Supporting Information

Computational Investigation of [FeFe]-Hydrogenase Models: Characterization of Singly and Doubly Protonated Intermediates and Mechanistic Insights

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EXPERIMENTAL METHODS

Compounds of $Fe_2(pdt)(dppv)_2(CO)_2$ ($[(Fe^IFe^I)pdt]^0$) and $Fe_2(adt)(dppv)_2(CO)_2$ ($[(Fe^IFe^I)adt]^0$) as referenced in the main text were prepared according to reported procedures.[1] FT-IR measurements (in cm^{-1}) are reported for the ν_{CO} region only. Additional experimental conditions are provided in the captions of the following spectra.

Low-temperature Protonation. Protonation of $[(Fe^IFe^I)adt]^0$ followed a previously described procedure.[2] In a typical experiment, in a J. Young NMR tube ca. 0.6 mL CD_2Cl_2 was distilled onto $[(Fe^IFe^I)adt]^0$ (6 mg , 0.006 mmol) and $[H(OEt_2)_2]BAr^F{}_4$ (6 mg, 0.006 mmol). The J. Young tube was then placed directly into a CH_3CN/dry ice bath and analyzed with low temperature NMR spectroscopy at –40 °C.

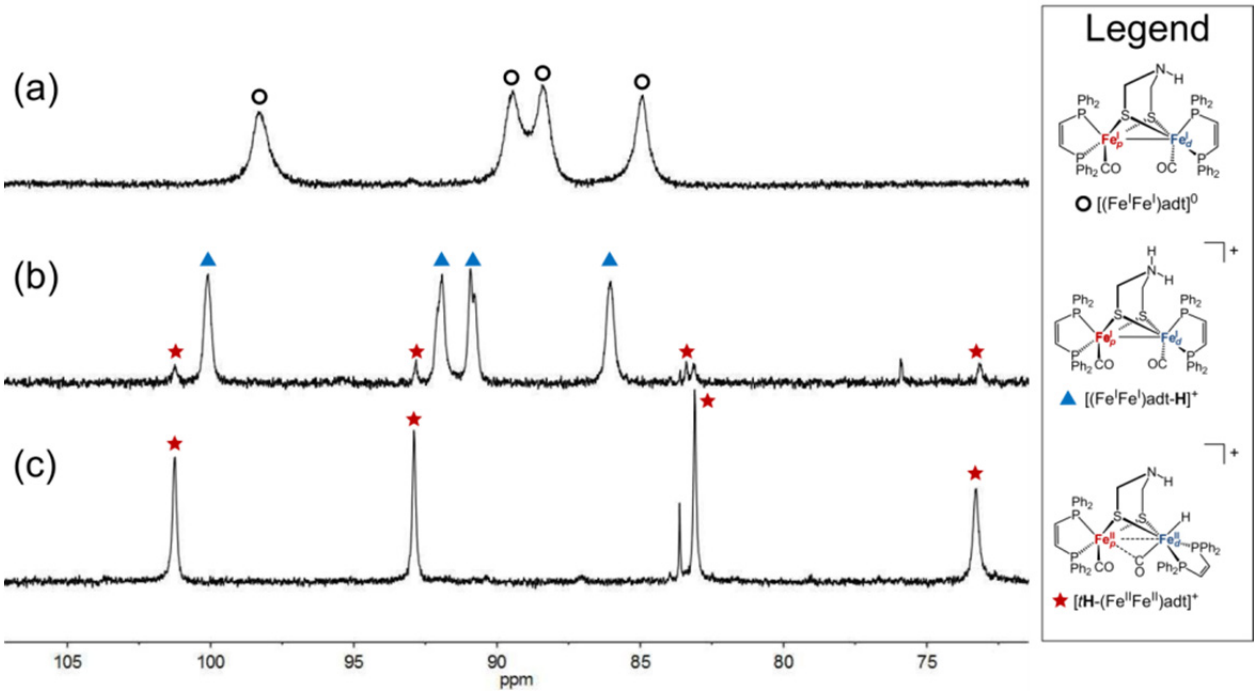


Figure S1. ^{31}P NMR spectra at –40 °C of [(FeFe)adt] species in CD_2Cl_2 solvent. The Fe centers have been labeled Fe_p and Fe_d for consistency, although this labeling is only chemically meaningful for the terminal hydride species. (a) Spectrum for $[(Fe^IFe^I)adt]^0$ before addition of acid. Four nonequivalent phosphine peaks are observed, consistent with both diphosphine ligands occupying the apical-basal sites with relatively rigid azadithiolate (adt) bridge enforcing nonequivalency. (b) $[(Fe^IFe^I)adt]^0$ after treatment with $[Me_3NH]^+$ acid. The blue triangles correspond to the ammonium tautomer, $[(Fe^IFe^I)adt\text{-}\mathbf{H}]^+$, which exhibits heteroconjugation via hydrogen bonding between $[Me_3NH]^+$ and the adt (see **Figure S14b**). A small fraction of the terminal hydride species, $[t\mathbf{H}\text{-}(Fe^{II}Fe^{II})adt]^+$, is evident (red stars). (c) Spectrum of the terminal hydride species (red stars), $[t\mathbf{H}\text{-}(Fe^{II}Fe^{II})adt]^+$, produced by treatment of $[(Fe^IFe^I)adt]^0$ with the strong acid $[H(OEt_2)_2]BAr^F{}_4$. No traces of the ammonium tautomer are seen.

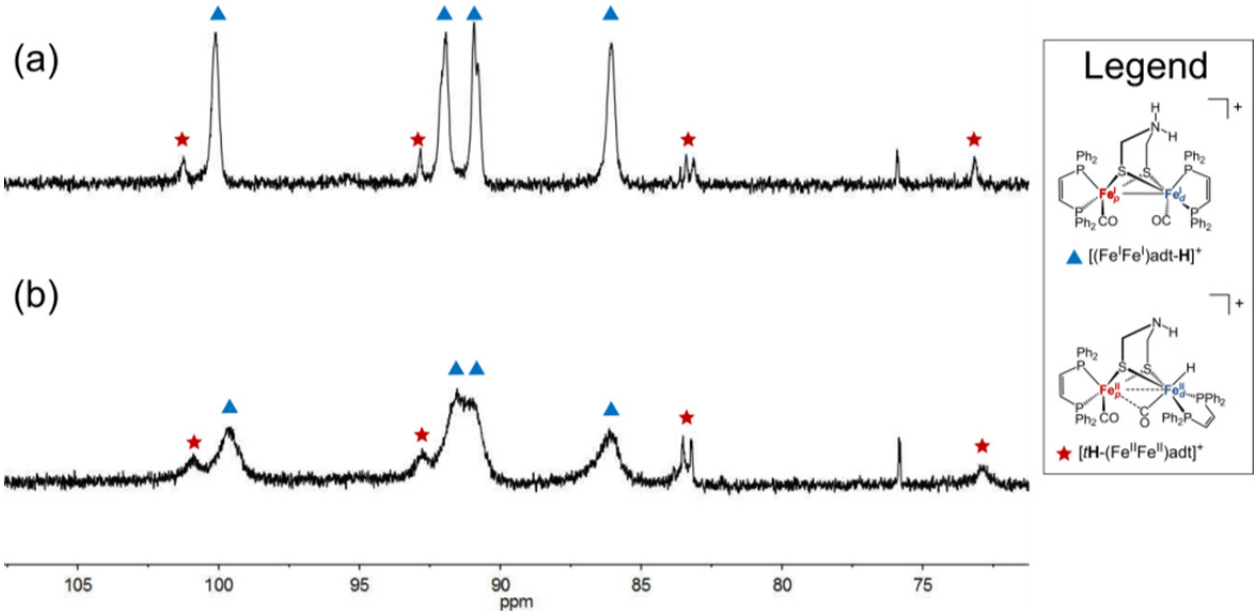


Figure S2. ^{31}P NMR spectra of $[(Fe^{I}Fe^{I})adt]^{0}$ species (in CD_2Cl_2 solvent) with $[Me_3NH]BAr^{F}_4$ at (a) –40 °C and (b) –20 °C. All species, $[t\mathbf{H}\text{-}(Fe^{II}Fe^{II})adt]^{+}$ (red stars) and $[(Fe^{I}Fe^{I})adt\text{-}\mathbf{H}]^{+}$ (blue triangles), experience dynamical broadening. The Fe centers have been labeled Fe_p and Fe_d for consistency, although this labeling is only chemically meaningful for the terminal hydride species.

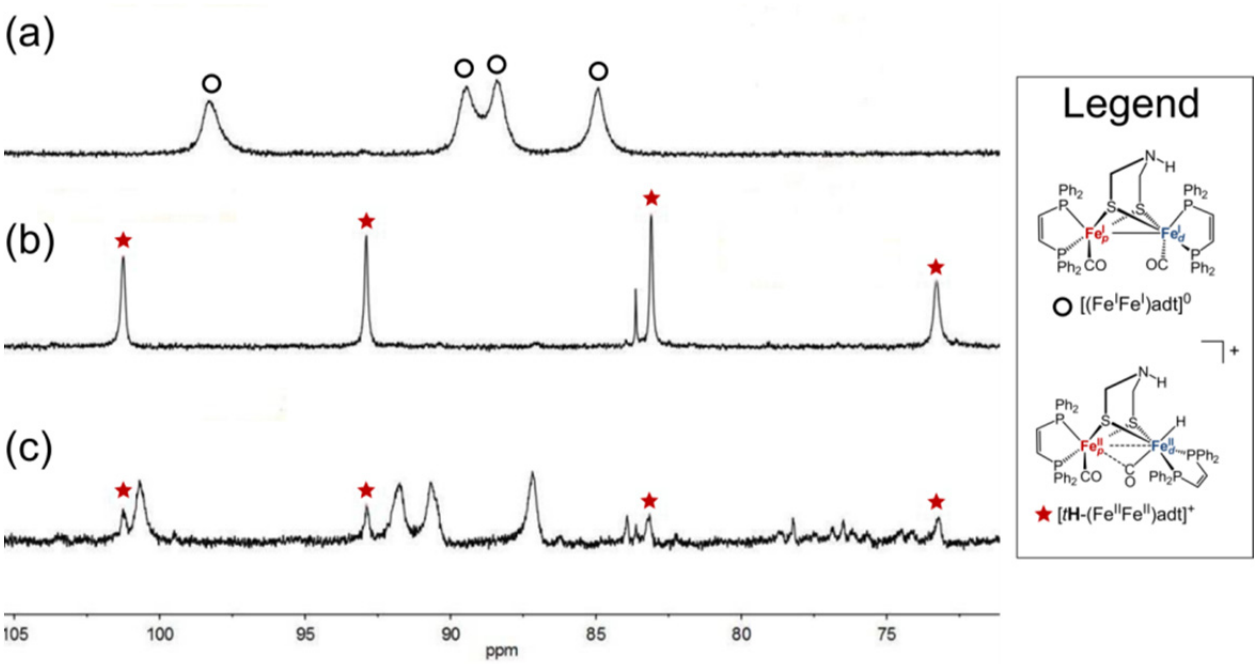


Figure S3. ^{31}P NMR spectra (at –40 °C in CD_2Cl_2 solvent) of $[(Fe^IFe^I)adt]^0$ species (a) before the addition of acid, (b) after treatment with 1 equiv of the strong acid $[H(OEt_2)_2]BAr^F{}_4$, and (c) after treatment with 0.5 equiv of $[H(OEt_2)_2]BAr^F{}_4$. The spectrum in (b) shows that in CD_2Cl_2, one equiv of strong acid exclusively results in protonation at a single Fe site to form the terminal hydride, $[t\mathbf{H}\text{-}(Fe^{II}Fe^{II})adt]^+$ (red stars). The spectrum in (c) shows that treating $[(Fe^IFe^I)adt]^0$ with 0.5 equiv of strong acid results in only a fraction of the terminal hydride species forming. The main species is assigned to the homoconjugated species, $[(Fe^IFe^I)adt\text{-}\mathbf{H}\text{-}adt(Fe^IFe^I)]^+$ (see **Figure S14c**). The Fe centers have been labeled Fe_p and Fe_d for consistency, although this labeling is only chemically meaningful for the terminal hydride species.

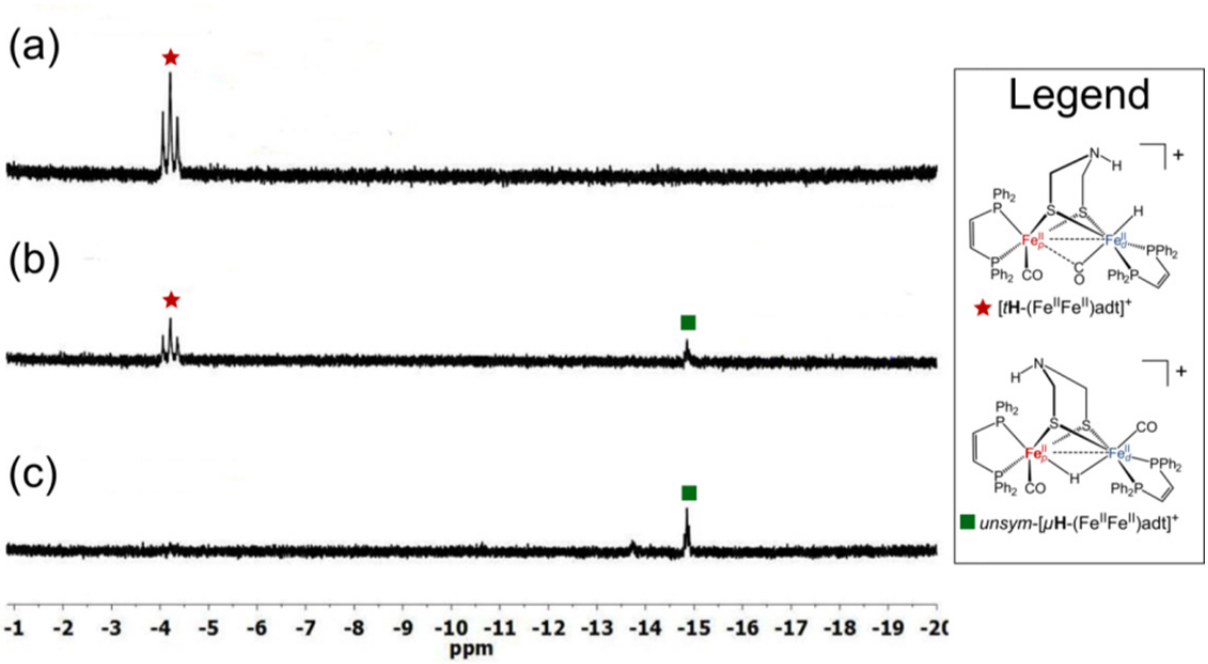


Figure S4. ^{1}H NMR spectra (at –40 °C in CD_2Cl_2 solvent) of $[(Fe^{I}Fe^{I})adt]^{0}$ species (a) after treatment with 1 equiv of $[PhNH_3]BAr^{F}_4$ for 30 min. showing exclusive formation of the terminal hydride, $[t\mathbf{H}\text{-}(Fe^{II}Fe^{II})adt]^{+}$ (red stars); (b) after treatment with 2 equiv of $[PhNH_3]BAr^{F}_4$ for 30 min. showing formation of the terminal hydride, $[t\mathbf{H}\text{-}(Fe^{II}Fe^{II})adt]^{+}$ (red stars), and bridging hydride, *unsym*-$[\mu\mathbf{H}\text{-}(Fe^{II}Fe^{II})adt]^{+}$ (green squares); (c) after treatment with 2 equiv of $[PhNH_3]BAr^{F}_4$ for 90 min. showing exclusive formation of the bridging hydride, *unsym*-$[\mu\mathbf{H}\text{-}(Fe^{II}Fe^{II})adt]^{+}$ (green squares). The Fe centers have been labeled Fe_p and Fe_d for consistency, although this labeling is only chemically meaningful for the terminal hydride species.

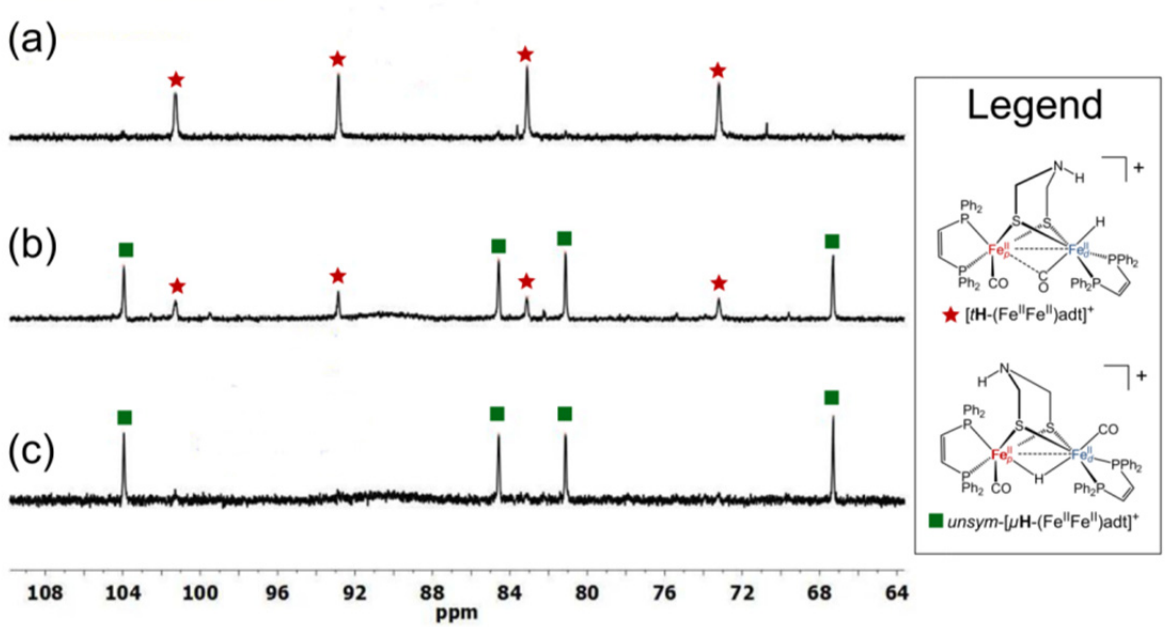


Figure S5. ^{31}P NMR spectra (at –40°C in CD_2Cl_2 solvent) of $[(Fe^IFe^I)adt]^0$ species (a) after treatment with 1 equiv of $[PhNH_3]BAr^F_4$ for 30 min. showing exclusive formation of the terminal hydride $[t\mathbf{H}\text{-}(Fe^{II}Fe^{II})adt]^+$ (red stars); (b) after treatment with 2 equiv of $[PhNH_3]BAr^F_4$ for 50 minutes showing formation of the terminal hydride, $[t\mathbf{H}\text{-}(Fe^{II}Fe^{II})adt]^+$ (red stars), and bridging hydride, *unsym*-$[\mu\mathbf{H}\text{-}(Fe^{II}Fe^{II})adt]^+$ (green squares); (c) after treatment with 2 equiv of $[PhNH_3]BAr^F_4$ for 90 min. showing exclusive formation of the bridging hydride, *unsym*-$[\mu\mathbf{H}\text{-}(Fe^{II}Fe^{II})adt]^+$ (green squares). The Fe centers have been labeled Fe_p and Fe_d for consistency, although this labeling is only chemically meaningful for the terminal hydride species.

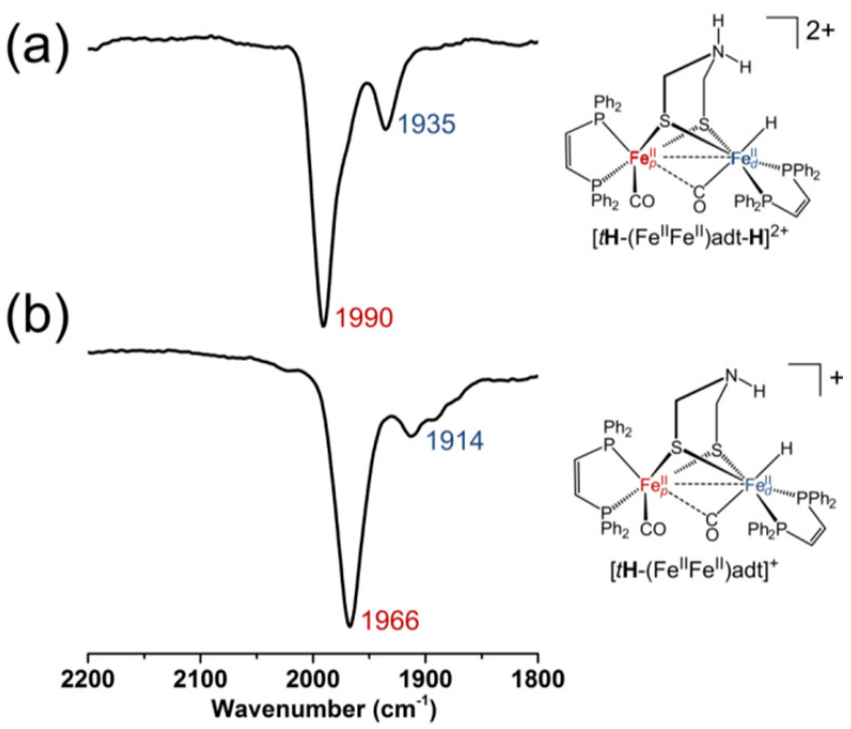


Figure S6. FT-IR spectrum (at –40°C in CH_2Cl_2 solvent) of (a) the doubly protonated terminal hydride species, [*t***H**-($Fe^{II}Fe^{II}$)adt-**H**](BAr^F_4)$_2$, showing bands at 1990 (terminal Fe_pCO) and 1935 cm^{-1} (semi-bridging Fe_dCO); and (b) the singly protonated terminal hydride species, [*t***H**-($Fe^{II}Fe^{II}$)adt]BAr^F_4, showing bands at 1966 (terminal Fe_pCO) and 1914 cm^{-1} (semi-bridging Fe_dCO). We had previously shown that the ν_{CO} band assigned to the semibridging Fe_dCO group in [*t***H**-($Fe^{II}Fe^{II}$)adt-**H**]$^{2+}$ was highly sensitive to the nature of the acid used and anions in CH_2Cl_2 solution: ν_{CO} = 1986 and 1950 cm^{-1} if using CF_3CO_2H; ν_{CO} = 1986 and 1925 cm^{-1} if using $HBF_4{\cdot}Et_2O$.[1]

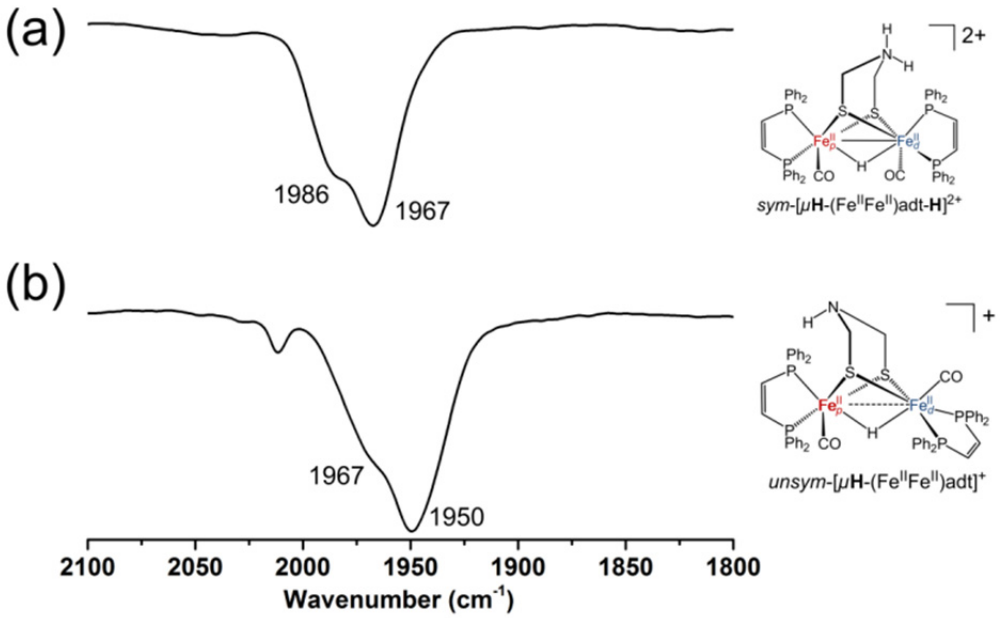


Figure S7. FT-IR spectrum (at room temperature in CH_2Cl_2 solvent) of (a) the doubly protonated bridging hydride species, *sym*-[μ**H**-($Fe^{II}Fe^{II}$)adt-**H**](BAr^F_4)$_2$, showing bands at 1986 and 1967 cm^{-1}; and (b) the singly protonated bridging hydride species, *unsym*-[μ**H**-($Fe^{II}Fe^{II}$)adt]BAr^F_4, showing bands at 1967 and 1950 cm^{-1}. The ν_{CO} bands are assigned to terminal FeCO species in these isomers. The Fe centers have been labeled Fe_p and Fe_d for consistency, although this labeling is only chemically meaningful for the terminal hydride species.

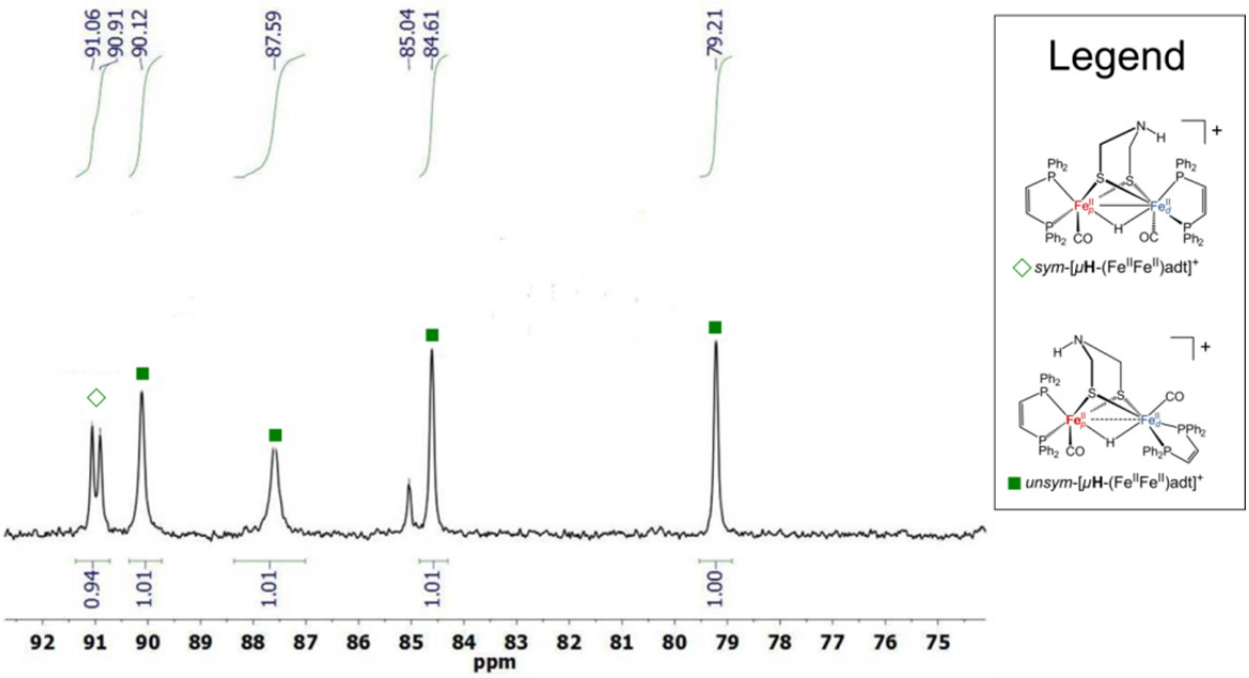


Figure S8. ^{31}P NMR spectrum (in CD_2Cl_2 solvent) of the major isomer, *unsym*-[μ**H**-($Fe^{II}Fe^{II}$)adt]BAr^F_4 (green squares), and minor isomer, *sym*-[μ**H**-($Fe^{II}Fe^{II}$)adt]BAr^F_4 (green diamonds), of the singly protonated bridging hydride species. The spectrum was collected after the sample, obtained from the protonation experiment, was equilibrated for ca. 24 hours at room temperature.[1] The Fe centers have been labeled Fe_p and Fe_d for consistency, although this labeling is only chemically meaningful for the terminal hydride species.

