Supplementary Material

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1 Contracted Functions

The contracted functions for the ANO-GS/GSn basis are given in Table 1.

Table 1: Contracted functions for the ANO-GS/GSn basis (continued on pp. 2-7).

System	Ang. Mom.	Exp.	Coef.
H	S	0.04070137	0.01743456
		0.07733261	0.13755618
		0.14693196	0.28886067
		0.27917073	0.28678168
		0.53042439	0.19955827
		1.00780633	0.10852968
		1.91483203	0.05910120
		3.63818086	0.03532391
		6.91254363	0.00208123
He	S	0.10248700	0.04089897
		0.20497400	0.17797219
		0.40994800	0.28528092
		0.81989600	0.26264576
		1.63979200	0.19034914
		3.27958400	0.11079007
		6.55916800	0.05972012
		13.11833600	0.03700733
		26.23667200	0.00265480
Li	S	0.00974359	0.00352139
		0.01851281	0.12903815
		0.03517434	0.44387349
		0.06683125	0.41065480
		0.12697938	0.14370785
		0.24126082	-0.03788394
		0.45839556	-0.07998393
		0.87095157	-0.08019704
		1.65480799	0.02185181
Be	S	0.02562891	-0.02641643
		0.05125781	-0.26469665
		0.10251562	-0.47655474
		0.20503125	-0.31422835
		0.41006250	-0.05743711
		0.82012500	0.08548245
		1.64025000	0.10710350
		3.28050000	-0.01780919
		6.56100000	0.00028805
	P	0.05062500	-0.08962232
		0.10631250	-0.39006883

Table 1 – continued

System	Ang. Mom.	Exp.	Coef.
		0.22325625	-0.41374468
		0.46883813	-0.16909999
		0.98456006	-0.04840263
		2.06757613	-0.03205887
		4.34190988	-0.01041324
		9.11801074	0.00084677
		19.14782255	-0.00015624
B	S	0.05062500	0.06617715
		0.09618750	0.27188133
		0.18275625	0.40697090
		0.34723687	0.30816297
		0.65975006	0.09364792
		1.25352512	-0.05802092
		2.38169773	-0.11533259
		4.52522568	0.01237330
		8.59792879	0.00199977
	P	0.02560000	0.01149541
		0.05376000	0.12869896
		0.11289600	0.30526053
		0.23708160	0.33323434
		0.49787136	0.24570299
		1.04552986	0.12565772
		2.19561270	0.05791787
		4.61078666	0.01337759
		9.68265200	0.00106468
C	S	0.05062500	0.01598887
		0.10125000	0.16450719
		0.20250000	0.38233257
		0.40500000	0.38771387
		0.81000000	0.19273788
		1.62000000	-0.02854048
		3.24000000	-0.12573272
		6.48000000	0.01468002
		12.96000000	-0.00056068
	P	0.05062500	0.03428372
		0.10125000	0.15793083
		0.20250000	0.28424988
		0.40500000	0.30233996
		0.81000000	0.23261465
		1.62000000	0.13296708
		3.24000000	0.06080434
		6.48000000	0.02999658
		12.96000000	0.00135166
N	S	0.07593750	-0.02686191
		0.15187500	-0.19321557
		0.30375000	-0.38474373
		0.60750000	-0.36536725
		1.21500000	-0.18207788
		2.43000000	0.04616424

Table 1 – continued

System	Ang. Mom.	Exp.	Coef.
		4.86000000	0.11528896
		9.72000000	-0.01986060
		19.44000000	0.00078409
	P	0.06750000	0.02961387
		0.13500000	0.14041850
		0.27000000	0.26447192
		0.54000000	0.30063044
		1.08000000	0.24703829
		2.16000000	0.15132427
		4.32000000	0.07150723
		8.64000000	0.03478525
		17.28000000	0.00432024
O	S	0.11000000	0.03202827
		0.19800000	0.15307475
		0.35640000	0.29474179
		0.64152000	0.34704224
		1.15473600	0.23419103
		2.07852480	0.10026844
		3.74134464	-0.07861470
		6.73442035	-0.08350949
		12.12195663	0.01763657
	P	0.07700000	0.04130711
		0.15400000	0.14327885
		0.30800000	0.24460946
		0.61600000	0.27683163
		1.23200000	0.24908259
		2.46400000	0.17441826
		4.92800000	0.08867451
		9.85600000	0.04488804
		19.71200000	0.00673237
F	S	0.13641020	0.02797138
		0.24553835	0.13686497
		0.44196904	0.29018339
		0.79554427	0.33878915
		1.43197968	0.25867176
		2.57756343	0.10284827
		4.63961418	-0.05605286
		8.35130552	-0.09992039
		15.03234993	0.01670889
	P	0.10248700	0.04254657
		0.20497400	0.13947990
		0.40994800	0.24175240
		0.81989600	0.27636193
		1.63979200	0.25237549
		3.27958400	0.17542641
		6.55916800	0.09152650
		13.11833600	0.04443016
		26.23667200	0.00513978
Ne	S	0.17569200	0.03422377

