Supplemental Information
Removing Orbital-dependence to Improve Exchange-Correlation Functional Accuracy

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We provide

1. Discussion of saturation magnetization calcualations for bcc Fe, hcp Co, and fcc Ni with MVS, MVS-L(CRopt), and PBE;
2. Detailed, system-by-system tabulation of the numerical results for the testing against standard molecular and crystalline system data sets reported in the main paper;
3. Discussion of ffects of self-consistent reparametrization of the CR de-orbitalizer.

LIST OF TABLES

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I. MAGNETIZATION

One of our original motivations for reconsidering MVS was the hope of gaining insight into the over-magnetization of *3d* elemental solids exhibited by the more sophisticated SCAN functional, which also uses only the α indicator. See Ref. 1 and references therein.

We computed the saturation magnetization of bcc Fe, hcp Co, and fcc Ni at their respective equilibrium lattice constants (ideal c/a for hcp) using the MVS, MVS-L(CRopt), and PC functionals. The reported experimental magnetic moments for bcc Fe, fcc Co, and fcc Ni are 2.22, 1.72, and 0.62 μ_B, respectively [2, 3]. Figs. 1-3 present the corresponding magnetization curves. The MVS functional gives notably different energetic behavior compared both to its de-orbitalized version, MVS-L(CRopt), and to PBE. MVS-L magnetization curves, in contrast, have qualitatively similar behavior to that of PBE.

Table I presents the magnetization values. For bcc Fe, MVS overestimates the saturation magnetization by 20.7 % relative to the experimental value, whereas MVS-L(CRopt) overestimates it by 1.02 %. For hcp Co, the MVS error relative to experimental values are less severe, 6.4 % versus the MVS-L(CRopt) underestimate by 8.2 %. For fcc Ni, both MVS and MVS-L overestimate, 27.42 % and 9.78 %, respectively. Clearly MVS-L(CRopt) subsumes quite different physics of saturation magnetization than its parent functional. Detailed investigation is a distinct exploration outside the scope of this work.

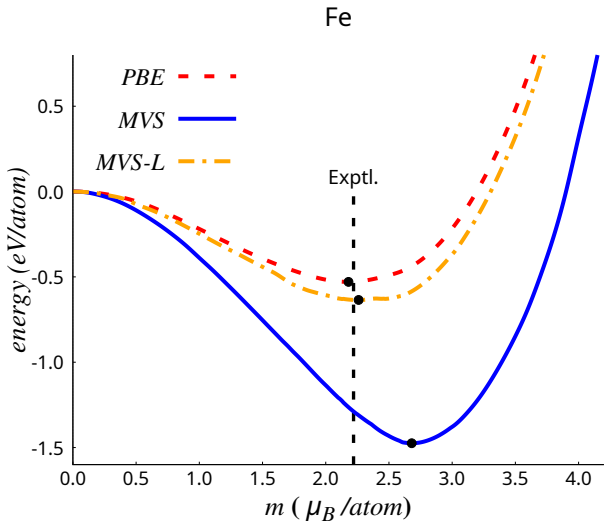

FIG. 1. Fixed spin moment energy on a per-atom basis for bcc Fe from MVS, MVS-L(CRopt), and PBE XC functionals using the calculated equilibrium lattice parameters.

TABLE I. Magnetic moments in μ_B for three elemental *3d* solids as determined from different XC functionals. Exp. refers to the experimental data [2, 3].

	Exp.	MVS	MVS-L(CRopt)	PBE
Fe	2.22	2.68	2.26	2.18
Co	1.72	1.83	1.56	1.64
Ni	0.62	0.79	0.68	0.63

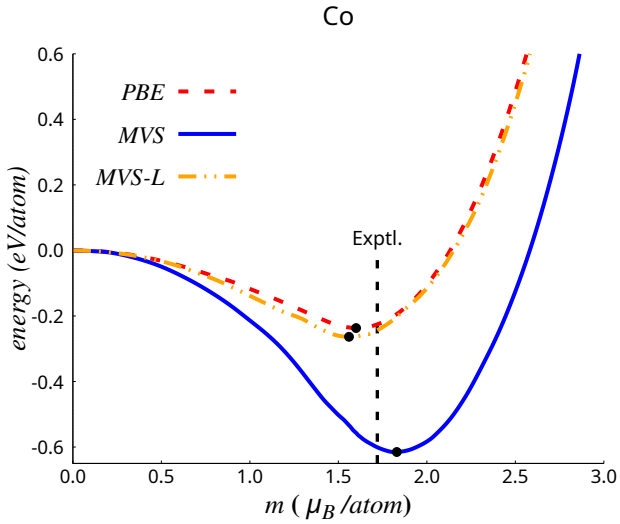

FIG. 2. As in Fig. 1 for hcp Co.

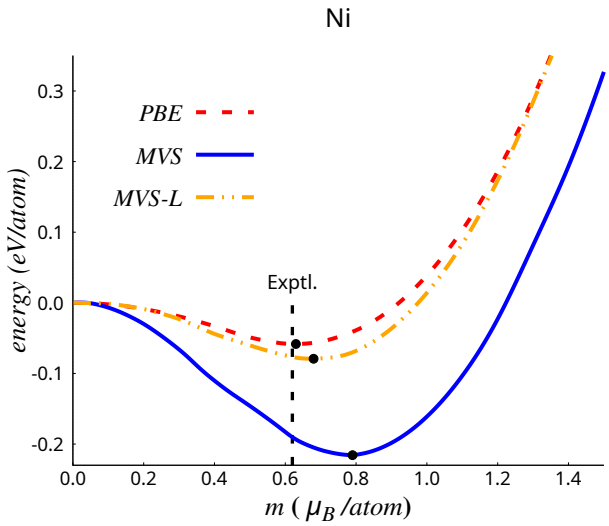

FIG. 3. As in Fig. 1 for fcc Ni.

II. MOLECULAR TEST SET RESULTS CALCULATED IN NWCHEM-7.0.2

Here we provide the molecule-by-molecule tabulation for the G3X/99, T96R, and T82-F test sets for MVS and MVS-L(CRopt) compared r^{2}SCAN, r^{2}SCAN-L, and PBE. Calculations were performed in NWChem-7.0.2.

TABLE II: Standard enthalpies of formation (ΔH_f) in kcal/mol for the G3X/99 molecular test set [4, 5] obtained with different approximate exchange-correlation functionals.

Molecule	Exptl.	MVS	MVS-L(CRopt)	MVS-L(CR)	r^{2}SCAN	r^{2}SCAN-L	PBE
LiH	33.30	39.644	39.676	40.809	36.369	37.109	38.353
^{2}BeH	81.70	74.917	74.223	76.606	72.401	71.715	77.032
^{2}CH	142.50	146.270	140.313	142.907	145.246	142.898	141.870
CH_2 ($^3B_1$)	93.60	85.826	87.529	91.631	87.380	89.023	89.512
CH_2 ($^1A_1$)	102.60	109.125	103.426	106.602	109.080	107.423	104.969
$^3CH_3$	35.00	32.325	31.729	35.814	31.093	33.253	32.400
CH_4	-17.80	-15.517	-13.108	-10.718	-16.985	-13.443	-18.051

(continued)

^{3}NH	85.80	89.320	80.892	83.528	84.819	80.954	80.598
NH_2 ($^2B_1$)	44.50	50.533	40.337	43.178	43.940	39.917	38.882
NH_3	-10.90	-2.195	-8.902	-6.866	-6.591	-9.316	-14.099
^{2}OH	9.00	9.688	6.978	8.151	7.906	6.770	6.631
H_2O	-57.80	-56.870	-54.687	-52.815	-54.083	-53.778	-58.172
HF	-65.20	-67.870	-62.315	-59.064	-61.578	-59.851	-65.067
SiH_2 ($^1A_1$)	65.20	67.663	67.579	66.746	67.659	68.505	68.485
SiH_2 ($^3B_1$)	86.20	77.310	79.759	80.185	78.683	80.106	84.892
$^2SiH_3$	47.90	43.203	45.973	45.797	43.307	45.945	50.421
SiH_4	8.20	7.931	13.718	11.999	8.051	12.501	16.150
$^2PH_2$	33.10	34.237	30.570	31.603	30.299	30.770	31.274
PH_3	1.30	7.299	4.810	4.783	2.916	4.561	3.543
H_2S	-4.90	-5.589	-5.126	-4.175	-4.802	-2.944	-4.615
HCl	-22.00	-25.520	-22.654	-21.862	-21.697	-20.269	-21.934
Li_2	51.60	61.132	59.069	58.900	57.947	57.313	56.191
LiF	-80.10	-79.678	-72.782	-66.255	-75.223	-72.315	-79.231
HC≡CH	54.60	47.712	45.509	54.977	57.291	58.892	45.137
$H_2C{=}CH_2$	12.50	10.285	11.770	15.834	13.271	16.661	4.228
$H_3C{-}CH_3$	-20.10	-20.081	-12.199	-12.452	-19.899	-14.284	-25.214
CN	105.20	102.352	95.157	99.637	108.285	101.621	89.042
HCN	30.90	30.058	23.560	29.601	36.347	32.404	18.183
CO	-26.40	-34.016	-27.956	-26.476	-22.494	-22.436	-36.038
^{2}HCO	10.00	-0.813	4.772	4.582	6.377	5.781	-6.433
H_2CO	-26.10	-31.968	-25.849	-26.483	-25.490	-24.880	-38.135
CH_3OH	-48.00	-50.225	-40.154	-43.899	-46.549	-44.067	-55.038
N_2	0.00	5.633	-4.901	-2.201	8.712	-0.240	-14.907
H_2NNH_2	23.30	40.922	29.608	25.912	32.413	24.446	12.788
NO	21.80	19.827	17.659	13.871	22.182	15.018	2.200
$^3O_2$	0.00	-16.507	-6.611	-16.765	-9.495	-12.965	-23.578
H_2O_2	-32.40	-33.092	-30.687	-39.539	-31.281	-35.353	-45.264
F_2	0.00	-4.679	-0.199	-11.806	-0.542	-4.667	-14.480
CO_2	-94.00	-121.849	-102.398	-101.200	-99.730	-97.447	-121.233
Na_2	34.00	39.159	39.988	39.161	37.360	37.546	33.232
Si_2	139.90	133.844	131.994	130.369	140.050	135.912	133.292
P_2	34.30	33.647	31.626	32.474	38.906	36.922	29.927
$^3S_2$	30.70	15.392	22.846	19.920	21.557	22.318	17.177
Cl_2	0.00	-6.352	1.866	-5.460	-1.173	-1.638	-7.570
NaCl	-43.60	-49.876	-44.155	-40.441	-42.209	-39.092	-39.225
SiO	-24.60	-27.708	-19.414	-20.346	-19.627	-19.149	-27.933
CS	66.90	59.211	60.872	62.159	69.398	68.214	58.405
SO	1.20	-11.699	-2.159	-6.708	-5.788	-6.158	-14.255
ClO	24.30	17.198	22.233	14.266	18.816	15.974	7.982
ClF	-13.30	-18.508	-10.485	-18.323	-13.513	-14.553	-23.623
Si_2H_6	19.10	12.485	24.823	19.810	16.115	23.590	28.424
CH_3Cl	-19.60	-25.341	-16.046	-19.263	-20.538	-17.375	-25.231
$H_3C{-}SH$	-5.50	-8.803	-2.394	-4.292	-6.357	-2.615	-10.501
HOCl	-18.40	-21.685	-16.133	-23.786	-18.260	-20.219	-28.359
SO_2	-71.00	-89.087	-65.313	-73.428	-72.660	-71.378	-91.183
BF_3	-271.40	-306.748	-261.571	-260.888	-273.732	-261.174	-282.638
BCl_3	-96.30	-130.663	-102.480	-109.299	-106.297	-100.432	-110.171
AlF_3	-289.00	-319.666	-282.272	-273.831	-286.282	-273.842	-287.468
$AlCl_3$	-139.70	-177.785	-149.509	-150.213	-147.936	-138.944	-140.648

