Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting Information: The one-electron self interaction error in 74 density functional approximations: a case study on hydrogenic mono- and dinuclear systems

Dale R. Lonsdale and Lars Goerigk*

School of Chemistry, The University of Melbourne, Victoria 3010, Australia;

Ph: +61-(0)3-83446784

E-mail: lars.goerigk@unimelb.edu.au

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SI.1 Technical Details

For the mononuclear series calculations, grids were chosen to replicate the 'grid 7' option from ORCA consisting of a 770 Lebedev scheme which is an angular grid of 770 points and a series of radial grid points that change depending on the nuclei: H,He 45 radial points, Li-Ne 50 radial points, Na-Ar 55 radial points, K-Kr 60 radial points. Such points were initially chosen to compare QCHEM results with ORCA results to ensure trends were correct. For the diatomic calculations the grid was defined as XC_Grid 000060000770. For the SIE4x4 test set, QCHEM 5.2 was used with the SG-3 grid and the aug-cc-pVQZ (regular, not decontracted) was used. Several DFAs are not defined for use in QCHEM 5.2, namely RSX-QIDH, ωB2PLYP, and ωB2GPPLYP, which we used ORCA 4.2.1 with the grid 7 option instead.

SI.2 Mononuclear series

SI.2.1 Mononuclear series plots

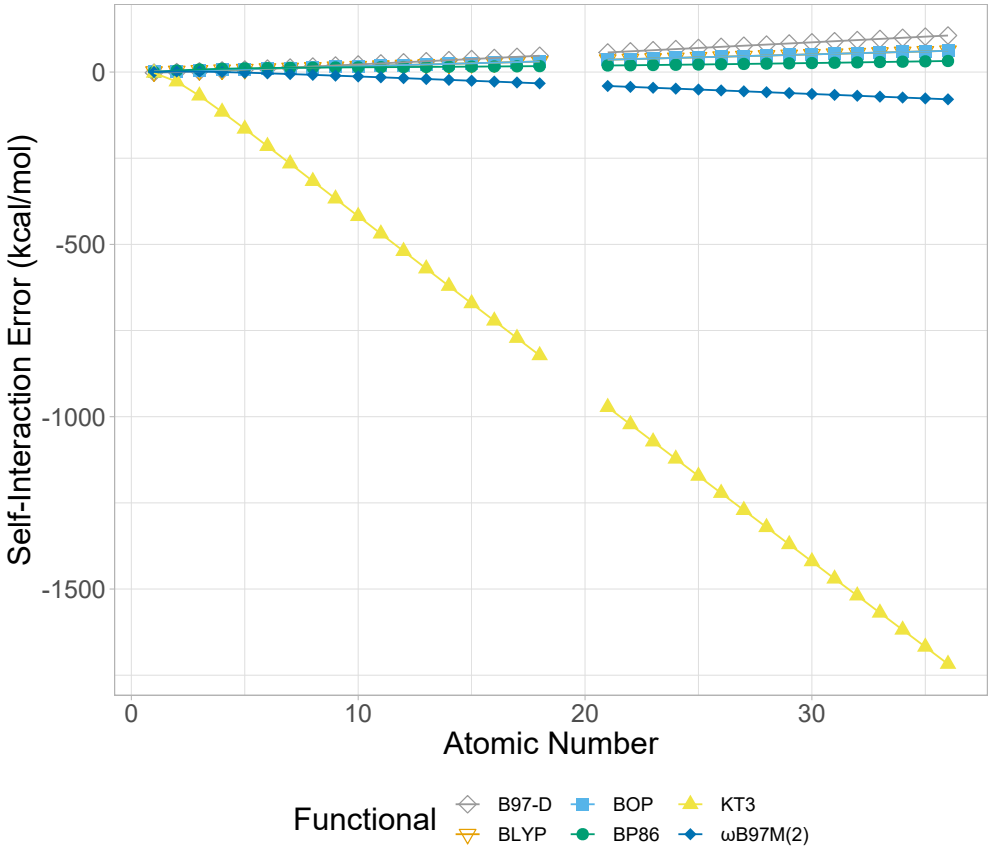

Figure S1: Self-interaction error vs increasing atomic charge of one-electron ions for B97-D, BLYP, BOP, BP86, KT3 with ωB97M(2) as a reference.

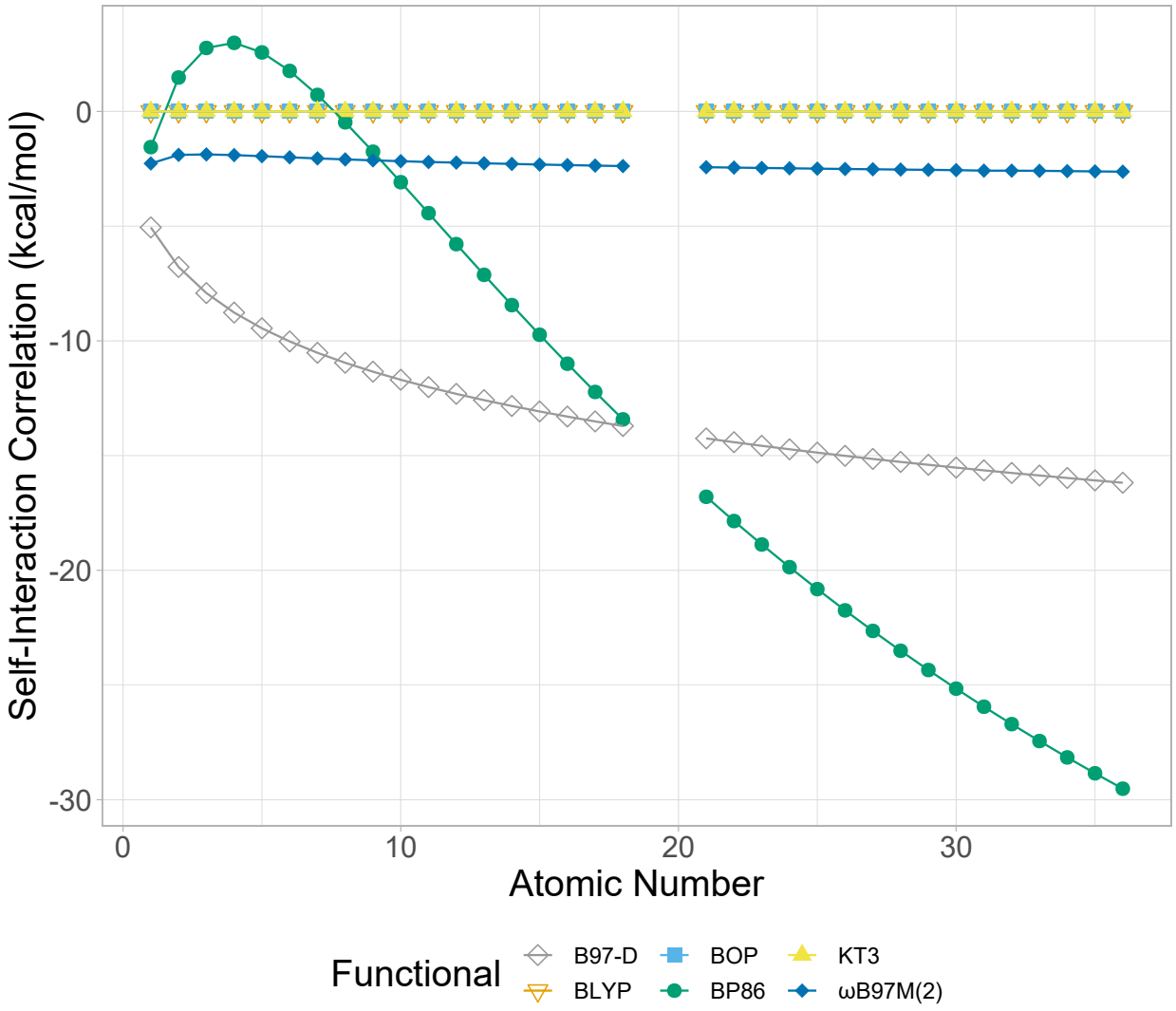

Figure S2: Self-interaction correlation vs increasing atomic charge of one-electron ions for B97-D, BLYP, BOP, BP86, KT3 with ωB97M(2) as a reference.

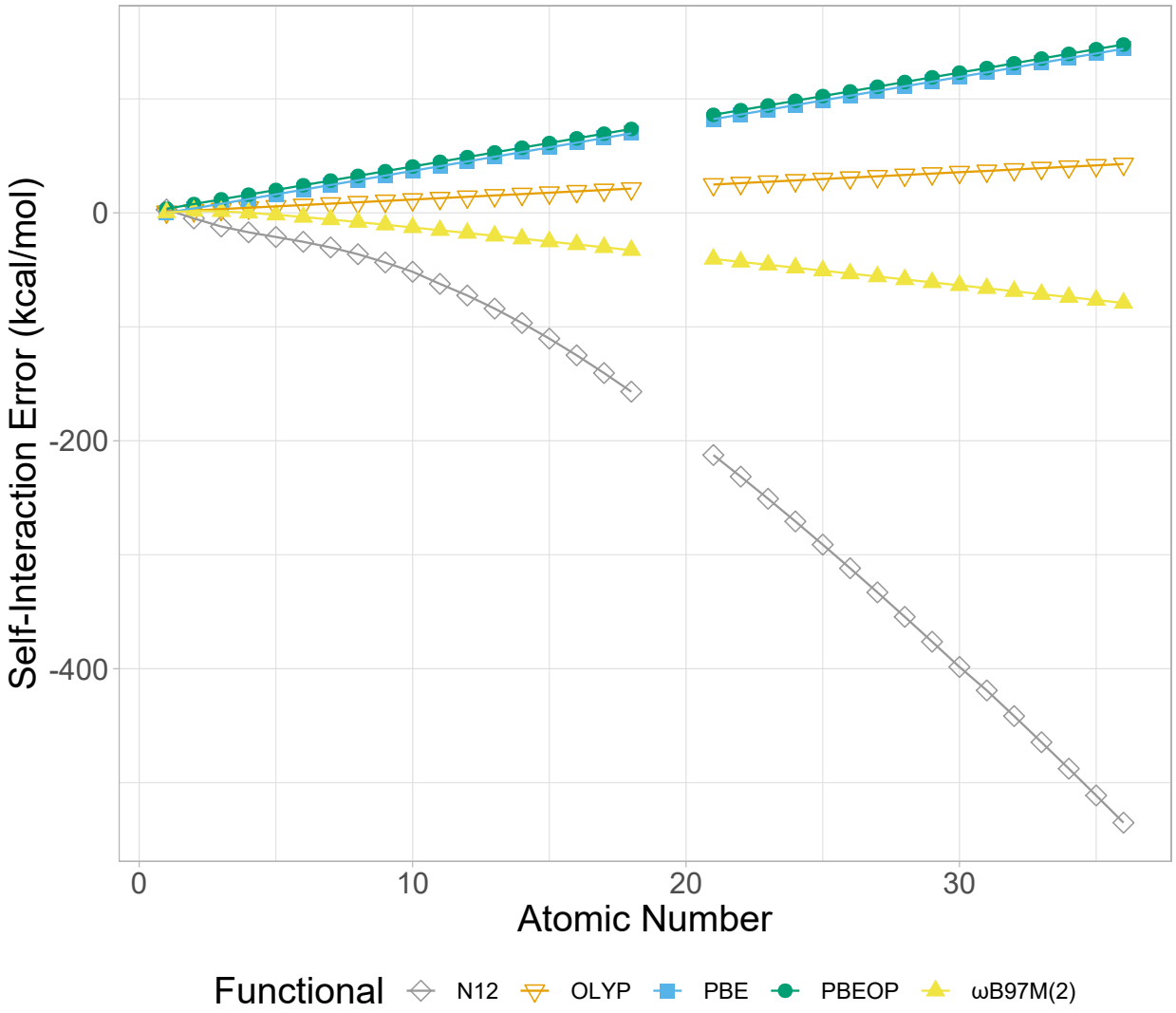

Figure S3: Self-interaction error vs increasing atomic charge of one-electron ions for N12, OLYP, PBE, PBEOP with ωB97M(2) as a reference.

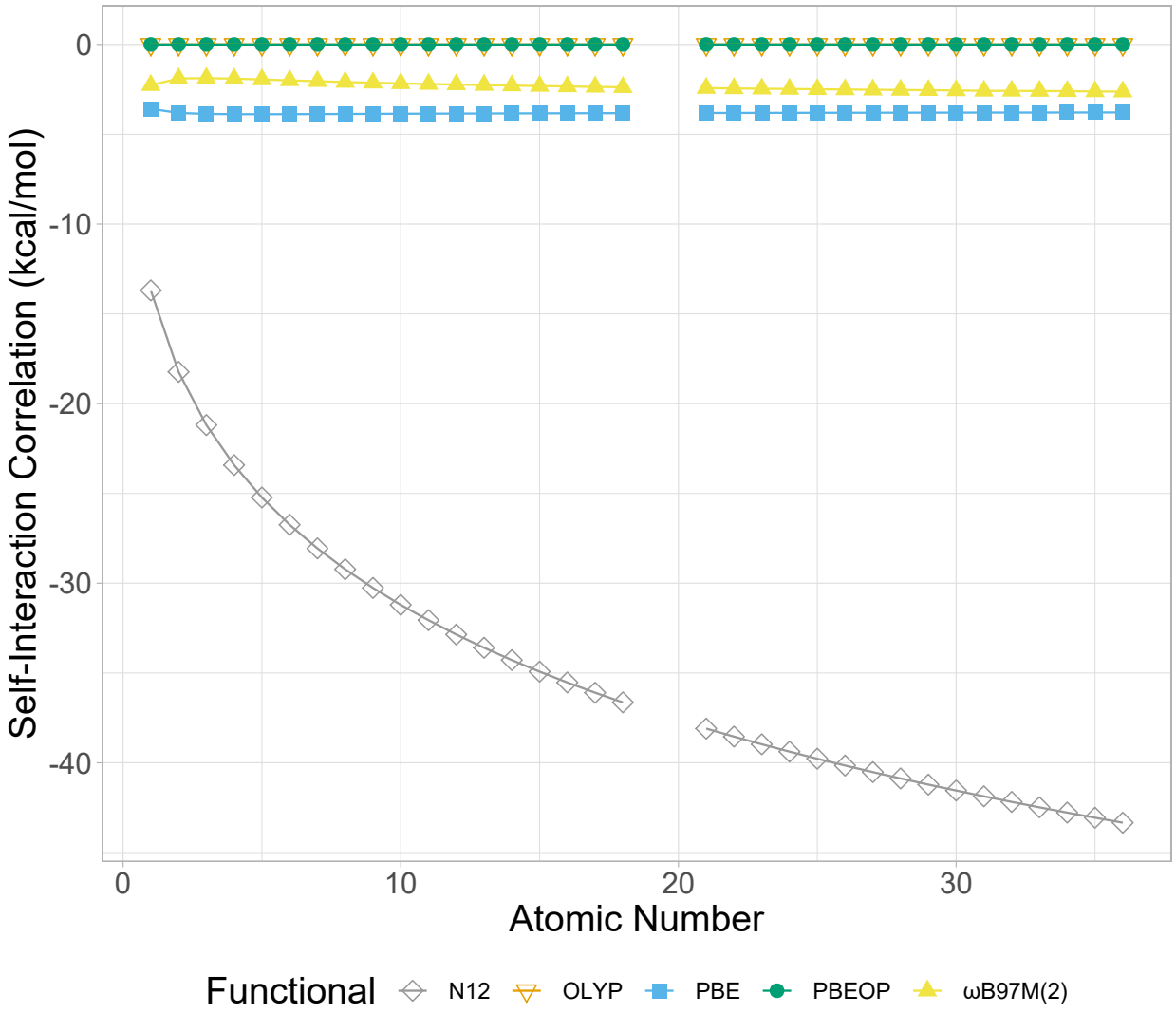

Figure S4: Self-interaction correlation vs increasing atomic charge of one-electron ions for N12, OLYP, PBE, PBEOP with ωB97M(2) as a reference.

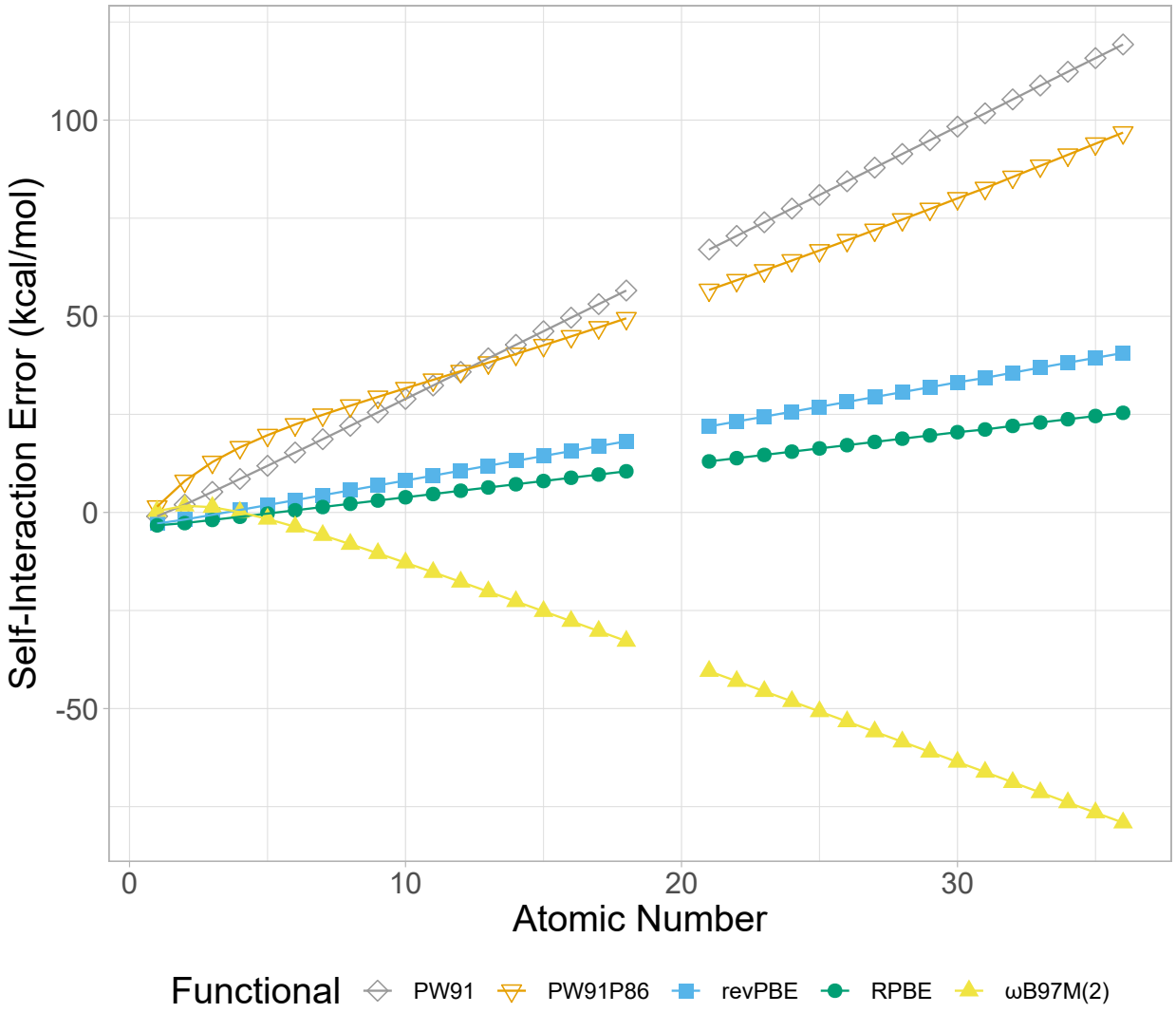

Figure S5: Self-interaction error vs increasing atomic charge of one-electron ions for PW91, PW91P86, revPBE, RPBE with ωB97M(2) as a reference.

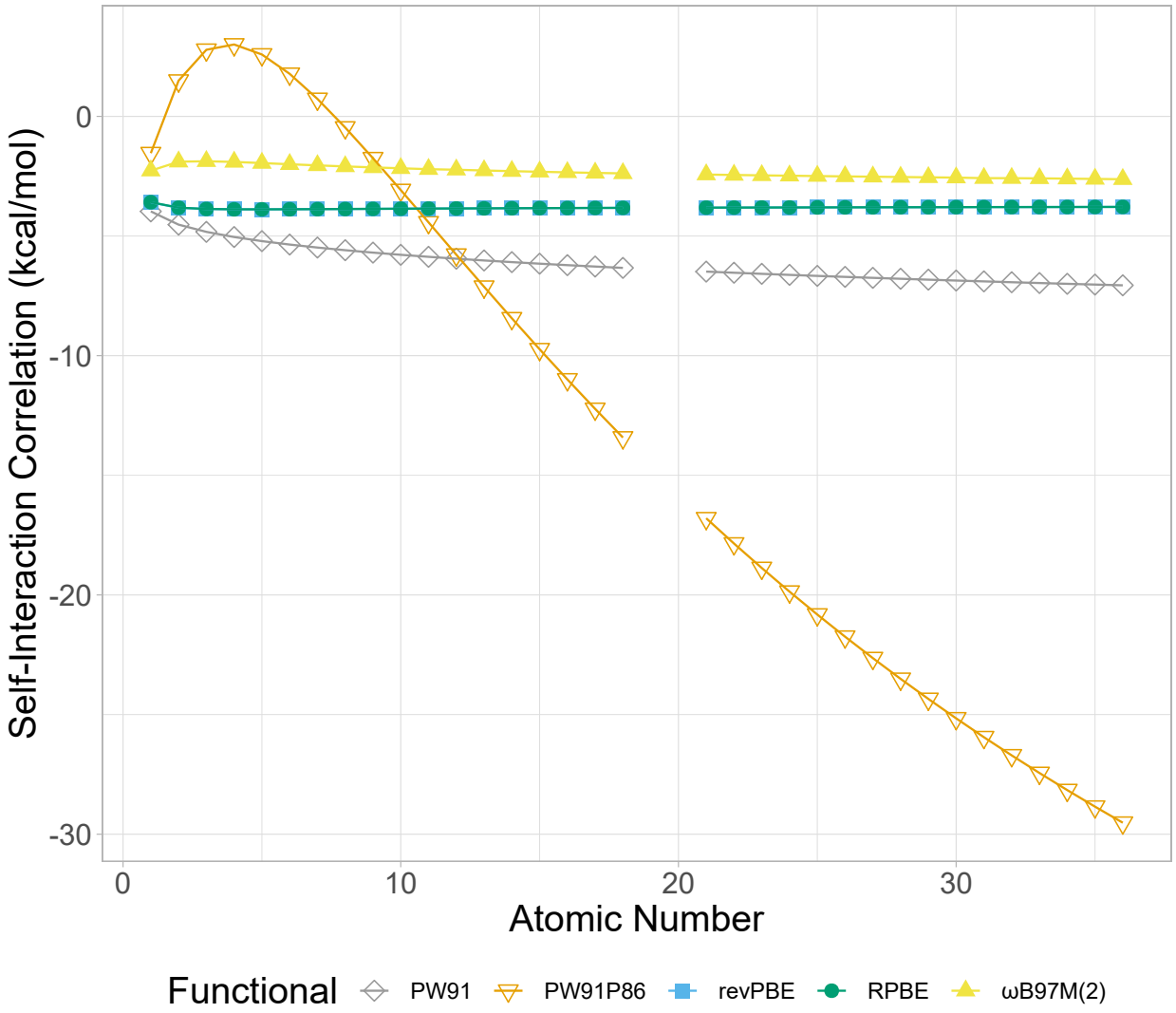

Figure S6: Self-interaction correlation vs increasing atomic charge of one-electron ions for PW91, PW91P86, revPBE, RPBE with ωB97M(2) as a reference.

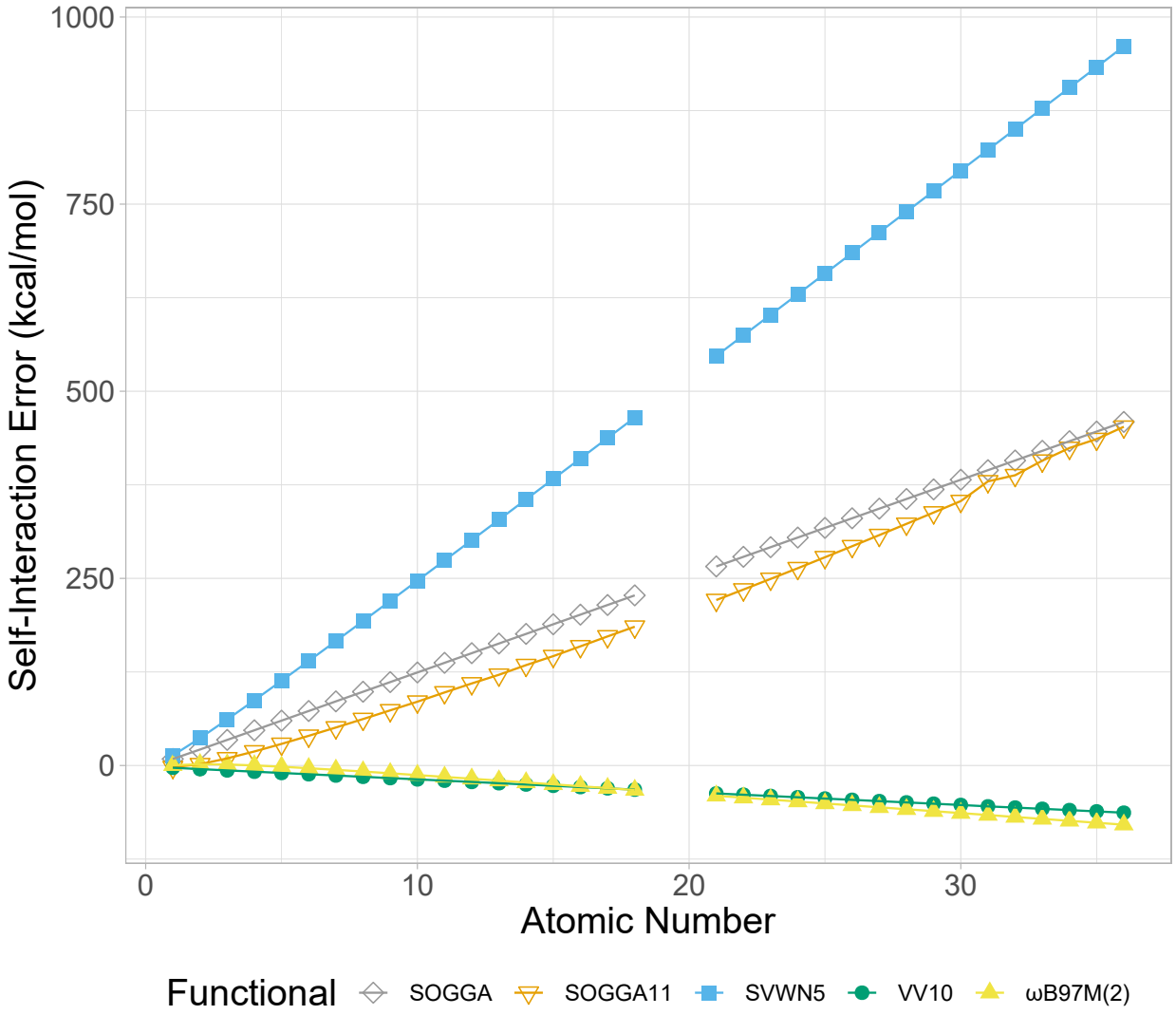

Figure S7: Self-interaction error vs increasing atomic charge of one-electron ions for SOGGA, SOGGA11, SVWN5, VV10 with ωB97M(2) as a reference.

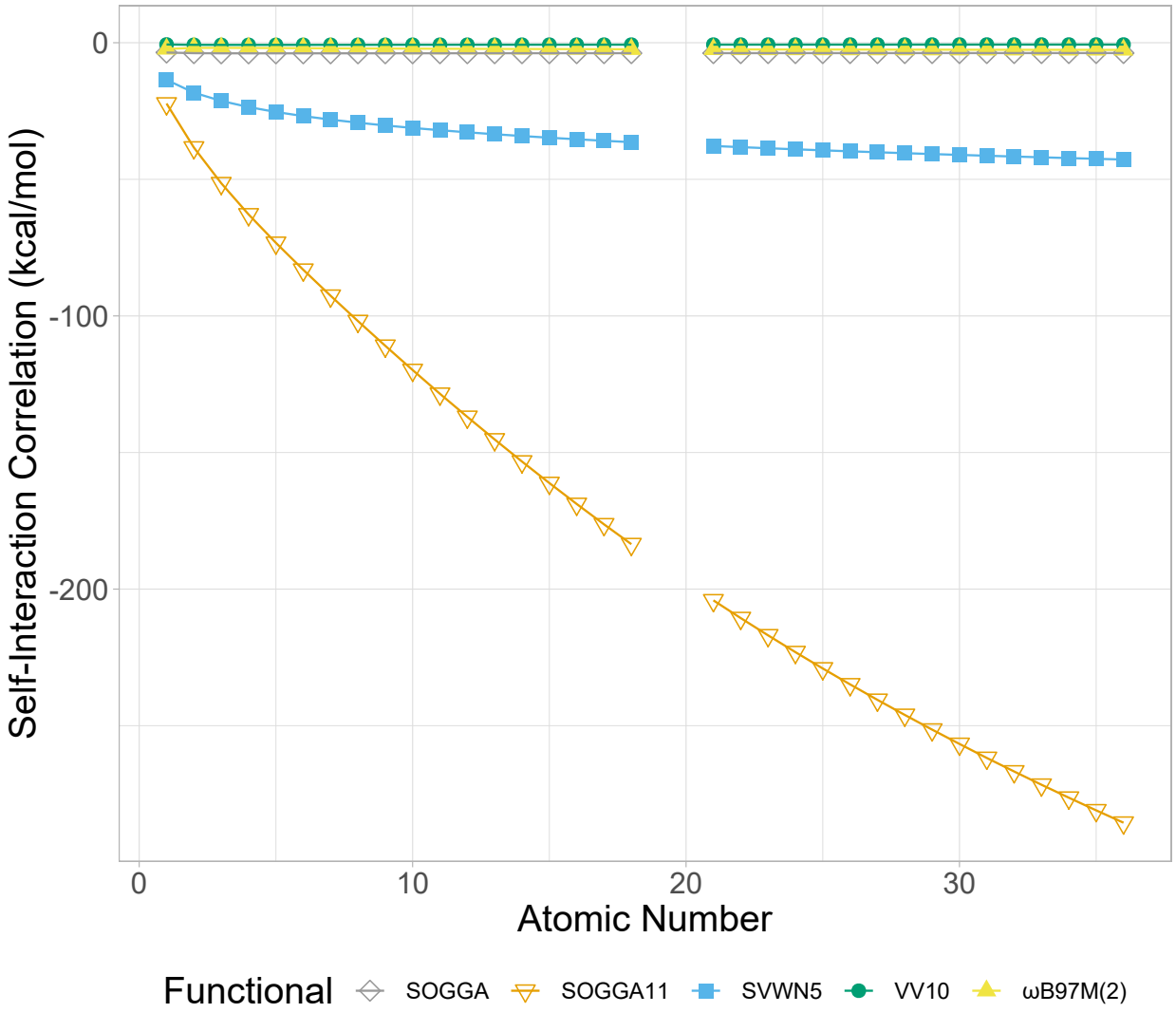

Figure S8: Self-interaction correlation vs increasing atomic charge of one-electron ions for SOGGA, SOGGA11, SVWN5, VV10 with ωB97M(2) as a reference.

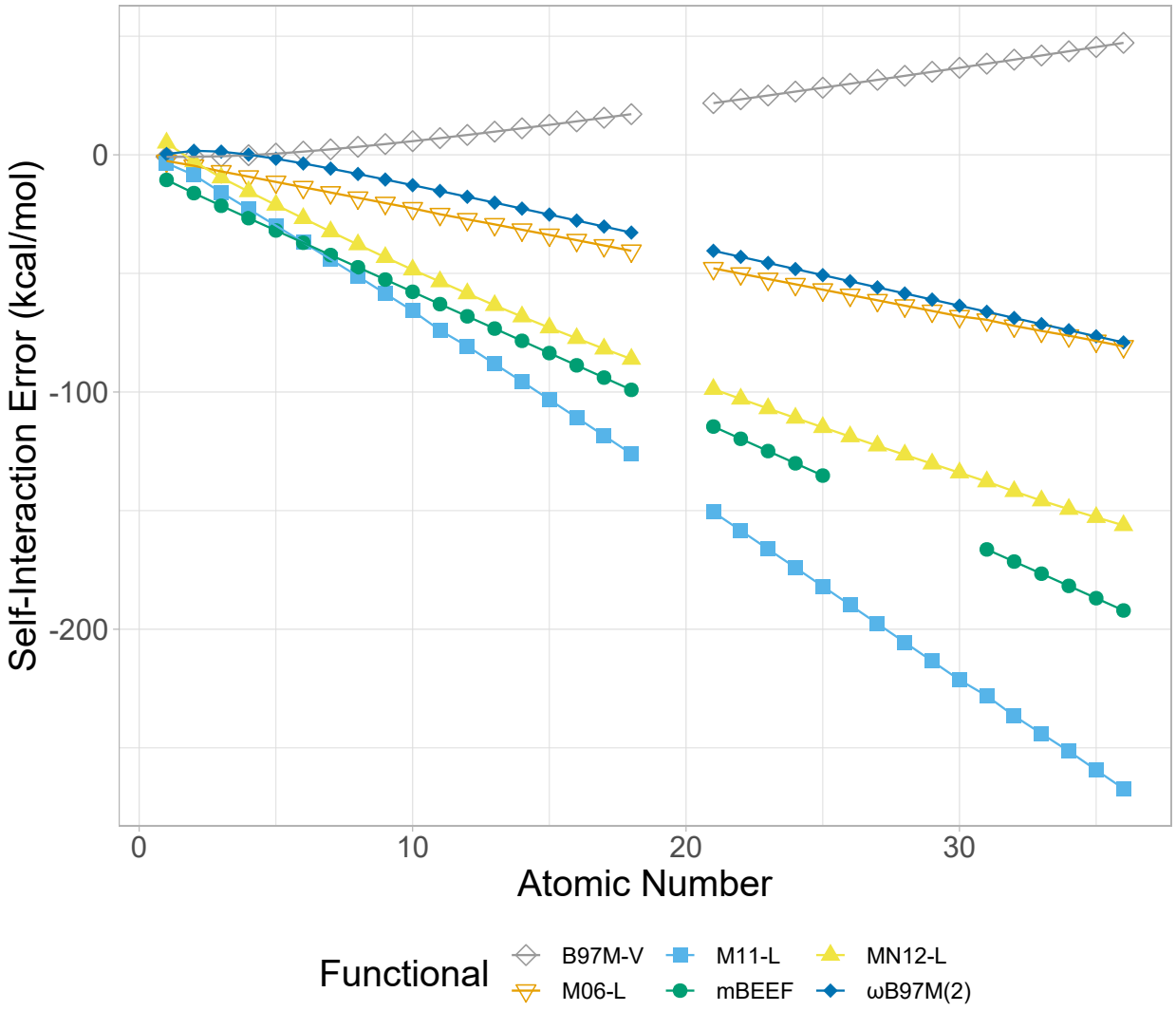

Figure S9: Self-interaction error vs increasing atomic charge of one-electron ions for B97M-V, M06-L, M11-L, mBEEF, MN12-L with ωB97M(2) as a reference.

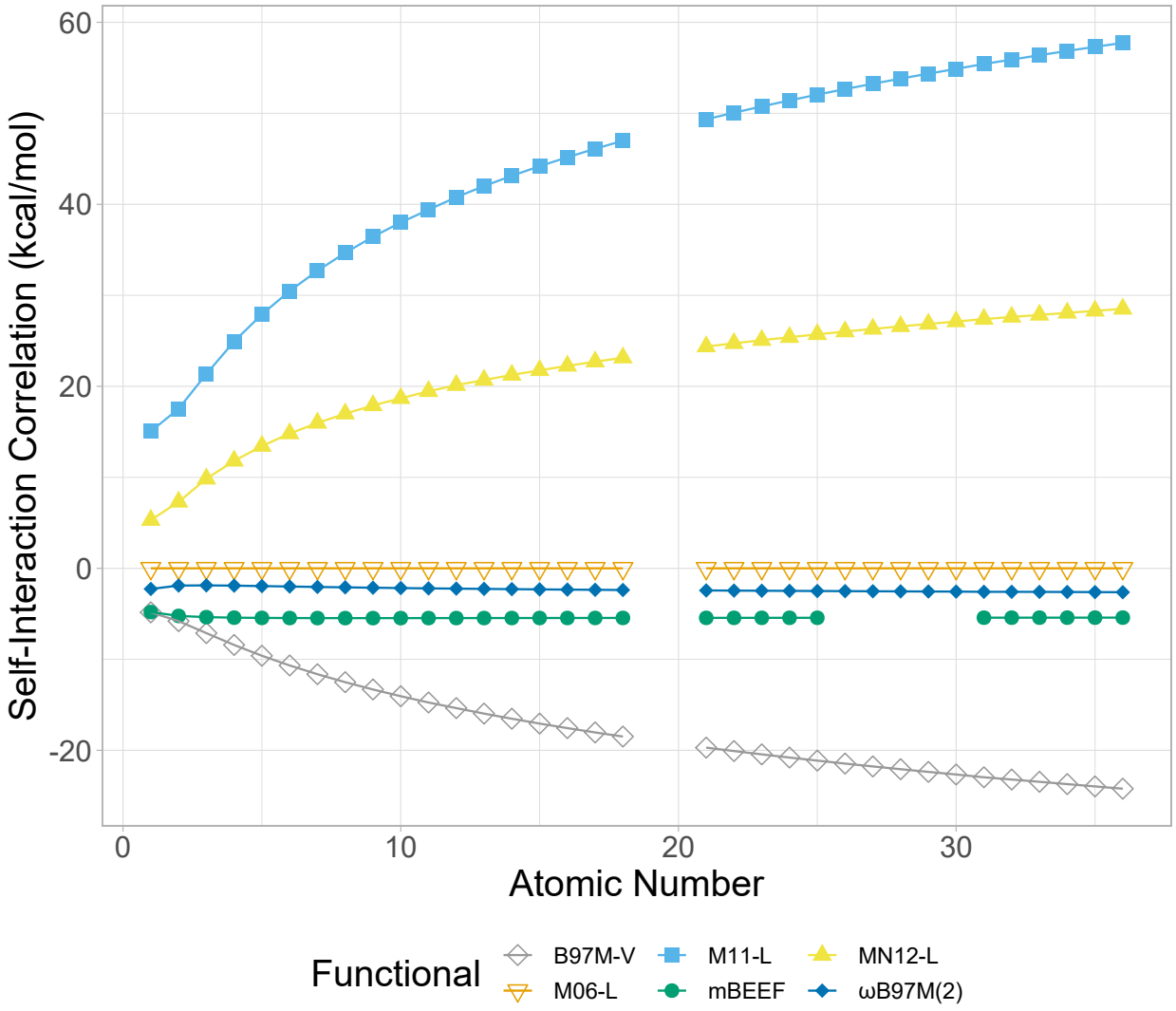

Figure S10: Self-interaction correlation vs increasing atomic charge of one-electron ions for B97M-V, M06-L, M11-L, mBEEF, MN12-L with ωB97M(2) as a reference.

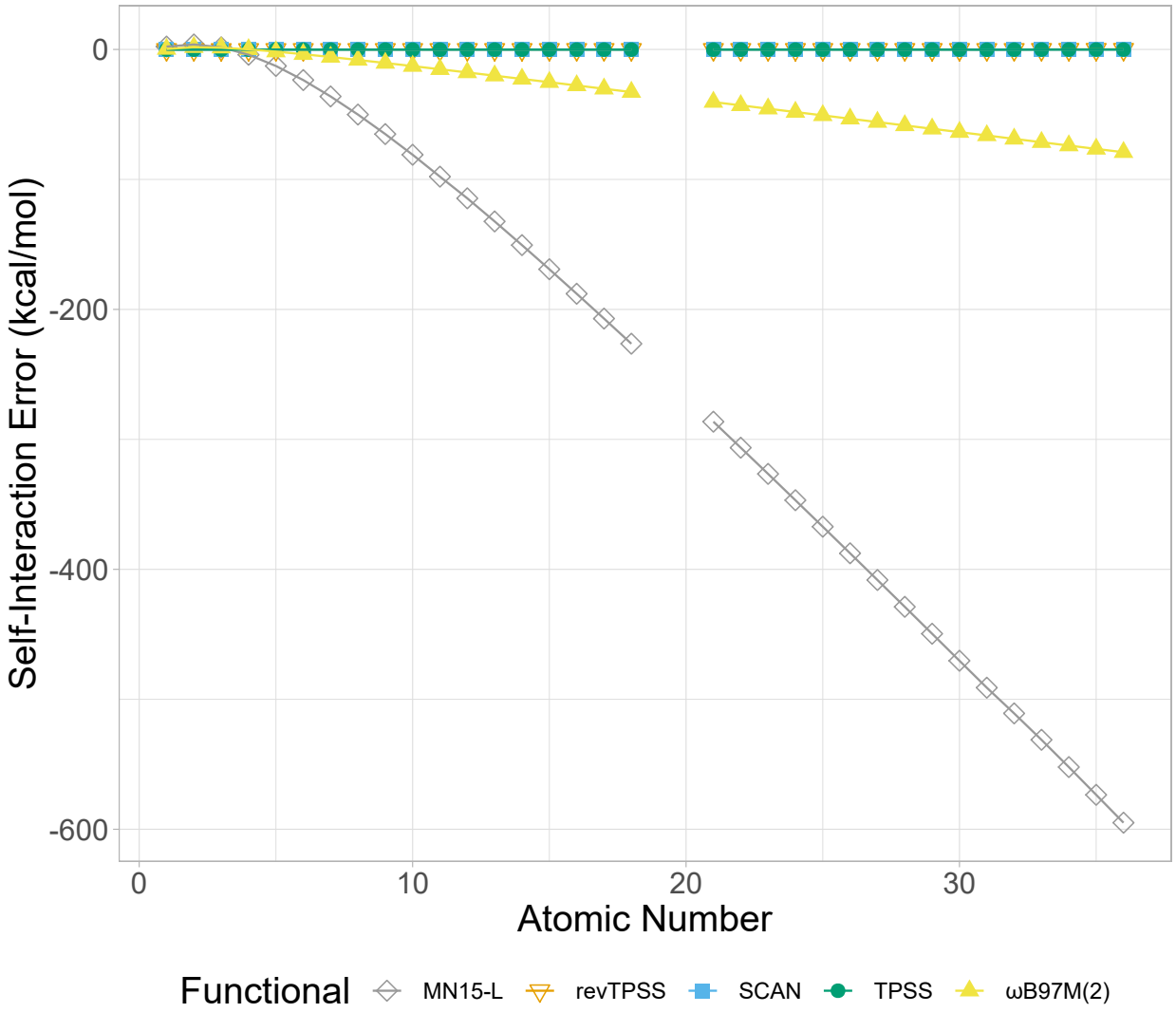

Figure S11: Self-interaction error vs increasing atomic charge of one-electron ions for MN15-L, revTPSS, SCAN, TPSS with ωB97M(2) as a reference.

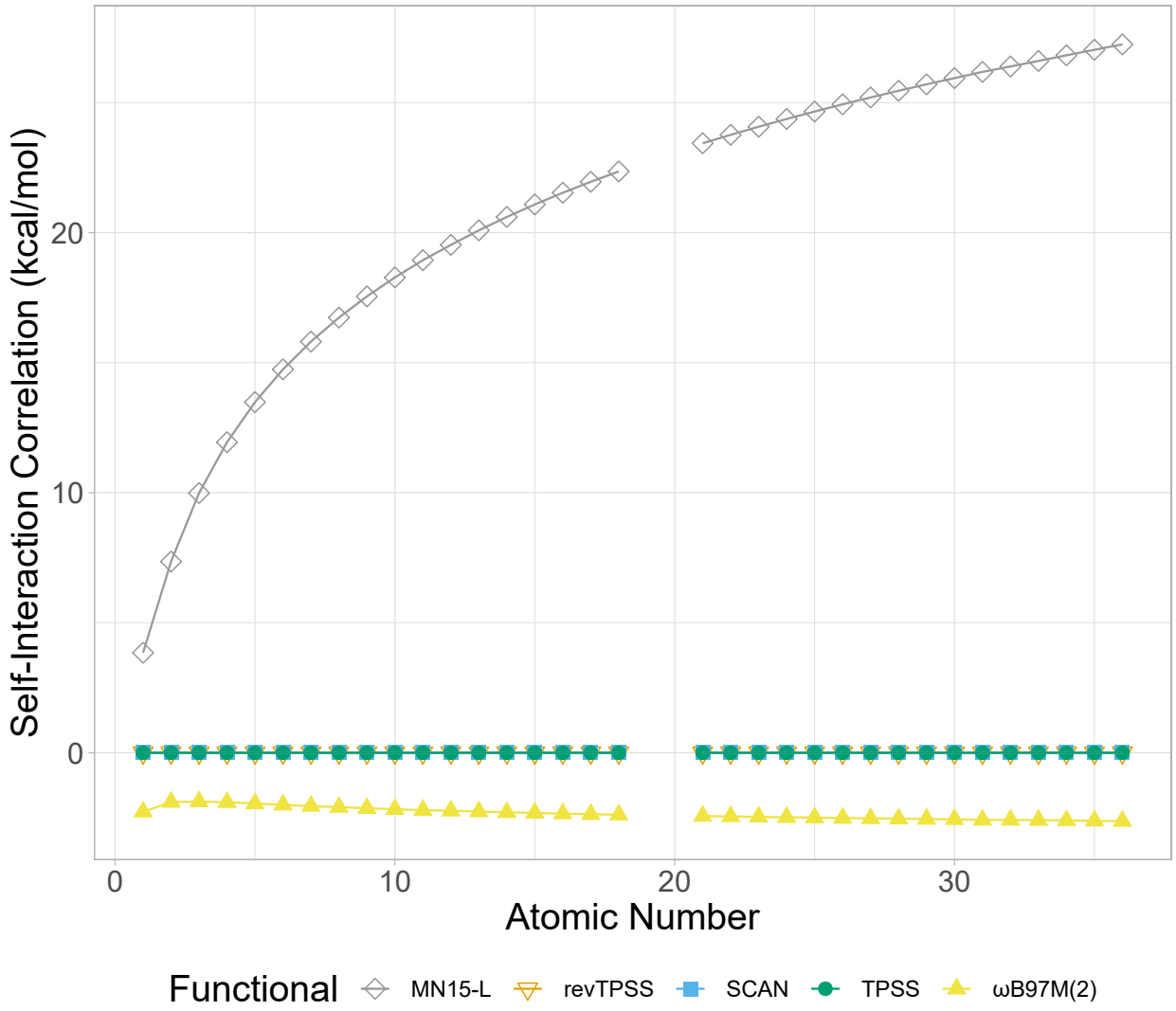

Figure S12: Self-interaction correlation vs increasing atomic charge of one-electron ions for MN15-L, revTPSS, SCAN, TPSS with ωB97M(2) as a reference.

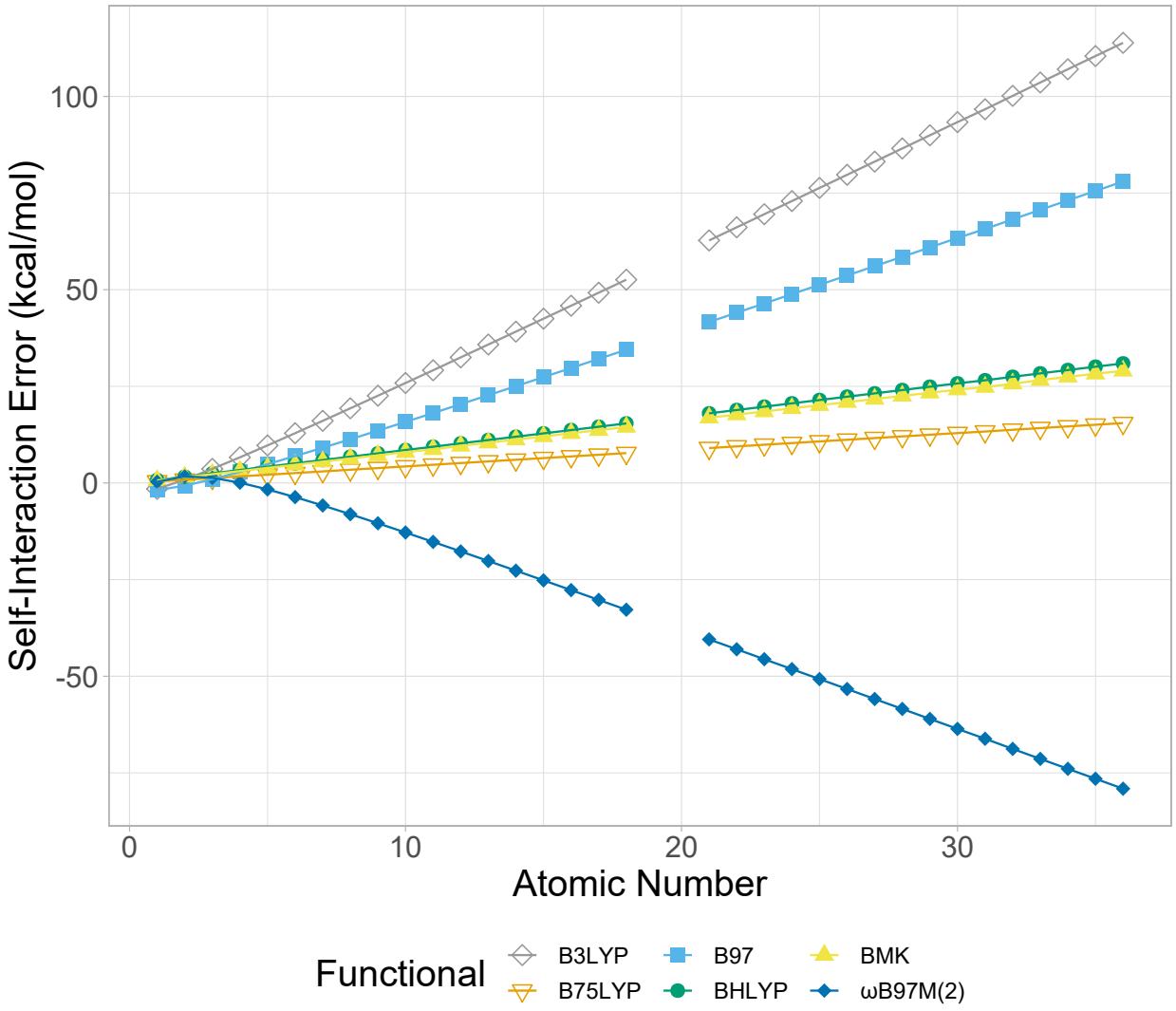

Figure S13: Self-interaction error vs increasing atomic charge of one-electron ions for B3LYP, B75LYP, B97, BHLYP, BMK with ωB97M(2) as a reference.

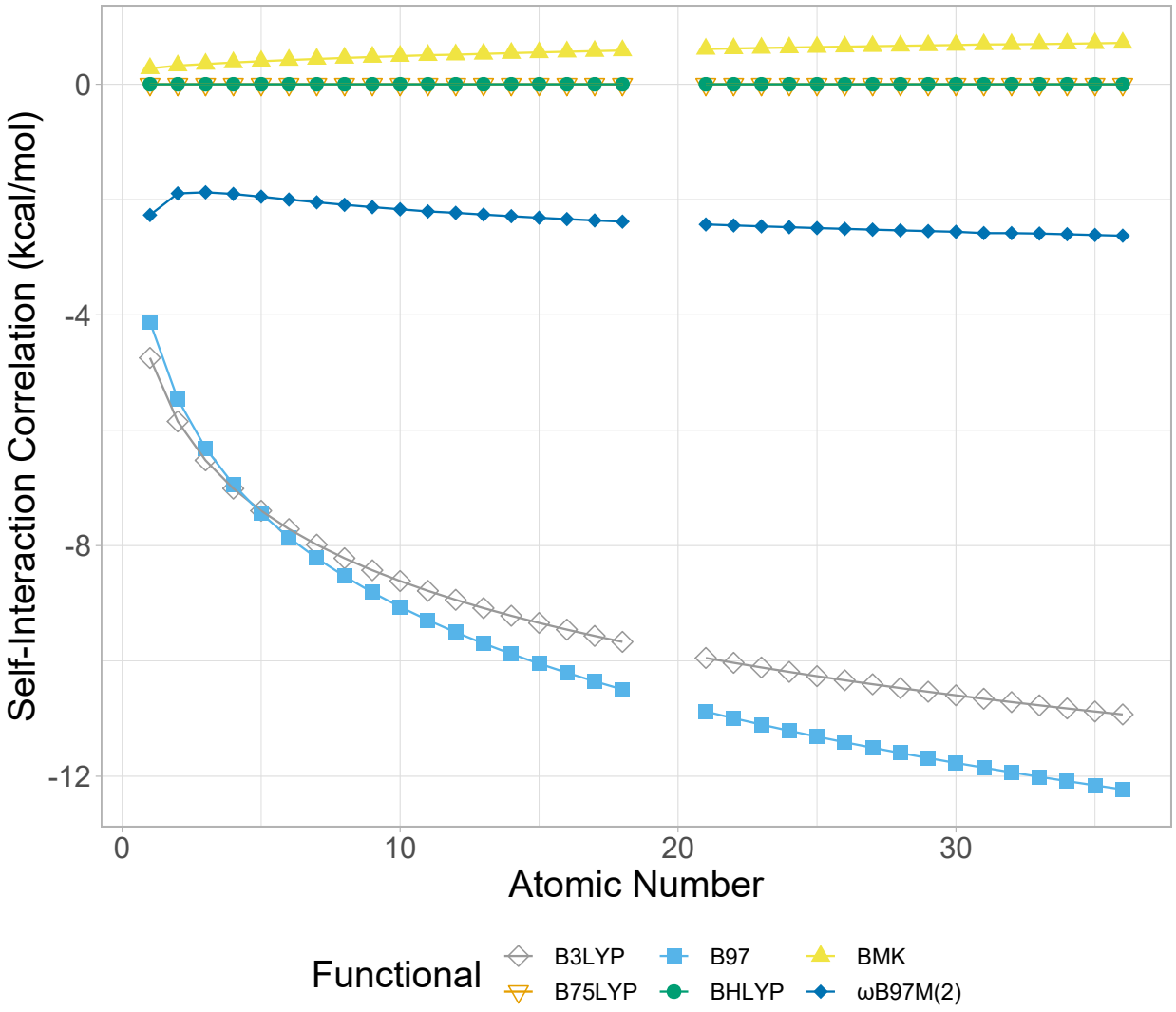

Figure S14: Self-interaction correlation vs increasing atomic charge of one-electron ions for B3LYP, B75LYP, B97, BHLYP, BMK with ωB97M(2) as a reference.

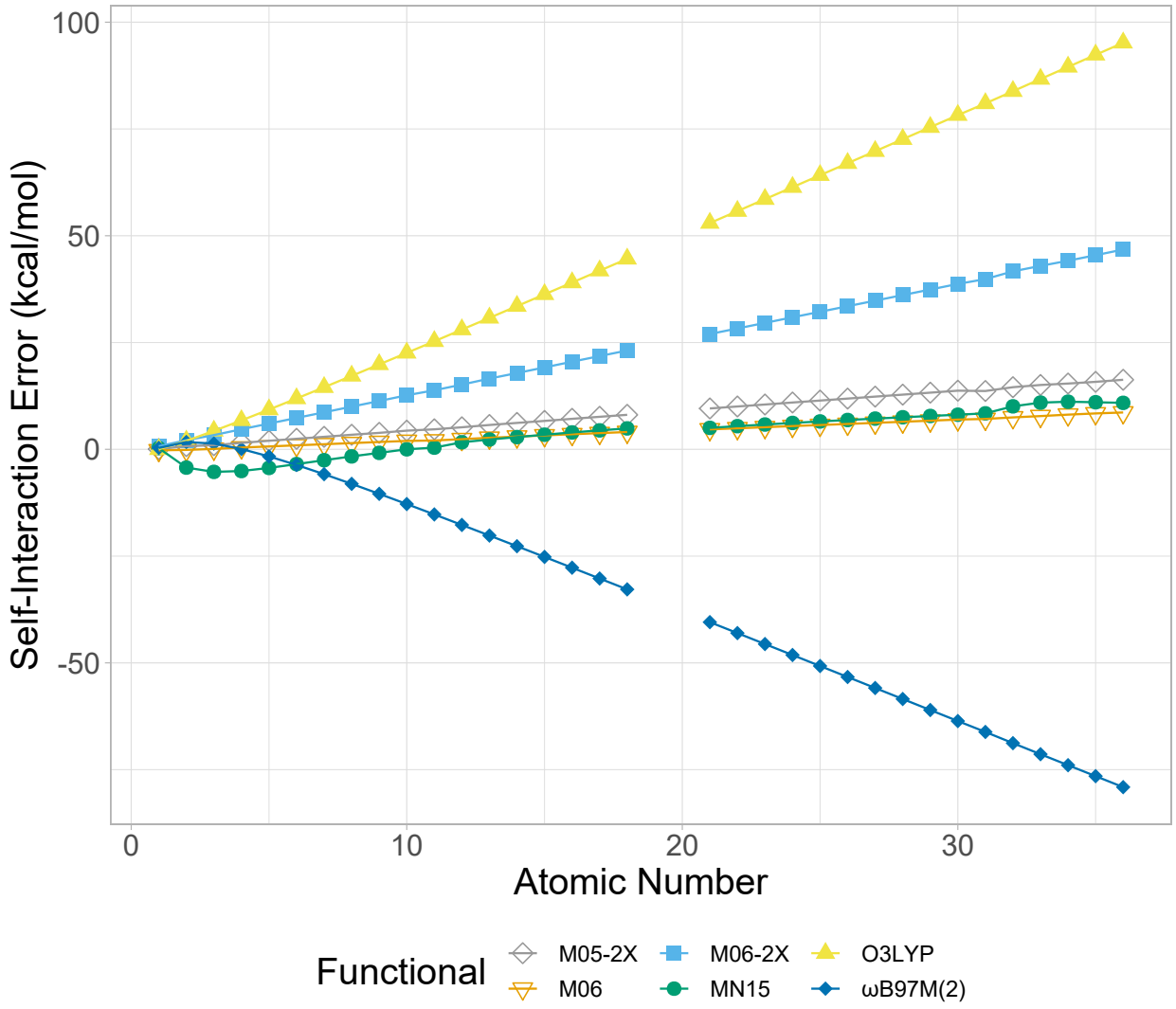

Figure S15: Self-interaction error vs increasing atomic charge of one-electron ions for M05-2X, M06, M06-2X, MN15, O3LYP with ωB97M(2) as a reference.

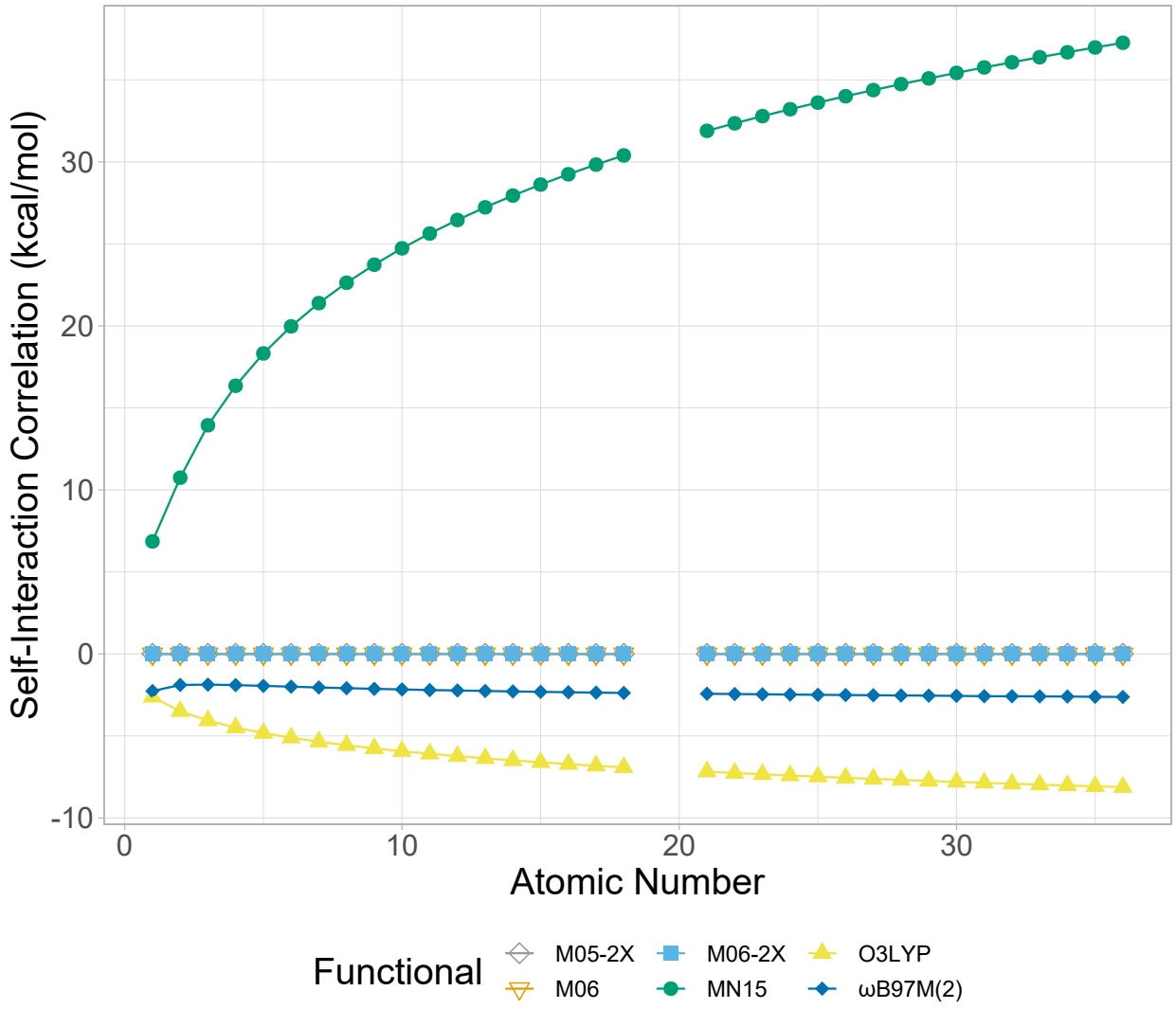

Figure S16: Self-interaction correlation vs increasing atomic charge of one-electron ions for M05-2X, M06, M06-2X, MN15, O3LYP with ωB97M(2) as a reference.

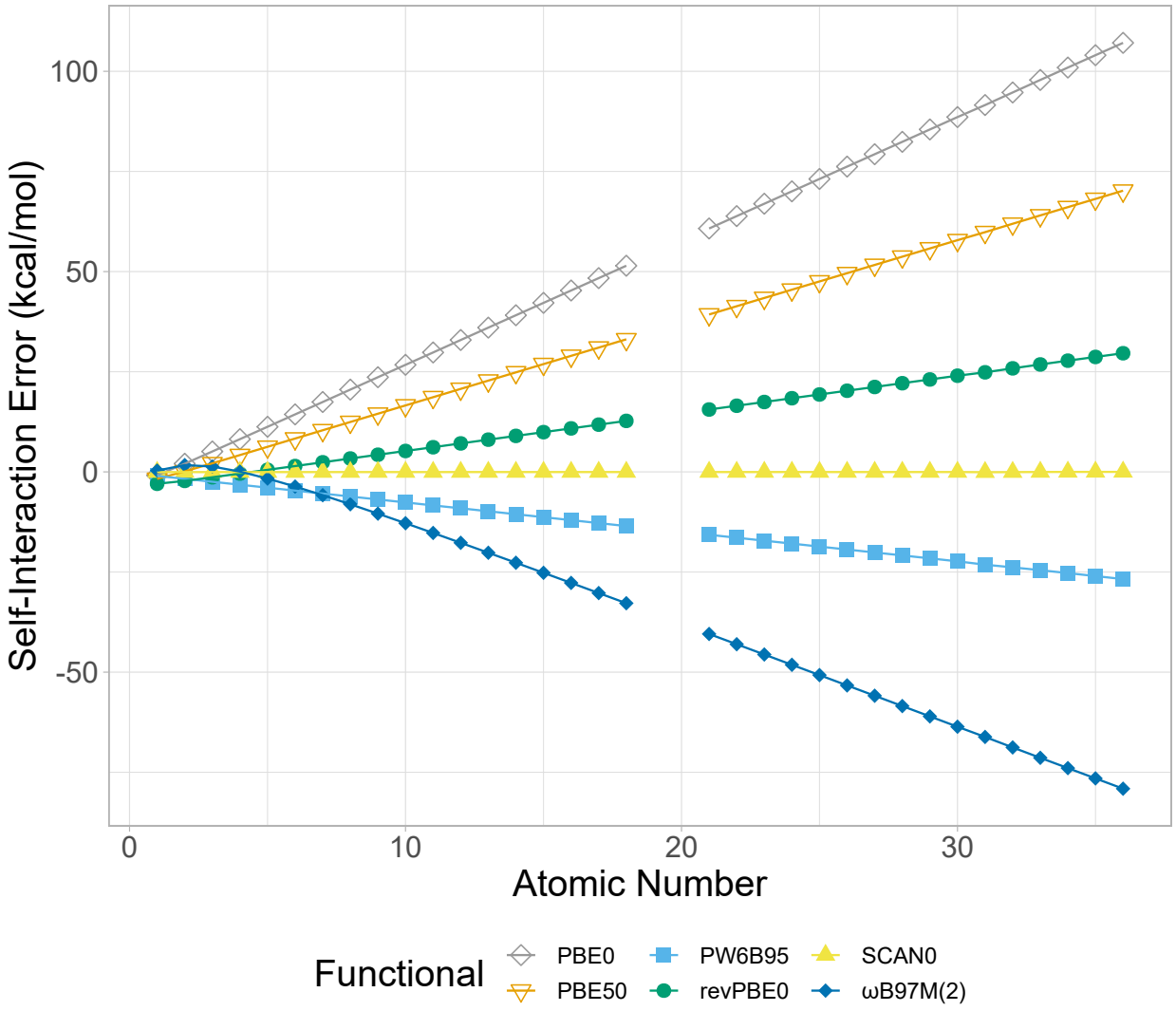

Figure S17: Self-interaction error vs increasing atomic charge of one-electron ions for PBE0, PBE50, PW6B95, revPBE0, SCAN0 with ωB97M(2) as a reference.

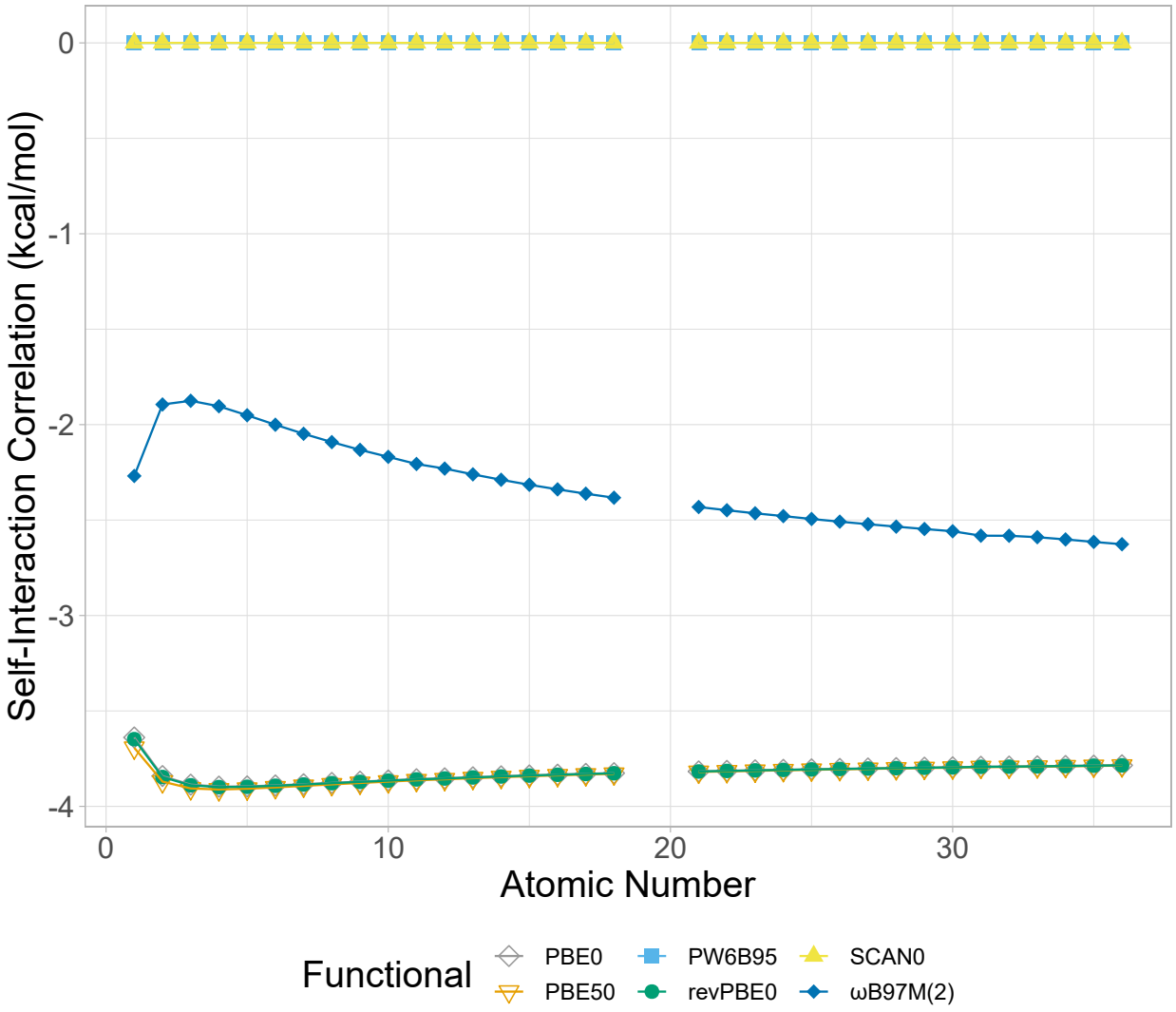

Figure S18: Self-interaction correlation vs increasing atomic charge of one-electron ions for PBE0, PBE50, PW6B95, revPBE0, SCAN0 with ωB97M(2) as a reference.

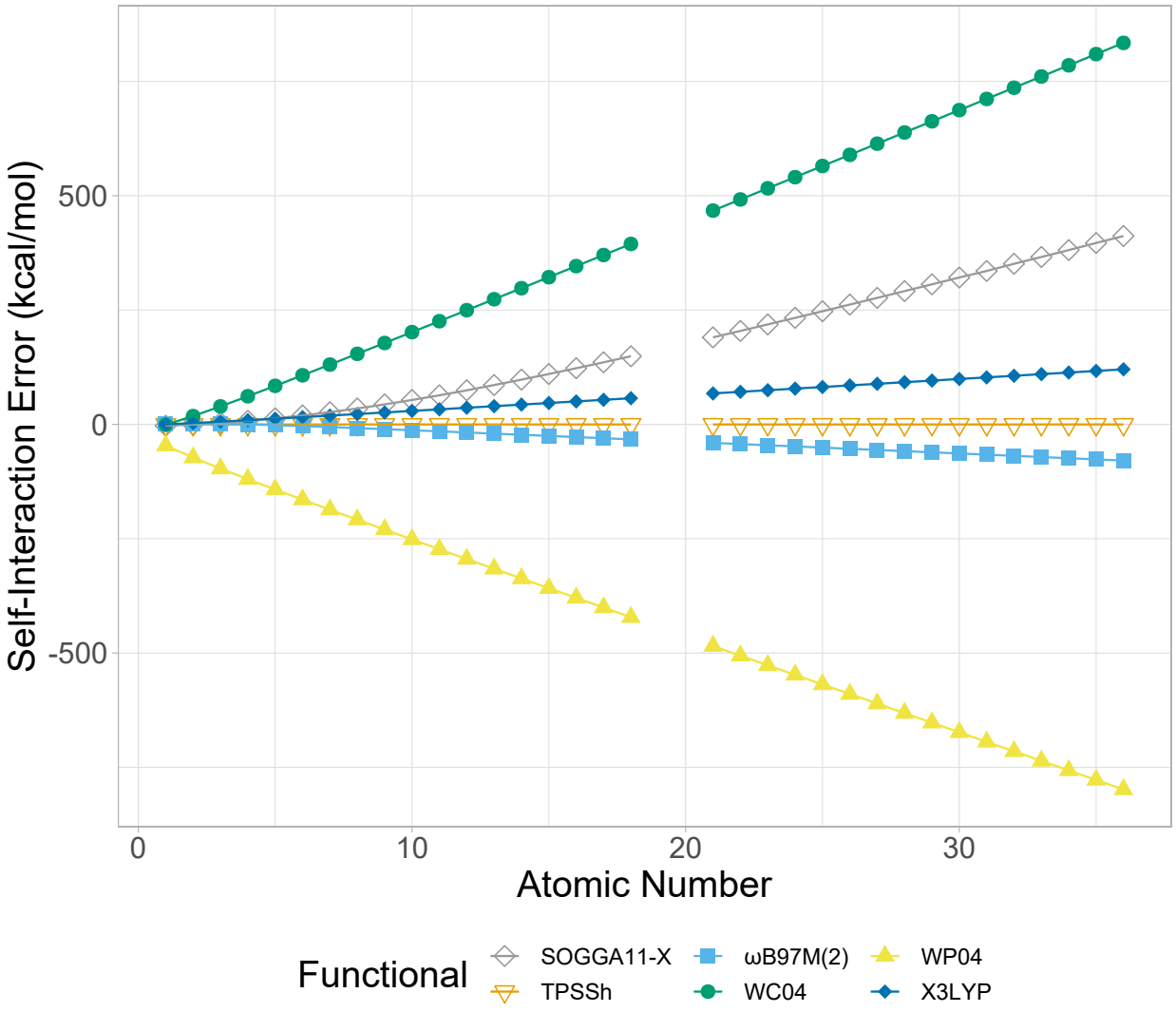

Figure S19: Self-interaction error vs increasing atomic charge of one-electron ions for SOGGA11-X, TPSSh, WC04, WP04, X3LYP with ωB97M(2) as a reference.

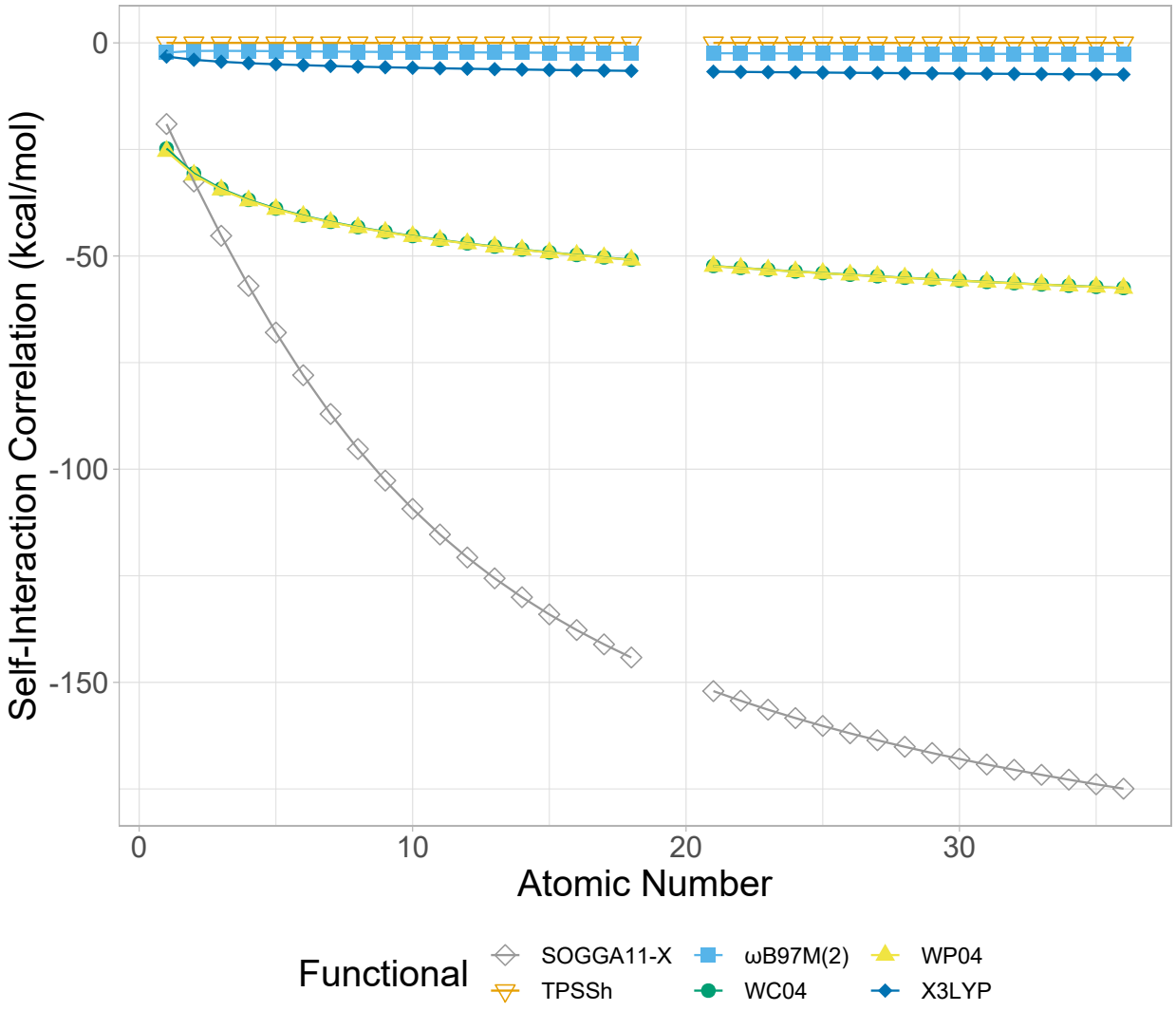

Figure S20: Self-interaction correlation vs increasing atomic charge of one-electron ions for SOGGA11-X, TPSSh, WC04, WP04, X3LYP with ωB97M(2) as a reference.

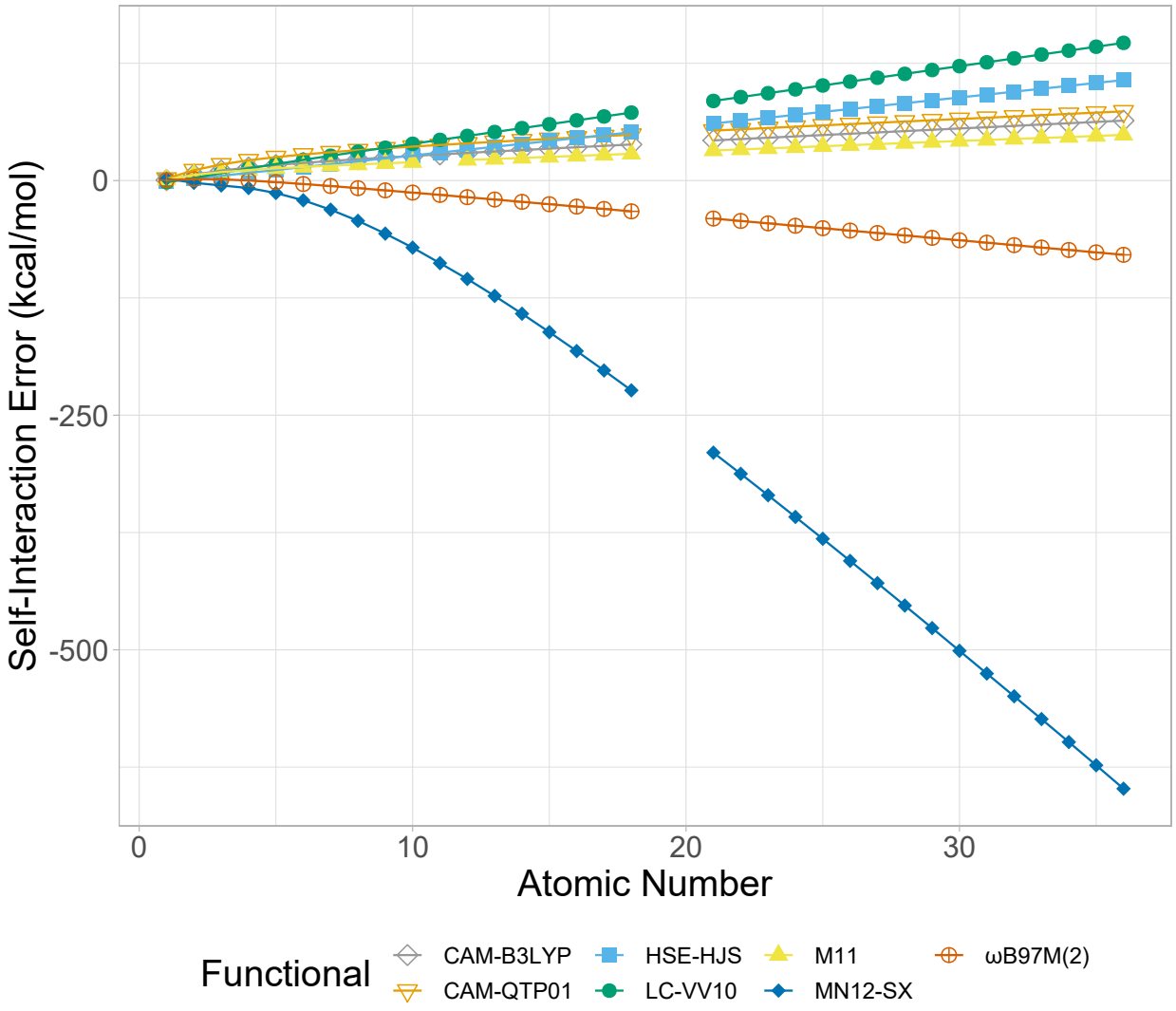

Figure S21: Self-interaction error vs increasing atomic charge of one-electron ions for CAM-B3LYP, CAM-QTP01, HSE-HJS, LC-VV10, M11, MN12-SX with ωB97M(2) as a reference.

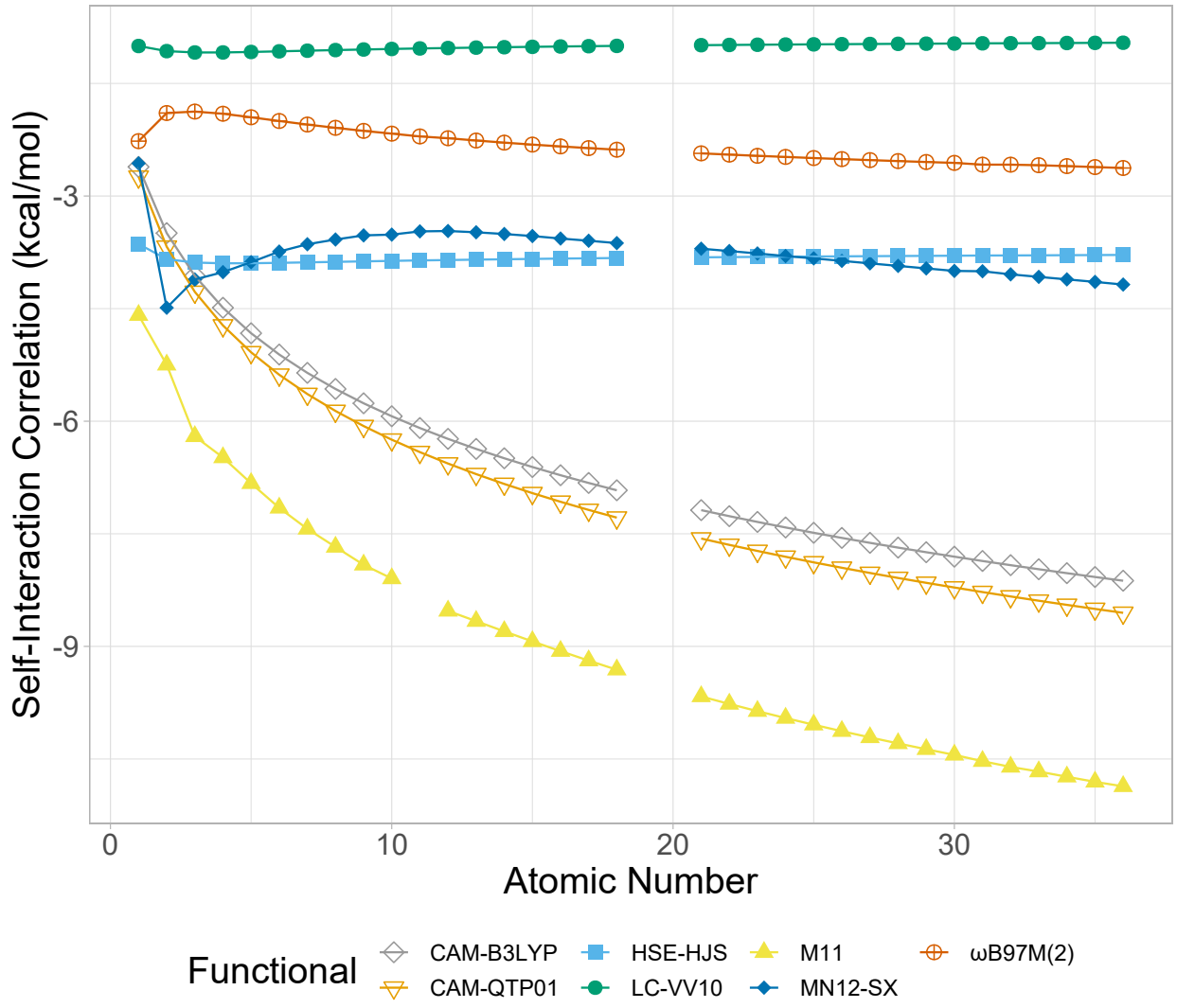

Figure S22: Self-interaction correlation vs increasing atomic charge of one-electron ions for CAM-B3LYP, CAM-QTP01, HSE-HJS, LC-VV10, M11, MN12-SX with ωB97M(2) as a reference.

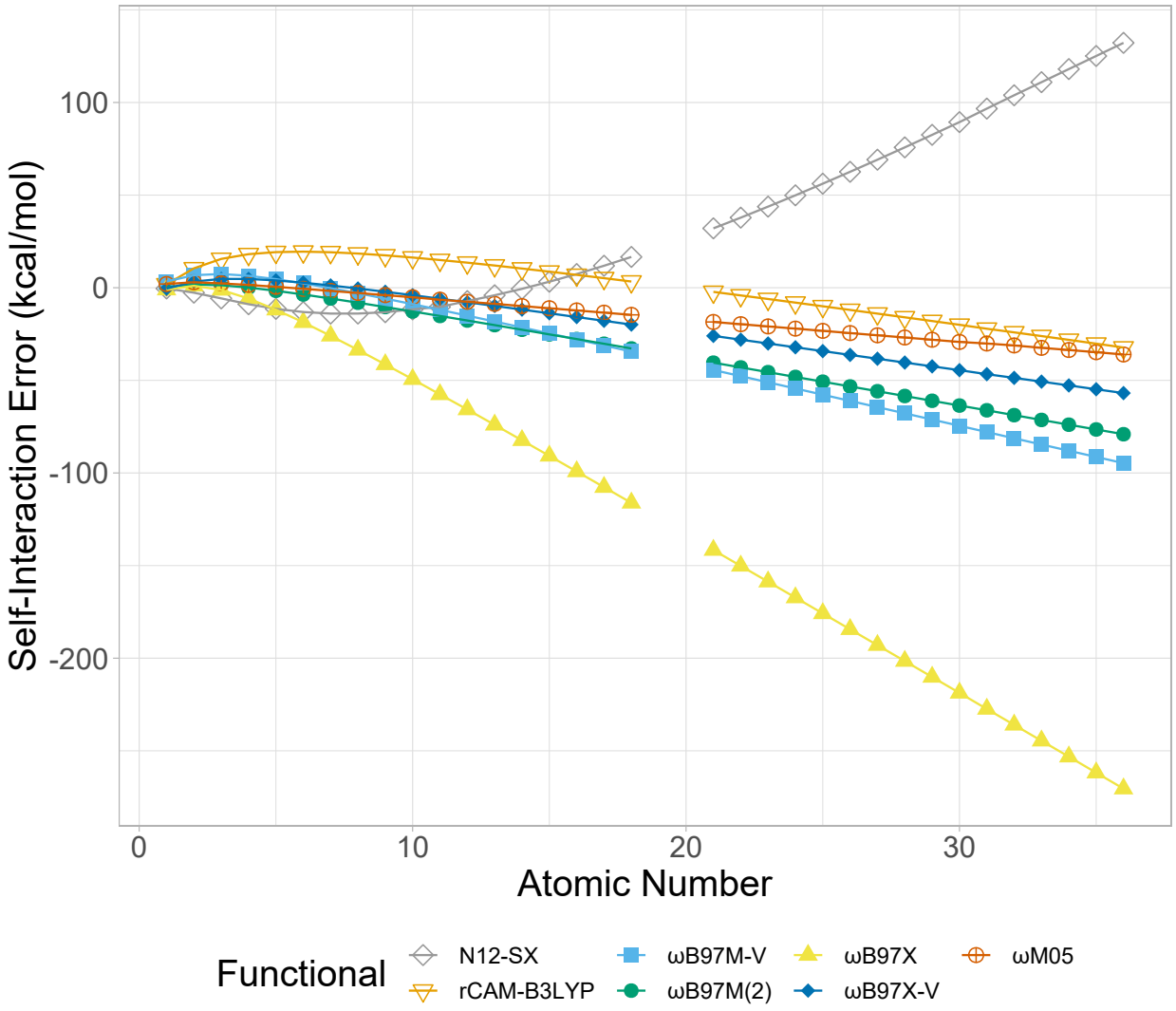

Figure S23: Self-interaction error vs increasing atomic charge of one-electron ions for N12-SX, rCAM-B3LYP, ωB97M-V, ωB97X, ωB97X-V, ωM05 with ωB97M(2) as a reference.

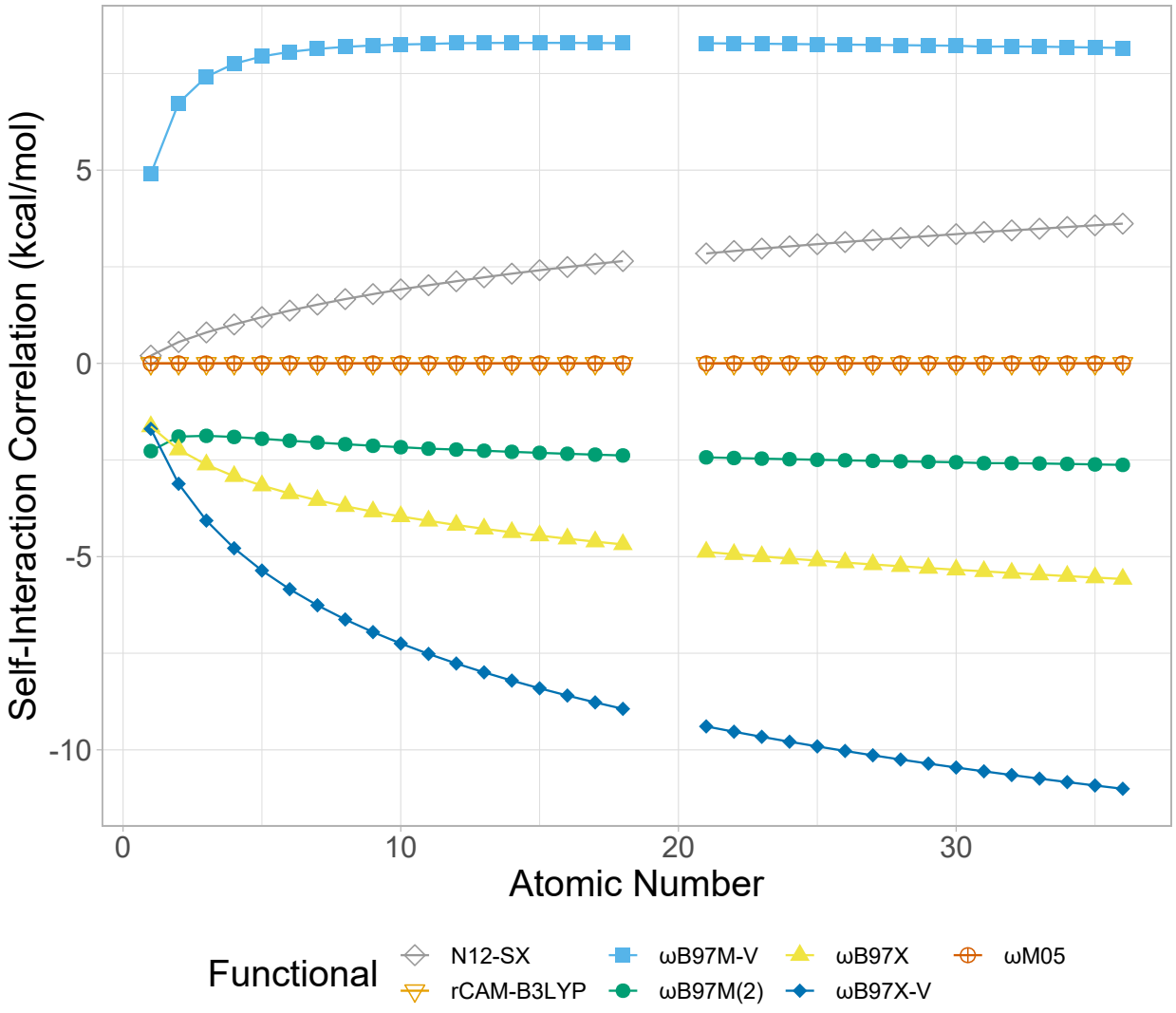

Figure S24: Self-interaction correlation vs increasing atomic charge of one-electron ions for N12-SX, rCAM-B3LYP, ωB97M-V, ωB97X, ωB97X-V, ωM05 with ωB97M(2) as a reference.

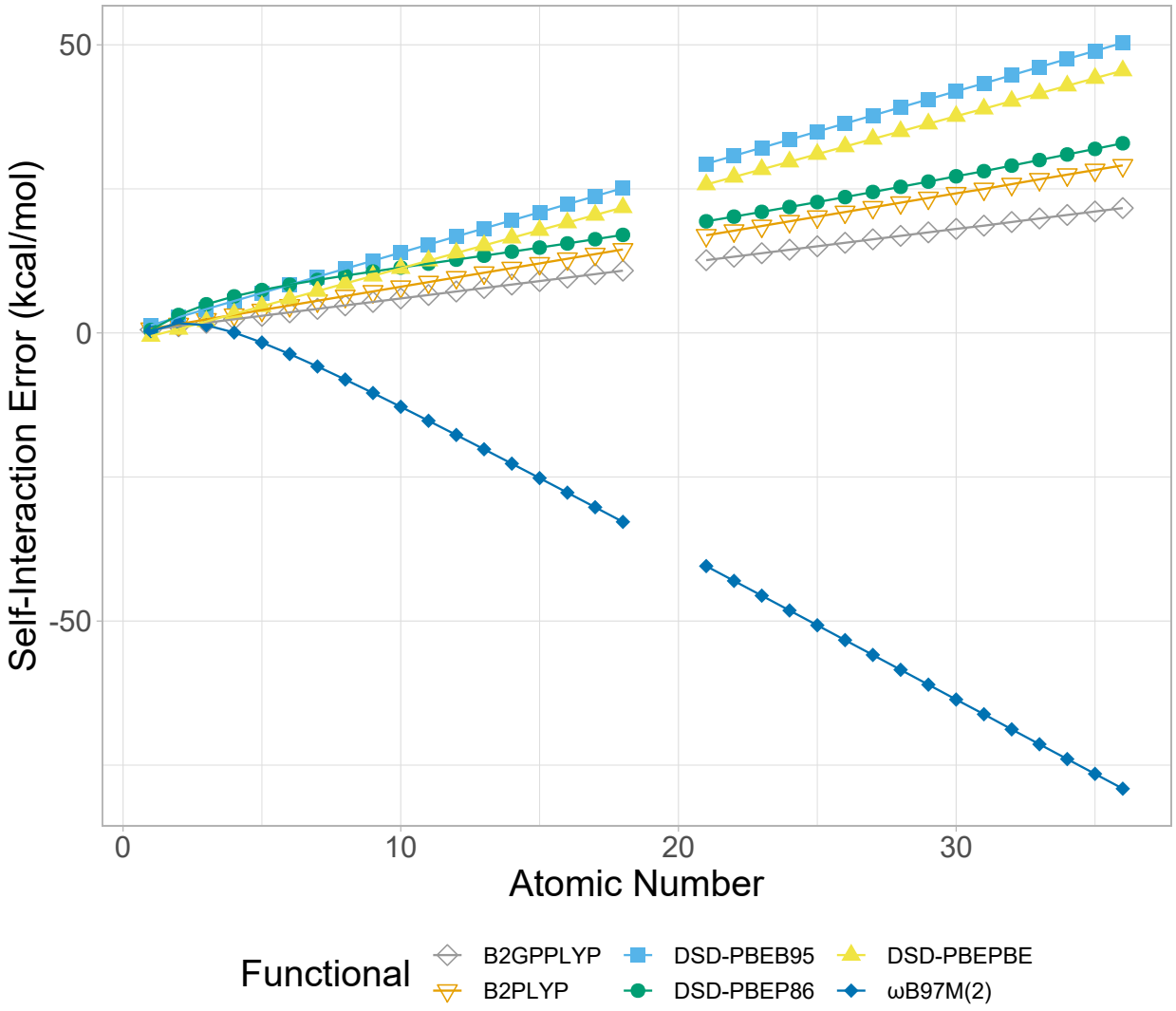

Figure S25: Self-interaction error vs increasing atomic charge of one-electron ions for B2GPPLYP, B2PLYP, DSD-PBEB95, DSD-PBEP86, DSD-PBEPBE with ωB97M(2) as a reference.

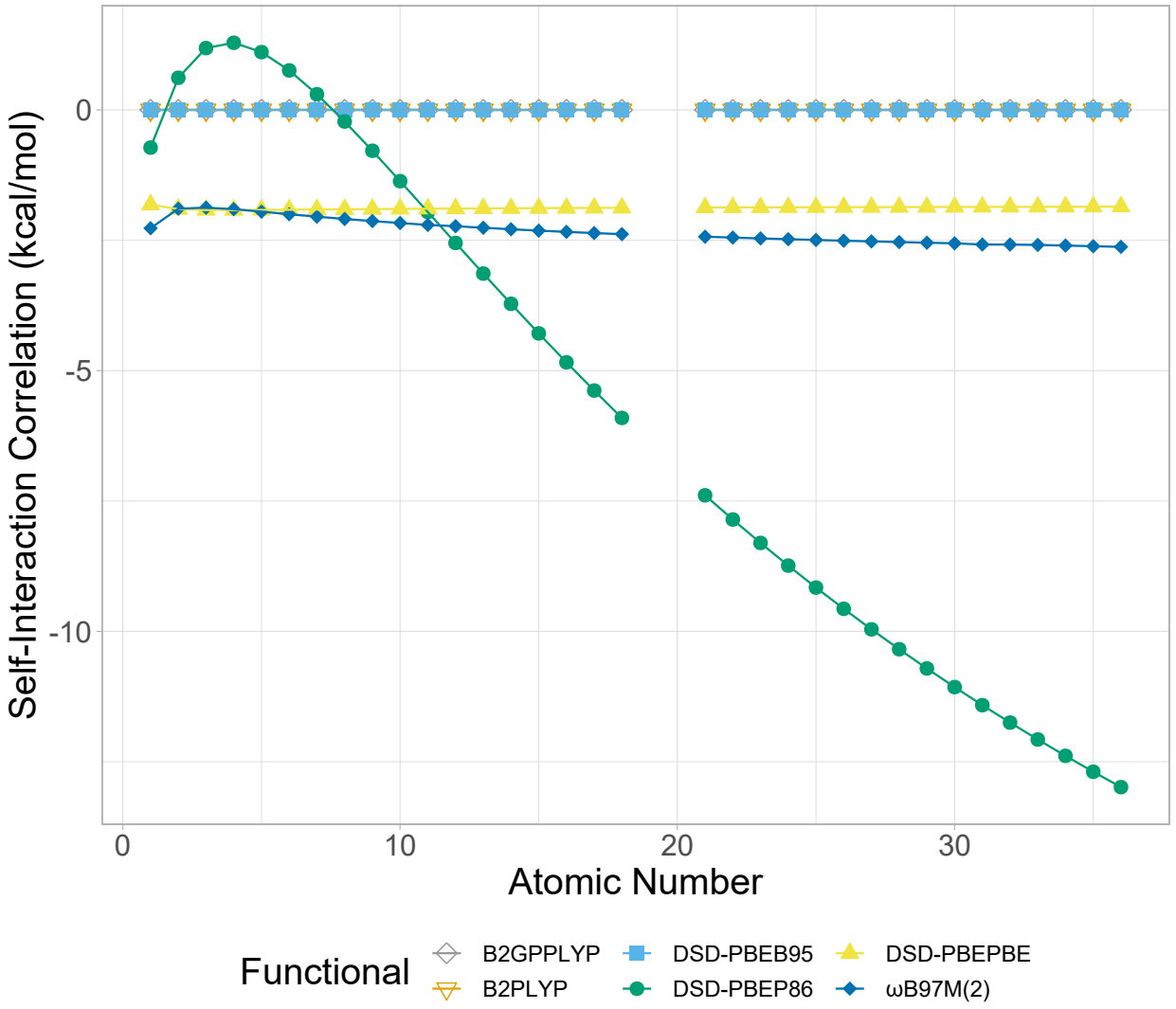

Figure S26: Self-interaction correlation vs increasing atomic charge of one-electron ions for B2GPPLYP, B2PLYP, DSD-PBEB95, DSD-PBEP86, DSD-PBEPBE with ωB97M(2) as a reference.

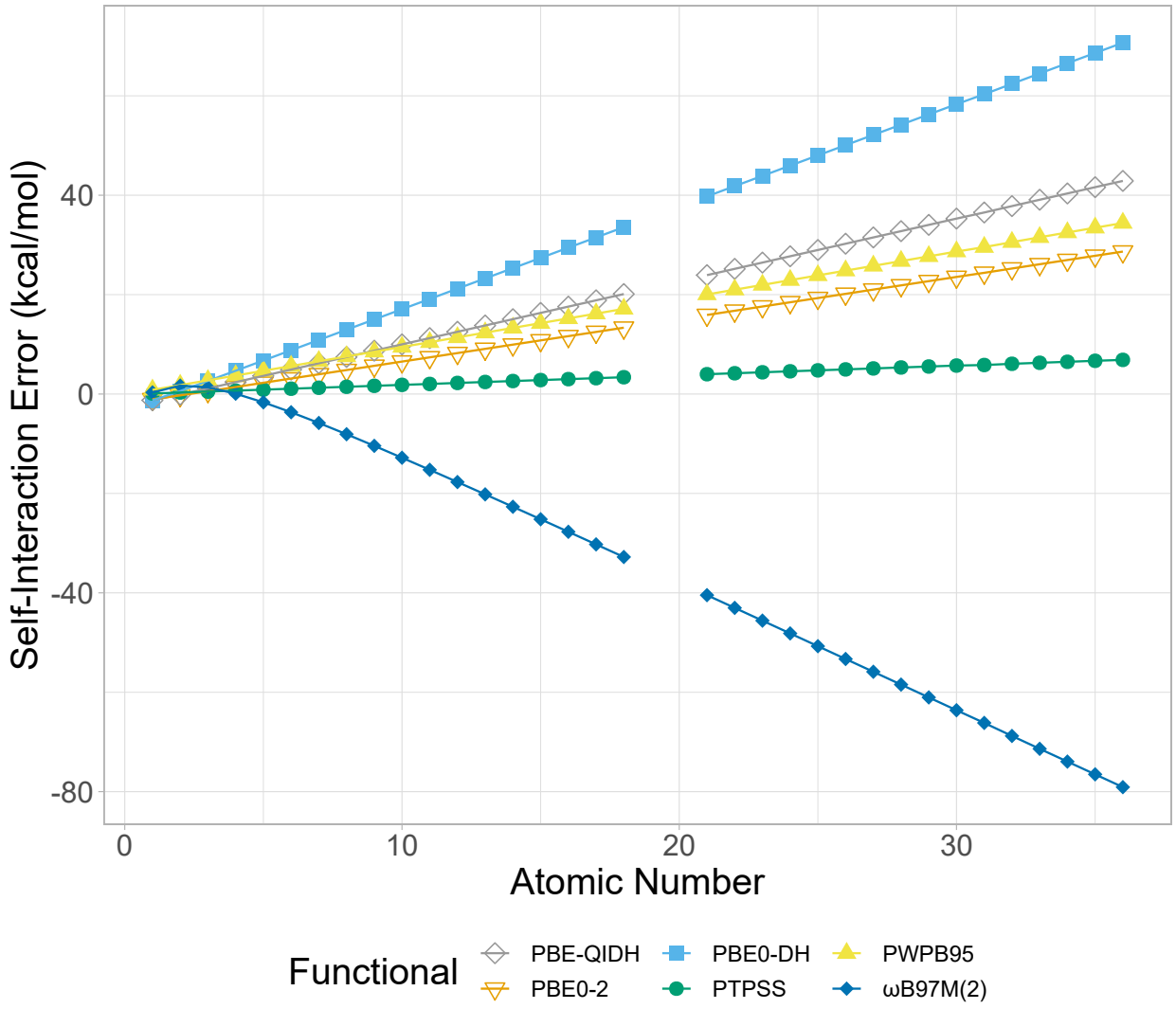

Figure S27: Self-interaction error vs increasing atomic charge of one-electron ions for PBE-QIDH, PBE0-2, PBE0-DH, PTPSS, PWPB95 with ωB97M(2) as a reference.

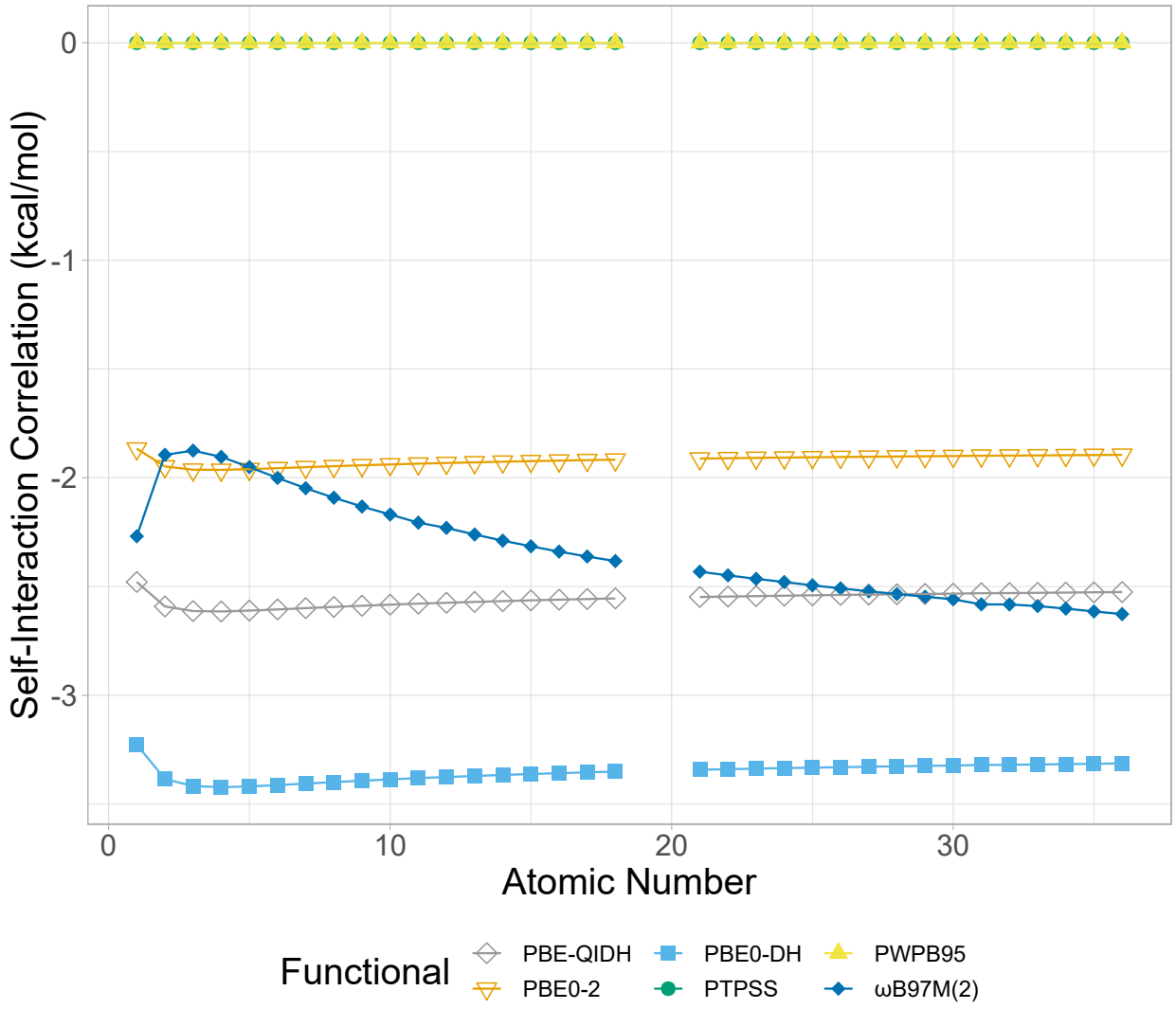

Figure S28: Self-interaction correlation vs increasing atomic charge of one-electron ions for PBE-QIDH, PBE0-2, PBE0-DH, PTPSS, PWPB95 with ωB97M(2) as a reference.

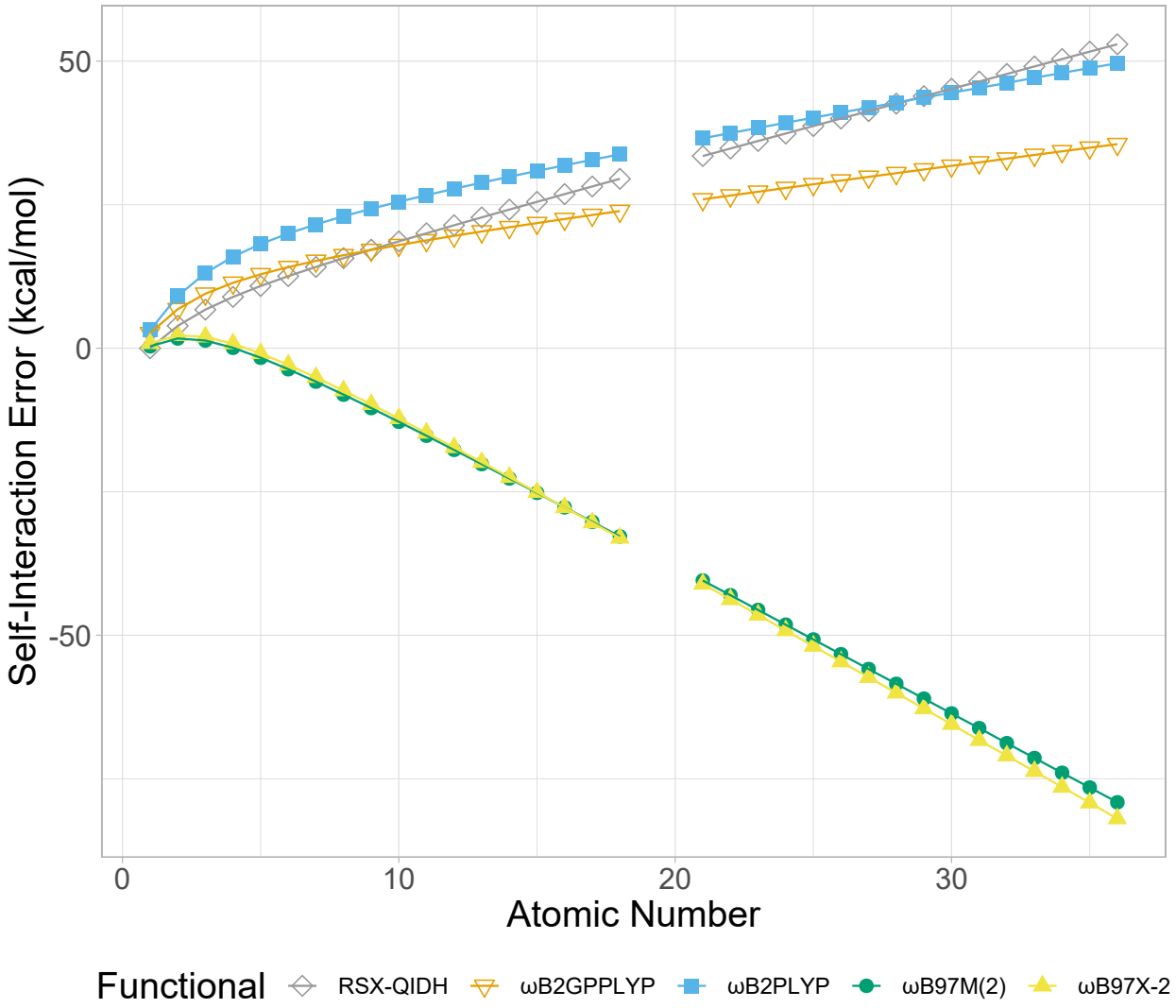

Figure S29: Self-interaction error vs increasing atomic charge of one-electron ions for RSX-QIDH, ωB2GPPLYP, ωB2PLYP, ωB97X-2 with ωB97M(2) as a reference.

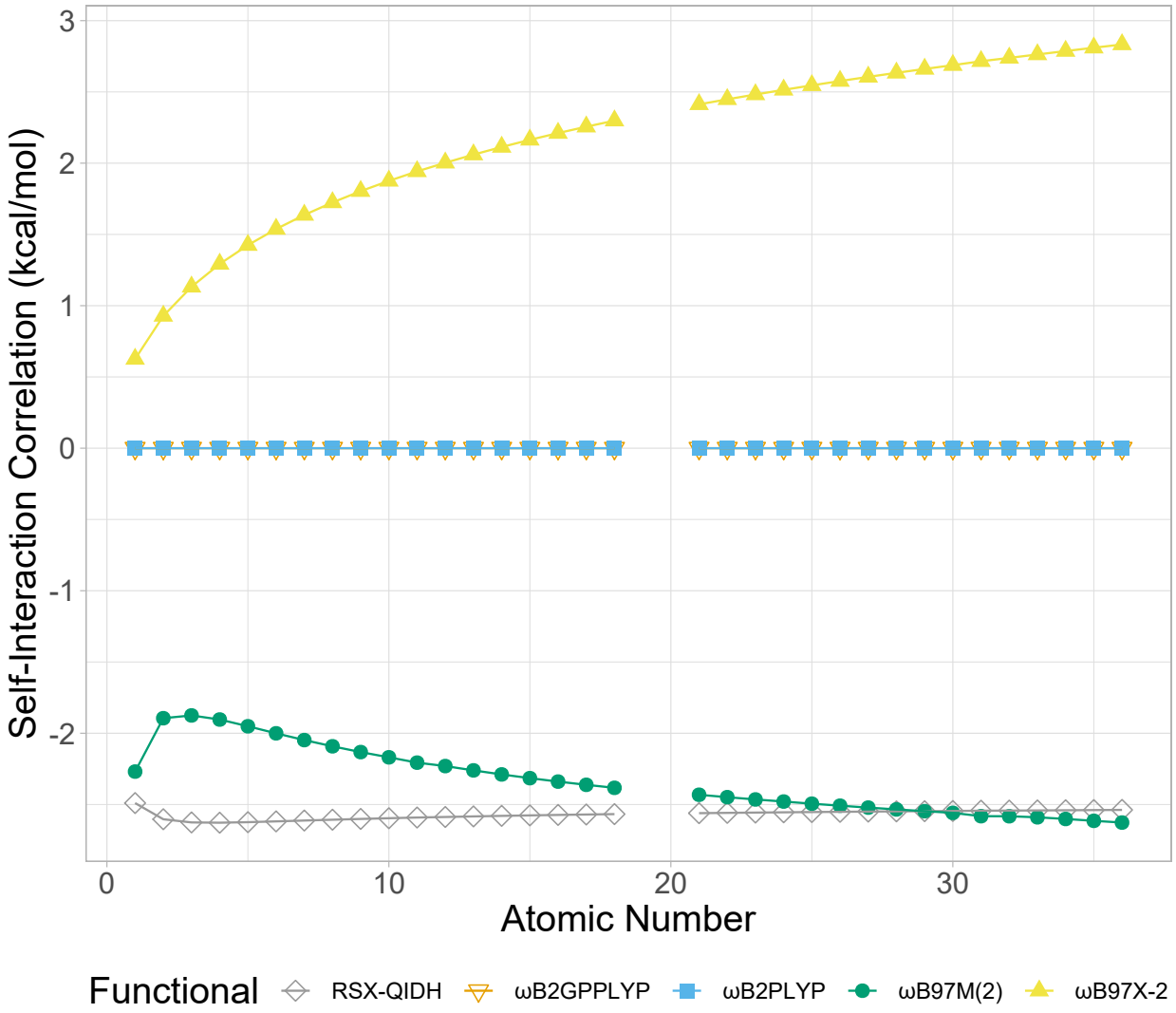

Figure S30: Self-interaction correlation vs increasing atomic charge of one-electron ions for RSX-QIDH, ωB2GPPLYP, ωB2PLYP, ωB97X-2 with ωB97M(2) as a reference.

SI.2.2 Density and Functional error

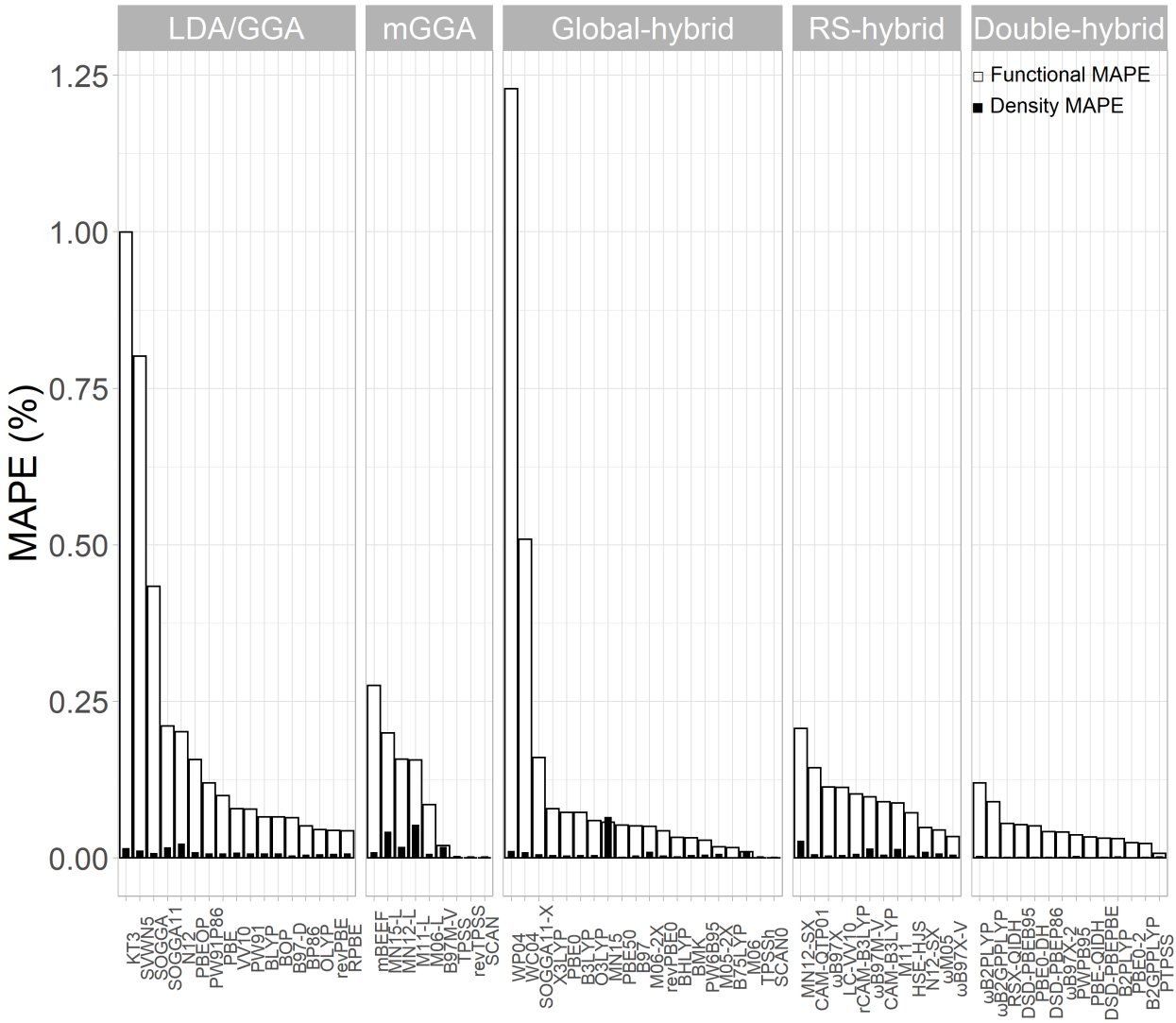

Figure S31: Functional (white) and density (black) mean absolute percentage errors across the entire mononuclear dataset, from H to Kr^{35+}, for each density functional approximation.

SI.2.3 Self-interaction basis set error for select DFAs

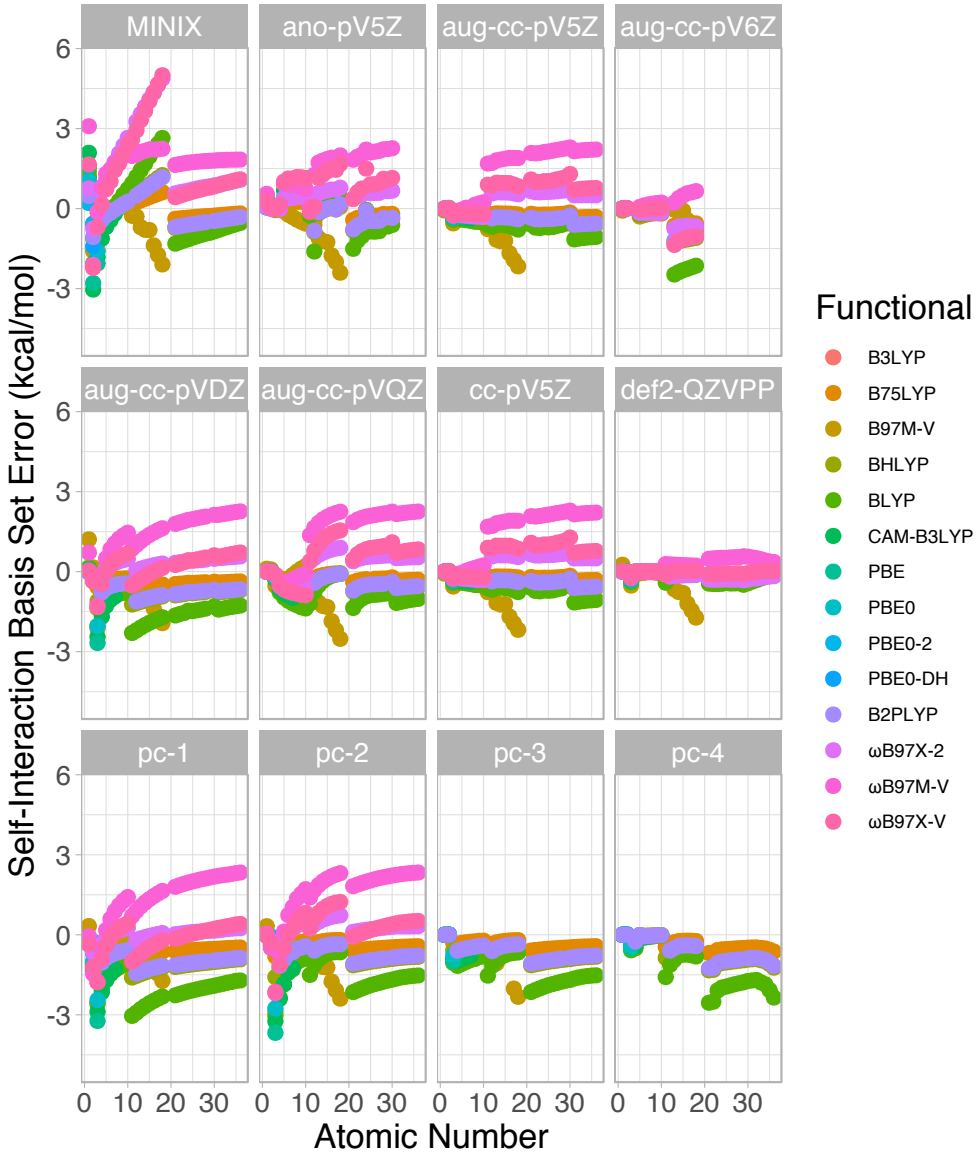

Figure S32: Self-interaction basis set error across the entire mononuclear series dataset, from H to Kr^{35+}, for select density functional approximations. Select DFAs do not give values for all systems, likewise, certain basis sets are only defined for a set number of atoms. The scale was adjusted to ensure the majority of points are legible, although some results lie outside these bounds.

SI.2.3.1 Basis set contraction of 1s-orbitals

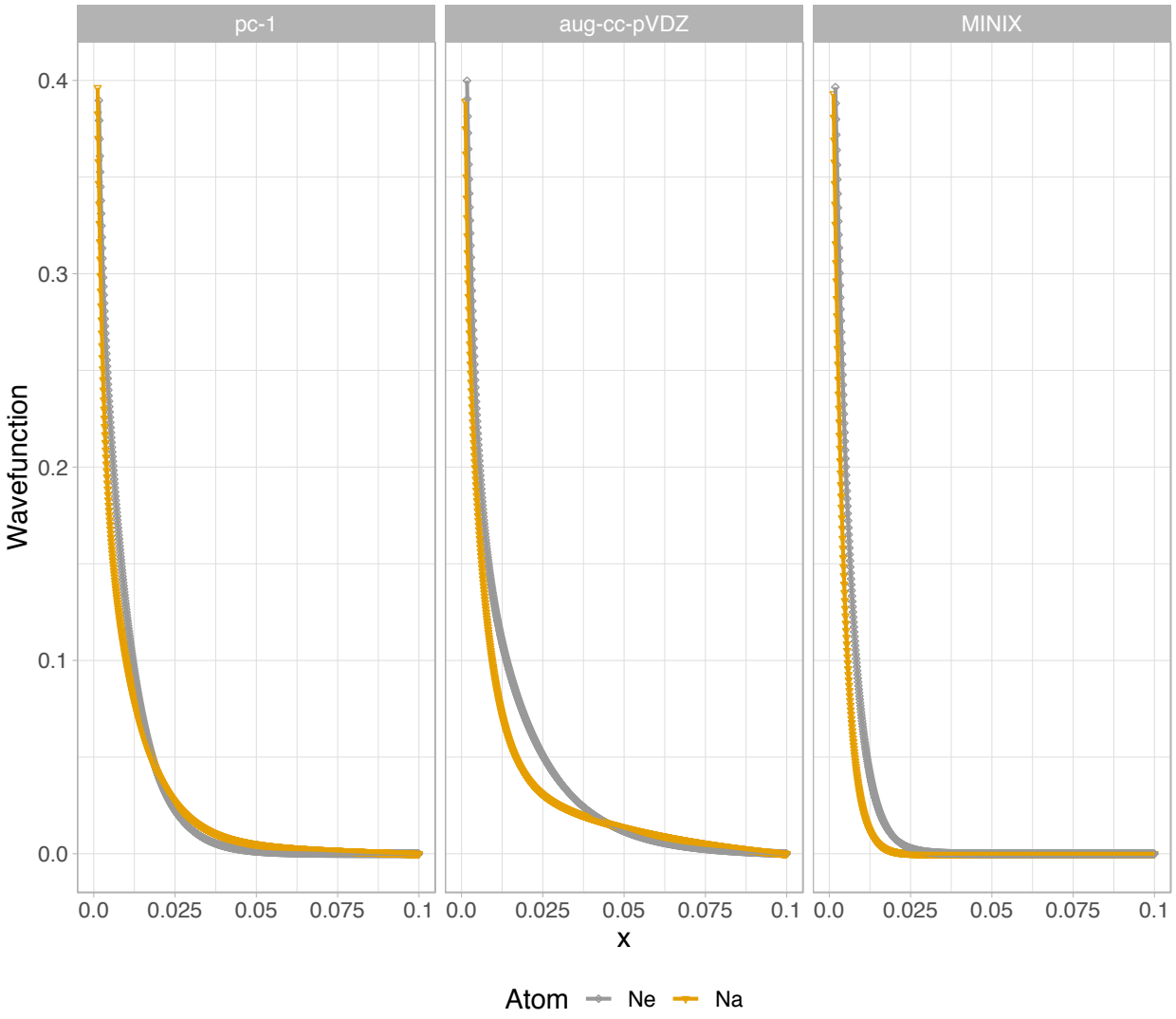

Figure S33: Normalised radial part of the contracted Gaussian-type 1s-orbitals. This is shown for the basis sets pc-1, aug-cc-pVDZ, and MINIX for neon and sodium.

Exponents and coefficients for the basis sets found in Fig. S33 are courtesy of the basis set exchange. [1]

[1] 1. B. P. Pritchard, D. Altarawy, B. Didier, T. D. Gibson and T. L. Windus. *J. Chem. Inf. Model.*, 2019, **59**, 4814–4820

SI.2.4 Dispersion correction and non-local correlation

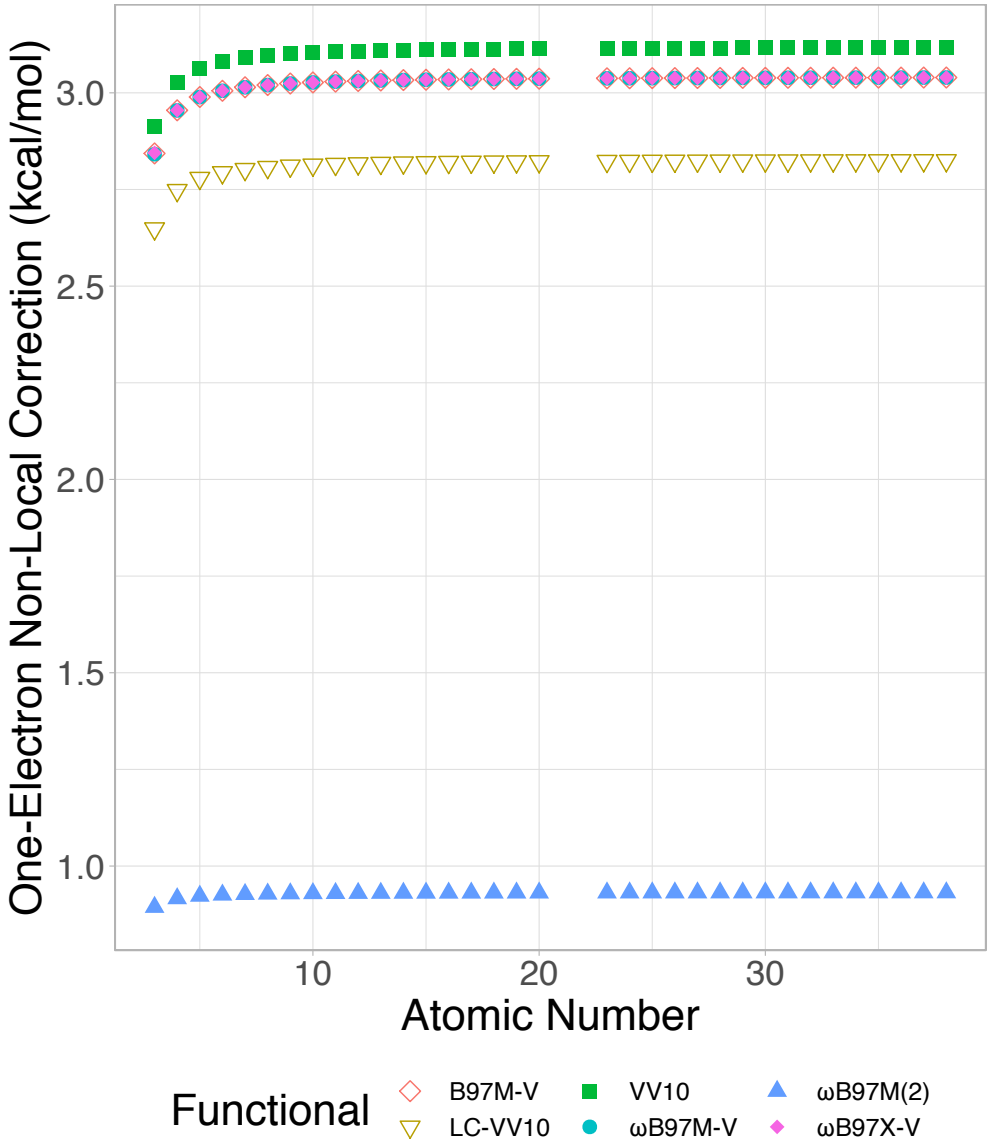

Figure S34: Non-local van der Waals correction values for the mononuclear series dataset for B97M-V, LC-VV10, VV10, ωB97M-V, ωB97M(2), and ωB97X-V.

SI.2.5 Linear relationship between the self-interaction error and the nuclear charge

The radius of hydrogenic systems is inversely proportional to the nuclear charge Z, and the same therefore also applies to the average extension $\bar{r}$ of 1s orbitals.
We can also see this from the expectation value of the distance r,[2] $\bar{r}$, which is:

$$\bar{r} = \langle \psi_{1s} | r | \psi_{1s} \rangle = \frac{3a_0}{2Z}, \tag{1}$$

where a_0 is the bohr radius and Z is the nuclear charge.
Substituting 1 into the nuclear-electron attraction expression we recover the Z^2 dependence of the energy of one-electron nuclei:

$$\frac{eZ}{\bar{r}} = \frac{eZ^2}{3a_0}, \tag{2}$$

where e is the charge of the electron.
Given that self-interaction is an electron-electron interaction, we can write an equivalent expression to 2 as:

$$\frac{e \times e}{\bar{r}} = \frac{e^2 Z}{3a_0}. \tag{3}$$

In order to assess the validity of the above, we fit straight lines to data from the mononuclear series dataset, hydrogenic systems from H to Kr^{35+}. To get an unbiased representation of the actual exchange and Coulomb energy we fit the functional error, SIE^{func}, which is free from any density error components, and subtracted away any self-correlation which is not accounted for in the above. This fit led to table S1, which contains the R^2 result for those fits. TPSS- and SCAN-based DFAs can be ignored as they are essentially one-electron SIE free. Among the remaining DFAs, we can see that several Minnesota and range-separated functionals have non-linearities and go against the rationale of the above.

Table S1: The table of R^2 values from fitting the functional error to a straight line, SIE^{func}, subtracted by the self-interaction correlation error, E_C^{SIE}. Density functional approximations based off TPSS and SCAN can be ignored as they are essentially one-electron SIE free, and their R^2 values come primarily from computational noise or minor imperfections in the cancellation between the self-exchange and self-Coulomb values.

DFA	R^2	DFA	R^2	DFA	R^2	DFA	R^2
B2GPPLYP	0.9999985	KT3	0.9998616	PBE-QIDH	0.9999997	SCAN0	0.2549449
B2PLYP	0.9999985	LC-VV10	0.999685	PBE0	0.9999996	SOGGA	1
B3LYP	0.9999998	M05-2X	0.9995702	PBE0-2	0.9999997	SOGGA11	0.9998677
B75LYP	0.9999985	M06	0.9998611	PBE0-DH	0.9999997	SOGGA11-X	0.9999987
B97	0.9999995	M06-2X	0.9999779	PBE50	0.9999997	SVWN5	1
B97-D	0.9999998	M06-L	0.9998829	PBEOP	0.9999997	TPSS	0.003219561
B97M-V	0.9986407	M11	0.9790161	PTPSS	0.9999406	TPSSh	0.003219562
BHLYP	0.9999985	M11-L	0.9999071	PW6B95	0.9999943	VV10	0.9999959
BLYP	0.9999985	mBEEF	0.9999996	PW91	0.9999993	ωB2GPPLYP	0.9637068
BMK	0.9999768	MN12-L	0.9878491	PW91P86	0.9999994	ωB2PLYP	0.9568088
BOP	0.9999985	MN12-SX	0.9797302	PWPB95	0.9999986	ωB97M-V	0.9950312
BP86	0.9999977	MN15	0.8988953	rCAM-B3LYP	0.8860715	ωB97M(2)	0.9950312
CAM-B3LYP	0.9783641	MN15-L	0.9893211	revPBE	0.9999945	ωB97X	0.9962682
CAM-QTP01	0.9545367	N12	0.949041	revPBE0	0.9999955	ωB97X-2	0.9958574
DSD-PBEB95	0.9999997	N12-SX	0.901572	revTPSS	0.7687985	ωB97X-V	0.9658405
DSD-PBEP86	0.9999997	O3LYP	0.9999997	RSX-QIDH	0.9936078	WC04	1
DSD-PBEPBE	0.9999997	OLYP	0.9999982	RPBE	0.9999856	ωM05	0.9978063
HSE-HJS	0.9999996	PBE	0.9999995	SCAN	0.2549449	WP04	0.9999999
						X3LYP	0.9999998

[2] Y. B. Bend, *Quantum mechanics with applications to nanotechnology and information science*, Academic Press, 1st edn., 2012.

SI.3 Diatomic series

SI.3.1 Hydrogen cation plots

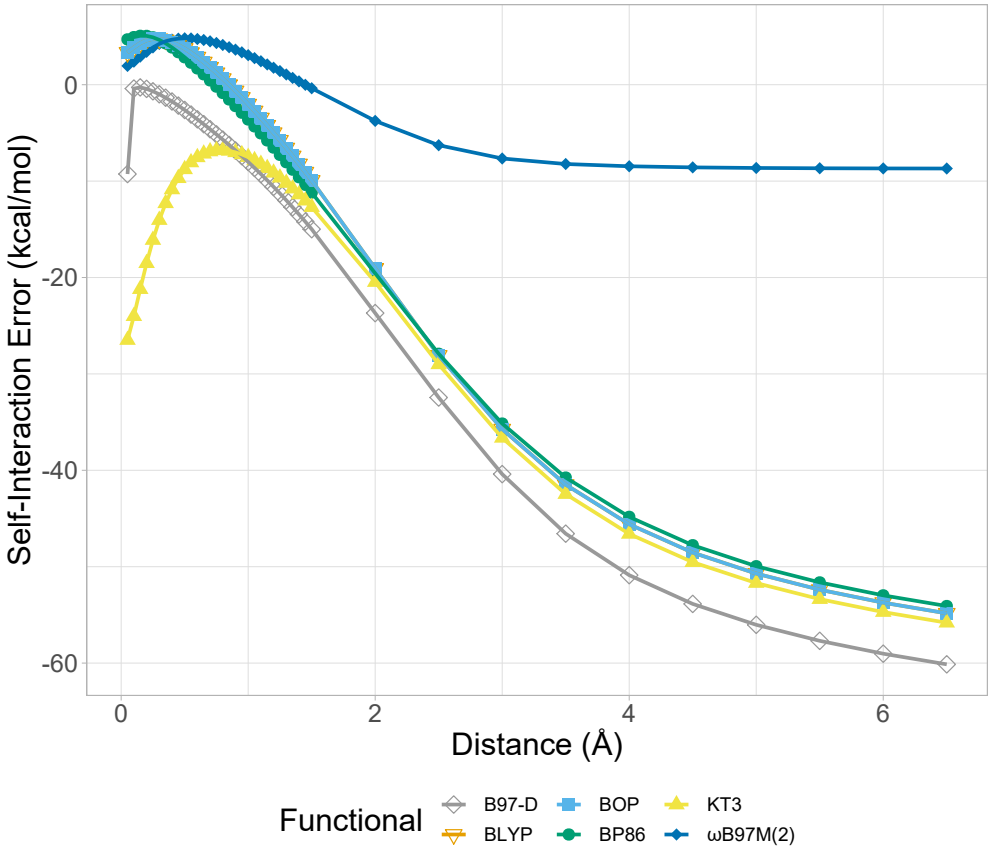

Figure S35: Self-interaction error vs internuclear distance of the H_2^+ molecule for B97-D, BLYP, BOP, BP86, KT3 with ωB97M(2) as a reference.

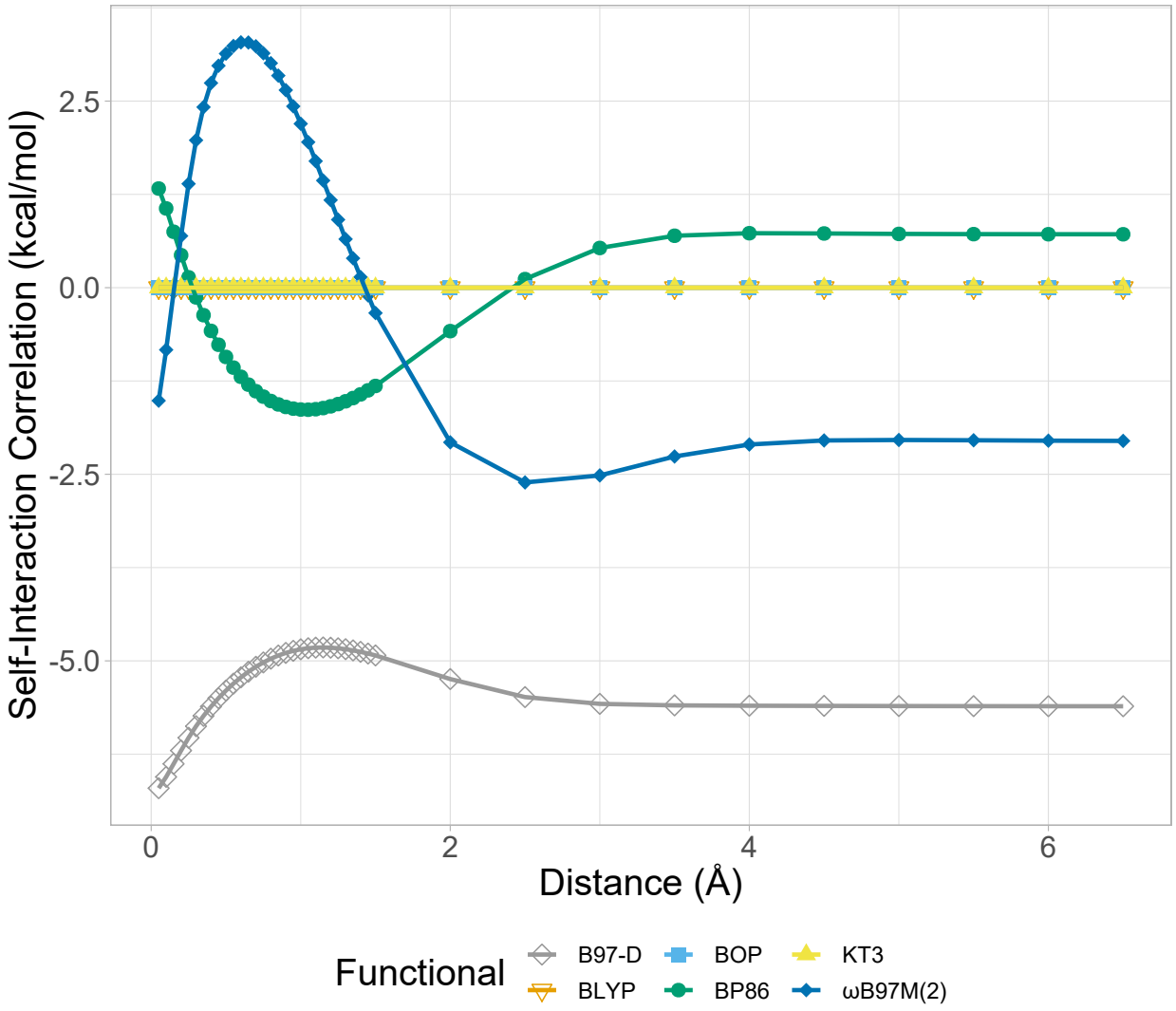

Figure S36: Self-interaction correlation error vs internuclear distance of the H_2^+ molecule for B97-D, BLYP, BOP, BP86, KT3 with ωB97M(2) as a reference.

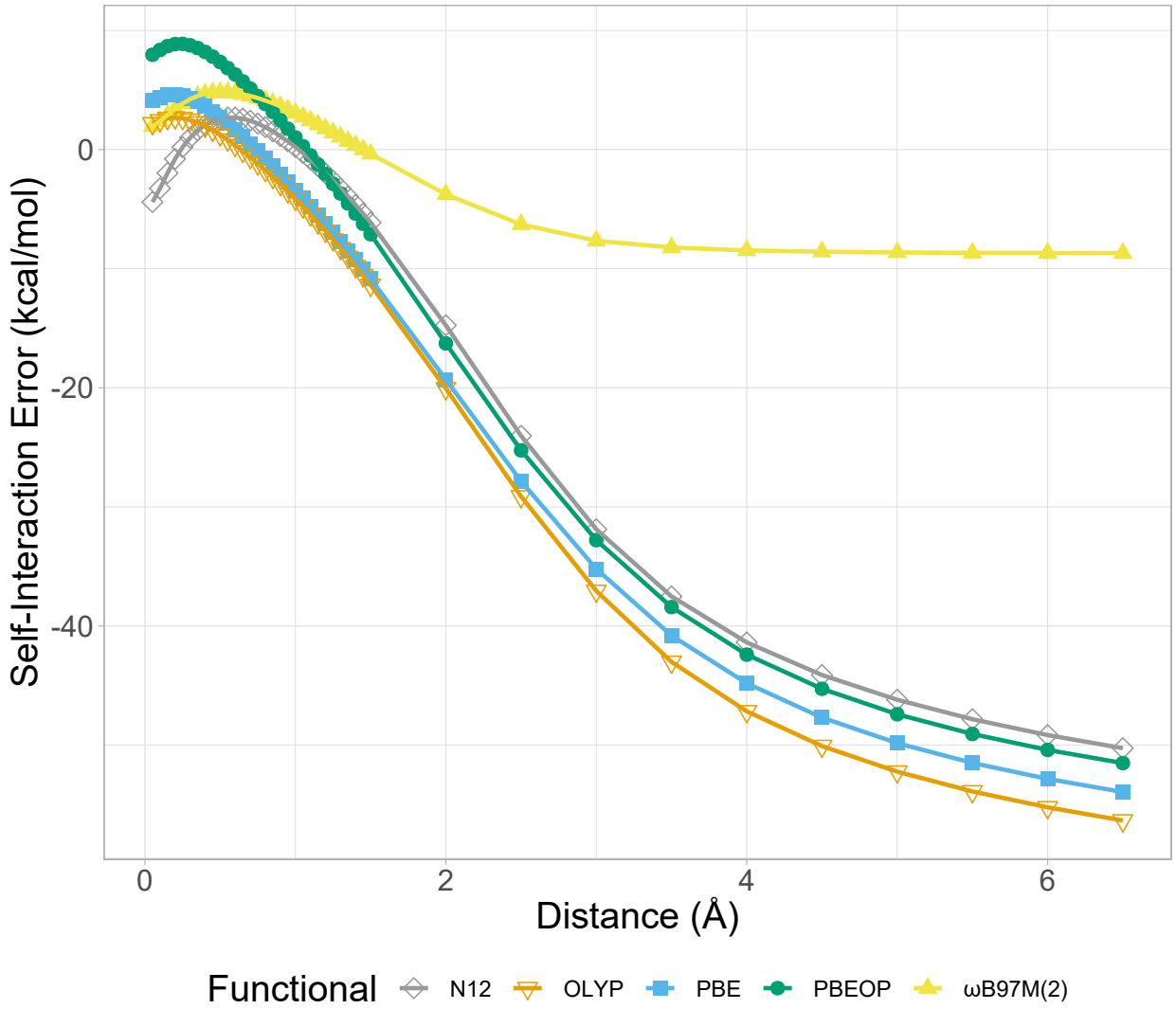

Figure S37: Self-interaction error vs internuclear distance of the H_2^+ molecule for N12, OLYP, PBE, PBEOP with ωB97M(2) as a reference.

