

## Investigating the Influence of Oriented External Electric Fields on Modulating Spin-Transition Temperatures in Fe(II) SCO Complexes: A Theoretical Perspective

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**Table S1:** DFT optimised geometrical parameters of **1** in the presence of an Oriented External Electric Fields

High spin								
$F_z$ (V/Å)	Fe-N1	Fe-N2	Fe-N3	Fe-N4	Fe-N5	Fe-N6	Average Fe-N	$\angle$ SCN-Fe-NCS
<b>1</b> <sub>xray</sub>	2.060	2.059	2.269	2.269	2.198	2.197	2.175	98.114
<b>1</b> <sub>opt</sub>	2.064	2.064	2.261	2.261	2.199	2.199	2.175	97.353
0.1285 <b>1</b> <sub>opt</sub>	2.096	2.096	2.255	2.255	2.193	2.192	2.181	90.379
0.2570 <b>1</b> <sub>opt</sub>	2.134	2.134	2.249	2.249	2.196	2.196	2.193	86.145
0.3855 <b>1</b> <sub>opt</sub>	2.186	2.186	2.239	2.239	2.197	2.197	2.207	81.872
0.5140 <b>1</b> <sub>opt</sub>	2.264	2.266	2.225	2.225	2.194	2.193	2.228	77.767
0.6425 <b>1</b> <sub>opt</sub>	2.429	2.470	2.206	2.207	2.163	2.162	2.272	71.636

**Note:** Here, <sup>N</sup>**M**<sub>opt</sub> denoted the optimised geometry at N V/Å applied OEEF of molecule **M**.

**Table S2:** Geometrical parameters of **1** in the presence of an Oriented External Electric Field

Low spin								
$F_z$ (V/Å)	Fe-N1	Fe-N2	Fe-N3	Fe-N4	Fe-N5	Fe-N6	Average Fe-N	$\angle$ SCN-Fe-NCS
<b>1</b> <sub>xray</sub>	1.93	1.93	1.979	1.979	1.976	1.976	1.962	90.734
<b>1</b> <sub>opt</sub>	1.93	1.93	1.98	1.98	1.976	1.976	1.962	90.833
0.1285 <b>1</b> <sub>opt</sub>	1.937	1.937	1.988	1.988	1.977	1.977	1.967	88.668
0.2570 <b>1</b> <sub>opt</sub>	1.945	1.945	1.995	1.995	1.980	1.980	1.973	86.542
0.3855 <b>1</b> <sub>opt</sub>	1.955	1.955	2.003	2.003	1.985	1.985	1.981	84.851
0.5140 <b>1</b> <sub>opt</sub>	1.966	1.966	2.011	2.011	1.992	1.992	1.990	83.137
0.6425 <b>1</b> <sub>opt</sub>	1.978	1.978	2.018	2.018	1.999	1.999	1.998	81.154

**Table S3:** Geometrical parameters of **2** in the presence of an Oriented External Electric Field

High spin								
$F_z$ (V/Å)	Fe-N1	Fe-N2	Fe-N3	Fe-N4	Fe-N5	Fe-N6	Average Fe-N	$\angle$ SCN-Fe-NCS
<b>2</b> <sub>xray</sub>	2.021	2.02	2.246	2.243	2.178	2.172	2.147	98.395
<b>2</b> <sub>opt</sub>	2.051	2.051	2.249	2.249	2.192	2.192	2.164	97.888
0.1285 <b>2</b> <sub>opt</sub>	2.085	2.085	2.242	2.242	2.187	2.187	2.171	91.319
0.2570 <b>2</b> <sub>opt</sub>	2.128	2.128	2.235	2.235	2.182	2.182	2.181	86.683
0.3855 <b>2</b> <sub>opt</sub>	2.184	2.184	2.227	2.227	2.176	2.176	2.196	82.488
0.5140 <b>2</b> <sub>opt</sub>	2.269	2.269	2.212	2.212	2.171	2.171	2.217	78.063
0.6425 <b>2</b> <sub>opt</sub>	2.502	2.501	2.177	2.176	2.149	2.149	2.276	70.529

**Table S4:** Geometrical parameters of **2** in the presence of an Oriented External Electric Field

Low spin								
$F_z$ (V/Å)	Fe-N1	Fe-N2	Fe-N3	Fe-N4	Fe-N5	Fe-N6	Average Fe-N	$\angle$ SCN-Fe-NCS
$\mathbf{2}_{\text{xray}}$	1.913	1.913	1.944	1.944	1.942	1.943	1.933	89.561
$\mathbf{2}_{\text{opt}}$	1.929	1.929	1.962	1.962	1.957	1.957	1.949	90.256
0.1285 $\mathbf{2}_{\text{opt}}$	1.937	1.937	1.966	1.966	1.960	1.960	1.954	88.198
0.2570 $\mathbf{2}_{\text{opt}}$	1.947	1.947	1.971	1.971	1.964	1.964	1.961	86.182
0.3855 $\mathbf{2}_{\text{opt}}$	1.959	1.959	1.978	1.978	1.968	1.968	1.968	84.627
0.5140 $\mathbf{2}_{\text{opt}}$	1.972	1.972	1.985	1.985	1.974	1.974	1.977	82.908
0.6425 $\mathbf{2}_{\text{opt}}$	1.987	1.987	1.994	1.994	1.980	1.980	1.987	80.990

**Table S5:** Geometrical parameters of **3** in the presence of an Oriented External Electric Field

High spin								
$F_z$ (V/Å)	Fe-N1	Fe-N2	Fe-N3	Fe-N4	Fe-N5	Fe-N6	Average Fe-N	$\angle$ SCN-Fe-NCS
$\mathbf{3}_{\text{xray}}$	2.034	2.062	2.277	2.248	2.243	2.172	2.173	97.999
$\mathbf{3}_{\text{opt}}$	2.063	2.090	2.294	2.259	2.233	2.202	2.190	97.755
0.1285 $\mathbf{3}_{\text{opt}}$	2.102	2.114	2.291	2.258	2.233	2.208	2.201	90.295
0.2570 $\mathbf{3}_{\text{opt}}$	2.141	2.159	2.276	2.249	2.229	2.210	2.211	86.045
0.3855 $\mathbf{3}_{\text{opt}}$	2.190	2.225	2.253	2.241	2.226	2.210	2.224	81.919
0.5140 $\mathbf{3}_{\text{opt}}$	2.239	2.359	2.225	2.236	2.222	2.207	2.248	77.012
0.6425 $\mathbf{3}_{\text{opt}}$	2.375	2.523	2.203	2.203	2.218	2.192	2.285	72.272

**Table S6:** Geometrical parameters of **3** in the presence of an Oriented External Electric Field

Low spin								
$F_z$ (V/Å)	Fe-N1	Fe-N2	Fe-N3	Fe-N4	Fe-N5	Fe-N6	Average Fe-N	$\angle$ SCN-Fe-NCS
$\mathbf{3}_{\text{xray}}$	1.912	1.920	1.998	1.975	1.988	1.968	1.960	89.968
$\mathbf{3}_{\text{opt}}$	1.931	1.937	2.010	1.998	2.008	1.983	1.978	90.307
0.1285 $\mathbf{3}_{\text{opt}}$	1.938	1.942	2.017	2.004	2.005	1.986	1.982	88.248
0.2570 $\mathbf{3}_{\text{opt}}$	1.947	1.951	2.024	2.014	2.004	1.989	1.988	86.483
0.3855 $\mathbf{3}_{\text{opt}}$	1.958	1.960	2.032	2.022	2.007	1.992	1.995	84.593
0.5140 $\mathbf{3}_{\text{opt}}$	1.969	1.972	2.039	2.030	2.011	1.997	2.003	83.090
0.6425 $\mathbf{3}_{\text{opt}}$	1.983	1.985	2.046	2.039	2.018	2.003	2.012	81.243