(continued)

CF_4	-223.10	-273.220	-205.950	-225.607	-229.516	-216.625	-250.932
CCl_4	-22.90	-55.988	-23.023	-43.217	-29.324	-27.076	-44.635
OCS	-33.10	-62.382	-45.976	-46.625	-42.142	-40.244	-59.555
CS_2	28.00	-2.671	11.714	10.238	16.922	18.742	3.687
F_2CO	-145.00	-182.394	-140.028	-150.050	-151.353	-144.425	-174.364
SiF_4	-386.00	-428.963	-367.251	-367.192	-381.350	-362.250	-383.670
$SiCl_4$	-158.40	-203.546	-158.238	-170.514	-165.540	-154.410	-162.572
NNO	19.70	2.779	7.305	2.046	14.882	5.088	-20.819
ClNO	12.60	-4.062	-0.012	-10.515	5.366	-3.159	-21.066
NF_3	-31.60	-58.914	-35.174	-58.353	-40.578	-45.794	-71.956
PF_3	-229.10	-253.980	-207.954	-218.751	-226.170	-216.140	-239.427
O_3	33.90	18.045	30.193	8.521	29.247	18.630	-3.619
F_2O	5.90	-9.110	1.062	-19.028	-1.016	-7.977	-24.698
ClF_3	-38.00	-67.901	-39.980	-61.329	-52.045	-52.659	-76.974
$F_2C{=}CF_2$	-161.30	-213.454	-149.497	-170.441	-170.535	-160.217	-204.597
$Cl_2C{=}CCl_2$	-5.50	-48.703	-7.703	-28.136	-14.830	-10.984	-38.126
$F_3C{-}CN$	-118.40	-162.441	-116.639	-129.951	-121.756	-117.287	-160.508
$HC{\equiv}C{-}CH_3$	44.40	32.992	38.857	45.500	44.560	49.244	28.038
$H_2C{=}C{=}CH_2$	45.40	31.562	36.216	42.470	41.757	46.143	24.612
C_3H_4 (cyclopropene)	67.80	53.557	64.296	66.130	65.517	69.956	47.295
$H_2C{=}CH{-}CH_3$	4.80	-1.109	7.452	8.846	4.184	10.216	-8.811
C_3H_6 (cyclopropane)	12.80	1.342	18.202	16.081	10.333	17.650	-2.957
$CH_3{-}CH_2{-}CH_3$	-25.10	-27.763	-13.750	-16.675	-25.256	-17.376	-34.289
C_4H_6 (Z-1,3-butadiene)	26.40	13.433	22.372	26.238	24.253	30.580	3.800
C_4H_6 (2-butyne)	34.80	19.578	33.621	37.641	33.601	41.191	12.610
C_4H_6 (methylenecyclopropane)	47.90	27.357	45.840	45.206	41.192	49.145	19.512
C_4H_6 (bicyclo[1.1.0]butane)	51.90	30.501	55.767	51.871	48.326	57.165	26.702
C_4H_6 (cyclobutene)	38.20	24.119	38.027	36.447	36.791	43.603	14.490
C_4H_8 (cyclobutane)	6.60	-4.317	13.686	8.500	5.204	13.579	-12.675
C_4H_8 (isobutene)	-4.10	-13.463	1.866	0.920	-5.292	3.326	-21.712
C_4H_{10} (trans butane)	-30.10	-36.131	-15.879	-20.726	-30.572	-20.449	-43.284
C_4H_{10} (isobutane)	-32.20	-37.676	-17.607	-22.860	-32.187	-21.989	-44.374
C_5H_8 (spiropentane)	44.30	18.634	51.347	44.497	37.527	49.222	11.935
C_6H_6 (benzene)	19.90	-14.539	10.420	10.687	10.403	21.101	-22.848
CH_2F_2	-107.70	-126.671	-95.915	-105.531	-109.028	-102.636	-122.140
CHF_3	-166.30	-200.286	-151.722	-166.469	-170.262	-160.693	-187.903
CH_2Cl_2	-22.60	-37.038	-20.148	-28.723	-25.527	-22.453	-33.914
$CHCl_3$	-24.50	-48.204	-22.993	-37.300	-29.246	-26.387	-41.183
CH_3NH_2	-5.20	9.081	8.610	7.577	5.683	4.732	-5.273
CH_3CN	18.00	11.824	13.591	17.259	20.217	19.696	-2.068
CH_3NO_2 (nitromethane)	-17.90	-39.079	-19.635	-34.958	-25.879	-29.863	-59.555
CH_3ONO (methyl nitrite)	-16.10	-31.746	-14.791	-33.602	-21.287	-27.059	-54.847
CH_3SiH_3	-7.00	-10.941	1.321	-0.522	-7.007	0.193	-2.305
HCO_2H	-90.40	-108.178	-87.783	-93.457	-93.046	-90.478	-111.954
HCO_2CH_3	-85.50	-106.328	-77.667	-88.436	-89.540	-84.805	-111.885
CH_3CONH_2	-57.00	-70.085	-54.988	-58.459	-58.876	-56.622	-84.231
C_2H_5N (aziridine)	30.20	25.655	34.673	29.352	30.296	31.194	10.031
NCCN (cyanogen)	74.10	62.366	53.233	61.070	77.596	68.762	37.081
$NH(CH_3)_2$	-4.30	-0.156	6.048	0.875	-1.721	-0.130	-17.806
$CH_3CH_2NH_2$	-11.30	-8.991	-2.941	-6.985	-9.440	-7.713	-25.203
$H_2C{=}C{=}O$ (ketene)	-11.60	-33.661	-21.464	-18.225	-17.674	-14.523	-37.267
C_2H_4O (oxirane)	-12.60	-24.941	-6.336	-14.583	-15.115	-11.040	-32.026

(continued)

CH_3CHO	-39.50	-50.781	-36.891	-39.100	-41.209	-37.277	-57.475
$O{=}CH{-}CH{=}O$	-50.80	-70.357	-52.821	-57.743	-52.935	-51.983	-80.457
CH_3CH_2OH	-56.10	-61.842	-45.491	-51.306	-55.501	-50.440	-67.563
$(CH_3)_2O$	-44.00	-49.220	-30.694	-39.716	-44.134	-39.194	-56.457
C_2H_4S (thiooxirane)	19.60	5.536	20.762	15.519	15.131	20.141	2.869
$(CH_3)_2S{=}O$	-36.20	-52.343	-26.307	-35.154	-40.593	-33.373	-55.337
CH_3CH_2SH	-11.10	-16.787	-4.701	-8.898	-12.118	-6.069	-19.965
$(CH_3)_2S$	-8.90	-15.454	-2.249	-7.205	-10.926	-4.989	-19.041
$H_2C{=}CHF$	-34.00	-48.740	-32.005	-33.943	-35.880	-30.799	-51.223
CH_3CH_2Cl	-26.60	-35.512	-19.985	-25.708	-28.392	-22.771	-36.728
$H_2C{=}CHCl$	5.20	-7.637	3.102	1.511	2.760	6.443	-9.818
$H_2C{=}CHCN$	43.20	32.871	33.743	39.422	46.042	45.318	15.936
$(CH_3)_2C{=}O$	-51.70	-67.503	-46.105	-50.232	-54.629	-47.703	-73.961
CH_3CO_2H	-103.40	-125.671	-98.271	-105.038	-106.948	-101.283	-128.749
CH_3CFO	-105.70	-131.475	-99.961	-106.996	-109.997	-102.930	-131.162
CH_3COCl	-57.70	-83.119	-59.022	-66.144	-64.631	-59.903	-83.546
$CH_3CH_2CH_2Cl$	-31.50	-43.802	-22.118	-29.797	-33.686	-25.818	-45.698
$(CH_3)_2CHOH$	-65.20	-74.592	-51.289	-59.890	-65.178	-57.559	-80.323
$CH_3{-}O{-}CH_2CH_3$	-51.70	-60.998	-35.401	-46.880	-52.970	-45.482	-68.837
$(CH_3)_3N$	-6.60	-4.882	8.470	-0.194	-4.148	-0.171	-23.659
C_4H_4O (furan)	-8.30	-39.733	-11.063	-18.015	-15.438	-8.334	-46.264
C_4H_4S (thiophene)	27.50	-4.796	19.722	15.644	18.924	27.078	-6.991
C_4H_5N (pyrrole)	25.90	2.061	20.053	17.701	20.536	25.053	-13.692
C_5H_5N (pyridine)	33.60	4.856	24.004	20.858	25.893	30.671	-14.365
H_2	0.00	3.993	5.086	4.993	2.029	3.370	4.886
^2SH	34.20	32.897	31.540	32.924	32.445	33.084	33.105
$^2C{\equiv}CH$	135.80	125.061	124.396	132.133	134.490	134.664	123.889
$'HC{=}CH_2$ ($^2A''$)'	71.00	62.129	63.810	68.618	66.139	68.536	58.415
$'CH_3C{=}O$ ($^2A''$)'	-2.40	-17.868	-4.933	-6.693	-8.016	-5.418	-24.710
$CH_2{-}OH$ (^2A)	-4.00	-11.462	-3.213	-5.857	-7.899	-6.516	-15.885
$'CH_3O$ ($^2A''$)'	5.20	-1.038	6.207	2.013	-1.003	0.761	-8.513
$'CH_3CH_2O$ ($^2A''''$)'	-2.90	-13.347	0.129	-6.113	-10.758	-6.431	-22.343
$'CH_3S$ ($^2A''$)'	29.80	23.873	29.420	27.825	25.434	28.251	21.234
$'CH_2CH_3$ ($^2A''$)'	28.60	22.797	29.120	30.460	23.478	28.166	19.913
$'(CH_3)_2CH$ ($^2A''$)'	21.10	11.687	25.257	23.787	14.611	21.949	6.724
$^2C(CH_3)_3$	12.00	-0.383	20.368	16.306	5.238	15.230	-6.483
NO_2 ($^2A_1$)	8.10	-14.137	-2.509	-14.825	-3.219	-11.088	-34.634
$CH_3CH{=}C{=}CH_2$	38.80	22.846	33.749	37.065	34.599	41.379	13.314
C_5H_8 (isoprene)	18.00	1.624	17.392	18.796	15.537	24.329	-8.185
C_5H_{10} (cyclopentane twist)	-18.50	-32.124	-4.485	-14.643	-20.086	-8.784	-40.154
C_5H_{12} (n-pentane)	-35.00	-44.048	-18.474	-24.938	-35.811	-23.446	-52.206
$C(CH_3)_4$ (neopentane)	-40.10	-48.850	-22.692	-30.300	-39.783	-27.331	-54.531
C_6H_8 (1,3-cyclohexadiene)	25.40	0.906	25.987	22.449	21.575	32.086	-9.671
C_6H_8 (1,4-cyclohexadiene)	25.00	1.109	26.897	23.083	21.779	32.585	-9.517
C_6H_{12} (cyclohexane chair)	-29.40	-46.450	-12.051	-23.868	-30.893	-17.059	-53.494
C_6H_{14} (n-hexane)	-39.90	-50.270	-19.888	-29.253	-41.090	-26.405	-61.144
C_6H_{14} (3-methylpentane)	-41.10	-52.341	-20.933	-29.753	-41.004	-26.565	-60.442
$C_6H_5{-}CH_3$ (toluene)	12.00	-25.772	6.629	4.144	2.192	15.416	-34.726
C_7H_{16} (n-heptane)	-44.80	-59.387	-23.267	-33.465	-46.317	-29.454	-70.074
C_8H_8 (cyclooctatetraene)	70.70	36.492	62.419	60.859	65.076	76.464	18.532
C_8H_{18} (n-octane)	-49.80	-67.174	-25.357	-37.646	-51.578	-32.464	-79.015
$C_{10}H_8$ (naphtalene)	35.90	-29.386	17.071	13.837	17.424	34.814	-40.015

