Supporting Information

Stabilization of different redox levels of a tridentate benzoxazole amidophenoxide ligand when bound to Co(III) or V(V).

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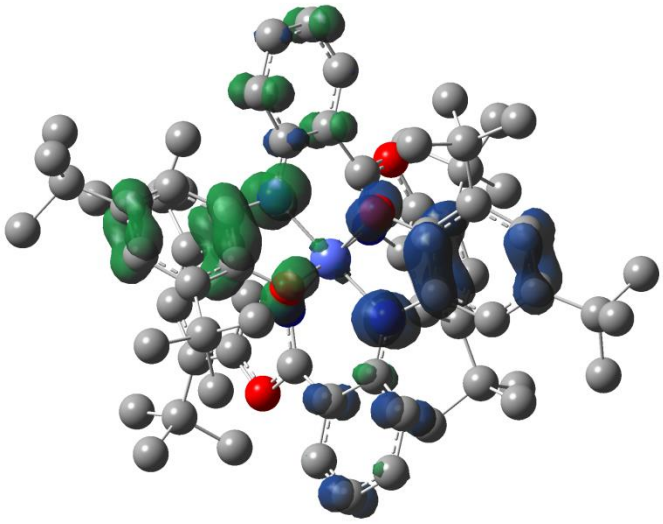

Figure S1. DFT-computed spin density for the lowest energy broken symmetry solution for $[Co^{III}(\mathbf{NNO^{ISQ}})_2]^+$. DFT calculations predict that diamagnetic $[Co^{III}(\mathbf{NNO^{AP}})_2]^-$ forms upon one-electron reduction.

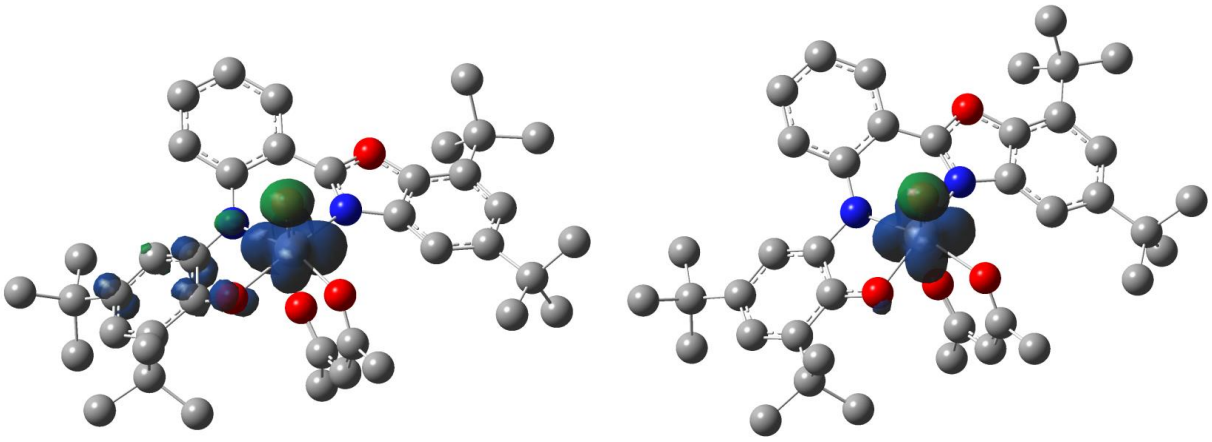

Figure S2. DFT-computed spin density for the lowest energy solution for oxidized $[VO(\mathbf{NNO})(acac)]^{+}$ (left), predicting the formation of V^{IV} and an **NNO** ligand in the iminioquinone oxidation level. The DFT-computed spin density for the reduced complex $[VO(\mathbf{NNO})(acac)]^{-}$ (right) predicts metal reduction to V^{IV}.

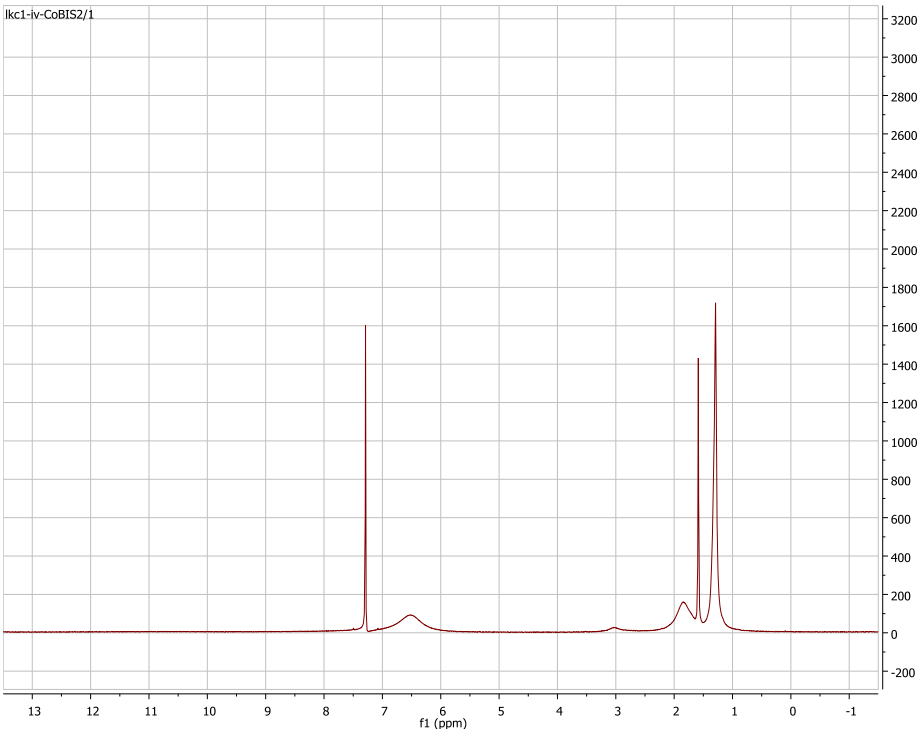

Figure S3. ^{1}H NMR of $Co(\mathbf{NNO})_2$ in $CDCl_3$. Note solvent peaks at 7.26 ($CDCl_3$) and 1.54 (H_2O) ppm.

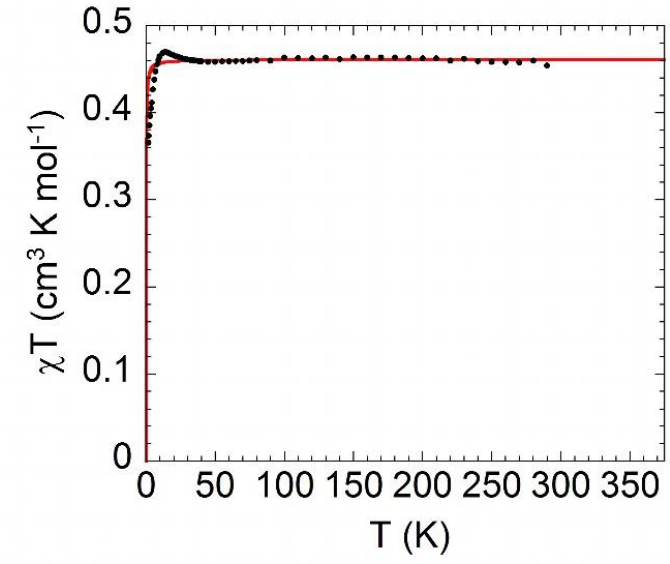

Figure S4. $\chi_M T$ *vs T* data for Co(**NNO**)$_2$ at 10 000 Oe between 1.8 and 300 K. The solid line represents the fit to the data. Above 30 K, the magnetic data was fitted to the Curie-Weiss law, which indicated the absence of intermolecular magnetic interactions in the system.