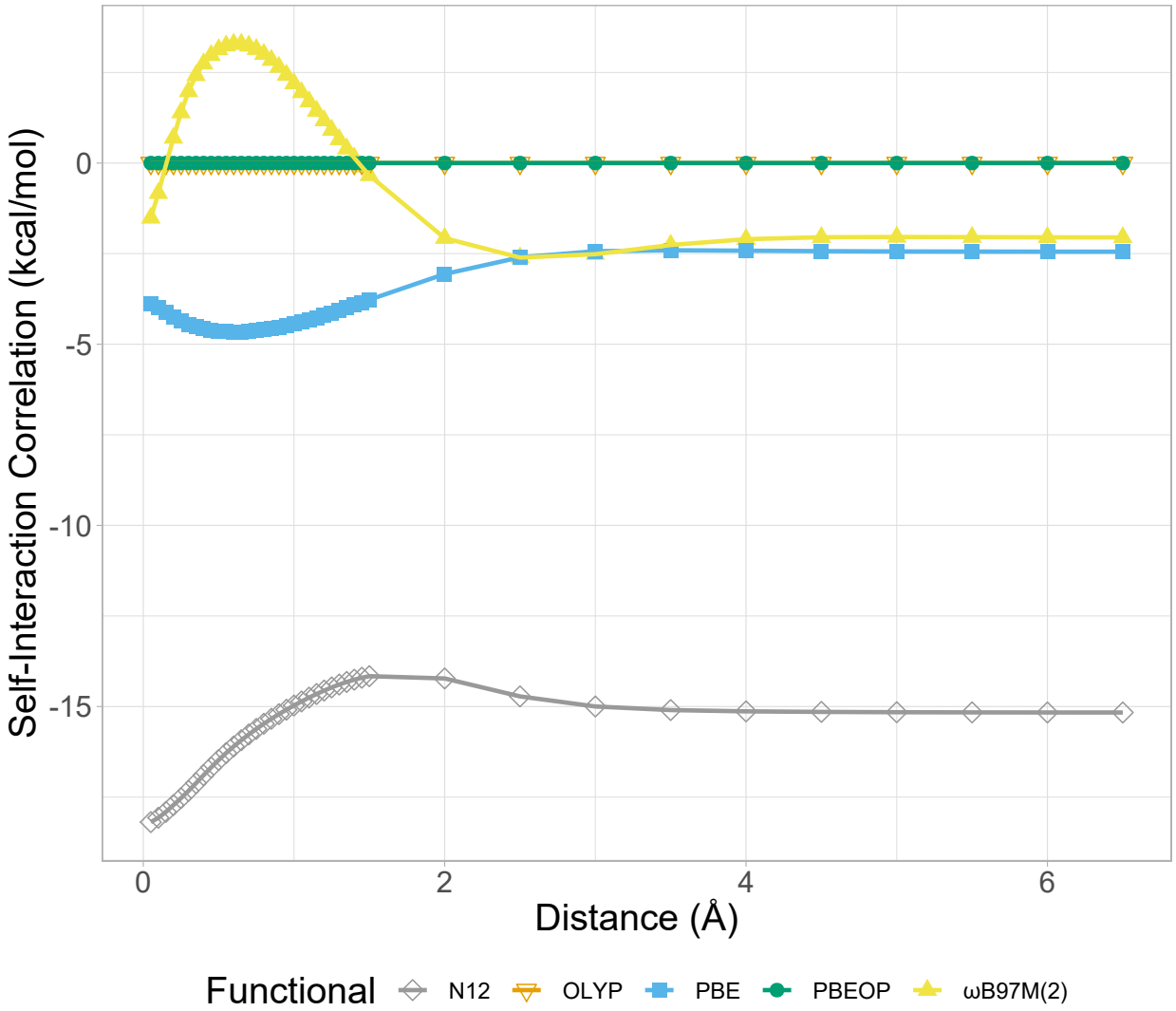

Figure S38: Self-interaction correlation error vs internuclear distance of the H_2^+ molecule for N12, OLYP, PBE, PBEOP with ωB97M(2) as a reference.

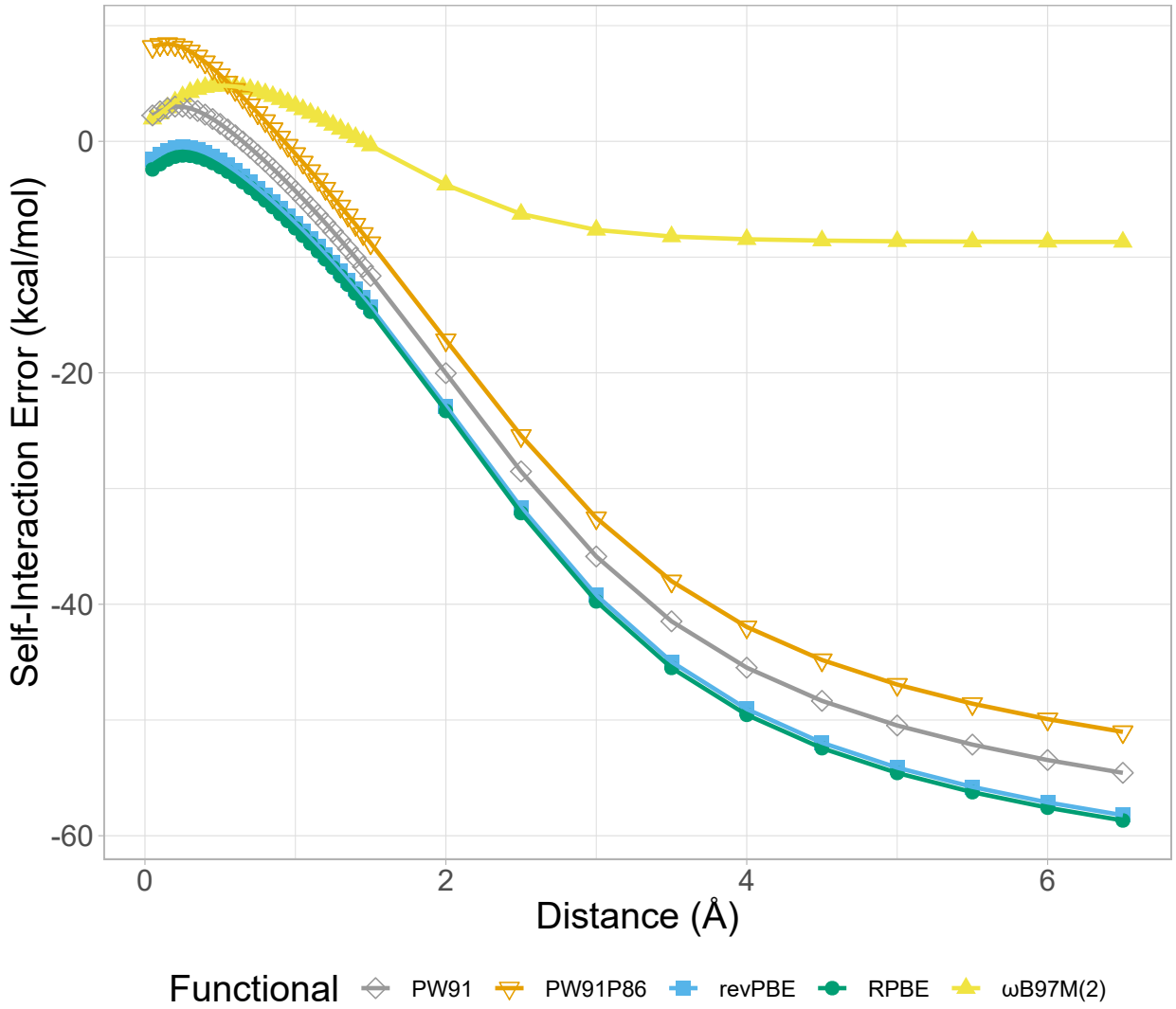

Figure S39: Self-interaction error vs internuclear distance of the H_2^+ molecule for PW91, PW91P86, revPBE, RPBE with ωB97M(2) as a reference.

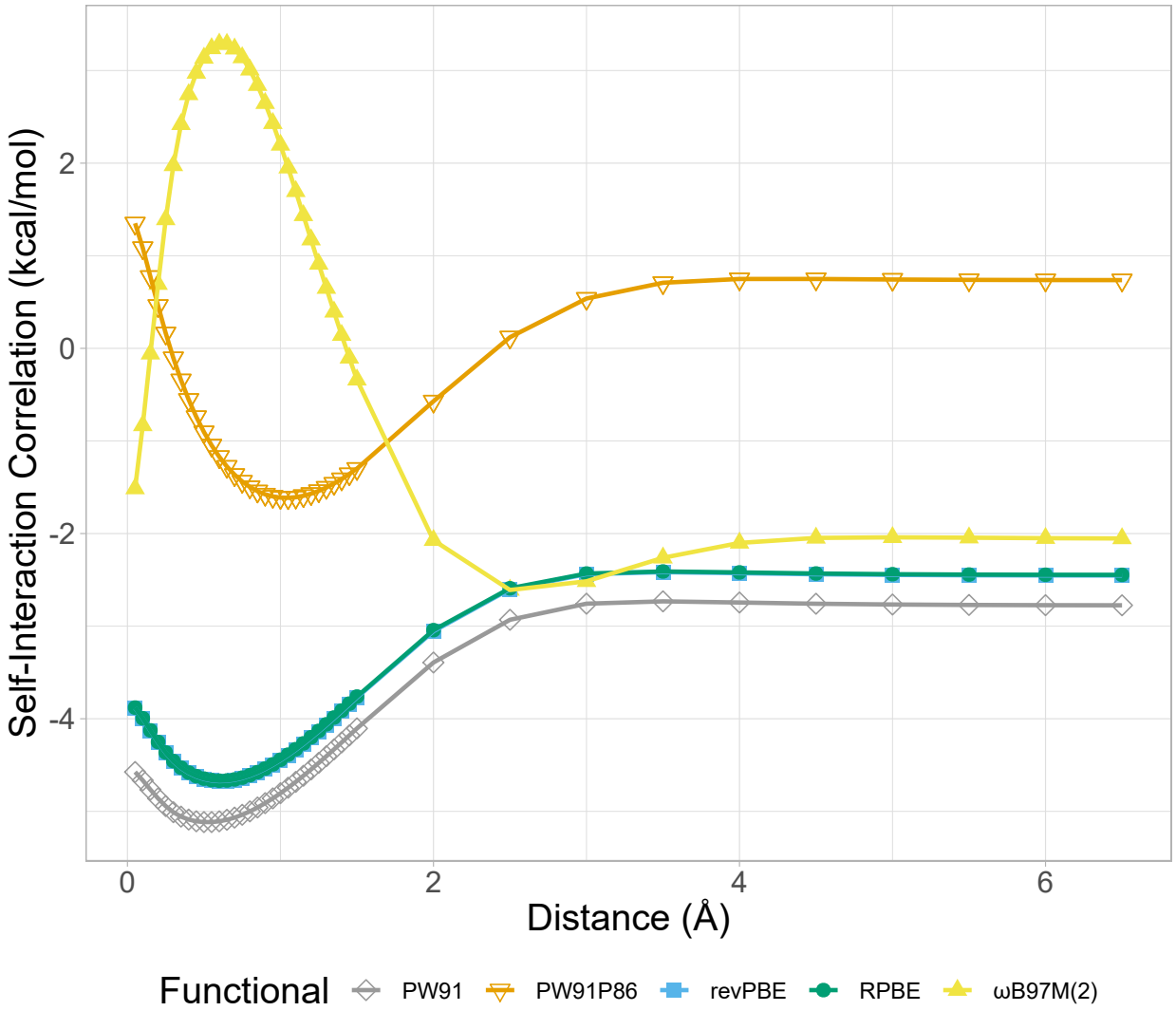

Figure S40: Self-interaction correlation error vs internuclear distance of the H_2^+ molecule for PW91, PW91P86, revPBE, RPBE with ωB97M(2) as a reference.

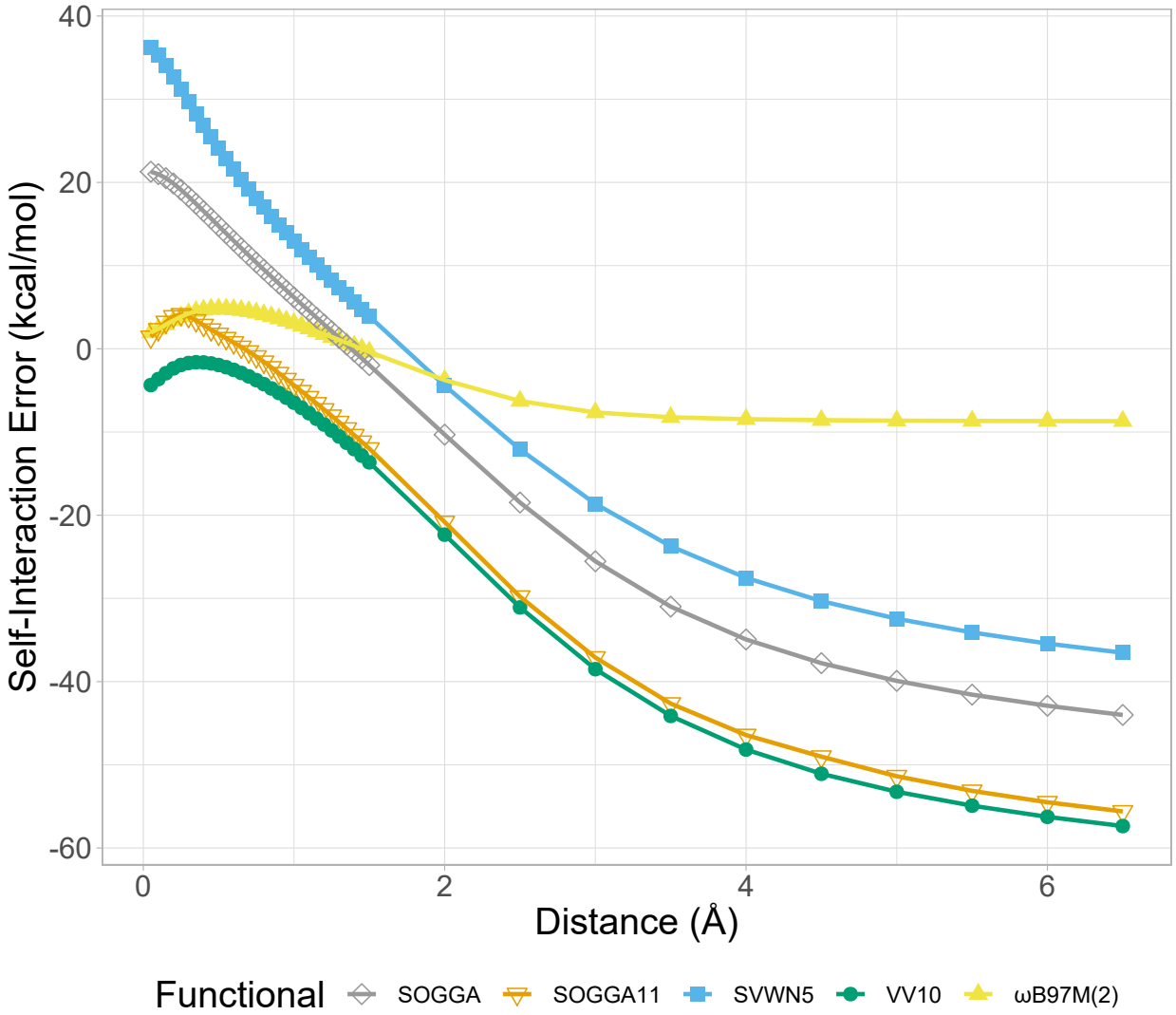

Figure S41: Self-interaction error vs internuclear distance of the H_2^+ molecule for SOGGA, SOGGA11, SVWN5, VV10 with ωB97M(2) as a reference.

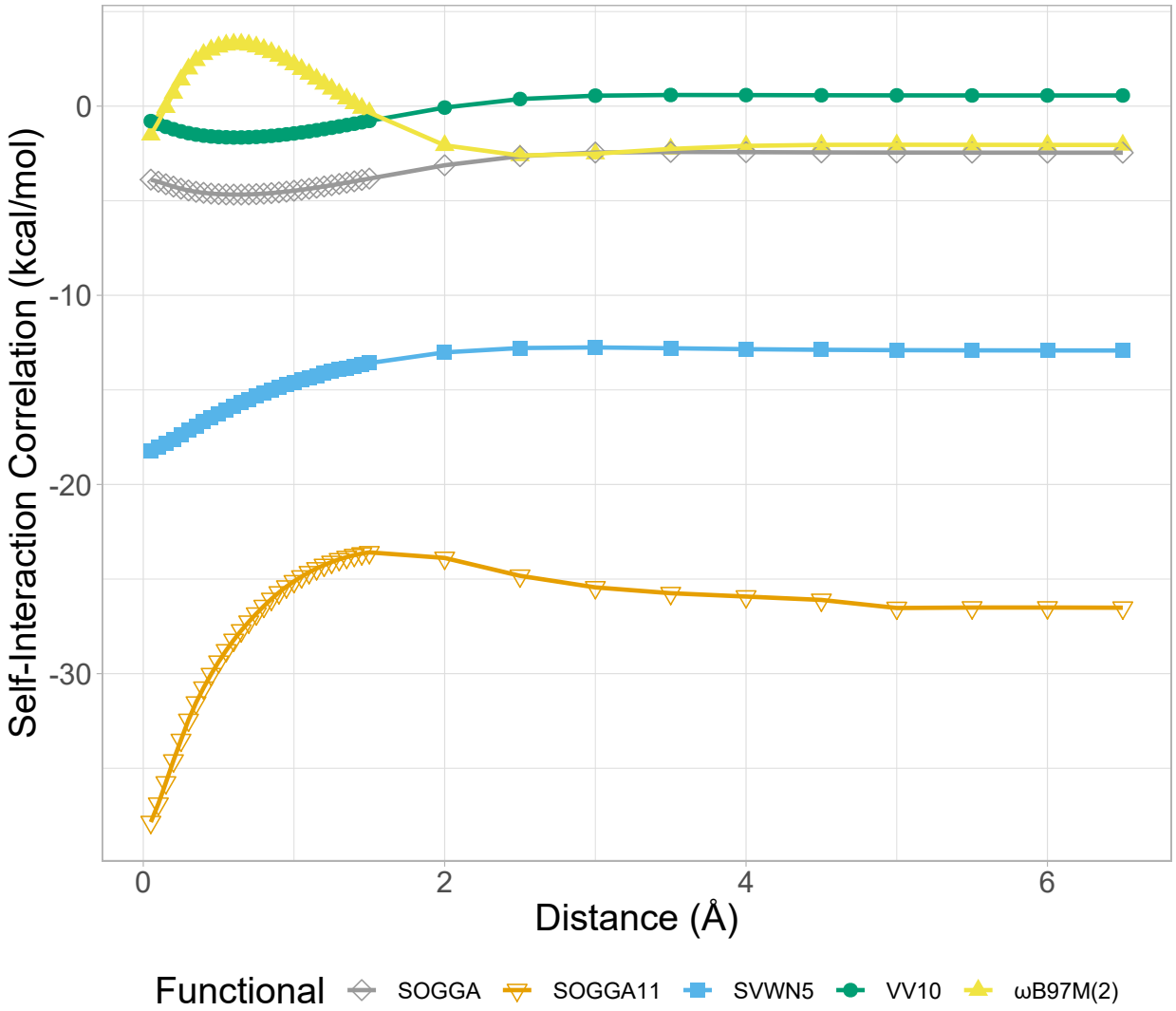

Figure S42: Self-interaction correlation error vs internuclear distance of the H_2^+ molecule for SOGGA, SOGGA11, SVWN5, VV10 with ωB97M(2) as a reference.

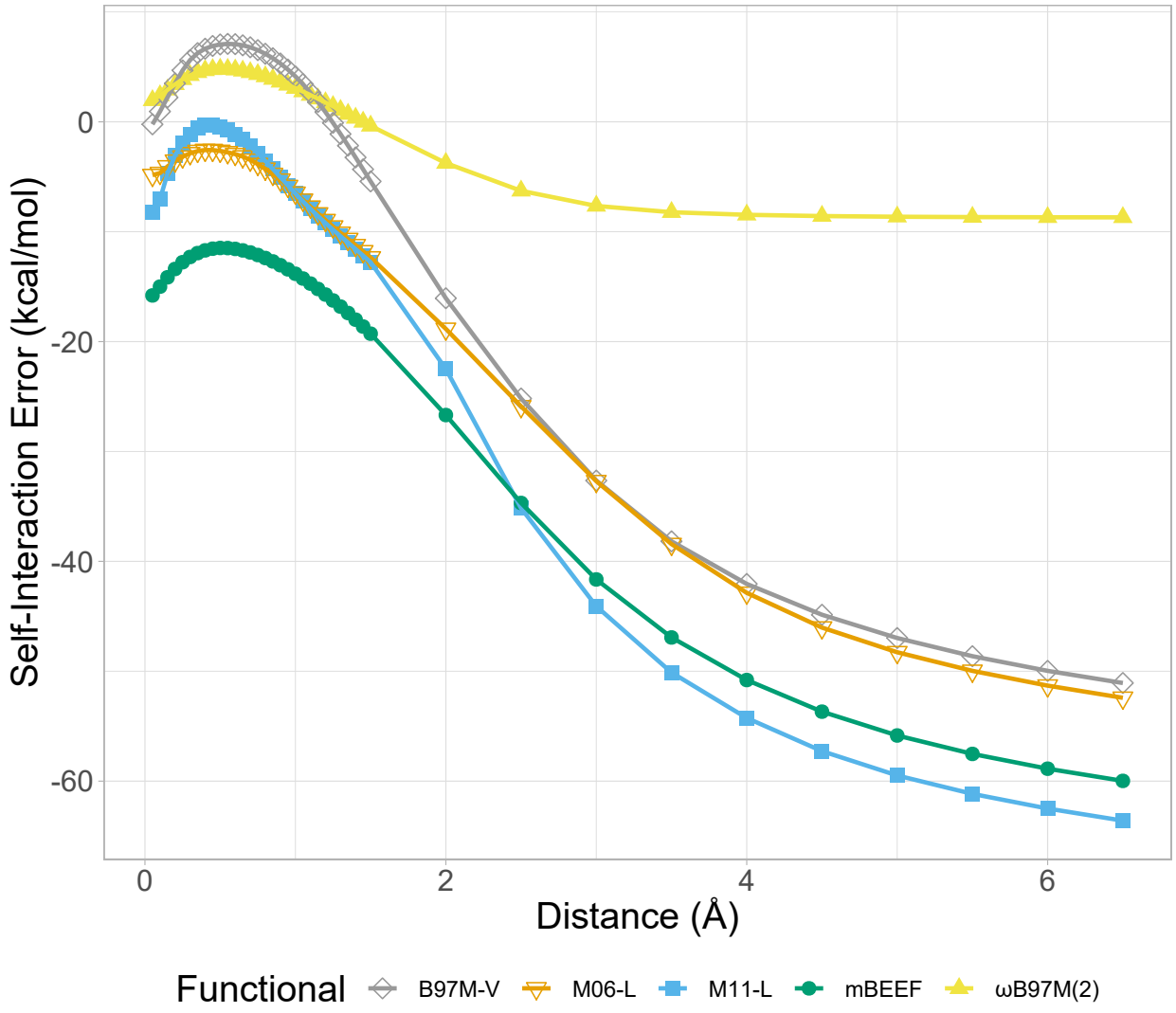

Figure S43: Self-interaction error vs internuclear distance of the H_2^+ molecule for B97M-V, M06-L, M11-L, mBEEF, MN12-L with ωB97M(2) as a reference.

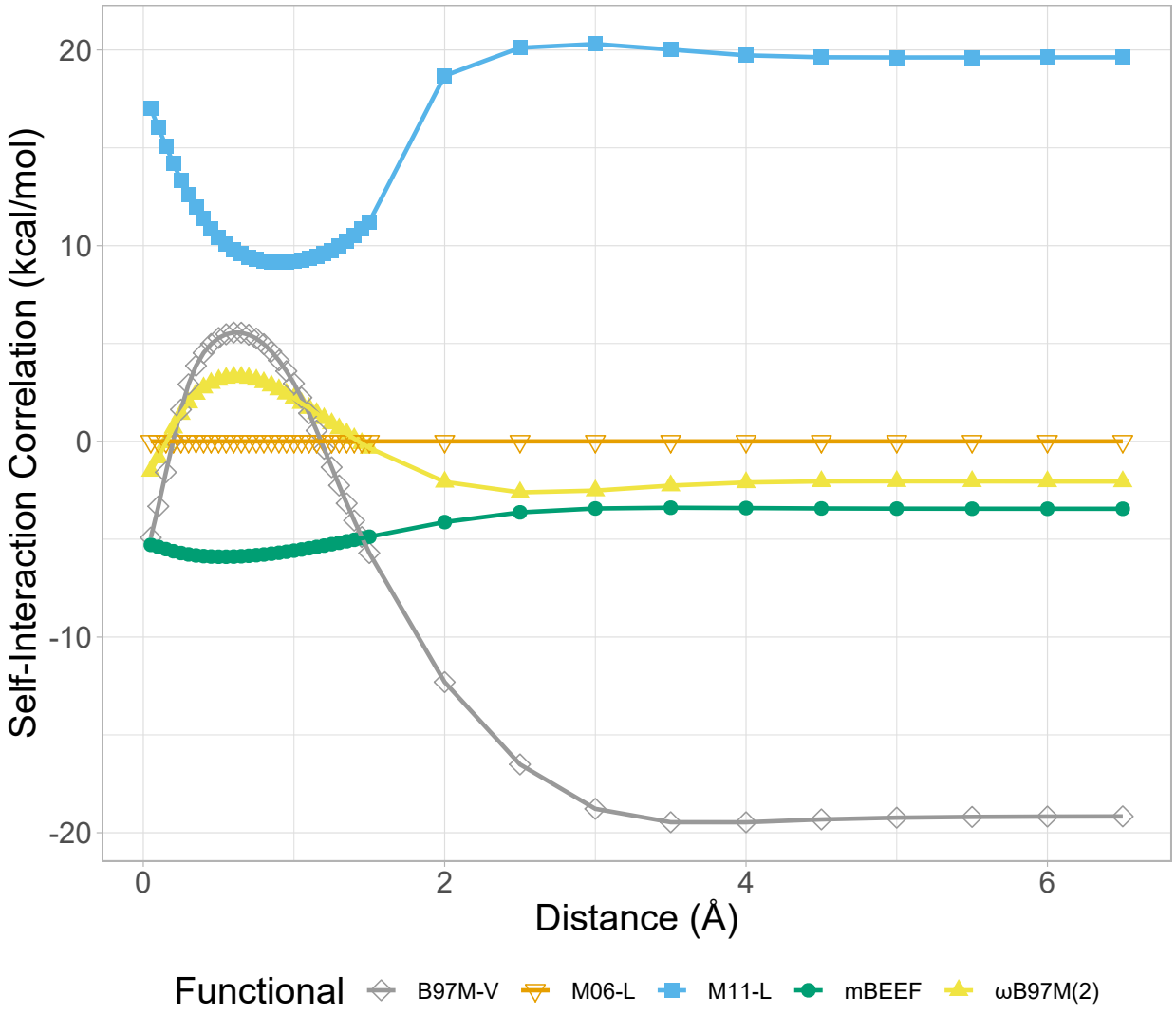

Figure S44: Self-interaction correlation error vs internuclear distance of the H_2^+ molecule for B97M-V, M06-L, M11-L, mBEEF, MN12-L with ωB97M(2) as a reference.

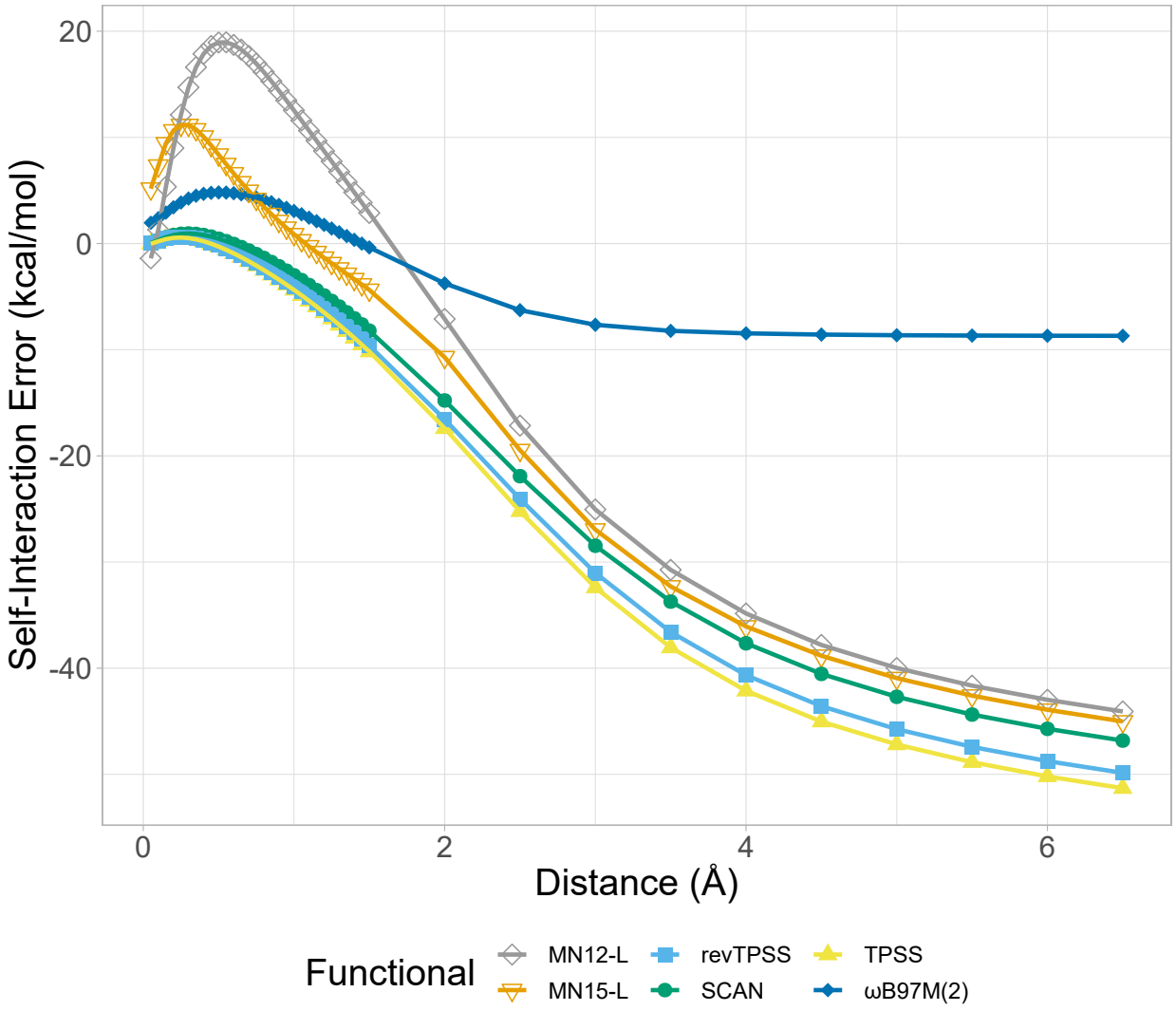

Figure S45: Self-interaction error vs internuclear distance of the H_2^+ molecule for MN15-L, revTPSS, SCAN, TPSS with ωB97M(2) as a reference.

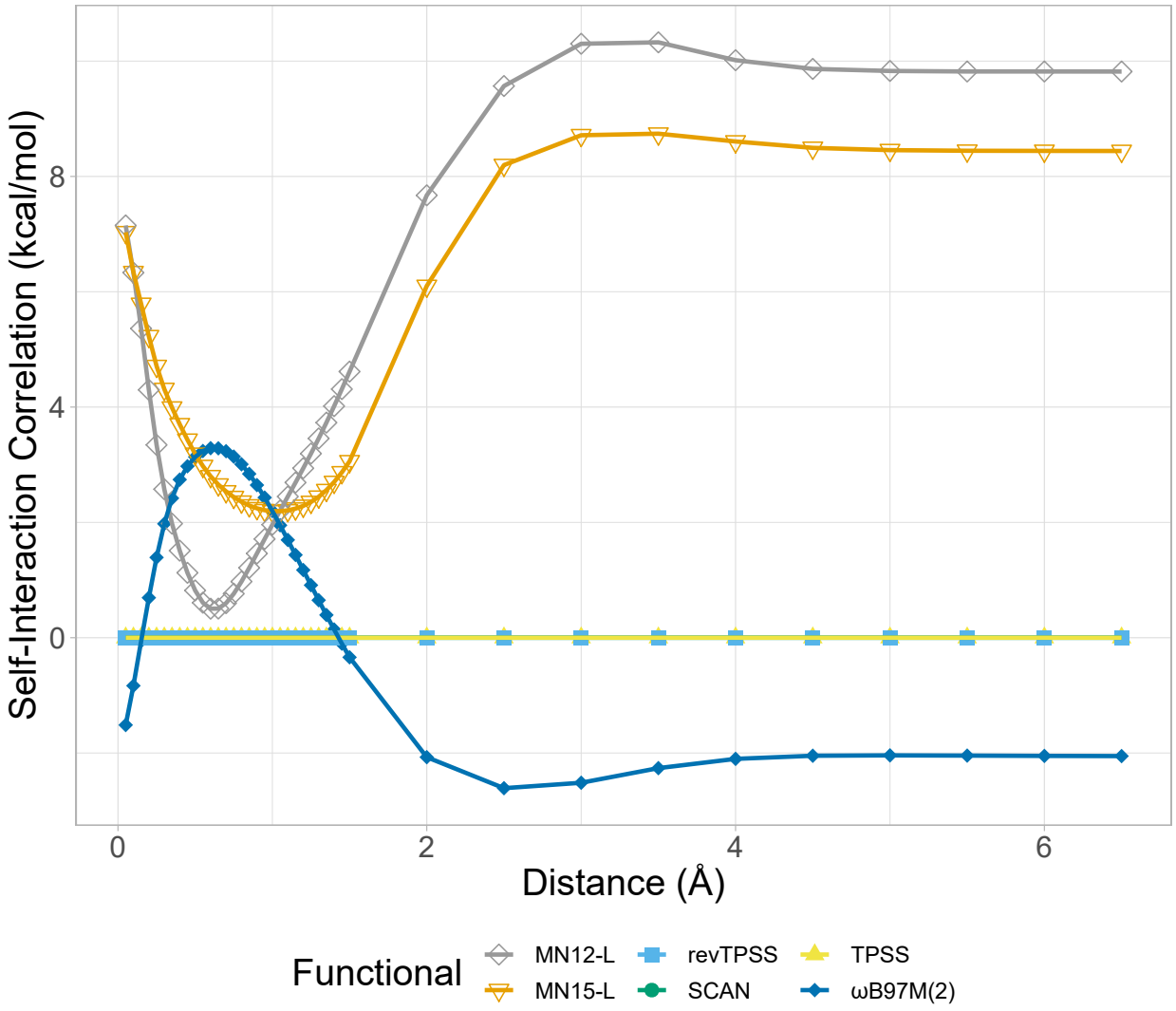

Figure S46: Self-interaction correlation error vs internuclear distance of the H_2^+ molecule for MN15-L, revTPSS, SCAN, TPSS with ωB97M(2) as a reference.

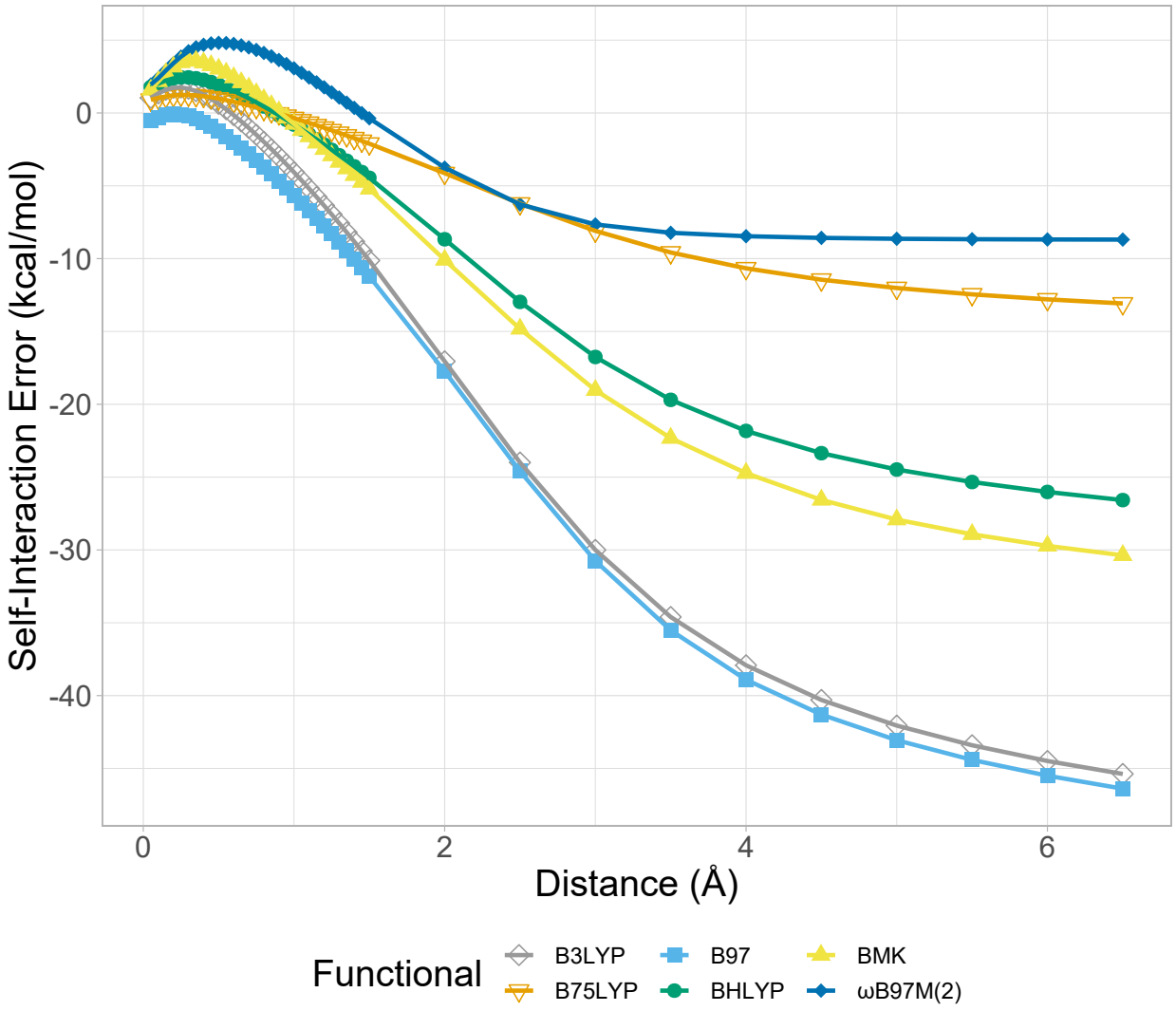

Figure S47: Self-interaction error vs internuclear distance of the H_2^+ molecule for B3LYP, B75LYP, B97, BHLYP, BMK with ωB97M(2) as a reference.

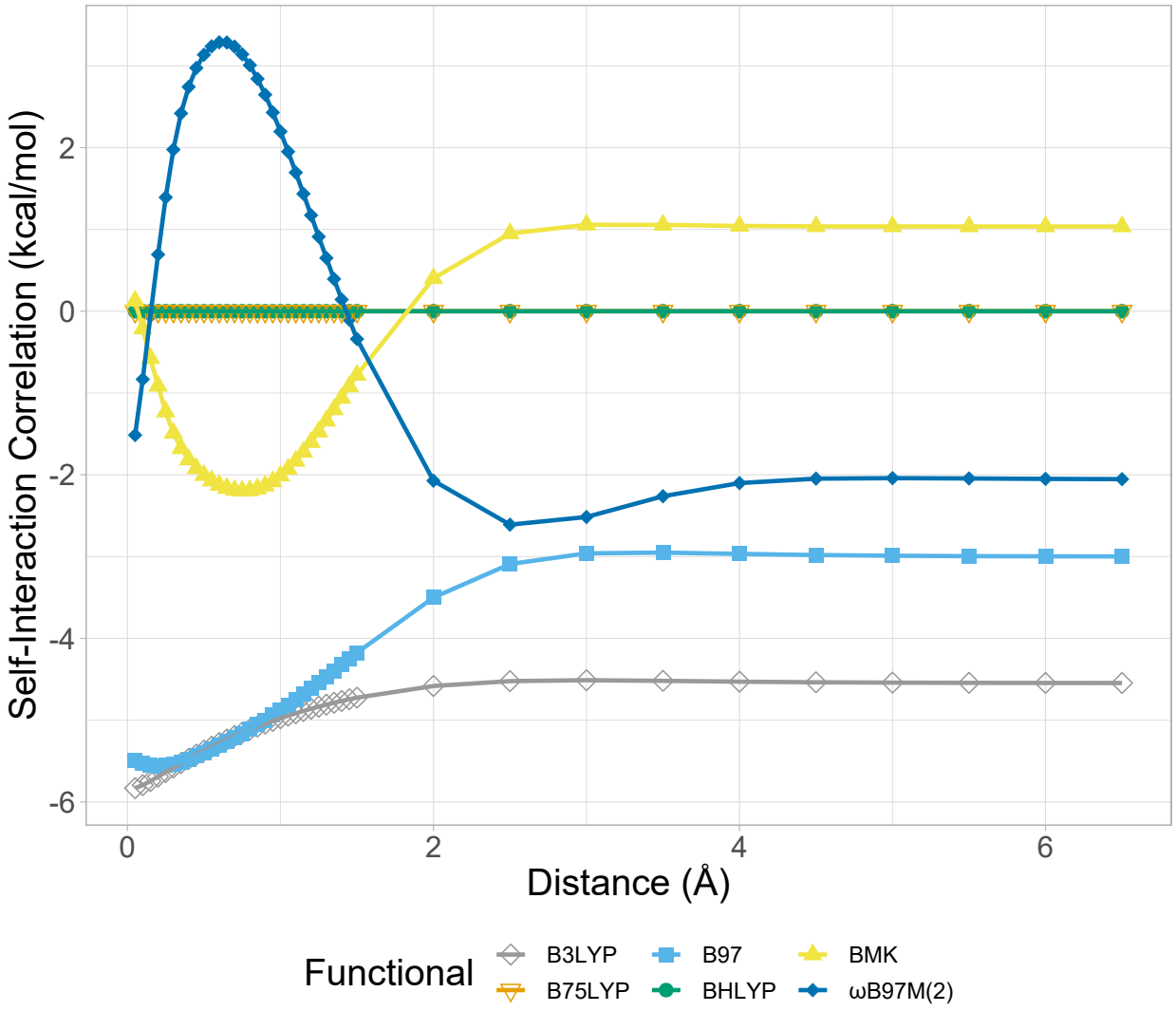

Figure S48: Self-interaction correlation error vs internuclear distance of the H_2^+ molecule for B3LYP, B75LYP, B97, BHLYP, BMK with ωB97M(2) as a reference.

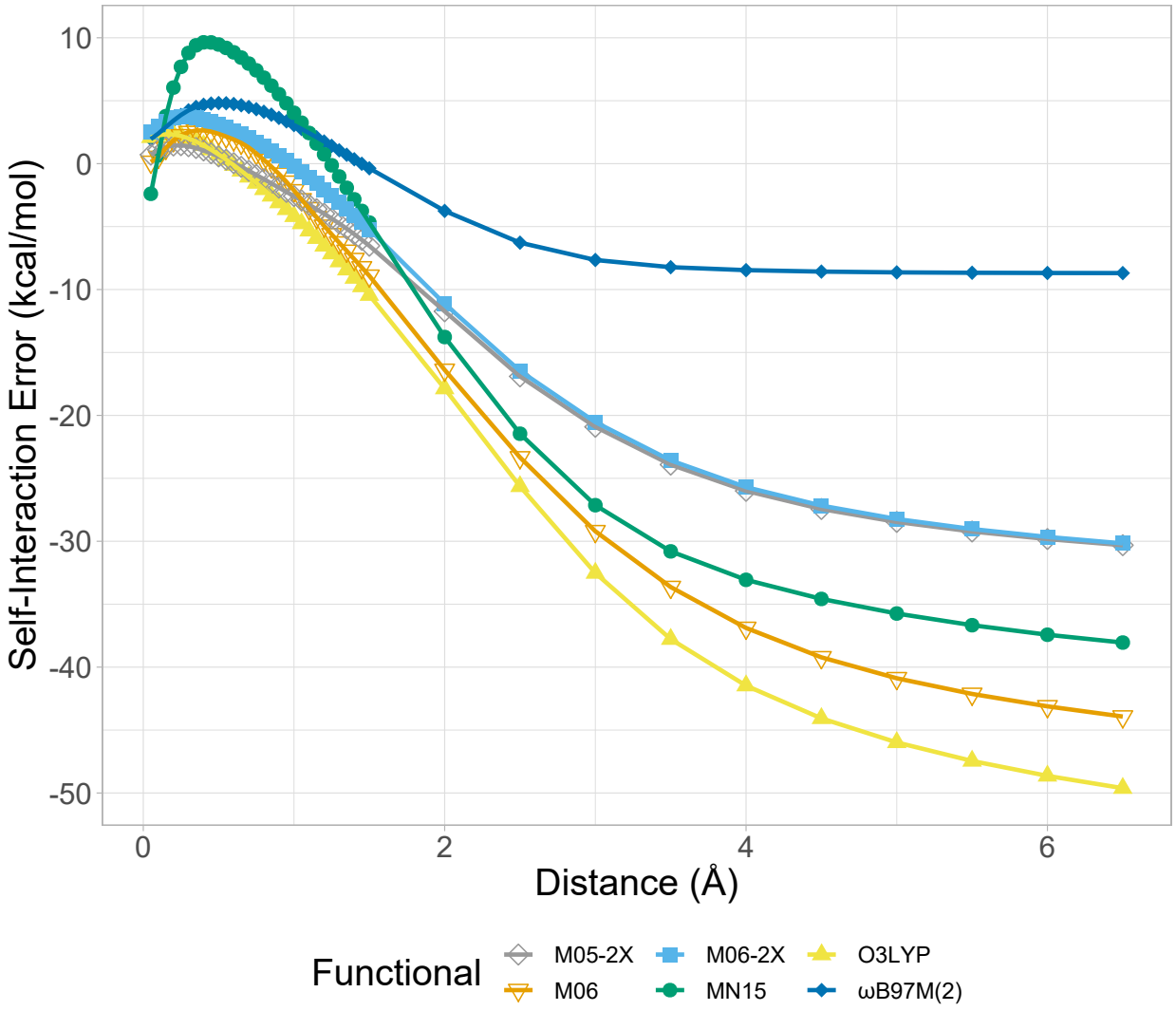

Figure S49: Self-interaction error vs internuclear distance of the H_2^+ molecule for M05-2X, M06, M06-2X, MN15, O3LYP with ωB97M(2) as a reference.

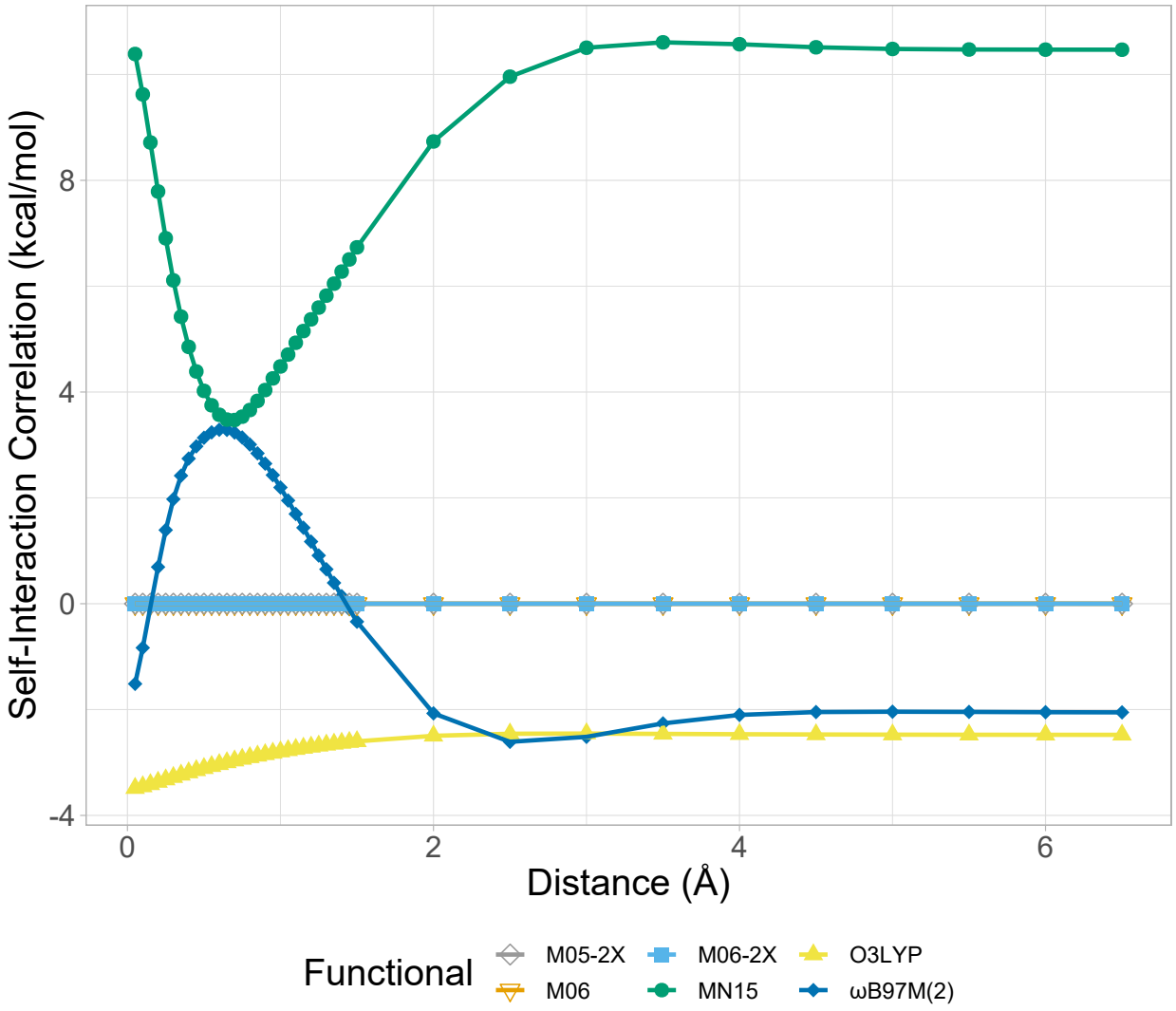

Figure S50: Self-interaction correlation error vs internuclear distance of the H_2^+ molecule for M05-2X, M06, M06-2X, MN15, O3LYP with ωB97M(2) as a reference.

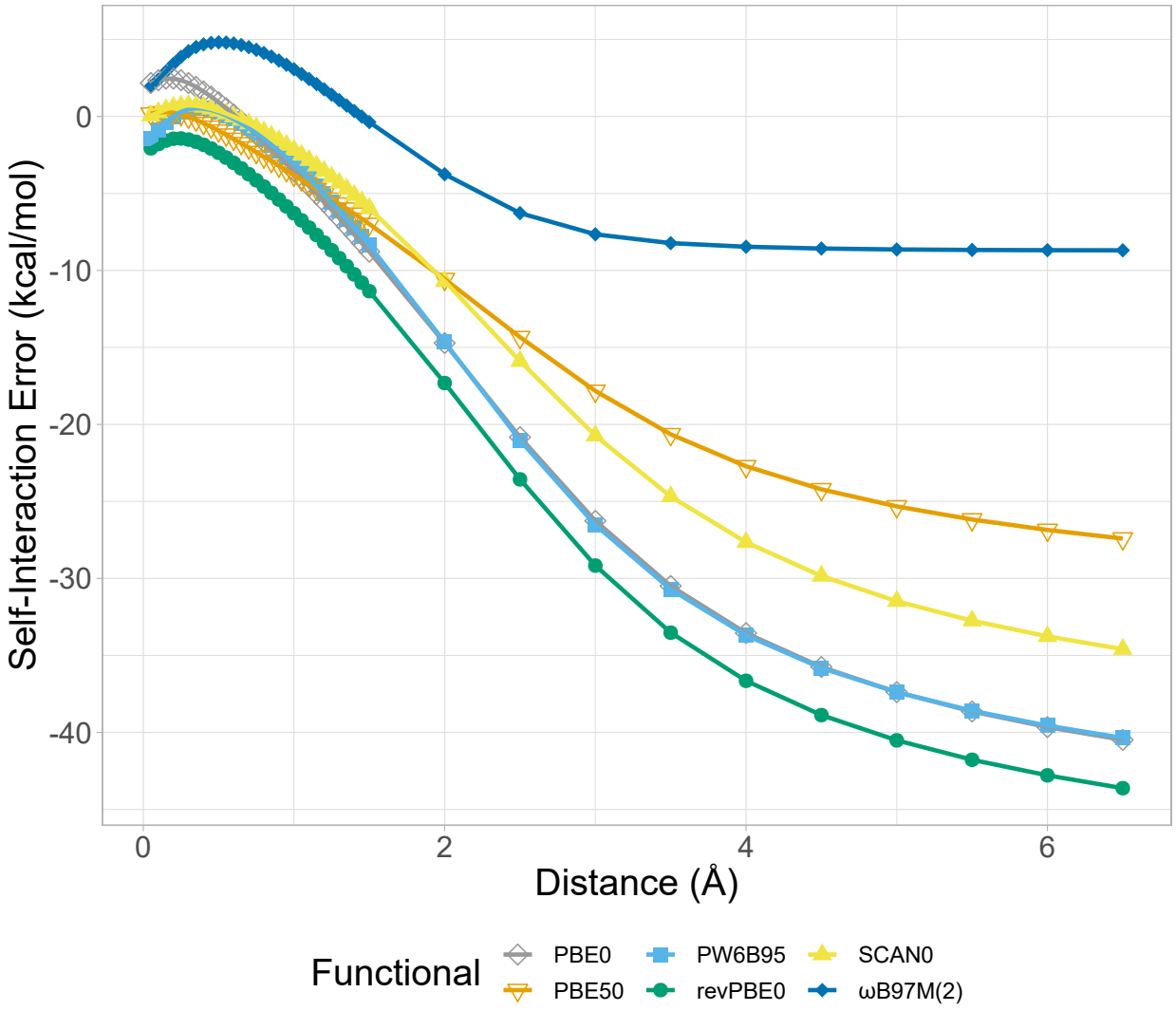

Figure S51: Self-interaction error vs internuclear distance of the H_2^+ molecule for PBE0, PBE50, PW6B95, revPBE0, SCAN0 with ωB97M(2) as a reference.

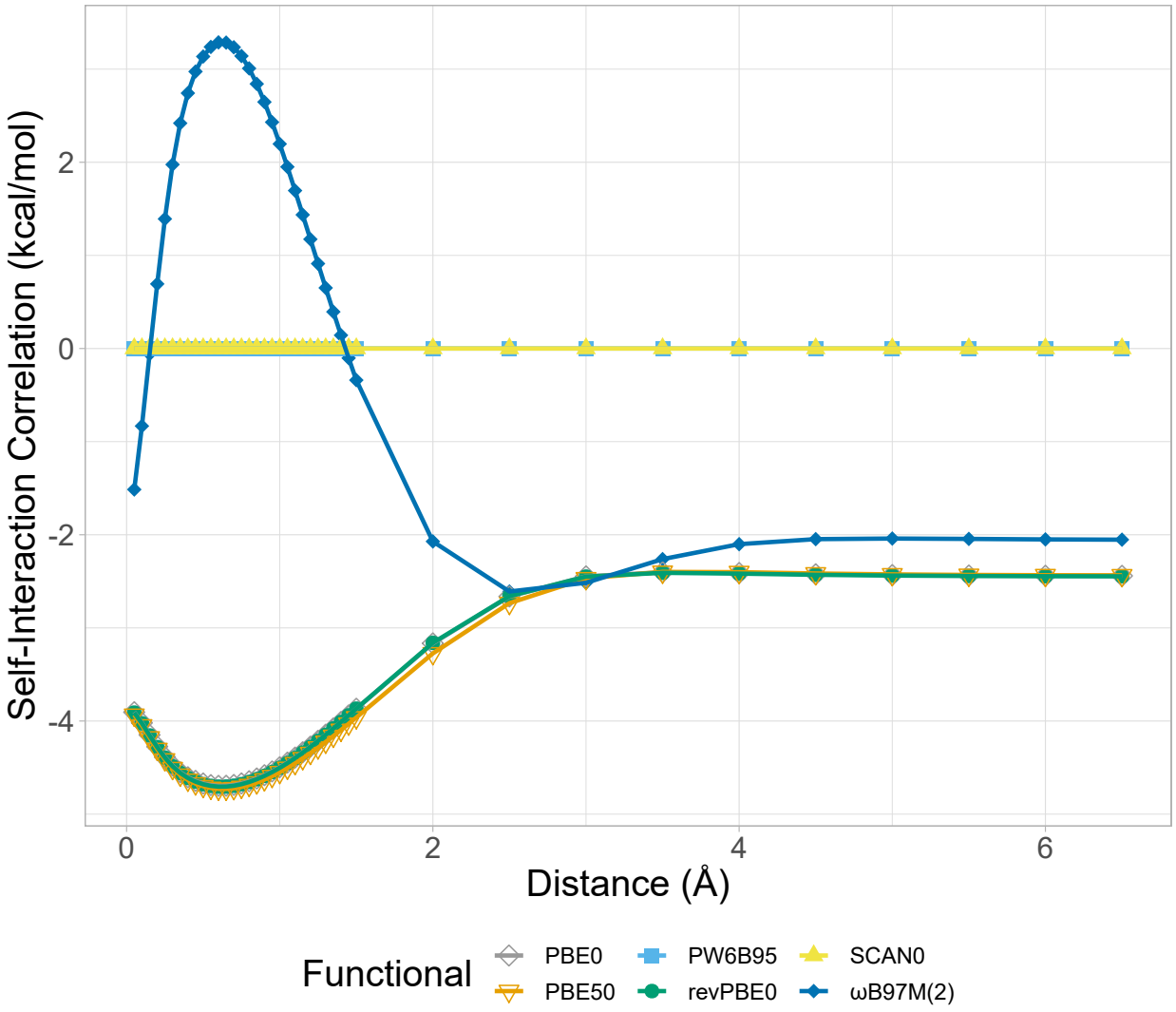

Figure S52: Self-interaction correlation error vs internuclear distance of the H_2^+ molecule for PBE0, PBE50, PW6B95, revPBE0, SCAN0 with ωB97M(2) as a reference.

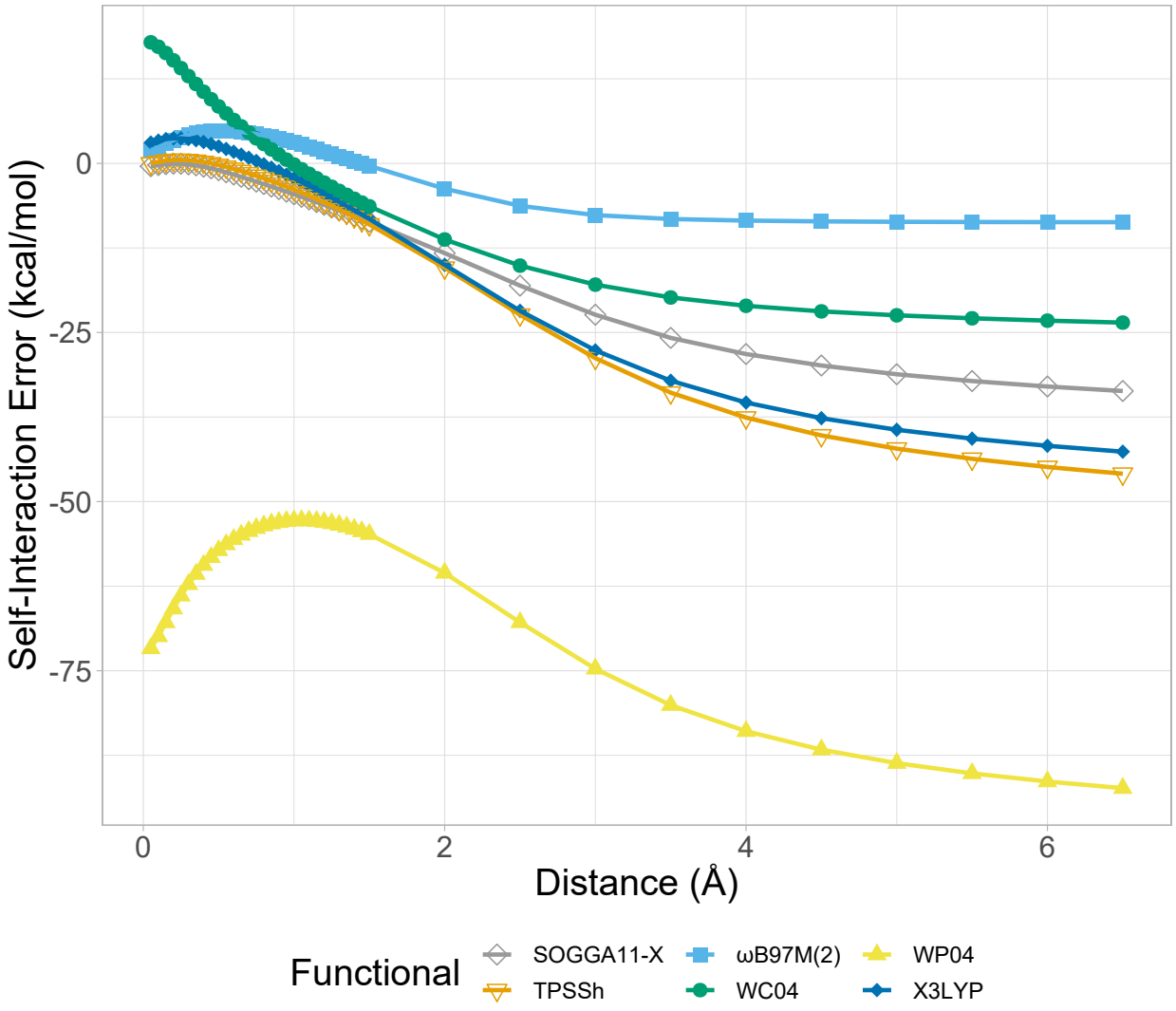

Figure S53: Self-interaction error vs internuclear distance of the H_2^+ molecule for SOGGA11-X, TPSSh, WC04, WP04, X3LYP with ωB97M(2) as a reference.

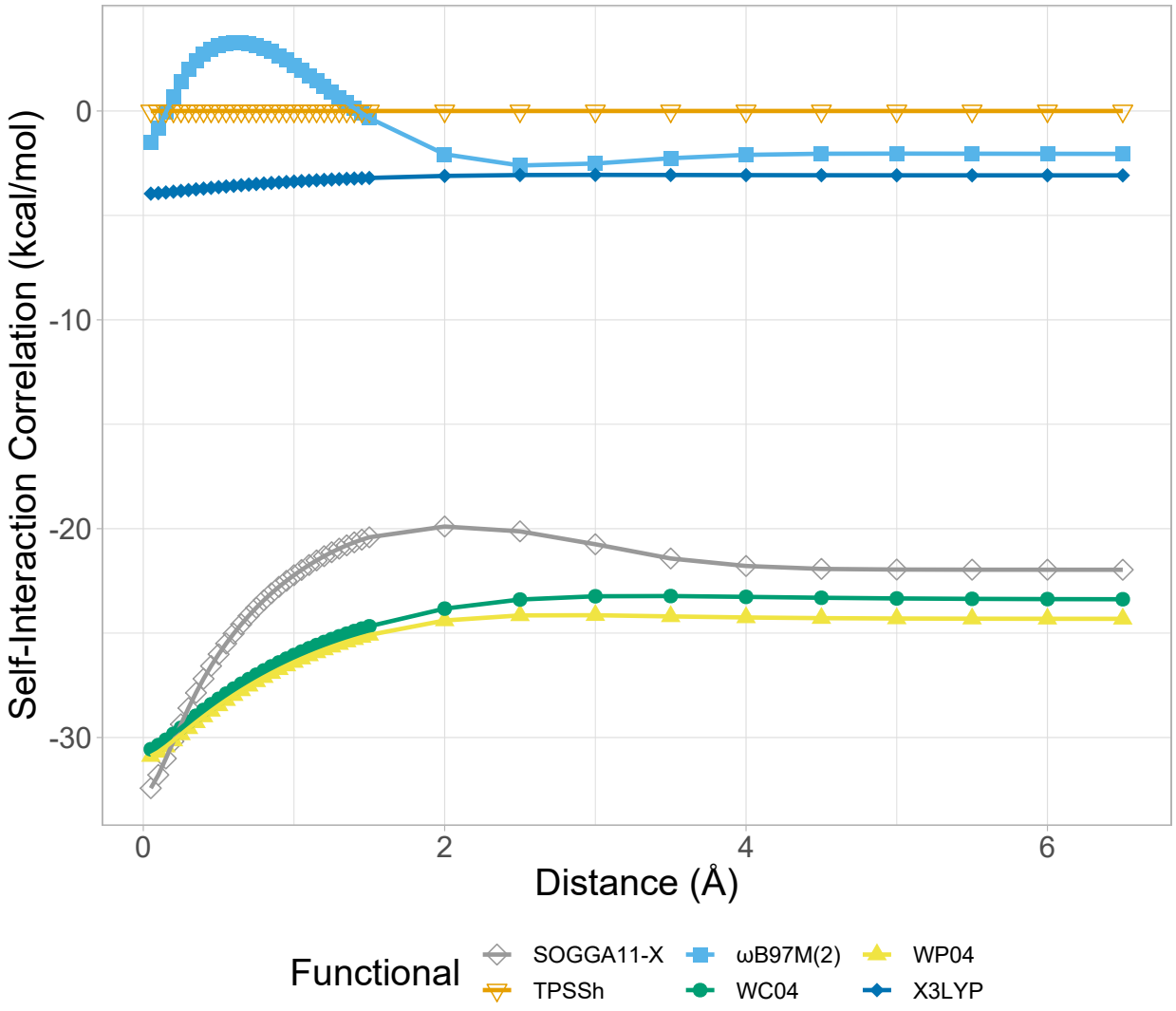

Figure S54: Self-interaction correlation error vs internuclear distance of the H_2^+ molecule for SOGGA11-X, TPSSh, WC04, WP04, X3LYP with ωB97M(2) as a reference.

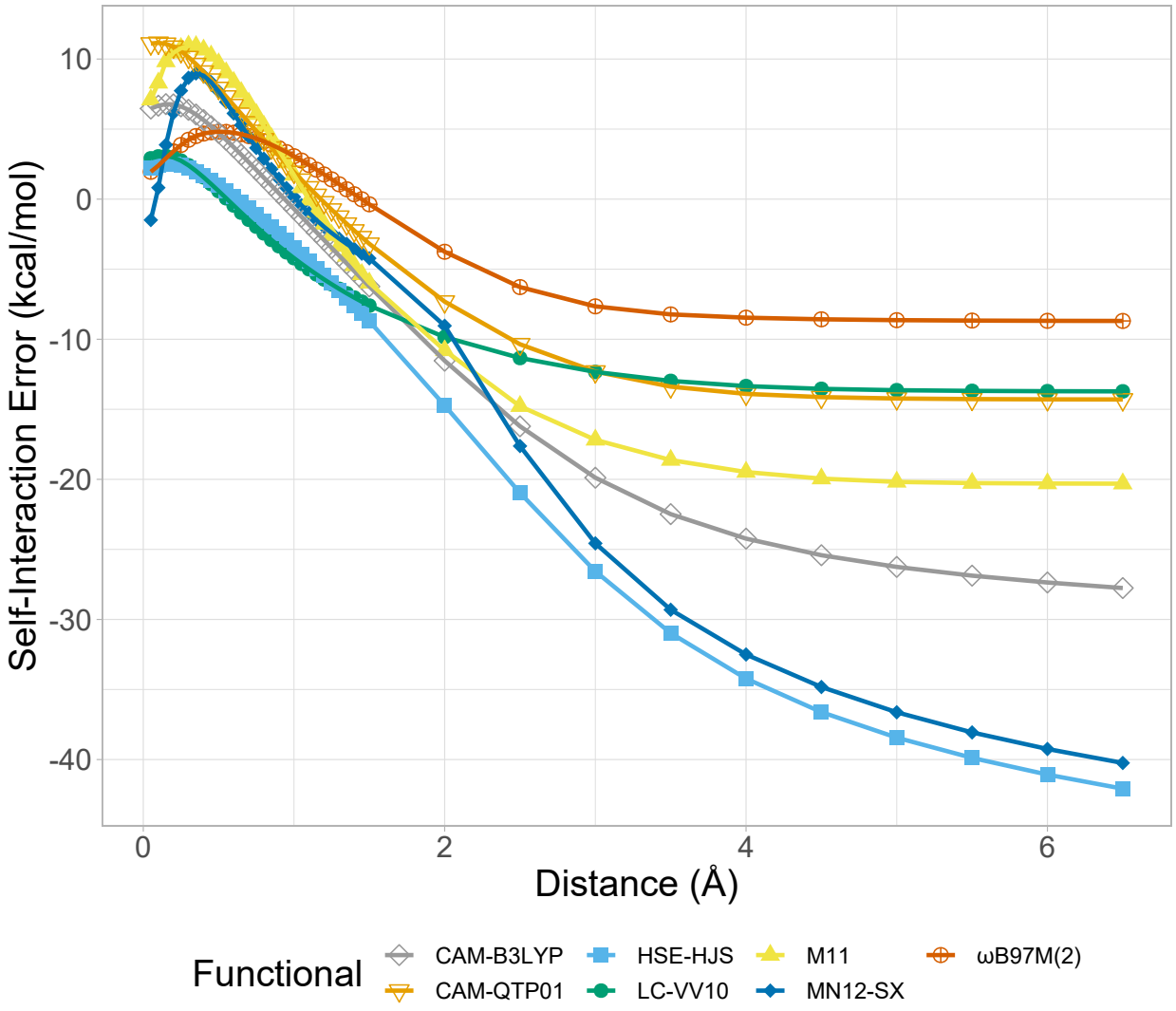

Figure S55: Self-interaction error vs internuclear distance of the H_2^+ molecule for CAM-B3LYP, CAM-QTP01, HSE-HJS, LC-VV10, M11, MN12-SX with ωB97M(2) as a reference.

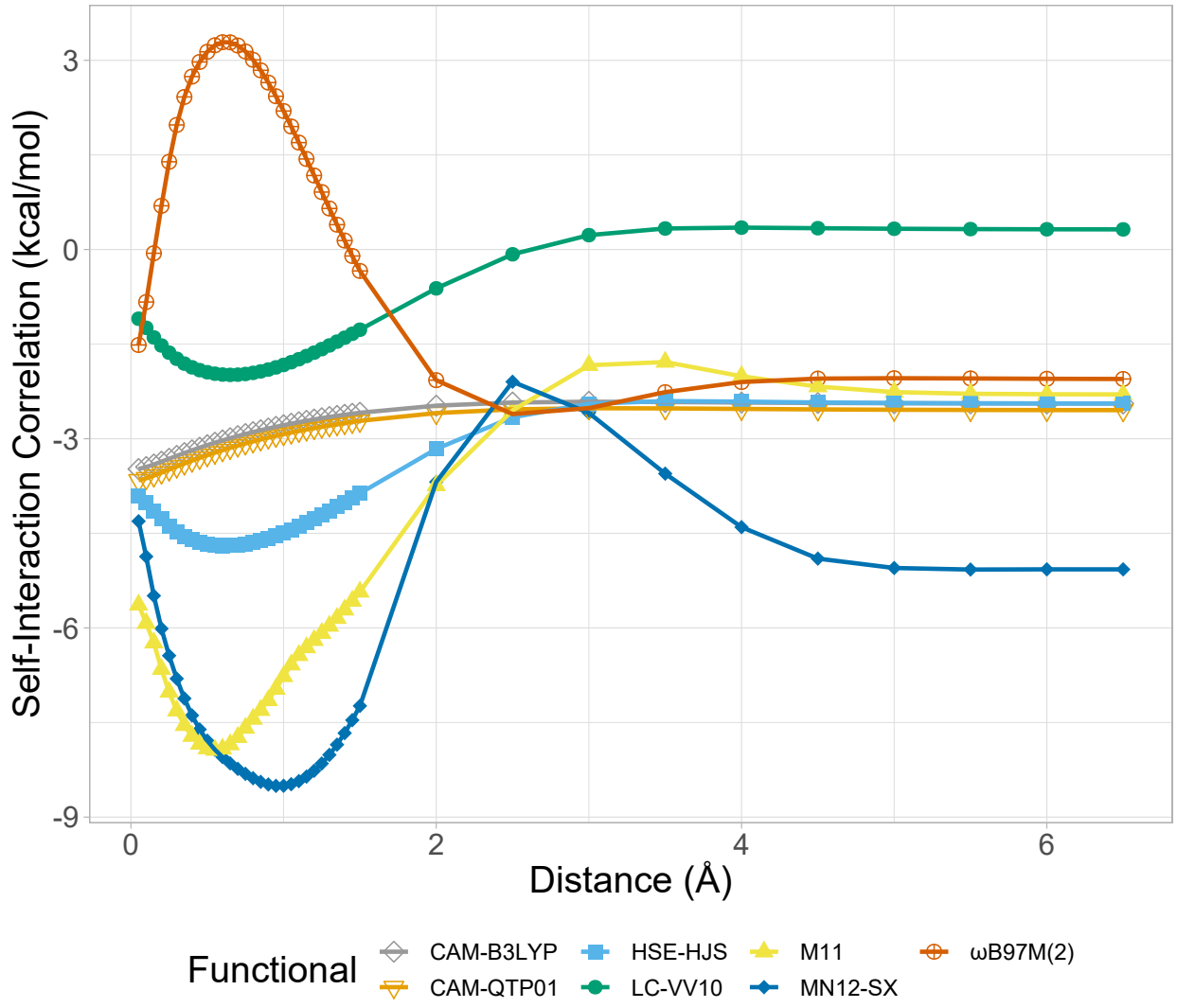

Figure S56: Self-interaction correlation error vs internuclear distance of the H_2^+ molecule for CAM-B3LYP, CAM-QTP01, HSE-HJS, LC-VV10, M11, MN12-SX with ωB97M(2) as a reference.

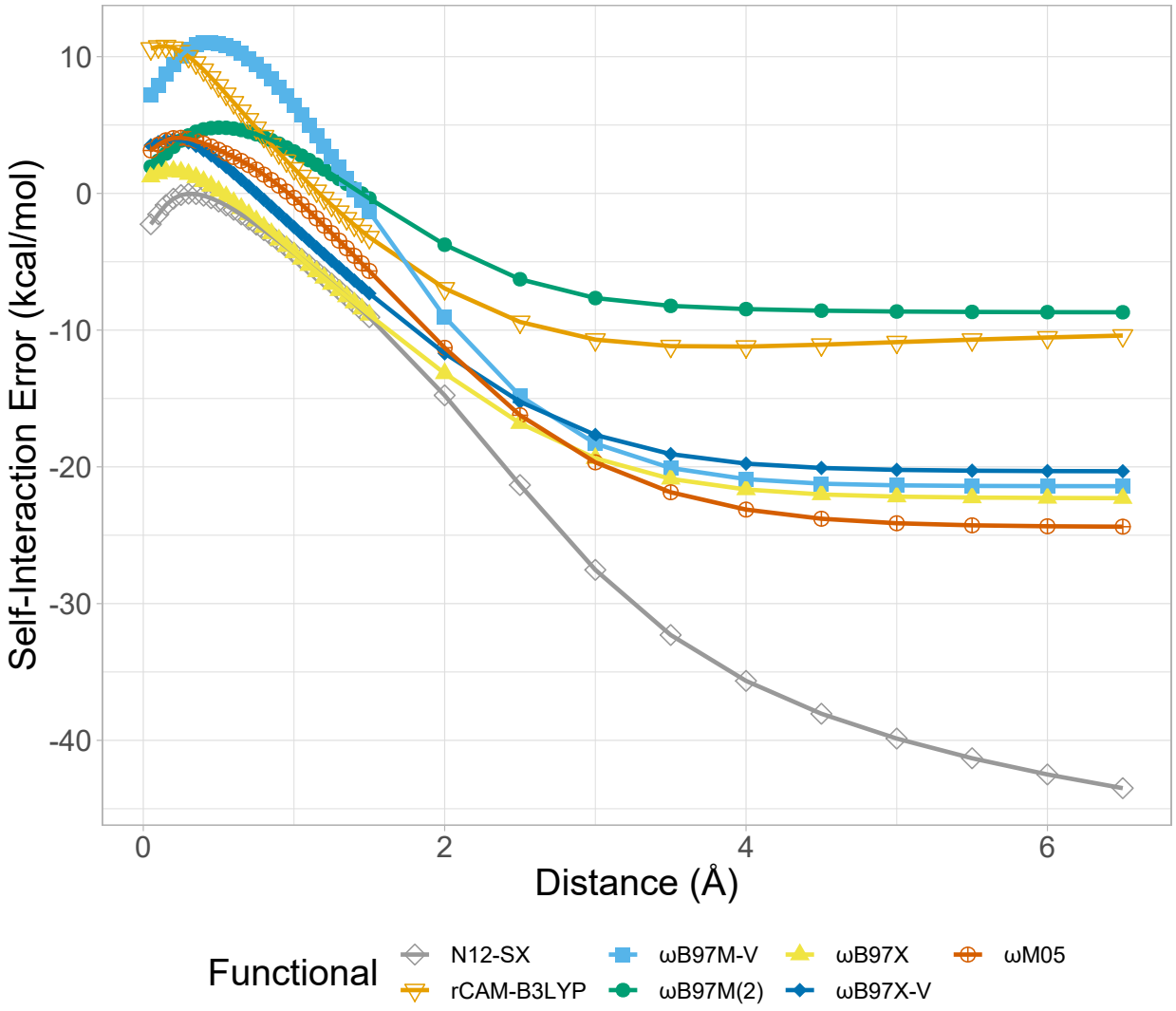

Figure S57: Self-interaction error vs internuclear distance of the H_2^+ molecule for N12-SX, rCAM-B3LYP, ωB97M-V, ωB97X, ωB97X-V, ωM05 with ωB97M(2) as a reference.

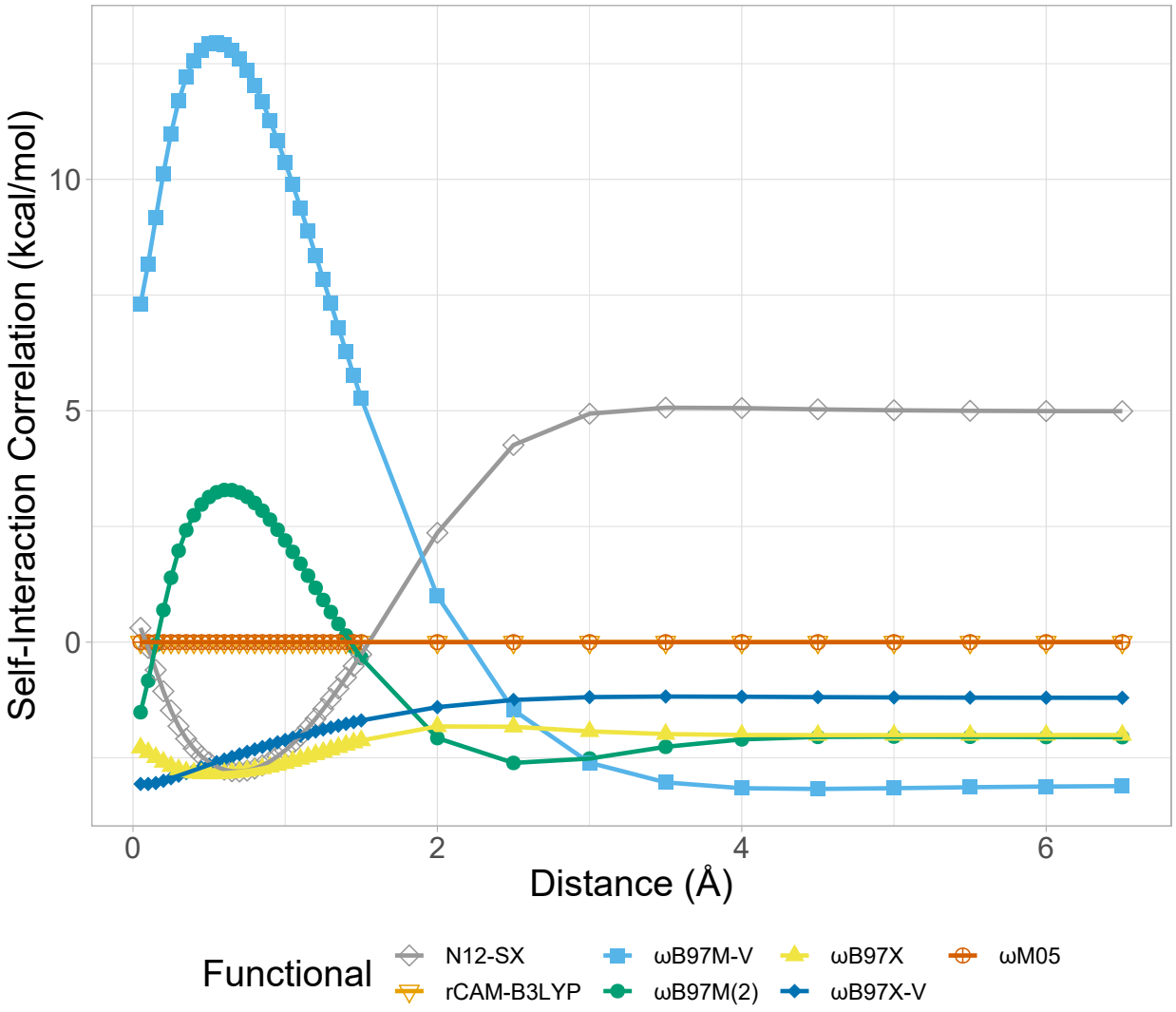

Figure S58: Self-interaction correlation error vs internuclear distance of the H_2^+ molecule for N12-SX, rCAM-B3LYP, ωB97M-V, ωB97X, ωB97X-V, ωM05 with ωB97M(2) as a reference.

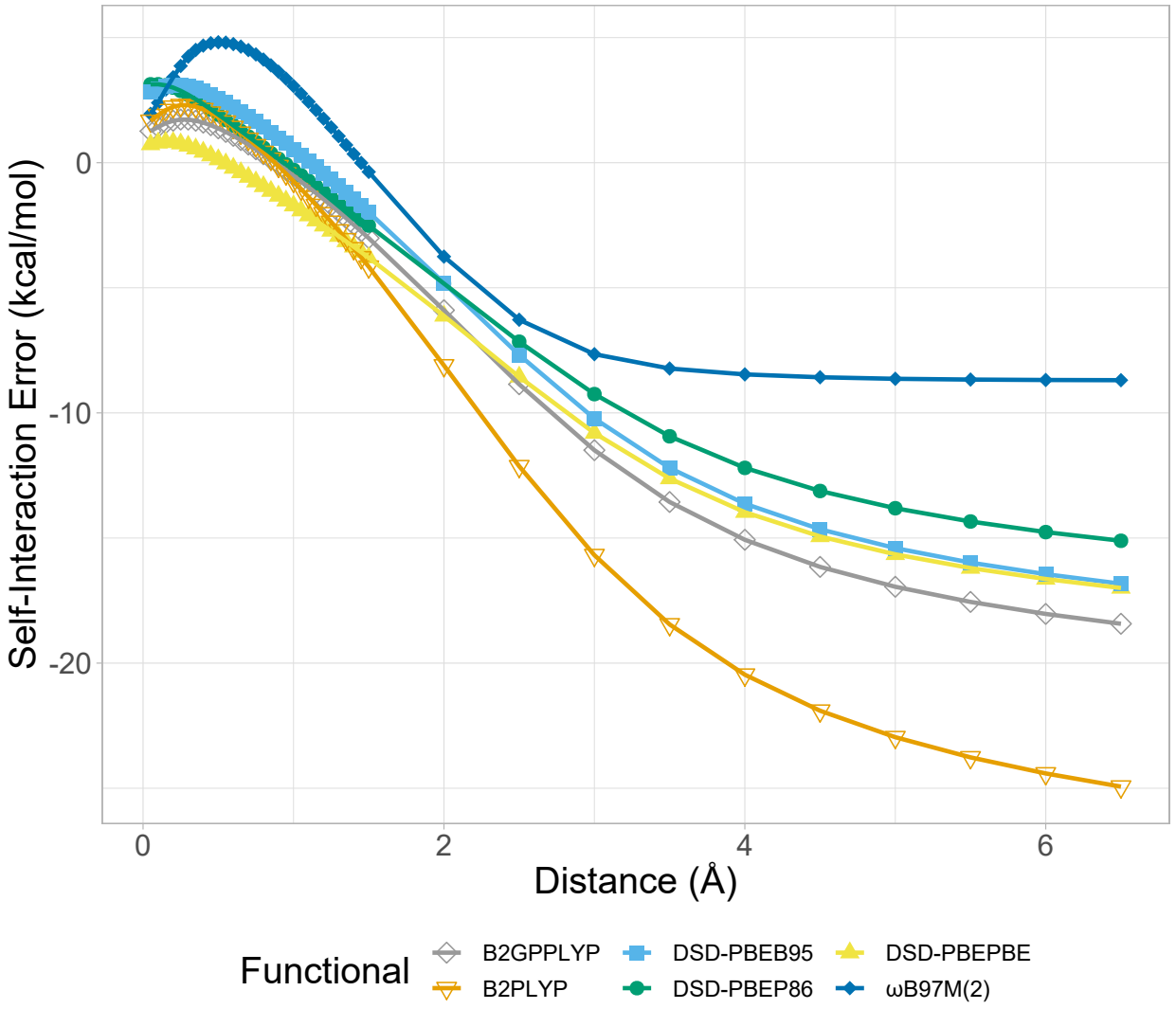

Figure S59: Self-interaction error vs internuclear distance of the H_2^+ molecule for B2GPPLYP, B2PLYP, DSD-PBEB95, DSD-PBEP86, DSD-PBEPBE with ωB97M(2) as a reference.

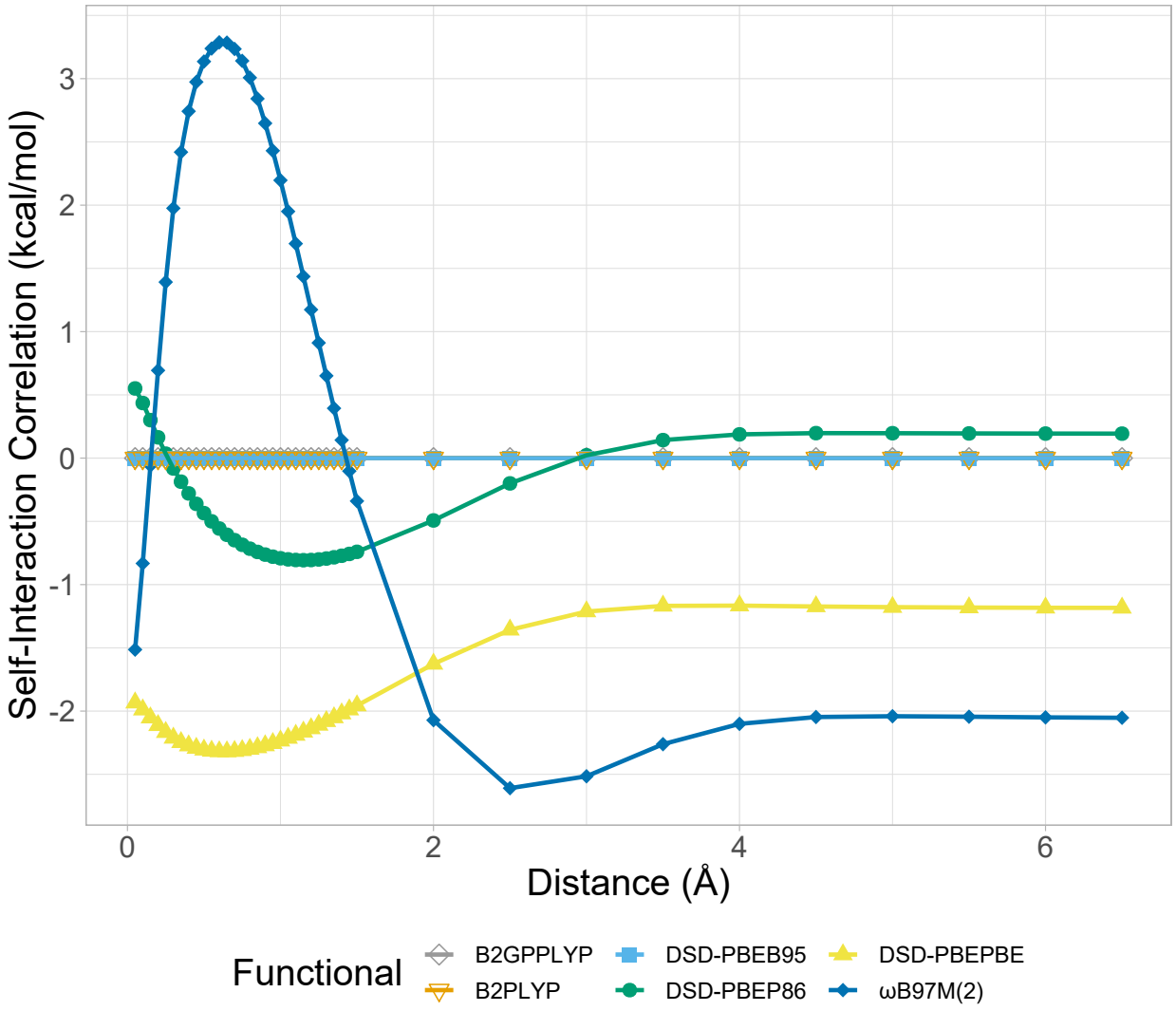

Figure S60: Self-interaction correlation error vs internuclear distance of the H_2^+ molecule for B2GPPLYP, B2PLYP, DSD-PBEB95, DSD-PBEP86, DSD-PBEPBE with ωB97M(2) as a reference.

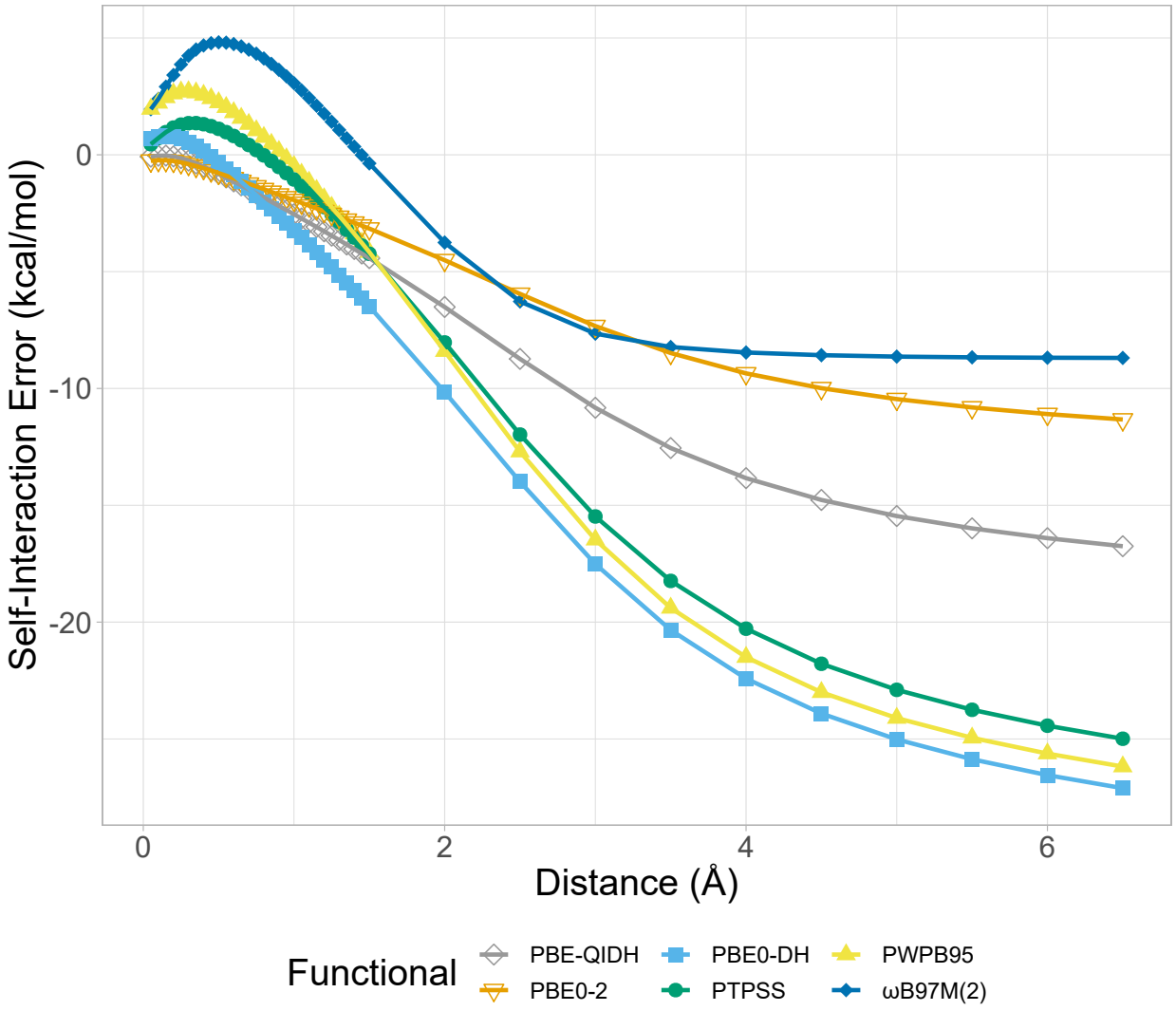

Figure S61: Self-interaction error vs internuclear distance of the H_2^+ molecule for PBE-QIDH, PBE0-2, PBE0-DH, PTPSS, PWPB95 with ωB97M(2) as a reference.

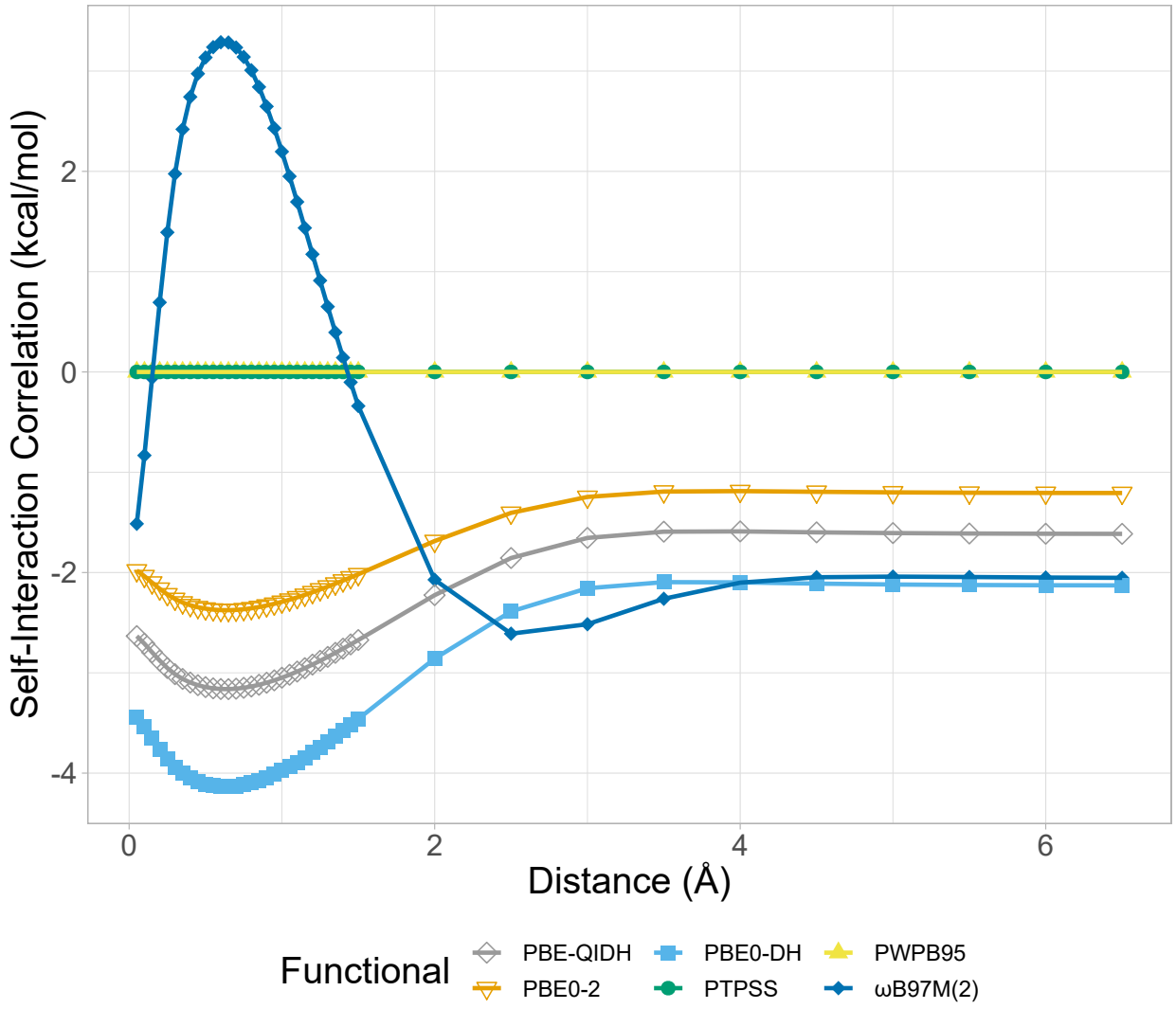

Figure S62: Self-interaction correlation error vs internuclear distance of the H_2^+ molecule for PBE-QIDH, PBE0-2, PBE0-DH, PTPSS, PWPB95 with ωB97M(2) as a reference.

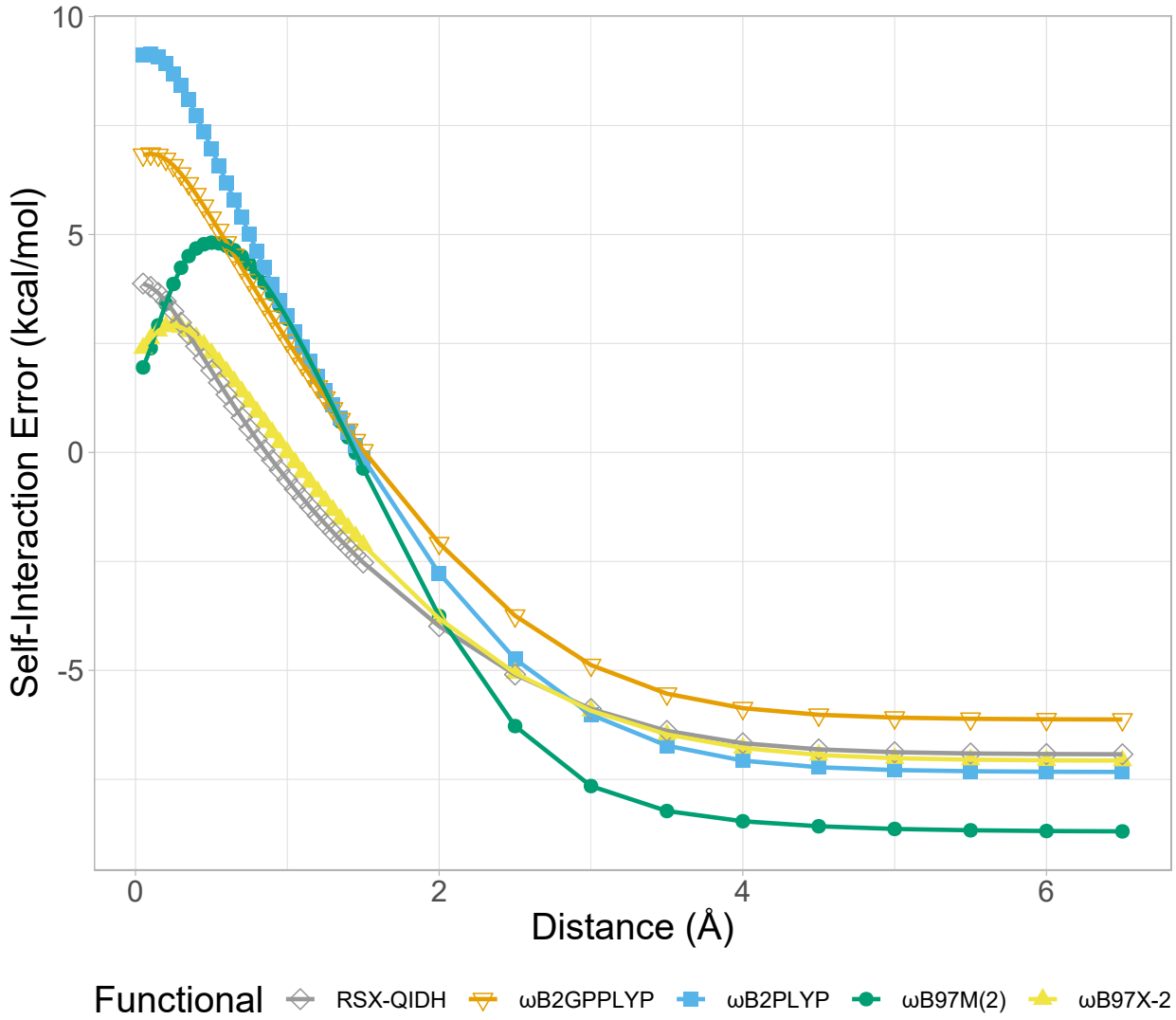

Figure S63: Self-interaction error vs internuclear distance of the H_2^+ molecule for RSX-QIDH, ωB2GPPLYP, ωB2PLYP, ωB97X-2 with ωB97M(2) as a reference.

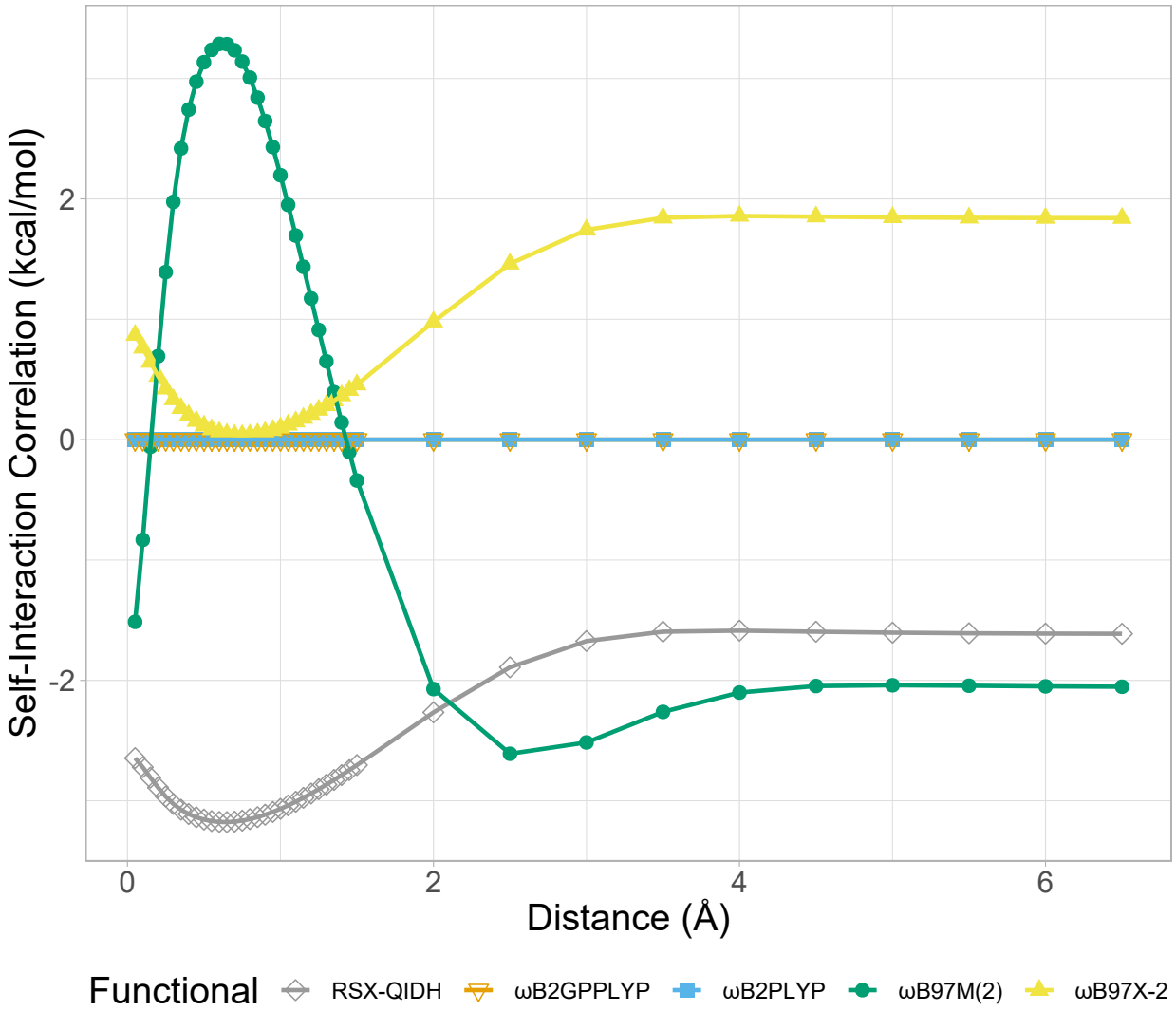

Figure S64: Self-interaction correlation error vs internuclear distance of the H_2^+ molecule for RSX-QIDH, ωB2GPPLYP, ωB2PLYP, ωB97X-2 with ωB97M(2) as a reference.

SI.3.2 Homonuclear Density and Functional error

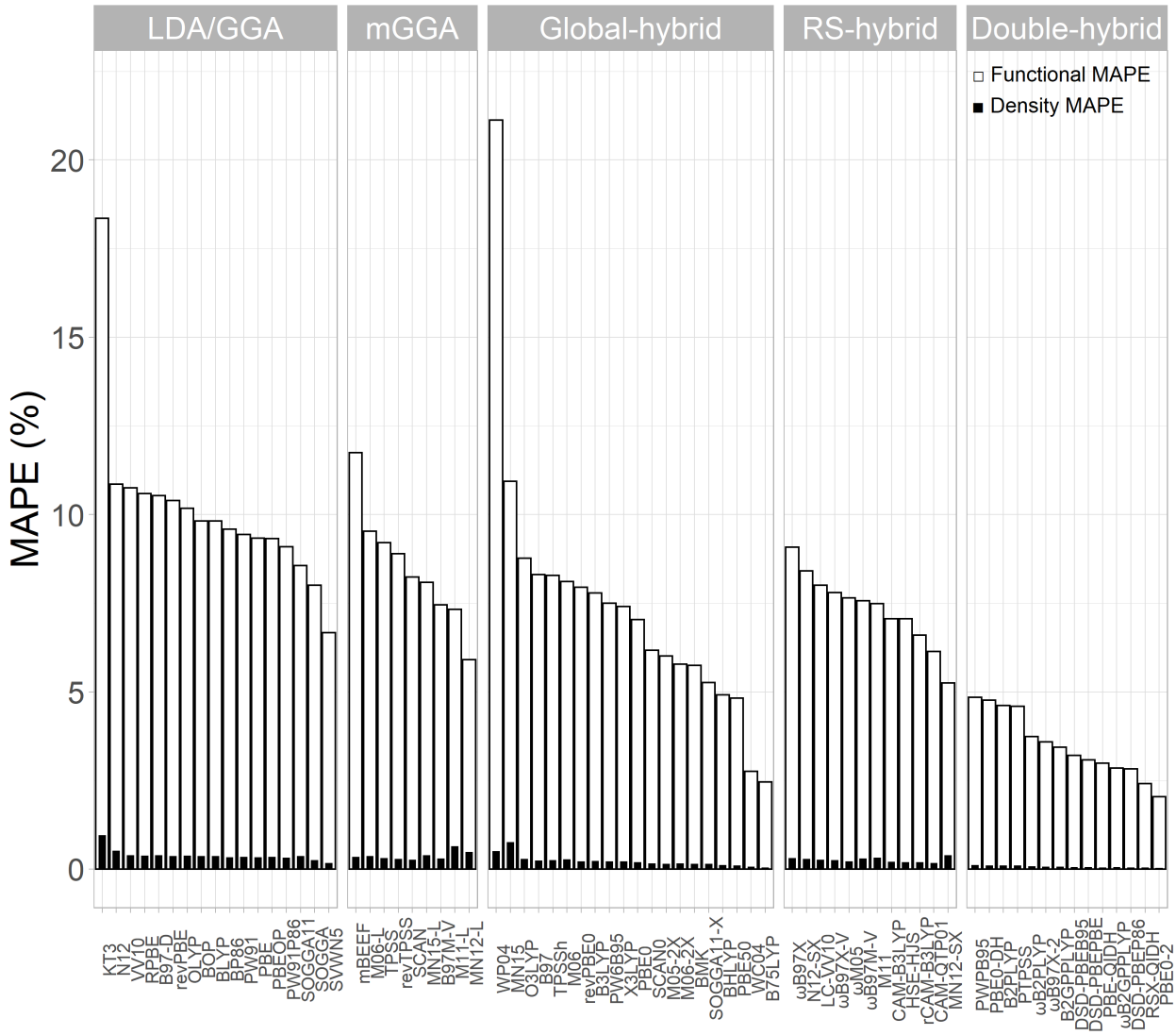

Figure S65: Functional (white) and density (black) mean absolute percentage errors across the homonuclear diatomic dataset, from H_2^+ to C_2^{11+}, for each density functional approximation.

SI.3.3 Diatomic self-interaction basis set error for select DFAs

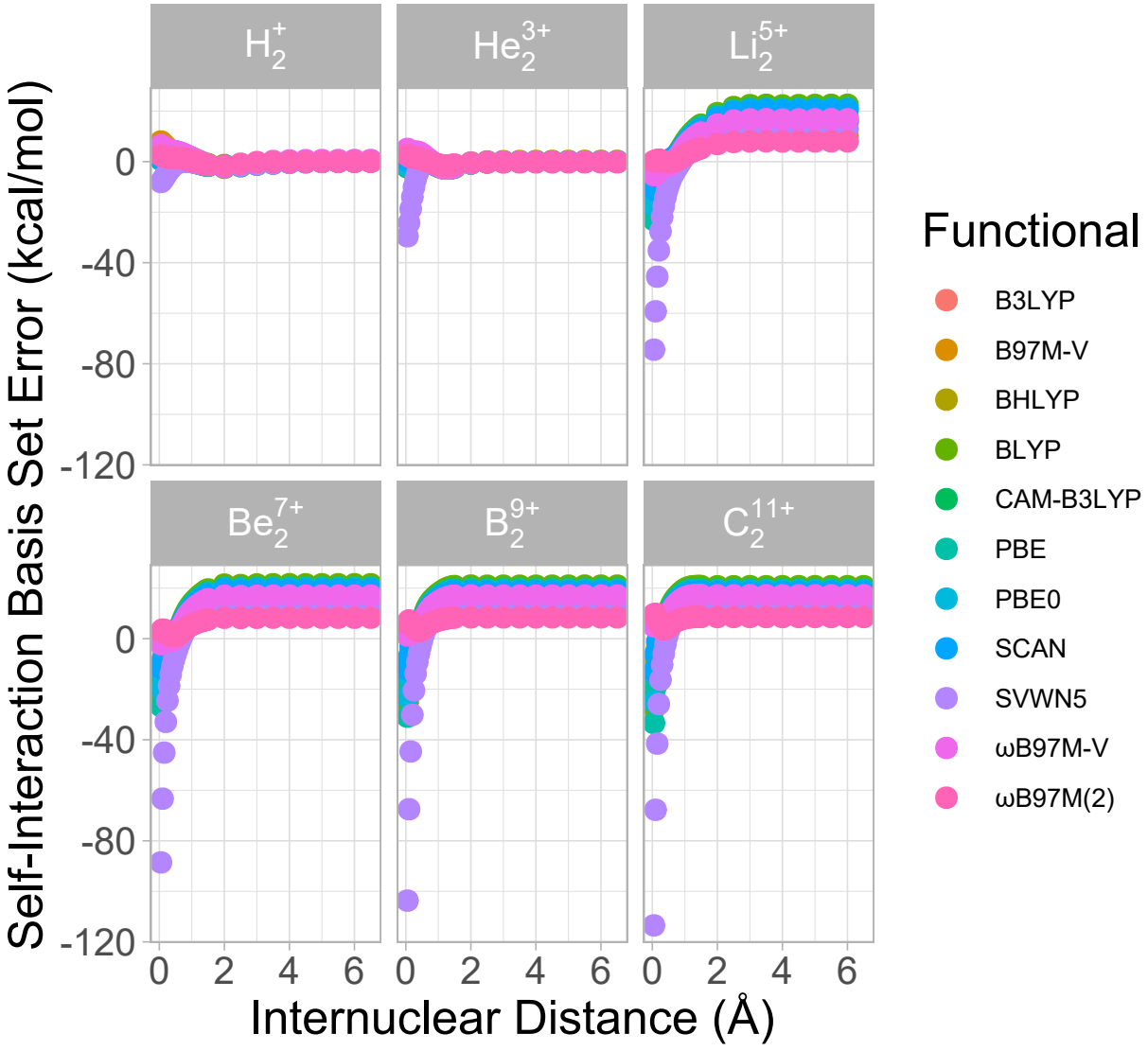

Figure S66: Self-interaction basis set error vs distance for the cc-pVDZ basis set across different homonuclear diatomics and density functional approximations.

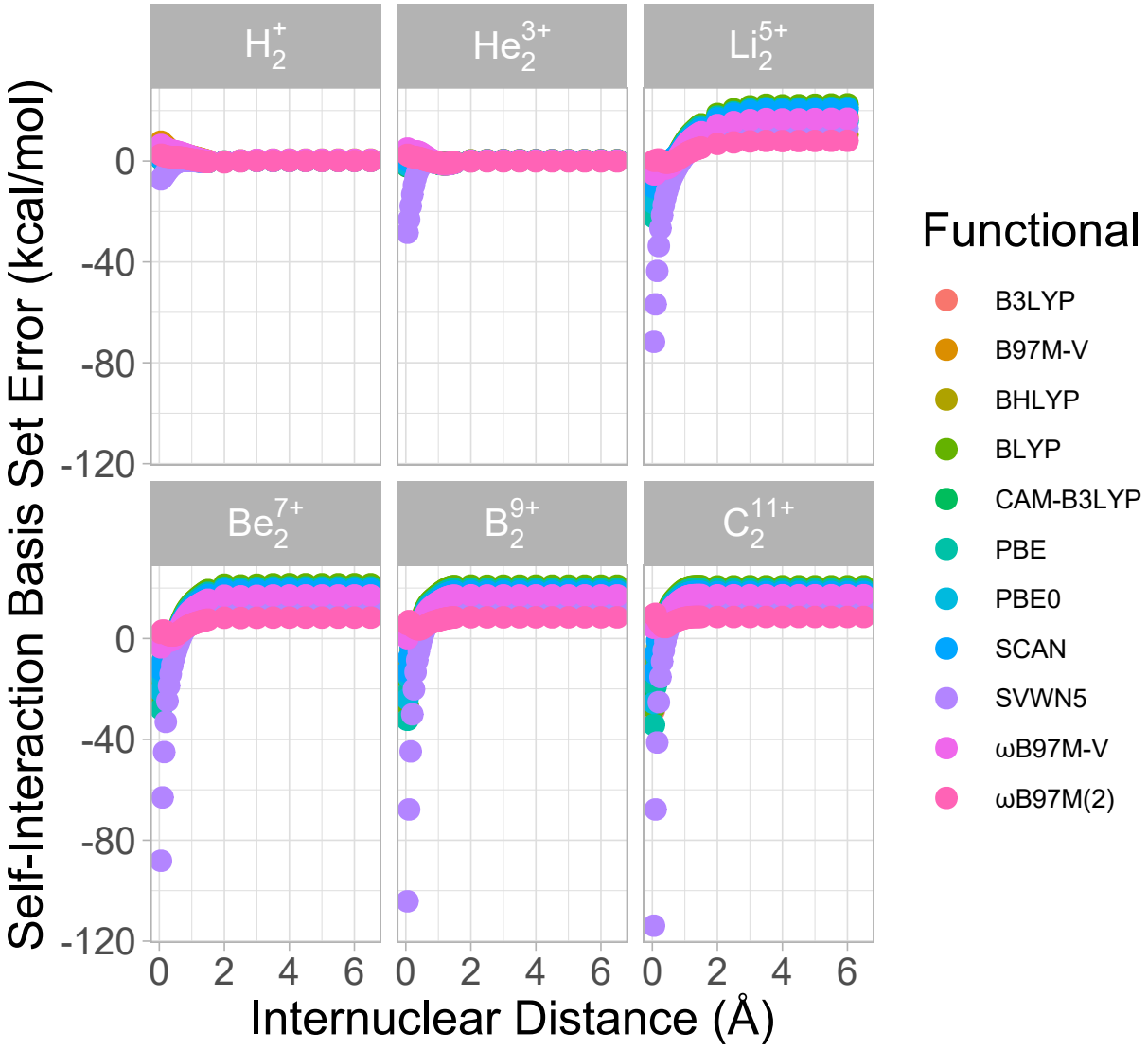

Figure S67: Self-interaction basis set error vs distance for the aug-cc-pVDZ basis set across different homonuclear diatomics and density functional approximations.

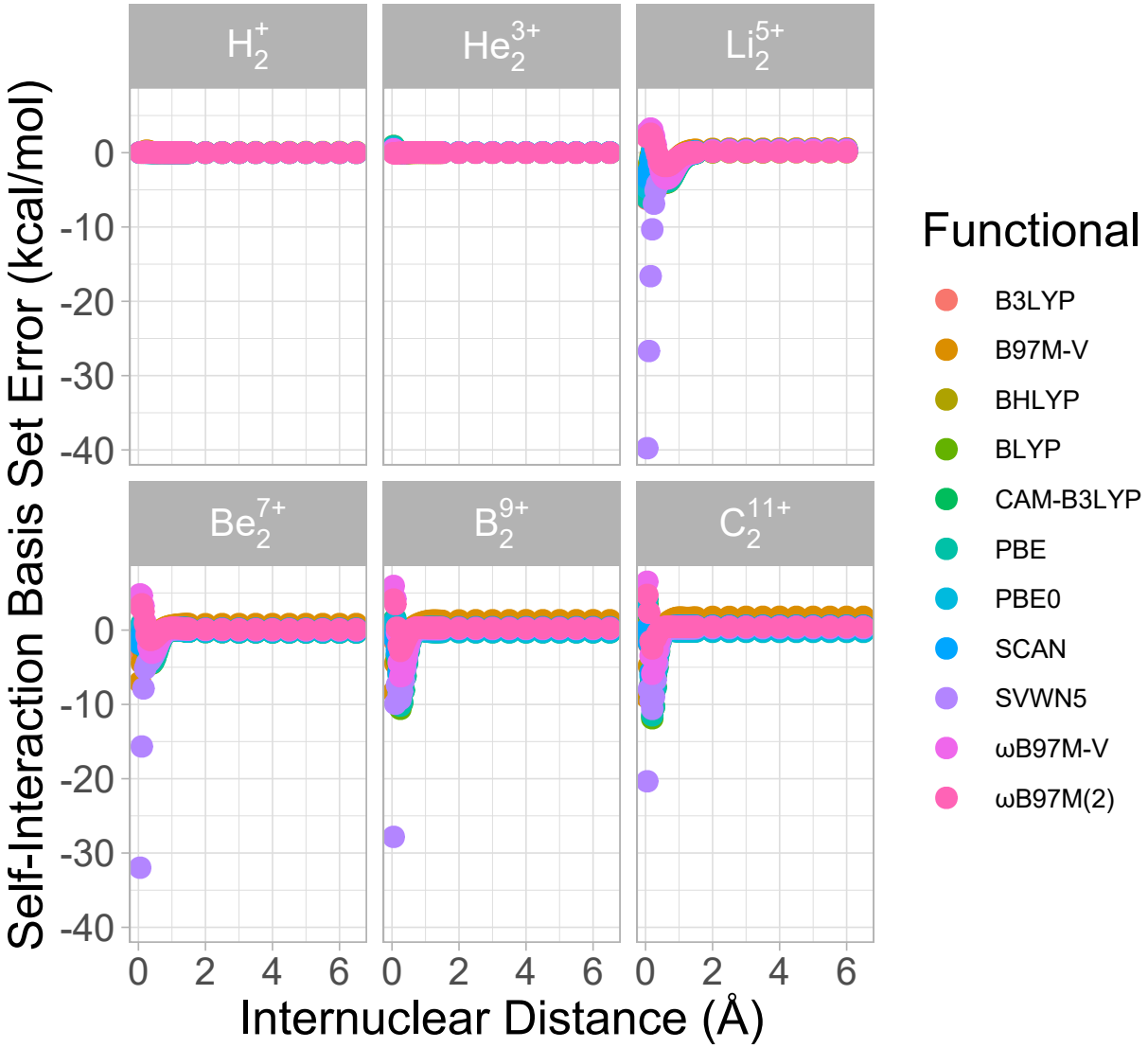

Figure S68: Self-interaction basis set error vs distance for the aug-cc-pVQZ basis set across different homonuclear diatomics and density functional approximations.

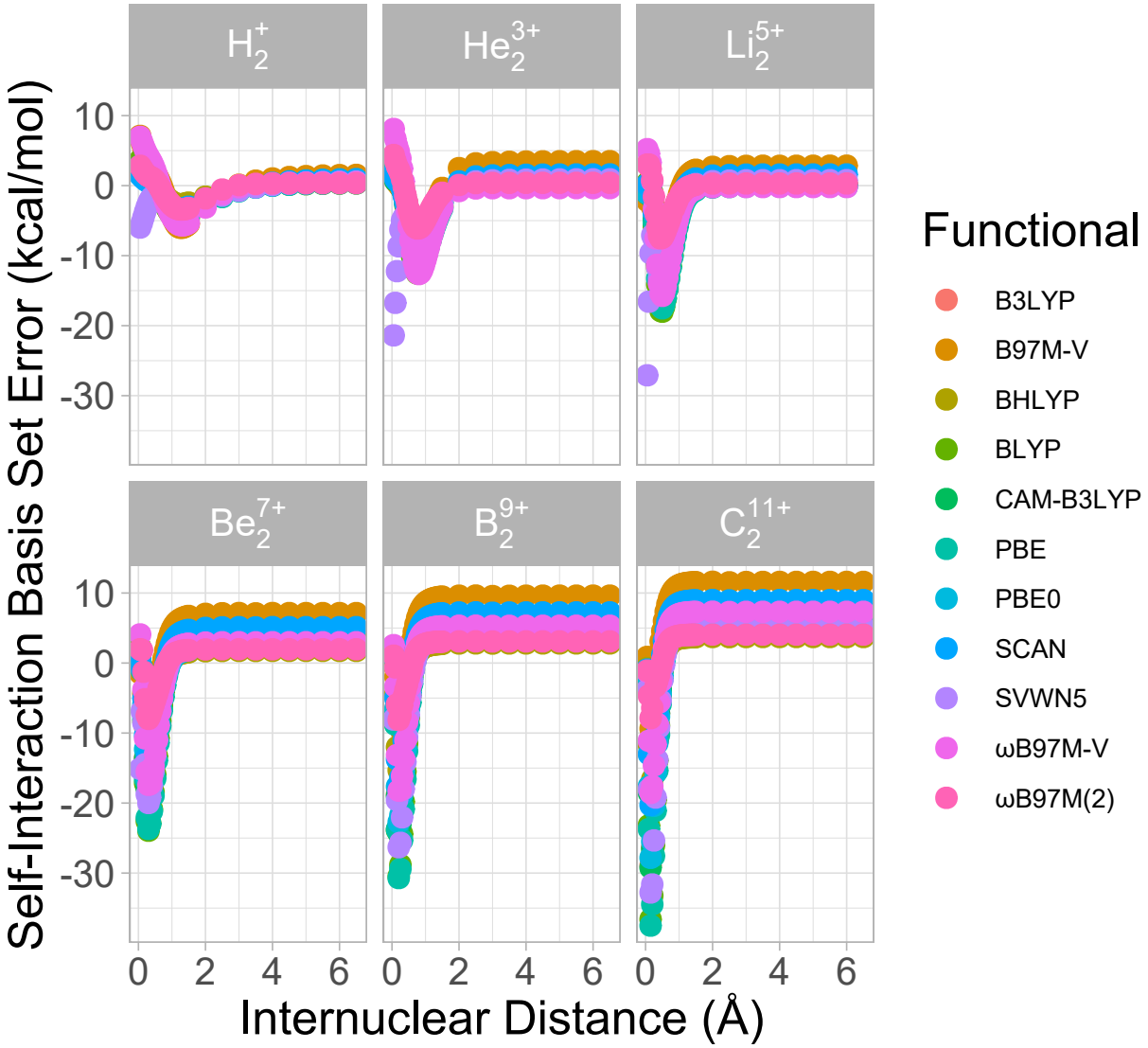

Figure S69: Self-interaction basis set error vs distance for the pc-0 basis set across different homonuclear diatomics and density functional approximations.

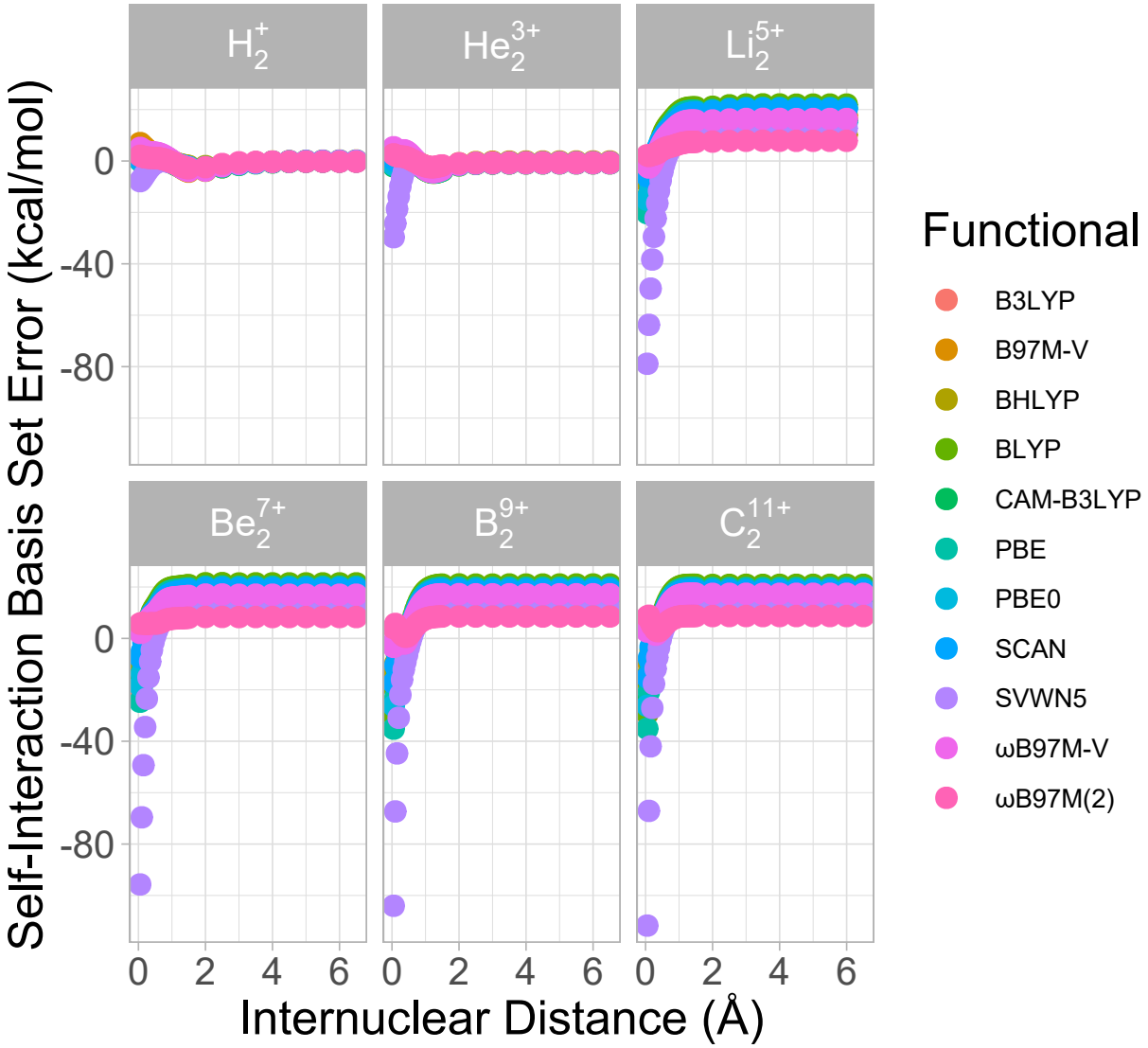

Figure S70: Self-interaction basis set error vs distance for the pc-1 basis set across different homonuclear diatomics and density functional approximations.

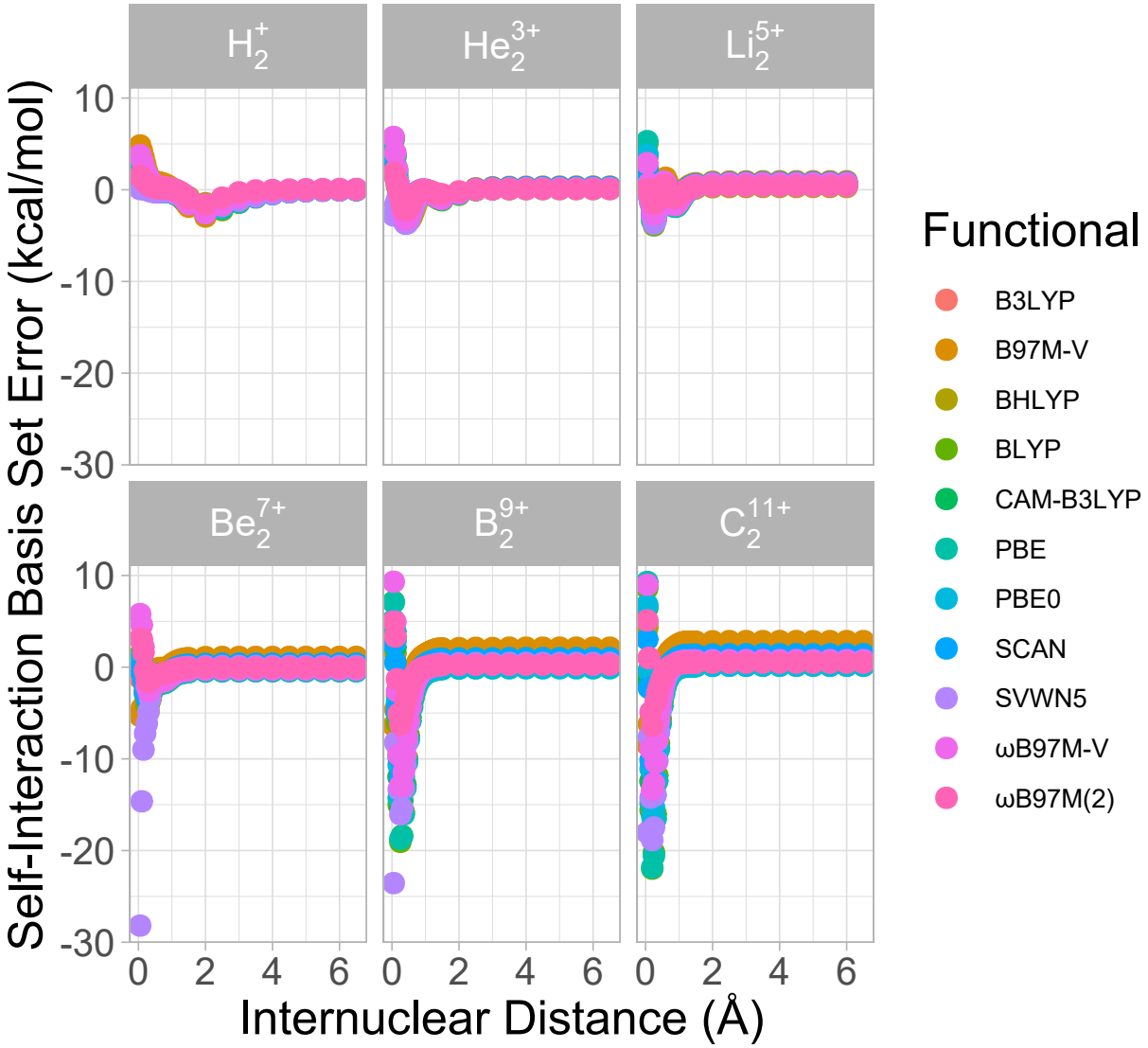

Figure S71: Self-interaction basis set error vs distance for the def2-TZVP basis set across different homonuclear diatomics and density functional approximations.

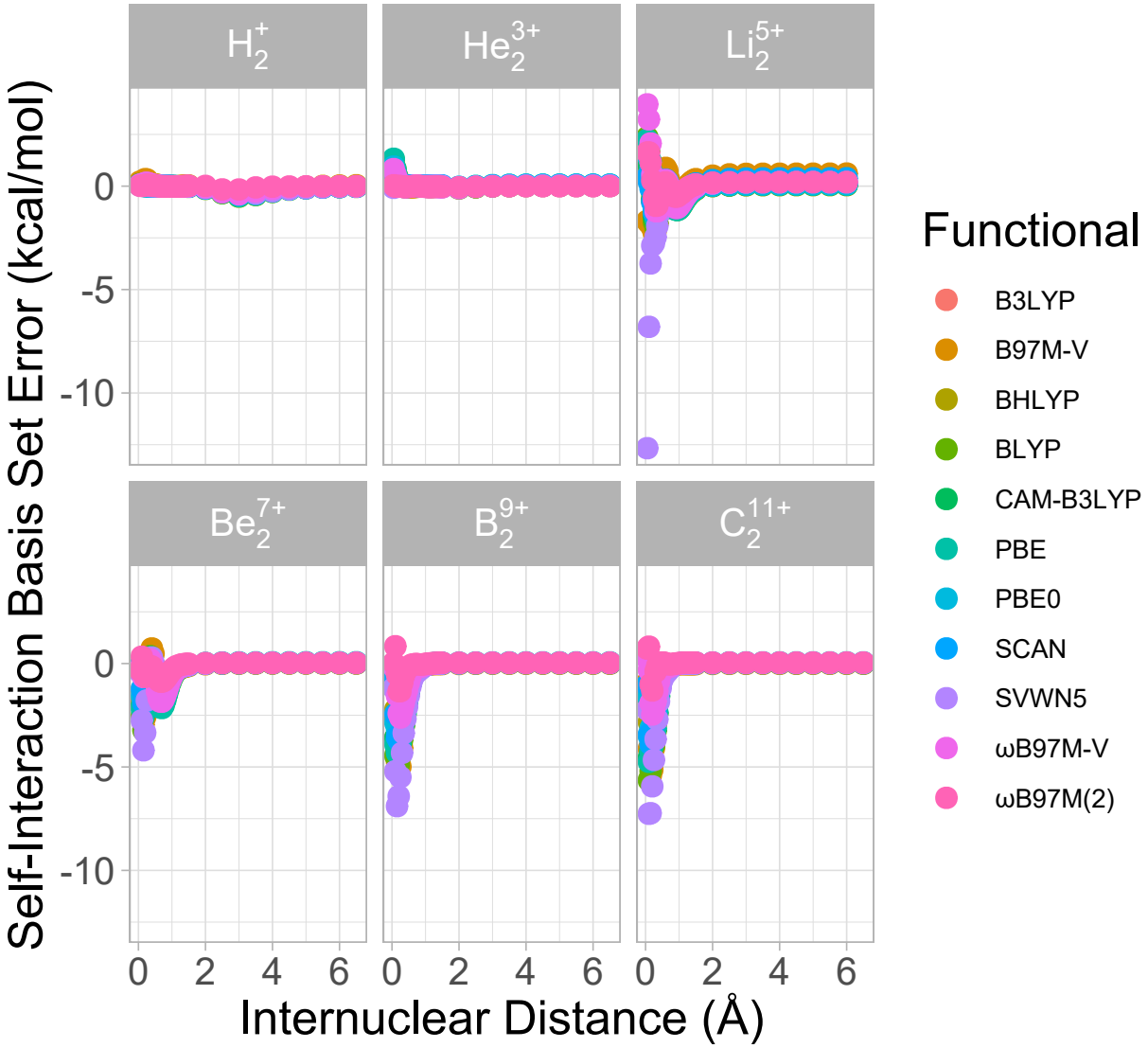

Figure S72: Self-interaction basis set error vs distance for the def2-QZVP basis set across different homonuclear diatomics and density functional approximations.

SI.3.4 Effects of dispersion corrections on diatomics

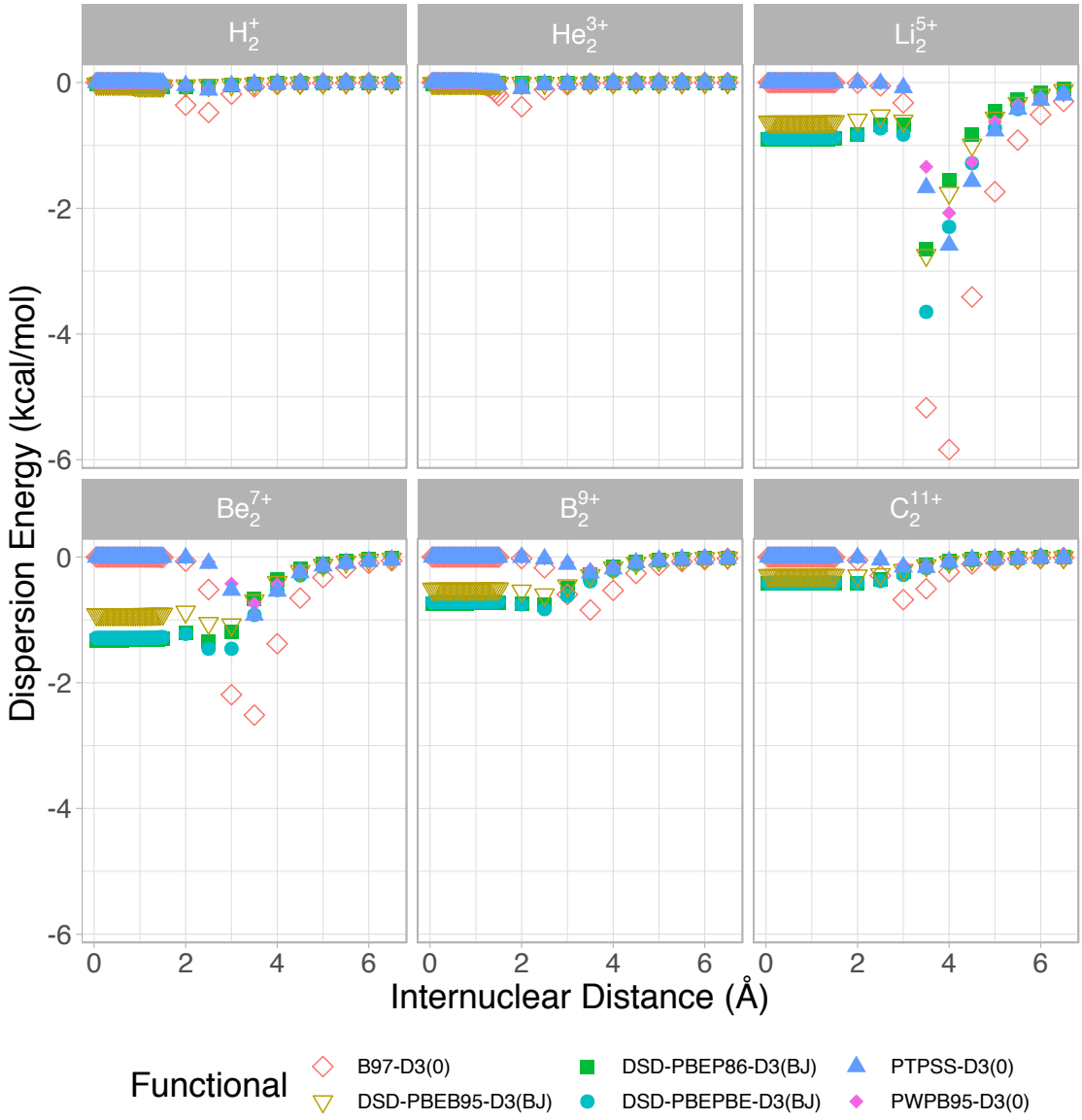

Figure S73: Non-local van der Waals correction based on Grimme's DFT-D3 correction for the diatomic series dataset for the relevant density functional approximations. The DFT-D3(0) correction makes the dispersion correction go to zero at the close range, whereas DFT-D3(BJ) makes the correction tend to a constant at the close range.

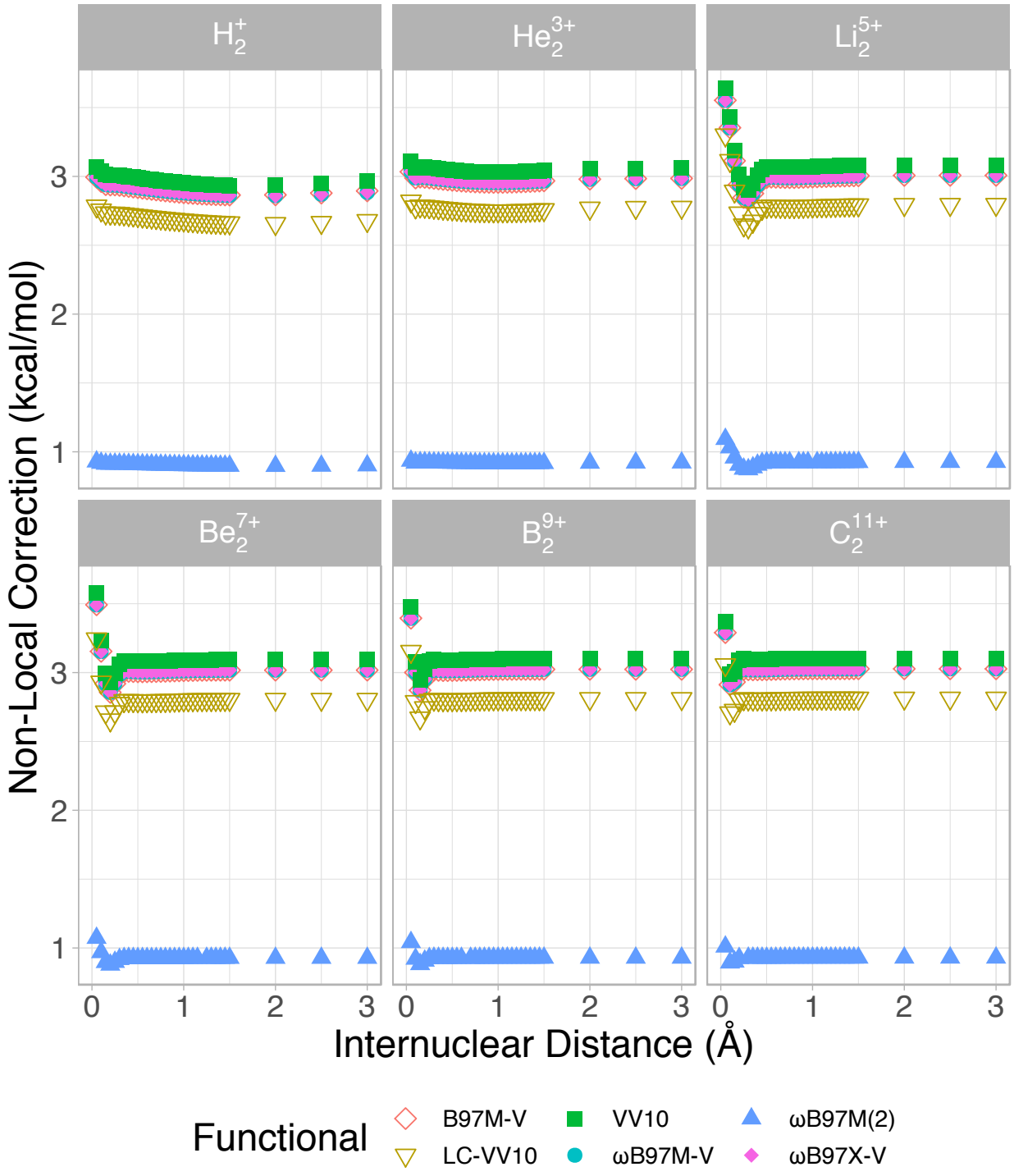

Figure S74: Non-local van der Waals correction based on the VV10 non-local kernel for the diatomic series dataset for the relevant density functional approximations.

