Supporting Information

A Computational Study of a High-Spin Iron(I) Complex for Possible Dinitrogen Reduction to Ammonia

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Contents:

SAMPLE INPUT FILES

Geometry Optimizations

BP86

This is the input for all complexes except $[Fe]_2(\mu_2\text{-}N_2)$.

```
!UKS BP86 def2-SVPD TightSCF NormalOPT def2/J

%MaxCore 3900

%Basis
 newGTO H “def2-SVP” end
end

%pal
 nprocs 32
end

*xyz # #
  COORDINATES
*
```

This is the input for $[Fe]_2(\mu_2\text{-}N_2)$.

```
!UKS BP86 def2-SVPD TightSCF TightOPT FinalGrid6 SlowConv def2/J

%MaxCore 3900

%scf
 MaxIter 1500
end

%Basis
 newGTO H “def2-SVP” end
end

%pal
 nprocs 64
end

*xyz # #
  COORDINATES
*
```

BP86 ZORA

This is the input for all of the complexes of interest.

```
! UKS BP86 ZORA D3BJ ZORA-def2-TZVP SARC/J TightSCF TightOpt Grid5
FinalGrid6 SlowConv PrintBasis

%scf
MaxIter 2000
end

%MaxCore 3900

%pal
nprocs 64
end

*xyz  # #
COORDINATES
*
```

BP86 ZORA CPCM(THF)

This is the input for all of the complexes of interest.

```
! UKS BP86 ZORA D3BJ ZORA-def2-TZVP SARC/J TightSCF TightOpt Grid7
SlowConv PrintBasis CPCM(THF)

%scf
MaxIter 3000
end

%MaxCore 7800

%pal
nprocs 64
end

*xyz  0 7
```

Plotting orbitals with .cube files

This block was added at the end of geometry optimization inputs.

```
*
COORDINATES
*

%plots
 dim1 #
 dim2 #
 dim3 #
 min1 #
 max1 #
 min2 #
 max2 #
 min3 #
 max3 #
 Format Gaussian_Cube
 MO("name.cube",orb#,a/b);
 MO("name.cube",orb#,a/b);
end
```

The values of dim1-dim3 were changed between $[Fe]_2(\mu_2\text{-}N_2)$ (175) and the rest of the complexes (150) due to the drastic size difference. The values of min1-min3 and max1-max3 were changed job to job to center them in the cube when plotted in VESTA. Names of files were assigned accordingly for easy organization. The orbital numbers were decided based on relevant occupancy, and each alpha and beta orbital was plotted, where alpha=0 and beta =1.

Vibrational Frequency Analyses

BP86

This input file was used for all complexes of interest.

```
!UKS BP86 def2-SVPD TightSCF def2/J Grid7

%MaxCore 3900

%method
 Z_Solver DIIS
 Z_MaxIter 250
 Z_MaxDIIS 12
 Z_Tol 1.e-6
end

%scf
 MaxIter 2500
end

%Basis
 newGTO H "def2-SVP" end
end

%Freq
 AnFreq True
end

%pal
 nprocs 64
end

*xyz  0 4
COORDINATES
*
```

BP86 ZORA

This input file was used for all complexes of interest.

```
! UKS BP86 ZORA D3BJ ZORA-def2-TZVP SARC/J TightSCF Grid5 FinalGrid6
SlowConv PrintBasis

%method
 Z_Solver DIIS
 Z_MaxIter 250
 Z_MaxDIIS 12
 Z_Tol 1.0e-7
end

%scf
MaxIter 2000
end

%MaxCore 3900

%Freq
 AnFreq true
end

%pal
nprocs 64
end

*xyz  # #
COORDINATES
*
```

BP86 ZORA CPCM(THF)

This input file was used for all complexes of interest.

```
! UKS BP86 ZORA D3BJ ZORA-def2-TZVP SARC/J TightSCF Grid7 SlowConv
PrintBasis CPCM(THF)

%scf
MaxIter 2500
end

%MaxCore 3900

%Freq
 AnFreq true
end

%pal
nprocs 128
end

*xyz  # #
COORDINATES
*
```

SINGLE-POINT ENERGY CALCULATIONS

BP86

This input file was used for all complexes on interest.

```
!UKS BP86 def2-SVPD TightSCF def2/J

%MaxCore 3900

%scf
 MaxIter 2500
end

%pal
 nprocs 32
end

*xyz  # #
COORDINATES
*
```

BP86 ZORA

This file was used for all complexes on interest.

```
! UKS BP86 ZORA D3BJ def2-TZVP SARC/J TightSCF Grid7 SlowConv PrintBasis

%scf
MaxIter 2000
end

%MaxCore 3900

%pal
nprocs 64
end

*xyz  # #
COORDINATES
*
```

BP86 ZORA CPCM(THF)

This input file was used for all complexes of interest.

```
! UKS BP86 ZORA D3BJ def2-TZVP SARC/J TightSCF Grid7 SlowConv PrintBasis
CPCM(THF)

%scf
MaxIter 3000
end

%MaxCore 3900

%pal
nprocs 128
end

*xyz  # #
COORDINATES
*
```

CASSCF

This file was used for all complexes of interest, and the coordinates used were given by the BP86 ZORA CPCM(THF) geometry optimizations.

```
!EPR-II TightSCF

%MaxCore 7800

%casscf
nel #
norb #
MaxIter 4000
#nroots 10
#rel
 #dosoc true
 #gtensor ture
 #amatrix true
#end
ci
  MaxIter 2000
 end
end

%basis
 NewGTO Fe "CP(PPP)" end
 NewGTO S "def2-TZVP" end
end

%pal
 nprocs 64
end

*xyz # #
COORDINATES
*
```

The values for norb and nel were subject to the specific complex, which is specified in the CASSCF coordinate section of this document. The same %plot block was used to plot the CASSCF-derived orbitals.

SI 2

xyz COORDINATES OF GEOMETRY OPTIMIZATION OUTPUT FILES

BP86

$[Fe]_2(\mu_2\text{-}N_2)$

CARTESIAN COORDINATES (ANGSTROEM)

C	-5.471440	-0.145038	0.088860
C	-4.965256	1.151060	0.272407
C	-3.580378	1.389378	0.194528
C	-2.645673	0.358767	-0.071658
C	-4.573034	-1.194780	-0.168951
C	-3.194517	-0.938798	-0.244035
H	-2.521695	-1.788675	-0.443155
H	-5.651768	1.986261	0.479621
H	-3.231609	2.421193	0.351321
H	-6.553176	-0.336749	0.147623
H	-4.949258	-2.218897	-0.314283
B	-1.006065	0.576341	-0.129488
C	-0.679483	2.217123	-0.044109
S	0.940808	2.745553	-0.694368
C	1.252461	4.483224	-0.032887
C	0.069138	5.373391	-0.440945
H	-0.096005	5.352505	-1.535608
H	0.274627	6.422739	-0.144616
H	-0.869496	5.064778	0.056672
C	2.541126	4.930996	-0.741509
C	1.441306	4.468421	1.487858
H	0.549095	4.075180	2.012097
H	1.610533	5.501823	1.855933
H	2.316323	3.854357	1.773466
C	-0.479561	-0.203092	1.254054
C	-0.478808	-0.071867	-1.572723
S	1.329301	-0.360599	1.568676
S	1.287651	-0.549561	-1.598600
C	1.571919	-0.071494	3.415424
C	1.827930	-0.481930	-3.401541
C	1.789645	0.951996	-3.944668
H	0.778764	1.397346	-3.876581
H	2.080407	0.949929	-5.015914
H	2.489911	1.609117	-3.396029
C	0.900721	-1.408848	-4.203136

H	-0.140108	-1.034680	-4.220771
H	0.897202	-2.434546	-3.786693
H	1.253363	-1.467661	-5.253541
C	3.267213	-1.022727	-3.391005
H	3.928929	-0.395376	-2.763302
H	3.672352	-1.024174	-4.423473
H	3.305881	-2.060701	-3.007878
C	1.138693	1.340154	3.828241
H	1.743304	2.109104	3.314074
H	1.276547	1.468348	4.922759
H	0.069808	1.529706	3.609679
C	0.767047	-1.146499	4.163489
H	-0.321656	-1.042472	3.990423
H	0.943159	-1.052504	5.255876
H	1.072436	-2.164555	3.853817
C	3.080739	-0.262932	3.637441
H	3.671365	0.476008	3.062161
H	3.407490	-1.277337	3.338450
H	3.318200	-0.130424	4.713189
Fe	2.257925	0.951348	-0.089886
N	4.087645	1.068085	-0.141996
N	5.245435	1.113027	-0.149709
H	-1.045115	-0.980341	-1.852364
H	-0.638831	0.675165	-2.376766
H	-0.880849	-1.233220	1.316319
H	-0.899054	0.354052	2.115789
H	2.817429	5.949868	-0.401792
H	2.409693	4.958675	-1.839952
H	3.386143	4.253989	-0.509030
H	-1.425797	2.780212	-0.638093
Fe	7.076247	1.092346	-0.177005
S	8.059947	-0.892832	-0.843410
S	8.217878	2.545862	-1.572382
S	8.275398	1.483315	1.764034
C	9.786097	-0.631642	-0.283808
C	9.949665	1.800235	1.090517
C	9.871718	1.769505	-1.670614
B	10.346229	0.949297	-0.298265
H	10.414827	-1.260464	-0.944112
H	9.865264	-1.044397	0.742585
H	10.650938	1.569129	1.914564
H	10.026531	2.887723	0.884732
H	10.569957	2.599918	-1.891813
H	9.871817	1.086235	-2.544557
C	7.486584	-2.491320	-0.024274
C	7.386084	-2.319636	1.496132

H	8.354301	-2.032836	1.947721
H	7.075206	-3.279137	1.959544
H	6.639773	-1.548162	1.763550
C	8.477322	-3.602298	-0.403860
H	9.483626	-3.423403	0.018882
H	8.574689	-3.700091	-1.502470
H	8.114340	-4.571957	-0.005077
C	6.103693	-2.765449	-0.638202
H	5.392204	-1.944932	-0.424302
H	5.686015	-3.696671	-0.204158
H	6.168597	-2.896939	-1.735424
C	7.831138	2.998324	2.793368
C	6.487043	2.636774	3.447073
H	5.707137	2.447249	2.683814
H	6.144847	3.477106	4.085468
H	6.577054	1.737455	4.086580
C	7.689913	4.243304	1.909046
H	6.872117	4.122511	1.173691
H	8.619896	4.467572	1.353064
H	7.461028	5.125534	2.542760
C	8.922255	3.184670	3.858892
H	9.048041	2.272260	4.474000
H	8.640076	4.017373	4.535730
H	9.899683	3.438316	3.407758
C	7.618254	2.740422	-3.347970
C	7.375139	1.371085	-3.994719
H	8.293344	0.754342	-4.015912
H	7.041955	1.507097	-5.044886
H	6.593684	0.804541	-3.454232
C	8.669640	3.549178	-4.122668
H	9.622664	2.996320	-4.221966
H	8.878918	4.519150	-3.631779
H	8.296323	3.758494	-5.145939
C	6.304405	3.528289	-3.220357
H	5.563934	2.978610	-2.607357
H	5.866760	3.685068	-4.226868
H	6.469255	4.522235	-2.761311
C	11.989587	0.788158	-0.295942
C	12.640730	0.186122	0.812729
C	14.027413	-0.031153	0.850128
C	12.828919	1.156166	-1.375849
C	14.220390	0.946791	-1.354737
C	14.829502	0.351304	-0.239143
H	12.043389	-0.131875	1.683039
H	14.485380	-0.503214	1.733073
H	15.916693	0.184314	-0.218799

H	14.831639	1.252062	-2.217957
H	12.394789	1.620675	-2.274137
H	-0.748647	2.563648	1.007207

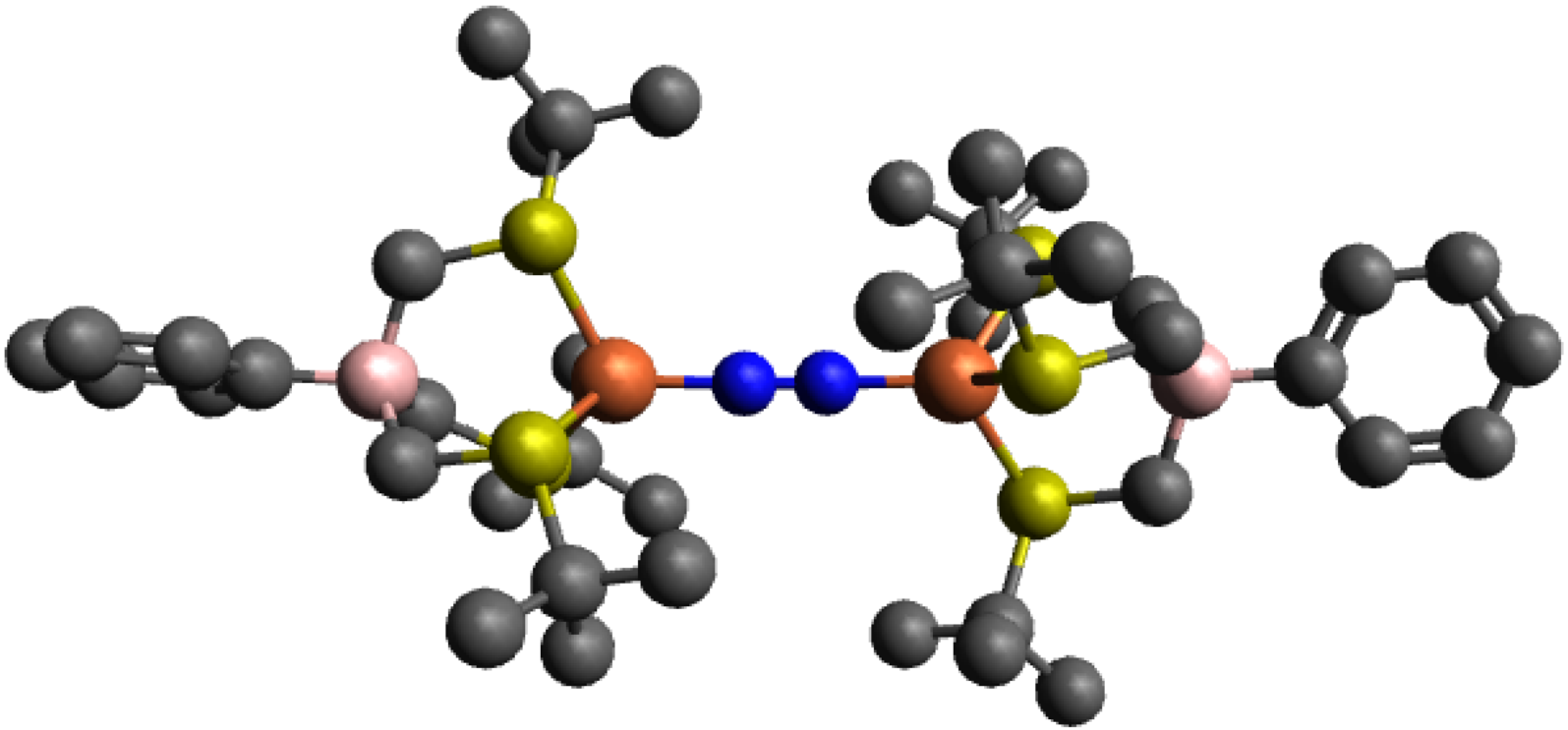

Figure SI1. Optimized structure of $[Fe]_2(\mu\text{-}N_2)$ where hydrogen atoms were not pictured for clarity.

[Fe]

CARTESIAN COORDINATES (ANGSTROEM)

C	-2.611128	-0.531821	-0.662050
C	-1.236127	-0.250869	-0.601703
C	-0.278319	-1.209677	-0.179904
C	-3.088095	-1.803959	-0.302136
C	-2.168921	-2.780958	0.110919
C	-0.795518	-2.482944	0.167160
H	-0.903572	0.756398	-0.898511
H	-2.521977	-3.784295	0.393604
H	-0.112616	-3.282063	0.493244
H	-3.314784	0.247547	-0.992705
B	1.337674	-0.853500	-0.192925
C	2.145229	-2.072003	0.621735
C	1.680155	-0.825849	-1.831103
S	3.862432	-1.726371	1.207334
H	1.591284	-2.354970	1.540871
H	2.225466	-2.982911	-0.006993
S	3.425608	-0.634598	-2.477980
H	1.129199	0.000229	-2.324887
H	1.322947	-1.760477	-2.309331
C	3.767340	-2.301840	-3.144614
H	3.102520	-2.491719	-4.009183
H	3.616831	-3.076396	-2.371137
H	4.819834	-2.313188	-3.483942
C	4.563283	-3.415282	1.292672
H	5.606712	-3.327200	1.647164
H	4.543863	-3.900366	0.298849
H	3.977233	-4.013122	2.016297
Fe	4.335737	-0.301248	-0.441169
H	-4.163631	-2.030761	-0.345933
C	1.533970	0.613969	0.569166
S	3.098441	1.504902	0.151652
H	1.553562	0.460648	1.667602
H	0.718864	1.332834	0.355554
C	3.544525	2.260819	1.752833
H	3.685007	1.481313	2.526017
H	2.744966	2.962179	2.058329
H	4.488245	2.819669	1.610334

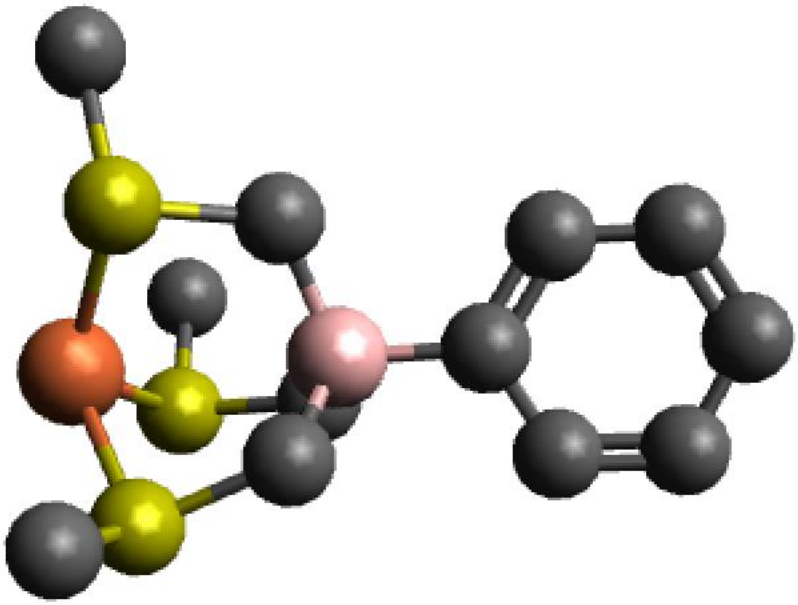

Figure SI2. Optimized structure of [Fe] where hydrogen atoms were not pictured for clarity.