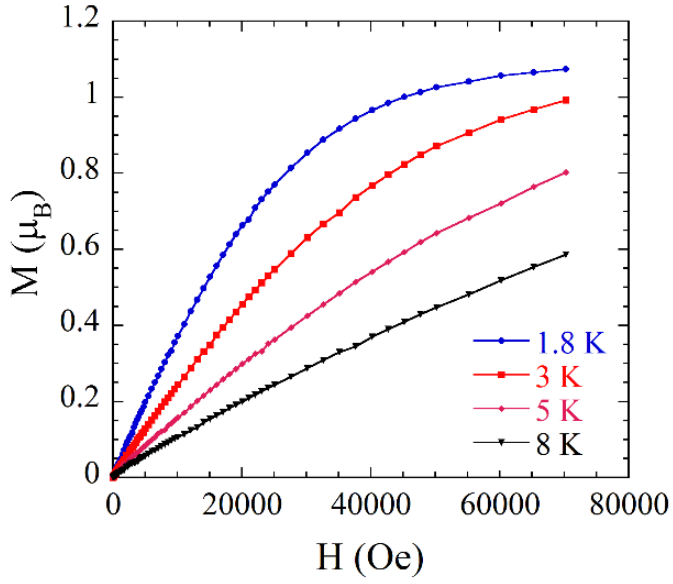

Figure S5. Field dependence of the magnetization for Co(**NNO**)$_2$ at 1.8, 3, 5 and 8 K between 0 and 7 T. The saturated value of 1.07 μ_β at 7 T observed in field dependence of the magnetization data (left) further suggested the presence of a single unpaired electron in complex Co(**NNO**)$_2$.

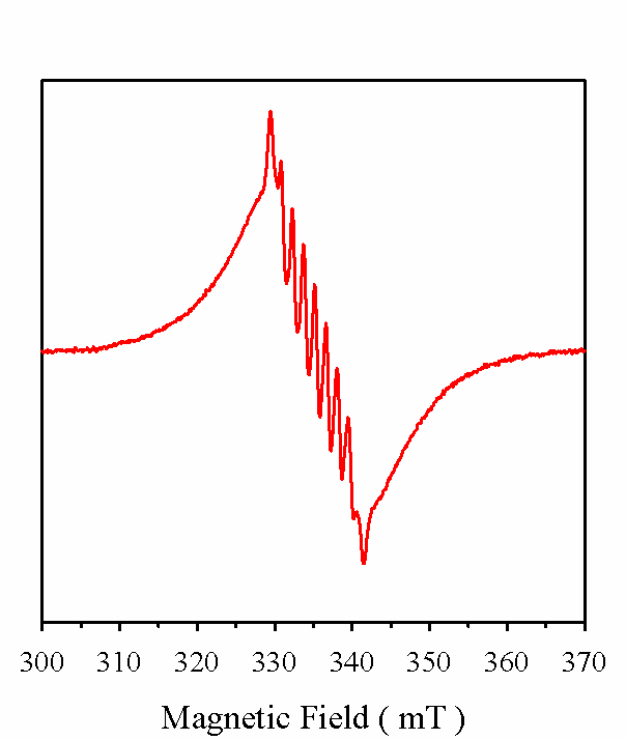

Figure S6. Solution EPR of Co(**NNO**)$_2$ in toluene at 298 K. Conditions: 1.0 mM; Frequency: 9.38 GHz; Power: 2.0 mW; modulation frequency: 100 kHz; amplitude 0.2 mT.

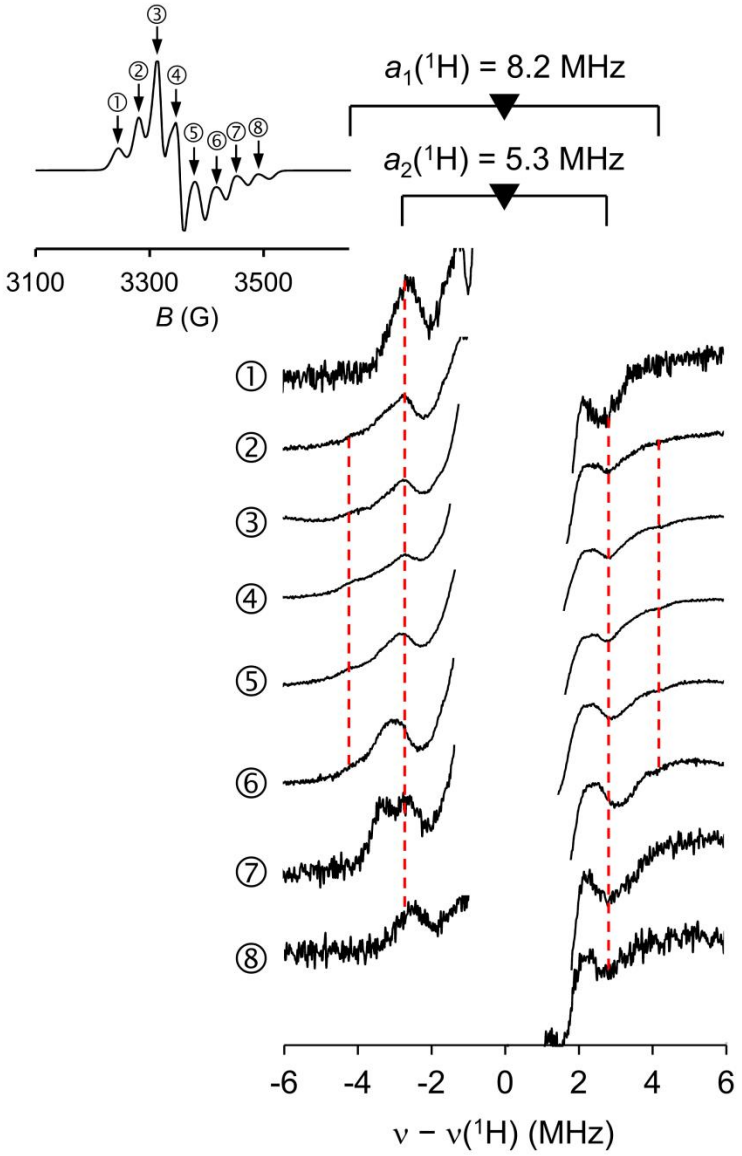

Figure S7. Field dependence of ^{1}H ENDOR of Co(**NNO**)$_2$, scaled to show most strongly coupled protons. Dashed lines show isotropic hyperfine interaction components. Field positions indicated on EPR spectrum (inset). ENDOR measurement settings: microwave modulation frequency = 25 kHz, microwave modulation amplitude = 0 G, microwave power = 20 mW, frequency = 9.468 GHz, time constant = 1.28 ms, RF modulation depth = 250 kHz, RF attenuation = 8 db, average of 15 five-second scans, temperature = 20 K.