SI.3.5 Incremental diatomics

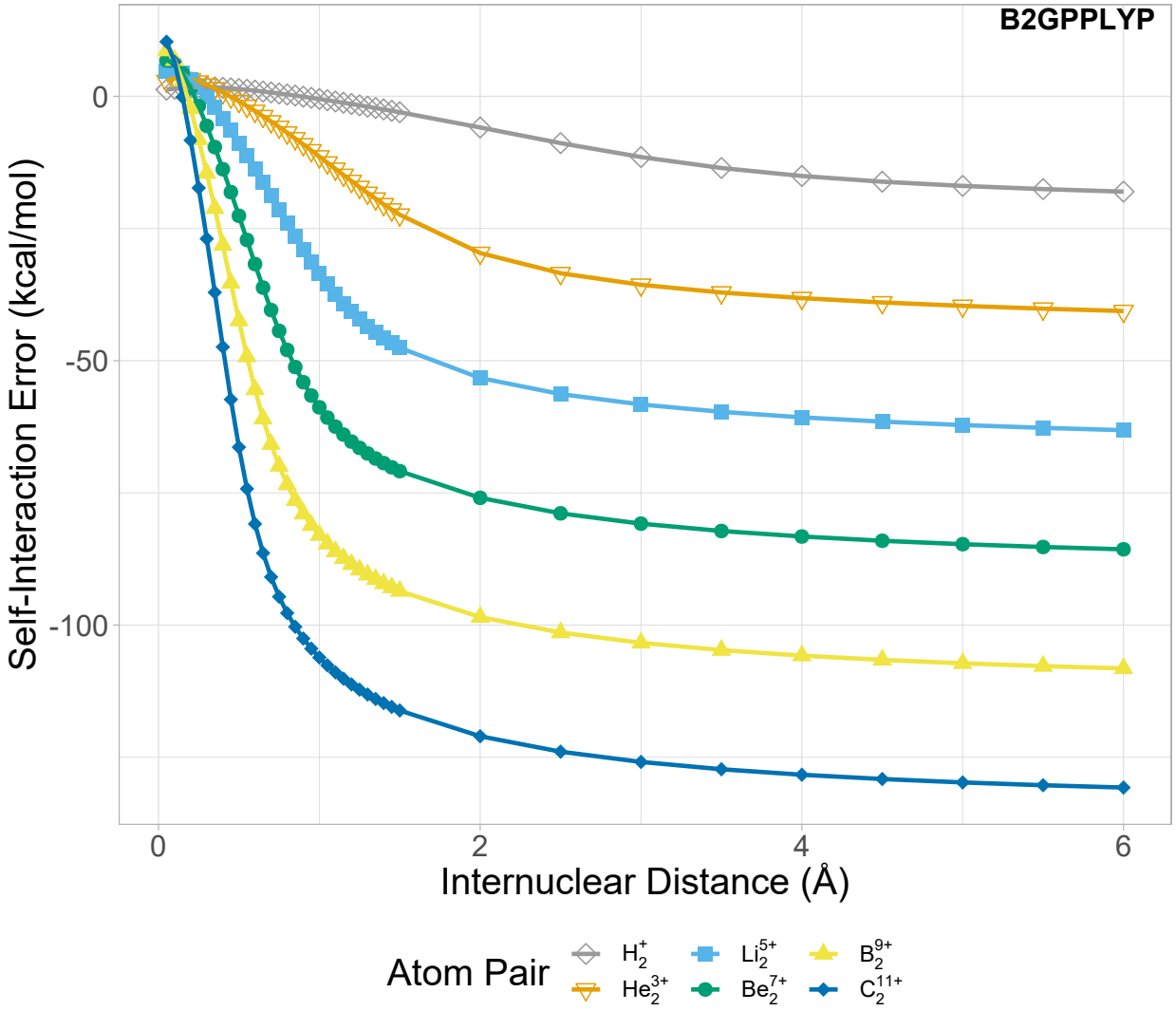

Figure S75: Self-interaction error vs internuclear distance for incremental homonuclear pairs for B2GPPLYP.

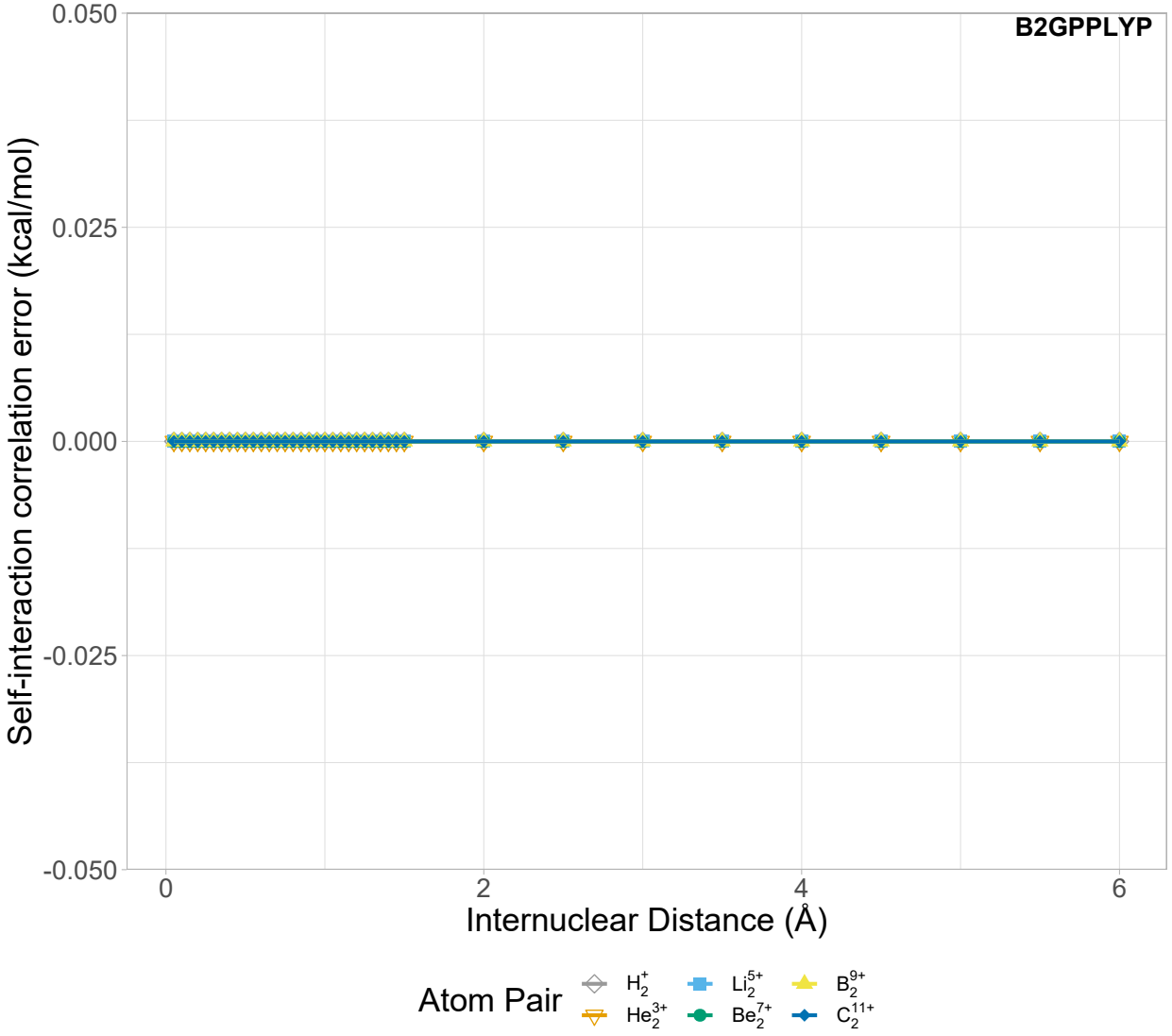

Figure S76: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for B2GPPLYP.

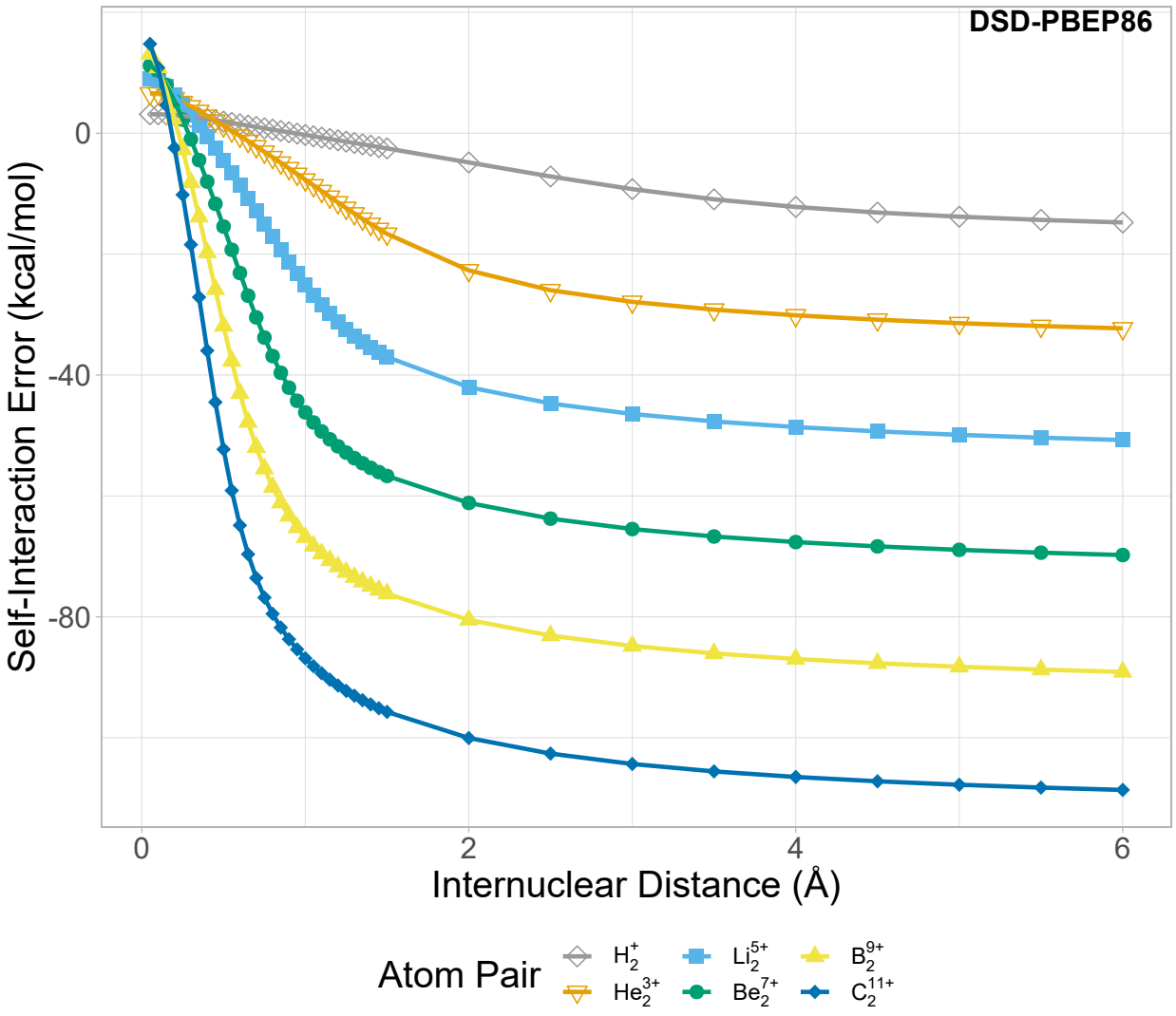

Figure S77: Self-interaction error vs internuclear distance for incremental homonuclear pairs for DSD-PBEP86.

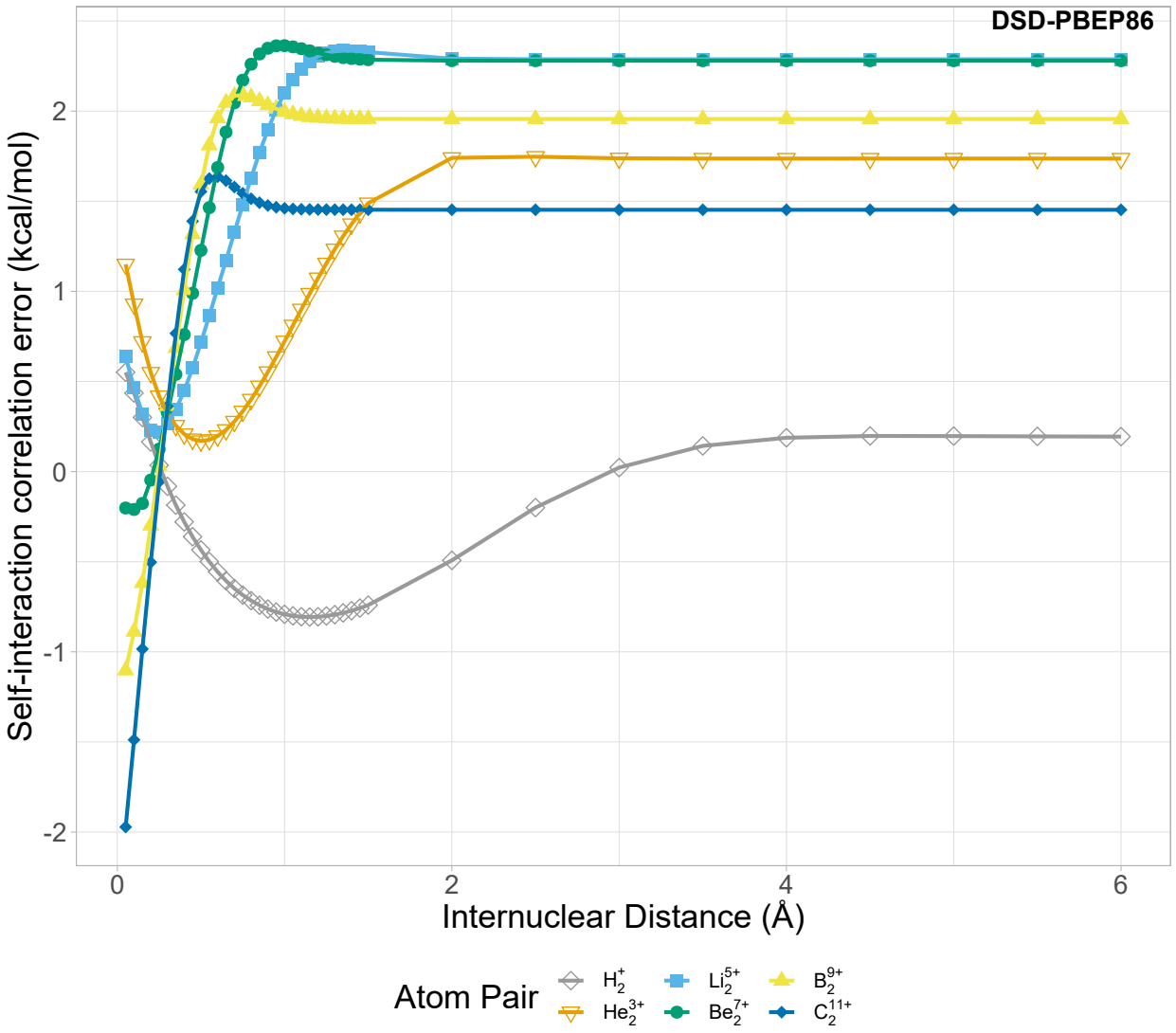

Figure S78: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for DSD-PBEP86.

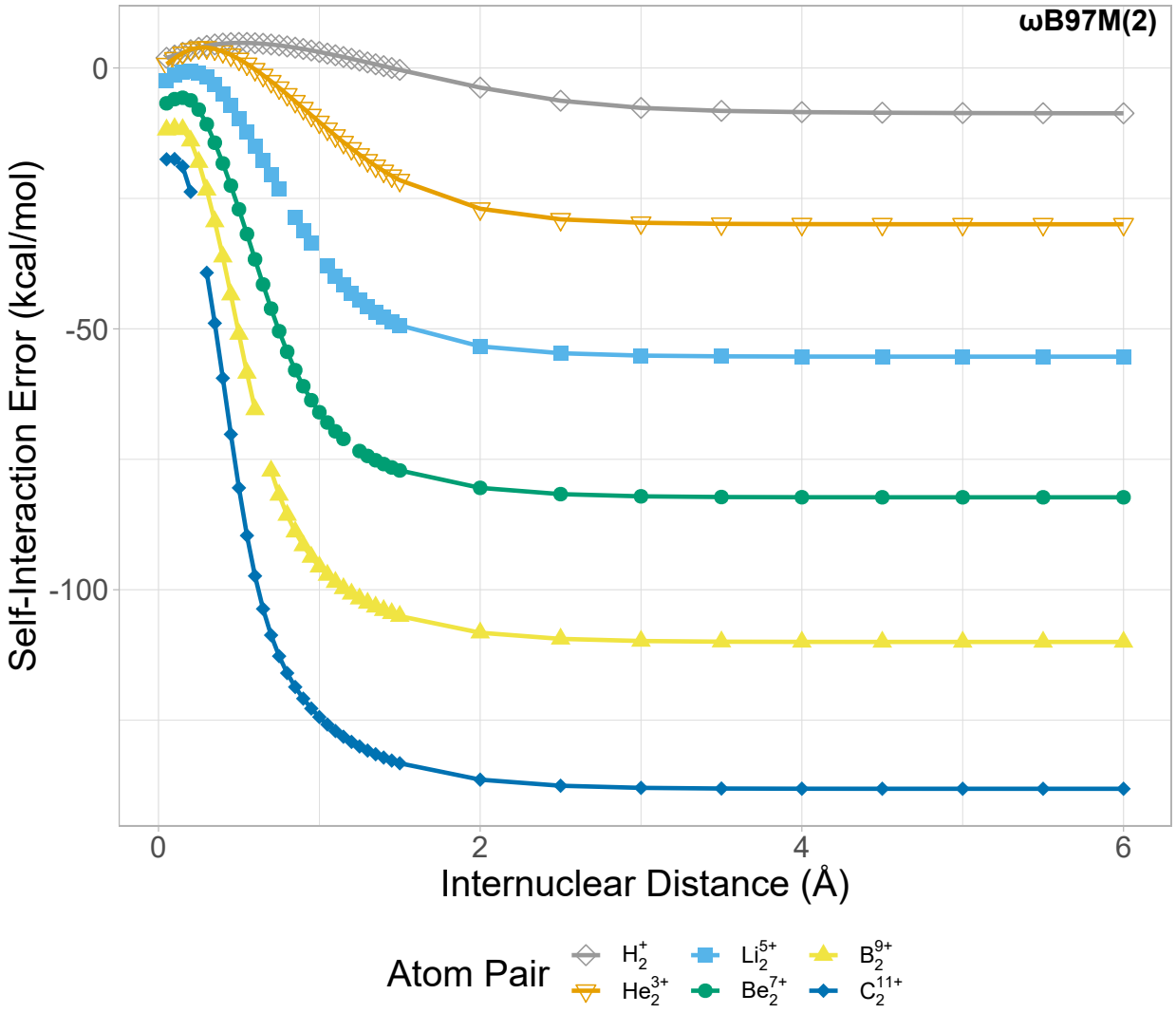

Figure S79: Self-interaction error vs internuclear distance for incremental homonuclear pairs for ωB97M(2).

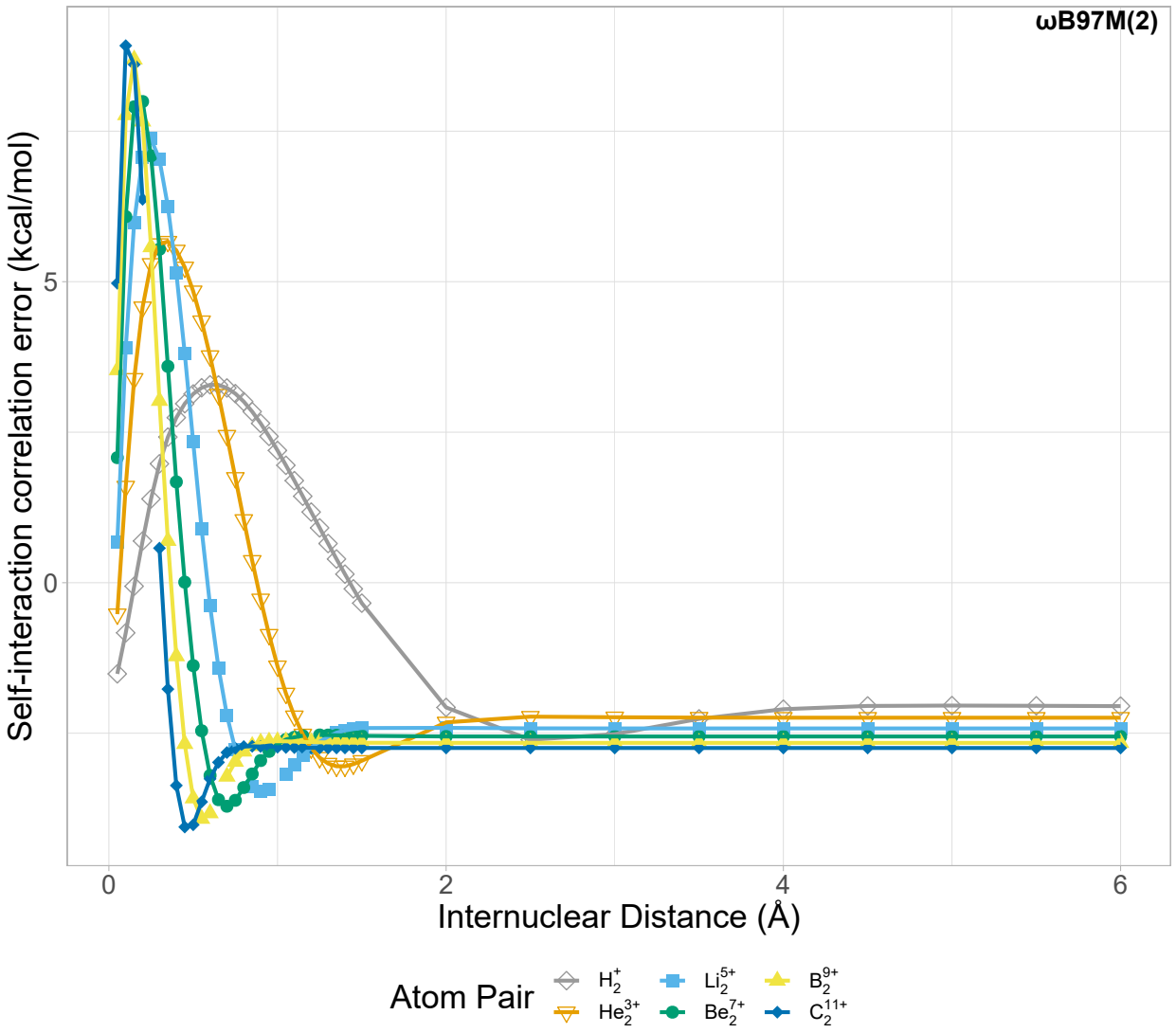

Figure S80: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for ωB97M(2).

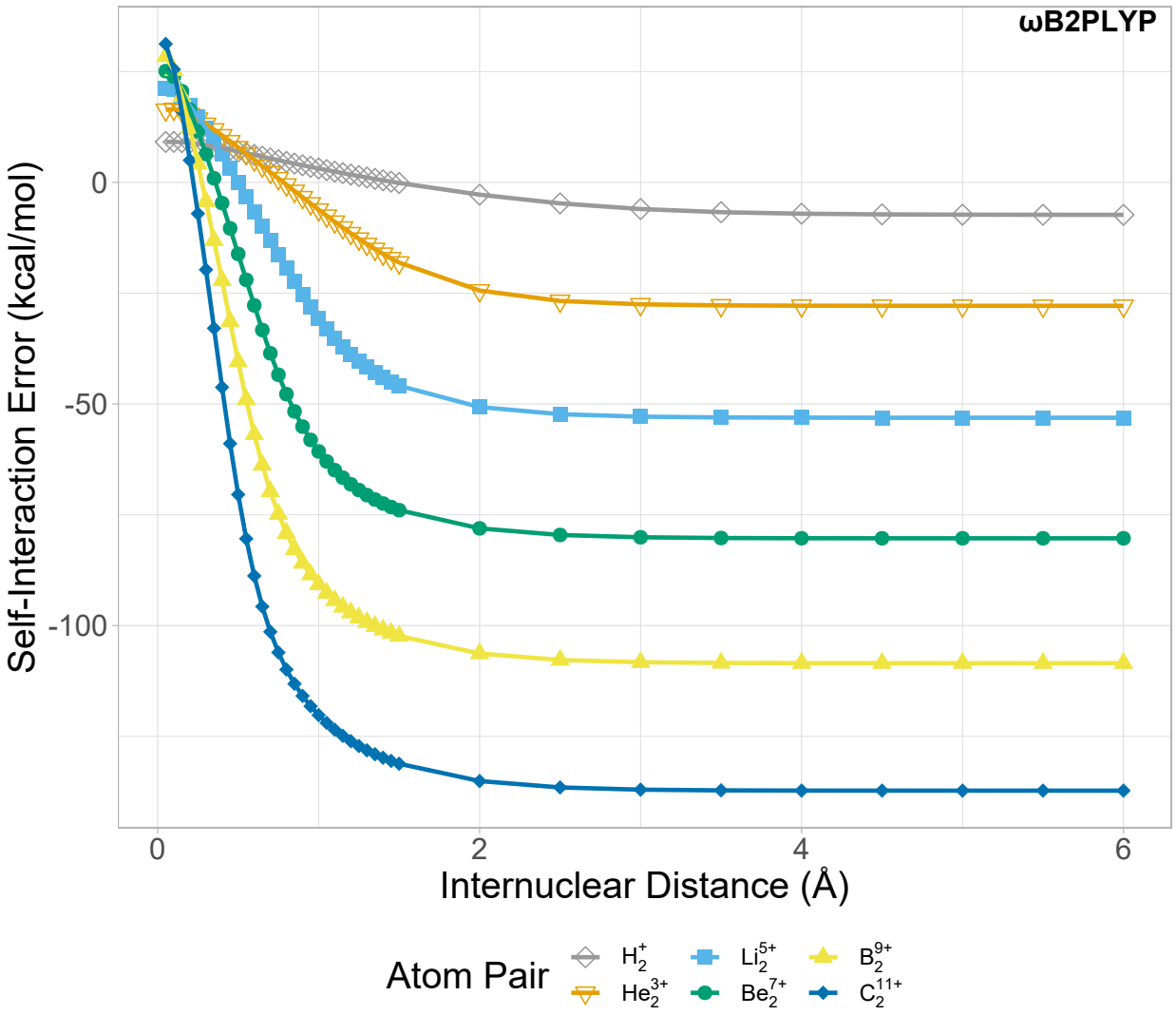

Figure S81: Self-interaction error vs internuclear distance for incremental homonuclear pairs for ωB2PLYP.

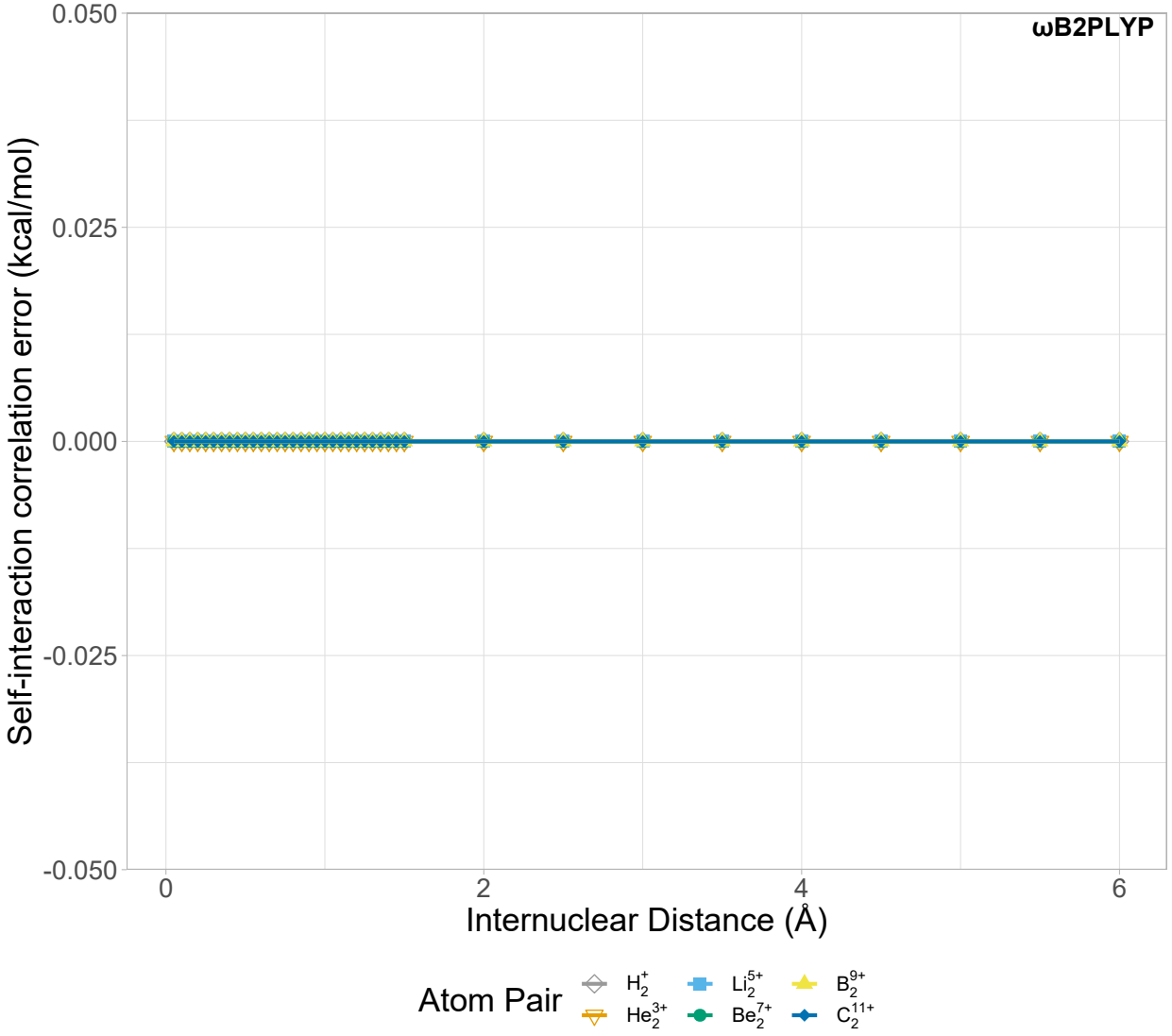

Figure S82: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for ωB2PLYP.

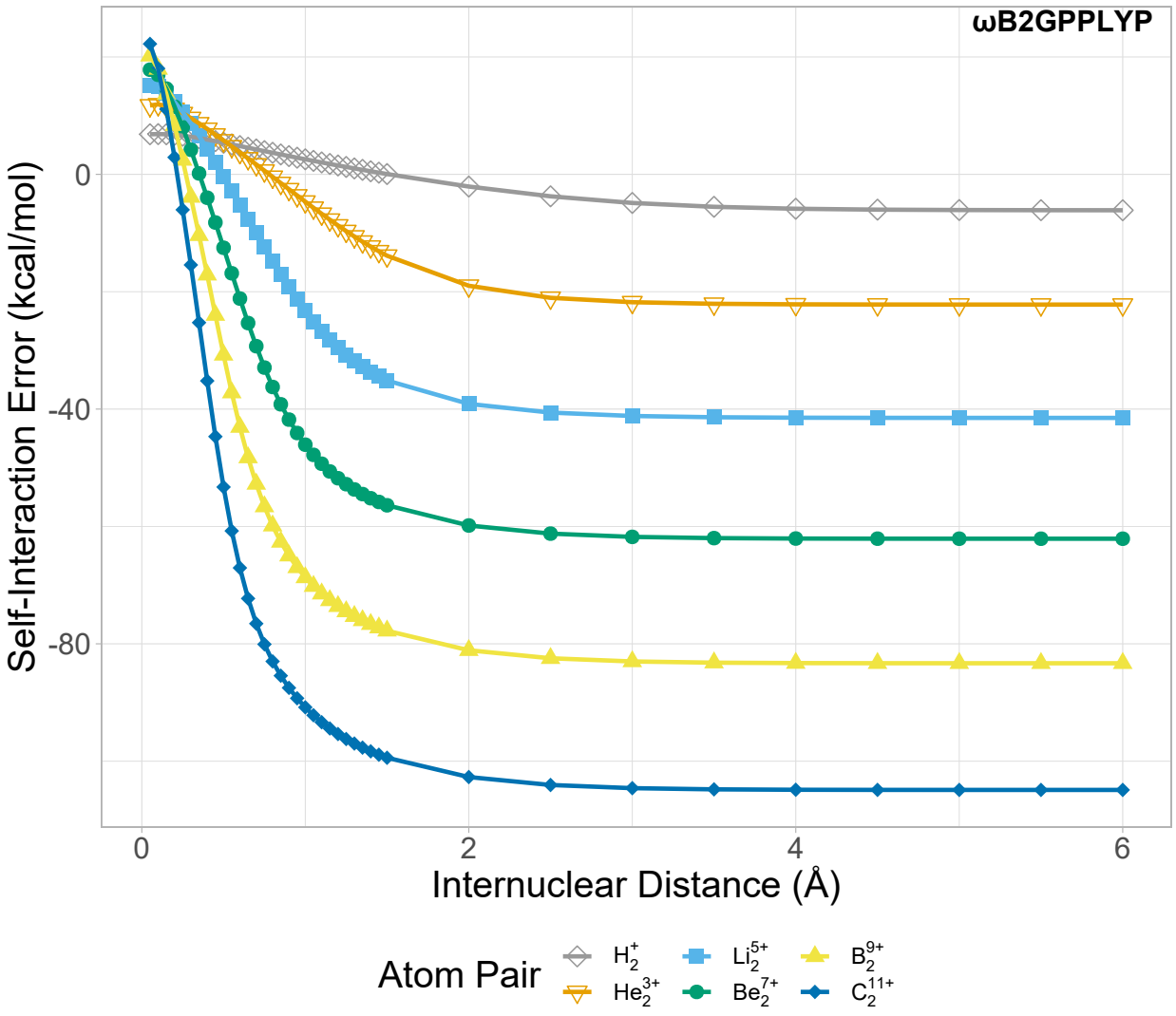

Figure S83: Self-interaction error vs internuclear distance for incremental homonuclear pairs for ωB2GPPLYP.

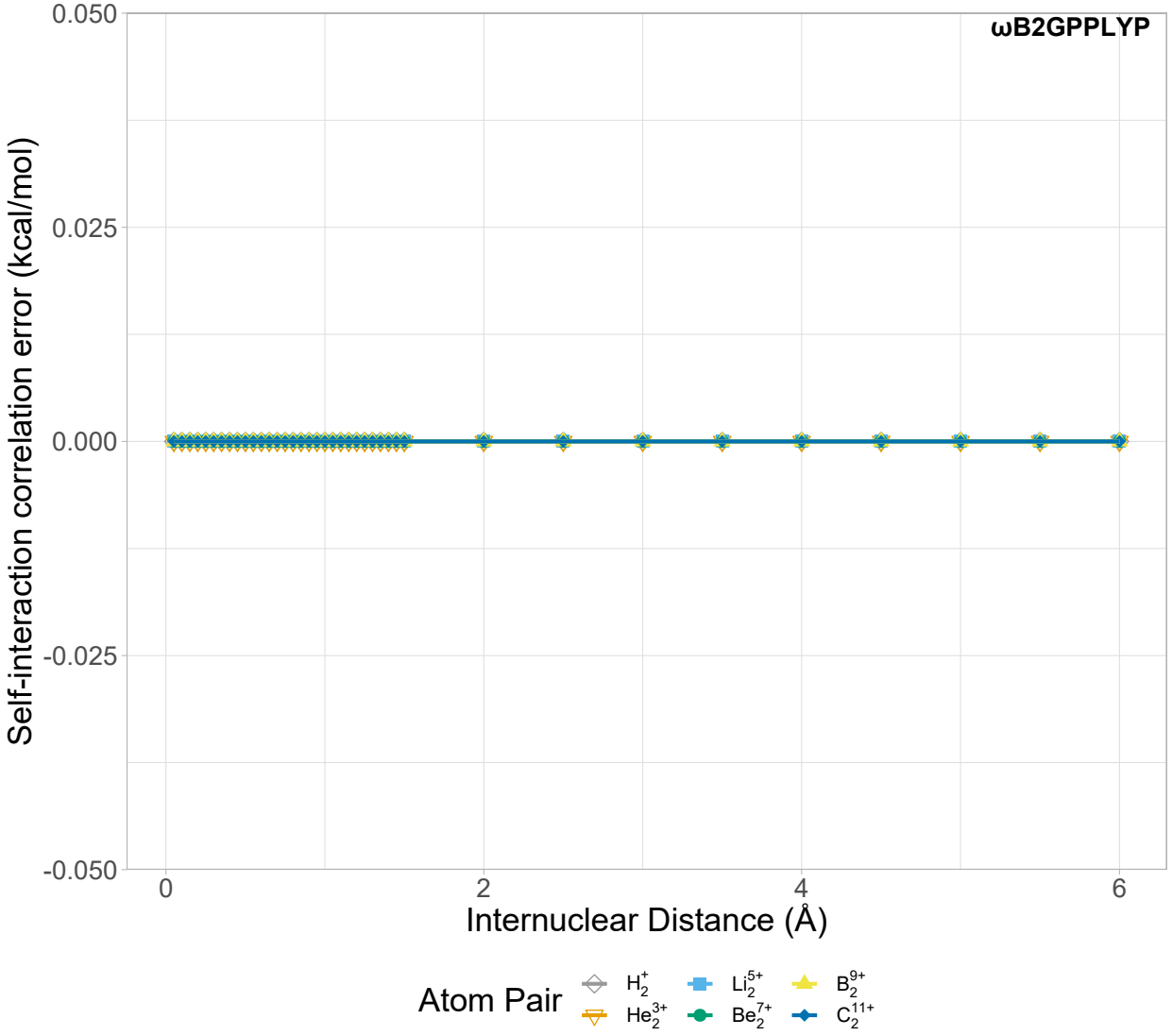

Figure S84: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for ωB2GPPLYP.

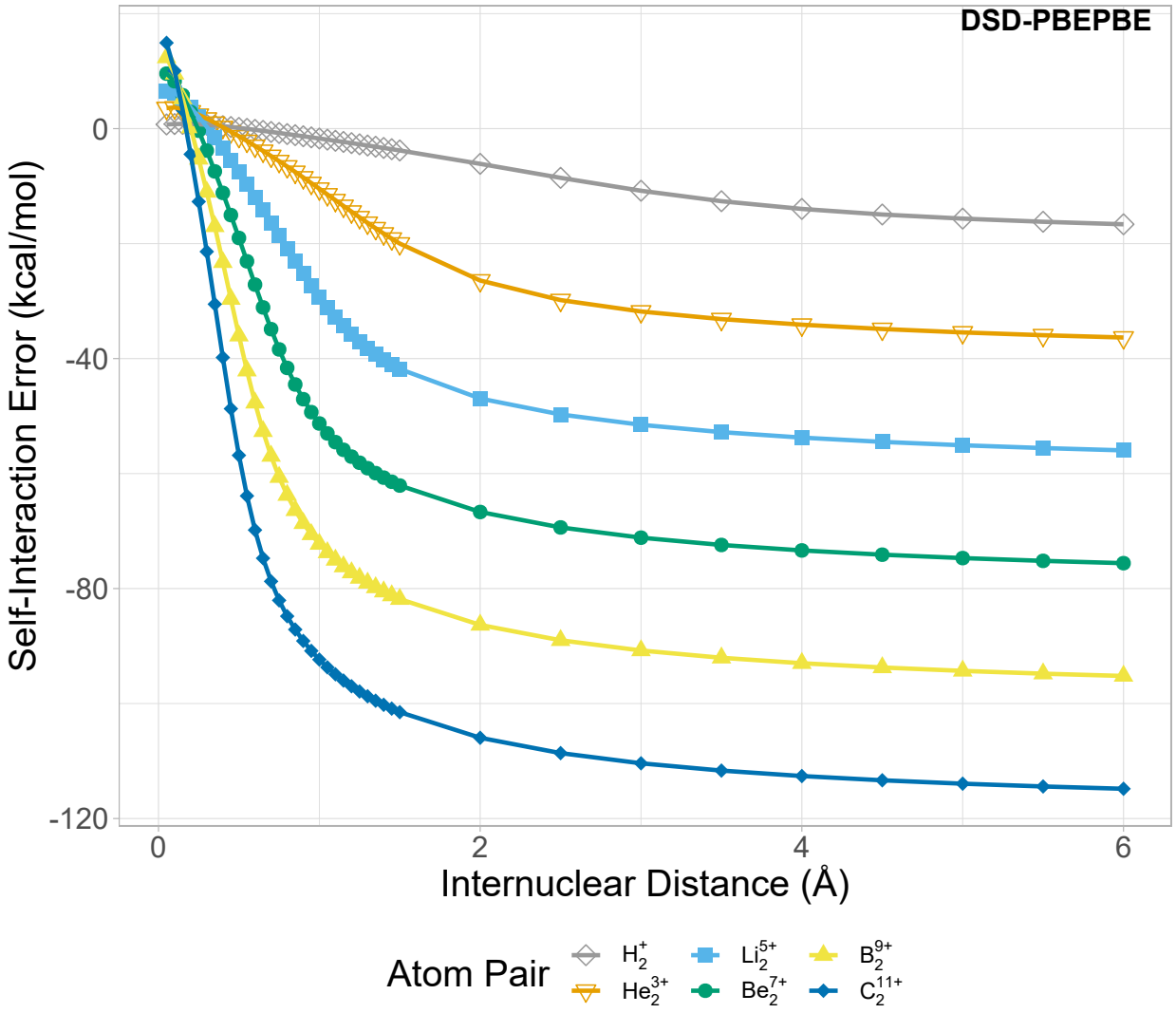

Figure S85: Self-interaction error vs internuclear distance for incremental homonuclear pairs for DSD-PBEPBE.

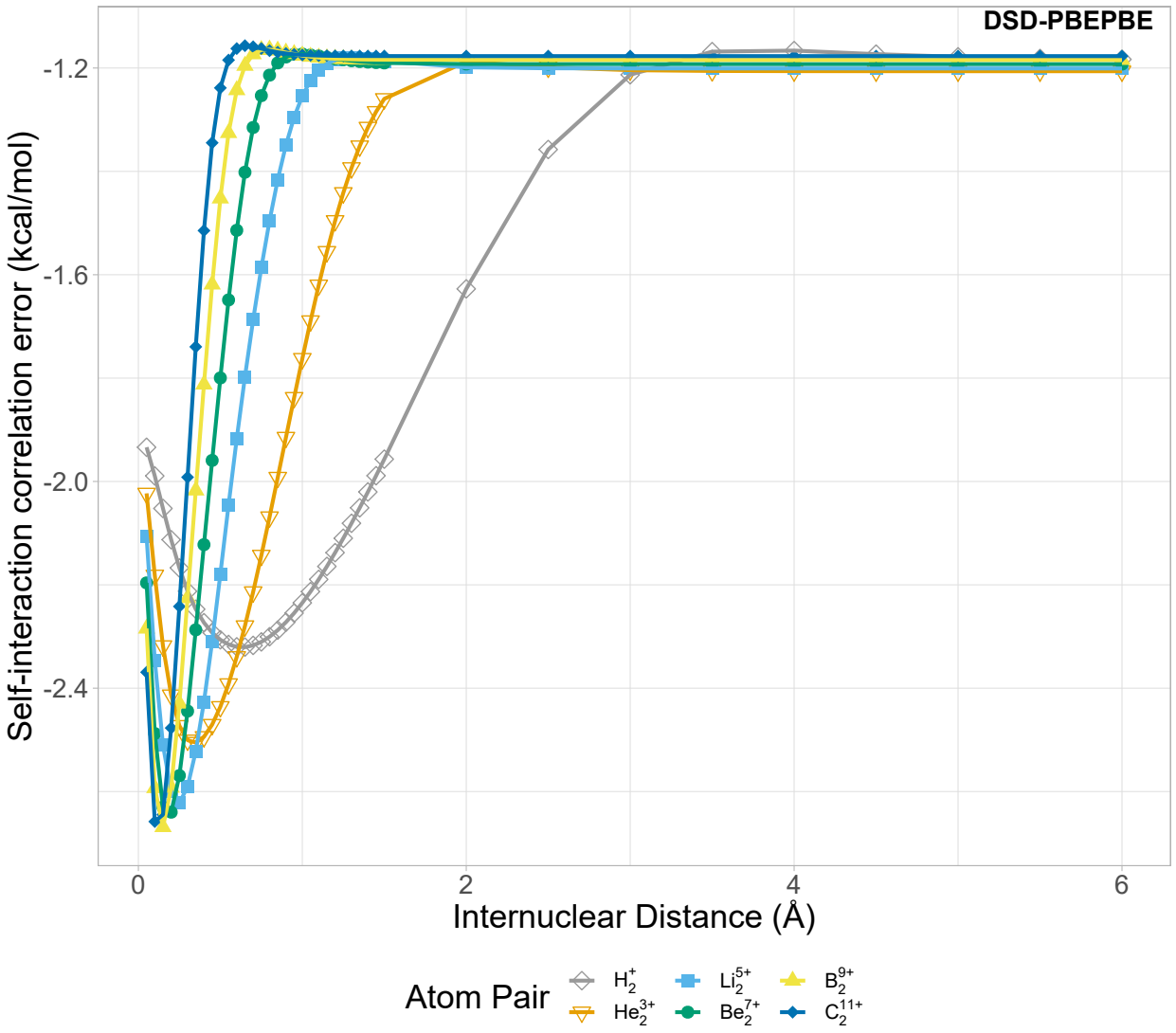

Figure S86: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for DSD-PBEPBE.

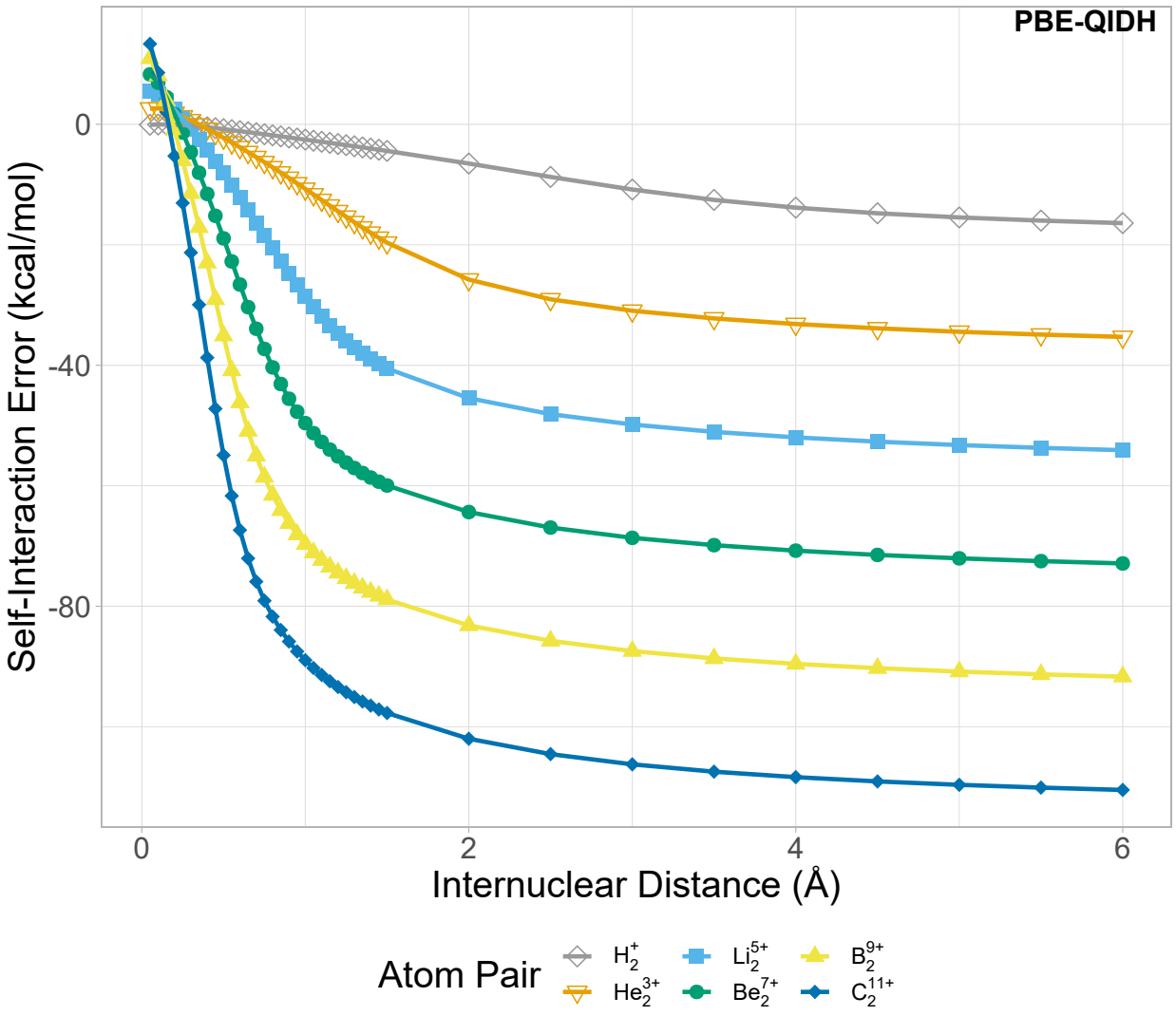

Figure S87: Self-interaction error vs internuclear distance for incremental homonuclear pairs for PBE-QIDH.

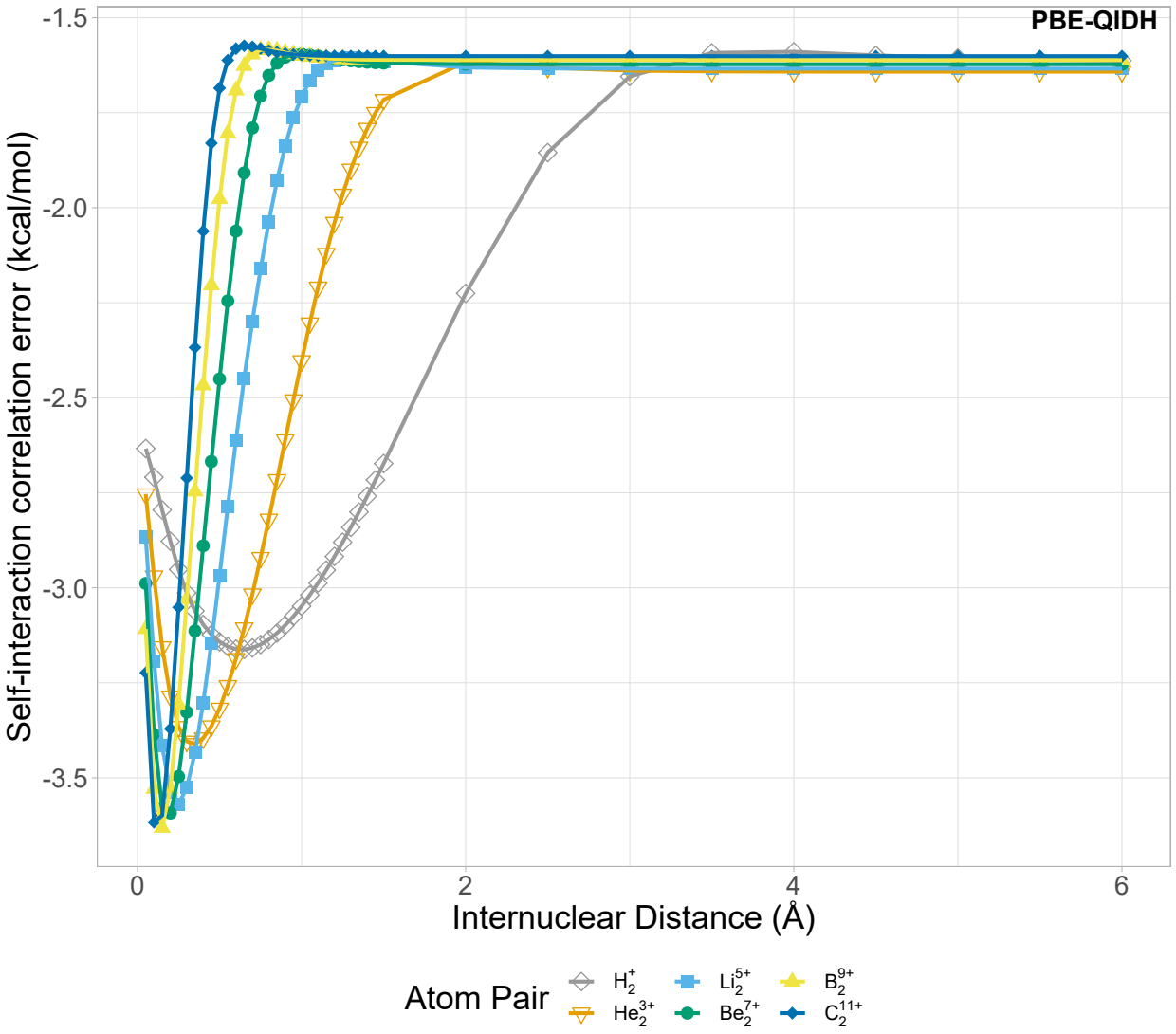

Figure S88: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for PBE-QIDH.

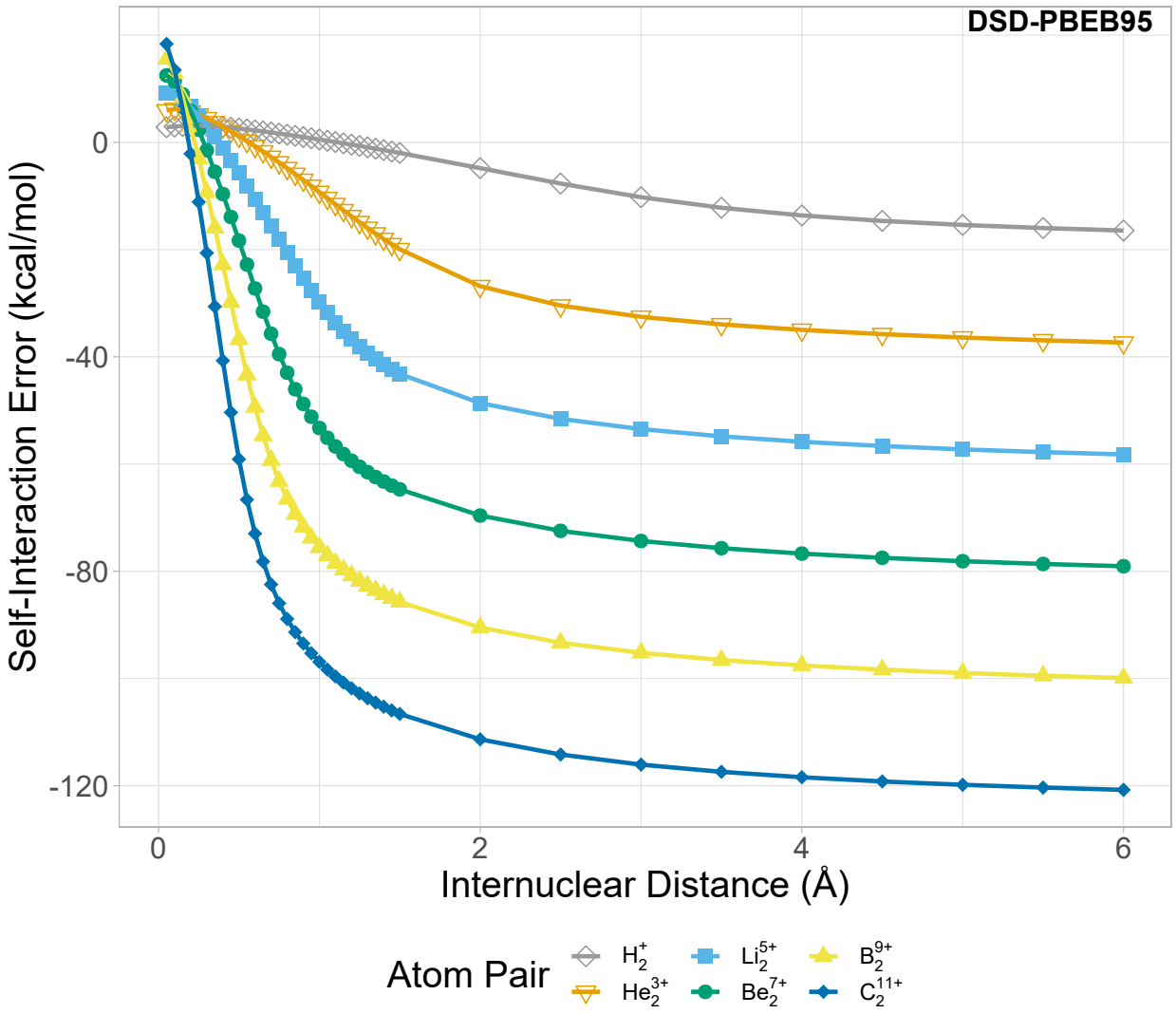

Figure S89: Self-interaction error vs internuclear distance for incremental homonuclear pairs for DSD-PBEB95.

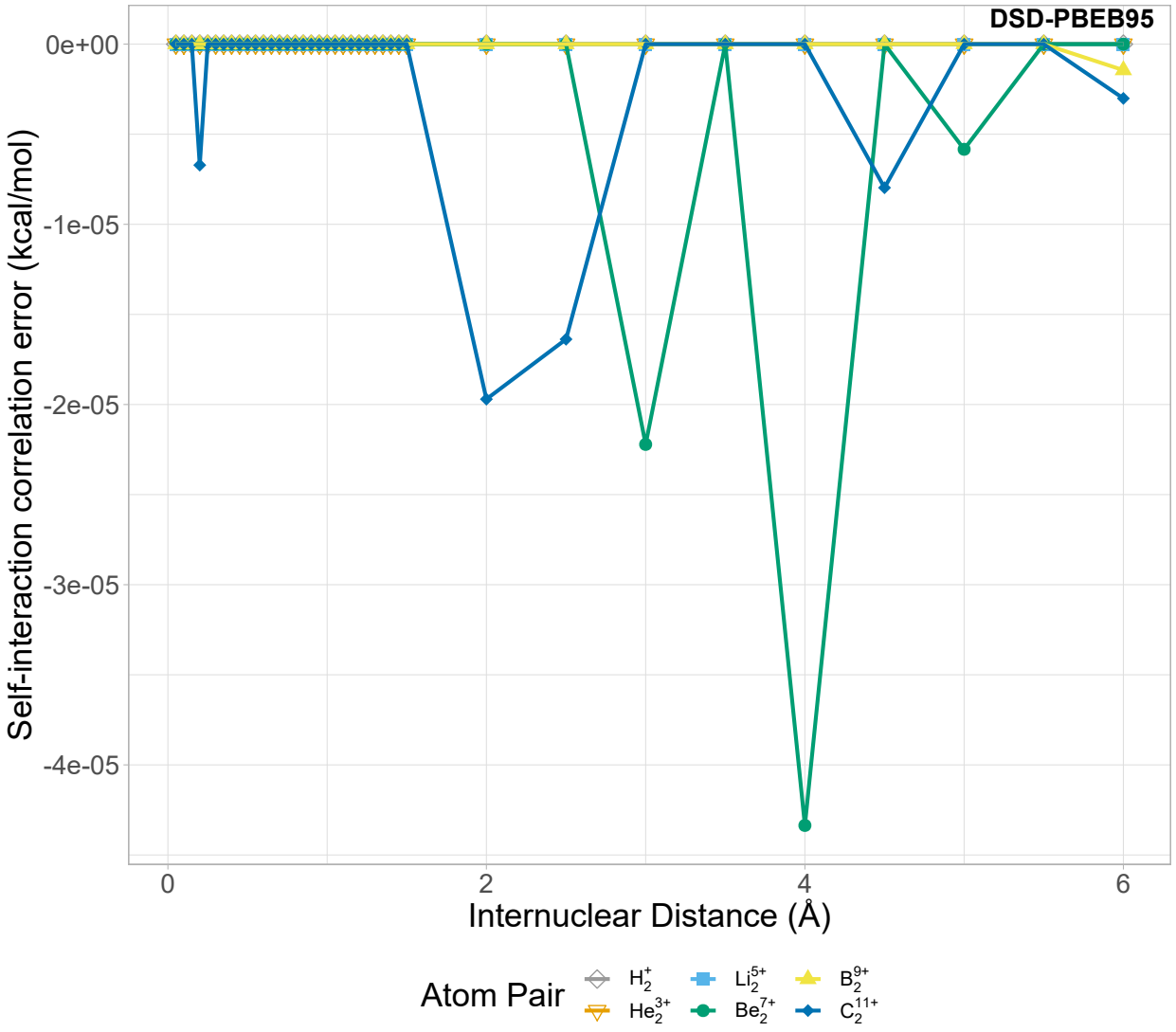

Figure S90: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for DSD-PBEB95. Formally this DFA is self-correlation free, any deviations from this is likely computational noise.

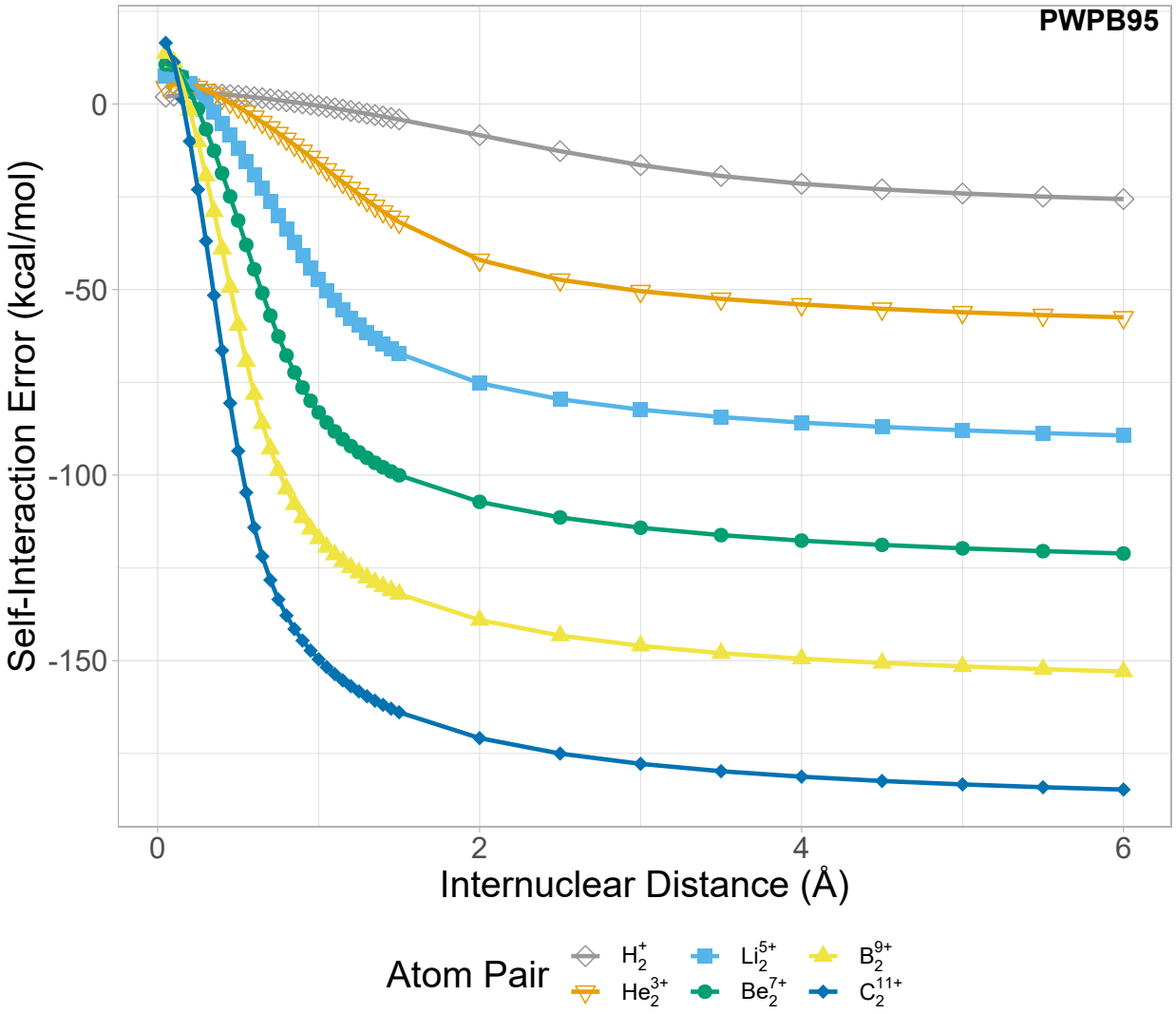

Figure S91: Self-interaction error vs internuclear distance for incremental homonuclear pairs for PWPB95.

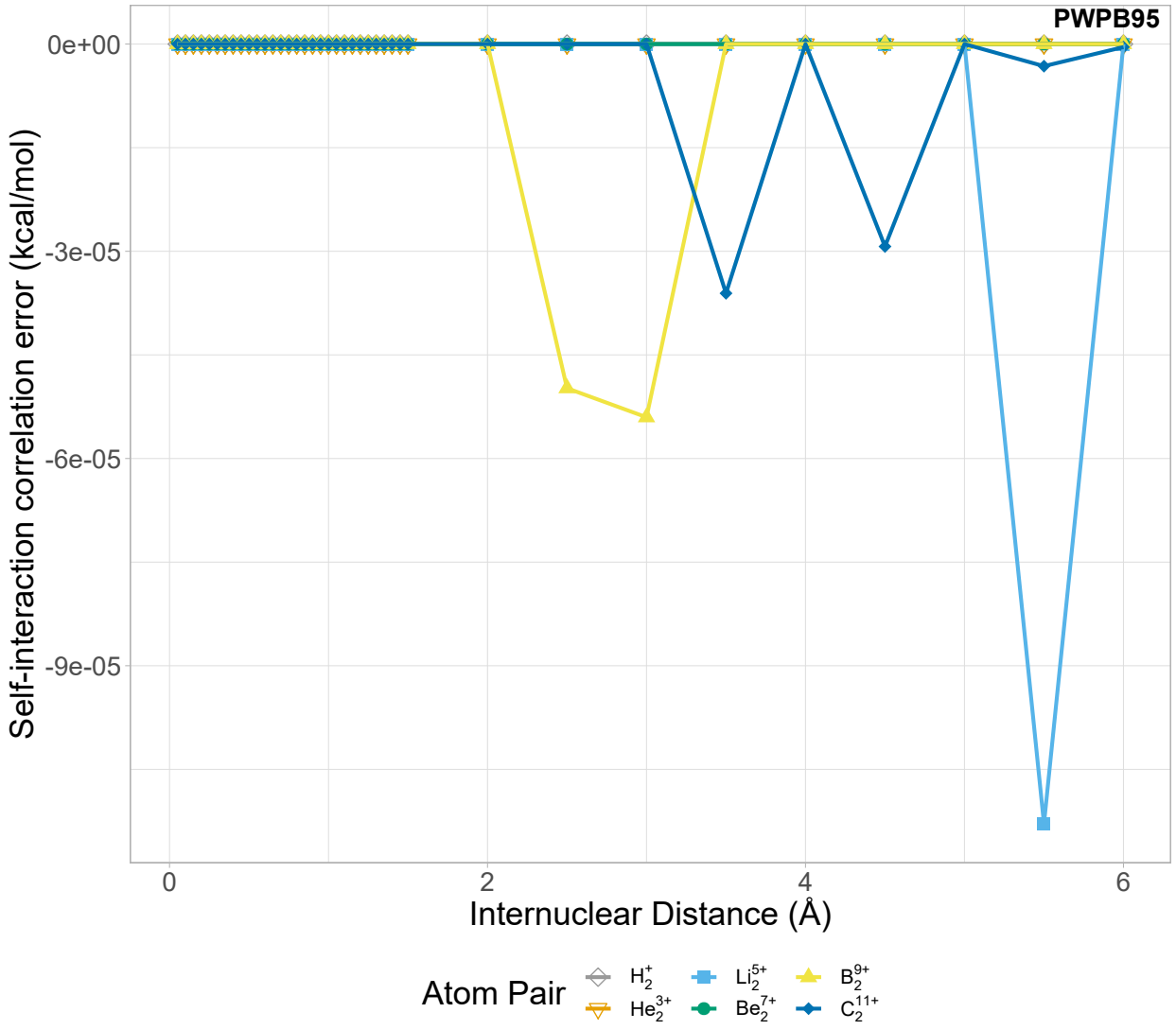

Figure S92: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for PWPB95. Formally this DFA is self-correlation free, any deviations from this is likely computational noise.

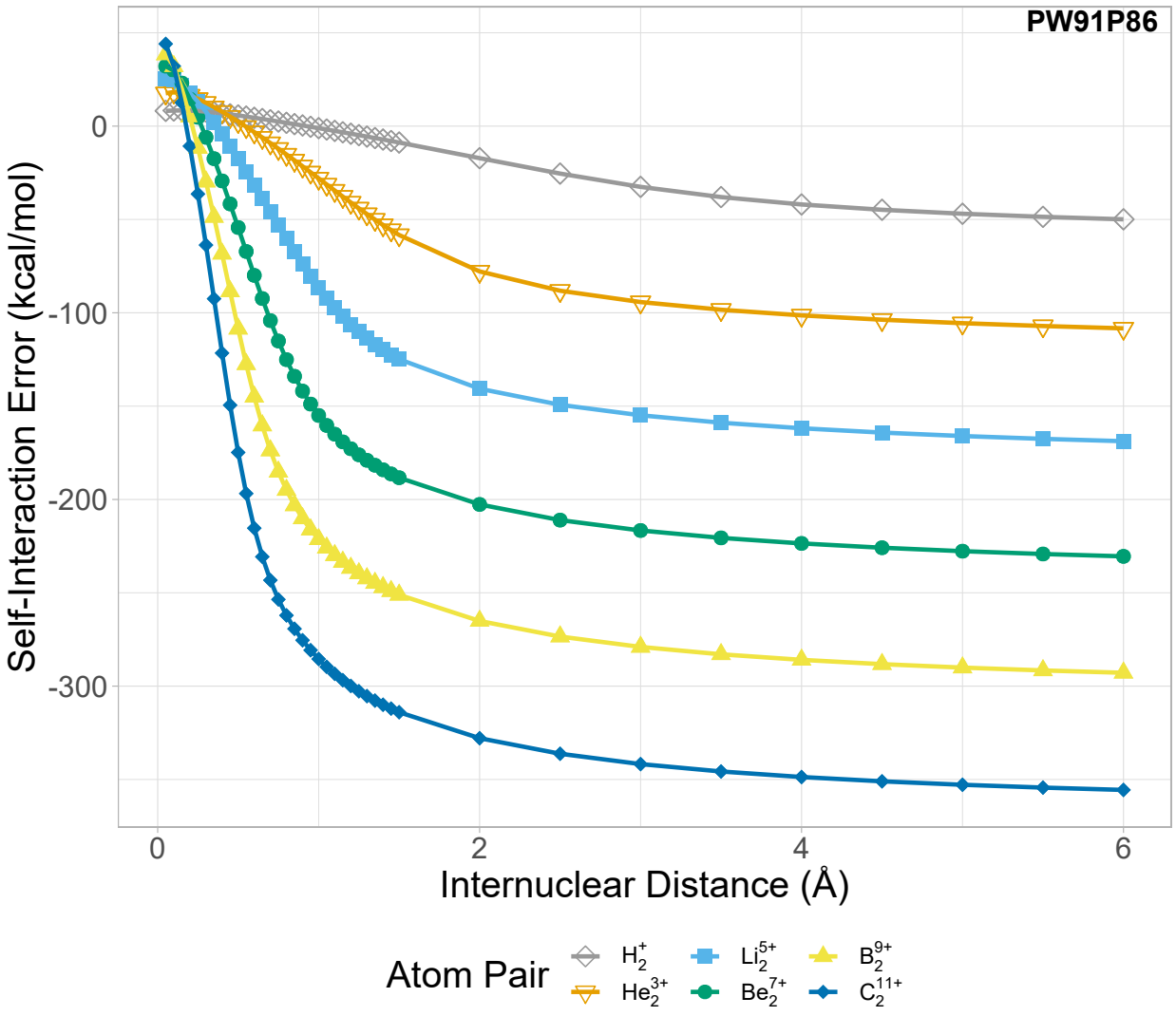

Figure S93: Self-interaction error vs internuclear distance for incremental homonuclear pairs for PW91P86.

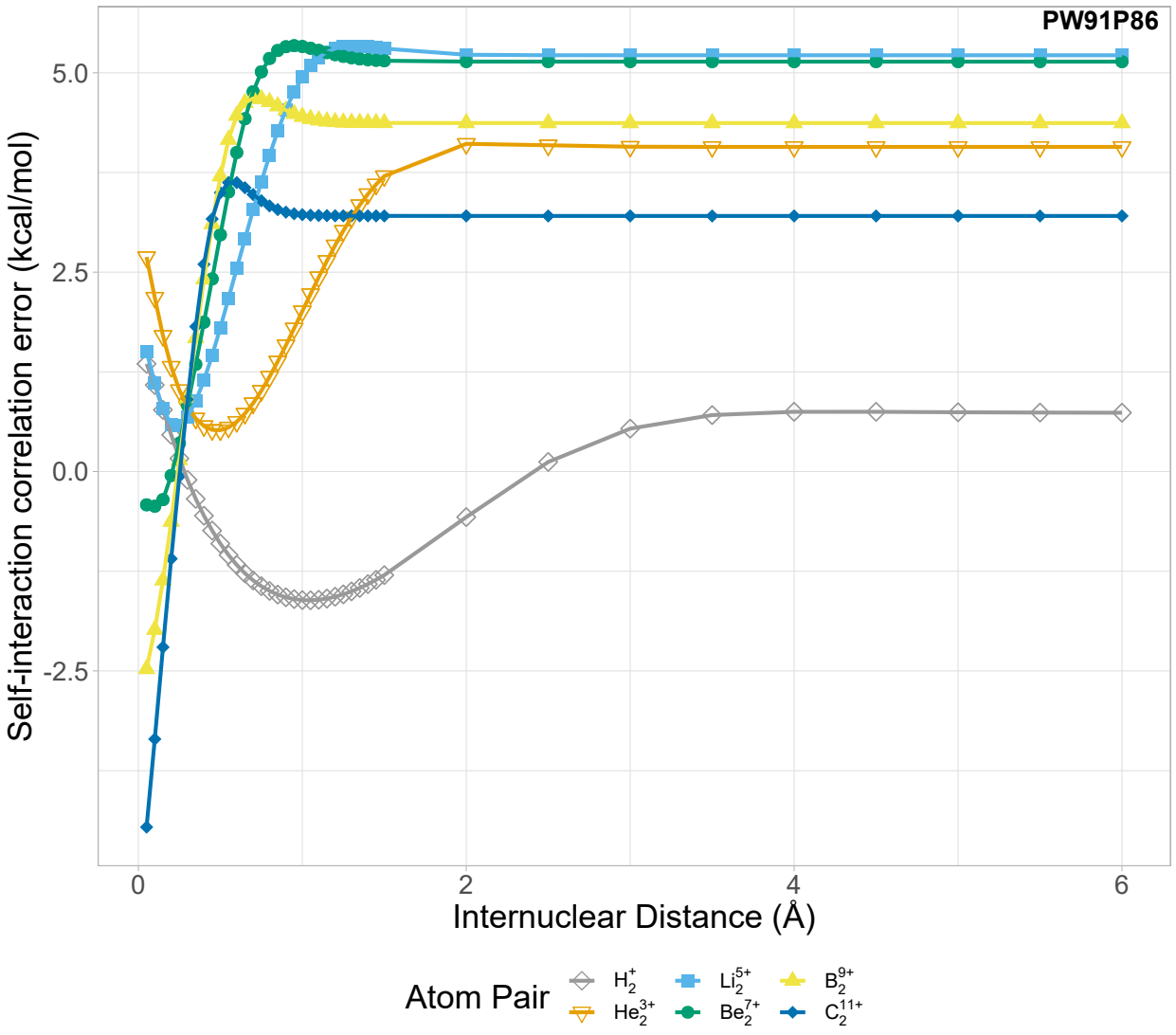

Figure S94: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for PW91P86.

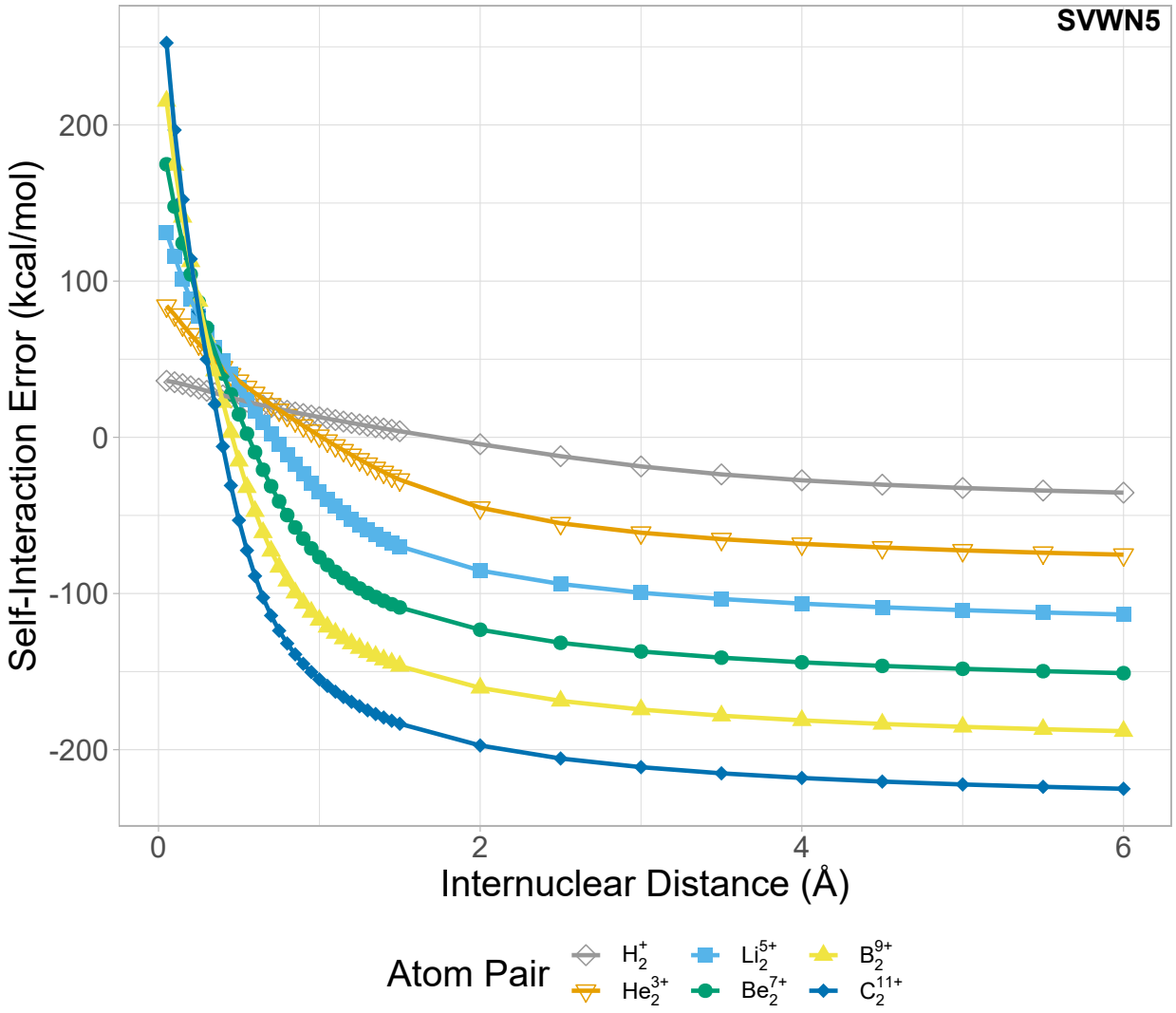

Figure S95: Self-interaction error vs internuclear distance for incremental homonuclear pairs for SVWN5.

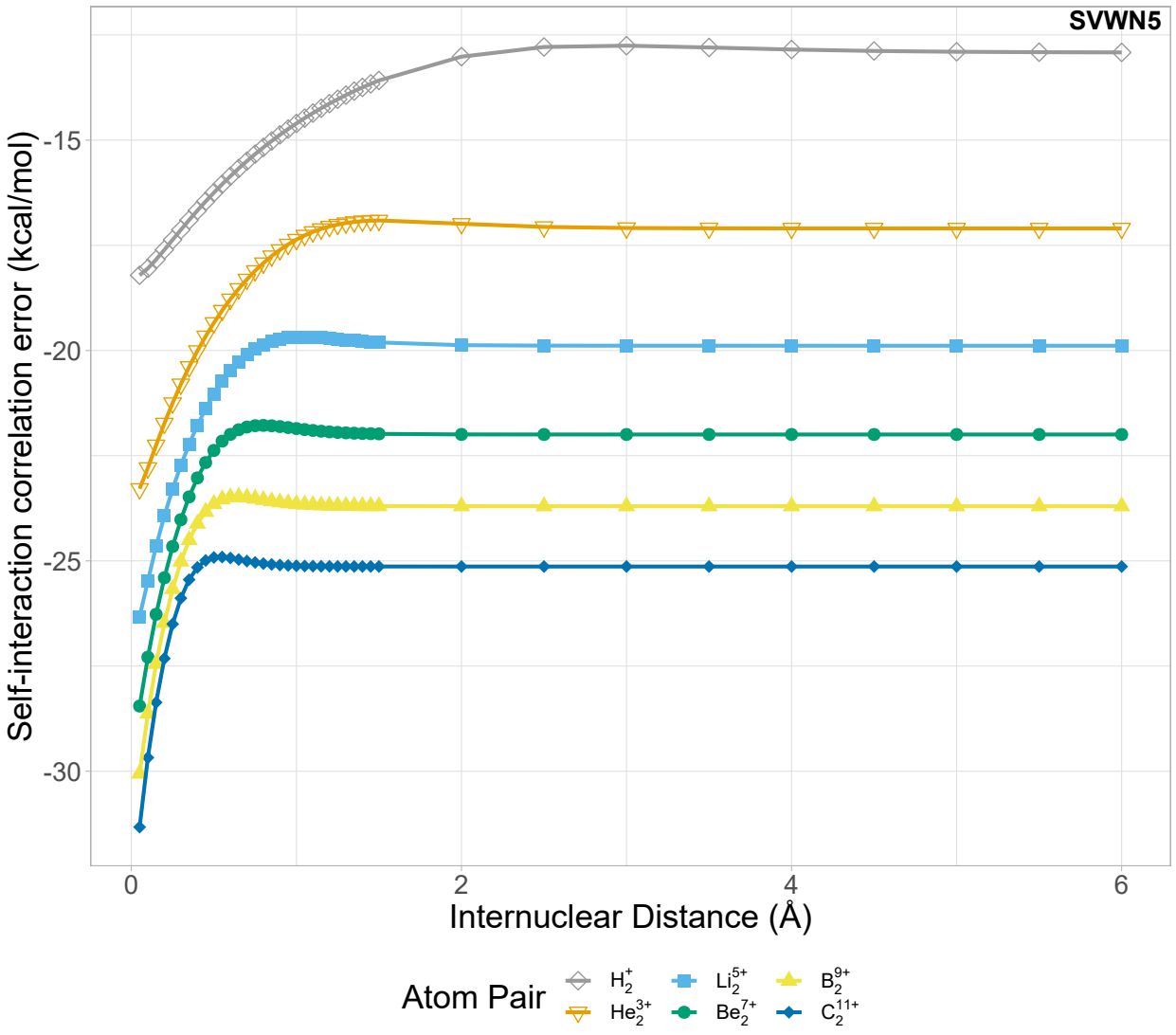

Figure S96: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for SVWN5.

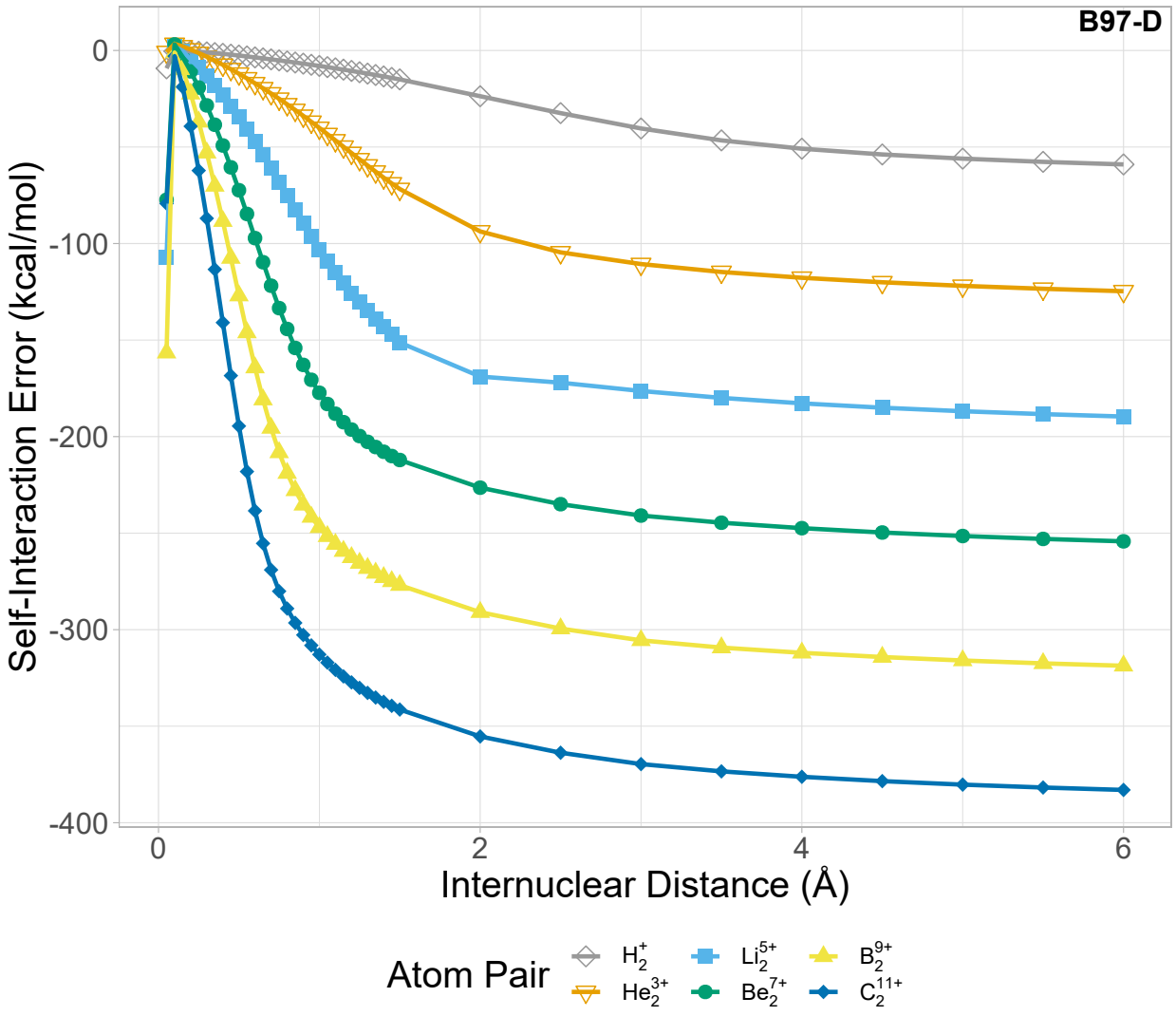

Figure S97: Self-interaction error vs internuclear distance for incremental homonuclear pairs for B97-D.

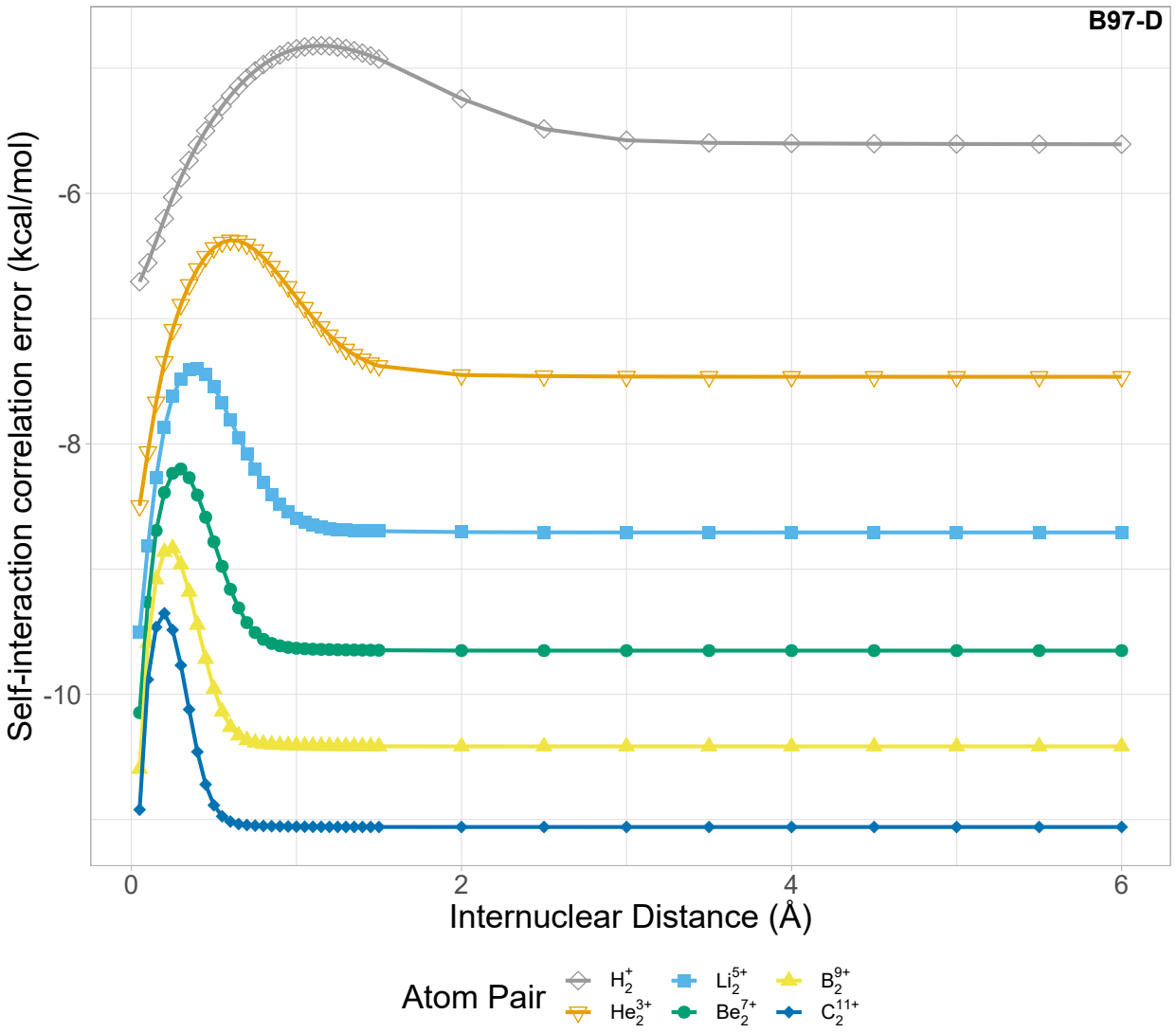

Figure S98: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for B97-D.

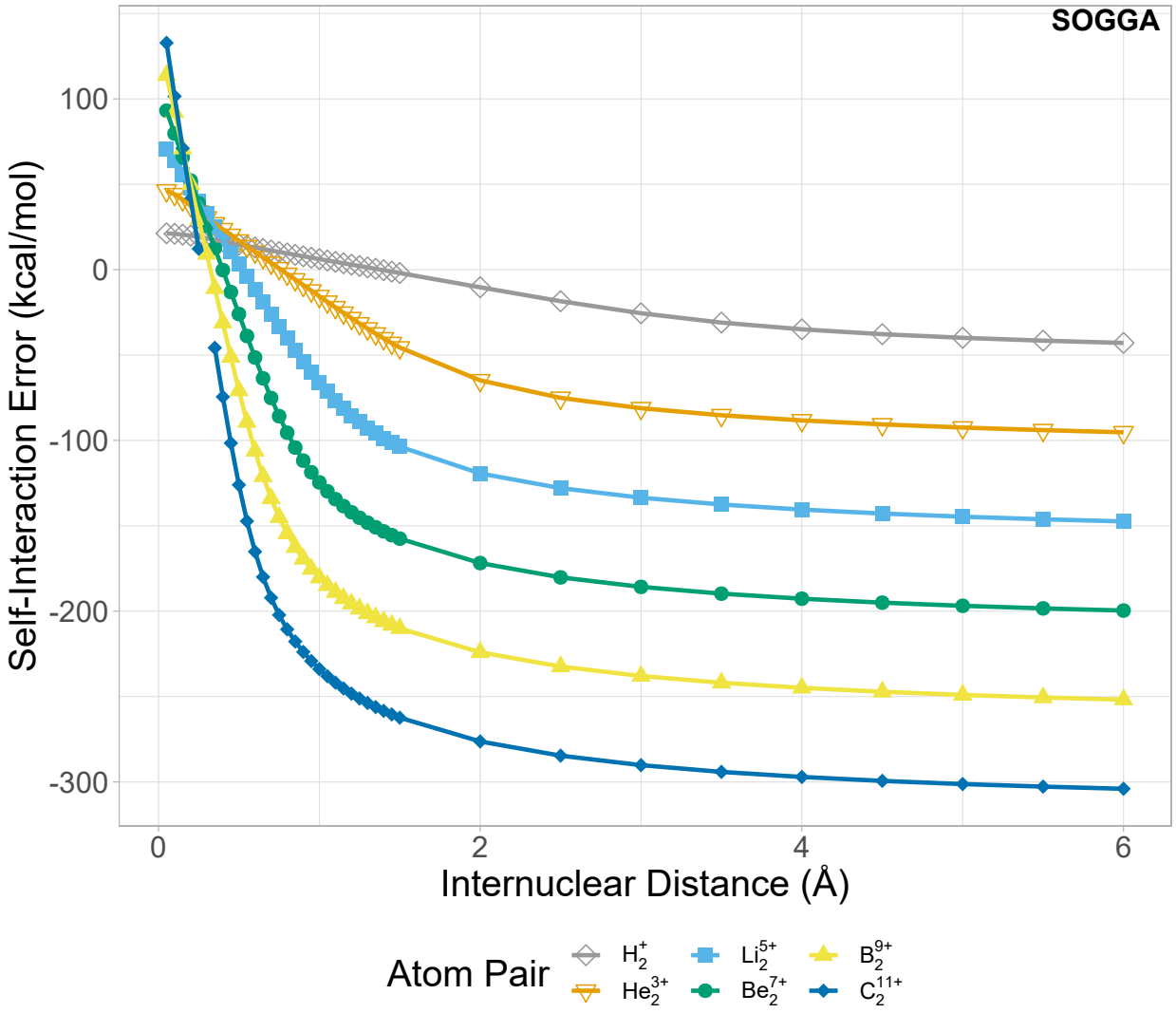

Figure S99: Self-interaction error vs internuclear distance for incremental homonuclear pairs for SOGGA.

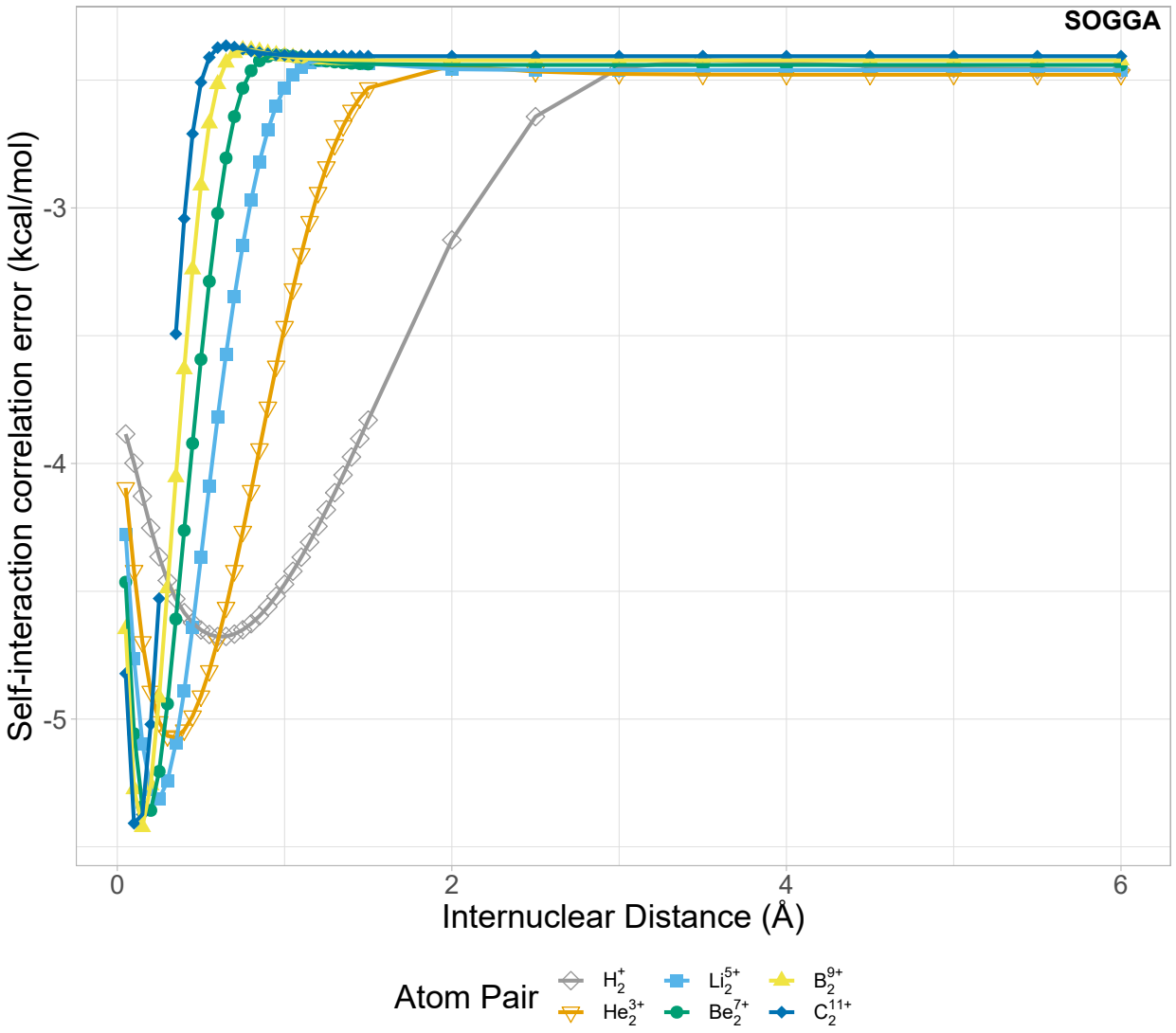

Figure S100: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for SOGGA.

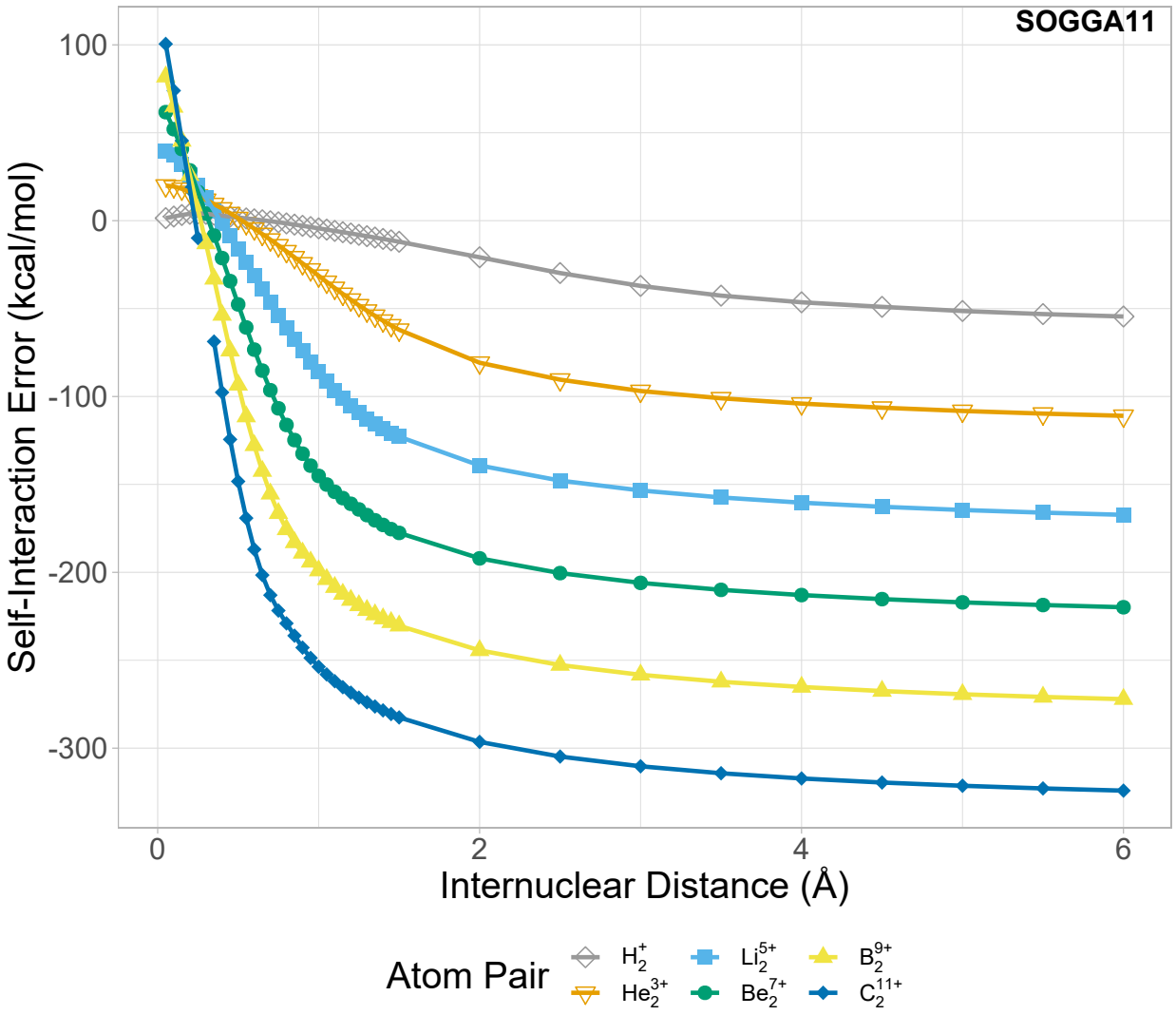

Figure S101: Self-interaction error vs internuclear distance for incremental homonuclear pairs for SOGGA11.

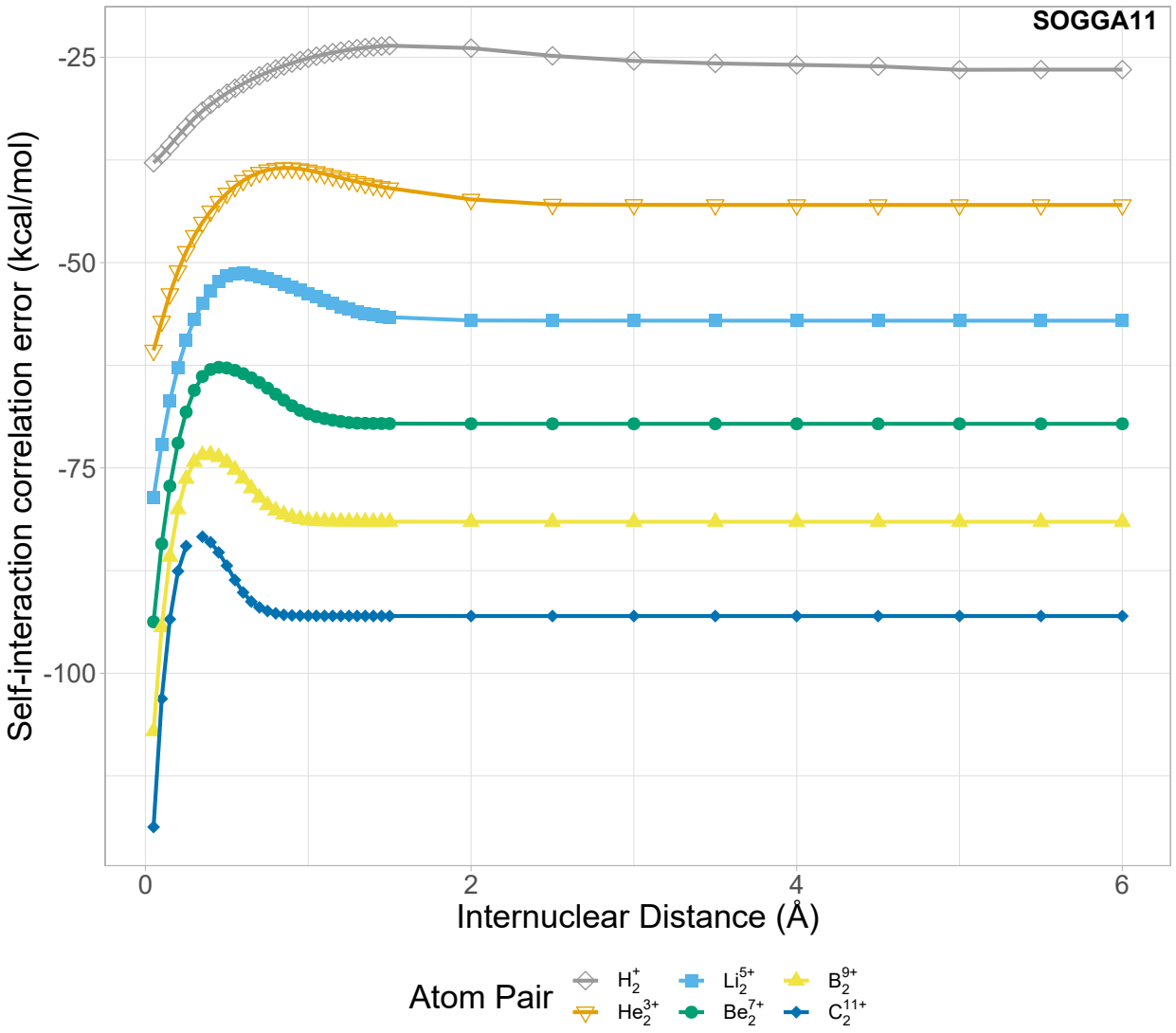

Figure S102: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for SOGGA11.

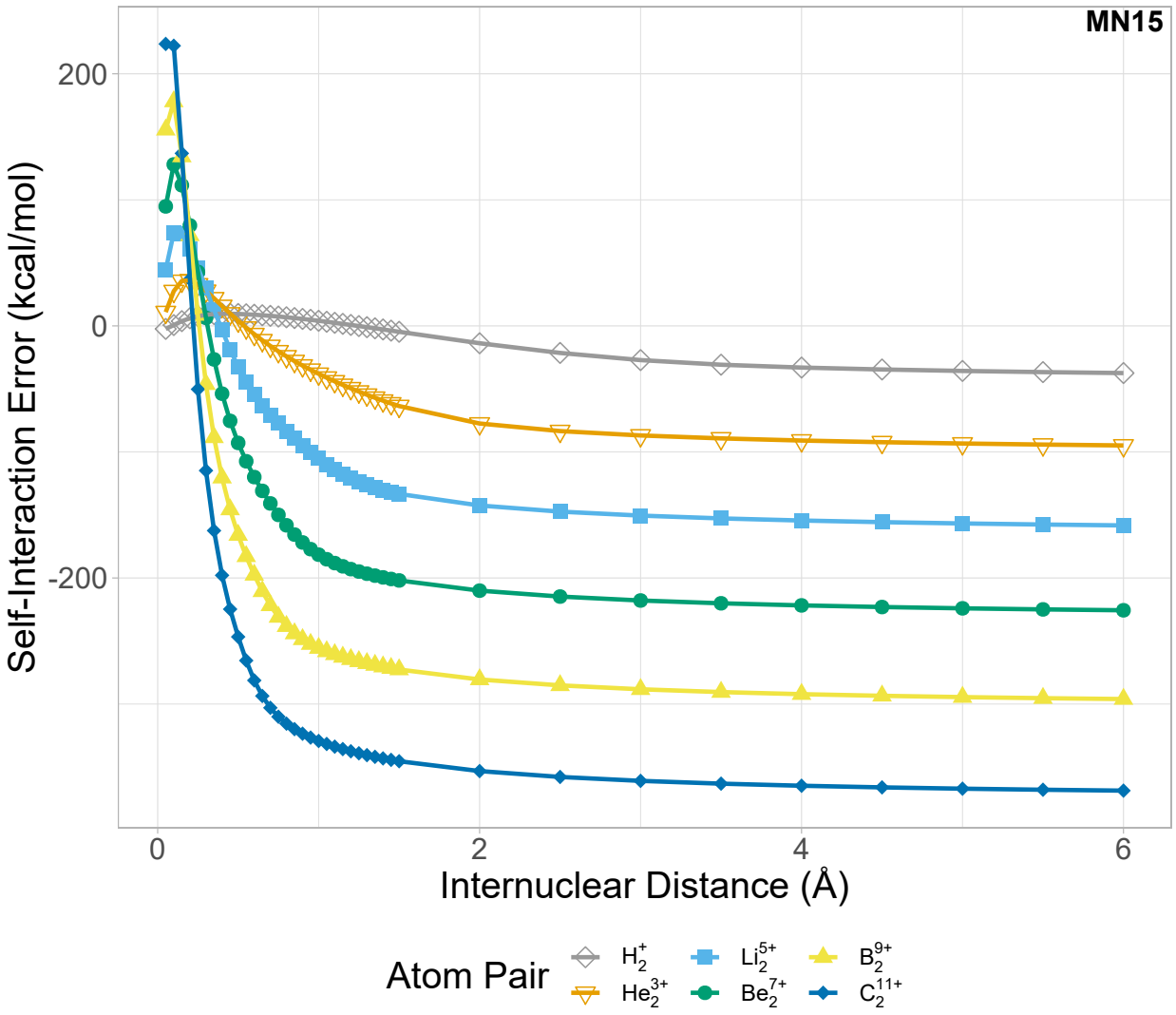

Figure S103: Self-interaction error vs internuclear distance for incremental homonuclear pairs for MN15.

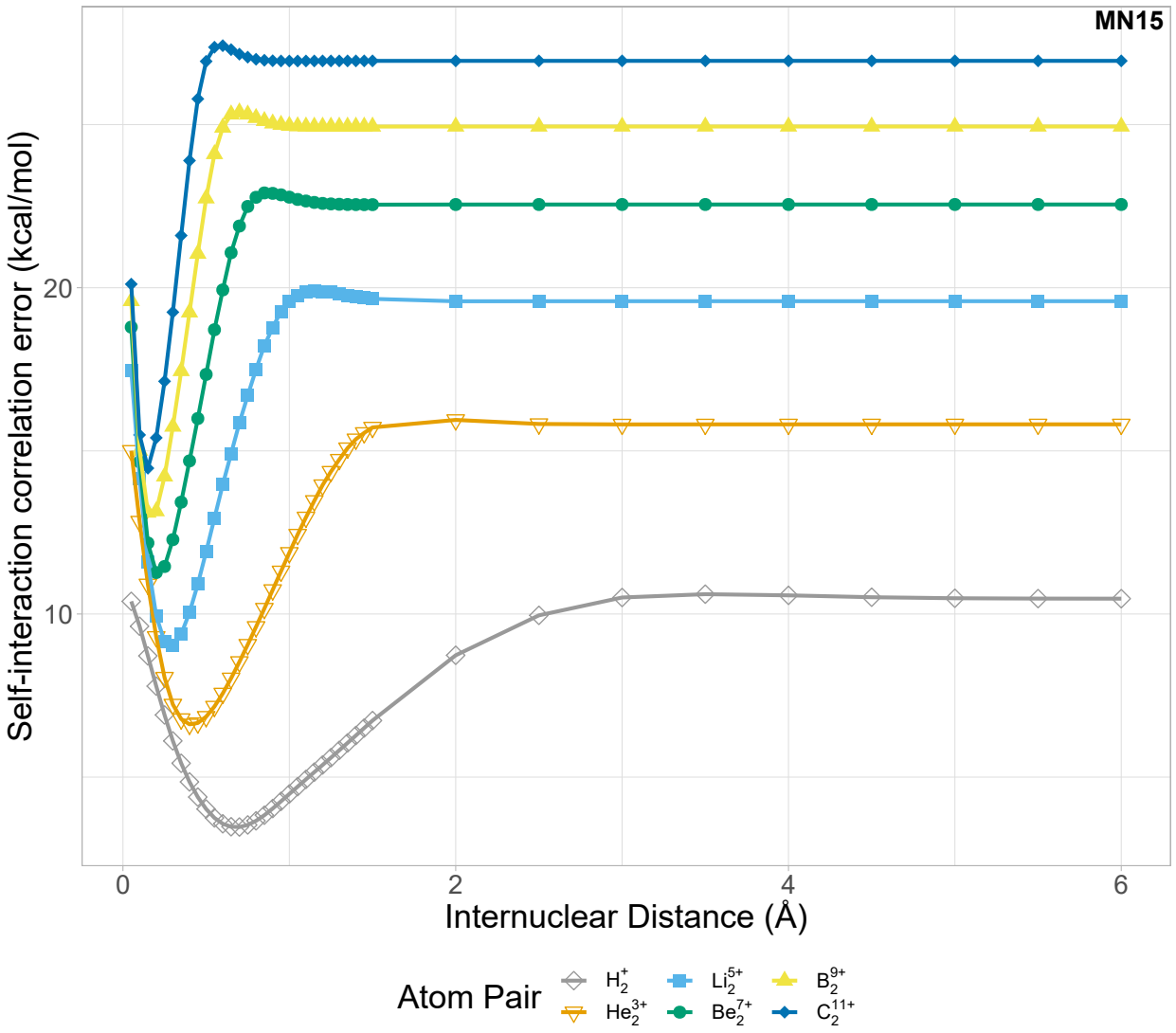

Figure S104: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for MN15.

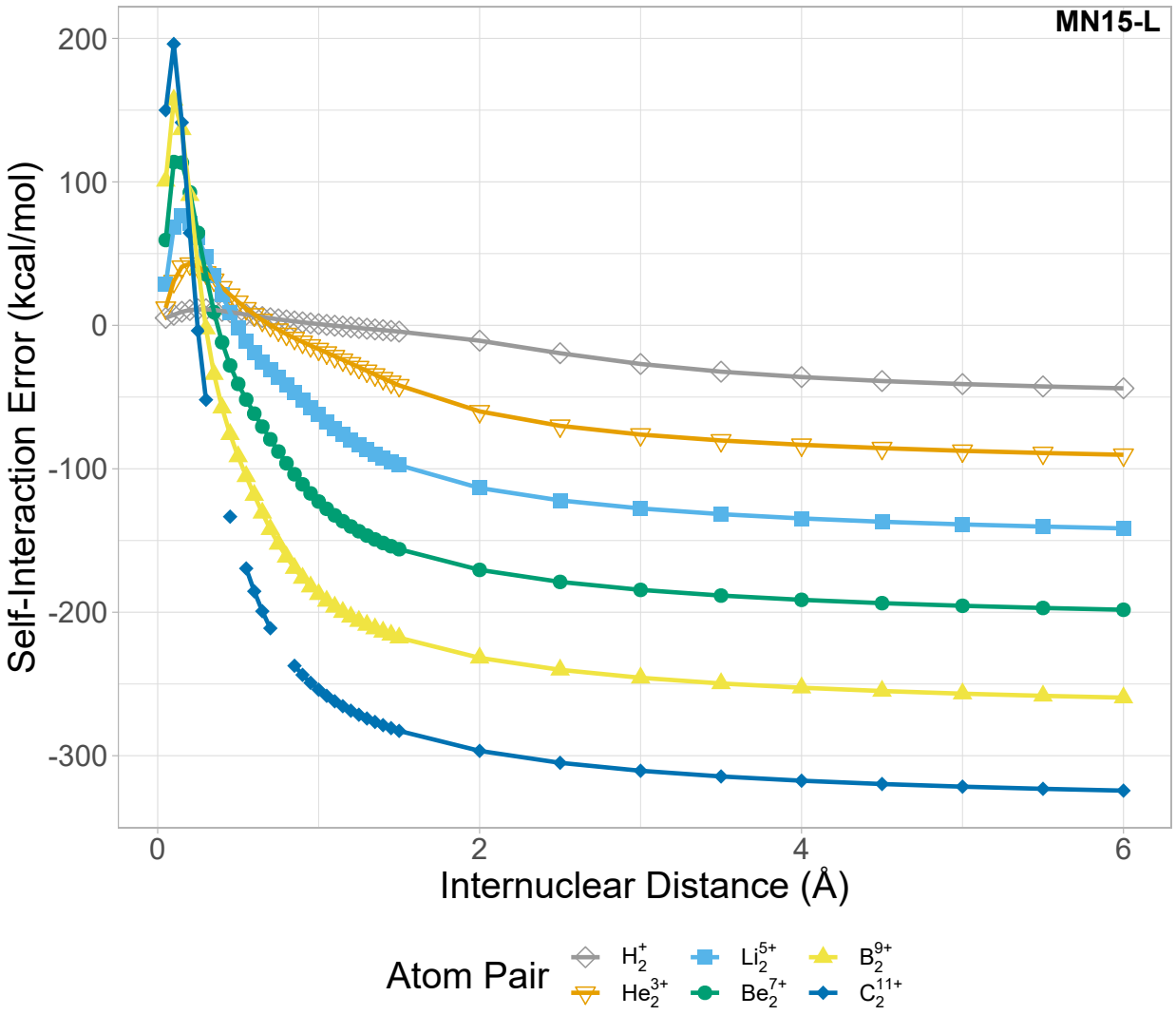

Figure S105: Self-interaction error vs internuclear distance for incremental homonuclear pairs for MN15-L.

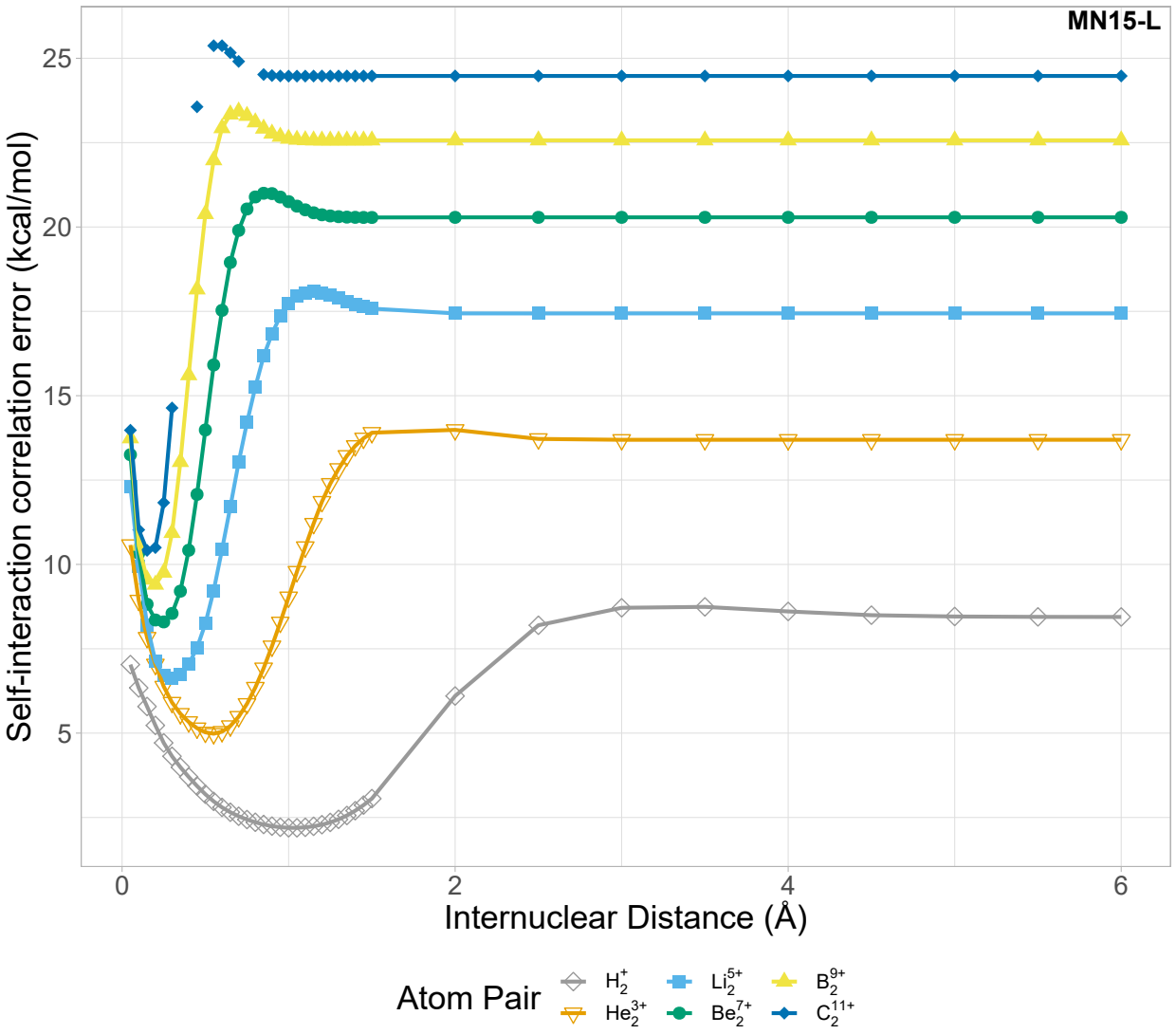

Figure S106: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for MN15-L.

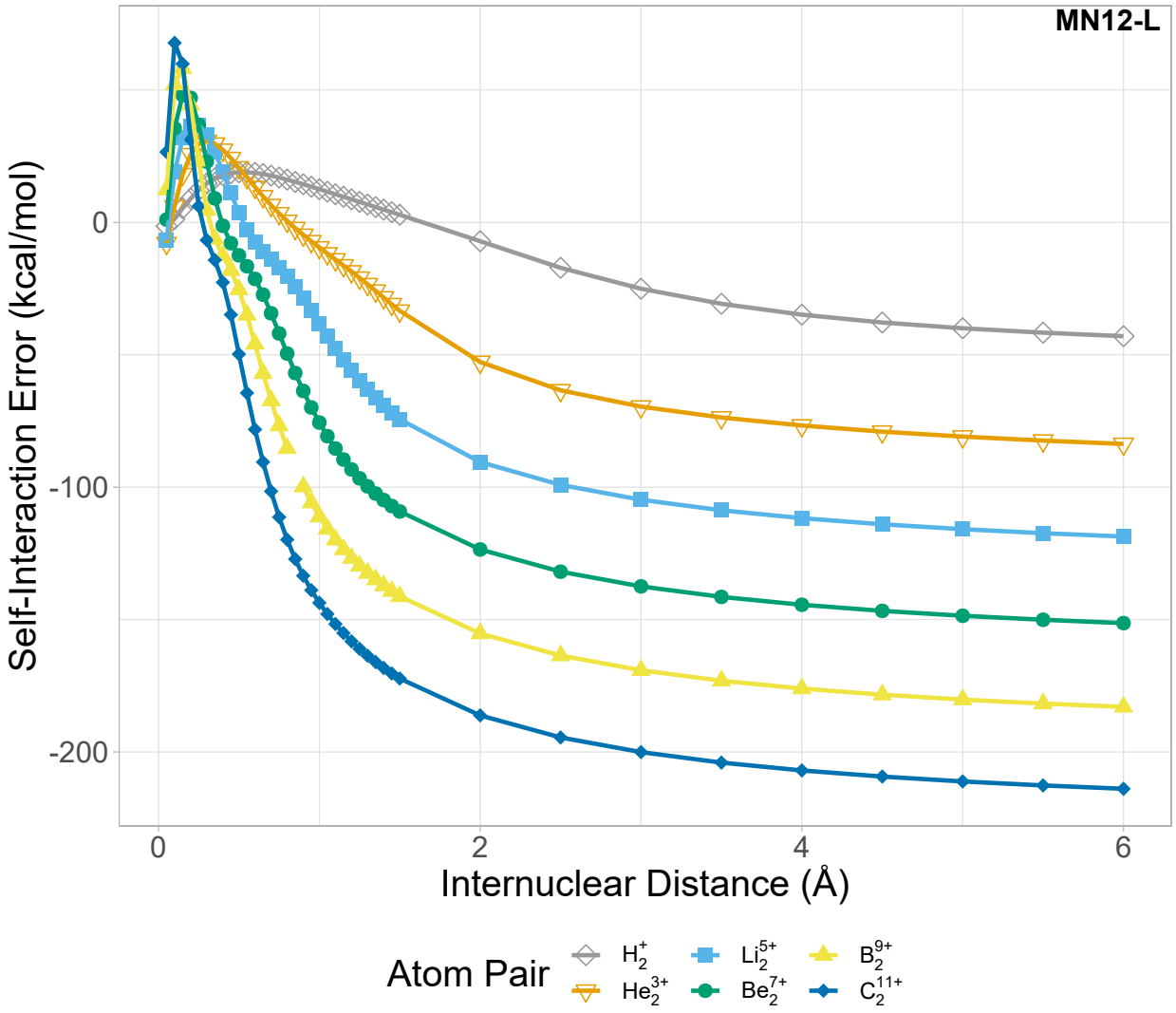

Figure S107: Self-interaction error vs internuclear distance for incremental homonuclear pairs for MN12-L.

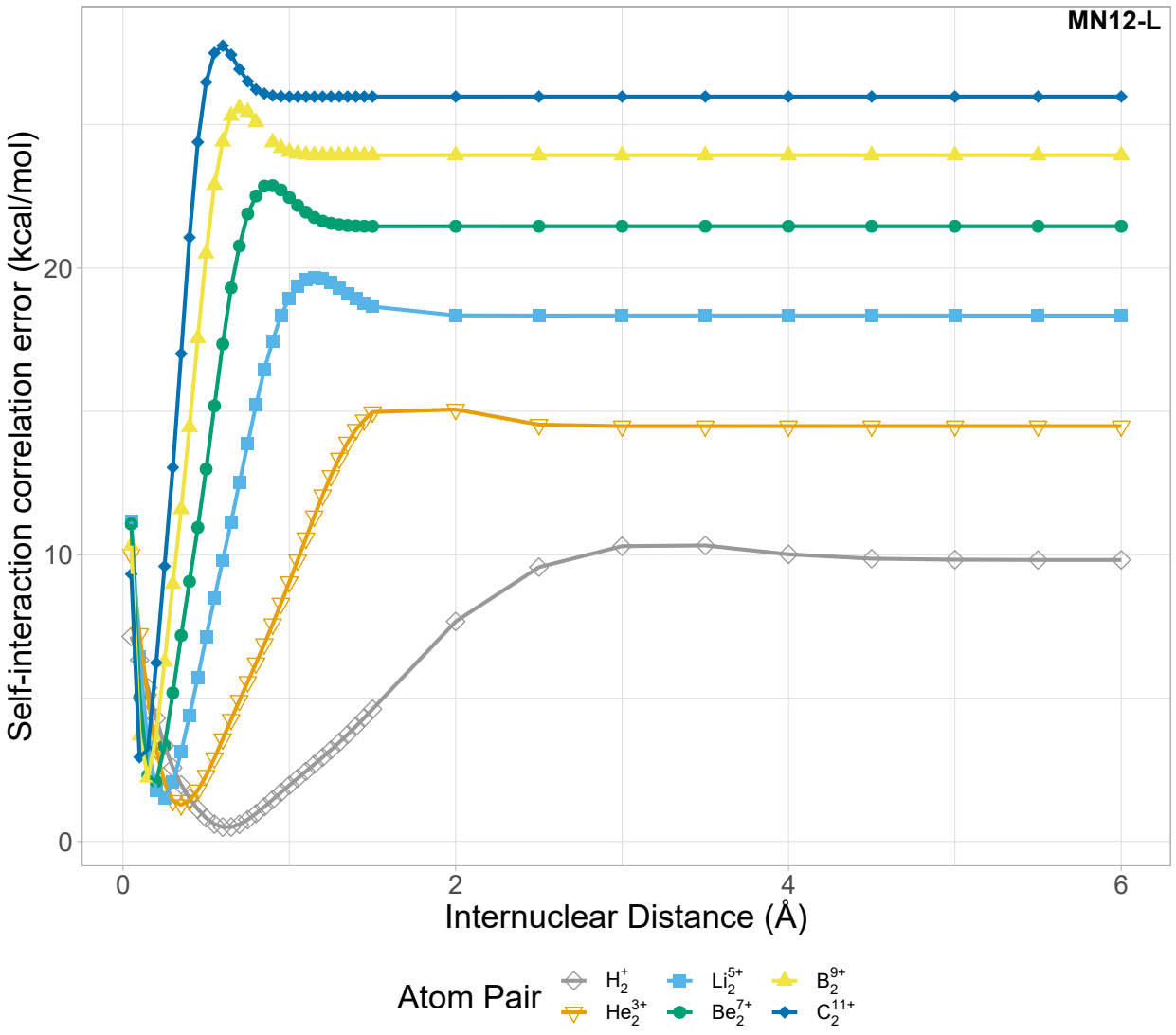

Figure S108: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for MN12-L.

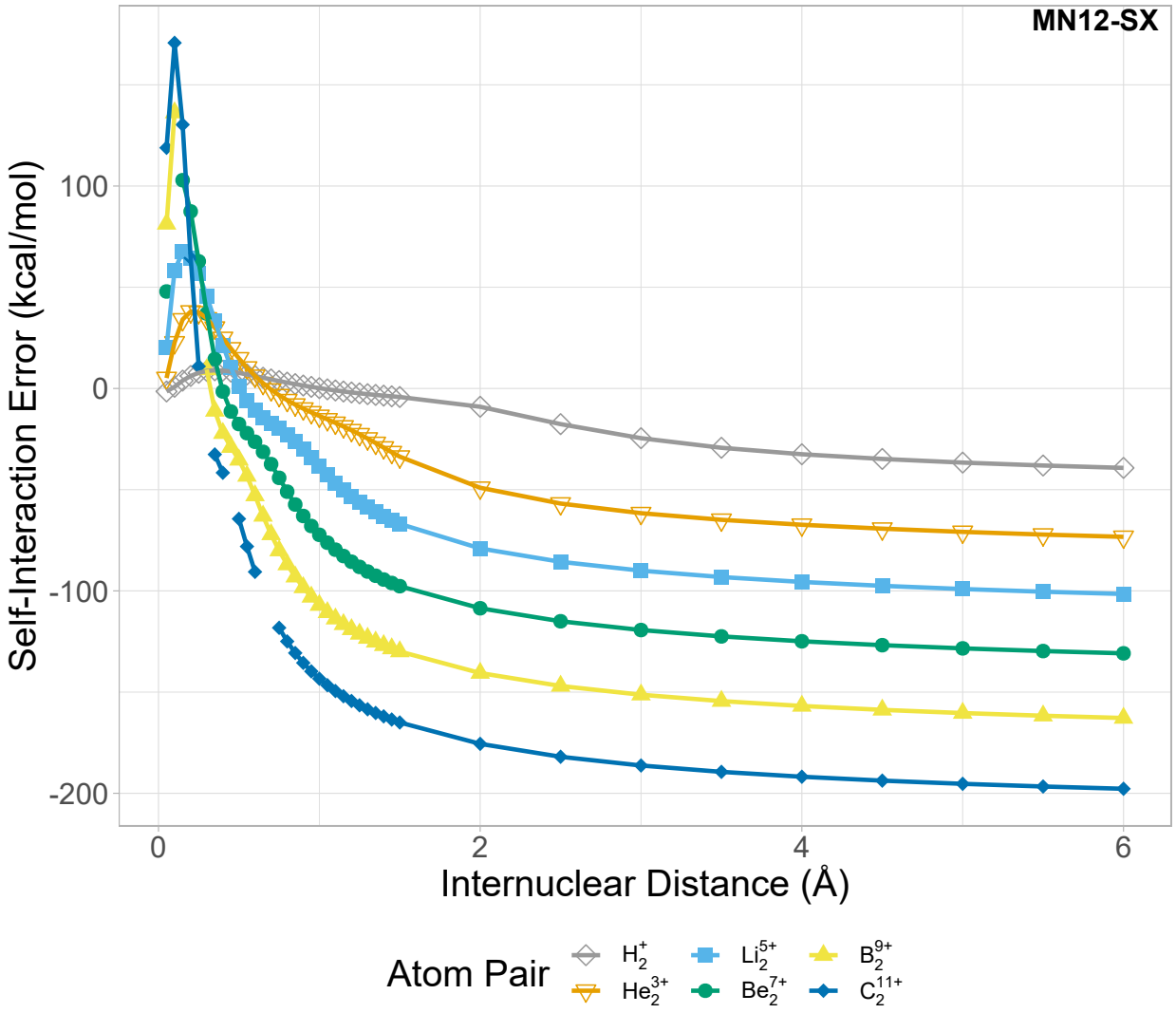

Figure S109: Self-interaction error vs internuclear distance for incremental homonuclear pairs for MN12-SX.

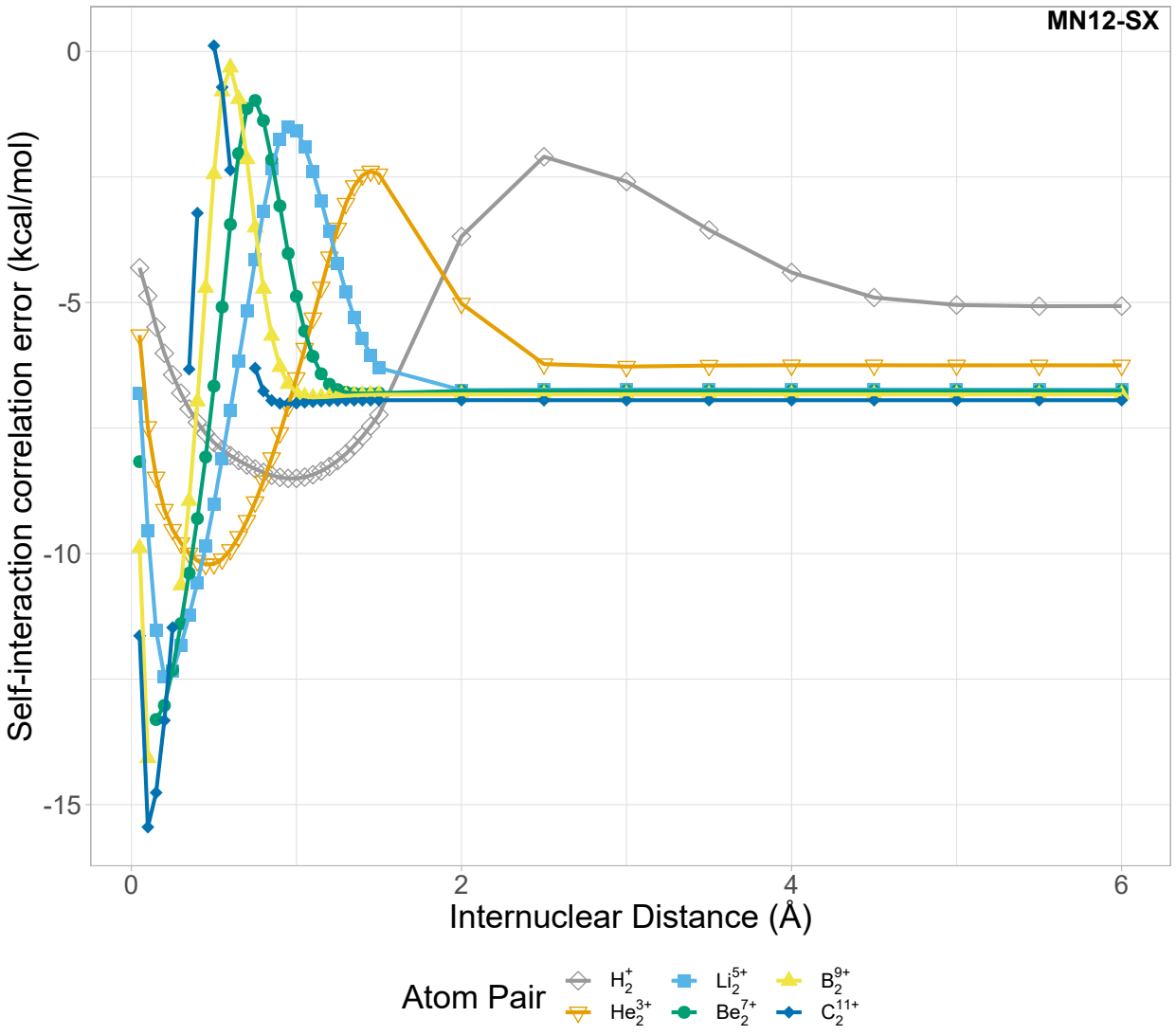

Figure S110: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for MN12-SX.

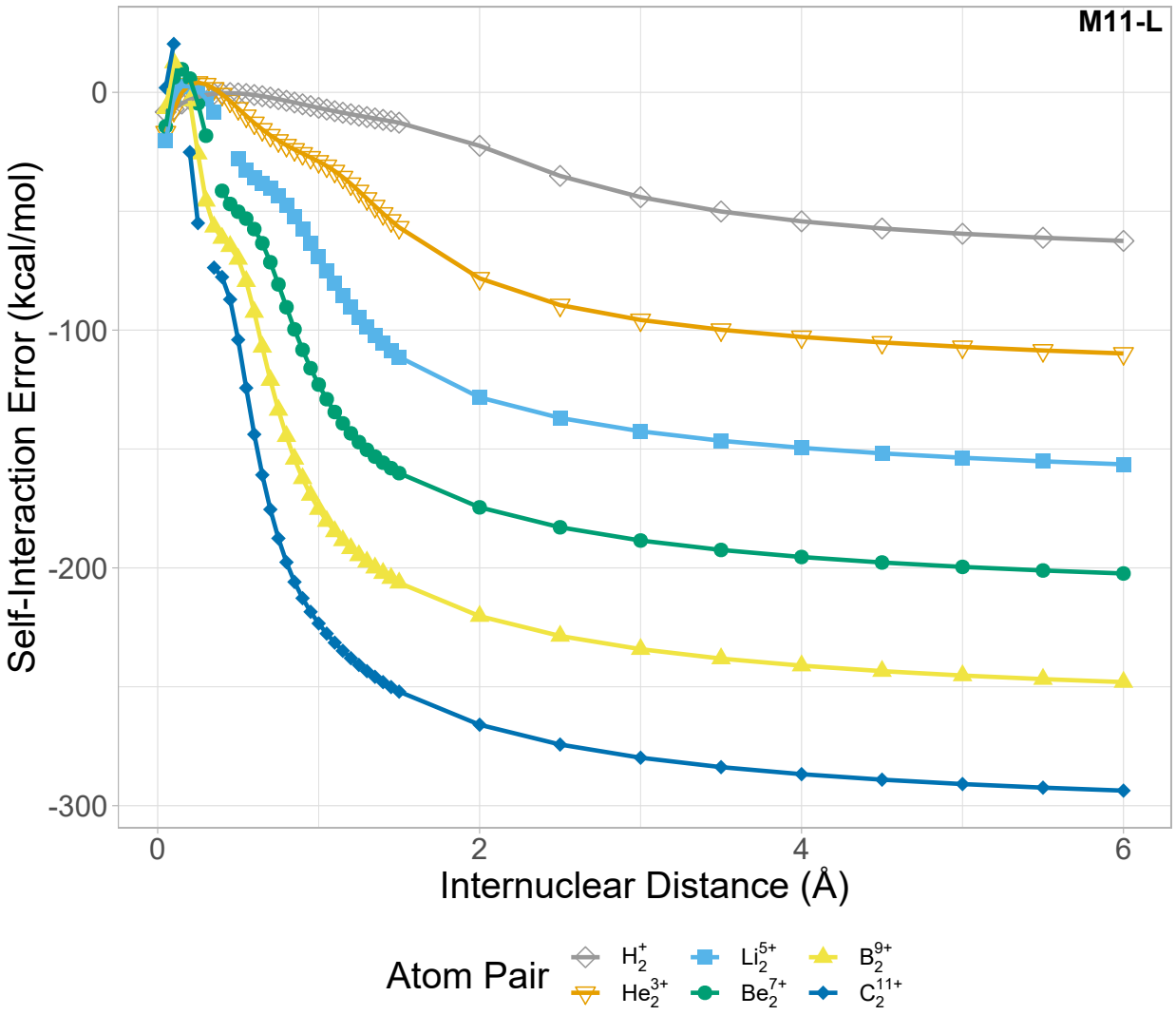

Figure S111: Self-interaction error vs internuclear distance for incremental homonuclear pairs for M11-L.

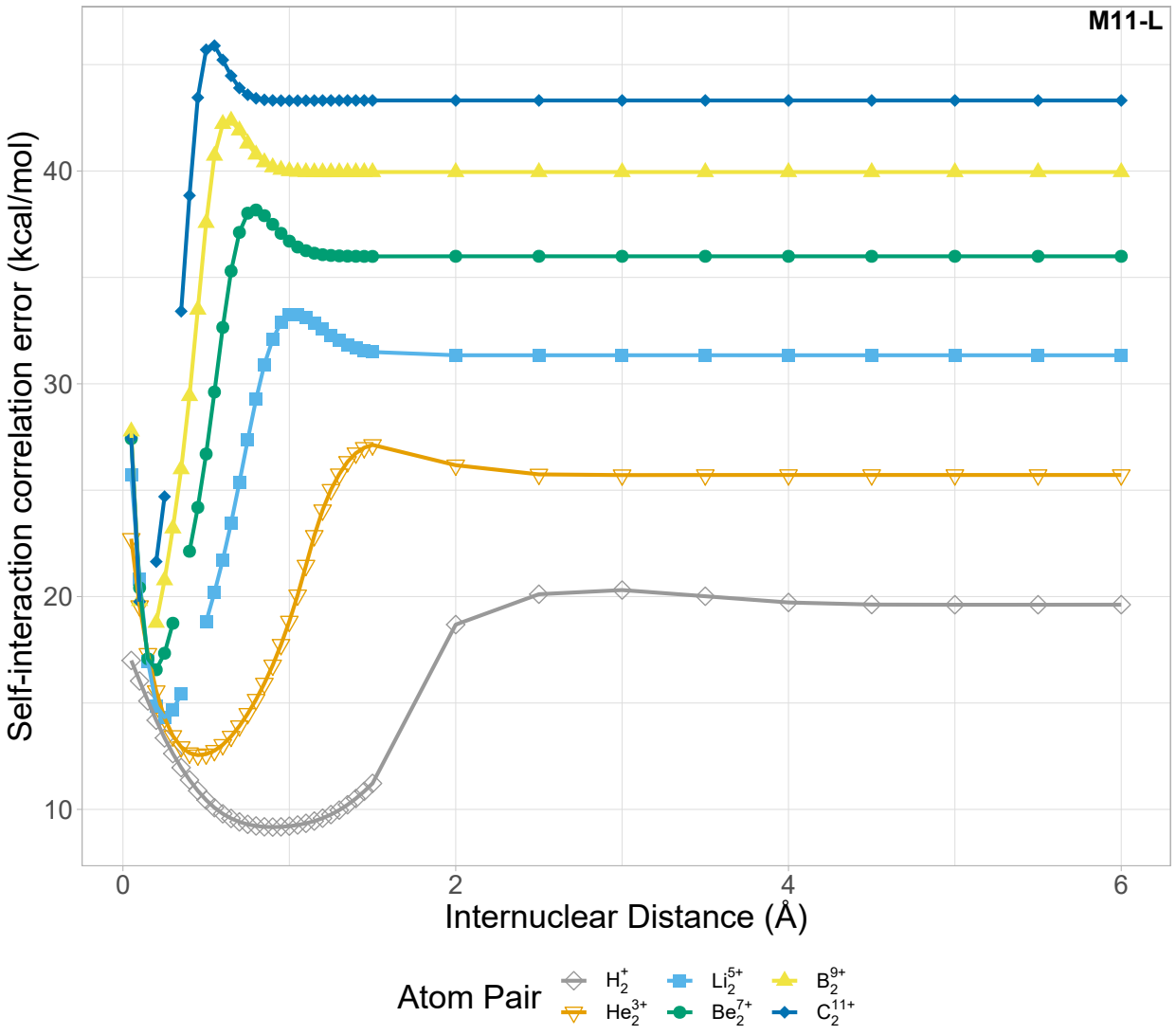

Figure S112: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for M11-L.

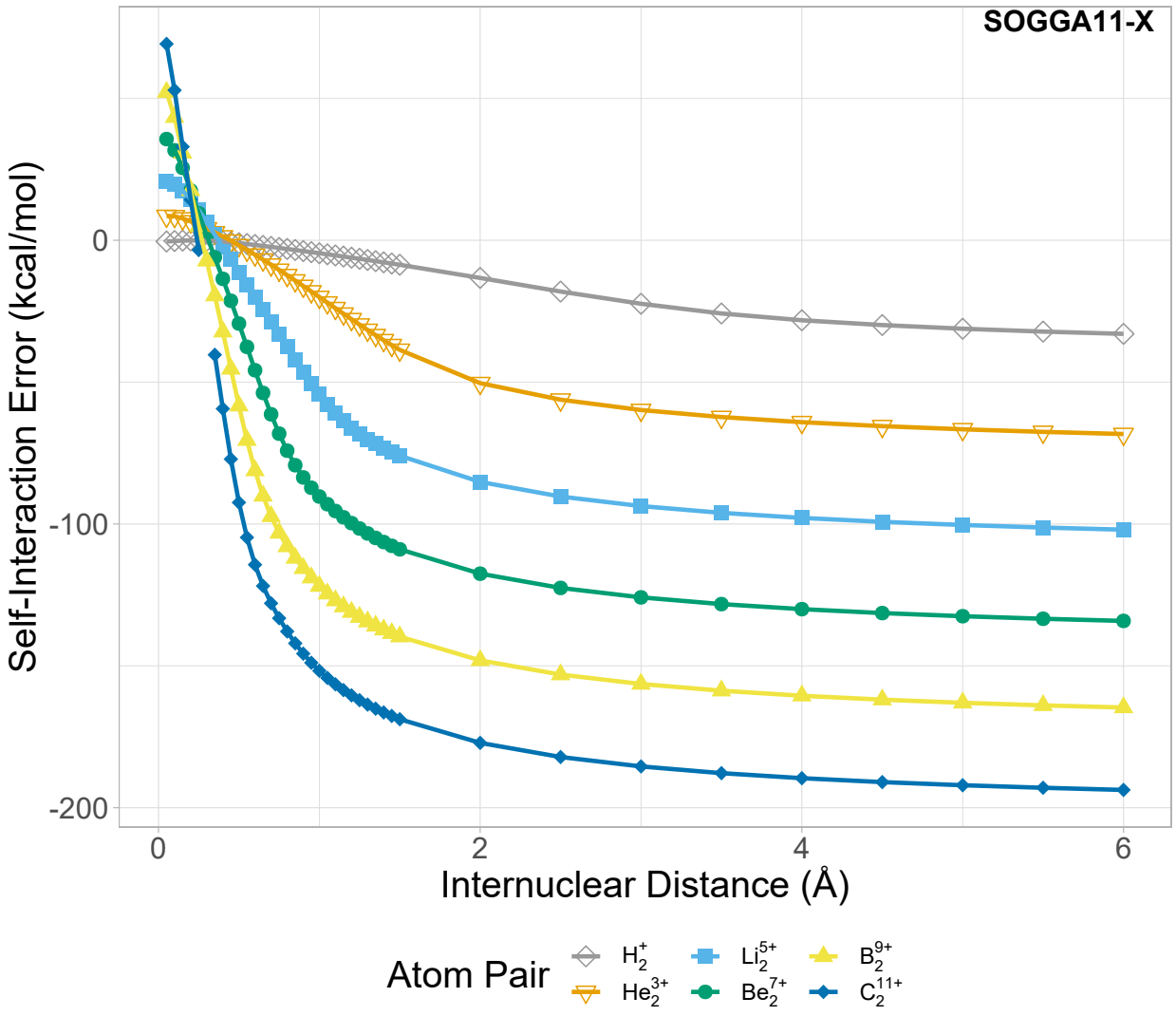

Figure S113: Self-interaction error vs internuclear distance for incremental homonuclear pairs for SOGGA11-X.

