Supporting Information for

Models of the Ni-L and Ni-SI$_a$ States of the [NiFe]-Hydrogenase Active Site

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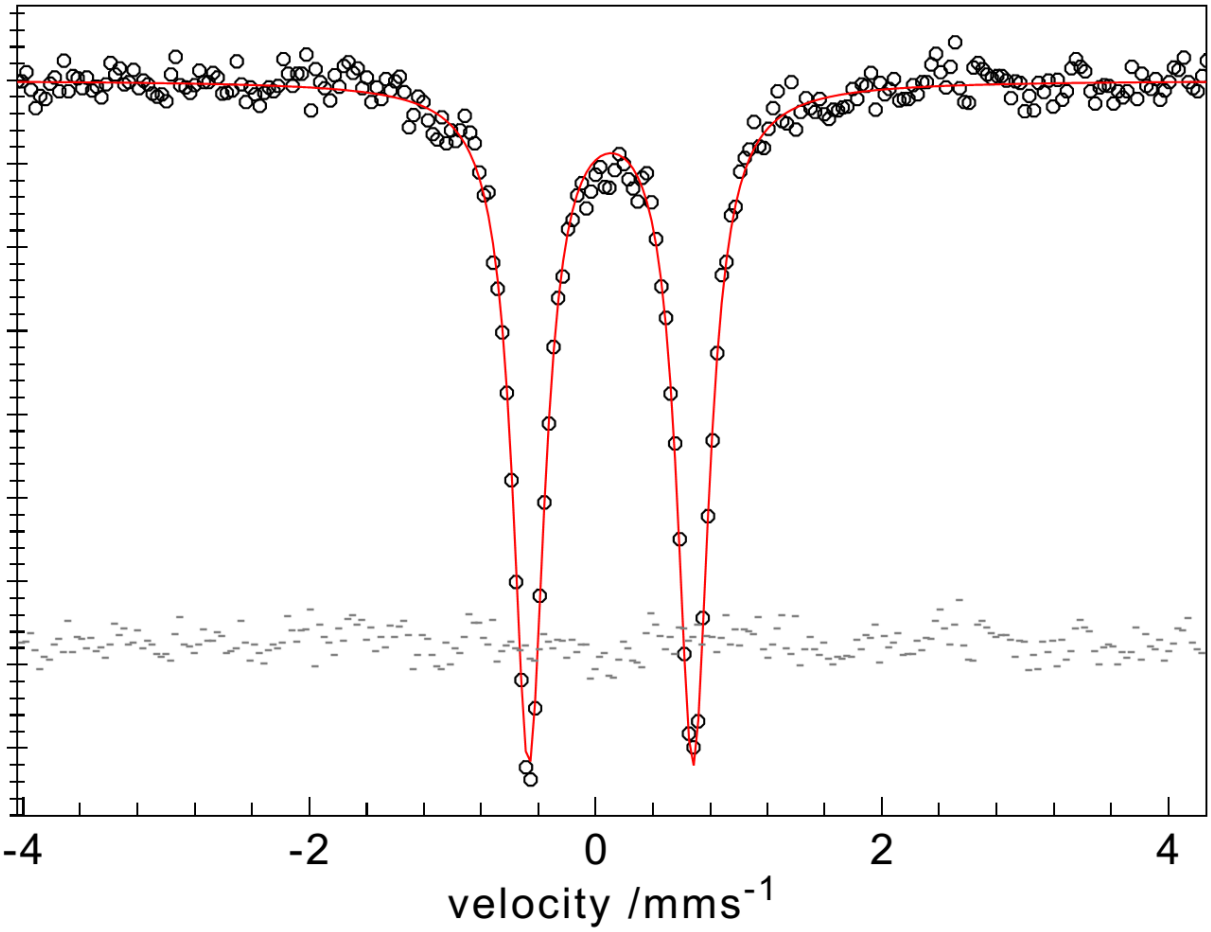

Figure S1. Mössbauer spectrum of [**1a**]$^{+}$ at 0T, 80K. Simulation parameters: $\delta = 0.11$, ΔE_q = 1.15 mm/s.

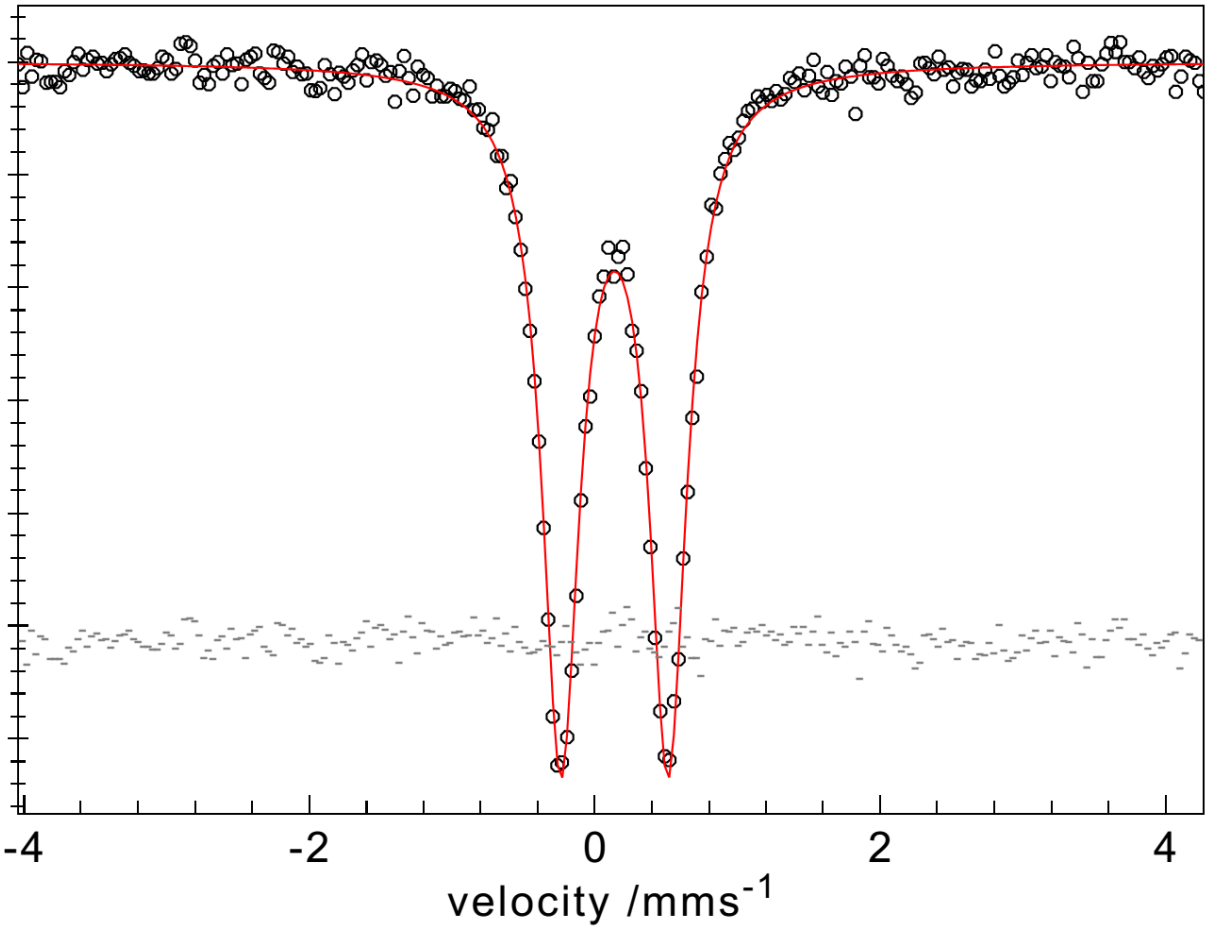

Figure S2. Mössbauer spectrum of [**1a**]^{0} at 0T, 80K. Simulation parameters: δ = 0.14, ΔE_q = -0.75 mm/s (sign obtained from Magnetic Mossbauer Spectra).

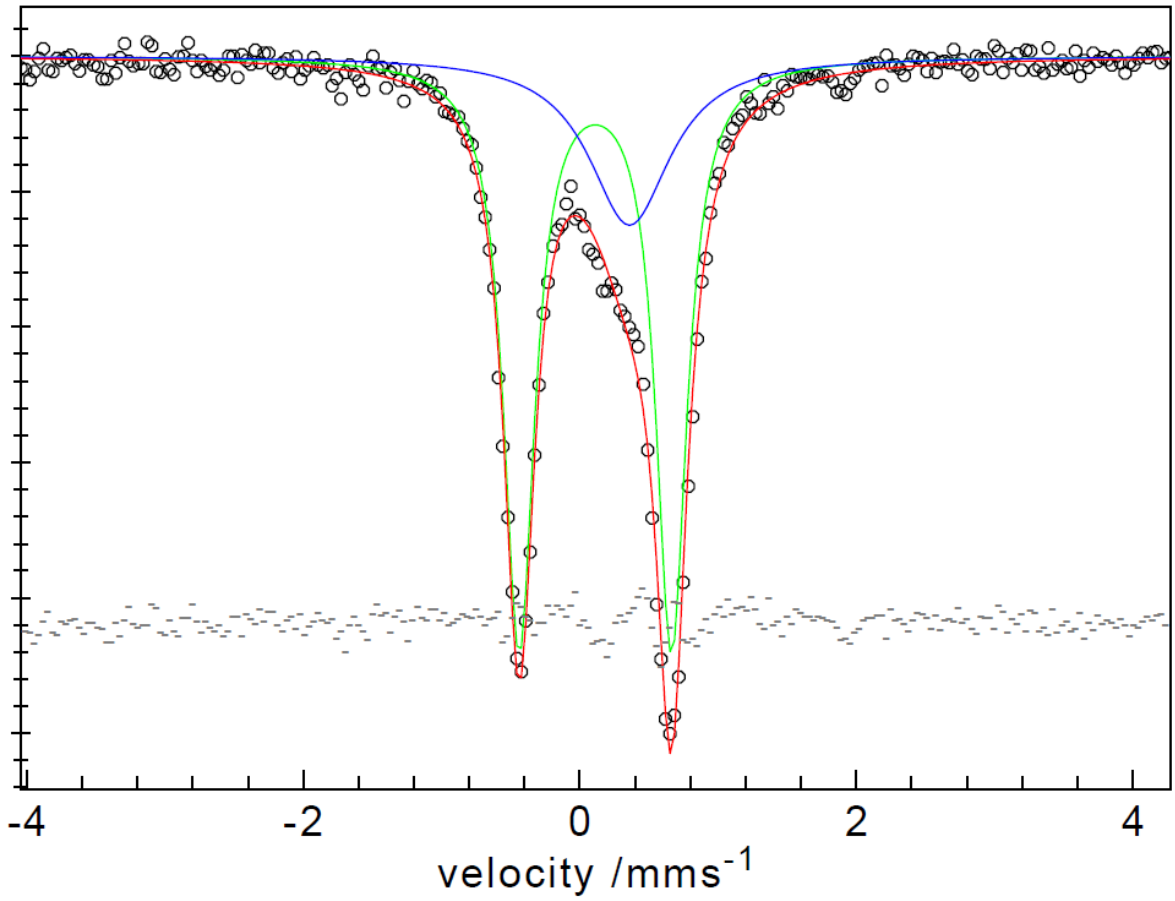

Figure S3. Mössbauer spectrum of [**1b**]$^{+}$ at 0T, 80K. Simulation parameters: $\delta = 0.11$, $\Delta E_q =$ 1.10 mm/s at 73% contribution. An unknown impurity was simulated with parameters $\delta =$ 0.36, $\Delta E_q = 0$ mm/s at 27% contribution.

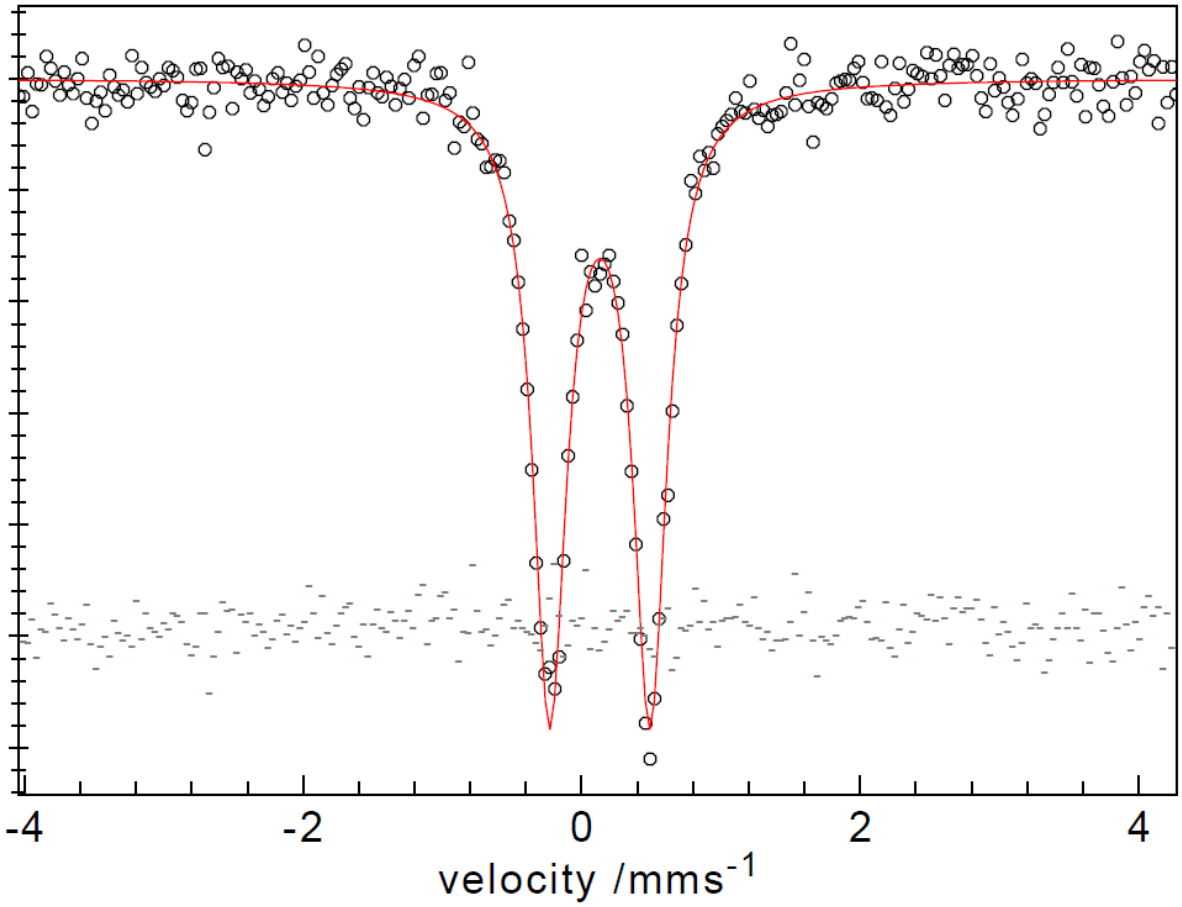

Figure S4. Mössbauer spectrum of [**1b**]^{0} at 0T, 80K. Simulation parameters: $\delta = 0.13$, $\Delta E_q =$ 0.72 mm/s.

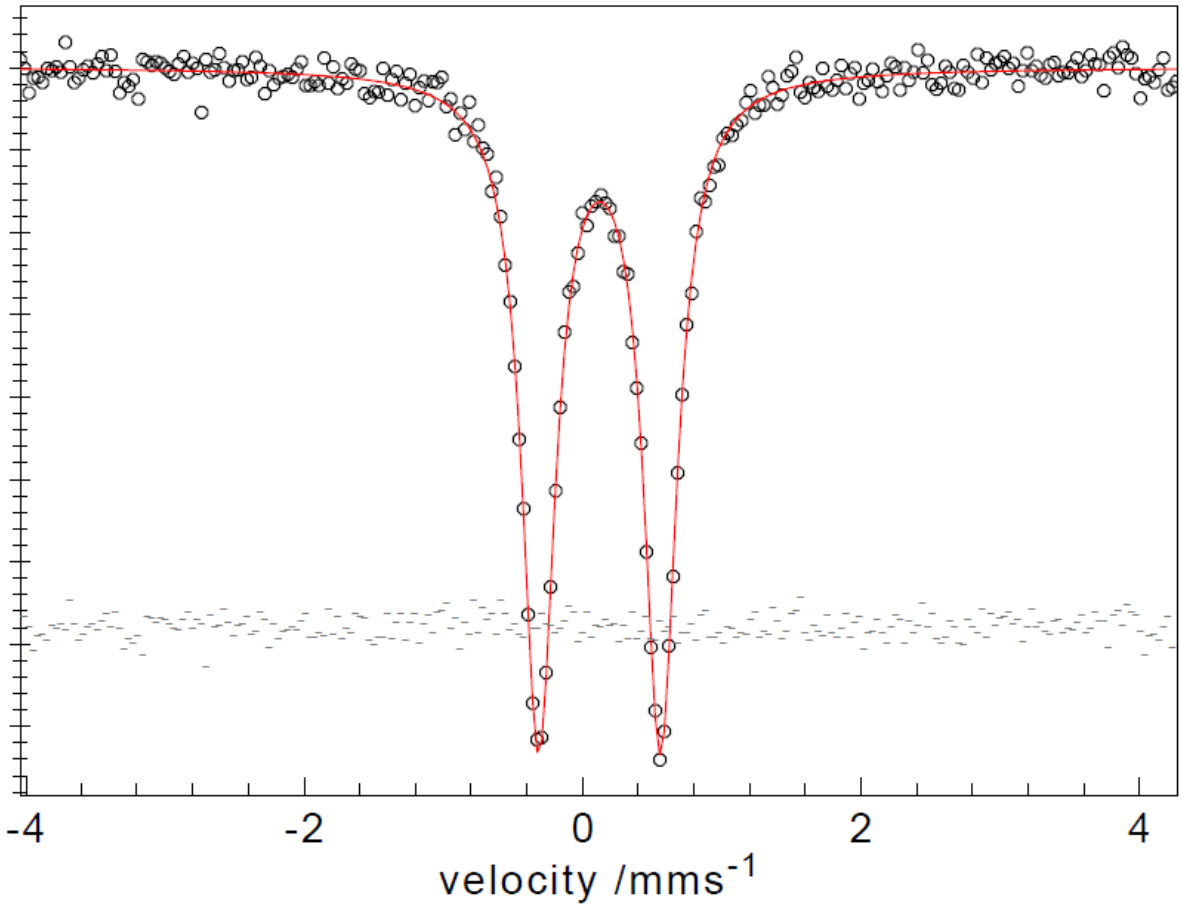

Figure S5. Mössbauer spectrum of $[\mathbf{1c}]^+$ at 0T, 80K. Simulation parameters: $\delta = 0.12$, $\Delta E_q = 0.87$ mm/s

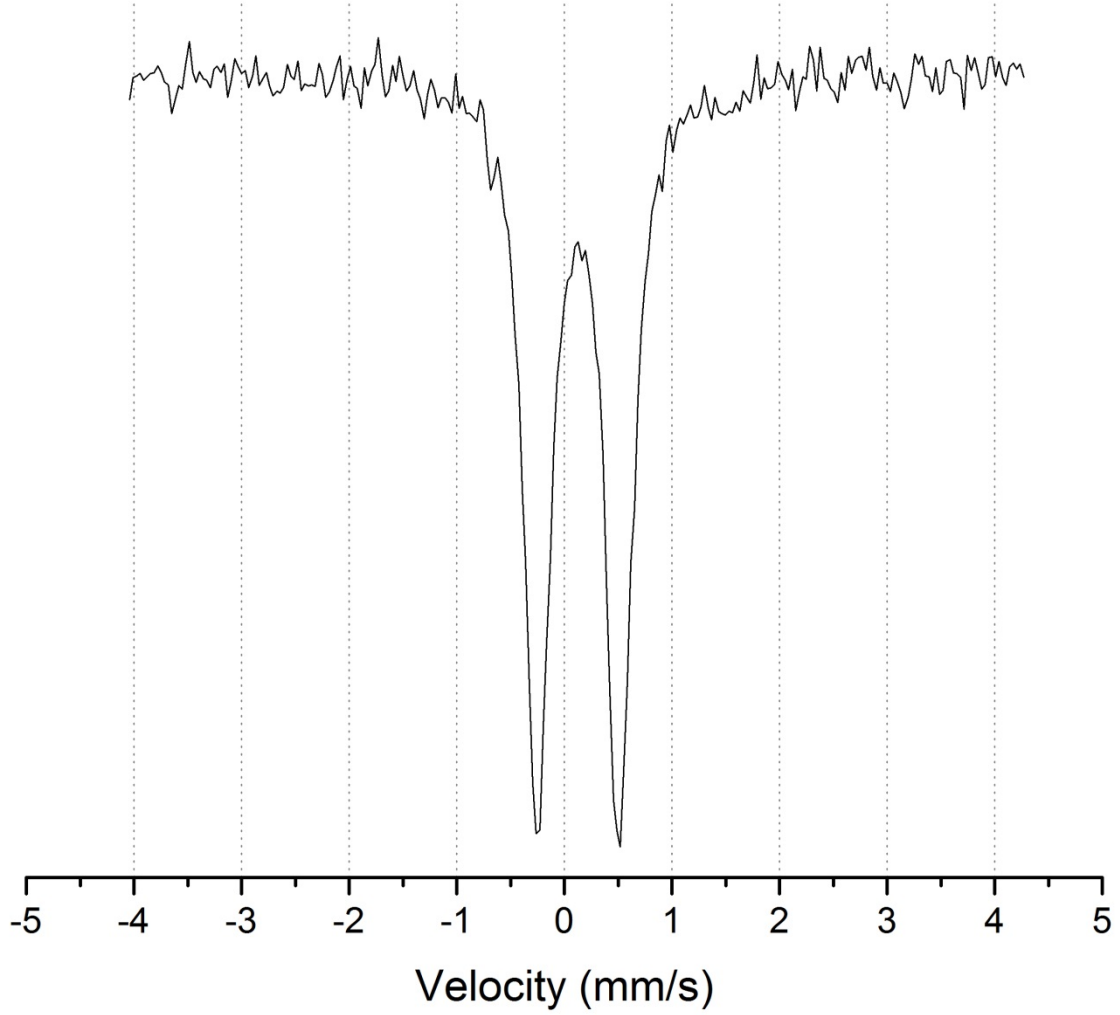

Figure S6. Mössbauer spectrum of $[\mathbf{1c}]^{\mathbf{0}}$ at 0T, 80K. Simulation parameters: $\delta = 0.13$, $\Delta E_q = 0.78$ mm/s.

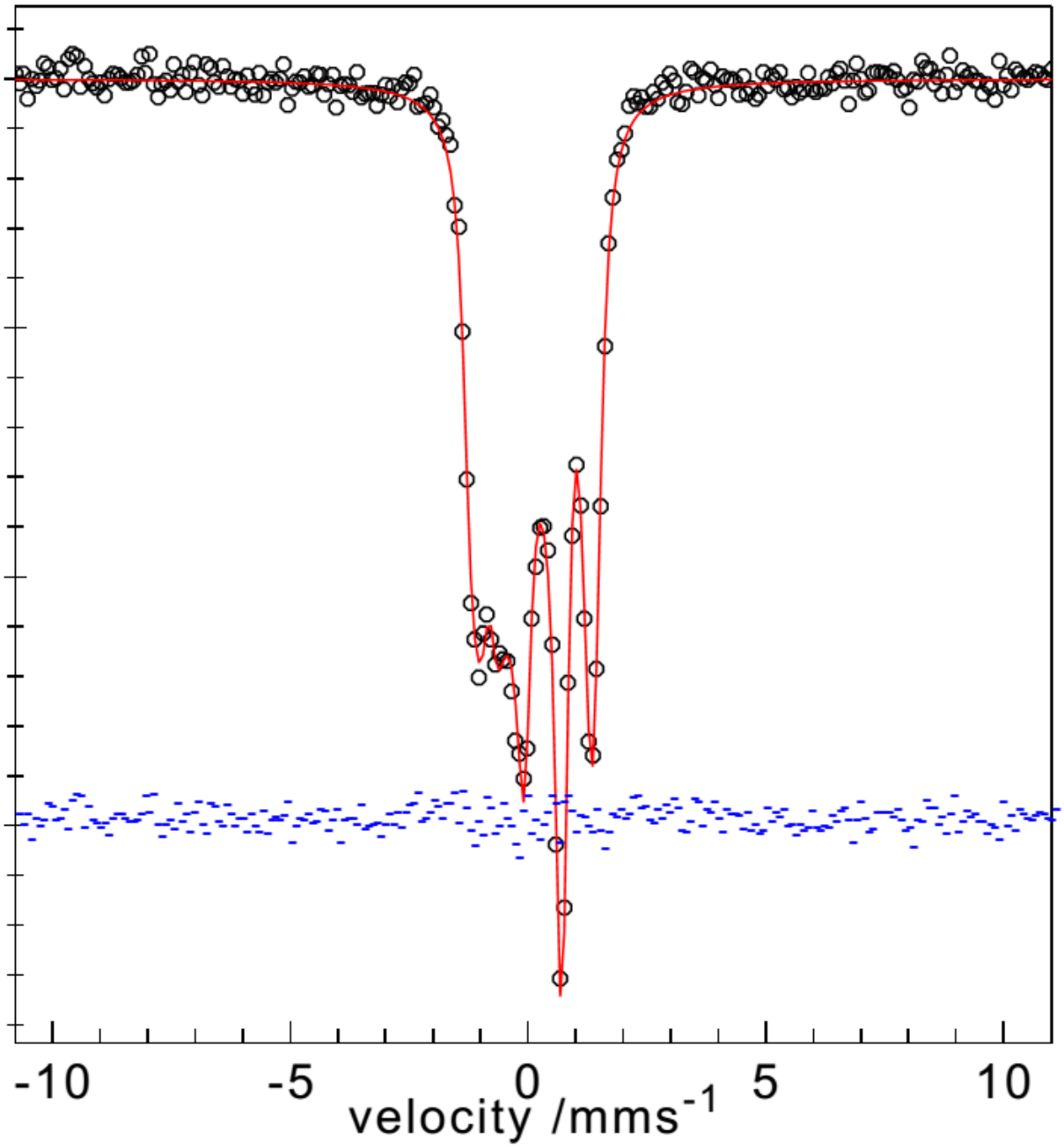

Figure S7. Mössbauer spectrum of [**1a**]^{0} at 7T, 4.2K. Simulation parameters: δ = 0.13, ΔE_q = -0.73 mm/s, η = 0.13. HFI (^{57}Fe) = (-28.81, -2.17, -6.02) kG = (-4.0 -0.30 -0.83) MHz.

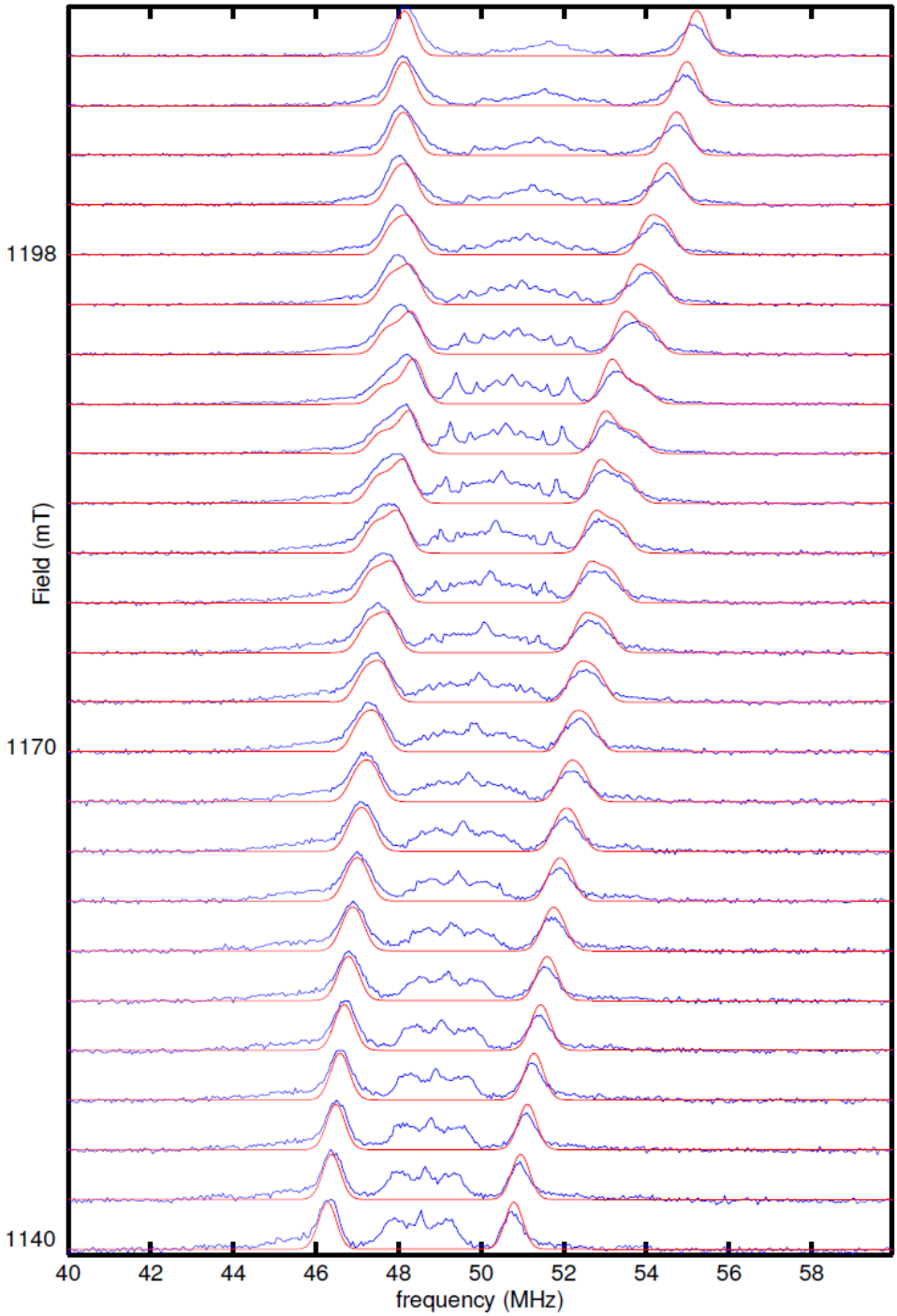

Figure S8. Q-band Davis ENDOR of $[\mathbf{1a}\text{-}d_5]^0$ in proton frequency range. Simulation parameters: A_{pdt} = (7.3, 4.4, 4.36) MHz; MW frequency = 34.0434 GHz. Temperature 10K. τ = 300 ns, T_{RF} = 18 µs.

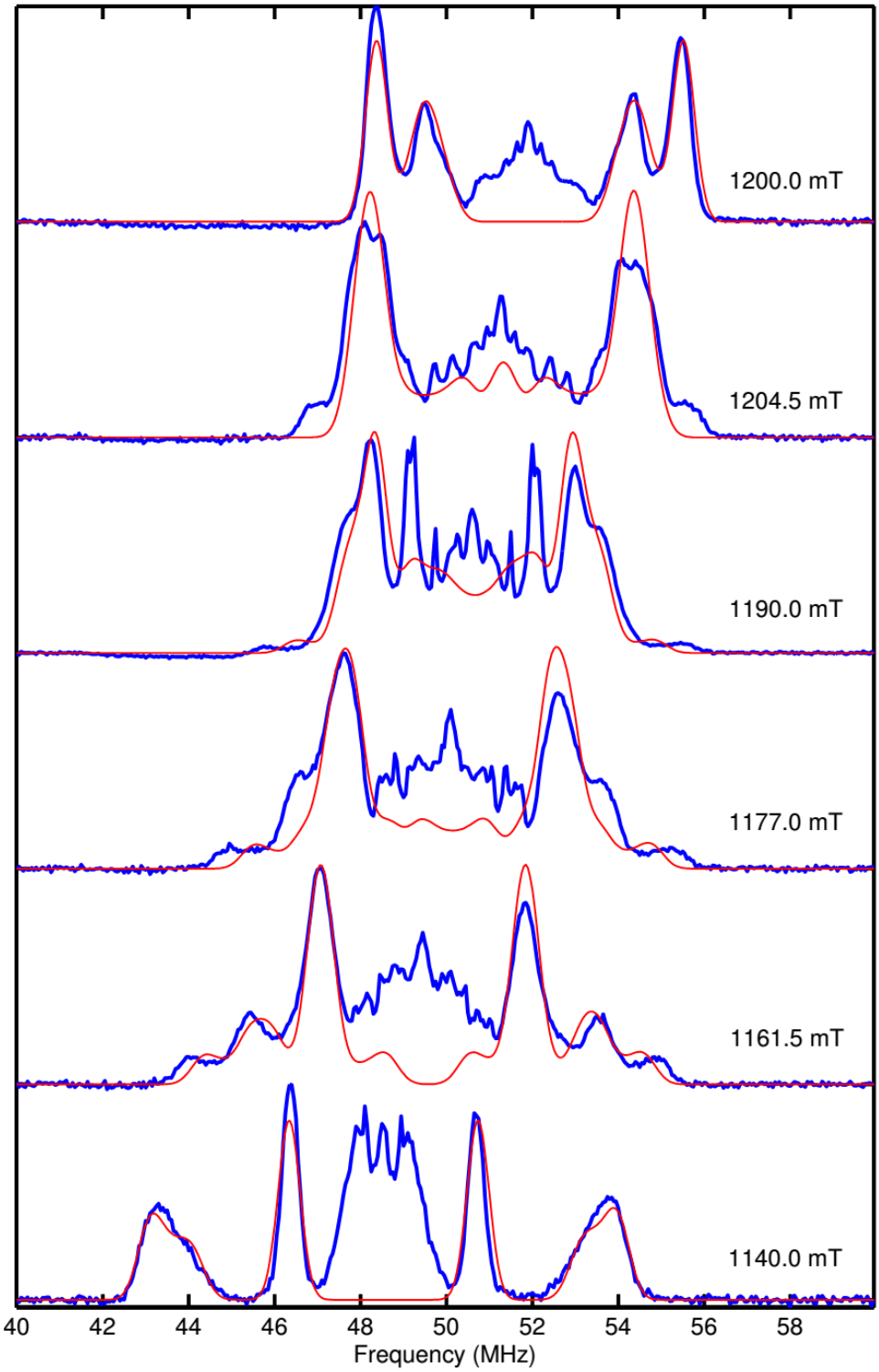

Figure S9. Q-band Davis ENDOR of [**1a**]^0 in proton range. Simulation parameters: A_{CP} = (-4.86, 3.25, -11.25) MHz; A_{pdt} = (7.3, 4.4, 4.36) MHz; MW frequency = 34.0434 GHz. Temperature 10K. τ = 300 ns, T_{RF} = 18 µs.

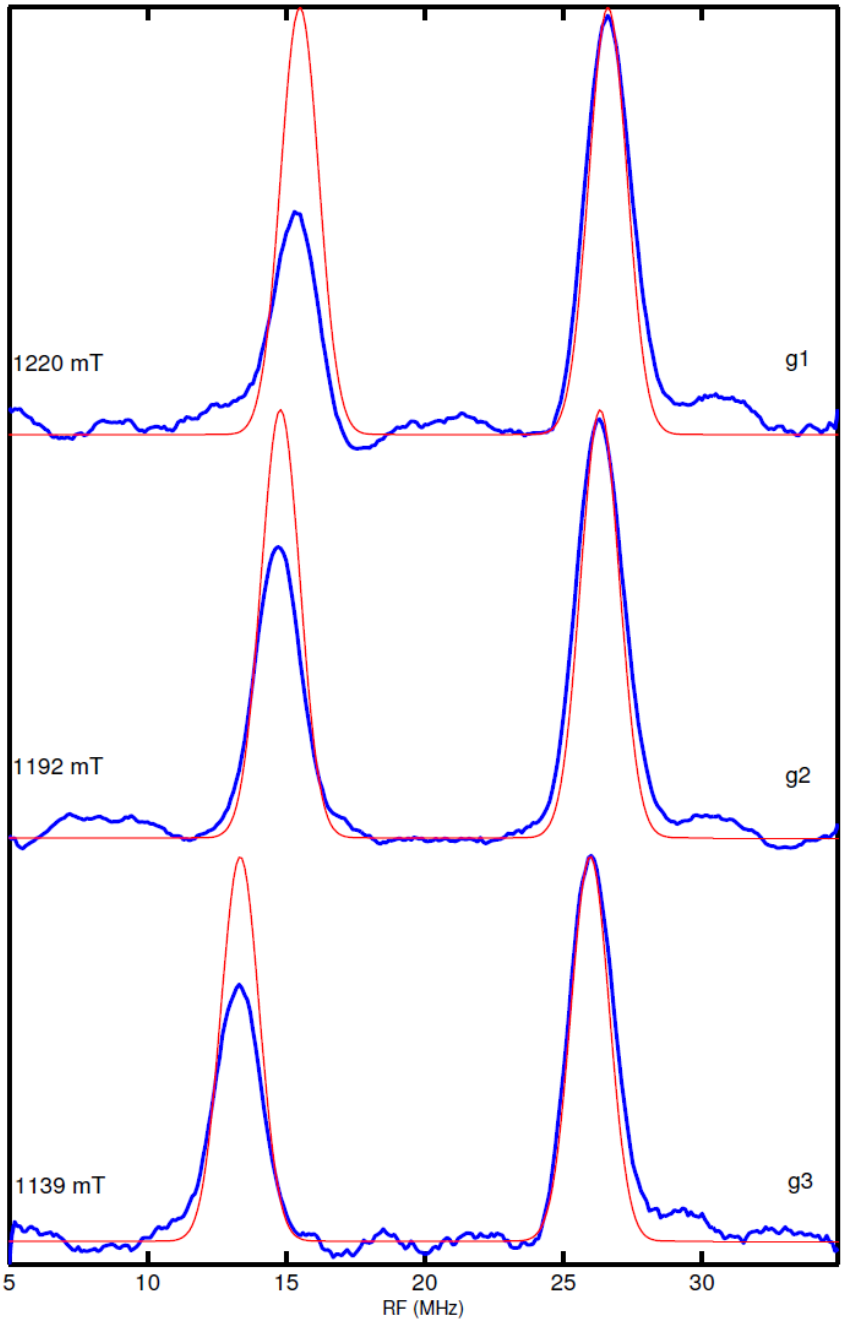

Figure S10. Q-band Davies ENDOR of [**1a**]^{0} in ^{31}P range. Simulation parameters: A= (11.1, 11.3, 12.7) MHz. MW frequency = 34.0434 GHz. Temperature 10K. τ = 300 ns, T_{RF} = 18 µs.

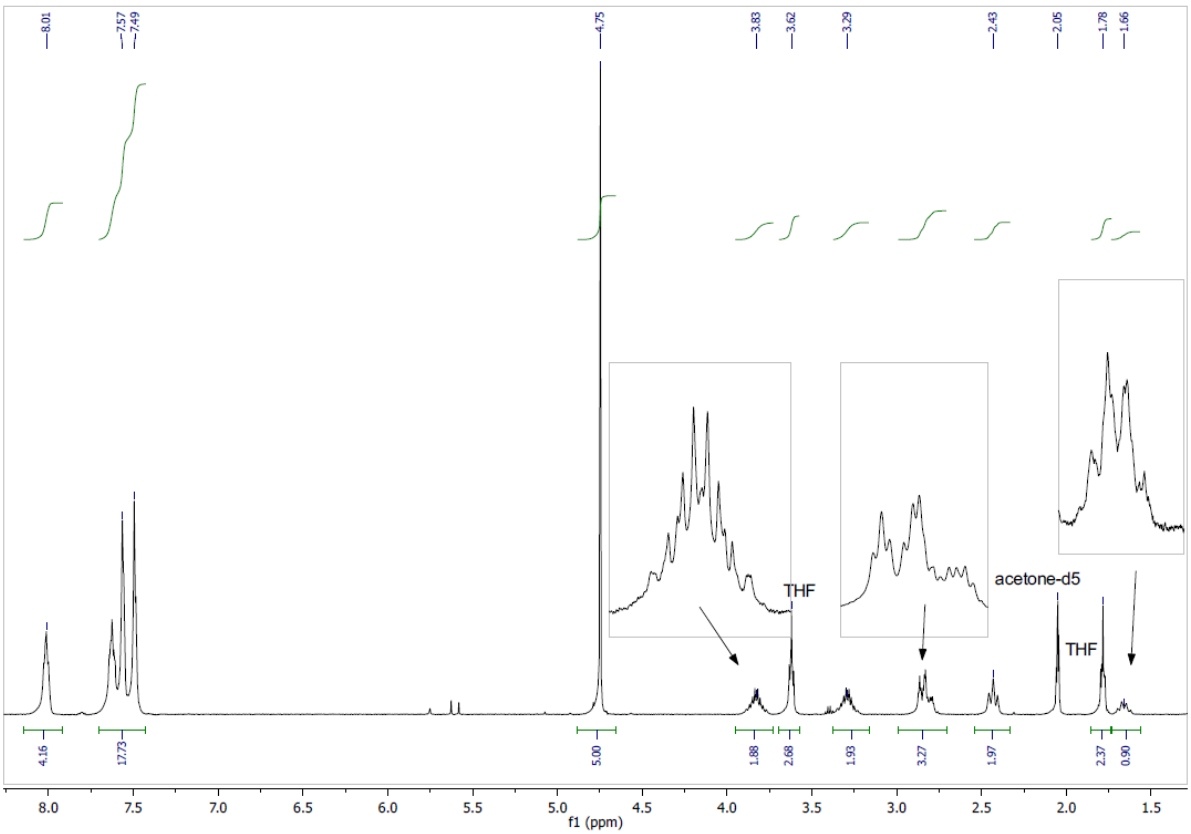

Figure S11. ^{1}H NMR spectrum of [**1a**]BF_4 in acetone-d_6.

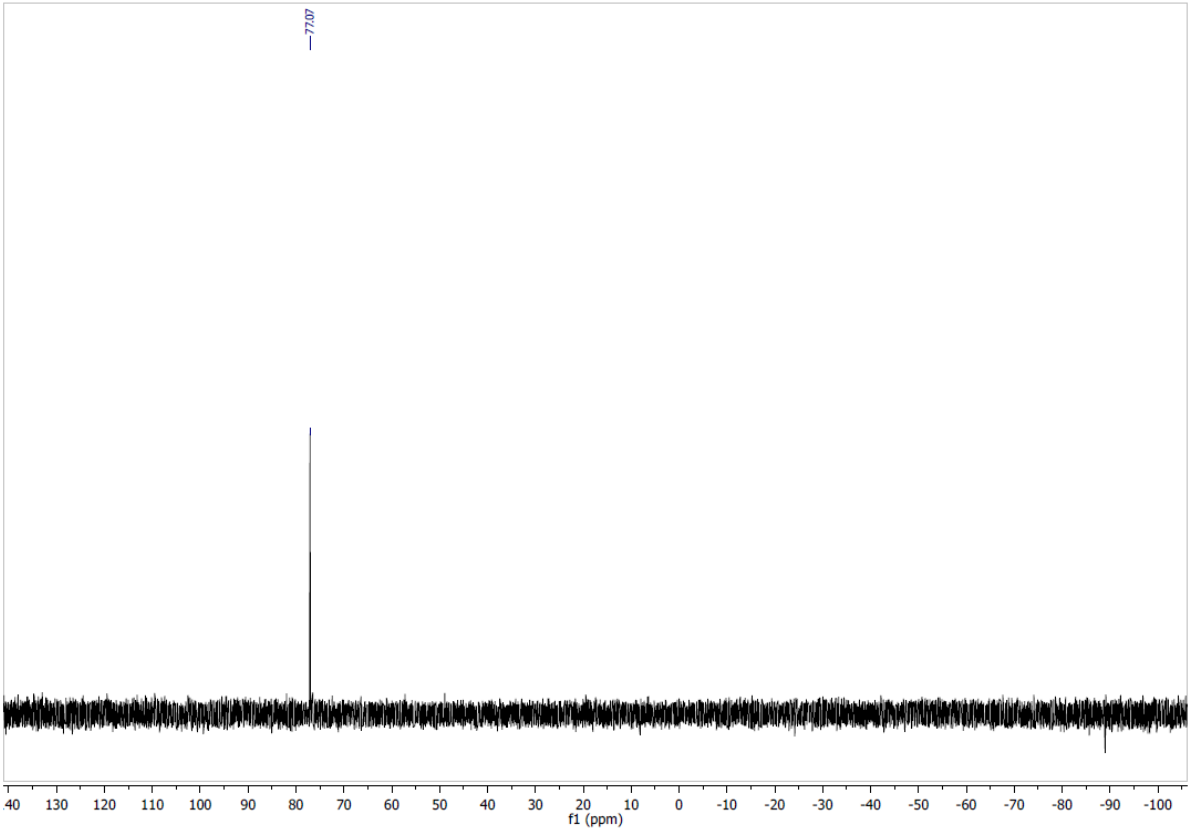

Figure S12. ^{31}P{H} NMR spectrum of [**1a**]BF_4 in acetone-d_6.