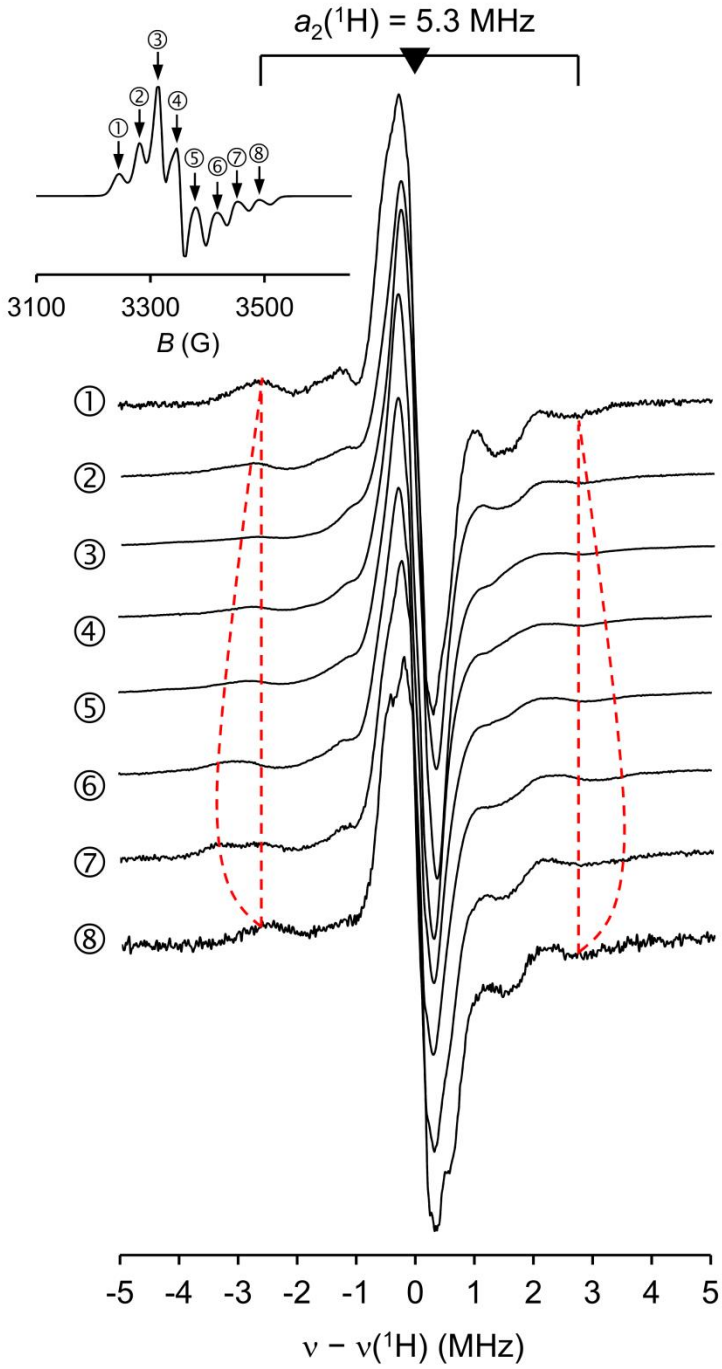

Figure S8. Field dependence of ^{1}H ENDOR of Co(**NNO**)$_2$, showing more weakly coupled protons. Red dashed lines indicate field dependence of second strongest proton coupling, showing a predominantly isotropic interaction an additional splitting from a dipolar contribution. Field positions indicated on EPR spectrum (inset). ENDOR measurement settings: microwave modulation frequency = 25 kHz, microwave modulation amplitude = 0 G, microwave power = 20 mW, frequency = 9.468 GHz, time constant = 1.28 ms, RF modulation depth = 250 kHz, RF attenuation = 8 db, average of 15 five-second scans, temperature = 20 K.

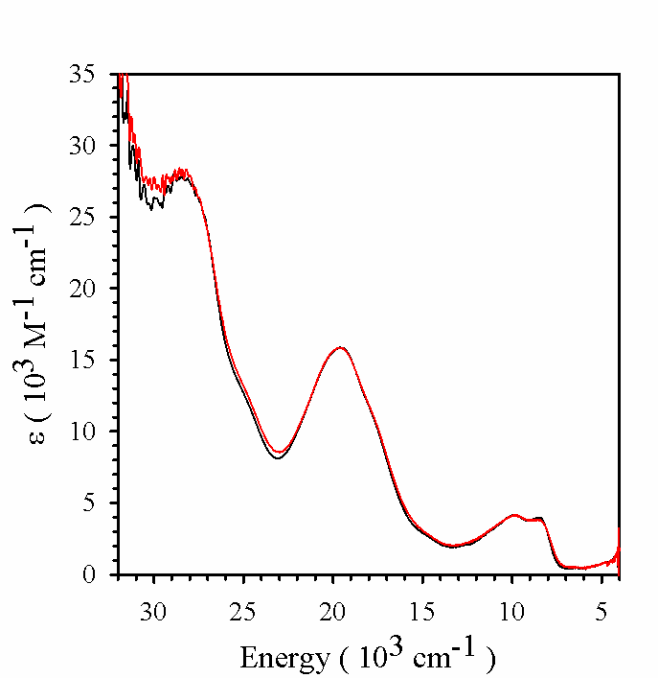

Figure S9. Variable temperature (black: 298 K; red: 353 K) UV-Vis-NIR spectrum of Co(**NNO**)$_2$ in toluene.

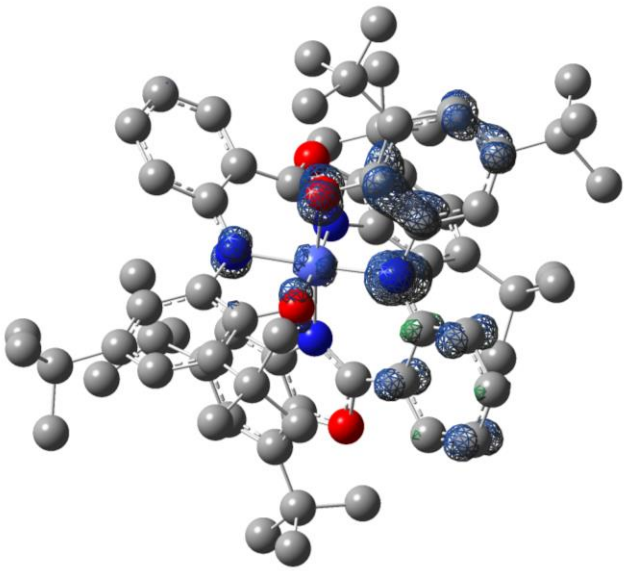

Figure S10. Spin density plot of Co(**NNO**)$_2$ using a PCM solvent model for CH_2Cl_2. The single-point calculation was completed on the S = ½ geometry discussed in the text, and results in localization of the ligand radical on one of the **NNO** ligands. The energy difference between the symmetric structure (**Figure 6**), and that discussed here is very small (Δ = 0.16 kcal / mol).

Table S1. Crystallographic data for Co(**NNO**)$_2$ and VO(**NNO**)(acac).

