

Benjamin Cahier, Rémi Maurice, Hélène Bolvin, Talal Mallah and Nathalie Guihéry

2Å	2.1Å	2.2Å	2.3Å	2.4Å	2.5Å	2.6Å	2.7Å	2.8Å	2.9Å	3.0Å	3.1Å	3.2Å	3.3Å	3.4Å	3.5Å
0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
59.369	77.6	98.2	119.645	65.822	35.298	92.243	119.198	128.221	125.902	117.086	107.011	95.882	85.379	75.718	66.86
59.369	77.6	98.2	119.645	65.822	35.298	92.243	119.198	128.221	125.902	117.086	107.011	95.882	85.379	75.718	66.86
1376.968	851.706	482.413	230.385	138.996	187.885	249.5	271.372	267.058	246.637	218.543	189.494	161.951	138.008	117.435	99.906
1376.968	851.706	482.413	230.385	138.996	187.885	249.5	271.372	267.058	246.637	218.543	189.494	161.951	138.008	117.435	99.906
1860.772	1338.594	968.352	711.838	539.173	461.108	443.839	420.625	390.065	352.681	310.481	265.583	223.065	185.764	153.726	127.053
1860.772	1338.594	968.352	711.838	539.173	461.108	443.839	420.625	390.065	352.681	310.481	265.583	223.065	185.764	153.726	127.053
2356.49	1836.916	1467.251	1208.666	1029.081	931.814	862.215	746.83	619.864	505.652	409.269	328.762	264.682	214.752	175.147	143.69
2356.49	1836.916	1467.251	1208.666	1029.081	931.814	862.215	746.83	619.864	505.652	409.269	328.762	264.682	214.752	175.147	143.69
2854.65	2343.436	1979.771	1725.514	1546.924	1251.171	1098.201	1023.916	984.241	955.163	930.004	907.118	887.001	869.939	855.421	843.113
2854.65	2343.436	1979.771	1725.514	1546.924	1251.171	1098.201	1023.916	984.241	955.163	930.004	907.118	887.001	869.939	855.421	843.113
4503.075	3467.118	2646.69	2017.617	1550.49	1439.274	1348.734	1248.861	1162.5	1091.709	1033.469	985.735	947.302	917.048	883.388	858.102
4503.075	3467.118	2646.69	2017.617	1550.49	1439.274	1348.734	1248.861	1162.5	1091.709	1033.469	985.735	947.302	917.048	883.388	858.102
4712.203	3694.81	2896.958	2296.941	1868.654	1629.144	1509.779	1366.633	1250.992	1158.978	1085.584	1026.366	970.814	919.195	892.654	872.985
4712.203	3694.81	2896.958	2296.941	1868.654	1629.144	1509.779	1366.633	1250.992	1158.978	1085.584	1026.366	970.814	919.195	892.654	872.985
4882.407	3863.547	3058.629	2443.983	1988.425	1691.65	1553.551	1524.266	1505.578	1336.461	1166.098	1048.607	979.653	943.292	914.324	890.972
4882.407	3863.547	3058.629	2443.983	1988.425	1691.65	1553.551	1524.266	1505.578	1336.461	1166.098	1048.607	979.654	943.292	914.324	890.972
5082.169	4088.151	3315.827	2742.596	2336.176	2093.363	1965.834	1872.411	1585.356	1489.961	1475.684	1462.807	1451.369	1441.616	1433.186	1425.834
5082.169	4088.151	3315.827	2742.596	2336.176	2093.363	1965.834	1872.411	1585.356	1489.961	1475.684	1462.807	1451.369	1441.616	1433.186	1425.834
14029.288	10235.753	7495.928	5515.887	4091.047	3102.588	2428.232	1940.411	1799.193	1738.817	1685.7					

14715.631	10943.794	8225.682	6269.528	4872.222	3915.895	3278.204	2829.678	2513.837	2299.214	2155.046	2055.966	1989.488	1944.458	1912.498	1889.365
15026.883	11252.601	8527.656	6560.136	5146.468	4168.227	3502.976	3022.409	2671.919	2424.335	2252.345	2131.684	2050.019	1994.568	1955.092	1926.195
15026.883	11252.601	8527.656	6560.136	5146.468	4168.227	3502.976	3022.409	2671.919	2424.335	2252.345	2131.684	2050.019	1994.568	1955.092	1926.195

Table S2. ($^4\text{F} + ^4\text{P}$) SO spectrum (cm^{-1}).