(continued)

$C_{10}H_8$ (azulene)	69.10	6.870	50.705	45.098	50.165	66.252	-9.667
$CH_3CO_2CH_3$ (Z-methylacetate)	-98.40	-123.362	-86.911	-99.407	-102.893	-95.109	-128.162
$(CH_3)_3COH$ (t-butanol)	-74.70	-87.723	-58.067	-68.880	-74.627	-64.578	-92.259
$C_6H_5NH_2$ (aniline)	20.80	-14.191	11.101	8.016	11.151	18.846	-32.284
C_6H_5OH (phenol)	-22.30	-66.231	-29.599	-35.294	-33.497	-22.547	-73.497
C_4H_6O (divinyl ether)	-3.30	-24.295	-2.521	-6.827	-7.406	-0.764	-35.231
C_4H_8O (tetrahydrofuran)	-44.00	-60.951	-27.912	-43.199	-45.741	-36.685	-68.725
C_5H_8O (cyclopentanone)	-45.90	-74.474	-39.441	-49.452	-51.377	-40.705	-81.653
$C_6H_4O_2$ (benzoquinone)	-29.40	-77.153	-40.740	-47.934	-38.774	-31.421	-90.551
$C_6H_4N_2$ (pyrimidine)	46.80	22.133	36.293	29.208	39.251	38.453	-7.724
$(CH_3)_2SO_2$	-89.20	-120.164	-74.676	-86.240	-95.574	-83.642	-113.367
C_6H_5Cl (chlorobenzene)	12.50	-32.110	1.870	-4.128	0.253	11.245	-36.164
$NC(CH_2)_2CN$ (succinonitrile)	50.10	33.977	40.289	42.943	54.670	51.985	7.507
$C_4H_4N_2$ (pyrazine)	46.90	27.099	40.176	32.711	43.407	42.011	-4.086
C_4H_4O (3-butyn-2-one)	15.60	-8.975	8.578	11.434	15.244	20.204	-15.729
C_4H_6O (E-crotonaldehyde)	-24.00	-46.458	-25.149	-28.052	-29.433	-22.649	-57.070
$C_4H_6O_3$ (acetic anhydride)	-136.80	-180.026	-129.114	-144.984	-146.708	-136.689	-185.596
C_4H_6S (2,5-dihydrothiophene)	20.80	-1.054	21.803	13.585	15.945	23.923	-7.106
$(CH_3)_2CHCN$	5.60	-4.363	9.090	7.982	9.229	13.050	-20.146
C_4H_8O (methyl ethyl ketone)	-57.10	-76.258	-48.092	-54.611	-60.325	-51.134	-83.160
$(CH_3)_2CHCHO$	-51.60	-66.973	-40.909	-48.266	-52.368	-43.945	-75.639
$C_4H_8O_2$ (1,4-dioxane)	-75.50	-99.349	-53.480	-78.569	-78.669	-69.832	-107.583
C_4H_8S (tetrahydrothiophene)	-8.20	-25.148	2.316	-10.138	-10.968	-1.369	-29.721
$(CH_3)_3CCl$	-43.50	-59.491	-31.510	-41.943	-45.684	-35.297	-59.576
C_4H_9Cl (n-butyl chloride)	-37.00	-50.552	-24.426	-34.158	-38.869	-28.722	-54.560
C_4H_9N (tetrahydropyrrole)	-0.80	-8.693	11.337	-0.294	-0.164	5.179	-27.032
$C_4H_9NO_2$ (2-nitrobutane)	-39.10	-69.382	-30.246	-53.047	-47.460	-44.384	-90.951
$(CH_3CH_2)_2O$	-60.30	-71.765	-40.518	-54.240	-61.704	-51.628	-81.093
$CH_3CH(OCH_3)_2$ (dimethyl acetal)	-93.10	-112.592	-67.836	-90.118	-95.091	-85.050	-119.944
$(CH_3)_3CSH$	-26.20	-38.755	-14.212	-23.404	-27.472	-16.801	-41.107
$C_4H_{10}S_2$ (diethyl disulfide)	-17.90	-36.309	-3.328	-18.186	-22.447	-11.194	-41.111
$(CH_3)_3CNH_2$	-28.90	-32.478	-13.930	-22.892	-26.378	-19.879	-47.647
$(CH_3)_4Si$	-55.70	-69.846	-38.544	-40.787	-54.175	-38.488	-58.530
C_5H_6S (2-methyl thiophene)	20.00	-15.414	14.801	9.094	10.745	21.461	-18.880
C_5H_7N (N-methyl pyrrole)	24.60	-1.939	24.644	18.094	18.955	25.792	-19.073
$C_5H_{10}O$ (tetrahydropyran)	-53.40	-73.996	-33.582	-51.872	-55.557	-44.128	-81.279
$(CH_3CH_2)_2C{=}O$	-61.60	-84.519	-50.626	-59.138	-65.921	-54.472	-92.252
$C_5H_{10}O_2$ (isopropyl acetate)	-115.10	-145.709	-97.705	-114.663	-120.534	-107.716	-152.246
$C_5H_{10}S$ (tetrahydrothiopyran)	-15.20	-35.555	-1.958	-16.174	-18.399	-6.440	-40.167
$C_5H_{11}N$ (piperidine)	-11.30	-21.997	5.690	-8.985	-10.425	-2.564	-39.964
$C_5H_{12}O$ (t-butyl methyl ether)	-67.80	-83.641	-45.388	-61.222	-68.525	-56.363	-89.665
$C_6H_4F_2$ (1,3-difluorobenzene)	-73.90	-134.648	-79.269	-90.073	-88.648	-74.292	-133.695
$C_6H_4F_2$ (1,4-difluorobenzene)	-73.30	-133.604	-78.435	-89.341	-87.757	-73.541	-132.983
C_6H_5F (fluorobenzene)	-27.40	-74.853	-34.683	-39.992	-39.430	-26.907	-78.603
$C_6H_{14}O$ (diisopropyl ether)	-76.30	-94.033	-50.814	-68.535	-77.839	-62.933	-103.079
PF_5	-381.10	-435.491	-350.331	-369.126	-380.254	-360.011	-391.911
SF_6	-291.70	-367.019	-261.613	-301.329	-303.621	-284.615	-323.703
P_4	14.10	-1.805	15.139	2.423	9.944	11.530	-6.429
SO_3	-94.60	-132.353	-90.506	-101.604	-102.558	-98.006	-125.370
SCl_2	-4.20	-19.006	-2.272	-14.320	-9.243	-8.592	-18.677
$POCl_3$	-133.80	-172.551	-128.376	-143.744	-140.862	-133.935	-151.623
PCl_5	-86.10	-132.893	-84.791	-111.267	-99.199	-93.756	-108.417

(continued)

Cl_2O_2S	-84.80	-128.879	-80.247	-100.421	-96.999	-91.340	-116.410
PCl_3	-69.00	-91.855	-64.797	-79.276	-74.777	-71.565	-83.704
Cl_2S_2	-4.00	-31.774	-8.558	-24.869	-18.120	-16.570	-31.124
$SiCl_2$ ($^1A_1$)	-40.30	-57.771	-36.680	-44.168	-42.734	-38.579	-46.440
CF_3Cl	-169.70	-216.262	-158.176	-178.504	-177.683	-167.722	-197.941
C_2F_6	-320.90	-399.572	-295.880	-330.040	-332.636	-313.857	-367.896
$^2CF_3$	-111.80	-151.134	-104.631	-117.888	-122.127	-113.933	-139.910
$^2C_6H_5$ (phenyl radical)	80.50	39.553	64.711	65.231	65.334	74.912	33.117
ME		-17.226	3.012	-2.933	-3.145	1.845	-20.878
MAD		18.242	7.845	6.906	4.488	5.300	21.385

TABLE III: Bond lengths in $\AA$ for the T96-R molecular test set [6, 7] from different approximate XC functionals.

Molecule	Exptl.	MVS	MVS-L(CRopt)	MVS-L(CR)	r^{2}SCAN	r^{2}SCAN-L	PBE
H_2	0.741	0.741	0.742	0.747	0.742	0.742	0.751
Li_2	2.673	2.800	2.742	2.737	2.762	2.729	2.738
LiH	1.595	1.616	1.595	1.601	1.605	1.595	1.607
LiF	1.564	1.574	1.564	1.558	1.574	1.569	1.576
LiCl	2.021	2.030	2.016	2.001	2.029	2.016	2.021
LiO	1.688	1.698	1.686	1.681	1.696	1.691	1.699
Be_2	2.440	2.527	2.384	2.413	2.489	2.395	2.441
BeH	1.343	1.359	1.352	1.357	1.357	1.349	1.362
BeF	1.361	1.373	1.365	1.366	1.374	1.373	1.382
BeO	1.331	1.333	1.328	1.332	1.335	1.336	1.346
BeS	1.742	1.743	1.733	1.734	1.749	1.744	1.759
B_2	1.590	1.620	1.599	1.606	1.618	1.609	1.619
BH	1.232	1.243	1.247	1.259	1.242	1.240	1.252
BF	1.263	1.265	1.264	1.270	1.267	1.270	1.276
BF_3	1.313	1.307	1.309	1.312	1.314	1.318	1.325
BCl	1.715	1.719	1.715	1.723	1.724	1.722	1.732
BCl_3	1.742	1.732	1.734	1.730	1.742	1.740	1.748
BN	1.281	1.319	1.321	1.325	1.321	1.323	1.333
BO	1.204	1.205	1.203	1.207	1.206	1.209	1.215
BS	1.609	1.607	1.603	1.606	1.612	1.610	1.621
C_2	1.242	1.246	1.398	1.398	1.249	1.394	1.404
CH	1.120	1.125	1.126	1.133	1.126	1.125	1.137
CH_4	1.087	1.088	1.089	1.095	1.088	1.088	1.096
CF	1.272	1.270	1.282	1.286	1.278	1.285	1.287
CF_4	1.323	1.312	1.328	1.330	1.325	1.333	1.337
CCl	1.645	1.641	1.645	1.653	1.654	1.657	1.660
CCl_4	1.767	1.761	1.774	1.770	1.774	1.778	1.782
CN	1.172	1.163	1.164	1.168	1.165	1.169	1.174
CO	1.128	1.126	1.129	1.133	1.127	1.132	1.136
CO^+	1.115	1.110	1.110	1.115	1.112	1.116	1.122
CO_2	1.160	1.158	1.158	1.168	1.162	1.167	1.171
CP	1.562	1.549	1.547	1.552	1.555	1.556	1.567
CS	1.535	1.533	1.533	1.537	1.536	1.539	1.546
CS_2	1.553	1.546	1.544	1.549	1.553	1.554	1.562
N_2	1.098	1.092	1.095	1.099	1.093	1.099	1.103
N_2^+	1.116	1.103	1.103	1.108	1.106	1.110	1.114
NH	1.036	1.037	1.036	1.046	1.041	1.043	1.051
NH^+	1.070	1.074	1.079	1.084	1.078	1.080	1.091
NF	1.317	1.308	1.331	1.330	1.319	1.330	1.327
NCl	1.611	1.600	1.614	1.622	1.616	1.622	1.618
NO	1.151	1.143	1.150	1.155	1.148	1.155	1.158
NO^+	1.063	1.058	1.061	1.066	1.059	1.066	1.070
NS	1.494	1.486	1.491	1.498	1.494	1.500	1.506
O_2	1.208	1.198	1.216	1.220	1.207	1.218	1.219
O_2^+	1.116	1.104	1.115	1.121	1.109	1.119	1.122
OH	0.970	0.968	0.966	0.972	0.973	0.975	0.983
OH^+	1.029	1.029	1.032	1.037	1.035	1.038	1.048
OF	1.358	1.332	1.375	1.364	1.350	1.364	1.360
F_2	1.412	1.382	1.426	1.412	1.399	1.415	1.413
F_2^+	1.322	1.281	1.327	1.319	1.298	1.318	1.316