Table 1 – continued

System	Ang. Mom.	Exp.	Coef.
		0.31624560	0.14694149
		0.56924208	0.29359528
		1.02463574	0.33571250
		1.84434434	0.24448271
		3.31981981	0.09720106
		5.97567566	-0.06977995
		10.75621619	-0.06483557
		19.36118914	0.00842959
	P	0.10717944	0.02332263
		0.21435888	0.10697547
		0.42871776	0.21486030
		0.85743552	0.27023961
		1.71487105	0.26653350
		3.42974210	0.20120113
		6.85948419	0.11703442
		13.71896838	0.06254500
		27.43793677	0.00658784
Na	S	0.01726136	0.15962472
		0.03279658	0.46763739
		0.06231350	0.41453230
		0.11839564	0.10369471
		0.22495172	-0.06422858
		0.42740827	-0.09822361
		0.81207570	-0.08827260
		1.54294384	0.03021669
		2.93159329	-0.00088115
Mg	S	0.02297486	-0.05834372
		0.04365224	-0.34313617
		0.08293926	-0.48833841
		0.15758460	-0.26608755
		0.29941074	0.01344630
		0.56888040	0.13225689
		1.08087276	0.14219414
		2.05365825	-0.05439608
		3.90195067	0.00405301
	P	0.03057955	0.02695661
		0.05198523	0.17682202
		0.08837489	0.38169561
		0.15023731	0.37158738
		0.25540342	0.14842290
		0.43418581	-0.00124050
		0.73811588	-0.05271299
		1.25479700	-0.04013899
		2.13315490	0.01107743
Al	S	0.03700125	-0.07202255
		0.06660225	-0.28464829
		0.11988405	-0.45758868
		0.21579129	-0.31670557
		0.38842432	-0.06157971

Table 1 – continued

System	Ang. Mom.	Exp.	Coef.
		0.69916377	0.14261991
		1.25849479	0.17701513
		2.26529063	-0.05806300
		4.07752313	0.00616560
	P	0.02779959	0.10096225
		0.05281921	0.27373295
		0.10035651	0.37457865
		0.19067736	0.27468527
		0.36228699	0.12036825
		0.68834529	-0.00528008
		1.30785604	-0.04982463
		2.48492648	0.00733696
		4.72136031	-0.00113882
Si	S	0.03240000	-0.00874843
		0.06156000	-0.13111530
		0.11696400	-0.41542276
		0.22223160	-0.45858890
		0.42224004	-0.21179781
		0.80225608	0.13567942
		1.52428654	0.21148601
		2.89614443	-0.04725110
		5.50267443	0.00229557
	P	0.03240000	-0.04259816
		0.06156000	-0.19674204
		0.11696400	-0.35101560
		0.22223160	-0.33929651
		0.42224004	-0.19292528
		0.80225608	-0.02815469
		1.52428654	0.05474493
		2.89614443	-0.00309097
		5.50267443	0.00067220
P	S	0.05187485	-0.02375243
		0.09337473	-0.17887230
		0.16807451	-0.39775410
		0.30253412	-0.42608111
		0.54456142	-0.19606826
		0.98021055	0.09284981
		1.76437899	0.24630885
		3.17588218	-0.03921506
		5.71658793	-0.00215415
	P	0.04594973	0.04535552
		0.07811454	0.13096467
		0.13279472	0.26611330
		0.22575102	0.29765470
		0.38377674	0.25786675
		0.65242046	0.13484429
		1.10911478	0.02027525
		1.88549512	-0.05247665
		3.20534170	-0.00416241

Table 1 – continued

System	Ang. Mom.	Exp.	Coef.
S	S	0.06152788	-0.01886366
		0.10459739	-0.11497881
		0.17781556	-0.31264645
		0.30228646	-0.39617586
		0.51388697	-0.31902178
		0.87360786	-0.06793635
		1.48513336	0.18081152
		2.52472671	0.18001334
		4.29203540	-0.05451818
	P	0.05350250	-0.06374846
		0.09630450	-0.16240617
		0.17334810	-0.28676895
		0.31202659	-0.30277046
		0.56164786	-0.24696068
		1.01096614	-0.11183221
		1.81973905	0.03134041
		3.27553029	0.02960453
		5.89595452	-0.00246437
Cl	S	0.09257500	-0.05241911
		0.16663500	-0.21568577
		0.29994300	-0.44405450
		0.53989740	-0.38395406
		0.97181532	-0.17146038
		1.74926758	0.22058963
		3.14868164	0.20298184
		5.66762695	-0.07946711
		10.20172850	0.00689586
	P	0.06118046	-0.04168476
		0.11012482	-0.14017625
		0.19822468	-0.26402369
		0.35680443	-0.31550738
		0.64224797	-0.26657997
		1.15604634	-0.14441887
		2.08088342	0.02261610
		3.74559015	0.04758516
		6.74206227	-0.00681694
Ar	S	0.10000000	-0.03266327
		0.18000000	-0.18332457
		0.32400000	-0.40217886
		0.58320000	-0.43059024
		1.04976000	-0.21192824
		1.88956800	0.14966838
		3.40122240	0.27291260
		6.12220032	-0.08305932
		11.01996058	0.00413157
	P	0.06939182	-0.03009514
		0.12490528	-0.11975400
		0.22482951	-0.24832306
		0.40469311	-0.31388823

Table 1 – continued

System	Ang. Mom.	Exp.	Coef.
		0.72844760	-0.28705129
		1.31120568	-0.16471762
		2.36017023	0.00637195
		4.24830641	0.05891777
		7.64695153	-0.00725650

2 GS Functions

2.1 Normalization of GS functions

The normalization factors for the radial part of the GS basis functions with unit exponent are given in Table 2.

