*GW*100: Benchmarking G_0W_0 for molecular systems, Supplementary material

Michiel J. van Setten,[*,†,‡] Fabio Caruso,[¶,§] Sahar Sharifzadeh,[‖] Xinguo Ren,[#,@] Matthias Scheffler,[@] Fang Liu,[△] Johannes Lischner,[∇,1] Lin Lin,[2] Jack R. Deslippe,[3] Steven G. Louie,[4,∇] Chao Yang,[2] Florian Weigend,[5,6] Jeffrey B. Neaton,[1,‖,7] Ferdinand Evers,[8] and Patrick Rinke[9,@]

Nanoscopic Physics, Institute of Condensed Matter and Nanosciences, Université Catholique de Louvain, Louvain-la-Neuve, 1348, Belgium, Institute of Nanotechnology, Karlsruhe Institute of Technology Campus North, Karlsruhe, 76344 Germany, Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, 4195, Germany, Department of Materials, University of Oxford, Oxford, OX1 3PH, United Kingdom, Molecular Foundry, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA, Department of Electrical and Computer Engineering, Boston University, Boston, MA 02215, USA, Key Laboratory of Quantum Information, University of Science and Technology of China, Hefei, 230026, China, Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, 14195, Germany, School of Applied Mathematics, Central University of Finance and Economics, Beijing, Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA, Department of Physics, University of California, Berkeley, CA 94720, USA, Computational Research Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA, National Energy Research Scientific Computing Center, Berkeley, CA 94720, USA, Department of Physics, University of California, Berkeley, CA 94720, USA , Institute of Nanotechnology, Karlsruhe Institute of Technology Campus North, Karlsruhe, 76344, Germany, Institute of Physical Chemistry, Karlsruhe Institute of Technology Campus South, Karlsruhe, 76021, Germany, Kavli Energy NanoSciences Institute at Berkeley, Berkeley, CA 94720 USA, Institute of Theoretical Physics, University of Regensburg, Regensburg, 93040, Germany, and COMP/Department of Applied Physics, Aalto University School of Science, Aalto, 00076, Finland

1 Convergence studies

This section contains various convergence studies of critical numerical parameters for four representative molecules.

1.1 Basis set extrapolation

Figure 1 addresses the basis set extrapolation used for the local orbital calculations. Figures 2-5 compare the extrapolation of the inverse of the number of basis functions to the inverse of the cube of the cardinal number of the basis set. The difference between the two extrapolation schemes is used for all molecules as an estimate of the extrapolation error. To validate this error estimate Figures 2-5 also contain results calculated using Dunning basis sets of triple, quadruple and quintuple zeta quality.[1]

*To whom correspondence should be addressed

†Nanoscopic Physics, Institute of Condensed Matter and Nanosciences, Université Catholique de Louvain, Louvain-la-Neuve, 1348, Belgium

‡Institute of Nanotechnology, Karlsruhe Institute of Technology Campus North, Karlsruhe, 76344 Germany

¶Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, 4195, Germany

§Department of Materials, University of Oxford, Oxford, OX1 3PH, United Kingdom

‖Molecular Foundry, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA

⊥Department of Electrical and Computer Engineering, Boston University, Boston, MA 02215, USA

#Key Laboratory of Quantum Information, University of Science and Technology of China, Hefei, 230026, China

@Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, 14195, Germany

△School of Applied Mathematics, Central University of Finance and Economics, Beijing

▽Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA

[1]Department of Physics, University of California, Berkeley, CA 94720, USA

[2]Computational Research Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA

[3]National Energy Research Scientific Computing Center, Berkeley, CA 94720, USA

[4]Department of Physics, University of California, Berkeley, CA 94720, USA

[5]Institute of Nanotechnology, Karlsruhe Institute of Technology Campus North, Karlsruhe, 76344, Germany

[6]Institute of Physical Chemistry, Karlsruhe Institute of Technology Campus South, Karlsruhe, 76021, Germany

[7]Kavli Energy NanoSciences Institute at Berkeley, Berkeley, CA 94720 USA

[8]Institute of Theoretical Physics, University of Regensburg, Regensburg, 93040, Germany

[9]COMP/Department of Applied Physics, Aalto University School of Science, Aalto, 00076, Finland

