SUPPORTING INFORMATION FOR

Scrutinizing Low-Spin Cr(II) Complexes

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Figure S1. Temperature dependence of the magnetic moment of $[Cr(phen)_3][PF_6]_2$ (**1**) and $[CrCl_2(^t bpy)_2]$ (**3**) in a 1T magnetic field. Solid lines represent best fits of the experimental data.

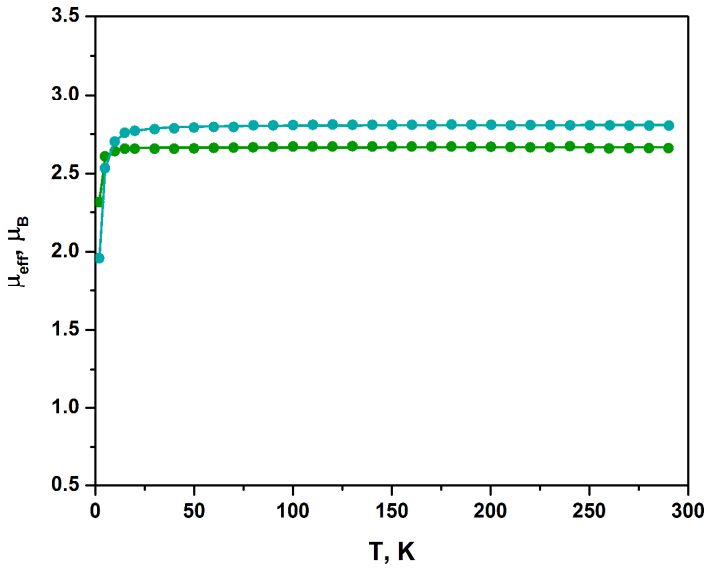

Table S1. Fitting Parameters for Magnetic Susceptibility Data for Complexes **1** and **3**.

	1	**3**
g	1.972	1.885
$\|D\|$ (cm^{-1})	5.623	2.732
TIP (10^{-6} cm^3 mol^{-1})	340.6	450.0
mass (mg)	55.9	31.4
MW (g mol^{-1})	882.55	659.71
χ_{dia} (10^{-6} emu)	-529.0	–463.0

Geometry-optimized molecular xyz coordinates

1, S = 1, BS(3,1), COSMO(water)

Cr	-0.054560	-0.081373	-0.030800
N	-0.046574	-0.099677	2.024377
C	1.205126	0.088581	2.540636
C	1.451491	0.201792	3.932915
C	0.341174	0.097771	4.797593
C	-0.924200	-0.114563	4.257667
C	-1.077337	-0.206973	2.866226
N	1.953708	0.009593	0.285436
C	2.278896	0.151345	1.609559
C	3.609166	0.330971	2.063980
C	4.632531	0.352236	1.090807
C	4.290844	0.192234	-0.249832
C	2.946949	0.028438	-0.614284
C	3.838963	0.466301	3.479174
C	2.806098	0.403540	4.375494
H	0.485908	0.181796	5.877334
H	-1.804381	-0.204779	4.896262
H	-2.061342	-0.360095	2.421199
H	5.672283	0.488348	1.396788
H	5.052849	0.197200	-1.031193
H	2.661453	-0.088386	-1.659588
H	4.865487	0.613273	3.823363
H	2.993658	0.499537	5.447624
N	-0.082265	1.951673	-0.125456
C	0.188409	2.428277	-1.381600
C	0.330548	3.808272	-1.670678
C	0.170601	4.717385	-0.601679
C	-0.121428	4.221380	0.666314
C	-0.235829	2.838644	0.867114
C	0.304907	1.462018	-2.420054
N	0.143155	0.152296	-2.063881
C	0.199273	-0.783147	-3.014233
C	0.437553	-0.471688	-4.361790
C	0.627577	0.855178	-4.736880
C	0.559959	1.867241	-3.755141
C	0.722863	3.270990	-4.026972
C	0.613653	4.200174	-3.027419
H	0.929644	3.579561	-5.054479
H	0.732273	5.264543	-3.244150
H	0.272883	5.790164	-0.780113
H	-0.258173	4.891458	1.516898
H	-0.452219	2.433920	1.856227
H	0.824542	1.122442	-5.777870
H	0.477990	-1.278873	-5.094878
H	0.062411	-1.816602	-2.693869
N	-2.119256	-0.332084	-0.181833
C	-2.487082	-1.641608	-0.243899
C	-3.837351	-2.053167	-0.337524
C	-4.821473	-1.034046	-0.365581
C	-4.429332	0.293527	-0.304355
C	-3.057992	0.605965	-0.215226
C	-1.439082	-2.620634	-0.209527
N	-0.160214	-2.161905	-0.119868
C	0.839039	-3.035545	-0.088965
C	0.623409	-4.426564	-0.152429
C	-0.671777	-4.910587	-0.241893
C	-1.755335	-3.998185	-0.272081
C	-4.135442	-3.458926	-0.396628
C	-3.135271	-4.393301	-0.365290
H	-5.877432	-1.306066	-0.435213
H	-5.160252	1.103373	-0.324442
H	-2.717517	1.641229	-0.166453
H	-0.869882	-5.984047	-0.290424
H	1.481178	-5.100466	-0.127409
H	1.847063	-2.624892	-0.012887
H	-5.181791	-3.764303	-0.467781
H	-3.368115	-5.459592	-0.411396

