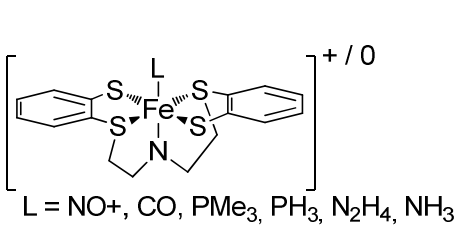
# Predicting the spin state of paramagnetic iron complexes by DFT calculation of proton NMR spectra

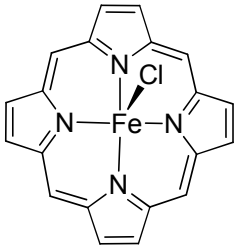
Andrea Borgogno, Federico Rastrelli and Alessandro Bagno*
*Dipartimento di Scienze Chimiche, Università di Padova, via Marzolo, 1 - 35131 Padova (Italy)*
E-mail: alessandro.bagno@unipd.it

## Electronic Supplementary Information

**Scheme S1**




**1-6**

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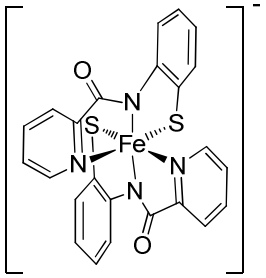


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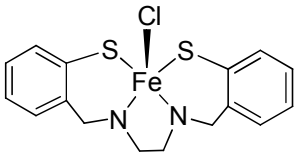


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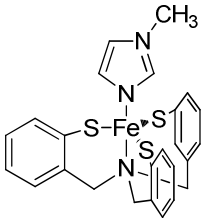


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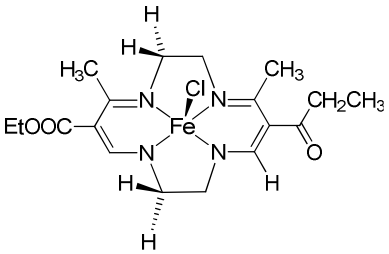


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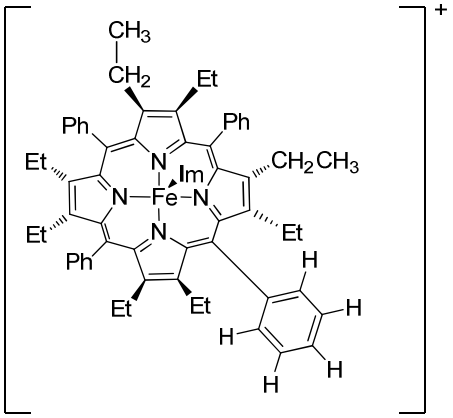


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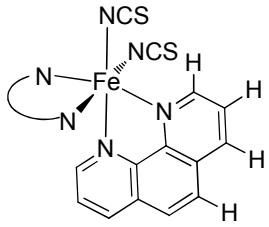
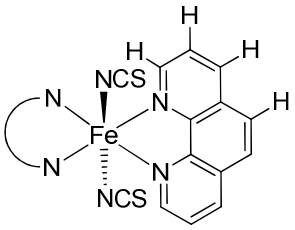
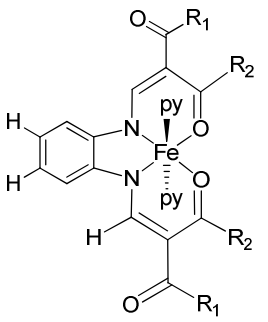



**13a**

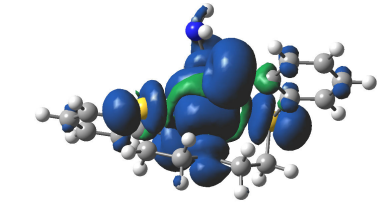



**13b**




**14-16**

**Scheme S1.** Structures of complexes **1**-**16**. **1**-**6**: $[Fe^{II}(N_HS_4)(L)]$. L = $NO^+$ (**1**), CO (**2**), $PMe_3$ (**3**), $PH_3$ (**4**), $N_2H_4$ (**5**), $NH_3$ (**6**), $N_HS_4$ = 2,2'-bis(2-mercaptophenylthio)diethylamine dianion. **7**: $[Fe^{III}(por)Cl]$. por: porphyrinate. **8**: $[Fe^{III}(pyPepS)_2]^-$. pyPepS = *N*-(2-mercaptophenyl)-2-pyridinecarboxamide). **9**: $[Fe^{III}(tsalen)Cl]$. tsalen = *N*,*N*'-ethylenebis-(thiosalicylideneiminato). **10**: $[Fe^{III}(N(CH_2$*o*$-C_6H_4S)_3(1\text{-MeIm})]$. $N(CH_2$*o*$-C_6H_4S)_3(1\text{-MeIm})$ = 2.2'.2"-(nitrilo-tris-(methylene))-tri-benzenethiolato; 1-MeIm = 1-methylimidazole. **11**: $[Fe^{III}(L)Cl]$. L = diethyl 5,14-dimethyl-1,4,8,11-tetraazacyclotetradeca-4,7,11,14-tetraene-6,13-dicarboxylato. **12**: $[Fe^{III}(oetpp)(HIm)]^+$. oetpp = [2,3,7,8,12,13,17,18-octaethyl-5,10,15,20-tetraphenyl-21H,23H-porphinato; HIm = imidazole. **13**: $[Fe^{II}(phen)_2(NCS)_2]$. phen: 1.10-phenanthroline. **14**-**16**: $[Fe^{II}(L1R_1R_2)(py)_2]$. L1 = *N*,*N*'-*o*-phenylenebis(R,R'-vinylamido). **14**: $R_1$ = OEt. $R_2$ = $CH_3$; **15**: $R_1$ = $CH_3$. $R_2$ = $CH_3$; **16**: $R_1$ = OEt. $R_2$ = OEt.



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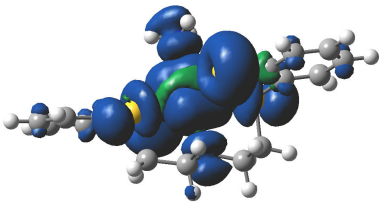

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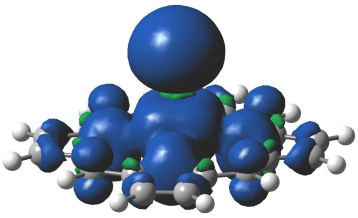

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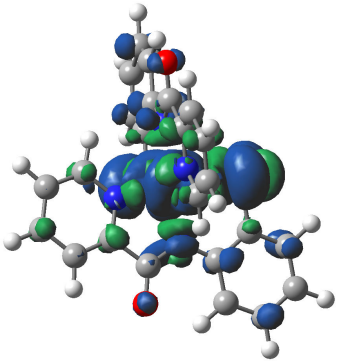

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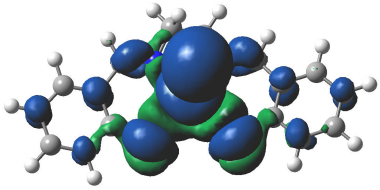

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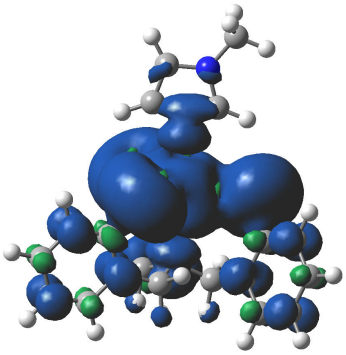

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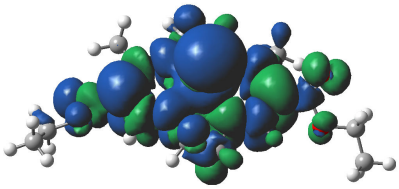

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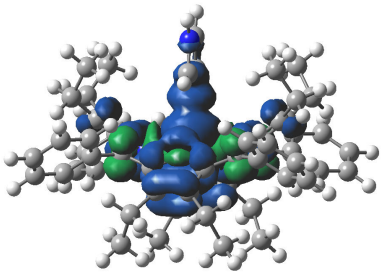

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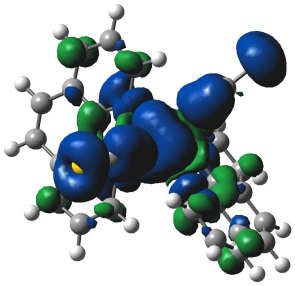
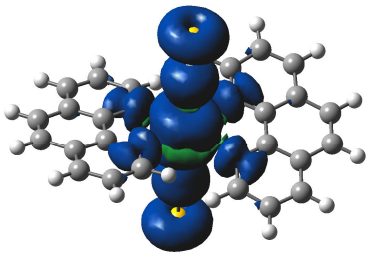
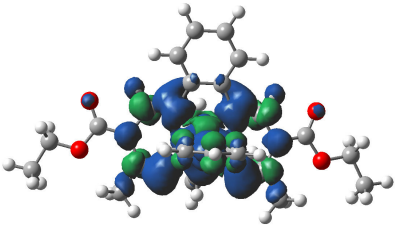


**13a**



**13b**



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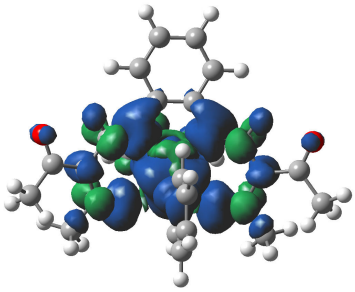

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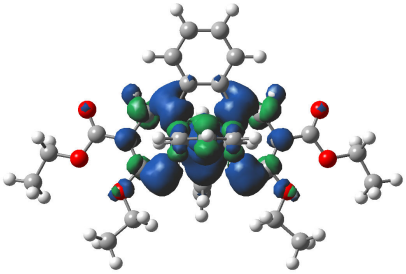


**16**

**Figure S1.** Isosurfaces (isovalue $4 \times 10^{-4}$) of the spin densities for **5**-**16** in the most stable spin state (see Table 1).

**Table S1.** Summary of relative energies of spin states of iron complexes at the B3LYP*/cc-pVTZ level.[a]

| | $\Delta E$ [b] | $\Delta E_{tot}$ [b,c] | $\Delta E_{tot}$ [d] | | $\Delta E$ [b] | $\Delta E_{tot}$ [b,c] | $\Delta E_{tot}$ [d] |
|---|---|---|---|---|---|---|---|
| $[Fe^{II}(N_HS_4)(NO)]^+$ (**1**) | | | | $[Fe^{III}(N(CH_2$*o*$-C_6H_4S)_3(1\text{-MeIm})]$ (**10**) | | | |
| ***S* = 0** | (0.0) | (0.0) | (0.0) | *S* = 1/2 | (0.0) | (0.0) | (0.0) |
| *S* = 1 | 32.2 | 26.4 | 35.1 | *S* = 3/2 | –5.0 | –7.1 | –6.7 |
| *S* = 2 | 72.0 | 60.7 | 64.9 | ***S* = 5/2** | –15.9 | –20.9 | –20.5 |
| $[Fe^{II}(N_HS_4)(CO)]$ (**2**) | | | | $[Fe^{III}(L)Cl]$ (**11**) | | | |
| ***S* = 0** | (0.0) | (0.0) | (0.0) | *S* = 1/2 | (0.0) | (0.0) | (0.0) |
| *S* = 1 | 52.7 | 49.4 | 57.3 | *S* = 3/2 | –37.7 | –38.9 | –46.4 |
| *S* = 2 | 73.2 | 65.3 | 64.0 | *S* = 5/2 | 10.5 | 4.2 | 10.5 |
| $[Fe^{II}(N_HS_4)(PMe_3)]$ (**3**) | | | | $[Fe^{III}(oetpp)(HIm)]^+$ (**12**) [e] | | | |
| ***S* = 0** | (0.0) | (0.0) | (0.0) | ***S* = 3/2** | (0.0) | (0.0) | (0.0) |
| *S* = 1 | 36.4 | 30.5 | 36.0 | *S* = 5/2 | 46.4 | 43.5 | 46.9 |
| *S* = 2 | 17.9 | 12.1 | 8.4 | *cis*-$[Fe^{II}(phen)_2(NCS)_2]$ (**13a**) | | | |
| $[Fe^{II}(N_HS_4)(PH_3)]$ (**4**) | | | | *S* = 0 | (0.0) | (0.0) | (0.0) |
| ***S* = 0** | (0.0) | (0.0) | (0.0) | S = 1 | 44.8 | 43.5 | |
| *S* = 1 | 38.9 | 35.6 | 25.5 | ***S* = 2** | 2.1 | –7.5 | –5.0 |
| *S* = 2 | 17.6 | 9.6 | 5.9 | *trans*-$[Fe^{II}(phen)_2(NCS)_2]$ (**13b**) | | | |
| $[Fe^{II}(N_HS_4)(N_2H_4)]$ (**5**) | | | | *S* = 0 | (0.0) | (0.0) | |
| *S* = 0 | (0.0) | (0.0) | (0.0) | *S* = 1 | 8.8 | 3.8 | |
| *S* = 1 | 35.6 | 32.6 | 16.7 | ***S* = 2** | –25.9 | –35.1 | |
| ***S* = 2** | 2.5 | –4.2 | -12.1 | $[Fe^{II}(L1R_1R_2)(py)_2]$ $R_1$ = OEt, $R_2$ = $CH_3$ (**14**) | | | |
| $[Fe^{II}(N_HS_4)(NH_3)]$ (**6**) | | | | *S* = 0 | (0.0) | (0.0) | |
| *S* = 0 | (0.0) | (0.0) | (0.0) | ***S* = 2** | 13.8 | 2.9 | |
| *S* = 1 | 27.2 | 19.7 | 19.7 | $[Fe^{II}(L1R_1R_2)(py)_2]$ $R_1$, $R_2 = CH_3$ (**15**) | | | |
| ***S* = 2** | 0.8 | -6.7 | -10.5 | *S* = 0 | (0.0) | (0.0) | |
| $[Fe^{III}(por)Cl]$ (**7**) | | | | ***S* = 2** | 14.2 | 3.3 | |
| *S* = 3/2 | (0.0) | (0.0) | (0.0) | $[Fe^{II}(L1R_1R_2)(py)_2]$ $R_1$, $R_2$ = OEt (**16**) | | | |
| ***S* = 5/2** | 9.2 | 5.4 | 1.7 | *S* = 0 | (0.0) | (0.0) | |
| $[Fe^{III}(pyPepS)_2]^-$ (**8**) | | | | ***S* = 2** | 10.5 | –0.8 | |
| ***S* = 1/2** | (0.0) | (0.0) | (0.0) | | | | |
| *S* = 3/2 | 79.5 | 76.1 | 39.7 | | | | |
| *S* = 5/2 | 34.7 | 29.7 | 20.9 | | | | |
| $[Fe^{III}(tsalen)Cl]$ (**9**) | | | | | | | |
| *S* = 1/2 | (0.0) | (0.0) | (0.0) | | | | |
| ***S* = 3/2** | –37.2 | –38.9 | –49.0 | | | | |
| *S* = 5/2 | –15.1 | –19.7 | –28.0 | | | | |

[a]Relative energies with respect to lowest *S* value (*S* = 0 for Fe(II), *S* = 1/2 for Fe(III)) in kJ $mol^{-1}$. See Tables S2-S3 for results with other density functionals and Scheme 1 for structures. The known ground-state multiplicity is highlighted in boldface. [b]B3LYP*/cc-pVTZ//B3LYP*/6-31G(d,p) calculations. [c]Including zero-point vibrational energy. [d]B3LYP*/TZ2P//OPBE/TZP calculations, without vibrational corrections. [e]SCF convergence could not be achieved for the *S* = 1/2 state; however, according to the OPBE calculation this state is higher in energy by ca. 40 kJ/mol (Table S3).

**Table S2.** Relative energies (with respect to lowest *S* value) of spin states of iron complexes with gaussian basis sets. The known ground-state multiplicity is highlighted in boldface. Energy and geometry calculated with the cc-pVTZ and 6-31G(d,p) basis sets, respectively, with the given functional.

| Complex | E (au) | ΔE (au) | ZPE (Joule) | ΔE tot (kJ/mol) | E (au) | ΔE (au) | ZPE (Joule) | ΔE tot (kJ/mol) | E (au) | ΔE (au) | ZPE (Joule) | ΔE tot (kJ/mol) |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | **B3LYP** | | | | **B3LYP*** | | | | **X3LYP** | | | |
| **1** | | | | | | | | | | | | |
| **S=0** | -3661.45 | 0.00 | 817583.30 | 0.00 | -3648.26 | 0.00 | 805094.20 | 0.00 | -3660.81 | 0.00 | 819455.50 | 0.0 |
| S=1 | -3661.44 | 3.56 | 811467.90 | 8.8 | -3648.25 | 7.68 | 799297.50 | 26.4 | -3660.80 | 2.23 | 813172.20 | 37.7 |
| S=2 | -3661.43 | 13.03 | 805595.10 | 42.7 | -3648.24 | 17.17 | 793856.90 | 60.7 | -3660.79 | 12.03 | 807089.90 | 25.5 |
| **2** | | | | | | | | | | | | |
| **S=0** | -3645.09 | 0.00 | 813356.90 | 0.00 | -3631.95 | 0.00 | 801397.80 | 0.00 | -3644.45 | 0.00 | 815069.30 | 22.2 |
| S=1 | -3645.07 | 10.55 | 809639.00 | 40.6 | -3631.93 | 12.64 | 797963.80 | 49.4 | -3644.44 | 9.88 | 811425.00 | 44.4 |
| S=2 | -3645.07 | 10.42 | 803766.90 | 33.9 | -3631.92 | 17.48 | 793311.20 | 65.3 | -3644.44 | 8.47 | 805566.60 | 0.0 |
| **3** | | | | | | | | | | | | |
| **S=0** | -3992.90 | 0.00 | 1093129.10 | 15.5 | -3978.36 | 0.00 | 1080219.30 | 0.00 | -3992.19 | 3.57 | 1095275.60 | 24.7 |
| S=1 | -3992.89 | 7.21 | 1086506.30 | 39.3 | -3978.35 | 8.72 | 1074359.80 | 30.5 | -3992.18 | 10.50 | 1088624.50 | 48.5 |
| S=2 | -3992.90 | -2.05 | 1086063.00 | 0.00 | -3978.35 | 4.33 | 1074014.80 | 12.1 | -3992.20 | 0.00 | 1088136.20 | 0.0 |
| **4** | | | | | | | | | | | | |
| **S=0** | -3874.89 | 2.17 | 863214.20 | 0.00 | -3861.23 | 0.00 | 851144.80 | 0.00 | -3874.24 | 3.81 | 864999.90 | 37.7 |
| S=1 | -3874.88 | 9.22 | 860111.20 | 44.4 | -3861.22 | 9.27 | 848045.80 | 35.6 | -3874.23 | 10.22 | 861988.90 | 70.7 |
| S=2 | -3874.89 | 0.00 | 854515.80 | 17.9 | -3861.23 | 4.20 | 843185.80 | 9.6 | -3874.25 | 0.00 | 856311.50 | 0.0 |
| **5** | | | | | | | | | | | | |
| S=0 | -3643.64 | 5.62 | 940935.40 | 31.8 | -3630.43 | 0.00 | 927543.30 | 4.2 | -3642.99 | 7.01 | 943029.40 | 38.9 |
| S=1 | -3643.62 | 14.80 | 937224.40 | 66.5 | -3630.42 | 8.53 | 924198.60 | 36.8 | -3642.98 | 15.82 | 939226.60 | 51.0 |
| **S=2** | -3643.64 | 0.00 | 932670.10 | 0.00 | -3630.43 | 0.60 | 920684.00 | 0.0 | -3643.00 | 0.00 | 934768.00 | 0.0 |
| **6** | | | | | | | | | | | | |
| S=0 | -3588.30 | 5.92 | 893224.40 | 33.1 | -3575.44 | 0.00 | 881844.30 | 6.7 | -3587.68 | 7.33 | 895109.00 | 13.0 |
| S=1 | -3588.29 | 10.99 | 884947.30 | 46.0 | -3575.43 | 6.51 | 874627.60 | 26.4 | -3587.67 | 12.19 | 886998.40 | 0.0 |
| **S=2** | -3588.31 | 0.00 | 885064.70 | 0.00 | -3575.44 | 0.20 | 874491.80 | 0.0 | -3587.69 | 0.00 | 887071.00 | 0.0 |
| **7** | | | | | | | | | | | | |
| S=3/2 | -2712.83 | 1.32 | 727185.30 | 9.2 | -2702.04 | -2.17 | 715172.30 | 0.0 | -2712.26 | 2.18 | 728958.80 | 27.2 |
| **S=5/2** | -2712.84 | 0.00 | 723449.00 | 0.00 | -2702.04 | 0.00 | 711386.70 | 5.4 | -2712.26 | 0.00 | 725051.20 | 0.8 |
| **8** | | | | | | | | | | | | |
| **S=1/2** | -3354.56 | 0.00 | 931854.10 | 0.00 | -3340.72 | 0.00 | 917103.40 | 0.0 | -3353.82 | 0.00 | 934200.20 | 51.0 |
| S=3/2 | -3354.55 | 8.09 | 927805.70 | 29.7 | -3340.69 | 19.03 | 913448.20 | 76.1 | -3353.81 | 7.49 | 930166.00 | 0.0 |
| S=5/2 | -3354.56 | 2.81 | 926705.60 | 6.7 | -3340.71 | 8.27 | 912261.80 | 29.7 | -3353.82 | 1.44 | 929074.80 | 6.3 |
| **9** | | | | | | | | | | | | |
| S=1/2 | -3248.46 | 11.64 | 707649.90 | 50.6 | -3236.84 | 8.89 | 696891.10 | 38.9 | -3247.90 | 11.81 | 708821.30 | 37.7 |
| **S=3/2** | -3248.48 | 0.00 | 705767.50 | 0.00 | -3236.86 | 0.00 | 695203.10 | 0.0 | -3247.92 | 0.00 | 707316.50 | 25.5 |
| S=5/2 | -3248.48 | 2.81 | 709284.80 | 15.1 | -3236.85 | 5.31 | 692613.30 | 19.2 | -3247.91 | 2.14 | 704836.00 | 0.0 |

**Table S2 (continued).**

| Complex | E (au) | ΔE (au) | ZPE (Joule) | ΔE tot (kJ/mol) | E (au) | ΔE (au) | ZPE (Joule) | ΔE tot (kJ/mol) | E (au) | ΔE (au) | ZPE (Joule) | ΔE tot (kJ/mol) |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | **B3LYP** | | | | **B3LYP*** | | | | **X3LYP** | | | |
| **10** | | | | | | | | | | | | |
| S=1/2 | -3590.30 | 9.91 | 1161602.00 | 46.4 | -3575.96 | 3.82 | 1145348.70 | 20.9 | -3589.54 | 10.57 | 1164185.70 | 49.4 |
| S=3/2 | -3590.30 | 5.39 | 1160094.70 | 25.9 | -3575.97 | 2.58 | 1143867.40 | 13.8 | -3589.55 | 5.95 | 1162647.50 | 28.5 |
| **S=5/2** | -3590.31 | 0.00 | 1156564.40 | 0.0 | -3575.97 | 0.00 | 1140575.50 | 0.0 | -3589.56 | 0.00 | 1159115.80 | 0.0 |
| **11** | | | | | | | | | | | | |
| S=1/2 | | | | | -2933.91 | 9.10 | 1141584.40 | 38.8 | | | | |
| **S=3/2** | | | | | -2933.92 | 0.00 | 1140876.20 | 0.0 | | | | |
| S=5/2 | | | | | -2.933.90 | 11.51 | 1135715.40 | 43.0 | | | | |
| **12** | | | | | | | | | | | | |
| **S=3/2** | | | | | -4010.29 | 0.00 | 2914927.40 | 0.0 | | | | |
| S=5/2 | | | | | -4010.27 | 11.09 | 2912188.10 | 43.5 | | | | |
| **13a** | | | | | | | | | | | | |
| **S=0** | -3389.76 | 6.42 | 971295.00 | 37.2 | -3375.57 | 0.00 | 956899.10 | 7.5 | -3388.99 | 7.72 | 973541.40 | 42.3 |
| S=1 | | | | 0.0 | -3375.55 | 10.67 | 951481.90 | 46.7 | | | | 0.0 |
| S=2 | -3389.77 | 0.00 | 961081.60 | 0.0 | -3375.57 | 0.45 | 947524.30 | 0.0 | -3389.00 | 0.00 | 963391.30 | 0.0 |
| **13b** | | | | | | | | | | | | |
| **S=0** | | | | | -3375.55 | 6.18 | 957253.90 | 35.3 | | | | |
| S=1 | | | | | -3375.55 | 8.28 | 952104.10 | 39.0 | | | | |
| S=2 | | | | | -3375.56 | 0.00 | 947792.30 | 0.0 | | | | |
| **14** | | | | | | | | | | | | |
| S=0 | -3097.40 | 3.63 | 1528838.50 | 27.2 | -3082.59 | 0.00 | 1510976.50 | 0.0 | | | | |
| **S=2** | -3097.41 | 0.00 | 1516497.90 | 0.0 | -3082.59 | 3.31 | 1500123.20 | 2.9 | | | | |
| **15** | | | | | | | | | | | | |
| S=0 | -2868.21 | 3.65 | 1351221.90 | 25.9 | -2854.81 | 0.00 | 1336481.30 | 0.0 | | | | |
| **S=2** | -2868.21 | 0.00 | 1340336.20 | 0.0 | -2854.80 | 3.35 | 1325952.60 | 3.3 | | | | |
| **16** | | | | | | | | | | | | |
| S=0 | -3326.57 | 4.47 | 1702990.40 | 30.5 | -3310.35 | 0.00 | 1683134.00 | 0.8 | | | | |
| **S=2** | -3326.58 | 0.00 | 1691093.50 | 0.0 | -3310.34 | 2.43 | 1672189.30 | 0.0 | | | | |

**Table S2 (continued).**

| Complex | E (au) | ΔE (au) | ZPE (Joule) | ΔE tot (kJ/mol) | E (au) | ΔE (au) | ZPE (Joule) | ΔE tot (kJ/mol) |
|---|---|---|---|---|---|---|---|---|
| | **PBE0** | | | | **M06** | | | |
| **1** | | | | | | | | |
| **S=0** | -3659.64 | 0.00 | 825946.30 | 0.0 | -3660.63 | 0.00 | 813583.00 | 0.0 |
| S=1 | -3659.63 | 2.05 | 819050.50 | 1.7 | -3660.62 | 7.70 | 807927.40 | 26.4 |
| S=2 | -3659.62 | 9.44 | 812940.00 | 26.4 | -3660.63 | 3.52 | 801943.50 | 2.9 |
| **2** | | | | | | | | |
| **S=0** | -3643.29 | 0.00 | 821093.90 | 0.0 | -3644.28 | 0.00 | 808775.10 | 0.6 |
| S=1 | -3643.27 | 10.36 | 816870.70 | 39.3 | -3644.27 | 7.37 | 806422.40 | 29.3 |
| S=2 | -3643.28 | 7.27 | 810795.20 | 20.1 | -3644.27 | 2.11 | 799306.70 | 0.0 |
| **3** | | | | | | | | |
| **S=0** | -3990.93 | 4.89 | 1102246.40 | 27.6 | -3992.02 | 7.04 | 1088534.20 | 34.7 |
| S=1 | -3990.92 | 12.47 | 1095448.40 | 52.7 | -3992.01 | 13.74 | 1081765.40 | 56.1 |
| S=2 | -3990.94 | 0.00 | 1094933.10 | 0.0 | -3992.03 | 0.00 | 1083195.20 | 0.0 |
| **4** | | | | | | | | |
| **S=0** | -3873.06 | 5.09 | 871320.10 | 30.5 | -3874.10 | 8.47 | 859716.50 | 43.9 |
| S=1 | -3873.05 | 12.02 | 867515.60 | 52.7 | -3874.09 | 15.24 | 856618.90 | 69.0 |
| S=2 | -3873.07 | 0.00 | 862103.30 | 0.0 | -3874.12 | 0.00 | 851269.30 | 0.0 |
| **5** | | | | | | | | |
| S=0 | -3641.83 | 10.25 | 950110.00 | 51.5 | -3642.81 | 13.36 | 936367.30 | 63.2 |
| S=1 | -3641.82 | 18.49 | 945680.40 | 81.2 | -3642.80 | 22.32 | 933782.70 | 97.9 |
| **S=2** | -3641.85 | 0.00 | 941690.40 | 0.0 | -3642.83 | 0.00 | 929092.80 | 0.0 |
| **6** | | | | | | | | |
| S=0 | -3586.56 | 10.62 | 901936.20 | 52.7 | -3587.51 | 14.30 | 889628.80 | 66.5 |
| S=1 | -3586.55 | 14.50 | 893685.50 | 60.7 | -3587.51 | 15.79 | 881290.90 | 64.4 |
| **S=2** | -3586.58 | 0.00 | 893704.60 | 0.0 | -3587.53 | 0.00 | 882778.60 | 0.0 |
| **7** | | | | | | | | |
| S=3/2 | -2711.26 | 5.96 | 734478.80 | 28.5 | -2711.97 | 13.17 | 725879.80 | 58.2 |
| **S=5/2** | -2711.27 | 0.00 | 730830.90 | 0.0 | -2712.00 | 0.00 | 722636.10 | 0.0 |
| **8** | | | | | | | | |
| **S=1/2** | -3352.50 | 2.10 | 941386.20 | 13.8 | -3353.47 | 14.88 | 931263.90 | 68.6 |
| S=3/2 | -3352.49 | 8.66 | 937336.10 | 37.2 | -3353.47 | 15.62 | 925976.20 | 66.5 |
| S=5/2 | -3352.51 | 0.00 | 936249.30 | 0.0 | -3353.50 | 0.00 | 924991.00 | 0.0 |
| **9** | | | | | | | | |
| S=1/2 | -3246.86 | 16.42 | 713408.30 | 71.1 | -3247.70 | 28.42 | 701482.10 | 119.7 |
| **S=3/2** | -3246.89 | 0.00 | 711906.00 | 1.3 | -3247.74 | 5.58 | 702851.90 | 25.5 |
| S=5/2 | -3246.89 | 0.38 | 709284.80 | 0.0 | -3247.74 | 0.00 | 700541.30 | 0.0 |
| **10** | | | | | | | | |
| S=1/2 | -3588.24 | 15.10 | 1171480.20 | 68.2 | -3589.24 | 25.30 | 1157052.60 | 110.5 |
| S=3/2 | -3588.25 | 7.15 | 1169909.90 | 33.5 | -3589.26 | 14.90 | 1155263.20 | 65.3 |
| **S=5/2** | -3588.26 | 0.00 | 1166425.80 | 0.0 | -3589.28 | 0.00 | 1152481.60 | 0.0 |
| **13a** | | | | | | | | |
| **S=0** | -3387.63 | 11.46 | 980643.00 | 58.6 | -3388.61 | 16.30 | 969546.50 | 74.9 |
| S=2 | -3387.64 | 0.00 | 970041.90 | 0.0 | -3388.64 | 0.00 | 962712.00 | 0.0 |