Table 2: Normalization factors for GS basis functions with unit exponent and principal quantum number n.

n	N_n^1
1	1.126467421
2	0.576609950
3	0.196581141
4	0.050275655
5	0.010280772

2.2 Gaussian fits of GS functions

The expansions of the GS functions with unit exponent in a single Gaussian are given in Table 3. The expansions of the GS functions with unit exponent in six Gaussians are given in Table 4.

Table 3: The expansions of the GS functions with unit exponent in a single Gaussian.

Function	Exp.	Coef.
GS-1S	0.20708437	1.00000000
GS-3S	0.04833286	1.00000000
GS-2P	0.15168443	1.00000000
GS-3P	0.08304123	1.00000000
GS-4P	0.05235243	1.00000000
GS-3D	0.11860705	1.00000000
GS-4D	0.07451191	1.00000000
GS-4F	0.09694663	1.00000000

2.3 Exponents

The exponents for the $2z$ ANO-GS basis are given in Table 5. The exponents for the $2z$ ANO-GSn basis of lithium and sodium are given in Table 6. These are the only elements where the $2z$ ANO-GSn basis differs from the $2z$ ANO-GS basis. The exponents for the $3z$ ANO-GS basis are given in Table 7. The exponents for the $3z$ ANO-GSn basis are given in Table 8.

Table 4: The expansions of the GS functions with unit exponent in six Gaussians.

Function	Exp.	Coef.
GS-1S	0.06689139	0.18918908
	0.16047444	0.48208792
	0.39737459	0.32148998
	1.07508975	0.09402732
	3.46365258	0.01304374
	17.18649824	0.00063827
GS-3S	0.02615360	0.10206590
	0.05465914	0.81024134
	0.11465363	0.31331649
	0.24039781	-0.22526824
	0.49848703	-0.08108560
	1.06677389	-0.03103457
GS-2P	0.05676262	0.20261442
	0.12307372	0.48590726
	0.26955306	0.32471268
	0.62972336	0.09930363
	1.67428436	0.01515182
	5.98403593	0.00089381
GS-3P	0.03085706	0.08339441
	0.06586273	0.57673030
	0.14128985	0.39763641
	0.28846603	0.01475027
	0.61860693	-0.01504486
	1.44581552	-0.01698559
GS-4P	0.02525014	0.09557434
	0.04880620	0.65897768
	0.09457222	0.36633078
	0.18377564	-0.08042587
	0.35102086	-0.04341420
	0.69569372	-0.01916470
GS-3D	0.05169545	0.24526750
	0.10571780	0.49873695
	0.21472412	0.29991138
	0.45696287	0.08654449
	1.08040987	0.01296466
	3.24061913	0.00078847
GS-4D	0.03709411	0.22140826
	0.07022210	0.55582268
	0.13371057	0.28014977
	0.24564044	0.03219304
	0.40027045	-0.00451244
	0.96625100	-0.00763014
GS-4F	0.04845121	0.30307423
	0.09536478	0.50746119
	0.18409468	0.26328334
	0.36676059	0.06853641
	0.79713347	0.00963494
	2.12180291	0.00057001

Table 5: Exponents for $2z$ ANO-GS basis (continued on next page).

System	Function	Exp.
H	GS-1S	1.8008
	GS-2P	2.0063
He	GS-1S	3.0477
	GS-2P	2.7297
Li	GS-1S	0.4713
	GS-2P	0.7183
	GS-2P	1.9282
	GS-3D	0.8379
Be	GS-1S	0.4940
	GS-2P	0.6767
	GS-3D	1.2406
B	GS-1S	0.6835
	GS-2P	0.8468
	GS-3D	1.7179
C	GS-1S	0.8335
	GS-2P	1.0750
	GS-3D	2.2359
N	GS-1S	0.8903
	GS-2P	1.2640
	GS-3D	2.5937
O	GS-1S	0.9835
	GS-2P	1.2960
	GS-3D	2.8726
F	GS-1S	1.0734
	GS-2P	1.5617
	GS-3D	3.3773
Ne	GS-1S	1.2705
	GS-2P	1.8062
	GS-3D	4.1117
Na	GS-1S	0.4594
	GS-2P	0.7359
	GS-2P	1.1328
	GS-3D	0.7719
Mg	GS-1S	0.3548
	GS-2P	0.5323
	GS-3D	1.0473
Al	GS-1S	0.4798
	GS-2P	0.6281
	GS-3D	1.2257
Si	GS-1S	0.5335
	GS-2P	0.7775
	GS-3D	1.4937
P	GS-1S	0.6320
	GS-2P	0.9073
	GS-3D	1.7950
S	GS-1S	0.6656
	GS-2P	0.9748
	GS-3D	1.9992
Cl	GS-1S	0.6882

Table 5 – continued

System	Function	Exp.
	GS-2P	1.0867
	GS-3D	2.1929
Ar	GS-1S	0.6914
	GS-2P	1.2242
	GS-3D	2.4625

Table 6: Exponents for $2z$ ANO-GSn bases of lithium and sodium.