**Table S7:**  $t_{2g}$ - $e_g$  gap of complex **1,2** and **3** in presence of OEEFs.

$F_z$ (V/Å)	LS	HS
$\mathbf{1}_{\text{opt}}$	4.14	1.18
0.1285 $\mathbf{1}_{\text{opt}}$	4.26	1.17
0.2570 $\mathbf{1}_{\text{opt}}$	4.24	0.42

0.3855 <b>1</b> <sub>opt</sub>	4.12	0.56
0.5140 <b>1</b> <sub>opt</sub>	4.10	0.62
0.6425 <b>1</b> <sub>opt</sub>	3.95	0.49
<b>2</b> <sub>opt</sub>	3.90	1.40
0.1285 <b>2</b> <sub>opt</sub>	4.03	1.18
0.2570 <b>2</b> <sub>opt</sub>	4.02	1.30
0.3855 <b>2</b> <sub>opt</sub>	4.01	1.19
0.5140 <b>2</b> <sub>opt</sub>	3.89	0.62
0.6425 <b>2</b> <sub>opt</sub>	3.71	0.51
<b>3</b> <sub>opt</sub>	4.15	1.08
0.1285 <b>3</b> <sub>opt</sub>	4.23	1.00
0.2570 <b>3</b> <sub>opt</sub>	4.21	0.97
0.3855 <b>3</b> <sub>opt</sub>	4.12	0.98
0.5140 <b>3</b> <sub>opt</sub>	4.08	1.03
0.6425 <b>3</b> <sub>opt</sub>	3.95	0.68