2, $S = {}^{3}/_{2}$, UKS, COSMO(water)

Cr	0.001423	-0.015729	-0.023920
N	-0.035342	-0.028185	2.035371
C	1.218866	0.003692	2.569179
C	1.451358	0.000058	3.963455
C	0.313830	-0.041422	4.806352
C	-0.952818	-0.076993	4.244922
C	-1.089577	-0.070560	2.844877
N	2.031278	0.042070	0.325226
C	2.320151	0.044300	1.658212
C	3.645077	0.082466	2.148690
C	4.686363	0.122510	1.188952
C	4.373003	0.123296	-0.161345
C	3.023928	0.083462	-0.558853
C	3.860746	0.078292	3.569844
C	2.806333	0.038666	4.442183
H	0.447411	-0.044524	5.890738
H	-1.849630	-0.109302	4.864973
H	-2.075017	-0.096283	2.378959
H	5.726270	0.152051	1.522517
H	5.150192	0.153680	-0.926076
H	2.750447	0.081115	-1.613860
H	4.888693	0.107637	3.937103
H	2.976331	0.035685	5.521046
N	-0.062414	2.039124	-0.156850
C	0.078410	2.471747	-1.442483
C	0.088512	3.842873	-1.786351
C	-0.054599	4.773553	-0.727949
C	-0.194928	4.314556	0.572480
C	-0.193008	2.929911	0.821950
C	0.220556	1.476104	-2.458905
N	0.209898	0.170900	-2.063730
C	0.335762	-0.789284	-2.975128

C	0.487916	-0.500450	-4.343786
C	0.506341	0.820258	-4.764296
C	0.369409	1.859591	-3.811369
C	0.376881	3.259012	-4.140720
C	0.241484	4.212365	-3.167382
H	0.493779	3.544082	-5.188409
H	0.248071	5.274241	-3.422271
H	-0.052693	5.843657	-0.947948
H	-0.307647	5.003805	1.410338
H	-0.301933	2.543810	1.835819
H	0.626248	1.070621	-5.820940
H	0.591427	-1.324491	-5.050884
H	0.321788	-1.819406	-2.618428
N	-2.036732	-0.242797	-0.207062
C	-2.402381	-1.555919	-0.253643
C	-3.750066	-1.966063	-0.370134
C	-4.729890	-0.945470	-0.443749
C	-4.338313	0.383548	-0.400293
C	-2.972313	0.699415	-0.281482
C	-1.359778	-2.531492	-0.177179
N	-0.079766	-2.073442	-0.071068
C	0.922128	-2.944620	0.005240
C	0.703829	-4.334106	-0.026033
C	-0.590129	-4.818866	-0.135591
C	-1.673264	-3.909349	-0.215300
C	-4.048437	-3.371900	-0.406660
C	-3.050274	-4.306061	-0.331788
H	-5.784381	-1.216441	-0.533987
H	-5.066622	1.193679	-0.455376
H	-2.638432	1.736420	-0.242217
H	-0.786442	-5.893256	-0.161977
H	1.560249	-5.006688	0.037153
H	1.930416	-2.538381	0.090188
H	-5.093091	-3.677334	-0.495645
H	-3.282280	-5.372849	-0.359662

$[CrCl_2(bpy)_2]^0$, S = 1, BS(3,1)

