Supporting Information

Solvent- and metal-gated coordination diversity in a dynamic aminal ligand

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General Considerations

All chemicals and solvents were purchased from commercial suppliers and used without further purification. Deuterated solvents ($CDCl_3$, CD_3OD, CD_3CN, acetone-d_6, and DMSO-d_6) were dried over 3 Å molecular sieves prior to use. All reactions in this study were performed under ambient conditions because our laboratory currently does not have access to a glovebox after the 2024 disaster.[1] NMR spectra were recorded on Bruker 400 MHz and Varian 600 MHz spectrometers. Chemical shifts (δ) are referenced to the residual ^1H and ^{13}C NMR signals of the deuterated solvent. Coupling constants (J) are reported in Hz, with multiplicities indicated as s (singlet), d (doublet), t (triplet), q (quartet), and m (multiplet). UV-Vis spectra were acquired on SHIMADZU UV-2600 spectrophotometer, Agilent Cary-60 and a JASCO V-550 UV/VIS spectrophotometer using quartz cuvettes (path length = 1 cm) in solution. FT-IR/ATR spectra were measured on a SHIMADZU IRSpirit spectrometer. Electrospray ionization (ESI) mass spectra were obtained using a Bruker micrOTOF II instrument. Cyclic voltammet was performed at room temperature under an nitrogen atmosphere on a CH Instruments CHI6111F system equipped with a 3 mm diameter glassy carbon working electrode (CHI104), a platinum wire counter electrode, an Ag/AgCl reference electrode (RE-1B), and [n-Bu_4N][ClO_4] as the supporting electrolyte. Single-crystal X-ray single-crystal diffraction data were collected on Rigaku XtaLAB Synergy-S, Bruker or Oxford diffractometers. Elemental analysis was performed using a Thermo Scientific FLASH 2000 CHNS analyzer.

Computational Details

All calculations were performed using the Gaussian 16 program suite. The geometries of all stationary states were optimized at the UB3LYP level. The LANL2DZ basis set was used for the central metal ions, while 6-31g* was applied to the other atoms. To account for dispersion interactions and solvent effects, we incorporated Grimme's D3 empirical correction and the polarizable continuum model (PCM), respectively. The absence of imaginary frequencies confirmed that all optimized structures correspond to local minima. Intermolecular interactions involving $n \rightarrow \pi^*$ transitions were assessed using second-order perturbation energy from NBO analysis.

Synthesis and Spectral Characterization

Preparation and characterization of $[Co(L^{O1})_2](ClO_4)$. $Co(ClO_4)_2 \cdot 6H_2O$ (185.6 mg, 0.52 mmol) was dissolved methanol (15 mL) in a 25 mL round-bottom flask equipped with a magnetic stir bar. L^{A1} (280.0 mg, 1.01 mmol) was then added to the solution, and the reaction mixture was stirred at room temperature for 4 hours. Upon completion, the solvent was removed by rotary evaporation, and the resulting pink solid was washed three times with CH_2Cl_2 and Et_2O, then dried under vacuum to afford **$[Co(L^{O1})_2](ClO_4)$** as a pink powder (167.5 mg, 51% yield). Recrystallization from acetone yielded single crystals suitable for X-ray diffraction. ^1H

NMR (500 MHz, DMSO-d_6): δ:8.84 (d, $J_{H\text{-}H}$=3.6 Hz, 2H), 8.40 (d, $J_{H\text{-}H}$=5.4 Hz, 2H), 8.31 (d, $J_{H\text{-}H}$=6.0 Hz, 2H), 7.97 (dd, $J_{H\text{-}H}$=7.4, 7.4 Hz, 2H), 7.60 (d, $J_{H\text{-}H}$=7.6 Hz, 2H), 7.41 (m, 4H), 6.50 (dd, $J_{H\text{-}H}$=6.4, 6.4 Hz, 2H), 6.41 (d, $J_{H\text{-}H}$=8.4 Hz 2H), 5.54 (d, $J_{H\text{-}H}$=3.9 Hz, 2H) ppm. ^{1}H NMR (400 MHz, Acetonitrile-d_3): δ:8.43 (m, 4H),7.87 (t, $J_{H\text{-}H}$ =7.7 Hz, 2H), 7.50 (d, $J_{H\text{-}H}$ =7.7 Hz, 2H), 7.37 (dd, $J_{H\text{-}H}$ =7.7, 7.7 Hz, 2H), 7.29 (d, $J_{H\text{-}H}$ =6.6 Hz, 2H), 7.17 (s, 2H), 6.49 (dd, $J_{H\text{-}H}$ =6.5, 6.6 Hz, 2H), 6.36 (d, $J_{H\text{-}H}$ =8.4 Hz 2H), 5.56 (d, $J_{H\text{-}H}$ =4.1 Hz, 2H) ppm. ^{13}C NMR (500 MHz, DMSO-d_6): δ: 169.4, 156.1, 148.2, 147.4, 140.5, 138.7, 124.6, 118.4, 113.6, 112.1, 83.7 ppm.UV-Vis (EtOH): λ_{max} (ε in $M^{-1}\cdot cm^{-1}$) = 514 (90) nm ; UV-Vis (MeCN) : λ_{max} (ε in $M^{-1}\cdot cm^{-1}$) = 519 (52) nm ; UV-Vis (MeOH): λ_{max} (ε in $M^{-1}\cdot cm^{-1}$) = 510 (64) nm ; UV-Vis (DMSO): λ_{max} (ε in $M^{-1}\cdot cm^{-1}$) = 513(93) nm; HR-ESIMS: m/z 459.0968 $[M\text{-}ClO_4]^+$calcd. for $(C_{22}H_{20}CoN_6O_2)^+$: 459.0980. Elemental Anal. Calcd. for $C_{22}H_{20}CoN_6ClO_6$: C, 47.29; H, 3.61; N, 15.04. Found: C, 47.66; H, 3.67; N, 15.29.

Preparation and characterization of $[Co(L^{O2Me})_2](ClO_4)_2$. $Co(ClO_4)_2\cdot 6H_2O$ (101.5 mg, 0.28 mmol) was dissolved in methanol (15 mL) in a 25 mL round-bottom flask equipped with a magnetic stir bar. A solution of 2-pyridinecarboxaldehyde (59.4 mg, 0.56 mmol) and 2-aminopyrimidine (52.8 mg, 0.56 mmol) in MeOH was then added, and the reaction mixture was stirred at room temperature for 0.5 h. The solvent was removed under reduced pressure, and the residue was left undisturbed and exposed to air at room temperature. Orange crystals suitable for X-ray diffraction were obtained within 1 day. The sample was further left in air for a total of 7 days to allow slow oxidation, consistent with the transformation shown in Scheme 2. The isolated yield of the Co(III) product is difficult to determine because the Co(II) → Co(III) oxidation proceeds slowly and both species coexist in a dynamic mixture. However, the ^{1}H NMR spectra clearly show gradual changes over time, with the broad, shifted paramagnetic resonances of Co(II) diminishing and the well-defined diamagnetic signals of Co(III) appearing, confirming the slow oxidation process. UV-Vis (MeOH): λ_{max} (ε in $M^{-1}\cdot cm^{-1}$) = 486 (97) nm. The ESIMS and elemental analysis could not be obtained because the product is dynamic and unstable.

Preparation and characterization of $[Co(L^{O2})_2](ClO_4)$. $Co(ClO_4)_2\cdot 6H_2O$ (184.2 mg, 0.50 mmol) was dissolved methanol (15 mL) in a 25 mL round-bottom flask equipped with a magnetic stir bar. L^{A2} (279.8 mg, 1.00 mmol) and TBHP (1 equiv, 70% aqueous solution) were then added. The reaction mixture was stirred at room temperature for 4 hours. Upon completion, the solvent was removed by rotary evaporation, and the resulting pink solid was washed three times with CH_2Cl_2 and Et_2O, then dried under vacuum to afford **$[Co(L^{O2})_2](ClO_4)$** as a pink powder (205.4 mg, 74% yield). Recrystallization from acetone yielded single crystals suitable for X-ray diffraction. ^{1}H NMR of major form (500 MHz, DMSO-d_6): δ 9.48 (d, $J_{H\text{-}H}$ = 4.2 Hz, 2H), 8.63 (dd, $J_{H\text{-}H}$ = 6.2, 2.3 Hz, 2H), 8.37 (dd, $J_{H\text{-}H}$ = 5.8, 1.5 Hz, 2H), 8.35 (dd, $J_{H\text{-}H}$ = 4.4, 2.2 Hz, 2H), 8.02 (dd, $J_{H\text{-}H}$ = 7.6, 1.5 Hz, 2H), 7.61 (d, $J_{H\text{-}H}$ = 7.7 Hz, 2H), 7.44 (dd, $J_{H\text{-}H}$ = 6.6, 1.5 Hz,2H), 6.78 (dd, $J_{H\text{-}H}$ = 6.1, 4.4 Hz, 2H), 5.65 (d, $J_{H\text{-}H}$ = 4.2 Hz, 2H) ppm. ^{1}H NMR of

major form (400 MHz, Acetonitrile-d_3):δ 8.71 (dd, $J_{H\text{-}H}$ = 6.1, 2.2 Hz, 2H),δ 8.39 (d, $J_{H\text{-}H}$ = 5.5 Hz, 2H), δ 8.24 (dd, $J_{H\text{-}H}$ = 2.2, 4.2 Hz, 2H), 7.90 (td, $J_{H\text{-}H}$ = 1.2, 7.7 Hz, 2H), 7.66(d, $J_{H\text{-}H}$ = 3.1 Hz , 2H), 7.53(d, $J_{H\text{-}H}$ = 7.7 Hz, 2H), 7.32(td, $J_{H\text{-}H}$ = 1.4, 6.7Hz , 2H), 6.66(dd, $J_{H\text{-}H}$ = 5.3, 4.5 Hz, 2H) , 5.69(d, $J_{H\text{-}H}$ =4.2 Hz, 2H) ppm. ^{1}H NMR of minor form (400 MHz, DMSO-d_6): δ: 9.48 (d, $J_{H\text{-}H}$ = 4.4 Hz, 2H), 8.70 (d, $J_{H\text{-}H}$ = 6.0 Hz, 2H), 8.17 (d, $J_{H\text{-}H}$ = 6.0, 2H,), 8.14 (d, $J_{H\text{-}H}$ = 6.0, 2H), 8.09 (dd, $J_{H\text{-}H}$ = 7.6, 6.6 Hz, 2H), 7.71 (d, $J_{H\text{-}H}$ = 7.6 Hz, 2H), 7.55 (dd, $J_{H\text{-}H}$ = 6.6, 6.6 Hz, 2H), 6.58 (dd, $J_{H\text{-}H}$ = 6.0, 6.0 Hz, 2H), 5.71 (d, $J_{H\text{-}H}$ = 4.4 Hz, 2H). ^{13}C NMR of major form (500 MHz, DMSO-d_6): δ: 168.9, 161.6, 158.2, 157.9, 147.9, 140.9, 125.17, 118.9, 112.8, 84.6 ppm.^{13}C NMR of minor form (100 MHz, DMSO-d_6): δ: 167.2, 159.2, 158.6, 152.0, 144.4, 140.5 138.3, 123.2, 116.0, 81.3 ppm. UV-Vis (EtOH): λ_{max} (ε in $M^{-1}\cdot cm^{-1}$) = 509 (77) nm ; UV-Vis(MeCN) : λmax (ε in $M^{-1}\cdot cm^{-1}$) = 511 (76) nm ; UV-Vis (MeOH): λ_{max} (ε in $M^{-1}\cdot cm^{-1}$) = 510 (69) nm ; UV-Vis (DMSO): λ_{max} (ε in $M^{-1}\cdot cm^{-1}$) = 512 (69) nm ; HR-ESIMS: *m/z* 461.0873 $[M\text{-}ClO_4]^+$calcd. for $(C_{20}H_{18}CoN_8O_2)^+$:461.0885. Elemental Anal. Calcd. for $C_{20}H_{18}CoN_8ClO_6$: C, 42.84; H, 3.24; N, 19.98. Found: C, 42.97; H, 3.57; N, 20.16.

Preparation and characterization of $[Co(L^M)](ClO_4)_3$. $Co(ClO_4)_2\cdot 6H_2O$ (92.6 mg, 0.25 mmol) was dissolved in acetonitrile (15 mL) in a 25 mL round-bottom flask equipped with a magnetic stirrer. L^{A2} (208.9 mg, 0.75 mmol) and $NaClO_4$ (30.6 mg, 0.25 mmol) were then added. The reaction mixture was stirred and heated to reflux for 1 hour. After completion, the mixture was cooled to room temperature, and the solvent was removed by rotary evaporation. Methanol (15 mL) was then added to the residue, inducing the precipitation of the crude product, **$[Co(L^M)](ClO_4)_3$**. The solid was purified by washing with methanol and diethyl ether, yielding the desired product as a solid (133.4 mg, 59% yield). Recrystallization via slow diffusion of diethyl ether into an acetonitrile solution afforded single crystals suitable for X-ray diffraction analysis. ^{1}H NMR (400 MHz, DMSO-d_6): δ: 8.61 (m, 6H), 8.60 (d, $J_{H\text{-}H}$=4.4 Hz, 3H), 8.55 (d, $J_{H\text{-}H}$=6.6 Hz, 3H), 8.33 (ddd, $J_{H\text{-}H}$=8.0, 7.6, 1.4 Hz, 3H), 8.11 (dd, $J_{H\text{-}H}$=8.0, 1.4 Hz, 3H), 7.93 (ddd, $J_{H\text{-}H}$=7.6, 5.9, 1.4 Hz, 3H), 7.67 (s, 3H), 6.95 (dd, $J_{H\text{-}H}$= 6.6, 4.4 Hz, 3H) ppm. ^{13}C NMR (100 MHz, DMSO-*d6*): δ: 165.3, 157.6, 155.8, 150.7, 147.9, 143.0, 129.3, 123.9, 110.0, 80.3 ppm. UV-Vis (CH_3CN): λ_{max} (ε in $M^{-1}\cdot cm^{-1}$) = 480 (315) nm; HR-ESIMS: *m/z* 203.7187 $[M\text{-}3ClO_4]^{+3}$calcd. for $(C_{30}H_{24}CoN_{12})^{+3}$: 203.7193. Elemental Anal. Calcd. for $C_{30}H_{26}CoN_{12}Cl_3O_{13}$: C, 38.83; H, 2.82; N, 18.11. Found: C, 39.02; H, 2.62; N, 18.24.

Preparation and characterization of $[Co(L^{O2})_2](NO_3)$. A solution of $Co(NO_3)_2\cdot 6H_2O$ (124.3 mg, 0.43 mmol) in methanol (15 mL) was placed in a 25 mL round-bottom flask equipped with a magnetic stirrer. L^{A2} (240.5mg, 0.86 mmol) was then added. The reaction mixture was stirred at room temperature for 4 hours. Upon completion, the solvent was removed by rotary evaporation. Acetone (15 mL) was then added to the residue, inducing the precipitation of the crude product, **$[Co(L^{O2})_2](NO_3)$**. The solid was washed with CH_2Cl_2 and Et_2O to afford the desired product as a pink powder (166.4 mg, 74 yield). ^{1}H NMR (300 MHz, CD_3CN) : δ 8.69

(dd, $J_{H\text{-}H}$ = 3.0, 2.1 Hz, 2H), 8.38 (d, $J_{H\text{-}H}$ = 5.7 Hz, 2H), 8.26 (dd, $J_{H\text{-}H}$ = 2.3, 2.1 Hz, 2H), 7.90 (td, $J_{H\text{-}H}$ = 1.5, 7.7 Hz, 2H), 7.77 (s, 2H), 7.53 (d, $J_{H\text{-}H}$ = 7.8 Hz, 2H), 7.32 (td, $J_{H\text{-}H}$ = 1.4, 6.6 Hz, 2H), 6.66 (dd, $J_{H\text{-}H}$ = 3.0, 4.4 Hz, 2H), 5.65 (d, $J_{H\text{-}H}$ =4.1 Hz, 2H). Recrystallization via slow diffusion of CH_2Cl_2 into a methanol solution provided single crystals suitable for X-ray diffraction. UV-Vis (MeOH): λ_{max} (ε in $M^{-1}\cdot cm^{-1}$) = 514 (75) nm. HR-ESIMS: *m/z* 461.0872 $[M\text{-}NO_3]^+$calcd. for $(C_{20}H_{18}CoN_8O_2)^+$: 461.0885. Elemental Anal. Calcd. for $C_{20}H_{18}CoN_9O_5$: C, 45.90; H, 3.47; N, 24.09. Found: C, 45.70; H, 3.32; N, 24.33.

Preparation and characterization of $[Cu(L^{O1Me})_2](ClO_4)_2$. A solution of $Cu(ClO_4)_2\cdot 6H_2O$ (145.8 mg, 0.39 mmol) in methanol (15 mL) was placed in a 25 mL round-bottom flask equipped with a magnetic stir bar. L^{A1} (218.4 mg, 0.78 mmol) was then added, and the reaction mixture was stirred at room temperature for 4 hours. Upon completion, the reaction yielded a clear green solution. The solvent was removed by rotary evaporation, and CH_2Cl_2 (15 mL) was added to the residue, inducing the precipitation of the crude product, **$[Cu(L^{O1Me})_2](ClO_4)_2$**. The solid was washed three times with CH_2Cl_2 and Et_2O, affording the desired product as a green powder (234.1mg, 86 % yield). UV-Vis (MeOH): λ_{max} (ε in $M^{-1}\cdot cm^{-1}$) = 633 (48) nm; UV-Vis (CH_3CN): λ_{max} (ε in $M^{-1}\cdot cm^{-1}$) = 689 (72) nm. The ESIMS and elemental analysis could not be obtained because the product is dynamic and unstable.

Preparation and characterization of $[Cu(L^{O2Me})_2](ClO_4)_2$. A solution of $Cu(ClO_4)_2\cdot 6H_2O$ (219.5 mg, 0.59 mmol) in methanol (15 mL) was placed in a 25 mL round-bottom flask equipped with a magnetic stir bar. L^{A2} (330.1 mg, 1.18 mmol) was then added, and the reaction mixture was stirred at room temperature for 4 hours. Upon completion, the reaction yielded a clear blue solution. The solvent was removed by rotary evaporation, and CH_2Cl_2 (15 mL) was added to the residue, inducing the precipitation of the crude product, **$[Cu(L^{O2H})_2](ClO_4)_2$**. The solid was washed three times with CH_2Cl_2 and Et_2O, affording the desired product as a blue powder (321.7 mg, 81% yield). UV-Vis (MeOH): λ_{max} (ε in $M^{-1}\cdot cm^{-1}$) = 656 (144) nm. The HR-ESIMS: *m/z* 594.0795 $[M\text{-}ClO_4]^+$calcd. for $(C_{22}H_{24}N_8\,ClO_6Cu)^+$: 594.0803. and elemental analysis could not be obtained because the product is dynamic and unstable.

Preparation and characterization of $[Cu(L^{O2H})_2](ClO_4)_2$. A solution of $Cu(ClO_4)_2\cdot 6H_2O$ (200.6 mg, 0.54 mmol) in acetonitrile (15 mL) was placed in a 25 mL round-bottom flask equipped with a magnetic stir bar. L^{A2} (302.5 mg, 1.08 mmol) was then added, and the reaction mixture was stirred at room temperature for 4 hours. Upon completion, the reaction yielded a clear blue solution. The solvent was removed by rotary evaporation, and CH_2Cl_2 (15 mL) was added to the residue, inducing the precipitation of the crude product, **$[Cu(L^{O2H})_2](ClO_4)_2$**. The solid was washed three times with CH_2Cl_2 and Et_2O, affording the desired product as a blue powder (290.0 mg, 80% yield). UV-Vis (CH_3CN): λ_{max} (ε in $M^{-1}\cdot cm^{-1}$) = 689 (89) nm; HR-ESIMS: *m/z* 432.0872 243.04 $[M\text{-}H_2O\text{-}OH\text{-}2ClO_4]^+$ calcd. for $(C_{20}H_{17}N_8Cu)^+$: 432.0842. Elemental Anal. Calcd. for $C_{20}H_{20}CuN_8O_{10}Cl_2$: C, 36.02; H, 3.02; N, 16.80. Found: C, 35.88; H, 2.91; N, 17.12.

Preparation and characterization of *cis*-[4^{pym}](ClO$_4$). A solution of $Fe(ClO_4)_3 \cdot 6H_2O$ (119.2 mg, 0.258 mmol) in acetonitrile (15 mL) was placed in a 25 mL round-bottom flask equipped with a magnetic stirrer. L^{A2} (216.2 mg, 0.772 mmol) was then added, and the reaction mixture was stirred and heated to reflux for 3 hours. After completion, the mixture was cooled to room temperature, and the solvent was removed by rotary evaporation. Dichloromethane (15 mL) was then added to the residue, inducing the precipitation of the Fe(III)-containing product. The precipitate was filtered off, and the volatiles were removed by rotary evaporation. Ethanol was then added to the residue, leading to precipitation, followed by washing with diethyl ether three times to afford desired pale yellow solid, **[4^{pym}](ClO$_4$)** (73.2 mg, 40% yield). Recrystallization via slow diffusion of diethyl ether into an acetonitrile solution provided single crystals suitable for X-ray diffraction analysis. ^{1}H NMR (400 MHz, DMSO-d_6): δ 9.42 (d, $J_{H\text{-}H}$ = 6.4 Hz, 1H), 8.72 (d, $J_{H\text{-}H}$ = 4.8 Hz, 1H), 8.64 (dd, $J_{H\text{-}H}$ = 8.0, 8.0 Hz, 1H), 8.58 (d, $J_{H\text{-}H}$ = 4.8 Hz, 2H), 8.33 (d, $J_{H\text{-}H}$ = 7.20 Hz, 1H), 8.22 (d, $J_{H\text{-}H}$ = 8.0 Hz, 1H), 8.17 (dd, $J_{H\text{-}H}$ = 6.4, 8.0 Hz, 1H), 8.05 (br, 2H), 7.86 (dd, $J_{H\text{-}H}$ = 8.0, 8.0 Hz, 1H), 7.82 (s, 1H, *CH*), 7.68 (d, $J_{H\text{-}H}$ = 8.00 Hz, 1H), 7.61 (d, $J_{H\text{-}H}$ = 7.20 Hz, 1H), 7.50 (dd, $J_{H\text{-}H}$ = 4.8, 8.0 Hz, 1H), 7.06 (t, $J_{H\text{-}H}$ = 4.8 Hz, 1H), 6.69 (t, $J_{H\text{-}H}$ = 4.8 Hz, 1H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ 159.9, 159.0, 158.6, 157.8, 153.6, 153.6, 149.4, 147.5, 139.9, 137.7, 127.0, 124.8, 123.3, 122.4, 114.5, 112.6, 82.6, 69.5 ppm. Elemental Anal. Calcd. for $C_{20}H_{17}N_8O_4Cl$: C, 51.24; H, 3.65; N, 23.90. Found: C, 51.08; H, 3.32; N, 24.02.

Demetalation reaction of [Co(L^M)](ClO$_4$)$_3$. A solution of **[Co(L^M)](ClO$_4$)$_3$** (39.5 mg, 0.043 mmol) in water (10 mL) was placed in a 25 mL round-bottom flask equipped with a magnetic stirrer. $NaBH_4$ (34.7 mg, 0.917 mmol) was then added to the flask, and the mixture was stirred at room temperature for 15 minutes. Subsequently, NaCN (74.7 mg, 1.52 mmol) was added, and the reaction was continued at room temperature for an additional hour. Upon completion, the mixture was extracted with CH_2Cl_2 (3 × 20 mL), and the combined organic layers were dried over $MgSO_4$. After filtration, the solvent was removed by rotary evaporation, affording L^{A2}-re as a pale-yellow solid (10.1 mg, 42%). ^{1}H NMR of **L^{A2}-re** (400 MHz, $CDCl_3$): δ 8.57 (d, $J_{H\text{-}H}$ = 4.00 Hz, 1H), 8.31 (d, $J_{H\text{-}H}$ = 4.00 Hz, 2H), 7.64 (dd, $J_{H\text{-}H}$ = 4.00, 8.00 Hz, 1H), 7.31 (d, $J_{H\text{-}H}$ = 8.00 Hz, 1H), 7.18 (dd, $J_{H\text{-}H}$ =8.00, 8.00 Hz, 1H), 6.55 (t, $J_{H\text{-}H}$ =4.00 Hz, 1H), 4.75 (s, 2H). ^{13}C NMR of **6c** (100 MHz, $CDCl_3$): δ 162.3, 158.3, 157.9, 149.3, 136.7, 122.3, 121.7, 111.0, 46.6 ppm. HR-ESIMS: *m/z* 187.0985 [M+H$^+$] calcd. for $(C_{10}H_{11}N_4)^+$: 187.0984.

