

Supporting Information

Dinuclear Ru^{II} complexes with quinonoid bridges: tuning the electrochemical and spectroscopic properties of redox-switchable NIR dyes through judicious bridge design

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Crystallographic Data

Table S1 Crystallographic data of **mono2b**^{O,N(Mes)}

Complex	mono2b ^{O,N(Mes)} .0.5CH ₂ Cl ₂
Chemical formula	C _{33.50} H ₃₃ Cl ₂ N ₅ O ₇ Ru
<i>M</i> _r	789.62
Crystal System	triclinic
Space group	<i>P</i> -1
<i>a</i> (Å)	11.800(5)
<i>b</i> (Å)	12.969(5)
<i>c</i> (Å)	24.229(10)
α (°)	89.705(16)
β (°)	76.042(11)
γ (°)	79.798(12)
<i>V</i> (Å ³)	3539(3)
<i>Z</i>	4
Density (g cm ⁻³)	1.482
F(000)	1612
Radiation Type	MoK α
μ (mm ⁻¹)	0.647
Crystal size	0.48 x 0.32 x 0.17
Meas. Refl.	71270
Indep. Refl.	13010
Obsvd. [<i>I</i> > 2 σ (<i>I</i>)] refl.	8734
R _{int}	0.1503
R [<i>F</i> ² > 2 σ (<i>F</i> ²)], wR(<i>F</i> ²), S	0.0637
$\Delta\rho_{\max}$, $\Delta\rho_{\min}$ (e Å ⁻³)	0.759, -0.878

Table S2 (a) Selected bond lengths (Å) and selected bond angles (°) of **mono2b**^{O,N(Mes)}.0.5CH₂Cl₂

Bond	Bond lengths	Bond Angle	
C1- C2	1.501(7)	N3- Ru1- O1	99.62(15)
C1- C6	1.390(7)	N4 -Ru1- O1	93.50(15)
C3- C2	1.402(7)	N2- Ru1- O1	101.13(16)
C3- C4	1.377(7)	N1- Ru1- O1	175.31(15)
C5- C6	1.416(7)	O2- Ru1- O1	77.82(13)
C4- C5	1.521(7)	O1 -C1 -C6	125.3(5)
C1- O1	1.274(6)	O1- C1- C2	115.6(4)
O2 -C2	1.282(6)	O2- C2- C1	116.2(4)
Ru1- O1	2.088(3)	O3- C5- C4	116.8(5)
Ru1- O2	2.085(3)	N5- C4- C5	115.0(5)
O3- C5	1.243(6)	N3 -Ru1- N1	82.47(17)
C4 -N5	1.336(6)	N4- Ru1- N1	82.18(17)
Ru1- N3	2.010(4)	N3- Ru1- N4	92.52(17)
Ru1- N4	2.047(5)	N4- Ru1- N2	165.07(17)
Ru1- N2	2.049(4)	N3- Ru1- N2	87.97(17)
Ru1- N1	2.054(4)	N2- Ru1- N1	83.09(17)

(b) Hydrogen bonding parameters [bond lengths (Å) and bond angles (°)]

D-H...A	d(D-H)	d(H...A)	d(D...A)	<(DHA)
N105-H105.....O3	0.88	2.03	2.845(6)	153.2
N5-H5.....O103	0.88	2.07	2.862(6)	149.0

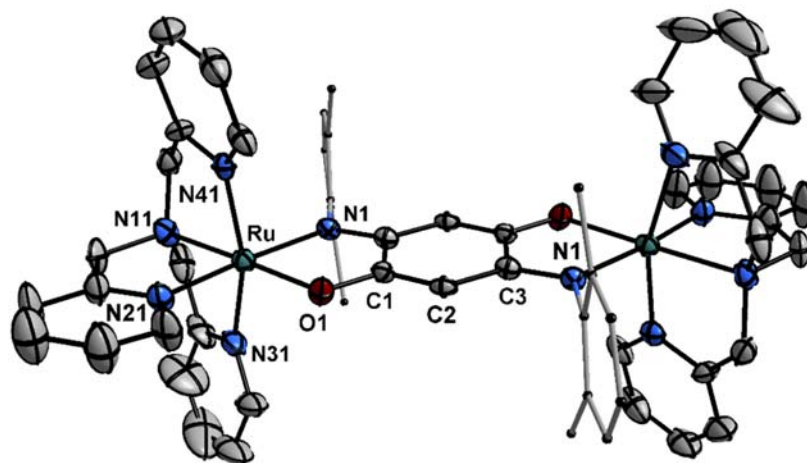


Figure S1: Crystal Structure of $[3^{N(\text{PhMe}_2)}](\text{ClO}_4)_2$ with Ellipsoids Drawn at the 30% Probability Level. H atoms are omitted for clarity.

NMR Spectra

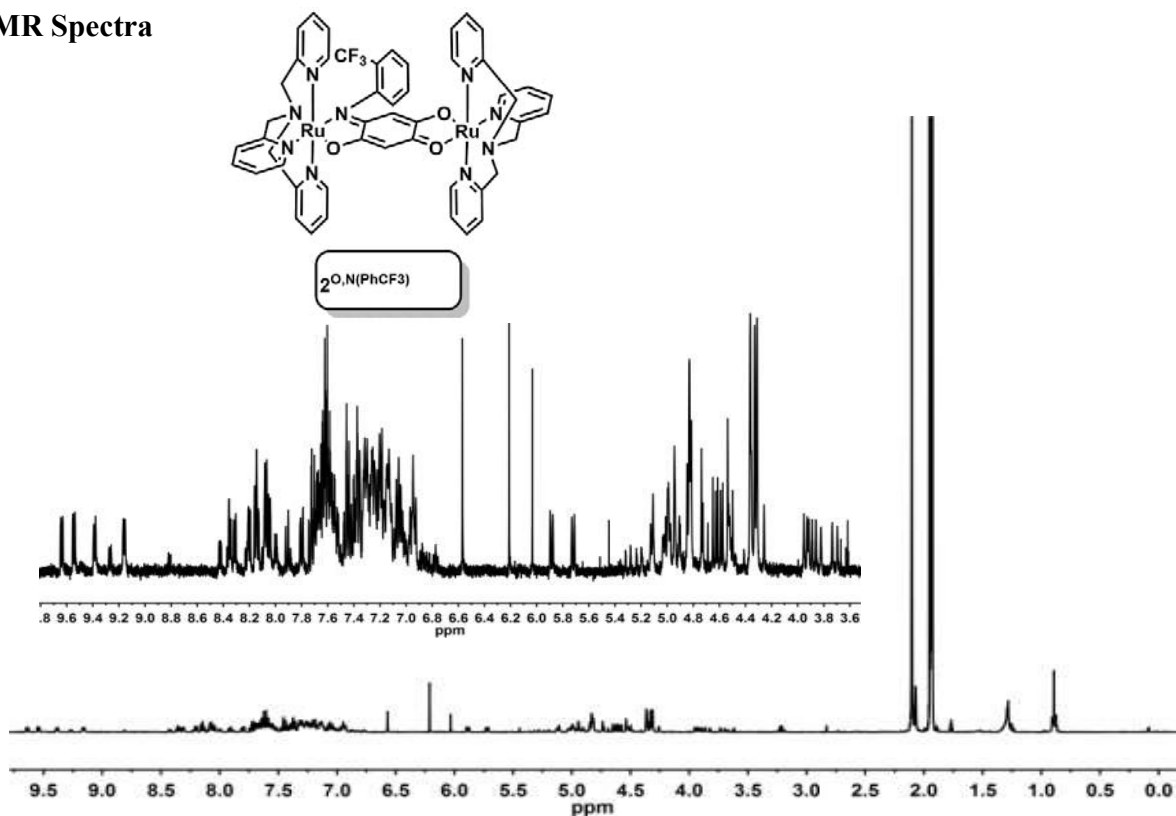


Figure S2: 1H NMR spectrum of $2^{O,N(PhCF_3)}$ recorded in CD_3CN . Inset contains expanded region for better visibility. Bottom: Full spectrum.

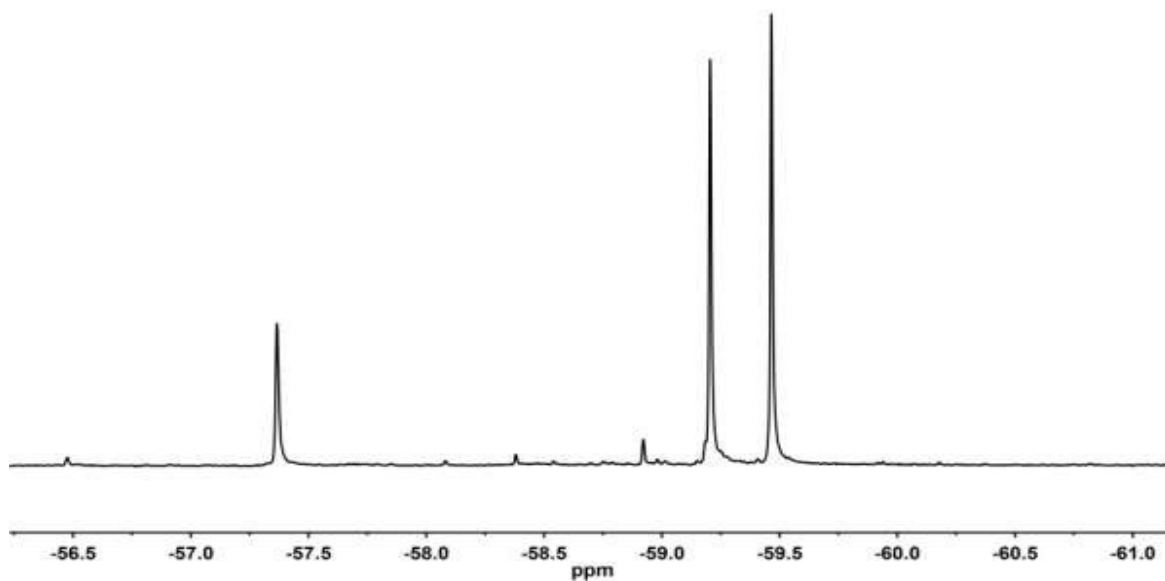


Figure S3: ^{19}F NMR spectrum of $2^{O,N(PhCF_3)}$ recorded in CD_3CN .

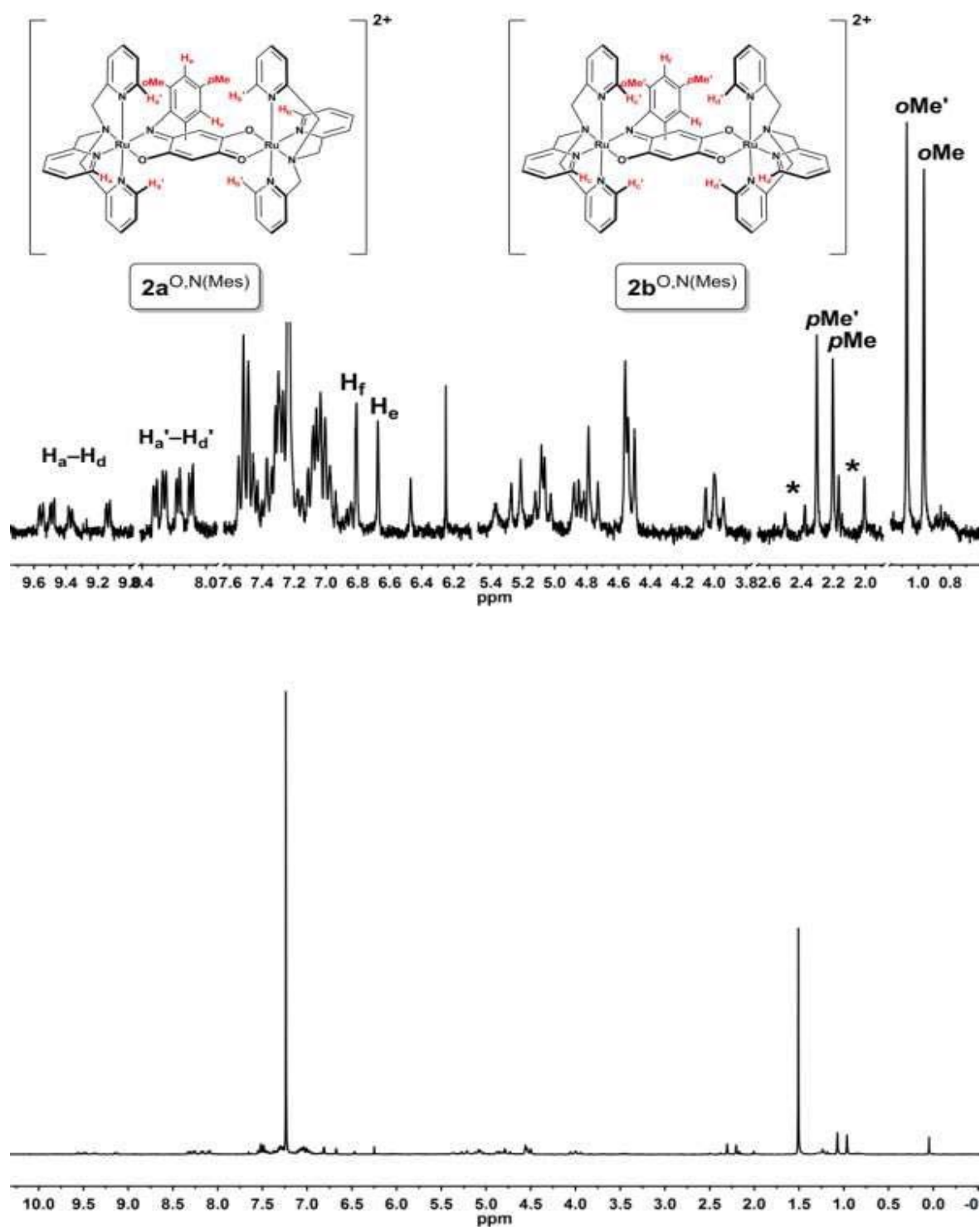


Figure S4: ¹H NMR spectrum of **2**^{O,N(Mes)} recorded in CDCl₃. Top: Parts are cut for better visibility; “*” mark impurities of mono-**2**^{O,N(Mes)}. Bottom: Full spectrum.

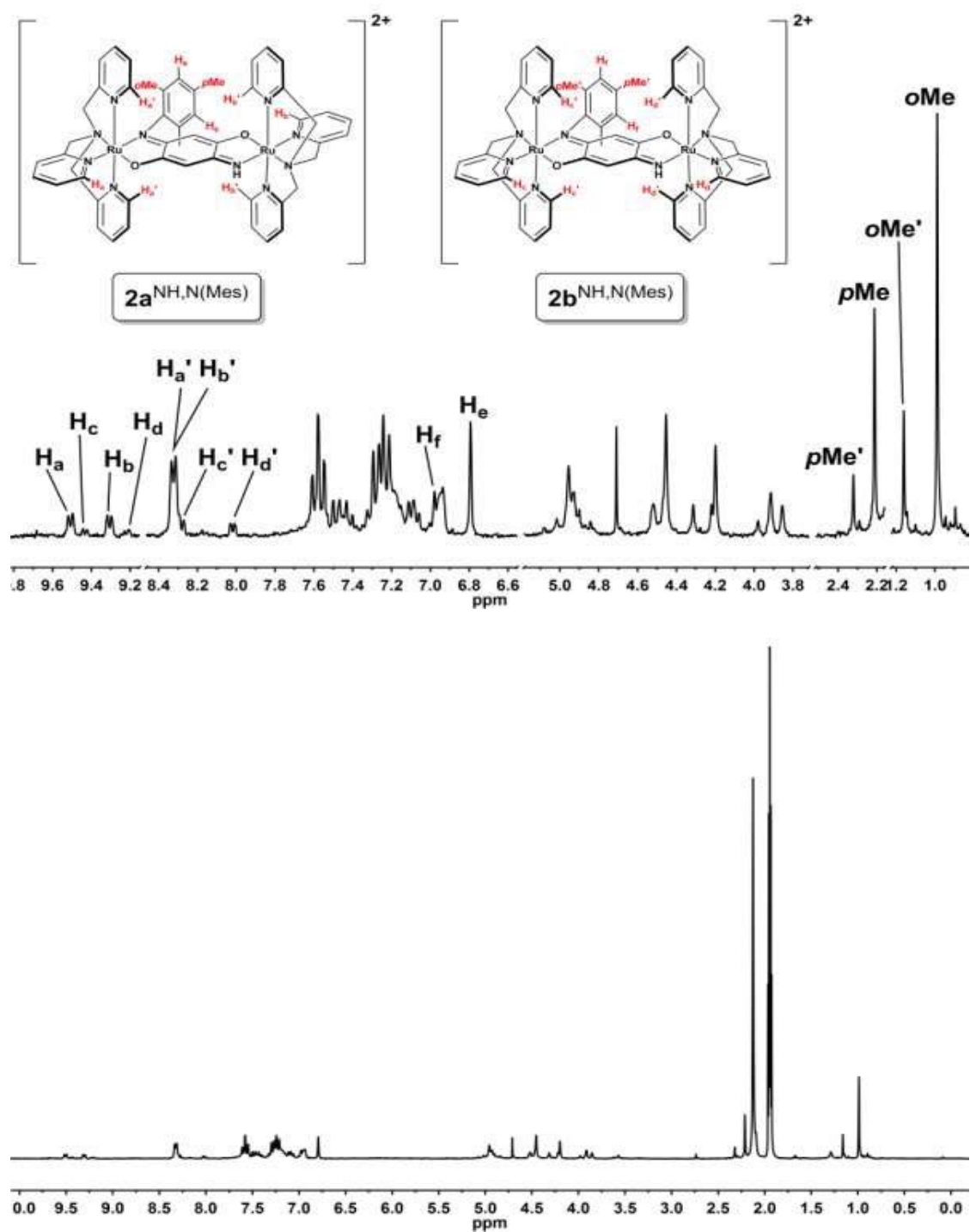


Figure S5: ^1H NMR spectrum of $2^{\text{NH,N(Mes)}}$ recorded in CD_3CN . Top: Parts are cut for better visibility. Bottom: Full spectrum.

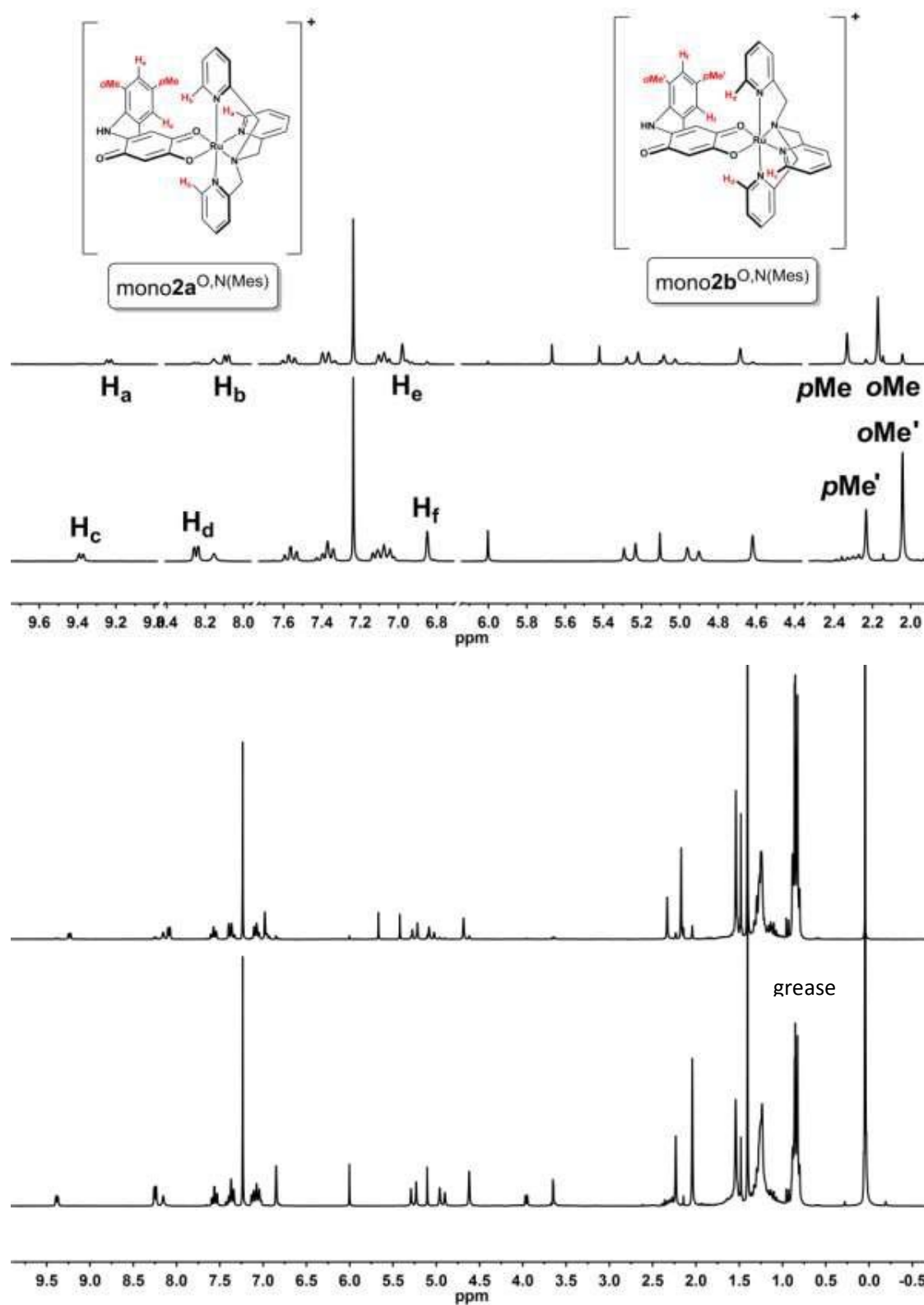


Figure S6: ^1H NMR spectrum of **mono2**^{O,N(Mes)} recorded in CDCl_3 . Top: Parts are cut for better visibility. Bottom: Full spectrum.