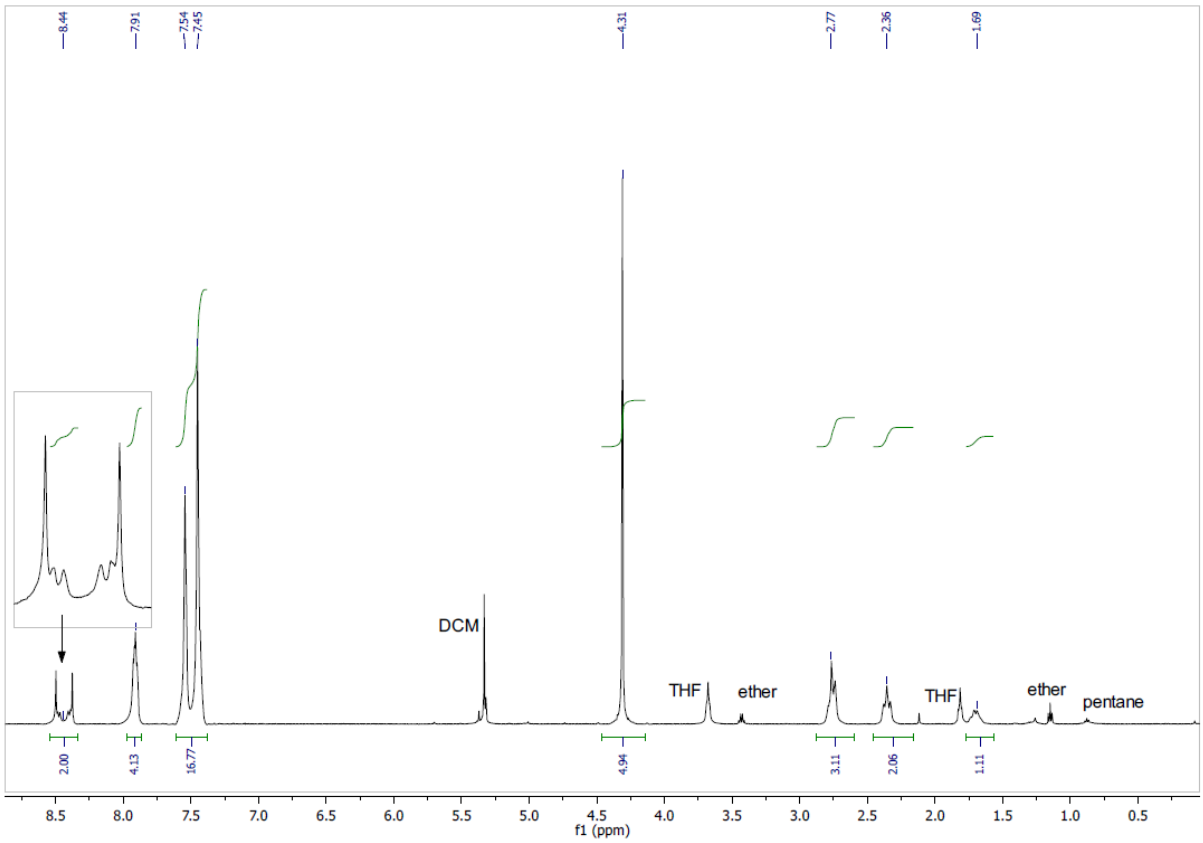

Figure S13. ^{1}H NMR spectrum of [**1b**]BF_4 in CD_2Cl_2.

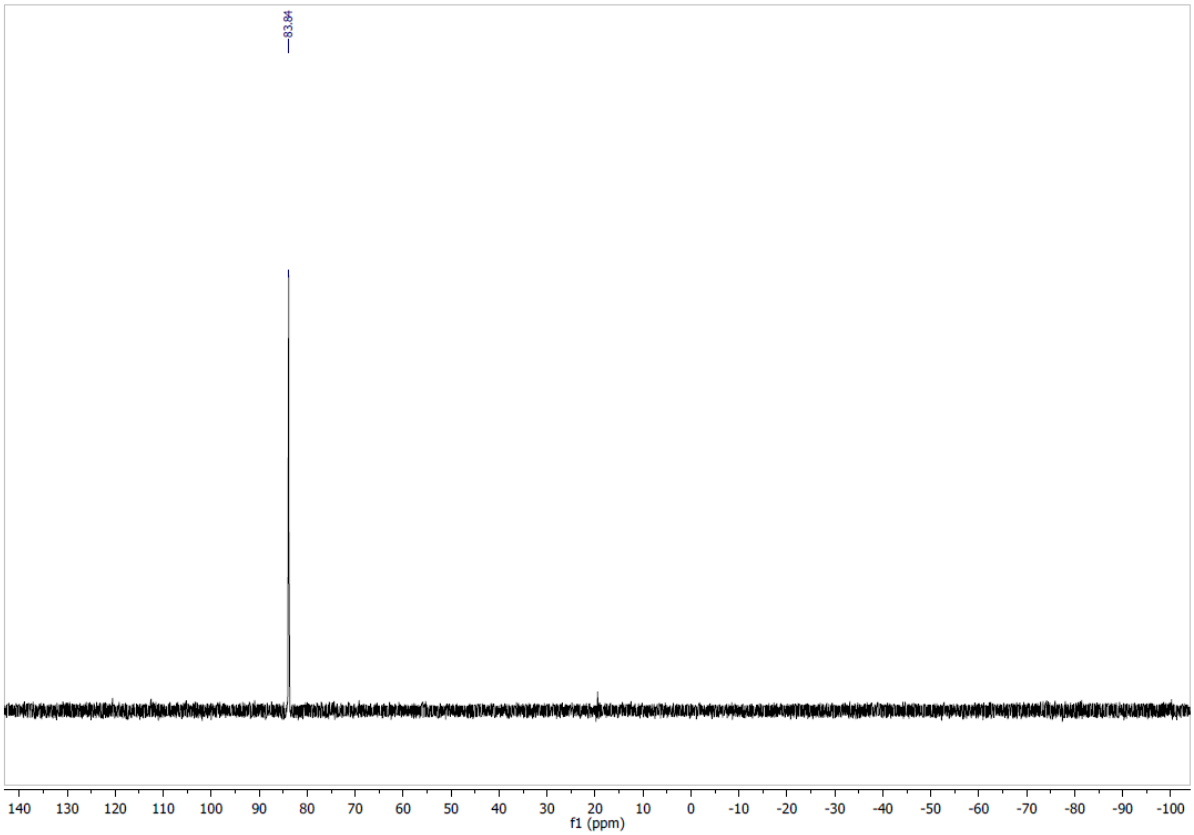

Figure S14. ^{31}P{H} NMR spectrum of [**1b**]BF_4 in CD_2Cl_2.

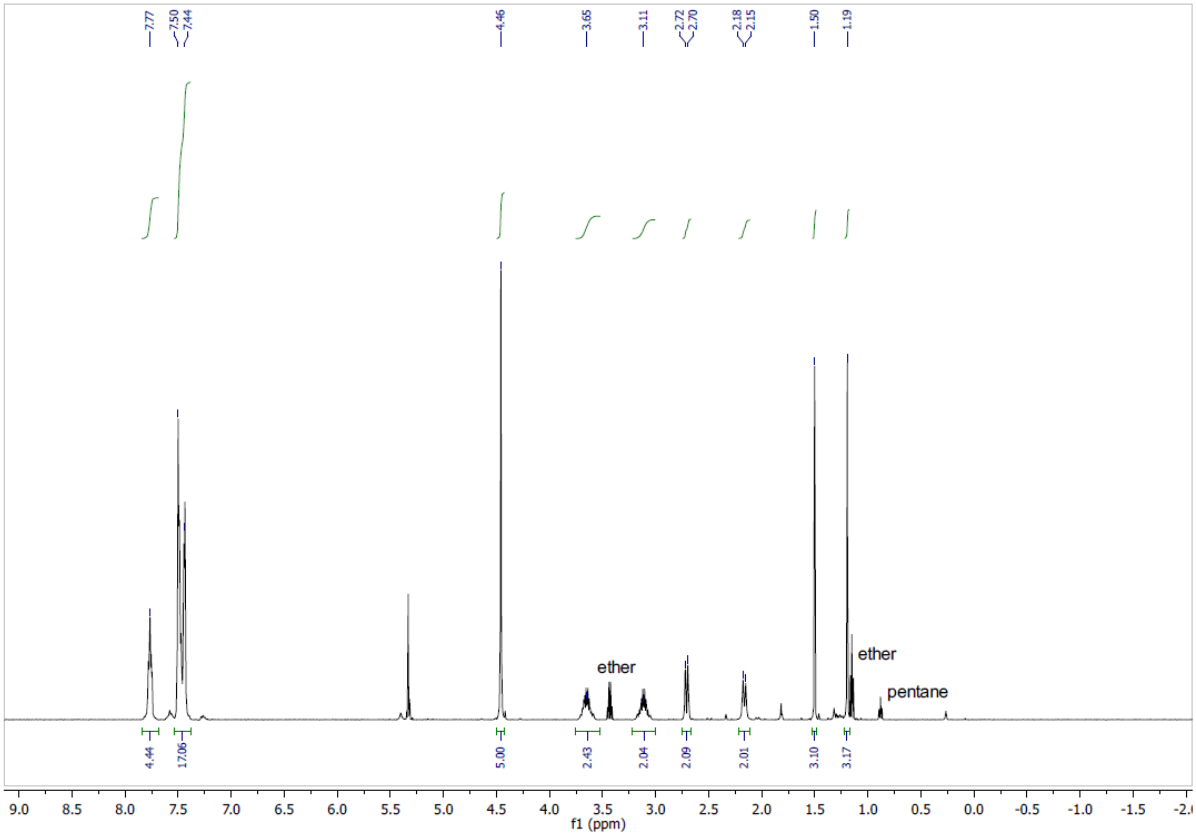

Figure S15. ^{1}H NMR spectrum of [**1c**]BF_4 in CD_2Cl_2.

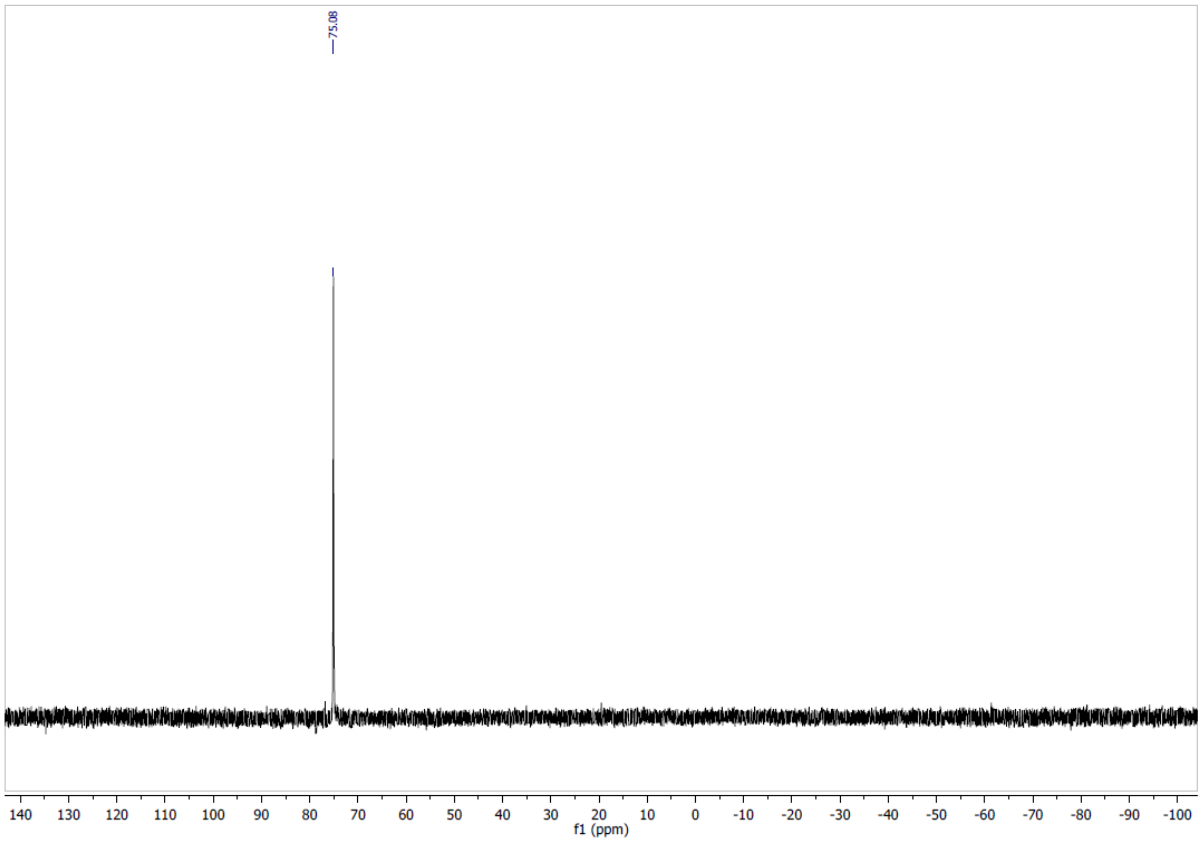

Figure S16. ^{31}P{H} NMR spectrum of [**1c**]BF_4 in CD_2Cl_2.

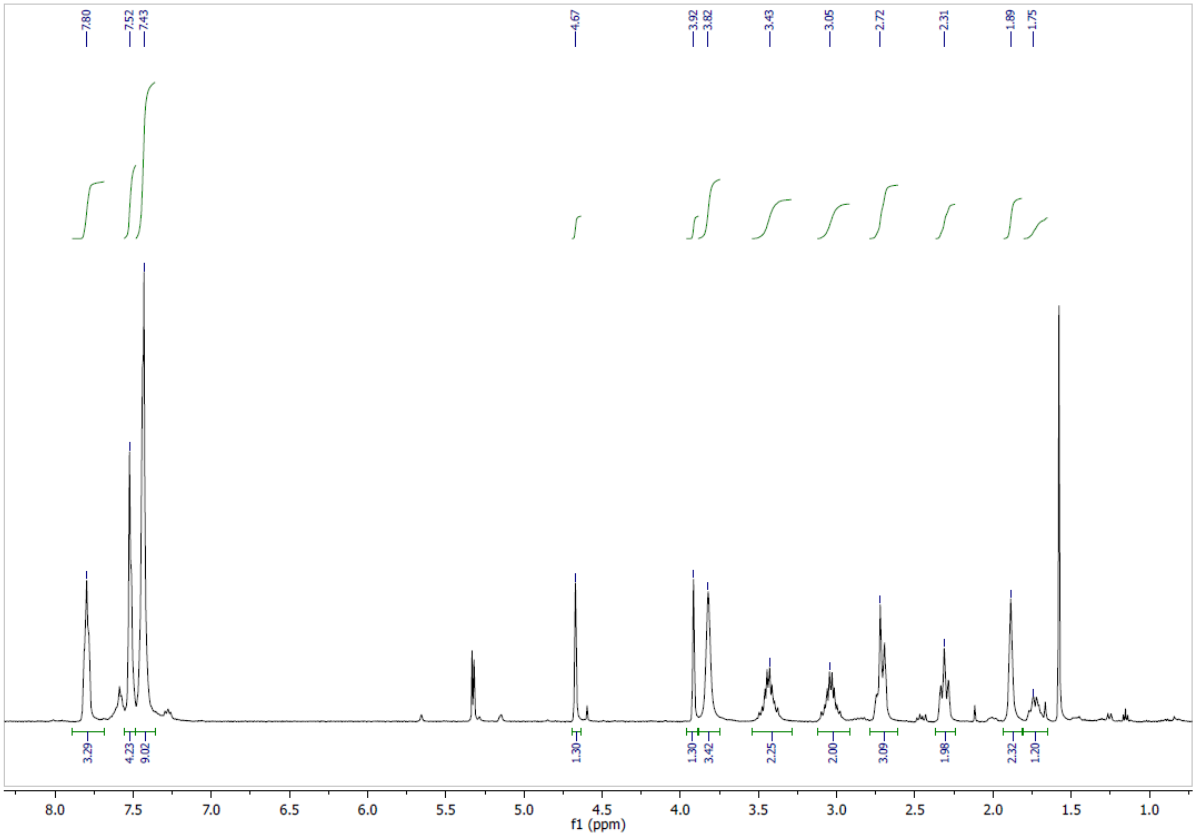

Figure S17. ^{1}H NMR spectrum of [**1d**]BF_4 in CD_2Cl_2.

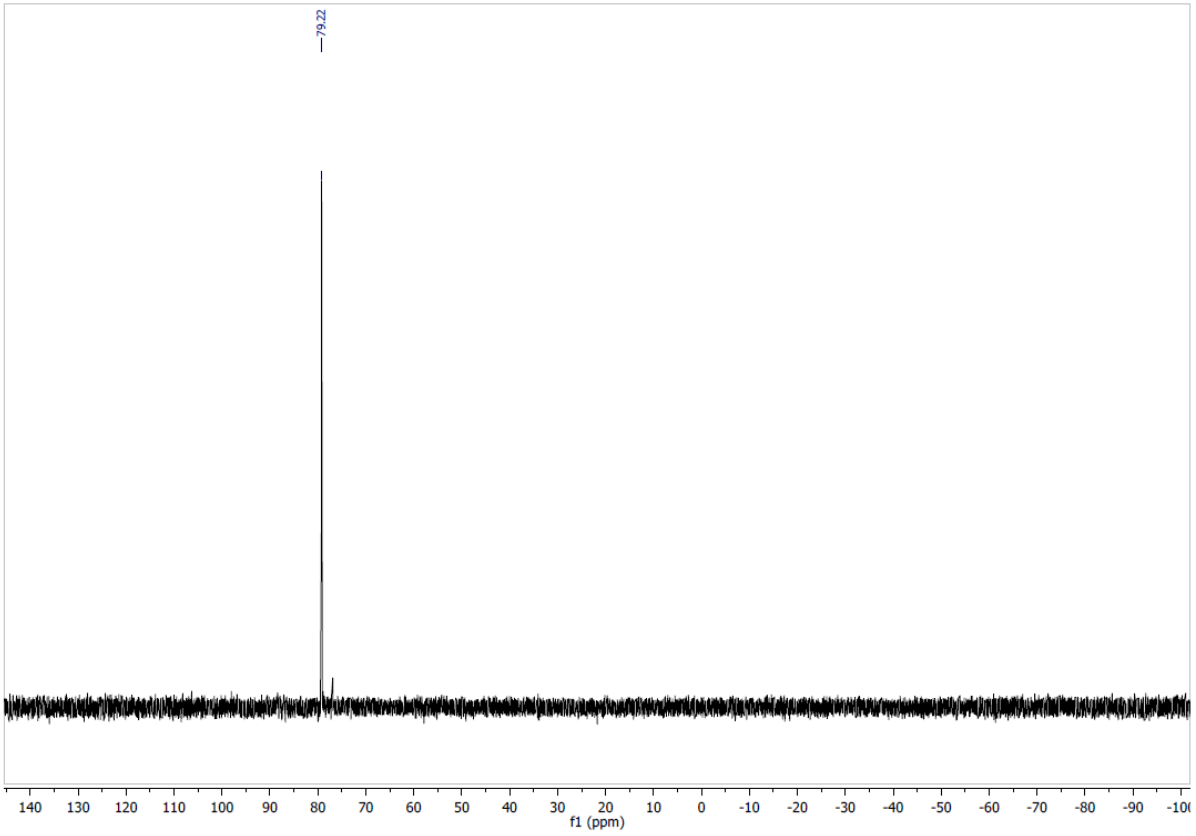

Figure S18. ^{31}P{H} NMR spectrum of [**1d**]BF_4 in CD_2Cl_2.

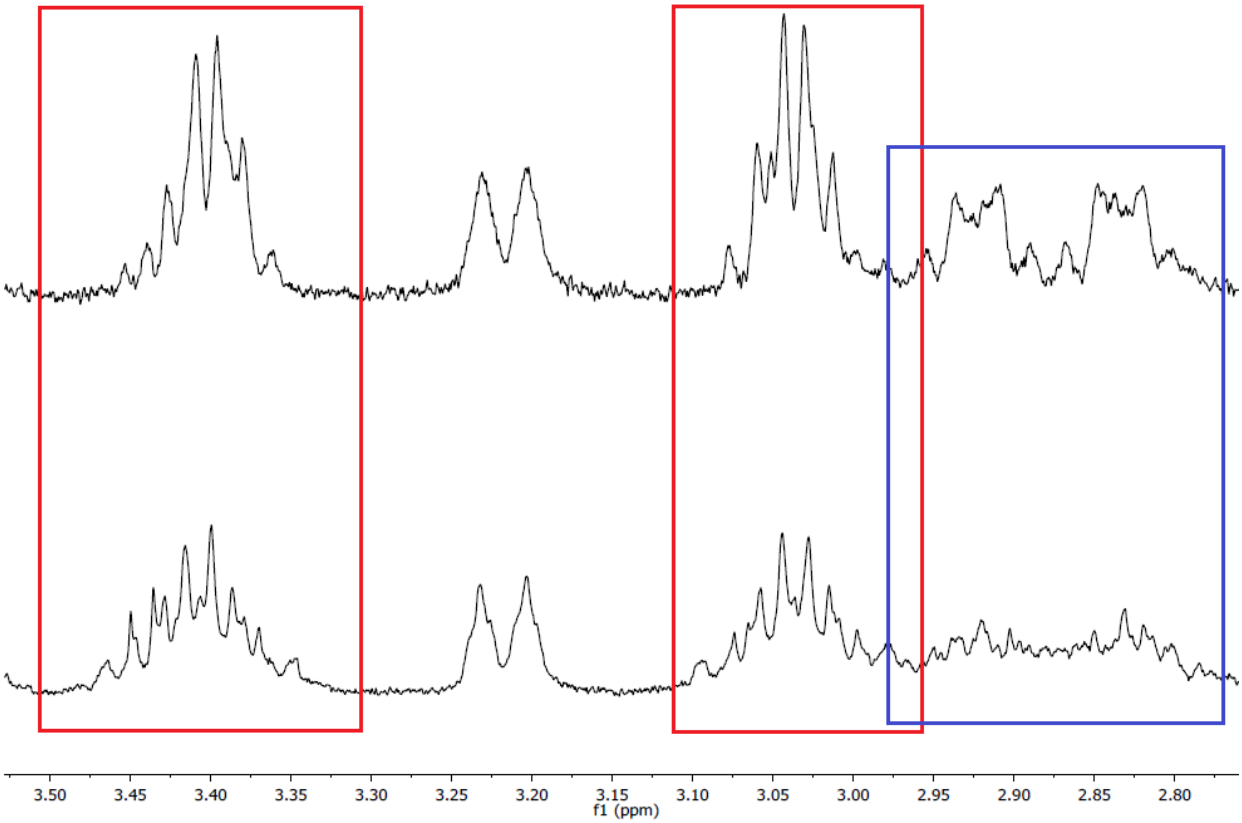

Figure S19. ^{1}H NMR spectrum of [**1a**]^{0} after treatment with excess acid, focusing on the ethylene protons of the diphosphine (bottom). ^{1}H{^{31}P}-NMR spectrum focusing on the ethylene protons of the diphosphine (top). The red box highlights the resonances assigned to the dibasal isomer and the blue box highlights the resonances assigned to the proposed apical-basal isomer.

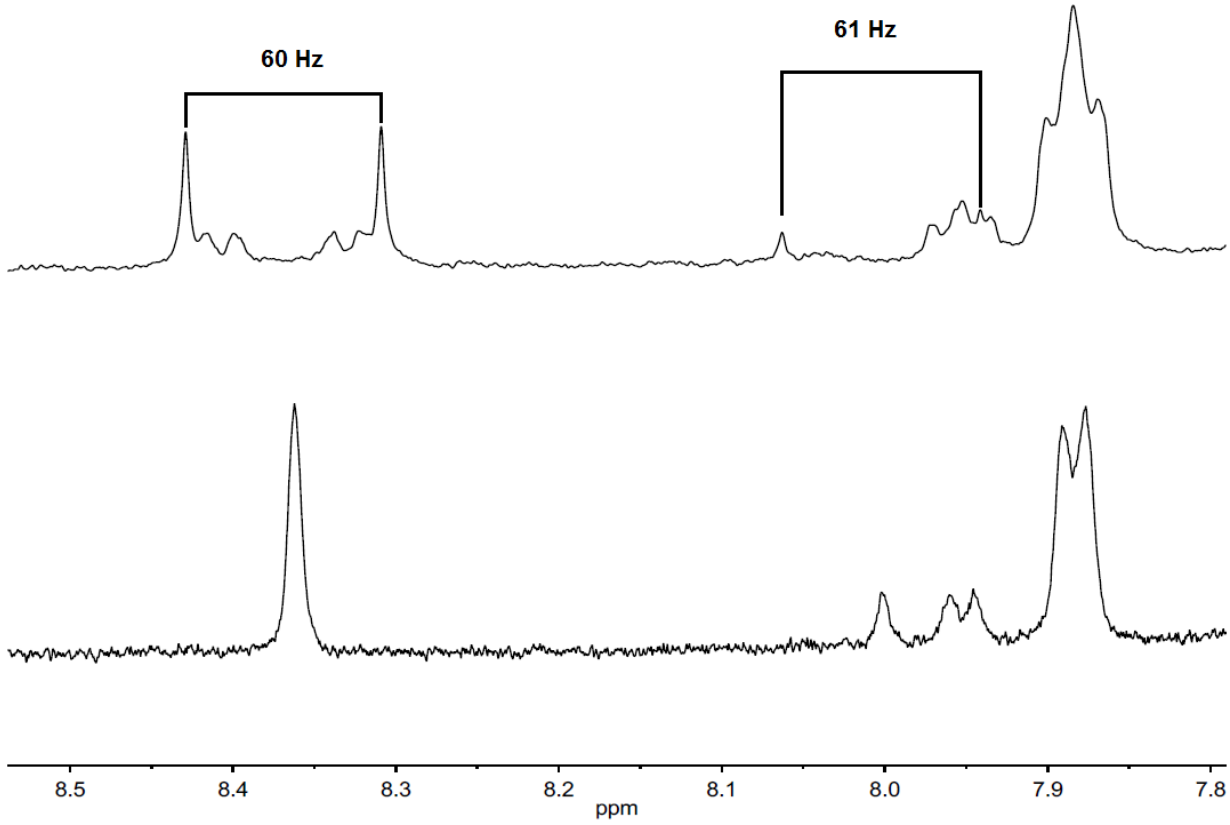

Figure S20. ^{1}H NMR Spectrum of [**1b**]^{0} after treatment with excess acid, highlighting the vinylidene protons of the diphosphine (top). The ^{1}H{^{31}P} NMR spectrum highlighting the vinylidene protons of the diphosphine (bottom). The dibasal isomer is assigned to the resonance at c.a. δ8.35 and the proposed apical-basal isomer is assigned to the resonance at c.a. δ8.00.

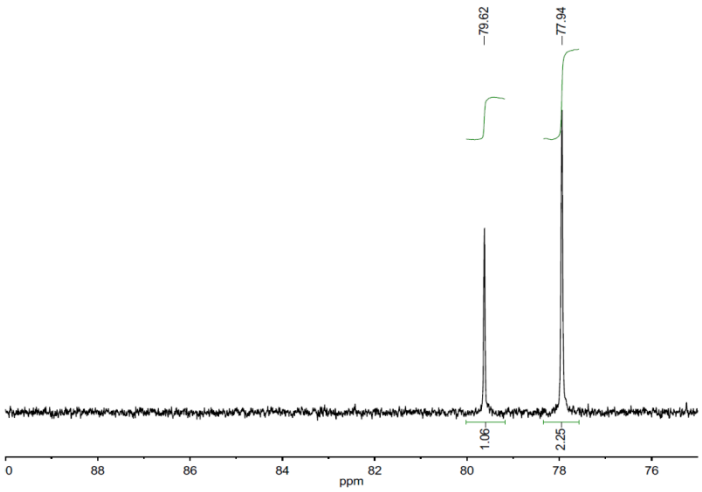

Figure S21. $^{31}P\{H\}$ NMR spectrum of [**1a**]^{0} after treatment with excess acid.

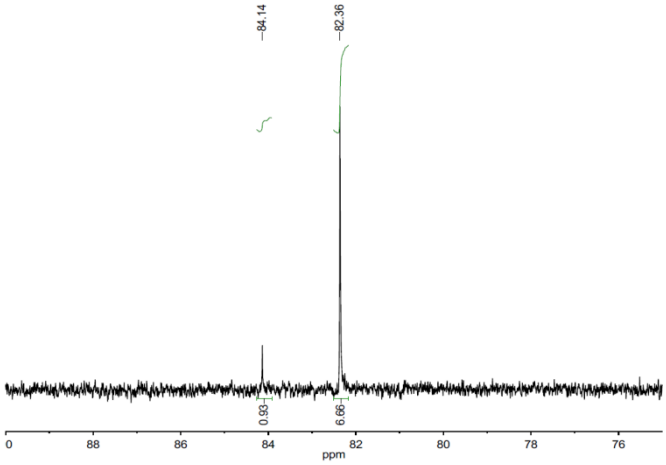

Figure S22. $^{31}P\{H\}$ NMR spectrum of [**1b**]^{0} after treatment with excess acid.

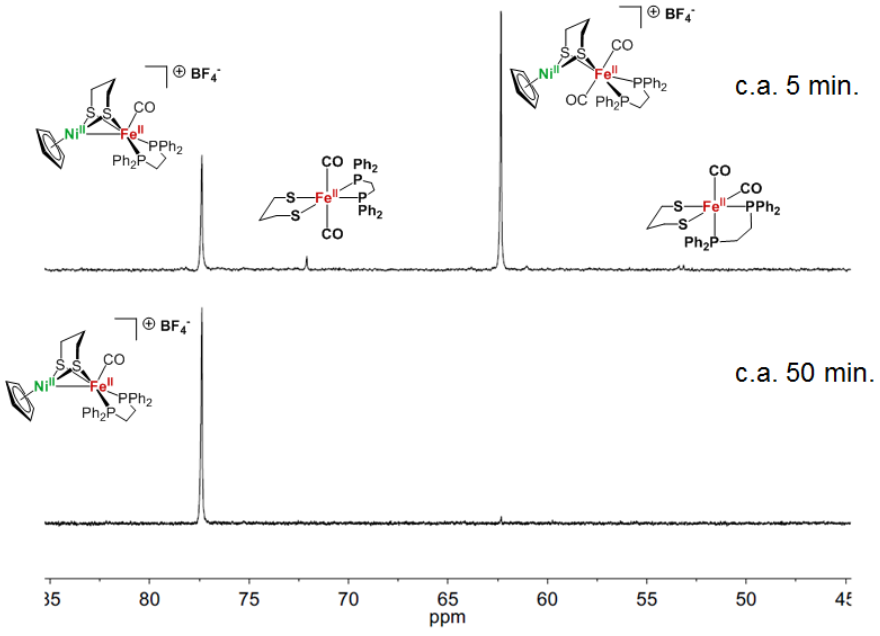

Figure S23. $^{31}P\{H\}$ NMR spectrum of the reaction $[Cp_3Ni_2]BF_4$ and $Fe(pdt)(dppe)(CO)_2$.

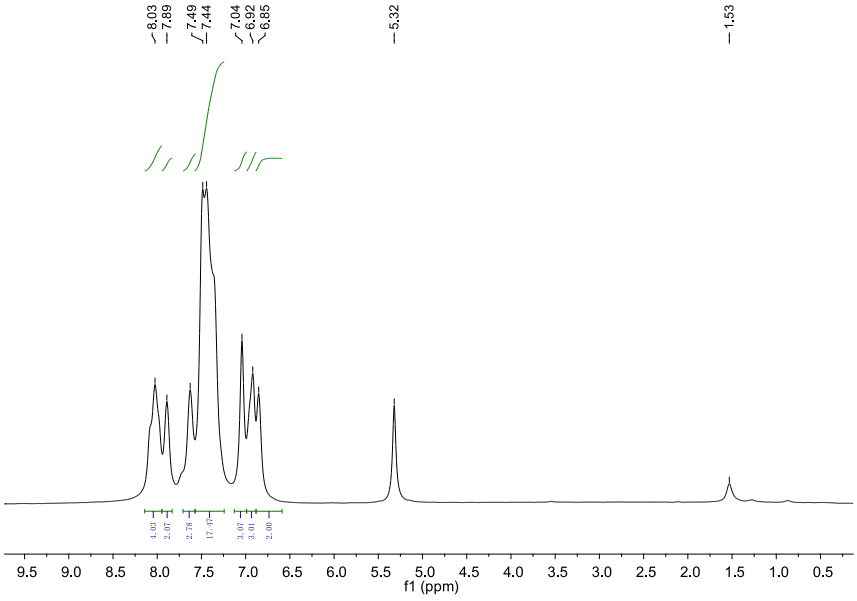

Figure S24. ^{1}H NMR spectrum of Fe(dppv)$(SPh)_2(CO)_2$.

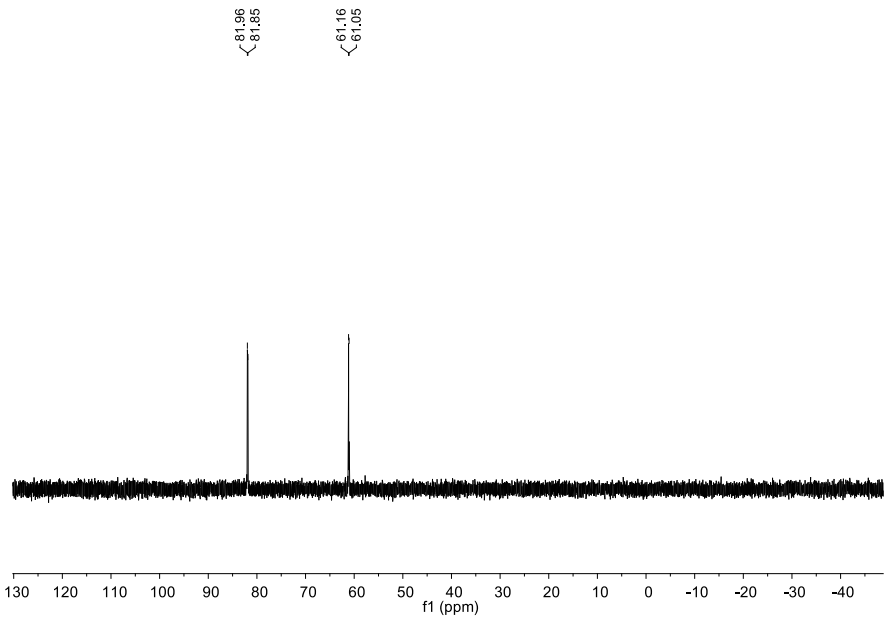

Figure S25. ^{31}P NMR spectrum of Fe(dppv)$(SPh)_2(CO)_2$.

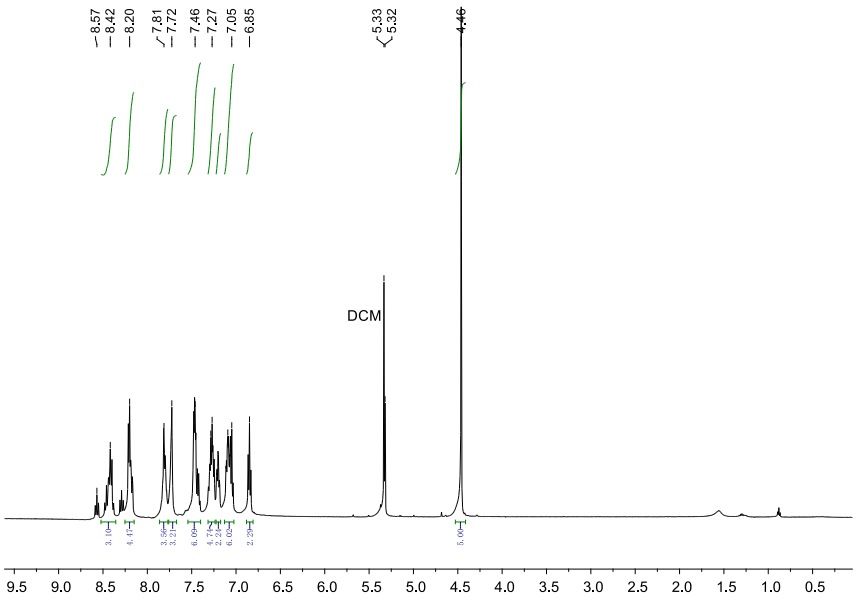

Figure S26. ^{1}H NMR spectrum of [**2b**CO]BF_4.

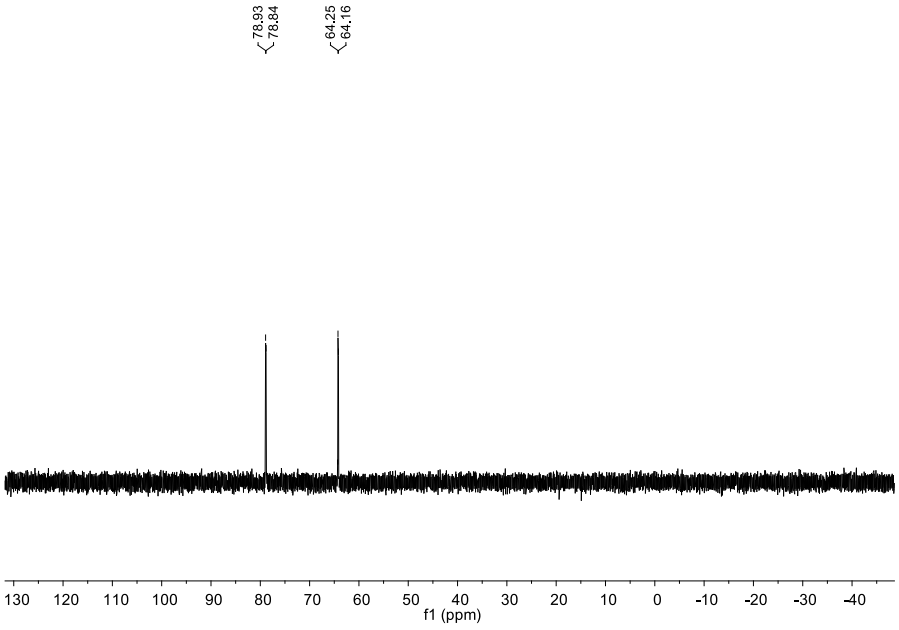

Figure S27. ^{31}P NMR spectrum of [**2b**CO]BF_4.

Cyclic Voltammograms

The following data were recorded according to the procedure outlined in the main text. All voltammograms were recorded in dichloromethane and began at ca. –0.5 V vs $Fc^{0/+}$ at 500 mVs^{-1}. $E_{½}$ and E_p values for each voltammogram are tabulated in Table 2 in the main text.

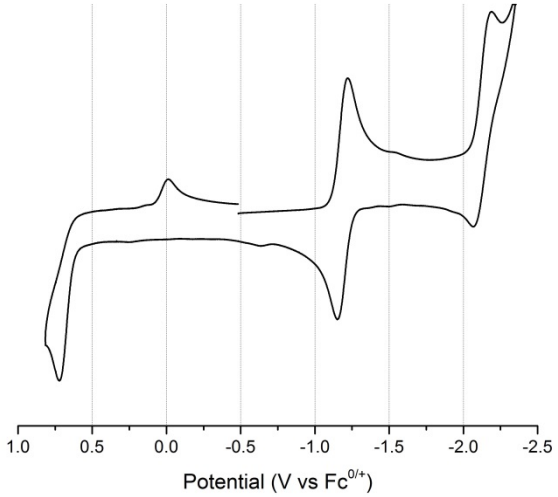

Figure S28. Voltammogram of [**1b**]BF_4.

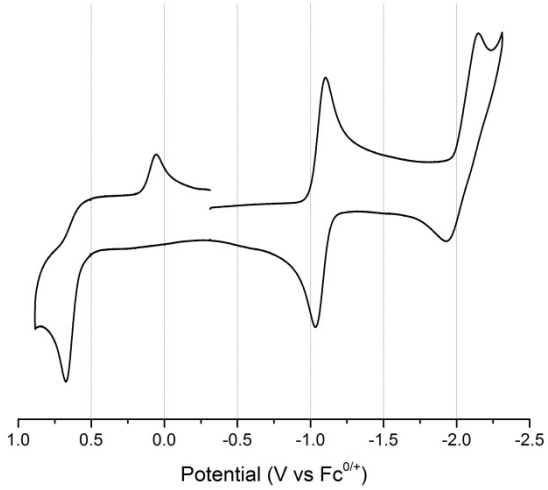

Figure S29. Voltammogram of [**1c**]BF_4.

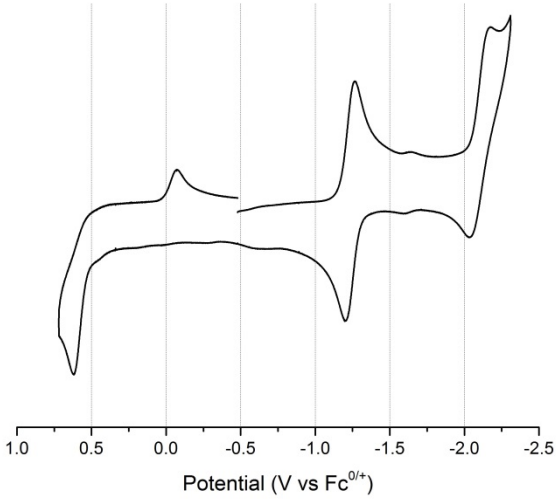

Figure S30. Voltammogram of [**1d**]BF_4.

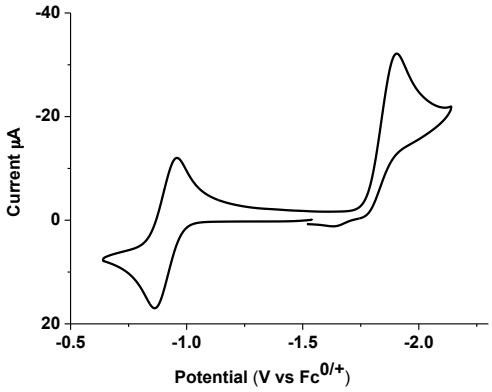

Figure S31. Cyclic voltammogram of [**2b**]BF_4 at 100 mV s^{-1}.

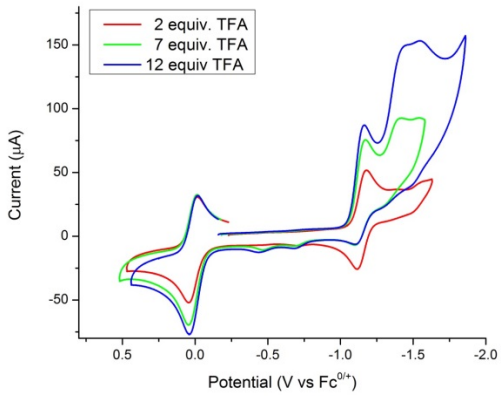

Figure S32. Cyclic voltammogram of [**1a**]BF_4 in CH_2Cl_2 at 500 mV s^{-1} with increasing amounts of trifluoroacetic acid showing a catalytic wave centered at the $[\mathbf{1a}]^{+/0}$ couple. The maximum i_c/i_p observed in the acid independent region is approximately 2, yielding a rate of 3.88 s^{-1}.[1] Glassy carbon is known to catalyze proton reduction of trifluoroacetic acid at −1.81 V under these conditions.[2]

IR Spectra

The following spectra were recorded using the equipment described in the main text. All spectra were recorded as solutions in dichloromethane.

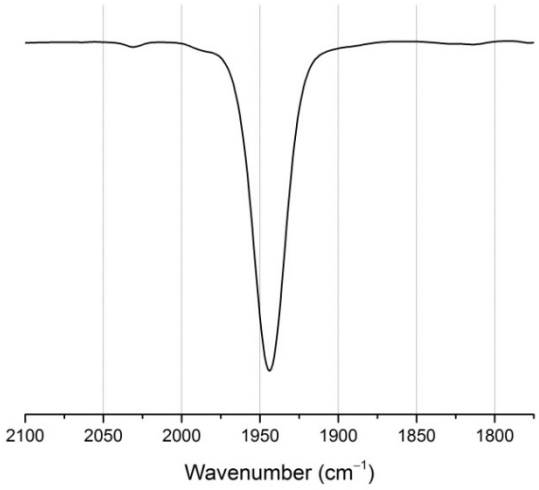

Figure S33. IR spectrum of [**1a**]BF_4.