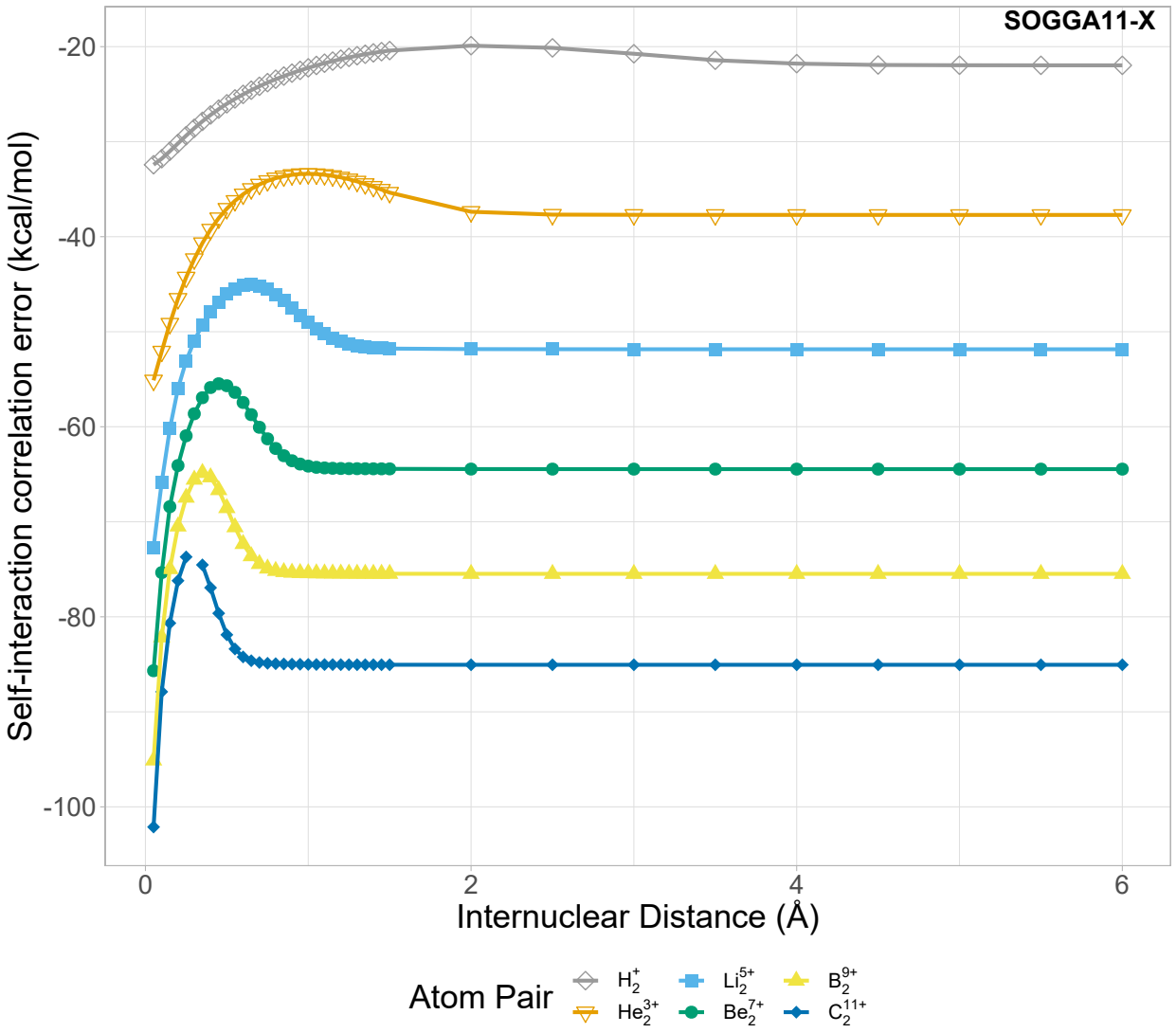

Figure S114: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for SOGGA11-X.

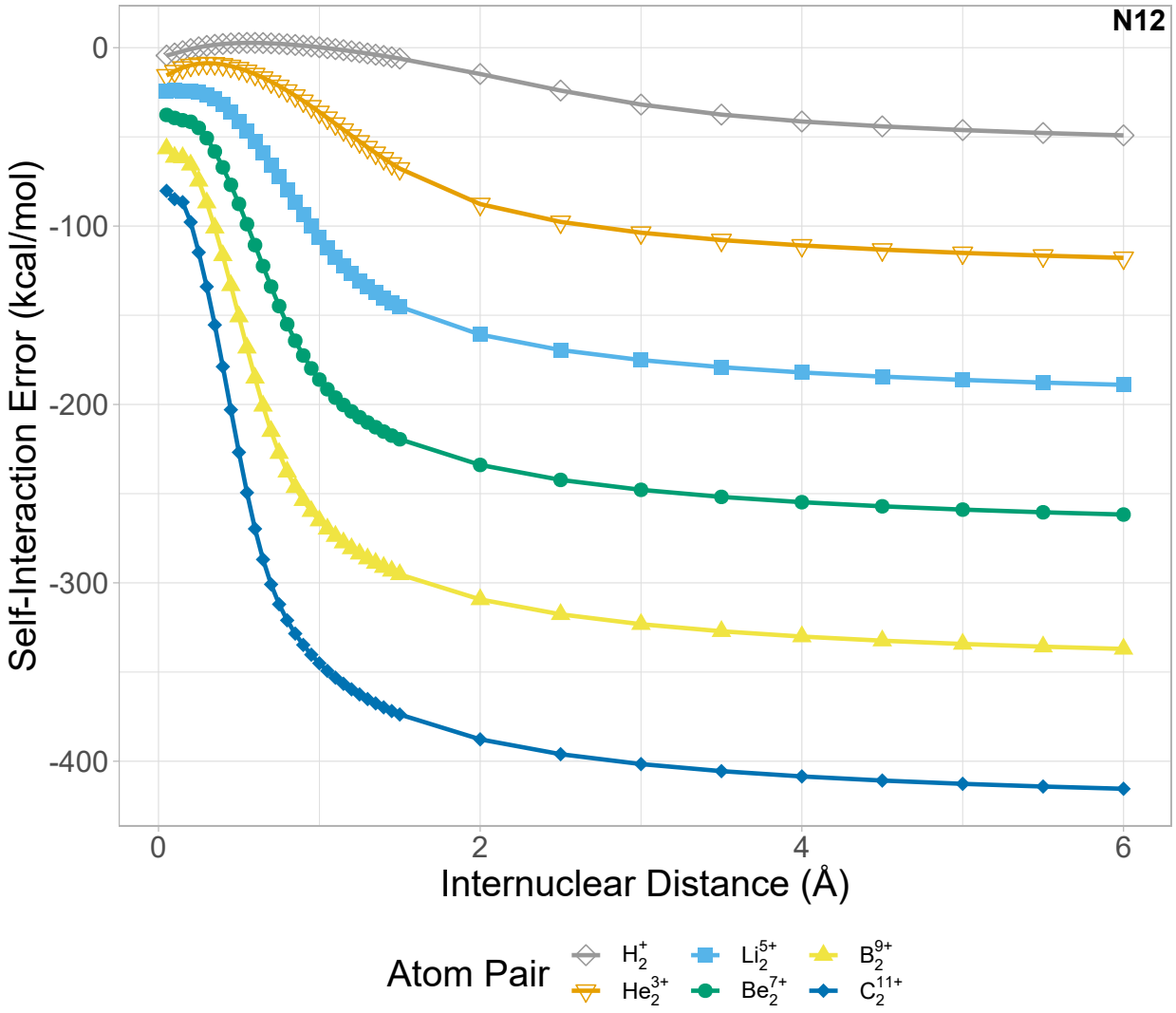

Figure S115: Self-interaction error vs internuclear distance for incremental homonuclear pairs for N12.

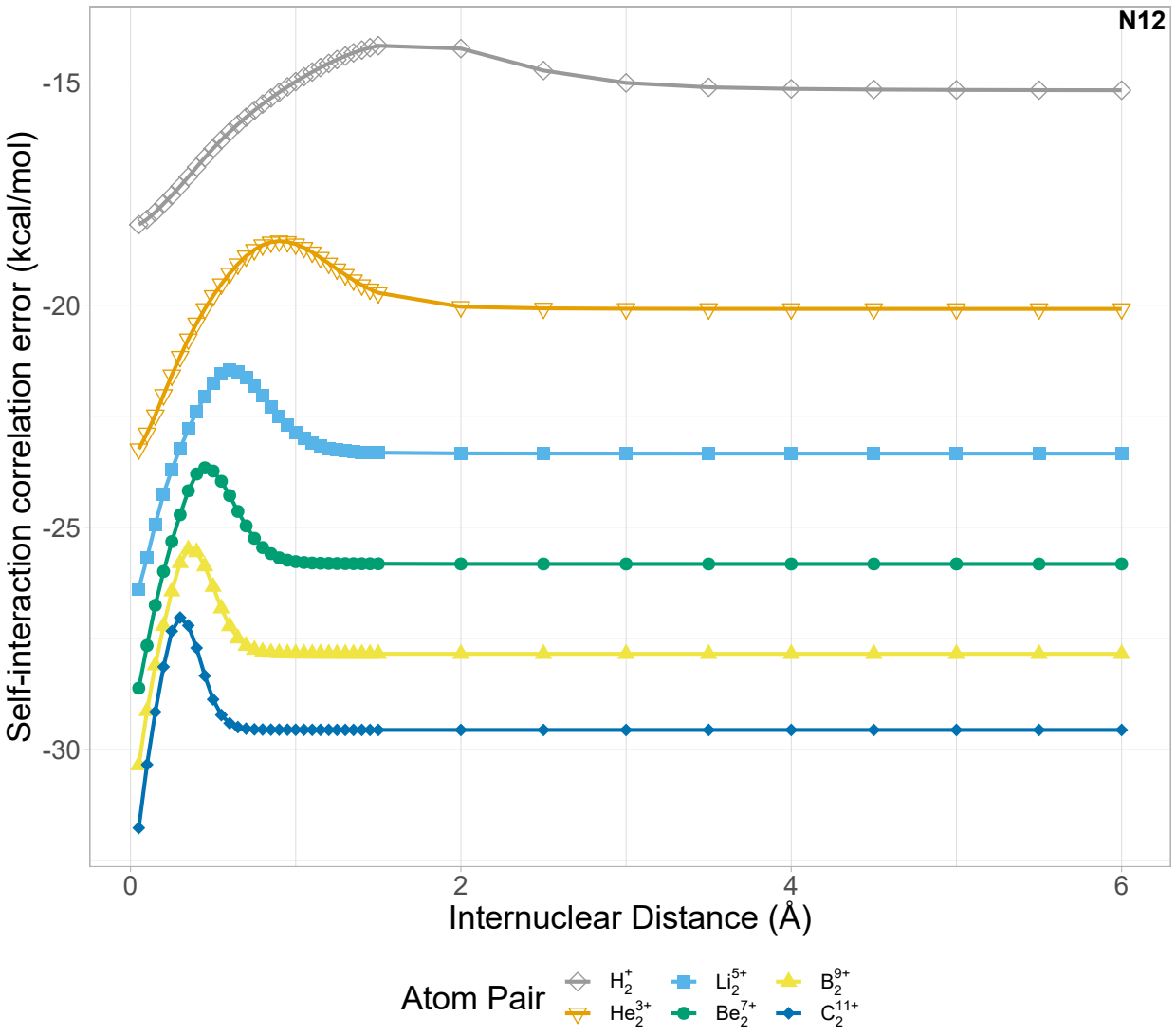

Figure S116: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for N12.

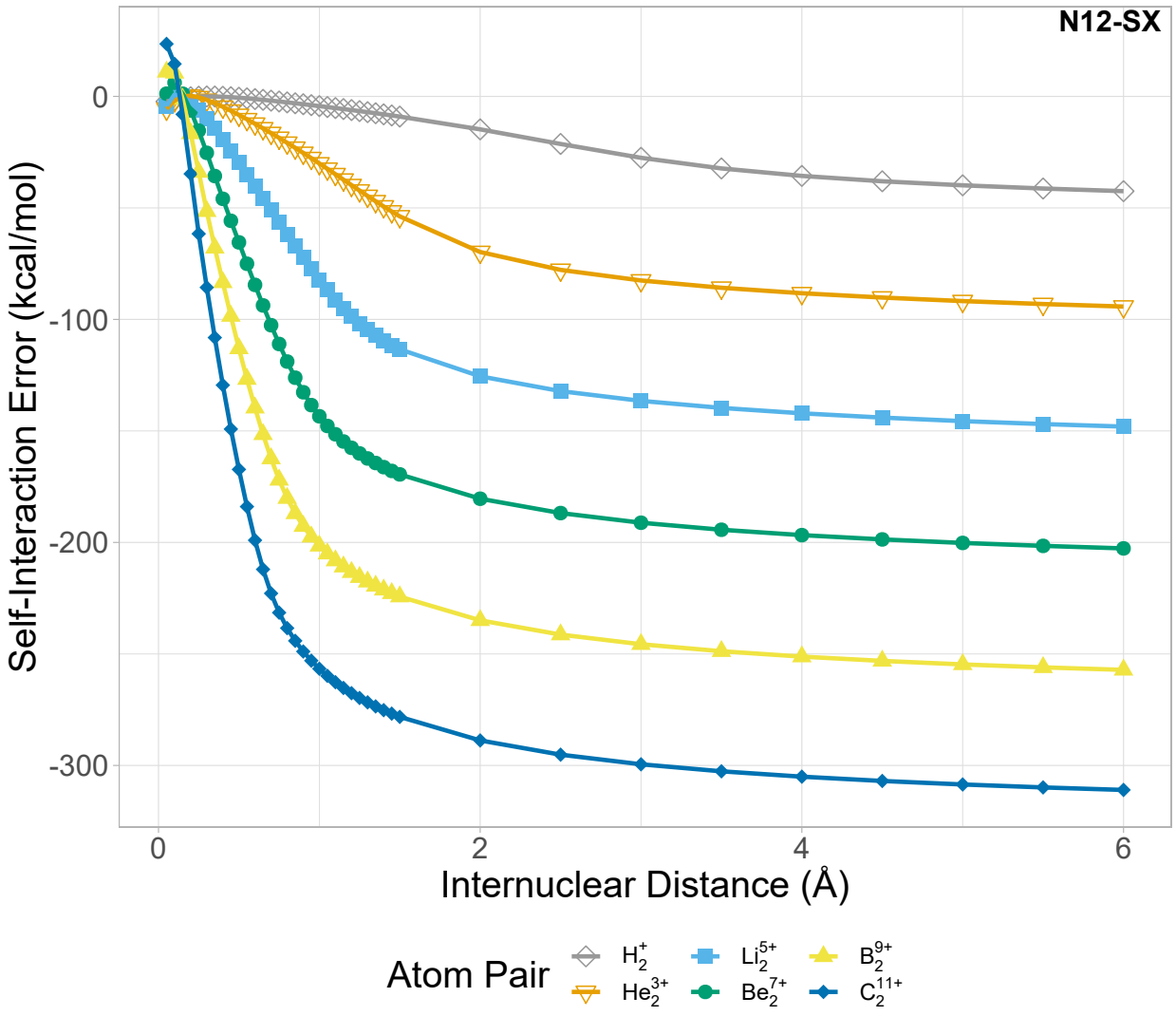

Figure S117: Self-interaction error vs internuclear distance for incremental homonuclear pairs for N12-SX.

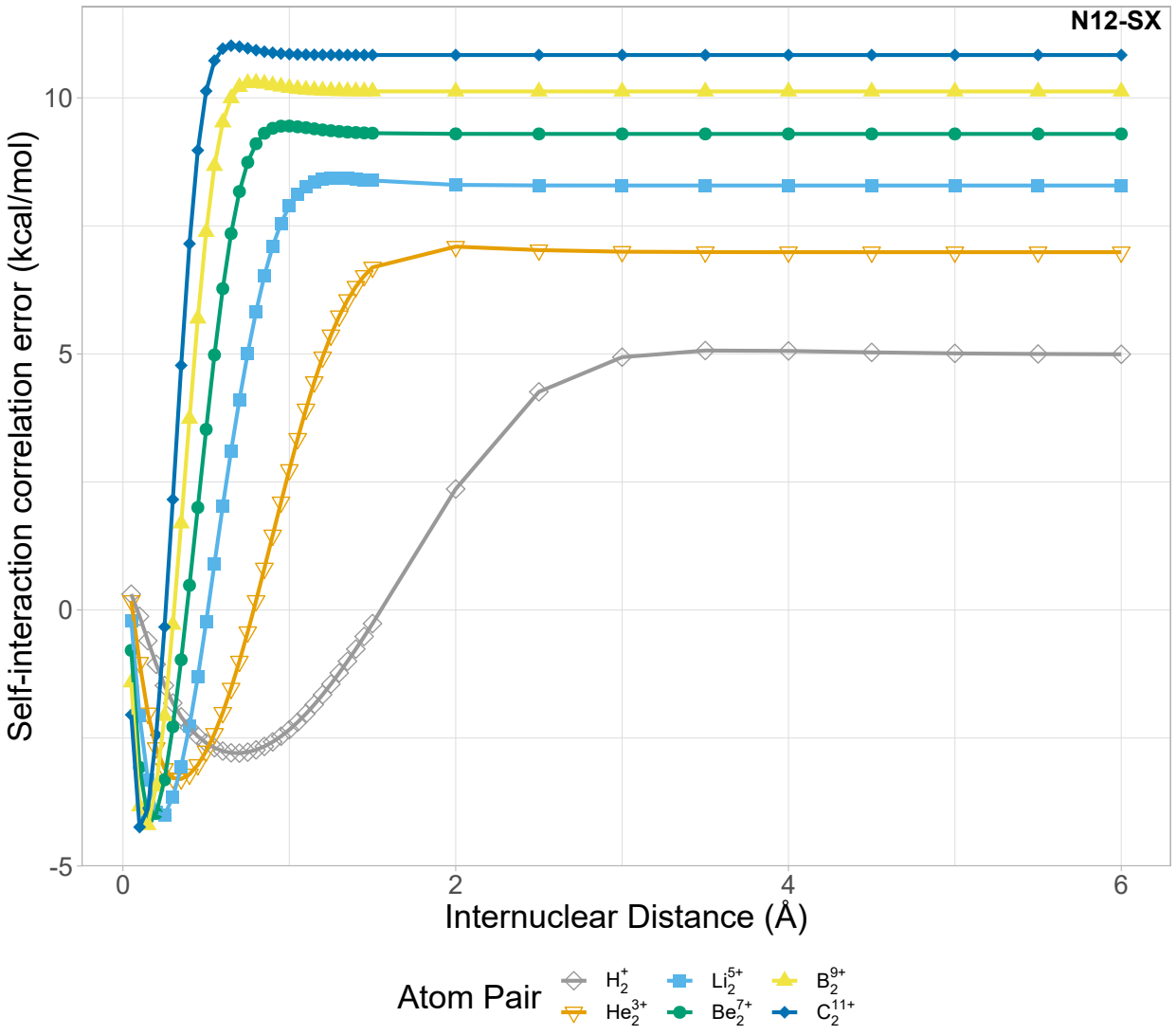

Figure S118: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for N12-SX.

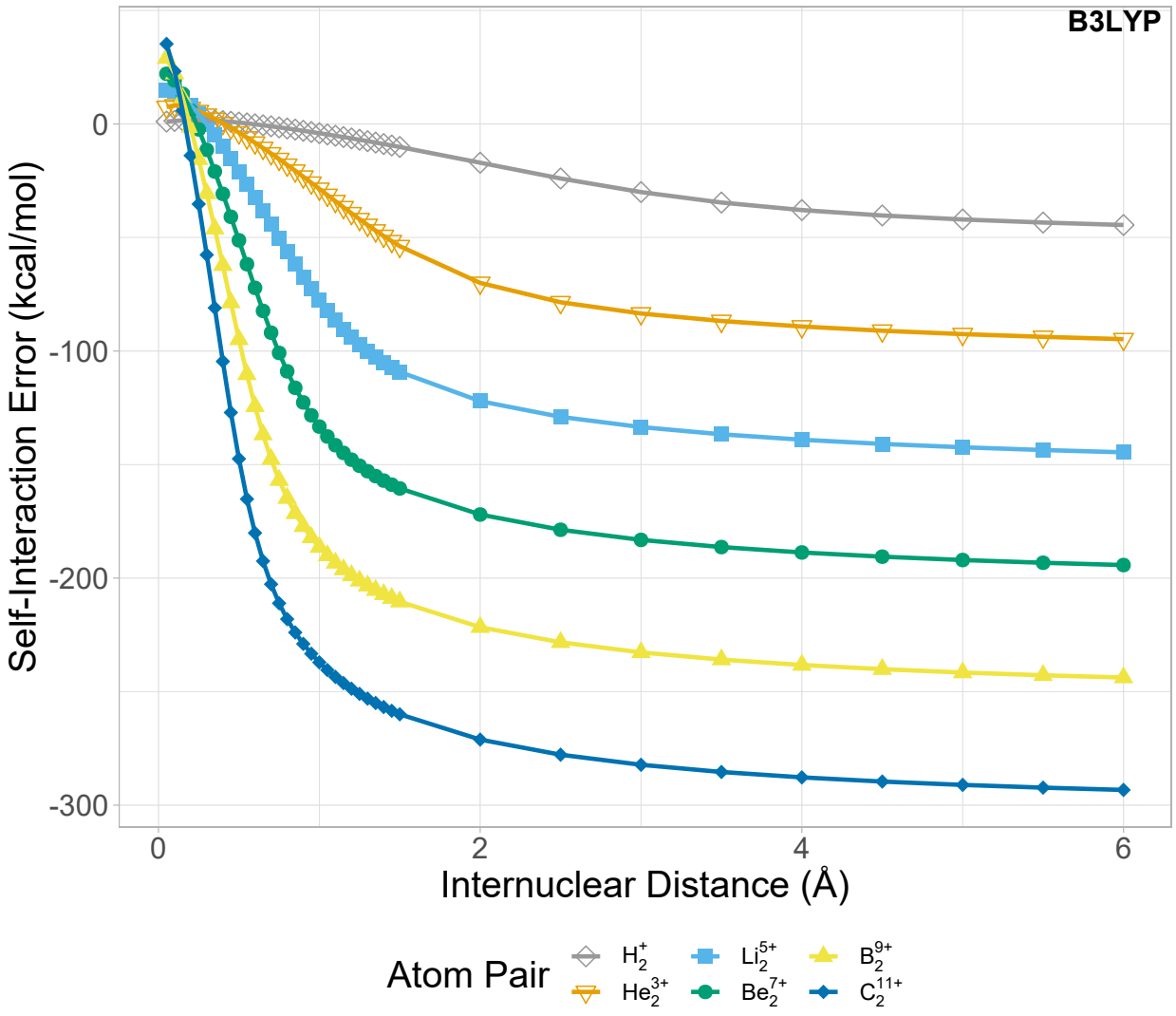

Figure S119: Self-interaction error vs internuclear distance for incremental homonuclear pairs for B3LYP.

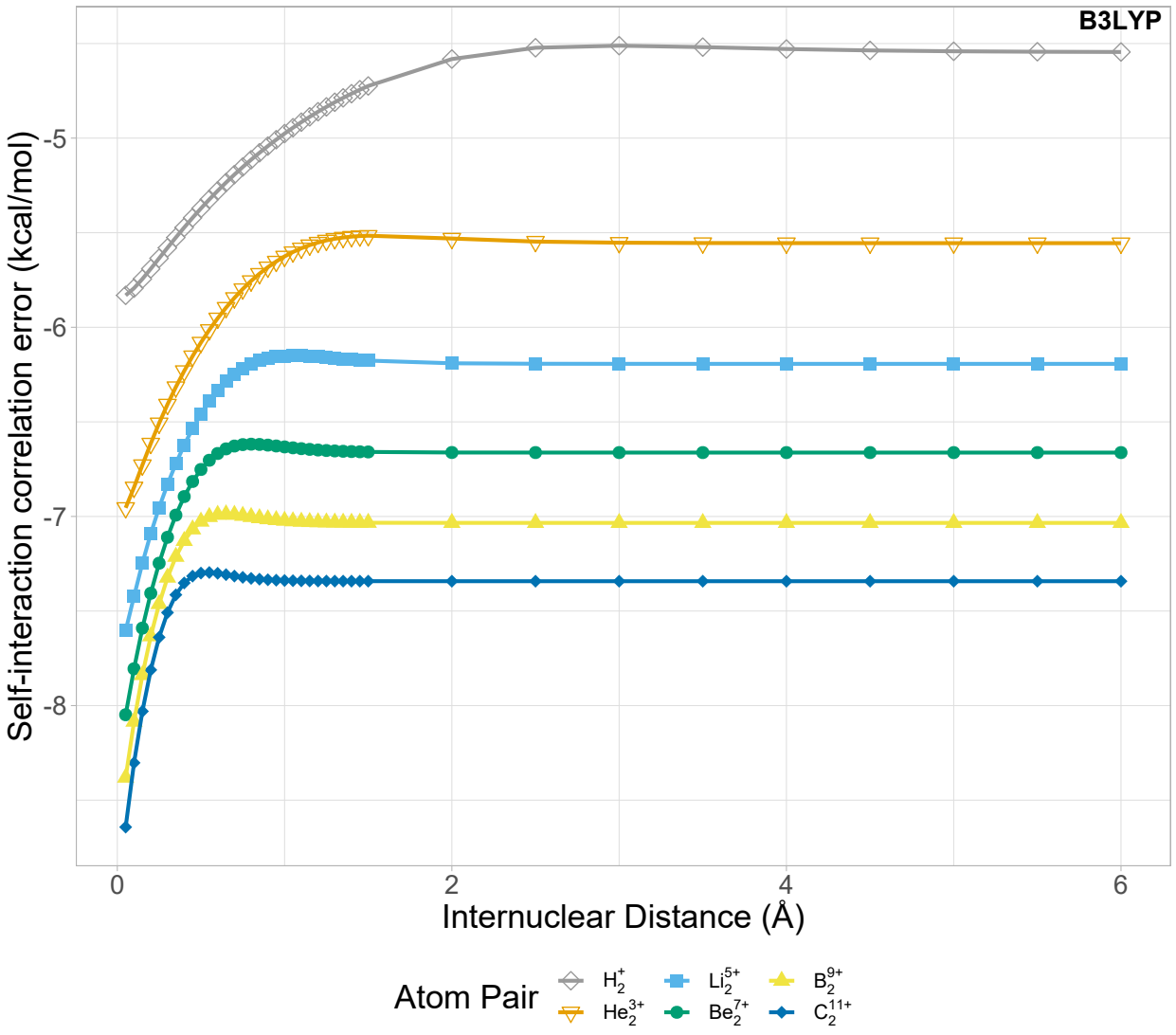

Figure S120: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for B3LYP.

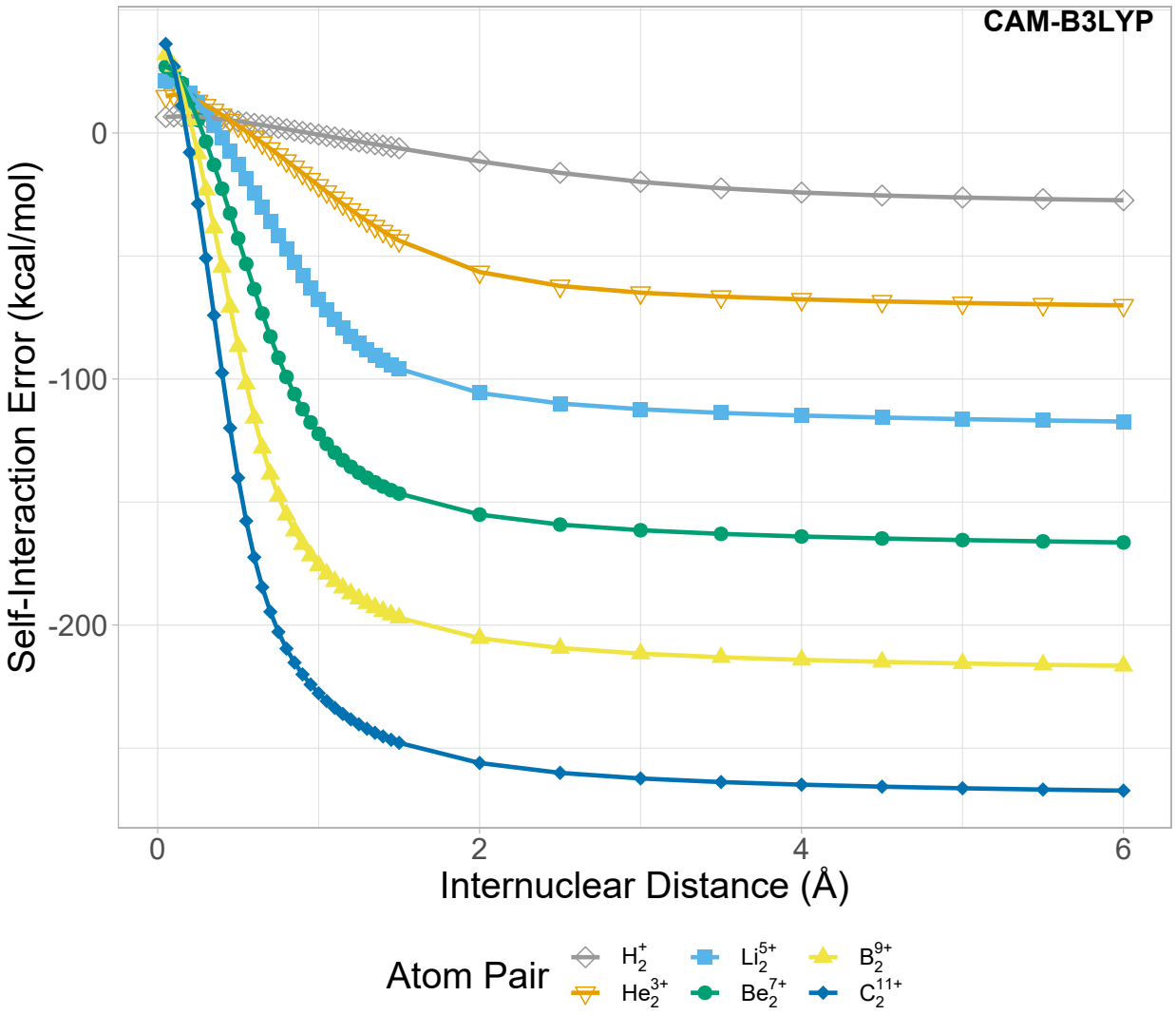

Figure S121: Self-interaction error vs internuclear distance for incremental homonuclear pairs for CAM-B3LYP.

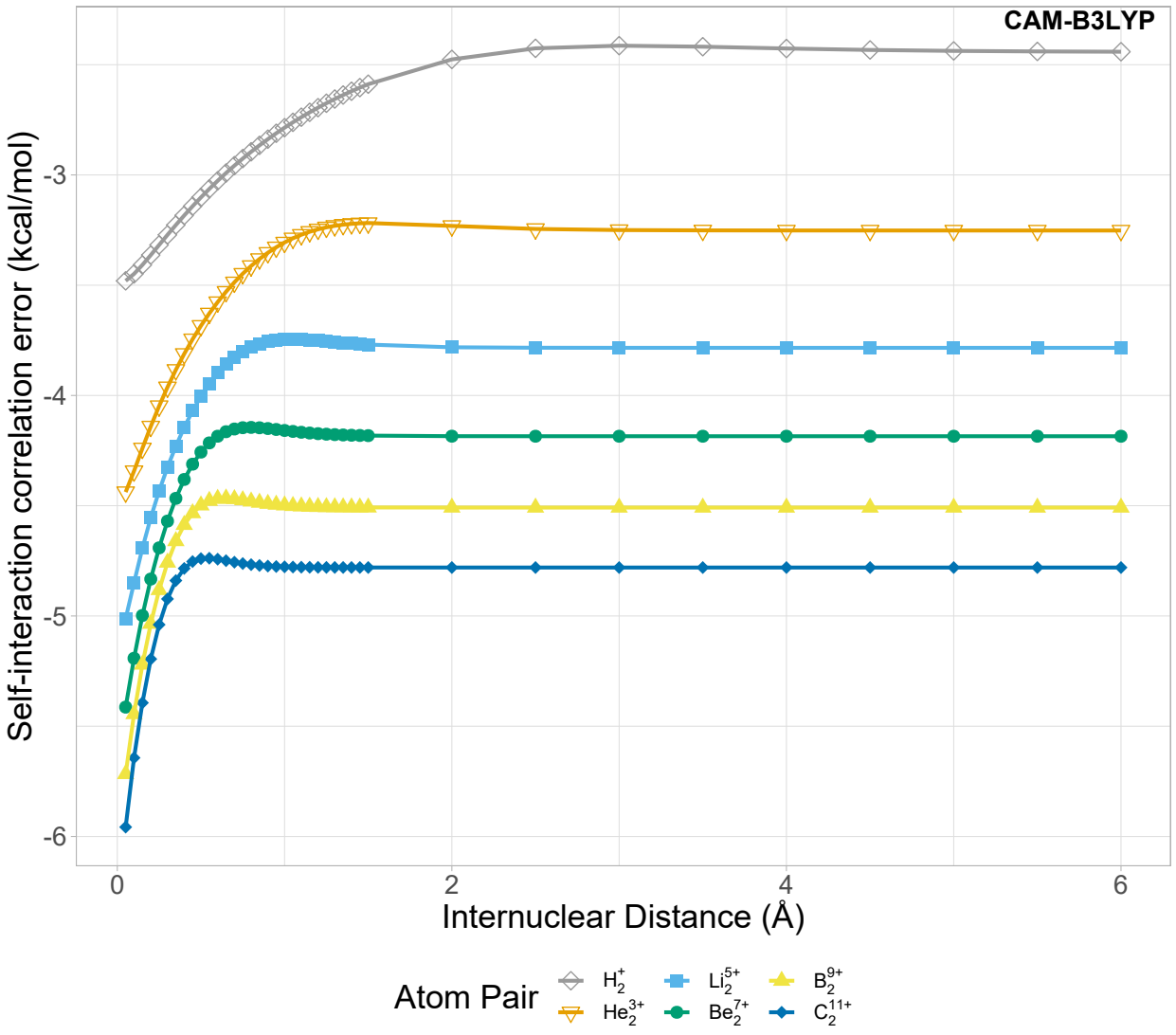

Figure S122: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for CAM-B3LYP.

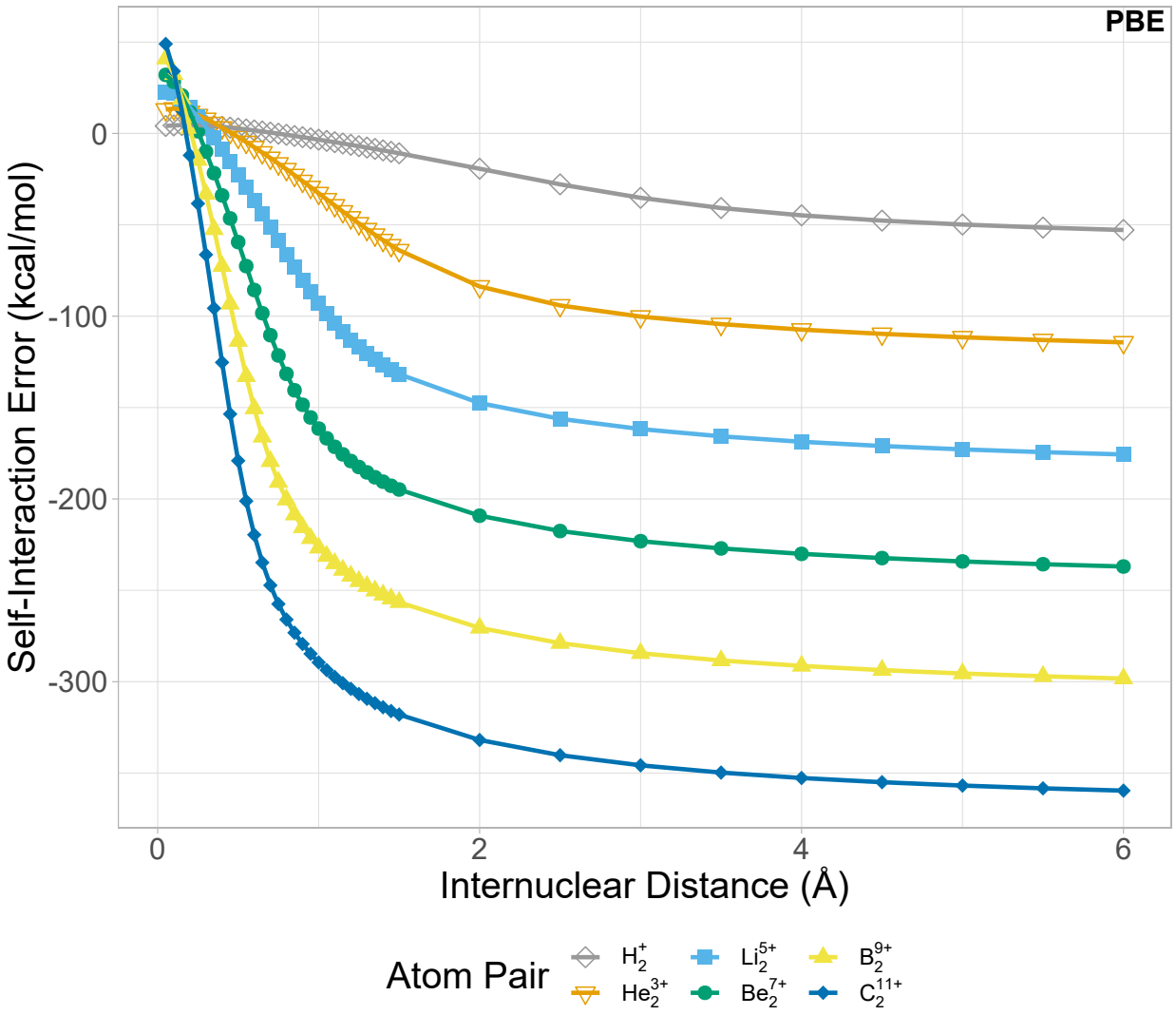

Figure S123: Self-interaction error vs internuclear distance for incremental homonuclear pairs for PBE.

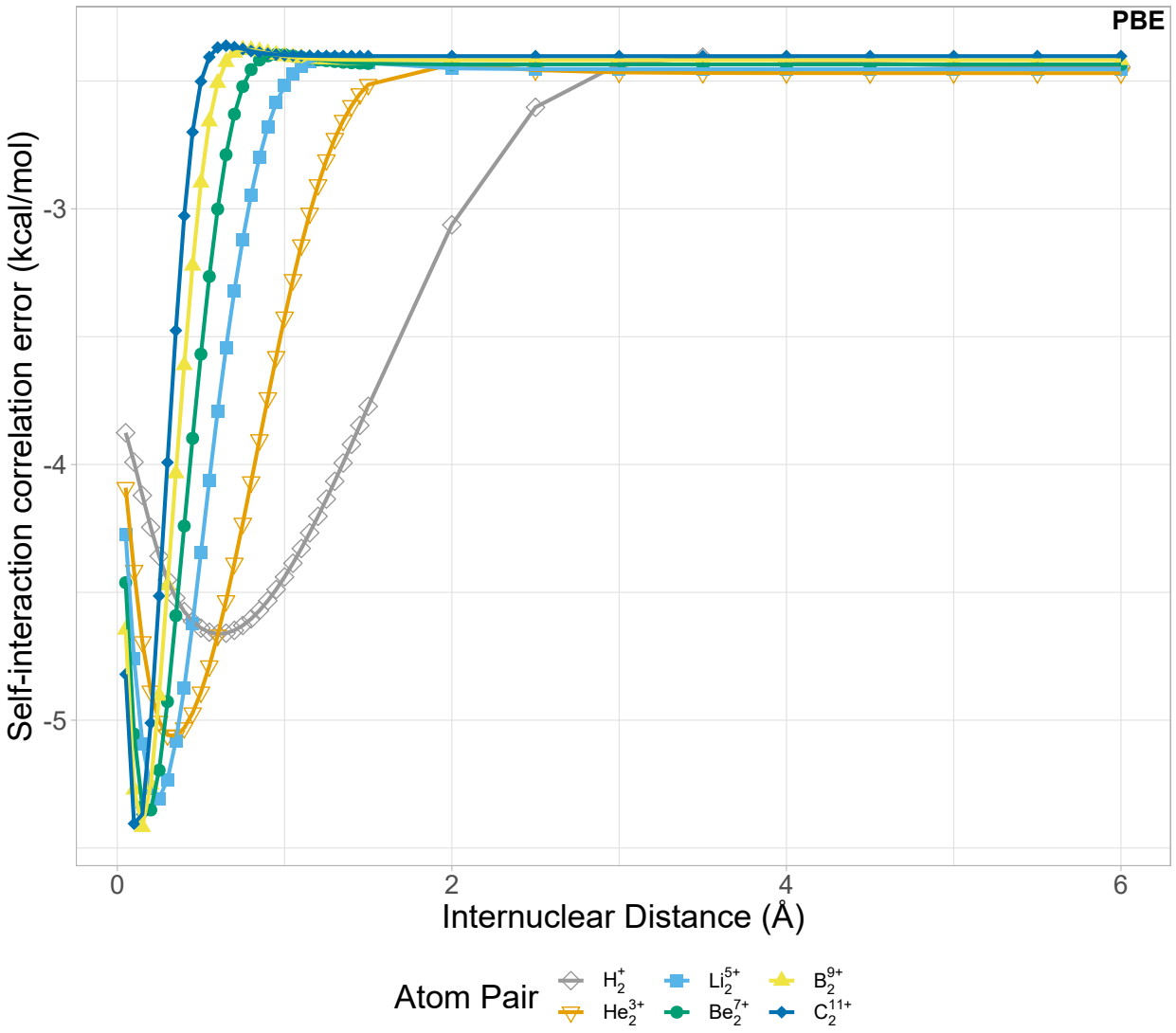

Figure S124: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for PBE.

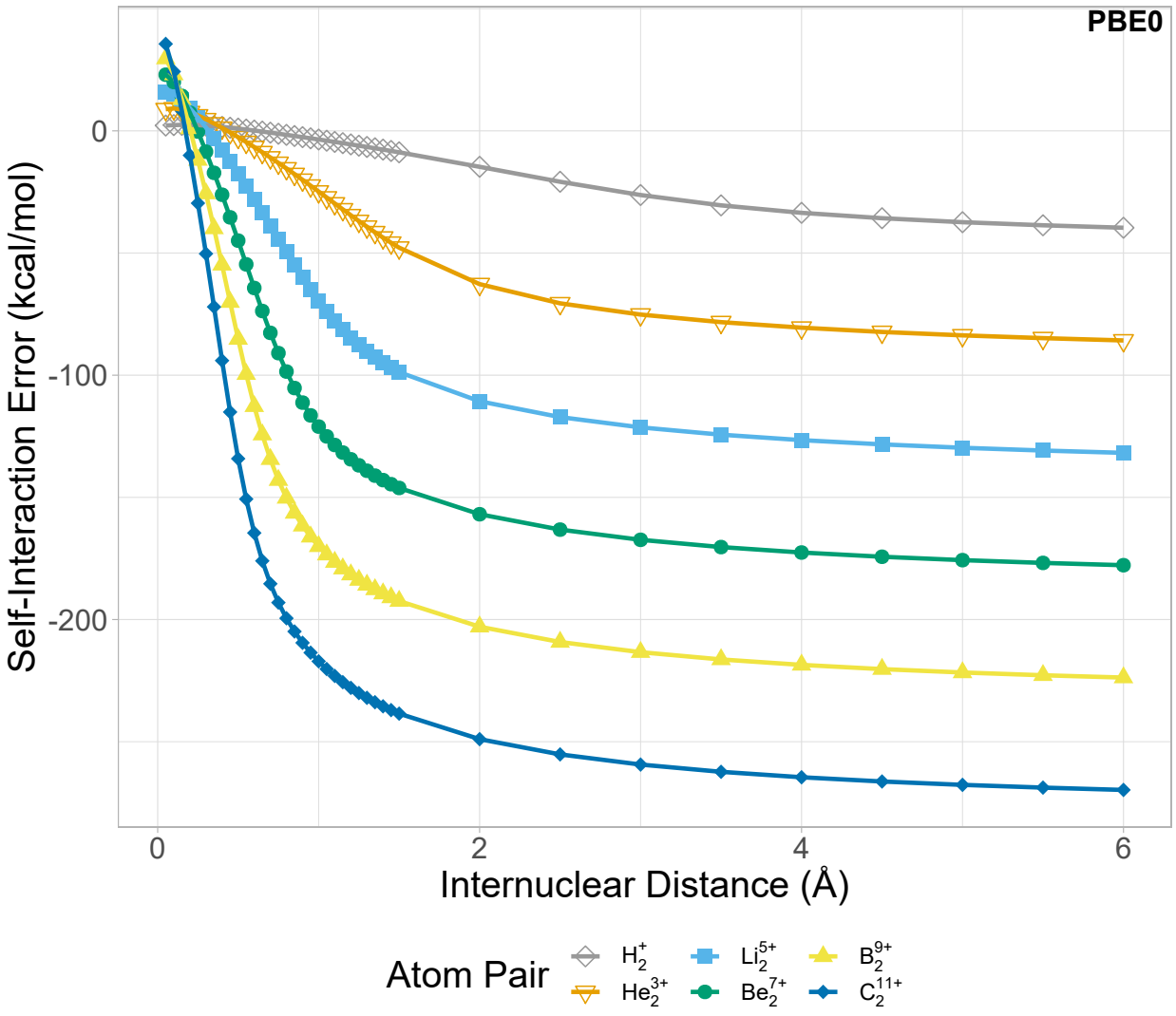

Figure S125: Self-interaction error vs internuclear distance for incremental homonuclear pairs for PBE0.

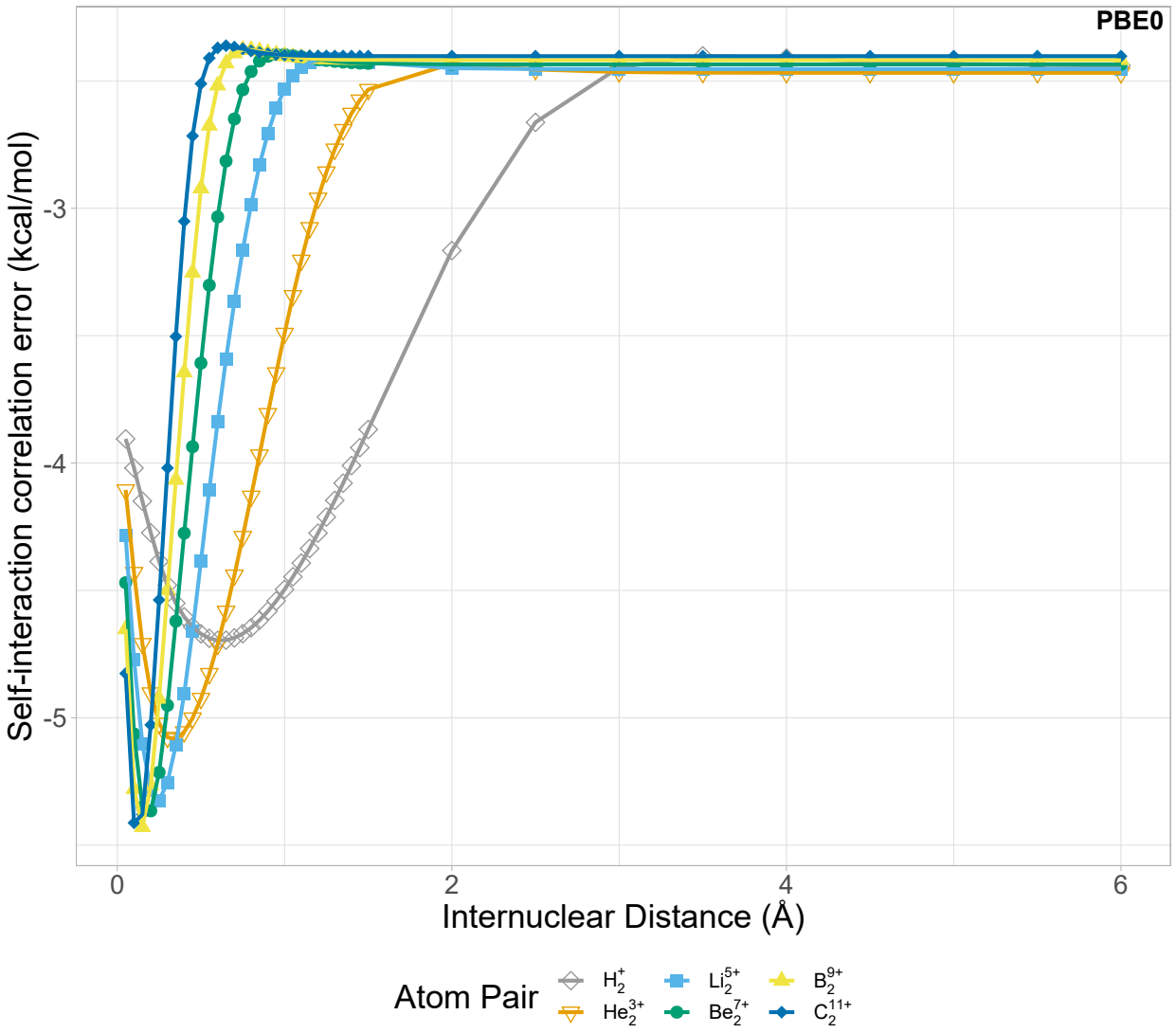

Figure S126: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for PBE0.

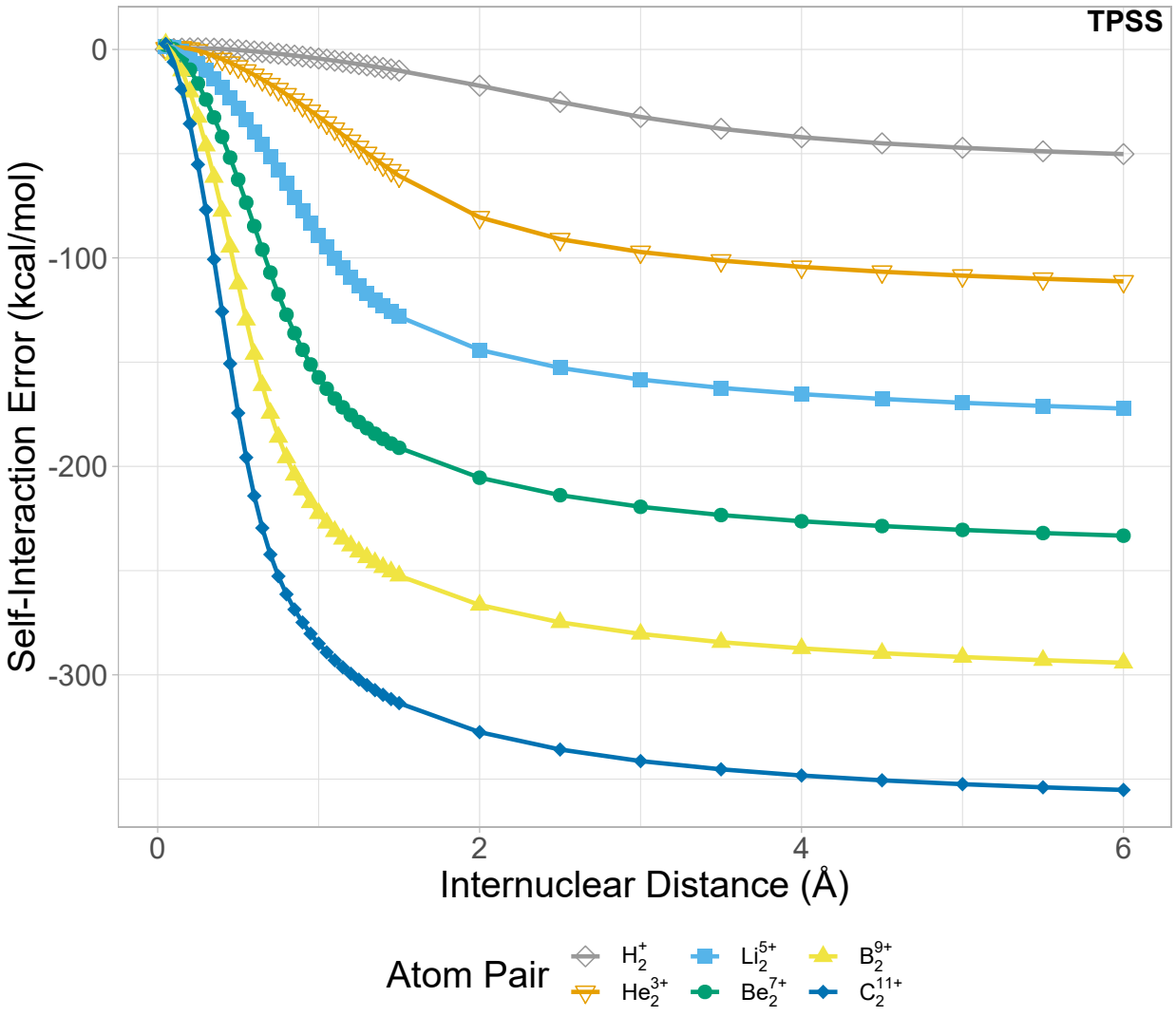

Figure S127: Self-interaction error vs internuclear distance for incremental homonuclear pairs for TPSS.

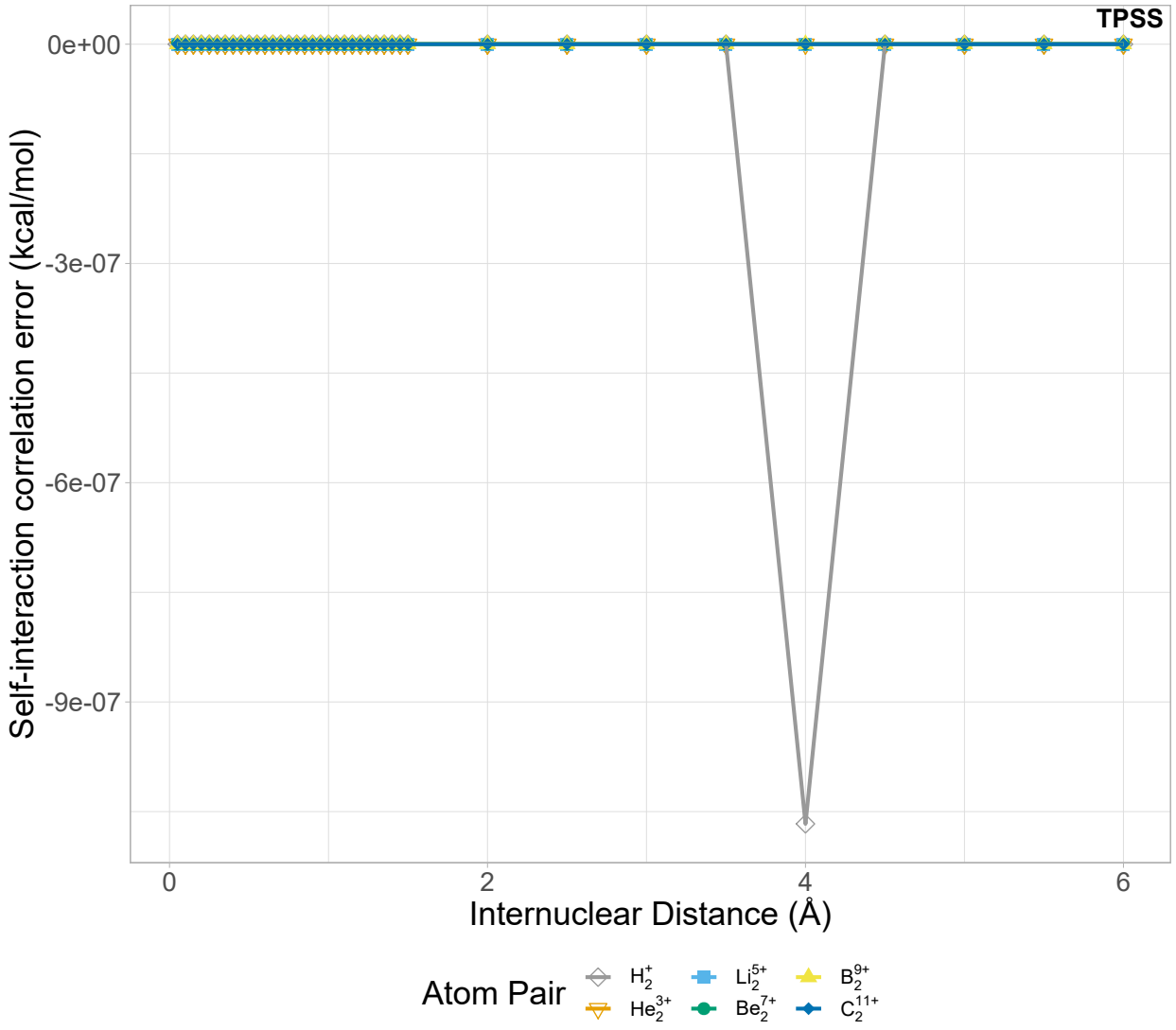

Figure S128: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for TPSS. Formally, this DFA is self-correlation free, any deviations from this is likely computational noise.

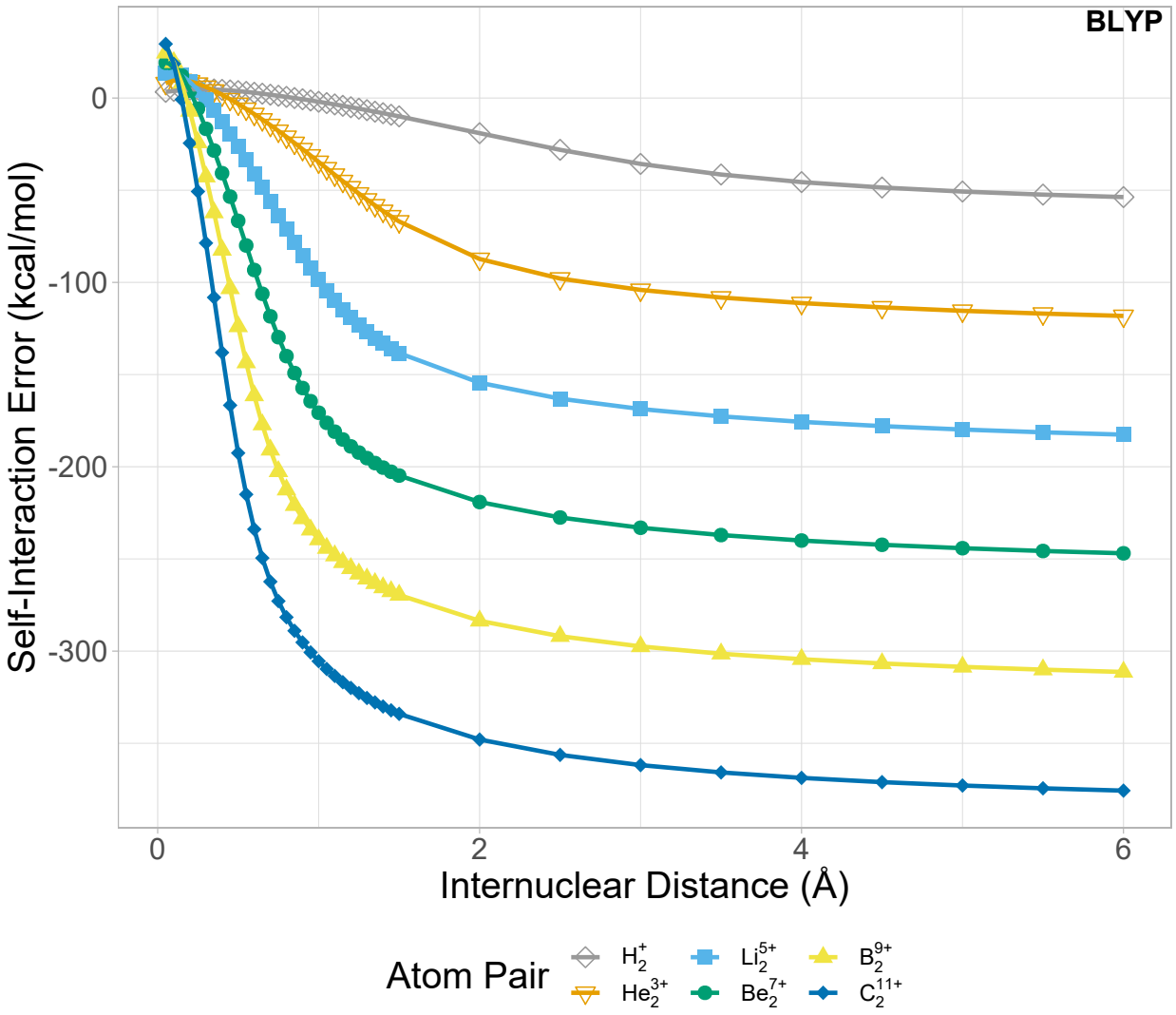

Figure S129: Self-interaction error vs internuclear distance for incremental homonuclear pairs for BLYP.

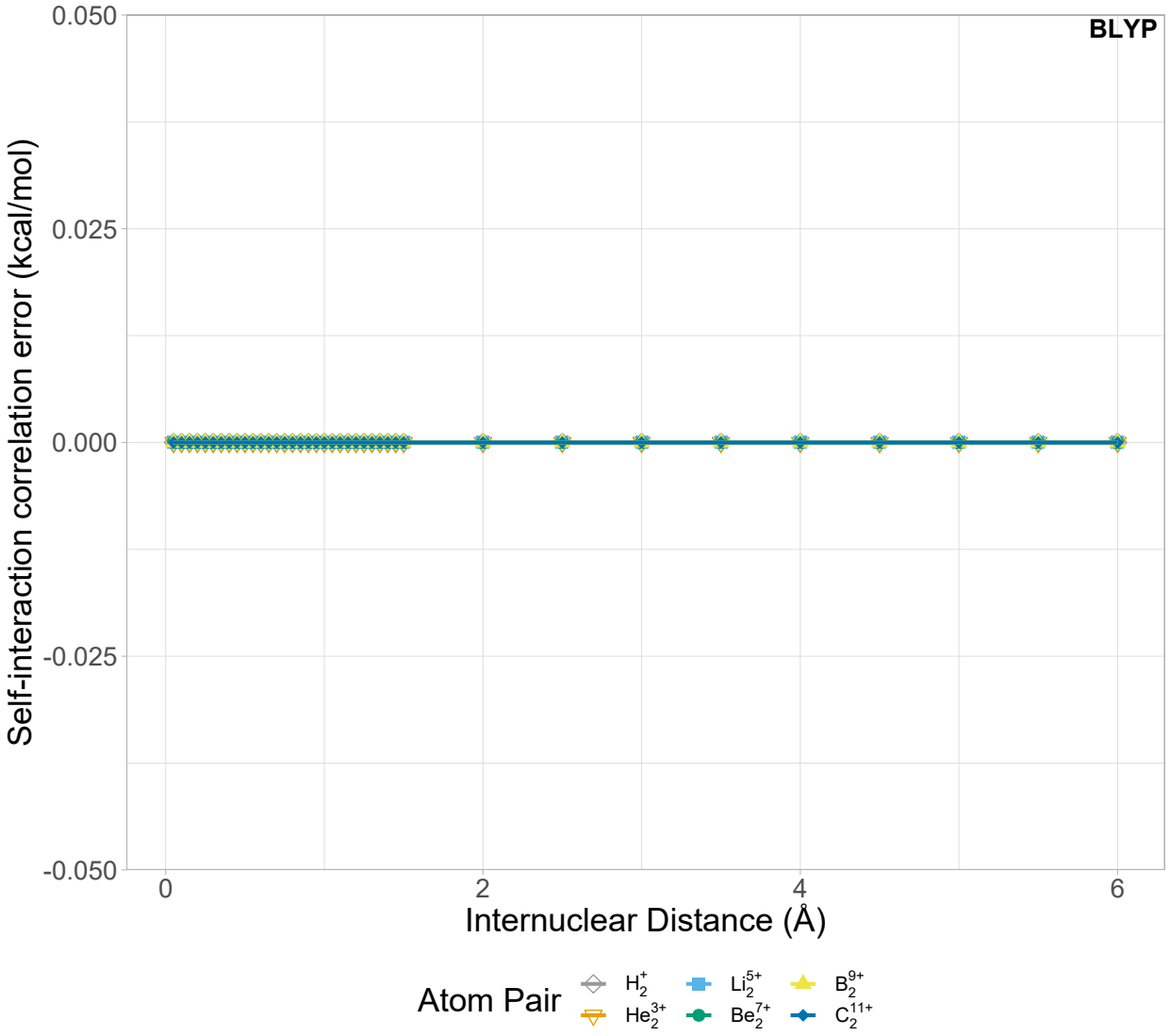

Figure S130: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for BLYP.

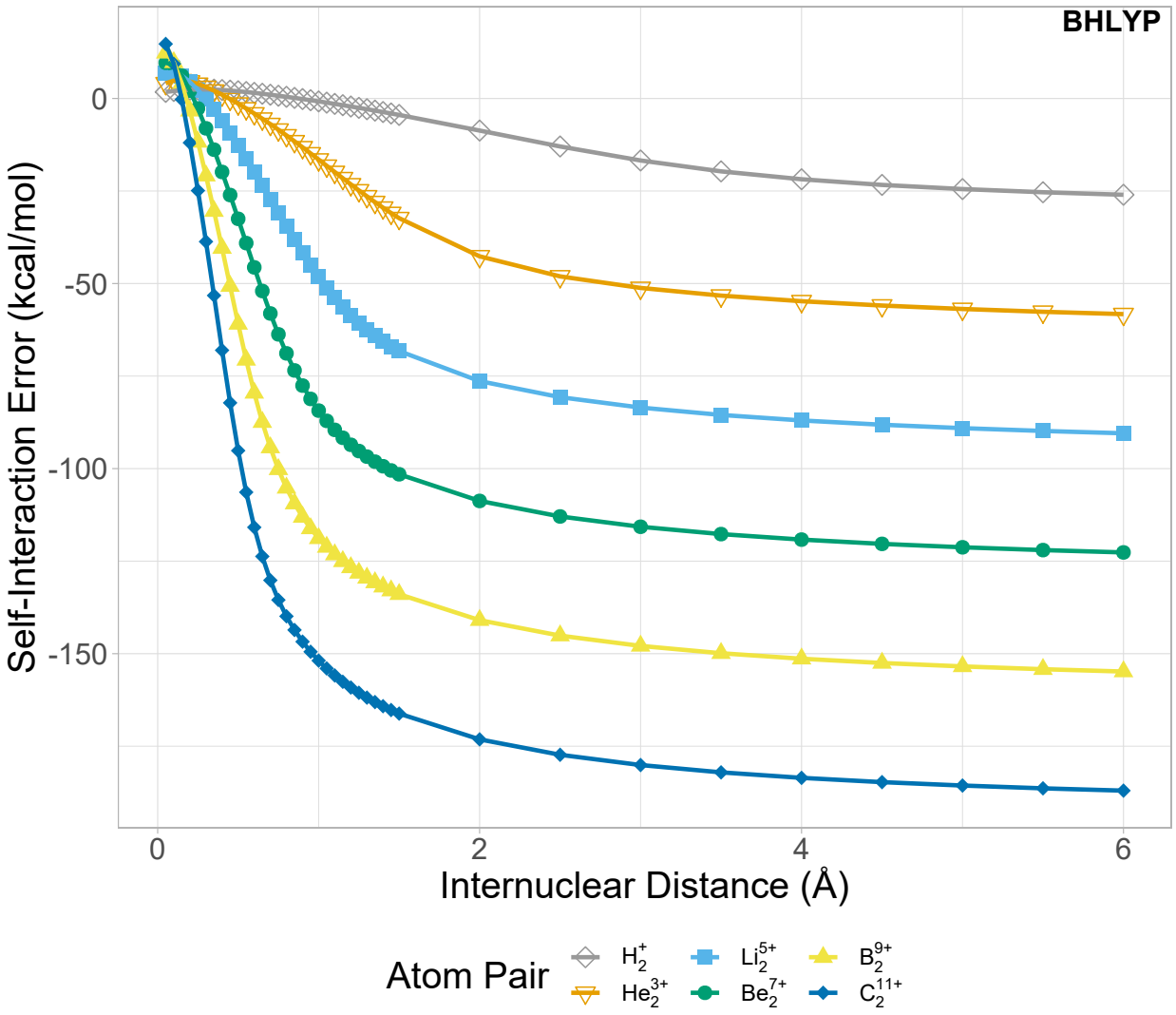

Figure S131: Self-interaction error vs internuclear distance for incremental homonuclear pairs for BHHLYP.

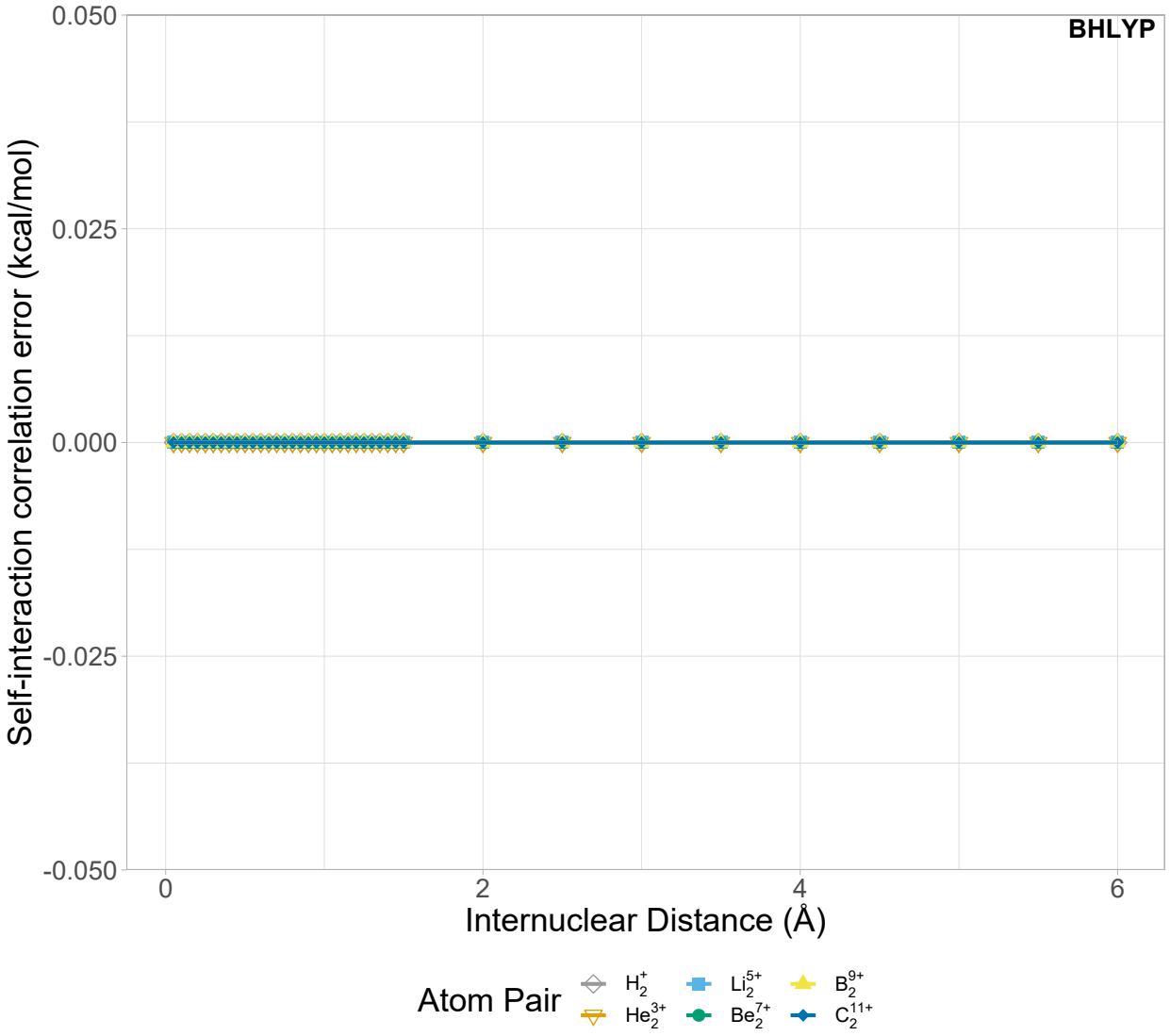

Figure S132: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for BHHLYP.

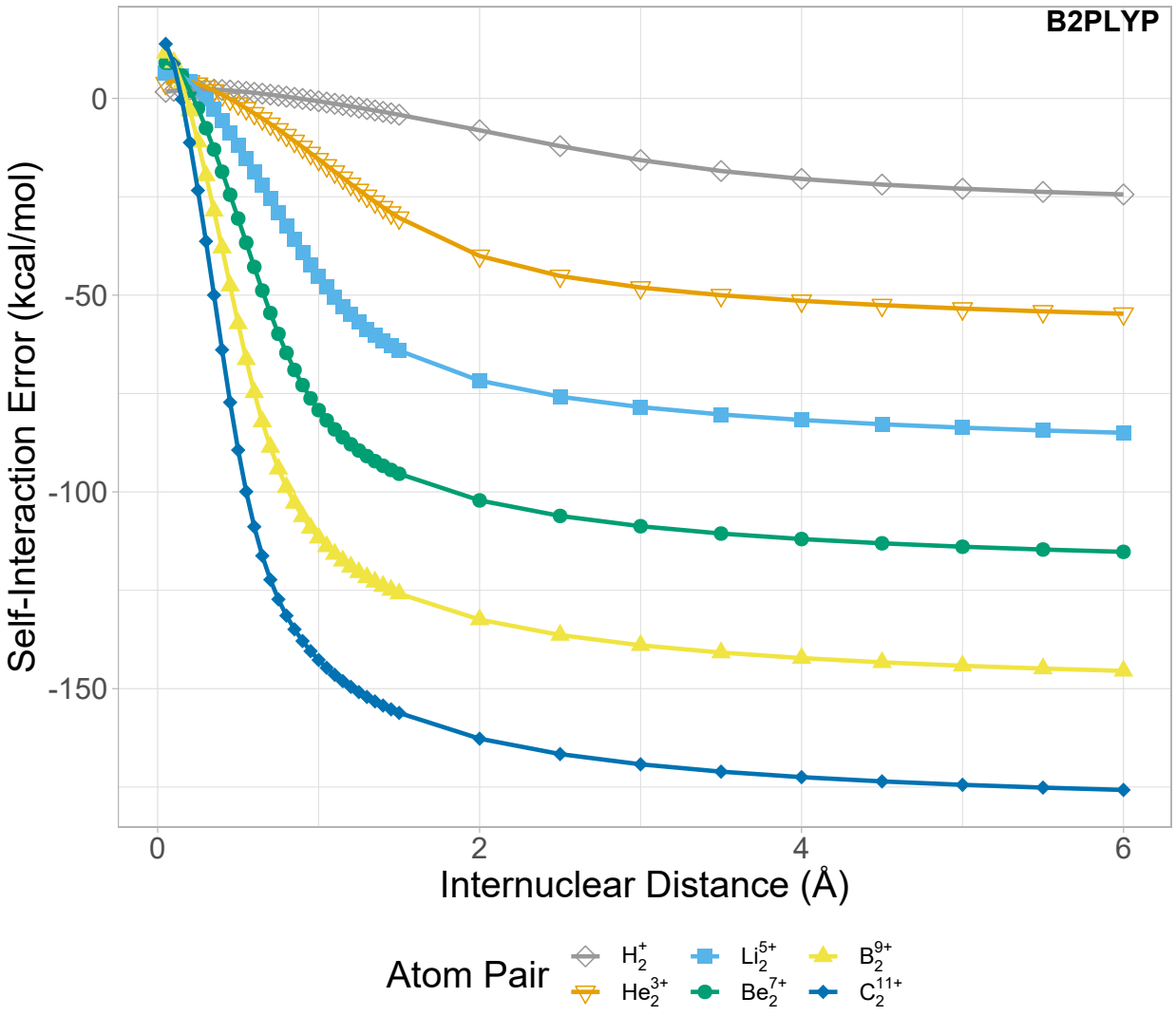

Figure S133: Self-interaction error vs internuclear distance for incremental homonuclear pairs for B2PLYP.

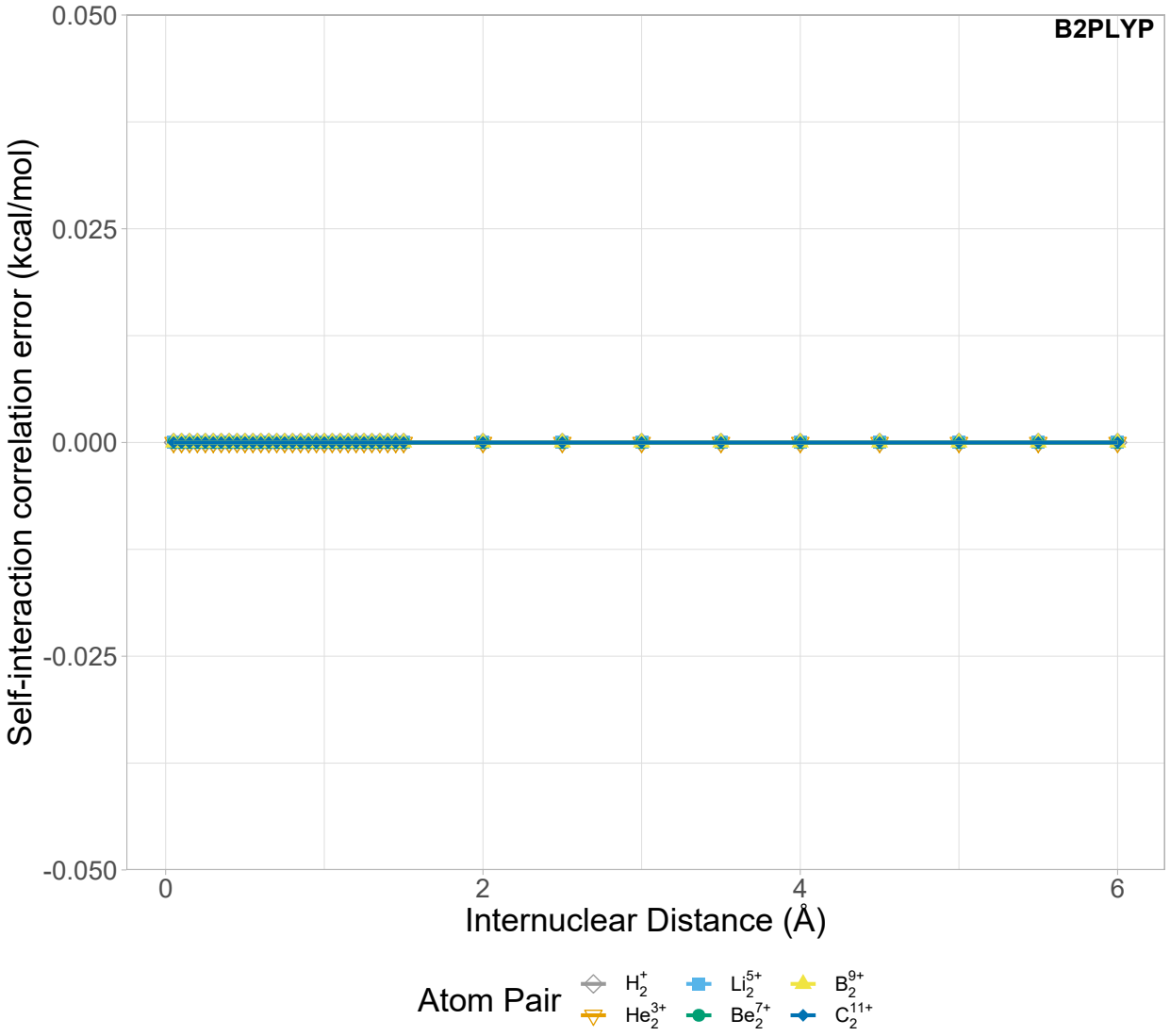

Figure S134: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for B2PLYP.

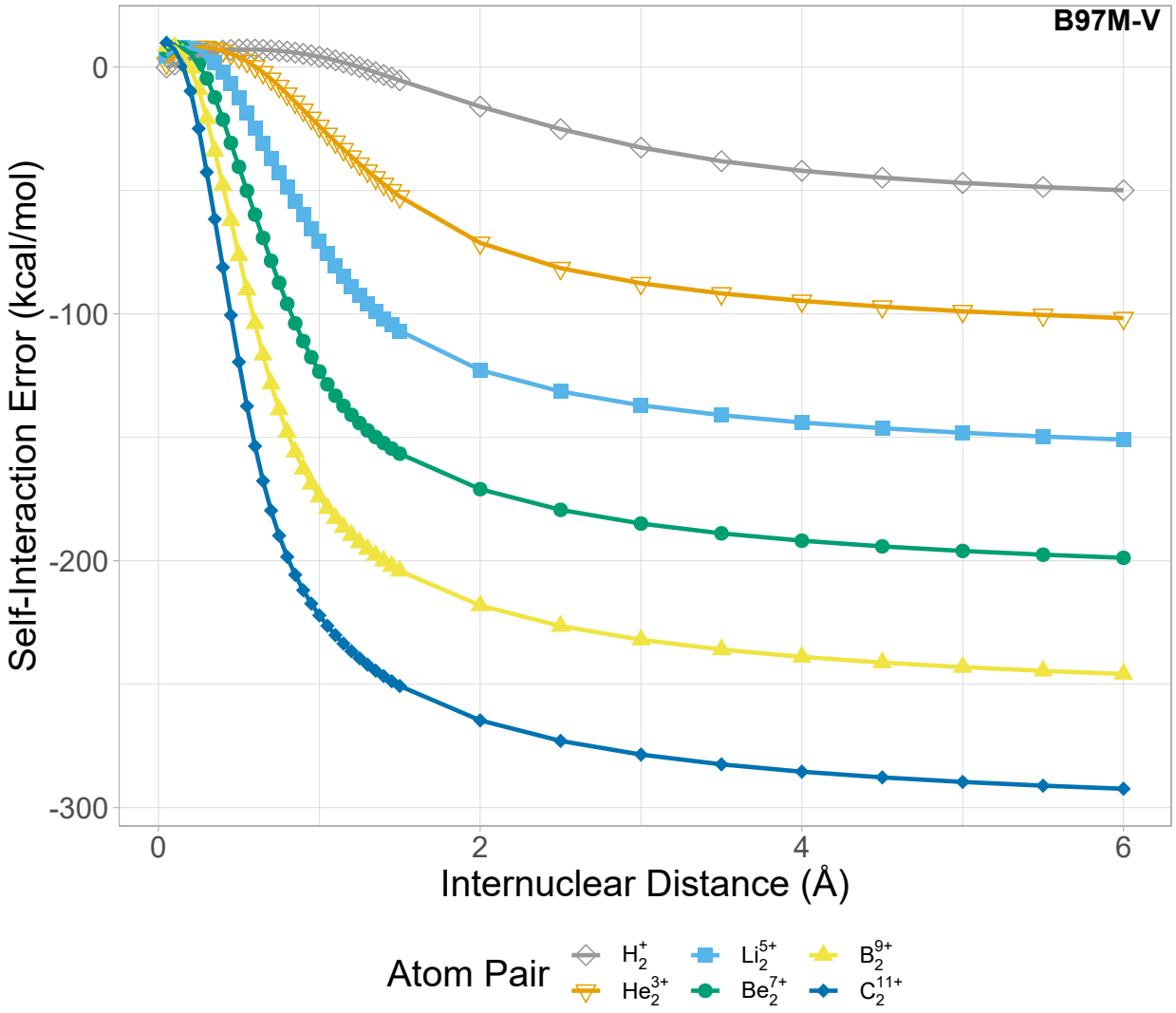

Figure S135: Self-interaction error vs internuclear distance for incremental homonuclear pairs for B97M-V.

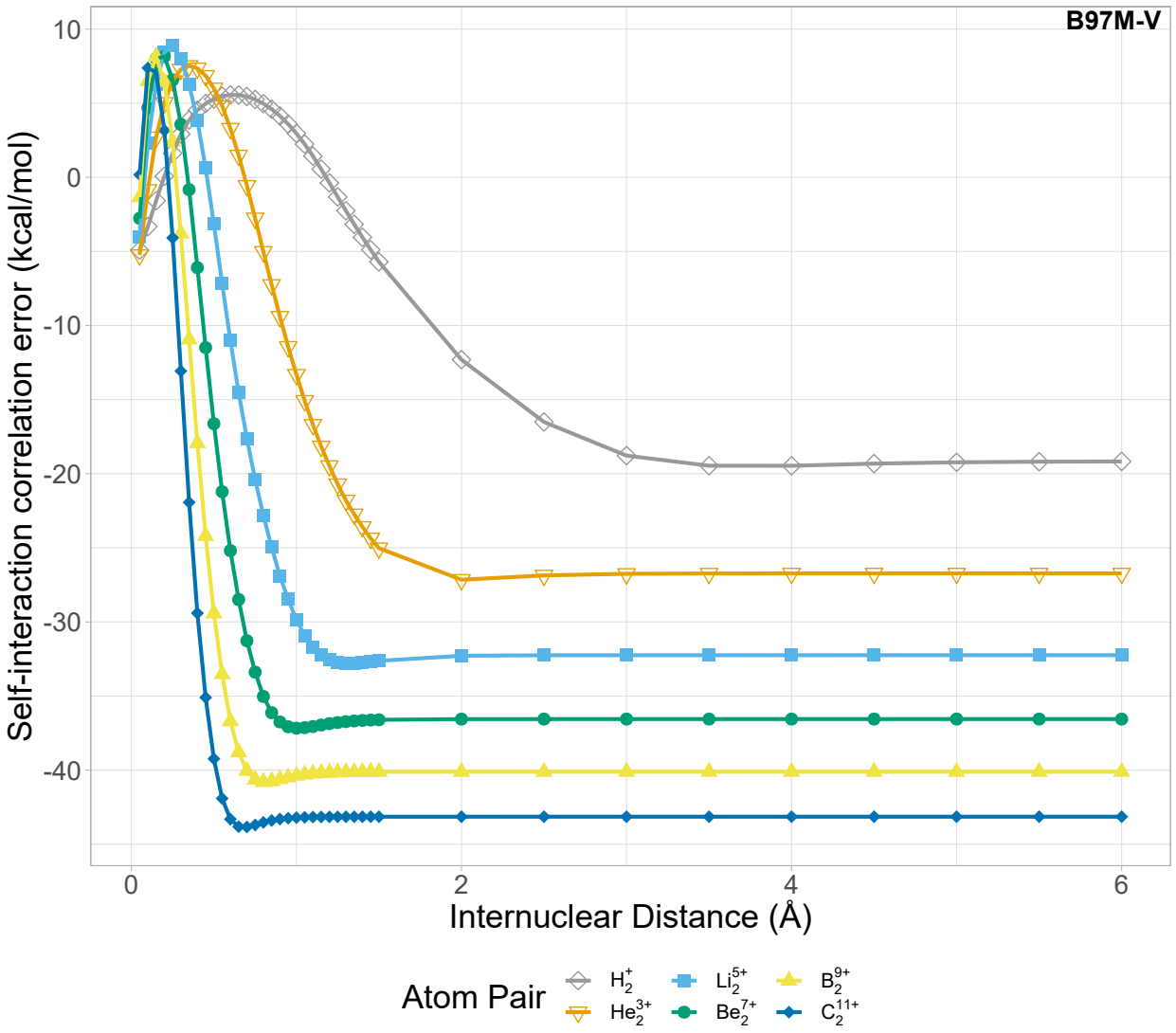

Figure S136: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for B97M-V.

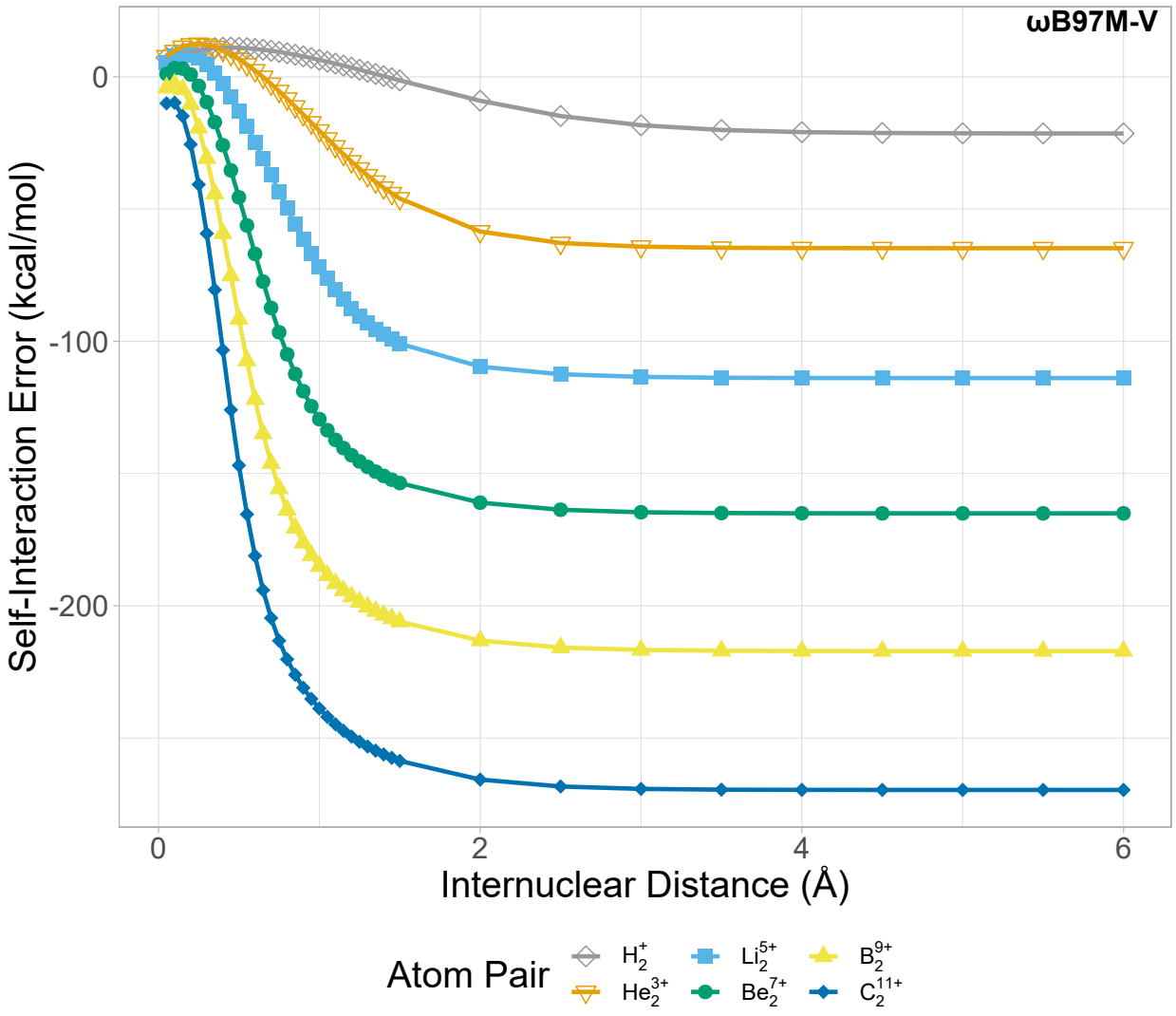

Figure S137: Self-interaction error vs internuclear distance for incremental homonuclear pairs for ωB97M-V.

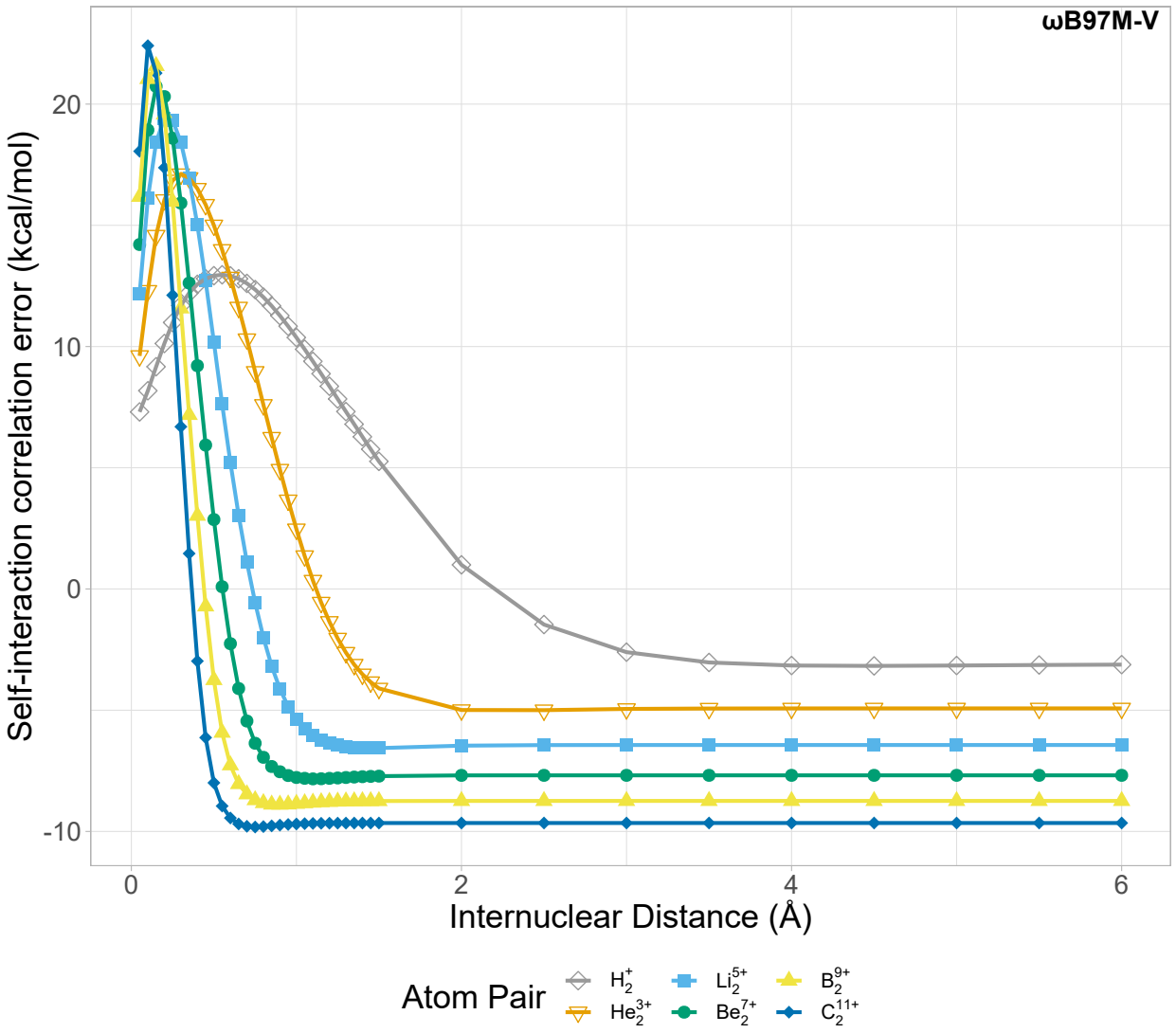

Figure S138: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for ωB97M-V.

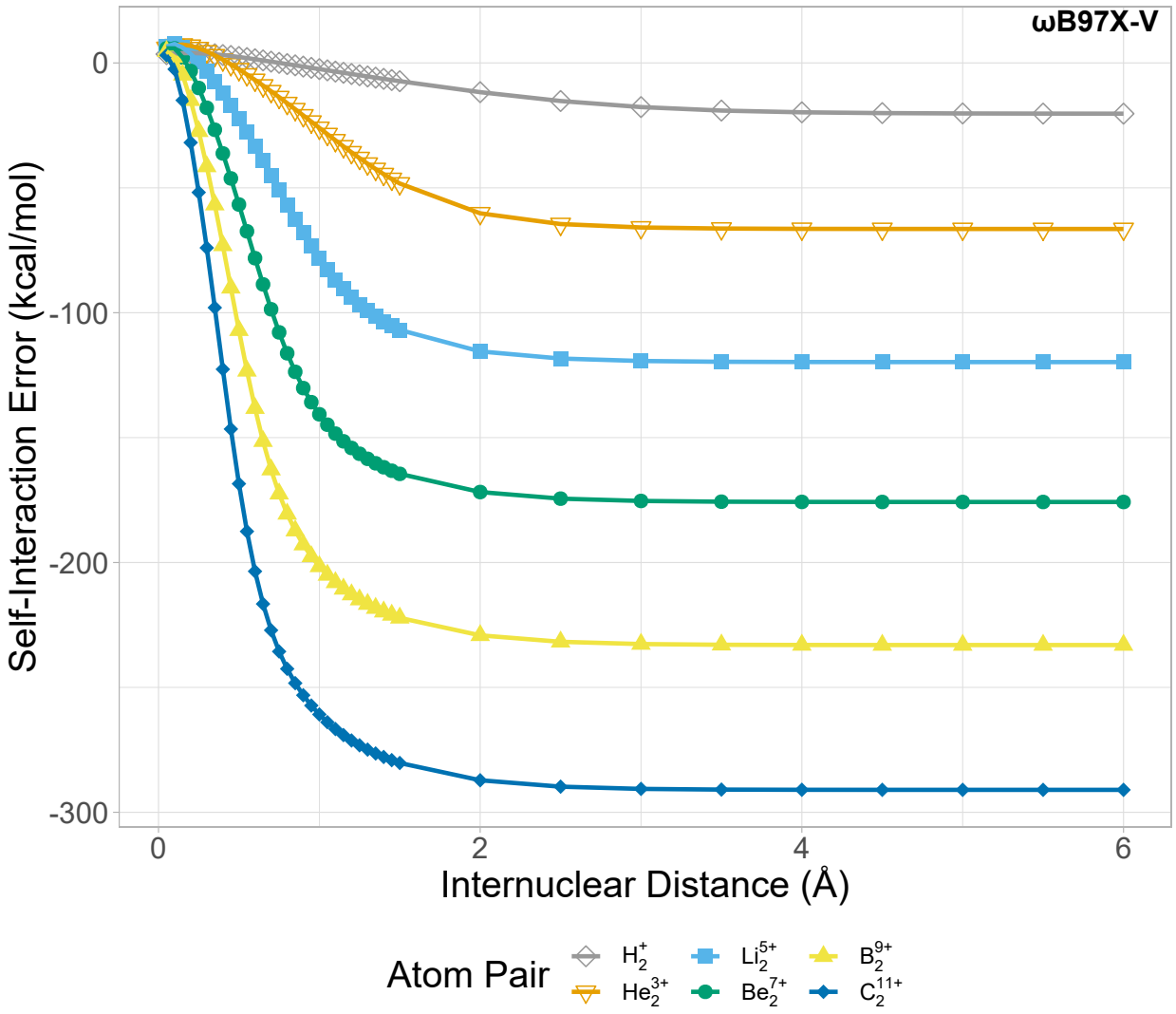

Figure S139: Self-interaction error vs internuclear distance for incremental homonuclear pairs for ωB97X-V.

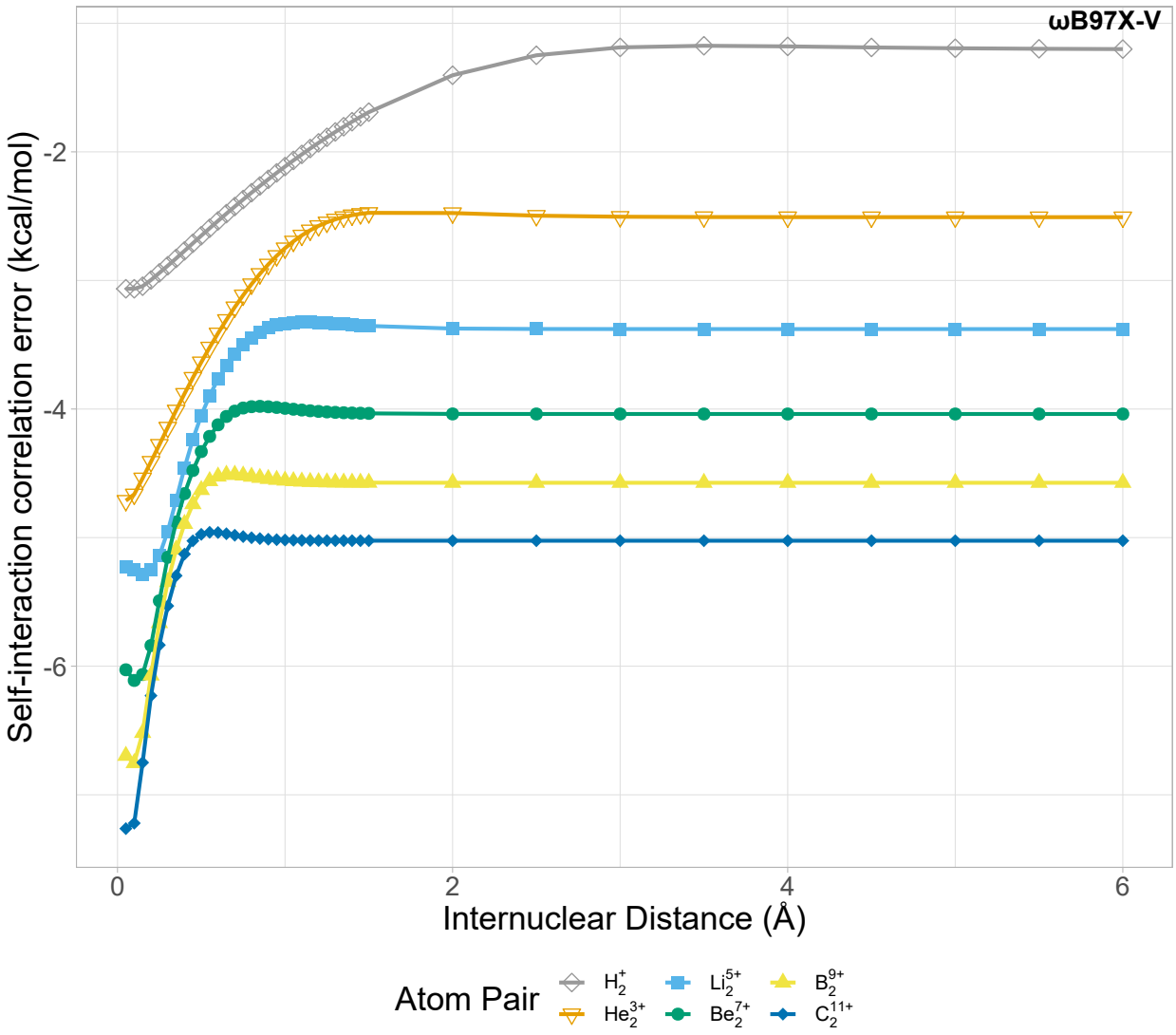

Figure S140: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for ωB97X-V.

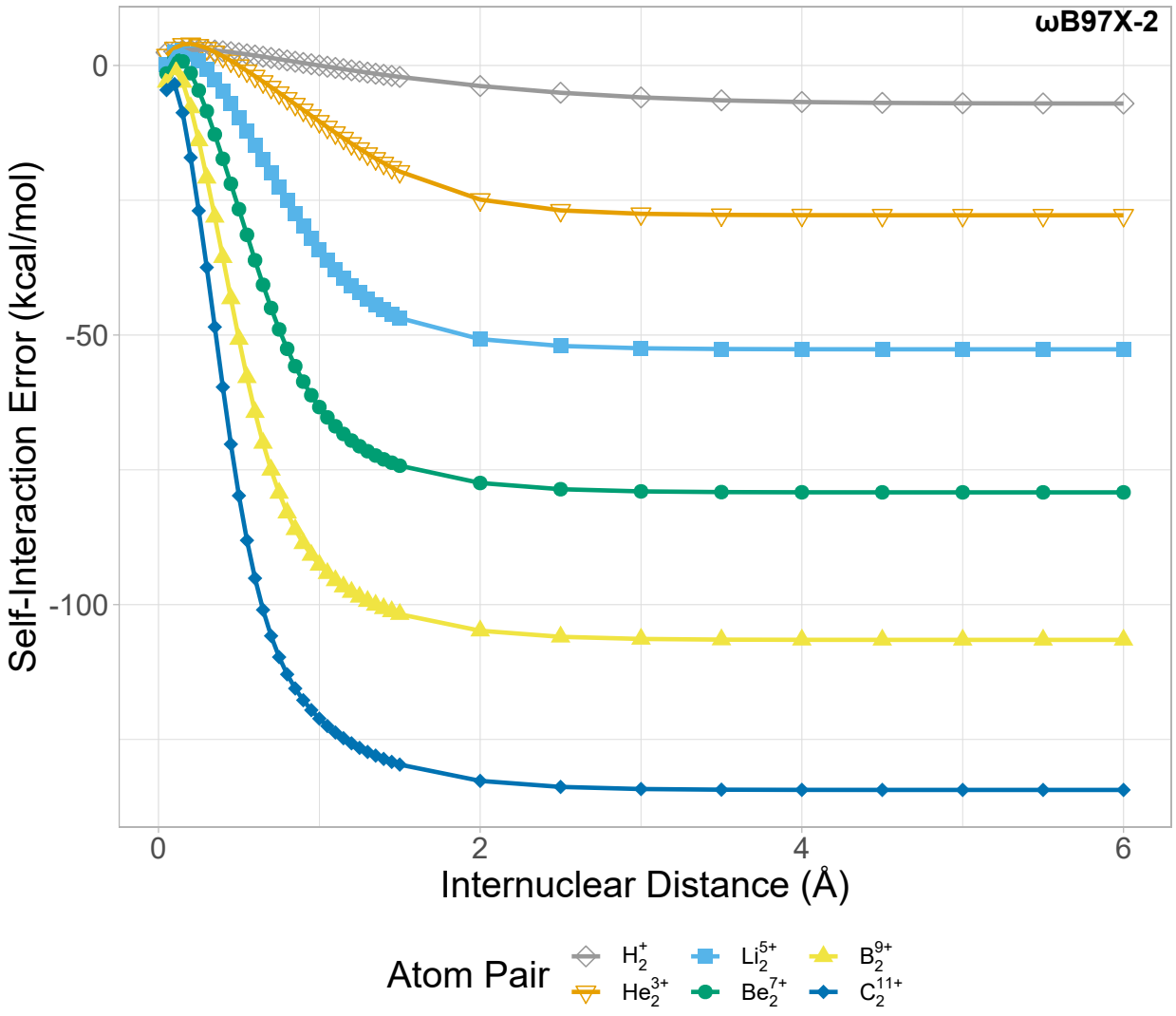

Figure S141: Self-interaction error vs internuclear distance for incremental homonuclear pairs for ωB97X-2.

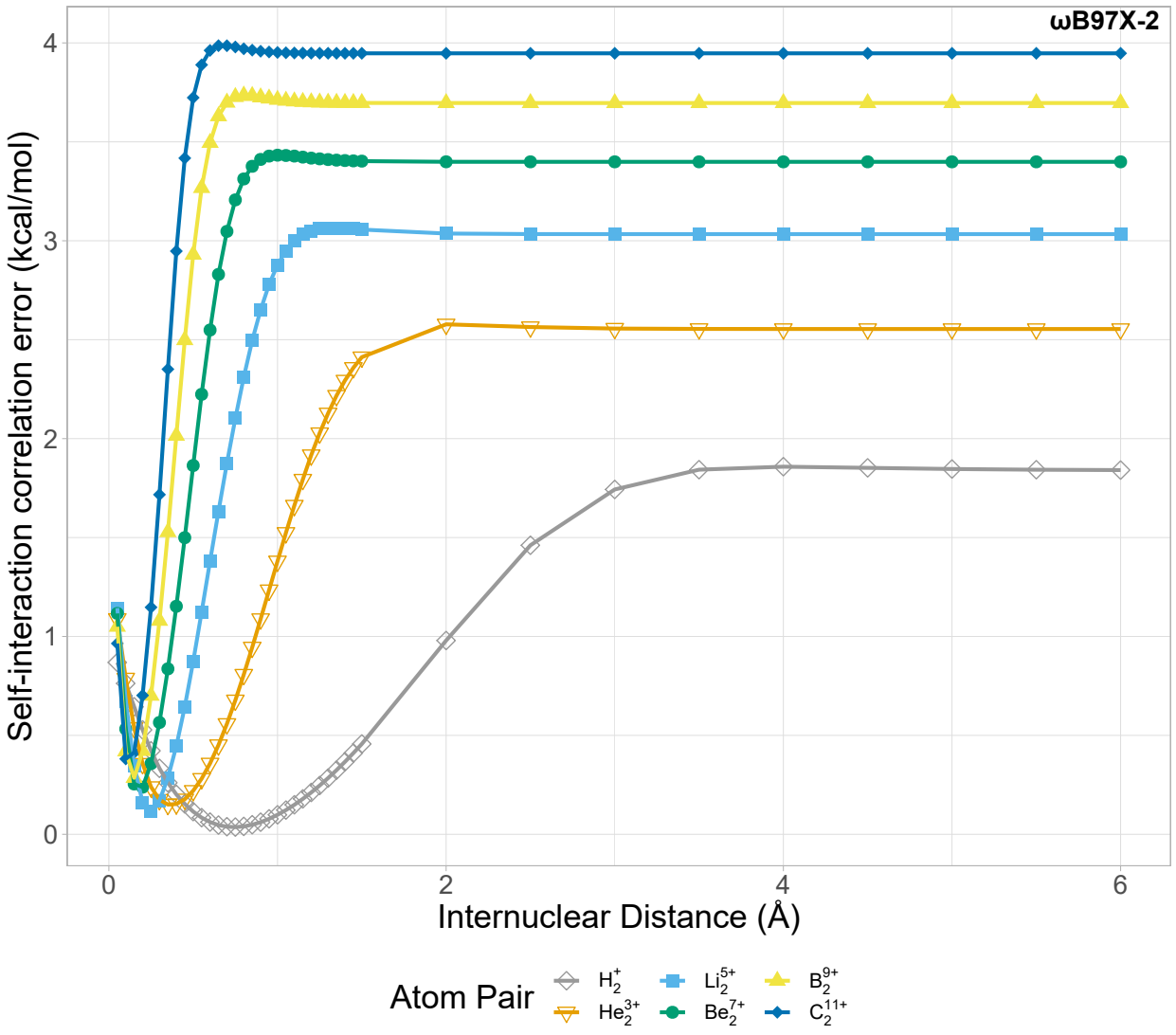

Figure S142: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for ωB97X-2.

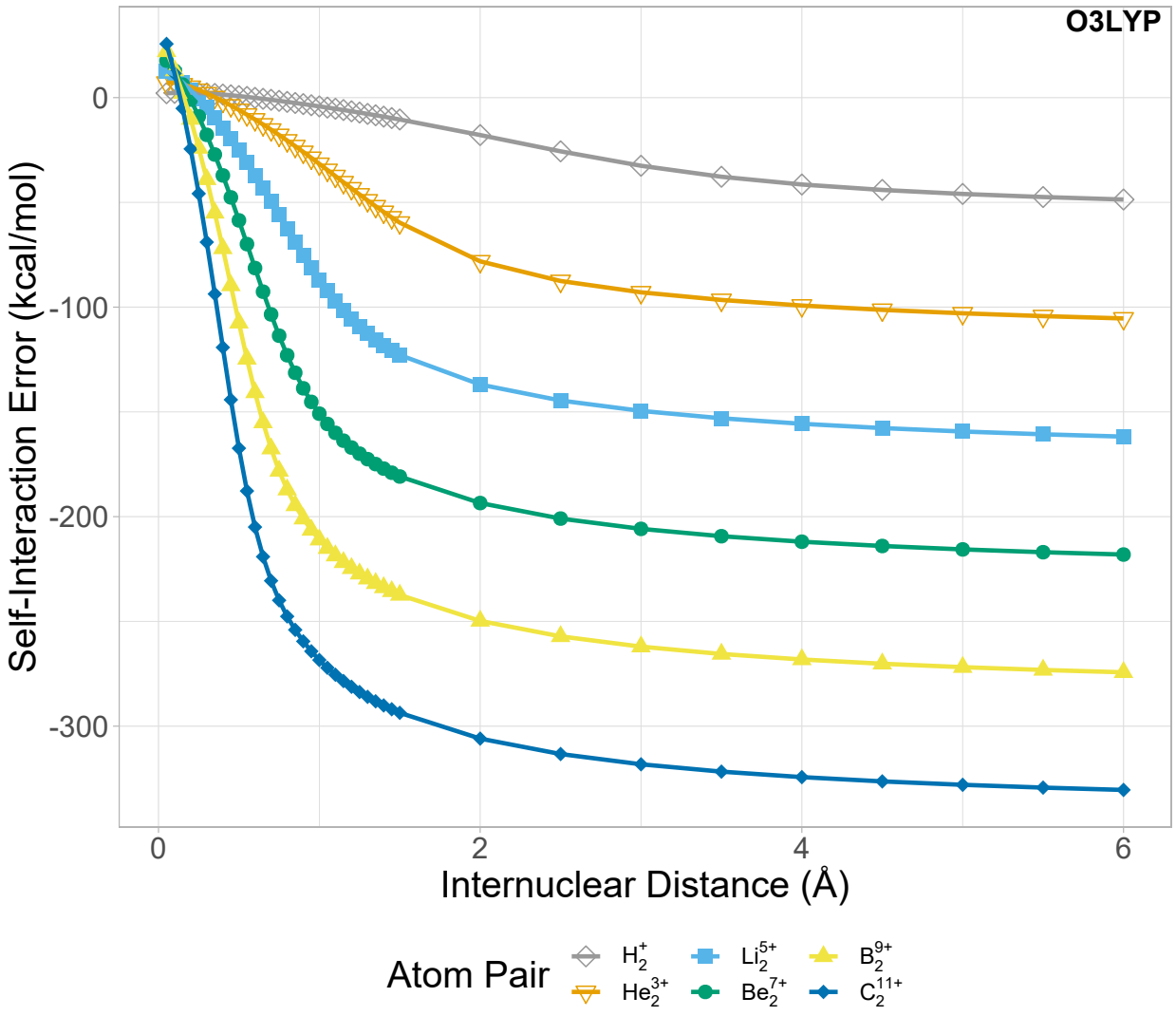

Figure S143: Self-interaction error vs internuclear distance for incremental homonuclear pairs for O3LYP.

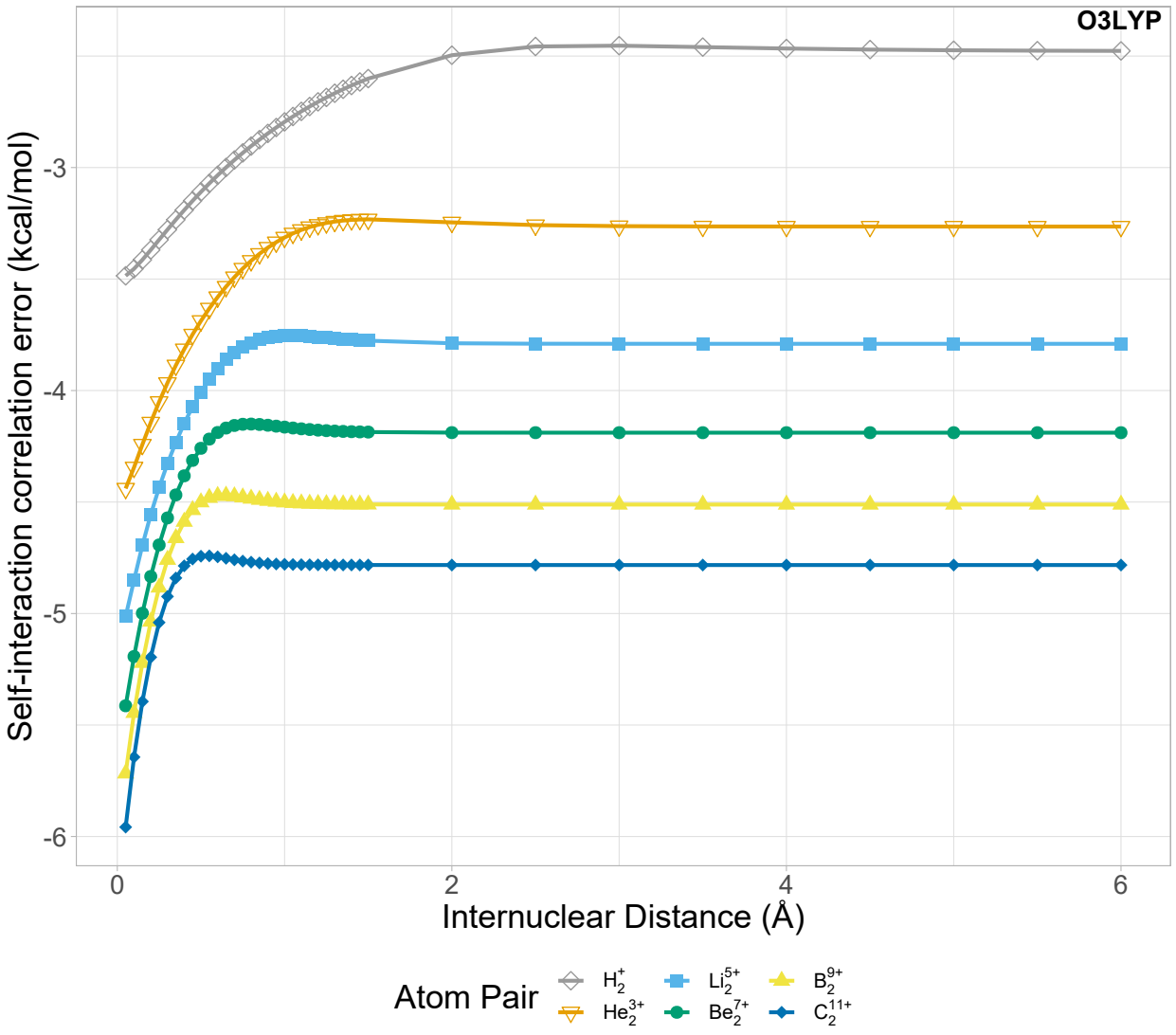

Figure S144: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for O3LYP.

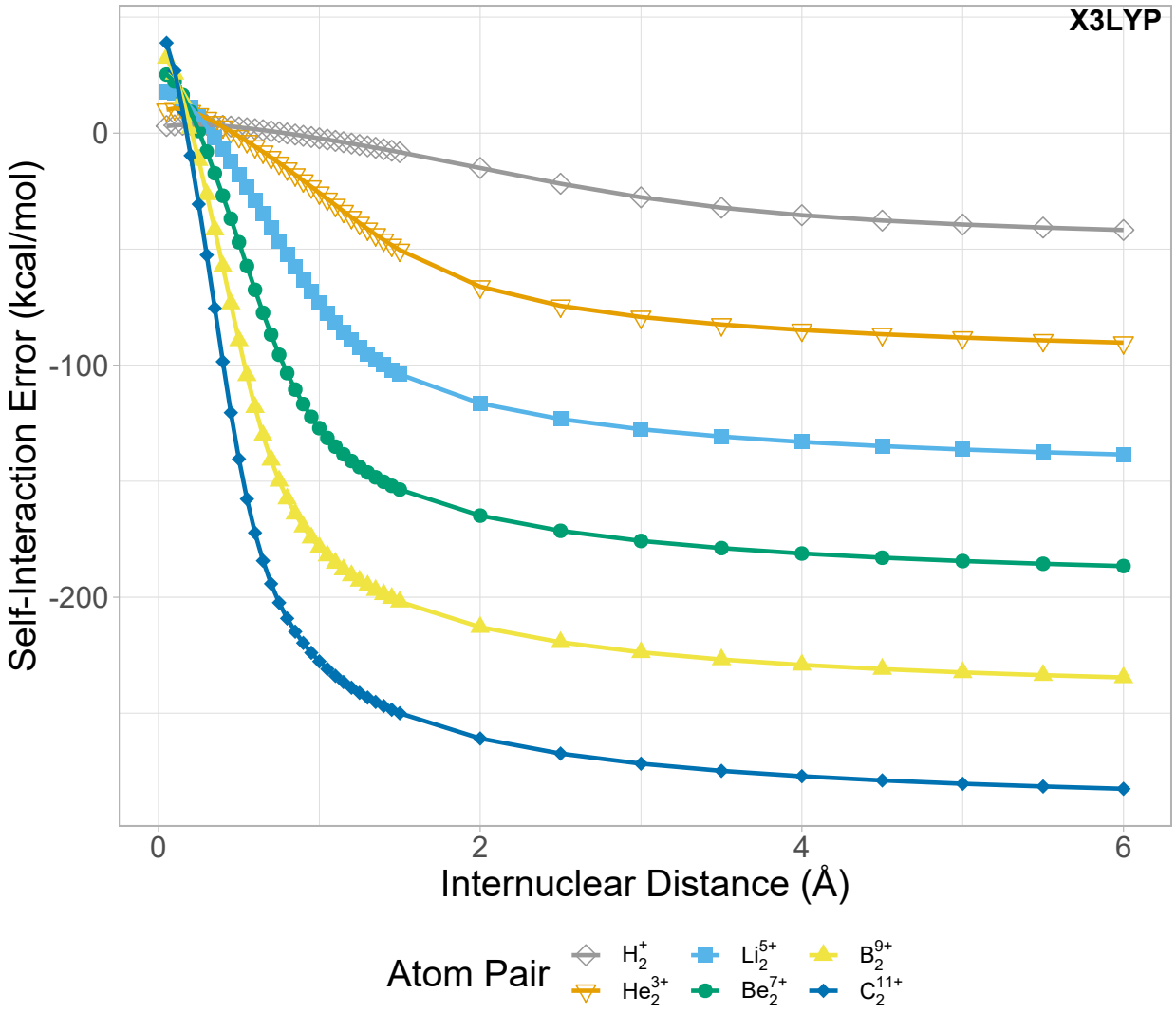

Figure S145: Self-interaction error vs internuclear distance for incremental homonuclear pairs for X3LYP.

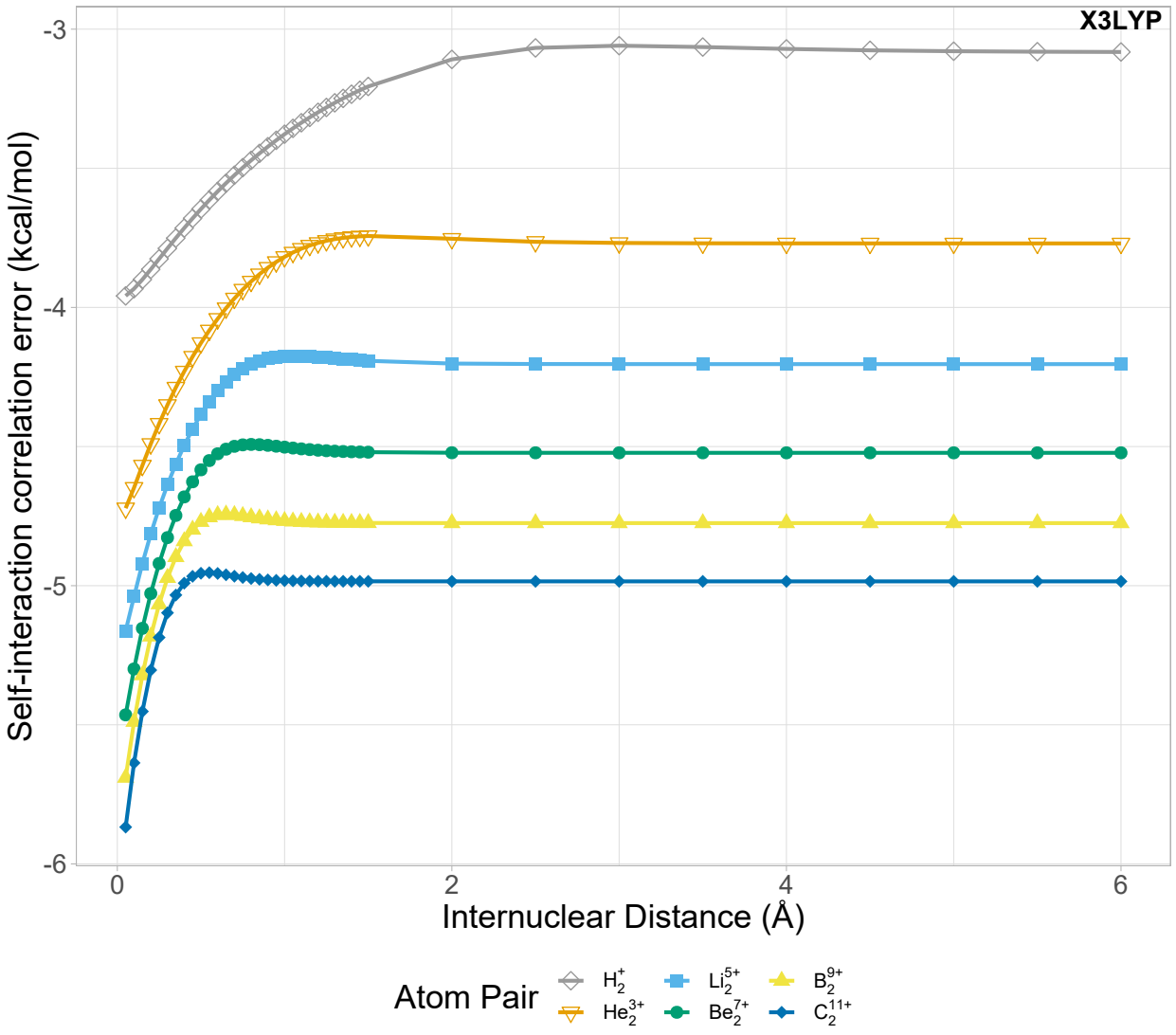

Figure S146: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for X3LYP.

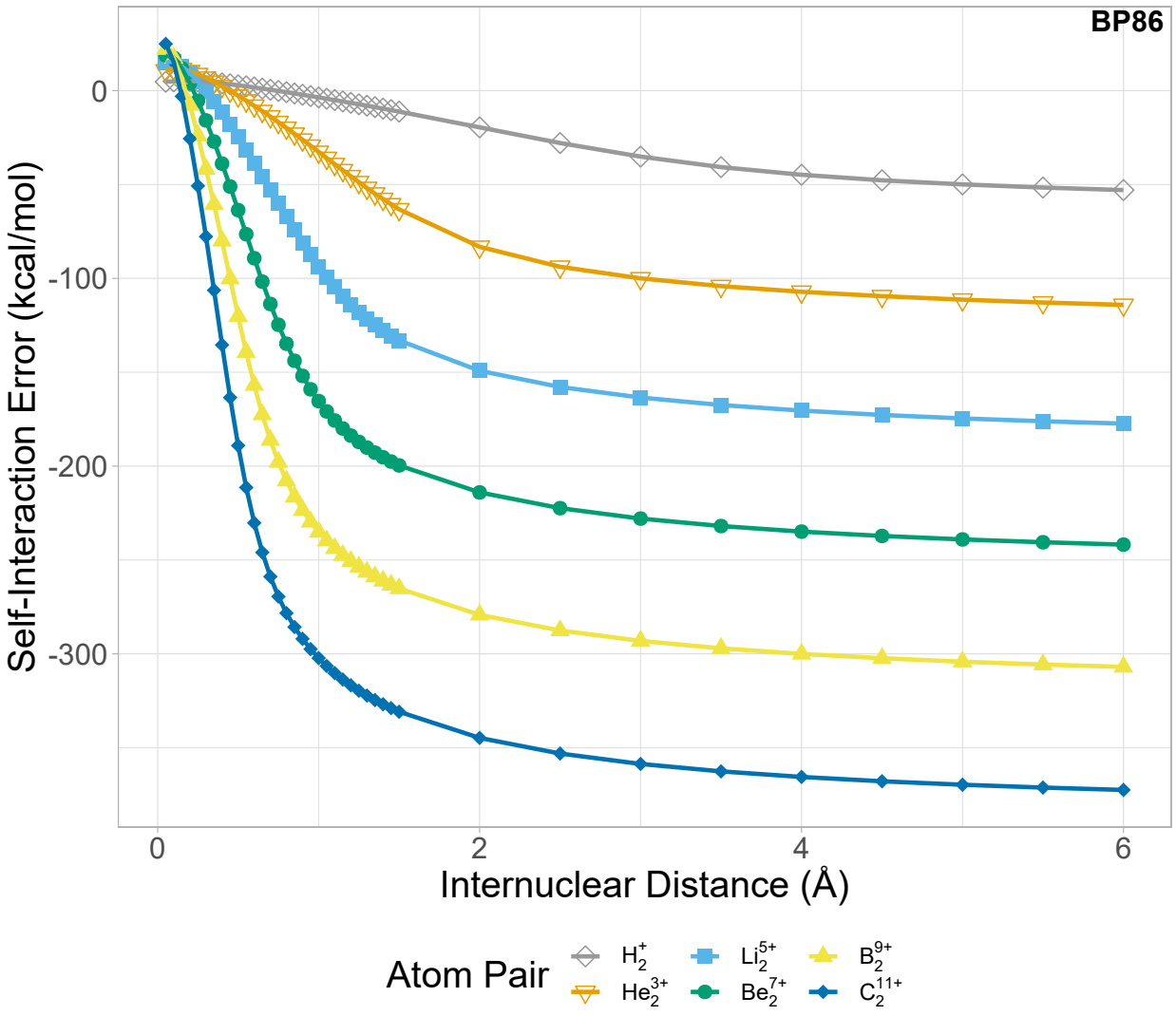

Figure S147: Self-interaction error vs internuclear distance for incremental homonuclear pairs for BP86.

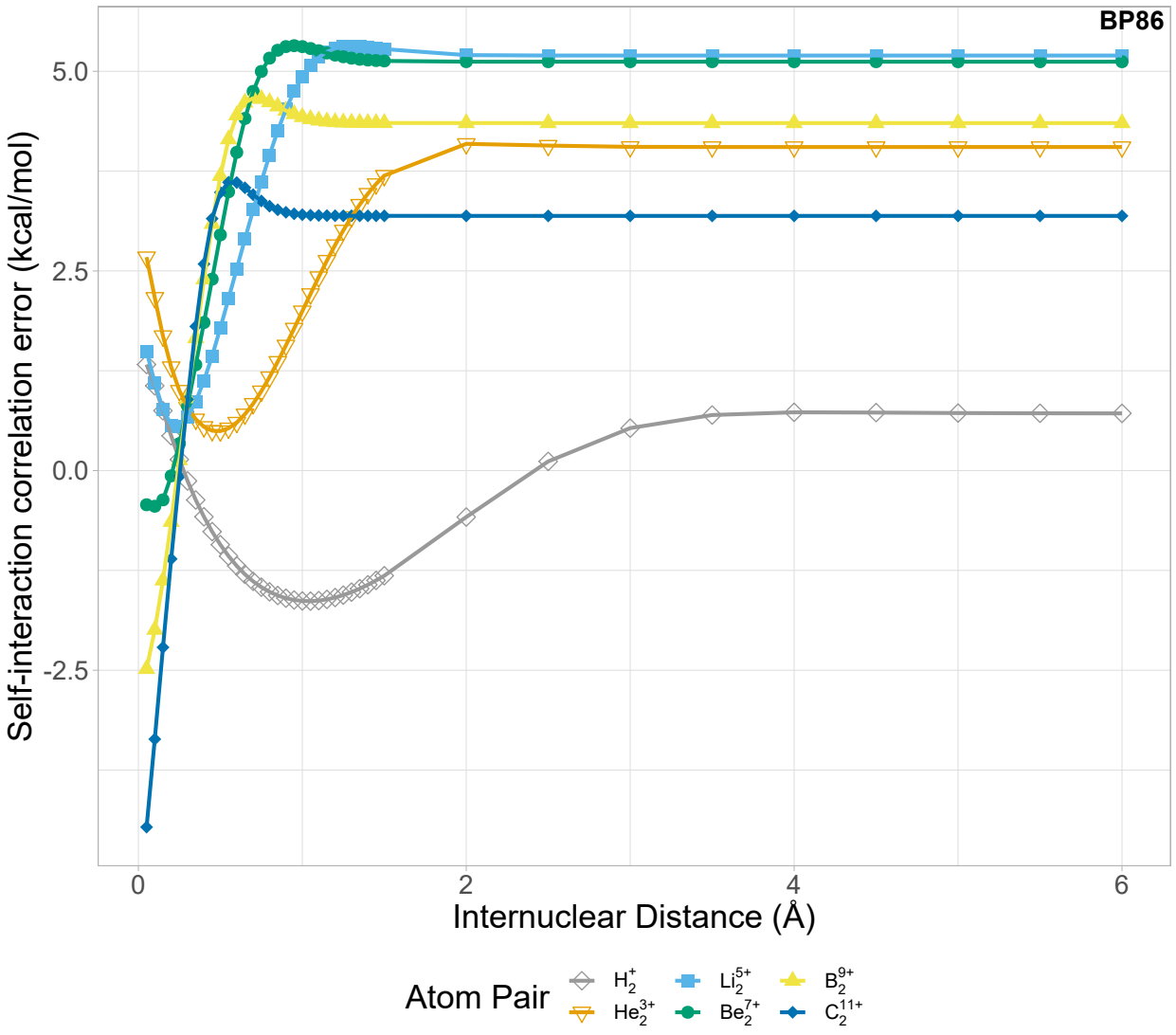

Figure S148: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for BP86.

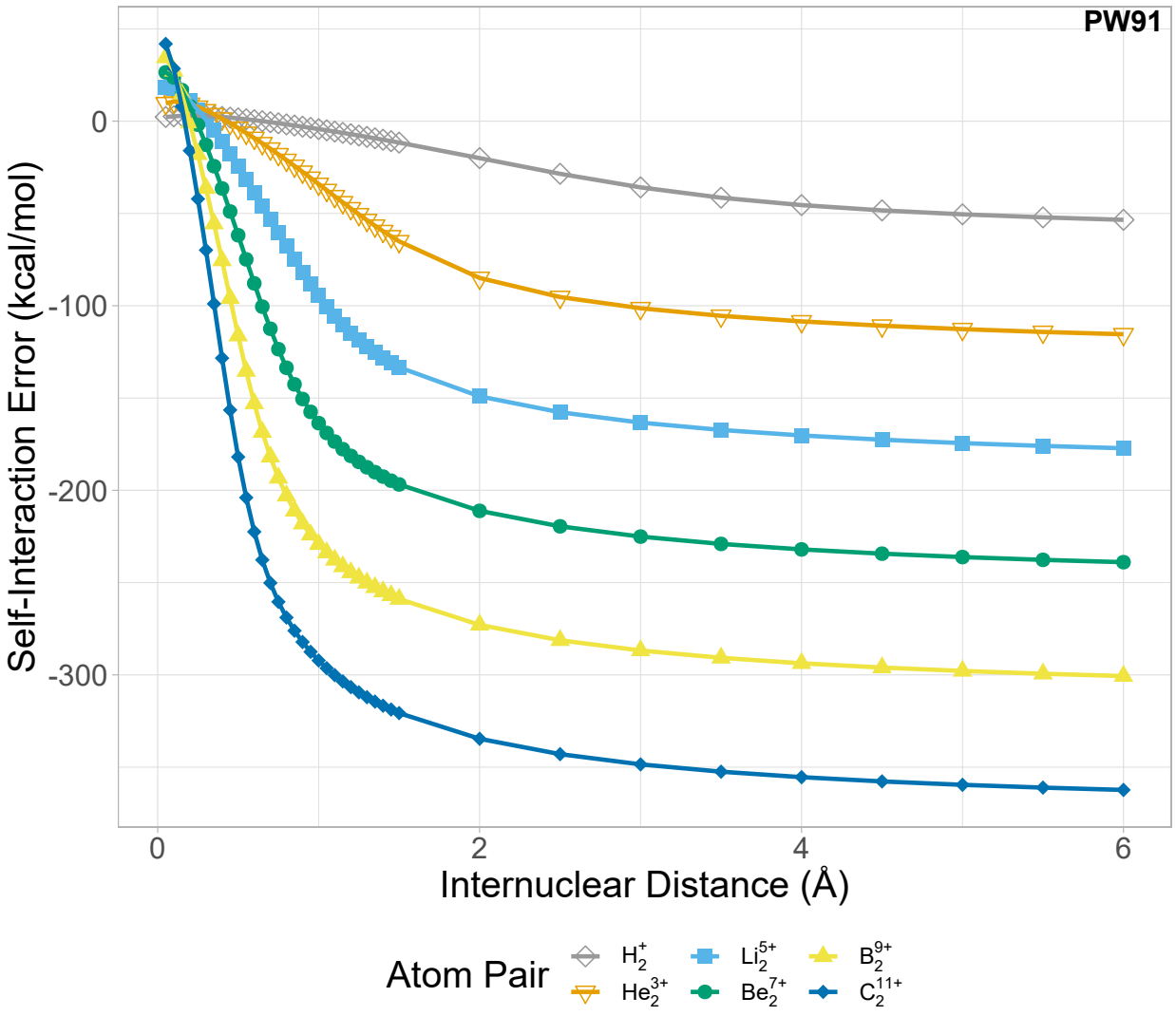

Figure S149: Self-interaction error vs internuclear distance for incremental homonuclear pairs for PW91.

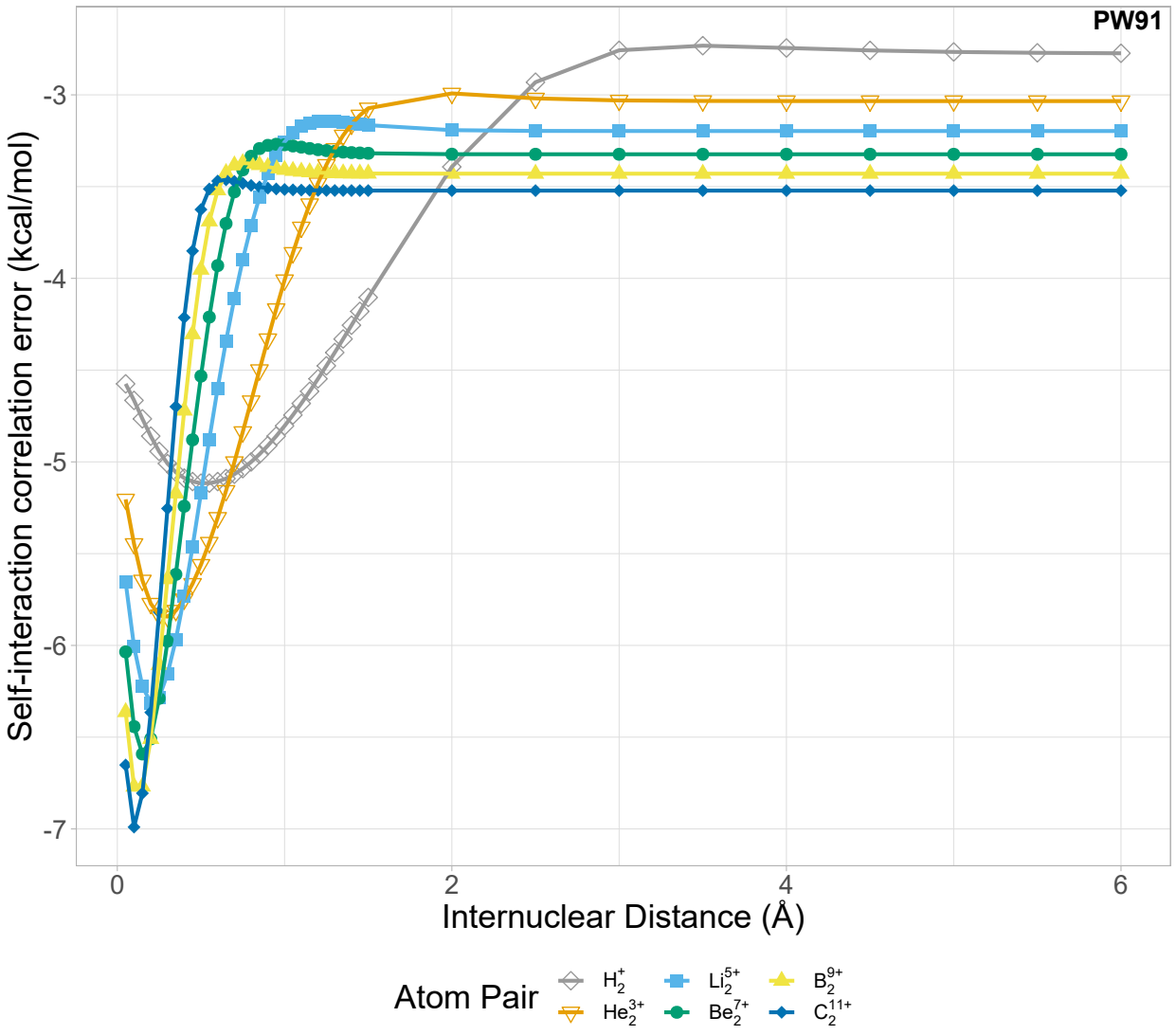

Figure S150: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for PW91.

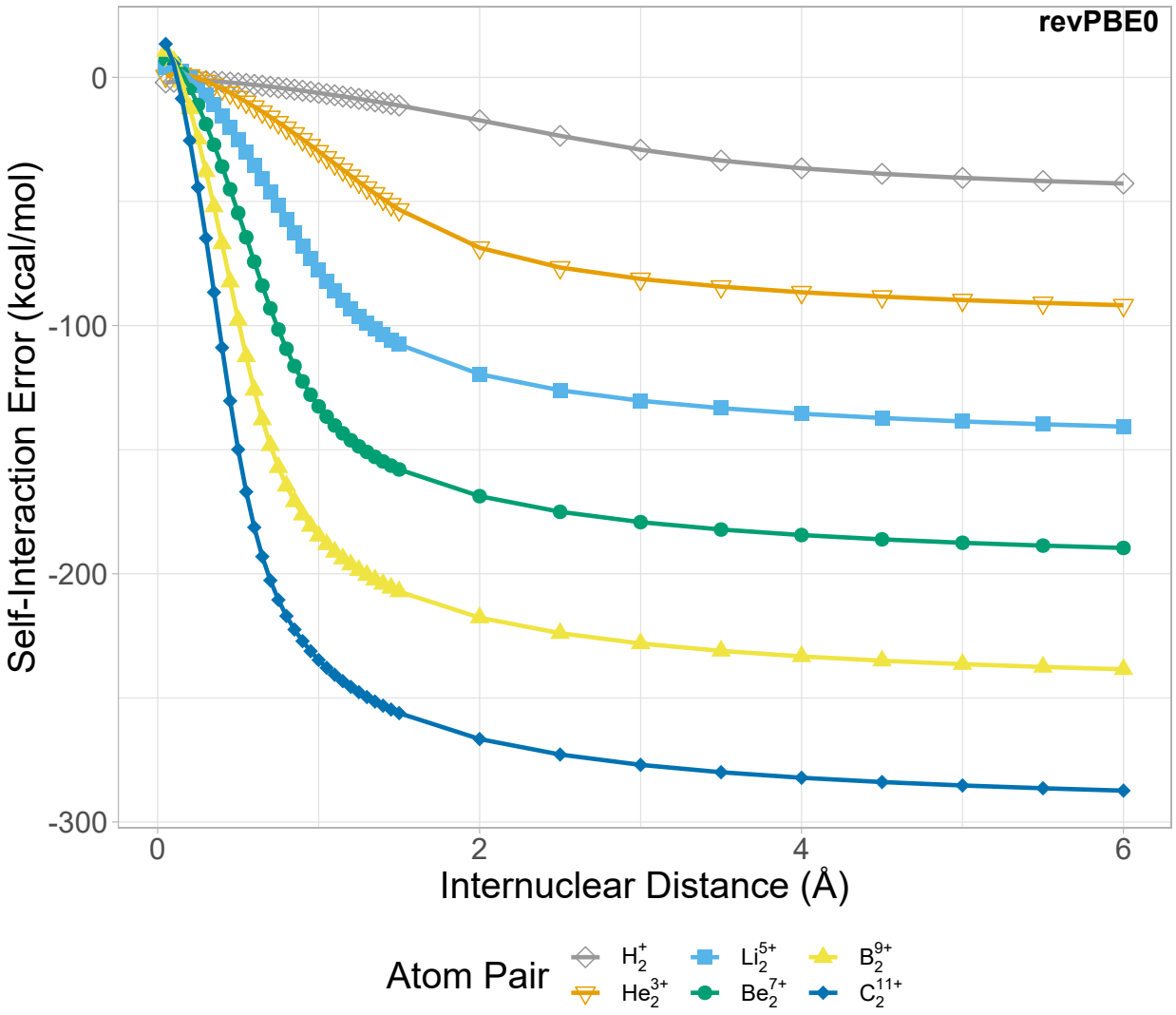

Figure S151: Self-interaction error vs internuclear distance for incremental homonuclear pairs for revPBE0.

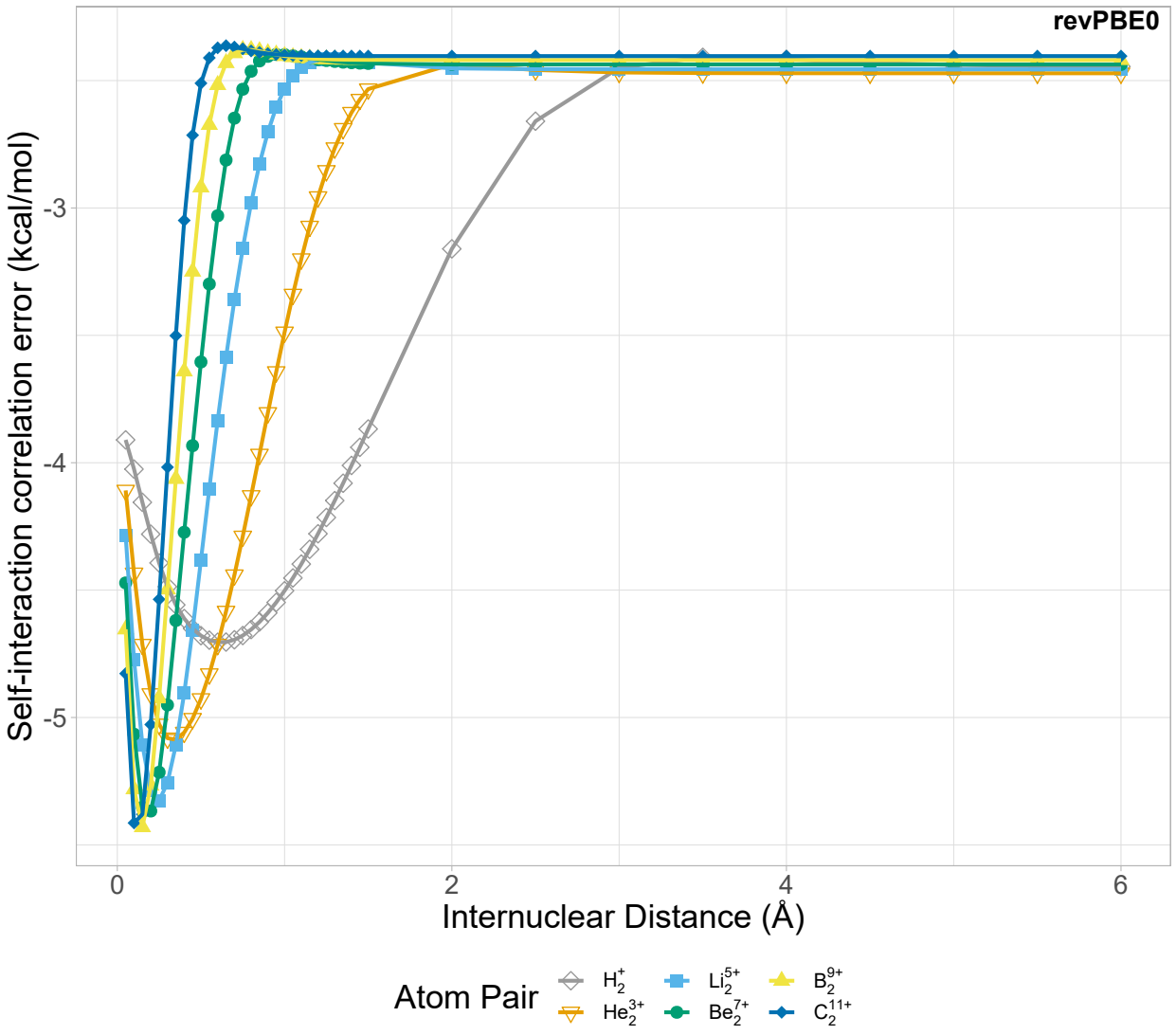

Figure S152: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for revPBE0.