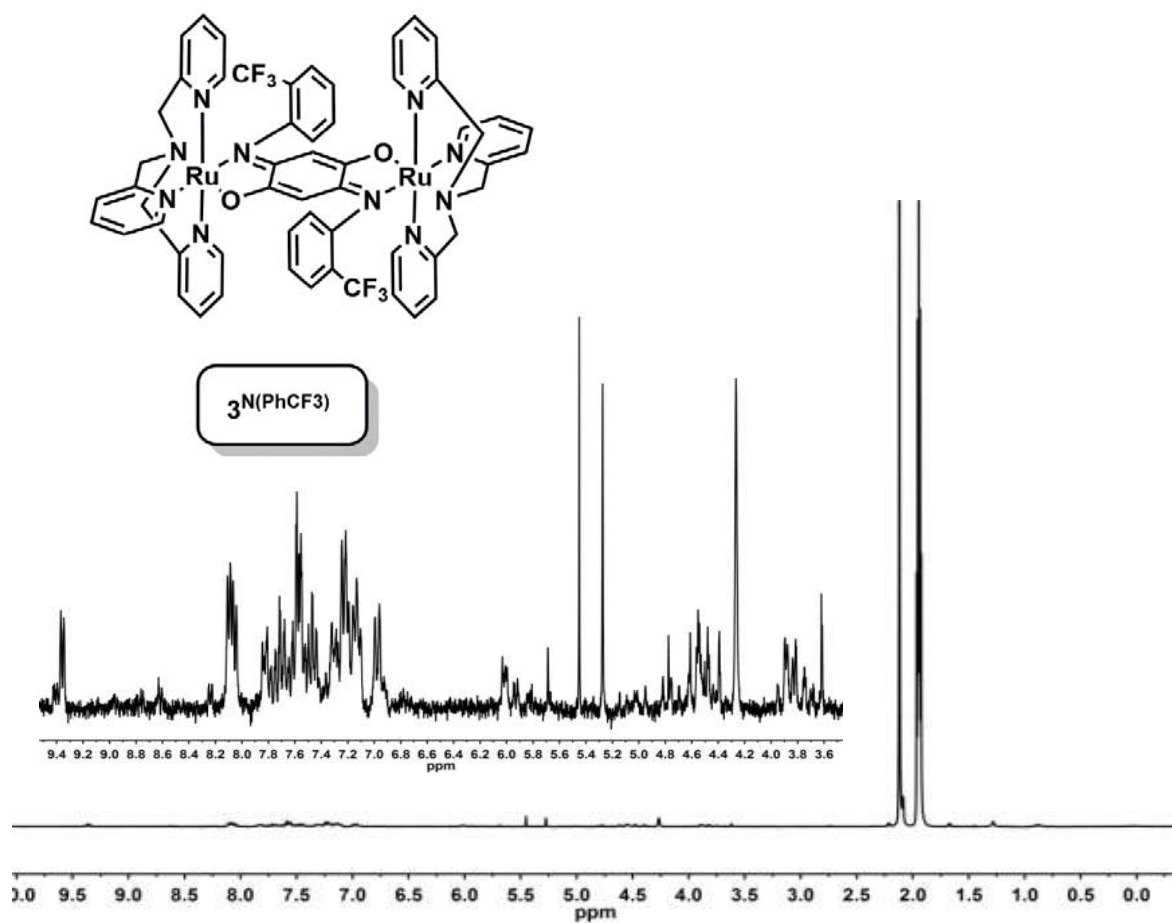


Figure S7: ^1H NMR spectrum of $3^{N(\text{PhCF}_3)}$ recorded in CD_3CN . Inset contains expanded region for better visibility. Bottom: Full spectrum.

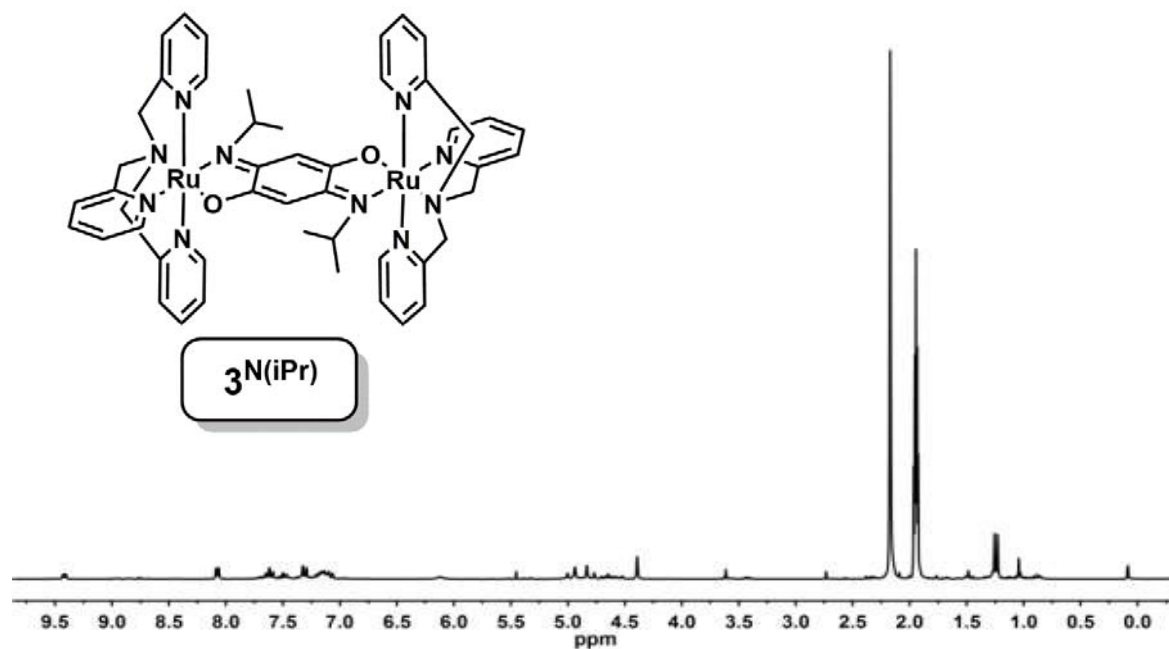
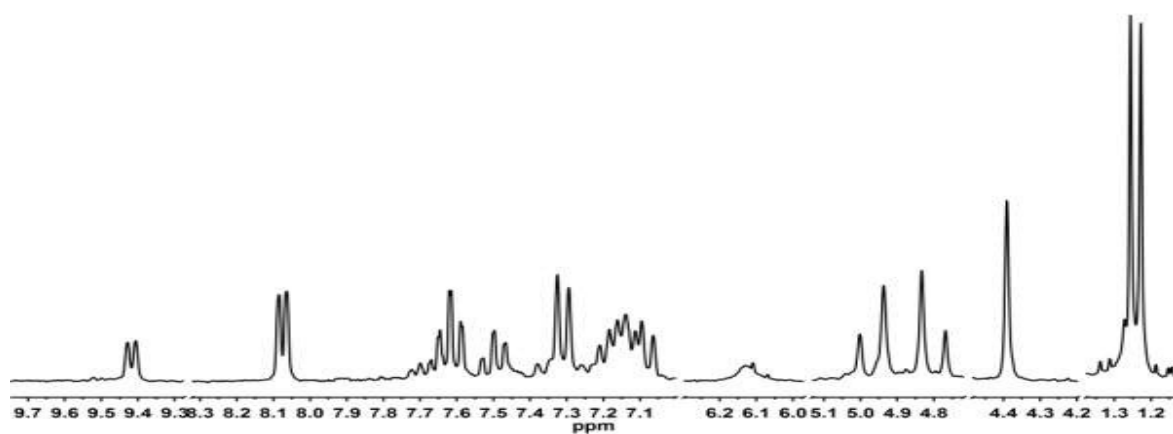


Figure S8: ^1H NMR spectrum of $3^{\text{N(iPr)}}$ recorded in CD_3CN

CV Data

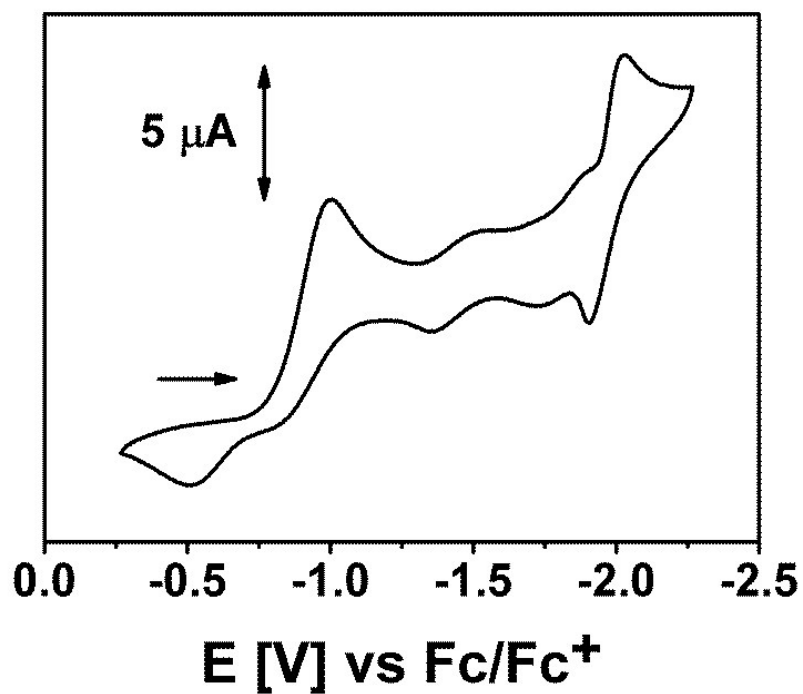


Figure S9: Cyclic voltammogram of $\text{H}_2\text{L}_2^{\text{O,N(PhCF}_3)}$ recorded in CD_3CN (0.1 M Bu_4NPF_6) at 10 mV/s.

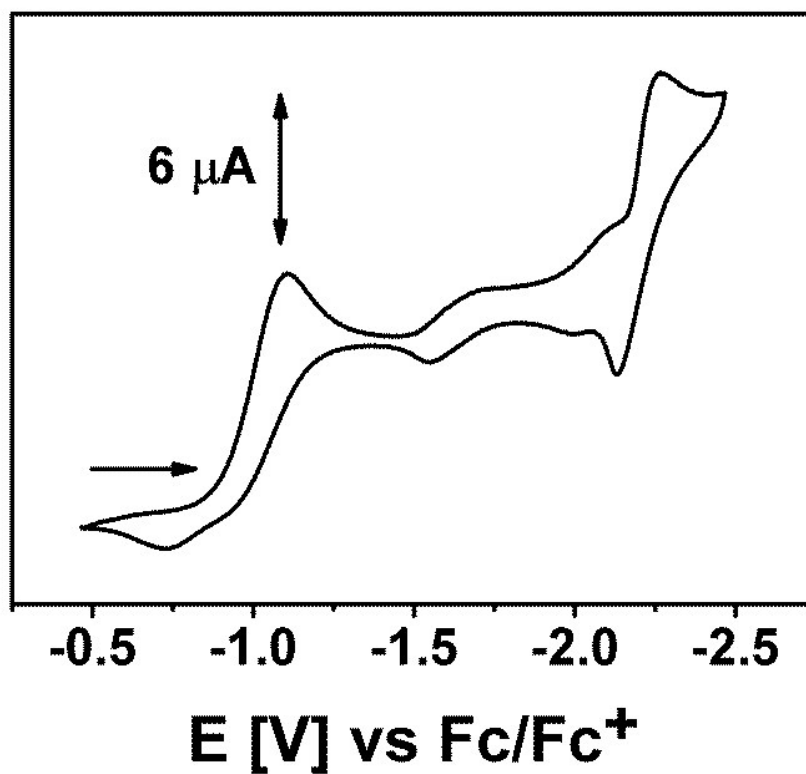


Figure S10: Cyclic voltammogram of $\text{H}_2\text{L}_2^{\text{O,N(Mes)}}$ recorded in CD_3CN ($0.1 \text{ M Bu}_4\text{NPF}_6$) at 10 mV/s .

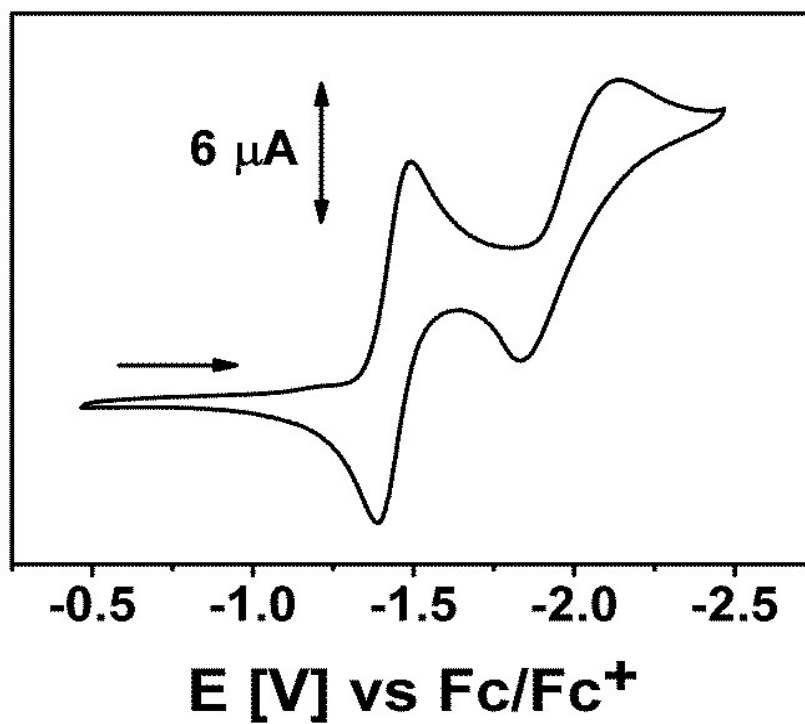


Figure S11: Cyclic voltammogram of $\text{H}_2\text{L2}^{\text{NH}_2\text{N}(\text{Mes})}$ recorded in CD_3CN (0.1 M Bu_4NPF_6) at 10 mV/s.

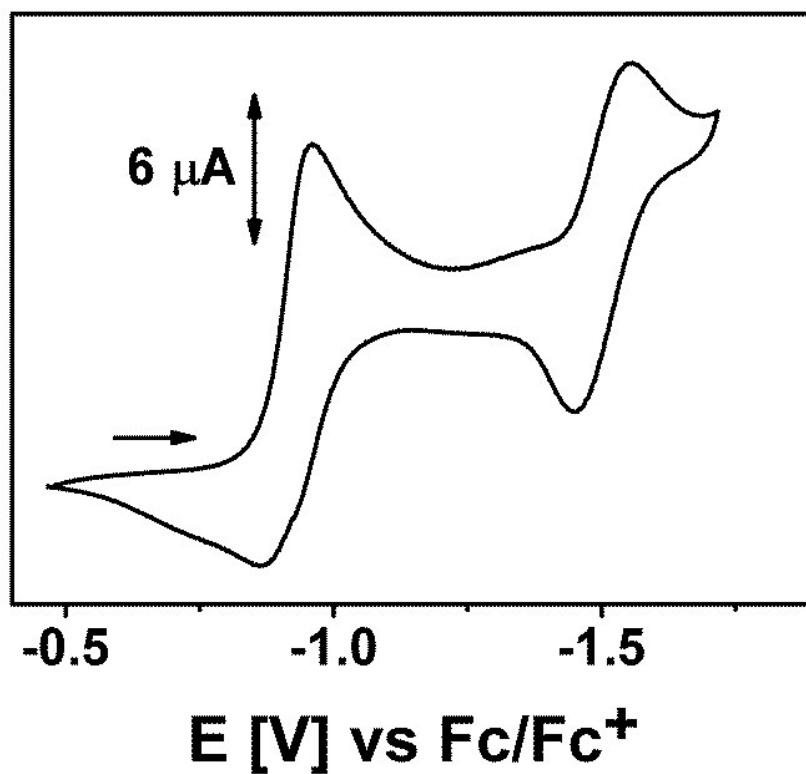


Figure S12: Cyclic voltammogram of $\text{H}_2\text{L3}^{\text{N(PhCF}_3)}$ recorded in CD_3CN (0.1 M Bu_4NPF_6) at 10 mV/s.

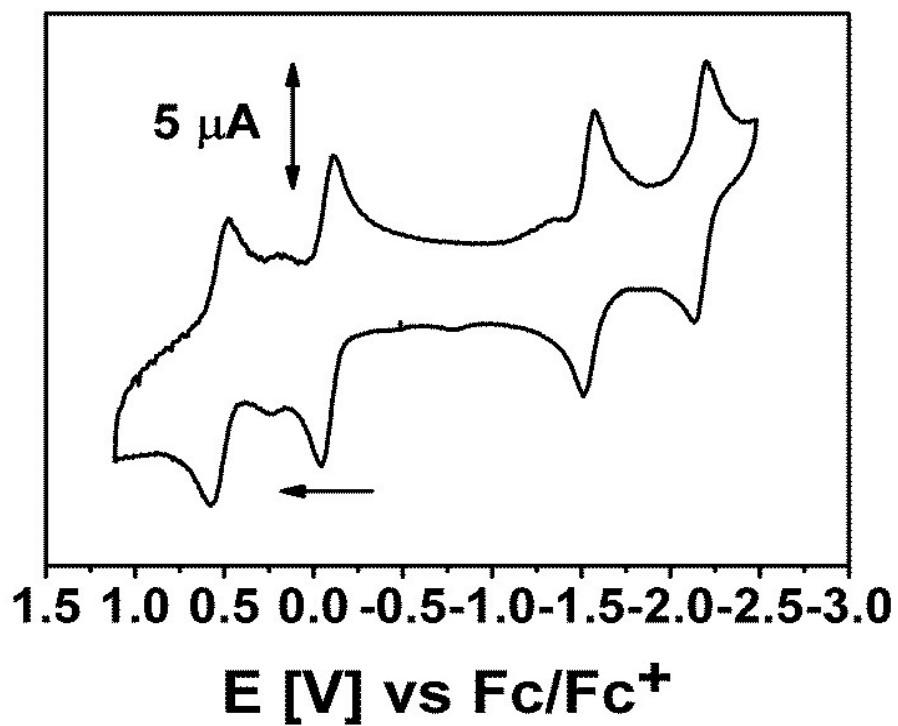


Figure S13: Cyclic voltammogram of $2^{O,N(PhCF_3)}$ recorded in CD_3CN (0.1 M Bu_4NPF_6) at 100 mV/s.

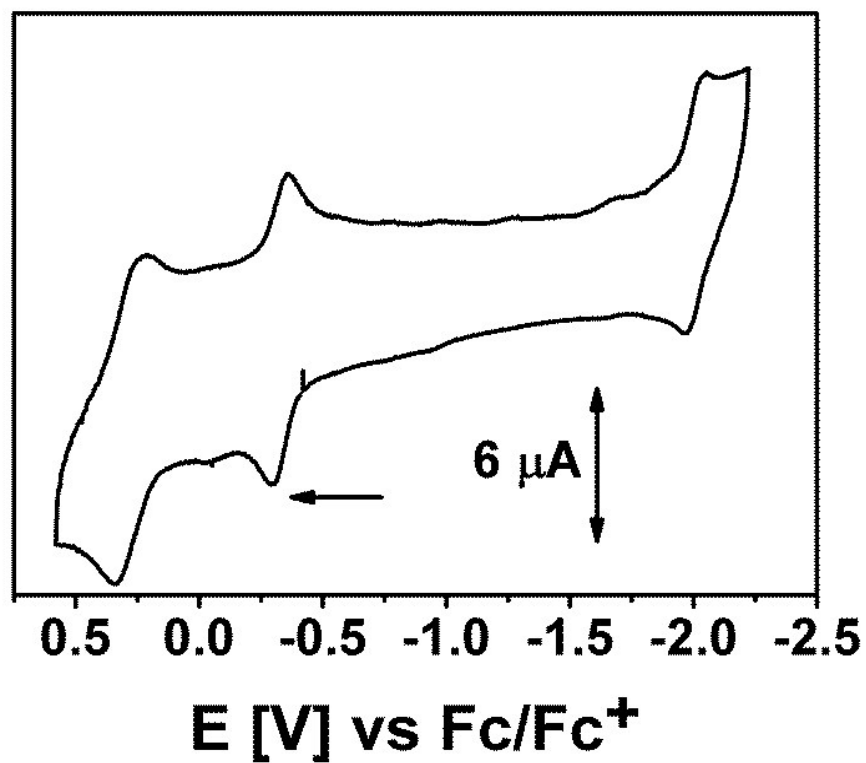


Figure S14: Cyclic voltammogram of $2^{\text{NH,N(Mes)}}$ recorded in CD_3CN (0.1 M Bu_4NPF_6) at 100 mV/s.

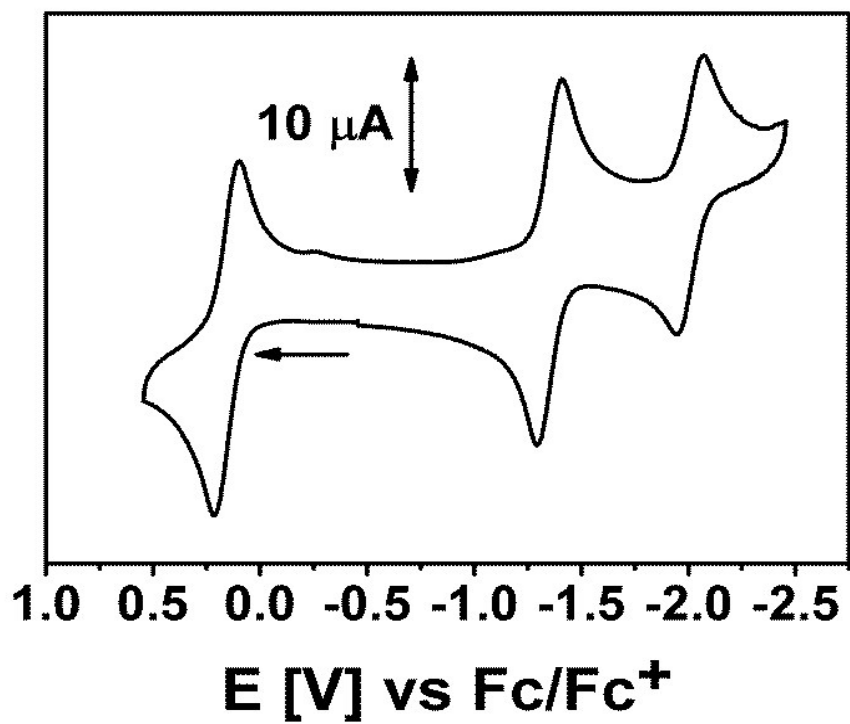


Figure S15: Cyclic voltammogram of **mono2b**^{O,N(Mes)} recorded in CD₃CN (0.1 M Bu₄NPF₆) at 100 mV/s.

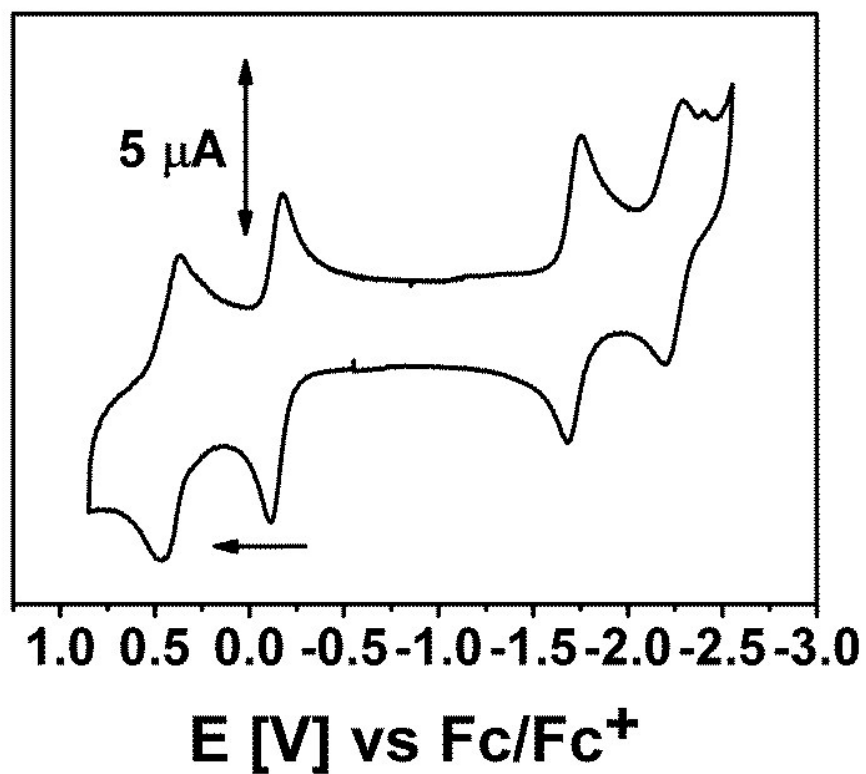


Figure S16: Cyclic voltammogram of $3^{N(\text{PhCF}_3)}$ recorded in CD_3CN (0.1 M Bu_4NPF_6) at 100 mV/s.