System	Function	Exp.
Li	GS-1S	0.4719
	GS-2P	1.5344
	GS-3P	0.9062
	GS-3D	0.8438
Na	GS-1S	0.4594
	GS-2P	1.0875
	GS-3P	0.8750
	GS-3D	0.7750

Table 7: Exponents for $3z$ ANO-GS basis (continued on pp. 11-12).

System	Function	Exp.
H	GS-1S	1.0180
	GS-1S	1.3266
	GS-2P	1.8750
	GS-2P	2.5328
	GS-3D	2.7219
He	GS-1S	1.5133
	GS-1S	2.4992
	GS-2P	2.5180
	GS-2P	4.0086
	GS-3D	3.8367
Li	GS-1S	0.5938
	GS-1S	0.6000
	GS-2P	0.8031
	GS-2P	1.1641
	GS-2P	1.9031
	GS-3D	0.7625
	GS-3D	0.7750
	GS-4F	1.0875
Be	GS-1S	0.8344
	GS-1S	0.8516
	GS-2P	0.7875
	GS-2P	2.2375
	GS-3D	1.0906
	GS-3D	1.0938
	GS-4F	1.5781
B	GS-1S	0.9961

Table 7 – continued

System	Function	Exp.
	GS-1S	1.2078
	GS-2P	0.9828
	GS-2P	1.3445
	GS-3D	1.4289
	GS-3D	1.8094
	GS-4F	2.2172
C	GS-1S	1.1414
	GS-1S	1.6055
	GS-2P	1.2047
	GS-2P	1.6383
	GS-3D	1.9766
	GS-3D	2.4367
	GS-4F	2.7523
N	GS-1S	1.2094
	GS-1S	1.8875
	GS-2P	1.3188
	GS-2P	3.7000
	GS-3D	2.2062
	GS-3D	3.0000
	GS-4F	3.2437
O	GS-1S	1.3344
	GS-1S	2.3312
	GS-2P	1.3219
	GS-2P	4.0687
	GS-3D	2.4906
	GS-3D	3.6063
	GS-4F	3.5219
F	GS-1S	1.7125
	GS-1S	2.5938
	GS-2P	1.6531
	GS-2P	2.4000
	GS-3D	2.7812
	GS-3D	4.2594
	GS-4F	4.0438
Ne	GS-1S	2.1031
	GS-1S	2.8125
	GS-2P	1.8844
	GS-2P	3.2000
	GS-3D	3.2656
	GS-3D	4.9531
	GS-4F	4.8812
Na	GS-1S	0.5406
	GS-1S	0.5719
	GS-2P	0.5813
	GS-2P	0.9031
	GS-2P	0.9187
	GS-3D	0.6687
	GS-3D	0.6656
	GS-4F	1.0531

Table 7 – continued

System	Function	Exp.
Mg	GS-1S	0.7063
	GS-1S	0.7125
	GS-2P	1.0344
	GS-2P	1.0469
	GS-3D	1.1477
	GS-3D	1.1586
	GS-4F	1.2180
Al	GS-1S	0.8500
	GS-1S	0.8656
	GS-2P	0.7188
	GS-2P	1.4000
	GS-3D	1.0437
	GS-3D	1.2594
	GS-4F	1.4656
Si	GS-1S	0.9281
	GS-1S	0.9500
	GS-2P	0.8656
	GS-2P	1.7219
	GS-3D	1.1812
	GS-3D	1.6187
	GS-4F	1.7875
P	GS-1S	1.0844
	GS-1S	1.1125
	GS-2P	1.0250
	GS-2P	2.0312
	GS-3D	1.4781
	GS-3D	1.9625
	GS-4F	2.0781
S	GS-1S	1.2125
	GS-1S	1.2281
	GS-2P	1.0125
	GS-2P	2.2687
	GS-3D	1.5750
	GS-3D	2.2000
	GS-4F	2.2781
Cl	GS-1S	1.3719
	GS-1S	1.3844
	GS-2P	1.1812
	GS-2P	2.5656
	GS-3D	1.8125
	GS-3D	2.3750
	GS-4F	2.5250
Ar	GS-1S	1.5688
	GS-1S	1.5734
	GS-2P	1.3609
	GS-2P	2.8422
	GS-3D	2.0656
	GS-3D	2.6148
	GS-4F	2.9375

Table 8: Exponents for $3z$ ANO-GSn basis (continued on pp. 14-15).

System	Function	Exp.
H	GS-1S	0.9250
	GS-3S	1.7437
	GS-2P	1.8594
	GS-3P	2.1437
	GS-3D	2.7094
He	GS-1S	2.6875
	GS-3S	4.4625
	GS-2P	2.5187
	GS-3P	3.0000
	GS-3D	3.8344
Li	GS-1S	0.5781
	GS-3S	0.7250
	GS-2P	1.6438
	GS-3P	0.8656
	GS-4P	1.4031
	GS-3D	0.7750
	GS-4D	1.2750
	GS-4F	1.1875
Be	GS-1S	0.6031
	GS-3S	1.0625
	GS-2P	2.0219
	GS-3P	0.9344
	GS-3D	1.0500
	GS-4D	1.7781
	GS-4F	1.5781
B	GS-1S	0.7875
	GS-3S	1.4094
	GS-2P	2.2500
	GS-3P	1.1156
	GS-3D	1.8094
	GS-4D	1.6344
	GS-4F	2.2172
C	GS-1S	0.9469
	GS-3S	1.7500
	GS-2P	2.9219
	GS-3P	1.3438
	GS-3D	1.9719
	GS-4D	2.1719
	GS-4F	2.7531
N	GS-1S	1.0312
	GS-3S	1.9000
	GS-2P	3.5063
	GS-3P	1.5688
	GS-3D	3.0094
	GS-4D	2.5469
	GS-4F	3.2594
O	GS-1S	1.0469
	GS-3S	2.4906