	Co(**NNO**)$_2$	VO(**NNO**)(acac)
empirical formula	$C_{70}H_{88}N_4O_4Co$	$C_{40}H_{51}N_2O_5V$
formula weight	1108.40	690.78
space group	Pbca	Pbcn
crystal system	orthorhombic	orthorhombic
a (Å)	21.8910(16)	23.0329(19)
b (Å)	24.2657(18)	14.5586(11)
c (Å)	24.9070(19)	5.2546(18)
α (deg)	90	90
β (deg)	90	90
γ (deg)	90	90
V (Å^3)	13230.6	8468.54
Z, Z'	8, 0	12, 0
T (K)	RT (283-303)	RT (283-303)
ρ_{calcd} (g cm^{-3})	1.117	1.1335
λ (Å)	0.71073	1.54178
μ (cm^{-1})	0.0307	0.2322
Final R indexes [I > 2.0σ(I)]	0.0489	0.0427
wR_2	0.0427	0.1238
R_1	0.0695	0.0560
Goodness-of-fit on F^2	1.068	1.030

Computed Metrical Parameters:

1. Co(**NNO**)$_2$ (S = ½)

Co	0.03258000	-0.89039700	-0.00369300
O	1.03358600	-2.18908800	0.93281700
O	-1.45129800	1.66872000	-2.83343600
O	-0.92176400	-2.20421100	-0.96643300
O	1.42525400	1.65803800	2.88209400
N	1.42462700	-1.01810100	-1.32907800
N	-0.90128400	0.50527200	-1.01205700
N	-1.34695600	-1.09615500	1.32068200
N	0.91205800	0.51838000	1.03379000
C	2.24784400	-2.32634400	0.44419700
C	3.25424100	-3.13413500	1.05917700
C	4.52331700	-3.09219900	0.49180500
C	4.86706900	-2.32471200	-0.64937800
C	3.85470900	-1.61032900	-1.27806700
C	2.54080800	-1.64282100	-0.77524500
C	1.20293100	-0.92560100	-2.67367700
C	1.94696200	-1.66367400	-3.63947300
C	1.72503900	-1.52488900	-4.99548200
C	0.74779700	-0.64047500	-5.48541300
C	-0.02710900	0.05836500	-4.57988200
C	0.16020900	-0.08003700	-3.18695400
C	-0.71185900	0.65105900	-2.30808200
C	-2.14713300	2.22091000	-1.77478100
C	-3.03255300	3.30112800	-1.80193600
C	-3.54581300	3.62447400	-0.54050400
C	-3.21960100	2.94566300	0.65854900
C	-2.33245900	1.86850900	0.61431600
C	-1.80068700	1.50950100	-0.62707500
C	2.93865200	-3.98724800	2.30152000
C	6.32556200	-2.31382500	-1.14044700
C	-3.38971900	4.04784500	-3.09558600
C	-3.86041600	3.42047200	1.97847300
C	2.54465100	-3.07197500	3.48495600
C	1.77353500	-4.95583000	1.98284000
C	4.14542900	-4.83706700	2.74600800
C	7.24670300	-1.77038500	-0.02157200
C	6.76265100	-3.75305000	-1.50594600
C	6.51794200	-1.42847700	-2.38617700
C	-2.10639900	4.65427300	-3.71345600
C	-4.03362400	3.06047100	-4.09886900
C	-4.38780100	5.19268400	-2.84127000
C	-3.48099000	4.89829400	2.23744100
C	-5.39956500	3.29489500	1.87965300
C	-3.39091800	2.59100700	3.18855000
C	-2.13371000	-2.38756200	-0.48144200
C	-3.11816800	-3.20548300	-1.11602700
C	-4.38884800	-3.21135000	-0.54821600

C	-4.75253000	-2.48206100	0.61052700
C	-3.75788800	-1.75751400	1.25696400
C	-2.44510900	-1.74328400	0.75294900
C	-1.11841400	-1.03765500	2.66489200
C	-1.83211400	-1.82782400	3.61269200
C	-1.60398800	-1.72321800	4.97041700
C	-0.65134600	-0.82319800	5.48148100
C	0.09262000	-0.07120100	4.59326100
C	-0.10083400	-0.17314000	3.19789600
C	0.73026400	0.61998400	2.33533500
C	2.07701100	2.27718400	1.83140700
C	2.89747600	3.40236400	1.87698100
C	3.37289700	3.79684700	0.61541400
C	3.06653500	3.13732700	-0.59394200
C	2.24344600	2.00421300	-0.56610000
C	1.75211900	1.57898000	0.66596900
C	-2.78060400	-4.01854800	-2.37937400
C	-6.21056400	-2.51993800	1.10191500
C	3.23194200	4.13179300	3.18689700
C	3.60494400	3.63488100	-1.95222900
C	-2.41125500	-3.06334100	-3.53903300
C	-1.58976200	-4.96310000	-2.08479200
C	-3.96417900	-4.88885500	-2.84582900
C	-7.14791400	-1.98361400	-0.00680700
C	-6.60711700	-3.97662600	1.44349100
C	-6.42655400	-1.66077500	2.36209700
C	4.15727500	5.34036100	2.95277900
C	1.92596900	4.64383000	3.84183300
C	3.94842900	3.15532600	4.15091400
C	4.49796300	4.88319600	-1.81707000
C	4.44348000	2.51799200	-2.61770800
C	2.41483800	3.99586900	-2.87365000
H	5.30972500	-3.67142700	0.95996900
H	4.06116300	-1.00756300	-2.15323000
H	2.67940300	-2.37917500	-3.28959400
H	2.30699100	-2.12650100	-5.68921800
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H	-2.06270700	1.31037900	1.49747100
H	3.36616400	-2.39005200	3.73767300
H	2.32441300	-3.67749800	4.37348400
H	1.66128200	-2.47765900	3.24814200
H	1.52959800	-5.55505700	2.86937300
H	0.87898200	-4.41088900	1.67559900
H	2.05233100	-5.64503100	1.17657100
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H	6.96898500	-0.74492700	0.24802600
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H	8.29251400	-1.76459900	-0.35409800
H	7.80431600	-3.76133000	-1.85100800
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H	6.69065100	-4.43218500	-0.65018000
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H	7.56870600	-1.45002900	-2.69740000
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H	-4.95450200	2.63136600	-3.68798400
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H	-3.98074800	5.94701700	-2.15836600
H	-2.39431500	5.01416400	2.32091000
H	-3.93277500	5.24784200	3.17374600
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H	-5.86948600	3.62979800	2.81240300
H	-5.69618800	2.25439100	1.70408900
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H	-3.66016800	1.53358500	3.09054700
H	-3.86766600	2.96875500	4.09997600
H	-2.30667900	2.65312500	3.33339300
H	-5.15951100	-3.79946700	-1.03139100
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H	-0.48210100	-0.74054200	6.55033100
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H	4.01139300	4.66739100	0.57365500
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H	-3.25071900	-2.39740000	-3.77484700
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H	-1.32903800	-5.53246000	-2.98616300
H	-1.85005100	-5.68017500	-1.29684400
H	-0.71071800	-4.40182400	-1.76238400
H	-4.83272400	-4.28616500	-3.13694300
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H	-7.07529700	-2.57687100	-0.92413500
H	-6.89844100	-0.94678100	-0.26014200
H	-8.19303400	-2.01151600	0.32684200
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H	-6.51501900	-4.63860800	0.57623700
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H	-6.18997100	-0.60571900	2.18065400
H	-5.81535000	-2.00774700	3.20318200
H	-7.47626300	-1.71517900	2.67278300

H	4.37038400	5.82545500	3.91175000
H	5.11595100	5.04321200	2.51287800
H	3.69593400	6.08944100	2.29929000
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H	4.88580100	2.79285200	3.71394900
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H	5.38182200	4.69000300	-1.19841100
H	4.85128100	5.18919800	-2.80804800
H	3.95488500	5.73191600	-1.38576500
H	4.83456300	2.85995700	-3.58371000
H	5.29431100	2.23797500	-1.98588500
H	3.84992000	1.61666800	-2.80057700
H	1.76578600	3.13378800	-3.05879400
H	2.77984800	4.35170800	-3.84490000
H	1.80178900	4.78873500	-2.42973400

2. Computed transitions for Co**(NNO)**$_2$ (S = ½)

Excited State 1: 2.002-A 0.3445 eV 3598.99 nm f=0.0293 <S**2>=0.752
297B -> 298B 1.02148
297B <- 298B -0.23114
This state for optimization and/or second-order correction.
Total Energy, E(TD-HF/TD-DFT) = -4624.37841599
Copying the excited state density for this state as the 1-particle RhoCI density.