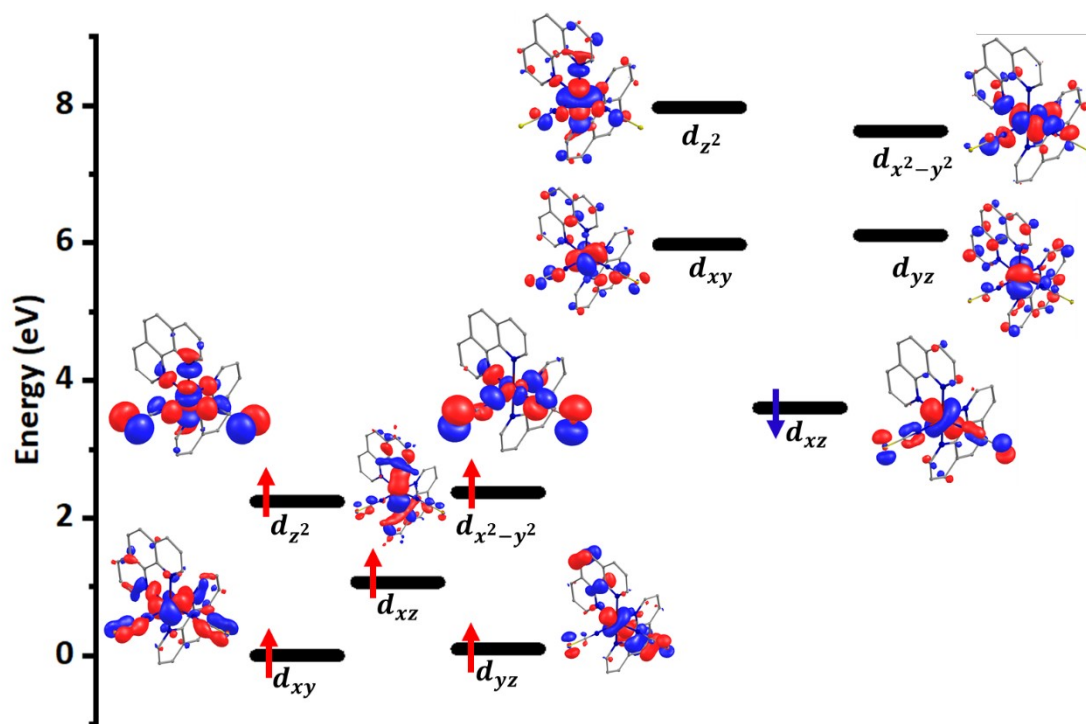


Figure S1: *d* orbital splitting diagram of complex 1 in High Spin

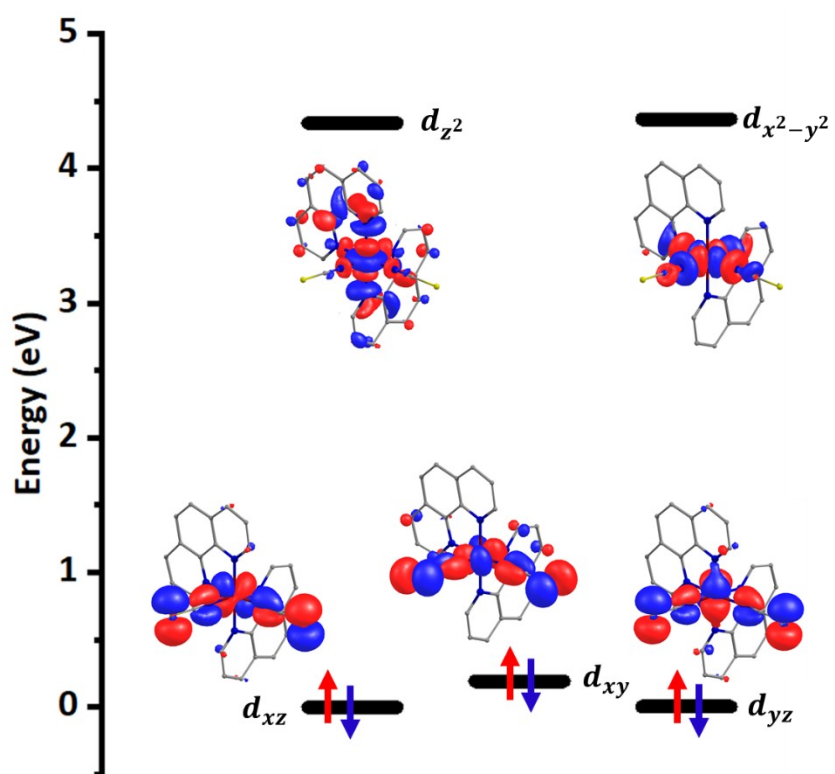


Figure S2: *d* orbital splitting diagram of complex 1 in Low Spin

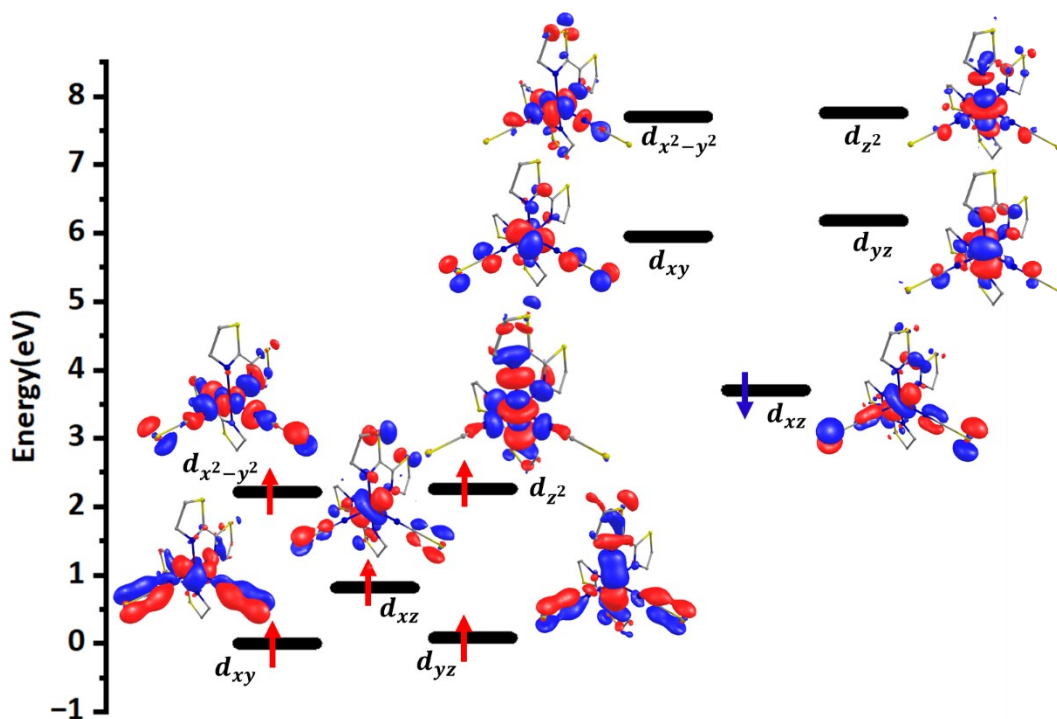


Figure S3: *d* orbital splitting diagram of complex 2 in High Spin

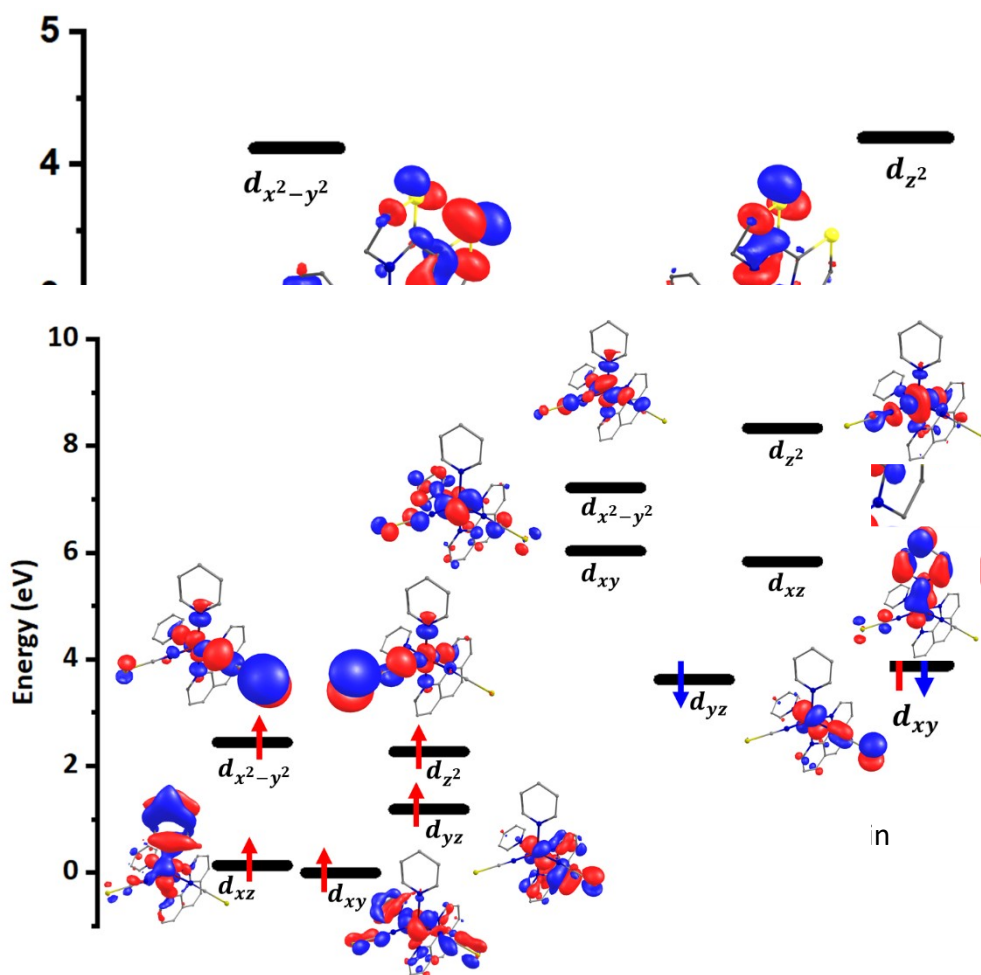


Figure S5: *d* orbital splitting diagram of complex 3 in High Spin

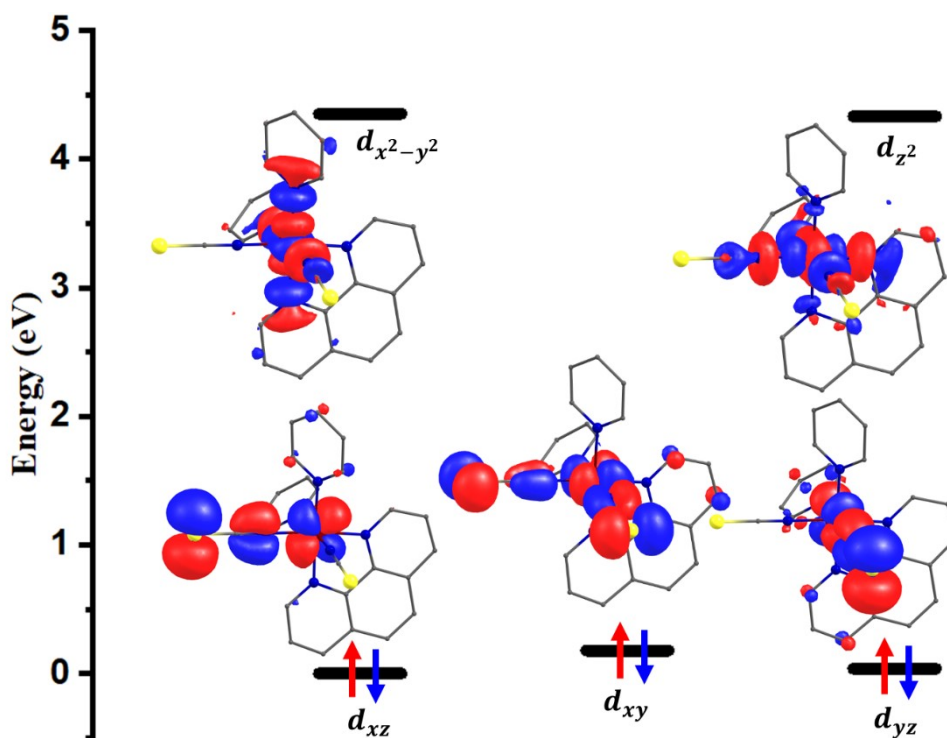


Figure S6: d orbital splitting diagram of complex **3** in Low Spin

Table S8: Energy parameters of **1** in the presence of an Oriented External Electric Field

$F_z$ (V/Å)	$\Delta E_{HL}$ (kJ mol <sup>-1</sup> )	$\Delta H_{HL}$ (kJ mol <sup>-1</sup> )	$\Delta S_{HL}$ (kJ mol <sup>-1</sup> K <sup>-1</sup> )	$\Delta ZPE_{HL}$ (kJ mol <sup>-1</sup> )
<b>1</b> <sub>xray</sub>	35.426	-5.170	0.0952	-11.074
<b>1</b> <sub>opt</sub>	35.553	-5.238	0.0863	-10.951
0.1285 <b>1</b> <sub>opt</sub>	33.694	-5.038	0.0836	-10.633
0.2570 <b>1</b> <sub>opt</sub>	27.364	-4.918	0.0832	-10.536
0.3855 <b>1</b> <sub>opt</sub>	18.602	-4.550	0.0730	-9.985
0.5140 <b>1</b> <sub>opt</sub>	9.258	-4.755	0.0769	-10.576
0.6425 <b>1</b> <sub>opt</sub>	-5.217	-7.265	0.0723	-11.374

