Supporting Information

Table 1: Rotational g tensors determined by experiment and calculated using DFT methods. Basis set used is aug-cc-pVTZ.

Molecule	KT2	B3LYP	B97-2	B97-3	O-B3LYP	O-B97-2	O-B97-3	Exp.
Linear molecules								
CO	-0.2673	-0.2811	-0.2827	-0.2861	-0.2704	-0.2717	-0.2719	-0.2689
CS	-0.2782	-0.2923	-0.2932	-0.2989	-0.2730	-0.2740	-0.2728	-0.2702
CSe	-0.2548	-0.2733	-0.2713	-0.2793	-0.2480	-0.2463	-0.2463	-0.2431
$HC^{15}N$	-0.0912	-0.0980	-0.0983	-0.0978	-0.0936	-0.0941	-0.0924	-0.0904
$FC^{15}N$	-0.0481	-0.0515	-0.0499	-0.0502	-0.0509	-0.0493	-0.0495	-0.0504
$ClC^{15}N$	-0.0393	-0.0408	-0.0396	-0.0397	-0.0407	-0.0394	-0.0395	-0.04121
$BrC^{15}N$	-0.0331	-0.0344	-0.0334	-0.0335	-0.0339	-0.0329	-0.0330	-0.0325
OCS	-0.0287	-0.0304	-0.0290	-0.0293	-0.0299	-0.0284	-0.0285	-0.028839
$OC^{34}S$	-0.0281	-0.0298	-0.0284	-0.0287	-0.0293	-0.0278	-0.0280	-0.028242
$O^{13}CS$	-0.0286	-0.0303	-0.0288	-0.0292	-0.0298	-0.0283	-0.0284	-0.028710
$^{15}N^{15}NO$	-0.0769	-0.0797	-0.0772	-0.0774	-0.0782	-0.0756	-0.0754	-0.07606
$^{14}N^{14}NO$	-0.0796	-0.0825	-0.0799	-0.0801	-0.0810	-0.0783	-0.0780	-0.07887
$OC^{80}Se$	-0.0196	-0.0210	-0.0198	-0.0200	-0.0204	-0.0192	-0.0192	-0.01952
$OC^{76}Se$	-0.0198	-0.0212	-0.0200	-0.0202	-0.0206	-0.0194	-0.0194	-0.01969
HCP	-0.0419	-0.0468	-0.0461	-0.0458	-0.0450	-0.0442	-0.0437	-0.0430
DCP	-0.0359	-0.0400	-0.0394	-0.0391	-0.0385	-0.0378	-0.0374	-0.0353
HBS	-0.0406	-0.0433	-0.0409	-0.0428	-0.0412	-0.0388	-0.0395	-0.0414
FCCH	-0.0071	-0.0092	-0.0073	-0.0072	-0.0094	-0.0075	-0.0075	-0.0077
$^{35}ClCCH$	-0.0070	-0.0080	-0.0062	-0.0062	-0.0083	-0.0065	-0.0066	-0.00630
$^{37}ClCCH$	-0.0068	-0.0077	-0.0060	-0.0060	-0.0081	-0.0063	-0.0065	-0.00601
$^{79}BrCCH$	-0.0055	-0.0059	-0.0045	-0.0045	-0.0061	-0.0046	-0.0046	-0.00395
$^{81}BrCCH$	-0.0055	-0.0058	-0.0044	-0.0045	-0.0060	-0.0046	-0.0046	-0.00388
Symmetric top molecules								
$^{15}NH_3$	0.5508	0.5615	0.5654	0.5677	0.5592	0.5629	0.5653	0.5654
	0.4870	0.4930	0.4963	0.4977	0.4916	0.4948	0.4951	0.5024
CHF_3	-0.0394	-0.0389	-0.0375	-0.0374	-0.0394	-0.0381	-0.0381	-0.0359
$CH_3\ ^{14}NC$	-0.0589	-0.0608	-0.0588	-0.0589	-0.0601	-0.0582	-0.0580	-0.0546
$CH_3C^{15}N$	-0.0333	-0.0351	-0.0336	-0.0335	-0.0346	-0.0331	-0.0328	-0.0317
$CH_3C\ ^{14}N$	-0.0353	-0.0371	-0.0356	-0.0355	-0.0367	-0.0351	-0.0348	-0.0338
$CD_3C\ ^{14}N$	-0.0331	-0.0347	-0.0334	-0.0333	-0.0343	-0.0330	-0.0327	-0.0315
CH_3F	-0.0657	-0.0645	-0.0596	-0.0592	-0.0660	-0.0612	-0.0609	-0.0620
	0.2494	0.2567	0.2629	0.2642	0.2522	0.2583	0.2588	0.265
CH_3Cl	-0.0186	-0.0180	-0.0137	-0.0136	-0.0196	-0.0153	-0.0155	-0.0165
Asymmetric top molecules								
acrolein	-0.5574	-0.5738	-0.5656	-0.5688	-0.5550	-0.5466	-0.5451	-0.5512
C_3H_4O	-0.0578	-0.0609	-0.0599	-0.0605	-0.0592	-0.0582	-0.0583	-0.0567
	-0.0079	-0.0079	-0.0062	-0.0061	-0.0086	-0.0069	-0.0070	-0.0080

