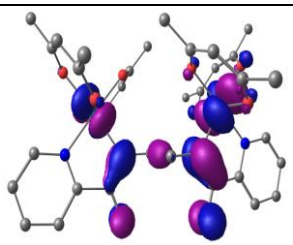
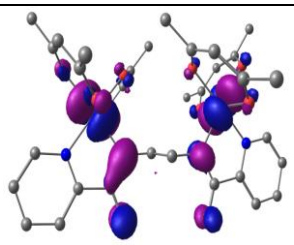
α-spin			
			
SOMO 1	SOMO 2	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S15 Composition and energies of selected molecular orbitals of **2a**$^{2-}$ (*S*=0)

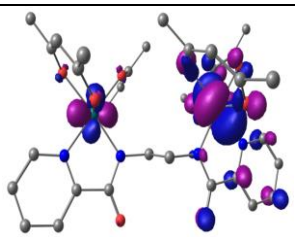
MO	Energy (eV)	% Composition		
		Ru	acac	L
LUMO+5	4.544	7	80	13
LUMO+4	4.523	6	65	29
LUMO+3	4.498	4	26	69
LUMO+2	4.455	3	12	85
LUMO+1	4.078	6	14	80
LUMO	4.051	7	9	84
HOMO	1.995	52	11	37
HOMO-1	1.898	62	14	24
HOMO-2	1.159	70	18	11
HOMO-3	1.121	72	18	10
HOMO-4	0.975	76	15	9
HOMO-5	0.919	76	15	9



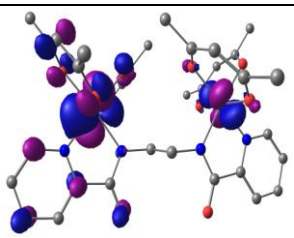
HOMO



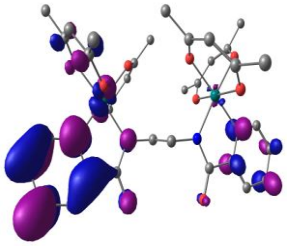
HOMO-1



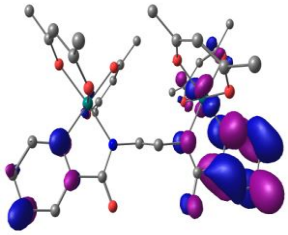
HOMO-2



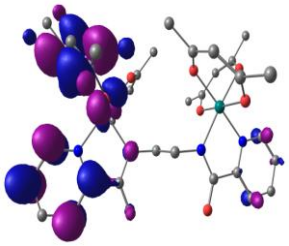
HOMO-3



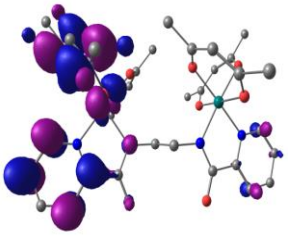
LUMO



LUMO+1



LUMO+2



LUMO+3

Table S16 Composition and energies of selected molecular orbitals of $\mathbf{3}^{2+}$ (*S*=1)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	-5.941	41	52	8
LUMO+4	-5.986	50	39	11
LUMO+3	-6.022	3	2	95
LUMO+2	-6.138	2	1	96
LUMO+1	-7.427	7	2	91
LUMO	-7.622	7	2	91
SOMO 1	-10.698	18	80	2
SOMO 2	-10.808	17	80	3
HOMO-2	-10.911	21	75	4
HOMO-3	-10.924	19	77	4
HOMO-4	-11.422	51	38	11
HOMO-5	-11.546	73	18	8
		β-spin		
LUMO+5	-5.984	3	1	95
LUMO+4	-6.107	3	2	95
LUMO+3	-7.407	9	2	89
LUMO+2	-7.596	9	2	88
LUMO+1	-8.404	68	25	6
LUMO	-8.572	69	23	8
HOMO	-10.650	25	70	5
HOMO-1	-10.684	25	70	5
HOMO-2	-10.905	41	54	5
HOMO-3	-10.955	37	57	6
HOMO-4	-11.131	51	39	10
HOMO-5	-11.291	52	37	10

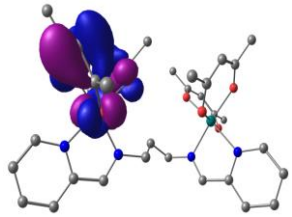
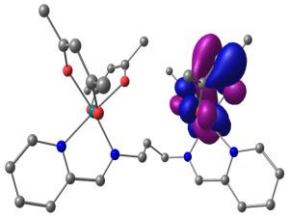
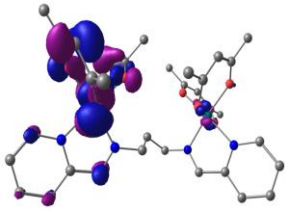
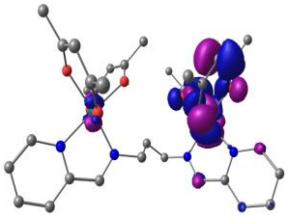
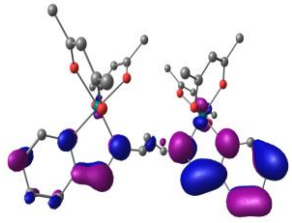
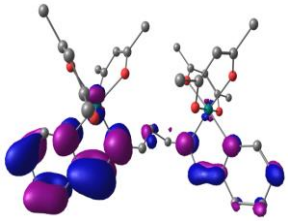
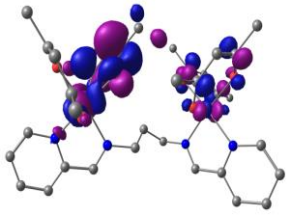
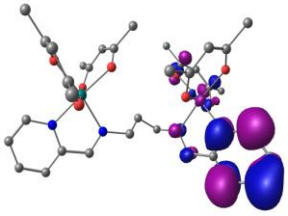
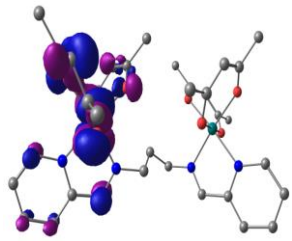
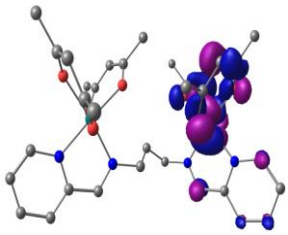
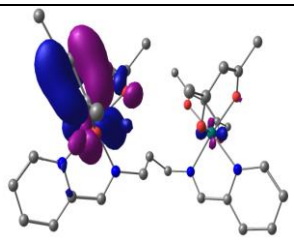
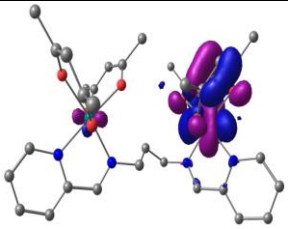
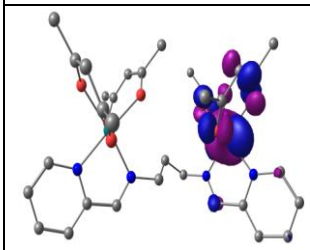
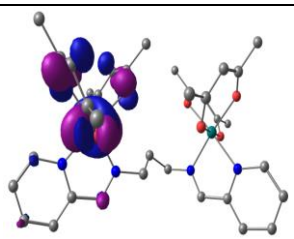
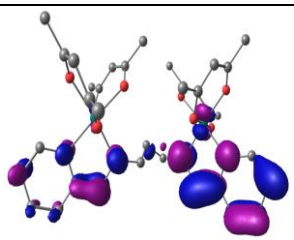
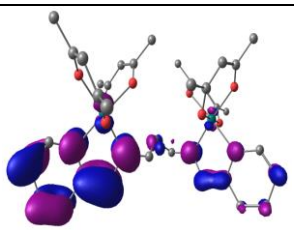
α-spin			
			
SOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S17 Composition and energies of selected molecular orbitals of $\mathbf{3}^{+}$ (S=1/2)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	-3.147	7	89	2
LUMO+4	-3.167	4	94	2
LUMO+3	-3.297	3	1	96
LUMO+2	-3.597	2	1	97
LUMO+1	-4.543	11	3	86
LUMO	-4.847	9	2	88
SOMO	-7.469	47	48	5
HOMO-1	-7.609	52	37	10
HOMO-2	-7.841	77	15	8
HOMO-3	-7.893	36	61	3
HOMO-4	-8.123	47	45	8
HOMO-5	-8.266	26	68	6
		β-spin		
LUMO+5	-3.145	3	95	2
LUMO+4	-3.293	4	2	95
LUMO+3	-3.580	3	1	95
LUMO+2	-4.532	13	3	84
LUMO+1	4.834	11	3	86
LUMO	-6.152	67	26	7
HOMO	-6.584	68	26	7
HOMO-1	-7.473	54	34	12
HOMO-2	-7.665	69	23	8
HOMO-3	-7.936	51	39	10
HOMO-4	-8.127	64	29	6
HOMO-5	-8.153	22	72	6

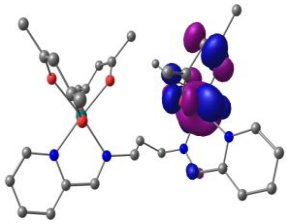
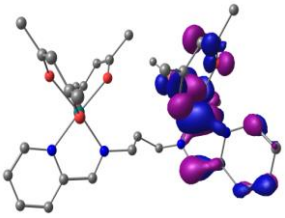
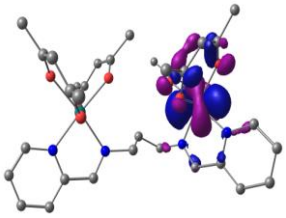
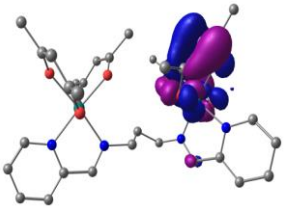
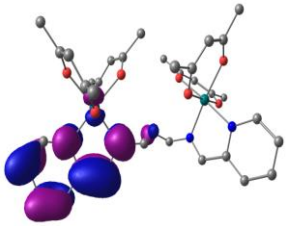
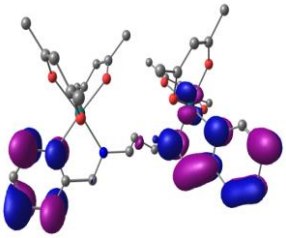
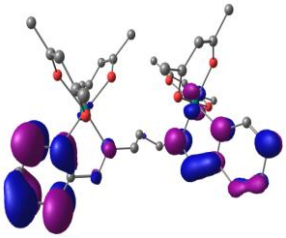
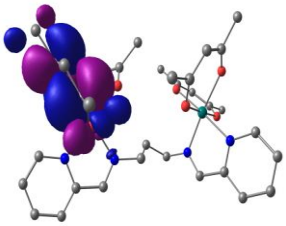
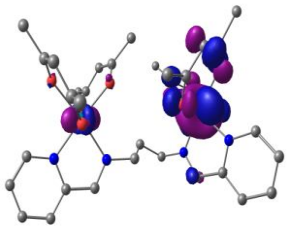
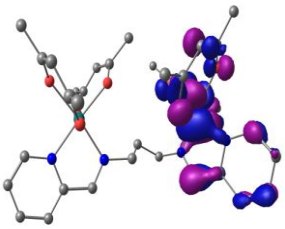
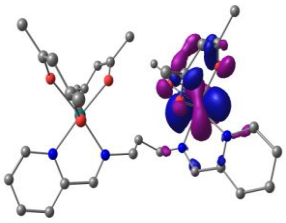
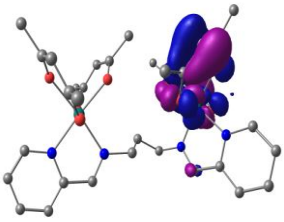
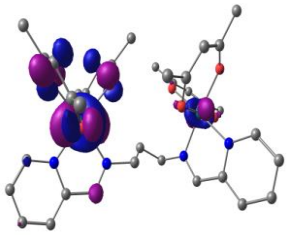
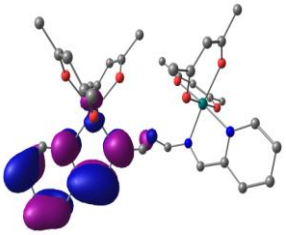
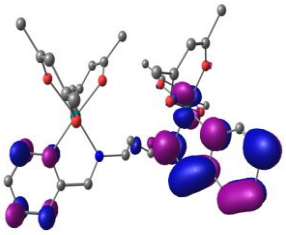
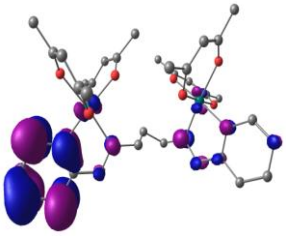
α-spin			
			
SOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S18 Composition and energies of selected molecular orbitals of **3** (*S*=0)

MO	Energy (eV)	% Composition		
		Ru	acac	L
LUMO+5	-0.324	5	91	4
LUMO+4	-0.361	4	93	3
LUMO+3	-0.702	4	1	95
LUMO+2	-0.815	4	1	95
LUMO+1	-1.713	14	4	82
LUMO	-1.893	14	3	83
HOMO	-4.034	63	30	8
HOMO-1	-4.251	65	30	5
HOMO-2	-4.559	63	21	15
HOMO-3	-4.675	67	20	13
HOMO-4	-4.736	79	15	6
HOMO-5	-4.792	73	16	12

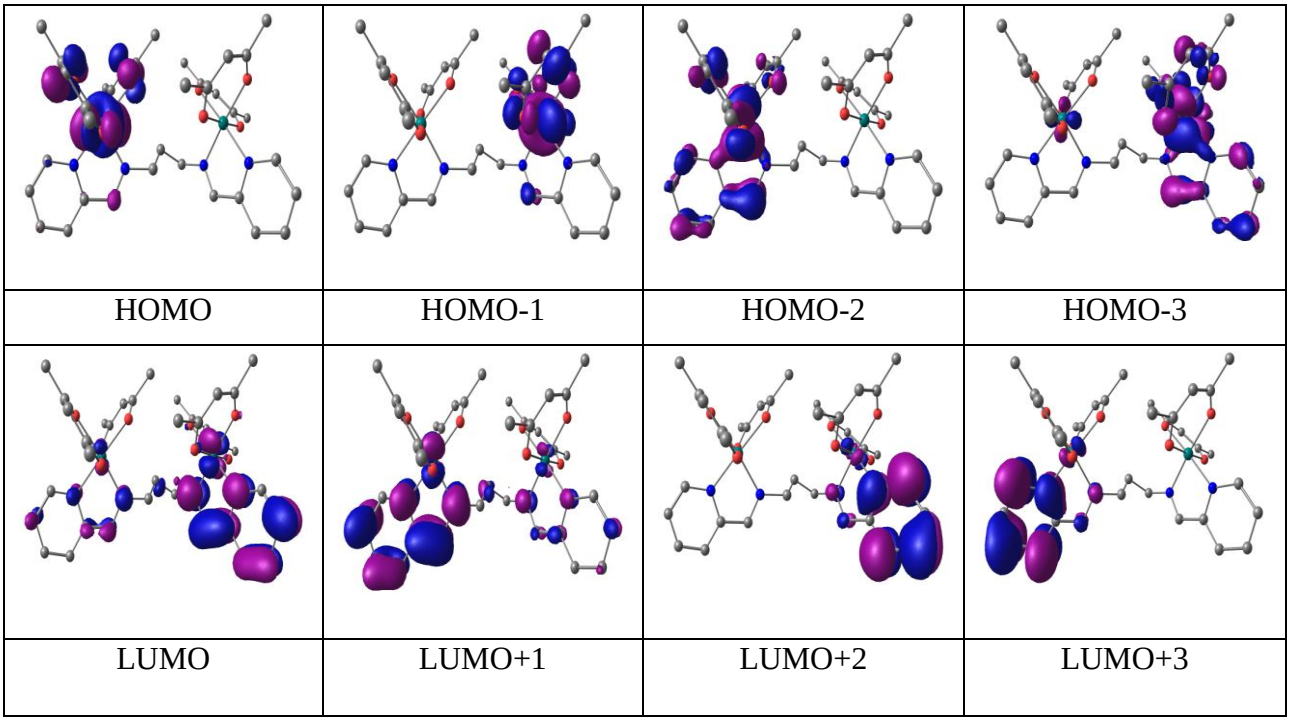


Table S19 Composition and energies of selected molecular orbitals of **3**$^{-}$ (*S*=1/2)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	2.278	5	84	10
LUMO+4	2.141	3	52	45
LUMO+3	2.111	6	58	35
LUMO+2	2.087	5	66	29
LUMO+1	1.985	3	33	64
LUMO	0.728	14	4	82
SOMO 1	0.215	14	4	82
SOMO 2	-1.471	72	21	7
HOMO-2	-1.479	70	24	6
HOMO-3	-1.863	75	16	9
HOMO-4	-1.884	67	18	15
HOMO-5	-1.970	78	15	8
		β-spin		
LUMO+5	2.196	3	38	59
LUMO+4	2.140	7	83	11
LUMO+3	2.119	6	43	51
LUMO+2	2.025	3	56	41
LUMO+1	1.467	11	7	82
LUMO	1.306	11	6	83
HOMO	-1.444	72	20	8
HOMO-1	-1.459	70	23	6
HOMO-2	-1.710	68	18	14
HOMO-3	-1.831	70	16	13
HOMO-4	-1.847	76	16	8
HOMO-5	-1.951	78	15	7

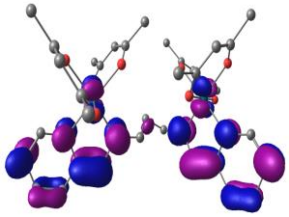
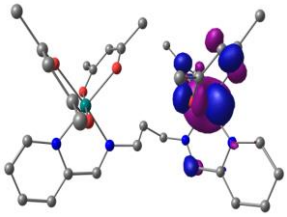
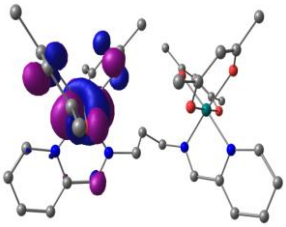
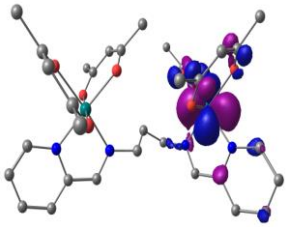
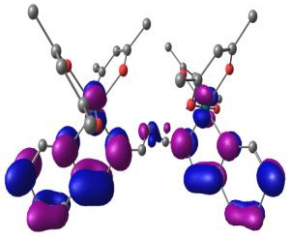
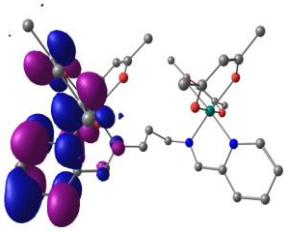
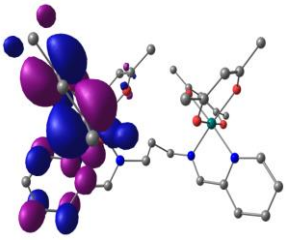
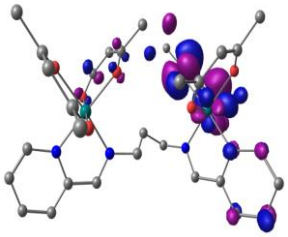
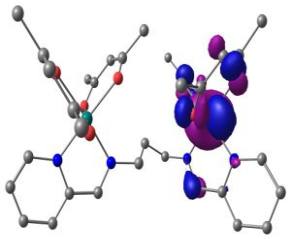
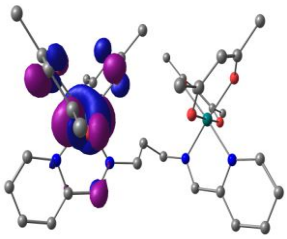
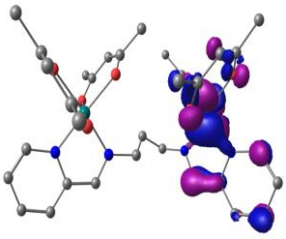
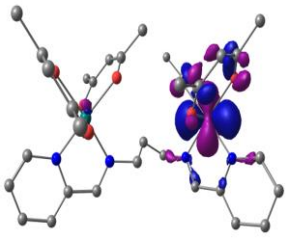
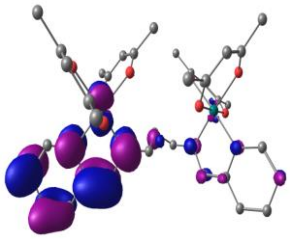
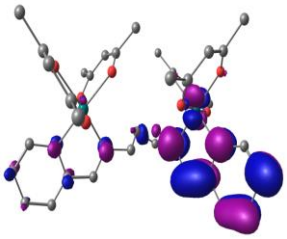
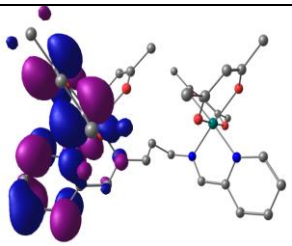
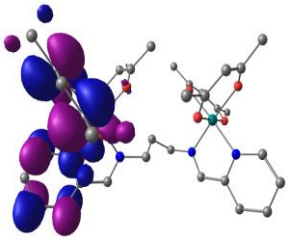
α-spin			
			
SOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S20 Composition and energies of selected molecular orbitals of $\mathbf{3}^{2-}$ (*S*=1)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	5.037	8	3	89
LUMO+4	4.949	9	5	86
LUMO+3	4.801	6	91	3
LUMO+2	4.640	6	88	6
LUMO+1	4,511	4	92	3
LUMO	4.457	6	92	3
SOMO 1	2.708	13	4	83
SOMO 2	2.490	13	4	83
HOMO-2	1.318	75	19	6
HOMO-3	1.148	76	17	7
HOMO-4	0.888	79	15	6
HOMO-5	0.790	75	15	10
		β-spin		
LUMO+5	4.856	8	80	11
LUMO+4	4.671	6	80	13
LUMO+3	4.624	9	61	30
LUMO+2	4.540	10	71	19
LUMO+1	4.436	3	65	32
LUMO	4.270	5	34	60
HOMO	1.376	74	18	9
HOMO-1	1.217	74	16	10
HOMO-2	1.091	74	15	12
HOMO-3	1.000	72	16	12
HOMO-4	0.930	78	15	7
HOMO-5	0.831	77	15	7

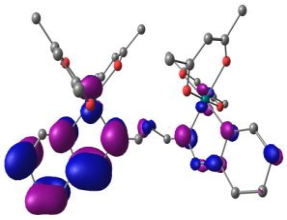
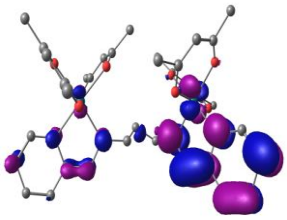
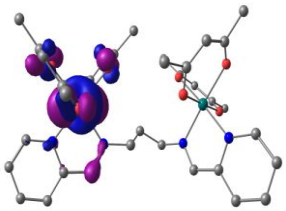
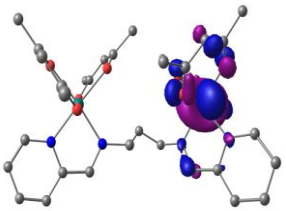
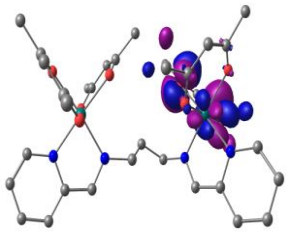
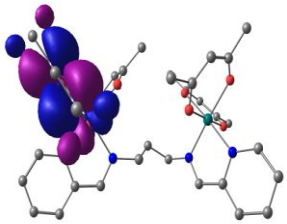
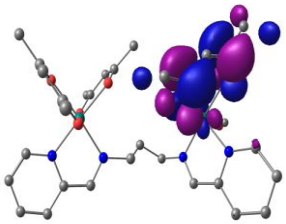
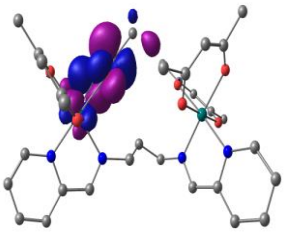
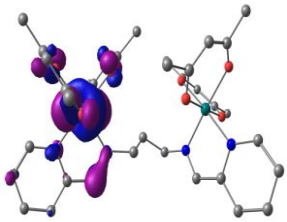
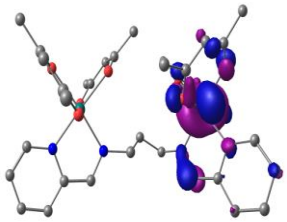
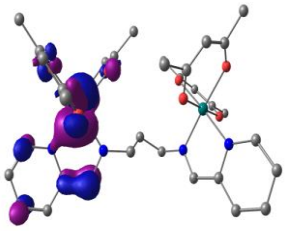
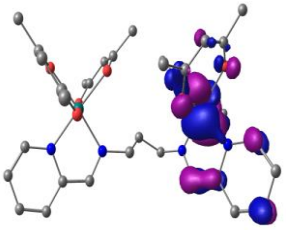
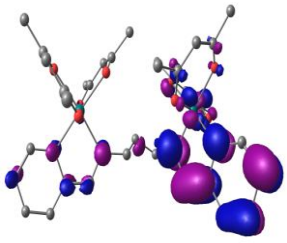
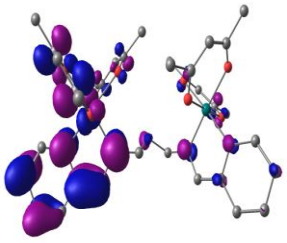
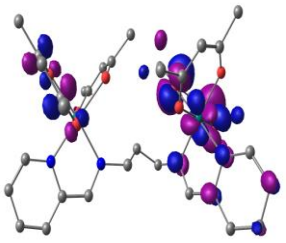
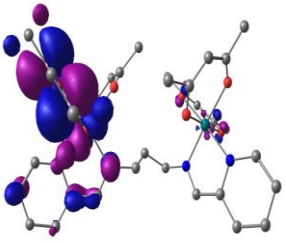
α-spin			
			
SOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S21 Composition and energies of selected molecular orbitals of **3a**$^{4+}$ (*S*=1)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	-11.340	13	50	37
LUMO+4	-11.365	14	57	29
LUMO+3	-11.477	35	56	9
LUMO+2	-11.491	35	54	10
LUMO+1	-14.590	44	42	14
LUMO	-14.647	42	48	10
SOMO 1	-16.301	22	41	37
SOMO 2	-16.407	25	43	32
HOMO-2	-16.626	21	71	8
HOMO-3	-16.654	23	69	8
HOMO-4	-16.863	4	3	94
HOMO-5	-16.885	3	2	94
		β-spin		
LUMO+5	-11.435	20	71	9
LUMO+4	-11.436	19	73	8
LUMO+3	-14.092	42	11	46
LUMO+2	-14.282	46	13	41
LUMO+1	-14.458	47	49	4
LUMO	-14.496	45	51	4
HOMO	-16.431	27	66	7
HOMO-1	-16.448	30	61	9
HOMO-2	-16.656	16	52	32
HOMO-3	-16.713	13	49	38
HOMO-4	-16.811	9	10	81
HOMO-5	-16.833	10	7	83

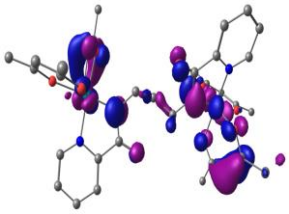
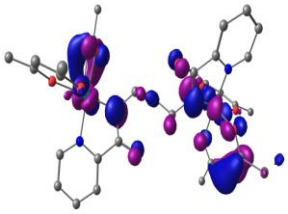
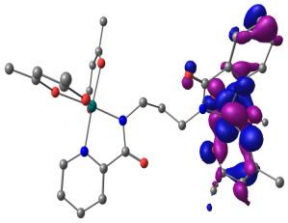
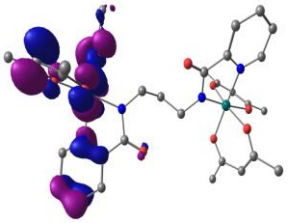
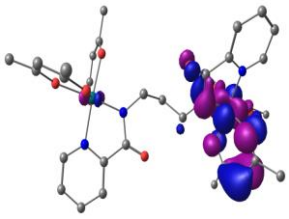
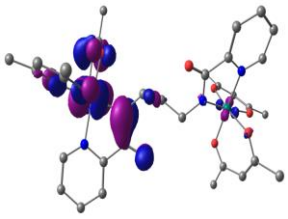
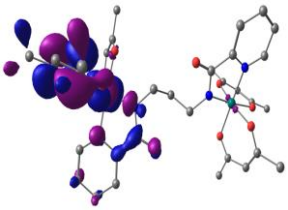
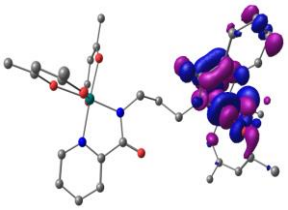
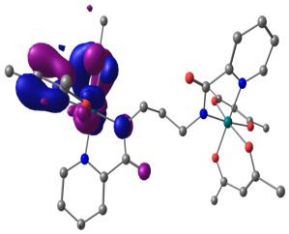
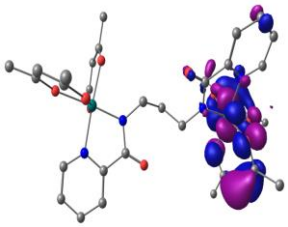
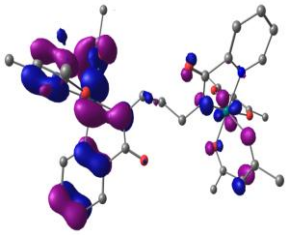
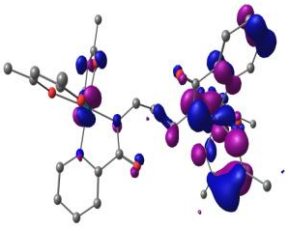
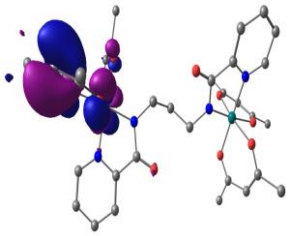
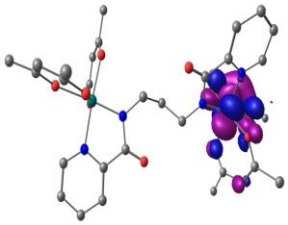
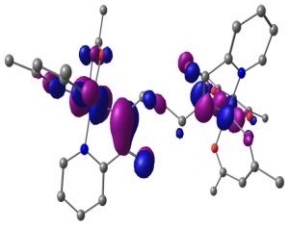
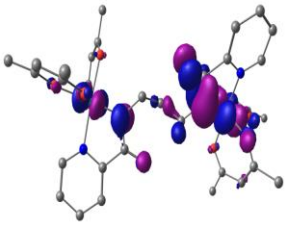
α-spin			
			
SOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S22 Composition and energies of selected molecular orbitals of **3a**$^{3+}$ (*S*=3/2)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	-8.753	39	49	12
LUMO+4	-8.963	5	89	6
LUMO+3	-9.188	16	49	35
LUMO+2	-9.233	17	26	57
LUMO+1	-9.524	45	41	14
LUMO	-12.600	36	26	37
SOMO 1	-13.067	32	48	20
SOMO 2	-13.535	22	37	40
SOMO 3	-13.707	2	85	13
HOMO-3	-14.173	9	59	32
HOMO-4	-14.243	15	21	64
HOMO-5	-14.350	14	8	78
		β-spin		
LUMO+5	-9.121	20	30	50
LUMO+4	-9.334	26	42	33
LUMO+3	-10.994	56	22	22
LUMO+2	-11.338	58	29	13
LUMO+1	-11.715	52	23	25
LUMO	-12.428	54	37	9
HOMO	-13.437	2	89	9
HOMO-1	-13.654	36	48	16
HOMO-2	-14.048	39	14	46
HOMO-3	-14.131	5	87	8
HOMO-4	-14.250	11	28	61
HOMO-5	-14.319	7	9	84

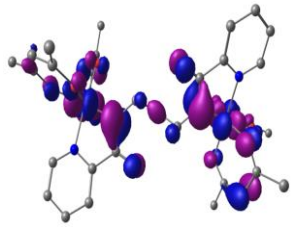
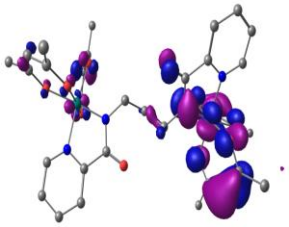
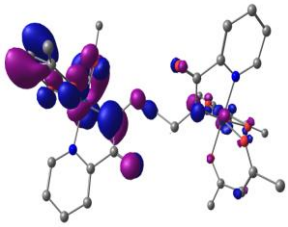
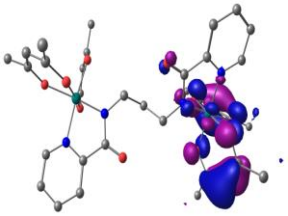
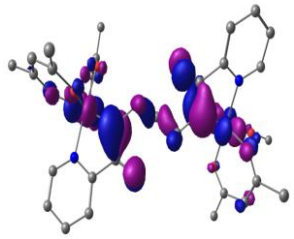
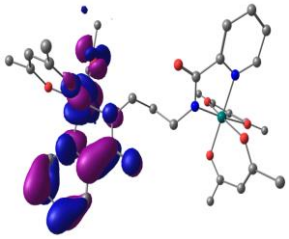
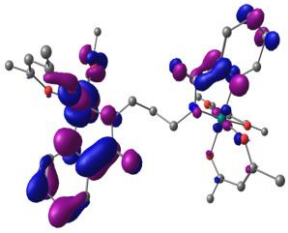
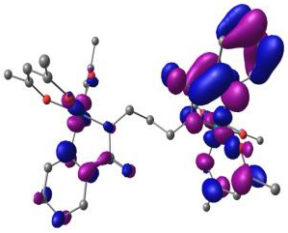
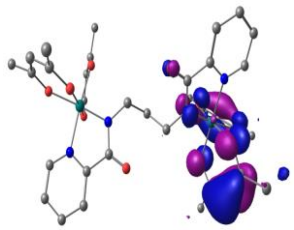
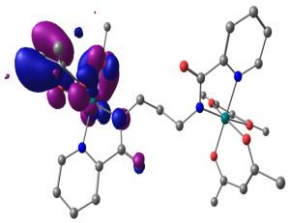
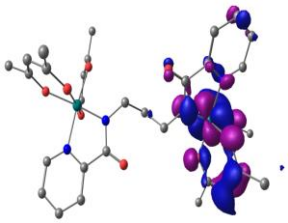
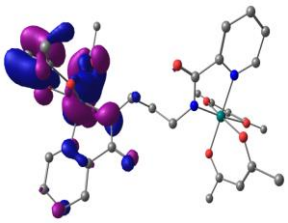
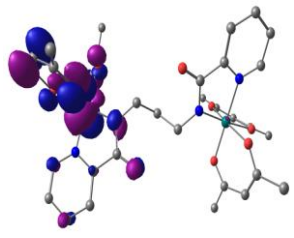
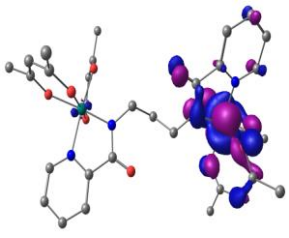
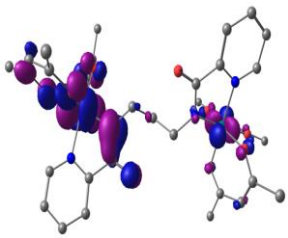
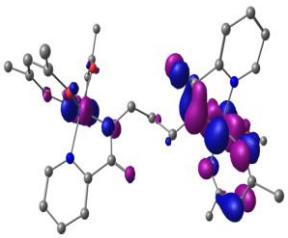
α-spin			
			
SOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S23 Composition and energies of selected molecular orbitals of **3a**$^{2+}$ (*S*=1)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	-6.384	6	90	4
LUMO+4	-6.529	5	19	76
LUMO+3	-6.570	18	66	16
LUMO+2	-6.755	51	39	10
LUMO+1	-6.841	3	2	95
LUMO	-9.164	52	22	26
SOMO 1	-11.030	34	51	15
SOMO 2	-11.389	23	65	12
SOMO 3	-11.470	2	84	14
HOMO-3	-11.688	52	20	28
HOMO-4	-11.731	29	45	26
HOMO-5	-11.794	18	65	18
		β-spin		
LUMO+5	-6.485	11	80	9
LUMO+4	-6.531	5	18	78
LUMO+3	-6.669	4	10	87
LUMO+2	-8.923	54	16	30
LUMO+1	-9.166	53	22	24
LUMO	-9.485	59	26	15
HOMO	-11.033	35	51	14
HOMO-1	-11.332	10	75	15
HOMO-2	-11.491	4	85	11
HOMO-3	-11.683	47	41	11
HOMO-4	-11.717	50	34	16
HOMO-5	-11.960	43	25	32

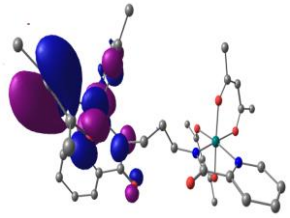
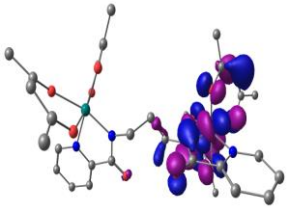
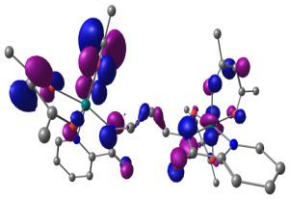
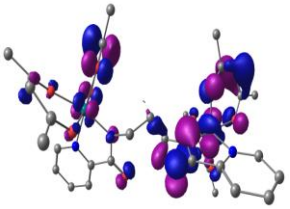
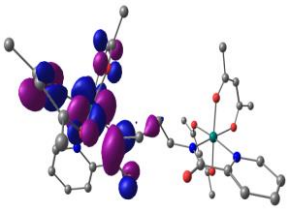
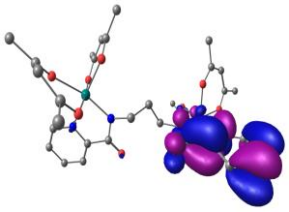
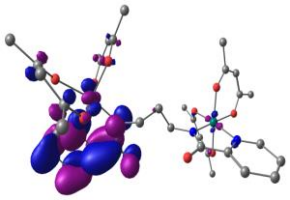
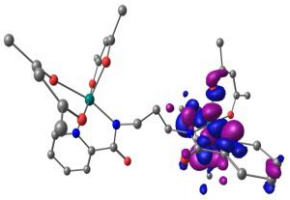
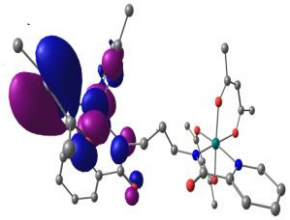
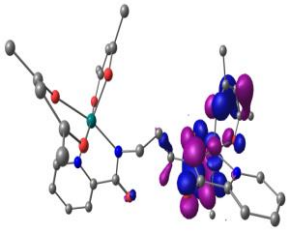
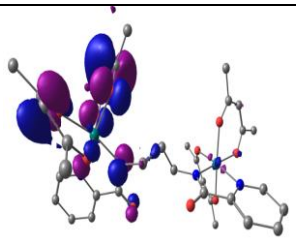
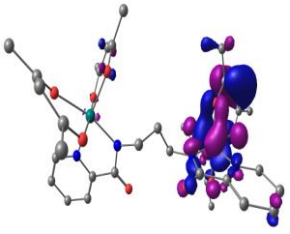
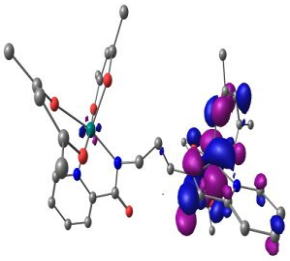
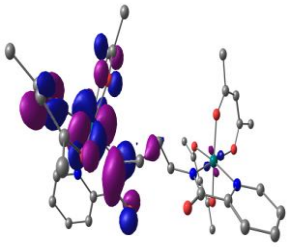
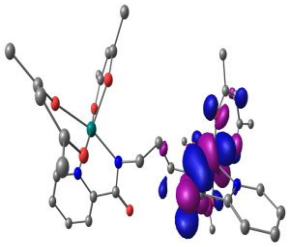
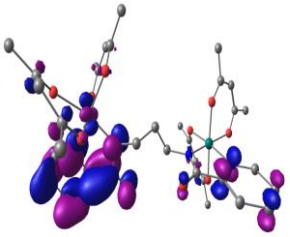
α-spin			
			
SOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S24 Composition and energies of selected molecular orbitals of **3a**$^{+}$ (*S*=1/2)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	-3.494	4	6	90
LUMO+4	-3.684	5	90	6
LUMO+3	-3.767	16	78	7
LUMO+2	-3.897	3	2	94
LUMO+1	-4.168	4	4	93
LUMO	-7.084	37	10	53
SOMO	-7.788	46	15	40
HOMO-1	-8.167	33	62	5
HOMO-2	-8.481	22	65	13
HOMO-3	-8.568	30	66	5
HOMO-4	-8.830	53	35	12
HOMO-5	-8.881	13	73	14
		β-spin		
LUMO+5	-3.662	5	88	8
LUMO+4	-3.733	10	85	5
LUMO+3	-3.923	3	2	94
LUMO+2	-4.243	3	3	94
LUMO+1	-5.635	68	17	15
LUMO	-6.152	67	21	12
HOMO	-8.001	46	40	14
HOMO-1	-8.348	41	49	9
HOMO-2	-8.483	42	43	15
HOMO-3	-8.640	36	51	12
HOMO-4	-8.834	21	70	9
HOMO-5	-8.984	52	35	13

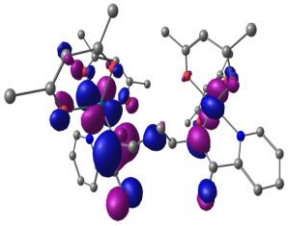
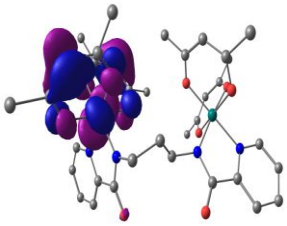
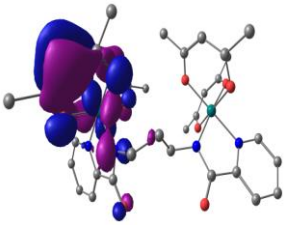
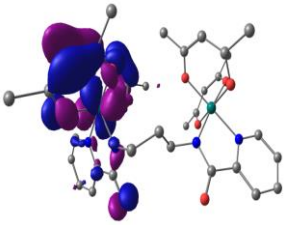
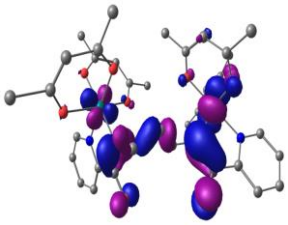
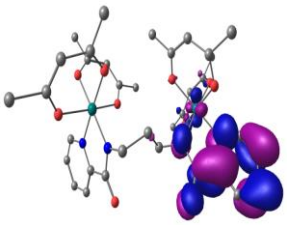
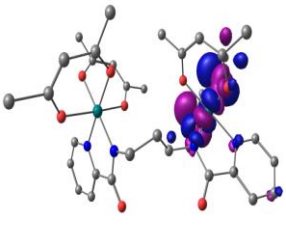
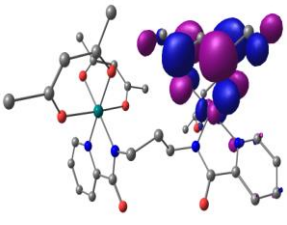
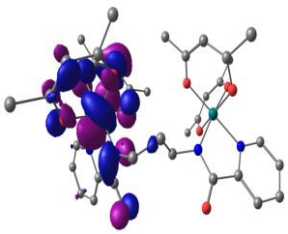
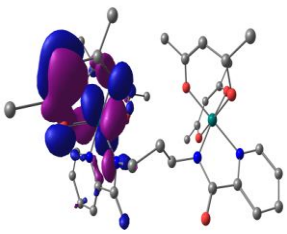
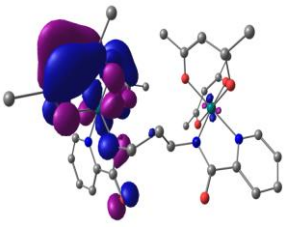
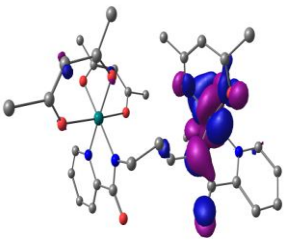
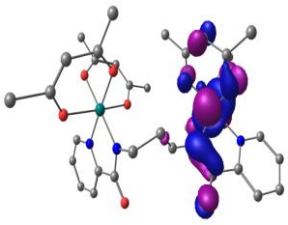
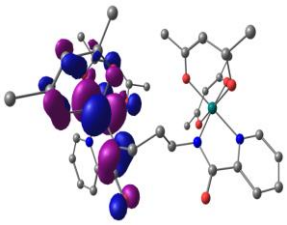
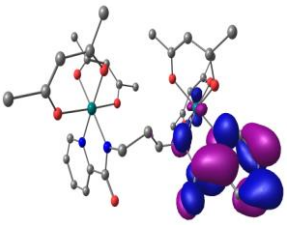
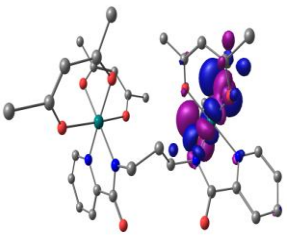
α-spin			
			
SOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S25 Composition and energies of selected molecular orbitals of **3a** (*S*=1)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	-0.691	3	4	94
LUMO+4	-0.811	6	88	6
LUMO+3	-0.906	5	89	7
LUMO+2	-0.956	4	90	7
LUMO+1	-1.215	4	6	90
LUMO	-1.244	4	7	89
SOMO 1	-4.930	36	32	32
SOMO 2	-5.099	37	36	27
SOMO 3	-5.264	46	16	38
HOMO-3	-5.346	49	37	14
HOMO-4	-5.675	48	20	32
HOMO-5	-5.712	63	22	15
		β-spin		
LUMO+5	-0.863	8	80	12
LUMO+4	-0.890	5	86	9
LUMO+3	-1.166	5	10	86
LUMO+2	-1.200	4	12	84
LUMO+1	-2.359	62	17	20
LUMO	-2.537	65	17	18
HOMO	-4.820	41	38	21
HOMO-1	-5.010	57	28	15
HOMO-2	-5.420	68	19	13
HOMO-3	-5.427	66	16	18
HOMO-4	-5.654	26	65	9
HOMO-5	-5.744	18	25	57

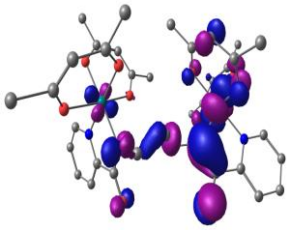
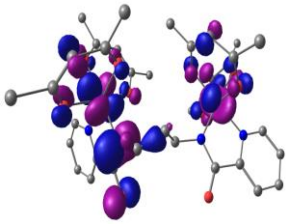
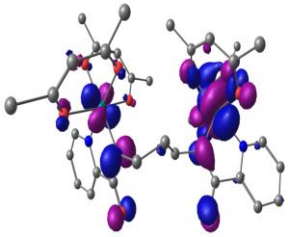
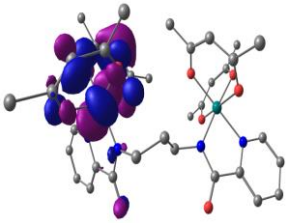
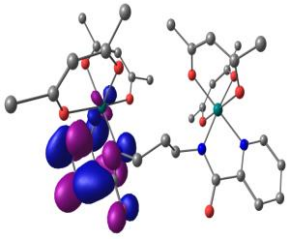
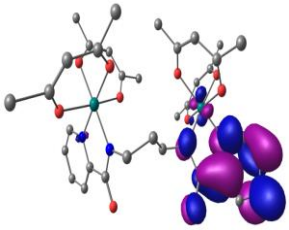
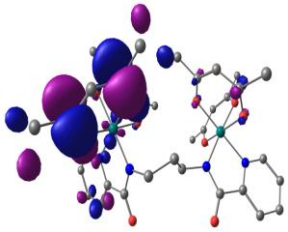
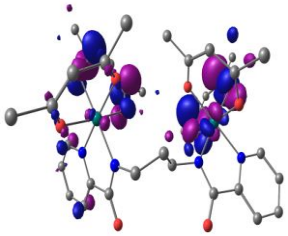
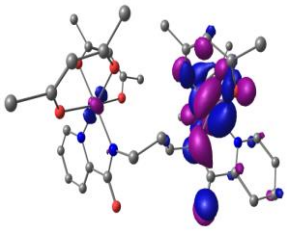
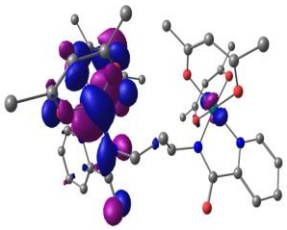
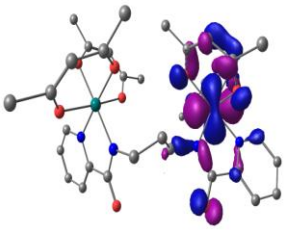
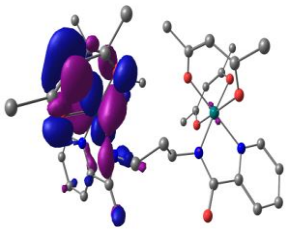
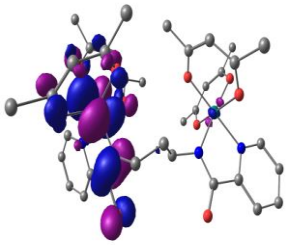
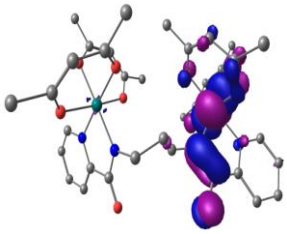
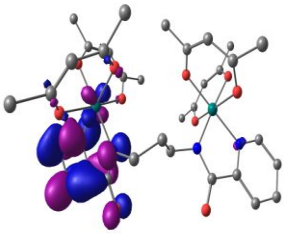
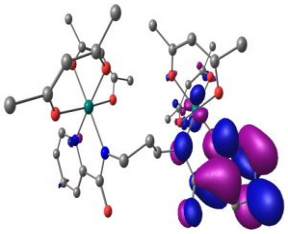
α-spin			
			
SOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S26 Composition and energies of selected molecular orbitals of **3a**$^{2-}$ (*S*=0)

MO	Energy (eV)	% Composition		
		Ru	acac	L
LUMO+5	4.475	6	81	13
LUMO+4	4.440	6	53	42
LUMO+3	4.419	5	41	54
LUMO+2	4.319	5	10	85
LUMO+1	4.003	6	13	81
LUMO	3.916	7	7	86
HOMO	1.911	57	12	30
HOMO-1	1.701	59	13	28
HOMO-2	1.079	70	18	11
HOMO-3	1.012	69	20	11
HOMO-4	0.873	77	14	9
HOMO-5	0.781	76	15	9

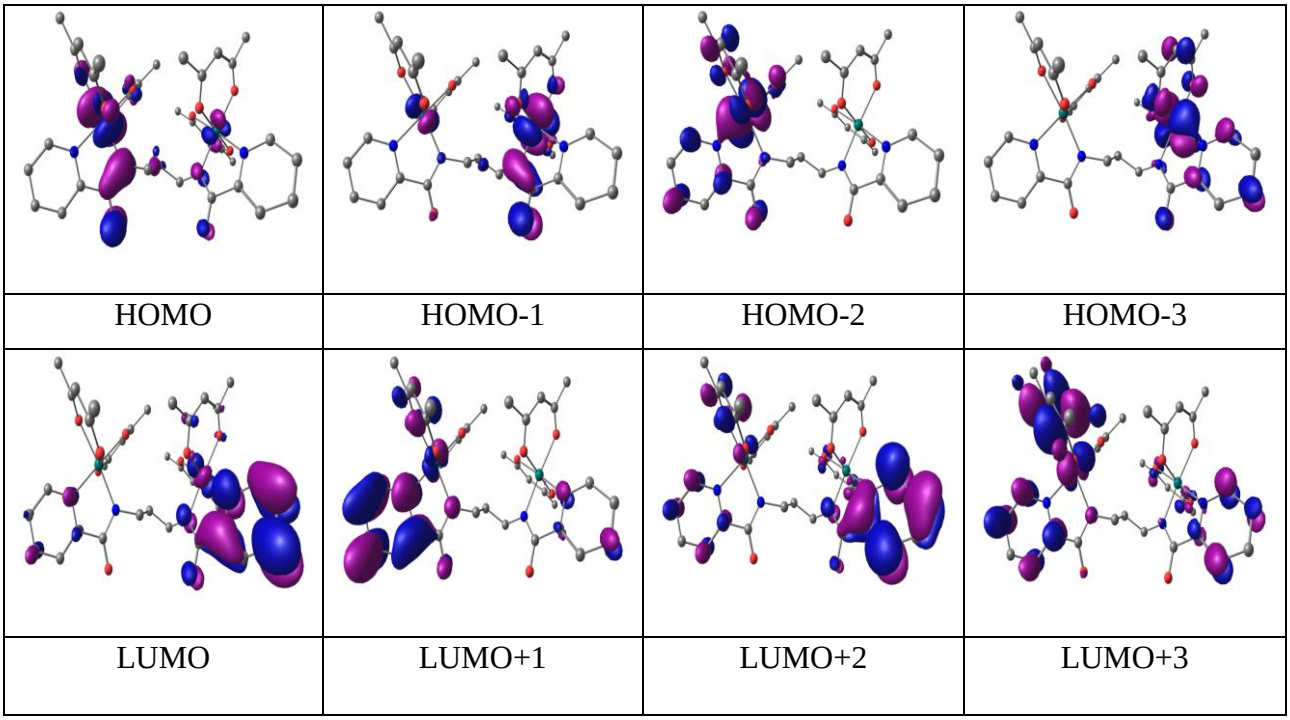


Table S27 Composition and energies of selected molecular orbitals of $\mathbf{4}^{2+}$ (*S*=1)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	-5.715	5	91	4
LUMO+4	-5.746	5	89	6
LUMO+3	-5.903	3	2	95
LUMO+2	-5.954	2	2	95
LUMO+1	-7.227	7	2	91
LUMO	-7.273	7	3	90
SOMO 1	-10.499	30	67	3
SOMO 2	-10.580	29	68	3
HOMO-2	-10.816	43	50	6
HOMO-3	-10.908	50	42	8
HOMO-4	-11.219	74	19	7
HOMO-5	-11.254	72	20	8
		β-spin		
LUMO+5	-5.867	4	2	93
LUMO+4	-5.925	3	5	92
LUMO+3	-7.200	10	3	88
LUMO+2	-7.253	9	3	88
LUMO+1	-7.941	63	30	7
LUMO	-8.028	65	27	8
HOMO	-10.544	51	41	8
HOMO-1	-10.623	58	33	9
HOMO-2	-10.799	60	33	7
HOMO-3	-10.804	59	32	7
HOMO-4	-11.029	21	74	5
HOMO-5	-11.080	27	67	6

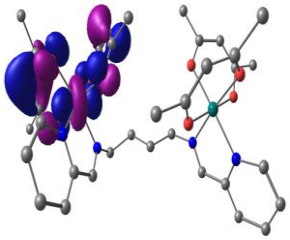
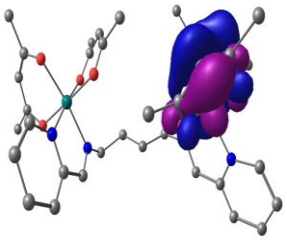
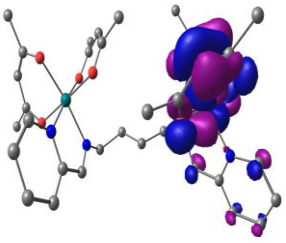
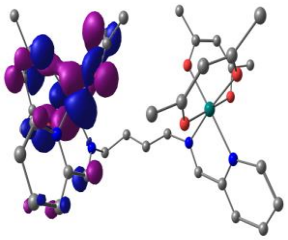
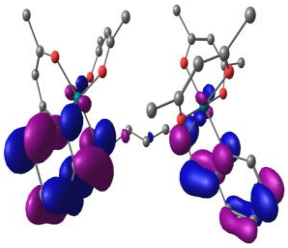
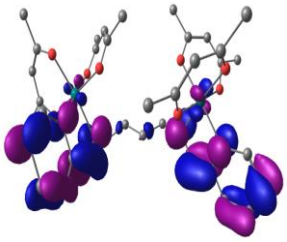
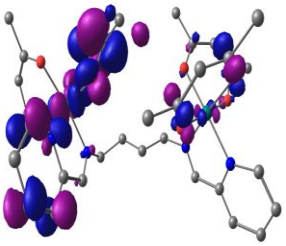
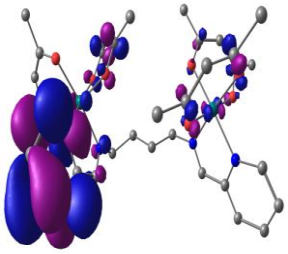
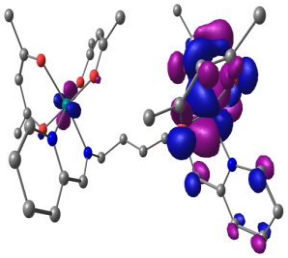
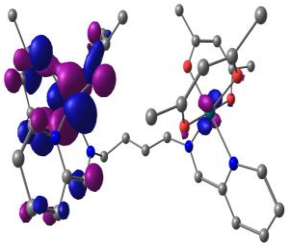
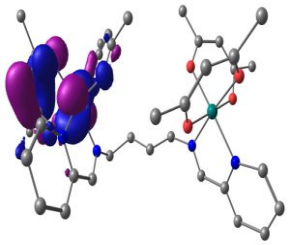
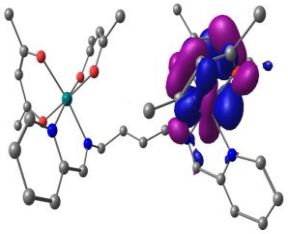
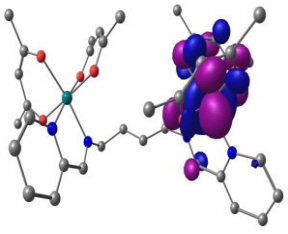
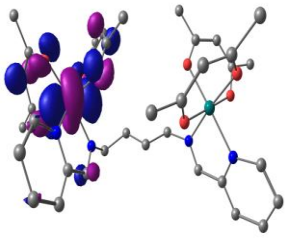
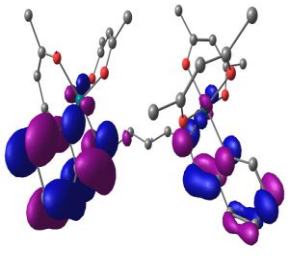
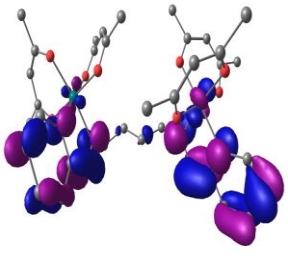
α-spin			
			
SOMO 1	SOMO 2	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S28 Composition and energies of selected molecular orbitals of **4** (*S*=0)

MO	Energy (eV)	% Composition		
		Ru	acac	L
LUMO+5	-0.362	4	92	3
LUMO+4	-0.405	5	91	4
LUMO+3	-0.719	4	2	94
LUMO+2	-0,722	4	2	94
LUMO+1	-1.702	13	4	83
LUMO	-1.734	13	4	83
HOMO	-3.806	62	33	5
HOMO-1	-3.835	63	32	5
HOMO-2	-4.468	67	17	16
HOMO-3	-4.473	68	17	15
HOMO-4	-4.602	78	15	6
HOMO-5	-4.603	78	16	6

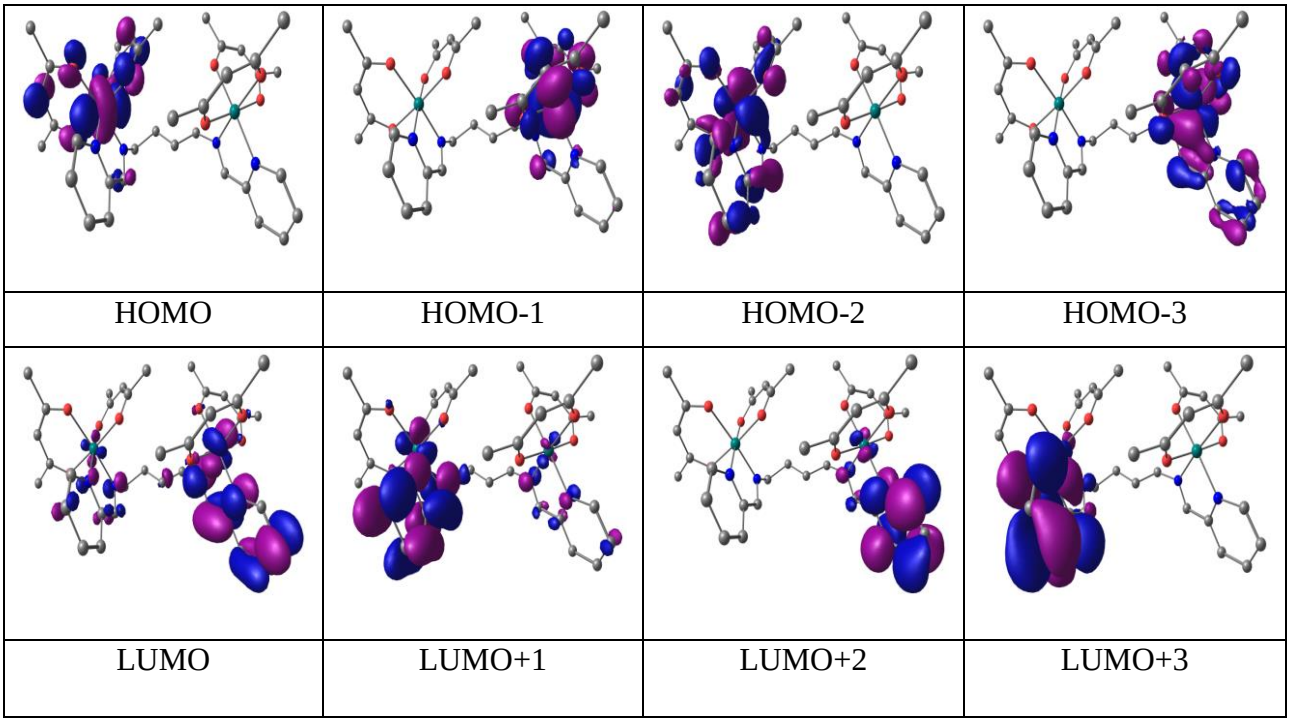


Table S29 Composition and energies of selected molecular orbitals of **4**$^{-}$ (*S*=1/2)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	2.251	5	91	4
LUMO+4	2.099	6	28	66
LUMO+3	2.078	3	30	67
LUMO+2	2.049	6	72	23
LUMO+1	2.003	3	76	21
LUMO	0.656	14	4	82
SOMO	0.223	14	4	82
HOMO-1	-1.505	71	23	7
HOMO-2	-1.512	71	22	7
HOMO-3	-1.932	71	17	11
HOMO-4	-1.960	72	17	12
HOMO-5	-1.968	74	16	9
		β-spin		
LUMO+5	2.155	7	13	81
LUMO+4	2.134	3	20	76
LUMO+3	2.069	6	87	7
LUMO+2	2.023	4	84	12
LUMO+1	1.376	12	7	82
LUMO	1.331	11	7	82
HOMO	-1.472	70	21	9
HOMO-1	-1.481	71	22	7
HOMO-2	-1.779	68	18	14
HOMO-3	-1.818	69	17	14
HOMO-4	-1.922	78	15	7
HOMO-5	-1.949	78	15	7

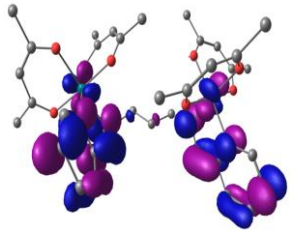
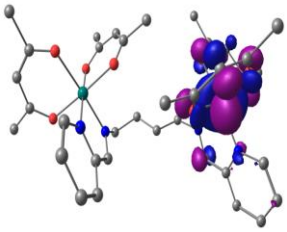
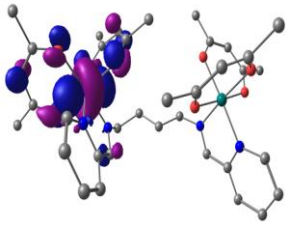
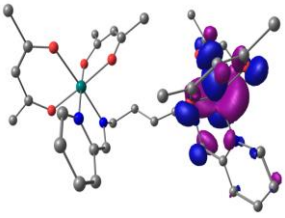
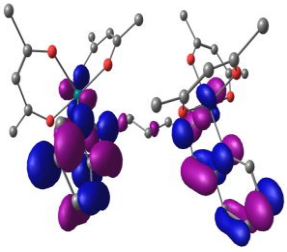
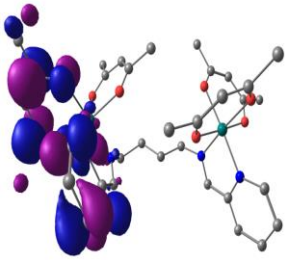
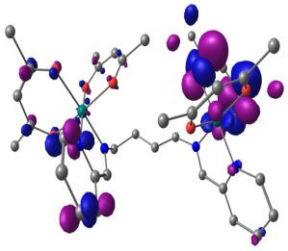
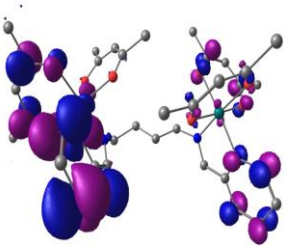
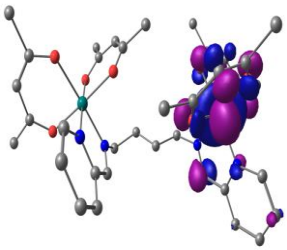
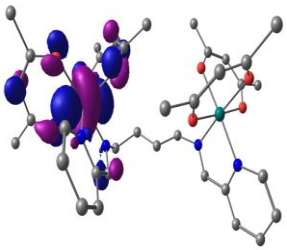
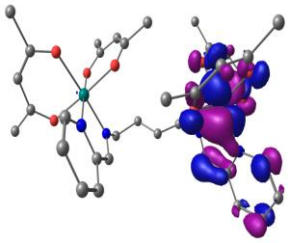
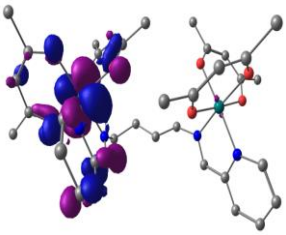
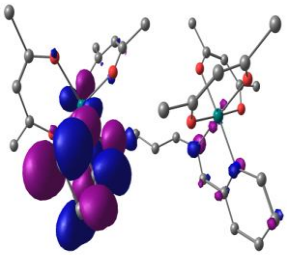
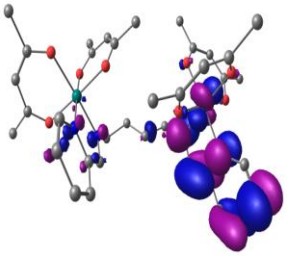
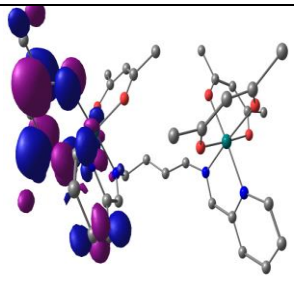
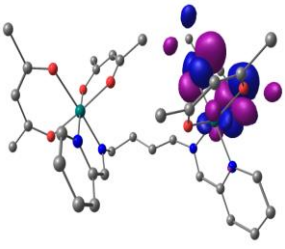
α-spin			
			
SOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S30 Composition and energies of selected molecular orbitals of $\mathbf{4}^{2-}$ (*S*=1)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	4.985	9	6	85
LUMO+4	4.969	12	8	80
LUMO+3	4.738	5	84	11
LUMO+2	4.670	6	90	4
LUMO+1	4.457	6	92	2
LUMO	4.341	5	92	3
SOMO 1	2.751	13	5	82
SOMO 2	2.714	13	5	82
HOMO-2	1.269	74	18	8
HOMO-3	1.229	74	19	7
HOMO-4	0.864	77	16	7
HOMO-5	0.789	78	16	7
		β-spin		
LUMO+5	4.808	8	71	21
LUMO+4	4.774	10	69	22
LUMO+3	4.605	12	47	41
LUMO+2	4.531	10	48	42
LUMO+1	4.399	3	72	25
LUMO	4.305	3	82	16
HOMO	1.352	72	17	11
HOMO-1	1.283	73	18	9
HOMO-2	1.086	72	16	11
HOMO-3	1.003	73	15	12
HOMO-4	0.907	77	16	7
HOMO-5	0.826	77	16	7

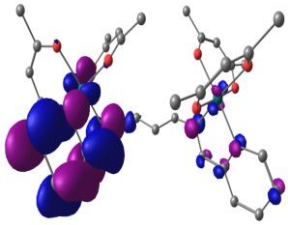
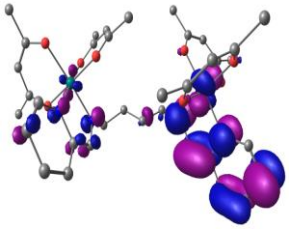
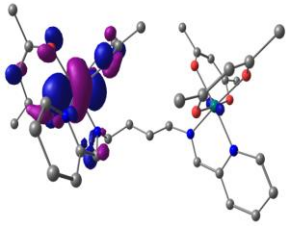
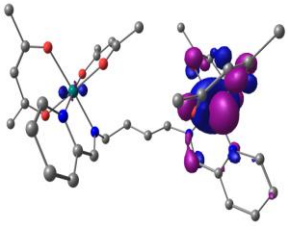
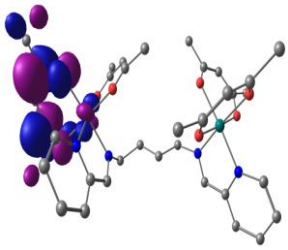
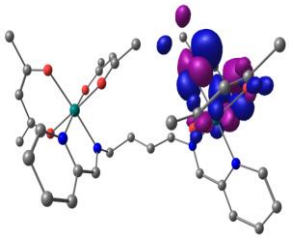
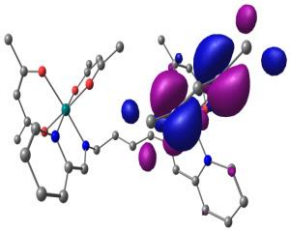
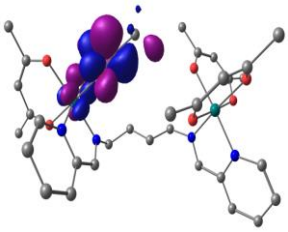
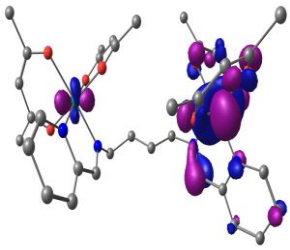
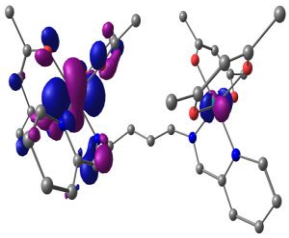
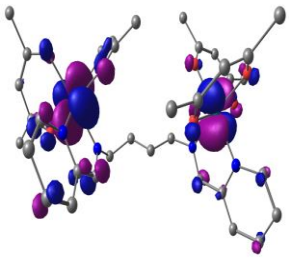
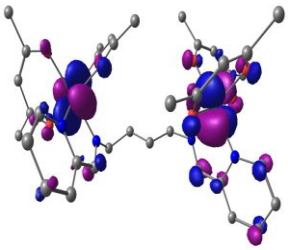
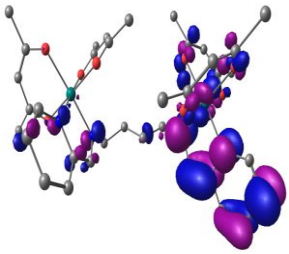
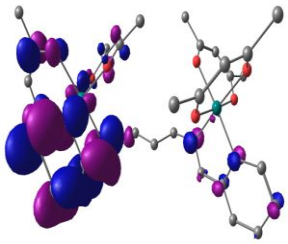
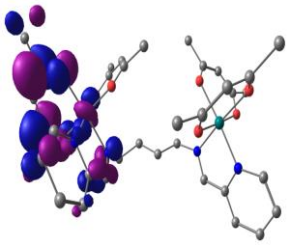
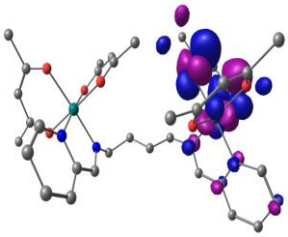
α-spin			
			
SOMO 1	SOMO 2	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S31 Composition and energies of selected molecular orbitals of **4a**$^{4+}$ (*S*=1)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	-11.415	28	61	11
LUMO+4	-11.418	29	60	11
LUMO+3	-11.755	42	51	7
LUMO+2	-11.784	41	51	8
LUMO+1	-14.805	42	43	15
LUMO	-14.824	42	45	13
SOMO 1	-16.417	18	49	33
SOMO 2	-16.490	20	53	26
HOMO-2	-16.730	3	4	92
HOMO-3	-16.748	3	5	92
HOMO-4	-16.863	23	55	22
HOMO-5	-16.883	26	54	20
		β-spin		
LUMO+5	-11.618	39	54	7
LUMO+4	-11.650	38	55	7
LUMO+3	-14.257	39	11	49
LUMO+2	-14.359	42	14	44
LUMO+1	-14.797	47	45	8
LUMO	-14.824	48	42	10
HOMO	-16.565	32	42	26
HOMO-1	-16.581	31	45	24
HOMO-2	-16.657	8	5	87
HOMO-3	-16.674	7	4	89
HOMO-4	-16.811	11	74	15
HOMO-5	-16.865	6	67	28