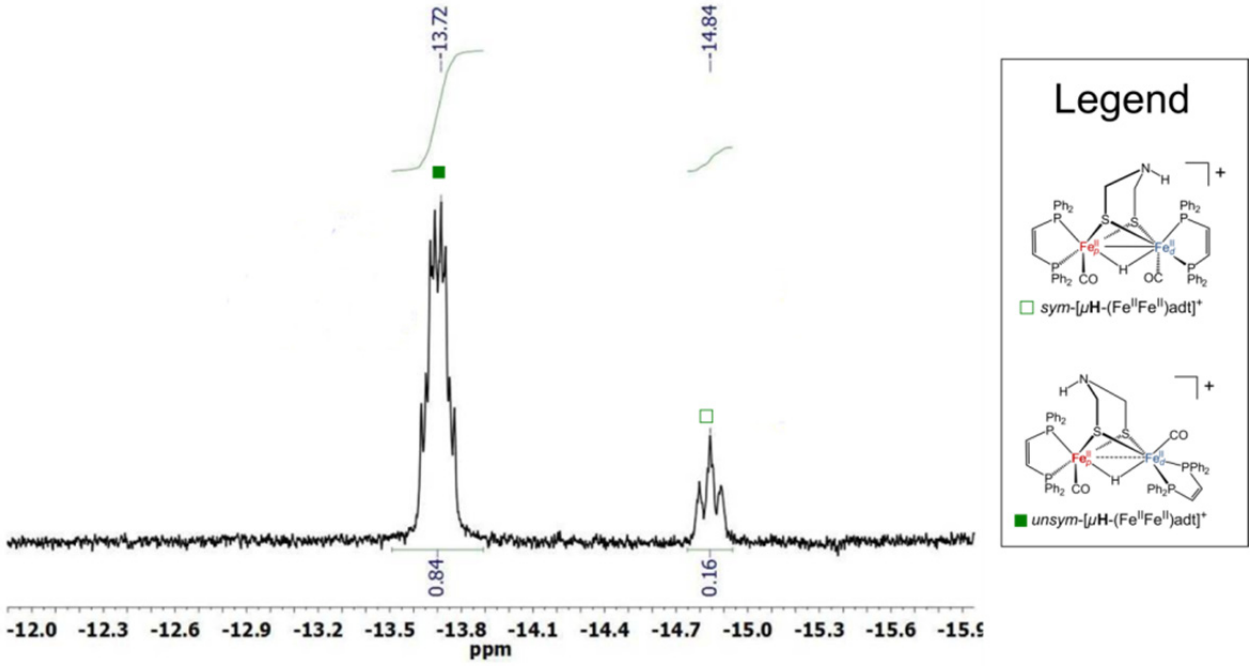


Figure S9. ^{1}H NMR spectrum (in CD_2Cl_2 solvent) of the hydride region of the singly protonated bridging hydride species *unsym*-[μ**H**-($Fe^{II}Fe^{II}$)adt]BAr^{F}_{4} (filled green square) and *sym*-[μ**H**-($Fe^{II}Fe^{II}$)adt]BAr^{F}_{4} (open green square). This is the same sample as shown in **Figure S8**. The Fe centers have been labeled Fe_p and Fe_d for consistency, although this labeling is only chemically meaningful for the terminal hydride species.

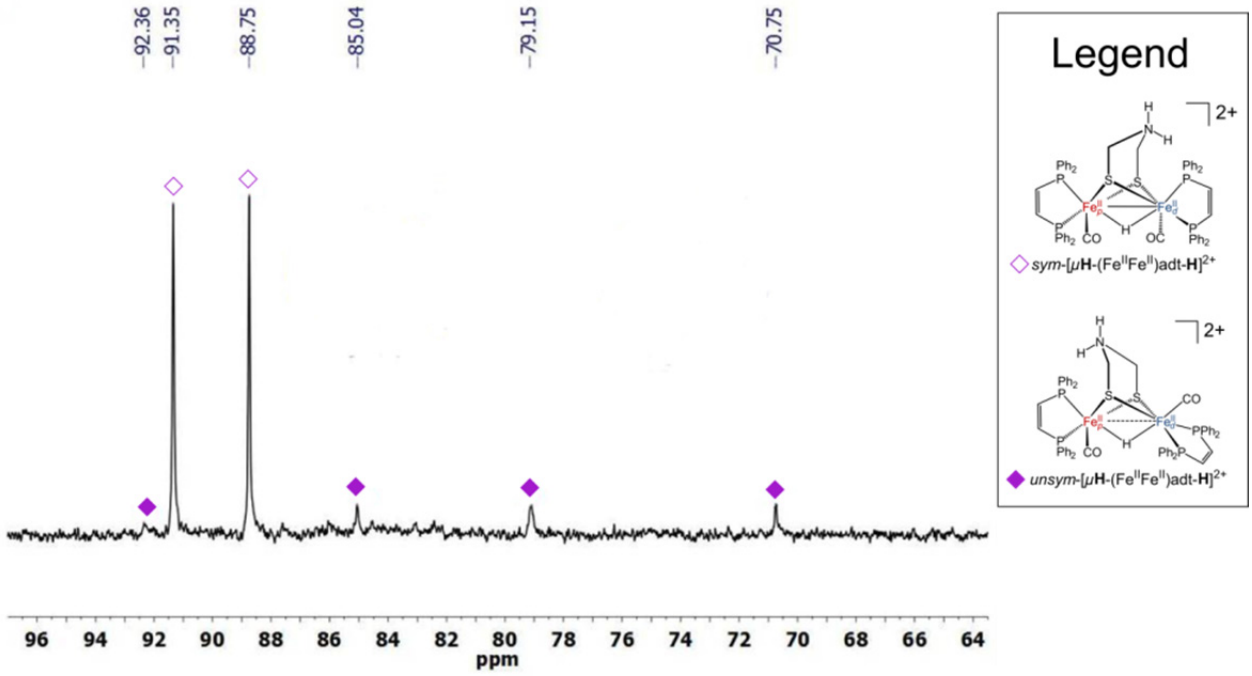


Figure S10. ^{31}P NMR spectrum (in CD_2Cl_2 solvent) of the doubly protonated bridging hydride species *unsym*-[μ**H**-($Fe^{II}Fe^{II}$)adt-**H**](BAr^F_4)$_2$ (filled purple diamonds) and *sym*-[μ**H**-($Fe^{II}Fe^{II}$)adt-**H**](BAr^F_4)$_2$ (open purple diamonds). The spectrum was collected after the sample, obtained from the protonation experiment, was equilibrated for ca. 24 hours at room temperature.[1] The Fe centers have been labeled Fe_p and Fe_d for consistency, although this labeling is only chemically meaningful for the terminal hydride species.

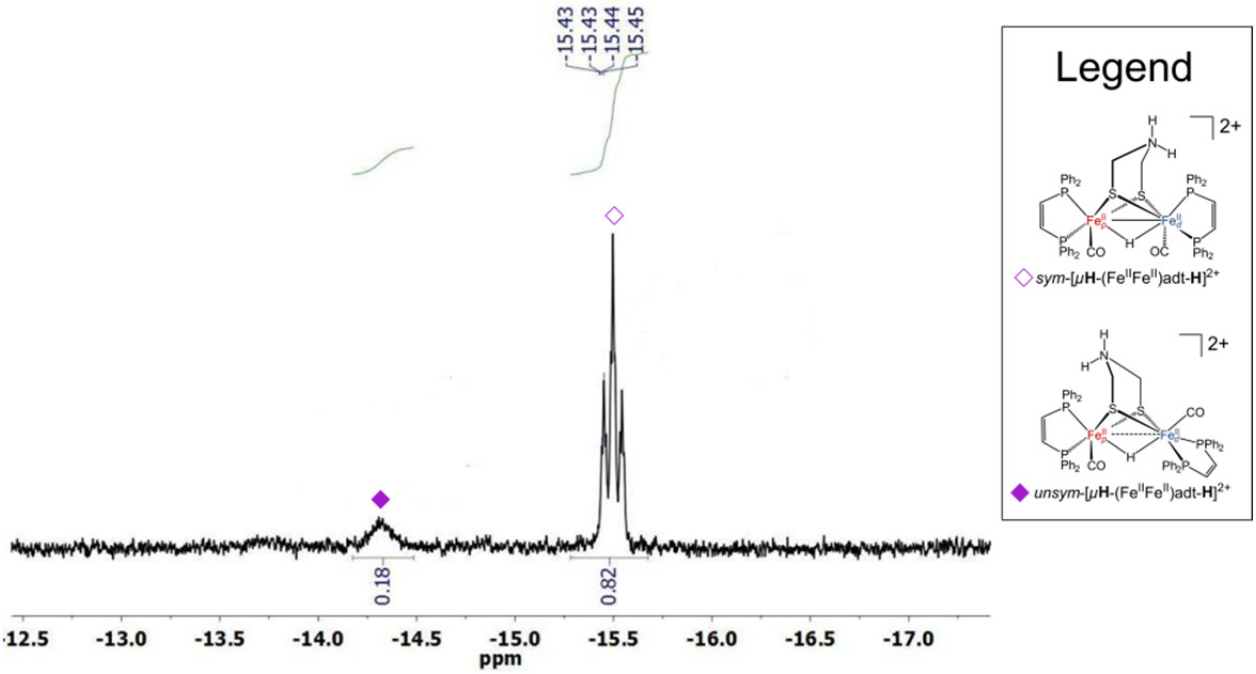


Figure S11. ^{1}H NMR spectrum (in CD_2Cl_2 solvent) of the hydride region of the doubly protonated bridging hydride species *unsym*-[μ**H**-($Fe^{II}Fe^{II}$)adt-**H**](BAr^{F}_{4})$_2$ (filled purple diamonds) and *sym*-[μ**H**-($Fe^{II}Fe^{II}$)adt-**H**](BAr^{F}_{4})$_2$ (open purple diamonds). This the same sample as shown in **Figure S10**. The Fe centers have been labeled Fe_p and Fe_d for consistency, although this labeling is only chemically meaningful for the terminal hydride species.

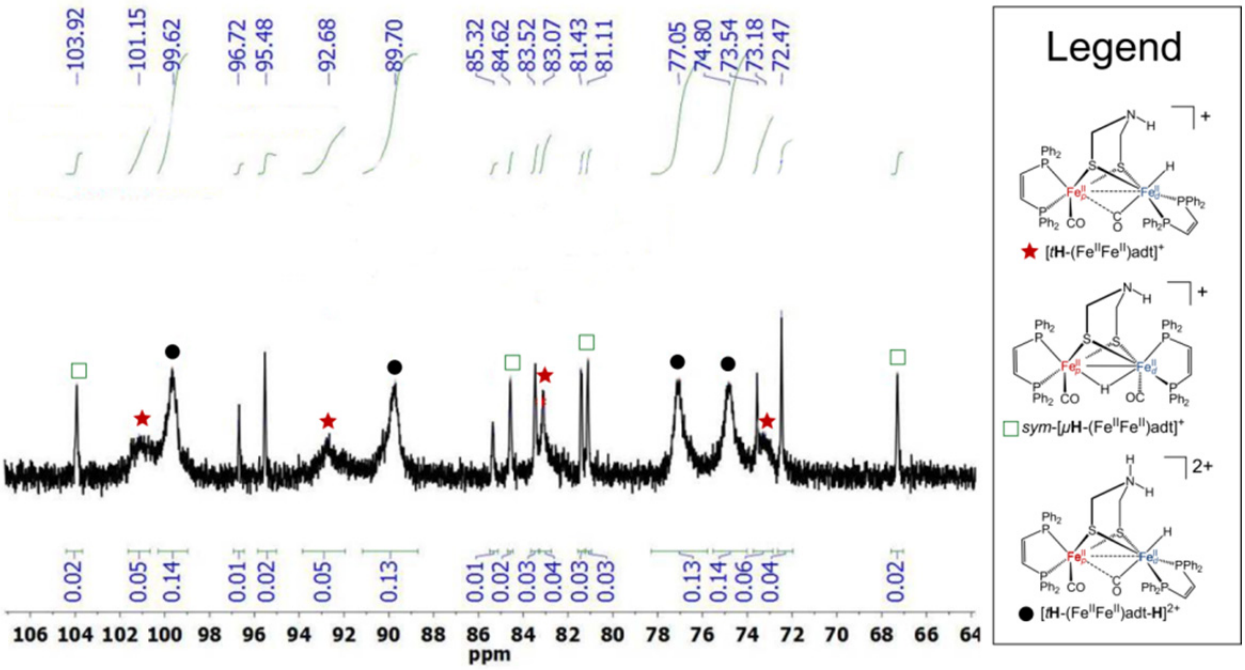


Figure S12. ^{31}P NMR spectrum (at –40 °C in CD_2Cl_2 solvent) of $[(Fe^IFe^I)adt]^0$ after treatment with 2 equiv of $[Ph_2NH_2]BAr^F{}_4$, showing formation of the single protonated terminal hydride species, $[t\mathbf{H}\text{-}(Fe^{II}Fe^{II})adt]^+$ (red stars), the singly protonated bridging hydride species *sym*-$[\mu\mathbf{H}\text{-}(Fe^{II}Fe^{II})adt]^+$ (open green squares), and the doubly protonated terminal hydride species, $[t\mathbf{H}\text{-}(Fe^{II}Fe^{II})adt\text{-}\mathbf{H}]^{2+}$ (black circles). About 17% of an unknown species is observed at δ 96.72, 95.48, 85.32, 83.52, 81.43, and 72.47. The Fe centers have been labeled Fe_p and Fe_d for consistency, although this labeling is only chemically meaningful for the terminal hydride species.

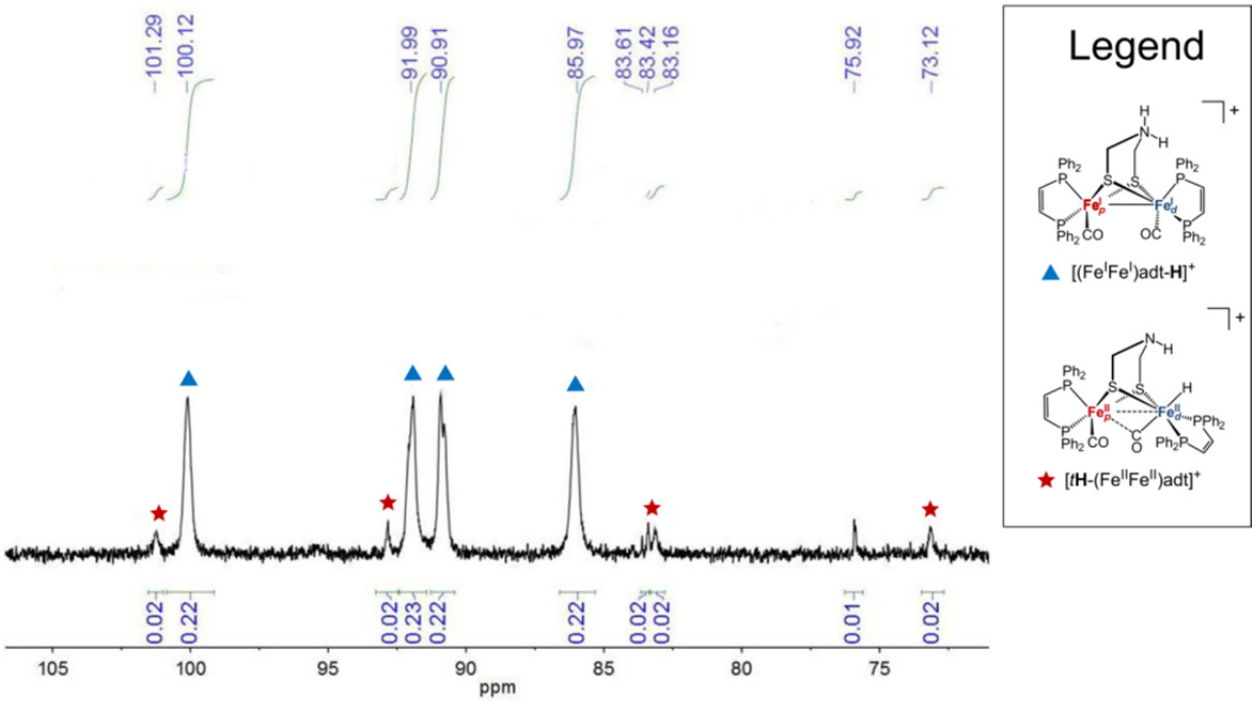


Figure S13. ^{31}P NMR spectrum (at –40 °C in CD_2Cl_2 solvent) of $[(Fe^{I}Fe^{I})adt]^0$ after treatment with 1 equiv of $[Me_3NH]BAr^F{}_4$, showing formation of the singly protonated terminal hydride species $[t\mathbf{H}\text{-}(Fe^{II}Fe^{II})adt]^+$ (red stars) and its ammonium tautomer $[(Fe^{II}Fe^{II})adt\text{-}\mathbf{H}]^+$ (blue triangles). About 3% of an unknown species is observed at δ 83.61, 83.42, and 75.92. The Fe centers have been labeled Fe_p and Fe_d for consistency, although this labeling is only chemically meaningful for the terminal hydride species.

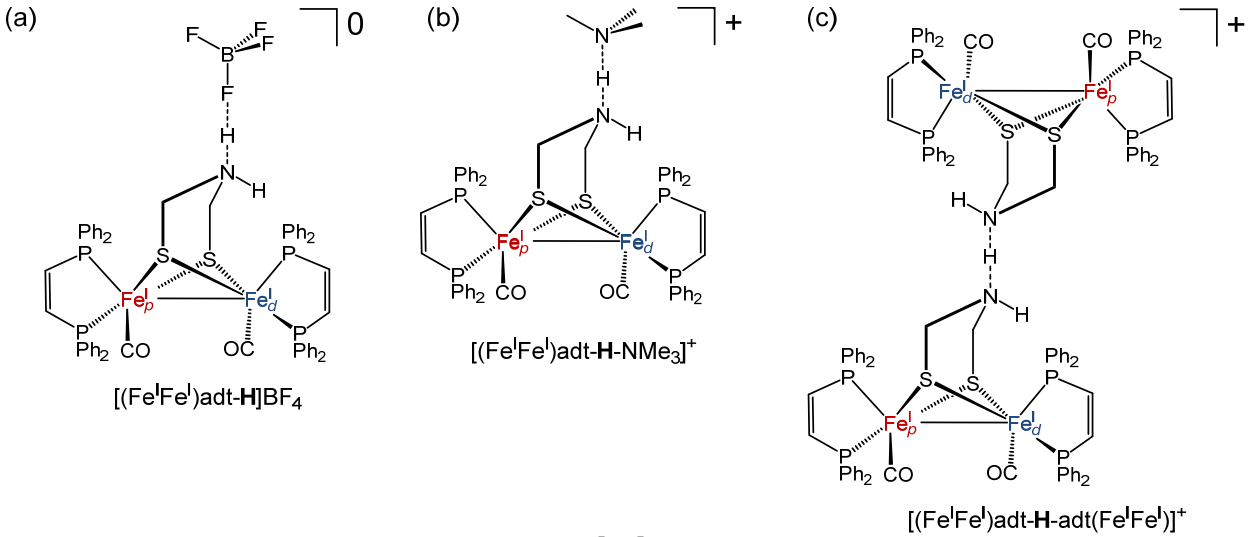


Figure S14. (a) The ion-paired product, $[(Fe^{I}Fe^{I})adt\text{-}\mathbf{H}]BF_4$, arising from the interaction of the ammonium of the $[(Fe^{I}Fe^{I})adt\text{-}\mathbf{H}]^+$ species and BF_4^- ions. (b) The heteroconjugated product, $[(Fe^{I}Fe^{I})adt\text{-}\mathbf{H}\text{-}NMe_3]^+$, formed upon addition of $[(Fe^{I}Fe^{I})adt]^0$ and $[Me_3NH]BAr^F{}_4$ (see **Figures S1b, S2,** and **S13**). (c) The homoconjugated product, $[(Fe^{I}Fe^{I})adt\text{-}\mathbf{H}\text{-}adt(Fe^{I}Fe^{I})]^+$, formed upon addition of $[(Fe^{I}Fe^{I})adt]^0$ and 0.5 equiv of $HBAr^F{}_4$ (see **Figure S3c**). The Fe centers have been labeled Fe_p and Fe_d for consistency, although this labeling is only chemically meaningful for the terminal hydride species.

Estimation of pK_a of [*t*H-(Fe^{II}Fe^{II})adt]$^+$

Initial Conditions:

$$\left[[(Fe^{I}Fe^{I})adt]^{0}\right]_0 = \left[[Me_3NH]BAr_4^F\right]$$

$$\left[[(Fe^{I}Fe^{I})adt]^{0}\right]_0 = \frac{0.006g / 1068\,{}^{g}/_{mol}}{0.0006L}$$

$$\left[[(Fe^{I}Fe^{I})adt]^{0}\right]_0 = 0.0093M$$

Integrated Ratio: from **Figure S13**

89% for [(Fe^{I}Fe^{I})adt-**H**]$^+$ (ammonium tautomer and heteroconjugates)
8% for [*t***H**-(Fe^{II}Fe^{II})adt]$^+$
~3% for unknown species observed in ^{31}P NMR with integration at δ 83.61, 83.42, and 75.92

Combining equimolar amounts of [Me$_3$NH]BAr$^F_4$ and [(Fe^{I}Fe^{I})adt]^0 gives [*t***H**-(Fe^{II}Fe^{II})adt]$^+$ in 8% yield, indicating that the pK_a of [*t***H**-(Fe^{II}Fe^{II})adt]$^+$ is approximately 2.1 pK_a units less than that of [Me$_3$NH]$^+$ (p$K_a^{CH_3CN} = 17.6$[3]).

$$\log K_a\left([t\text{H-}(Fe^{II}Fe^{II})adt]^+\right) = \log\frac{\left[[(Fe^{I}Fe^{I})adt]^0\right]\left[H^+\right]}{\left[[t\text{H-}(Fe^{II}Fe^{II})adt]^+\right]}$$

$$= \log\frac{\left[[(Me_3N]^0\right]\left[H^+\right]}{\left[[Me_3NH]^+\right]} + \log\frac{\left[[Me_3NH]^+\right]\left[(Fe^{I}Fe^{I})adt]^0\right]}{\left[[(Me_3N]^0\right]\left[[t\text{H-}(Fe^{II}Fe^{II})adt]^+\right]}$$

$$= \log K_a\left([Me_3\ NH]^+\right) + \log\frac{\left(0.0093\times 0.89\right)^2}{\left(0.0093\times 0.08\right)^2}$$

$$= -17.6 + \log(123)$$

$$pK_a\left([t\text{H-}(Fe^{II}Fe^{II})adt]^+\right) = 15.5$$

Estimation of pK_a of [($Fe^{I}Fe^{I}$)adt-H]$^+$

Since the ammonium tautomer, [($Fe^{I}Fe^{I}$)adt-**H**]$^+$, cannot be detected in the NMR spectrum (**Figure S1C**) of a solution prepared from treating [($Fe^{I}Fe^{I}$)adt]^0 with 1 equiv of the strong acid [$H(OEt_2)_2$]BAr^F_4, we know that the Fe is at least 20x (conservative NMR detection limits), or 1.3 pK_a units, more basic than the amine.

$$K_a\left([(Fe^{I}Fe^{I})\text{adt-H}]^+\right) < 20 \times K_a\left([t\text{H-}(Fe^{II}Fe^{II})\text{adt}]^+\right)$$

$$pK_a\left([(Fe^{I}Fe^{I})\text{adt-H}]^+\right) < 15.5 - 1.3$$

$$pK_a\left([(Fe^{I}Fe^{I})\text{adt-H}]^+\right) < 14.2$$

Estimation of pK_a of [*t*H-($Fe^{II}Fe^{II}$)adt-H]$^{2+}$

Initial Conditions:

$$\left[[(Fe^{I}Fe^{I})adt]^{0}\right]_0 = \frac{0.006g / 1068\,{}^{g}\!/_{mol}}{0.0006L}$$

$$\left[[(Fe^{I}Fe^{I})adt]^{0}\right]_0 = 0.0093M$$

Integrated Ratio: from **Figure S12**

54% for [*t***H**-($Fe^{II}Fe^{II}$)adt-**H**]$^{2+}$
20% for [*t***H**-($Fe^{II}Fe^{II}$)adt]$^{+}$
~9% for bridging hydride, *sym*-[μ**H**-($Fe^{II}Fe^{II}$)adt]$^{+}$
~17% for unknown species observed in ^{31}P NMR with integration at δ 96.72, 95.48, 85.32, 83.52, 81.43, 72.47

Using one or more equiv of $PhNH_3^+$ (p$K_a^{CH_3CN}$ = 10.6[4]), which has a low self-association constant) in CH_3CN solution, gave only the singly protonated terminal hydride species, [*t***H**-($Fe^{II}Fe^{II}$)adt]$^{+}$. The ammonium tautomer, [($Fe^{I}Fe^{I}$)adt-**H**]$^{+}$, was not present. The ready formation of the singly protonated terminal hydride species, [*t***H**-($Fe^{II}Fe^{II}$)adt]$^{+}$, is consistent with its high pK_a ~ 15.5. The nonobservation of the doubly protonated terminal hydride species, [*t***H**-($Fe^{II}Fe^{II}$)adt-**H**]$^{2+}$, is consistent with the second pK_a being <10.

The pK_a of N-protonation of [*t***H**-($Fe^{II}Fe^{II}$)adt]$^{+}$ is indicated by the observation that the doubly protonated species, [*t***H**-($Fe^{II}Fe^{II}$)adt-**H**]$^{2+}$, is partially generated upon treatment of [($Fe^{I}Fe^{I}$)adt]^{0} with 2 equivalents of $[Ph_2NH_2]BAr^F{}_4$ (p$K_a^{CH_3CN}$ = 5.97[5]).

Computational Details

The details of the methodology are provided in the main text. Unless otherwise noted, all structures were optimized in the gas phase with the B3P86 functional using the SDD pseudopotential and associated basis set for the Fe centers, the 6-31G** basis set for the active hydrogens, and the 6-31G* basis set for all other atoms. Solvation free energies in dichloromethane (CH_2Cl_2) were obtained using C-PCM with Bondi atomic radii and including nonelectrostatic contributions of dispersion, repulsion, and cavitation energies. Thermochemical data were calculated at T = 298.15 K unless otherwise noted.