Table 8 – continued

System	Function	Exp.
	GS-2P	3.8625
	GS-3P	1.5781
	GS-3D	2.5969
	GS-4D	2.9562
	GS-4F	3.5344
F	GS-1S	1.1375
	GS-3S	2.8906
	GS-2P	4.4125
	GS-3P	1.8469
	GS-3D	4.3156
	GS-4D	3.3312
	GS-4F	4.0594
Ne	GS-1S	1.4500
	GS-3S	3.3563
	GS-2P	5.0750
	GS-3P	2.1656
	GS-3D	5.0031
	GS-4D	3.8500
	GS-4F	4.8750
Na	GS-1S	0.5219
	GS-3S	0.6687
	GS-2P	0.5875
	GS-3P	0.7406
	GS-4P	1.2937
	GS-3D	0.6281
	GS-4D	1.0875
	GS-4F	1.0469
Mg	GS-1S	0.4125
	GS-3S	0.9125
	GS-2P	0.6781
	GS-3P	0.9656
	GS-3D	1.1125
	GS-4D	1.1531
	GS-4F	1.2437
Al	GS-1S	0.6281
	GS-3S	1.1156
	GS-2P	0.6188
	GS-3P	0.9250
	GS-3D	1.0969
	GS-4D	1.1719
	GS-4F	1.4688
Si	GS-1S	0.6188
	GS-3S	1.2531
	GS-2P	0.7531
	GS-3P	1.2031
	GS-3D	1.6187
	GS-4D	1.3594
	GS-4F	1.7969
P	GS-1S	0.7375

Table 8 – continued

System	Function	Exp.
	GS-3S	1.4594
	GS-2P	0.8688
	GS-3P	1.4375
	GS-3D	1.9625
	GS-4D	1.6687
	GS-4F	2.0812
S	GS-1S	0.7875
	GS-3S	1.6687
	GS-2P	0.9156
	GS-3P	1.6313
	GS-3D	2.1969
	GS-4D	1.8469
	GS-4F	2.2844
Cl	GS-1S	0.8000
	GS-3S	1.8781
	GS-2P	1.0063
	GS-3P	1.7906
	GS-3D	2.3484
	GS-4D	2.1000
	GS-4F	2.5375
Ar	GS-1S	0.8094
	GS-3S	2.0875
	GS-2P	1.1156
	GS-3P	1.9500
	GS-3D	2.1281
	GS-4D	3.8687
	GS-4F	2.9312

3 Results

The total energies for the atoms hydrogen through argon using CCSD with the BFD, ANO-GS, and ANO-GSn bases are shown in Table 9. The total energies for the homonuclear dimers of hydrogen through argon using CCSD with the BFD, ANO-GS, and ANO-GSn bases are shown in Table 10. The total energies for Li, O, F, P, and S using several different electronic structure methods with the BFD, ANO-GS, and ANO-GSn bases are shown in Table 11. The total energies for LiF, O_2, P_2, S_2, and SO_2 using several different electronic structure methods with the BFD, ANO-GS, and ANO-GSn bases are shown in Table 12. The atomization energies for LiF, O_2, P_2, S_2, and SO_2 using several different electronic structure methods with the BFD, ANO-GS, and ANO-GSn bases are shown in Table 13.

Table 9: The total energies (in Hartrees) for the atoms hydrogen through argon using CCSD with the BFD, ANO-GS, and ANO-GSn bases. The results for the $2z$ ANO-GS/GSn are identical. In particular, there is no difference for lithium or sodium since their atoms are treated exactly in both cases.

	$2z$ BFD	$2z$ ANO-GS/GSn	$3z$ BFD	$3z$ ANO-GS	$3z$ ANO-GSn	$5z$ BFD
H	-0.499045	-0.500008	-0.499043	-0.500008	-0.500008	-0.499905
He	-2.878934	-2.897624	-2.898728	-2.901811	-2.901593	-2.902789
Li	-0.195611	-0.196326	-0.196093	-0.196326	-0.196326	-0.196315
Be	-1.000525	-1.008573	-1.008436	-1.009838	-1.009939	-1.009957
B	-2.608084	-2.606818	-2.615667	-2.616105	-2.616217	-2.617624
C	-5.408172	-5.406662	-5.425180	-5.426030	-5.426303	-5.429634
N	-9.759589	-9.761698	-9.788099	-9.789509	-9.790472	-9.796294
O	-15.828908	-15.833195	-15.879079	-15.881652	-15.884088	-15.896580
F	-24.091656	-24.099703	-24.159640	-24.164668	-24.166728	-24.186059
Ne	-34.899475	-34.911426	-34.972194	-34.990730	-34.992328	-35.018857
Na	-0.174227	-0.182144	-0.181799	-0.182144	-0.182144	-0.182034
Mg	-0.816857	-0.818098	-0.819095	-0.819352	-0.819531	-0.819679
Al	-1.928593	-1.928912	-1.935164	-1.935209	-1.935676	-1.936678
Si	-3.746217	-3.749091	-3.759962	-3.760189	-3.761051	-3.763306
P	-6.440629	-6.445748	-6.464683	-6.465607	-6.466748	-6.470915
S	-10.062475	-10.072356	-10.109733	-10.110522	-10.113578	-10.123942
Cl	-14.872848	-14.884950	-14.938980	-14.940167	-14.943319	-14.961472
Ar	-21.040743	-21.055925	-21.123540	-21.124919	-21.129124	-21.155927