Crystallographic Data and the Parameters of the Single Crystal X-Ray Diffraction

Table S1 Crystal data and structure refinements for $[Co(L^{O1})_2](ClO_4)$, $[Co(L^{O2})_2](ClO_4)$ and $[Co(L^{O2Me})_2](ClO_4)_2$

	$[Co(L^{O1})_2](ClO_4)$	**$[Co(L^{O2})_2](ClO_4)$**	**$[Co(L^{O2Me})_2](ClO_4)_2$**
Formula	$C_{22}H_{20}ClCoN_6O_6$	$C_{20}H_{18}ClCoN_8O_6$	$C_{22}H_{24}Cl_2CoN_8O_{10}$ Cl
Crystal system	Orthorhombic	Monoclinic	Triclinic
Space group	*P*ca2$_1$	*P21/c*	*P -1*
***a, b, c* (Å)**	14.935(3), 8.6856(17), 17.997(4)	8.6366(2), 18.5010(3), 17.997(4)	9.0344(3), 9.3458(3), 9.7176(3)
α, β, γ (°)	90, 90, 90	90, 92.2990(10), 90	111.912(1), 106.149(1), 98.145(1)
Temperature (K)	295(2)	295(2)	100 K
Volume (Å^3), Z	2334.7(8), 4	2253.78(8), 4	702.89(4), 1
Final *R* indices [i>2 sigma(i)]	R_1=0.0828, wR_2=0.2097	R1=0.0499, wR2=0.1294	R_1=0.0423, wR_2=0.1161
***R* indices (all data)**	R_1=0.1066, wR_2=0.2302	R1=0.0630, wR2=0.1473	R_1=0.0435, wR_2=0.1164

Table S2 Crystal data and structure refinements for $[Co(L^{M})](ClO_4)_3$, $[Co(L^{O2})_2](NO_3)$ major and $[Co(L^{O2})_2](NO_3)$ minor

	$[Co(L^{M})](ClO_4)_3$	**$[Co(L^{O2})_2](NO_3)$ major**	**$[Co(L^{O2})_2](NO_3)$ minor**
Formula	$C_{30}H_{24}Cl_3CoN_{12}O_{12}$	$C_{20}H_{18}CoN_9O_5$	$C_{20}H_{18}CoN_9O_5$
Crystal system	Orthorhombic	Triclinic	Trigonal
Space group	$P2_12_12_1$	*P -1*	*R-3c*
***a, b, c* (Å)**	13.266(3), 15.857(3) , 20.048(4)	8.4274(3), 9.5402(3), 15.1235(6)	24.4927(7),24.4927(7), 20.8894(7)
α, β, γ (°)	90, 90, 90	72.323(2), 86.796(2), 78.310(2)	90, 90, 120
Temperature (K)	200(2)	100.0(2)	100.00(10)
Volume (Å^3), Z	4217.3(15), 4	1134.45(7), 2	10852.5(7), 6
Final *R* indices [i>2 sigma(i)]	R1 = 0.0539, wR2 = 0.1323	R1 = 0.0525, wR2 = 0.1437	R_1=0.0638, wR_2=0.1611
***R* indices (all data)**	R1 = 0.1114, wR2 = 0.1769	R1 = 0.0565, wR2 = 0.1468	R_1=0.0672, wR_2=0.1632

Table S3 Crystal data and structure refinements for **$[Cu(L^{O2H})_2](ClO_4)_2$** and $[Cu(L^{O2Me})_2](ClO_4)_2$ and $[Cu(L^{O1Me})_2](ClO_4)_2$

	$[Cu(L^{O2H})_2](ClO_4)_2$	**$[Cu(L^{O2Me})_2](ClO_4)_2$**	**$[Cu(L^{O1Me})_2](ClO_4)_2$**
Formula	$C_{20}H_{22}Cl_2CuN_8O_{12}$	$C_{22}H_{25}Cl_2CuN_8O_{10}$	$C_{24}H_{26}Cl_2CuN_6O_{10}$
Crystal system	Triclinic	Monoclinic	Monoclinic
Space group	*P*-1	$P2_1/c$	$P2_1/c$
***a, b, c* (Å)**	8.1805(6),8.9423(6), 9.7820(8)	12.4953(2),18.2777(3), 12.5438(2)	12.0096(2),18.7829(3), 12.7611(2)
α, β, γ (°)	106.172(2),94.489(2), 92.988(2)	90, 105.642(2), 90	90, 104.737(2), 90
Temperature (K)	200(2)	100.00(10)	200(2)
Volume (Å^3), Z	683.11(9), 1	2758.72(8), 4	2783.89(8), 4
Final *R* indices [i>2 sigma(i)]	R_1=0.0581, wR_2=0.1534	R1 = 0.0541, wR2 = 0.1371	R_1=0.0866, wR_2=0.1746
***R* indices (all data)**	R_1=0.0808, wR_2=0.1352	R1 = 0.0581, wR2 = 0.1399	R_1=0.0875, wR_2=0.1750

Table S4 Crystal data and structure refinements for $Co(L^{O2})_2$ and *cis*-$[4^{pym}](ClO_4)$

	***cis*-$[4^{pym}](ClO_4)$**	**$[Co(L^{O2})_2]$**	
Formula	$C_{20}H_{17}ClN_8O_4$	$C_{20}H_{22}CoN_8O_4$	
Crystal system	Orthorhombic	Trigonal	
Space group	*P*ccn	*R*-3c	
***a, b, c* (Å)**	15.2487(7), 22.7472(7), 12.0738(6)	24.5386(7), 24.5386(7), 20.9738(5)	
α, β, γ (°)	90, 90, 90	90, 90, 120	
Temperature (K)	200(2)	295(2)	
Volume (Å^3), Z	4188.0(3), 8	11113.6(7), 18	
Final *R* indices [i>2 sigma(i)]	R_1=0.0492, wR_2=0.099	R_1=0.0446, wR_2=0.1443	
***R* indices (all data)**	R_1=0.0824, wR_2=0.1186	R_1=0.0551, wR_2=0.1550	

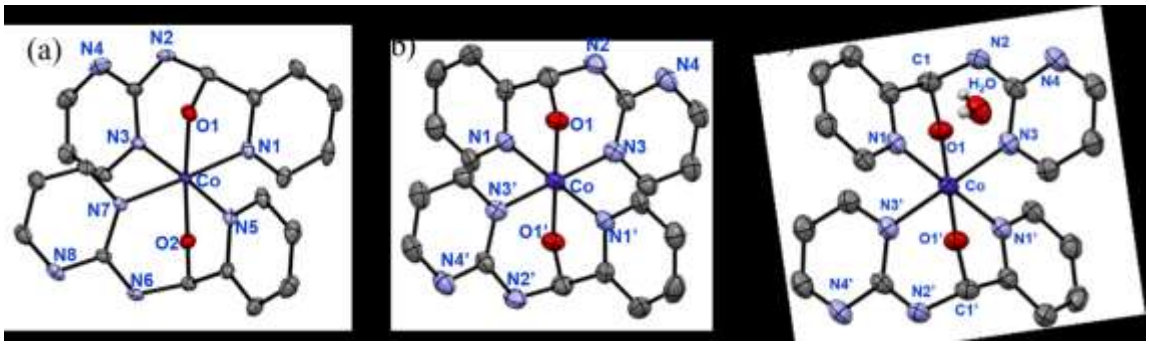

Fig. S1. ORTEP drawings of (a) $[Co(L^{O2})_2](NO_3)$, (b) iso1-$[Co(L^{O2})_2](NO_3)$ and (c) $[Co(L^{O2})_2]$. The counteranion (NO_3^-) and hydrogen atoms are omitted for clarity. Thermal ellipsoids are displayed at 50 % probability level. Selected bond lengths (Å) and bond angles (°) for (a) $[Co(L^{O2})_2](NO_3)$: Co-O1: 1.899(2); Co-O2: 1.869(2); Co-N1: 1.923(2); Co-N5: 1.926(3); Co-N3: 1.957(2); Co-N7: 1.976(2); N1-Co-O1: 83.75(10); N3-Co-O1: 89.50(9); N(5)-Co-O(1): 94.40(10); N(7)-Co-O(1): 94.53(9); N(1)-Co-O(2): 91.98(10); N(3)-Co-O(2): 92.21(9); N(7)-Co-O(2): 89.78(9); N1-Co-N3: 90.87(10); N(1)-Co-N(5); 90.48(10); N(3)-Co-N(7): 87.69(9); N(5)-Co-N(7): 91.07(10); O1-Co-O2 175.43(8), and (b) iso1-$[Co(L^{O2})_2](NO_3)$: Co-O1: 1.880(3); Co-N1: 1.930(3); Co-N3: 1.971(3); N1-Co-O1: 84.59(13); N3-Co-O1: 88.51(12). (c) $[Co(L^{O2})_2]$: Co-O1: 1.886 (2); Co-N1: 1.940(4); Co-N4: 1.989(3); N1-Co-N4: 90.77(10); O1-Co-N1: 95.98(10); O1-Co-N4 88.65(10). The O1…H_{H2O} is 1.819 Å.

- **UV-vis spectra**

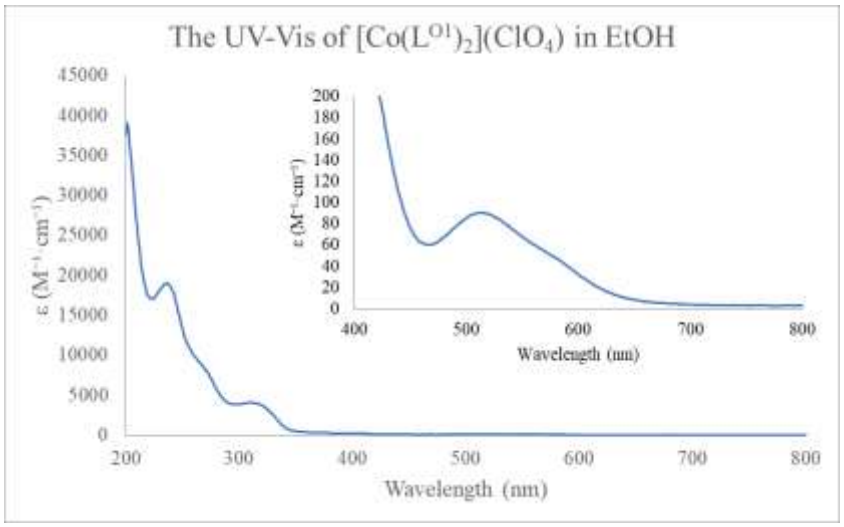

Fig. S2. The UV-Vis absorption spectrum of $[Co(L^{O1})_2](ClO_4)$ in EtOH. *Inset:* enlarged view showing the d–d transition bands.

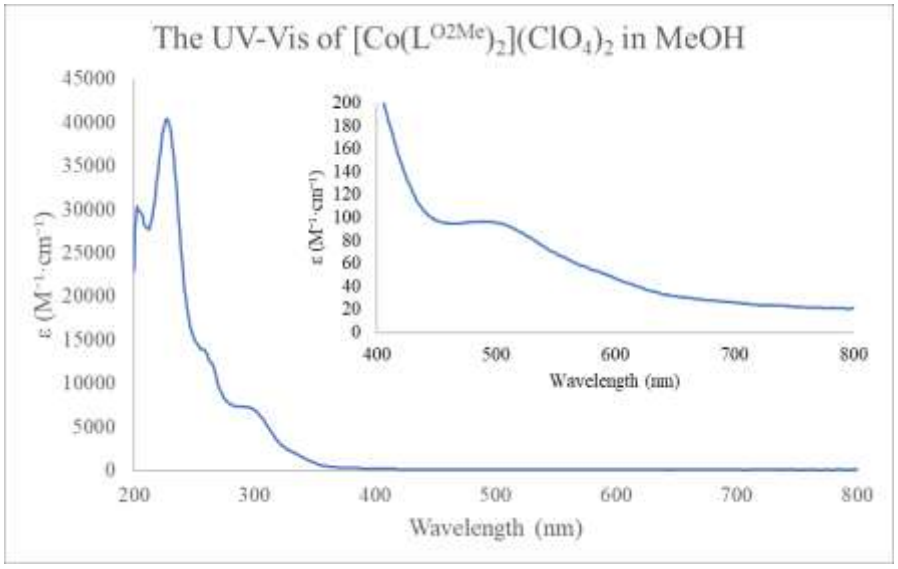

Fig. S3. The UV-Vis absorption spectrum of $[Co(L^{O2Me})_2](ClO_4)_2$ in MeOH *Inset:* enlarged view showing the d–d transition bands.

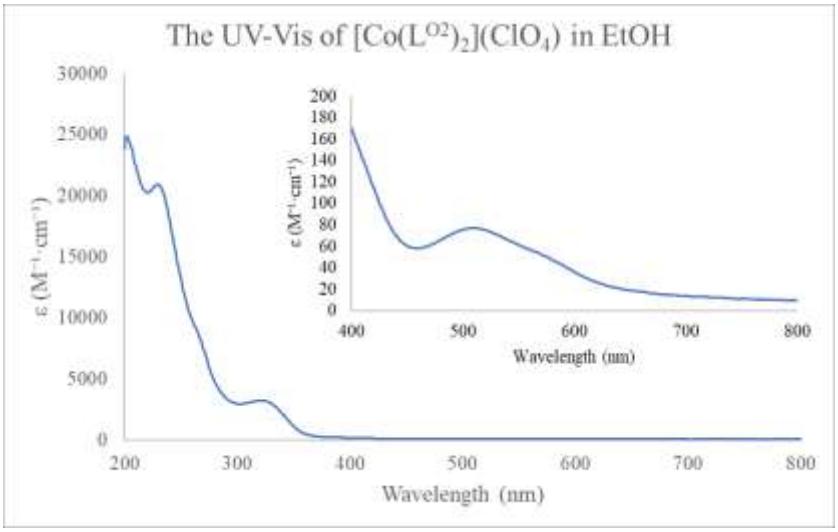

Fig. S4. The UV-Vis absorption spectrum of $[Co(L^{O2})_2](ClO_4)$ in EtOH. *Inset:* enlarged view showing the d–d transition bands.

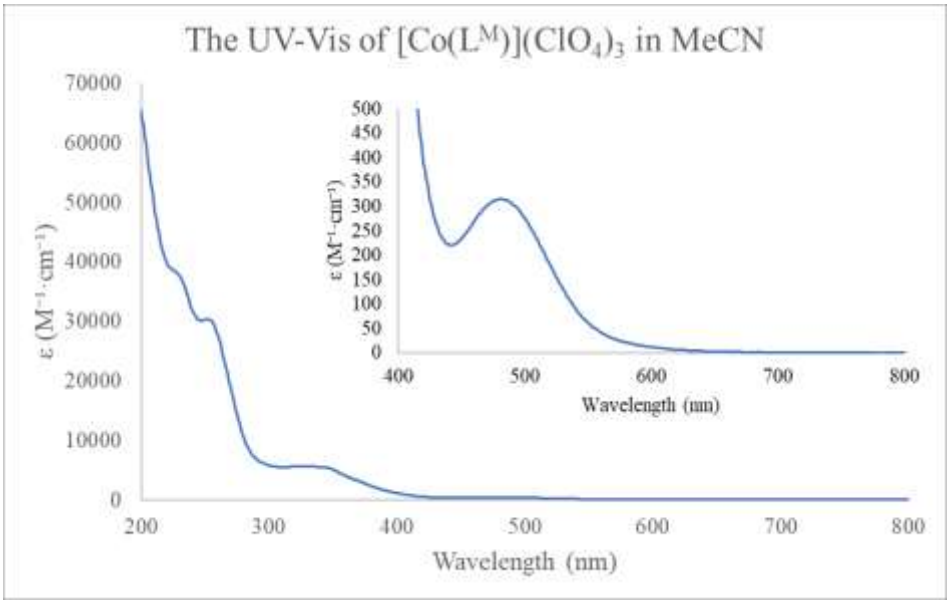

Fig. S5. The UV-Vis absorption spectrum of $[Co(L^{M})](ClO_4)_3$ in MeCN. *Inset:* enlarged view showing the d–d transition bands.

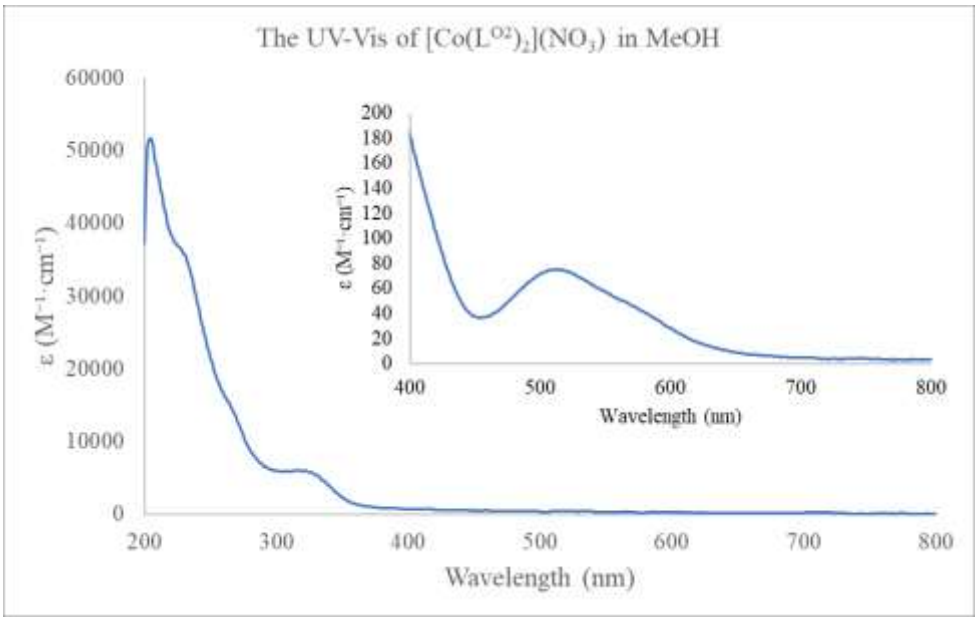

Fig. S6. The UV-Vis absorption spectrum of UV-Vis of $[Co(L^{O2})_2](NO_3)$ in MeOH. *Inset:* enlarged view showing the d–d transition bands.

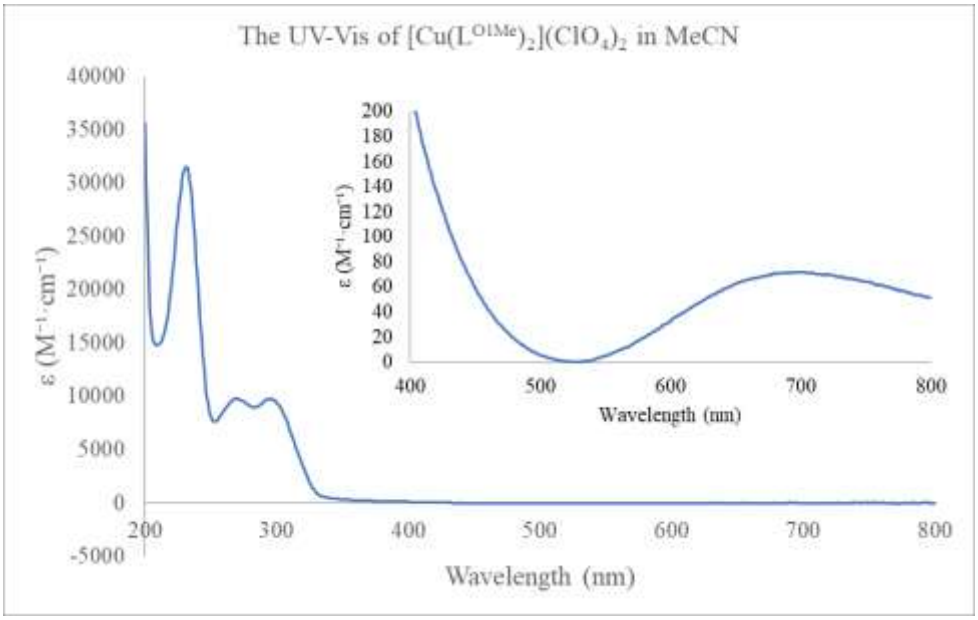

Fig. S7. The UV-Vis absorption spectrum of $[Cu(L^{O1Me})_2](ClO_4)_2$ in MeCN. *Inset:* enlarged view showing the d–d transition bands.

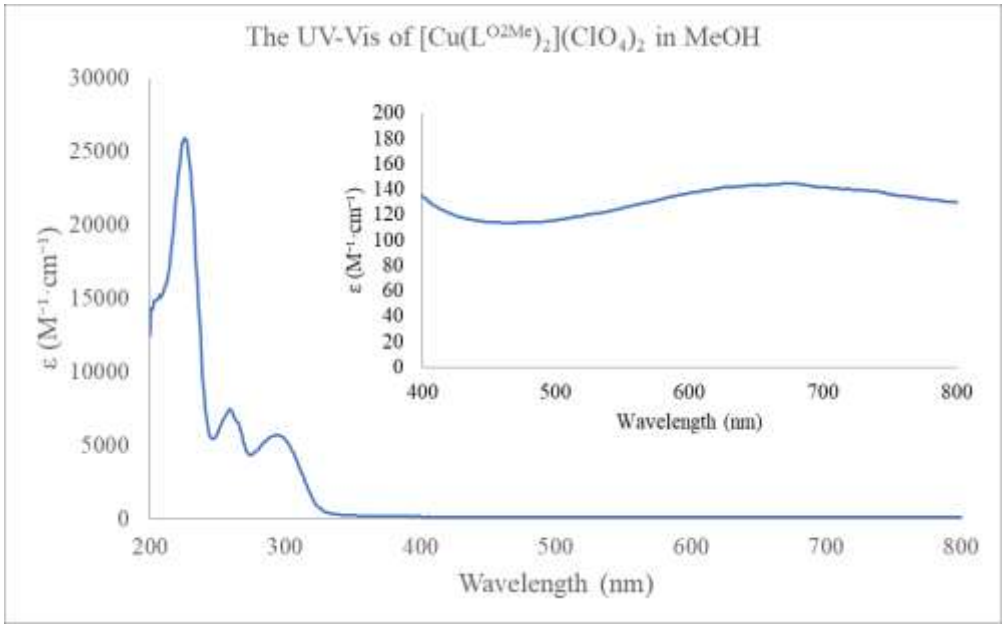

Fig. S8. The UV-Vis absorption spectrum of $[Cu(L^{OMe2})_2](ClO_4)_2$ in MeOH. *Inset:* enlarged view showing the d–d transition bands.

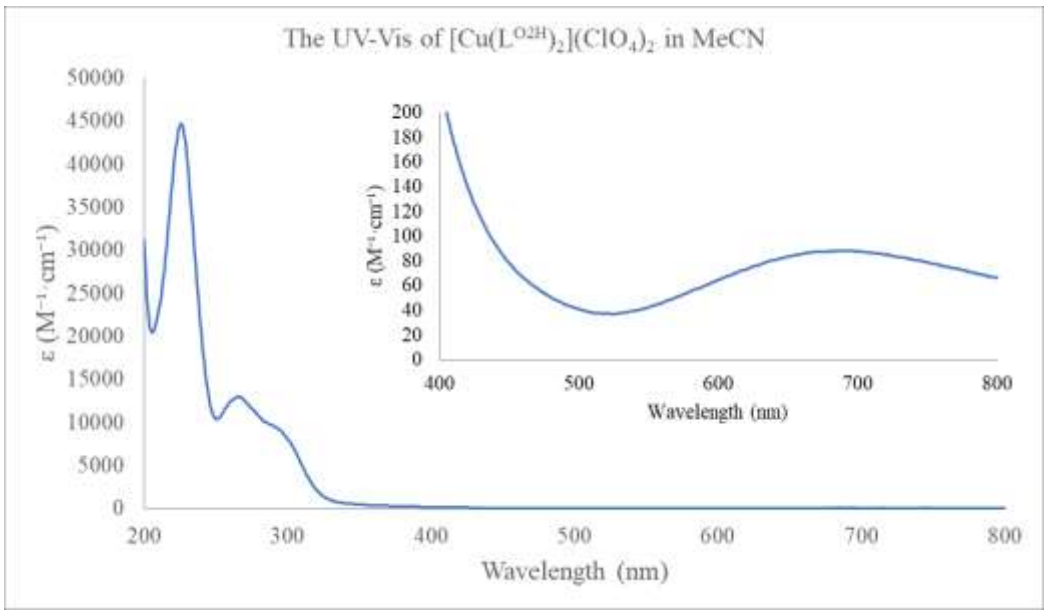

Fig. S9. The UV-Vis absorption spectrum of $[Cu(L^{O2H})_2](ClO_4)_2$ in MeCN. *Inset:* enlarged view showing the d–d transition bands.