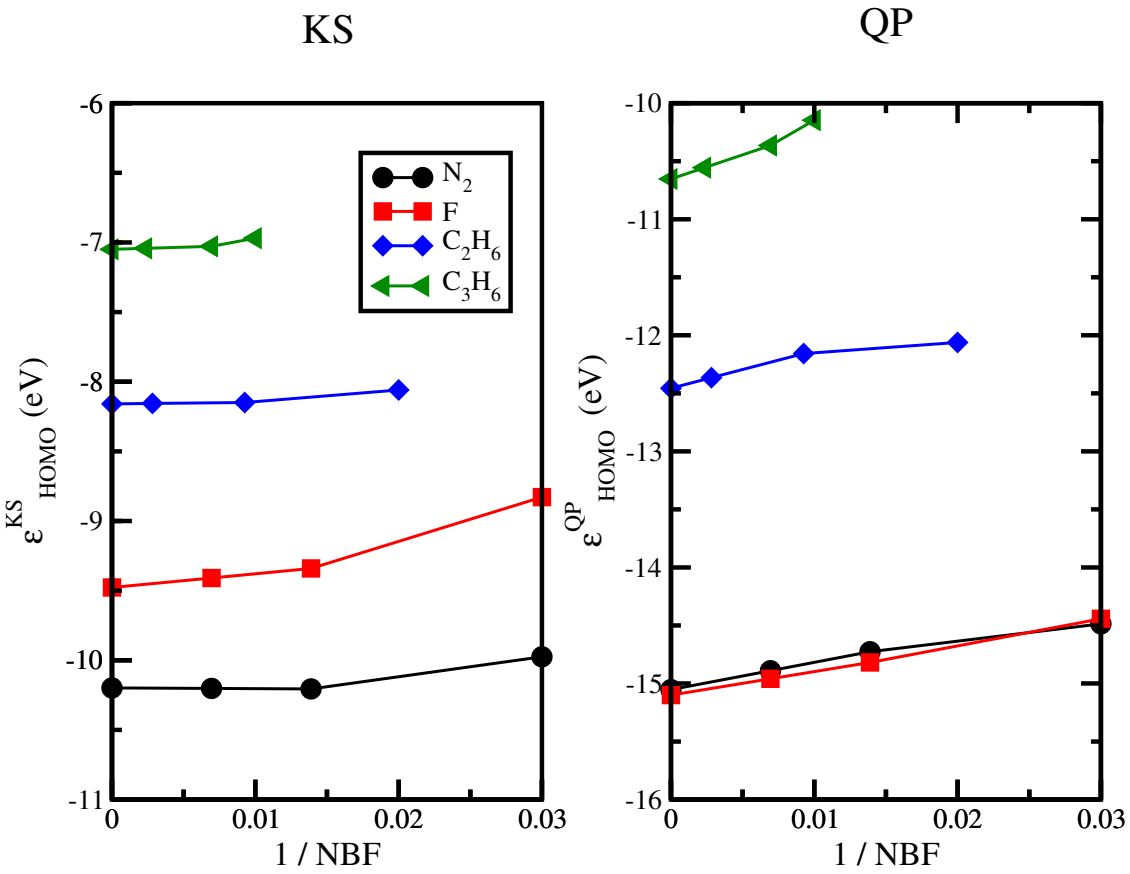

Figure 1: Basis set extrapolation of the KS and QP homo energies. The extrapolation is obtained from a linear extrapolation of the TZVP and QZVP results in the inverse of the number of basis functions (NBF).

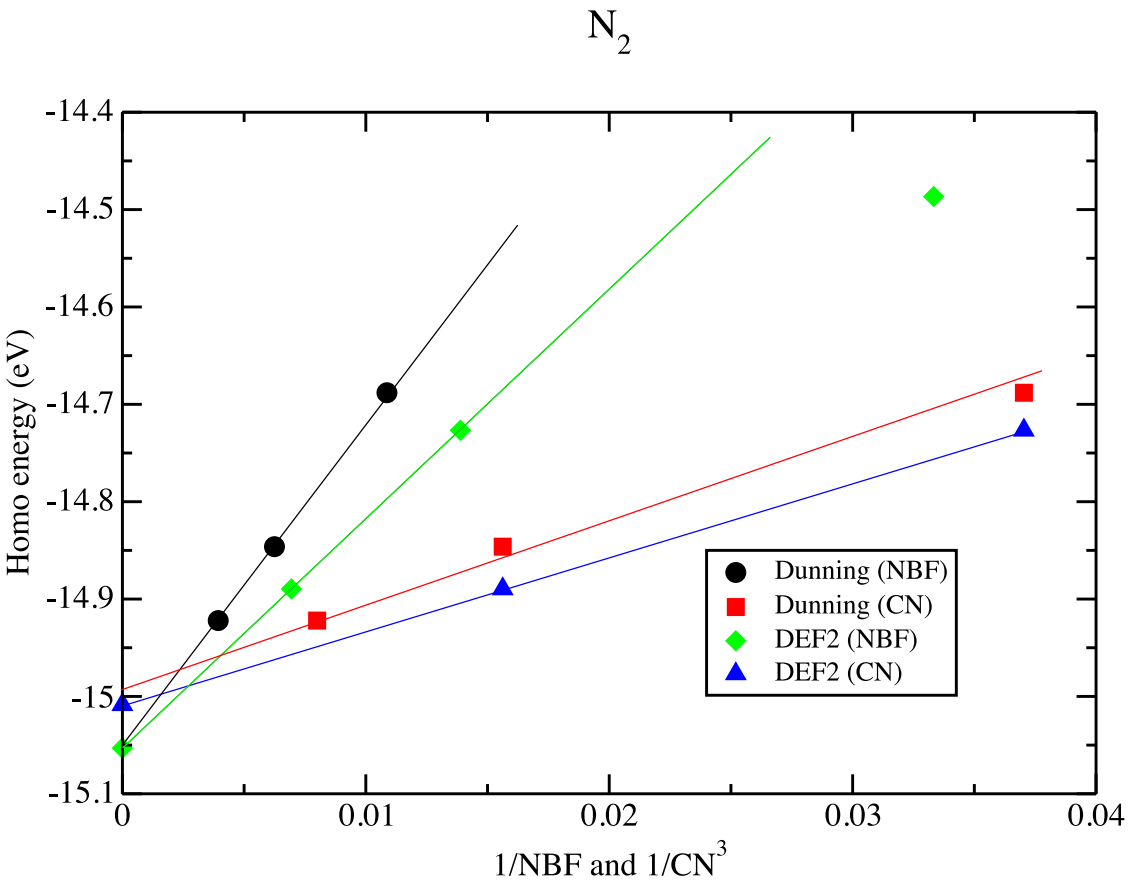

Figure 2: Basis set extrapolation of the QP homo energies comparing the def2 and Dunning basis sets (triple, quadruple, and quintuple). The points at 0 are the results of the extrapolation used in this work. For both basis sets we compare the extrapolation in the absolute size of the basis set (NBF) and the cube of the cardinal number.