Table S9: Energy parameters of **2** in the presence of an Oriented External Electric Field

$F_z$ (V/Å)	$\Delta E_{HL}$ (kJ mol <sup>-1</sup> )	$\Delta H_{HL}$ (kJ mol <sup>-1</sup> )	$\Delta S_{HL}$ (kJ mol <sup>-1</sup> K <sup>-1</sup> )	$\Delta ZPE_{HL}$ (kJ mol <sup>-1</sup> )
<b>2</b> <sub>xray</sub>	26.695	-6.133	0.023	-11.512
<b>2</b> <sub>opt</sub>	27.156	-6.083	0.083	-11.707
0.1285 <b>2</b> <sub>opt</sub>	26.115	-5.833	0.091	-11.549

0.2570 $\mathbf{2}_{\text{opt}}$	21.319	-5.749	0.085	-11.541
0.3855 $\mathbf{2}_{\text{opt}}$	13.818	-5.264	0.078	-10.767
0.5140 $\mathbf{2}_{\text{opt}}$	3.791	-4.904	0.062	-9.921
0.6425 $\mathbf{2}_{\text{opt}}$	-10.040	10.268	0.066	-12.15

**Table S10:** Energy parameters of **3** in the presence of an Oriented External Electric Field

$F_z$ (V/Å)	$\Delta E_{\text{HL}}$ (kJ mol <sup>-1</sup> )	$\Delta H_{\text{HL}}$ (kJ mol <sup>-1</sup> )	$\Delta S_{\text{HL}}$ (kJ mol <sup>-1</sup> K <sup>-1</sup> )	$\Delta ZPE_{\text{HL}}$ (kJ mol <sup>-1</sup> )
$\mathbf{3}_{\text{xray}}$	27.267	-5.997	0.082	-12.369
$\mathbf{3}_{\text{opt}}$	26.803	-4.920	0.096	-11.389
0.1285 $\mathbf{3}_{\text{opt}}$	25.681	-5.114	0.099	-11.791
0.2570 $\mathbf{3}_{\text{opt}}$	19.952	-5.083	0.093	-11.586
0.3855 $\mathbf{3}_{\text{opt}}$	11.685	-4.970	0.089	-11.418
0.5140 $\mathbf{3}_{\text{opt}}$	-0.507	-4.826	0.083	-11.122
0.6425 $\mathbf{3}_{\text{opt}}$	-16.218	-7.078	0.067	-11.177

**Table S11:** DFT computed HS-LS energy gap ( $\Delta U_{\text{HL}}$ ) for complex **2** at various oriented external electric fields using several functionals.

	0% HF	0% HF	10% HF	15% HF	20% HF	25% HF
$+F_z$ (V/Å)	PBE	BLYP	TPSSh	B3LYP*	B3LYP	PBE0
<b>Opt</b>	65.446	53.120	15.44973	48.268	-30.071	-57.694
0.1285	68.684	61.581	14.5662	50.729	-27.493	-58.055
0.2570	64.112	58.208	9.77781	48.942	-32.052	-63.486
0.3855	58.234	51.004	3.05102	42.616	-39.125	-71.219
0.514	48.560	41.828	-6.13081	34.081	-48.291	-80.953

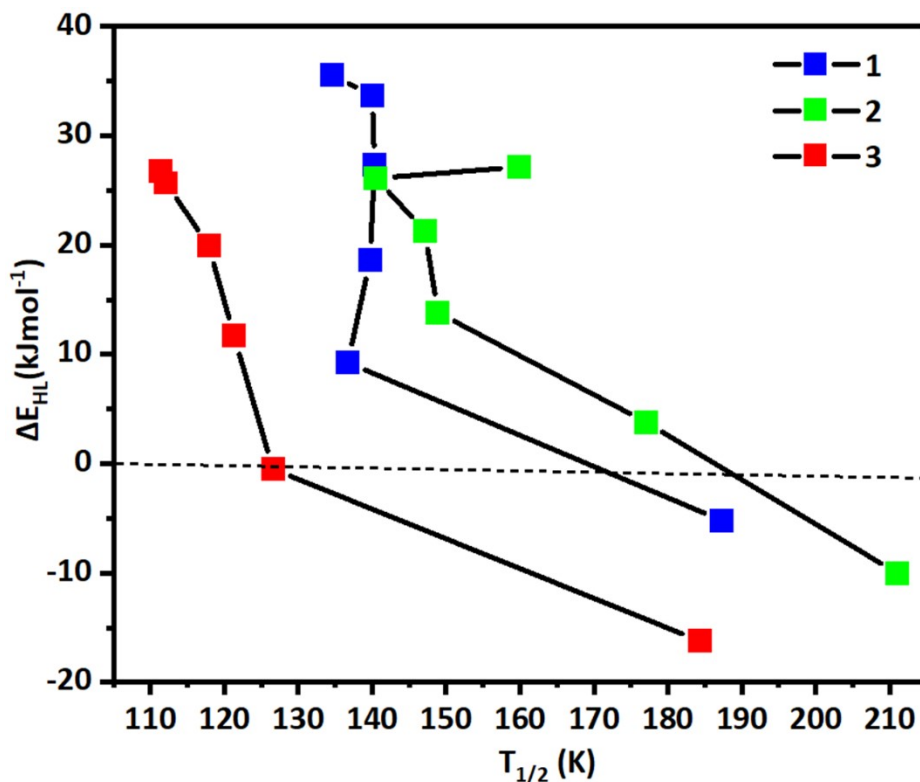


Figure S7.  $\Delta E_{HL}$  vs  $T_{1/2}$  for complexes 1, 2 and 3.

Table S12: A few selected parameters of 1 in HS and LS state adsorbed on Au(111) surface

	Fe-N1	Fe-N2	Fe-N3	Fe-N4	Fe-N5	Fe-N6	Average Fe-N	Au-S1	Au-S2
$\mathbf{1}_{HS}@Au(111)$	1.920	1.913	1.939	1.937	2.107	2.090	1.984	2.861	2.921
$\mathbf{1}_{LS}@Au(111)$	1.936	1.906	1.964	1.957	1.967	1.966	1.950	2.517	2.580

Table S13: A few selected parameters of 1 in HS and LS state adsorbed on Au(111) surface

	S-S	N-Fe-N	S-Fe-S
$\mathbf{1}_{HS}$	7.361	97.353	98.010
$\mathbf{1}_{LS}$	6.778	90.833	91.057
$\mathbf{1}_{HS}@Au(111)$	7.687	90.988	105.295
$\mathbf{1}_{LS}@Au(111)$	8.144	99.282	123.852

## Geometrical Coordinates

$HS\mathbf{1}_{opt}$

Fe 0.00000000 0.00000000 0.00000000



N	-1.854350000	1.134940000	-0.326700000
N	-1.080210000	-1.083070000	-1.665460000
N	-0.738060000	-1.363040000	1.362900000
C	-2.708780000	0.624880000	-1.251580000
C	-2.293020000	-0.554730000	-1.969230000
C	-2.226700000	2.209420000	0.367670000
C	-3.468630000	2.845260000	0.178290000
C	-4.345320000	2.345740000	-0.770940000
C	-3.978860000	1.202600000	-1.521190000
C	-4.825210000	0.608130000	-2.519190000
C	-4.430400000	-0.509150000	-3.202900000
C	-3.157930000	-1.125020000	-2.942840000
C	-2.709290000	-2.294000000	-3.603500000
C	-1.472980000	-2.826020000	-3.276550000
C	-0.688310000	-2.185340000	-2.297200000
C	-1.255000000	-2.098940000	2.138460000
S	-1.964690000	-3.111670000	3.199430000
H	-1.506350000	2.579540000	1.101010000
H	-3.720640000	3.721950000	0.776410000
H	-5.314000000	2.818960000	-0.945590000
H	-5.796330000	1.066420000	-2.717560000
H	-5.080960000	-0.957630000	-3.956610000
H	-3.342160000	-2.764880000	-4.358940000
H	-1.097430000	-3.727760000	-3.762200000
H	0.290040000	-2.586150000	-2.021360000
N	1.854240000	-1.135140000	-0.326940000
N	1.080050000	1.082940000	-1.665530000
N	0.738060000	1.363040000	1.362840000
C	2.708550000	-0.625150000	-1.251960000
C	2.226620000	-2.209630000	0.367380000
C	2.292750000	0.554500000	-1.969540000
C	0.688110000	2.185250000	-2.297180000