(continued)

HF	0.917	0.914	0.912	0.920	0.921	0.924	0.930
HF^+	1.001	1.003	1.003	1.004	1.010	1.011	1.023
Na_2	3.079	3.228	3.248	3.176	3.142	3.120	3.079
NaH	1.887	1.914	1.920	1.900	1.898	1.887	1.893
NaF	1.926	1.921	1.946	1.915	1.926	1.926	1.940
NaCl	2.361	2.361	2.381	2.342	2.365	2.357	2.367
NaO	2.052	2.050	2.075	2.042	2.056	2.053	2.070
MgH	1.730	1.737	1.737	1.730	1.745	1.735	1.757
MgF	1.750	1.754	1.764	1.752	1.766	1.766	1.784
MgCl	2.196	2.196	2.201	2.181	2.212	2.204	2.226
MgO	1.748	1.722	1.729	1.728	1.736	1.737	1.750
Al_2	2.466	2.444	2.434	2.427	2.471	2.462	2.488
AlH	1.648	1.649	1.661	1.657	1.661	1.656	1.679
AlF	1.654	1.651	1.657	1.654	1.664	1.666	1.684
AlCl	2.130	2.121	2.120	2.115	2.145	2.140	2.163
AlO	1.618	1.603	1.604	1.611	1.619	1.619	1.633
AlS	2.029	2.010	2.010	2.008	2.031	2.028	2.049
Si_2	2.246	2.134	2.246	2.248	2.152	2.263	2.287
SiH	1.520	1.522	1.521	1.527	1.530	1.524	1.545
SiH_4	1.480	1.482	1.480	1.486	1.482	1.479	1.493
SiF	1.601	1.598	1.604	1.606	1.613	1.617	1.632
SiF_4	1.553	1.553	1.555	1.557	1.564	1.567	1.580
SiCl	2.058	2.045	2.047	2.048	2.070	2.067	2.084
$SiCl_4$	2.019	2.007	2.009	2.004	2.025	2.024	2.039
SiN	1.572	1.560	1.557	1.563	1.569	1.570	1.580
SiO	1.510	1.506	1.508	1.514	1.515	1.519	1.529
SiS	1.929	1.923	1.922	1.924	1.937	1.937	1.951
P_2	1.893	1.878	1.878	1.881	1.891	1.892	1.905
P_4	2.210	2.165	2.185	2.187	2.190	2.193	2.208
PH	1.421	1.420	1.418	1.426	1.427	1.424	1.440
PF	1.589	1.581	1.596	1.601	1.599	1.607	1.617
PCl	2.015	1.995	2.005	2.010	2.019	2.021	2.031
PN	1.491	1.482	1.484	1.488	1.487	1.492	1.500
PO	1.476	1.473	1.476	1.483	1.482	1.488	1.497
S_2	1.889	1.877	1.882	1.889	1.896	1.900	1.912
SH	1.341	1.340	1.335	1.342	1.345	1.342	1.356
SF	1.601	1.586	1.620	1.617	1.605	1.617	1.622
SF_6	1.561	1.552	1.580	1.582	1.576	1.589	1.596
SO	1.481	1.478	1.488	1.498	1.490	1.499	1.505
SO_3	1.420	1.416	1.421	1.433	1.429	1.438	1.444
Cl_2	1.988	1.967	2.008	2.001	1.996	2.005	2.008
Cl_2^+	1.891	1.871	1.904	1.904	1.898	1.908	1.914
HCl	1.275	1.273	1.268	1.275	1.279	1.277	1.288
HCl^+	1.315	1.315	1.313	1.318	1.321	1.319	1.333
ClF	1.628	1.615	1.662	1.654	1.636	1.651	1.652
ClO	1.570	1.551	1.590	1.591	1.575	1.587	1.583
ME		-0.002	0.005	0.005	0.005	0.008	0.018
MAD		0.014	0.013	0.012	0.010	0.011	0.018

TABLE IV: Harmonic vibrational frequencies in cm^{-1} for the T82-F molecular test set [6, 7] obtained with different approximate exchange-correlation functionals.

Molecule	Exptl.	MVS	MVS-L(CRopt)	MVS-L(CR)	r^{2}SCAN	r^{2}SCAN-L	PBE
H_2	4401.20	4462.35	4448.81	4396.56	4429.67	4433.00	4316.60
Li_2	351.40	328.15	341.18	342.53	332.37	338.50	329.83
LiH	1405.70	1392.59	1410.30	1393.95	1394.28	1398.39	1377.44
LiF	910.60	902.31	942.72	915.35	904.77	900.09	894.04
LiCl	643.00	644.28	650.59	664.84	642.66	647.69	644.19
LiO	814.60	806.61	823.04	819.00	810.73	805.43	797.92
LiNa	256.80	244.11	245.64	251.39	246.33	249.04	245.92
Be_2	267.90	319.87	398.91	380.43	327.20	378.91	347.20
BeH	2060.80	2043.58	2040.64	2032.59	2009.73	2032.61	1974.63
BeH^+	2221.70	2251.21	2224.89	2191.03	2203.03	2204.43	2109.72
BeF	1247.40	1235.66	1263.14	1243.13	1222.55	1216.82	1196.15
BeCl	846.70	835.51	856.89	856.59	816.44	828.52	809.36
BeO	1487.30	1536.64	1552.10	1508.14	1508.31	1485.15	1455.69
BeS	997.90	1033.75	1050.20	1036.82	1008.22	1002.21	975.83
B_2	1051.30	1043.46	1075.90	1041.38	1012.77	1026.00	1011.60
BH	2366.90	2339.86	2325.62	2269.38	2301.01	2311.73	2245.71
BF	1402.10	1427.25	1413.12	1378.17	1398.46	1374.72	1354.73
BCl	840.30	849.99	860.67	828.80	826.55	822.02	810.20
BN	1514.60	1597.55	1561.71	1543.76	1570.16	1547.01	1513.24
BO	1885.70	1924.08	1933.23	1883.33	1896.33	1866.83	1844.00
BS	1180.20	1222.01	1231.90	1210.01	1195.46	1185.04	1161.49
C_2	1854.70	1942.46	1977.44	1917.07	1891.01	1873.68	1857.11
CH	2858.50	2846.12	2845.51	2795.20	2803.97	2802.58	2730.18
CF	1308.10	1346.83	1256.71	1243.24	1297.42	1257.20	1257.32
CN	2068.60	2184.49	2191.12	2136.85	2149.60	2113.03	2093.57
CO	2169.80	2225.84	2211.03	2147.17	2195.55	2145.55	2129.62
CO^+	2214.20	2315.61	2320.06	2246.76	2277.95	2226.89	2202.58
CP	1239.70	1322.31	1332.48	1297.43	1286.41	1269.35	1250.18
CS	1285.20	1321.15	1321.12	1301.12	1304.75	1279.88	1263.69
N_2	2358.60	2457.59	2431.63	2381.29	2432.68	2371.77	2353.66
N_2^+	2207.00	2381.60	2377.46	2303.89	2326.82	2282.72	2264.91
NH	3282.30	3312.92	3343.24	3240.55	3246.50	3217.63	3171.82
NF	1141.40	1209.65	1092.08	1121.08	1171.33	1122.99	1128.53
NCl	828.00	867.57	815.79	822.63	836.47	823.16	835.85
NO	1904.20	2007.28	1930.91	1889.84	1959.47	1893.38	1892.16
NO^+	2376.40	2485.26	2449.06	2389.06	2456.14	2379.66	2363.83
NS	1218.70	1292.75	1274.58	1245.58	1254.42	1220.84	1213.17
O_2	1580.20	1696.64	1536.38	1545.81	1624.96	1552.32	1555.90
O_2^+	1904.80	2089.28	1952.04	1918.34	2026.58	1932.96	1931.33
OH	3737.80	3785.85	3822.68	3741.07	3718.29	3689.78	3602.70
OH^+	3113.40	3129.13	3096.30	3039.46	3060.89	3023.95	2953.36
F_2	916.60	1079.03	980.36	1018.76	1048.14	1006.03	997.29
F_2^+	1073.30	1266.81	1130.44	1163.24	1219.83	1154.32	1151.83
HF	4138.30	4202.39	4252.25	4093.61	4093.34	4047.58	3977.20
HF^+	3090.50	3090.14	3096.86	3074.62	3024.75	3000.15	2910.03
Na_2	159.10	155.37	154.09	157.34	155.29	156.89	158.96
NaH	1172.20	1156.84	1153.75	1160.49	1167.88	1170.47	1143.28
NaF	535.70	545.19	516.69	539.28	539.36	533.81	519.85
NaO	492.30	496.45	472.09	491.04	488.05	483.38	466.16

(continued)