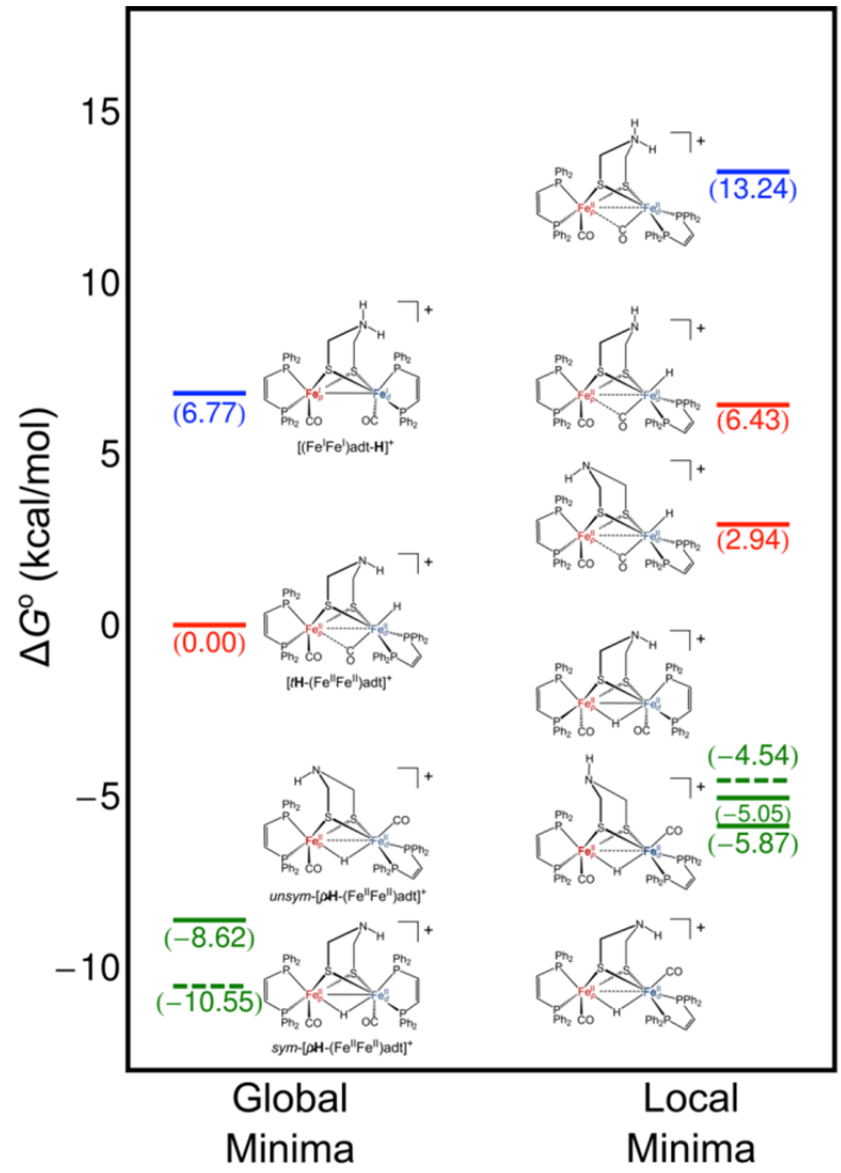


Figure S15. Calculated free energies (in kcal/mol) of the singly protonated isomers relative to the terminal hydride species, [*t*H-($Fe^{II}Fe^{II}$)adt]$^+$. On the left are the lowest energy isomers, which are discussed in the main text; on the right are higher energy isomers. Blue lines refer to the ammonium tautomers, red lines refer to the terminal hydride species, solid green lines refer to the unsymmetric bridging hydrides, and dashed green lines refer to the symmetric bridging hydrides. The local minimum for the symmetric bridging hydride (green dashed lines) differs from the global minimum in terms of the orientation of the dppv ligands. In the global minimum, the dppv ligands are oriented in opposite directions: the Fe_p(dppv) tilts inward and the Fe_d(dppv) ring tilts outward, giving the molecule pseudo-C_2 symmetry if the adt bridge is not considered. In contrast, the local minimum form has both Fe_p(dppv) and Fe_d(dppv) rings tilting in the same orientation (as depicted, they are both tilting outward), which gives the molecule pseudo-C_s symmetry if the adt bridge is not considered.

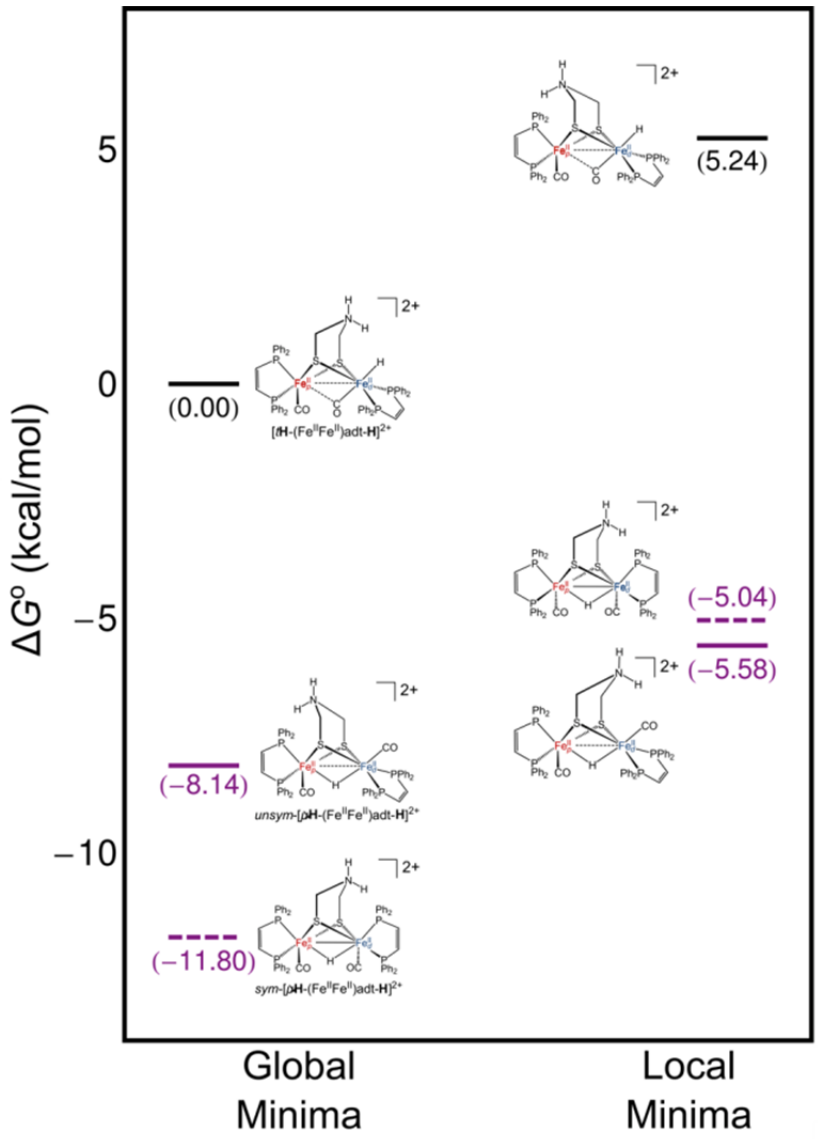


Figure S16. Calculated free energies (in kcal/mol) of the doubly protonated isomers relative to the doubly protonated terminal hydride species, [*t***H**-($Fe^{II}Fe^{II}$)adt-**H**]$^{2+}$. On the left are the lowest energy isomers, which are discussed in the main text; on the right are higher energy isomers. Black lines refer to the doubly protonated terminal hydrides species, solid purple lines refer to the unsymmetric bridging hydrides, and dashed purple lines refer to the symmetric bridging hydrides. The local minimum for the symmetric bridging hydride (purple dashed lines) differs from the global minimum in terms of the orientation of the dppv ligands. In the global minimum, the dppv ligands are oriented in opposite directions: the Fe_p(dppv) tilts inward and the Fe_d(dppv) ring tilts outward, giving the molecule pseudo-C_2 symmetry if the adt bridge is not considered. In contrast, the local minimum form has both Fe_p(dppv) and Fe_d(dppv) rings tilting in the same orientation (as depicted, they are both tilting outward), which gives the molecule pseudo-C_s symmetry if the adt bridge is not considered.

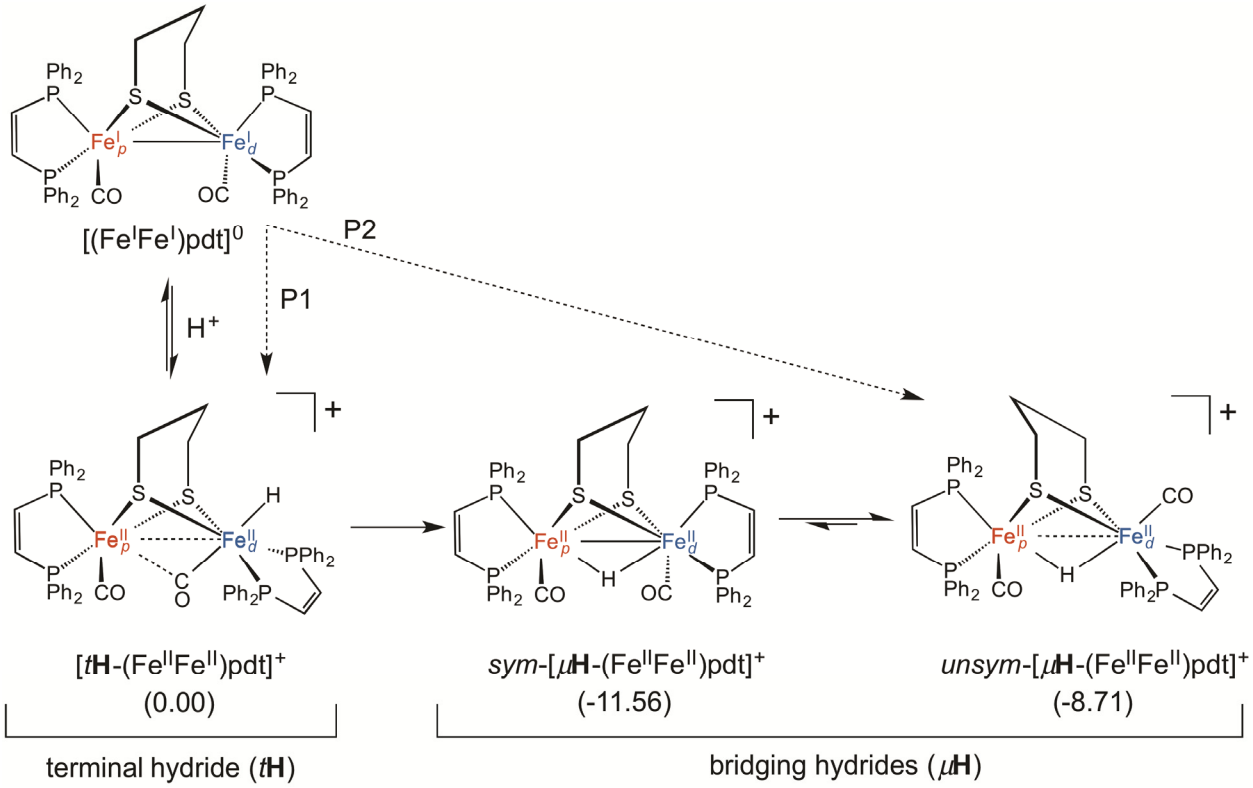


Figure S17: Relative Free Energies of [(FeFe)pdt] Species
Schematic depiction of the various protonated states of the [(FeFe)pdt] species, where each row represents a protonation state of the catalyst. The free energies are provided for the singly protonated states in kcal/mol relative to the terminal hydride species, $[t\mathbf{H}\text{-}(Fe^{II}Fe^{II})pdt]^{+}$. The dashed lines (P1 and P2) correspond to pathways for which structural and/or electronic changes can be monitored by $\Delta\nu_{CO}$ values; they do not necessarily correspond to reaction pathways. The $\Delta\nu_{CO}$ values are given in Table S1.

Table S1: Experimental and Calculated $\Delta\nu_{CO}$ Values[a] upon Protonation of the [(FeFe)pdt] Species[b,c]

	expt[d]	calc[e]
P1[f]	77, 37	87, 42
P2	83, 80	90, 77

[a]Values given in cm^{-1}. [b]All geometries were optimized in the gas phase using the B3P86 functional with the SDD pseudopotential and associated basis set for the Fe centers, the 6-31G** basis set for the active hydrogen, and the 6-31G* basis set for all other atoms. [c]The P1 and P2 protonation reactions are defined in Figure S17. [d]Experimental data from ref 1. [e]Frequencies scaled by a factor of 0.9850.[6] [f]The smaller $\Delta\nu_{CO}$ value is assigned to the semibridging CO.

Table S2: Comparison of Structures Optimized in the Gas Phase and Solution Phase[a]

species		$Fe_p\cdots Fe_d$	$Fe_p\cdots CO$	$Fe_d\cdots CO$	$Fe_d\cdots H$
$[(Fe^{I}Fe^{I})adt]^{0}$	expt[b]	2.60	1.74	1.75	
	gas-opt[c]	2.53	1.73	1.73	n/a
	sol-opt[c,d]	2.54	1.72	1.72	
$[t\mathbf{H}\text{-}(Fe^{II}Fe^{II})adt\text{-}\mathbf{H}]^{2+}$	expt[b]	2.62	1.77	1.79	1.44
	gas-opt[c]	2.65	1.77	1.78	1.53
	sol-opt[c,d]	2.61	1.76	1.77	1.51

[a]Values given in Å. [b]Experimental data from ref 1. [c]Geometries were optimized using the B3P86 functional with the SDD pseudopotential and associated basis set for the Fe centers, the 6-31G** basis set for the active hydrogen, and the 6-31G* basis set for all other atoms. [d]Geometries optimized in CH_2Cl_2 solvent using C-PCM.

Table S3: Bond Lengths[a] for the Doubly Protonated [(FeFe)adt] Species using Various Functionals[b]

	$Fe_p\cdots Fe_d$	$Fe_d\cdots H$	$H\cdots H$
Expt[c]	2.62	1.44	1.88
B3P86	2.65	1.53	1.40
M06-L	2.61	1.55	1.49
TPSSh	2.64	1.54	1.43
B3LYP	2.75	1.54	1.40
ωB97XD	2.65	1.52	1.43

[a]Values given in Å. [b]All geometry optimizations were performed in the gas phase. [c]Experimental data from ref 1.

Table S4: Experimental and Calculated Absolute ν_{CO} Values[a]

species	ν_{CO} (expt)[b]	ν_{CO} (calc)[c,d]
$[(Fe^{I}Fe^{I})adt]^{0}$	1868, 1888	1942, 1964
$[(Fe^{II}Fe^{II})adt\text{-}\mathbf{H}]^{+}$	1890, 1910	1973, 1995
$[t\mathbf{H}\text{-}(Fe^{II}Fe^{II})adt]^{+}$	1914, 1966	1979, 2050
$sym\text{-}[\mu\mathbf{H}\text{-}(Fe^{II}Fe^{II})adt]^{+}$	n/a[e]	2031, 2040
$unsym\text{-}[\mu\mathbf{H}\text{-}(Fe^{II}Fe^{II})adt]^{+}$	1950, 1967	2031, 2041
$[t\mathbf{H}\text{-}(Fe^{II}Fe^{II})adt\text{-}\mathbf{H}]^{2+}$	1935, 1990	2013, 2077
$sym\text{-}[\mu\mathbf{H}\text{-}(Fe^{II}Fe^{II})adt\text{-}\mathbf{H}]^{2+}$	1967, 1986	2060, 2071
$unsym\text{-}[\mu\mathbf{H}\text{-}(Fe^{II}Fe^{II})adt\text{-}\mathbf{H}]^{2+}$	n/a[e]	1991, 2068
$[(Fe^{I}Fe^{I})pdt]^{0}$	1868, 1888	1941, 1964
$[t\mathbf{H}\text{-}(Fe^{II}Fe^{II})pdt]^{+}$	1905, 1965	1983, 2051
$sym\text{-}[\mu\mathbf{H}\text{-}(Fe^{II}Fe^{II})adt]^{+}$	n/a[e]	2030, 2039
$unsym\text{-}[\mu\mathbf{H}\text{-}(Fe^{II}Fe^{II})adt]^{+}$	1951, 1968	2032, 2041

[a]Values given in cm^{-1}. [b]Experimental data from ref 1. [c]Geometries were optimized using the B3P86 functional with the SDD pseudopotential and associated basis set for the Fe centers, the 6-31G** basis set for the active hydrogen, and the 6-31G* basis set for all other atoms. [d]Frequencies scaled by a factor of 0.9850.[6] [e]These isomers were observed using NMR spectroscopy but were not observed using IR spectroscopy.

Table S5: Experimental and Calculated Reduction Potentials[a,b]

species		$E°$ $[(FeFe)]^{0/-}$	$E°$ $[t\mathbf{H}\text{-}(FeFe)]^{+/0}$	$E°$ $[\mu\mathbf{H}\text{-}(FeFe)]^{+/0}$
[(FeFe)pdt]	expt[c]	n/a	–1.70[e]	–1.80
	calc[d]		*–1.70*	*–1.80*
[(FeFe)adt]	expt[f]	n/a	–1.63[g]	–1.79
	calc		–1.66	–1.77
$[(FeFe)pdt(CO)_3(PMe_3)(dppv)]$	expt[i]	n/a	n/a	–1.68
	calc			–1.62
$[(FeFe)pdt(CO)_6]$	expt[h]	–1.65	n/a	n/a
	calc[d]	*–1.65*		
$[(FeFe)pdt(CO)_4(dppv)]$	expt[i]	–2.09	n/a	–1.27
	calc	–2.13		–1.33
$[(FeFe)pdt(CO)_4(PMe_3)_2]$	expt[i]	–2.63	n/a	–1.52
	calc	–2.34		–1.40

[a]Values given in Volts vs Fc^0/Fc^+ in dichloromethane (CH_2Cl_2) solvent. [b]All geometries were optimized in the gas phase using the B3P86 functional with the SDD pseudopotential and associated basis set for the Fe centers, the 6-31G** basis set for the active hydrogen, and the 6-31G* basis set for all other atoms. [c]Experimental data from ref 7. [d]These reactions were used as the reference reactions, so the calculated and experimental values agree by construction [e]Quasi-reversible reaction so E_p is reported. [f]Experimental data from ref 1. [g]Irreversible reaction, so E_p reported. [h]Experimental data from ref 8. [i]Experimental data from ref 2.

Table S6: Spin Densities and Bond Lengths[a] of Oxidized and Reduced Mixed-Valence [(FeFe)pdt] Species[b]

Oxidized					
species	$\rho(Fe_p)$	$\rho(Fe_d)$	$Fe_p\cdots Fe_d$	$Fe_p\cdots H$	$Fe_d\cdots H$
$[(Fe^{II}Fe^{II})pdt]^0$	0.00	0.00	2.53	n/a	n/a
$[t\mathbf{H}\text{-}(Fe^{II}Fe^{II})pdt]^+$	0.00	0.00	2.59	n/a	1.50
sym-$[\mu\mathbf{H}\text{-}(Fe^{II}Fe^{II})pdt]^+$	0.00	0.00	2.61	1.65	1.64
unsym-$[\mu\mathbf{H}\text{-}(Fe^{II}Fe^{II})pdt]^+$	0.00	0.00	2.66	1.68	1.71
Reduced					
species	$\rho(Fe_p)$	$\rho(Fe_d)$	$Fe_p\cdots Fe_d$	$Fe_p\cdots H$	$Fe_d\cdots H$
$[(Fe^{II}Fe^{I})pdt]^+$	0.03	1.11	2.59	n/a	n/a
$[t\mathbf{H}\text{-}(Fe^{I}Fe^{II})pdt]^0$	1.02	0.07	2.94	n/a	1.52
sym-$[\mu\mathbf{H}\text{-}(Fe^{1.5}Fe^{1.5})pdt]^0$	0.78	0.37	2.74	1.76	1.59
unsym-$[\mu\mathbf{H}\text{-}(Fe^{I}Fe^{II})pdt]^0$	0.96	0.18	2.85	2.01	1.59

[a]Values given in Å. [b]All geometries were optimized in the gas phase using the B3P86 functional with the SDD pseudopotential and associated basis set for the Fe centers, the 6-31G** basis set for the active hydrogen, and the 6-31G* basis set for all other atoms.

Table S7: Spin Densities and Bond Lengths[a] for the Reduced Mixed-Valence Bridging Hydride Forms of the [(FeFe)pdt] Species using the BP86 Functional and Various Basis Sets

Basis Set: SDD (Fe) / 6-31G** (μ**H**) / 6-31G* (all other atoms)					
species	$\rho(Fe_p)$	$\rho(Fe_d)$	$Fe_p\cdots Fe_d$	$Fe_p\cdots H$	$Fe_d\cdots H$
sym-[μ**H**-($Fe^{1.5}Fe^{1.5}$)pdt]^0	0.47	0.36	2.75	1.70	1.67
unsym-[μ**H**-($Fe^{I}Fe^{II}$)pdt]^0	0.57	0.31	2.81	1.76	1.71
Basis Set: SDD (Fe) / D95* (all other atoms)					
species	$\rho(Fe_p)$	$\rho(Fe_d)$	$Fe_p\cdots Fe_d$	$Fe_p\cdots H$	$Fe_d\cdots H$
sym-[μ**H**-($Fe^{1.5}Fe^{1.5}$)pdt]^0	0.51	0.42	2.75	1.71	1.67
unsym-[μ**H**-($Fe^{I}Fe^{II}$)pdt]^0	0.64	0.32	2.81	1.79	1.70
Basis Set: TZVP[b] (Fe) / 6-31G** (μ**H**) / 6-31G* (all other atoms)					
species	$\rho(Fe_p)$	$\rho(Fe_d)$	$Fe_p\cdots Fe_d$	$Fe_p\cdots H$	$Fe_d\cdots H$
sym-[μ**H**-($Fe^{1.5}Fe^{1.5}$)pdt]^0	0.47	0.39	2.75	1.70	1.67
unsym-[μ**H**-($Fe^{I}Fe^{II}$)pdt]^0	0.56	0.31	2.81	1.76	1.71
Basis Set: TZVP[b] (Fe) / D95* (all other atoms)					
species	$\rho(Fe_p)$	$\rho(Fe_d)$	$Fe_p\cdots Fe_d$	$Fe_p\cdots H$	$Fe_d\cdots H$
sym-[μ**H**-($Fe^{1.5}Fe^{1.5}$)pdt]^0	0.47	0.39	2.75	1.70	1.67
unsym-[μ**H**-($Fe^{I}Fe^{II}$)pdt]^0	0.57	0.31	2.81	1.76	1.71

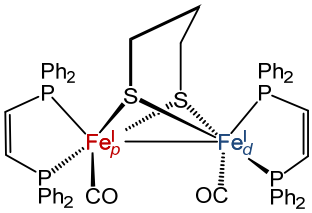
[a]Values given in Å. [b]No pseudopotential was used for the TZVP basis set on the Fe center.

Table S8. Experimental and Calculated ΔpK_a Values[a] of the [(FeFe)adt] Species using Different Functionals

		calc[c]							
		B3P86		BP86		M06-L		ωB97XD	
	expt[b]	233K	298K	233K	298K	233K	298K	233K	298K
ΔpK_a (P2 – P1)	>1.3	6	5	5	4	8	6	4	3
ΔpK_a (P4 – P2)	–9.5	–18	–14	–16	–13	–19	–15	–10	–8
ΔpK_a (P3 – P2)	>2.0	8	6	4	3	8	6	20	15

[a]The protonation pathways P1, P2, P3, and P4 are defined in Figure 3 in the main paper, and the pK_a is determined for the protonated species in each case. [b]These experiments were performed in CH_2Cl_2 solvent, but the reference pK_a values pertain to CH_3CN solvent. [c]The values were calculated in CH_2Cl_2 solvent.