Cr	0.163538	-0.001834	0.160750
N	0.017805	0.113785	-1.906246
C	-0.195770	1.364667	-2.415320
C	-0.483511	1.544101	-3.779583
C	-0.522536	0.444540	-4.627187
C	-0.258086	-0.831924	-4.092108
C	0.003654	-0.943749	-2.734898
N	0.175487	2.046687	-0.166823
C	-0.067278	2.445190	-1.450611
C	-0.141051	3.813651	-1.773254
C	0.066128	4.763473	-0.782141
C	0.352307	4.332480	0.527030
C	0.396885	2.968118	0.785529
H	-0.668988	2.546914	-4.165830
H	-0.746219	0.572093	-5.688763
H	-0.252065	-1.724272	-4.720868
H	0.216495	-1.909400	-2.276560
H	-0.342775	4.119571	-2.800239
H	0.018284	5.828591	-1.021756
H	0.537548	5.042296	1.335024
H	0.598994	2.566351	1.779400
N	-0.164998	-2.049690	0.172845
C	-1.448523	-2.448387	-0.070977
C	-1.771619	-3.816875	-0.141907
C	-0.781477	-4.766648	0.069767
C	0.527366	-4.335413	0.357370
C	0.786468	-2.971102	0.398572
C	-2.412732	-1.367939	-0.202502
N	-1.903852	-0.116969	0.011337
C	-2.732472	0.940588	-0.005084
C	-4.089172	0.828730	-0.269394
C	-4.623885	-0.447837	-0.534194
C	-3.776468	-1.547402	-0.492859
H	-4.162523	-2.550242	-0.678618
H	-5.685017	-0.575454	-0.759945
H	-4.717887	1.721118	-0.265094
H	-2.274658	1.906414	0.208067
H	-2.798410	-4.122884	-0.344516
H	-1.021555	-5.831769	0.024568
H	1.334628	-5.045035	0.546451
H	1.780125	-2.569318	0.601643
Cl	2.469527	-0.147692	0.009208
Cl	0.010265	0.143435	2.466277

$[CrCl_2(bpy)_2]^0$, S = 1, BS(3,1), COSMO(water)

Cr	0.103050	-0.042628	0.131870
N	0.102189	-0.348654	-1.892203
C	0.265232	0.613854	-2.822434
C	0.065736	0.388699	-4.176721
C	-0.331175	-0.892642	-4.602136
C	-0.479418	-1.895703	-3.650122
C	-0.243914	-1.609332	-2.295853
C	-0.306488	-2.594607	-1.225615
C	-0.551824	-3.964675	-1.424045
C	-0.522869	-4.829492	-0.337072
C	-0.240840	-4.309027	0.939593
C	-0.021983	-2.944495	1.071022
N	-0.065790	-2.099224	0.024150
H	0.567552	1.593451	-2.455617
H	0.215116	1.204897	-4.886011
H	-0.514660	-1.101220	-5.658338
H	-0.772404	-2.902446	-3.949608
H	-0.752845	-4.342136	-2.426803
H	-0.710612	-5.896416	-0.477427
H	-0.198018	-4.951514	1.820911
H	0.180770	-2.485998	2.039003
N	-1.909094	0.212413	-0.140029
C	-2.831739	-0.768450	-0.070199
C	-4.154686	-0.580625	-0.442795
C	-4.553861	0.680866	-0.921924
C	-3.612017	1.702823	-0.976465

C	-2.292159	1.454516	-0.566754
C	-1.243683	2.463772	-0.518988
C	-1.436779	3.821366	-0.829295
C	-0.381634	4.713367	-0.683305
C	0.857352	4.232656	-0.220384
C	0.987934	2.878035	0.053290
N	-0.025875	2.006105	-0.102718
H	-2.485108	-1.731842	0.300586
H	-4.859886	-1.410041	-0.362341
H	-3.893964	2.695163	-1.329980
H	-2.411685	4.168729	-1.172466
H	1.711131	4.898043	-0.080228
H	1.930333	2.448994	0.394265
H	-5.583189	0.860064	-1.239949
H	-0.518343	5.771044	-0.919670
Cl	-0.137975	0.114786	2.493690
Cl	2.478834	-0.146114	0.219220

4, $S = {}^{3}/_{2}$, UKS, COSMO(water)