Excited State 2: 2.032-A 1.1436 eV 1084.15 nm f=0.0138 <S**2>=0.782
295B -> 298B -0.15698
296B -> 298B 0.97595

Excited State 3: 2.089-A 1.1816 eV 1049.28 nm f=0.0276 <S**2>=0.841
295B -> 298B 0.96050
296B -> 298B 0.15349

Excited State 4: 3.420-A 1.2175 eV 1018.35 nm f=0.0008 <S**2>=2.674
278A -> 301A 0.12806
284A -> 301A -0.24184
286A -> 301A -0.38786
293A -> 301A 0.11282
295A -> 301A -0.22966
296A -> 301A -0.10873
297A -> 301A -0.27511
280B -> 301B -0.13547
284B -> 301B 0.19595
286B -> 301B 0.43060
291B -> 301B -0.11826
293B -> 301B -0.10664
295B -> 298B 0.17001
295B -> 301B 0.28212

297B -> 301B 0.40598
286A <- 301A -0.10397
286B <- 301B 0.10723

Excited State 5: 2.940-A 1.4662 eV 845.63 nm f=0.0004 <S**2>=1.910
262A -> 301A -0.11176
281A -> 301A 0.17143
283A -> 301A -0.31149
287A -> 301A 0.27357
296A -> 301A -0.19666
298A -> 301A 0.62822
281B -> 301B 0.15187
283B -> 301B 0.22062
287B -> 301B -0.25443
296B -> 301B 0.22075

3. VO(**NNO**)(acac) (*S* = 0)

V	0.39922200	-0.52627500	-1.39072100
O	-2.65044900	2.30777700	-0.37471600
O	-0.00029100	-0.14538800	-2.86173600
O	2.16353700	-1.20277700	-1.39403800
O	-0.32480100	-2.37060600	-1.52570800
O	0.35634600	-1.06309400	0.76090300
N	1.32928700	1.08444800	-0.79704600
N	-1.37012500	0.50556400	-0.68453900
C	-2.61397000	0.07570300	-0.21128200
C	-3.11343200	-1.20125500	0.05207000
C	-4.42698500	-1.30099800	0.51301900
C	-5.18960600	-0.11973900	0.69207600
C	-4.72891100	1.17594000	0.43201200
C	-3.40905200	1.20591600	-0.02667700
C	-1.43627500	1.81522100	-0.75201000
C	-0.40629900	2.75884300	-1.12634800
C	-0.77138600	4.08125400	-1.44702200
C	0.18672700	5.03008000	-1.76282400
C	1.54068800	4.66447400	-1.75992900
C	1.92492100	3.37340800	-1.43292400
C	0.96936800	2.38529500	-1.10619300
C	2.54342600	0.75353100	-0.19866800
C	3.29394600	1.54275500	0.68761300
C	4.52306200	1.08500400	1.15032200
C	4.97893200	-0.17411100	0.69216700
C	4.26456100	-1.00853300	-0.16834000
C	2.99287000	-0.53849900	-0.58476200
C	-5.07749900	-2.65980700	0.84157500
C	-5.44852500	-2.70262300	2.34327800
C	-4.13543000	-3.84215400	0.54602800
C	-6.35616300	-2.84720100	-0.00949200
C	-5.56948600	2.44647200	0.62462400
C	-4.86984200	3.37872900	1.64328200

C	-5.71519900	3.17626400	-0.73274100
C	-6.98018000	2.12791400	1.15338800
C	4.81306000	-2.36396800	-0.65101800
C	4.87999800	-2.37028400	-2.19796900
C	6.23147000	-2.64199800	-0.11639200
C	3.89227200	-3.50847200	-0.16641500
C	0.48193100	-2.25956500	2.79839100
C	0.28653200	-2.20258900	1.29712300
C	0.03115400	-3.40219000	0.58150600
C	-0.25856700	-3.41893400	-0.78015800
C	-0.55719300	-4.71944200	-1.48883900
C	5.39344400	1.89194900	2.13183100
C	6.76212900	2.20210500	1.47953500
C	5.61801700	1.06980700	3.42351200
C	4.74039700	3.22994300	2.52743200
H	-2.48621600	-2.06570000	-0.10503600
H	-6.20380900	-0.22832600	1.05542800
H	-1.82336400	4.34558300	-1.45112900
H	-0.11100200	6.04138500	-2.02181600
H	2.30035000	5.39334400	-2.02958400
H	2.97179600	3.09362600	-1.45264300
H	2.89059500	2.49652200	1.00482500
H	5.94589100	-0.51384100	1.04228800
H	-5.91775600	-3.66213800	2.59278500
H	-6.15211400	-1.90813700	2.61383300
H	-4.55700700	-2.58713800	2.97022000
H	-4.63675200	-4.78338700	0.79816800
H	-3.86288200	-3.88627000	-0.51464000
H	-3.21187700	-3.79014500	1.13276900
H	-6.82103600	-3.81532600	0.21309900
H	-6.12123000	-2.82117900	-1.07954800
H	-7.10154900	-2.06989000	0.18823300
H	-5.46183800	4.29086900	1.78541900
H	-4.76805700	2.88640900	2.61719500
H	-3.87233000	3.67371700	1.30606300
H	-6.31113600	4.08825600	-0.60716900
H	-6.22129200	2.53908100	-1.46681100
H	-4.74401300	3.46308100	-1.14593200
H	-7.54113100	3.06084700	1.27664700
H	-7.54387700	1.49315900	0.46036900
H	-6.94881900	1.63011100	2.12923900
H	5.25885800	-3.33678800	-2.55374500
H	3.89540000	-2.20158000	-2.63932900
H	5.55882600	-1.58866300	-2.55952100
H	6.58100900	-3.60561200	-0.50405600
H	6.94927700	-1.87799800	-0.43693500
H	6.25682700	-2.70046500	0.97818600
H	4.27597300	-4.47476000	-0.51806000
H	2.87348100	-3.38630200	-0.53823300
H	3.85508400	-3.54020600	0.92945400
H	0.41838600	-3.27574600	3.19623400

H	-0.27452800	-1.63398600	3.28569500
H	1.46024600	-1.83377700	3.04755800
H	0.02883100	-4.34025000	1.12390900
H	-0.55710600	-5.57494100	-0.80862900
H	-1.52964800	-4.65239500	-1.98922700
H	0.19297700	-4.88288100	-2.27134000
H	7.39526100	2.77252900	2.17063900
H	6.63460500	2.79554300	0.56684400
H	7.30303600	1.28908700	1.20992300
H	6.24234000	1.63012600	4.13074300
H	4.66391900	0.84675000	3.91475000
H	6.11880200	0.11764400	3.21971100
H	5.39226800	3.76779100	3.22525000
H	3.77516300	3.08105600	3.02464000
H	4.58076600	3.87880900	1.65860300

4. Computed transitions for VO(**NNO**)(acac) (S = 0)

Excited State 1: 3.000-A -0.6944 eV -1785.51 nm f=-0.0000 <S**2>=2.000
181A -> 185A -0.13452
184A -> 185A -0.71574
181B -> 185B 0.13452
184B -> 185B 0.71574
184A <- 185A -0.22503
184B <- 185B 0.22503
This state for optimization and/or second-order correction.
Total Energy, E(TD-HF/TD-DFT) = -2985.53627154
Copying the excited state density for this state as the 1-particle RhoCI density.