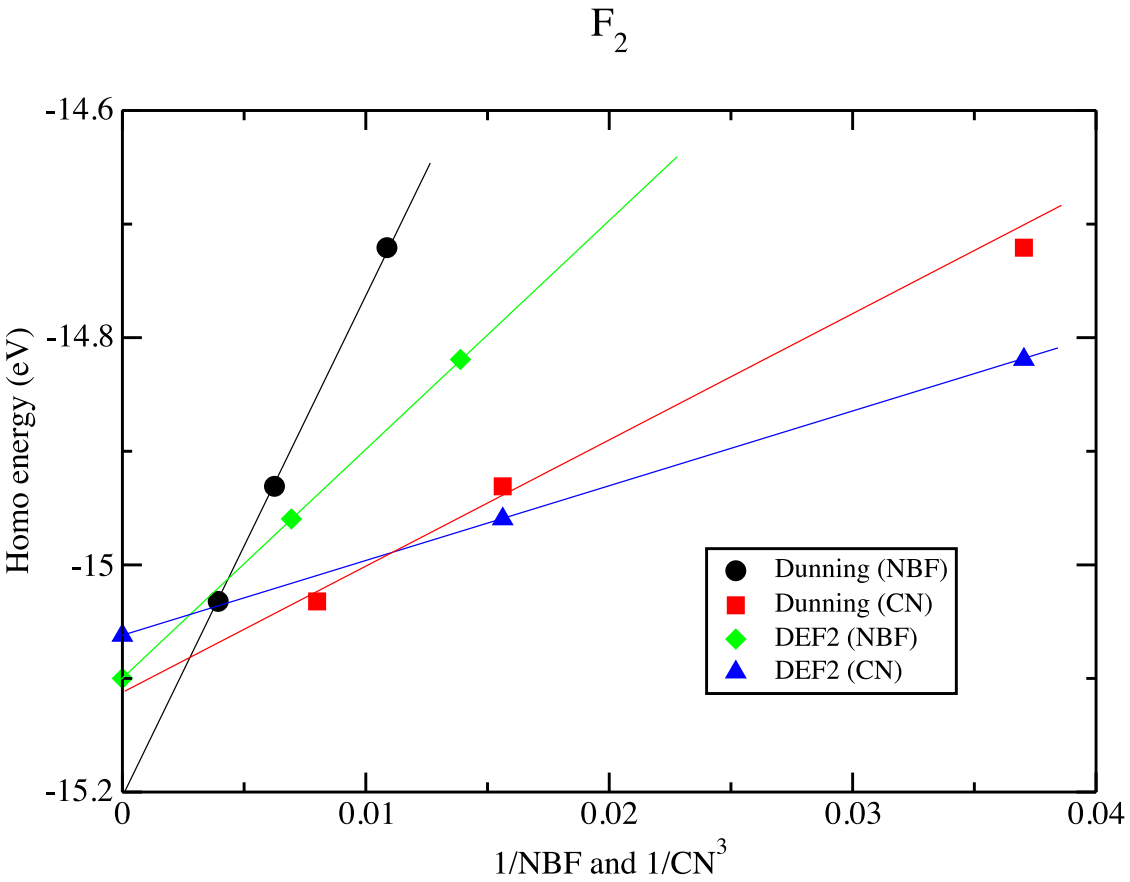

Figure 3: Basis set extrapolation of the QP homo energies comparing the def2 and Dunning basis sets (triple, quadruple, and quintuple). The points at 0 are the results of the extrapolation used in this work. The same as Figure 2, but for F_2.

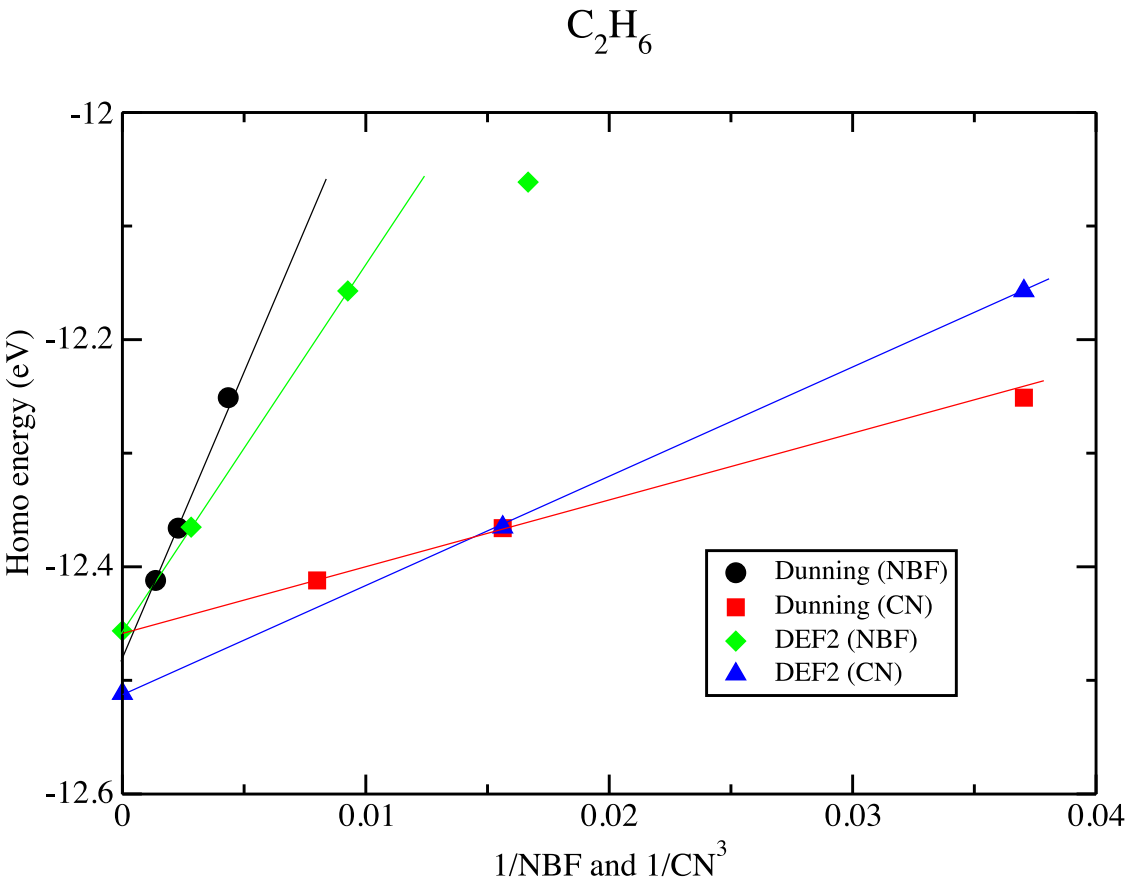

Figure 4: Basis set extrapolation of the QP homo energies comparing the def2 and Dunning basis sets (triple, quadruple, and quintuple). The points at 0 are the results of the extrapolation used in this work. The same as Figure 2, but for C_2H_6.