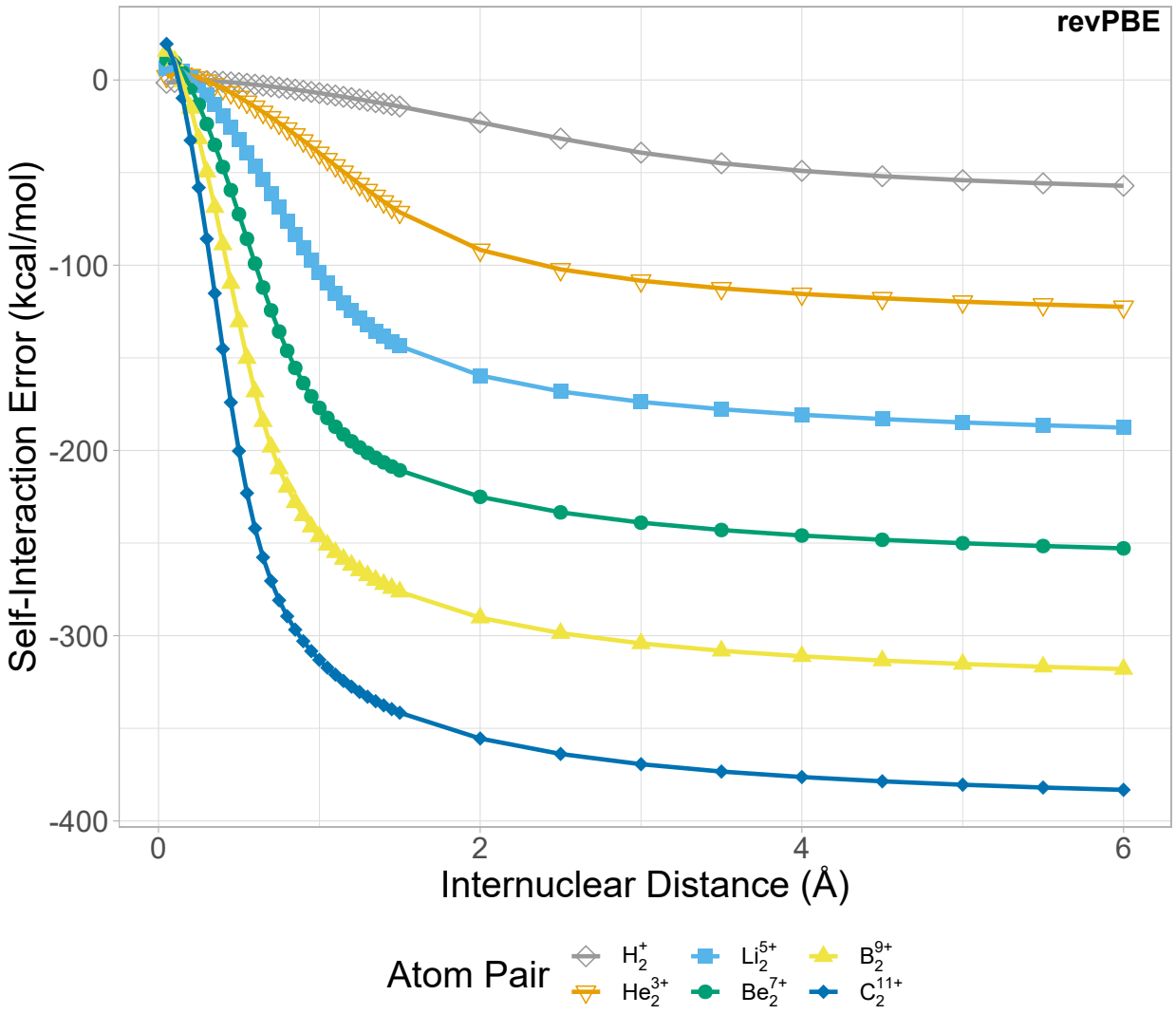

Figure S153: Self-interaction error vs internuclear distance for incremental homonuclear pairs for revPBE.

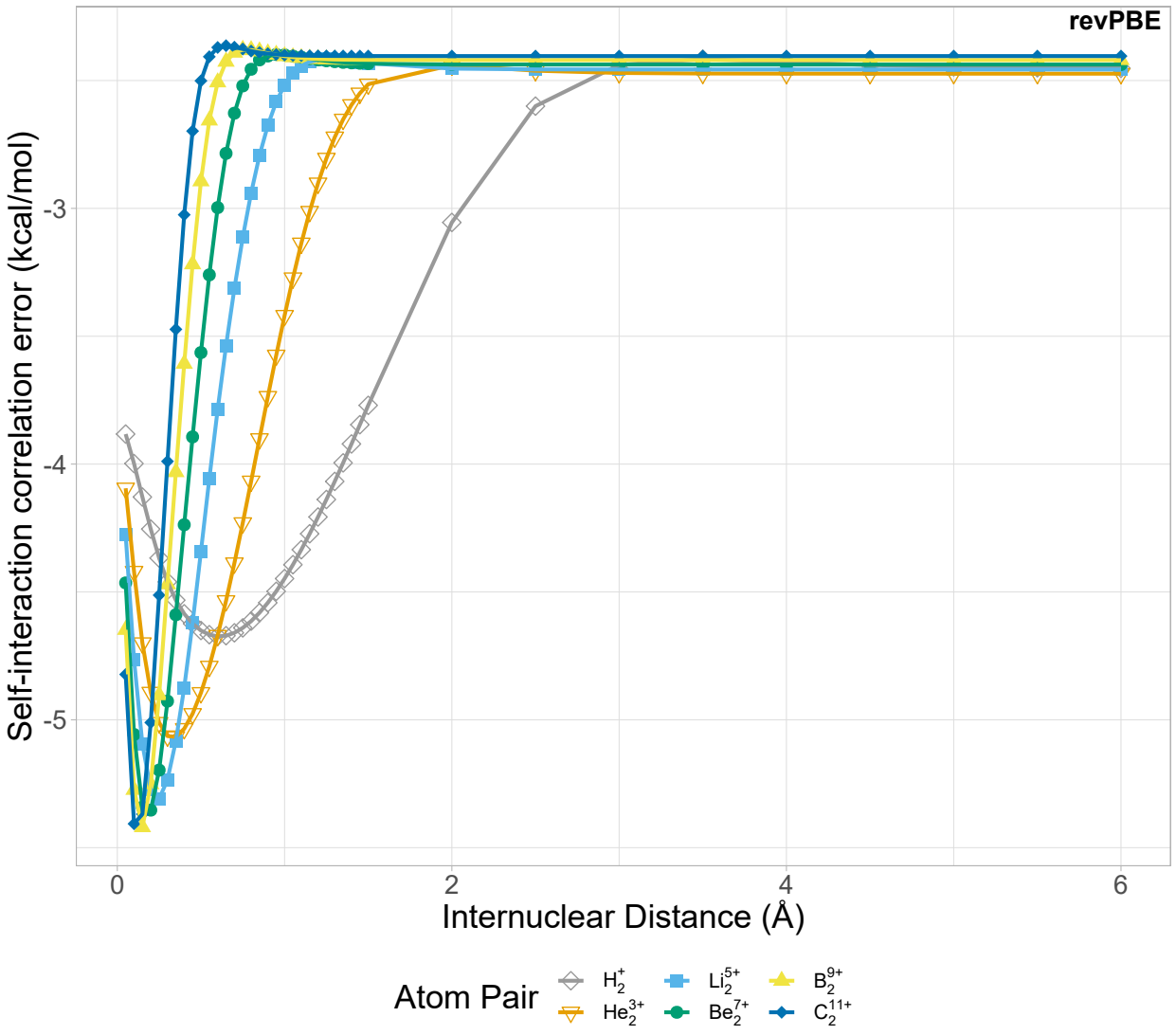

Figure S154: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for revPBE.

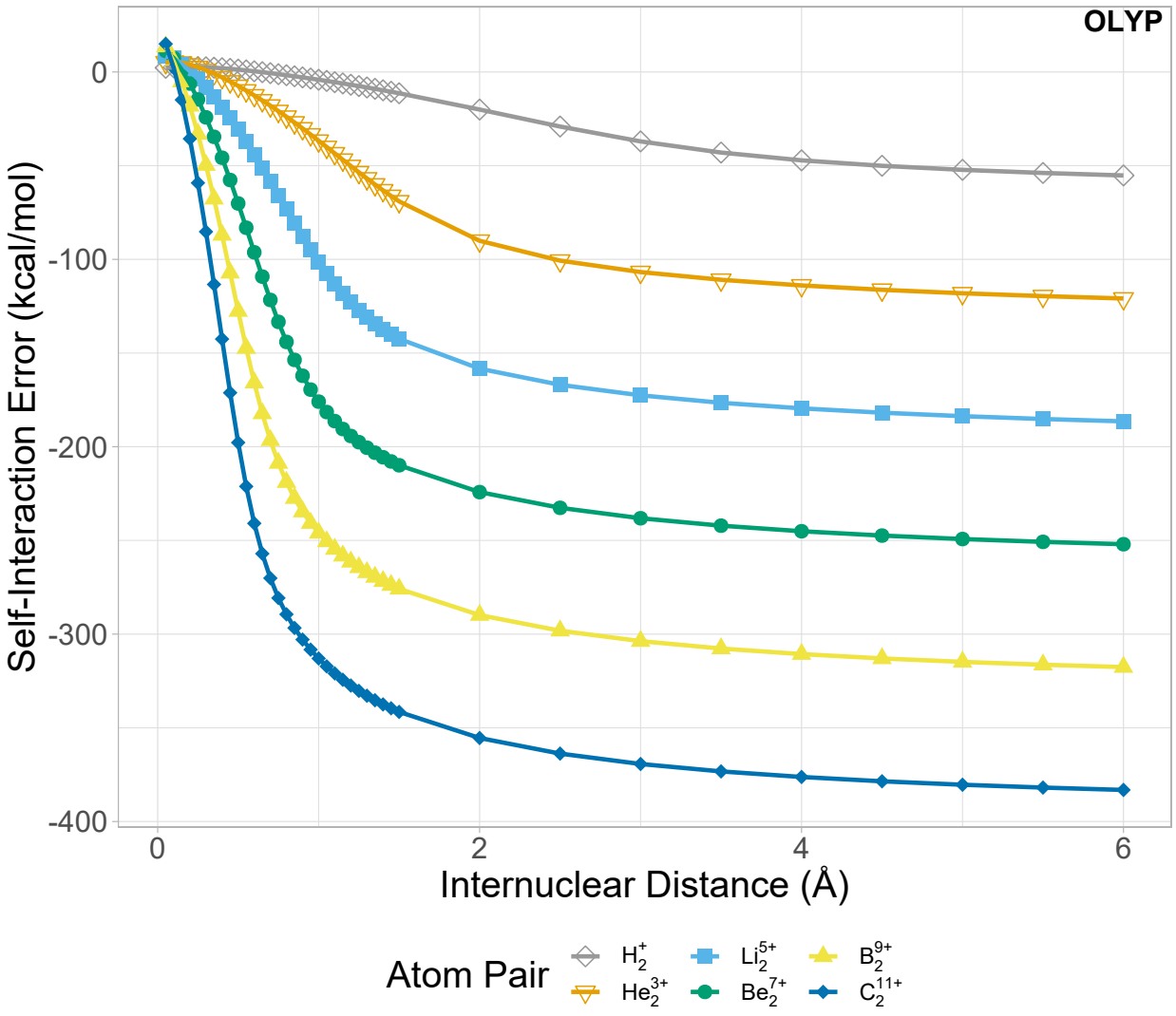

Figure S155: Self-interaction error vs internuclear distance for incremental homonuclear pairs for OLYP.

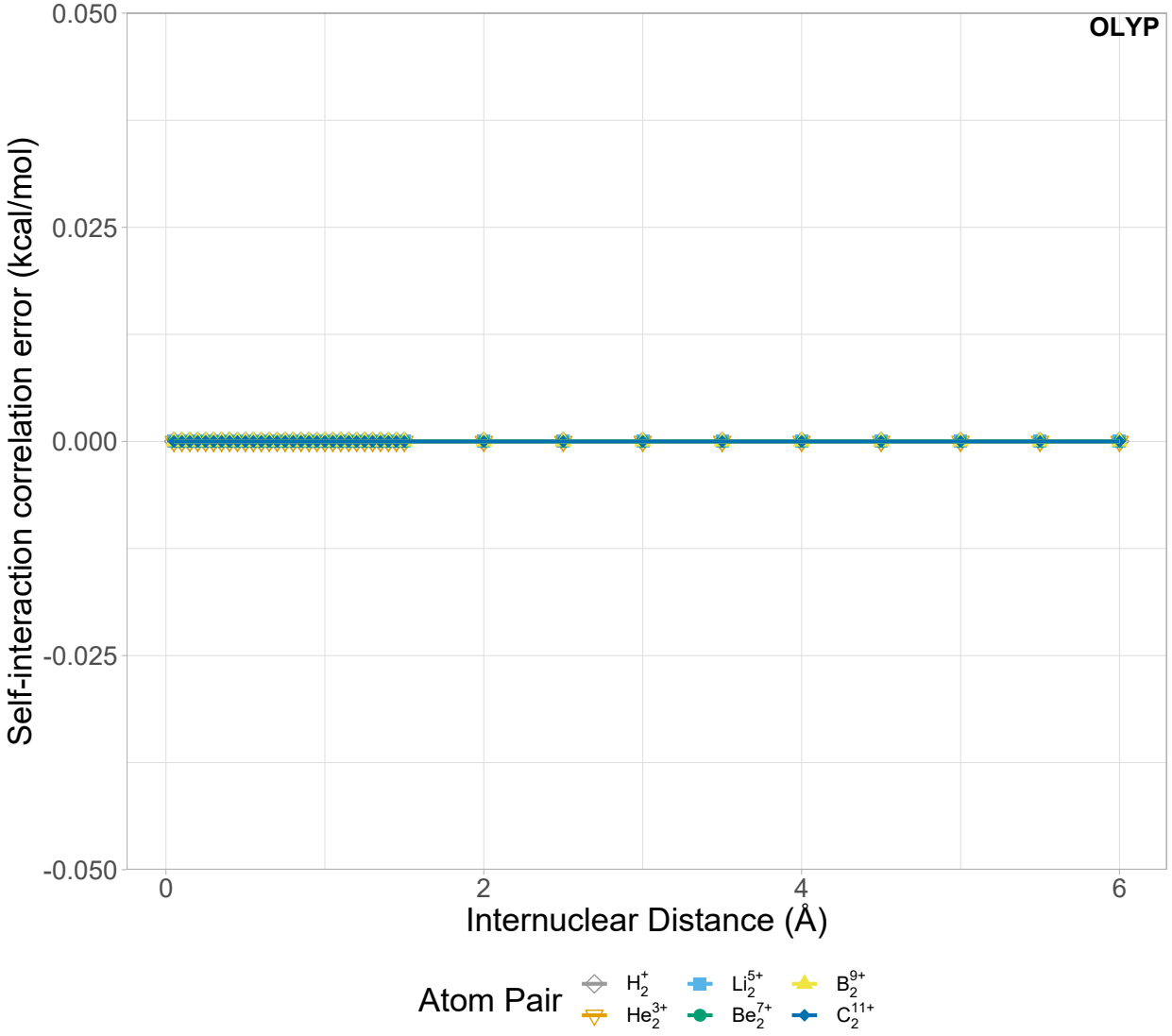

Figure S156: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for OLYP.

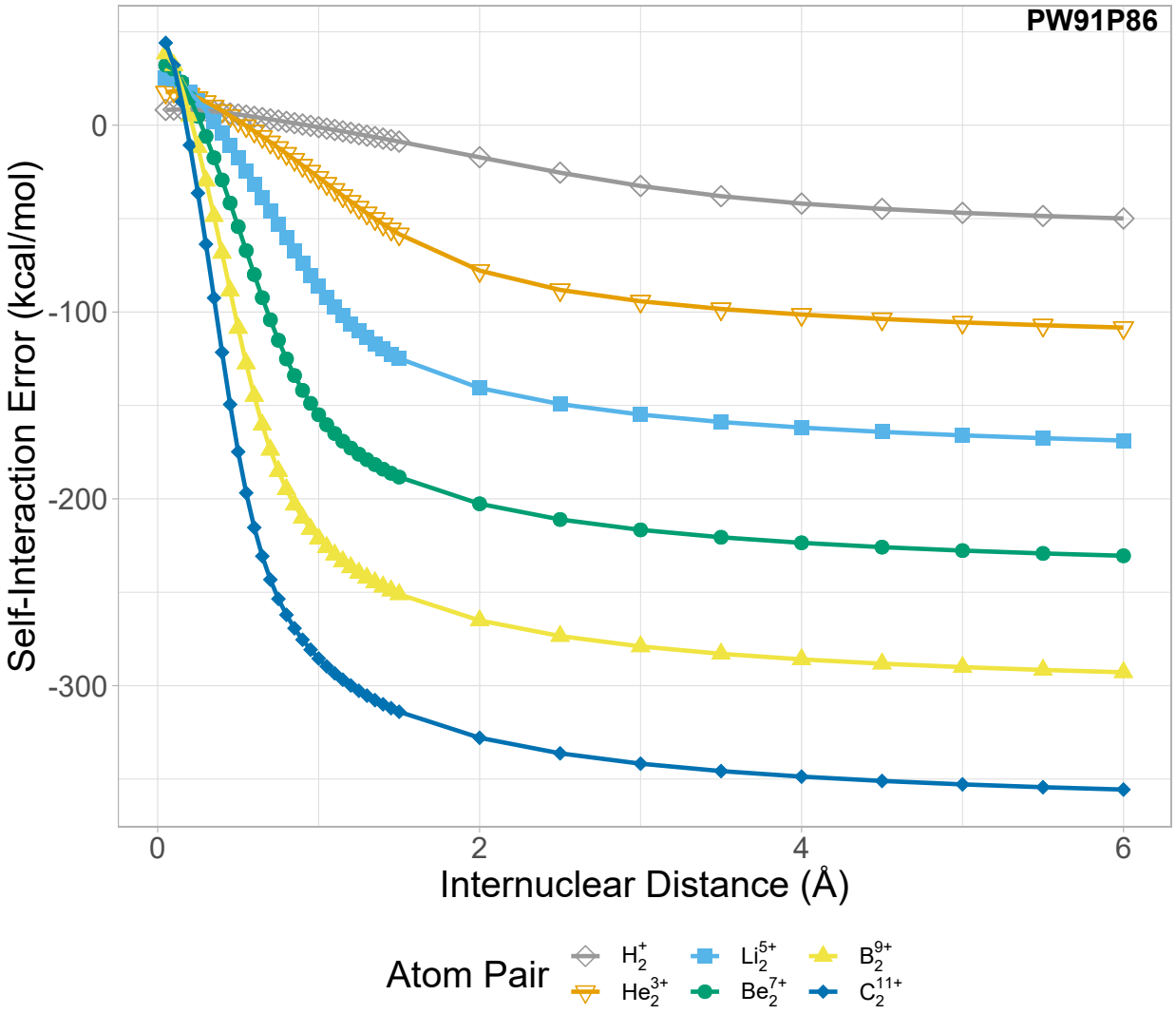

Figure S157: Self-interaction error vs internuclear distance for incremental homonuclear pairs for PW91P86.

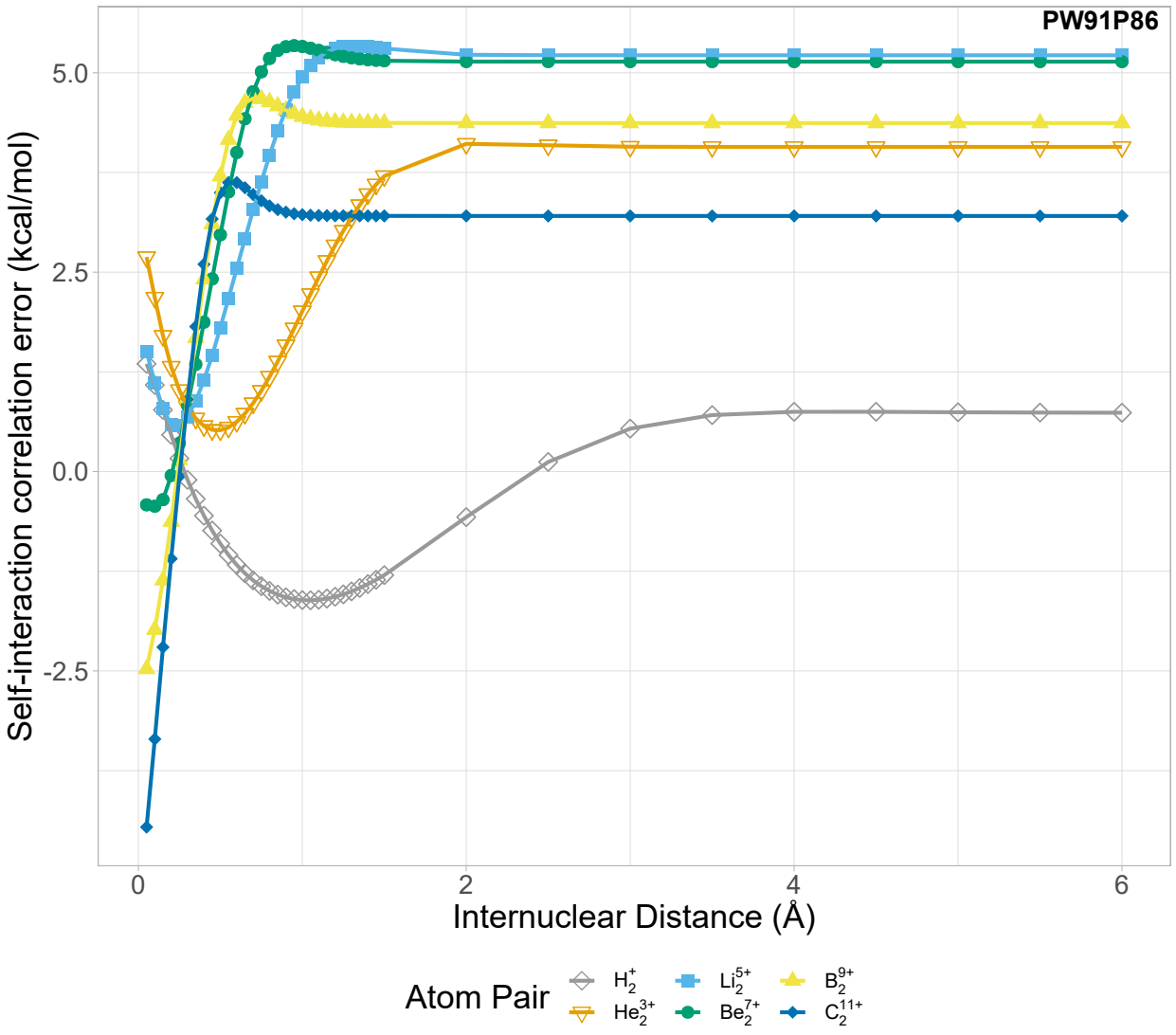

Figure S158: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for PW91P86.

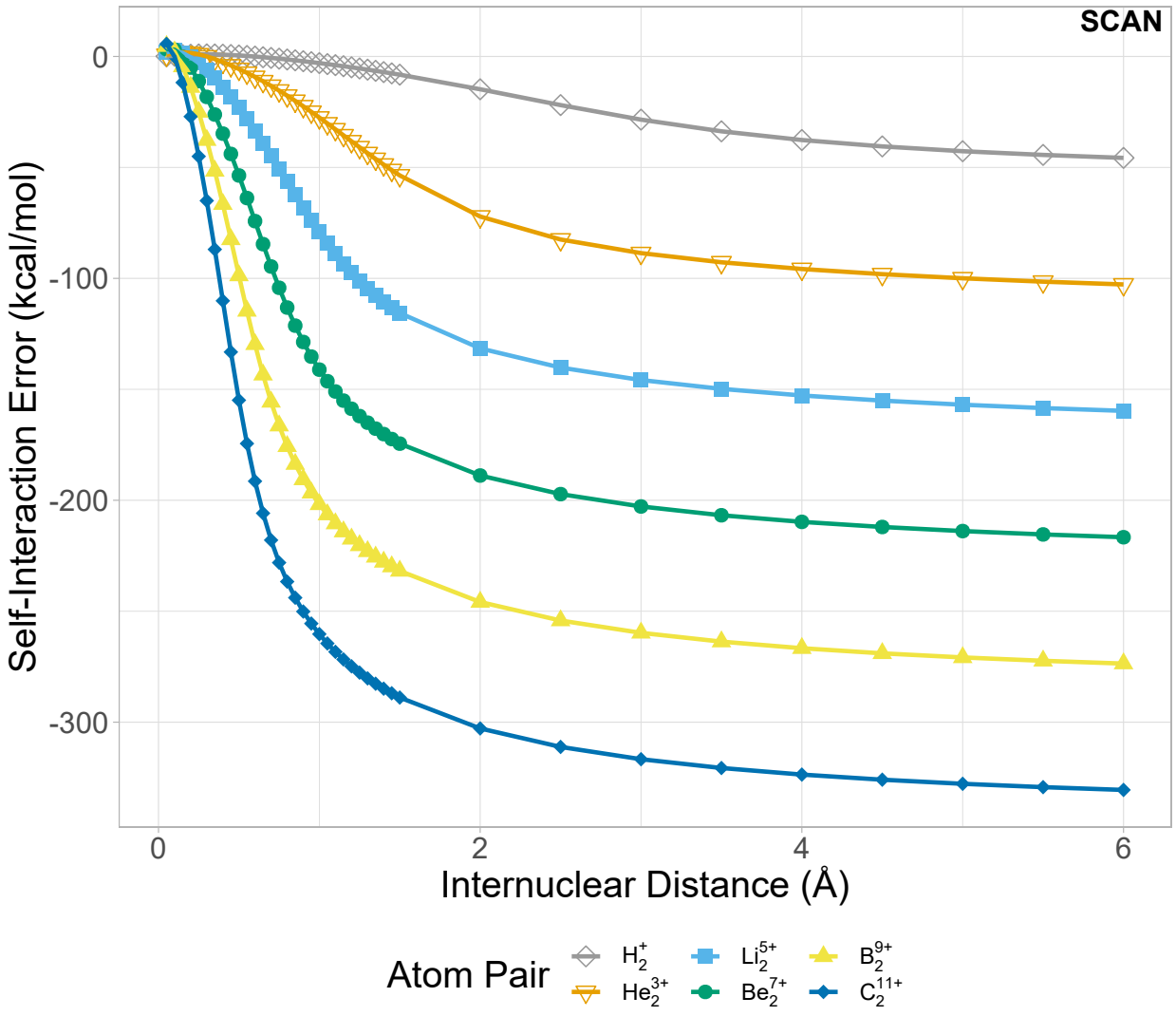

Figure S159: Self-interaction error vs internuclear distance for incremental homonuclear pairs for SCAN.

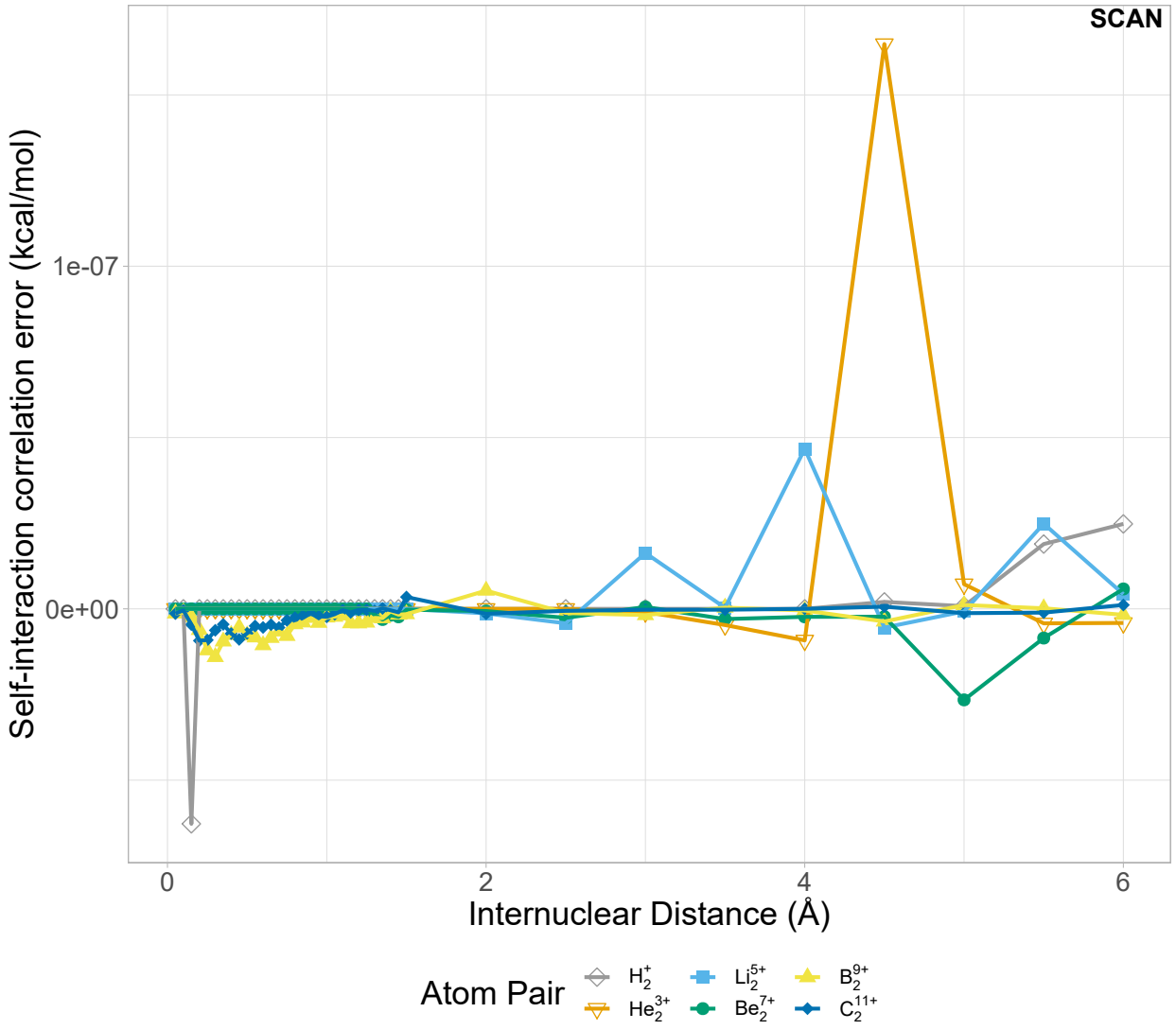

Figure S160: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for SCAN. Formally, this DFA is self-correlation free, any deviations from this is likely computational noise.

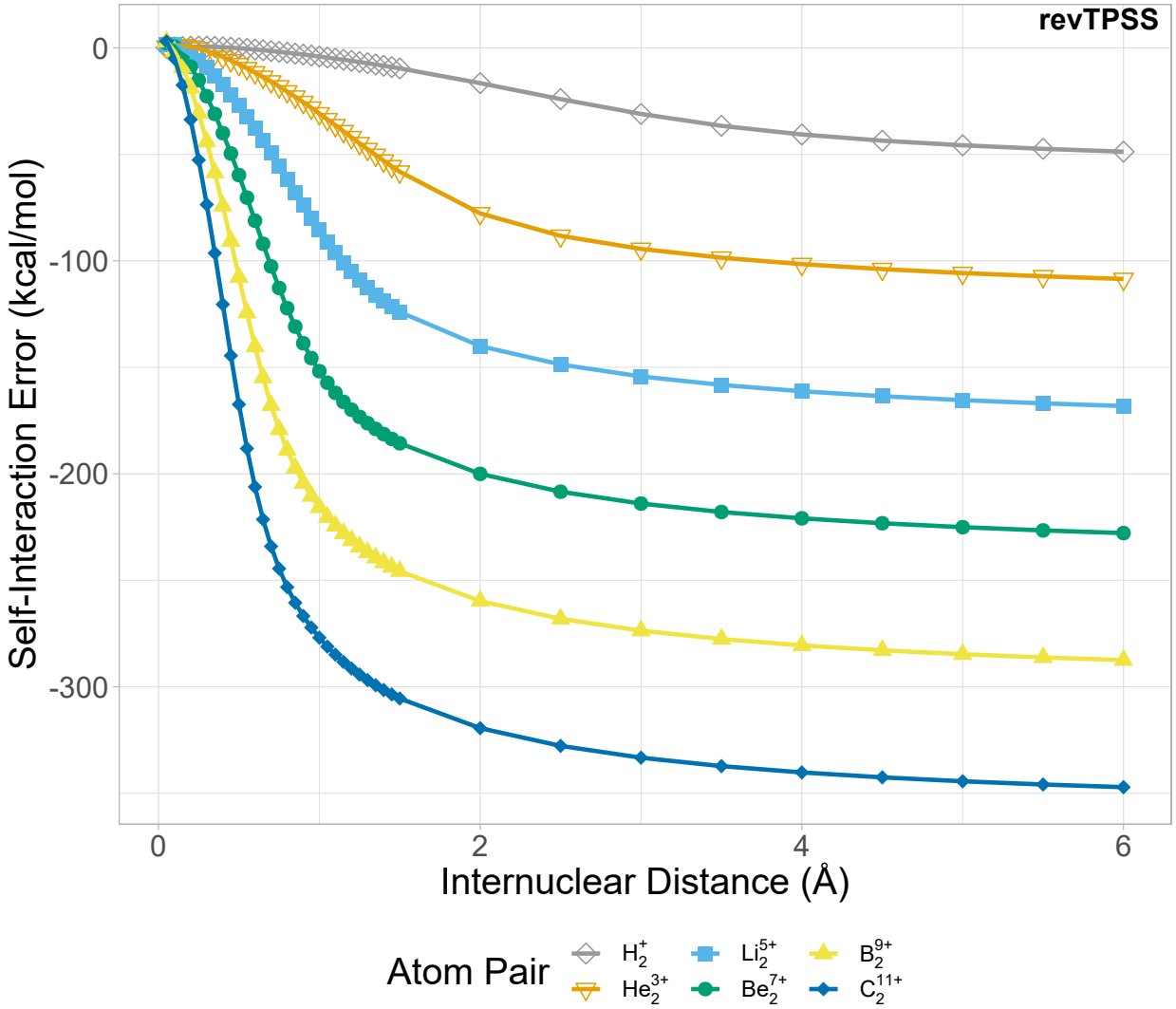

Figure S161: Self-interaction error vs internuclear distance for incremental homonuclear pairs for revTPSS. Formally, this DFA is self-correlation free, any deviations from this is likely computational noise.

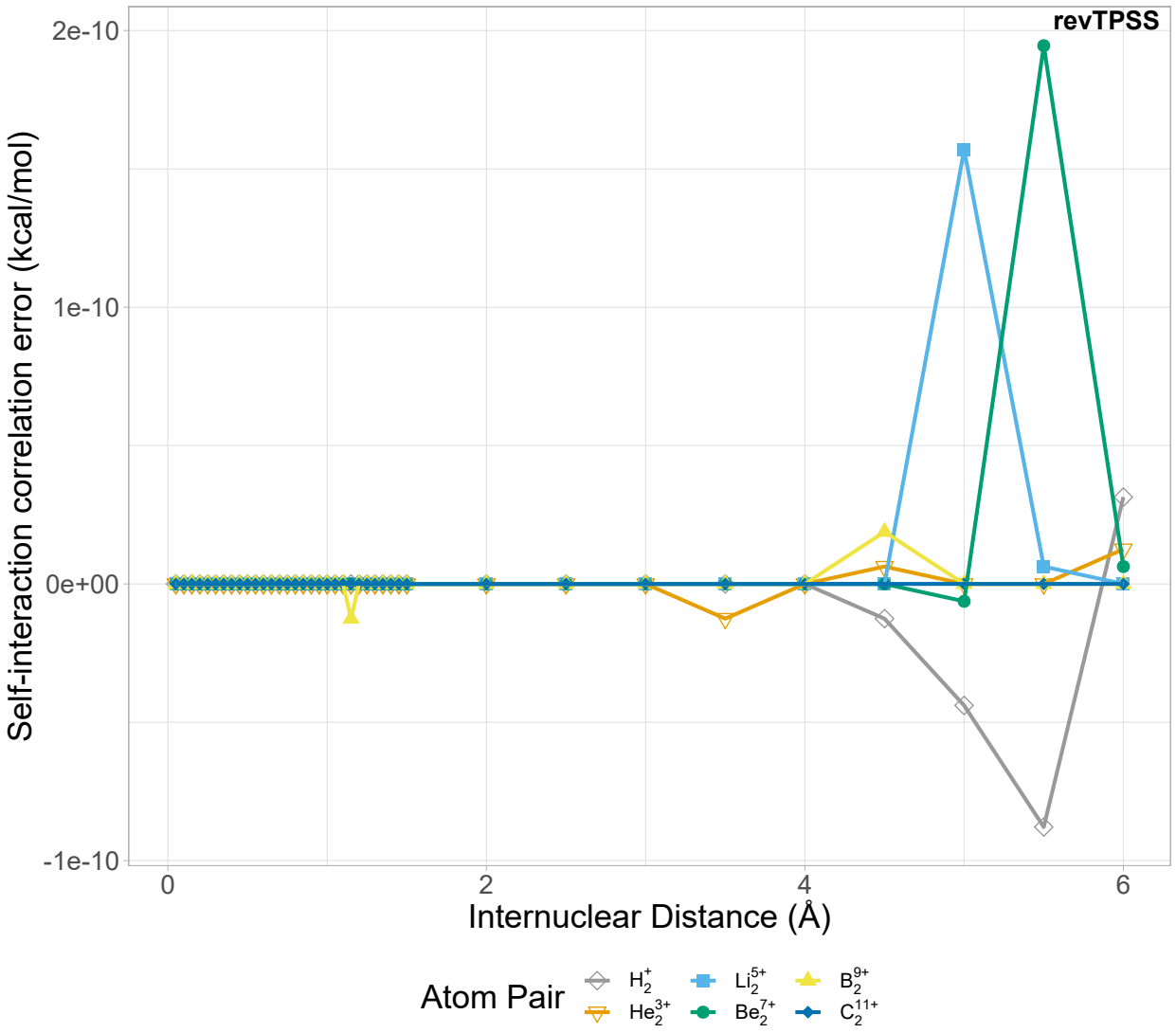

Figure S162: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for revTPSS.

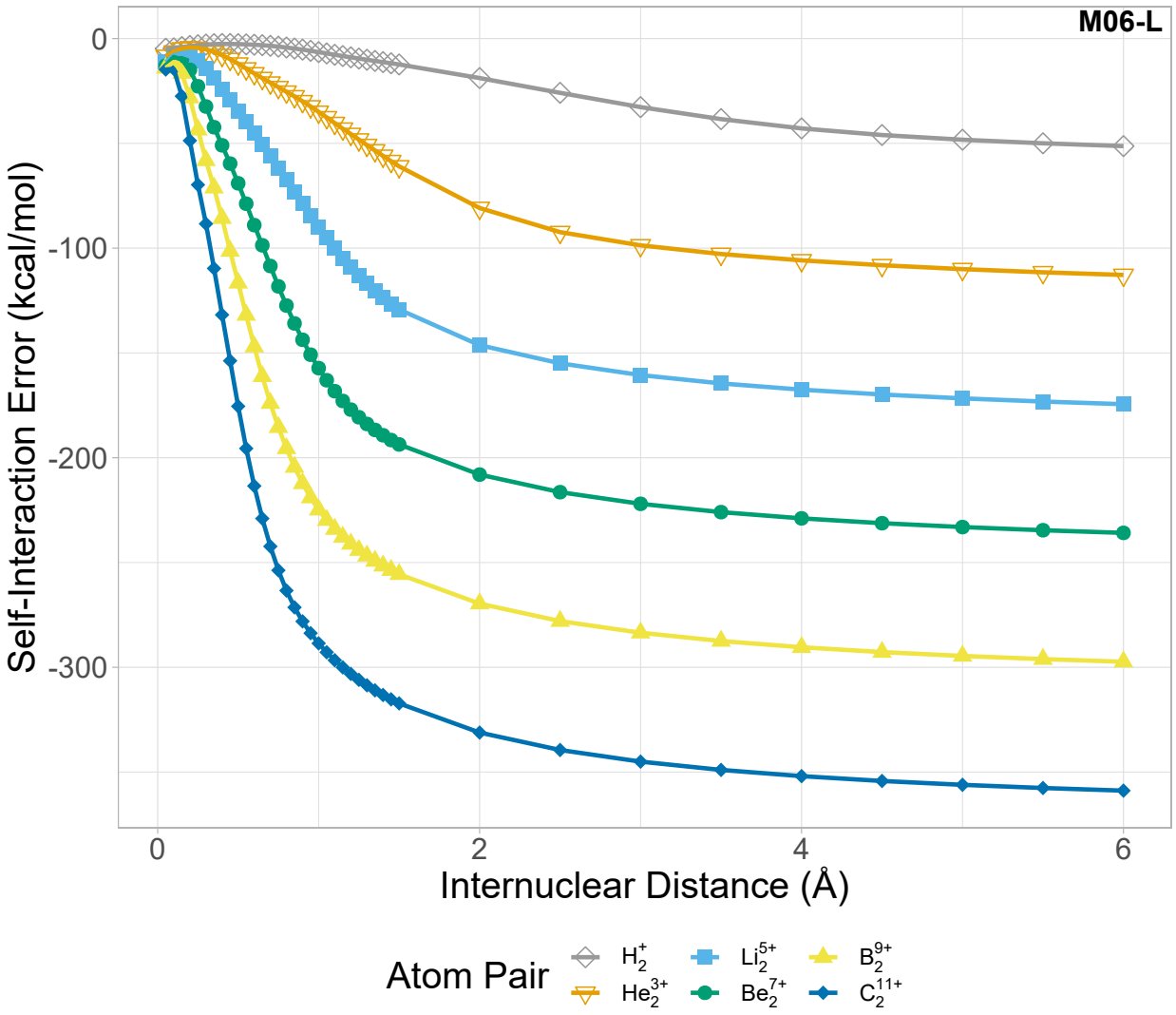

Figure S163: Self-interaction error vs internuclear distance for incremental homonuclear pairs for M06-L.

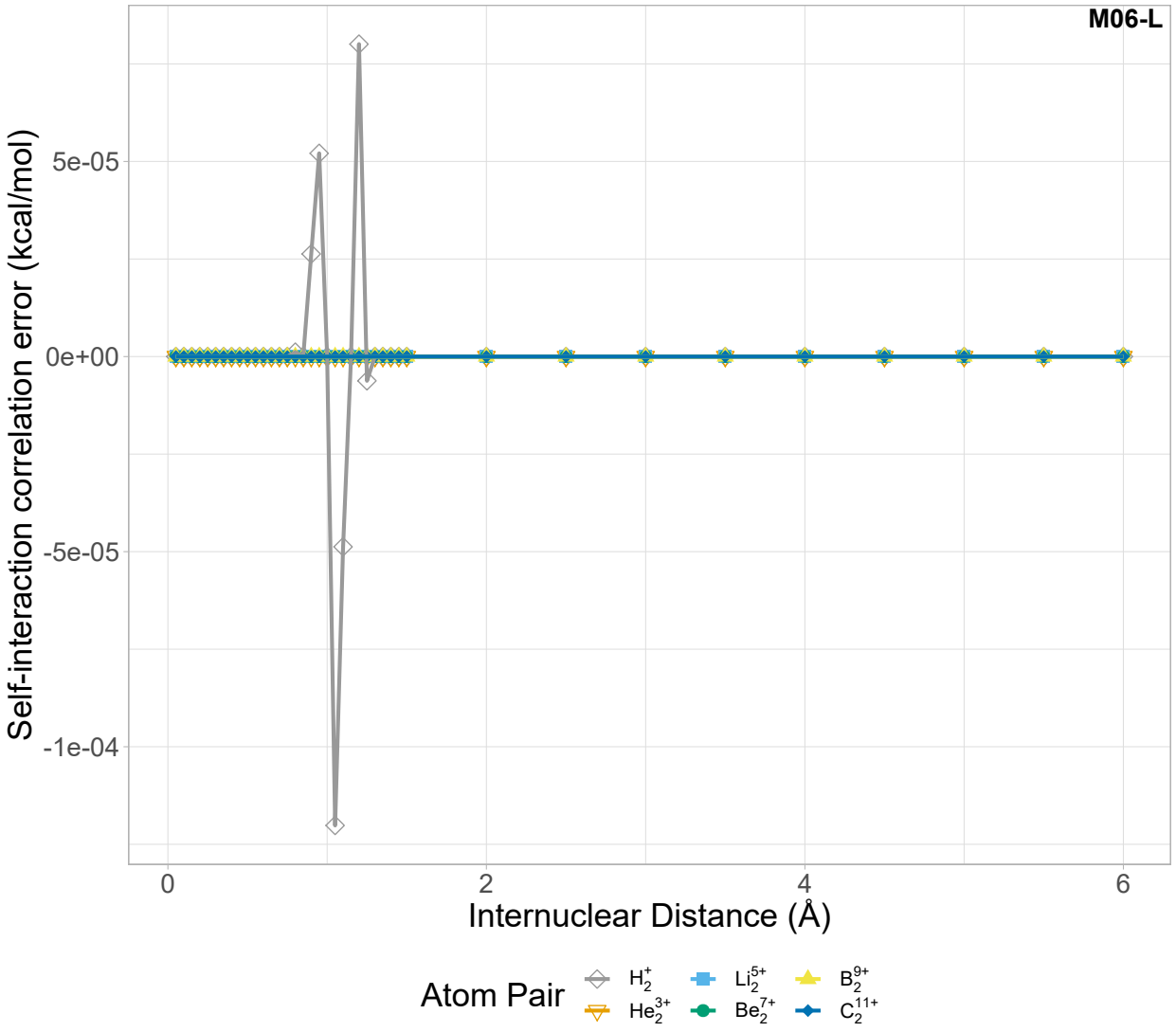

Figure S164: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for M06-L. Formally, this DFA is self-correlation free, any deviations from this is likely computational noise.

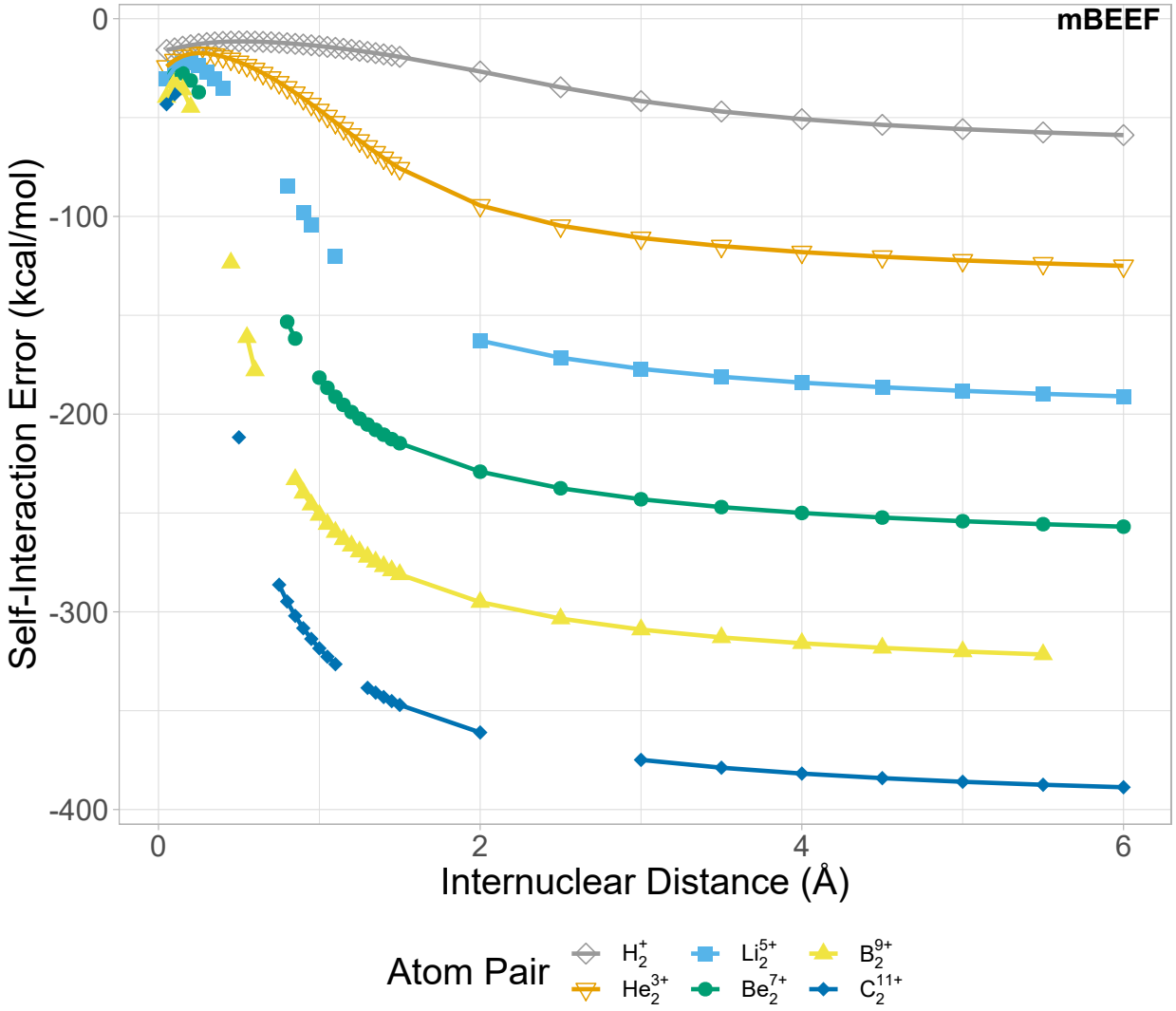

Figure S165: Self-interaction error vs internuclear distance for incremental homonuclear pairs for mBEEF. This DFA suffers from convergence issues.

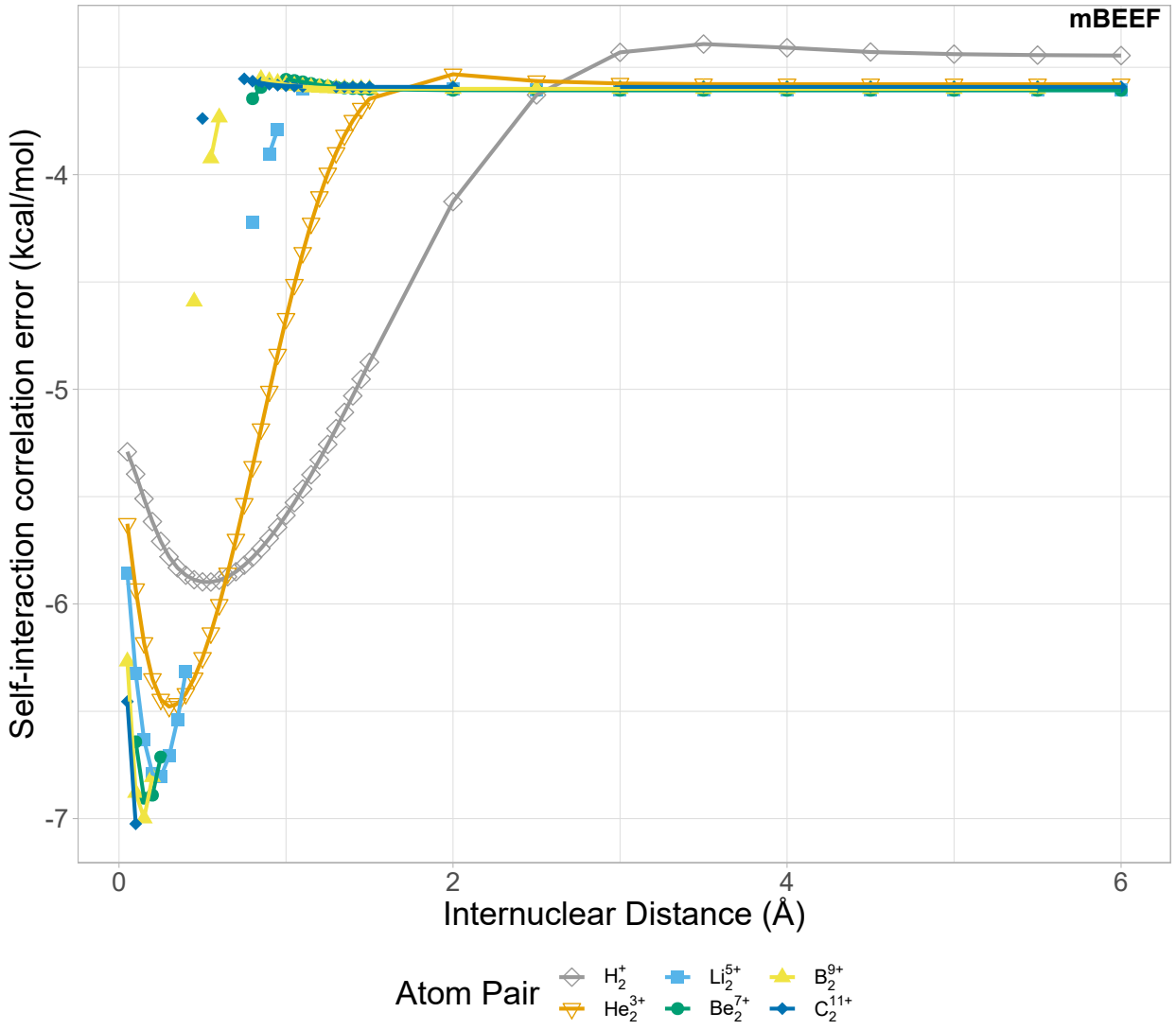

Figure S166: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for mBEEF. This DFA suffers from convergence issues.

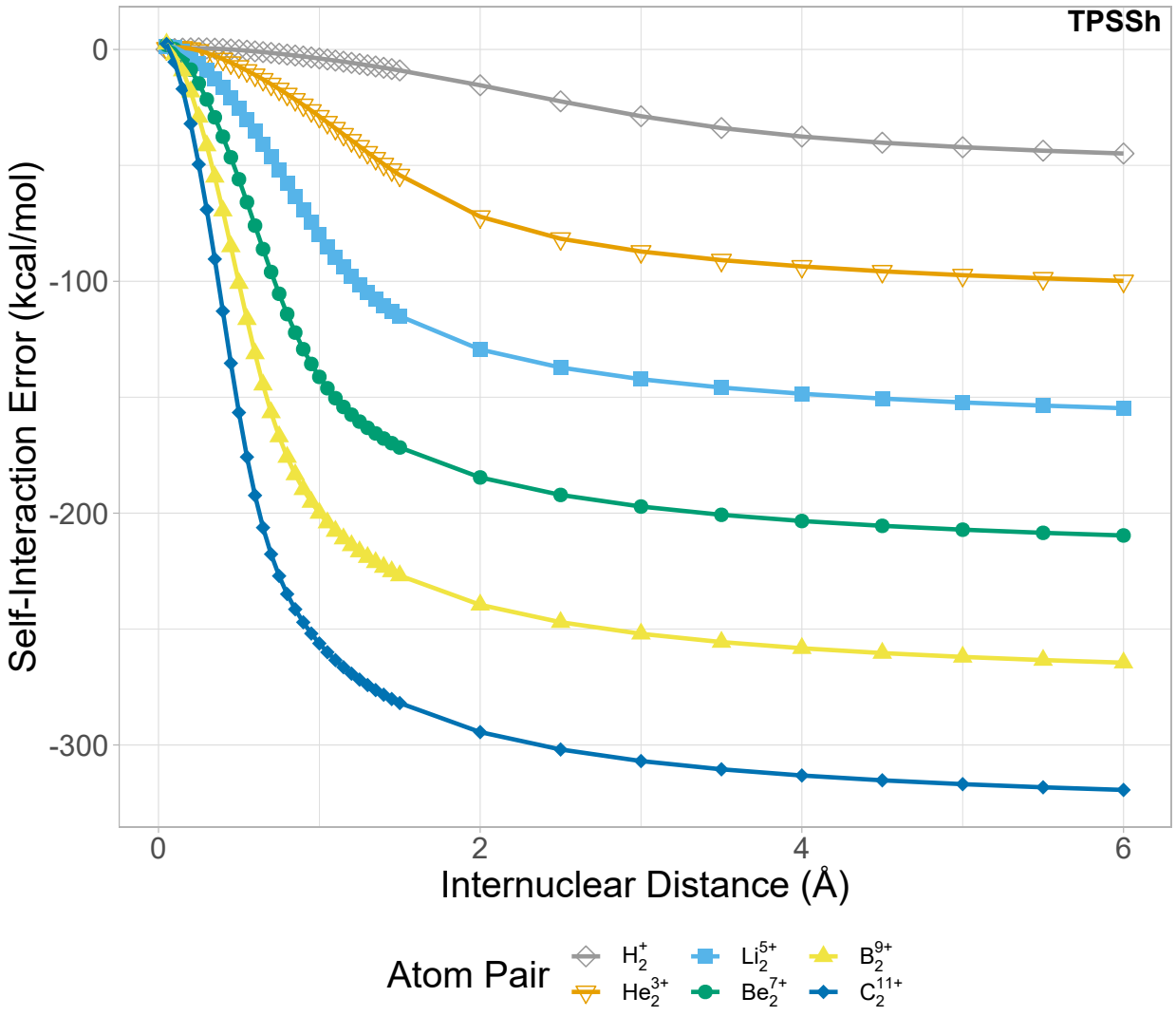

Figure S167: Self-interaction error vs internuclear distance for incremental homonuclear pairs for TPSSh.

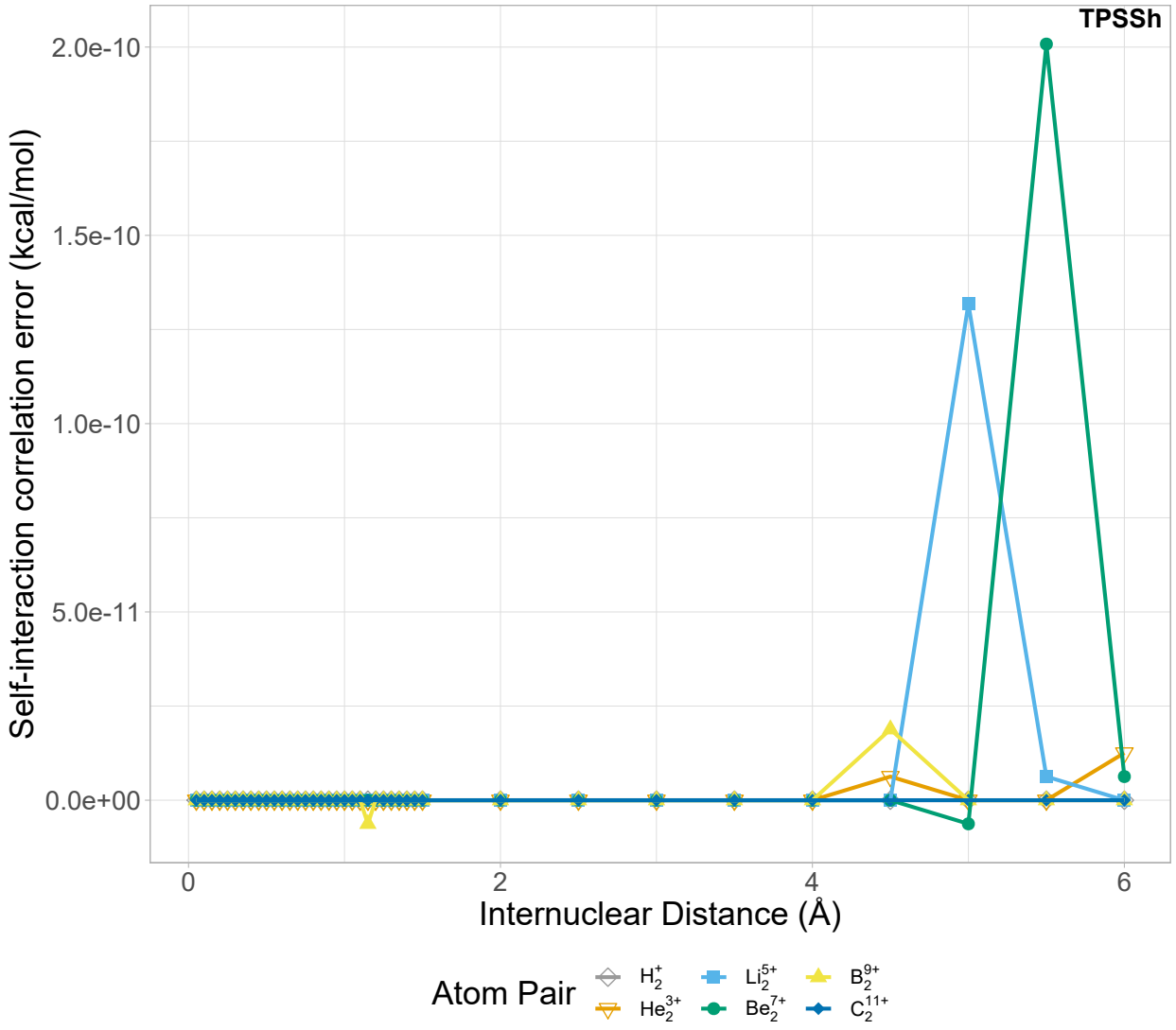

Figure S168: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for TPSSh. Formally, this DFA is self-correlation free, any deviations from this is likely computational noise.

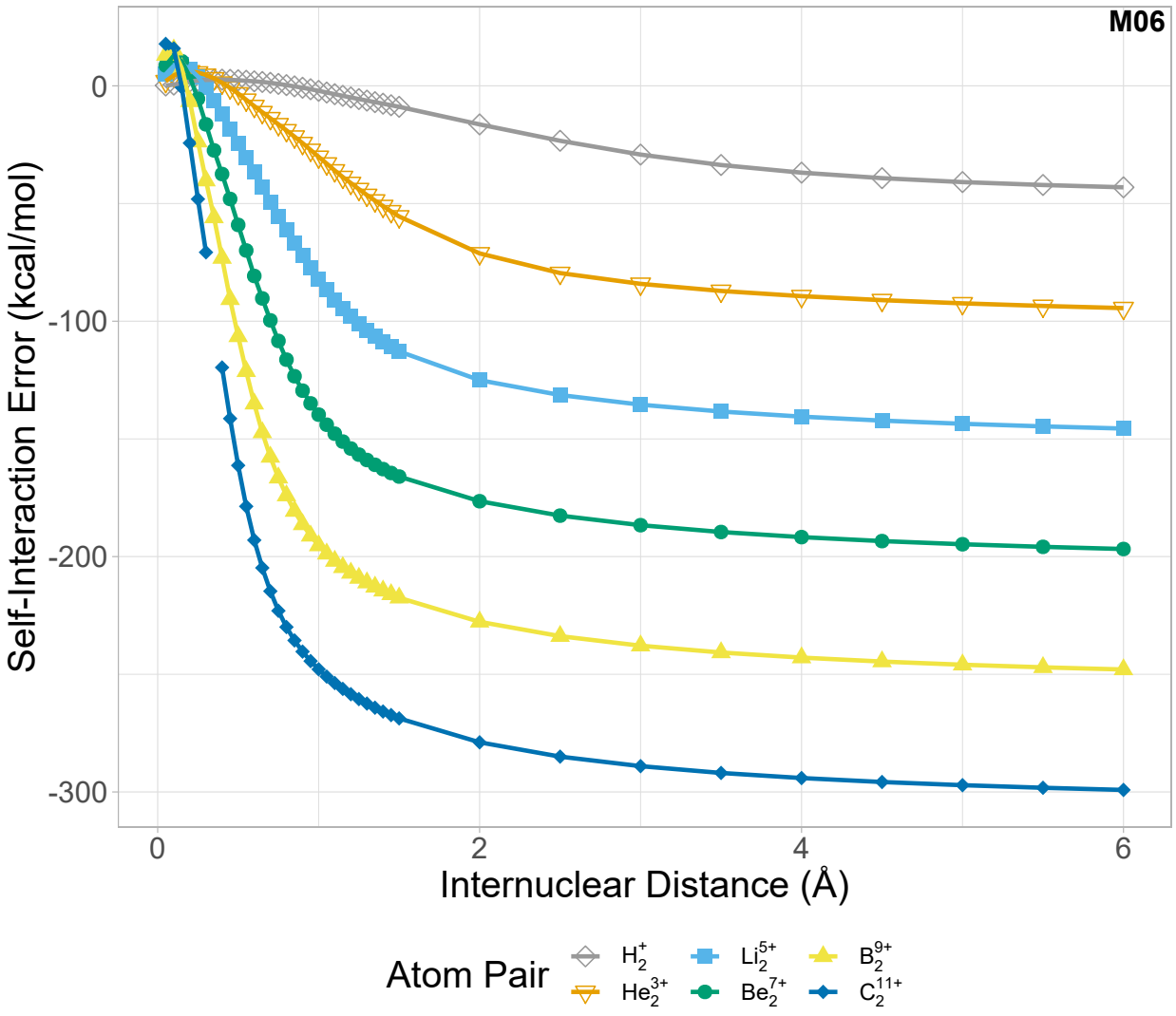

Figure S169: Self-interaction error vs internuclear distance for incremental homonuclear pairs for M06.

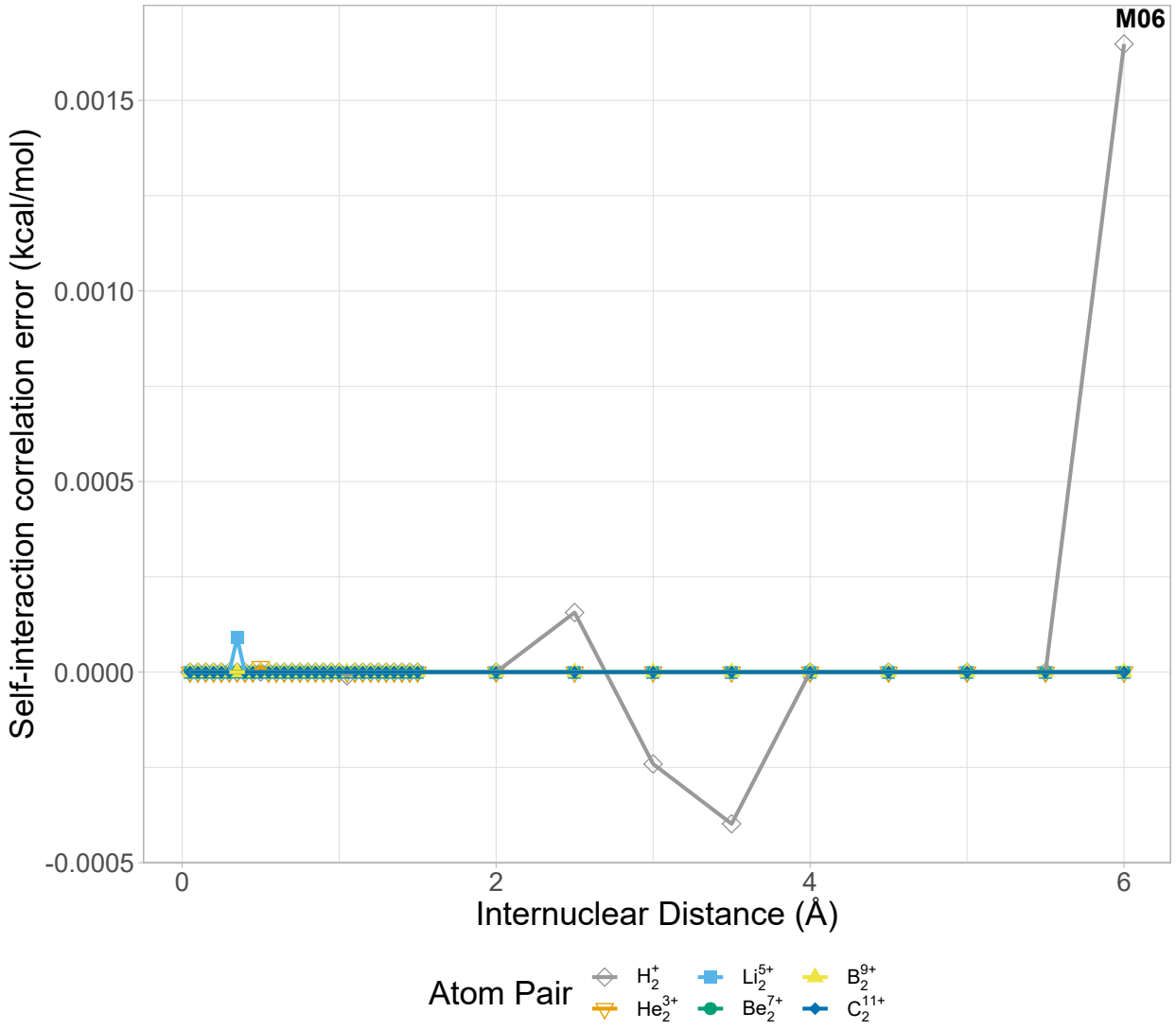

Figure S170: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for M06. Formally, this DFA is self-correlation free, any deviations from this is likely computational noise.

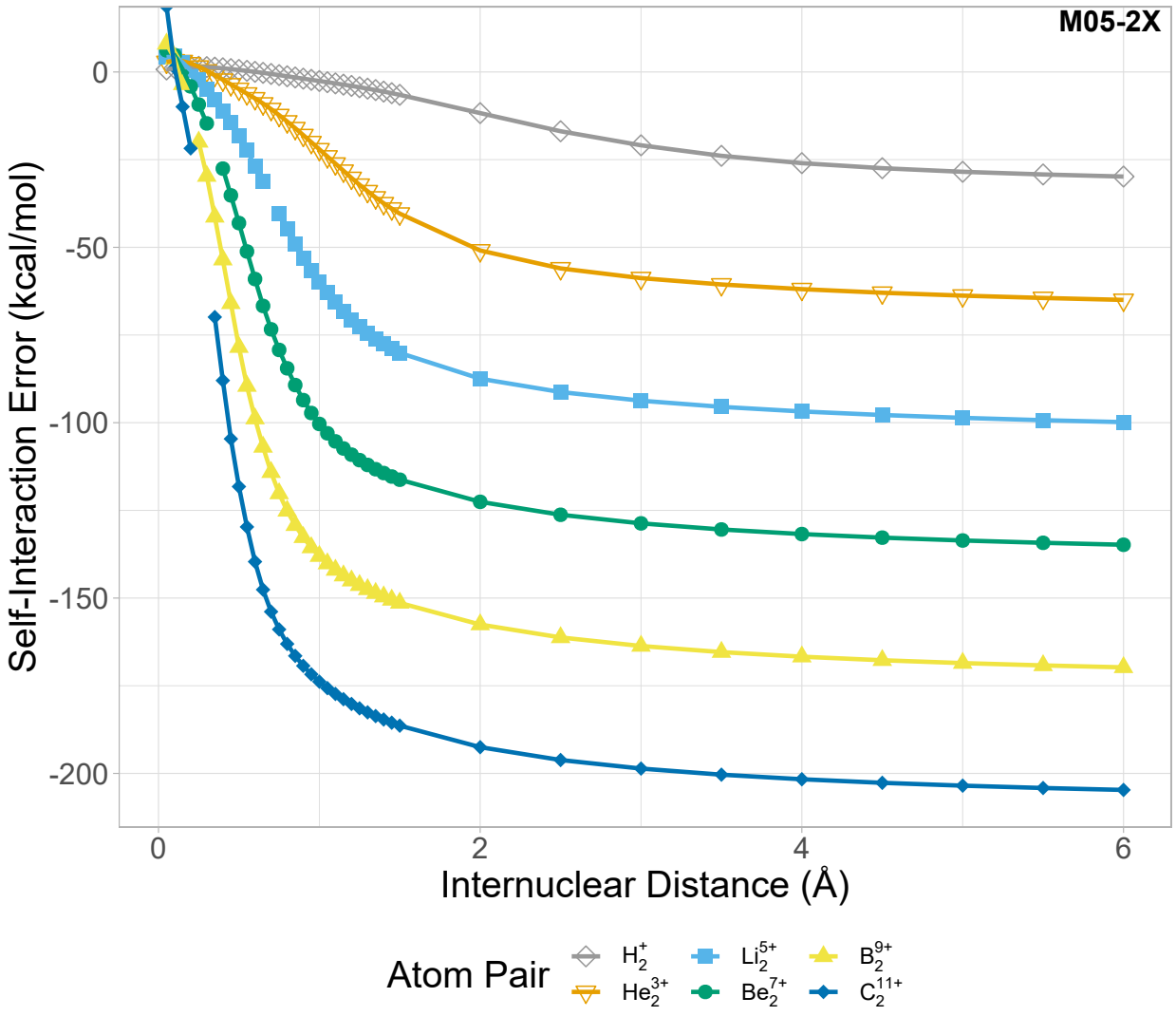

Figure S171: Self-interaction error vs internuclear distance for incremental homonuclear pairs for M05-2X.

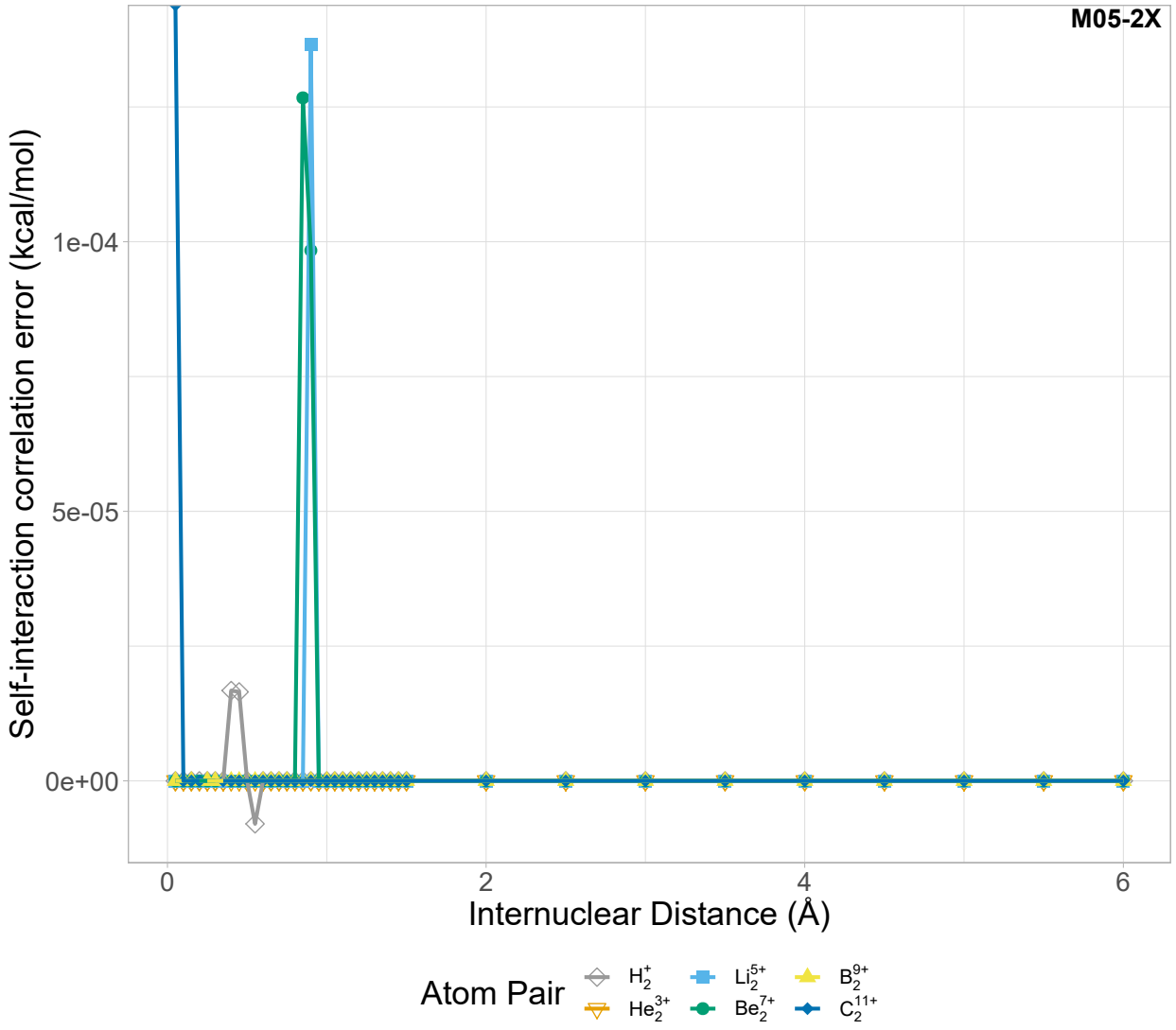

Figure S172: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for M05-2X. Formally, this DFA is self-correlation free, any deviations from this is likely computational noise.

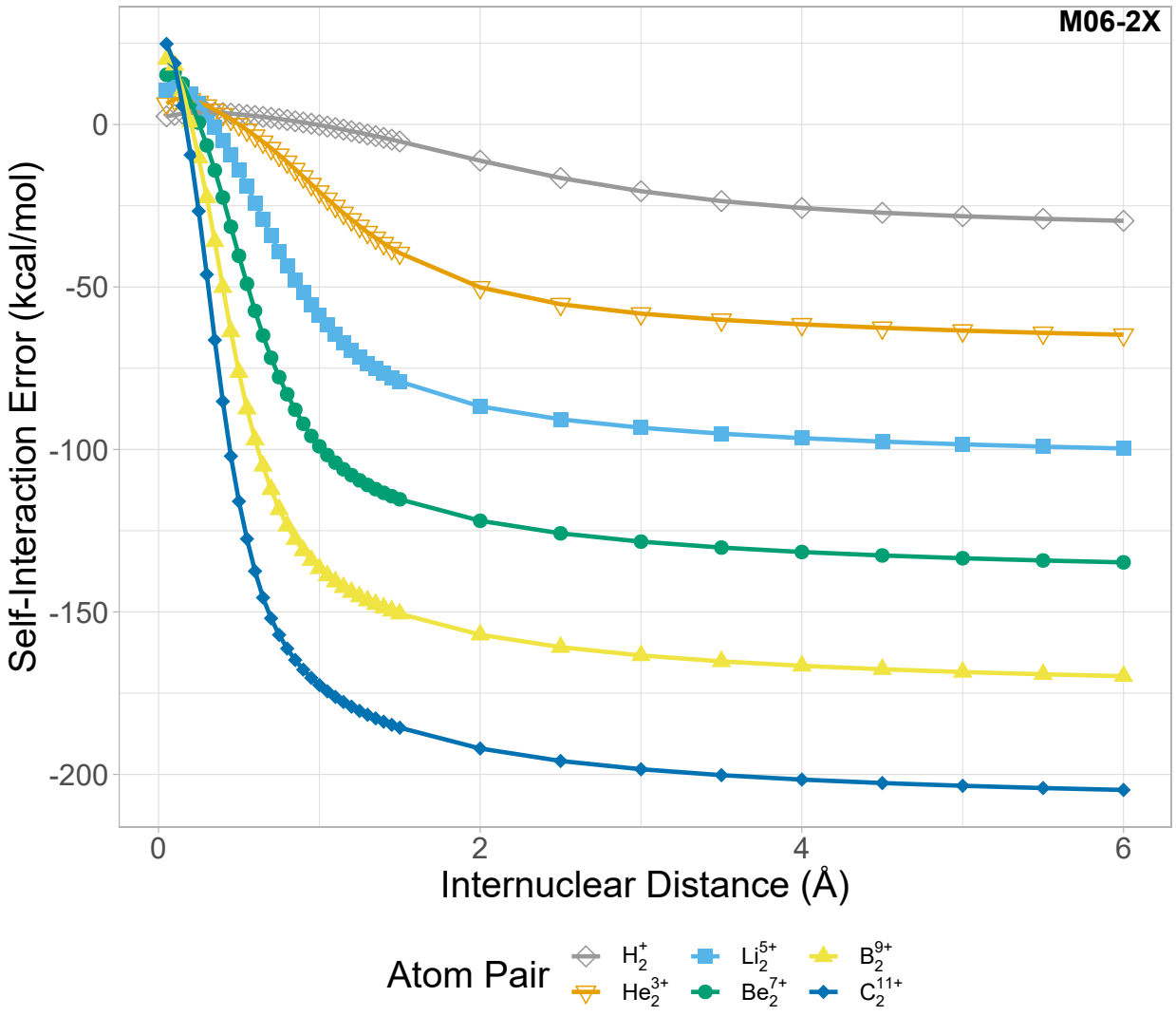

Figure S173: Self-interaction error vs internuclear distance for incremental homonuclear pairs for M06-2X.

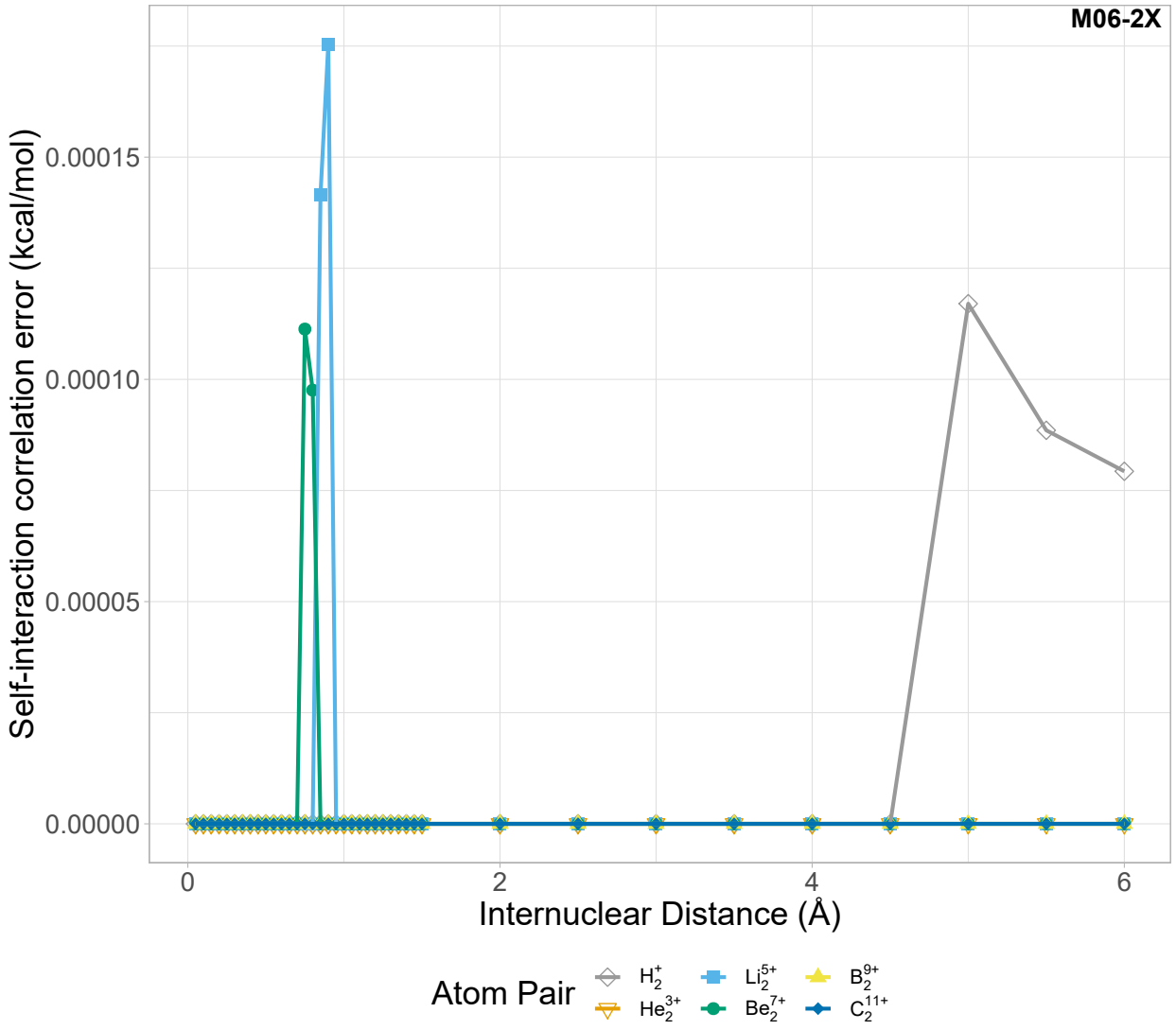

Figure S174: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for M06-2X. Formally, this DFA is self-correlation free, any deviations from this is likely computational noise.

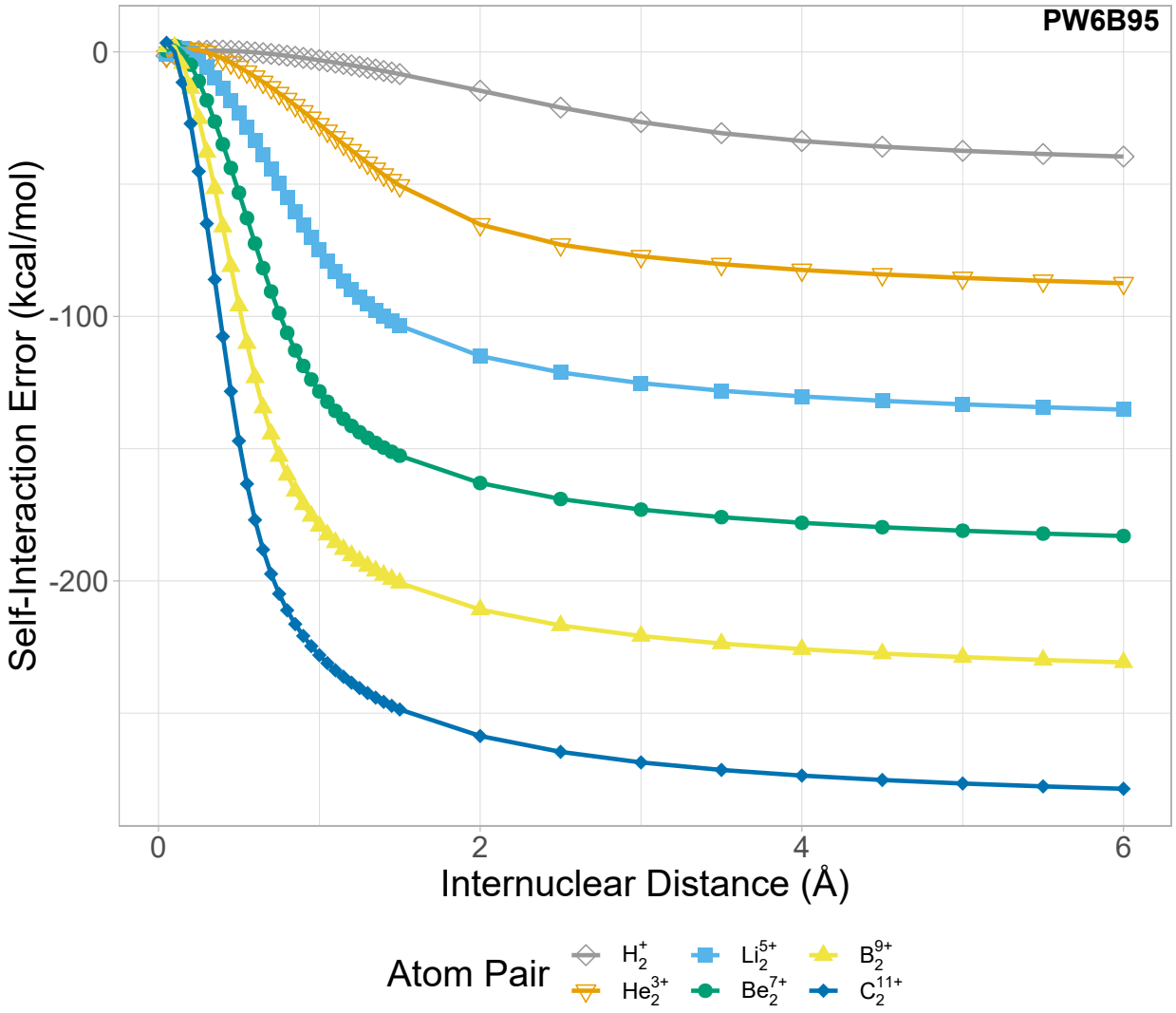

Figure S175: Self-interaction error vs internuclear distance for incremental homonuclear pairs for PW6B95.

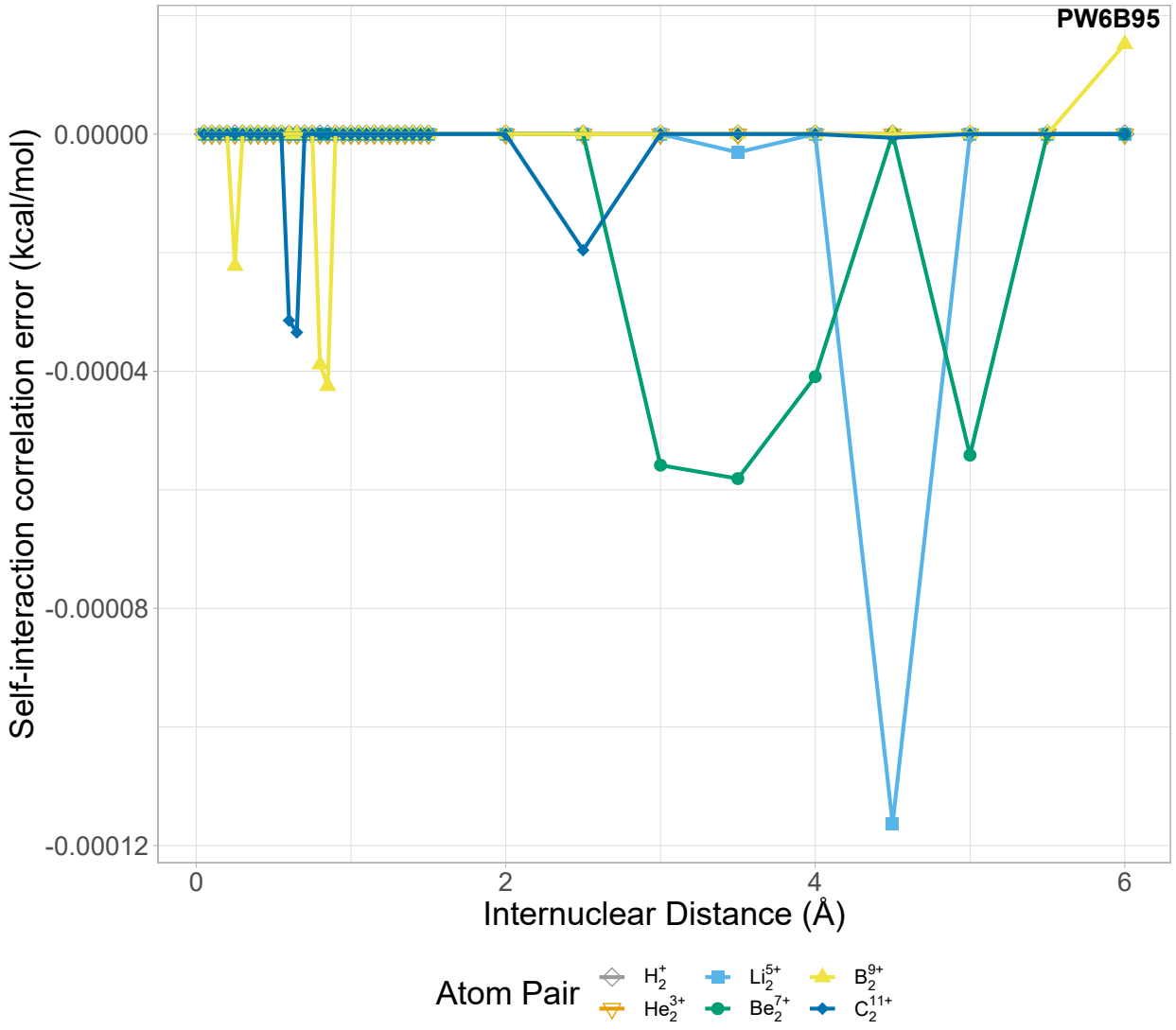

Figure S176: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for PW6B95. Formally, this DFA is self-correlation free, any deviations from this is likely computational noise.

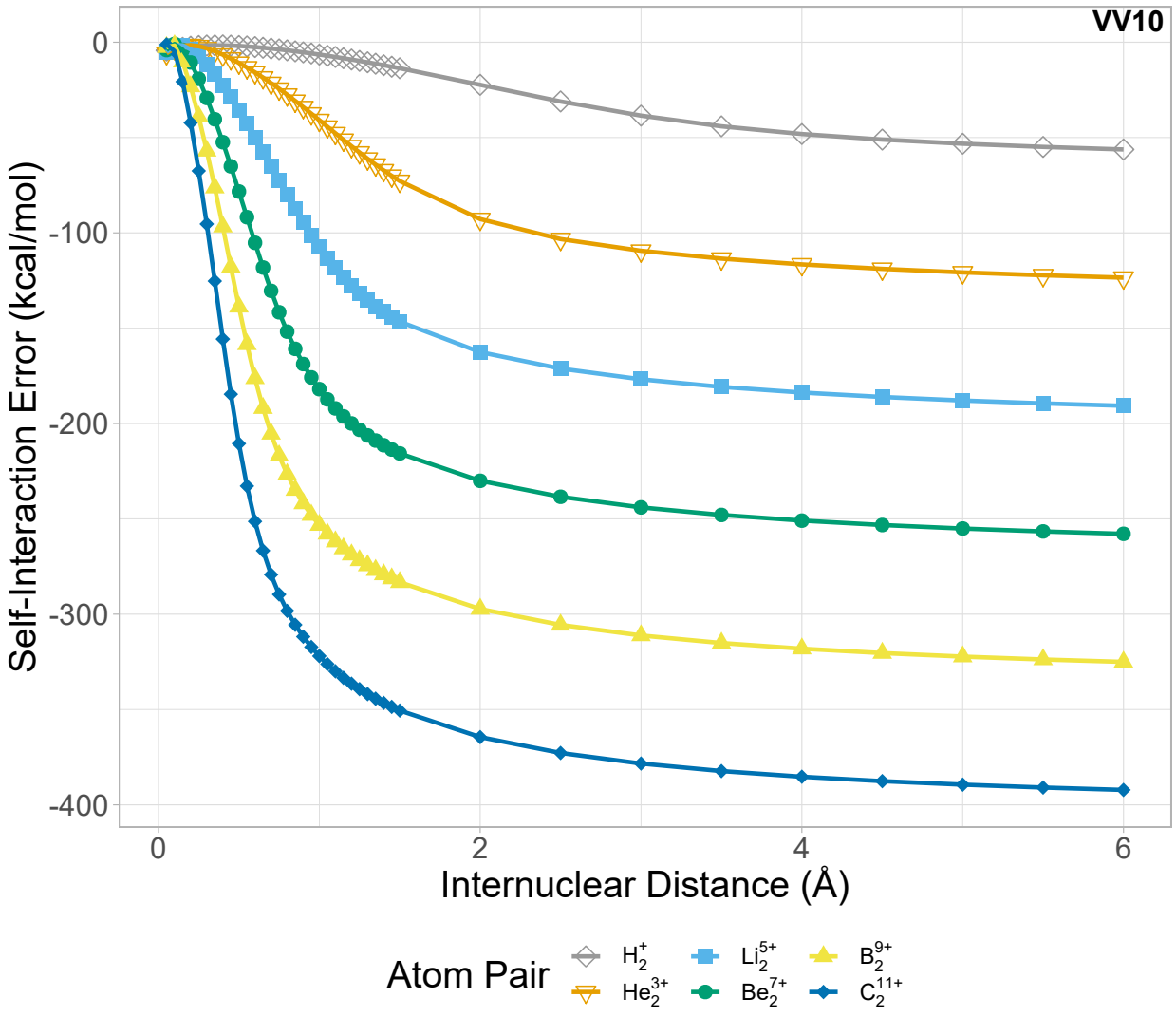

Figure S177: Self-interaction error vs internuclear distance for incremental homonuclear pairs for VV10.

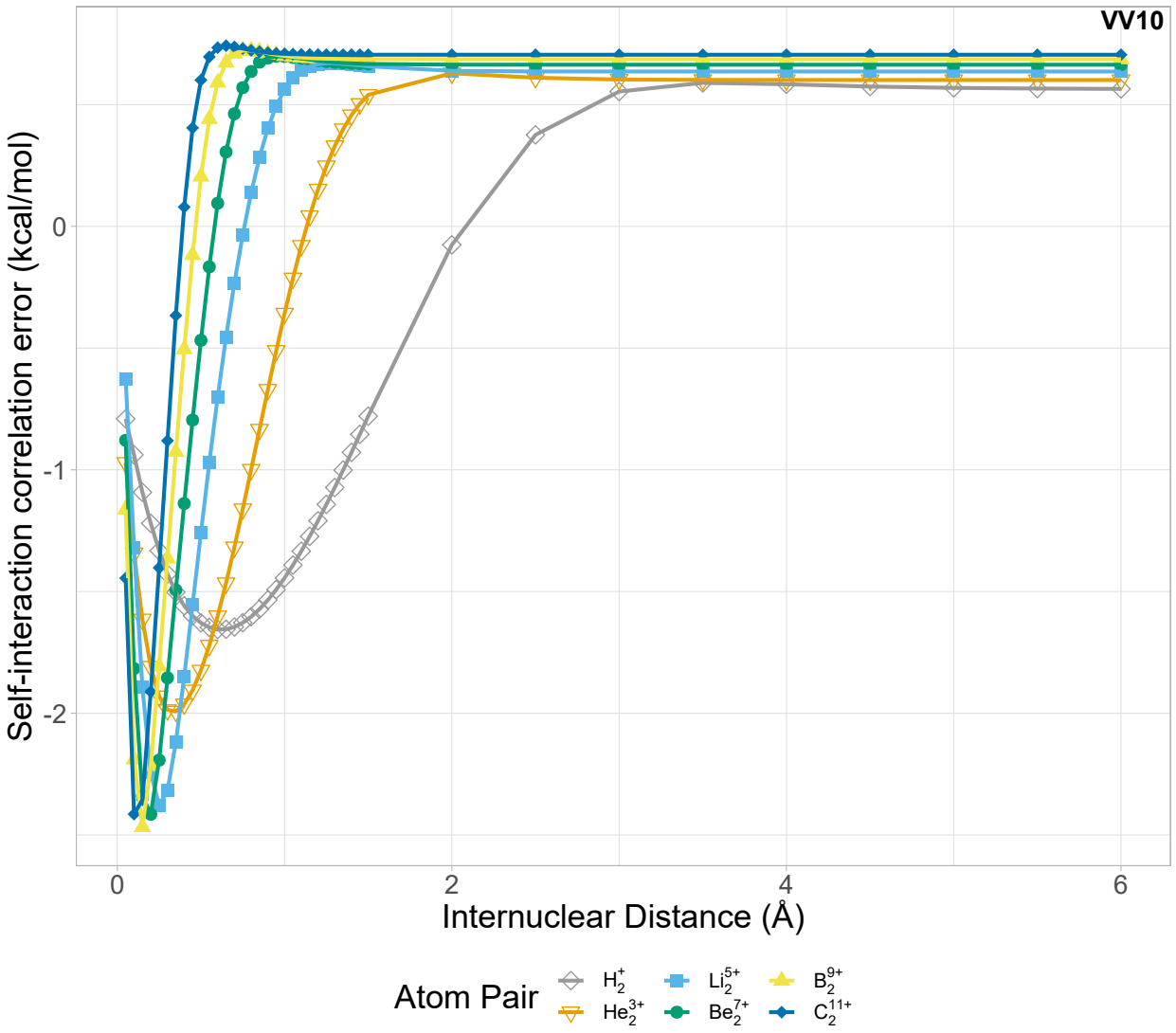

Figure S178: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for VV10.

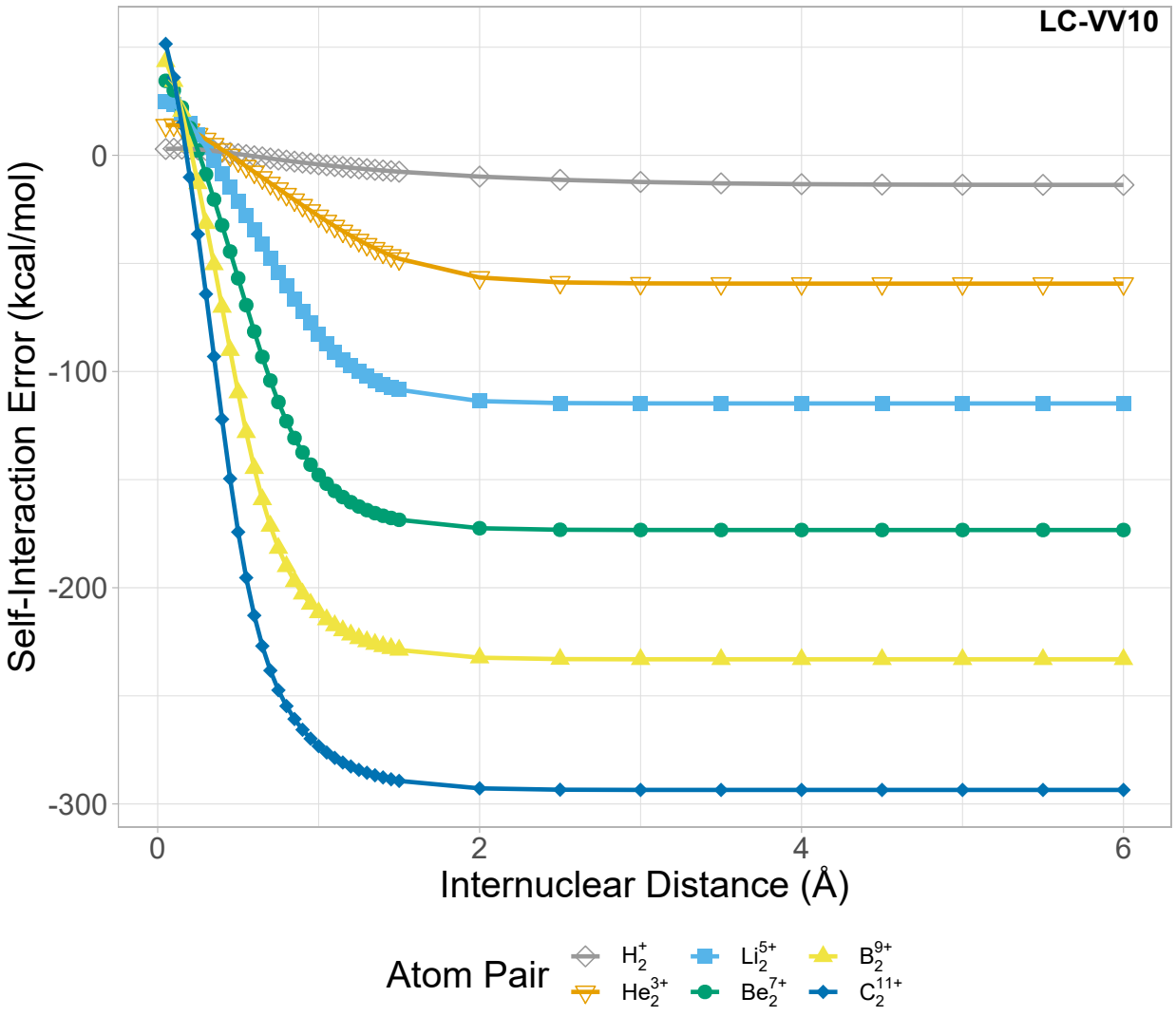

Figure S179: Self-interaction error vs internuclear distance for incremental homonuclear pairs for LC-VV10.

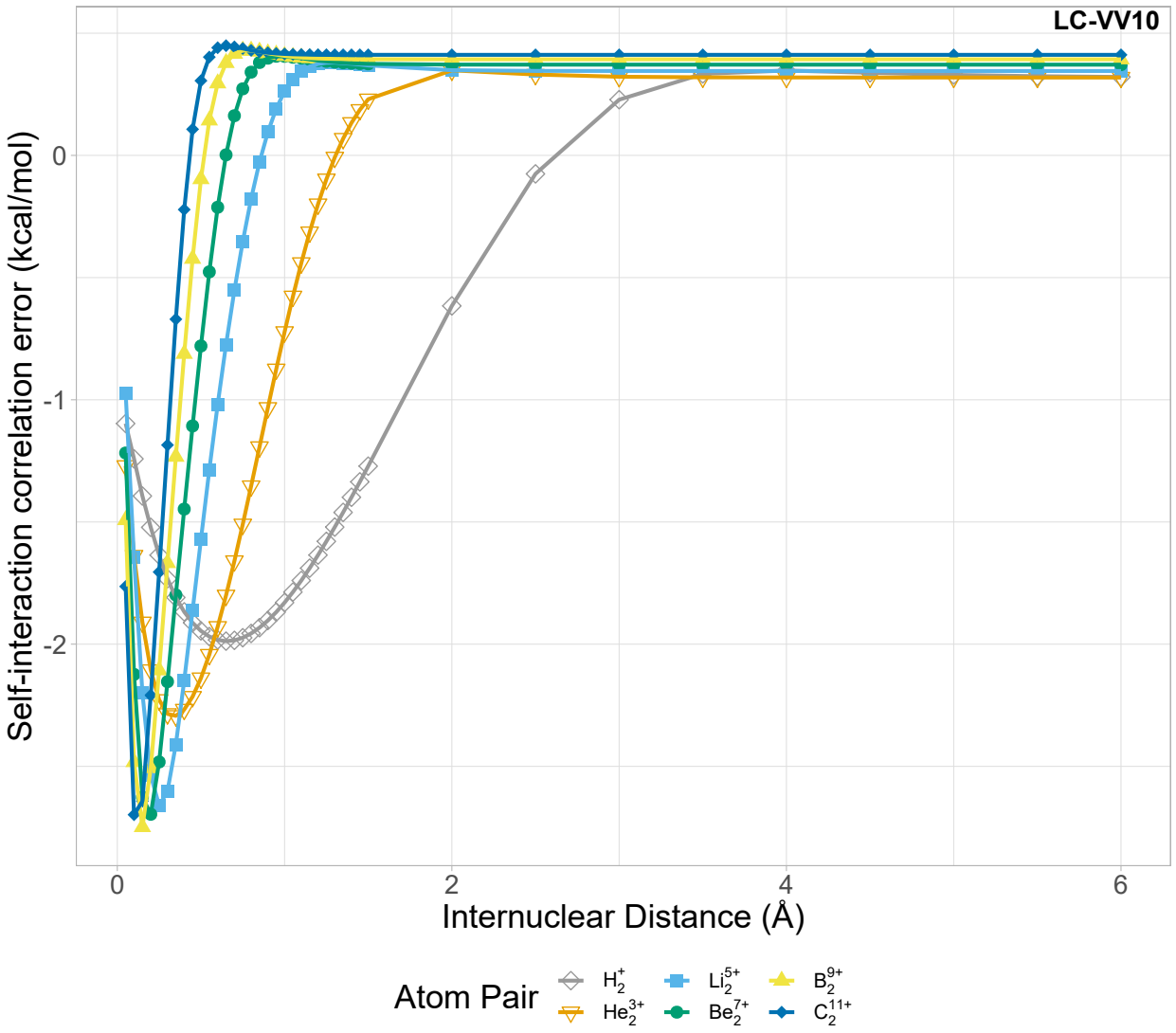

Figure S180: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for LC-VV10.

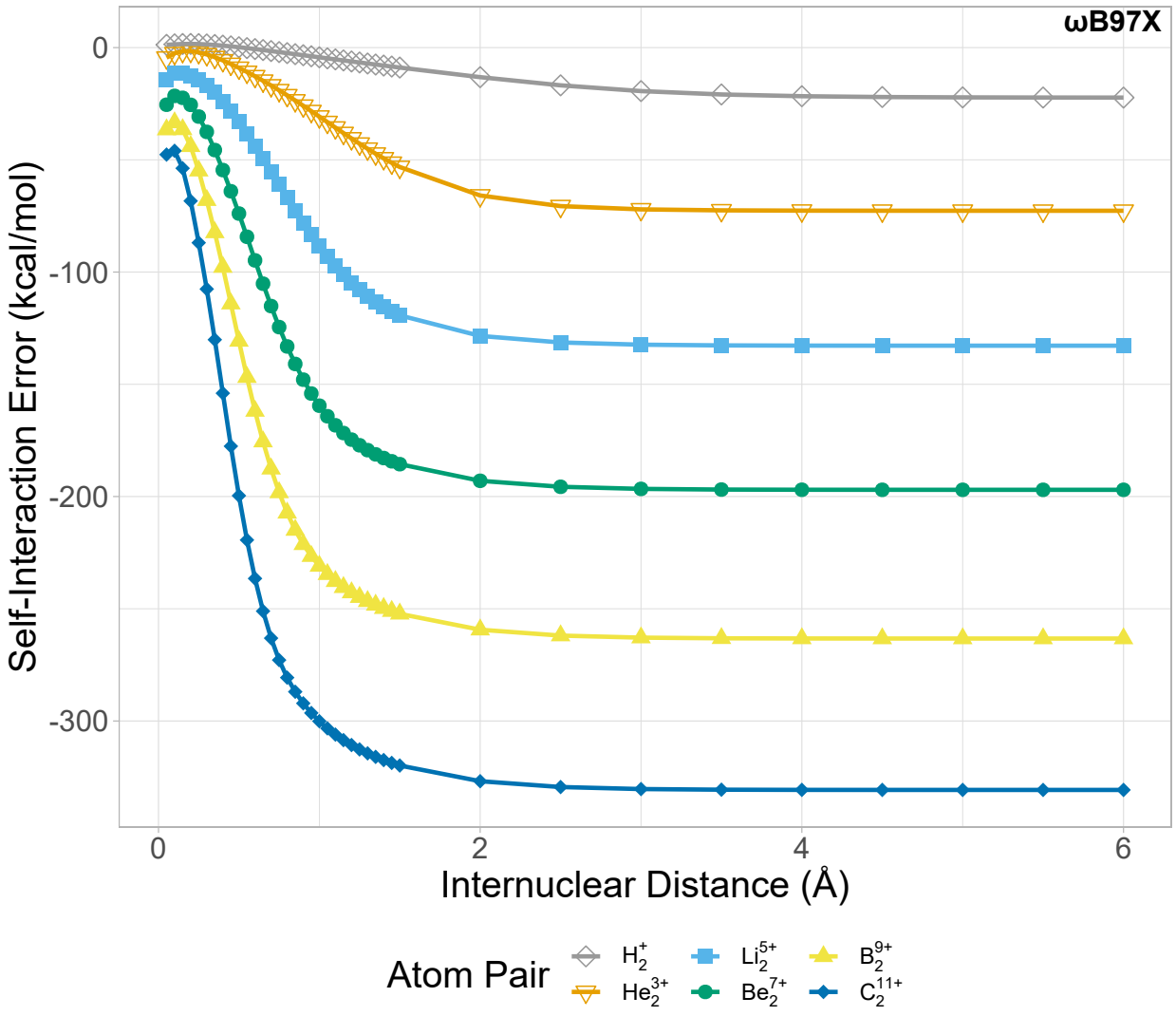

Figure S181: Self-interaction error vs internuclear distance for incremental homonuclear pairs for ωB97X.

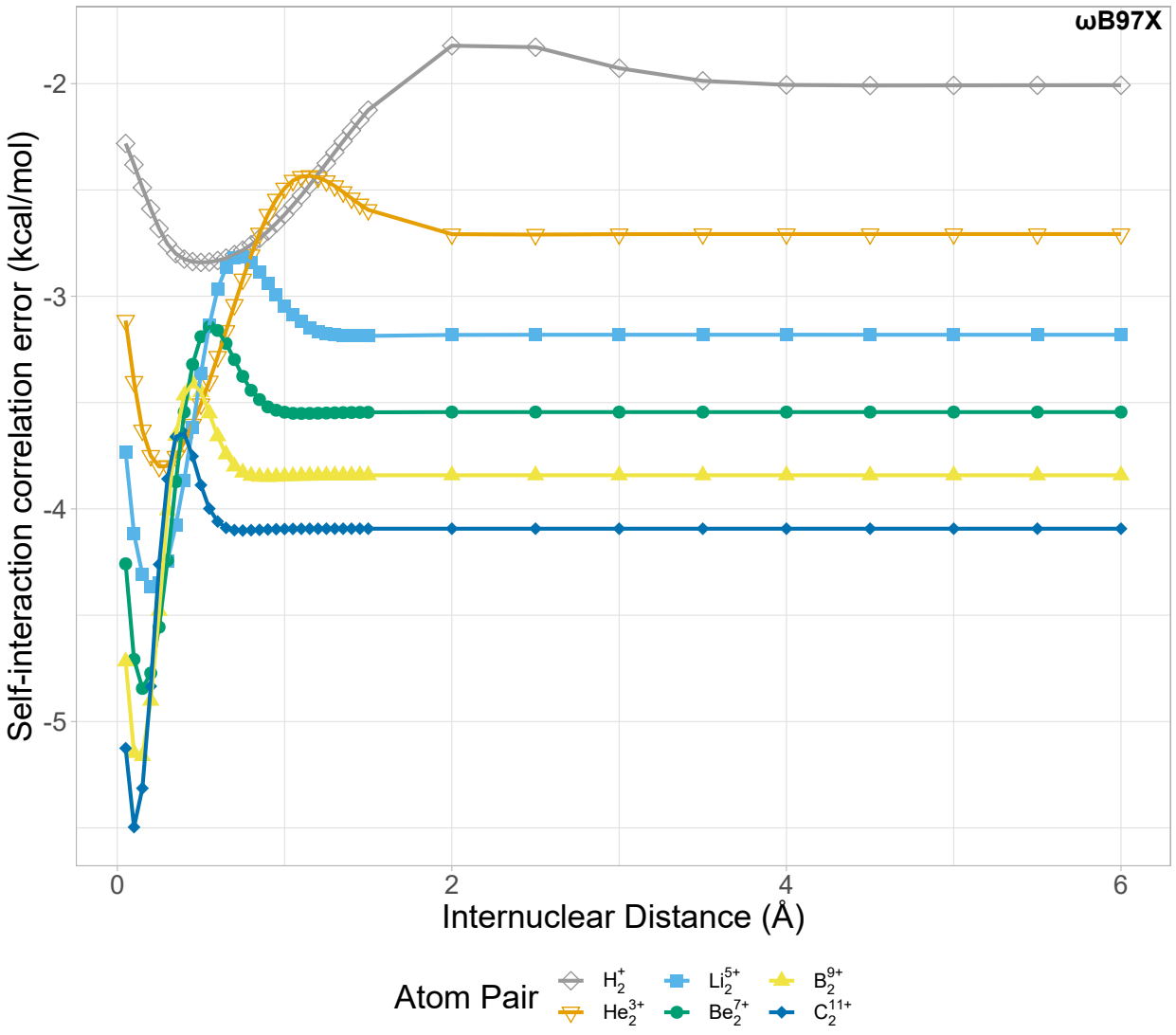

Figure S182: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for ωB97X.

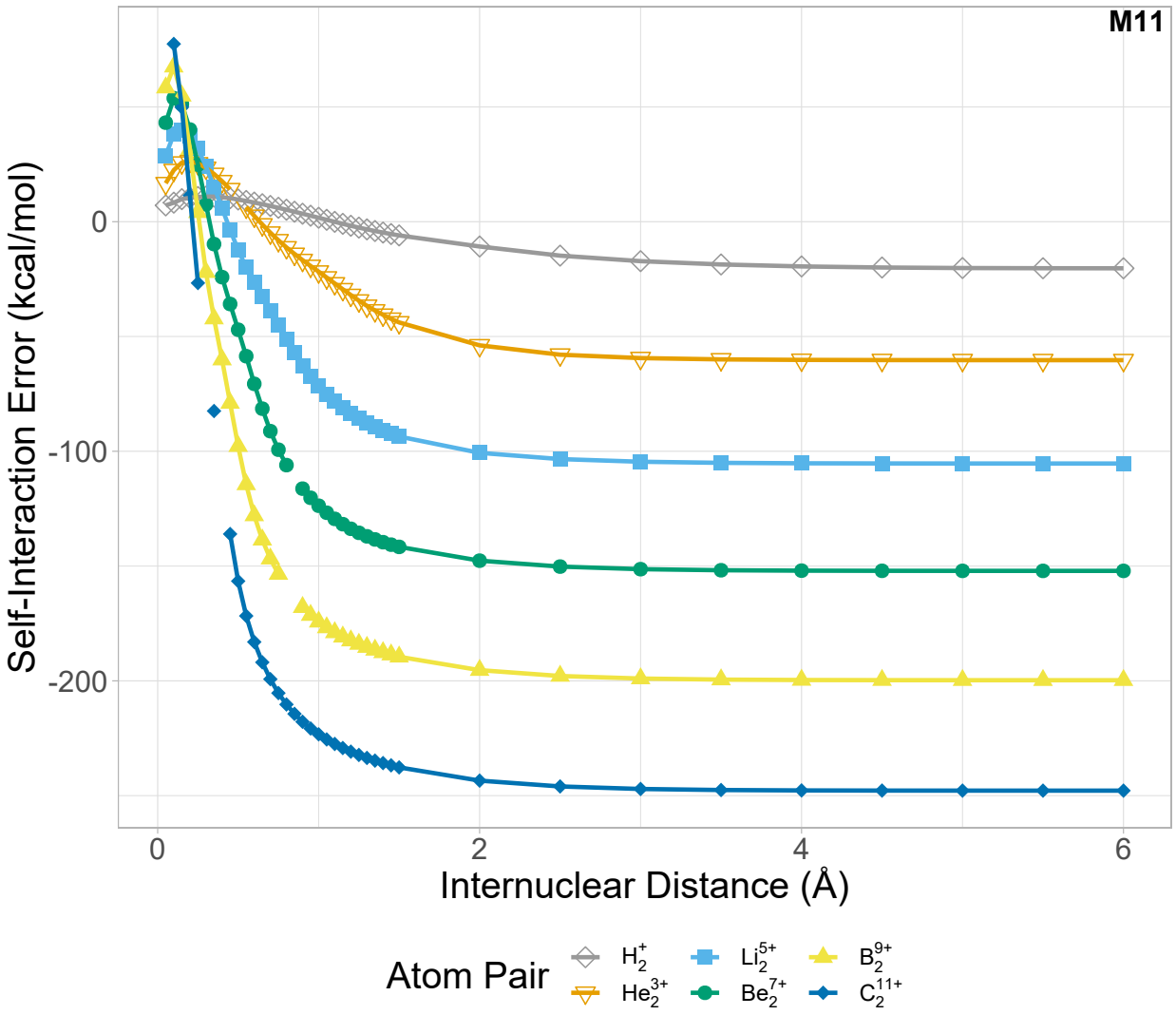

Figure S183: Self-interaction error vs internuclear distance for incremental homonuclear pairs for M11.

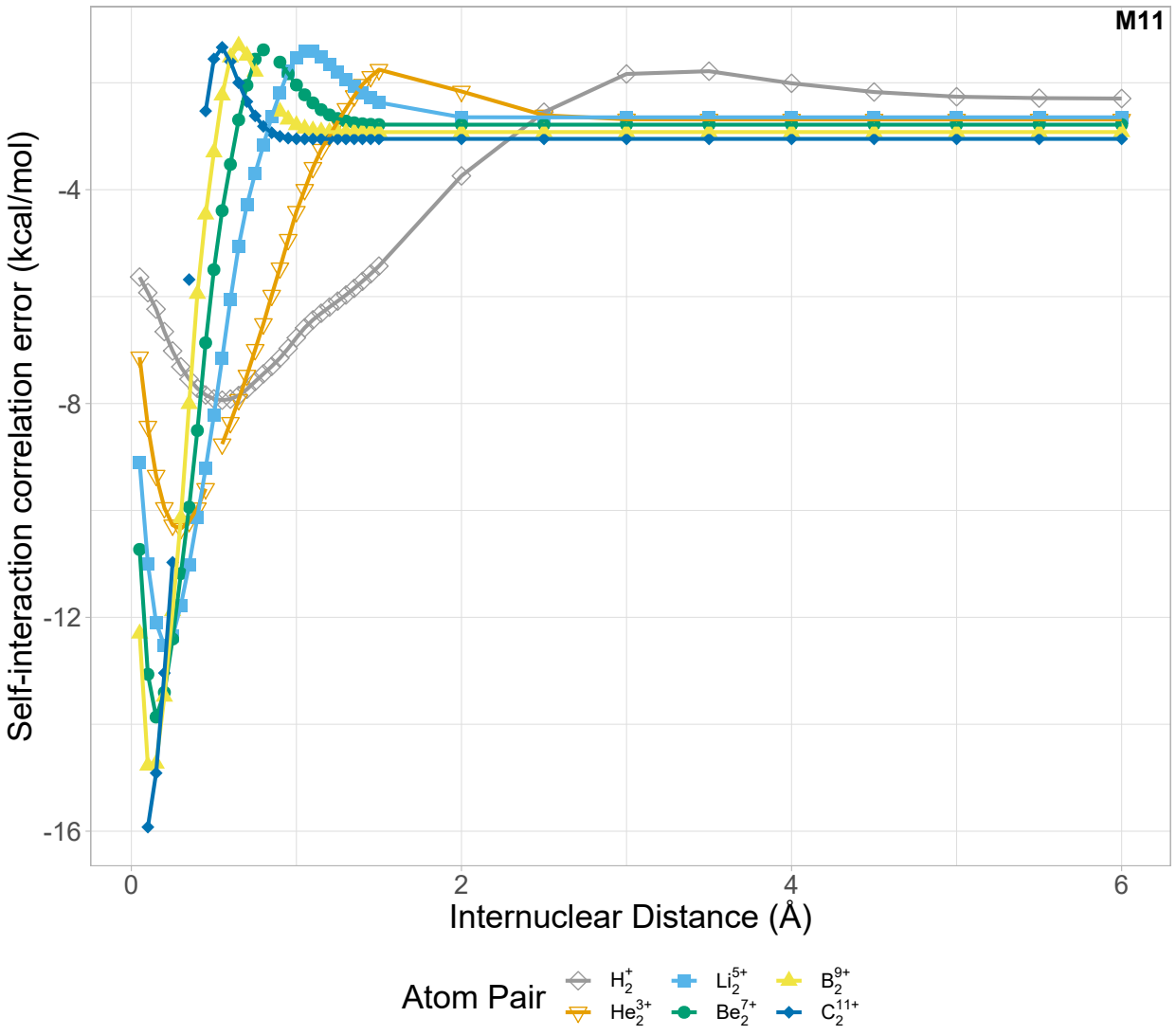

Figure S184: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for M11.

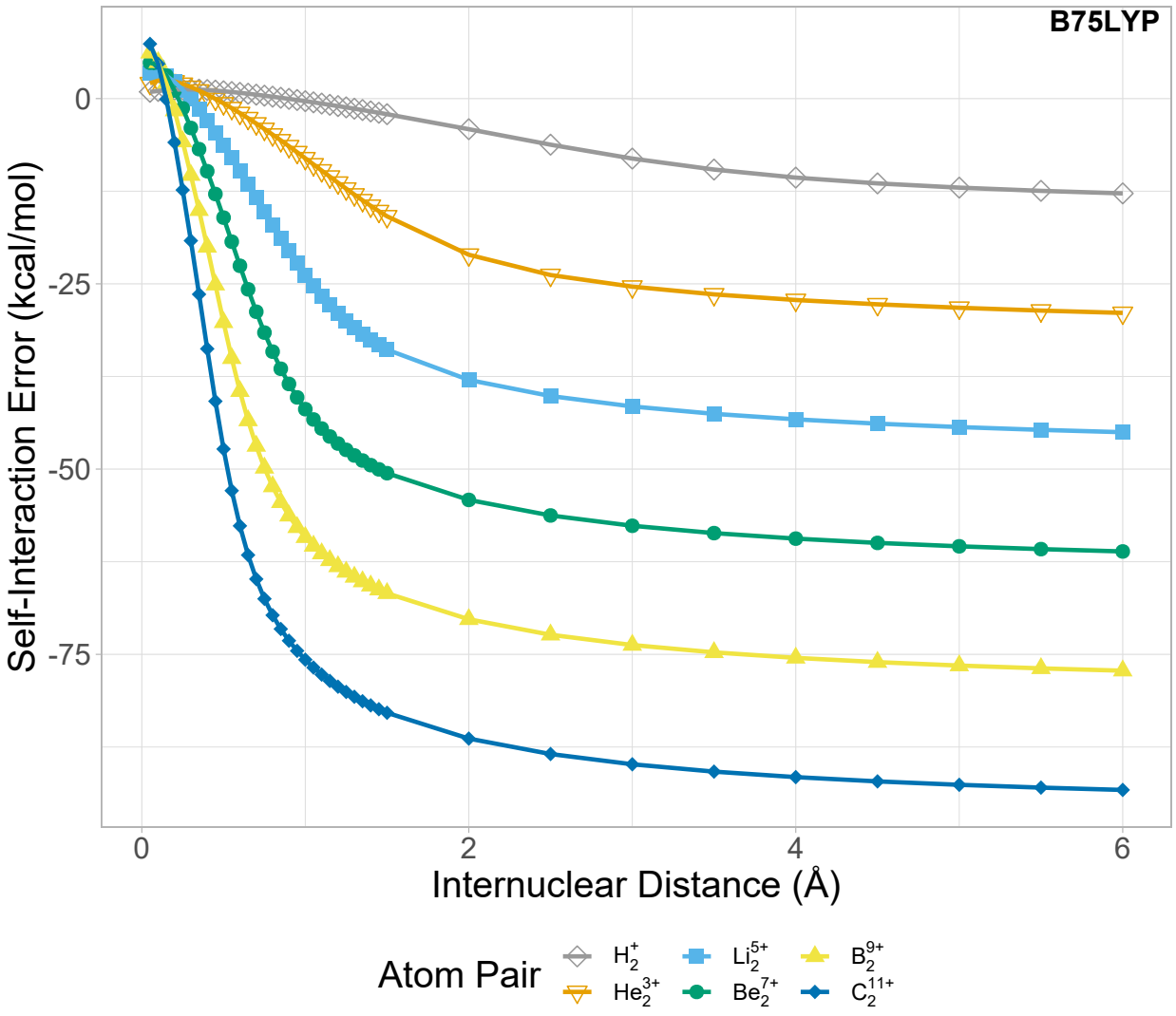

Figure S185: Self-interaction error vs internuclear distance for incremental homonuclear pairs for B75LYP.

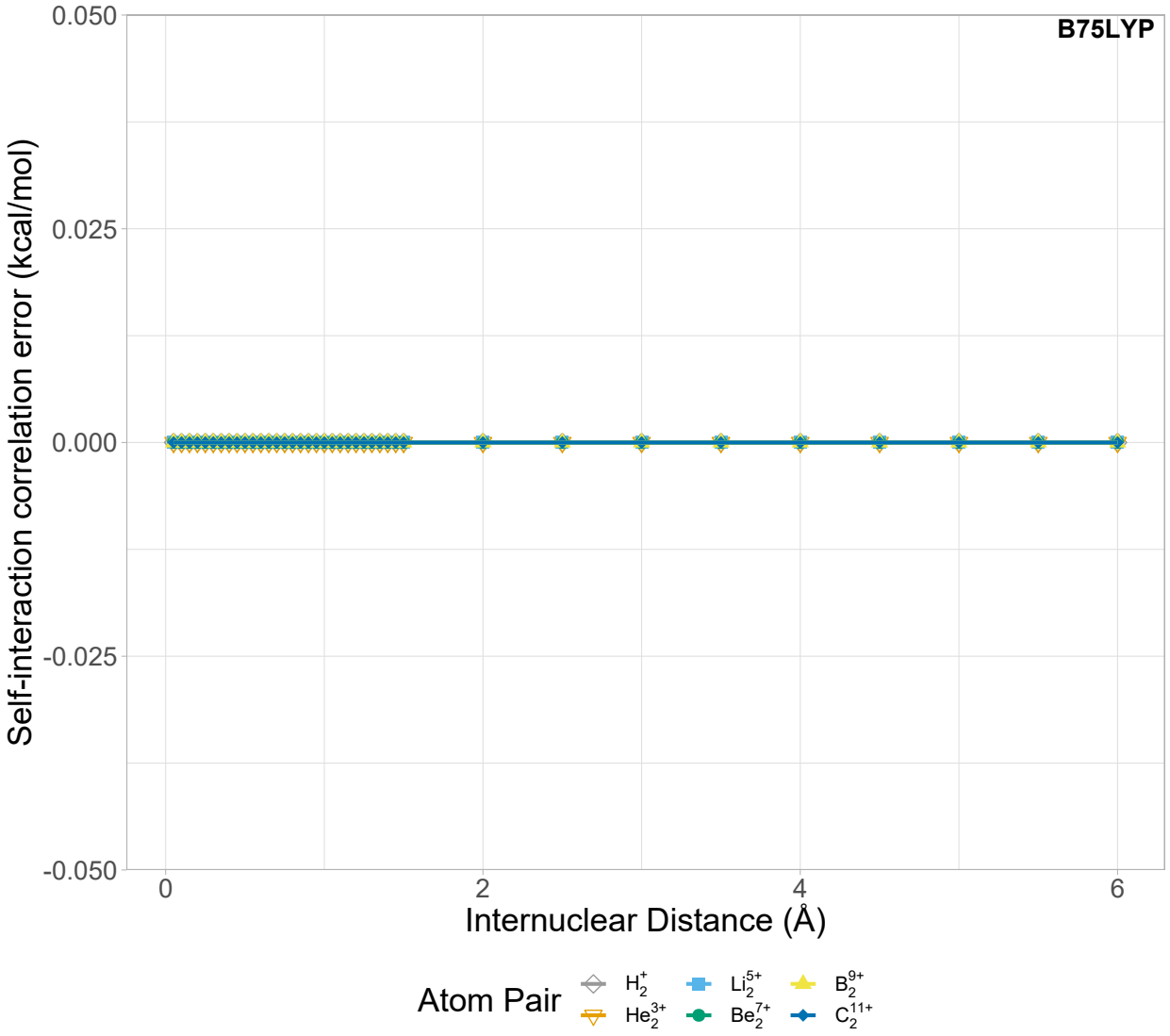

Figure S186: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for B75LYP.

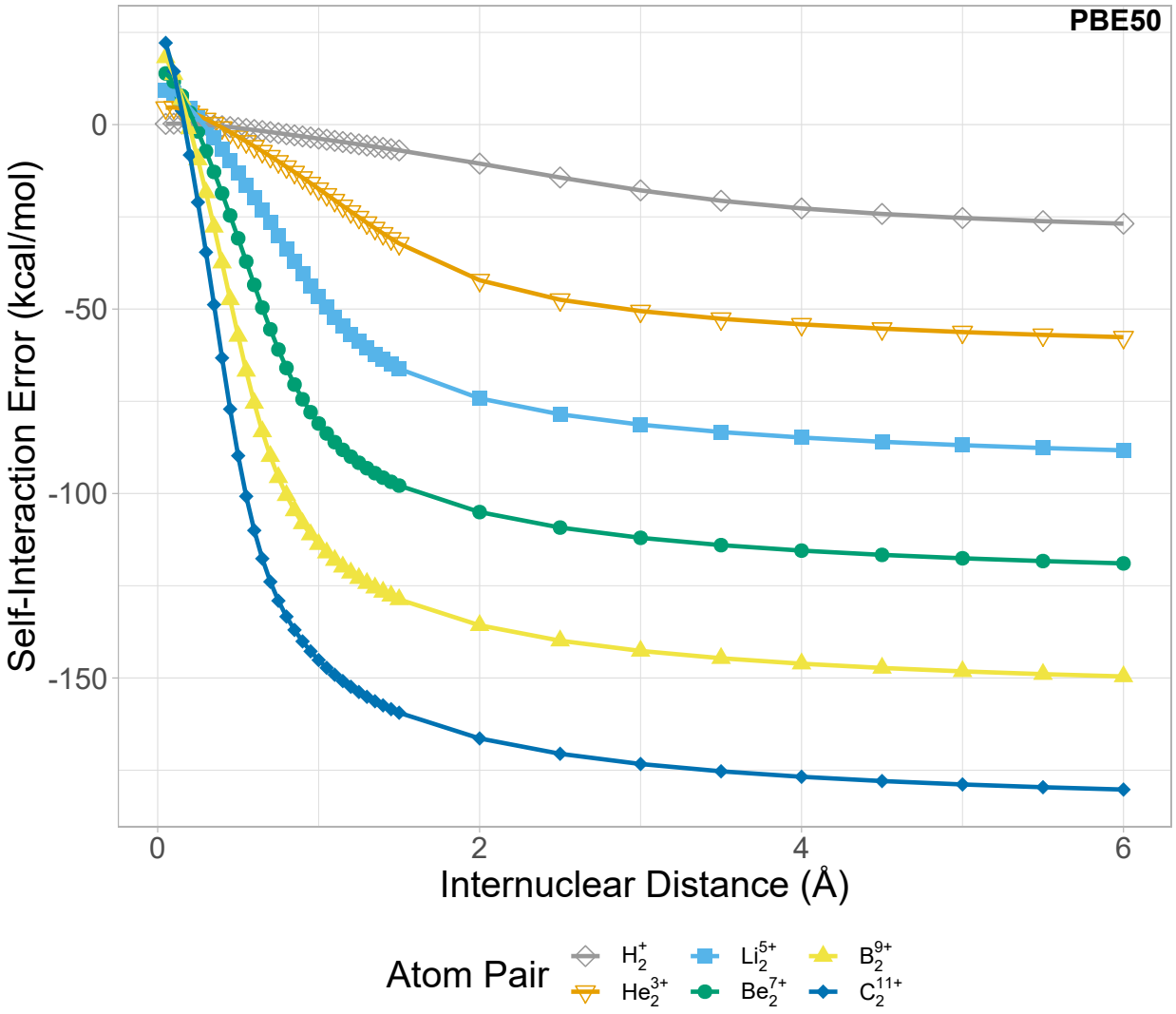

Figure S187: Self-interaction error vs internuclear distance for incremental homonuclear pairs for PBE50.

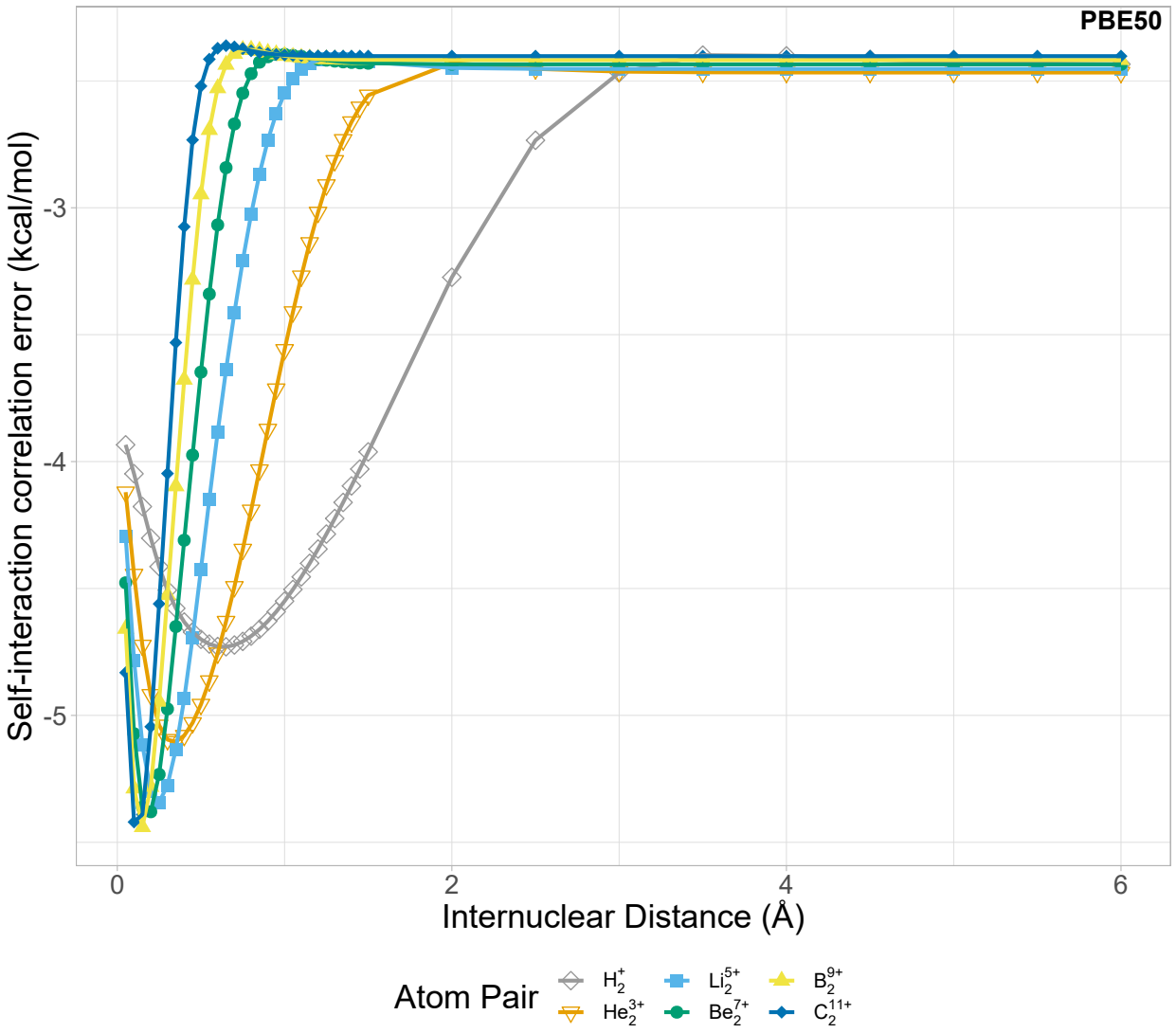

Figure S188: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for PBE50.

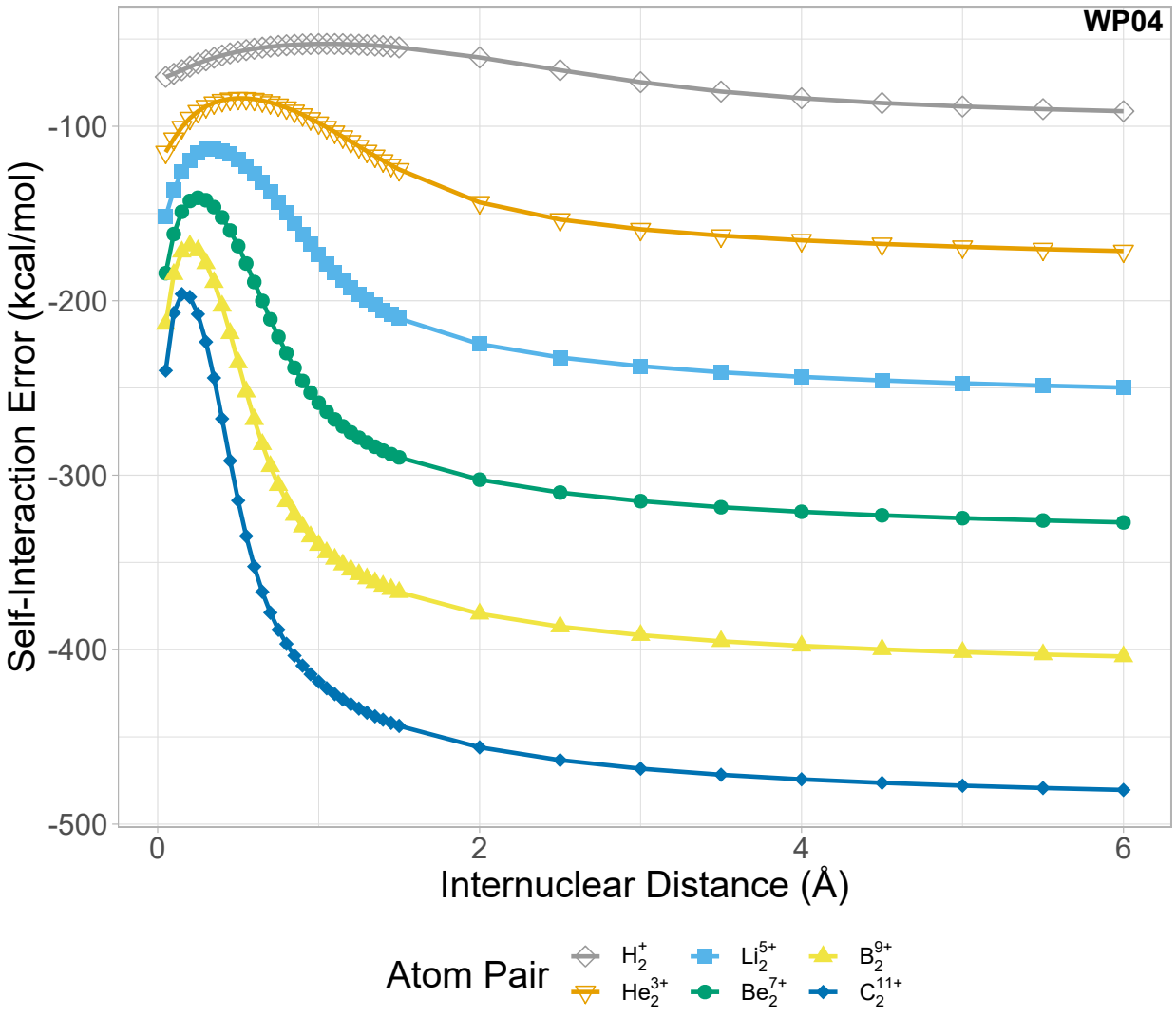

Figure S189: Self-interaction error vs internuclear distance for incremental homonuclear pairs for WP04.

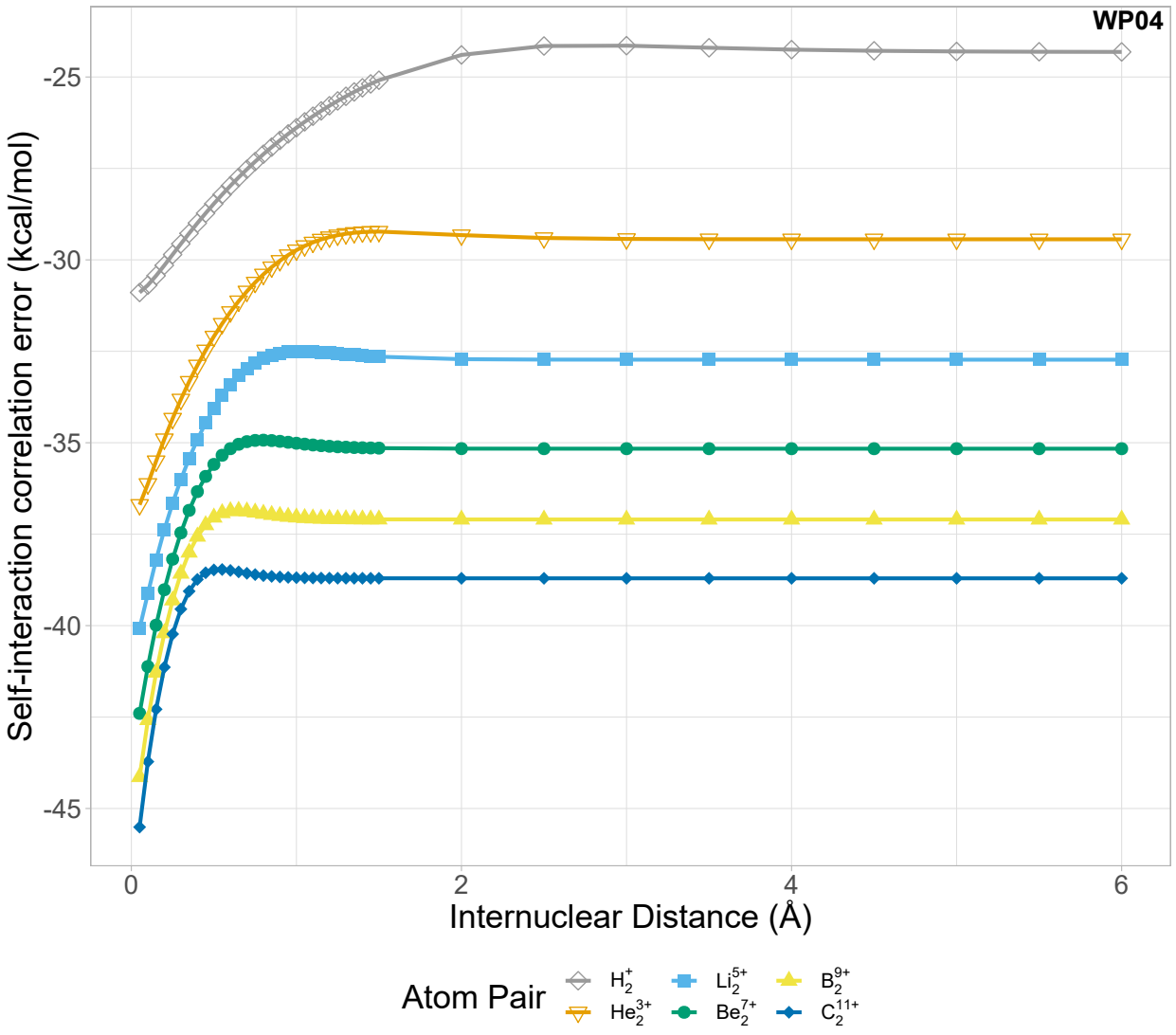

Figure S190: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for WP04.