Table 10: The total energies (in Hartrees) for the homonuclear dimers of hydrogen through argon using CCSD with the BFD, ANO-GS, and ANO-GSn bases. The results for the $2z$ ANO-GS/GSn are nearly identical. In particular, the lithium and sodium results differ by ~ 0.01 mH between the two basis sets.

	$2z$ BFD	$2z$ ANO-GS/GSn	$3z$ BFD	$3z$ ANO-GS	$3z$ ANO-GSn	$5z$ BFD
H_2	-1.170217	-1.170463	-1.173126	-1.173924	-1.173970	-1.174918
He_2	-5.757867	-5.795248	-5.797456	-5.803622	-5.803186	-5.805578
Li_2	-0.429057	-0.429487	-0.431324	-0.431277	-0.431339	-0.431475
Be_2	-1.994640	-2.012318	-2.013852	-2.017464	-2.017607	-2.018268
B_2	-5.294354	-5.300153	-5.318641	-5.320255	-5.320438	-5.325104
C_2	-10.993974	-11.005633	-11.043724	-11.047164	-11.047923	-11.059142
N_2	-19.821534	-19.837516	-19.905031	-19.911701	-19.912931	-19.934928
O_2	-31.803582	-31.818415	-31.922614	-31.930948	-31.934688	-31.966232
F_2	-48.209467	-48.230636	-48.361317	-48.370735	-48.376440	-48.419828
Ne_2	-69.798933	-69.822943	-69.944427	-69.981464	-69.984658	-70.037743
Na_2	-0.381541	-0.389296	-0.390139	-0.390741	-0.390815	-0.390813
Mg_2	-1.632893	-1.635623	-1.638339	-1.638985	-1.639314	-1.639841
Al_2	-3.889249	-3.891268	-3.908157	-3.908561	-3.909380	-3.912708
Si_2	-7.582780	-7.589569	-7.624903	-7.627154	-7.628772	-7.638618
P_2	-13.006848	-13.019340	-13.079636	-13.083205	-13.085725	-13.105334
S_2	-20.243451	-20.267665	-20.357425	-20.361329	-20.367137	-20.396481
Cl_2	-29.805005	-29.833665	-29.955047	-29.956971	-29.965052	-30.007852
Ar_2	-42.081394	-42.111987	-42.247056	-42.249899	-42.258331	-42.312045

Table 11: Total energies (in Hartree) for Li, O, F, P, and S using several different electronic structure methods. Calculations are performed with the BFD, ANO-GS, and ANO-GSn bases. However, the $5z$ BFD* calculations do not include the G or H functions from the $5z$ BFD basis. All diffusion Monte Carlo (DMC) calculations are performed with a trial wavefunction obtained by optimizing Jastrow and orbital parameters via the linear method [1, 2, 3] in variational Monte Carlo. The DMC calculations are for a single-CSF reference (DMC-1CSF). For these systems, this is equivalent to a full-valence complete active space reference (DMC-FVCAS). A 0.01 H^{-1} time step is used for DMC calculations.