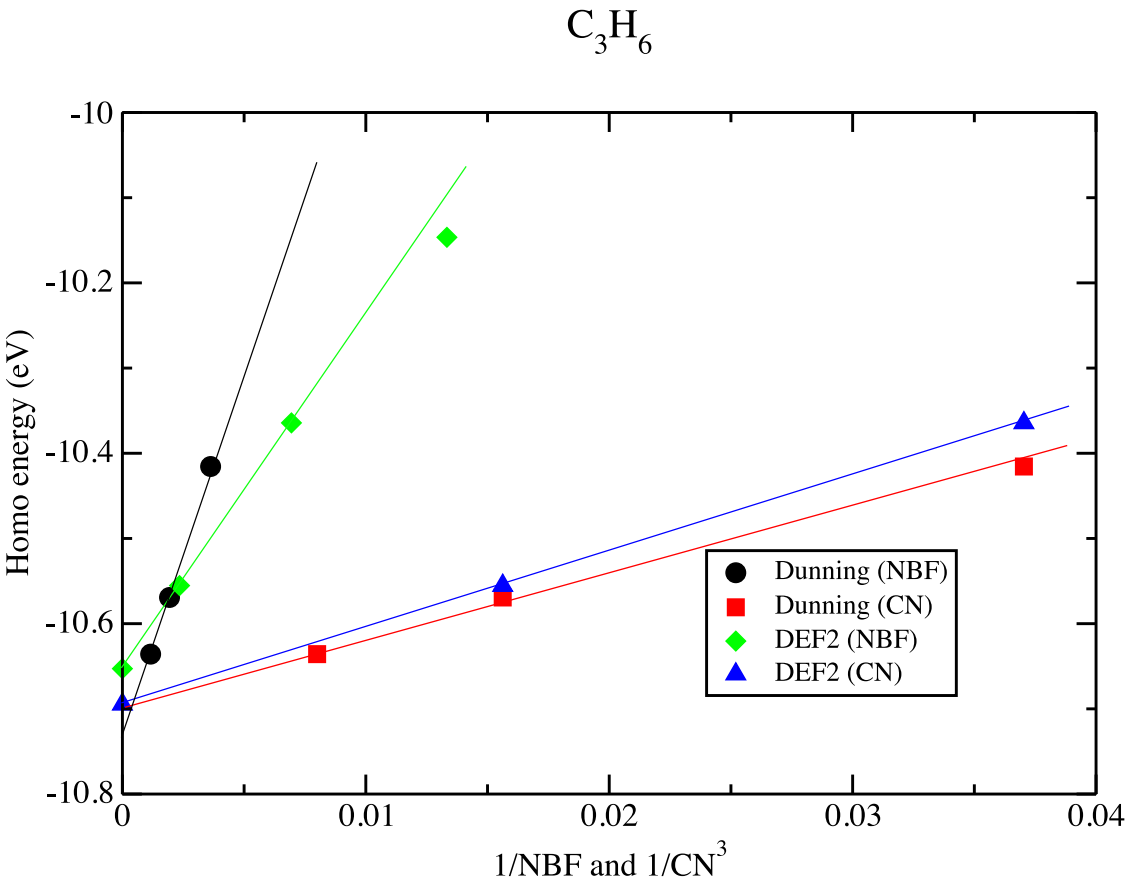

Figure 5: Basis set extrapolation of the QP homo energies comparing the def2 and Dunning basis sets (triple, quadruple, and quintuple). The points at 0 are the results of the extrapolation used in this work. The same as Figure 2, but for C_3H_6.

1.2 Ground state convergence parameters

The tables in this section report PBE KS-HOMO eigenvalues. The Tables 1 and 2 address ground state properties in TRUBOMOLE. It is shown that a level of meV convergence has been reached for the XC integration grid (grids m3, m4, m5) and that the error introduced by the RI on the DFT orbitals is also of this magnitude. Table 3 concerns FHI-AIMS, showing the dependence on different orders of the Hartree multipole expansion (l_hartree) in FHI-aims.

Table 1: PBE HOMO eigenvalue of four molecules for different XC integration grids (m3, m4, m5) in TURBOMOLE.

	m3	m4	m5
N_2	-10.20292396	-10.20192103	-10.20221994
F_2	-9.40915387	-9.40884945	-9.40869029
C_2H_6	-8.15643009	-8.15623074	-8.15630102
C_3H_6	-7.04459266	-7.04193216	-7.04189156

Table 2: PBE HOMO eigenvalue of four molecules obtained with and without the resolution of the identity (RI) in TURBOMOLE.

	RI	no RI
N_2	-10.20221994	-10.20052639
F_2	-9.40869029	-9.4084012
C_2H_6	-8.15630102	-8.15635867
C_3H_6	-7.04189156	-7.04225172

Table 3: PBE HOMO eigenvalue of four molecules for different orders of the Hartree multipole expansion (l_hartree) in FHI-aims.

	8	10	12
N_2	-10.20048075	-10.20051154	-10.20052298
F_2	-9.40826092	-9.40832362	-9.40830352
C_2H_6	-8.15644071	-8.15641251	-8.15640127
C_3H_6	-7.04265618	-7.0421901	-7.04233201

1.3 Convergence of the product-basis parameters in FHI-aims

The tables in this subsection report the G_0W_0 HOMO eigenvalue.

Table 4: Threshold of the Gram-Schmidt orthogonalization (prodbas_acc) in FHI-aims.

	1.d-5	1.d-6	1.d-7	1.d-8
N_2	-16.694478	-16.694478	-16.694478	-16.694477
F_2	-18.132832	-18.132834	-18.132837	-18.132838
C_2H_6	-13.247344	-13.247344	-13.247344	-13.247344
C_3H_6	-11.362216	-11.362215	-11.362215	-11.362215

Table 5: Threshold of the singular value decomposition (prodbas_threshold) in FHI-aims.

	1.d-5	1.d-6	1.d-7	1.d-8
N_2	-16.694478	-16.694478	-16.694477	-16.694477
F_2	-18.132819	-18.132819	-18.132820	-18.132820
C_2H_6	-13.247342	-13.247344	-13.247345	-13.247345
C_3H_6	-11.362216	-11.362217	-11.362216	-11.362191

Table 6: Comparison of the exchange correlation matrix elements (v_{XC}), exchange parts of the self-enery (Σ_x), and KS-energies (KS) of the HOMO level for the GW100 molecules.