Continued

Continued

Molecule	KT2	B3LYP	B97-2	B97-3	O-B3LYP	O-B97-2	O-B97-3	Exp.
propene	-0.0903	-0.0936	-0.0881	-0.0879	-0.0953	-0.0900	-0.0905	-0.0789
C_3H_6	-0.0441	-0.0455	-0.0430	-0.0431	-0.0449	-0.0425	-0.0424	-0.0424
	0.0101	0.0107	0.0135	0.0137	0.0096	0.0123	0.0122	0.0107
propynal	-0.5713	-0.5865	-0.5787	-0.5814	-0.5627	-0.5546	-0.5501	-0.553
C_3H_2O	-0.0398	-0.0426	-0.0416	-0.0419	-0.0409	-0.0398	-0.0397	-0.040
	-0.0147	-0.0153	-0.0139	-0.0138	-0.0156	-0.0142	-0.0141	-0.015
dimethylether	-0.0552	-0.0478	-0.0356	-0.0343	-0.0524	-0.0402	-0.0399	-0.0214
CH_3OCH_3	-0.0074	-0.0061	-0.0037	-0.0035	-0.0071	-0.0047	-0.0048	-0.0093
	-0.0238	-0.0228	-0.0205	-0.0203	-0.0240	-0.0217	-0.0218	-0.0210
dimethylsulfane	-0.0268	-0.0239	-0.0149	-0.0144	-0.0277	-0.0184	-0.0189	-0.0193
CH_3SCH_3	0.0006	0.0004	0.0038	0.0036	-0.0007	0.0026	0.0021	0.0000
	-0.0086	-0.0089	-0.0062	-0.0065	-0.0101	-0.0075	-0.0080	-0.0083
acetaldehyde	-0.3641	-0.3834	-0.3743	-0.3796	-0.3601	-0.3510	-0.3495	-0.3609
C_2H_4O	-0.0732	-0.0768	-0.0749	-0.0759	-0.0734	-0.0715	-0.0715	-0.0731
	-0.0253	-0.0251	-0.0225	-0.0225	-0.0257	-0.0232	-0.0233	-0.0245
formaldehyde	-2.9286	-3.0654	-2.9966	-3.0271	-2.9232	-2.8567	-2.8457	-2.9017
H_2CO	-0.2276	-0.2342	-0.2319	-0.2338	-0.2248	-0.2224	-0.2207	-0.2243
	-0.1101	-0.1034	-0.0939	-0.0921	-0.1057	-0.0962	-0.0949	-0.0994
thioformaldehyde	-5.4460	-5.6181	-5.4575	-5.5541	-5.4135	-5.2558	-5.2967	-5.2602
H_2CS	-0.1353	-0.1415	-0.1393	-0.1410	-0.1354	-0.1332	-0.1323	-0.1337
	-0.0280	-0.0245	-0.0170	-0.0169	-0.0271	-0.0195	-0.0200	-0.0239
formic acid	-0.3112	-0.3328	-0.3283	-0.3321	-0.3115	-0.3068	-0.3043	-0.2797
HCOOH	-0.0903	-0.0933	-0.0916	-0.0923	-0.0913	-0.0895	-0.0896	-0.0903
	-0.0292	-0.0295	-0.0277	-0.0278	-0.0294	-0.0278	-0.0278	-0.0270
formamide	-0.2843	-0.3000	-0.2939	-0.2950	-0.2857	-0.2798	-0.2770	-0.2843
$HCONH_2$	-0.0666	-0.0697	-0.0676	-0.0682	-0.0677	-0.0656	-0.0655	-0.0649
	-0.0119	-0.0124	-0.0100	-0.0101	-0.0124	-0.0102	-0.0102	-0.0117
glycoaldehyde	-0.1185	-0.1197	-0.1140	-0.1145	-0.1159	-0.1102	-0.1093	-0.1239
$C_2H_4O_2$	-0.0733	-0.0767	-0.0750	-0.0759	-0.0736	-0.0720	-0.0720	-0.0726
	-0.0130	-0.0144	-0.0125	-0.0127	-0.0148	-0.0130	-0.0132	-0.0178
methyl formate	-0.1370	-0.1394	-0.1340	-0.1347	-0.1365	-0.1311	-0.1308	-0.1267
$C_2H_4O_2$	-0.0397	-0.0413	-0.0398	-0.0402	-0.0403	-0.0389	-0.0389	-0.0391
	-0.0179	-0.0185	-0.0169	-0.0169	-0.0187	-0.0172	-0.0173	-0.0167
ketene	-0.3433	-0.3848	-0.3601	-0.3743	-0.3517	-0.3267	-0.3306	-0.4182
H_2C_2O	-0.0349	-0.0377	-0.0350	-0.0351	-0.0372	-0.0346	-0.0345	-0.0356
	-0.0247	-0.0268	-0.0250	-0.0255	-0.0256	-0.0238	-0.0238	-0.0238
difluoromethane	-0.0935	-0.0842	-0.0765	-0.0747	-0.0883	-0.0807	-0.0793	-0.0725
CH_2F_2	-0.0434	-0.0431	-0.0412	-0.0412	-0.0437	-0.0418	-0.0419	-0.0411
	-0.0413	-0.0415	-0.0401	-0.0401	-0.0419	-0.0405	-0.0405	-0.0398
carbonic difluoride	-0.0581	-0.0598	-0.0581	-0.0586	-0.0589	-0.0572	-0.0574	-0.0568
F_2CO	-0.0760	-0.0786	-0.0767	-0.0773	-0.0773	-0.0754	-0.0755	-0.0747
	-0.0331	-0.0335	-0.0323	-0.0325	-0.0337	-0.0326	-0.0328	-0.0328
formyl fluoride	-0.4276	-0.4435	-0.4377	-0.4414	-0.4247	-0.4186	-0.4165	-0.4227