**Table S2 (continued).**

| Complex | E (au) | ΔE (au) | ZPE (Joule) | ΔE tot (kJ/mol) | E (au) | ΔE (au) | ZPE (Joule) | ΔE tot (kJ/mol) |
|---|---|---|---|---|---|---|---|---|
| | **M06-2X** | | | | **M06-L** | | | |
| **1** | | | | | | | | |
| **S=0** | -3660.83 | 38.94 | 820653.50 | 170.7 | -3661.22 | 0.00 | 816056.40 | 0.0 |
| S=1 | -3660.86 | 17.66 | 820293.50 | 81.2 | -3661.20 | 12.02 | 808483.60 | 42.7 |
| S=2 | -3660.89 | 0.00 | 812796.30 | 0.0 | -3661.18 | 26.01 | 806547.70 | 99.2 |
| **2** | | | | | | | | |
| **S=0** | -3644.52 | 33.86 | 818947.40 | 149.0 | -3644.84 | 0.00 | 812186.10 | 0.0 |
| S=1 | -3644.53 | 26.90 | 816362.70 | 117.2 | -3644.81 | 15.41 | 808394.00 | 60.7 |
| S=2 | -3644.58 | 0.00 | 811781.00 | 0.0 | -3644.81 | 17.39 | 803484.10 | 64.0 |
| **3** | | | | | | | | |
| **S=0** | -3992.29 | 36.88 | 1101273.50 | 281.6 | -3992.62 | 0.00 | 1093423.50 | 0.0 |
| S=1 | -3992.30 | 33.50 | 1096433.40 | 262.3 | -3992.60 | 14.77 | 1092072.50 | 60.2 |
| S=2 | -3992.35 | 0.00 | 974206.00 | 0.0 | -3992.61 | 7.13 | 1086315.50 | 22.6 |
| **4** | | | | | | | | |
| **S=0** | -3874.33 | 39.66 | 869509.10 | 171.1 | -3874.63 | 0.00 | 863373.70 | 0.0 |
| S=1 | -3874.35 | 31.19 | 867989.90 | 134.3 | -3874.61 | 15.23 | 862321.00 | 62.8 |
| S=2 | -3874.40 | 0.00 | 864173.80 | 0.0 | -3874.62 | 5.33 | 855926.10 | 15.1 |
| **5** | | | | | | | | |
| S=0 | -3643.09 | 37.71 | 949805.70 | 164.0 | -3643.37 | 0.00 | 941201.80 | 6.7 |
| S=1 | -3643.09 | 37.05 | 948442.10 | 159.8 | -3643.35 | 14.03 | 936740.40 | 61.1 |
| **S=2** | -3643.15 | 0.00 | 943500.80 | 0.0 | -3643.37 | 0.36 | 932891.10 | 0.0 |
| **6** | | | | | | | | |
| S=0 | -3587.77 | 38.80 | 901701.00 | 169.9 | -3588.05 | 1.11 | 892288.90 | 10.0 |
| S=1 | -3587.78 | 34.31 | 894555.80 | 143.9 | -3588.03 | 9.37 | 884003.90 | 36.4 |
| **S=2** | -3587.84 | 0.00 | 894194.80 | 0.0 | -3588.05 | 0.00 | 886616.00 | 0.0 |
| **7** | | | | | | | | |
| S=3/2 | -2712.34 | 19.31 | 734086.10 | 83.3 | -2712.62 | 10.30 | 727066.20 | 45.6 |
| **S=5/2** | -2712.37 | 0.00 | 731360.20 | 0.0 | -2712.63 | 0.00 | 724732.70 | 0.0 |
| **8** | | | | | | | | |
| **S=1/2** | -3353.91 | 30.99 | 942706.00 | 132.6 | -3354.28 | 3.87 | 927564.90 | 16.7 |
| S=3/2 | -3353.92 | 21.76 | 939521.80 | 90.8 | -3354.27 | 13.31 | 927285.30 | 56.1 |
| S=5/2 | -3353.96 | 0.00 | 939957.00 | 0.0 | -3354.29 | 0.00 | 927055.80 | 0.0 |
| **9** | | | | | | | | |
| S=1/2 | -3247.95 | 42.86 | 715325.20 | 184.1 | -3248.23 | 17.34 | 705093.90 | 74.5 |
| **S=3/2** | -3248.00 | 10.88 | 711045.90 | 46.0 | -3248.25 | 4.41 | 703449.10 | 17.6 |
| S=5/2 | -3248.02 | 0.00 | 710565.30 | 0.0 | -3248.26 | 0.00 | 703363.10 | 0.0 |
| **10** | | | | | | | | |
| S=1/2 | -3589.64 | 23.00 | 1172701.20 | 98.3 | -3590.01 | 14.95 | 1159738.90 | 66.5 |
| S=3/2 | -3589.68 | 0.00 | 1171624.00 | 0.8 | -3590.02 | 12.35 | 1159111.10 | 55.2 |
| **S=5/2** | -3589.68 | 0.00 | 1170780.60 | 0.0 | -3590.04 | 0.00 | 1155647.10 | 0.0 |
| **13a** | | | | | | | | |
| **S=0** | -3389.09 | 37.89 | 979111.60 | 163.6 | -3389.48 | 2.70 | 970711.40 | 17.6 |
| S=2 | -3389.15 | 0.00 | 974206.60 | 0.0 | -3389.49 | 0.00 | 964562.20 | 0.0 |

**Table S3.** Relative energies (with respect to lowest *S* value) of spin states of iron complexes with Slater basis sets. The known ground-state multiplicity is highlighted in boldface. Energy and geometry calculated with the TZ2P basis set with the given functional at the OPBE/TZP geometry.

| Complex | E (au) | ΔE (kJ/mol) | E (au) | ΔE (kJ/mol) | Complex | E (au) | ΔE (kJ/mol) | E (au) | ΔE (kJ/mol) |
|---|---|---|---|---|---|---|---|---|---|
| | **OPBE** | | **M06-L** | | | **OPBE** | | **M06-L** | |
| **1** | | | | | **9** | | | | |
| **S=0** | -9.141843 | 0.00 | -9.63016 | 0.0 | S=1/2 | -8.512102 | 0.0 | -8.96228 | 0.0 |
| S=1 | -9.116660 | 15.80 | -9.60324 | 70.7 | **S=3/2** | -8.522772 | -28.0 | -8.98304 | -54.5 |
| S=2 | -9.092222 | 31.14 | -9.51406 | 304.8 | S=5/2 | -8.514030 | -5.1 | -8.99135 | -76.3 |
| **2** | | | | | **10** | | | | |
| **S=0** | -9.427992 | 0.00 | -9.91639 | 0.0 | S=1/2 | -13.172376 | 0.0 | -13.87953 | 0.0 |
| S=1 | -9.389178 | 24.36 | -9.88706 | 77.0 | S=3/2 | -13.169421 | 7.7 | -13.88140 | -4.9 |
| S=2 | -9.369650 | 36.61 | -9.89367 | 59.7 | **S=5/2** | -13.179933 | -19.8 | -13.89786 | -48.1 |
| **3** | | | | | **11** | | | | |
| **S=0** | -11.262982 | 0.00 | -11.89439 | 0.0 | S=0 | -13.223831 | 0.0 | -13.92308 | 0.0 |
| S=1 | -11.236432 | 16.66 | -11.86482 | 77.7 | **S=2** | -13.222437 | 3.6 | -13.93169 | -22.6 |
| S=2 | -11.227240 | 22.43 | -11.87529 | 50.2 | **12** | | | | |
| **4** | | | | | S = 0 | -17.289042 | 0.0 | -18.30955 | 0.0 |
| **S=0** | -9.409518 | 0.00 | -9.931298 | 0.0 | **S = 2** | -17.292500 | -9.1 | -18.28612 | 61.5 |
| S=1 | -9.385041 | 15.36 | -9.904149 | 71.3 | **13** | | | | |
| S=2 | -9.376769 | 20.55 | -9.905171 | 68.6 | S = 0 | -15.560082 | 0.0 | -16.46552 | 0.0 |
| **5** | | | | | **S = 2** | -15.563631 | -9.3 | -16.44109 | 64.1 |
| S=0 | -9.948673 | 0.00 | -10.49871 | 0.0 | **14** | | | | |
| S=1 | -9.937823 | 6.81 | -10.48295 | 41.4 | S = 0 | -18.989796 | 0.0 | -20.12500 | 0.0 |
| **S=2** | -9.932574 | 10.10 | -10.49696 | 4.6 | **S = 2** | -18.998276 | -22.3 | -20.10275 | 58.4 |
| **6** | | | | | **15** | | | | |
| S=0 | -9.550085 | 0.00 | -10.07311 | 0.0 | S=1/2 | -12.119127 | 0.0 | | |
| S=1 | -9.538047 | 7.55 | -10.05847 | 38.4 | **S=3/2** | -12.130049 | -28.7 | | |
| **S=2** | -9.526470 | 14.82 | -10.07349 | -1.0 | S=5/2 | -12.117507 | 4.3 | | |
| **7** | | | | | **16** | | | | |
| S=3/2 | -9.890682 | 0.00 | -10.38670 | 0.0 | S=1/2 | -31.418478 | 50.7 | | |
| **S=5/2** | -9.896617 | -3.72 | -10.40421 | -46.0 | **S=3/2** | -31.437796 | 0.0 | | |
| **8** | | | | | S=5/2 | -31.424471 | 35.0 | | |
| **S=1/2** | -12.557637 | 0.00 | -13.20257 | 0.0 | | | | | |
| S=3/2 | -12.533065 | 15.42 | -13.18618 | 43.0 | | | | | |
| S=5/2 | -12.538900 | 11.76 | -13.21094 | -22.0 | | | | | |

**Table S3 (continued).**

| Complex | E (au) | ΔE (kJ/mol) | E (au) | ΔE (kJ/mol) | E (au) | ΔE (kJ/mol) | E (au) | ΔE (kJ/mol) |
|---|---|---|---|---|---|---|---|---|
| | **PBE0** | | **B3LYP** | | **B3LYP*** | | **M06** | |
| **1** | | | | | | | | |
| **S=0** | -11.294259 | 0.0 | -10.407348 | 0.0 | -10.042735 | 0.0 | -10.994006 | 0.0 |
| S=1 | -11.286478 | 20.4 | -10.399378 | 20.9 | -10.029324 | 35.2 | -10.975367 | 49.0 |
| S=2 | -11.284088 | 26.7 | -10.394942 | 32.6 | -10.018053 | 64.8 | -10.989485 | 11.9 |
| **2** | | | | | | | | |
| **S=0** | -11.591679 | 0.0 | -10.702991 | 0.0 | -10.335796 | 0.0 | -11.283601 | 0.0 |
| S=1 | -11.572605 | 50.1 | -10.686144 | 44.2 | -10.313879 | 57.5 | -11.266374 | 45.2 |
| S=2 | -11.580073 | 30.5 | -10.691992 | 28.9 | -10.311348 | 64.2 | -11.285116 | -4.0 |
| **3** | | | | | | | | |
| **S=0** | -13.780024 | 0.0 | -12.739224 | 0.0 | -12.312605 | 0.0 | -13.424388 | 0.0 |
| S=1 | -13.766672 | 35.1 | -12.730093 | 24.0 | -12.298971 | 35.8 | -13.412412 | 31.4 |
| S=2 | -13.787239 | -19.0 | -12.749730 | -27.6 | -12.309395 | 8.4 | -13.432724 | -21.9 |
| **4** | | | | | | | | |
| **S=0** | -11.561884 | 0.0 | -10.668249 | 0.0 | -10.303246 | 0.0 | -11.253267 | 0.0 |
| S=1 | -11.550162 | 30.8 | -10.665225 | 7.9 | -10.293591 | 25.4 | -11.244980 | 21.8 |
| S=2 | -11.56506822 | -8.4 | -10.680498 | -32.2 | -10.301009 | 5.9 | -11.261672 | -22.0 |
| **5** | | | | | | | | |
| S=0 | -12.264363 | 0.0 | -11.325039 | 0.0 | -10.931831 | 0.0 | -11.943376 | 0.0 |
| S=1 | -12.259826 | 11.9 | -11.322833 | 5.8 | -10.925425 | 16.8 | -11.942474 | 2.4 |
| **S=2** | -12.282774 | -48.3 | -11.341835 | -44.1 | -10.936527 | -12.3 | -11.966406 | -60.5 |
| **6** | | | | | | | | |
| S=0 | -11.748367 | 0.0 | -10.852080 | 0.0 | -10.479052 | 0.0 | -11.436970 | 0.0 |
| S=1 | -11.743028 | 14.0 | -10.848517 | 9.4 | -10.471546 | 19.7 | -11.436028 | 2.5 |
| **S=2** | -11.766214 | -46.9 | -10.868114 | -42.1 | -10.483108 | -10.7 | -11.461045 | -63.2 |
| **7** | | | | | | | | |
| S=3/2 | -12.126014 | 0.0 | -11.228042 | 0.0 | -10.849060 | 0.0 | -11.802994 | 0.0 |
| **S=5/2** | -12.136552 | -27.7 | -11.232238 | -11.0 | -10.848441 | 1.6 | -11.824971 | -57.7 |
| **8** | | | | | | | | |
| **S=1/2** | -15.430622 | 0.0 | -14.290199 | 0.0 | -13.805237 | 0.0 | -15.038075 | 0.0 |
| S=3/2 | -15.420463 | 26.7 | -14.279606 | 27.8 | -13.790011 | 40.0 | -15.036331 | 4.6 |
| S=5/2 | -15.435351 | -12.4 | -14.291397 | -3.1 | -13.797251 | 21.0 | -15.064482 | -69.3 |

**Table S3 (continued).**

| Complex | E (au) | ΔE (kJ/mol) | E (au) | ΔE (kJ/mol) | E (au) | ΔE (kJ/mol) | E (au) | ΔE (kJ/mol) |
|---|---|---|---|---|---|---|---|---|
| | **PBE0** | | **B3LYP** | | **B3LYP*** | | **M06** | |
| **9** | | | | | | | | |
| S=1/2 | -10.464984 | 0.0 | -9.686188 | 0.0 | -9.355013 | 0.0 | -10.181326 | 0.0 |
| **S=3/2** | -10.492327 | -71.8 | -9.709268 | -60.6 | -9.373604 | -48.8 | -10.217907 | -96.1 |
| S=5/2 | -10.491507 | -69.6 | -9.706155 | -52.4 | -9.365710 | -28.1 | -10.227391 | -121.0 |
| **10** | | | | | | | | |
| S=1/2 | -16.114063 | 0.0 | -14.926728 | 0.0 | -14.432103 | 0.0 | -15.696870 | 0.0 |
| S=3/2 | -16.125015 | -28.7 | -14.933381 | -17.4 | -14.434671 | -6.7 | -15.712871 | -42.0 |
| **S=5/2** | -16.135694 | -56.8 | -14.942908 | -42.5 | -14.439924 | -20.5 | -15.734812 | -99.6 |
| **11a** | | | | | | | | |
| S=1/2 | | | | | -13.433343 | 0.0 | | |
| S=3/2 | | | | | -13.451015 | -46.4 | | |
| **S=5/2** | | | | | -13.435328 | -5.2 | | |
| **12** | | | | | | | | |
| **S=3/2** | | | | | -34.299285 | 0.0 | | |
| S=5/2 | | | | | -34.281389 | 47.0 | | |
| **13** | | | | | | | | |
| S=0 | -16.221224 | 0.0 | -15.037839 | 0.0 | -14.532262 | 0.0 | -15.804735 | 0.0 |
| **S=2** | -16.239244 | -47.3 | -15.052453 | -38.4 | -14.535142 | -7.6 | -15.833023 | -74.3 |
| **14** | | | | | | | | |
| S = 0 | -21.292287 | 0.0 | -19.789698 | 0.0 | -19.113057 | 0.0 | -20.826676 | 0.0 |
| **S = 2** | -21.296075 | -10.0 | -19.792556 | -7.5 | -19.103600 | 24.8 | -20.827731 | -2.8 |
| **15** | | | | | | | | |
| S = 0 | -19.123497 | 0.0 | -17.773708 | 0.0 | -17.172196 | 0.0 | -18.689077 | 0.0 |
| **S = 2** | -19.127893 | -11.5 | -17.777281 | -9.4 | -17.163201 | 23.6 | -18.690784 | -4.5 |
| **16** | | | | | | | | |
| S = 0 | -23.431967 | 0.0 | -21.775921 | 0.0 | -21.022975 | 0.0 | -22.935178 | 0.0 |
| **S = 2** | -23.439603 | -20.0 | -21.782764 | -17.9 | -21.017623 | 14.1 | -22.938922 | -9.8 |

**Table S4.** Calculated and experimental $^{1}$H shifts (ppm) for complexes **11-16**.[a]

| Complex | $\sigma_{orb}$ [b] | $A$ (MHz) | $\sigma_{FC}$ [c] | $\delta_{calc}$ [d] | $\delta_{exp}$ |
|---|---|---|---|---|---|
| [Fe$^{III}$(L)Cl] (**11**), $S$ = 1/2; $g_{iso}$ = 2.1336 | | | | | |
| H-1 | 29.70 | –0.004 | 0.09 | 1.29 | 2.00 |
| H-2 | 26.62 | 0.089 | –2.36 | 6.82 | 13.15 |
| H-3 | 27.75 | –0.662 | 17.51 | –14.18 | 55.82 |
| H-4 | 20.75 | 0.232 | –6.13 | 16.46 | –185.08 |
| H-5 | 26.36 | 3.501 | -92.65 | 97.37 | 74.97 |
| H-5’ | 26.34 | 2.611 | -69.11 | 73.84 | 58.42 |
| H-6 | 27.10 | -1.285 | 34.01 | -30.03 | 44.16 |
| H-6’ | 27.04 | -0.939 | 24.86 | -20.82 | 65.06 |
| [Fe$^{III}$(L)Cl] (**11**), $S$ = 3/2; $g_{iso}$ = 2.4307 | | | | | |
| H-1 | 29.77 | –0.001 | 0.14 | 1.16 | 2.00 |
| H-2 | 26.78 | 0.074 | –11.22 | 15.53 | 13.15 |
| H-3 | 28.31 | 0.431 | –64.91 | 67.68 | 55.82 |
| H-4 | 22.02 | –1.308 | 197.17 | –188.12 | –185.08 |
| H-5 | 27.38 | 0.409 | –61.69 | 65.38 | 74.97 |
| H-5’ | 27.63 | 0.334 | –50.30 | 54.00 | 58.42 |
| H-6 | 27.68 | 0.237 | –35.78 | 39.18 | 44.16 |
| H-6’ | 27.57 | 0.351 | –52.92 | 56.43 | 65.06 |
| [Fe$^{III}$(L)Cl] (**11**), $S$ = 5/2; $g_{iso}$ = 2.4489 | | | | | |
| H-1 | 29.83 | 0.001 | –0.27 | 1.53 | 2.00 |
| H-2 | 26.86 | 0.054 | –19.13 | 23.35 | 13.15 |
| H-3 | 28.58 | 0.023 | –8.17 | 10.68 | 55.82 |
| H-4 | 22.30 | 4.236 | –1501.03 | 1509.81 | –185.08 |
| H-5 | 27.16 | 0.938 | -332.43 | 336.34 | 58.42 |
| H-5’ | 27.14 | 1.169 | -414.33 | 418.27 | 44.16 |
| H-6 | 27.50 | 1.349 | -478.12 | 481.69 | 74.97 |
| H-6’ | 27.48 | 1.256 | -445.04 | 448.64 | 65.06 |

**Table S4 (continued).**

| Complex | $\sigma_{orb}$ [b] | *A* (MHz) | $\sigma_{FC}$ [c] | $\delta_{calc}$ [d] | $\delta_{exp}$ |
|---|---|---|---|---|---|
| $[Fe^{III}(oetpp)(HIm)]^+$ (**12**), $S = 3/2$; $g_{iso} = 2.6003$ | | | | | |
| $CH_3$ u [g] | 31.30 | –0.014 | 2.34 | –2.56 | –1 |
| $CH_3$ d [g] | 31.00 | –0.0002 | 0.03 | 0.05 | 1.7 |
| $CH_2$ (A) | 28.85 | 0.1781 | –30.65 | 32.88 | 24.3 |
| $CH_2$ (B) | 28.58 | 0.0773 | –13.30 | 15.80 | 18.3 |
| $CH_2$ (A) | 28.77 | 0.386 | –66.51 | 68.81 | 52.4 |
| $CH_2$ (B) | 28.25 | –0.018 | 3.09 | 0.26 | 9.4 |
| H-o1 | 22.38 | 0.0383 | –6.59 | 15.29 | 14.8 |
| H-o2 | 22.74 | 0.0377 | –6.48 | 14.83 | 11.8 |
| H-m1 | 22.97 | –0.027 | 4.65 | 3.47 | 6.9 |
| H-m2 | 23.05 | –0.023 | 3.09 | 4.12 | 7.4 |
| H-p | 22.95 | 0.026 | –4.40 | 12.53 | 9.8 |
| $L_1$ | 24.51 | 0.856 | –147.22 | 153.79 | 93.7 |
| $L_2$ | 27.34 | 0.206 | –35.48 | 39.22 | 42.9 |
| $L_3$ | 27.81 | 0.419 | –72.02 | 75.29 | n.a. |
| $L_4$ | 26.04 | 0.666 | –114.7 | 119.74 | 78.3 |
| $[Fe^{III}(oetpp)(HIm)]^+$ (**12**), S = 5/2; $g_{iso} = 2.4885$ | | | | | |
| $CH_3$ | 31.25 | 0.018 | –7.01 | 6.84 | –1 |
| $CH_3$ d | 31.04 | 0.025 | –9.45 | 9.49 | 1.7 |
| $CH_2$ (A) | 28.67 | 0.178 | –68.48 | 70.89 | 24.3 |
| $CH_2$ (B) | 28.68 | 0.179 | –68.77 | 71.18 | 18.3 |
| $CH_2$ (A) | 28.92 | 0.383 | –147.27 | 149.43 | 52.4 |
| $CH_2$ (B) | 28.32 | 0.129 | –49.48 | 52.24 | 9.4 |
| H-o1 | 22.36 | -0.026 | 10.05 | -1.33 | 14.8 |
| H-o2 | 22.66 | -0.027 | 10.50 | -2.09 | 11.8 |
| H-m1 | 22.97 | 0.018 | –6.93 | 15.03 | 6.9 |
| H-m2 | 23.05 | 0.022 | –8.60 | 16.63 | 7.4 |
| H-p | 22.95 | –0.019 | 7.44 | 0.69 | 9.8 |
| $L_1$ | 25.97 | 0.381 | –146.40 | 151.51 | 93.7 |
| $L_2$ | 27.42 | 0.118 | –45.19 | 48.84 | 42.9 |
| $L_3$ | 27.86 | 0.264 | –101.49 | 104.71 | n.a. |
| $L_4$ | 24.36 | 0.473 | –181.89 | 188.61 | 78.3 |

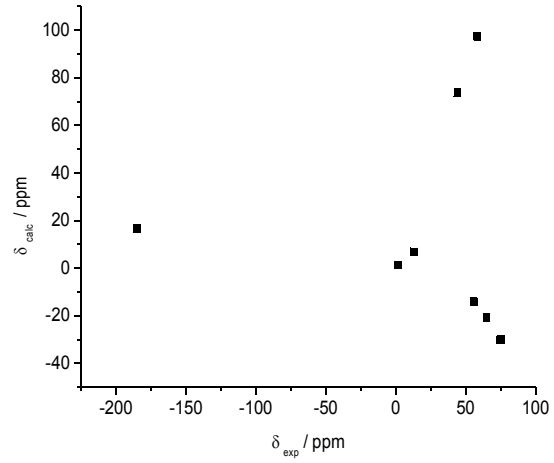
**Table S4 (continued).**

| Complex | $\sigma_{orb}$ [b] | *A* (MHz) | $\sigma_{FC}$ [c] | $\delta_{calc}$ [d] | $\delta_{exp}$ |
|---|---|---|---|---|---|
| *cis*-[$Fe^{II}$(phen)$_2$(NCS)$_2$] (**13a**), $S = 0$ [e] | | | | | |
| H-o | 21.83 | | | 10.31 | 9.18 |
| H-m | 23.47 | | | 8.67 | 7.58 |
| H-p | 22.84 | | | 9.29 | 8.20 |
| H-a | 23.02 | | | 9.11 | 8.22 |
| *cis*-[$Fe^{II}$(phen)$_2$(NCS)$_2$] (**13a**), $S = 1$; $g_{iso} = 2.3135$ | | | | | |
| H-o | 22.78 | 0.524 | -42.80 | 52.17 | 163.16 |
| H-m | 23.82 | 0.145 | -11.81 | 20.13 | 52.07 |
| H-p | 23.02 | 0.403 | -32.87 | 42.00 | 10.81 |
| H-a | 23.19 | 0.003 | -0.24 | 9.19 | 20.78 |
| *cis*-[$Fe^{II}$(phen)$_2$(NCS)$_2$] (**13a**), $S = 2$; $g_{iso} = 3.0033$ | | | | | |
| H-o | 19.15 | 0.698 | –221.91 | 234.90 | 163.16 |
| H-m | 23.33 | 0.266 | –84.58 | 93.39 | 52.07 |
| H-p | 22.91 | 0.196 | –62.22 | 71.44 | 10.81 |
| H-a | 23.33 | 0.091 | –28.84 | 37.65 | 20.78 |
| *trans*-[$Fe^{II}$(phen)$_2$(NCS)$_2$] (**13b**), $S = 0$ [e] | | | | | |
| H-o | 20.92 | | | 11.22 | 9.18 |
| H-m | 23.15 | | | 8.99 | 7.58 |
| H-p | 22.67 | | | 9.47 | 8.20 |
| H-a | 22.94 | | | 9.20 | 8.22 |
| *trans*-[$Fe^{II}$(phen)$_2$(NCS)$_2$] (**13b**), $S = 1$; $g_{iso} = 2.4410$ | | | | | |
| H-o | 22.84 | 0.928 | -79.96 | 89.26 | 163.16 |
| H-m | 23.67 | 0.140 | -12.06 | 20.53 | 52.07 |
| H-p | 22.94 | 0.382 | -32.92 | 42.12 | 10.81 |
| H-a | 23.19 | 0.028 | -2.43 | 11.38 | 20.78 |
| trans-[$Fe^{II}$(phen)$_2$(NCS)$_2$] (**13b**), $S = 2$; $g_{iso} = 3.0632$ | | | | | |
| H-o | 22.63 | 0.529 | –171.61 | 181.12 | 163.16 |
| H-m | 23.25 | 0.211 | –68.30 | 77.18 | 52.07 |
| H-p | 22.53 | 0.002 | –0.75 | 10.36 | 10.81 |
| H-a | 22.81 | 0.073 | –23.89 | 33.21 | 20.78 |