Table S9: Cartesian coordinates of the optimized structure of $[(Fe^{I}Fe^{I})pdt]^{0}$ (–4772.17273608 E_h)



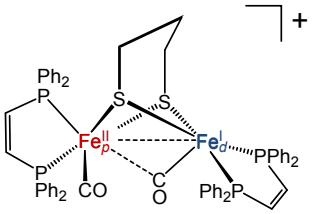
$[(Fe^{I}Fe^{I})pdt]^{0}$

atom	x	y	z
Fe	5.253654	4.186127	4.516163
Fe	5.000536	6.255282	5.953701
S	6.998949	5.193392	5.567261
S	4.875367	6.275426	3.677138
P	5.936775	3.123960	2.722515
P	5.588014	2.277931	5.654418
P	5.568139	8.214309	6.791629
P	2.859831	6.910070	6.190059
O	2.446414	3.560006	4.167961
O	4.866415	4.843985	8.478321
C	7.546086	7.270461	3.676303
H	7.253686	8.063670	4.368032
C	4.916876	3.138966	1.184753
C	3.904593	4.088918	1.021854
H	3.743648	4.833002	1.796373
C	3.121160	4.096191	-0.131695
H	2.341572	4.844460	-0.244948
C	3.333722	3.152365	-1.131185
H	2.718436	3.155797	-2.026839
C	4.344369	2.203235	-0.981824
H	4.521581	1.466463	-1.760641
C	5.131792	2.200268	0.164468
H	5.932183	1.470517	0.253378
C	7.638328	3.330396	2.026422
C	8.734378	2.970713	2.822166
H	8.568343	2.566871	3.817026
C	10.035713	3.153525	2.365076
H	10.872127	2.863810	2.995312
C	10.265072	3.717927	1.110940
H	11.281079	3.866079	0.755720
C	9.184039	4.096262	0.320054
H	9.352039	4.542658	-0.656410
C	7.879458	3.901046	0.771506
H	7.046162	4.188761	0.137588
C	5.942013	1.328653	3.110120
H	6.073834	0.589486	2.321024
C	5.769074	0.963302	4.383063
H	5.749494	-0.086568	4.674754
C	4.331018	1.507609	6.762768
C	4.527886	0.209042	7.255033
H	5.438998	-0.332558	7.012888
C	3.578872	-0.387920	8.076051
H	3.743804	-1.395085	8.449496
C	2.422195	0.309767	8.424185
H	1.680003	-0.155575	9.067464
C	2.226604	1.604146	7.954834

H	1.337565	2.163124	8.229874
C	3.177882	2.202661	7.129069
H	3.024526	3.216955	6.778092
C	7.110213	1.977183	6.657310
C	8.194156	1.245661	6.161403
H	8.149095	0.814983	5.164911
C	9.333339	1.044344	6.940187
H	10.163027	0.467258	6.540267
C	9.401356	1.570250	8.226280
H	10.287020	1.411617	8.835562
C	8.322395	2.294407	8.731141
H	8.362627	2.703093	9.737230
C	7.185769	2.497559	7.955477
H	6.351778	3.059407	8.363727
C	3.579843	3.803856	4.319567
C	8.135587	6.110891	4.454936
H	8.947320	6.476491	5.094619
H	8.566277	5.367578	3.776259
C	6.358437	6.918633	2.801769
H	6.638842	6.177689	2.044425
H	6.019061	7.816514	2.273562
C	4.912716	5.414137	7.459596
C	6.785941	8.314435	8.172166
C	6.883267	9.453606	8.985814
H	6.264484	10.324171	8.784624
C	7.784619	9.496064	10.044505
H	7.845860	10.385371	10.665997
C	8.607034	8.400519	10.304868
H	9.310600	8.433078	11.132375
C	8.522707	7.268140	9.501064
H	9.160628	6.410564	9.695684
C	7.616095	7.222211	8.442689
H	7.550002	6.334800	7.818786
C	6.020778	9.677296	5.746707
C	7.204039	10.407817	5.897057
H	7.906219	10.149502	6.683544
C	7.496966	11.465255	5.036533
H	8.423008	12.019435	5.164798
C	6.611601	11.809517	4.019661
H	6.841170	12.634552	3.350941
C	5.431619	11.083756	3.859080
H	4.736475	11.338655	3.063716
C	5.142057	10.022666	4.710049
H	4.234325	9.444578	4.555901
C	4.073089	8.911080	7.603008
H	4.151387	9.773119	8.264052
C	2.893878	8.348061	7.330947
H	1.965114	8.712829	7.769536
C	1.884065	7.597186	4.781542
C	1.486522	8.939945	4.758611

H	1.745864	9.599301	5.581104
C	0.762447	9.453142	3.684169
H	0.463353	10.497870	3.689372
C	0.418894	8.630772	2.615527
H	-0.150577	9.028319	1.779906
C	0.814261	7.295464	2.623689
H	0.556360	6.644553	1.792788
C	1.550258	6.784296	3.689082
H	1.861133	5.746049	3.666639
C	1.599033	5.897849	7.092405
C	0.508809	5.284912	6.466213
H	0.367863	5.380826	5.395766
C	-0.420883	4.561992	7.210472
H	-1.263851	4.098317	6.705666
C	-0.278540	4.442242	8.590715
H	-1.012176	3.888496	9.170505
C	0.810480	5.038673	9.222987
H	0.936732	4.948483	10.298329
C	1.742892	5.756808	8.480149
H	2.585576	6.215570	8.988713
H	8.336162	7.692125	3.035912

Table S10: Cartesian coordinates of the optimized structure of $[(Fe^{II}Fe^{I})pdt]^{+}$ (–4772.00017013 E_h)



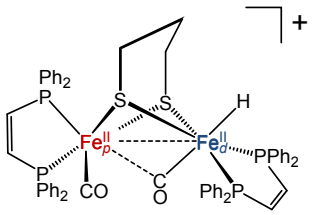
$[(Fe^{II}Fe^{I})pdt]^{+}$

atom	x	y	z
Fe	1.235518	6.242631	3.356462
Fe	2.665259	4.103853	3.628214
S	0.498432	4.149800	2.859644
S	1.860092	5.363329	5.379669
P	-0.683354	7.262904	3.857985
P	2.259539	8.262053	3.761124
P	4.551734	3.424500	4.709203
P	3.025931	2.371168	2.239175
O	0.987914	6.895661	0.528640
O	4.174740	5.953209	1.943212
C	-2.137656	7.121335	2.750258
C	-2.169236	6.195864	1.702874
H	-1.336174	5.517175	1.552733
C	-3.274842	6.128814	0.856144
H	-3.285896	5.405441	0.046297
C	-4.355444	6.983804	1.045589
H	-5.213980	6.932911	0.382460
C	-4.337009	7.905030	2.092807
H	-5.180697	8.570457	2.250035
C	-3.238306	7.972294	2.941124
H	-3.249518	8.679414	3.766297
C	-1.412820	7.063783	5.531434
C	-2.598157	6.348251	5.736137
H	-3.142023	5.939741	4.889580
C	-3.097239	6.170754	7.025259
H	-4.019604	5.616469	7.172206
C	-2.421694	6.708282	8.117819
H	-2.815283	6.573999	9.121032
C	-1.241065	7.423341	7.920028
H	-0.710839	7.846778	8.767983
C	-0.735685	7.598038	6.635633
H	0.194868	8.141209	6.495965
C	-0.358252	9.061590	3.828562
H	-1.194173	9.758640	3.854374
C	0.906505	9.487446	3.794737
H	1.152086	10.548396	3.768113
C	3.129884	8.559293	5.343638
C	4.208973	7.738873	5.695733
H	4.513443	6.922749	5.046531
C	4.904173	7.961051	6.880953
H	5.746732	7.323391	7.129430
C	4.517890	8.988371	7.738816
H	5.059619	9.158296	8.664715
C	3.434968	9.797031	7.405752
H	3.127958	10.600739	8.068695
C	2.746241	9.587165	6.213319

H	1.909419	10.233374	5.966899
C	3.356426	9.025030	2.499249
C	2.775568	9.522934	1.325338
H	1.700683	9.468504	1.177610
C	3.566449	10.104279	0.339043
H	3.101619	10.491221	-0.562958
C	4.945368	10.193081	0.511981
H	5.562663	10.648630	-0.256639
C	5.528232	9.702790	1.677527
H	6.601978	9.777427	1.822752
C	4.741006	9.122040	2.667701
H	5.210504	8.757431	3.575213
C	-0.568604	3.391030	4.144994
H	-1.433420	4.046934	4.295120
H	-0.929233	2.461184	3.694146
C	0.125357	3.094274	5.460549
H	0.970986	2.417333	5.281245
H	-0.579986	2.550059	6.105445
C	0.596210	4.323637	6.214565
H	1.065589	4.015706	7.154046
H	-0.246986	4.972395	6.466564
C	4.298287	2.597766	6.326961
C	4.578853	1.238631	6.511063
H	4.973539	0.644805	5.691730
C	4.363507	0.632988	7.747559
H	4.586993	-0.422197	7.874875
C	3.875026	1.378745	8.816335
H	3.712363	0.907052	9.780847
C	3.600031	2.734703	8.645136
H	3.225208	3.323885	9.477199
C	3.802939	3.341323	7.408856
H	3.579611	4.397529	7.281277
C	6.018704	4.500943	4.984838
C	6.603251	5.110502	3.864624
H	6.160547	4.991602	2.881555
C	7.760831	5.871032	4.002130
H	8.204285	6.332917	3.125004
C	8.351504	6.030196	5.253965
H	9.257809	6.619278	5.358797
C	7.781400	5.420485	6.368598
H	8.245018	5.526060	7.345408
C	6.621413	4.658623	6.238087
H	6.199345	4.176088	7.113297
C	5.322801	2.119324	3.684988
H	6.311699	1.750180	3.955332
C	4.674785	1.674154	2.604305
H	5.103722	0.911330	1.956243
C	1.910040	0.944908	2.547131
C	0.805250	0.715878	1.717335
H	0.644245	1.339441	0.843307

C	-0.084104	-0.316338	2.005035
H	-0.933974	-0.489992	1.351311
C	0.118663	-1.128771	3.119209
H	-0.572974	-1.937340	3.336650
C	1.214757	-0.904871	3.949152
H	1.381598	-1.537571	4.816075
C	2.104565	0.128720	3.667772
H	2.953997	0.295508	4.324304
C	2.689168	3.813651	-1.597875
H	2.411645	4.750632	-2.070974
C	2.672389	3.714806	-0.208018
H	2.380601	4.572304	0.384949
C	1.087416	6.663880	1.655964
C	3.427410	5.365613	2.623358
C	3.025857	2.517379	0.417543
C	3.387104	1.413080	-0.368722
H	3.635342	0.463564	0.098423
C	3.407829	1.515905	-1.754718
H	3.691369	0.656416	-2.354919
C	3.060853	2.718234	-2.370988
H	3.077371	2.796733	-3.454058

Table S11: Cartesian coordinates of the optimized structure of [*t***H**-($Fe^{II}Fe^{II}$)pdt]$^{+}$ (–4772.61253629 E_h)



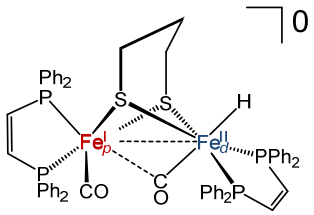
[*t***H**-($Fe^{II}Fe^{II}$)pdt]$^{+}$

atom	x	y	z
Fe	12.418874	3.295460	4.103115
Fe	14.030377	5.306776	4.363357
S	13.222384	4.560551	2.368542
S	14.611904	3.152060	4.697286
P	10.468949	2.893672	3.096764
P	11.900960	1.711493	5.570303
P	15.972006	6.233788	3.785380
P	13.106899	7.402976	4.111534
C	14.896913	2.255579	2.002317
O	11.246499	5.210619	5.960640
O	14.532724	5.777125	7.199300
C	10.617698	2.383294	1.345608
C	10.610449	1.028113	0.998135
H	10.468905	0.269147	1.762877
C	10.787497	0.637451	-0.327634
H	10.774480	-0.418028	-0.583439
C	10.973999	1.596207	-1.319584
H	11.107773	1.291854	-2.353458
C	10.985923	2.948610	-0.981347
H	11.130181	3.701659	-1.750760
C	10.813257	3.343247	0.342814
H	10.835727	4.399587	0.594744
C	8.961929	3.961763	3.123387
C	8.572710	4.522548	4.348563
H	9.181855	4.380498	5.234364
C	7.397952	5.262828	4.446442
H	7.112152	5.688657	5.403789
C	6.591551	5.450312	3.326052
H	5.674142	6.026375	3.403492
C	6.961462	4.884568	2.108621
H	6.330068	5.010942	1.233669
C	8.136562	4.141941	2.005676
H	8.400113	3.694591	1.053297
C	9.753321	1.424378	3.918204
H	8.828714	1.002150	3.525921
C	10.336622	0.949395	5.021956
H	9.931989	0.096348	5.564958
C	11.600995	2.174743	7.311816
C	11.857570	4.581516	5.192395
C	14.367197	3.529347	1.371881
H	13.797210	3.278976	0.471306
H	15.187087	4.184866	1.067753
C	15.605690	2.433558	3.330047
H	16.493700	3.065856	3.222070
H	15.937739	1.460790	3.706515
C	14.329354	5.622453	6.074510

C	16.608584	6.050376	2.072523
C	17.732603	5.266596	1.788449
H	18.281466	4.787349	2.593661
C	18.166233	5.112266	0.472978
H	19.041069	4.503546	0.264130
C	17.487646	5.742783	-0.566649
H	17.830943	5.626455	-1.590320
C	16.369048	6.527860	-0.289614
H	15.837194	7.023816	-1.096188
C	15.928001	6.678850	1.021198
H	15.043885	7.277140	1.223316
C	15.755918	8.046820	3.863628
H	16.627103	8.695411	3.789603
C	14.522144	8.545576	3.969130
H	14.341479	9.618908	4.012929
C	12.201731	8.203500	5.497574
H	12.559965	2.029899	3.307211
H	14.073918	1.545077	2.128253
C	12.997228	0.238607	5.670842
C	13.334086	-0.428098	4.484925
H	12.974377	-0.049263	3.532196
C	14.122959	-1.573065	4.517724
H	14.370811	-2.083418	3.591357
C	14.591819	-2.064191	5.735408
H	15.210604	-2.956332	5.761286
C	14.258455	-1.410145	6.917413
H	14.614633	-1.789974	7.870531
C	13.460401	-0.267809	6.889077
H	13.200075	0.224595	7.819963
C	12.645686	2.774168	8.029625
C	12.451093	3.182486	9.344595
H	13.268134	3.644303	9.890700
C	11.209187	3.008748	9.954040
H	11.056623	3.332275	10.979391
C	10.164057	2.424862	9.244425
H	9.193731	2.290610	9.713433
C	10.357218	2.007694	7.928812
H	9.529874	1.559187	7.387132
C	17.467017	5.959352	4.809815
C	17.510480	4.947453	5.773642
H	16.655024	4.294445	5.914008
C	18.654209	4.762336	6.548923
H	18.674553	3.972255	7.293603
C	19.761407	5.585271	6.371051
H	20.649923	5.442865	6.979002
C	19.731313	6.591322	5.405391
C	18.594926	6.775212	4.626793
C	12.092608	7.780396	2.637493
C	12.455766	8.774956	1.722641
H	13.364112	9.351837	1.868351

C	11.653397	9.041262	0.615076
H	11.945492	9.818670	-0.084892
C	10.477463	8.323997	0.414431
H	9.847712	8.540528	-0.443518
C	10.110071	7.329119	1.318621
H	9.193323	6.764676	1.175481
C	10.918256	7.049474	2.416836
H	10.625278	6.260841	3.104158
C	12.930959	8.583097	6.632418
H	14.002918	8.414114	6.679449
C	12.294110	9.193347	7.708394
H	12.873384	9.486577	8.578906
C	10.922461	9.430213	7.665172
H	10.425353	9.908201	8.504068
C	10.192975	9.059551	6.538821
H	9.124700	9.250228	6.494093
C	10.826209	8.450629	5.458944
H	10.244186	8.181763	4.584002
H	13.614979	2.919027	7.558388
H	20.595714	7.231537	5.256432
H	18.598222	7.546302	3.861269
H	15.606150	1.799330	1.296529

Table S12: Cartesian coordinates of the optimized structure of [*t*H-($Fe^{I}Fe^{II}$)pdt]^{0} (–4772.77741619 E_h)



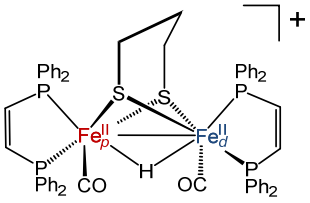
[*t*H-($Fe^{I}Fe^{II}$)pdt]^{0}

atom	x	y	z
Fe	12.203894	3.225618	4.203989
Fe	14.093258	5.460734	4.443761
S	13.135382	4.552239	2.528187
S	14.409692	3.236835	4.970841
P	10.362954	2.792279	3.095101
P	11.703783	1.591573	5.563273
P	16.108002	6.310077	3.712825
P	13.256432	7.517827	4.114950
C	14.874939	2.258628	2.321062
O	11.042214	5.290461	5.928213
O	14.549665	5.970882	7.269907
C	10.562816	2.342512	1.328462
C	10.532797	1.006720	0.914527
H	10.340286	0.218660	1.637548
C	10.758307	0.670729	-0.418737
H	10.730422	-0.372008	-0.723232
C	11.018026	1.667238	-1.355526
H	11.194344	1.406383	-2.395423
C	11.054320	3.000861	-0.952183
H	11.263262	3.784276	-1.675414
C	10.832033	3.339623	0.380138
H	10.886099	4.379997	0.686426
C	8.830977	3.835786	3.082489
C	8.398472	4.368390	4.305059
H	8.980799	4.203377	5.205338
C	7.226443	5.115433	4.380482
H	6.911015	5.521162	5.337603
C	6.466135	5.344208	3.235757
H	5.553366	5.930730	3.293660
C	6.881171	4.812847	2.017413
H	6.289971	4.977122	1.120327
C	8.052466	4.060595	1.940448
H	8.356304	3.644964	0.985281
C	9.628581	1.279706	3.830844
H	8.729069	0.848556	3.391422
C	10.185488	0.777401	4.937192
H	9.780032	-0.108744	5.423647
C	11.342309	1.907797	7.337193
C	11.612665	4.552387	5.229661
C	14.340461	3.498522	1.621997
H	13.835372	3.194708	0.698364
H	15.163445	4.161682	1.341436
C	15.490664	2.475824	3.691343
H	16.375631	3.119801	3.621571
H	15.814471	1.511786	4.098532
C	14.365298	5.808270	6.130137

C	16.745360	6.105846	1.991645
C	17.851842	5.299140	1.697760
H	18.418076	4.842795	2.504635
C	18.239728	5.080777	0.377694
H	19.101945	4.452975	0.168792
C	17.530543	5.665194	-0.668932
H	17.835238	5.495557	-1.697917
C	16.426181	6.467516	-0.387240
H	15.863482	6.925126	-1.196491
C	16.033826	6.683297	0.930288
H	15.158833	7.294467	1.135144
C	15.904886	8.137349	3.717398
H	16.771462	8.785442	3.586091
C	14.677547	8.650247	3.850764
H	14.510857	9.727715	3.854661
C	12.418277	8.440122	5.480040
H	12.503446	1.988110	3.372508
H	14.072409	1.520364	2.410264
C	12.858214	0.158042	5.640820
C	13.054222	-0.608932	4.485473
H	12.524037	-0.348847	3.572988
C	13.928237	-1.691142	4.494758
H	14.066524	-2.280092	3.592017
C	14.628174	-2.014322	5.656382
H	15.314975	-2.856282	5.663034
C	14.444116	-1.252528	6.806561
H	14.988542	-1.495310	7.715007
C	13.560448	-0.174642	6.802151
H	13.422075	0.410553	7.705684
C	12.119754	2.863646	8.003851
C	11.900536	3.138280	9.351144
H	12.511869	3.884407	9.850185
C	10.894644	2.471394	10.046268
H	10.717100	2.693076	11.095031
C	10.112965	1.522337	9.391478
H	9.324729	1.000265	9.927167
C	10.338705	1.237655	8.047222
H	9.722559	0.490726	7.556360
C	17.648007	6.107534	4.703623
C	17.657233	5.159255	5.731573
H	16.767720	4.561622	5.913459
C	18.794694	4.980683	6.518421
H	18.786953	4.240599	7.313712
C	19.931015	5.749940	6.288490
H	20.815860	5.614128	6.904512
C	19.934104	6.695449	5.262601
C	18.802203	6.869841	4.473579
C	12.170242	7.877014	2.675757
C	12.483779	8.840221	1.711218
H	13.398395	9.419682	1.796604

C	11.632609	9.067196	0.631039
H	11.892325	9.819392	-0.109153
C	10.451620	8.341046	0.506170
H	9.784504	8.524397	-0.331801
C	10.130516	7.375562	1.458743
H	9.213960	6.798356	1.372113
C	10.990201	7.135380	2.527117
H	10.739233	6.363996	3.249841
C	13.194768	8.859075	6.568693
H	14.261440	8.655043	6.585994
C	12.613978	9.536045	7.636656
H	13.233457	9.853757	8.470872
C	11.246661	9.800914	7.636844
H	10.791859	10.327704	8.471293
C	10.466990	9.387151	6.560427
H	9.399623	9.590876	6.549394
C	11.047183	8.713345	5.488639
H	10.425487	8.409115	4.653134
H	12.899867	3.396398	7.467604
H	20.821303	7.294702	5.075944
H	18.824094	7.592761	3.661892
H	15.644915	1.815341	1.670081

Table S13: Cartesian coordinates of the optimized structure of *sym*-[μ**H**-($Fe^{II}Fe^{II}$)pdt]$^+$ (–4772.63198301 E_h)



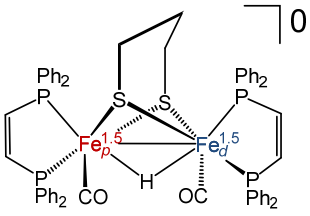
sym-[μ**H**-($Fe^{II}Fe^{II}$)pdt]$^+$

atom	x	y	z
Fe	5.155846	4.128140	4.506640
Fe	4.995650	6.194918	6.092929
S	6.946050	5.054845	5.611703
S	4.821468	6.284632	3.799051
P	6.064959	3.161669	2.686187
P	5.421891	2.123168	5.548439
P	5.623471	8.287389	6.696267
P	2.852875	6.913782	6.468464
O	2.512349	3.309907	3.618696
O	5.262330	5.280793	8.837915
C	7.545725	7.174819	3.786467
H	7.331788	7.975807	4.496104
C	5.220709	3.333217	1.066385
C	4.249238	4.317540	0.861327
H	3.995177	5.003408	1.664017
C	3.613022	4.431574	-0.373898
H	2.862515	5.202550	-0.521263
C	3.936016	3.563219	-1.411389
H	3.435674	3.650030	-2.371371
C	4.908954	2.582627	-1.218612
H	5.170367	1.906176	-2.026925
C	5.550796	2.470926	0.009087
H	6.325317	1.719127	0.133791
C	7.835059	3.369349	2.234173
C	8.822890	2.838085	3.073467
H	8.541821	2.301083	3.974273
C	10.170527	3.015407	2.775821
H	10.925968	2.593867	3.432394
C	10.548527	3.734584	1.643058
H	11.600439	3.872869	1.411437
C	9.572687	4.273322	0.808566
H	9.860006	4.832555	-0.077039
C	8.222003	4.090355	1.098496
H	7.470929	4.498492	0.428940
C	5.972444	1.357402	2.980826
H	6.153726	0.669745	2.156456
C	5.688804	0.911251	4.207595
H	5.621662	-0.154934	4.420189
C	4.040565	1.377914	6.495262
C	4.145925	0.049150	6.932854
H	5.049765	-0.521713	6.736188
C	3.108884	-0.541863	7.643536
H	3.199757	-1.572326	7.974146
C	1.959043	0.191036	7.937328
H	1.149665	-0.270721	8.495314
C	1.854657	1.514736	7.523537

H	0.971348	2.098394	7.761574
C	2.891080	2.108164	6.804278
H	2.801785	3.144825	6.496519
C	6.837870	1.788032	6.667275
C	7.933978	1.015234	6.270768
H	7.964216	0.575307	5.277749
C	8.987441	0.780262	7.153114
H	9.827540	0.168688	6.836639
C	8.955100	1.314797	8.437428
H	9.774146	1.128307	9.125773
C	7.861678	2.079542	8.841615
H	7.824242	2.491359	9.845839
C	6.808487	2.313868	7.964513
H	5.956954	2.898342	8.298693
C	3.571811	3.625464	3.958948
C	8.099808	5.975397	4.526741
H	8.917084	6.298019	5.180878
H	8.514954	5.241001	3.832664
C	6.313163	6.924916	2.939179
H	6.530399	6.224741	2.125355
H	5.995275	7.867351	2.482097
C	5.141601	5.670750	7.756748
C	7.009353	8.496196	7.877239
C	7.168271	9.694442	8.591798
H	6.492545	10.528979	8.425879
C	8.205524	9.838593	9.506366
H	8.315635	10.770267	10.053352
C	9.100302	8.790346	9.719080
H	9.909041	8.903954	10.434936
C	8.953091	7.600413	9.013604
H	9.645371	6.779634	9.175971
C	7.912454	7.451296	8.098458
H	7.797943	6.514458	7.560753
C	5.854504	9.645964	5.470159
C	6.995887	10.454824	5.437423
H	7.799053	10.297537	6.150120
C	7.114221	11.463560	4.483318
H	8.005782	12.083649	4.468033
C	6.097333	11.676862	3.556861
H	6.191118	12.466041	2.816705
C	4.959031	10.872581	3.582016
H	4.160879	11.030831	2.862582
C	4.838529	9.860561	4.529213
H	3.951339	9.233747	4.527811
C	4.210446	8.967659	7.643787
H	4.352147	9.859185	8.252129
C	3.014224	8.384986	7.534686
H	2.139970	8.761562	8.064124
C	1.793476	7.513298	5.095804
C	1.295594	8.822203	5.076914

H	1.545265	9.516640	5.873030
C	0.478111	9.252696	4.034284
H	0.097857	10.270026	4.036510
C	0.142513	8.381295	3.001792
H	-0.500801	8.716071	2.193441
C	0.635310	7.078458	3.010186
H	0.379402	6.391347	2.209089
C	1.462796	6.649119	4.043628
H	1.842426	5.633793	4.027366
C	1.697858	5.900378	7.484310
C	0.536453	5.323515	6.958999
H	0.287293	5.447621	5.910573
C	-0.333750	4.615317	7.785897
H	-1.239286	4.187165	7.365625
C	-0.055985	4.474442	9.143381
H	-0.742252	3.932930	9.787896
C	1.101195	5.041223	9.673243
H	1.323901	4.942233	10.731487
C	1.972316	5.749378	8.851257
H	2.859774	6.198692	9.286319
H	8.336552	7.559202	3.126060
H	4.372555	4.689175	5.838095

Table S14: Cartesian coordinates of the optimized structure of *sym*-[μ**H**-($Fe^{1.5}Fe^{1.5}$)pdt]^0 (–4772.78320098 E_h)



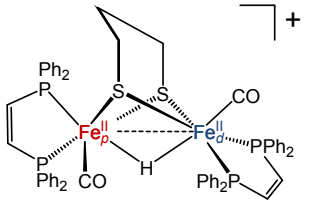
sym-[μ**H**-($Fe^{1.5}Fe^{1.5}$)pdt]^0

atom	x	y	z
Fe	5.140208	4.075987	4.459059
Fe	4.978115	6.224414	6.147703
S	6.938638	5.019307	5.627156
S	4.749204	6.280335	3.813517
P	6.141502	3.151248	2.630448
P	5.375872	2.085481	5.429998
P	5.607677	8.400991	6.696407
P	2.874849	6.918914	6.583815
O	2.502614	3.354385	3.507156
O	5.347925	5.317199	8.876057
C	7.470738	7.159639	3.801777
H	7.234594	7.930880	4.538335
C	5.386584	3.354143	0.962643
C	4.428387	4.351771	0.758770
H	4.154424	5.007802	1.581334
C	3.830653	4.510538	-0.491153
H	3.089249	5.291499	-0.634909
C	4.178997	3.673426	-1.546133
H	3.709200	3.795247	-2.518355
C	5.136823	2.677652	-1.354278
H	5.417069	2.023945	-2.175799
C	5.738392	2.521803	-0.110363
H	6.500804	1.757896	0.019232
C	7.943748	3.244302	2.244732
C	8.854738	2.708069	3.165419
H	8.492501	2.214184	4.062383
C	10.225185	2.830282	2.958843
H	10.915995	2.409249	3.684112
C	10.708736	3.498058	1.835080
H	11.778957	3.596265	1.675971
C	9.811530	4.043477	0.920080
H	10.178748	4.569161	0.042730
C	8.438739	3.918240	1.121636
H	7.750732	4.345107	0.397988
C	5.965088	1.336900	2.872009
H	6.138333	0.660326	2.035666
C	5.631028	0.872542	4.080018
H	5.514600	-0.195609	4.261673
C	3.978579	1.335525	6.365274
C	4.024760	-0.012256	6.748192
H	4.896254	-0.617093	6.509961
C	2.974475	-0.581263	7.458651
H	3.021209	-1.628049	7.746553
C	1.869427	0.194354	7.809202
H	1.050499	-0.249047	8.369263
C	1.823455	1.537469	7.450421

H	0.977856	2.156409	7.734515
C	2.872772	2.106369	6.729572
H	2.837390	3.157293	6.461101
C	6.766857	1.686318	6.568565
C	7.781754	0.784773	6.232923
H	7.772706	0.287780	5.266439
C	8.811326	0.510024	7.132749
H	9.590271	-0.196863	6.859257
C	8.835366	1.132548	8.376769
H	9.636488	0.918108	9.078820
C	7.826079	2.031616	8.718773
H	7.837619	2.525863	9.686018
C	6.799921	2.308709	7.822488
H	6.024709	3.015121	8.102707
C	3.572408	3.623554	3.880271
C	8.051590	5.945427	4.500803
H	8.902085	6.260112	5.116929
H	8.435004	5.230891	3.767572
C	6.243546	6.903126	2.945918
H	6.475313	6.188818	2.145636
H	5.938691	7.840892	2.468989
C	5.174501	5.701238	7.787741
C	7.010455	8.699696	7.851145
C	7.176226	9.903889	8.551173
H	6.483644	10.726765	8.393090
C	8.235770	10.068540	9.437467
H	8.351294	11.006724	9.973619
C	9.147550	9.031801	9.635331
H	9.974242	9.160430	10.328795
C	8.994107	7.833873	8.944310
H	9.698806	7.021056	9.096202
C	7.930483	7.666300	8.058522
H	7.802066	6.724629	7.529918
C	5.772771	9.778268	5.474831
C	6.910614	10.586316	5.362182
H	7.738871	10.457142	6.052019
C	6.994349	11.557529	4.366247
H	7.885444	12.175744	4.295095
C	5.945451	11.737021	3.468514
H	6.012004	12.496131	2.693970
C	4.812426	10.930613	3.565947
H	3.990191	11.054486	2.866379
C	4.728880	9.955183	4.554406
H	3.849386	9.319207	4.601900
C	4.200454	9.052315	7.681894
H	4.325727	9.946687	8.291561
C	3.022870	8.421863	7.622112
H	2.158195	8.773592	8.184997
C	1.746472	7.485086	5.242686
C	1.218510	8.781372	5.208644