Excited State 2: 1.000-A 1.7429 eV 711.36 nm f=0.0590 <S**2>=0.000
184A -> 185A 0.67862
184A -> 188A 0.11942
184B -> 185B 0.67862
184B -> 188B 0.11942

Excited State 3: 3.000-A 1.8869 eV 657.07 nm f=0.0000 <S**2>=2.000
183A -> 185A -0.66665
184A -> 187A -0.17086
183B -> 185B 0.66665
184B -> 187B 0.17086

Excited State 4: 3.000-A 2.1674 eV 572.05 nm f=0.0000 <S**2>=2.000
183A -> 185A 0.17950
184A -> 186A -0.31028
184A -> 187A -0.58259
183B -> 185B -0.17950
184B -> 186B 0.31028
184B -> 187B 0.58259

Excited State 5: 1.000-A 2.2720 eV 545.70 nm f=0.0397 <S**2>=0.000
183A -> 185A 0.69268

183B -> 185B 0.69268

Excited State 6: 3.000-A 2.2774 eV 544.41 nm f=0.0000 <S**2>=2.000
184A -> 186A 0.29847
184A -> 187A -0.24204
184A -> 188A -0.47967
184A -> 189A -0.31364
184B -> 186B -0.29847
184B -> 187B 0.24204
184B -> 188B 0.47967
184B -> 189B 0.31364

Excited State 7: 3.000-A 2.3955 eV 517.58 nm f=0.0000 <S**2>=2.000
181A -> 185A -0.64932
182A -> 185A 0.17018
184A -> 185A 0.12675
181B -> 185B 0.64932
182B -> 185B -0.17018
184B -> 185B -0.12675

Excited State 8: 1.000-A 2.4561 eV 504.80 nm f=0.0019 <S**2>=0.000
184A -> 186A 0.31170
184A -> 187A 0.61576
184B -> 186B 0.31170
184B -> 187B 0.61576

Excited State 9: 3.000-A 2.4641 eV 503.15 nm f=0.0000 <S**2>=2.000
182A -> 185A -0.12506
183A -> 186A -0.14254
184A -> 186A 0.48835
184A -> 187A -0.22094
184A -> 188A 0.27215
184A -> 189A 0.23827
184A -> 190A -0.10520
182B -> 185B 0.12506
183B -> 186B 0.14254
184B -> 186B -0.48835
184B -> 187B 0.22094
184B -> 188B -0.27215
184B -> 189B -0.23827
184B -> 190B 0.10520

Excited State 10: 1.000-A 2.5924 eV 478.26 nm f=0.0348 <S**2>=0.000
181A -> 185A -0.14107
182A -> 185A 0.11567
184A -> 186A 0.52134
184A -> 187A -0.31104
184A -> 188A -0.23318
184A -> 189A -0.13018
181B -> 185B -0.14107
182B -> 185B 0.11567

184B -> 186B	0.52134
184B -> 187B	-0.31104
184B -> 188B	-0.23318
184B -> 189B	-0.13018

5. VO(**NNO**)(acac) (*S* = 1)

V	0.29751500	-0.66320700	-1.26835600
O	-2.54344800	2.36464300	-0.17502800
O	0.20063500	-0.38806700	-2.81960800
O	2.10857800	-1.48582000	-0.99818600
O	-0.51683500	-2.49363700	-1.23250800
O	0.19849000	-0.95345700	0.95027000
N	1.44104100	1.02791100	-0.74297800
N	-1.42074400	0.51190100	-0.70829600
C	-2.71106200	0.14096900	-0.31528500
C	-3.32500800	-1.11020800	-0.23272600
C	-4.64912300	-1.15214300	0.20472200
C	-5.30852000	0.05835400	0.53715100
C	-4.73128300	1.33065400	0.45877800
C	-3.40576600	1.30222100	0.01550900
C	-1.37136200	1.81850500	-0.60932300
C	-0.29795200	2.73869800	-0.94590100
C	-0.64729100	4.07490900	-1.21745300
C	0.30007100	5.00567600	-1.61772700
C	1.62863500	4.59648100	-1.77569000
C	2.00125400	3.29197500	-1.48819700
C	1.06807600	2.33237000	-1.02905300
C	2.67784800	0.68924400	-0.29107400
C	3.59402700	1.54167500	0.38114100
C	4.82522200	1.07297900	0.78687300
C	5.16460000	-0.28758900	0.49655500
C	4.31726500	-1.19408800	-0.10987600
C	3.01142800	-0.71691300	-0.48133300
C	-5.42133600	-2.47928100	0.34223500
C	-5.86401300	-2.67011400	1.81271700
C	-4.56168700	-3.69307700	-0.05821600
C	-6.66959400	-2.45235200	-0.57158200
C	-5.46031800	2.63414700	0.81510700
C	-4.69848600	3.35768200	1.95189300
C	-5.51964600	3.54709100	-0.43347300
C	-6.90193000	2.37532400	1.29037700
C	4.71983200	-2.65657500	-0.37245500
C	4.59126200	-2.97472700	-1.88285700
C	6.17810200	-2.93625200	0.04145200
C	3.80731100	-3.60503800	0.44316600
C	0.27439500	-1.96756300	3.08949000
C	0.08814500	-2.03965700	1.58538400
C	-0.19646900	-3.29259600	0.98692100
C	-0.47102200	-3.44754800	-0.37299600
C	-0.77692800	-4.81962400	-0.93300100