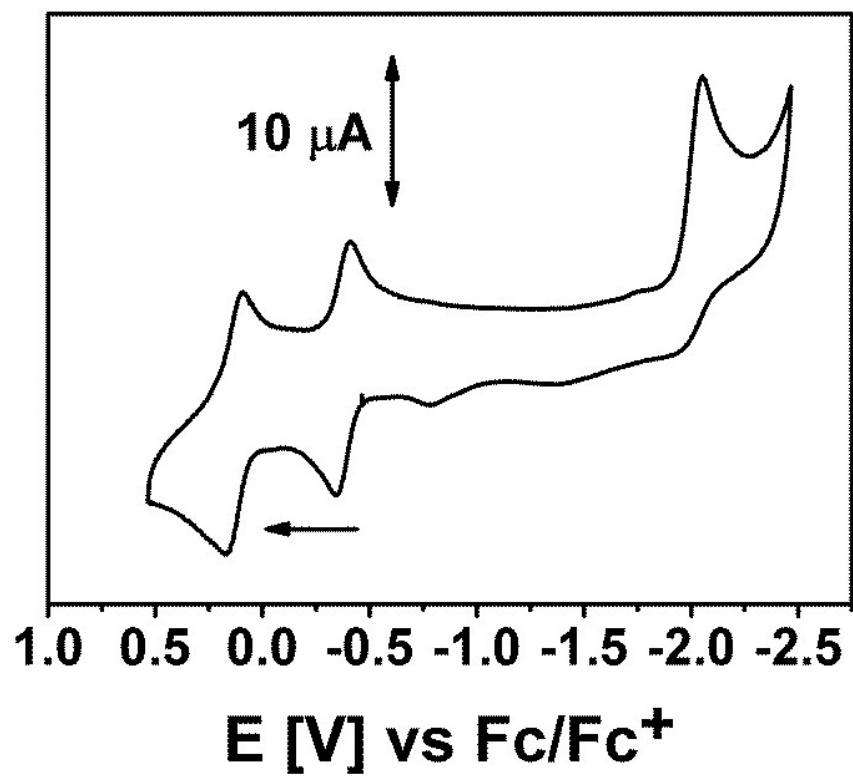


Figure S17: Cyclic voltammogram of $3^{N(iPr)}$ recorded in CD_3CN (0.1 M Bu_4NPF_6) at 100 mV/s.

EPR Data

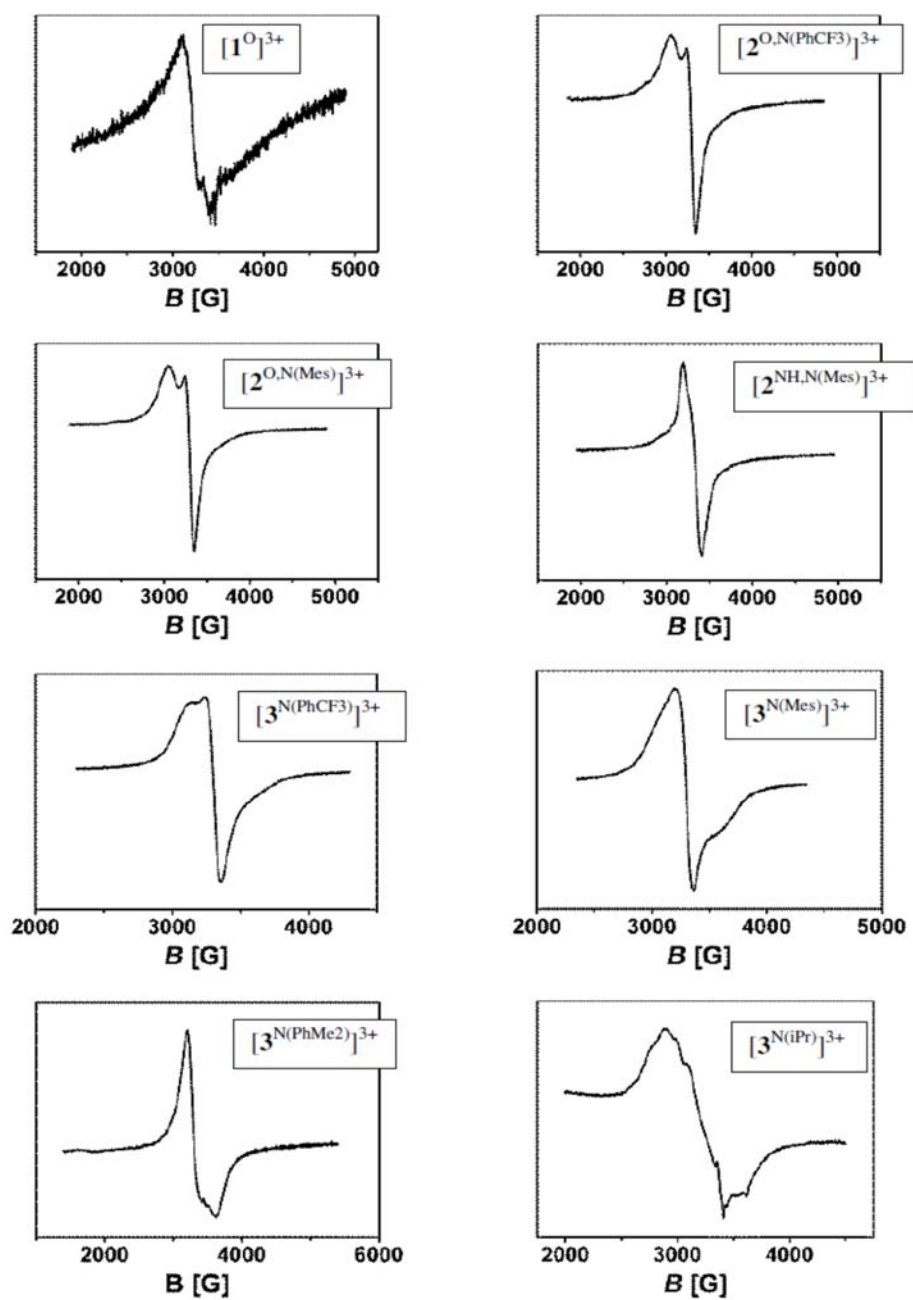


Figure S18: EPR spectra of mixed-valent dinuclear complexes recorded in frozen CH_3CN at 110 K.

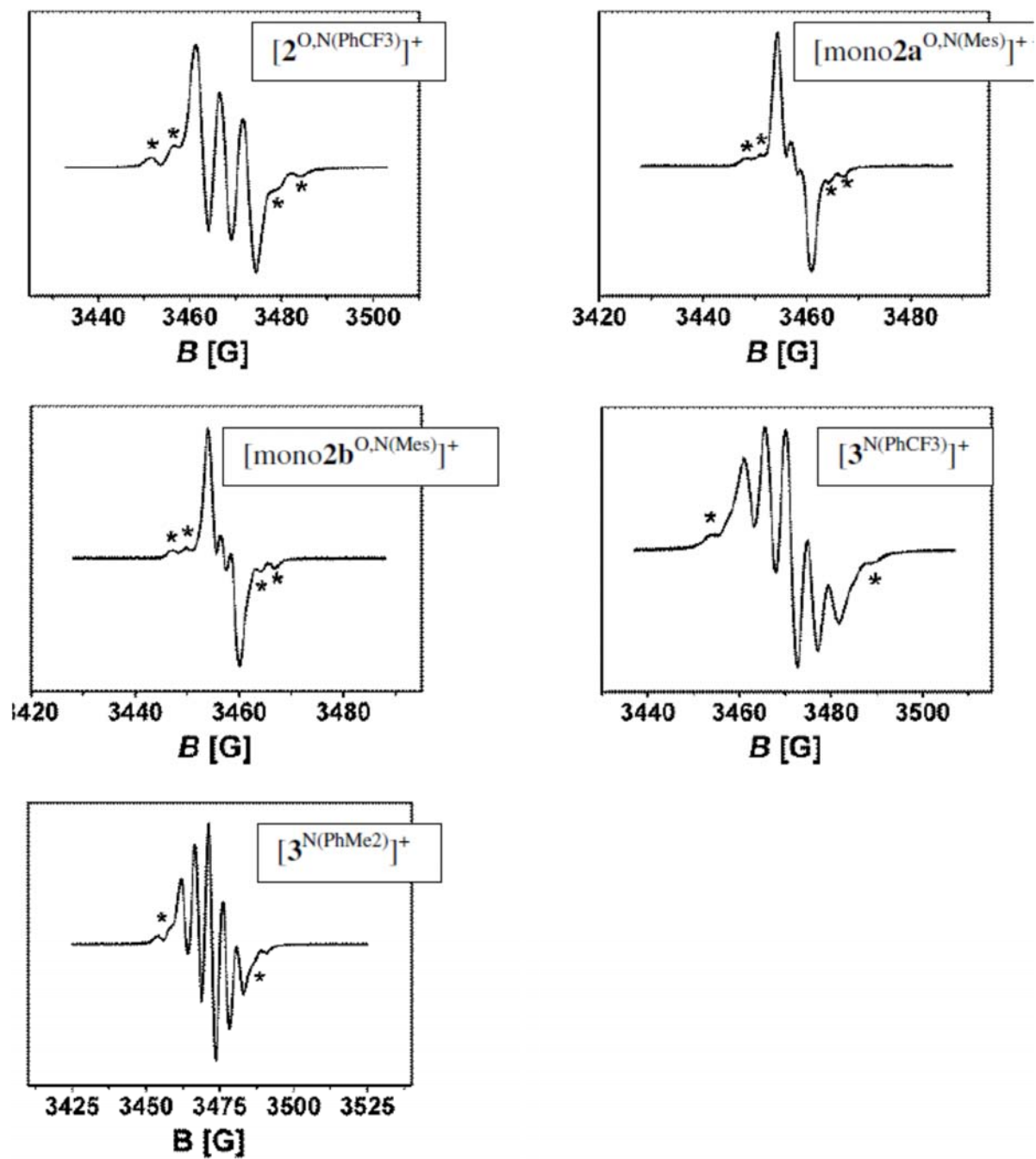
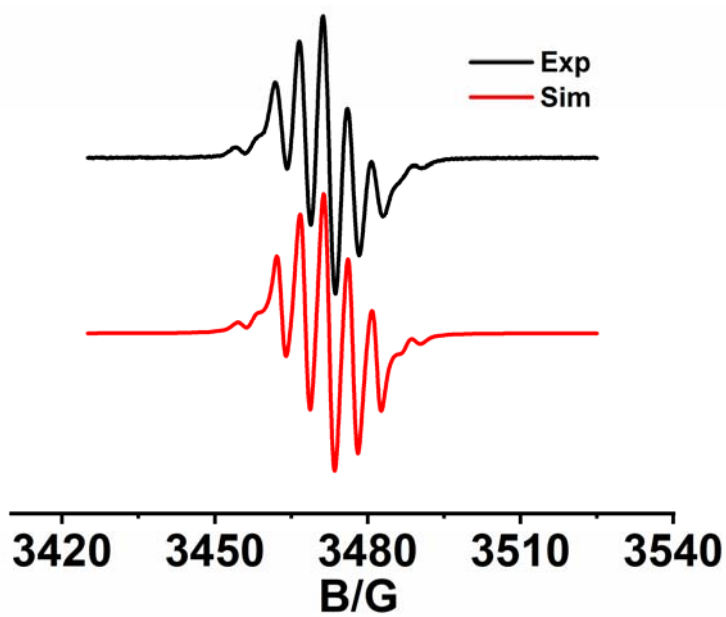


Figure S19: (a) EPR spectra of reduced complexes recorded in CH_3CN at room temperature.



	Simulated parameters of $\mathbf{2}^{\text{NH},\text{N}(\text{Mes})^+}$
g	1.9958
$A(99,101\text{Ru})^*$	3.2850
$A(99,101\text{Ru})^*$	2.9473
$A(14\text{N})^*$	4.9607
$A(14\text{N})^*$	4.4088

* in G

Figure S19: (b) EPR simulated spectra of complex $\mathbf{2}^{\text{NH},\text{N}(\text{Mes})^+}$

Spectroelectrochemistry Data

Table S3 UV/Vis–NIR absorption bands of various oxidation states of the complexes discussed in this paper (recorded in CH₃CN)

Compound	λ_{max} [nm] (ϵ [$10^4 \text{ cm}^{-1} \text{ m}^{-1}$])
[1 ^O] ⁴⁺	298 (0.89), 365 sh, 530 (0.66), 785 (0.23)
[1 ^O] ³⁺	305 (0.71), 375 (0.73), 780 (1.30), 1865 (0.23)
[1 ^O] ²⁺	316 (0.79), 437 (0.83), 765 (0.91)
[1 ^O] ⁺	340 (0.72), 442 (0.90), 515 sh, 750 (0.35)
[1 ^O] ⁰	405 sh, 450 sh, 505 (1.15), 800 sh
[2 ^{O,N(PhCF₃)}] ⁴⁺	300 sh, 543 (3.50), 1087 sh
[2 ^{O,N(PhCF₃)}] ³⁺	317 (2.26), 371 (2.19), 720 (4.61), 1564 (0.71)
[2 ^{O,N(PhCF₃)}] ²⁺	325 (2.63), 417 sh, 450 (2.65), 706 (4.15)
[2 ^{O,N(Mes)}] ⁴⁺	298 sh, 542 (2.88), 589 (2.36), 1132 sh
[2 ^{O,N(Mes)}] ³⁺	377 (2.13), 716 (3.49), 1542 (0.69)
[2 ^{O,N(Mes)}] ²⁺	323 (2.38), 421 (2.61), 449 sh, 686 (3.62)
[2 ^{NH,N(Mes)}] ⁴⁺	306 sh, 471 sh, 580 (4.95), 1126 sh
[2 ^{NH,N(Mes)}] ³⁺	371 (2.63), 580 sh, 708 (5.32), 752 sh, 1435 (1.28)
[2 ^{NH,N(Mes)}] ²⁺	311 (2.63), 376 sh, 422 sh, 479 (2.93), 645 (5.78)
[mono 2 ^{O,N(Mes)}] ²⁺	A: 304 (1.56), 460 (1.33), 496 sh, 1004 sh B: 305 (1.61), 364 sh, 431 sh, 459 (1.33), 496 sh, 685 sh, 966 sh
[mono 2 ^{O,N(Mes)}] ⁺	A: 327 (1.68), 436 (1.17), 648 (1.18) B: 326 (1.88), 437 (1.22), 639 (1.34)
[mono 2 ^{O,N(Mes)}] ⁰	A: 313 (0.99), 353 (0.91), 453 (1.23), 496 (1.27), 669 sh B: 330 (1.01), 355 (1.06), 458 (1.29), 489 (1.29), 659 sh

$[\mathbf{3}^{\text{N(PhCF}_3)}]^{4+}$	304 sh, 503 sh, 568 (4.16), 946 sh, 1091 sh
$[\mathbf{3}^{\text{N(PhCF}_3)}]^{3+}$	333 (2.17), 392 sh, 729 (6.15), 1248 sh, 1492 (1.47)
$[\mathbf{3}^{\text{N(PhCF}_3)}]^{2+}$	332 (2.80), 408 (1.99), 467 (2.26), 699 (5.39)
$[\mathbf{3}^{\text{N(PhCF}_3)}]^+$	310 (4.16), 433 (3.05), 790 (4.07)
$[\mathbf{3}^{\text{N(Mes)}}]^{4+}$	310 (1.79), 478 sh, 585 (2.86), 1129 sh
$[\mathbf{3}^{\text{N(Mes)}}]^{3+}$	318 sh, 376 (1.59), 724 (3.59), 850 sh, 1490 (1.35)
$[\mathbf{3}^{\text{N(Mes)}}]^{2+}$	320 (1.71), 418 (1.67), 483 (1.89), 680 (3.55)
$[\mathbf{3}^{\text{N(Mes)}}]^+$	350 (1.45), 468 (0.23), 780 (1.59), 860 sh
$[\mathbf{3}^{\text{N(PhMe}_2)}]^{4+}$	305 (1.73), 470 sh, 580 (3.15), 1085 sh
$[\mathbf{3}^{\text{N(PhMe}_2)}]^{3+}$	325 (1.53), 374 (1.55), 727 (4.06), 835 sh, 1485 (1.42)
$[\mathbf{3}^{\text{N(PhMe}_2)}]^{2+}$	324 (1.91), 410 sh, 485 (1.92), 685 (3.86)
$[\mathbf{3}^{\text{N(PhMe}_2)}]^+$	348 (1.66), 473 (2.53), 578 (1.51), 740 (1.92), 795 (1.86)
$[\mathbf{3}^{\text{N(iPr)}}]^{4+}$	301 sh, 483 sh, 549 (3.50), 1039 sh
$[\mathbf{3}^{\text{N(iPr)}}]^{3+}$	321 sh, 371 (2.95), 709 (4.41), 811 sh, 1403 (1.40)
$[\mathbf{3}^{\text{N(iPr)}}]^{2+}$	324 (2.89), 416 (3.04), 480 (2.72), 643 (5.55)

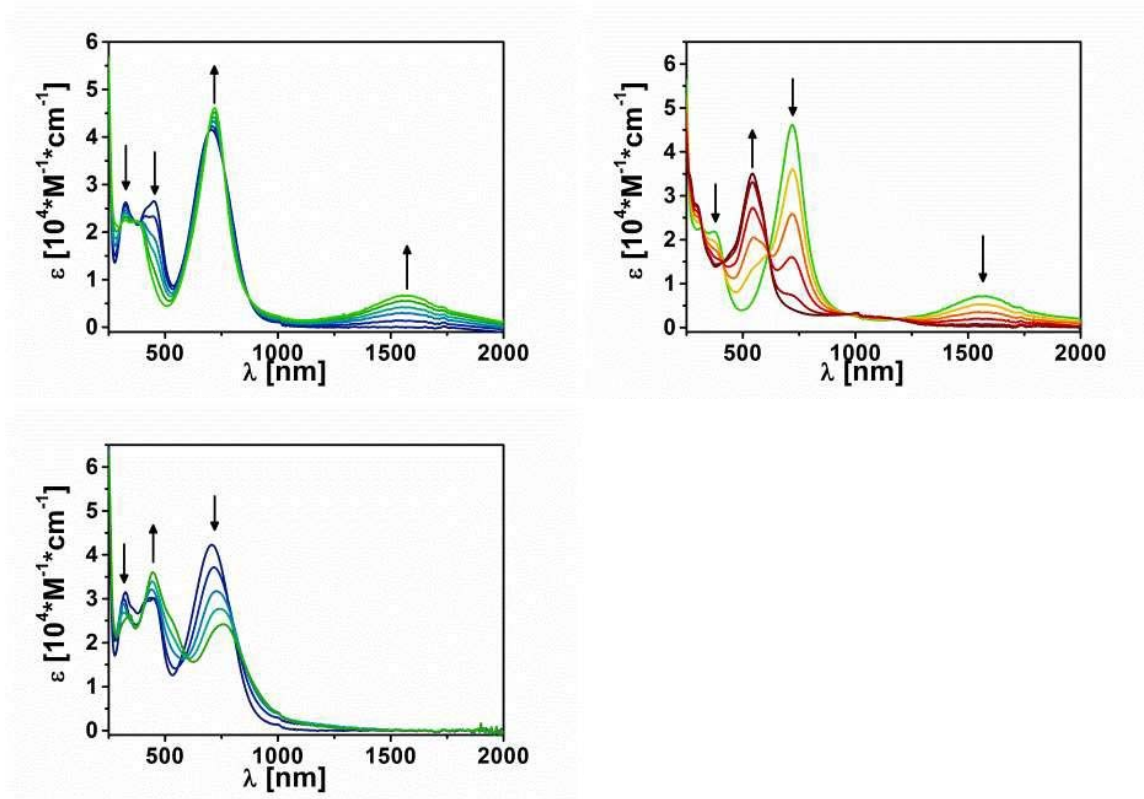


Figure S20: Changes in the UV/Vis–NIR spectrum of $[2^{O,N(\text{PhCF}_3)}]^{2+}$, observed upon oxidation to $[2^{O,N(\text{PhCF}_3)}]^{3+}$ (top left) and $[2^{O,N(\text{PhCF}_3)}]^{4+}$ (top right) and upon reduction to $[2^{O,N(\text{PhCF}_3)}]^{+}$ (bottom left) in acetonitrile (scan rate of 20 mV/s).

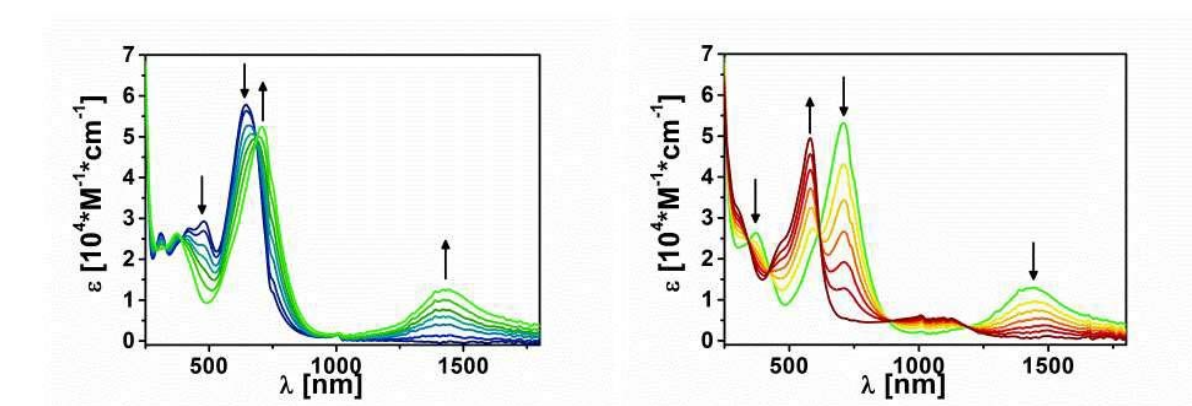


Figure S21: Changes in the UV/Vis–NIR spectrum of $[2^{\text{NH},\text{N}(\text{Mes})}]^{2+}$, observed upon oxidation to $[2^{\text{NH},\text{N}(\text{Mes})}]^{3+}$ (left) and $[2^{\text{O},\text{N}(\text{Mes})}]^{4+}$ (right) in acetonitrile (scan rate of 20 mV/s).

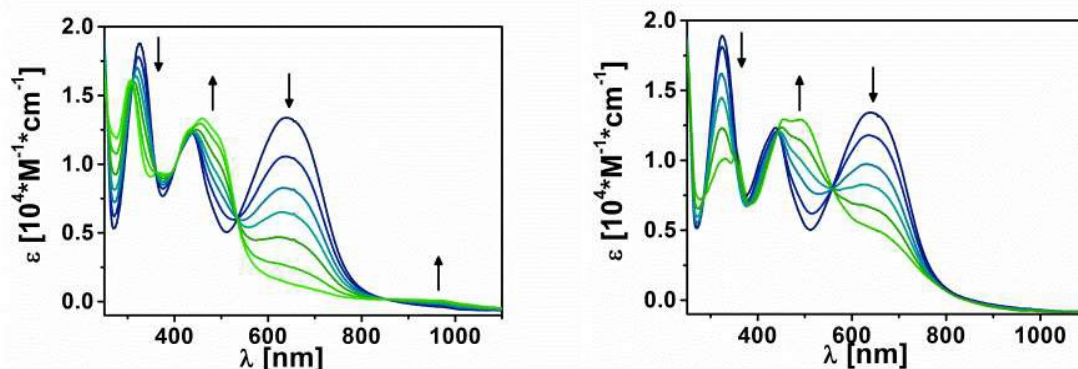


Figure S22: Changes in the UV/Vis–NIR spectrum of $[\text{mono2b}^{\text{O,N(Mes)}}]^+$, observed upon oxidation to $[\text{mono2b}^{\text{O,N(Mes)}}]^{2+}$ (left) and reduction to $[\text{mono2b}^{\text{O,N(Mes)}}]^0$ (right) in acetonitrile (scan rate of 20 mV/s).