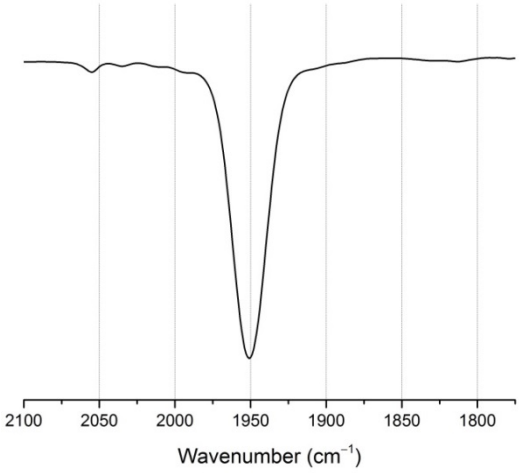

Figure S34. IR spectrum of [**1b**]BF_4

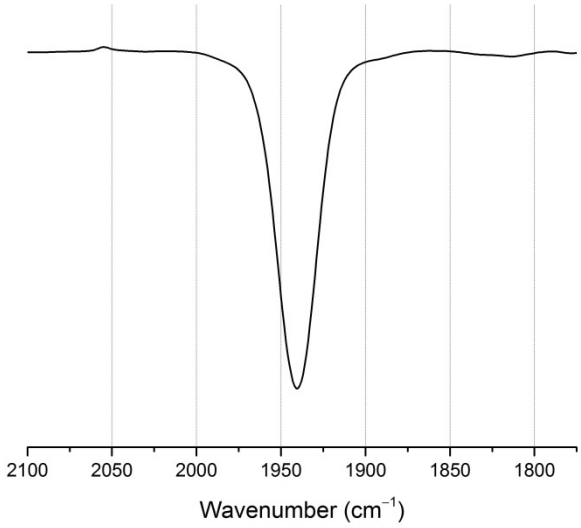

Figure S35. IR spectrum of [**1c**]BF_4

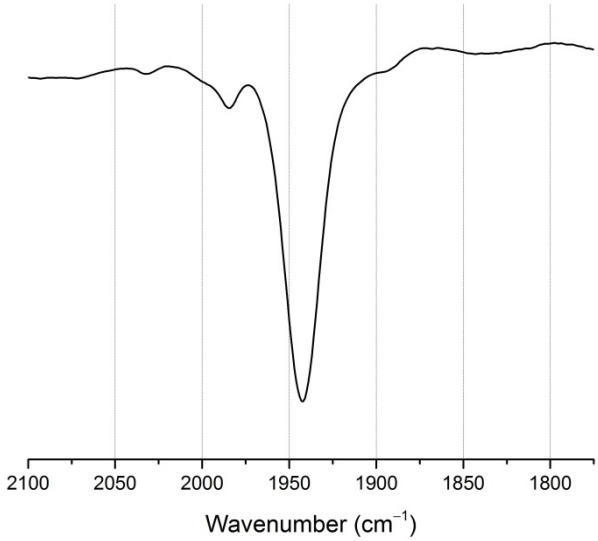

Figure S36. IR spectrum of [**1d**]BF_4

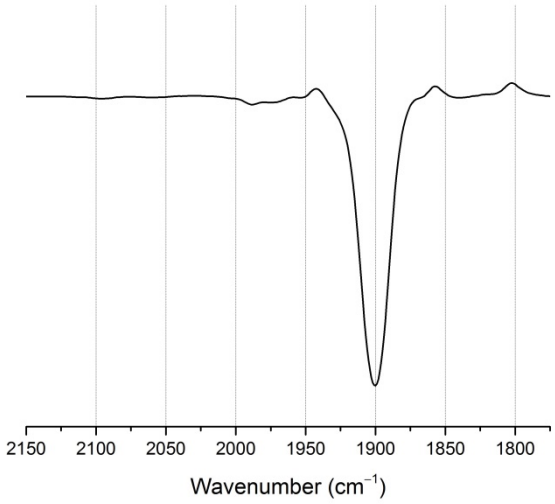

Figure S37. IR spectrum of [**1a**]^{0}.

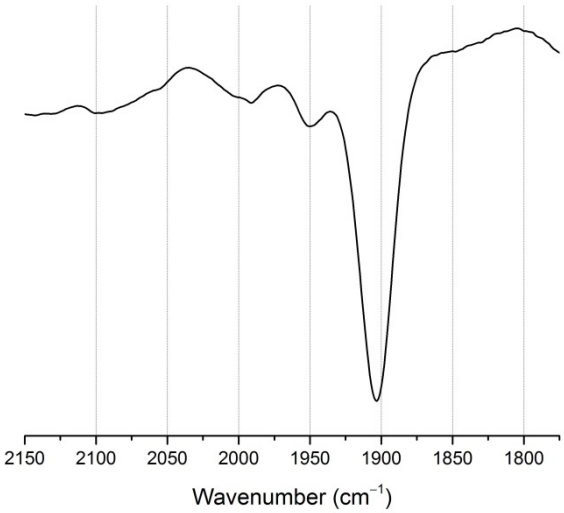

Figure S38. IR spectrum of [**1b**]^{0}.

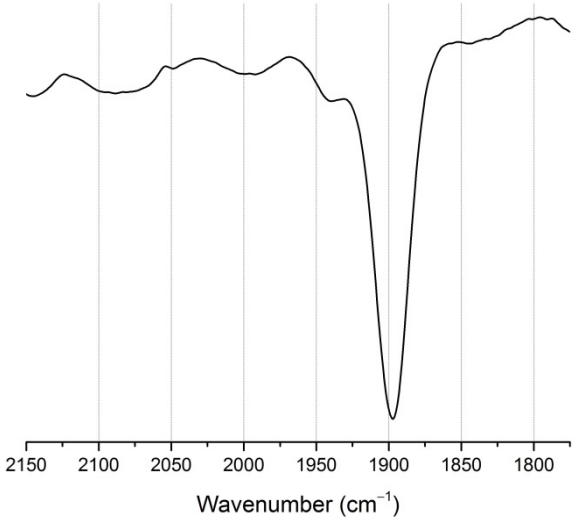

Figure S39. IR spectrum of [**1c**]^{0}.

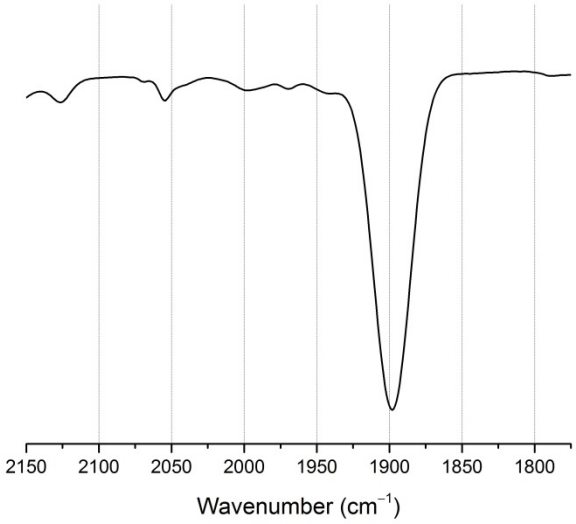

Figure S40. IR spectrum of [**1d**]^{0}.

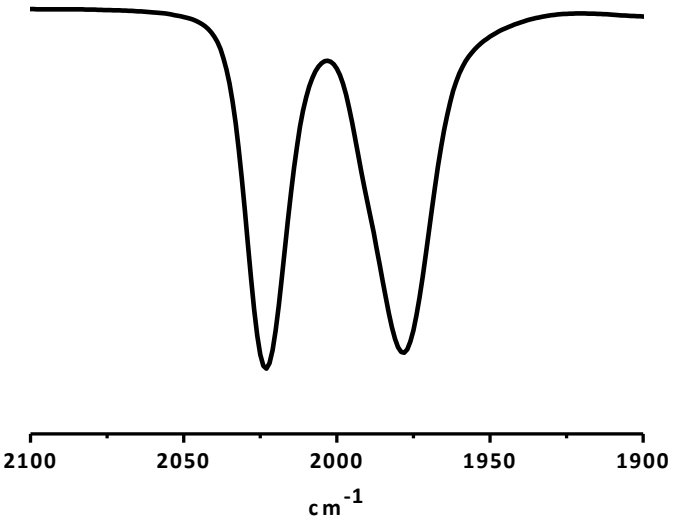

Figure S41. IR spectrum of $Fe(dppv)(SPh)_2(CO)_2$

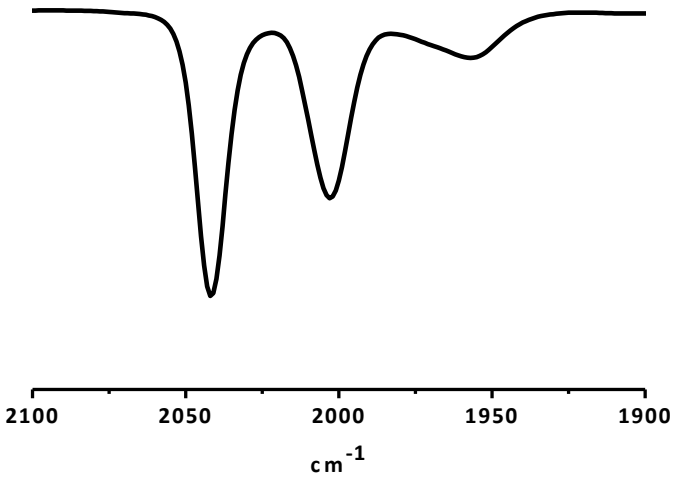

Figure S42. IR spectrum of [**2b**CO]BF_4.

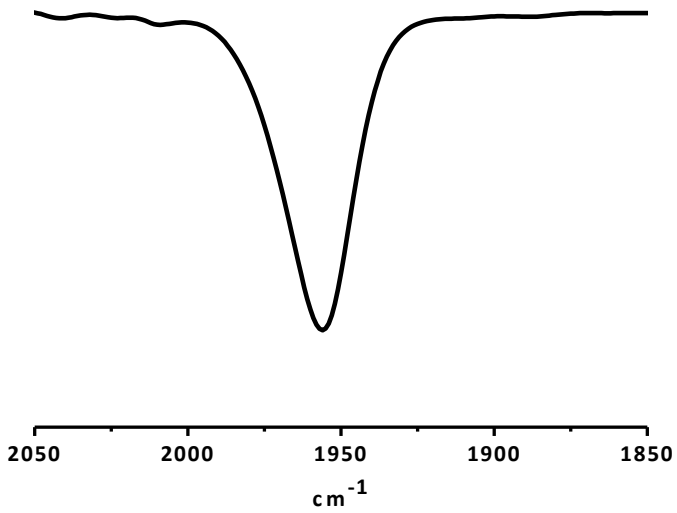

Figure S43. IR spectrum of [**2b**]BF_4.

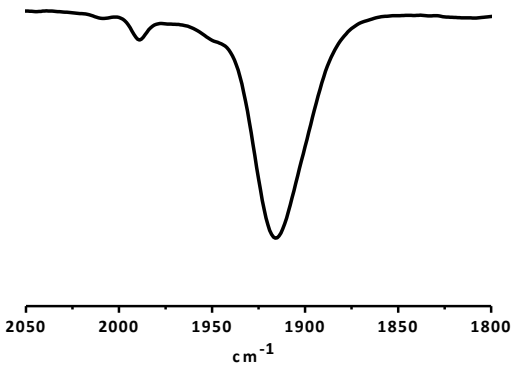

Figure S44. IR spectrum of [**2b**]^{0}.

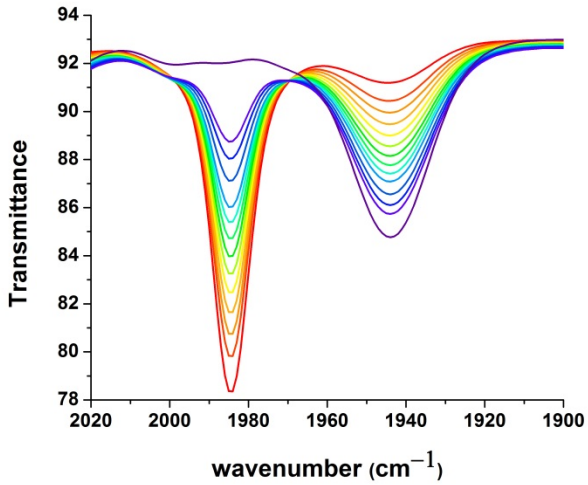

Figure S45. IR spectra from c.a. 5 minutes (red) to c.a. 50 minutes (purple) of the reaction of $[Cp_3Ni_2]BF_4$ and $Fe(pdt)(dppe)(CO)_2$. The product band ([**1a**]BF_4) occurs at 1943 cm^{-1}. Intervals for timepoint 1 to 10 are 1 minute, 11 to 13 2 minutes, and the final time point is after 50 minutes.

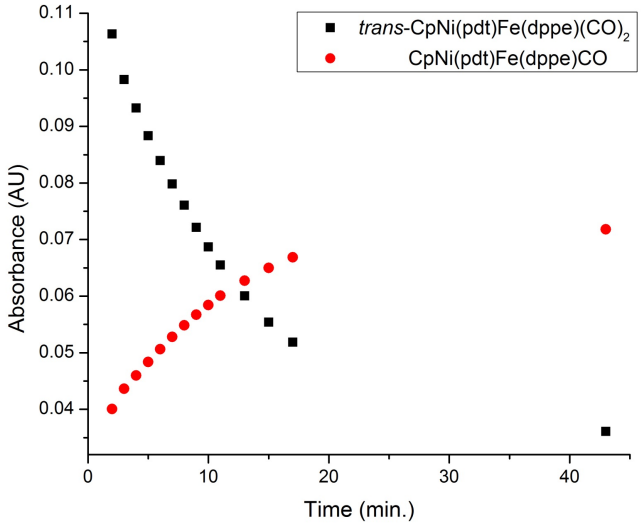

Figure S46. IR absorbance versus time plot of the CO bands observed during the reaction of $[Cp_3Ni_2]BF_4$ and $Fe(pdt)(dppe)(CO)_2$. The half-life of the *trans*-[CpNi(pdt)Fe(dppe)$(CO)_2$] product is ca. 480 sec., equating to an activation barrier of 21 kcal/mol under the experimental conditions.

CW X-Band EPR Spectra

EPR spectra were recorded according to the conditions described in the figure and using the instrumentation described in the main text.

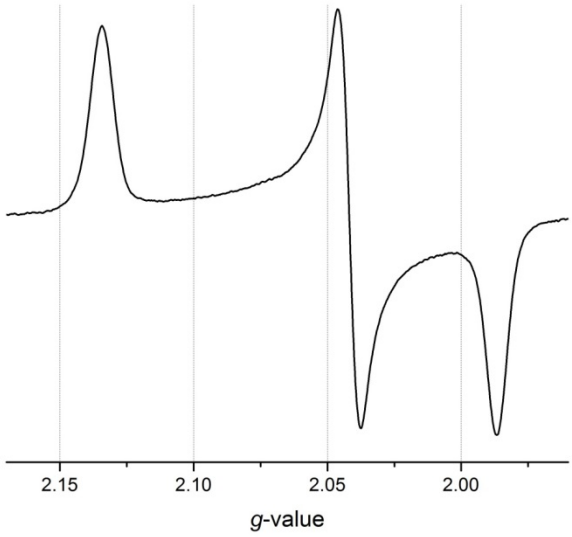

Figure S47. X-band CW-EPR Spectrum of **[1b]**^{0} as a ~1 mM frozen solution dichloromethane-toluene solution at 110K.

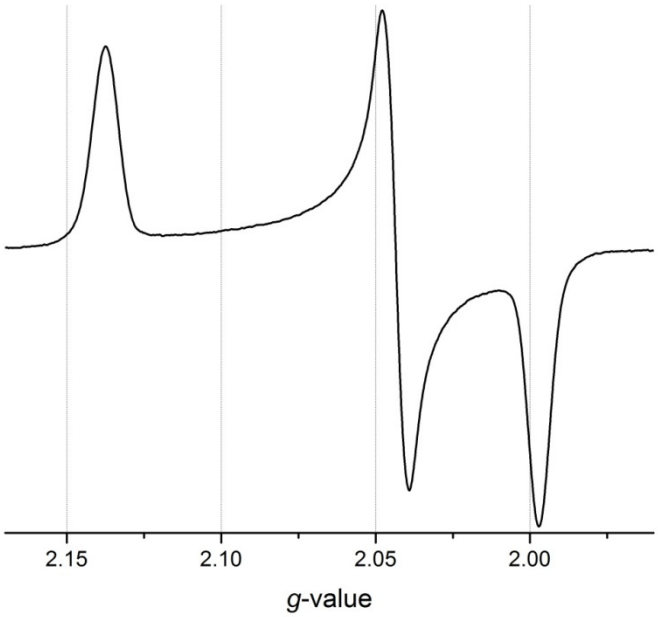

Figure S48. X-band CW-EPR spectrum of **[1c]**^{0} as a ~1 mM frozen solution dichloromethane-toluene solution at 110K.

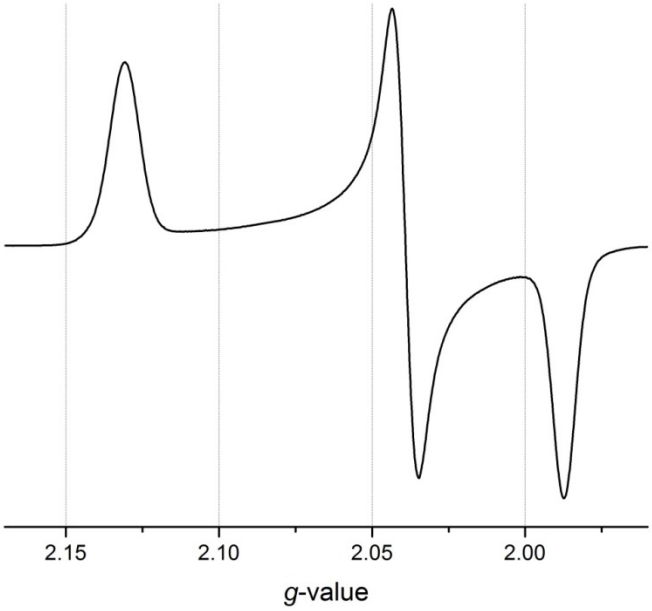

Figure S49. X-band CW-EPR spectrum of **[1d]**^{0} as a ~1 mM frozen solution dichloromethane-toluene solution at 110K.

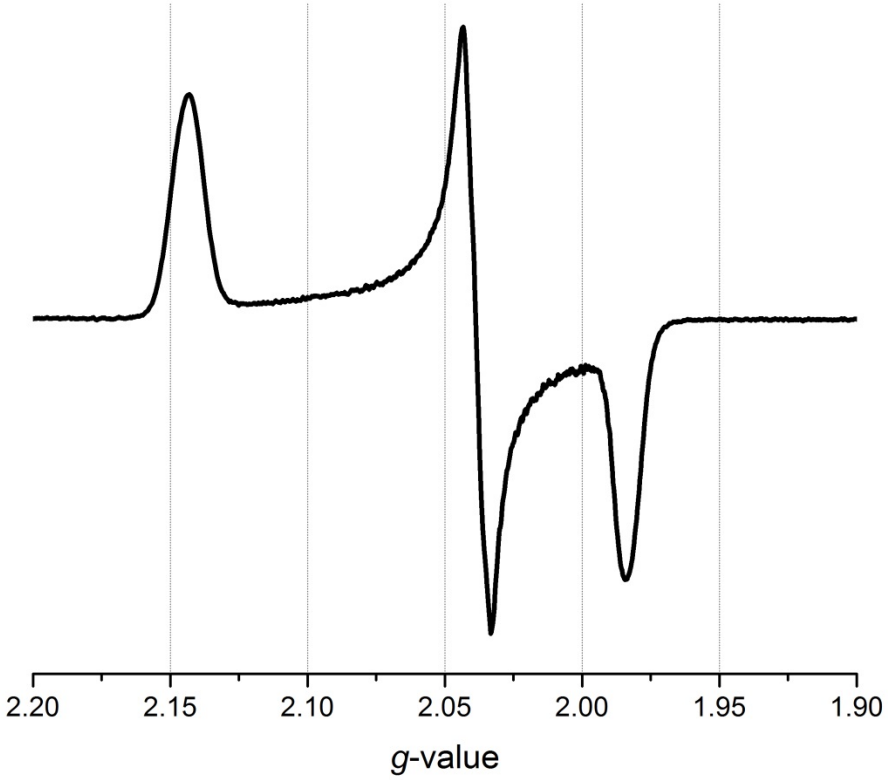

Figure S50. X-band CW-EPR spectrum of **[2b]**^{0} as a ~1 mM frozen solution dichloromethane-toluene solution at 110K.

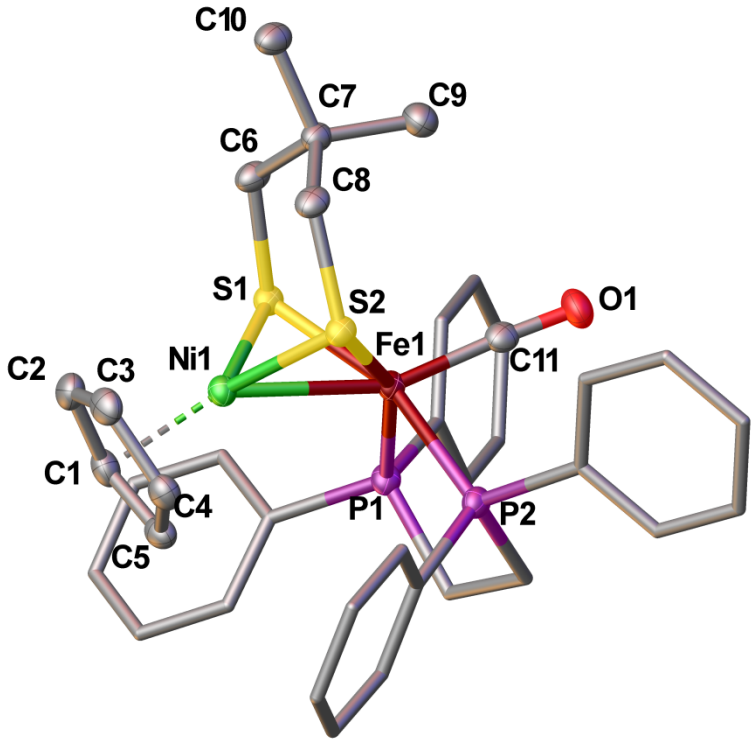

Figure S51. Solid-state structure of [**1c**]BF_4 shown at the 35% probability level with hydrogen atoms and counterion omitted for clarity. Selected bond distances (Å): Ni1-Fe1, 2.4825(9); Ni1-Cent, 1.723(5); Ni1-S_{avg}, 2.1762(9); Fe1-S_{avg}, 2.2359(9); Fe1-C11, 1.744(3); C11-O1, 1.160(3).

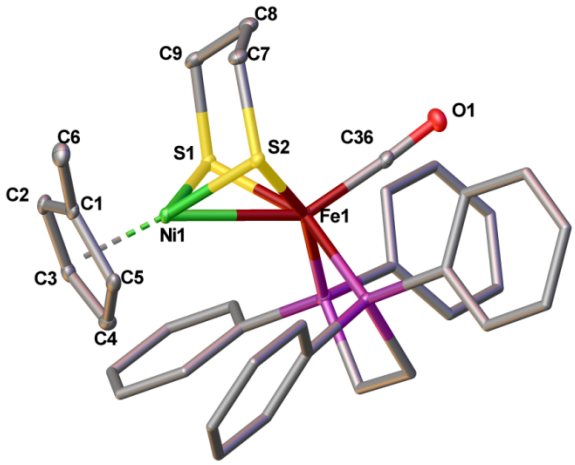

Figure S52. Solid-state structure of [**1d**]BF_4 shown at the 35% probability level with hydrogen atoms and counterion omitted for clarity. Selected bond distances (Å): Ni1-Fe1, 2.5214(6); Ni1-Cent, 1.724(4); Ni1-S_{avg}, 2.172(1); Fe1-S_{avg}, 2.219(1); Fe1-C36, 1.748(3); C36-O1, 1.151(4).

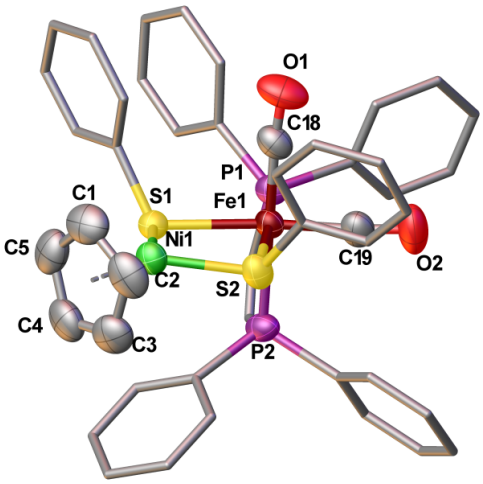

Figure S53. Solid-state structure of [**2b**CO]BF_4 shown at the 35% probability level with hydrogen atoms, counterion, and dichloromethane solvate omitted for clarity. The diffraction data were collected at room temperature (see CIF for more information). Selected bond distances (Å): Ni1-Fe1, 3.308(1); Ni1-cent, 1.754(9); Ni1-S_{avg}, 2.180(1); Fe1-S_{avg}, 2.353(1); Fe1-C18, 1.795(4); Fe1-C19, 1.796(4); C18-O1, 1.133(5); C19-O2, 1.129(5). Fe1-P1, 2.252(1); Fe1-P2, 2.291(1).

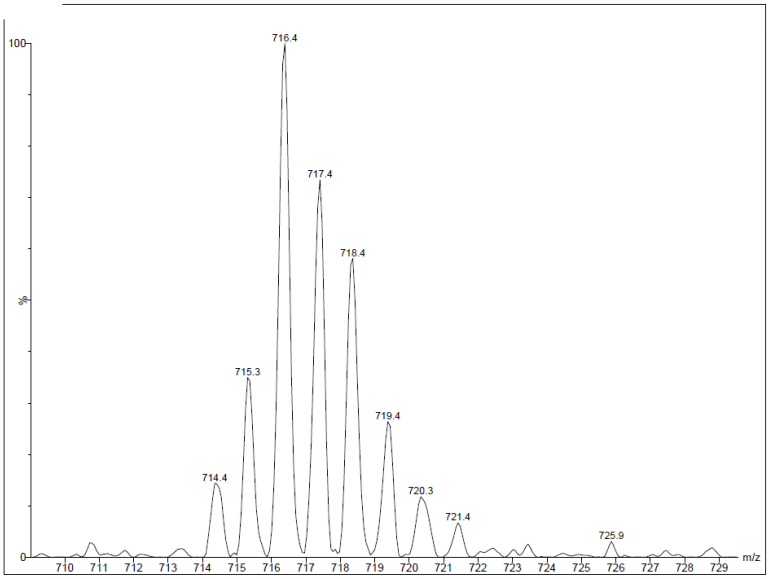

Figure S54. Mass spectrum of [(C_5D_5)Ni(pdt)Fe(CO)(dppe)]BF_4. ^2H incorperation ≥ 92%.

Table S1. Calculated Reduction Potentials[a] for [**1a-d**]$^{+/0}$ Using Different Functionals[b]

	$E_{1/2}$ ($Ni^{II}Fe^{II}/Ni^{I}Fe^{II}$)	$E°$[c] ($Ni^{II}Fe^{II}/Ni^{I}Fe^{II}$)					
	Expt	**BP86**	**BP86**[d]	**BP86-D2**	**B3P86**	**M06-L**	**ωB97X-D**
[**1a**]$^{+/0}$	–1.16	–1.16[e,f]	–1.16[e]	–1.16[e]	–1.16[e]	–1.16[e]	–1.16[e]
		–1.14[g]	–1.02	–1.02	–1.14	–1.10	–1.06
[**1b**]$^{+/0}$	–1.15	–1.12	–1.15	–1.11	–1.12	–1.14	–1.05
		–1.02	–1.09	–1.06	–1.05	–1.07	–0.94
[**1c**]$^{+/0}$	–1.09	–1.16	–1.11	–1.09	–1.15	–1.17	–1.05
		–1.09	–1.19	–1.08	–1.08	–1.08	–0.97
[**1d**]$^{+/0}$	–1.21	–1.23	–1.20	–1.26	–1.21	–1.23	–1.14
		–1.20	–1.21	–1.13	–1.15	–1.15	–1.03

[a]Units of V vs. $Fc^{0/+}$ in CH_2Cl_2. [b]Unless stated otherwise, the geometries were optimized in the gas phase using the SDD pseudopotential and associated basis set for Ni and Fe and the 6-31G* basis set for all other atoms, and the solvation free energies were calculated in CH_2Cl_2 using C-PCM. [c]For each complex, the first calculated potential corresponds to the species in which the pdt is oriented toward the Fe, and the second calculated potential corresponds to the species in which the pdt is oriented toward the Ni. [d]The geometries were optimized in CH_2Cl_2 solvent using C-PCM. [e]The [**1a**]$^{+/0}$ couple was used as the reference reaction so the experimental and calculated values agree by construction. [f]Reducing the Fe center has a calculated potential of $E°$ = –1.35 V vs. $Fc^{0/+}$ in CH_2Cl_2. [g]Reducing the Fe center has a calculated potential of $E°$ = –1.31 V vs. $Fc^{0/+}$ in CH_2Cl_2.

Table S2. Experimental and Calculated Reduction Potentials[a] for [**1a-d**]$^{+/0}$ Using BP86 Functional Assuming the pdt Ring Flips Concurrently with Reduction

	$E_{p,ox}$ ($Ni^{II}Fe^{II}/Ni^{III}Fe^{II}$)	Calc[b,c,d]	$E_{1/2}$ ($Ni^{II}Fe^{II}/Ni^{I}Fe^{II}$)	Calc[b,c,d]	$E_{p,red}$[e] ($Ni^{I}Fe^{II}/Ni^{I}Fe^{I}$)	Calc[b,c,d,e]
[**1a**]$^{+}$	0.70	0.70	–1.16	–1.16	–2.15	–2.15
[**1b**]$^{+}$	0.72	0.74	–1.15	–1.08	–2.18	–2.08
[**1c**]$^{+}$	0.67	0.67	–1.09	–1.23	–2.15	–2.15
[**1d**]$^{+}$	0.62	0.66	–1.21	–1.21	–2.17	–2.24

[a]Units of V vs. $Fc^{0/+}$ in CH_2Cl_2. [b]All geometries were optimized in the gas phase using the SDD pseudopotential and associated basis set for Ni and Fe and the 6-31G* basis set for all other atoms. Solvation free energies were calculated in CH_2Cl_2 using C-PCM. [c]For the calculated potentials, the pdt ring is oriented toward the Fe center before reduction (oxidation) and toward the Ni center after reduction (oxidation). [d]The [**1a**]$^{+/0}$ couple was used as the reference reaction so the experimental and calculated values agree by construction. [e]This wave is assigned to the $Ni^{I}Fe^{II}/Ni^{I}Fe^{I}$ couple, where the $Ni^{I}Fe^{I}$ state is an open-shell singlet.

Table S3. Calculated $\Delta\tilde{\nu}_{CO}$[a] for [**1a-d**]$^{+/0}$ Using Different Functionals[b]

	Expt	Calc[c]					
		BP86	**BP86**[d]	**BP86-D2**	**B3P86**	**M06-L**	**ωB97X-D**
[**1a**]$^{+/0}$	–42	–47/–45	–33/–31	–39/–32	–50/–63	–39/–36	–71/–68
[**1b**]$^{+/0}$	–48	–42/–43	–33/–32	–40/–36	–51/–52	–40/–39	–77/–81
[**1c**]$^{+/0}$	–43	–39/–37	–34/–34	–38/–35	–51/–50	–43/–41	–77/–73
[**1d**]$^{+/0}$	–45	–47/–45	–33/–32	–40/–24	–46/–47	–42/–41	–80/–68

[a]Units of cm^{-1}. [b]Unless stated otherwise, the geometries were optimized in the gas phase using the SDD pseudopotential and associated basis set for Ni and Fe and the 6-31G* basis set for all other atoms. [c]For each complex, the first calculated frequency corresponds to the species in which the pdt is oriented toward the Fe center, and the second calculated frequency corresponds to the species in which the pdt is oriented toward the Ni center. [d]The geometries were optimized in CH_2Cl_2 solvent using C-PCM.

Table S4. Selected Bond Lengths (Å) and Angles (°) for [**1a**]$^{+}$ Using Different Functionals[a]

	Expt	Calc					
		BP86	**BP86**[b]	**BP86-D2**	**B3P86**	**M06-L**	**ωB97X-D**
Ni–Fe	2.51	2.57	2.55	2.51	2.57	2.53	2.51
Ni–$Cp_{centroid}$	1.72	1.75	1.74	1.72	1.74	1.71	1.72
Ni–S[c]	2.16	2.19	2.19	2.18	2.18	2.20	2.16
Fe–S[c]	2.22	2.26	2.26	2.25	2.25	2.27	2.22
Fe–P[c]	2.23	2.27	2.26	2.22	2.27	2.25	2.23
Fe–C	1.76	1.73	1.72	1.73	1.73	1.73	1.76
S_1–Ni–S_2	90.3	89.8	89.8	91.5	86.6	90.8	90.3
S_1–Fe–S_2	87.4	86.3	86.5	87.9	90.0	87.1	87.4
P_1–Fe–P_2	86.7	87.2	86.8	86.9	87.0	86.3	86.7
S_1–Ni–S_2/ S_1–Fe–S_2[d]	106.8	106.7	105.6	105.8	107.6	104.5	106.8

[a]Unless stated otherwise, the geometries were optimized in the gas phase using the SDD pseudopotential and associated basis set for Ni and Fe and the 6-31G* basis set for all other atoms. [b]The geometries were optimized in CH_2Cl_2 solvent using C-PCM. [c]Bond distance represents an average value. [d]Defined as the dihedral angle between the planes generated by the groups of three atoms listed.

Table S5. Selected Bond Lengths (Å) and Angles (°) for [**1a**]^0 Using Different Functionals[a]

	Expt	Calc					
		BP86	**BP86**[b]	**BP86-D2**	**B3P86**	**M06-L**	**ωB97X-D**
Ni–Fe	2.46	2.53	2.50	2.41	2.58	2.45	2.50
Ni–$Cp_{centroid}$	1.80	1.83	1.83	1.78	1.87	1.79	1.84
Ni–S[c]	2.28	2.29	2.30	2.28	2.30	2.33	2.33
Fe–S[c]	2.25	2.28	2.27	2.28	2.29	2.30	2.32
Fe–P[c]	2.24	2.25	2.26	2.21	2.24	2.23	2.27
Fe–C	1.75	1.73	1.72	1.73	1.75	1.74	1.78
S_1–Ni–S_2	88.2	88.5	87.5	89.7	88.2	87.9	88.2
S_1–Fe–S_2	88.9	89.0	88.8	89.8	88.9	89.5	88.9
P_1–Fe–P_2	87.0	86.3	86.1	86.5	87.0	86.4	86.2
S_1–Ni–S_2/ S_1–Fe–S_2[d]	98.2	101.5	99.1	96.3	103.5	95.4	97.3

[a]Unless stated otherwise, the geometries were optimized in the gas phase using the SDD pseudopotential and associated basis set for Ni and Fe and the 6-31G* basis set for all other atoms. [b]The geometries were optimized in CH_2Cl_2 solvent using C-PCM. [c]Bond distance represents an average value. [d]Defined as the dihedral angle between the planes generated by the groups of three atoms listed.

Table S6. Selected Bond Lengths (Å) and Angles (°) for [**1c**]$^+$ Using Different Functionals[a]

	Expt	Calc					
		BP86	**BP86**[b]	**BP86-D2**	**B3P86**	**M06-L**	**ωB97X-D**
Ni–Fe	2.48	2.54	2.52	2.49	2.53	2.49	2.53
Ni–$Cp'_{centroid}$	1.72	1.75	1.74	1.72	1.74	1.71	1.73
Ni–S[c]	2.18	2.20	2.20	2.19	2.18	2.21	2.20
Fe–S[c]	2.24	2.40	2.26	2.25	2.25	2.28	2.26
Fe–P[c]	2.23	2.27	2.27	2.22	2.27	2.24	2.28
Fe–C	1.74	1.73	1.72	1.73	1.73	1.73	1.76
S_1–Ni–S_2	88.1	88.1	88.1	90.0	88.6	89.4	89.3
S_1–Fe–S_2	85.2	85.1	85.1	86.5	85.4	86.0	86.2
P_1–Fe–P_2	87.5	86.7	86.7	86.8	86.8	86.5	85.9
S_1–Ni–S_2/ S_1–Fe–S_2[d]	101.3	101.9	101.9	102.7	103.7	100.9	103.9

[a]Unless stated otherwise, the geometries were optimized in the gas phase using the SDD pseudopotential and associated basis set for Ni and Fe and the 6-31G* basis set for all other atoms. [b]The geometries were optimized in CH_2Cl_2 solvent using C-PCM. [c]Bond distance represents an average value. [d]Defined as the dihedral angle between the planes generated by the groups of three atoms listed.

Table S7. Selected Bond Lengths (Å) and Angles (°) for [**1d**]$^+$ Using Different Functionals[a]

	Expt	Calc					
		BP86	**BP86**[b]	**BP86-D2**	**B3P86**	**M06-L**	**ωB97X-D**
Ni–Fe	2.52	2.57	2.54	2.51	2.56	2.50	2.56
Ni–Cp′$_{centroid}$	1.72	1.75	1.74	1.72	1.74	1.70	1.73
Ni–S[c]	2.17	2.19	2.19	2.18	2.18	2.20	2.20
Fe–S[c]	2.22	2.26	2.26	2.25	2.25	2.28	2.26
Fe–P[c]	2.24	2.27	2.26	2.22	2.27	2.24	2.28
Fe–C	1.75	1.73	1.72	1.73	1.73	1.73	1.77
S_1–Ni–S_2	89.8	89.8	89.8	92.0	90.1	91.1	90.8
S_1–Fe–S_2	87.4	86.4	86.4	88.4	86.8	87.2	87.8
P_1–Fe–P_2	86.5	87.3	86.5	86.9	87.2	86.2	86.6
S_1–Ni–S_2/ S_1–Fe–S_2[d]	106.7	106.4	104.9	106.7	107.1	103.3	107.4

[a]Unless stated otherwise, the geometries were optimized in the gas phase using the SDD pseudopotential and associated basis set for Ni and Fe and the 6-31G* basis set for all other atoms. [b]The geometries were optimized in CH_2Cl_2 solvent using C-PCM. [c]Bond distance represents an average value. [d]Defined as the dihedral angle between the planes generated by the groups of three atoms listed.

Table S8. Selected Bond Lengths (Å) and Angles (°) for [**2b**]^{0a}

	Expt	**Calc**
Ni–Fe	2.46	2.45
Ni–$Cp_{centroid}$	1.82	1.82
Ni–S[b]	2.31	2.32
Fe–S[b]	2.29	2.33
Fe–P[b]	2.19	2.22
Fe–C	1.74	1.73
S_1–Ni–S_2	82.2	83.4
S_1–Fe–S_2	83.1	83.1
P_1–Fe–P_2	85.4	86.8
S_1–Ni–S_2/ S_1–Fe–S_2[c]	90.9	89.5

[a]The geometries were optimized in the gas phase using the BP86 DFT functional, the SDD pseudopotential and associated basis set for Ni and Fe, and the 6-31G* basis set for all other atoms. [b]Bond distance represents an average value. [c]Defined as the dihedral angle between the planes generated by the groups of three atoms listed.

Table S9. Calculated Reaction Free Energies and Free Energy Barriers using the BP86 Functional for Isomerization of Fe(CO)(dppe) from Dibasal to Apical-Basal for all Intermediates in the Proposed Catalytic Cycle when the pdt Ring is Oriented Toward Fe[a,b,c]

Compound	$\Delta G^{\circ d}$	$\Delta G^{\ddagger d,e}$
[**1a**]^0	4.66	11.29
[H**1a**]$^+$	3.99	21.02
[H**1a**]^0	5.16	22.21
[H_2**1a**]$^+$	9.41	27.26
[**1a**]$^+$	3.34	20.36

[a]See Figure 10 in main text for structures of all species. [b]The calculated values corresponding to the case where the pdt ring is oriented toward the Ni center are provided in the Main Text (Table 11). [c]All geometries were optimized in the gas phase using the SDD pseudopotential and associated basis set for Ni and Fe, the 6-31G** basis set for the μ-H ligand, and the 6-31G* basis set for all other atoms. Solvation free energies were calculated in CH_2Cl_2 using C-PCM. [d]Values given in kcal/mol relative to the dibasal isomer. [e]The potential energy surface was found to be relatively flat along the isomerization pathway, leading to relatively small imaginary frequencies for the transition states (TSs).

Table S10. Calculated Spin Densities and Bond Lengths (Å) of [**1a**]^0, [H**1a**]$^+$, and [H**1a**]^0 Using the BP86 Functional when the pdt Ring is Oriented Toward Fe[a,b]

	[**1a**]^0		[H**1a**]$^+$		[H**1a**]^0	
	Cycle A	**Cycle B**	**Cycle A**	**Cycle B**	**Cycle A**	**Cycle B**
***ρ*(Ni)**	0.71	0.70	0.45	0.40	0.00	0.00
***ρ*(Fe)**	–0.17	–0.17	0.23	0.33	0.00	0.00
***ρ*(2S)**	0.23	0.23	0.13	0.10	0.00	0.00
***ρ*(C_5H_5)**	0.25	0.24	0.17	0.15	0.00	0.00
***d*(Ni–Fe)**	2.53	2.72	2.60	2.60	3.08	2.87
***d*(Ni–H)**	n/a	n/a	1.80	1.77	2.53	2.04
***d*(Fe–H)**	n/a	n/a	1.61	1.58	1.54	1.53

[a]For these calculations, the pdt ring is oriented toward the Fe center. An analogous table for the pdt ring oriented toward the Ni center is provided in the Main Text (Table 10). [b]The geometries were optimized in the gas phase using the SDD pseudopotential and associated basis set for Ni and Fe, the 6-31G** basis set for the μ-H ligand, and the 6-31G* basis set for all other atoms.

Table S11. Calculated $\tilde{\nu}_{CO}$ (cm^{-1}) for all Intermediates in the Catalytic Cycle of [**1a**]$^+$ Using the BP86 Functional[a]

	Expt	**Calc**[b]	
		Cycle A	**Cycle B**
[**1a**]^0	1901	1901	1904
[H**1a**]$^+$	n/a	1947	1963
[H**1a**]^0	n/a	1910	1924
[H_2**1a**]$^+$	n/a	1957	1968
[**1a**]$^+$	1943	1946	1949

[a]See Figure 10 in Main Text for structures of all intermediates. [b]The geometries were optimized in the gas phase using the SDD pseudopotential and associated basis set for Ni and Fe, the 6-31G** basis set for the μ-H ligand, and the 6-31G* basis set for all other atoms.

Electronic Structure of $[\mathbf{1a}]^+$, $[\mathbf{1a}]^0$, $[\mathbf{2b}]^+$, and $[\mathbf{2b}]^0$

The unusually short Ni–Fe distance in both $[\mathbf{1a}]^+$ and $[\mathbf{1a}]^0$ (2.5145 and 2.4593 Å, respectively) is attributed to the σ-bond between the two metal centers. This σ-bond can be localized using the Pipek-Mezey criteria, and an analysis of the bonding nature is provided in the Main Text. It is also possible to localize the Ni–Fe bond using Natural Bond Orbitals (NBO). For $[\mathbf{1a}]^+$, the Pipek-Mezey localized MO (Figure 9, left) reveals that the 2-center, 2-electron bond is dative in nature, with Ni donating electron density to Fe. Interestingly, NBO cannot localize this dative bond and instead represents it as a lone pair on the Ni center. This result suggests that either no such bond exists or that the bond is highly polarized (<5% contribution from Fe). The observations that the bond can be localized using the Pipek-Mezey criteria and the Ni–Fe distance is very short suggest that the latter explanation is more accurate for $[\mathbf{1a}]^+$. In contrast, NBO is able localize the 2-center, 2-electron Ni–Fe σ-bond in $[\mathbf{1a}]^0$ (Figure S55), and it is qualitatively similar to the Pipek-Mezey localized MO (Figure 9, right). Again, the shortening of the bond upon reduction of $[\mathbf{1a}]^+$ to $[\mathbf{1a}]^0$ is due to the increased covalency (i.e., strengthening) of the Ni–Fe σ-bond, which has nearly equal contribution from Ni and Fe.

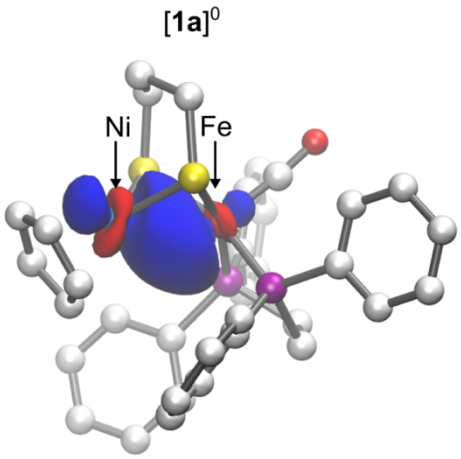

Figure S55. Natural Bond Orbital (NBO) of $[\mathbf{1a}]^0$ showing the Ni-Fe σ-bond from the structure optimized in the gas phase using BP86/SDD/6-31G*. The hydrogens have been removed for clarity.

As mentioned in the Main Text, the isomerization of the Fe(CO)(dppe) subunit from the dibasal (Cycle A) to apical-basal (Cycle B) orientation does not disrupt the Ni–Fe σ-bond in $[\mathbf{1a}]^+$ and $[\mathbf{1a}]^0$ (Figure 9). The Pipek-Mezey localized MOs for the apical-basal isomer are shown in Figure S56 and are in qualitative agreement with the bonding MOs shown in Figure 9 of the Main Text for the dibasal isomer. The composition of the bonding MO in $[\mathbf{1a}]^+$ is heavily Ni-based and in $[\mathbf{1a}]^0$ is nearly equally split between Ni and Fe.