NMR spectra

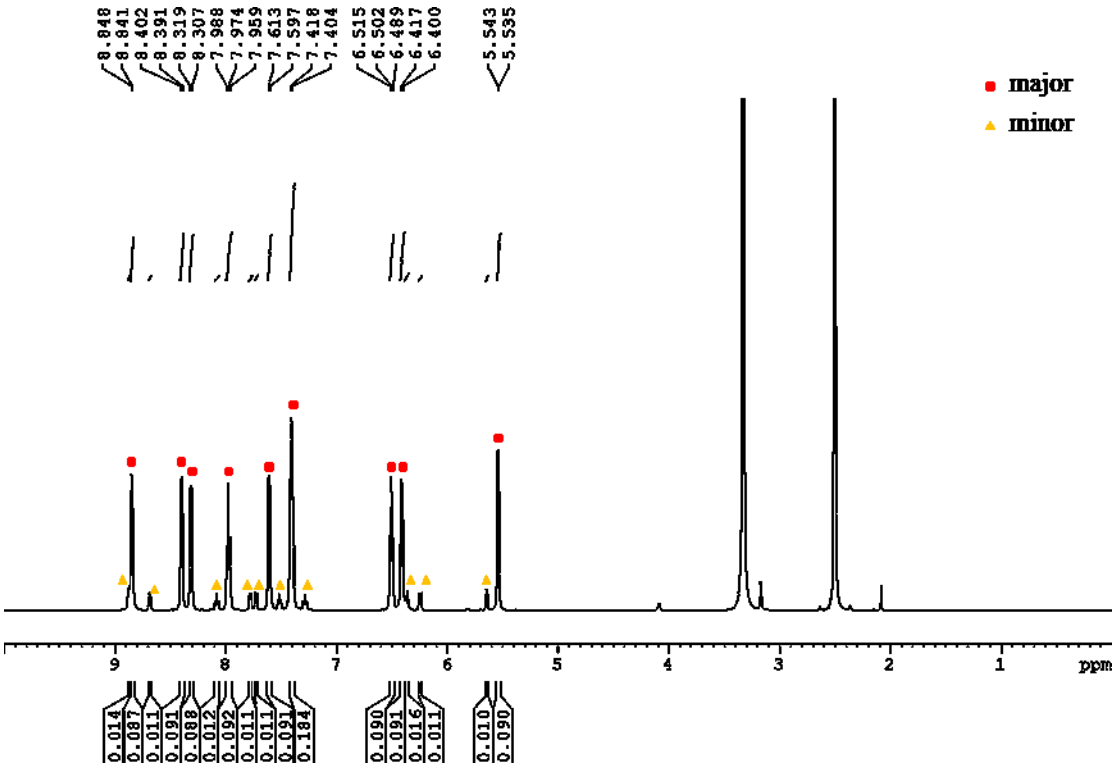

Fig. S10. ^{1}H NMR spectrum of $[Co(L^{O1})_2](ClO_4)$ in DMSO-d_6 at 25 °C

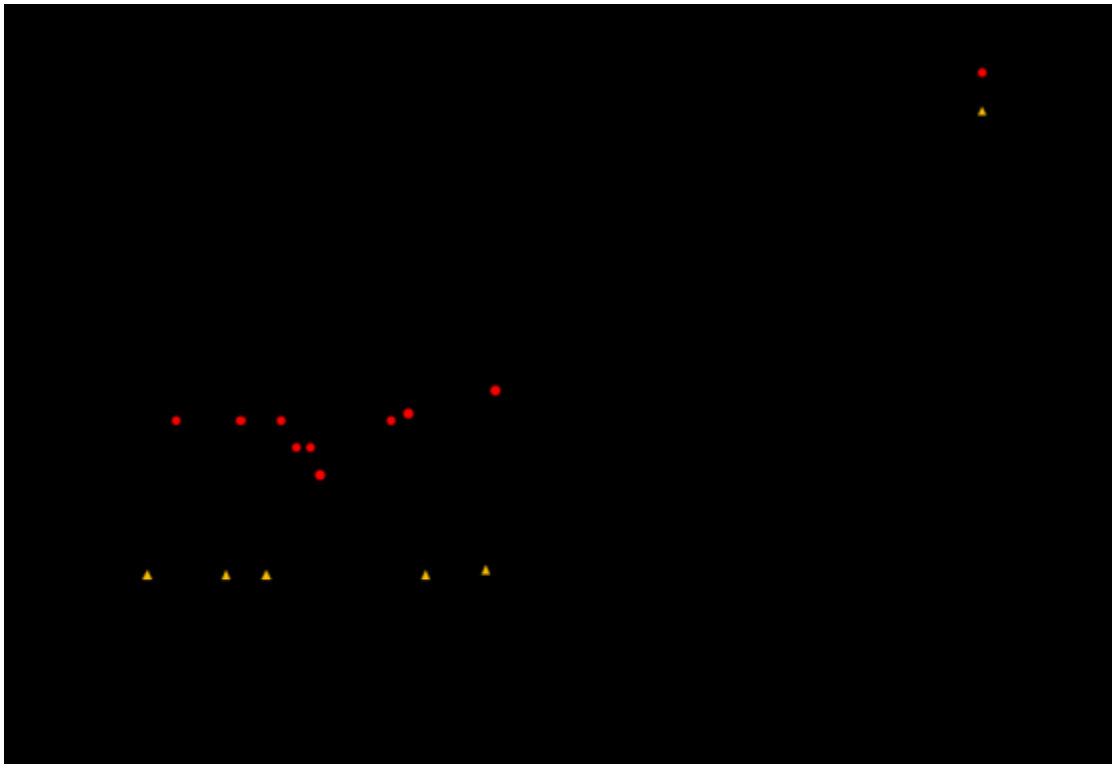

Fig. S11. ^{1}H NMR spectrum of $[Co(L^{O1})_2](ClO_4)$ in CD_3CN at 25 °C

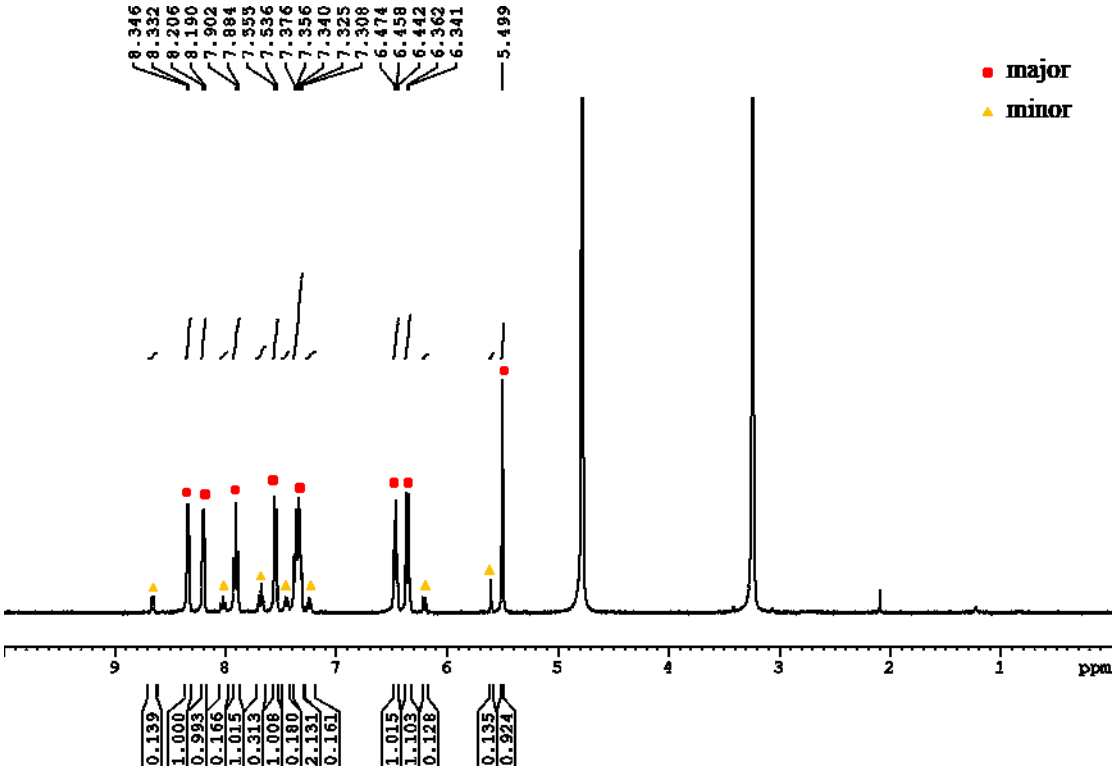

Fig. S12 ^{1}H NMR spectrum of $[Co(L^{O1})_2](ClO_4)$ in CD_3OD at 25 °C

Fig. S13 ^{13}C NMR spectrum of $[Co(L^{O1})_2](ClO_4)$ in DMSO-d_6 at 25 °C

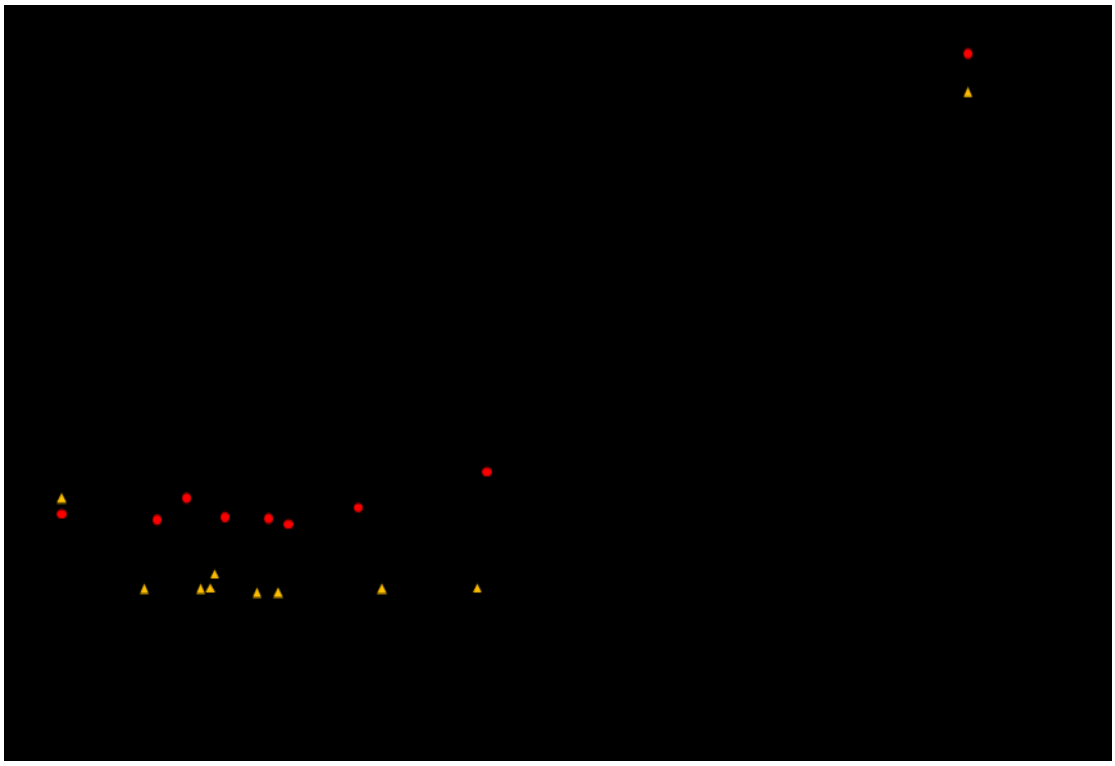

Fig. S14. ^{1}H NMR spectrum of $[Co(L^{O2})_2](ClO_4)$ in DMSO-d_6 at 25 °C

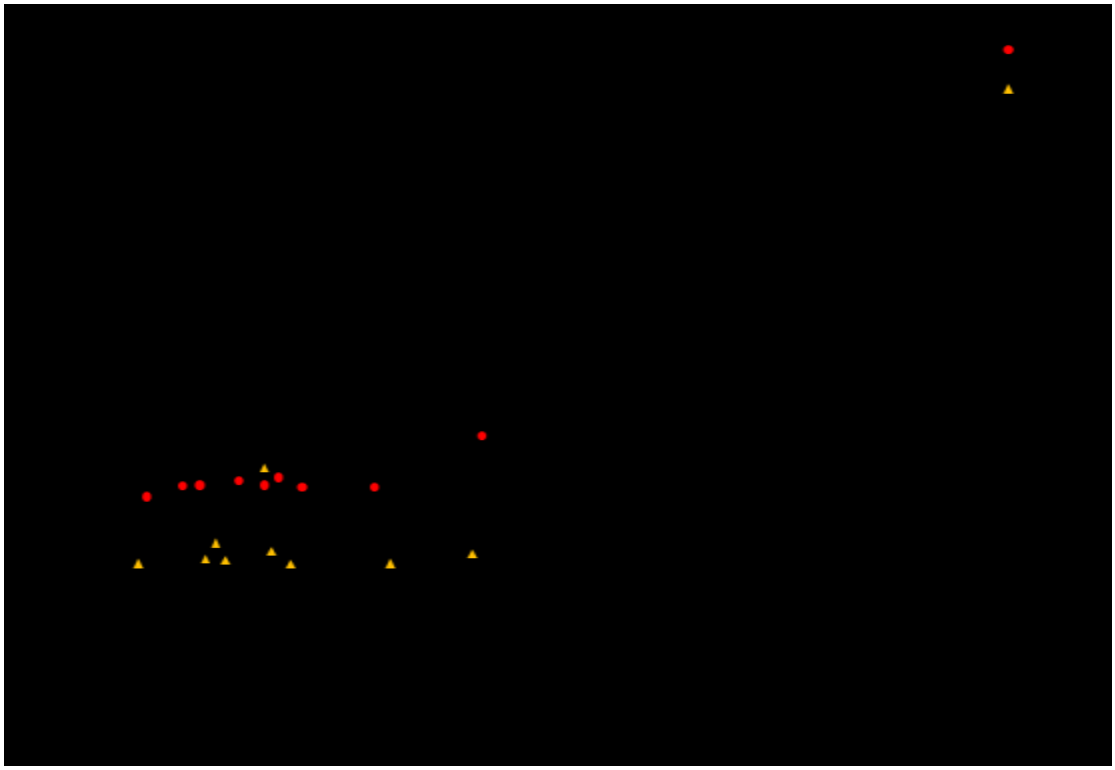

Fig. S15. ^{1}H NMR spectrum of $[Co(L^{O2})_2](ClO_4)$ in CD_3CN at 25 °C

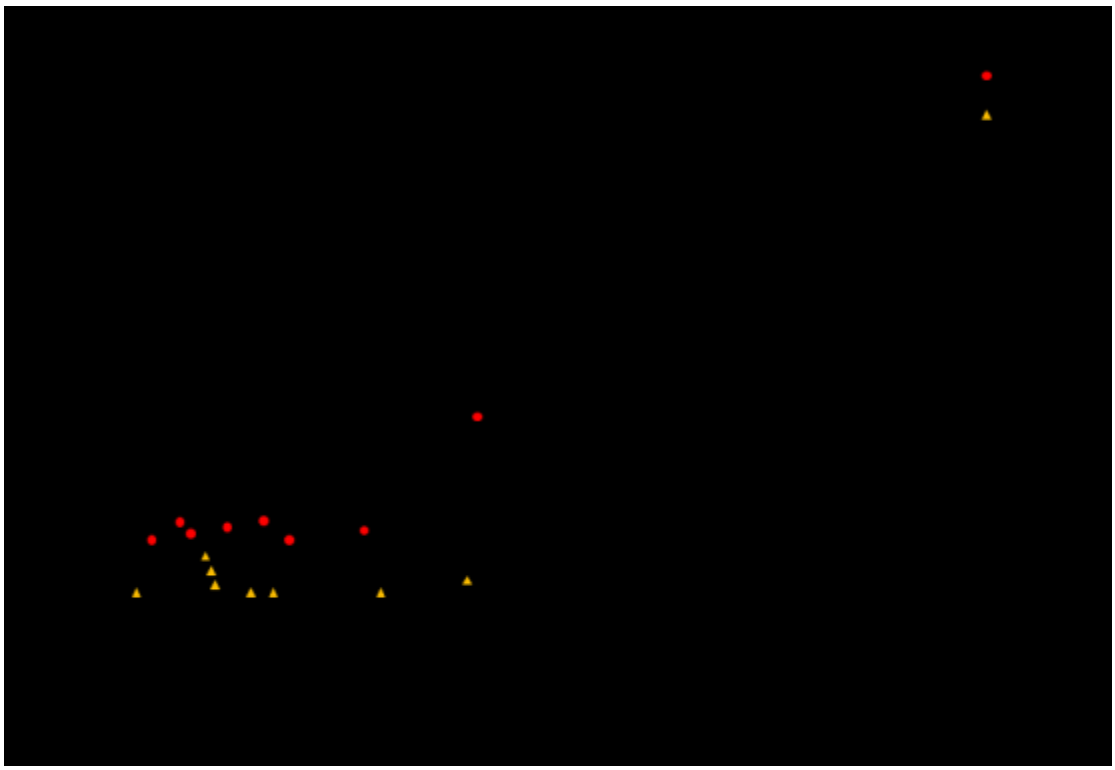

Fig. S16 ^{1}H NMR spectrum of $[Co(L^{O2})_2](ClO_4)$ in CD_3OD at 25 °C

Fig. S17 ^{13}C NMR spectrum of $[Co(L^{O2})_2](ClO_4)$ in DMSO-d_6 at 25 °C

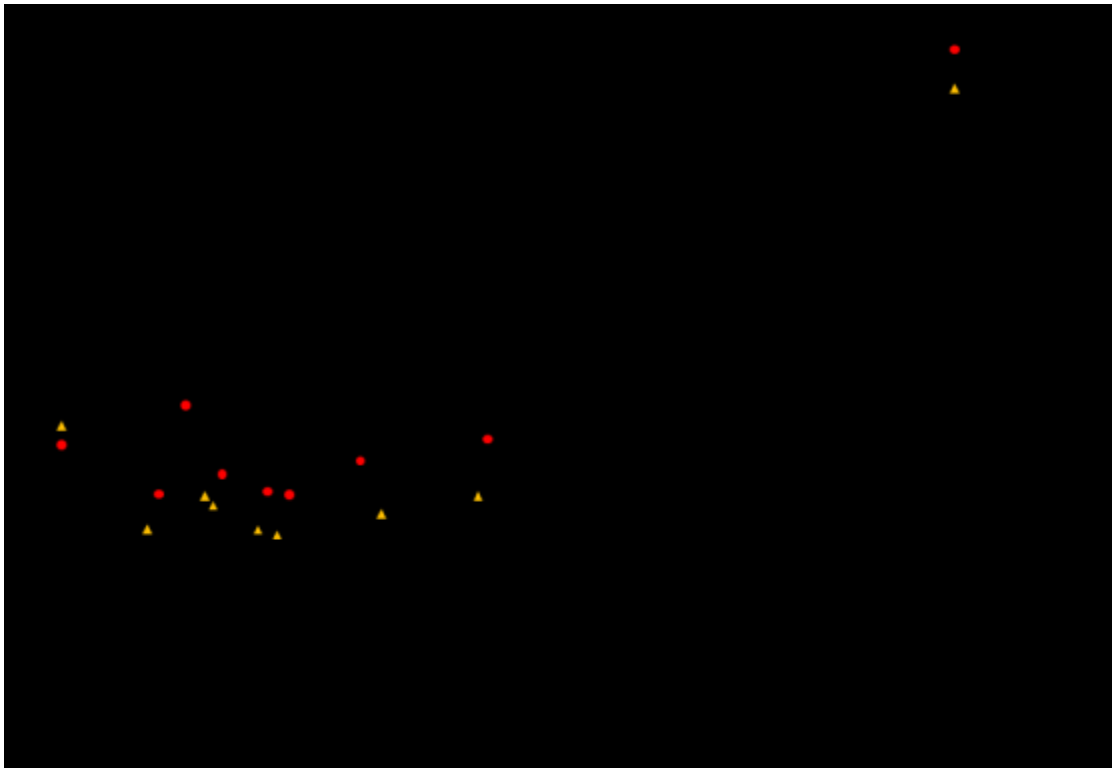

Fig. S18 ^{1}H NMR spectrum of $[Co(L^{O2})_2](NO_3)$ in DMSO-d_6 at 25 °C

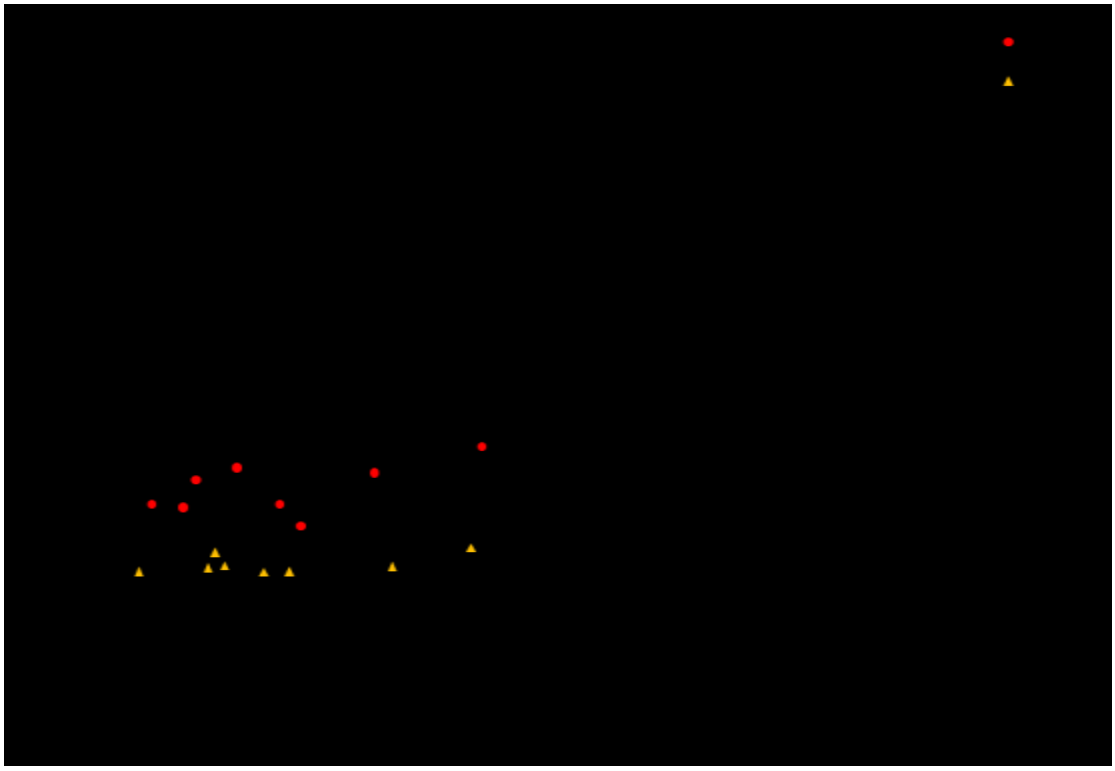

Fig. S19 ^{1}H NMR spectrum of $[Co(L^{O2})_2](NO_3)$ in CD_3CN at 25 °C

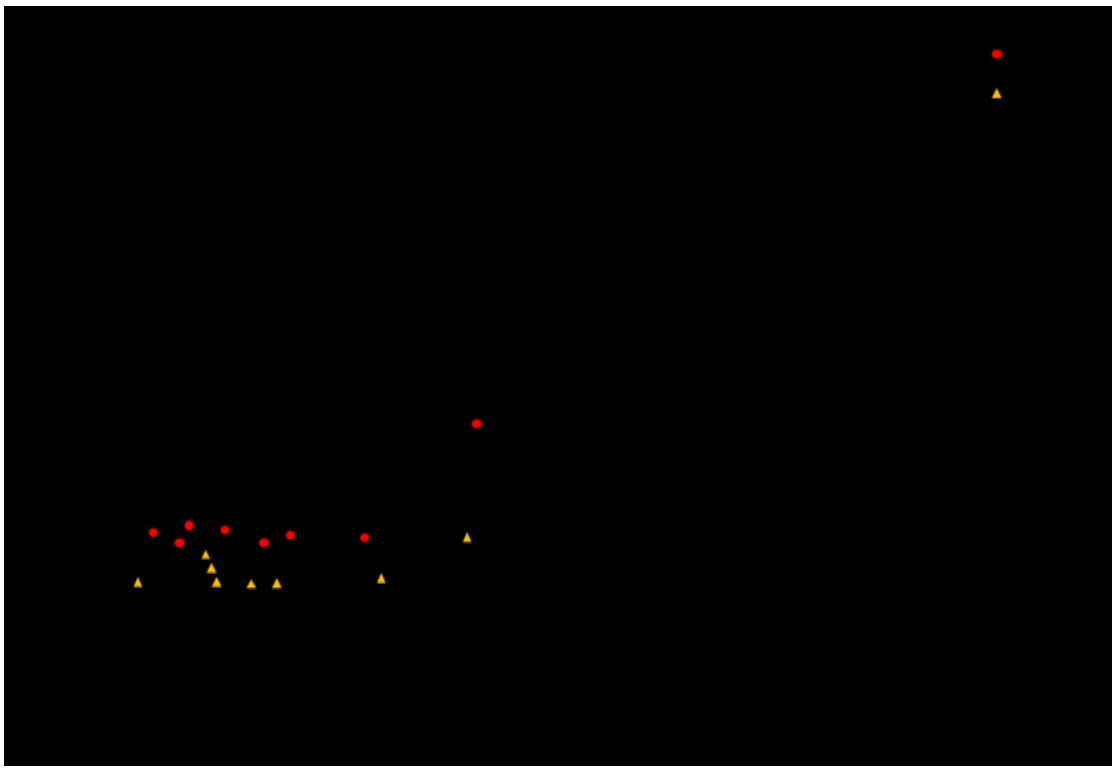

Fig. S20 ^{1}H NMR spectrum of $[Co(L^{O2})_2](NO_3)$ in CD_3OD at 25 °C

The VT-NMR Spectroscopic Studies of $[Co(L^{M})](ClO_4)_3$ in DMSO-d_6

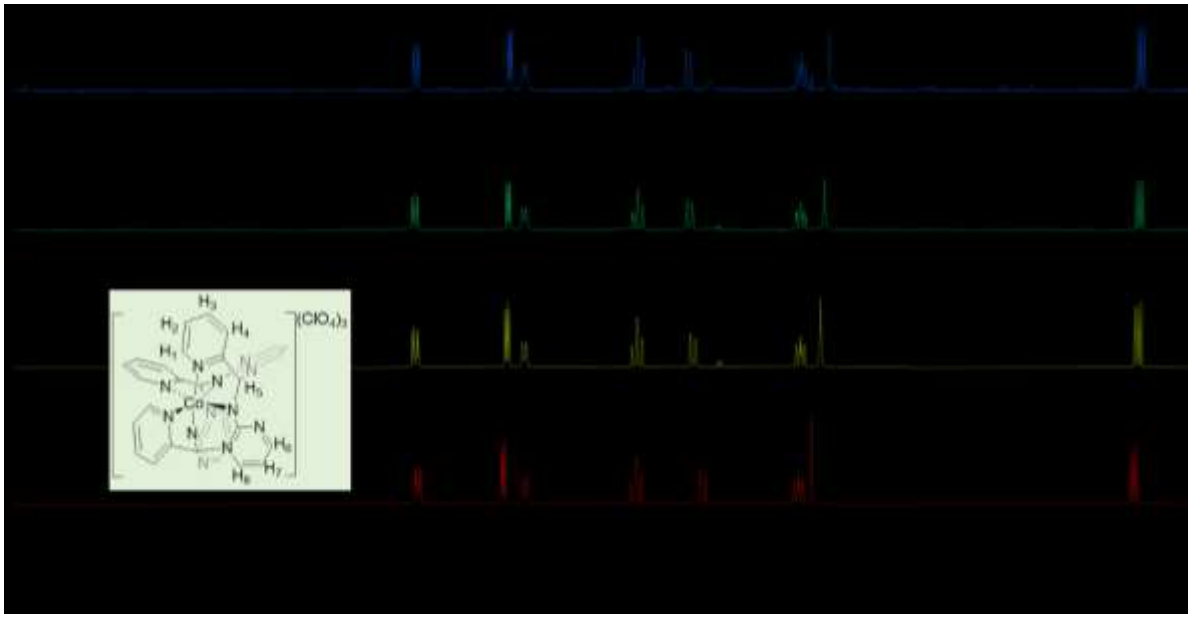

Fig. S21. Variable-temperature ^{1}H NMR spectra of $[Co(L^{M})](ClO_4)_3$ recorded in DMSO-d_6 from 273 K to 403 K. The signals marked with dots correspond to decomposed peaks.

Cyclic voltammogram of $[Co(L^M)](ClO_4)_3$

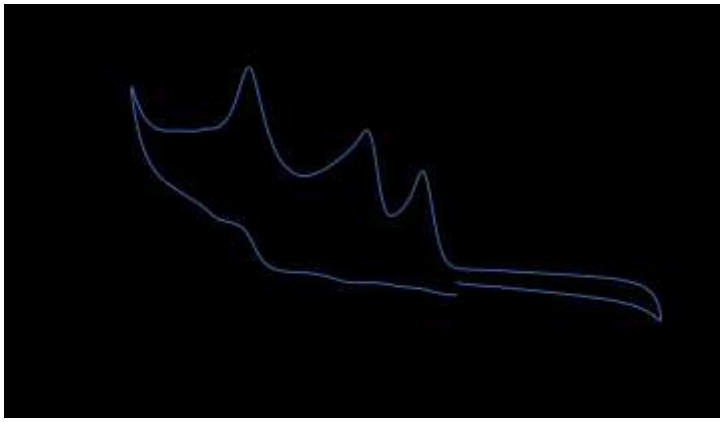

Fig. S22. The cyclic voltammogram of 10 mM $[Co(L^M)](ClO_4)_3$ measured in DMSO with 0.1 M $TBAClO_4$ as a supporting electrolyte at a scan rate 0.05 mV s^{-1} scanned in the positive direction.

HR-ESI-MS spectra

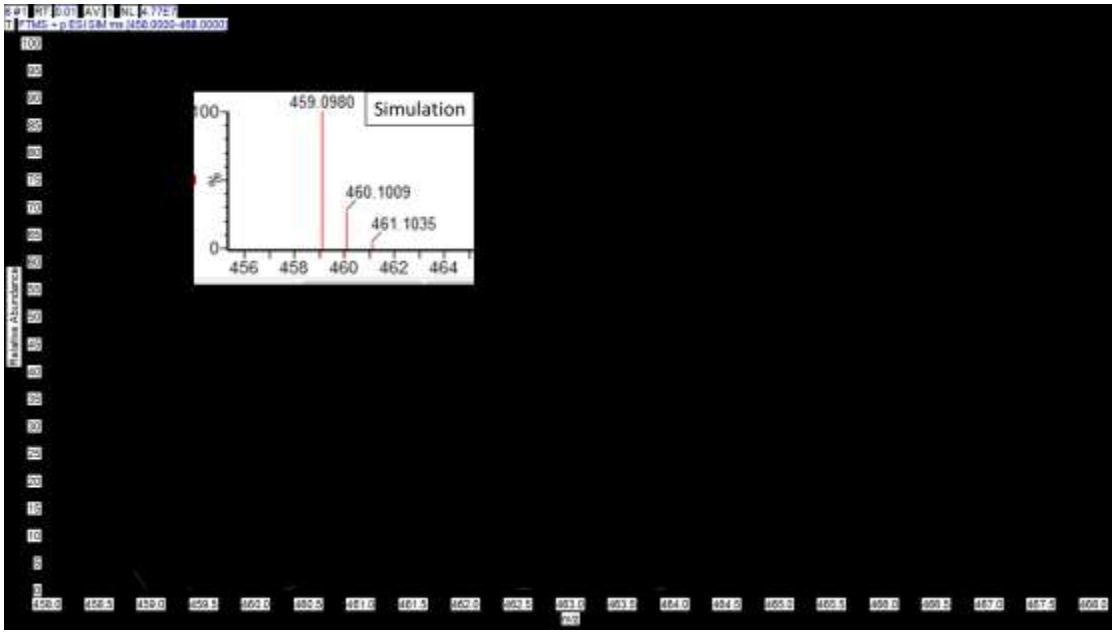

Fig. S23. Positive-ion mode HR-ESI-MS spectrum of the acetonitrile solution of $[Co(L^{O1})_2](ClO_4)$. The most aboundant peak at m/z = 459.0668 corresponds to $[M\text{-}ClO_4]^+$

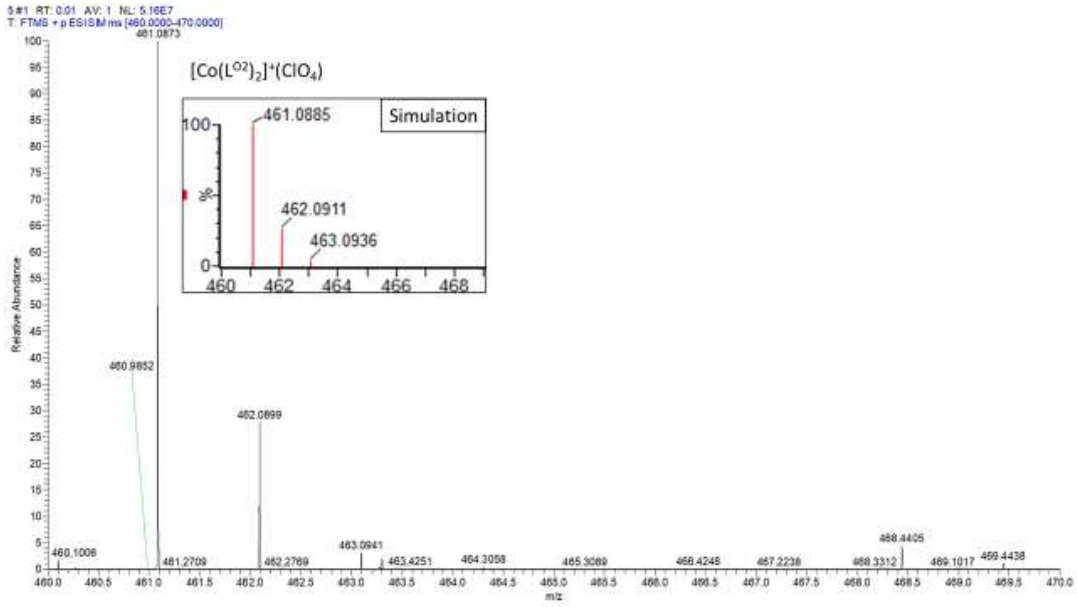

Fig. S24. Positive-ion mode HR-ESI-MS spectrum of the acetonitrile solution of $[Co(L^{O2})_2](ClO_4)$. The most aboundant peak at m/z = 461.0873 corresponds to $[M\text{-}ClO_4]^+$.