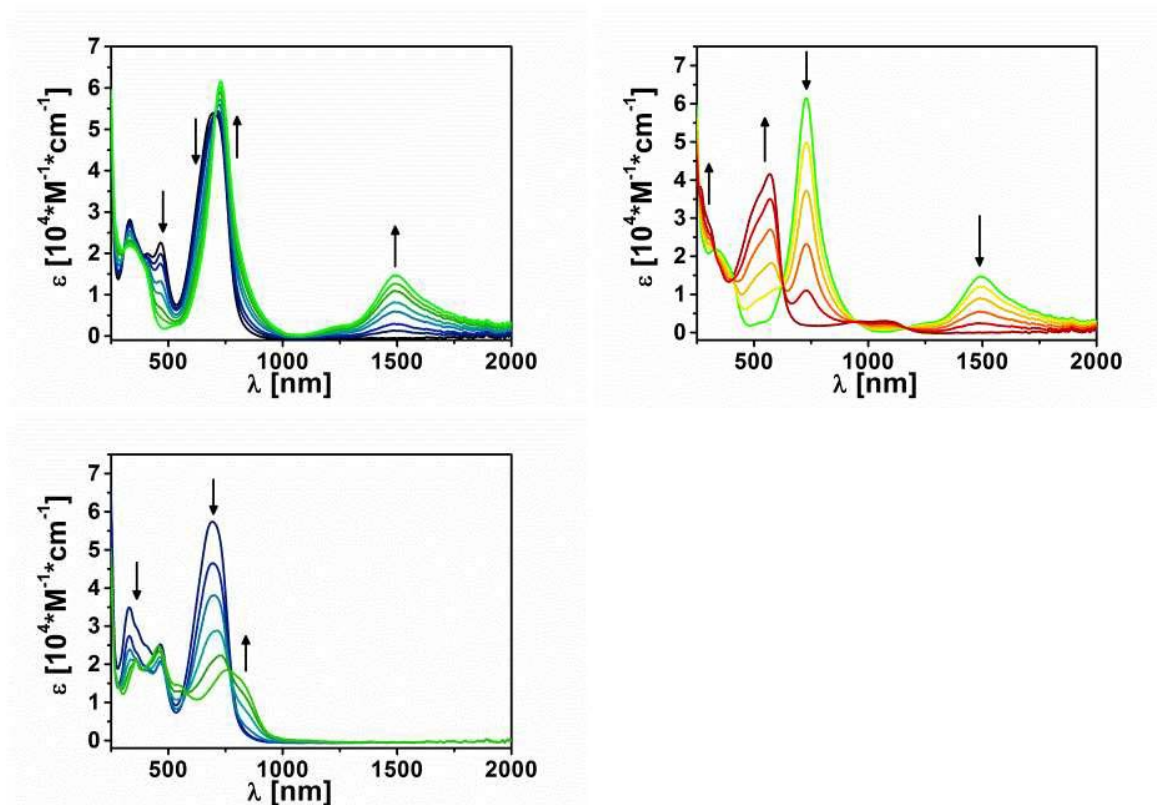


Figure S23: Changes in the UV/Vis–NIR spectrum of $[\mathbf{3}^{\text{N(PhCF}_3\text{)}}]^{2+}$, observed upon oxidation to $[\mathbf{3}^{\text{N(PhCF}_3\text{)}}]^{3+}$ (top left) and $[\mathbf{3}^{\text{N(PhCF}_3\text{)}}]^{4+}$ (top right) and upon reduction to $[\mathbf{3}^{\text{N(PhCF}_3\text{)}}]^+$ (bottom left) in acetonitrile (scan rate of 20 mV/s).

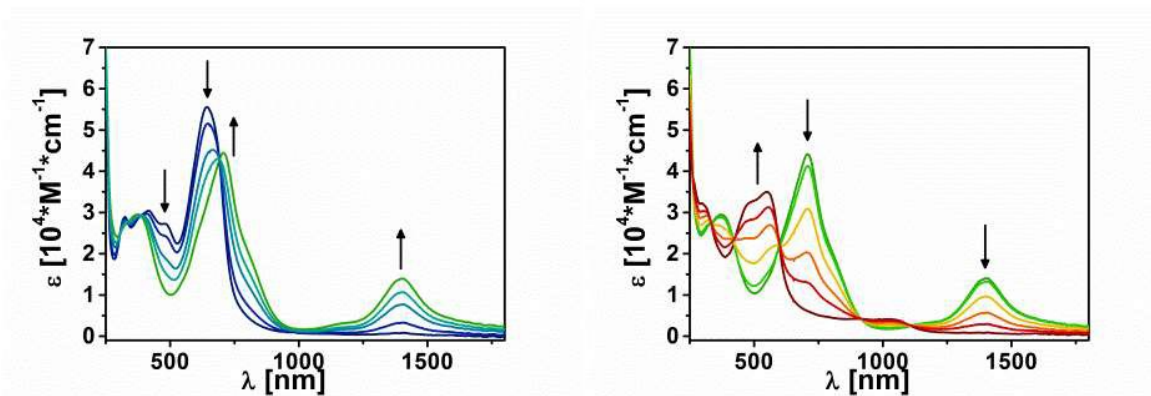


Figure S24: Changes in the UV/Vis–NIR spectrum of $[3^{\text{N(iPr)}}]^{2+}$, observed upon oxidation to $[3^{\text{N(iPr)}}]^{3+}$ (left) and $[3^{\text{N(iPr)}}]^{4+}$ (right) in acetonitrile (scan rate of 20 mV/s).

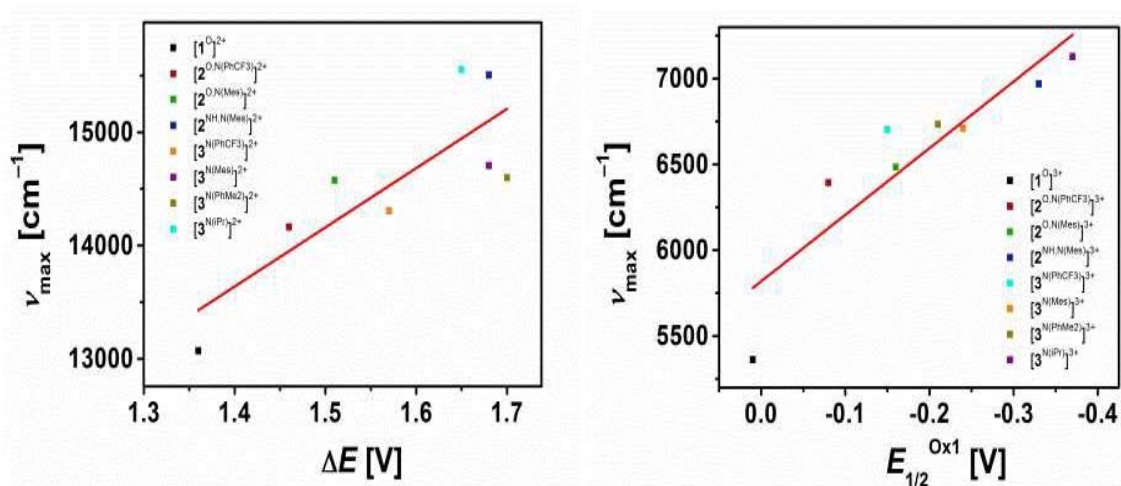


Figure S25: Left: ν_{max} of the MLCT bands of the native dinuclear species plotted against the potential gap between first oxidation and first reduction determined by cyclic voltammetry. Right: ν_{max} of the NIR bands of the mono-oxidized mixed-valent species plotted against the potential of the first oxidation.

DFT Calculations

Table S4 Bond lengths of the DFT-optimized geometries Of $3^{N(\text{Mes})}$ and $2^{NH,N(\text{Mes})}$ in different oxidation states (See Figure 11 for atom labeling).

Bond	Exp. [$3^{N(\text{Mes})}$] ²⁺	DFT $3^{N(\text{Mes})}$				DFT $2^{NH,N(\text{Mes})}$			
		2+	3+	4+	+	2+	3+	4+	+
Ru1–N10 _{ax}	2.044(4)	2.039	2.053	2.060	2.029	2.039	2.051	2.061	2.031
Ru1–N11 _{ax}	2.063(4)	2.054	2.062	2.069	2.050	2.053	2.062	2.069	2.049
Ru1–N12 _{eq}	2.034(4)	2.039	2.054	2.063	2.035	2.038	2.053	2.064	2.035
Ru1–N13 _{amine}	2.074(4)	2.093	2.096	2.108	2.095	2.093	2.102	2.107	2.095
Ru1–N14 _{Mes}	2.097(4)	2.083	2.072	2.045	2.079	2.085	2.088	2.044	2.079
Ru1–O1	2.076(3)	2.068	2.032	2.003	2.081	2.070	2.010	2.006	2.083
N14 _{Mes} –C1	1.330(5)	1.339	1.337	1.340	1.362	1.338	1.336	1.340	1.362
C1–C2	1.417(6)	1.410	1.412	1.410	1.413	1.411	1.411	1.411	1.413
C2–C3	1.375(6)	1.392	1.390	1.389	1.399	1.392	1.390	1.389	1.398
C3–O1 ⁱ /O2	1.305(5)	1.304	1.307	1.308	1.326	1.305	1.305	1.308	1.328
C1 ⁱ –C3/C3–C4	1.481(6)	1.476	1.471	1.467	1.450	1.482	1.478	1.473	1.455
Ru2–N20 _{ax}						2.040	2.057	2.064	2.029
Ru2–N21 _{ax}						2.049	2.059	2.066	2.045
Ru2–N22 _{eq}						2.039	2.056	2.060	2.037
Ru2–N23 _{amine}						2.079	2.077	2.088	2.080
Ru2–N24 _H						2.044	2.000	2.002	2.040
Ru2–O2						2.080	2.077	2.015	2.090
N24 _H –C4						1.332	1.334	1.332	1.356
C4–C5						1.413	1.411	1.411	1.415
C5–C6						1.391	1.390	1.389	1.399
C6–O1						1.305	1.309	1.308	1.326
C1–C6						1.478	1.472	1.469	1.451

Table S5 Calculated and experimental transition energies for different oxidation states of $3^{\text{N(Mes)}}$ and $2^{\text{NH,N(Mes)}}$.

Compound	Main contributing excitation (%)	Calcd. transition energy [nm]	Oscillator Strength	Exp. transition energ[nm] ($e/10^4 \text{ cm}^{-1} \text{ m}^{-1}$)	Character of the transition ^a
$[3^{\text{N(Mes)}}]^{2+}$	HOMO-2→LUMO (λ_3)	640.6	0.0584	680 (3.55)	Ru→Q
	HOMO-1→LUMO+1 (54)	519.8	0.1583	483 (1.89)	Ru→Q→TPA
	HOMO→LUMO +2 (35)				RuQ→TPA
	HOMO → LUMO +8(77)	429.1	0.0284	418 (1.67)	Ru-Q→TPA
	HOMO-5→LUMO +7 (21)	323.4	0.0129	320 (1.71)	Ru→TPA
	HOMO-4→LUMO +8 (28)				Ru→TPA
	HOMO-3→LUMO +7 (24)				Ru→TPA
	$[3^{\text{N(Mes)}}]^{3+}$	HOMO-2(β)→LUMO (β) (85)	1081.6	0.2787	1490 (1.35)
HOMO-5(β)→LUMO (β) (63)		770.4	0.0126	724 (3.59)	Mixed
HOMO-2(β)→LUMO +3(β)		377.9	0.0213	376 (1.59)	Ru→TPA
HOMO-1(β)→LUMO +6(β) (20)		337.6	0.0115	318 (sh)	Ru-Q→TPA
$[3^{\text{N(Mes)}}]^{4+}$		HOMO (α)→LUMO (α) (32)	927.6	0.0486	1129 (sh)
	HOMO-6(β)→LUMO +1(β) (33)	585.3	0.1602	585 (2.86)	Ru-Q→Ru
	HOMO (β)→LUMO +2(β) (27)				Ru→Q
	HOMO-13(β)→LUMO (β) (50)	476.3	0.0618	478 (sh)	TPA→Ru-Q
	HOMO-12(β)→LUMO (β) (22)				TPA→Ru-Q
	$[3^{\text{N(Mes)}}]^{+}$	HOMO (α)→LUMO +8(α) (8 λ)	823.2	0.0027	860 (sh)
HOMO (β)→LUMO (β) (7 λ)		742.1	0.0954	780 (1.59)	Ru-Q→TPA
HOMO-5(β)→LUMO +1(β) (36)		463.2	0.0260	468 (0.23)	Ru→TPA
HOMO-6(α)→LUMO (α) (53)		422.0	0.1144	350 (1.45)	Ru→TPA
$[2^{\text{NH,N(Mes)}}]^{2+}$		HOMO-1→LUMO (λ_4)	726.5	0.0028	763 (sh)
	HOMO-2→LUMO (λ_2)	636.8	0.0656	645 (5.78)	Ru→Q
	HOMO →LUMO +4 (42)	480.6	0.0117	479 (2.93)	Ru-Q→TPA
	HOMO-3→LUMO +2 (47)	423.0	0.0575	422 (sh)	Ru→TPA
	HOMO-3→LUMO +7 (22)	375.0	0.0036	376 (sh)	Ru→TPA
	HOMO-2→LUMO +5 (40)				Ru→TPA
	HOMO-8→LUMO +1 (35)	307.5	0.0276	311 (2.63)	Mes→TPA

	HOMO-6 → LUMO +3 (37)				Ru-Q → TPA
$[2^{\text{NH,N(Mes)}}]^{3+}$	HOMO-1(β) → LUMO (β) (58)	1065.3	0.2594	1435 (1.28)	Ru-Q → Ru-Q
	HOMO-5(β) → LUMO (β) (5 λ)	737.7	0.0070	708 (5.32)	Ru-Q → Ru-Q
	HOMO (β) → LUMO +10(β) (58)	370.0	0.0074	371 (2.63)	Ru-Q → TPA
$[2^{\text{NH,N(Me)}}]^{4+}$	HOMO (α) → LUMO (α) (33)	895.9	0.0599	1126 (sh)	Ru-Q → Q
	HOMO-4(β) → LUMO +1(β) (27)	585.0	0.1471	580 (4.95)	Ru-Q → Ru-Q
	HOMO → LUMO +2(β) (35)				Ru → Ru-Q
	HOMO-11(β) → LUMO (β) (20)	469.2	0.2155	471 (sh)	TPA → Ru-Q

^aQ = quinone; Mes = mesityl

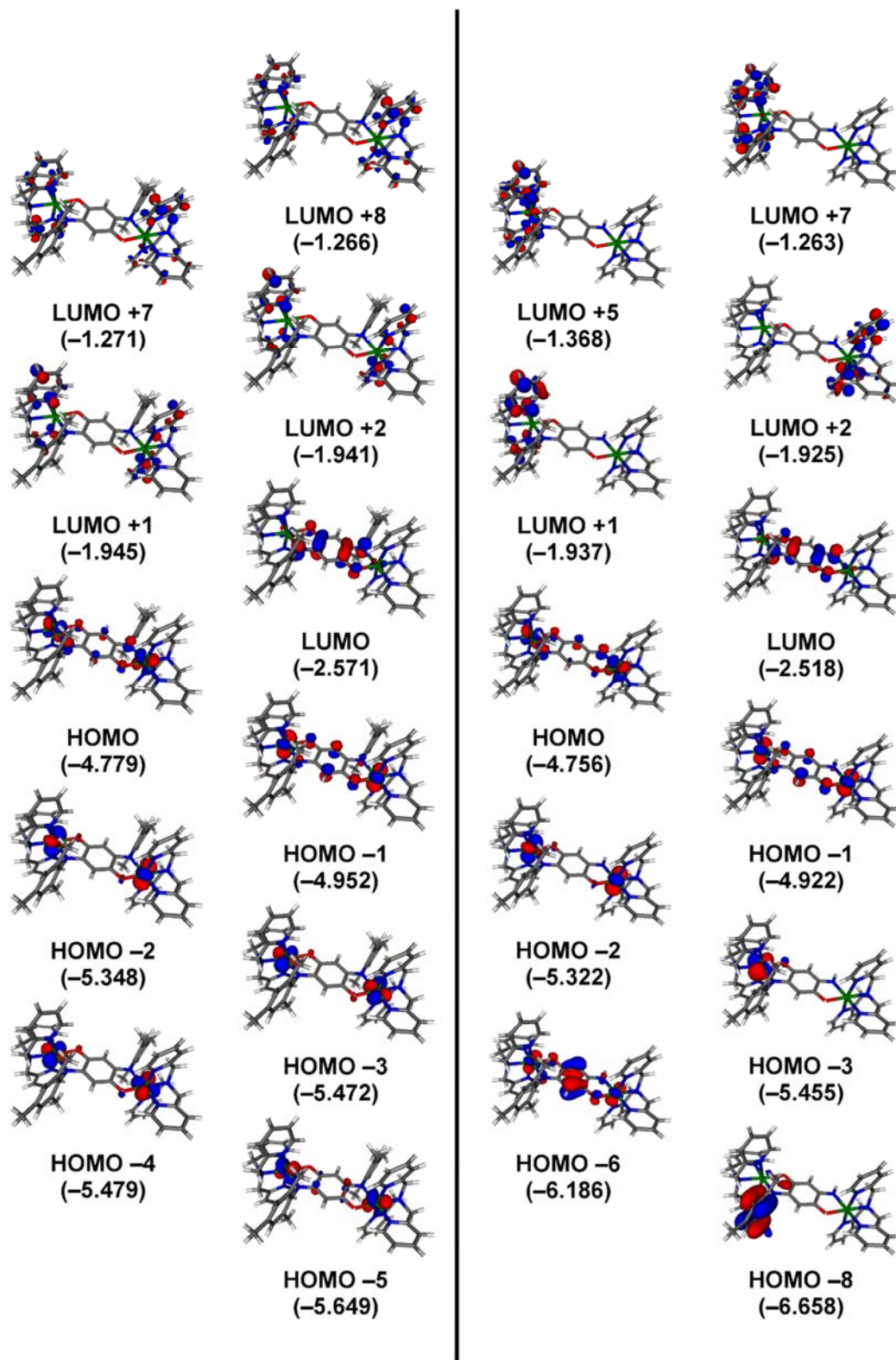


Figure S26: Calculated molecular orbitals of $[3^{\text{N(Mes)}}]^{2+}$ (left) and $[2^{\text{NH,N(Mes)}}]^{2+}$ (right). Orbital energies (in eV) are given in parentheses.

Table S6 Atomic coordinates of the optimised geometry of $[3^{N(\text{Mes})}]^{2+}$.

Ru	2.36455422707026	5.40917945817707	7.52891326578441	H	3.82879927387302	9.83181097763899	5.36307244590164
O	0.90342497984703	6.83122555401484	7.87212326092627	C	3.32071153857601	8.00178021735201	6.36054026757049
N	0.81774812898506	4.27491825215283	8.34173071990938	H	2.36239202315674	8.33801319206077	6.75512833627333
N	3.92545489534924	4.08251206004675	7.09852639518989	C	3.41796828052166	3.21168403544115	6.00078626884117
N	3.65948181570446	6.72804343333323	6.66872202991270	H	2.78736370152037	2.44333851586019	6.47262718783947
N	1.82846218287221	5.01082012651856	5.60231280686115	H	4.23846235072529	2.69986267345454	5.47618673406757
N	3.34465275572129	5.43719322322291	9.33351976315792	C	2.56994620026224	4.00796059156702	5.04231357539773
C	-0.23952013319418	6.38444470934418	8.31433976092895	C	2.47017503364230	3.70790331226118	3.68950613310421
C	-1.34612813354788	7.19839102940884	8.53930951282347	H	3.08486198327162	2.91220198094709	3.26936046510509
H	-1.28469446390487	8.26612627043437	8.32983746523047	C	1.57246563229886	4.42327029242053	2.89071577465823
C	-0.31629241452177	4.93798541340229	8.59947130136825	H	1.47739556813406	4.19477879688214	1.82955560500940
C	0.90603420979434	2.89516283356768	8.68138696011976	C	0.80894333144059	5.43529719836352	3.47379250422116
C	0.68064116518975	1.91140911031005	7.69552809730135	H	0.10045781653233	6.02004466571802	2.88850035381048
C	0.97920866745521	0.57624473356860	7.99819701019475	C	0.97065508976144	5.70930512625608	4.82839671410094
H	0.82916584362714	-0.18249898198911	7.22536270526082	H	0.41810884416656	6.49947251231028	5.33548787867064
C	1.45048970194214	0.18852999086650	9.25735294273611	C	4.19744153311507	3.36546261039499	8.37700591083078
C	1.56784424225126	1.17437368252788	10.24537039048523	H	5.15048213041102	2.81529337723756	8.34500316710684
H	1.89194721975005	0.88947766690151	11.25008140991874	H	3.38496847936297	2.64159582017535	8.52899101843097
C	1.28340395736229	2.52103138527499	9.99123985725022	C	4.18994410460838	4.37491055774004	9.49984901151377
C	0.06743612409472	2.25662055367276	6.36239253189978	C	4.96840674058645	4.24464867188473	10.64226809070639
H	0.63840230915215	1.82877091140695	5.52606910232387	H	5.64019798818638	3.39219370009835	10.73977221225043
H	-0.00793453931750	3.33827624660932	6.21871082186811	C	4.87496420728237	5.20785453904491	11.65243024807129
H	-0.94894101040605	1.83867165955010	6.30164730571912	H	5.47280387680207	5.11483662830421	12.55865236710944
C	1.76962506158915	-1.25309225747318	9.56029068565112	C	4.01288585448144	6.28941682899835	11.47393265358357
H	2.67148643347067	-1.33992622402550	10.18261226051380	H	3.91159866924099	7.06556875392703	12.23132750303010
H	1.92682798329191	-1.82853196732550	8.63837671791132	C	3.26846568069020	6.37446661409028	10.30093348869019
H	0.94605537779989	-1.73110524296510	10.11443436560600	H	2.57738275124754	7.19442461506905	10.10878286073972
C	1.35369741292337	3.51961054435546	11.11700448718715	C	-2.53389787466561	6.63686159896264	9.05162631272772
H	1.24719967177626	4.54748980681886	10.76027149441018	C	-1.50400625116741	4.37648895791763	9.11193970770782
H	2.30147738103075	3.43662268217322	11.66660859765753	N	-3.66797711699262	7.29989224573432	9.30936926953497
H	0.54260810337153	3.33058203336699	11.83664963765613	C	-2.61057417066926	5.19044690552129	9.33700377208811
C	5.15800570863815	4.84184207827451	6.67877495261681	H	-1.56539633665308	3.30878233565523	9.32156857140914
H	5.80611215830126	4.93010706139458	7.56253999807324	Ru	-5.21477141770761	6.16566689931224	10.12216911172191
H	5.71826601182537	4.27082949506706	5.92641932364614	C	-3.75623166641919	8.67980123888501	8.97042747881717
C	4.83143078168157	6.22320148092896	6.20231941218944	O	-3.75335494759588	4.74373681346769	9.77972611079830
C	5.68479237825881	6.97773477393950	5.40227407738308	N	-6.77605515383179	7.49227095521381	10.55125420524752
H	6.61002096749007	6.53491709014757	5.03381503586391	N	-6.50956809529208	4.84708230106512	10.98296982547366
C	5.33946567482315	8.29302862624609	5.08831873186094	N	-4.67928157001597	6.56531777678873	12.04867409671567
H	5.99351524073978	8.90097232059329	4.46374850138984	N	-6.19418360597400	6.13646623550947	8.31724295236431
C	4.13926137062288	8.81138720446604	5.58386551648914	C	-6.26905149916007	8.36385296240153	11.64864667180122