2Å	2.1Å	2.2Å	2.3Å	2.4Å	2.5Å	2.6Å	2.7Å	2.8Å	2.9Å	3.0Å	3.1Å	3.2Å	3.3Å	3.4Å	3.5Å
0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
60.494	78.335	98.654	119.915	66.11	35.131	92.147	119.143	128.19	125.884	117.075	107.004	95.878	85.377	75.717	66.859
60.494	78.335	98.654	119.915	66.11	35.131	92.147	119.143	128.19	125.884	117.075	107.004	95.878	85.377	75.717	66.859
1378.968	853.02	483.231	230.877	139.152	187.807	249.454	271.346	267.043	246.628	218.538	189.491	161.949	138.007	117.434	99.906
1378.968	853.02	483.231	230.877	139.152	187.807	249.454	271.346	267.043	246.628	218.538	189.491	161.949	138.007	117.434	99.906
1862.772	1339.909	969.171	712.329	539.462	461.108	443.839	420.625	390.065	352.681	310.481	265.583	223.065	185.764	153.726	127.053
1862.772	1339.909	969.171	712.329	539.462	461.108	443.839	420.625	390.065	352.681	310.481	265.583	223.065	185.764	153.726	127.053
2358.49	1838.23	1468.068	1209.156	1029.368	931.812	862.213	746.828	619.862	505.651	409.268	328.761	264.681	214.752	175.146	143.69
2358.49	1838.23	1468.068	1209.156	1029.368	931.812	862.213	746.828	619.862	505.651	409.268	328.761	264.681	214.752	175.146	143.69
2856.65	2344.75	1980.588	1726.004	1547.21	1251.171	1098.201	1023.916	984.241	955.163	930.004	907.118	887.001	869.939	855.421	843.113
2856.65	2344.75	1980.588	1726.004	1547.21	1251.171	1098.201	1023.916	984.241	955.163	930.004	907.118	887.001	869.939	855.421	843.113
4505.074	3468.431	2647.508	2018.107	1550.778	1439.268	1348.726	1248.856	1162.498	1091.709	1033.468	985.735	947.302	917.048	883.388	858.102
4505.074	3468.431	2647.508	2018.107	1550.778	1439.268	1348.726	1248.856	1162.498	1091.709	1033.468	985.735	947.302	917.048	883.388	858.102
4713.568	3695.744	2897.557	2297.311	1868.877	1629.114	1509.775	1366.63	1250.991	1158.977	1085.583	1026.366	970.814	919.195	892.654	872.985
4713.568	3695.744	2897.557	2297.311	1868.877	1629.114	1509.775	1366.63	1250.991	1158.977	1085.583	1026.366	970.814	919.195	892.654	872.985
4883.96	3864.643	3059.347	2444.43	1988.694	1691.641	1553.54	1524.261	1505.575	1336.461	1166.098	1048.607	979.653	943.292	914.324	890.972
4883.96	3864.643	3059.347	2444.43	1988.694	1691.641	1553.54	1524.261	1505.575	1336.461	1166.098	1048.607	979.653	943.292	914.324	890.972
5083.074	4088.749	3316.196	2742.816	2336.304	2093.272	1965.783	1872.383	1585.356	1489.958	1475.682	1462.806	1451.368	1441.615	1433.186	1425.834
5083.074	4088.749	3316.196	2742.816	2336.304	2093.272	1965.783	1872.383	1585.356	1489.958	1475.682	1462.806	1451.368	1441.615	1433.186	1425.834
14031.288	10237.068	7496.746	5516.379	4091.335	3102.588	2428.232	1940.411	1799.178	1738.809	1685.744	1637.956	1586.936	1536.235	1499.763	1473.119
14031.288	10237.068	7496.746	5516.379	4091.335	3102.588	2428.232	1940.411	1799.178	1738.809	1685.744	1637.956	1586.936	1536.235	1499.763	1473.119
14384.269	10605.883	7882.711	5922.393	4521.402	3561.279	2919.537	2465.851	2143.225	1920.502	1767.833	1660.486	1595.631	1559.768	1529.444	1504.175
14384.269	10605.883	7882.711	5922.393	4521.402	3561.279	2919.537	2465.851	2143.225	1920.502	1767.833	1660.486	1595.631	1559.768	1529.444	1504.175
14716.35	10944.436	8226.14	6269.821	4872.399	3915.83	3278.166	2829.655	2513.823	2299.206	2155.041	2055.963	1989.486	1944.456	1912.497	1889.365
14716.35	10944.436	8226.14	6269.821	4872.399	3915.83	3278.166	2829.655	2513.823	2299.206	2155.041	2055.963	1989.486	1944.456	1912.497	1889.365
15027.903	11253.402	8528.198	6560.475	5146.669	4168.175	3502.944	3022.389	2671.905	2424.326	2252.338	2131.68	2050.017	1994.565	1955.091	1926.194
15027.903	11253.402	8528.198	6560.475	5146.669	4168.175	3502.944	3022.389	2671.905	2424.326	2252.338	2131.68	2050.017	1994.565	1955.091	1926.194