C	1.255150000	2.099360000	2.137900000
C	3.978540000	-1.202950000	-1.521790000
C	3.468480000	-2.845560000	0.177800000
H	1.506360000	-2.579700000	1.100840000
C	3.157520000	1.124730000	-2.943310000
C	1.472650000	2.825870000	-3.276680000
H	-0.290150000	2.586130000	-2.021130000
S	1.965100000	3.112560000	3.198240000
C	4.345040000	-2.346110000	-0.771590000
C	4.824760000	-0.608550000	-2.519950000
H	3.720540000	-3.722260000	0.775890000
C	4.429900000	0.508750000	-3.203600000
C	2.708850000	2.293740000	-3.603880000
H	1.097090000	3.727650000	-3.762240000
H	5.313650000	-2.819420000	-0.946410000
H	5.795810000	-1.066900000	-2.718480000
H	5.080360000	0.957170000	-3.957440000
H	3.341620000	2.764590000	-4.359430000

LS<sub>1opt</sub>

Fe	0.000000000	0.000000000	0.000000000
N	-1.643610000	1.094600000	-0.056410000
N	-0.944200000	-1.042850000	-1.393040000
N	-0.809820000	-1.111030000	1.354990000
C	-2.572680000	0.609010000	-0.927210000
C	-2.187580000	-0.551240000	-1.662980000
C	-1.963260000	2.169290000	0.668730000
C	-3.210560000	2.812800000	0.550170000
C	-4.157060000	2.333840000	-0.343060000
C	-3.849030000	1.191640000	-1.119860000
C	-4.736760000	0.590230000	-2.078470000
C	-4.365410000	-0.520640000	-2.789750000
C	-3.075700000	-1.129940000	-2.602420000

C	-2.623080000	-2.280260000	-3.291560000
C	-1.357640000	-2.773100000	-3.012240000
C	-0.547130000	-2.131020000	-2.056230000
C	-1.352450000	-1.758850000	2.182660000
S	-2.091320000	-2.668390000	3.326710000
H	-1.201200000	2.527070000	1.361890000
H	-3.413360000	3.687960000	1.168880000
H	-5.127680000	2.822460000	-0.449440000
H	-5.720560000	1.039000000	-2.229830000
H	-5.050170000	-0.966440000	-3.513950000
H	-3.266880000	-2.766120000	-4.027460000
H	-0.973120000	-3.658750000	-3.519730000
H	0.445930000	-2.516490000	-1.823310000
N	1.643620000	-1.094600000	-0.056410000
N	0.944210000	1.042860000	-1.393020000
N	0.809820000	1.111030000	1.354990000
C	2.572690000	-0.609000000	-0.927190000
C	1.963260000	-2.169290000	0.668740000
C	2.187590000	0.551250000	-1.662960000
C	0.547140000	2.131030000	-2.056220000
C	1.352440000	1.758750000	2.182740000
C	3.849040000	-1.191630000	-1.119840000
C	3.210570000	-2.812790000	0.550170000
H	1.201200000	-2.527080000	1.361880000
C	3.075720000	1.129950000	-2.602400000
C	1.357650000	2.773110000	-3.012230000
H	-0.445920000	2.516500000	-1.823300000
S	2.090690000	2.666860000	3.328320000
C	4.157070000	-2.333830000	-0.343050000
C	4.736780000	-0.590210000	-2.078440000
H	3.413370000	-3.687960000	1.168890000
C	4.365430000	0.520660000	-2.789720000

C	2.623090000	2.280280000	-3.291550000
H	0.973130000	3.658760000	-3.519720000
H	5.127700000	-2.822440000	-0.449410000
H	5.720580000	-1.038980000	-2.229800000
H	5.050190000	0.966460000	-3.513920000
H	3.266900000	2.766130000	-4.027440000

HS<sub>2</sub><sub>opt</sub>

Fe	0.000000000	0.000000000	0.000000000
S	3.092640000	-1.553060000	-3.197930000
S	4.166520000	1.303260000	-1.675820000
S	-3.091490000	1.550570000	-3.200120000
S	-4.166150000	-1.304460000	-1.676050000
S	-1.362220000	3.365020000	3.240250000
S	1.363830000	-3.365910000	3.238860000
N	0.993140000	-1.138800000	-1.666420000
N	1.917440000	1.003390000	-0.347570000
N	-0.992890000	1.137730000	-1.667000000
N	-1.917410000	-1.003820000	-0.347400000
N	-0.575710000	1.435220000	1.346820000
N	0.575710000	-1.435220000	1.346990000
C	2.153930000	-0.682620000	-1.992560000
C	1.589260000	-2.557850000	-3.567970000
C	0.690670000	-2.407430000	-2.326680000
C	2.647740000	0.510710000	-1.291730000
C	3.986360000	2.285110000	-0.121720000
C	2.486610000	2.226680000	0.218400000
C	-2.153440000	0.681180000	-1.993500000
C	-1.587980000	2.555220000	-3.570030000
C	-0.690120000	2.405860000	-2.328080000
C	-2.647460000	-0.511710000	-1.292050000
C	-3.986390000	-2.285420000	-0.121340000
C	-2.486740000	-2.226740000	0.219190000

C	-0.907500000	2.248390000	2.146130000
C	0.907930000	-2.248650000	2.145840000
H	1.891350000	-3.596100000	-3.752170000
H	1.125000000	-2.144990000	-4.474680000
H	0.875500000	-3.211760000	-1.593320000
H	-0.372060000	-2.447150000	-2.604360000
H	4.344360000	3.305980000	-0.302590000
H	4.610460000	1.808660000	0.647300000
H	1.940400000	3.082560000	-0.214940000
H	2.320690000	2.246010000	1.304120000
H	-1.890070000	3.593290000	-3.755240000
H	-1.123130000	2.141670000	-4.476120000
H	-0.875450000	3.210770000	-1.595480000
H	0.372760000	2.445450000	-2.605170000
H	-4.344300000	-3.306400000	-0.301720000
H	-4.610730000	-1.808550000	0.647230000
H	-1.940380000	-3.082890000	-0.213440000
H	-2.321140000	-2.245370000	1.304970000

LS<sub>2opt</sub>

Fe	0.000000000	0.000000000	0.000000000
S	3.198210000	-1.394410000	-2.755470000
S	4.025310000	1.601790000	-0.996510000
S	-3.198530000	1.394540000	-2.754980000
S	-4.024640000	-1.602770000	-0.997480000
S	-2.002160000	2.685000000	3.364710000
S	2.002920000	-2.684260000	3.364870000
N	1.005250000	-0.988890000	-1.364340000
N	1.607580000	1.112950000	-0.086200000
N	-1.005360000	0.989010000	-1.364190000
N	-1.607320000	-1.113210000	-0.086480000
N	-0.794830000	1.112260000	1.360950000
N	0.794830000	-1.112260000	1.361000000