Continued

Continued

Molecule	KT2	B3LYP	B97-2	B97-3	O-B3LYP	O-B97-2	O-B97-3	Exp.
HFCO	-0.0786	-0.0812	-0.0798	-0.0804	-0.0794	-0.0780	-0.0780	-0.0771
	-0.0383	-0.0384	-0.0368	-0.0369	-0.0386	-0.0372	-0.0372	-0.0371
fluoroethene	-0.1717	-0.1751	-0.1712	-0.1700	-0.1754	-0.1715	-0.1709	-0.1533
C_2H_3F	-0.0545	-0.0569	-0.0549	-0.0552	-0.0558	-0.0539	-0.0539	-0.0526
	-0.0044	-0.0042	-0.0021	-0.0021	-0.0048	-0.0028	-0.0029	-0.0037
1,1-difluoroethene	-0.0436	-0.0450	-0.0431	-0.0432	-0.0454	-0.0435	-0.0437	-0.0421
CH_2CF_2	-0.0499	-0.0523	-0.0499	-0.0501	-0.0512	-0.0488	-0.0486	-0.0466
	-0.0128	-0.0129	-0.0115	-0.0116	-0.0133	-0.0120	-0.0121	-0.0119
cis-difluoroethene	-0.1169	-0.1153	-0.1096	-0.1091	-0.1165	-0.1109	-0.1105	-0.1015
CHFCHF	-0.0303	-0.0319	-0.0309	-0.0310	-0.0313	-0.0304	-0.0303	-0.0296
	-0.0168	-0.0169	-0.0155	-0.0155	-0.0174	-0.0160	-0.0161	-0.0158
fluoroethane	-0.0106	-0.0062	0.0024	0.0034	-0.0099	-0.0013	-0.0011	0.0185
CH_3CH_2F	-0.0121	-0.0123	-0.0100	-0.0100	-0.0130	-0.0108	-0.0109	-0.0124
	-0.0255	-0.0258	-0.0238	-0.0238	-0.0264	-0.0244	-0.0246	-0.0197
trifluoroethene	-0.0550	-0.0556	-0.0531	-0.0530	-0.0558	-0.0534	-0.0532	-0.0503
$CHFCF_2$	-0.0331	-0.0342	-0.0328	-0.0330	-0.0340	-0.0327	-0.0327	-0.0321
	-0.0169	-0.0175	-0.0166	-0.0167	-0.0178	-0.0169	-0.0171	-0.0170
ozone	-2.9230	-3.6369	-3.6351	-3.7980	-2.8729	-2.8508	-2.7752	-2.9877
O_3	-0.2313	-0.2657	-0.2635	-0.2729	-0.2259	-0.2226	-0.2195	-0.2295
	-0.0792	-0.0783	-0.0751	-0.0754	-0.0782	-0.0752	-0.0754	-0.0760
sulfur dioxide	-0.6586	-0.6836	-0.6934	-0.6925	-0.6434	-0.6510	-0.6388	-0.6043
SO_2	-0.1188	-0.1207	-0.1196	-0.1200	-0.1179	-0.1168	-0.1164	-0.11634
	-0.0845	-0.0863	-0.0846	-0.0853	-0.0861	-0.0845	-0.0851	-0.08865
difluorooxide	-0.2292	-0.2202	-0.2121	-0.2075	-0.2263	-0.2187	-0.2155	-0.213
OF_2	-0.0606	-0.0606	-0.0585	-0.0580	-0.0611	-0.0590	-0.0587	-0.058
	-0.0729	-0.0738	-0.0714	-0.0715	-0.0720	-0.0698	-0.0694	-0.068
hypofluoros acid	0.6573	0.6741	0.6742	0.6796	0.6726	0.6728	0.6758	0.642
HOF	-0.1188	-0.1212	-0.1164	-0.1164	-0.1211	-0.1165	-0.1161	-0.119
	-0.0684	-0.0676	-0.0633	-0.0629	-0.0692	-0.0650	-0.0648	-0.061
water	0.6331	0.6534	0.6539	0.6591	0.6514	0.6519	0.6547	0.657
H_2O	0.7083	0.7170	0.7179	0.7238	0.7160	0.7171	0.7167	0.718
	0.6227	0.6395	0.6405	0.6445	0.6377	0.6386	0.6401	0.645
hydrogendisulfide	0.4076	0.3951	0.4083	0.4031	0.3907	0.4033	0.3992	0.355
H_2S	0.2020	0.1738	0.1952	0.1871	0.1733	0.1936	0.1849	0.195
	0.2438	0.2243	0.2389	0.2331	0.2210	0.2349	0.2288	0.209
methylenecyclopropane	-0.0754	-0.0727	-0.0672	-0.0664	-0.0752	-0.0698	-0.0696	-0.0672
C_4H_6	-0.0265	-0.0269	-0.0247	-0.0246	-0.0270	-0.0249	-0.0248	-0.0231
	0.0254	0.0261	0.0283	0.0284	0.0248	0.0269	0.0267	0.0244
cyclopropene	-0.0910	-0.0973	-0.0955	-0.0955	-0.0925	-0.0908	-0.0890	-0.0897
C_3H_4	-0.1638	-0.1606	-0.1521	-0.1517	-0.1615	-0.1532	-0.1532	-0.1492
	0.0492	0.0529	0.0558	0.0564	0.0501	0.0528	0.0529	0.0536
aziridine	-0.0466	-0.0428	-0.0350	-0.0342	-0.0464	-0.0387	-0.0387	-0.0422
C_2H_5N	0.0171	0.0212	0.0266	0.0273	0.0179	0.0231	0.0232	0.0229