Table S3. ($^4F + ^2G$) SO spectrum (cm^{-1}).

2Å	2.1Å	2.2Å	2.3Å	2.4Å	2.5Å	2.6Å	2.7Å	2.8Å	2.9Å	3.0Å	3.1Å	3.2Å	3.3Å	3.4Å	3.5Å
0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
77.954	94.484	113.45	133.115	67.117	35.041	92.43	119.465	128.407	125.968	117.05	106.9	95.729	85.207	75.542	66.689
77.954	94.484	113.45	133.115	67.117	35.041	92.43	119.465	128.407	125.968	117.05	106.9	95.729	85.207	75.542	66.689
1385.522	858.952	487.595	233.402	150.505	196.982	256.781	276.618	270.44	248.61	219.583	189.938	162.066	137.952	117.295	99.734
1385.522	858.952	487.595	233.402	150.505	196.982	256.781	276.618	270.44	248.61	219.583	189.938	162.066	137.952	117.295	99.734
1876.265	1353.422	982.136	724.594	551.094	472.056	454.405	430.114	397.882	358.522	314.427	267.979	224.409	186.47	154.06	127.18
1876.265	1353.422	982.136	724.594	551.094	472.056	454.405	430.114	397.882	358.522	314.427	267.979	224.409	186.47	154.06	127.18
2367.762	1851.574	1483.332	1225.298	1045.135	944.223	867.64	747.848	619.762	505.213	408.703	328.172	264.125	214.252	174.71	143.315
2367.762	1851.574	1483.332	1225.298	1045.135	944.223	867.64	747.848	619.762	505.213	408.703	328.172	264.125	214.252	174.71	143.315
2851.119	2349.936	1991.606	1740.716	1558.431	1264.194	1120.033	1050.786	1012.237	983.283	958.037	935.044	914.844	897.73	883.187	870.869
2851.119	2349.936	1991.606	1740.716	1558.431	1264.194	1120.033	1050.786	1012.237	983.283	958.037	935.044	914.844	897.73	883.187	870.869
4485.484	3464.318	2649.457	2023.317	1564.045	1454.358	1365.334	1268.844	1185.153	1116.166	1059.088	1012.123	974.168	944.212	910.154	885.229
4485.484	3464.318	2649.457	2023.317	1564.045	1454.358	1365.334	1268.844	1185.153	1116.166	1059.088	1012.123	974.168	944.212	910.154	885.229
4722.387	3707.369	2911.137	2313.624	1889.729	1657.109	1534.5	1392.353	1277.361	1185.758	1112.632	1053.606	996.1	945.388	920.013	900.474