Formula	CAS	v_{xc}		Σ_x		$\Sigma_x - v_{\mathrm{xc}}$		KS		
		BGW	QZVP	BGW	QZVP	BGW	QZVP	BGW	QZVP	EXTRA
He	12597-35-2	-17.94	-18.22	-27.23	-27.59	-9.29	-9.37	-15.77	-15.75	-15.77
Ne	7440-01-9	-25.20	-26.96	-35.22	-37.42	-10.02	-10.45	-13.31	-13.31	-13.48
Ar	7440-37-1	-15.50	-18.05	-21.35	-23.80	-5.85	-5.75	-10.27	-10.27	-10.36
Kr	7439-90-9	-13.41	-16.59	-18.20	-21.50	-4.80	-4.91	-9.26	-9.27	-9.31
Xe	7740-63-3	-11.57	-14.62	-15.56	-18.57	-3.99	-3.94	-8.13	-8.22	-
H_2	1333-74-0	-12.04	-12.06	-17.84	-17.87	-5.80	-5.81	-10.37	-10.38	-10.38
Li_2	14452-59-6	-4.68	-5.35	-6.44	-7.03	-1.76	-1.68	-3.22	-3.22	-3.21
Na_2	25681-79-2	-4.22	-5.23	-5.69	-6.56	-1.47	-1.33	-3.12	-3.13	-3.11
Na_4	39297-86-4	-4.25	-5.18	-5.31	-6.12	-1.06	-0.94	-2.69	-2.68	-2.67
Na_6	39297-88-6	-4.40	-5.44	-5.43	-6.32	-1.03	-0.89	-3.00	-2.99	-2.98
K_2	25681-80-5	-3.50	-4.62	-4.58	-5.53	-1.08	-0.91	-2.49	-2.57	-2.56
Rb_2	25681-81-6	-3.36	-4.63	-4.33	-5.41	-0.98	-0.78	-2.47	-2.49	-
N_2	7727-37-9	-17.14	-18.00	-23.55	-24.50	-6.41	-6.49	-10.27	-10.20	-10.20
P_2	12185-09-0	-10.38	-11.93	-13.17	-14.59	-2.79	-2.66	-7.11	-7.11	-7.15
As_2	23878-46-8	-9.62	-11.71	-12.00	-14.10	-2.38	-2.39	-6.50	-6.56	-6.67
F_2	7782-41-4	-23.68	-24.73	-32.27	-33.56	-8.59	-8.83	-9.39	-9.41	-9.48
Cl_2	7782-50-5	-14.17	-16.59	-18.83	-21.13	-4.66	-4.53	-7.29	-7.29	-7.36
Br_2	7726-95-6	-12.54	-15.53	-16.55	-19.53	-4.01	-4.00	-6.80	-6.83	-6.88
I_2	7553-56-2	-10.80	-13.78	-14.07	-16.97	-3.27	-3.20	-6.24	-6.25	-