Continued

Continued

Molecule	KT2	B3LYP	B97-2	B97-3	O-B3LYP	O-B97-2	O-B97-3	Exp.
	0.0527	0.0520	0.0550	0.0549	0.0500	0.0528	0.0522	0.0539
oxirane	-0.1028	-0.1007	-0.0932	-0.0924	-0.1038	-0.0964	-0.0963	-0.0946
C_2H_4O	0.0096	0.0152	0.0201	0.0209	0.0121	0.0168	0.0169	0.0189
	0.0310	0.0310	0.0337	0.0337	0.0291	0.0317	0.0312	0.0318
thiirane	-0.0190	-0.0174	-0.0097	-0.0102	-0.0211	-0.0136	-0.0151	-0.0159
C_2H_4S	-0.0296	-0.0274	-0.0221	-0.0217	-0.0301	-0.0249	-0.0249	-0.0242
	0.0489	0.0483	0.0503	0.0502	0.0468	0.0487	0.0483	0.0487
cyclopropenone	-0.2905	-0.2969	-0.2930	-0.2937	-0.2892	-0.2852	-0.2844	-0.2900
C_3H_2O	-0.1000	-0.1019	-0.0996	-0.0999	-0.1007	-0.0984	-0.0984	-0.0963
	-0.0138	-0.0111	-0.0086	-0.0084	-0.0123	-0.0098	-0.0098	-0.0121
methylcyclopropene	-0.0908	-0.0904	-0.0835	-0.0832	-0.0893	-0.0824	-0.0816	-0.0813
C_4H_6	-0.0455	-0.0461	-0.0443	-0.0444	-0.0454	-0.0436	-0.0435	-0.0261
	0.0180	0.0188	0.0206	0.0205	0.0176	0.0193	0.0189	0.0166
cyclobutene	-0.0555	-0.0575	-0.0545	-0.0547	-0.0565	-0.0535	-0.0531	-0.0516
C_4H_6	-0.0762	-0.0746	-0.0697	-0.0692	-0.0763	-0.0713	-0.0713	-0.0663
	-0.0261	-0.0217	-0.0196	-0.0187	-0.0233	-0.0212	-0.0208	-0.0219
oxetane	-0.0156	-0.0124	-0.0075	-0.0069	-0.0144	-0.0097	-0.0094	-0.0073
C_3H_6O	-0.0500	-0.0472	-0.0420	-0.0411	-0.0496	-0.0444	-0.0440	-0.0429
	-0.0847	-0.0781	-0.0749	-0.0741	-0.0795	-0.0763	-0.0758	-0.0747
beta-propiolactone	-0.0820	-0.0833	-0.0800	-0.0803	-0.0821	-0.0788	-0.0786	-0.0758
$C_3H_4O_2$	-0.0386	-0.0395	-0.0376	-0.0378	-0.0391	-0.0372	-0.0372	-0.0356
	-0.0332	-0.0331	-0.0319	-0.0320	-0.0333	-0.0322	-0.0323	-0.0319
MAE	0.0082	0.0153	0.0125	0.0149	0.0077	0.0057	0.0064	