C	5.84012700	1.94418200	1.54472700
C	7.14632400	2.04808700	0.71980200
C	6.15130000	1.30093900	2.91815900
C	5.31700300	3.37124900	1.79371100
H	-2.77042800	-1.99251000	-0.51456300
H	-6.33553400	-0.00715800	0.87416800
H	-1.68764200	4.36702600	-1.13030500
H	0.00392900	6.02726800	-1.83438000
H	2.37679100	5.29619800	-2.13842200
H	3.02596200	2.97922500	-1.64836300
H	3.28688800	2.55519700	0.60015200
H	6.14863600	-0.62618900	0.79561300
H	-6.41546400	-3.61163700	1.92432100
H	-6.51810100	-1.86057800	2.15360400
H	-4.99627400	-2.70331900	2.48129200
H	-5.14382200	-4.61324000	0.06520800
H	-4.24700900	-3.63872200	-1.10657200
H	-3.66452200	-3.78009900	0.56472700
H	-7.22519500	-3.39397000	-0.48376600
H	-6.38303100	-2.32365800	-1.62143600
H	-7.35385700	-1.63841800	-0.30939500
H	-5.21085900	4.29172100	2.21225200
H	-4.65527300	2.73335700	2.85163200
H	-3.67375900	3.60537900	1.66129900
H	-6.03390300	4.48471100	-0.19037500
H	-6.06841200	3.06063300	-1.24782600
H	-4.52026900	3.79713700	-0.80107800
H	-7.38075000	3.32980600	1.53528500
H	-7.50794700	1.89126700	0.51620600
H	-6.92994800	1.75099700	2.19062000
H	4.88155700	-4.01614700	-2.06948800
H	3.56840600	-2.83256000	-2.23573400
H	5.25430600	-2.33073400	-2.47316900
H	6.42134100	-3.98093900	-0.18182800
H	6.89021100	-2.30817600	-0.50700600
H	6.34079000	-2.78577800	1.11535200
H	4.08521700	-4.64857900	0.24807500
H	2.75561600	-3.47301900	0.18367800
H	3.92306500	-3.42294600	1.51876100
H	0.15288700	-2.93698200	3.58020000
H	-0.44783400	-1.25884200	3.51041800
H	1.27405600	-1.57632700	3.31059400
H	-0.22724000	-4.17182600	1.61969600
H	-0.79235800	-5.59356000	-0.16119600
H	-1.74338100	-4.80327200	-1.44891000
H	-0.01987800	-5.07665300	-1.68315000
H	7.88341500	2.66066200	1.25322100
H	6.95821100	2.51391700	-0.25423900
H	7.59889800	1.06758100	0.53939300
H	6.88055200	1.91042000	3.46573700
H	5.24434000	1.22537100	3.52861600

H	6.57041700	0.29466500	2.81580600
H	6.07302600	3.95199600	2.33391800
H	4.40469200	3.37091600	2.40072800
H	5.10461700	3.89777200	0.85628500

6. Computed transitions for VO(**NNO**)(acac) (*S* = 1)

Excited State 1: 3.030-A 1.4360 eV 863.40 nm f=0.0529 <S**2>=2.046
181B -> 184B -0.12310
183B -> 184B 0.97406
This state for optimization and/or second-order correction.
Total Energy, E(TD-HF/TD-DFT) = -2985.45990018
Copying the excited state density for this state as the 1-particle RhoCI density.

Excited State 2: 3.018-A 1.8608 eV 666.30 nm f=0.0008 <S**2>=2.027
182A -> 186A 0.10574
182A -> 187A -0.12029
182A -> 188A 0.23258
182A -> 189A 0.27380
184A -> 186A 0.20076
184A -> 187A -0.19146
184A -> 188A 0.34854
184A -> 189A 0.41003
184A -> 193A -0.10141
185A -> 186A -0.31056
185A -> 187A 0.18300
185A -> 188A -0.32591
185A -> 189A -0.35347

Excited State 3: 3.015-A 1.8908 eV 655.71 nm f=0.0002 <S**2>=2.022
181A -> 190A 0.10444
182A -> 187A -0.12814
182A -> 188A -0.22877
182A -> 190A 0.33113
184A -> 187A -0.21430
184A -> 188A -0.35515
184A -> 190A 0.49130
185A -> 187A 0.19020
185A -> 188A 0.31618
185A -> 190A -0.41836

Excited State 4: 3.022-A 1.9626 eV 631.72 nm f=0.0010 <S**2>=2.033
182B -> 184B 0.99335

Excited State 5: 3.089-A 2.1827 eV 568.03 nm f=0.0023 <S**2>=2.136
182A -> 189A 0.11239
182A -> 194A -0.15844
182A -> 195A 0.17056
184A -> 189A 0.17592
184A -> 194A -0.21593
184A -> 195A 0.23482

185A -> 186A 0.53784
185A -> 189A -0.17754
185A -> 194A 0.18766
185A -> 195A -0.16892
178B -> 184B -0.20687
181B -> 184B -0.50137

Excited State 6: 3.055-A 2.2785 eV 544.15 nm f=0.0402 <S**2>=2.084
182A -> 194A -0.17792
182A -> 195A 0.17765
184A -> 186A -0.16402
184A -> 189A 0.10813
184A -> 194A -0.25207
184A -> 195A 0.24622
185A -> 186A 0.15352
185A -> 194A 0.18984
185A -> 195A -0.20176
178B -> 184B 0.22011
180B -> 184B 0.11128
181B -> 184B 0.69693
183B -> 184B 0.12448

Excited State 7: 3.058-A 2.4460 eV 506.89 nm f=0.0490 <S**2>=2.088
182A -> 194A 0.15046
182A -> 195A -0.15432
184A -> 186A 0.21618
184A -> 187A -0.15501
184A -> 194A 0.21243
184A -> 195A -0.21612
185A -> 186A 0.71388
185A -> 187A 0.12585
185A -> 194A -0.17048
185A -> 195A 0.17196
180B -> 184B 0.25255
181B -> 184B 0.23831

Excited State 8: 3.026-A 2.4657 eV 502.84 nm f=0.0072 <S**2>=2.040
185A -> 186A -0.18192
178B -> 184B -0.14742
179B -> 184B 0.21444
180B -> 184B 0.92266
181B -> 184B -0.11714

Excited State 9: 3.072-A 2.6717 eV 464.07 nm f=0.0176 <S**2>=2.110
185A -> 188A -0.10349
185A -> 189A 0.11189
177B -> 184B -0.15948
178B -> 184B 0.86395
180B -> 184B 0.12877
181B -> 184B -0.34389

Excited State 10: 3.077-A 2.8016 eV 442.55 nm f=0.0029 <S**2>=2.117
177B -> 184B -0.20283
179B -> 184B 0.93342
180B -> 184B -0.17852

7. VO(**NNO**)(acac) broken symmetry (*S* = 0)

V	0.33764800	-0.61250200	-1.35838600
O	-2.58739900	2.34328600	-0.28337400
O	0.07717800	-0.29075900	-2.88023900
O	2.15725400	-1.37922400	-1.21375900
O	-0.42437600	-2.47182700	-1.38793300
O	0.28705700	-1.00916700	0.83969600
N	1.39416500	1.05061700	-0.80607300
N	-1.39901600	0.50388900	-0.71413500
C	-2.66565900	0.10905800	-0.27372400
C	-3.22573600	-1.15733500	-0.09630300
C	-4.54052900	-1.22342600	0.36527900
C	-5.24426800	-0.02057600	0.62759100
C	-4.72181600	1.26592900	0.45370400
C	-3.40262800	1.26173600	-0.00902700
C	-1.39982700	1.81446500	-0.69733200
C	-0.34570400	2.74517300	-1.06027600
C	-0.70271700	4.07340300	-1.35763500
C	0.25106400	5.01151600	-1.72420000
C	1.59360900	4.62427700	-1.80769400
C	1.97175100	3.32586900	-1.49854000
C	1.02553100	2.35608600	-1.09812400
C	2.60264100	0.71903900	-0.26142100
C	3.43416200	1.55706600	0.52072700
C	4.65674700	1.10162200	0.97257900
C	5.06054300	-0.21984400	0.60969100
C	4.29190400	-1.10559200	-0.12639600
C	2.99824800	-0.64600300	-0.54120100
C	-5.25374700	-2.56848200	0.60774200
C	-5.63345100	-2.68720700	2.10321200
C	-4.36255000	-3.77074100	0.24269100
C	-6.53544000	-2.64409900	-0.25577500
C	-5.49906700	2.55984500	0.73511500
C	-4.75281100	3.38701000	1.80990400
C	-5.61161300	3.38506200	-0.56964600
C	-6.92258400	2.27556000	1.24875400
C	4.77533700	-2.52299400	-0.48309400
C	4.76805900	-2.70419200	-2.02127000
C	6.21041300	-2.78687000	0.01342500
C	3.84778700	-3.57457000	0.17193900
C	0.38540200	-2.09686700	2.94025000
C	0.19260500	-2.11916300	1.43585300
C	-0.08059300	-3.35240300	0.79424300
C	-0.36414400	-3.45809300	-0.56951000
C	-0.66315600	-4.80990800	-1.18011700