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Table 6 continued

Formula	CAS	v_{xc}		Σ_x		$\Sigma_x - v_{xc}$		KS		
		BGW	QZVP	BGW	QZVP	BGW	QZVP	BGW	QZVP	EXTRA
CH_4	74-82-8	-13.81	-14.07	-19.00	-19.29	-5.19	-5.21	-9.44	-9.45	-9.46
C_2H_6	74-84-0	-14.04	-14.31	-18.89	-19.18	-4.85	-4.88	-8.13	-8.16	-8.16
C_3H_8	74-98-6	-14.03	-14.29	-18.77	-19.05	-4.74	-4.76	-7.72	-7.75	-7.76
C_4H_{10}	106-97-8	-13.88	-14.13	-18.53	-18.81	-4.66	-4.68	-7.54	-7.58	-7.58
C_2H_4	74-85-1	-12.70	-13.10	-15.99	-16.39	-3.29	-3.29	-6.76	-6.77	-6.78
C_2H_2	74-86-2	-13.20	-13.64	-16.92	-17.37	-3.72	-3.73	-7.16	-7.19	-7.21
C_4	12184-80-4	-13.80	-14.57	-18.41	-19.16	-4.60	-4.59	-7.29	-7.24	-7.26
C_3H_6	75-19-4	-14.31	-14.77	-18.30	-18.79	-3.99	-4.01	-7.05	-7.04	-7.05
C_6H_6	71-43-2	-13.06	-13.48	-15.61	-16.03	-2.56	-2.55	-6.27	-6.33	-6.35
C_8H_8	629-20-9	-13.20	-13.67	-15.71	-16.20	-2.51	-2.52	-5.26	-5.30	-5.32
C_5H_6	542-92-7	-13.10	-13.54	-15.79	-16.24	-2.69	-2.70	-5.37	-5.40	-5.42
C_2H_3F	75-02-5	-14.88	-15.40	-18.67	-19.22	-3.79	-3.82	-6.53	-6.53	-6.56
C_2H_3Cl	75-01-4	-13.46	-14.63	-16.90	-18.02	-3.43	-3.38	-6.45	-6.43	-6.46
C_2H_3Br	593-60-2	-12.80	-14.27	-15.83	-17.30	-3.03	-3.03	-5.83	-5.84	-5.86
C_2H_3I	593-66-8	-11.50	-13.58	-14.53	-16.55	-3.03	-2.98	-6.04	-6.04	-
CF_4	75-73-0	-23.61	-24.59	-31.76	-32.94	-8.15	-8.35	-10.42	-10.40	-10.46
CCl_4	56-23-5	-14.43	-16.87	-19.13	-21.43	-4.70	-4.56	-7.65	-7.66	-7.72
CBr_4	558-13-4	-12.79	-15.84	-16.79	-19.84	-4.00	-4.00	-6.95	-6.98	-7.00
CI_4	507-25-5	-11.04	-14.12	-14.28	-17.31	-3.25	-3.19	-6.20	-6.25	-
SiH_4	7803-62-5	-11.31	-12.07	-15.84	-16.54	-4.53	-4.47	-8.52	-8.52	-8.52
GeH_4	7782-65-2	-10.97	-12.32	-15.17	-16.59	-4.20	-4.27	-8.29	-8.37	-8.37
Si_2H_6	1590-87-0	-10.69	-12.07	-14.09	-15.35	-3.40	-3.28	-7.29	-7.29	-7.29
Si_5H_{12}	14868-53-2	-10.63	-12.27	-13.36	-14.85	-2.73	-2.58	-6.58	-6.57	-6.58
LiH	7580-67-8	-8.82	-9.20	-12.78	-13.15	-3.97	-3.96	-4.44	-4.36	-4.35
KH	7693-26-7	-8.02	-8.87	-11.29	-12.16	-3.26	-3.30	-3.44	-3.45	-3.45
BH_3	13283-31-3	-12.28	-12.50	-17.20	-17.43	-4.92	-4.93	-8.50	-8.49	-8.49
B_2H_6	19287-45-7	-12.60	-12.82	-17.29	-17.52	-4.69	-4.70	-7.86	-7.86	-7.87
NH_3	7664-41-7	-15.27	-15.87	-20.72	-21.38	-5.45	-5.51	-6.14	-6.12	-6.19
HN_3	7782-79-8	-16.25	-16.81	-19.94	-20.55	-3.68	-3.74	-6.84	-6.78	-6.82
PH_3	7803-51-2	-10.73	-12.25	-14.42	-15.86	-3.69	-3.61	-6.72	-6.71	-6.72
AsH_3	7784-42-1	-10.35	-12.39	-13.76	-15.81	-3.41	-3.41	-6.73	-6.72	-6.75
SH_2	7783-06-4	-11.83	-13.63	-15.96	-17.65	-4.13	-4.03	-6.29	-6.28	-6.32
FH	7664-39-3	-21.99	-22.88	-30.35	-31.43	-8.36	-8.55	-9.68	-9.57	-9.70
ClH	7647-01-0	-13.62	-15.78	-18.58	-20.63	-4.96	-4.85	-8.06	-8.02	-8.09
LiF	7789-24-4	-20.79	-21.55	-28.37	-29.22	-7.58	-7.67	-6.28	-6.09	-6.21
F_2Mg	7783-40-6	-21.28	-22.10	-29.05	-30.00	-7.77	-7.90	-8.52	-8.29	-8.28
TiF_4	7783-63-3	-22.44	-23.34	-29.98	-31.04	-7.53	-7.70	-10.43	-10.43	-10.42
AlF_3	7784-18-1	-22.49	-23.38	-30.40	-31.46	-7.92	-8.08	-9.81	-9.69	-9.76
BF	13768-60-0	-10.45	-11.37	-14.42	-15.34	-3.97	-3.97	-6.80	-6.75	-6.75
SF_4	7783-60-0	-19.78	-21.43	-25.17	-26.89	-5.39	-5.46	-8.29	-8.19	-8.31
BrK	7758-2-3	-11.63	-14.13	-15.64	-18.14	-4.01	-4.01	-4.48	-4.71	-4.80
GaCl	17108-85-9	-10.45	-13.13	-13.54	-16.08	-3.09	-2.95	-6.62	-6.51	-6.58
NaCl	7647-14-5	-13.10	-15.11	-17.80	-19.70	-4.70	-4.58	-5.47	-5.25	-5.34
$MgCl_2$	7786-30-3	-13.42	-15.47	-18.15	-20.09	-4.74	-4.62	-7.73	-7.63	-7.65
AlI_3	7784-23-8	-10.85	-13.74	-14.12	-16.96	-3.27	-3.22	-6.64	-6.63	-
BN	10043-11-5	-14.09	-14.60	-18.22	-18.74	-4.13	-4.14	-7.50	-7.46	-7.47
NCH	74-90-8	-14.91	-15.42	-19.16	-19.69	-4.26	-4.27	-9.07	-9.03	-9.05
PN	17739-47-8	-15.21	-16.50	-20.42	-21.67	-5.21	-5.16	-7.76	-7.73	-7.79
H_2NNH_2	302-01-2	-16.04	-16.72	-21.37	-22.09	-5.33	-5.37	-5.26	-5.26	-5.32
H_2CO	50-00-0	-17.80	-18.31	-23.55	-24.14	-5.75	-5.83	-6.26	-6.25	-6.29
CH_4O	67-56-1	-18.12	-18.69	-24.21	-24.90	-6.10	-6.21	-6.32	-6.30	-6.36