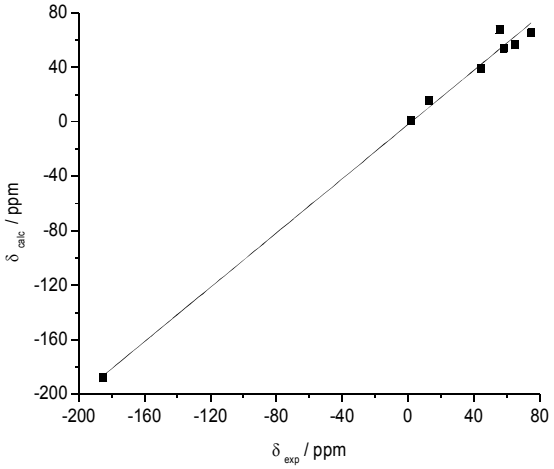
**Table S4 (continued).**

| Complex | $\sigma_{orb}$ [b] | $A$ (MHz) | $\sigma_{FC}$ [c] | $\delta_{calc}$ [d] | $\delta_{exp}$ |
|---|---|---|---|---|---|
| $[Fe^{II}(L1R_1R_2)(py)_2]$ $R_1$ = OEt, $R_2$ = $CH_3$ (**14**), $S$ = 0 [e] | | | | | |
| H-d | 29.80 | | | 1.28 | 1.3 |
| H-c | 26.94 | | | 4.13 | 4.2 |
| H-a | 28.37 | | | 2.71 | 2.5 |
| H-i | 20.98 | | | 10.10 | 8.3 |
| H-h | 23.16 | | | 7.91 | 7.2 |
| H-g | 23.99 | | | 7.08 | 7.3 |
| $[Fe^{II}(L1R_1R_2)(py)_2]$ $R_1$ = OEt, $R_2$ = $CH_3$ (**14**), $S$ = 2; $g_{iso}$ = 2.7145 | | | | | |
| H-d | 29.71 | 0.002 | –0.46 | 1.83 | 3.57 |
| H-c | 26.79 | 0.027 | –7.68 | 11.97 | 9.99 |
| H-a | 28.37 | –0.254 | 72.92 | –70.21 | –25.74 |
| H-i | 20.98 | 3.092 | –888.66 | 898.76 | 561 |
| H-h | 23.93 | 0.007 | –1.98 | 9.13 | 2.01 |
| H-g | 24.32 | 0.055 | –15.94 | 22.69 | 10.95 |
| $[Fe^{II}(L1R_1R_2)(py)_2]$ $R_1$, $R_2$ = $CH_3$ (**15**), $S$ = 0 [e] | | | | | |
| H-b | 28.82 | | | 2.26 | 2.5 |
| H-a | 28.44 | | | 2.64 | 2.5 |
| H-i | 20.97 | | | 10.11 | 8.3 |
| H-h | 23.17 | | | 7.91 | 7.1 |
| H-g | 23.99 | | | 7.08 | 7.2 |
| $[Fe^{II}(L1R_1R_2)(py)_2]$ $R_1$, $R_2$ = $CH_3$ (**15**), $S$ = 2; $g_{iso}$ = 2.7295 | | | | | |
| H-b | 28.71 | 0.022 | –6.51 | 8.89 | 13.36 |
| H-a | 28.43 | –0.257 | 74.39 | –71.74 | –23.94 |
| H-i | 21.06 | 3.095 | –894.59 | 904.60 | 551 |
| H-h | 23.97 | 0.006 | –1.77 | 8.88 | 1.52 |
| H-g | 24.32 | 0.055 | –15.89 | 22.65 | 10.46 |
| $[Fe^{II}(L1R_1R_2)(py)_2]$ $R_1$, $R_2$ = OEt (**16**), $S$ = 0 [f] | | | | | |
| H-d | 29.88 | | | 1.19 | 1.3 |
| H-c | 27.01 | | | 4.07 | 4.3 |
| H-f | 29.79 | | | 1.29 | 1.3 |
| H-e | 26.76 | | | 4.32 | 4.3 |
| H-i | 20.91 | | | 10.17 | 8.4 |
| H-h | 23.30 | | | 7.78 | 7.2 |
| H-g | 24.07 | | | 7.01 | 7.3 |
| $[Fe^{II}(L1R_1R_2)(py)_2]$ $R_1$, $R_2$ = OEt (**16**), $S$ = 2; $g_{iso}$ = 2.8265 | | | | | |
| H-d | 29.78 | 0.001 | –0.22 | 1.52 | 4.33 |
| H-c | 26.83 | 0.020 | –5.91 | 10.16 | 10.41 |
| H-f | 29.79 | 0.027 | –7.99 | 9.28 | 3.43 |
| H-e | 27.19 | –0.047 | 14.00 | –10.11 | –5.23 |
| H-i | 20.99 | 2.776 | –830.72 | 840.81 | 504 |
| H-h | 24.07 | –0.002 | 0.57 | 6.44 | –1.08 |
| H-g | 24.40 | 0.055 | –16.54 | 23.22 | 8.53 |

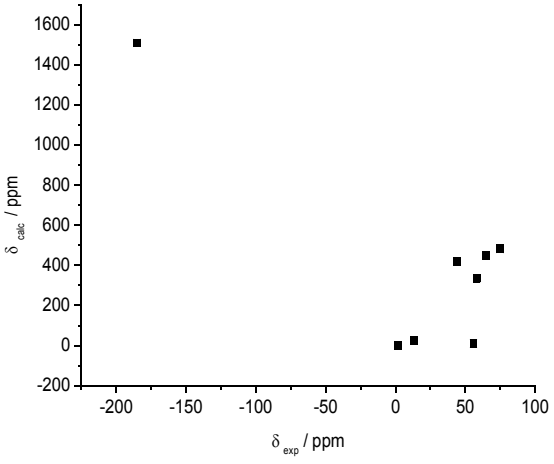
[a] B3LYP*/cc-pVTZ//B3LYP*/6-31G(d,p) calculations. See Scheme 1 for signal labelling. See text for the source of experimental shifts, except **11** (this work). [b] Orbital shielding. [c] Fermi contact shielding calculated from eq. (2) in the text. [d] $\sigma_{orb} + \sigma_{FC}$. [e] $-\sigma_{tot}$. [f] Experimental shifts are those of the diamagnetic free ligand.




(a)




(b)




(c)

**Figure S2a.** Experimental vs. calculated $^{1}$H chemical shifts for [Fe$^{III}$(L)Cl] (**11**), (a) $S$ = 1/2, (b) $S$ = 3/2 and (c) $S$ = 5/2. The fitting parameters of the correlation line $\delta_{calc} = a\delta_{exp} + b$ for (b) are: $a$ = 0.996, $b$ = –2.1 ppm ($r^2$ = 0.992).

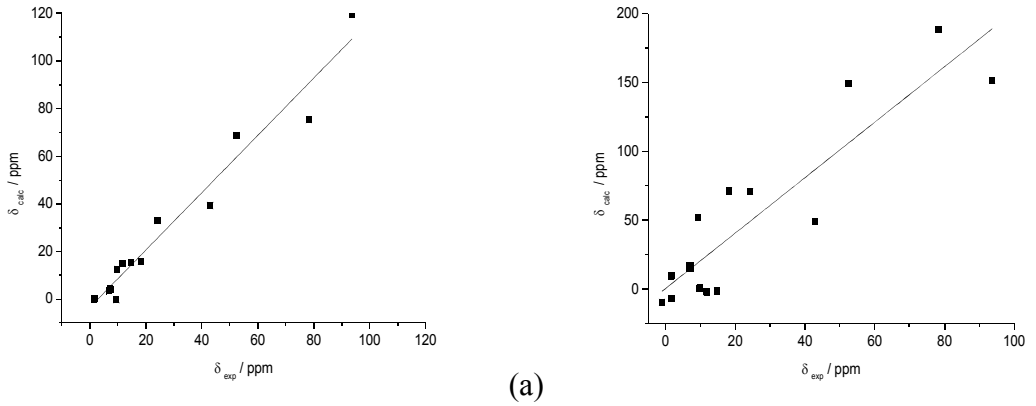



**Figure S2b.** Experimental and calculated $^1$H chemical shifts for $[Fe^{III}(oetpp)(HIm)]^+$ (**12**): (a) $S = 3/2$; (b) $S = 5/2$. The fitting parameters of the correlation lines $\delta_{calc} = a\delta_{exp} + b$ are (a) $a = 1.20$, $b = -3.6$ ppm ($r^2 = 0.968$); (b) $a = 2.02$, $b = 0.42$ ppm ($r^2 = 0.813$).

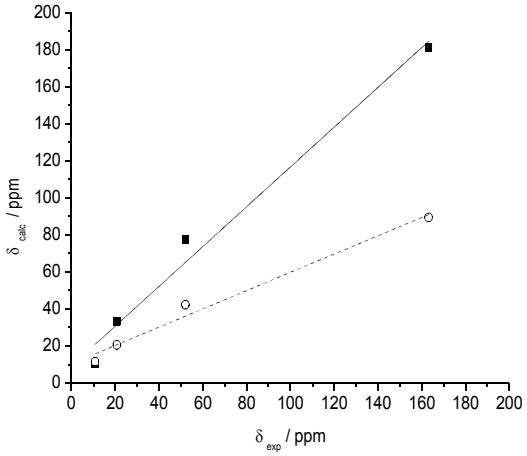
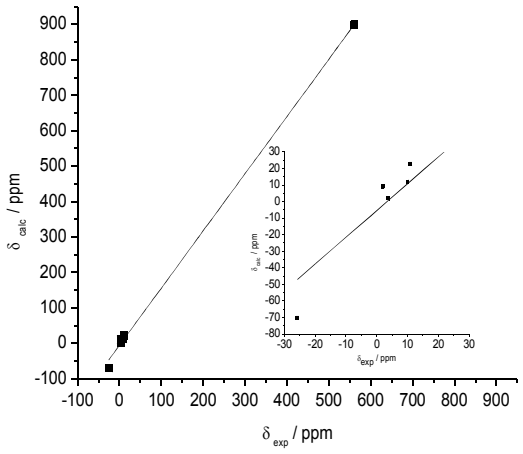
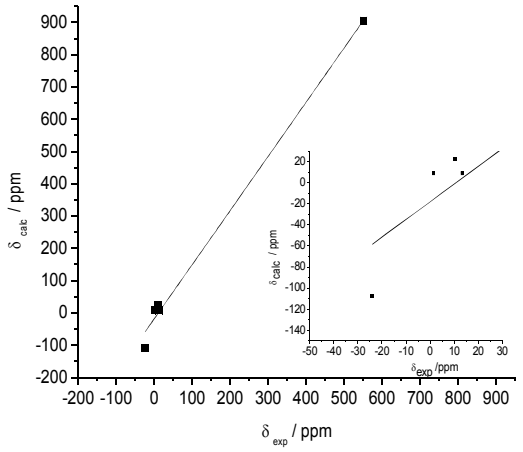


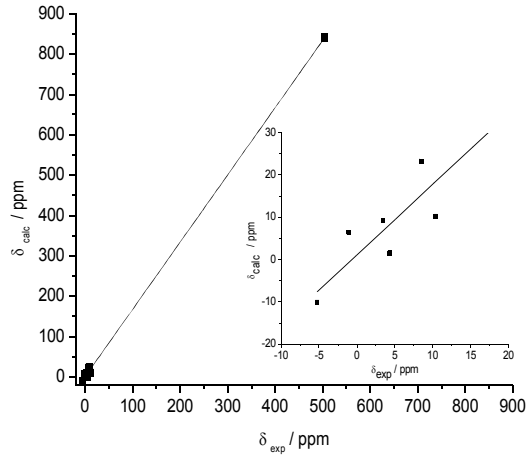

**Figure S2c.** Calculated $^1$H chemical shifts of *trans*-$[Fe^{II}(phen)_2(NCS)_2]$ (**13b**) with $S = 1$ (open circles) and $S = 2$ (filled squares), against experimental values. The fitting parameters of the correlation lines $\delta_{calc} = a\delta_{exp} + b$ are (a) $S = 1$: $a = 0.49$, $b = 10.37$ ppm ($r^2 = 0.976$); (b) $S = 2$: $a = 1.07$, $b = 9.16$ ppm ($r^2 = 0.977$).




(a)




(b)




(c)

**Figure S2d.** Experimental vs. calculated $^{1}$H chemical shifts of (a) **14**,(b) **15** and (c) **16** ($S = 2$). The inset shows an expansion of the most shielded region. The fitting parameters of the correlation lines $\delta_{calc} = a\delta_{exp} + b$ are: **14**, $a = 1.61$, $b = -5.4$ ppm ($r^2 = 0.999$). **15**, $a = 1.68$, $b = -18.1$ ppm ($r^2 = 0.993$); **16**, $a = 1.67$, $b = 1.1$ ppm ($r^2 = 0.999$).

**Estimating line widths from Solomon-Bloembergen equations**

The overall transverse relaxation rate $R_2 = 1/T_2$ can be expressed as the sum of a dipolar ($R_2^{dip}$) and a contact ($R_2^{FC}$) term, each term given by[1]

$$R_2^{dip} = \frac{1}{15}\left(\frac{\mu_0}{4\pi}\right)^2 \frac{\gamma_I^2 g_{iso}^2 \mu_B^2 S(S+1)}{r^6}\left[4\tau_c + J(\omega_I - \omega_S, \tau_c) + 3J(\omega_I, \tau_c) + 6J(\omega_I + \omega_S, \tau_c) + 6J(\omega_S, \tau_c)\right] \quad \text{(S1)}$$

$$R_2^{FC} = \frac{4\pi^2}{3} S(S+1) A^2 \left[\tau_e + J(\omega_S, \tau_e)\right] \quad \text{(S2)}$$

so that $R_2 = R_2^{dip} + R_2^{FC}$ and the line width $W = R_2/\pi$. $\omega_I$ and $\omega_S$ are the nuclear and electronic Larmor frequencies, respectively; $r$ is the distance between the observed nucleus and the paramagnetic centre (where it can be defined), and the other symbols have usual meanings. The spectral density function $J(\omega,\tau) = \frac{\tau}{1+\omega^2\tau^2}$ contains the correlation times for the dipolar and exchange interactions resulting from all the relevant dynamic processes:

$$\tau_c^{-1} = \tau_S^{-1} + \tau_r^{-1} + \tau_M^{-1}; \tau_e^{-1} = \tau_S^{-1} + \tau_M^{-1} \quad \text{(S3)}$$

$\tau_S$, $\tau_r$ and $\tau_M$ being the electronic relaxation, rotational and electron-spin exchange correlation times, respectively.[1] Since $\omega_S/\omega_I = -658$ for $^1$H. $J(\omega_I \pm \omega_S,\tau) \approx J(\omega_S,\tau)$. $\tau_r$ is given by

$$\tau_r = \frac{\eta V_m}{kT} \quad \text{(S4)}$$

where $\eta$ is the solvent viscosity and $V_m$ the molecular volume, which was calculated by DFT in this work.

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(1) I. Bertini, C. Luchinat, G. Parigi, *Solution NMR of Paramagnetic Molecules: Applications to Metallobiomolecules and Models.* Elsevier, Amsterdam, 2001.

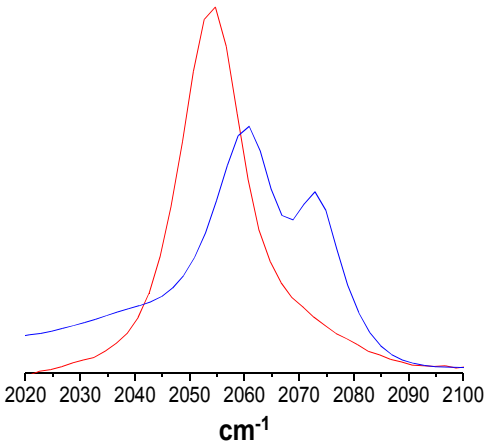


**Figure S3.** IR spectra of C≡N stretching band for complex **13**, in solid (blue) and in DMSO solution (red). A single band for $C\equiv N_{NCS}$ stretching indicates a *trans* configuration.

**Table S5.** Cartesian coordinates of the complexes investigated (B3LYP*/6-31G(d,p) geometries) and energies (B3LYP*/cc-pVTZ level, in au).

**$[Fe^{II}(N_HS_4)(NO)]^+$. S = 0. E =** -3648.26439139

| | | | |
|---|---|---|---|
| C | 4.280172 | 0.907311 | -1.467365 |
| C | 3.096368 | 0.454325 | -0.822416 |
| C | 3.197709 | -0.690533 | 0.013720 |
| C | 4.418664 | -1.390483 | 0.184134 |
| C | 5.578919 | -0.917467 | -0.456236 |
| C | 5.501500 | 0.233282 | -1.277276 |
| H | 4.229316 | 1.786559 | -2.126343 |
| H | 4.452818 | -2.296178 | 0.809039 |
| H | 6.532522 | -1.449252 | -0.328133 |
| H | 6.402986 | 0.600900 | -1.790116 |
| S | 1.720552 | -1.334765 | 0.853653 |
| S | 1.553973 | 1.333954 | -1.092626 |
| Fe | -0.030839 | -0.107594 | -0.103268 |
| S | -1.651614 | -1.400010 | 1.055349 |
| S | -1.760795 | 1.334068 | -0.754457 |
| C | -3.241145 | 0.303711 | -0.553329 |
| C | -4.434704 | 0.696986 | -1.206104 |
| C | -3.171489 | -0.866753 | 0.252621 |
| C | -5.601207 | -0.071121 | -1.025272 |
| H | -4.444153 | 1.591672 | -1.846778 |
| C | -4.360013 | -1.625830 | 0.418906 |
| C | -5.557195 | -1.224923 | -0.207775 |
| H | -6.534666 | 0.222763 | -1.526136 |
| H | -4.333089 | -2.539656 | 1.030397 |
| H | -6.464139 | -1.832583 | -0.069959 |
| C | 1.781771 | -0.471512 | 2.519531 |
| H | 2.811368 | -0.578790 | 2.910756 |
| H | 1.089816 | -1.045318 | 3.170072 |
| C | 1.363668 | 0.999358 | 2.371504 |
| H | 2.102604 | 1.561598 | 1.768613 |
| H | 1.275853 | 1.480500 | 3.370594 |
| N | 0.037911 | 1.058652 | 1.650876 |
| H | -0.668128 | 0.569950 | 2.241765 |
| C | -0.444785 | 2.471637 | 1.437405 |
| H | -0.497942 | 3.010538 | 2.408666 |
| H | 0.294556 | 2.982444 | 0.789577 |
| C | -1.835023 | 2.431062 | 0.785372 |
| H | -2.601071 | 1.986948 | 1.450503 |
| H | -2.165353 | 3.436623 | 0.462561 |
| O | -0.184361 | -1.709905 | -2.380806 |
| N | -0.124339 | -1.041546 | -1.433267 |

**$[Fe^{II}(N_HS_4)(NO)]^+$. S = 1. E** = -3648.25215160

| | | | |
|---|---|---|---|
| C | 4.210397 | -0.568328 | -1.746058 |
| C | 3.073486 | -0.384198 | -0.913889 |
| C | 3.236747 | -0.507942 | 0.492649 |
| C | 4.489698 | -0.832983 | 1.062227 |
| C | 5.610605 | -0.999087 | 0.222368 |
| C | 5.465418 | -0.863774 | -1.178110 |
| H | 4.099173 | -0.484062 | -2.837150 |
| H | 4.583644 | -0.959264 | 2.151560 |
| H | 6.589052 | -1.246484 | 0.658838 |
| H | 6.335960 | -1.004343 | -1.836004 |
| S | 1.792670 | -0.318339 | 1.583265 |
| S | 1.485281 | -0.014577 | -1.668500 |
| Fe | -0.014486 | -0.077733 | 0.042547 |
| S | -1.569214 | -0.457320 | 1.695181 |
| S | -1.748503 | 0.510806 | -1.421724 |
| C | -3.226984 | -0.107097 | -0.565934 |
| C | -4.446659 | -0.199684 | -1.274894 |
| C | -3.122154 | -0.512409 | 0.794672 |
| C | -5.597551 | -0.670036 | -0.609299 |
| H | -4.491411 | 0.090259 | -2.335397 |
| C | -4.291418 | -0.990797 | 1.444481 |
| C | -5.514603 | -1.058753 | 0.748243 |
| H | -6.551295 | -0.742867 | -1.151563 |
| H | -4.228574 | -1.318961 | 2.492428 |
| H | -6.409246 | -1.435642 | 1.265946 |
| C | 1.920366 | 1.483060 | 2.098227 |
| H | 2.967280 | 1.677887 | 2.398979 |
| H | 1.276518 | 1.570804 | 2.997884 |
| C | 1.465850 | 2.425258 | 0.967582 |
| H | 2.157009 | 2.360079 | 0.104239 |
| H | 1.454799 | 3.477633 | 1.329707 |
| N | 0.096644 | 2.012556 | 0.490267 |
| H | -0.560975 | 2.126434 | 1.285560 |
| C | -0.419758 | 2.858222 | -0.644167 |
| H | -0.473903 | 3.929273 | -0.347254 |
| H | 0.297355 | 2.767404 | -1.484269 |
| C | -1.820145 | 2.365236 | -1.052807 |
| H | -2.563956 | 2.503375 | -0.243236 |
| H | -2.178290 | 2.878480 | -1.965243 |
| O | -0.869788 | -2.622712 | -0.924320 |
| N | -0.212707 | -1.875923 | -0.293143 |

**$[Fe^{II}(N_HS_4)(NO)]^+$. S = 2. E** = -3648.23702407007

| | | | |
|---|---|---|---|
| C | 4.178978 | -0.756853 | -1.823725 |
| C | 3.115556 | -0.409580 | -0.941085 |
| C | 3.362924 | -0.476879 | 0.468666 |
| C | 4.622962 | -0.889926 | 0.963154 |
| C | 5.665612 | -1.210111 | 0.068268 |
| C | 5.437491 | -1.142073 | -1.324768 |
| H | 4.000247 | -0.719100 | -2.908027 |
| H | 4.782073 | -0.964928 | 2.049215 |
| H | 6.644706 | -1.522777 | 0.459247 |
| H | 6.241953 | -1.402145 | -2.028739 |
| S | 2.056871 | -0.150484 | 1.695237 |
| S | 1.569672 | 0.121366 | -1.673677 |
| Fe | 0.014262 | -0.016685 | 0.139699 |
| S | -1.743242 | -0.371767 | 1.755996 |
| S | -1.930424 | 0.883571 | -1.371857 |
| C | -3.324081 | 0.032276 | -0.577709 |
| C | -4.514137 | -0.124284 | -1.326494 |
| C | -3.210192 | -0.517869 | 0.742580 |
| C | -5.617157 | -0.803723 | -0.767231 |
| H | -4.572618 | 0.287351 | -2.344766 |
| C | -4.335992 | -1.203401 | 1.281743 |
| C | -5.523958 | -1.337700 | 0.537942 |
| H | -6.541833 | -0.917408 | -1.351467 |
| H | -4.260689 | -1.633997 | 2.290703 |
| H | -6.379024 | -1.872930 | 0.976997 |
| C | 2.203475 | 1.688220 | 2.049618 |
| H | 3.276617 | 1.919727 | 2.190556 |
| H | 1.695021 | 1.814561 | 3.028712 |
| C | 1.577978 | 2.588395 | 0.963639 |
| H | 2.142552 | 2.504040 | 0.014004 |
| H | 1.626708 | 3.648991 | 1.301874 |
| N | 0.157025 | 2.178755 | 0.687205 |
| H | -0.377164 | 2.253558 | 1.573342 |
| C | -0.518698 | 3.075700 | -0.313689 |
| H | -0.549471 | 4.127168 | 0.052562 |
| H | 0.088794 | 3.063036 | -1.242562 |
| C | -1.958784 | 2.598967 | -0.592748 |
| H | -2.560825 | 2.533105 | 0.334881 |
| H | -2.464102 | 3.282335 | -1.301709 |
| O | -1.029533 | -2.495342 | -0.929052 |
| N | -0.318610 | -1.785314 | -0.325229 |

**$[Fe^{II}(N_HS_4)(CO)]$. S = 0. E =** -3631.95200818

| | | | |
|---|---|---|---|
| C | 4.345622 | 1.199016 | -1.178809 |
| C | 3.128590 | 0.615621 | -0.712897 |
| C | 3.221215 | -0.669609 | -0.103226 |
| C | 4.453223 | -1.359003 | 0.011110 |
| C | 5.638499 | -0.758926 | -0.453022 |
| C | 5.572681 | 0.526390 | -1.046504 |
| H | 4.305323 | 2.190185 | -1.655236 |
| H | 4.472716 | -2.364247 | 0.461207 |
| H | 6.598981 | -1.288350 | -0.365975 |
| H | 6.492117 | 1.003603 | -1.421276 |
| S | 1.711778 | -1.479867 | 0.516862 |
| S | 1.582496 | 1.486371 | -0.906355 |
| Fe | -0.031696 | -0.103870 | -0.156973 |
| S | -1.677826 | -1.617929 | 0.694550 |
| S | -1.742798 | 1.411698 | -0.533926 |
| C | -3.262254 | 0.413048 | -0.465059 |
| C | -4.468189 | 0.961916 | -0.961881 |
| C | -3.203669 | -0.893469 | 0.103276 |
| C | -5.661658 | 0.219133 | -0.875849 |
| H | -4.462100 | 1.966025 | -1.413747 |
| C | -4.427031 | -1.620098 | 0.185222 |
| C | -5.630834 | -1.071444 | -0.293819 |
| H | -6.602689 | 0.636959 | -1.263593 |
| H | -4.411764 | -2.631776 | 0.617819 |
| H | -6.557522 | -1.663054 | -0.225390 |
| C | 1.764697 | -0.988061 | 2.329250 |
| H | 2.786728 | -1.175234 | 2.710741 |
| H | 1.058914 | -1.677014 | 2.838726 |
| C | 1.343938 | 0.483842 | 2.495218 |
| H | 2.087340 | 1.152652 | 2.017900 |
| H | 1.273363 | 0.745112 | 3.576777 |
| N | 0.028447 | 0.697312 | 1.804866 |
| H | -0.686527 | 0.111248 | 2.283552 |
| C | -0.431015 | 2.124745 | 1.848481 |
| H | -0.501500 | 2.491810 | 2.898790 |
| H | 0.328561 | 2.729247 | 1.312947 |
| C | -1.807533 | 2.241315 | 1.167949 |
| H | -2.598203 | 1.718925 | 1.741858 |
| H | -2.099887 | 3.300002 | 1.029619 |
| C | -0.130387 | -0.809231 | -1.764583 |
| O | -0.192876 | -1.282398 | -2.832464 |

**[$Fe^{II}(N_HS_4)(CO)$]. S = 1. E =** -3631.93185741

| | | | |
|---|---|---|---|
| C | 4.356841 | 1.082348 | -1.416674 |
| C | 3.195383 | 0.479392 | -0.842167 |
| C | 3.397678 | -0.697344 | -0.052486 |
| C | 4.695315 | -1.245242 | 0.125547 |
| C | 5.821070 | -0.627490 | -0.448216 |
| C | 5.639889 | 0.545198 | -1.220236 |
| H | 4.225226 | 1.985040 | -2.031794 |
| H | 4.807641 | -2.166704 | 0.717584 |
| H | 6.823190 | -1.058983 | -0.306081 |
| H | 6.508534 | 1.039968 | -1.682955 |
| S | 2.009630 | -1.572951 | 0.731831 |
| S | 1.609659 | 1.234541 | -1.189112 |
| Fe | -0.061922 | -0.156478 | -0.168348 |
| S | -1.744184 | -1.475276 | 0.898250 |
| S | -2.021832 | 1.605386 | -0.499129 |
| C | -3.423192 | 0.454715 | -0.373901 |
| C | -4.668852 | 0.882553 | -0.899460 |
| C | -3.269177 | -0.859208 | 0.178349 |
| C | -5.788900 | 0.030074 | -0.869230 |
| H | -4.747645 | 1.891390 | -1.331777 |
| C | -4.420470 | -1.698066 | 0.199984 |
| C | -5.656737 | -1.262336 | -0.311517 |
| H | -6.752353 | 0.369762 | -1.278063 |
| H | -4.320785 | -2.710913 | 0.617470 |
| H | -6.522676 | -1.942447 | -0.284556 |
| C | 1.984391 | -0.753523 | 2.413320 |
| H | 3.000678 | -0.816298 | 2.848967 |
| H | 1.302960 | -1.377779 | 3.029944 |
| C | 1.506303 | 0.711168 | 2.354131 |
| H | 2.202381 | 1.323098 | 1.746742 |
| H | 1.482191 | 1.132982 | 3.386616 |
| N | 0.146146 | 0.799575 | 1.725101 |
| H | -0.512101 | 0.237667 | 2.301942 |
| C | -0.376951 | 2.210350 | 1.715879 |
| H | -0.303353 | 2.656141 | 2.735633 |
| H | 0.271763 | 2.799711 | 1.034985 |
| C | -1.849977 | 2.258523 | 1.257166 |
| H | -2.499848 | 1.639071 | 1.906557 |
| H | -2.219350 | 3.302556 | 1.277461 |
| C | -0.359908 | -0.900597 | -1.753875 |
| O | -0.572822 | -1.377628 | -2.798073 |