C	2.190410000	-0.520230000	-1.624060000
C	1.716010000	-2.400340000	-3.216050000
C	0.747170000	-2.265390000	-2.031530000
C	2.538590000	0.686060000	-0.887590000
C	3.490830000	2.539220000	0.506820000
C	1.965610000	2.366830000	0.581080000
C	-2.190380000	0.520120000	-1.624100000
C	-1.716840000	2.401540000	-3.214850000
C	-0.747700000	2.265990000	-2.030620000
C	-2.538280000	-0.686510000	-0.888020000
C	-3.490220000	-2.540220000	0.505860000
C	-1.965070000	-2.367310000	0.580540000
C	-1.305480000	1.768320000	2.201940000
C	1.305820000	-1.768020000	2.202010000
H	2.032230000	-3.435560000	-3.391950000
H	1.303880000	-1.978530000	-4.143400000
H	0.903390000	-3.064150000	-1.285960000
H	-0.297150000	-2.313310000	-2.368670000
H	3.795990000	3.586780000	0.395570000
H	4.004760000	2.096950000	1.371560000
H	1.439140000	3.189190000	0.067140000
H	1.612190000	2.342680000	1.619730000
H	-2.033570000	3.436780000	-3.389740000
H	-1.304670000	1.980800000	-4.142650000
H	-0.903890000	3.064200000	-1.284460000
H	0.296530000	2.314360000	-2.367990000
H	-3.794970000	-3.587870000	0.394390000
H	-4.004520000	-2.098250000	1.370530000
H	-1.438190000	-3.189430000	0.066650000
H	-1.611930000	-2.343160000	1.619290000
$\text{HS}_3_{\text{opt}}$			
Fe	0.000000000	0.000000000	0.000000000

N	2.047050000	-0.801140000	-0.130300000
N	1.010100000	1.231770000	-1.602250000
N	-2.004390000	0.974650000	-0.135910000
N	-0.585870000	-1.386330000	-1.731280000
C	3.143440000	1.589330000	-2.727510000
C	2.314140000	0.902890000	-1.797440000
C	-1.087330000	-0.950930000	-2.901040000
C	2.865150000	-0.179360000	-1.018430000
C	1.237250000	2.984640000	-3.238210000
C	-2.190430000	2.278310000	0.143700000
C	5.033860000	0.157930000	-2.149430000
C	4.227290000	-0.548080000	-1.192500000
C	4.724180000	-1.605480000	-0.393490000
C	2.543950000	-1.787950000	0.613510000
C	2.564470000	2.655680000	-3.455710000
C	0.495470000	2.241810000	-2.300060000
C	-0.595140000	-3.626330000	-2.615450000
C	4.513400000	1.185590000	-2.886470000
C	-4.391520000	0.691780000	-0.235020000
C	-4.580910000	2.048940000	0.039990000
C	-1.112270000	-3.167250000	-3.830150000
C	-3.088980000	0.198850000	-0.317020000
C	-1.364600000	-1.800020000	-3.973290000
C	-3.456880000	2.857090000	0.232850000
C	-0.348650000	-2.704180000	-1.597340000
C	3.879440000	-2.223100000	0.513800000
S	1.677580000	3.316110000	3.201290000
S	-1.638390000	-3.145600000	3.333030000
N	0.608560000	1.440720000	1.385990000
N	-0.608560000	-1.440720000	1.345110000
C	-1.043340000	-2.158860000	2.187160000
C	1.058570000	2.228020000	2.151150000

H	0.757690000	3.801850000	-3.778430000
H	-1.285850000	2.865640000	0.316100000
H	6.076790000	-0.141470000	-2.272460000
H	5.765280000	-1.918600000	-0.497370000
H	1.851990000	-2.252150000	1.320560000
H	-1.771890000	-1.390350000	-4.899120000
H	3.169470000	3.208320000	-4.177770000
H	-0.554590000	2.481840000	-2.116800000
H	-0.383390000	-4.684320000	-2.451580000
H	5.132680000	1.721680000	-3.608660000
H	-5.587010000	2.468460000	0.107860000
H	-1.316410000	-3.861300000	-4.648300000
H	-3.553190000	3.920670000	0.456910000
H	0.046120000	-3.024170000	-0.630660000
H	-5.237240000	0.018740000	-0.385100000
H	4.228550000	-3.037430000	1.150210000
H	-2.897410000	-0.855440000	-0.526730000
H	-1.278360000	0.121660000	-2.980270000

LS<sub>3</sub><sub>opt</sub>

Fe	0.000000000	0.000000000	0.000000000
N	1.851330000	-0.710690000	0.008360000
N	0.750400000	1.215770000	-1.397030000
N	-1.836120000	0.811130000	0.042660000
N	-0.531170000	-1.312350000	-1.426940000
C	2.866960000	1.750840000	-2.518940000
C	2.083400000	1.002450000	-1.604930000
C	-1.147150000	-0.926240000	-2.564950000
C	2.682450000	-0.041330000	-0.837010000
C	0.858040000	2.971690000	-3.047610000
C	-2.021720000	2.125500000	0.295260000
C	4.840520000	0.444910000	-1.898510000
C	4.064350000	-0.327290000	-0.966620000



C	4.587170000	-1.362210000	-0.154940000
C	2.374640000	-1.670920000	0.772960000
C	2.212300000	2.766610000	-3.254460000
C	0.164020000	2.178690000	-2.114240000
C	-0.552350000	-3.576320000	-2.254200000
C	4.265400000	1.439730000	-2.643770000
C	-4.230020000	0.526610000	0.066660000
C	-4.416590000	1.889310000	0.311700000
C	-1.180280000	-3.168920000	-3.433890000
C	-2.933660000	0.029720000	-0.057490000
C	-1.484090000	-1.813670000	-3.584920000
C	-3.284590000	2.699230000	0.431450000
C	-0.248690000	-2.625350000	-1.281840000
C	3.736670000	-2.025240000	0.715380000
S	1.570450000	2.937020000	3.394020000
S	-1.472920000	-2.938550000	3.428530000
N	0.576920000	1.244020000	1.368070000
N	-0.576920000	-1.244020000	1.359840000
C	-0.959210000	-1.949090000	2.230900000
C	0.996870000	1.949650000	2.219220000
H	0.312060000	3.741030000	-3.594920000
H	-1.117360000	2.723250000	0.408390000
H	5.904540000	0.222230000	-2.000750000
H	5.645760000	-1.622760000	-0.215810000
H	1.689540000	-2.170930000	1.458830000
H	-1.980790000	-1.436630000	-4.480150000
H	2.772260000	3.371580000	-3.970260000
H	-0.901940000	2.340050000	-1.950430000
H	-0.296570000	-4.622120000	-2.077120000
H	4.864680000	2.019590000	-3.348570000
H	-5.419600000	2.308450000	0.414390000
H	-1.430500000	-3.890180000	-4.214410000

H	-3.367580000	3.768180000	0.633840000
H	0.227490000	-2.913550000	-0.345590000
H	-5.075830000	-0.156630000	-0.024680000
H	4.102760000	-2.821850000	1.364350000
H	-2.755130000	-1.031550000	-0.229070000
H	-1.383310000	0.133700000	-2.655720000

<sup>HS</sup>**1@Au(111)**<sub>opt</sub>

Au	0.000000000	0.000000000	0.000000000
Au	-2.949542000	5.108757000	0.000000000
Au	-5.899085000	10.217514000	0.000000000
Au	5.899075000	0.000000000	0.000000000
Au	2.949532000	5.108757000	0.000000000
Au	-0.000012000	10.217514000	0.000000000
Au	11.798149000	0.000000000	0.000000000
Au	8.848606000	5.108757000	0.000000000
Au	5.899062000	10.217514000	0.000000000
Au	-1.474771000	2.554378000	0.000000000
Au	-4.424314000	7.663137000	0.000000000
Au	-7.373856000	12.771894000	0.000000000
Au	4.424303000	2.554378000	0.000000000
Au	1.474758000	7.663137000	0.000000000
Au	-1.474785000	12.771894000	0.000000000
Au	10.323377000	2.554378000	0.000000000
Au	7.373833000	7.663137000	0.000000000
Au	4.424291000	12.771894000	0.000000000
Au	2.949537000	0.000000000	0.000000000
Au	-0.000006000	5.108757000	0.000000000
Au	-2.949549000	10.217514000	0.000000000
Au	8.848614000	0.000000000	0.000000000
Au	5.899071000	5.108757000	0.000000000
Au	2.949529000	10.217514000	0.000000000
Au	14.747688000	0.000000000	0.000000000