[Fe](N_2)

CARTESIAN COORDINATES (ANGSTROEM)

C	-5.843534	1.980661	-0.070057
C	-5.038869	3.116953	0.107160
C	-3.637090	3.007094	0.057196
C	-2.978128	1.773584	-0.170872
C	-3.824642	0.646584	-0.337786
C	-5.225331	0.738135	-0.292223
H	-5.837882	-0.165890	-0.430304
H	-6.940256	2.060866	-0.033106
H	-5.504162	4.097707	0.289418
H	-3.048048	3.924598	0.207644
H	-3.377616	-0.346713	-0.504709
B	-1.332419	1.593096	-0.184440
C	-0.641243	3.118798	-0.146271
S	1.091474	3.236128	-0.704121
H	-0.688739	3.532219	0.881958
H	-1.209091	3.798754	-0.811873
C	1.769215	4.868110	-0.049950
C	1.851562	4.856182	1.480416
H	0.859085	4.715437	1.949662
H	2.249391	5.827830	1.840003
H	2.530289	4.058809	1.836771
C	0.860875	5.999331	-0.555521
H	-0.154573	5.939482	-0.122462
H	0.770820	5.983985	-1.658929
H	1.292034	6.978199	-0.260182
C	3.170032	4.977788	-0.674790
H	3.820620	4.133348	-0.374801
H	3.654633	5.914981	-0.332806
H	3.118265	5.002230	-1.780364
C	-1.039797	0.769436	1.244779
C	-0.942560	0.790399	-1.594402
S	0.676302	-0.070423	-1.587251
H	-0.930495	1.524872	-2.426273
H	-1.691915	0.018814	-1.851811
S	0.654243	0.134250	1.586331
H	-1.708561	-0.106897	1.343433
H	-1.292544	1.462928	2.071992
C	0.957244	0.348837	3.430773
C	0.949845	1.828535	3.830189
H	1.767011	2.380077	3.330930
H	1.089357	1.920425	4.927987

H	-0.008923	2.320843	3.577601
C	-0.130288	-0.438352	4.179243
H	-1.138997	-0.017956	4.003905
H	0.065464	-0.393251	5.270890
H	-0.139011	-1.502546	3.875264
C	2.344191	-0.272718	3.666654
H	3.129655	0.254609	3.091330
H	2.364925	-1.342381	3.382624
H	2.604887	-0.200217	4.742361
C	1.261293	-0.147239	-3.376370
C	1.561398	1.253298	-3.923567
H	0.674179	1.913338	-3.886377
H	1.874747	1.177558	-4.985747
H	2.375902	1.741814	-3.357446
C	0.168285	-0.848836	-4.197745
H	-0.759543	-0.248794	-4.248343
H	-0.080995	-1.841404	-3.775764
H	0.527945	-0.999537	-5.236179
C	2.538344	-1.002172	-3.323140
H	3.313666	-0.534088	-2.686301
H	2.955478	-1.107072	-4.345079
H	2.332458	-2.017401	-2.932694
Fe	1.888609	1.188465	-0.048326
N	3.714288	0.933987	-0.029102
N	4.837101	0.802992	0.013089

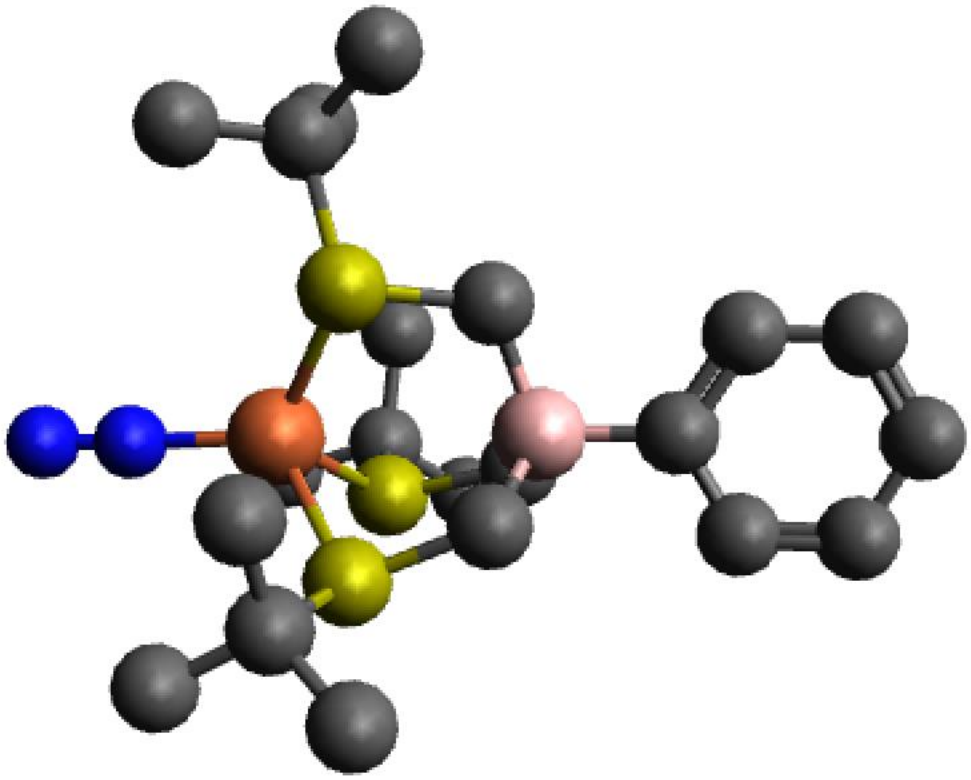

Figure SI3. Optimized structure of [Fe](N_2) where hydrogen atoms were not pictured for clarity.

[Fe](N)

CARTESIAN COORDINATES (ANGSTROEM)

C	-5.996936	-0.028834	-0.018791
C	-5.312343	1.192603	-0.113537
C	-3.905375	1.220029	-0.095331
C	-3.125475	0.043066	0.014698
C	-5.256012	-1.217598	0.099196
C	-3.852722	-1.172110	0.115856
H	-5.876108	2.133365	-0.205558
H	-3.411561	2.200546	-0.171413
H	-7.096714	-0.055685	-0.036018
H	-5.775494	-2.184741	0.178998
H	-3.304954	-2.123979	0.213441
B	-1.480130	0.019412	0.092433
C	-0.887465	1.567276	-0.109580
S	0.857932	1.657085	-0.633807
H	-0.984408	2.162586	0.821029
H	-1.451595	2.089648	-0.907202
C	1.515315	3.353572	-0.153900
C	0.604919	4.407634	-0.803006
H	0.542960	4.274510	-1.900569
H	1.022978	5.416103	-0.605238
H	-0.419277	4.383647	-0.386940
C	2.930513	3.388707	-0.754757
H	3.406974	4.359720	-0.508111
H	2.907004	3.291822	-1.858011
H	3.556963	2.577623	-0.332770
C	1.566203	3.504860	1.370696
H	2.248820	2.758409	1.817008
H	0.568253	3.400662	1.837805
H	1.943278	4.518325	1.619922
C	-0.946182	-0.984231	-1.137201
S	0.643802	-1.800849	-0.771883
H	-0.829599	-0.408930	-2.078138
H	-1.668571	-1.799280	-1.331159
C	1.433268	-2.276992	-2.411282
C	0.430786	-3.152056	-3.178479
H	0.126445	-4.036377	-2.586259
H	0.897946	-3.513997	-4.116741
H	-0.479918	-2.587640	-3.456288
C	2.683614	-3.081640	-2.019077
H	2.420032	-3.995039	-1.452060
H	3.379451	-2.477160	-1.403211
H	3.226567	-3.391439	-2.934822

C	1.814920	-1.025991	-3.213419
H	0.943529	-0.370720	-3.404279
H	2.227603	-1.327060	-4.197965
H	2.584313	-0.430393	-2.686452
C	-1.125611	-0.593123	1.620613
S	0.487778	-0.066668	2.280046
H	-1.167771	-1.701052	1.622559
H	-1.874635	-0.231034	2.351322
C	1.004286	-1.285054	3.616167
C	2.259086	-0.652383	4.242565
H	3.039803	-0.467692	3.477425
H	2.672449	-1.339297	5.009603
H	2.020601	0.308016	4.739138
C	1.328216	-2.652105	2.998520
H	2.192127	-2.580874	2.310056
H	0.468371	-3.073017	2.443560
H	1.589065	-3.369233	3.804029
C	-0.140730	-1.383485	4.635374
H	-0.405790	-0.390760	5.047587
H	0.180191	-2.027690	5.479213
H	-1.049941	-1.835271	4.196673
Fe	1.902265	-0.152911	0.398817
N	3.336895	0.381189	0.989333

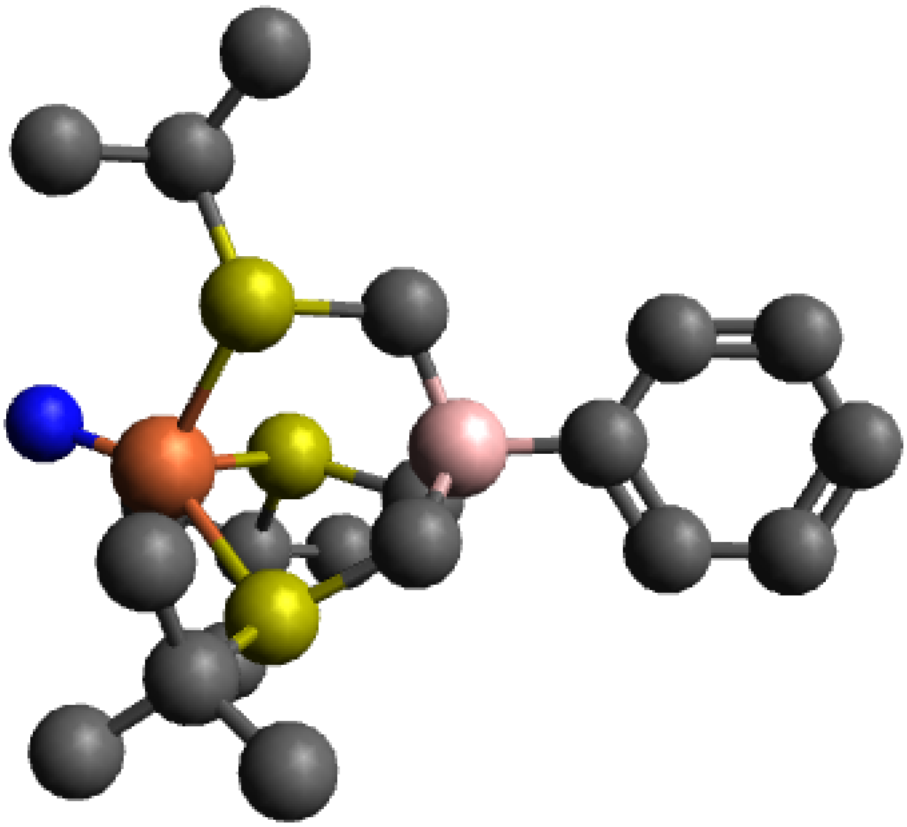

Figure SI4. Optimized structure of [Fe](N) where hydrogen atoms were not pictured for clarity.

[Fe](NH)

CARTESIAN COORDINATES (ANGSTROEM)

C	-5.980021	-0.040972	0.083465
C	-5.307774	1.188113	-0.003038
C	-3.901194	1.227709	-0.011673
C	-3.108461	0.056084	0.062184
C	-5.226488	-1.224677	0.165381
C	-3.823318	-1.166720	0.155618
H	-5.881285	2.125425	-0.066623
H	-3.416705	2.213464	-0.079152
H	-7.079704	-0.077399	0.087876
H	-5.735953	-2.197746	0.238190
H	-3.266299	-2.115526	0.226443
B	-1.461256	0.058579	0.118256
C	-0.888774	1.604327	-0.128625
S	0.836344	1.678407	-0.716018
H	-0.955633	2.207592	0.798955
H	-1.483690	2.115958	-0.910831
C	1.487588	3.389355	-0.288256
C	0.574043	4.419873	-0.971747
H	0.519362	4.253814	-2.065185
H	0.982843	5.437279	-0.800905
H	-0.452682	4.399509	-0.561214
C	2.902385	3.416856	-0.890578
H	3.367467	4.404971	-0.694367
H	2.878458	3.265730	-1.987870
H	3.537990	2.633588	-0.432445
C	1.536287	3.600028	1.229206
H	2.200218	2.856087	1.705746
H	0.534419	3.531855	1.694441
H	1.931019	4.614988	1.443757
C	-0.919049	-0.972828	-1.084327
S	0.688009	-1.740199	-0.688273
H	-0.809582	-0.423577	-2.041730
H	-1.621808	-1.809525	-1.256520
C	1.501954	-2.209169	-2.322428
C	0.511420	-3.094941	-3.093077
H	0.200306	-3.973430	-2.495362
H	0.993014	-3.466909	-4.020076
H	-0.396267	-2.535806	-3.390229
C	2.753775	-3.004326	-1.916293
H	2.490228	-3.922936	-1.357530
H	3.433407	-2.395495	-1.286982
H	3.313020	-3.304554	-2.825581

C	1.881575	-0.961166	-3.128427
H	1.008993	-0.309611	-3.325175
H	2.299987	-1.266289	-4.109319
H	2.646557	-0.359450	-2.602246
C	-1.092789	-0.507558	1.653128
S	0.555209	-0.071375	2.320698
H	-1.189784	-1.611605	1.685091
H	-1.811497	-0.091443	2.385342
C	0.951482	-1.413612	3.587368
C	2.251561	-0.939196	4.259168
H	3.079383	-0.872035	3.525744
H	2.549691	-1.663656	5.044329
H	2.123656	0.050260	4.739954
C	1.158747	-2.776324	2.915811
H	2.012484	-2.748345	2.212100
H	0.262049	-3.109872	2.359916
H	1.374047	-3.541677	3.690069
C	-0.199977	-1.454387	4.604448
H	-0.382957	-0.457891	5.051562
H	0.059861	-2.153644	5.425316
H	-1.142757	-1.810346	4.149528
Fe	1.905747	-0.088582	0.413595
N	3.381324	0.500969	0.989842
H	3.899284	0.614620	1.872987

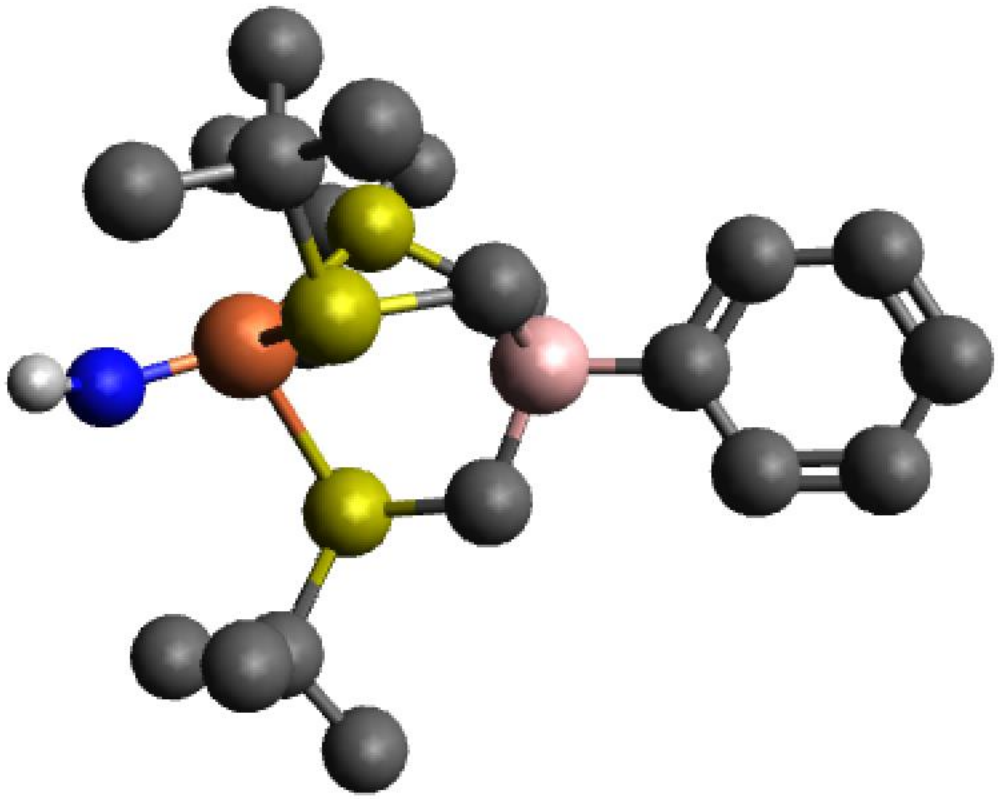

Figure SI5. Optimized structure of [Fe](NH) where hydrogen were not pictured for clarity.