H	1.474539	9.494210	5.986704
C	0.369800	9.176485	4.176419
H	-0.029320	10.187221	4.166938
C	0.029432	8.279311	3.167735
H	-0.636334	8.586117	2.365685
C	0.551214	6.987902	3.190083
H	0.299549	6.280648	2.404895
C	1.411400	6.596564	4.212227
H	1.820828	5.592338	4.202027
C	1.723521	5.940802	7.653593
C	0.528337	5.382967	7.186510
H	0.231693	5.517637	6.151496
C	-0.307794	4.673697	8.047537
H	-1.237281	4.258229	7.667105
C	0.036247	4.510790	9.387127
H	-0.619855	3.963657	10.058546
C	1.226578	5.058248	9.861023
H	1.507134	4.937789	10.903711
C	2.063236	5.765309	9.002923
H	2.987169	6.183579	9.389874
H	8.253469	7.583843	3.153770
H	4.395562	4.604584	5.761094

Table S15: Cartesian coordinates of the optimized structure of *unsym*-[μ**H**-($Fe^{II}Fe^{II}$)pdt]$^{+}$ (–4772.63153580 E_h)



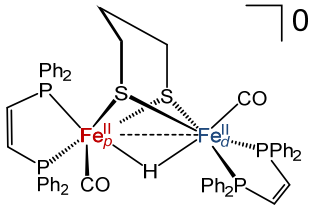
unsym-[μ**H**-($Fe^{II}Fe^{II}$)pdt]$^{+}$

atom	x	y	z
Fe	4.641740	13.836618	3.835663
Fe	5.845623	11.470123	3.739776
H	5.349018	12.645602	4.830471
S	3.745760	11.954920	2.917777
S	6.471207	13.349783	2.582851
P	2.870163	14.250103	5.189734
P	5.161166	9.875929	5.201064
P	5.668364	15.527198	4.927995
P	6.590353	9.749651	2.459318
O	3.508941	15.603739	1.841950
O	8.512313	11.456195	4.897654
C	3.684576	12.216050	1.097043
H	3.114004	11.374553	0.692492
H	3.084237	13.116780	0.936707
C	5.011188	12.302904	0.361352
H	4.791191	12.474627	-0.702477
H	5.501008	11.326695	0.414055
C	5.980691	13.379479	0.815171
H	5.579316	14.379770	0.624028
H	6.913690	13.295859	0.248635
C	7.439650	11.463169	4.469187
C	3.953985	14.930568	2.670856
C	6.436979	16.859762	3.923555
C	5.590185	17.764976	3.268810
H	4.513029	17.694022	3.390620
C	6.117104	18.770210	2.463908
H	5.447979	19.467574	1.968546
C	7.495537	18.881425	2.297709
H	7.907356	19.665927	1.669828
C	8.343184	17.987897	2.946804
H	9.419479	18.073980	2.829942
C	7.819972	16.983292	3.756916
H	8.497987	16.310093	4.269729
C	6.896437	15.168304	6.237471
C	6.709972	15.579112	7.562790
H	5.817458	16.127132	7.848217
C	7.667178	15.286089	8.531846
H	7.513165	15.617476	9.554715
C	8.819381	14.581936	8.189534
H	9.569983	14.364767	8.944018
C	9.007281	14.158618	6.874726
H	9.896225	13.599171	6.600168
C	8.050035	14.445545	5.905514
H	8.201545	14.103061	4.885953
C	1.264241	14.375064	4.328745
C	0.730314	15.616845	3.967379

H	1.243927	16.535232	4.237780
C	-0.461210	15.689829	3.249151
H	-0.865355	16.659918	2.975568
C	-1.129457	14.524425	2.885033
H	-2.059513	14.582180	2.327336
C	-0.601248	13.283108	3.236700
H	-1.118297	12.370949	2.953299
C	0.591533	13.204288	3.948977
H	0.998657	12.231934	4.211081
C	2.475533	13.386644	6.762606
C	3.540495	13.031498	7.598474
H	4.563115	13.199074	7.272317
C	3.298703	12.457345	8.843314
H	4.135615	12.171828	9.472932
C	1.989634	12.234139	9.265977
H	1.800153	11.783072	10.235588
C	0.924299	12.593812	8.443297
H	-0.098238	12.432452	8.772733
C	1.162512	13.171183	7.197770
H	0.325086	13.453482	6.568169
C	8.273624	9.930629	1.763569
C	9.386465	9.464382	2.472427
H	9.256986	8.949739	3.420174
C	10.673063	9.662684	1.976232
H	11.526703	9.293162	2.536541
C	10.862663	10.328743	0.769259
H	11.865740	10.480654	0.382141
C	9.760285	10.797758	0.056538
H	9.901047	11.313510	-0.888786
C	8.474819	10.604025	0.550939
H	7.625074	10.962180	-0.022872
C	5.631229	8.916708	1.125289
C	6.265731	8.219040	0.089509
H	7.347865	8.226400	0.006889
C	5.511469	7.514603	-0.845753
H	6.014416	6.982368	-1.647790
C	4.122262	7.489359	-0.752480
H	3.537511	6.939921	-1.484320
C	3.485568	8.168865	0.284126
H	2.402625	8.149389	0.366484
C	4.234054	8.879275	1.218379
H	3.730626	9.408171	2.023167
C	3.440475	9.239165	5.213862
C	2.465619	9.877102	5.990404
H	2.733684	10.718120	6.621356
C	1.150480	9.421405	5.977580
H	0.408192	9.922755	6.591500
C	0.793089	8.325489	5.194172
H	-0.232372	7.967583	5.191959
C	1.758976	7.682661	4.424766

H	1.492688	6.818514	3.822957
C	5.566471	10.023402	6.986480
C	5.160846	9.015616	7.873600
H	4.532767	8.201235	7.522395
C	5.546451	9.054687	9.209197
H	5.227393	8.266641	9.885022
C	6.343494	10.099405	9.676821
H	6.651380	10.123774	10.718240
C	6.741363	11.110157	8.805806
H	7.355572	11.933071	9.159280
C	6.352030	11.074690	7.467651
H	6.659621	11.877005	6.806173
C	6.106632	8.370471	4.765356
H	6.108032	7.526836	5.454580
C	6.758939	8.331203	3.600728
H	7.310618	7.449710	3.275475
C	4.363049	16.456501	5.804351
H	4.594730	17.426208	6.243537
C	3.138345	15.925770	5.874207
H	2.325578	16.423463	6.402574
C	3.076307	8.136187	4.432646
H	3.820096	7.620234	3.832613

Table S16: Cartesian coordinates of the optimized structure of *unsym*-[μ**H**-($Fe^{I}Fe^{II}$)pdt]^{0} (–4772.78280972 E_h)



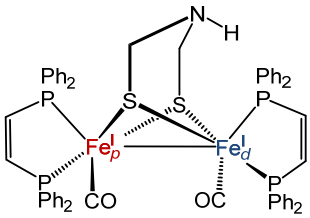
unsym-[μ**H**-($Fe^{I}Fe^{II}$)pdt]^{0}

atom	x	y	z
Fe	10.524401	0.209814	4.818341
H	10.727185	1.682356	4.249856
Fe	10.630234	2.854264	5.879598
S	12.002715	1.038103	6.399941
S	8.924852	1.265963	6.107792
P	12.094782	-0.616993	3.491528
P	9.118249	-0.126371	3.151194
P	10.589949	4.197188	7.841009
P	12.301206	4.207743	5.220320
O	10.219953	-2.414769	6.061735
O	8.636864	4.648735	4.757691
C	13.191683	-1.892963	4.214924
C	12.982040	-3.256802	3.986696
H	12.180090	-3.583333	3.330508
C	13.784971	-4.209863	4.609952
H	13.608540	-5.265754	4.424587
C	14.803907	-3.810304	5.469781
H	15.429638	-4.553603	5.956045
C	15.012877	-2.453333	5.711512
H	15.799544	-2.134989	6.389973
C	14.210693	-1.498464	5.094246
H	14.364581	-0.444094	5.305906
C	13.246771	0.374642	2.447325
C	12.775605	1.578818	1.912176
H	11.791216	1.940194	2.196479
C	13.570107	2.327704	1.047919
H	13.196074	3.271172	0.661557
C	14.845525	1.880682	0.709634
H	15.470105	2.469369	0.043389
C	15.321372	0.681006	1.234342
H	16.314819	0.326328	0.972649
C	14.526437	-0.072122	2.096009
H	14.905525	-1.006379	2.498385
C	11.237285	-1.500950	2.130578
H	11.801974	-2.136343	1.448245
C	9.937751	-1.244754	1.950451
H	9.373664	-1.677624	1.124492
C	7.496280	-0.975949	3.364545
C	6.628779	-1.123673	2.273677
H	6.887650	-0.686775	1.312342
C	5.426250	-1.808191	2.413768
H	4.763204	-1.915313	1.559606
C	5.070561	-2.347632	3.649290
H	4.129260	-2.878896	3.760476
C	5.920979	-2.197120	4.740456
H	5.646301	-2.607503	5.708104

C	7.127680	-1.514110	4.599468
H	7.780902	-1.388576	5.454507
C	10.329067	-1.372431	5.558265
C	11.532035	0.237360	7.987080
H	11.576633	-0.845483	7.826592
H	12.321495	0.495169	8.701078
C	10.192341	0.640683	8.587296
H	10.064187	0.078732	9.525466
H	10.236470	1.695675	8.868261
C	8.954482	0.411598	7.734724
H	8.076326	0.770191	8.284356
H	8.805211	-0.658730	7.550895
C	9.458163	3.935205	5.177885
C	11.532677	3.838640	9.392743
C	10.986367	3.909107	10.679886
H	9.959822	4.233722	10.817540
C	11.748027	3.560685	11.794055
H	11.305563	3.621951	12.784941
C	13.065790	3.138475	11.642300
H	13.658228	2.869962	12.512674
C	13.615574	3.051886	10.364158
H	14.639087	2.712203	10.229735
C	12.853750	3.388039	9.249567
H	13.285182	3.283152	8.257899
C	8.992089	4.858880	8.478828
C	7.812631	4.195845	8.123274
H	7.867767	3.317022	7.485882
C	6.575149	4.665230	8.560797
H	5.668908	4.139906	8.272321
C	6.500215	5.806323	9.353494
H	5.535286	6.175343	9.690460
C	7.668172	6.478652	9.711753
H	7.616528	7.370876	10.330066
C	8.903678	6.007910	9.278354
H	9.805578	6.534267	9.579971
C	11.417804	5.752209	7.318328
H	11.314736	6.657782	7.915654
C	12.144345	5.754467	6.196142
H	12.650237	6.656388	5.850081
C	14.084885	3.816587	5.502488
C	14.848188	4.458386	6.484657
H	14.398620	5.227964	7.105274
C	16.186544	4.118037	6.681666
H	16.762872	4.630474	7.447500
C	16.781521	3.134548	5.897725
H	17.825192	2.872082	6.048521
C	16.030124	2.490870	4.915465
H	16.481518	1.723840	4.292293
C	14.694006	2.825753	4.722395
H	14.123187	2.318526	3.952442

C	12.371521	4.922335	3.515967
C	13.440876	5.736407	3.118649
H	14.276639	5.898351	3.794644
C	13.451430	6.330176	1.860428
H	14.287309	6.960110	1.567809
C	12.392012	6.117351	0.978547
H	12.397615	6.585123	-0.002489
C	11.329888	5.301325	1.359573
H	10.501762	5.119623	0.680633
C	11.321080	4.704283	2.619941
H	10.492644	4.063577	2.903885
C	8.619499	1.290047	2.092288
C	7.725012	2.230720	2.619370
H	7.339140	2.103778	3.627560
C	7.338288	3.332733	1.864254
H	6.656076	4.061225	2.291539
C	7.832773	3.506277	0.571524
H	7.521077	4.362918	-0.020003
C	8.727143	2.578981	0.042996
H	9.117213	2.706733	-0.963381
C	9.123180	1.477415	0.801147
H	9.829116	0.765304	0.382530

Table S17: Cartesian coordinates of the optimized structure of $[(Fe^{I}Fe^{I})adt]^{0}$ (–4788.20514698 E_h)



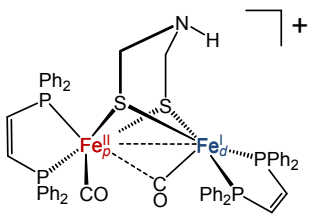
$[(Fe^{I}Fe^{I})adt]^{0}$

atom	x	y	z
Fe	5.236157	4.184566	4.510930
Fe	4.970985	6.243511	5.961275
S	6.979952	5.208321	5.558059
S	4.837397	6.275308	3.682349
P	5.940559	3.139686	2.716665
P	5.586922	2.277559	5.643984
P	5.566610	8.186115	6.791459
P	2.843321	6.916784	6.206264
O	2.435328	3.537600	4.150199
O	4.827369	4.805194	8.469565
N	7.469047	7.311144	3.704995
H	7.206778	8.063847	4.334402
C	4.928686	3.142296	1.174093
C	3.899498	4.073745	1.010134
H	3.721218	4.811688	1.786512
C	3.121400	4.070762	-0.147013
H	2.328157	4.804395	-0.260987
C	3.356813	3.135334	-1.149305
H	2.745994	3.130947	-2.047997
C	4.384481	2.204844	-0.998916
H	4.579453	1.474950	-1.779949
C	5.166095	2.211773	0.151337
H	5.979569	1.496783	0.241537
C	7.642793	3.383144	2.035907
C	8.738530	3.056045	2.846215
H	8.571763	2.653491	3.841409
C	10.039548	3.271766	2.403213
H	10.876106	3.008162	3.044616
C	10.268241	3.836423	1.148982
H	11.283900	4.011952	0.805617
C	9.187045	4.181625	0.343624
H	9.354476	4.629682	-0.632074
C	7.882929	3.953808	0.780870
H	7.049777	4.218317	0.136785
C	5.972929	1.343403	3.099059
H	6.122928	0.609191	2.308560
C	5.795388	0.970471	4.369270
H	5.791246	-0.080766	4.656578
C	4.326422	1.495624	6.739645
C	4.520590	0.193025	7.221955
H	5.431489	-0.348002	6.977795
C	3.568653	-0.409135	8.035887
H	3.731183	-1.419559	8.401478
C	2.412148	0.287327	8.387149
H	1.668042	-0.182245	9.025107
C	2.219250	1.585780	7.927868

H	1.330542	2.144035	8.205902
C	3.173124	2.189173	7.108866
H	3.022643	3.206653	6.765885
C	7.104327	1.990414	6.658231
C	8.207495	1.289491	6.160343
H	8.179864	0.874191	5.156766
C	9.343855	1.099246	6.945804
H	10.188738	0.546023	6.544016
C	9.390104	1.605622	8.240724
H	10.273696	1.455473	8.855131
C	8.291996	2.298801	8.747598
H	8.314856	2.691203	9.760718
C	7.157831	2.490771	7.965318
H	6.308603	3.027869	8.375299
C	3.566627	3.787161	4.306413
C	8.057306	6.226511	4.426392
H	8.855146	6.621952	5.062069
C	6.364507	6.970035	2.866262
H	6.027149	7.872495	2.349052
C	4.879272	5.383641	7.455554
C	6.778156	8.282621	8.177585
C	6.911753	9.444529	8.952993
H	6.327951	10.330164	8.715658
C	7.805478	9.489015	10.017832
H	7.895562	10.395841	10.609764
C	8.584229	8.372788	10.321828
H	9.282256	8.407086	11.153924
C	8.464857	7.218215	9.554956
H	9.070230	6.345357	9.782595
C	7.565376	7.170233	8.490439
H	7.475218	6.267859	7.892139
C	6.079038	9.612629	5.724105
C	7.321486	10.250024	5.824669
H	8.026016	9.947568	6.593399
C	7.666241	11.273093	4.941656
H	8.637248	11.752595	5.030688
C	6.774873	11.677652	3.953014
H	7.045060	12.475326	3.266778
C	5.536277	11.045902	3.841380
H	4.835929	11.348845	3.067807
C	5.194241	10.017554	4.712634
H	4.238411	9.512356	4.597206
C	4.083056	8.928722	7.582355
H	4.175217	9.800952	8.228129
C	2.896793	8.373361	7.324761
H	1.973946	8.756148	7.760561
C	1.858984	7.591105	4.797815
C	1.465281	8.934600	4.760784
H	1.731134	9.603581	5.573396
C	0.735002	9.436468	3.685068

H	0.439044	10.482094	3.679092
C	0.380911	8.601601	2.629726
H	-0.193427	8.990272	1.793277
C	0.772238	7.265211	2.652053
H	0.506554	6.604653	1.831304
C	1.514789	6.765492	3.718260
H	1.824172	5.726528	3.705767
C	1.588614	5.919624	7.132287
C	0.498629	5.291828	6.520457
H	0.348892	5.375921	5.450103
C	-0.419128	4.568404	7.278914
H	-1.262182	4.092923	6.785330
C	-0.264421	4.462612	8.659044
H	-0.988146	3.907500	9.249842
C	0.823735	5.075199	9.277109
H	0.959022	4.996296	10.352225
C	1.743896	5.794349	8.520204
H	2.587085	6.264697	9.017411
H	8.500741	5.517857	3.720602
H	6.688742	6.240409	2.117129

Table S18: Cartesian coordinates of the optimized structure of $[(Fe^{II}Fe^{I})adt]^{+}$ (–4788.03034271 E_h)



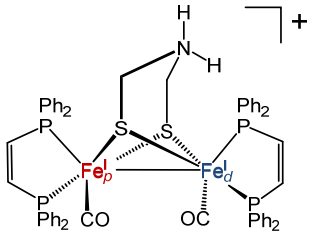
$[(Fe^{II}Fe^{I})adt]^{+}$

atom	x	y	z
Fe	12.304486	3.493007	4.261011
Fe	13.942596	5.491778	4.482835
S	13.180310	4.664203	2.484524
S	14.487546	3.341454	4.973996
P	10.352667	2.963697	3.215825
P	11.854826	1.772796	5.631919
P	15.929647	6.324848	3.924779
P	13.094637	7.588803	4.087441
N	14.688840	2.256841	2.378353
O	11.006983	5.464045	5.992057
O	14.302568	6.108794	7.306645
C	10.510164	2.155590	1.575696
C	10.153130	0.816086	1.377822
H	9.749628	0.229693	2.198336
C	10.302139	0.221074	0.126263
H	10.019563	-0.818568	-0.011055
C	10.799720	0.958321	-0.944175
H	10.911406	0.495214	-1.919986
C	11.149262	2.295368	-0.759840
H	11.530929	2.878496	-1.593016
C	11.012293	2.891088	0.490862
H	11.293949	3.932009	0.627138
C	8.952418	4.138123	2.998934
C	8.458381	4.786255	4.140708
H	8.935186	4.640891	5.103983
C	7.347122	5.619476	4.050734
H	6.973919	6.111313	4.944116
C	6.712949	5.813600	2.825388
H	5.842948	6.460041	2.757638
C	7.192489	5.165845	1.689740
H	6.694179	5.299487	0.733703
C	8.305696	4.331013	1.772836
H	8.656903	3.821090	0.882030
C	9.520753	1.688826	4.230815
H	8.506258	1.386708	3.972330
C	10.159158	1.184225	5.290903
H	9.695677	0.437688	5.934004
C	11.907451	1.885589	7.453901
C	11.668096	4.803243	5.291774
C	14.330270	3.435204	1.659666
H	13.809645	3.146222	0.744243
H	15.232112	3.988533	1.389466
C	15.394332	2.449531	3.597554
H	16.314085	3.010645	3.407625
H	15.651702	1.478788	4.027271
C	14.163691	5.895036	6.180042

C	16.599623	6.009761	2.244781
C	17.707538	5.179307	2.040066
H	18.229881	4.747317	2.888572
C	18.157159	4.914532	0.747676
H	19.017945	4.268999	0.600515
C	17.509768	5.479269	-0.348032
H	17.864556	5.275795	-1.354017
C	16.406842	6.309445	-0.150352
H	15.899680	6.754817	-1.001204
C	15.949948	6.571209	1.137455
H	15.078358	7.204377	1.278715
C	15.769074	8.145730	3.904691
H	16.662279	8.763628	3.829432
C	14.549248	8.685013	3.962734
H	14.400718	9.764179	3.961764
C	12.138433	8.469753	5.386671
H	13.900114	1.626886	2.501745
C	12.877486	0.292582	5.257696
C	12.654388	-0.424817	4.075244
H	11.825699	-0.159575	3.423795
C	13.484887	-1.487521	3.727974
H	13.297000	-2.041258	2.812716
C	14.549090	-1.841295	4.555016
H	15.195225	-2.671454	4.285698
C	14.778002	-1.130183	5.731384
H	15.603098	-1.404956	6.382107
C	13.948944	-0.067423	6.083617
H	14.133597	0.480401	7.002394
C	12.366419	3.043235	8.086447
C	12.387712	3.122214	9.477433
H	12.746667	4.027938	9.956564
C	11.949958	2.046973	10.244468
H	11.963417	2.110480	11.328553
C	11.497522	0.884151	9.620938
H	11.161598	0.040362	10.216337
C	11.478759	0.800863	8.233502
H	11.145801	-0.119178	7.760061
C	17.387220	6.076531	5.008498
C	17.360634	5.161285	6.065031
H	16.475888	4.557638	6.237779
C	18.473426	5.007146	6.890647
H	18.439058	4.292988	7.707978
C	19.619341	5.764185	6.670466
H	20.483798	5.645725	7.317061
C	19.658810	6.673451	5.613440
C	18.552847	6.827170	4.786017
C	12.174851	7.933418	2.543835
C	12.611807	8.894279	1.624472
H	13.520571	9.459040	1.808352
C	11.884745	9.141185	0.462199

H	12.234057	9.892452	-0.240132
C	10.711006	8.436982	0.209368
H	10.140053	8.636816	-0.692698
C	10.271389	7.475465	1.116647
H	9.358111	6.919067	0.930345
C	11.003517	7.215367	2.271568
H	10.655849	6.450409	2.960318
C	12.820694	8.915687	6.526564
H	13.890379	8.753846	6.625502
C	12.139551	9.582220	7.540350
H	12.682669	9.927248	8.415202
C	10.769967	9.808822	7.429514
H	10.238349	10.330570	8.219771
C	10.086833	9.370977	6.298107
H	9.020476	9.552770	6.201097
C	10.764464	8.705714	5.280290
H	10.219059	8.384372	4.399324
H	12.711026	3.884032	7.497428
H	20.553377	7.261798	5.432046
H	18.608383	7.522253	3.952505

Table S19: Cartesian coordinates of the optimized structure of $[(Fe^{I}Fe^{I})adt\text{-}\mathbf{H}]^{+}$ (–4788.62478532 E_h)



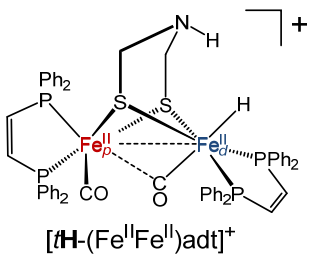
$[(Fe^{I}Fe^{I})adt\text{-}\mathbf{H}]^{+}$

atom	x	y	z
Fe	5.211985	4.185860	4.488292
Fe	4.956720	6.266998	5.932230
S	6.962320	5.217254	5.509899
S	4.749694	6.245290	3.648955
P	5.916842	3.122792	2.686674
P	5.604891	2.279164	5.650208
P	5.566787	8.175269	6.850385
P	2.820090	6.966534	6.192595
O	2.423754	3.460992	4.161587
O	4.730675	4.744262	8.384249
N	7.274808	7.347556	3.733938
H	6.827555	7.930986	4.462828
C	4.877371	3.045915	1.172832
C	3.782702	3.900963	1.014811
H	3.552647	4.627788	1.786452
C	2.992695	3.831341	-0.131849
H	2.145160	4.501529	-0.241906
C	3.283950	2.905564	-1.128425
H	2.663602	2.847713	-2.018016
C	4.377395	2.052116	-0.983804
H	4.613112	1.330226	-1.760236
C	5.170522	2.124365	0.155660
H	6.031666	1.467228	0.241688
C	7.574949	3.524391	1.979019
C	8.723827	3.256728	2.736649
H	8.630378	2.781646	3.709070
C	9.985265	3.608618	2.263520
H	10.865833	3.379211	2.857081
C	10.118618	4.249946	1.031755
H	11.102893	4.520822	0.661098
C	8.983449	4.529786	0.275500
H	9.078495	5.018377	-0.690246
C	7.719958	4.165678	0.741812
H	6.845807	4.362404	0.127492
C	6.086741	1.351498	3.110727
H	6.294170	0.616545	2.334708
C	5.917903	0.988546	4.385012
H	5.983789	-0.056050	4.686589
C	4.335700	1.475636	6.706824
C	4.545691	0.168372	7.171435
H	5.465691	-0.358111	6.931222
C	3.593392	-0.458884	7.965136
H	3.765965	-1.472227	8.315899
C	2.423380	0.215470	8.314600
H	1.679791	-0.275128	8.936147
C	2.216215	1.518325	7.874493

H	1.316624	2.058514	8.152810
C	3.168518	2.147894	7.074538
H	3.001886	3.168380	6.750211
C	7.107311	2.087712	6.701642
C	8.296642	1.557476	6.189869
H	8.339263	1.207609	5.162312
C	9.425378	1.436242	6.998143
H	10.336798	1.008962	6.589336
C	9.378314	1.842560	8.328512
H	10.255361	1.741268	8.961212
C	8.193966	2.360914	8.849231
H	8.142013	2.661173	9.892036
C	7.064558	2.481538	8.044884
H	6.144428	2.870052	8.468457
C	3.544505	3.738339	4.305040
C	8.001997	6.231606	4.417291
H	8.783309	6.706748	5.016924
C	6.196204	6.934184	2.786646
H	5.892878	7.842256	2.259414
C	4.817739	5.344908	7.390977
C	6.727797	8.212175	8.271785
C	6.911642	9.386731	9.018157
H	6.393820	10.301079	8.739740
C	7.770936	9.401765	10.111078
H	7.901138	10.315906	10.682893
C	8.462783	8.245076	10.470606
H	9.132427	8.257215	11.325610
C	8.293170	7.078283	9.732087
H	8.830343	6.175065	10.005840
C	7.428155	7.058587	8.638727
H	7.299554	6.145114	8.067503
C	6.239910	9.522276	5.767057
C	7.572982	9.954042	5.834938
H	8.217646	9.593177	6.631373
C	8.075195	10.854539	4.894055
H	9.107920	11.184636	4.965098
C	7.252843	11.344943	3.881867
H	7.642461	12.054117	3.157465
C	5.921306	10.930673	3.811341
H	5.270019	11.319202	3.033471
C	5.420740	10.022110	4.739257
H	4.388992	9.686445	4.660454
C	4.083076	8.987165	7.547193
H	4.181977	9.868499	8.178779
C	2.892698	8.452634	7.266058
H	1.967664	8.865222	7.667790
C	1.855096	7.602251	4.759748
C	1.518751	8.958498	4.661105
H	1.813017	9.654788	5.440409
C	0.796055	9.436001	3.569413