4722.387	3707.369	2911.137	2313.624	1889.729	1657.109	1534.5	1392.353	1277.361	1185.758	1112.632	1053.606	996.1	945.388	920.013	900.474
4906.262	3885.38	3079.835	2465.497	2010.79	1714.846	1586.351	1558.479	1540.645	1355.617	1187.878	1072.467	1007.027	970.763	941.868	918.572
4906.262	3885.38	3079.835	2465.497	2010.79	1714.846	1586.351	1558.479	1540.645	1355.617	1187.878	1072.467	1007.027	970.763	941.868	918.572
5110.992	4117.717	3345.651	2772.732	2366.977	2124.923	1998.993	1906.73	1601.479	1525.642	1511.798	1499.237	1488.012	1478.404	1470.075	1462.794
5110.992	4117.717	3345.651	2772.732	2366.977	2124.923	1998.993	1906.73	1601.479	1525.642	1511.798	1499.237	1488.012	1478.404	1470.075	1462.794
14000.779	10223.812	7490.932	5515.218	4093.887	3108.457	2437.795	1953.309	1834.339	1774.538	1721.867	1674.371	1622.938	1572.604	1536.376	1509.895
14000.779	10223.812	7490.932	5515.218	4093.887	3108.457	2437.795	1953.309	1834.339	1774.538	1721.867	1674.371	1622.938	1572.604	1536.376	1509.895
14397.723	10620.723	7900.677	5942.398	4543.415	3585.467	2946.582	2495.405	2174.916	1953.862	1802.406	1695.921	1632.252	1596.534	1566.315	1541.122
14397.723	10620.723	7900.677	5942.398	4543.415	3585.467	2946.582	2495.405	2174.916	1953.862	1802.406	1695.921	1632.252	1596.534	1566.315	1541.122
14745.247	10971.082	8254.192	6298.716	4902.198	3946.654	3310.637	2863.441	2548.646	2334.787	2191.136	2092.405	2026.147	1981.258	1949.395	1926.331
14745.247	10971.082	8254.192	6298.716	4902.198	3946.654	3310.637	2863.441	2548.646	2334.787	2191.136	2092.405	2026.147	1981.258	1949.395	1926.331
15046.456	11277.671	8554.813	6588.687	5176.369	4199.286	3535.861	3056.602	2707.048	2460.113	2288.557	2168.194	2086.719	2031.392	1992.003	1963.169
15046.456	11277.671	8554.813	6588.687	5176.369	4199.286	3535.861	3056.602	2707.048	2460.113	2288.557	2168.194	2086.719	2031.392	1992.003	1963.169

Table S4. Difference between the (⁴F) and (⁴F +⁴P) SO spectra (cm⁻¹).