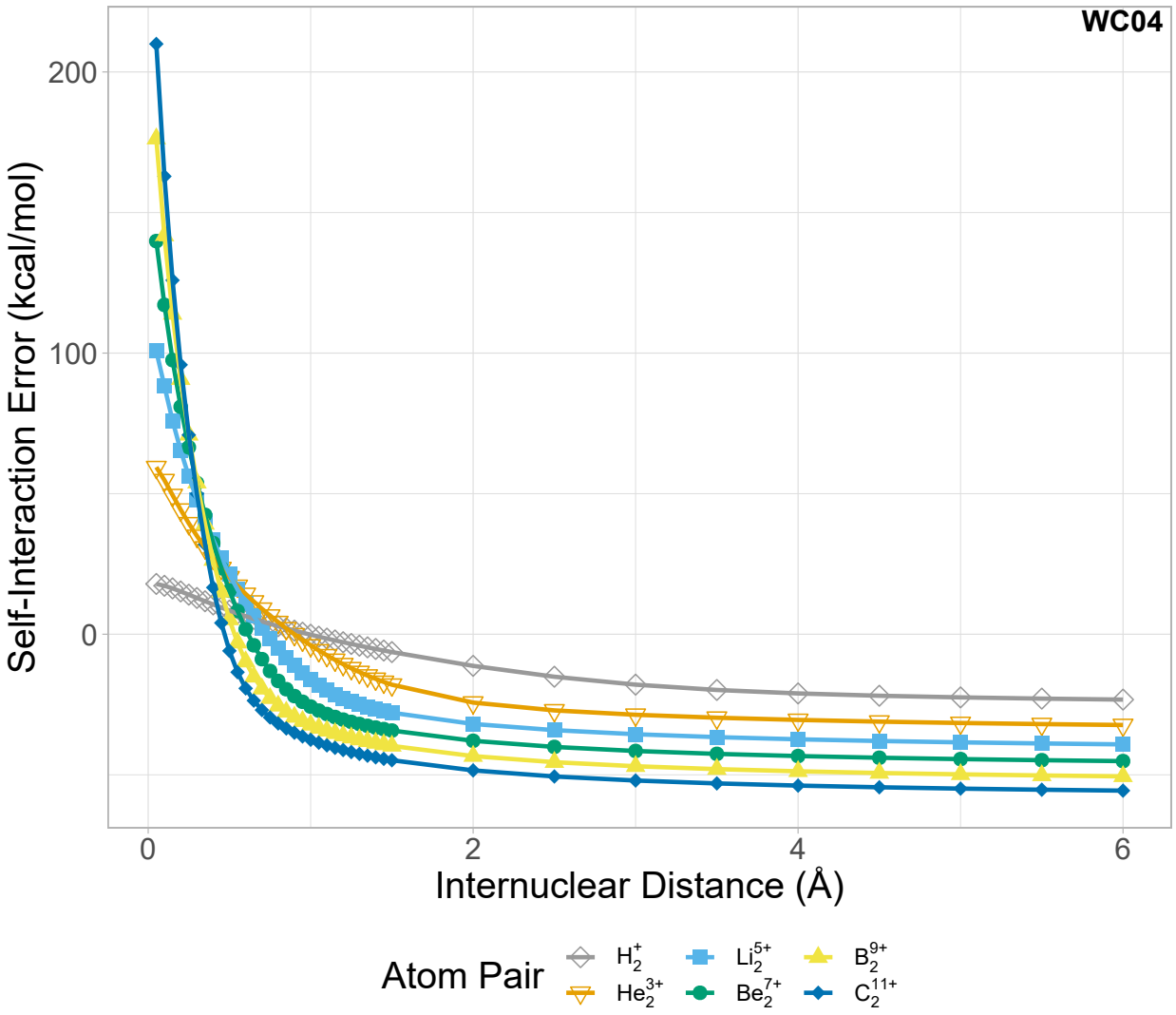

Figure S191: Self-interaction error vs internuclear distance for incremental homonuclear pairs for WC04.

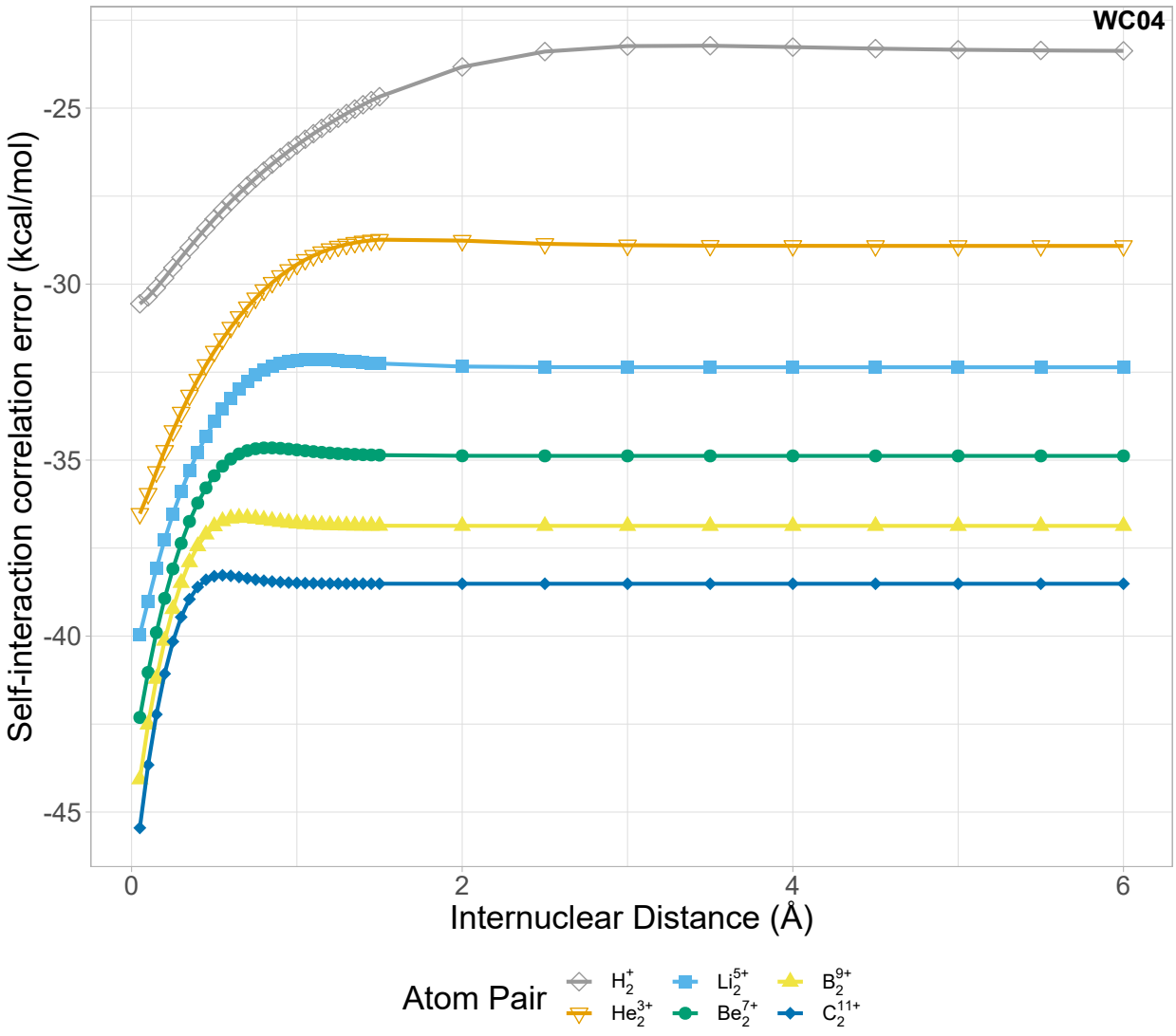

Figure S192: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for WC04.

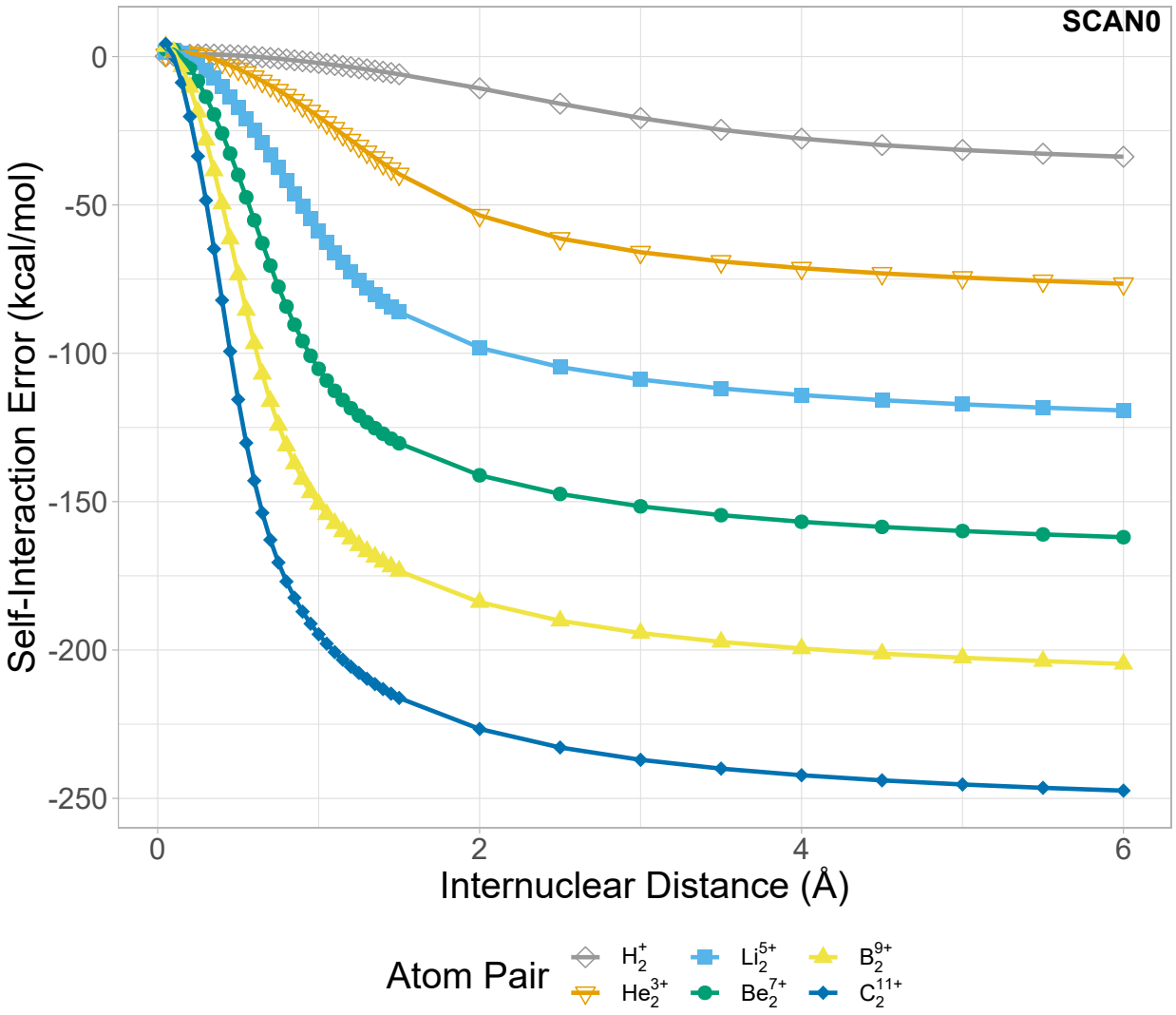

Figure S193: Self-interaction error vs internuclear distance for incremental homonuclear pairs for SCAN0.

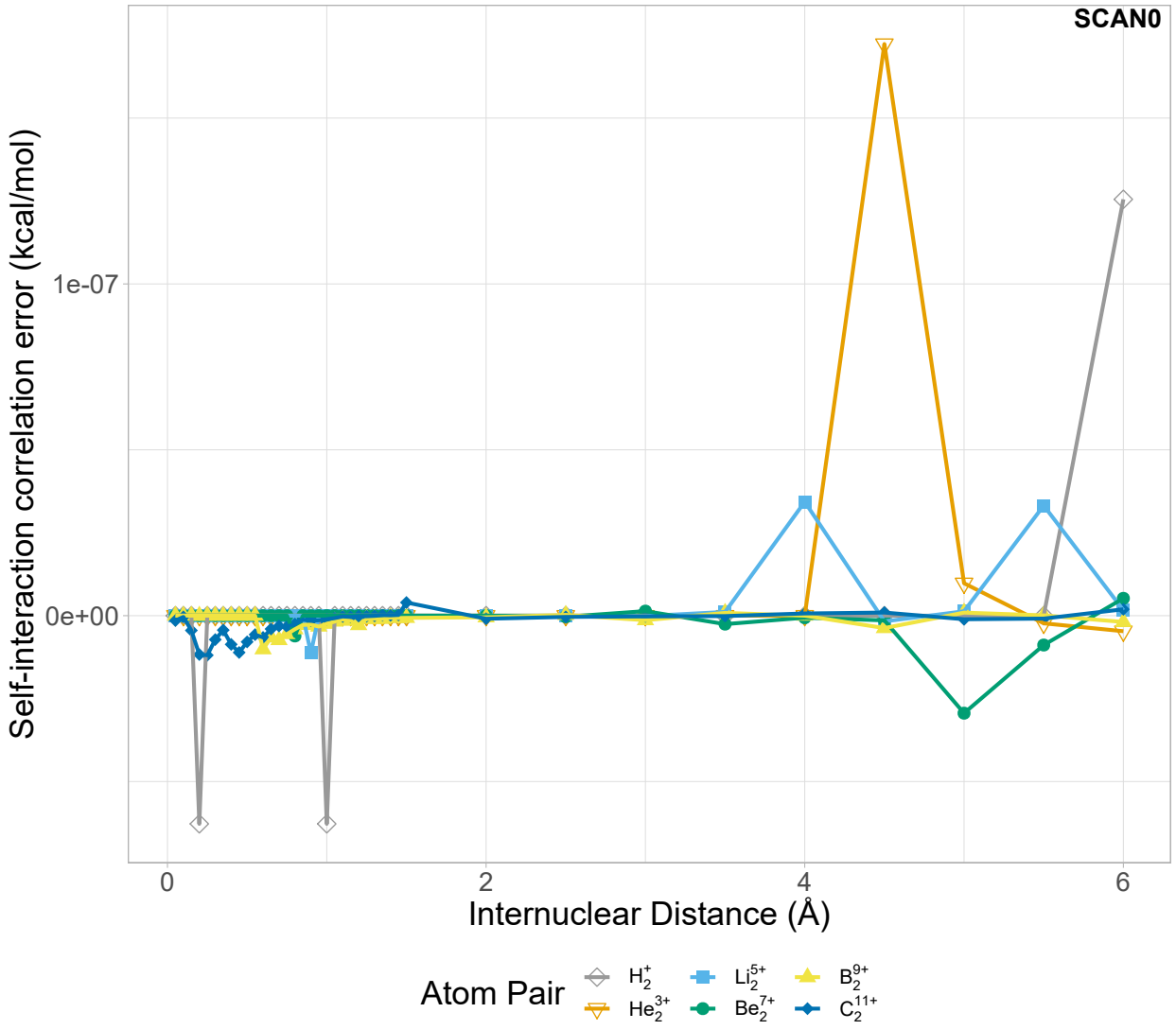

Figure S194: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for SCAN0. Formally, this DFA is self-correlation free, any deviations from this is likely computational noise.

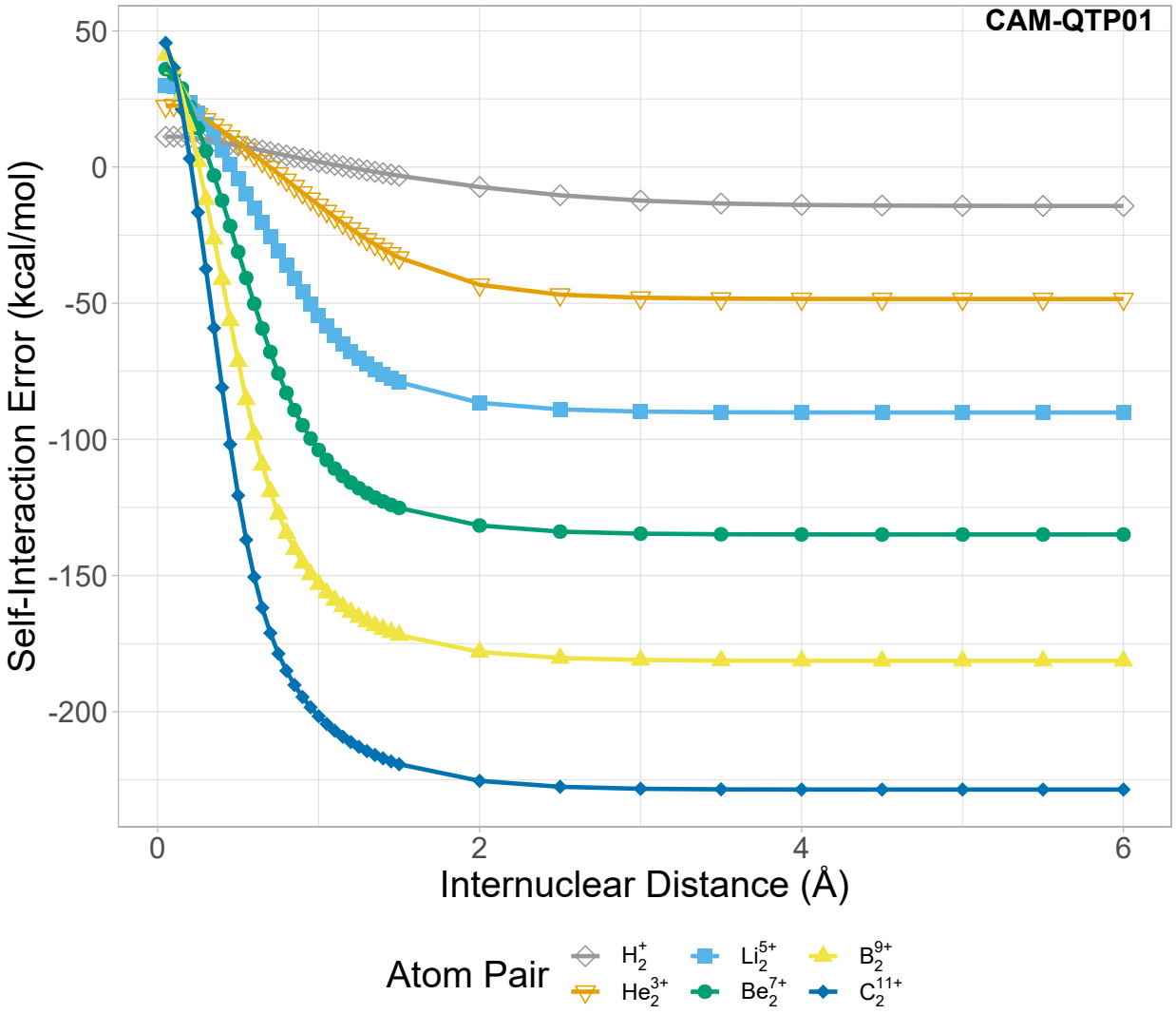

Figure S195: Self-interaction error vs internuclear distance for incremental homonuclear pairs for CAM-QTP01.

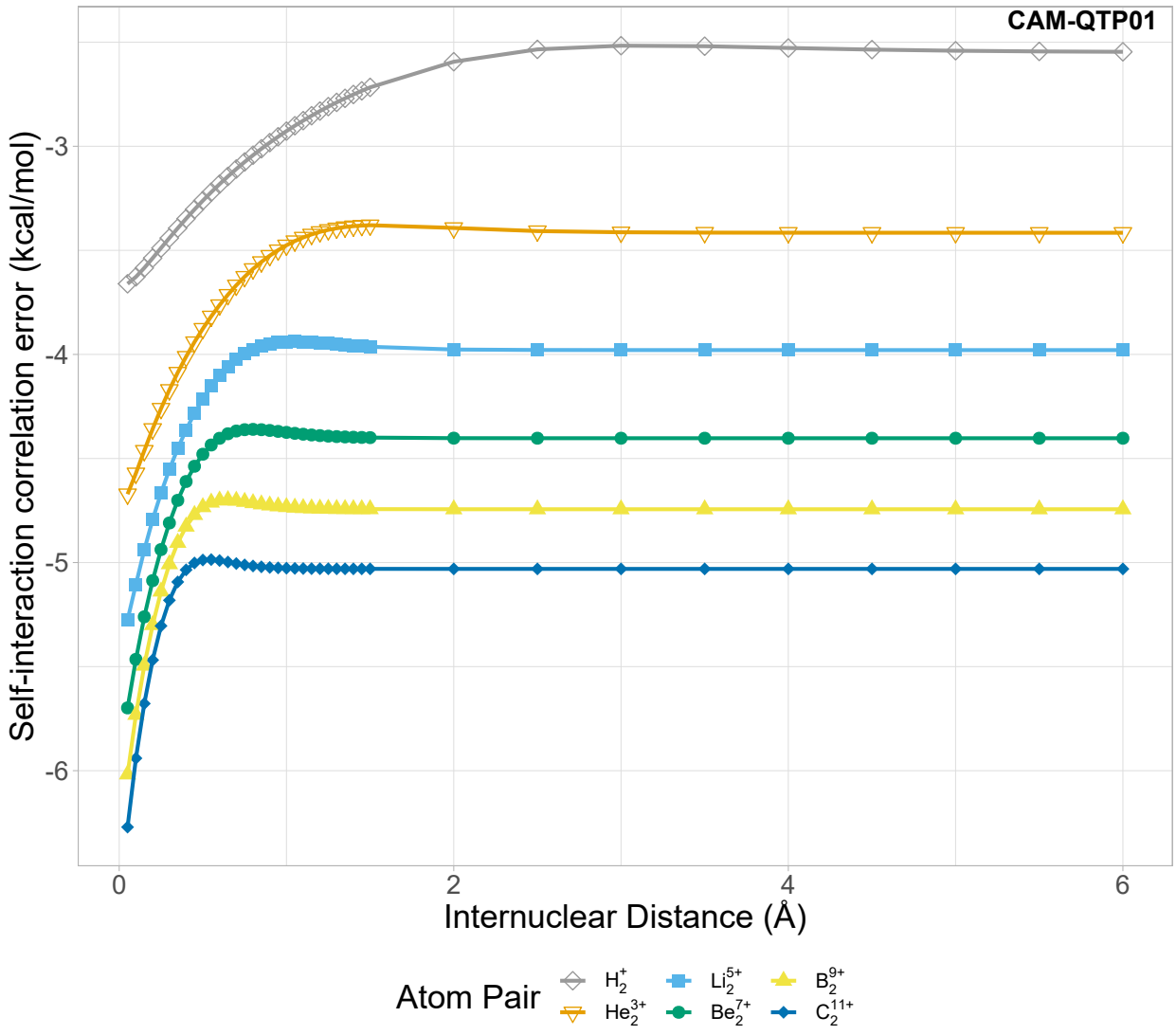

Figure S196: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for CAM-QTP01.

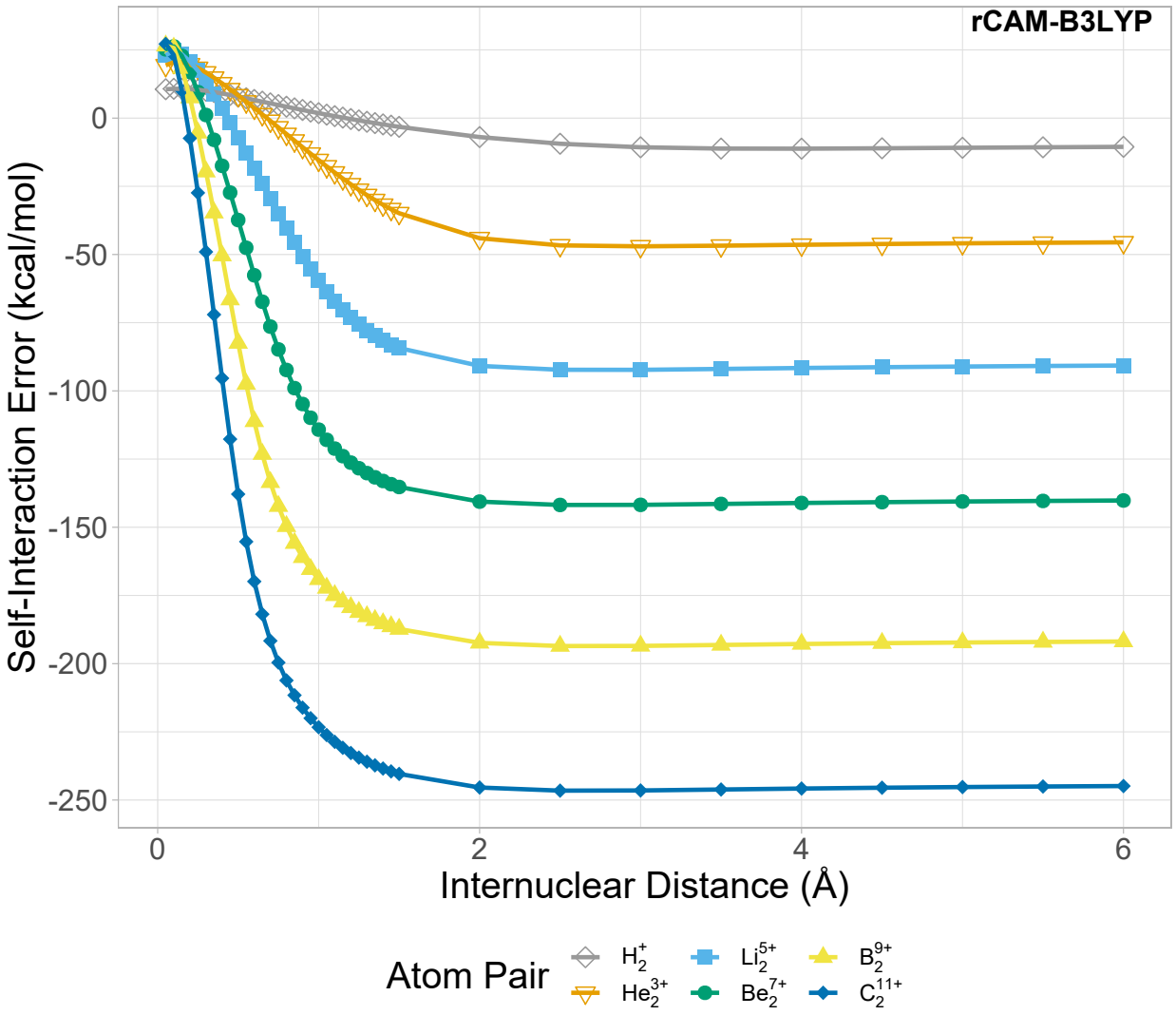

Figure S197: Self-interaction error vs internuclear distance for incremental homonuclear pairs for rCAM-B3LYP.

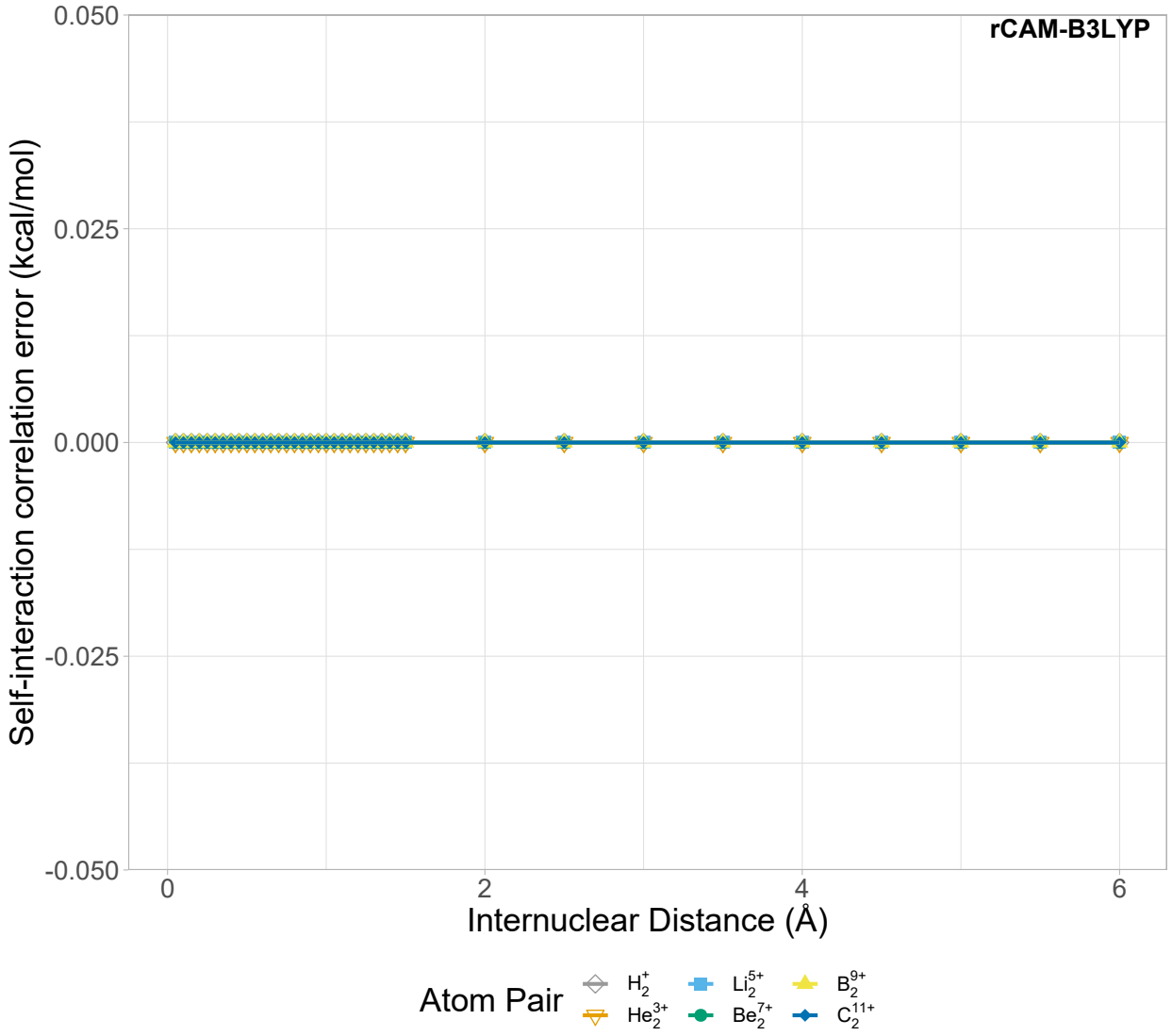

Figure S198: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for rCAM-B3LYP.

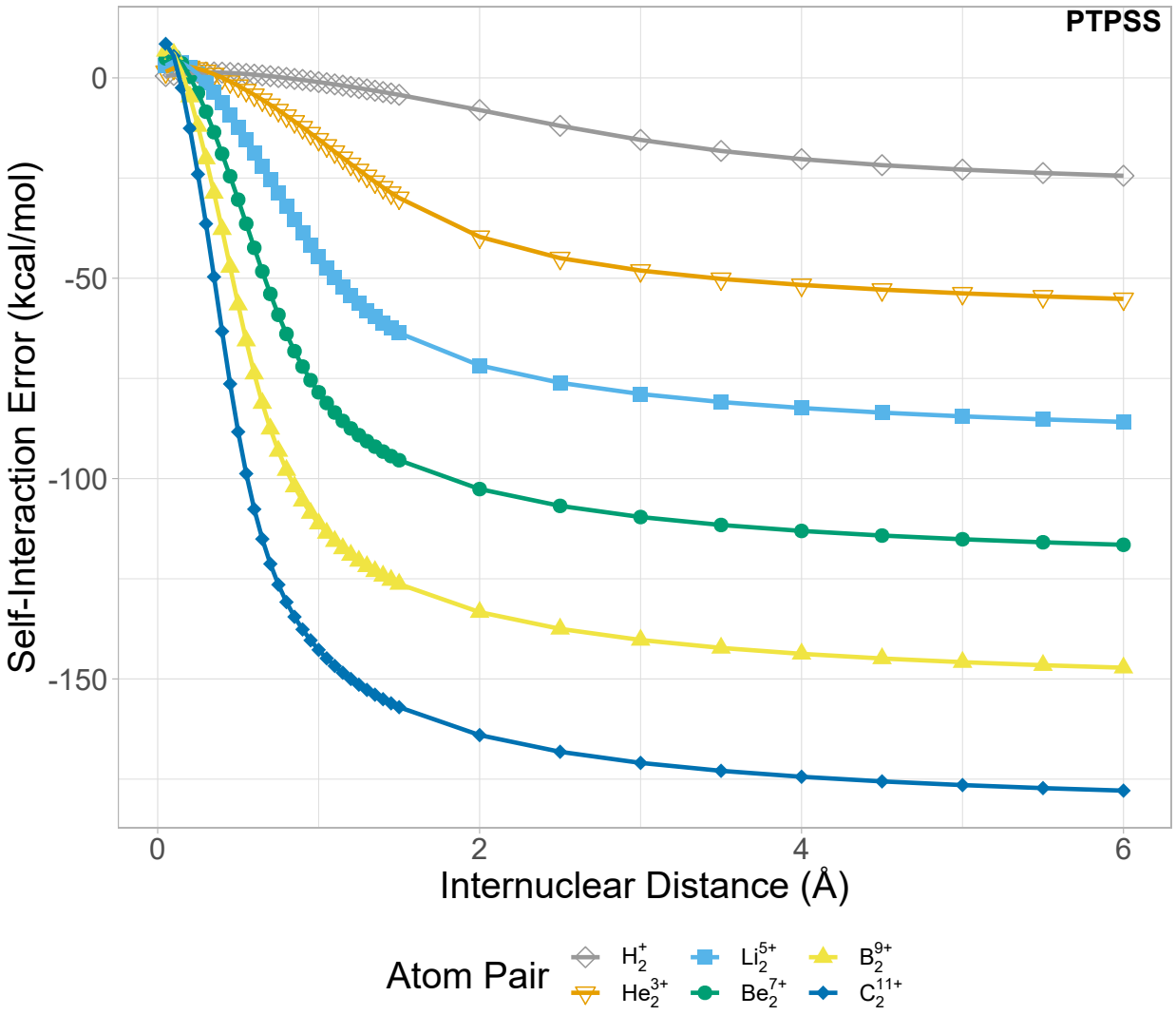

Figure S199: Self-interaction error vs internuclear distance for incremental homonuclear pairs for PTPSS.

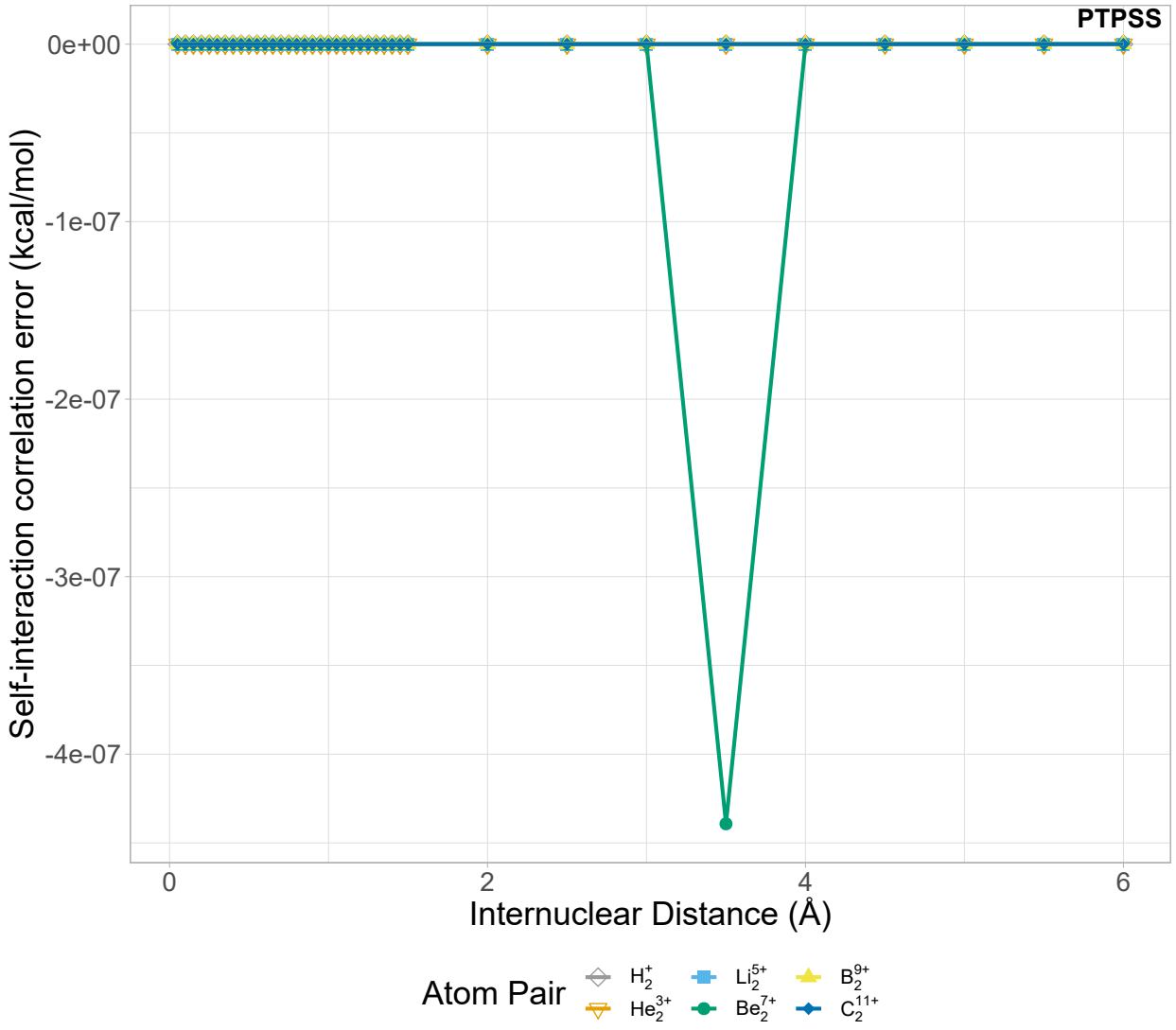

Figure S200: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for PTPSS. Formally, this DFA is self-correlation free, any deviations from this is likely computational noise.

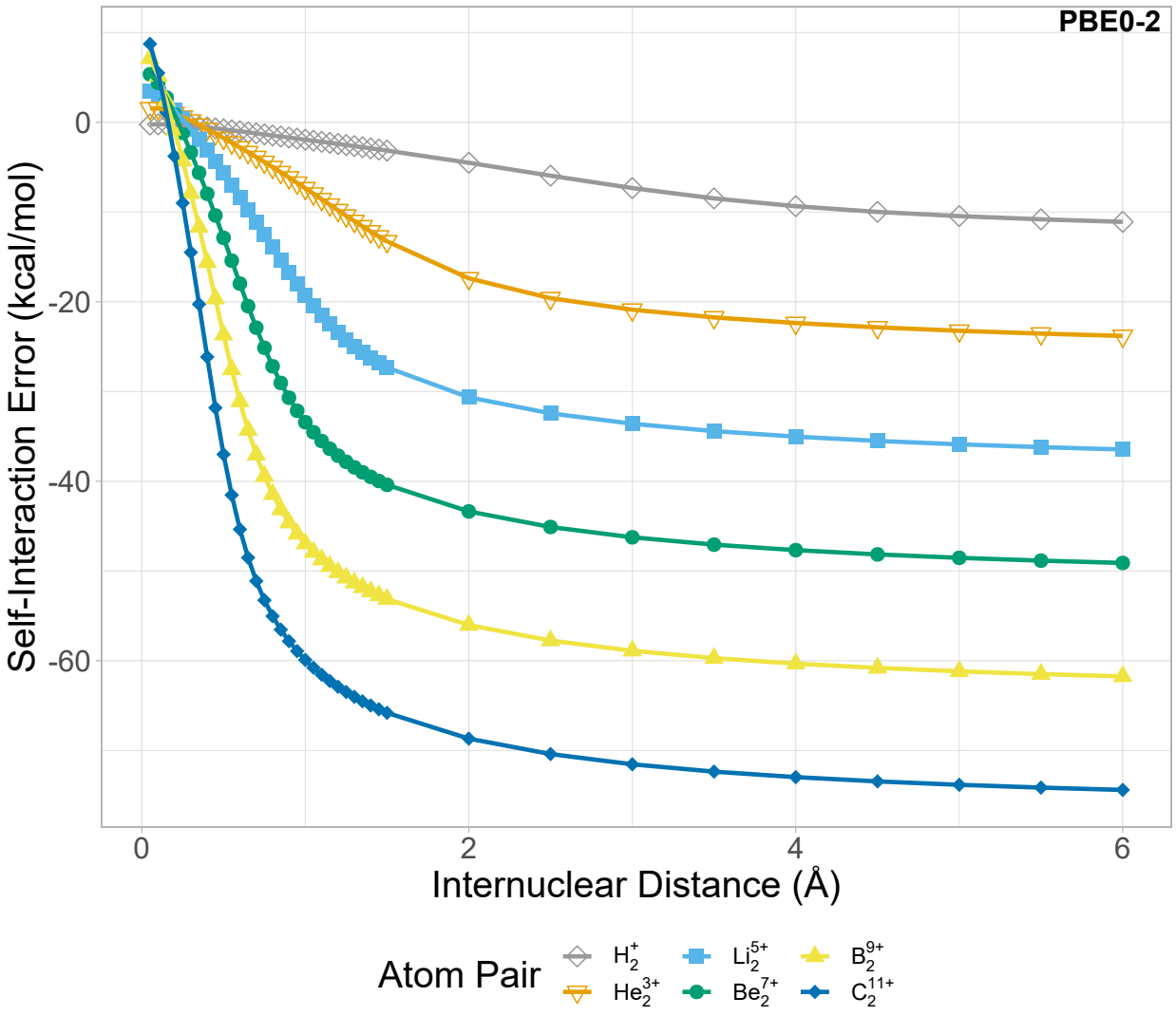

Figure S201: Self-interaction error vs internuclear distance for incremental homonuclear pairs for PBE0-2.

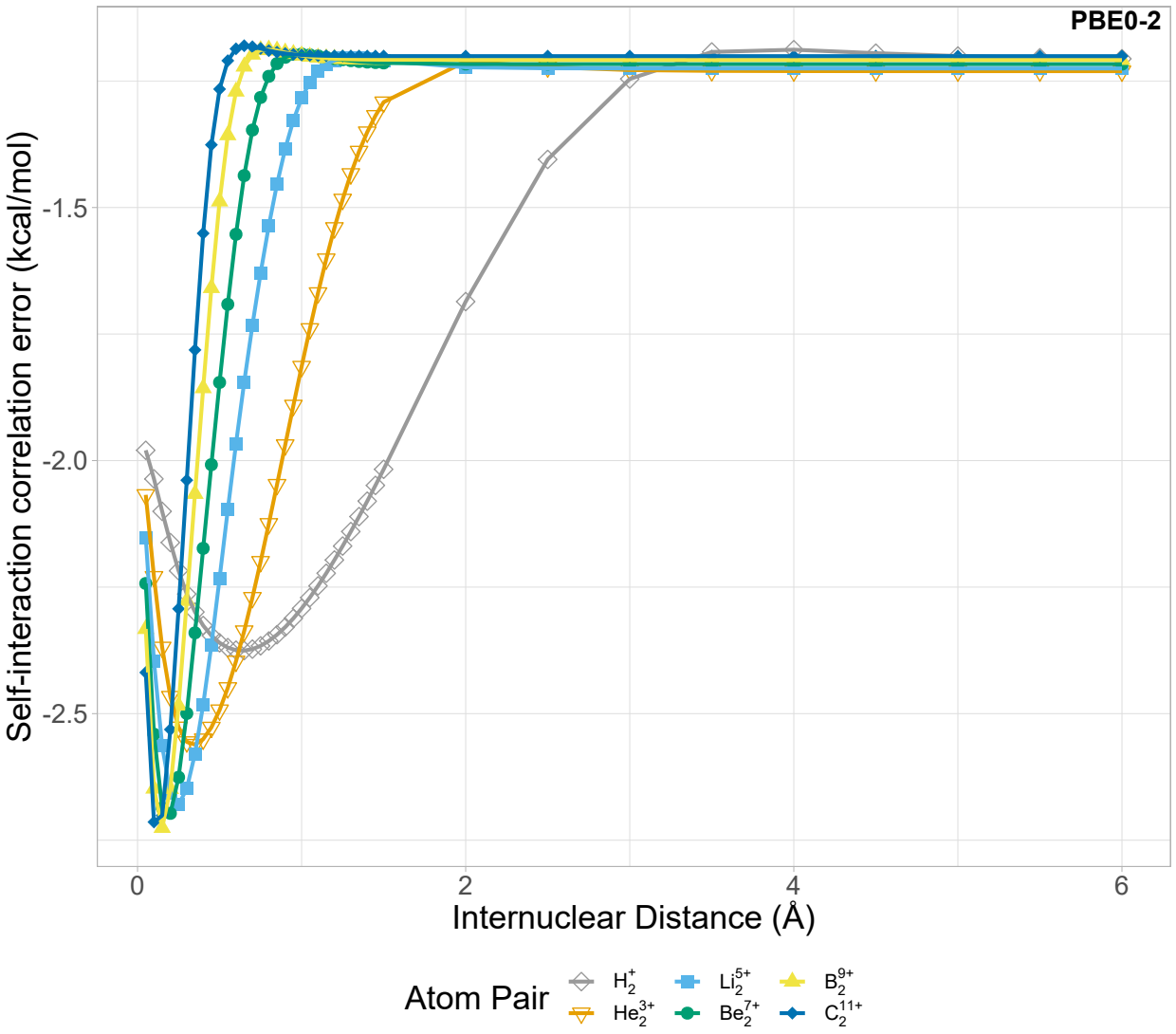

Figure S202: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for PBE0-2.

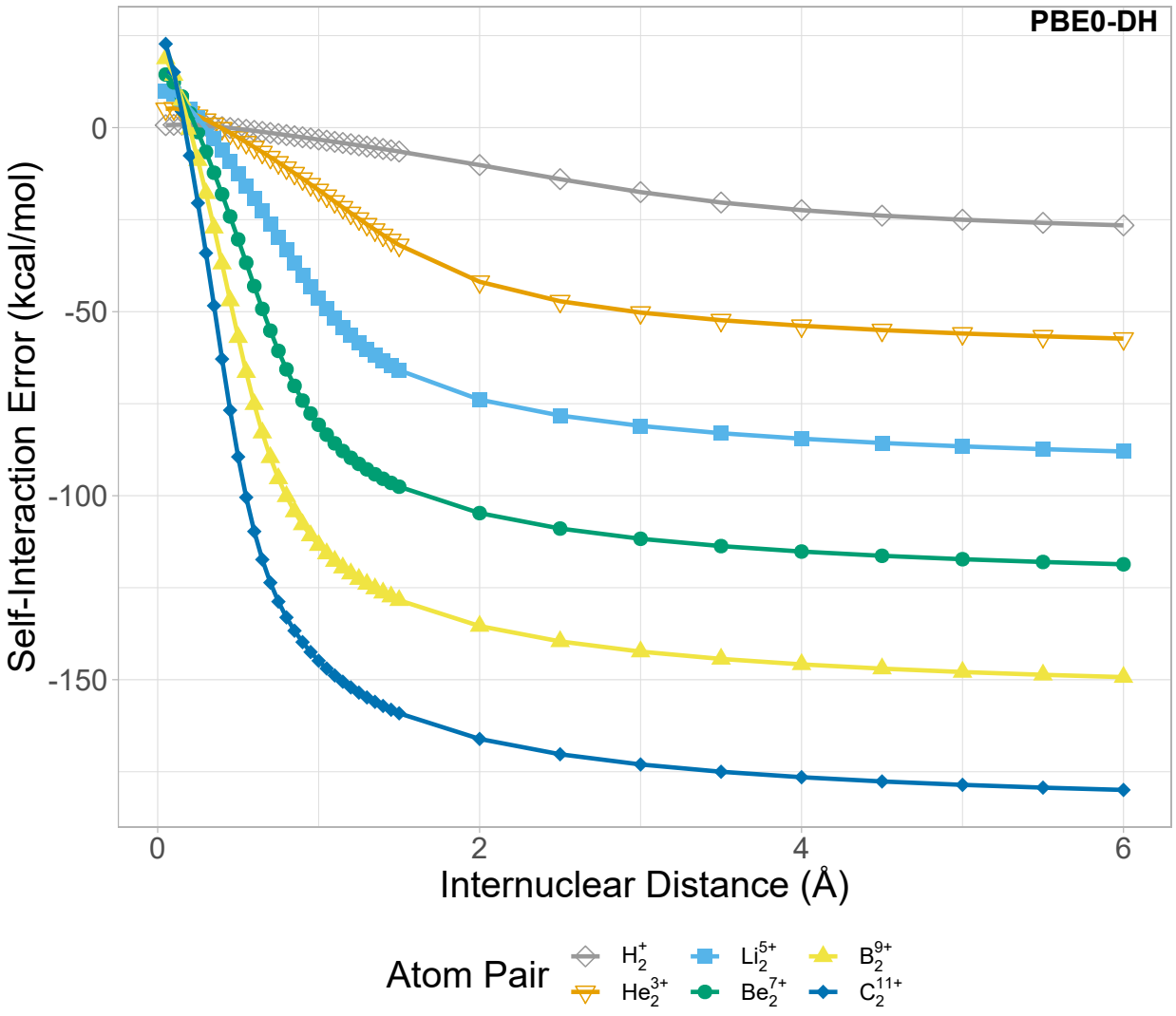

Figure S203: Self-interaction error vs internuclear distance for incremental homonuclear pairs for PBE0-DH.

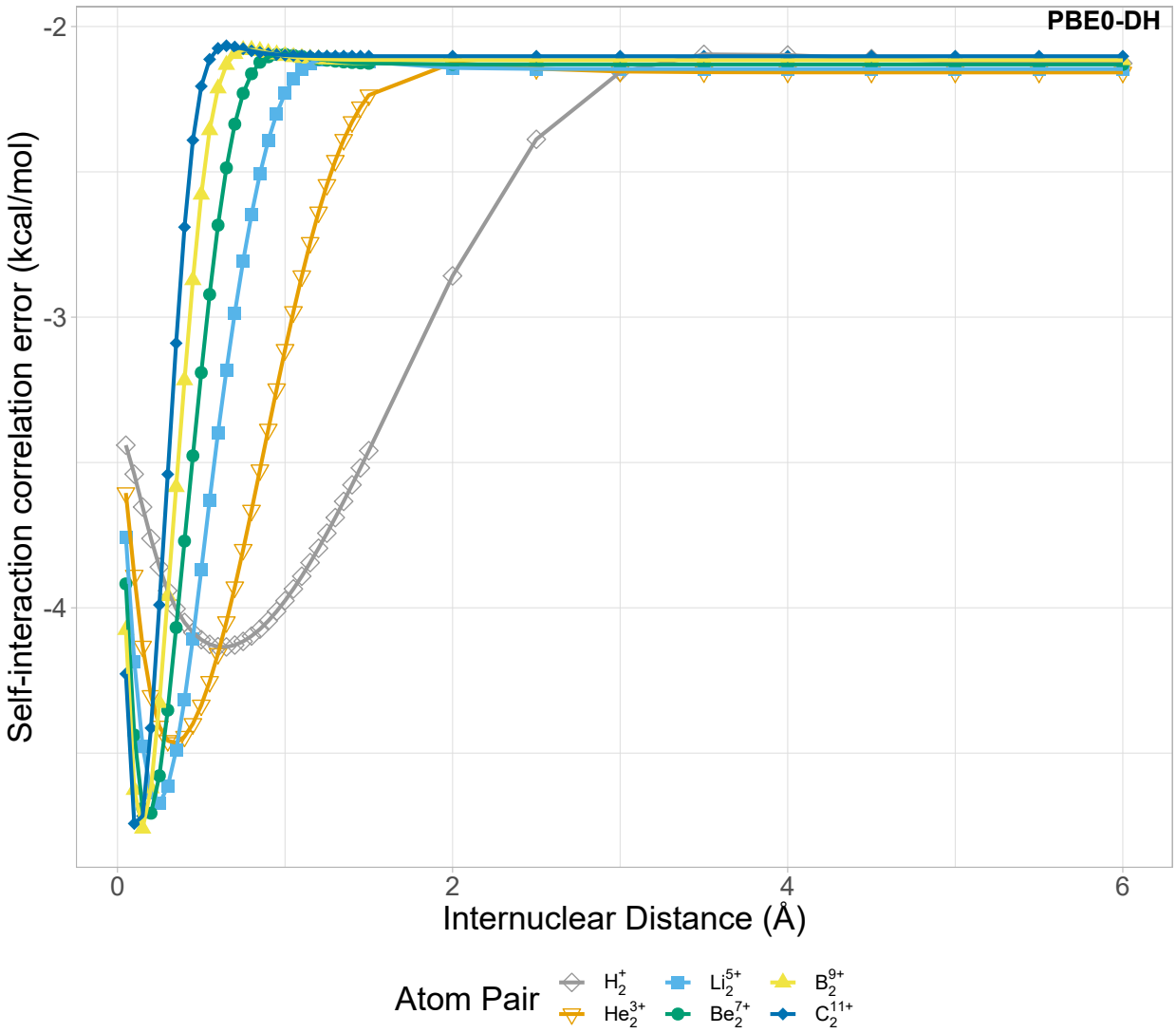

Figure S204: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for PBE0-DH.

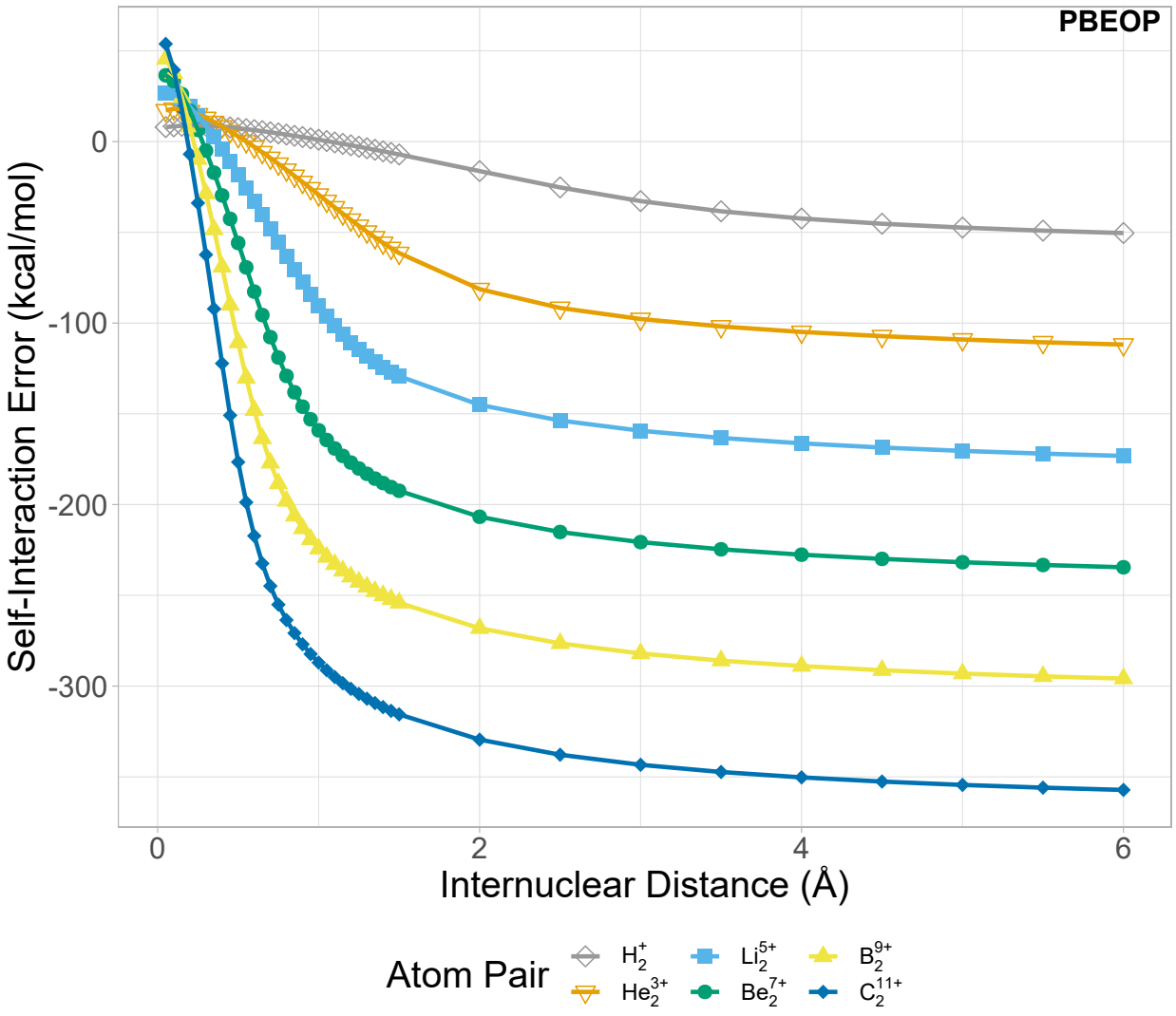

Figure S205: Self-interaction error vs internuclear distance for incremental homonuclear pairs for PBEOP.

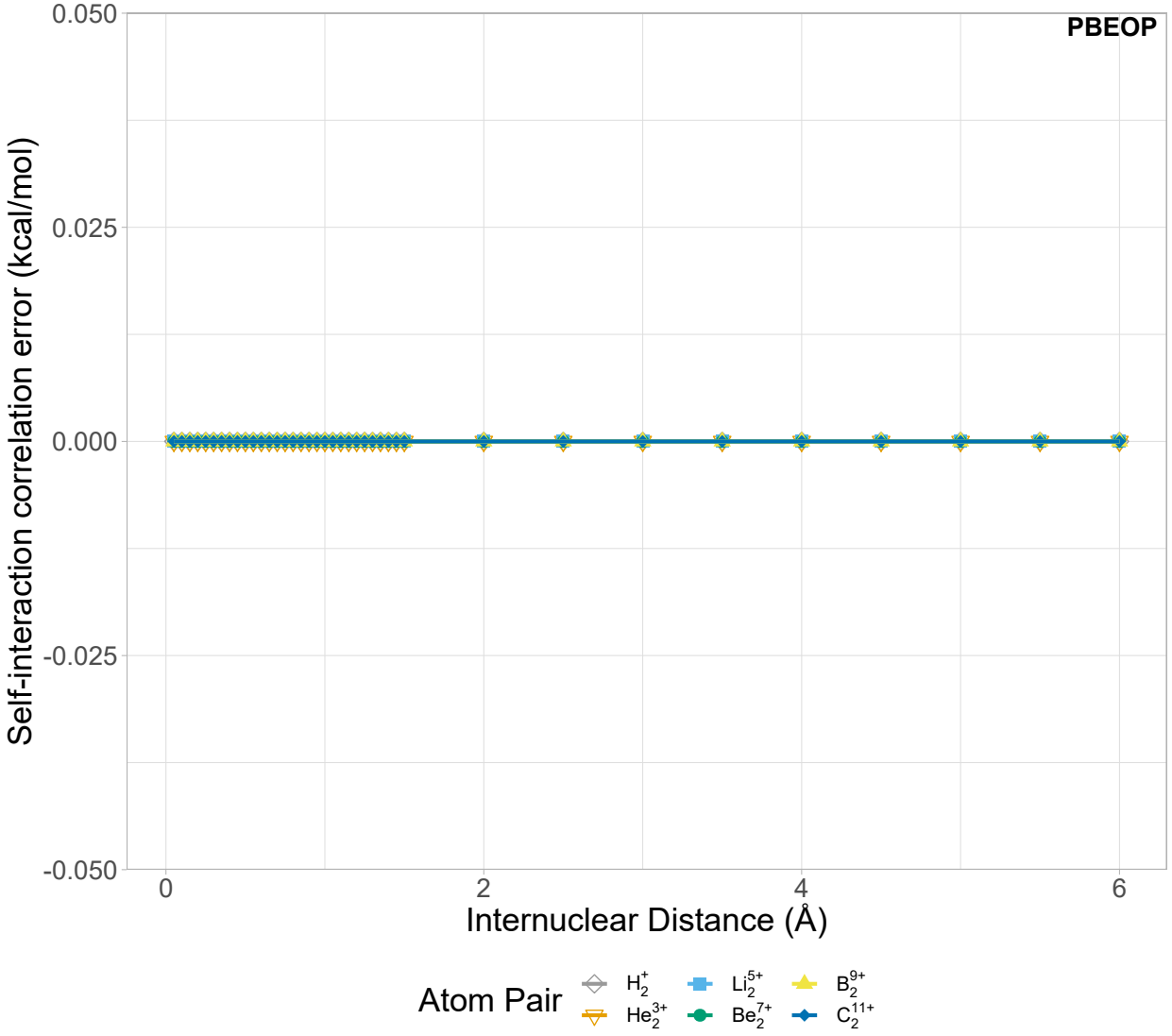

Figure S206: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for PBEOP.

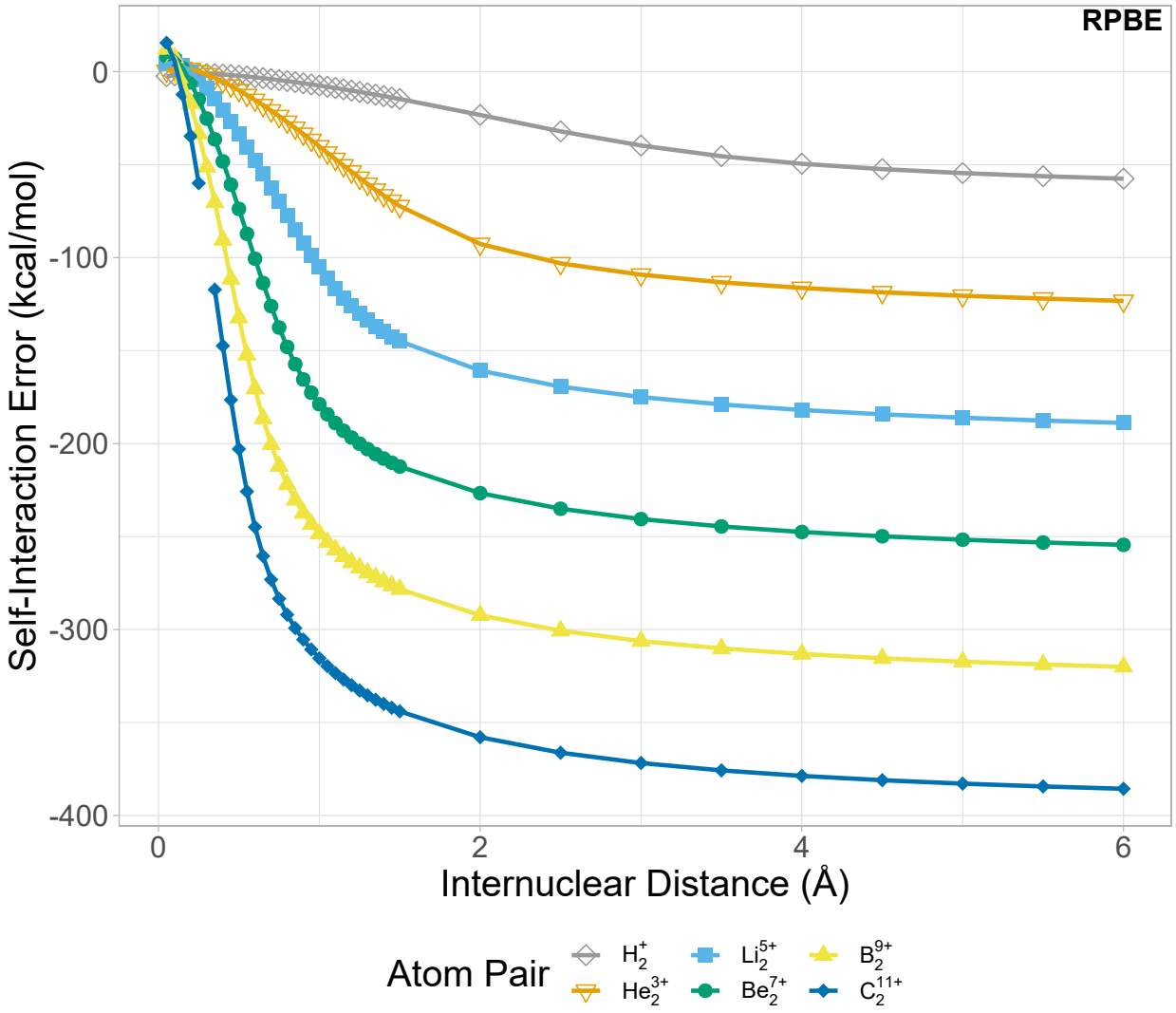

Figure S207: Self-interaction error vs internuclear distance for incremental homonuclear pairs for RPBE.

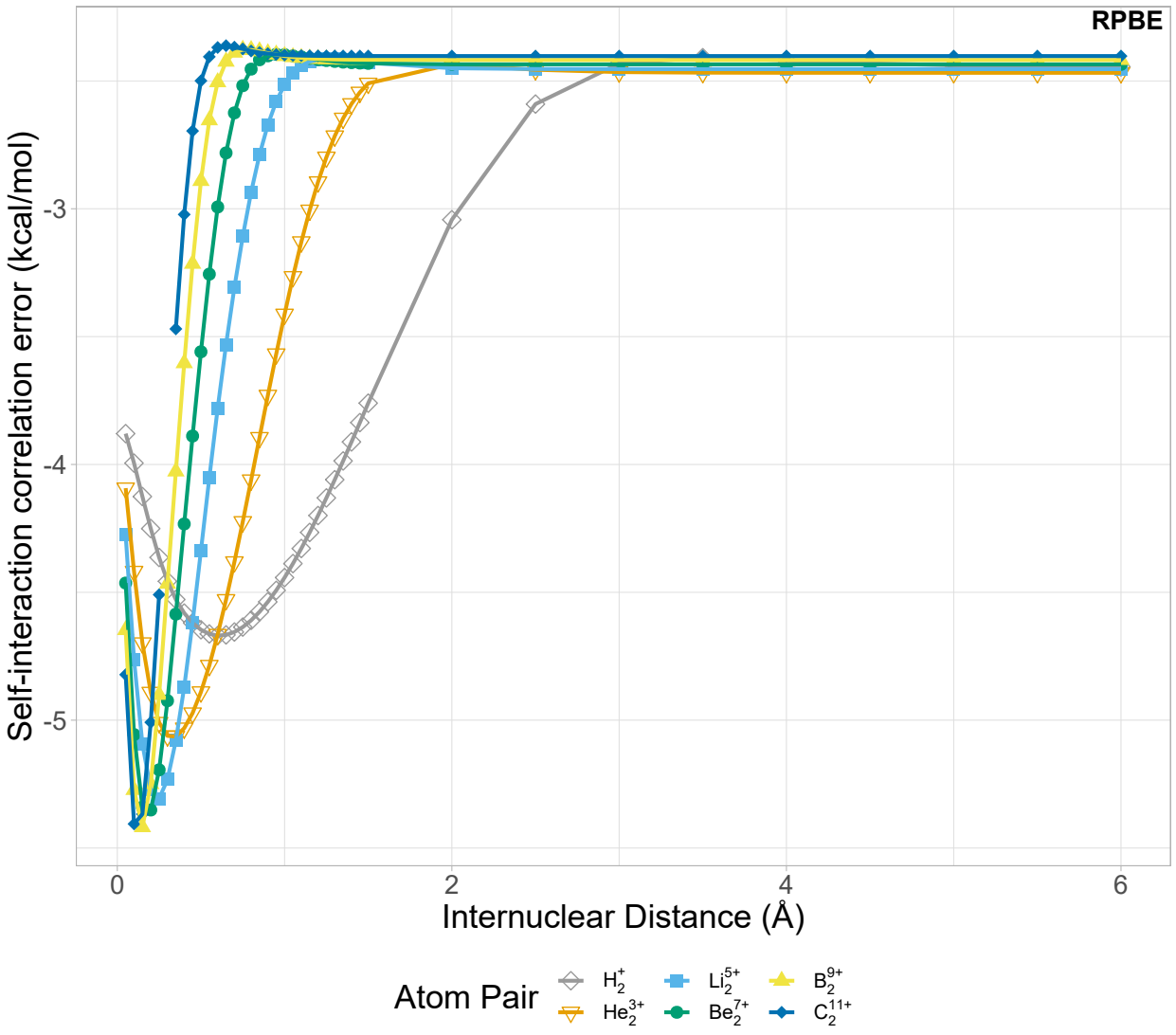

Figure S208: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for RPBE.

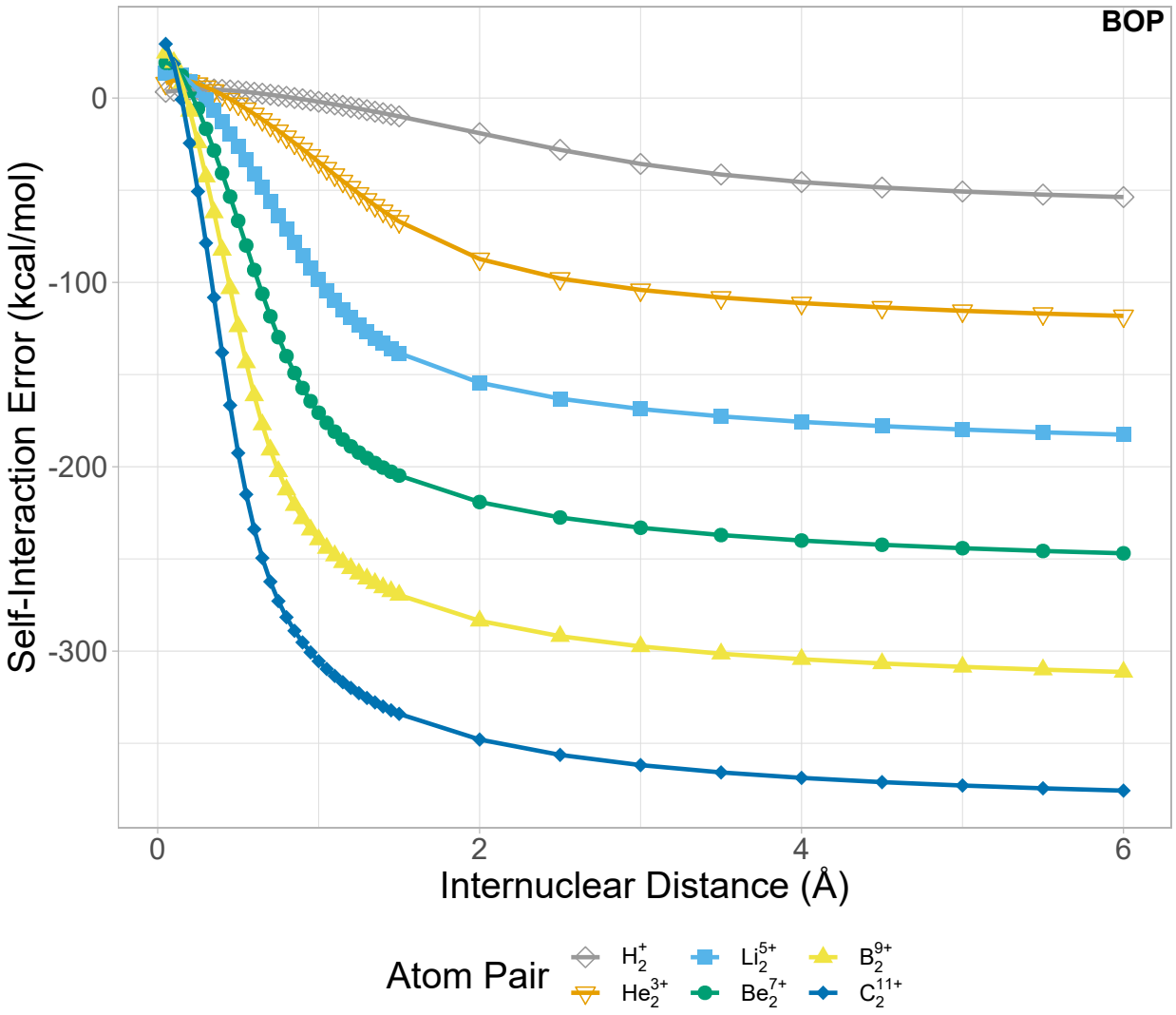

Figure S209: Self-interaction error vs internuclear distance for incremental homonuclear pairs for BOP.

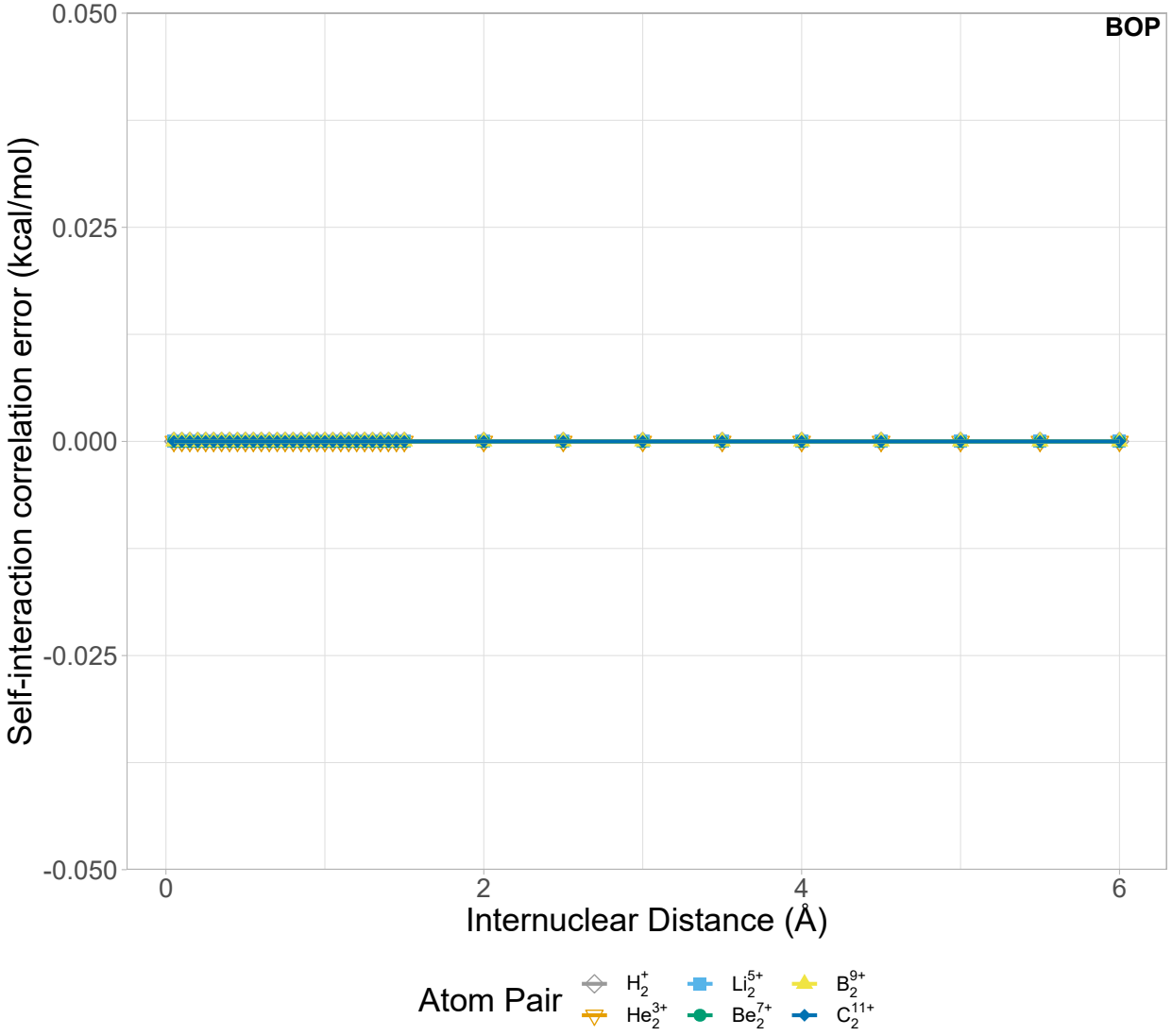

Figure S210: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for BOP.

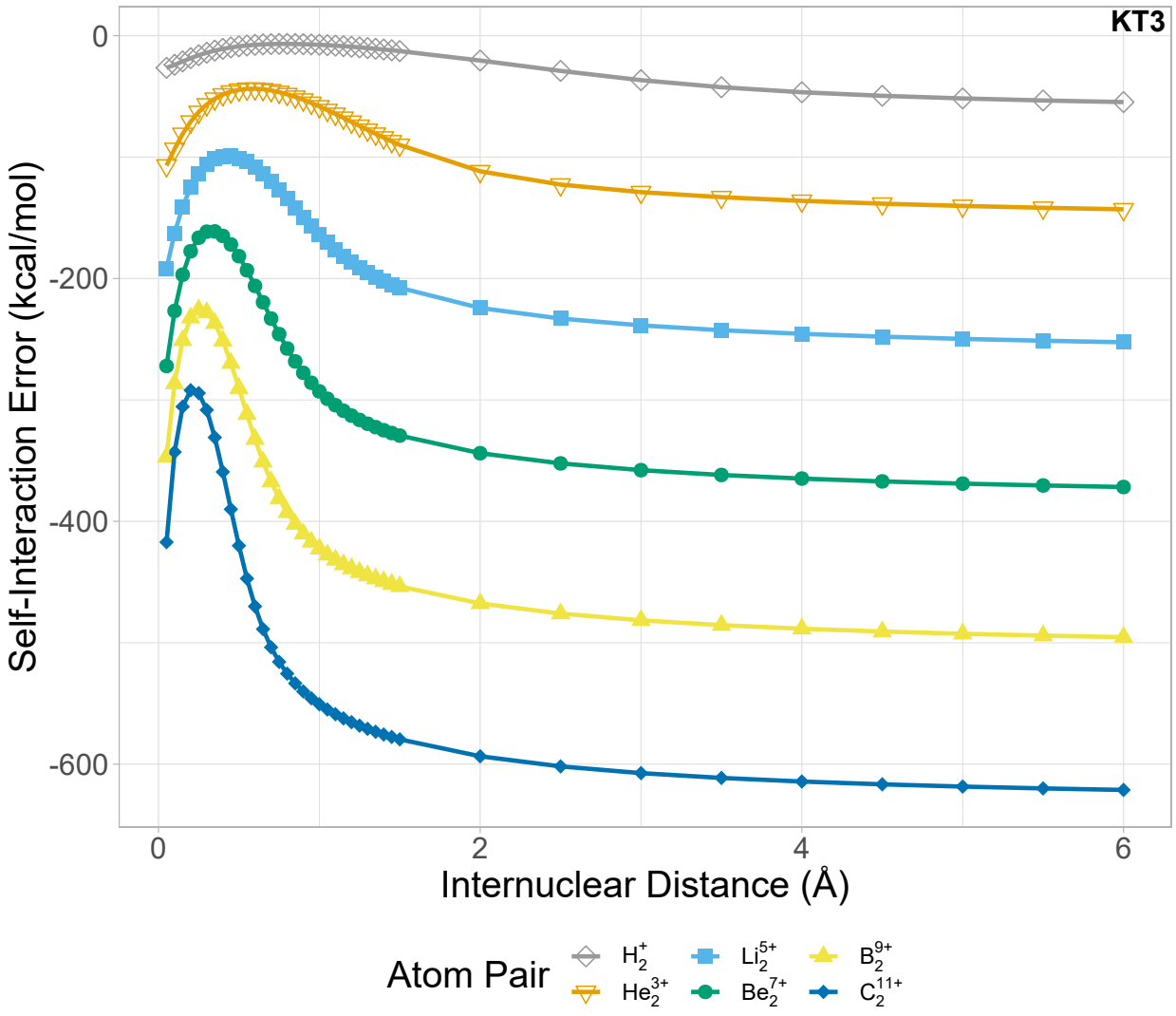

Figure S211: Self-interaction error vs internuclear distance for incremental homonuclear pairs for KT3.

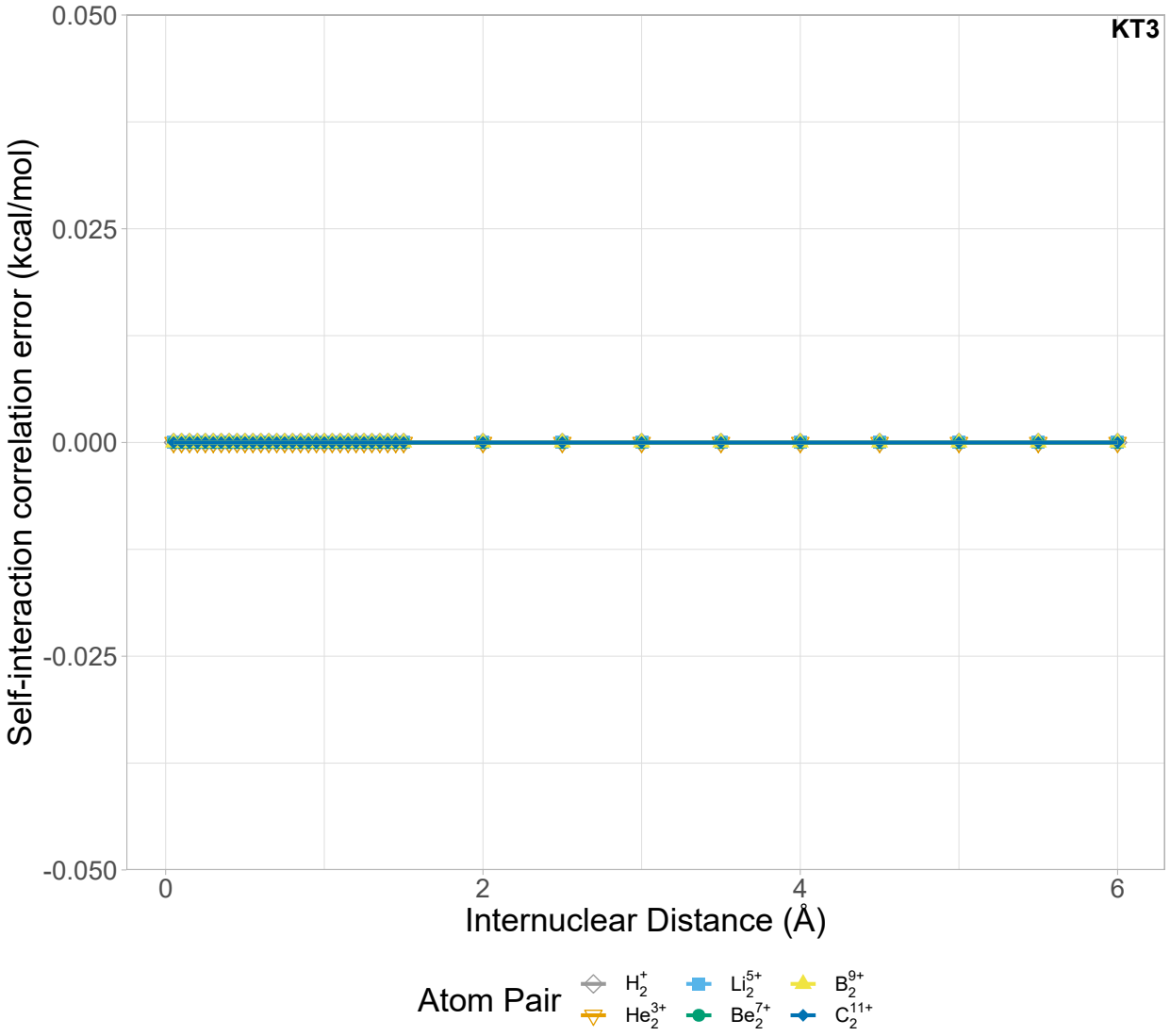

Figure S212: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for KT3.

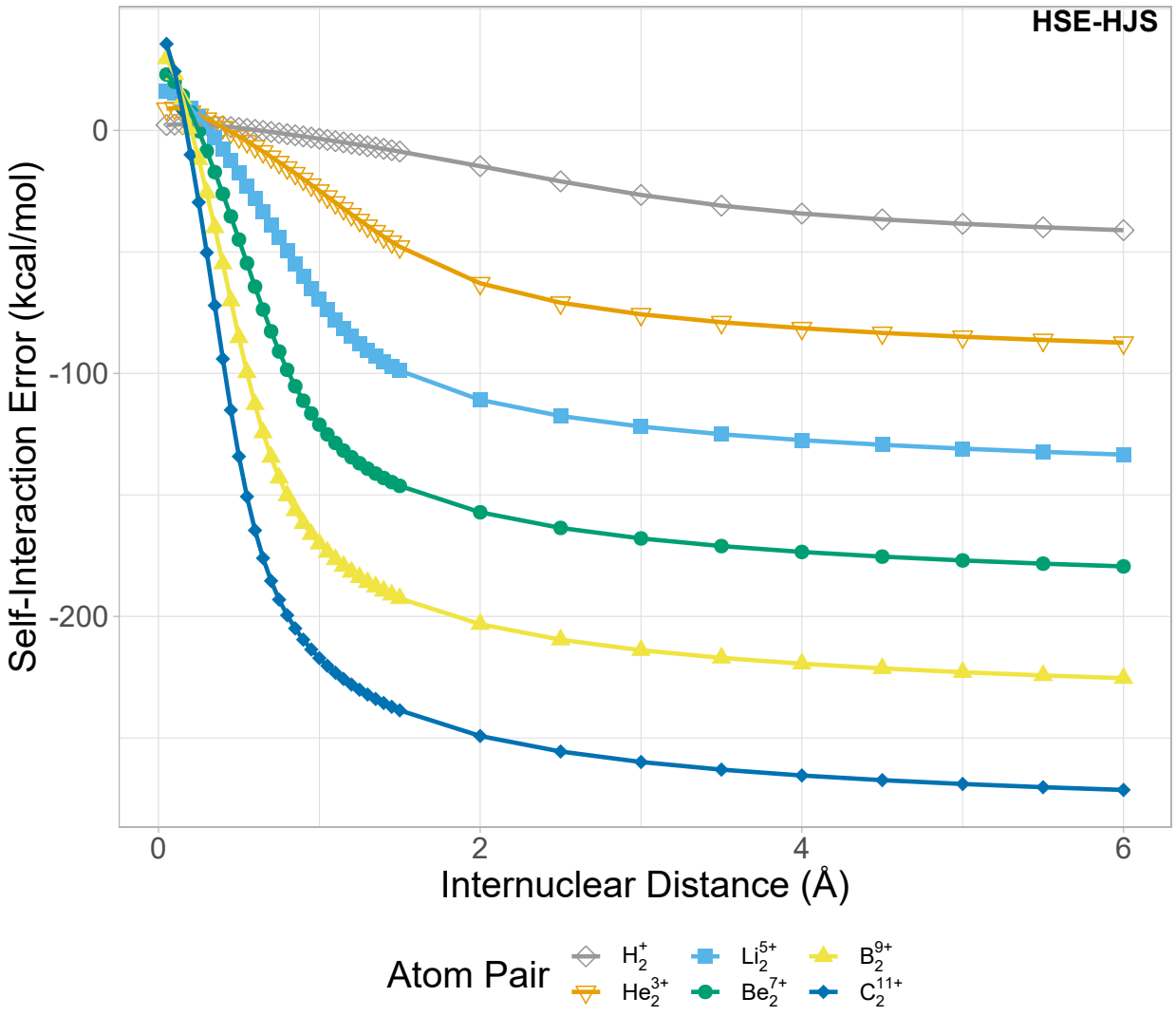

Figure S213: Self-interaction error vs internuclear distance for incremental homonuclear pairs for HSE-HJS.

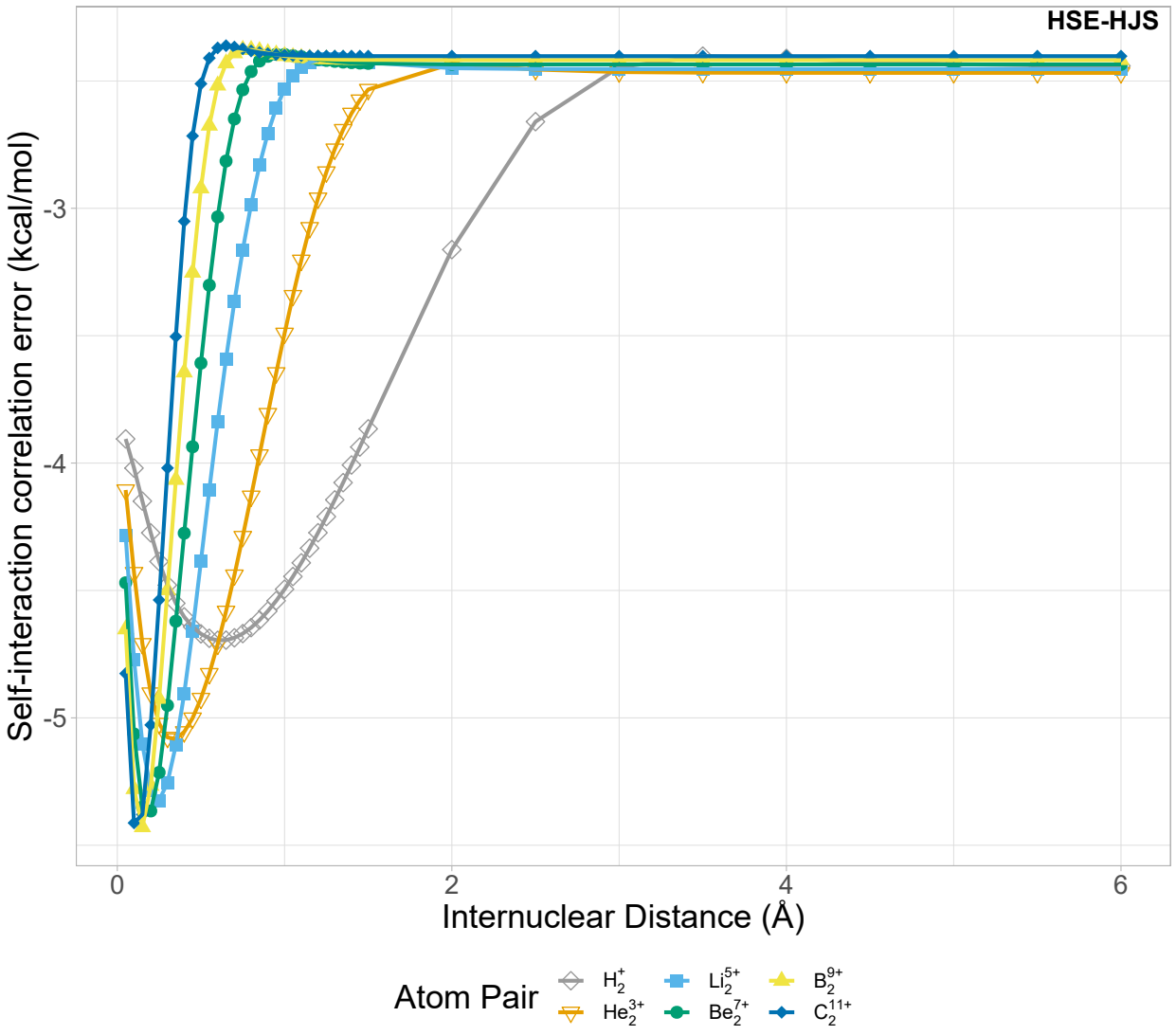

Figure S214: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for HSE-HJS.

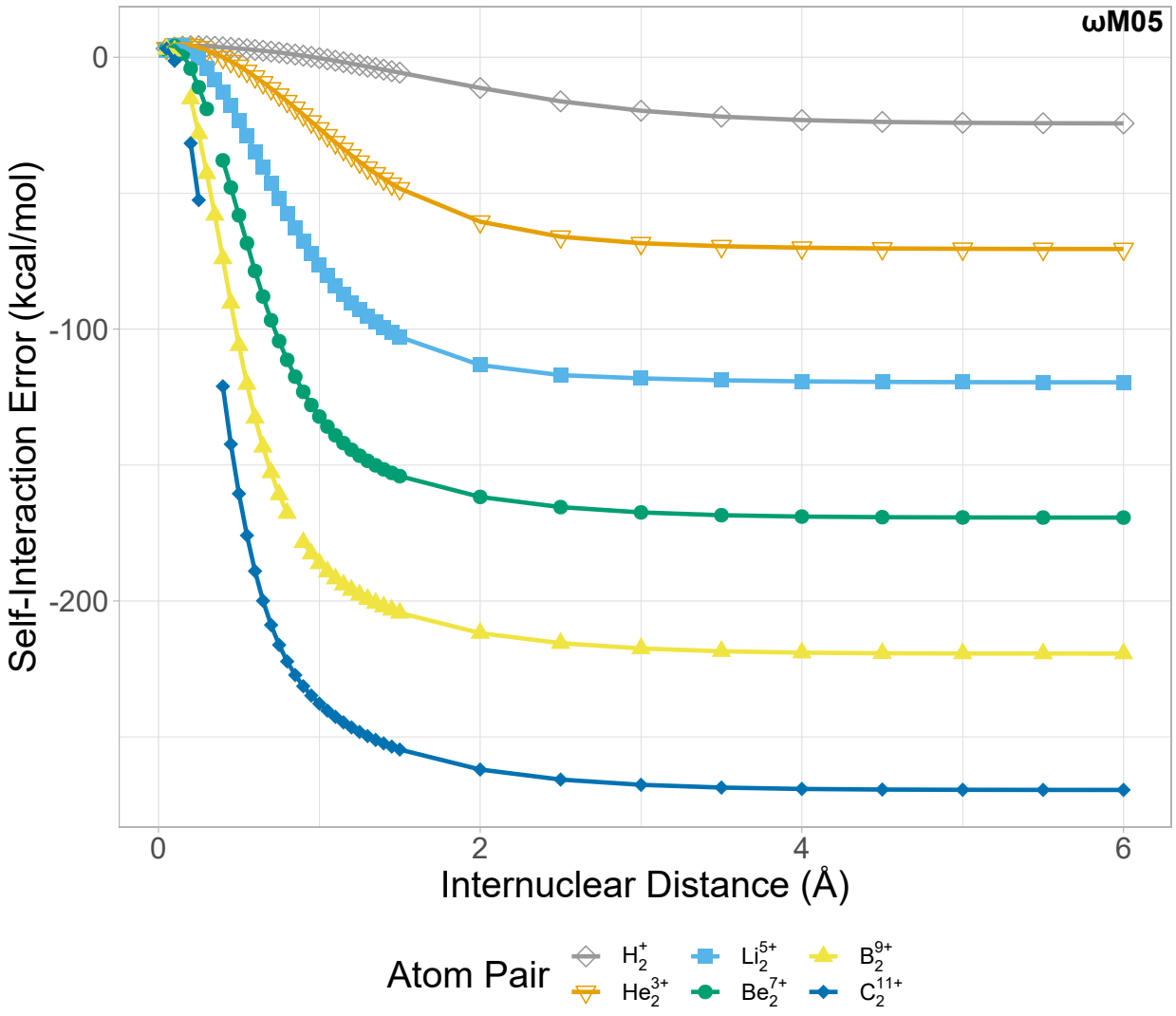

Figure S215: Self-interaction error vs internuclear distance for incremental homonuclear pairs for ωM05-D.

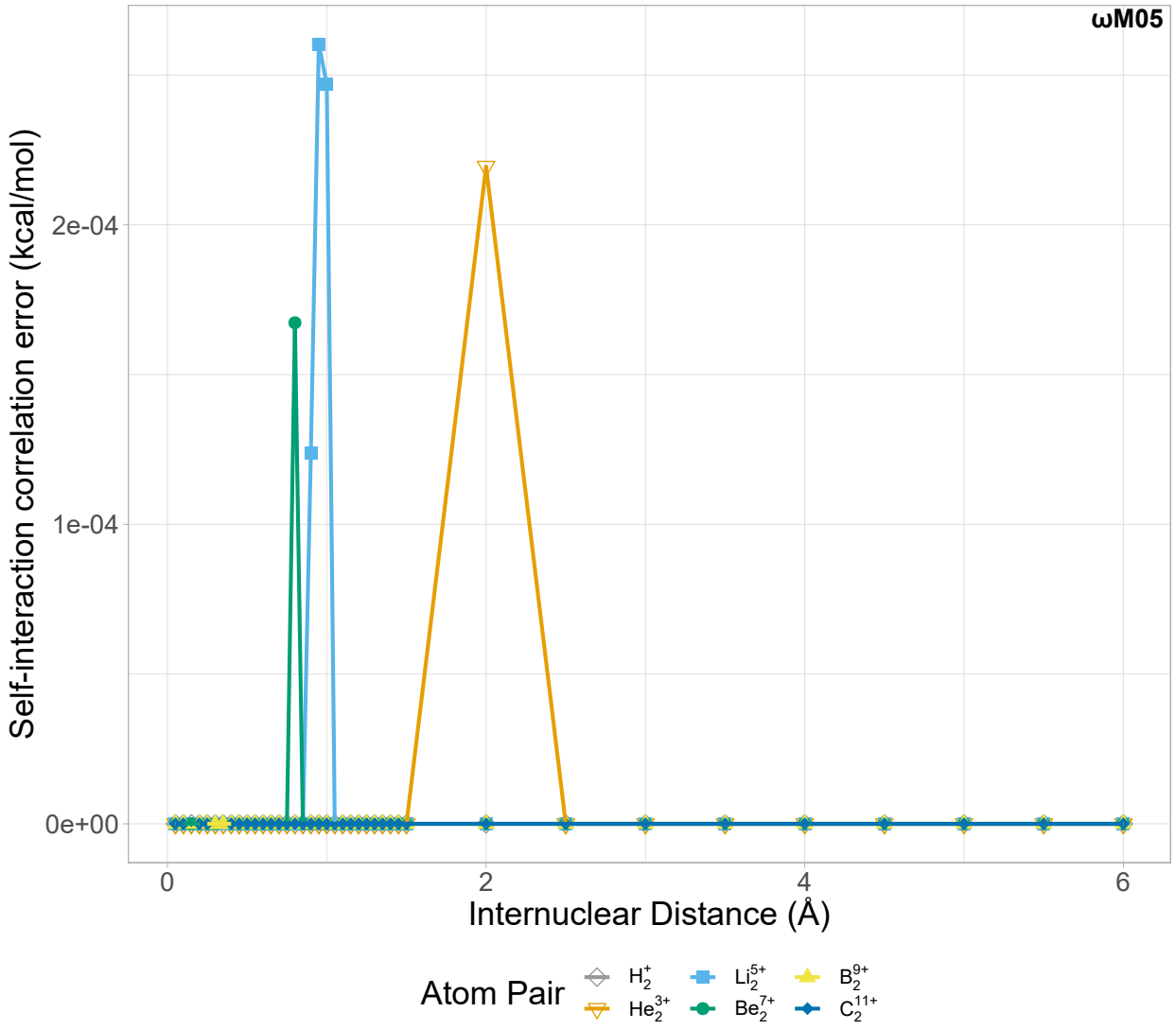

Figure S216: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for ωM05-D. Formally, this DFA is self-correlation free, any deviations from this is likely computational noise.

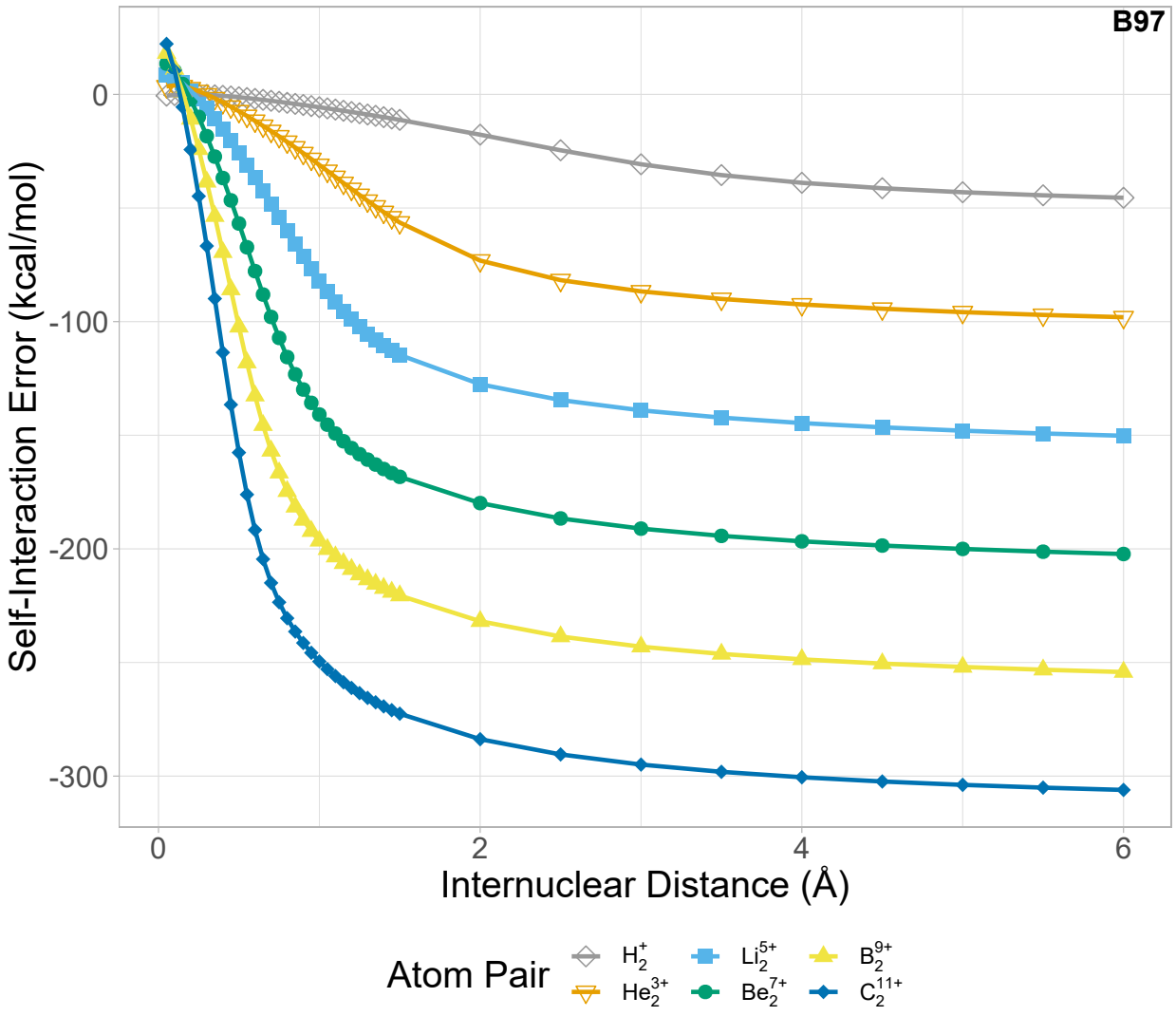

Figure S217: Self-interaction error vs internuclear distance for incremental homonuclear pairs for B97.

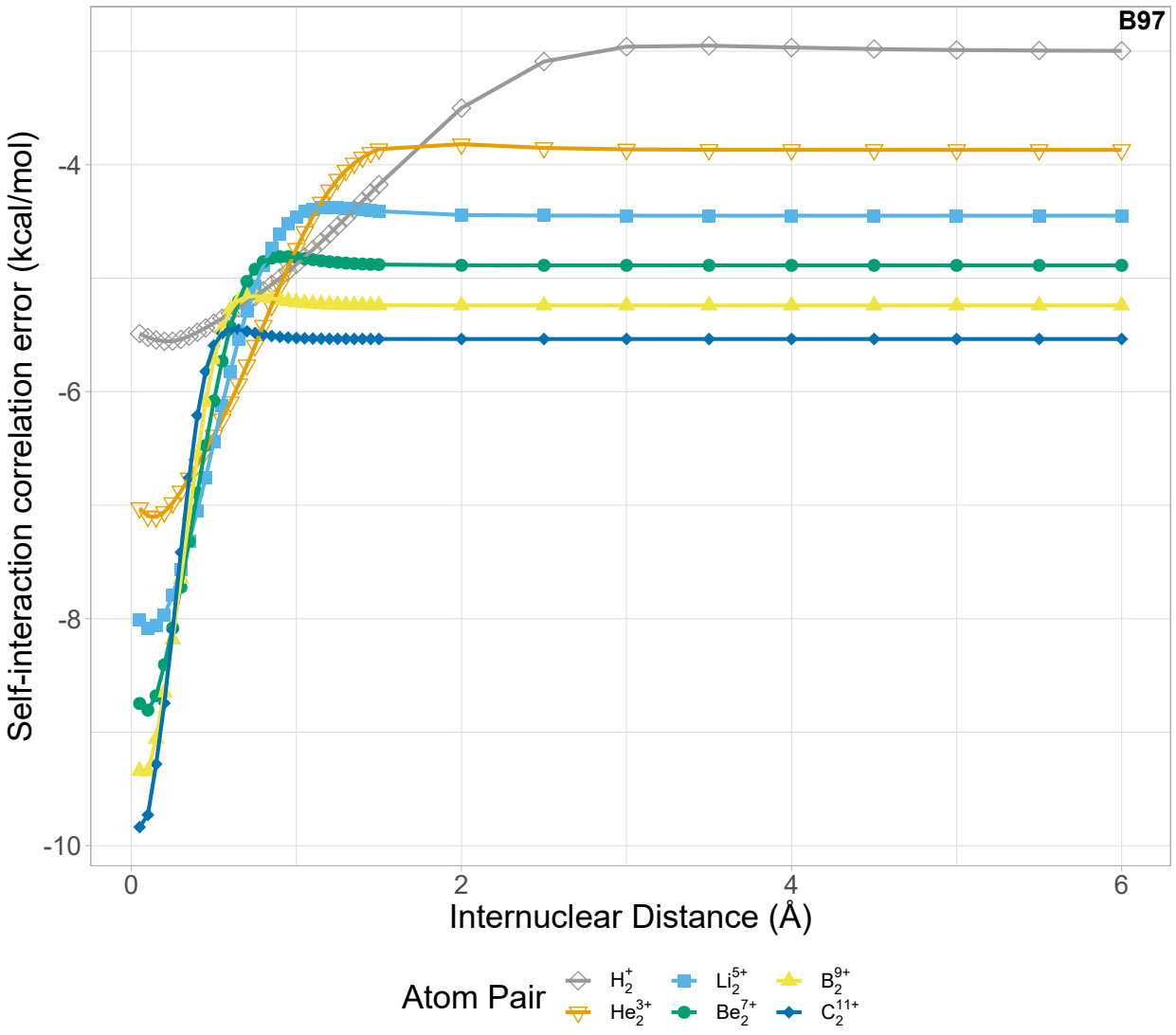

Figure S218: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for B97.

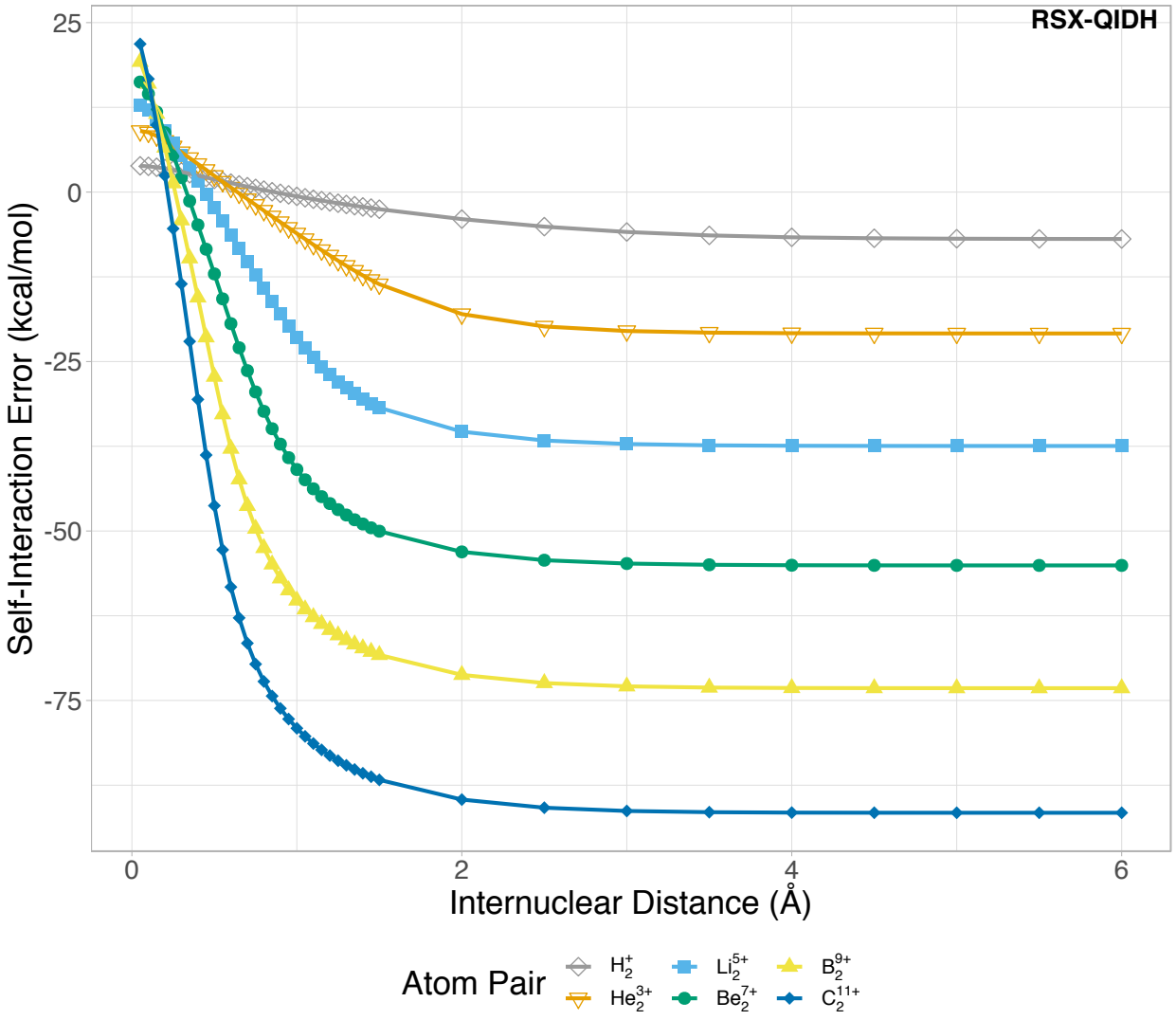

Figure S219: Self-interaction error vs internuclear distance for incremental homonuclear pairs for RSX-QIDH.

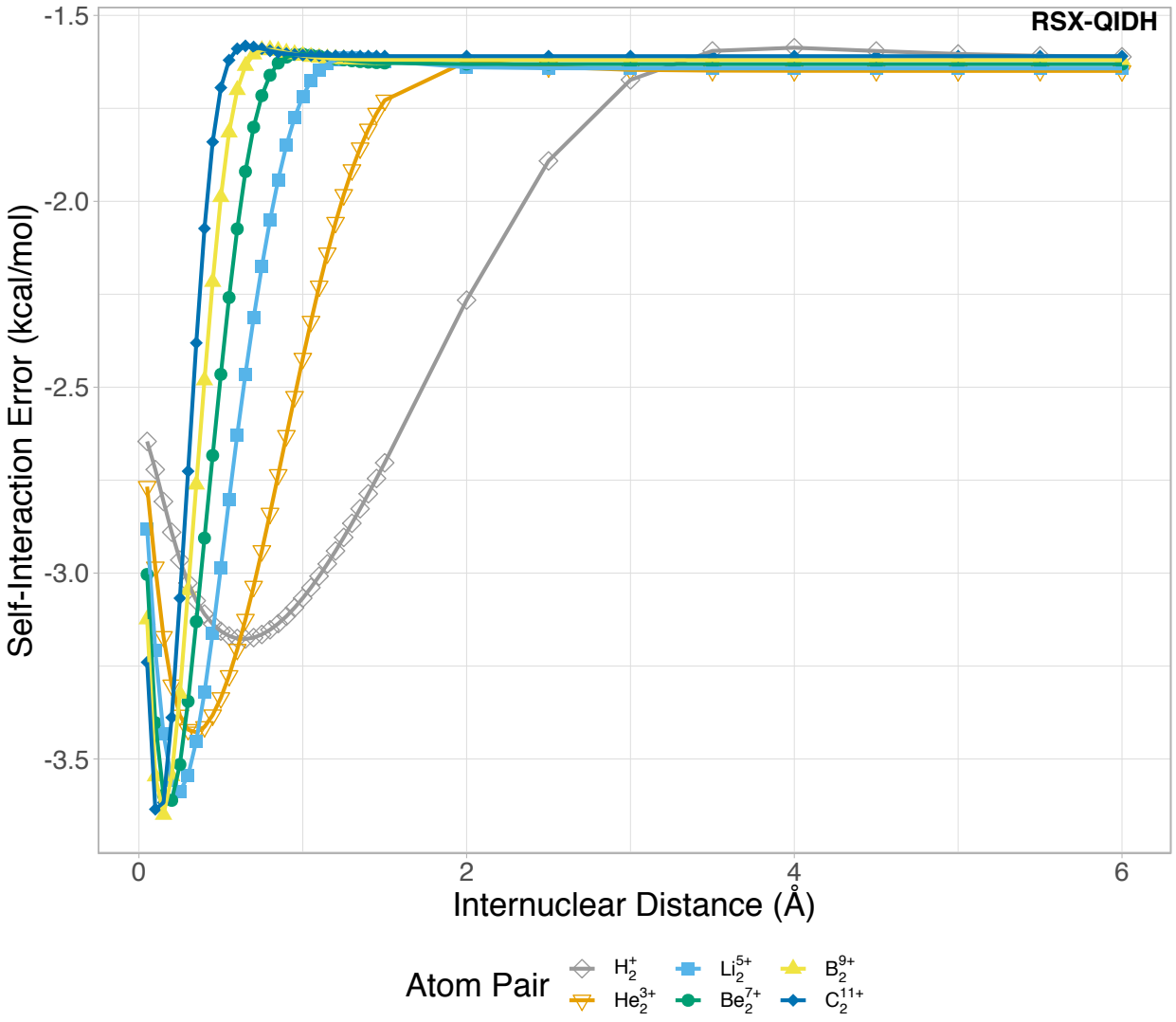

Figure S220: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for RSX-QIDH.

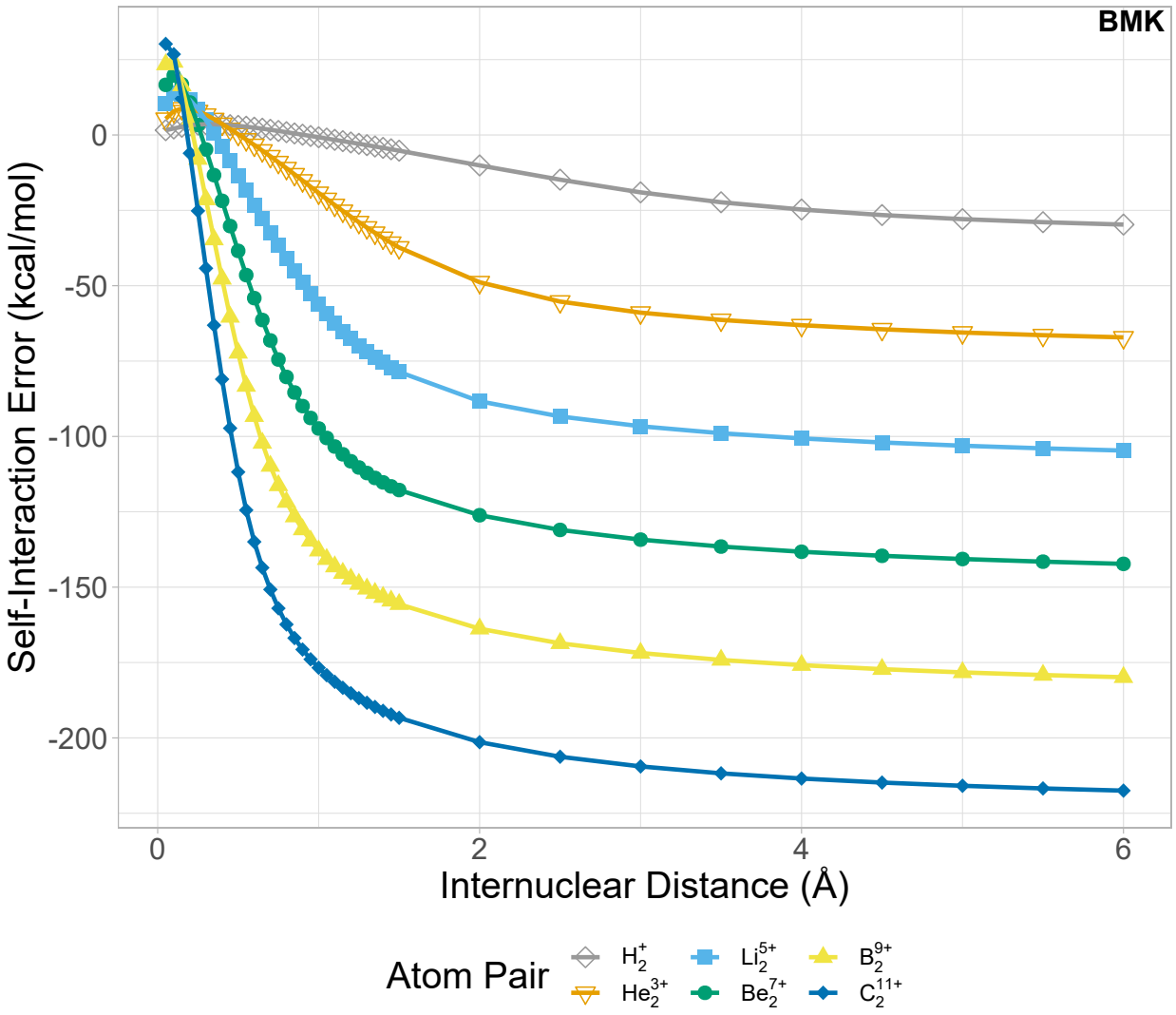

Figure S221: Self-interaction error vs internuclear distance for incremental homonuclear pairs for BMK.

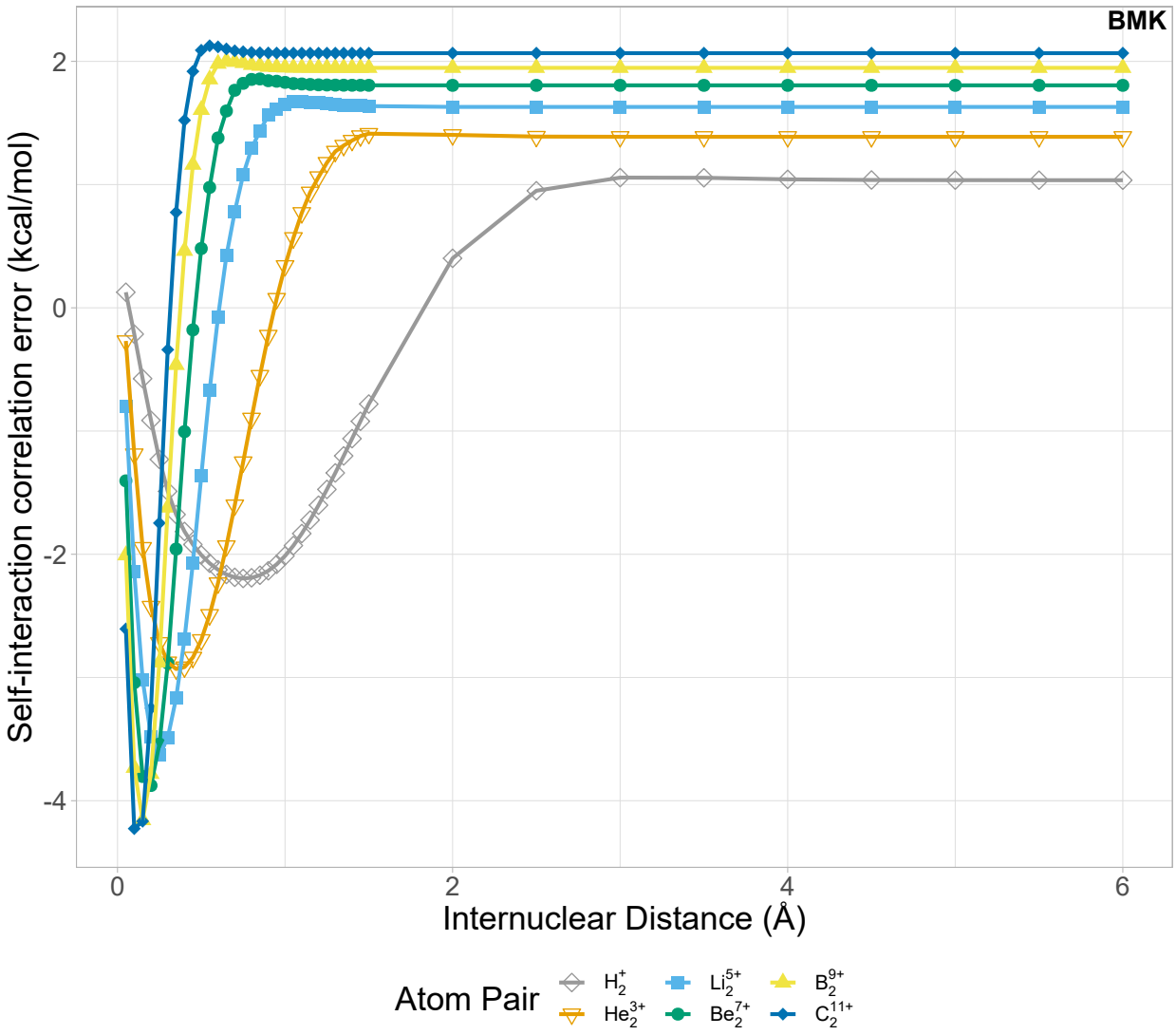

Figure S222: Self-interaction correlation error vs internuclear distance for incremental homonuclear pairs for BMK.

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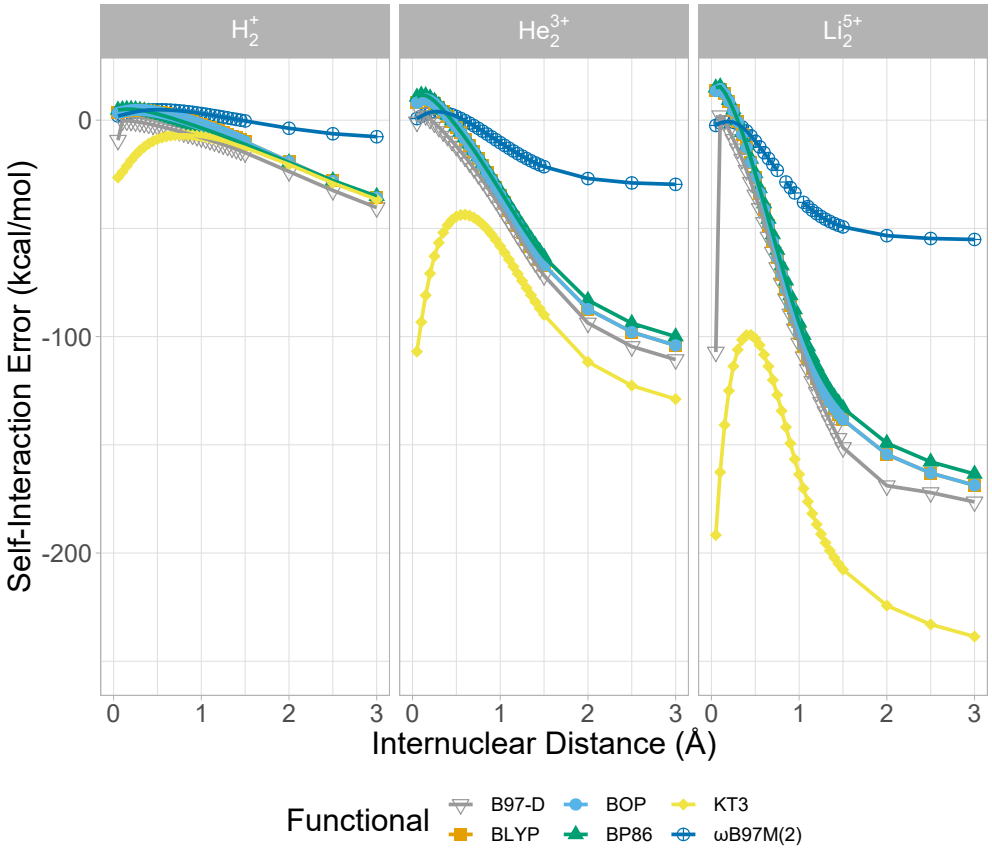

Figure S223: Self-interaction error vs internuclear distance of the homonuclear pairs, H_2^+, He_2^{3+}, Li_2^{5+}, for B97-D, BLYP, BOP, BP86, KT3 with ωB97M(2) as a reference.

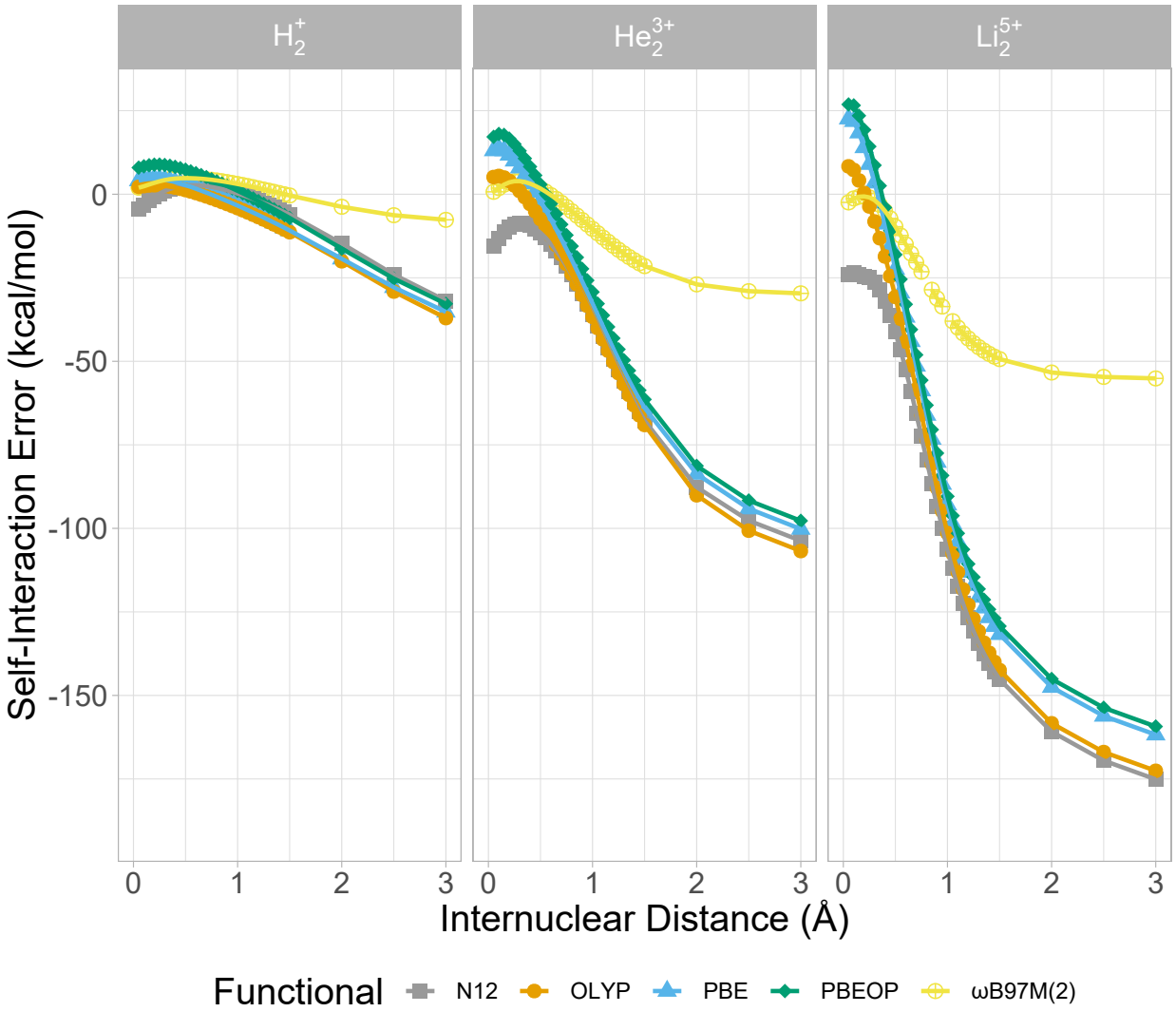

Figure S224: Self-interaction error vs internuclear distance of the homonuclear pairs, H_2^+, He_2^{3+}, Li_2^{5+}, for N12, OLYP, PBE, PBEOP with ωB97M(2) as a reference.

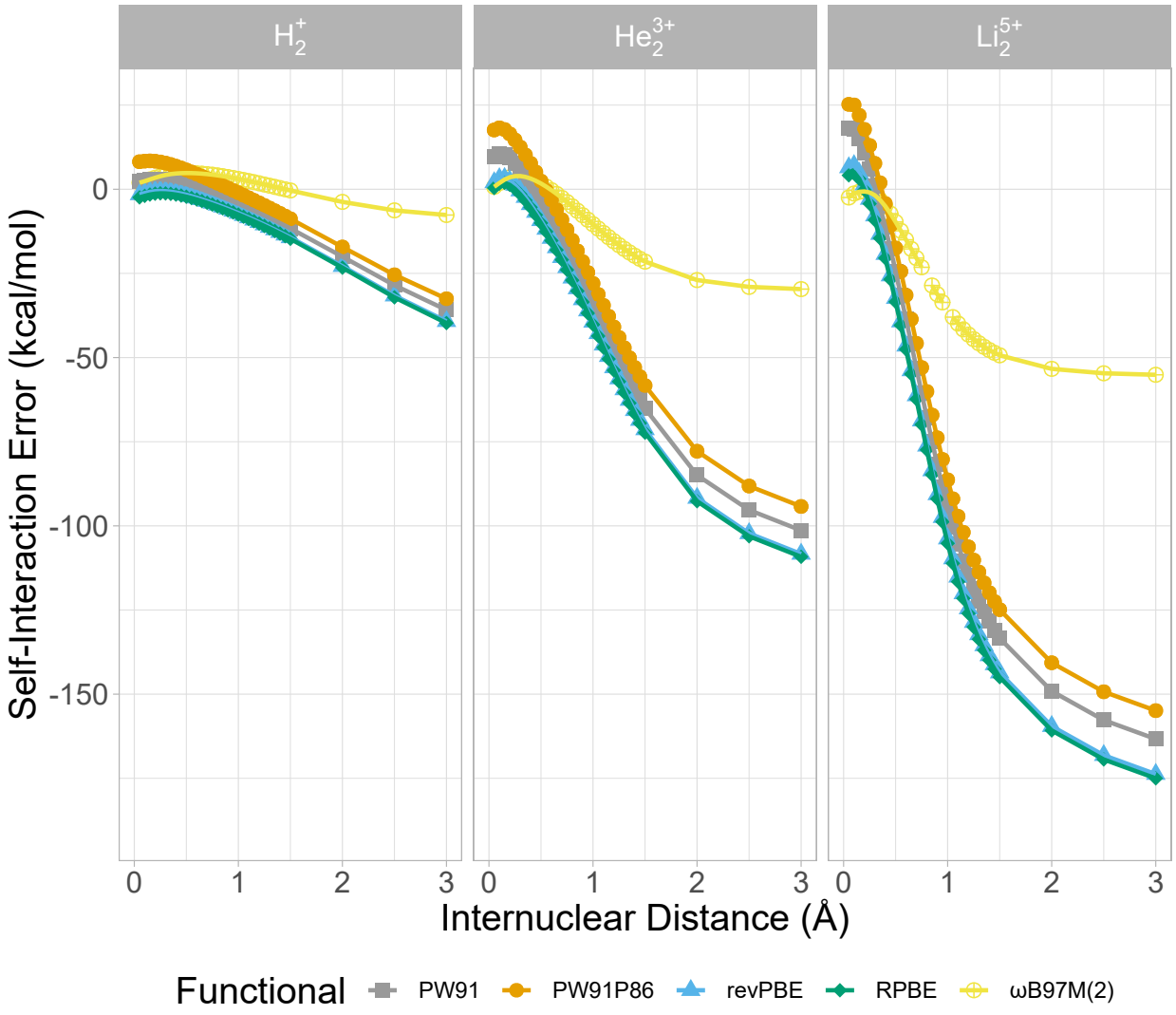

Figure S225: Self-interaction error vs internuclear distance of the homonuclear pairs, H_2^+, He_2^{3+}, Li_2^{5+}, for PW91, PW91P86, revPBE, RPBE with ωB97M(2) as a reference.

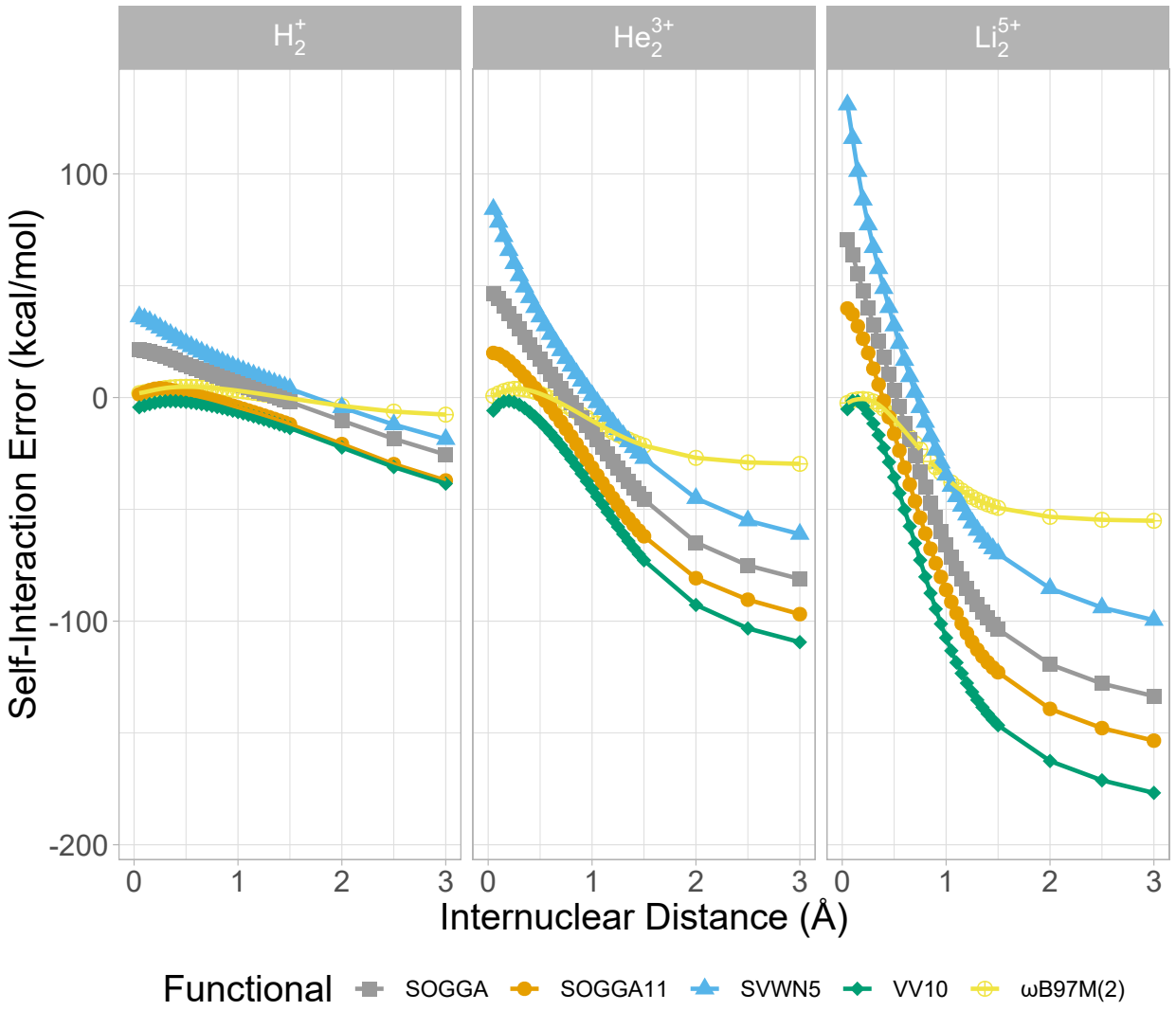

Figure S226: Self-interaction error vs internuclear distance of the homonuclear pairs, H_2^+, He_2^{3+}, Li_2^{5+}, for SOGGA, SOGGA11, SVWN5, VV10 with ωB97M(2) as a reference.

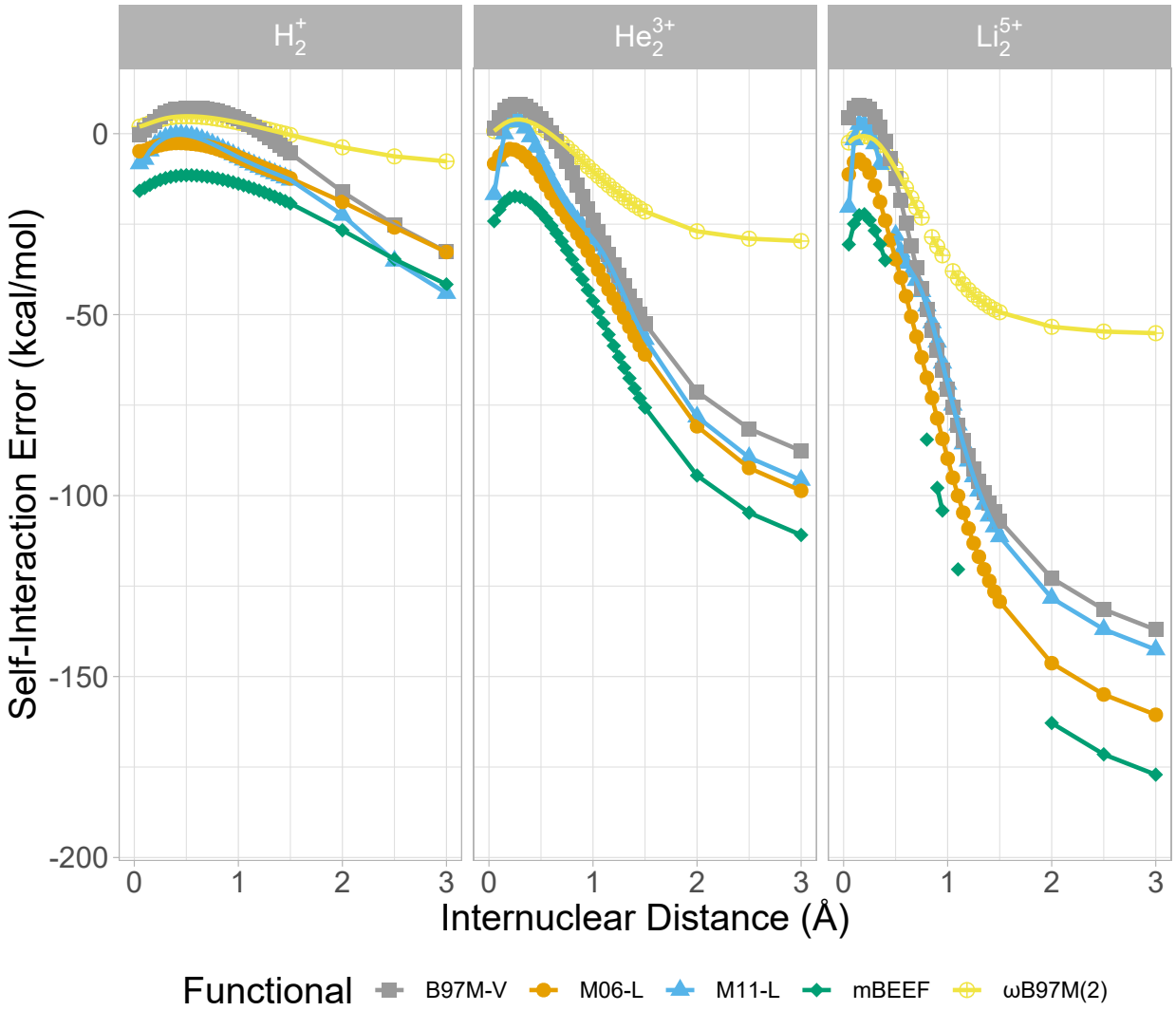

Figure S227: Self-interaction error vs internuclear distance of the homonuclear pairs, H_2^+, He_2^{3+}, Li_2^{5+}, for B97M-V, M06-L, M11-L, mBEEF, MN12-L with ωB97M(2) as a reference.

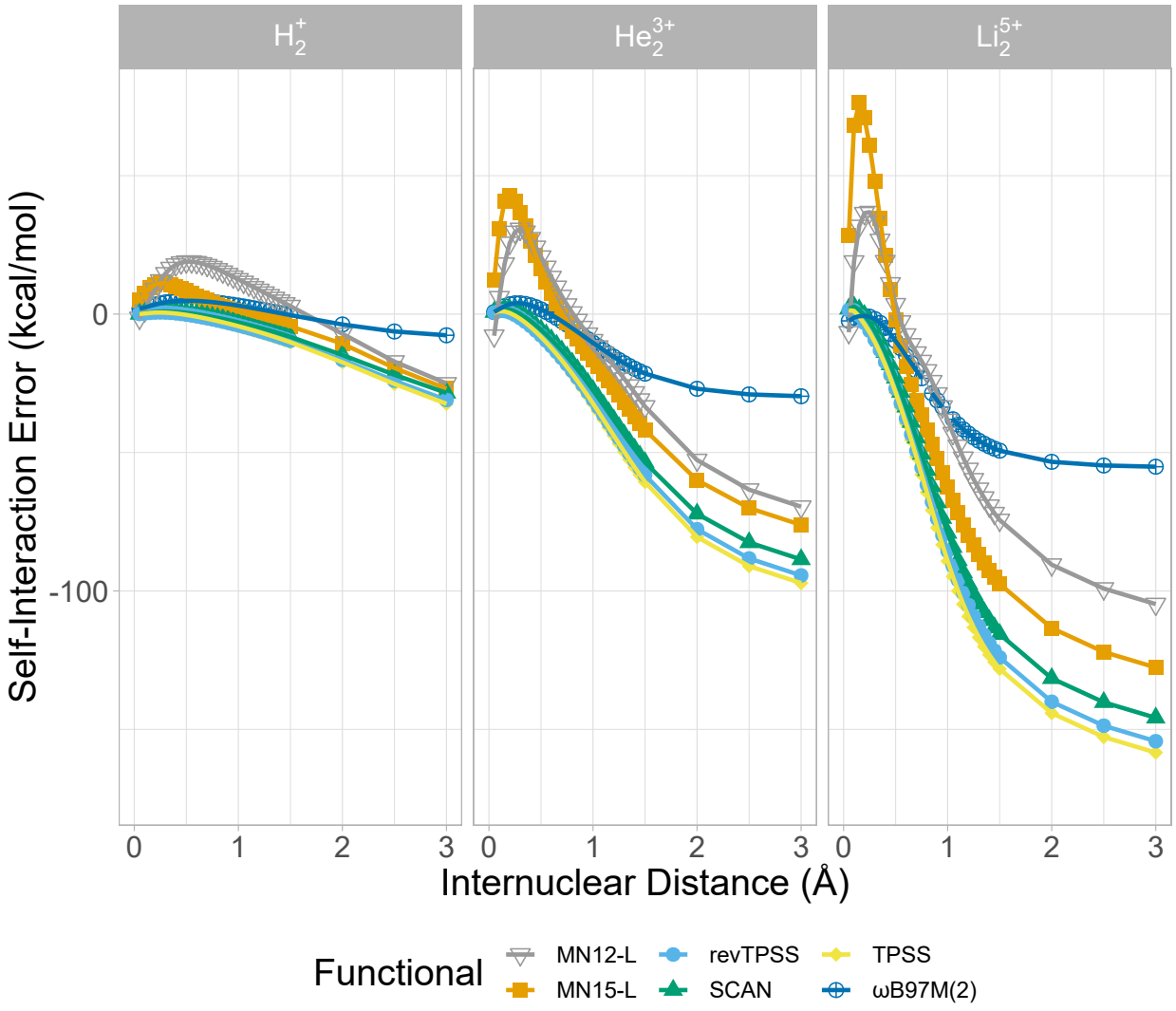

Figure S228: Self-interaction error vs internuclear distance of the homonuclear pairs, H_2^+, He_2^{3+}, Li_2^{5+}, for MN15-L, revTPSS, SCAN, TPSS with ωB97M(2) as a reference.

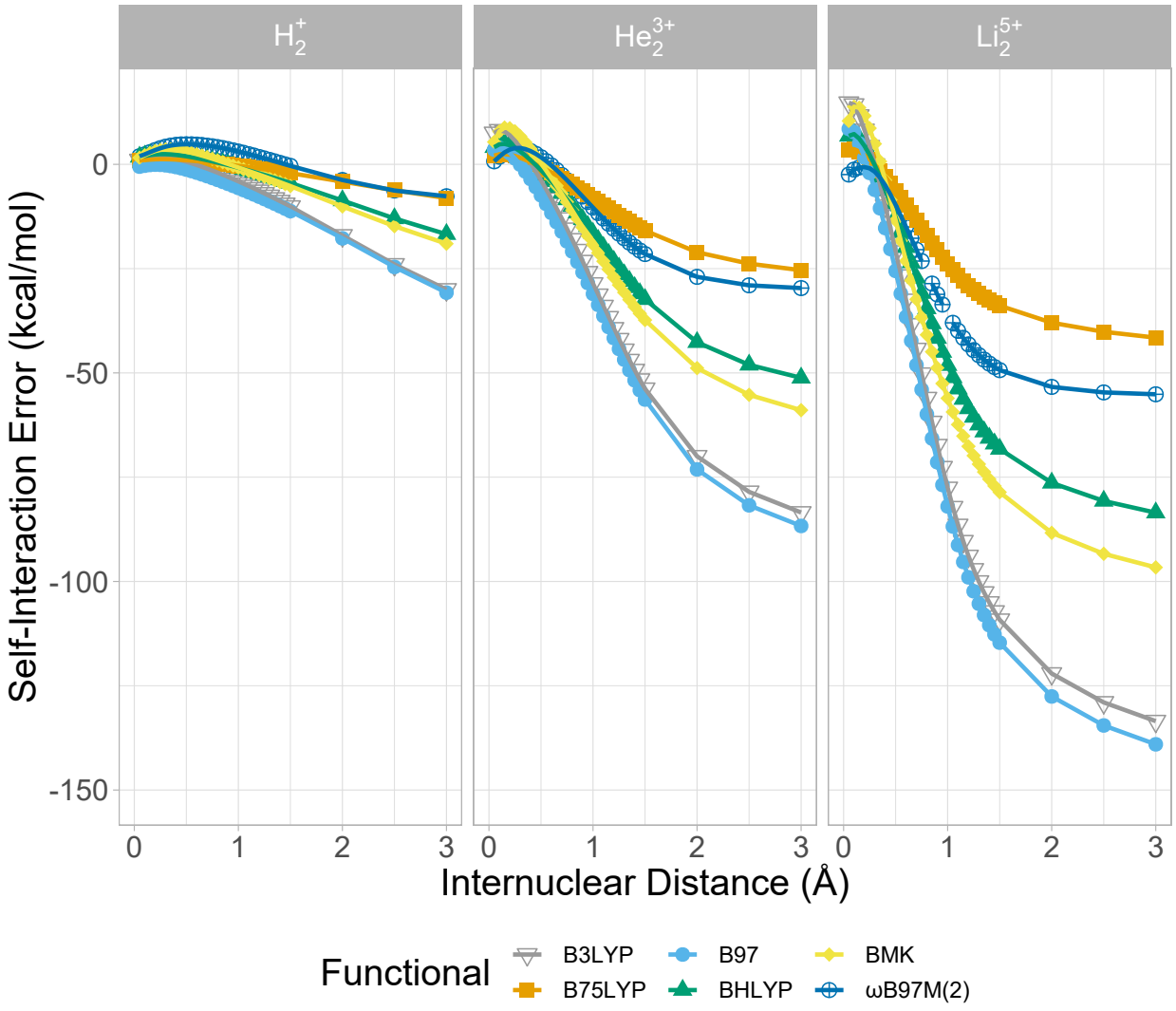

Figure S229: Self-interaction error vs internuclear distance of the homonuclear pairs, H_2^+, He_2^{3+}, Li_2^{5+}, for B3LYP, B75LYP, B97, BHLYP, BMK with ωB97M(2) as a reference.