[Fe](NH$_2$)

CARTESIAN COORDINATES (ANGSTROEM)

C	-5.937621	-0.134763	-0.220569
C	-5.280645	1.089122	-0.420658
C	-3.878387	1.165247	-0.328412
C	-3.074354	0.036359	-0.035756
C	-5.172780	-1.276486	0.075919
C	-3.774613	-1.182249	0.164161
H	-5.862167	1.994023	-0.654352
H	-3.406821	2.145333	-0.493484
H	-7.033436	-0.199835	-0.295275
H	-5.669406	-2.245262	0.239711
H	-3.209492	-2.099246	0.398455
B	-1.433205	0.081262	0.134340
C	-0.884334	1.619789	-0.180641
S	0.905687	1.785394	-0.541703
H	-1.122766	2.286445	0.674097
H	-1.383496	2.040530	-1.076554
C	1.359746	3.516790	0.068517
C	0.478341	4.522632	-0.688976
H	0.573933	4.399353	-1.785677
H	0.792178	5.555978	-0.431576
H	-0.589407	4.421505	-0.417778
C	2.836047	3.691056	-0.324420
H	3.169074	4.717497	-0.068278
H	2.988838	3.544250	-1.411308
H	3.486708	2.983475	0.224867
C	1.185010	3.649486	1.585365
H	1.853566	2.961145	2.134341
H	0.146033	3.442064	1.904678
H	1.424448	4.687701	1.897374
C	-0.807965	-1.036494	-0.940287
S	0.822911	-1.788553	-0.543019
H	-0.704902	-0.559384	-1.935821
H	-1.495969	-1.895469	-1.057252
C	1.517116	-2.298683	-2.227956
C	0.471290	-3.187105	-2.920008
H	0.167678	-4.036576	-2.277541
H	0.903227	-3.604143	-3.852677
H	-0.436084	-2.620036	-3.200192
C	2.773474	-3.120572	-1.897527
H	2.530585	-4.003794	-1.275672
H	3.525783	-2.513209	-1.359417
H	3.242326	-3.479614	-2.836176

C	1.864083	-1.070584	-3.075963
H	0.978618	-0.432105	-3.258774
H	2.250016	-1.395030	-4.064298
H	2.633651	-0.452769	-2.577335
C	-1.154002	-0.340791	1.724000
S	0.544448	0.031316	2.274526
H	-1.326063	-1.425320	1.872433
H	-1.834577	0.205689	2.405582
C	0.971459	-1.267838	3.580156
C	2.373394	-0.869689	4.071672
H	3.113019	-0.896484	3.246537
H	2.707779	-1.577359	4.857672
H	2.376640	0.148374	4.508939
C	0.983108	-2.687572	3.002119
H	1.765128	-2.805958	2.228549
H	0.010639	-2.962587	2.551246
H	1.188545	-3.414668	3.814989
C	-0.063784	-1.129988	4.707226
H	-0.108975	-0.092302	5.090991
H	0.218228	-1.793556	5.550295
H	-1.077711	-1.423290	4.375525
Fe	1.860008	-0.046589	0.476163
N	3.609355	0.024289	-0.045351
H	4.183324	-0.760900	-0.365827
H	4.122172	0.884228	-0.262156

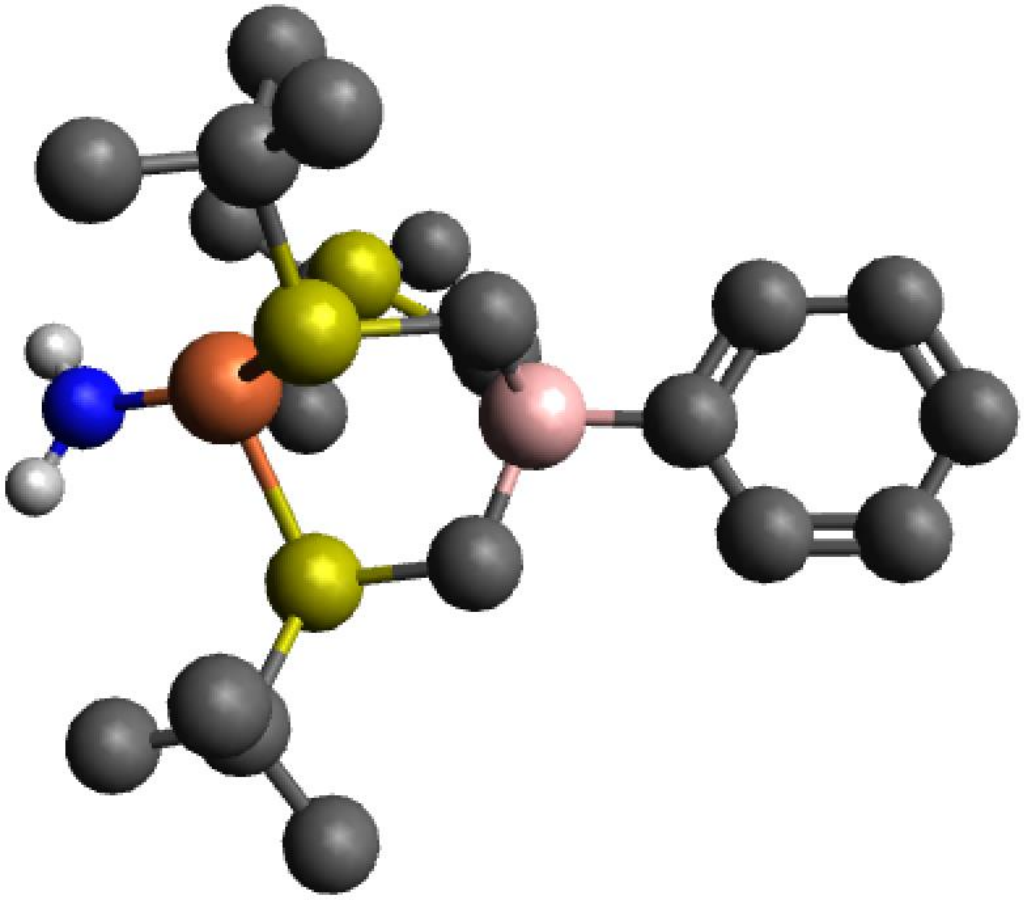

Figure SI6. Optimized structure of [Fe](NH_2) where hydrogens were not pictured for clarity.

[Fe](NH$_3$)

CARTESIAN COORDINATES (ANGSTROEM)

H	1.714612	-1.226972	-4.451993
H	3.250662	-3.070044	-3.419776
H	0.799865	-3.620521	-4.053383
H	-0.545475	-2.892189	-3.128504
C	1.456868	-2.212436	-2.527978
C	0.485754	-3.282817	-3.043460
C	1.436240	-0.960511	-3.410603
H	0.431900	-0.497471	-3.442538
H	3.112480	3.614038	-1.540841
H	3.193506	4.798694	-0.197305
C	2.879462	-2.779521	-2.416063
H	3.527751	3.068874	0.121564
H	-0.939365	-0.880954	-1.907175
C	2.894972	3.764191	-0.465050
S	0.978135	-1.777805	-0.730353
C	-0.784687	-1.193872	-0.854074
H	3.578810	-2.024928	-2.002921
H	0.722026	4.364060	-2.077744
H	2.909594	-3.675558	-1.765619
H	0.466689	-4.166640	-2.377061
H	0.771502	5.554911	-0.734744
H	2.147123	-0.196584	-3.042877
H	-1.402121	-2.100612	-0.700255
C	0.537321	4.500299	-0.994017
C	1.406198	3.551644	-0.156792
H	-1.247000	1.803533	-1.391737
S	1.056315	1.762448	-0.727989
Fe	1.962691	-0.028017	0.362276
C	-0.763603	1.539459	-0.429181
H	-0.541699	4.342427	-0.803060
B	-1.271799	0.061478	0.127960
H	-3.262044	2.064582	-0.646310
C	-3.735240	1.121686	-0.334909
C	1.134385	3.708408	1.342450
C	-2.929160	0.026891	0.063469
C	-5.141028	1.058428	-0.349499
H	-1.054814	2.318121	0.305624
H	-5.723338	1.936658	-0.668633
H	0.076245	3.504388	1.595524
H	1.352035	4.751294	1.656275
C	-3.632796	-1.144216	0.450154
C	-5.801356	-0.117997	0.038310

H	1.761431	3.023224	1.942592
C	-5.034771	-1.225149	0.441932
H	-3.066573	-2.032726	0.774384
H	-6.900382	-0.172011	0.027295
S	0.812595	0.021952	2.334842
H	-5.532362	-2.157084	0.752081
C	-0.946312	-0.156646	1.747321
H	1.547451	-2.913288	2.208396
H	-1.535904	0.552677	2.361905
H	-1.263453	-1.175103	2.044634
C	0.829230	-2.736245	3.031068
C	1.020342	-1.346466	3.648078
H	3.191477	-1.345235	3.333519
H	0.981704	-3.517144	3.805980
H	-0.191178	-2.872065	2.624757
C	2.461816	-1.174603	4.150475
H	2.630751	-0.160629	4.564599
C	0.016559	-1.086996	4.779835
H	2.672210	-1.910796	4.953337
H	0.166618	-1.825948	5.595428
H	-1.028643	-1.182447	4.429713
H	0.146307	-0.074083	5.208355
N	4.099629	-0.077924	0.516630
H	4.564200	0.223934	-0.348518
H	4.458564	-1.018129	0.723730
H	4.451100	0.535690	1.262158

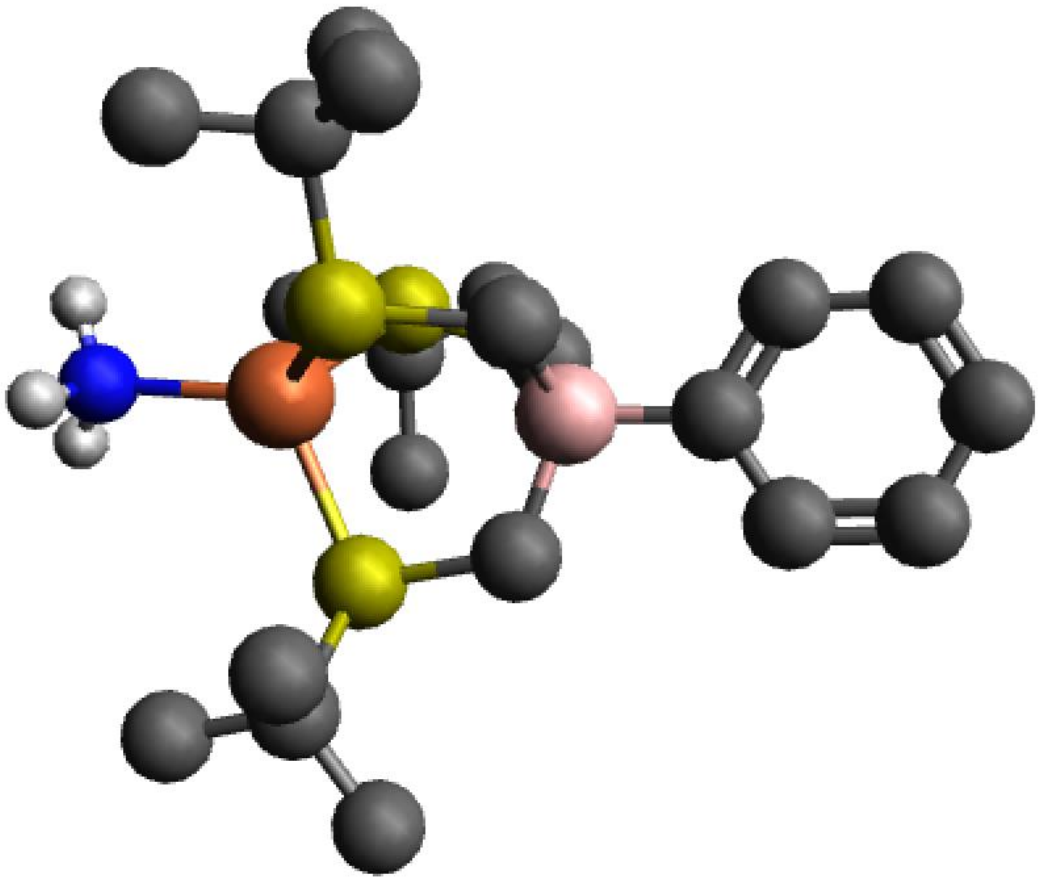

Figure SI7. Optimized structure of [Fe](NH_3) where hydrogen atoms were not pictured for clarity.

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$[Fe]_2(\mu_2\text{-}N_2)$

CARTESIAN COORDINATES (ANGSTROEM)

C	-4.461024	-1.922073	0.444138
C	-4.273093	-0.640856	0.965515
C	-3.054359	0.020448	0.783624
C	-1.982595	-0.556589	0.076344
C	-3.413284	-2.530209	-0.252274
C	-2.203228	-1.855242	-0.425577
H	-1.400805	-2.363982	-0.967434
H	-5.079577	-0.152642	1.517397
H	-2.943715	1.019191	1.213280
H	-5.410831	-2.442017	0.580495
H	-3.540544	-3.534664	-0.662166
B	-0.518710	0.154551	-0.065872
C	-0.652034	1.763965	0.357228
S	0.619556	2.805848	-0.419819
C	0.615401	4.481696	0.412648
C	-0.760490	5.112612	0.199360
H	-1.002511	5.183498	-0.869892
H	-0.775171	6.128004	0.625798
H	-1.548083	4.528328	0.694782
C	1.708030	5.253907	-0.331693
C	0.950207	4.366459	1.894954
H	0.177764	3.804117	2.436037
H	1.006450	5.370374	2.344993
H	1.918687	3.867718	2.039851
C	0.393115	-0.700840	1.027915
C	-0.007065	-0.006321	-1.636564
S	2.129639	-0.165435	1.371931
S	1.815269	-0.024894	-1.784989
C	2.215882	0.198094	3.213977
C	2.224097	0.588581	-3.509847
C	1.839146	2.056542	-3.673422
H	0.770662	2.224816	-3.484993
H	2.058447	2.380701	-4.703422
H	2.409291	2.689449	-2.980369
C	1.489994	-0.299226	-4.514267
H	0.400777	-0.190680	-4.425182
H	1.746345	-1.357810	-4.368650
H	1.775828	-0.012256	-5.538547
C	3.740809	0.417498	-3.625132
H	4.263430	0.994991	-2.850696

H	4.077933	0.780901	-4.607881
H	4.035323	-0.635888	-3.528234
C	1.054620	1.066836	3.682548
H	1.034268	2.016192	3.137392
H	1.174131	1.287990	4.755169
H	0.084695	0.568790	3.554960
C	2.227953	-1.147781	3.940886
H	1.303158	-1.710554	3.751297
H	2.307836	-0.986722	5.027934
H	3.078553	-1.762880	3.617735
C	3.542671	0.939091	3.389110
H	3.545037	1.880940	2.822807
H	4.391088	0.333310	3.045169
H	3.698488	1.172623	4.453527
Fe	2.426428	1.466781	-0.155334
N	4.152569	1.948553	-0.328397
N	5.312855	1.935358	-0.412446
H	-0.360813	-0.936071	-2.100628
H	-0.385018	0.833098	-2.240669
H	0.477003	-1.758127	0.735720
H	-0.117181	-0.689690	2.001348
H	1.782953	6.273808	0.075266
H	1.482827	5.327017	-1.404903
H	2.684641	4.761944	-0.215637
H	-1.617647	2.164117	0.015072
Fe	7.062116	1.536585	-0.407845
S	7.452885	-0.652046	-0.931734
S	8.592977	2.520707	-1.770810
S	8.136814	1.687393	1.586169
C	9.111347	-0.821200	-0.166149
C	9.870576	1.584357	1.015099
C	9.957645	1.303808	-1.710840
B	10.097566	0.525077	-0.252748
H	9.586836	-1.672021	-0.673615
H	8.955787	-1.105087	0.886201
H	10.463150	1.300131	1.894722
H	10.183450	2.596804	0.715988
H	10.867415	1.874234	-1.947600
H	9.789701	0.579668	-2.522806
C	6.311931	-1.814225	0.000456
C	6.124622	-1.333361	1.435884
H	7.074235	-1.294770	1.986416
H	5.445554	-2.018338	1.967484
H	5.680430	-0.329838	1.448618
C	6.906141	-3.220336	-0.053161
H	7.867251	-3.273803	0.475810

H	7.063010	-3.544685	-1.091291
H	6.214181	-3.928979	0.428351
C	4.992290	-1.734862	-0.767770
H	4.602484	-0.709678	-0.785001
H	4.233447	-2.360184	-0.275120
H	5.112287	-2.082305	-1.803060
C	7.916660	3.379463	2.360804
C	6.480600	3.359070	2.891008
H	5.764001	3.197379	2.073615
H	6.248583	4.323959	3.367443
H	6.342075	2.564008	3.636758
C	8.089871	4.475359	1.312005
H	7.334374	4.382326	0.520009
H	9.081480	4.438860	0.841295
H	7.978319	5.462903	1.787474
C	8.929438	3.520841	3.496374
H	8.814743	2.713510	4.233010
H	8.777407	4.482492	4.011246
H	9.960364	3.500407	3.117988
C	8.022113	2.602977	-3.554053
C	7.503005	1.246501	-4.022571
H	8.275347	0.468157	-3.966357
H	7.179186	1.322182	-5.072737
H	6.648664	0.919110	-3.417416
C	9.201050	3.076555	-4.404181
H	10.027341	2.353096	-4.379298
H	9.578785	4.046785	-4.053314
H	8.881296	3.187323	-5.452229
C	6.892854	3.636137	-3.537098
H	6.074684	3.311608	-2.878594
H	6.490379	3.757336	-4.554322
H	7.252053	4.615140	-3.189863
C	11.602295	-0.087638	-0.101778
C	12.004334	-0.682602	1.111751
C	13.254033	-1.282948	1.274142
C	12.535127	-0.140329	-1.153523
C	13.792967	-0.734886	-1.006951
C	14.160951	-1.308743	0.210878
H	11.314841	-0.683314	1.961473
H	13.523664	-1.731620	2.232888
H	15.141496	-1.772667	0.330876
H	14.488520	-0.749762	-1.849119
H	12.281036	0.290685	-2.124864
H	-0.585862	1.898423	1.446919