MgH	1495.20	1523.40	1517.79	1514.18	1461.59	1480.39	1409.21
MgH^+	1699.10	1759.75	1733.14	1730.95	1720.88	1721.29	1642.69
MgO	784.80	874.87	845.32	833.86	837.42	823.41	803.21
MgS	528.70	579.38	572.59	568.81	550.24	543.50	526.22
Al_2	350.00	383.61	390.63	384.33	355.77	354.35	341.93
AlH	1682.60	1716.22	1656.10	1639.67	1646.51	1651.24	1585.99
AlF	802.30	817.74	803.33	798.14	788.75	776.71	747.66
AlCl	481.30	501.69	499.97	497.41	475.87	473.41	458.80
AlO	979.20	1057.49	1049.82	1007.74	992.26	985.14	956.88
AlS	617.10	674.63	672.54	662.57	630.55	625.83	600.43
Si_2	511.00	593.22	530.55	513.93	564.20	499.32	483.24
SiH	2041.80	2081.80	2058.90	2040.75	2020.24	2030.81	1951.71
SiH^+	2157.20	2206.87	2148.55	2101.33	2119.64	2110.97	2047.83
SiF	857.20	885.43	854.08	843.95	847.30	831.01	807.65
SiCl	535.60	557.72	542.10	538.78	529.27	524.43	515.10
SiN	1151.40	1222.56	1230.12	1190.49	1179.68	1166.46	1150.13
SiO	1241.50	1282.66	1272.23	1236.17	1247.89	1219.92	1196.56
SiS	749.60	781.92	780.45	768.23	753.33	741.63	726.42
P_2	780.80	842.35	838.25	823.76	813.84	798.52	782.42
$P_2{}^+$	672.20	736.43	727.53	717.01	707.78	693.66	674.31
PH	2365.20	2422.09	2450.94	2387.03	2367.90	2363.18	2288.59
PF	846.80	889.81	816.67	818.21	846.90	821.17	811.02
PCl	551.40	578.16	545.13	542.96	550.12	539.08	538.06
PN	1337.20	1422.60	1410.70	1382.57	1390.71	1357.51	1339.51
PO	1233.30	1292.48	1272.72	1237.03	1250.59	1214.09	1200.39
S_2	725.60	769.47	738.24	723.09	732.65	713.94	701.88
SO	1149.20	1208.12	1131.16	1108.83	1159.28	1115.80	1110.40
Cl_2	559.70	586.59	524.81	541.35	555.49	535.88	540.40
$Cl_2{}^+$	645.60	696.17	621.19	632.68	649.26	629.91	624.21
HCl	2990.90	3050.84	3078.80	3011.07	2981.86	2965.12	2901.24
HCl^+	2673.70	2695.80	2709.30	2673.66	2643.90	2630.45	2571.65
ClF	786.10	825.54	738.84	761.58	791.24	758.60	760.84
ClO	853.80	923.71	816.42	839.42	872.53	843.62	862.59
ME		47.24	28.58	5.61	11.34	-7.25	-33.78
MAD		51.44	42.46	28.24	30.90	25.71	43.61

III. MOLECULAR TEST SET RESULTS CALCULATED IN VASP-5.4.4

Here we provide the molecule-by-molecule tabulation for the G3X/99 and T46-R test sets for MVS and MVS-L(CRopt) compared with r^{2}SCAN and r^{2}SCAN-L. Calculations were performed with VASP-5.4.4.

TABLE V: Standard enthalpies of formation (ΔH_f) in kcal/mol for the G3X/99 molecular test set [4, 5] obtained with different approximate exchange-correlation functionals obtained with VASP.

Molecule	Exptl.	MVS	MVS-L(CRopt)	r^{2}SCAN	r^{2}SCAN-L	PBE
LiH	33.30	39.454	39.245	35.824	36.608	37.632
^{2}BeH	81.70	74.046	75.653	71.053	70.806	76.084
^{2}CH	142.50	153.785	144.132	153.807	148.257	144.870
CH_2 ($^3B_1$)	93.60	85.782	87.047	87.275	89.007	89.651
CH_2 ($^1A_1$)	102.60	108.802	102.384	108.988	107.210	104.980
$^3CH_3$	35.00	32.158	29.913	31.199	32.787	32.764
CH_4	-17.80	-15.528	-17.440	-15.896	-12.713	-17.164
^{3}NH	85.80	88.363	79.525	83.928	80.149	80.274
NH_2 ($^2B_1$)	44.50	49.030	38.021	43.224	39.170	38.402
NH_3	-10.90	-4.083	-12.835	-6.642	-9.447	-14.565
^{2}OH	9.00	21.339	13.595	22.318	16.353	11.739
H_2O	-57.80	-57.232	-56.610	-52.908	-52.814	-57.980
HF	-65.20	-67.966	-63.000	-60.362	-58.172	-64.352
SiH_2 ($^1A_1$)	65.20	62.803	64.843	67.664	69.202	68.822
SiH_2 ($^3B_1$)	86.20	73.162	77.553	79.097	80.531	85.495
$^2SiH_3$	47.90	38.622	43.213	43.519	46.011	51.089
SiH_4	8.20	3.025	9.505	8.091	12.823	16.944
$^2PH_2$	33.10	33.075	28.326	29.401	30.075	31.164
PH_3	1.30	5.937	0.215	2.315	4.653	3.771
H_2S	-4.90	-5.999	-6.949	-3.867	-2.506	-4.716
HCl	-22.00	-24.745	-23.721	-20.387	-19.530	-22.133
Li_2	51.60	60.948	58.703	57.326	56.995	55.654
LiF	-80.10	-79.692	-73.817	-76.825	-72.906	-81.522
HC≡CH	54.60	47.913	46.227	58.943	59.974	45.992
$H_2C{=}CH_2$	12.50	10.121	7.686	15.149	18.108	5.523
$H_3C{-}CH_3$	-20.10	-20.300	-20.361	-17.937	-12.701	-23.626
CN	105.20	103.369	95.428	109.663	103.427	90.150
HCN	30.90	30.841	23.446	38.182	34.466	19.640
CO	-26.40	-30.949	-26.510	-18.147	-17.591	-32.208
^{2}HCO	10.00	1.268	4.167	9.792	8.900	-3.503
H_2CO	-26.10	-30.013	-27.711	-21.924	-21.501	-35.320
CH_3OH	-48.00	-49.982	-46.304	-44.240	-41.965	-53.549
N_2	0.00	9.077	-3.395	12.592	4.607	-11.349
H_2NNH_2	23.30	33.311	18.901	28.699	21.402	8.879
NO	21.80	25.689	21.234	32.112	23.379	7.870
$^3O_2$	0.00	-15.603	-5.958	-4.660	-8.124	-19.231
H_2O_2	-32.40	-34.202	-32.646	-28.583	-32.615	-43.529
F_2	0.00	-6.962	-1.679	0.714	-2.029	-12.815
CO_2	-94.00	-115.700	-100.281	-91.108	-89.236	-114.290
Na_2	34.00	38.607	34.322	36.717	37.917	32.860
Si_2	139.90	120.130	128.213	136.593	136.984	133.456
P_2	34.30	32.551	24.852	38.081	37.685	29.813
$^3S_2$	30.70	14.550	20.051	22.307	22.380	16.754
Cl_2	0.00	-4.502	-1.520	1.029	0.001	-7.806

(continued)

NaCl	-43.60	-48.897	-43.348	-40.635	-36.757	-39.280
SiO	-24.60	-32.034	-20.938	-18.866	-16.991	-27.425
CS	66.90	58.776	58.759	70.738	69.555	58.738
SO	1.20	-14.062	-5.661	-6.059	-6.536	-15.234
ClO	24.30	20.444	21.091	25.688	20.123	9.247
ClF	-13.30	-19.370	-13.853	-12.671	-13.460	-24.038
Si_2H_6	19.10	3.154	16.803	16.433	25.297	30.157
CH_3Cl	-19.60	-24.229	-21.618	-18.337	-15.622	-24.473
H_3C-SH	-5.50	-9.282	-8.715	-4.609	-1.213	-9.814
HOCl	-18.40	-21.106	-19.367	-16.094	-18.340	-27.944
SO_2	-71.00	-93.812	-71.611	-73.155	-70.639	-93.378
BF_3	-271.40	-302.049	-260.733	-267.819	-251.254	-276.380
BCl_3	-96.30	-127.705	-107.493	-101.253	-94.054	-108.210
AlF_3	-289.00	-321.105	-284.511	-286.122	-270.572	-286.910
$AlCl_3$	-139.70	-174.158	-149.651	-142.181	-132.486	-138.030
CF_4	-223.10	-267.173	-211.587	-218.245	-203.843	-240.820
CCl_4	-22.90	-52.427	-31.516	-23.610	-21.843	-43.629
OCS	-33.10	-59.527	-48.082	-36.935	-35.254	-56.227
CS_2	28.00	-2.672	6.452	19.086	20.754	3.715
F_2CO	-145.00	-176.386	-142.058	-141.750	-134.141	-166.200
SiF_4	-386.00	-432.796	-366.671	-376.630	-352.682	-378.640
$SiCl_4$	-158.40	-205.009	-163.469	-158.400	-145.393	-159.470
NNO	19.70	6.544	7.814	20.800	12.452	-15.446
ClNO	12.60	0.593	0.466	12.621	4.526	-15.603
NF_3	-31.60	-59.785	-39.633	-36.356	-39.397	-67.368
PF_3	-229.10	-257.249	-212.087	-225.840	-211.489	-238.980
O_3	33.90	18.170	31.900	35.011	24.990	1.356
F_2O	5.90	-11.237	-0.299	1.559	-3.811	-22.051
ClF_3	-38.00	-72.834	-51.571	-52.986	-51.955	-78.687
$F_2C=CF_2$	-161.30	-207.703	-155.843	-159.606	-146.737	-194.820
$Cl_2C=CCl_2$	-5.50	-44.328	-20.173	-8.608	-4.517	-36.929
F_3C-CN	-118.40	-157.737	-122.052	-111.499	-105.193	-151.370
$HC\equiv C-CH_3$	44.40	33.040	34.399	47.161	51.471	29.706
$H_2C=C=CH_2$	45.40	31.531	31.755	44.410	48.469	26.459
C_3H_4 (cyclopropene)	67.80	53.706	57.504	68.072	72.655	49.078
$H_2C=CH-CH_3$	4.80	-1.474	-1.264	6.947	12.623	-6.801
C_3H_6 (cyclopropane)	12.80	1.396	8.259	12.987	20.397	-0.916
$CH_3-CH_2-CH_3$	-25.10	-28.142	-26.121	-22.437	-14.961	-32.013
C_4H_6 (Z-1,3-butadiene)	26.40	13.634	13.867	27.894	33.881	6.333
C_4H_6 (2-butyne)	34.80	20.143	24.366	37.197	44.650	15.145
C_4H_6 (methylenecyclopropane)	47.90	27.239	34.427	44.709	52.779	22.074
C_4H_6 (bicyclo[1.1.0]butane)	51.90	30.937	44.664	51.592	61.014	29.220
C_4H_6 (cyclobutene)	38.20	24.299	27.553	40.203	47.106	16.937
C_4H_8 (cyclobutane)	6.60	-4.156	0.278	8.732	17.125	-9.955
C_4H_8 (isobutene)	-4.10	-13.927	-11.089	-1.646	6.663	-18.953
C_4H_{10} (trans butane)	-30.10	-35.929	-31.830	-26.887	-17.168	-40.316
C_4H_{10} (isobutane)	-32.20	-38.133	-33.954	-28.505	-18.795	-41.416
C_5H_8 (spiropentane)	44.30	18.723	34.281	41.784	54.044	15.217
C_6H_6 (benzene)	19.90	-13.994	-2.470	15.665	26.629	-19.162
CH_2F_2	-107.70	-124.686	-101.736	-104.763	-97.680	-118.370
CHF_3	-166.30	-196.483	-157.638	-162.856	-152.177	-181.290
CH_2Cl_2	-22.60	-35.233	-26.941	-22.160	-19.601	-33.122

(continued)