C	-5.42127254698376	7.56824189951355	12.60787621837565	H	-3.49226961821966	9.74189185074954	12.12669458644964
C	-7.04760311907497	8.20859845515076	9.27226250813290	H	-2.84171057507949	8.23505579671709	11.43239821646917
C	-7.03936624197991	7.19865601756582	8.14986639540775	H	-1.90384375979439	9.73696404246290	11.35333004500575
C	-3.53126735081677	9.66292078605000	9.95699683423832	H	-4.74288705059991	10.68700849841299	6.40314877065207
C	-4.13403589668564	9.05467166590182	7.66089952482639	H	-4.09968562679469	7.02847531102256	6.89105016497619
C	-8.00859442255906	6.73288083661336	10.97104621378802	H	-5.15271194143574	8.14074432838270	5.98497906573341
C	-7.68172608791756	5.35200743591656	11.44878253739177	H	-3.39367039614471	8.24515328638025	5.81533298430791
C	-6.17031175872547	3.57382183950854	11.29257751595889	C	-5.32232763801127	7.86881294568625	13.96062871790224
C	-6.11724264655453	5.19882056381359	7.35026125812505	H	-8.48854493627901	8.18096889362091	6.90841910672046
C	-3.82975492548467	10.99827950348931	9.65507726888450	C	-7.72259169845162	6.36493967574584	5.99700976418059
C	-2.91916335094724	9.31680065374233	11.29040798538420	H	-9.46027678807648	5.04084451665216	12.61753846984959
C	-4.41845497668714	10.40148122261995	7.40757461270144	C	-8.18892928317680	3.28325587939130	12.56516773194641
C	-4.20510504266041	8.05661115405943	6.53472570191787	H	-6.67763608041607	1.74477936056521	12.29228583869090
H	-5.63843841987160	9.13199034832709	11.17646230091261	H	-6.75873175382231	4.50707917077605	5.41948023496749
H	-7.08978046088994	8.87587471158977	12.17267412612176	C	-4.62024030328610	12.82855300451733	8.09409922011347
H	-8.00071761201953	8.75867706497328	9.30358256551536	H	-8.84264243483805	2.67576383768940	13.19051891615122
H	-6.23515079118362	8.93252055005389	9.12028186139961	H	-8.31969905232728	6.45772884051947	5.09027772372593
C	-7.81689102327028	7.32850297501768	7.00676063494095	C	-3.82164402158823	5.86743753529907	12.82329571336802
H	-8.65627233582261	6.64370829662376	10.08706120842561	H	-5.93747582881735	8.66449982975286	14.38011801406218
H	-8.56931443088875	7.30430120865675	11.72274908851044	C	-4.42495667418224	7.15390336656866	14.76021233532261
C	-8.53482327587860	4.59802858147768	12.24963269835166	C	-3.66082293662719	6.14188768435065	14.17792015929630
C	-6.98848563034121	2.76482253939148	12.07027438287228	H	-3.26844826935592	5.07733399117245	12.31683235268468
H	-5.21178264876615	3.23758666088854	10.89852546442453	H	-4.33064207475711	7.38280667895572	15.82134651905867
C	-6.86066389961868	5.28343422071422	6.17659493278538	H	-2.95244338832467	5.55759985844580	14.76381542388287
H	-5.42629604913320	4.37894799068966	7.54324048811483	H	-5.52255411000051	12.91568299065231	7.47247351158227
H	-3.67993646660130	11.75658027519642	10.42839128872734	H	-4.77682951833427	13.40353265592232	9.01640550312002
C	-4.30092635287338	11.38677873580186	8.39613193144765	H	-3.79705490368721	13.30681711600206	7.53959728577814

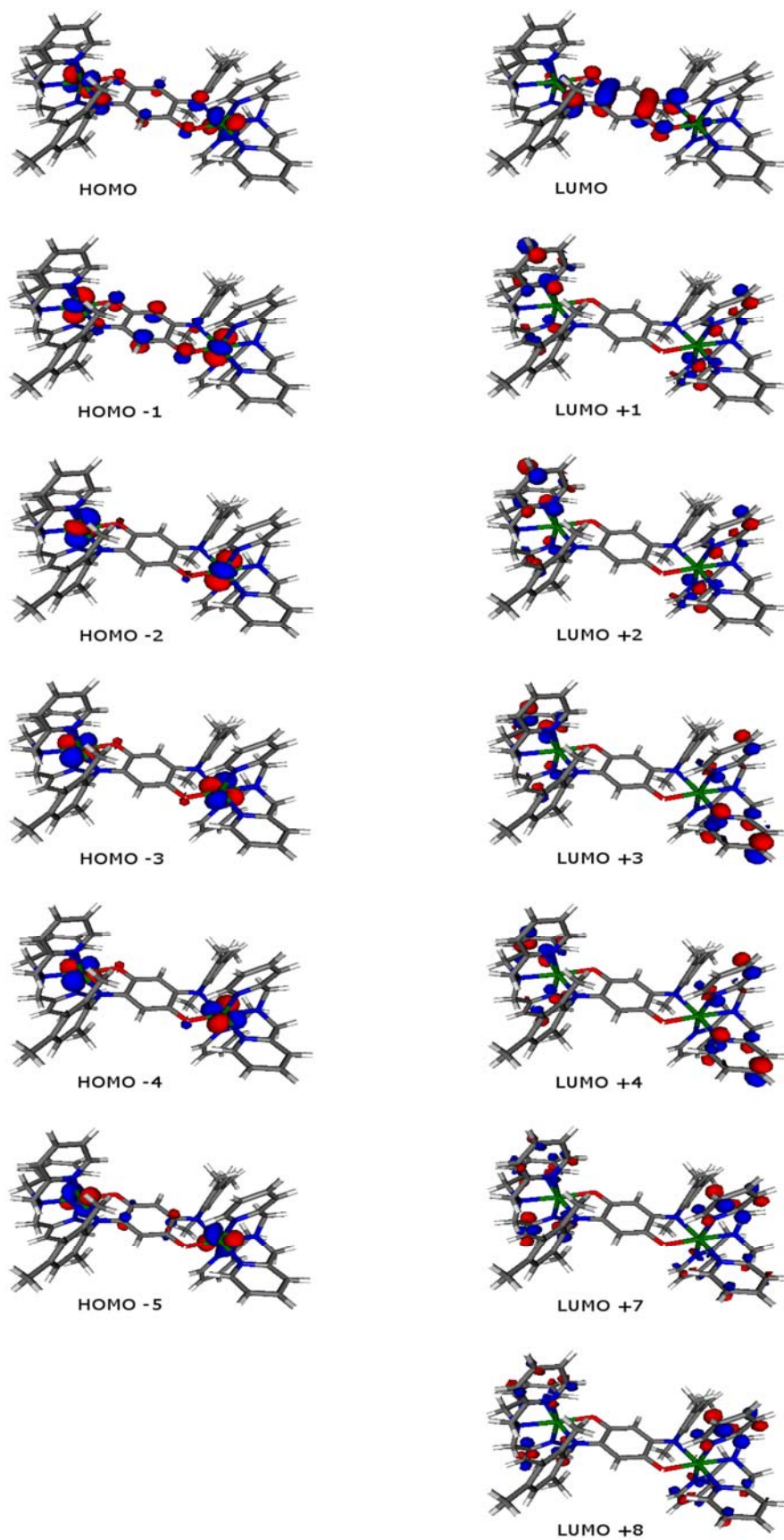


Figure S27: Contributing molecular orbitals of $[3^{\text{N(Mes)}}]^{2+}$ for the main absorption transitions.

Table S7 Atomic coordinates of the optimised geometry of $[3^{N(\text{Mes})}]^{3+}$.

Ru	2.32504760349012	5.42208295314722	7.50283247127081	C	4.13548082346873	8.82709667057750	5.57160213328713
O	0.88366227280645	6.81826076919952	7.82427720574826	H	3.83051437196297	9.84736127955211	5.34459989695811
N	0.80117215880324	4.26904516463790	8.30292274751513	C	3.30295233551196	8.01766357718281	6.33364712522364
N	3.90095886533174	4.09892255671930	7.10243046270121	H	2.33904777670804	8.35744310579420	6.71045029517889
N	3.64008113131469	6.74646667268654	6.64499735893654	C	3.41575018964476	3.20392330179860	6.00989281278259
N	1.81539962062997	4.98176843558548	5.56351674496270	H	2.79445540126184	2.42931477788041	6.48376150966092
N	3.28849304024217	5.45790343266680	9.32546865491683	H	4.25127308283498	2.70007991347909	5.50437580122538
C	-0.25218280348986	6.37614806823862	8.29539265103358	C	2.56918933075016	3.97513424354360	5.03275566555069
C	-1.34425018432080	7.19987464810801	8.53995066613582	C	2.48415248591535	3.65282598639110	3.68392485348878
H	-1.27784506428711	8.26746484354569	8.33369992854224	H	3.10519641570220	2.85280490823241	3.28268960215979
C	-0.32469795701384	4.93440467202768	8.57929875171761	C	1.59583704381124	4.35575220720511	2.86589700827917
C	0.88969349992032	2.88381895633246	8.64036982524071	H	1.51447126684750	4.11144744075530	1.80724590026042
C	0.67159732720206	1.90690796624152	7.64770109359158	C	0.82423637902321	5.37662055209333	3.42134011925131
C	0.96635329754631	0.57151283597583	7.95195189400274	H	0.12397734651740	5.95248484647283	2.81842618928480
H	0.81929021718283	-0.18571680896201	7.17767112901799	C	0.96697418954413	5.67037642485535	4.77320963417662
C	1.42840972449921	0.18258034145926	9.21380438720129	H	0.40672675236417	6.46853026477265	5.25817234446568
C	1.53699208215576	1.16539911838995	10.20590035045490	C	4.16913436574759	3.39031955652279	8.38938937825483
H	1.85127795348897	0.87825555171561	11.21260085866825	H	5.12825434672591	2.85324512826290	8.36325670996961
C	1.25405374391831	2.51235192352493	9.95415699884381	H	3.36680617984213	2.65466007144705	8.53753336240420
C	0.06065892385269	2.25023248506197	6.31327966277200	C	4.13948874190894	4.40487044427999	9.50479368818381
H	0.64883908302247	1.84561675090968	5.47767434238687	C	4.90226244399422	4.28963100554904	10.65957448681596
H	-0.04868210517706	3.32971236907615	6.17522671121847	H	5.58074567323183	3.44486833824243	10.77286594920817
H	-0.94196326150374	1.80278807522874	6.24144496717783	C	4.78609576579450	5.26046103133356	11.65829018165703
C	1.74559089400200	-1.25856255253551	9.51640236174948	H	5.37313290797045	5.18016174146363	12.57260492526492
H	2.63975105595210	-1.34492026618270	10.14937594063824	C	3.91866550246658	6.33433610036450	11.46169822583536
H	1.91295266554136	-1.83169888614402	8.59515304475106	H	3.80300292212329	7.11629631236170	12.21044253800432
H	0.91518319915656	-1.73615950929261	10.06027934951295	C	3.18768943954961	6.40442750105648	10.28021423878608
C	1.30394152719301	3.50391005576671	11.08684686527094	H	2.49435686656171	7.21875928905512	10.07388334961637
H	1.27999427328863	4.53963313536936	10.73835508683337	C	-2.52763808812979	6.63933575842258	9.06849269128064
H	2.20290337831272	3.36122670494821	11.70104673052298	C	-1.50818792652169	4.37385945982118	9.10763759232100
H	0.43418791358532	3.36021086545698	11.74624738835213	N	-3.65323964860851	7.30493262307987	9.34552923352101
C	5.13619350295196	4.85852414504193	6.68801793777311	C	-2.60013626105200	5.19762414603591	9.35260703552648
H	5.77987702225147	4.95215456734069	7.57439592907133	H	-1.57456798244570	3.30627680710531	9.31392308737425
H	5.69968345467822	4.28261623004849	5.94286626176736	Ru	-5.17637313727031	6.15268529500694	10.14758186300738
C	4.81625352764413	6.23746369616124	6.20357633458217	C	-3.74124555810659	8.69040440568249	9.00887360058901
C	5.68423899064598	6.99079198808859	5.41885190371536	O	-3.73567419438943	4.75579685346871	9.82464471308645
H	6.61620517083694	6.54935140027776	5.06734739620935	N	-6.75141150924564	7.47632677606840	10.54950339924007
C	5.34204775814732	8.30511154173723	5.09926646467650	N	-6.49038705249353	4.82906272253599	11.00834384825537
H	6.00735525467885	8.91194269451969	4.48564939418929	N	-4.66387614572242	6.59362231013322	12.08607012755154
				N	-6.14215714560496	6.11633524901887	8.32623856957888

C	-6.26457114842805	8.37159674469351	11.64111336182037	C	-4.27779966837028	11.39248661291810	8.43735859571328
C	-5.41693183034294	7.60048959675890	12.61742654360571	H	-3.49497799424901	9.72513315930146	12.17159823672655
C	-7.02138065729552	8.18446472993191	9.26265877546081	H	-2.79670673110287	8.24276206973775	11.47127018354681
C	-6.99301131714911	7.16959720860871	8.14747625396400	H	-1.90518944352064	9.77084843182075	11.40603082959188
C	-3.52022574682515	9.66669314567959	10.00147641204101	H	-4.70512788067818	10.69808429476728	6.43912450744801
C	-4.10749064809237	9.06275433164166	7.69585119169726	H	-4.13792352776200	7.03576407485600	6.91115733496758
C	-7.98621977602290	6.71709155981301	10.96593895102985	H	-5.05987558000977	8.21608612662184	5.94988263463415
C	-7.66562847388533	5.33871724295041	11.45153115309417	H	-3.29116217940311	8.21393085850729	5.90262017489726
C	-6.15261074616538	3.55833728911359	11.32085535697217	C	-5.33044944342788	7.92295474358043	13.96613310203269
C	-6.04247753941437	5.16961710325914	7.37158510218952	H	-8.43493141519639	8.1298632397852	6.88044189237869
C	-3.81393669764149	11.00256378042786	9.69820072231959	C	-7.64149351333804	6.31391882513562	5.99456621593796
C	-2.90734556769258	9.32230096143319	11.33473935820072	H	-9.46318498292281	5.02852905473414	12.59205066667342
C	-4.38926137139137	10.41012104465650	7.44509335193823	C	-8.18892640600543	3.27283108750899	12.56026506374756
C	-4.16043466712428	8.07162800942822	6.56296577398325	H	-6.67791261173730	1.73025154957065	12.31417584061600
H	-5.64384323146915	9.14592373212929	11.16604800294937	H	-6.65941521047683	4.45776367117065	5.44177405155810
H	-7.09931369956222	8.87575814586494	12.14760369940776	C	-4.59412661094054	12.83403969137451	8.13584175606264
H	-7.98057524512066	8.72134110540915	9.28996704248422	H	-8.85272146852003	2.66694824118177	13.17643612303360
H	-6.21965363741804	8.92051762228943	9.11324935539532	H	-8.22919005803974	6.39441014646186	5.08068542777092
C	-7.75663847434770	7.28488803778438	6.99327237102238	C	-3.81471521130399	5.90509612381457	12.87555937758949
H	-8.63073103004451	6.62253756004627	10.08025740039667	H	-5.95099783312687	8.72309933630554	14.36787257575808
H	-8.54908717646530	7.29372977919050	11.71100182393092	C	-4.44142165170083	7.21997797230135	14.78337272496888
C	-8.53193480602876	4.58661340824728	12.23924294046116	C	-3.67053665791249	6.19892746287889	14.22727835442162
C	-6.98336269089649	2.75014923202420	12.08616284322888	H	-3.25505041324826	5.10672989629353	12.39027984218158
H	-5.18940595828255	3.21808221392427	10.94272074372150	H	-4.35904332406269	7.46443777500754	15.84190257223251
C	-6.77425956626742	5.23977680828759	6.1906077570309	H	-2.96966101228407	5.62310440064354	14.82953129601296
H	-5.34927712653269	4.35506669222315	7.57748825430259	H	-5.49091350998683	12.92157328958123	7.50674190044486
H	-3.66455928983761	11.75937928369976	10.47244113766945	H	-4.75673480546900	13.40756690577816	9.05769385336132
				H	-3.76539719213644	13.31029848562764	7.58824722646197

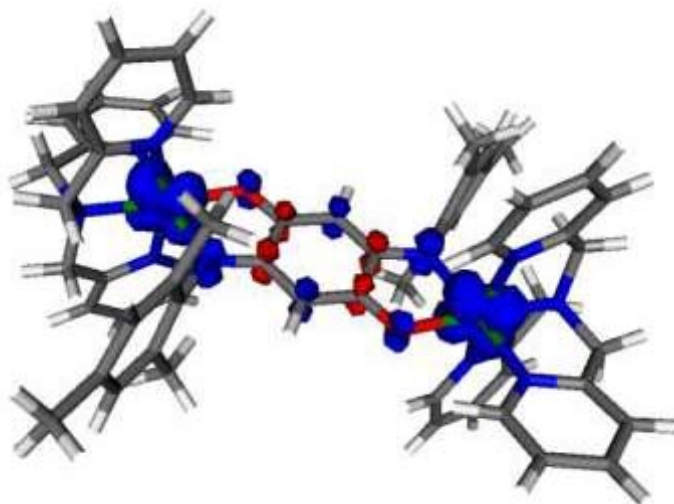


Figure S28: Spin-density distribution of $[3^{N(\text{Mes})}]^{3+}$.

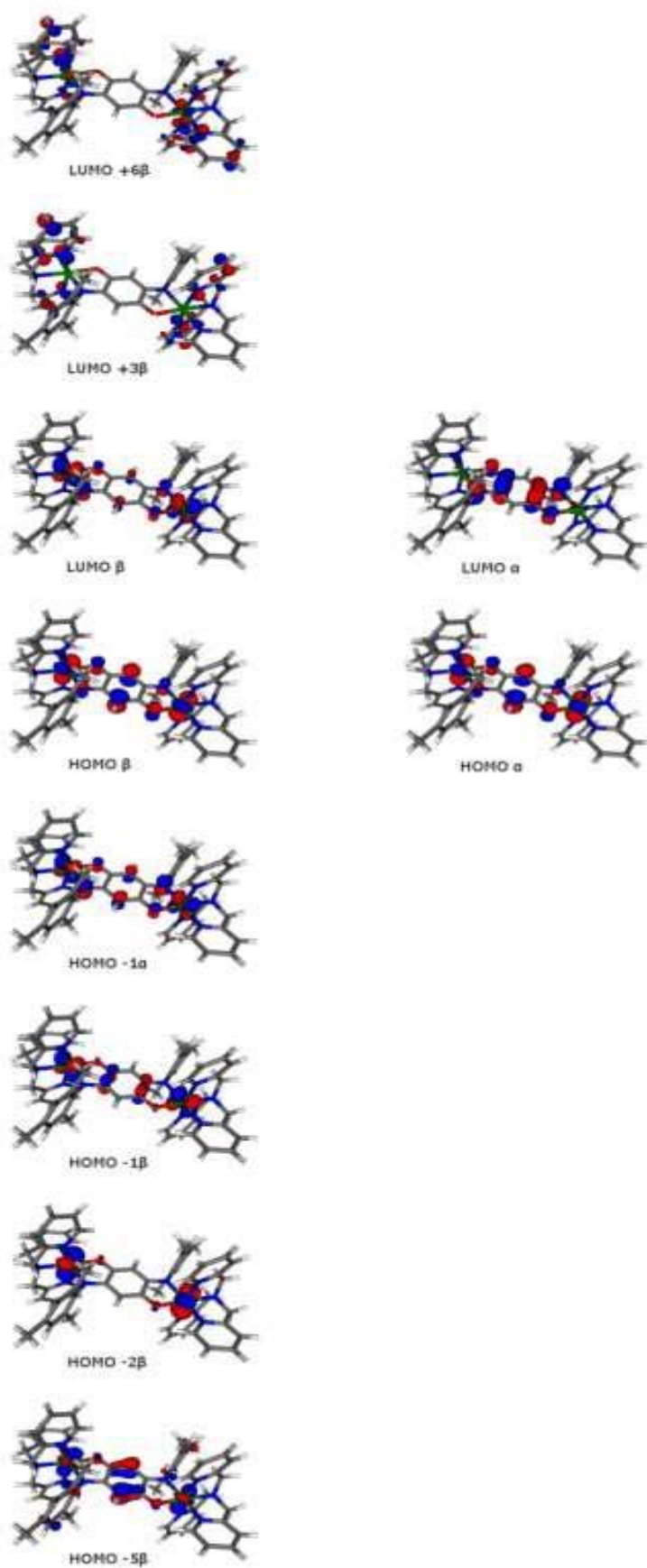


Figure S29: Contributing molecular orbitals of $[3^{\text{N(Mes)}}]^{3+}$ for the main absorption transitions.