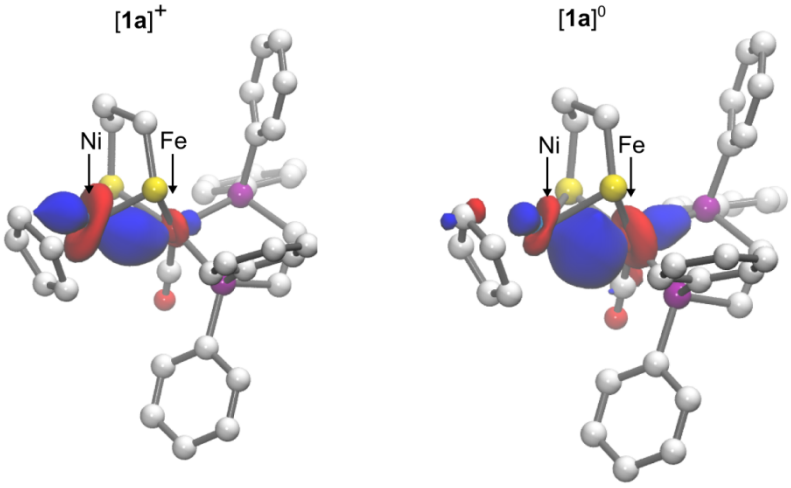

Figure S56. Pipek-Mezey localized molecular orbitals showing the Ni–Fe σ-bonds in [**1a**]$^{+}$ (left) and [**1a**]^{0} (right) for the apical-basal isomer. The composition of the MO in [**1a**]$^{+}$ is 74% Ni and 26% Fe, and the MO in [**1a**]^{0} is 48% Ni and 52% Fe. The analogous figure for the dibasal isomer is provided in the Main Text (Figure 9). The hydrogens have been removed for clarity.

In addition to the preservation of the Ni–Fe bond, the apical-basal (Cycle B) isomer of [**1a**]^{0} also maintains the localization of the unpaired spin density about the Ni coordination sphere (Table 10 in Main Text and Table S10), suggesting that the reduction from [**1a**]$^{+}$ to [**1a**]^{0} is Ni-based regardless of the Fe(CO)(dppe) and pdt orientations. The plot of the spin density is depicted in Figure S57.

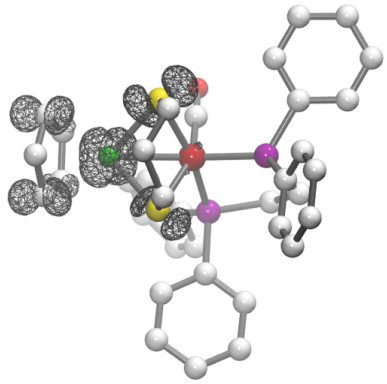

Figure S57. Top-down view of the isocontour plot of the unpaired spin density for the apical-basal isomer of [**1a**]^{0}, showing the localization about the Ni coordination sphere. Color Scheme: green, Ni; maroon, Fe; yellow, S; purple, P; red, O; white, C. The analogous figure for the dibasal isomer is provided in Figure 6 of Main Text. The hydrogens have been removed for clarity.

As mentioned in the Main Text, the chemical bonding pattern is qualitatively similar for the [**2b**]$^{+}$ and [**2b**]^{0} species. The Pipek-Mezey localized MOs featuring the Ni–Fe σ-bonds are shown in Figure S58. Again, the composition of the bonding MO in [**2b**]$^{+}$ is heavily Ni-based and in [**2a**]^{0} is nearly equally split between Ni and Fe.

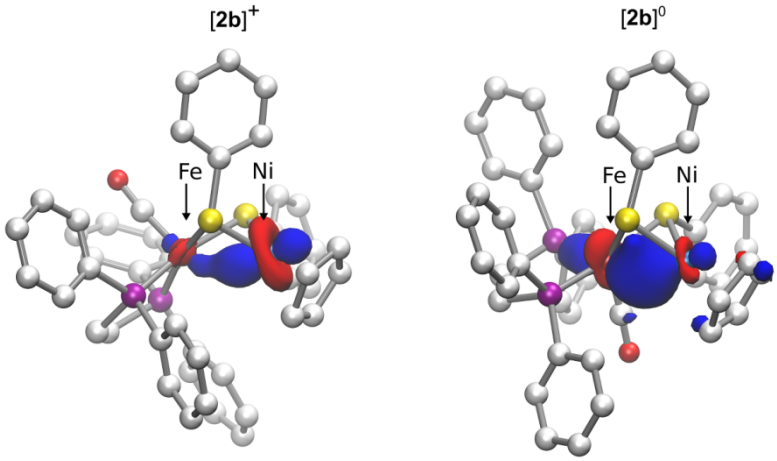

Figure S58. Pipek-Mezey localized molecular orbitals showing the Ni–Fe σ-bonds in [**2b**]$^{+}$ (left) and [**2b**]^{0} (right). The composition of the MO in [**2b**]$^{+}$ is 88% Ni and 12% Fe, and the MO in [**2b**]^{0} is 42% Ni and 58% Fe. The hydrogens have been removed for clarity.

Comparing the Isomerization between Cycle A and Cycle B for $[\mathbf{1a}]^+$ and $[\mathbf{1a}]^0$.

The structures of $[\mathbf{1a}]^+$ and $[\mathbf{1a}]^0$ are similar; however, the main difference is that the Ni center is reduced from Ni(II) to Ni(I) in $[\mathbf{1a}]^0$, which contracts the Ni–Fe bond and elongates the Ni-ligand bonds. The isomerization of the Fe(CO)(dppe) subunit from dibasal (Cycle A) to apical-basal (Cycle B) proceeds through a lower free energy barrier for $[\mathbf{1a}]^0$ (11.76 kcal/mol) than for $[\mathbf{1a}]^+$ (21.54 kcal/mol). While the structures of $[\mathbf{1a}]^+$ and $[\mathbf{1a}]^0$ are similar, the transition states (TSs) associated with their respective isomerizations are quite different. As seen in Figure S59, the TS for $[\mathbf{1a}]^0$ features an elongated Ni-Fe distance and intramolecular electron transfer from the Ni to the Fe center to form a Ni(II)Fe(I) TS. The pentacoordinate Fe(I) center of the TS of $[\mathbf{1a}]^0$ is more accommodating of rotation than the hexacoordinate Fe(II) center in the TS of $[\mathbf{1a}]^+$. Similar arguments could be made for the other intermediates in the catalytic cycle, which also feature hexacoordinate Fe(II) centers and further rigidifying bridging hydride ligands.

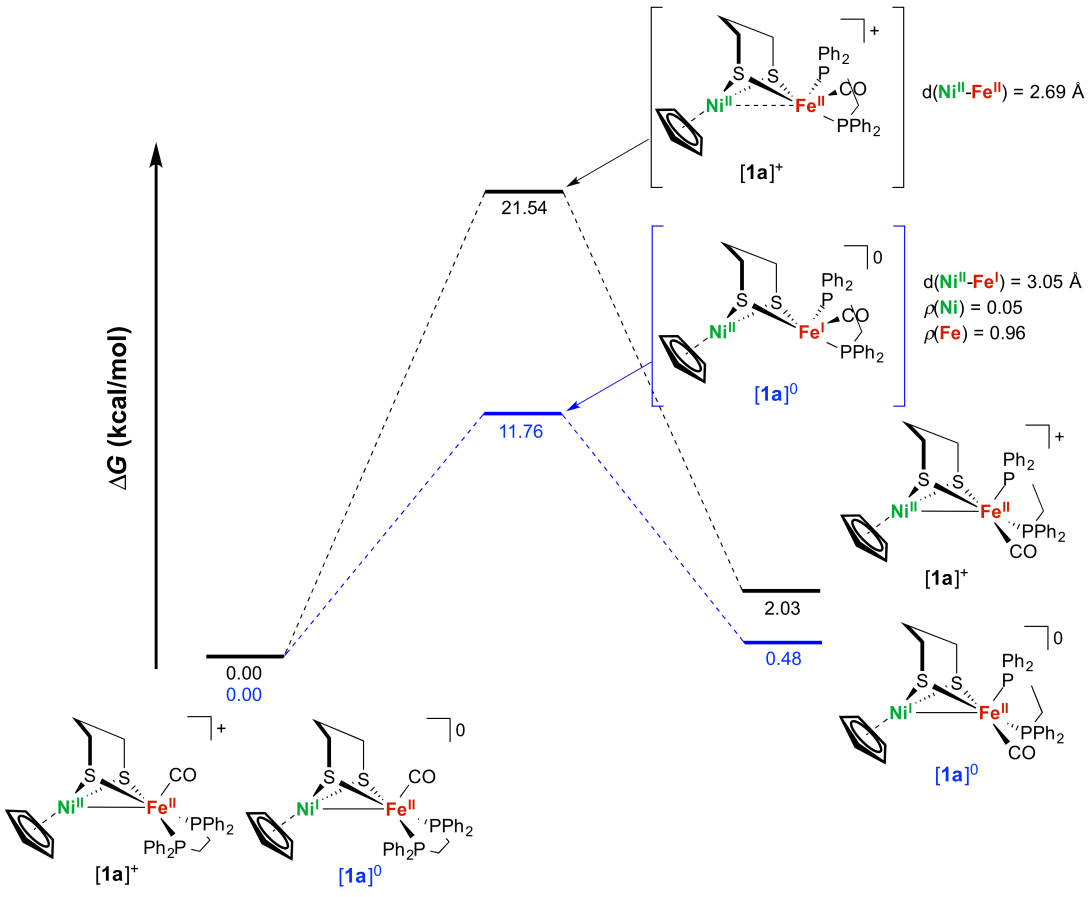

Figure S59. Free energy diagram for the isomerization of the Fe(CO)(dppe) subunit from dibasal (Cycle A) to apical-basal (Cycle B) for $[\mathbf{1a}]^+$ (black numbers) and $[\mathbf{1a}]^0$ (blue numbers).

The following tables provide the optimized Cartesian coordinates of all species studied herein. The geometries were optimized in the gas phase using the BP86 functional with the SDD pseudopotential and associated basis set for the Ni and Fe centers, the 6-31G** basis set for the μ-H ligands, and the 6-31G* basis set for all other atoms.

Table S12. $[CpNi^{II}(pdt)Fe^{II}(dppe)(CO)]^+$ in Cycle A with pdt Oriented Toward Ni ($[\mathbf{1a}]^+$, –3204.21402316 E_h)

atom	x	y	z
Ni	10.278976	8.685769	7.506633
Fe	11.605186	6.865203	8.768546
P	11.263092	6.829652	11.024459
S	11.987742	9.091285	8.831761
P	10.560518	4.854142	8.701351
S	11.442101	7.097433	6.527789
C	11.113074	1.111088	7.031790
H	10.570711	0.159524	7.024950
C	12.338349	1.231423	6.353184
H	12.754288	0.371789	5.816998
C	8.134972	9.289818	12.145532
H	7.080481	9.395395	11.866868
C	7.701224	4.824240	9.026236
H	7.861467	4.769000	10.107699
C	8.687911	10.112983	13.142266
H	8.066944	10.865528	13.639630
C	9.299120	10.580691	7.258654
H	9.790755	11.548872	7.366544
C	10.035699	9.961105	13.500969
H	10.472054	10.594259	14.280905
C	13.211065	6.246680	8.899772
C	12.492450	3.558715	7.048198
H	13.035621	4.506723	7.048386
C	9.153014	9.835937	6.064131
H	9.500491	10.119053	5.069690
C	13.192655	9.363756	6.225170
H	12.339751	9.980391	5.885046
H	14.090783	9.735658	5.691013
C	6.139257	4.876385	7.157791
H	5.111439	4.879506	6.779900
C	11.267019	3.445608	7.735024
C	13.024396	2.455020	6.360487
H	13.977533	2.555602	5.830424
C	10.833104	8.988205	12.874802
H	11.876553	8.867991	13.180845
C	8.492769	8.589646	6.409997
H	8.261360	7.780497	5.716321
O	14.339087	5.909988	8.918349
C	10.524630	4.174101	10.451898
H	9.782377	3.360675	10.541610
H	11.523267	3.726437	10.607420
C	7.222391	4.889727	6.261278
H	7.040858	4.892537	5.180703
C	10.265875	5.299962	11.461768
H	10.459527	4.985166	12.502085
H	9.212991	5.628480	11.431394

C	10.288302	8.160544	11.871609
C	10.576537	2.210909	7.715299
H	9.610758	2.110252	8.223519
C	8.710647	9.810802	8.347974
H	8.688047	10.116955	9.394790
C	8.164613	8.608110	7.806899
H	7.642808	7.817789	8.346388
C	6.382154	4.840026	8.539818
H	5.544973	4.807805	9.245654
C	12.966342	7.907578	5.816400
H	13.819221	7.270857	6.112116
H	12.844323	7.831040	4.722037
C	8.932163	8.327676	11.507926
H	8.494535	7.701678	10.720704
C	8.795718	4.846283	8.134463
C	8.539984	4.876979	6.743093
H	9.375697	4.867593	6.034657
C	12.834435	6.716880	11.989741
C	13.793833	7.744989	11.832438
H	13.585213	8.586646	11.161864
C	13.132433	5.633860	12.843363
H	12.420865	4.814478	12.984155
C	15.295793	6.616145	13.377681
H	16.248939	6.575600	13.915210
C	14.358003	5.583782	13.528183
H	14.575218	4.733295	14.183123
C	15.009307	7.699293	12.529984
H	15.737015	8.508295	12.404909
C	13.411493	9.573910	7.724135
H	13.587282	10.642530	7.938008
H	14.285679	9.003152	8.085456

Table S13. $[CpNi^{II}(pdt)Fe^{II}(dppe)(CO)]^+$ in Cycle A with pdt Oriented Toward Fe ($[\mathbf{1a}]^+$, –3204.21506968 E_h)

atom	x	y	z
Ni	10.288890	8.654004	7.546586
Fe	11.647336	6.844533	8.768726
P	11.280366	6.824961	11.020321
S	12.012952	9.076378	8.836414
P	10.569520	4.855162	8.688245
S	11.443890	7.092480	6.530323
C	11.125432	1.107971	7.031393
H	10.599190	0.147638	7.052975
C	12.320888	1.243015	6.304079
H	12.729993	0.385922	5.758667
C	8.129038	9.262273	12.134497
H	7.069939	9.349535	11.867319
C	7.712015	4.826237	9.024997
H	7.876651	4.769102	10.105634
C	8.683337	10.108971	13.110498
H	8.058433	10.860810	13.603980
C	9.344119	10.561041	7.299863
H	9.854654	11.518463	7.416333
C	10.037855	9.981885	13.453225
H	10.475838	10.634021	14.216402
C	13.210121	6.130976	8.959611
C	12.463055	3.578321	6.974529
H	12.988300	4.536157	6.945034
C	9.198698	9.821323	6.101981
H	9.566151	10.098448	5.112984
C	13.904206	8.580172	6.718733
H	14.712853	9.044886	6.119051
H	14.373063	7.773581	7.308531
C	6.142140	4.877333	7.163316
H	5.112735	4.878517	6.789770
C	11.268419	3.450113	7.711244
C	12.985532	2.477993	6.274317
H	13.914922	2.589766	5.705710
C	10.840013	9.009273	12.832925
H	11.888850	8.908143	13.127133
C	8.502461	8.591160	6.437067
H	8.262294	7.788350	5.738989
O	14.297249	5.688899	9.050502
C	10.540058	4.170834	10.436419
H	9.798677	3.356865	10.527412
H	11.539565	3.724211	10.589179
C	7.221355	4.895097	6.262277
H	7.035325	4.899699	5.182511
C	10.281198	5.294236	11.448952
H	10.472012	4.974580	12.488067
H	9.228629	5.623007	11.418537
C	10.293190	8.157027	11.851439
C	10.598032	2.204343	7.727295
H	9.654327	2.091827	8.273157
C	8.719312	9.804637	8.378631

H	8.688894	10.110502	9.425209
C	8.153329	8.617530	7.827886
H	7.608584	7.837372	8.359410
C	6.390926	4.839000	8.544221
H	5.556881	4.803033	9.253567
C	12.883587	7.985443	5.749995
H	13.362466	7.226760	5.105036
H	12.447688	8.758254	5.091527
C	8.930745	8.300550	11.501876
H	8.491751	7.659270	10.728046
C	8.802453	4.853117	8.128526
C	8.540954	4.884812	6.738481
H	9.373478	4.879134	6.026331
C	12.839321	6.723872	12.008485
C	13.814246	7.732855	11.825717
H	13.627267	8.547658	11.116957
C	13.109383	5.673876	12.911163
H	12.385555	4.869730	13.074589
C	15.276665	6.650350	13.440223
H	16.220663	6.619940	13.994310
C	14.323099	5.636896	13.617464
H	14.518732	4.811295	14.309948
C	15.017752	7.701170	12.544439
H	15.757933	8.495160	12.398142
C	13.343119	9.645757	7.658865
H	12.920274	10.500415	7.100614
H	14.142473	10.036431	8.314051

Table S14. $[CpNi^{II}(pdt)Fe^{II}(dppe)(CO)]^+$ in Cycle B with pdt Oriented Toward Ni ($[\mathbf{1a}]^+$, – 3204.21250942 E_h)

atom	x	y	z
Ni	10.402720	8.717211	7.549389
Fe	11.457749	6.819719	8.815851
P	13.193832	5.525003	9.311297
S	12.107824	9.003699	8.924943
P	10.891035	6.791479	11.026377
S	11.530868	7.099384	6.572706
C	7.506110	6.490987	13.390587
H	7.290024	6.468939	14.464115
C	6.461053	6.380029	12.457223
H	5.427495	6.269408	12.802046
C	16.485192	7.932358	10.076622
H	16.758910	8.691519	10.817385
C	12.545502	7.879507	13.111382
H	12.990164	6.886190	13.229592
C	17.384651	7.588085	9.052241
H	18.363293	8.076112	8.993772
C	9.511111	10.389802	6.516619
H	10.040619	11.125946	5.911171
C	17.023744	6.615649	8.106575
H	17.719994	6.339653	7.307242
C	10.379738	5.459577	8.592522
C	8.070555	6.556293	10.640382
H	8.285698	6.581121	9.568736
C	8.961356	9.149411	6.063305
H	9.012835	8.757568	5.045908
C	13.363190	9.296494	6.338834
H	12.532715	9.926560	5.968900
H	14.284716	9.647159	5.831282
C	12.412205	10.197263	13.846039
H	12.751341	11.005528	14.502508
C	9.125567	6.661913	11.569383
C	6.744575	6.414872	11.083421
H	5.933704	6.329869	10.351909
C	15.773372	5.979268	8.186610
H	15.513582	5.203813	7.458475
C	8.317934	8.486809	7.182074
H	7.797423	7.529499	7.133939
O	9.639047	4.562436	8.418867
C	11.633968	5.187493	11.654892
H	11.596990	5.136056	12.757809
H	10.964469	4.396283	11.271310
C	11.394499	10.432083	12.905939
H	10.935463	11.423607	12.829024
C	13.063496	4.991107	11.123343
H	13.374643	3.936673	11.215876
H	13.789238	5.598453	11.690103
C	14.867300	6.312556	9.218002
C	8.830020	6.635093	12.952272
H	9.633858	6.739526	13.689604
C	9.263967	10.465269	7.927551

H	9.592848	11.267927	8.590007
C	8.499259	9.305111	8.332426
H	8.144095	9.092876	9.341265
C	12.983735	8.920014	13.948950
H	13.765251	8.723911	14.691072
C	13.119232	7.842296	5.939930
H	13.934888	7.189612	6.299195
H	13.054551	7.746432	4.842118
C	15.231325	7.307329	10.155333
H	14.532946	7.610209	10.944876
C	11.528370	8.106605	12.159411
C	10.957670	9.397069	12.067545
H	10.153079	9.586516	11.348292
C	13.422246	3.937418	8.388616
C	12.720293	3.672333	7.195065
H	12.016963	4.410147	6.797972
C	14.338756	2.973503	8.873848
H	14.922583	3.168465	9.780769
C	13.815774	1.507174	7.004061
H	13.963799	0.562097	6.470859
C	14.530035	1.766745	8.186754
H	15.240432	1.028992	8.574891
C	12.916436	2.462365	6.507995
H	12.358768	2.268253	5.585654
C	13.541024	9.503311	7.843123
H	13.697825	10.572987	8.066358
H	14.413599	8.943460	8.223250

Table S15. $[CpNi^{II}(pdt)Fe^{II}(dppe)(CO)]^+$ in Cycle B with pdt Oriented Toward Fe ($[\mathbf{1a}]^+$, –3204.21195089 E_h)

atom	x	y	z
Ni	10.329214	8.711862	7.628986
Fe	11.495072	6.804895	8.777204
P	13.248253	5.518554	9.302176
S	12.136376	8.987428	8.858769
P	10.922614	6.797971	10.981182
S	11.354933	7.116386	6.527692
C	7.525084	6.563514	13.332718
H	7.302782	6.596327	14.404693
C	6.489782	6.372132	12.401864
H	5.457075	6.253723	12.746626
C	16.462542	7.990453	10.148369
H	16.652853	8.836382	10.818048
C	12.568699	7.905356	13.061059
H	13.013139	6.913336	13.189247
C	17.482482	7.511768	9.307029
H	18.470146	7.984887	9.317306
C	9.462298	10.450441	6.709078
H	10.008776	11.198142	6.133397
C	17.230356	6.424847	8.456042
H	18.020413	6.044554	7.799510
C	10.456279	5.410983	8.577741
C	8.106787	6.488627	10.586057
H	8.326015	6.462244	9.515467
C	8.860397	9.258373	6.204994
H	8.874727	8.918044	5.168042
C	13.884382	8.468116	6.617894
H	14.683552	8.917050	5.993862
H	14.335153	7.604945	7.135483
C	12.434659	10.230301	13.772712
H	12.771832	11.044467	14.422860
C	9.152395	6.675487	11.513251
C	6.782259	6.336338	11.029816
H	5.979212	6.188112	10.299766
C	15.966265	5.810889	8.444586
H	15.785341	4.958545	7.782315
C	8.219585	8.559308	7.305531
H	7.670189	7.620860	7.219747
O	9.749219	4.485342	8.417716
C	11.647680	5.196275	11.631879
H	11.600806	5.159683	12.735080
H	10.973561	4.406770	11.253056
C	11.421825	10.457512	12.825447
H	10.964776	11.448977	12.736802
C	13.078347	4.984269	11.113624
H	13.379098	3.927797	11.215633
H	13.804411	5.588701	11.683246
C	14.938687	6.283178	9.289252
C	8.847944	6.717383	12.894002
H	9.643859	6.881394	13.628903
C	9.241745	10.460994	8.130589

H	9.609091	11.220207	8.823479
C	8.440195	9.314259	8.489885
H	8.096620	9.059863	9.492789
C	13.004140	8.953420	13.890534
H	13.782523	8.763614	14.637502
C	12.772918	7.985633	5.691987
H	13.164609	7.241406	4.975168
H	12.329793	8.813189	5.108921
C	15.196726	7.386276	10.136312
H	14.407416	7.781433	10.787597
C	11.556357	8.124741	12.102151
C	10.987701	9.415053	11.995038
H	10.186996	9.598003	11.269887
C	13.471694	3.934901	8.373046
C	12.840679	3.733408	7.128126
H	12.198485	4.514982	6.710066
C	14.304000	2.914188	8.891895
H	14.834247	3.055870	9.840422
C	13.839045	1.521233	6.951472
H	13.975882	0.582487	6.404309
C	14.482328	1.715852	8.185888
H	15.126759	0.933580	8.601022
C	13.023365	2.532614	6.422543
H	12.519765	2.388242	5.460793
C	13.452039	9.509310	7.647804
H	13.077535	10.431308	7.167462
H	14.309860	9.784497	8.286868

Table S16. $[CpNi^{II}(pdt)Fe^{II}(dppe)(CO)]^+$ Transition State with pdt Oriented Toward Ni ($[\mathbf{1a}]^+$, –3204.18116822 E_h)

atom	x	y	z
Ni	10.144336	8.382023	6.946605
Fe	11.391763	7.082924	8.949983
P	12.005481	7.340579	11.116823
S	11.352001	9.364678	8.486299
P	9.554369	6.102962	9.695009
S	11.732148	6.910882	6.624899
C	7.606676	2.545646	8.800631
H	6.969003	1.830452	9.331177
C	7.958636	2.319563	7.459358
H	7.597138	1.425245	6.940879
C	11.535858	11.212425	12.622813
H	10.968012	12.114147	12.368842
C	8.214989	8.352978	10.767518
H	9.215431	8.702006	11.041067
C	12.343630	11.187010	13.769583
H	12.406919	12.066914	14.418626
C	9.326908	9.245240	5.117491
H	9.912026	9.602841	4.269698
C	13.074145	10.028188	14.079404
H	13.707739	9.997987	14.972380
C	12.128542	5.513632	8.938855
C	9.250261	4.382383	7.456260
H	9.910459	5.079891	6.931226
C	8.876778	7.889401	5.330242
H	9.063900	7.048982	4.659096
C	13.181224	9.369485	6.255281
H	12.379008	9.733474	5.586986
H	14.132212	9.801692	5.881808
C	5.810021	8.726654	10.734690
H	4.940714	9.339485	10.995473
C	8.901378	4.621528	8.801972
C	8.781041	3.238583	6.789763
H	9.069642	3.062248	5.748031
C	13.001595	8.901952	13.246255
H	13.583104	8.009796	13.499328
C	8.105179	7.834525	6.546783
H	7.611464	6.952108	6.951579
O	12.647818	4.460544	8.864672
C	9.999664	5.369274	11.377625
H	9.087946	5.051537	11.912464
H	10.600176	4.467024	11.161182
C	5.641331	7.528063	10.023580
H	4.639268	7.201409	9.725184
C	10.790545	6.390917	12.202213
H	11.310867	5.928561	13.059706
H	10.112108	7.157632	12.615944
C	12.189461	8.917447	12.087019
C	8.074141	3.686726	9.469754
H	7.783941	3.846234	10.513789
C	8.923502	9.996385	6.252011

H	9.145798	11.047616	6.443666
C	8.167514	9.123105	7.142902
H	7.723223	9.418599	8.093970
C	7.101469	9.136718	11.107457
H	7.245234	10.070657	11.661638
C	13.279983	7.848125	6.155472
H	14.092729	7.450415	6.788382
H	13.479402	7.543456	5.113497
C	11.455856	10.085796	11.787204
H	10.839749	10.132428	10.885801
C	8.055165	7.136697	10.065696
C	6.753194	6.735525	9.691716
H	6.600819	5.799184	9.146053
C	13.660778	6.557540	11.367860
C	14.806736	7.262794	10.931060
H	14.703712	8.277280	10.527612
C	13.815787	5.261982	11.902542
H	12.946080	4.686586	12.237288
C	16.225062	5.395135	11.576737
H	17.219394	4.944204	11.660558
C	15.093049	4.686949	12.008413
H	15.199805	3.681767	12.429793
C	16.079594	6.685749	11.038694
H	16.960470	7.246439	10.707743
C	12.949014	9.897890	7.670048
H	12.905992	11.000544	7.664649
H	13.763165	9.590939	8.350048

Table S17. $[CpNi^{II}(pdt)Fe^{II}(dppe)(CO)]^+$ Transition State with pdt Oriented Toward Fe ($[\mathbf{1a}]^+$, –3204.18536467 E_h)

atom	x	y	z
Ni	10.128052	8.350780	6.973368
Fe	11.408357	7.080048	8.942431
P	12.021156	7.337268	11.111976
S	11.348941	9.356855	8.482025
P	9.564547	6.106750	9.681695
S	11.712460	6.890707	6.616337
C	7.591971	2.559601	8.803664
H	6.948895	1.852430	9.338428
C	7.942437	2.322872	7.463865
H	7.574510	1.428176	6.950592
C	11.547240	11.214479	12.609928
H	10.994181	12.121804	12.343561
C	8.238699	8.358199	10.766526
H	9.241644	8.702457	11.037130
C	12.326376	11.179790	13.775978
H	12.382407	12.058149	14.427767
C	9.363356	9.251452	5.160093
H	9.977311	9.625312	4.340104
C	13.037593	10.013300	14.101676
H	13.648999	9.975586	15.009705
C	12.152373	5.513414	8.937068
C	9.248726	4.376516	7.448989
H	9.913115	5.065983	6.919041
C	8.889163	7.897191	5.323313
H	9.086968	7.071330	4.637327
C	13.808990	8.785015	7.114750
H	14.719027	9.246443	6.680668
H	14.131856	8.182718	7.985896
C	5.835511	8.743709	10.742276
H	4.970002	9.360091	11.007307
C	8.901867	4.626335	8.793163
C	8.771453	3.232092	6.789180
H	9.058760	3.047950	5.748440
C	12.974633	8.889132	13.265054
H	13.542176	7.991568	13.530455
C	8.080095	7.821672	6.514648
H	7.569026	6.933252	6.883514
O	12.670192	4.459665	8.864066
C	10.011178	5.367012	11.361607
H	9.099406	5.048206	11.895695
H	10.609995	4.464587	11.141265
C	5.658833	7.547822	10.028437
H	4.654273	7.226908	9.732184
C	10.803422	6.383259	12.190941
H	11.323965	5.915297	13.045232
H	10.125800	7.148246	12.609034
C	12.191612	8.914042	12.085915
C	8.067758	3.701129	9.466046
H	7.778000	3.868737	10.508894
C	8.929781	9.978799	6.301396

H	9.154827	11.023089	6.525313
C	8.133888	9.093460	7.142977
H	7.664532	9.370559	8.087383
C	7.130111	9.146431	11.112074
H	7.280317	10.078298	11.668012
C	13.196638	7.866219	6.060568
H	13.923969	7.095654	5.748896
H	12.889242	8.430120	5.160974
C	11.476536	10.089572	11.770897
H	10.880337	10.142873	10.856454
C	8.070743	7.144830	10.061746
C	6.765847	6.750834	9.690923
H	6.607317	5.817216	9.142477
C	13.677248	6.560988	11.380230
C	14.828871	7.283330	10.989089
H	14.729190	8.307238	10.609993
C	13.827185	5.253374	11.886428
H	12.953223	4.665254	12.186257
C	16.242597	5.407381	11.621749
H	17.237336	4.959852	11.718114
C	15.104901	4.682658	12.008641
H	15.207466	3.667935	12.407590
C	16.102212	6.710316	11.112906
H	16.987288	7.284076	10.817498
C	12.883138	9.907039	7.578892
H	12.550344	10.535604	6.733129
H	13.399652	10.560738	8.304410

Table S18. $[CpNi^{II}(pdt)Fe^{II}(dppe)(CO)]^+$ Transition State for Interconverting ^{31}P Sites in Cycle B with pdt Oriented Toward Ni
($[\mathbf{1a}]^+$, –3204.19703182 E_h)

atom	x	y	z
Ni	9.959845	8.635246	7.737942
Fe	11.361653	6.834973	8.962178
P	11.576109	4.634731	8.668420
S	11.737520	9.113903	8.977579
P	12.304073	6.555967	10.943862
S	11.108269	7.071974	6.671667
C	12.215189	8.278021	14.704764
H	12.820559	8.229809	15.616266
C	11.057404	9.073408	14.666475
H	10.757855	9.647513	15.549763
C	15.248507	4.562617	6.769055
H	16.091172	5.252007	6.648067
C	14.984016	5.535441	11.060852
H	14.556760	4.535294	11.182849
C	15.310741	3.269654	6.224634
H	16.199907	2.949534	5.671049
C	9.023850	10.501783	7.181511
H	9.555023	11.439805	7.014615
C	14.232101	2.385254	6.393705
H	14.278538	1.374669	5.973820
C	9.817417	6.577987	9.650764
C	10.670837	8.400660	12.360021
H	10.068069	8.453795	11.448562
C	8.718879	9.516061	6.216639
H	8.969682	9.541404	5.155144
C	12.748030	9.422599	6.289984
H	11.859528	10.030839	6.038843
H	13.592166	9.830449	5.696581
C	16.959530	6.950677	10.905065
H	18.049130	7.060288	10.894753
C	11.829395	7.597698	12.392166
C	10.287972	9.134525	13.494105
H	9.387481	9.757166	13.459537
C	13.088649	2.790130	7.100092
H	12.252101	2.093957	7.222291
C	8.036930	8.419241	6.898183
H	7.690256	7.500246	6.422494
O	8.768217	6.344679	10.147198
C	11.867975	4.817884	11.501655
H	12.499785	4.509355	12.353509
H	10.835004	4.910535	11.882723
C	16.135519	8.084492	10.792431
H	16.580794	9.081423	10.703551
C	11.933854	3.809490	10.339207
H	11.240431	2.965558	10.495596
H	12.944129	3.377785	10.241308
C	13.013737	4.093777	7.639125
C	12.603621	7.545472	13.573834
H	13.516151	6.939861	13.608812

C	8.543039	10.017576	8.468882
H	8.647999	10.547783	9.417197
C	7.863552	8.774124	8.269261
H	7.384197	8.168136	9.036881
C	16.381511	5.679253	11.042487
H	17.017088	4.793042	11.144017
C	12.519847	7.976390	5.849698
H	13.419346	7.356379	6.014414
H	12.283272	7.941815	4.771932
C	14.104770	4.973282	7.472711
H	14.060461	5.977543	7.911270
C	14.148487	6.666897	10.942790
C	14.740609	7.945175	10.807617
H	14.103381	8.835012	10.743916
C	10.122421	3.750964	7.959906
C	9.970485	3.643613	6.557866
H	10.755269	4.013293	5.890698
C	9.092606	3.272845	8.800515
H	9.170333	3.366089	9.888968
C	7.808347	2.564477	6.858482
H	6.914552	2.096857	6.432018
C	7.945838	2.679319	8.251199
H	7.159765	2.305497	8.915833
C	8.820935	3.049553	6.014621
H	8.720854	2.961855	4.927360
C	13.082417	9.597530	7.770991
H	13.280908	10.661339	7.989724
H	13.982381	9.021332	8.053113

Table S19. $[CpNi^{I}(pdt)Fe^{II}(dppe)(CO)]^{0}$ in Cycle A with pdt Oriented Toward Ni ($[\mathbf{1a}]^{0}$, –3204.37976648 E_h)

atom	x	y	z
Ni	2.427036	6.004331	2.118879
Fe	1.588207	5.961511	4.502278
S	0.975732	7.540924	2.984157
P	2.627744	7.504127	5.753736
O	-0.868802	5.881075	6.064364
C	4.561116	6.325510	1.826157
H	5.249270	6.579572	2.633134
C	3.863975	7.260232	0.995473
H	3.886564	8.346118	1.093196
C	3.061243	6.516002	0.070480
H	2.394383	6.937405	-0.684594
C	-0.723425	7.178498	2.312340
H	-1.422847	7.181838	3.168662
H	-0.970227	8.047455	1.675614
C	-0.856287	5.885180	1.504918
H	-0.142466	5.908047	0.659575
H	-1.878395	5.851592	1.070217
C	0.166206	5.909646	5.486631
C	3.070098	6.656885	7.370965
C	5.468059	8.041438	5.784650
H	5.547975	7.381509	6.655240
C	6.633204	8.656598	5.291542
H	7.596750	8.457566	5.774833
C	6.559882	9.529982	4.196452
H	7.466291	10.013015	3.814909
C	5.314820	9.785503	3.594367
H	5.247814	10.472741	2.743288
C	4.154324	9.164799	4.077122
H	3.188741	9.363275	3.597550
C	2.429787	9.979139	7.139531
H	3.507984	9.872105	7.305982
C	1.733748	9.011518	6.380103
C	0.354347	9.185933	6.152221
H	-0.191090	8.453397	5.553006
C	-0.318218	10.299777	6.682607
H	-1.390853	10.422935	6.494978
C	0.378699	11.250978	7.442956
H	-0.147232	12.120190	7.854117
C	1.755929	11.088559	7.670635
H	2.308912	11.830197	8.258511
H	3.730653	7.284280	7.997203
H	4.741263	5.341374	6.852192
H	2.105495	6.566198	7.902587
S	1.086910	4.349431	2.968035
C	-0.638432	4.604429	2.314836
H	-1.331190	4.560838	3.175504
H	-0.836969	3.721085	1.680936
C	3.670672	5.268430	7.107135
C	4.178668	3.524913	4.810709
C	5.428827	4.147704	4.598598

H	5.597218	5.179228	4.929002
C	6.464521	3.469238	3.938455
H	7.430562	3.965651	3.792362
C	6.259989	2.165622	3.454878
H	7.067940	1.637361	2.936429
C	5.010843	1.549626	3.631832
H	4.836851	0.537483	3.249181
C	3.978178	2.221058	4.306926
H	3.012255	1.725995	4.447424
C	2.594546	2.200028	7.400060
H	3.658387	2.372413	7.601762
C	1.905121	3.022091	6.478446
C	0.551904	2.744819	6.204368
H	0.015279	3.357242	5.476132
C	-0.104853	1.683708	6.851295
H	-1.159228	1.485156	6.628452
C	0.583317	0.885003	7.776501
H	0.069390	0.059961	8.282702
C	1.938185	1.143310	8.047401
H	2.487111	0.517741	8.760623
H	3.590215	4.623558	7.998978
C	4.232011	4.996992	1.377608
H	4.601749	4.065548	1.806500
C	3.307684	5.117747	0.295858
H	2.834898	4.290533	-0.237231
C	4.215389	8.283718	5.181823
P	2.786978	4.437343	5.644757

Table S20. $[CpNi^{I}(pdt)Fe^{II}(dppe)(CO)]^{0}$ in Cycle A with pdt Oriented Toward Fe ($[\mathbf{1a}]^{0}$, –3204.37814099 E_h)

atom	x	y	z
Ni	2.441321	6.001201	2.157813
Fe	1.562657	5.961024	4.517917
S	0.977259	7.538793	2.980689
P	2.621495	7.504762	5.745603
O	-0.749390	5.891834	6.295963
C	4.591935	6.322925	1.804073
H	5.305023	6.577629	2.589072
C	3.864863	7.256764	1.000271
H	3.891111	8.342817	1.095967
C	3.028909	6.512419	0.103740
H	2.338229	6.933406	-0.629979
C	-0.666135	7.176356	2.172506
H	-1.320118	8.030175	2.427666
H	-0.490558	7.201104	1.081179
C	-1.340918	5.866190	2.581676
H	-2.341989	5.832511	2.101242
H	-1.514273	5.863359	3.672507
C	0.209048	5.914780	5.598343
C	3.057700	6.658588	7.364625
C	5.461157	8.035697	5.784956
H	5.535752	7.378819	6.658299
C	6.629307	8.649109	5.296924
H	7.589780	8.452030	5.787041
C	6.562656	9.518115	4.197924
H	7.471389	9.999846	3.820268
C	5.321587	9.770772	3.586617
H	5.260065	10.453939	2.731900
C	4.158088	9.151671	4.064169
H	3.195791	9.347102	3.576904
C	2.423013	9.970631	7.145047
H	3.496191	9.849494	7.333214
C	1.731229	9.016993	6.364460
C	0.359501	9.210054	6.107253
H	-0.179213	8.489039	5.487808
C	-0.310401	10.328593	6.630992
H	-1.377058	10.466931	6.421119
C	0.381538	11.265370	7.413513
H	-0.142515	12.137980	7.819840
C	1.751534	11.084624	7.669474
H	2.300680	11.815427	8.274242
H	3.720783	7.283264	7.990698
H	4.724513	5.332602	6.856588
H	2.091995	6.574133	7.895079
S	1.088910	4.350253	2.968816
C	-0.582512	4.600640	2.177752
H	-1.178020	3.707682	2.442427
H	-0.414858	4.579672	1.084889
C	3.651909	5.266367	7.104772
C	4.171950	3.535316	4.802626
C	5.416395	4.169657	4.592391

H	5.571912	5.204903	4.917100
C	6.461495	3.497621	3.940749
H	7.423185	4.002649	3.795670
C	6.271453	2.189472	3.463454
H	7.086705	1.666212	2.951389
C	5.027503	1.562447	3.637792
H	4.864857	0.546847	3.259396
C	3.985485	2.227324	4.305037
H	3.023696	1.723883	4.443793
C	2.580265	2.195182	7.389023
H	3.640562	2.374864	7.602632
C	1.894673	3.013610	6.461503
C	0.546364	2.727317	6.172122
H	0.014600	3.337622	5.437983
C	-0.109347	1.660339	6.810155
H	-1.159883	1.454427	6.576061
C	0.574720	0.864957	7.741360
H	0.061335	0.035370	8.240630
C	1.924627	1.132470	8.027346
H	2.470323	0.509612	8.745380
H	3.561680	4.622793	7.996574
C	4.246155	4.995790	1.370942
H	4.629281	4.064235	1.787778
C	3.282919	5.114964	0.322206
H	2.794577	4.286816	-0.195374
C	4.212601	8.275594	5.172850
P	2.774277	4.435933	5.638186

Table S21. $[CpNi^{I}(pdt)Fe^{II}(dppe)(CO)]^{0}$ in Cycle B with pdt Oriented Toward Ni ($[\mathbf{1a}]^{0}$, –3204.37876445 E_h)

atom	x	y	z
Ni	10.355618	8.715226	7.626818
Fe	11.483121	6.852139	8.785355
P	13.212069	5.567964	9.280533
S	12.204943	9.026626	8.928084
P	10.915510	6.811888	10.958497
S	11.555965	7.070642	6.491699
C	7.545981	6.375246	13.357529
H	7.338598	6.295279	14.430846
C	6.492469	6.320642	12.428527
H	5.460442	6.195549	12.775362
C	16.538296	7.918852	10.090302
H	16.789767	8.707367	10.808765
C	12.473741	7.876470	13.144125
H	12.896586	6.876036	13.284704
C	17.475059	7.521430	9.120399
H	18.463000	7.993950	9.082355
C	9.400355	10.372801	6.558322
H	9.928952	11.064034	5.898907
C	17.133803	6.521206	8.197238
H	17.854552	6.207329	7.433422
C	10.371934	5.535373	8.516101
C	8.089677	6.596501	10.611552
H	8.302690	6.693708	9.543623
C	8.765147	9.148194	6.164115
H	8.782439	8.709655	5.165038
C	13.362176	9.282093	6.289057
H	12.489877	9.881516	5.965388
H	14.248725	9.669836	5.743057
C	12.350698	10.201439	13.856238
H	12.671936	11.008929	14.523762
C	9.152739	6.643703	11.534250
C	6.766573	6.433804	11.057179
H	5.949993	6.398459	10.327307
C	15.869138	5.910157	8.248925
H	15.621028	5.120772	7.531822
C	8.200006	8.548239	7.330207
H	7.653533	7.603816	7.359678
O	9.608846	4.656045	8.309021
C	11.653203	5.207337	11.614036
H	11.597021	5.147293	12.716373
H	10.996566	4.414755	11.210699
C	11.396581	10.449858	12.854995
H	10.972142	11.453143	12.736428
C	13.090517	5.027279	11.101574
H	13.422695	3.980492	11.211418
H	13.795074	5.659117	11.669791
C	14.925256	6.293999	9.225857
C	8.866404	6.538593	12.914448
H	9.678134	6.601165	13.648569
C	9.161371	10.562784	7.964740

H	9.532836	11.393709	8.567329
C	8.439302	9.433732	8.447870
H	8.109702	9.268448	9.474851
C	12.888384	8.913479	13.998517
H	13.627518	8.707442	14.781347
C	13.145211	7.821848	5.890365
H	13.980384	7.190690	6.245760
H	13.100784	7.730902	4.789705
C	15.271092	7.318661	10.137051
H	14.537771	7.672221	10.871629
C	11.516580	8.114807	12.135551
C	10.986990	9.418556	11.998461
H	10.246282	9.621507	11.217275
C	13.454344	3.952160	8.389925
C	12.760019	3.693384	7.190630
H	12.090943	4.457271	6.780276
C	14.339904	2.969174	8.890950
H	14.917773	3.160198	9.802889
C	13.802467	1.497674	7.029846
H	13.932880	0.543881	6.505983
C	14.509605	1.750322	8.218253
H	15.197335	0.997982	8.621279
C	12.934055	2.472317	6.516382
H	12.382931	2.286039	5.587799
C	13.599421	9.489789	7.785446
H	13.781627	10.560136	7.991902
H	14.491244	8.931548	8.121790

Table S22. $[CpNi^{I}(pdt)Fe^{II}(dppe)(CO)]^{0}$ in Cycle B with pdt Oriented Toward Fe ($[\mathbf{1a}]^{0}$, –3204.37350150 E_h)

atom	x	y	z
Ni	10.267441	8.881138	7.595412
Fe	11.436435	6.729504	8.782279
P	13.287988	5.516831	9.299823
S	12.131492	8.939494	8.758362
P	10.925912	6.747890	10.949665
S	11.035075	7.114724	6.526076
C	7.541099	6.370541	13.349601
H	7.324529	6.301962	14.421959
C	6.493325	6.324422	12.413579
H	5.457178	6.217701	12.754407
C	16.442068	8.051270	10.200992
H	16.587019	8.912253	10.863954
C	12.504357	7.883980	13.087958
H	12.937862	6.891164	13.247618
C	17.490041	7.612412	9.372979
H	18.457519	8.126887	9.387201
C	9.521723	10.794435	6.809274
H	10.139148	11.548596	6.319439
C	17.285056	6.512748	8.524782
H	18.094145	6.162247	7.873214
C	10.409133	5.333152	8.631349
C	8.106857	6.558779	10.605445
H	8.333528	6.641737	9.537419
C	8.855751	9.707503	6.191365
H	8.863281	9.461093	5.127860
C	13.702615	8.165502	6.461897
H	14.529877	8.491585	5.796568
H	14.055503	7.261246	6.987335
C	12.375040	10.227044	13.739051
H	12.700872	11.054826	14.378901
C	9.164419	6.597251	11.535844
C	6.778140	6.421061	11.042771
H	5.965492	6.389276	10.308111
C	16.047067	5.848608	8.508085
H	15.906383	4.988046	7.845722
C	8.164468	8.941433	7.221663
H	7.565970	8.046452	7.042544
O	9.719600	4.377735	8.504505
C	11.682936	5.165842	11.639291
H	11.637002	5.133018	12.743586
H	11.025137	4.361804	11.261927
C	11.402124	10.440521	12.747402
H	10.966227	11.436761	12.610804
C	13.120402	4.972534	11.124218
H	13.437047	3.920827	11.237336
H	13.830195	5.592825	11.699001
C	14.989437	6.273816	9.342264
C	8.867209	6.509358	12.914290
H	9.675438	6.564370	13.653075
C	9.302785	10.664899	8.232067