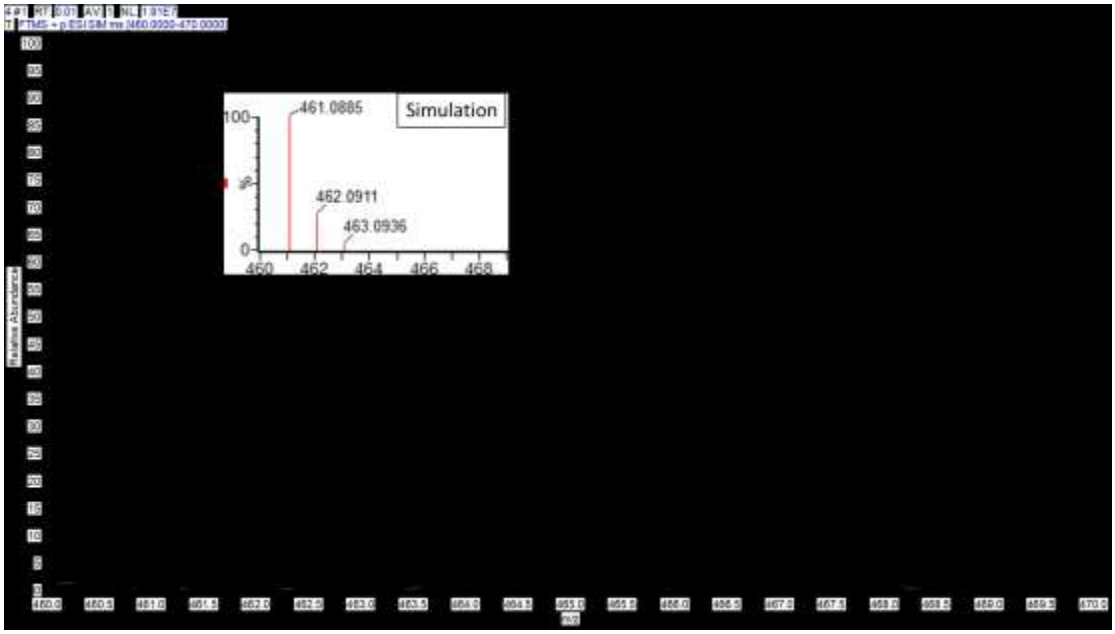

Fig. S25. Positive-ion mode HR-ESI-MS spectrum of the acetonitrile solution of $[Co(L^{O2})_2](NO_3)$. The most aboundant peak at m/z = 461.0872 corresponds to $[M\text{-}NO_3]^+$.

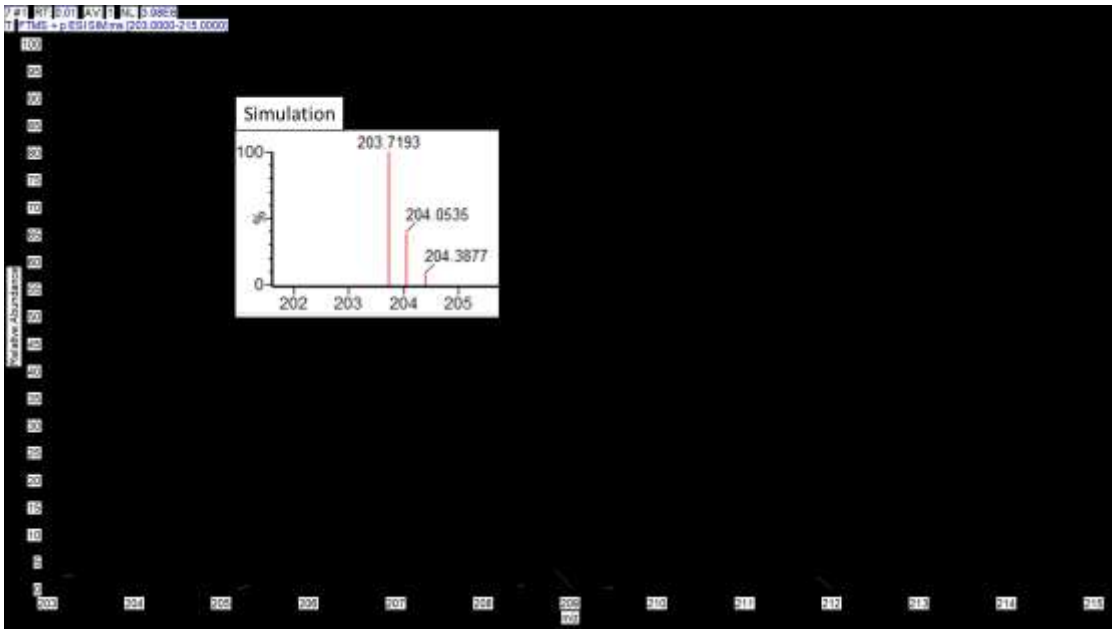

Fig. S26. Positive-ion mode HR-ESI-MS spectrum of the acetonitrile solution of $[Co(L^{M})](ClO_4)_3$. The most aboundant peak at m/z = 203.7187 corresponds to $[M\text{-}3ClO_4]^{3+}$.

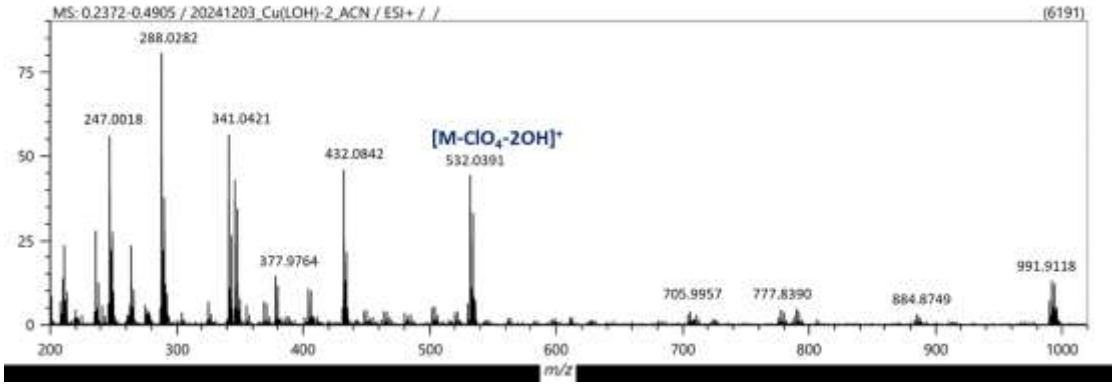

Fig. S27. Positive-ion mode HR-ESI-MS spectrum of the acetonitrile solution of $[Cu(L^{O2H})](ClO_4)_2$. The moderate aboundant peak at m/z = 532.0391 corresponds to $[M\text{-}ClO_4\text{-}2OH]^{+}$. The loss of hydroxyl radicals has been reported in the ESI-MS fragmentation of various Cu(II) complexes, consistent with the observed behavior here.[2]

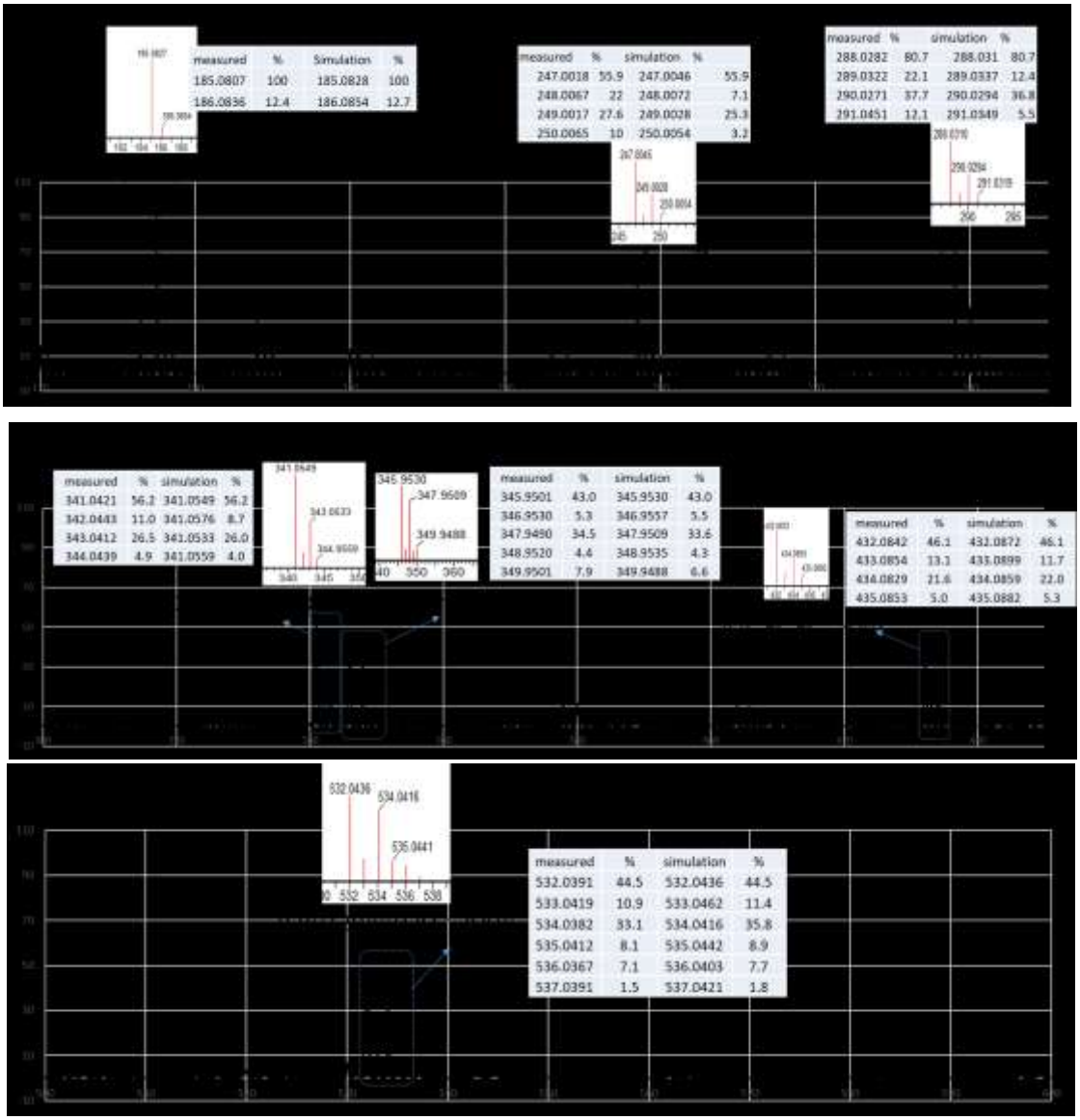

Fig. S28. Positive-ion mode HR-ESI-MS fragmentation pattern analysis of the acetonitrile solution of $[Cu(L^{O2H})](ClO_4)_2$. The hemiaminal ligand undergoes extensive fragmentation to generate multiple Cu(I) and Cu(II) species. Previous studies have shown that Cu(II) complexes can be readily reduced to Cu(I) during ESI-MS analysis, particularly in the presence of hydroxyl-containing ligands. (a) m/z = 130-300: The most aboundant peak at m/z = 185.0827 corresponds to $[L^{I2}+H]^+$, while the peaks at m/z= 247.0018 and 288.0282 correspond to $[Cu+L^{I2}]^+$and $[Cu+L^{I2}+MeCN]^+$, respectively. (b) m/z = 300-450: The peaks at m/z= 341.0421, 345.9501 and 432.0842 correspond to $[Cu+L^{O2H}+OH+MeCN+H_2O]^+$, $[Cu+L^{I2}+ClO_4]^+$ and $[Cu+L^{I2}+L^{O2H}-OH]^+$, respectively. (c) m/z = 500-600: The peaks at m/z= 532.0391 correspond to $[Cu+2L^{O2H}-2OH+ClO_4]^+$.

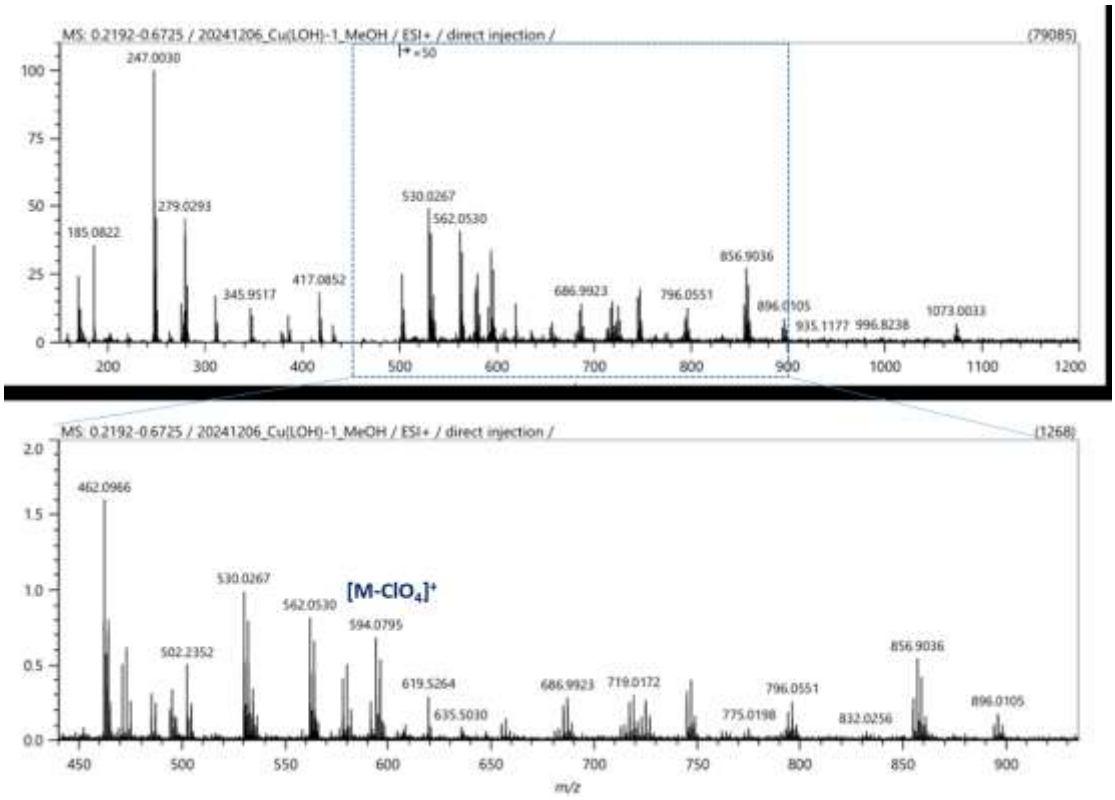

Fig. S29. Positive-ion mode HR-ESI-MS spectrum of the methanol solution of $[Cu(L^{O2H})](ClO_4)_2$. Although the aboundant is small, The peak corresponding to $[M\text{-}ClO_4]^+$ can be observed at m/z = 594.0795.

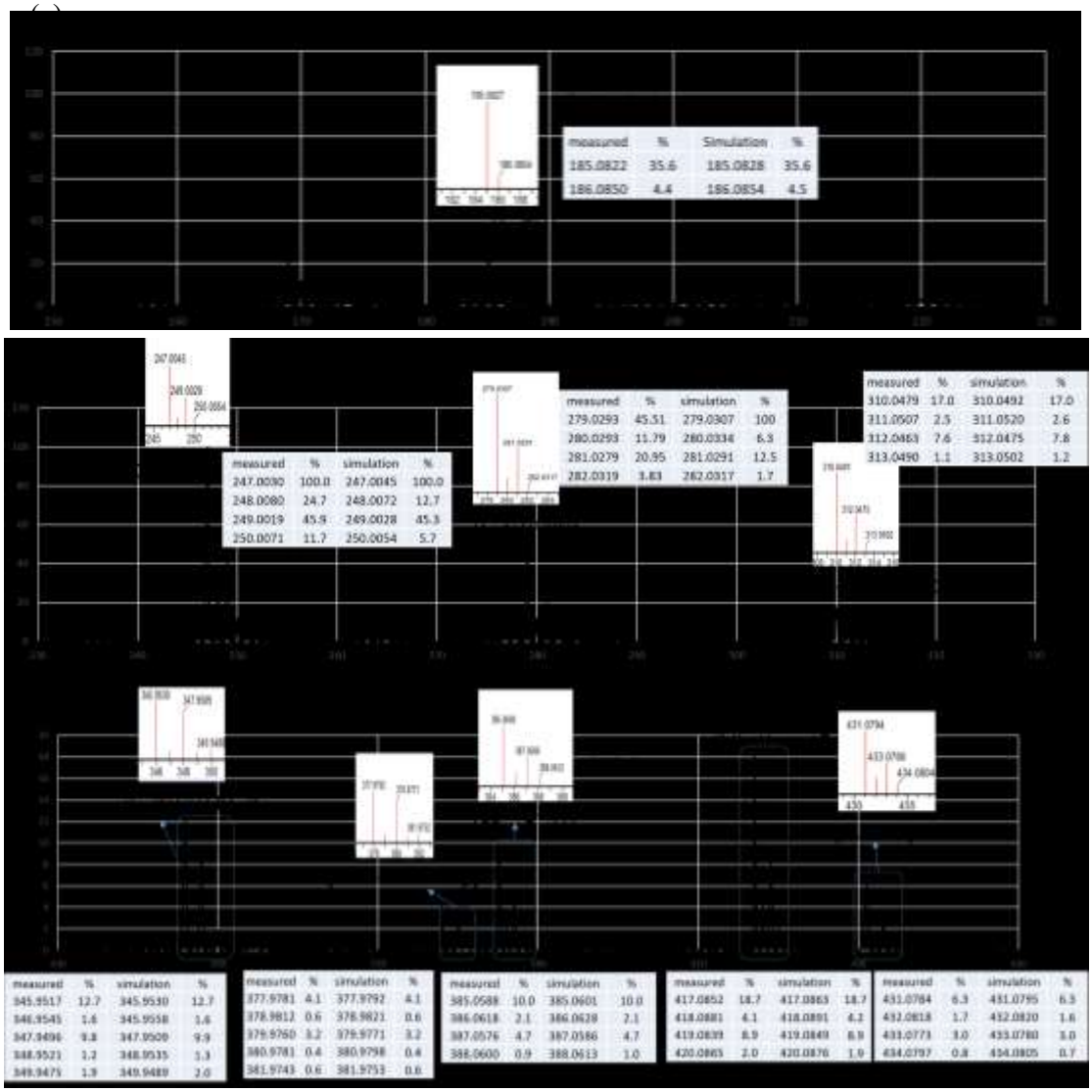

Fig. S30. Positive-ion mode HR-ESI-MS fragmentation pattern analysis of the methanol solution of $[Cu(L^{O2H})](ClO_4)_2$. The hemiaminal ether ligand undergoes extensive fragmentation to generate multiple Cu(I) and Cu(II) species.[3] (a) m/z = 150-230: The peak at m/z = 185.0827 corresponds to $[L^{I2}+H]^+$. (b) m/z = 230-330: The peaks at m/z= 247.0045, 279.0293 and 310.0479 correspond to $[Cu+L^{I2}]^+$, $[Cu+L^{O2Me}]^+$ and $[Cu+L^{O2Me}+MeO]^+$, respectively. (c) m/z = 330-450: The peaks at m/z= 345.9517, 377.9781 and 385.0588 correspond to $[Cu+L^{I2}+ClO_4]^+$, $[Cu+L^{O2Me}+ClO_4]^+$ and $[Cu+L^{O2Me}+A]^+$ (A = 2-pyridinecarboxaldehyde methyl hemiacetal), respectively.

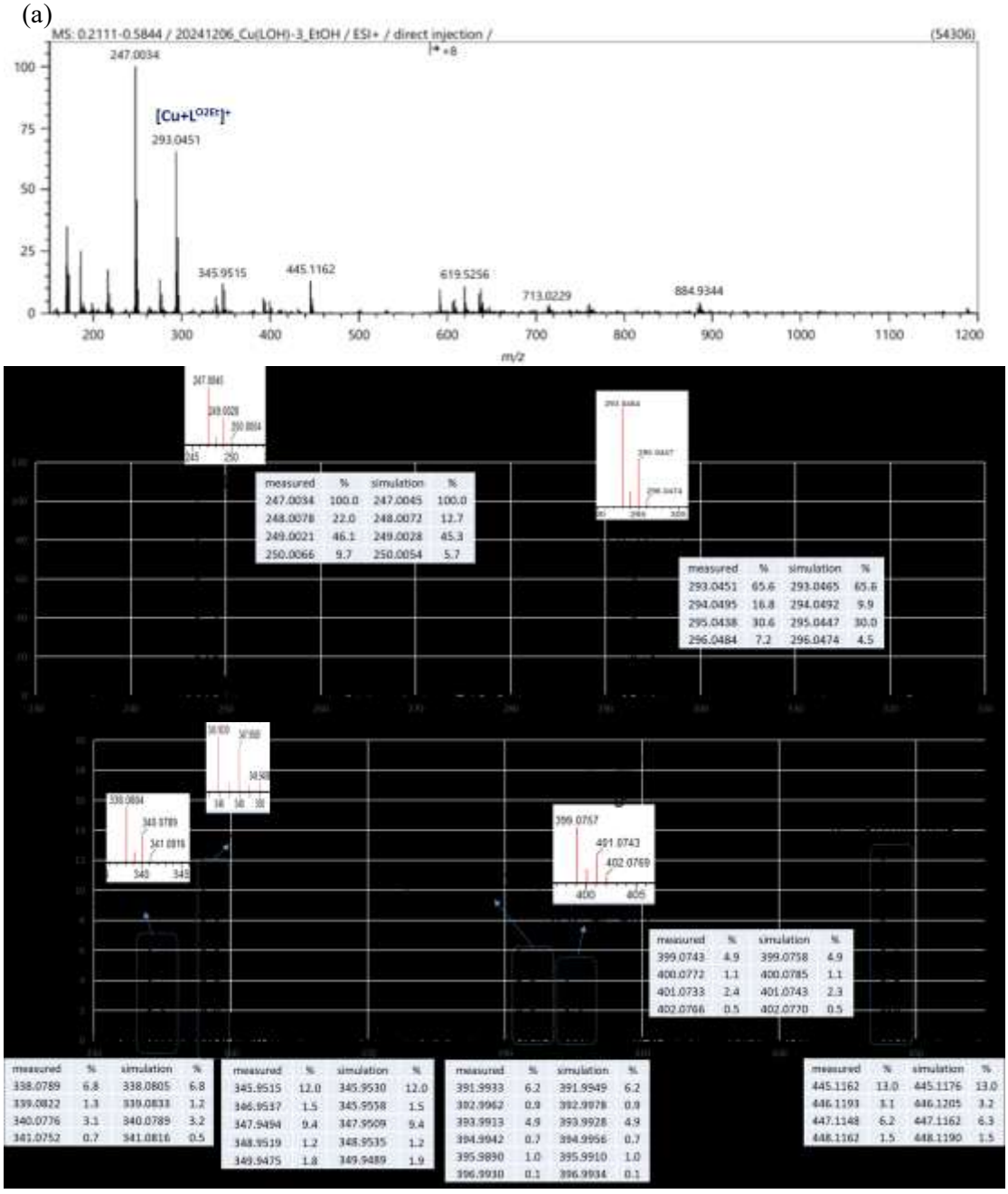

Fig. S31. Positive-ion mode HR-ESI-MS fragmentation pattern analysis of the ethanol solution of $[Cu(L^{O2H})](ClO_4)_2$. The fragmentation pattern is similar to that observed in methanol, except that the hemiaminal ether ligand contains an ethyl group rather than a methyl group. In the range of *m/z* = 150–230, only the $[L^{I2} + H]^+$ peak is observed, consistent with Fig. S30(a) (a) The full spectrum. (b) m/z = 230-330: The peaks at m/z= 247.0045 and 293.0451 correspond to $[Cu+L^{I2}]^+$ and $[Cu+L^{O2Et}]^+$ and, respectively. (c) m/z = 330-450: The peaks at m/z= 338.0789, 345.9515, 391.9933, 399.0743 and 445.1162 correspond to $[Cu+L^{O2Et}+EtO]^+$, $[Cu+L^{I2}+ClO_4]^+$, $[Cu+L^{O2Et}+ClO_4]^+$, $[Cu+L^{I2}+B]^+$ and $[Cu+L^{O2Et}+B]^+$ (B = 2-pyridinecarboxaldehyde ethyl hemiacetal), respectively.

The coordinates of optimized structures.