H	0.538735	10.490039	3.516414
C	0.393781	8.565193	2.561408
H	-0.178392	8.935134	1.715685
C	0.728165	7.215687	2.645636
H	0.418393	6.527239	1.864653
C	1.461729	6.738305	3.727867
H	1.722853	5.686111	3.763433
C	1.592454	5.980963	7.154327
C	0.499722	5.332761	6.569118
H	0.325513	5.400743	5.501633
C	-0.396297	4.612177	7.355747
H	-1.243780	4.121747	6.885829
C	-0.216373	4.532536	8.734490
H	-0.924205	3.980988	9.346632
C	0.870761	5.172571	9.325836
H	1.019442	5.119541	10.400324
C	1.769985	5.889077	8.542369
H	2.609725	6.384439	9.021108
H	8.461316	5.616261	3.639673
H	6.633390	6.223886	2.080888
H	7.950799	7.950864	3.253548

Table S20: Cartesian coordinates of the optimized structure of [*t***H**-($Fe^{II}Fe^{II}$)adt]$^{+}$ (–4788.64475156 E_h)



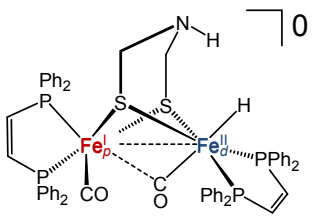
[*t***H**-($Fe^{II}Fe^{II}$)adt]$^{+}$

atom	x	y	z
Fe	12.395358	3.324029	4.129008
Fe	14.026934	5.341232	4.384241
S	13.223283	4.580207	2.396097
S	14.585914	3.186047	4.749359
P	10.456384	2.921823	3.099933
P	11.879247	1.716225	5.567098
P	15.972774	6.246286	3.802564
P	13.114378	7.435744	4.103601
N	14.818044	2.244853	2.090580
O	11.217225	5.242587	5.981300
O	14.508176	5.833248	7.219675
C	10.627899	2.422253	1.347441
C	10.633625	1.068985	0.991542
H	10.480064	0.304705	1.748761
C	10.835735	0.686850	-0.333176
H	10.832992	-0.367088	-0.595459
C	11.033858	1.652588	-1.316143
H	11.188302	1.355061	-2.349060
C	11.030539	3.003052	-0.970051
H	11.182899	3.761405	-1.732611
C	10.833083	3.389210	0.353261
H	10.844125	4.444174	0.611206
C	8.945619	3.982695	3.118001
C	8.547751	4.544658	4.339742
H	9.153659	4.408838	5.228710
C	7.367848	5.277695	4.430095
H	7.074893	5.704494	5.384813
C	6.565448	5.456520	3.305431
H	5.644078	6.026964	3.377013
C	6.944380	4.889678	2.091209
H	6.316297	5.009888	1.213038
C	8.124540	4.154215	1.995778
H	8.395936	3.706277	1.045820
C	9.741467	1.441248	3.901775
H	8.819723	1.021058	3.500547
C	10.322656	0.952123	5.000458
H	9.919194	0.087272	5.524984
C	11.584909	2.128926	7.322805
C	11.821125	4.603338	5.217120
C	14.381885	3.446127	1.458395
H	13.830040	3.189893	0.551203
H	15.244844	4.055835	1.184320
C	15.513624	2.397653	3.318770
H	16.411601	3.003570	3.167961
H	15.805729	1.415374	3.696815
C	14.315168	5.671705	6.093813

C	16.611866	6.009149	2.097991
C	17.731956	5.210444	1.840644
H	18.277330	4.753925	2.661335
C	18.164221	5.009280	0.531012
H	19.033911	4.386937	0.342535
C	17.488109	5.607463	-0.528918
H	17.829261	5.453236	-1.548238
C	16.373736	6.407608	-0.278477
H	15.844480	6.878819	-1.101451
C	15.933845	6.605489	1.026413
H	15.053619	7.215711	1.208875
C	15.765498	8.061772	3.835152
H	16.639332	8.704521	3.743642
C	14.534354	8.568662	3.933127
H	14.358978	9.643581	3.953180
C	12.221094	8.259298	5.483590
H	12.566797	2.059480	3.322626
H	14.035761	1.605192	2.208999
C	12.996534	0.257445	5.617426
C	13.178955	-0.483772	4.441846
H	12.659985	-0.191503	3.532178
C	14.010016	-1.599040	4.431531
H	14.136239	-2.169564	3.515830
C	14.675114	-1.984518	5.594906
H	15.325798	-2.853954	5.587363
C	14.495457	-1.255864	6.766714
H	15.004580	-1.554719	7.678315
C	13.656552	-0.142745	6.782455
H	13.516466	0.408801	7.706225
C	12.583263	2.828004	8.015440
C	12.399395	3.182304	9.347823
H	13.181009	3.723120	9.872819
C	11.211701	2.855421	9.999965
H	11.065564	3.137974	11.038275
C	10.210525	2.171766	9.315979
H	9.281307	1.918940	9.818073
C	10.394949	1.806766	7.984080
H	9.601187	1.277696	7.466152
C	17.460472	5.987981	4.840888
C	17.495309	4.994085	5.823677
H	16.637221	4.346272	5.971047
C	18.634477	4.819529	6.608042
H	18.648340	4.043516	7.367503
C	19.745630	5.634920	6.420174
H	20.630666	5.500628	7.034994
C	19.724136	6.622617	5.435558
C	18.592250	6.796107	4.648050
C	12.093240	7.794421	2.629625
C	12.458075	8.770260	1.695511
H	13.371900	9.342183	1.825212

C	11.650819	9.023770	0.588501
H	11.944488	9.786763	-0.126492
C	10.468293	8.312232	0.407392
H	9.834640	8.519096	-0.450070
C	10.099352	7.335384	1.330461
H	9.177663	6.775404	1.202250
C	10.912412	7.068243	2.428288
H	10.618377	6.293154	3.130340
C	12.959716	8.654395	6.607118
H	14.032138	8.486178	6.647270
C	12.331816	9.279461	7.679806
H	12.918263	9.584781	8.541307
C	10.959762	9.515551	7.644799
H	10.469587	10.004868	8.481224
C	10.220934	9.129531	6.529762
H	9.152304	9.319534	6.491401
C	10.845241	8.505940	5.453024
H	10.256079	8.225882	4.586408
H	13.507663	3.096102	7.509877
H	20.591802	7.256465	5.278643
H	18.602437	7.5525	3.868096

Table S21: Cartesian coordinates of the optimized structure of [*t***H**-($Fe^{I}Fe^{II}$)adt]^{0} (–4788.81184262 E_h)



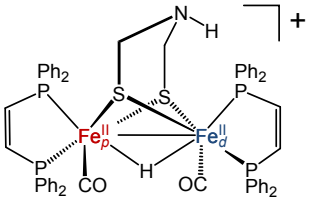
[*t***H**-($Fe^{I}Fe^{II}$)adt]^{0}

atom	x	y	z
Fe	12.188181	3.224418	4.213202
Fe	14.098443	5.494543	4.470433
S	13.136526	4.574367	2.559556
S	14.382627	3.268375	5.011610
P	10.359227	2.779077	3.087057
P	11.692022	1.584578	5.567118
P	16.111773	6.331851	3.732083
P	13.270434	7.553183	4.133733
N	14.808334	2.268444	2.390841
O	10.994506	5.298982	5.899640
O	14.552956	6.009184	7.295856
C	10.576427	2.318847	1.325443
C	10.525636	0.982927	0.913375
H	10.304538	0.200150	1.633880
C	10.766835	0.639627	-0.415259
H	10.723593	-0.402996	-0.718162
C	11.062263	1.628619	-1.349416
H	11.251564	1.361792	-2.385434
C	11.118390	2.962136	-0.948164
H	11.355200	3.739488	-1.669299
C	10.881191	3.308414	0.379710
H	10.952693	4.347894	0.685475
C	8.830990	3.825328	3.058409
C	8.370540	4.333889	4.281071
H	8.928402	4.144957	5.192234
C	7.201572	5.086631	4.342943
H	6.863829	5.473364	5.300282
C	6.472605	5.345385	3.184218
H	5.562123	5.936337	3.231632
C	6.915804	4.838348	1.965494
H	6.348878	5.026026	1.057433
C	8.084178	4.080258	1.901892
H	8.409942	3.683189	0.946077
C	9.621565	1.270667	3.828065
H	8.721193	0.840065	3.389850
C	10.175123	0.770506	4.937297
H	9.764352	-0.112370	5.425153
C	11.333807	1.886876	7.344136
C	11.568982	4.547309	5.220777
C	14.372510	3.450465	1.711051
H	13.896481	3.152003	0.773364
H	15.235966	4.078674	1.481759
C	15.413905	2.462024	3.668358
H	16.306059	3.087899	3.562692
H	15.708189	1.488515	4.069346
C	14.370135	5.845829	6.155859

C	16.725487	6.116039	2.003850
C	17.835630	5.318594	1.698908
H	18.419900	4.874814	2.499789
C	18.202091	5.090211	0.374385
H	19.065744	4.467616	0.156731
C	17.467518	5.655155	-0.665111
H	17.754798	5.476545	-1.697555
C	16.359055	6.447876	-0.372265
H	15.776457	6.889901	-1.176085
C	15.987832	6.673655	0.949613
H	15.110001	7.277589	1.163498
C	15.920526	8.160600	3.735655
H	16.790709	8.803482	3.602843
C	14.696206	8.680215	3.871295
H	14.534767	9.758473	3.874889
C	12.427880	8.478730	5.493551
H	12.515641	1.981972	3.379268
H	14.012361	1.641656	2.501719
C	12.856664	0.160030	5.633973
C	12.994231	-0.655504	4.503827
H	12.398419	-0.450867	3.617810
C	13.890320	-1.719803	4.504229
H	13.983293	-2.346889	3.621698
C	14.669848	-1.975877	5.631324
H	15.373765	-2.803509	5.630778
C	14.543189	-1.165886	6.756260
H	15.149857	-1.356709	7.637199
C	13.638733	-0.105509	6.761178
H	13.545992	0.518607	7.644335
C	12.063294	2.884858	8.002480
C	11.845699	3.144492	9.353362
H	12.420093	3.923251	9.846380
C	10.889706	2.419206	10.059764
H	10.713294	2.628458	11.111258
C	10.157176	1.425721	9.413493
H	9.409073	0.856675	9.958755
C	10.381743	1.156526	8.066233
H	9.806809	0.372571	7.583029
C	17.658453	6.119953	4.709420
C	17.668873	5.172929	5.738543
H	16.776321	4.582138	5.927984
C	18.811052	4.987191	6.516793
H	18.804340	4.248246	7.313148
C	19.950994	5.748075	6.276992
H	20.839666	5.606605	6.886175
C	19.952788	6.692453	5.250083
C	18.816020	6.874091	4.469694
C	12.190985	7.910122	2.688899
C	12.510004	8.871484	1.724201
H	13.424311	9.450824	1.813506

C	11.664956	9.096525	0.638835
H	11.928923	9.847278	-0.101302
C	10.484682	8.370098	0.508696
H	9.822460	8.551686	-0.333534
C	10.158030	7.406673	1.461377
H	9.242041	6.829282	1.370392
C	11.011504	7.168458	2.535212
H	10.756164	6.399186	3.258512
C	13.200816	8.900881	6.583540
H	14.267814	8.698636	6.603880
C	12.615987	9.578657	7.648793
H	13.232705	9.899156	8.483988
C	11.248120	9.840805	7.645058
H	10.790209	10.368259	8.477381
C	10.471914	9.423517	6.567464
H	9.404183	9.625139	6.553413
C	11.056158	8.749072	5.498268
H	10.437304	8.442224	4.661584
H	12.806067	3.460472	7.457885
H	20.842761	7.285123	5.055835
H	18.836597	7.596123	3.657172

Table S22: Cartesian coordinates of the optimized structure of *sym*-[μ**H**-($Fe^{II}Fe^{II}$)adt]$^+$ (–4788.66309126 E_h)



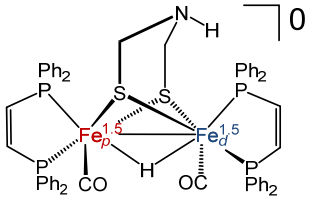
sym-[μ**H**-($Fe^{II}Fe^{II}$)adt]$^+$

atom	x	y	z
Fe	5.135294	4.124930	4.498327
Fe	4.961012	6.181140	6.102054
S	6.920521	5.075616	5.599829
S	4.771795	6.283035	3.804877
P	6.068021	3.178300	2.683183
P	5.419656	2.122881	5.535578
P	5.618717	8.254873	6.698270
P	2.832075	6.916961	6.482901
O	2.504062	3.285974	3.593892
O	5.223360	5.243522	8.838800
N	7.452801	7.209473	3.796523
H	7.306259	8.012227	4.397296
C	5.236352	3.341287	1.056569
C	4.252419	4.312071	0.846022
H	3.982221	4.992095	1.648349
C	3.624451	4.419875	-0.393942
H	2.863726	5.179955	-0.545434
C	3.968459	3.558924	-1.430887
H	3.474690	3.640925	-2.394685
C	4.953790	2.591925	-1.232601
H	5.231466	1.921407	-2.040430
C	5.587056	2.486134	0.000102
H	6.370951	1.744894	0.129420
C	7.839756	3.420877	2.257109
C	8.822714	2.934233	3.128795
H	8.536417	2.409534	4.035123
C	10.171047	3.141858	2.855804
H	10.923101	2.755493	3.537467
C	10.554141	3.846926	1.715651
H	11.606676	4.010364	1.504273
C	9.582941	4.341123	0.849251
H	9.874135	4.891966	-0.040183
C	8.231573	4.127972	1.114501
H	7.484721	4.505011	0.422473
C	6.000344	1.371701	2.971810
H	6.199818	0.689404	2.147192
C	5.711264	0.917377	4.194399
H	5.659066	-0.150390	4.403007
C	4.036918	1.368200	6.472985
C	4.138130	0.034635	6.896351
H	5.038523	-0.538377	6.690464
C	3.101114	-0.559169	7.604914
H	3.188595	-1.593506	7.924135
C	1.955888	0.175686	7.911621
H	1.147042	-0.288334	8.468488
C	1.855626	1.504134	7.512096

H	0.976132	2.089468	7.760652
C	2.891453	2.099943	6.794074
H	2.805910	3.140024	6.496872
C	6.830656	1.799277	6.664539
C	7.944210	1.051123	6.269010
H	7.991450	0.623719	5.271214
C	8.994010	0.825126	7.157973
H	9.847817	0.232613	6.841929
C	8.940797	1.343917	8.448071
H	9.757039	1.164133	9.141524
C	7.829927	2.083517	8.851348
H	7.775741	2.482074	9.860168
C	6.780140	2.308693	7.967655
H	5.914412	2.871974	8.301486
C	3.558913	3.608477	3.941553
C	8.018690	6.103089	4.487790
H	8.821029	6.463969	5.136304
C	6.315406	6.959818	2.983559
H	5.998393	7.897226	2.521853
C	5.107633	5.642469	7.760403
C	6.999918	8.455937	7.885961
C	7.181202	9.668086	8.571303
H	6.525839	10.513214	8.379005
C	8.215435	9.811786	9.489126
H	8.343109	10.754048	10.013666
C	9.085450	8.749455	9.733390
H	9.892162	8.862926	10.451530
C	8.917159	7.546413	9.055508
H	9.591186	6.715499	9.241245
C	7.878932	7.397257	8.137366
H	7.750280	6.451475	7.618542
C	5.909552	9.580402	5.449150
C	7.104590	10.307406	5.378075
H	7.905147	10.115117	6.085628
C	7.275631	11.284167	4.397905
H	8.208443	11.838247	4.351914
C	6.257395	11.550233	3.487454
H	6.391792	12.314699	2.728034
C	5.064583	10.830528	3.552419
H	4.265269	11.032095	2.845274
C	4.891952	9.847787	4.521519
H	3.963258	9.284475	4.549928
C	4.211380	8.981364	7.618397
H	4.362937	9.883620	8.208285
C	3.009135	8.409167	7.519063
H	2.138830	8.806871	8.039563
C	1.766254	7.497190	5.107736
C	1.277782	8.808956	5.065250
H	1.535618	9.517499	5.846274
C	0.456681	9.223999	4.018992

H	0.083522	10.243838	4.002708
C	0.107823	8.333927	3.007158
H	-0.538495	8.656637	2.196311
C	0.591358	7.027883	3.039183
H	0.325722	6.326464	2.253826
C	1.422919	6.614052	4.075519
H	1.797791	5.596679	4.076506
C	1.684304	5.924430	7.526080
C	0.526227	5.326668	7.017028
H	0.269651	5.430990	5.968203
C	-0.330816	4.621876	7.860356
H	-1.234015	4.176978	7.452595
C	-0.043141	4.505444	9.218107
H	-0.718658	3.965445	9.875078
C	1.109477	5.095414	9.732258
H	1.339036	5.016503	10.790722
C	1.967612	5.800114	8.893823
H	2.852665	6.266262	9.316085
H	8.445842	5.394166	3.775696
H	6.569668	6.248682	2.191949
H	4.334629	4.673891	5.838514

Table S23: Cartesian coordinates of the optimized structure of *sym*-[μ**H**-($Fe^{1.5}Fe^{1.5}$)adt]^0 (–4788.81452017 E_h)



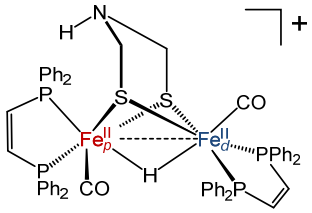
sym-[μ**H**-($Fe^{1.5}Fe^{1.5}$)adt]^0

atom	x	y	z
Fe	5.119535	4.075973	4.454549
Fe	4.950370	6.214133	6.159062
S	6.916772	5.033302	5.619210
S	4.708219	6.281497	3.824565
P	6.144190	3.171160	2.631933
P	5.366868	2.085206	5.418698
P	5.606856	8.371203	6.697875
P	2.859036	6.923129	6.599835
O	2.493117	3.341047	3.481397
O	5.315197	5.289791	8.882423
N	7.396606	7.186886	3.815779
H	7.205181	7.936725	4.471925
C	5.407666	3.372713	0.956368
C	4.443501	4.363250	0.745767
H	4.155496	5.013736	1.567896
C	3.857907	4.521858	-0.509876
H	3.111490	5.297057	-0.658772
C	4.224543	3.691574	-1.564076
H	3.764366	3.813262	-2.540895
C	5.188181	2.702773	-1.365488
H	5.482528	2.054459	-2.186342
C	5.777457	2.546935	-0.115668
H	6.544211	1.788354	0.019458
C	7.950641	3.292033	2.278142
C	8.850237	2.796457	3.232459
H	8.477190	2.317590	4.133025
C	10.222382	2.940549	3.053260
H	10.904352	2.551362	3.804232
C	10.718327	3.590148	1.924224
H	11.789747	3.706660	1.787174
C	9.832145	4.095755	0.976420
H	10.208911	4.609565	0.096231
C	8.457767	3.948696	1.150290
H	7.778720	4.347570	0.402753
C	5.986092	1.353674	2.863946
H	6.175663	0.682677	2.026661
C	5.645205	0.880122	4.066556
H	5.540258	-0.190161	4.242433
C	3.968721	1.322739	6.342708
C	4.016523	-0.028170	6.713966
H	4.889010	-0.629795	6.471391
C	2.966385	-0.604889	7.418522
H	3.014357	-1.654118	7.697153
C	1.860039	0.165986	7.775323
H	1.041490	-0.283486	8.331064
C	1.812487	1.512159	7.428281

H	0.965999	2.127510	7.717855
C	2.861249	2.088665	6.712793
H	2.824973	3.141820	6.453147
C	6.753044	1.695737	6.566645
C	7.782585	0.809917	6.233454
H	7.788466	0.319925	5.263433
C	8.808481	0.543197	7.139821
H	9.598965	-0.151430	6.868059
C	8.814479	1.158099	8.387887
H	9.612823	0.949797	9.094937
C	7.790652	2.041478	8.727398
H	7.787725	2.529547	9.697850
C	6.767888	2.310373	7.824684
H	5.980708	3.003879	8.103350
C	3.558201	3.615034	3.863935
C	7.972376	6.052806	4.468637
H	8.815186	6.396055	5.075827
C	6.249458	6.940129	3.002970
H	5.947890	7.880378	2.534068
C	5.146810	5.679513	7.795560
C	7.010856	8.668972	7.850984
C	7.199642	9.889682	8.515852
H	6.525167	10.721845	8.330195
C	8.259690	10.057935	9.400603
H	8.393563	11.008753	9.909486
C	9.149289	9.008595	9.631367
H	9.976753	9.140381	10.323277
C	8.973731	7.794831	8.974245
H	9.662056	6.973031	9.150678
C	7.909137	7.623272	8.090121
H	7.765311	6.670427	7.585770
C	5.814882	9.715212	5.448041
C	6.998333	10.448352	5.289009
H	7.830045	10.291495	5.969256
C	7.120491	11.383510	4.262511
H	8.046741	11.941411	4.154584
C	6.064890	11.603433	3.382435
H	6.161477	12.334197	2.584331
C	4.885530	10.873155	3.526759
H	4.057466	11.030012	2.840886
C	4.763324	9.931559	4.542943
H	3.847556	9.352831	4.626513
C	4.205472	9.061484	7.664009
H	4.341499	9.964822	8.257935
C	3.021615	8.441993	7.614159
H	2.160869	8.812376	8.171070
C	1.722283	7.474153	5.260033
C	1.192086	8.769100	5.215001
H	1.450367	9.490926	5.984013
C	0.336564	9.151477	4.183474

H	-0.064228	10.161432	4.165096
C	-0.008685	8.242502	3.187181
H	-0.679787	8.539336	2.385845
C	0.515284	6.952156	3.220652
H	0.260475	6.235975	2.444656
C	1.382618	6.573715	4.241500
H	1.795945	5.570974	4.238805
C	1.716359	5.961283	7.691957
C	0.531103	5.375014	7.234466
H	0.233274	5.485410	6.196858
C	-0.293622	4.667960	8.108097
H	-1.215899	4.230166	7.735179
C	0.052139	4.535729	9.450653
H	-0.594788	3.989478	10.131576
C	1.231733	5.113267	9.915387
H	1.512859	5.017585	10.960480
C	2.057127	5.818177	9.044693
H	2.973481	6.259596	9.424156
H	8.357000	5.364471	3.712631
H	6.513904	6.226503	2.214304
H	4.361605	4.590526	5.76707

Table S24: Cartesian coordinates of the optimized structure of *unsym*-[μ**H**-($Fe^{II}Fe^{II}$)adt]$^+$ (–4788.66283980 E_h)



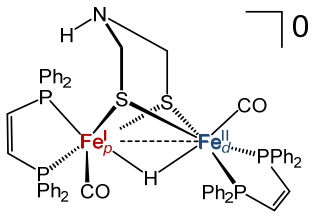
unsym-[μ**H**-($Fe^{II}Fe^{II}$)adt]$^+$

atom	x	y	z
Fe	4.635587	13.824517	3.824910
Fe	5.838108	11.464927	3.754654
H	5.344078	12.640031	4.829002
S	3.729376	11.931754	2.934450
S	6.458247	13.322471	2.561781
P	2.870996	14.248857	5.183904
P	5.154296	9.869118	5.209521
P	5.669086	15.522294	4.900766
P	6.590490	9.757125	2.467678
O	3.493144	15.579487	1.825153
O	8.503603	11.462633	4.913443
C	3.732451	12.167637	1.073018
C	5.901568	13.239398	0.775106
C	7.431515	11.468153	4.483438
C	3.942815	14.910843	2.655400
C	6.439198	16.847776	3.888309
C	5.591882	17.741398	3.218415
H	4.514309	17.665024	3.332568
C	6.118714	18.742206	2.408044
H	5.449070	19.430290	1.900585
C	7.497702	18.860764	2.251697
H	7.909449	19.641692	1.619343
C	8.345807	17.979264	2.916345
H	9.422437	18.071359	2.807409
C	7.822612	16.979012	3.731885
H	8.500921	16.315588	4.256856
C	6.895495	15.168688	6.213367
C	6.702483	15.577524	7.538417
H	5.806698	16.121743	7.820876
C	7.657055	15.287257	8.510869
H	7.497863	15.617166	9.533424
C	8.813272	14.587916	8.172292
H	9.561897	14.373004	8.929385
C	9.007781	14.166659	6.857814
H	9.899918	13.610896	6.586170
C	8.053089	14.450709	5.885240
H	8.209295	14.109203	4.866012
C	1.260334	14.367389	4.331105
C	0.727702	15.606625	3.959184
H	1.245315	16.526497	4.216607
C	-0.467563	15.675031	3.246756
H	-0.870508	16.643163	2.964650
C	-1.140874	14.507553	2.899036
H	-2.073719	14.561745	2.345674
C	-0.613999	13.268714	3.261276
H	-1.134969	12.354955	2.990489

C	0.582514	13.194380	3.967727
H	0.988664	12.223869	4.237963
C	2.486744	13.394407	6.764185
C	3.557058	13.037887	7.592621
H	4.577604	13.199345	7.256867
C	3.323031	12.470603	8.842153
H	4.163744	12.183867	9.466136
C	2.016539	12.255565	9.276897
H	1.833165	11.809894	10.250171
C	0.946002	12.616521	8.461547
H	-0.074472	12.461529	8.800305
C	1.176477	13.187083	7.211445
H	0.335134	13.470380	6.587526
C	8.286180	9.934817	1.801399
C	9.383096	9.446469	2.519989
H	9.233096	8.915650	3.455672
C	10.679687	9.644036	2.050235
H	11.520383	9.257362	2.618429
C	10.895988	10.331513	0.859902
H	11.906841	10.483047	0.493546
C	9.810161	10.822717	0.136919
H	9.970748	11.355316	-0.795723
C	8.515251	10.629282	0.605886
H	7.681030	11.004027	0.019387
C	5.645877	8.967591	1.095369
C	6.283824	8.329869	0.022474
H	7.364910	8.357688	-0.066420
C	5.534742	7.655725	-0.938901
H	6.040848	7.171604	-1.768836
C	4.147286	7.598351	-0.835836
H	3.566941	7.072607	-1.588184
C	3.506441	8.215517	0.236579
H	2.424994	8.169918	0.326879
C	4.249035	8.897152	1.196572
H	3.741976	9.376782	2.029837
C	3.429800	9.242037	5.228909
C	2.464262	9.879434	6.017312
H	2.741528	10.714288	6.652444
C	1.146330	9.431701	6.010006
H	0.411354	9.932776	6.632859
C	0.776724	8.344279	5.220479
H	-0.250890	7.992649	5.222561
C	1.733371	7.701589	4.439545
H	1.457676	6.843407	3.833427
C	5.571405	10.001341	6.993130
C	5.171747	8.986970	7.875463
H	4.543882	8.173438	7.521834
C	5.562307	9.018903	9.209764
H	5.247771	8.226045	9.882053
C	6.358219	10.062991	9.680780

H	6.669670	10.081918	10.721234
C	6.750135	11.080177	8.814610
H	7.363414	11.902537	9.171003
C	6.355752	11.051931	7.477786
H	6.658183	11.859252	6.820051
C	6.082112	8.357037	4.754898
H	6.074754	7.505961	5.434955
C	6.729819	8.319285	3.587599
H	7.268709	7.433809	3.251926
C	4.366704	16.459748	5.773392
H	4.601062	17.433027	6.203169
C	3.142364	15.929818	5.854006
H	2.332092	16.432146	6.381802
C	3.053453	8.147185	4.441960
H	3.790252	7.630063	3.834233
N	4.989353	12.212531	0.403273
H	5.422266	11.294856	0.384115
H	3.152525	11.336349	0.668253
H	3.179110	13.092275	0.889633
H	5.467324	14.219427	0.557809
H	6.818729	13.137843	0.190948

Table S25: Cartesian coordinates of the optimized structure of *unsym*-[μ**H**-($Fe^{I}Fe^{II}$)adt]^{0} (–4788.81465481 E_h)