ME	-0.0043	-0.0133	-0.0084	-0.0107	-0.0037	0.0013	0.0018	
PMAE	10.9	11.3	8.8	9.2	12.0	7.1	6.8	
SD	0.0203	0.0636	0.0583	0.0735	0.0189	0.0165	0.0214	

Table 2: Rotational g tensors for a subset of molecules determined by experiment and calculated using DFT methods and the WY(BD) approach. Basis set used is aug-cc-pVTZ.

Molecule	KT2	B3LYP	B97-2	B97-3	O-B3LYP	O-B97-2	O-B97-3	WY(BD)	Exp.
CO	-0.2673	-0.2811	-0.2827	-0.2861	-0.2704	-0.2717	-0.2719	-0.2677	-0.2689
CSe	-0.2548	-0.2733	-0.2713	-0.2793	-0.2480	-0.2463	-0.2463	-0.2505	-0.2431
$O^{13}CS$	-0.0286	-0.0303	-0.0288	-0.0292	-0.0298	-0.0283	-0.0284	-0.0280	-0.028710
$^{15}N^{15}NO$	-0.0769	-0.0797	-0.0772	-0.0774	-0.0782	-0.0756	-0.0754	-0.0758	-0.07606
$^{15}NH_3$	0.5508	0.5615	0.5654	0.5677	0.5592	0.5629	0.5653	0.5693	0.5654
	0.4870	0.4930	0.4963	0.4977	0.4916	0.4948	0.4951	0.4988	0.5024
SO_2	-0.6586	-0.6836	-0.6934	-0.6925	-0.6434	-0.6510	-0.6388	-0.6121	-0.6043
	-0.1188	-0.1207	-0.1196	-0.1200	-0.1179	-0.1168	-0.1164	-0.1145	-0.11634
	-0.0845	-0.0863	-0.0846	-0.0853	-0.0861	-0.0845	-0.0851	-0.0854	-0.08865
HOF	0.6573	0.6741	0.6742	0.6796	0.6726	0.6728	0.6758	0.6866	0.642
	-0.1188	-0.1212	-0.1164	-0.1164	-0.1211	-0.1165	-0.1161	-0.1156	-0.119
	-0.0684	-0.0676	-0.0633	-0.0629	-0.0692	-0.0650	-0.0648	-0.0644	-0.061
water	0.6331	0.6534	0.6539	0.6591	0.6514	0.6519	0.6547	0.6632	0.657
H_2O	0.7083	0.7170	0.7179	0.7238	0.7160	0.7171	0.7167	0.7270	0.718
	0.6227	0.6395	0.6405	0.6445	0.6377	0.6386	0.6401	0.6474	0.645
MAE	0.0123	0.0132	0.0127	0.0139	0.0084	0.0079	0.0068	0.0066	
ME	-0.0094	-0.0086	-0.0075	-0.0067	-0.0040	-0.0028	-0.0013	0.0036	
PMAE	3.4	4.6	3.6	4.0	3.0	2.3	2.1	2.2	
SD	0.0162	0.0232	0.0257	0.0270	0.0136	0.0151	0.0132	0.0122	