**[$Fe^{II}(N_HS_4)(CO)$]. S = 2. E** = -3631.92414691

| | | | |
|---|---|---|---|
| C | 4.388346 | -0.256902 | -1.780493 |
| C | 3.214725 | -0.211746 | -0.967786 |
| C | 3.389491 | -0.449100 | 0.434309 |
| C | 4.670247 | -0.732711 | 0.976488 |
| C | 5.809005 | -0.760379 | 0.150796 |
| C | 5.656780 | -0.519870 | -1.235120 |
| H | 4.277417 | -0.081196 | -2.860659 |
| H | 4.760502 | -0.936326 | 2.054680 |
| H | 6.798685 | -0.978105 | 0.579536 |
| H | 6.535449 | -0.546067 | -1.898941 |
| S | 1.982966 | -0.468199 | 1.590905 |
| S | 1.650828 | 0.143765 | -1.769783 |
| Fe | -0.064555 | -0.242811 | -0.055495 |
| S | -1.822502 | -0.587639 | 1.676967 |
| S | -1.998152 | 0.777715 | -1.442798 |
| C | -3.433865 | 0.033743 | -0.611034 |
| C | -4.662030 | 0.023723 | -1.320473 |
| C | -3.325133 | -0.546383 | 0.696246 |
| C | -5.814635 | -0.544902 | -0.746846 |
| H | -4.700312 | 0.465252 | -2.327848 |
| C | -4.512079 | -1.112742 | 1.249848 |
| C | -5.730148 | -1.110679 | 0.546634 |
| H | -6.764351 | -0.549607 | -1.302530 |
| H | -4.452094 | -1.566613 | 2.250012 |
| H | -6.621559 | -1.563038 | 1.009459 |
| C | 1.961673 | 1.299353 | 2.210315 |
| H | 2.988118 | 1.557865 | 2.535899 |
| H | 1.310181 | 1.271574 | 3.109693 |
| C | 1.440445 | 2.318930 | 1.174279 |
| H | 2.105041 | 2.336616 | 0.286680 |
| H | 1.453055 | 3.337527 | 1.634329 |
| N | 0.074183 | 1.944793 | 0.708864 |
| H | -0.566873 | 1.909214 | 1.523036 |
| C | -0.490390 | 2.892321 | -0.295606 |
| H | -0.498655 | 3.941637 | 0.088335 |
| H | 0.171912 | 2.864666 | -1.186661 |
| C | -1.935199 | 2.498534 | -0.675366 |
| H | -2.596065 | 2.479542 | 0.213986 |
| H | -2.348127 | 3.216957 | -1.409981 |
| C | -0.161441 | -2.202132 | -0.557452 |
| O | 0.020534 | -3.343056 | -0.642712 |

**[$Fe^{II}(N_HS_4)$ $P(CH_3)_3$]. S = 0. E** = -3978.36083775

| | | | |
|---|---|---|---|
| C | 4.422627 | 0.133336 | 1.534935 |
| C | 3.186051 | 0.017916 | 0.827440 |
| C | 3.260174 | -0.018553 | -0.597204 |
| C | 4.493942 | 0.083700 | -1.285063 |
| C | 5.697362 | 0.192918 | -0.562857 |
| C | 5.649357 | 0.215016 | 0.853781 |
| H | 4.396423 | 0.162435 | 2.634966 |
| H | 4.500663 | 0.078387 | -2.386919 |
| H | 6.658342 | 0.269772 | -1.093418 |
| H | 6.582875 | 0.306485 | 1.431894 |
| S | 1.721646 | -0.167006 | -1.569951 |
| S | 1.638936 | -0.056622 | 1.715863 |
| Fe | -0.019995 | -0.107162 | -0.037783 |
| S | -1.677172 | -0.339906 | -1.774852 |
| S | -1.726574 | -0.421941 | 1.488128 |
| C | -3.270895 | -0.179775 | 0.549885 |
| C | -4.488731 | -0.015792 | 1.252094 |
| C | -3.215916 | -0.183356 | -0.876952 |
| C | -5.696307 | 0.128506 | 0.541015 |
| H | -4.480794 | -0.004619 | 2.353478 |
| C | -4.454887 | -0.043160 | -1.571457 |
| C | -5.668087 | 0.107932 | -0.875170 |
| H | -6.645945 | 0.259379 | 1.081365 |
| H | -4.443266 | -0.043923 | -2.672056 |
| H | -6.605446 | 0.221703 | -1.443003 |
| C | 1.734457 | -2.014713 | -1.906072 |
| H | 2.737323 | -2.298900 | -2.278449 |
| H | 0.989472 | -2.172505 | -2.713583 |
| C | 1.353104 | -2.773626 | -0.623020 |
| H | 2.129080 | -2.625027 | 0.154155 |
| H | 1.265535 | -3.866003 | -0.830557 |
| N | 0.062354 | -2.221582 | -0.095659 |
| H | -0.678474 | -2.420902 | -0.799589 |
| C | -0.344515 | -2.850138 | 1.203615 |
| H | -0.387229 | -3.961530 | 1.116993 |
| H | 0.428171 | -2.582189 | 1.951814 |
| C | -1.724623 | -2.314371 | 1.622224 |
| H | -2.529499 | -2.678335 | 0.953488 |
| H | -1.967868 | -2.592911 | 2.665600 |
| P | -0.147343 | 2.162311 | 0.036829 |
| C | 1.430465 | 3.093879 | -0.347581 |
| H | 2.228313 | 2.764412 | 0.345373 |
| H | 1.753325 | 2.884641 | -1.387250 |
| H | 1.279432 | 4.187976 | -0.236161 |
| C | -0.564741 | 2.889395 | 1.709198 |
| H | -0.526975 | 3.998431 | 1.681305 |
| H | -1.581423 | 2.574843 | 2.019096 |
| H | 0.164867 | 2.512336 | 2.452013 |
| C | -1.368731 | 3.041875 | -1.072315 |
| H | -1.145372 | 2.805310 | -2.130441 |
| H | -2.394716 | 2.684557 | -0.858469 |
| H | -1.323302 | 4.140471 | -0.918116 |

**[$Fe^{II}(N_HS_4)$ $P(CH_3)_3$]. S = 1. E = -3978.34693766**

| | | | |
|---|---|---|---|
| C | 4.366740 | 0.254503 | 1.559034 |
| C | 3.160651 | 0.067642 | 0.821509 |
| C | 3.273621 | -0.072795 | -0.590587 |
| C | 4.526373 | -0.009400 | -1.246089 |
| C | 5.705491 | 0.170331 | -0.496183 |
| C | 5.614851 | 0.301696 | 0.910817 |
| H | 4.305099 | 0.363888 | 2.652571 |
| H | 4.568474 | -0.097320 | -2.343467 |
| H | 6.681670 | 0.218423 | -1.001941 |
| H | 6.528945 | 0.449956 | 1.507644 |
| S | 1.756706 | -0.296961 | -1.581534 |
| S | 1.579134 | 0.022129 | 1.669942 |
| Fe | -0.011049 | -0.099451 | -0.062168 |
| S | -1.632028 | -0.293426 | -1.770071 |
| S | -1.705657 | -0.547823 | 1.490933 |
| C | -3.248514 | -0.300136 | 0.552709 |
| C | -4.477566 | -0.204894 | 1.247790 |
| C | -3.181938 | -0.210333 | -0.868947 |
| C | -5.679981 | -0.035838 | 0.532526 |
| H | -4.482350 | -0.262564 | 2.347614 |
| C | -4.412343 | -0.044250 | -1.569574 |
| C | -5.636888 | 0.040031 | -0.880478 |
| H | -6.637638 | 0.041280 | 1.069196 |
| H | -4.387603 | 0.027195 | -2.667699 |
| H | -6.569665 | 0.175719 | -1.450900 |
| C | 1.819147 | -2.159848 | -1.852861 |
| H | 2.834760 | -2.414760 | -2.213303 |
| H | 1.095174 | -2.348037 | -2.672973 |
| C | 1.446115 | -2.959788 | -0.582439 |
| H | 2.183100 | -2.755583 | 0.221744 |
| H | 1.493920 | -4.053014 | -0.816146 |
| N | 0.112504 | -2.528943 | -0.103718 |
| H | -0.605807 | -2.747265 | -0.815234 |
| C | -0.312717 | -3.038793 | 1.220204 |
| H | -0.392028 | -4.154547 | 1.255118 |
| H | 0.454694 | -2.729748 | 1.960166 |
| C | -1.692367 | -2.447997 | 1.593188 |
| H | -2.481190 | -2.796403 | 0.896867 |
| H | -1.976348 | -2.732634 | 2.624918 |
| P | -0.226376 | 2.471825 | 0.028373 |
| C | 1.332093 | 3.421785 | -0.408772 |
| H | 2.162287 | 3.079266 | 0.240410 |
| H | 1.604185 | 3.208380 | -1.462628 |
| H | 1.201853 | 4.517999 | -0.285218 |
| C | -0.639480 | 3.184607 | 1.712922 |
| H | -0.626260 | 4.295248 | 1.711945 |
| H | -1.647571 | 2.835981 | 2.016644 |
| H | 0.094642 | 2.807364 | 2.452088 |
| C | -1.495818 | 3.325433 | -1.056169 |
| H | -1.291600 | 3.084784 | -2.117987 |
| H | -2.504630 | 2.935468 | -0.814851 |
| H | -1.486035 | 4.427482 | -0.917683 |

**$[Fe^{II}(N_HS_4)\ P(CH_3)_3]$. S = 2. E** = -3978.35392968

| | | | |
|---|---|---|---|
| C | 4.456932 | 0.404627 | 1.602832 |
| C | 3.279928 | 0.124182 | 0.839950 |
| C | 3.456590 | -0.040884 | -0.574331 |
| C | 4.737211 | 0.086106 | -1.174419 |
| C | 5.875953 | 0.355742 | -0.393674 |
| C | 5.723587 | 0.514294 | 1.004377 |
| H | 4.347881 | 0.535158 | 2.689814 |
| H | 4.826721 | -0.026222 | -2.266123 |
| H | 6.864495 | 0.449837 | -0.867908 |
| H | 6.601484 | 0.731789 | 1.633652 |
| S | 2.054481 | -0.362124 | -1.691563 |
| S | 1.719136 | 0.001702 | 1.710497 |
| Fe | -0.024680 | -0.097488 | -0.080867 |
| S | -1.822596 | -0.454660 | -1.803558 |
| S | -1.958060 | -0.733589 | 1.596067 |
| C | -3.409399 | -0.322367 | 0.580572 |
| C | -4.635186 | -0.099204 | 1.259753 |
| C | -3.317822 | -0.202063 | -0.848037 |
| C | -5.801686 | 0.236265 | 0.546933 |
| H | -4.660528 | -0.194206 | 2.356083 |
| C | -4.522144 | 0.132569 | -1.540498 |
| C | -5.735272 | 0.346253 | -0.861886 |
| H | -6.748135 | 0.407120 | 1.081809 |
| H | -4.478126 | 0.227752 | -2.635677 |
| H | -6.637879 | 0.606174 | -1.437845 |
| C | 2.036029 | -2.234170 | -1.768924 |
| H | 3.060486 | -2.574801 | -2.016655 |
| H | 1.375855 | -2.469131 | -2.631143 |
| C | 1.529451 | -2.913353 | -0.477320 |
| H | 2.202400 | -2.670879 | 0.370376 |
| H | 1.551701 | -4.021880 | -0.625813 |
| N | 0.164949 | -2.432981 | -0.126290 |
| H | -0.485907 | -2.639647 | -0.906585 |
| C | -0.376573 | -3.048262 | 1.117253 |
| H | -0.357338 | -4.165401 | 1.068428 |
| H | 0.280552 | -2.736550 | 1.956857 |
| C | -1.833670 | -2.600666 | 1.366751 |
| H | -2.486727 | -2.860802 | 0.510043 |
| H | -2.233126 | -3.084454 | 2.279468 |
| P | -0.249554 | 2.403271 | 0.000159 |
| C | 1.298150 | 3.366582 | -0.428847 |
| H | 2.129752 | 3.036506 | 0.224284 |
| H | 1.575633 | 3.156562 | -1.481748 |
| H | 1.148415 | 4.460110 | -0.306648 |
| C | -0.701336 | 3.088814 | 1.681743 |
| H | -0.709863 | 4.199128 | 1.688368 |
| H | -1.707924 | 2.718287 | 1.963130 |
| H | 0.029246 | 2.719133 | 2.427984 |
| C | -1.524222 | 3.209097 | -1.107897 |
| H | -1.304661 | 2.953136 | -2.162995 |
| H | -2.527129 | 2.804181 | -0.868566 |
| H | -1.533428 | 4.312954 | -0.987470 |

**[$Fe^{II}(N_HS_4)(PH_3)$]. S = 0. E =** -3861.23347661

| | | | |
|---|---|---|---|
| C | 4.34623600 | 0.97637100 | -1.29285700 |
| C | 3.12989100 | 0.49283200 | -0.76135700 |
| C | 3.20911600 | -0.63791800 | 0.07870300 |
| C | 4.42762700 | -1.27246500 | 0.35258500 |
| C | 5.60993000 | -0.77308600 | -0.18258700 |
| C | 5.55666600 | 0.35899800 | -1.00685600 |
| H | 4.31670800 | 1.84630500 | -1.94149900 |
| H | 4.43822500 | -2.15697300 | 0.98388100 |
| H | 6.55623000 | -1.26066200 | 0.02871000 |
| H | 6.47182300 | 0.75687600 | -1.43716000 |
| S | 1.70418000 | -1.31371600 | 0.79300900 |
| S | 1.60758900 | 1.28456100 | -1.15980100 |
| Fe | -0.02219600 | -0.08923800 | -0.12061600 |
| S | -1.65974700 | -1.46393600 | 0.94597000 |
| S | -1.73709300 | 1.30376000 | -0.75615000 |
| C | -3.23992200 | 0.36094800 | -0.46559300 |
| C | -4.44619900 | 0.82917600 | -0.99966800 |
| C | -3.17604100 | -0.82208200 | 0.30146900 |
| C | -5.63003200 | 0.13885700 | -0.75834200 |
| H | -4.44652700 | 1.73332900 | -1.60169500 |
| C | -4.39137600 | -1.49784200 | 0.53571600 |
| C | -5.59186000 | -1.02545500 | 0.01806500 |
| H | -6.56695400 | 0.49792500 | -1.17205100 |
| H | -4.37174000 | -2.40938600 | 1.12481700 |
| H | -6.50855200 | -1.57558600 | 0.21254600 |
| C | 1.71857900 | -0.52278800 | 2.46241200 |
| H | 2.71365900 | -0.63788800 | 2.89655900 |
| H | 1.00362900 | -1.09505100 | 3.06241900 |
| C | 1.31054800 | 0.93855600 | 2.34976100 |
| H | 2.05976500 | 1.49489100 | 1.78040900 |
| H | 1.22856300 | 1.38938100 | 3.34948400 |
| N | 0.02530300 | 1.03274900 | 1.61935000 |
| H | -0.69240200 | 0.56331700 | 2.18138600 |
| C | -0.40147500 | 2.43132500 | 1.39457300 |
| H | -0.45656100 | 2.98617000 | 2.34240600 |
| H | 0.35478900 | 2.90271800 | 0.76090400 |
| C | -1.76329900 | 2.44905400 | 0.71714500 |
| H | -2.55110600 | 2.08586700 | 1.38196300 |
| H | -2.02316400 | 3.45323800 | 0.37640300 |
| P | -0.19935000 | -1.42665400 | -1.91845300 |
| H | -1.48033700 | -1.71195500 | -2.44939500 |
| H | 0.45877500 | -1.12128400 | -3.13438300 |
| H | 0.23621500 | -2.76807800 | -1.79577600 |

**[$Fe^{II}(N_HS_4)(PH_3)$]. S = 1. E** = -3861.21871165

| | | | |
|---|---|---|---|
| C | -4.381912 | -1.059400 | 1.400055 |
| C | -3.225210 | -0.679113 | 0.653049 |
| C | -3.440875 | -0.154376 | -0.660572 |
| C | -4.750829 | -0.042632 | -1.195567 |
| C | -5.873118 | -0.423362 | -0.436845 |
| C | -5.677003 | -0.931296 | 0.869637 |
| H | -4.237434 | -1.465495 | 2.412553 |
| H | -4.876447 | 0.344221 | -2.218848 |
| H | -6.884980 | -0.334649 | -0.860298 |
| H | -6.543050 | -1.237715 | 1.477776 |
| S | -2.052432 | 0.374110 | -1.714121 |
| S | -1.617090 | -0.935900 | 1.407197 |
| Fe | 0.055927 | -0.190148 | -0.110390 |
| S | 1.731914 | 0.526618 | -1.617433 |
| S | 2.064822 | -0.093146 | 1.704626 |
| C | 3.460685 | -0.189318 | 0.542430 |
| C | 4.727354 | -0.552435 | 1.068531 |
| C | 3.284976 | 0.033390 | -0.863501 |
| C | 5.844468 | -0.687369 | 0.222364 |
| H | 4.824297 | -0.725619 | 2.151045 |
| C | 4.435755 | -0.102202 | -1.694217 |
| C | 5.690233 | -0.454874 | -1.164175 |
| H | 6.823243 | -0.968571 | 0.639609 |
| H | 4.319821 | 0.066685 | -2.775294 |
| H | 6.554115 | -0.557247 | -1.840071 |
| C | -1.997284 | 2.195111 | -1.296436 |
| H | -3.004155 | 2.626155 | -1.461871 |
| H | -1.301163 | 2.643882 | -2.036754 |
| C | -1.526553 | 2.457111 | 0.147689 |
| H | -2.232130 | 2.005046 | 0.873200 |
| H | -1.498849 | 3.557614 | 0.331023 |
| N | -0.172532 | 1.855637 | 0.382347 |
| H | 0.490710 | 2.288372 | -0.291038 |
| C | 0.343744 | 2.164511 | 1.761342 |
| H | 0.252248 | 3.256948 | 1.969663 |
| H | -0.297432 | 1.622634 | 2.487197 |
| C | 1.825566 | 1.760212 | 1.922385 |
| H | 2.467445 | 2.265245 | 1.173410 |
| H | 2.180085 | 2.038574 | 2.934422 |
| P | 0.428770 | -2.400890 | -0.500800 |
| H | 1.489725 | -2.827987 | -1.370068 |
| H | 0.719878 | -3.266694 | 0.605397 |
| H | -0.619009 | -3.193191 | -1.076541 |

**[$Fe^{II}(N_HS_4)(PH_3)$]. S = 2. E =** -3861.22678066

| | | | |
|---|---|---|---|
| C | 0.006411 | 0.067546 | 0.002155 |
| C | 0.014395 | 0.086931 | 1.416753 |
| C | 1.248542 | 0.057971 | 2.092298 |
| C | 2.468105 | 0.010592 | 1.368536 |
| C | 2.481144 | 0.008729 | -0.066597 |
| C | 1.212106 | 0.030675 | -0.720512 |
| S | 4.008924 | -0.013378 | 2.333150 |
| C | 4.466740 | -1.843117 | 2.299377 |
| C | 5.971945 | -2.048821 | 2.019570 |
| N | 6.350142 | -1.483082 | 0.692504 |
| C | 7.776561 | -1.718164 | 0.329470 |
| C | 8.098389 | -1.106912 | -1.052020 |
| S | 7.745902 | 0.730086 | -1.173792 |
| Fe | 5.736220 | 0.732217 | 0.529084 |
| P | 4.846881 | 3.142771 | 0.039727 |
| S | 3.963313 | -0.028289 | -1.077251 |
| C | 9.086748 | 1.435042 | -0.163640 |
| C | 8.904193 | 1.750029 | 1.222956 |
| C | 10.023638 | 2.322864 | 1.902710 |
| C | 11.243180 | 2.568673 | 1.249648 |
| C | 11.400660 | 2.260575 | -0.122758 |
| C | 10.315694 | 1.702454 | -0.822590 |
| S | 7.398883 | 1.484631 | 2.159632 |
| H | 9.907853 | 2.573770 | 2.967561 |
| H | 10.407508 | 1.468863 | -1.894719 |
| H | 12.350585 | 2.461258 | -0.640705 |
| H | 12.078721 | 3.012249 | 1.814359 |
| H | 1.282582 | 0.071675 | 3.192296 |
| H | -0.927309 | 0.123594 | 1.984902 |
| H | 1.196947 | 0.027897 | -1.820544 |
| H | -0.950227 | 0.088596 | -0.543913 |
| H | 9.162598 | -1.267432 | -1.313534 |
| H | 7.474147 | -1.563705 | -1.849438 |
| H | 8.406769 | -1.245372 | 1.110310 |
| H | 8.020572 | -2.808927 | 0.304438 |
| H | 5.738690 | -1.904240 | -0.031486 |
| H | 6.194596 | -3.142504 | 2.079716 |
| H | 6.588169 | -1.536587 | 2.788760 |
| H | 3.843061 | -2.315828 | 1.514500 |
| H | 4.196160 | -2.280890 | 3.279990 |
| H | 3.435171 | 3.397318 | 0.080438 |
| H | 5.255024 | 4.314009 | 0.765899 |
| H | 5.069561 | 3.675323 | -1.274153 |

**$[Fe^{II}(N_{II}S_4)(N_2H_4)]$. S = 0. E =** -3630.43178181

| | | | |
|---|---|---|---|
| C | -4.38657400 | -1.57204300 | 0.46326500 |
| C | -3.16641700 | -0.88527700 | 0.18160700 |
| C | -3.25916600 | 0.29838900 | -0.60893800 |
| C | -4.49647200 | 0.76361200 | -1.11713100 |
| C | -5.68513400 | 0.06962500 | -0.82083900 |
| C | -5.61798200 | -1.10206200 | -0.02594300 |
| H | -4.34546000 | -2.48537600 | 1.07617600 |
| H | -4.51826600 | 1.66890500 | -1.74501500 |
| H | -6.64958700 | 0.42893700 | -1.21033200 |
| H | -6.53994700 | -1.65820200 | 0.20806600 |
| S | -1.73066600 | 1.20995300 | -1.02404300 |
| S | -1.60818700 | -1.50066600 | 0.80762200 |
| Fe | 0.02064200 | -0.00251000 | -0.13053900 |
| S | 1.68453600 | 1.42883600 | -1.10996700 |
| S | 1.74143000 | -1.07624300 | 0.98561100 |
| C | 3.27770500 | -0.52607900 | 0.17207100 |
| C | 4.48762700 | -1.20693000 | 0.44213100 |
| C | 3.22267000 | 0.58741500 | -0.71697600 |
| C | 5.68740700 | -0.77166900 | -0.15531000 |
| H | 4.47930100 | -2.07635100 | 1.11794500 |
| C | 4.45117600 | 1.01380000 | -1.30086600 |
| C | 5.65884400 | 0.34607200 | -1.02379100 |
| H | 6.63136400 | -1.29939900 | 0.04850700 |
| H | 4.43786900 | 1.87484100 | -1.98644100 |
| H | 6.59008000 | 0.69546100 | -1.49801700 |
| C | -1.77994800 | 2.57490000 | 0.26072400 |
| H | -2.79883200 | 3.00635100 | 0.28590400 |
| H | -1.06944200 | 3.34628200 | -0.10292000 |
| C | -1.35384400 | 2.00719400 | 1.62751500 |
| H | -2.10044600 | 1.26958800 | 1.98388700 |
| H | -1.27817400 | 2.82594200 | 2.38085700 |
| N | -0.04031900 | 1.29152500 | 1.47602300 |
| H | 0.67451400 | 2.00074800 | 1.21262200 |
| C | 0.41175300 | 0.64421200 | 2.75443400 |
| H | 0.46441400 | 1.38789600 | 3.58396700 |
| H | -0.34253500 | -0.12462800 | 3.01723800 |
| C | 1.79669200 | 0.00666900 | 2.53824400 |
| H | 2.57744700 | 0.77459500 | 2.36796500 |
| H | 2.09120700 | -0.62066500 | 3.40158400 |
| H | 0.99256200 | -0.81485500 | -2.32650000 |
| H | -0.64146200 | -1.05512700 | -2.42488100 |
| N | 0.17888000 | -1.17920200 | -1.79938200 |
| N | 0.43349600 | -2.61750600 | -1.57437800 |
| H | -0.25735400 | -2.88339400 | -0.84710000 |
| H | 0.17784400 | -3.13707100 | -2.43408400 |

**$[Fe^{II}(N_HS_4)(N_2H_4)]$. S = 1. E** = -3630.41818962

| | | | |
|---|---|---|---|
| C | 4.38460200 | 1.67125900 | 0.58021600 |
| C | 3.24388000 | 0.91827200 | 0.16488400 |
| C | 3.48600400 | -0.23139100 | -0.65413300 |
| C | 4.80412700 | -0.58307400 | -1.04661700 |
| C | 5.90945200 | 0.17808300 | -0.62297000 |
| C | 5.68784900 | 1.30967000 | 0.19779800 |
| H | 4.22049600 | 2.55548800 | 1.21441200 |
| H | 4.94953900 | -1.46212100 | -1.69369500 |
| H | 6.92764900 | -0.10213400 | -0.93270100 |
| H | 6.54030300 | 1.91879600 | 0.53883500 |
| S | 2.11949700 | -1.27170000 | -1.26033900 |
| S | 1.61986400 | 1.48595400 | 0.67650100 |
| Fe | -0.05385000 | 0.02494200 | -0.19117000 |
| S | -1.76291600 | -1.36468700 | -1.10133100 |
| S | -2.05584500 | 0.95590700 | 1.35410400 |
| C | -3.46620300 | 0.42580800 | 0.33843900 |
| C | -4.72213500 | 1.03560000 | 0.58699500 |
| C | -3.31135700 | -0.53126600 | -0.71713100 |
| C | -5.84977600 | 0.70079500 | -0.18710300 |
| H | -4.80192000 | 1.77597900 | 1.39726200 |
| C | -4.47166900 | -0.85878600 | -1.47796200 |
| C | -5.71664700 | -0.25624900 | -1.21962400 |
| H | -6.82060100 | 1.17855200 | 0.01455400 |
| H | -4.37170200 | -1.59584100 | -2.28907400 |
| H | -6.58926700 | -0.53098700 | -1.83364100 |
| C | 2.04200300 | -2.57054800 | 0.08066900 |
| H | 3.04906800 | -3.01750900 | 0.19594200 |
| H | 1.35908000 | -3.35253600 | -0.31461400 |
| C | 1.54262500 | -2.01818900 | 1.43029600 |
| H | 2.22948000 | -1.23306200 | 1.80416500 |
| H | 1.52202100 | -2.84578300 | 2.17888200 |
| N | 0.17818500 | -1.40777000 | 1.29265600 |
| H | -0.47365600 | -2.14817800 | 0.96539900 |
| C | -0.35126800 | -0.92856400 | 2.61977900 |
| H | -0.26269400 | -1.73988900 | 3.38049200 |
| H | 0.28685500 | -0.08125400 | 2.94588800 |
| C | -1.83283800 | -0.50141200 | 2.52163300 |
| H | -2.46546600 | -1.33006200 | 2.14545600 |
| H | -2.20110500 | -0.20820600 | 3.52478300 |
| H | -1.21086300 | 1.03511100 | -2.23000200 |
| H | 0.40117200 | 1.43496100 | -2.32384300 |
| N | -0.41308200 | 1.39852700 | -1.68022900 |
| N | -0.78939100 | 2.75795500 | -1.24130600 |
| H | -0.05999600 | 3.00747400 | -0.54781200 |
| H | -0.65974900 | 3.40407700 | -2.04208300 |

**[$Fe^{II}(N_HS_4)(N_2H_4)$]. S = 2. E** = -3630.43083146

| | | | |
|---|---|---|---|
| C | 4.45179400 | 1.14852700 | 1.25808600 |
| C | 3.27167200 | 0.68421100 | 0.59956400 |
| C | 3.45925300 | -0.02443200 | -0.63103200 |
| C | 4.75511800 | -0.23168300 | -1.17244300 |
| C | 5.89775900 | 0.23317400 | -0.49598500 |
| C | 5.73415100 | 0.92690500 | 0.72699000 |
| H | 4.33323500 | 1.69245900 | 2.20713500 |
| H | 4.85472400 | -0.76008200 | -2.13355900 |
| H | 6.89975300 | 0.06543600 | -0.91907600 |
| H | 6.61662600 | 1.30359700 | 1.26859300 |
| S | 2.04209000 | -0.63171600 | -1.60066000 |
| S | 1.68070700 | 1.01460300 | 1.36388000 |
| Fe | -0.04183300 | 0.19421800 | -0.17463400 |
| S | -1.85971400 | -0.60458500 | -1.69193300 |
| S | -2.02034800 | 0.31277500 | 1.58808600 |
| C | -3.48244800 | 0.26150900 | 0.50707800 |
| C | -4.72252700 | 0.65316100 | 1.07468400 |
| C | -3.38298900 | -0.12883900 | -0.86912800 |
| C | -5.89858900 | 0.64611800 | 0.30195000 |
| H | -4.75085400 | 0.96406300 | 2.13023100 |
| C | -4.59678200 | -0.13415600 | -1.62202700 |
| C | -5.82564100 | 0.24325200 | -1.05248100 |
| H | -6.85716500 | 0.95090600 | 0.74848000 |
| H | -4.54688000 | -0.43619600 | -2.67891200 |
| H | -6.73605200 | 0.23016100 | -1.67312900 |
| C | 1.90486700 | -2.41025900 | -1.02815600 |
| H | 2.89568400 | -2.88781900 | -1.15921400 |
| H | 1.19540900 | -2.87835600 | -1.74284600 |
| C | 1.41781700 | -2.56450400 | 0.42966300 |
| H | 2.12016500 | -2.05642900 | 1.12180900 |
| H | 1.40510700 | -3.65099900 | 0.69148600 |
| N | 0.07294900 | -1.94608100 | 0.61068200 |
| H | -0.60798500 | -2.40399800 | -0.02433700 |
| C | -0.43781100 | -2.03357100 | 2.01151600 |
| H | -0.42817000 | -3.08782500 | 2.38197100 |
| H | 0.24829700 | -1.44179300 | 2.65341000 |
| C | -1.88227500 | -1.49660300 | 2.10834800 |
| H | -2.56845600 | -2.06580500 | 1.44998100 |
| H | -2.24603100 | -1.57929100 | 3.15126400 |
| H | -1.16407200 | 2.17717800 | -1.64305800 |
| H | 0.46107000 | 2.23035400 | -1.96922200 |
| N | -0.23207100 | 2.18837800 | -1.19717900 |
| N | -0.16256300 | 3.35818100 | -0.31219800 |
| H | 0.56367900 | 3.11579300 | 0.38864000 |
| H | 0.18738900 | 4.17154500 | -0.84804400 |