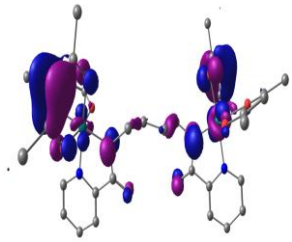
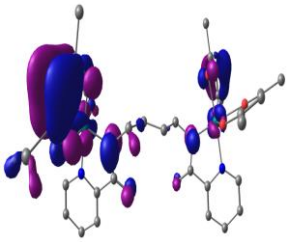
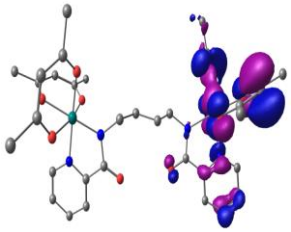
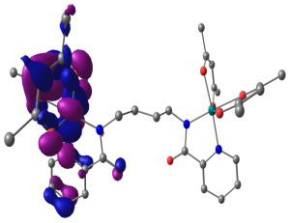
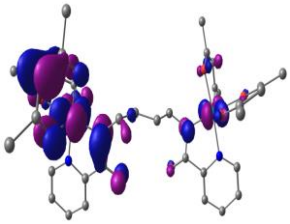
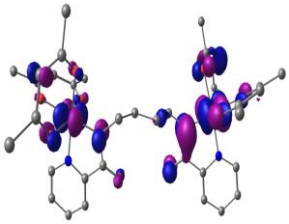
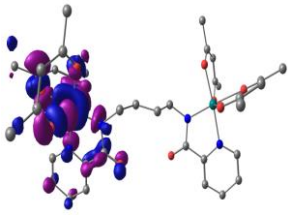
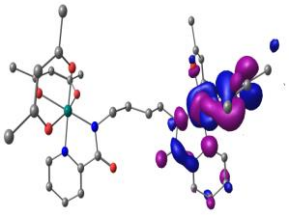
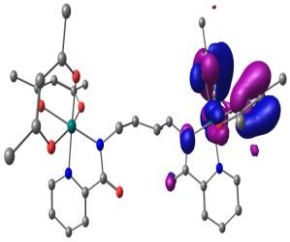
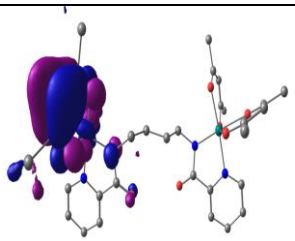
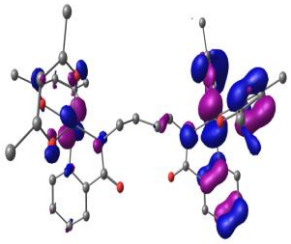
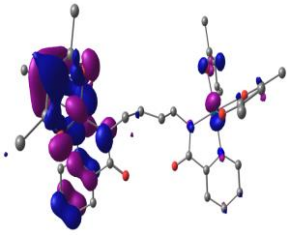
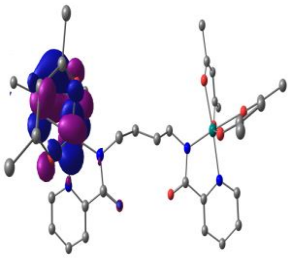
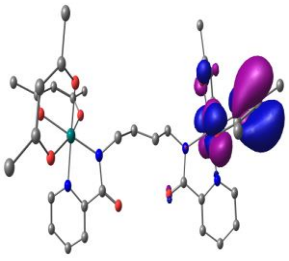
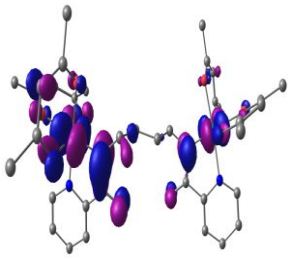
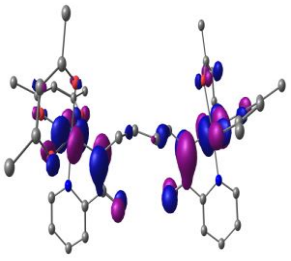
α-spin			
			
SOMO 1	SOMO 2	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S32 Composition and energies of selected molecular orbitals of **4a**$^{3+}$ (*S*=3/2)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	-8.594	34	58	8
LUMO+4	-8.645	27	63	10
LUMO+3	-8.864	14	64	22
LUMO+2	-8.984	6	26	68
LUMO+1	-9.164	25	64	12
LUMO	-12.008	53	26	21
SOMO 1	-13.230	30	55	15
SOMO 2	-13.469	27	38	35
SOMO 3	-13.836	4	70	26
HOMO-3	-13.953	23	35	42
HOMO-4	-13.991	20	32	48
HOMO-5	-14.139	4	2	94
		β-spin		
LUMO+5	-8.918	8	46	46
LUMO+4	-9.107	24	65	11
LUMO+3	-10.560	54	18	28
LUMO+2	-11.016	57	29	13
LUMO+1	-11.933	44	27	28
LUMO	-12.642	31	44	25
HOMO	-13.040	18	61	21
HOMO-1	-13.349	36	53	11
HOMO-2	-13.713	37	15	48
HOMO-3	-13.927	19	39	42
HOMO-4	-13.985	4	83	13
HOMO-5	-14.051	2	3	95

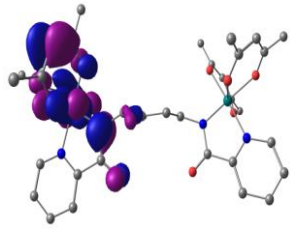
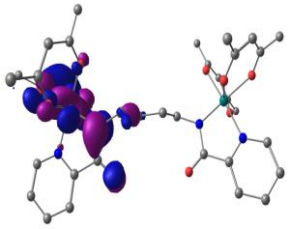
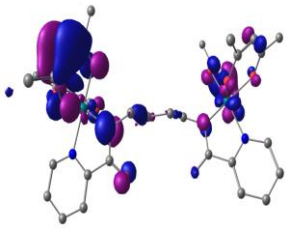
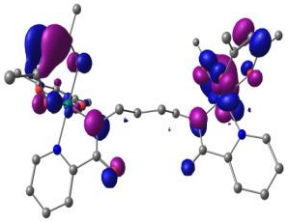
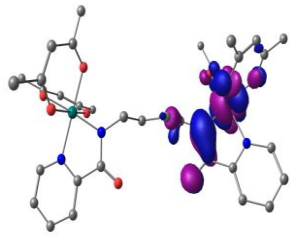
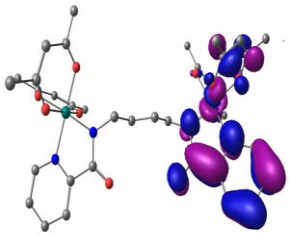
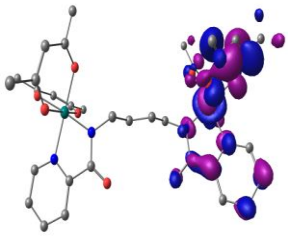
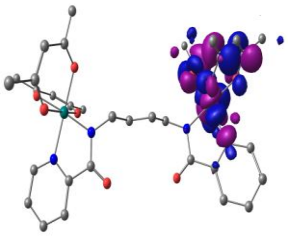
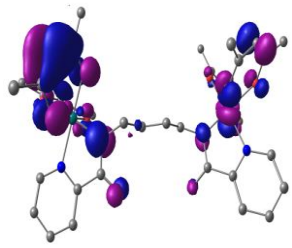
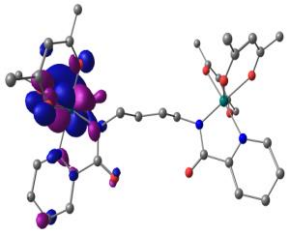
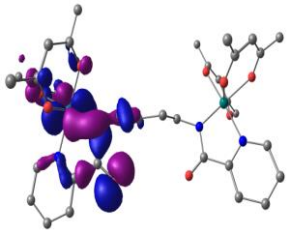
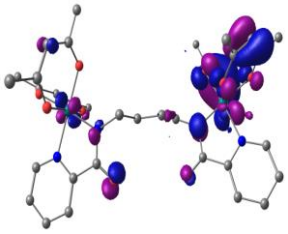
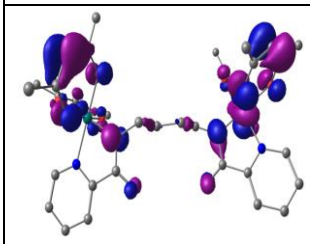
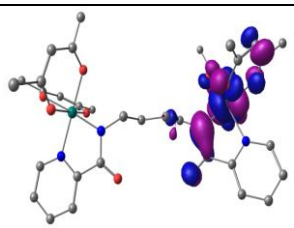
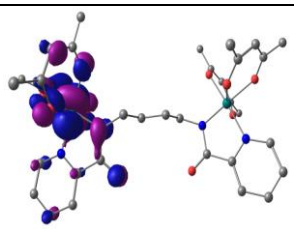
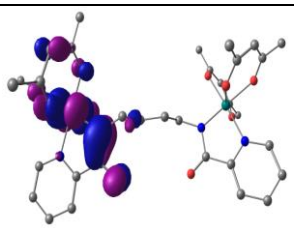
α-spin			
			
SOMO 1	SOMO 1	SOMO 1	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S33 Composition and energies of selected molecular orbitals of **4a**$^{2+}$ (*S*=1)

MO	Energy (eV)	% Composition		
		Ru	acac	L
	α-spin			
LUMO+5	-6.157	20	69	10
LUMO+4	-6.208	24	62	14
LUMO+3	-6.398	18	20	62
LUMO+2	-6.415	16	18	66
LUMO+1	-6.553	23	25	52
LUMO	-8.855	58	31	11
SOMO 1	-10.843	31	44	25
SOMO 2	-10.964	26	57	18
HOMO-2	-11.159	12	66	22
HOMO-3	-11.373	18	42	39
HOMO-4	-11.400	5	87	8
HOMO-5	-11.447	55	33	12
	β-spin			
LUMO+5	-6.175	19	56	25
LUMO+4	-6.305	9	27	65
LUMO+3	-6.357	8	27	65
LUMO+2	-8.331	53	17	31
LUMO+1	-8.846	49	13	13
LUMO	-9.096	62	25	13
HOMO	-10.954	23	69	7
HOMO-1	-11.049	10	72	18
HOMO-2	-11.247	15	72	13
HOMO-3	-11.277	18	71	11
HOMO-4	-11.513	44	15	41
HOMO-5	-11.597	56	17	28

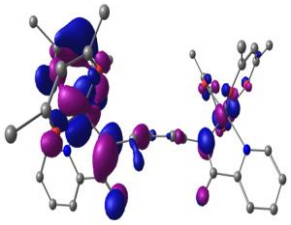
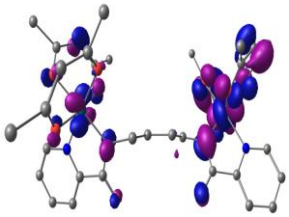
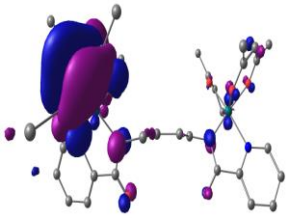
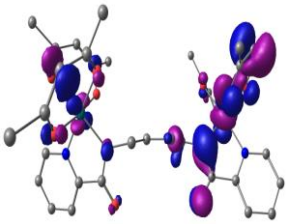
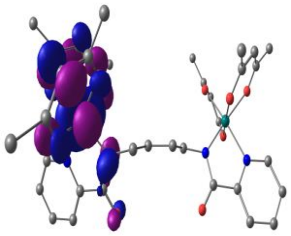
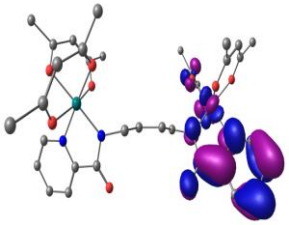
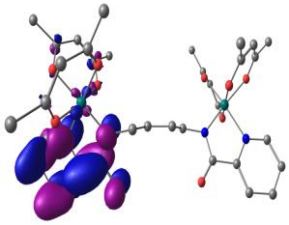
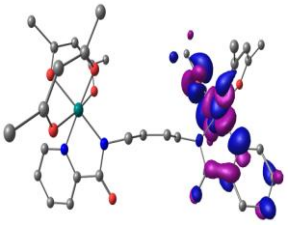
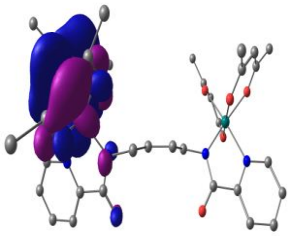
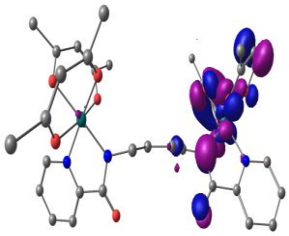
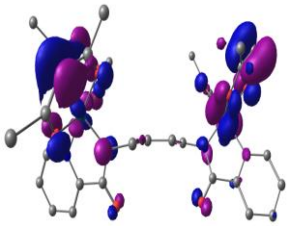
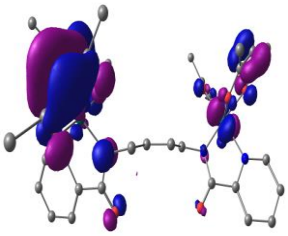
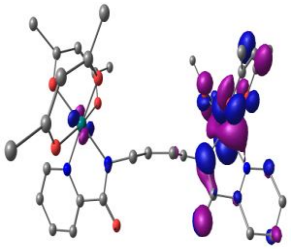
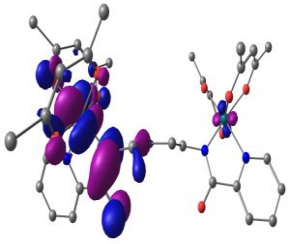
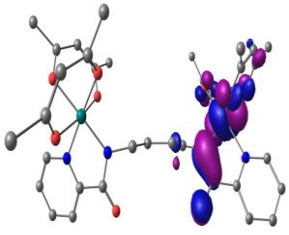
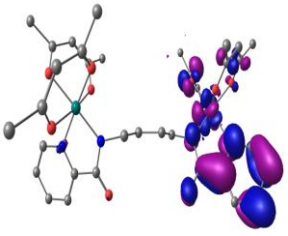
α-spin			
			
SOMO 1	SOMO 2	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S34 Composition and energies of selected molecular orbitals of **4a**$^{+}$ (*S*=1/2)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	-3.565	4	92	4
LUMO+4	-3.650	9	85	6
LUMO+3	-3.683	9	85	6
LUMO+2	-3.948	3	5	93
LUMO+1	-3.976	3	3	94
LUMO	-7.188	51	35	14
SOMO	-7.553	51	34	15
HOMO-1	-8.306	36	26	39
HOMO-2	-8.408	40	34	26
HOMO-3	-8.605	22	64	14
HOMO-4	-8.673	25	53	23
HOMO-5	-8.890	51	32	17
		β-spin		
LUMO+5	-3.650	8	84	9
LUMO+4	-3.681	8	83	9
LUMO+3	-3.869	3	12	85
LUMO+2	-3.893	3	9	87
LUMO+1	-5.858	58	13	29
LUMO	-5.921	59	13	28
HOMO	-8.287	42	48	10
HOMO-1	-8.340	43	49	8
HOMO-2	-8.535	44	40	16
HOMO-3	-8.590	45	43	12
HOMO-4	-8.811	41	26	33
HOMO-5	-8.851	40	26	34

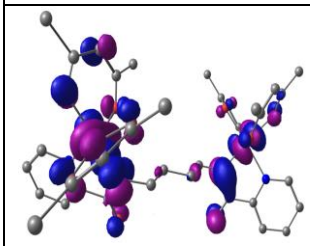
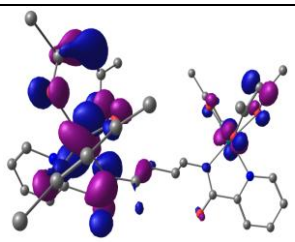
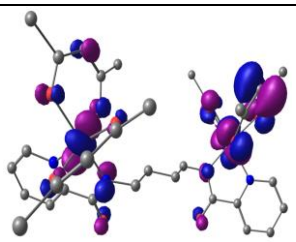
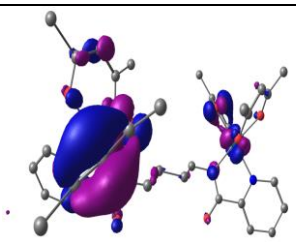
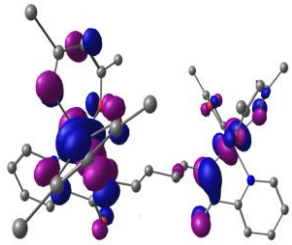
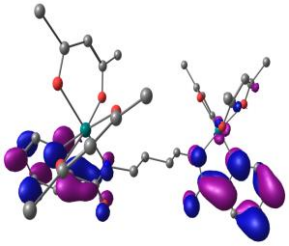
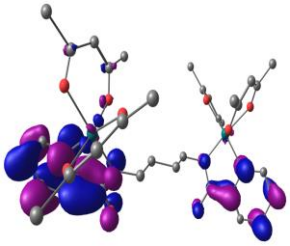
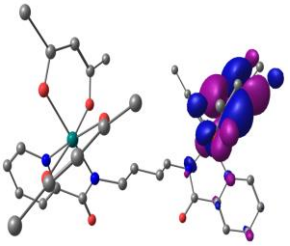
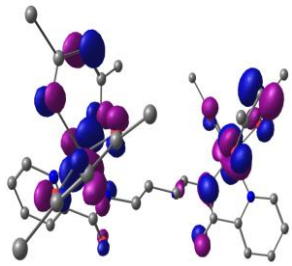
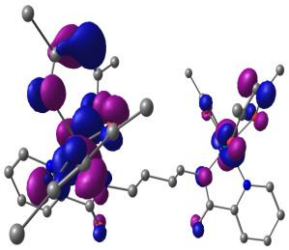
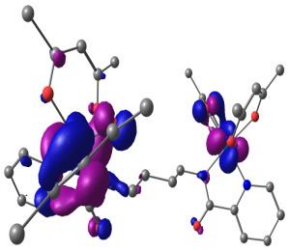
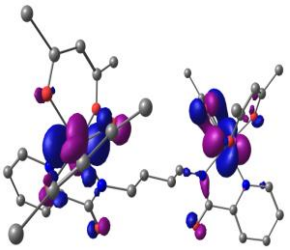
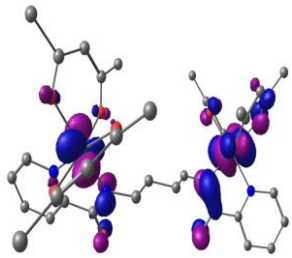
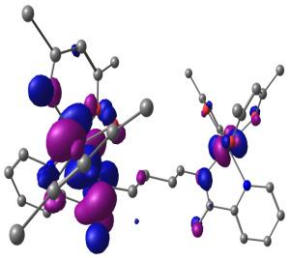
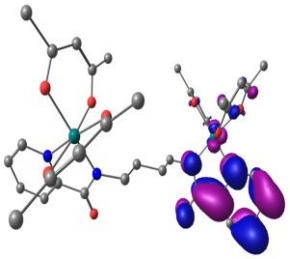
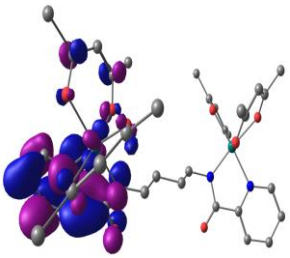
α-spin			
			
SOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S35 Composition and energies of selected molecular orbitals of **4a** (*S*=1)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	-0.859	5	91	4
LUMO+4	-0.876	4	93	3
LUMO+3	-1.072	6	11	83
LUMO+2	-1.250	5	3	92
LUMO+1	-1.324	4	84	12
LUMO	-1.537	3	2	96
SOMO 1	-4.817	28	7	65
SOMO 2	-5.067	37	13	50
HOMO-2	-5.212	54	40	6
HOMO-3	-5.215	54	39	6
HOMO-4	-5.472	62	25	13
HOMO-5	-5.594	18	4	78
		β-spin		
LUMO+5	-1.042	7	13	80
LUMO+4	-1.246	6	3	91
LUMO+3	-1.320	4	82	14
LUMO+2	-1.429	4	4	92
LUMO+1	-2.215	65	19	16
LUMO	-2.523	60	21	19
HOMO	-4.744	42	20	38
HOMO-1	-4.864	54	24	22
HOMO-2	-5.158	64	22	13
HOMO-3	-5.352	49	18	33
HOMO-4	-5.445	36	11	53
HOMO-5	-5.554	19	19	62

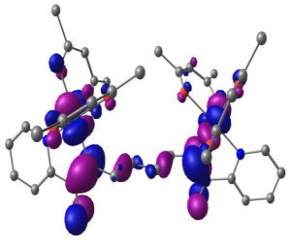
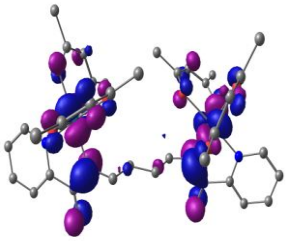
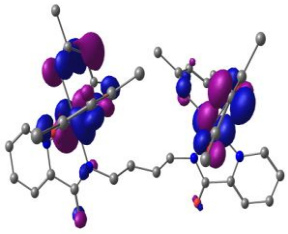
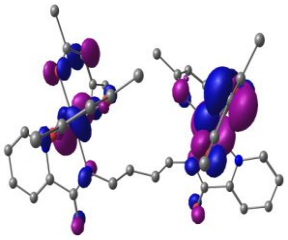
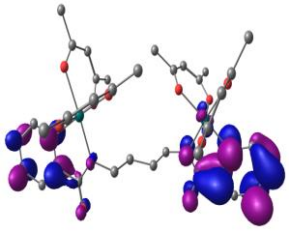
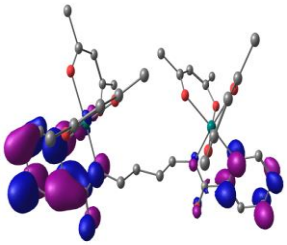
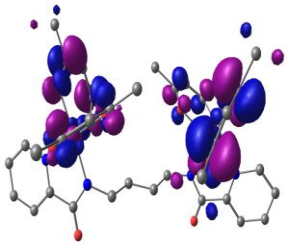
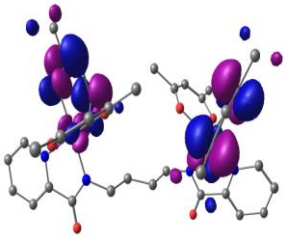
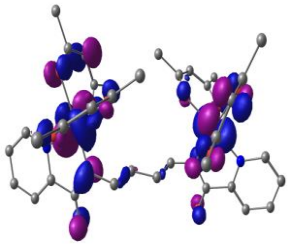
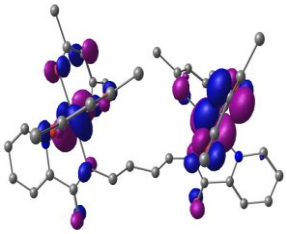
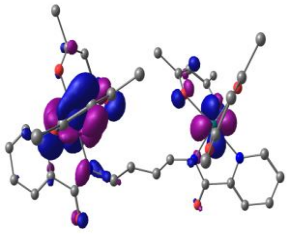
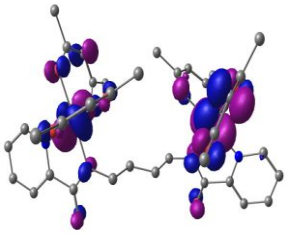
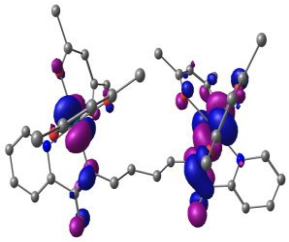
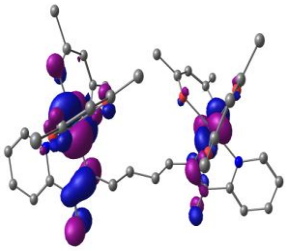
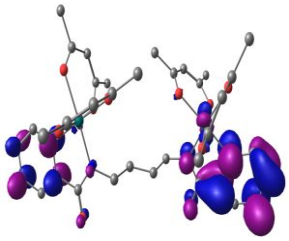
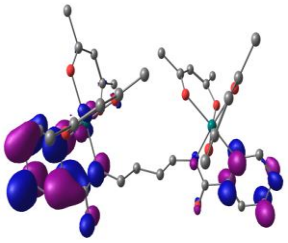
α-spin			
			