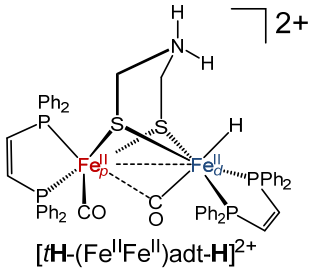
unsym-[μ**H**-($Fe^{I}Fe^{II}$)adt]^{0}

atom	x	y	z
Fe	4.611717	13.983800	3.888902
Fe	5.923450	11.445299	3.783111
H	5.227004	12.981408	4.943166
S	3.775144	12.006958	3.014854
S	6.519829	13.418738	2.711366
P	2.844595	14.356473	5.163897
P	5.269146	9.836472	5.191512
P	5.589762	15.645272	4.965946
P	6.644674	9.659739	2.402459
O	3.551184	15.736383	1.805152
O	8.602201	11.402588	4.905995
C	3.821842	12.268286	1.165166
C	6.008534	13.337556	0.921378
C	7.510768	11.422980	4.489991
C	3.971684	15.039340	2.634551
C	6.379479	16.998458	4.000342
C	5.560577	17.950273	3.379374
H	4.482462	17.905963	3.507794
C	6.114366	18.955099	2.591689
H	5.465188	19.687297	2.119526
C	7.493464	19.017191	2.406108
H	7.926399	19.799602	1.789001
C	8.314233	18.071390	3.014996
H	9.390889	18.111997	2.874962
C	7.762929	17.068587	3.808421
H	8.415891	16.343664	4.283473
C	6.825681	15.301863	6.280459
C	6.742311	15.853353	7.564019
H	5.924363	16.517854	7.824320
C	7.700075	15.545974	8.528090
H	7.620447	15.979948	9.521198
C	8.754459	14.688892	8.220812
H	9.503095	14.454047	8.972728
C	8.843666	14.131772	6.946162
H	9.649036	13.447981	6.695993
C	7.883147	14.432961	5.984046
H	7.949675	13.981801	4.998731
C	1.227157	14.484969	4.314104
C	0.677769	15.725976	3.975280
H	1.183655	16.643207	4.264220
C	-0.512056	15.801017	3.254239
H	-0.925180	16.772507	2.997418
C	-1.165066	14.636138	2.861852
H	-2.092771	14.694254	2.299323
C	-0.620648	13.394922	3.187617
H	-1.120847	12.481713	2.877226

C	0.569925	13.315790	3.903833
H	0.996677	12.344728	4.137310
C	2.435344	13.436886	6.708674
C	3.496757	12.987792	7.502303
H	4.516135	13.110803	7.147380
C	3.252762	12.364919	8.723695
H	4.088505	12.004182	9.315794
C	1.943502	12.180966	9.163832
H	1.752001	11.686299	10.112094
C	0.880735	12.628073	8.381813
H	-0.142691	12.490942	8.721014
C	1.123285	13.257505	7.162474
H	0.288078	13.605002	6.562516
C	8.301865	9.769991	1.625494
C	9.454152	9.436711	2.347497
H	9.366187	9.028921	3.350448
C	10.719016	9.636680	1.798854
H	11.601172	9.370560	2.374746
C	10.853567	10.177688	0.523609
H	11.840276	10.333790	0.096723
C	9.713847	10.523414	-0.201137
H	9.807765	10.948784	-1.196600
C	8.450538	10.327096	0.346488
H	7.572953	10.595191	-0.237293
C	5.657080	8.754717	1.122906
C	6.224762	7.958482	0.118854
H	7.304704	7.881978	0.034167
C	5.414312	7.267639	-0.777698
H	5.869541	6.657127	-1.553070
C	4.026509	7.353615	-0.680665
H	3.396674	6.814623	-1.383137
C	3.451850	8.133935	0.319198
H	2.371009	8.206193	0.404431
C	4.260491	8.832889	1.213070
H	3.807930	9.443722	1.990937
C	3.557131	9.152300	5.185796
C	2.545881	9.812859	5.895200
H	2.782822	10.690473	6.487988
C	1.235570	9.346119	5.858069
H	0.468191	9.871102	6.420120
C	0.912557	8.215324	5.109374
H	-0.110895	7.850974	5.082681
C	1.910301	7.553154	4.400155
H	1.671653	6.667353	3.817468
C	5.621837	9.934127	7.002111
C	5.196097	8.929137	7.881159
H	4.585233	8.109786	7.510698
C	5.535824	8.977008	9.229675
H	5.199346	8.189948	9.899286
C	6.306941	10.031096	9.719723

H	6.576265	10.065044	10.772100
C	6.727151	11.039657	8.856176
H	7.321770	11.870609	9.225587
C	6.383452	10.992491	7.505598
H	6.706487	11.786758	6.840413
C	6.242570	8.335567	4.767271
H	6.294205	7.513609	5.481625
C	6.865361	8.273434	3.585260
H	7.445623	7.398033	3.291320
C	4.280830	16.568385	5.858158
H	4.496155	17.531653	6.319844
C	3.063343	16.018409	5.913418
H	2.241004	16.490237	6.451417
C	3.224259	8.017457	4.438118
H	3.992681	7.490550	3.880076
N	5.106229	12.295394	0.528020
H	5.551038	11.388584	0.641119
H	3.246096	11.448021	0.728545
H	3.295341	13.204191	0.958921
H	5.570160	14.308099	0.669547
H	6.937734	13.220912	0.356701

Table S26: Cartesian coordinates of the optimized structure of [*t*H-($Fe^{II}Fe^{II}$)adt-**H**]$^{2+}$ (–4788.96705002 E_h)



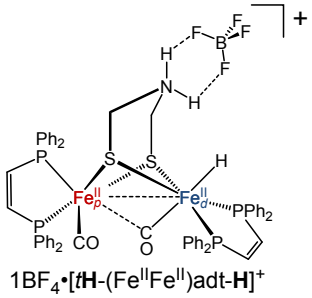
[*t*H-($Fe^{II}Fe^{II}$)adt-**H**]$^{2+}$

atom	x	y	z
Fe	12.397076	3.291206	4.065225
Fe	14.038075	5.354641	4.350989
S	13.209995	4.641881	2.355941
S	14.606118	3.204997	4.723784
P	10.405577	2.914359	3.064778
P	11.871701	1.673596	5.527631
P	16.004433	6.270695	3.811683
P	13.120575	7.467967	4.077737
N	14.627779	2.275989	2.133202
O	11.200143	5.218629	5.901907
O	14.438524	5.892144	7.194995
C	10.591317	2.472132	1.300794
C	10.609216	1.128755	0.906018
H	10.448918	0.340326	1.636661
C	10.817115	0.788267	-0.429653
H	10.811661	-0.256574	-0.725623
C	11.010859	1.785066	-1.382987
H	11.160782	1.519864	-2.425183
C	10.995202	3.125554	-0.997554
H	11.131025	3.906915	-1.739616
C	10.789856	3.471456	0.336418
H	10.777029	4.519816	0.621168
C	8.907556	3.971885	3.152364
C	8.497479	4.435381	4.411641
H	9.090296	4.228704	5.296170
C	7.310759	5.149495	4.546110
H	7.003392	5.498349	5.527237
C	6.515157	5.402704	3.430464
H	5.586478	5.954824	3.537582
C	6.905580	4.929304	2.180107
H	6.278569	5.103519	1.310702
C	8.093411	4.214639	2.037642
H	8.369750	3.834509	1.060133
C	9.740702	1.403865	3.843985
H	8.819808	0.979914	3.444824
C	10.334857	0.902639	4.930237
H	9.944277	0.023162	5.439653
C	11.562504	2.095045	7.272241
C	11.801068	4.568145	5.154354
C	14.305422	3.538073	1.410317
H	13.769971	3.249785	0.502029
H	15.236750	4.044564	1.151700
C	15.491360	2.423545	3.333948
H	16.376773	3.000100	3.057933
H	15.778869	1.417430	3.647599
C	14.287363	5.709262	6.070246

C	16.666054	5.915691	2.138897
C	17.761696	5.060113	1.960934
H	18.278332	4.648740	2.824146
C	18.225245	4.771064	0.677485
H	19.085079	4.119948	0.550102
C	17.606908	5.339065	-0.434484
H	17.979851	5.126841	-1.431865
C	16.520324	6.197563	-0.262943
H	16.046400	6.655009	-1.126257
C	16.047534	6.482206	1.015408
H	15.196648	7.147216	1.136738
C	15.776322	8.076988	3.753034
H	16.646038	8.719995	3.630972
C	14.543793	8.580880	3.845124
H	14.365036	9.655284	3.829348
C	12.274836	8.295493	5.477158
H	12.511263	1.988612	3.268949
H	15.020463	1.582769	1.487932
H	13.688039	1.941704	2.518096
C	13.033468	0.254584	5.556912
C	13.134198	-0.556502	4.416833
H	12.502991	-0.360758	3.552502
C	14.006962	-1.640398	4.398641
H	14.057924	-2.279848	3.521897
C	14.795160	-1.920549	5.515161
H	15.471236	-2.770043	5.504299
C	14.696550	-1.121016	6.650461
H	15.295894	-1.344476	7.527794
C	13.816644	-0.039993	6.676861
H	13.733952	0.561312	7.576364
C	12.524335	2.845174	7.963788
C	12.326769	3.182345	9.298572
H	13.079729	3.759968	9.825780
C	11.161345	2.785979	9.952623
H	11.004935	3.052817	10.993234
C	10.196367	2.050622	9.269614
H	9.286406	1.742315	9.775113
C	10.393702	1.702403	7.935492
H	9.628865	1.128984	7.421871
C	17.454822	6.050145	4.899993
C	17.470247	5.096129	5.923240
H	16.612829	4.451858	6.084487
C	18.591432	4.959169	6.739727
H	18.591402	4.217631	7.532712
C	19.703435	5.772000	6.542922
H	20.573623	5.668544	7.183780
C	19.701616	6.718381	5.518120
C	18.588072	6.855583	4.698220
C	12.069890	7.782255	2.617981
C	12.443797	8.709926	1.638181

H	13.375863	9.259836	1.724196
C	11.616351	8.950264	0.543541
H	11.913631	9.679742	-0.203747
C	10.406824	8.272605	0.418561
H	9.755940	8.473493	-0.426975
C	10.030536	7.341950	1.385579
H	9.085389	6.813923	1.302805
C	10.860918	7.087956	2.473900
H	10.551571	6.354698	3.214124
C	13.055363	8.728294	6.558980
H	14.132279	8.584111	6.557700
C	12.461772	9.371399	7.640198
H	13.077631	9.710832	8.467334
C	11.085662	9.587094	7.654522
H	10.623363	10.092939	8.496566
C	10.306872	9.165110	6.580139
H	9.236147	9.344922	6.579681
C	10.894763	8.523540	5.493713
H	10.274972	8.223414	4.656043
H	13.433587	3.165255	7.461198
H	20.569719	7.349882	5.356521
H	18.614660	7.582602	3.891337

Table S27: Cartesian coordinates of the optimized structure of [*t*H-($Fe^{II}Fe^{II}$)adt-**H**]$^{2+}$ and $1BF_4^-$ (–5214.45104427 E_h)



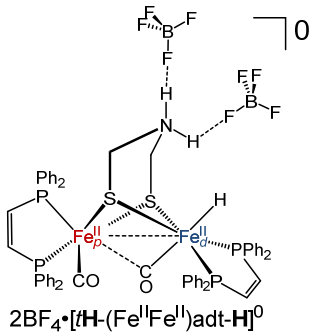
$1BF_4$•[*t*H-($Fe^{II}Fe^{II}$)adt-**H**]$^+$

atom	x	y	z
Fe	8.954269	14.343289	4.890860
Fe	7.249073	16.326675	4.729789
S	8.188919	15.609460	6.670528
S	6.734237	14.165091	4.376366
P	10.915174	13.912889	5.895796
P	9.496353	12.777443	3.379560
P	5.299395	17.175417	5.396821
P	8.125820	18.441499	4.993432
N	6.778930	13.228619	6.996753
O	10.057573	16.365918	3.103140
O	6.710240	16.811875	1.902095
C	10.716469	13.544631	7.672467
C	10.415887	12.239304	8.076404
H	10.368060	11.432204	7.351294
C	10.133720	11.961323	9.412848
H	9.871110	10.949087	9.702932
C	10.158891	12.985535	10.356012
H	9.939263	12.768616	11.397134
C	10.456309	14.290428	9.960445
H	10.473956	15.092126	10.693251
C	10.725055	14.575167	8.624251
H	10.932832	15.598610	8.325199
C	12.461459	14.905633	5.774505
C	12.801139	15.468971	4.536459
H	12.131391	15.376503	3.688611
C	14.008927	16.142045	4.374313
H	14.256761	16.572832	3.408712
C	14.897322	16.254200	5.441604
H	15.840386	16.777327	5.313819
C	14.577888	15.678859	6.669340
H	15.273820	15.744433	7.500699
C	13.369878	15.004932	6.837180
H	13.142931	14.549793	7.795319
C	11.525956	12.358256	5.157888
H	12.389679	11.870694	5.607990
C	10.953017	11.903036	4.040863
H	11.302900	11.001674	3.540077
C	9.991184	13.302314	1.702187
C	9.084379	14.050866	0.939021
H	8.106164	14.299704	1.343260
C	9.425458	14.479805	-0.339038
H	8.712585	15.055387	-0.921692
C	10.681015	14.178172	-0.864520
H	10.948714	14.516417	-1.861041
C	11.592064	13.446076	-0.108580
H	12.572006	13.210220	-0.512790

C	11.250403	13.006987	1.169008
H	11.972823	12.437665	1.745757
C	9.490055	15.666826	3.839956
C	7.167429	14.481438	7.671059
H	7.766658	14.194072	8.538936
H	6.264416	14.992372	8.006565
C	5.929202	13.352533	5.798623
H	5.019270	13.888594	6.074834
H	5.676917	12.335692	5.488131
C	6.912477	16.656319	3.025276
C	4.684087	16.767317	7.076190
C	3.634622	15.857783	7.259264
H	3.120512	15.437340	6.399155
C	3.225159	15.507235	8.545003
H	2.413412	14.798235	8.676443
C	3.848501	16.071541	9.655759
H	3.525683	15.799992	10.656086
C	4.883009	16.990403	9.479897
H	5.364810	17.439901	10.343080
C	5.304320	17.333779	8.198178
H	6.123158	18.036828	8.072393
C	5.492957	18.985267	5.524974
H	4.623138	19.604986	5.735277
C	6.710160	19.517949	5.394104
H	6.871536	20.593148	5.460842
C	9.317686	18.748479	6.343981
C	9.021047	19.616805	7.400715
H	8.065876	20.131535	7.439989
C	9.950487	19.835372	8.414961
H	9.709142	20.515562	9.226412
C	11.186165	19.194927	8.382381
H	11.913429	19.375825	9.168290
C	11.487307	18.322624	7.337932
H	12.447713	17.816656	7.301339
C	10.554851	18.090507	6.330520
C	7.911651	19.743039	2.539232
H	6.851497	19.521197	2.624144
C	8.369978	20.437415	1.424400
H	7.667098	20.739969	0.654001
C	9.722555	20.746482	1.302044
H	10.080447	21.289657	0.432581
C	10.612071	20.365308	2.302830
H	11.666138	20.612451	2.218712
C	10.157988	19.673128	3.422119
H	10.862568	19.400054	4.199839
H	8.863303	13.032687	5.633194
H	6.330851	12.556073	7.682234
H	7.646916	12.743227	6.708237
B	6.738946	10.190199	8.193870
F	5.995966	9.325177	7.416998

F	7.243119	9.632085	9.339434
F	7.797817	10.748692	7.403481
F	5.895330	11.334985	8.546679
C	8.300021	11.415177	3.124348
C	7.912299	10.659009	4.239460
H	8.315842	10.865632	5.226132
C	7.003176	9.614218	4.102537
H	6.711501	9.049122	4.983184
C	6.475582	9.314280	2.846740
H	5.764799	8.500454	2.737314
C	6.866734	10.053327	1.733396
H	6.466930	9.815141	0.751958
C	7.777436	11.100120	1.865878
H	8.079776	11.660668	0.987565
C	3.809572	16.966452	4.354760
C	3.758490	15.992193	3.352367
H	4.610101	15.342727	3.178708
C	2.608604	15.836937	2.579995
H	2.581354	15.075809	1.806182
C	1.503362	16.652680	2.799576
H	0.609733	16.533589	2.194391
C	1.541079	17.619520	3.804175
H	0.677503	18.252093	3.985919
C	2.682984	17.772933	4.581244
H	2.683435	18.512531	5.376792
H	10.803013	17.397721	5.532106
C	8.803175	19.350959	3.547542

Table S28: Cartesian coordinates of the optimized structure of [*t*H-($Fe^{II}Fe^{II}$)adt-**H**]$^{2+}$ and $2BF_4^-$ (–5639.82929386 E_h)



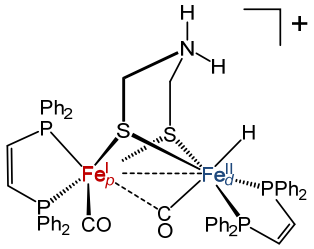
2BF$_4$•[*t*H-($Fe^{II}Fe^{II}$)adt-**H**]^0

atom	x	y	z
Fe	8.844445	14.311357	4.995428
Fe	7.183486	16.284840	4.781803
S	8.116487	15.606353	6.744594
S	6.631765	14.116502	4.560144
P	10.762731	13.801472	6.011726
P	9.385402	12.765921	3.456688
P	5.253942	17.190500	5.415718
P	8.083109	18.394382	4.953643
N	6.559243	13.307890	7.273421
O	10.083097	16.347650	3.320675
O	6.658287	16.617717	1.935248
C	10.617828	13.550876	7.810087
C	10.380926	12.266118	8.309650
H	10.343484	11.406468	7.647218
C	10.134642	12.078671	9.669582
H	9.909864	11.078645	10.025317
C	10.136839	13.167297	10.536665
H	9.929189	13.021071	11.592434
C	10.380468	14.450404	10.044408
H	10.372396	15.303770	10.716656
C	10.610679	14.647637	8.686277
H	10.765801	15.654971	8.310684
C	12.338942	14.737145	5.780125
C	12.708353	15.099488	4.477163
H	12.056297	14.872009	3.640210
C	13.918949	15.742311	4.236312
H	14.187380	16.014266	3.219533
C	14.781756	16.029378	5.291863
H	15.727080	16.530508	5.103778
C	14.432845	15.656951	6.587465
H	15.109027	15.856673	7.414194
C	13.222077	15.010992	6.832337
H	12.973264	14.707929	7.843745
C	11.279436	12.189688	5.330390
H	12.067112	11.629350	5.830226
C	10.706167	11.757720	4.205218
H	10.977867	10.805754	3.753507
C	10.095448	13.301572	1.851222
C	9.392958	14.227647	1.069027
H	8.453119	14.640707	1.424629
C	9.890783	14.635910	-0.164382
H	9.331559	15.353096	-0.758007
C	11.106102	14.135895	-0.627024
H	11.498031	14.458629	-1.587302
C	11.818913	13.225336	0.148114
H	12.768591	12.833120	-0.204498

C	11.316599	12.807234	1.378484
H	11.884286	12.095416	1.969514
C	9.450987	15.642205	4.001977
C	7.085346	14.575086	7.829812
H	7.695801	14.303198	8.692395
H	6.238768	15.160059	8.187986
C	5.794003	13.393784	6.013334
H	4.879323	13.953802	6.216905
H	5.536007	12.367599	5.742937
C	6.854813	16.523175	3.069760
C	4.676050	16.955763	7.131704
C	3.626376	16.076457	7.427717
H	3.082844	15.582728	6.626874
C	3.276974	15.824840	8.751429
H	2.504294	15.101158	8.984215
C	3.957896	16.460848	9.786541
H	3.706555	16.222187	10.813828
C	4.982311	17.360152	9.497111
H	5.516728	17.854552	10.302949
C	5.349986	17.601796	8.176878
H	6.176023	18.274431	7.962364
C	5.450670	19.006427	5.369241
H	4.578985	19.644118	5.506421
C	6.675233	19.519807	5.231186
H	6.848236	20.595237	5.219094
C	9.237418	18.772809	6.322291
C	8.914072	19.706294	7.313244
H	7.964518	20.232164	7.283509
C	9.804002	19.970765	8.351782
H	9.538147	20.697156	9.114277
C	11.028469	19.311440	8.408994
H	11.723628	19.522760	9.216286
C	11.357046	18.376011	7.429805
H	12.307176	17.850584	7.464412
C	10.463616	18.097827	6.399099
C	7.979427	19.555055	2.419699
H	6.918464	19.327722	2.470134
C	8.483359	20.188761	1.288179
H	7.813549	20.440090	0.470923
C	9.838225	20.500780	1.207909
H	10.232491	20.995512	0.324980
C	10.683385	20.181862	2.267152
H	11.740155	20.427391	2.215412
C	10.182420	19.549716	3.401859
H	10.853271	19.318379	4.221830
H	8.693137	13.022235	5.749117
H	5.874799	12.930075	7.989717
H	7.296848	12.583115	7.176485
B	8.382324	9.607450	7.288820
F	7.530412	8.667545	6.726096

F	9.596742	9.664081	6.565726
F	7.760738	10.918373	7.140699
F	8.630537	9.380067	8.630450
B	4.664194	12.896288	10.254593
F	5.739268	13.806866	10.360421
F	3.473545	13.493129	10.666665
F	4.924752	11.727883	10.929273
F	4.544724	12.607038	8.821565
C	8.123426	11.529659	2.985608
C	7.781283	10.560657	3.935572
H	8.278812	10.528116	4.898322
C	6.803931	9.611623	3.647783
H	6.566546	8.867709	4.401976
C	6.159345	9.629507	2.412722
H	5.396202	8.889837	2.186708
C	6.501023	10.590153	1.462477
H	6.009179	10.599790	0.493556
C	7.481933	11.538157	1.742355
H	7.750568	12.269015	0.986309
C	3.740883	16.908667	4.420347
C	3.682972	15.898453	3.455779
H	4.540428	15.254565	3.290469
C	2.517982	15.695543	2.717379
H	2.485921	14.903274	1.975474
C	1.403791	16.499356	2.933809
H	0.496981	16.340567	2.357550
C	1.448334	17.502714	3.901627
H	0.576694	18.124147	4.084805
C	2.605809	17.703165	4.644103
H	2.613274	18.465657	5.418032
H	10.731522	17.353326	5.655625
C	8.825495	19.225432	3.487068

Table S29: Cartesian coordinates of the optimized structure of [*t***H**-($Fe^{I}Fe^{II}$)adt-**H**]$^{+}$ (–4789.23851241 E_h)



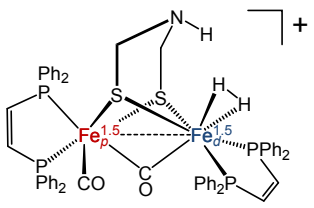
[*t***H**-($Fe^{I}Fe^{II}$)adt-**H**]$^{+}$

atom	x	y	z
Fe	12.180853	3.192851	4.148116
Fe	14.073312	5.543279	4.463268
S	13.107068	4.646144	2.532523
S	14.365460	3.321832	5.015796
P	10.288585	2.772837	3.052065
P	11.684764	1.530206	5.521919
P	16.127114	6.359427	3.780224
P	13.268888	7.625611	4.100793
N	14.633960	2.338561	2.462014
O	10.941694	5.272435	5.797353
O	14.380019	6.085735	7.305907
C	10.511087	2.345057	1.286637
C	10.451764	1.015518	0.853852
H	10.214206	0.222394	1.557390
C	10.695040	0.693736	-0.480047
H	10.636989	-0.341789	-0.802787
C	11.000790	1.696605	-1.396373
H	11.183877	1.446464	-2.437269
C	11.063406	3.023848	-0.974000
H	11.295288	3.811961	-1.684717
C	10.824005	3.349969	0.358985
H	10.886739	4.386667	0.676762
C	8.785979	3.836016	3.087583
C	8.304728	4.240993	4.340952
H	8.831608	3.959705	5.247177
C	7.141784	4.998066	4.440743
H	6.782605	5.302626	5.419204
C	6.441593	5.359556	3.291702
H	5.533686	5.950276	3.369741
C	6.904447	4.950762	2.043668
H	6.354589	5.214198	1.144615
C	8.067834	4.190215	1.939100
H	8.404444	3.864843	0.960522
C	9.588065	1.255697	3.797334
H	8.672558	0.836683	3.380540
C	10.162397	0.740659	4.889318
H	9.748583	-0.138635	5.380056
C	11.367352	1.795153	7.304484
C	11.523106	4.510041	5.147697
C	14.304410	3.565834	1.683840
H	13.855488	3.217893	0.749330
H	15.226527	4.110446	1.475774
C	15.386755	2.528522	3.729233
H	16.275638	3.129958	3.525380
H	15.677054	1.531342	4.070801
C	14.259760	5.912914	6.163396

C	16.766563	6.005536	2.086581
C	17.835093	5.124054	1.871495
H	18.387328	4.726090	2.718809
C	18.216678	4.775020	0.576878
H	19.055241	4.100364	0.427281
C	17.540275	5.303125	-0.520837
H	17.844845	5.038474	-1.529171
C	16.478945	6.184254	-0.317387
H	15.952283	6.608337	-1.167690
C	16.091499	6.530037	0.974623
H	15.254759	7.208448	1.119418
C	15.932766	8.179136	3.676667
H	16.807122	8.809553	3.517556
C	14.711453	8.710972	3.786810
H	14.557652	9.789052	3.742323
C	12.463074	8.575388	5.457690
H	12.468597	1.913937	3.302261
H	15.098969	1.649923	1.864102
H	13.659550	1.979662	2.787667
C	12.886796	0.141886	5.511288
C	12.908120	-0.752096	4.433243
H	12.174832	-0.656267	3.635991
C	13.853598	-1.772794	4.382646
H	13.848157	-2.473613	3.552359
C	14.796076	-1.902555	5.402179
H	15.531846	-2.700620	5.365117
C	14.782694	-1.013906	6.474792
H	15.509811	-1.114644	7.275265
C	13.830152	0.001809	6.534291
H	13.820223	0.683407	7.379274
C	11.958448	2.886862	7.950707
C	11.760684	3.090513	9.314583
H	12.225863	3.942346	9.801186
C	10.966986	2.209749	10.043573
H	10.806995	2.372342	11.105342
C	10.379417	1.115762	9.409880
H	9.763392	0.423228	9.975842
C	10.582613	0.904488	8.050371
H	10.130418	0.037304	7.578811
C	17.637663	6.183995	4.808160
C	17.607687	5.327097	5.913366
H	16.699716	4.775723	6.140078
C	18.731133	5.184049	6.726794
H	18.694246	4.518774	7.584523
C	19.891660	5.897728	6.444280
H	20.765305	5.791076	7.080734
C	19.933323	6.751748	5.341649
C	18.815611	6.891936	4.526730
C	12.183324	7.937373	2.654236
C	12.529989	8.835798	1.639266

H	13.463633	9.387639	1.691736
C	11.682708	9.037349	0.551301
H	11.965232	9.741638	-0.225990
C	10.474993	8.350418	0.467616
H	9.810488	8.517468	-0.375221
C	10.121170	7.449833	1.470774
H	9.180431	6.909101	1.418874
C	10.974437	7.235247	2.549721
H	10.689994	6.520059	3.317093
C	13.263911	9.018695	6.519079
H	14.332102	8.820350	6.518666
C	12.704173	9.722328	7.580746
H	13.339498	10.063062	8.393053
C	11.337096	9.988196	7.599855
H	10.899898	10.537680	8.428396
C	10.535188	9.552089	6.548836
H	9.469586	9.762676	6.552045
C	11.092628	8.851866	5.481969
H	10.454995	8.536921	4.662812
H	12.580227	3.581008	7.395020
H	20.838911	7.307893	5.117128
H	18.866728	7.545622	3.659657