**[$Fe^{II}(N_HS_4)(NH_3)$]. S = 0. E =** -3575.44413536

| | | | |
|---|---|---|---|
| C | -4.37221800 | -1.50327200 | -0.79447800 |
| C | -3.14823300 | -0.80368600 | -0.56402300 |
| C | -3.24159200 | 0.60149500 | -0.33886700 |
| C | -4.48225700 | 1.28387600 | -0.36745100 |
| C | -5.67424000 | 0.56884500 | -0.59108100 |
| C | -5.60675400 | -0.83099700 | -0.80425000 |
| H | -4.33120300 | -2.58950000 | -0.96772500 |
| H | -4.50425300 | 2.37509000 | -0.21526200 |
| H | -6.64139600 | 1.09355000 | -0.60926000 |
| H | -6.53135700 | -1.40181200 | -0.98714400 |
| S | -1.70878800 | 1.55672800 | -0.06578700 |
| S | -1.58488500 | -1.67323600 | -0.57445700 |
| Fe | 0.03628900 | 0.05510600 | -0.21035200 |
| S | 1.69899700 | 1.76912700 | 0.03212500 |
| S | 1.74199500 | -1.48937400 | -0.11076300 |
| C | 3.28386900 | -0.54208000 | -0.33489800 |
| C | 4.49238900 | -1.23191900 | -0.59439200 |
| C | 3.23038900 | 0.88207800 | -0.24538300 |
| C | 5.69340200 | -0.51221500 | -0.74859700 |
| H | 4.48159200 | -2.33054300 | -0.67342200 |
| C | 4.46320500 | 1.58486800 | -0.39679300 |
| C | 5.66741400 | 0.90044000 | -0.64236000 |
| H | 6.63590700 | -1.04222000 | -0.95294400 |
| H | 4.45358900 | 2.68334000 | -0.32784400 |
| H | 6.59983000 | 1.47522100 | -0.76254100 |
| C | -1.73916800 | 1.72122100 | 1.80227500 |
| H | -2.75069200 | 2.04080100 | 2.11792100 |
| H | -1.01423900 | 2.52727400 | 2.04046400 |
| C | -1.32830800 | 0.38071700 | 2.44038800 |
| H | -2.08520800 | -0.39740700 | 2.21589300 |
| H | -1.24804800 | 0.48640400 | 3.54759000 |
| N | -0.02237100 | -0.06777600 | 1.84512400 |
| H | 0.70411500 | 0.62210100 | 2.12896500 |
| C | 0.40246000 | -1.41772800 | 2.35061700 |
| H | 0.45931900 | -1.43069900 | 3.46443900 |
| H | -0.36938800 | -2.14490100 | 2.02721200 |
| C | 1.77856100 | -1.78093500 | 1.76027600 |
| H | 2.58520000 | -1.13688700 | 2.16374300 |
| H | 2.03307200 | -2.84098900 | 1.95172800 |
| H | 1.14273100 | 0.14601200 | -2.61564900 |
| H | -0.40825100 | -0.45711300 | -2.72764300 |
| H | -0.12735300 | 1.18112200 | -2.58836500 |
| N | 0.17978800 | 0.25238300 | -2.25152500 |

**[$Fe^{II}(N_HS_4)(NH_3)$]. S = 1. E =** -3575.43375996

| | | | |
|---|---|---|---|
| C | 4.35235900 | 1.40382300 | -0.97351900 |
| C | 3.13881600 | 0.72450300 | -0.65830000 |
| C | 3.24320200 | -0.63238700 | -0.23978800 |
| C | 4.49082000 | -1.29642100 | -0.16442100 |
| C | 5.67642400 | -0.60303200 | -0.47791500 |
| C | 5.59595700 | 0.75189300 | -0.88192400 |
| H | 4.29963000 | 2.45482700 | -1.29612600 |
| H | 4.52435400 | -2.35529900 | 0.13843300 |
| H | 6.64950600 | -1.11344700 | -0.41749600 |
| H | 6.51539400 | 1.30334100 | -1.13583200 |
| S | 1.71537500 | -1.54998900 | 0.16064100 |
| S | 1.56030100 | 1.56817800 | -0.80395900 |
| Fe | -0.03598200 | -0.10664700 | -0.28408600 |
| S | -1.69094100 | -1.77764300 | 0.15637100 |
| S | -1.73042900 | 1.46184500 | -0.26700400 |
| C | -3.28000900 | 0.50579400 | -0.36035900 |
| C | -4.49465600 | 1.17607600 | -0.64022800 |
| C | -3.22857400 | -0.90359900 | -0.14750900 |
| C | -5.70188500 | 0.45124400 | -0.69070200 |
| H | -4.48444600 | 2.26314000 | -0.81723300 |
| C | -4.46533500 | -1.61181600 | -0.19573300 |
| C | -5.67639300 | -0.94597600 | -0.46162500 |
| H | -6.64933400 | 0.96664700 | -0.90921300 |
| H | -4.45509600 | -2.69997100 | -0.03055300 |
| H | -6.61343900 | -1.52437100 | -0.49997900 |
| C | 1.75890700 | -1.45630400 | 2.04008300 |
| H | 2.77249100 | -1.75256700 | 2.37297000 |
| H | 1.03491500 | -2.22676300 | 2.37830300 |
| C | 1.37029400 | -0.05337300 | 2.56310300 |
| H | 2.10500900 | 0.69758000 | 2.20596500 |
| H | 1.40089000 | -0.05827600 | 3.68074200 |
| N | 0.03802200 | 0.32050200 | 2.02527900 |
| H | -0.68031500 | -0.33534700 | 2.37928500 |
| C | -0.39810400 | 1.72101400 | 2.25127400 |
| H | -0.49816800 | 1.97546100 | 3.33551800 |
| H | 0.37612500 | 2.38461000 | 1.81327200 |
| C | -1.76438600 | 1.96189400 | 1.56820500 |
| H | -2.56298400 | 1.35191500 | 2.03599700 |
| H | -2.05131900 | 3.02968800 | 1.62451000 |
| H | -1.19827600 | -0.54029100 | -2.92362700 |
| H | 0.34049400 | 0.06740800 | -3.10443100 |
| H | 0.09626000 | -1.55034700 | -2.80185200 |
| N | -0.23226500 | -0.60098200 | -2.56285600 |

**[$Fe^{II}(N_HS_4)(NH_3)$]. S = 2. E =** -3575.44382353

| | | | |
|---|---|---|---|
| C | 4.45522900 | 1.47195800 | -0.97189600 |
| C | 3.26920500 | 0.73482600 | -0.66486300 |
| C | 3.45280400 | -0.61319300 | -0.21456300 |
| C | 4.74683100 | -1.18659600 | -0.10527900 |
| C | 5.89375600 | -0.43169700 | -0.41107600 |
| C | 5.73538100 | 0.90639000 | -0.84608100 |
| H | 4.34176700 | 2.51038600 | -1.31746500 |
| H | 4.84164900 | -2.23441300 | 0.22055700 |
| H | 6.89448900 | -0.88057100 | -0.32064600 |
| H | 6.62121300 | 1.51225700 | -1.09594200 |
| S | 2.03038800 | -1.68149800 | 0.17634300 |
| S | 1.68121600 | 1.54230800 | -0.86944800 |
| Fe | -0.03297500 | -0.14951600 | -0.36557400 |
| S | -1.85861900 | -1.75966700 | 0.13971700 |
| S | -2.03400900 | 1.64019000 | -0.13748900 |
| C | -3.49160300 | 0.56074500 | -0.27691200 |
| C | -4.73892500 | 1.18682400 | -0.53672300 |
| C | -3.38335200 | -0.86562800 | -0.16060100 |
| C | -5.91272000 | 0.42208600 | -0.66789900 |
| H | -4.77417700 | 2.28289200 | -0.63277100 |
| C | -4.59625100 | -1.61039400 | -0.29089600 |
| C | -5.83062800 | -0.98491200 | -0.53738200 |
| H | -6.87638300 | 0.91458400 | -0.86778700 |
| H | -4.53996000 | -2.70549500 | -0.20014600 |
| H | -6.73896800 | -1.60106300 | -0.63507300 |
| C | 1.88366500 | -1.47087400 | 2.03208200 |
| H | 2.87043100 | -1.70097600 | 2.47978800 |
| H | 1.16610600 | -2.25727700 | 2.34816100 |
| C | 1.40763300 | -0.06677400 | 2.46488100 |
| H | 2.12075200 | 0.70312200 | 2.10514900 |
| H | 1.38820700 | -0.02028800 | 3.58144800 |
| N | 0.06896300 | 0.24445900 | 1.88437300 |
| H | -0.62253000 | -0.45408100 | 2.21659900 |
| C | -0.42039800 | 1.61191500 | 2.22719700 |
| H | -0.39638500 | 1.78829300 | 3.33061600 |
| H | 0.26933000 | 2.34003500 | 1.75045800 |
| C | -1.86850700 | 1.82166600 | 1.73399800 |
| H | -2.55872800 | 1.07855700 | 2.18086800 |
| H | -2.21764500 | 2.83642300 | 2.00824300 |
| H | -1.16792200 | -0.54013300 | -2.89503700 |
| H | 0.41647300 | -0.02645800 | -3.07754100 |
| H | 0.06378300 | -1.62475200 | -2.72607500 |
| N | -0.20617900 | -0.64632100 | -2.53271500 |

**[$Fe^{III}$(por)Cl]. S = 3/2. E =** -2702.04475616

| | | | |
|---|---|---|---|
| Fe | 0.000000 | 0.000000 | 0.059067 |
| Cl | 0.000002 | -0.000002 | 2.370727 |
| N | 1.370205 | -1.468637 | -0.241927 |
| N | -1.370206 | 1.468639 | -0.241920 |
| N | -1.468640 | -1.370203 | -0.241928 |
| C | 1.139337 | -2.846998 | -0.251464 |
| C | -0.120117 | -3.456815 | -0.250660 |
| H | -0.158133 | -4.555684 | -0.255757 |
| C | -1.334281 | -2.761286 | -0.251565 |
| C | -2.847001 | -1.139336 | -0.251394 |
| C | -3.577793 | -2.396265 | -0.276381 |
| H | -4.671059 | -2.478140 | -0.293656 |
| C | -2.639009 | -3.402805 | -0.276513 |
| H | -2.796825 | -4.487718 | -0.293874 |
| C | -3.456818 | 0.120118 | -0.250528 |
| H | -4.555687 | 0.158134 | -0.255567 |
| C | -2.761289 | 1.334281 | -0.251480 |
| C | -1.139338 | 2.846999 | -0.251462 |
| C | -2.396267 | 3.577792 | -0.276422 |
| H | -2.478143 | 4.671057 | -0.293746 |
| C | -3.402808 | 2.639008 | -0.276448 |
| H | -4.487722 | 2.796824 | -0.293757 |
| C | 2.396267 | -3.577790 | -0.276417 |
| H | 2.478142 | -4.671056 | -0.293737 |
| C | 3.402808 | -2.639006 | -0.276443 |
| H | 4.487722 | -2.796823 | -0.293749 |
| C | 2.761288 | -1.334279 | -0.251481 |
| C | 3.456817 | -0.120116 | -0.250528 |
| H | 4.555686 | -0.158132 | -0.255565 |
| C | 2.847000 | 1.139338 | -0.251393 |
| N | 1.468638 | 1.370205 | -0.241927 |
| C | 1.334280 | 2.761287 | -0.251565 |
| C | 2.639007 | 3.402807 | -0.276513 |
| H | 2.796823 | 4.487720 | -0.293875 |
| C | 3.577792 | 2.396266 | -0.276381 |
| H | 4.671058 | 2.478142 | -0.293655 |
| C | 0.120116 | 3.456817 | -0.250660 |
| H | 0.158132 | 4.555685 | -0.255759 |

**[$Fe^{III}$(por)Cl]. S = 5/2. E =** -2702.04129532

| | | | |
|---|---|---|---|
| Fe | 0.000000 | 0.000000 | 0.255824 |
| Cl | 0.000001 | -0.000003 | 2.510598 |
| N | -0.962353 | 1.808896 | -0.253748 |
| N | 0.962353 | -1.808895 | -0.253749 |
| N | 1.808895 | 0.962353 | -0.253748 |
| C | -0.372257 | 3.072426 | -0.288624 |
| C | 1.013525 | 3.317291 | -0.294926 |
| H | 1.335018 | 4.369288 | -0.322862 |
| C | 2.025901 | 2.339795 | -0.288623 |
| C | 3.072426 | 0.372257 | -0.288618 |
| C | 4.103657 | 1.402203 | -0.344646 |
| H | 5.181766 | 1.205475 | -0.385595 |
| C | 3.456375 | 2.619205 | -0.344683 |
| H | 3.895862 | 3.623136 | -0.385580 |
| C | 3.317290 | -1.013525 | -0.294916 |
| H | 4.369288 | -1.335018 | -0.322848 |
| C | 2.339795 | -2.025900 | -0.288619 |
| C | 0.372256 | -3.072426 | -0.288626 |
| C | 1.402202 | -4.103657 | -0.344652 |
| H | 1.205475 | -5.181765 | -0.385606 |
| C | 2.619204 | -3.456374 | -0.344684 |
| H | 3.623136 | -3.895862 | -0.385579 |
| C | -1.402202 | 4.103657 | -0.344648 |
| H | -1.205475 | 5.181766 | -0.385600 |
| C | -2.619205 | 3.456375 | -0.344680 |
| H | -3.623136 | 3.895862 | -0.385574 |
| C | -2.339795 | 2.025901 | -0.288616 |
| C | -3.317291 | 1.013526 | -0.294914 |
| H | -4.369288 | 1.335019 | -0.322845 |
| C | -3.072427 | -0.372256 | -0.288617 |
| N | -1.808896 | -0.962353 | -0.253748 |
| C | -2.025901 | -2.339795 | -0.288624 |
| C | -3.456375 | -2.619204 | -0.344683 |
| H | -3.895863 | -3.623135 | -0.385580 |
| C | -4.103658 | -1.402202 | -0.344646 |
| H | -5.181766 | -1.205474 | -0.385595 |
| C | -1.013526 | -3.317290 | -0.294927 |
| H | -1.335018 | -4.369287 | -0.322864 |

**$[Fe^{III}(pyPepS)_2]^-$. S = 1/2. E** = -3340.71924212

| | | | |
|---|---|---|---|
| Fe | 4.890611 | 1.865257 | 3.158600 |
| S | 6.031087 | 3.555711 | 2.116678 |
| S | 5.963934 | 1.987673 | 5.177780 |
| O | 1.372402 | 3.423703 | 4.672451 |
| O | 6.721816 | -1.074726 | 0.899951 |
| N | 3.538435 | 3.229522 | 3.673421 |
| N | 3.521702 | 0.626510 | 3.977048 |
| N | 4.091504 | 1.328438 | 1.383357 |
| N | 6.234652 | 0.494356 | 2.640100 |
| C | 5.011837 | 4.900672 | 2.716513 |
| C | 5.355979 | 6.249372 | 2.448410 |
| C | 4.541462 | 7.310026 | 2.894315 |
| C | 3.361825 | 7.027164 | 3.619619 |
| C | 2.994519 | 5.693939 | 3.897580 |
| C | 3.810870 | 4.611632 | 3.453278 |
| C | 2.381903 | 2.785915 | 4.269990 |
| C | 2.407227 | 1.275143 | 4.431171 |
| C | 1.337343 | 0.580706 | 5.036518 |
| C | 1.425111 | -0.814905 | 5.181949 |
| C | 2.584389 | -1.479118 | 4.717180 |
| C | 3.609538 | -0.723356 | 4.119402 |
| C | 7.355379 | 0.947707 | 4.741952 |
| C | 8.437575 | 0.767186 | 5.639485 |
| C | 9.521264 | -0.072899 | 5.312174 |
| C | 9.531143 | -0.747957 | 4.070474 |
| C | 8.465528 | -0.588301 | 3.160177 |
| C | 7.363716 | 0.260095 | 3.478698 |
| C | 6.044088 | -0.165262 | 1.448953 |
| C | 4.795600 | 0.343725 | 0.748054 |
| C | 4.395118 | -0.166067 | -0.506164 |
| C | 3.244778 | 0.359269 | -1.120430 |
| C | 2.525194 | 1.383567 | -0.461547 |
| C | 2.979880 | 1.839794 | 0.789278 |
| H | 8.417615 | 1.302287 | 6.602450 |
| H | 10.355100 | -0.197255 | 6.023413 |
| H | 10.374771 | -1.405859 | 3.802839 |
| H | 8.458025 | -1.112114 | 2.198245 |
| H | 2.911282 | -0.015416 | -2.101418 |
| H | 1.625686 | 1.831926 | -0.908698 |
| H | 5.014946 | -0.956972 | -0.950699 |
| H | 2.469687 | 2.641971 | 1.340810 |
| H | 0.477224 | 1.174832 | 5.374954 |
| H | 0.607961 | -1.382603 | 5.655162 |
| H | 2.701703 | -2.568114 | 4.819214 |
| H | 4.538083 | -1.182917 | 3.752547 |
| H | 2.078833 | 5.461742 | 4.451975 |
| H | 2.718228 | 7.848988 | 3.975374 |
| H | 4.828877 | 8.352693 | 2.677873 |
| H | 6.280090 | 6.454684 | 1.884585 |

**$[Fe^{III}(pyPepS)_2]^-$. S = 3/2. E** = -3340.68892253

| | | | |
|---|---|---|---|
| Fe | 5.266479 | 2.174509 | 3.329221 |
| S | 6.131336 | 4.213479 | 2.411098 |
| S | 6.668062 | 1.997214 | 5.266346 |
| O | 1.472063 | 3.376984 | 4.958201 |
| O | 6.850484 | -0.843621 | 0.706449 |
| N | 3.569725 | 3.401080 | 3.813651 |
| N | 3.786333 | 0.729799 | 4.239107 |
| N | 4.340729 | 1.642992 | 1.337643 |
| N | 6.460868 | 0.535294 | 2.618923 |
| C | 4.679496 | 5.233954 | 2.634332 |
| C | 4.676645 | 6.555754 | 2.114037 |
| C | 3.575479 | 7.414448 | 2.292284 |
| C | 2.448494 | 6.952475 | 3.007829 |
| C | 2.422293 | 5.646029 | 3.536111 |
| C | 3.527329 | 4.754637 | 3.360592 |
| C | 2.527067 | 2.847525 | 4.520510 |
| C | 2.697536 | 1.347152 | 4.768433 |
| C | 1.723020 | 0.637909 | 5.508727 |
| C | 1.889259 | -0.745404 | 5.698759 |
| C | 3.022100 | -1.382991 | 5.144139 |
| C | 3.949231 | -0.605617 | 4.423475 |
| C | 7.572969 | 0.527484 | 4.797220 |
| C | 8.480475 | -0.051224 | 5.724260 |
| C | 9.245236 | -1.186244 | 5.394770 |
| C | 9.109750 | -1.761277 | 4.111609 |
| C | 8.217606 | -1.209168 | 3.170427 |
| C | 7.425781 | -0.060501 | 3.486765 |
| C | 6.246609 | 0.056410 | 1.346927 |
| C | 5.063267 | 0.724792 | 0.643640 |
| C | 4.733197 | 0.362437 | -0.683196 |
| C | 3.624321 | 0.971455 | -1.297029 |
| C | 2.874843 | 1.923938 | -0.570043 |
| C | 3.270845 | 2.233398 | 0.745500 |
| H | 8.575258 | 0.415581 | 6.717227 |
| H | 9.943334 | -1.614819 | 6.133262 |
| H | 9.705119 | -2.646775 | 3.832505 |
| H | 8.117843 | -1.646309 | 2.172201 |
| H | 3.344053 | 0.710823 | -2.330477 |
| H | 2.000823 | 2.425878 | -1.010939 |
| H | 5.362705 | -0.387953 | -1.179458 |
| H | 2.738233 | 2.979389 | 1.352800 |
| H | 0.869737 | 1.204491 | 5.904370 |
| H | 1.147613 | -1.324181 | 6.272754 |
| H | 3.192877 | -2.462585 | 5.268864 |
| H | 4.858216 | -1.042452 | 3.985398 |
| H | 1.554861 | 5.288704 | 4.099476 |
| H | 1.578758 | 7.612606 | 3.163580 |
| H | 3.599967 | 8.436562 | 1.878675 |
| H | 5.567194 | 6.898310 | 1.564080 |

**$[Fe^{III}(pyPepS)_2]^-$. S = 5/2. E** = -3340.70606078

| | | | |
|---|---|---|---|
| Fe | 5.266479 | 2.174509 | 3.329221 |
| S | 6.131336 | 4.213479 | 2.411098 |
| S | 6.668062 | 1.997214 | 5.266346 |
| O | 1.472063 | 3.376984 | 4.958201 |
| O | 6.850484 | -0.843621 | 0.706449 |
| N | 3.569725 | 3.401080 | 3.813651 |
| N | 3.786333 | 0.729799 | 4.239107 |
| N | 4.340729 | 1.642992 | 1.337643 |
| N | 6.460868 | 0.535294 | 2.618923 |
| C | 4.679496 | 5.233954 | 2.634332 |
| C | 4.676645 | 6.555754 | 2.114037 |
| C | 3.575479 | 7.414448 | 2.292284 |
| C | 2.448494 | 6.952475 | 3.007829 |
| C | 2.422293 | 5.646029 | 3.536111 |
| C | 3.527329 | 4.754637 | 3.360592 |
| C | 2.527067 | 2.847525 | 4.520510 |
| C | 2.697536 | 1.347152 | 4.768433 |
| C | 1.723020 | 0.637909 | 5.508727 |
| C | 1.889259 | -0.745404 | 5.698759 |
| C | 3.022100 | -1.382991 | 5.144139 |
| C | 3.949231 | -0.605617 | 4.423475 |
| C | 7.572969 | 0.527484 | 4.797220 |
| C | 8.480475 | -0.051224 | 5.724260 |
| C | 9.245236 | -1.186244 | 5.394770 |
| C | 9.109750 | -1.761277 | 4.111609 |
| C | 8.217606 | -1.209168 | 3.170427 |
| C | 7.425781 | -0.060501 | 3.486765 |
| C | 6.246609 | 0.056410 | 1.346927 |
| C | 5.063267 | 0.724792 | 0.643640 |
| C | 4.733197 | 0.362437 | -0.683196 |
| C | 3.624321 | 0.971455 | -1.297029 |
| C | 2.874843 | 1.923938 | -0.570043 |
| C | 3.270845 | 2.233398 | 0.745500 |
| H | 8.575258 | 0.415581 | 6.717227 |
| H | 9.943334 | -1.614819 | 6.133262 |
| H | 9.705119 | -2.646775 | 3.832505 |
| H | 8.117843 | -1.646309 | 2.172201 |
| H | 3.344053 | 0.710823 | -2.330477 |
| H | 2.000823 | 2.425878 | -1.010939 |
| H | 5.362705 | -0.387953 | -1.179458 |
| H | 2.738233 | 2.979389 | 1.352800 |
| H | 0.869737 | 1.204491 | 5.904370 |
| H | 1.147613 | -1.324181 | 6.272754 |
| H | 3.192877 | -2.462585 | 5.268864 |
| H | 4.858216 | -1.042452 | 3.985398 |
| H | 1.554861 | 5.288704 | 4.099476 |
| H | 1.578758 | 7.612606 | 3.163580 |
| H | 3.599967 | 8.436562 | 1.878675 |
| H | 5.567194 | 6.898310 | 1.564080 |

**[$Fe^{III}$(tsalen)Cl]. S = 1/2. E =** -3236.84094505

| | | | |
|---|---|---|---|
| Fe | -0.259983 | 1.148059 | -0.751389 |
| S | 1.219167 | 1.864067 | -2.273658 |
| N | 0.995995 | -0.111901 | 0.101385 |
| C | 2.275982 | -0.362223 | -0.067678 |
| H | 2.735545 | -1.103164 | 0.617862 |
| C | 2.821207 | 1.143867 | -2.063802 |
| C | 3.826353 | 1.556816 | -2.992783 |
| C | 3.189917 | 0.190271 | -1.051540 |
| C | 5.136973 | 1.067981 | -2.920230 |
| H | 3.548272 | 2.283058 | -3.770837 |
| C | 4.539883 | -0.294188 | -1.000657 |
| C | 5.505955 | 0.133952 | -1.913307 |
| H | 5.886152 | 1.415142 | -3.648892 |
| H | 4.807590 | -1.021165 | -0.216341 |
| H | 6.536292 | -0.246813 | -1.854721 |
| C | 0.327975 | -0.809412 | 1.239425 |
| H | 0.281705 | -0.100125 | 2.092908 |
| H | 0.888544 | -1.718200 | 1.546500 |
| C | -1.093653 | -1.159684 | 0.772627 |
| H | -1.723295 | -1.520323 | 1.614477 |
| H | -1.050558 | -1.959393 | -0.000037 |
| N | -1.664351 | 0.070050 | 0.151880 |
| S | -1.783879 | 2.082431 | -2.121571 |
| C | -2.930697 | 0.322176 | 0.385978 |
| H | -3.468128 | -0.367682 | 1.068749 |
| C | -3.745495 | 1.389759 | -0.171342 |
| C | -5.056101 | 1.553508 | 0.383458 |
| C | -3.340378 | 2.207269 | -1.283484 |
| C | -5.950135 | 2.500000 | -0.124993 |
| H | -5.351833 | 0.916332 | 1.233046 |
| C | -4.281676 | 3.146144 | -1.802419 |
| C | -5.554018 | 3.297364 | -1.232312 |
| H | -6.948175 | 2.620402 | 0.321618 |
| H | -3.980076 | 3.768777 | -2.657512 |
| H | -6.249051 | 4.043432 | -1.648392 |
| Cl | -0.169040 | 2.573984 | 1.000620 |

**[$Fe^{III}$(tsalen)Cl]. S = 3/2. E =** -3236.85511442

| | | | |
|---|---|---|---|
| Fe | -0.223025 | 1.299783 | -0.656050 |
| S | 1.230208 | 1.807895 | -2.342495 |
| N | 1.024204 | -0.066298 | 0.132272 |
| C | 2.288706 | -0.358637 | -0.063954 |
| H | 2.738122 | -1.113463 | 0.614368 |
| C | 2.833148 | 1.101195 | -2.093189 |
| C | 3.842314 | 1.490614 | -3.027322 |
| C | 3.201918 | 0.167131 | -1.062066 |
| C | 5.151618 | 0.998491 | -2.941740 |
| H | 3.570731 | 2.201642 | -3.821472 |
| C | 4.548806 | -0.325227 | -1.001254 |
| C | 5.518802 | 0.081385 | -1.919155 |
| H | 5.901987 | 1.330730 | -3.676262 |
| H | 4.810880 | -1.038985 | -0.203045 |
| H | 6.547554 | -0.301774 | -1.853104 |
| C | 0.330212 | -0.757960 | 1.256862 |
| H | 0.253217 | -0.042267 | 2.102692 |
| H | 0.889389 | -1.659533 | 1.588118 |
| C | -1.077121 | -1.130295 | 0.754690 |
| H | -1.712879 | -1.517294 | 1.580682 |
| H | -0.999100 | -1.919370 | -0.026401 |
| N | -1.667225 | 0.092270 | 0.137617 |
| S | -1.755535 | 2.299765 | -2.005097 |
| C | -2.943413 | 0.312684 | 0.344714 |
| H | -3.480896 | -0.401983 | 1.003227 |
| C | -3.771928 | 1.378051 | -0.194262 |
| C | -5.110604 | 1.457278 | 0.312188 |
| C | -3.348629 | 2.285242 | -1.227814 |
| C | -6.020187 | 2.401143 | -0.171474 |
| H | -5.415619 | 0.752938 | 1.103453 |
| C | -4.303392 | 3.225064 | -1.719902 |
| C | -5.605455 | 3.286842 | -1.201967 |
| H | -7.040436 | 2.454198 | 0.235812 |
| H | -3.991041 | 3.920477 | -2.512778 |
| H | -6.309263 | 4.035218 | -1.598812 |
| Cl | 0.016923 | 2.773614 | 1.115553 |