Table S5. Optimized structure of **Cry-[Co(L^{O2})$_2$]$^+$** (Energy= -2744.15359230 Hartree, Charge= 1, Multiplicity=1)

Center Number	Coordinates (Angstroms) X	Y	Z
Co	-0.579160	4.368973	7.338227
O	-0.361620	5.202911	9.018950
O	-0.741787	3.658066	5.595855
N	-1.311822	6.069988	6.812204
N	-2.654993	5.378424	9.529557
H	-3.276483	5.762657	10.228890
N	-2.374558	3.745260	7.866617
N	-4.311041	3.810968	9.274783
N	1.165269	4.917455	6.736284
N	0.862517	1.951871	5.851968
H	1.334522	1.230525	5.323003
N	0.168123	2.659868	7.979889
N	1.173591	0.516703	7.614884
C	-1.469494	6.532193	5.564204
H	-1.202550	5.838494	4.776320
C	-1.944619	7.819021	5.333215
H	-2.059169	8.174684	4.315606
C	-2.257681	8.630311	6.425932
H	-2.622447	9.640912	6.271774
C	-2.098524	8.131430	7.720267
H	-2.338681	8.730037	8.592305
C	-1.618162	6.838274	7.877638
C	-1.402669	6.084305	9.184046
H	-1.222455	6.783109	10.009586
C	-3.116421	4.293838	8.870703
C	-2.880987	2.660073	7.243238
H	-2.265886	2.269084	6.442420
C	-4.097182	2.110502	7.597975
H	-4.492901	1.239243	7.092035
C	-4.779472	2.739839	8.645725
H	-5.741543	2.365452	8.989410

C	1.952545	5.853947	7.283334
H	1.574651	6.301985	8.194197
C	3.164332	6.201714	6.695427
H	3.783325	6.965702	7.151834
C	3.553616	5.558768	5.518448
H	4.490727	5.818061	5.036029
C	2.728020	4.577807	4.965770
H	3.001504	4.052619	4.057060
C	1.531472	4.281397	5.604663
C	0.503483	3.230807	5.202608
H	0.549255	3.035759	4.124635
C	0.731961	1.714641	7.174893
C	0.058556	2.369726	9.293905
H	-0.378782	3.155718	9.896794
C	0.489505	1.167323	9.818784
H	0.395210	0.945357	10.874004
C	1.046873	0.259188	8.911082
H	1.406172	-0.712578	9.243422

Table S6. Optimized structure of **iso1-[Co(L^{O2})$_2$]$^+$** (Energy= -2744.15044525 Hartree, Charge= 1, Multiplicity=1)

Atomic Number	Coordinates (Angstroms) X	Y	Z
Co	-3.499716	0.576712	8.469163
O	-4.238780	0.562316	6.734559
N	-4.539041	2.171130	8.707543
N	-1.981642	1.667366	7.771598
M	-3.168539	2.488404	5.917958
H	-3.132346	3.054692	5.081075
C	-4.433360	1.868314	6.348149
H	-5.109509	1.944965	5.488093
C	-4.982479	2.662666	7.531998
C	-5.807581	3.778718	7.485510
H	-6.153536	4.158736	6.530245
C	-6.175403	4.386236	8.687759
H	-6.826331	5.254793	8.682587

C	-5.700982	3.868742	9.895339
H	-5.967855	4.318429	10.844882
C	-4.877215	2.748405	9.868990
H	-4.469914	2.280282	10.757305
C	-2.016850	2.388008	6.611975
C	-0.806132	1.640694	8.434760
H	-0.825293	1.091586	9.366055
C	0.324250	2.283168	7.969091
H	1.258140	2.246700	8.515228
C	0.191573	2.970711	6.758307
H	1.036956	3.492403	6.314427
O	-2.760651	0.591108	10.203768
N	-2.460391	-1.017706	8.230783
N	-5.017789	-0.513942	9.166728
N	-3.830892	-1.334980	11.020368
H	-3.867086	-1.901268	11.857251
C	-2.566071	-0.714890	10.590177
H	-1.889923	-0.791540	11.450234
C	-2.016953	-1.509242	9.406329
C	-1.191851	-2.625294	9.452816
H	-0.845896	-3.005312	10.408081
C	-0.824029	-3.232812	8.250568
H	-0.173101	-4.101369	8.255740
C	-1.298449	-2.715318	7.042987
H	-1.031577	-3.165005	6.093445
C	-2.122217	-1.594980	7.069337
H	-2.529518	-1.126858	6.181021
C	-4.982582	-1.234584	10.326351
C	-6.193299	-0.487270	8.503566
H	-6.174139	0.061838	7.572271
C	-7.323681	-1.129744	8.969235
H	-8.257571	-1.093276	8.423098
C	-7.191005	-1.817287	10.180020
H	-8.036387	-2.338979	10.623900
N	-0.952092	3.033071	6.088245
N	-6.047340	-1.879647	10.850081

Table S7. Optimized structure of **iso2-[Co(L^{O2})$_2$]$^+$** (Energy= -2744.13600869 Hartree, Charge= 1, Multiplicity=1)

Atomic Number	Coordinates (Angstroms) X	Y	Z
Co	-3.549673	0.733775	8.670890
O	-2.099079	1.903993	8.494087
N	-4.537559	2.338951	9.165636
C	-2.581243	3.116197	8.069214
C	-3.837861	3.449840	8.852977
C	-4.261345	4.715942	9.234392
H	-3.674535	5.586040	8.960904
C	-5.434923	4.832436	9.979498
H	-5.788548	5.808041	10.297495
C	-6.137689	3.677430	10.324236
H	-7.043126	3.721772	10.918432
C	-5.652238	2.443361	9.906206
H	-6.147063	1.519811	10.172630
O	-3.220229	0.459382	10.493017
N	-2.295930	-0.736220	8.418359
N	-5.170232	-0.433749	8.894943
N	-4.234453	-1.673076	10.652272
H	-4.360732	-2.442572	11.296766
C	-2.985239	-0.878768	10.684942
H	-2.543005	-1.072441	11.670070
C	-2.072487	-1.383241	9.581456
C	-1.107121	-2.374638	9.696238
H	-0.958096	-2.875603	10.646523
C	-0.337270	-2.691384	8.576810
H	0.428925	-3.457745	8.636900
C	-0.553921	-1.998143	7.385535
H	0.035324	-2.199349	6.498281
C	-1.538612	-1.017274	7.346233
H	-1.722052	-0.434183	6.454465
C	-5.262752	-1.434493	9.819751
C	-6.294704	-0.159281	8.191977
H	-6.230455	0.679819	7.510716

C	-7.456939	-0.887520	8.325005
H	-8.341381	-0.651070	7.747991
C	-7.416462	-1.950484	9.238349
H	-8.278493	-2.599191	9.378421
N	-6.353096	-2.220358	9.980737
N	-2.923087	3.104917	6.628387
H	-2.718563	3.912237	6.053858
C	-3.490174	2.065156	5.992020
C	-4.261883	-0.113180	5.962486
C	-4.139933	1.169637	3.987301
C	-4.434259	-0.058459	4.596450
H	-4.463747	-1.019169	6.520013
H	-4.282893	1.310892	2.918141
H	-4.784792	-0.915374	4.036179
N	-3.830143	0.942932	6.692170
N	-3.668989	2.210297	4.658078
H	-1.832335	3.909407	8.184904

Table S8. Optimized structure of **iso3-[Co(L^{O2})$_2$]$^+$** (Energy= -2744.13169008 Hartree, Charge= 1, Multiplicity=1)

Atomic Number	Coordinates (Angstroms) X	Y	Z
Co	-2.822649	1.143168	8.902958
O	-1.770440	2.322101	7.903720
N	-3.899568	2.727678	9.512319
C	-4.012459	3.895935	8.813413
C	-5.284646	4.906791	10.426292
H	-5.857826	5.778108	10.736091
C	-5.150610	3.794217	11.262855
H	-5.581239	3.757130	12.255160
C	-4.439878	2.727528	10.750923
H	-4.286931	1.837461	11.339599
O	-1.812761	1.328184	10.465298
N	-1.563499	-0.320326	8.344011
N	-3.917396	-0.064850	10.021828

C	-0.829688	-1.077412	9.212602
C	0.233563	-2.225131	7.541068
H	0.926470	-3.016447	7.262850
C	-0.395642	-1.440304	6.569630
H	-0.206297	-1.562301	5.510979
C	-1.288275	-0.494890	7.032587
H	-1.805632	0.153979	6.344592
C	-3.236207	-0.435588	11.128347
C	-5.216630	-0.393962	9.909839
H	-5.746722	-0.000337	9.052875
C	-5.866633	-1.161279	10.869010
H	-6.915748	-1.401063	10.740787
C	-5.152526	-1.586454	11.990180
H	-5.635163	-2.182751	12.757706
C	-3.533900	1.980198	6.371212
C	-4.574633	-0.062421	6.761012
C	-4.846852	0.996179	4.625586
C	-5.109220	-0.077054	5.478318
H	-4.709868	-0.898223	7.434450
H	-5.246936	1.009558	3.616949
H	-5.703126	-0.924651	5.156598
N	-3.828160	0.962405	7.209776
N	0.035520	-2.047032	8.839444
N	-0.923489	-0.860960	10.537796
C	-1.825183	0.133200	11.142143
H	-1.478227	0.257978	12.175335
C	-3.816970	-1.208276	12.126218
H	-3.235571	-1.488073	12.998009
C	-4.036519	2.037570	5.077379
H	-3.778594	2.873334	4.436071
C	-2.575657	2.977562	7.004677
H	-1.981555	3.475325	6.228420
N	-4.720590	4.970210	9.228543
N	-3.371180	4.041217	7.638756
H	-3.541827	4.909468	7.148604
H	-0.393091	-1.490656	11.125363

Table S9. Optimized structure of $[Sc(L^{I2})_3]^{3+}$ (Energy= -1859.946883 Hartree, Charge= 3, Multiplicity=1)

Atomic Number	Coordinates (Angstroms) X	Y	Z
C	-1.02871200	2.70690000	0.98729000
N	0.24671100	2.30531100	1.13621000
C	1.07105100	3.17413200	1.74637300
C	0.60300800	4.40971600	2.19293400
C	-0.74872900	4.70807300	1.97644700
N	-1.57748700	3.84804900	1.36250900
H	2.10655100	2.87273100	1.87246800
H	1.26181000	5.11454100	2.68804800
H	-1.17953500	5.65278700	2.29791000
N	-1.72566100	1.68866000	0.33091200
C	-5.06635400	-0.75036400	-1.96757300
C	-4.12549300	-1.76606500	-2.08924900
C	-4.69483200	0.42262500	-1.29816200
C	-2.84492100	-1.58343200	-1.54168800
H	-4.36308200	-2.69520300	-2.59694400
C	-3.40224400	0.52469200	-0.78552500
H	-5.39479000	1.24430700	-1.17899300
N	-2.46914200	-0.47156600	-0.90108300
H	-6.06501900	-0.85931300	-2.37907800
C	-2.94316900	1.71746300	-0.08599200
H	-2.10294600	-2.36859900	-1.62365300
C	0.61043300	0.54382900	-2.73766500
N	0.17457900	1.55799000	-1.96711000
C	0.19769500	2.77907800	-2.52821600
C	0.64719400	2.95487700	-3.83570300
C	1.07552400	1.81676400	-4.53171800
N	1.06082800	0.59416800	-3.97655500
H	-0.14765300	3.61321300	-1.92480100
H	0.66410000	3.93597500	-4.29712600
H	1.43751100	1.88175900	-5.55459800
N	0.50649100	-0.63214100	-1.99052200
C	0.70080700	-5.18725800	-0.70116300

C	0.11315200	-4.79185400	0.49506400
C	0.96159100	-4.20802800	-1.66704100
C	-0.18591800	-3.43368800	0.69553400
H	-0.12036300	-5.51242900	1.27218000
C	0.63137600	-2.88126600	-1.38696600
H	1.41120600	-4.46823200	-2.62071900
N	0.06447600	-2.48217300	-0.20817200
H	0.94690100	-6.22798800	-0.88763600
C	0.84716300	-1.81774300	-2.35795900
H	-0.65148200	-3.11433300	1.62123800
C	3.00275300	-0.13505200	0.70207000
N	3.02742500	0.36038200	-0.53713300
C	4.25260800	0.55472500	-1.05780200
C	5.41362600	0.25343600	-0.34916500
C	5.25121300	-0.26076600	0.94151500
N	4.03759500	-0.45479600	1.47165100
H	4.29499500	0.95938400	-2.06542500
H	6.39730400	0.41064700	-0.77747100
H	6.10327000	-0.52244900	1.56362400
N	1.68503400	-0.31122300	1.21972000
C	-1.33161300	-1.27750800	4.80709500
C	-2.38390400	-1.05140000	3.92392700
C	-0.02165000	-1.14661100	4.33297900
C	-2.09584700	-0.69528800	2.60030700
H	-3.41695200	-1.14365700	4.24280000
C	0.18481700	-0.79168700	2.99887000
H	0.82852600	-1.31674400	4.98653200
N	-0.84612500	-0.56154800	2.13274800
H	-1.51976400	-1.55068300	5.84091900
C	1.52909800	-0.64819700	2.45725700
H	-2.89962000	-0.51581300	1.89506300
H	2.39534500	-0.81814200	3.09815500
H	1.26662800	-2.02008900	-3.34487400
H	-3.59290500	2.58118800	0.06616100
Sc	-0.20340100	0.09232100	-0.01535500

Table S10. Optimized structure of **[Sc(L^{M})]$^{3+}$** (Energy= -1859.930796 Hartree, Charge= 3, Multiplicity=1)

Atomic Number	Coordinates (Angstroms) X	Y	Z
C	2.59241500	0.24730200	-3.13906800
H	1.93057700	-0.17046000	-3.89283900
C	3.91555100	0.54905600	-3.44497900
H	4.29608400	0.37335600	-4.44546300
C	4.72463000	1.08551200	-2.44312700
H	5.75840600	1.34675400	-2.64815200
C	4.18443900	1.29221400	-1.16986300
H	4.79396100	1.72042000	-0.38037100
C	2.85347600	0.96206400	-0.93754700
C	2.20919300	1.16195000	0.42917600
H	2.70106000	1.98484100	0.94912900
C	0.19958700	2.10085800	1.28087200
C	0.35019300	3.52368700	3.10266000
H	1.00514000	4.03000300	3.80849900
C	-1.04678200	3.68771400	3.16956600
H	-1.52128500	4.32135800	3.90893400
C	-1.78651000	2.96464800	2.27197500
H	-2.87039000	2.97454700	2.26905900
C	-1.51416600	2.12197700	-3.13694100
H	-0.82237300	1.75781900	-3.89161900
C	-2.43736600	3.11709400	-3.44151000
H	-2.47661500	3.53476900	-4.44183500
C	-3.30530100	3.54933200	-2.43853100
H	-4.04863800	4.31407200	-2.64248800
C	-3.21274100	2.97785400	-1.16552100
H	-3.88743100	3.29142100	-0.37517300
C	-2.26105300	1.99023400	-0.93455500
C	-2.11031700	1.33206000	0.43183700
H	-3.06824600	1.34639900	0.95298900
C	-1.08431000	-2.37082300	-3.13648000
H	-1.11569200	-1.58934600	-3.89082600
C	-1.48499800	-3.66774100	-3.44105100

H	-1.82831100	-3.91016800	-4.44105500
C	-1.42422500	-4.63589700	-2.43850700
H	-1.71518200	-5.66191100	-2.64250200
C	-0.97403100	-4.27051000	-1.16589900
H	-0.90738400	-5.01189200	-0.37590400
C	-0.59417200	-2.95262700	-0.93488900
C	-0.09796800	-2.49358500	0.43114000
H	0.36915500	-3.33061600	0.95134000
C	1.72074000	-1.22210600	1.28046100
C	2.87951700	-2.06285900	3.10155500
H	2.99130500	-2.88282700	3.80772700
C	3.72013600	-0.93502100	3.16689800
H	4.50692000	-0.84047300	3.90538000
C	3.46287400	0.06662300	2.26899100
H	4.01336300	1.00035800	2.26495900
C	-1.67188800	-3.03034100	2.27262000
H	-1.13842300	-3.97390400	2.26889100
C	-2.66727300	-2.75170200	3.17109200
H	-2.97813000	-3.47977600	3.91041600
C	-3.22393200	-1.45992300	3.10510500
H	-3.98943700	-1.14628800	3.81161500
C	-1.91787400	-0.87805300	1.28255600
N	2.06365600	0.45315500	-1.91419100
N	0.79024700	1.38619600	0.31016200
N	0.93780000	2.74969100	2.20462000
N	-1.19215100	2.16905500	1.33199200
N	-1.42662500	1.56083500	-1.91229500
N	-1.59509600	-0.00882500	0.31188200
N	-2.84820500	-0.56361500	2.20716400
N	-1.28094000	-2.11739600	1.33268000
N	-0.64060900	-2.01494300	-1.91223500
N	0.80558100	-1.37690600	0.31069400
N	1.91443800	-2.18526200	2.20463000
N	2.47566900	-0.05087200	1.33015400
Sc	-0.00056400	-0.00000900	-1.13170200

Table S11. Optimized structure of $[Ti(L^{I2})_3]^{2+}$ (Energy= -1871.904575 Hartree, Charge= 2, Multiplicity=3)

Atomic Number	Coordinates (Angstroms) X	Y	Z
C	1.47192100	1.13548300	-2.31558000
N	2.02229100	1.72893800	-1.25489400
C	3.10100700	2.48596300	-1.50133800
C	3.61876500	2.64890900	-2.78413800
C	2.95672000	1.97503900	-3.81257600
N	1.88284100	1.21380000	-3.58221700
H	3.55591800	2.97322200	-0.64220200
H	4.48946200	3.26583700	-2.97498000
H	3.29123600	2.04335000	-4.84500300
N	0.32879700	0.33860200	-2.01833800
C	-3.15865200	-2.62947200	-3.37260600
C	-3.52312300	-2.72348300	-2.02788400
C	-2.08093100	-1.81600500	-3.71536500
C	-2.79952500	-2.00340000	-1.07750800
H	-4.35323700	-3.34645600	-1.71232600
C	-1.40064600	-1.12381900	-2.70673700
H	-1.76063100	-1.71282900	-4.74733400
N	-1.75683500	-1.21378800	-1.39061300
H	-3.70112200	-3.17875200	-4.13546800
C	-0.26517700	-0.27447700	-2.99776100
H	-3.06107800	-2.05993800	-0.02676800
C	1.46397100	-2.57808500	0.17086200
N	2.01379800	-1.95754000	-0.87451200
C	3.08912500	-2.55287300	-1.40994600
C	3.60406000	-3.74732200	-0.91160500
C	2.94300300	-4.29928700	0.18768500
N	1.87248300	-3.71581000	0.73479000
H	3.54355000	-2.05360400	-2.26241800
H	4.47198000	-4.22382600	-1.35295800
H	3.27554800	-5.22889600	0.64344000
N	0.32448500	-1.91864500	0.71550800
C	-3.15428600	-1.59626400	3.97118600

C	-3.51653400	-0.38378800	3.38072200
C	-2.07983200	-2.30316800	3.43588300
C	-2.79394000	0.07684400	2.28027000
H	-4.34412100	0.20347800	3.76409500
C	-1.40039100	-1.77804100	2.33051900
H	-1.76145800	-3.24935000	3.86199900
N	-1.75428300	-0.59225700	1.75099800
H	-3.69599900	-1.98047100	4.82960000
C	-0.26849000	-2.45836900	1.73777900
H	-3.05376400	1.01564500	1.80403600
C	1.47531700	1.43256600	2.14308200
N	2.01901000	0.21420700	2.13064500
C	3.09620300	0.04545300	2.91080300
C	3.61905600	1.07431300	3.69070300
C	2.96392200	2.30535900	3.61712500
N	1.89165200	2.49014900	2.84131900
H	3.54564000	-0.94473500	2.90650400
H	4.48845200	0.92814500	4.32151000
H	3.30278500	3.16515500	4.19018500
N	0.33357000	1.57754100	1.30311000
C	-3.14112600	4.24738800	-0.59547100
C	-3.50957500	3.13075400	-1.34869100
C	-2.06460200	4.13375900	0.28141400
C	-2.79086600	1.94486800	-1.19882000
H	-4.33900600	3.17180000	-2.04663000
C	-1.38937400	2.91148400	0.37802900
H	-1.74143600	4.97500400	0.88644900
N	-1.74931600	1.81768300	-0.35758300
H	-3.67969700	5.18482200	-0.69087900
C	-0.25552100	2.73472800	1.26038900
H	-3.05546300	1.06368100	-1.77254100
H	0.10244800	3.56564800	1.86587400
H	0.08459500	-3.40012200	2.15444600
H	0.08987200	-0.16576800	-4.02113700
Ti	-0.47985500	0.00083900	0.00056600

Table S12. Optimized structure of **[Ti (L^M)]$^{2+}$** (Energy= -1871.872383 Hartree, Charge= 2, Multiplicity=3)

Atomic Number	Coordinates (Angstroms) X	Y	Z
C	3.08148800	-2.53179300	0.52195900
H	3.81953200	-1.94968200	-0.02299100
C	3.39847200	-3.79001300	1.02427100
H	4.39431800	-4.19486400	0.87930900
C	2.41724000	-4.50462800	1.71048200
H	2.62916400	-5.48552300	2.12352800
C	1.15226900	-3.92967800	1.86048100
H	0.36951100	-4.45663300	2.39752900
C	0.90903500	-2.66868200	1.32638200
C	-0.45430200	-2.00591100	1.45846100
H	-0.96604200	-2.39603100	2.33987200
C	-1.29352500	0.11280400	2.08845900
C	-3.16836400	0.21495700	3.44868100
H	-3.91938500	-0.33920000	4.00855600
C	-3.18508900	1.62533100	3.43178000
H	-3.92239500	2.20332200	3.97395200
C	-2.24333900	2.23042600	2.64369200
H	-2.20213900	3.30492100	2.50769000
C	3.15592700	1.62872300	1.88777600
H	3.86843700	0.84477200	1.64635900
C	3.51507800	2.68364000	2.72088800
H	4.51858500	2.73209900	3.12984800
C	2.56553400	3.66310100	3.00966700
H	2.81073600	4.50510700	3.64886500
C	1.28863500	3.54166000	2.45469400
H	0.52981900	4.29248000	2.65288200
C	1.00253500	2.45571000	1.63409500
C	-0.37535000	2.27767200	1.01354100
H	-0.86106400	3.25021400	0.91758900
C	3.14495000	0.73207200	-2.39785200
H	3.87345700	0.89388100	-1.60814600
C	3.49079800	0.91744700	-3.73274500

H	4.49968100	1.21920200	-3.99298300
C	2.52106400	0.70589000	-4.71214100
H	2.75521000	0.83274200	-5.76416700
C	1.23829600	0.32173200	-4.31197500
H	0.46365600	0.14031900	-5.05078500
C	0.96647100	0.16062100	-2.95746200
C	-0.41693000	-0.24932100	-2.47420600
H	-0.92918800	-0.80427700	-3.26197900
C	-1.32575600	-1.83020700	-0.96941100
C	-3.22245500	-3.00836500	-1.59396500
H	-3.96546700	-3.19595800	-2.36683100
C	-3.27920200	-3.69802700	-0.36479200
H	-4.04057900	-4.43635400	-0.14841000
C	-2.34367300	-3.34346400	0.56964600
H	-2.33120800	-3.76366900	1.56864500
C	-2.25554700	1.23734600	-3.22127500
H	-2.24508000	0.58168200	-4.08424200
C	-3.16775600	2.24795100	-3.07754900
H	-3.91102800	2.44902300	-3.83831300
C	-3.11339100	2.96664500	-1.86501500
H	-3.84006100	3.74899800	-1.65416900
C	-1.25951400	1.78799900	-1.12377000
N	1.86532300	-1.96645800	0.66922000
N	-0.32707500	-0.57440400	1.49802600
N	-2.27847400	-0.50414400	2.79437200
N	-1.30716100	1.51018800	1.96443100
N	1.92798900	1.50828300	1.34186000
N	-0.28539600	1.59300900	-0.24743700
N	-2.21586100	2.73466200	-0.92811200
N	-1.31201600	0.98319800	-2.27171200
N	1.91125000	0.35387400	-2.00375600
N	-0.33126700	-1.00204500	-1.25228900
N	-2.30292600	-2.10651900	-1.87376600
N	-1.37747300	-2.42075500	0.30211900
Ti	1.10779700	-0.02039200	0.00140500

Table S13. Optimized structure of **[Ti(L^{12})$_3$]$^{4+}$** (Energy= -1870.954701 Hartree, Charge= 4, Multiplicity=1)

Atomic Number	Coordinates (Angstroms) X	Y	Z
C	0.89204000	-2.36933900	1.51377500
N	-0.33126100	-1.81150100	1.47185300
C	-1.28782300	-2.39244500	2.22279000
C	-0.97045300	-3.50915900	2.99796200
C	0.34894400	-3.99126700	2.96134500
N	1.29602000	-3.41034800	2.19848700
H	-2.28724100	-1.97233900	2.19256700
H	-1.72546700	-3.99314300	3.60954200
H	0.65537600	-4.85586400	3.54534000
N	1.66024600	-1.60556400	0.63595600
C	4.85761700	0.11552600	-2.36001400
C	3.89893600	1.07393100	-2.66331500
C	4.53886700	-0.85513600	-1.39624100
C	2.65579900	1.04920900	-2.00212200
H	4.09063100	1.84802600	-3.40033200
C	3.28451700	-0.81677900	-0.78927200
H	5.25075400	-1.63088700	-1.12704200
N	2.32996400	0.13486800	-1.07989700
H	5.82644600	0.11185600	-2.85171800
C	2.85993100	-1.78771700	0.19129800
H	1.91479600	1.80155200	-2.23417200
C	-0.70983200	-1.07694200	-2.36899200
N	-0.15976000	-1.86191000	-1.42552600
C	-0.15240400	-3.18754600	-1.67275500
C	-0.68976100	-3.67109400	-2.86451500
C	-1.22904000	-2.74238800	-3.77161000
N	-1.24091400	-1.41982200	-3.51349400
H	0.28228800	-3.84799600	-0.92892500
H	-0.68741500	-4.73361000	-3.08603600
H	-1.65592400	-3.05839600	-4.72056400
N	-0.57628900	0.22307700	-1.87546000
C	-0.38434500	4.91423900	-1.39818200

C	0.37598300	4.66085100	-0.26245700
C	-0.85758900	3.81768000	-2.13314800
C	0.62242000	3.32900200	0.12876700
H	0.79215300	5.47185300	0.32786500
C	-0.56599700	2.52825000	-1.68206000
H	-1.43570100	3.95964100	-3.04229900
N	0.15914700	2.27217100	-0.54290100
H	-0.59474000	5.93048700	-1.71928000
C	-0.93277300	1.34330500	-2.41693200
H	1.22840000	3.13447800	1.00503200
C	-2.90681800	0.41724400	0.45029600
N	-3.04680100	-0.53900400	-0.46395900
C	-4.31165200	-0.74496000	-0.88594300
C	-5.38418700	0.00212900	-0.39957300
C	-5.09478400	0.98664100	0.55248600
N	-3.83952500	1.19386100	0.97916100
H	-4.45880800	-1.52630200	-1.62627800
H	-6.39802400	-0.17072400	-0.74542900
H	-5.86847200	1.61908600	0.98024800
N	-1.55949800	0.58835800	0.94625500
C	1.48910400	2.03679500	4.33107800
C	2.51633700	1.52889000	3.53804100
C	0.17199700	1.93243000	3.86312500
C	2.20407700	0.91855700	2.31571200
H	3.55409400	1.59234900	3.85151000
C	-0.06623100	1.31875800	2.63270600
H	-0.65965800	2.32255000	4.44329900
N	0.94360900	0.80085600	1.86137800
H	1.70114300	2.50627600	5.28781300
C	-1.39848700	1.18467100	2.08656600
H	2.99439300	0.52157800	1.69109100
H	-2.26553500	1.57412600	2.62118000
H	-1.44241000	1.37299200	-3.38131800
H	3.48595800	-2.61586200	0.52872900
Ti	0.25509200	-0.15441000	0.00560500

Table S14. Optimized structure of **[Ti (L^M)]$^{4+}$** (Energy= -1870.985432 Hartree, Charge= 4, Multiplicity=1)

Atomic Number	Coordinates (Angstroms) X	Y	Z
C	-2.15089400	0.80669100	3.06963300
H	-1.58143500	0.17253800	3.74187100
C	-3.30576300	1.45018500	3.50288200
H	-3.64021200	1.33016400	4.52853300
C	-4.00944900	2.25104600	2.60030400
H	-4.90474400	2.78095200	2.91360900
C	-3.54301800	2.37056000	1.28299200
H	-4.07270700	2.99495500	0.56989700
C	-2.38436400	1.69983000	0.92191000
C	-1.78906000	1.74603300	-0.47450700
H	-2.00667800	2.69039600	-0.97418200
C	0.44796400	2.09462000	-1.32129800
C	0.73572500	3.63222100	-3.01960900
H	0.26385500	4.36639200	-3.66895700
C	2.11478700	3.36991500	-3.09530000
H	2.76357700	3.88557900	-3.79527200
C	2.60754300	2.40200700	-2.25324900
H	3.64930700	2.09986200	-2.25684200
C	1.77065300	1.45618500	3.07310800
H	0.93620300	1.27879800	3.74434600
C	2.90484300	2.13410100	3.50847600
H	2.96704500	2.48228900	4.53469500
C	3.95115900	2.34442900	2.60726400
H	4.85736000	2.85444200	2.92222600
C	3.82293600	1.88233500	1.28923600
H	4.62940600	2.02964900	0.57729800
C	2.66318100	1.21462500	0.92605400
C	2.40734300	0.67694300	-0.47103300
H	3.33467300	0.39356900	-0.96966300
C	0.37185000	-2.26453500	3.07155800
H	0.63448800	-1.45387300	3.74400900
C	0.39175900	-3.58625300	3.50537800