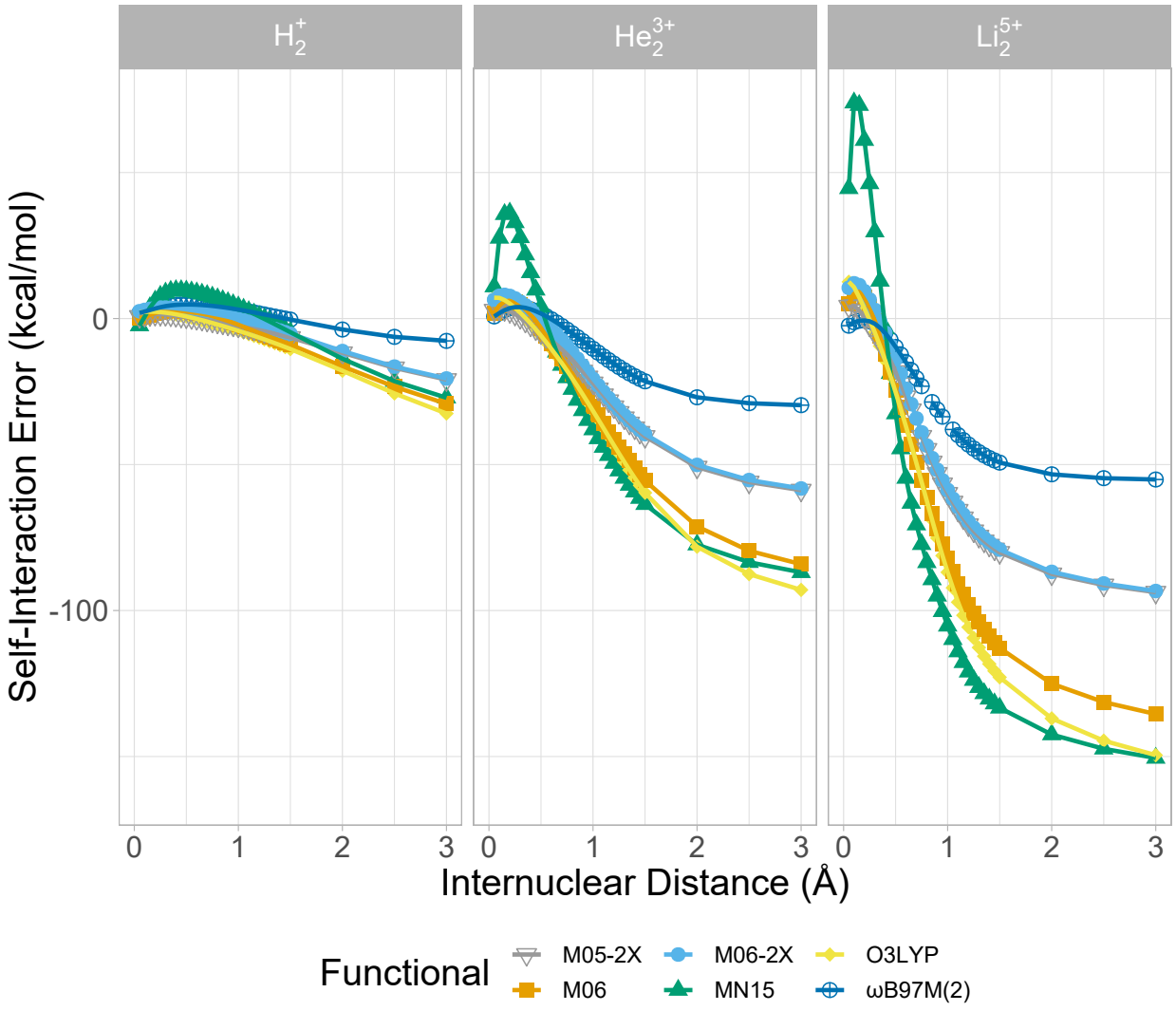

Figure S230: Self-interaction error vs internuclear distance of the homonuclear pairs, H_2^+, He_2^{3+}, Li_2^{5+}, for M05-2X, M06, M06-2X, MN15, O3LYP with ωB97M(2) as a reference.

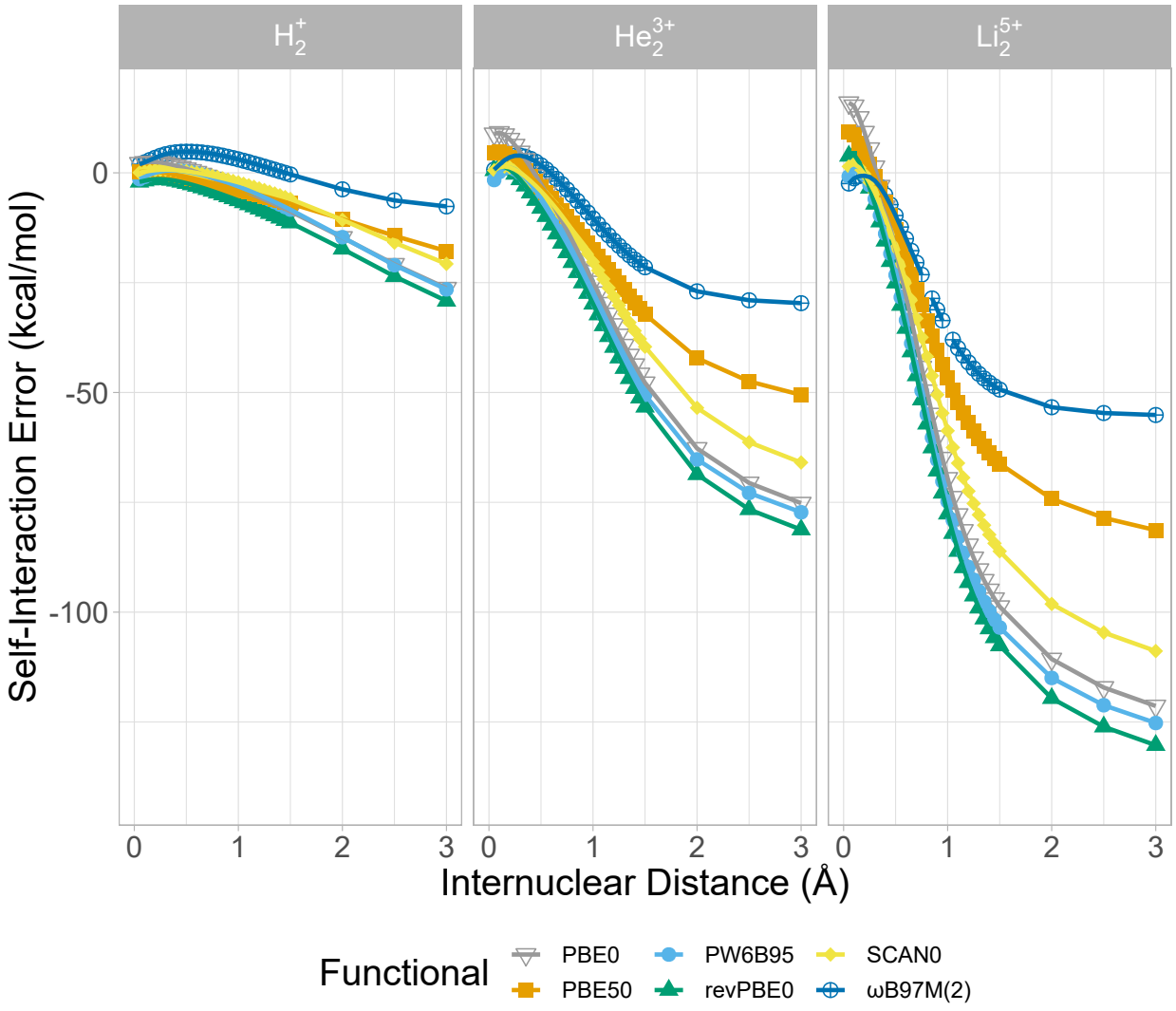

Figure S231: Self-interaction error vs internuclear distance of the homonuclear pairs, H_2^+, He_2^{3+}, Li_2^{5+}, for PBE0, PBE50, PW6B95, revPBE0, SCAN0 with ωB97M(2) as a reference.

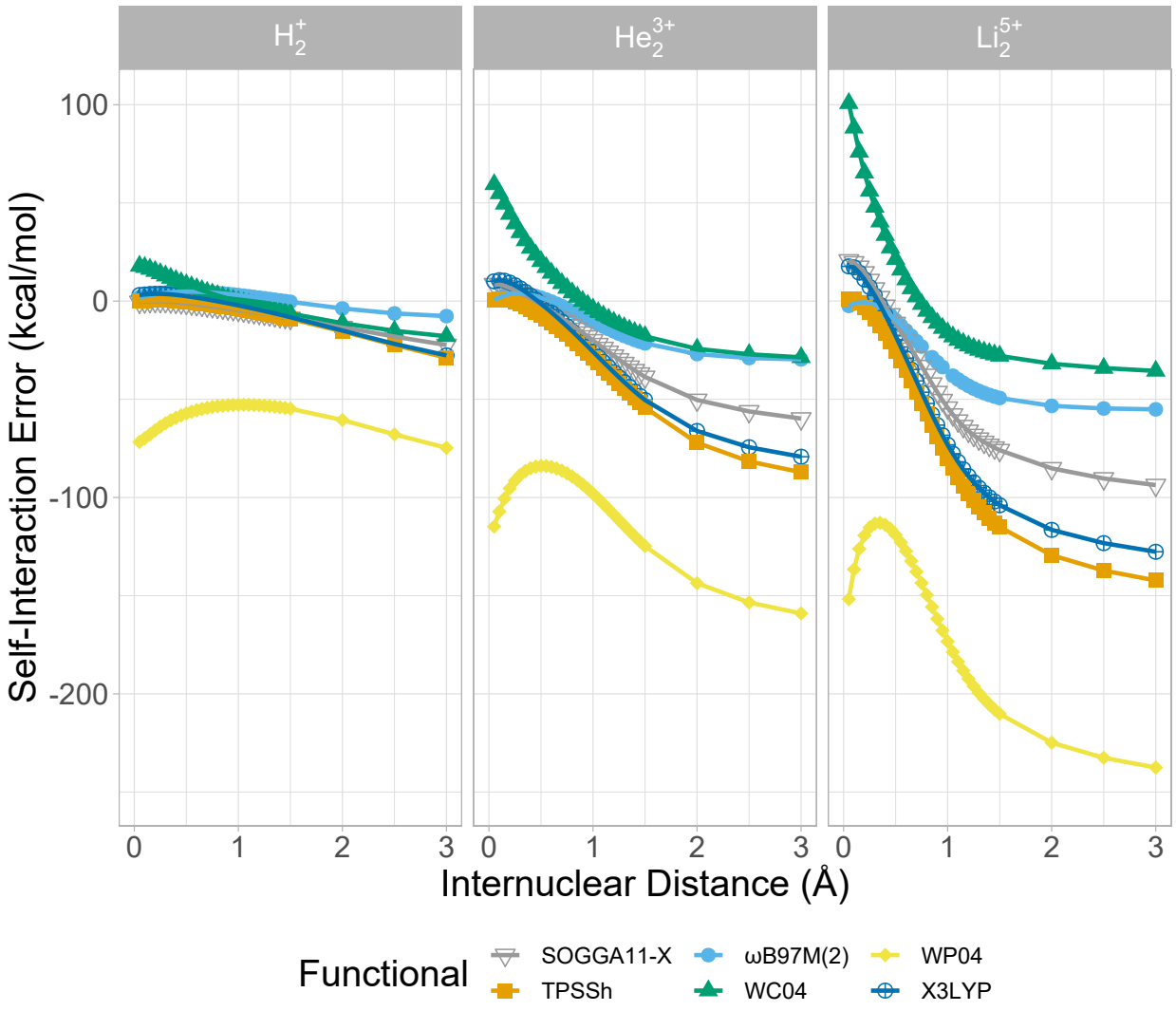

Figure S232: Self-interaction error vs internuclear distance of the homonuclear pairs, H_2^+, He_2^{3+}, Li_2^{5+}, for SOGGA11-X, TPSSh, WC04, WP04, X3LYP with ωB97M(2) as a reference.

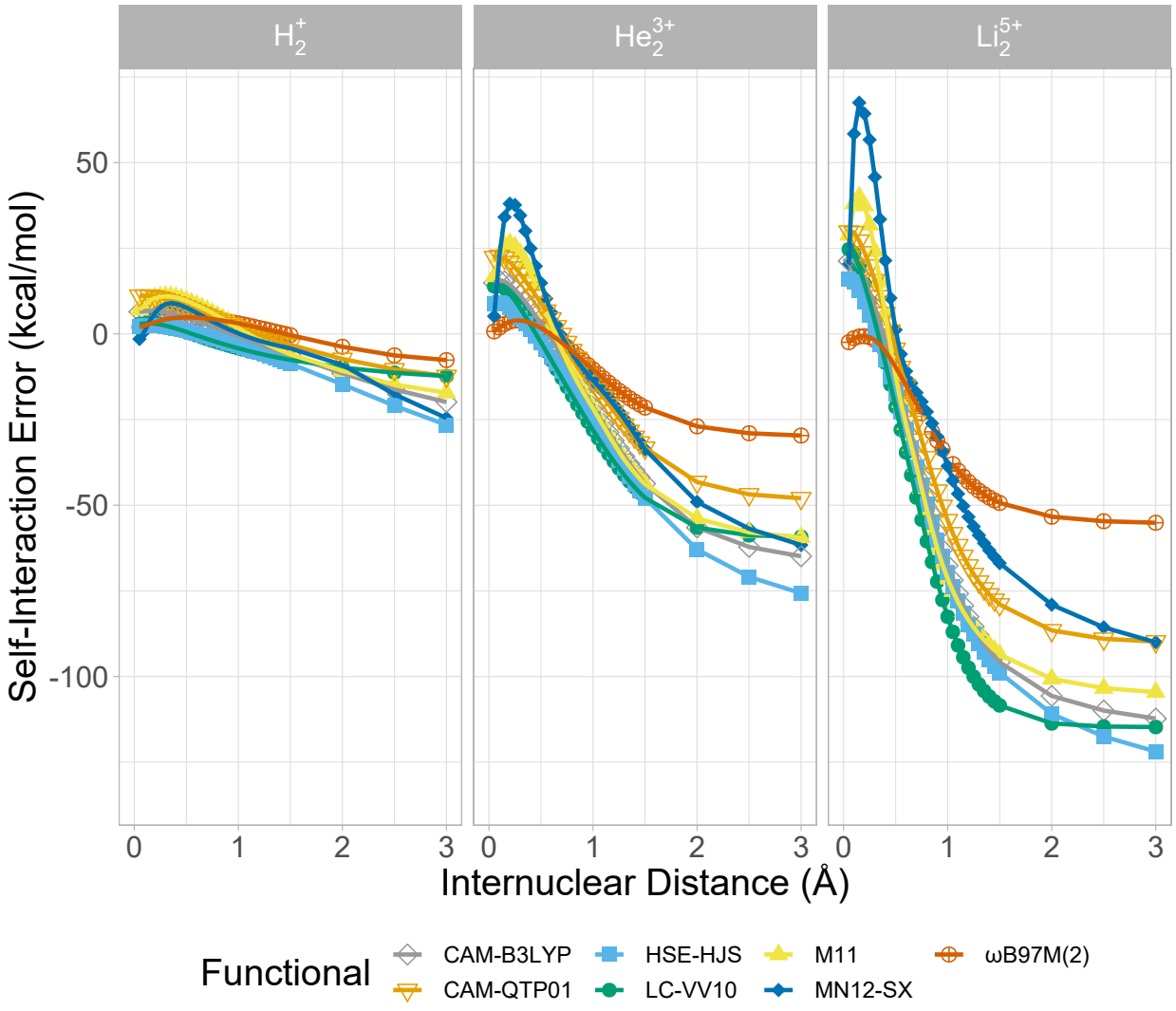

Figure S233: Self-interaction error vs internuclear distance of the homonuclear pairs, H_2^+, He_2^{3+}, Li_2^{5+}, for CAM-B3LYP, CAM-QTP01, HSE-HJS, LC-VV10, M11, MN12-SX with ωB97M(2) as a reference.

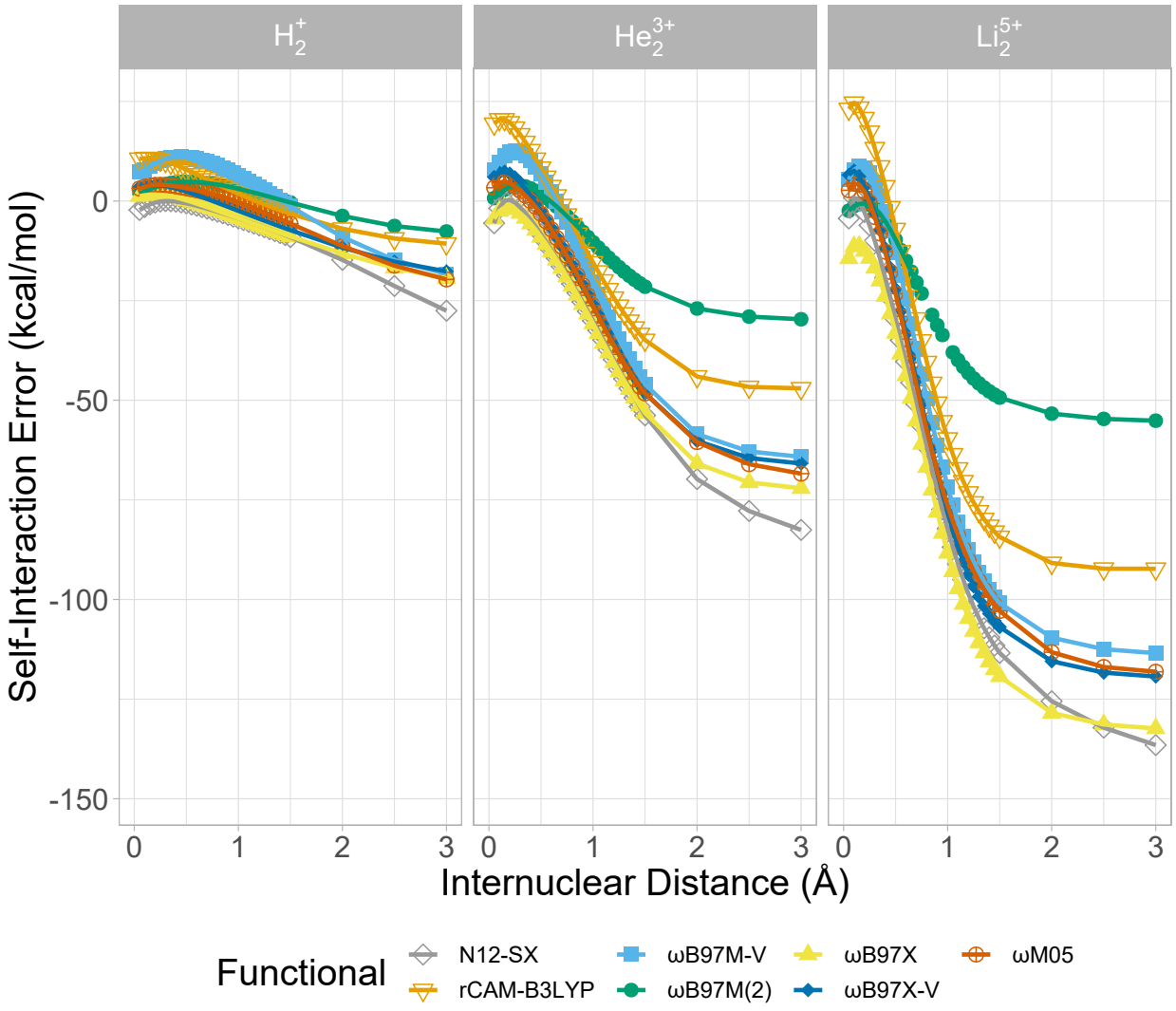

Figure S234: Self-interaction error vs internuclear distance of the homonuclear pairs, H_2^+, He_2^{3+}, Li_2^{5+}, for N12-SX, rCAM-B3LYP, ωB97M-V, ωB97X, ωB97X-V, ωM05 with ωB97M(2) as a reference.

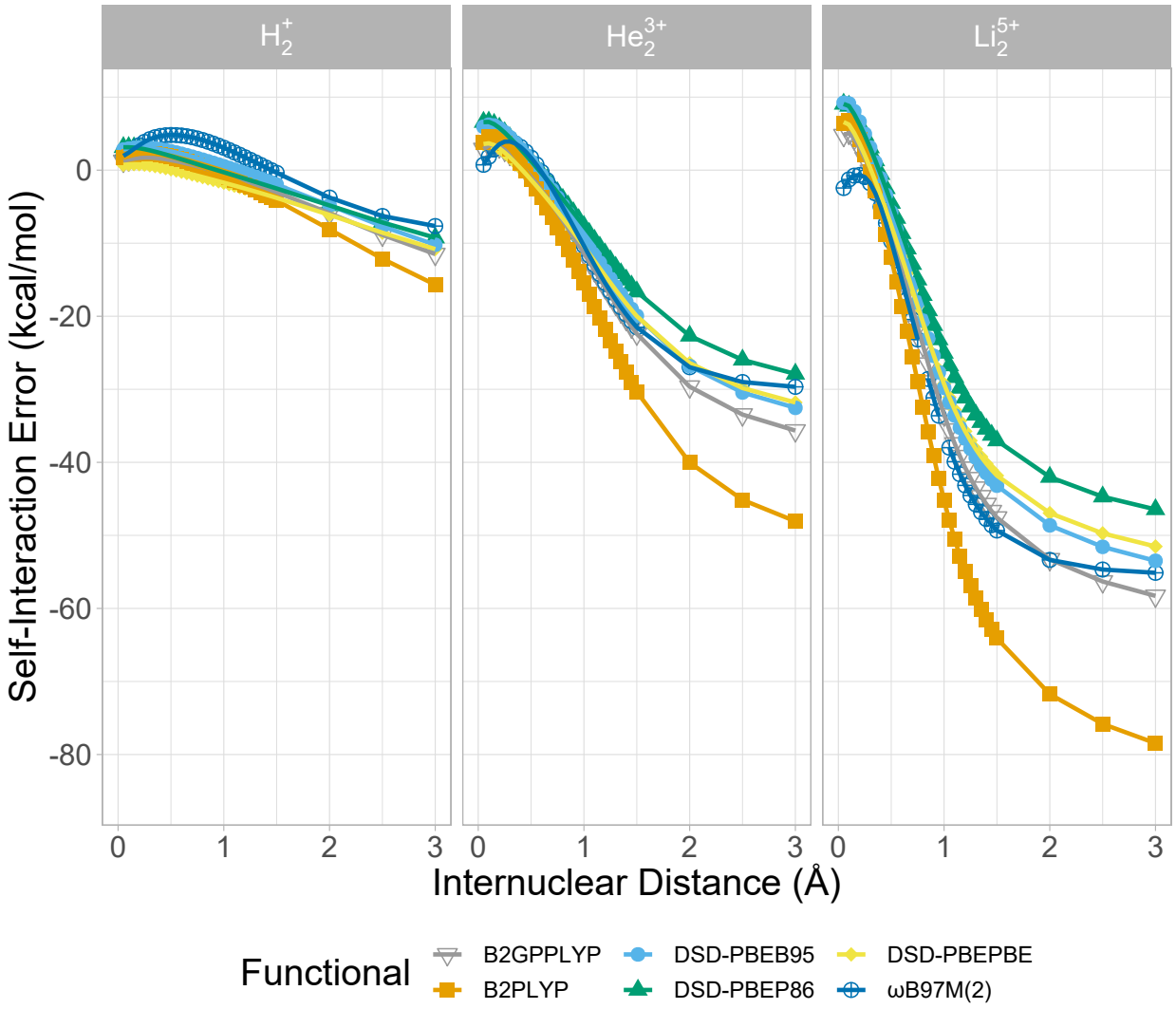

Figure S235: Self-interaction error vs internuclear distance of the homonuclear pairs, H_2^+, He_2^{3+}, Li_2^{5+}, for B2GPPLYP, B2PLYP, DSD-PBEB95, DSD-PBEP86, DSD-PBEPBE with ωB97M(2) as a reference.

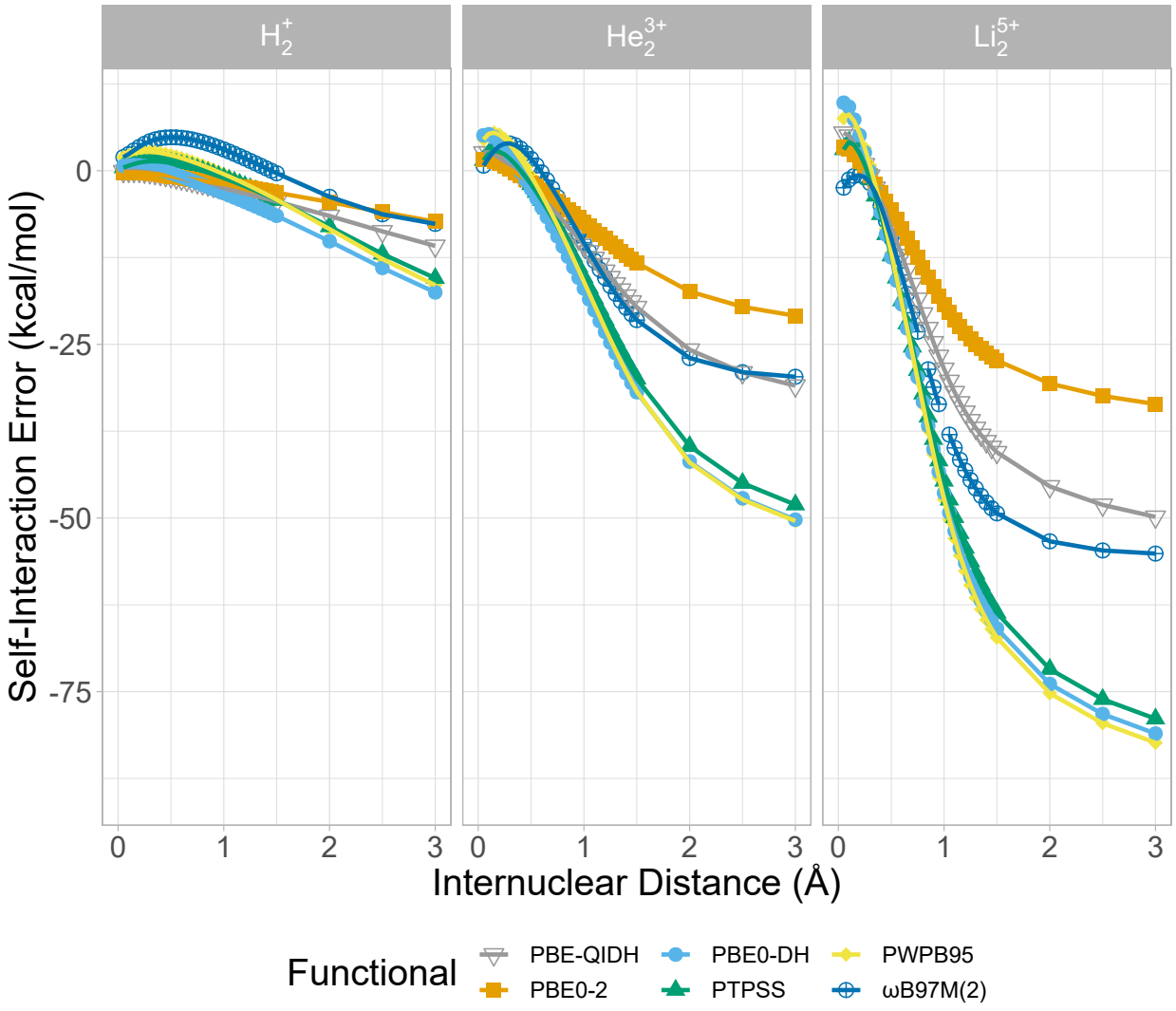

Figure S236: Self-interaction error vs internuclear distance of the homonuclear pairs, H_2^+, He_2^{3+}, Li_2^{5+}, for PBE-QIDH, PBE0-2, PBE0-DH, PTPSS, PWPB95 with ωB97M(2) as a reference.

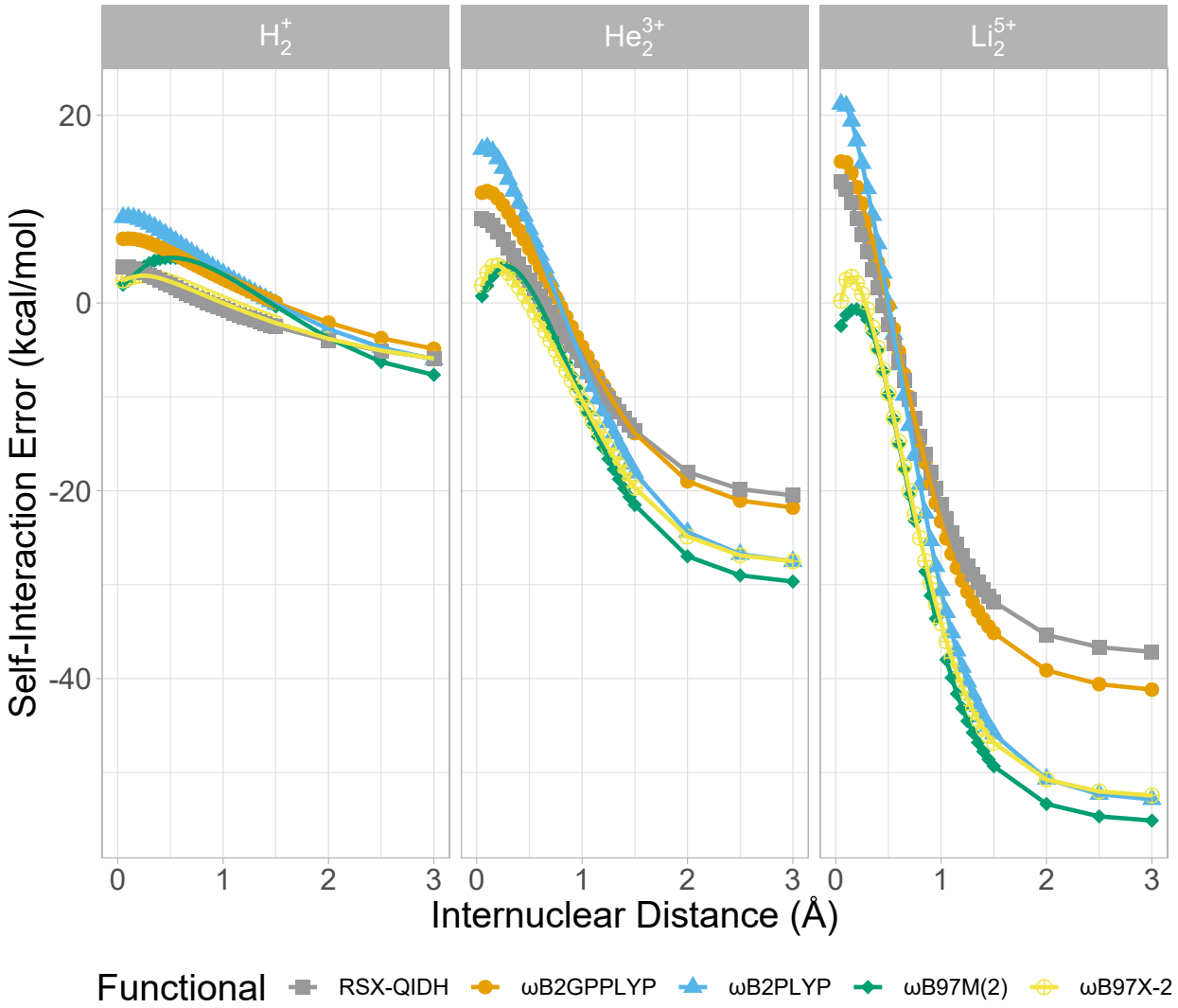

Figure S237: Self-interaction error vs internuclear distance of the homonuclear pairs, H_2^+, He_2^{3+}, Li_2^{5+}, for RSX-QIDH, ωB2GPPLYP, ωB2PLYP, ωB97X-2 with ωB97M(2) as a reference.

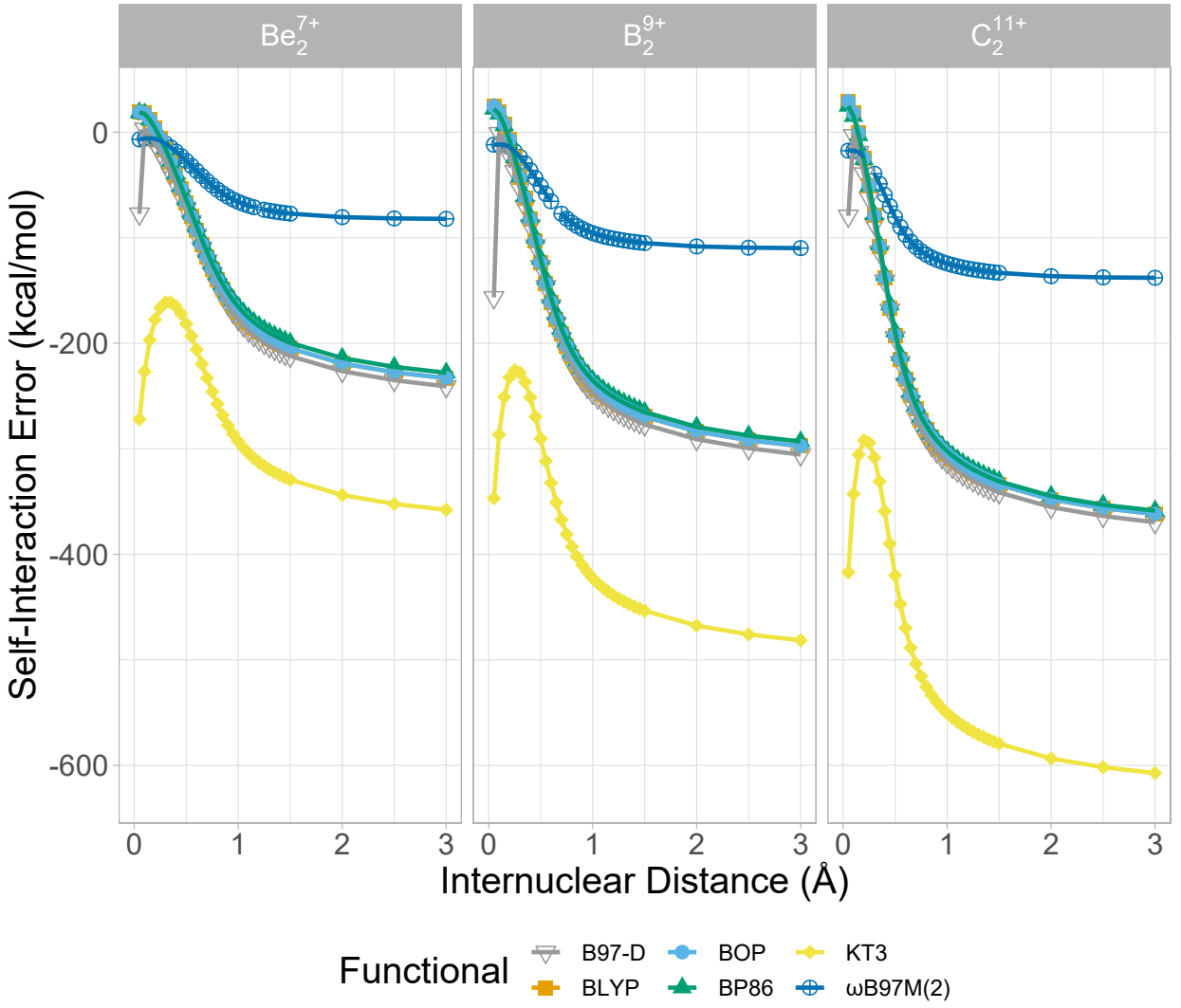

Figure S238: Self-interaction error vs internuclear distance of the homonuclear pairs, Be_2^{7+} B_2^{9+}, C_2^{11+}, for B97-D, BLYP, BOP, BP86, KT3 with ωB97M(2) as a reference.

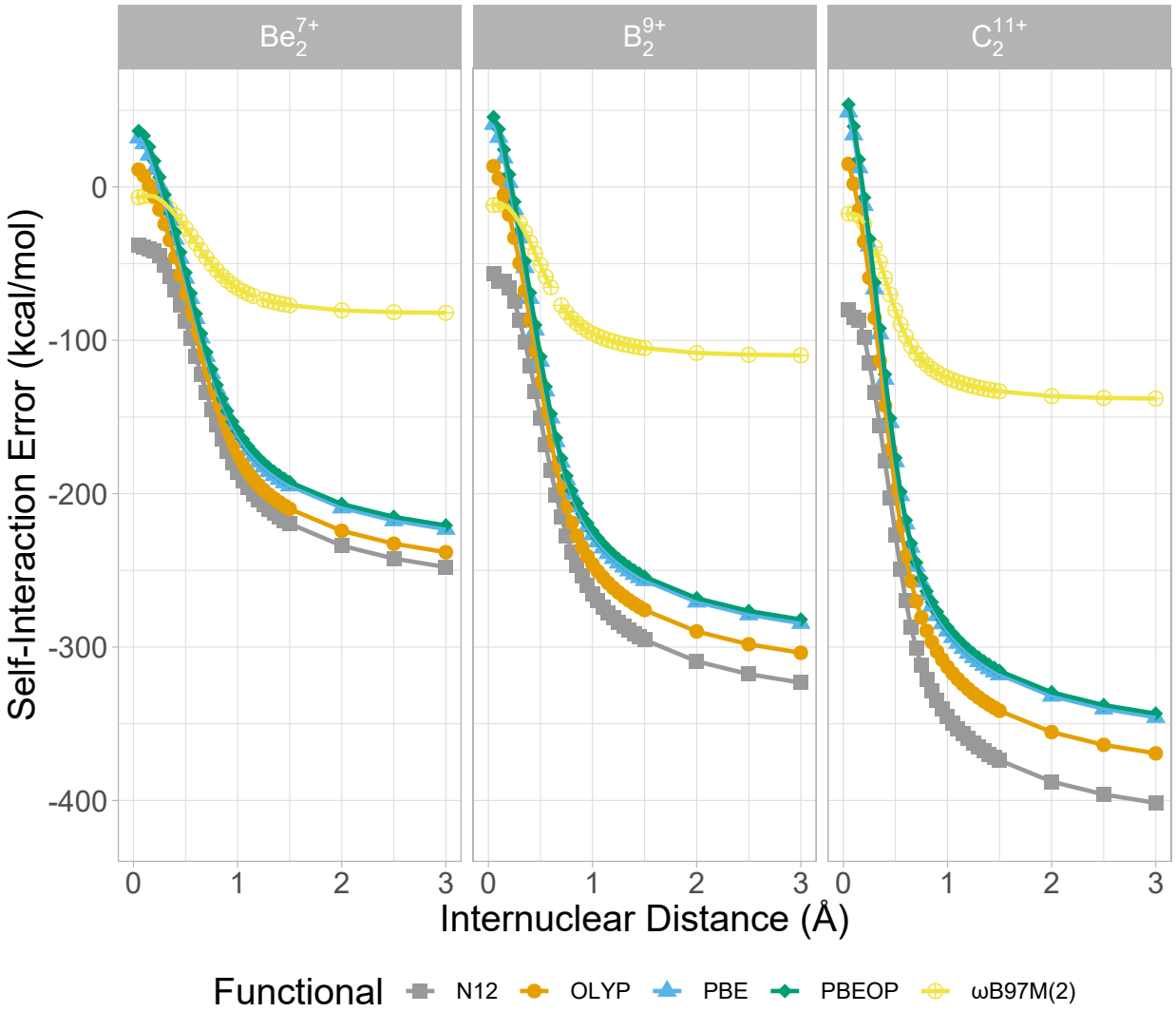

Figure S239: Self-interaction error vs internuclear distance of the homonuclear pairs, Be_2^{7+} B_2^{9+}, C_2^{11+}, for N12, OLYP, PBE, PBEOP with ωB97M(2) as a reference.

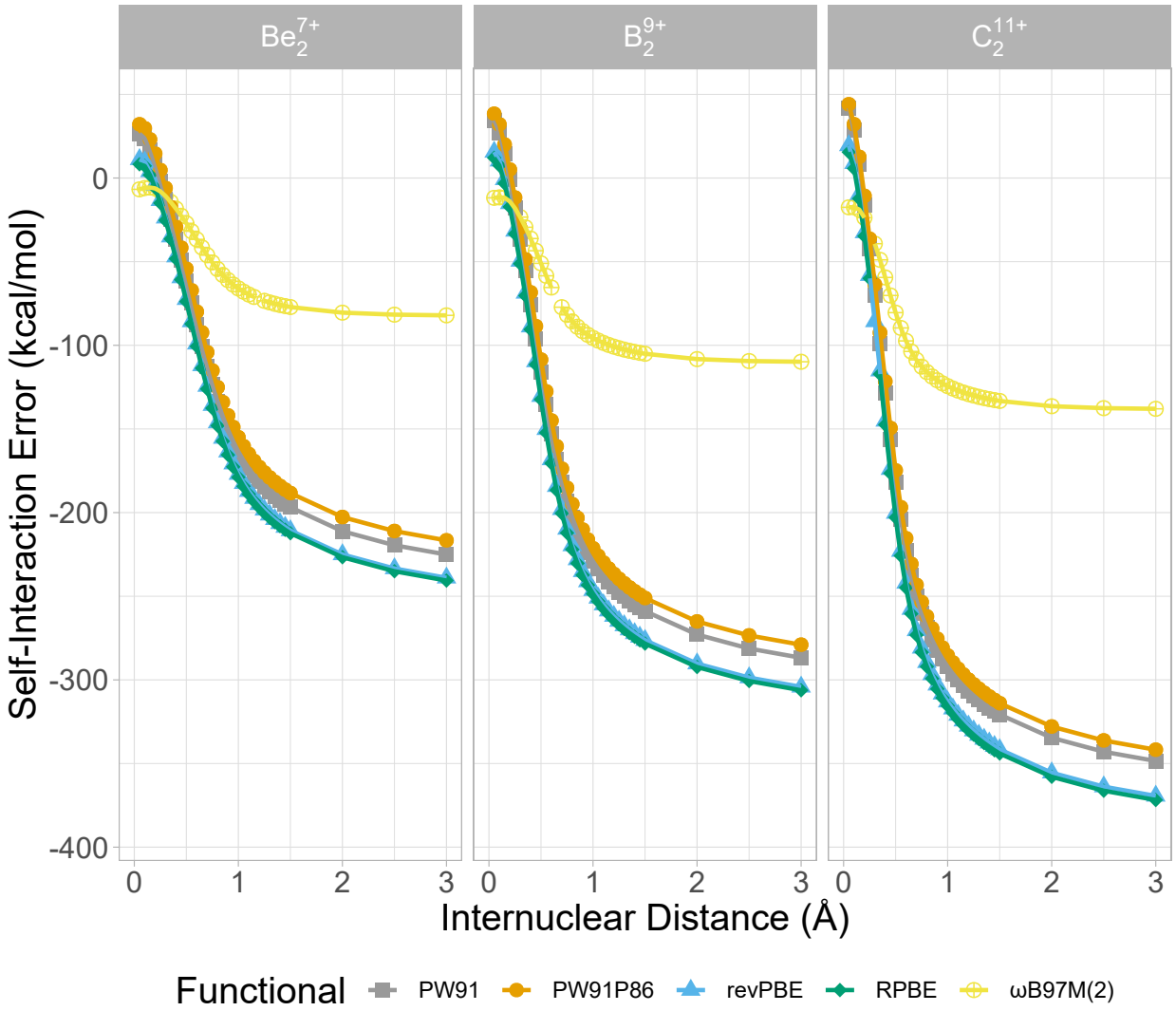

Figure S240: Self-interaction error vs internuclear distance of the homonuclear pairs, Be_2^{7+} B_2^{9+}, C_2^{11+}, for PW91, PW91P86, revPBE, RPBE with ωB97M(2) as a reference.

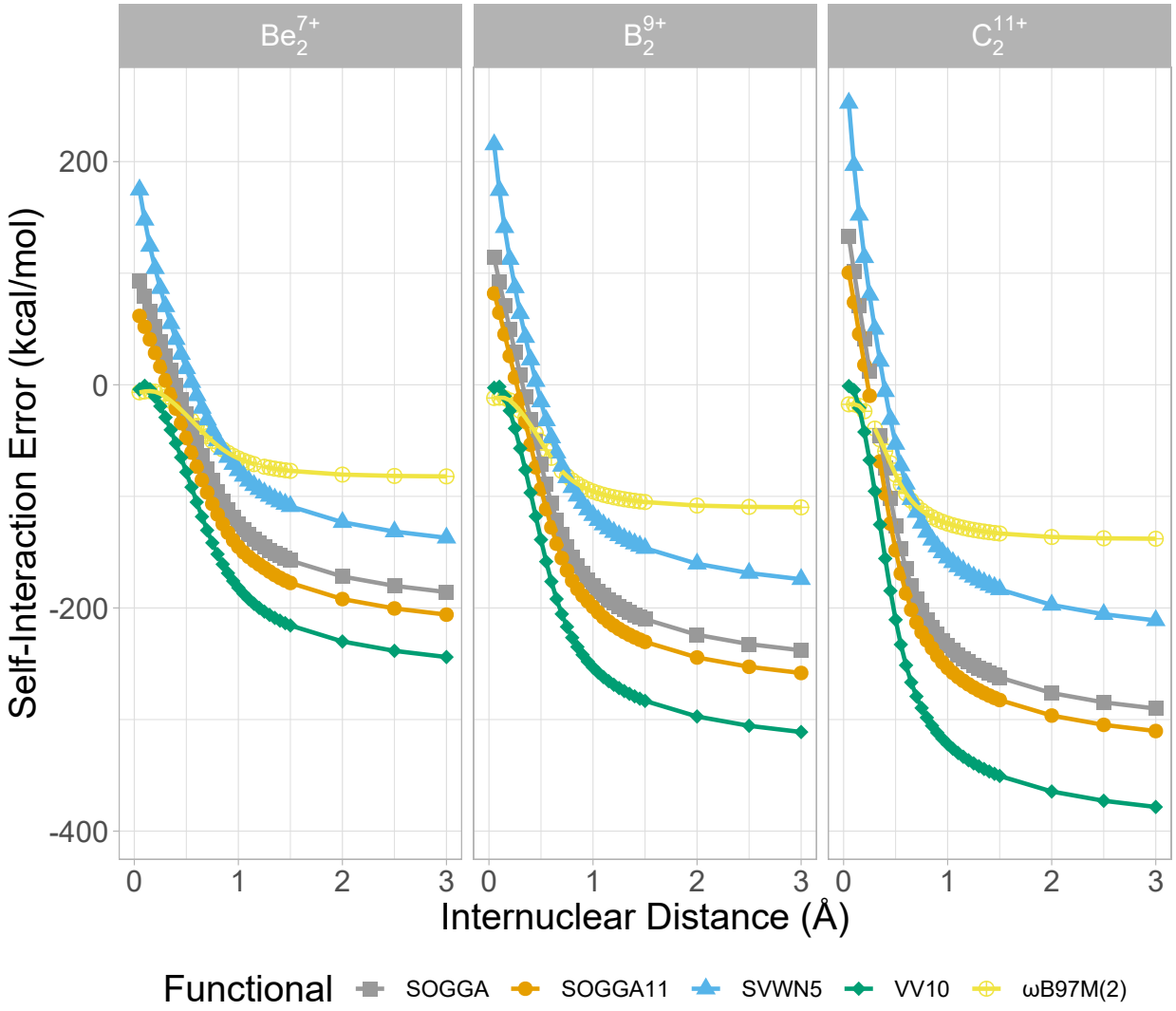

Figure S241: Self-interaction error vs internuclear distance of the homonuclear pairs, Be_2^{7+} B_2^{9+}, C_2^{11+}, for SOGGA, SOGGA11, SVWN5, VV10 with ωB97M(2) as a reference.

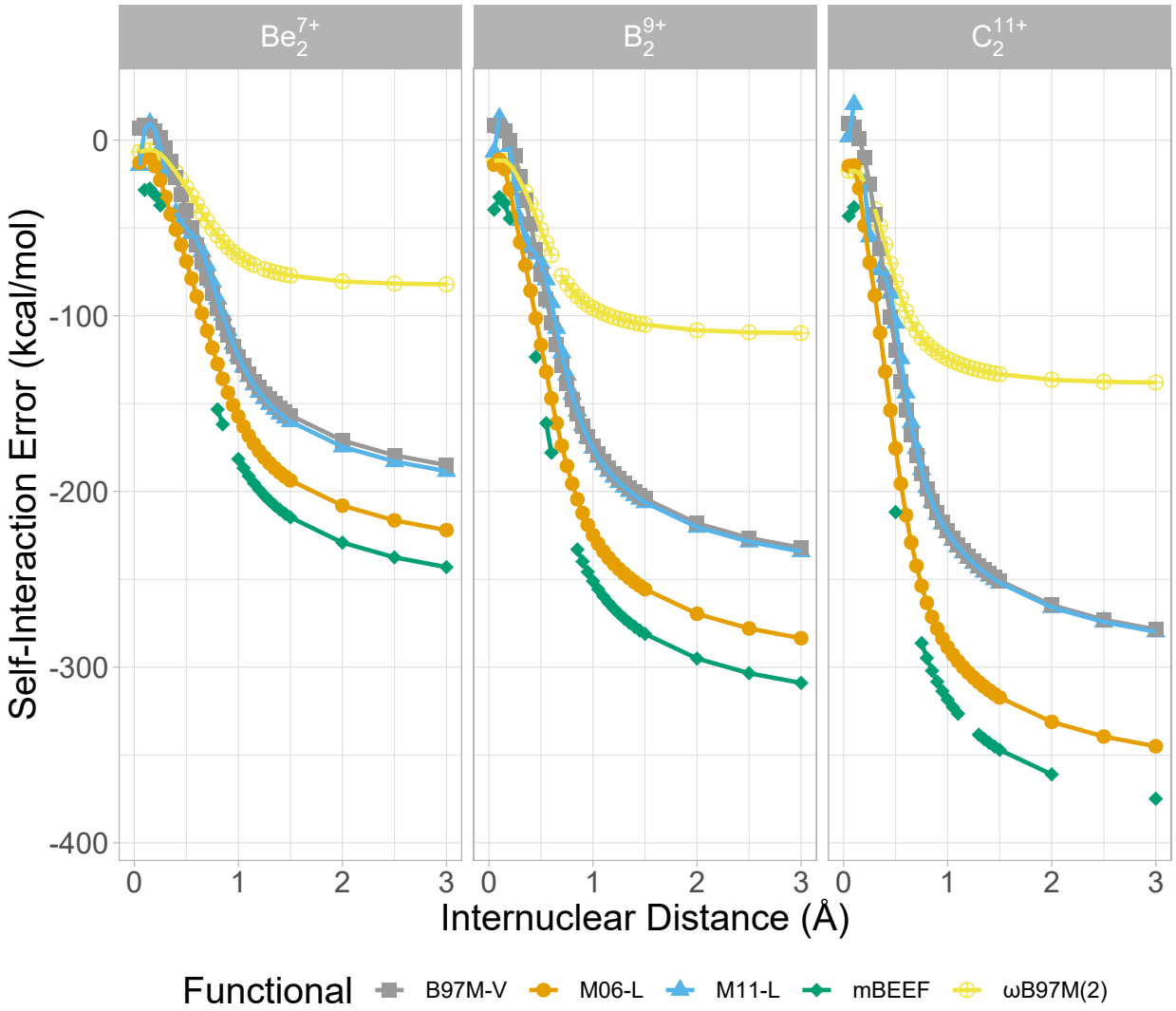

Figure S242: Self-interaction error vs internuclear distance of the homonuclear pairs, Be_2^{7+} B_2^{9+}, C_2^{11+}, for B97M-V, M06-L, M11-L, mBEEF, MN12-L with ωB97M(2) as a reference.

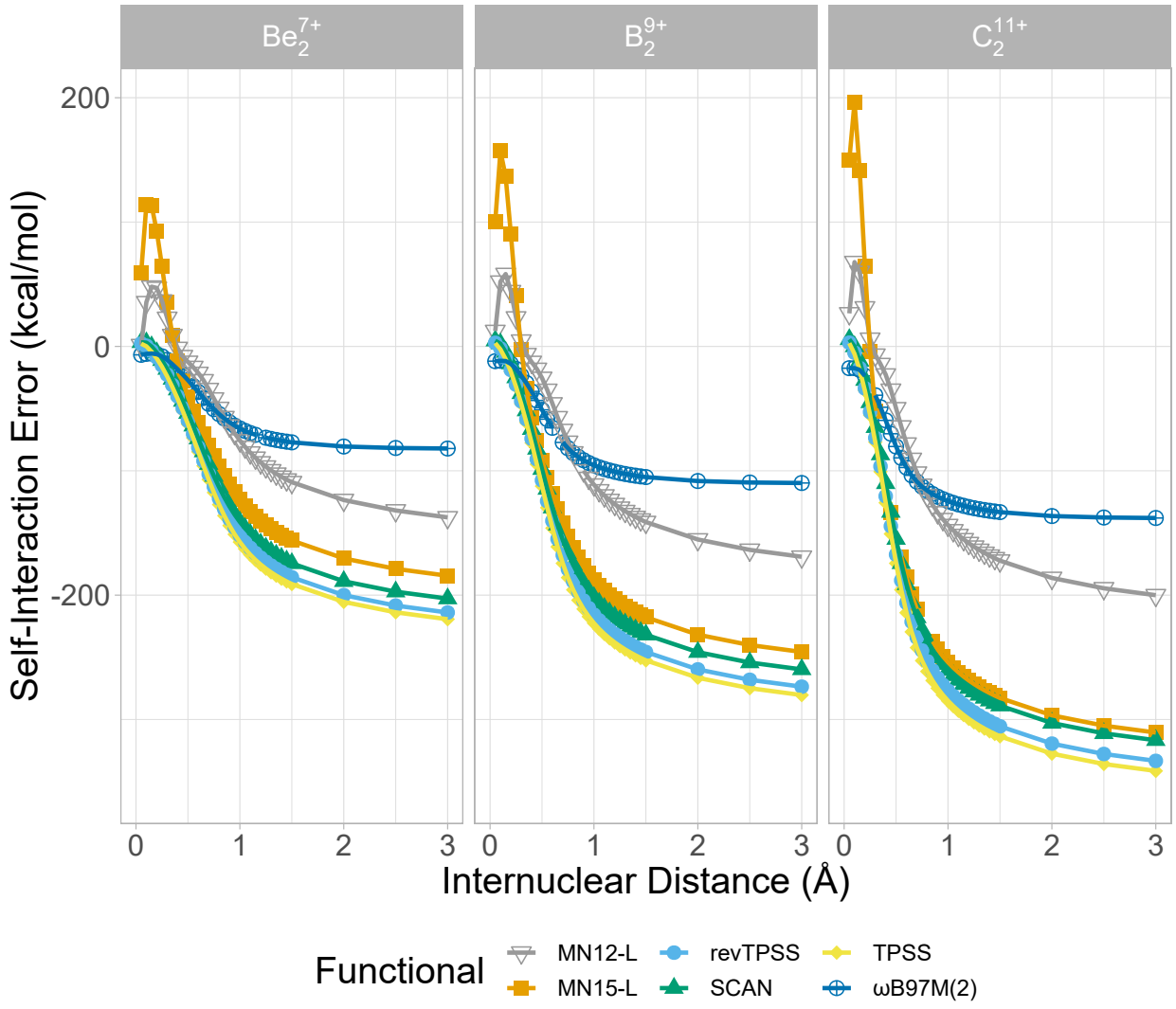

Figure S243: Self-interaction error vs internuclear distance of the homonuclear pairs, Be_2^{7+} B_2^{9+}, C_2^{11+}, for MN15-L, revTPSS, SCAN, TPSS with ωB97M(2) as a reference.

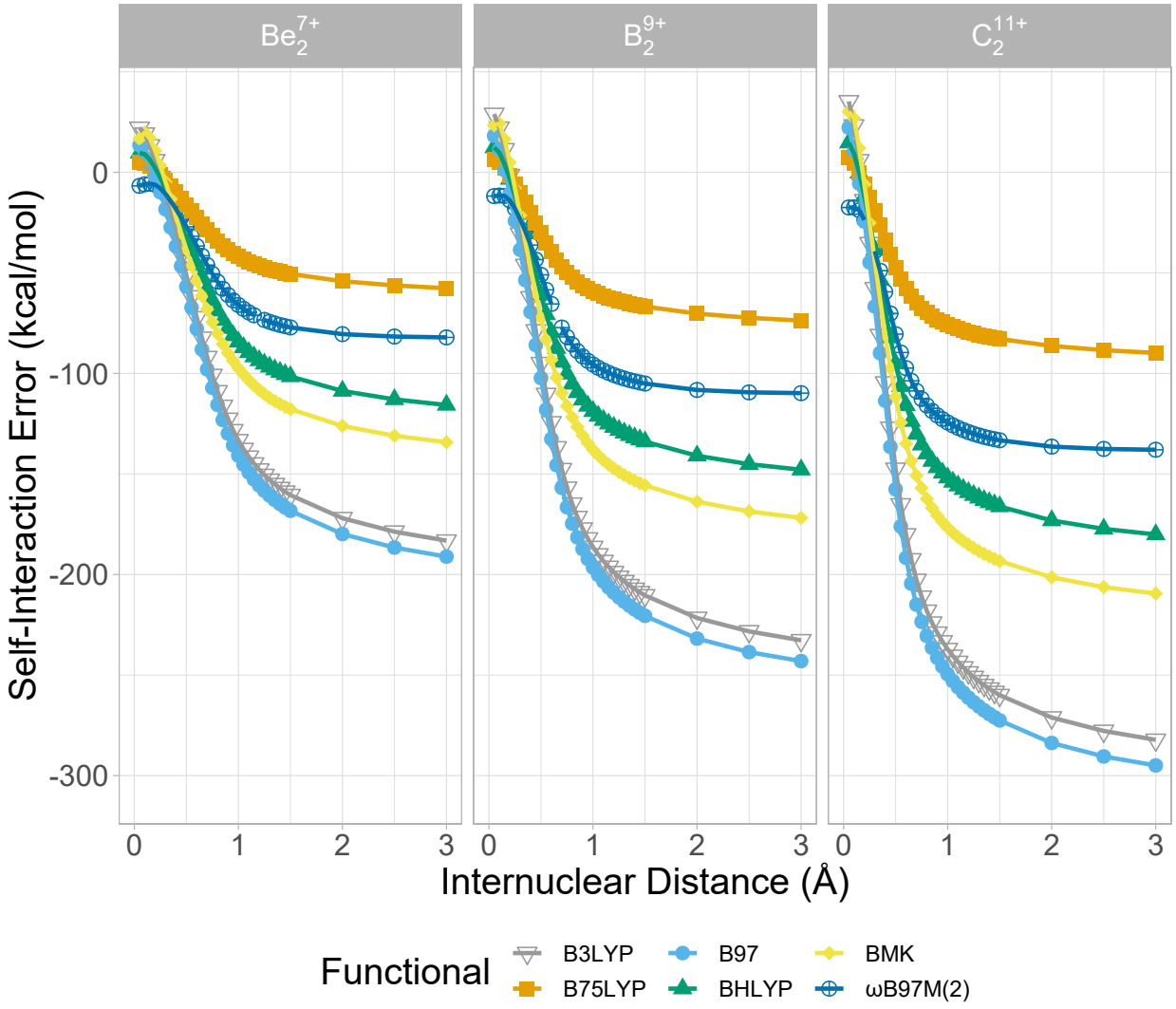

Figure S244: Self-interaction error vs internuclear distance of the homonuclear pairs, Be_2^{7+} B_2^{9+}, C_2^{11+}, for B3LYP, B75LYP, B97, BHLYP, BMK with ωB97M(2) as a reference.

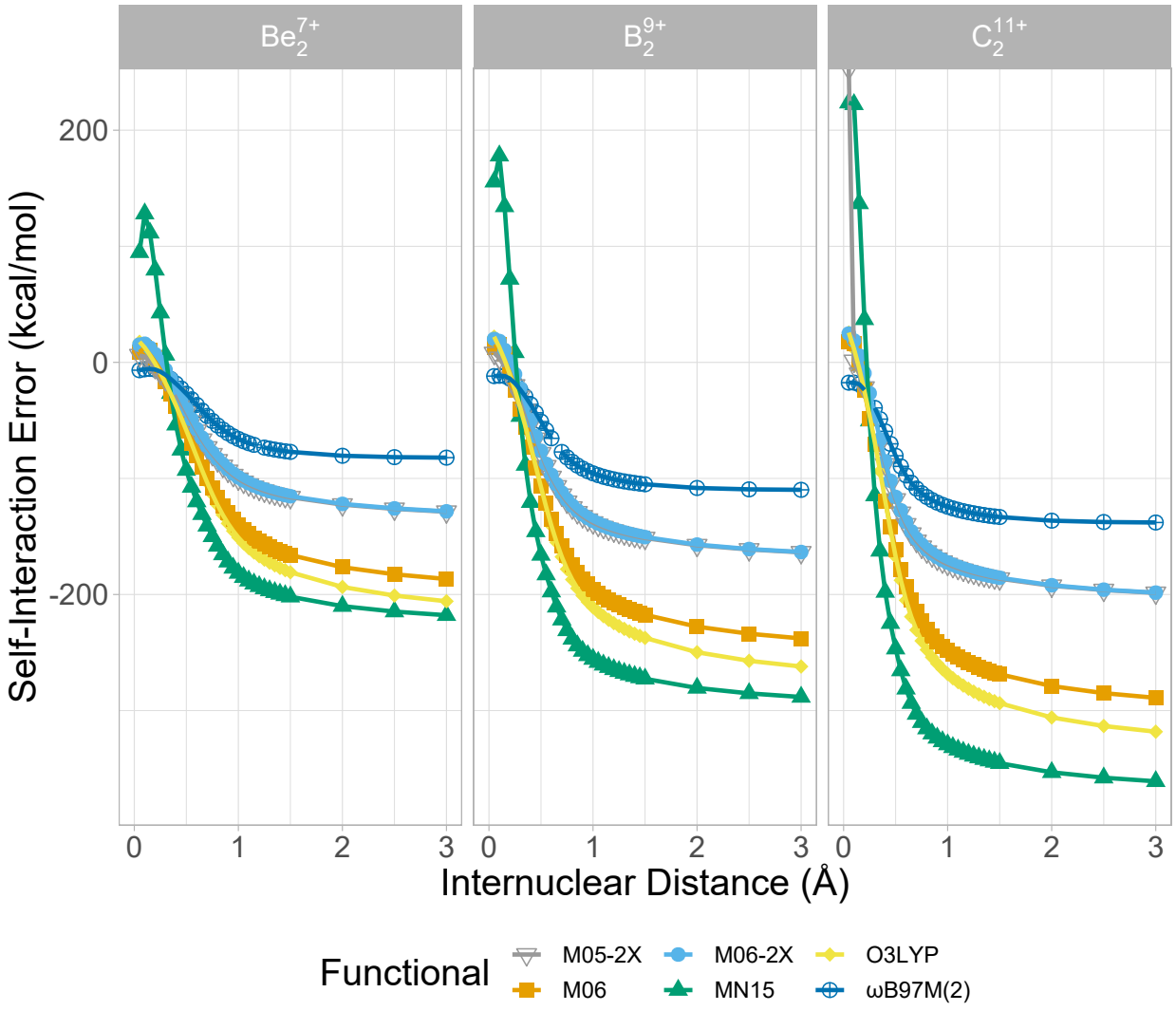

Figure S245: Self-interaction error vs internuclear distance of the homonuclear pairs, Be_2^{7+} B_2^{9+}, C_2^{11+}, for M05-2X, M06, M06-2X, MN15, O3LYP with ωB97M(2) as a reference.

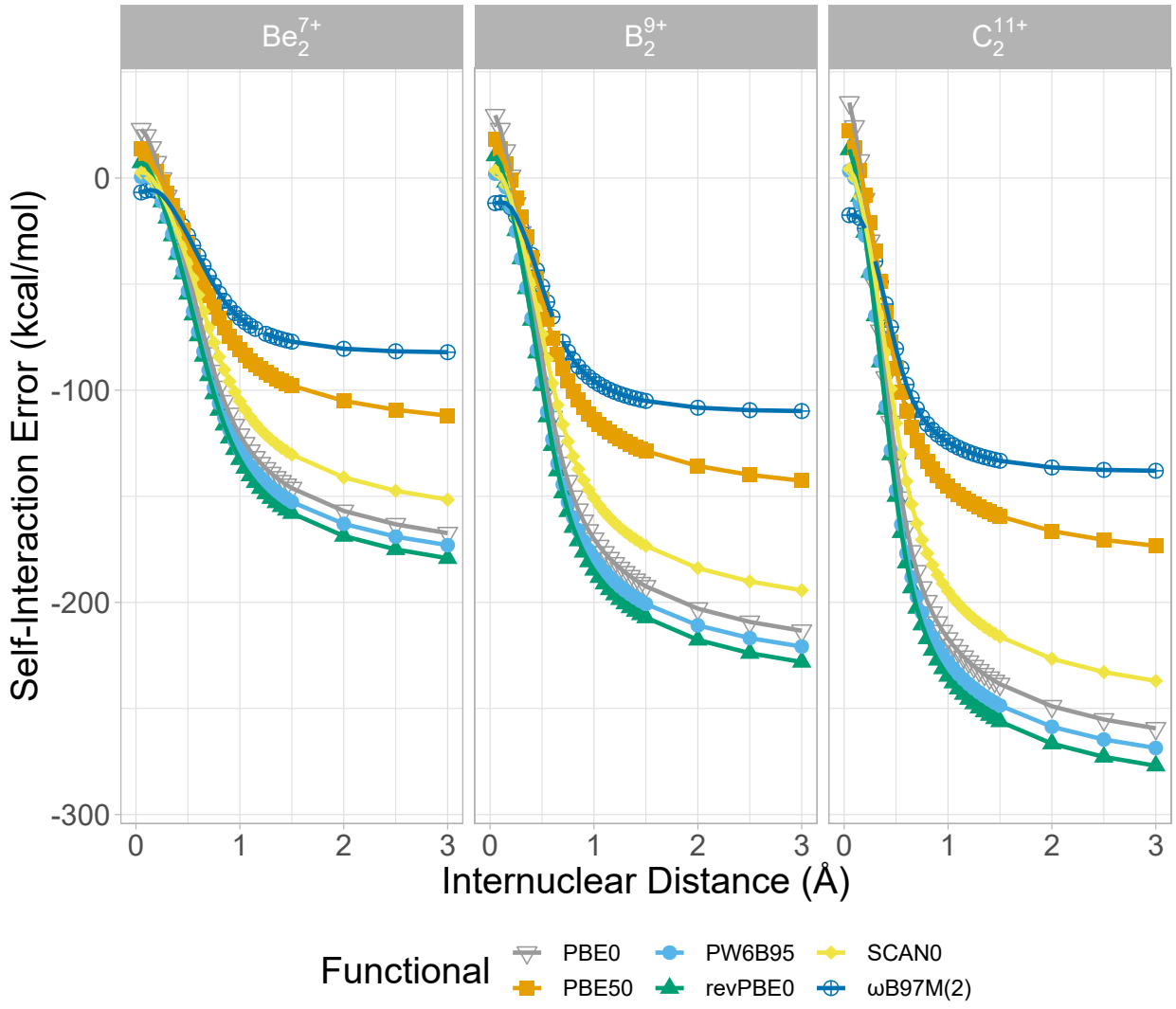

Figure S246: Self-interaction error vs internuclear distance of the homonuclear pairs, Be_2^{7+} B_2^{9+}, C_2^{11+}, for PBE0, PBE50, PW6B95, revPBE0, SCAN0 with ωB97M(2) as a reference.

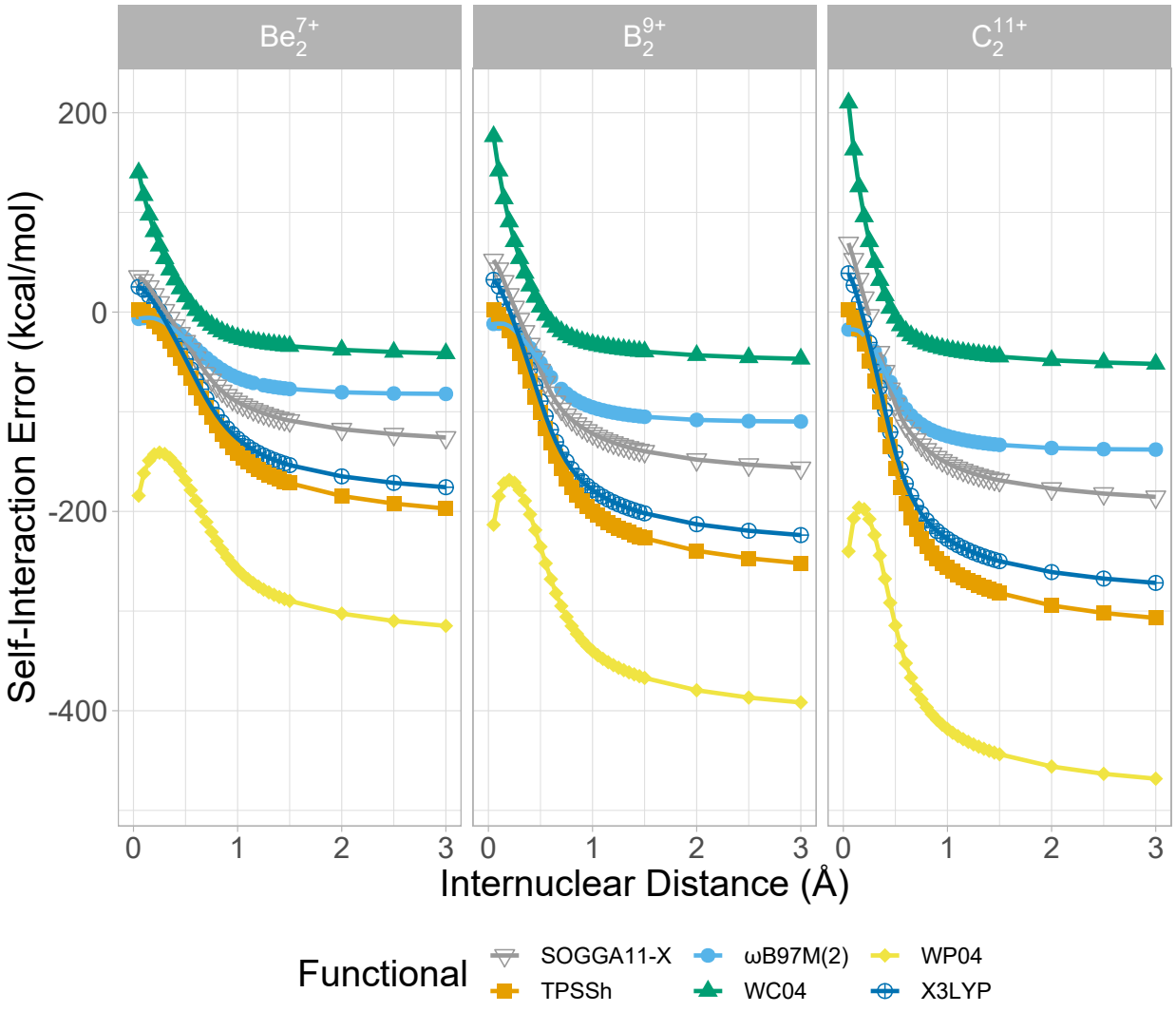

Figure S247: Self-interaction error vs internuclear distance of the homonuclear pairs, Be_2^{7+} B_2^{9+}, C_2^{11+}, for SOGGA11-X, TPSSh, WC04, WP04, X3LYP with ωB97M(2) as a reference.

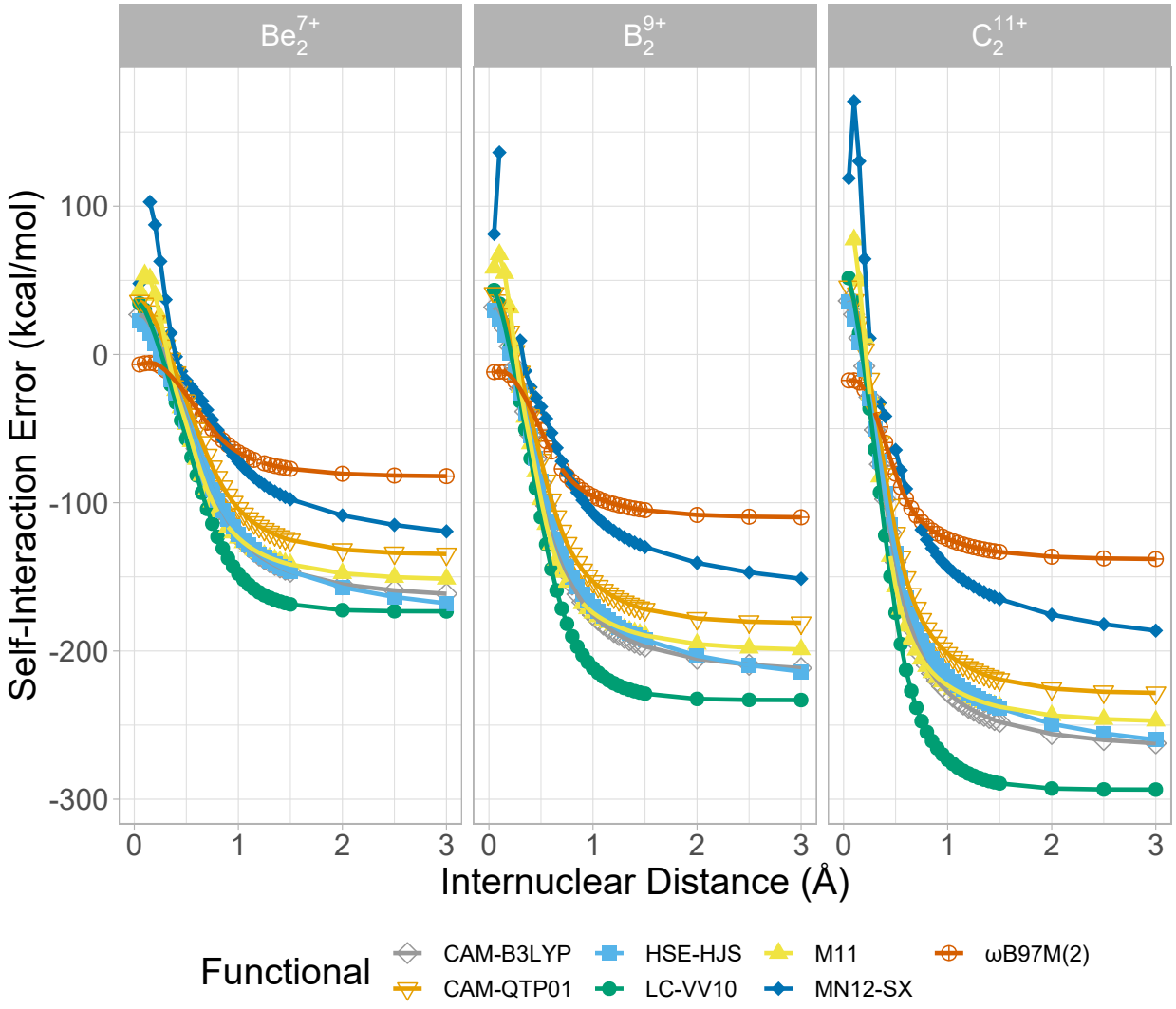

Figure S248: Self-interaction error vs internuclear distance of the homonuclear pairs, Be_2^{7+} B_2^{9+}, C_2^{11+}, for CAM-B3LYP, CAM-QTP01, HSE-HJS, LC-VV10, M11, MN12-SX with ωB97M(2) as a reference.

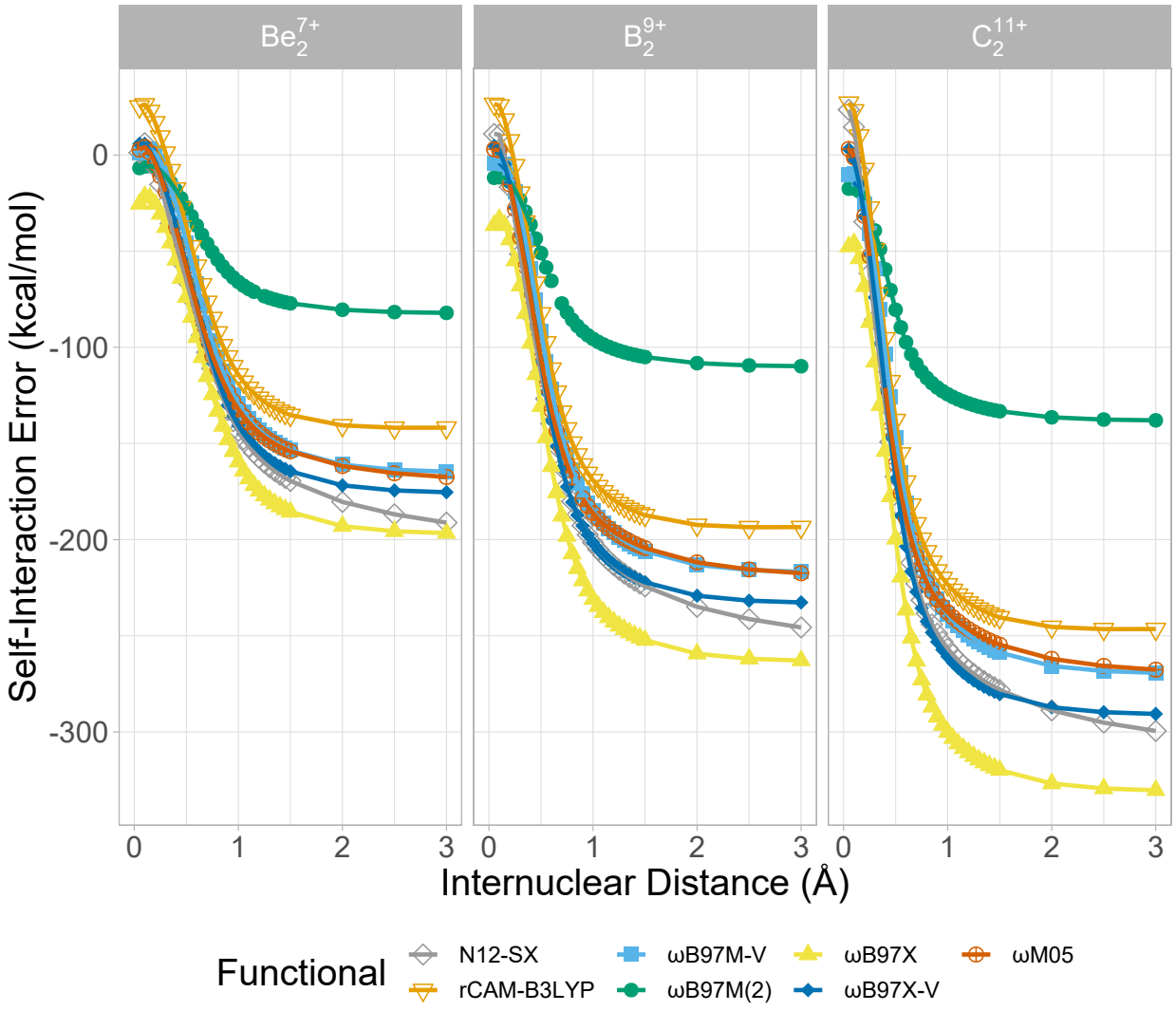

Figure S249: Self-interaction error vs internuclear distance of the homonuclear pairs, Be_2^{7+} B_2^{9+}, C_2^{11+}, for N12-SX, rCAM-B3LYP, ωB97M-V, ωB97X, ωB97X-V, ωM05 with ωB97M(2) as a reference.

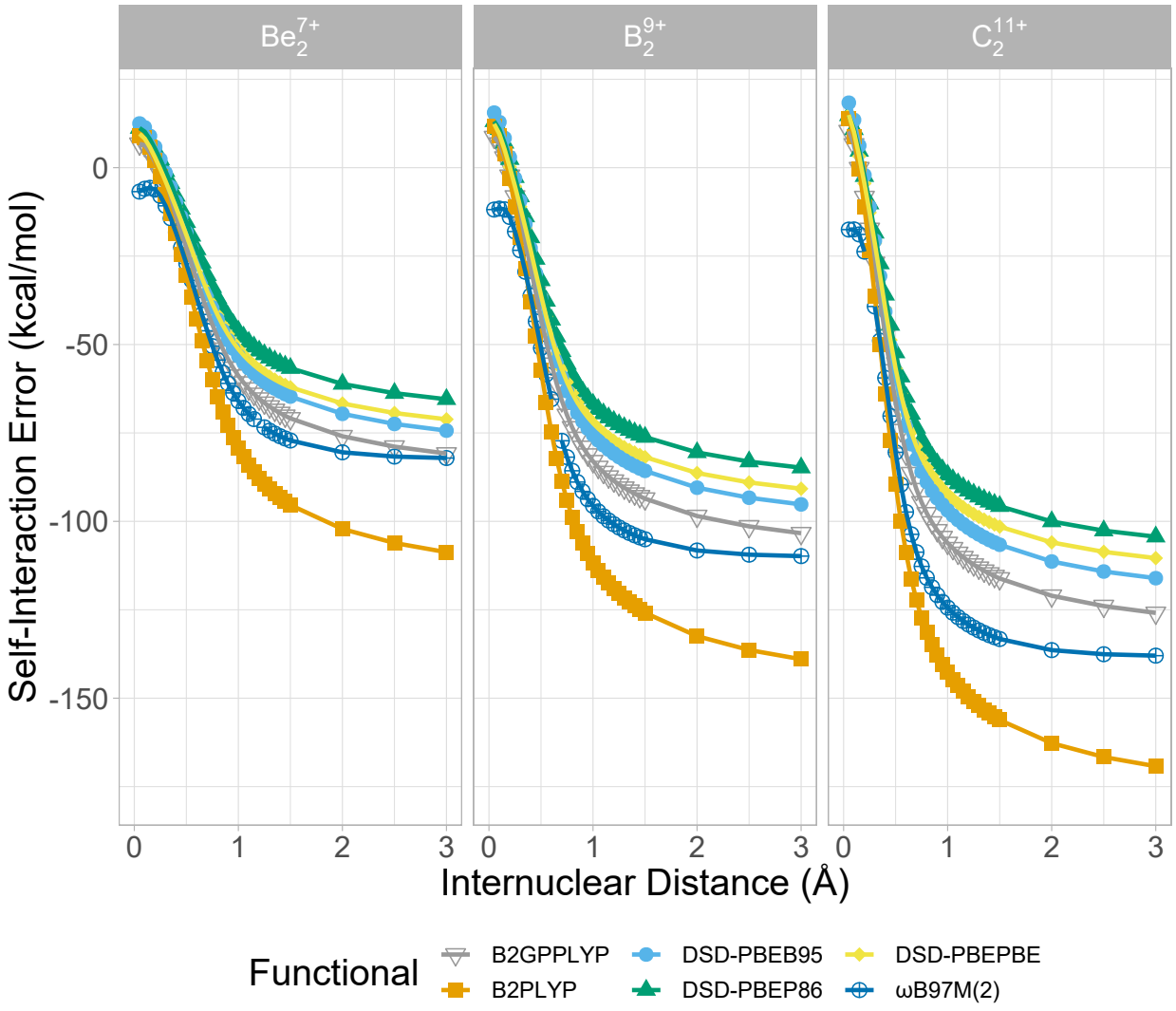

Figure S250: Self-interaction error vs internuclear distance of the homonuclear pairs, Be_2^{7+} B_2^{9+}, C_2^{11+}, for B2GPPLYP, B2PLYP, DSD-PBEB95, DSD-PBEP86, DSD-PBEPBE with ωB97M(2) as a reference.

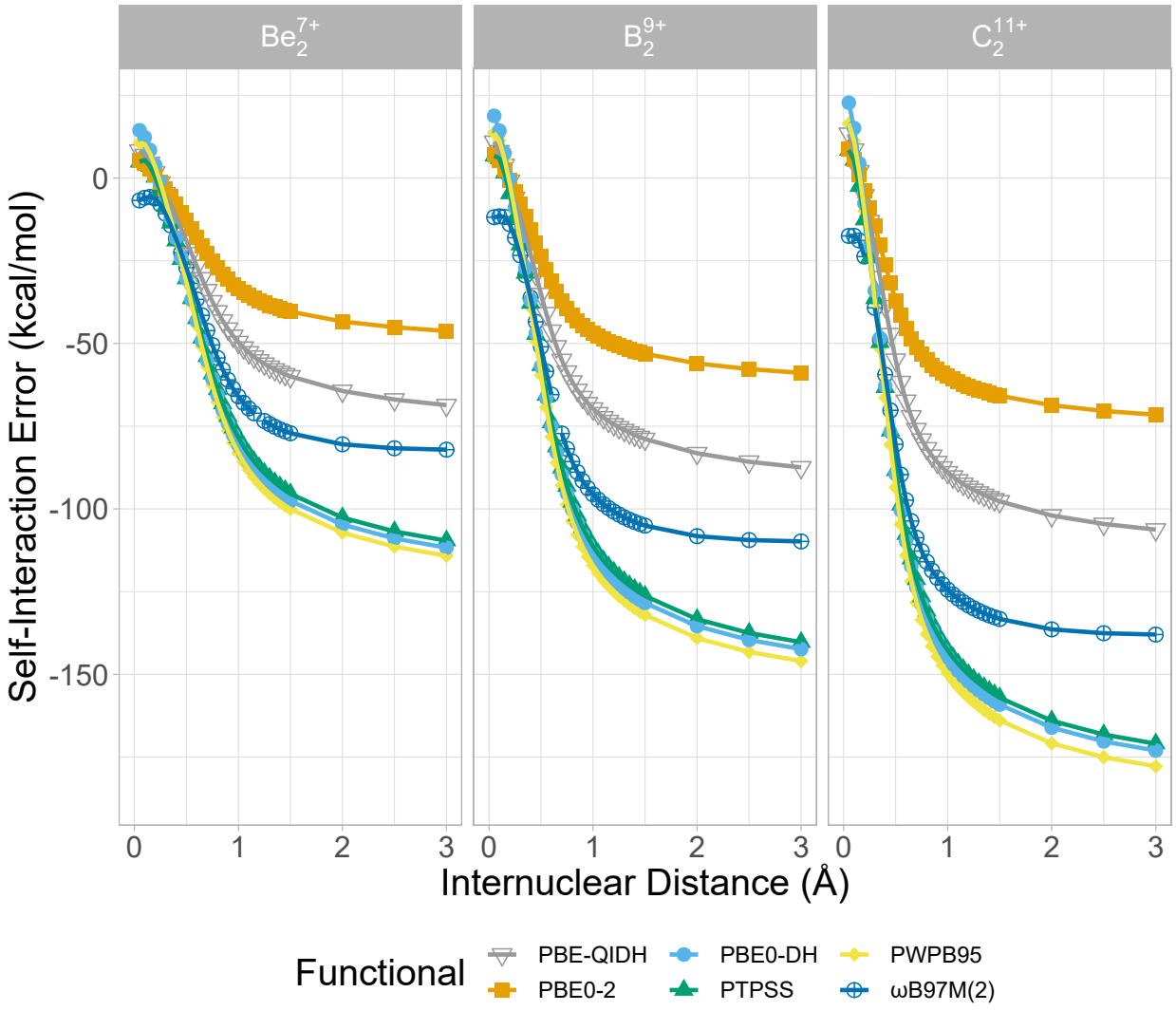

Figure S251: Self-interaction error vs internuclear distance of the homonuclear pairs, Be_2^{7+} B_2^{9+}, C_2^{11+}, for PBE-QIDH, PBE0-2, PBE0-DH, PTPSS, PWPB95 with ωB97M(2) as a reference.

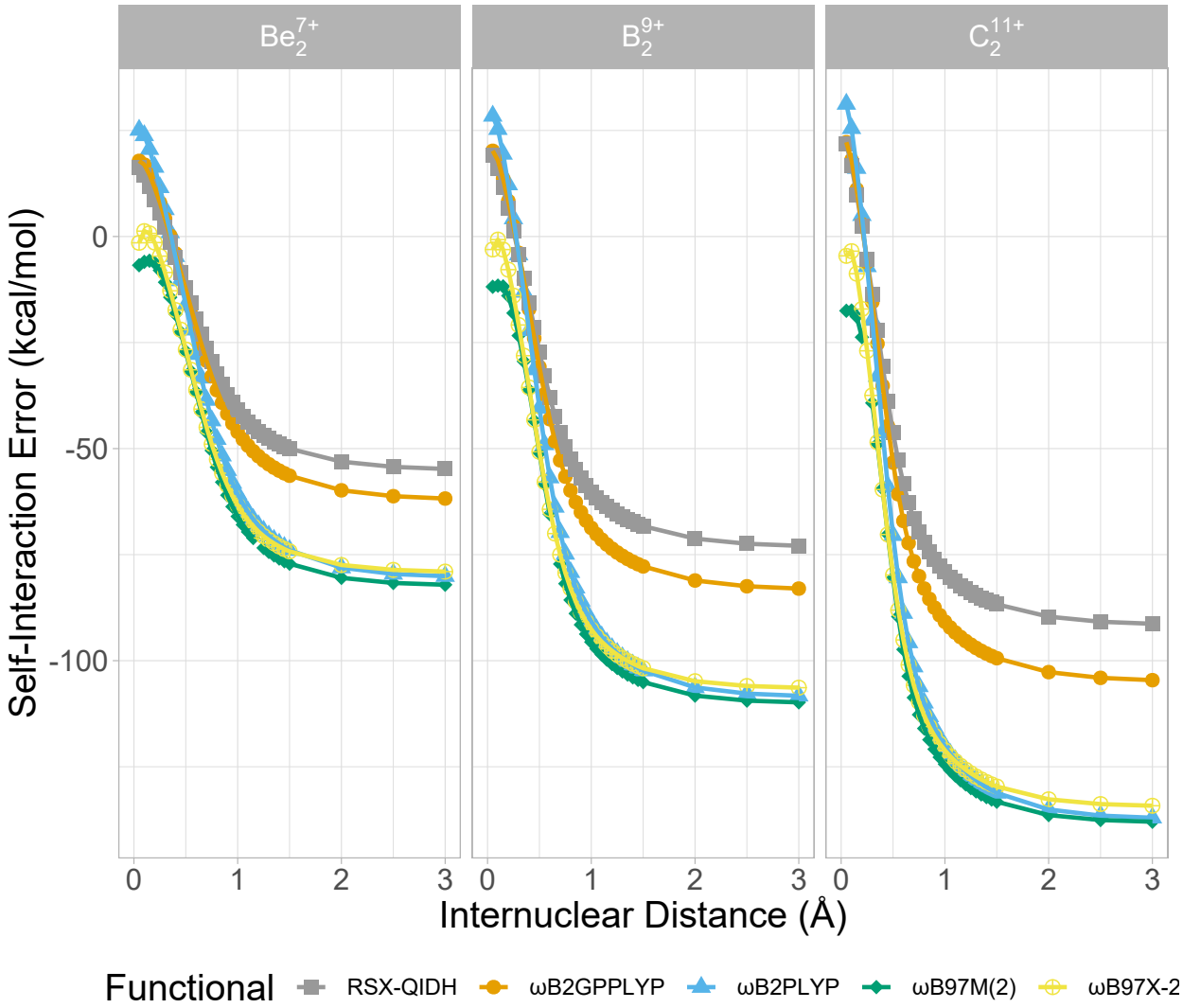

Figure S252: Self-interaction error vs internuclear distance of the homonuclear pairs, Be_2^{7+} B_2^{9+}, C_2^{11+}, for RSX-QIDH, ωB2GPPLYP, ωB2PLYP, ωB97X-2 with ωB97M(2) as a reference.

SI.3.7 Heteronuclear diatomics

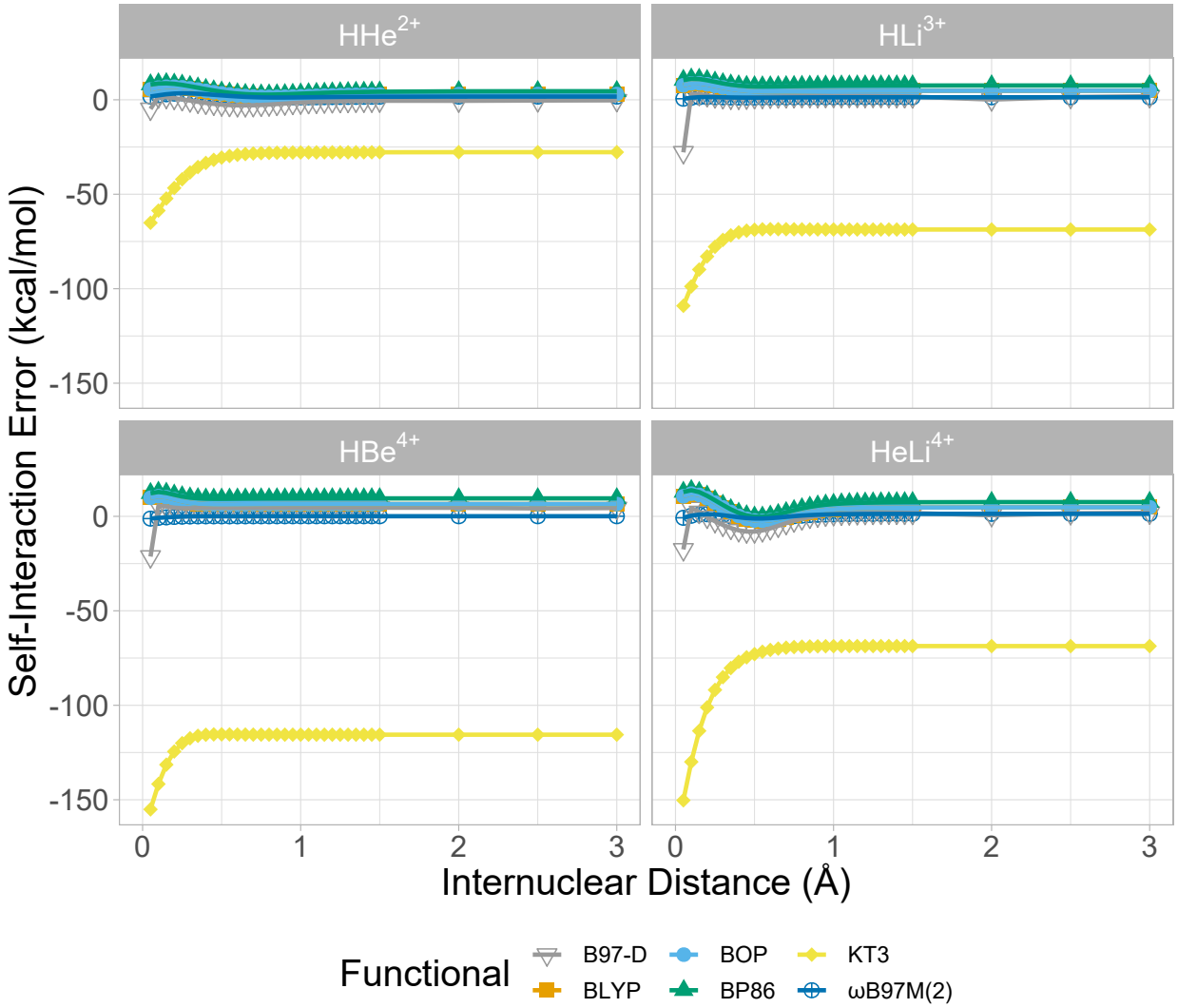

Figure S253: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, $HLi^{3}+$, $HBe^{4}+$, $HeLi^{4}+$, for B97-D, BLYP, BOP, BP86, KT3 with ωB97M(2) as a reference.

v

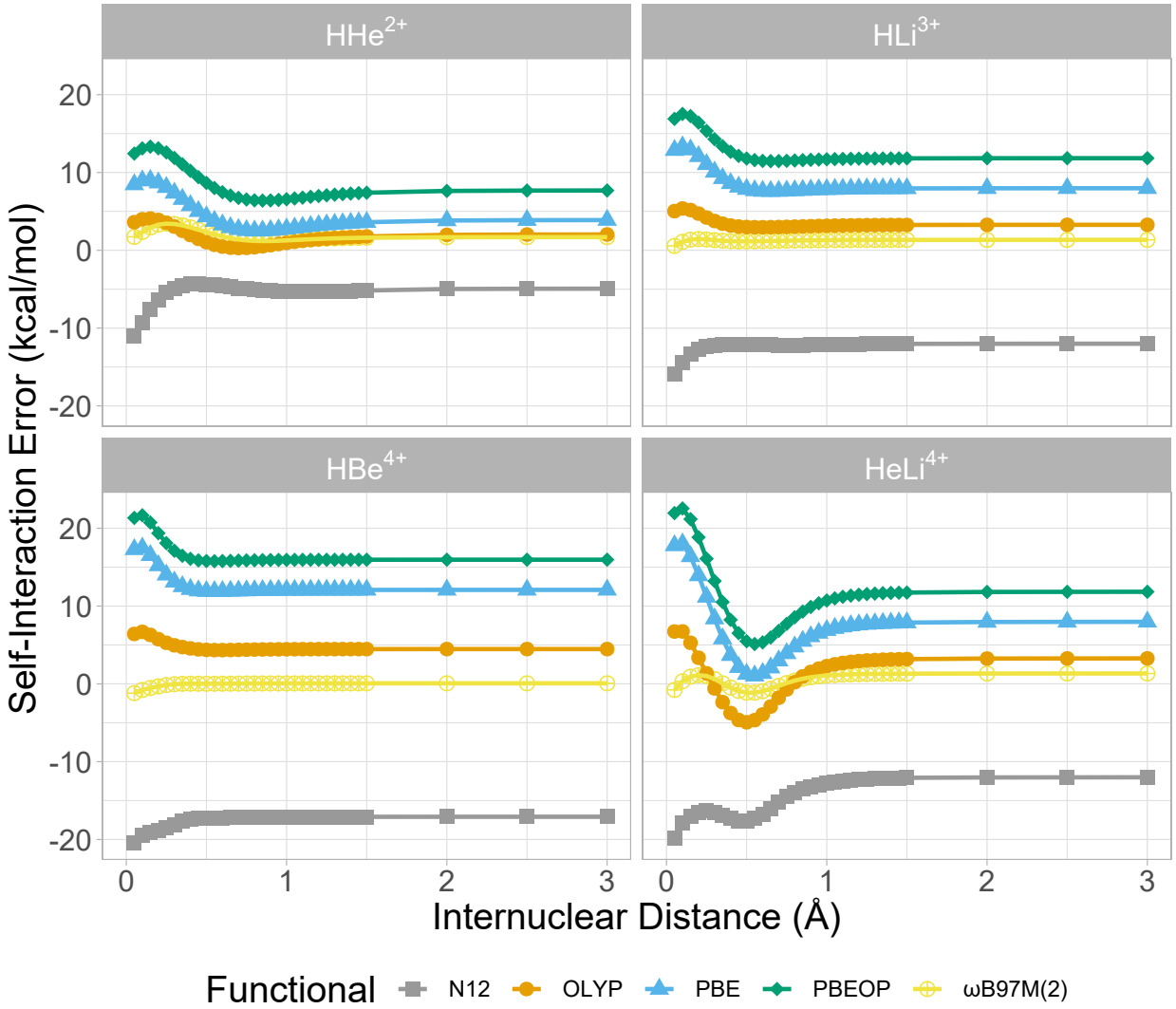

Figure S254: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, $HLi^{3}+$, $HBe^{4}+$, $HeLi^{4}+$, for N12, OLYP, PBE, PBEOP with ωB97M(2) as a reference.

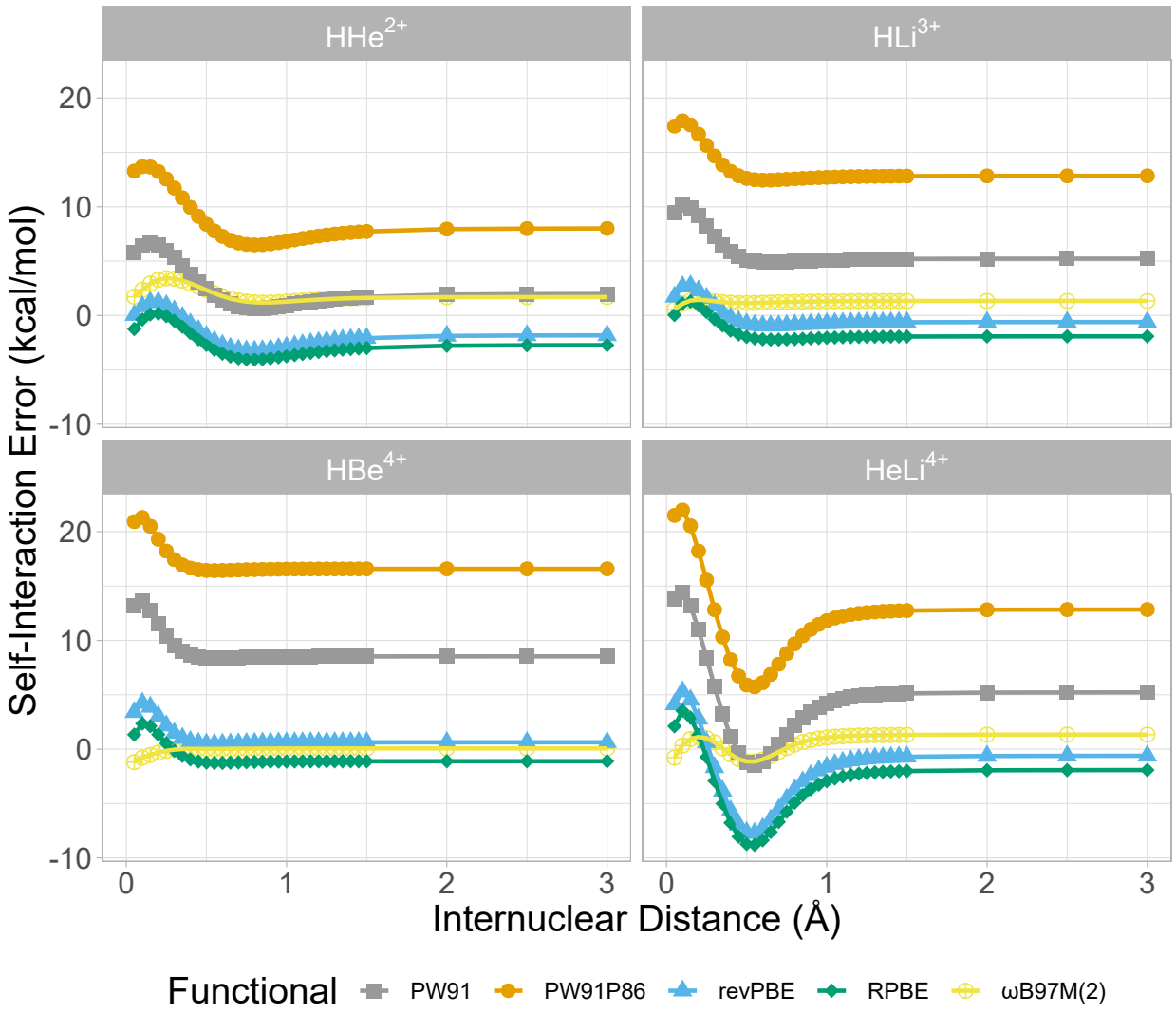

Figure S255: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, $HLi^{3}+$, $HBe^{4}+$, $HeLi^{4}+$, for PW91, PW91P86, revPBE, RPBE with ωB97M(2) as a reference.

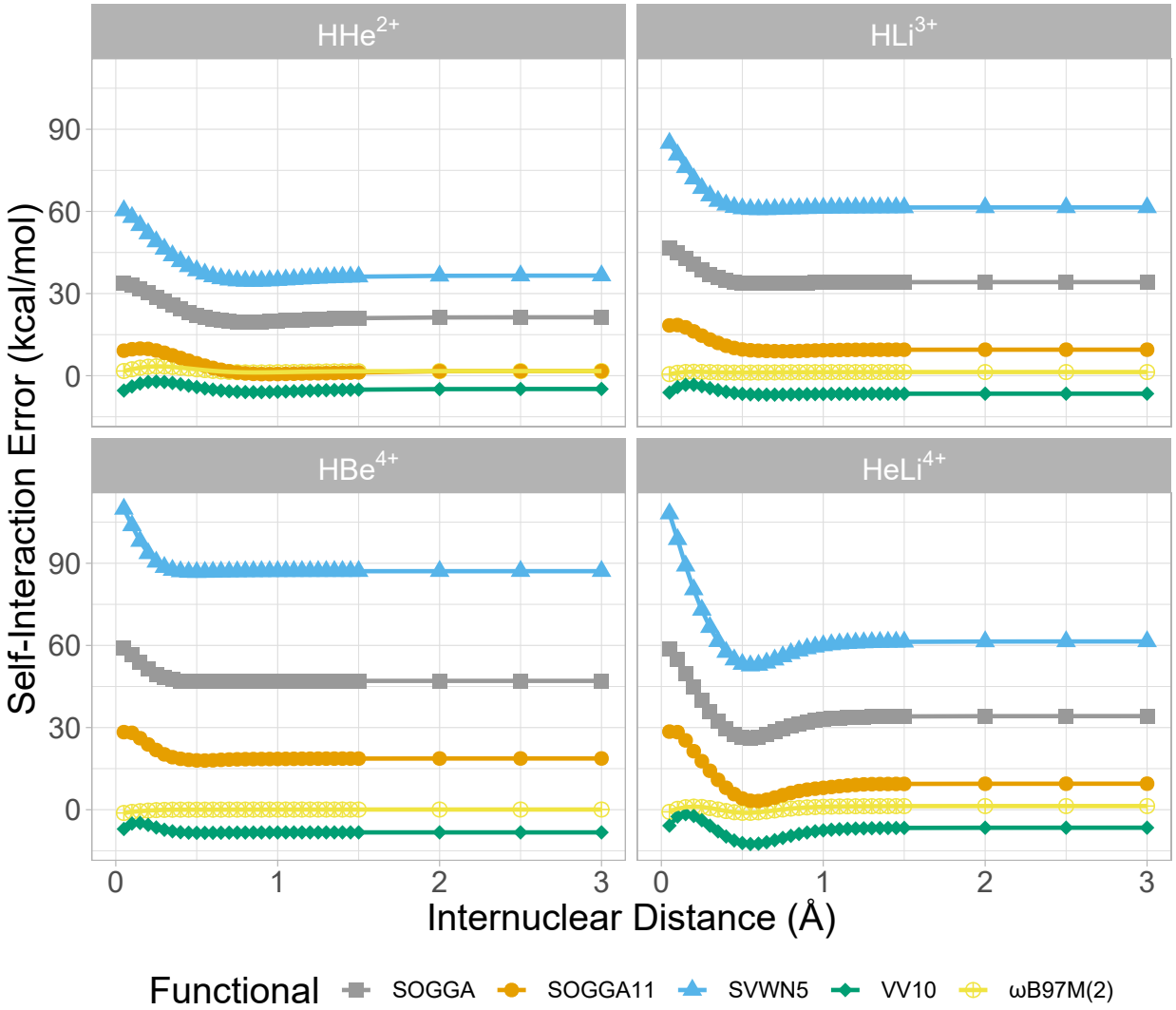

Figure S256: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, $HLi^{3}+$, $HBe^{4}+$, $HeLi^{4}+$, for SOGGA, SOGGA11, SVWN5, VV10 with ωB97M(2) as a reference.

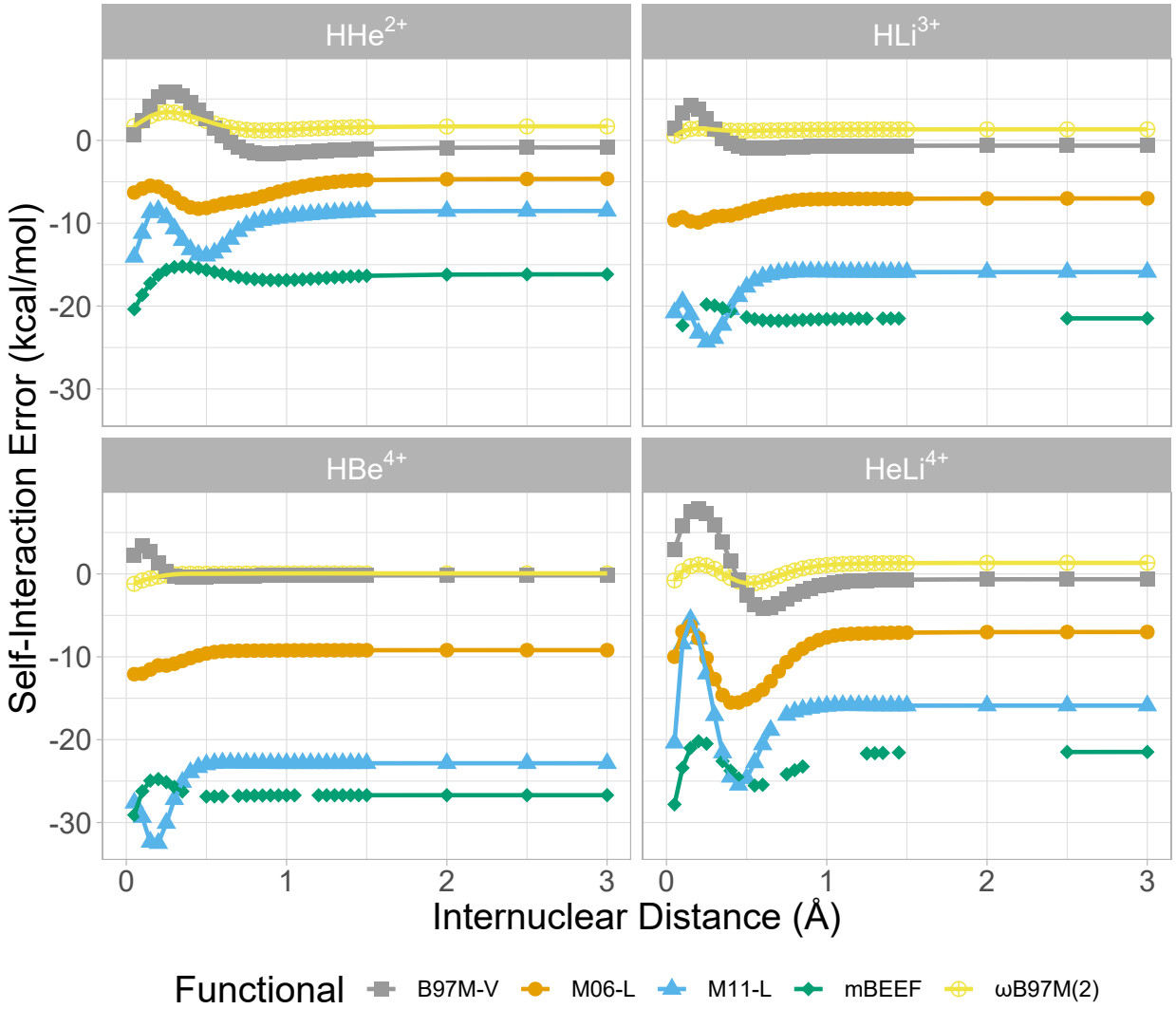

Figure S257: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, $HLi^{3}+$, $HBe^{4}+$, $HeLi^{4}+$, for B97M-V, M06-L, M11-L, mBEEF, MN12-L with ωB97M(2) as a reference.

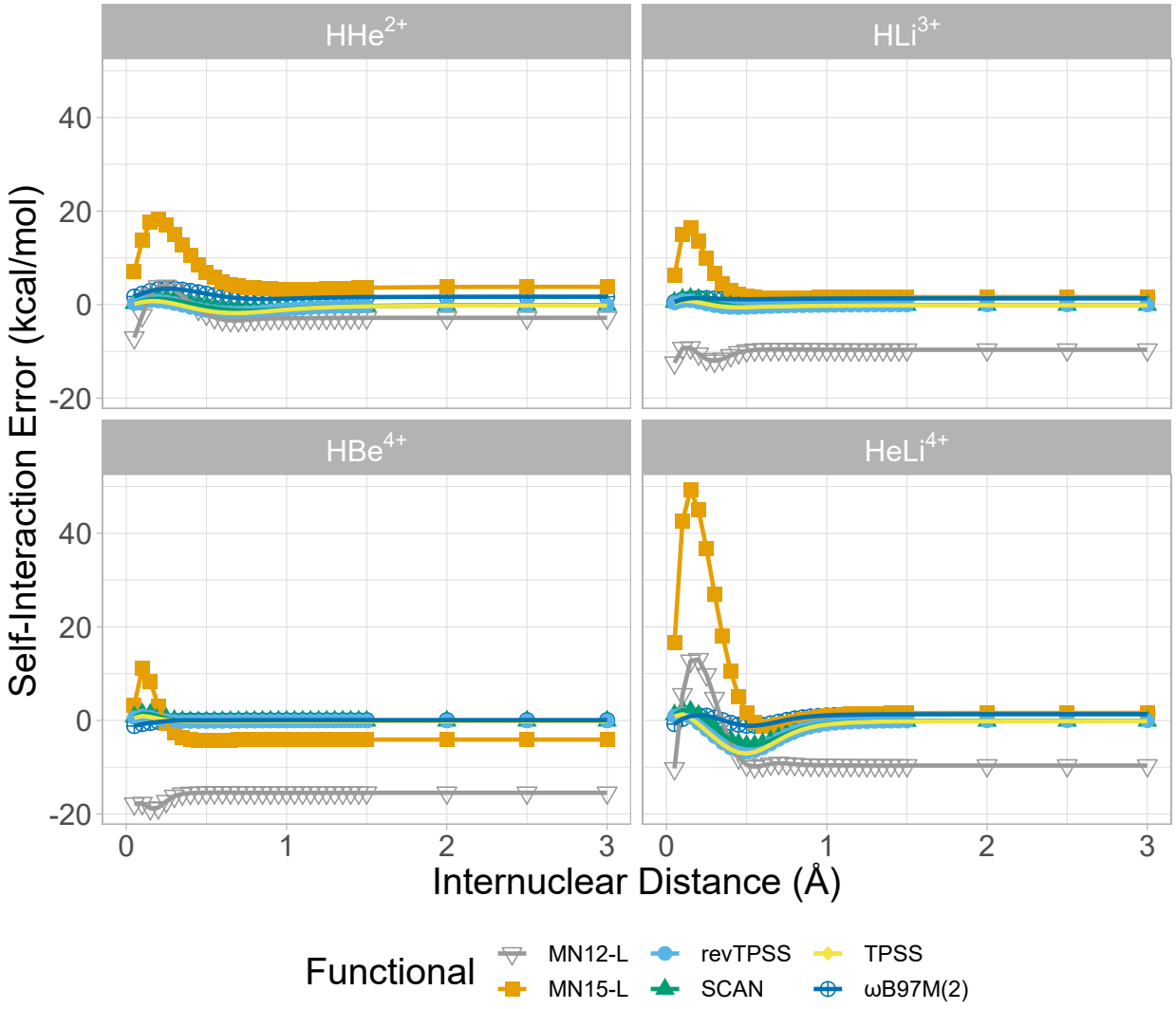

Figure S258: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, $HLi^{3}+$, $HBe^{4}+$, $HeLi^{4}+$, for MN15-L, revTPSS, SCAN, TPSS with ωB97M(2) as a reference.

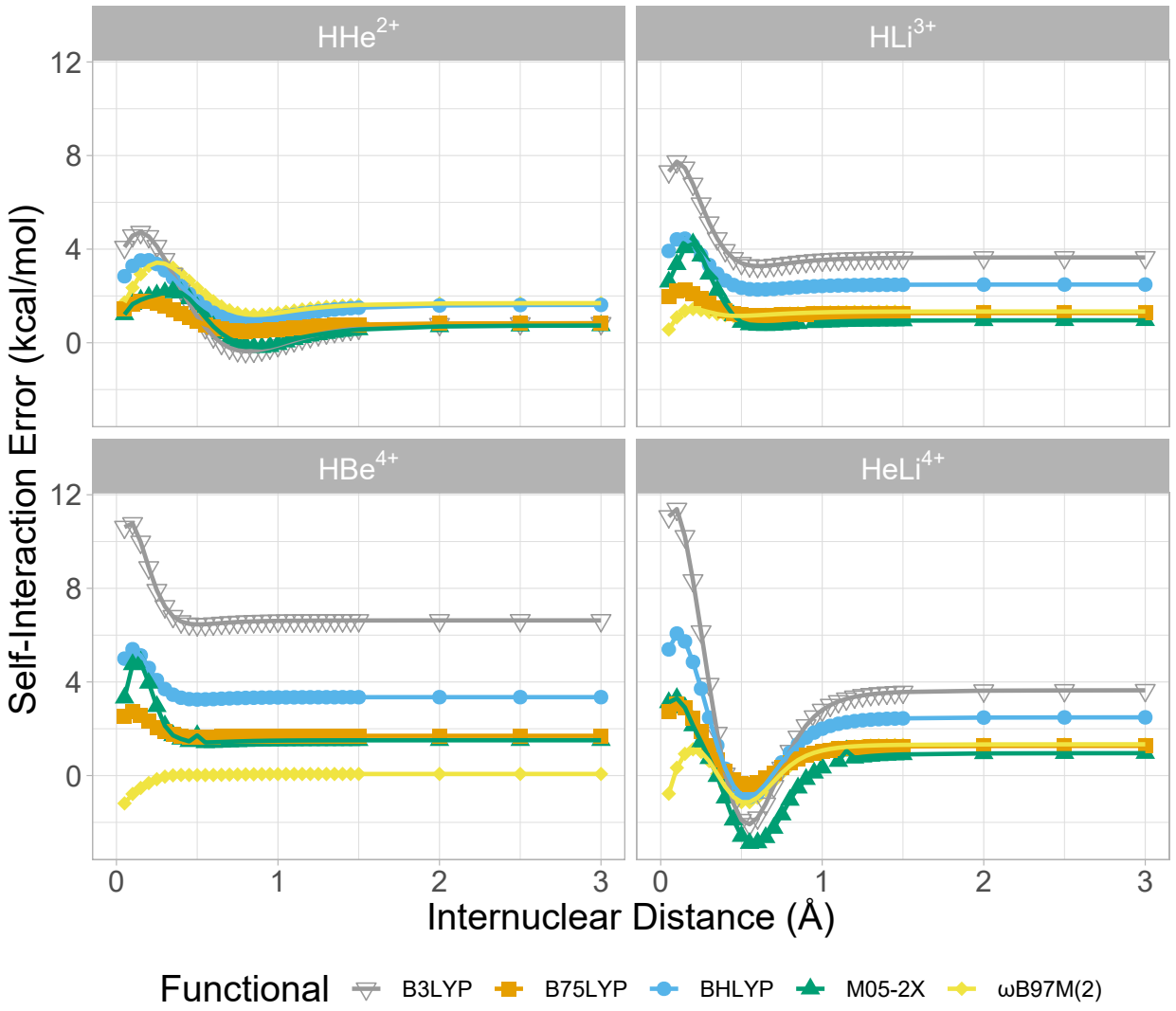

Figure S259: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, $HLi^{3}+$, $HBe^{4}+$, $HeLi^{4}+$, for B3LYP, B75LYP, B97, BHLYP, BMK with ωB97M(2) as a reference.

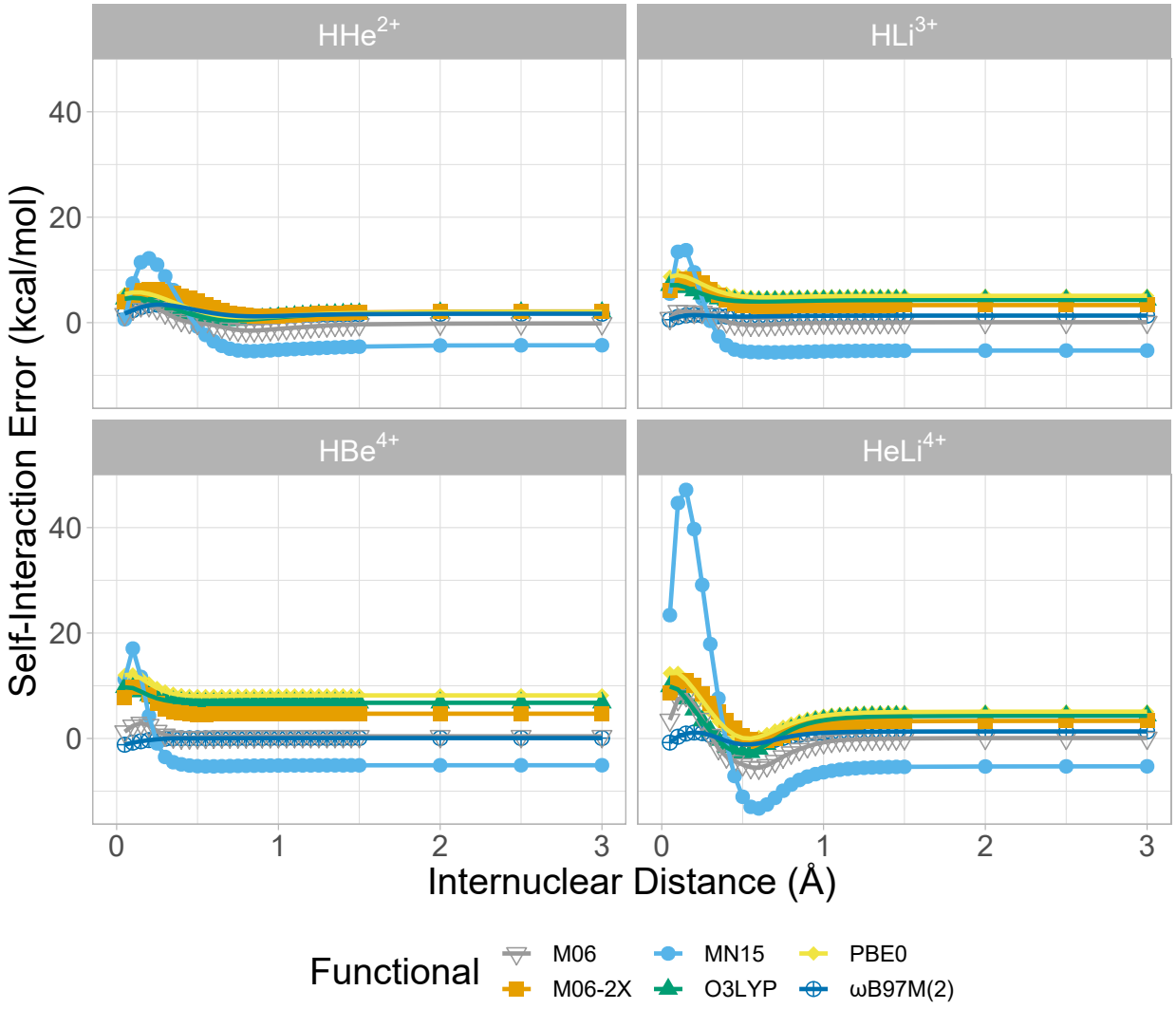

Figure S260: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, $HLi^{3}+$, $HBe^{4}+$, $HeLi^{4}+$, for M05-2X, M06, M06-2X, MN15, O3LYP with ωB97M(2) as a reference.

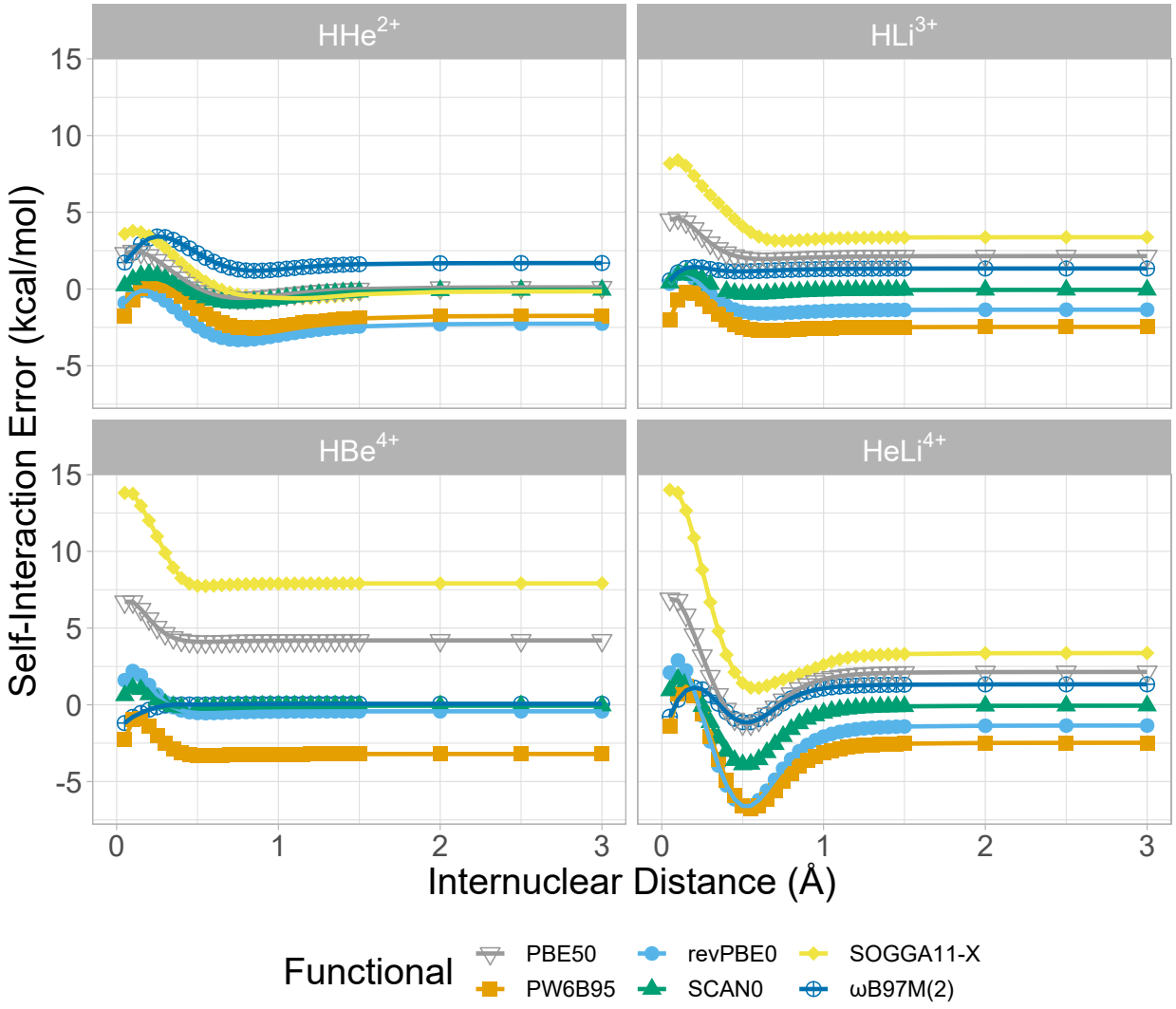

Figure S261: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, $HLi^{3}+$, $HBe^{4}+$, $HeLi^{4}+$, for PBE0, PBE50, PW6B95, revPBE0, SCAN0 with ωB97M(2) as a reference.

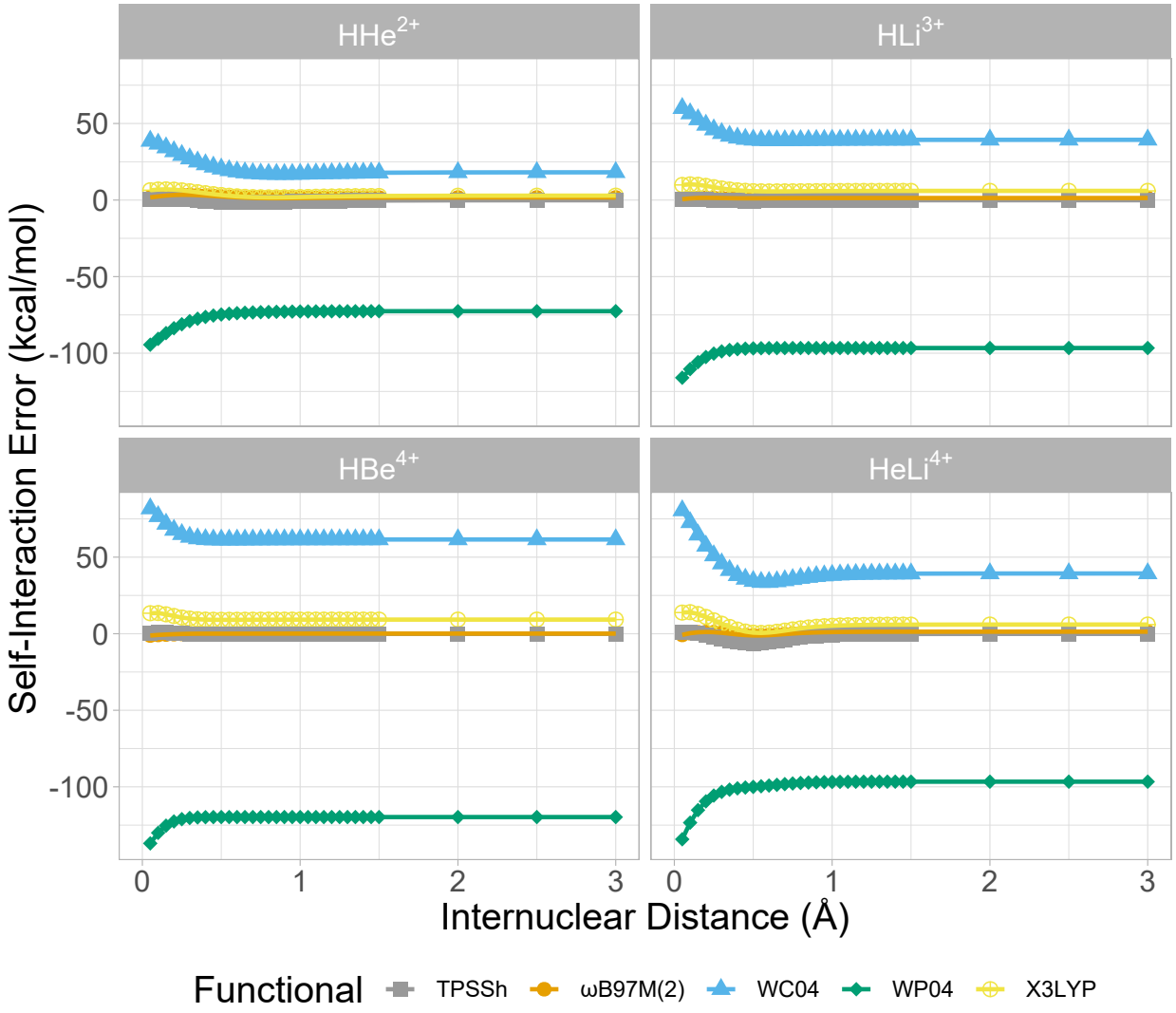

Figure S262: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, $HLi^{3}+$, $HBe^{4}+$, $HeLi^{4}+$, for SOGGA11-X, TPSSh, WC04, WP04, X3LYP with ωB97M(2) as a reference.

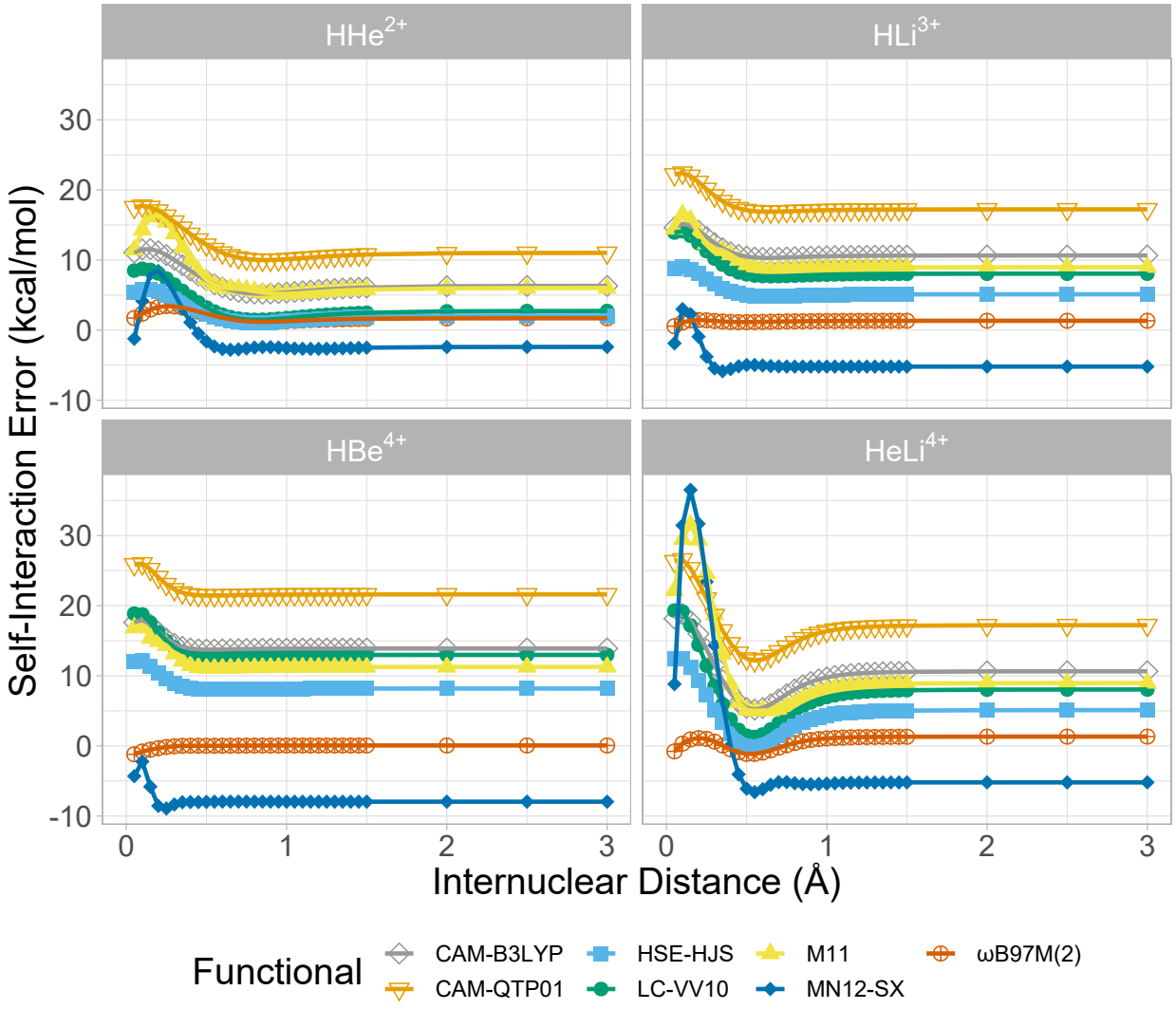

Figure S263: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, $HLi^{3}+$, $HBe^{4}+$, $HeLi^{4}+$, for CAM-B3LYP, CAM-QTP01, HSE-HJS, LC-VV10, M11, MN12-SX with ωB97M(2) as a reference.

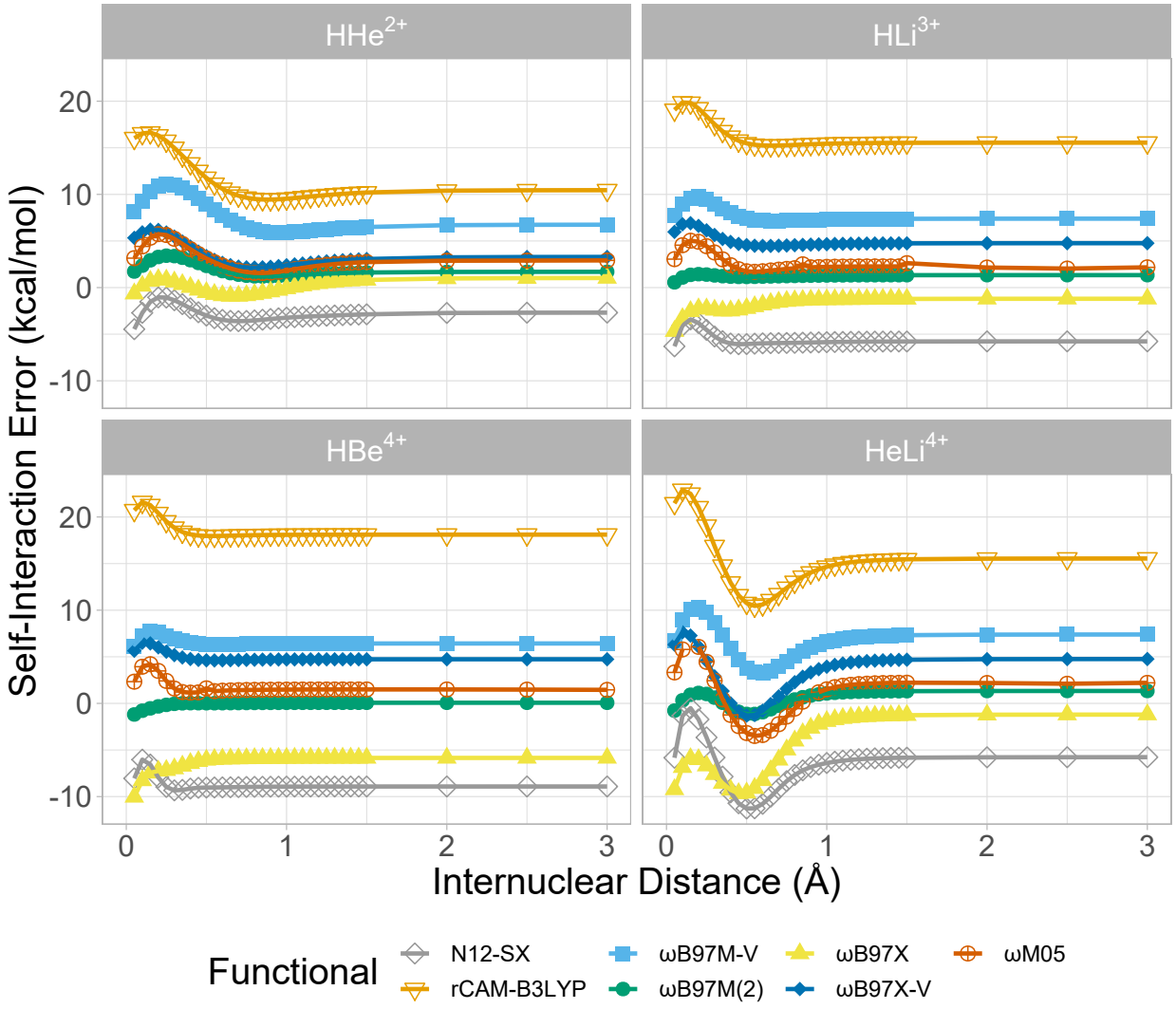

Figure S264: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, $HLi^{3}+$, $HBe^{4}+$, $HeLi^{4}+$, for N12-SX, rCAM-B3LYP, ωB97M-V, ωB97X, ωB97X-V, ωM05 with ωB97M(2) as a reference.

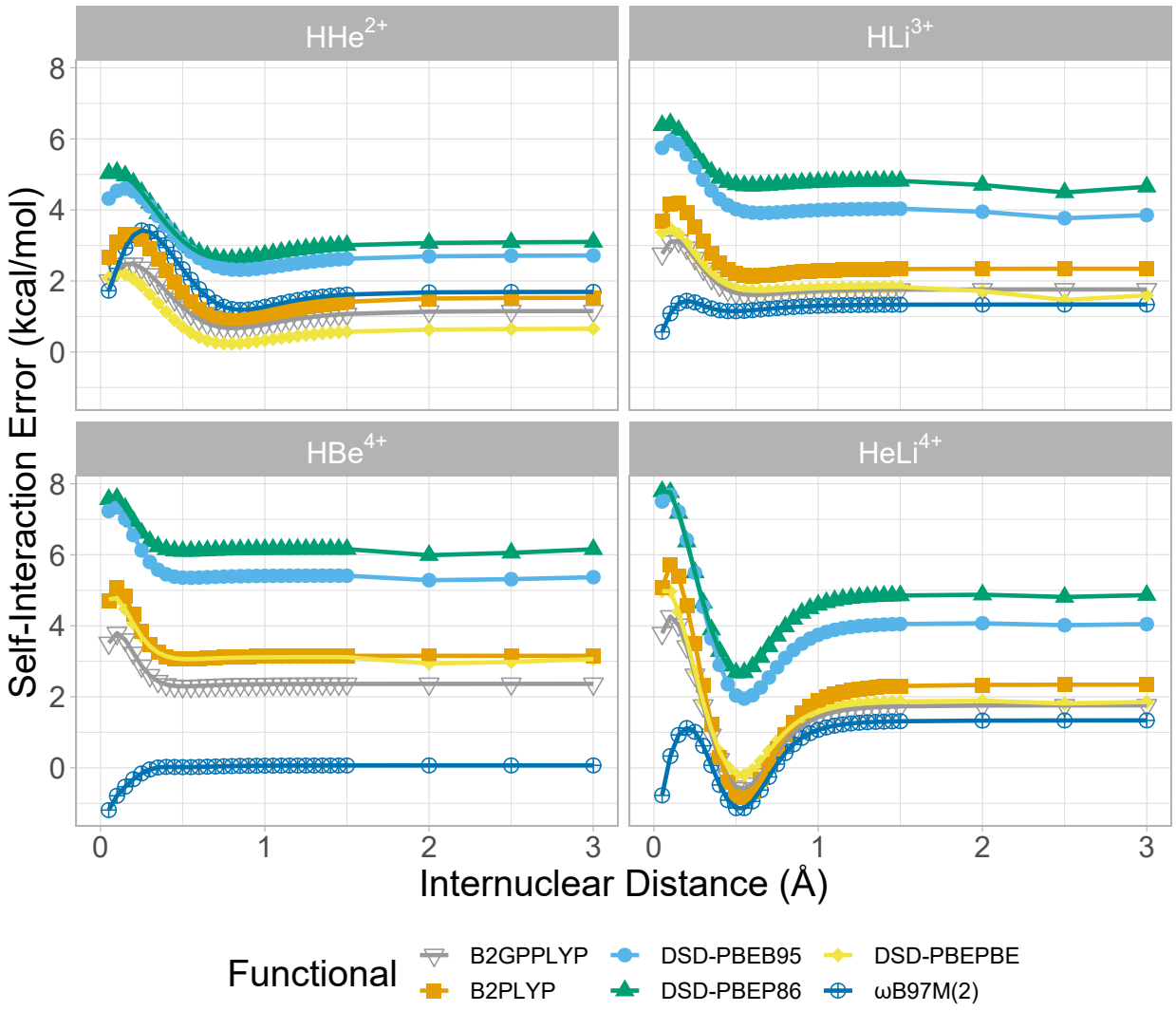

Figure S265: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, $HLi^{3}+$, $HBe^{4}+$, $HeLi^{4}+$, for B2GPPLYP, B2PLYP, DSD-PBEB95, DSD-PBEP86, DSD-PBEPBE with ωB97M(2) as a reference.

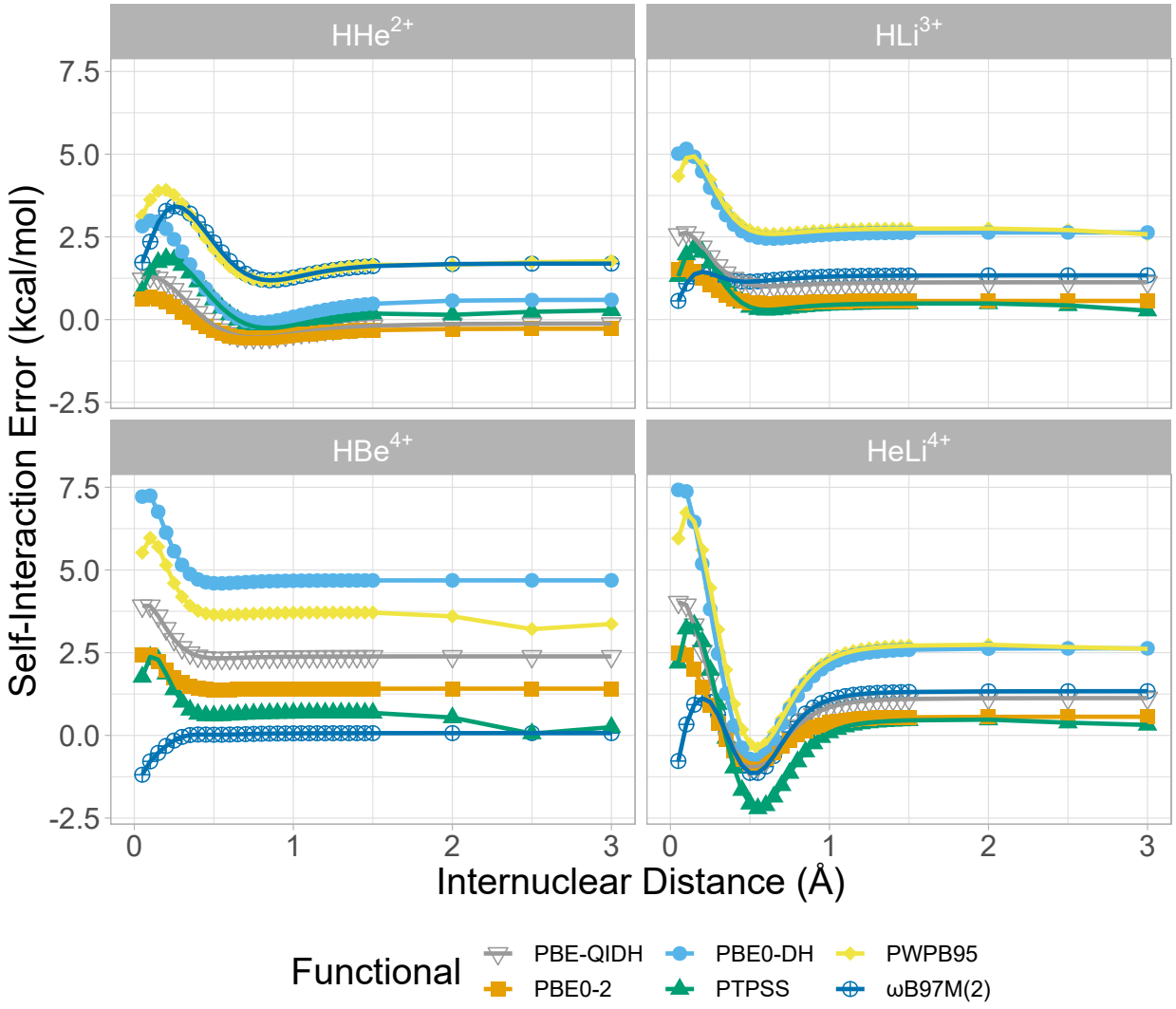

Figure S266: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, $HLi^{3}+$, $HBe^{4}+$, $HeLi^{4}+$, for PBE-QIDH, PBE0-2, PBE0-DH, PTPSS, PWPB95 with ωB97M(2) as a reference.