2Å	2.1Å	2.2Å	2.3Å	2.4Å	2.5Å	2.6Å	2.7Å	2.8Å	2.9Å	3.0Å	3.1Å	3.2Å	3.3Å	3.4Å	3.5Å
0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
1.125	0.735	0.454	0.27	0.288	0.167	0.096	0.055	0.031	0.018	0.011	0.007	0.004	0.002	0.001	0.001
1.125	0.735	0.454	0.27	0.288	0.167	0.096	0.055	0.031	0.018	0.011	0.007	0.004	0.002	0.001	0.001
2	1.314	0.818	0.492	0.156	0.078	0.046	0.026	0.015	0.009	0.005	0.003	0.002	0.001	0.001	0
2	1.314	0.818	0.492	0.156	0.078	0.046	0.026	0.015	0.009	0.005	0.003	0.002	0.001	0.001	0
2	1.315	0.819	0.491	0.289	0	0	0	0	0	0	0	0	0	0	0
2	1.315	0.819	0.491	0.289	0	0	0	0	0	0	0	0	0	0	0
2	1.314	0.817	0.49	0.287	0.002	0.002	0.002	0.002	0.001	0.001	0.001	0.001	0	0.001	0
2	1.314	0.817	0.49	0.287	0.002	0.002	0.002	0.002	0.001	0.001	0.001	0.001	0	0.001	0
2	1.314	0.817	0.49	0.286	0	0	0	0	0	0	0	0	0	0	0
2	1.314	0.817	0.49	0.286	0	0	0	0	0	0	0	0	0	0	0
1.999	1.313	0.818	0.49	0.288	0.006	0.008	0.005	0.002	0	0.001	0	0	0	0	0
1.999	1.313	0.818	0.49	0.288	0.006	0.008	0.005	0.002	0	0.001	0	0	0	0	0
1.365	0.934	0.599	0.37	0.223	0.03	0.004	0.003	0.001	0.001	0.001	0	0	0	0	0
1.365	0.934	0.599	0.37	0.223	0.03	0.004	0.003	0.001	0.001	0.001	0	0	0	0	0
1.553	1.096	0.718	0.447	0.269	0.009	0.011	0.005	0.003	0	0	0	0	0	0	0
1.553	1.096	0.718	0.447	0.269	0.009	0.011	0.005	0.003	0	0	0	0.001	0	0	0
0.905	0.598	0.369	0.22	0.128	0.091	0.051	0.028	0	0.003	0.002	0.001	0.001	0.001	0	0
0.905	0.598	0.369	0.22	0.128	0.091	0.051	0.028	0	0.003	0.002	0.001	0.001	0.001	0	0
2	1.315	0.818	0.492	0.288	0	0	0	0.015	0.008	0.004	0.001	0.001	0.001	0	0
2	1.315	0.818	0.492	0.288	0	0	0	0.015	0.008	0.004	0.001	0.001	0.001	0	0
1.065	0.824	0.558	0.351	0.211	0.043	0.024	0.014	0.008	0.005	0.003	0.002	0.001	0.001	0	0
1.065	0.824	0.558	0.351	0.211	0.043	0.024	0.014	0.008	0.005	0.003	0.002	0.001	0.001	0	0
0.719	0.642	0.458	0.293	0.177	0.065	0.038	0.023	0.014	0.008	0.005	0.003	0.002	0.002	0.001	0
0.719	0.642	0.458	0.293	0.177	0.065	0.038	0.023	0.014	0.008	0.005	0.003	0.002	0.002	0.001	0
1.02	0.801	0.542	0.339	0.201	0.052	0.032	0.02	0.014	0.009	0.007	0.004	0.002	0.003	0.001	0.001
1.02	0.801	0.542	0.339	0.201	0.052	0.032	0.02	0.014	0.009	0.007	0.004	0.002	0.003	0.001	0.001

Table S5. Difference between the (4F) and ($^4F + ^2G$) SO spectra (cm^{-1}).