H	0.66098700	-3.81534800	4.53166000
C	0.05262200	-4.59664300	2.60242500
H	0.04136700	-5.63684000	2.91607500
C	-0.28183800	-4.25314000	1.28435600
H	-0.55596300	-5.02447100	0.57102000
C	-0.28056100	-2.91448100	0.92285900
C	-0.61677400	-2.42256700	-0.47412500
H	-1.32510600	-3.08355400	-0.97436000
C	-2.03668000	-0.66030300	-1.32308700
C	-3.51046800	-1.18105600	-3.02249600
H	-3.90966000	-1.95722100	-3.67175500
C	-3.97265800	0.14437800	-3.09963800
H	-4.74293000	0.44792700	-3.80059000
C	-3.38155200	1.05568500	-2.25772900
H	-3.64069500	2.10897000	-2.26238500
C	0.77912700	-3.45645900	-2.25596400
H	-0.00355800	-4.20742700	-2.26149900
C	1.86492600	-3.51298200	-3.09653300
H	1.98788400	-4.33188100	-3.79730400
C	2.78179700	-2.45018200	-3.01829000
H	3.65436900	-2.40798400	-3.66648600
C	1.59198400	-1.43388200	-1.32045300
N	-1.68590600	0.93524700	1.80449400
N	-0.36185500	1.50494300	-0.38873100
N	-0.06032600	2.99463400	-2.16263400
N	1.79154200	1.75843200	-1.36160400
N	1.65083000	0.99097600	1.80719100
N	1.48496500	-0.43861800	-0.38713000
N	2.62669000	-1.44291300	-2.16042300
N	0.62880200	-2.42901500	-1.36328600
N	0.03038600	-1.92683300	1.80559200
N	-1.12203700	-1.06613100	-0.38933600
N	-2.56112900	-1.55109300	-2.16434100
N	-2.41701000	0.67141000	-1.36492100
Ti	-0.00064600	-0.00004500	0.86769200

Table S15. Optimized structure of $[V(L^{I2})_3]^{2+}$ (Energy= -1885.146746 Hartree, Charge= 2, Multiplicity=4)

Atomic Number	Coordinates (Angstroms) X	Y	Z
C	1.65222100	0.76143500	-2.25602800
N	2.01078300	1.71610700	-1.39874600
C	3.16528600	2.34287700	-1.66646300
C	3.94762000	2.02111500	-2.77308700
C	3.46747700	1.00201500	-3.59766300
N	2.32082200	0.36417200	-3.34136700
H	3.46213000	3.12105200	-0.96728300
H	4.87996200	2.53268700	-2.98345800
H	4.01227900	0.68338700	-4.48336200
N	0.41368100	0.11641700	-1.95693000
C	-3.18701000	-2.73494500	-3.30999800
C	-3.71034700	-2.60939700	-2.02447400
C	-2.00129700	-2.06539000	-3.61648900
C	-3.03148300	-1.82169400	-1.09086300
H	-4.62904000	-3.11049700	-1.73861500
C	-1.38044800	-1.29673000	-2.62876100
H	-1.55686500	-2.13519200	-4.60430200
N	-1.89105500	-1.17389500	-1.37231700
H	-3.68856100	-3.34011700	-4.05851100
C	-0.13755400	-0.58313100	-2.89218500
H	-3.41409400	-1.70507300	-0.08251200
C	1.28287300	-2.56787800	0.39625500
N	1.99799900	-2.00477700	-0.57777200
C	3.05738500	-2.70570100	-1.00823100
C	3.39569400	-3.94734500	-0.47647200
C	2.57040800	-4.43341500	0.53991900
N	1.51227900	-3.74693200	0.97921800
H	3.64133900	-2.25197100	-1.80535100
H	4.25135000	-4.50909800	-0.83349100
H	2.75638900	-5.39473500	1.01302200
N	0.15961400	-1.80435300	0.83866300
C	-3.33590100	-1.08330100	4.03291100

C	-3.68935500	0.06291300	3.32337000
C	-2.26971100	-1.85474100	3.56768800
C	-2.96702900	0.39774500	2.17462500
H	-4.50970700	0.69502600	3.64629800
C	-1.59411200	-1.45339000	2.41258100
H	-1.95875600	-2.75430400	4.08940600
N	-1.94151300	-0.33541200	1.71783900
H	-3.87607300	-1.37157900	4.92910400
C	-0.46490700	-2.21240900	1.89397900
H	-3.21829900	1.28333800	1.60032700
C	1.51268600	1.44870900	2.04719900
N	1.63126000	0.24952400	2.61852900
C	2.66275800	0.10516500	3.46335700
C	3.56341300	1.13439000	3.72665700
C	3.33849100	2.34026900	3.06157100
N	2.31473600	2.50335500	2.21730900
H	2.76375200	-0.86717300	3.94004000
H	4.39538900	1.00578500	4.40960400
H	3.99227700	3.19780100	3.20261000
N	0.39922600	1.59937000	1.16589600
C	-2.63775800	4.50173300	-1.13785500
C	-3.19592800	3.37354500	-1.73595300
C	-1.58076900	4.32911400	-0.24273500
C	-2.67820300	2.11428400	-1.42114900
H	-4.01929200	3.45667200	-2.43736600
C	-1.11735200	3.03783600	0.02003600
H	-1.11572100	5.17919000	0.24636300
N	-1.66185500	1.93569100	-0.56449300
H	-3.01531800	5.49447700	-1.36136500
C	-0.01448200	2.80182400	0.94228600
H	-3.09110400	1.21736200	-1.87062800
H	0.46429600	3.65539000	1.42127300
H	-0.14796200	-3.11586900	2.41376400
H	0.32595500	-0.66671500	-3.87482100
V	-0.66531100	0.05792500	-0.01894600

Table S16. Optimized structure of **[V(L^M)]$^{2+}$** (Energy= -1885.126255 Hartree, Charge= 2, Multiplicity=4)

Atomic Number	Coordinates (Angstroms) X	Y	Z
C	-3.12966400	-0.46272700	-2.39105900
H	-3.83696100	-0.72325100	-1.60946200
C	-3.49855900	-0.50321800	-3.73375900
H	-4.50661200	-0.79229100	-4.01091700
C	-2.55095400	-0.16120300	-4.69661800
H	-2.80320100	-0.17070800	-5.75231700
C	-1.26814800	0.20151500	-4.27927300
H	-0.51204600	0.48294400	-5.00574700
C	-0.97548000	0.21110100	-2.91836900
C	0.41107100	0.59204300	-2.41043400
H	0.89254300	1.26073900	-3.12639700
C	1.29665800	1.97778300	-0.70546400
C	3.15822800	3.28289800	-1.15808200
H	3.88987600	3.60256600	-1.89782800
C	3.21134500	3.78224200	0.16114900
H	3.96030700	4.49508500	0.48126200
C	2.28899300	3.27614700	1.03648700
H	2.27366700	3.54559700	2.08624600
C	-3.12284400	2.30615500	0.81033100
H	-3.83399800	1.76029400	0.19769400
C	-3.48696500	3.48991300	1.44807700
H	-4.49506800	3.87575600	1.34130600
C	-2.53458200	4.15176500	2.22068500
H	-2.78298300	5.07127900	2.74126900
C	-1.25198900	3.60730000	2.31985100
H	-0.49219300	4.09493000	2.92283200
C	-0.96425700	2.42336000	1.64660700
C	0.42182000	1.79124200	1.71590300
H	0.90821500	2.07658800	2.65056100
C	-3.11678100	-1.85188000	1.60754000
H	-3.82793000	-1.04762100	1.44488800
C	-3.47853200	-2.99582000	2.31543200

H	-4.48478000	-3.09531500	2.70800400
C	-2.52624800	-3.99685200	2.49734500
H	-2.77287900	-4.90729600	3.03445200
C	-1.24606700	-3.81164500	1.96994600
H	-0.48645100	-4.57843800	2.08683000
C	-0.96056800	-2.63673000	1.28011400
C	0.42282900	-2.38190900	0.69124000
H	0.90726600	-3.33440600	0.46843000
C	1.29769700	-1.59513000	-1.36473100
C	3.15939300	-2.63315200	-2.27582900
H	3.89422500	-3.43102800	-2.18562600
C	3.20513300	-1.74017000	-3.36808700
H	3.95134400	-1.81676200	-4.14838300
C	2.27926400	-0.73229100	-3.36392000
H	2.25832400	0.04202100	-4.12209300
C	2.29781000	-2.53805900	2.30767700
H	2.27986900	-3.58189600	2.01626400
C	3.22516700	-2.03404300	3.17890100
H	3.97577000	-2.66850000	3.63235200
C	3.17471600	-0.64183300	3.40633500
H	3.91017600	-0.16183000	4.04930800
C	1.30661000	-0.37918200	2.05919500
N	-1.89814900	-0.10962000	-1.98248500
N	0.32583200	1.17255300	-1.09891300
N	2.25815400	2.41182400	-1.56570700
N	1.33988500	2.38233600	0.63938100
N	-1.89152400	1.77416200	0.90580300
N	0.33399900	0.36525400	1.56363000
N	2.27269600	0.14770600	2.86041100
N	1.34666900	-1.74619400	1.73696300
N	-1.88782300	-1.66965500	1.09303200
N	0.33005800	-1.53679500	-0.46697700
N	2.26279800	-2.55371300	-1.31413500
N	1.33359100	-0.63256300	-2.38762200
V	-1.01827700	-0.00092300	0.00305500

Table S17. Optimized structure of $[Cr(L^{I2})_3]^{2+}$ (Energy= -1900.078879 Hartree, Charge= 2, Multiplicity=3)

Atomic Number	Coordinates (Angstroms) X	Y	Z
C	1.78850400	1.60215100	-1.44827600
N	2.12877800	2.13502900	-0.27470800
C	3.33915800	2.70919700	-0.21866600
C	4.19255800	2.76051000	-1.31771400
C	3.71898100	2.18107400	-2.49552300
N	2.51963900	1.59450900	-2.56586400
H	3.62404700	3.14019900	0.73812300
H	5.16864000	3.22882300	-1.26344300
H	4.31171800	2.18117500	-3.40746700
N	0.49233900	1.00572600	-1.50860400
C	-3.34876200	-0.29784000	-4.00927900
C	-3.92569300	-0.66405600	-2.78610100
C	-2.06007800	0.21988000	-4.00377500
C	-3.19517900	-0.51498000	-1.61271100
H	-4.93243600	-1.06530100	-2.73905800
C	-1.37760800	0.35427400	-2.78605000
H	-1.57033900	0.52393900	-4.92348700
N	-1.93781400	-0.01975300	-1.58991300
H	-3.89652500	-0.41339300	-4.93870600
C	-0.06206000	0.91386600	-2.68990000
H	-3.62210200	-0.79506000	-0.65775500
C	0.96203400	-2.32206800	-1.08144500
N	2.00030900	-1.49537400	-1.20677300
C	3.04084000	-1.96795800	-1.90758100
C	3.04069200	-3.23884300	-2.47847000
C	1.89373900	-4.00859700	-2.28058300
N	0.84898600	-3.55484300	-1.57999700
H	3.89273800	-1.30117800	-2.01288000
H	3.88490400	-3.60936400	-3.04862400
H	1.80264800	-5.01120700	-2.69149300
N	-0.12900100	-1.82147600	-0.30976200
C	-3.72083100	-2.51445800	2.73445900

C	-3.73385400	-1.12656800	2.91701000
C	-2.82425100	-3.06385300	1.82413100
C	-2.86202100	-0.33866600	2.17046900
H	-4.41238200	-0.65817100	3.62161300
C	-1.96052400	-2.21457600	1.11929000
H	-2.77513000	-4.13533400	1.65868400
N	-1.99557300	-0.85736700	1.28211200
H	-4.39576800	-3.15210600	3.29645900
C	-0.94031200	-2.69475500	0.22486300
H	-2.85795700	0.74063700	2.27524400
C	1.51720900	-0.26192000	2.41251000
N	1.28451100	-1.57274800	2.37925700
C	2.18022100	-2.33672700	3.02140700
C	3.28945800	-1.80099300	3.67257000
C	3.42397400	-0.41309000	3.63157300
N	2.53760900	0.36543400	3.00066200
H	1.99625200	-3.40843500	3.01065800
H	4.00949200	-2.42885000	4.18493100
H	4.25834600	0.09252200	4.11166100
N	0.54388200	0.55826300	1.75694700
C	-1.74386600	4.73961400	1.35960600
C	-2.49822700	4.16978700	0.33633900
C	-0.76024200	3.96449900	1.97904800
C	-2.24312800	2.84816700	-0.04149900
H	-3.27421100	4.73209500	-0.17194400
C	-0.56677000	2.65147400	1.55079600
H	-0.15163500	4.36700500	2.78246300
N	-1.30215000	2.09812600	0.54829900
H	-1.91603600	5.76454400	1.67249500
C	0.41950900	1.77420400	2.16800100
H	-2.80613100	2.37688600	-0.83923800
H	1.03629400	2.15266700	2.98233800
H	-0.81537500	-3.75803000	0.03752800
H	0.46303400	1.26038400	-3.57651000
Cr	-0.66097900	0.14476600	0.02457600

Table S18. Optimized structure of **[Cr(L^{M})]$^{2+}$** (Energy= -1900.058993 Hartree, Charge= 2, Multiplicity=3)

Atomic Number	Coordinates (Angstroms) X	Y	Z
C	-3.25907700	-0.17677300	-1.98184600
H	-3.84642100	-0.59750900	-1.17148500
C	-3.81395000	0.01137200	-3.24453000
H	-4.85131700	-0.25249900	-3.42106400
C	-3.01765200	0.53670000	-4.26165700
H	-3.41894300	0.70060600	-5.25633300
C	-1.68751100	0.85167200	-3.97037000
H	-1.04009100	1.27128400	-4.73448600
C	-1.20403600	0.63637400	-2.68514800
C	0.22866400	0.96314400	-2.29820200
H	0.62886100	1.74933100	-2.94037000
C	1.27085800	2.06940400	-0.46676200
C	3.07733500	3.46660600	-0.84986600
H	3.73505500	3.92577200	-1.58548200
C	3.25447300	3.72461400	0.52602200
H	4.03317200	4.37855700	0.89700900
C	2.40734700	3.06941300	1.37921200
H	2.48031100	3.15682100	2.45717900
C	-3.01408800	2.04789000	1.24032300
H	-3.73326000	1.57403500	0.57940000
C	-3.37672400	3.12603900	2.04363800
H	-4.39540700	3.49803800	2.02519100
C	-2.40699000	3.70576500	2.86091200
H	-2.65444100	4.54265400	3.50644700
C	-1.10967100	3.18950100	2.84035600
H	-0.33622800	3.61624700	3.47133300
C	-0.82589900	2.10777700	2.01087000
C	0.56685300	1.49554100	1.93695200
H	1.09547200	1.64635600	2.88064400
C	-2.95838200	-1.95715300	1.48075100
H	-3.63356000	-1.10732000	1.50655400
C	-3.33656800	-3.18921200	2.01237900

H	-4.32010400	-3.30740800	2.45431800
C	-2.43535200	-4.24919400	1.95602600
H	-2.69863700	-5.22539000	2.35034300
C	-1.17917400	-4.03235400	1.37912300
H	-0.45485500	-4.83856200	1.31537700
C	-0.87111900	-2.77193100	0.88342400
C	0.48261000	-2.45051200	0.26777100
H	0.93575900	-3.35458500	-0.14333300
C	1.20354200	-1.34331600	-1.70741800
C	2.95146600	-2.22043600	-2.94550200
H	3.67698700	-3.02385100	-3.05796100
C	2.91118900	-1.15916300	-3.87629000
H	3.58004300	-1.10875400	-4.72575700
C	2.00323100	-0.16618400	-3.62392000
H	1.91874700	0.71971500	-4.24298700
C	2.44777300	-2.87064200	1.72405800
H	2.40104900	-3.85580100	1.27430000
C	3.42989100	-2.50953900	2.60658900
H	4.19931600	-3.20799400	2.90959900
C	3.40771200	-1.17070300	3.05255300
H	4.18523900	-0.79814000	3.71669300
C	1.46493100	-0.70109600	1.88276900
N	-1.97948100	0.13279000	-1.68993900
N	0.27705200	1.31486200	-0.90357200
N	2.14269900	2.66592900	-1.32234500
N	1.42284300	2.25021100	0.91540700
N	-1.76890100	1.54226900	1.22299300
N	0.46909800	0.11142400	1.56955600
N	2.48102700	-0.30437400	2.69367400
N	1.47381300	-1.99663800	1.34298900
N	-1.75520600	-1.74536100	0.91743100
N	0.32870300	-1.42227500	-0.72011300
N	2.14965200	-2.30024900	-1.90276200
N	1.15525900	-0.22982300	-2.56007000
Cr	-0.91386400	-0.01810900	0.08161800

Table S19. Optimized structure of $[Mn(L^{12})_3]^{2+}$ (Energy= -1917.669807 Hartree, Charge= 2, Multiplicity=2)

Atomic Number	Coordinates (Angstroms) X	Y	Z
C	1.85012200	1.21914100	-1.70942100
N	2.08796900	2.11695500	-0.75630100
C	3.28409300	2.72119500	-0.80569900
C	4.22246100	2.43678200	-1.79463000
C	3.85224600	1.48450900	-2.74470400
N	2.66674100	0.86612600	-2.70483400
H	3.48642000	3.45287300	-0.02751400
H	5.18711300	2.93030300	-1.82703900
H	4.51770500	1.20390400	-3.55789300
N	0.56091500	0.59158600	-1.66798100
C	-3.08693400	-1.47328400	-3.93674400
C	-3.70564000	-1.58524800	-2.69202400
C	-1.82339000	-0.88705200	-4.00093100
C	-3.04265700	-1.11623400	-1.55590400
H	-4.68957700	-2.03076700	-2.59097600
C	-1.22416900	-0.43825600	-2.82115400
H	-1.30232800	-0.77389300	-4.94631600
N	-1.82508800	-0.55174500	-1.60387700
H	-3.57686700	-1.83185600	-4.83630700
C	0.07978200	0.19582300	-2.80106000
H	-3.50224000	-1.19260900	-0.57759500
C	0.97671700	-2.54260700	-0.38017900
N	2.00937700	-1.79097600	-0.75882100
C	3.03486900	-2.44938600	-1.31864500
C	3.02343500	-3.83050500	-1.50052800
C	1.88197800	-4.50630200	-1.06670800
N	0.85112500	-3.86563200	-0.50459700
H	3.88105700	-1.84349600	-1.63158500
H	3.85506000	-4.35316200	-1.95925300
H	1.78308700	-5.58394500	-1.17269200
N	-0.10105900	-1.83523700	0.23688600
C	-3.72189800	-1.62482100	3.31636100

C	-3.82953800	-0.27527400	2.98119300
C	-2.74300100	-2.39272500	2.68614100
C	-2.96114100	0.26141500	2.02795900
H	-4.57277600	0.36256800	3.44763000
C	-1.90886200	-1.78508400	1.74500100
H	-2.61890000	-3.44609000	2.91636800
N	-2.01890800	-0.46888300	1.41211900
H	-4.38348900	-2.06905000	4.05314000
C	-0.84371400	-2.49414000	1.06477800
H	-3.02293200	1.30647800	1.74677800
C	1.42043200	0.43542600	2.38170200
N	1.20442400	-0.82300000	2.76465000
C	2.11426000	-1.34002400	3.60321500
C	3.22294200	-0.61975400	4.04187400
C	3.34042300	0.68609900	3.56706900
N	2.44071700	1.22176500	2.73407000
H	1.94203600	-2.36249700	3.93145900
H	3.95377800	-1.05020800	4.71695100
H	4.17267100	1.32363700	3.85595700
N	0.43593400	1.00952000	1.51388500
C	-1.95729100	4.82133000	-0.04976300
C	-2.58207400	3.96750300	-0.95843900
C	-1.01201900	4.28696000	0.82529600
C	-2.24250400	2.61305400	-0.96463500
H	-3.32390200	4.33664900	-1.65861400
C	-0.72410400	2.92212200	0.75895400
H	-0.49985900	4.91146200	1.55030900
N	-1.33537300	2.08786300	-0.12621400
H	-2.20061900	5.87864300	-0.02250500
C	0.24425600	2.28213500	1.62792600
H	-2.71181200	1.92663500	-1.66008700
Mn	-0.67239200	0.14029700	-0.02295200
H	0.79614000	2.87257800	2.35692100
H	-0.66868400	-3.54981600	1.26282000
H	0.63682100	0.32906300	-3.72685400

Table S20. Optimized structure of **[Mn(L^{M})]$^{2+}$** (Energy= -1917.661665 Hartree, Charge= 2, Multiplicity=2)

Atomic Number	Coordinates (Angstroms) X	Y	Z
C	3.04008400	-1.56596900	1.55220100
H	3.66843300	-1.40960300	0.68103100
C	3.51274200	-2.26476800	2.66168200
H	4.52672100	-2.65009500	2.66502200
C	2.66296600	-2.45216000	3.74977200
H	2.99980700	-2.98491200	4.63330200
C	1.36441700	-1.94035600	3.68261800
H	0.67645700	-2.06668300	4.51290200
C	0.96731700	-1.25607000	2.53913600
C	-0.42930800	-0.68439000	2.36988600
H	-0.87981500	-0.46309100	3.33907000
C	-1.31298600	1.37799700	1.59039200
C	-3.14538600	2.31421800	2.65161400
H	-3.86357300	2.24157400	3.46617600
C	-3.20471200	3.39247000	1.74207100
H	-3.94664900	4.17654300	1.82157600
C	-2.29345600	3.37412400	0.72069700
H	-2.27645400	4.12407800	-0.06191600
C	3.04365600	2.12168300	0.58131000
H	3.67013100	1.28791100	0.88222400
C	3.51893400	3.43098800	0.63130200
H	4.53314800	3.62466800	0.96372100
C	2.67142000	4.46850500	0.24861600
H	3.01034900	5.49936400	0.26781200
C	1.37242500	4.15687100	-0.16145800
H	0.68625900	4.94033800	-0.46800900
C	0.97261900	2.82523800	-0.18180100
C	-0.42466900	2.39537500	-0.59271500
H	-0.87360400	3.12477400	-1.26923400
C	3.04390800	-0.56254200	-2.12650400
H	3.67139900	0.11340700	-1.55420100
C	3.51850100	-1.17404700	-3.28559500

H	4.53320900	-0.98471000	-3.61900400
C	2.66966700	-2.02216600	-3.99364700
H	3.00798300	-2.52097600	-4.89622500
C	1.37013800	-2.21939400	-3.51932100
H	0.68292800	-2.87497400	-4.04522800
C	0.97111100	-1.57123400	-2.35563300
C	-0.42663200	-1.71019200	-1.77849400
H	-0.87681300	-2.66005200	-2.07234500
C	-1.31477100	-2.06566500	0.39557400
C	-3.14918900	-3.45095000	0.67205100
H	-3.86709900	-4.11930700	0.20036600
C	-3.21102500	-3.20242700	2.06050900
H	-3.95472000	-3.66255200	2.69829400
C	-2.29978700	-2.30972000	2.55714100
H	-2.28443800	-2.00708000	3.59799600
C	-2.29480700	-1.05931300	-3.28228400
H	-2.27947200	-2.11208900	-3.54040100
C	-3.20506700	-0.18265400	-3.80842900
H	-3.94790100	-0.50480800	-4.52674600
C	-3.14344200	1.14411700	-3.32957800
H	-3.86062800	1.88697500	-3.67365600
C	-1.31110100	0.69049800	-1.98893600
N	1.79670400	-1.05881200	1.48786500
N	-0.36054900	0.46530000	1.51301500
N	-2.25549600	1.34562500	2.57061800
N	-1.35514200	2.39162600	0.62172600
N	1.79987200	1.81468900	0.17376000
N	-0.35799400	1.07832800	-1.15987400
N	-2.25245200	1.55696100	-2.45076200
N	-1.35538900	-0.65512500	-2.38248600
N	1.79957500	-0.75993000	-1.65748200
N	-0.36028300	-1.54320200	-0.35423500
N	-2.25705000	-2.89746000	-0.12442900
N	-1.35930000	-1.73367300	1.75766100
Mn	0.86661900	-0.00069600	0.00067100

Table S21. Optimized structure of $[Fe(L^{I2})_3]^{2+}$ (Energy= -1937.194313 Hartree, Charge= 2, Multiplicity=1)

Atomic Number	Coordinates (Angstroms) X	Y	Z
C	1.82141800	1.35238900	-1.57702600
N	1.99327900	2.27538800	-0.63396600
C	3.16507500	2.92705800	-0.65808800
C	4.14379400	2.66533500	-1.61324700
C	3.83999500	1.68586500	-2.55899100
N	2.67962200	1.02052700	-2.54419900
H	3.31353500	3.68039700	0.11142500
H	5.08816900	3.19745300	-1.62536900
H	4.53913800	1.42228700	-3.34931800
N	0.55408100	0.67812800	-1.57152400
C	-3.01077800	-1.36766900	-3.99009600
C	-3.66809600	-1.50398800	-2.76933100
C	-1.74855000	-0.77250200	-4.00293200
C	-3.04353300	-1.05144300	-1.60322800
H	-4.65329400	-1.95386300	-2.70746700
C	-1.19127600	-0.34431900	-2.79673500
H	-1.19856700	-0.63840300	-4.92899400
N	-1.82887700	-0.48387600	-1.60286400
H	-3.46920400	-1.71243600	-4.91141800
C	0.10398400	0.31081700	-2.72439100
H	-3.53489300	-1.14457400	-0.64214700
C	1.06979300	-2.42998000	-0.51346500
N	2.12530000	-1.65300200	-0.74851100
C	3.17532600	-2.25751200	-1.32308600
C	3.16483500	-3.60891300	-1.66192700
C	1.99657100	-4.31436300	-1.37176000
N	0.94023300	-3.72838800	-0.79602300
H	4.04203300	-1.63163000	-1.51828400
H	4.01712700	-4.08660800	-2.13163600
H	1.89558200	-5.37123700	-1.60657300
N	-0.03806100	-1.79223800	0.12831700
C	-3.68172400	-1.89230800	3.18745800

C	-3.85187900	-0.53718700	2.91174500
C	-2.66661000	-2.58529800	2.52570500
C	-3.00919400	0.08107500	1.98282400
H	-4.62467600	0.04458900	3.40280000
C	-1.86233500	-1.89727300	1.61673000
H	-2.49240400	-3.64051800	2.71037500
N	-2.03475400	-0.57651400	1.34044700
H	-4.32179800	-2.39933200	3.90239300
C	-0.75995700	-2.52389300	0.90800900
H	-3.12212700	1.13221900	1.74455500
C	1.41891700	0.29069400	2.34256500
N	1.18794400	-0.96257600	2.73438300
C	2.11830600	-1.50177900	3.53535500
C	3.26151000	-0.80891200	3.92758200
C	3.39003200	0.49427500	3.44944500
N	2.46931800	1.05336600	2.65489500
H	1.93417000	-2.51939800	3.87210700
H	4.00841600	-1.25736900	4.57269600
H	4.24762900	1.11196900	3.70545600
N	0.41089400	0.89422700	1.52309900
C	-2.08023400	4.73853300	0.20991900
C	-2.73011100	3.91578300	-0.70783500
C	-1.09448700	4.18030700	1.02544000
C	-2.37558600	2.56573000	-0.78268600
H	-3.50383900	4.30304700	-1.36217500
C	-0.79420800	2.82499300	0.88915100
H	-0.56137800	4.78115700	1.75528700
N	-1.43043500	2.02131800	-0.00415300
H	-2.33410700	5.79061900	0.29111400
C	0.21224700	2.15619800	1.69806800
H	-2.86527600	1.90107300	-1.48494300
H	0.77955700	2.72113300	2.43551500
H	-0.54323100	-3.58035800	1.05424200
H	0.67514900	0.48678500	-3.63455000
Fe	-0.70749900	0.13031600	-0.01328600

Table S22. Optimized structure of **[Fe(L^{M})]$^{2+}$** (Energy= -1937.193209 Hartree, Charge= 2, Multiplicity=1)