Cr	0.108456	-0.000511	0.106451
N	-1.979456	-0.110225	0.020270
C	-2.484967	-1.365261	-0.035065
C	-3.863531	-1.581681	-0.114622
C	-4.725938	-0.484006	-0.132161
C	-4.188328	0.802780	-0.065257
C	-2.803710	0.942873	0.010917
N	-0.193944	-2.061499	0.096821
C	-1.489513	-2.454380	0.011042
C	-1.828621	-3.809599	-0.021192
C	-0.814998	-4.768150	0.045244
C	0.512635	-4.346817	0.145078
C	0.780039	-2.978373	0.168094
H	-4.261635	-2.594359	-0.159360
H	-5.805364	-0.637625	-0.194396
H	-4.821095	1.691581	-0.070080
H	-2.345329	1.928697	0.071112
H	-2.870808	-4.117131	-0.095311
H	-1.064750	-5.831019	0.020319
H	1.337661	-5.058482	0.203794
H	1.796681	-2.593330	0.239433
N	0.099017	2.060682	-0.194322
C	0.008514	2.454415	-1.489316
C	-0.026929	3.809812	-1.827285
C	0.040664	4.767683	-0.813078
C	0.145126	4.345498	0.513926
C	0.171586	2.976880	0.780171
C	-0.039664	1.365932	-2.485411
N	0.018565	0.110595	-1.980924
C	0.008177	-0.942051	-2.805757
C	-0.072333	-0.801240	-4.190039
C	-0.143176	0.485809	-4.726586
C	-0.124526	1.583031	-3.863585
H	-0.104085	4.117978	-2.869055
H	0.012985	5.830699	-1.061905
H	0.204785	5.056628	1.339348
H	0.246500	2.591176	1.796308
H	-0.172919	2.595855	-4.260916
H	-0.209660	0.639957	-5.805678
H	-0.077952	-1.689599	-4.823410
H	0.071120	-1.927991	-2.347962
Cl	-0.010066	0.049016	2.424143
Cl	2.425813	-0.051646	-0.016657

5, $S = 1$, BS(3,1)

Cr	3.553925	-0.002248	8.439680
N	1.772754	-1.019462	8.581656
N	4.368991	-1.552489	7.395987
C	0.613403	-0.584376	9.192626
C	1.516615	-2.281967	8.100673
C	3.753534	-2.754220	7.104412
C	5.671835	-1.651310	6.932498
C	-0.389615	-1.595969	9.092988
C	0.166656	-2.647793	8.400626
C	4.704349	-3.638872	6.456485
C	5.877114	-2.963006	6.351115
C	2.439853	-3.110877	7.408201
C	0.444796	0.670531	9.836925
C	1.965103	-4.463089	6.987831
C	1.584597	-5.422486	7.943725
C	1.142743	-6.689814	7.552308
C	1.074162	-7.023291	6.193999
C	1.450660	-6.078648	5.232362
C	1.889308	-4.810401	5.627236
C	-0.893730	0.962686	10.429687
C	-1.035856	1.185690	11.810725
C	-2.287478	1.465107	12.368278
C	-3.423134	1.523774	11.552797
C	-3.296373	1.301488	10.176495
C	-2.043518	1.023739	9.621987
N	5.335051	1.015025	8.297587
N	2.738916	1.547937	9.483474
C	6.663109	-0.675066	7.042536
C	1.436166	1.646635	9.947247
C	6.494332	0.580036	7.686420
C	5.591069	2.277682	8.778234
C	3.354398	2.749625	9.775163
C	8.001692	-0.967275	6.449934
C	1.231021	2.958154	10.529089
C	7.497143	1.591900	7.785491
C	6.940816	2.643795	8.477703
C	4.667934	3.106450	9.470996
C	2.403769	3.634043	10.423679
C	8.143898	-1.190912	5.069003
C	9.151477	-1.027756	7.257688
C	5.142653	4.458692	9.891285
C	9.395581	-1.470389	4.511617
C	10.404391	-1.305568	6.703347
C	5.522680	5.418210	8.935326