$CHCl_3$	-24.50	-45.547	-30.981	-24.708	-22.366	-40.300
CH_3NH_2	-5.20	-0.345	-6.879	-0.943	-1.606	-13.358
CH_3CN	18.00	12.401	8.407	22.880	22.713	0.076
CH_3NO_2 (nitromethane)	-17.90	-37.856	-24.153	-19.286	-22.757	-53.916
CH_3ONO (methyl nitrite)	-16.10	-30.041	-21.883	-15.139	-20.183	-49.377
CH_3SiH_3	-7.00	-16.036	-6.800	-6.135	1.530	-0.877
HCO_2H	-90.40	-104.947	-91.534	-87.112	-84.704	-107.360
HCO_2CH_3	-85.50	-102.627	-86.026	-83.084	-78.494	-106.770
CH_3CONH_2	-57.00	-69.688	-64.263	-54.269	-51.913	-80.812
C_2H_5N (aziridine)	30.20	24.654	24.984	32.121	33.681	11.524
NCCN (cyanogen)	74.10	64.252	51.649	81.599	73.825	40.450
$NH(CH_3)_2$	-4.30	-1.660	-5.522	0.202	1.924	-16.221
$CH_3CH_2NH_2$	-11.30	-10.526	-14.584	-7.517	-5.753	-23.752
$H_2C{=}C{=}O$ (ketene)	-11.60	-31.045	-23.035	-12.453	-9.383	-33.102
C_2H_4O (oxirane)	-12.60	-24.125	-13.290	-11.693	-7.446	-29.497
CH_3CHO	-39.50	-49.028	-43.053	-36.818	-33.098	-54.041
$O{=}CH{-}CH{=}O$	-50.80	-66.233	-56.903	-45.639	-44.729	-74.662
CH_3CH_2OH	-56.10	-61.638	-55.276	-52.192	-47.377	-65.192
$(CH_3)_2O$	-44.00	-48.459	-41.330	-40.807	-36.143	-53.843
C_2H_4S (thiooxirane)	19.60	5.273	11.660	17.453	22.511	3.919
$(CH_3)_2S{=}O$	-36.20	-55.997	-39.608	-38.836	-30.968	-54.799
CH_3CH_2SH	-11.10	-17.467	-14.609	-9.449	-3.788	-18.527
$(CH_3)_2S$	-8.90	-16.199	-13.328	-8.385	-2.653	-17.588
$H_2C{=}CHF$	-34.00	-47.896	-37.003	-32.329	-26.945	-48.331
CH_3CH_2Cl	-26.60	-34.740	-29.472	-25.233	-20.150	-35.149
$H_2C{=}CHCl$	5.20	-6.787	-2.884	5.817	9.171	-8.483
$H_2C{=}CHCN$	43.20	33.539	28.593	49.584	49.271	18.622
$(CH_3)_2C{=}O$	-51.70	-65.965	-56.669	-49.418	-42.689	-69.890
CH_3CO_2H	-103.40	-122.421	-105.574	-100.199	-94.661	-123.520
CH_3CFO	-105.70	-128.170	-106.589	-103.283	-95.940	-125.650
CH_3COCl	-57.70	-79.868	-66.085	-58.235	-53.756	-79.307
$CH_3CH_2CH_2Cl$	-31.50	-42.484	-35.096	-29.626	-22.258	-43.378
$(CH_3)_2CHOH$	-65.20	-74.578	-65.572	-60.934	-53.626	-77.143
$CH_3{-}O{-}CH_2CH_3$	-51.70	-60.074	-50.276	-48.714	-41.522	-65.428
$(CH_3)_3N$	-6.60	-6.155	-7.238	-1.323	2.781	-21.221
C_4H_4O (furan)	-8.30	-38.192	-20.873	-10.117	-2.473	-42.264
C_4H_4S (thiophene)	27.50	-5.725	7.938	22.834	31.527	-4.752
C_4H_5N (pyrrole)	25.90	0.902	8.413	24.104	29.423	-11.188
C_5H_5N (pyridine)	33.60	4.729	10.949	30.645	36.441	-10.766
H_2	0.00	4.493	4.850	1.986	3.157	4.947
^2SH	34.20	39.944	35.129	40.254	37.588	35.543
$^2C{\equiv}CH$	135.80	125.853	125.875	135.948	135.720	124.683
$'HC{=}CH_2$ ($^2A''$)'	71.00	61.962	61.047	67.263	69.046	59.335
$'CH_3C{=}O$ ($^2A''$)'	-2.40	-16.083	-9.976	-3.976	-1.878	-21.411
$CH_2{-}OH$ (^2A)	-4.00	-11.396	-7.545	-6.202	-5.097	-14.430
$'CH_3O$ ($^2A''$)'	5.20	-0.159	0.928	0.986	2.184	-7.137
$'CH_3CH_2O$ ($^2A''''$)'	-2.90	-12.562	-8.903	-7.766	-4.145	-20.105
$'CH_3S$ ($^2A''$)'	29.80	23.540	24.205	26.301	28.828	21.779
$'CH_2CH_3$ ($^2A''$)'	28.60	22.509	22.729	24.480	28.598	21.021
$'(CH_3)_2CH$ ($^2A''$)'	21.10	11.186	14.330	16.516	23.285	8.579
$^2C(CH_3)_3$	12.00	-1.040	4.999	8.053	17.358	-3.895
NO_2 ($^2A_1$)	8.10	-10.821	-1.730	3.586	-3.898	-28.707

(continued)

$CH_3CH{=}C{=}CH_2$	38.80	22.346	24.748	38.121	44.673	15.859
C_5H_8 (isoprene)	18.00	1.873	4.734	20.050	28.549	-4.905
C_5H_{10} (cyclopentane twist)	-18.50	-32.623	-23.482	-15.759	-4.376	-36.684
C_5H_{12} (n-pentane)	-35.00	-43.655	-37.449	-31.267	-19.338	-48.550
$C(CH_3)_4$ (neopentane)	-40.10	-49.361	-43.095	-35.247	-23.318	-50.892
C_6H_8 (1,3-cyclohexadiene)	25.40	1.238	10.648	26.710	37.508	-5.951
C_6H_8 (1,4-cyclohexadiene)	25.00	1.417	11.513	26.940	38.072	-5.765
C_6H_{12} (cyclohexane chair)	-29.40	-45.918	-33.495	-25.613	-11.758	-49.255
C_6H_{14} (n-hexane)	-39.90	-51.383	-43.109	-35.655	-21.473	-56.791
C_6H_{14} (3-methylpentane)	-41.10	-51.807	-43.823	-35.561	-21.611	-56.054
$C_6H_5{-}CH_3$ (toluene)	12.00	-25.152	-11.050	8.311	21.885	-30.313
C_7H_{16} (n-heptane)	-44.80	-59.133	-48.758	-40.053	-23.639	-65.036
C_8H_8 (cyclooctatetraene)	70.70	35.530	45.540	71.916	83.800	23.184
C_8H_{18} (n-octane)	-49.80	-66.853	-54.397	-44.438	-25.807	-73.277
$C_{10}H_8$ (naphtalene)	35.90	-28.053	-4.032	26.108	44.554	-33.814
$C_{10}H_8$ (azulene)	69.10	6.066	26.951	58.747	75.684	-3.610
$CH_3CO_2CH_3$ (Z-methylacetate)	-98.40	-119.531	-99.471	-95.607	-87.919	-122.370
$(CH_3)_3COH$ (t-butanol)	-74.70	-87.753	-76.282	-69.469	-59.790	-88.315
$C_6H_5NH_2$ (aniline)	20.80	-15.180	-5.921	16.532	25.034	-28.585
C_6H_5OH (phenol)	-22.30	-64.696	-44.818	-26.372	-14.866	-68.395
C_4H_6O (divinyl ether)	-3.30	-23.500	-13.015	-2.028	4.555	-31.248
C_4H_8O (tetrahydrofuran)	-44.00	-60.281	-44.831	-40.906	-31.721	-64.869
C_5H_8O (cyclopentanone)	-45.90	-72.337	-55.277	-44.448	-33.523	-76.219
$C_6H_4O_2$ (benzoquinone)	-29.40	-73.116	-52.360	-28.447	-20.463	-82.738
$C_6H_4N_2$ (pyrimidine)	46.80	21.207	22.637	43.594	44.504	-4.099
$(CH_3)_2SO_2$	-89.20	-125.035	-87.730	-92.920	-79.620	-111.970
C_6H_5Cl (chlorobenzene)	12.50	-31.009	-13.130	6.805	18.129	-32.272
$NC(CH_2)_2CN$ (succinonitrile)	50.10	35.406	30.909	60.277	58.642	12.115
$C_4H_4N_2$ (pyrazine)	46.90	25.873	26.274	47.752	48.091	-0.444
C_4H_4O (3-butyn-2-one)	15.60	-6.419	2.682	21.240	26.045	-11.364
C_4H_6O (E-crotonaldehyde)	-24.00	-44.573	-36.133	-23.368	-16.676	-52.464
$C_4H_6O_3$ (acetic anhydride)	-136.80	-174.964	-143.461	-135.400	-125.357	-176.620
C_4H_6S (2,5-dihydrothiophene)	20.80	-1.796	7.730	20.135	28.400	-4.516
$(CH_3)_2CHCN$	5.60	-3.833	-3.628	13.625	17.802	-16.590
C_4H_8O (methyl ethyl ketone)	-57.10	-74.061	-62.376	-54.148	-45.135	-78.262
$(CH_3)_2CHCHO$	-51.60	-65.209	-55.055	-46.154	-37.985	-70.713
$C_4H_8O_2$ (1,4-dioxane)	-75.50	-97.437	-73.368	-72.181	-63.126	-102.360
C_4H_8S (tetrahydrothiophene)	-8.20	-25.766	-15.081	-6.693	3.034	-26.855
$(CH_3)_3CCl$	-43.50	-58.844	-48.443	-40.635	-30.849	-56.428
C_4H_9Cl (n-butyl chloride)	-37.00	-50.086	-40.642	-33.915	-24.363	-51.531
C_4H_9N (tetrahydropyrrole)	-0.80	-10.175	-6.442	3.272	9.154	-24.256
$C_4H_9NO_2$ (2-nitrobutane)	-39.10	-67.633	-46.581	-38.079	-34.502	-82.942
$(CH_3CH_2)_2O$	-60.30	-71.589	-59.135	-56.532	-46.809	-76.903
$CH_3CH(OCH_3)_2$ (dimethyl acetal)	-93.10	-104.592	-83.757	-82.767	-73.070	-109.520
$(CH_3)_3CSH$	-26.20	-39.437	-31.938	-22.972	-12.825	-38.140
$C_4H_{10}S_2$ (diethyl disulfide)	-17.90	-36.405	-24.990	-17.207	-6.018	-37.915
$(CH_3)_3CNH_2$	-28.90	-34.378	-33.745	-22.659	-16.267	-44.723
$(CH_3)_4Si$	-55.70	-75.634	-57.397	-50.729	-34.157	-55.118
C_5H_6S (2-methyl thiophene)	20.00	-16.435	-0.170	15.599	26.934	-15.831
C_5H_7N (N-methyl pyrrole)	24.60	-2.485	8.162	23.425	31.220	-15.647
$C_5H_{10}O$ (tetrahydropyran)	-53.40	-72.597	-54.170	-49.696	-38.147	-76.563
$(CH_3CH_2)_2C{=}O$	-61.60	-82.060	-67.999	-58.788	-47.500	-86.539

(continued)