Atomic Number	Coordinates (Angstroms) X	Y	Z
C	3.02533200	-1.98863100	-0.77117300
H	3.63787100	-1.11988100	-0.98812000
C	3.51759600	-3.27937700	-0.95878600
H	4.53153300	-3.42405500	-1.31602400
C	2.68718800	-4.36200900	-0.67722300
H	3.04012400	-5.38073200	-0.80264000
C	1.38750700	-4.11251800	-0.22978100
H	0.71395800	-4.93240200	-0.00031300
C	0.97035900	-2.79485000	-0.07390100
C	-0.43248200	-2.42666800	0.38016600
H	-0.86462100	-3.21669600	0.99718200
C	-1.32001500	-0.85327900	1.92376500
C	-3.12678500	-1.42887200	3.25074800
H	-3.83199000	-2.20320000	3.54699300
C	-3.19507800	-0.14284100	3.83178000
H	-3.93374400	0.11518000	4.57962400
C	-2.29499000	0.77935200	3.37076900
H	-2.28070300	1.80779100	3.71355300
C	3.02644800	0.32551600	2.10608400
H	3.63844500	-0.29674900	1.46168500
C	3.51960600	0.80826300	3.31740300
H	4.53372700	0.57109700	3.62059600
C	2.68981600	1.59342900	4.11483900
H	3.04343800	1.99404900	5.05958400
C	1.38987200	1.85638700	3.67595900
H	0.71687600	2.46524600	4.27169700
C	0.97188500	1.33284200	2.45704100
C	-0.43116100	1.54255400	1.91181000
H	-0.86279100	2.47205100	2.28776800
C	3.02604900	1.66100000	-1.33618300
H	3.63842700	1.41427000	-0.47528100
C	3.51877000	2.46862500	-2.36015100

H	4.53287900	2.84986700	-2.30673600
C	2.68856700	2.76658500	-3.43855300
H	3.04183700	3.38448500	-4.25799500
C	1.38865500	2.25492700	-3.44635700
H	0.71523100	2.46652200	-4.27114000
C	0.97112800	1.46110100	-2.38331400
C	-0.43198300	0.88433800	-2.29161700
H	-0.86416800	0.74533200	-3.28434800
C	-1.32020100	-1.23907300	-1.70118900
C	-3.12725600	-2.09970500	-2.86328200
H	-3.83218600	-1.96891900	-3.68220100
C	-3.19638900	-3.24566600	-2.03978400
H	-3.93549500	-4.02192100	-2.19019200
C	-2.29663200	-3.30772600	-1.01036100
H	-2.28305900	-4.11858500	-0.29083200
C	-2.29522100	2.53021800	-2.35921400
H	-2.28161300	2.31269700	-3.42123700
C	-3.19454900	3.39113200	-1.79071800
H	-3.93326200	3.91008000	-2.38777000
C	-3.12556000	3.53099100	-0.38649600
H	-3.83027700	4.17499400	0.13628200
C	-1.31927000	2.09330700	-0.22229000
N	1.78169600	-1.74273500	-0.32807100
N	-0.38000500	-1.16031300	1.05164000
N	-2.24538500	-1.76481200	2.33116800
N	-1.36337800	0.45358400	2.43196800
N	1.78261100	0.58664500	1.67237300
N	-0.37935200	1.49100900	0.47932400
N	-2.24408300	2.90222300	0.36383800
N	-1.36357800	1.87963600	-1.60810700
N	1.78219600	1.15482300	-1.34497000
N	-0.37991800	-0.33056600	-1.53090100
N	-2.24533500	-1.13581700	-2.69448400
N	-1.36446700	-2.33232100	-0.82313800
Fe	0.81751300	-0.00023500	-0.00017500

Table S23. Optimized structure of **[Fe(L^{I2})$_3$]$^{3+}$** (Energy= -1936.756075 Hartree, Charge= 3, Multiplicity=2)

Atomic Number	Coordinates (Angstroms) X	Y	Z
C	1.92985900	1.65452200	-1.09249900
N	2.15265300	2.21379800	0.09057800
C	3.35619100	2.79449900	0.24214400
C	4.30215300	2.81718700	-0.77942400
C	3.94097100	2.20345800	-1.98076700
N	2.74988900	1.61082800	-2.13806200
H	3.55461500	3.25420900	1.20638000
H	5.26885200	3.29100600	-0.65028700
H	4.61234600	2.17983900	-2.83539900
N	0.62064200	1.07015400	-1.29547900
C	-3.00542900	-0.03721500	-4.18666900
C	-3.67398000	-0.49415400	-3.05228300
C	-1.71676300	0.48526500	-4.03995900
C	-3.03900200	-0.42829400	-1.80743500
H	-4.67828400	-0.90020100	-3.11583300
C	-1.14573800	0.52386700	-2.76794500
H	-1.16051000	0.85899200	-4.89407000
N	-1.80108000	0.06863200	-1.66134700
H	-3.47508700	-0.08127600	-5.16458400
C	0.17065500	1.06927400	-2.50760300
H	-3.54106500	-0.77621700	-0.91337800
C	0.88765700	-2.24292400	-1.21896700
N	1.97040800	-1.47696700	-1.28107400
C	2.98636200	-1.97789800	-2.00552900
C	2.90102200	-3.21222700	-2.64756200
C	1.70075000	-3.91400100	-2.50930400
N	0.68036500	-3.42551400	-1.79040600
H	3.88222900	-1.36693700	-2.06739100
H	3.72481000	-3.60752000	-3.23146900
H	1.54405200	-4.88024600	-2.98164500
N	-0.18174400	-1.74072200	-0.39697900
C	-3.81747300	-2.34363500	2.60585500

C	-3.85291000	-0.95392400	2.71132500
C	-2.87184200	-2.93045200	1.75867900
C	-2.95092900	-0.18602600	1.96747200
H	-4.56709100	-0.45634200	3.35926500
C	-1.99989200	-2.10545000	1.05088600
H	-2.80520000	-4.00856700	1.65073600
N	-2.04829100	-0.74560600	1.14830500
H	-4.50818400	-2.96087000	3.17227300
C	-0.95247700	-2.60041200	0.18222700
H	-2.96006200	0.89518800	2.02830400
C	1.33512100	-0.56410400	2.32752100
N	0.96259000	-1.83976000	2.34439800
C	1.78620100	-2.66963200	3.00895900
C	2.95171300	-2.22360500	3.62890500
C	3.23121400	-0.85973300	3.53071400
N	2.41856400	-0.02009600	2.87350800
H	1.49419700	-3.71591400	3.04548500
H	3.60750700	-2.90207400	4.16305400
H	4.11795200	-0.42349400	3.98315400
N	0.43163000	0.36402400	1.68577000
C	-1.64632700	4.64041900	1.41284000
C	-2.34859300	4.14146400	0.31631900
C	-0.72138900	3.81075800	2.05415300
C	-2.11067900	2.83170100	-0.11172400
H	-3.07763100	4.74942300	-0.20936600
C	-0.53347200	2.51660600	1.57280400
H	-0.15368100	4.15836600	2.91156200
N	-1.22773300	2.03179700	0.50534800
H	-1.81413300	5.65387800	1.76440100
C	0.38742300	1.56536900	2.16290300
H	-2.64497500	2.41756200	-0.95814800
H	1.00316700	1.84262900	3.01585200
H	-0.80970800	-3.66749600	0.02368600
H	0.77246700	1.47569400	-3.31850400
Fe	-0.66785400	0.16662100	0.01799200

Table S24. Optimized structure of **[Fe(L^{M})]$^{3+}$** (Energy= -1936.787253 Hartree, Charge= 3, Multiplicity=2)

Atomic Number	Coordinates (Angstroms) X	Y	Z
C	-2.96898400	2.10120200	-0.16275600
H	-3.54719500	1.35390400	-0.69489300
C	-3.49183400	3.36705400	0.09513500
H	-4.49733100	3.61091500	-0.23106200
C	-2.70538300	4.29610900	0.77552700
H	-3.08895000	5.28624400	1.00181800
C	-1.40984900	3.93512600	1.15955600
H	-0.77236400	4.63887600	1.68553700
C	-0.95607100	2.65556800	0.86411100
C	0.44273400	2.16282600	1.17565600
H	0.87704600	2.67688900	2.03368900
C	1.31676000	0.13677600	2.11913200
C	3.06925200	0.25350000	3.61685300
H	3.74254600	0.89382000	4.18248900
C	3.15336200	-1.15000700	3.72062300
H	3.87865000	-1.63887900	4.35971200
C	2.28342100	-1.87417000	2.94885100
H	2.27969500	-2.95821100	2.92422800
C	-2.96986100	-0.90315800	1.90280100
H	-3.54634700	-0.06716900	1.52233900
C	-3.49428500	-1.75871500	2.86991000
H	-4.49928600	-1.59608500	3.24464100
C	-2.71001700	-2.81445100	3.33352100
H	-3.09485400	-3.50499100	4.07766900
C	-1.41497800	-2.96916000	2.82840200
H	-0.77919700	-3.77815300	3.17426700
C	-0.95955000	-2.07410700	1.86823700
C	0.43895400	-2.10042200	1.28512000
H	0.87124800	-3.10141300	1.30096700
C	-2.97327400	-1.19037600	-1.73165700
H	-3.54941900	-1.27791000	-0.81712800
C	-3.49919600	-1.59905500	-2.95585600

H	-4.50496200	-2.00306400	-3.00178200
C	-2.71545900	-1.47393300	-4.10244700
H	-3.10146300	-1.77230000	-5.07233300
C	-1.41940900	-0.96151000	-3.98463400
H	-0.78395900	-0.85762500	-4.85854200
C	-0.96246800	-0.57854800	-2.72964400
C	0.43722800	-0.06334900	-2.46168400
H	0.87008500	0.42236800	-3.33679300
C	1.31522400	1.76606700	-1.18064200
C	3.06638600	3.00396300	-2.03446300
H	3.73781600	3.17344200	-2.87335400
C	3.15360500	3.79537200	-0.87096500
H	3.87953400	4.59289700	-0.76872800
C	2.28584600	3.48940300	0.14403700
H	2.28453600	4.00999300	1.09522000
C	2.27739400	-1.62179500	-3.09906500
H	2.27416200	-1.05851100	-4.02559800
C	3.14497300	-2.65443200	-2.85859600
H	3.86879400	-2.96530000	-3.60217900
C	3.06053900	-3.26603900	-1.59112100
H	3.73213900	-4.07764300	-1.31995200
C	1.31193600	-1.90632800	-0.94184000
N	-1.73213300	1.74617600	0.22859800
N	0.38733800	0.73913800	1.36529300
N	2.19443600	0.86649300	2.83391300
N	1.36507300	-1.25210200	2.15001200
N	-1.73356800	-1.06713600	1.39911800
N	0.38397200	-1.55260300	-0.04255900
N	2.18749900	-2.89226900	-0.66799900
N	1.36088200	-1.23867500	-2.16009100
N	-1.73593500	-0.67448100	-1.62252500
N	0.38477300	0.81275400	-1.32344000
N	2.19079600	2.01984000	-2.17194100
N	1.36660900	2.48711700	0.00667000
Fe	-0.75042700	0.00068000	0.00083900

Table S25. Optimized structure of $[Co(L^{I2})_3]^{2+}$ (Energy= -1958.821906 Hartree, Charge= 2, Multiplicity=2)

Atomic Number	Coordinates (Angstroms) X	Y	Z
C	-2.60333600	1.43744600	0.34773300
N	-2.80467800	1.38679700	-0.97223500
C	-4.08345300	1.43278300	-1.37034700
C	-5.14347900	1.52185400	-0.47037400
C	-4.80733700	1.56364000	0.88273800
N	-3.53696900	1.51898500	1.30072900
H	-4.25735000	1.39524500	-2.44300100
H	-6.17446900	1.55605000	-0.80369300
H	-5.56960600	1.63332100	1.65550700
N	-1.24184800	1.40693800	0.75178100
C	2.17105500	2.62183500	3.88512900
C	3.09717800	1.94856000	3.09205100
C	0.83559800	2.63762000	3.48128500
C	2.65544700	1.31140300	1.93002400
H	4.14772900	1.90850800	3.35932300
C	0.46668400	1.98030700	2.30509400
H	0.07948400	3.15237100	4.06569800
N	1.37335700	1.31981800	1.53647100
H	2.47867600	3.12508000	4.79632100
C	-0.92479900	1.99316700	1.84676200
H	3.34789600	0.77779600	1.28986000
C	-0.35136400	-1.76988700	1.93621200
N	-1.59040600	-1.37783200	1.64720700
C	-2.54275900	-1.78754800	2.49831900
C	-2.25807500	-2.56973800	3.61562700
C	-0.91984100	-2.91292100	3.81093600
N	0.04315700	-2.51193700	2.97281100
H	-3.55700700	-1.46936900	2.27254300
H	-3.03441500	-2.89113500	4.30061100
H	-0.60367200	-3.51688700	4.65800700
N	0.66164900	-1.34865000	1.01634800
C	4.81574500	-2.28837800	-1.19067900

C	4.68005700	-1.07864100	-1.86735200
C	3.83874500	-2.64944000	-0.25942900
C	3.57401200	-0.26585700	-1.58521800
H	5.41308100	-0.75859400	-2.60045700
C	2.76394800	-1.78279900	-0.04697500
H	3.90401700	-3.58452100	0.28820800
N	2.63744500	-0.60152400	-0.69607200
H	5.66223700	-2.94020900	-1.38219300
C	1.69262700	-2.11406600	0.88470600
H	3.44206500	0.68678900	-2.09077500
C	-0.87201200	-1.76696800	-1.93668300
N	-0.19573400	-2.80811900	-1.45173300
C	-0.73107600	-4.01314300	-1.69644700
C	-1.92196500	-4.17430300	-2.40088300
C	-2.53625300	-3.00739800	-2.85410000
N	-2.01478000	-1.79660700	-2.62552500
H	-0.18193600	-4.87133900	-1.31609900
H	-2.34642700	-5.15381100	-2.58940700
H	-3.46768600	-3.03558300	-3.41455700
N	-0.28353700	-0.47778700	-1.72451800
C	0.64218700	3.94744400	-3.22529300
C	1.35768000	4.12235700	-2.04302600
C	0.00148600	2.72652700	-3.44582000
C	1.40462600	3.07814000	-1.11441900
H	1.87596300	5.05119900	-1.83018200
C	0.09799700	1.73078400	-2.47501400
H	-0.56311600	2.54321600	-4.35419500
N	0.78974400	1.90678200	-1.31845300
H	0.58514300	4.74158300	-3.96290300
C	-0.49977000	0.41429000	-2.63123200
H	1.94826300	3.18505900	-0.18317100
H	-1.09489400	0.18706800	-3.51355700
H	1.76533600	-3.03053700	1.46820200
H	-1.66576400	2.50679700	2.46275100
Co	0.65212300	0.32449700	-0.09668400

Table S26. Optimized structure of **[Co(L^M)]$^{2+}$** (Energy= -1958.813880 Hartree, Charge= 2, Multiplicity=2)

Atomic Number	Coordinates (Angstroms) X	Y	Z
C	-2.90332300	-1.67828100	1.54943100
H	-3.61067700	-1.25728300	0.84250900
C	-3.33167800	-2.43861200	2.63608900
H	-4.38956100	-2.63091600	2.77886500
C	-2.37696500	-2.93896700	3.51900500
H	-2.67188000	-3.54325200	4.37116500
C	-1.02992300	-2.64775800	3.29126400
H	-0.26229700	-3.02118900	3.96192100
C	-0.68021300	-1.87904100	2.18511600
C	0.76632700	-1.49012700	1.89499000
H	1.44988800	-2.18951600	2.38442800
C	2.16126300	-1.39244300	-0.02359900
C	4.44494100	-1.74739000	0.08139100
H	5.30428500	-1.99761500	0.70103700
C	4.61354600	-1.49971200	-1.29969800
H	5.58072400	-1.55838000	-1.78206600
C	3.49327000	-1.12999700	-1.99468300
H	3.51699600	-0.85922800	-3.04427400
C	-1.93565200	-2.61589100	-1.86705900
H	-2.76891100	-2.62577300	-1.17276000
C	-1.86005500	-3.53268100	-2.91425800
H	-2.64869900	-4.26482400	-3.04992400
C	-0.76205500	-3.48218800	-3.77012500
H	-0.67277400	-4.17634300	-4.59980000
C	0.22194500	-2.51574300	-3.54800200
H	1.08343100	-2.44303900	-4.20442800
C	0.07563700	-1.63744300	-2.47962600
C	1.08550900	-0.53913300	-2.18064300
H	1.52815600	-0.17281600	-3.10890600
C	-3.64397900	1.04801500	-1.16865300
H	-3.93424200	0.07487200	-1.55596000
C	-4.49592100	2.14657700	-1.28515700

H	-5.46478400	2.03736000	-1.76086600
C	-4.07377600	3.37188800	-0.77279200
H	-4.71228200	4.24785200	-0.83161000
C	-2.81271400	3.45491800	-0.17809200
H	-2.45887900	4.39441300	0.23600500
C	-2.02376200	2.30752800	-0.11489700
C	-0.62488200	2.34203700	0.48759200
H	-0.54247200	3.16308600	1.20083300
C	0.57497500	1.07268200	2.08997600
C	1.79773200	2.20350000	3.70468200
H	2.08162700	3.16781700	4.12197400
C	2.29610100	1.00804100	4.26301100
H	2.96541000	0.99927000	5.11360300
C	1.91529600	-0.15070900	3.64243200
H	2.27157700	-1.12500100	3.95621500
C	0.87028800	3.96602100	-0.65390600
H	0.42542400	4.66507400	0.04505300
C	1.85109100	4.32819800	-1.53721100
H	2.21771700	5.34538200	-1.58707800
C	2.38476100	3.29324600	-2.33540000
H	3.20010400	3.49650500	-3.02711700
C	0.92625900	1.72229000	-1.46117500
Co	-0.78518500	-0.35118900	-0.16976200
N	-1.60794700	-1.41424700	1.31825800
N	0.95709600	-1.39000700	0.49077500
N	3.28177700	-1.67839000	0.69732300
N	2.27529000	-1.06544400	-1.38868900
N	-0.99251700	-1.68685900	-1.65223800
N	0.41043900	0.50676000	-1.45558900
N	1.95728900	2.04772500	-2.28780900
N	0.40528800	2.68711200	-0.59059300
N	-2.43745300	1.11972800	-0.58991700
N	-0.28671000	1.08076600	1.08381600
N	0.99146200	2.23428000	2.66323500
N	1.06996700	-0.14675400	2.57353100

Table S27. Optimized structure of $[Co(L^{I2})_3]^{3+}$ (Energy= -1958.395341 Hartree, Charge= 3, Multiplicity=1)

Atomic Number	Coordinates (Angstroms) X	Y	Z
C	-1.83544700	1.67489900	1.13992600
N	-1.99615500	2.35677400	0.01263700
C	-3.15671900	3.02618000	-0.10285300
C	-4.12115300	3.01532000	0.90135400
C	-3.82501200	2.27117500	2.04539700
N	-2.67804400	1.58933700	2.16456800
H	-3.30394400	3.58654900	-1.02192900
H	-5.05258400	3.56155800	0.80259600
H	-4.51381200	2.21390600	2.88444200
N	-0.56508800	1.00044100	1.31470700
C	2.98240900	-0.45153400	4.15015100
C	3.64896800	-0.84155600	2.99055500
C	1.71308800	0.12407000	4.03220800
C	3.03109100	-0.65964600	1.74772500
H	4.63911600	-1.28372300	3.03130000
C	1.15838000	0.27701800	2.76203700
H	1.15986100	0.45118400	4.90716500
N	1.81182900	-0.11451000	1.63163400
H	3.43881600	-0.58590900	5.12609900
C	-0.12999700	0.89760400	2.52702100
H	3.53232700	-0.95346600	0.83426300
C	-1.05058500	-2.19720000	1.08127100
N	-2.11910300	-1.41106800	1.10848700
C	-3.17070300	-1.89831800	1.79002600
C	-3.13269400	-3.13855500	2.42552500
C	-1.94095500	-3.86080900	2.32897200
N	-0.88447500	-3.38651400	1.65276500
H	-4.05703800	-1.27139900	1.82264000
H	-3.98530200	-3.52224900	2.97479500
H	-1.81939300	-4.83243100	2.80061700
N	0.06084600	-1.72148300	0.29775300
C	3.68224800	-2.40823300	-2.70529200

C	3.82268800	-1.02201200	-2.72060300
C	2.68324800	-2.97438200	-1.90607200
C	2.97041100	-0.23602500	-1.93628800
H	4.58091400	-0.53883600	-3.32816500
C	1.86510900	-2.13216400	-1.15643800
H	2.53444800	-4.04900100	-1.86774700
N	2.01746100	-0.77817700	-1.16627000
H	4.33275700	-3.03882600	-3.30366400
C	0.77316700	-2.59837100	-0.32556500
H	3.06269100	0.84261500	-1.92658500
C	-1.38063500	-0.37821000	-2.28952000
N	-1.06609800	-1.66216900	-2.43137900
C	-1.95165900	-2.39713700	-3.12657700
C	-3.12055900	-1.85167000	-3.65437600
C	-3.33441200	-0.49135000	-3.42911200
N	-2.45937800	0.25533700	-2.73967900
H	-1.70753900	-3.44711800	-3.26539400
H	-3.82658600	-2.45392300	-4.21522300
H	-4.21777700	0.01844300	-3.80476300
N	-0.40631400	0.45633200	-1.62494300
C	1.95900200	4.56506600	-1.17354100
C	2.64253200	3.96424000	-0.11758900
C	0.96913100	3.83403700	-1.83839200
C	2.32293900	2.65247200	0.24777100
H	3.41940900	4.49323900	0.42467800
C	0.69946100	2.53345200	-1.41897000
H	0.41303300	4.26332300	-2.66586600
N	1.37671900	1.95036400	-0.39219600
H	2.19006200	5.58171100	-1.47672900
C	-0.29062100	1.67307700	-2.04131600
H	2.84112400	2.15912100	1.06069100
H	-0.89815900	2.03340200	-2.86857300
H	0.54988300	-3.65938900	-0.23555000
H	-0.72140700	1.27892400	3.35753800
Co	0.69879200	0.13263000	-0.00769700

Table S28. Optimized structure of **[Co(L^{M})]$^{3+}$** (Energy= -1958.427051 Hartree, Charge= 3, Multiplicity=1)

Atomic Number	Coordinates (Angstroms) X	Y	Z
C	-2.96971800	-1.87641600	-0.83935400
H	-3.53992500	-1.47034100	-0.01190500
C	-3.50176300	-2.86671200	-1.66298200
H	-4.50722900	-3.23241500	-1.48348900
C	-2.72465500	-3.36496200	-2.70810900
H	-3.11576200	-4.12791700	-3.37419800
C	-1.42874900	-2.86971800	-2.88394900
H	-0.79818100	-3.24154500	-3.68543200
C	-0.96398800	-1.88572200	-2.01988700
C	0.43577600	-1.30773700	-2.07361400
H	0.85266900	-1.33682700	-3.08111900
C	1.32657300	0.91135000	-1.89787700
C	3.08501200	1.53379700	-3.25573200
H	3.76244800	1.24787400	-4.05739500
C	3.16835700	2.81191300	-2.66541000
H	3.89803300	3.54698900	-2.98255100
C	2.29361600	3.07259700	-1.64429600
H	2.28756400	4.00768400	-1.09526200
C	-2.96766900	1.66624100	-1.20920000
H	-3.53797100	0.74673900	-1.27242800
C	-3.49895400	2.87493600	-1.65525600
H	-4.50392900	2.90266900	-2.06290300
C	-2.72169700	4.02897500	-1.56295700
H	-3.11225000	4.98748200	-1.89079000
C	-1.42633600	3.93321400	-1.04482600
H	-0.79564900	4.81307400	-0.96524800
C	-0.96230900	2.69270500	-0.62449700
C	0.43672800	2.44969100	-0.09543000
H	0.85347300	3.33650800	0.38370400
C	-2.97031500	0.21497900	2.04361200
H	-3.53956400	0.72974200	1.27830100
C	-3.50346900	-0.00363700	3.31253400

H	-4.50880900	0.33535400	3.53901500
C	-2.72764100	-0.66107200	4.26661900
H	-3.11961600	-0.85686900	5.25997800
C	-1.43184300	-1.06194000	3.92627400
H	-0.80234200	-1.57137900	4.64908100
C	-0.96596400	-0.80518300	2.64258500
C	0.43349400	-1.14238600	2.16934700
H	0.84931700	-2.00084700	2.69834600
C	1.32357900	-2.10113600	0.16004200
C	3.07972400	-3.59094700	0.30043600
H	3.75589200	-4.14357000	0.94908400
C	3.16359700	-3.71856900	-1.10160600
H	3.89258100	-4.36173800	-1.57936900
C	2.29031500	-2.96316400	-1.83819100
H	2.28480800	-2.95491200	-2.92251900
C	2.28866000	-0.11286100	3.48620700
H	2.28186300	-1.05606900	4.02115800
C	3.16289600	0.90177500	3.77275100
H	3.89112100	0.80878900	4.56923700
C	3.08101300	2.05218000	2.96107100
H	3.75808200	2.88943900	3.11568400
C	1.32450000	1.18780800	1.74008500
Co	-0.74549600	0.00067300	-0.00047500
N	-1.73250000	-1.38425100	-1.02445300
N	0.38743900	0.02613300	-1.54562600
N	2.20898500	0.61724300	-2.87295900
N	1.37261700	2.14106000	-1.25379200
N	-1.73107000	1.58011700	-0.68899300
N	0.38709500	1.32541900	0.79562600
N	2.20658200	2.17920900	1.97452400
N	1.36933800	0.01495400	2.48277400
N	-1.73325800	-0.19236600	1.71051400
N	0.38558900	-1.35178400	0.75014400
N	2.20455200	-2.80006100	0.90257500
N	1.37022500	-2.15796900	-1.22697800

Table S29. Optimized structure of $[Ni(L^{12})_3]^{2+}$ (Energy= -1983.063967 Hartree, Charge= 2, Multiplicity=3)

Atomic Number	Coordinates (Angstroms) X	Y	Z
C	-1.80326300	1.36353500	1.76326700
N	-2.08112200	2.14776300	0.72276600
C	-3.27346200	2.76034000	0.75309600
C	-4.16975200	2.59548000	1.80641600
C	-3.76218000	1.75334800	2.84181500
N	-2.57997700	1.12831300	2.82425700
H	-3.50845700	3.39789800	-0.09564500
H	-5.13197900	3.09454500	1.82252700
H	-4.39514600	1.56882000	3.70683700
N	-0.52400500	0.73064500	1.72710200
C	3.12041900	-1.30829700	4.08087100
C	3.74614800	-1.51612100	2.85401500
C	1.87298100	-0.68050900	4.09868700
C	3.10193000	-1.09246900	1.68646500
H	4.71635100	-1.99723500	2.79002000
C	1.29673700	-0.28765700	2.88851900
H	1.35029700	-0.49704000	5.03220900
N	1.90683100	-0.49293500	1.69540500
H	3.59100800	-1.62578000	5.00604900
C	-0.00438400	0.37837700	2.84961700
H	3.56084600	-1.23845400	0.71425500
C	-1.07550300	-2.57592700	0.49275100
N	-1.98415300	-1.76415200	1.03328200
C	-3.01196600	-2.36614300	1.64963900
C	-3.12703400	-3.75204000	1.72752800
C	-2.10973100	-4.49539100	1.12604600
N	-1.07876900	-3.91159100	0.50784400
H	-3.75416000	-1.70933700	2.09608200
H	-3.95868900	-4.22940400	2.23319400
H	-2.11379500	-5.58273300	1.14139800
N	0.00789000	-1.92324300	-0.16568400
C	3.57632800	-2.04586600	-3.36073300