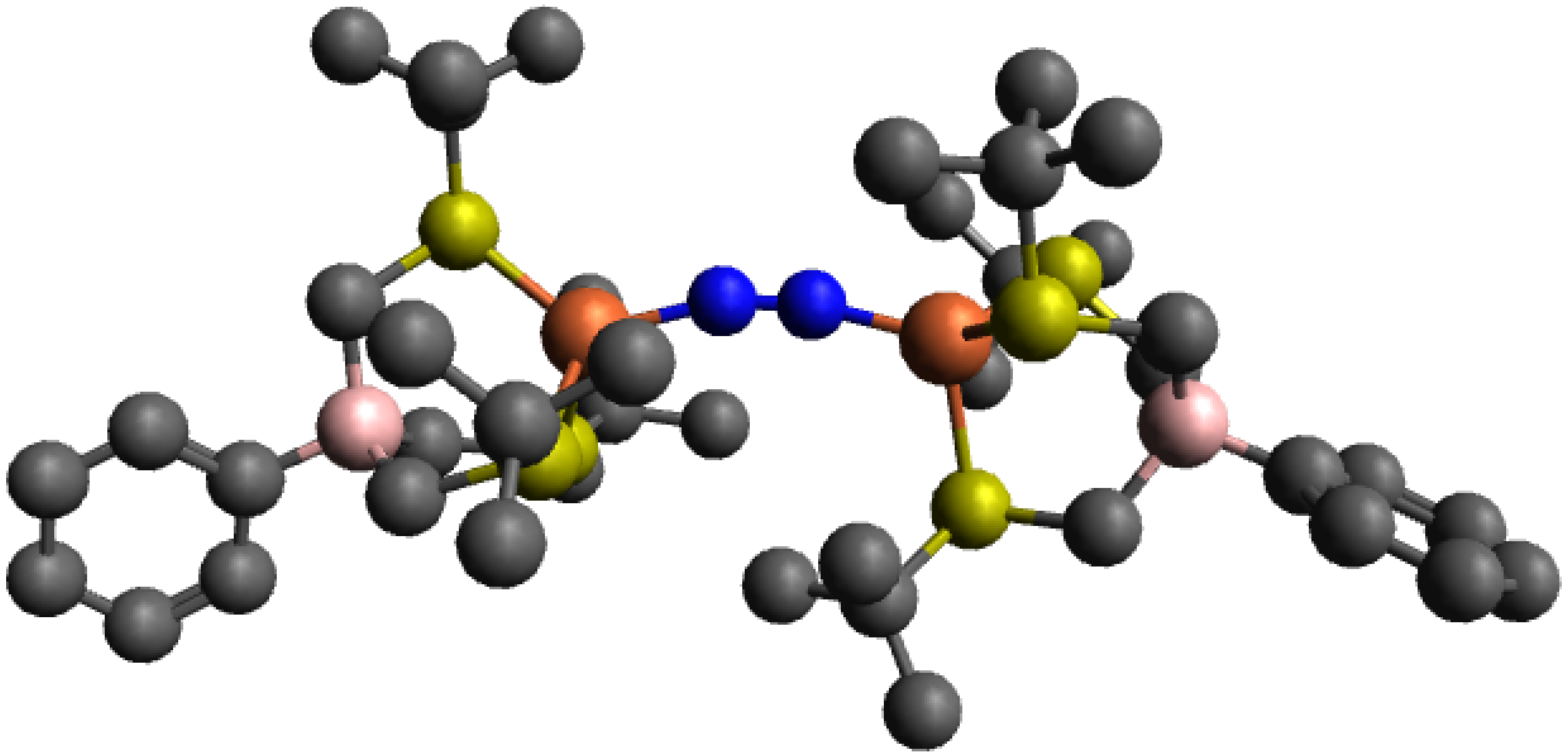


Figure SI8. Optimized structure of $[Fe]_2(\mu\text{-}N_2)$. H atoms not pictured for clarity.

[Fe]

CARTESIAN COORDINATES (ANGSTROEM)

Fe	3.148075	11.126020	1.937168
S	3.130107	9.004643	2.510356
S	5.210944	11.697342	1.435928
B	4.985463	10.641162	4.271717
C	3.918481	11.781825	4.805446
C	4.260414	9.183486	3.971591
H	3.648299	8.868578	4.829890
H	5.017241	8.397852	3.836514
C	3.983710	7.748460	1.387778
C	3.891840	6.383991	2.071867
H	4.346353	5.612117	1.430414
H	2.846104	6.105666	2.261496
H	4.428235	6.379229	3.031058
C	5.430953	8.137042	1.106824
H	5.880306	7.401612	0.420428
H	6.036182	8.146814	2.022837
C	3.155283	7.781617	0.102875
H	3.540187	7.034095	-0.607783
H	3.214033	8.770716	-0.374590
C	5.964939	11.192249	3.054197
H	6.727864	10.450834	2.777072
H	6.514070	12.077560	3.407881
S	2.537240	12.302120	3.690171
C	5.636961	13.527159	1.245032
H	4.421771	12.724348	5.069125
H	3.422225	11.428772	5.722131
H	5.496068	9.129368	0.640740
H	2.098147	7.553194	0.301326
C	0.958441	11.833788	4.614603
C	5.220319	14.334898	2.469138
C	4.841117	13.961296	0.013309
C	7.142466	13.623472	0.995627
C	0.963099	10.367821	5.032891
C	-0.150558	12.102363	3.596328
C	0.838365	12.762513	5.823188
H	4.144313	14.237540	2.666332
H	5.765667	14.023239	3.369793
H	5.446246	15.399612	2.297844
H	3.759865	13.874040	0.195309
H	5.096482	13.350742	-0.864784
H	5.065162	15.012870	-0.223153
H	7.434246	13.048183	0.106261

H	7.714977	13.244852	1.853837
H	7.429611	14.675521	0.839048
H	1.766330	10.148818	5.748824
H	1.083935	9.705037	4.165468
H	0.007237	10.124822	5.524036
H	-1.132279	11.910941	4.056040
H	-0.047102	11.442556	2.722384
H	-0.132161	13.145473	3.249003
H	-0.094738	12.549008	6.368605
H	1.673675	12.619231	6.522850
H	0.825730	13.816495	5.513336
C	6.037396	10.320364	5.490278
C	6.012540	10.960582	6.743534
C	6.936428	10.665981	7.752120
C	7.930315	9.710133	7.538069
C	7.986329	9.056039	6.304057
C	7.055976	9.361406	5.309688
H	5.252892	11.717340	6.950944
H	6.878436	11.187138	8.710444
H	8.653162	9.476834	8.321736
H	8.757839	8.305495	6.117941
H	7.129332	8.831706	4.354963

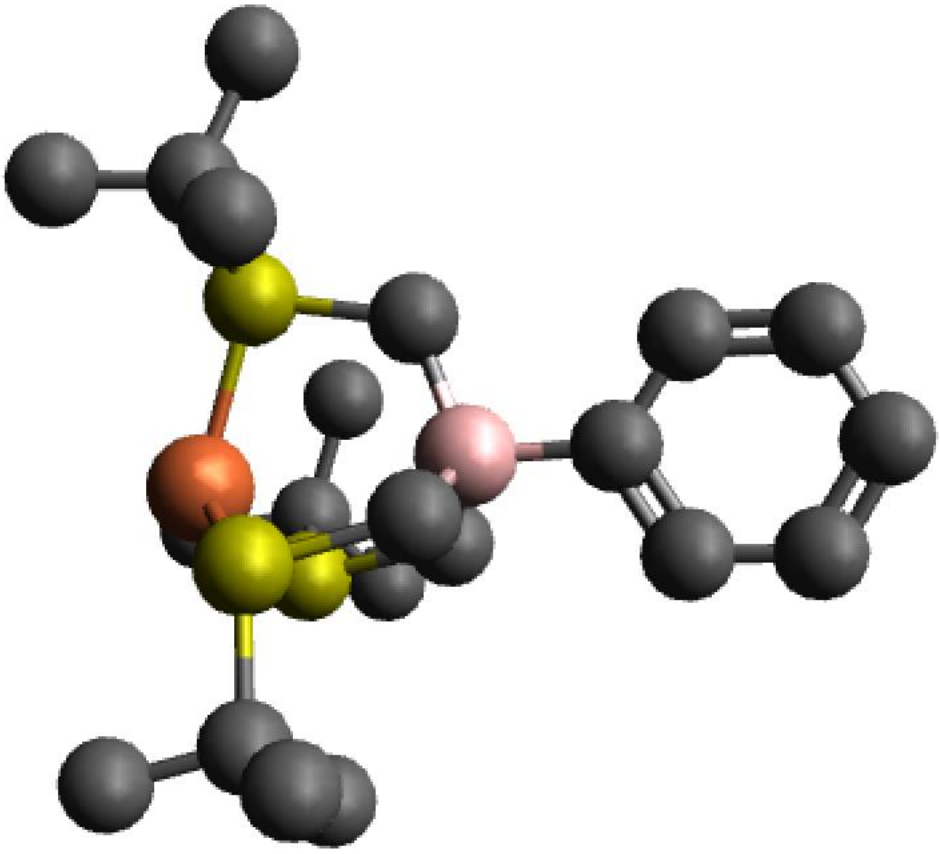


Figure SI9. Optimized structure of [Fe]. H atoms not pictured for clarity.

[Fe](N_2)

CARTESIAN COORDINATES (ANGSTROEM)

C	-5.960333	-0.094295	-0.009809
C	-5.308189	1.138621	-0.059013
C	-3.910490	1.196513	-0.049267
C	-3.111412	0.039890	0.003280
C	-5.196788	-1.263291	0.053810
C	-3.802926	-1.187611	0.059534
H	-5.889992	2.061964	-0.105150
H	-3.437334	2.180866	-0.080634
H	-7.050460	-0.145113	-0.018869
H	-5.690789	-2.236515	0.097578
H	-3.233825	-2.120596	0.115138
B	-1.482244	0.066272	0.095473
C	-0.933356	1.609475	-0.163695
S	0.807886	1.693599	-0.716368
H	-1.017756	2.213371	0.753204
H	-1.511958	2.109362	-0.953710
C	1.456047	3.362559	-0.163482
C	0.545135	4.444550	-0.743075
H	0.483095	4.367718	-1.837427
H	0.946757	5.437834	-0.488819
H	-0.470993	4.377029	-0.332104
C	2.860771	3.438419	-0.766323
H	3.328923	4.394546	-0.486956
H	2.827797	3.378416	-1.863057
H	3.497677	2.625493	-0.391335
C	1.513702	3.433548	1.360597
H	2.196997	2.674614	1.764289
H	0.526555	3.277380	1.815955
H	1.874460	4.427273	1.669950
C	-0.908985	-0.977133	-1.069829
S	0.680889	-1.780792	-0.651529
H	-0.764492	-0.432198	-2.015559
H	-1.614998	-1.795454	-1.262989
C	1.499441	-2.220267	-2.279882
C	0.534065	-3.097179	-3.076921
H	0.245651	-3.991001	-2.506508
H	1.019595	-3.424846	-4.009392
H	-0.377910	-2.548167	-3.346946
C	2.752111	-2.999721	-1.872990
H	2.491844	-3.907939	-1.311748
H	3.417414	-2.384073	-1.252018
H	3.307919	-3.298874	-2.774712

C	1.872492	-0.954635	-3.048213
H	0.994489	-0.330060	-3.260720
H	2.331252	-1.230095	-4.011030
H	2.591936	-0.348112	-2.482048
C	-1.166912	-0.451285	1.650345
S	0.466769	0.036115	2.322752
H	-1.229373	-1.549113	1.701635
H	-1.910777	-0.040556	2.347034
C	0.934305	-1.269603	3.585194
C	2.236151	-0.750078	4.199484
H	3.019516	-0.642580	3.436563
H	2.594563	-1.462658	4.958032
H	2.086622	0.223957	4.685743
C	1.154958	-2.616243	2.900543
H	1.981155	-2.560785	2.179182
H	0.258424	-2.954784	2.364111
H	1.403347	-3.377878	3.656602
C	-0.180796	-1.344316	4.627861
H	-0.352466	-0.365447	5.096574
H	0.100512	-2.059986	5.416134
H	-1.125050	-1.686348	4.183485
Fe	1.698282	-0.055933	0.422980
N	3.506126	-0.115863	0.588836
N	4.634463	-0.157320	0.693633

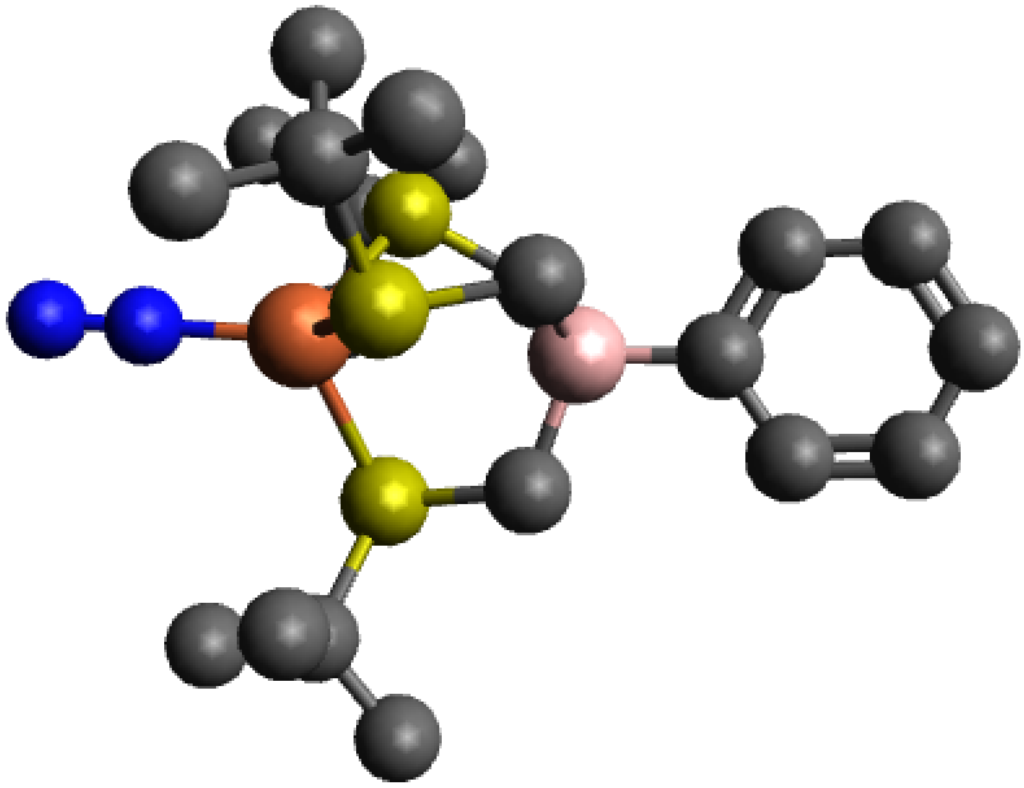


Figure SI10. Optimized structure of [Fe](N_2) where H atoms are not pictured for clarity.

[Fe](N)

CARTESIAN COORDINATES (ANGSTROEM)

C	-6.009638	-0.001880	0.016983
C	-5.324020	1.213670	0.023383
C	-3.925220	1.232608	0.018392
C	-3.159362	0.052938	0.001455
C	-5.278934	-1.193433	0.010160
C	-3.883582	-1.156769	0.002232
H	-5.880247	2.153636	0.032539
H	-3.424857	2.203923	0.032451
H	-7.100752	-0.022084	0.019290
H	-5.799862	-2.153438	0.009618
H	-3.339733	-2.106358	0.002675
B	-1.532761	0.026378	0.069231
C	-0.934327	1.562226	-0.115152
S	0.796462	1.617756	-0.684355
H	-0.988577	2.134282	0.824152
H	-1.503731	2.108242	-0.881040
C	1.505950	3.267739	-0.152089
C	0.619026	4.363164	-0.745294
H	0.547461	4.273011	-1.838094
H	1.055001	5.346133	-0.509021
H	-0.395272	4.335576	-0.324668
C	2.909906	3.280410	-0.760487
H	3.410052	4.223009	-0.490266
H	2.871373	3.213072	-1.857084
H	3.504602	2.443163	-0.365111
C	1.574352	3.355285	1.370295
H	2.245296	2.583577	1.769865
H	0.585255	3.239819	1.834325
H	1.964567	4.345122	1.654868
C	-0.990637	-0.955616	-1.159632
S	0.555761	-1.809110	-0.717518
H	-0.809055	-0.366705	-2.071689
H	-1.714455	-1.744249	-1.404357
C	1.442193	-2.254210	-2.305094
C	0.506559	-3.134191	-3.133579
H	0.200494	-4.028191	-2.572975
H	1.024493	-3.460427	-4.048662
H	-0.396491	-2.586111	-3.434585
C	2.678920	-3.029080	-1.842844
H	2.399487	-3.934520	-1.286490
H	3.318360	-2.405854	-1.199776
H	3.270575	-3.331238	-2.720250

C	1.846033	-0.992909	-3.066046
H	0.978171	-0.366362	-3.311331
H	2.335337	-1.276491	-4.011093
H	2.552001	-0.387183	-2.480767
C	-1.176507	-0.599103	1.581178
S	0.410206	-0.001498	2.237315
H	-1.161744	-1.699755	1.562486
H	-1.936479	-0.283993	2.309736
C	1.016759	-1.225808	3.518666
C	2.249074	-0.550165	4.125434
H	2.990780	-0.318932	3.345986
H	2.707341	-1.228399	4.861672
H	1.976162	0.382284	4.638922
C	1.388326	-2.546406	2.846722
H	2.222524	-2.404700	2.144791
H	0.540944	-2.980477	2.298658
H	1.703330	-3.271521	3.613619
C	-0.094394	-1.414006	4.550744
H	-0.386275	-0.455675	5.001890
H	0.267162	-2.075848	5.353109
H	-0.986191	-1.877062	4.107205
Fe	1.808559	-0.164924	0.380718
N	3.256420	0.354488	0.935002

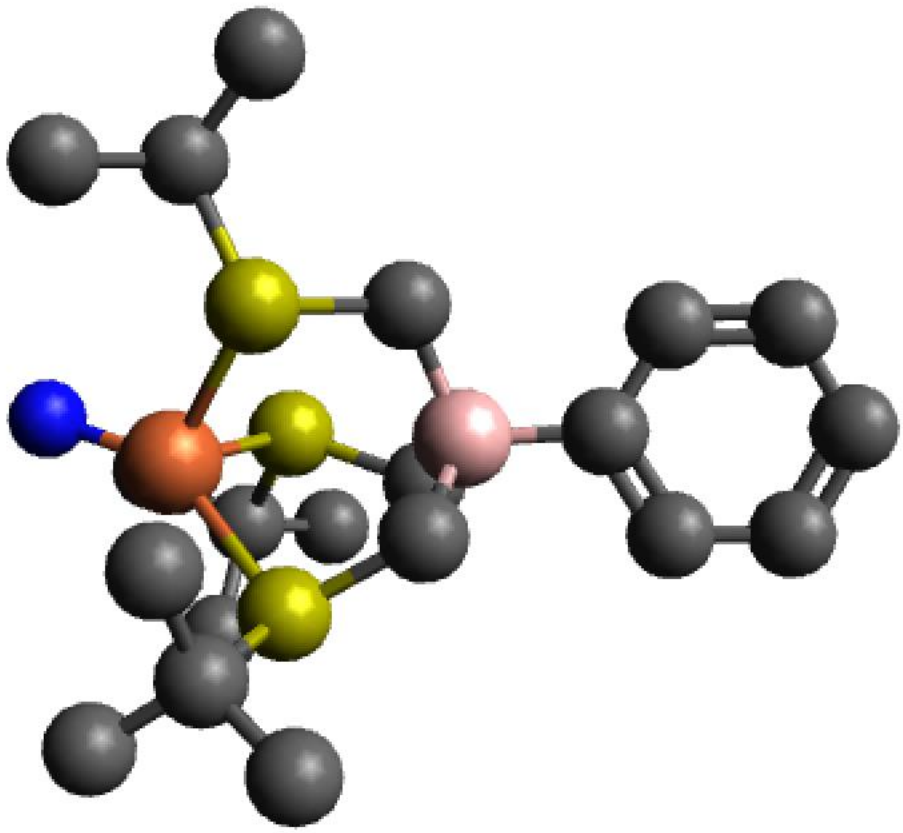


Figure SI11. Optimized structure of [Fe](N) where H atoms are not pictured for clarity.