			RHF	B3LYP	CCSD	DMC-1CSF
Li	2z	BFD	-0.19561	-0.19754	-0.19561	-0.1963293(8)
Li	2z	ANO-GS	-0.19633	-0.19778	-0.19633	-0.1963299(5)
Li	2z	ANO-GSn	-0.19633	-0.19778	-0.19633	-0.1963304(8)
Li	3z	BFD	-0.19609	-0.19767	-0.19609	-0.1963303(4)
Li	3z	ANO-GS	-0.19633	-0.19781	-0.19633	-0.1963294(4)
Li	3z	ANO-GSn	-0.19633	-0.19781	-0.19633	-0.1963295(4)
Li	5z	BFD*	-0.19631	-0.19785	-0.19631	-0.1963306(3)
O	2z	BFD	-15.70594	-15.89680	-15.82891	-15.89316(9)
O	2z	ANO-GS/GSn	-15.70470	-15.89796	-15.83319	-15.89320(9)
O	3z	BFD	-15.70800	-15.89890	-15.87908	-15.89324(7)
O	3z	ANO-GS	-15.70785	-15.89924	-15.88165	-15.89312(7)
O	3z	ANO-GSn	-15.70799	-15.89925	-15.88409	-15.89321(7)
O	5z	BFD*	-15.70845	-15.89940	-15.89188	-15.89320(7)
F	2z	BFD	-23.93702	-24.19552	-24.09166	-24.18623(5)
F	2z	ANO-GS/GSn	-23.93594	-24.19760	-24.09970	-24.18621(5)
F	3z	BFD	-23.93822	-24.19875	-24.15964	-24.18642(7)
F	3z	ANO-GS	-23.93797	-24.19927	-24.16467	-24.18651(7)
F	3z	ANO-GSn	-23.93815	-24.19927	-24.16673	-24.18650(7)
F	5z	BFD*	-23.93849	-24.19952	-24.17812	-24.18658(8)
P	2z	BFD	-6.35908	-6.44809	-6.44063	-6.47696(9)
P	2z	ANO-GS/GSn	-6.35899	-6.44816	-6.44575	-6.47698(9)
P	3z	BFD	-6.35908	-6.44841	-6.46468	-6.47705(5)
P	3z	ANO-GS	-6.35907	-6.44825	-6.46561	-6.47705(5)
P	3z	ANO-GSn	-6.35907	-6.44823	-6.46675	-6.47702(6)
P	5z	BFD*	-6.35908	-6.44850	-6.46905	-6.47697(5)
S	2z	BFD	-9.95531	-10.09844	-10.06248	-10.1318(1)
S	2z	ANO-GS/GSn	-9.95541	-10.09874	-10.07236	-10.1319(1)
S	3z	BFD	-9.95714	-10.09970	-10.10973	-10.13191(8)
S	3z	ANO-GS	-9.95661	-10.09981	-10.11052	-10.13191(7)
S	3z	ANO-GSn	-9.95727	-10.09986	-10.11358	-10.13191(8)
S	5z	BFD*	-9.95742	-10.10005	-10.11845	-10.13195(7)

Table 12: Total energies (in Hartree) of several systems from G2 set [4] at their experimental geometries [5] using different electronic structure methods. Calculations are performed with the BFD, ANO-GS, and ANO-GSn bases. All diffusion Monte Carlo calculations are performed with a trial wavefunction obtained by optimizing Jastrow, orbital, and configuration state function (CSF) parameters (where applicable) via the linear method [1, 2, 3] in variational Monte Carlo. For each system, the DMC calculations are performed with both a single-CSF reference (DMC-1CSF) and full-valence complete active space reference (DMC-FVCAS). A 0.01 H^{-1} time step is used for DMC calculations.

			RHF	B3LYP	CCSD	DMC 1-CSF	DMC FVCAS
LiF	2z	BFD	-24.27255	-24.59507	-24.47966	-24.6034(1)	-24.6073(1)
LiF	2z	ANO-GS	-24.27844	-24.60193	-24.49408	-24.60552(8)	-24.60829(7)
LiF	2z	ANO-GSn	-24.27946	-24.60327	-24.49568	-24.6057(2)	-24.60852(7)
LiF	3z	BFD	-24.28220	-24.60833	-24.56394	-24.60567(9)	-24.60985(6)
LiF	3z	ANO-GS	-24.28439	-24.61057	-24.57235	-24.6060(1)	-24.61005(7)
LiF	3z	ANO-GSn	-24.28430	-24.61052	-24.57310	-24.6059(1)	-24.6102(1)
LiF	5z	BFD*	-24.28550	-24.61160	-24.58740	-24.6060(1)	-24.61026(7)
O_2	2z	BFD	-31.43491	-31.96283	-31.80358	-31.9601(1)	-31.9715(1)
O_2	2z	ANO-GS/GSn	-31.43861	-31.97019	-31.81842	-31.9642(1)	-31.97439(9)
O_2	3z	BFD	-31.45438	-31.97639	-31.92261	-31.9697(1)	-31.97562(9)
O_2	3z	ANO-GS	-31.45616	-31.97892	-31.93095	-31.9709(1)	-31.97695(7)
O_2	3z	ANO-GSn	-31.45657	-31.97930	-31.93469	-31.9712(1)	-31.97731(8)
O_2	5z	BFD*	-31.45869	-31.98096	-31.95414	-31.9721(1)	-31.97816(5)
P_2	2z	BFD	-12.76276	-13.06483	-13.00685	-13.12686(9)	-13.13125(8)
P_2	2z	ANO-GS/GSn	-12.77082	-13.07018	-13.01934	-13.12796(9)	-13.13256(8)
P_2	3z	BFD	-12.77495	-13.07404	-13.07964	-13.1283(1)	-13.13272(8)
P_2	3z	ANO-GS	-12.77794	-13.07675	-13.08320	-13.13017(9)	-13.13472(8)
P_2	3z	ANO-GSn	-12.77785	-13.07642	-13.08573	-13.1299(1)	-13.13456(8)
P_2	5z	BFD*	-12.77890	-13.07769	-13.09576	-13.13082(9)	-13.13535(4)
S_2	2z	BFD	-19.97287	-20.34439	-20.24345	-20.4186(1)	-20.4193(1)
S_2	2z	ANO-GS/GSn	-19.98230	-20.35322	-20.26766	-20.4212(1)	-20.42199(9)
S_2	3z	BFD	-19.98990	-20.35785	-20.35743	-20.4218(1)	-20.4225(1)
S_2	3z	ANO-GS	-19.99188	-20.36111	-20.36133	-20.4237(1)	-20.4246(1)
S_2	3z	ANO-GSn	-19.99353	-20.36134	-20.36714	-20.4236(1)	-20.4246(1)
S_2	5z	BFD*	-19.99451	-20.36265	-20.38135	-20.4249(1)	-20.42569(9)
SO_2	2z	BFD	-41.48746	-42.22186	-42.00477	-42.2939(1)	-42.2983(1)
SO_2	2z	ANO-GS/GSn	-41.52259	-42.26068	-42.05531	-42.3126(1)	-42.3180(1)
SO_2	3z	BFD	-41.55800	-42.28125	-42.22455	-42.3171(1)	-42.3224(1)
SO_2	3z	ANO-GS	-41.56769	-42.29184	-42.24229	-42.32256(8)	-42.32781(6)
SO_2	3z	ANO-GSn	-41.56777	-42.29155	-42.24683	-42.32216(8)	-42.32734(6)
SO_2	5z	BFD*	-41.57326	-42.29628	-42.27913	-42.32436(8)	-42.33020(6)