**[$Fe^{III}$(tsalen)Cl]. S = 5/2. E =** -3236.84665703

| | | | |
|---|---|---|---|
| Fe | -0.198755 | 1.680086 | -0.368084 |
| S | 1.573038 | 2.449812 | -1.771715 |
| N | 1.100009 | 0.097667 | 0.302289 |
| C | 2.286536 | -0.333575 | -0.047737 |
| H | 2.689768 | -1.210392 | 0.505272 |
| C | 2.956663 | 1.349109 | -1.884844 |
| C | 3.954984 | 1.692414 | -2.849646 |
| C | 3.180235 | 0.171790 | -1.082263 |
| C | 5.113281 | 0.922426 | -3.021020 |
| H | 3.791731 | 2.589646 | -3.464268 |
| C | 4.379666 | -0.592149 | -1.276028 |
| C | 5.336427 | -0.235678 | -2.229001 |
| H | 5.855363 | 1.223359 | -3.777132 |
| H | 4.536804 | -1.486150 | -0.650286 |
| H | 6.246213 | -0.839457 | -2.361056 |
| C | 0.406292 | -0.619293 | 1.404831 |
| H | 0.294162 | 0.082249 | 2.259131 |
| H | 0.982329 | -1.507948 | 1.746837 |
| C | -0.989438 | -1.036321 | 0.884504 |
| H | -1.617255 | -1.446528 | 1.707692 |
| H | -0.868113 | -1.828400 | 0.111873 |
| N | -1.617519 | 0.147136 | 0.246587 |
| S | -1.735422 | 2.203620 | -2.089738 |
| C | -2.913903 | 0.288409 | 0.356317 |
| H | -3.466426 | -0.449972 | 0.978817 |
| C | -3.754850 | 1.315654 | -0.254227 |
| C | -5.104568 | 1.374713 | 0.222672 |
| C | -3.339681 | 2.206895 | -1.310386 |
| C | -6.028109 | 2.288677 | -0.294011 |
| H | -5.408387 | 0.682107 | 1.024736 |
| C | -4.307226 | 3.116927 | -1.831043 |
| C | -5.618387 | 3.164357 | -1.332906 |
| H | -7.056102 | 2.323623 | 0.095789 |
| H | -3.997709 | 3.796066 | -2.638723 |
| H | -6.332459 | 3.887907 | -1.756500 |
| Cl | -0.406199 | 3.051777 | 1.405902 |

**[$Fe^{III}$(N($CH_2$-*o*-$C_6H_4S)_3$(1-MeIm)]. S = 1/2. E =** -3575.96455453

| | | | |
|---|---|---|---|
| Fe | 0.78580600 | -0.07818800 | -0.24163400 |
| S | 1.16397400 | -1.84535300 | 1.10571300 |
| S | 1.28410400 | 2.08530800 | 0.16755500 |
| S | 0.43104500 | -0.96875800 | -2.26906300 |
| N | -1.23026600 | 0.10552000 | 0.22383300 |
| N | 2.77784100 | -0.15123400 | -0.68620600 |
| N | 4.99700200 | -0.20354400 | -0.40810200 |
| C | -1.89602800 | -1.23597900 | 0.57633700 |
| C | -1.53424900 | -1.80700200 | 1.93842900 |
| C | -2.55224600 | -2.07707200 | 2.88673700 |
| C | -2.25823800 | -2.68233400 | 4.12540900 |
| C | -0.91942000 | -3.01760000 | 4.42879400 |
| C | 0.10672900 | -2.74383100 | 3.50375700 |
| C | -0.18531900 | -2.13942600 | 2.25068800 |
| C | -1.44031200 | 1.03648200 | 1.42798200 |
| C | -1.31932500 | 2.53058100 | 1.15624300 |
| C | -2.39641300 | 3.38744400 | 1.49624700 |
| C | -2.30302700 | 4.78422100 | 1.32885600 |
| C | -1.11448100 | 5.34225300 | 0.80674300 |
| C | -0.03712100 | 4.50530500 | 0.45792100 |
| C | -0.12352600 | 3.09649200 | 0.62712400 |
| C | -1.98459400 | 0.69596800 | -0.97689500 |
| C | -1.66097000 | -1.95050700 | -3.77914500 |
| C | -2.99399300 | -2.03828500 | -4.22354700 |
| C | -3.99987000 | -1.26416200 | -3.60290900 |
| C | -3.65242900 | -0.40726500 | -2.53878400 |
| C | -2.31260900 | -0.28935900 | -2.09205600 |
| C | -1.29966700 | -1.07056800 | -2.72163500 |
| C | 3.77594400 | -0.34881000 | 0.19404100 |
| C | 4.76569800 | 0.11306300 | -1.75113100 |
| C | 3.39323600 | 0.14928400 | -1.90844000 |
| C | 6.30968500 | -0.35608400 | 0.24415600 |
| H | -2.99757600 | -1.08864100 | 0.52748700 |
| H | -1.61472700 | -1.94764100 | -0.22272400 |
| H | -3.59472400 | -1.81641600 | 2.63849800 |
| H | -3.06423500 | -2.88947600 | 4.84575000 |
| H | -0.67138400 | -3.49017400 | 5.39243600 |
| H | 1.15073700 | -2.99807000 | 3.74273000 |
| H | -2.45567100 | 0.83774700 | 1.83534600 |
| H | -0.70769200 | 0.71809800 | 2.19695900 |
| H | -3.32091200 | 2.94775800 | 1.90600300 |
| H | -3.15100100 | 5.43080600 | 1.60197000 |
| H | -1.02735600 | 6.43157700 | 0.66890500 |
| H | 0.88977600 | 4.93313000 | 0.04668300 |
| H | -2.93563900 | 1.13419700 | -0.60391100 |
| H | -1.36179800 | 1.53005700 | -1.35853300 |
| H | -0.87670900 | -2.56256200 | -4.24989400 |
| H | -3.24931700 | -2.72039100 | -5.04982000 |
| H | -5.04608100 | -1.33334500 | -3.93783400 |
| H | -4.43383500 | 0.19352400 | -2.04430800 |
| H | 3.64325900 | -0.60411900 | 1.24803700 |
| H | 5.58457600 | 0.28444700 | -2.45438500 |
| H | 2.80191800 | 0.35859600 | -2.80242000 |
| H | 6.15287500 | -0.58166900 | 1.31510500 |
| H | 6.87797800 | -1.18738600 | -0.21927900 |
| H | 6.89385800 | 0.58166400 | 0.15620000 |

**[$Fe^{III}$(N($CH_2$-*o*-$C_6H_4S)_3$(1-MeIm)]. S = 3/2. E =** -3575.96654445

| | | | |
|---|---|---|---|
| Fe | 0.790761 | 0.386391 | -0.008428 |
| S | 1.344144 | -0.764301 | -2.033642 |
| S | 1.394281 | -0.724596 | 2.018693 |
| S | 0.249907 | 2.640246 | -0.034161 |
| N | -1.155434 | -0.253080 | 0.017316 |
| N | 2.744311 | 0.926838 | -0.032034 |
| N | 4.955002 | 0.624682 | -0.048065 |
| C | -1.744600 | -0.224747 | -1.417729 |
| C | -1.393687 | -1.405863 | -2.310739 |
| C | -2.443307 | -2.190595 | -2.853923 |
| C | -2.183750 | -3.249689 | -3.746110 |
| C | -0.846213 | -3.540535 | -4.098173 |
| C | 0.209237 | -2.781024 | -3.559357 |
| C | -0.043634 | -1.704120 | -2.663838 |
| C | -1.257268 | -1.702257 | 0.546825 |
| C | -1.188548 | -1.866513 | 2.058010 |
| C | -2.277145 | -2.469813 | 2.738307 |
| C | -2.234654 | -2.708303 | 4.126498 |
| C | -1.085757 | -2.327214 | 4.856044 |
| C | -0.000686 | -1.715576 | 4.200059 |
| C | -0.030490 | -1.481371 | 2.796810 |
| C | -2.035428 | 0.645512 | 0.915623 |
| C | -1.970652 | 4.183688 | -0.628752 |
| C | -3.346443 | 4.474426 | -0.696369 |
| C | -4.297125 | 3.517978 | -0.273545 |
| C | -3.850518 | 2.274877 | 0.217625 |
| C | -2.468403 | 1.970840 | 0.307996 |
| C | -1.512199 | 2.936108 | -0.122670 |
| C | 3.736958 | 0.012724 | -0.046941 |
| C | 4.732381 | 2.009208 | -0.033194 |
| C | 3.363433 | 2.183599 | -0.023162 |
| C | 6.265892 | -0.051914 | -0.064265 |
| H | -2.848013 | -0.178578 | -1.306545 |
| H | -1.412298 | 0.730291 | -1.870312 |
| H | -3.484328 | -1.954213 | -2.576974 |
| H | -3.014252 | -3.841340 | -4.160240 |
| H | -0.623166 | -4.366415 | -4.792227 |
| H | 1.251357 | -3.011518 | -3.827468 |
| H | -2.226833 | -2.108137 | 0.189067 |
| H | -0.448435 | -2.269861 | 0.047025 |
| H | -3.168728 | -2.767033 | 2.161125 |
| H | -3.088484 | -3.183299 | 4.633207 |
| H | -1.036598 | -2.501623 | 5.942637 |
| H | 0.890448 | -1.408962 | 4.768172 |
| H | -2.941536 | 0.057349 | 1.172861 |
| H | -1.462573 | 0.803363 | 1.850982 |
| H | -1.229239 | 4.922899 | -0.967546 |
| H | -3.677835 | 5.449457 | -1.087285 |
| H | -5.374361 | 3.736643 | -0.328758 |
| H | -4.585555 | 1.523347 | 0.551088 |
| H | 3.595001 | -1.070222 | -0.057157 |
| H | 5.556425 | 2.726930 | -0.031675 |
| H | 2.778635 | 3.106288 | -0.010877 |
| H | 6.103672 | -1.145264 | -0.071834 |
| H | 6.833879 | 0.235655 | -0.971177 |
| H | 6.848206 | 0.220850 | 0.838093 |

**[$Fe^{III}$(N($CH_2$-*o*-$C_6H_4S)_3$(1-MeIm)]. S = 5/2. E =** -3575.97064928

| | | | |
|---|---|---|---|
| Fe | -0.709941 | 0.142037 | 0.109827 |
| S | -1.203272 | -2.156240 | -0.180266 |
| S | -0.745018 | 1.744948 | -1.624033 |
| S | -0.509434 | 0.904851 | 2.323443 |
| N | 1.605629 | -0.234329 | -0.139932 |
| N | -2.908674 | 0.555818 | 0.227107 |
| N | -5.133566 | 0.346147 | 0.192134 |
| C | 1.920384 | -1.727172 | -0.175416 |
| C | 1.336251 | -2.478978 | -1.366422 |
| C | 2.192099 | -3.001639 | -2.368243 |
| C | 1.686079 | -3.750145 | -3.450730 |
| C | 0.296321 | -3.983521 | -3.541104 |
| C | -0.571756 | -3.471686 | -2.556110 |
| C | -0.067925 | -2.720052 | -1.461658 |
| C | 2.125232 | 0.382883 | -1.436361 |
| C | 2.081511 | 1.903951 | -1.506805 |
| C | 3.291041 | 2.643256 | -1.540411 |
| C | 3.292522 | 4.045974 | -1.679331 |
| C | 2.061735 | 4.730585 | -1.786466 |
| C | 0.849178 | 4.014563 | -1.751108 |
| C | 0.840109 | 2.599928 | -1.612223 |
| C | 2.363168 | 0.391777 | 1.027327 |
| C | 0.646661 | -0.602641 | 4.346571 |
| C | 1.680082 | -1.313766 | 4.987325 |
| C | 2.922837 | -1.493273 | 4.340911 |
| C | 3.114195 | -0.953445 | 3.053067 |
| C | 2.088410 | -0.231822 | 2.391015 |
| C | 0.834384 | -0.052014 | 3.050131 |
| C | -3.931934 | -0.272344 | -0.035251 |
| C | -4.856270 | 1.649722 | 0.624653 |
| C | -3.478835 | 1.764640 | 0.638495 |
| C | -6.469281 | -0.249884 | 0.007136 |
| H | 3.030217 | -1.843327 | -0.176289 |
| H | 1.533339 | -2.157121 | 0.768826 |
| H | 3.276905 | -2.820830 | -2.289417 |
| H | 2.370691 | -4.147936 | -4.215370 |
| H | -0.114997 | -4.565224 | -4.381128 |
| H | -1.654914 | -3.652914 | -2.621260 |
| H | 3.179400 | 0.044622 | -1.574081 |
| H | 1.519921 | -0.050160 | -2.256734 |
| H | 4.248182 | 2.100984 | -1.463953 |
| H | 4.244392 | 4.598148 | -1.705644 |
| H | 2.044315 | 5.826474 | -1.896139 |
| H | -0.111612 | 4.544282 | -1.833534 |
| H | 3.454247 | 0.318876 | 0.806274 |
| H | 2.092142 | 1.465799 | 1.039493 |
| H | -0.323751 | -0.461836 | 4.845119 |
| H | 1.512340 | -1.729949 | 5.993232 |
| H | 3.735875 | -2.047261 | 4.834568 |
| H | 4.083786 | -1.085044 | 2.544748 |
| H | -3.827230 | -1.304506 | -0.382805 |
| H | -5.652439 | 2.356867 | 0.871929 |
| H | -2.858035 | 2.623776 | 0.905349 |
| H | -6.349880 | -1.298896 | -0.321266 |
| H | -7.033090 | -0.234301 | 0.961207 |
| H | -7.038668 | 0.306752 | -0.764073 |

**[$Fe^{III}$(L)Cl]. S = 1/2. E =** -2933.90713950

| | | | |
|---|---|---|---|
| Fe | 0.002456 | 0.169864 | 0.088023 |
| N | 1.249677 | -1.249095 | -0.178181 |
| C | 2.569850 | -1.154670 | -0.261286 |
| C | 3.316994 | 0.059570 | -0.383091 |
| C | 4.809244 | -0.068652 | -0.382160 |
| O | 5.216810 | -1.384005 | -0.556689 |
| N | -1.316350 | -1.141329 | -0.337242 |
| C | -2.635413 | -0.963880 | -0.368558 |
| C | -3.332748 | 0.260889 | -0.145183 |
| C | -4.827379 | 0.193268 | -0.254386 |
| O | -5.294652 | -1.106370 | -0.139388 |
| C | 2.680295 | 1.354137 | -0.510061 |
| C | 3.489020 | 2.576509 | -0.964772 |
| C | -2.649190 | 1.522649 | 0.075154 |
| C | -3.438176 | 2.797429 | 0.401238 |
| C | 0.616653 | -2.585303 | -0.018716 |
| C | -0.769929 | -2.486703 | -0.689480 |
| O | 5.627325 | 0.844734 | -0.229419 |
| O | -5.605476 | 1.134074 | -0.442659 |
| H | 3.152199 | -2.091263 | -0.256540 |
| H | -3.257593 | -1.843256 | -0.607058 |
| H | 4.484280 | 2.291585 | -1.333086 |
| H | 2.938850 | 3.123666 | -1.757110 |
| H | 3.646041 | 3.279443 | -0.117856 |
| H | -3.455920 | 3.488258 | -0.469788 |
| H | -4.484396 | 2.569411 | 0.648178 |
| H | -2.960486 | 3.329860 | 1.247953 |
| C | -6.739580 | -1.266998 | -0.312909 |
| C | -7.050861 | -2.763442 | -0.168310 |
| H | -7.262866 | -0.656734 | 0.452013 |
| H | -7.033428 | -0.872426 | -1.308196 |
| H | -8.141270 | -2.933801 | -0.284741 |
| H | -6.522501 | -3.357497 | -0.942002 |
| H | -6.744472 | -3.138039 | 0.829484 |
| C | 6.660098 | -1.625657 | -0.468664 |
| C | 7.117917 | -1.793992 | 0.992035 |
| H | 6.813043 | -2.555456 | -1.052176 |
| H | 7.193571 | -0.788897 | -0.960140 |
| H | 8.197057 | -2.055111 | 1.021731 |
| H | 6.972677 | -0.851206 | 1.554782 |
| H | 6.550324 | -2.603613 | 1.494888 |
| N | -1.306059 | 1.588673 | 0.027552 |
| N | 1.354372 | 1.502638 | -0.302670 |
| C | -0.609899 | 2.884800 | 0.260182 |
| C | 0.736243 | 2.850480 | -0.489505 |
| H | -0.431172 | 2.999911 | 1.353084 |
| H | 1.386172 | 3.662464 | -0.104611 |
| H | -1.449654 | -3.298029 | -0.350547 |
| H | 1.235915 | -3.390699 | -0.469313 |
| H | -0.662525 | -2.561683 | -1.795731 |
| H | 0.497391 | -2.787816 | 1.068265 |
| H | 0.567244 | 3.040232 | -1.575591 |
| H | -1.195682 | 3.757624 | -0.090570 |
| Cl | 0.048293 | 0.015903 | 2.360350 |

**[$Fe^{III}$(L)Cl]. S = 3/2. E =** -2933.92164369

| | | | |
|---|---|---|---|
| Fe | 0.006235 | 0.166883 | 0.272640 |
| N | 1.258325 | -1.252585 | -0.026173 |
| C | 2.569277 | -1.157460 | -0.213408 |
| C | 3.308576 | 0.042619 | -0.431702 |
| C | 4.797011 | -0.090228 | -0.507962 |
| O | 5.193964 | -1.415791 | -0.624176 |
| N | -1.319352 | -1.166567 | -0.206424 |
| C | -2.629915 | -0.983034 | -0.321673 |
| C | -3.340149 | 0.247092 | -0.174846 |
| C | -4.826830 | 0.174217 | -0.346604 |
| O | -5.293362 | -1.128124 | -0.250966 |
| C | 2.658100 | 1.334985 | -0.576237 |
| C | 3.442138 | 2.539701 | -1.112514 |
| C | -2.660902 | 1.512680 | 0.038552 |
| C | -3.458839 | 2.799358 | 0.285300 |
| C | 0.625011 | -2.581107 | 0.188318 |
| C | -0.753874 | -2.516053 | -0.505785 |
| O | 5.623861 | 0.827000 | -0.458830 |
| O | -5.602576 | 1.110590 | -0.566384 |
| H | 3.148931 | -2.096020 | -0.214098 |
| H | -3.241882 | -1.869279 | -0.563201 |
| H | 4.422598 | 2.242375 | -1.509291 |
| H | 2.855445 | 3.054974 | -1.899896 |
| H | 3.629951 | 3.273932 | -0.298983 |
| H | -3.423191 | 3.463235 | -0.605986 |
| H | -4.518115 | 2.581696 | 0.480058 |
| H | -3.022083 | 3.352922 | 1.140315 |
| C | -6.729806 | -1.294583 | -0.479832 |
| C | -7.040687 | -2.791905 | -0.343259 |
| H | -7.284745 | -0.684427 | 0.262452 |
| H | -6.986193 | -0.904038 | -1.486958 |
| H | -8.125016 | -2.966917 | -0.501808 |
| H | -6.480207 | -3.386056 | -1.093959 |
| H | -6.772344 | -3.162436 | 0.666936 |
| C | 6.639855 | -1.657223 | -0.601682 |
| C | 7.177873 | -1.739282 | 0.838835 |
| H | 6.759604 | -2.621036 | -1.135714 |
| H | 7.145651 | -0.853809 | -1.171653 |
| H | 8.256318 | -2.004479 | 0.824273 |
| H | 7.066781 | -0.763362 | 1.350636 |
| H | 6.637271 | -2.513577 | 1.420805 |
| N | -1.318901 | 1.578411 | 0.057353 |
| N | 1.351101 | 1.492197 | -0.307571 |
| C | -0.616313 | 2.877656 | 0.252408 |
| C | 0.713356 | 2.824597 | -0.527978 |
| H | -0.414908 | 3.014576 | 1.338320 |
| H | 1.364867 | 3.656455 | -0.190116 |
| H | -1.431009 | -3.321897 | -0.148571 |
| H | 1.251363 | -3.403257 | -0.220190 |
| H | -0.626323 | -2.628629 | -1.606715 |
| H | 0.492612 | -2.738668 | 1.281101 |
| H | 0.513546 | 2.970277 | -1.615510 |
| H | -1.207070 | 3.743346 | -0.105865 |
| Cl | 0.082909 | 0.191962 | 2.608115 |

**[$Fe^{III}$(L)Cl]. S = 5/2. E =** -2933.90330200

| | | | |
|---|---|---|---|
| Fe | 0.015780 | 0.173135 | 0.505141 |
| N | 1.301805 | -1.302383 | -0.061324 |
| C | 2.603564 | -1.190458 | -0.277016 |
| C | 3.331143 | 0.034558 | -0.441845 |
| C | 4.822510 | -0.076009 | -0.491571 |
| O | 5.245444 | -1.389851 | -0.645175 |
| N | -1.327945 | -1.166182 | -0.272527 |
| C | -2.638299 | -0.975383 | -0.392440 |
| C | -3.356516 | 0.250415 | -0.207574 |
| C | -4.844169 | 0.165046 | -0.357524 |
| O | -5.301738 | -1.140117 | -0.249453 |
| C | 2.681537 | 1.328729 | -0.593422 |
| C | 3.453945 | 2.510251 | -1.198723 |
| C | -2.701224 | 1.536124 | -0.005284 |
| C | -3.519640 | 2.815811 | 0.213626 |
| C | 0.638686 | -2.625675 | 0.000178 |
| C | -0.745460 | -2.470571 | -0.690218 |
| O | 5.631690 | 0.852736 | -0.388559 |
| O | -5.628334 | 1.096830 | -0.568040 |
| H | 3.197466 | -2.119069 | -0.358983 |
| H | -3.253075 | -1.847773 | -0.681433 |
| H | 4.446747 | 2.211283 | -1.561352 |
| H | 2.867159 | 2.952383 | -2.030465 |
| H | 3.606948 | 3.306876 | -0.438721 |
| H | -3.614180 | 3.389671 | -0.734206 |
| H | -4.543453 | 2.586657 | 0.543343 |
| H | -3.019042 | 3.463146 | 0.959953 |
| C | -6.738709 | -1.316174 | -0.465481 |
| C | -7.038797 | -2.815341 | -0.324442 |
| H | -7.291228 | -0.708803 | 0.280996 |
| H | -7.007329 | -0.928460 | -1.470599 |
| H | -8.123345 | -2.997884 | -0.472901 |
| H | -6.481025 | -3.406579 | -1.079455 |
| H | -6.758574 | -3.183078 | 0.683562 |
| C | 6.694565 | -1.606503 | -0.604431 |
| C | 7.208461 | -1.729283 | 0.842081 |
| H | 6.841600 | -2.548939 | -1.168994 |
| H | 7.196251 | -0.774957 | -1.136532 |
| H | 8.291764 | -1.974401 | 0.838391 |
| H | 7.069936 | -0.773810 | 1.384768 |
| H | 6.671979 | -2.533047 | 1.386768 |
| N | -1.363151 | 1.613993 | 0.007136 |
| N | 1.381296 | 1.502196 | -0.298317 |
| C | -0.648349 | 2.912025 | 0.107623 |
| C | 0.712795 | 2.793150 | -0.631169 |
| H | -0.474176 | 3.160611 | 1.180355 |
| H | 1.338122 | 3.667808 | -0.353920 |
| H | -1.417944 | -3.319218 | -0.431285 |
| H | 1.243489 | -3.418178 | -0.493624 |
| H | -0.603821 | -2.467855 | -1.795171 |
| H | 0.494864 | -2.910431 | 1.067157 |
| H | 0.533095 | 2.842753 | -1.730459 |
| H | -1.215806 | 3.750568 | -0.345471 |
| Cl | 0.033913 | 0.173223 | 2.771496 |

**[$Fe^{III}$(oetpp)(HIm)]$^{+}$. S = 3/2. E** = -4010.28963518

| | | | |
|---|---|---|---|
| C | -0.021632 | 0.609149 | 4.381570 |
| C | -0.063710 | 1.063628 | 3.075047 |
| N | 0.003513 | -0.024450 | 2.199406 |
| C | 0.085318 | -1.123708 | 2.961953 |
| N | 0.074166 | -0.783342 | 4.288934 |
| Fe | -0.001196 | 0.004792 | 0.033982 |
| N | -0.027327 | 2.019701 | -0.279018 |
| C | -1.154820 | 2.711616 | -0.740788 |
| C | -0.741400 | 3.810929 | -1.630891 |
| C | 0.653023 | 3.831949 | -1.619853 |
| C | 1.085565 | 2.743634 | -0.725243 |
| C | -2.451960 | 2.395006 | -0.246021 |
| C | -3.542375 | 3.433873 | -0.287409 |
| C | -3.392199 | 4.669606 | 0.398425 |
| C | -4.416237 | 5.639236 | 0.362577 |
| C | -5.598101 | 5.395435 | -0.372687 |
| C | -5.754834 | 4.174041 | -1.066252 |
| C | -4.738668 | 3.196783 | -1.017796 |
| C | 2.386523 | 2.458514 | -0.220733 |
| C | 2.700038 | 1.163039 | 0.287987 |
| N | 1.980817 | 0.029559 | -0.143793 |
| C | 2.731846 | -1.090431 | 0.267516 |
| C | 3.846888 | -0.656162 | 1.111475 |
| C | 3.826686 | 0.745731 | 1.125375 |
| C | 4.708734 | 1.640819 | 1.992381 |
| C | 3.947392 | 2.248336 | 3.211123 |
| C | 4.754385 | -1.542035 | 1.961742 |
| C | 4.013780 | -2.181041 | 3.177054 |
| C | 2.450536 | -2.387164 | -0.257646 |
| C | 3.542957 | -3.423631 | -0.307055 |
| C | 4.738646 | -3.178715 | -1.035973 |
| C | 5.757379 | -4.152941 | -1.091223 |
| C | 5.604022 | -5.379628 | -0.406306 |
| C | 4.422723 | -5.631756 | 0.327041 |
| C | 3.396426 | -4.664795 | 0.369779 |
| C | -1.639626 | 4.633961 | -2.550248 |
| C | -2.188557 | 3.808870 | -3.756013 |
| C | 1.540281 | 4.683534 | -2.523713 |
| C | 2.117030 | 3.882064 | -3.732347 |
| N | -1.983064 | -0.021694 | -0.140972 |
| C | -2.702588 | -1.156639 | 0.286919 |
| C | -3.827843 | -0.741382 | 1.127070 |
| C | -3.848240 | 0.660484 | 1.118527 |
| C | -2.733232 | 1.096886 | 0.275423 |
| C | -4.706780 | -1.639556 | 1.994047 |
| C | -3.941214 | -2.244607 | 3.211381 |
| C | -2.390358 | -2.450627 | -0.226196 |
| C | -3.452671 | -3.518718 | -0.254044 |
| C | -3.261811 | -4.747933 | 0.433470 |
| C | -4.257844 | -5.746661 | 0.409235 |
| C | -5.452634 | -5.538659 | -0.316027 |
| C | -5.649962 | -4.324079 | -1.011236 |
| C | -4.661673 | -3.318189 | -0.974440 |
| C | -4.756416 | 1.543346 | 1.971098 |
| C | -4.015685 | 2.186174 | 3.184338 |
| N | 0.024772 | -2.016014 | -0.278824 |
| C | -1.089125 | -2.735754 | -0.731076 |
| C | -0.657778 | -3.816881 | -1.633788 |
| C | 0.737171 | -3.795896 | -1.646609 |
| C | 1.152012 | -2.703748 | -0.749605 |
| C | -1.545931 | -4.662143 | -2.542736 |
| C | -2.120784 | -3.853298 | -3.747395 |
| C | 1.633762 | -4.612522 | -2.573318 |
| C | 2.178564 | -3.779826 | -3.775757 |
| C | 3.446626 | 3.528887 | -0.242817 |
| C | 3.253632 | 4.752366 | 0.454201 |
| C | 4.247210 | 5.753664 | 0.435012 |
| C | 5.440881 | 5.554109 | -0.294463 |
| C | 5.640060 | 4.345134 | -0.998871 |
| C | 4.654542 | 3.336359 | -0.967107 |
| H | 1.053083 | -5.471463 | -2.967009 |
| H | 2.486063 | -5.050364 | -2.021489 |
| H | 1.349558 | -3.357627 | -4.380563 |
| H | 2.798796 | -4.418003 | -4.438485 |

| | | | |
|---|---|---|---|
| H | 2.808432 | -2.935858 | -3.427256 |
| H | -2.382571 | -5.112398 | -1.977343 |
| H | -0.951145 | -5.512561 | -2.934013 |
| H | -2.763502 | -3.018443 | -3.400332 |
| H | -2.735202 | -4.510604 | -4.396701 |
| H | -1.307102 | -3.420767 | -4.365486 |
| H | 4.299366 | -6.584817 | 0.863848 |
| H | 6.401551 | -6.137039 | -0.445263 |
| H | 6.672552 | -3.954638 | -1.669755 |
| H | 4.860242 | -2.220925 | -1.564691 |
| H | 2.470693 | -4.866682 | 0.930495 |
| H | 2.318720 | 4.912358 | 1.013028 |
| H | 6.212643 | 6.338465 | -0.315162 |
| H | 4.088530 | 6.692352 | 0.987590 |
| H | 4.811604 | 2.392563 | -1.511450 |
| H | 6.565128 | 4.187667 | -1.574235 |
| H | -4.290632 | 6.587807 | 0.906718 |
| H | -2.466600 | 4.864200 | 0.961750 |
| H | -6.393716 | 6.155136 | -0.406142 |
| H | -6.670612 | 3.982186 | -1.646016 |
| H | -4.862801 | 2.243027 | -1.553241 |
| H | -5.598758 | 0.930961 | 2.351524 |
| H | -5.215017 | 2.345474 | 1.363623 |
| H | -3.553833 | 1.411605 | 3.831256 |
| H | -4.724824 | 2.775660 | 3.801397 |
| H | -3.210811 | 2.869279 | 2.844315 |
| H | -5.144424 | -2.462851 | 1.399715 |
| H | -5.564402 | -1.047444 | 2.372053 |
| H | -3.114596 | -2.903428 | 2.874210 |
| H | -4.626970 | -2.852477 | 3.837045 |
| H | -3.507827 | -1.445999 | 3.849328 |
| H | 5.599501 | -0.932995 | 2.341039 |
| H | 5.208392 | -2.346853 | 1.354530 |
| H | 3.562594 | -1.401719 | 3.826552 |
| H | 4.719733 | -2.776856 | 3.791719 |
| H | 3.203622 | -2.858943 | 2.837405 |
| H | 5.565692 | 1.045868 | 2.367821 |
| H | 5.147960 | 2.462572 | 1.396992 |
| H | 3.122249 | 2.909943 | 2.877497 |
| H | 4.637840 | 2.852198 | 3.835457 |
| H | 3.508991 | 1.453167 | 3.849396 |
| H | -2.325556 | -4.915215 | 0.988061 |
| H | -6.226477 | -6.320827 | -0.340775 |
| H | -4.100287 | -6.690012 | 0.954188 |
| H | -6.575799 | -4.159992 | -1.583487 |
| H | -4.817424 | -2.370064 | -1.511522 |
| H | 2.375734 | 5.132366 | -1.955482 |
| H | 0.944130 | 5.534874 | -2.910790 |
| H | -2.489753 | 5.069720 | -1.993515 |
| H | -1.058863 | 5.494358 | -2.940538 |
| H | -0.136340 | 2.086269 | 2.696473 |
| H | 0.153006 | -2.148207 | 2.586460 |
| H | 0.125640 | -1.437436 | 5.074268 |
| H | -0.049004 | 1.127973 | 5.342727 |
| H | -1.361604 | 3.389182 | -4.365398 |
| H | -2.809757 | 4.451603 | -4.413412 |
| H | -2.818619 | 2.963714 | -3.410747 |
| H | 2.760794 | 3.046321 | -3.389379 |
| H | 2.730935 | 4.543755 | -4.377675 |
| H | 1.304326 | 3.451740 | -4.353293 |