[Fe](NH)

CARTESIAN COORDINATES (ANGSTROEM)

C	-5.984695	-0.032885	0.105595
C	-5.317348	1.192493	0.069216
C	-3.919360	1.231874	0.041573
C	-3.135669	0.063601	0.043181
C	-5.236190	-1.213186	0.118175
C	-3.841638	-1.155920	0.087160
H	-5.887683	2.124073	0.062313
H	-3.432238	2.209751	0.021183
H	-7.075269	-0.069089	0.125627
H	-5.742474	-2.180531	0.150783
H	-3.283814	-2.097284	0.103244
B	-1.506698	0.070572	0.087946
C	-0.931352	1.600834	-0.156486
S	0.787512	1.643242	-0.762717
H	-0.973349	2.192126	0.771367
H	-1.520753	2.126075	-0.921977
C	1.478884	3.307557	-0.259497
C	0.583208	4.384298	-0.875318
H	0.517283	4.271773	-1.966383
H	1.006256	5.376928	-0.655467
H	-0.432819	4.351949	-0.459303
C	2.883649	3.332922	-0.866043
H	3.366915	4.291236	-0.620735
H	2.845723	3.237879	-1.960738
H	3.492001	2.515356	-0.452231
C	1.541015	3.431696	1.260645
H	2.201739	2.662648	1.679834
H	0.548716	3.330820	1.721227
H	1.934662	4.425376	1.527149
C	-0.963066	-0.954411	-1.103982
S	0.613146	-1.738537	-0.642275
H	-0.808386	-0.403969	-2.044623
H	-1.665316	-1.774629	-1.302827
C	1.509294	-2.181663	-2.229479
C	0.588592	-3.094924	-3.038342
H	0.295581	-3.980919	-2.458026
H	1.113939	-3.434202	-3.944451
H	-0.322351	-2.568518	-3.353444
C	2.762477	-2.923549	-1.758034
H	2.502276	-3.831427	-1.195992
H	3.381259	-2.278367	-1.116591
H	3.364927	-3.217660	-2.630945

C	1.885435	-0.927023	-3.013549
H	1.006638	-0.316510	-3.260116
H	2.370792	-1.218962	-3.958200
H	2.586932	-0.300703	-2.445513
C	-1.135562	-0.485752	1.612861
S	0.508580	-0.008543	2.257633
H	-1.207297	-1.583517	1.649808
H	-1.850206	-0.080451	2.343266
C	0.963170	-1.357965	3.484548
C	2.268471	-0.874693	4.120094
H	3.063391	-0.798350	3.365207
H	2.591993	-1.593926	4.888020
H	2.141740	0.106939	4.597605
C	1.170453	-2.693489	2.776610
H	1.996866	-2.630449	2.055583
H	0.270887	-3.017524	2.236794
H	1.415737	-3.468768	3.519754
C	-0.155989	-1.436502	4.523812
H	-0.322897	-0.461456	5.002512
H	0.122410	-2.160209	5.305690
H	-1.101530	-1.769885	4.076373
Fe	1.832837	-0.101903	0.388471
N	3.340737	0.453040	0.888879
H	3.937318	0.463832	1.726952

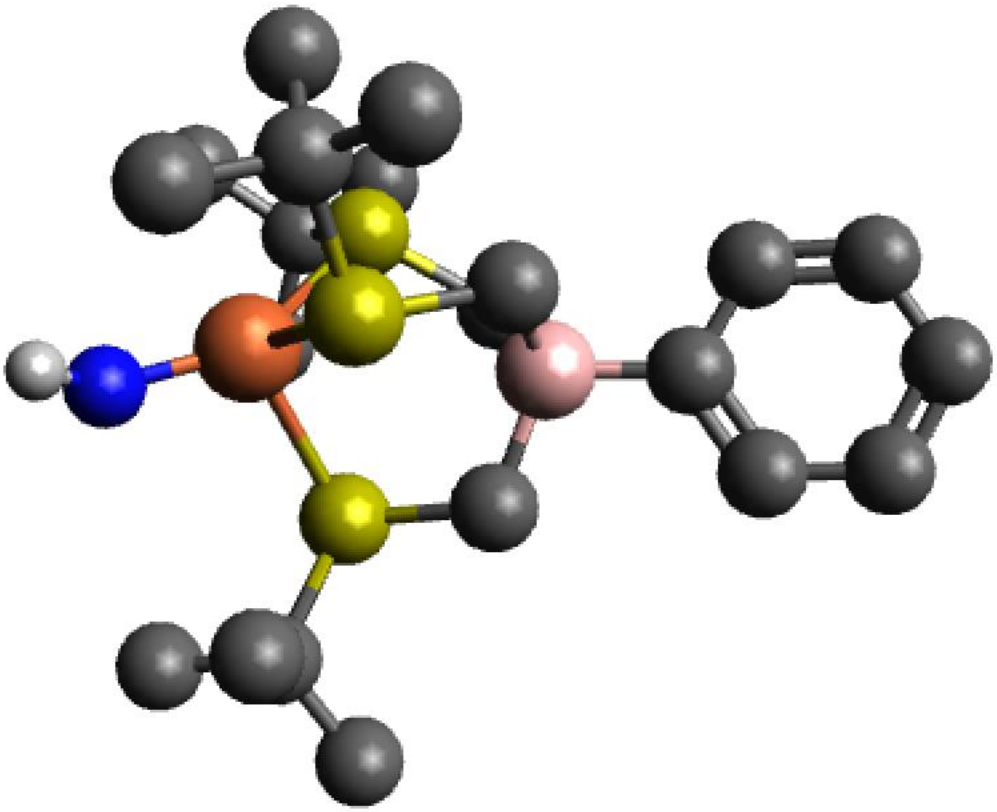


Figure SI12. Optimized structure of [Fe](NH) where H atoms are not pictured for clarity.

[Fe](NH$_2$)

CARTESIAN COORDINATES (ANGSTROEM)

C	-5.998048	-0.127492	-0.039670
C	-5.357116	1.112305	-0.047516
C	-3.959986	1.182296	-0.031384
C	-3.150505	0.031899	-0.013330
C	-5.223899	-1.290983	-0.011089
C	-3.830765	-1.203040	0.001181
H	-5.947366	2.031298	-0.066379
H	-3.495301	2.171346	-0.029997
H	-7.087667	-0.187888	-0.053985
H	-5.709274	-2.269517	-0.000266
H	-3.252462	-2.131598	0.027856
B	-1.523059	0.069603	0.086365
C	-0.985359	1.620243	-0.145713
S	0.756940	1.723432	-0.695535
H	-1.073891	2.217480	0.775055
H	-1.564803	2.124484	-0.932191
C	1.389108	3.393925	-0.125781
C	0.506974	4.475666	-0.748206
H	0.489318	4.392174	-1.843707
H	0.899144	5.470067	-0.483195
H	-0.525575	4.411947	-0.379069
C	2.818033	3.452104	-0.671770
H	3.279439	4.412292	-0.394725
H	2.831816	3.366374	-1.767480
H	3.428520	2.643046	-0.244554
C	1.396051	3.481146	1.398585
H	2.066409	2.726704	1.831618
H	0.394602	3.333697	1.824428
H	1.748972	4.478037	1.707191
C	-0.919765	-0.941438	-1.088131
S	0.632446	-1.779272	-0.619328
H	-0.728018	-0.370260	-2.009716
H	-1.630276	-1.740883	-1.336736
C	1.494677	-2.214329	-2.222190
C	0.546800	-3.069728	-3.062870
H	0.225009	-3.963462	-2.510471
H	1.060790	-3.396814	-3.980498
H	-0.347262	-2.505076	-3.359954
C	2.724443	-3.013275	-1.781610
H	2.432461	-3.927545	-1.245794
H	3.363217	-2.405818	-1.123199
H	3.311175	-3.304544	-2.666614

C	1.914018	-0.949267	-2.968267
H	1.054881	-0.306140	-3.201345
H	2.397785	-1.224374	-3.919105
H	2.627589	-0.364105	-2.372829
C	-1.203312	-0.480424	1.633650
S	0.382439	0.109211	2.320288
H	-1.191527	-1.581050	1.650434
H	-1.978518	-0.139364	2.333423
C	0.960822	-1.146221	3.580275
C	2.225022	-0.521820	4.177441
H	2.976082	-0.326912	3.398021
H	2.662591	-1.211008	4.915879
H	1.998906	0.427193	4.682978
C	1.276240	-2.478314	2.902589
H	2.087048	-2.370151	2.169169
H	0.402494	-2.886642	2.377716
H	1.589595	-3.212362	3.661881
C	-0.134694	-1.303841	4.634526
H	-0.375911	-0.340180	5.103748
H	0.207621	-1.995773	5.419977
H	-1.054091	-1.716915	4.198146
Fe	1.788154	0.003726	0.452345
N	3.643785	-0.216948	0.504864
H	4.144495	-0.857152	1.121636
H	4.297184	0.113710	-0.205288

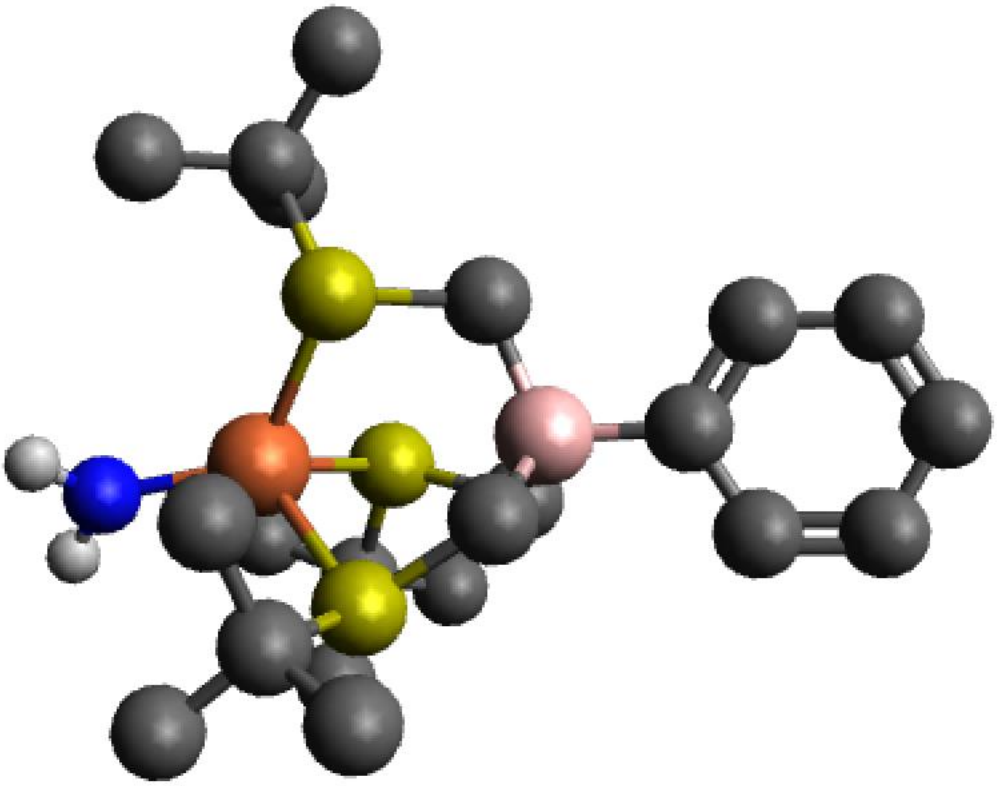


Figure SI13. Optimized structure of [Fe](NH_2) where H atoms were not pictured for clarity.

[Fe](NH3)

CARTESIAN COORDINATES (ANGSTROEM)

H	1.105605	-1.007425	-4.340757
H	3.175906	-2.374796	-3.648429
H	0.856887	-3.533932	-3.933072
H	-0.387479	-3.271335	-2.691498
C	1.393449	-2.031997	-2.461054
C	0.696383	-3.332392	-2.861439
C	0.879069	-0.848401	-3.273890
H	-0.208753	-0.732597	-3.181193
H	3.383473	3.500596	-0.869538
H	3.065361	4.669116	0.439213
C	2.911205	-2.155638	-2.602446
H	3.284702	2.943145	0.825667
H	-1.088779	-1.514656	-1.468307
C	2.864549	3.644682	0.089682
S	1.128678	-1.778806	-0.607417
C	-0.677714	-1.403228	-0.453834
H	3.401613	-1.213538	-2.315593
H	1.269560	4.310871	-2.045504
H	3.307124	-2.964789	-1.971039
H	1.085954	-4.183084	-2.285195
H	0.855434	5.431186	-0.717771
H	1.352382	0.090059	-2.953651
H	-1.085928	-2.240335	0.130701
C	0.758604	4.393430	-1.076175
C	1.357778	3.426456	-0.054942
H	-0.855053	1.214755	-1.812060
S	1.222044	1.677362	-0.755996
Fe	2.049372	-0.027104	0.388605
C	-0.590879	1.324162	-0.749096
H	-0.310293	4.193648	-1.234362
B	-1.129078	0.062080	0.161068
H	-3.047811	1.718786	-1.241940
C	-3.542095	0.934650	-0.664019
C	0.671293	3.547596	1.301470
C	-2.768020	0.032463	0.090181
C	-4.939068	0.868069	-0.707805
H	-1.062854	2.258672	-0.409687
H	-5.498870	1.591156	-1.305742
H	-0.412116	3.386402	1.225888
H	0.827730	4.561041	1.705548
C	-3.489471	-0.950653	0.799715
C	-5.618954	-0.117773	0.008851

H	1.077204	2.820829	2.018715
C	-4.882314	-1.033064	0.766386
H	-2.943366	-1.683484	1.401610
H	-6.708575	-0.173858	-0.021696
S	0.975201	0.099724	2.321170
H	-5.397038	-1.812514	1.333058
C	-0.783809	0.258534	1.763419
H	0.866681	-2.867545	1.860650
H	-1.098976	1.251939	2.116721
H	-1.342511	-0.466061	2.372754
C	0.343969	-2.621648	2.795299
C	0.961488	-1.400739	3.469073
H	2.976982	-1.925601	2.848540
H	0.410521	-3.489828	3.471074
H	-0.717511	-2.464306	2.563602
C	2.440581	-1.648731	3.768266
H	2.916804	-0.754807	4.197621
C	0.201312	-1.016657	4.738662
H	2.546470	-2.474407	4.488832
H	0.201409	-1.858870	5.449641
H	-0.845602	-0.767504	4.516287
H	0.663258	-0.148029	5.228048
N	4.156420	-0.082321	0.539817
H	4.611500	0.197951	-0.333023
H	4.498924	-1.019653	0.767269
H	4.500058	0.546355	1.270759

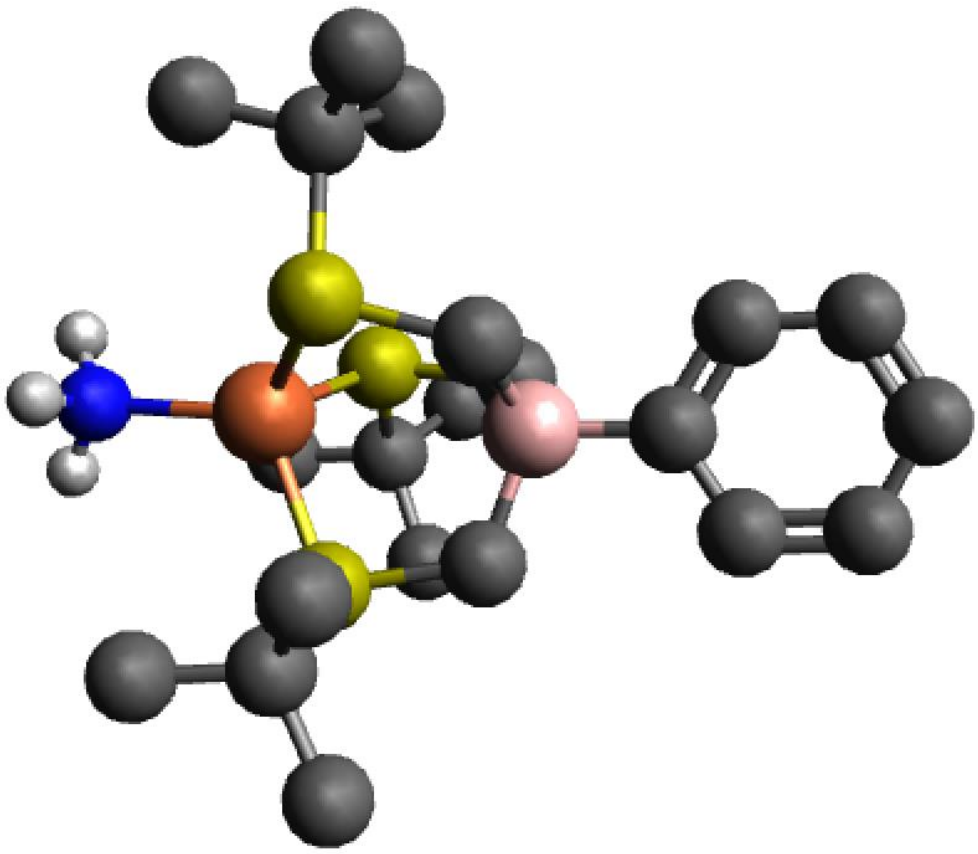


Figure SI14. Optimized structure of [Fe](NH$_3$) where H atoms were not pictured for clarity.