C	5.218910	4.805916	11.251874
C	10.531225	-1.528487	5.327154
C	5.964517	6.685566	9.326668
C	5.657531	6.074192	11.646680
C	6.033554	7.018958	10.684975
H	-1.394698	-1.523352	9.499801
H	-0.304246	-3.586144	8.121008
H	4.488795	-4.657820	6.145733
H	6.818032	-3.316997	5.937460
H	1.641868	-5.161858	9.003245
H	0.855431	-7.422934	8.311486
H	0.728719	-8.014545	5.887027
H	1.397097	-6.328084	4.168753
H	2.179220	-4.069727	4.877826
H	-0.148680	1.138039	12.447283
H	-2.377362	1.633952	13.445166
H	-4.402879	1.741210	11.987080
H	-4.177391	1.348628	9.530046
H	-1.943304	0.848718	8.548140
H	0.290206	3.312000	10.943103
H	8.502115	1.519422	7.378383
H	7.411569	3.582350	8.756893
H	2.619407	4.652883	10.734731
H	7.256737	-1.143700	4.432393
H	9.051209	-0.852243	8.331448
H	9.485519	-1.639726	3.434810
H	11.285397	-1.352261	7.349844
H	5.465062	5.157644	7.875806
H	4.929358	4.065153	12.001339
H	11.511016	-1.745972	4.892998
H	6.251466	7.418771	8.567436
H	5.711444	6.323559	12.710287
H	6.378981	8.010234	10.991891
N	2.918997	0.885855	6.636608
C	3.764015	0.996718	5.598095
C	3.389616	1.587369	4.392703
C	2.088362	2.079054	4.254878
C	1.212969	1.957967	5.337707
C	1.667672	1.357129	6.510151
H	4.764377	0.596523	5.750776
H	4.115513	1.655886	3.579873
H	1.763492	2.547755	3.322118
H	0.186508	2.326348	5.285311
H	1.022895	1.247437	7.379857
N	4.188844	-0.890360	10.242755
C	3.343708	-1.001570	11.281137
C	5.440273	-1.361326	10.369338
C	3.718087	-1.592273	12.486509
C	5.894958	-1.962203	11.541768
C	5.019442	-2.083654	12.624457
H	2.343265	-0.601612	11.128368
H	6.085150	-1.251358	9.499744
H	2.992089	-1.661076	13.299226
H	6.921504	-2.330335	11.594261
H	5.344296	-2.552398	13.557201

[Cr(CNMe)$_6$]$^{2+}$, *S* = 1, UKS, COSMO(water)

Cr	-0.001305	0.009818	0.003087
C	2.052481	0.049006	0.053989
C	-2.055012	-0.030007	-0.049438
C	-0.037383	-0.025562	-2.050474
C	0.036050	0.045352	2.056472
C	0.034528	2.060958	0.037584
C	-0.036321	-2.041297	-0.032547
N	3.208085	0.083352	0.091706
N	-3.210477	-0.065241	-0.090723
N	-0.079688	-0.054322	-3.206148
N	0.080322	0.074709	3.212067
N	0.069335	3.217097	0.067812
N	-0.070193	-3.197431	-0.064009
C	0.116128	4.638076	0.108411
C	4.629239	0.126788	0.131402
C	-0.136517	-0.086437	-4.627024
C	0.139393	0.106172	4.632856
C	-0.115380	-4.618410	-0.106048
C	-4.631040	-0.111289	-0.144910
H	1.035491	4.958274	0.621812
H	-0.764734	5.010447	0.653359
H	0.106828	5.029841	-0.920031
H	4.960791	0.154014	1.180429
H	4.974816	1.033056	-0.388950
H	5.034811	-0.767363	-0.365894
H	0.772964	0.377103	-5.038373
H	-1.024099	0.471556	-4.962408
H	-0.208933	-1.131936	-4.962982
H	1.033375	-0.442806	4.966121
H	-0.764491	-0.367058	5.045491
H	0.201471	1.152088	4.969575
H	0.776719	-4.990185	-0.632814
H	-1.023876	-4.938993	-0.638228
H	-0.127222	-5.010398	0.922285
H	-4.999650	0.786338	-0.664352
H	-5.030543	-0.135444	0.880313
H	-4.945962	-1.014423	-0.689414

7, *S* = 1, UKS

Cr	0.000000	-0.000000	0.000000
Cl	-0.077190	0.024959	2.391810
Cl	0.077191	-0.024960	-2.391810
P	0.262480	2.375582	0.056491
P	2.389510	0.013565	-0.003444
C	-0.582726	3.405545	1.311743
C	-0.025669	3.306781	-1.493803
C	2.051519	2.741734	0.403093
C	3.252839	-0.345400	1.570858
C	3.354022	-0.949025	-1.225948
C	2.939572	1.751630	-0.363043
P	-0.262480	-2.375582	-0.056491
P	-2.389509	-0.013565	0.003444