SOMO 1	SOMO 2	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S36 Composition and energies of selected molecular orbitals of **4a**$^{2-}$ (*S*=0)

MO	Energy (eV)	% Composition		
		Ru	acac	L
LUMO+5	4.260	6	86	7
LUMO+4	4.242	5	89	6
LUMO+3	4.140	3	4	93
LUMO+2	4.126	2	5	93
LUMO+1	3.701	7	7	86
LUMO	3.689	7	6	87
HOMO	1.348	60	12	28
HOMO-1	1.311	63	12	25
HOMO-2	0.735	70	18	12
HOMO-3	0.724	71	18	11
HOMO-4	0.559	76	16	9
HOMO-5	0.537	76	15	8

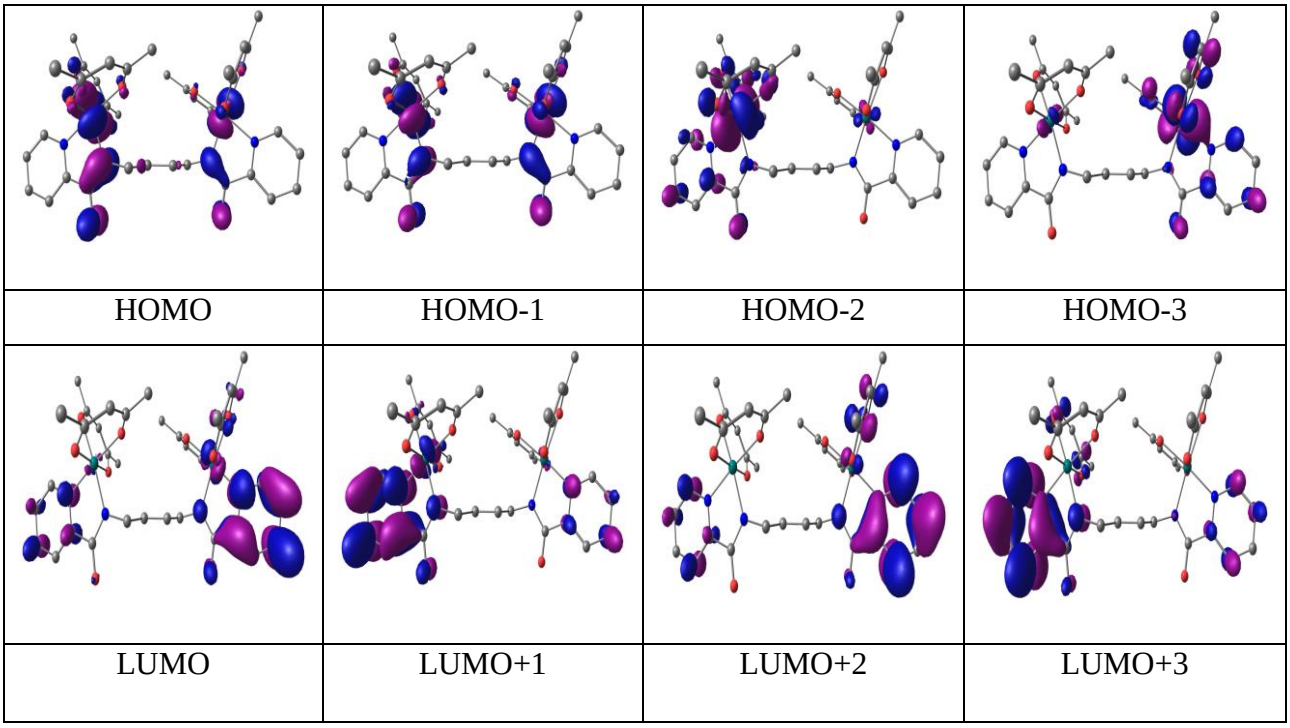


Table S37 Composition and energies of selected molecular orbitals of $\mathbf{5}^{2+}$ (*S*=1)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	-5.808	7	88	5
LUMO+4	-5.812	7	82	11
LUMO+3	-5.838	3	7	90
LUMO+2	-5.838	2	1	97
LUMO+1	-7.161	8	2	90
LUMO	-7.167	8	2	90
SOMO 1	-10.318	25	72	2
SOMO 2	-10.320	25	72	3
HOMO-2	-10.876	48	44	8
HOMO-3	-10.878	48	43	9
HOMO-4	-11.066	34	59	7
HOMO-5	-11.073	34	60	6
		β-spin		
LUMO+5	-5.801	4	1	95
LUMO+4	-5.804	4	5	91
LUMO+3	-7.146	9	3	88
LUMO+2	-7.152	9	3	88
LUMO+1	-7.951	61	32	7
LUMO	-7.951	62	32	6
HOMO	-10.498	23	71	5
HOMO-1	-10.500	23	71	6
HOMO-2	-10.642	66	25	10
HOMO-3	-10.643	66	24	10
HOMO-4	-10.782	55	39	7
HOMO-5	-10.799	55	38	7

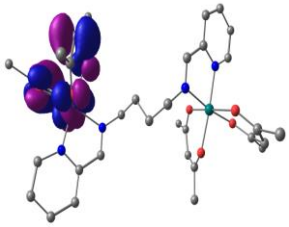
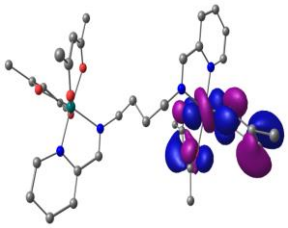
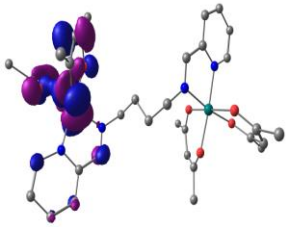
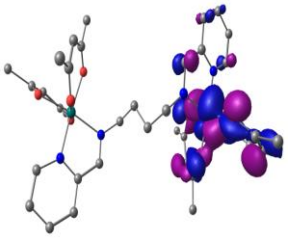
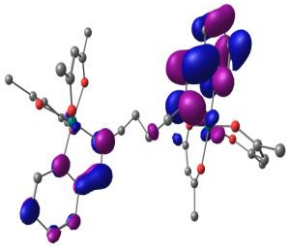
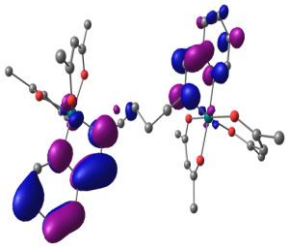
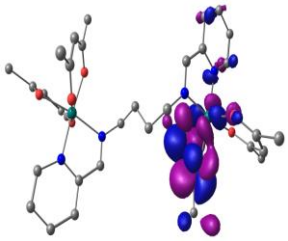
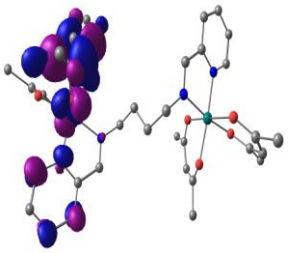
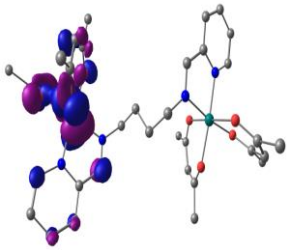
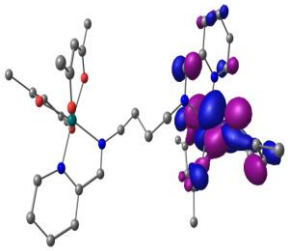
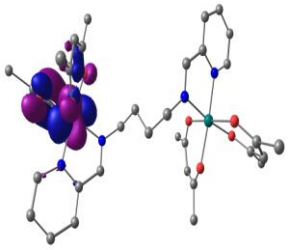
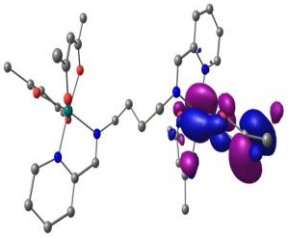
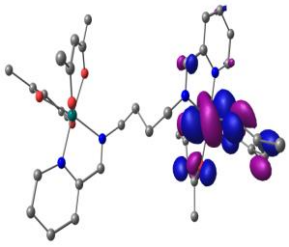
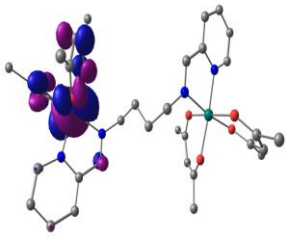
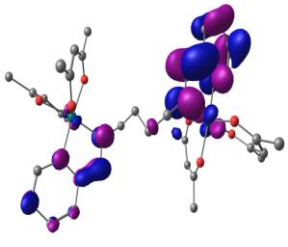
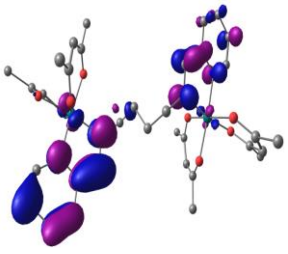
α-spin			
			
SOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S38 Composition and energies of selected molecular orbitals of **5** (*S*=0)

MO	Energy (eV)	% Composition		
		Ru	acac	L
LUMO+5	-0.422	4	92	4
LUMO+4	-0.462	4	92	4
LUMO+3	-0.722	4	1	95
LUMO+2	-0.754	4	2	94
LUMO+1	-1.714	12	4	83
LUMO	-1.751	13	4	83
HOMO	-3.831	63	32	5
HOMO-1	-3.866	63	32	5
HOMO-2	-4.480	68	17	15
HOMO-3	-4.514	68	17	15
HOMO-4	-4.619	78	15	7
HOMO-5	-4.650	78	16	6

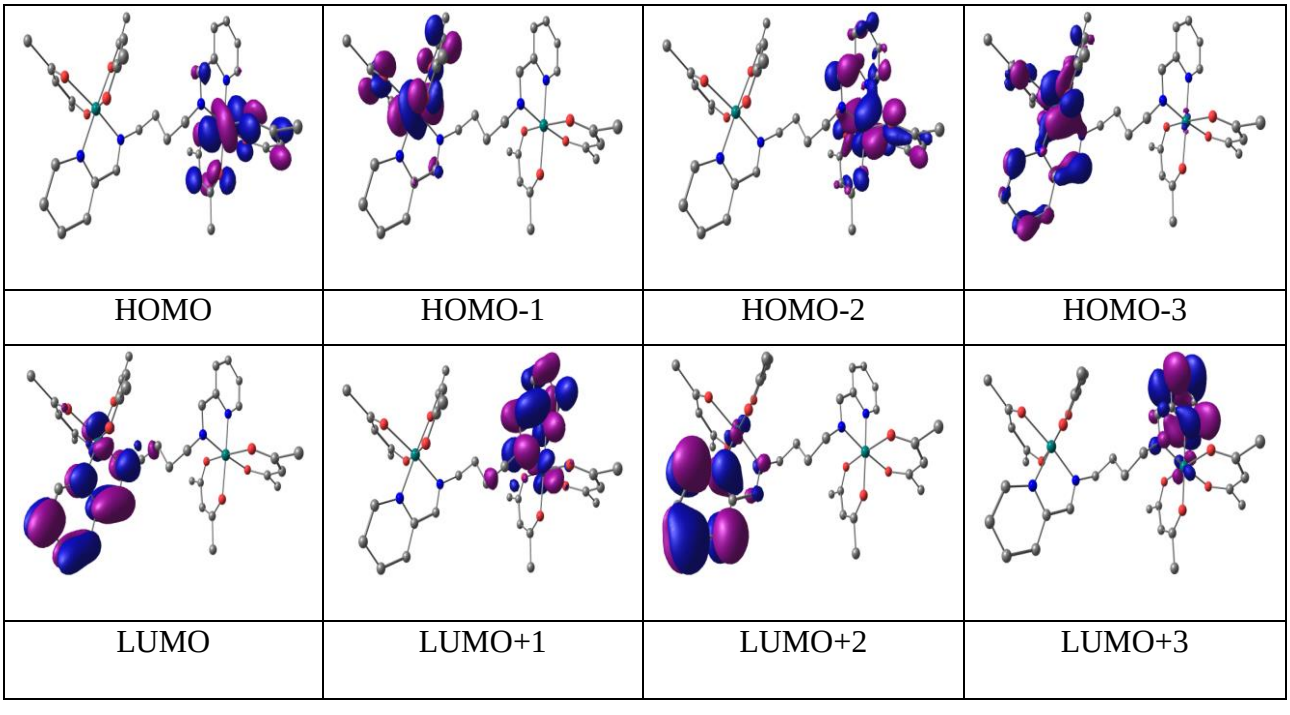


Table S39 Composition and energies of selected molecular orbitals of **5**$^{-}$ (*S*=1/2)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	2,057	6	73	21
LUMO+4	2.042	5	44	52
LUMO+3	1.998	5	44	51
LUMO+2	1.951	3	79	18
LUMO+1	1.918	3	75	23
LUMO	0.581	13	4	83
SOMO	0.204	13	4	82
HOMO-1	-1.561	72	22	6
HOMO-2	-1.605	71	22	6
HOMO-3	-2.022	75	16	9
HOMO-4	-2.037	70	16	14
HOMO-5	-2.062	77	15	8
		β-spin		
LUMO+5	2.088	5	72	22
LUMO+4	2.044	5	59	35
LUMO+3	1.975	4	85	12
LUMO+2	1.943	3	83	14
LUMO+1	1.339	10	9	81
LUMO	1.270	10	8	82
HOMO	-1.538	71	21	7
HOMO-1	-1.584	71	22	7
HOMO-2	-1.876	70	16	14
HOMO-3	-1.927	70	16	14
HOMO-4	-2.006	78	15	8
HOMO-5	-2.044	78	15	7

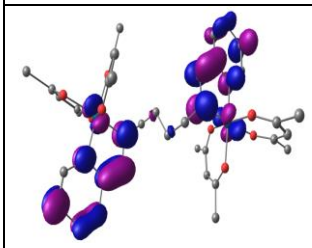
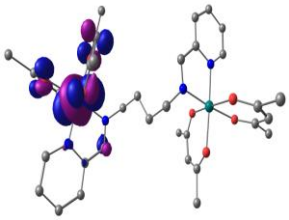
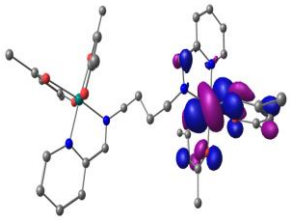
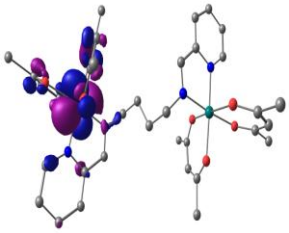
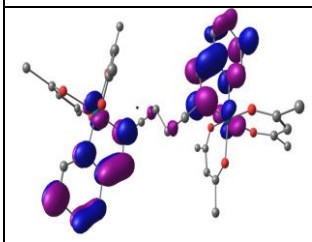
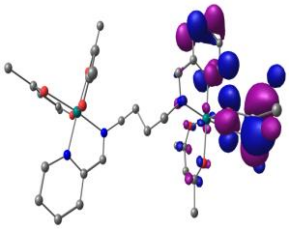
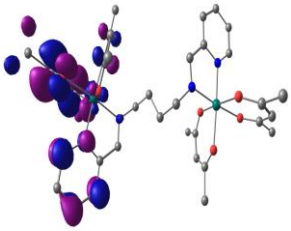
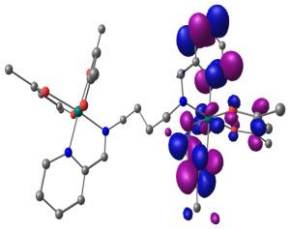
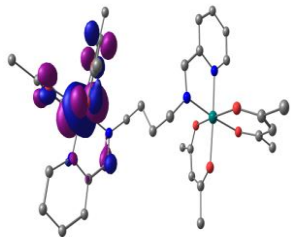
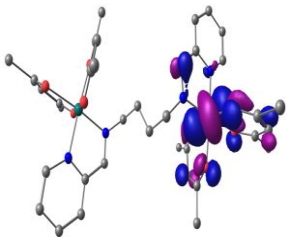
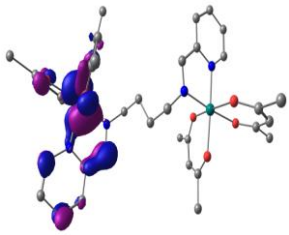
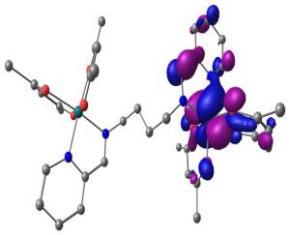
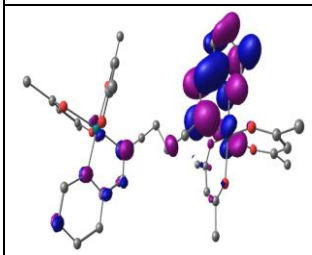
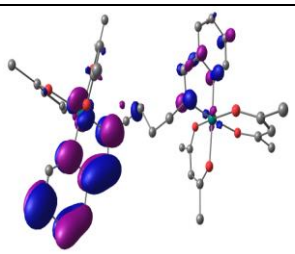
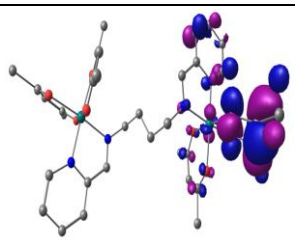
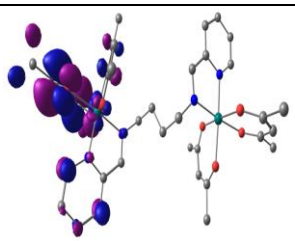
α-spin			
			
SOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S40 Composition and energies of selected molecular orbitals of $\mathbf{5}^{2-}$ (*S*=1)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	4.855	10	2	88
LUMO+4	4.823	9	2	89
LUMO+3	4.431	5	91	3
LUMO+2	4.411	6	91	3
LUMO+1	4.236	4	93	3
LUMO	4.221	4	93	3
SOMO 1	2.469	12	4	84
SOMO 2	2.397	13	4	83
HOMO-2	1.020	76	18	7
HOMO-3	1.009	75	18	7
HOMO-4	0.620	79	15	7
HOMO-5	0.596	79	15	7
		β-spin		
LUMO+5	4.502	8	75	17
LUMO+4	4.499	10	67	24
LUMO+3	4.341	8	66	26
LUMO+2	4.318	8	66	25
LUMO+1	4.153	2	67	32
LUMO	4.122	3	58	40
HOMO	1.086	74	16	9
HOMO-1	1.070	74	17	9
HOMO-2	0.820	74	15	11
HOMO-3	0.802	74	15	11
HOMO-4	0.661	78	15	7
HOMO-5	0.637	78	15	7

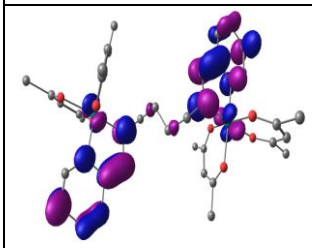
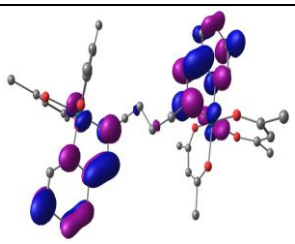
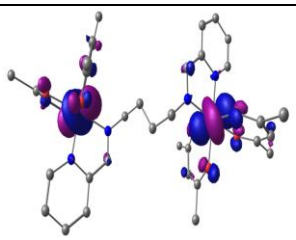
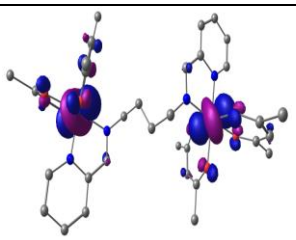
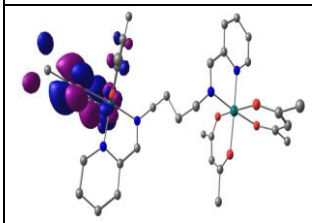
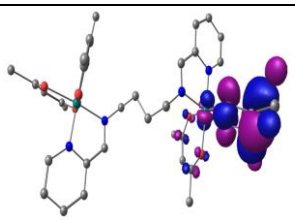
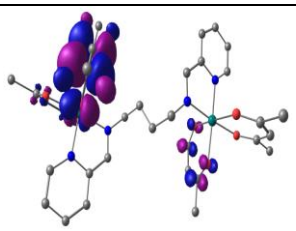
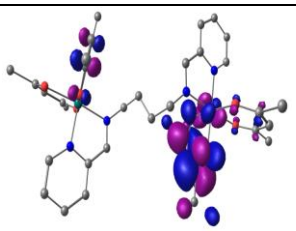
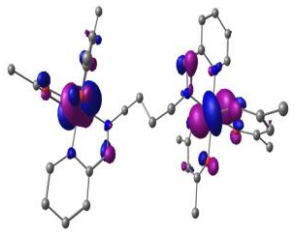
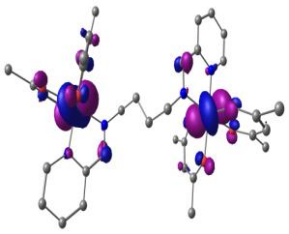
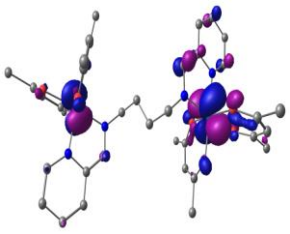
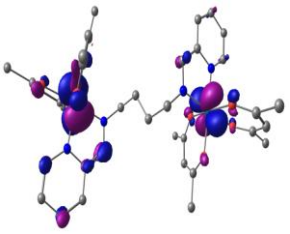
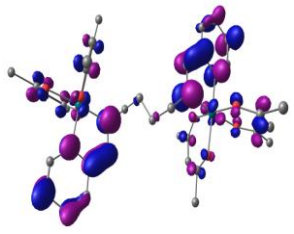
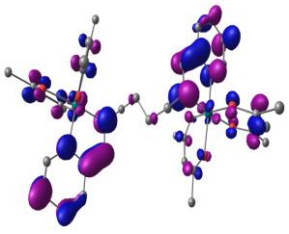
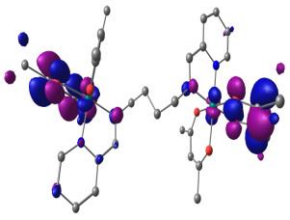
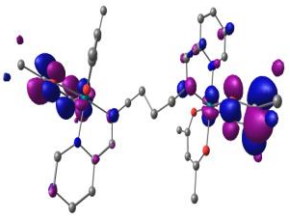
α-spin			
			
SOMO 1	SOMO 2	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S41 Composition and energies of selected molecular orbitals of **5a**$^{4+}$ (*S*=1)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	-11.203	14	43	44
LUMO+4	-11.223	14	39	47
LUMO+3	-11.323	32	56	12
LUMO+2	-11.336	33	55	12
LUMO+1	-14.394	45	41	14
LUMO	-14.442	45	43	11
SOMO 1	-16.125	18	42	40
SOMO 2	-16.272	26	46	28
HOMO-2	-16.469	21	71	7
HOMO-3	-16.482	22	71	7
HOMO-4	-16.740	3	2	95
HOMO-5	-16.767	3	2	95
		β-spin		
LUMO+5	-11.277	21	70	9
LUMO+4	-11.289	21	70	9
LUMO+3	-13.984	38	11	51
LUMO+2	-14.178	44	13	43
LUMO+1	-14.320	45	51	4
LUMO	-14.332	45	50	4
HOMO	-16.256	25	68	7
HOMO-1	-16.274	25	68	6
HOMO-2	-16.420	21	50	30
HOMO-3	-16.534	16	54	30
HOMO-4	-16.668	9	5	86
HOMO-5	-16.700	4	2	94