H	9.723477	11.321976	8.996264
C	8.399604	9.568788	8.474639
H	8.027783	9.241419	9.446001
C	12.924679	8.947286	13.906601
H	13.679046	8.768230	14.681535
C	12.496907	7.806028	5.596796
H	12.774247	7.020740	4.869271
H	12.131508	8.681663	5.028529
C	15.202007	7.395633	10.180100
H	14.387400	7.769169	10.812651
C	11.529528	8.086594	12.087751
C	10.987240	9.382631	11.926817
H	10.230069	9.557053	11.154106
C	13.596355	3.906068	8.419233
C	12.936234	3.666668	7.196479
H	12.254320	4.428283	6.801392
C	14.470043	2.915995	8.924800
H	15.015460	3.089717	9.860037
C	13.997820	1.484078	7.014682
H	14.149445	0.542851	6.474002
C	14.667245	1.713442	8.229446
H	15.346348	0.954699	8.635447
C	13.135695	2.463437	6.498514
H	12.608455	2.290751	5.553405
C	13.452230	9.274879	7.484905
H	13.169769	10.220867	6.986618
H	14.372381	9.455446	8.069031

Table S23. $[CpNi^{I}(pdt)Fe^{II}(dppe)(CO)]^{0}$ Transition State with pdt Oriented Toward Ni ($[\mathbf{1a}]^{0}$, –3204.36221151 E_h)

atom	x	y	z
Ni	10.112252	8.666263	6.812686
Fe	11.466957	7.033514	8.945521
P	12.110353	7.144118	11.082137
S	10.875819	9.298471	8.771078
P	9.536081	6.064507	9.618858
S	11.882235	7.379888	6.628355
C	8.062952	2.286833	8.566730
H	7.678618	1.393865	9.073213
C	8.162623	2.315427	7.166558
H	7.856494	1.444557	6.575897
C	11.591339	10.926415	12.765214
H	10.745128	11.577574	13.012755
C	7.954621	8.292700	10.381377
H	8.909385	8.829900	10.391936
C	12.906747	11.363394	12.988476
H	13.094075	12.356095	13.413048
C	9.453732	9.440969	4.823330
H	10.121685	9.687749	3.996289
C	13.979710	10.517995	12.663153
H	15.011440	10.846527	12.834034
C	12.127798	5.441630	8.720776
C	9.071559	4.569735	7.285728
H	9.494264	5.451969	6.790678
C	8.953836	8.107865	5.135565
H	9.162760	7.209636	4.550367
C	13.043574	9.983037	7.025200
H	12.320082	10.401006	6.299478
H	13.978701	10.576312	6.935628
C	5.535178	8.313415	10.658240
H	4.612518	8.843953	10.919597
C	8.965350	4.558449	8.693525
C	8.667743	3.459857	6.527811
H	8.761848	3.483910	5.436188
C	13.739507	9.243593	12.125483
H	14.586913	8.591604	11.888350
C	8.093274	8.189264	6.276509
H	7.540004	7.371806	6.738950
O	12.568530	4.362480	8.513995
C	9.942741	5.355979	11.329133
H	9.029823	5.045583	11.868939
H	10.566549	4.459222	11.155333
C	5.486626	6.972976	10.242369
H	4.524201	6.452170	10.176169
C	10.719247	6.405008	12.141283
H	11.108731	5.973449	13.080244
H	10.047456	7.237682	12.417737
C	12.419983	8.787624	11.903562
C	8.458795	3.401068	9.325343
H	8.372616	3.360286	10.416768
C	8.987511	10.313997	5.822597

H	9.219762	11.375625	5.924548
C	8.188623	9.531213	6.759132
H	7.703140	9.921288	7.655462
C	6.773548	8.971886	10.724821
H	6.823201	10.022470	11.034256
C	13.357011	8.535716	6.636627
H	14.110349	8.095242	7.314505
H	13.762635	8.502665	5.609312
C	11.350171	9.654951	12.221382
H	10.316008	9.342367	12.045249
C	7.921035	6.938306	9.980434
C	6.667522	6.290528	9.907201
H	6.610498	5.246355	9.582139
C	13.619535	6.203431	11.647383
C	14.610618	5.861110	10.704767
H	14.456240	6.119215	9.651677
C	13.836070	5.873338	13.004408
H	13.103159	6.162027	13.766396
C	15.972757	4.849534	12.448451
H	16.880045	4.318771	12.758558
C	14.998822	5.196007	13.400746
H	15.147150	4.942729	14.456854
C	15.779407	5.189502	11.101046
H	16.534561	4.926467	10.351772
C	12.494609	10.169851	8.442848
H	12.306392	11.241336	8.634523
H	13.211117	9.810146	9.203392

Table S24. $[CpNi^{I}(pdt)Fe^{I}(dppe)(CO)]^{-}$ in Cycle A with pdt Oriented Toward Ni ($[\mathbf{1a}]^{-}$, –3204.41087145 E_h)

atom	x	y	z
Ni	2.425506	5.998244	2.007580
Fe	1.557297	5.969306	4.577002
S	1.043390	7.551304	2.950210
P	2.609388	7.470217	5.790091
O	-0.958394	5.904422	6.096925
C	4.553784	6.319355	1.592047
H	5.255465	6.585545	2.383935
C	3.841808	7.239277	0.753459
H	3.864025	8.328408	0.834521
C	3.047447	6.484400	-0.169551
H	2.368072	6.898010	-0.919407
C	-0.697496	7.194360	2.386745
H	-1.347580	7.192419	3.282905
H	-0.995068	8.052819	1.754486
C	-0.874820	5.891543	1.597518
H	-0.194406	5.906194	0.723805
H	-1.917859	5.859594	1.207640
C	0.081114	5.927198	5.508810
C	3.038481	6.643097	7.429811
C	5.448460	8.093726	5.955717
H	5.488719	7.518821	6.887906
C	6.633072	8.673764	5.467565
H	7.570999	8.537451	6.020877
C	6.614578	9.431966	4.286270
H	7.536982	9.886404	3.905287
C	5.401237	9.596548	3.591771
H	5.375336	10.180600	2.663986
C	4.223406	9.005741	4.068071
H	3.288547	9.116501	3.505711
C	2.416781	9.954870	7.212008
H	3.485216	9.823090	7.421762
C	1.739567	9.007625	6.413127
C	0.376712	9.214458	6.122891
H	-0.144587	8.489109	5.492314
C	-0.297516	10.338888	6.630095
H	-1.357403	10.488031	6.390725
C	0.381582	11.269744	7.432857
H	-0.145762	12.147354	7.827299
C	1.742550	11.075648	7.723551
H	2.282775	11.801052	8.344881
H	3.691002	7.267786	8.068883
H	4.715524	5.345887	6.895616
H	2.066747	6.543954	7.946272
S	1.138915	4.353572	2.953945
C	-0.623004	4.610080	2.402320
H	-1.267673	4.588444	3.302081
H	-0.878339	3.729543	1.781883
C	3.645290	5.260500	7.152165
C	4.176168	3.564750	4.846278
C	5.377001	4.256728	4.562601

H	5.478401	5.311582	4.844294
C	6.434273	3.623492	3.894123
H	7.357785	4.179693	3.692835
C	6.303488	2.291199	3.461901
H	7.126431	1.798857	2.929889
C	5.100253	1.606815	3.698844
H	4.977144	0.573633	3.350789
C	4.047461	2.235713	4.384173
H	3.116307	1.689025	4.564186
C	2.579991	2.182102	7.396965
H	3.645199	2.350201	7.597265
C	1.897470	3.014405	6.479951
C	0.542956	2.744127	6.205433
H	0.016335	3.379078	5.487659
C	-0.118342	1.677569	6.839000
H	-1.173241	1.483042	6.610830
C	0.565568	0.866465	7.759981
H	0.047408	0.037624	8.258232
C	1.919197	1.120208	8.036762
H	2.464899	0.487637	8.748274
H	3.566806	4.597625	8.032749
C	4.202832	4.989733	1.178262
H	4.570516	4.062620	1.620959
C	3.273745	5.097159	0.091352
H	2.787284	4.259637	-0.415175
C	4.223646	8.249909	5.266922
P	2.754480	4.466454	5.659336

Table S25. $[CpNi^{I}(pdt)Fe^{I}(dppe)(CO)]^{-}$ in Cycle A with pdt Oriented Toward Fe ($[\mathbf{1a}]^{-}$, –3204.40706696 E_h)

atom	x	y	z
Ni	10.410532	8.648857	7.356811
Fe	11.803418	6.711890	8.773338
P	11.414786	6.759169	10.938595
S	12.097277	9.044302	8.856538
P	10.593560	4.860355	8.731470
S	11.663863	6.963443	6.482931
C	10.683791	1.150964	6.797510
H	10.050350	0.256143	6.750140
C	11.894116	1.197843	6.085469
H	12.210302	0.339367	5.479679
C	8.235837	9.332102	11.625085
H	7.445243	9.780015	11.011320
C	7.799229	4.451176	9.437882
H	8.127733	4.097160	10.421514
C	8.279540	9.574010	13.007659
H	7.521024	10.209631	13.481427
C	9.584265	10.709659	6.738283
H	10.195549	11.615298	6.764603
C	9.303748	9.002366	13.782235
H	9.346957	9.188902	14.862860
C	13.369860	5.947309	8.958519
C	12.287910	3.453700	6.920561
H	12.899397	4.359306	6.963530
C	9.449078	9.814800	5.633437
H	9.925296	9.919668	4.655145
C	14.128978	8.433031	6.895300
H	15.013903	8.852149	6.364699
H	14.505260	7.630413	7.552975
C	5.963299	4.953992	7.916116
H	4.888291	5.006430	7.706059
C	11.078034	3.414942	7.642705
C	12.693060	2.351334	6.148384
H	13.634527	2.400044	5.587792
C	10.275749	8.190580	13.175630
H	11.074874	7.753015	13.785177
C	8.596696	8.735095	6.040484
H	8.310972	7.880883	5.423043
O	14.433323	5.418886	9.070023
C	10.795706	4.052533	10.430033
H	10.106016	3.199153	10.572994
H	11.824616	3.649066	10.421765
C	6.899271	5.330027	6.933578
H	6.554559	5.678304	5.952447
C	10.633033	5.122220	11.518827
H	11.042732	4.806328	12.496467
H	9.564109	5.354477	11.673952
C	10.236769	7.941440	11.786061
C	10.278332	2.253187	7.567917
H	9.323078	2.218014	8.105288
C	8.821559	10.183409	7.831125

H	8.750027	10.628302	8.826069
C	8.210374	8.957127	7.402626
H	7.543571	8.320861	7.988212
C	6.420885	4.519948	9.170753
H	5.703671	4.223916	9.946897
C	13.194360	7.829367	5.840944
H	13.753641	7.085546	5.240752
H	12.833031	8.616574	5.151502
C	9.209619	8.522303	11.015728
H	9.197399	8.350117	9.930577
C	8.749896	4.825355	8.460289
C	8.272647	5.278747	7.204552
H	8.992625	5.601704	6.443507
C	12.922614	6.951942	12.007868
C	13.468402	8.243646	12.201899
H	12.954145	9.111646	11.775064
C	13.619894	5.842127	12.529960
H	13.239955	4.827303	12.364869
C	15.341598	7.300760	13.444811
H	16.276280	7.435649	14.002731
C	14.815727	6.013378	13.246658
H	15.340522	5.135806	13.643811
C	14.665742	8.412656	12.914407
H	15.073226	9.421578	13.054426
C	13.522471	9.545222	7.757038
H	13.153490	10.370017	7.117264
H	14.306075	9.956375	8.422260

Table S26. $[CpNi^{III}(pdt)Fe^{II}(dppe)(CO)]^{2+}$ in Cycle A with pdt Oriented Toward Ni ($[\mathbf{1a}]^{2+}$, –3203.88969329 E_h)

atom	x	y	z
Ni	10.262166	8.706945	7.461582
Fe	11.519647	6.919971	8.753215
P	11.241399	6.810031	11.104792
S	11.898147	9.150931	8.856414
P	10.524108	4.797063	8.733090
S	11.385341	7.082263	6.499479
C	11.098703	1.144697	6.938001
H	10.536833	0.208308	6.860020
C	12.368705	1.259368	6.346144
H	12.798601	0.409940	5.805661
C	8.099892	9.272133	12.119785
H	7.046263	9.368780	11.836110
C	7.682770	4.894698	9.085683
H	7.847202	4.856431	10.166750
C	8.640089	10.103060	13.118651
H	8.009308	10.853425	13.606076
C	9.303165	10.590799	7.203014
H	9.791188	11.559742	7.322290
C	9.984122	9.960231	13.498197
H	10.404101	10.595283	14.284840
C	13.194720	6.415417	8.836930
C	12.534307	3.554494	7.136745
H	13.103780	4.484450	7.206261
C	9.184418	9.846043	6.002635
H	9.553954	10.130848	5.016450
C	13.250501	9.264373	6.314047
H	12.454874	9.916328	5.909525
H	14.195616	9.582934	5.829158
C	6.113049	4.927083	7.222741
H	5.082976	4.938486	6.851964
C	11.261289	3.445290	7.734708
C	13.083649	2.463272	6.444485
H	14.071699	2.556328	5.982273
C	10.796092	8.990297	12.887768
H	11.833607	8.872002	13.213141
C	8.497322	8.607492	6.329442
H	8.271447	7.804972	5.625826
O	14.333111	6.152634	8.845185
C	10.539522	4.147854	10.488691
H	9.809308	3.323970	10.583777
H	11.544485	3.708125	10.623278
C	7.188568	4.888823	6.316705
H	6.997346	4.855740	5.238712
C	10.278278	5.260907	11.510012
H	10.503133	4.939030	12.542250
H	9.220733	5.573703	11.510656
C	10.259787	8.154627	11.884128
C	10.540604	2.230459	7.626827
H	9.543316	2.131820	8.069488
C	8.672524	9.825572	8.275309

H	8.618353	10.134602	9.320583
C	8.132471	8.628700	7.717948
H	7.586324	7.844765	8.243184
C	6.362668	4.924676	8.605642
H	5.528796	4.923665	9.315403
C	12.996695	7.815452	5.889255
H	13.802629	7.138875	6.220951
H	12.924131	7.747787	4.790272
C	8.906159	8.308679	11.497037
H	8.475770	7.666050	10.718822
C	8.769327	4.861141	8.181303
C	8.509438	4.859741	6.788509
H	9.337781	4.793682	6.074567
C	12.852218	6.713407	11.974979
C	13.761388	7.793330	11.856273
H	13.492908	8.678883	11.268402
C	13.222462	5.580540	12.733890
H	12.548831	4.725454	12.844637
C	15.362689	6.616899	13.259989
H	16.335051	6.579579	13.761319
C	14.473565	5.536540	13.370013
H	14.748068	4.654663	13.957616
C	15.003359	7.746741	12.504572
H	15.692278	8.593446	12.419638
C	13.412066	9.516118	7.815647
H	13.613591	10.585123	8.001169
H	14.242475	8.931362	8.247331

Table S27. $[CpNi^{III}(pdt)Fe^{II}(dppe)(CO)]^{2+}$ in Cycle A with pdt Oriented Toward Fe ($[\mathbf{1a}]^{2+}$, –3203.88593808 E_h)

atom	x	y	z
Ni	10.273744	8.686062	7.511060
Fe	11.582654	6.897311	8.753160
P	11.254326	6.813773	11.104627
S	11.955462	9.134576	8.839949
P	10.526985	4.801665	8.712185
S	11.388536	7.095062	6.501880
C	11.155897	1.119166	7.004479
H	10.633977	0.156816	6.997304
C	12.371554	1.270342	6.314467
H	12.799679	0.422914	5.769478
C	8.107450	9.269514	12.131601
H	7.046244	9.348602	11.871946
C	7.683936	4.897164	9.049782
H	7.842253	4.873310	10.132233
C	8.663487	10.129772	13.096383
H	8.036858	10.884676	13.582156
C	9.331994	10.578913	7.269474
H	9.833237	11.541250	7.387758
C	10.018565	10.010785	13.443707
H	10.451772	10.669176	14.203508
C	13.213729	6.256997	8.915650
C	12.486792	3.596563	7.018617
H	13.012023	4.554929	7.012829
C	9.196574	9.838391	6.068178
H	9.564214	10.121027	5.080637
C	13.905365	8.536023	6.758819
H	14.692447	9.019700	6.144968
H	14.416743	7.747596	7.333604
C	6.124896	4.898270	7.177714
H	5.096874	4.900029	6.801157
C	11.270817	3.449750	7.718177
C	13.033061	2.508079	6.319385
H	13.977279	2.629560	5.778780
C	10.824816	9.034379	12.836464
H	11.870998	8.933981	13.139031
C	8.491043	8.610235	6.396371
H	8.249209	7.812353	5.692721
O	14.297602	5.838988	9.031528
C	10.536858	4.158201	10.469652
H	9.805044	3.335983	10.565768
H	11.540674	3.718323	10.611864
C	7.205454	4.855502	6.278194
H	7.020744	4.810003	5.199577
C	10.268828	5.273793	11.486682
H	10.470368	4.947141	12.522068
H	9.214453	5.595821	11.468609
C	10.272140	8.167958	11.867853
C	10.600830	2.201587	7.700580
H	9.643240	2.073882	8.217602
C	8.694205	9.821768	8.342756

H	8.649571	10.129649	9.388656
C	8.133270	8.635674	7.785712
H	7.580145	7.857733	8.312230
C	6.366609	4.913146	8.562006
H	5.528668	4.915543	9.266959
C	12.910723	7.909254	5.780214
H	13.402553	7.112632	5.194772
H	12.509663	8.649781	5.065755
C	8.908063	8.300224	11.510773
H	8.465027	7.639107	10.756007
C	8.775314	4.858490	8.151528
C	8.523759	4.837955	6.757692
H	9.356280	4.768042	6.049085
C	12.845435	6.706592	12.009698
C	13.785061	7.757092	11.867199
H	13.555012	8.620544	11.232186
C	13.167766	5.599219	12.825768
H	12.470593	4.766228	12.956380
C	15.321176	6.603790	13.359846
H	16.280315	6.563739	13.885825
C	14.401855	5.551873	13.493968
H	14.639484	4.689451	14.124902
C	15.009530	7.708089	12.547656
H	15.722675	8.532161	12.442552
C	13.368817	9.609087	7.707895
H	12.998181	10.495664	7.163541
H	14.169822	9.945942	8.389084

Table S28. $[CpNi^{III}(pdt)HFe^{II}(dppe)(CO)]^+$ in Cycle A with pdt Oriented Toward Ni ($[H\mathbf{1a}]^+$, –3204.79612707 E_h)

atom	x	y	z
Ni	2.523314	5.994772	2.044962
Fe	1.735917	5.969888	4.520335
S	1.135799	7.505895	2.905084
P	2.665693	7.527218	5.848970
O	-0.832560	5.880103	5.959476
C	4.632773	6.204597	1.500013
H	5.417140	6.375151	2.236683
C	3.832640	7.212716	0.876185
H	3.932736	8.289058	1.021163
C	2.895367	6.565849	-0.027804
H	2.167760	7.075335	-0.660855
C	-0.595558	7.155533	2.327350
H	-1.237322	7.161462	3.227180
H	-0.872175	8.021249	1.700794
C	-0.778895	5.856013	1.538497
H	-0.150238	5.879211	0.628976
H	-1.833271	5.810419	1.198401
C	0.208097	5.910544	5.412489
C	3.096924	6.646685	7.444955
C	5.494734	8.059142	5.860635
H	5.580412	7.399315	6.730205
C	6.656491	8.677351	5.363925
H	7.621270	8.480232	5.843965
C	6.577431	9.555768	4.273043
H	7.481047	10.044593	3.893726
C	5.329339	9.819825	3.680970
H	5.257104	10.525398	2.845638
C	4.170186	9.199167	4.168991
H	3.199902	9.424327	3.710835
C	2.365449	9.967169	7.233357
H	3.432855	9.865930	7.460364
C	1.713223	9.001269	6.431935
C	0.348026	9.170712	6.126415
H	-0.168214	8.437531	5.501843
C	-0.357621	10.279865	6.621590
H	-1.418383	10.400597	6.376976
C	0.293670	11.228040	7.424739
H	-0.257335	12.092495	7.810205
C	1.656680	11.069982	7.729703
H	2.171583	11.810438	8.351217
H	3.742815	7.271091	8.088297
H	4.785902	5.367691	6.877421
H	2.130687	6.532007	7.969573
S	1.267956	4.372538	2.911630
C	-0.487051	4.581730	2.336149
H	-1.125837	4.529543	3.236716
H	-0.694021	3.692542	1.715459
C	3.724601	5.272850	7.162285
C	4.185677	3.512948	4.845613
C	5.394607	4.173804	4.530022

H	5.550839	5.217964	4.827054
C	6.408548	3.505810	3.826469
H	7.350327	4.022178	3.609567
C	6.219475	2.177117	3.406095
H	7.012862	1.655396	2.860212
C	5.012469	1.521259	3.695725
H	4.857293	0.485931	3.373476
C	4.001597	2.181778	4.414752
H	3.072208	1.653664	4.649659
C	2.531326	2.243803	7.522250
H	3.579823	2.420540	7.788058
C	1.893651	3.046792	6.546827
C	0.559913	2.758687	6.194297
H	0.057236	3.357273	5.430638
C	-0.131789	1.706124	6.816928
H	-1.170046	1.499257	6.536380
C	0.503389	0.927656	7.795716
H	-0.037363	0.110388	8.284606
C	1.838525	1.195701	8.144138
H	2.344116	0.584979	8.899774
H	3.682218	4.628832	8.056816
C	4.119003	4.937915	1.090686
H	4.468231	3.962201	1.432292
C	3.066745	5.168997	0.110139
H	2.491124	4.390696	-0.393093
C	4.240110	8.307118	5.264586
P	2.832016	4.418676	5.731836
H	3.136123	6.048236	3.738760

Table S29. $[CpNi^{III}(pdt)HFe^{II}(dppe)(CO)]^+$ in Cycle A with pdt Oriented Toward Fe ($[H\mathbf{1a}]^+$, –3204.79478996 E_h)

atom	x	y	z
Ni	10.093192	8.677953	7.623824
Fe	11.529079	6.860436	8.806726
P	11.268926	6.812053	11.043321
S	11.896346	9.154630	8.840928
P	10.555674	4.837979	8.710876
S	11.268119	7.153491	6.520527
C	11.440298	1.057248	7.283247
H	11.033140	0.048028	7.407115
C	12.554698	1.273069	6.453899
H	13.020413	0.430308	5.932076
C	8.307480	9.474213	12.165590
H	7.270149	9.668765	11.870379
C	7.692934	4.867722	8.872981
H	7.800091	4.921927	9.961478
C	8.897906	10.220630	13.200220
H	8.324993	11.003153	13.708952
C	9.304868	10.642731	7.131291
H	9.911093	11.549772	7.122693
C	10.221212	9.953743	13.581822
H	10.686837	10.526702	14.390827
C	13.181879	6.218207	8.900424
C	12.471659	3.650571	6.965037
H	12.874940	4.658528	6.834987
C	9.074475	9.771022	6.042288
H	9.466725	9.875431	5.029699
C	13.759916	8.608189	6.700817
H	14.560473	9.068390	6.086811
H	14.236196	7.797311	7.276097
C	6.228239	4.771536	6.929764
H	5.220609	4.758105	6.500841
C	11.359431	3.442566	7.804904
C	13.065713	2.569486	6.292473
H	13.930999	2.743490	5.643923
C	10.955688	8.942092	12.940561
H	11.980140	8.733432	13.262607
C	8.257647	8.667493	6.524436
H	7.929273	7.811685	5.932933
O	14.268322	5.782029	8.985086
C	10.433063	4.186456	10.470851
H	9.652572	3.408986	10.551006
H	11.406536	3.702089	10.669883
C	7.353830	4.680045	6.093309
H	7.227423	4.587766	5.008916
C	10.185888	5.337291	11.456733
H	10.321748	5.024843	12.506898
H	9.151640	5.714916	11.377244
C	10.373404	8.192174	11.897772
C	10.841135	2.134366	7.950748
H	9.960097	1.954376	8.577151
C	8.629356	10.091377	8.296707

H	8.642592	10.523594	9.298267
C	7.927237	8.915368	7.891895
H	7.328195	8.262061	8.526387
C	6.401108	4.860891	8.320633
H	5.528961	4.910707	8.981870
C	12.717791	8.025097	5.746706
H	13.184214	7.263242	5.095811
H	12.292404	8.804781	5.088658
C	9.043248	8.474438	11.512162
H	8.579687	7.909000	10.695366
C	8.829878	4.779315	8.040723
C	8.645884	4.682310	6.642632
H	9.514423	4.588215	5.982327
C	12.825550	6.609083	12.013141
C	13.851581	7.565645	11.828768
H	13.699435	8.397332	11.130719
C	13.045532	5.537512	12.903859
H	12.277143	4.775808	13.069093
C	15.269324	6.387711	13.415490
H	16.216328	6.299899	13.958253
C	14.263285	5.426627	13.595529
H	14.420912	4.585649	14.279186
C	15.059388	7.460633	12.532740
H	15.841006	8.213679	12.385690
C	13.229221	9.678018	7.655264
H	12.828134	10.547459	7.102892
H	14.047270	10.046575	8.300578
H	9.994060	7.358191	8.838881

Table S30. $[CpNi^{III}(pdt)HFe^{II}(dppe)(CO)]^+$ in Cycle B with pdt Oriented Toward Ni ($[H\mathbf{1a}]^+$, –3204.79237812 E_h)

atom	x	y	z
Ni	2.510082	5.989219	2.091223
Fe	1.891500	6.233924	4.596869
S	1.343538	7.707653	2.899931
P	2.851650	4.742845	5.997903
O	3.458262	8.378735	5.812292
C	4.605970	5.690097	1.511227
H	5.416354	5.571215	2.231051
C	4.098099	6.924805	1.003010
H	4.474355	7.922432	1.235720
C	3.018452	6.627480	0.073887
H	2.444919	7.368201	-0.485165
C	-0.421857	7.537937	2.342482
H	-1.055187	7.622878	3.243521
H	-0.601353	8.423764	1.708283
C	-0.739841	6.257258	1.570762
H	-0.093839	6.190970	0.675600
H	-1.785582	6.322781	1.208105
C	2.818836	7.508187	5.351600
C	2.281998	5.252941	7.708292
C	1.720727	2.232441	6.822326
H	1.368517	2.729180	7.732163
C	1.459662	0.861184	6.654943
H	0.902422	0.318876	7.426505
C	1.923237	0.188491	5.514251
H	1.723052	-0.880710	5.387813
C	2.654932	0.888971	4.540042
H	3.029278	0.366340	3.653210
C	2.913617	2.256950	4.702233
H	3.498027	2.792369	3.945508
C	5.280191	3.747841	7.030090
H	4.651700	3.012341	7.544820
C	4.694490	4.703484	6.167425
C	5.527870	5.620078	5.493953
H	5.092268	6.363662	4.820712
C	6.919267	5.589762	5.686824
H	7.555143	6.311121	5.162145
C	7.491266	4.642777	6.549954
H	8.575984	4.619867	6.699199
C	6.668947	3.721311	7.220541
H	7.109667	2.976670	7.892019
H	2.571127	4.504011	8.467457
H	0.193251	4.594209	7.791809
H	2.852596	6.172864	7.930688
S	1.073359	4.615159	3.097516
C	-0.613359	4.982850	2.408033
H	-1.306429	5.008253	3.265209
H	-0.859887	4.098970	1.794254
C	0.770668	5.531808	7.726808
C	-1.436312	5.607011	5.823110
C	-1.511181	4.195875	5.756415

H	-0.616725	3.582453	5.918900
C	-2.727682	3.561727	5.461051
H	-2.771976	2.467847	5.422573
C	-3.881758	4.324806	5.213256
H	-4.830525	3.828028	4.984295
C	-3.813021	5.726176	5.262748
H	-4.708780	6.328245	5.075265
C	-2.599171	6.366075	5.565097
H	-2.563886	7.459124	5.619742
C	-1.076188	8.249343	7.905900
H	-1.427987	7.365458	8.450396
C	-0.281625	8.104870	6.743126
C	0.120853	9.260979	6.043550
H	0.730188	9.168557	5.139131
C	-0.249347	10.536572	6.502599
H	0.076941	11.425421	5.952155
C	-1.025873	10.669874	7.662958
H	-1.309637	11.664353	8.023429
C	-1.441675	9.523253	8.362492
H	-2.054248	9.620667	9.265243
H	0.497848	6.145037	8.602505
C	3.774277	4.645471	1.000425
H	3.863593	3.582105	1.227974
C	2.820052	5.227450	0.070504
H	2.068273	4.675776	-0.495343
C	2.448824	2.945953	5.845990
P	0.198021	6.418653	6.149933
H	3.189950	5.962249	3.746230

Table S31. $[CpNi^{III}(pdt)HFe^{II}(dppe)(CO)]^+$ in Cycle B with pdt Oriented Toward Fe ($[H\mathbf{1a}]^+$, –3204.79069048 E_h)

atom	x	y	z
Ni	10.226775	8.766062	7.619574
Fe	11.327749	6.745687	8.820907
P	13.240841	5.523048	9.312295
S	12.142555	8.941542	8.746457
P	10.905525	6.726311	11.038409
S	11.084581	7.042774	6.513796
C	7.588193	6.526418	13.500920
H	7.402872	6.575167	14.579282
C	6.521146	6.326770	12.608678
H	5.500337	6.217197	12.989903
C	16.336037	8.117280	10.225812
H	16.472912	8.972493	10.896751
C	12.558522	7.858555	13.098964
H	12.967876	6.858291	13.272817
C	17.395509	7.680493	9.411684
H	18.362042	8.194487	9.444592
C	9.711311	10.772262	6.948921
H	10.444543	11.545407	6.715686
C	17.209917	6.581184	8.558979
H	18.031458	6.232158	7.923977
C	10.385896	5.282766	8.649990
C	8.075917	6.412090	10.737176
H	8.254618	6.364778	9.659396
C	9.165551	9.824853	6.051672
H	9.400380	9.721819	4.991165
C	13.747190	8.105434	6.480734
H	14.570349	8.438807	5.816783
H	14.116563	7.212980	7.011704
C	12.489442	10.213448	13.715143
H	12.841151	11.041289	14.339818
C	9.153556	6.607861	11.625742
C	6.767009	6.270773	11.228201
H	5.939779	6.113627	10.527537
C	15.973583	5.914632	8.518878
H	15.845650	5.053740	7.854938
C	8.258242	8.967640	6.799981
H	7.695314	8.130087	6.384667
O	9.735901	4.317109	8.497102
C	11.669229	5.138082	11.668187
H	11.643480	5.092281	12.771928
H	11.002553	4.338459	11.297630
C	11.505477	10.437017	12.736829
H	11.086311	11.439542	12.598962
C	13.093718	4.961565	11.120877
H	13.414709	3.909161	11.201999
H	13.815880	5.569532	11.691641
C	14.906007	6.343801	9.336394
C	8.895764	6.668664	13.015064
H	9.716494	6.840792	13.720333
C	9.150790	10.504401	8.263107

H	9.385521	11.060811	9.171891
C	8.202989	9.439627	8.146452
H	7.618674	9.007431	8.959541
C	13.012516	8.924101	13.895671
H	13.769549	8.738871	14.665348
C	12.556580	7.722714	5.609036
H	12.841921	6.920275	4.904723
H	12.191428	8.577545	5.010962
C	15.097182	7.459144	10.185485
H	14.278494	7.819769	10.820050
C	11.574923	8.074596	12.109776
C	11.053475	9.377575	11.938292
H	10.276075	9.556468	11.187001
C	13.529131	3.955572	8.374311
C	12.875712	3.737032	7.143676
H	12.187981	4.493736	6.752110
C	14.413966	2.965824	8.864965
H	14.957182	3.122104	9.803983
C	13.960176	1.570305	6.923529
H	14.122593	0.642325	6.365055
C	14.624227	1.781353	8.144476
H	15.309185	1.022242	8.537229
C	13.090048	2.550354	6.423092
H	12.568558	2.391242	5.473203
C	13.479760	9.225264	7.486198
H	13.215115	10.169519	6.976061
H	14.387137	9.409171	8.087869
H	9.957179	7.528568	8.851672

Table S32. $[CpNi^{III}(pdt)HFe^{II}(dppe)(CO)]^+$ Transition State with pdt Oriented Toward Ni ($[H\mathbf{1a}]^+$, –3204.75933368 E_h)

atom	x	y	z
Ni	10.309093	8.679596	7.042980
Fe	11.313257	6.978439	8.939014
P	11.912875	7.131780	11.131083
S	11.619292	9.319790	8.711672
P	9.489257	5.897493	9.658602
S	11.708922	7.027288	6.568988
C	7.998461	2.255981	8.311845
H	7.719678	1.289312	8.744641
C	7.876794	2.472611	6.930589
H	7.501655	1.674739	6.281107
C	10.772613	10.645033	13.051008
H	9.841323	11.094224	13.412887
C	8.029189	8.248105	10.283529
H	8.958100	8.777672	10.042191
C	11.990675	11.326775	13.204138
H	12.013576	12.310877	13.684111
C	9.740729	9.592106	5.068334
H	10.361054	9.591113	4.170857
C	13.179026	10.738052	12.743825
H	14.134756	11.259618	12.864037
C	12.051450	5.398635	8.775470
C	8.737480	4.730355	7.211951
H	9.048110	5.685804	6.775237
C	8.869873	8.501814	5.491779
H	8.701525	7.580095	4.931916
C	13.552754	9.228503	6.574757
H	12.860895	9.792677	5.922163
H	14.581021	9.533105	6.292064
C	5.704681	8.289632	11.004849
H	4.823294	8.849040	11.335992
C	8.855241	4.525442	8.604226
C	8.247761	3.711519	6.382176
H	8.166168	3.880712	5.303088
C	13.152927	9.471613	12.139495
H	14.090276	9.015055	11.805236
C	8.210761	8.886164	6.702115
H	7.475510	8.303372	7.257023
O	12.526160	4.338803	8.589736
C	9.999119	5.065417	11.264041
H	9.118497	4.646809	11.782720
H	10.675143	4.233337	10.996617
C	5.638655	6.892922	10.862367
H	4.705457	6.361747	11.078325
C	10.709068	6.096867	12.151155
H	11.241146	5.607731	12.984567
H	9.972689	6.791129	12.590929
C	11.932691	8.776025	11.985884
C	8.484305	3.274974	9.147251
H	8.576558	3.080660	10.220622
C	9.688840	10.589559	6.055445

H	10.259423	11.519232	6.076584
C	8.782661	10.133689	7.105175
H	8.534467	10.692793	8.008530
C	6.900111	8.966442	10.714253
H	6.954475	10.055765	10.819254
C	13.410400	7.730379	6.298567
H	14.106579	7.139951	6.921587
H	13.638037	7.514623	5.239804
C	10.740578	9.382312	12.439539
H	9.775596	8.878610	12.319571
C	7.973051	6.845077	10.148467
C	6.764747	6.172286	10.436321
H	6.695902	5.085804	10.311354
C	13.589049	6.464191	11.552513
C	14.564991	6.289661	10.550233
H	14.314395	6.515276	9.507954
C	13.928415	6.179926	12.895499
H	13.202833	6.345527	13.699919
C	16.168558	5.520912	12.208664
H	17.166100	5.147605	12.463315
C	15.207307	5.703913	13.217900
H	15.454855	5.479651	14.260965
C	15.848430	5.820924	10.876027
H	16.594556	5.684202	10.085962
C	13.328372	9.631581	8.033658
H	13.486932	10.717149	8.155171
H	14.034288	9.106472	8.702360
H	9.924217	7.405922	8.208524

Table S33. $[CpNi^{III}(pdt)HFe^{II}(dppe)(CO)]^+$ Transition State with pdt Oriented Toward Fe ($[H\mathbf{1a}]^+$, –3204.76094325 E_h)

atom	x	y	z
Ni	10.253148	8.702348	7.076186
Fe	11.340618	6.991811	8.917689
P	11.937112	7.132018	11.114491
S	11.609695	9.345595	8.700876
P	9.521964	5.892238	9.631650
S	11.657961	7.083127	6.532500
C	8.120259	2.224981	8.262907
H	7.886861	1.242191	8.686115
C	7.953607	2.460033	6.889488
H	7.588996	1.660274	6.236340
C	10.890466	10.690315	13.017046
H	9.981302	11.150022	13.419674
C	8.051391	8.215483	10.328710
H	8.985431	8.754243	10.131242
C	12.114899	11.373597	13.083513
H	12.165059	12.369838	13.535499
C	9.672761	9.641123	5.143587
H	10.291442	9.659683	4.245161
C	13.276032	10.770287	12.573623
H	14.237369	11.292627	12.628142
C	12.085489	5.419234	8.720175
C	8.741340	4.742213	7.186988
H	9.003682	5.715926	6.758765
C	8.791373	8.550051	5.539090
H	8.618376	7.641796	4.959066
C	14.001246	8.555365	7.316674
H	14.985811	8.917207	6.957647
H	14.200248	7.823772	8.121777
C	5.711833	8.229346	11.001042
H	4.824900	8.774930	11.340344
C	8.905104	4.517752	8.571812
C	8.265132	3.720583	6.353087
H	8.146894	3.904978	5.279908
C	13.214984	9.488835	12.005469
H	14.132154	9.019220	11.634005
C	8.139050	8.908737	6.760515
H	7.404732	8.314725	7.304568
O	12.544290	4.358730	8.498938
C	10.029390	5.073761	11.243735
H	9.150732	4.637442	11.751083
H	10.731729	4.258120	10.993370
C	5.645308	6.841871	10.787649
H	4.706294	6.303972	10.956678
C	10.698363	6.133350	12.128804
H	11.204569	5.673009	12.994069
H	9.938951	6.832983	12.518304
C	11.987079	8.791755	11.937307
C	8.592773	3.246612	9.102962
H	8.719441	3.038811	10.170179
C	9.626767	10.616962	6.155956

H	10.202400	11.542881	6.197394
C	8.714035	10.145007	7.191268
H	8.470809	10.682256	8.108970
C	6.915385	8.915363	10.770907
H	6.970265	9.997914	10.931300
C	13.303312	7.855630	6.150815
H	13.923509	7.020718	5.778175
H	13.133769	8.552297	5.309245
C	10.823283	9.411520	12.441228
H	9.852901	8.906839	12.391356
C	7.994563	6.820826	10.123314
C	6.778938	6.138712	10.351692
H	6.711759	5.059412	10.174699
C	13.576841	6.423138	11.619165
C	14.549237	6.049287	10.671222
H	14.336738	6.155988	9.603681
C	13.875234	6.285814	12.995844
H	13.151621	6.605297	13.754637
C	16.067383	5.381008	12.450574
H	17.030436	4.971525	12.773383
C	15.109191	5.762879	13.406083
H	15.324635	5.657486	14.474782
C	15.787975	5.529878	11.084348
H	16.530858	5.238209	10.334305
C	13.237580	9.745957	7.894714
H	13.036950	10.511755	7.123456
H	13.825903	10.222922	8.698956
H	9.924117	7.426988	8.232187

Table S34. $[CpNi^{II}(pdt)HFe^{II}(dppe)(CO)]^0$ in Cycle A with pdt Oriented Toward Ni ($[H\mathbf{1a}]^0$, –3204.96518567 E_h)

atom	x	y	z
Ni	2.459552	6.042756	1.605011
Fe	1.836147	5.971505	4.553767
S	1.329295	7.481681	2.832344
P	2.685323	7.486714	5.908894
O	-0.807368	5.845126	5.865925
C	4.480742	6.342023	0.978953
H	5.267611	6.578298	1.695272
C	3.605875	7.282942	0.334336
H	3.639120	8.367986	0.455726
C	2.736418	6.572138	-0.588884
H	1.986248	7.028513	-1.236723
C	-0.461418	7.196372	2.399469
H	-1.035469	7.222301	3.343312
H	-0.769966	8.060901	1.784142
C	-0.730221	5.897204	1.629734
H	-0.189765	5.938254	0.664732
H	-1.815821	5.848296	1.398466
C	0.241904	5.891814	5.317955
C	3.139556	6.585157	7.493279
C	5.470049	8.237475	6.122438
H	5.518843	7.661436	7.052640
C	6.631486	8.890034	5.671080
H	7.558087	8.808249	6.251072
C	6.602118	9.646112	4.490390
H	7.505739	10.158065	4.140795
C	5.405175	9.743770	3.759267
H	5.371298	10.335836	2.837535
C	4.249180	9.085109	4.201697
H	3.324084	9.156534	3.617955
C	2.327906	9.870516	7.428751
H	3.382818	9.756183	7.704725
C	1.723527	8.943422	6.550036
C	0.378596	9.133139	6.177159
H	-0.093519	8.426473	5.489649
C	-0.354215	10.219891	6.683493
H	-1.399744	10.354936	6.384205
C	0.250037	11.128135	7.565782
H	-0.321708	11.975896	7.960175
C	1.594249	10.951860	7.937437
H	2.074594	11.662343	8.620041
H	3.754039	7.201247	8.175165
H	4.844396	5.438033	6.742731
H	2.174625	6.398792	8.000347
S	1.473916	4.466366	2.786968
C	-0.336780	4.606994	2.360569
H	-0.905757	4.499813	3.302116
H	-0.566231	3.736745	1.719115
C	3.826383	5.258736	7.132061
C	4.188283	3.426584	4.888779
C	5.296586	4.108608	4.337886

H	5.352731	5.201334	4.406329
C	6.315578	3.402973	3.681383
H	7.173176	3.947004	3.269183
C	6.231725	2.006400	3.542610
H	7.024499	1.456112	3.023233
C	5.124232	1.323450	4.066877
H	5.044571	0.235930	3.956167
C	4.110078	2.027066	4.739112
H	3.254303	1.480518	5.146997
C	2.423745	2.366523	7.704149
H	3.440689	2.568322	8.059124
C	1.891429	3.090174	6.612686
C	0.599200	2.762165	6.153591
H	0.182554	3.303864	5.300026
C	-0.151062	1.753258	6.779559
H	-1.155276	1.516802	6.410319
C	0.380175	1.056751	7.875799
H	-0.207495	0.274721	8.369783
C	1.672179	1.363322	8.334824
H	2.099331	0.817480	9.183920
H	3.919820	4.599377	8.011764
C	4.028389	5.046718	0.598365
H	4.426994	4.094624	0.953404
C	2.971542	5.199575	-0.402375
H	2.441624	4.374615	-0.882931
C	4.265331	8.323655	5.393241
P	2.859476	4.417516	5.736038
H	3.241933	6.054995	3.919996

Table S35. $[CpNi^{II}(pdt)HFe^{II}(dppe)(CO)]^0$ in Cycle A with pdt Oriented Toward Fe ($[H\mathbf{1a}]^0$, –3204.96246943 E_h)

atom	x	y	z
Ni	10.311748	9.055122	7.210982
Fe	11.524935	6.725881	8.826819
P	11.294318	6.742363	11.012568
S	11.898360	9.049312	8.728527
P	10.588237	4.734572	8.789334
S	11.192428	7.160939	6.535917
C	11.059342	1.042737	6.928037
H	10.541980	0.076413	6.927202
C	12.211721	1.227459	6.144613
H	12.596975	0.404001	5.532542
C	8.339702	9.556459	11.810893
H	7.587363	10.108050	11.235434
C	7.826281	4.126175	9.378581
H	8.149943	3.766013	10.360909
C	8.421253	9.720942	13.202431
H	7.733074	10.400328	13.718343
C	9.824335	11.148412	6.623055
H	10.525447	11.984680	6.652428
C	9.392066	9.014063	13.931681
H	9.464105	9.138417	15.018347
C	13.184007	6.100084	8.911302
C	12.368117	3.527277	6.926679
H	12.874747	4.496529	6.925464
C	9.548040	10.316740	5.523157
H	9.991400	10.376594	4.527631
C	13.856331	8.276548	6.731394
H	14.698313	8.671004	6.123612
H	14.266988	7.437267	7.313424
C	6.008848	4.526632	7.808661
H	4.942517	4.487402	7.559204
C	11.218343	3.349185	7.721191
C	12.862443	2.470624	6.143522
H	13.757579	2.623142	5.529943
C	10.276207	8.147409	13.271458
H	11.036720	7.608959	13.847826
C	8.614520	9.294376	5.973712
H	8.224997	8.483891	5.354135
O	14.245244	5.603532	9.080900
C	10.788175	4.016857	10.517787
H	10.169736	3.114339	10.675458
H	11.846850	3.700806	10.566340
C	6.935978	5.024954	6.876673
H	6.595070	5.373402	5.895021
C	10.491926	5.115241	11.551946
H	10.807264	4.833756	12.572742
H	9.408613	5.329223	11.590465
C	10.198309	7.974361	11.870938
C	10.562485	2.097694	7.707120
H	9.649200	1.951690	8.295871
C	9.046764	10.662483	7.758842