Au	11.798146000	5.108757000	0.000000000
Au	8.848602000	10.217514000	0.000000000
Au	1.474766000	2.554378000	0.000000000
Au	-1.474779000	7.663137000	0.000000000
Au	-4.424321000	12.771894000	0.000000000
Au	7.373843000	2.554378000	0.000000000
Au	4.424298000	7.663137000	0.000000000
Au	1.474756000	12.771894000	0.000000000
Au	13.272917000	2.554378000	0.000000000
Au	10.323372000	7.663137000	0.000000000
Au	7.373830000	12.771894000	0.000000000
Au	1.474789000	0.851469000	2.408288000
Au	-1.474755000	5.960227000	2.408288000
Au	-4.424299000	11.068983000	2.408288000
Au	7.373864000	0.851469000	2.408288000
Au	4.424322000	5.960227000	2.408288000
Au	1.474778000	11.068983000	2.408288000
Au	13.272939000	0.851469000	2.408288000
Au	10.323396000	5.960227000	2.408288000
Au	7.373853000	11.068983000	2.408288000
Au	0.000017000	3.405848000	2.408288000
Au	-2.949526000	8.514602000	2.408288000
Au	-5.899071000	13.623364000	2.408288000
Au	5.899093000	3.405848000	2.408288000
Au	2.949550000	8.514602000	2.408288000
Au	0.000005000	13.623364000	2.408288000
Au	11.798166000	3.405848000	2.408288000
Au	8.848626000	8.514602000	2.408288000
Au	5.899080000	13.623364000	2.408288000
Au	4.424325000	0.851469000	2.408288000
Au	1.474782000	5.960227000	2.408288000
Au	-1.474761000	11.068983000	2.408288000

Au	10.323399000	0.851469000	2.408288000
Au	7.373856000	5.960227000	2.408288000
Au	4.424313000	11.068983000	2.408288000
Au	16.222478000	0.851469000	2.408288000
Au	13.272936000	5.960227000	2.408288000
Au	10.323393000	11.068983000	2.408288000
Au	2.949553000	3.405848000	2.408288000
Au	0.000011000	8.514602000	2.408288000
Au	-2.949535000	13.623364000	2.408288000
Au	8.848626000	3.405848000	2.408288000
Au	5.899086000	8.514602000	2.408288000
Au	2.949540000	13.623364000	2.408288000
Au	14.747706000	3.405848000	2.408288000
Au	11.798165000	8.514602000	2.408288000
Au	8.848619000	13.623364000	2.408288000
Au	-0.022616000	1.654148000	4.796401000
Au	-2.971844000	6.810778000	4.796850000
Au	-5.906402000	11.905682000	4.799448000
Au	5.911992000	1.689176000	4.807148000
Au	2.968163000	6.794069000	4.740280000
Au	-0.019402000	11.918592000	4.817812000
Au	11.789354000	1.730364000	4.809846000
Au	8.857921000	6.816291000	4.822719000
Au	5.933822000	11.860655000	4.818700000
Au	-1.478300000	4.247684000	4.786242000
Au	-4.427706000	9.367034000	4.794368000
Au	-7.383163000	14.439379000	4.805344000
Au	4.490224000	4.225078000	4.775256000
Au	1.465203000	9.372968000	4.818707000
Au	-1.476958000	14.473254000	4.794542000
Au	10.338878000	4.261661000	4.813233000
Au	7.384714000	9.336603000	4.813089000

Au	4.402970000	14.555687000	4.745431000
Au	2.976466000	1.626951000	4.789472000
Au	-0.073080000	6.876469000	4.773796000
Au	-2.938679000	11.913222000	4.802610000
Au	8.849269000	1.707670000	4.796057000
Au	5.938209000	6.781607000	4.817590000
Au	2.883065000	11.921474000	4.789943000
Au	14.748649000	1.717895000	4.816366000
Au	11.796707000	6.814220000	4.793448000
Au	8.858447000	11.910568000	4.811579000
Au	1.510061000	4.125362000	4.749086000
Au	-1.483443000	9.393214000	4.826590000
Au	-4.416458000	14.444448000	4.798073000
Au	7.409847000	4.253316000	4.809259000
Au	4.420056000	9.277418000	4.765074000
Au	1.460174000	14.487312000	4.789261000
Au	13.272743000	4.257455000	4.809768000
Au	10.315211000	9.369451000	4.814299000
Au	7.361245000	14.467009000	4.778791000
Fe	3.822038000	8.456417000	9.936853000
N	1.922554000	9.029973000	10.054959000
N	4.010129000	9.696972000	11.478962000
N	4.228293000	9.857310000	8.709086000
N	5.690167000	7.832354000	9.985240000
N	3.580387000	7.030415000	11.284365000
N	3.370946000	7.210527000	8.545719000
C	1.660737000	9.748653000	11.186904000
C	2.800561000	10.115278000	11.959111000
C	0.891023000	8.767828000	9.240994000
C	-0.436032000	9.102898000	9.568612000
C	-0.714790000	9.761202000	10.754441000
C	0.360038000	10.124006000	11.599825000

C	0.221870000	10.865033000	12.818614000
C	1.315961000	11.234795000	13.552303000
C	2.642072000	10.895498000	13.131171000
C	3.819804000	11.268182000	13.817470000
C	5.042658000	10.863489000	13.314444000
C	5.102387000	10.078452000	12.149205000
C	4.447172000	10.814940000	8.051424000
C	5.923183000	6.950394000	11.005596000
C	6.717181000	8.132509000	9.179019000
C	4.774259000	6.538906000	11.731836000
C	2.460937000	6.571599000	11.853875000
C	2.658618000	6.511379000	7.910885000
C	7.200268000	6.431761000	11.326141000
C	8.012000000	7.624184000	9.393096000
C	4.888919000	5.623247000	12.804021000
C	2.482497000	5.655541000	12.919262000
C	8.270227000	6.795380000	10.472892000
C	7.304519000	5.557870000	12.458132000
C	6.198910000	5.181098000	13.174785000
C	3.691237000	5.194249000	13.417611000
S	4.742766000	12.174833000	7.191362000
S	1.532789000	5.627671000	7.118739000
H	1.105994000	8.249134000	8.311328000
H	-1.229143000	8.833586000	8.874487000
H	-1.738239000	10.013037000	11.033921000
H	-0.779376000	11.140073000	13.150616000
H	1.190080000	11.808828000	14.475202000
H	3.758992000	11.862465000	14.729765000
H	5.970901000	11.136571000	13.809921000
H	6.059212000	9.742493000	11.751215000
H	6.509891000	8.791962000	8.340516000
H	1.521192000	6.936847000	11.450289000

H	8.802077000	7.885014000	8.686928000
H	1.534904000	5.323266000	13.338493000
H	9.269718000	6.403786000	10.653630000
H	8.286830000	5.179942000	12.738368000
H	6.304732000	4.510162000	14.027826000
H	3.712216000	4.498097000	14.256444000