C	3.83912700	-0.71176800	-3.05882400
C	2.54047000	-2.69513100	-2.68509100
C	3.05822900	-0.07185900	-2.08973700
H	4.63257700	-0.16621000	-3.55849300
C	1.80509200	-1.98454100	-1.73422800
H	2.30080200	-3.73369500	-2.89041700
N	2.06699000	-0.68811300	-1.43927000
H	4.16296900	-2.57268000	-4.10674300
C	0.69866500	-2.60549300	-1.01019400
H	3.23519800	0.96623200	-1.82753000
C	-1.50331900	0.44157100	-2.42559700
N	-1.38492000	-0.85671200	-2.70912900
C	-2.34670700	-1.37155200	-3.48903800
C	-3.41118800	-0.61057700	-3.96670900
C	-3.42809900	0.73363600	-3.59476000
N	-2.47593800	1.26763700	-2.82209100
H	-2.25491700	-2.42684800	-3.73562900
H	-4.18487600	-1.03948400	-4.59328400
H	-4.22125500	1.40323800	-3.91916600
N	-0.46355200	0.99680100	-1.62178000
C	2.10417200	4.83189500	-0.32246000
C	2.78109900	4.02376400	0.58788500
C	1.10455700	4.26017500	-1.11288500
C	2.43385300	2.67153400	0.67868500
H	3.56589200	4.42289900	1.22163200
C	0.81652000	2.90270200	-0.96010200
H	0.55333800	4.85321200	-1.83584100
N	1.47736800	2.11921400	-0.07408900
H	2.34755300	5.88535000	-0.41846400
C	-0.21620800	2.25044900	-1.76342400
H	2.93899700	2.01203300	1.37689800
H	-0.77363700	2.85260600	-2.48127800
H	0.46507700	-3.65223500	-1.20496100
H	-0.52777400	0.55863800	3.78916400
Ni	0.68843200	0.12169600	0.00420700

Table S30. Optimized structure of **[Ni (L^M)]$^{2+}$** (Energy= -1983.052103 Hartree, Charge= 2, Multiplicity=3)

Atomic Number	Coordinates (Angstroms) X	Y	Z
C	-3.09925200	1.36698900	1.89205300
H	-3.78107300	1.28957500	1.05041700
C	-3.49879900	1.96015300	3.08832000
H	-4.50726900	2.34516000	3.19454100
C	-2.57786800	2.03861700	4.13137800
H	-2.85356800	2.48420800	5.08210200
C	-1.29203000	1.52969600	3.93665200
H	-0.55637600	1.57006600	4.73409900
C	-0.97082100	0.95634000	2.70842200
C	0.42151900	0.39295600	2.43460300
H	0.87630700	0.06724700	3.37265300
C	1.32421900	-1.54124100	1.40899300
C	3.20445300	-2.55488900	2.30807900
H	3.94685900	-2.55790100	3.10413400
C	3.25161600	-3.52633200	1.28552300
H	4.00673900	-4.30130800	1.25734000
C	2.31514000	-3.40716700	0.29414200
H	2.29322000	-4.06160400	-0.56970400
C	-3.09914800	-2.32260500	0.23767100
H	-3.78096400	-1.55519100	0.59185700
C	-3.49859000	-3.65521200	0.15279100
H	-4.50699300	-3.93991500	0.43311900
C	-2.57765200	-4.59750400	-0.30130100
H	-2.85328800	-5.64363500	-0.39115400
C	-1.29188000	-4.17424000	-0.64463800
H	-0.55620300	-4.88487500	-1.00865500
C	-0.97072500	-2.82393900	-0.52653500
C	0.42167500	-2.30494100	-0.87701200
H	0.87678900	-2.95420400	-1.62812800
C	-3.09896300	0.95556900	-2.13043000
H	-3.78100000	0.26535400	-1.64293500
C	-3.49828200	1.69536300	-3.24210500

H	-4.50671300	1.59510800	-3.62880500
C	-2.57725300	2.55965000	-3.83110200
H	-2.85282000	3.16058700	-4.69213500
C	-1.29150400	2.64520100	-3.29279900
H	-0.55579400	3.31579000	-3.72613700
C	-0.97041300	1.86766500	-2.18248500
C	0.42186200	1.91195900	-1.55758800
H	0.87680000	2.88715500	-1.74434900
C	1.32473700	1.99006500	0.63000300
C	3.20449900	3.27610900	1.05848900
H	3.94693200	3.96700700	0.66310900
C	3.25131000	2.87670900	2.41120400
H	4.00607700	3.24023000	3.09660300
C	2.31482500	1.95859900	2.80374000
H	2.29246600	1.53827600	3.80264800
C	2.31593400	1.44889100	-3.09704900
H	2.29404400	2.52419600	-3.23205700
C	3.25270300	0.64991400	-3.69546400
H	4.00808500	1.06180000	-4.35223200
C	3.20545400	-0.72134100	-3.36536300
H	3.94803900	-1.40923200	-3.76567200
C	1.32473600	-0.44950900	-2.03864800
N	-1.86796700	0.86984200	1.70652000
N	0.34797200	-0.65871200	1.46819000
N	2.29874800	-1.59907000	2.35866000
N	1.35944400	-2.43697700	0.32817800
N	-1.86789200	-1.91320600	-0.10008300
N	0.34812800	-0.94214100	-1.30442600
N	2.29951500	-1.24302300	-2.56313000
N	1.35991500	0.93433100	-2.27419800
N	-1.86770300	1.04314700	-1.60694100
N	0.34808900	1.60069500	-0.16369500
N	2.29924000	2.84143700	0.20524100
N	1.35956200	1.50244900	1.94631000
Ni	-0.95173800	-0.00015800	-0.00008200

Table S31. Optimized structure of $[Cu(L^{I2})_3]^{2+}$ (Energy= -2009.853213 Hartree, Charge= 2, Multiplicity=2)

Atomic Number	Coordinates (Angstroms) X	Y	Z
C	-0.08551700	2.26412600	1.69168600
N	1.01565000	1.62907700	2.08798200
C	1.90105500	2.36550300	2.77423500
C	1.68415100	3.71071600	3.06401900
C	0.48692500	4.26047100	2.60236600
N	-0.40211400	3.54118900	1.90942900
H	2.80602100	1.85538800	3.09584700
H	2.40449800	4.29949100	3.62042300
H	0.23038500	5.30113000	2.78673700
N	-1.01364500	1.45462700	0.96869600
C	-5.44199300	0.97936700	-0.87398800
C	-5.12165600	-0.31678200	-1.26827800
C	-4.47933300	1.72683600	-0.19143800
C	-3.84795800	-0.81893300	-0.96476900
H	-5.83759500	-0.93751100	-1.79698600
C	-3.23162200	1.14568100	0.06073600
H	-4.68738600	2.74031300	0.13808900
N	-2.91907700	-0.11428800	-0.31783300
H	-6.41869200	1.40164600	-1.08820300
C	-2.20536700	1.90498900	0.77376300
H	-3.57239500	-1.83035800	-1.25283600
C	0.24960900	1.58089400	-2.26599600
N	0.53764700	2.47288400	-1.32071400
C	0.81900600	3.71104200	-1.75376200
C	0.80598900	4.04986300	-3.10442900
C	0.48212500	3.02785400	-3.99874900
N	0.20082300	1.78923000	-3.58343900
H	1.05503000	4.44585600	-0.98824400
H	1.02932800	5.05481900	-3.44401700
H	0.44116400	3.20087600	-5.07146500
N	-0.03536800	0.25961700	-1.80175500
C	-0.48424100	-4.42842000	-2.67079400

C	-0.78634100	-4.60101400	-1.32238200
C	-0.20860700	-3.14086700	-3.13824300
C	-0.79993600	-3.48086600	-0.48191000
H	-1.00781900	-5.58198200	-0.91547400
C	-0.24547100	-2.07646400	-2.23596900
H	0.03047700	-2.96095900	-4.18155600
N	-0.53699100	-2.25022900	-0.92480200
H	-0.46270500	-5.27643000	-3.34795800
C	0.02718000	-0.70710200	-2.65239300
H	-1.02747300	-3.57838000	0.57448000
C	3.07447100	-0.93303100	-0.00166500
N	3.00680200	-0.90089500	-1.33750100
C	4.17972700	-0.77710700	-1.97538500
C	5.39350000	-0.66946100	-1.29984300
C	5.33355400	-0.70151800	0.09381400
N	4.17627900	-0.83386400	0.74967200
H	4.13941300	-0.76100300	-3.06219700
H	6.33396500	-0.56274300	-1.82837200
H	6.23038400	-0.61789000	0.70340500
N	1.82836000	-1.08018900	0.66074200
C	-0.64284400	-2.39345800	4.56161300
C	-1.81594500	-1.96962000	3.94103100
C	0.56575000	-2.26069300	3.87570700
C	-1.73737200	-1.42280800	2.65782500
H	-2.77910900	-2.05416600	4.43272400
C	0.57010700	-1.70477000	2.59402800
H	1.50102700	-2.58197200	4.32318100
N	-0.57822700	-1.29275800	2.00088100
H	-0.66530100	-2.82065200	5.55930700
C	1.82561200	-1.56134600	1.84843400
H	-2.62376500	-1.07927000	2.13538500
H	2.74755100	-1.87162800	2.34531200
H	0.27668000	-0.50598800	-3.69346200
H	-2.46624300	2.89504600	1.15053700
Cu	-0.42157700	-0.39561800	0.14242300

Table S32. Optimized structure of **[Cu(L^M)]$^{2+}$** (Energy= -2009.834457 Hartree, Charge= 2, Multiplicity=2)

Atomic Number	Coordinates (Angstroms) X	Y	Z
C	-3.82778900	0.81056500	-1.26367400
H	-4.06187800	-0.17328600	-1.66163200
C	-4.73473500	1.86288300	-1.38581000
H	-5.68896100	1.70706900	-1.87774600
C	-4.38552800	3.10343400	-0.85558600
H	-5.06804600	3.94544500	-0.91665000
C	-3.14201800	3.24540100	-0.23673300
H	-2.84845400	4.19720900	0.19597500
C	-2.29237800	2.14112100	-0.16955000
C	-0.91534600	2.26475300	0.47753300
H	-0.92719100	3.08631600	1.19513400
C	0.35883900	1.11227100	2.10166900
C	1.44066900	2.36160900	3.73295800
H	1.63483200	3.35194600	4.14073800
C	2.01351500	1.21852200	4.32515200
H	2.65107700	1.27328900	5.19816000
C	1.76104900	0.02771000	3.69921100
H	2.19887800	-0.90657200	4.03037500
C	-2.75481300	-2.19040200	1.49882500
H	-3.51025400	-1.89734100	0.77608200
C	-3.07175100	-3.00556000	2.58342000
H	-4.08535800	-3.36957000	2.71206100
C	-2.05983800	-3.33719200	3.48243700
H	-2.26381700	-3.97805300	4.33460500
C	-0.77501900	-2.83269300	3.27057400
H	0.03293500	-3.07750700	3.95313800
C	-0.53594100	-2.02011300	2.16440900
C	0.84892200	-1.42496700	1.90622200
H	1.59993000	-2.02644000	2.42704900
C	-1.51180700	-2.87210300	-2.13060500
H	-2.41216800	-2.99681100	-1.53740500
C	-1.19017200	-3.76695100	-3.14873300

H	-1.84837000	-4.60077200	-3.36746500
C	-0.01526800	-3.55960200	-3.86923000
H	0.26672300	-4.23133900	-4.67399600
C	0.79205900	-2.46666700	-3.54763200
H	1.70524200	-2.27400500	-4.10213500
C	0.40296700	-1.61730500	-2.51498900
C	1.23068600	-0.39014400	-2.13932100
H	1.67407000	0.03080100	-3.04466900
C	0.79352200	1.82454400	-1.40687600
C	2.15289100	3.54657900	-2.15377900
H	2.99469000	3.83885200	-2.77879500
C	1.45998100	4.51058400	-1.39213400
H	1.72301200	5.56044500	-1.40776900
C	0.46508300	4.03646600	-0.58017400
H	-0.08989200	4.67697100	0.09550600
C	3.68240700	-0.64426900	-1.83019800
H	3.72103100	-0.36529000	-2.87712000
C	4.80755200	-0.84783500	-1.07724900
H	5.79637900	-0.76159100	-1.50891700
C	4.60602300	-1.11567500	0.29563300
H	5.46005600	-1.23084800	0.96099500
C	2.30128300	-1.11368700	0.06977100
N	-2.63557400	0.94068100	-0.66503900
N	-0.49507300	1.04008100	1.09061000
N	0.66803200	2.31124000	2.66669100
N	0.95522400	-0.05334300	2.60304200
N	-1.51673700	-1.71897200	1.28883200
N	1.08626700	-1.30009500	0.51434900
N	3.41559000	-1.22267900	0.84994000
N	2.43807600	-0.76337800	-1.28910600
N	-0.73555000	-1.82331200	-1.82343700
N	0.39416500	0.56782100	-1.46816000
N	1.84827100	2.26430600	-2.14722500
N	0.12596300	2.71712100	-0.55884600
Cu	-0.85949400	-0.47113400	-0.24587300

Table S33. Optimized structure of $[Zn(L^{I2})_3]^{2+}$ (Energy= -1879.350921 Hartree, Charge= 2, Multiplicity=1)

Atomic Number	Coordinates (Angstroms) X	Y	Z
C	1.52972400	-2.54742400	-0.11763700
N	1.98554200	-1.88923500	-1.18439700
C	3.08689900	-2.39518000	-1.75843900
C	3.72348700	-3.53530000	-1.27437000
C	3.15578400	-4.13220300	-0.14677300
N	2.05954800	-3.64035300	0.43710500
H	3.46169900	-1.86504600	-2.63085800
H	4.61284300	-3.93798400	-1.74564500
H	3.58714700	-5.02368800	0.30265100
N	0.35157900	-1.99739800	0.46447500
C	-3.14849200	-2.28664300	3.73954600
C	-3.62519100	-1.05444000	3.29968000
C	-2.02411200	-2.83074600	3.11468200
C	-2.95996000	-0.40989700	2.24990800
H	-4.49477000	-0.59142600	3.75398100
C	-1.41799200	-2.11978700	2.07541100
H	-1.61799100	-3.78814200	3.42603300
N	-1.88333600	-0.92256300	1.64662800
H	-3.63820200	-2.81541400	4.55123700
C	-0.22895400	-2.65199400	1.40727900
H	-3.30591100	0.55066600	1.88104000
C	1.54107000	1.16610400	2.26201700
N	1.98207600	-0.09246000	2.24016900
C	3.08382800	-0.33974800	2.96367900
C	3.73533400	0.65185300	3.69287000
C	3.18240200	1.93264900	3.63129600
N	2.08607000	2.19568300	2.91468300
H	3.44678900	-1.36483000	2.95267000
H	4.62493500	0.44248300	4.27595000
H	3.62612400	2.76978900	4.16526000
N	0.36181700	1.39907600	1.49732100
C	-3.12932100	4.39417300	0.11823300

C	-3.61244700	3.39879100	-0.72726400
C	-2.00382900	4.12079200	0.89876400
C	-2.95232900	2.16470100	-0.76202800
H	-4.48315000	3.56404300	-1.35296900
C	-1.40309800	2.86278100	0.80143000
H	-1.59274900	4.86770900	1.57077800
N	-1.87466900	1.89445200	-0.01931700
H	-3.61494300	5.36350800	0.17133100
C	-0.21330200	2.54554000	1.59339600
H	-3.30335700	1.36622800	-1.40828000
C	1.53883300	1.36895100	-2.14564600
N	1.98683900	1.97428400	-1.04485900
C	3.09082800	2.72072900	-1.19475600
C	3.73771400	2.85751700	-2.42043700
C	3.17756100	2.16915000	-3.49859000
N	2.07881300	1.42097000	-3.36577800
H	3.45927200	3.21967500	-0.30147900
H	4.62920100	3.46404000	-2.53262800
H	3.61725200	2.21433900	-4.49229000
N	0.35737400	0.59437300	-1.96202600
C	-3.14995100	-2.08385500	-3.85583200
C	-3.63011800	-2.31648500	-2.56960700
C	-2.02175800	-1.27559300	-4.01274200
C	-2.96448400	-1.73206000	-1.48547600
H	-4.50270200	-2.93763100	-2.39722200
C	-1.41546300	-0.73327700	-2.87641000
H	-1.61284700	-1.06865200	-4.99683800
N	-1.88416900	-0.95802500	-1.62600600
H	-3.63991400	-2.52061000	-4.72037700
C	-0.22261300	0.10640200	-3.00121100
H	-3.31319000	-1.89094200	-0.46982000
H	0.17739400	0.29437200	-3.99830600
H	0.18881900	3.31317700	2.25563000
H	0.16787200	-3.61151900	1.74098000
Zn	-0.54974400	0.00137500	0.00024500

Table S34. Optimized structure of **[Zn(L^M)]$^{2+}$** (Energy= -1879.326224 Hartree, Charge= 2, Multiplicity=1)

Atomic Number	Coordinates (Angstroms) X	Y	Z
C	3.13504300	2.51293100	0.24031200
H	3.87981500	1.77682100	0.52900700
C	3.44689900	3.86975700	0.21350500
H	4.44446400	4.20479900	0.47606100
C	2.45190300	4.77055500	-0.16241900
H	2.65614900	5.83583800	-0.20672500
C	1.18699400	4.28032500	-0.49017900
H	0.39749800	4.95877200	-0.79844200
C	0.95241900	2.90730700	-0.43322100
C	-0.42532600	2.34260100	-0.78755400
H	-0.90140900	3.01303000	-1.50669500
C	-1.32006400	0.53799000	-2.00664800
C	-3.23862200	0.85640800	-3.27240300
H	-3.99880300	1.55550800	-3.61672500
C	-3.27866500	-0.49531700	-3.67495300
H	-4.04402700	-0.88098200	-4.33606100
C	-2.32485200	-1.31447400	-3.13390200
H	-2.30191600	-2.38213100	-3.31923300
C	3.14302900	-1.05087400	-2.28387900
H	3.88629600	-0.43329700	-1.78785100
C	3.45887500	-1.75266700	-3.44434300
H	4.45809500	-1.69346900	-3.86193100
C	2.46561400	-2.52783700	-4.04041700
H	2.67293400	-3.09890600	-4.94008700
C	1.19839700	-2.56570000	-3.45692200
H	0.41013100	-3.17123300	-3.89348800
C	0.95984300	-1.82976700	-2.29720600
C	-0.42055900	-1.85312600	-1.63646900
H	-0.89666000	-2.81067400	-1.85950000
C	3.13368400	-1.45575900	2.06378200
H	3.88018100	-1.33523500	1.28397800
C	3.44415800	-2.11083400	3.25265600

H	4.44236300	-2.50313700	3.41393400
C	2.44692200	-2.23903600	4.21798200
H	2.64998400	-2.73340400	5.16290100
C	1.18126000	-1.71341400	3.95425000
H	0.38995300	-1.78841200	4.69372400
C	0.94821800	-1.07607500	2.73648700
C	-0.43033000	-0.49049300	2.42130200
H	-0.90949300	-0.20443500	3.36038300
C	-1.32829900	1.46493900	1.46576400
C	-3.25135200	2.39694600	2.36998200
H	-4.01313400	2.34367600	3.14588400
C	-3.29177100	3.42134200	1.40054800
H	-4.05907100	4.18475500	1.39537400
C	-2.33551700	3.36488000	0.42283500
H	-2.31239000	4.05938000	-0.40896900
C	-2.33757100	-2.05425500	2.69627000
H	-2.31825600	-1.68080700	3.71358400
C	-3.29042100	-2.93163500	2.25378400
H	-4.05871700	-3.31054300	2.91539400
C	-3.24516900	-3.25930900	0.88214900
H	-4.00398400	-3.90677200	0.44607200
C	-1.32426200	-2.00548100	0.53227100
N	1.91974100	2.03878500	-0.07917400
N	-0.32799400	1.00330000	-1.27635900
N	-2.31971100	1.34436300	-2.46473200
N	-1.35454200	-0.83497200	-2.30634800
N	1.92547500	-1.08961600	-1.71826100
N	-0.32865500	-1.60682800	-0.23184800
N	-2.32258100	-2.80483600	0.05927400
N	-1.36361000	-1.57834800	1.87093100
N	1.91766000	-0.94518000	1.80996000
N	-0.33374100	0.60285300	1.50623300
N	-2.33004800	1.45588400	2.39084500
N	-1.36277800	2.41091300	0.42654100
Zn	1.11175400	0.00090200	0.00232700

Table S35. Optimized structure of **[Al (L^{I2})$_3$]$^{3+}$** (Energy= -1815.409266 Hartree, Charge= 3, Multiplicity=1)

Center Number	Coordinates (Angstroms) X	Y	Z
C	-1.66476700	1.67332600	1.53622100
N	-1.99720000	2.19134400	0.36080900
C	-3.14180600	2.89751100	0.34108100
C	-3.92008200	3.07861800	1.48162100
C	-3.45619600	2.48427400	2.65844300
N	-2.32341900	1.77078800	2.68631800
H	-3.42948300	3.32441100	-0.61603400
H	-4.83966100	3.65271000	1.45953400
H	-3.99798900	2.57466100	3.59653400
N	-0.41564400	0.94828000	1.56319800
C	3.23874100	-0.87925000	4.04794400
C	3.79399200	-1.27110300	2.83156700
C	2.01313600	-0.20449600	4.03805700
C	3.11000400	-0.98628000	1.64529600
H	4.74512300	-1.79142200	2.78896100
C	1.39096400	0.04252000	2.81533100
H	1.54578500	0.12506000	4.96084800
N	1.92979400	-0.34498000	1.62524100
H	3.74588400	-1.08855800	4.98490400
C	0.12557500	0.75447700	2.72109400
H	3.52467400	-1.27792000	0.68781300
C	-1.17939100	-2.36017100	0.88323100
N	-2.01419800	-1.43035500	1.33265900
C	-3.07963000	-1.88982100	2.01269600
C	-3.28849700	-3.24946600	2.23155300
C	-2.33219700	-4.12532600	1.70869300
N	-1.26917500	-3.67795300	1.02994500
H	-3.77156000	-1.14149700	2.38899300
H	-4.14804000	-3.61208800	2.78422300
H	-2.41210600	-5.20190900	1.83545400
N	-0.05342200	-1.86385800	0.13494600
C	3.47317500	-2.57010600	-3.01074900

C	3.73457500	-1.20351800	-2.93415600
C	2.44542000	-3.10238200	-2.22399300
C	2.96676900	-0.40905500	-2.07616600
H	4.52074400	-0.74701500	-3.52638100
C	1.71972400	-2.24666700	-1.39726100
H	2.20643900	-4.16092500	-2.25351500
N	1.97890700	-0.91120300	-1.31835200
H	4.05317500	-3.21140600	-3.66744800
C	0.61692400	-2.71360200	-0.57193200
H	3.15307000	0.65558800	-1.99427500
C	-1.51502900	0.12318100	-2.39184900
N	-1.58383200	-1.19979700	-2.29553700
C	-2.57438400	-1.77865700	-2.99804600
C	-3.46720900	-1.04066700	-3.77098400
C	-3.28934400	0.34518100	-3.78273400
N	-2.30897500	0.93304800	-3.08627200
H	-2.64348100	-2.86165600	-2.93655000
H	-4.26133900	-1.51666500	-4.33530000
H	-3.94105000	1.00031300	-4.35510700
N	-0.42798200	0.74807600	-1.67654000
C	2.41683700	4.53181400	-0.99661100
C	3.02399800	3.81745200	0.03432800
C	1.37466600	3.92777400	-1.70894400
C	2.57391700	2.52613200	0.32672100
H	3.83722500	4.24448100	0.61178900
C	0.97818000	2.63927700	-1.35691800
H	0.87570400	4.44629600	-2.52172300
N	1.57213300	1.94050300	-0.34954100
H	2.74507600	5.53638200	-1.24565700
C	-0.09374100	1.93984200	-2.04862000
H	3.03088200	1.94796500	1.12128000
H	-0.61934100	2.42380900	-2.86959400
H	0.34361900	-3.76723200	-0.56302600
H	-0.36744500	1.11653900	3.62174100
Al	0.65609400	0.08445900	-0.02115900

Table S36. Optimized structure of **[Al(L^M)]$^{3+}$** (Energy= -1815.428231 Hartree, Charge= 3, Multiplicity=1)

Atomic Number	Coordinates (Angstroms) X	Y	Z
C	3.09188700	-1.45591600	-1.61391100
H	3.72525900	-0.60465500	-1.38789200
C	3.55892800	-2.50698300	-2.39617600
H	4.57477900	-2.48938700	-2.77600200
C	2.69926800	-3.57044600	-2.67261000
H	3.03506200	-4.41229600	-3.27047800
C	1.39561400	-3.53837200	-2.16788800
H	0.70701000	-4.35333800	-2.36738300
C	0.99885500	-2.45485300	-1.39450600
C	-0.40692600	-2.32365100	-0.81795500
H	-0.84649600	-3.30580300	-0.64025400
C	-1.29473700	-1.66824700	1.30105600
C	-3.09050700	-2.81892100	2.18713000
H	-3.78708100	-3.64676500	2.07333700
C	-3.16211200	-1.96798000	3.30832700
H	-3.89897800	-2.10438000	4.09046600
C	-2.26925700	-0.92965700	3.34583100
H	-2.26117900	-0.18985400	4.13852100
C	3.08713800	-0.67497000	2.07345100
H	3.72195400	-0.90531200	1.22443000
C	3.55128000	-0.82831200	3.37571200
H	4.56627900	-1.16762500	3.55227600
C	2.68984800	-0.53541200	4.43332100
H	3.02334700	-0.63344300	5.46195100
C	1.38743100	-0.11215400	4.15074300
H	0.69748900	0.12312800	4.95496100
C	0.99364100	0.01730500	2.82497000
C	-0.41062600	0.45368100	2.42047000
H	-0.85040400	1.09947200	3.18135900
C	3.09167600	2.12780700	-0.44668900
H	3.72402500	1.50707600	0.17919900
C	3.55906400	3.33105300	-0.96491500

H	4.57417600	3.65198900	-0.75735700
C	2.70072200	4.10140000	-1.74991300
H	3.03684500	5.04027900	-2.17937600
C	1.39795400	3.64723700	-1.97755100
H	0.71030200	4.22686300	-2.58526100
C	1.00076700	2.43555700	-1.42655600
C	-0.40433100	1.86977700	-1.60410700
H	-0.84222400	2.20651400	-2.54445500
C	-1.28953900	-0.29383300	-2.09789500
C	-3.08118200	-0.48748000	-3.54239700
H	-3.77651200	0.02440200	-4.20419800
C	-3.15160300	-1.88408100	-3.36677200
H	-3.88632700	-2.49393900	-3.87821500
C	-2.26060200	-2.43495400	-2.48396600
H	-2.25206700	-3.49139700	-2.23985200
C	-2.25976400	3.36808400	-0.87151300
H	-2.24920700	3.68463700	-1.90854000
C	-3.15257500	3.85739200	0.04508700
H	-3.88675500	4.60519000	-0.22895500
C	-3.08483100	3.31133700	1.34260700
H	-3.78185700	3.62852400	2.11529000
C	-1.29189000	1.96367100	0.79197500
N	1.84199000	-1.43190900	-1.10773500
N	-0.34306700	-1.52384000	0.37164000
N	-2.20105600	-2.66143100	1.21877500
N	-1.33689500	-0.76392500	2.36036300
N	1.83841900	-0.24666300	1.79724600
N	-0.34308100	1.08404000	1.13320400
N	-2.19863400	2.39132600	1.69172100
N	-1.33079000	2.42892500	-0.52095300
N	1.84264000	1.67652000	-0.68182500
N	-0.33985300	0.43969800	-1.50590200
N	-2.19428500	0.27321400	-2.91918500
N	-1.33092300	-1.66352600	-1.84484000
Al	0.87152000	-0.00041100	0.00128600

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