H	9.061870	11.100630	8.758983
C	8.228285	9.575947	7.324752
H	7.513755	9.011055	7.924305
C	6.457283	4.080179	9.060099
H	5.743733	3.689669	9.794836
C	12.791193	7.742887	5.765407
H	13.203774	6.881334	5.207475
H	12.496457	8.511974	5.028748
C	9.223226	8.689254	11.146272
H	9.167198	8.566748	10.058514
C	8.763753	4.628762	8.452572
C	8.299962	5.081435	7.195871
H	9.014000	5.484324	6.468062
C	12.845964	6.864677	12.018955
C	13.473819	8.126796	12.132126
H	13.001266	9.008230	11.683481
C	13.475732	5.739886	12.590060
H	13.023240	4.745836	12.502910
C	15.307254	7.131173	13.386911
H	16.259867	7.234310	13.918545
C	14.697522	5.873016	13.271089
H	15.172442	4.987811	13.709292
C	14.692033	8.257603	12.813905
H	15.162506	9.243921	12.898382
C	13.417725	9.386376	7.696195
H	13.197828	10.323500	7.153237
H	14.237941	9.590936	8.409068
H	10.051282	7.175035	8.882086

Table S36. $[CpNi^{II}(pdt)HFe^{II}(dppe)(CO)]^0$ in Cycle B with pdt Oriented Toward Ni ($[H\mathbf{1a}]^0$, –3204.96011758 E_h)

atom	x	y	z
Ni	2.505936	6.027790	1.859034
Fe	1.973376	6.230903	4.655664
S	1.509595	7.716014	2.892030
P	2.844904	4.752795	6.037451
O	3.515391	8.355479	5.915032
C	4.538137	5.758413	1.249538
H	5.335253	5.659532	1.986848
C	4.015089	6.975102	0.713867
H	4.367551	7.982622	0.944300
C	2.993265	6.647706	-0.277808
H	2.427836	7.376723	-0.861363
C	-0.296791	7.622150	2.449739
H	-0.874490	7.703206	3.388998
H	-0.501015	8.523231	1.843049
C	-0.693708	6.366108	1.670118
H	-0.095826	6.315081	0.740050
H	-1.759867	6.459466	1.372730
C	2.872139	7.486369	5.436579
C	2.248393	5.238055	7.752725
C	1.753055	2.212550	6.869203
H	1.409191	2.701893	7.786856
C	1.491113	0.842440	6.691319
H	0.945572	0.290320	7.465362
C	1.934301	0.184490	5.534163
H	1.732122	-0.883684	5.396626
C	2.640887	0.901794	4.553705
H	2.990923	0.395265	3.647232
C	2.897344	2.269037	4.726786
H	3.447439	2.821048	3.956978
C	5.240117	3.800178	7.237679
H	4.585548	3.129261	7.806442
C	4.687692	4.681773	6.281120
C	5.551841	5.508148	5.535770
H	5.130647	6.183250	4.784506
C	6.939802	5.464216	5.751314
H	7.599580	6.114183	5.165400
C	7.478511	4.593114	6.710339
H	8.561155	4.559138	6.877195
C	6.625153	3.758771	7.452916
H	7.039566	3.070363	8.198403
H	2.512094	4.486666	8.519705
H	0.160841	4.587549	7.794422
H	2.816670	6.156053	7.988527
S	1.198250	4.666575	3.017930
C	-0.532583	5.060026	2.454480
H	-1.178745	5.062956	3.347245
H	-0.832164	4.205671	1.820941
C	0.738585	5.525610	7.730383
C	-1.456318	5.570533	5.862294
C	-1.516494	4.157867	5.790070

H	-0.607076	3.559779	5.924409
C	-2.727154	3.503882	5.515757
H	-2.750620	2.409095	5.467443
C	-3.899489	4.246148	5.294511
H	-4.844296	3.734850	5.078592
C	-3.849124	5.648538	5.347164
H	-4.756939	6.238521	5.175101
C	-2.639596	6.305599	5.626341
H	-2.619290	7.399629	5.671075
C	-1.116821	8.238439	7.868358
H	-1.482854	7.359304	8.411823
C	-0.307617	8.074959	6.719646
C	0.115977	9.223825	6.020995
H	0.741203	9.111414	5.128528
C	-0.249900	10.506518	6.464771
H	0.093757	11.388208	5.912179
C	-1.043596	10.656837	7.611516
H	-1.324616	11.657061	7.960439
C	-1.479424	9.518463	8.312263
H	-2.105385	9.627713	9.205480
H	0.444959	6.149506	8.592434
C	3.719875	4.704053	0.732341
H	3.809902	3.643816	0.978124
C	2.817564	5.257410	-0.271953
H	2.087415	4.686139	-0.847957
C	2.459748	2.942860	5.890116
P	0.201052	6.389453	6.110250
H	3.214647	6.028044	3.783052

Table S37. $[CpNi^{II}(pdt)HFe^{II}(dppe)(CO)]^0$ in Cycle B with pdt Oriented Toward Fe ($[H\mathbf{1a}]^0$, –3204.95690824 E_h)

atom	x	y	z
Ni	10.223777	8.952914	7.469037
Fe	11.265461	6.682839	8.885089
P	13.230307	5.547099	9.312824
S	12.065938	8.929285	8.693801
P	10.913309	6.670358	11.052086
S	10.900661	7.053858	6.565333
C	7.655097	6.274955	13.600788
H	7.497144	6.211651	14.683566
C	6.560112	6.209441	12.722003
H	5.544837	6.092127	13.117657
C	16.304435	8.157559	10.264772
H	16.412039	9.023307	10.928390
C	12.509196	7.844817	13.152767
H	12.917442	6.848036	13.351483
C	17.382824	7.740133	9.466025
H	18.338382	8.275168	9.505275
C	9.749619	11.048613	6.768627
H	10.491791	11.815434	6.540149
C	17.224866	6.635343	8.614281
H	18.058516	6.301911	7.985219
C	10.347252	5.224421	8.731784
C	8.072656	6.452811	10.829020
H	8.237054	6.530441	9.749964
C	9.180655	10.130540	5.870865
H	9.389547	10.044558	4.802809
C	13.635030	7.910984	6.473756
H	14.479321	8.165229	5.798524
H	13.951773	7.023237	7.044854
C	12.432870	10.213101	13.712735
H	12.773114	11.056027	14.324691
C	9.177387	6.511526	11.701777
C	6.771495	6.301028	11.337890
H	5.922246	6.255262	10.646736
C	16.001905	5.945596	8.564548
H	15.896652	5.084157	7.896813
C	8.300277	9.245983	6.629315
H	7.721756	8.422263	6.206166
O	9.693181	4.249773	8.589314
C	11.716278	5.104355	11.709304
H	11.716821	5.061275	12.814076
H	11.059221	4.290369	11.352790
C	11.478140	10.414592	12.701190
H	11.072345	11.416280	12.520194
C	13.132175	4.954985	11.130685
H	13.476964	3.908560	11.200363
H	13.850168	5.575239	11.694757
C	14.914846	6.348405	9.370330
C	8.953957	6.428048	13.094792
H	9.798508	6.500316	13.790198
C	9.244466	10.729626	8.097427

H	9.516227	11.258093	9.013718
C	8.266363	9.689458	7.985744
H	7.687187	9.253284	8.800215
C	12.947864	8.927233	13.935740
H	13.689523	8.759104	14.725164
C	12.426375	7.540074	5.617301
H	12.678022	6.676578	4.973709
H	12.124782	8.376320	4.959601
C	15.079867	7.474212	10.212886
H	14.241950	7.829007	10.824925
C	11.551081	8.036273	12.134620
C	11.044566	9.337969	11.915181
H	10.303424	9.503211	11.125156
C	13.568311	3.967378	8.389130
C	12.884799	3.733602	7.177984
H	12.168250	4.479839	6.816498
C	14.483653	2.993555	8.851298
H	15.045913	3.162564	9.777294
C	14.007369	1.589392	6.921670
H	14.173705	0.665140	6.356685
C	14.699824	1.813120	8.124451
H	15.411180	1.067011	8.497112
C	13.102855	2.552242	6.449042
H	12.556639	2.383539	5.514135
C	13.437396	9.084023	7.438288
H	13.227502	10.017462	6.883795
H	14.362562	9.233346	8.022352
H	9.953479	7.467292	8.843108

Table S38. $[CpNi^{II}(pdt)HFe^{II}(dppe)(CO)]^0$ Transition State with pdt Oriented Toward Ni ($[H\mathbf{1a}]^0$, –3204.92842649 E_h)

atom	x	y	z
Ni	10.386115	8.488705	6.450412
Fe	11.206965	6.773296	8.946701
P	12.081704	7.128308	10.952948
S	11.385180	9.065630	8.322202
P	9.538891	5.923354	9.965837
S	11.577463	6.657422	6.579128
C	6.703053	2.986559	9.150436
H	5.827657	2.555373	9.649759
C	7.106807	2.505363	7.893097
H	6.547637	1.695694	7.410334
C	10.358554	10.631642	12.390903
H	9.508438	11.265134	12.114287
C	7.794295	6.948259	12.027497
H	8.271179	6.249536	12.722925
C	11.119126	10.928066	13.532091
H	10.869261	11.797166	14.151495
C	10.055674	9.276911	4.367297
H	10.792306	9.269119	3.562306
C	12.199881	10.100924	13.880900
H	12.795037	10.318344	14.775453
C	12.093462	5.288052	8.874685
C	8.938950	4.108640	7.872126
H	9.808370	4.553397	7.375027
C	9.192111	8.167287	4.735186
H	9.155225	7.202071	4.225303
C	13.574833	8.699062	6.499642
H	13.008096	9.229502	5.710517
H	14.651866	8.909321	6.328154
C	6.144502	8.697517	11.643134
H	5.350946	9.354038	12.017357
C	8.543628	4.592311	9.135382
C	8.223295	3.068823	7.255345
H	8.540303	2.701960	6.272521
C	12.522775	8.990121	13.086239
H	13.366991	8.352882	13.370712
C	8.339346	8.573307	5.816620
H	7.549818	7.976434	6.274600
O	12.678960	4.261257	8.758158
C	10.251515	4.989819	11.448701
H	9.472836	4.628586	12.144415
H	10.714234	4.097535	10.990923
C	6.539252	8.761172	10.295227
H	6.052113	9.467294	9.612886
C	11.307549	5.855242	12.158646
H	12.099233	5.241570	12.622348
H	10.846585	6.454203	12.962633
C	11.771842	8.692030	11.925480
C	7.415484	4.025733	9.767635
H	7.081358	4.408864	10.739128
C	9.830195	10.307804	5.297321

H	10.356428	11.262402	5.356443
C	8.794076	9.861915	6.225914
H	8.405496	10.452042	7.058381
C	6.778434	7.792897	12.507637
H	6.483654	7.739559	13.562033
C	13.348261	7.190928	6.340191
H	13.952951	6.610795	7.059162
H	13.627665	6.878140	5.317463
C	10.679747	9.520211	11.593691
H	10.077434	9.291679	10.710974
C	8.198020	7.001739	10.676117
C	7.562056	7.928827	9.819065
H	7.887647	8.003707	8.774337
C	13.915601	6.895981	11.165587
C	14.798964	7.997394	11.074582
H	14.396263	9.013624	11.000326
C	14.468883	5.600357	11.273843
H	13.815236	4.722703	11.313382
C	16.725643	6.515267	11.222034
H	17.811042	6.367495	11.246804
C	15.859295	5.413107	11.305807
H	16.265070	4.398810	11.393395
C	16.189436	7.808020	11.104867
H	16.854823	8.676867	11.041413
C	13.185341	9.278081	7.864929
H	13.379315	10.366325	7.873438
H	13.778746	8.811136	8.670467
H	9.778918	6.986320	8.328362

Table S39. $[CpNi^{II}(pdt)HFe^{II}(dppe)(CO)]^0$ Transition State with pdt Oriented Toward Fe ($[H\mathbf{1a}]^0$, –3204.93021804 E_h)

atom	x	y	z
Ni	10.413099	8.829738	6.589482
Fe	11.248861	6.867737	8.936903
P	12.017960	7.049421	11.012062
S	11.343785	9.222576	8.533936
P	9.564360	5.864145	9.769298
S	11.631793	7.022438	6.568959
C	6.900213	2.910893	8.538766
H	6.021780	2.400050	8.949773
C	7.382906	2.569826	7.263535
H	6.882580	1.789746	6.678365
C	10.296754	10.448862	12.657434
H	9.416720	11.065838	12.444056
C	7.575373	7.869414	9.733245
H	7.992556	8.099032	8.744881
C	11.112098	10.738401	13.762644
H	10.873794	11.584491	14.417409
C	9.299148	8.687034	4.794935
H	9.294309	7.782564	4.182620
C	12.232654	9.934675	14.028515
H	12.870476	10.146381	14.894560
C	12.210501	5.436309	8.749244
C	9.142884	4.237174	7.487952
H	10.013932	4.761223	7.078956
C	8.386332	8.976235	5.864486
H	7.574728	8.335441	6.210983
C	13.928290	8.392725	7.599007
H	14.959025	8.732061	7.364390
H	14.004360	7.674586	8.435825
C	5.979855	8.354479	11.507756
H	5.142731	8.944665	11.897226
C	8.668546	4.579990	8.770033
C	8.502158	3.235517	6.739581
H	8.880095	2.978874	5.743467
C	12.541259	8.852552	13.188887
H	13.415206	8.231781	13.411439
C	8.812073	10.211302	6.436186
H	8.376524	10.707466	7.305940
O	12.827663	4.442316	8.549623
C	10.226011	4.845799	11.218037
H	9.427783	4.359776	11.807745
H	10.809677	4.041418	10.735608
C	6.548593	7.336193	12.287088
H	6.158629	7.126529	13.289821
C	11.121232	5.744069	12.085705
H	11.844968	5.165701	12.686214
H	10.507046	6.329613	12.792041
C	11.735327	8.562607	12.064896
C	7.537067	3.912162	9.287012
H	7.139972	4.186790	10.271338
C	10.184779	9.822677	4.604536

H	10.964984	9.901335	3.845703
C	9.902630	10.748764	5.628021
H	10.421075	11.690186	5.819522
C	6.496148	8.616658	10.227042
H	6.060730	9.412186	9.611443
C	13.375947	7.654642	6.375454
H	14.001447	6.768983	6.160734
H	13.368294	8.300487	5.478378
C	10.604276	9.369066	11.813813
H	9.961313	9.148511	10.957321
C	8.148761	6.830966	10.502025
C	7.620764	6.577343	11.786897
H	8.042094	5.785646	12.415349
C	13.824446	6.706371	11.282634
C	14.770464	7.750340	11.153573
H	14.425791	8.779891	11.003308
C	14.300306	5.390428	11.471885
H	13.597746	4.553243	11.543452
C	16.605358	6.175525	11.436364
H	17.679773	5.970100	11.498818
C	15.677340	5.128501	11.549909
H	16.023392	4.099249	11.697950
C	16.146032	7.488272	11.236759
H	16.861043	8.314071	11.145306
C	13.112190	9.603033	8.063189
H	13.065627	10.386385	7.284974
H	13.579336	10.045230	8.963097
H	9.828263	7.124685	8.316785

Table S40. $[CpNi^{II}(pdt)(H_2)Fe^{II}(dppe)(CO)]^+$ in Cycle A with pdt Oriented Toward Ni ($[H_2\mathbf{1a}]^+$, –3205.37137313 E_h)

atom	x	y	z
Ni	2.331744	6.013534	1.528735
Fe	1.974198	5.980039	4.513685
S	1.279773	7.485457	2.800823
P	2.740016	7.550047	5.947787
O	-0.416172	5.862045	6.178346
C	4.236571	6.130325	0.614054
H	5.167713	6.202802	1.176482
C	3.405868	7.215891	0.185117
H	3.596837	8.276836	0.357837
C	2.307928	6.682807	-0.617579
H	1.535653	7.279897	-1.105140
C	-0.500306	7.184288	2.325701
H	-1.079569	8.010646	2.775424
H	-0.524897	7.314914	1.229449
C	-1.115128	5.842934	2.739346
H	-2.130435	5.795829	2.294809
H	-1.264635	5.827462	3.830192
C	0.508588	5.901752	5.452201
C	2.996532	6.630021	7.557360
C	5.523032	8.166758	6.327048
H	5.476761	7.639290	7.285658
C	6.738701	8.752953	5.932685
H	7.616682	8.665364	6.581724
C	6.823434	9.455259	4.721015
H	7.769974	9.915664	4.418872
C	5.688146	9.573245	3.900025
H	5.745987	10.132193	2.959815
C	4.475184	8.984447	4.284367
H	3.592117	9.086588	3.641813
C	2.309166	9.993253	7.276765
H	3.346996	9.891084	7.613794
C	1.738266	9.023586	6.420303
C	0.413988	9.193529	5.968398
H	-0.031811	8.459639	5.291996
C	-0.334542	10.308390	6.378968
H	-1.362568	10.432553	6.022366
C	0.233219	11.260687	7.239050
H	-0.351671	12.130193	7.557161
C	1.556275	11.102374	7.686285
H	2.006027	11.847335	8.351250
H	3.548928	7.243931	8.291218
H	4.762990	5.419742	7.090386
H	1.980647	6.480170	7.966975
S	1.430582	4.431942	2.780207
C	-0.370399	4.575574	2.305627
H	-0.870135	3.689701	2.737663
H	-0.381345	4.462256	1.207164
C	3.690331	5.278753	7.308762
C	4.362424	3.517702	5.033646
C	5.559378	4.218324	4.758813

H	5.668103	5.274251	5.037606
C	6.628435	3.571096	4.122071
H	7.556356	4.119966	3.928255
C	6.510388	2.223860	3.738583
H	7.346704	1.719632	3.242922
C	5.318695	1.527702	3.993256
H	5.219457	0.478343	3.695767
C	4.247378	2.168210	4.637290
H	3.325342	1.613852	4.837375
C	2.416276	2.264407	7.605471
H	3.410671	2.466032	8.019295
C	1.913715	3.035178	6.530706
C	0.651745	2.714395	5.989434
H	0.262524	3.286761	5.143112
C	-0.104413	1.659421	6.525357
H	-1.085189	1.423931	6.098846
C	0.393476	0.912494	7.603646
H	-0.198809	0.093006	8.024201
C	1.657268	1.213631	8.139640
H	2.056113	0.627104	8.974043
H	3.619251	4.632966	8.200340
C	3.564251	4.929688	0.223248
H	3.895583	3.911279	0.434729
C	2.402490	5.284761	-0.591049
H	1.717708	4.572100	-1.053786
C	4.380863	8.274609	5.504422
P	2.936914	4.394790	5.818303
H	3.257283	6.059140	3.286218
H	3.583541	6.089894	4.072041

Table S41. $[CpNi^{II}(pdt)(H_2)Fe^{II}(dppe)(CO)]^+$ in Cycle A with pdt Oriented Toward Fe ($[H_2\mathbf{1a}]^+$, –3205.37563093 E_h)

atom	x	y	z
Ni	2.277670	6.021159	1.501438
Fe	2.004562	5.977391	4.494989
S	1.270530	7.489604	2.821045
P	2.743963	7.547573	5.950428
O	-0.544675	5.830260	5.898502
C	4.180839	6.130759	0.596140
H	5.110068	6.185929	1.163812
C	3.363257	7.229095	0.175507
H	3.562856	8.286138	0.362278
C	2.270121	6.717077	-0.649747
H	1.507917	7.329284	-1.134396
C	-0.549502	7.170507	2.547455
H	-1.049580	7.215791	3.530862
H	-0.901418	8.029067	1.949630
C	-0.873261	5.862916	1.817136
H	-0.431156	5.897286	0.804225
H	-1.973943	5.807703	1.689734
C	0.479645	5.885181	5.324281
C	2.990964	6.625446	7.559545
C	5.524923	8.167694	6.352065
H	5.470560	7.645924	7.313379
C	6.744089	8.751251	5.964365
H	7.616787	8.666722	6.620898
C	6.839009	9.447037	4.749693
H	7.788230	9.905221	4.452631
C	5.710260	9.561519	3.919248
H	5.775772	10.115880	2.976812
C	4.494105	8.975128	4.297126
H	3.615801	9.075058	3.647717
C	2.340956	10.013534	7.248088
H	3.391416	9.926474	7.547775
C	1.748632	9.024465	6.428979
C	0.406557	9.174950	6.025819
H	-0.058984	8.427295	5.379489
C	-0.337650	10.289058	6.446205
H	-1.379422	10.396960	6.126132
C	0.252357	11.260923	7.268492
H	-0.328669	12.130292	7.593974
C	1.592730	11.122003	7.667926
H	2.059907	11.881908	8.303336
H	3.529541	7.242558	8.301156
H	4.769984	5.435630	7.092630
H	1.971817	6.464789	7.957548
S	1.428244	4.430349	2.785368
C	-0.416134	4.578661	2.517699
H	-0.909644	4.460623	3.498694
H	-0.681755	3.703687	1.899276
C	3.699313	5.282431	7.312370
C	4.382360	3.509456	5.052147
C	5.580582	4.208193	4.778006

H	5.688336	5.265739	5.051187
C	6.652627	3.556984	4.150388
H	7.581310	4.104729	3.956993
C	6.536545	2.207176	3.775507
H	7.375063	1.699817	3.286823
C	5.343990	1.512465	4.030013
H	5.246365	0.461005	3.739453
C	4.269693	2.157151	4.664911
H	3.347079	1.603794	4.865334
C	2.425801	2.283147	7.634274
H	3.416145	2.493124	8.053385
C	1.929468	3.037728	6.544997
C	0.674273	2.702525	5.997358
H	0.291776	3.256029	5.136363
C	-0.082603	1.652022	6.541175
H	-1.057837	1.405968	6.108026
C	0.408233	0.923125	7.634773
H	-0.184330	0.107237	8.061876
C	1.666376	1.237002	8.176779
H	2.060826	0.663651	9.022367
H	3.636777	4.636747	8.204490
C	3.498263	4.942536	0.183336
H	3.818352	3.917873	0.382093
C	2.351503	5.319727	-0.643391
H	1.664949	4.619549	-1.122112
C	4.389367	8.271040	5.519740
P	2.952736	4.393002	5.822515
H	3.277500	6.061604	3.287746
H	3.618269	6.090193	4.062797

Table S42. $[CpNi^{II}(pdt)(H_2)Fe^{II}(dppe)(CO)]^+$ in Cycle B with pdt Oriented Toward Ni ($[H_2\mathbf{1a}]^+$, –3205.36262279 E_h)

atom	x	y	z
Ni	2.459279	6.112966	1.723823
Fe	2.126568	6.226291	4.698586
S	1.518089	7.717471	2.928500
P	2.926773	4.687246	6.185451
O	3.376033	8.464367	6.103395
C	4.399163	5.987892	0.906725
H	5.301162	5.946304	1.518429
C	3.738524	7.166398	0.433693
H	4.050502	8.195661	0.621718
C	2.632526	6.767573	-0.434497
H	1.967200	7.455041	-0.959403
C	-0.305540	7.562590	2.569260
H	-0.842640	7.621436	3.531325
H	-0.552733	8.468502	1.988035
C	-0.695312	6.311839	1.779199
H	-0.171831	6.324265	0.805359
H	-1.782062	6.365449	1.563845
C	2.855144	7.556687	5.572446
C	2.303657	5.284630	7.840224
C	1.707270	2.243895	7.059279
H	1.441436	2.752679	7.991563
C	1.361534	0.890519	6.904070
H	0.825662	0.373957	7.707639
C	1.714371	0.202117	5.732826
H	1.447790	-0.853557	5.615467
C	2.419876	0.868039	4.716366
H	2.706675	0.333471	3.804627
C	2.761914	2.219710	4.864438
H	3.320788	2.724818	4.069924
C	5.318238	3.382892	6.974658
H	4.676612	2.529615	7.216231
C	4.760161	4.557552	6.419639
C	5.613524	5.639595	6.108787
H	5.211346	6.566163	5.688721
C	6.993630	5.548787	6.349006
H	7.641389	6.397056	6.103477
C	7.538891	4.377752	6.898226
H	8.616391	4.306477	7.080402
C	6.698289	3.297434	7.211731
H	7.116285	2.380177	7.640068
H	2.534869	4.560829	8.642270
H	0.203342	4.676710	7.859129
H	2.889665	6.196588	8.052998
S	1.336641	4.674168	3.004191
C	-0.426657	4.980364	2.487905
H	-1.054652	4.869634	3.383955
H	-0.659120	4.143085	1.806546
C	0.802922	5.597145	7.763425
C	-1.305585	5.536245	5.776473
C	-1.350168	4.122299	5.784311

H	-0.446700	3.537369	5.991503
C	-2.548516	3.448242	5.504606
H	-2.568733	2.353253	5.521191
C	-3.714081	4.172587	5.201700
H	-4.648684	3.644596	4.984882
C	-3.675529	5.575763	5.180347
H	-4.580618	6.148224	4.950339
C	-2.479930	6.256244	5.464569
H	-2.469865	7.350943	5.462786
C	-1.097338	8.250836	7.739118
H	-1.448090	7.373119	8.293873
C	-0.246851	8.097699	6.616997
C	0.149385	9.245761	5.900517
H	0.799510	9.152391	5.025139
C	-0.280392	10.521135	6.305250
H	0.042213	11.403040	5.741777
C	-1.110453	10.663649	7.426523
H	-1.440124	11.658523	7.744132
C	-1.521421	9.524781	8.141168
H	-2.175989	9.627538	9.013255
H	0.501510	6.271541	8.582599
C	3.604946	4.877180	0.473168
H	3.799072	3.825655	0.693410
C	2.551473	5.369348	-0.411609
H	1.810929	4.746265	-0.915536
C	2.407504	2.924305	6.038453
P	0.300652	6.400250	6.113460
H	3.291928	6.105003	3.403795
H	3.637511	5.954081	4.204562

Table S43. $[CpNi^{II}(pdt)(H_2)Fe^{II}(dppe)(CO)]^+$ in Cycle B with pdt Oriented Toward Fe ($[H_2\mathbf{1a}]^+$, –3205.35830320 E_h)

atom	x	y	z
Ni	2.538775	5.997677	1.744333
Fe	2.105919	6.276014	4.703687
S	1.828679	7.798789	2.821611
P	2.901170	4.733635	6.186769
O	3.266846	8.538116	6.142455
C	4.437984	5.513102	0.945184
H	5.321109	5.340415	1.561367
C	3.986232	6.770042	0.431433
H	4.469012	7.737034	0.585398
C	2.817622	6.537974	-0.413971
H	2.275398	7.309844	-0.962711
C	0.091850	7.937852	2.174453
H	-0.269321	8.938534	2.472057
H	0.186436	7.923700	1.074095
C	-0.875550	6.870541	2.677481
H	-1.860790	7.046628	2.199411
H	-1.032223	7.019055	3.757083
C	2.796972	7.611638	5.596169
C	2.282147	5.331260	7.840729
C	1.781658	2.257385	7.096216
H	1.591868	2.740376	8.060188
C	1.434771	0.905586	6.935082
H	0.976176	0.362287	7.768179
C	1.684926	0.252987	5.717435
H	1.417179	-0.801885	5.595486
C	2.286301	0.954047	4.658508
H	2.489972	0.447819	3.709020
C	2.627972	2.305396	4.812412
H	3.101167	2.840967	3.982564
C	5.340214	3.379182	6.743771
H	4.729416	2.477864	6.850739
C	4.738729	4.610032	6.396093
C	5.554571	5.756955	6.262847
H	5.119744	6.726610	6.004019
C	6.939078	5.673054	6.475250
H	7.556142	6.571821	6.370833
C	7.528497	4.445060	6.816318
H	8.609979	4.380222	6.975895
C	6.726165	3.301281	6.951175
H	7.177517	2.339659	7.217885
H	2.529756	4.619150	8.648185
H	0.200507	4.671905	7.847689
H	2.850422	6.256409	8.044136
S	1.165535	4.813422	3.025547
C	-0.496457	5.411053	2.412997
H	-1.239944	4.748479	2.887483
H	-0.485630	5.194672	1.329768
C	0.775061	5.609275	7.759950
C	-1.298464	5.431764	5.764258
C	-1.200791	4.032965	5.580186

H	-0.226628	3.534211	5.625454
C	-2.350783	3.267697	5.331435
H	-2.259289	2.184596	5.196599
C	-3.609636	3.887097	5.253693
H	-4.506200	3.288704	5.060053
C	-3.712891	5.276391	5.426807
H	-4.690402	5.767470	5.370776
C	-2.565993	6.047167	5.679264
H	-2.664091	7.128647	5.815950
C	-1.164682	8.247829	7.728211
H	-1.464205	7.384728	8.333521
C	-0.349746	8.074562	6.583635
C	-0.013141	9.204082	5.808708
H	0.622048	9.088938	4.924616
C	-0.470035	10.481242	6.173793
H	-0.191427	11.349784	5.567635
C	-1.269485	10.643825	7.315008
H	-1.620846	11.640341	7.602737
C	-1.618189	9.524318	8.090279
H	-2.245606	9.644055	8.979965
H	0.454318	6.272843	8.580668
C	3.464548	4.539168	0.550586
H	3.478240	3.478077	0.806760
C	2.499633	5.173778	-0.343198
H	1.660140	4.669909	-0.824817
C	2.378350	2.972736	6.034170
P	0.255655	6.393831	6.103073
H	3.300702	5.990086	3.437145
H	3.638443	6.031978	4.251732

Table S44. $[CpNi^{II}(pdt)(H_2)Fe^{II}(dppe)(CO)]^+$ Transition State with pdt Oriented Toward Ni ($[H_2\mathbf{1a}]^+$, –3205.32955903 E_h)

atom	x	y	z
Ni	10.736536	9.120781	6.620028
Fe	10.968802	6.950181	8.888038
P	12.006783	6.977916	10.871523
S	11.704249	9.294353	8.596854
P	9.315712	5.921268	10.013912
S	11.618290	7.100712	6.467995
C	7.197889	2.438969	9.286744
H	6.550811	1.769089	9.862949
C	7.455055	2.179475	7.929116
H	7.011782	1.302198	7.446185
C	10.339350	10.222337	12.889993
H	9.659971	11.049640	12.658393
C	6.725678	6.892607	9.462033
H	6.785225	6.264775	8.566243
C	10.865147	10.081270	14.182983
H	10.597806	10.797784	14.967031
C	9.549669	9.251419	4.890054
H	9.244550	8.375254	4.314271
C	11.739354	9.018278	14.470352
H	12.153929	8.901941	15.477433
C	11.683866	5.416020	8.564101
C	8.852684	4.168523	7.814303
H	9.499269	4.836579	7.235230
C	8.842174	9.819152	5.995839
H	7.905920	9.462673	6.426237
C	13.903161	8.779907	6.805956
H	13.487448	9.481064	6.058617
H	15.007530	8.828112	6.715973
C	5.467922	8.445223	10.851959
H	4.564210	9.036085	11.034943
C	8.609297	4.428743	9.178276
C	8.276146	3.047407	7.193827
H	8.476312	2.852443	6.134947
C	12.090836	8.105142	13.465884
H	12.792310	7.296456	13.700446
C	9.646920	10.904044	6.475212
H	9.425191	11.531789	7.340581
O	12.220624	4.394837	8.311320
C	9.956336	5.300139	11.676240
H	9.520902	5.935989	12.465052
H	9.597778	4.269665	11.837828
C	6.522548	8.465288	11.778023
H	6.446116	9.070880	12.687462
C	11.488995	5.379444	11.709055
H	11.942381	4.543456	11.149644
H	11.874999	5.342984	12.741991
C	11.565375	8.239960	12.157631
C	7.764357	3.560478	9.908190
H	7.530577	3.764872	10.959529
C	10.705797	10.094289	4.587553

H	11.413050	9.921715	3.774693
C	10.762489	11.107167	5.553920
H	11.524133	11.882644	5.648608
C	5.573849	7.659098	9.692334
H	4.751876	7.630442	8.968960
C	13.463662	7.355087	6.457535
H	13.900571	6.610324	7.144819
H	13.782063	7.101931	5.431176
C	10.682992	9.304328	11.882083
H	10.267106	9.419126	10.877621
C	7.791465	6.906074	10.391468
C	7.681871	7.707194	11.548329
H	8.497128	7.756483	12.276345
C	13.855263	6.922660	10.932082
C	14.608498	8.019533	11.410364
H	14.099999	8.917257	11.776023
C	14.537677	5.776746	10.459448
H	13.983805	4.920357	10.062898
C	16.679607	6.815308	10.973802
H	17.773607	6.770691	10.995768
C	15.939399	5.724095	10.488420
H	16.452776	4.824815	10.131507
C	16.011520	7.962472	11.430425
H	16.581131	8.818041	11.808978
C	13.532656	9.253850	8.214033
H	13.869371	10.295422	8.358513
H	14.015370	8.629941	8.983532
H	9.435307	7.273173	8.247320
H	9.764850	7.974065	7.885597

Table S45. $[CpNi^{II}(pdt)(H_2)Fe^{II}(dppe)(CO)]^+$ Transition State with pdt Oriented Toward Fe ($[H_2\mathbf{1a}]^+$, –3205.33003368 E_h)

atom	x	y	z
Ni	10.687939	9.134404	6.643926
Fe	10.981739	6.937034	8.889657
P	12.021742	6.951737	10.876097
S	11.670891	9.295399	8.608514
P	9.315736	5.923339	10.010106
S	11.593078	7.137372	6.435338
C	7.215020	2.425084	9.308493
H	6.564367	1.760968	9.887328
C	7.485509	2.148696	7.956833
H	7.049279	1.263910	7.481248
C	10.359761	10.200803	12.896350
H	9.662597	11.015782	12.674044
C	6.733494	6.886979	9.412286
H	6.806266	6.249858	8.524032
C	10.922247	10.082057	14.175988
H	10.665338	10.803369	14.959119
C	9.523777	9.308375	4.900272
H	9.221400	8.446742	4.301693
C	11.820282	9.035721	14.450904
H	12.263862	8.937120	15.447462
C	11.706863	5.407478	8.557592
C	8.877948	4.140570	7.829141
H	9.526847	4.803408	7.246886
C	8.802097	9.859536	6.005522
H	7.856375	9.500733	6.412491
C	14.061350	8.012015	7.590852
H	15.134219	8.190669	7.375710
H	14.022320	7.231096	8.368861
C	5.454284	8.452374	10.767912
H	4.547197	9.043568	10.932130
C	8.621953	4.417541	9.187617
C	8.310548	3.009771	7.217946
H	8.520598	2.802264	6.163375
C	12.158766	8.117007	13.447265
H	12.878268	7.321111	13.670537
C	9.605915	10.926836	6.522836
H	9.373770	11.537495	7.397547
O	12.253721	4.393064	8.302994
C	9.934605	5.327388	11.688918
H	9.514713	5.993864	12.460662
H	9.550785	4.310531	11.876458
C	6.496143	8.484389	11.707924
H	6.406183	9.099604	12.609647
C	11.468133	5.369888	11.722333
H	11.898330	4.518162	11.168314
H	11.853104	5.332346	12.755738
C	11.595787	8.229184	12.152653
C	7.772551	3.556180	9.920581
H	7.527640	3.773419	10.966756
C	10.696316	10.141126	4.641786

H	11.417888	9.979702	3.839276
C	10.744394	11.133371	5.632171
H	11.512609	11.897942	5.757902
C	5.577373	7.653803	9.618493
H	4.765389	7.615723	8.884348
C	13.412745	7.475921	6.313205
H	13.881017	6.517522	6.027651
H	13.528274	8.179481	5.469168
C	10.690315	9.276933	11.889195
H	10.247377	9.376316	10.894725
C	7.786364	6.911823	10.356163
C	7.659528	7.725457	11.502579
H	8.464261	7.783873	12.241667
C	13.866773	6.872621	10.957677
C	14.626164	8.010053	11.315243
H	14.121680	8.940258	11.597146
C	14.541266	5.677090	10.615663
H	13.982098	4.785994	10.313752
C	16.689630	6.754705	11.006930
H	17.783431	6.706992	11.031431
C	15.942940	5.620248	10.647731
H	16.450856	4.684265	10.391846
C	16.028608	7.948623	11.338136
H	16.604066	8.835616	11.624454
C	13.483049	9.312397	8.152774
H	13.592348	10.148705	7.439402
H	14.018852	9.580704	9.079377
H	9.457852	7.228382	8.219722
H	9.766304	7.947089	7.859746

Table S46. $[CpNi^{II}(pdt)Fe^{II}(dppv)(CO)]^+$ with pdt Oriented Toward Ni ($[\mathbf{1b}]^+$, –3202.98799965 E_h)

atom	x	y	z
Ni	10.441312	8.705691	7.422905
Fe	11.667542	6.832880	8.733993
P	11.413405	6.788047	10.989003
S	12.148330	9.033524	8.771636
P	10.502159	4.898555	8.718405
S	11.522215	7.034432	6.490883
C	10.785404	1.067761	7.180391
H	10.174633	0.158764	7.196859
C	12.022686	1.076407	6.512905
H	12.379166	0.171727	6.009012
C	7.969491	8.865625	11.930218
H	7.010591	9.064832	11.438688
C	7.697670	5.222690	9.125201
H	7.928996	5.293972	10.193905
C	8.206462	9.341511	13.231048
H	7.435464	9.917370	13.753915
C	9.589890	10.655005	7.148093
H	10.136312	11.589706	7.284249
C	9.432556	9.073302	13.860290
H	9.620581	9.437165	14.876115
C	13.247299	6.147881	8.859112
C	12.343287	3.406524	7.137808
H	12.955816	4.311084	7.117288
C	9.446896	9.928612	5.941340
H	9.852810	10.195573	4.964805
C	13.399328	9.198657	6.178730
H	12.587787	9.856036	5.814731
H	14.324543	9.510379	5.652257
C	6.052822	5.236884	7.328336
H	5.012887	5.319309	6.994928
C	11.105408	3.405081	7.811552
C	12.797927	2.245546	6.490519
H	13.761056	2.258347	5.969313
C	10.424252	8.333057	13.196356
H	11.372759	8.124792	13.700757
C	8.697671	8.724883	6.249898
H	8.446122	7.934961	5.540751
O	14.360405	5.765932	8.873788
C	7.077811	5.013966	6.391776
H	6.837481	4.917583	5.327196
C	10.802590	5.110080	11.445900
C	10.197333	7.863381	11.884602
C	10.325317	2.224917	7.823690
H	9.349862	2.212171	8.323335
C	8.905891	9.919609	8.205648
H	8.854735	10.225392	9.251632
C	8.310740	8.756952	7.632202
H	7.713706	7.999206	8.140162
C	6.364799	5.337864	8.694340
H	5.568591	5.492696	9.430754

C	13.097966	7.750799	5.788120
H	13.910357	7.072527	6.104256
H	12.985522	7.665084	4.693438
C	8.960987	8.133473	11.259110
H	8.780425	7.765183	10.243340
C	8.730563	4.997270	8.189152
C	8.410078	4.892622	6.816329
H	9.199591	4.697279	6.082272
C	12.983255	6.975341	11.931577
C	13.545943	8.260317	12.109436
H	13.014645	9.148575	11.749814
C	13.684105	5.842351	12.396757
H	13.272243	4.837650	12.249797
C	15.470170	7.272095	13.227005
H	16.434479	7.387711	13.732992
C	14.921046	5.993500	13.044099
H	15.454937	5.107621	13.404000
C	14.781308	8.404052	12.758265
H	15.205753	9.403917	12.899502
C	13.609057	9.425109	7.676902
H	13.835996	10.487236	7.874017
H	14.446432	8.817802	8.064146
C	10.376493	4.299249	10.457655
H	9.956655	3.303885	10.657004
H	10.740027	4.827846	12.505939

Table S47. $[CpNi^{II}(pdt)Fe^{II}(dppv)(CO)]^+$ with pdt Oriented Toward Fe ($[\mathbf{1b}]^+$, –3202.98899445 E_h)

atom	x	y	z
Ni	10.451857	8.673567	7.459182
Fe	11.711725	6.813202	8.732220
P	11.433190	6.790466	10.984203
S	12.175125	9.019610	8.772047
P	10.512600	4.902969	8.711069
S	11.527227	7.027871	6.491719
C	10.787459	1.065713	7.189825
H	10.189499	0.149317	7.236822
C	11.995458	1.085938	6.470873
H	12.342286	0.182668	5.957819
C	7.960004	8.829352	11.902463
H	6.995180	9.002556	11.412714
C	7.708051	5.215978	9.133156
H	7.942783	5.279853	10.201507
C	8.195559	9.333609	13.192887
H	7.417278	9.905343	13.709450
C	9.630606	10.632015	7.199801
H	10.190579	11.555534	7.356344
C	9.429596	9.098869	13.819749
H	9.616995	9.484864	14.827474
C	13.243025	6.032989	8.923646
C	12.311707	3.424023	7.067885
H	12.909074	4.337499	7.014209
C	9.498354	9.920659	5.982523
H	9.928397	10.190349	5.017075
C	14.056027	8.403226	6.678097
H	14.887047	8.822282	6.075319
H	14.487392	7.586122	7.281321
C	6.055931	5.233607	7.343106
H	5.014161	5.311999	7.014498
C	11.104363	3.410395	7.794755
C	12.753222	2.264958	6.408182
H	13.692753	2.286770	5.845813
C	10.430393	8.363433	13.164119
H	11.384519	8.180313	13.667421
C	8.717828	8.730225	6.264859
H	8.464790	7.952285	5.543054
O	14.309261	5.545049	9.020417
C	7.078510	5.024915	6.400634
H	6.834525	4.935687	5.336298
C	10.820546	5.112783	11.438740
C	10.204402	7.865608	11.862599
C	10.340108	2.220914	7.845275
H	9.385687	2.199075	8.383825
C	8.907324	9.902130	8.235107
H	8.841589	10.199461	9.282600
C	8.302250	8.758768	7.638649
H	7.684463	8.005719	8.128380
C	6.372660	5.325440	8.708567
H	5.578474	5.468859	9.449440

C	13.015828	7.838778	5.711338
H	13.465151	7.047340	5.084739
H	12.621541	8.618313	5.034907
C	8.960368	8.102402	11.238978
H	8.781049	7.714094	10.230662
C	8.738338	5.005660	8.191000
C	8.413105	4.908919	6.818870
H	9.200843	4.724881	6.080010
C	12.991509	6.987322	11.943809
C	13.559504	8.273003	12.099152
H	13.039774	9.155204	11.708894
C	13.677172	5.862220	12.448716
H	13.262628	4.856388	12.319319
C	15.458362	7.301479	13.273055
H	16.414932	7.423825	13.791985
C	14.904225	6.022140	13.112621
H	15.426674	5.142190	13.502538
C	14.784561	8.425612	12.765072
H	15.212784	9.426163	12.889008
C	13.543640	9.507220	7.601911
H	13.171576	10.377880	7.032438
H	14.356370	9.858321	8.262722
C	10.392943	4.302893	10.450371
H	9.971052	3.308719	10.650919
H	10.756097	4.830305	12.498555

Table S48. $[CpNi^{I}(pdt)Fe^{II}(dppv)(CO)]^{0}$ with pdt Oriented Toward Ni ($[\mathbf{1b}]^{0}$, – 3203.15408796 E_h)

atom	x	y	z
Ni	2.575026	5.928760	2.106496
Fe	1.620047	5.949297	4.417138
S	1.108430	7.525918	2.855222
P	2.710520	7.506230	5.600043
O	-0.908307	5.944944	5.862380
C	4.743490	5.954527	2.047189
H	5.373589	5.979391	2.936157
C	4.266302	7.099089	1.323041
H	4.427602	8.141872	1.598610
C	3.480048	6.624124	0.226493
H	2.949592	7.244083	-0.499163
C	-0.572127	7.192363	2.127416
H	-1.303414	7.216105	2.956254
H	-0.777647	8.060168	1.474566
C	-0.699490	5.894816	1.325570
H	0.053577	5.890776	0.514545
H	-1.700542	5.881514	0.843563
C	0.156575	5.948051	5.343276
C	5.428681	8.136401	4.950180
H	5.697356	7.231750	5.507647
C	6.437891	8.905005	4.347644
H	7.485127	8.595019	4.440193
C	6.106872	10.067546	3.632860
H	6.894165	10.667505	3.162949
C	4.761185	10.459030	3.526589
H	4.494472	11.366355	2.972705
C	3.751392	9.696767	4.133770
H	2.706310	10.012745	4.050716
C	2.446394	9.675850	7.429687
H	3.518706	9.544155	7.615607
C	1.781219	8.852316	6.492996
C	0.415359	9.078155	6.235507
H	-0.103045	8.462813	5.496626
C	-0.277539	10.095308	6.914067
H	-1.340027	10.259201	6.701926
C	0.385498	10.894647	7.857034
H	-0.157108	11.684501	8.388759
C	1.751723	10.684471	8.112223
H	2.279552	11.311606	8.839797
S	1.132411	4.327863	2.893401
C	-0.552778	4.618815	2.158145
H	-1.284064	4.603685	2.987188
H	-0.744972	3.732776	1.526043
C	4.117889	3.427236	4.911728
C	5.462133	3.847954	4.988233
H	5.713561	4.775587	5.515076
C	6.484574	3.083775	4.402810
H	7.525123	3.419752	4.478463
C	6.175316	1.892605	3.726321
H	6.972991	1.296085	3.269730