**[$Fe^{III}$(oetpp)(HIm)]$^{+}$. S = 5/2. E** = -4010.27195819

| | | | |
|---|---|---|---|
| C | 0.035792 | 0.604288 | 4.459761 |
| C | 0.091176 | 1.060723 | 3.155949 |
| N | -0.006753 | -0.030086 | 2.282980 |
| C | -0.119502 | -1.130594 | 3.045348 |
| N | -0.098250 | -0.784829 | 4.367219 |
| Fe | 0.001123 | 0.005602 | 0.161128 |
| N | 0.054729 | 2.017267 | -0.299223 |
| C | -1.055862 | 2.771393 | -0.709026 |
| C | -0.607414 | 3.871747 | -1.571095 |
| C | 0.794901 | 3.830539 | -1.587663 |
| C | 1.198245 | 2.707884 | -0.731836 |
| C | -2.375315 | 2.493686 | -0.225807 |
| C | -3.403898 | 3.597343 | -0.238268 |
| C | -3.197446 | 4.787433 | 0.510795 |
| C | -4.158488 | 5.820373 | 0.499505 |
| C | -5.334919 | 5.685578 | -0.271611 |
| C | -5.548636 | 4.509899 | -1.026358 |
| C | -4.594687 | 3.470754 | -1.004535 |
| C | 2.507148 | 2.363981 | -0.264031 |
| C | 2.817320 | 1.057912 | 0.229370 |
| N | 2.042031 | -0.049187 | -0.135384 |
| C | 2.754202 | -1.191417 | 0.250005 |
| C | 3.927356 | -0.791713 | 1.045563 |
| C | 3.969171 | 0.608037 | 1.029542 |
| C | 4.917956 | 1.475625 | 1.852939 |
| C | 4.238358 | 2.082462 | 3.119749 |
| C | 4.816722 | -1.696152 | 1.895049 |
| C | 4.085161 | -2.251406 | 3.156784 |
| C | 2.379122 | -2.485351 | -0.229730 |
| C | 3.411259 | -3.585542 | -0.250299 |
| C | 4.604591 | -3.446268 | -1.010497 |
| C | 5.562922 | -4.481178 | -1.039946 |
| C | 5.351234 | -5.666056 | -0.299180 |
| C | 4.172226 | -5.814132 | 0.465517 |
| C | 3.207234 | -4.785035 | 0.484419 |
| C | -1.472501 | 4.765249 | -2.455986 |
| C | -2.038761 | 4.010261 | -3.699584 |
| C | 1.689278 | 4.670389 | -2.495668 |
| C | 2.203730 | 3.874039 | -3.736194 |
| N | -2.040133 | 0.057015 | -0.140610 |
| C | -2.815950 | -1.050645 | 0.219297 |
| C | -3.968148 | -0.603002 | 1.019956 |
| C | -3.927233 | 0.796762 | 1.040483 |
| C | -2.752264 | 1.198576 | 0.248441 |
| C | -4.915947 | -1.473693 | 1.841290 |
| C | -4.233145 | -2.087898 | 3.102763 |
| C | -2.505207 | -2.355991 | -0.276815 |
| C | -3.597816 | -3.394747 | -0.330757 |
| C | -3.479139 | -4.614325 | 0.389564 |
| C | -4.504728 | -5.582003 | 0.341056 |
| C | -5.660059 | -5.351833 | -0.439343 |
| C | -5.787405 | -4.146250 | -1.165777 |
| C | -4.768142 | -3.172572 | -1.106809 |
| C | -4.821263 | 1.697864 | 1.888652 |
| C | -4.100694 | 2.241680 | 3.161650 |
| N | -0.052050 | -2.011938 | -0.298698 |
| C | -1.194884 | -2.699013 | -0.741093 |
| C | -0.789265 | -3.814345 | -1.604759 |
| C | 0.613178 | -3.855219 | -1.585929 |
| C | 1.059995 | -2.762273 | -0.714230 |
| C | -1.681720 | -4.646370 | -2.521930 |
| C | -2.193821 | -3.839479 | -3.756599 |
| C | 1.480106 | -4.740296 | -2.477577 |
| C | 2.047085 | -3.974114 | -3.713993 |
| C | 3.597941 | 3.405143 | -0.308725 |
| C | 3.476565 | 4.617310 | 0.423431 |
| C | 4.500253 | 5.587405 | 0.383171 |
| C | 5.655362 | 5.367204 | -0.400468 |
| C | 5.785029 | 4.168812 | -1.138304 |
| C | 4.768038 | 3.192305 | -1.087543 |
| H | 0.873095 | -5.596632 | -2.835207 |
| H | 2.319622 | -5.179892 | -1.909698 |
| H | 1.230547 | -3.552158 | -4.335441 |
| H | 2.646325 | -4.658157 | -4.349629 |

| | | | |
|---|---|---|---|
| H | 2.702069 | -3.136305 | -3.399230 |
| H | -2.549502 | -5.055147 | -1.973410 |
| H | -1.108186 | -5.524271 | -2.882190 |
| H | -2.816123 | -2.977283 | -3.441491 |
| H | -2.812549 | -4.488023 | -4.410527 |
| H | -1.349355 | -3.446842 | -4.359792 |
| H | 4.003151 | -6.733996 | 1.046157 |
| H | 6.101147 | -6.471357 | -0.319311 |
| H | 6.476329 | -4.363243 | -1.642830 |
| H | 4.771597 | -2.522173 | -1.584822 |
| H | 2.283094 | -4.906535 | 1.070609 |
| H | 2.571955 | 4.794575 | 1.025487 |
| H | 6.450896 | 6.126893 | -0.436738 |
| H | 4.395586 | 6.517689 | 0.962335 |
| H | 4.871409 | 2.256932 | -1.658655 |
| H | 6.679934 | 3.994631 | -1.755151 |
| H | -3.988258 | 6.732769 | 1.091432 |
| H | -2.275782 | 4.897843 | 1.102872 |
| H | -6.081528 | 6.494088 | -0.285742 |
| H | -6.460197 | 4.402437 | -1.633996 |
| H | -4.763161 | 2.553662 | -1.589613 |
| H | -5.716798 | 1.124915 | 2.204062 |
| H | -5.199295 | 2.552733 | 1.299051 |
| H | -3.739960 | 1.412414 | 3.805824 |
| H | -4.794171 | 2.866806 | 3.761281 |
| H | -3.226540 | 2.867915 | 2.889152 |
| H | -5.339203 | -2.291309 | 1.229816 |
| H | -5.778860 | -0.859322 | 2.169081 |
| H | -3.402159 | -2.764822 | 2.814728 |
| H | -4.964264 | -2.679873 | 3.691228 |
| H | -3.821737 | -1.294445 | 3.761825 |
| H | 5.706211 | -1.122170 | 2.224784 |
| H | 5.204045 | -2.545677 | 1.303531 |
| H | 3.713127 | -1.426393 | 3.800271 |
| H | 4.774792 | -2.875658 | 3.761789 |
| H | 3.219252 | -2.882994 | 2.869032 |
| H | 5.783464 | 0.860783 | 2.173372 |
| H | 5.337888 | 2.296486 | 1.243638 |
| H | 3.403473 | 2.757273 | 2.839897 |
| H | 4.971279 | 2.673339 | 3.707030 |
| H | 3.829912 | 1.286851 | 3.777472 |
| H | -2.573695 | -4.800247 | 0.987938 |
| H | -6.457268 | -6.109401 | -0.482208 |
| H | -4.401395 | -6.518279 | 0.910775 |
| H | -6.682270 | -3.964327 | -1.780428 |
| H | -4.869760 | -2.231826 | -1.669297 |
| H | 2.555805 | 5.074449 | -1.941814 |
| H | 1.116385 | 5.551249 | -2.849603 |
| H | -2.312136 | 5.200785 | -1.885233 |
| H | -0.864234 | 5.623983 | -2.805575 |
| H | 0.192234 | 2.080087 | 2.775837 |
| H | -0.214801 | -2.152535 | 2.669303 |
| H | -0.167743 | -1.436831 | 5.153398 |
| H | 0.077602 | 1.123048 | 5.420243 |
| H | -1.221914 | 3.593140 | -4.323911 |
| H | -2.636865 | 4.700290 | -4.329794 |
| H | -2.694747 | 3.170276 | -3.392780 |
| H | 2.825202 | 3.008997 | -3.427275 |
| H | 2.824014 | 4.528068 | -4.383144 |
| H | 1.360416 | 3.486889 | -4.344556 |

***cis*-[Fe$^{II}$(phen)$_2$(NCS)$_2$]. S = 0. E =** -3375.57135153

| | | | |
|---|---|---|---|
| Fe | 0.000051 | 0.000374 | 0.455695 |
| N | -1.559950 | 1.239206 | 0.429204 |
| C | -1.783636 | 2.335144 | 1.188939 |
| H | -0.962544 | 2.624054 | 1.858494 |
| C | -3.007216 | 3.058984 | 1.133939 |
| H | -3.128855 | 3.941478 | 1.777880 |
| C | -4.033752 | 2.642272 | 0.278918 |
| H | -4.987194 | 3.190481 | 0.227151 |
| C | -3.835623 | 1.478917 | -0.525341 |
| C | -2.576932 | 0.813266 | -0.401733 |
| C | -2.300622 | -0.381099 | -1.158368 |
| C | -3.278757 | -0.909000 | -2.057960 |
| C | -4.548252 | -0.212115 | -2.179470 |
| H | -5.304204 | -0.619321 | -2.868551 |
| C | -4.815585 | 0.927289 | -1.445184 |
| H | -5.787102 | 1.436014 | -1.543237 |
| N | -1.062028 | -0.956865 | -0.938903 |
| C | -0.774931 | -2.084961 | -1.627553 |
| H | 0.204988 | -2.539921 | -1.429771 |
| C | -1.679930 | -2.678065 | -2.547632 |
| H | -1.381984 | -3.599184 | -3.068520 |
| C | -2.934545 | -2.098186 | -2.768331 |
| H | -3.653364 | -2.549273 | -3.469465 |
| N | 1.062221 | 0.954543 | -0.940981 |
| C | 0.775106 | 2.081040 | -1.632231 |
| H | -0.204854 | 2.536392 | -1.435558 |
| C | 1.680150 | 2.672115 | -2.553572 |
| H | 1.382193 | 3.592035 | -3.076569 |
| C | 2.934822 | 2.091817 | -2.772837 |
| H | 3.653680 | 2.541366 | -3.474919 |
| C | 3.279041 | 0.904265 | -2.059739 |
| C | 2.300856 | 0.378345 | -1.159041 |
| C | 2.577175 | -0.814285 | -0.399676 |
| C | 3.835945 | -1.480099 | -0.521604 |
| C | 4.815957 | -0.930496 | -1.442605 |
| H | 5.787527 | -1.439362 | -1.539397 |
| C | 4.548600 | 0.207196 | -2.179532 |
| H | 5.304586 | 0.612884 | -2.869470 |
| N | 1.560145 | -1.238420 | 0.432119 |
| C | 1.783851 | -2.332558 | 1.194433 |
| H | 0.962695 | -2.619961 | 1.864562 |
| C | 3.007508 | -3.056402 | 1.141243 |
| H | 3.129176 | -3.937391 | 1.787237 |
| C | 4.034095 | -2.641569 | 0.285367 |
| H | 4.987595 | -3.189805 | 0.234966 |
| N | 0.921715 | 1.080352 | 1.784518 |
| C | 1.629978 | 1.827496 | 2.397358 |
| S | 2.582517 | 2.880989 | 3.239877 |
| N | -0.921634 | -1.076641 | 1.786878 |
| C | -1.630612 | -1.821917 | 2.401177 |
| S | -2.583960 | -2.872850 | 3.245963 |

***cis*-[Fe$^{II}$(phen)$_2$(NCS)$_2$]. S = 1. E** = -3375.55434601

| | | | |
|---|---|---|---|
| Fe | 0.000670 | 0.005516 | -0.642200 |
| N | 1.552908 | 1.232236 | -0.592313 |
| C | 1.726358 | 2.284499 | -1.430496 |
| H | 0.884034 | 2.484649 | -2.105890 |
| C | 2.906686 | 3.070980 | -1.439687 |
| H | 2.979626 | 3.909088 | -2.147245 |
| C | 3.955909 | 2.766288 | -0.566764 |
| H | 4.885230 | 3.356589 | -0.563671 |
| C | 3.814559 | 1.660208 | 0.323554 |
| C | 2.587719 | 0.921584 | 0.276021 |
| C | 2.398826 | -0.217481 | 1.152637 |
| C | 3.443436 | -0.615175 | 2.052581 |
| C | 4.674703 | 0.152340 | 2.081473 |
| H | 5.472796 | -0.158162 | 2.773574 |
| C | 4.848356 | 1.243380 | 1.255612 |
| H | 5.787783 | 1.817168 | 1.280238 |
| N | 1.196204 | -0.874744 | 1.061810 |
| C | 0.999576 | -1.950890 | 1.843064 |
| H | 0.027807 | -2.458121 | 1.737282 |
| C | 1.974530 | -2.430638 | 2.761259 |
| H | 1.757054 | -3.319274 | 3.371059 |
| C | 3.196858 | -1.759296 | 2.869226 |
| H | 3.972556 | -2.105467 | 3.570213 |
| N | -1.200120 | 0.860805 | 1.074980 |
| C | -1.006535 | 1.925637 | 1.872210 |
| H | -0.035335 | 2.435776 | 1.775512 |
| C | -1.983766 | 2.390606 | 2.795623 |
| H | -1.768753 | 3.270654 | 3.418609 |
| C | -3.205121 | 1.715771 | 2.891705 |
| H | -3.982593 | 2.050360 | 3.596343 |
| C | -3.448431 | 0.583220 | 2.058094 |
| C | -2.401758 | 0.200379 | 1.154048 |
| C | -2.587724 | -0.926077 | 0.260355 |
| C | -3.813568 | -1.667169 | 0.295683 |
| C | -4.849208 | -1.265784 | 1.232434 |
| H | -5.787674 | -1.841469 | 1.247458 |
| C | -4.678480 | -0.186567 | 2.074246 |
| H | -5.477977 | 0.112564 | 2.769728 |
| N | -1.551667 | -1.222578 | -0.611410 |
| C | -1.722894 | -2.262493 | -1.465314 |
| H | -0.879579 | -2.451535 | -2.142687 |
| C | -2.902068 | -3.050452 | -1.487352 |
| H | -2.973123 | -3.878058 | -2.207353 |
| C | -3.952466 | -2.760203 | -0.611004 |
| H | -4.880948 | -3.351808 | -0.617542 |
| N | -1.071533 | 1.268891 | -1.796584 |
| C | -2.011809 | 1.909325 | -2.188643 |
| S | -3.287497 | 2.806039 | -2.713743 |
| N | 1.074636 | -1.241808 | -1.814856 |
| C | 2.016738 | -1.875464 | -2.213573 |
| S | 3.294755 | -2.763236 | -2.748244 |

***cis*-[Fe$^{II}$(phen)$_2$(NCS)$_2$]. S = 2. E** = -3375.57063246

| | | | |
|---|---|---|---|
| Fe | 0.00001300 | 0.00006900 | 0.78883400 |
| N | -1.81040700 | 1.23286700 | 0.44457000 |
| C | -2.13632700 | 2.31327200 | 1.18752800 |
| H | -1.37444600 | 2.63505200 | 1.91268900 |
| C | -3.37384900 | 2.99778900 | 1.04914600 |
| H | -3.57986200 | 3.87176100 | 1.68332900 |
| C | -4.30487600 | 2.54827800 | 0.10661000 |
| H | -5.27134200 | 3.05974300 | -0.02512900 |
| C | -3.99885600 | 1.40183000 | -0.68749700 |
| C | -2.72872700 | 0.76978700 | -0.47165000 |
| C | -2.38004100 | -0.42611400 | -1.22319400 |
| C | -3.30872500 | -0.96088300 | -2.17982300 |
| C | -4.57494400 | -0.28350700 | -2.39325100 |
| H | -5.27526800 | -0.70162800 | -3.13309700 |
| C | -4.90582700 | 0.84755600 | -1.67699300 |
| H | -5.87448100 | 1.34645700 | -1.83664500 |
| N | -1.16524500 | -1.01027200 | -0.95316300 |
| C | -0.84858600 | -2.14320300 | -1.60641800 |
| H | 0.12424500 | -2.59108200 | -1.35254700 |
| C | -1.69947400 | -2.75187200 | -2.56914900 |
| H | -1.38240300 | -3.67985600 | -3.06619600 |
| C | -2.93062500 | -2.15621200 | -2.86141100 |
| H | -3.61483100 | -2.60153400 | -3.60069600 |
| N | 1.16556500 | 1.00976100 | -0.95336400 |
| C | 0.84904700 | 2.14244500 | -1.60711700 |
| H | -0.12380800 | 2.59045800 | -1.35357900 |
| C | 1.70011000 | 2.75070700 | -2.56995000 |
| H | 1.38314700 | 3.67850600 | -3.06741200 |
| C | 2.93129100 | 2.15489500 | -2.86177900 |
| H | 3.61562900 | 2.59990700 | -3.60112900 |
| C | 3.30924800 | 0.95983000 | -2.17965200 |
| C | 2.38039400 | 0.42546600 | -1.22296100 |
| C | 2.72893200 | -0.77012900 | -0.47086800 |
| C | 3.99908700 | -1.40228400 | -0.68623500 |
| C | 4.90622900 | -0.84843700 | -1.67581300 |
| H | 5.87490100 | -1.34742100 | -1.83509900 |
| C | 4.57548700 | 0.28233700 | -2.39259300 |
| H | 5.27594200 | 0.70013900 | -3.13249500 |
| N | 1.81044800 | -1.23281100 | 0.44539400 |
| C | 2.13623300 | -2.31290700 | 1.18886500 |
| H | 1.37422000 | -2.63435600 | 1.91403500 |
| C | 3.37376800 | -2.99749600 | 1.05098200 |
| H | 3.57966600 | -3.87120000 | 1.68557300 |
| C | 4.30495900 | -2.54839900 | 0.10840700 |
| H | 5.27144000 | -3.05993300 | -0.02295500 |
| N | 0.80622100 | 1.39001300 | 2.00166900 |
| C | 1.39079700 | 2.27925600 | 2.56402100 |
| S | 2.16946900 | 3.50729100 | 3.32742900 |
| N | -0.80642600 | -1.38933100 | 2.00212600 |
| C | -1.39154000 | -2.27791500 | 2.56496300 |
| S | -2.17092600 | -3.50497500 | 3.32919700 |

***trans*-[Fe$^{II}$(phen)$_2$(NCS)$_2$]. S = 0. E = -3375.55451274**

| | | | |
|---|---|---|---|
| C | 2.677098 | -0.982758 | 3.328771 |
| C | 3.936587 | -0.794897 | 2.753155 |
| C | 4.011790 | -0.383057 | 1.389677 |
| C | 2.775755 | -0.157018 | 0.703234 |
| C | 1.506988 | -0.705472 | 2.572849 |
| C | 5.254256 | -0.191264 | 0.662906 |
| C | 2.775760 | 0.157158 | -0.703259 |
| C | 4.011800 | 0.383229 | -1.389684 |
| C | 5.254261 | 0.191414 | -0.662911 |
| C | 3.936605 | 0.795145 | -2.753139 |
| H | 4.858877 | 0.981944 | -3.324360 |
| C | 2.677118 | 0.983081 | -3.328735 |
| C | 1.507003 | 0.705763 | -2.572833 |
| H | 6.203109 | -0.352033 | 1.197534 |
| H | 2.570518 | -1.341174 | 4.362359 |
| H | 4.858857 | -0.981666 | 3.324390 |
| H | 0.516626 | -0.880983 | 3.008580 |
| H | 6.203118 | 0.352194 | -1.197529 |
| H | 2.570544 | 1.341584 | -4.362294 |
| H | 0.516649 | 0.881351 | -3.008542 |
| N | 1.535829 | -0.245224 | 1.302629 |
| N | 1.535836 | 0.245396 | -1.302654 |
| C | -2.775763 | 0.157160 | 0.703191 |
| C | -1.507011 | 0.706069 | 2.572686 |
| C | -2.775761 | -0.157325 | -0.703233 |
| C | -4.011805 | 0.383326 | 1.389582 |
| C | -2.677129 | 0.983500 | 3.328542 |
| H | -0.516653 | 0.881695 | 3.008382 |
| C | -4.011797 | -0.383580 | -1.389605 |
| C | -5.254267 | 0.191341 | 0.662856 |
| C | -3.936613 | 0.795480 | 2.752965 |
| H | -2.570559 | 1.342151 | 4.362050 |
| C | -3.936596 | -0.795790 | -2.752970 |
| C | -5.254263 | -0.191639 | -0.662873 |
| C | -1.506996 | -0.706295 | -2.572697 |
| H | -6.203124 | 0.352206 | 1.197448 |
| H | -4.858887 | 0.982357 | 3.324157 |
| C | -2.677107 | -0.983808 | -3.328534 |
| H | -4.858865 | -0.982740 | -3.324146 |
| H | -6.203116 | -0.352563 | -1.197454 |
| H | -0.516639 | -0.881938 | -3.008381 |
| H | -2.570527 | -1.342525 | -4.362018 |
| N | -1.535840 | 0.245528 | 1.302571 |
| N | -1.535835 | -0.245658 | -1.302615 |
| Fe | -0.000004 | 0.000016 | -0.000025 |
| N | -0.000090 | 1.948350 | -0.000213 |
| C | -0.000025 | 3.145615 | -0.000172 |
| S | -0.000426 | 4.798163 | -0.000474 |
| N | 0.000109 | -1.948319 | 0.000193 |
| C | 0.000304 | -3.145585 | 0.000360 |
| S | 0.000361 | -4.798135 | 0.000495 |

***trans*-[Fe$^{II}$(phen)$_2$(NCS)$_2$]. S = 1. E = -3375.55115036**

| | | | |
|---|---|---|---|
| C | -3.216789 | -0.807351 | -3.405383 |
| C | -4.333360 | -1.008615 | -2.592134 |
| C | -4.240887 | -0.727302 | -1.195997 |
| C | -2.977541 | -0.255024 | -0.697838 |
| C | -2.020060 | -0.311151 | -2.815866 |
| C | -5.357717 | -0.881620 | -0.283505 |
| C | -2.831501 | 0.026899 | 0.724049 |
| C | -3.971676 | -0.081918 | 1.590666 |
| C | -5.231530 | -0.558965 | 1.049746 |
| C | -3.812425 | 0.296288 | 2.956885 |
| H | -4.668110 | 0.223835 | 3.645763 |
| C | -2.572826 | 0.774477 | 3.390535 |
| C | -1.491079 | 0.819204 | 2.476912 |
| H | -6.315692 | -1.249022 | -0.683356 |
| H | -3.247765 | -1.017768 | -4.484094 |
| H | -5.282205 | -1.379981 | -3.010203 |
| H | -1.129582 | -0.144095 | -3.441683 |
| H | -6.088305 | -0.657357 | 1.734167 |
| H | -2.417585 | 1.106806 | 4.426700 |
| H | -0.506429 | 1.196237 | 2.782927 |
| N | -1.892436 | -0.031770 | -1.508686 |
| N | -1.592476 | 0.422817 | 1.187646 |
| C | 2.831563 | 0.020852 | -0.723837 |
| C | 1.491827 | 0.801566 | -2.482397 |
| C | 2.976755 | -0.252588 | 0.699771 |
| C | 3.972156 | -0.093430 | -1.589194 |
| C | 2.573929 | 0.750811 | -3.395281 |
| H | 0.507302 | 1.176539 | -2.791285 |
| C | 4.239799 | -0.721835 | 1.201518 |
| C | 5.231789 | -0.566868 | -1.044608 |
| C | 3.813433 | 0.275767 | -2.957936 |
| H | 2.419054 | 1.076134 | -4.433719 |
| C | 4.331234 | -0.995268 | 2.599289 |
| C | 5.357249 | -0.881344 | 0.290672 |
| C | 2.017544 | -0.297198 | 2.817321 |
| H | 6.088976 | -0.669326 | -1.727915 |
| H | 4.669432 | 0.198952 | -3.645949 |
| C | 3.213940 | -0.789811 | 3.410511 |
| H | 5.279835 | -1.364043 | 3.020193 |
| H | 6.315061 | -1.246108 | 0.693316 |
| H | 1.126456 | -0.126943 | 3.441401 |
| H | 3.244105 | -0.994276 | 4.490388 |
| N | 1.592767 | 0.413889 | -1.190468 |
| N | 1.890989 | -0.024977 | 1.508519 |
| Fe | 0.000137 | 0.381882 | -0.001267 |
| N | 0.001654 | 2.302654 | -0.006630 |
| C | 0.002715 | 3.503711 | -0.010444 |
| S | 0.004173 | 5.145991 | -0.015317 |
| N | -0.001366 | -1.568318 | 0.004112 |
| C | -0.001571 | -2.767628 | 0.007022 |
| S | -0.001790 | -4.414024 | 0.011066 |

***trans*-[Fe$^{II}$(phen)$_2$(NCS)$_2$]. S = 2. E = -3375.56435742**

| | | | |
|---|---|---|---|
| C | -3.058655 | -0.737699 | -3.429662 |
| C | -4.295953 | -0.603827 | -2.798361 |
| C | -4.341704 | -0.298904 | -1.404064 |
| C | -3.088743 | -0.137260 | -0.718209 |
| C | -1.875315 | -0.542250 | -2.664729 |
| C | -5.585814 | -0.147700 | -0.672718 |
| C | -3.088716 | 0.137325 | 0.718214 |
| C | -4.341653 | 0.298786 | 1.404154 |
| C | -5.585791 | 0.147365 | 0.672901 |
| C | -4.295851 | 0.603768 | 2.798437 |
| H | -5.236230 | 0.736422 | 3.356243 |
| C | -3.058528 | 0.737887 | 3.429639 |
| C | -1.875211 | 0.542599 | 2.664629 |
| H | -6.533482 | -0.270079 | -1.220059 |
| H | -2.984230 | -0.986523 | -4.497945 |
| H | -5.236352 | -0.736621 | -3.356102 |
| H | -0.886998 | -0.654682 | -3.135592 |
| H | -6.533440 | 0.269588 | 1.220309 |
| H | -2.984067 | 0.986777 | 4.497904 |
| H | -0.886875 | 0.655226 | 3.135407 |
| N | -1.877913 | -0.235867 | -1.356890 |
| N | -1.877857 | 0.236143 | 1.356807 |
| C | 3.088942 | 0.145716 | -0.716483 |
| C | 1.875519 | 0.574147 | -2.657988 |
| C | 3.088885 | -0.145894 | 0.716562 |
| C | 4.341916 | 0.315225 | -1.400417 |
| C | 3.058875 | 0.778253 | -3.420626 |
| H | 0.887187 | 0.692532 | -3.127336 |
| C | 4.341802 | -0.315589 | 1.400556 |
| C | 5.586012 | 0.155170 | -0.670923 |
| C | 4.296167 | 0.636574 | -2.791011 |
| H | 2.984488 | 1.039682 | -4.485896 |
| C | 4.295938 | -0.636933 | 2.791147 |
| C | 5.585956 | -0.155718 | 0.671123 |
| C | 1.875305 | -0.574150 | 2.658007 |
| H | 6.533699 | 0.283807 | -1.216792 |
| H | 5.236572 | 0.775674 | -3.347203 |
| C | 3.058595 | -0.778431 | 3.420702 |
| H | 5.236296 | -0.776172 | 3.347385 |
| H | 6.533598 | -0.284495 | 1.217038 |
| H | 0.886934 | -0.692391 | 3.127308 |
| H | 2.984117 | -1.039850 | 4.485969 |
| N | 1.878117 | 0.252236 | -1.353891 |
| N | 1.878013 | -0.252237 | 1.353911 |
| Fe | 0.000098 | 0.000102 | -0.000048 |
| N | 0.000086 | 2.021799 | 0.011868 |
| C | 0.000018 | 3.224170 | 0.018173 |
| S | -0.000329 | 4.867827 | 0.027018 |
| N | -0.000064 | -2.021599 | -0.012054 |
| C | -0.000239 | -3.223968 | -0.018735 |
| S | -0.000732 | -4.867629 | -0.026989 |