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Table 6 continued

Formula	CAS	v_{xc}		Σ_x		$\Sigma_x - v_{xc}$		KS		
		BGW	QZVP	BGW	QZVP	BGW	QZVP	BGW	QZVP	EXTRA
C_2H_6O	64-17-5	-17.98	-18.54	-24.01	-24.68	-6.03	-6.14	-6.13	-6.12	-6.17
C_2H_4O	75-07-0	-17.92	-18.53	-23.45	-24.14	-5.52	-5.61	-5.99	-5.96	-5.99
$C_4H_{10}O$	60-29-7	-17.66	-18.15	-23.29	-23.86	-5.63	-5.71	-5.75	-5.79	-5.82
CH_2O_2	64-18-6	-18.78	-19.40	-24.63	-25.37	-5.86	-5.97	-6.96	-6.91	-6.96
HOOH	7722-84-1	-19.79	-20.49	-26.60	-27.43	-6.81	-6.94	-6.44	-6.39	-6.47
H_2O	7732-18-5	-18.52	-19.15	-25.26	-26.02	-6.73	-6.87	-7.23	-7.16	-7.25
CO_2	124-38-9	-19.43	-20.08	-24.79	-25.55	-5.36	-5.47	-9.12	-9.07	-9.12
CS_2	75-15-0	-12.16	-14.10	-15.16	-16.97	-3.01	-2.87	-6.77	-6.80	-6.82
OCS	463-58-1	-13.84	-15.40	-17.49	-18.98	-3.65	-3.58	-7.46	-7.48	-7.51
OCSe	1603-84-5	-12.55	-17.80	-15.76	-14.57	-3.21	3.23	-6.91	-6.98	-7.00
CO	630-08-0	-15.01	-15.91	-20.32	-21.25	-5.31	-5.34	-9.34	-9.31	-9.34
O_3	10028-15-6	-19.86	-20.72	-26.21	-27.20	-6.35	-6.48	-8.00	-7.89	-7.94
SO_2	7446-09-5	-17.51	-18.62	-22.62	-23.77	-5.11	-5.15	-8.13	-8.03	-8.16
BeO	1304-56-9	-16.29	-16.87	-21.46	-22.10	-5.18	-5.22	-6.23	-6.13	-6.16
MgO	1309-48-4	-16.93	-17.57	-22.62	-23.29	-5.69	-5.71	-4.85	-4.79	-4.79
C_7H_8	108-88-3	-13.31	-13.74	-15.84	-16.27	-2.53	-2.54	-5.95	-6.00	-6.01
C_8H_{10}	100-41-4	-13.37	-13.83	-15.86	-16.32	-2.49	-2.50	-5.98	-6.00	-6.02
C_6F_6	392-56-3	-16.96	-17.60	-20.32	-21.01	-3.36	-3.41	-6.74	-6.63	-6.69
C_6H_5OH	108-95-2	-15.07	-15.59	-18.14	-18.69	-3.07	-3.10	-5.57	-5.62	-5.65
$C_6H_5NH_2$	62-53-3	-14.49	-15.00	-17.45	-18.00	-2.97	-3.00	-4.93	-5.00	-5.04
C_5H_5N	110-86-1	-16.44	-17.19	-21.55	-22.32	-5.11	-5.12	-5.84	-5.93	-5.97
$C_5H_5N_5O$	73-40-5	-15.40	-15.94	-17.83	-18.40	-2.43	-2.46	-5.23	-5.27	-5.32
$C_5H_5N_5O$	73-24-5	-15.17	-15.67	-17.77	-18.30	-2.61	-2.63	-5.46	-5.50	-5.55
$C_4H_5N_3O$	71-30-7	-16.46	-16.98	-19.80	-20.35	-3.34	-3.38	-5.67	-5.70	-5.75
$C_5H_6N_2O_2$	65-71-4	-15.68	-16.20	-18.68	-19.24	-2.99	-3.04	-6.07	-6.04	-6.07
$C_4H_4N_2O_2$	66-22-8	-19.14	-19.84	-24.97	-25.75	-5.83	-5.91	-6.26	-6.26	-6.31
CH_4N_2O	57-13-6	-18.75	-19.39	-24.57	-25.31	-5.83	-5.91	-5.88	-5.89	-5.95
Ag_2	12187-06-3	-9.44	-9.43	-10.97	-10.79	-1.53	-1.35	-5.24	-5.26	-
Cu_2	12190-70-4	-32.91	-35.67	-41.84	-45.06	-8.94	-9.39	-4.66	-4.77	-4.72
NCCu	544-92-3	-24.56	-26.30	-31.19	-33.23	-6.63	-6.93	-6.79	-6.71	-6.69

2 Parameters for the plane wave calculations

In this section the most important pseudo potential parameters and plane wave function cutoffs used in the BerkeleyGW calculations are summarized.

Table 8: Wave function cutoffs for BerkeleyGW calculations.

Molecule	Wavefunction cutoff (Ry)
Ethybenzene	80
Ozone	110
Boron nitride	110
Buthane	80

Continues on next page

Table 8 continued

Molecule	Wavefunction cutoff (Ry)
Tuloene	80
Phenol	100
Pyridine	100
Tetracarbon	80
Cesium dimer	50
Phosphorus dimer	50
Carbon dioxide	110
Helium atom	80
Beryllium monoxide	110
Magnesium monoxide	110
Borane	80
Hydrogen	90
Boron monofluoride	90
Lithium dimer	50
Pentasilane	80
Disilane	80
Carbon oxyselenide	110
Gallium monochloride	60
Phosphorus mononitride	110
Diborane	80
Arsenic dimer	50
Sodium dimer	50
Potassium dimer	50
Rubidium dimer	50
Hydrazene	110
Hexafluorobenzene	90
Sodium tetramer	50
Sodium Hexamer	50
Carbon oxysulfide	110
Formaldehyde	110
Carbon tetraiodide	60
Cyclopentadiene	80
Carbon tetrabromide	60
Carbon tetrachloride	80
Urea	110
Vinyl bromide	80
Vinyl iodide	80
Ethoxy ehtane	100
Aniline	100
cyclooctatetraene	80
Carbon monoxide	110
Ethanol	100
Formic acid	110
Thymine	110
Uracil	110
Methanol	100
Cytosine	110
Benzene	80
Adenine	110
Guanine	110
Methane	90
Ethane	80
Ethylene	80

Continues on next page

Table 8 continued

Molecule	Wavefunction cutoff (Ry)
Acetylen	80
Hydrogen cyanide	110
Propane	80
Krypton atom	60
Neon atom	80
Argon atom	90
Sulfer dioxide	110
Vinyl chloride	70
Vinyl fluoride	90
Acetaldehyde	100
Carbon disulfide	70
Cyclopropane	80
Carbon tetrafluoride	90
Iodine	50
Lithium hydride	60
Hydrogen chloride	70
Sodium chloride	60
Hydrogen fluoride	90
Amonia	110
Potassium hydride	60
Hydrogen peroxide	110
Bromine	60
Nitrogen	120
Water	110
Potassium bromide	70
Fluorine	90
chlorine	60
Germane	80
Hydrogen azide	120
Hydrogen sulfide	80
Magnesium fluoride	90
Sulfer tetrafluoride	90
Titanium fluoride	80
Aluminum trifluoride	90
Aluminum triiodide	50
Arsine	80
Magnesium chloride	60
Lithium fluoride	90
Phosphine	80
Silane	80
Copper dimer	350
Copper cyanide	350
Silver dimer	200

Table 7: Pseudopotential parameters from the Quantum Espresso library[2] for BerkeleyGW calculations. N_c is the number of valence electrons, $R_{\text{cut-s}}$ the s-orbital, $R_{\text{cut-p}}$ the p-orbital, and $R_{\text{cut-d}}$ the d-orbital cutoff, respectively.