$C_5H_{10}O_2$ (isopropyl acetate)	-115.10	-143.237	-117.851	-111.445	-98.807	-144.960
$C_5H_{10}S$ (tetrahydrothiopyran)	-15.20	-35.658	-22.150	-13.289	-1.193	-36.612
$C_5H_{11}N$ (piperidine)	-11.30	-23.583	-16.536	-6.665	1.711	-36.957
$C_5H_{12}O$ (t-butyl methyl ether)	-67.80	-82.590	-68.006	-62.551	-50.773	-84.845
$C_6H_4F_2$ (1,3-difluorobenzene)	-73.90	-131.950	-93.046	-79.584	-63.533	-126.300
$C_6H_4F_2$ (1,4-difluorobenzene)	-73.30	-130.721	-92.339	-78.777	-62.864	-125.660
C_6H_5F (fluorobenzene)	-27.40	-73.315	-48.252	-32.369	-18.879	-73.164
$C_6H_{14}O$ (diisopropyl ether)	-76.30	-94.154	-77.033	-70.916	-56.521	-97.458
PF_5	-381.10	-437.129	-355.224	-375.692	-348.671	-386.360
SF_6	-291.70	-372.898	-271.978	-297.199	-272.287	-317.050
P_4	14.10	-5.639	-0.339	9.887	14.664	-6.651
SO_3	-94.60	-136.234	-95.425	-100.385	-94.494	-124.720
SCl_2	-4.20	-18.357	-9.146	-6.497	-5.958	-18.976
$POCl_3$	-133.80	-171.895	-135.374	-136.967	-126.980	-150.050
PCl_5	-86.10	-129.612	-94.766	-91.638	-84.449	-106.150
Cl_2O_2S	-84.80	-131.024	-89.208	-92.830	-85.974	-115.460
PCl_3	-69.00	-91.695	-72.503	-71.039	-66.634	-83.314
Cl_2S_2	-4.00	-31.588	-17.465	-14.684	-12.866	-31.356
$SiCl_2$ ($^1A_1$)	-40.30	-61.304	-43.528	-39.538	-34.144	-45.238
CF_3Cl	-169.70	-211.456	-165.326	-167.895	-156.905	-190.150
C_2F_6	-320.90	-390.725	-306.329	-316.239	-294.755	-352.990
$^2CF_3$	-111.80	-147.481	-108.729	-115.035	-105.651	-133.220
$^2C_6H_5$ (phenyl radical)	80.50	39.970	52.824	69.965	78.823	36.489
ME		-16.845	-5.745	0.870	6.194	-18.020
MAD		17.864	7.675	3.671	7.881	18.693

TABLE VI. Bond lengths in $\AA$ for the T46-R molecular test set from different approximate XC functionals with VASP. T46-R is a subset of the T96-R test set [6, 7]; see main text.

Molecule	Exptl.	MVS	MVS-L(CRopt)	r^{2}SCAN	r^{2}SCAN-L	PBE
H_2	0.741	0.745	0.742	0.741	0.742	0.750
Li_2	2.673	2.690	2.725	2.748	2.717	2.728
LiH	1.595	1.621	1.600	1.607	1.593	1.604
LiF	1.564	1.593	1.584	1.581	1.584	1.579
LiCl	2.021	2.040	2.022	2.033	2.025	2.025
LiO	1.688	1.713	1.703	1.703	1.700	1.701
Be_2	2.440	2.522	2.415	2.470	2.395	2.432
BeF	1.361	1.365	1.355	1.364	1.315	1.370
BeO	1.331	1.329	1.323	1.326	1.327	1.335
BeS	1.742	1.737	1.730	1.739	1.736	1.748
B_2	1.590	1.590	1.607	1.616	1.610	1.618
BH	1.232	1.244	1.245	1.239	1.238	1.251
BF	1.263	1.267	1.273	1.267	1.274	1.279
BCl	1.715	1.714	1.715	1.717	1.719	1.726
BN	1.281	1.320	1.324	1.321	1.322	1.332
BO	1.204	1.205	1.206	1.204	1.208	1.213
BS	1.609	1.603	1.602	1.608	1.606	1.616
C_2	1.242	1.248	1.249	1.251	1.252	1.262
CH	1.120	1.126	1.128	1.126	1.126	1.137
CH_4	1.087	1.092	1.088	1.089	1.089	1.096
CCl	1.645	1.633	1.640	1.646	1.651	1.653
CO	1.128	1.135	1.140	1.134	1.140	1.143
N_2	1.098	1.106	1.110	1.106	1.109	1.113
NH	1.036	1.040	1.037	1.041	1.043	1.051
NF	1.317	1.313	1.342	1.332	1.344	1.340
O_2	1.208	1.207	1.229	1.222	1.233	1.233
OH	0.970	0.971	0.972	0.977	0.979	0.987
F_2	1.412	1.377	1.431	1.410	1.426	1.423
NaH	1.887	1.915	1.933	1.897	1.885	1.896
NaF	1.926	1.918	1.953	1.925	1.926	1.941
NaCl	2.361	2.353	2.381	2.361	2.352	2.368
NaO	2.052	2.048	2.089	2.054	2.054	2.071
MgH	1.730	1.734	1.730	1.742	1.731	1.754
MgF	1.750	1.752	1.764	1.761	1.773	1.778
MgCl	2.196	2.189	2.190	2.205	2.200	2.220
MgO	1.748	1.722	1.736	1.736	1.737	1.750
Al_2	2.466	2.435	2.434	2.467	2.458	2.487
AlF	1.654	1.651	1.663	1.666	1.669	1.685
AlO	1.618	1.606	1.619	1.623	1.626	1.634
SiH	1.520	1.520	1.518	1.527	1.522	1.543
SiF	1.601	1.598	1.612	1.616	1.620	1.635
SiO	1.510	1.506	1.515	1.514	1.519	1.529
P_2	1.893	1.871	1.871	1.881	1.884	1.900
Cl_2	1.988	1.954	1.981	1.980	1.990	1.994
ClF	1.628	1.604	1.654	1.633	1.648	1.649
ClO	1.570	1.537	1.572	1.566	1.578	1.575
ME		0.001	0.007	0.008	0.006	0.016
MAD		0.013	0.014	0.010	0.012	0.016

IV. SOLID TEST SET RESULTS

Here we provide the solid-by-solid tabulation for the KS (or gKS) band gaps (21 systems), equilibrium lattice constants and cohesive energies (55 solids), and bulk moduli (44 cubic solids) for MVS and MVS-L(CRopt) compared wtih r^{2}SCAN, r^{2}SCAN-L, and PBE. All calculations done with VASP 5.4.4.

TABLE VII. Band Gap (eV) of 21 insulators and semiconductors. Experimental lattice parameters were used with all functionals. Experimental values Exptl. and lattice constants are from Ref. [10].

Solid	Exptl.	MVS	MVS-L(CRopt)	MVS-L(CR)	r^{2}SCAN	r^{2}SCAN-L	PBE
C	5.50	4.11	4.16	3.97	4.32	4.23	4.13
Si	1.17	0.90	0.87	0.81	0.78	0.83	0.58
Ge	0.74	1.14	0.87	0.70	0.50	0.35	0.09
SiC	2.42	1.87	1.66	1.42	1.75	1.69	1.38
BN	6.36	5.09	5.02	4.67	5.00	4.81	4.48
BP	2.10	1.43	1.35	1.39	1.45	1.42	1.24
AlN	4.90	4.12	3.68	3.39	3.97	3.73	3.35
AlP	2.50	2.15	1.95	1.75	1.92	1.88	1.59
AlAs	2.23	2.17	1.88	1.68	1.80	1.73	1.44
GaN	3.28	2.53	1.95	1.59	2.19	1.90	1.74
GaP	2.35	2.14	1.95	1.76	1.89	1.86	1.62
GaAs	1.52	2.16	1.76	1.24	1.09	0.90	0.61
InP	1.42	1.92	1.47	1.16	1.18	1.01	0.75
InAs	0.42	1.36	0.87	0.46	0.25	0.12	0.00
InSb	0.24	1.57	0.66	0.52	0.27	0.16	0.00
LiH	4.94	4.07	3.64	3.47	3.72	3.60	3.07
LiF	14.20	10.66	10.29	9.39	10.19	9.66	9.20
LiCl	9.40	7.85	7.38	7.20	7.31	6.97	6.41
NaF	11.50	7.77	6.96	6.14	7.21	6.70	6.37
NaCl	8.50	6.61	6.07	5.81	5.98	5.67	5.19
MgO	7.83	5.97	5.72	4.99	5.65	5.28	4.77
ME		-0.76	-1.11	-1.43	-1.20	-1.38	-1.69
MAD		1.12	1.24	1.46	1.20	1.38	1.69

TABLE VIII. Equilibrium lattice constants, $a_0(\AA)$ compared for 55 solids. Experimental values Exptl. are from Ref. [8], include zero-point effects.

Solid	Exptl.	MVS	MVS-L(CRopt)	MVS-L(CR)	r^{2}SCAN	r^{2}SCAN-L	PBE
C	3.553	3.548	3.572	3.576	3.562	3.569	3.572
Si	5.421	5.386	5.415	5.392	5.440	5.430	5.469
Ge	5.644	5.585	5.613	5.583	5.681	5.709	5.782
Sn	6.477	6.459	6.546	6.489	6.565	6.593	6.653
SiC	4.346	4.306	4.344	4.346	4.355	4.356	4.379
BN	3.592	3.589	3.623	3.614	3.614	3.619	3.626
BP	4.525	4.500	4.560	4.549	4.533	4.538	4.547
AlN	4.368	4.328	4.357	4.348	4.368	4.366	4.402
AlP	5.451	5.417	5.453	5.420	5.472	5.463	5.506
AlAs	5.649	5.605	5.654	5.598	5.677	5.678	5.733
GaN	4.520	4.476	4.534	4.524	4.527	4.548	4.589
GaP	5.439	5.400	5.472	5.445	5.467	5.485	5.534
GaAs	5.640	5.588	5.675	5.639	5.672	5.701	5.762
InP	5.858	5.858	5.925	5.893	5.947	5.948	6.001
InAs	6.047	6.033	6.105	6.035	6.131	6.151	6.211
InSb	6.468	6.481	6.503	6.428	6.569	6.581	6.647
LiH	3.979	4.019	3.977	3.971	4.012	3.988	4.016
LiF	3.972	3.971	4.008	3.967	3.994	4.003	4.071
LiCl	5.070	5.065	5.077	4.997	5.111	5.079	5.153
NaF	4.582	4.452	4.569	4.485	4.503	4.508	4.632
NaCl	5.569	5.474	5.555	5.436	5.545	5.519	5.655
MgO	4.189	4.158	4.189	4.170	4.196	4.192	4.248
Li	3.443	3.484	3.444	3.443	3.479	3.432	3.439
Na	4.214	4.194	4.190	4.097	4.200	4.140	4.193
K	5.212	5.174	5.325	5.242	5.350	5.269	5.286
Rb	5.577	5.691	5.628	5.667	5.751	5.667	5.669
Cs	6.039	6.418	6.060	6.077	6.271	6.174	6.161
Ca	5.556	5.322	5.538	5.461	5.574	5.503	5.525
Ba	5.002	5.145	5.003	4.988	5.077	5.061	5.030
Sr	6.040	6.062	6.106	6.034	6.101	6.051	6.027
Al	4.018	3.931	3.968	3.967	3.988	3.977	4.040
Fe	2.853	2.844	2.836	2.776	2.865	2.841	2.829
Co	3.524	3.484	3.509	3.445	3.514	3.516	3.517
Ni	3.508	3.476	3.508	3.437	3.479	3.509	3.518
Sc	3.270	3.237	3.256	3.214	3.260	3.226	3.249
Y	3.594	3.625	3.617	3.583	3.623	3.600	3.592
Ti	2.915	2.860	2.905	2.856	2.887	2.877	2.893
Zr	3.198	3.199	3.242	3.213	3.215	3.216	3.210
Hf	3.151	3.119	3.196	3.148	3.141	3.150	3.158
V	3.021	2.942	2.969	2.950	2.965	2.965	2.978
Nb	3.294	3.301	3.343	3.311	3.320	3.322	3.322
Ta	3.299	3.267	3.343	3.301	3.290	3.301	3.309
Mo	3.141	3.117	3.173	3.145	3.144	3.149	3.151
W	3.160	3.132	3.212	3.160	3.156	3.167	3.172
Tc	2.716	2.692	2.738	2.715	2.716	2.723	2.725
Re	2.744	2.731	2.790	2.740	2.750	2.760	2.764
Ru	2.669	2.658	2.699	2.672	2.672	2.683	2.683
Os	2.699	2.687	2.744	2.711	2.708	2.723	2.723
Rh	3.794	3.785	3.841	3.781	3.804	3.824	3.824
Ir	3.831	3.840	3.905	3.850	3.852	3.877	3.872
Pd	3.876	3.867	3.939	3.864	3.912	3.938	3.939
Pt	3.913	3.924	3.987	3.911	3.943	3.974	3.968
Cu	3.595	3.505	3.633	3.529	3.582	3.611	3.634
Ag	4.062	4.035	4.142	4.025	4.108	4.141	4.147
Au	4.062	4.092	4.147	4.070	4.128	4.167	4.156
ME		-0.015	0.024	-0.019	0.026	0.022	0.046
MAD		0.045	0.034	0.032	0.037	0.039	0.053

TABLE IX. Cohesive energies, $E_{coh}(eV/atom)$ of 55 solids. Experimental values Exptl. are from Ref. [8]. They include zero-point effects.