BP86 ZORA CPCM(THF)

$[Fe]_2(\mu_2\text{-}N_2)$

CARTESIAN COORDINATES (ANGSTROEM)

Fe	3.314189	11.161610	1.996237
S	3.389118	8.976745	2.601055
S	5.524566	11.619368	1.805247
B	5.155159	10.372282	4.459387
C	6.875526	8.884475	5.889129
H	6.751302	8.072363	5.168658
C	7.769903	8.692671	6.950088
H	8.312171	7.748313	7.040046
C	7.970566	9.704712	7.892383
H	8.665135	9.560251	8.722079
C	7.268418	10.907651	7.754811
H	7.416234	11.711848	8.479581
C	6.377690	11.080581	6.691927
H	5.847541	12.034093	6.609778
C	6.145725	10.077989	5.725407
C	3.710004	10.931356	5.085217
C	4.931962	8.975813	3.585974
H	4.859709	8.094883	4.240482
H	5.766565	8.806301	2.887748
C	3.678871	7.871845	1.113717
C	4.053534	6.478062	1.615635
H	4.163822	5.797125	0.757269
H	3.277319	6.073722	2.280387
H	5.007785	6.492739	2.160010
C	4.763128	8.454839	0.211196
H	4.922174	7.783659	-0.647357
H	5.720748	8.563029	0.738257
C	2.326490	7.864801	0.394786
H	2.397463	7.241837	-0.509468
H	2.039751	8.880997	0.089438
C	5.979345	11.533970	3.579511
H	7.061875	11.343090	3.602371
H	5.807025	12.532129	4.010806
S	2.848304	12.126842	3.999171
C	5.803210	13.387663	1.240319
H	3.849527	11.444776	6.046295
H	3.022528	10.088087	5.255763
H	4.467672	9.442149	-0.168614
H	1.533338	7.455233	1.035878
C	1.014551	12.012121	4.378014
C	4.875188	14.344352	1.983823

C	5.458827	13.353479	-0.251507
C	7.272745	13.738097	1.468867
C	0.474052	10.631073	4.016628
C	0.387944	13.086259	3.483942
C	0.809365	12.326579	5.859162
H	3.822427	14.099913	1.788739
H	5.038632	14.313259	3.069580
H	5.063339	15.374028	1.641368
H	4.414653	13.046945	-0.405985
H	6.111616	12.658741	-0.798406
H	5.587901	14.359232	-0.678991
H	7.934856	13.033988	0.945672
H	7.524094	13.725924	2.538427
H	7.471982	14.751134	1.085726
H	0.978087	9.833651	4.579160
H	0.602285	10.428702	2.944692
H	-0.600577	10.586948	4.253496
H	-0.702212	13.095179	3.633652
H	0.586072	12.878732	2.422995
H	0.778010	14.085468	3.723427
H	-0.267931	12.328545	6.087536
H	1.288898	11.571828	6.497583
H	1.218835	13.313685	6.115920
H	2.119093	8.639414	-3.683728
H	3.242304	9.735149	-5.693268
C	1.560688	9.568190	-3.487714
H	4.275406	10.023349	-3.316821
H	0.583166	9.486390	-3.981843
C	2.643470	10.639578	-5.503374
H	1.391977	9.651123	-2.405483
C	3.656814	10.926425	-3.203841
H	1.711297	10.554689	-6.078785
C	2.362878	10.758961	-4.006076
H	3.441020	11.062285	-2.134702
H	3.204451	11.509641	-5.872455
H	-1.440996	8.900412	-0.231149
H	4.238361	11.790000	-3.555892
H	-3.647553	9.446128	-1.385657
C	-1.515087	9.838035	0.337361
H	-0.413593	11.243692	-4.748043
H	-2.005941	9.622824	1.298355
C	-3.715288	10.368008	-0.791165
H	-4.279019	10.146849	0.128565
H	-0.499113	10.202248	0.545470
S	1.437279	12.362999	-3.705885
C	-0.104760	12.299203	-4.690443

S	-1.354754	11.148589	-2.005632
C	-2.327302	10.890461	-0.423154
H	0.172273	12.619243	-5.704103
H	-4.283579	11.113146	-1.364715
Fe	0.514904	12.377832	-1.629974
H	-3.025683	11.713423	-3.591807
C	-2.301942	12.327813	-3.036490
B	-1.345856	13.223296	-4.059913
C	-3.642949	13.671609	-5.377901
C	-4.383288	14.202721	-6.441840
C	-2.238143	13.756806	-5.320807
H	-4.187445	13.179091	-4.568585
C	-2.408271	12.187434	0.377610
H	-5.472059	14.110377	-6.446155
C	-3.734005	14.850525	-7.495975
C	-1.616186	14.423245	-6.398846
C	-2.339137	14.961658	-7.466800
H	-2.991979	12.013507	1.295158
H	-4.305534	15.264900	-8.328678
H	-0.527432	14.530917	-6.403452
H	-1.406031	12.534112	0.663195
H	-1.814996	15.469295	-8.280099
H	-2.869795	12.977866	-2.352536
H	-2.899631	12.989510	-0.189740
C	-0.807333	14.634015	-3.336660
H	0.080145	15.017903	-3.863088
H	2.639283	14.472437	-1.742337
S	-0.362889	14.465361	-1.565839
C	2.227018	15.457637	-1.998514
H	2.040236	15.480202	-3.080678
H	-1.582489	15.412624	-3.378269
H	1.584104	14.559166	0.525821
C	0.960986	15.743589	-1.195583
H	2.984775	16.223042	-1.768636
C	1.224480	15.573625	0.303368
H	0.316949	15.760276	0.894440
C	0.394151	17.126774	-1.511941
H	0.172398	17.231878	-2.582957
H	1.997906	16.288943	0.621242
H	-0.526769	17.317792	-0.943395
H	1.134035	17.896496	-1.242041
N	2.256009	11.596084	0.620160
N	1.566056	11.892298	-0.265654

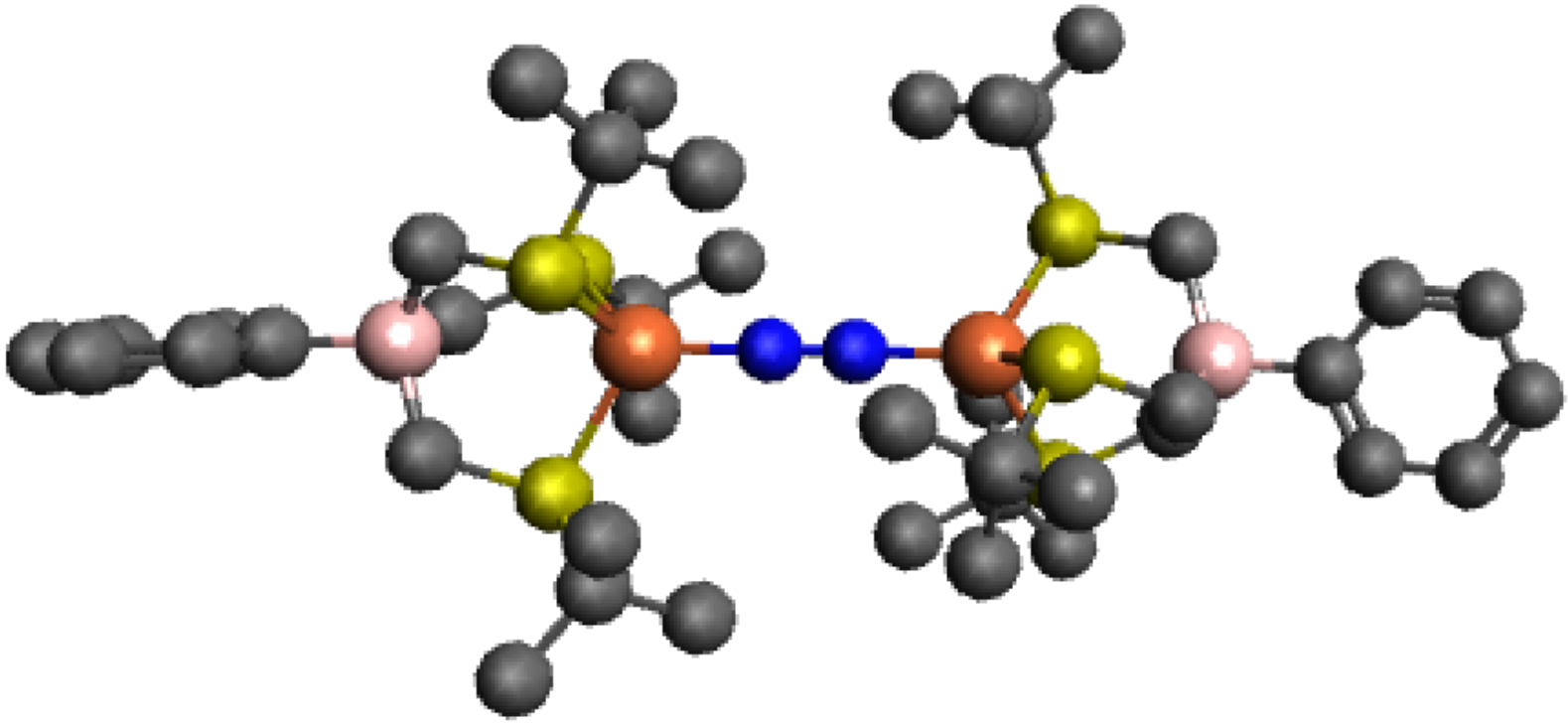


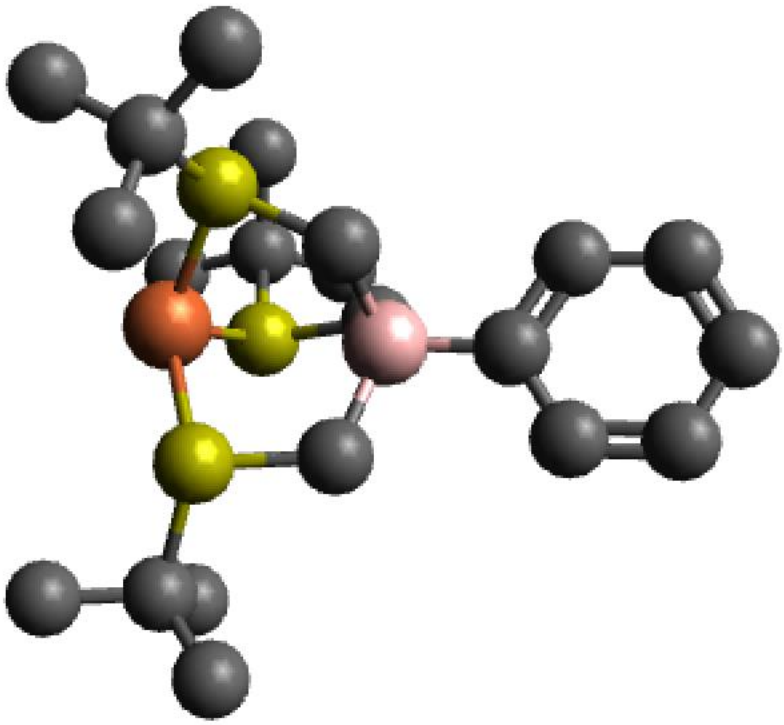
Figure SI15. Optimized structure of $[Fe]_2(\mu\text{-}N_2)$ where H atoms are left out for clarity.

[Fe]

CARTESIAN COORDINATES (ANGSTROEM)

Fe	3.137798	11.128405	1.926946
S	3.131764	9.006134	2.507127
S	5.205337	11.698438	1.434553
B	4.983235	10.643063	4.268220
C	3.920629	11.787441	4.801047
C	4.256744	9.184704	3.971675
H	3.638009	8.877705	4.828831
H	5.010269	8.394822	3.839831
C	3.985942	7.746084	1.387236
C	3.904679	6.383892	2.076953
H	4.358584	5.614771	1.432037
H	2.861166	6.100396	2.272830
H	4.450258	6.384941	3.031263
C	5.430244	8.139906	1.099289
H	5.877623	7.401577	0.415055
H	6.039277	8.157751	2.012903
C	3.150771	7.771627	0.106305
H	3.543425	7.028471	-0.604503
H	3.195595	8.761902	-0.370719
C	5.963836	11.186252	3.047597
H	6.715641	10.434626	2.766081
H	6.523118	12.068773	3.393455
S	2.535644	12.301586	3.687917
C	5.637761	13.528093	1.245092
H	4.428035	12.730650	5.055368
H	3.424632	11.444148	5.721777
H	5.486138	9.129633	0.626487
H	2.097509	7.529103	0.309559
C	0.957778	11.835865	4.616625
C	5.218050	14.335088	2.468422
C	4.846257	13.962752	0.010575
C	7.144231	13.623632	1.001474
C	0.962145	10.368855	5.030750
C	-0.152459	12.109815	3.600840
C	0.842296	12.759952	5.829226
H	4.139541	14.243493	2.654627
H	5.754030	14.016656	3.372603
H	5.452942	15.397922	2.298791
H	3.764169	13.870447	0.186103
H	5.110597	13.357645	-0.868876
H	5.069240	15.016267	-0.217662
H	7.439244	13.047971	0.113024

H	7.713585	13.249368	1.864047
H	7.427610	14.676324	0.843142
H	1.771423	10.146023	5.739006
H	1.073118	9.710282	4.158675
H	0.008758	10.129696	5.528077
H	-1.131451	11.909347	4.062265
H	-0.046841	11.458813	2.720280
H	-0.138955	13.156899	3.264862
H	-0.092126	12.544305	6.371267
H	1.676899	12.609494	6.528626
H	0.830112	13.815423	5.523405
C	6.036306	10.321723	5.488585
C	6.007920	10.958026	6.745653
C	6.930678	10.661967	7.757389
C	7.928363	9.707670	7.543150
C	7.987959	9.056917	6.305411
C	7.058433	9.363736	5.308296
H	5.245212	11.711869	6.953074
H	6.868575	11.180099	8.717496
H	8.649630	9.472750	8.328199
H	8.761383	8.307962	6.118844
H	7.134264	8.836069	4.352897



SI16. Optimized structure of [Fe] where H atoms were not pictured for clarity.

[Fe](N_2)

CARTESIAN COORDINATES (ANGSTROEM)

C	-5.962426	-0.087157	-0.009029
C	-5.306820	1.145088	-0.072427
C	-3.907159	1.200221	-0.063280
C	-3.108522	0.042049	0.003576
C	-5.199369	-1.257933	0.067005
C	-3.803820	-1.184601	0.072070
H	-5.886863	2.069330	-0.128791
H	-3.431784	2.183099	-0.106958
H	-7.052927	-0.136143	-0.016663
H	-5.695065	-2.230077	0.121776
H	-3.236219	-2.117685	0.136831
B	-1.476406	0.066559	0.095464
C	-0.929624	1.608523	-0.168755
S	0.813274	1.697922	-0.717252
H	-1.015211	2.219912	0.743035
H	-1.504033	2.107666	-0.962528
C	1.455365	3.369120	-0.159385
C	0.548235	4.448984	-0.748760
H	0.499044	4.372831	-1.843995
H	0.949730	5.440551	-0.488376
H	-0.471367	4.381251	-0.346137
C	2.865211	3.445016	-0.749914
H	3.326690	4.402407	-0.464635
H	2.842458	3.385558	-1.847073
H	3.499120	2.633094	-0.367141
C	1.497561	3.440883	1.364869
H	2.177250	2.682388	1.776067
H	0.504122	3.289307	1.808411
H	1.856914	4.435381	1.672482
C	-0.907865	-0.983805	-1.065031
S	0.687375	-1.780629	-0.652553
H	-0.768221	-0.449399	-2.017582
H	-1.608827	-1.809511	-1.246972
C	1.500522	-2.221425	-2.284480
C	0.538736	-3.112744	-3.069883
H	0.263811	-4.006713	-2.492780
H	1.026238	-3.438923	-4.001648
H	-0.379278	-2.573542	-3.339871
C	2.762553	-2.986656	-1.879641
H	2.514419	-3.894129	-1.311522
H	3.426312	-2.359760	-1.268048
H	3.312446	-3.284766	-2.785065

C	1.855568	-0.956487	-3.061920
H	0.968006	-0.346331	-3.277536
H	2.314149	-1.237117	-4.023021
H	2.570673	-0.337955	-2.502806
C	-1.164210	-0.448684	1.651434
S	0.470709	0.033693	2.324426
H	-1.228263	-1.546132	1.707962
H	-1.903384	-0.034224	2.351595
C	0.934776	-1.276988	3.584961
C	2.241339	-0.764437	4.194907
H	3.023156	-0.664303	3.429133
H	2.594118	-1.481108	4.951916
H	2.099548	0.210574	4.681791
C	1.144366	-2.624172	2.898495
H	1.966304	-2.572399	2.171703
H	0.240689	-2.959644	2.371738
H	1.395402	-3.383287	3.655788
C	-0.177544	-1.345518	4.630933
H	-0.342451	-0.365846	5.100804
H	0.107520	-2.062556	5.416340
H	-1.123737	-1.686543	4.189542
Fe	1.703047	-0.053807	0.422387
N	3.501779	-0.113615	0.590153
N	4.632351	-0.154017	0.697270

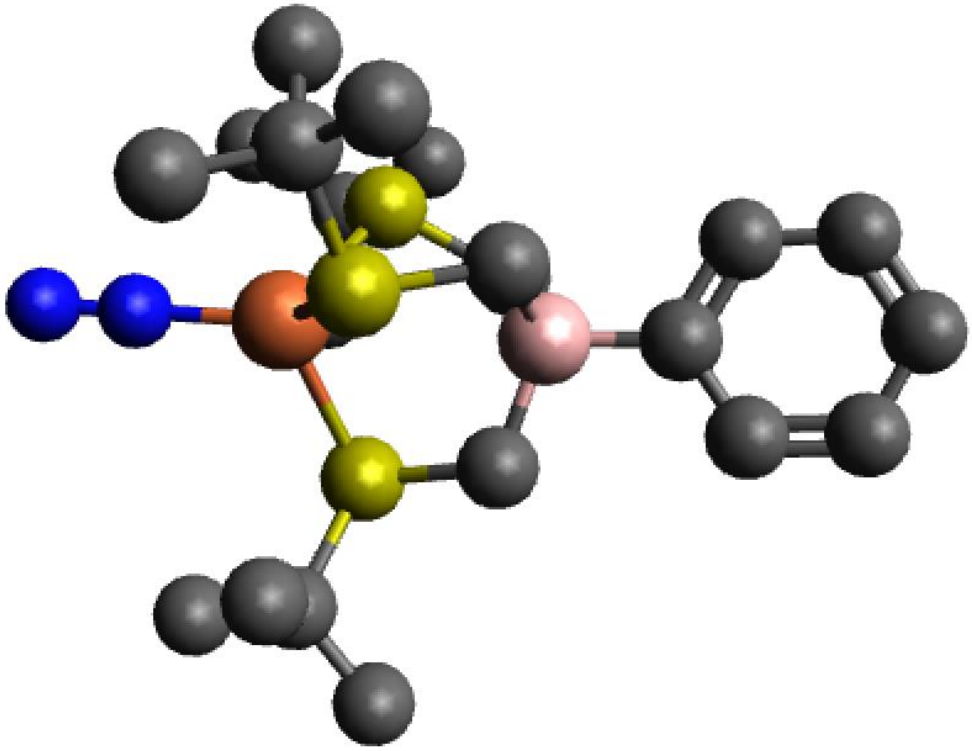


Figure SI17. Optimized structure of [Fe](N_2) where H atoms are not pictured for clarity.