$2\text{\AA}$	$2.1\text{\AA}$	$2.2\text{\AA}$	$2.3\text{\AA}$	$2.4\text{\AA}$	$2.5\text{\AA}$	$2.6\text{\AA}$	$2.7\text{\AA}$	$2.8\text{\AA}$	$2.9\text{\AA}$	$3.0\text{\AA}$	$3.1\text{\AA}$	$3.2\text{\AA}$	$3.3\text{\AA}$	$3.4\text{\AA}$	$3.5\text{\AA}$
0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
18.585	16.884	15.25	13.47	1.295	0.257	0.187	0.267	0.186	0.066	0.036	0.111	0.153	0.172	0.176	0.171
18.585	16.884	15.25	13.47	1.295	0.257	0.187	0.267	0.186	0.066	0.036	0.111	0.153	0.172	0.176	0.171
8.554	7.246	5.182	3.017	11.509	9.097	7.281	5.246	3.382	1.973	1.04	0.444	0.115	0.056	0.14	0.172
8.554	7.246	5.182	3.017	11.509	9.097	7.281	5.246	3.382	1.973	1.04	0.444	0.115	0.056	0.14	0.172
15.493	14.828	13.784	12.756	11.921	10.948	10.566	9.489	7.817	5.841	3.946	2.396	1.344	0.706	0.334	0.127
15.493	14.828	13.784	12.756	11.921	10.948	10.566	9.489	7.817	5.841	3.946	2.396	1.344	0.706	0.334	0.127
11.272	14.658	16.081	16.632	16.054	12.409	5.425	1.018	0.102	0.439	0.566	0.59	0.557	0.5	0.437	0.375
11.272	14.658	16.081	16.632	16.054	12.409	5.425	1.018	0.102	0.439	0.566	0.59	0.557	0.5	0.437	0.375
3.531	6.5	11.835	15.202	11.507	13.023	21.832	26.87	27.996	28.12	28.033	27.926	27.843	27.791	27.766	27.756
3.531	6.5	11.835	15.202	11.507	13.023	21.832	26.87	27.996	28.12	28.033	27.926	27.843	27.791	27.766	27.756
17.591	2.8	2.767	5.7	13.555	15.084	16.6	19.983	22.653	24.457	25.619	26.388	26.866	27.164	26.766	27.127
17.591	2.8	2.767	5.7	13.555	15.084	16.6	19.983	22.653	24.457	25.619	26.388	26.866	27.164	26.766	27.127
10.184	12.559	14.179	16.683	21.075	27.965	24.721	25.72	26.369	26.78	27.048	27.24	25.286	26.193	27.359	27.489
10.184	12.559	14.179	16.683	21.075	27.965	24.721	25.72	26.369	26.78	27.048	27.24	25.286	26.193	27.359	27.489
23.855	21.833	21.206	21.514	22.365	23.196	32.8	34.213	35.067	19.156	21.78	23.86	27.374	27.471	27.544	27.6
23.855	21.833	21.206	21.514	22.365	23.196	32.8	34.213	35.067	19.156	21.78	23.86	27.373	27.471	27.544	27.6
28.823	29.566	29.824	30.136	30.801	31.56	33.159	34.319	16.123	35.681	36.114	36.43	36.643	36.788	36.889	36.96
28.823	29.566	29.824	30.136	30.801	31.56	33.159	34.319	16.123	35.681	36.114	36.43	36.643	36.788	36.889	36.96
28.509	11.941	4.996	-0.669	2.84	5.869	9.563	12.898	35.146	35.721	36.119	36.414	36.001	36.368	36.613	36.776
28.509	11.941	4.996	-0.669	2.84	5.869	9.563	12.898	35.146	35.721	36.119	36.414	36.001	36.368	36.613	36.776
14.519	15.664	18.524	20.356	22.224	24.145	27.021	29.54	31.683	33.355	34.57	35.433	36.62	36.765	36.871	36.947
14.519	15.664	18.524	20.356	22.224	24.145	27.021	29.54	31.683	33.355	34.57	35.433	36.62	36.765	36.871	36.947
29.616	27.288	28.51	29.188	29.976	30.759	32.433	33.763	34.809	35.573	36.09	36.439	36.659	36.8	36.897	36.966
29.616	27.288	28.51	29.188	29.976	30.759	32.433	33.763	34.809	35.573	36.09	36.439	36.659	36.8	36.897	36.966
19.573	25.07	27.157	28.551	29.901	31.059	32.885	34.193	35.129	35.778	36.212	36.51	36.7	36.824	36.911	36.974
19.573	25.07	27.157	28.551	29.901	31.059	32.885	34.193	35.129	35.778	36.212	36.51	36.7	36.824	36.911	36.974

Table S6. $\Delta_{(4F+4P)-(4F)}$ (cm^{-1}).