Table S8 Atomic coordinates of the optimised geometry of $[3^{N(\text{Mes})}]^{4+}$

Ru	2.27952361372964	5.39815359905460	7.47118365257209	H	3.87579599399773	9.89560365523061	5.51523774959338	C	3.30270311484745	8.03109725715474	6.40623760573815
O	0.87781881313715	6.79029100585174	7.80216378187183	H	2.33894461749222	8.37075419690829	6.78248239221102	C	3.37184628678455	3.18402121900584	5.93466975976065
N	0.78535578609927	4.25978094454150	8.28060721837747	H	2.74802665736981	2.40123670753943	6.39097207029570	H	4.21148375557430	2.68730588961752	5.43093290432954
N	3.85322793755547	4.06372178244457	7.04082678714951	C	2.53809314683934	3.97557946593759	4.96740380879907	C	2.43758163566387	3.66654025748628	3.61686071138056
N	3.61844775260288	6.74198887179881	6.65993815360987	H	3.03762423671525	2.85613337540169	3.20548491816541	H	3.03762423671525	2.85613337540169	3.20548491816541
N	1.80968085711287	4.99458202440437	5.50588169011821	C	1.56151711708993	4.39745027419626	2.81180208763060	H	1.46844330139312	4.16366694861444	1.75188276932313
N	3.30595143034529	5.39828537983921	9.26780864816591	C	0.81737627503332	5.43315802776583	3.37775473263840	H	0.12829061818690	6.03034417009543	2.78333088714812
C	-0.26094363733300	6.36507231350337	8.28575092625581	C	0.97490299989461	5.71542230762595	4.72901836374662	H	0.43924057217559	6.52641912630064	5.21939376173743
C	-1.33760666611496	7.20615511814452	8.53539693897573	H	4.13176273065134	3.32469543286770	8.30908672786397	C	4.13176273065134	3.32469543286770	8.30908672786397
H	-1.26435702255446	8.27271993938778	8.32823913499728	H	5.08695614237677	2.78484459813987	8.25268103512576	H	5.08695614237677	2.78484459813987	8.25268103512576
C	-0.33664631912686	4.92909039503871	8.57853038890010	H	3.33035197117293	2.58796732258264	8.45459251183933	H	3.33035197117293	2.58796732258264	8.45459251183933
C	0.89402275844580	2.88259882174652	8.65096587517693	C	4.12735384493544	4.32270143016474	9.43609099597480	C	4.12735384493544	4.32270143016474	9.43609099597480
C	0.71889393225492	1.88750160182339	7.66787678654572	C	4.88971397072131	4.18429741473911	10.58827895126897	C	4.88971397072131	4.18429741473911	10.58827895126897
C	1.04442440968636	0.56788284417049	8.00118263292801	H	5.54719524967335	3.32297771539472	10.69682631515995	H	5.54719524967335	3.32297771539472	10.69682631515995
H	0.92754816995216	-0.20722679500847	7.24023969088709	C	4.80183788153420	5.15677186566039	11.58713722104312	C	4.80183788153420	5.15677186566039	11.58713722104312
C	1.49987256116718	0.21622746459178	9.27730226716096	H	5.38934561049409	5.05878648964273	12.49929587798161	C	3.96664583771304	6.25711782768646	11.39611162294826
C	1.56494034070618	1.21791294409586	10.25421205957453	C	3.96664583771304	6.25711782768646	11.39611162294826	H	3.87738375506315	7.04163081808716	12.14531248167310
H	1.87688320312673	0.95837102148688	11.26860411681708	H	3.87738375506315	7.04163081808716	12.14531248167310	C	3.23482697314455	6.35180115381762	10.21827082345576
C	1.24618006174920	2.55178110502364	9.97827788337044	C	3.23482697314455	6.35180115381762	10.21827082345576	H	2.56778744670460	7.18826419099365	10.01540576177142
C	0.10130709272455	2.19511607642386	6.32870596429602	H	2.56778744670460	7.18826419099365	10.01540576177142	C	-2.51039631339812	6.64602162138138	9.08315377894165
H	0.61958008756269	1.67477940859918	5.51213694038273	C	-1.50855250224548	4.36933904257703	9.12852132928706	C	-1.50855250224548	4.36933904257703	9.12852132928706
H	0.08098554701053	3.26743741068272	6.11283865784373	N	-3.63353234090536	7.31473950791974	9.37821527603549	N	-3.63353234090536	7.31473950791974	9.37821527603549
H	-0.94219962401965	1.84506810789330	6.31818438896410	C	-2.58521351282601	5.21045515673973	9.37819090309673	C	-2.58521351282601	5.21045515673973	9.37819090309673
C	1.85684706094549	-1.20767435281947	9.60886799074986	H	-1.58154375935831	3.30297697681830	9.33674153941716	H	-1.58154375935831	3.30297697681830	9.33674153941716
H	2.74693541091002	-1.25479078132549	10.25125340777285	Ru	-5.12986224176325	6.17537953918973	10.18240765580288	C	-3.74175901120282	8.69192272476189	9.00728278765089
H	2.04755750689290	-1.7928000280353	8.70016289360609	C	-3.74175901120282	8.69192272476189	9.00728278765089	O	-3.72322610309019	4.78557737848819	9.86381353418604
H	1.03498560256346	-1.69640663586625	10.15560170957875	O	-3.72322610309019	4.78557737848819	9.86381353418604	N	-6.71249807528901	7.50554622844416	10.59271609154869
C	1.26061673027072	3.56291503178439	11.09381359779079	N	-6.71249807528901	7.50554622844416	10.59271609154869	N	-6.46950351055158	4.83071261538035	10.99158490507757
H	1.25237970278686	4.59564090419976	10.73491898272567	N	-6.46950351055158	4.83071261538035	10.99158490507757	N	-4.67765684698672	6.58905040450768	12.14911555403930
H	2.13709718392185	3.42450324267461	11.73983379665152	N	-4.67765684698672	6.58905040450768	12.14911555403930	N	-6.14132579068338	6.16386483003836	8.37700036471633
H	0.36930144368896	3.43201483079521	11.72646845035124	N	-6.14132579068338	6.16386483003836	8.37700036471633	C	-6.24572640408300	8.39012788679342	11.70121590437262
C	5.09191338060839	4.82839486837541	6.63473547850406	C	-6.24572640408300	8.39012788679342	11.70121590437262	C	-5.41668947373654	7.60533707832044	12.67809882703112
H	5.75832125094659	4.86934985498000	7.50786016154988	C	-5.41668947373654	7.60533707832044	12.67809882703112				
H	5.62687337947344	4.27562165608882	5.85263234652787								
C	4.79145117259161	6.23251655244186	6.21609231816853								
C	5.68430041255830	7.00858167861236	5.48253223985677								
H	6.61553256250853	6.57002825727807	5.12626502332895								
C	5.36631861568504	8.34067744721754	5.22163881998495								
H	6.05117240215630	8.96523349265386	4.64882733751574								
C	4.16033016336721	8.86099549290766	5.69819882338025								

C	-6.98142569837361	8.23929512863740	9.31952851947526	H	-1.93733305498062	9.75901585721731	11.36483576846326
C	-6.96201442482195	7.23776386885193	8.19569797120668	H	-4.70685247043372	10.61417078531101	6.38133378404610
C	-3.57763598578614	9.68719813492778	9.99175681374077	H	-4.10573244232318	6.97846428737000	6.91862747378433
C	-4.08303819702725	9.02207064619627	7.67667967556779	H	-4.93446874018318	8.16769108783048	5.88752191307662
C	-7.95199771160670	6.73699941216241	10.98890370486497	H	-3.16823967792327	8.12371152836504	5.95725523274782
C	-7.64801825914825	5.33833653626250	11.42301993391320	C	-5.33018237786515	7.91858044974084	14.02867909714923
C	-6.14900579569657	3.54569928354990	11.25916162716612	H	-8.36716827219486	8.23217271034058	6.91439676603339
C	-6.05811942776141	5.20701615522242	7.43086775269532	C	-7.60899359638809	6.39646202309373	6.03903193845217
C	-3.90311224837833	11.00637688859325	9.65543598075926	H	-9.47718950294877	5.00193331097503	12.50452752767779
C	-2.97102728803341	9.38223594354884	11.33666108526642	C	-8.21843471960199	3.23677679190080	12.43378354507220
C	-4.40302697668976	10.35482641972372	7.39822550063826	H	-6.71833344864716	1.68675108673301	12.16423581448996
C	-4.08208129242124	8.01138940771632	6.56088625028217	H	-6.67658646531266	4.51045996285883	5.49825438002928
H	-5.62197841063188	9.17526500408933	11.24890639434301	C	-4.70872976864591	12.77920030072510	8.04085146966231
H	-7.09260117050823	8.88347244527661	12.19605860579076	H	-8.90354433574511	2.61419451490332	13.00842377383334
H	-7.93937838091221	8.77525717264621	9.36453732822640	H	-8.18508836123691	6.49137876115503	5.11929324949949
H	-6.18174313537334	8.97891085463367	9.17930811635423	C	-3.84522873605287	5.87570452221726	12.93534326085204
C	-7.71022871604990	7.37186833267838	7.03374615570386	H	-5.93871324911352	8.72653095011432	14.43234868714564
H	-8.60613279808268	6.68444701324962	10.10715080276174	C	-4.45708449988853	7.19509439997722	14.84362695968372
H	-8.50029315362249	7.29250778358146	11.75951805574704	C	-3.70144564545621	6.16258614444352	14.28713947348951
C	-8.54159696962458	4.56474150567406	12.15817898978329	H	-3.29968774597744	5.06716819082977	12.45188703811240
C	-7.00691081575813	2.71813577347164	11.96973124544787	H	-4.37491714217914	7.43226408736098	15.90368665409573
H	-5.18106315966524	3.20745722912318	10.89252187758683	H	-3.01359004705328	5.57155258739009	14.88907400212796
C	-6.77555245460118	5.29725813281019	6.24384901617261	H	-5.61478444134420	12.82357880204867	7.42071411210564
H	-5.39311428847806	4.37162173306091	7.64447642042443	H	-4.87518440058896	13.37284460655818	8.94871789904414
H	-3.79385501632546	11.78167633997905	10.41733046716990	H	-3.89980438926722	13.25997837367716	7.46853787647669
C	-4.34890468871050	11.35686419477620	8.37594196170136				
H	-3.51296839401126	9.88141116908603	12.15118754772443				
H	-2.92733780736686	8.30870655758252	11.54283817954291				

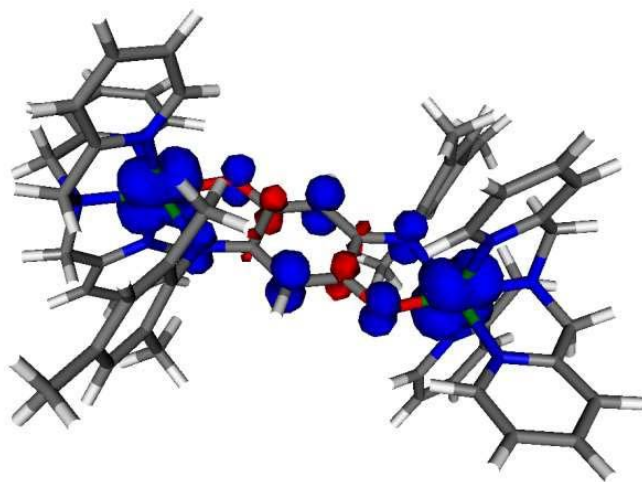


Figure S30: Spin-density distribution of $[3^{N(\text{Mes})}]^{4+}$.

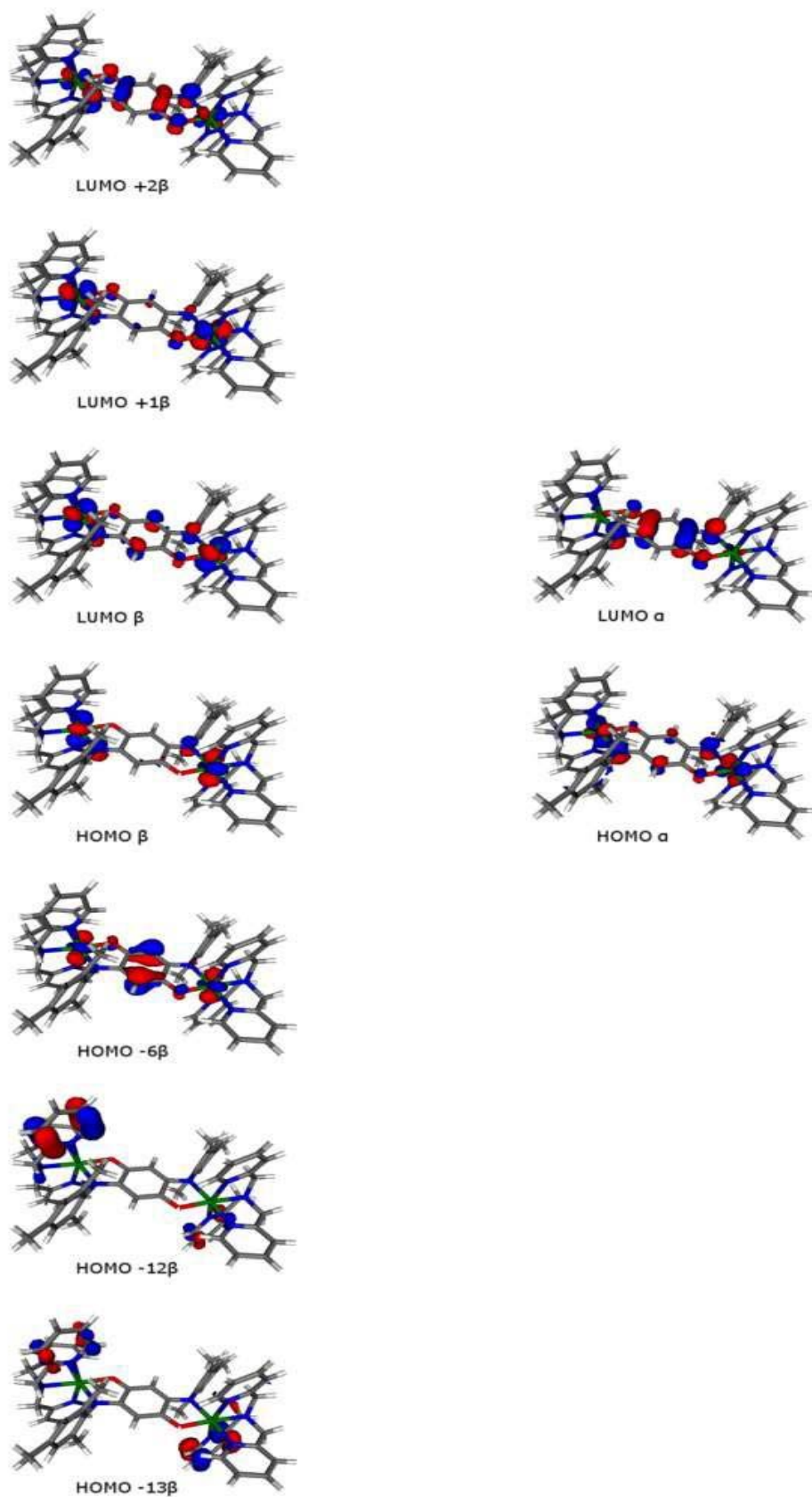


Figure S31: Contributing molecular orbitals of $[3^{N(\text{Mes})}]^{4+}$ for the main absorption transitions.

Table S9 Atomic coordinates of the optimised geometry of $[3^{\text{N(Mes)}}]^+$.

Ru	2.37086702164298	5.41094031803325	7.52043660256048	H	3.77586803032406	9.74355409434714	5.15190339947385	C	3.30560048367552	7.96994681283444	6.26169862020016
O	0.90778748051922	6.85482056990603	7.84606908938810	H	2.34722544488984	8.31397067377454	6.65060295925807				
N	0.83445269715361	4.25874134236627	8.31799696903099	C	3.45757456224677	3.22358905160744	6.00865676374386				
N	3.94269400556012	4.08469953186857	7.12429275940012	H	2.81229024804138	2.45818417559208	6.46420031609149				
N	3.66116225959979	6.71725223986245	6.64246425945739	H	4.28712051760914	2.71077782318642	5.49742949635250				
N	1.86774343353039	5.02070877850108	5.59323027411253	C	2.63933986418689	4.03496425548029	5.03557645789117				
N	3.30489070411954	5.44435608350516	9.34415085190151	C	2.60277728057751	3.77632017881728	3.67143953810875				
C	-0.24053215508339	6.37183246463649	8.30130515574227	H	3.24019676164926	2.99533182411200	3.25664390996994				
C	-1.36043672311686	7.17623305318884	8.53595814570134	C	1.74217344234971	4.51480753000236	2.85092706946474				
H	-1.30314222731454	8.24605210653191	8.32757964157275	H	1.69812407955880	4.31893687919951	1.77984907312937				
C	-0.31067789049630	4.94986947647064	8.57602150140546	C	0.95143260837716	5.51077931565300	3.42979370399193				
C	0.90989772698578	2.88892823531971	8.65957240321022	H	0.26985332013680	6.11259778452053	2.82922035371054				
C	0.70830309515402	1.89826573042622	7.67172701309962	C	1.04690564342246	5.74250495794846	4.79805701496058				
C	1.00500897384909	0.56172297941466	7.97449522152635	H	0.46920116656301	6.51253046118265	5.30912325232021				
H	0.87857075043962	-0.19549520520825	7.19510412387265	C	4.18400768839979	3.36980974400345	8.40785710351053				
C	1.44642793511711	0.17008846094920	9.24297198675911	H	5.14202257639274	2.82520662539766	8.40564367596076				
C	1.53266923499927	1.15388281145604	10.23762088279816	H	3.37092821811902	2.64224045298512	8.53818662333064				
H	1.83457953588038	0.86620036523466	11.24908753092050	C	4.13930228654835	4.37660385408437	9.53508437261322				
C	1.24851567283901	2.49946141078643	9.97924674321806	C	4.87504706431262	4.23654737569449	10.70419053398814				
C	0.12717766737809	2.25052131918188	6.32586484407039	H	5.53767354817981	3.37920875450014	10.82210928869909				
H	0.73463832242071	1.85944357898382	5.49645248940041	C	4.75165873666552	5.19560122878834	11.71645358555776				
H	0.03266018005738	3.33433401467691	6.21036782963164	H	5.31591026080287	5.09412706734740	12.64322885087348				
H	-0.87504086804918	1.80632570259876	6.22358963692798	C	3.90228390585523	6.28303728334142	11.51236815996928				
C	1.77148207394366	-1.27065976959146	9.54787046516186	H	3.77744420220476	7.05505354124301	12.27092436090550				
H	2.70923513413825	-1.35785004371287	10.11592598226525	C	3.19950737086027	6.37661343460108	10.31416770568293				
H	1.87103542967599	-1.85980631044771	8.62635784017615	H	2.51390231825402	7.19591497985802	10.09903435911421				
H	0.98105067704029	-1.73654436180429	10.15794201927414	C	-2.55501114298515	6.62018326943055	9.04525417427057				
C	1.28093079752541	3.49789433970123	11.10586820295809	C	-1.50609721529196	4.39347643664874	9.08296965781760				
H	1.25582972861013	4.52397372726494	10.72909602373135	N	-3.69978993625437	7.31124637704737	9.30522000340764				
H	2.17513014194368	3.36995556796138	11.73148083605775	C	-2.62542843171981	5.19804739170595	9.31907021752260				
H	0.40186345873049	3.36180812063212	11.75502245465174	H	-1.56312395374691	3.32369886122866	9.29167645032079				
C	5.17741763057949	4.85419614898192	6.73767984938807	Ru	-5.22654997958815	6.16366034246459	10.12530036141888				
H	5.76535461069913	5.00100542160965	7.65561867694364	C	-3.77450896555382	8.68167337478833	8.96582669486727				
H	5.79667814351307	4.26591202716788	6.04633320708449	O	-3.77376443389416	4.71541305685645	9.77461918400646				
C	4.83643665869646	6.20120140538295	6.18133424251196	N	-6.78580340042266	7.49501026861741	10.55406115413635				
C	5.66493146177629	6.91529295450047	5.32321220136292	N	-6.50528540800415	4.86027893964443	11.02452338683151				
H	6.58729212281574	6.45873404301898	4.96324404497430	N	-4.68593619992582	6.54841169481776	12.04322664921320				
C	5.30254113970742	8.20969503892463	4.93826819625759	N	-6.19442150777035	6.13752111442829	8.31877597719763				
H	5.93848851027168	8.78678397990491	4.26752831055096	C	-6.27402254296880	8.35378218296752	11.65960146638256				
C	4.10276955064365	8.74087819673195	5.42624393724474	C	-5.44123541696300	7.53791328930444	12.61636617839631				

C	-7.05088724319950	8.21144714947164	9.27617159461056	H	-1.93831216340520	9.75627343235133	11.37078798318501
C	-7.03370184420980	7.20489703614328	8.14812703635140	H	-4.73496435877156	10.71061294622400	6.39432224486401
C	-3.54628403126443	9.67126443627469	9.94943777120371	H	-4.09728615189782	7.05025656049253	6.89950562404976
C	-4.13738736630112	9.07263142151576	7.65363171129670	H	-5.15381645417559	8.15066430968761	5.97784569697455
C	-8.01536427978458	6.73020697195138	10.96563617183603	H	-3.39321202779884	8.25837277922893	5.80946492582155
C	-7.66910428661009	5.37971578127172	11.51018907693611	C	-5.37658718569869	7.79543950070803	13.97967775945032
C	-6.14749522926992	3.60499960055267	11.39478209713634	H	-8.46475490644746	8.20121658876871	6.89666534683889
C	-6.10970894983665	5.20747973311683	7.34471410629078	C	-7.69757779862166	6.38699312127731	5.98149542436226
C	-3.83905159862654	11.00952099680786	9.65192345040870	H	-9.39705236213690	5.12551183147219	12.76125779890103
C	-2.94329868030235	9.31506086478915	11.28462933073700	C	-8.11943184538948	3.36921252246697	12.75588344309099
C	-4.41551172293159	10.42077179428433	7.39978562012923	H	-6.60339327183710	1.82991331247729	12.50815100446866
C	-4.20588408615516	8.07323022709933	6.52884948027008	H	-6.73219295633601	4.53094945800656	5.40092869354955
H	-5.63351223393614	9.11518629597655	11.19071008968816	C	-4.61531296872301	12.84839970763302	8.09142468154408
H	-7.09035479744401	8.87144303015735	12.18699138594500	H	-8.74461685006781	2.79255940774333	13.43703546268767
H	-8.00625467156337	8.76017087818533	9.29859272750185	H	-8.28452602806780	6.48819665118419	5.06889331118270
H	-6.23756136651549	8.93560542890120	9.12919941466905	C	-3.85367971975580	5.82155961432756	12.82177985209025
C	-7.79818960826962	7.34476417091340	6.99754665271493	H	-6.00150487257586	8.57959423845025	14.40731725239541
H	-8.62457766386772	6.58943516098402	10.06071935286342	C	-4.50410682037837	7.05177046253543	14.78277895616301
H	-8.61597080922599	7.31928625295773	11.67257338415533	C	-3.73029743691321	6.05178276966713	14.18802509703792
C	-8.48376733087227	4.66629426990648	12.38202919369638	H	-3.29032518107631	5.04892557761742	12.29874024485838
C	-6.93142290050593	2.83475857213723	12.24325418756478	H	-4.43803130802256	7.24674460528241	15.85288010098672
H	-5.19802223225168	3.25855705751546	10.98663630331454	H	-3.04054278867568	5.44594866246034	14.77508085911846
C	-6.84061206725592	5.30154344670614	6.16346152748873	H	-5.52274024781047	12.94164083111511	7.47751177415576
H	-5.41864291331063	4.38846954724937	7.54273571272258	H	-4.76236998825463	13.42441431672891	9.01500592079448
H	-3.69226138220226	11.76602518312341	10.42845551442330	H	-3.79534056474770	13.32494436757215	7.53021777328228
C	-4.30144779414766	11.40414532437207	8.39152260117190				
H	-3.53578895275274	9.70652923493273	12.12465459083526				
H	-2.85005052741223	8.23083185025362	11.39719709921927				

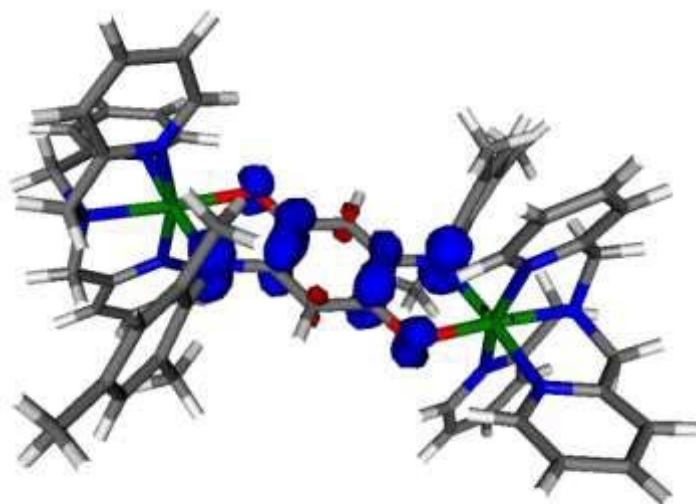


Figure S32: Spin-density distribution of $[3^{N(\text{Mes})}]^+$.