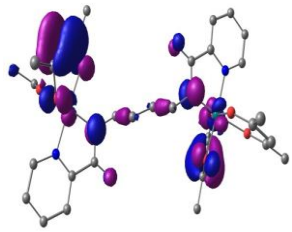
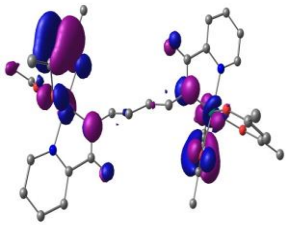
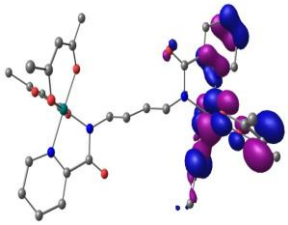
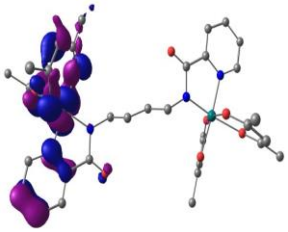
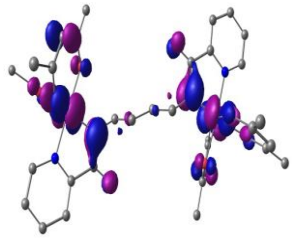
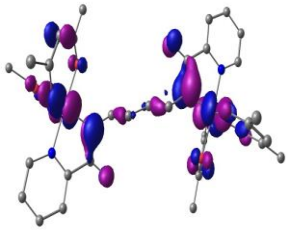
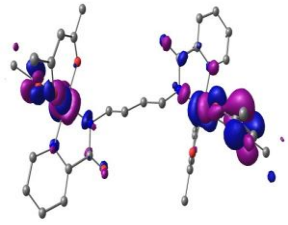
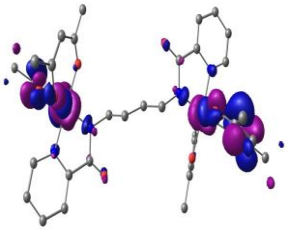
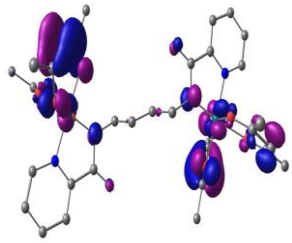
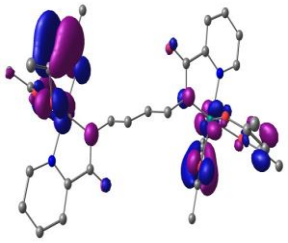
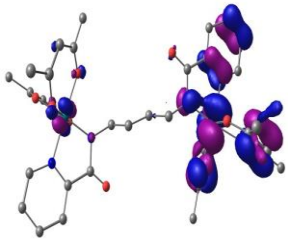
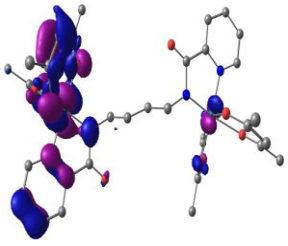
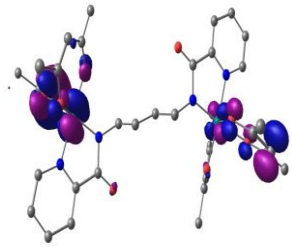
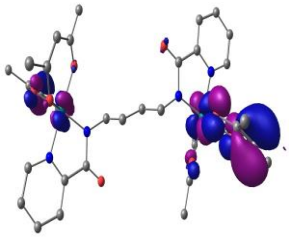
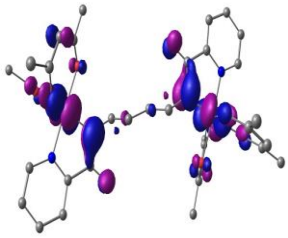
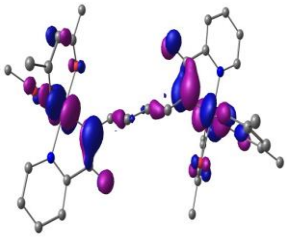
α-spin			
			
SOMO 1	SOMO 2	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S42 Composition and energies of selected molecular orbitals of **5a**$^{3+}$ (*S*=3/2)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	-8.769	21	59	19
LUMO+4	-8.785	9	84	7
LUMO+3	-8.922	40	49	11
LUMO+2	-9.092	6	20	74
LUMO+1	-9.311	24	56	20
LUMO	-11.983	49	24	27
SOMO 1	-13.344	33	46	21
SOMO 2	-13.619	25	42	33
SOMO 3	-13.978	2	82	15
HOMO-3	-14.050	9	69	22
HOMO-4	-14.185	45	15	39
HOMO-5	-14.337	26	67	7
		β-spin		
LUMO+5	-9.029	8	61	67
LUMO+4	-9.243	22	25	26
LUMO+3	-10.693	56	52	23
LUMO+2	-11.179	56	29	15
LUMO+1	-11.852	47	24	29
LUMO	-12.822	31	54	14
HOMO	-13.159	16	72	12
HOMO-1	-13.484	37	46	17
HOMO-2	-13.884	56	17	27
HOMO-3	-14.027	6	67	27
HOMO-4	-14.088	13	48	38
HOMO-5	-14.288	5	7	88

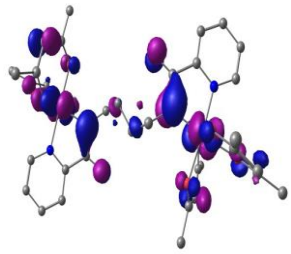
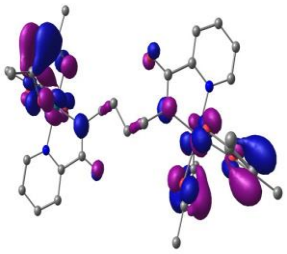
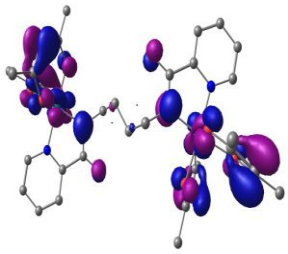
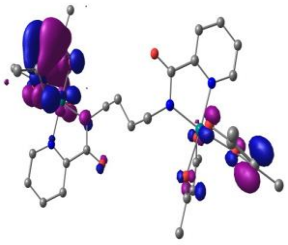
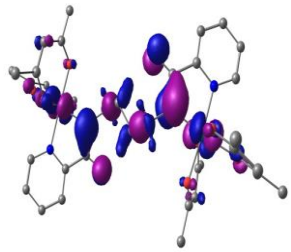
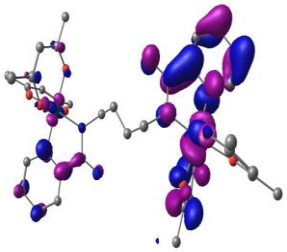
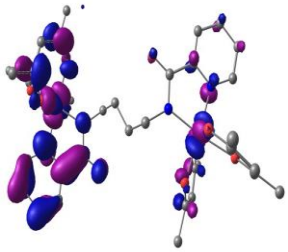
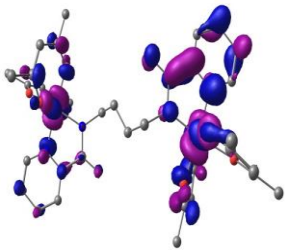
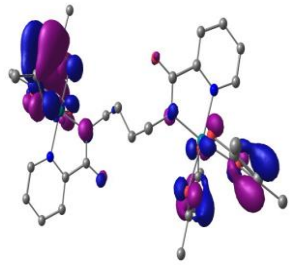
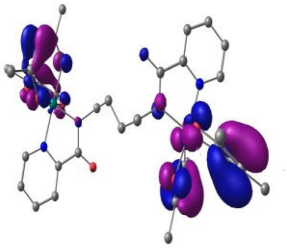
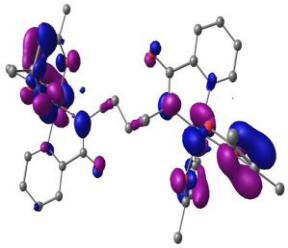
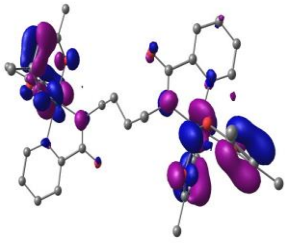
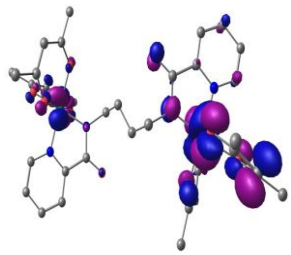
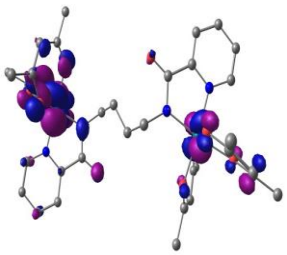
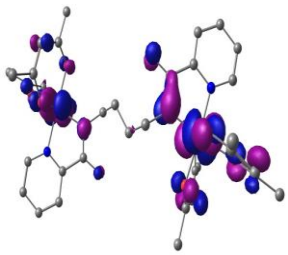
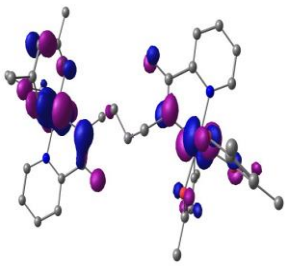
α-spin			
			
SOMO	SOMO 2	SOMO 3	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S43 Composition and energies of selected molecular orbitals of **5a**$^{2+}$ (*S*=1)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	-6.084	15	78	7
LUMO+4	-6.278	20	66	14
LUMO+3	-6.308	4	7	89
LUMO+2	-6.532	48	44	8
LUMO+1	-6.660	3	3	95
LUMO	-8.889	52	24	24
SOMO 1	-10.751	34	51	15
SOMO 2	-11.076	25	61	13
HOMO-2	-11.142	3	84	13
HOMO-3	-11.420	60	23	17
HOMO-4	-11.473	20	44	35
HOMO-5	-11.529	7	67	26
		β-spin		
LUMO+5	-6.225	25	70	5
LUMO+4	-6.308	4	8	89
LUMO+3	-6.493	4	9	87
LUMO+2	-8.646	57	21	22
LUMO+1	-8.888	52	24	23
LUMO	-9.217	58	26	16
HOMO	-10.751	34	51	15
HOMO-1	-11.138	3	82	14
HOMO-2	-11.154	4	82	13
HOMO-3	-11.353	31	54	15
HOMO-4	-11.423	62	23	15
HOMO-5	-11.698	46	27	27

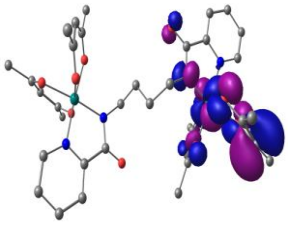
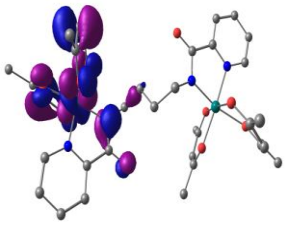
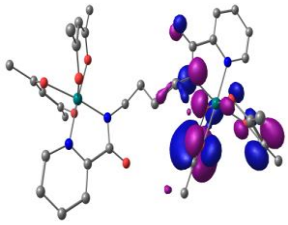
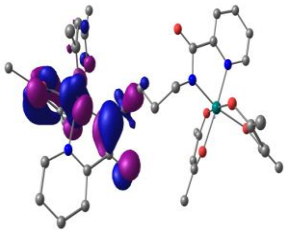
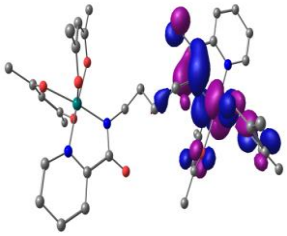
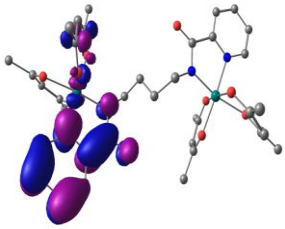
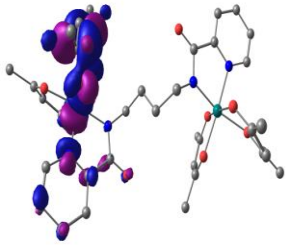
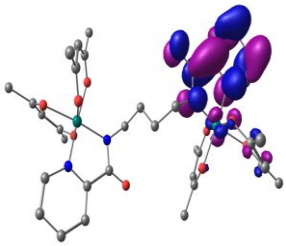
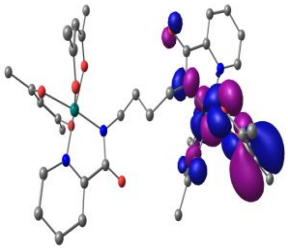
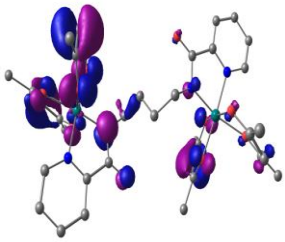
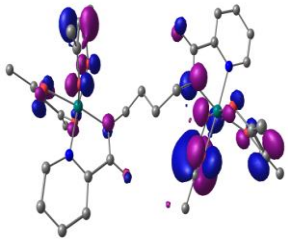
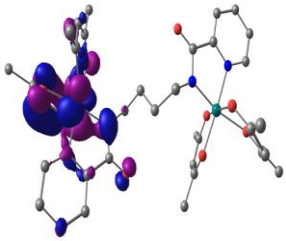
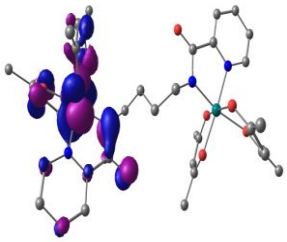
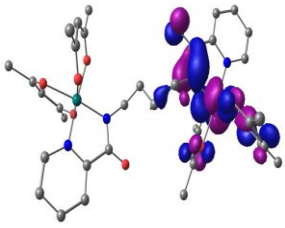
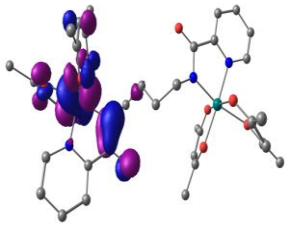
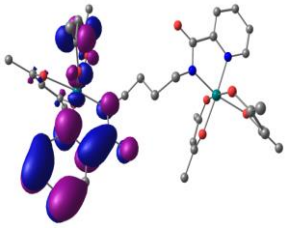
α-spin			
			
SOMO 1	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S44 Composition and energies of selected molecular orbitals of **5a**$^{+}$ (*S*=1/2)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	-3.369	3	76	21
LUMO+4	-3.463	15	80	6
LUMO+3	-3.524	12	83	6
LUMO+2	-3.921	3	3	94
LUMO+1	-4.006	3	2	94
LUMO	-5.884	66	22	13
SOMO	-8.056	35	39	27
HOMO-1	-8.231	42	40	19
HOMO-2	-8.350	32	45	23
HOMO-3	-8.558	13	75	12
HOMO-4	-8.592	11	73	16
HOMO-5	-8.719	57	31	12
		β-spin		
LUMO+5	-3.383	8	86	6
LUMO+4	-3.562	16	77	7
LUMO+3	-3.827	4	5	91
LUMO+2	-3.959	4	3	93
LUMO+1	-5.411	64	17	19
LUMO	-7.000	48	20	32
HOMO	-7.399	52	24	24
HOMO-1	-8.307	30	64	6
HOMO-2	-8.321	34	52	14
HOMO-3	-8.492	41	48	12
HOMO-4	-8.594	7	76	17
HOMO-5	-8.766	39	36	25

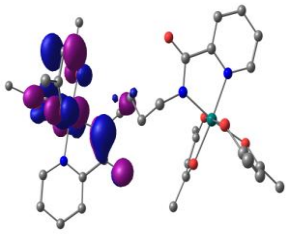
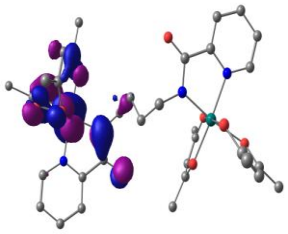
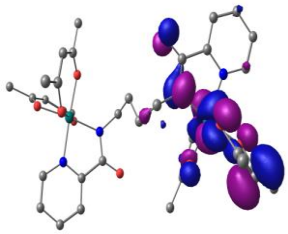
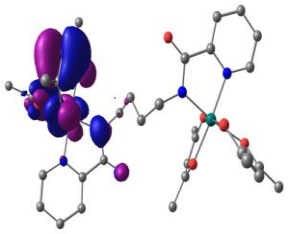
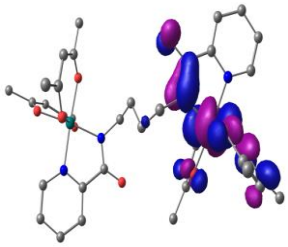
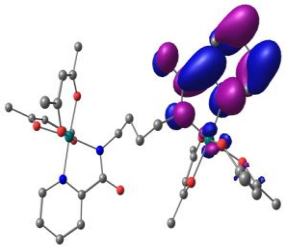
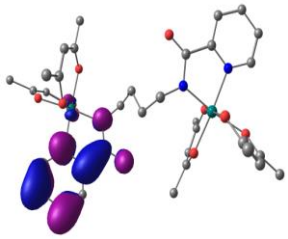
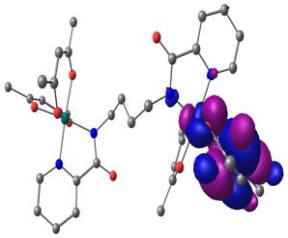
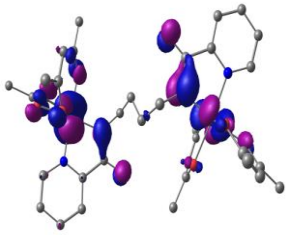
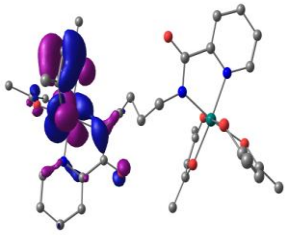
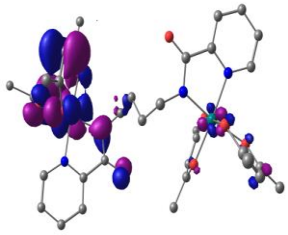
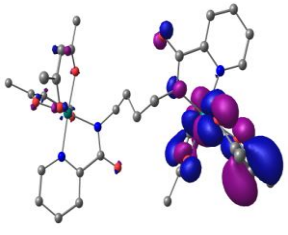
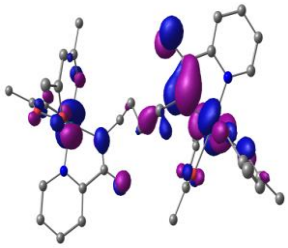
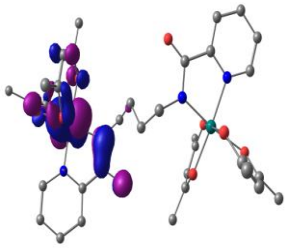
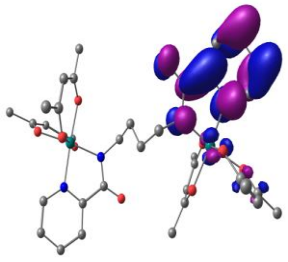
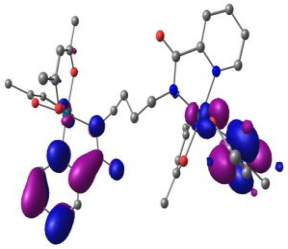
α-spin			
			
SOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S45 Composition and energies of selected molecular orbitals of **5a** (*S*=1)

MO	Energy (eV)	% Composition		
		Ru	acac	L
		α-spin		
LUMO+5	-0.704	4	93	3
LUMO+4	-0.841	3	92	5
LUMO+3	-0.865	4	93	3
LUMO+2	-1.012	5	89	6
LUMO+1	-1.265	16	8	76
LUMO	-1.437	4	4	92
SOMO 1	-5.143	22	6	72
SOMO 2	-5.275	33	15	52
HOMO-2	-5.321	41	47	12
HOMO-3	-5.390	49	43	8
HOMO-4	-5.562	30	15	55
HOMO-5	-5.782	41	24	35
		β-spin		
LUMO+5	-0.809	5	80	15
LUMO+4	-0.991	5	88	7
LUMO+3	-1.026	15	22	63
LUMO+2	-1.379	4	6	90
LUMO+1	-2.618	64	17	19
LUMO	-2.795	62	13	25
HOMO	-4.943	40	18	42
HOMO-1	-5.072	51	25	24
HOMO-2	-5.279	34	19	47
HOMO-3	-5.544	60	18	23
HOMO-4	-5.575	50	29	21
HOMO-5	-5.898	23	46	31

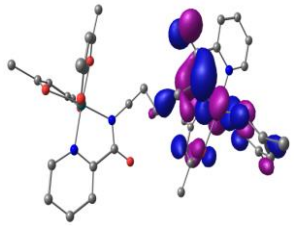
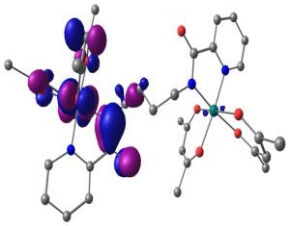
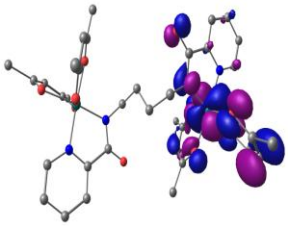
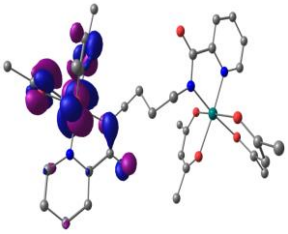
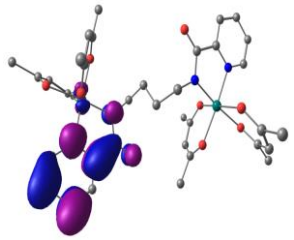
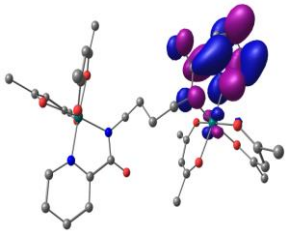
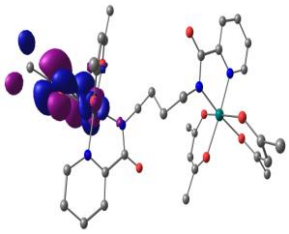
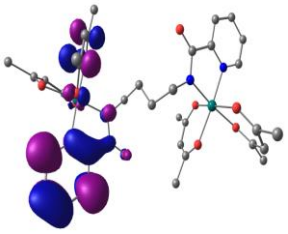
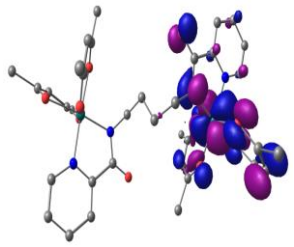
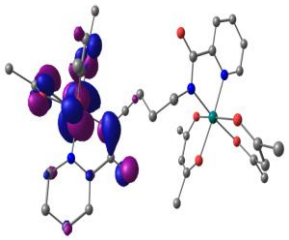
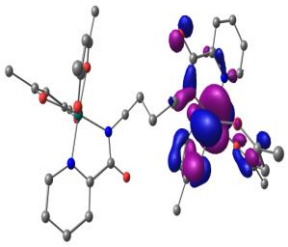
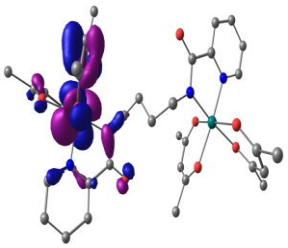
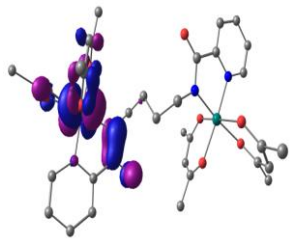
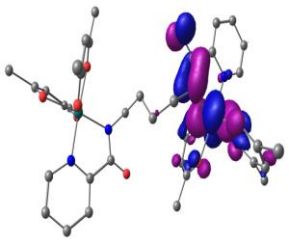
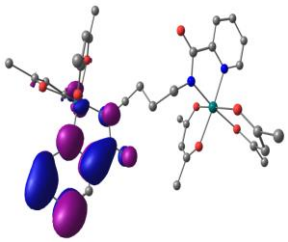
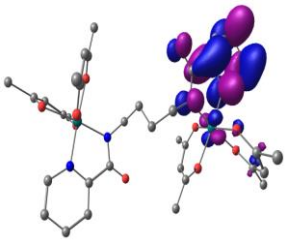
α-spin			
			
SOMO 1	SOMO 2	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3
β-spin			
			
HOMO	HOMO-1	HOMO-2	HOMO-3
			
LUMO	LUMO+1	LUMO+2	LUMO+3

Table S46 Composition and energies of selected molecular orbitals of **5a**$^{2-}$ (*S*=0)

MO	Energy (eV)	% Composition		
		Ru	acac	L
LUMO+5	4.330	4	10	85
LUMO+4	4.302	6	75	19
LUMO+3	4.199	4	4	92
LUMO+2	4.151	7	82	11
LUMO+1	3.893	6	14	80
LUMO	3.761	6	15	80
HOMO	1.781	61	14	25
HOMO-1	1.575	60	14	25
HOMO-2	1.007	70	18	12
HOMO-3	0.835	70	18	12
HOMO-4	0.780	77	14	9
HOMO-5	0.605	77	15	9