Atom	N_c	$R_{\text{cut-s}}(a.u.)$	$R_{\text{cut-p}}$ (a.u.)	$R_{\text{cut-d}}$ (a.u.)
H	1	0.50	N/A	N/A
He	2	1.00	N/A	N/A
Li	1	2.45	2.45	N/A
Be	2	2.10	2.10	N/A
B	3	1.40	1.40	N/A
C	4	1.30	1.30	N/A
N	5	1.00	1.00	N/A
O	6	1.10	1.10	N/A
F	7	1.30	1.30	N/A
Ne	8	1.55	1.55	N/A
Na	1	2.30	2.30	N/A
Mg	2	2.80	2.80	2.80
Al	3	2.60	2.60	2.60
Si	4	1.80	1.80	1.80
P	5	1.95	1.95	1.95
S	6	1.70	1.70	1.70
Cl	7	1.75	1.75	1.75
Ar	8	2.05	2.05	2.05
K	1	3.51	3.76	3.76
Ca	2	3.30	3.30	3.30
Ga	3	2.05	2.45	2.75
Ge	4	2.10	2.10	2.40
As	5	2.10	2.10	2.10
Se	6	1.90	1.90	1.90
Br	7	1.95	1.95	1.95
Kr	8	3.25	3.25	3.25
Rb	1	3.82	3.82	3.72
I	7	2.40	2.40	2.40
Xe	8	3.25	3.25	3.25
Cs	1	4.40	4.60	N/A

3 Graphical solutions QPE

This section contains the plots of the self-energies used to graphically solve the QP-equation for the two remaining molecules that exhibit multi-solution behaviour, but were not presented in the main manuscript.

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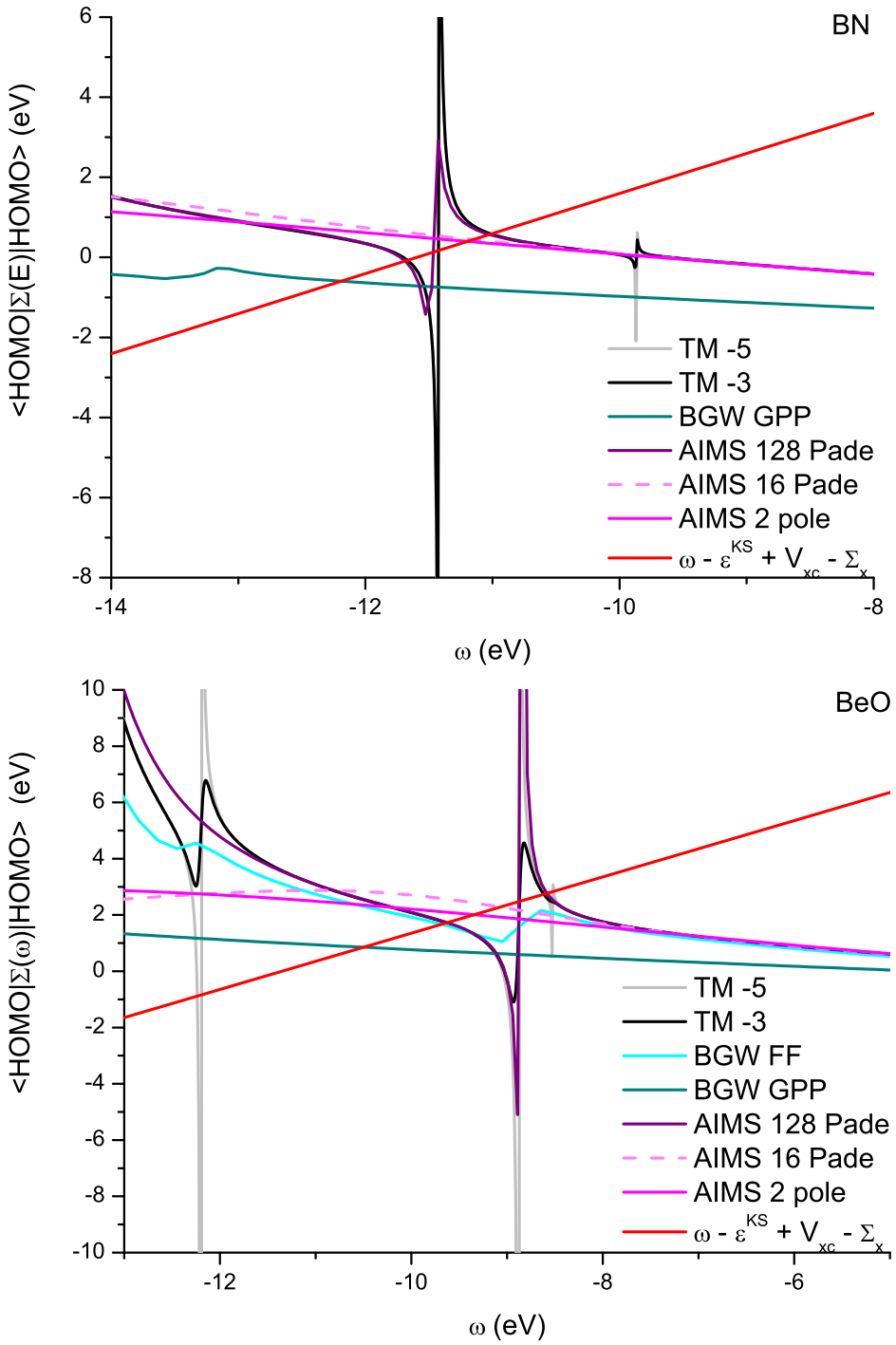

Figure 6: Comparison of the energy dependent correlation part of the self-energy $\Sigma_c(\varepsilon)$ calculated with the three different codes using different procedures for boron nitride and beryllium oxide. "TM -5" and "TM -3" indicate TURBOMOLE results calculated with imaginary shifts of $1e-3$ and $1e-5$ H respectively, "BGW-GPP" and "BGW-FF" indicate BerkeleyGW results using the Generalize Plasmon Pole model and Full Frequency integration method respectively, "AIMS-2P", "AIMS 128 Pade", and " AIMS 16 pade" indicate FHI-aims results using 2 Pole, 128 parameter Padé, and 16 parameter Padé analytical continuation.