Solid	Exptl.	MVS	MVS-L(CRopt)	MVS-L(CR)	r^{2}SCAN	r^{2}SCAN-L	PBE
C	7.550	7.885	7.595	7.741	7.467	7.354	7.714
Si	4.680	5.093	4.765	4.948	4.675	4.505	4.561
Ge	3.890	4.228	4.054	4.387	3.891	3.534	3.725
Sn	3.160	3.567	3.267	3.667	3.295	3.076	3.200
SiC	6.480	7.048	6.470	6.606	6.432	6.231	6.408
BN	6.760	7.141	6.823	6.911	6.788	6.704	6.936
BP	5.140	5.575	5.460	5.514	5.281	5.141	5.293
AlN	5.850	6.257	5.895	5.914	5.809	5.717	5.711
AlP	4.320	4.676	4.543	4.561	4.249	4.137	4.093
AlAs	3.820	4.212	4.029	4.119	3.867	3.682	3.693
GaN	4.550	5.071	4.561	4.684	4.465	4.308	4.385
GaP	3.610	4.155	3.876	3.969	3.642	3.444	3.478
GaAs	3.340	3.718	3.413	3.567	3.328	3.064	3.161
InP	3.470	3.803	3.464	3.660	3.245	3.103	3.123
InAs	3.080	3.452	3.063	3.324	3.026	2.811	2.893
InSb	2.810	3.016	2.589	3.057	2.729	2.580	2.650
LiH	2.490	2.361	2.414	2.399	2.420	2.420	2.348
LiF	4.460	4.549	4.315	4.203	4.395	4.293	4.321
LiCl	3.590	3.626	3.469	3.526	3.467	3.403	3.352
NaF	3.970	4.258	3.950	3.851	3.992	3.875	3.860
NaCl	3.340	3.516	3.324	3.344	3.260	3.177	3.088
MgO	5.200	5.651	5.220	5.255	5.249	5.141	4.998
Li	1.670	1.492	1.652	1.655	1.569	1.605	1.600
Na	1.120	1.076	1.097	1.186	1.041	1.017	1.038
K	0.940	0.855	0.938	1.060	0.841	0.826	0.857
Rb	0.860	1.015	0.824	0.984	0.734	0.733	0.760
Cs	0.810	0.726	0.828	0.883	0.661	0.637	0.693
Ca	1.870	2.172	2.263	2.313	1.917	1.986	1.846
Ba	1.910	2.046	2.154	2.279	1.817	1.845	1.703
Sr	1.730	2.130	2.039	2.119	1.786	1.729	1.604
Al	3.430	4.058	3.762	3.931	3.609	3.437	3.442
Fe	4.300	5.689	4.805	5.579	4.635	4.501	4.871
Co	4.420	5.888	4.719	5.442	4.871	4.450	5.165
Ni	4.480	5.819	4.352	5.189	4.645	4.322	4.732
Sc	3.930	4.702	4.193	4.595	4.017	3.874	4.142
Y	4.390	4.592	4.194	4.533	4.014	3.775	4.100
Ti	4.880	5.704	5.001	5.860	5.060	4.925	5.442
Zr	6.270	4.610	4.700	5.467	4.378	4.656	5.344
Hf	6.460	6.880	6.392	6.792	6.434	6.058	6.455
V	5.350	5.365	4.871	5.906	4.660	4.807	6.520
Nb	7.600	6.746	6.070	7.104	6.361	6.187	6.919
Ta	8.130	8.490	7.748	8.576	8.251	7.733	8.314
Mo	6.860	5.918	5.718	6.833	5.690	5.637	6.351
W	8.940	8.456	7.322	8.731	8.067	7.631	8.485
Tc	6.880	6.808	6.374	7.337	6.337	6.176	6.971
Re	8.050	8.050	6.383	8.464	7.514	7.165	7.815
Ru	6.770	7.183	6.313	7.290	6.338	6.040	6.773
Os	8.200	8.217	7.054	8.535	8.072	7.530	8.356
Rh	5.780	6.767	5.216	6.014	5.826	5.287	5.748
Ir	6.990	7.099	5.697	7.396	7.142	6.556	7.494
Pd	3.930	4.722	3.489	4.785	4.163	3.696	3.738
Pt	5.870	6.043	4.490	5.938	5.716	5.067	5.531
Cu	3.510	4.853	3.446	4.319	3.855	3.487	3.479
Ag	2.960	3.560	2.287	3.353	2.873	2.484	2.511
Au	3.830	4.160	2.773	3.860	3.417	2.919	3.033
ME		0.292	-0.236	0.270	-0.134	-0.331	-0.070
MAD		0.457	0.385	0.345	0.238	0.346	0.252

TABLE X. Bulk modulus, $B_0(GPa)$ of 44 cubic solids. Experimental values Exptl. are from Ref. [9]. Theoe values were obtained by subtracting the zero-point phonon effect from the experimental zero-temperature values.

Solid	Exptl.	MVS	MVS-L(CRopt)	MVS-L(CR)	r^{2}SCAN	r^{2}SCAN-L	PBE
C	454.700	463.777	447.252	460.565	449.244	433.521	433.061
Si	101.300	108.168	109.695	111.381	97.272	92.931	88.798
Ge	79.400	93.554	89.04	91.21	73.095	68.252	59.083
Sn	42.800	57.047	46.402	46.659	43.526	39.541	36.172
SiC	229.100	251.785	231.915	234.699	226.927	219.406	212.361
BN	410.200	417.275	382.791	374.969	389.901	377.584	372.412
BP	168.000	178.003	158.897	168.87	170.052	165.254	161.851
AlN	206.000	229.842	201.092	225.381	212.380	207.043	193.670
AlP	87.400	101.278	95.904	95.813	91.095	88.727	82.678
AlAs	75.000	88.049	79.327	89.555	76.387	73.888	67.252
GaN	213.000	240.326	207.43	205.915	211.373	197.740	182.466
GaP	89.600	106.971	94.55	99.173	90.466	87.723	78.712
GaAs	76.700	91.906	78.485	83.761	73.804	69.055	62.019
InP	72.000	84.672	77.89	79.436	70.286	66.749	60.930
InAs	58.600	73.560	62.083	62.277	58.929	56.038	49.497
InSb	46.100	55.202	49.155	56.809	44.017	40.168	37.074
LiH	40.100	35.323	45.576	37.841	36.485	38.008	36.343
LiF	76.300	81.254	95.111	81.431	78.714	78.913	67.393
LiCl	38.700	36.474	35.257	46.929	35.064	37.084	31.863
NaF	53.100	65.172	56.365	58.419	59.895	59.433	47.016
NaCl	27.600	31.655	29.698	34.309	29.140	31.114	24.264
MgO	169.800	182.891	173.361	177.299	170.619	170.667	151.596
Li	13.100	14.009	20.164	15.034	13.116	14.902	13.957
Na	7.900	8.299	8.796	10.788	7.930	8.527	7.877
K	3.800	5.979	4.453	4.466	3.422	3.900	3.593
Rb	3.600	3.180	3.576	4.259	2.631	2.689	2.798
Cs	2.300	1.767	2.101	2.425	1.867	2.288	1.960
Ca	15.900	33.749	19.378	21.82	17.595	19.379	17.458
Ba	10.600	8.556	11.076	12.609	8.569	11.738	8.844
Sr	12.000	11.840	14.11	13.298	11.832	12.184	11.598
Al	77.100	116.650	90.633	95.273	92.466	90.178	77.996
Ni	192.500	260.509	199.791	263.721	216.804	249.963	196.258
V	165.800	210.292	214.221	159.28	201.269	200.164	187.488
Nb	173.200	184.327	162.17	203.799	178.078	176.731	173.453
Ta	202.700	219.987	234.442	196.672	210.119	208.545	201.055
Mo	276.200	294.933	244.192	277.258	281.995	269.301	268.535
W	327.500	349.759	302.695	303.653	329.918	316.703	312.477
Rh	277.100	296.668	278.289	303.514	281.584	257.708	258.115
Ir	362.200	388.070	342.792	366.851	378.905	343.185	349.818
Pd	187.200	216.254	167.812	217.532	186.087	165.310	169.148
Pt	285.500	286.179	253.819	311.54	273.641	243.359	249.877
Cu	144.300	260.458	197.176	193.439	159.969	146.405	138.007
Ag	105.700	125.441	95.960	131.872	103.562	92.108	90.318
Au	182.000	177.135	157.253	189.493	153.795	133.245	139.596
ME		16.012	0.647	8.582	1.367	-4.008	-9.704
MAD		16.695	11.143	12.262	5.963	10.345	11.022

V. REPARMETRIZATION OF THE CR DE-ORBITALIZER

The question of parameter optimization in the de-orbitalizer may be germane. The CRopt parameters were set by optimization against the Hartree-Fock densities of the first 18 neutral atoms [11]. It is possible instead to reparametrize consistently, that is, against the densities from the parent XC functional [12]. We did so for MVS following essentially the procedure of Ref. 11 with NWChem. The reparametrized de-orbitalizer is referred to as "CRc" (latter "c" for "consistent"). The resulting parameter values are $a = -0.33134$ and $b = 2.65093$. The a value is changed 12.1% compared to the CRopt value (-0.295491), while b is changed only 1.34% compared to the CRopt value (2.615740). These parameter changes nevertheless have an effect on the results obtained for the molecular test sets.

The performance of the reparametrized version (MVS-L-CRc) on the molecular test set gave heat- of-formation MAD as 7.86 kcal/mol, bond distances as 0.013 Å, and frequencies, 43.425 cm^{-1}. A comparison with the results tabulated in the main paper shows that the CRc de-orbitalization is not quite as good as CRopt. The two are very close on bond distances and frequencies, but for heats of formation, CRopt outperforms CRc by 27%. It is not straight-forward to identify the reason for that difference but we speculate that it may be associated with the more realistic atomic densities used for the CRopt parameters.

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