[Fe](N)

CARTESIAN COORDINATES (ANGSTROEM)

C	-6.001656	0.005832	0.027544
C	-5.313991	1.222127	0.024557
C	-3.913478	1.239667	0.015919
C	-3.147340	0.058231	0.005107
C	-5.270424	-1.187460	0.025087
C	-3.873489	-1.151983	0.013489
H	-5.869371	2.162959	0.029571
H	-3.411740	2.210386	0.021486
H	-7.093050	-0.013689	0.033533
H	-5.792001	-2.147404	0.031429
H	-3.330394	-2.101747	0.016405
B	-1.517960	0.033066	0.070525
C	-0.925657	1.567362	-0.126715
S	0.807218	1.628812	-0.687208
H	-0.985298	2.152152	0.804231
H	-1.488143	2.106014	-0.902948
C	1.496396	3.290310	-0.158320
C	0.599396	4.372173	-0.761407
H	0.535564	4.276090	-1.854183
H	1.027436	5.358551	-0.525651
H	-0.415452	4.335843	-0.343257
C	2.902027	3.319667	-0.760854
H	3.389328	4.266832	-0.484290
H	2.868646	3.254891	-1.857649
H	3.503353	2.486654	-0.367030
C	1.549940	3.383607	1.363959
H	2.216985	2.614492	1.774874
H	0.555566	3.268245	1.816467
H	1.935062	4.375805	1.646065
C	-0.978578	-0.959052	-1.149389
S	0.574593	-1.800216	-0.711151
H	-0.804296	-0.384006	-2.071645
H	-1.693966	-1.760179	-1.378793
C	1.454659	-2.254684	-2.302217
C	0.517036	-3.150658	-3.111169
H	0.221807	-4.039949	-2.537283
H	1.036005	-3.483191	-4.023083
H	-0.389837	-2.610968	-3.415839
C	2.698774	-3.017134	-1.840301
H	2.429001	-3.918158	-1.272175
H	3.341746	-2.383053	-1.211393
H	3.281453	-3.324039	-2.721672

C	1.839767	-0.997685	-3.078660
H	0.962860	-0.384864	-3.326944
H	2.325341	-1.291383	-4.022073
H	2.545315	-0.379385	-2.505865
C	-1.165420	-0.578584	1.586257
S	0.433213	-0.008380	2.241266
H	-1.169880	-1.679334	1.583431
H	-1.912186	-0.240835	2.318820
C	1.010933	-1.254136	3.519120
C	2.258986	-0.611620	4.128483
H	3.007265	-0.402574	3.349512
H	2.695908	-1.304001	4.863990
H	2.010887	0.327799	4.642012
C	1.345972	-2.581280	2.842302
H	2.177046	-2.460585	2.132813
H	0.482866	-2.994655	2.303068
H	1.649238	-3.311538	3.608491
C	-0.103950	-1.414899	4.551870
H	-0.370592	-0.450118	5.005087
H	0.246636	-2.086765	5.350491
H	-1.005777	-1.858130	4.108616
Fe	1.816946	-0.152222	0.372645
N	3.294931	0.329808	0.897061

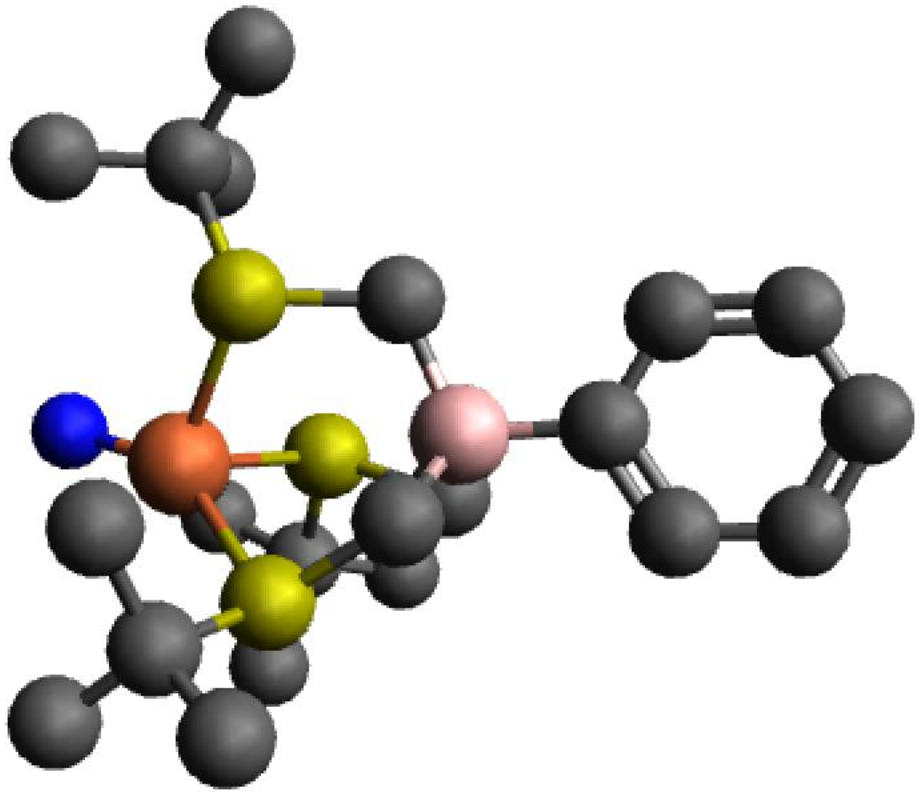


Figure SI18. Optimized structure of [Fe](N) where H atoms are not pictured for clarity.

[Fe](NH)

CARTESIAN COORDINATES (ANGSTROEM)

C	-6.010539	-0.068123	-0.008989
C	-5.350308	1.163240	0.003816
C	-3.950425	1.212364	0.008884
C	-3.157213	0.048946	-0.003626
C	-5.252328	-1.244522	-0.012915
C	-3.856518	-1.177180	-0.010913
H	-5.926859	2.091302	0.010359
H	-3.470686	2.194016	0.026426
H	-7.101246	-0.112260	-0.013945
H	-5.752023	-2.216104	-0.018379
H	-3.292082	-2.114515	-0.009481
B	-1.527333	0.059858	0.081606
C	-0.963257	1.608577	-0.101864
S	0.758607	1.683381	-0.695317
H	-1.012821	2.173104	0.842154
H	-1.547780	2.158147	-0.853627
C	1.472249	3.326800	-0.151921
C	0.578119	4.436745	-0.704161
H	0.493651	4.374063	-1.798022
H	1.017405	5.412707	-0.445883
H	-0.430203	4.395164	-0.270266
C	2.865354	3.353377	-0.785110
H	3.373673	4.285183	-0.494144
H	2.806159	3.319764	-1.882273
H	3.463765	2.500853	-0.429528
C	1.567154	3.382527	1.371227
H	2.237601	2.597812	1.745711
H	0.584186	3.261853	1.846638
H	1.968183	4.363639	1.670260
C	-0.958370	-0.924656	-1.137598
S	0.564600	-1.820447	-0.684946
H	-0.743350	-0.336540	-2.042801
H	-1.683669	-1.703714	-1.407589
C	1.469346	-2.257519	-2.266213
C	0.538234	-3.115394	-3.122094
H	0.215157	-4.014851	-2.579996
H	1.072580	-3.431409	-4.031208
H	-0.352691	-2.552517	-3.432165
C	2.687535	-3.052689	-1.789229
H	2.387132	-3.960941	-1.248576
H	3.322949	-2.442881	-1.129138
H	3.288710	-3.350793	-2.661255

C	1.900529	-0.990555	-3.002089
H	1.043564	-0.352572	-3.256292
H	2.404319	-1.270679	-3.940178
H	2.604097	-0.401620	-2.395738
C	-1.190586	-0.543656	1.604453
S	0.395587	0.040426	2.288036
H	-1.177621	-1.644492	1.600198
H	-1.957230	-0.220746	2.322594
C	0.967987	-1.209438	3.560108
C	2.217833	-0.570602	4.171117
H	2.967610	-0.366026	3.391891
H	2.653264	-1.263199	4.907427
H	1.973076	0.370297	4.683717
C	1.309399	-2.533430	2.878824
H	2.141922	-2.404957	2.171888
H	0.449560	-2.947166	2.334621
H	1.613726	-3.267390	3.641281
C	-0.142721	-1.380942	4.595546
H	-0.405591	-0.420342	5.059837
H	0.205203	-2.064129	5.385831
H	-1.047505	-1.814420	4.148126
Fe	1.816911	-0.127875	0.392978
N	3.500378	0.158040	0.796356
H	4.403133	-0.300253	0.618962

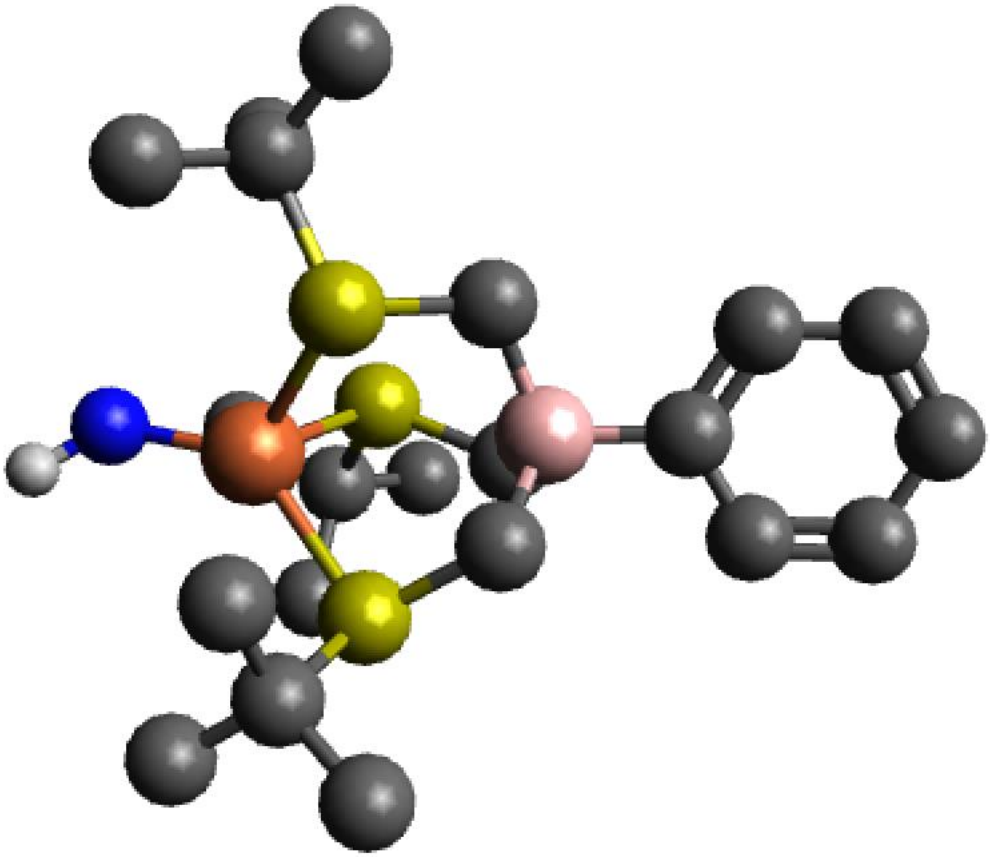


Figure SI19. Optimized structure of [Fe](NH) where H atoms are not pictured for clarity.

[Fe](NH$_2$)

CARTESIAN COORDINATES (ANGSTROEM)

C	-5.996180	-0.131702	-0.049399
C	-5.355820	1.110076	-0.069462
C	-3.956959	1.182245	-0.047940
C	-3.143964	0.032756	-0.011327
C	-5.218680	-1.294708	-0.004195
C	-3.824226	-1.204038	0.013363
H	-5.947322	2.028253	-0.101712
H	-3.493334	2.171813	-0.057633
H	-7.085906	-0.194095	-0.067031
H	-5.702473	-2.274211	0.016326
H	-3.244445	-2.131120	0.052414
B	-1.513841	0.073043	0.093281
C	-0.978922	1.623746	-0.137562
S	0.761759	1.730284	-0.692989
H	-1.062183	2.221678	0.783302
H	-1.558191	2.134503	-0.920249
C	1.386977	3.407200	-0.131534
C	0.490658	4.482927	-0.744153
H	0.461802	4.399476	-1.839657
H	0.884629	5.477383	-0.482755
H	-0.536489	4.415326	-0.360568
C	2.808843	3.476157	-0.694562
H	3.267718	4.434757	-0.408732
H	2.808769	3.406138	-1.791549
H	3.429140	2.663651	-0.288111
C	1.406282	3.493152	1.392639
H	2.083787	2.741120	1.819659
H	0.407506	3.342972	1.824465
H	1.758624	4.491505	1.696150
C	-0.911118	-0.940228	-1.079271
S	0.645932	-1.771822	-0.613245
H	-0.719837	-0.374352	-2.004394
H	-1.616486	-1.745956	-1.324354
C	1.502379	-2.205309	-2.221106
C	0.555483	-3.066119	-3.057635
H	0.240054	-3.961512	-2.503699
H	1.072064	-3.390457	-3.974636
H	-0.340849	-2.504930	-3.354978
C	2.736225	-3.001066	-1.785752
H	2.449671	-3.917346	-1.250111
H	3.376741	-2.393402	-1.129617
H	3.318813	-3.287797	-2.674658

C	1.912408	-0.939201	-2.970238
H	1.046296	-0.306659	-3.207533
H	2.399647	-1.216629	-3.918339
H	2.620110	-0.344808	-2.376578
C	-1.198778	-0.477818	1.640036
S	0.388280	0.106584	2.329913
H	-1.188186	-1.578392	1.661222
H	-1.970736	-0.135487	2.343672
C	0.954617	-1.155138	3.592520
C	2.224080	-0.542927	4.190656
H	2.985555	-0.373963	3.415266
H	2.641769	-1.232810	4.939503
H	2.009907	0.415153	4.684586
C	1.257502	-2.489651	2.914680
H	2.068261	-2.387103	2.180262
H	0.377088	-2.892354	2.396155
H	1.568270	-3.221907	3.676238
C	-0.143466	-1.300944	4.645737
H	-0.374090	-0.334782	5.115666
H	0.195970	-1.995973	5.429394
H	-1.065791	-1.707860	4.209293
Fe	1.799952	0.016625	0.466467
N	3.660863	-0.203857	0.527586
H	4.104405	-0.957052	1.057861
H	4.264080	0.010445	-0.269271

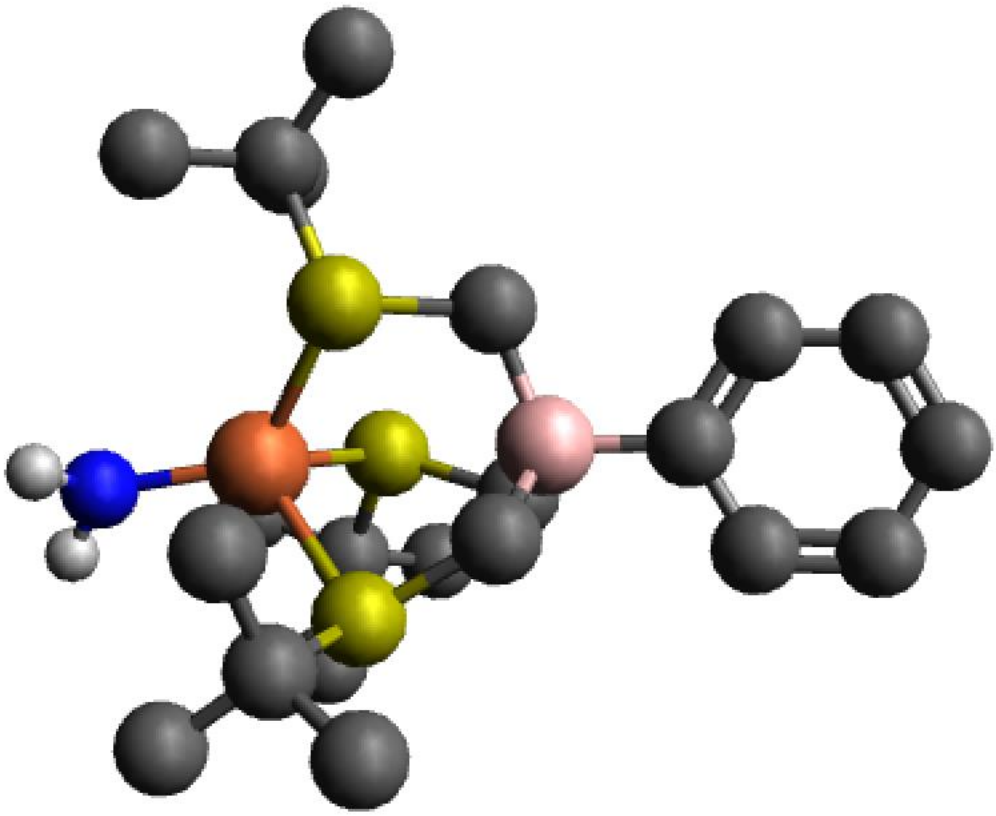


Figure SI20. Optimized structure of [Fe](NH_2) where H atoms are not pictured for clarity.