2Å	2.1Å	2.2Å	2.3Å	2.4Å	2.5Å	2.6Å	2.7Å	2.8Å	2.9Å	3.0Å	3.1Å	3.2Å	3.3Å	3.4Å	3.5Å
39.502	27.03	17.21	10.47	6.182	1.086	0.624	0.362	0.21	0.124	0.08	0.044	0.029	0.022	0.01	0.004

Table S7. $\Delta_{(4F+2G)-(4F)}$ (cm^{-1}).

2Å	2.1Å	2.2Å	2.3Å	2.4Å	2.5Å	2.6Å	2.7Å	2.8Å	2.9Å	3.0Å	3.1Å	3.2Å	3.3Å	3.4Å	3.5Å
460.21	413.674	418.59	425.072	450.046	470.742	508.946	535.038	552.924	565.88	574.346	580.362	584.321	587.196	589.406	590.88

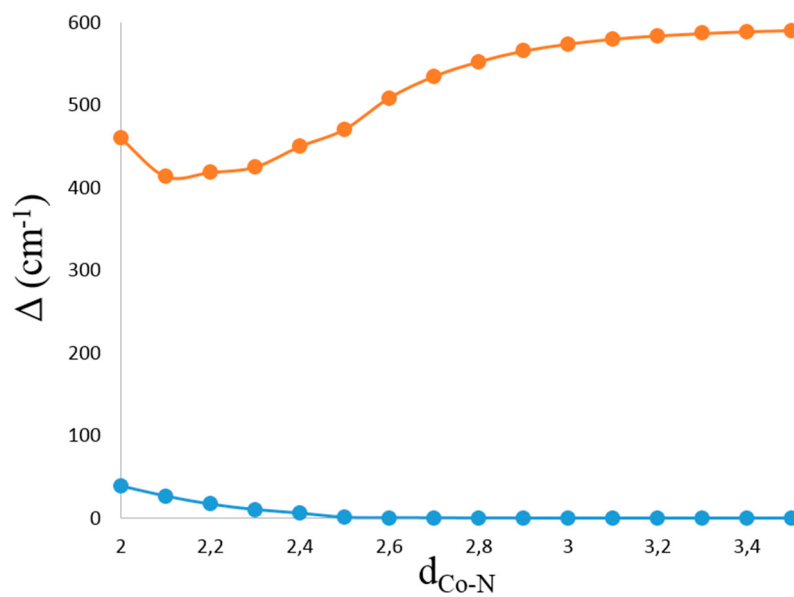


Figure S1. $\Delta(^4F + ^4P) - (^4F)$ (blue) and $\Delta(^4F + ^2G) - (^4F)$ (red) as a function of $d_{\text{Co-N}}$.

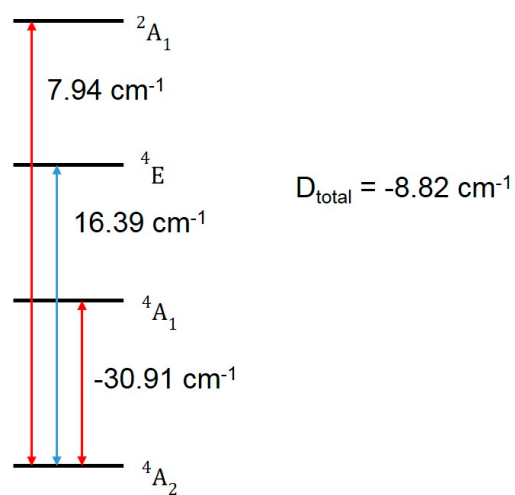


Figure S2. Most important contributions to D for the $[\text{Co}(\text{NCH})_4\text{Cl}]^+$ ($\alpha = 3^\circ$) complex.