C	4.838096	1.468423	3.640895
H	4.588358	0.539002	3.116534
C	3.815181	2.226450	4.231294
H	2.776932	1.885087	4.164769
C	2.506687	2.311781	7.519777
H	3.576329	2.466327	7.703411
C	1.828880	3.099994	6.561970
C	0.467379	2.844597	6.309027
H	-0.060245	3.432263	5.554295
C	-0.209131	1.833329	7.012462
H	-1.268417	1.646157	6.803620
C	0.466106	1.069464	7.975960
H	-0.063767	0.284286	8.527099
C	1.828252	1.309101	8.226953
H	2.365618	0.709618	8.970676
C	4.294559	4.771573	1.367371
H	4.481876	3.744240	1.681699
C	3.498320	5.185074	0.253539
H	2.983027	4.525129	-0.447347
C	4.075934	8.524146	4.852304
P	2.735802	4.439095	5.636383
C	3.585832	6.669932	6.998669
C	3.597228	5.322836	7.014460
H	4.086489	4.758465	7.820360
H	4.065173	7.261274	7.791087

Table S49. $[CpNi^{I}(pdt)Fe^{II}(dppv)(CO)]^{0}$ with pdt Oriented Toward Fe ($[\mathbf{1b}]^{0}$, –3203.15227771 E_h)

atom	x	y	z
Ni	2.576851	5.929252	2.147481
Fe	1.592883	5.951091	4.439259
S	1.102322	7.524294	2.861651
P	2.708555	7.502910	5.599147
O	-0.776324	5.956392	6.142674
C	4.764502	6.037312	2.014416
H	5.422972	6.139981	2.877054
C	4.213893	7.116623	1.248952
H	4.340432	8.179047	1.460037
C	3.401675	6.549500	0.214264
H	2.824524	7.105779	-0.527350
C	-0.524814	7.193340	2.010566
H	-1.164071	8.066594	2.235405
H	-0.315921	7.200880	0.924701
C	-1.246834	5.906139	2.411775
H	-2.232610	5.893010	1.899818
H	-1.453956	5.919401	3.496610
C	0.206628	5.955041	5.482314
C	5.443827	8.136387	5.019576
H	5.702202	7.250281	5.610595
C	6.466098	8.902505	4.435673
H	7.512605	8.607696	4.574626
C	6.148652	10.043265	3.681265
H	6.945903	10.641411	3.226087
C	4.803484	10.416056	3.516659
H	4.547390	11.307107	2.932193
C	3.780570	9.656372	4.104311
H	2.735628	9.957175	3.974696
C	2.439092	9.668834	7.434239
H	3.502248	9.513542	7.651758
C	1.784285	8.863349	6.475174
C	0.432004	9.119970	6.176956
H	-0.074689	8.519295	5.417399
C	-0.258282	10.149959	6.838267
H	-1.310008	10.338395	6.595042
C	0.393351	10.931012	7.804345
H	-0.147680	11.730679	8.322838
C	1.746284	10.690133	8.099691
H	2.265474	11.303095	8.845318
S	1.124682	4.331992	2.900660
C	-0.508352	4.617987	2.044807
H	-1.135761	3.742899	2.294554
H	-0.301114	4.583450	0.959109
C	4.111766	3.439674	4.892565
C	5.444597	3.899502	4.931357
H	5.679200	4.848050	5.427246
C	6.475875	3.149667	4.343686
H	7.507870	3.515893	4.389342
C	6.186153	1.934761	3.701081
H	6.990839	1.349699	3.241966

C	4.859738	1.473270	3.651019
H	4.625132	0.526580	3.151233
C	3.828120	2.216701	4.245134
H	2.798486	1.846468	4.206426
C	2.479048	2.302560	7.505496
H	3.541169	2.472168	7.717044
C	1.814390	3.081585	6.531297
C	0.463353	2.806902	6.243322
H	-0.051004	3.387074	5.473097
C	-0.215943	1.785483	6.928851
H	-1.266846	1.583099	6.693475
C	0.445583	1.030756	7.909061
H	-0.086900	0.237861	8.446453
C	1.797282	1.289504	8.194648
H	2.324056	0.697290	8.951624
C	4.335079	4.800547	1.424453
H	4.577467	3.800508	1.785529
C	3.483787	5.117046	0.319430
H	2.969714	4.397517	-0.321183
C	4.091551	8.504886	4.861780
P	2.722273	4.433240	5.626457
C	3.563882	6.657827	7.004618
C	3.572844	5.310547	7.015115
H	4.055494	4.742804	7.822466
H	4.038011	7.243892	7.803990

Table S50. $[CpNi^{I}(pdt)Fe^{I}(dppv)(CO)]^{-}$ with pdt Oriented Toward Ni ($[\mathbf{1b}]^{-}$, –3203.18570180 E_h)

atom	x	y	z
Ni	10.348631	8.717371	7.388539
Fe	11.770476	6.794174	8.730061
P	11.516365	6.762535	10.906509
S	12.102583	9.104987	8.815906
P	10.525621	4.973432	8.705358
S	11.558995	7.021526	6.445919
C	10.746024	1.063847	7.222329
H	10.119194	0.163231	7.239935
C	11.997592	1.040187	6.584011
H	12.352987	0.120791	6.102216
C	8.258473	9.159304	11.818442
H	7.442831	9.618595	11.247227
C	7.690092	5.146677	9.138076
H	7.934156	5.231390	10.203785
C	8.324726	9.322325	13.212147
H	7.558647	9.906474	13.737082
C	9.367828	10.739449	6.858104
H	9.924632	11.679842	6.852123
C	9.383043	8.739760	13.931686
H	9.444405	8.866391	15.020091
C	13.399650	6.156163	8.770963
C	12.335953	3.378742	7.183056
H	12.950326	4.283469	7.174369
C	9.202690	9.849674	5.752580
H	9.598145	9.994456	4.743913
C	13.414189	9.212458	6.260796
H	12.567999	9.837668	5.914784
H	14.329141	9.570744	5.735956
C	6.024442	5.140413	7.359251
H	4.979391	5.209595	7.034384
C	11.083160	3.413433	7.826331
C	12.788729	2.201436	6.563819
H	13.763475	2.195168	6.061092
C	10.365407	7.994734	13.260569
H	11.191572	7.549723	13.827406
C	8.456636	8.711881	6.206088
H	8.184114	7.840143	5.608600
O	14.529273	5.777692	8.734721
C	7.052511	4.972721	6.413350
H	6.810187	4.909696	5.345208
C	10.867085	5.104382	11.431831
C	10.302550	7.823781	11.859473
C	10.290846	2.242708	7.834383
H	9.301918	2.260197	8.308178
C	8.730957	10.152599	7.999178
H	8.706238	10.582170	9.003175
C	8.166715	8.893102	7.597967
H	7.587280	8.208074	8.220348
C	6.351240	5.230764	8.723250
H	5.559518	5.366121	9.470938

C	13.161615	7.758971	5.839041
H	13.983958	7.106117	6.187213
H	13.123518	7.697402	4.734431
C	9.242350	8.416592	11.144243
H	9.216124	8.307205	10.051483
C	8.728971	4.960414	8.197978
C	8.389763	4.886252	6.825664
H	9.184497	4.763854	6.080792
C	13.039835	6.948344	11.946916
C	13.555698	8.238420	12.210947
H	13.012892	9.118584	11.849217
C	13.768027	5.822468	12.385805
H	13.394800	4.815755	12.163150
C	15.472524	7.263613	13.358242
H	16.414565	7.386098	13.906630
C	14.972766	5.978132	13.090638
H	15.525243	5.091269	13.424657
C	14.762594	8.391032	12.911003
H	15.151407	9.398090	13.106058
C	13.609068	9.430363	7.767068
H	13.888628	10.485566	7.951393
H	14.426650	8.790934	8.149515
C	10.378857	4.326241	10.443987
H	9.907826	3.351380	10.640621
H	10.824907	4.817344	12.492897

Table S51. $[CpNi^{I}(pdt)Fe^{I}(dppv)(CO)]^{-}$ with pdt Oriented Toward Fe ([**1b**]$^{-}$, –3203.18219354 E_h)

atom	x	y	z
Ni	10.391213	8.644329	7.389750
Fe	11.834520	6.769388	8.731128
P	11.557108	6.747895	10.904389
S	12.093182	9.110641	8.847847
P	10.520476	4.981581	8.689324
S	11.688552	7.020654	6.452105
C	10.707613	1.071050	7.208296
H	10.101266	0.158842	7.275651
C	11.904743	1.070151	6.472551
H	12.238150	0.156829	5.964193
C	8.259021	9.112726	11.754906
H	7.440555	9.552243	11.172412
C	7.700398	5.188208	9.206051
H	7.977826	5.291222	10.261900
C	8.313260	9.294597	13.146719
H	7.534836	9.873780	13.658845
C	9.425895	10.644547	6.815686
H	9.983662	11.584407	6.818871
C	9.374764	8.737000	13.880907
H	9.427037	8.878474	14.967907
C	13.392483	5.985939	8.861007
C	12.244766	3.416355	7.040978
H	12.831480	4.336869	6.973311
C	9.316409	9.731307	5.721157
H	9.758688	9.857000	4.729462
C	14.118709	8.550460	6.869921
H	14.994748	8.986669	6.338427
H	14.511306	7.761978	7.535384
C	5.979388	5.165525	7.480133
H	4.925636	5.241232	7.186425
C	11.047507	3.427775	7.783581
C	12.668919	2.246507	6.388119
H	13.599779	2.257965	5.808263
C	10.372821	7.998538	13.225899
H	11.201937	7.573587	13.803546
C	8.535972	8.608005	6.157203
H	8.288155	7.726824	5.562866
O	14.457579	5.454338	8.919237
C	6.975998	4.965799	6.506428
H	6.699710	4.882821	5.447893
C	10.908161	5.088254	11.418184
C	10.321843	7.809080	11.826822
C	10.280714	2.241915	7.855006
H	9.332022	2.241068	8.405279
C	8.722270	10.084797	7.930628
H	8.653929	10.530810	8.925486
C	8.173823	8.821044	7.526263
H	7.565229	8.148550	8.134333
C	6.350462	5.280135	8.831106
H	5.584296	5.440299	9.600300

C	13.202735	7.917301	5.816181
H	13.782062	7.183728	5.222653
H	12.826676	8.692010	5.120193
C	9.257679	8.375811	11.096318
H	9.237338	8.252434	10.004703
C	8.707979	4.968566	8.238383
C	8.323949	4.872450	6.878996
H	9.092750	4.722282	6.112073
C	13.064821	6.951019	11.961435
C	13.576129	8.247345	12.206240
H	13.038531	9.118979	11.816881
C	13.785404	5.834848	12.436072
H	13.416813	4.823372	12.228893
C	15.473417	7.297345	13.405957
H	16.405774	7.431783	13.967936
C	14.978354	6.006024	13.157088
H	15.525407	5.126577	13.518351
C	14.770953	8.414791	12.923229
H	15.156038	9.425920	13.104292
C	13.480768	9.653052	7.720769
H	13.074271	10.453435	7.072806
H	14.255096	10.103861	8.370875
C	10.408143	4.321950	10.427211
H	9.932400	3.349177	10.622939
H	10.869566	4.792036	12.476763

Table S52. $[CpNi^{III}(pdt)Fe^{II}(dppv)(CO)]^{2+}$ with pdt Oriented Toward Ni ($[\mathbf{1b}]^{2+}$, –3202.66201673 E_h)

atom	x	y	z
Ni	10.394333	8.728738	7.402955
Fe	11.567755	6.899220	8.721660
P	11.385208	6.766558	11.065829
S	12.031005	9.107471	8.815095
P	10.453148	4.850540	8.734269
S	11.443282	7.040630	6.467737
C	10.836123	1.096459	7.100193
H	10.222230	0.191504	7.047806
C	12.119574	1.115320	6.526312
H	12.507493	0.222041	6.026281
C	7.962748	8.874597	11.961357
H	6.974997	9.015861	11.509295
C	7.663419	5.218091	9.118919
H	7.881930	5.279386	10.190719
C	8.257012	9.462069	13.205051
H	7.502108	10.068260	13.716148
C	9.526398	10.653950	7.165594
H	10.052525	11.596265	7.328698
C	9.515367	9.263041	13.795897
H	9.743192	9.709644	14.769128
C	13.223118	6.332093	8.799943
C	12.408045	3.425188	7.233573
H	13.029654	4.322559	7.280248
C	9.417654	9.947949	5.939607
H	9.834062	10.242889	4.975386
C	13.409743	9.136883	6.286817
H	12.650019	9.822038	5.868552
H	14.373548	9.406401	5.808811
C	6.037180	5.214355	7.304823
H	4.999213	5.283797	6.963293
C	11.121246	3.411953	7.811503
C	12.902455	2.278718	6.591530
H	13.901173	2.296409	6.143535
C	10.487797	8.482092	13.150973
H	11.458194	8.322198	13.629595
C	8.665771	8.735092	6.208168
H	8.429745	7.962155	5.474952
O	14.351212	6.029856	8.815146
C	7.070353	4.995724	6.374610
H	6.836463	4.886527	5.310248
C	10.765908	5.088324	11.469713
C	10.198869	7.898442	11.896793
C	10.332097	2.237755	7.738822
H	9.324247	2.214109	8.167884
C	8.819803	9.893169	8.193120
H	8.738198	10.180180	9.242861
C	8.250975	8.737044	7.584227
H	7.650976	7.964526	8.066640
C	6.334601	5.320877	8.675066
H	5.528966	5.462644	9.403127

C	13.092349	7.698468	5.867624
H	13.863692	6.986780	6.207539
H	13.024593	7.629521	4.768408
C	8.929568	8.098570	11.305727
H	8.697348	7.634503	10.340353
C	8.703573	4.994817	8.186793
C	8.400275	4.885753	6.808387
H	9.194374	4.679768	6.082277
C	12.993376	6.923223	11.920691
C	13.593981	8.196866	12.068510
H	13.073292	9.100081	11.730549
C	13.676666	5.769614	12.367956
H	13.233248	4.774951	12.248838
C	15.524631	7.159219	13.129031
H	16.506340	7.251760	13.604530
C	14.936887	5.893436	12.973715
H	15.456801	4.997205	13.326931
C	14.852821	8.308907	12.676261
H	15.307639	9.296825	12.802695
C	13.569884	9.398262	7.787302
H	13.813536	10.460098	7.962752
H	14.369819	8.785354	8.236515
C	10.344606	4.282392	10.476125
H	9.936636	3.281070	10.671105
H	10.703764	4.795786	12.527015

Table S53. $[CpNi^{III}(pdt)Fe^{II}(dppv)(CO)]^{2+}$ with pdt Oriented Toward Fe ($[\mathbf{1b}]^{2+}$, –3202.65799897 E_h)

atom	x	y	z
Ni	10.420149	8.709988	7.443894
Fe	11.629452	6.871471	8.721284
P	11.403817	6.769485	11.066426
S	12.100271	9.082098	8.794709
P	10.460635	4.853788	8.722574
S	11.453873	7.046002	6.469413
C	10.855794	1.075622	7.153117
H	10.271140	0.150198	7.169975
C	12.086124	1.123628	6.473985
H	12.462573	0.232107	5.962212
C	7.946036	8.832152	11.933095
H	6.952524	8.941935	11.484999
C	7.669185	5.207198	9.118251
H	7.889136	5.265413	10.189883
C	8.236831	9.456022	13.159867
H	7.472972	10.058486	13.662004
C	9.575939	10.645626	7.241044
H	10.116501	11.576875	7.420054
C	9.503541	9.298139	13.745308
H	9.729151	9.772962	14.705603
C	13.236270	6.172563	8.876796
C	12.350853	3.458261	7.105982
H	12.939493	4.378608	7.077812
C	9.461465	9.959632	6.003368
H	9.887608	10.262592	5.045862
C	14.035095	8.379146	6.730973
H	14.840543	8.827457	6.113884
H	14.516592	7.574626	7.308515
C	6.039265	5.197389	7.307575
H	5.000108	5.260741	6.968525
C	11.119649	3.414554	7.792890
C	12.829383	2.314014	6.447786
H	13.785178	2.354503	5.915544
C	10.487243	8.521611	13.112377
H	11.463365	8.392359	13.588245
C	8.685196	8.758201	6.251107
H	8.438647	8.000071	5.505941
O	14.302033	5.714792	9.006435
C	7.071870	4.989716	6.374339
H	6.836579	4.883008	5.310081
C	10.780580	5.090797	11.460552
C	10.201230	7.901119	11.875116
C	10.367454	2.214168	7.808131
H	9.397807	2.167860	8.316504
C	8.847577	9.885267	8.252725
H	8.765354	10.159046	9.305749
C	8.262802	8.749552	7.624618
H	7.648772	7.979768	8.093236
C	6.338694	5.300550	8.677633
H	5.533484	5.432777	9.407948

C	13.018249	7.785557	5.753698
H	13.479331	6.962097	5.180461
H	12.654091	8.534002	5.027917
C	8.924147	8.061133	11.288566
H	8.695709	7.572087	10.335044
C	8.708696	4.996056	8.182725
C	8.403323	4.887724	6.804895
H	9.197247	4.690046	6.076302
C	12.994599	6.935288	11.950944
C	13.603207	8.208208	12.071009
H	13.100275	9.103530	11.688012
C	13.655491	5.792052	12.454507
H	13.208022	4.797178	12.355906
C	15.495627	7.191364	13.216713
H	16.465628	7.291749	13.714148
C	14.900567	5.926039	13.088659
H	15.403299	5.037835	13.484398
C	14.846519	8.330609	12.707686
H	15.307140	9.318379	12.812433
C	13.548975	9.479019	7.677042
H	13.231844	10.385988	7.132616
H	14.360273	9.768050	8.367817
C	10.359541	4.286683	10.465142
H	9.949850	3.286499	10.662054
H	10.714383	4.794987	12.516682

Table S54. $[CpNi^{II}(Me_2pdt)Fe^{II}(dppe)(CO)]^+$ with pdt Oriented Toward Ni ($[\mathbf{1c}]^+$, –3282.83197161 E_h)

atom	x	y	z
Ni	10.523474	8.725938	7.620294
Fe	11.750575	6.768791	8.716330
P	11.355233	6.700785	10.952905
S	12.327991	8.960389	8.850132
P	10.629513	4.808392	8.585478
S	11.570415	7.162965	6.491550
C	10.906086	1.204438	6.574648
H	10.329908	0.273682	6.537586
C	12.076247	1.342633	5.808217
H	12.415496	0.517959	5.172384
C	7.868792	8.683179	12.091242
H	6.789617	8.502310	12.039814
C	7.825557	4.623919	9.207718
H	8.107416	4.395094	10.240764
C	8.359310	9.868102	12.665866
H	7.664108	10.613576	13.066221
C	9.495333	10.622145	7.620127
H	9.986877	11.590035	7.731365
C	9.745270	10.088947	12.727543
H	10.136787	11.008123	13.176605
C	13.284523	5.987962	8.802656
C	12.371958	3.598846	6.671213
H	12.947796	4.526878	6.700450
C	9.206475	9.957222	6.404308
H	9.422477	10.313211	5.396186
C	13.731577	9.179580	6.248309
C	6.067672	4.907713	7.545960
H	5.005041	4.926444	7.282196
C	11.202196	3.467657	7.446458
C	12.805531	2.540057	5.856109
H	13.715835	2.654890	5.257971
C	10.639094	9.126207	12.230655
H	11.717064	9.301136	12.304049
C	8.614013	8.677098	6.740599
H	8.305879	7.914367	6.024701
O	14.365934	5.517229	8.791662
C	10.803298	4.006416	10.271362
H	10.122617	3.142063	10.372754
H	11.834439	3.608720	10.278676
C	7.045181	5.113352	6.555913
H	6.745175	5.281894	5.515645
C	10.611090	5.022236	11.408455
H	11.084506	4.662958	12.337871
H	9.542681	5.178458	11.630749
C	10.155305	7.927853	11.658221
C	10.467556	2.260251	7.385657
H	9.543196	2.147328	7.963478
C	9.061614	9.768016	8.717276
H	9.163393	10.001328	9.777932
C	8.467140	8.592556	8.165619

H	8.033905	7.759432	8.716797
C	6.460755	4.660121	8.869964
H	5.706794	4.477096	9.643324
C	13.186558	7.801940	5.806351
H	13.917773	7.010431	6.051703
H	13.043294	7.799989	4.710759
C	8.758964	7.724377	11.582044
H	8.350518	6.818765	11.119370
C	8.813854	4.841600	8.223587
C	8.406936	5.084342	6.890379
H	9.162020	5.226354	6.108291
C	12.790431	6.870609	12.112607
C	14.054237	7.309515	11.670466
H	14.208825	7.555028	10.617191
C	12.608615	6.579838	13.485616
H	11.625973	6.272937	13.862115
C	14.936359	7.128750	13.931410
H	15.769741	7.223359	14.635586
C	13.675664	6.703924	14.385875
H	13.520893	6.472719	15.445202
C	15.121661	7.436043	12.575655
H	16.099641	7.772835	12.215604
C	13.829312	9.287915	7.786947
H	14.143949	10.311166	8.060629
H	14.594122	8.586745	8.167140
C	12.902417	10.335518	5.654225
H	13.345303	11.309847	5.929108
H	12.883664	10.271960	4.551188
H	11.862133	10.315890	6.018030
C	15.180756	9.283058	5.698521
H	15.827848	8.478212	6.091837
H	15.183323	9.216699	4.595625
H	15.632897	10.252555	5.974957

Table S55. $[CpNi^{II}(Me_2pdt)Fe^{II}(dppe)(CO)]^+$ with pdt Oriented Toward Fe ($[\mathbf{1c}]^+$, –3282.82962339 E_h)

atom	x	y	z
Ni	6.844859	9.044809	8.267013
Fe	6.321722	11.456350	7.672758
S	8.346696	10.646651	8.281713
P	6.226015	12.635279	9.610918
S	6.261839	9.699156	6.253436
P	4.080400	11.728083	7.536047
O	6.620804	13.915320	6.148280
C	6.469545	11.740172	11.217083
C	7.744122	11.736203	11.827131
H	8.570238	12.291957	11.372756
C	2.950016	10.450150	8.252728
C	5.629412	10.336372	13.035185
H	4.799846	9.796829	13.504981
C	5.628966	7.347104	8.526170
H	4.794862	7.101821	7.867980
C	6.510384	12.916886	6.765422
C	9.097818	10.109081	5.452363
C	7.955030	11.039719	13.028424
H	8.946634	11.053725	13.493484
C	6.868829	8.262349	10.233862
H	7.170987	8.837278	11.109839
C	7.763982	7.474503	9.394166
H	8.840768	7.375257	9.542020
C	9.529801	10.113992	6.936125
H	10.400272	10.784846	7.061147
H	9.848165	9.102345	7.246378
C	10.203074	9.342527	4.672274
H	10.317203	8.304673	5.034545
H	11.178180	9.851288	4.778241
H	9.963358	9.304404	3.594325
C	4.505858	13.406574	9.779593
H	3.984673	12.883260	10.598035
H	4.621907	14.458121	10.092383
C	5.545269	8.134454	9.722483
H	4.643683	8.598752	10.119891
C	1.895990	8.256208	8.018842
H	1.737030	7.341446	7.437087
C	7.781849	9.330074	5.229256
H	7.946211	8.247707	5.378470
H	7.444975	9.470927	4.185283
C	5.418642	11.018972	11.826796
H	4.427718	10.974598	11.361387
C	9.017047	11.537862	4.879684
H	9.989388	12.052000	4.986570
H	8.254999	12.147895	5.382725
H	8.772498	11.509097	3.802424
C	2.752433	9.252108	7.527179
H	3.254576	9.110178	6.563237
C	3.716341	13.316252	8.463049
H	4.031842	14.114908	7.767826

H	2.630927	13.440215	8.626537
C	2.264852	10.628304	9.473782
H	2.365725	11.560244	10.039703
C	6.998894	6.897789	8.352061
H	7.365649	6.261235	7.545640
C	1.227804	8.436841	9.243072
H	0.555833	7.660088	9.622877
C	1.411126	9.625222	9.966552
H	0.877018	9.784089	10.909683
C	6.898140	10.345271	13.639862
H	7.060481	9.815357	14.584422
C	9.339799	15.354314	9.109815
H	10.185800	15.490155	8.427643
C	9.115868	16.271599	10.146996
H	9.786013	17.127849	10.277763
C	8.031191	16.087090	11.022002
H	7.856320	16.794131	11.839970
C	7.170932	14.993274	10.854376
H	6.345277	14.850187	11.561069
C	8.477214	14.258676	8.936682
H	8.661046	13.547639	8.126826
C	3.436952	12.394281	3.478857
H	4.040165	12.456520	2.566790
C	4.060541	12.114260	4.705989
H	5.141919	11.958421	4.740172
C	3.297039	12.030172	5.888096
C	1.897187	12.222182	5.820743
H	1.284278	12.138119	6.725369
C	1.280097	12.506477	4.594709
H	0.195790	12.655907	4.555061
C	2.049345	12.593085	3.421478
H	1.565112	12.812003	2.463844
C	7.379180	14.072405	9.800438

Table S56. $[CpNi^{I}(Me_2pdt)Fe^{II}(dppe)(CO)]^0$ with pdt Oriented Toward Ni ($[\mathbf{1c}]^0$, –3282.99649843 E_h)

atom	x	y	z
Ni	10.492675	8.685293	7.653980
Fe	11.788570	6.791168	8.683987
P	11.358316	6.750333	10.892459
S	12.423256	8.980204	8.862982
P	10.646353	4.867315	8.561122
S	11.626980	7.126822	6.434762
C	10.903035	1.197889	6.624500
H	10.321885	0.268774	6.603930
C	12.077956	1.313981	5.862032
H	12.417557	0.474230	5.245052
C	7.880858	8.697425	12.135012
H	6.801264	8.516576	12.082949
C	7.842269	4.531467	9.172597
H	8.134049	4.262311	10.193510
C	8.372530	9.870368	12.731892
H	7.679066	10.606107	13.154206
C	9.420780	10.624979	7.669736
H	9.936485	11.580322	7.785595
C	9.757744	10.096877	12.774494
H	10.152217	11.012906	13.228769
C	13.330500	6.011341	8.763733
C	12.375471	3.589047	6.678159
H	12.943317	4.522021	6.687796
C	9.150102	9.958182	6.425494
H	9.402525	10.328880	5.429916
C	13.777076	9.153745	6.242651
C	6.071961	4.853728	7.531849
H	5.008126	4.857686	7.269872
C	11.202543	3.480252	7.450754
C	12.809957	2.510537	5.889131
H	13.723153	2.611438	5.291800
C	10.647890	9.149525	12.243362
H	11.725680	9.333751	12.291207
C	8.484278	8.725975	6.718267
H	8.165056	7.980395	5.989821
O	14.421359	5.546724	8.738702
C	10.814020	4.051789	10.246780
H	10.143490	3.178872	10.348850
H	11.851178	3.670426	10.260622
C	7.041438	5.156192	6.559045
H	6.734647	5.392409	5.533706
C	10.604124	5.074506	11.373174
H	11.058064	4.725101	12.316402
H	9.530385	5.232643	11.568523
C	10.164935	7.959857	11.653949
C	10.465863	2.274580	7.409536
H	9.536927	2.180944	7.984052
C	8.946604	9.795585	8.731062
H	9.020242	10.010832	9.796996
C	8.384709	8.606512	8.142727

H	7.927523	7.776253	8.681564
C	6.475913	4.542121	8.838469
H	5.729500	4.295099	9.602304
C	13.258108	7.766873	5.794541
H	14.004769	6.993207	6.052892
H	13.149852	7.764647	4.693306
C	8.768632	7.756613	11.590638
H	8.358497	6.868996	11.095698
C	8.822597	4.841290	8.206100
C	8.403095	5.155807	6.892544
H	9.152474	5.397234	6.129561
C	12.777596	6.894353	12.094233
C	14.059989	7.286912	11.664944
H	14.223145	7.527613	10.612049
C	12.575303	6.618724	13.466607
H	11.577231	6.349385	13.831960
C	14.915222	7.087833	13.936476
H	15.744874	7.158290	14.649044
C	13.635996	6.709313	14.379331
H	13.461837	6.489309	15.438864
C	15.122314	7.381190	12.580605
H	16.114266	7.684765	12.227512
C	13.917435	9.248219	7.780378
H	14.280004	10.260268	8.043008
H	14.679396	8.525412	8.126266
C	12.898624	10.291898	5.688668
H	13.317787	11.276668	5.968746
H	12.849737	10.242834	4.584521
H	11.871046	10.227734	6.084119
C	15.205182	9.305815	5.652969
H	15.883970	8.513660	6.020374
H	15.181246	9.249592	4.548565
H	15.640768	10.284151	5.929620

Table S57. $[CpNi^{I}(Me_2pdt)Fe^{II}(dppe)(CO)]^0$ with pdt Oriented Toward Fe ($[\mathbf{1c}]^0$, –3282.99012011 E_h)

atom	x	y	z
Ni	10.524619	8.566196	7.752792
Fe	11.888558	6.712790	8.729138
P	11.398985	6.724246	10.922183
S	12.473113	8.911941	8.902673
P	10.670780	4.837853	8.597629
S	11.667339	7.044977	6.486381
C	10.856213	1.180102	6.633648
H	10.259219	0.261280	6.606771
C	12.031722	1.282252	5.870088
H	12.355703	0.441530	5.246049
C	7.880196	8.645279	12.071601
H	6.803791	8.458105	11.986613
C	7.867204	4.540167	9.214393
H	8.159418	4.245793	10.228180
C	8.346832	9.811994	12.700487
H	7.636559	10.535503	13.115993
C	9.599257	10.570383	7.716803
H	10.181249	11.478717	7.886225
C	9.728269	10.048851	12.783081
H	10.103127	10.960924	13.261224
C	13.313012	5.757423	8.985187
C	12.371135	3.545569	6.702904
H	12.950972	4.471870	6.716787
C	9.389813	9.927009	6.450075
H	9.756823	10.276188	5.482431
C	14.103816	8.759215	6.408640
C	6.095229	4.919113	7.587962
H	5.030556	4.941212	7.330208
C	11.197605	3.449387	7.476845
C	12.784537	2.465771	5.904637
H	13.697868	2.555177	5.305680
C	10.639968	9.115846	12.262745
H	11.714558	9.306673	12.343540
C	8.587397	8.760095	6.677436
H	8.271614	8.044031	5.918104
O	14.275397	5.087485	9.162832
C	10.827209	4.021077	10.282127
H	10.138066	3.163581	10.388035
H	11.855285	3.616643	10.291397
C	7.064492	5.231636	6.618160
H	6.756551	5.495324	5.599980
C	10.644232	5.049446	11.409170
H	11.115008	4.699843	12.343960
H	9.575634	5.215385	11.625169
C	10.181340	7.932503	11.643631
C	10.439057	2.257926	7.428042
H	9.509841	2.176171	8.003967
C	8.954664	9.789132	8.725651
H	8.951001	10.001577	9.794608
C	8.342472	8.658560	8.083261

H	7.769740	7.870868	8.574237
C	6.499797	4.574415	8.886067
H	5.753354	4.319733	9.647301
C	12.941456	8.107567	5.625007
H	13.367124	7.465050	4.830933
H	12.346952	8.896735	5.129389
C	8.789223	7.720342	11.535748
H	8.400698	6.840152	11.011240
C	8.846666	4.860186	8.250616
C	8.427218	5.208599	6.946101
H	9.176751	5.461675	6.187317
C	12.801593	6.896825	12.137971
C	14.081635	7.304691	11.714545
H	14.247357	7.541216	10.660096
C	12.591286	6.626953	13.510058
H	11.594607	6.343879	13.868844
C	14.919996	7.134332	13.994951
H	15.742772	7.221761	14.713607
C	13.643239	6.739553	14.430543
H	13.464192	6.523513	15.490056
C	15.134145	7.421277	12.638668
H	16.124739	7.736488	12.291982
C	13.607587	9.617397	7.595292
H	13.057390	10.493775	7.205968
H	14.487726	9.998791	8.147216
C	14.806314	9.735047	5.423434
H	15.659548	10.239648	5.914104
H	15.199352	9.189312	4.545344
H	14.112517	10.514850	5.058445
C	15.143728	7.714444	6.857460
H	14.718143	6.987591	7.561368
H	15.534518	7.157812	5.985009
H	16.000437	8.209341	7.352346

Table S58. $[CpNi^{I}(Me_2pdt)Fe^{I}(dppe)(CO)]^{-}$ with pdt Oriented Toward Ni ($[\mathbf{1c}]^{-}$, –3283.02866040 E_h)

atom	x	y	z
Ni	10.478629	8.761776	7.613549
Fe	11.848082	6.726621	8.706592
P	11.392154	6.722411	10.864146
S	12.387957	8.988831	8.865989
P	10.693903	4.861488	8.597796
S	11.558134	7.142792	6.430315
C	10.917018	1.178740	6.629579
H	10.350134	0.239233	6.623221
C	12.060483	1.319170	5.824981
H	12.390795	0.488919	5.188277
C	7.838326	8.604969	11.991886
H	6.765922	8.418289	11.858082
C	7.894128	4.379129	9.224864
H	8.211651	4.003207	10.204227
C	8.275655	9.764655	12.656282
H	7.548190	10.483632	13.051728
C	9.415267	10.784713	7.605119
H	9.958555	11.721117	7.755703
C	9.653894	10.002443	12.787298
H	10.010340	10.911919	13.286725
C	13.423823	5.975896	8.739651
C	12.351485	3.593640	6.650377
H	12.898475	4.540238	6.658821
C	9.123766	10.173494	6.346580
H	9.396921	10.561325	5.361828
C	13.775090	9.092036	6.261805
C	6.080218	4.874212	7.675082
H	5.009387	4.902743	7.440837
C	11.209862	3.461590	7.464691
C	12.773518	2.528862	5.835361
H	13.661639	2.649949	5.203367
C	10.585483	9.079971	12.284068
H	11.656440	9.278049	12.394618
C	8.451048	8.935833	6.604288
H	8.125526	8.215951	5.850392
O	14.513651	5.486421	8.728380
C	10.875006	4.010084	10.272052
H	10.212212	3.130549	10.379020
H	11.916547	3.641291	10.275284
C	7.027198	5.303145	6.726098
H	6.694973	5.668367	5.746860
C	10.664122	5.036732	11.394257
H	11.135030	4.707507	12.338105
H	9.588913	5.182456	11.595876
C	10.159134	7.897294	11.637322
C	10.492741	2.245212	7.438715
H	9.584779	2.137366	8.044453
C	8.924328	9.927263	8.644856
H	8.996579	10.111525	9.717778
C	8.325835	8.779118	8.023727

H	7.843608	7.945209	8.535368
C	6.520133	4.416483	8.927143
H	5.793824	4.077052	9.676570
C	13.224799	7.713252	5.815019
H	13.948027	6.928086	6.108370
H	13.156767	7.707144	4.708720
C	8.768941	7.691392	11.476160
H	8.404562	6.820706	10.918179
C	8.855042	4.807955	8.281080
C	8.395430	5.280638	7.026798
H	9.124821	5.640876	6.291303
C	12.791680	6.913563	12.099656
C	14.073497	7.302836	11.666004
H	14.226687	7.513827	10.604143
C	12.593239	6.657428	13.475921
H	11.594279	6.390655	13.842009
C	14.929933	7.149396	13.942971
H	15.759836	7.237089	14.655190
C	13.653951	6.767786	14.390144
H	13.481188	6.560868	15.453856
C	15.134817	7.421899	12.580297
H	16.125282	7.727982	12.222531
C	13.906119	9.199082	7.802911
H	14.310543	10.201687	8.048180
H	14.640710	8.448573	8.152927
C	12.927421	10.245886	5.692359
H	13.365813	11.224323	5.971100
H	12.885972	10.189328	4.586775
H	11.895213	10.200821	6.083182
C	15.210074	9.209174	5.681031
H	15.866300	8.402367	6.059293
H	15.193947	9.146116	4.575431
H	15.667995	10.179247	5.956455

Table S59. $[CpNi^{I}(Me_2pdt)Fe^{I}(dppe)(CO)]^{-}$ with pdt Oriented Toward Fe ($[\mathbf{1c}]^{-}$, –3283.01962480 E_h)

atom	x	y	z
Ni	10.585685	8.692379	7.753356
Fe	11.945569	6.689170	8.748400
P	11.350609	6.764153	10.922520
S	12.580083	8.882782	8.834521
P	10.760533	4.840451	8.639189
S	11.568779	7.088213	6.405379
C	11.010392	1.141824	6.707286
H	10.458964	0.193274	6.713771
C	12.144796	1.294450	5.891366
H	12.482704	0.463821	5.259165
C	7.741413	8.574106	11.977756
H	6.676914	8.381371	11.795819
C	7.966583	4.284729	9.204379
H	8.285549	3.835534	10.151507
C	8.138805	9.721522	12.687732
H	7.389719	10.425757	13.068734
C	9.723691	10.769547	7.776202
H	10.351883	11.640505	7.980579
C	9.510705	9.964968	12.880686
H	9.839457	10.863050	13.418697
C	13.404461	5.771511	9.022900
C	12.407909	3.581084	6.692972
H	12.929077	4.543339	6.680592
C	9.442079	10.218667	6.483596
H	9.805144	10.600516	5.525702
C	14.077408	8.623070	6.239075
C	6.153844	4.867991	7.686417
H	5.084538	4.887966	7.443648
C	11.277304	3.433458	7.521117
C	12.837892	2.515225	5.883266
H	13.717224	2.644351	5.240764
C	10.469028	9.063758	12.393301
H	11.532566	9.269260	12.554580
C	8.616909	9.065604	6.673656
H	8.268491	8.394278	5.885706
O	14.381103	5.122374	9.251299
C	10.917306	4.020717	10.325350
H	10.279641	3.122698	10.430102
H	11.968921	3.684553	10.365603
C	7.094868	5.407054	6.790924
H	6.760873	5.852826	5.846459
C	10.637763	5.061174	11.419960
H	11.067502	4.747768	12.388347
H	9.551478	5.188478	11.567385
C	10.082980	7.888442	11.704063
C	10.577091	2.207828	7.511824
H	9.676860	2.090154	8.127392
C	9.072755	9.959808	8.763889
H	9.100143	10.115695	9.843073
C	8.396553	8.895688	8.080316

H	7.800084	8.108401	8.543336
C	6.593962	4.311156	8.896724
H	5.870879	3.889694	9.606257
C	12.755041	8.237725	5.536162
H	13.006513	7.742374	4.576345
H	12.187517	9.157660	5.300964
C	8.698934	7.679314	11.480189
H	8.362637	6.817499	10.891608
C	8.920576	4.823434	8.313227
C	8.462115	5.394471	7.102185
H	9.186085	5.844151	6.411777
C	12.725120	6.950695	12.186930
C	14.006602	7.376570	11.785990
H	14.174828	7.614613	10.731362
C	12.504843	6.671027	13.555485
H	11.505755	6.373634	13.896367
C	14.816890	7.217822	14.079842
H	15.628384	7.318800	14.811322
C	13.541208	6.799734	14.494490
H	13.349791	6.577992	15.552018
C	15.044310	7.510050	12.725261
H	16.034617	7.844034	12.392776
C	13.847447	9.408019	7.560032
H	13.533462	10.439021	7.306368
H	14.817323	9.485159	8.091267
C	14.835593	9.586893	5.284752
H	15.782105	9.933567	5.743758
H	15.088444	9.080068	4.333174
H	14.226727	10.479281	5.045055
C	14.965140	7.383040	6.459624
H	14.475497	6.660066	7.126940
H	15.172834	6.880867	5.494400
H	15.935297	7.669794	6.910077

Table S60. $[CpNi^{III}(Me_2pdt)Fe^{II}(dppe)(CO)]^{2+}$ with pdt Oriented Toward Ni ($[\mathbf{1c}]^{2+}$, –3282.50779488 E_h)

atom	x	y	z
Ni	10.494606	8.771781	7.591556
Fe	11.664983	6.835433	8.692741
P	11.335292	6.694550	11.015293
S	12.252283	9.028945	8.874118
P	10.608291	4.751402	8.601306
S	11.481390	7.179393	6.455157
C	10.924403	1.226425	6.513746
H	10.344400	0.300587	6.443121
C	12.115192	1.373274	5.781248
H	12.465544	0.559531	5.138199
C	7.846424	8.689944	12.061081
H	6.767406	8.513616	11.999409
C	7.819078	4.629434	9.249782
H	8.100506	4.396701	10.281533
C	8.336167	9.865218	12.659104
H	7.638200	10.606031	13.062576
C	9.499613	10.659885	7.586771
H	10.000307	11.623139	7.699081
C	9.722500	10.081758	12.747495
H	10.108429	10.989861	13.222117
C	13.272952	6.158102	8.695883
C	12.404810	3.609764	6.694292
H	12.989772	4.530598	6.752978
C	9.218541	9.990542	6.367146
H	9.451702	10.340025	5.360355
C	13.718213	9.117000	6.297647
C	6.061425	4.899539	7.584588
H	4.998561	4.911963	7.322499
C	11.213181	3.467568	7.435430
C	12.851896	2.564598	5.870216
H	13.777518	2.683371	5.297785
C	10.621446	9.123819	12.251865
H	11.698817	9.288251	12.353749
C	8.586924	8.727433	6.700983
H	8.269307	7.970278	5.982843
O	14.369211	5.751082	8.651440
C	10.836207	3.999127	10.294858
H	10.189632	3.109488	10.402433
H	11.880737	3.637443	10.302154
C	7.035544	5.096308	6.588570
H	6.731926	5.246700	5.546965
C	10.601811	5.012079	11.424879
H	11.059525	4.654325	12.362857
H	9.528387	5.158752	11.625848
C	10.135072	7.934886	11.659021
C	10.468117	2.267107	7.334307
H	9.529459	2.144989	7.885941
C	9.024659	9.822146	8.680686
H	9.108969	10.061045	9.742178
C	8.417572	8.655570	8.124242

H	7.955222	7.836723	8.673798
C	6.455375	4.660647	8.911523
H	5.702046	4.474699	9.684142
C	13.159936	7.745886	5.843146
H	13.853003	6.931335	6.116169
H	13.050232	7.740620	4.744399
C	8.738336	7.733673	11.551970
H	8.335136	6.830447	11.080196
C	8.803556	4.836831	8.256690
C	8.398180	5.069599	6.919078
H	9.150109	5.186637	6.130009
C	12.805917	6.865885	12.107623
C	14.042949	7.359535	11.643799
H	14.164954	7.649489	10.597526
C	12.666783	6.523841	13.475475
H	11.706780	6.175559	13.872920
C	14.990453	7.129640	13.872656
H	15.839919	7.225387	14.556457
C	13.755987	6.653115	14.347857
H	13.637915	6.383964	15.402425
C	15.130378	7.487971	12.523421
H	16.087798	7.865981	12.150638
C	13.798014	9.258463	7.837517
H	14.130578	10.279975	8.093292
H	14.533747	8.550270	8.256863
C	12.945249	10.288808	5.660465
H	13.414011	11.251658	5.929420
H	12.957116	10.207139	4.559383
H	11.893562	10.318031	5.989833
C	15.190507	9.163952	5.788516
H	15.802163	8.346894	6.210506
H	15.219264	9.083223	4.687889
H	15.661307	10.124219	6.062309

Table S61. $[CpNi^{III}(Me_2pdt)Fe^{II}(dppe)(CO)]^{2+}$ with pdt Oriented Toward Fe ($[\mathbf{1c}]^{2+}$, –3282.49907273 E_h)

atom	x	y	z
Ni	10.452161	8.636393	7.706434
Fe	11.832651	6.760167	8.718675
P	11.347994	6.715521	11.048407
S	12.274474	8.979484	8.876427
P	10.587544	4.748881	8.595646
S	11.518436	7.144215	6.507606
C	11.021149	1.165895	6.639567
H	10.479286	0.214891	6.616797
C	12.188058	1.334826	5.874014
H	12.558191	0.512619	5.253193
C	7.945355	8.822770	12.152336
H	6.860041	8.684030	12.107770
C	7.782336	4.761715	9.144895
H	8.023240	4.623165	10.203635
C	8.485302	9.975738	12.751158
H	7.819870	10.736257	13.172490
C	9.539173	10.564224	7.609777
H	10.070986	11.511927	7.711433
C	9.878863	10.144065	12.817022
H	10.303995	11.035227	13.290093
C	13.250563	5.745877	9.025394
C	12.404411	3.614662	6.697197
H	12.946900	4.564002	6.704801
C	9.277874	9.868020	6.404206
H	9.560965	10.176268	5.396586
C	14.034299	8.754330	6.473466
C	6.089106	4.925618	7.401628
H	5.036162	4.943727	7.102462
C	11.237937	3.448708	7.473178
C	12.874703	2.558599	5.900365
H	13.779907	2.694223	5.299730
C	10.735669	9.158827	12.300793
H	11.819409	9.284780	12.387705
C	8.576476	8.642492	6.754759
H	8.250357	7.876155	6.050367
O	14.157782	5.045025	9.250065
C	10.736963	4.025198	10.311523
H	10.036073	3.178302	10.422145
H	11.757165	3.603076	10.348516
C	7.100272	5.001448	6.426733
H	6.835967	5.065644	5.365783
C	10.534566	5.062710	11.426539
H	10.955457	4.687748	12.375313
H	9.466168	5.260566	11.608251
C	10.198883	7.993511	11.704428
C	10.539593	2.216744	7.432097
H	9.617980	2.077048	8.007887
C	8.990250	9.781272	8.712196
H	9.044686	10.053784	9.767151
C	8.351374	8.626946	8.169991