[Fe](NH3)

CARTESIAN COORDINATES (ANGSTROEM)

H	1.111661	-1.011555	-4.339075
H	3.173513	-2.374902	-3.644975
H	0.857863	-3.531736	-3.931720
H	-0.392446	-3.266871	-2.695894
C	1.393700	-2.033713	-2.459160
C	0.692571	-3.331607	-2.860589
C	0.882193	-0.849982	-3.273366
H	-0.205879	-0.731736	-3.181897
H	3.378471	3.516093	-0.881146
H	3.060194	4.672121	0.438954
C	2.911550	-2.161458	-2.597108
H	3.293811	2.942666	0.807020
H	-1.086708	-1.522961	-1.463160
C	2.864076	3.649268	0.082041
S	1.130837	-1.773088	-0.604321
C	-0.675866	-1.403547	-0.448896
H	3.407812	-1.225637	-2.301435
H	1.258496	4.319200	-2.044410
H	3.303658	-2.976665	-1.971219
H	1.079902	-4.184312	-2.285071
H	0.849566	5.433271	-0.710824
H	1.358971	0.086043	-2.951015
H	-1.081416	-2.235703	0.145746
C	0.750104	4.397087	-1.072923
C	1.356825	3.429809	-0.056408
H	-0.855426	1.209848	-1.812560
S	1.222812	1.677834	-0.755058
Fe	2.070281	-0.023459	0.389908
C	-0.590283	1.325034	-0.749888
H	-0.320231	4.197552	-1.224758
B	-1.127338	0.064427	0.161820
H	-3.048850	1.720264	-1.242868
C	-3.544522	0.936156	-0.665797
C	0.673597	3.548040	1.301805
C	-2.768731	0.034097	0.090320
C	-4.943358	0.870465	-0.711368
H	-1.060636	2.262655	-0.415664
H	-5.502268	1.593583	-1.310691
H	-0.410067	3.384883	1.228445
H	0.831989	4.561195	1.705472
C	-3.493668	-0.948261	0.801421
C	-5.625702	-0.115319	0.006348

H	1.085029	2.819916	2.014670
C	-4.888215	-1.030472	0.766789
H	-2.948691	-1.679278	1.406183
H	-6.715575	-0.171071	-0.024836
S	0.976717	0.096916	2.318234
H	-5.403813	-1.808919	1.334650
C	-0.782607	0.260196	1.765076
H	0.871034	-2.868744	1.858089
H	-1.094069	1.256983	2.114533
H	-1.340510	-0.463546	2.376982
C	0.344322	-2.625438	2.791216
C	0.961116	-1.405369	3.467074
H	2.982801	-1.916375	2.846183
H	0.411706	-3.492640	3.467885
H	-0.717081	-2.468695	2.556920
C	2.440868	-1.650766	3.765575
H	2.914121	-0.759644	4.203835
C	0.198076	-1.026320	4.736467
H	2.543851	-2.482210	4.479948
H	0.203427	-1.871296	5.444069
H	-0.850844	-0.782823	4.514748
H	0.658130	-0.158367	5.229683
N	4.153790	-0.076692	0.541404
H	4.621330	0.149576	-0.341057
H	4.501876	-0.998002	0.821882
H	4.512173	0.587610	1.233453

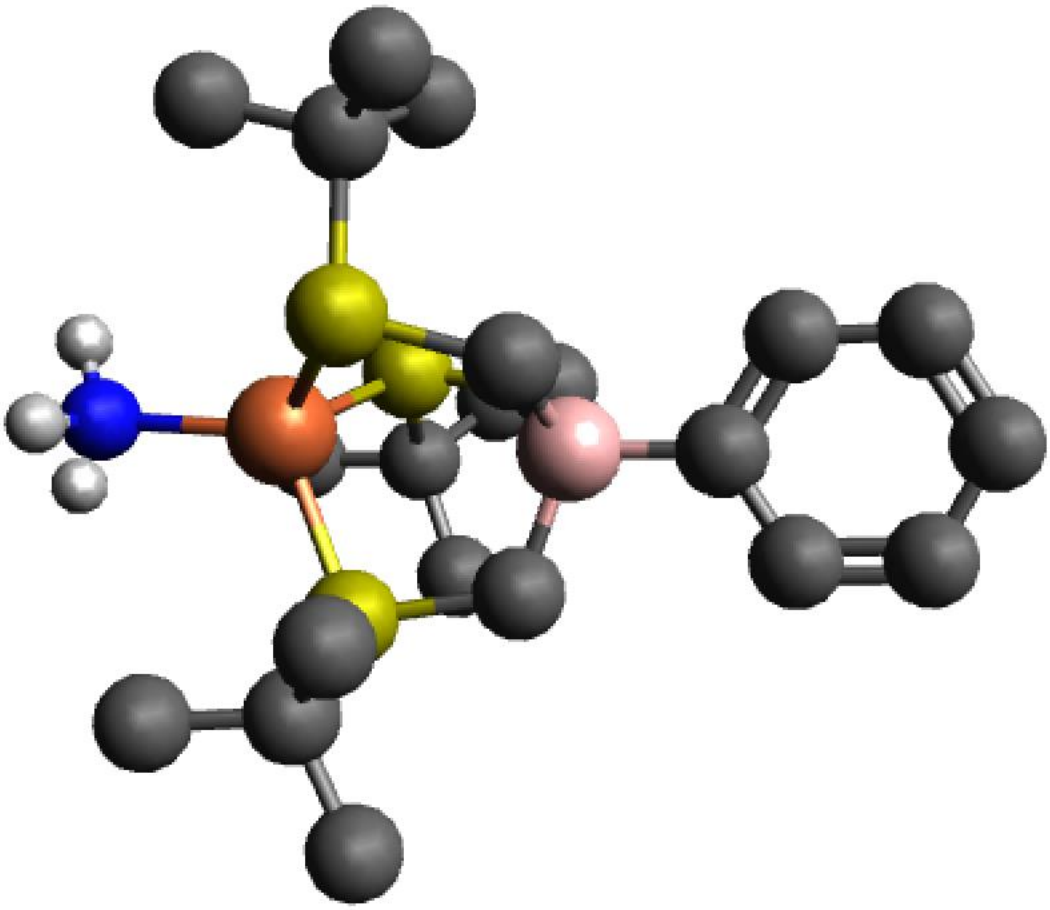


Figure SI21. Optimized structure of [Fe](NH_3) where H atoms are not pictured for clarity.

SELECT ORBITAL ENERGIES OF CASSCF OUTPUT FILES

All CASSCF jobs used BP86 ZORA CPCM(THF) def2-TZVP optimized coordinates for input file coordinates.

$[Fe]_2(\mu_2\text{-}N_2)$

ORBITAL ENERGIES

NO	OCC	E(Eh)	E(eV)
130	2.0000	-0.410618	-11.1735
131	2.0000	-0.404283	-11.0011
132	2.0000	-0.398314	-10.8387
133	2.0000	-0.389091	-10.5877
134	2.0000	-0.384897	-10.4736
135	2.0000	-0.370206	-10.0738
136	2.0000	-0.365561	-9.9474
137	2.0000	-0.359981	-9.7956
138	2.0000	-0.356975	-9.7138
139	2.0000	-0.348208	-9.4752
140	2.0000	-0.343622	-9.3504
141	2.0000	-0.333660	-9.0793
142	2.0000	-0.328108	-8.9283
143	1.0000	-0.014389	-0.3916
144	1.0000	-0.012115	-0.3297
145	1.0000	-0.031099	-0.8463
146	1.0000	-0.031870	-0.8672
147	1.0000	-0.011867	-0.3229
148	1.0000	-0.015109	-0.4111
149	0.0000	0.084295	2.2938
150	0.0000	0.098610	2.6833
151	0.0000	0.101829	2.7709
152	0.0000	0.102326	2.7844
153	0.0000	0.104421	2.8414
154	0.0000	0.136944	3.7264
155	0.0000	0.139058	3.7840
156	0.0000	0.140504	3.8233
157	0.0000	0.154451	4.2028
158	0.0000	0.158963	4.3256
159	0.0000	0.165130	4.4934
160	0.0000	0.168012	4.5718

[Fe]

ORBITAL ENERGIES

NO	OCC	E(Eh)	E(eV)
110	2.0000	-0.412364	-11.2210
111	2.0000	-0.399595	-10.8735
112	2.0000	-0.372478	-10.1356
113	2.0000	-0.349519	-9.5109
114	2.0000	-0.344100	-9.3634
115	2.0000	-0.305500	-8.3131
116	1.9849	-0.411762	-11.2046
117	1.9734	-0.383058	-10.4235
118	1.9723	-0.380935	-10.3658
119	1.9445	-0.287779	-7.8309
120	0.9916	-0.022004	-0.5988
121	0.9914	-0.036524	-0.9939
122	0.9910	-0.021922	-0.5965
123	0.0421	0.209713	5.7066
124	0.0292	0.807026	21.9603
125	0.0261	1.017136	27.6777
126	0.0143	0.757321	20.6077
127	0.0134	0.207181	5.6377
128	0.0094	1.308913	35.6173
129	0.0089	1.360558	37.0227
130	0.0075	1.326677	36.1007
131	0.0000	0.081122	2.2074
132	0.0000	0.088623	2.4116
133	0.0000	0.089874	2.4456
134	0.0000	0.095796	2.6067
135	0.0000	0.122470	3.3326
136	0.0000	0.123089	3.3494
137	0.0000	0.128007	3.4832
138	0.0000	0.130037	3.5385
139	0.0000	0.131564	3.5800
140	0.0000	0.139776	3.8035

[Fe](N_2)

ORBITAL ENERGIES

NO	OCC	E(Eh)	E(eV)
110	2.0000	-0.470729	-12.8092
111	2.0000	-0.466406	-12.6915
112	2.0000	-0.458812	-12.4849
113	2.0000	-0.449593	-12.2340
114	2.0000	-0.444243	-12.0885
115	2.0000	-0.438189	-11.9237
116	2.0000	-0.405717	-11.0401
117	2.0000	-0.401019	-10.9123
118	2.0000	-0.384224	-10.4553
119	2.0000	-0.372289	-10.1305
120	2.0000	-0.362769	-9.8715
121	2.0000	-0.307818	-8.3762
122	2.0000	-0.292685	-7.9644
123	1.9788	-0.416144	-11.3239
124	1.9554	-0.554694	-15.0940
125	1.9019	-0.412623	-11.2281
126	1.8494	-0.341620	-9.2960
127	0.9984	-0.103149	-2.8068
128	0.9961	-0.102584	-2.7915
129	0.9916	-0.078286	-2.1303
130	0.1413	0.191768	5.2183
131	0.1215	0.211326	5.7505
132	0.0202	0.466891	12.7048
133	0.0163	1.173170	31.9236
134	0.0088	1.248911	33.9846
135	0.0079	1.407361	38.2962
136	0.0067	1.551105	42.2077
137	0.0057	1.694852	46.1193
138	0.0000	0.074895	2.0380
139	0.0000	0.086886	2.3643
140	0.0000	0.088577	2.4103
141	0.0000	0.106884	2.9085
142	0.0000	0.115351	3.1389
143	0.0000	0.115771	3.1503
144	0.0000	0.125769	3.4224
145	0.0000	0.126010	3.4289
146	0.0000	0.130216	3.5434
147	0.0000	0.136933	3.7261
148	0.0000	0.140518	3.8237
149	0.0000	0.146517	3.9869

150	0.0000	0.148172	4.0320

[Fe](N)

ORBITAL ENERGIES

NO	OCC	E(Eh)	E(eV)
110	2.0000	-0.446212	-12.1420
111	2.0000	-0.426672	-11.6103
112	2.0000	-0.406944	-11.0735
113	2.0000	-0.398366	-10.8401
114	2.0000	-0.388643	-10.5755
115	2.0000	-0.379291	-10.3210
116	2.0000	-0.362949	-9.8763
117	2.0000	-0.361593	-9.8395
118	2.0000	-0.305534	-8.3140
119	1.9533	-0.336586	-9.1590
120	1.9174	-0.453281	-12.3344
121	1.9166	-0.447812	-12.1856
122	1.7926	-0.368137	-10.0175
123	1.0637	-0.137153	-3.7321
124	1.0620	-0.130594	-3.5536
125	0.9988	-0.162858	-4.4316
126	0.9977	-0.141466	-3.8495
127	0.2088	0.176481	4.8023
128	0.0401	0.254753	6.9322
129	0.0153	1.196513	32.5588
130	0.0131	1.286583	35.0097
131	0.0074	0.680332	18.5128
132	0.0069	1.239763	33.7357
133	0.0063	1.294539	35.2262
134	0.0000	0.075491	2.0542
135	0.0000	0.088709	2.4139
136	0.0000	0.089675	2.4402
137	0.0000	0.107254	2.9185
138	0.0000	0.114069	3.1040
139	0.0000	0.118150	3.2150
140	0.0000	0.125974	3.4279

[Fe](NH)

ORBITAL ENERGIES

NO	OCC	E(Eh)	E(eV)
110	2.0000	-0.451577	-12.2880
111	2.0000	-0.445057	-12.1106
112	2.0000	-0.439234	-11.9522
113	2.0000	-0.405527	-11.0349
114	2.0000	-0.403883	-10.9902
115	2.0000	-0.388096	-10.5606
116	2.0000	-0.377156	-10.2629
117	2.0000	-0.363808	-9.8997
118	2.0000	-0.358063	-9.7434
119	1.9402	-0.300434	-8.1752
120	1.9375	-0.291093	-7.9210
121	1.8983	-0.399202	-10.8628
122	1.8931	-0.435559	-11.8522
123	1.0905	-0.152892	-4.1604
124	1.0904	-0.146454	-3.9852
125	1.0001	-0.168038	-4.5725
126	1.0001	-0.151874	-4.1327
127	0.9969	-0.115483	-3.1425
128	0.0619	0.187338	5.0977
129	0.0604	0.192173	5.2293
130	0.0162	1.010790	27.5050
131	0.0143	0.628060	17.0904
132	0.0000	0.075409	2.0520
133	0.0000	0.089191	2.4270
134	0.0000	0.090731	2.4689
135	0.0000	0.107468	2.9243
136	0.0000	0.113525	3.0892
137	0.0000	0.118749	3.2313
138	0.0000	0.125740	3.4216
139	0.0000	0.128449	3.4953
140	0.0000	0.132903	3.6165

[Fe](NH$_2$)

ORBITAL ENERGIES

NO	OCC	E(Eh)	E(eV)
110	2.0000	-0.456783	-12.4297
111	2.0000	-0.429792	-11.6952
112	2.0000	-0.421175	-11.4608
113	2.0000	-0.403570	-10.9817
114	2.0000	-0.397314	-10.8115
115	2.0000	-0.388051	-10.5594
116	2.0000	-0.376658	-10.2494
117	2.0000	-0.360317	-9.8047
118	2.0000	-0.306862	-8.3502
119	2.0000	-0.291524	-7.9328
120	1.9982	-0.630399	-17.1540
121	1.9979	-0.477699	-12.9988
122	1.9970	-0.535750	-14.5785
123	1.9834	-0.485566	-13.2129
124	1.0008	-0.127554	-3.4709
125	0.9968	-0.108636	-2.9561
126	0.9967	-0.109224	-2.9721
127	0.9955	-0.092935	-2.5289
128	0.0160	1.346864	36.6500
129	0.0065	1.382610	37.6227
130	0.0056	1.646234	44.7963
131	0.0055	1.603123	43.6232
132	0.0000	0.075502	2.0545
133	0.0000	0.087761	2.3881
134	0.0000	0.089569	2.4373
135	0.0000	0.107003	2.9117
136	0.0000	0.114442	3.1141
137	0.0000	0.116186	3.1616
138	0.0000	0.126456	3.4410
139	0.0000	0.126831	3.4512
140	0.0000	0.130749	3.5579

[Fe](NH$_3$)

ORBITAL ENERGIES

NO	OCC	E(Eh)	E(eV)
110	2.0000	-0.442505	-12.0412
111	2.0000	-0.432913	-11.7802
112	2.0000	-0.428553	-11.6615
113	2.0000	-0.421995	-11.4831
114	2.0000	-0.402900	-10.9635
115	2.0000	-0.400488	-10.8978
116	2.0000	-0.387819	-10.5531
117	2.0000	-0.378285	-10.2937
118	2.0000	-0.336577	-9.1587
119	2.0000	-0.329635	-8.9698
120	2.0000	-0.296997	-8.0817
121	1.9974	-0.408861	-11.1257
122	1.9695	-0.351125	-9.5546
123	1.9693	-0.350396	-9.5348
124	1.9599	-0.313710	-8.5365
125	0.9926	0.003373	0.0918
126	0.9909	0.003685	0.1003
127	0.9907	-0.001916	-0.0521
128	0.0401	0.263269	7.1639
129	0.0299	0.929546	25.2942
130	0.0297	0.937311	25.5055
131	0.0105	1.245005	33.8783
132	0.0098	1.287158	35.0254
133	0.0098	1.282863	34.9085
134	0.0000	0.070574	1.9204
135	0.0000	0.088731	2.4145
136	0.0000	0.089621	2.4387
137	0.0000	0.099325	2.7028
138	0.0000	0.121545	3.3074
139	0.0000	0.125234	3.4078
140	0.0000	0.126591	3.4447