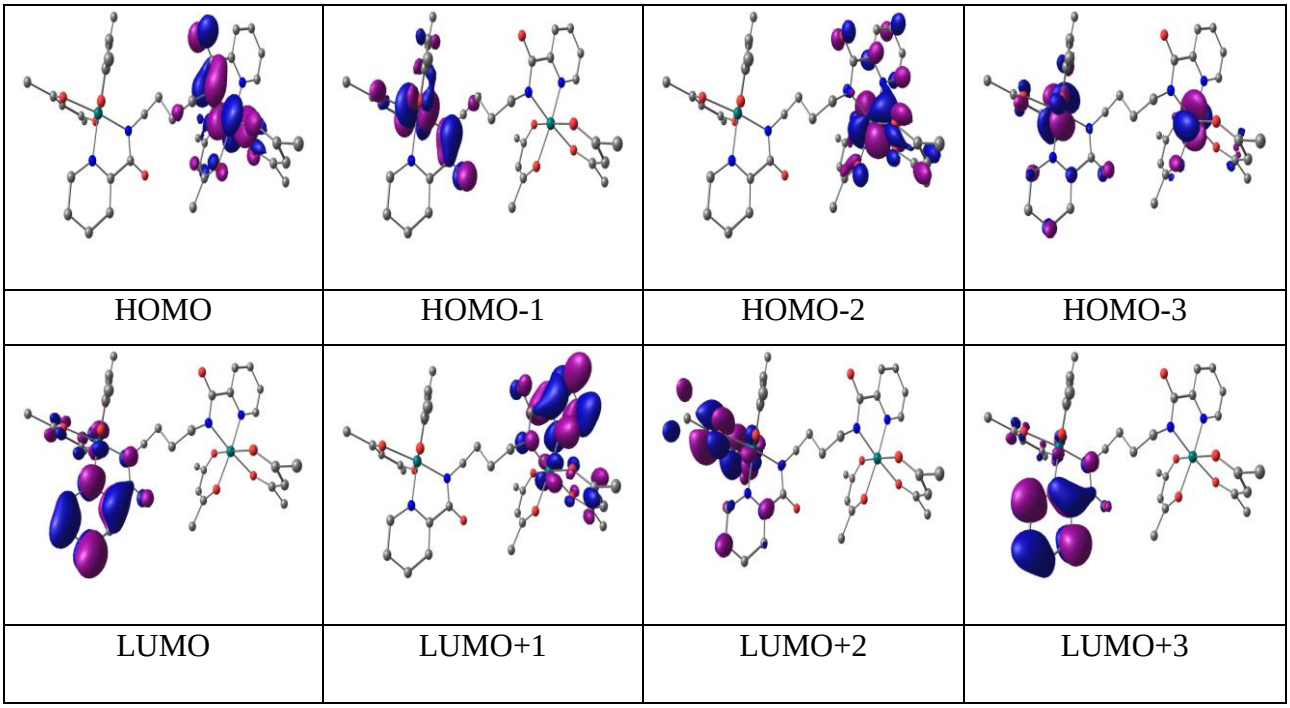


Table S47 DFT (UB3LYP/LANL2DZ/6-31G**) calculated Mulliken spin density distributions

Complex	Ru1	Ru2	L	acac
$\mathbf{2}^{2+}$(*S*=1)	0.769	0.769	-0.004	0.493
$\mathbf{2}^{2-}$(*S*=1)	0.071	0.071	1.844	0.016
$\mathbf{2a}^{4+}$(*S*=1)	0.415	0.400	0.533	0.651
$\mathbf{2a}^{3+}$(*S*=3/2)	1.137	1.134	-0.681	0.920
$\mathbf{2a}^{2+}$(*S*=1)	0.007	1.218	0.351	0.426
$\mathbf{2a}^{+}$(*S*=1/2)	-0.634	0.938	0.605	0.221
2a(*S*=1)	0.750	0.750	0.411	0.135
$\mathbf{3}^{2+}$(*S*=1)	0.771	0.774	-0.021	0.302
$\mathbf{3}^{2-}$(*S*=1)	0.078	0.081	1.924	0.016
$\mathbf{3a}^{4+}$(*S*=1)	0.275	0.343	0.834	0.473
$\mathbf{3a}^{3+}$(*S*=3/2)	1.058	1.034	1.059	-0.097
$\mathbf{3a}^{2+}$(*S*=1)	0	1.213	0.226	0.416
$\mathbf{3a}^{+}$(*S*=1/2)	0.711	0.401	-0.327	0.216
3a(*S*=1)	0.744	0.756	0.379	0.141
$\mathbf{4}^{2+}$(*S*=1)	0.764	0.774	-0.016	0.482
$\mathbf{4}^{2-}$(*S*=1)	0.079	0.092	1.813	0.015
$\mathbf{4a}^{4+}$(*S*=1)	0.304	0.321	0.251	0.302
$\mathbf{4a}^{3+}$(*S*=3/2)	0.238	1.180	0.418	1.154
$\mathbf{4a}^{2+}$(*S*=1)	-0.122	1.203	0.754	0.234
$\mathbf{4a}^{+}$(*S*=1/2)	0.477	0.403	0.136	0.025
4a(*S*=1)	0.755	0.753	0.356	0.143
$\mathbf{5}^{2+}$(*S*=1)	0.765	0.766	-0.020	0.493
$\mathbf{5}^{2-}$(*S*=1)	0.076	0.077	1.826	0.018
$\mathbf{5a}^{4+}$(*S*=1)	0.281	0.281	0.073	0.136
$\mathbf{5a}^{3+}$(*S*=3/2)	1.082	1.081	0.968	-0.148
$\mathbf{5a}^{2+}$(*S*=1)	1.217	0	0.419	0.380
$\mathbf{5a}^{+}$(*S*=1/2)	0.923	-0.398	0.350	0.137
5a(*S*=1)	0.766	0.760	0.304	0.177

Table S48 Simulation data for EPR

Parameter	**2^{2+}**		**3^{2+}**		**4^{2+}**		**5^{2+}**	
	Ru	**Ru**	**Ru**	**Ru**	**Ru**	**Ru**	**Ru**	**Ru**
Sys. weight	50%	50%	25%	75%	58%	42%	31%	50%
S	1		1		1		1	
g_x	2.3062	2.2551	2.3875	2.2986	2.3121	2.4194	2.2572	2.3183
g_y	2.2561	2.0846	2.1522	2.2974	2.3120	2.2899	2.2572	2.2956
g_z	1.9154	1.7925	2.1519	1.8990	2.1078	1.8906	2.1581	1.9024
lwpp (peak to peak)	21.8126 11.1672	4.6423 7.1842	15.5150 11.8349	14.2103 6.9250	10.2630 12.2964	4.6593 0.0700	16.7859 14.8432	17.4424 2.2250
A	48.4898	111.8380	38.4919	35.8921	28.2692	63.2253	63.4902	84.1727
Parameter	**2a**		**3a**		**4a**		**5a**	
	Ru	**Ru**	**Ru**	**Ru**	**Ru**	**Ru**	**Ru**	**Ru**
Sys. weight	20%	80%	70%	30%	30%	70%	50%	50%
S	1		1		1		1	
g_x	2.4121	2.2400	2.4935	2.4488	2.3493	2.2678	2.3844	2.3104
g_y	2.2311	2.0365	2.2466	2.2681	2.3335	2.0907	2.2298	2.2231
g_z	2.0574	1.7960	1.8540	2.2680	2.1357	1.8821	2.0603	1.8098
lwpp (peak to peak)	0.0137 6.6630	6.9747 4.4994	0.0880 11.5717	0.0205 4.6167	4.5832 10.8569	9.5237 5.7337	0.2665 9.5865	2.0679 2.4984
A	75.6892	0.0949	67.1555	9.2405	21.2639	65.0723	43.6927	61.6109

EPR simulation:

Spectral simulations were performed using the EasySpin package (verson number 5.2.35)[1] based on Matlab R2023a (The MathWorks, Massachusetts,USA). The EPR data was fit using a spin Hamiltonian, H, containing the electron Zeeman interaction with the applied magnetic field B_o, and the hyperfine coupling (hfc) interactions with two ^{99}Ru/^{101}Ru (natural abundance 12.74 and 17.05 % respectively).

$$H = \beta_e \boldsymbol{S}.\boldsymbol{g}.\boldsymbol{B_0} + h\,\boldsymbol{S}.\boldsymbol{A}^{Ru}{}_i.\boldsymbol{I}^{Ru}{}_i$$

EasySpin calculates EPR resonance fields using the energies of the states of the spin system obtained by direct diagonalization of the spin Hamiltonian (Eq above). The EPR fitting procedure used a Monte Carlo type iteration to minimize the root-mean-square deviation between measured and simulated spectra. The optimum values of the following parameters were obtained from the simulation: the principal components of g (g_x, g_y, g_z), the principal components of the hfc tensors $\boldsymbol{A}^{Ru}$ (A_x^{Ru}, A_y^{Ru}, A_z^{Ru}), and the peak-to peak line widths (ΔB_x, ΔB_y and ΔB_z)

A. *Optimised cartesian coordinates using M06(SMD)/TZVP/SDD//B3LYP/6-31G**/LANL2DZ level of theory for the free energy calculations*

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2 (*S*=0)

Zero-point correction= 0.714681 (Hartree/Particle)
Thermal correction to Energy= 0.765213
Thermal correction to Enthalpy= 0.766157
Thermal correction to Gibbs Free Energy= 0.626954
Sum of electronic and zero-point Energies= -2330.280640
Sum of electronic and thermal Energies= -2330.230108
Sum of electronic and thermal Enthalpies= -2330.229164
Sum of electronic and thermal Free Energies= -2330.368368

Cartesian Coordinates

Ru	-1.209350000	3.758345000	14.375988000
O	-0.707345000	2.166261000	15.631081000
O	-0.883080000	2.609036000	12.682139000
O	-1.592720000	4.859999000	16.083002000
O	-3.151220000	2.981284000	14.360130000
N	-1.573988000	5.431214000	13.263335000
N	0.616356000	4.577487000	14.279236000
C	-0.497709000	6.253565000	13.122709000
C	0.709053000	5.731728000	13.704366000
H	1.653613000	6.267266000	13.627441000
C	1.794239000	3.961849000	14.862865000
H	2.702900000	4.489138000	14.555502000
H	1.860442000	2.938610000	14.483486000
C	-0.531432000	1.393569000	12.723155000
C	-2.892929000	6.997506000	12.050066000
H	-3.859683000	7.257295000	11.638938000
C	-1.800260000	7.844434000	11.909021000
H	-1.896543000	8.784761000	11.380986000
C	-0.356931000	1.028480000	15.209465000
C	-0.589120000	7.464195000	12.448707000
H	0.291411000	8.088994000	12.360322000
C	-2.742028000	5.806686000	12.729635000
H	-3.567450000	5.115845000	12.863247000
C	-4.032733000	3.296965000	15.207445000
C	-2.718822000	4.878045000	16.663753000
C	-3.871108000	4.176837000	16.288865000
H	-4.741400000	4.324752000	16.916557000
C	-0.271126000	0.627866000	13.867564000
H	0.031081000	-0.398225000	13.694763000

C	-0.381232000	0.742651000	11.379342000
H	-1.329279000	0.801115000	10.839550000
H	-0.075448000	-0.300308000	11.447707000
H	0.356141000	1.292352000	10.789612000
C	0.024467000	0.046275000	16.277393000
H	0.955507000	0.379298000	16.751455000
H	0.175339000	-0.961470000	15.891525000
H	-0.741978000	0.028446000	17.054451000
C	-5.376688000	2.664234000	14.995155000
H	-5.796922000	3.023813000	14.051832000
H	-6.079436000	2.884131000	15.797542000
H	-5.262252000	1.582325000	14.901526000
C	-2.776360000	5.756812000	17.878401000
H	-2.011560000	5.441119000	18.591872000
H	-3.749183000	5.736621000	18.367221000
H	-2.542839000	6.785696000	17.594100000
N	5.063991000	5.430874000	17.995626000
N	2.873649000	4.577136000	16.979698000
C	3.987651000	6.253115000	18.136440000
C	2.780914000	5.731304000	17.554709000
H	1.836339000	6.266809000	17.631685000
C	1.695798000	3.961601000	16.395888000
H	0.787095000	4.488717000	16.703419000
H	1.629673000	2.938240000	16.774941000
C	6.382811000	6.996975000	19.209274000
H	7.349543000	7.256730000	19.620475000
C	5.290081000	7.843791000	19.350510000
H	5.386289000	8.783995000	19.878778000
C	4.078974000	7.463595000	18.810722000
H	3.198397000	8.088310000	18.899248000
C	6.232001000	5.806307000	18.529419000
H	7.057474000	5.115559000	18.395644000
Ru	4.699427000	3.758154000	16.882728000
O	4.197472000	2.166156000	15.627487000
O	4.373602000	2.608549000	18.576464000
O	5.082444000	4.860124000	15.175844000
O	6.641413000	2.981359000	16.898269000
C	4.022094000	1.393049000	18.535337000
C	3.847242000	1.028280000	16.049003000
C	7.522810000	3.297387000	16.050961000
C	6.208515000	4.878508000	14.595037000
C	7.360960000	4.177467000	14.969744000
H	8.231207000	4.325714000	14.342068000
C	3.761616000	0.627498000	17.390862000
H	3.459495000	-0.398633000	17.563576000
C	3.872130000	0.741935000	19.879082000
H	4.820202000	0.800560000	20.418811000
H	3.566577000	-0.301084000	19.810612000
H	3.134669000	1.291392000	20.468931000
C	3.465829000	0.046166000	14.980990000

H	2.535088000	0.379525000	14.506588000
H	3.314473000	-0.961512000	15.366842000
H	4.232546000	0.028005000	14.204204000
C	8.866931000	2.664951000	16.263091000
H	9.287152000	3.024537000	17.206419000
H	9.569569000	2.885099000	15.460676000
H	8.752756000	1.583007000	16.356621000
C	6.265830000	5.757549000	13.380580000
H	5.501150000	5.441773000	12.667016000
H	7.238674000	5.737748000	12.891786000
H	6.031989000	6.786305000	13.665084000

2a (*S*=1)

Zero-point correction= 0.704482 (Hartree/Particle)
Thermal correction to Energy= 0.755487
Thermal correction to Enthalpy= 0.756431
Thermal correction to Gibbs Free Energy= 0.618457
Sum of electronic and zero-point Energies= -2479.582031
Sum of electronic and thermal Energies= -2479.531026
Sum of electronic and thermal Enthalpies= -2479.530081
Sum of electronic and thermal Free Energies= -2479.668056

Cartesian Coordinates

Ru	7.417388000	6.352415000	9.978916000
Ru	13.353591000	6.788317000	9.439259000
O	12.431198000	7.988614000	8.067975000
O	8.371746000	5.288670000	11.438323000
O	6.418504000	7.416874000	8.566104000
O	14.319547000	5.646482000	10.813936000
N	8.523702000	5.422804000	8.584797000
O	14.578179000	8.390375000	9.945528000
O	8.725817000	7.894967000	10.371058000
O	6.209079000	7.193913000	11.446209000
O	12.033045000	7.531695000	10.834021000
N	14.618060000	5.949334000	8.054005000
O	12.278537000	3.362723000	7.482650000
N	6.158384000	4.802585000	9.484816000
N	12.243567000	5.225094000	8.841321000
O	8.491492000	3.674654000	7.082006000
C	10.864094000	5.009728000	9.224811000
H	10.714309000	5.365277000	10.248332000
H	10.634933000	3.938764000	9.201383000
C	4.954317000	4.575160000	10.009830000
H	4.656879000	5.227898000	10.822769000
C	7.975324000	4.363258000	7.960327000
C	9.680668000	10.018962000	10.256891000
H	9.408233000	10.369949000	11.256508000

H	9.748143000	10.880660000	9.592949000
H	10.655381000	9.529021000	10.339974000
C	9.904873000	5.742043000	8.288673000
H	10.059158000	6.820287000	8.386058000
H	10.135131000	5.461053000	7.255235000
C	6.717699000	8.620887000	8.263828000
C	6.602059000	4.046569000	8.467648000
C	8.666408000	9.018598000	9.798518000
C	6.511980000	7.242961000	12.677857000
C	14.284103000	9.605819000	9.729241000
C	8.388156000	5.606117000	12.667840000
C	12.089604000	7.256331000	12.064773000
C	14.171219000	4.792936000	7.538127000
C	5.838517000	3.020815000	7.947925000
H	6.240083000	2.436287000	7.130303000
C	14.019093000	5.649146000	12.055173000
C	15.191882000	10.604678000	10.373598000
H	15.252003000	10.404678000	11.445462000
H	14.864801000	11.630769000	10.214348000
H	16.200392000	10.487730000	9.969406000
C	7.746790000	9.389421000	8.801954000
H	7.846796000	10.394496000	8.411144000
C	12.794223000	4.374266000	7.955026000
C	12.410124000	9.257373000	8.127648000
C	7.564227000	6.550778000	13.284607000
H	7.695559000	6.689922000	14.350098000
C	13.231475000	10.056441000	8.926575000
H	13.102515000	11.127878000	8.840602000
C	4.141571000	3.562942000	9.537611000
H	3.171209000	3.402455000	9.988389000
C	14.936972000	4.056278000	6.657091000
H	14.532611000	3.131331000	6.265786000
C	5.838932000	9.216441000	7.209755000
H	5.907244000	8.615678000	6.300028000
H	6.101372000	10.247110000	6.978074000
H	4.798959000	9.176068000	7.541298000
C	15.826857000	6.404974000	7.724516000
H	16.126298000	7.339160000	8.185783000
C	13.002240000	6.375432000	12.670623000
H	12.901421000	6.244946000	13.740933000
C	16.641500000	5.717677000	6.846215000
H	17.615591000	6.116592000	6.596261000
C	16.190917000	4.524222000	6.306377000
H	16.812634000	3.964663000	5.618453000
C	11.072566000	7.951262000	12.913021000
H	11.314293000	9.017025000	12.957796000
H	11.035861000	7.563473000	13.930514000
H	10.092860000	7.863344000	12.432510000
C	9.376813000	4.841060000	13.489319000
H	9.143699000	3.774954000	13.440813000

H	9.389575000	5.156592000	14.531500000
H	10.376177000	4.968920000	13.060279000
C	5.618455000	8.098737000	13.518287000
H	5.620612000	9.118340000	13.126148000
H	5.917495000	8.115267000	14.564846000
H	4.592335000	7.731018000	13.445516000
C	4.588708000	2.776231000	8.488925000
H	3.967808000	1.979302000	8.098694000
C	14.879737000	4.753496000	12.888904000
H	14.796252000	3.728073000	12.521727000
H	14.612177000	4.778189000	13.943605000
H	15.925284000	5.048814000	12.775886000
C	11.422004000	9.902966000	7.208808000
H	11.675373000	9.665254000	6.173064000
H	11.386405000	10.984926000	7.327967000
H	10.427660000	9.485301000	7.398601000

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3 (*S*=0)

Zero-point correction= 0.746042 (Hartree/Particle)
Thermal correction to Energy= 0.797135
Thermal correction to Enthalpy= 0.798080
Thermal correction to Gibbs Free Energy= 0.659921
Sum of electronic and zero-point Energies= -2369.550703
Sum of electronic and thermal Energies= -2369.499609
Sum of electronic and thermal Enthalpies= -2369.498665
Sum of electronic and thermal Free Energies= -2369.636824

Cartesian Coordinates

Ru	9.970007000	15.754552000	8.704409000
Ru	8.483099000	9.950258000	10.695954000
O	8.975295000	8.588105000	12.175290000
O	8.862815000	17.506791000	8.944320000
O	8.322002000	14.849402000	9.597399000
O	10.800007000	16.190802000	10.551650000
O	9.038861000	15.325442000	6.908566000
O	6.484479000	9.564609000	11.172661000
O	8.388432000	11.504829000	12.088137000
O	7.927231000	11.284027000	9.223220000
N	11.551656000	16.634475000	7.769211000
N	8.695735000	8.489559000	9.289084000
N	10.410655000	10.194691000	10.165534000
N	11.207751000	14.168916000	8.416821000
C	12.324462000	14.429288000	7.820246000
C	10.854600000	9.399185000	9.247377000
C	9.933791000	8.426388000	8.728439000
C	11.671116000	17.932723000	7.464503000

H	10.835396000	18.562427000	7.750304000
C	8.064006000	14.930292000	10.829221000
C	12.569903000	15.794160000	7.448244000
C	13.720201000	16.247265000	6.815132000
H	14.508031000	15.541993000	6.578987000
C	9.121809000	8.894192000	13.395493000
C	12.786817000	18.440589000	6.833000000
H	12.834192000	19.497378000	6.604783000
C	5.507341000	10.112710000	10.585073000
C	10.957657000	16.398619000	12.869276000
H	11.779679000	15.688719000	13.002568000
H	10.348035000	16.392855000	13.772374000
H	11.404228000	17.386149000	12.737430000
C	13.831747000	17.585790000	6.501557000
H	14.717714000	17.964173000	6.007408000
C	10.246197000	7.486874000	7.754581000
H	11.245017000	7.470909000	7.335102000
C	8.016504000	6.660534000	7.925937000
H	7.229405000	5.976902000	7.635756000
C	6.729236000	11.580061000	8.945876000
C	9.277885000	6.593681000	7.345724000
H	9.497448000	5.851287000	6.588926000
C	7.764954000	7.615955000	8.888567000
H	6.797692000	7.702388000	9.371709000
C	7.979048000	17.885597000	8.123859000
C	8.661088000	11.365192000	13.313025000
C	10.942029000	12.788905000	8.792355000
H	11.718675000	12.144963000	8.356777000
H	9.980186000	12.490770000	8.361873000
C	10.180271000	16.014530000	11.644017000
C	4.154391000	9.672963000	11.063922000
H	4.040417000	8.600262000	10.885832000
H	3.339101000	10.200274000	10.570420000
H	4.079738000	9.826100000	12.142886000
C	9.002659000	10.168346000	13.961400000
H	9.185516000	10.237260000	15.026889000
C	11.301172000	11.204999000	10.720735000
H	12.331159000	10.995641000	10.405584000
H	11.258341000	11.120244000	11.810630000
C	8.088013000	16.015917000	6.439851000
C	5.576951000	11.053409000	9.548740000
H	4.630863000	11.421845000	9.170459000
C	10.907885000	12.613074000	10.304556000
H	9.918094000	12.852772000	10.698357000
H	11.621691000	13.304663000	10.765610000
C	9.463317000	7.739622000	14.291188000
H	8.669451000	6.990966000	14.236969000
H	9.598334000	8.037428000	15.329986000
H	10.378499000	7.260813000	13.935129000
C	7.580535000	17.215108000	6.958913000

H	6.781216000	17.676803000	6.391856000
C	7.451771000	15.447551000	5.205416000
H	8.223314000	15.114734000	4.508815000
H	6.791188000	16.155735000	4.706960000
H	6.866259000	14.565880000	5.484494000
C	8.898149000	15.486088000	11.812993000
H	8.521142000	15.456645000	12.828282000
C	7.325469000	19.195370000	8.454111000
H	6.958055000	19.174809000	9.482280000
H	6.503705000	19.438261000	7.781890000
H	8.072427000	19.992112000	8.397022000
C	6.579812000	12.609622000	7.866630000
H	7.026171000	13.548125000	8.219093000
H	5.540721000	12.778860000	7.584420000
H	7.148001000	12.297706000	6.985837000
C	8.627742000	12.628674000	14.121090000
H	9.388440000	13.316282000	13.735493000
H	8.804200000	12.457919000	15.182115000
H	7.663222000	13.125577000	13.989705000
C	6.767185000	14.315544000	11.260426000
H	6.020593000	14.417747000	10.471014000
H	6.389846000	14.751839000	12.186184000
H	6.940379000	13.245725000	11.430835000
H	11.881081000	9.445365000	8.887798000
H	13.053431000	13.652618000	7.596635000

3a (*S*=1)

Zero-point correction= 0.731738 (Hartree/Particle)
Thermal correction to Energy= 0.784764
Thermal correction to Enthalpy= 0.785708
Thermal correction to Gibbs Free Energy= 0.641729
Sum of electronic and zero-point Energies= -2518.846976
Sum of electronic and thermal Energies= -2518.793950
Sum of electronic and thermal Enthalpies= -2518.793006
Sum of electronic and thermal Free Energies= -2518.936985

Cartesian Coordinates

Ru	10.498997000	15.309408000	10.674690000
Ru	8.278622000	9.388549000	10.824140000
O	8.455622000	7.671272000	11.890376000
O	9.405951000	17.031047000	11.083956000
O	8.746504000	14.320313000	11.113328000
O	11.086561000	15.294044000	12.621038000
O	9.864462000	15.412088000	8.731971000
O	6.210936000	9.194930000	10.972417000
O	8.163702000	10.534692000	12.542475000