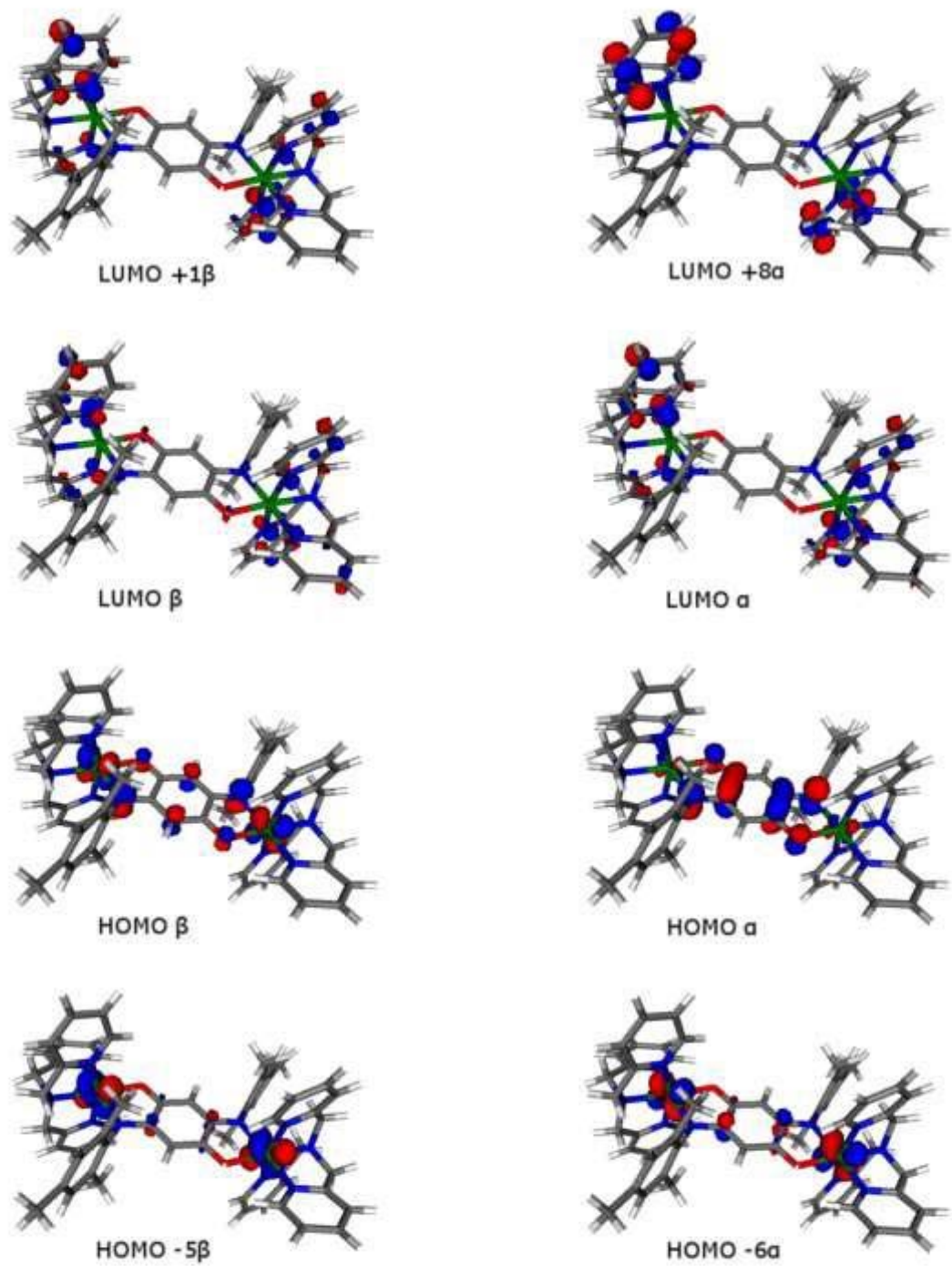


Figure S33: Contributing molecular orbitals of $[3^{N(\text{Mes})}]^+$ for the main absorption transitions.

Table S10 Atomic coordinates of the optimised geometry of $[2^{\text{NH,N(Mes)}}]^{2+}$.

Ru	2.43446	5.53513	7.49051	C	1.13318	5.72811	4.73202	C	-7.57420	5.23813	11.47516
O	0.93138	6.94272	7.78036	H	0.58705	6.55973	5.17605	C	-6.00416	3.50901	11.34760
N	0.92963	4.39602	8.27620	C	4.17074	3.52053	8.51750	C	-5.99309	5.10193	7.38131
N	3.96593	4.16346	7.18481	H	5.10454	2.93856	8.55445	C	-3.89189	10.98223	9.68666
N	3.77339	6.79430	6.60878	H	3.33340	2.82723	8.67876	C	-2.92931	9.32569	11.31733
N	1.93773	5.04541	5.57362	C	4.15035	4.58963	9.58408	C	-4.46055	10.37444	7.43703
N	3.32695	5.65111	9.33164	C	4.88824	4.51051	10.75773	C	-4.16051	8.04221	6.55549
C	-0.18163	6.47140	8.27235	H	5.54886	3.65954	10.92214	H	-5.65604	9.07488	11.18977
C	-1.30580	7.25291	8.52104	C	4.76987	5.52666	11.71273	H	-7.10365	8.77101	12.17771
H	-1.28367	8.32042	8.30194	H	5.33831	5.47786	12.64116	H	-7.98814	8.61814	9.29938
C	-0.21139	5.01935	8.56630	C	3.92023	6.60140	11.45208	H	-6.22775	8.84705	9.12867
C	5.22619	4.86971	6.75547	H	3.79862	7.41467	12.16644	C	-7.73669	7.19033	7.00667
H	5.85581	4.98846	7.64914	C	3.21853	6.63333	10.24962	H	-8.57436	6.47438	10.08197
H	5.78999	4.24859	6.04668	H	2.54441	7.44863	9.98931	H	-8.53668	7.16063	11.70934
C	4.94379	6.23392	6.19971	C	-2.46864	6.65805	9.05464	C	-8.40321	4.46841	12.28610
C	5.83444	6.92115	5.38029	C	-1.37874	4.42689	9.09726	C	-6.79667	2.68488	12.13605
H	6.75577	6.43573	5.05860	N	-3.61497	7.29135	9.32965	H	-5.03429	3.19909	10.95975
C	5.53173	8.22560	4.98545	C	-2.50256	5.20918	9.34361	C	-6.72515	5.16287	6.19907
H	6.21594	8.78035	4.34391	H	-1.40681	3.35542	9.30586	H	-5.28486	4.29997	7.58660
C	4.33518	8.80193	5.42233	Ru	-5.12975	6.11582	10.14904	H	-3.76618	11.74229	10.46253
H	4.05769	9.81604	5.13766	C	-3.74433	8.66931	8.99489	C	-4.37561	11.35964	8.42951
C	3.47830	8.05765	6.22297	O	-3.62492	4.73464	9.81147	H	-3.52977	9.70679	12.15574
H	2.52157	8.44185	6.57569	N	-6.73348	7.39597	10.56132	H	-2.79431	8.24789	11.44560
C	3.45917	3.23443	6.13080	N	-6.38467	4.76560	11.01881	H	-1.93945	9.80072	11.39289
H	2.78882	2.51942	6.62941	N	-4.61432	6.54414	12.07466	H	-4.79435	10.65253	6.43363
H	4.27683	2.66315	5.66659	N	-6.10097	6.04397	8.34126	H	-4.13868	7.01059	6.91652
C	2.66411	3.99138	5.09663	C	-6.26281	8.28638	11.65959	H	-5.05151	8.17772	5.92789
C	2.60864	3.62337	3.75770	C	-5.39435	7.52235	12.62602	H	-3.27949	8.18231	5.90993
H	3.21410	2.78961	3.40298	C	-7.01787	8.09832	9.27759	C	-5.31543	7.82997	13.97856
C	1.77177	4.32845	2.88665	C	-6.97024	7.08430	8.15960	H	-8.42698	8.02624	6.89591
H	1.71461	4.05165	1.83424	C	-3.55131	9.65563	9.98443	C	-7.60833	6.22418	6.00319
C	1.01955	5.39252	3.38615	C	-4.13177	9.03723	7.68555	H	-9.34345	4.88564	12.64632
H	0.35688	5.96993	2.74272	C	-7.94431	6.59946	10.97437	C	-8.01442	3.17062	12.62174
								H	-6.45272	1.67907	12.37367
								H	-6.59738	4.38476	5.44769
								C	-4.74447	12.79064	8.13268
								H	-8.64865	2.55121	13.25543
								H	-8.19626	6.29898	5.08882

C -3.73825 5.87885 12.85732
H -5.96086 8.60488 14.39141
C -4.39937 7.14897 14.78644

C -3.59640 6.16276 14.21220
H -3.15327 5.10764 12.35746
H -4.32042 7.38385 15.84755
H -2.87185 5.60529 14.80444

H -5.66956 12.84970 7.54171
H -4.88621 13.36529 9.05753
H -3.95468 13.28877 7.54805
H 0.91190 3.39200 8.47351

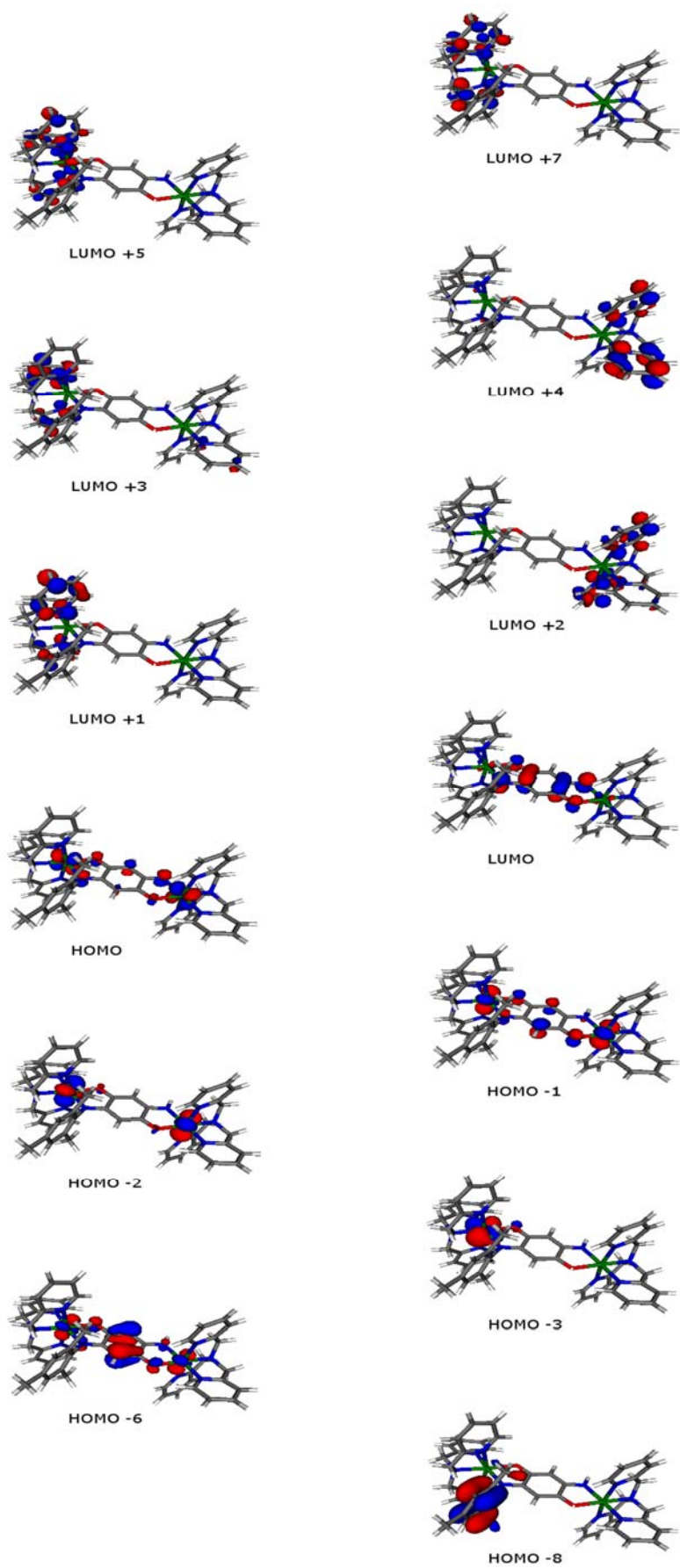


Figure S34: Contributing molecular orbitals of $[2^{\text{NH,N(Mes)}}]^{2+}$ for the main absorption transitions.

Table S11 Atomic coordinates of the optimised geometry of $[2^{\text{NH,N(Mes)}}]^{3+}$.

Ru	2.39595994820262	5.54066276742966	7.46013348835931	C	1.13680787143287	5.71013056695801	4.66722283918281
O	0.92538340492842	6.98112181550243	7.73485733555833	H	0.62348117648111	6.57897212596804	5.07752349902103
N	0.91431872990054	4.44230134943386	8.23280328320420	C	4.10608748986175	3.53056285315599	8.53758341852657
N	3.89741567700306	4.13203468246576	7.18384966944306	H	5.02769979004487	2.93184818333321	8.57940132827825
N	3.78490093173791	6.76810190920342	6.57105642215450	H	3.25957605238418	2.85832020591803	8.73629946347815
N	1.90220250978988	5.01785896835365	5.53330498702228	C	4.12444788938514	4.63764716401083	9.56368777949595
N	3.31172365678384	5.70208683115885	9.29738544539885	C	4.88746545975886	4.59175408609522	10.72337307687999
C	-0.18568880398368	6.52555686032060	8.24479853296454	H	5.53969229674028	3.73862526944654	10.90661757476343
C	-1.30006681236830	7.31235049950734	8.51150949635410	C	4.80628338145343	5.64742135419782	11.63660629178061
H	-1.27804974366378	8.38016379333125	8.29669332742083	H	5.39696717519733	5.62623410340307	12.55205977516093
C	-0.21923150282766	5.07652920779216	8.53646940642047	C	3.96996623286256	6.72760894168284	11.35583134931116
C	5.17202309153589	4.79003364093063	6.71450445305929	H	3.88233596390669	7.57071046654610	12.03930166990391
H	5.84480041049109	4.86995760188662	7.58049217939229	C	3.23859682974708	6.72418757714858	10.17189376455110
H	5.67638375962966	4.15125185752833	5.97791008849834	H	2.57236148100717	7.54011468574371	9.89435447942143
C	4.93812175315055	6.17099893790324	6.17920001820467	C	-2.45643779152229	6.71594874168735	9.05801957940364
C	5.86527235832346	6.84034534193083	5.38518222952613	C	-1.37386274414389	4.47545223834982	9.07969673893870
H	6.77766892045199	6.33304366651036	5.07344321193737	N	-3.60143683282529	7.33607268797802	9.35826430512462
C	5.60614047177210	8.15701286360724	5.00367385514359	C	-2.48214467704156	5.27213364586200	9.34219688606939
H	6.31882891526572	8.69758570731202	4.38125880432994	H	-1.40324728714205	3.40534970887604	9.28929388670669
C	4.42231324437491	8.76910401005617	5.42430230553779	Ru	-5.08454538251508	6.10374249936249	10.15978041719505
H	4.18368228166871	9.79443253845130	5.14660176122773	C	-3.74812607692221	8.72109784415639	9.05214636243886
C	3.52983640742747	8.04241353078117	6.20089041257503	O	-3.59541754217279	4.79322306451499	9.83753496787244
H	2.58017652308888	8.45197975469317	6.54363402180448	N	-6.72989710715543	7.35440346488452	10.54561282911281
C	3.35954502685434	3.17639427967013	6.16715297508590	N	-6.34037564181770	4.71762521787061	11.00499061657658
H	2.66898323735641	2.49980836095461	6.69114852523639	N	-4.63486177332102	6.57393537883844	12.10515905371236
H	4.15889656984057	2.56364443430186	5.72690945281049	N	-6.04457659001990	6.03131708870914	8.33690360835976
C	2.59147970879348	3.92151525341118	5.10457339872359	C	-6.30534134318819	8.27265926523628	11.64291551493152
C	2.52741545519738	3.50910050552351	3.77912960048511	C	-5.44090808243317	7.54048721649438	12.63134685908088
H	3.09864276825604	2.63942374464949	3.45643902349696	C	-7.01823013295630	8.05610384806330	9.26020236639930
C	1.72865984709237	4.22166344639425	2.88006449798642	C	-6.93295355925465	7.04886594130106	8.14304780575148
H	1.66780792316331	3.91183092438171	1.83714309295833	C	-3.56813617955991	9.68417351443717	10.06560027603248
C	1.02077600596914	5.33537787512850	3.33234853271759	C	-4.13332955271998	9.11003905135552	7.74816040902203
H	0.39124725113910	5.91911410497224	2.66263652263268	C	-7.92924915234766	6.53095249342239	10.94446442598122

C	-7.54500350066248	5.16946331811689	11.43340394151921	H	-1.99234174768706	9.86592759751115	11.50730879633252
C	-5.94886200463330	3.46350738695086	11.32261755057665	H	-4.80032850554117	10.74655332880726	6.52877965416351
C	-5.88563848717233	5.09299833173048	7.38147281854385	H	-4.15273081337867	7.10016853702151	6.92358631650607
C	-3.91646010251709	11.01420205330103	9.79276893171310	H	-5.01018499573856	8.31377077573550	5.94103219475097
C	-2.94905619557184	9.33476941196810	11.39468973053093	H	-3.24036997310588	8.28151557956310	5.98142311206596
C	-4.47082684979055	10.44943250741726	7.52777841997556	C	-5.38829705811635	7.86128797687443	13.98204949060601
C	-4.14462318522420	8.14263527111445	6.59421461448992	H	-8.38649565857888	7.95518013507182	6.85063170793507
H	-5.71154104518916	9.07428193882506	11.17812752489667	C	-7.49846557763958	6.17920116666322	5.97307329787809
H	-7.17271627226432	8.73699206847296	12.13201743397693	H	-9.34081208701529	4.77436813120676	12.54928434651696
H	-8.00030831457130	8.54992376379991	9.28230455535150	C	-7.98584625238072	3.08010557721989	12.53558314421585
H	-6.24737011847837	8.82676041199514	9.12024613248792	H	-6.39959487595424	1.61195374623461	12.30786493500931
C	-7.67650514062301	7.13819553648887	6.97393576163721	H	-6.42663751732273	4.36903051373469	5.43424473159503
H	-8.55577879182777	6.40629172356665	10.04975521734108	C	-4.78037334924053	12.84573379589167	8.27388841981476
H	-8.53041611148535	7.07664946624597	11.68282073221610	H	-8.62830047569348	2.44293686306900	13.14266359746572
C	-8.38513979083255	4.37635831660336	12.20956154189497	H	-8.06821294107949	6.24027748089119	5.04646361638189
C	-6.75054710510296	2.61644858349546	12.07669001047787	C	-3.76072878506078	5.92586909579131	12.90289581711326
H	-4.96654787872210	3.16714647918167	10.95714689574621	H	-6.05302601358820	8.62852776412478	14.37723564180169
C	-6.59146175971006	5.14134067249379	6.18379397150290	C	-4.47459577211433	7.20193880718976	14.80795102788142
H	-5.16691938237467	4.30433403950168	7.59948233949359	C	-3.64618683617067	6.22440705011476	14.25627665626522
H	-3.79755116054931	11.75895091632480	10.58392781080485	H	-3.15358910758966	5.15880458831152	12.42452779795148
C	-4.39776346292085	11.41361438478242	8.54206635319807	H	-4.41643834043100	7.44643060083597	15.86802562500126
H	-3.58338562351274	9.64481896294134	12.23730746224619	H	-2.92219276935712	5.68359818406689	14.86363989314453
H	-2.74904355394026	8.26313024436559	11.48443785037532	H	-5.74316348411488	12.90781296860813	7.74661320129889
				H	-4.85805363106043	13.41863974618861	9.20699039182418
				H	-4.03054844414245	13.34021338212497	7.63655499512498
				H	0.88855160422909	3.43536206697828	8.42171325022300

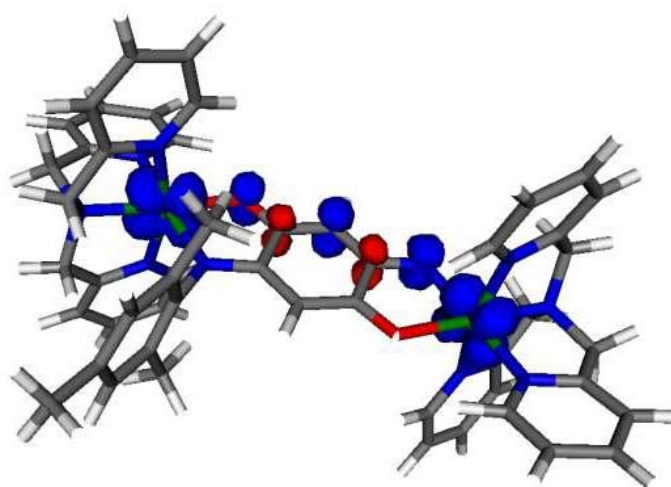


Figure S35: Spin-density distribution of $[2^{NH,N(Mes)}]^{3+}$.

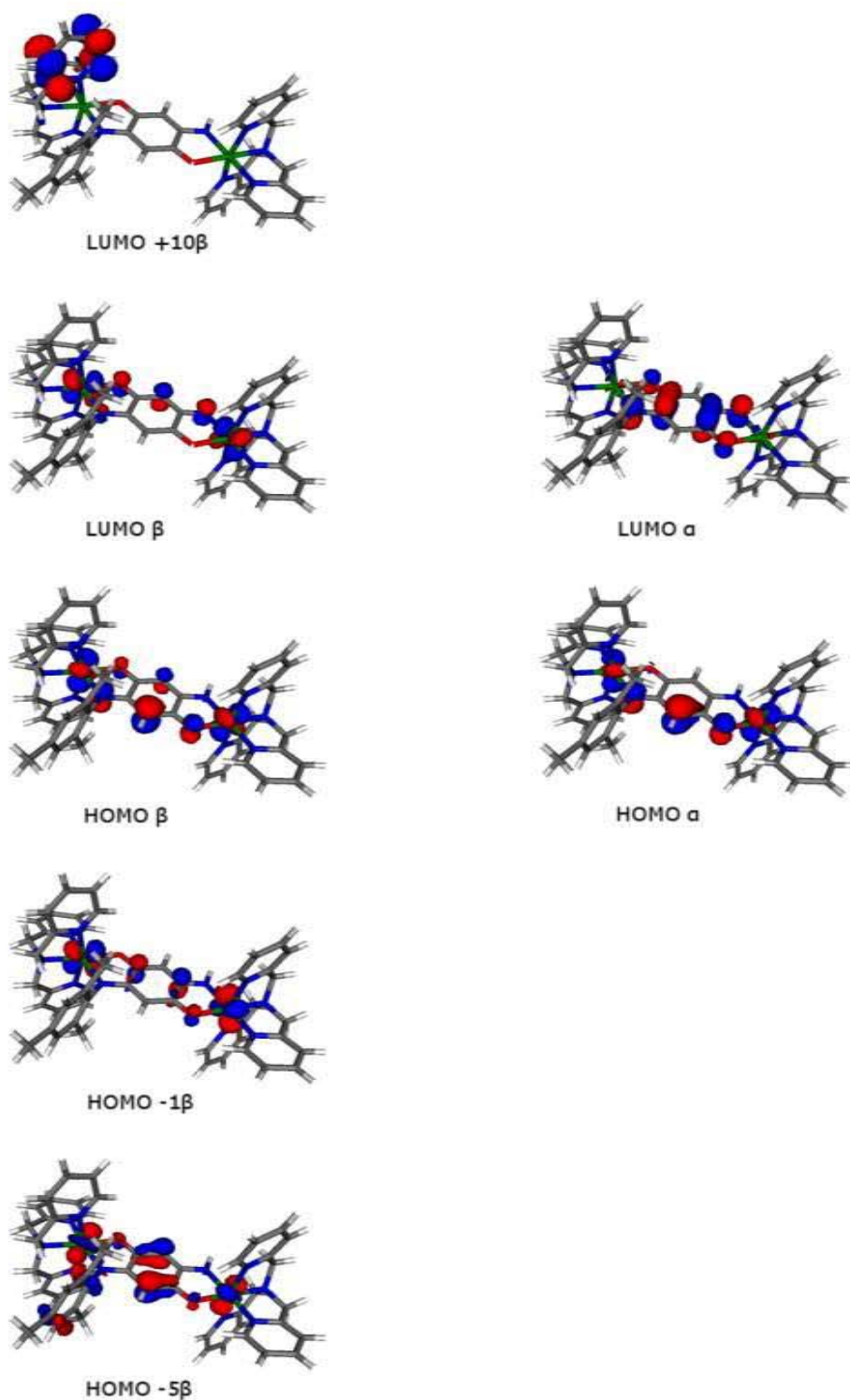


Figure S36: Contributing molecular orbitals of $[2^{\text{NH,N(Mes)}}]^{3+}$ for the main absorption transitions.