Table S30: Cartesian coordinates of the optimized structure of [**H**$_2$-(Fe$^{1.5}$Fe$^{1.5}$)adt-**H**]$^+$ (–4789.23330398 E_h)



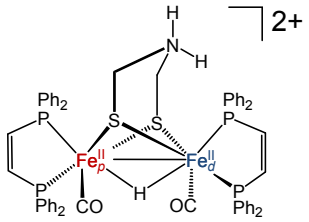
[**H**$_2$-(Fe$^{1.5}$Fe$^{1.5}$)adt]$^+$

atom	x	y	z
Fe	12.521617	3.111542	4.008774
Fe	13.926125	5.361107	4.441476
S	13.296807	4.528865	2.341009
S	14.684053	3.196306	4.822016
P	10.377775	2.912701	3.236793
P	11.906264	1.678434	5.661757
P	15.961796	6.403047	3.927887
P	13.024127	7.457539	4.123823
N	15.330279	2.523384	2.140259
O	11.247453	5.081199	5.722196
O	14.435992	5.987811	7.235696
C	10.264408	1.888955	1.716835
C	9.460217	0.745832	1.644401
H	8.863268	0.438980	2.497823
C	9.417455	-0.012678	0.476418
H	8.790928	-0.898810	0.434159
C	10.169052	0.365576	-0.633033
H	10.131706	-0.225627	-1.543121
C	10.967977	1.505856	-0.572188
H	11.552187	1.808462	-1.436498
C	11.021579	2.261470	0.596264
H	11.647110	3.150420	0.642411
C	9.169648	4.262926	2.925058
C	8.665070	4.982696	4.018140
H	9.028591	4.782353	5.020182
C	7.694937	5.962313	3.823954
H	7.307259	6.505264	4.680890
C	7.216484	6.235194	2.544380
H	6.453189	6.993512	2.396941
C	7.711872	5.521598	1.455695
H	7.331212	5.716214	0.456951
C	8.683025	4.540248	1.641575
H	9.043946	3.977730	0.786682
C	9.451662	1.944569	4.480392
H	8.373066	1.837024	4.370681
C	10.104298	1.415927	5.519404
H	9.583845	0.842560	6.285384
C	12.122821	1.919375	7.461848
C	12.090378	4.641494	5.022781
C	14.727096	3.647166	1.501138
H	14.335852	3.328553	0.531783
H	15.481964	4.415337	1.327919
C	15.864066	2.713711	3.443879
H	16.635750	3.488959	3.415544
H	16.323564	1.781878	3.782035
C	14.222426	5.752453	6.123704

C	16.632734	6.428729	2.216478
C	17.820931	5.770406	1.875977
H	18.414899	5.282655	2.642956
C	18.255224	5.744353	0.552497
H	19.179084	5.231502	0.301688
C	17.511529	6.374320	-0.442226
H	17.853637	6.354914	-1.472713
C	16.327740	7.031167	-0.110162
H	15.742371	7.525713	-0.879766
C	15.886900	7.056259	1.209415
H	14.953294	7.557048	1.448983
C	15.665923	8.182620	4.223748
H	16.517320	8.855471	4.314290
C	14.411495	8.629064	4.320367
H	14.192142	9.679455	4.508090
C	11.838690	8.129828	5.354624
H	12.948342	1.523691	3.391001
H	14.763090	1.686448	2.079882
H	12.741228	1.927565	2.745369
C	12.592736	-0.006875	5.407507
C	11.982644	-0.905862	4.524559
H	11.049803	-0.640937	4.033559
C	12.563834	-2.146489	4.272660
H	12.076705	-2.841206	3.594647
C	13.760889	-2.496515	4.892962
H	14.211842	-3.465006	4.698085
C	14.375292	-1.603727	5.769409
H	15.305962	-1.874724	6.259393
C	13.796454	-0.364436	6.028615
H	14.274868	0.322354	6.720777
C	12.729807	3.064004	7.978692
C	12.868572	3.228418	9.355281
H	13.344937	4.124699	9.740554
C	12.398164	2.249815	10.224833
H	12.503702	2.378233	11.298076
C	11.794669	1.099023	9.717187
H	11.432860	0.328964	10.392073
C	11.660447	0.931010	8.344169
H	11.211421	0.018593	7.960178
C	17.453098	6.118507	4.959289
C	17.480146	5.104479	5.921687
H	16.624464	4.447663	6.042352
C	18.605100	4.926205	6.725871
H	18.612317	4.134792	7.469493
C	19.709643	5.758680	6.579350
H	20.583838	5.620861	7.208833
C	19.694154	6.771101	5.619806
C	18.575793	6.949521	4.814204
C	12.308759	7.962197	2.512356
C	12.759000	9.105865	1.839506

H	13.558258	9.710765	2.255864
C	12.191030	9.482145	0.625044
H	12.550794	10.372343	0.117327
C	11.162253	8.725322	0.071011
H	10.716737	9.020742	-0.874430
C	10.706668	7.589103	0.734505
H	9.903516	6.992886	0.313739
C	11.278787	7.203539	1.943731
H	10.916157	6.309317	2.441018
C	12.317983	8.490923	6.620505
H	13.371252	8.381512	6.861926
C	11.452235	9.003547	7.581819
H	11.838799	9.285458	8.556779
C	10.099228	9.159872	7.291271
H	9.424289	9.564006	8.039979
C	9.617882	8.804643	6.034041
H	8.566029	8.936390	5.796755
C	10.479963	8.291752	5.068730
H	10.089864	8.031264	4.090364
H	13.098878	3.829853	7.311703
H	20.555395	7.421398	5.498130
H	18.586227	7.729106	4.056879

Table S31: Cartesian coordinates of the optimized structure of *sym*-[μ**H**-($Fe^{II}Fe^{II}$)adt-**H**]$^{2+}$ (–4788.98367639 E_h)



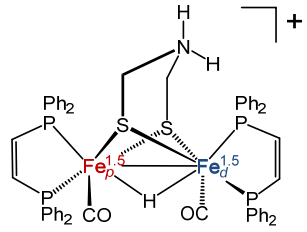
sym-[μ**H**-($Fe^{II}Fe^{II}$)adt-**H**]$^{2+}$

atom	x	y	z
Fe	5.121901	4.130346	4.461599
Fe	4.958153	6.186505	6.090015
S	6.916903	5.092002	5.545008
S	4.708245	6.280362	3.765877
P	6.032463	3.166224	2.630550
P	5.446039	2.136188	5.531322
P	5.623842	8.239064	6.786589
P	2.813203	6.926043	6.466509
O	2.506583	3.167693	3.625200
O	5.135725	5.175849	8.810039
N	7.322575	7.244619	3.765946
H	6.994111	7.951792	4.447011
C	5.132910	3.247024	1.040218
C	4.106860	4.175779	0.834297
H	3.828091	4.865788	1.624637
C	3.436101	4.222933	-0.386589
H	2.640278	4.946368	-0.535589
C	3.779306	3.342650	-1.408025
H	3.250522	3.374969	-2.355683
C	4.806061	2.418535	-1.214603
H	5.080222	1.732714	-2.010242
C	5.483194	2.373086	-0.001737
H	6.295001	1.661767	0.121712
C	7.760278	3.562990	2.141659
C	8.822545	3.160155	2.962975
H	8.625976	2.597646	3.870712
C	10.135068	3.483907	2.628374
H	10.950493	3.150797	3.263578
C	10.402750	4.223652	1.476104
H	11.427270	4.466729	1.211138
C	9.352792	4.631913	0.656561
H	9.556381	5.189170	-0.253229
C	8.037601	4.300316	0.982405
H	7.231573	4.586085	0.311989
C	6.109072	1.377315	2.981173
H	6.365071	0.687659	2.178864
C	5.838858	0.937226	4.213287
H	5.866225	-0.125981	4.446790
C	4.054509	1.370649	6.433308
C	4.143514	0.022279	6.812068
H	5.031086	-0.559552	6.579200
C	3.105190	-0.578934	7.512409
H	3.180557	-1.624332	7.795311
C	1.974240	0.161439	7.856513
H	1.165183	-0.310097	8.406010
C	1.888072	1.504234	7.503096

H	1.018871	2.091685	7.781106
C	2.923019	2.108646	6.792152
H	2.844742	3.158995	6.530227
C	6.837833	1.910900	6.701366
C	8.059362	1.361534	6.295492
H	8.195213	1.020145	5.273096
C	9.097448	1.202501	7.210868
H	10.033766	0.755086	6.890715
C	8.926232	1.593491	8.536326
H	9.732779	1.460243	9.250923
C	7.708467	2.131980	8.949079
H	7.560972	2.416290	9.986641
C	6.666724	2.286305	8.039632
H	5.711815	2.672675	8.382463
C	3.546260	3.547298	3.945478
C	7.982174	6.110687	4.491828
H	8.749782	6.568190	5.121793
C	6.164359	6.907533	2.883271
H	5.892979	7.840887	2.384437
C	5.060708	5.601676	7.743677
C	6.935669	8.374040	8.049961
C	7.103554	9.569164	8.769014
H	6.470667	10.429705	8.570937
C	8.096736	9.673174	9.736095
H	8.213726	10.599507	10.289977
C	8.938087	8.590761	9.994011
H	9.710556	8.673818	10.752349
C	8.784988	7.406060	9.280123
H	9.436600	6.560847	9.479503
C	7.787680	7.294653	8.312900
H	7.670456	6.361137	7.770390
C	6.102535	9.499876	5.524451
C	7.371867	10.099785	5.506787
H	8.095319	9.865993	6.282487
C	7.702106	11.016842	4.507827
H	8.682520	11.483976	4.511298
C	6.770380	11.352411	3.528143
H	7.024648	12.078271	2.761690
C	5.503439	10.765507	3.541968
H	4.769015	11.037409	2.789521
C	5.170766	9.840959	4.527613
H	4.182279	9.387469	4.525494
C	4.180515	9.029693	7.571282
H	4.313696	9.953149	8.131546
C	2.978661	8.466506	7.429218
H	2.088431	8.896536	7.886620
C	1.784007	7.434133	5.042453
C	1.383734	8.766397	4.877634
H	1.680193	9.525931	5.594628
C	0.585469	9.133432	3.796069

H	0.272655	10.167570	3.687570
C	0.175147	8.177381	2.871140
H	-0.457643	8.463608	2.036674
C	0.573611	6.850824	3.024191
H	0.252979	6.097765	2.310643
C	1.380017	6.481947	4.096440
H	1.680871	5.444351	4.196115
C	1.687719	5.967053	7.554466
C	0.530124	5.338904	7.080411
H	0.257663	5.397889	6.032596
C	-0.312618	4.665614	7.962795
H	-1.217200	4.198553	7.584697
C	-0.012045	4.613124	9.321850
H	-0.677989	4.098806	10.008052
C	1.136431	5.239728	9.801307
H	1.370043	5.216741	10.861289
C	1.981656	5.913172	8.925405
H	2.858545	6.415020	9.323514
H	8.461169	5.481262	3.739165
H	6.518316	6.186683	2.143790
H	4.327000	4.685614	5.805010
H	8.037550	7.715852	3.200054

Table S32: Cartesian coordinates of the optimized structure of *sym*-[μ**H**-($Fe^{1.5}Fe^{1.5}$)adt-**H**]$^+$ (–4789.23545763 E_h)



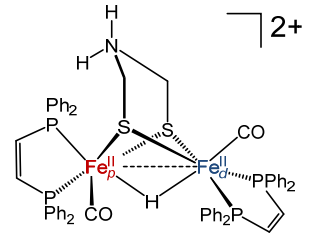
sym-[μ**H**-($Fe^{1.5}Fe^{1.5}$)adt-**H**]$^+$

atom	x	y	z
Fe	5.106096	4.054364	4.428101
Fe	4.970540	6.213700	6.148997
S	6.926035	4.974999	5.585459
S	4.666735	6.251942	3.803067
P	6.102819	3.145643	2.592118
P	5.362734	2.054339	5.407226
P	5.636612	8.390029	6.753884
P	2.855973	6.928512	6.597894
O	2.478620	3.246577	3.510201
O	5.289339	5.273740	8.877260
N	7.249730	7.188939	3.952175
H	6.810467	7.674173	4.754041
C	5.328027	3.310216	0.936462
C	4.310833	4.247834	0.730942
H	3.989688	4.883995	1.550487
C	3.706293	4.369461	-0.519613
H	2.916001	5.099986	-0.665709
C	4.107994	3.554103	-1.572747
H	3.632143	3.644735	-2.544807
C	5.124544	2.618551	-1.379143
H	5.443393	1.981770	-2.199119
C	5.733155	2.499357	-0.135027
H	6.537751	1.780312	-0.004921
C	7.888528	3.406681	2.218366
C	8.846873	2.948008	3.133488
H	8.536039	2.396124	4.015681
C	10.199092	3.212558	2.933513
H	10.929371	2.843474	3.647893
C	10.614347	3.946939	1.823406
H	11.669793	4.149346	1.666719
C	9.669382	4.411256	0.910974
H	9.985779	4.972941	0.036442
C	8.315077	4.142595	1.104407
H	7.590586	4.489770	0.373013
C	6.045959	1.333689	2.859216
H	6.276867	0.661739	2.033789
C	5.712769	0.861931	4.064534
H	5.660682	-0.209642	4.252296
C	3.952680	1.293382	6.298632
C	3.989168	-0.067895	6.633438
H	4.851081	-0.675570	6.370824
C	2.936091	-0.649899	7.328552
H	2.972759	-1.706302	7.577936
C	1.841041	0.123991	7.712363
H	1.020662	-0.331292	8.259564
C	1.806584	1.479638	7.402655

H	0.968037	2.095163	7.713543
C	2.856824	2.063413	6.696234
H	2.826748	3.123704	6.466939
C	6.735669	1.734600	6.584370
C	7.862099	0.982956	6.234726
H	7.942689	0.545367	5.243292
C	8.881528	0.768359	7.161118
H	9.745534	0.172087	6.881507
C	8.785070	1.303026	8.442441
H	9.577452	1.131514	9.165171
C	7.662228	2.048607	8.798855
H	7.574990	2.462264	9.799209
C	6.642207	2.262153	7.877963
H	5.767847	2.833853	8.174007
C	3.536804	3.555233	3.868772
C	7.959078	5.985719	4.495092
H	8.804574	6.371619	5.071282
C	6.141411	6.915588	2.984145
H	5.894893	7.878507	2.530247
C	5.141130	5.668860	7.795259
C	6.998880	8.660128	7.951132
C	7.194693	9.890535	8.596680
H	6.546504	10.734756	8.375888
C	8.230056	10.048888	9.511318
H	8.370710	11.004612	10.007736
C	9.085294	8.982385	9.790388
H	9.891318	9.107597	10.507477
C	8.902728	7.759659	9.151818
H	9.563196	6.925575	9.370108
C	7.862916	7.596786	8.237282
H	7.712046	6.635666	7.752100
C	5.953286	9.683545	5.471390
C	7.204709	10.296266	5.302307
H	7.993898	10.126546	6.029708
C	7.438338	11.143807	4.219152
H	8.408436	11.621336	4.111468
C	6.428095	11.397061	3.293397
H	6.608730	12.065828	2.457075
C	5.179789	10.794466	3.453354
H	4.383056	10.992925	2.742010
C	4.945375	9.937688	4.525111
H	3.973130	9.461079	4.624546
C	4.196058	9.097589	7.630808
H	4.318555	10.017016	8.201764
C	3.014931	8.476823	7.561617
H	2.141132	8.864954	8.084397
C	1.744975	7.435301	5.225799
C	1.273509	8.747611	5.099095
H	1.554952	9.504070	5.825510
C	0.434711	9.099953	4.043318

H	0.074048	10.121463	3.962972
C	0.049314	8.145922	3.105762
H	-0.611907	8.419911	2.288920
C	0.514294	6.837640	3.221803
H	0.220474	6.084914	2.496111
C	1.363259	6.486920	4.267264
H	1.723492	5.465469	4.333897
C	1.736528	5.982791	7.717835
C	0.555302	5.373788	7.280024
H	0.246898	5.455378	6.243245
C	-0.257478	4.683924	8.178049
H	-1.178722	4.229835	7.823780
C	0.096511	4.593930	9.521878
H	-0.542632	4.062738	10.221082
C	1.269549	5.199496	9.966856
H	1.551465	5.142211	11.014105
C	2.084113	5.887701	9.073149
H	2.990576	6.358589	9.440906
H	8.330678	5.417818	3.640391
H	6.532549	6.234566	2.223817
H	4.368327	4.547405	5.744094
H	7.924212	7.838726	3.535297

Table S33: Cartesian coordinates of the optimized structure of *unsym*-[μ**H**-($Fe^{II}Fe^{II}$)adt-**H**]$^{2+}$ (–4788.98064496 E_h)



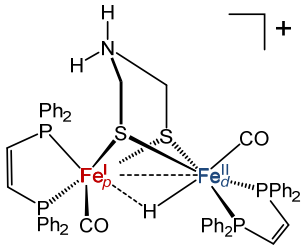
unsym-[μ**H**-($Fe^{II}Fe^{II}$)adt-**H**]$^{2+}$

atom	x	y	z
Fe	4.583752	13.791232	3.803022
Fe	5.854101	11.434970	3.765144
H	5.334713	12.628474	4.814009
S	3.691159	11.840958	3.006981
S	6.381047	13.287249	2.513582
P	2.826438	14.263989	5.185646
P	5.161437	9.861259	5.270622
P	5.619423	15.551124	4.814014
P	6.663693	9.696406	2.525023
O	3.387857	15.424157	1.739331
O	8.486291	11.447130	5.008086
C	3.560275	12.038996	1.206740
C	5.839739	13.162833	0.784051
C	7.434685	11.455439	4.541021
C	3.849482	14.830742	2.618650
C	6.416649	16.799784	3.734550
C	5.587183	17.631649	2.967987
H	4.506206	17.552481	3.043244
C	6.137805	18.589671	2.122510
H	5.484183	19.237406	1.546106
C	7.521447	18.725352	2.027994
H	7.951079	19.476688	1.372530
C	8.350471	17.906977	2.790557
H	9.428874	18.021139	2.735655
C	7.804480	16.949859	3.642691
H	8.466564	16.344459	4.252039
C	6.812082	15.233771	6.161100
C	6.593915	15.708390	7.460302
H	5.696092	16.269474	7.699536
C	7.530923	15.465797	8.462552
H	7.355440	15.848475	9.463452
C	8.692054	14.750112	8.179953
H	9.427998	14.576253	8.959284
C	8.911820	14.264255	6.891518
H	9.812748	13.702586	6.664832
C	7.975224	14.500822	5.889339
H	8.159286	14.121172	4.888282
C	1.213233	14.289598	4.335213
C	0.665352	15.490920	3.869909
H	1.172550	16.434831	4.049451
C	-0.540402	15.490188	3.171790
H	-0.959717	16.428748	2.822551
C	-1.207562	14.292154	2.931452
H	-2.151345	14.294007	2.394668
C	-0.665670	13.091073	3.388119
H	-1.188366	12.155938	3.209783

C	0.540671	13.084532	4.082036
H	0.948667	12.143158	4.440035
C	2.500967	13.488192	6.811237
C	3.594323	13.163559	7.623253
H	4.606431	13.296982	7.251686
C	3.391173	12.668200	8.908123
H	4.246025	12.406954	9.523659
C	2.095941	12.497634	9.393922
H	1.937501	12.111905	10.396441
C	1.004016	12.829893	8.594478
H	-0.006380	12.712261	8.974862
C	1.201147	13.326535	7.307921
H	0.343493	13.590819	6.698027
C	8.391825	9.756316	1.929244
C	9.057328	8.569863	1.579594
H	8.556309	7.609077	1.651961
C	10.367907	8.608597	1.116925
H	10.873498	7.683878	0.856595
C	11.029570	9.829535	0.989193
H	12.053991	9.856468	0.630856
C	10.375062	11.011500	1.322900
H	10.887178	11.964088	1.227003
C	9.063789	10.976884	1.792907
H	8.570887	11.905206	2.065806
C	5.731165	9.126385	1.034660
C	6.320748	9.072692	-0.239044
H	7.366478	9.335500	-0.366954
C	5.574561	8.658865	-1.343376
H	6.045269	8.612190	-2.320896
C	4.242530	8.282136	-1.189294
H	3.669606	7.944780	-2.047693
C	3.648715	8.328427	0.073465
H	2.614007	8.023409	0.199974
C	4.382222	8.755097	1.176744
H	3.904692	8.800386	2.152091
C	3.416869	9.302137	5.325839
C	2.492165	10.001142	6.111093
H	2.810496	10.849965	6.707294
C	1.163042	9.589238	6.159490
H	0.460341	10.133453	6.783047
C	0.743051	8.476718	5.432277
H	-0.291440	8.150767	5.482836
C	1.660787	7.768757	4.660661
H	1.347489	6.883584	4.115047
C	5.619190	10.022118	7.034312
C	5.264695	8.993162	7.920648
H	4.659416	8.157975	7.578206
C	5.672776	9.039121	9.248611
H	5.397018	8.235639	9.924676
C	6.437208	10.111625	9.708427

H	6.763431	10.140344	10.743770
C	6.778588	11.144252	8.839192
H	7.366633	11.987009	9.189801
C	6.368568	11.103066	7.507733
H	6.631730	11.924334	6.850463
C	6.013788	8.318245	4.791646
H	5.986763	7.471753	5.476842
C	6.634184	8.235144	3.612569
H	7.126535	7.318472	3.294490
C	4.300749	16.527176	5.606738
H	4.527192	17.529228	5.968724
C	3.083687	15.989016	5.726256
H	2.267873	16.514482	6.221668
C	2.991593	8.177773	4.606540
H	3.701213	7.594093	4.027139
N	4.871617	12.058368	0.476024
H	5.325456	11.143704	0.648996
H	3.015878	11.186975	0.794148
H	3.030167	12.960755	0.959708
H	5.371373	14.100223	0.475230
H	6.714235	12.976383	0.155199
H	4.675439	12.081512	-0.531658

Table S34: Cartesian coordinates of the optimized structure of *unsym*-[μ**H**-($Fe^{I}Fe^{II}$)adt-**H**]$^{+}$ (–4789.23512869 E_h)



unsym-[μ**H**-($Fe^{I}Fe^{II}$)adt-**H**]$^{+}$

atom	x	y	z
Fe	4.566097	13.944536	3.859759
Fe	5.939545	11.395469	3.778873
H	5.215482	12.989288	4.932288
S	3.753071	11.916881	3.089418
S	6.475588	13.402288	2.683760
P	2.795735	14.361583	5.155704
P	5.264791	9.805047	5.238033
P	5.525443	15.687597	4.869542
P	6.693127	9.580286	2.417726
O	3.438811	15.519064	1.677341
O	8.588264	11.390424	4.982905
C	3.726451	12.077069	1.281735
C	5.969557	13.276063	0.945231
C	7.517491	11.390493	4.530620
C	3.876050	14.927459	2.576694
C	6.345866	16.952713	3.822678
C	5.546594	17.819534	3.065515
H	4.463369	17.763180	3.132969
C	6.129761	18.765917	2.228408
H	5.498636	19.438503	1.654781
C	7.517082	18.852079	2.131228
H	7.972379	19.592040	1.479544
C	8.317466	17.992369	2.879079
H	9.399567	18.060118	2.814218
C	7.737533	17.048181	3.722915
H	8.373366	16.396398	4.313710
C	6.716219	15.411391	6.234726
C	6.622005	16.081882	7.459658
H	5.817300	16.786954	7.641846
C	7.556062	15.844470	8.465524
H	7.469769	16.370052	9.412098
C	8.597524	14.943031	8.257899
H	9.329169	14.766676	9.041162
C	8.699189	14.269936	7.041391
H	9.501964	13.559372	6.869126
C	7.760440	14.499294	6.038938
H	7.841649	13.962441	5.099505
C	1.174686	14.356754	4.310749
C	0.613750	15.542920	3.824363
H	1.110628	16.494026	3.995655
C	-0.583453	15.517969	3.112344
H	-1.009970	16.446615	2.744703
C	-1.230347	14.308083	2.876529
H	-2.165991	14.289500	2.325356
C	-0.674724	13.120950	3.351497
H	-1.176102	12.174326	3.171127

C	0.522800	13.140563	4.060367
H	0.947673	12.208682	4.422806
C	2.466425	13.547556	6.768823
C	3.559333	13.143781	7.544392
H	4.564699	13.235337	7.143078
C	3.361396	12.609124	8.814760
H	4.217456	12.282303	9.396903
C	2.070777	12.474145	9.322192
H	1.916112	12.051904	10.310836
C	0.978380	12.881189	8.558998
H	-0.029301	12.784909	8.953273
C	1.172206	13.419699	7.288950
H	0.314631	13.738071	6.704536
C	8.404835	9.570515	1.757504
C	9.035074	8.392239	1.329913
H	8.505105	7.443886	1.361511
C	10.337272	8.426052	0.843072
H	10.816126	7.506946	0.518249
C	11.026060	9.637116	0.772519
H	12.044045	9.660683	0.394874
C	10.408700	10.812380	1.189290
H	10.943656	11.756395	1.142129
C	9.105292	10.779756	1.682595
H	8.634657	11.696381	2.029026
C	5.710559	8.927748	0.990982
C	6.204842	8.889170	-0.321422
H	7.247059	9.125361	-0.515796
C	5.373945	8.526154	-1.381659
H	5.777117	8.490385	-2.389958
C	4.041838	8.191178	-1.149780
H	3.400243	7.898448	-1.975564
C	3.538730	8.229552	0.150895
H	2.502409	7.965991	0.343110
C	4.360243	8.606124	1.209902
H	3.945970	8.657744	2.213709
C	3.521543	9.214817	5.302497
C	2.570383	9.982149	5.986319
H	2.870502	10.891118	6.497901
C	1.240172	9.576642	6.036140
H	0.520419	10.180344	6.581448
C	0.838736	8.400595	5.403791
H	-0.198738	8.082269	5.449143
C	1.778256	7.627714	4.728327
H	1.479503	6.699639	4.249013
C	5.686062	9.920659	7.024141
C	5.321904	8.893172	7.905916
H	4.736575	8.049938	7.547984
C	5.690125	8.950402	9.245553
H	5.405309	8.146534	9.918149
C	6.423581	10.036747	9.723153

H	6.715968	10.076925	10.768581
C	6.776756	11.068625	8.857863
H	7.341620	11.922676	9.220465
C	6.406762	11.012676	7.514768
H	6.678043	11.827162	6.851590
C	6.127963	8.259520	4.762975
H	6.146462	7.428418	5.467853
C	6.710828	8.154951	3.565151
H	7.217269	7.238270	3.265742
C	4.185234	16.654540	5.650453
H	4.379129	17.657930	6.027034
C	2.978928	16.086292	5.742725
H	2.140739	16.584064	6.229570
C	3.112503	8.030500	4.678851
H	3.836926	7.404533	4.165542
N	5.108952	12.072830	0.693479
H	5.593217	11.272035	1.143025
H	3.224479	11.209031	0.848109
H	3.229851	12.994222	0.957570
H	5.414941	14.157366	0.615067
H	6.856271	13.145846	0.318788
H	5.055367	11.878758	-0.311669

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