C	0.582726	-3.405546	-1.311742
C	0.025667	-3.306782	1.493803
C	-2.051520	-2.741734	-0.403094
C	-3.252838	0.345401	-1.570858
C	-3.354021	0.949026	1.225948
C	-2.939572	-1.751630	0.363042
H	-1.652116	3.482453	1.060877
H	-0.488439	2.907874	2.288658
H	-0.151110	4.419860	1.357595
H	0.246518	4.369980	-1.380738
H	-1.088503	3.225147	-1.770339
H	0.557782	2.844597	-2.303353
H	2.291153	3.789125	0.150052
H	2.190390	2.617590	1.490745
H	3.054865	-1.389651	1.859125
H	2.836482	0.294818	2.362200
H	4.340551	-0.184951	1.478407
H	4.404010	-0.612761	-1.267189
H	2.878702	-0.831355	-2.211621
H	3.330275	-2.015018	-0.950345
H	4.007593	1.883660	-0.116297
H	2.823957	1.895159	-1.450992
H	1.652116	-3.482454	-1.060875
H	0.488439	-2.907876	-2.288658
H	0.151110	-4.419861	-1.357594
H	-0.246519	-4.369981	1.380738
H	1.088501	-3.225147	1.770341
H	-0.557785	-2.844598	2.303353
H	-2.291154	-3.789125	-0.150054
H	-2.190390	-2.617589	-1.490746
H	-3.054863	1.389652	-1.859124
H	-2.836481	-0.294817	-2.362200
H	-4.340550	0.184953	-1.478407
H	-4.404010	0.612762	1.267189
H	-2.878701	0.831355	2.211622
H	-3.330274	2.015018	0.950345
H	-4.007593	-1.883659	0.116296
H	-2.823957	-1.895160	1.450991

8, $S = 1$, UKS

Cr	-0.000000	0.000000	0.000000
C	-1.502421	0.557679	-1.553354
C	-1.598751	1.416996	-0.428398
C	-0.345777	2.090126	-0.278365
C	-0.206252	0.684133	-2.102402
C	1.502421	-0.557679	1.553354
C	1.598751	-1.416996	0.428398
C	0.345777	-2.090126	0.278365
C	-0.519266	-1.628117	1.322756
C	0.206252	-0.684133	2.102403
C	0.519266	1.628117	-1.322756
H	-2.266429	-0.124705	-1.893468
H	-2.463281	1.531150	0.206594
H	-0.107254	2.833425	0.465644
H	1.536648	1.942350	-1.495063
H	0.181960	0.121609	-2.937628
H	2.266429	0.124705	1.893468
H	2.463281	-1.531150	-0.206594
H	0.107254	-2.833425	-0.465644
H	-1.536648	-1.942350	1.495063
H	-0.181960	-0.121609	2.937628

Table S1. Calculated Cr 3d and 4p contributions to orbitals excited into in calculated X-ray absorption spectra.

	1		**2**		**3**		**4**		**5**		$[Cr(CNMe)_6]^{2+}$		**7**		**8**	
	% Cr 3d	% Cr 4p	% Cr 3d	% Cr 4p	% Cr 3d	% Cr 4p	% Cr 3d	% Cr 4p	% Cr 3d	% Cr 4p	% Cr 3d	% Cr 4p	% Cr 3d	% Cr 4p	% Cr 3d	% Cr 4p
e_g (β)	67.9	2.2	71.9	0.1	63.3	0.2	72.1	0.2	43.9	0.0	63.8	0.0	38.0	0.0	65.0	0.0
	66.2	3.5	72.4	0.1	62.3	0.9	72.9	0.0	54.8	0.0	62.7	0.0	45.2	0.0	22.6	0.0
e_g(α)	69.8	0.0	67.1	0.0	68.3	0.0	67.8	0.0	65.3	0.0	49.3	0.0	64.3	0.0	61.5	0.0
	64.7	0.1	67.4	0.0	57.5	0.2	65.5	0.2	66.6	0.0	51.6	0.0	55.4	0.0	61.2	0.0
t_{2g} (β)	21.8	0.3	33.0	0.2	27.6	0.2	34.1	0.1	86.1	0.0	47.8	0.0	54.7	0.0	67.2	0.0
	42.3	0.0	33.0	0.2	32.7	0.1	24.1	0.1	35.9	0.0	47.7	0.0	65.1	0.0	47.4	0.0
	31.1	0.1	38.4	0.0	38.2	0.2	42.1	0.2	27.9	0.0						