C	5.59125800	1.95159600	1.84971300
C	6.94254800	2.15273500	1.12160100
C	5.83763100	1.22830300	3.19600600
C	5.00176700	3.34129700	2.15570600
H	-2.63992300	-2.03494200	-0.32368300
H	-6.26209800	-0.10448900	0.98753700
H	-1.74903500	4.35401900	-1.31065700
H	-0.04775100	6.02726400	-1.96377900
H	2.34810900	5.33704600	-2.12935300
H	3.00887400	3.02641300	-1.59390200
H	3.07424400	2.54520200	0.77649400
H	6.03346400	-0.55054100	0.95136700
H	-6.14531700	-3.63884700	2.29152700
H	-6.30323300	-1.88116800	2.42140300
H	-4.74056000	-2.65026100	2.73761600
H	-4.90411400	-4.70299500	0.43878900
H	-4.08933200	-3.76361800	-0.81865600
H	-3.43957400	-3.79130400	0.83259200
H	-7.04614500	-3.60100500	-0.09318400
H	-6.29428000	-2.56467100	-1.32180100
H	-7.24442700	-1.84565400	-0.01268600
H	-5.29909100	4.31543400	2.01566700
H	-4.67311100	2.82637900	2.74828000
H	-3.74266800	3.65488100	1.48784100
H	-6.16160400	4.31483900	-0.38068700
H	-6.14958600	2.82386100	-1.34202700
H	-4.62815200	3.65088600	-0.96780000
H	-7.43712500	3.22423100	1.43729800
H	-7.51795200	1.71688100	0.51778000
H	-6.91343700	1.71219000	2.18873400
H	5.11093400	-3.71400300	-2.27916300
H	3.76787900	-2.56246000	-2.43465800
H	5.44595000	-1.98697300	-2.49966700
H	6.51270400	-3.79822500	-0.28038300
H	6.93320800	-2.08758500	-0.42340300
H	6.28890600	-2.72693800	1.10550500
H	4.18269100	-4.58534400	-0.09352200
H	2.81328500	-3.45648500	-0.15441500
H	3.87518700	-3.48843400	1.26522300
H	0.29309300	-3.08649600	3.39564300
H	-0.35384500	-1.42425500	3.39008000
H	1.37497100	-1.68573800	3.16997600
H	-0.09550900	-4.25480100	1.39413000
H	-0.67221000	-5.61256900	-0.43814600
H	-1.63097800	-4.77840900	-1.69270400
H	0.09310300	-5.03408700	-1.94148500
H	7.62222300	2.75110800	1.74064900
H	6.80002400	2.67660600	0.16950000
H	7.44028500	1.20125600	0.90772700
H	6.50868100	1.82167400	3.82917000
H	4.89691100	1.08345300	3.73921800

H	6.29750000	0.24461100	3.05563300
H	5.70024600	3.90919300	2.78050900
H	4.05343000	3.27088100	2.70028300
H	4.82969600	3.92183500	1.24215600

8. Computed transitions for VO(**NNO**)(acac) broken symmetry (S = 0)

Excited State 1: 1.618-A 1.1888 eV 1042.98 nm f=0.0241 <S**2>=0.404
182A -> 185A 0.14364
184A -> 185A 0.96307
184A -> 189A 0.10030
This state for optimization and/or second-order correction.
Total Energy, E(TD-HF/TD-DFT) = -2985.47331872
Copying the excited state density for this state as the 1-particle RhoCI density.

Excited State 2: 2.283-A 1.5537 eV 797.98 nm f=0.0453 <S**2>=1.053
182A -> 185A -0.10509
183A -> 185A 0.97217

Excited State 3: 2.297-A 1.9839 eV 624.94 nm f=0.0004 <S**2>=1.069
182A -> 188A 0.36944
182A -> 189A 0.11271
183A -> 188A -0.13737
184A -> 186A 0.20721
184A -> 187A 0.19141
184A -> 188A 0.77816
184A -> 189A 0.22607

Excited State 4: 2.295-A 2.0976 eV 591.06 nm f=0.0023 <S**2>=1.067
182A -> 185A 0.36870
182A -> 187A 0.16111
182A -> 188A -0.13841
182A -> 189A 0.25156
182A -> 190A -0.14832
184A -> 185A -0.21384
184A -> 186A 0.16405
184A -> 187A 0.35494
184A -> 188A -0.29681
184A -> 189A 0.52576
184A -> 190A -0.29587

Excited State 5: 2.230-A 2.2718 eV 545.75 nm f=0.0061 <S**2>=0.994
181A -> 185A -0.11759
182A -> 185A 0.88249
182A -> 189A -0.11218
184A -> 187A -0.19454
184A -> 188A 0.12269
184A -> 189A -0.22493
184A -> 190A 0.12989

Excited State 6: 2.402-A 2.3090 eV 536.97 nm f=0.0150 <S**2>=1.193

178A -> 185A 0.28363
180A -> 185A 0.12491
181A -> 185A 0.85066
182A -> 185A 0.12100
183A -> 185A 0.10111
184B -> 185B -0.27071

Excited State 7: 2.398-A 2.4815 eV 499.64 nm f=0.0024 <S**2>=1.188
180A -> 185A -0.17538
181A -> 185A -0.12192
182A -> 186A 0.11733
182A -> 189A -0.10373
182A -> 193A 0.27073
182A -> 194A -0.13883
184A -> 186A 0.50036
184A -> 187A 0.13136
184A -> 189A -0.24943
184A -> 191A 0.13265
184A -> 192A -0.12429
184A -> 193A 0.51660
184A -> 194A -0.25784
184B -> 185B -0.20186

Excited State 8: 2.291-A 2.5200 eV 492.01 nm f=0.0005 <S**2>=1.062
179A -> 185A 0.17946
180A -> 185A 0.95177

Excited State 9: 2.249-A 2.5623 eV 483.89 nm f=0.0748 <S**2>=1.014
181A -> 185A 0.25464
183A -> 186A 0.10375
184A -> 186A 0.10491
184A -> 193A 0.14128
184B -> 185B 0.90570

Excited State 10: 2.339-A 2.7757 eV 446.68 nm f=0.0219 <S**2>=1.118
178A -> 185A 0.87731
179A -> 185A -0.12446
181A -> 185A -0.32124
184B -> 186B 0.11662
184B -> 187B -0.15013