H	7.835777	7.844524	8.725344
C	6.432113	4.799985	8.758148
H	5.648606	4.708964	9.517662
C	12.901983	8.086655	5.660173
H	13.333296	7.352298	4.956399
H	12.349326	8.834862	5.064703
C	8.794708	7.841930	11.618888
H	8.351301	6.960844	11.142080
C	8.803744	4.845220	8.171285
C	8.450595	4.962964	6.804846
H	9.229998	4.985714	6.034948
C	12.824396	6.826464	12.141260
C	14.064035	7.322034	11.683987
H	14.184669	7.634908	10.642836
C	12.684752	6.452754	13.500261
H	11.722892	6.104808	13.893219
C	15.014774	7.028873	13.904613
H	15.867013	7.100414	14.587927
C	13.778329	6.550069	14.371656
H	13.661262	6.254670	15.419286
C	15.153754	7.420454	12.564296
H	16.113176	7.800605	12.198894
C	13.521499	9.633543	7.636675
H	13.025105	10.543389	7.254910
H	14.374365	9.956294	8.260572
C	14.744966	9.731030	5.485168
H	15.586657	10.240077	5.986320
H	15.158032	9.175088	4.625497
H	14.056318	10.502762	5.098112
C	15.073375	7.725015	6.955118
H	14.648865	7.000254	7.665022
H	15.484838	7.158266	6.101570
H	15.917564	8.230516	7.456081

Table S62. [Cp'Ni^{II}(pdt)Fe^{II}(dppe)(CO)]$^{+}$ with pdt Oriented Toward Ni ([**1d**]$^{+}$, –3243.53112609 E_h)

atom	x	y	z
Ni	12.058793	8.651948	2.018238
Fe	10.977570	9.463622	4.213653
S	12.799230	10.348382	3.211936
S	10.019935	9.462892	2.161256
P	12.199960	9.028180	6.088662
P	9.456399	7.980526	5.004553
O	9.769977	11.890664	5.244190
C	11.791399	10.417054	8.566624
H	11.347459	9.501178	8.968821
C	4.907827	9.014940	5.224491
H	3.840890	9.252947	5.291534
C	5.384071	7.780408	5.699441
H	4.690549	7.053151	6.135140
C	15.040147	9.126886	6.524861
H	14.878659	10.083231	7.030973
C	15.483601	6.650321	5.270888
H	15.649754	5.678739	4.792242
C	6.748498	7.472106	5.611419
H	7.103802	6.497696	5.965633
C	13.758443	7.452714	1.846139
H	14.616801	7.548869	2.512891
C	13.948368	8.418195	5.982353
C	10.243641	10.873765	4.886411
C	8.684961	6.140454	3.016871
H	8.133018	6.988039	2.596106
C	9.316159	3.789288	2.918499
H	9.269197	2.814455	2.421749
C	8.634800	4.893014	2.376069
H	8.046116	4.778888	1.458972
C	12.285483	10.465681	7.246101
C	12.919093	12.801848	7.602842
H	13.360993	13.728911	7.222123
C	5.800583	9.938476	4.660800
H	5.434634	10.900105	4.285387
C	9.417171	6.302511	4.216615
C	12.423132	12.742830	8.916023
H	12.474827	13.624453	9.563453
C	11.753363	8.222699	-1.291108
H	10.663364	8.379539	-1.223518
H	11.923912	7.440018	-2.055054
H	12.219775	9.151421	-1.660242
C	11.370065	7.639970	7.039417
H	11.817582	6.713104	6.640969
H	11.638147	7.675984	8.109930
C	16.566323	7.366251	5.810896
H	17.580303	6.957032	5.749920
C	7.169984	9.635365	4.575514
H	7.857020	10.361982	4.135360
C	10.042052	3.938655	4.110708
H	10.559304	3.079597	4.551890

C	12.624992	6.602522	2.045397
H	12.441942	5.925255	2.878845
C	13.587875	8.158415	0.583289
H	14.300191	8.866822	0.155733
C	10.093093	5.186295	4.756197
H	10.645703	5.268560	5.697793
C	16.340700	8.602963	6.433799
H	17.178090	9.163589	6.862876
C	12.374859	11.893995	2.255440
H	11.943116	12.598081	2.989410
H	13.347105	12.298449	1.924048
C	11.716364	6.867769	0.973080
H	10.725802	6.423824	0.858513
C	11.856376	11.551409	9.392815
H	11.463576	11.498154	10.413779
C	10.055361	11.172567	1.411554
H	9.429126	11.100691	0.505567
H	9.542869	11.836813	2.130029
C	9.852309	7.665190	6.815159
H	9.388217	8.505869	7.362599
H	9.369017	6.735995	7.166516
C	12.334475	7.796580	0.027520
C	14.186056	7.177433	5.348119
H	13.350406	6.617930	4.911609
C	11.444035	11.705791	1.056605
H	11.921749	11.033012	0.320004
H	11.319265	12.689823	0.560081
C	7.656964	8.402795	5.054367
C	12.841388	11.676558	6.770005
H	13.217697	11.735586	5.742107

Table S63. [Cp'Ni^{II}(pdt)Fe^{II}(dppe)(CO)]$^+$ with pdt Oriented Toward Fe ([**1d**]$^+$, –3243.53221453 E_h)

atom	x	y	z
Ni	12.067660	8.632248	2.080501
Fe	10.954919	9.497154	4.224798
S	12.790833	10.362132	3.226488
S	10.031926	9.440754	2.155960
P	12.188778	9.039194	6.086009
P	9.444567	7.991765	4.987021
O	9.655707	11.794669	5.438724
C	11.897944	10.360286	8.620549
H	11.519846	9.419821	9.032736
C	4.896959	9.036863	5.178515
H	3.829944	9.276774	5.236732
C	5.378345	7.835091	5.726662
H	4.688689	7.134927	6.210201
C	15.032169	9.181148	6.447779
H	14.868048	10.156148	6.916026
C	15.483707	6.659776	5.291226
H	15.653109	5.671122	4.850246
C	6.742996	7.524686	5.650928
H	7.101434	6.574385	6.062813
C	13.793251	7.456845	1.890983
H	14.654801	7.561485	2.552220
C	13.938348	8.433919	5.963405
C	10.175475	10.843495	4.979610
C	8.652481	6.158954	3.000088
H	8.096967	7.007770	2.586959
C	9.279343	3.807287	2.889630
H	9.225552	2.833554	2.391412
C	8.593900	4.913048	2.356927
H	7.995532	4.802104	1.445791
C	12.287676	10.457347	7.268103
C	12.866103	12.806222	7.637034
H	13.245061	13.757423	7.248130
C	5.784578	9.924876	4.553103
H	5.414207	10.860414	4.120440
C	9.398758	6.317505	4.191281
C	12.472044	12.699271	8.981170
H	12.540772	13.567291	9.645190
C	11.741707	8.238860	-1.211907
H	10.647807	8.362368	-1.133403
H	11.927306	7.479632	-1.995777
H	12.179421	9.188682	-1.562676
C	11.359354	7.637252	7.015580
H	11.803894	6.715179	6.603682
H	11.627235	7.655009	8.086285
C	16.567735	7.412922	5.774873
H	17.585913	7.015151	5.708393
C	7.154258	9.620073	4.480185
H	7.838386	10.317782	3.990849
C	10.018335	3.953035	4.074298
H	10.538732	3.092187	4.508227

C	12.682232	6.577818	2.084484
H	12.521845	5.885970	2.910405
C	13.596324	8.179519	0.641802
H	14.290015	8.908550	0.218175
C	10.078971	5.199311	4.721176
H	10.642364	5.278707	5.656578
C	16.338000	8.672527	6.348542
H	17.176379	9.263195	6.733015
C	12.418834	11.830754	2.137894
H	12.943148	12.673347	2.623759
H	12.919761	11.638190	1.171993
C	11.758003	6.838826	1.026806
H	10.775990	6.375683	0.913902
C	11.984828	11.477093	9.468106
H	11.671792	11.386042	10.513668
C	10.123760	11.074552	1.261019
H	10.511991	10.852114	0.250383
H	9.071213	11.392422	1.152155
C	9.841210	7.667513	6.795020
H	9.380610	8.505666	7.349193
H	9.359015	6.736514	7.142649
C	12.345309	7.799874	0.092257
C	14.180287	7.171153	5.376184
H	13.345670	6.581551	4.979844
C	10.940664	12.167250	1.948289
H	10.875440	13.083591	1.327193
H	10.486734	12.414988	2.923499
C	7.646649	8.421244	5.033465
C	12.764956	11.697995	6.783980
H	13.058708	11.794029	5.732479

Table S64. $[Cp'Ni^{I}(pdt)Fe^{II}(dppe)(CO)]^{0}$ with pdt Oriented Toward Ni ($[\mathbf{1d}]^{0}$, –3243.69475486 E_h)

atom	x	y	z
Ni	2.418910	6.014624	2.113318
Fe	1.587422	5.966243	4.499639
S	0.976136	7.552749	2.988160
P	2.628793	7.504514	5.754164
O	-0.867106	5.881013	6.066544
C	4.535889	6.358528	1.823239
H	5.206282	6.638242	2.636561
C	3.825104	7.262718	0.972240
H	3.814205	8.350152	1.054769
C	3.045873	6.480629	0.056886
H	2.377144	6.871132	-0.714294
C	-0.727783	7.200883	2.322898
H	-1.423873	7.207476	3.181967
H	-0.972160	8.071887	1.688013
C	-0.872392	5.909074	1.515049
H	-0.162399	5.929232	0.666297
H	-1.896956	5.881798	1.085644
C	0.167000	5.911299	5.487040
C	3.069221	6.653634	7.369910
C	5.469418	8.040895	5.788424
H	5.547882	7.382389	6.660251
C	6.635572	8.654264	5.295339
H	7.598476	8.455388	5.779993
C	6.564090	9.525526	4.198441
H	7.471319	10.006941	3.816785
C	5.319858	9.780769	3.594420
H	5.254340	10.465806	2.741486
C	4.158391	9.161946	4.077215
H	3.193559	9.359584	3.595799
C	2.435369	9.978888	7.142189
H	3.513929	9.871279	7.305883
C	1.737145	9.012172	6.383648
C	0.357231	9.187295	6.159419
H	-0.189886	8.455424	5.560925
C	-0.313642	10.300858	6.692518
H	-1.386724	10.424514	6.507743
C	0.385501	11.251190	7.451933
H	-0.139106	12.120212	7.865202
C	1.763234	11.088092	7.675972
H	2.317969	11.829020	8.263109
H	3.730217	7.278760	7.997962
H	4.739218	5.337276	6.850361
H	2.104142	6.563145	7.900686
S	1.074696	4.361197	2.956344
C	-0.656533	4.626680	2.322902
H	-1.340283	4.589370	3.190991
H	-0.869133	3.744735	1.691692
C	3.668080	5.265062	7.103229
C	4.181965	3.528336	4.805858
C	5.430311	4.155226	4.594993

H	5.593598	5.188689	4.921936
C	6.470423	3.478756	3.939705
H	7.434780	3.978676	3.794442
C	6.272339	2.172866	3.459638
H	7.083801	1.646008	2.945231
C	5.024997	1.552734	3.635005
H	4.856175	0.538466	3.255544
C	3.987932	2.222302	4.305287
H	3.023753	1.723673	4.445438
C	2.590606	2.196845	7.387144
H	3.653745	2.369381	7.592388
C	1.903576	3.020346	6.465023
C	0.551277	2.742874	6.186509
H	0.016669	3.356505	5.457778
C	-0.106841	1.680185	6.829451
H	-1.160484	1.481568	6.603174
C	0.578967	0.880061	7.755207
H	0.063930	0.053860	8.258405
C	1.932881	1.138568	8.030602
H	2.479990	0.512031	8.744387
H	3.585186	4.617849	7.993156
C	4.234208	5.016667	1.392448
H	4.619678	4.099087	1.838830
C	3.320550	5.085643	0.293261
C	4.217567	8.282970	5.183810
P	2.786192	4.438547	5.636536
C	2.771645	3.920852	-0.486436
H	1.802873	4.169727	-0.954906
H	3.462323	3.619445	-1.299131
H	2.618521	3.041081	0.162566

Table S65. $[Cp'Ni^{I}(pdt)Fe^{II}(dppe)(CO)]^{0}$ with pdt Oriented Toward Fe ($[\mathbf{1d}]^{0}$, –3243.69298779 E_h)

atom	x	y	z
Ni	2.441699	6.009024	2.152533
Fe	1.564009	5.964721	4.513226
S	0.980855	7.547784	2.979778
P	2.622538	7.505509	5.743942
O	-0.746272	5.889819	6.294450
C	4.576444	6.343177	1.811498
H	5.271461	6.613274	2.607335
C	3.842890	7.257784	0.992643
H	3.841496	8.344617	1.083243
C	3.030341	6.487681	0.095365
H	2.341750	6.887676	-0.653261
C	-0.664426	7.190747	2.173162
H	-1.314810	8.047449	2.427846
H	-0.489876	7.213593	1.081559
C	-1.344049	5.883813	2.584612
H	-2.347377	5.854731	2.108506
H	-1.512640	5.881606	3.676168
C	0.211099	5.915001	5.595232
C	3.058259	6.656705	7.361721
C	5.462271	8.036338	5.784030
H	5.536776	7.378501	6.656682
C	6.630593	8.649655	5.296187
H	7.591132	8.451654	5.785824
C	6.564057	9.519631	4.197950
H	7.472950	10.001091	3.820316
C	5.322902	9.773477	3.587238
H	5.261478	10.457081	2.732881
C	4.159232	9.154659	4.064722
H	3.196897	9.350778	3.577809
C	2.424371	9.969067	7.147867
H	3.497584	9.847570	7.335625
C	1.732513	9.016869	6.365610
C	0.360753	9.210424	6.108962
H	-0.177866	8.490575	5.488066
C	-0.309112	10.327933	6.634953
H	-1.375819	10.466637	6.425529
C	0.382911	11.263244	7.419166
H	-0.141118	12.135042	7.827283
C	1.752956	11.082060	7.674534
H	2.302185	11.811729	8.280609
H	3.721863	7.279836	7.988788
H	4.724467	5.330209	6.853681
H	2.092463	6.572257	7.891994
S	1.083974	4.360215	2.955228
C	-0.593242	4.614871	2.177297
H	-1.190315	3.724603	2.447571
H	-0.434370	4.591792	1.083199
C	3.651355	5.264430	7.099772
C	4.176704	3.537323	4.797872
C	5.421305	4.172473	4.590953

H	5.574852	5.208388	4.914499
C	6.469369	3.500656	3.943776
H	7.430904	4.006685	3.801175
C	6.282516	2.191623	3.467750
H	7.100180	1.668387	2.959496
C	5.038589	1.563586	3.638768
H	4.878598	0.546915	3.261990
C	3.993565	2.228415	4.301412
H	3.032090	1.723821	4.438397
C	2.578518	2.193406	7.377196
H	3.638096	2.373328	7.594156
C	1.895263	3.012821	6.448828
C	0.547934	2.726165	6.155192
H	0.018099	3.337444	5.420446
C	-0.109049	1.657894	6.789756
H	-1.158818	1.451776	6.552370
C	0.572704	0.861568	7.721872
H	0.058292	0.031044	8.218542
C	1.921604	1.129460	8.012191
H	2.465516	0.505985	8.731060
H	3.559005	4.619069	7.990071
C	4.253637	5.008228	1.382347
H	4.646048	4.084780	1.810399
C	3.303895	5.090104	0.313658
C	4.213617	8.277440	5.172597
P	2.775701	4.437384	5.629592
C	2.727056	3.934801	-0.460268
H	1.751182	4.196210	-0.907256
H	3.396567	3.631444	-1.289887
H	2.578820	3.052212	0.186189

Table S66. [Cp'Ni^{I}(pdt)Fe^{I}(dppe)(CO)]$^{-}$ with pdt Oriented Toward Ni ([**1d**]$^{-}$, $-E_h$)

atom	x	y	z
Ni	2.424831	6.026410	2.016487
Fe	1.559902	5.978417	4.579562
S	1.042710	7.564710	2.966099
P	2.614350	7.474650	5.796996
O	-0.951623	5.906030	6.106366
C	4.527669	6.376167	1.580157
H	5.214680	6.685514	2.369148
C	3.786558	7.246838	0.715527
H	3.766040	8.337869	0.766138
C	3.020370	6.433737	-0.183923
H	2.331771	6.796890	-0.952660
C	-0.697482	7.210704	2.396220
H	-1.349195	7.208562	3.291151
H	-0.991447	8.071491	1.765491
C	-0.876926	5.910091	1.603923
H	-0.196948	5.926017	0.729826
H	-1.920304	5.881081	1.214819
C	0.085179	5.928095	5.513361
C	3.036619	6.643118	7.436302
C	5.454314	8.091909	5.969539
H	5.491434	7.516356	6.901500
C	6.641224	8.669078	5.483627
H	7.577886	8.529920	6.038393
C	6.626729	9.428350	4.302868
H	7.550985	9.880482	3.923663
C	5.414818	9.596844	3.606562
H	5.391870	10.181317	2.678985
C	4.234705	9.008732	4.080489
H	3.301287	9.121920	3.516228
C	2.427562	9.962162	7.215136
H	3.497503	9.832066	7.418046
C	1.746378	9.012468	6.422496
C	0.381172	9.216896	6.141590
H	-0.143813	8.489762	5.516193
C	-0.291221	10.341132	6.651551
H	-1.353023	10.488133	6.419415
C	0.392064	11.274620	7.447648
H	-0.133780	12.152242	7.844050
C	1.755315	11.082977	7.729114
H	2.298895	11.810366	8.345177
H	3.689695	7.264654	8.077871
H	4.711386	5.339989	6.909496
H	2.063207	6.546919	7.950114
S	1.140408	4.364629	2.940461
C	-0.625593	4.626649	2.405819
H	-1.262720	4.608556	3.311042
H	-0.890867	3.747859	1.787243
C	3.639071	5.258642	7.158498
C	4.182253	3.578661	4.844977
C	5.383021	4.274877	4.572151

H	5.479936	5.328985	4.858179
C	6.446291	3.646945	3.908068
H	7.369581	4.206369	3.714935
C	6.321647	2.316128	3.469624
H	7.149278	1.827977	2.941033
C	5.118738	1.627731	3.695869
H	5.000599	0.595661	3.342855
C	4.059950	2.251199	4.377101
H	3.129169	1.701365	4.549342
C	2.576372	2.177822	7.382375
H	3.639764	2.347846	7.590543
C	1.897507	3.012611	6.464972
C	0.545566	2.739942	6.180104
H	0.022274	3.376794	5.461523
C	-0.116867	1.668518	6.804183
H	-1.169660	1.472058	6.568095
C	0.563167	0.855147	7.726078
H	0.043974	0.022655	8.217109
C	1.914182	1.111301	8.013083
H	2.456812	0.477082	8.725470
H	3.551681	4.593485	8.036493
C	4.212868	5.025396	1.205832
H	4.608413	4.122267	1.675052
C	3.287449	5.058712	0.106558
C	4.230822	8.251963	5.278999
P	2.754491	4.472046	5.657614
C	2.705658	3.861639	-0.598210
H	1.857229	4.154353	-1.244463
H	3.450407	3.350000	-1.243820
H	2.330561	3.114773	0.126660

Table S67. [Cp'Ni^{I}(pdt)Fe^{I}(dppe)(CO)]$^{-}$ with pdt Oriented Toward Fe ([**1d**]$^{-}$, –3243.72112875 E_h)

atom	x	y	z
Ni	10.365344	8.676106	7.402488
Fe	11.781234	6.738697	8.784741
P	11.413896	6.756595	10.954051
S	12.053144	9.073015	8.901104
P	10.578463	4.882637	8.730467
S	11.636962	7.024125	6.495469
C	10.672919	1.194584	6.755649
H	10.045901	0.295334	6.706633
C	11.873865	1.258520	6.029106
H	12.189054	0.409010	5.410298
C	8.249131	9.332250	11.696822
H	7.460940	9.797929	11.093316
C	7.793236	4.459244	9.464669
H	8.133173	4.108738	10.445629
C	8.296531	9.545804	13.083923
H	7.542975	10.176706	13.571723
C	9.525851	10.715983	6.740439
H	10.134172	11.624630	6.710799
C	9.318013	8.951782	13.845262
H	9.363869	9.116077	14.929385
C	13.352026	5.978172	8.945555
C	12.260722	3.508308	6.883586
H	12.865522	4.418290	6.928601
C	9.353753	9.797382	5.657886
C	14.091243	8.507268	6.934376
H	14.973762	8.940295	6.410956
H	14.473015	7.702029	7.585617
C	5.939545	4.949349	7.960302
H	4.862261	4.995807	7.760836
C	11.060299	3.452486	7.620212
C	12.664547	2.417516	6.094417
H	13.598534	2.479583	5.522738
C	10.284136	8.146587	13.220791
H	11.081966	7.692497	13.819940
C	8.517617	8.730820	6.137598
H	8.208990	7.861462	5.552144
O	14.417535	5.450413	9.040142
C	10.800542	4.054444	10.417153
H	10.114547	3.197368	10.555683
H	11.830397	3.654037	10.393406
C	6.863763	5.328128	6.967779
H	6.507409	5.672115	5.989265
C	10.646072	5.108926	11.521963
H	11.069281	4.781851	12.490104
H	9.578196	5.334369	11.693397
C	10.241182	7.925651	11.826528
C	10.268652	2.285335	7.542747
H	9.320524	2.236882	8.091580
C	8.806290	10.223872	7.878335
H	8.784089	10.690107	8.865797

C	8.179192	8.987449	7.506402
H	7.541269	8.361568	8.133858
C	6.411974	4.520093	9.211041
H	5.704129	4.221627	9.994812
C	13.166312	7.905613	5.870407
H	13.733914	7.170442	5.267403
H	12.805252	8.696073	5.184947
C	9.216472	8.528127	11.069895
H	9.200740	8.375893	9.981762
C	8.732262	4.836637	8.477131
C	8.239921	5.284338	7.225315
H	8.950782	5.608603	6.456333
C	12.930873	6.942783	12.011590
C	13.468510	8.234956	12.224104
H	12.943216	9.106585	11.818665
C	13.641476	5.829121	12.506870
H	13.267745	4.814569	12.326609
C	15.360392	7.284325	13.432334
H	16.299123	7.416361	13.984106
C	14.842507	5.996644	13.215657
H	15.377587	5.116137	13.592034
C	14.671218	8.400338	12.928419
H	15.072240	9.409702	13.083171
C	13.469803	9.604068	7.804665
H	13.088871	10.428718	7.171680
H	14.247289	10.021122	8.473288
C	9.896083	9.940641	4.259325
H	10.251440	8.971169	3.862624
H	9.134196	10.324311	3.546919
H	10.748829	10.644364	4.232159

Table S68. [Cp'Ni^{III}(pdt)Fe^{II}(dppe)(CO)]$^{2+}$ with pdt Oriented Toward Ni ([**1d**]$^{2+}$, –3243.20978117 E_h)

atom	x	y	z
Ni	10.255351	8.732063	7.486629
Fe	11.512810	6.936698	8.763755
P	11.231458	6.812331	11.110996
S	11.888721	9.166757	8.888071
P	10.524860	4.814414	8.725487
S	11.384928	7.124800	6.503288
C	11.166344	1.141591	6.995481
H	10.627926	0.189579	6.946482
C	12.421019	1.277753	6.375881
H	12.862578	0.429299	5.843308
C	8.146228	9.332228	12.163083
H	7.091857	9.449526	11.890363
C	7.677095	4.914910	9.024085
H	7.823055	4.905147	10.108563
C	8.714131	10.154317	13.153678
H	8.104268	10.918694	13.645945
C	9.297238	10.603728	7.132658
H	9.789163	11.578324	7.155424
C	10.058978	9.984560	13.519030
H	10.500442	10.612526	14.299591
C	13.188714	6.430990	8.837989
C	12.541043	3.591480	7.119048
H	13.085591	4.537926	7.158303
C	9.100789	9.792317	5.980743
C	13.252609	9.304778	6.352941
H	12.455818	9.957676	5.951996
H	14.198799	9.630636	5.875077
C	6.140305	4.890852	7.134063
H	5.116821	4.885662	6.745325
C	11.284413	3.460554	7.745885
C	13.105186	2.501703	6.436183
H	14.080810	2.611673	5.951786
C	10.844231	8.997045	12.901966
H	11.882903	8.858902	13.215619
C	8.438210	8.573228	6.449954
H	8.171202	7.724987	5.816624
O	14.327235	6.167572	8.840234
C	10.505057	4.163593	10.481451
H	9.765248	3.347183	10.564747
H	11.503488	3.714716	10.633087
C	7.231766	4.837929	6.248248
H	7.060102	4.777291	5.168130
C	10.238061	5.278765	11.499220
H	10.438113	4.952712	12.535261
H	9.184742	5.605350	11.479237
C	10.280351	8.170020	11.906386
C	10.593914	2.225651	7.674942
H	9.607831	2.109929	8.138358
C	8.736391	9.916649	8.290170
H	8.748022	10.293029	9.314645

C	8.152033	8.688888	7.847302
H	7.635432	7.945546	8.454381
C	6.365597	4.924652	8.520651
H	5.519344	4.936209	9.215538
C	13.003704	7.858854	5.915546
H	13.807142	7.181146	6.250734
H	12.944497	7.798595	4.815419
C	8.925983	8.350842	11.534757
H	8.473810	7.713669	10.764899
C	8.779674	4.864941	8.140086
C	8.544485	4.828736	6.743663
H	9.385776	4.751161	6.046313
C	12.837879	6.672680	11.984529
C	13.777980	7.725090	11.861067
H	13.536973	8.612899	11.264706
C	13.172557	5.536343	12.754486
H	12.473817	4.702362	12.870358
C	15.340003	6.514824	13.279982
H	16.309263	6.453526	13.784959
C	14.419746	5.461458	13.395430
H	14.666758	4.576807	13.991031
C	15.015795	7.648391	12.514438
H	15.729131	8.474157	12.425013
C	13.406053	9.543259	7.857467
H	13.606022	10.610602	8.053850
H	14.235601	8.955311	8.286678
C	9.428833	10.123931	4.555549
H	9.732780	9.231509	3.983668
H	8.529674	10.536521	4.058010
H	10.221253	10.886506	4.481801

Table S69. [Cp'Ni^{III}(pdt)Fe^{II}(dppe)(CO)]$^{2+}$ with pdt Oriented Toward Fe ([**1d**]$^{2+}$, –3243.20601950 E_h)

atom	x	y	z
Ni	10.248362	8.718785	7.546427
Fe	11.564028	6.918457	8.764927
P	11.238996	6.814054	11.109595
S	11.929672	9.154243	8.881183
P	10.527850	4.819728	8.702769
S	11.368444	7.151697	6.508585
C	11.274220	1.112583	7.095564
H	10.797073	0.128035	7.135261
C	12.456559	1.296971	6.357266
H	12.903916	0.453161	5.822186
C	8.177382	9.360528	12.181257
H	7.115949	9.472157	11.935004
C	7.677205	4.939001	8.954890
H	7.806953	4.961391	10.041344
C	8.772910	10.205665	13.135518
H	8.177121	10.981886	13.626495
C	9.318866	10.597480	7.178055
H	9.831420	11.561668	7.174955
C	10.127803	10.044051	13.466194
H	10.591468	10.690357	14.218377
C	13.199131	6.281769	8.900829
C	12.489300	3.646435	6.989101
H	12.967875	4.627613	6.935839
C	9.093234	9.765894	6.045642
C	13.880390	8.597380	6.788135
H	14.665999	9.094038	6.182756
H	14.393644	7.798339	7.346224
C	6.169665	4.862062	7.043127
H	5.152150	4.846855	6.639279
C	11.308061	3.466477	7.738253
C	13.059726	2.562799	6.301475
H	13.977447	2.710003	5.722933
C	10.894629	9.040849	12.851738
H	11.941321	8.908778	13.140181
C	8.402801	8.573671	6.545666
H	8.109991	7.718749	5.932815
O	14.284008	5.861528	8.999691
C	10.482755	4.177291	10.461481
H	9.733474	3.369432	10.541494
H	11.474822	3.720379	10.629723
C	7.274079	4.784381	6.175649
H	7.119045	4.695478	5.095059
C	10.210235	5.298599	11.471454
H	10.378284	4.968051	12.511604
H	9.162922	5.641040	11.426106
C	10.302547	8.189427	11.893264
C	10.695672	2.189740	7.780440
H	9.763374	2.034621	8.334936
C	8.754611	9.946905	8.353685
H	8.786164	10.342507	9.370345

C	8.135132	8.726374	7.940979
H	7.608435	8.008505	8.569012
C	6.373805	4.934111	8.431714
H	5.516953	4.965193	9.112894
C	12.884290	7.988424	5.799991
H	13.378283	7.209793	5.192741
H	12.476755	8.744757	5.105516
C	8.938890	8.363978	11.554571
H	8.465203	7.712670	10.810565
C	8.792563	4.862335	8.088997
C	8.579201	4.786572	6.691035
H	9.430592	4.689074	6.008854
C	12.824470	6.646175	12.016256
C	13.806753	7.655421	11.865246
H	13.613991	8.519446	11.218677
C	13.098036	5.537074	12.847471
H	12.365962	4.736026	12.986806
C	15.289702	6.458172	13.376582
H	16.244967	6.384764	13.906015
C	14.327279	5.446826	13.520216
H	14.527313	4.583031	14.162238
C	15.026139	7.564597	12.550521
H	15.773099	8.357078	12.437659
C	13.343310	9.650195	7.759105
H	12.973654	10.548064	7.232963
H	14.144181	9.973408	8.447071
C	9.421872	10.058258	4.611963
H	9.692954	9.145071	4.055882
H	8.533806	10.489085	4.110008
H	10.237558	10.794032	4.518607

Table S70. $[CpNi^{II}(SPh)_2Fe^{II}(dppv)(CO)]^+$ with CO in Apical Position ($[\mathbf{2b}]^+$, –3548.32358251 E_h)

atom	x	y	z
Ni	3.243579	3.626153	9.816348
Fe	2.463742	4.118076	7.341523
P	3.380404	5.827987	6.156320
P	3.505304	2.747667	5.852180
S	2.060285	2.237824	8.593957
S	1.763482	5.151470	9.231929
O	-0.081577	4.421410	5.982636
C	0.019524	2.575541	10.550687
H	0.729987	3.171056	11.130632
C	2.603945	1.385196	4.979698
C	0.397406	2.056337	9.297532
C	4.155603	4.400367	11.573440
H	3.853307	5.337452	12.044790
C	5.156441	4.276829	10.520962
H	5.745948	5.096846	10.111390
C	-2.178142	1.550291	10.304090
H	-3.181374	1.354175	10.696481
C	1.077836	9.276575	5.958730
H	0.901333	10.162288	6.578224
C	5.044051	1.900119	6.435800
C	1.197685	1.291853	5.015463
H	0.612888	1.998561	5.608287
C	0.975488	4.290094	6.478122
C	2.244186	7.165337	5.593337
C	4.953641	6.684242	6.649780
C	4.089599	3.778020	4.446902
H	4.499056	3.291403	3.551432
C	1.999661	8.308856	6.386689
H	2.533234	8.452564	7.331266
C	-0.506370	1.265837	8.556336
H	-0.200347	0.836477	7.596290
C	3.499565	7.376564	9.675827
H	4.348764	6.730861	9.442527
C	7.467787	7.809642	7.265089
H	8.442356	8.248287	7.504596
C	-1.268971	2.319580	11.047073
H	-1.559433	2.724897	12.022537
C	3.979332	5.114866	4.560043
H	4.306813	5.802755	3.768149
C	5.275418	7.987406	6.215105
H	4.552712	8.569779	5.635188
C	1.089518	7.699583	9.902759
H	0.072721	7.294157	9.861707
C	3.706132	8.719528	10.030172
H	4.727336	9.111994	10.083623
C	2.668931	-0.527998	3.461254
H	3.247211	-1.235482	2.857325
C	0.386185	9.112536	4.747365
H	-0.332738	9.870385	4.418349
C	-1.792544	1.022993	9.060204

H	-2.492081	0.411990	8.479639
C	5.236261	2.911758	10.148860
H	5.873846	2.473911	9.380205
C	7.466358	1.862911	6.750966
H	8.446668	2.325837	6.593433
C	4.269279	2.191865	10.955662
H	4.062828	1.122438	10.884860
C	6.525742	8.545893	6.528995
H	6.763524	9.560161	6.190421
C	1.540652	7.003455	4.378787
H	1.707871	6.116552	3.757049
C	2.189781	6.863720	9.607971
C	6.312051	2.496099	6.260034
H	6.402677	3.449232	5.727366
C	0.534401	0.294708	4.280969
H	-0.558729	0.235030	4.315604
C	1.267186	-0.616142	3.504784
H	0.748547	-1.393268	2.933364
C	3.652644	3.104306	11.880036
H	2.900084	2.855823	12.628453
C	3.335797	0.465073	4.193102
H	4.429697	0.515223	4.159430
C	7.366177	0.633840	7.424125
H	8.267672	0.137838	7.799416
C	6.106216	0.034785	7.601040
H	6.023611	-0.930534	8.112380
C	4.949793	0.660794	7.110405
H	3.975062	0.178693	7.238163
C	0.618636	7.974369	3.959059
H	0.082173	7.838373	3.014036
C	2.611749	9.555672	10.312580
H	2.777984	10.601403	10.592053
C	5.908606	5.944960	7.381394
H	5.665014	4.928745	7.713002
C	1.304835	9.043060	10.246013
H	0.447751	9.686483	10.471955
C	7.159372	6.504458	7.686322
H	7.896047	5.920901	8.250118

Table S71. $[CpNi^{II}(SPh)_2Fe^{II}(dppv)(CO)]^+$ with CO in Basal Position ($[\mathbf{2b}]^+$, –3548.32555524 E_h)

atom	x	y	z
Ni	2.878177	3.503015	9.719936
Fe	2.399186	4.248917	7.398816
P	3.083344	2.862557	5.731861
P	1.429031	5.466835	5.810708
S	1.463176	2.375218	8.455764
S	1.622112	5.274361	9.257231
O	5.006940	5.514048	7.158519
C	-0.362620	2.851460	10.599011
H	0.458621	3.326293	11.143344
C	1.768177	7.279329	5.795996
C	-0.144912	2.359331	9.296979
C	3.656348	3.802864	11.676821
H	3.201863	4.465956	12.414501
C	4.618522	4.212024	10.659755
H	4.998348	5.225865	10.523694
C	-2.676917	2.090613	10.502466
H	-3.659374	1.982772	10.973796
C	6.615338	0.979949	6.748728
H	6.927939	0.210253	7.462860
C	-0.399580	5.370307	5.568153
C	2.975801	7.764248	5.246362
H	3.695151	7.070386	4.796463
C	3.924918	5.055806	7.242653
C	4.847410	2.336344	5.745793
C	2.126476	1.336196	5.282550
C	2.116890	4.920082	4.188316
H	1.878210	5.482281	3.275581
C	5.266490	1.359675	6.678708
H	4.531269	0.886515	7.340263
C	-1.194904	1.715411	8.605006
H	-1.016014	1.301455	7.606708
C	3.220386	7.637259	9.138036
H	3.525531	7.364939	8.126537
C	0.572664	-0.867643	4.452195
H	-0.029684	-1.723654	4.129921
C	-1.627735	2.716191	11.193986
H	-1.789005	3.098728	12.207938
C	2.879208	3.809053	4.162942
H	3.276319	3.391906	3.227490
C	2.745238	0.111079	4.959850
H	3.832710	0.007260	5.016289
C	1.928912	7.196300	11.164702
H	1.227338	6.563692	11.720198
C	3.678554	8.836601	9.706722
H	4.350749	9.480072	9.129295
C	1.142414	9.559144	6.393385
H	0.425524	10.256553	6.839681
C	7.562382	1.583267	5.902448
H	8.615339	1.288007	5.959645
C	-2.455002	1.587929	9.208850

H	-3.261598	1.078884	8.669606
C	4.979928	3.064654	9.893163
H	5.674684	3.036176	9.053834
C	-2.395843	5.890468	4.267230
H	-2.861034	6.360415	3.394033
C	4.178797	1.972512	10.368212
H	4.165650	0.960091	9.961028
C	1.966669	-0.986066	4.553464
H	2.458466	-1.933776	4.309206
C	5.804647	2.940907	4.905002
H	5.503116	3.712719	4.188705
C	2.353067	6.806493	9.872919
C	-1.007900	5.976531	4.444516
H	-0.404044	6.529076	3.716021
C	3.256634	9.139449	5.265173
H	4.189476	9.508385	4.825175
C	2.341087	10.037833	5.838645
H	2.559503	11.111068	5.848479
C	3.395206	2.421343	11.505482
H	2.706050	1.807285	12.086322
C	0.853715	8.186100	6.374079
H	-0.088413	7.821981	6.797761
C	-3.187937	5.210033	5.209588
H	-4.272545	5.149875	5.068949
C	-2.589649	4.615635	6.331080
H	-3.199427	4.091000	7.074162
C	-1.197967	4.691997	6.510653
H	-0.733766	4.229527	7.387555
C	7.155560	2.564475	4.985399
H	7.889669	3.039971	4.326314
C	3.277533	9.213735	10.998841
H	3.641811	10.149383	11.435960
C	0.721586	1.451666	5.174273
H	0.227290	2.401856	5.406614
C	2.397686	8.392651	11.725194
H	2.068771	8.686456	12.727779
C	-0.047865	0.356525	4.756888
H	-1.134492	0.461375	4.664481

Table S72. $[CpNi^{I}(SPh)_2Fe^{II}(dppv)(CO)]^0$ with CO in Apical Position ($[\mathbf{2b}]^0$, –3548.49276969 E_h)

atom	x	y	z
Ni	3.335341	3.634989	9.745748
Fe	2.412508	4.085211	7.373762
P	3.344839	5.789927	6.246404
P	3.485309	2.747433	5.920755
S	2.004300	2.141955	8.585921
S	1.686268	5.119376	9.275875
O	-0.140676	4.371959	5.997127
C	0.057853	2.482456	10.600137
H	0.849258	2.960380	11.185607
C	2.619870	1.380954	4.996070
C	0.341706	2.042280	9.290443
C	4.276460	4.284776	11.655884
H	3.891073	5.124259	12.237692
C	5.245402	4.365048	10.608910
H	5.777867	5.264503	10.297390
C	-2.237990	1.691671	10.384157
H	-3.239527	1.557530	10.808275
C	1.174352	9.332962	5.964145
H	1.046675	10.245563	6.556769
C	5.041221	1.884822	6.453064
C	1.215596	1.270735	5.013332
H	0.624689	1.958487	5.622127
C	0.921357	4.247943	6.497125
C	2.247868	7.161629	5.660005
C	4.935999	6.650381	6.709069
C	4.074028	3.776166	4.507721
H	4.488421	3.297241	3.609882
C	2.067425	8.345239	6.408609
H	2.625471	8.501978	7.336882
C	-0.673364	1.411356	8.539255
H	-0.449764	1.045929	7.531387
C	3.353523	7.371867	9.729867
H	4.206602	6.697333	9.626030
C	7.479708	7.736991	7.279963
H	8.464696	8.161595	7.504862
C	-1.226701	2.306952	11.139325
H	-1.435843	2.656000	12.157435
C	3.956395	5.111173	4.631918
H	4.279019	5.810597	3.847530
C	5.307512	7.909137	6.190066
H	4.608775	8.473067	5.563480
C	0.944925	7.729496	9.704692
H	-0.068086	7.331116	9.580649
C	3.547364	8.729905	10.031727
H	4.565942	9.113592	10.161849
C	2.718735	-0.499214	3.435245
H	3.311624	-1.188504	2.823170
C	0.446283	9.151553	4.777326
H	-0.251536	9.924307	4.435518
C	-1.955371	1.242612	9.082996

H	-2.735497	0.754221	8.487320
C	5.429348	3.042016	10.068005
H	6.113217	2.759168	9.265992
C	7.473176	1.825681	6.690782
H	8.451362	2.284412	6.506016
C	4.591337	2.149567	10.805261
H	4.475301	1.081517	10.615929
C	6.570440	8.450037	6.481618
H	6.843867	9.432437	6.079591
C	1.505532	6.983333	4.471459
H	1.619980	6.063002	3.887352
C	2.050460	6.861212	9.558007
C	6.308569	2.466596	6.235620
H	6.387509	3.423922	5.708238
C	0.569374	0.284701	4.249249
H	-0.523724	0.211990	4.274741
C	1.317902	-0.602419	3.460417
H	0.812666	-1.372986	2.867198
C	3.854404	2.917552	11.766871
H	3.118780	2.526872	12.472400
C	3.366146	0.484661	4.197700
H	4.460099	0.546221	4.181249
C	7.385167	0.600583	7.371929
H	8.293808	0.100341	7.725177
C	6.125559	0.018167	7.596583
H	6.048875	-0.937795	8.126957
C	4.960063	0.652417	7.140612
H	3.982426	0.192015	7.318495
C	0.612903	7.973001	4.032582
H	0.044950	7.818708	3.108239
C	2.445484	9.592130	10.168129
H	2.599740	10.650358	10.407677
C	5.856615	5.936510	7.504171
H	5.563303	4.962435	7.913547
C	1.143968	9.085594	10.004860
H	0.277591	9.748298	10.114329
C	7.121757	6.476495	7.786822
H	7.826833	5.911965	8.407888

Table S73. $[CpNi^{I}(SPh)_2Fe^{II}(dppv)(CO)]^0$ with CO in Basal Position ($[\mathbf{2b}]^0$, –3548.4954561 E_h)

atom	x	y	z
Ni	3.033132	3.461907	9.691595
Fe	2.434672	4.162630	7.426474
P	3.062561	2.855927	5.721793
P	1.470828	5.453708	5.920275
S	1.555553	2.241328	8.465781
S	1.527425	5.223696	9.249375
O	5.084684	5.352461	7.261298
C	-0.230697	2.699877	10.627916
H	0.620066	3.130319	11.164960
C	1.846232	7.265147	5.928016
C	-0.047916	2.246166	9.304463
C	3.847048	3.897696	11.667472
H	3.352585	4.586066	12.355248
C	4.806340	4.256215	10.668786
H	5.185344	5.260086	10.474491
C	-2.575905	2.032195	10.555728
H	-3.553996	1.947512	11.042644
C	6.601643	0.958564	6.671329
H	6.935065	0.260860	7.447789
C	-0.369152	5.429809	5.668635
C	3.064307	7.727153	5.383408
H	3.763075	7.017062	4.926282
C	3.988579	4.911171	7.348763
C	4.818750	2.282815	5.650518
C	2.096721	1.340104	5.226302
C	2.094200	4.958329	4.247355
H	1.843050	5.553819	3.358582
C	5.267463	1.391536	6.651987
H	4.567639	1.044359	7.421293
C	-1.142664	1.671602	8.618326
H	-0.997342	1.291321	7.600937
C	3.308476	7.464164	9.371107
H	3.982401	6.957208	8.679170
C	0.520813	-0.847866	4.383905
H	-0.090214	-1.697348	4.058525
C	-1.488184	2.593196	11.244169
H	-1.614954	2.950597	12.273031
C	2.833522	3.833926	4.167166
H	3.194177	3.438235	3.207514
C	2.698406	0.103756	4.919877
H	3.783023	-0.015556	5.002190
C	1.234451	7.541444	10.643048
H	0.287776	7.074500	10.938714
C	3.651060	8.729169	9.874773
H	4.597190	9.188650	9.565428
C	1.298217	9.552263	6.578200
H	0.610347	10.259665	7.054376
C	7.509857	1.426636	5.705625
H	8.553992	1.095209	5.727074
C	-2.396187	1.568343	9.240911

H	-3.232862	1.110947	8.699080
C	5.171263	3.058148	9.954037
H	5.877067	3.002788	9.124164
C	-2.374281	6.067221	4.430420
H	-2.843385	6.614477	3.604663
C	4.447239	1.965655	10.521588
H	4.472060	0.927770	10.184820
C	1.911111	-0.985230	4.506841
H	2.392939	-1.942582	4.277738
C	5.737694	2.751703	4.690204
H	5.417063	3.462489	3.921126
C	2.094490	6.854391	9.751983
C	-0.984770	6.133791	4.609006
H	-0.380852	6.748206	3.931158
C	3.387910	9.091895	5.426209
H	4.333166	9.439020	4.993508
C	2.505377	10.007012	6.023269
H	2.759643	11.072341	6.058974
C	3.601875	2.486869	11.561307
H	2.911364	1.905354	12.175114
C	0.967622	8.189375	6.531746
H	0.023464	7.843036	6.964920
C	-3.163315	5.305964	5.311540
H	-4.249427	5.259742	5.171657
C	-2.559105	4.612793	6.371594
H	-3.163775	4.023862	7.069987
C	-1.166052	4.671741	6.549338
H	-0.693354	4.135729	7.379430
C	7.076810	2.327332	4.720812
H	7.781877	2.704796	3.971575
C	2.798180	9.404005	10.763406
H	3.073803	10.389828	11.154580
C	0.695216	1.470980	5.102607
H	0.212778	2.425674	5.340323
C	1.586259	8.802439	11.144407
H	0.907817	9.317180	11.835083
C	-0.084962	0.386094	4.678428
H	-1.170023	0.505271	4.579969

References:

(1) Pool, D. H.; DuBois, D. L. *J. Organomet. Chem.* **2009**, *694*, 2858.

(2) McCarthy, B. D.; Martin, D. J.; Rountree, E. S.; Ullman, A. C.; Dempsey, J. L. *Inorg. Chem.* **2014**, *53*, 8350.