**LS1@Au(111)<sub>opt</sub>**

Au	0.000000000	0.000000000	0.000000000
Au	-2.949542000	5.108757000	0.000000000
Au	-5.899085000	10.217514000	0.000000000
Au	5.899075000	0.000000000	0.000000000
Au	2.949532000	5.108757000	0.000000000
Au	-0.000012000	10.217514000	0.000000000
Au	11.798149000	0.000000000	0.000000000
Au	8.848606000	5.108757000	0.000000000
Au	5.899062000	10.217514000	0.000000000
Au	-1.474771000	2.554378000	0.000000000
Au	-4.424314000	7.663137000	0.000000000
Au	-7.373856000	12.771894000	0.000000000
Au	4.424303000	2.554378000	0.000000000
Au	1.474758000	7.663137000	0.000000000
Au	-1.474785000	12.771894000	0.000000000
Au	10.323377000	2.554378000	0.000000000
Au	7.373833000	7.663137000	0.000000000
Au	4.424291000	12.771894000	0.000000000
Au	2.949537000	0.000000000	0.000000000
Au	-0.000006000	5.108757000	0.000000000
Au	-2.949549000	10.217514000	0.000000000
Au	8.848614000	0.000000000	0.000000000
Au	5.899071000	5.108757000	0.000000000
Au	2.949529000	10.217514000	0.000000000
Au	14.747688000	0.000000000	0.000000000

Au	11.798146000	5.108757000	0.000000000
Au	8.848602000	10.217514000	0.000000000
Au	1.474766000	2.554378000	0.000000000
Au	-1.474779000	7.663137000	0.000000000
Au	-4.424321000	12.771894000	0.000000000
Au	7.373843000	2.554378000	0.000000000
Au	4.424298000	7.663137000	0.000000000
Au	1.474756000	12.771894000	0.000000000
Au	13.272917000	2.554378000	0.000000000
Au	10.323372000	7.663137000	0.000000000
Au	7.373830000	12.771894000	0.000000000
Au	1.474789000	0.851469000	2.408288000
Au	-1.474755000	5.960227000	2.408288000
Au	-4.424299000	11.068983000	2.408288000
Au	7.373864000	0.851469000	2.408288000
Au	4.424322000	5.960227000	2.408288000
Au	1.474778000	11.068983000	2.408288000
Au	13.272939000	0.851469000	2.408288000
Au	10.323396000	5.960227000	2.408288000
Au	7.373853000	11.068983000	2.408288000
Au	0.000017000	3.405848000	2.408288000
Au	-2.949526000	8.514602000	2.408288000
Au	-5.899071000	13.623364000	2.408288000
Au	5.899093000	3.405848000	2.408288000
Au	2.949550000	8.514602000	2.408288000
Au	0.000005000	13.623364000	2.408288000
Au	11.798166000	3.405848000	2.408288000
Au	8.848626000	8.514602000	2.408288000
Au	5.899080000	13.623364000	2.408288000
Au	4.424325000	0.851469000	2.408288000
Au	1.474782000	5.960227000	2.408288000
Au	-1.474761000	11.068983000	2.408288000



Au	10.323399000	0.851469000	2.408288000
Au	7.373856000	5.960227000	2.408288000
Au	4.424313000	11.068983000	2.408288000
Au	16.222478000	0.851469000	2.408288000
Au	13.272936000	5.960227000	2.408288000
Au	10.323393000	11.068983000	2.408288000
Au	2.949553000	3.405848000	2.408288000
Au	0.000011000	8.514602000	2.408288000
Au	-2.949535000	13.623364000	2.408288000
Au	8.848626000	3.405848000	2.408288000
Au	5.899086000	8.514602000	2.408288000
Au	2.949540000	13.623364000	2.408288000
Au	14.747706000	3.405848000	2.408288000
Au	11.798165000	8.514602000	2.408288000
Au	8.848619000	13.623364000	2.408288000
Au	0.007674000	1.707931000	4.800431000
Au	-2.961471000	6.817804000	4.809912000
Au	-5.910522000	11.902291000	4.808145000
Au	5.928450000	1.600394000	4.774894000
Au	2.863809000	6.818111000	4.755379000
Au	-0.060658000	11.906690000	4.819039000
Au	11.784743000	1.733414000	4.822715000
Au	8.883917000	6.820946000	4.811686000
Au	6.000710000	11.908071000	4.780649000
Au	-1.511641000	4.259935000	4.821306000
Au	-4.427013000	9.364467000	4.793317000
Au	-7.393690000	14.441136000	4.818131000
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Au	1.432047000	9.346320000	4.813743000
Au	-1.466139000	14.429256000	4.821481000
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Au	4.443737000	14.607836000	4.731547000
Au	2.927368000	1.613096000	4.756106000
Au	-0.046145000	6.807285000	4.802162000
Au	-2.961444000	11.904382000	4.813145000
Au	8.838426000	1.712064000	4.806467000
Au	5.985282000	6.789939000	4.785997000
Au	2.805129000	11.911495000	4.760767000
Au	14.768915000	1.746352000	4.823084000
Au	11.800549000	6.814691000	4.795776000
Au	8.878958000	11.914577000	4.814611000
Au	1.394971000	4.276872000	4.768284000
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Au	-4.429091000	14.421392000	4.805884000
Au	7.455343000	4.266404000	4.787236000
Au	4.443286000	9.202824000	4.701004000
Au	1.448776000	14.528448000	4.781139000
Au	13.271724000	4.252629000	4.819360000
Au	10.317936000	9.363265000	4.817605000
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Fe	3.793534000	8.093076000	9.181919000
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N	4.061755000	9.508790000	7.932863000
N	5.701830000	7.614136000	9.160563000
N	3.705210000	6.729590000	10.592758000
N	3.564002000	6.622818000	7.942995000
C	1.596069000	9.609265000	9.980730000
C	2.678279000	10.088057000	10.769382000
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C	-0.371947000	8.669099000	8.344108000
C	-0.676334000	9.724281000	9.186824000
C	0.325524000	10.224639000	10.049876000

C	0.156002000	11.311366000	10.970574000
C	1.189052000	11.752865000	11.754631000
C	2.492617000	11.160559000	11.673738000
C	3.622832000	11.579830000	12.409664000
C	4.835711000	10.940070000	12.207026000
C	4.933155000	9.893847000	11.275121000
C	4.154941000	10.597286000	7.483818000
C	5.982460000	6.562166000	9.983122000
C	6.663886000	8.038295000	8.334058000
C	4.903415000	6.104080000	10.787593000
C	2.657985000	6.327633000	11.319868000
C	3.844590000	5.508592000	7.648833000
C	7.240331000	5.919924000	10.032187000
C	7.939638000	7.449083000	8.305981000
C	5.085684000	5.035215000	11.696420000
C	2.755819000	5.297530000	12.269342000
C	8.237553000	6.393692000	9.149953000
C	7.404796000	4.832957000	10.953361000
C	6.377631000	4.416096000	11.758163000
C	3.964108000	4.645576000	12.459886000
S	4.296390000	12.111431000	6.857576000
S	4.237775000	3.977395000	7.266839000
H	1.181187000	7.278949000	7.693419000
H	-1.108130000	8.271210000	7.644961000
H	-1.667677000	10.179691000	9.171994000
H	-0.821593000	11.792435000	11.030024000
H	1.036929000	12.582359000	12.446831000
H	3.537146000	12.404167000	13.119190000
H	5.729047000	11.245372000	12.750279000
H	5.881882000	9.395730000	11.079772000
H	6.407592000	8.853022000	7.657573000
H	1.710586000	6.830027000	11.128475000

H	8.670208000	7.827557000	7.590456000
H	1.868108000	5.013477000	12.833066000
H	9.219384000	5.919230000	9.122387000
H	8.373749000	4.333117000	10.997395000
H	6.524392000	3.587313000	12.452590000
H	4.051121000	3.829466000	13.178964000