O	8.008932000	11.051083000	9.676282000
O	13.820942000	13.041892000	9.978851000
O	12.000584000	8.789871000	9.235379000
N	12.303020000	16.221218000	10.291024000
N	8.505897000	8.272540000	9.118677000
N	10.269213000	9.521966000	10.577714000
N	11.629147000	13.714480000	10.222897000
C	12.954496000	13.906782000	10.115306000
C	10.809658000	8.862314000	9.536561000
C	9.777588000	8.165753000	8.702330000
C	12.538505000	17.533869000	10.304207000
H	11.677250000	18.175170000	10.451334000
C	8.288615000	14.224351000	12.287091000
C	13.320396000	15.358306000	10.131683000
C	14.618457000	15.799966000	9.970736000
H	15.404971000	15.066606000	9.847178000
C	8.341729000	7.614425000	13.161784000
C	13.812639000	18.040543000	10.140221000
H	13.967379000	19.111201000	10.149972000
C	5.366544000	10.079763000	10.639122000
C	10.905949000	15.120523000	14.936953000
H	11.719834000	14.397326000	15.035349000
H	10.185248000	14.948299000	15.734608000
H	11.345285000	16.113718000	15.049546000
C	14.869113000	17.160330000	9.973483000
H	15.878285000	17.533005000	9.848308000
C	10.103664000	7.442005000	7.572696000
H	11.143215000	7.382835000	7.277301000
C	7.785673000	6.936998000	7.301142000
H	6.968638000	6.464626000	6.772274000
C	6.911332000	11.662277000	9.488321000
C	9.093571000	6.820061000	6.860543000
H	9.322590000	6.247054000	5.970461000
C	7.526765000	7.672738000	8.440950000
H	6.524599000	7.794905000	8.836614000
C	8.414962000	17.415193000	10.392217000
C	8.125230000	10.050135000	13.707761000
C	11.132847000	12.354935000	10.136591000
H	11.829452000	11.763037000	9.532347000
H	10.165301000	12.359964000	9.623266000
C	10.297096000	14.987350000	13.577180000
C	3.945889000	9.766766000	10.989083000
H	3.653795000	8.828859000	10.511017000
H	3.256872000	10.552532000	10.683993000
H	3.863670000	9.614795000	12.067654000
C	8.180180000	8.685715000	14.035713000
H	8.126700000	8.441572000	15.088949000
C	11.152802000	10.211760000	11.502609000
H	12.185497000	9.968116000	11.232262000
H	10.972804000	9.817682000	12.510644000

C	8.829219000	16.044300000	8.352891000
C	5.657189000	11.265166000	9.958201000
H	4.821059000	11.907835000	9.713725000
C	10.984625000	11.720833000	11.516617000
H	10.018076000	11.996824000	11.951749000
H	11.758232000	12.132258000	12.176300000
C	8.417931000	6.227351000	13.715642000
H	7.646169000	5.609635000	13.251038000
H	8.296730000	6.200880000	14.796942000
H	9.381668000	5.785404000	13.452563000
C	8.082795000	16.944470000	9.117842000
H	7.222894000	17.394253000	8.638217000
C	8.437527000	15.799950000	6.929851000
H	9.197159000	16.224264000	6.268478000
H	7.473184000	16.241406000	6.683143000
H	8.408315000	14.725410000	6.738205000
C	8.981948000	14.543831000	13.466313000
H	8.451324000	14.374213000	14.395142000
C	7.576443000	18.481367000	11.022000000
H	7.152082000	18.094320000	11.952492000
H	6.768519000	18.819380000	10.375322000
H	8.206302000	19.332767000	11.288560000
C	7.033836000	12.902490000	8.664532000
H	7.725592000	13.582152000	9.171325000
H	6.077734000	13.402177000	8.509877000
H	7.469013000	12.648173000	7.694694000
C	8.026737000	11.060780000	14.806909000
H	8.830253000	11.794266000	14.689323000
H	8.081337000	10.613928000	15.798004000
H	7.083756000	11.606045000	14.712710000
C	6.900522000	13.679357000	12.388047000
H	6.275802000	14.116778000	11.605731000
H	6.452133000	13.869138000	13.362780000
H	6.933146000	12.596403000	12.220819000

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4 (*S*=0)

Zero-point correction= 0.773963 (Hartree/Particle)
Thermal correction to Energy= 0.826625
Thermal correction to Enthalpy= 0.827569
Thermal correction to Gibbs Free Energy= 0.685393
Sum of electronic and zero-point Energies= -2408.816139
Sum of electronic and thermal Energies= -2408.763477
Sum of electronic and thermal Enthalpies= -2408.762533
Sum of electronic and thermal Free Energies= -2408.904709

Cartesian Coordinates

Ru	6.379257000	22.516573000	6.337672000
Ru	2.083227000	23.164908000	1.099736000
O	6.266178000	22.685992000	4.271838000
O	7.540583000	20.782252000	6.156567000
O	0.888790000	24.466395000	0.020740000
O	4.713537000	21.283195000	6.225695000
O	6.512040000	22.273167000	8.383223000
O	3.221121000	21.770975000	2.110029000
O	0.398681000	22.734681000	2.262033000
O	1.510519000	21.545012000	-0.092274000
N	5.362262000	24.216800000	6.615341000
N	7.902282000	23.869449000	6.509544000
N	3.747716000	23.672873000	0.042780000
N	2.703246000	24.797213000	2.098294000
C	6.786284000	21.869526000	3.456384000
C	7.865522000	20.256426000	5.054019000
C	4.114490000	20.830658000	7.242825000
C	0.722526000	21.650007000	-1.073709000
C	6.061962000	25.258013000	6.935057000
H	5.601539000	26.223294000	7.139654000
C	5.655841000	21.632902000	9.062782000
C	7.490050000	25.103314000	6.914066000
C	7.539240000	20.731373000	3.776715000
H	7.908760000	20.151860000	2.939351000
C	9.212262000	23.646863000	6.358712000
H	9.487207000	22.649278000	6.033956000
C	3.934630000	24.367588000	6.373198000
H	3.520321000	25.147831000	7.023260000
H	3.440031000	23.421969000	6.611927000
C	4.515566000	20.980087000	8.577829000
H	3.884878000	20.507119000	9.320804000
C	2.733106000	20.933802000	2.925149000
C	0.188813000	24.109658000	-0.972011000
C	8.696201000	19.011318000	5.164668000
H	9.641208000	19.250129000	5.659448000
H	8.909362000	18.557844000	4.197616000
H	8.180545000	18.284366000	5.796176000
C	4.376473000	24.796118000	0.483053000
C	4.288384000	22.988374000	-0.972127000
H	3.752802000	22.094993000	-1.275104000
C	9.747319000	25.872475000	7.044497000
H	10.473621000	26.646970000	7.256572000
C	1.975892000	25.386984000	3.211630000
H	0.913042000	25.331180000	2.963126000
H	2.252915000	26.443713000	3.313749000
C	0.093071000	22.822791000	-1.516797000
H	-0.554479000	22.719293000	-2.378895000
C	3.728027000	25.414256000	1.606540000
H	4.084052000	26.360551000	2.009642000
C	2.856133000	20.070754000	6.947370000

H	2.217487000	20.679195000	6.299103000
H	2.307180000	19.794790000	7.846550000
H	3.098675000	19.162783000	6.388429000
C	10.159187000	24.617683000	6.610833000
H	11.207941000	24.387834000	6.475136000
C	8.398294000	26.117175000	7.193077000
H	8.030482000	27.082875000	7.518861000
C	0.371897000	21.795498000	3.107846000
C	5.933579000	21.591088000	10.536429000
H	6.888878000	21.089507000	10.709716000
H	5.156968000	21.072212000	11.096372000
H	6.033516000	22.608931000	10.919980000
C	5.546580000	25.255016000	-0.108006000
H	6.013777000	26.156230000	0.270895000
C	1.418881000	20.915214000	3.415668000
H	1.189652000	20.129460000	4.126096000
C	5.454169000	23.389129000	-1.590899000
H	5.853164000	22.801333000	-2.407217000
C	-0.611623000	25.213178000	-1.599295000
H	-1.321188000	25.606876000	-0.867313000
H	-1.160586000	24.886808000	-2.481342000
H	0.052295000	26.035856000	-1.874181000
C	0.468536000	20.374404000	-1.822517000
H	1.409852000	20.009792000	-2.242304000
H	-0.254174000	20.492805000	-2.628619000
H	0.111052000	19.609350000	-1.129692000
C	6.093793000	24.544649000	-1.156731000
H	7.008297000	24.879255000	-1.630038000
C	2.247904000	24.661854000	4.517896000
H	1.627941000	25.133851000	5.288950000
H	1.921220000	23.616971000	4.441109000
C	6.536710000	22.186024000	2.012909000
H	5.458283000	22.125241000	1.827535000
H	7.056119000	21.508583000	1.335453000
H	6.844832000	23.213526000	1.798979000
C	3.696432000	19.898850000	3.420447000
H	4.387497000	19.621330000	2.622728000
H	3.187901000	19.009437000	3.794055000
H	4.282123000	20.327707000	4.242697000
C	-0.916973000	21.658753000	3.864032000
H	-0.998970000	22.490010000	4.571378000
H	-0.981170000	20.725440000	4.422351000
H	-1.762472000	21.734418000	3.177950000
C	3.711064000	24.717546000	4.912310000
H	4.303963000	24.031605000	4.295578000
H	4.112828000	25.725019000	4.736645000

4a (*S*=1)

Zero-point correction= 0.761612 (Hartree/Particle)

Thermal correction to Energy= 0.814197
Thermal correction to Enthalpy= 0.815141
Thermal correction to Gibbs Free Energy= 0.674587
Sum of electronic and zero-point Energies= -2558.111170
Sum of electronic and thermal Energies= -2558.058585
Sum of electronic and thermal Enthalpies= -2558.057641
Sum of electronic and thermal Free Energies= -2558.198195

Cartesian Coordinates

Ru	13.441905000	6.985929000	33.074751000
Ru	13.929865000	14.347554000	32.818416000
O	14.795069000	6.042375000	31.894121000
N	12.372271000	7.621969000	31.499630000
O	12.103902000	7.865798000	34.340868000
O	14.614158000	8.678514000	33.228990000
N	12.208174000	5.366396000	32.784441000
O	10.591311000	7.078111000	30.141757000
O	14.458024000	6.263856000	34.742879000
N	12.157189000	13.783985000	32.070361000
O	14.872913000	13.481660000	31.230024000
O	10.488709000	14.409998000	30.609783000
C	11.392484000	6.826492000	31.041873000
C	11.343135000	5.515666000	31.766752000
N	13.462117000	15.998606000	31.686652000
C	11.603226000	12.463025000	32.289142000
H	10.519025000	12.490676000	32.135340000
H	11.776707000	12.189004000	33.336831000
O	13.058619000	15.282443000	34.397484000
O	14.279101000	12.623633000	33.905567000
C	15.942362000	6.530268000	31.617599000
O	15.789147000	14.988060000	33.503839000
C	11.540408000	14.615755000	31.215108000
C	11.877527000	10.011215000	31.833739000
H	12.230687000	9.862095000	32.861975000
H	10.789029000	9.864308000	31.849841000
C	12.513867000	8.955342000	30.949415000
H	12.073184000	8.984994000	29.947449000
H	13.584177000	9.174634000	30.841516000
C	11.829707000	16.924169000	30.231935000
H	10.885472000	16.797218000	29.718175000
C	12.241473000	4.223234000	33.469602000
H	12.969168000	4.167110000	34.271239000
C	12.353464000	8.178524000	35.547611000
C	13.433625000	7.724752000	36.305868000
H	13.490928000	8.069995000	37.330357000
C	12.292320000	15.899304000	31.032362000
C	14.376563000	6.773796000	35.900860000
C	11.392088000	3.176366000	33.168629000

H	11.442185000	2.263757000	33.747479000
C	12.968830000	14.756199000	35.557391000
C	14.194845000	17.107336000	31.581389000
H	15.123564000	17.121038000	32.140552000
C	16.101870000	13.158136000	31.202165000
C	15.777568000	8.767449000	32.743321000
C	11.346797000	9.095359000	36.166724000
H	11.592930000	9.355211000	37.195121000
H	10.360304000	8.627321000	36.140460000
H	11.282171000	10.010108000	35.568773000
C	12.587649000	18.075674000	30.112248000
H	12.247300000	18.894132000	29.489964000
C	16.800156000	5.630928000	30.785260000
H	16.905420000	4.665469000	31.284961000
H	17.787409000	6.049851000	30.599185000
H	16.305950000	5.443094000	29.829128000
C	16.434750000	7.777188000	31.994381000
H	17.433550000	8.019535000	31.653999000
C	10.490314000	3.321575000	32.127487000
H	9.813658000	2.515398000	31.872320000
C	16.484467000	12.304784000	30.034577000
H	15.862965000	11.403734000	30.028223000
H	17.535338000	12.020153000	30.047808000
H	16.272335000	12.842761000	29.107738000
C	16.892171000	14.458715000	33.171486000
C	13.786971000	18.169686000	30.798377000
H	14.406991000	19.053622000	30.731399000
C	10.467004000	4.508258000	31.416452000
H	9.785285000	4.674756000	30.592492000
C	12.331185000	15.645221000	36.577401000
H	12.905947000	16.570701000	36.657895000
H	12.265477000	15.178177000	37.558305000
H	11.328590000	15.920546000	36.242491000
C	12.232468000	11.413256000	31.390512000
H	13.323526000	11.532302000	31.419113000
H	11.926263000	11.589704000	30.351125000
C	13.387910000	13.482908000	35.935352000
H	13.213273000	13.210464000	36.968491000
C	15.369645000	6.259321000	36.894075000
H	16.380310000	6.434015000	36.518000000
H	15.250282000	5.178502000	36.998179000
H	15.264224000	6.725141000	37.872421000
C	17.075044000	13.551405000	32.121739000
H	18.081871000	13.192359000	31.950324000
C	13.998044000	12.505354000	35.131735000
C	18.077939000	14.900366000	33.969290000
H	18.200769000	15.981061000	33.867220000
H	18.998010000	14.407405000	33.659996000
H	17.897829000	14.698070000	35.027541000
C	16.483461000	10.057981000	33.013181000

H	15.796946000	10.896183000	32.860352000
H	17.374122000	10.185374000	32.399320000
H	16.780345000	10.088688000	34.065988000
C	14.361717000	11.190120000	35.745011000
H	14.016540000	10.372730000	35.104255000
H	13.957221000	11.067528000	36.749117000
H	15.451406000	11.107157000	35.794643000

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5 (*S*=0)

Zero-point correction= 0.772740 (Hartree/Particle)
Thermal correction to Energy= 0.825176
Thermal correction to Enthalpy= 0.826120
Thermal correction to Gibbs Free Energy= 0.684424
Sum of electronic and zero-point Energies= -2408.817438
Sum of electronic and thermal Energies= -2408.765002
Sum of electronic and thermal Enthalpies= -2408.764057
Sum of electronic and thermal Free Energies= -2408.905754

Cartesian Coordinates

Ru	12.256729000	8.563959000	8.576256000
O	10.988321000	7.342742000	7.483838000
O	13.494155000	9.846351000	9.625535000
O	12.162494000	9.865983000	6.944709000
O	10.642659000	9.672256000	9.321129000
N	13.843686000	7.475659000	7.901859000
N	12.499528000	7.217084000	10.078642000
C	10.268510000	6.866109000	11.067153000
H	9.750729000	6.899842000	12.031965000
H	9.831281000	7.649279000	10.443047000
C	14.234431000	6.463839000	8.720632000
C	11.729263000	7.185231000	11.313604000
H	11.820826000	8.173211000	11.776789000
H	12.165831000	6.447161000	12.001355000
C	15.285107000	5.619245000	8.388268000
H	15.563559000	4.825434000	9.070919000
C	14.491732000	7.651319000	6.744585000
H	14.137565000	8.467444000	6.124490000
C	9.035216000	6.606546000	6.448046000
H	9.433895000	6.707029000	5.435613000
H	7.958471000	6.770714000	6.422364000
H	9.233189000	5.579889000	6.768331000
C	15.949133000	5.810549000	7.194351000
H	16.771346000	5.165413000	6.911572000
C	13.466653000	6.374373000	9.931097000
H	13.705115000	5.632010000	10.690828000
C	9.747038000	7.558969000	7.363787000

C	15.542533000	6.844681000	6.359980000
H	16.035605000	7.027964000	5.414141000
C	14.757423000	11.745566000	10.104273000
H	14.200833000	11.969808000	11.017560000
H	15.110421000	12.679011000	9.668450000
H	15.617779000	11.138910000	10.396124000
C	13.884431000	10.960845000	9.169422000
C	9.001380000	8.560378000	7.998878000
H	7.941858000	8.581888000	7.771467000
C	9.454153000	9.512711000	8.925602000
C	12.779883000	10.967898000	6.904568000
C	13.586495000	11.509896000	7.915163000
H	14.029340000	12.475473000	7.703186000
C	8.435312000	10.394370000	9.585739000
H	8.262769000	10.025662000	10.602985000
H	7.480740000	10.402747000	9.060244000
H	8.817551000	11.412659000	9.672732000
C	12.624816000	11.735162000	5.624364000
H	13.082491000	11.168429000	4.808702000
H	13.083619000	12.722058000	5.664494000
H	11.564434000	11.839061000	5.385112000
N	6.010675000	5.864079000	13.023809000
N	7.668511000	5.553792000	11.085750000
C	10.054670000	5.512434000	10.417455000
H	10.656295000	5.441892000	9.503150000
H	10.396599000	4.719415000	11.090668000
C	5.954210000	6.805027000	12.044222000
C	8.608832000	5.281288000	10.008333000
H	8.465153000	4.241007000	9.702597000
H	8.366884000	5.927512000	9.154829000
C	5.070267000	7.874033000	12.109512000
H	5.058397000	8.602481000	11.307281000
C	5.192370000	5.980774000	14.076069000
H	5.284850000	5.207951000	14.831844000
C	4.228051000	7.983646000	13.196812000
H	3.530653000	8.808345000	13.272200000
C	6.883471000	6.574406000	10.972818000
H	6.919134000	7.235115000	10.106772000
C	4.293038000	7.019459000	14.195888000
H	3.651004000	7.070071000	15.065552000
Ru	7.407632000	4.410198000	12.732370000
O	8.745502000	5.495865000	13.879353000
O	6.075459000	3.261988000	11.641712000
O	7.000277000	3.345430000	14.483036000
O	8.835559000	2.931059000	12.352180000
C	10.699833000	6.019725000	15.034301000
H	10.166355000	6.193962000	15.971670000
H	11.698701000	5.646194000	15.255730000
H	10.785976000	6.988358000	14.532770000
C	9.908614000	5.085023000	14.167220000

C	4.513089000	1.599054000	11.167373000
H	5.107681000	1.221247000	10.332525000
H	3.969837000	0.771334000	11.620839000
H	3.794710000	2.309727000	10.751530000
C	5.409572000	2.307627000	12.140058000
C	10.509188000	3.892391000	13.748093000
H	11.514722000	3.712339000	14.109071000
C	9.977955000	2.918088000	12.887923000
C	6.182600000	2.383694000	14.536156000
C	5.428482000	1.870632000	13.471048000
H	4.781632000	1.033899000	13.705295000
C	10.853099000	1.762987000	12.498840000
H	11.211535000	1.920646000	11.476589000
H	11.718379000	1.652136000	13.151174000
H	10.272801000	0.838910000	12.498558000
C	6.013684000	1.768613000	15.894444000
H	5.581418000	2.509629000	16.572546000
H	5.372650000	0.888251000	15.883420000
H	6.991502000	1.498260000	16.298815000

5a (*S*=1)

Zero-point correction= 0.759089 (Hartree/Particle)
Thermal correction to Energy= 0.813813
Thermal correction to Enthalpy= 0.814758
Thermal correction to Gibbs Free Energy= 0.664920
Sum of electronic and zero-point Energies= -2558.113085
Sum of electronic and thermal Energies= -2558.058360
Sum of electronic and thermal Enthalpies= -2558.057416
Sum of electronic and thermal Free Energies= -2558.207254

Cartesian Coordinates

Ru	12.391795000	8.830444000	8.499801000
O	11.101599000	7.741857000	7.371191000
O	13.718293000	9.960508000	9.569477000
O	12.696244000	10.070997000	6.863248000
O	10.927293000	10.181468000	9.053620000
N	13.787218000	7.416737000	7.978944000
N	12.244426000	7.570320000	10.066538000
C	9.931534000	7.640852000	10.955297000
H	9.391326000	7.782920000	11.899832000
H	9.569967000	8.417650000	10.273284000
C	13.834597000	6.361002000	8.806739000
C	11.416753000	7.832904000	11.229813000
H	11.601823000	8.863505000	11.552354000
H	11.720669000	7.163580000	12.043520000
C	14.649685000	5.278234000	8.542820000

H	14.654249000	4.449581000	9.239067000
C	14.526778000	7.429793000	6.869577000
H	14.426857000	8.305647000	6.239121000
C	9.145565000	7.192972000	6.234831000
H	9.729363000	6.959728000	5.342292000
H	8.175758000	7.592218000	5.943061000
H	8.991800000	6.252579000	6.772698000
C	15.428225000	5.288141000	7.399541000
H	16.075558000	4.451049000	7.169006000
C	12.962626000	6.435663000	10.021865000
C	9.912738000	8.130266000	7.112165000
C	15.365647000	6.380544000	6.549991000
H	15.956246000	6.424124000	5.644658000
C	14.851717000	11.853753000	10.317280000
H	14.303827000	11.845058000	11.261880000
H	15.084598000	12.881568000	10.044599000
H	15.784624000	11.309973000	10.483772000
C	14.049130000	11.150103000	9.269231000
C	9.287330000	9.282266000	7.582069000
H	8.272452000	9.447430000	7.242639000
C	9.784505000	10.206710000	8.515340000
C	13.166488000	11.247041000	6.940163000
C	13.766834000	11.809244000	8.070064000
H	14.134140000	12.822883000	7.974488000
C	8.874988000	11.290335000	9.001885000
H	8.513674000	11.017951000	9.998842000
H	8.012827000	11.434364000	8.352825000
H	9.426884000	12.225995000	9.100859000
C	13.084536000	12.047904000	5.679789000
H	13.632078000	11.532768000	4.887077000
H	13.486979000	13.052809000	5.795077000
H	12.042190000	12.110888000	5.358916000
N	5.723198000	6.115418000	13.393582000
N	7.322595000	6.205695000	11.344426000
C	9.614353000	6.276145000	10.379822000
H	10.125487000	6.159991000	9.415592000
H	10.028825000	5.504970000	11.034720000
C	5.588223000	7.196122000	12.608997000
C	8.131508000	6.033643000	10.151801000
H	7.982254000	5.009516000	9.789340000
H	7.765851000	6.718975000	9.378319000
C	4.656993000	8.175928000	12.889977000
H	4.584113000	9.027695000	12.225789000
C	4.958394000	5.975845000	14.477017000
H	5.131931000	5.087250000	15.073387000
C	3.854182000	8.035792000	14.007878000
H	3.117217000	8.790873000	14.251923000
C	6.481693000	7.253264000	11.407989000
C	4.006024000	6.918255000	14.812382000
H	3.398554000	6.773568000	15.695747000

Ru	7.286811000	4.892028000	12.870169000
O	8.395129000	6.091387000	14.075743000
O	6.143478000	3.648445000	11.713808000
O	7.072212000	3.571635000	14.456754000
O	8.918450000	3.736782000	12.330607000
C	10.246057000	6.863848000	15.257528000
H	9.645685000	6.951533000	16.165351000
H	11.270920000	6.609784000	15.522520000
H	10.238577000	7.844098000	14.772693000
C	9.625843000	5.862279000	14.335982000
C	5.258135000	1.664528000	10.872740000
H	5.836892000	1.762684000	9.951941000
H	5.132023000	0.609473000	11.109325000
H	4.277115000	2.106149000	10.682421000
C	5.936431000	2.421866000	11.970201000
C	10.405943000	4.825954000	13.837888000
H	11.435158000	4.795185000	14.175506000
C	10.051178000	3.864437000	12.874043000
C	6.735550000	2.354162000	14.331124000
C	6.243589000	1.763467000	13.164430000
H	5.984637000	0.714023000	13.220416000
C	11.128791000	2.957766000	12.370580000
H	11.739545000	3.517079000	11.651729000
H	11.789702000	2.634292000	13.175724000
H	10.697164000	2.091714000	11.870698000
C	6.851496000	1.532102000	15.575035000
H	6.181473000	1.935785000	16.338044000
H	6.607212000	0.484419000	15.408312000
H	7.867366000	1.609756000	15.968510000
O	12.981838000	5.527560000	10.854272000
O	6.368165000	8.171042000	10.595088000

References

1 S. Stoll, A. Schweiger, *J. Magn. Reson.*, 2006, **178**, 42.