**$[Fe^{II}(L1R_1R_2)(py)_2]$ ($R_1$ = OEt. $R_2$ = $CH_3$). S = 0. E =** -3082.59213048

| | | | |
|---|---|---|---|
| N | -0.000001 | -1.536202 | 1.038781 |
| C | -0.000009 | -2.811717 | 0.539937 |
| C | -0.000008 | -3.953193 | 1.360505 |
| C | 0.000002 | -3.796869 | 2.762168 |
| C | 0.000010 | -2.487303 | 3.284351 |
| C | 0.000008 | -1.394662 | 2.398752 |
| Fe | 0.000000 | 0.012757 | -0.253195 |
| N | 0.000003 | 1.472089 | -1.655088 |
| C | -1.167137 | 1.971961 | -2.164948 |
| C | -1.208234 | 2.954536 | -3.170098 |
| C | 0.000009 | 3.460622 | -3.690683 |
| C | 1.208249 | 2.954531 | -3.170096 |
| C | 1.167146 | 1.971956 | -2.164946 |
| N | 1.306516 | 0.969764 | 0.793026 |
| C | 2.615445 | 0.732958 | 0.792073 |
| C | 3.315230 | -0.197437 | -0.060398 |
| C | 4.783073 | -0.219679 | 0.229908 |
| O | 5.510112 | -1.123440 | -0.522771 |
| N | -1.306520 | 0.969764 | 0.793021 |
| C | -2.615450 | 0.732963 | 0.792057 |
| C | -3.315233 | -0.197427 | -0.060421 |
| C | -4.783079 | -0.219655 | 0.229873 |
| O | -5.510118 | -1.123439 | -0.522778 |
| O | 1.369675 | -0.941892 | -1.270381 |
| C | 2.645478 | -0.984499 | -1.078506 |
| C | 3.376599 | -1.929038 | -2.041399 |
| O | -1.369670 | -0.941893 | -1.270386 |
| C | -2.645475 | -0.984495 | -1.078522 |
| C | -3.376590 | -1.929020 | -2.041434 |
| C | 0.718453 | 1.961713 | 1.645682 |
| C | -0.718461 | 1.961712 | 1.645680 |
| C | -1.416552 | 2.905224 | 2.442743 |
| C | -0.707327 | 3.826736 | 3.237613 |
| C | 0.707314 | 3.826738 | 3.237614 |
| C | 1.416542 | 2.905227 | 2.442745 |
| O | 5.339521 | 0.495456 | 1.074340 |
| O | -5.339520 | 0.495462 | 1.074326 |
| H | -2.515507 | 2.933428 | 2.435603 |
| H | 2.515497 | 2.933434 | 2.435605 |
| H | -1.260817 | 4.556600 | 3.848117 |
| H | 1.260801 | 4.556604 | 3.848118 |
| H | 3.271574 | 1.281016 | 1.488666 |
| H | -3.271582 | 1.281024 | 1.488645 |
| H | 0.000012 | 4.230486 | -4.477128 |
| H | -2.093176 | 1.566822 | -1.739708 |
| H | -2.182756 | 3.313045 | -3.532640 |
| H | 2.182774 | 3.313036 | -3.532636 |
| H | 2.093182 | 1.566813 | -1.739705 |
| H | 0.000003 | -4.671944 | 3.429673 |
| H | -0.000016 | -2.885705 | -0.554199 |
| H | -0.000015 | -4.950561 | 0.896614 |
| H | 0.000018 | -2.302429 | 4.368458 |
| H | 0.000014 | -0.364848 | 2.777523 |
| H | 4.171878 | -1.404590 | -2.606253 |
| H | 3.876255 | -2.751660 | -1.492231 |
| H | 2.625581 | -2.343616 | -2.739714 |
| H | -3.876269 | -2.751637 | -1.492278 |
| H | -4.171849 | -1.404560 | -2.606304 |
| H | -2.625562 | -2.343607 | -2.739734 |
| C | -6.945087 | -1.151172 | -0.237331 |
| C | -7.575343 | -2.199072 | -1.166504 |
| H | -7.097405 | -1.399425 | 0.833796 |
| H | -7.369019 | -0.138766 | -0.402901 |
| H | -8.668003 | -2.258865 | -0.980783 |
| H | -7.418444 | -1.933809 | -2.232097 |
| H | -7.137583 | -3.202834 | -0.989755 |
| C | 6.945086 | -1.151161 | -0.237348 |
| C | 7.575336 | -2.199054 | -1.166533 |
| H | 7.369007 | -0.138752 | -0.402924 |
| H | 7.097424 | -1.399414 | 0.833776 |
| H | 8.668001 | -2.258834 | -0.980835 |
| H | 7.137592 | -3.202821 | -0.989774 |
| H | 7.418411 | -1.933793 | -2.232123 |

**$[Fe^{II}(L1R_1R_2)(py)_2]$ ($R_1$ = OEt. $R_2$ = $CH_3$). S = 2. E =** -3082.58685436

| | | | |
|---|---|---|---|
| N | -0.000115 | -2.006067 | 0.303665 |
| C | -0.000153 | -3.031693 | -0.588255 |
| C | -0.000219 | -4.385469 | -0.198651 |
| C | -0.000248 | -4.693471 | 1.178220 |
| C | -0.000208 | -3.634659 | 2.109630 |
| C | -0.000142 | -2.310096 | 1.627168 |
| Fe | -0.000004 | 0.190012 | -0.540425 |
| N | 0.000097 | 2.159864 | -1.614813 |
| C | -1.165948 | 2.787365 | -1.936461 |
| C | -1.211922 | 4.040142 | -2.576937 |
| C | 0.000225 | 4.682762 | -2.905311 |
| C | 1.212308 | 4.040028 | -2.576920 |
| C | 1.166207 | 2.787256 | -1.936445 |
| N | 1.343048 | 0.834629 | 0.957529 |
| C | 2.622770 | 0.487292 | 1.047228 |
| C | 3.413385 | -0.202217 | 0.051322 |
| C | 4.815692 | -0.440239 | 0.519164 |
| O | 5.619736 | -1.115831 | -0.381330 |
| N | -1.343033 | 0.834745 | 0.957500 |
| C | -2.622786 | 0.487513 | 1.047170 |
| C | -3.413432 | -0.201934 | 0.051245 |
| C | -4.815725 | -0.440000 | 0.519104 |
| O | -5.619734 | -1.115660 | -0.381370 |
| O | 1.651314 | -0.375548 | -1.561730 |
| C | 2.883004 | -0.597160 | -1.231891 |
| C | 3.703431 | -1.297972 | -2.324230 |
| O | -1.651347 | -0.375392 | -1.561777 |
| C | -2.883073 | -0.596864 | -1.231978 |
| C | -3.703497 | -1.297776 | -2.324256 |
| C | 0.719745 | 1.596313 | 1.996200 |
| C | -0.719687 | 1.596372 | 1.996188 |
| C | -1.411674 | 2.375700 | 2.959863 |
| C | -0.706291 | 3.118980 | 3.927202 |
| C | 0.706443 | 3.118921 | 3.927214 |
| C | 1.411781 | 2.375582 | 2.959888 |
| O | 5.259299 | -0.079383 | 1.617149 |
| O | -5.259334 | -0.079161 | 1.617094 |
| H | -2.510213 | 2.421651 | 2.939421 |
| H | 2.510325 | 2.421440 | 2.939465 |
| H | -1.261744 | 3.718846 | 4.664636 |
| H | 1.261933 | 3.718740 | 4.664658 |
| H | 3.188045 | 0.718791 | 1.968483 |
| H | -3.188056 | 0.719029 | 1.968423 |
| H | 0.000275 | 5.663741 | -3.404755 |
| H | -2.085787 | 2.252171 | -1.661026 |
| H | -2.184003 | 4.499130 | -2.809996 |
| H | 2.184435 | 4.498924 | -2.809965 |
| H | 2.085991 | 2.251974 | -1.660997 |
| H | -0.000300 | -5.740412 | 1.519086 |
| H | -0.000129 | -2.737545 | -1.648375 |
| H | -0.000248 | -5.178137 | -0.961399 |
| H | -0.000229 | -3.825463 | 3.192812 |
| H | -0.000110 | -1.451859 | 2.316138 |
| H | 4.601288 | -0.712585 | -2.602770 |
| H | 4.071230 | -2.284962 | -1.979260 |
| H | 3.046999 | -1.428262 | -3.205150 |
| H | -4.071387 | -2.284693 | -1.979168 |
| H | -4.601296 | -0.712361 | -2.602917 |
| H | -3.047032 | -1.428240 | -3.205126 |
| C | -6.991432 | -1.349432 | 0.071098 |
| C | -7.719317 | -2.105778 | -1.050008 |
| H | -6.968268 | -1.928951 | 1.017492 |
| H | -7.471825 | -0.374284 | 0.295778 |
| H | -8.768096 | -2.308868 | -0.748660 |
| H | -7.736224 | -1.511796 | -1.986709 |
| H | -7.224830 | -3.075775 | -1.262292 |
| C | 6.991452 | -1.349524 | 0.071124 |
| C | 7.719381 | -2.105794 | -1.050005 |
| H | 7.471781 | -0.374349 | 0.295825 |
| H | 6.968332 | -1.929067 | 1.017503 |
| H | 8.768177 | -2.308815 | -0.748670 |
| H | 7.224962 | -3.075821 | -1.262307 |
| H | 7.736238 | -1.511789 | -1.986693 |

**$[Fe^{II}(L1R_1R_2)(py)_2]$ ($R_1$. $R_2 = CH_3$). S = 0. E =** -2854.80681959

| | | | |
|---|---|---|---|
| Fe | -0.000077 | 0.067229 | -0.387009 |
| N | -1.306613 | -0.594076 | 0.864324 |
| N | 1.306570 | -0.595156 | 0.863591 |
| C | 0.718081 | -1.357256 | 1.927762 |
| C | -0.718189 | -1.356501 | 1.928268 |
| C | 1.417041 | -2.090105 | 2.920837 |
| C | -1.417103 | -2.088348 | 2.922126 |
| C | 0.707364 | -2.801552 | 3.908034 |
| H | 2.515923 | -2.121431 | 2.917047 |
| C | -0.707383 | -2.800616 | 3.908699 |
| H | -2.516027 | -2.118290 | 2.919501 |
| H | 1.260656 | -3.371128 | 4.670362 |
| H | -1.260703 | -3.369401 | 4.671601 |
| C | -2.611561 | -0.345539 | 0.818020 |
| C | -3.315744 | 0.375120 | -0.219234 |
| H | -3.268488 | -0.702615 | 1.628944 |
| C | -2.647500 | 0.826744 | -1.421515 |
| O | -1.374248 | 0.718074 | -1.614909 |
| C | 2.611534 | -0.346634 | 0.817324 |
| C | 3.315841 | 0.373745 | -0.220082 |
| H | 3.268389 | -0.703495 | 1.628399 |
| C | 2.647472 | 0.825469 | -1.422271 |
| O | 1.374199 | 0.716892 | -1.615516 |
| C | 1.165735 | -2.319743 | -1.733802 |
| C | 1.206576 | -3.529057 | -2.450341 |
| C | -0.002088 | -4.152039 | -2.822074 |
| C | -1.210194 | -3.528230 | -2.449936 |
| C | -1.168272 | -2.318929 | -1.733421 |
| N | -0.001011 | -1.704922 | -1.370448 |
| H | -0.002513 | -5.099325 | -3.382237 |
| H | 2.091698 | -1.818998 | -1.426550 |
| H | 2.180861 | -3.969933 | -2.707264 |
| H | -2.184862 | -3.968454 | -2.706515 |
| H | -2.093803 | -1.817493 | -1.425959 |
| C | 0.000450 | 2.996417 | -0.342412 |
| C | 0.001088 | 4.309251 | 0.159889 |
| C | 0.002203 | 4.515833 | 1.555360 |
| C | 0.002578 | 3.382466 | 2.393908 |
| C | 0.001881 | 2.100019 | 1.816198 |
| N | 0.000880 | 1.889788 | 0.464996 |
| H | 0.002753 | 5.532162 | 1.977545 |
| H | -0.000474 | 2.789311 | -1.419428 |
| H | 0.000686 | 5.155485 | -0.542950 |
| H | 0.003389 | 3.479857 | 3.489324 |
| H | 0.002053 | 1.200993 | 2.445509 |
| C | -4.778962 | 0.540435 | 0.101001 |
| O | -5.302065 | -0.110142 | 1.027499 |
| C | 4.779106 | 0.538720 | 0.100116 |
| O | 5.301769 | -0.111122 | 1.027402 |
| C | -5.680189 | 1.561595 | -0.631124 |
| H | -5.999937 | 1.190815 | -1.627199 |
| H | -6.584704 | 1.693332 | -0.008466 |
| H | -5.185101 | 2.542485 | -0.779252 |
| C | 5.681156 | 1.558373 | -0.633065 |
| H | 6.586589 | 1.688621 | -0.011436 |
| H | 5.999046 | 1.187284 | -1.629607 |
| H | 5.187481 | 2.540109 | -0.780436 |
| C | -3.381170 | 1.416741 | -2.637879 |
| H | -4.275861 | 0.828941 | -2.921696 |
| H | -3.710351 | 2.460752 | -2.455148 |
| H | -2.667686 | 1.422524 | -3.482854 |
| C | 3.380908 | 1.415624 | -2.638698 |
| H | 3.711584 | 2.459048 | -2.455399 |
| H | 4.274649 | 0.826911 | -2.923675 |
| H | 2.666787 | 1.422900 | -3.483114 |

**$[Fe^{II}(L1R_1R_2)(py)_2]$ ($R_1$. $R_2 = CH_3$). S = 2. E** = -2854.80147499

| | | | |
|---|---|---|---|
| Fe | -0.000001 | 0.100587 | -0.619173 |
| N | 1.341466 | 0.564732 | 0.949260 |
| N | -1.341488 | 0.564896 | 0.949205 |
| C | -0.719209 | 1.229724 | 2.053049 |
| C | 0.719221 | 1.229632 | 2.053083 |
| C | -1.412804 | 1.917843 | 3.082131 |
| C | 1.412851 | 1.917662 | 3.082200 |
| C | -0.706443 | 2.570627 | 4.112370 |
| H | -2.511464 | 1.964332 | 3.063432 |
| C | 0.706522 | 2.570537 | 4.112405 |
| H | 2.511517 | 1.964015 | 3.063560 |
| H | -1.261303 | 3.102077 | 4.900919 |
| H | 1.261411 | 3.101914 | 4.900981 |
| C | 2.607548 | 0.171002 | 1.023436 |
| C | 3.397460 | -0.446066 | -0.023241 |
| H | 3.164273 | 0.291862 | 1.971205 |
| C | 2.889992 | -0.611009 | -1.361192 |
| O | 1.668323 | -0.329714 | -1.688164 |
| C | -2.607607 | 0.171280 | 1.023343 |
| C | -3.397540 | -0.445720 | -0.023357 |
| H | -3.164348 | 0.292195 | 1.971096 |
| C | -2.890040 | -0.610692 | -1.361294 |
| O | -1.668350 | -0.329447 | -1.688235 |
| C | -1.165712 | 2.834289 | -1.718860 |
| C | -1.211761 | 4.146254 | -2.226852 |
| C | 0.000520 | 4.819047 | -2.487652 |
| C | 1.212648 | 4.146172 | -2.226352 |
| C | 1.166301 | 2.834211 | -1.718376 |
| N | 0.000220 | 2.176387 | -1.464844 |
| H | 0.000636 | 5.846092 | -2.883744 |
| H | -2.084647 | 2.272531 | -1.499530 |
| H | -2.183795 | 4.627067 | -2.410832 |
| H | 2.184791 | 4.626920 | -2.409928 |
| H | 2.085112 | 2.272400 | -1.498657 |
| C | -0.000031 | -3.096087 | -1.027642 |
| C | -0.000133 | -4.484018 | -0.785604 |
| C | -0.000374 | -4.937849 | 0.550468 |
| C | -0.000500 | -3.984968 | 1.589912 |
| C | -0.000385 | -2.616326 | 1.252384 |
| N | -0.000157 | -2.172173 | -0.030860 |
| H | -0.000460 | -6.015233 | 0.777229 |
| H | 0.000171 | -2.689901 | -2.050358 |
| H | -0.000025 | -5.190727 | -1.628678 |
| H | -0.000683 | -4.290885 | 2.646365 |
| H | -0.000475 | -1.836522 | 2.028807 |
| C | 4.770199 | -0.843655 | 0.456145 |
| O | 5.217905 | -0.409696 | 1.534967 |
| C | -4.770332 | -0.843166 | 0.456009 |
| O | -5.217975 | -0.409199 | 1.534853 |
| C | 5.650510 | -1.861093 | -0.307650 |
| H | 6.229589 | -1.369238 | -1.117571 |
| H | 6.374187 | -2.269542 | 0.422616 |
| H | 5.068624 | -2.687945 | -0.760285 |
| C | -5.650723 | -1.860560 | -0.307754 |
| H | -6.374361 | -2.269009 | 0.422553 |
| H | -6.229850 | -1.368667 | -1.117616 |
| H | -5.068892 | -2.687415 | -0.760450 |
| C | 3.745279 | -1.066858 | -2.557424 |
| H | 4.761599 | -0.629538 | -2.557071 |
| H | 3.853337 | -2.172260 | -2.582347 |
| H | 3.215398 | -0.757799 | -3.478206 |
| C | -3.745343 | -1.066317 | -2.557598 |
| H | -3.853532 | -2.171702 | -2.582631 |
| H | -4.761607 | -0.628868 | -2.557249 |
| H | -3.215388 | -0.757240 | -3.478332 |

**$[Fe^{II}(L1R_1R_2)(py)_2]$ ($R_1$. $R_2$ = OEt). S = 0. E =** -3310.34830088

| | | | |
|---|---|---|---|
| N | 0.000024 | -0.321982 | -1.769933 |
| C | 0.000033 | -1.658631 | -2.067933 |
| C | 0.000048 | -2.151569 | -3.384096 |
| C | 0.000054 | -1.241621 | -4.461777 |
| C | 0.000046 | 0.136711 | -4.165750 |
| C | 0.000031 | 0.552300 | -2.822268 |
| Fe | 0.000000 | 0.256769 | 0.156113 |
| N | -0.000023 | 0.708603 | 2.120922 |
| C | 1.167262 | 0.849118 | 2.821895 |
| C | 1.208307 | 1.117908 | 4.201481 |
| C | -0.000050 | 1.253722 | 4.915078 |
| C | -1.208393 | 1.117897 | 4.201459 |
| C | -1.167321 | 0.849107 | 2.821874 |
| N | -1.308985 | 1.633543 | -0.192039 |
| C | -2.616323 | 1.437274 | -0.359664 |
| C | -3.329945 | 0.203262 | -0.183569 |
| C | -4.788026 | 0.325594 | -0.497572 |
| O | -5.488629 | -0.858496 | -0.489647 |
| N | 1.308985 | 1.633550 | -0.192012 |
| C | 2.616324 | 1.437284 | -0.359631 |
| C | 3.329947 | 0.203273 | -0.183529 |
| C | 4.788027 | 0.325603 | -0.497539 |
| O | 5.488625 | -0.858491 | -0.489635 |
| O | -1.400171 | -1.108894 | 0.493158 |
| C | -2.656303 | -0.997617 | 0.281286 |
| O | -3.440751 | -2.080639 | 0.552749 |
| O | 1.400173 | -1.108885 | 0.493195 |
| C | 2.656306 | -0.997603 | 0.281336 |
| O | 3.440755 | -2.080623 | 0.552805 |
| C | -0.718567 | 2.932272 | -0.330831 |
| C | 0.718563 | 2.932275 | -0.330816 |
| C | 1.416117 | 4.162597 | -0.442884 |
| C | 0.707169 | 5.373428 | -0.569915 |
| C | -0.707177 | 5.373425 | -0.569932 |
| C | -1.416123 | 4.162590 | -0.442918 |
| O | -5.341114 | 1.400880 | -0.772252 |
| O | 5.341118 | 1.400892 | -0.772203 |
| H | 2.515042 | 4.181125 | -0.414263 |
| H | -2.515049 | 4.181114 | -0.414324 |
| H | 1.261024 | 6.321275 | -0.651564 |
| H | -1.261035 | 6.321269 | -0.651596 |
| H | -3.261765 | 2.279753 | -0.659644 |
| H | 3.261765 | 2.279762 | -0.659614 |
| H | -0.000061 | 1.466389 | 5.994846 |
| H | 2.093415 | 0.750759 | 2.242963 |
| H | 2.183042 | 1.221421 | 4.700350 |
| H | -2.183138 | 1.221400 | 4.700311 |
| H | -2.093463 | 0.750740 | 2.242926 |
| H | 0.000066 | -1.597013 | -5.503305 |
| H | 0.000028 | -2.330470 | -1.201431 |
| H | 0.000054 | -3.238406 | -3.553762 |
| H | 0.000051 | 0.893868 | -4.963304 |
| H | 0.000025 | 1.618726 | -2.564518 |
| C | 6.908695 | -0.733175 | -0.810354 |
| C | 7.509589 | -2.145544 | -0.751732 |
| H | 7.018875 | -0.275186 | -1.815905 |
| H | 7.390742 | -0.045720 | -0.083576 |
| H | 8.593089 | -2.107304 | -0.990168 |
| H | 7.389197 | -2.585604 | 0.259235 |
| H | 7.014678 | -2.816900 | -1.482982 |
| C | 2.768523 | -3.280743 | 1.046203 |
| C | 3.863807 | -4.327908 | 1.297852 |
| H | 2.206229 | -3.036457 | 1.971601 |
| H | 2.032115 | -3.628303 | 0.290920 |
| H | 3.408154 | -5.268412 | 1.671763 |
| H | 4.417997 | -4.551752 | 0.364157 |
| H | 4.589673 | -3.965619 | 2.053719 |
| C | -2.768520 | -3.280758 | 1.046149 |
| C | -3.863804 | -4.327923 | 1.297799 |
| H | -2.032112 | -3.628320 | 0.290866 |
| H | -2.206225 | -3.036471 | 1.971546 |
| H | -3.408152 | -5.268426 | 1.671710 |
| H | -4.589669 | -3.965633 | 2.053666 |
| H | -4.417995 | -4.551767 | 0.364105 |

| | | | |
|---|---|---|---|
| C | -6.908700 | -0.733180 | -0.810364 |
| C | -7.509602 | -2.145544 | -0.751712 |
| H | -7.390741 | -0.045707 | -0.083598 |
| H | -7.018881 | -0.275210 | -1.815924 |
| H | -8.593102 | -2.107303 | -0.990145 |
| H | -7.014696 | -2.816918 | -1.482950 |
| H | -7.389209 | -2.585583 | 0.259264 |

**$[Fe^{II}(L1R_1R_2)(py)_2]$ ($R_1$. $R_2$ = OEt). S = 2. E =** -3310.34442873

| | | | |
|---|---|---|---|
| N | -0.000005 | -1.194780 | -1.497632 |
| C | -0.000011 | -2.542054 | -1.322027 |
| C | -0.000014 | -3.455943 | -2.393670 |
| C | -0.000009 | -2.951519 | -3.711367 |
| C | -0.000002 | -1.554564 | -3.902225 |
| C | 0.000000 | -0.716456 | -2.768864 |
| Fe | 0.000000 | 0.213214 | 0.373759 |
| N | 0.000004 | 1.296836 | 2.319990 |
| C | 1.166267 | 1.659202 | 2.926311 |
| C | 1.211989 | 2.376669 | 4.136392 |
| C | 0.000009 | 2.743855 | 4.758307 |
| C | -1.211973 | 2.376675 | 4.136394 |
| C | -1.166256 | 1.659208 | 2.926313 |
| N | -1.349162 | 1.542900 | -0.557253 |
| C | -2.626110 | 1.293229 | -0.838987 |
| C | -3.427197 | 0.189857 | -0.377319 |
| C | -4.819369 | 0.212921 | -0.927980 |
| O | -5.610136 | -0.853489 | -0.564474 |
| N | 1.349164 | 1.542898 | -0.557255 |
| C | 2.626113 | 1.293227 | -0.838986 |
| C | 3.427199 | 0.189855 | -0.377319 |
| C | 4.819374 | 0.212918 | -0.927976 |
| O | 5.610141 | -0.853489 | -0.564463 |
| O | -1.684470 | -0.823523 | 0.962240 |
| C | -2.891191 | -0.801438 | 0.532937 |
| O | -3.744778 | -1.767534 | 0.984358 |
| O | 1.684468 | -0.823528 | 0.962235 |
| C | 2.891189 | -0.801444 | 0.532932 |
| O | 3.744771 | -1.767549 | 0.984343 |
| C | -0.720373 | 2.741387 | -1.020808 |
| C | 0.720377 | 2.741385 | -1.020809 |
| C | 1.411151 | 3.917128 | -1.415765 |
| C | 0.706253 | 5.063197 | -1.835050 |
| C | -0.706247 | 5.063199 | -1.835049 |
| C | -1.411147 | 3.917130 | -1.415763 |
| O | -5.250604 | 1.110005 | -1.667025 |
| O | 5.250611 | 1.109995 | -1.667027 |
| H | 2.509798 | 3.944513 | -1.372655 |
| H | -2.509793 | 3.944518 | -1.372650 |
| H | 1.262134 | 5.964901 | -2.135380 |
| H | -1.262126 | 5.964904 | -2.135377 |
| H | -3.181655 | 1.975592 | -1.508011 |
| H | 3.181659 | 1.975592 | -1.508006 |
| H | 0.000011 | 3.307156 | 5.704001 |
| H | 2.086334 | 1.358157 | 2.405872 |
| H | 2.184340 | 2.642729 | 4.576456 |
| H | -2.184322 | 2.642740 | 4.576459 |
| H | -2.086325 | 1.358166 | 2.405874 |
| H | -0.000011 | -3.635683 | -4.573914 |
| H | -0.000015 | -2.883617 | -0.275814 |
| H | -0.000019 | -4.538140 | -2.196384 |
| H | 0.000002 | -1.115157 | -4.910385 |
| H | 0.000005 | 0.379182 | -2.867644 |
| C | 6.966004 | -0.824786 | -1.107980 |
| C | 7.682840 | -2.082665 | -0.594745 |
| H | 6.917699 | -0.796839 | -2.217050 |
| H | 7.474124 | 0.106478 | -0.779826 |
| H | 8.724118 | -2.109854 | -0.978642 |
| H | 7.718353 | -2.093501 | 0.513760 |
| H | 7.161706 | -3.001415 | -0.933460 |
| C | 3.195737 | -2.754514 | 1.911005 |
| C | 4.349602 | -3.694991 | 2.290196 |
| H | 2.773980 | -2.236792 | 2.797928 |
| H | 2.363693 | -3.301360 | 1.418363 |
| H | 3.987327 | -4.472906 | 2.994212 |
| H | 4.761252 | -4.197151 | 1.391596 |
| H | 5.170721 | -3.133599 | 2.779940 |
| C | -3.195750 | -2.754493 | 1.911029 |
| C | -4.349620 | -3.694961 | 2.290227 |
| H | -2.363708 | -3.301348 | 1.418393 |
| H | -2.773991 | -2.236766 | 2.797947 |
| H | -3.987350 | -4.472870 | 2.994252 |
| H | -5.170737 | -3.133561 | 2.779964 |
| H | -4.761270 | -4.197129 | 1.391632 |

| | | | |
|---|---|---|---|
| C | -6.965999 | -0.824787 | -1.107990 |
| C | -7.682833 | -2.082668 | -0.594756 |
| H | -7.474121 | 0.106475 | -0.779835 |
| H | -6.917696 | -0.796839 | -2.217060 |
| H | -8.724112 | -2.109858 | -0.978651 |
| H | -7.161699 | -3.001417 | -0.933472 |
| H | -7.718346 | -2.093506 | 0.513749 |