Table 13: Atomization energies (in kcal/mol) of several systems from G2 set [4] at their experimental geometries [5] using different electronic structure methods. Calculations are performed with the BFD, ANO-GS, and ANO-GSn bases. Calculated atomization energies are corrected with the experimental zero point energies [5, 6]. All diffusion Monte Carlo calculations are performed with a trial wavefunction obtained by optimizing Jastrow, orbital, and configuration state function (CSF) parameters (where applicable) via the linear method [1, 2, 3] in variational Monte Carlo. For each system, the DMC calculations are performed with both a single-CSF reference (DMC-1CSF) and full-valence complete active space reference (DMC-FVCAS). A 0.01 H^{-1} time step is used for DMC calculations.

			RHF	B3LYP	CCSD	DMC 1-CSF	DMC FVCAS	Experiment
LiF	2z	BFD	86.5	125.5	119.4	137.28(7)	139.73(7)	138(2)
LiF	2z	ANO-GS	90.4	128.3	123.0	138.62(6)	140.36(5)	138(2)
LiF	2z	ANO-GSn	91.1	129.2	124.0	138.7(1)	140.51(5)	138(2)
LiF	3z	BFD	91.5	131.7	129.4	138.59(7)	141.21(6)	138(2)
LiF	3z	ANO-GS	92.9	132.7	131.3	138.74(8)	141.28(6)	138(2)
LiF	3z	ANO-GSn	92.7	132.6	130.5	138.68(8)	141.38(8)	138(2)
LiF	5z	BFD*	93.3	133.1	132.3	138.69(8)	141.37(7)	138(2)
O_2	2z	BFD	12.2	103.9	89.2	106.8(1)	114.0(1)	117.96(2)
O_2	2z	ANO-GS/GSn	16.1	107.1	93.1	109.3(1)	115.7(1)	117.96(2)
O_2	3z	BFD	21.8	109.8	100.9	112.7(1)	116.4(1)	117.96(2)
O_2	3z	ANO-GS	23.1	111.0	102.9	113.6(1)	117.4(1)	117.96(2)
O_2	3z	ANO-GSn	23.2	111.2	102.2	113.7(1)	117.5(1)	117.96(2)
O_2	5z	BFD*	24.0	112.1	104.7	114.3(1)	118.08(9)	117.96(2)
P_2	2z	BFD	26.9	104.7	77.7	107.4(1)	110.2(1)	116.1(5)
P_2	2z	ANO-GS/GSn	32.0	108.0	79.1	108.1(1)	111.0(1)	116.1(5)
P_2	3z	BFD	34.5	110.1	93.2	108.20(9)	110.97(8)	116.1(5)
P_2	3z	ANO-GS	36.4	112.0	94.3	109.37(8)	112.23(8)	116.1(5)
P_2	3z	ANO-GSn	36.4	111.8	94.4	109.2(1)	112.16(9)	116.1(5)
P_2	5z	BFD*	37.0	112.3	97.8	109.88(8)	112.72(7)	116.1(5)
S_2	2z	BFD	38.0	91.5	73.3	96.2(1)	96.7(1)	100.66(7)
S_2	2z	ANO-GS/GSn	43.8	96.7	76.1	97.7(1)	98.2(1)	100.66(7)
S_2	3z	BFD	46.4	98.4	85.5	98.1(1)	98.5(1)	100.66(7)
S_2	3z	ANO-GS	48.3	100.3	87.0	99.3(1)	99.9(1)	100.66(7)
S_2	3z	ANO-GSn	48.5	100.4	86.8	99.2(1)	99.9(1)	100.66(7)
S_2	5z	BFD*	49.0	101.0	89.6	100.0(1)	100.5(1)	100.66(7)
SO_2	2z	BFD	71.1	202.6	174.2	231.5(1)	234.2(1)	254.0(2)
SO_2	2z	ANO-GS/GSn	94.7	225.3	194.3	243.1(1)	246.5(1)	254.0(2)
SO_2	3z	BFD	111.7	236.5	219.5	245.9(1)	249.2(1)	254.0(2)
SO_2	3z	ANO-GS	118.3	242.6	226.9	249.4(1)	252.7(1)	254.0(2)
SO_2	3z	ANO-GSn	117.7	242.4	224.8	249.1(1)	252.3(1)	254.0(2)
SO_2	5z	BFD*	120.5	245.1	232.2	250.4(1)	254.1(1)	254.0(2)

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