Table S12 Atomic coordinates of the optimised geometry of $[2^{\text{NH,N(Mes)}}]^{4+}$.

Ru	2.36172515606884	5.53441037572969	7.47524105920949	C	1.11434984616822	5.71641137308172	4.68725816577496
O	0.92518066848282	6.91595208624542	7.77080576974897	H	0.60475042455132	6.58333983597701	5.10479209208998
N	0.91036285727858	4.39766361860536	8.25693014662195	C	4.14052853545022	3.52249898704217	8.49299906314707
N	3.88987216118566	4.14780161714044	7.15612079289372	H	5.07002409939625	2.93703461727245	8.49025914942212
N	3.72777663636485	6.80581825567929	6.60286799496326	H	3.30988867753325	2.83427830311002	8.70399930630644
N	1.89103430023282	5.01306450625467	5.53496018415767	C	4.17152227523247	4.61309403647402	9.52976568357873
N	3.34399514093380	5.66873897698423	9.28751738568660	C	4.95240238674693	4.56845039458795	10.67657666192724
C	-0.19033516944634	6.46294627657083	8.28305130613519	H	5.61826628684443	3.72270506427463	10.84232064872875
C	-1.28993803348645	7.27009867114005	8.54456148438577	C	4.86938332206150	5.61571726864222	11.59798618345346
H	-1.25372321697879	8.33817086396254	8.33525096387740	H	5.47354753731641	5.59559189360153	12.50441681898556
C	-0.22441115704265	5.01916113973996	8.57253668313048	C	4.01600602794308	6.68780957547084	11.33866891336523
C	5.14339180327533	4.83680309004960	6.66746387680479	H	3.92775059471585	7.52504407163461	12.02873265042686
H	5.84986796511383	4.88330461074728	7.50833941674300	C	3.26977777030972	6.68823928763855	10.16545794621257
H	5.61921917955756	4.22860473459909	5.88804678137366	H	2.59621484038213	7.50320773659660	9.90504278823550
C	4.88737679951212	6.23617948044074	6.19372094396856	C	-2.44253867453453	6.67154042144903	9.09499668384283
C	5.81072778025161	6.94909177546734	5.43412705939305	C	-1.37722249413644	4.42252092217195	9.12490103469621
H	6.73088392048566	6.46612609901986	5.10766965987346	N	-3.58420811055435	7.30656655098902	9.39513043038828
C	5.53792330136832	8.27684137380130	5.10750853859279	C	-2.47560687319010	5.23216967414021	9.38612994434475
H	6.24795785829051	8.85101630258545	4.51301927047577	H	-1.41331503210240	3.35397594812139	9.33699426833295
C	4.34572003783440	8.85870681883388	5.54667552962246	Ru	-5.04895502535875	6.12388921209920	10.19205461851073
H	4.09733357152804	9.89216556102124	5.31188818166032	C	-3.72778710454729	8.68404667828044	9.03484424621374
C	3.45754875521455	8.09148620478219	6.28665670534796	O	-3.59645070617244	4.77616848476428	9.88218727507256
H	2.50343764252568	8.47987957452749	6.64060598356109	N	-6.67737044801675	7.40326443117446	10.58099242069858
C	3.35739733110094	3.18309034370204	6.14304449265786	N	-6.35131910126510	4.74107707110196	10.99938808713603
H	2.67830530862416	2.49610787408660	6.66845999495613	N	-4.62849120005680	6.55796860753304	12.16203069950857
H	4.16660676327965	2.58451021266990	5.70443811430304	N	-6.04459516112546	6.06510561170684	8.37913367167913
C	2.58007702291592	3.92505680319595	5.09150481604147	C	-6.24847712768173	8.30677260976501	11.68908894149866
C	2.50281063881057	3.52556131336898	3.76358360789602	C	-5.40754042320342	7.55079568314569	12.67872962089395
H	3.07400326487620	2.66149770161154	3.42710639581853	C	-6.95809303441053	8.11972039637315	9.30100165360192
C	1.69047988470491	4.24351546769472	2.88254129065337	C	-6.89703356875684	7.11180952606580	8.18400491911882
H	1.61827647621777	3.94391678513385	1.83755575628197	C	-3.59160659937839	9.67617569396353	10.02612496908418
C	0.98355438460232	5.35039559934611	3.35267649538066	C	-4.07319463474518	9.01526715316389	7.70520596427367
H	0.34378500077134	5.93815240903965	2.69686817658152	C	-7.89490060092629	6.59744453058154	10.97083832341168
				C	-7.54896963095321	5.21297796935453	11.41862991399112
				C	-5.99096477628788	3.46938343293198	11.27922217024037
				C	-5.92312372026290	5.10615932375636	7.43930259194443
				C	-3.94812960128062	10.98961716688217	9.69737212963294
				C	-2.98490876193311	9.37797528870415	11.37249907292486
				C	-4.42458999182696	10.34137511906228	7.43435155191720

C	-4.04036811979939	8.01229251291651	6.58306881118027	C	-5.35017693599329	7.86934120158613	14.02952381212924
H	-5.64248005313164	9.10708651065151	11.23902843514175	H	-8.31991748456091	8.05502177571292	6.88386010156782
H	-7.11424946240422	8.77793317968696	12.17281019132062	C	-7.49661120070387	6.23920128827373	6.02601694582932
H	-7.93284317893063	8.62555913392176	9.33331404297304	H	-9.37379647553173	4.82507946283545	12.49032071015560
H	-6.18075492939807	8.88301722145333	9.16212385578130	C	-8.05735597330770	3.10175532657574	12.44219165232582
C	-7.63778343164345	7.21620196808292	7.01432039858806	H	-6.50553613851285	1.59969181467742	12.19517910296152
H	-8.53775541990730	6.51627755489821	10.08293989784345	H	-6.50120909403415	4.38015627264548	5.50482480087978
H	-8.46939350815914	7.14023899237340	11.73139738632362	C	-4.79219772405152	12.75310542077683	8.09217291252203
C	-8.42195497887752	4.41621790091535	12.15377208651544	H	-8.72564211300167	2.46146738866804	13.01721063651957
C	-6.82625086513393	2.61954288394635	11.99042629361784	H	-8.06636875021571	6.31126485505039	5.10026567608137
H	-5.00982567736819	3.16051515395834	10.92181570460044	C	-3.78325167018087	5.87466575616091	12.96094007766222
C	-6.63155224921261	5.16752174441231	6.24498705678516	H	-5.99041974799463	8.65764500582441	14.42288964778069
H	-5.23433825979063	4.29327669689735	7.66409897601297	C	-4.46495622590160	7.17653201600379	14.85808920620960
H	-3.86090078860245	11.76191128291003	10.46514800621341	C	-3.66767338670300	6.16913127673615	14.31406762092519
C	-4.39780513787562	11.33800733913243	8.41907713284675	H	-3.20140889635268	5.08595420252015	12.48687029904661
H	-3.55499697185869	9.84206347558767	12.18867133849937	H	-4.40521421272474	7.41819717067118	15.91866233020050
H	-2.89863085976520	8.30443828284506	11.56430837368552	H	-2.96827003986384	5.60303842642900	14.92657890222031
H	-1.96869263660043	9.79810385115564	11.41617719906583	H	-5.70844021235370	12.77925476526587	7.48595333081298
H	-4.73140624100792	10.59992628446189	6.41817420798244	H	-4.95774592313214	13.34149049539268	9.00359202311073
H	-4.08839151428076	6.97713747289086	6.93213558112470	H	-4.00270247094212	13.25178649616624	7.50825934789453
H	-4.86428135621280	8.18349421956282	5.87895445826139	H	0.90622209888874	3.38985083861263	8.44219932856691
H	-3.10208371726423	8.11696141429065	6.01626880282369				

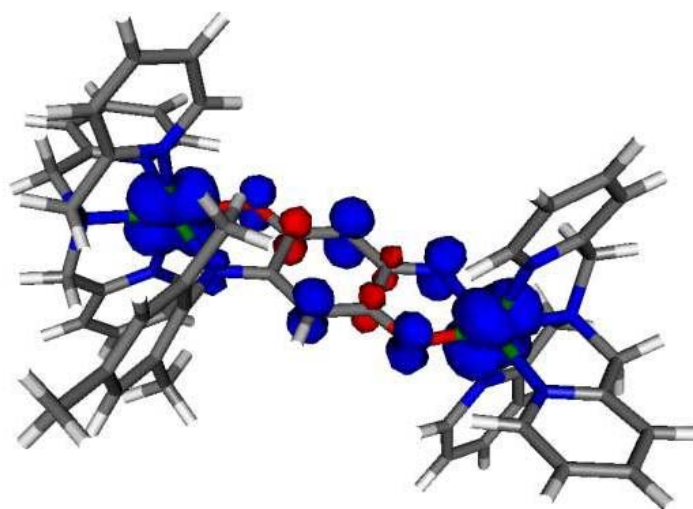


Figure S37: Spin-density distribution of $[2^{\text{NH,N(Mes)}}]^{4+}$.

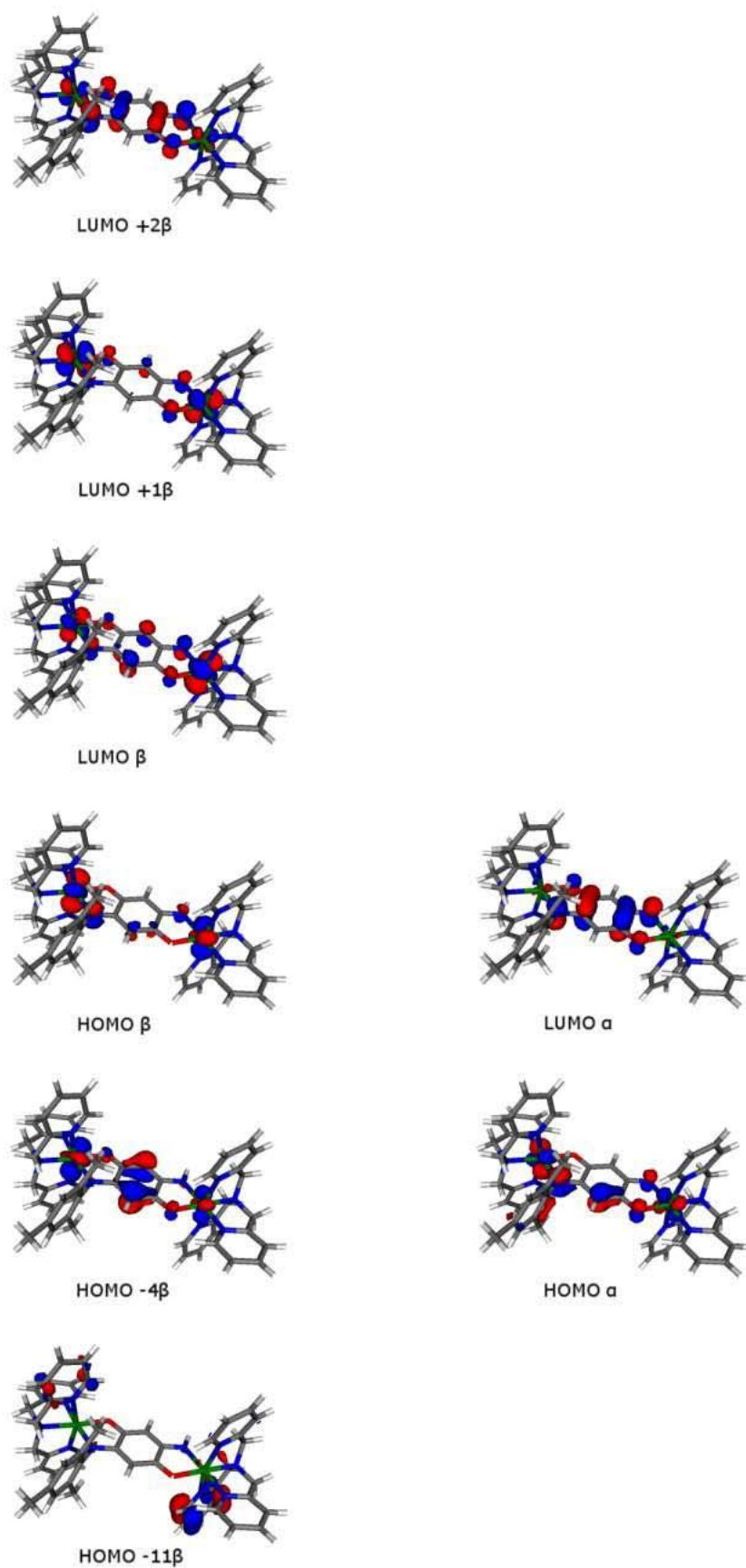


Figure S38: Contributing molecular orbitals of $[2^{\text{NH},\text{N}(\text{Mes})}]^{4+}$ for the main absorption transitions.

Table S13 Atomic coordinates of the optimized geometry of $[2^{\text{NH,N(Mes)}}]^+$.

Ru	2.46802750505337	5.55265664488606	7.53994353621201	C	1.11283527685289	5.83353501379649	4.82657255712705
O	0.98707482903542	6.98842432266854	7.87436526924708	H	0.58307318532676	6.65391076558031	5.31068900000177
N	1.00277804851803	4.40362633063748	8.37272754593424	C	4.20390128252826	3.50468883622116	8.49470588191208
N	3.96932410231154	4.16036816325216	7.17546384342726	H	5.13378811040598	2.91343584181943	8.50548474379747
N	3.77910197807984	6.78872948413406	6.59028671707809	H	3.36361710094206	2.81906126396593	8.67179514296896
N	1.90411816083552	5.10282451042056	5.64320564728280	C	4.22115263025161	4.56253507947086	9.57543418066743
N	3.41108218448812	5.64172465255312	9.35246416851002	C	4.98105240734025	4.45530727405867	10.73272140632973
C	-0.13917329610630	6.47395477183044	8.35381976914729	H	5.62958622543007	3.59050215122834	10.87298792445350
C	-1.28365946132141	7.24154273451370	8.59059293458340	C	4.90119527605725	5.46119905674718	11.70410581196351
H	-1.26583449052926	8.31035054029804	8.36921267823504	H	5.48768078930300	5.39022345083171	12.61990953225666
C	-0.16015987251135	5.04758917622954	8.63923721492850	C	4.06399128519018	6.55309420767668	11.47447455911214
C	5.22135908106334	4.85889595187884	6.71647540056251	H	3.96986050648996	7.35788937370833	12.20291984551918
H	5.84840399580464	5.01114324456376	7.60741784621039	C	3.33746340041296	6.61165322004922	10.28731644717215
H	5.78942694817989	4.22051230270291	6.02554652761019	H	2.66595612200892	7.43645916283706	10.04994755380811
C	4.91928045582329	6.20102366946417	6.11984451287675	C	-2.45830581637463	6.65056740286203	9.10651166761173
C	5.75379921376973	6.84120524846816	5.21131192204059	C	-1.33853373099129	4.45928950742203	9.15696616095885
H	6.64452062038353	6.32872729919562	4.84670870810021	N	-3.62528280231265	7.30584676855335	9.36018283189530
C	5.43944958262271	8.13449488555459	4.78058989246504	C	-2.48298418033359	5.22807732996152	9.39255356309603
H	6.08161657022446	8.65343534400722	4.06951263701013	H	-1.36374622717170	3.38614092116585	9.36804255908925
C	4.27852831829077	8.73962609589986	5.27691121659341	Ru	-5.13010700324068	6.11179038396259	10.15479479050257
H	3.98898168001462	9.74410026975532	4.96897044500485	C	-3.74145532053469	8.67154628904220	9.01257211771205
C	3.47244467170521	8.04075226065527	6.16549218062783	O	-3.61387720992939	4.71451666833871	9.85697147474730
H	2.54321927643518	8.44814393397790	6.56373361202706	N	-6.75267595079211	7.38654194465322	10.51615403627905
C	3.41085630765335	3.25687444825919	6.12431335957252	N	-6.38580757034086	4.77097846768075	11.03089945801442
H	2.73651843317173	2.55633215570687	6.63702874745138	N	-4.67092734691261	6.53926443778389	12.08643797635690
H	4.19960137951493	2.67000639214648	5.62831647880722	N	-6.03834215781005	6.02385780729350	8.32034283281360
C	2.61151107389042	4.05264713684317	5.12165734460992	C	-6.31021874256877	8.27902983963174	11.62482346294482
C	2.54770897879059	3.73921293617654	3.76944780959854	C	-5.48450492526404	7.50453677960807	12.62077181264409
H	3.13943379232393	2.90987229529643	3.38155201737570	C	-7.00513929309180	8.07472725293275	9.22055736445243
C	1.72374178843914	4.49300169100060	2.92530392232419	C	-6.91330556058473	7.05461884716168	8.10841465757099
H	1.66109118684030	4.25892139035521	1.86283482174683	C	-3.57398999953054	9.67090433027032	9.99802237395238
C	0.99335338213649	5.55337158333505	3.46924823037662	C	-4.08275182842342	9.04645469242508	7.68942706473512
H	0.34098014055300	6.16767227405975	2.84911965735988	C	-7.96412833704343	6.57987380144273	10.90108053258732
				C	-7.58298080671811	5.25110976384061	11.47474560966292
				C	-5.99240580806802	3.53460801616987	11.42810627956074
				C	-5.88436886433474	5.08641219026398	7.36196929532545
				C	-3.90402424599903	10.99790019844387	9.68783069262808
				C	-2.99617785325457	9.33959874406715	11.35068256614267

C	-4.40038737649064	10.38325923092456	7.42375044410036	H	0.98111472026743	3.40177657985598	8.56995445284888
C	-4.08533459866379	8.04338314548962	6.56601608747669				
H	-5.68126425317108	9.05630964161022	11.16646635380514				
H	-7.16215358084037	8.77416531617679	12.11638148940575				
H	-7.98121631135662	8.58596744384077	9.20555726806265				
H	-6.21605790270156	8.82795759620263	9.08773162791461				
C	-7.64446069465654	7.14968617649964	6.93197806126550				
H	-8.53944735124762	6.40405193023752	9.98021650313900				
H	-8.60815219462204	7.15429927929042	11.58135801006432				
C	-8.39581727920545	4.51773806958816	12.33151723025328				
C	-6.77172661499016	2.74536620353297	12.26310913088446				
H	-5.01888290099618	3.21937258929715	11.05268650671491				
C	-6.57901008746558	5.13707088793438	6.15636424806553				
H	-5.16794991600618	4.29855215239342	7.59347342271831				
H	-3.80277710777396	11.76131178332811	10.46486138109873				
C	-4.34642460506615	11.37295958928084	8.41473689108709				
H	-3.64009455147241	9.68495968826189	12.17300122491896				
H	-2.84219565219818	8.26175974504177	11.45838150633405				
H	-2.02432780284917	9.84134513821932	11.47564832756112				
H	-4.70277629485297	10.65903263818841	6.40909439419172				
H	-3.98861521642520	7.02204783469610	6.94409966417538				
H	-5.00301439192048	8.11442784105469	5.96548548088497				
H	-3.23578851783032	8.23088411849936	5.89107091735718				
C	-5.48518724292724	7.77407938563220	13.98321990396542				
H	-8.34075666843203	7.97748291338008	6.79803099760670				
C	-7.47233427537774	6.18467785562971	5.93249850302807				
H	-9.33696869948780	4.94624601736435	12.67711502762837				
C	-7.99409677490794	3.24016918084332	12.73346357339429				
H	-6.41398295475296	1.75705943603581	12.55083496568836				
H	-6.41475421403890	4.36257588358806	5.40788721130136				
C	-4.70605764715090	12.80368478333119	8.10150516723196				
H	-8.61713630075981	2.64842944365292	13.40354356726447				
H	-8.03243812338882	6.25125219965976	5.00005734297519				
C	-3.84626282999649	5.84873177024101	12.90527852262786				
H	-6.15564251123460	8.53764977507727	14.37793498549290				
C	-4.62026752676342	7.06862533898818	14.82794004757054				
C	-3.78760471566000	6.09296255246380	14.27341537532873				
H	-3.23352195282811	5.09299700669494	12.41413530884336				
H	-4.60525031329234	7.27332459505301	15.89816346464431				
H	-3.10109508793356	5.51637898353395	14.89288095771038				
H	-5.64128813272398	12.86450518347939	7.52591330748616				
H	-4.82791907758135	13.39356647163874	9.01986792712621				
H	-3.92310763762622	13.28740390657208	7.49566551744542				

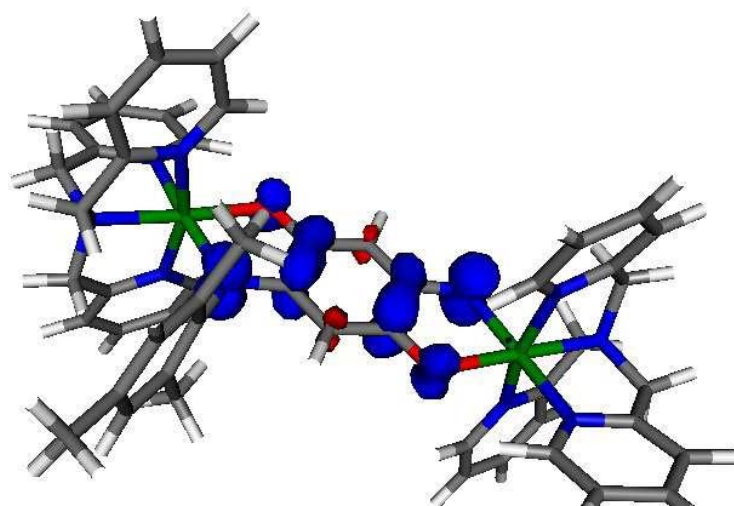


Figure S39: Spin-density distribution of $[2^{\text{NH},\text{N}(\text{Mes})}]^+$.