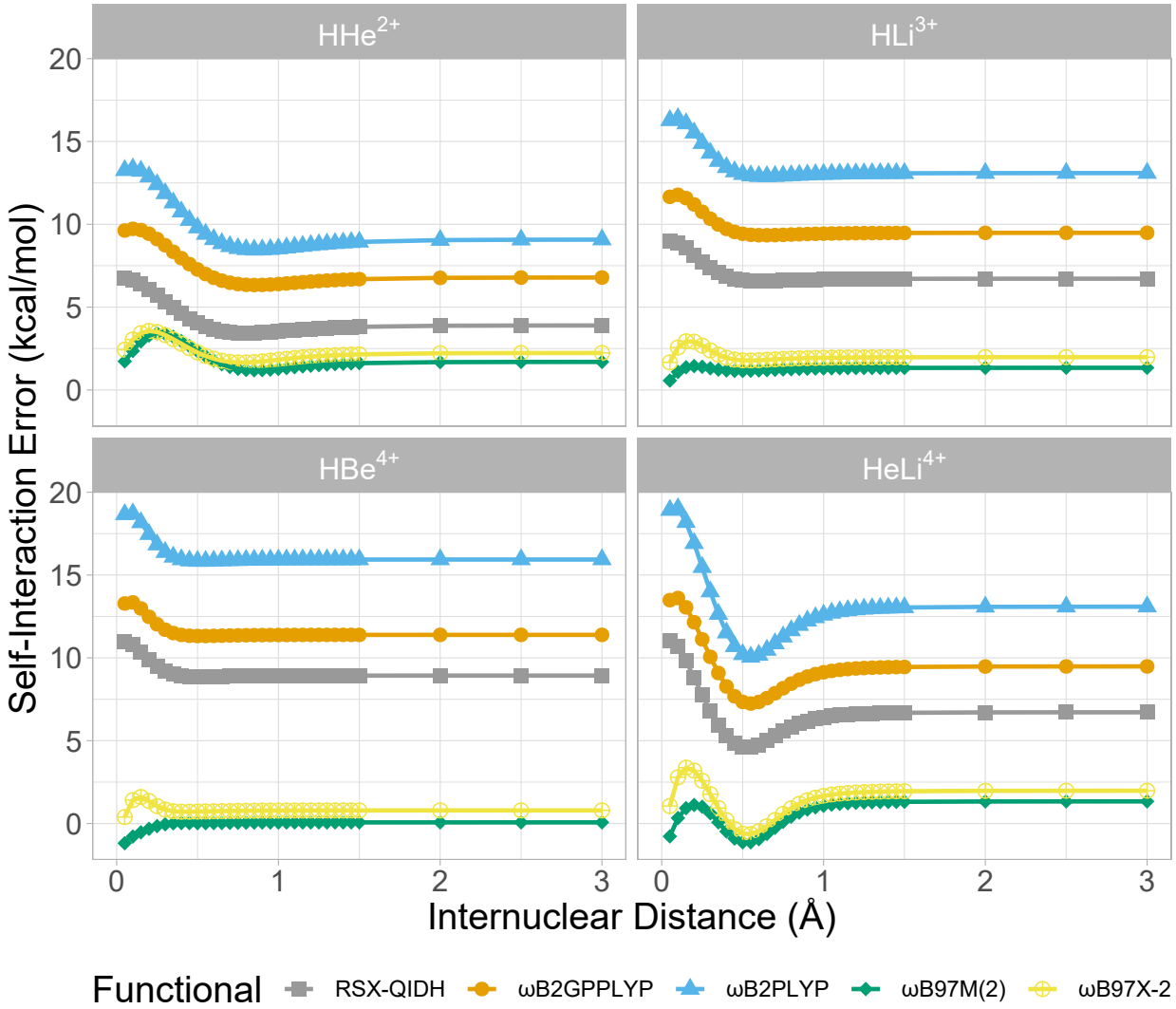

Figure S267: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, $HLi^{3}+$, $HBe^{4}+$, $HeLi^{4}+$, for RSX-QIDH, ωB2GPPLYP, ωB2PLYP, ωB97X-2 with ωB97M(2) as a reference.

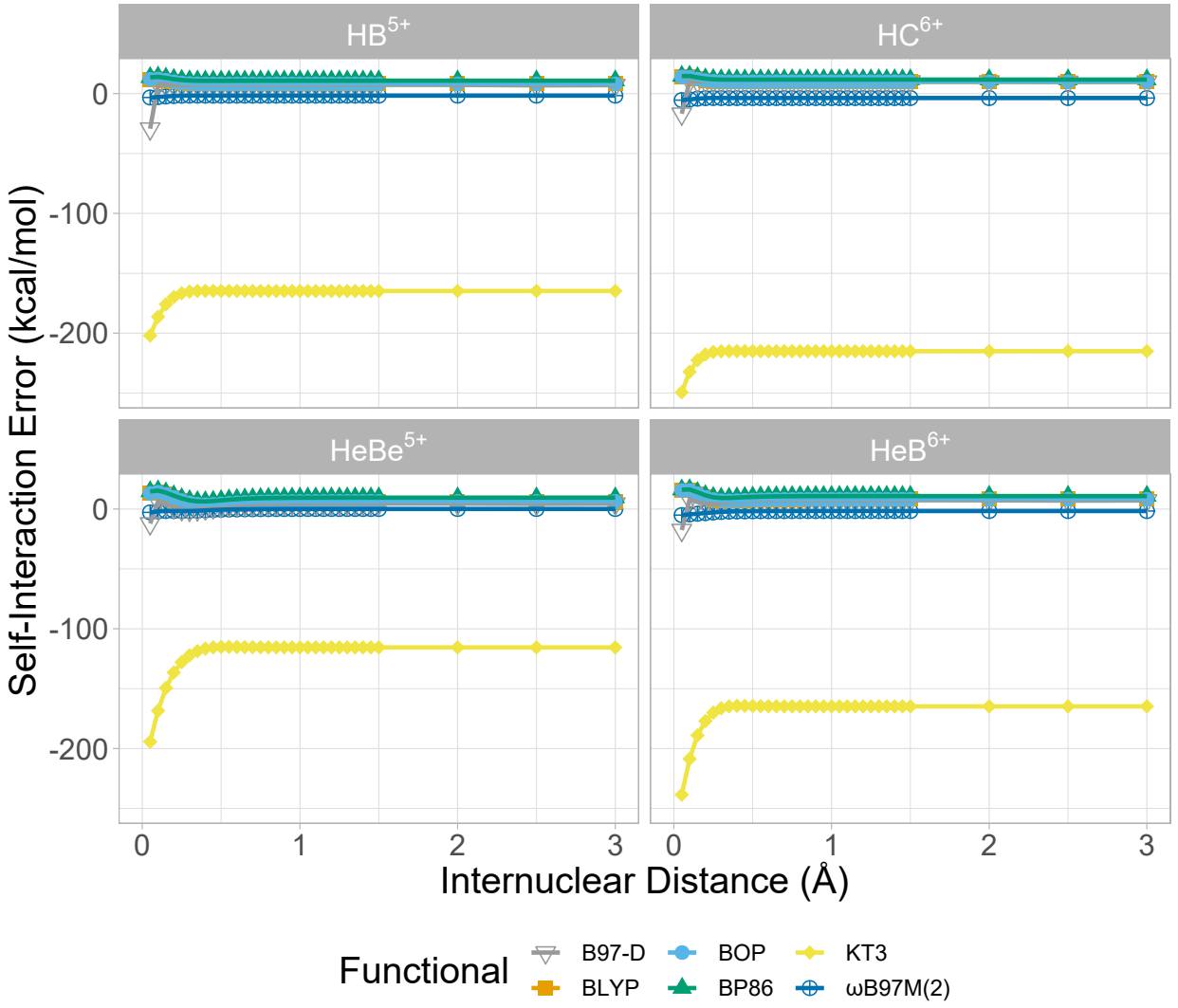

Figure S268: Self-interaction error vs internuclear distance of the heteronuclear pairs, HB^{5+}, HC^{6+}, $HeBe^{5+}$, HeB^{6+}, for B97-D, BLYP, BOP, BP86, KT3 with ωB97M(2) as a reference.

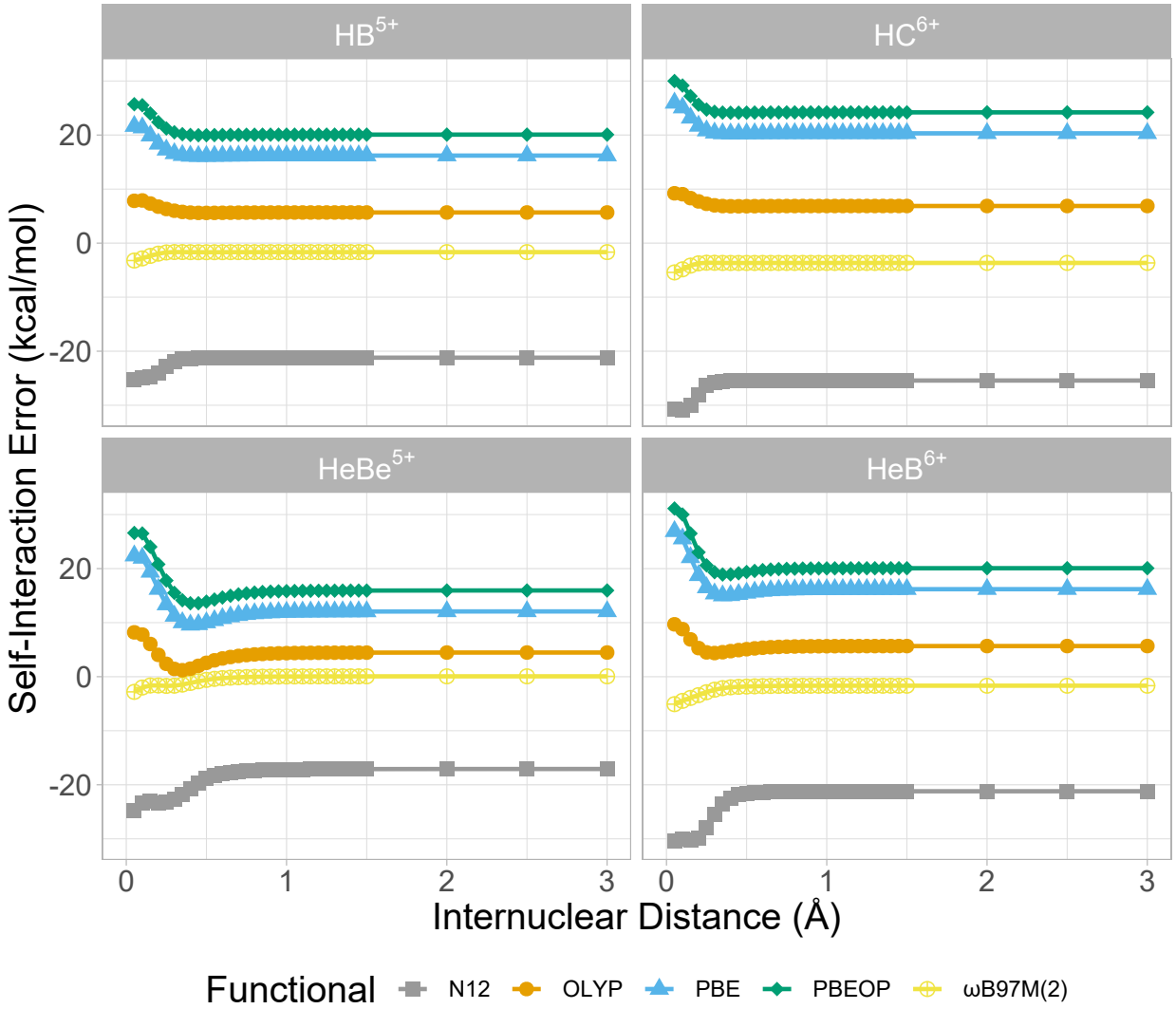

Figure S269: Self-interaction error vs internuclear distance of the heteronuclear pairs, HB^{5+}, HC^{6+}, $HeBe^{5+}$, HeB^{6+}, for N12, OLYP, PBE, PBEOP with ωB97M(2) as a reference.

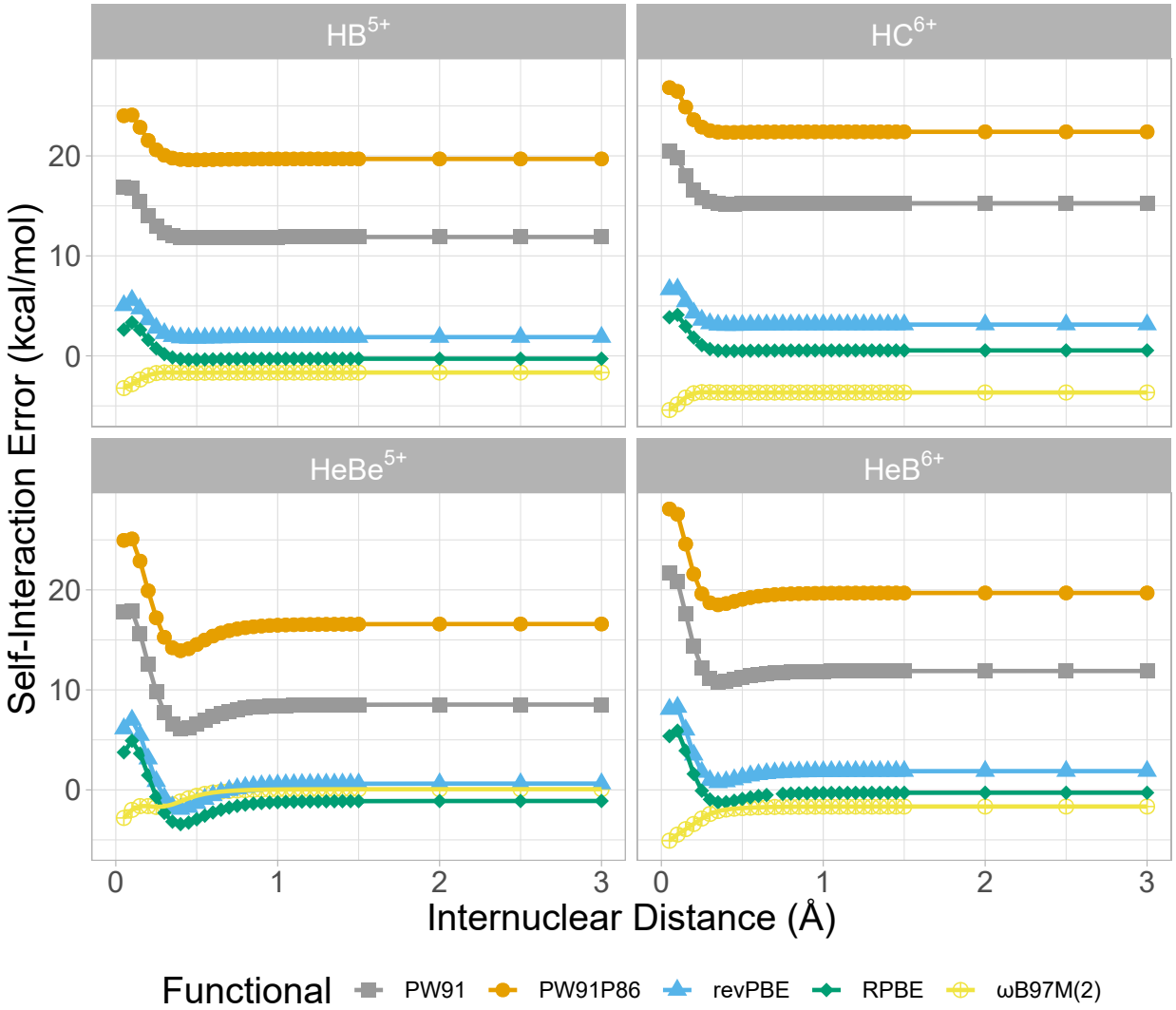

Figure S270: Self-interaction error vs internuclear distance of the heteronuclear pairs, HB^{5+}, HC^{6+}, $HeBe^{5+}$, HeB^{6+}, for PW91, PW91P86, revPBE, RPBE with ωB97M(2) as a reference.

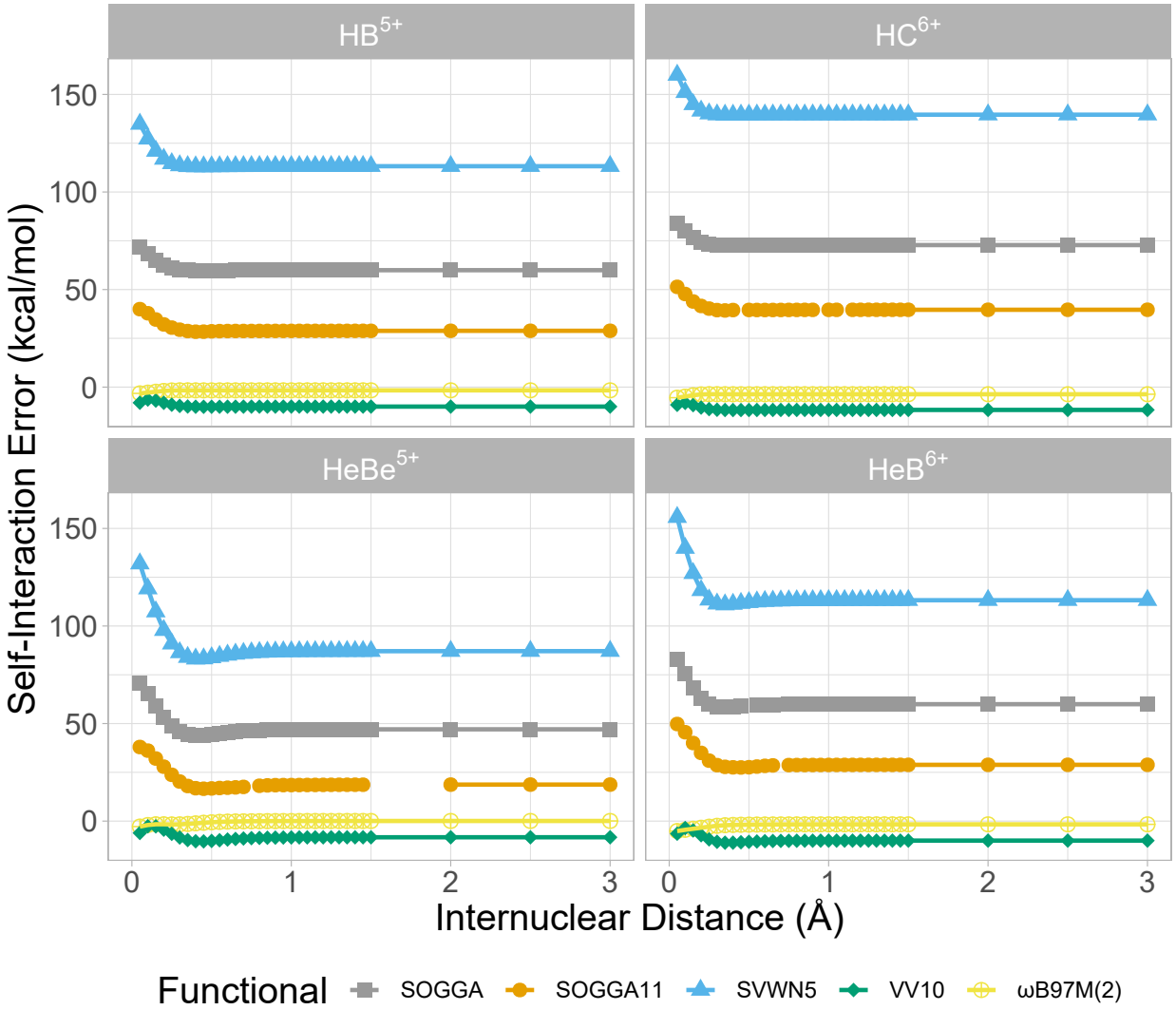

Figure S271: Self-interaction error vs internuclear distance of the heteronuclear pairs, HB^{5+}, HC^{6+}, $HeBe^{5+}$, HeB^{6+}, for SOGGA, SOGGA11, SVWN5, VV10 with ωB97M(2) as a reference.

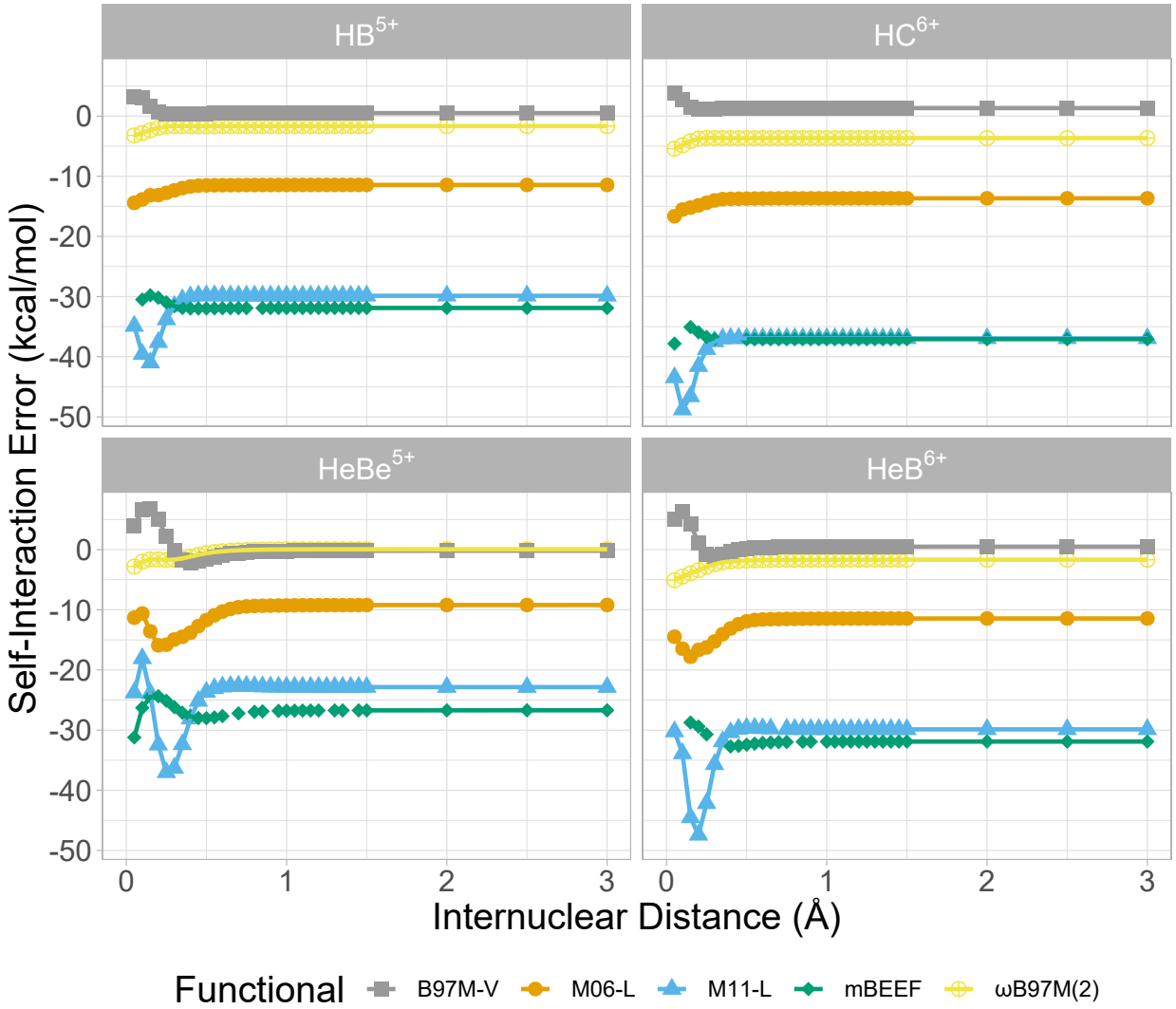

Figure S272: Self-interaction error vs internuclear distance of the heteronuclear pairs, HB^{5+}, HC^{6+}, $HeBe^{5+}$, HeB^{6+}, for B97M-V, M06-L, M11-L, mBEEF, MN12-L with ωB97M(2) as a reference.

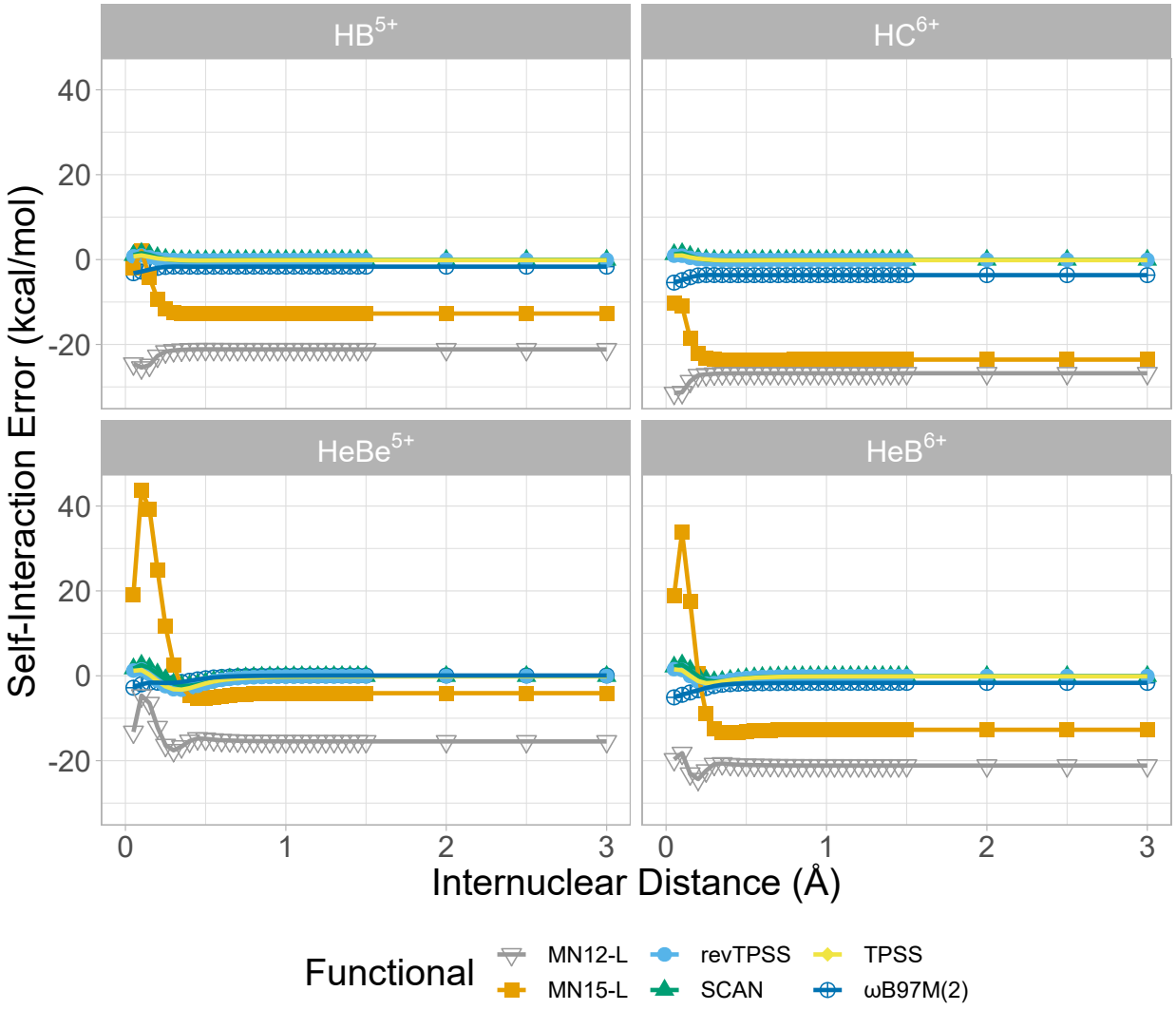

Figure S273: Self-interaction error vs internuclear distance of the heteronuclear pairs, HB^{5+}, HC^{6+}, $HeBe^{5+}$, HeB^{6+}, for MN15-L, revTPSS, SCAN, TPSS with ωB97M(2) as a reference.

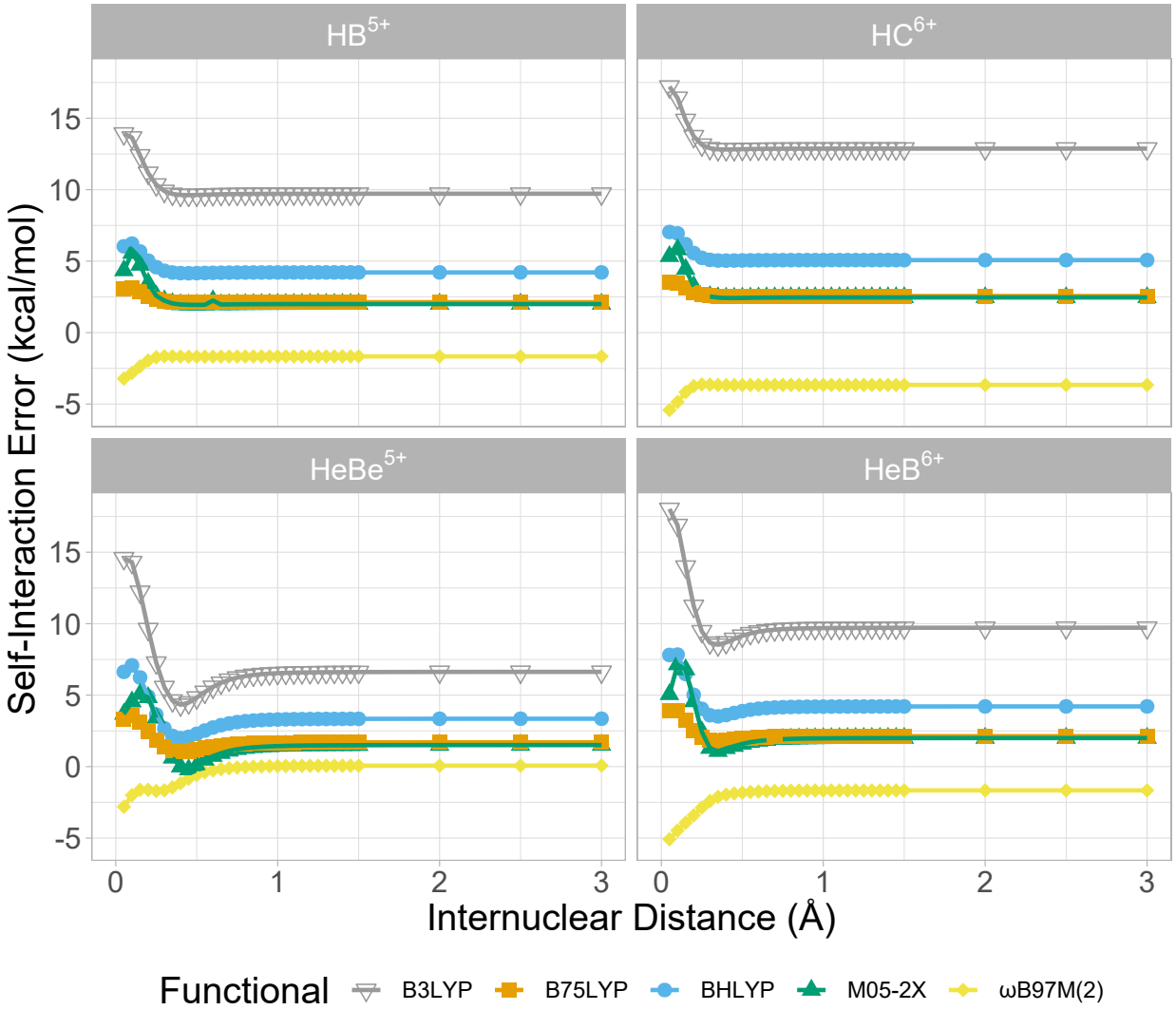

Figure S274: Self-interaction error vs internuclear distance of the heteronuclear pairs, HB^{5+}, HC^{6+}, $HeBe^{5+}$, HeB^{6+}, for B3LYP, B75LYP, B97, BHLYP, BMK with ωB97M(2) as a reference.

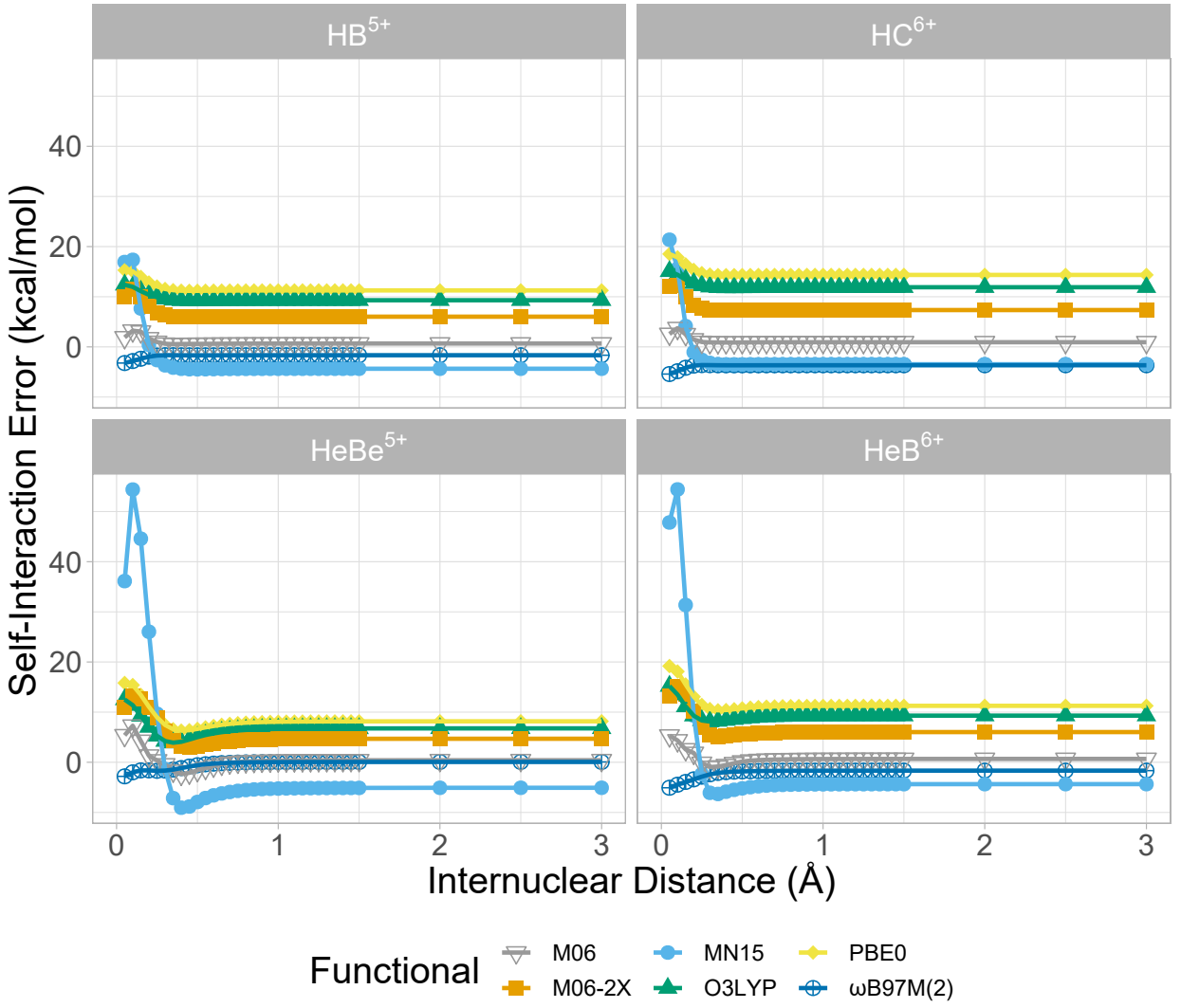

Figure S275: Self-interaction error vs internuclear distance of the heteronuclear pairs, HB^{5+}, HC^{6+}, $HeBe^{5+}$, HeB^{6+}, for M05-2X, M06, M06-2X, MN15, O3LYP with ωB97M(2) as a reference.

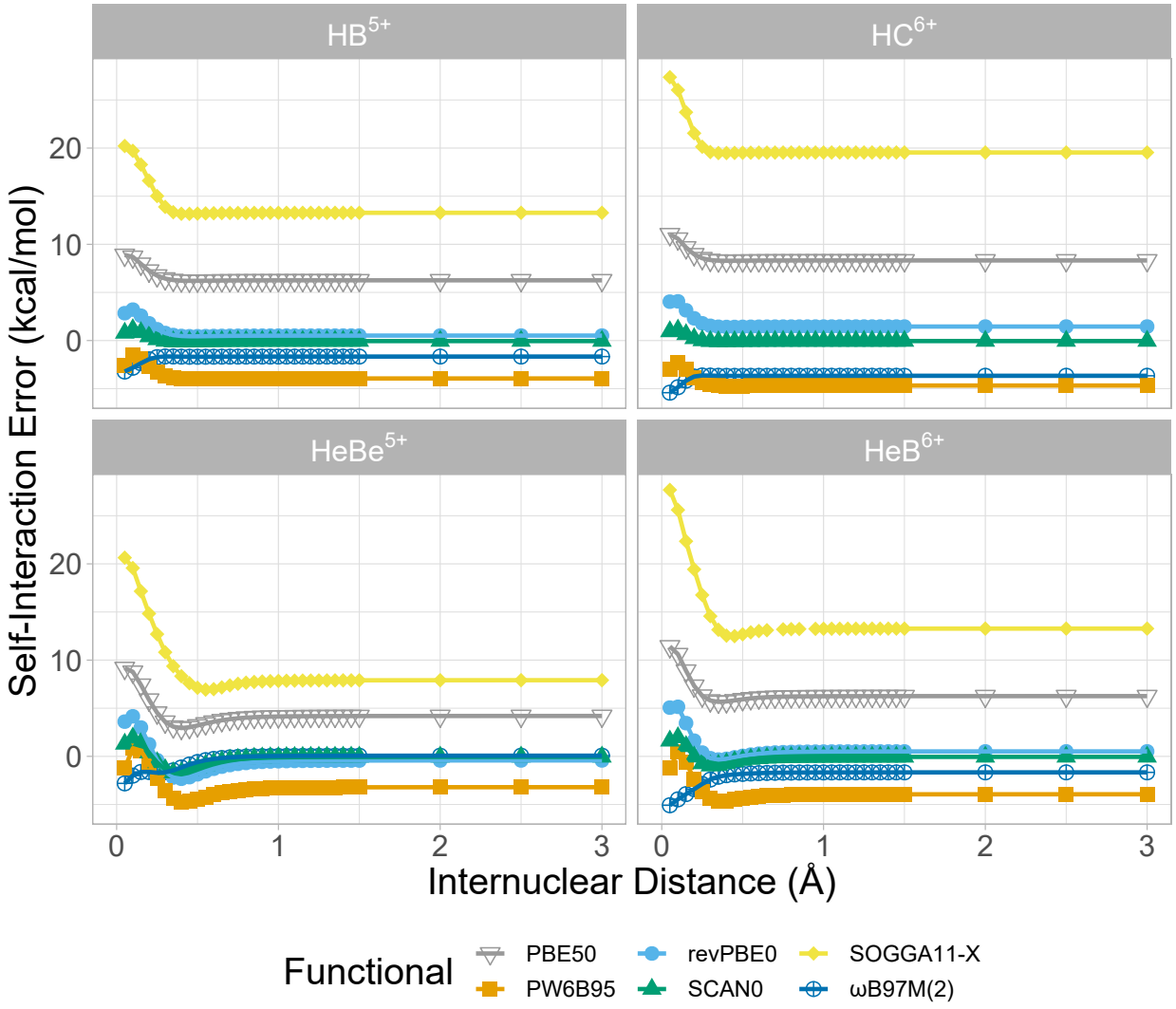

Figure S276: Self-interaction error vs internuclear distance of the heteronuclear pairs, HB^{5+}, HC^{6+}, $HeBe^{5+}$, HeB^{6+}, for PBE0, PBE50, PW6B95, revPBE0, SCAN0 with ωB97M(2) as a reference.

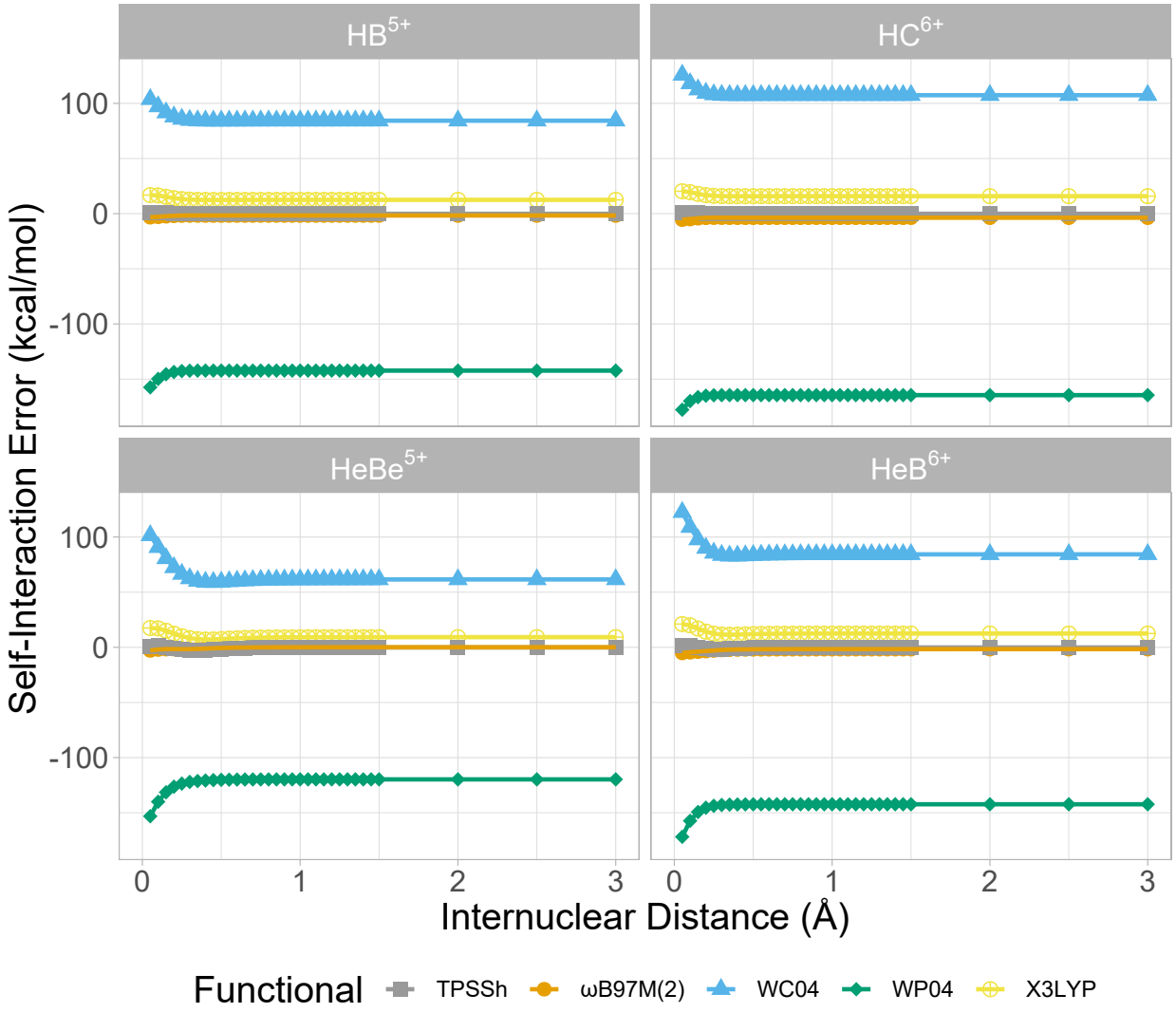

Figure S277: Self-interaction error vs internuclear distance of the heteronuclear pairs, HB^{5+}, HC^{6+}, $HeBe^{5+}$, HeB^{6+}, for SOGGA11-X, TPSSh, WC04, WP04, X3LYP with ωB97M(2) as a reference.

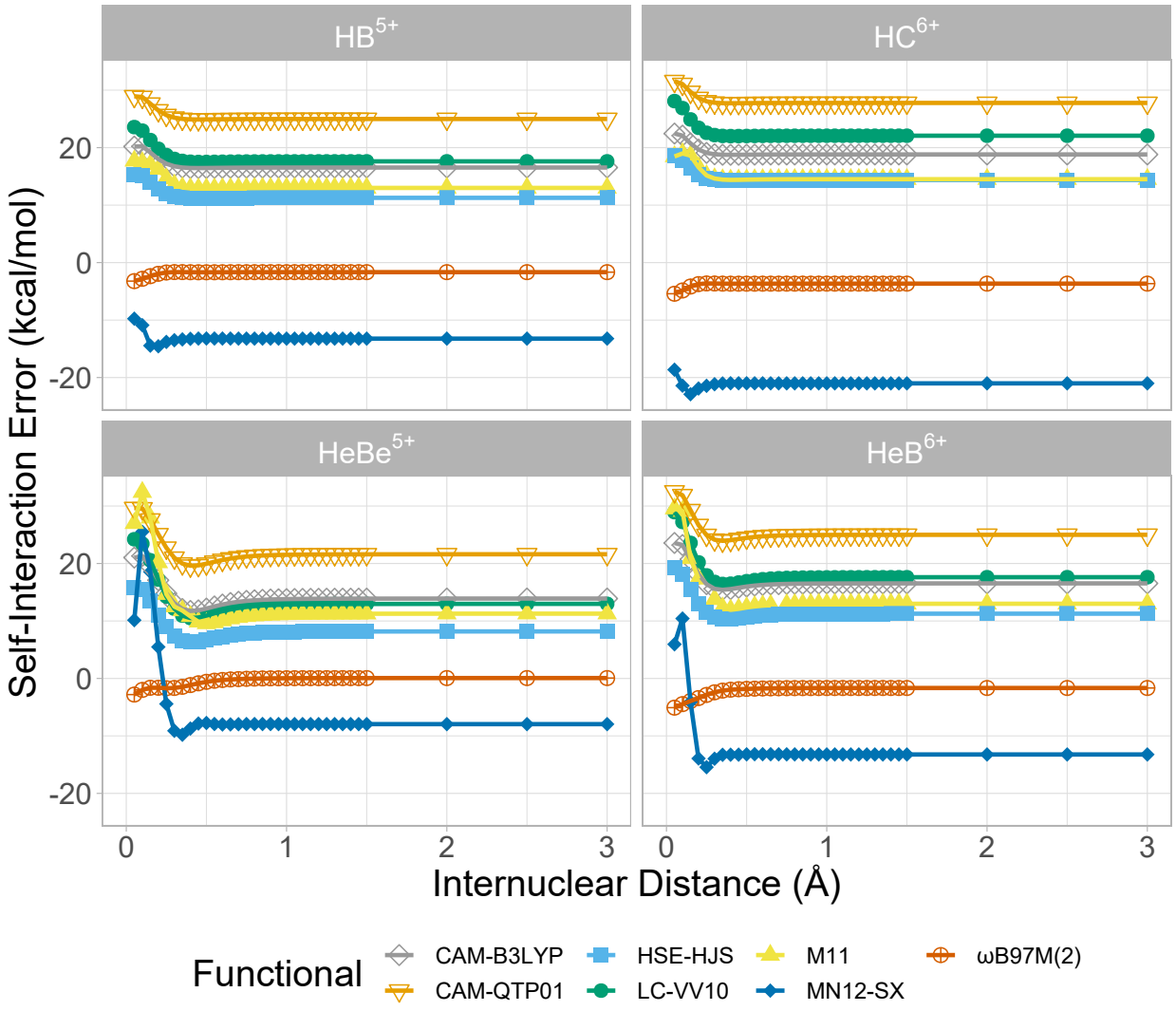

Figure S278: Self-interaction error vs internuclear distance of the heteronuclear pairs, HB^{5+}, HC^{6+}, $HeBe^{5+}$, HeB^{6+}, for CAM-B3LYP, CAM-QTP01, HSE-HJS, LC-VV10, M11, MN12-SX with ωB97M(2) as a reference.

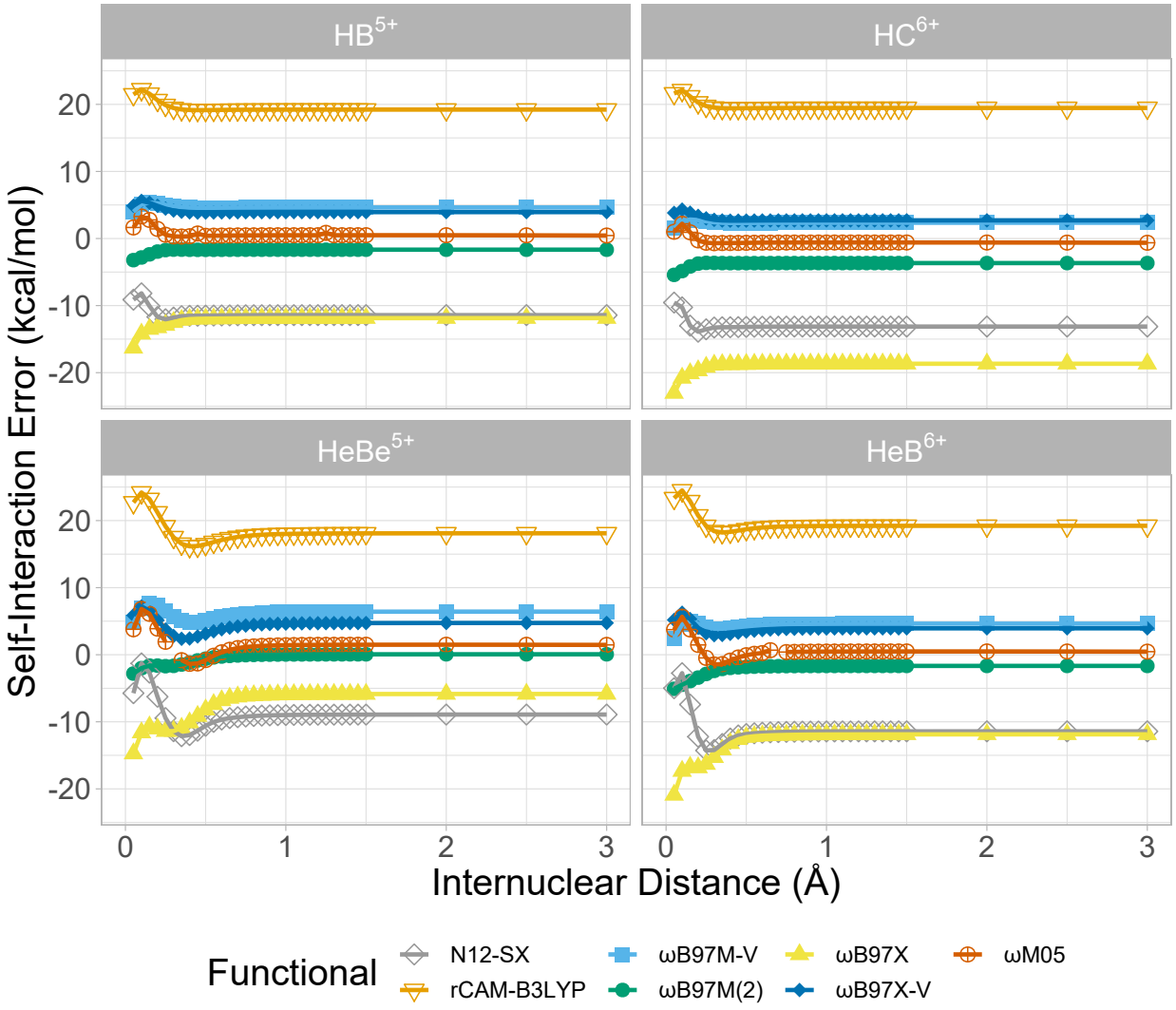

Figure S279: Self-interaction error vs internuclear distance of the heteronuclear pairs, HB^{5+}, HC^{6+}, $HeBe^{5+}$, HeB^{6+}, for N12-SX, rCAM-B3LYP, ωB97M-V, ωB97X, ωB97X-V, ωM05 with ωB97M(2) as a reference.

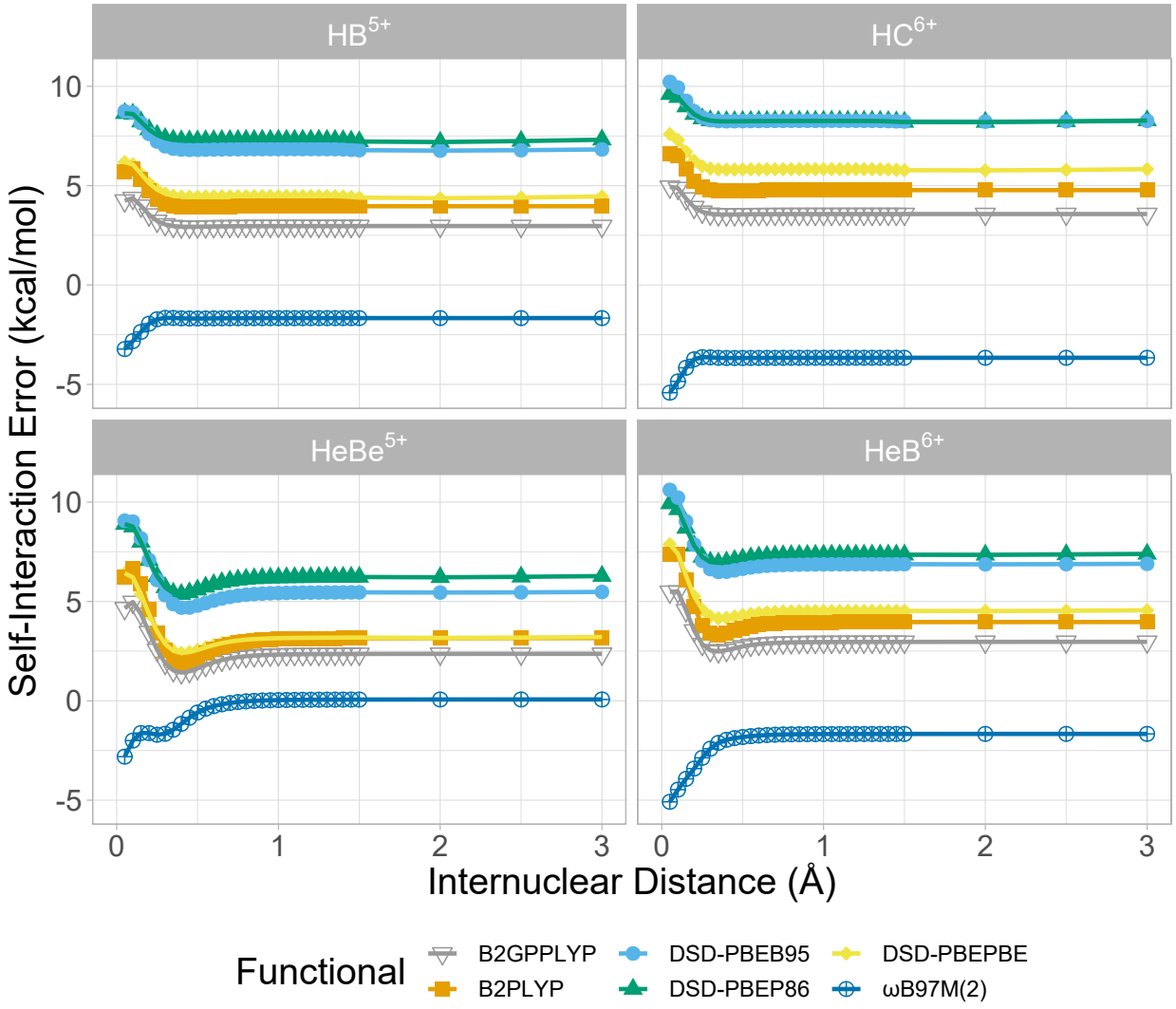

Figure S280: Self-interaction error vs internuclear distance of the heteronuclear pairs, HB^{5+}, HC^{6+}, $HeBe^{5+}$, HeB^{6+}, for B2GPPLYP, B2PLYP, DSD-PBEB95, DSD-PBEP86, DSD-PBEPBE with ωB97M(2) as a reference.

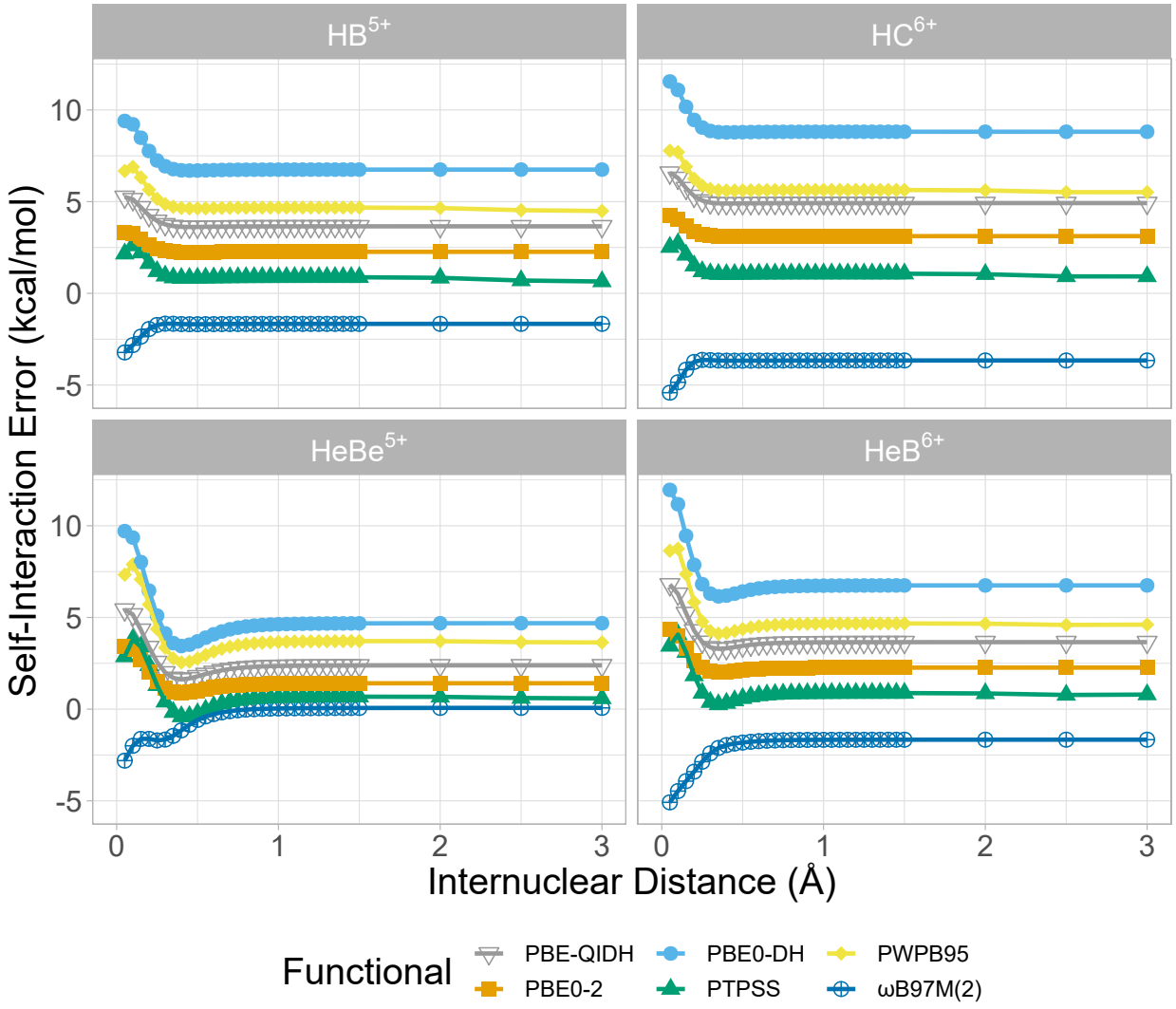

Figure S281: Self-interaction error vs internuclear distance of the heteronuclear pairs, HB^{5+}, HC^{6+}, $HeBe^{5+}$, HeB^{6+}, for PBE-QIDH, PBE0-2, PBE0-DH, PTPSS, PWPB95 with ωB97M(2) as a reference.

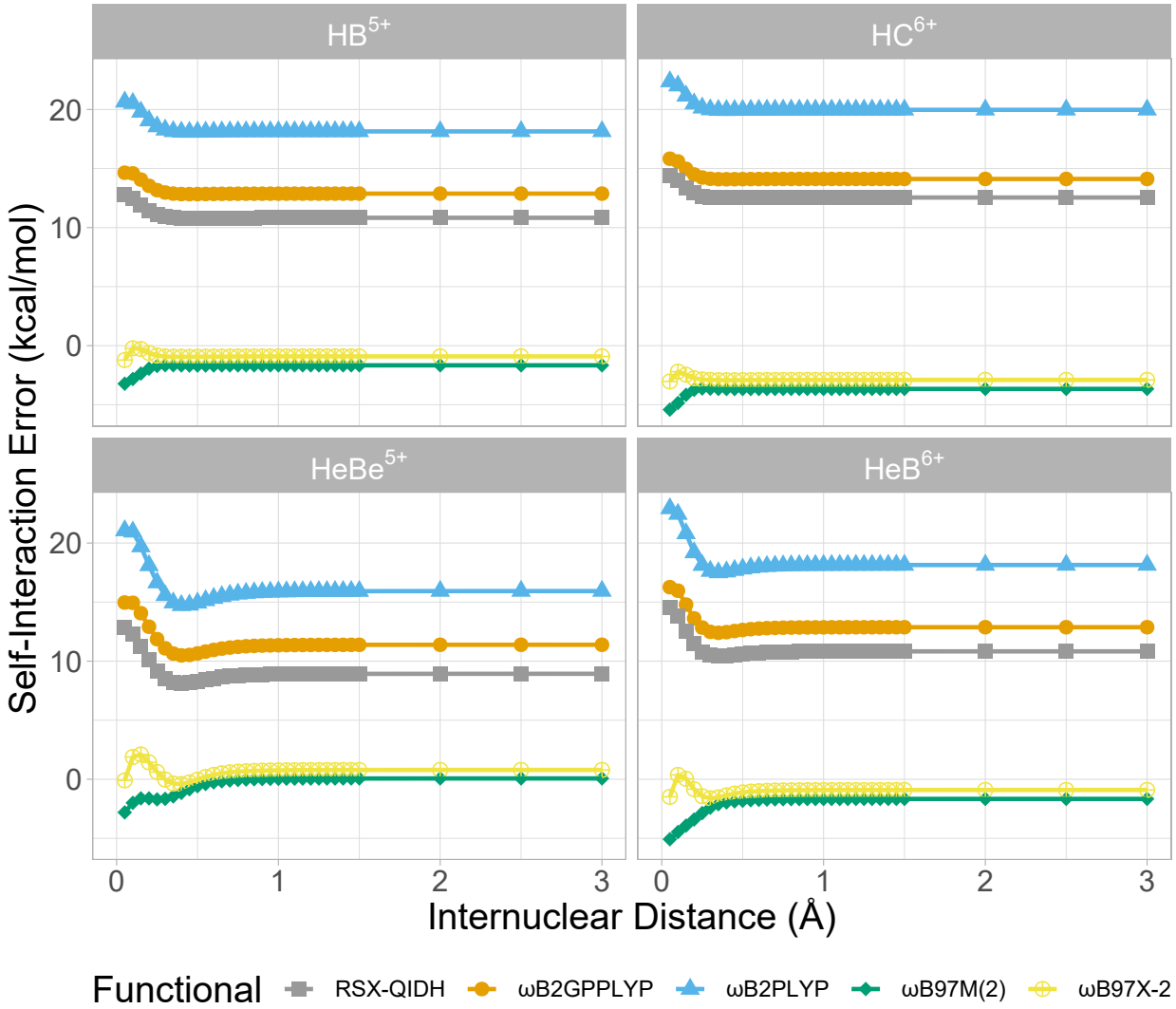

Figure S282: Self-interaction error vs internuclear distance of the heteronuclear pairs, HB^{5+}, HC^{6+}, $HeBe^{5+}$, HeB^{6+}, for RSX-QIDH, ωB2GPPLYP, ωB2PLYP, ωB97X-2 with ωB97M(2) as a reference.

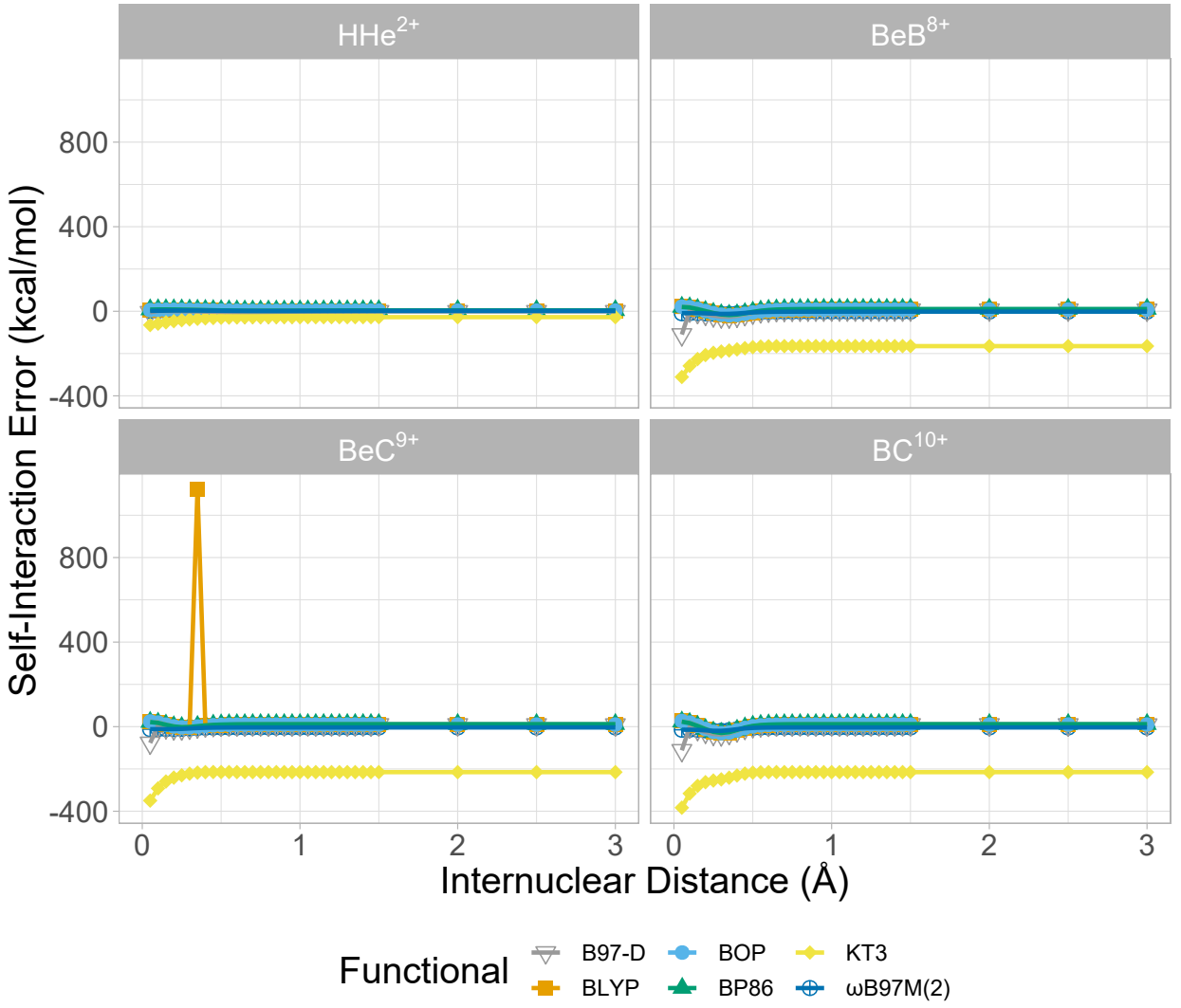

Figure S283: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, BeB^{8+}, BeC^{9+}, BC^{10+}, for B97-D, BLYP, BOP, BP86, KT3 with ωB97M(2) as a reference.

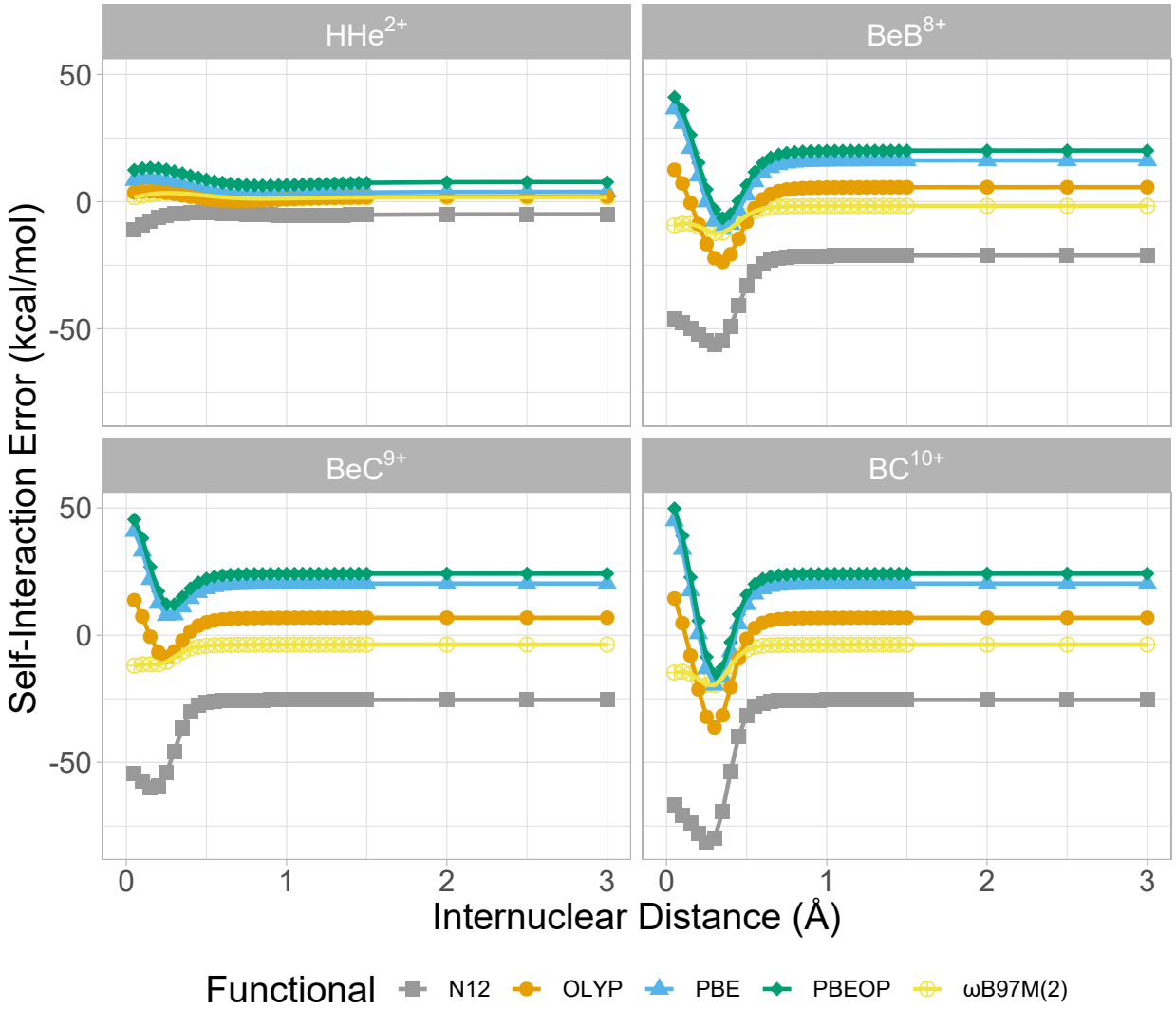

Figure S284: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, BeB^{8+}, BeC^{9+}, BC^{10+}, for N12, OLYP, PBE, PBEOP with ωB97M(2) as a reference.

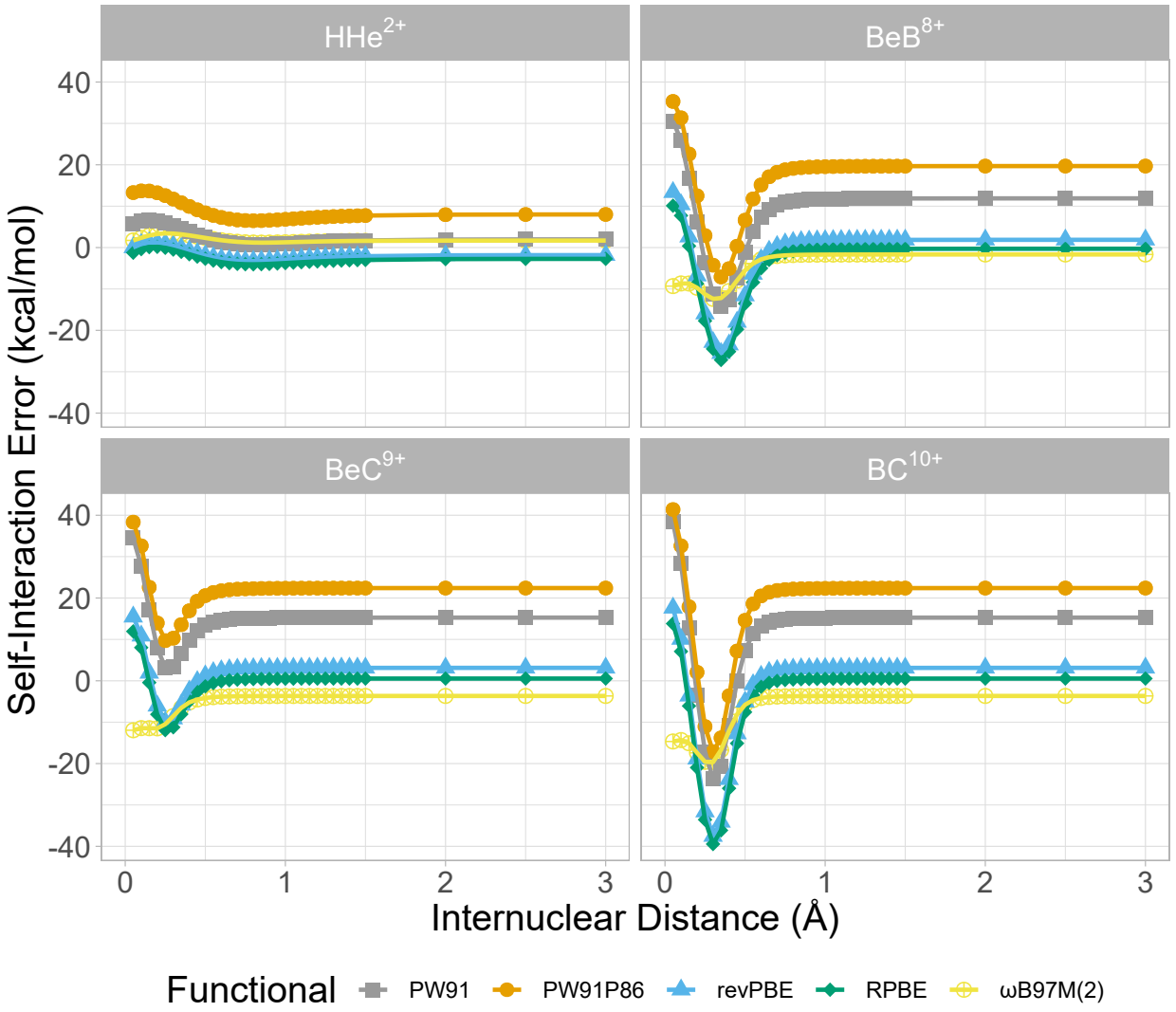

Figure S285: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, BeB^{8+}, BeC^{9+}, BC^{10+}, for PW91, PW91P86, revPBE, RPBE with ωB97M(2) as a reference.

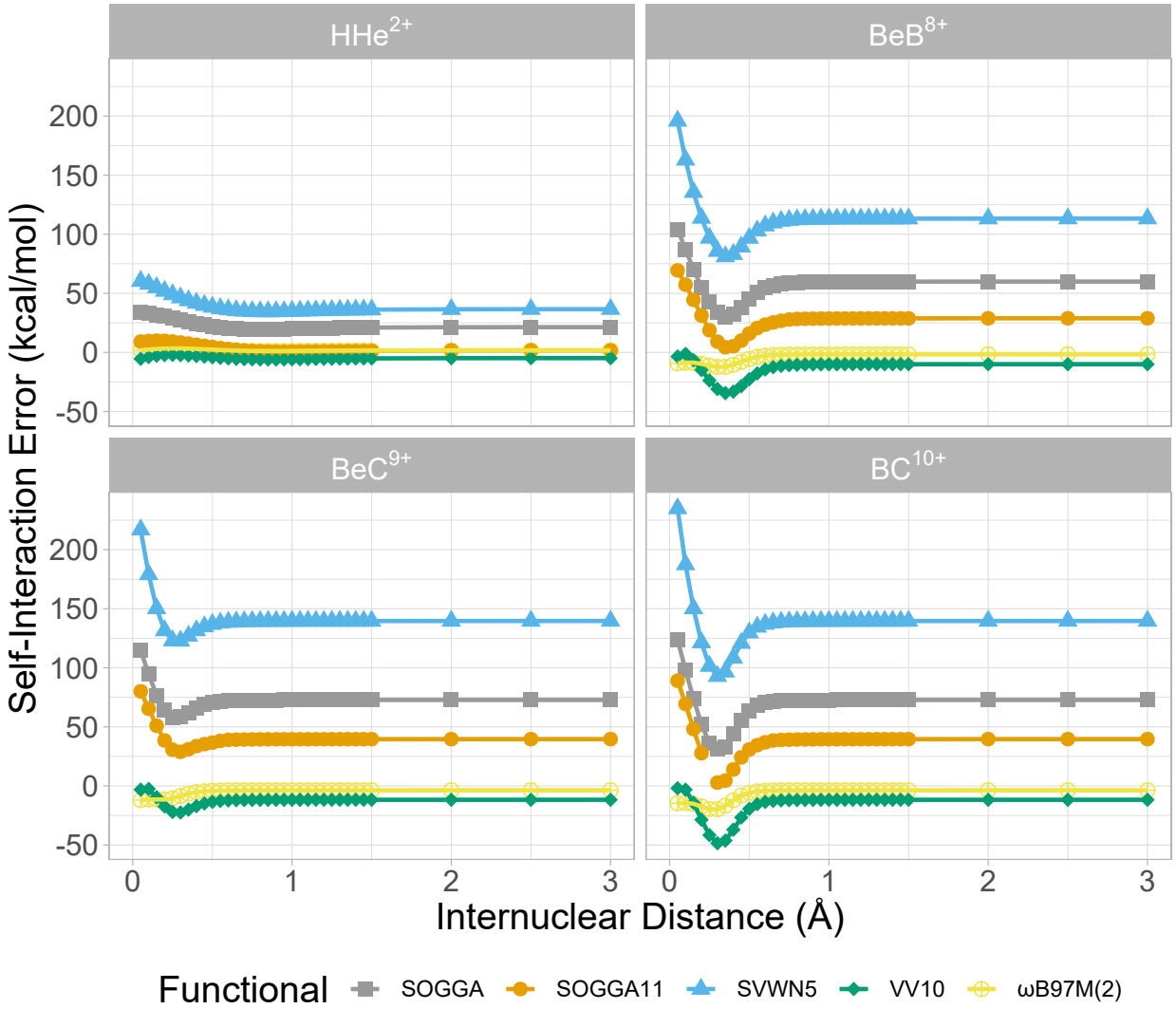

Figure S286: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, BeB^{8+}, BeC^{9+}, BC^{10+}, for SOGGA, SOGGA11, SVWN5, VV10 with ωB97M(2) as a reference.

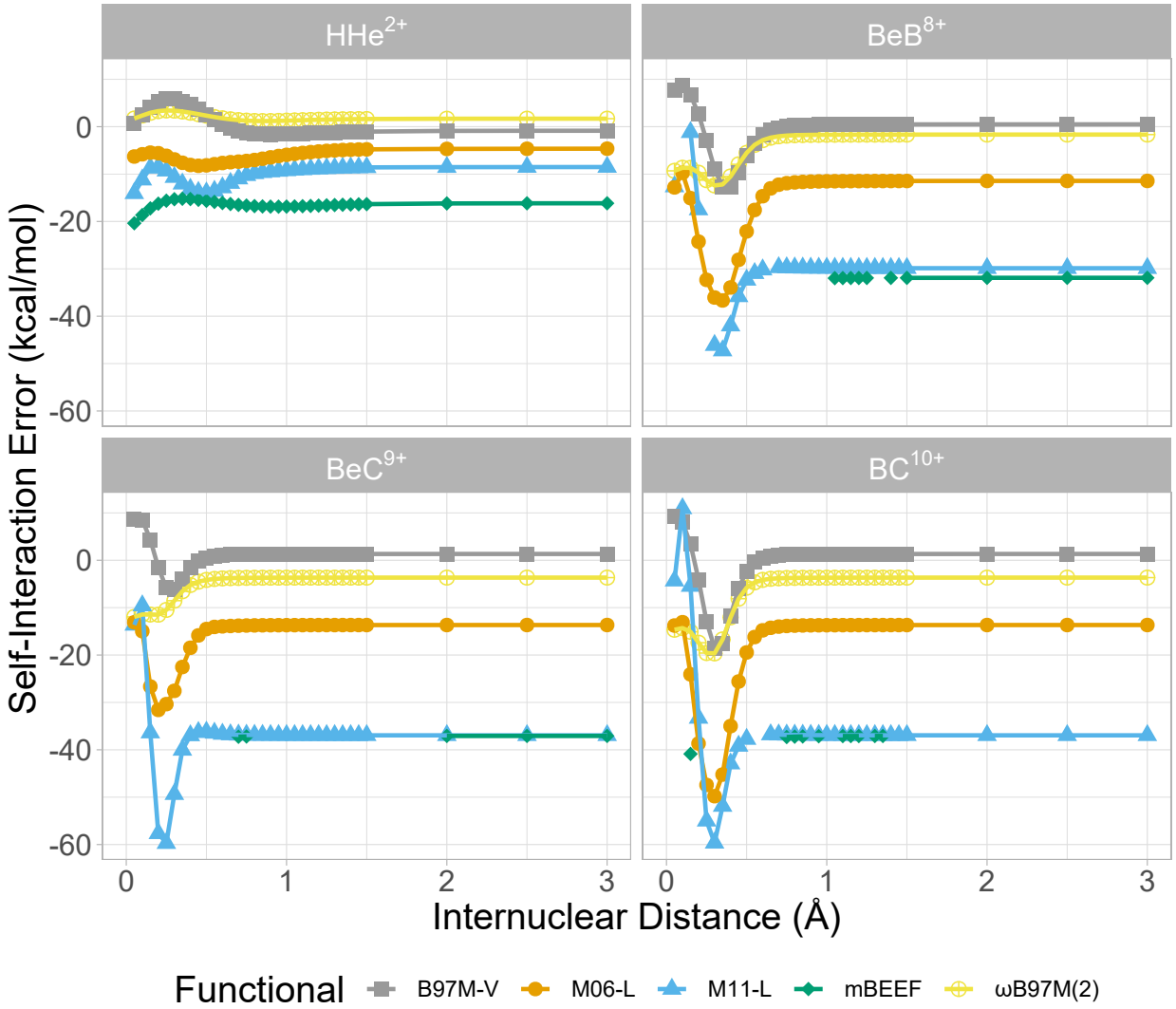

Figure S287: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, BeB^{8+}, BeC^{9+}, BC^{10+}, for B97M-V, M06-L, M11-L, mBEEF, MN12-L with ωB97M(2) as a reference.

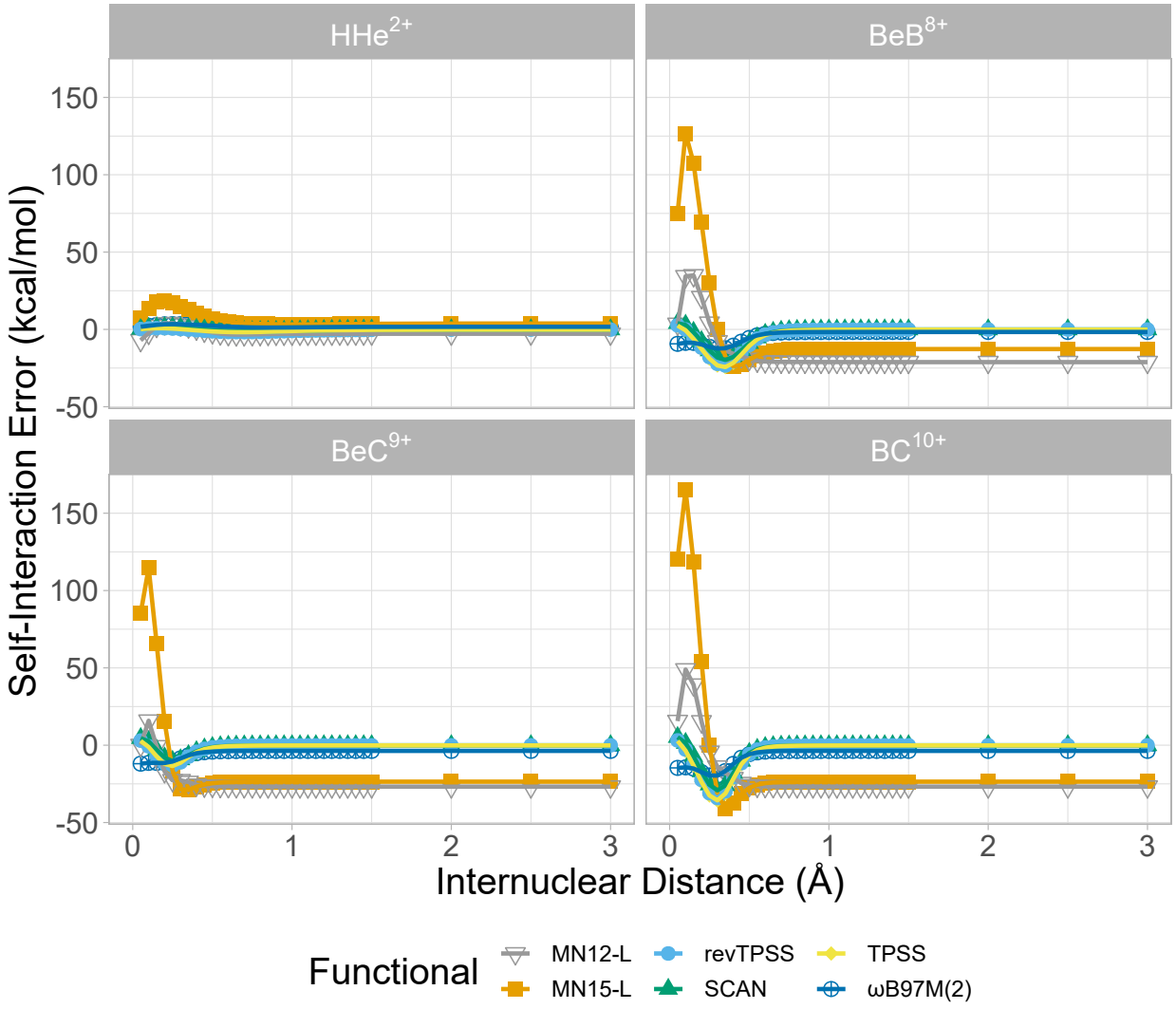

Figure S288: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, BeB^{8+}, BeC^{9+}, BC^{10+}, for MN15-L, revTPSS, SCAN, TPSS with ωB97M(2) as a reference.

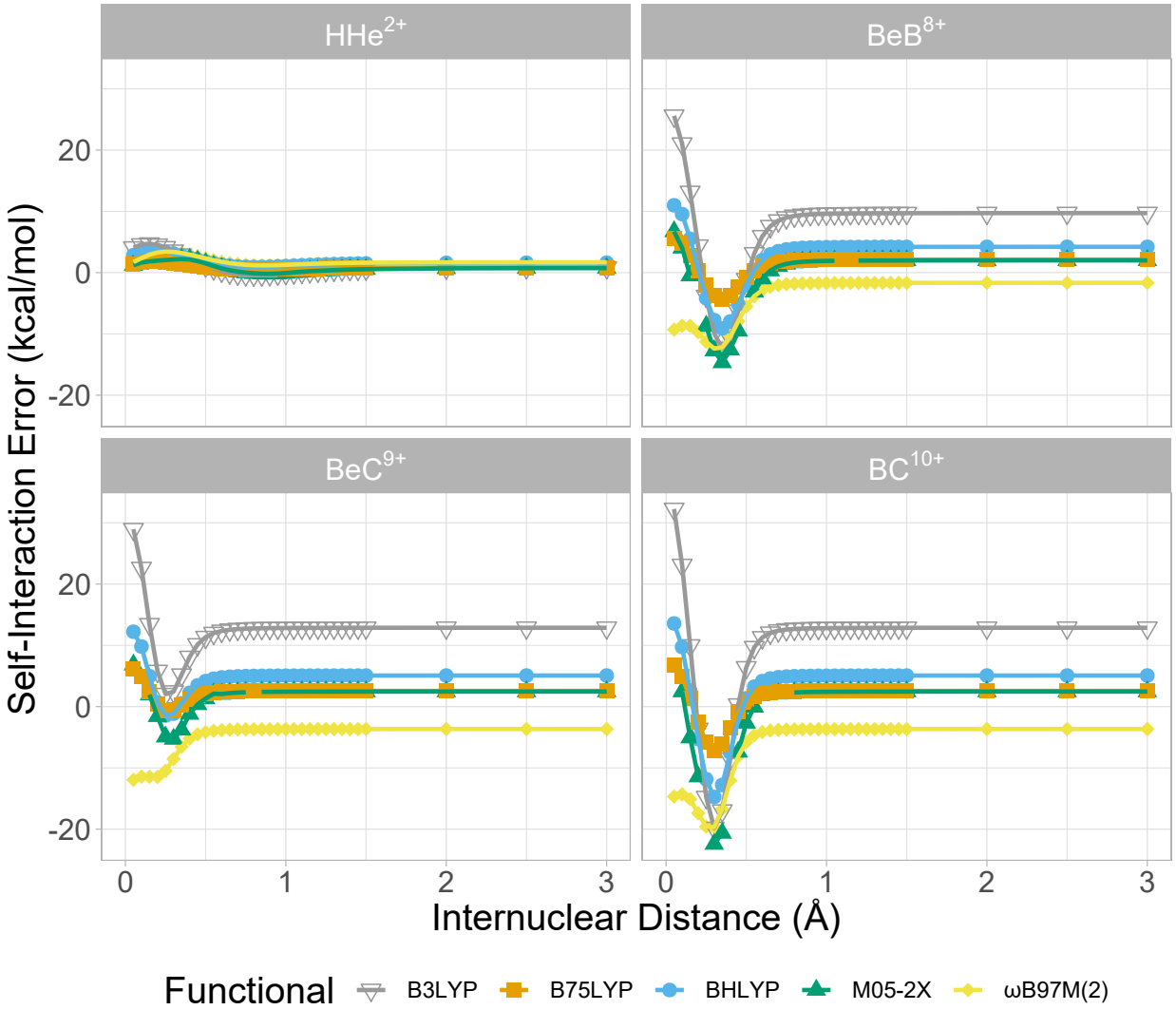

Figure S289: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, BeB^{8+}, BeC^{9+}, BC^{10+}, for B3LYP, B75LYP, B97, BHLYP, BMK with ωB97M(2) as a reference.

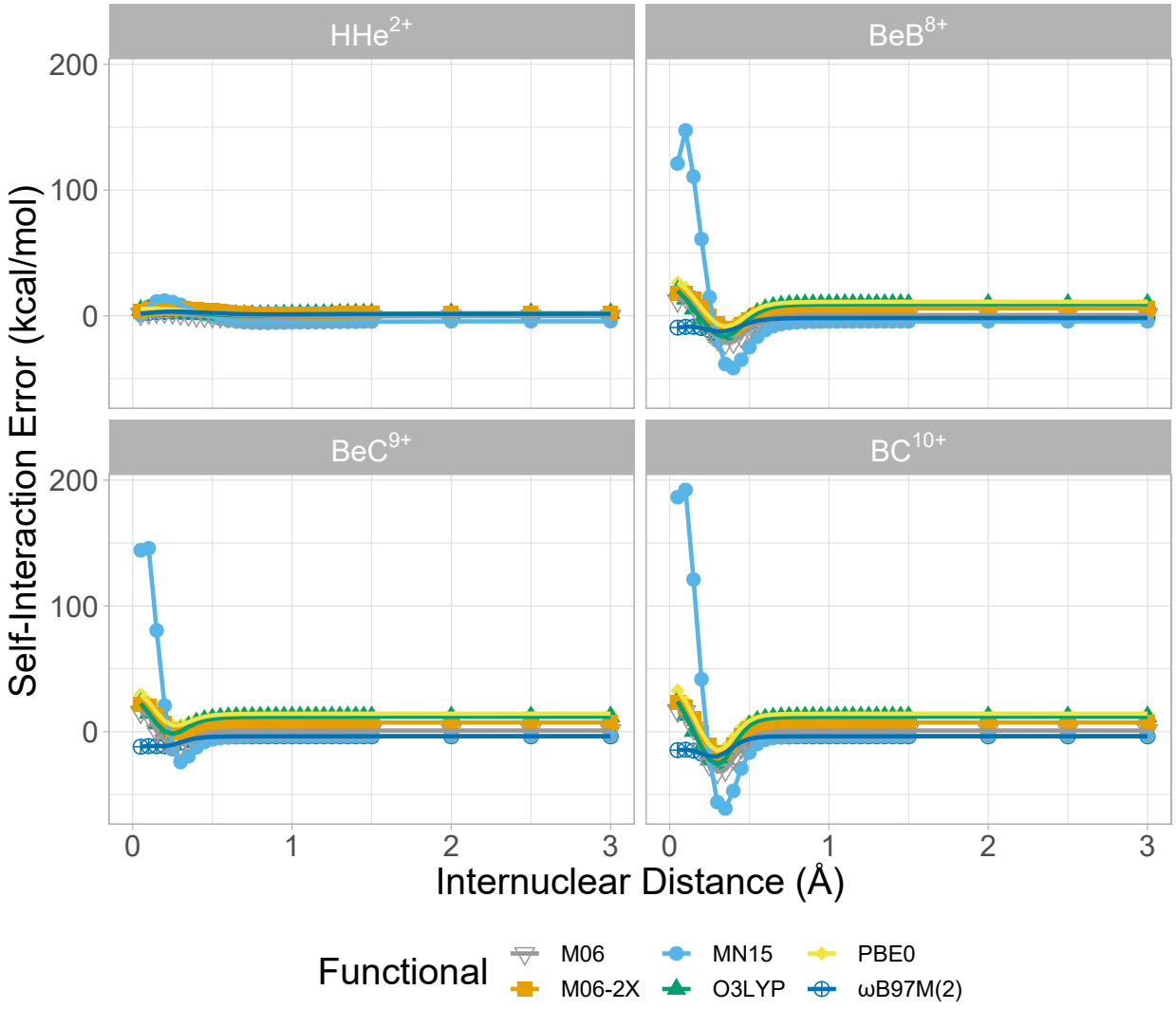

Figure S290: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, BeB^{8+}, BeC^{9+}, BC^{10+}, for M05-2X, M06, M06-2X, MN15, O3LYP with ωB97M(2) as a reference.

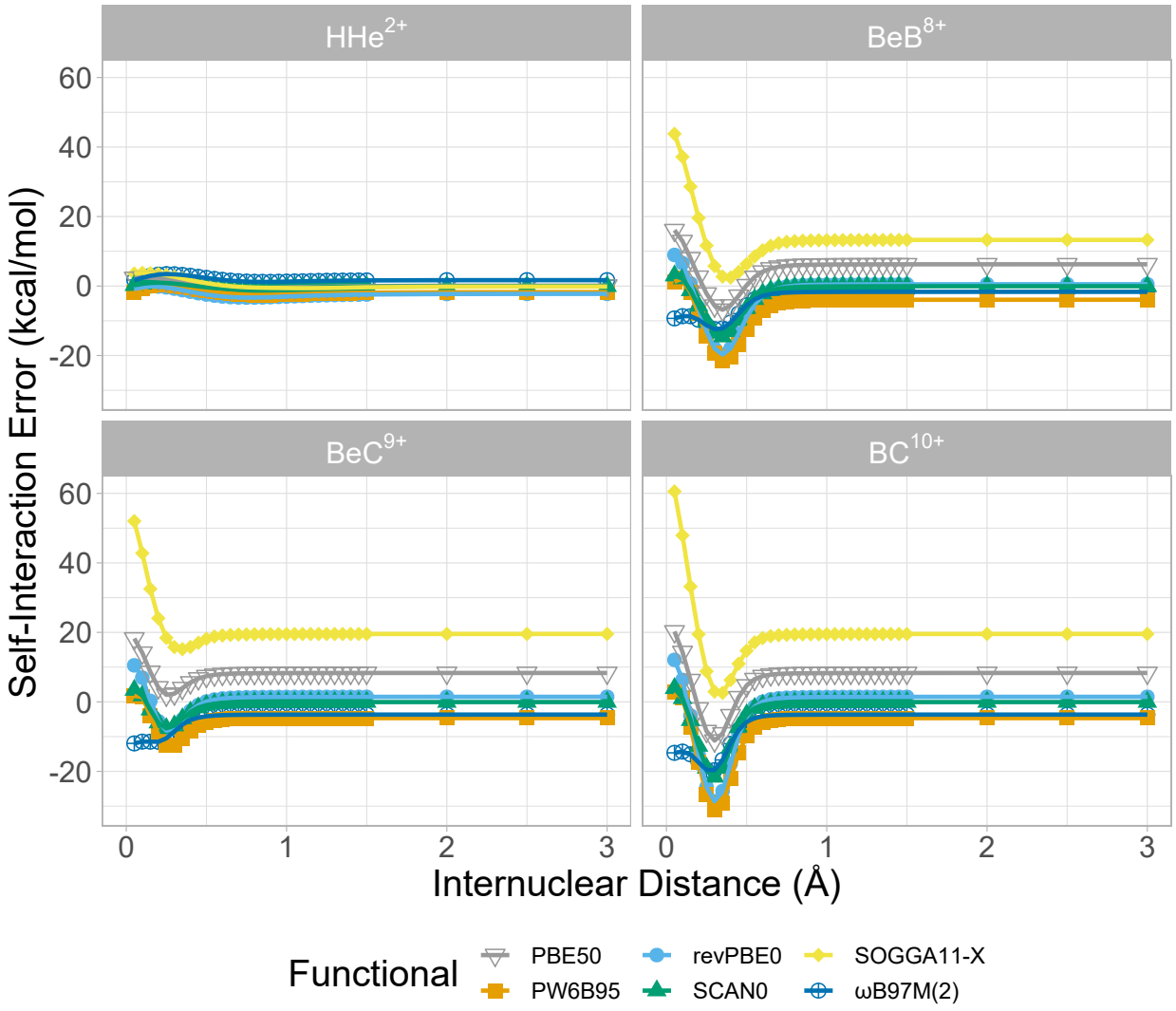

Figure S291: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, BeB^{8+}, BeC^{9+}, BC^{10+}, for PBE0, PBE50, PW6B95, revPBE0, SCAN0 with ωB97M(2) as a reference.

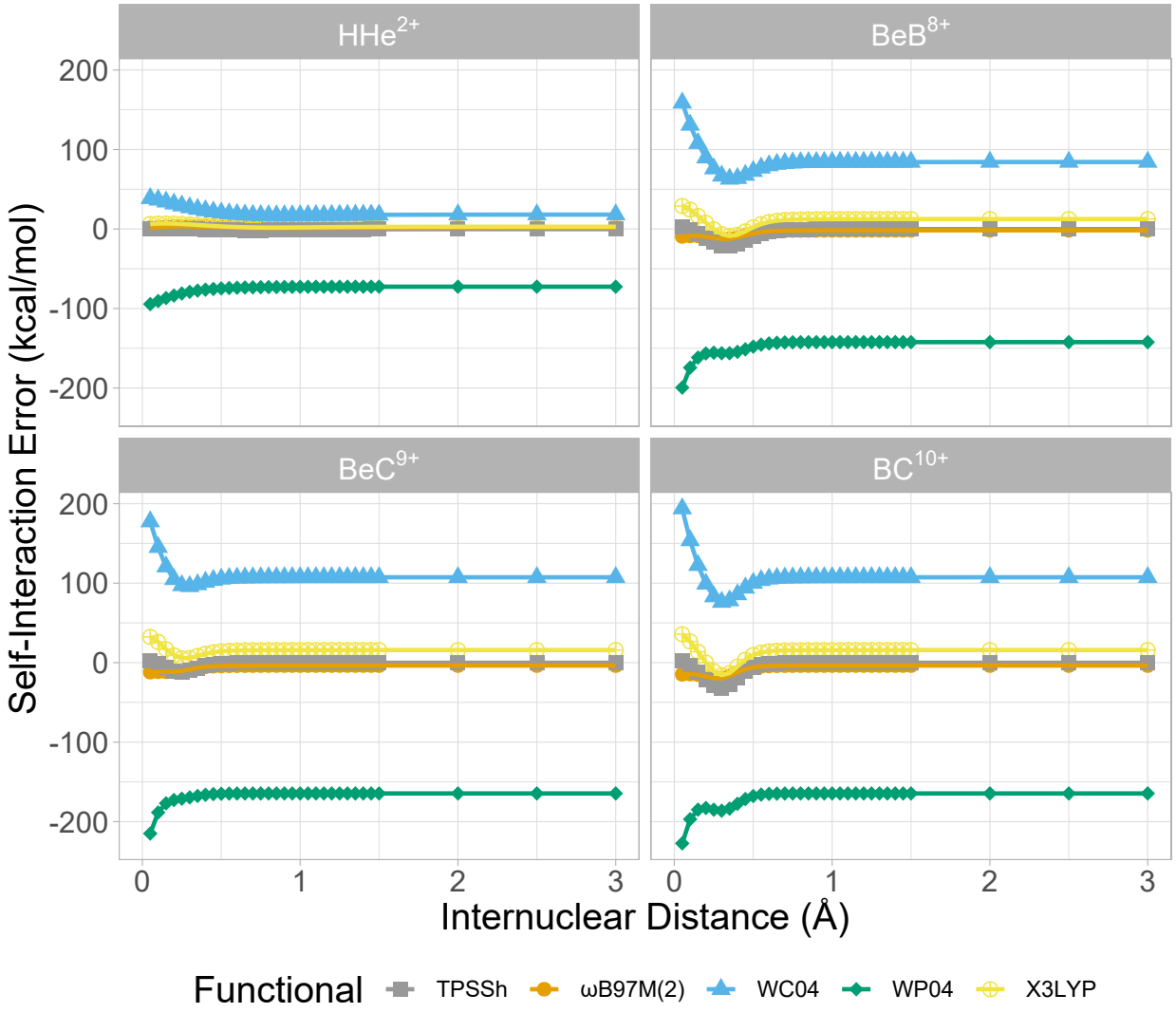

Figure S292: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, BeB^{8+}, BeC^{9+}, BC^{10+}, for SOGGA11-X, TPSSh, WC04, WP04, X3LYP with ωB97M(2) as a reference.

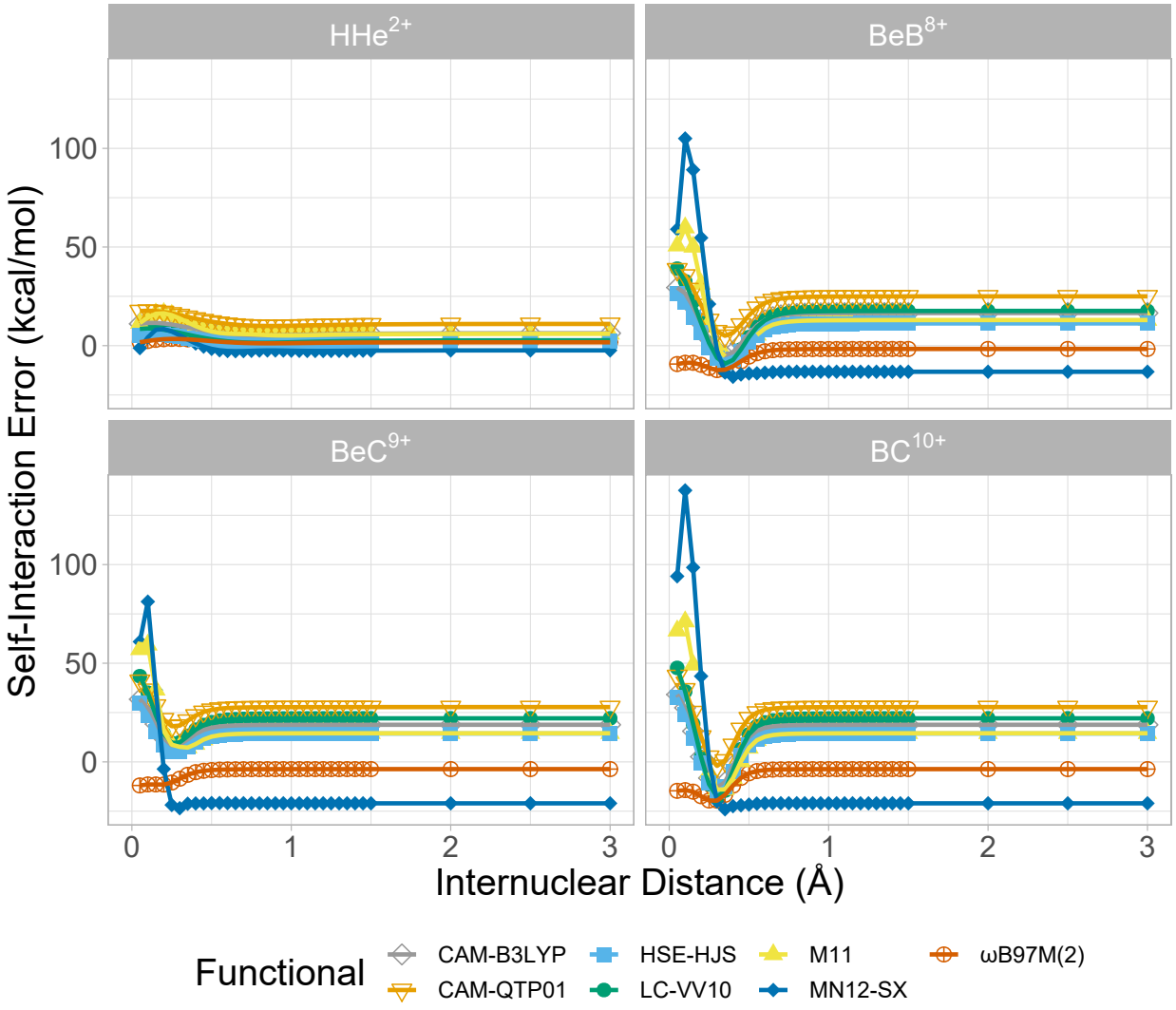

Figure S293: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, BeB^{8+}, BeC^{9+}, BC^{10+}, for CAM-B3LYP, CAM-QTP01, HSE-HJS, LC-VV10, M11, MN12-SX with ωB97M(2) as a reference.

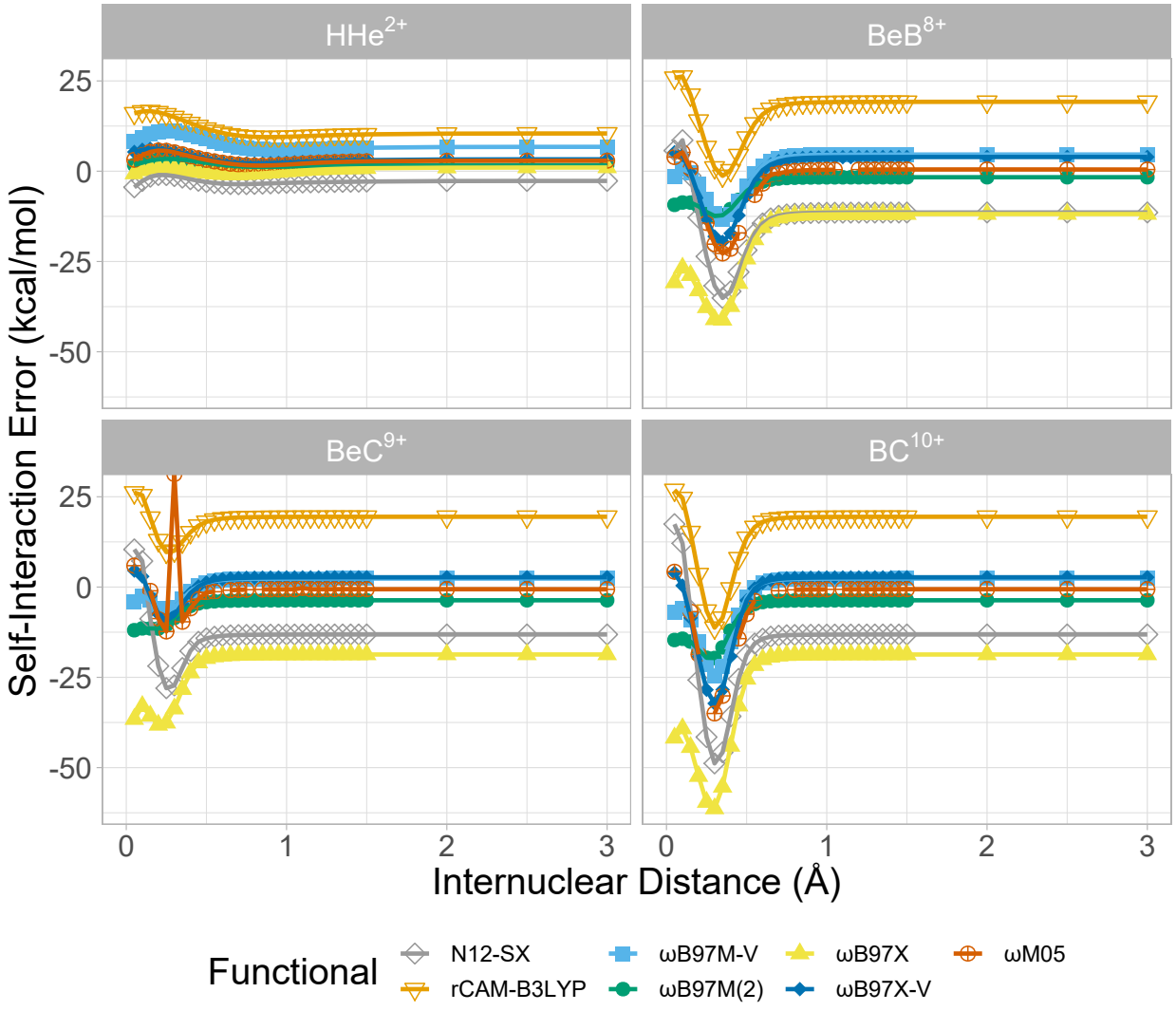

Figure S294: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, BeB^{8+}, BeC^{9+}, BC^{10+}, for N12-SX, rCAM-B3LYP, ωB97M-V, ωB97X, ωB97X-V, ωM05 with ωB97M(2) as a reference.

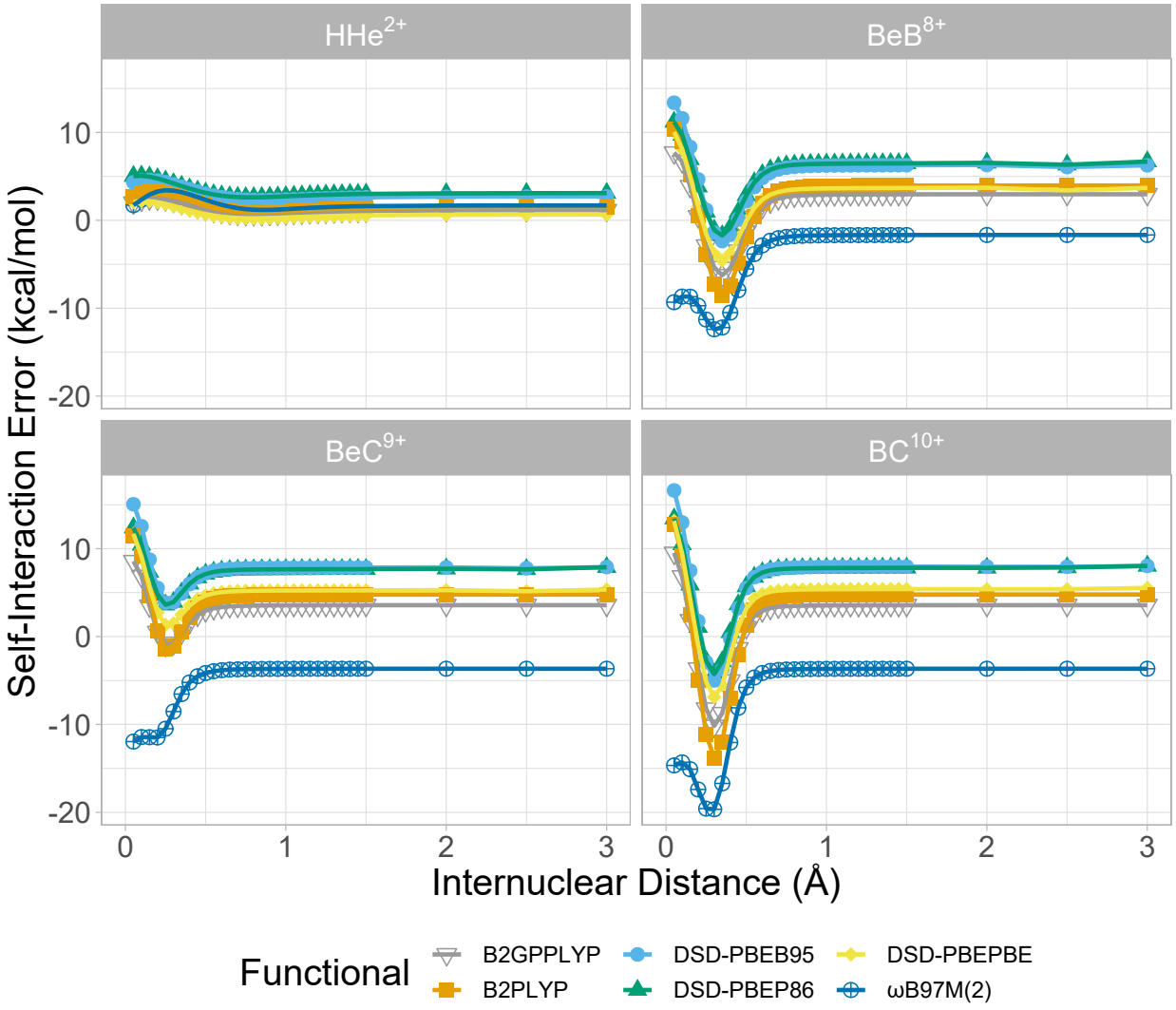

Figure S295: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, BeB^{8+}, BeC^{9+}, BC^{10+}, for B2GPPLYP, B2PLYP, DSD-PBEB95, DSD-PBEP86, DSD-PBEPBE with ωB97M(2) as a reference.

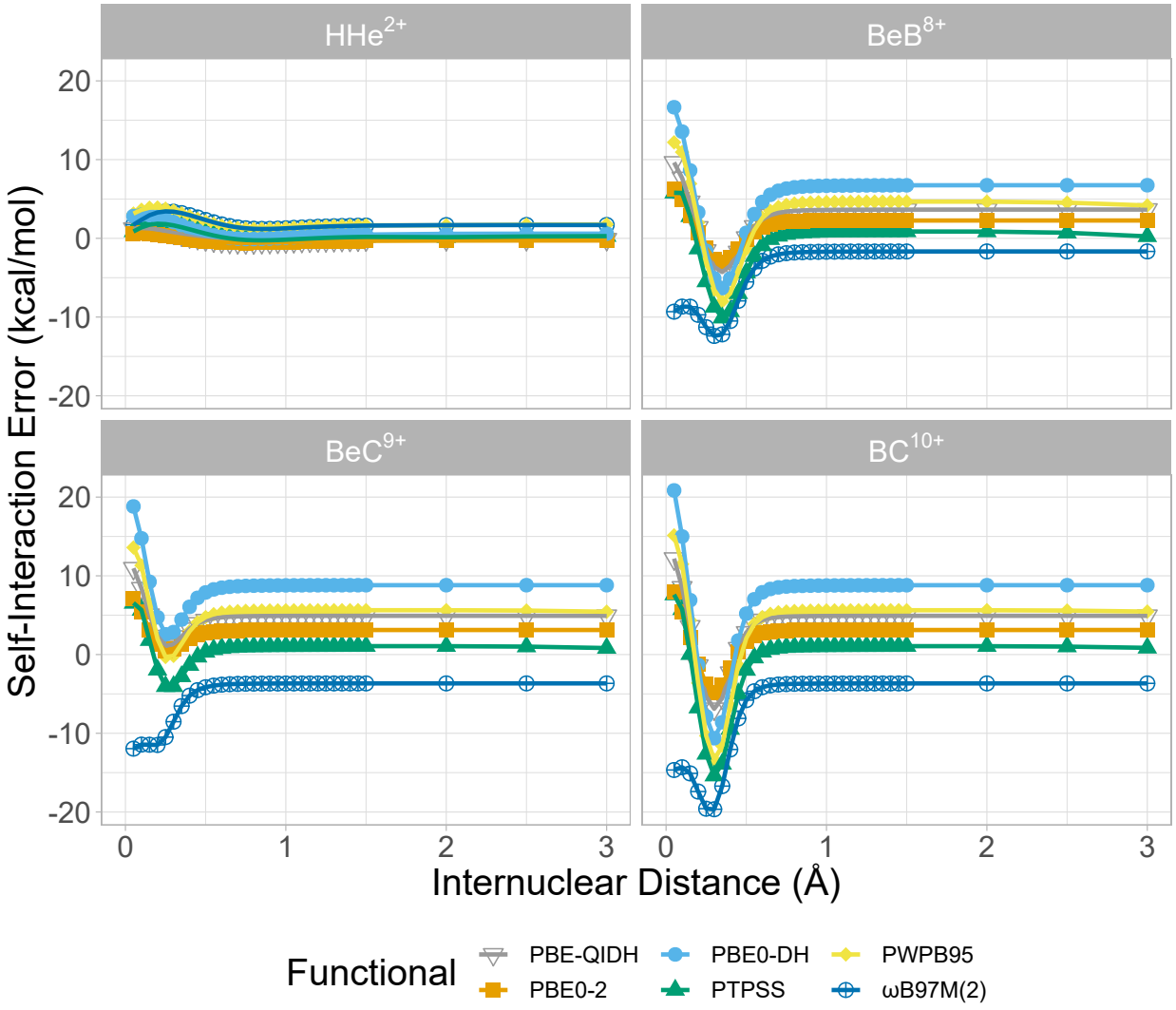

Figure S296: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, BeB^{8+}, BeC^{9+}, BC^{10+}, for PBE-QIDH, PBE0-2, PBE0-DH, PTPSS, PWPB95 with ωB97M(2) as a reference.

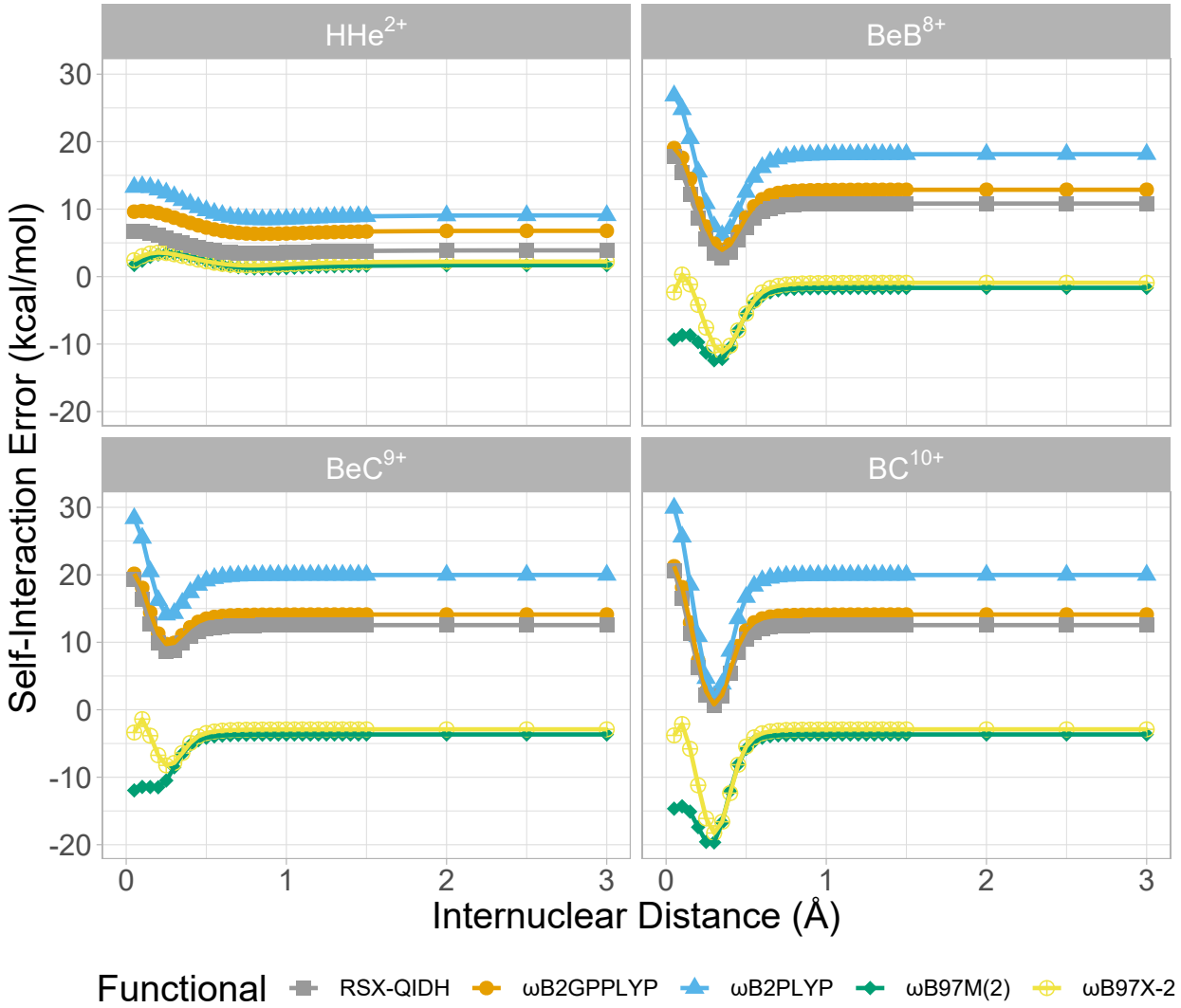

Figure S297: Self-interaction error vs internuclear distance of the heteronuclear pairs, HHe^{2+}, BeB^{8+}, BeC^{9+}, BC^{10+}, for RSX-QIDH, ωB2GPPLYP, ωB2PLYP, ωB97X-2 with ωB97M(2) as a reference.

SI.3.8 Heteronuclear Density and Functional error

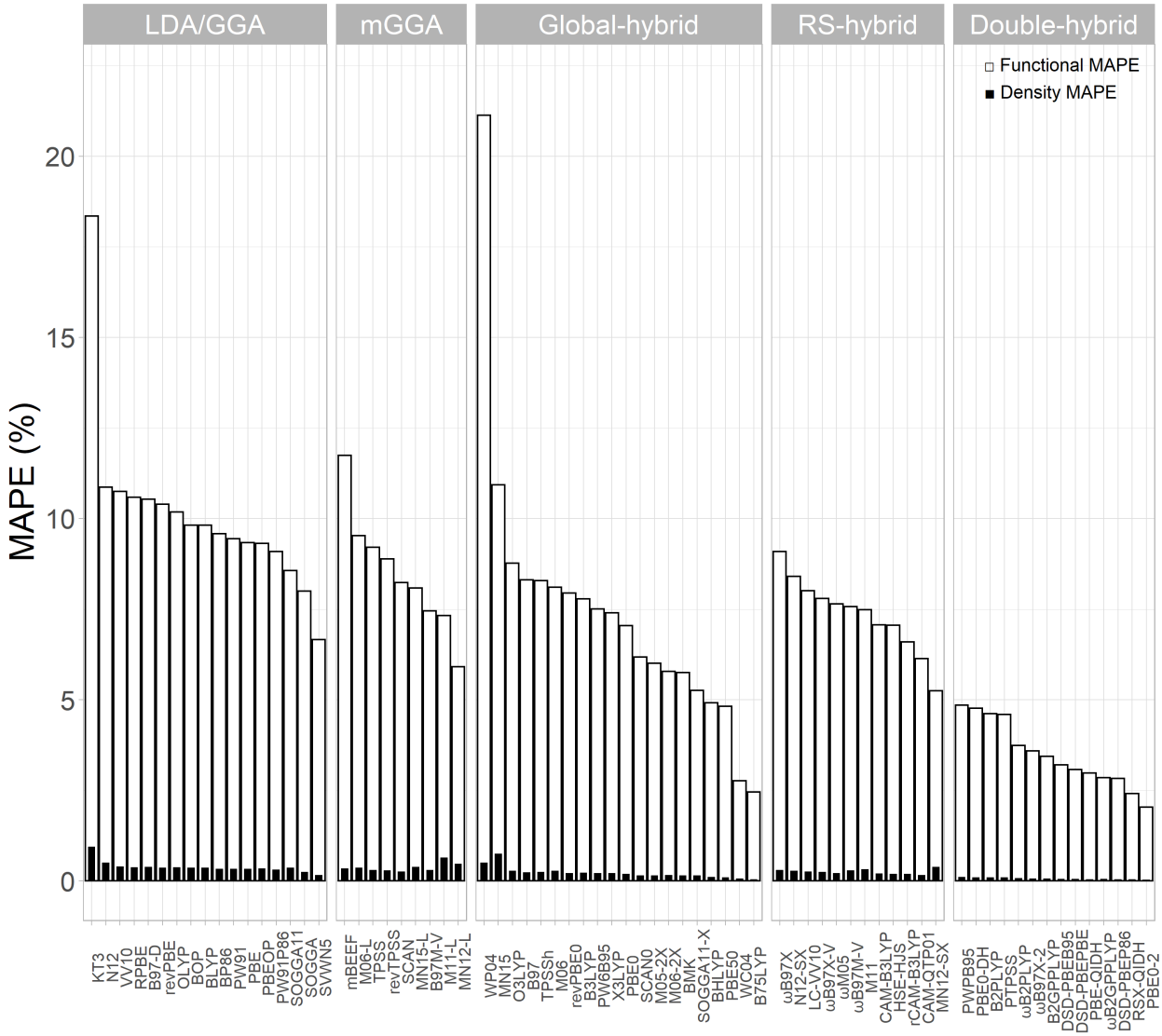

Figure S298: Functional (white) and density (black) mean absolute percentage errors across the heteronuclear diatomic dataset, from HHe_2^{2+} to BC_2^{10+}, for each density functional approximation.

SI.4 Correlation between SIE and fractional electron number

Firstly, we plot the total energy vs the fractional electron number for several DFAs and mononuclear ions in Fig. S299 which includes fractional electron number greater than 1. The convex shapes are difficult to see for the higher charged systems, but we can assure that they are not strictly linear with fractional electron number.
We then wanted to probe if there was a connection between the magnitude of the SIE, the fractional electron number, and the following scaling relation as discussed in section 4.3 of the paper:

$$E_{XC}[q\rho] - q^2 E_{XC}[\rho] = 0 \tag{4}$$

At a fractional electron number of q=0.5 there is a maximum deviation from eqn. 4, so we chose this value to compare with our integer electron systems. There appears to be little correlation between the SIE of the integer electron and the maximum deviation from eqn. 4 as per Fig. S300. However, we see much better correlation between the SIE at a fractional electron number of q=0.5 and the maximum deviation from eqn. 4 as seen in Fig. S301 as well as the difference between SIE(q=0.5) and SIE(q=1.0) vs the same maximum deviation in Fig. S302. In the latter case we note that the SIE at q=0.5 is normally much larger in magnitude than for SIE at q=1.0, which leads to the fractional SIE value dominating that equation. Overall we see there appears to be connection between the SIE at a particular fractional electron number and the value given by the scaling relation in eqn. 4 in that a smaller value for the scaling relation leads to a smaller SIE. We will not conduct a formal statistical analysis with this dataset as we believe that should be accompanied by a deeper theoretical analysis of the underlying exchange-correlation functionals with special attention given the outliers and why they do not behave as the majority of DFAs do.
Given homonuclear diatomics delocalise to yield a system of fractional electron number of q=0.5, we decided to include an additional analysis to see if there was a connection between a fractionally charged atom, the equivalent hydrogenic homonuclear diatomic, and eqn. 4.
Across Figs. S303 - S306 we can see that for each hydrogenic mono- and di-nuclear system there does seem to be a connection between satisfying the relation of eqn. 4 and the SIE of the homonuclear system at a large internuclear separation. Likewise, we refrain from formally correlating the two quantities here.

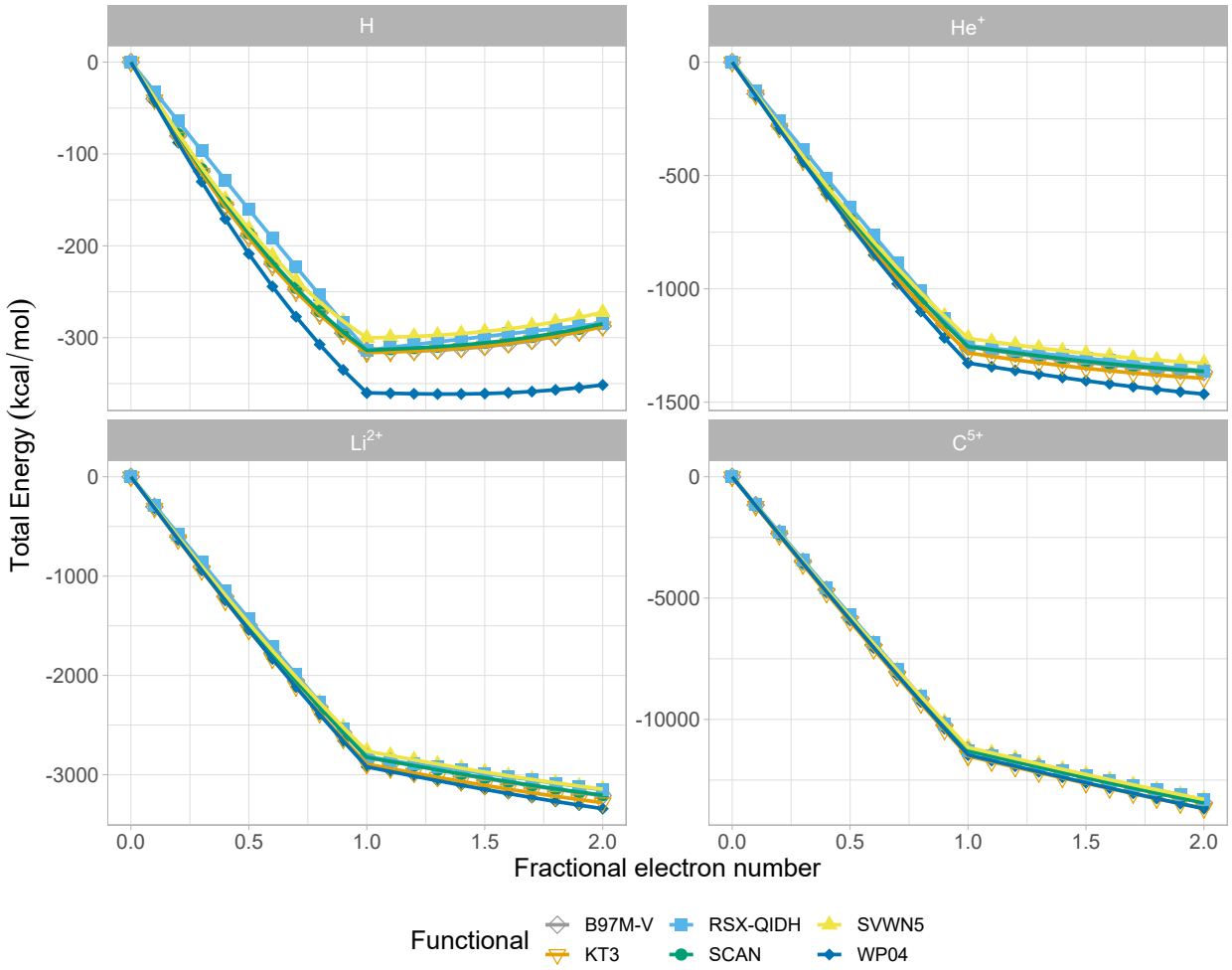

Figure S299: Total energy vs fractional electron number for the following DFAs: B97M-V, KT3, RSX-QIDH, SCAN, SVWN5, and WP04.

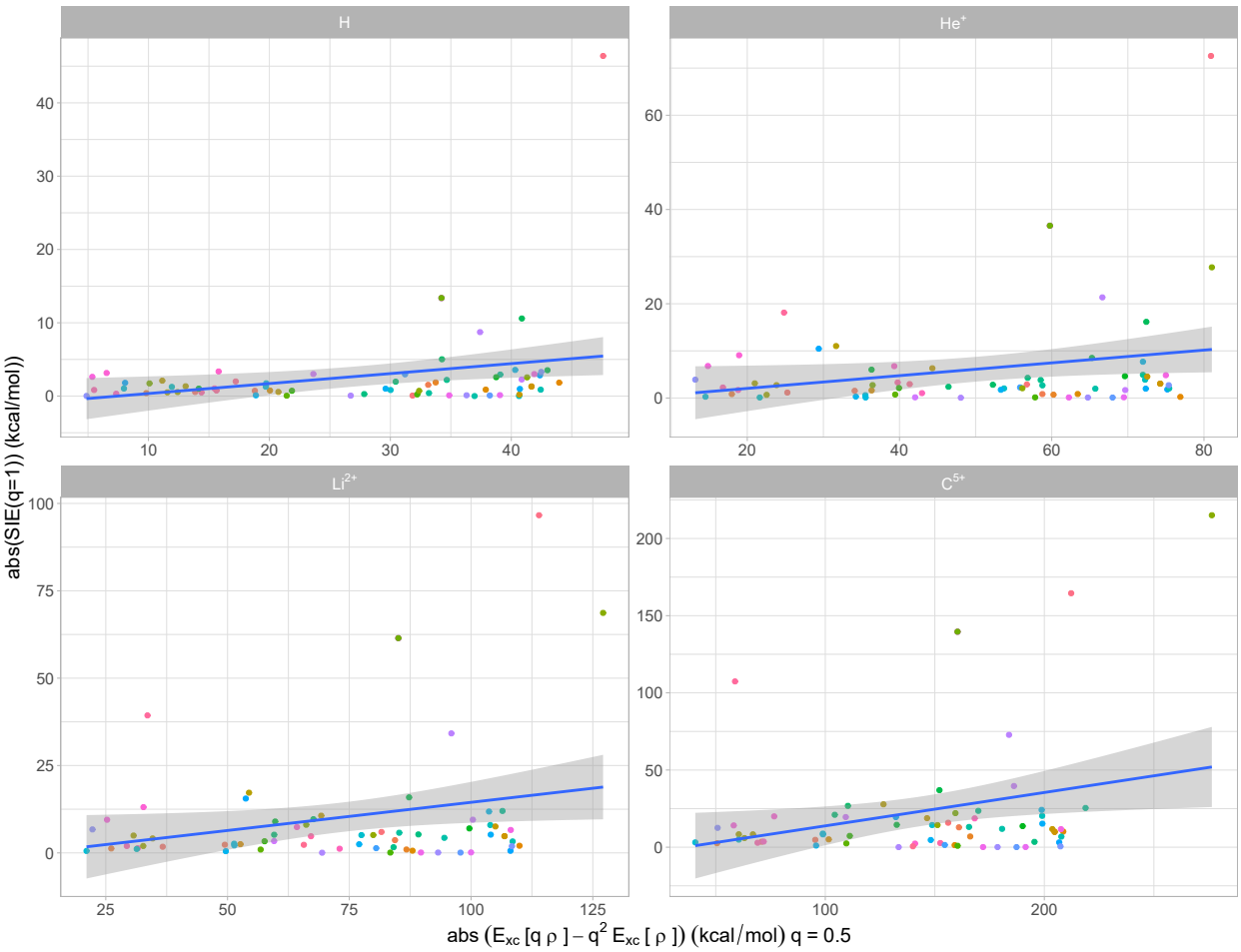

Figure S300: The absolute value of the self-interaction error of the integer electron system vs the absolute value of eqn. 4 at a fractional electron number of q=0.5 for all tested density functional approximations.

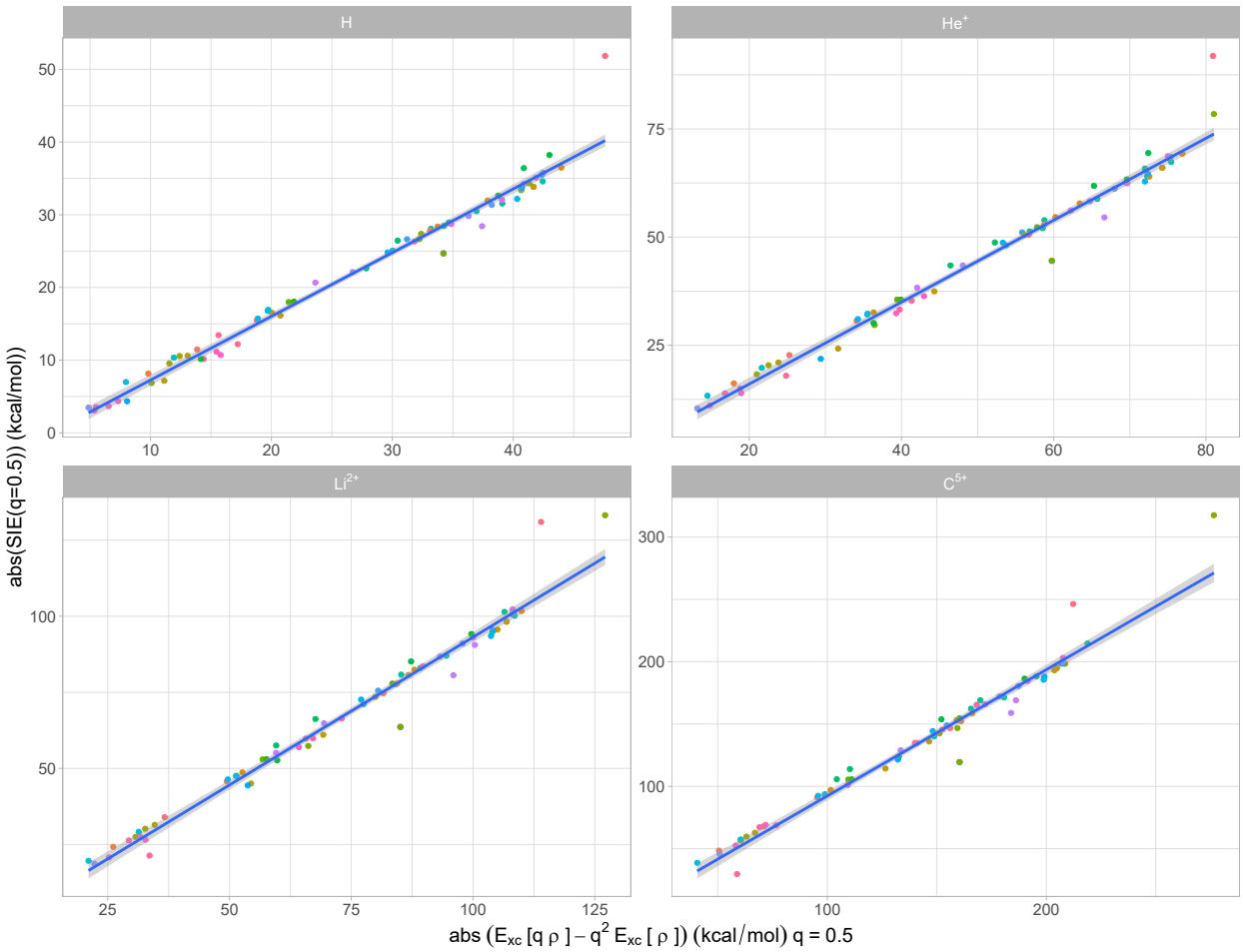

Figure S301: The absolute value of the self-interaction error at a fractional electron number of q=0.5 vs the absolute value of eqn. 4 at a fractional electron number of q=0.5 for all tested density functional approximations.

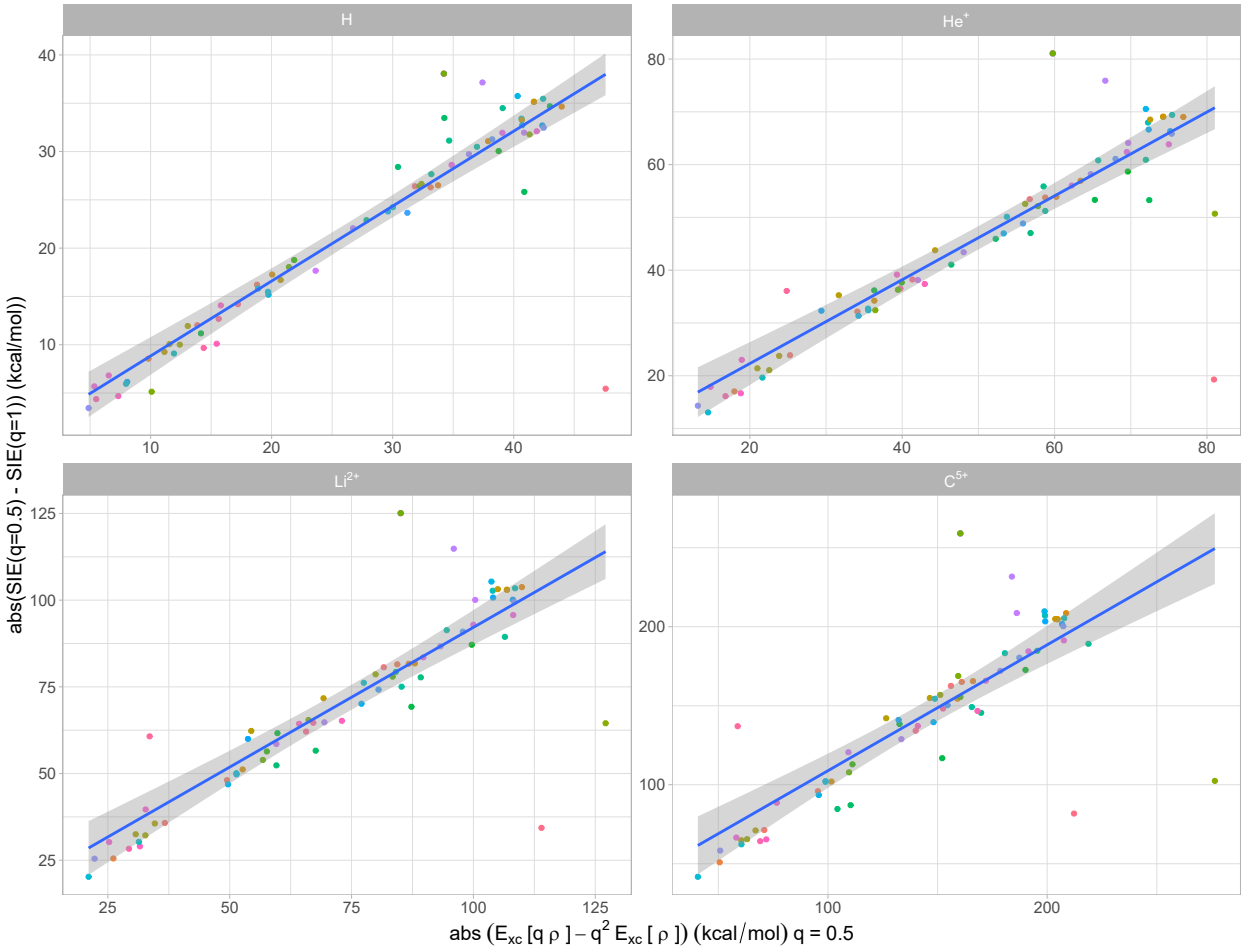

Figure S302: The absolute value of the difference between the self-interaction error at fractional electron number q=0.5 and SIE at q=1.0 vs the absolute value of eqn. 4 at a fractional electron number of q=0.5 for all tested density functional approximations.

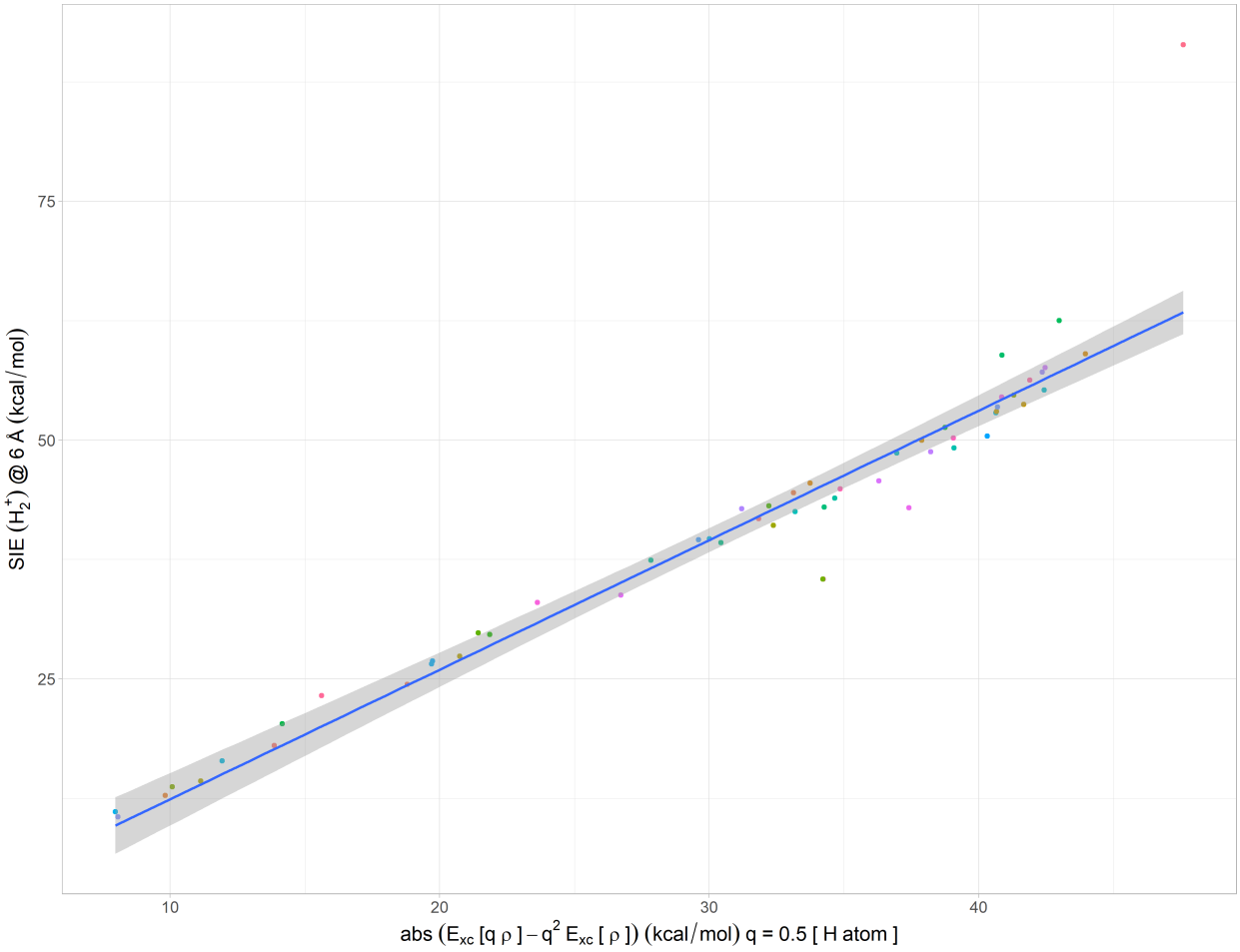

Figure S303: Self-interaction error of the H_2^+ diatomic at an internuclear separation of 6 Å vs the absolute value of eqn. 4 at a fractional electron number of q=0.5 for the hydrogen atom across all tested density functional approximations.

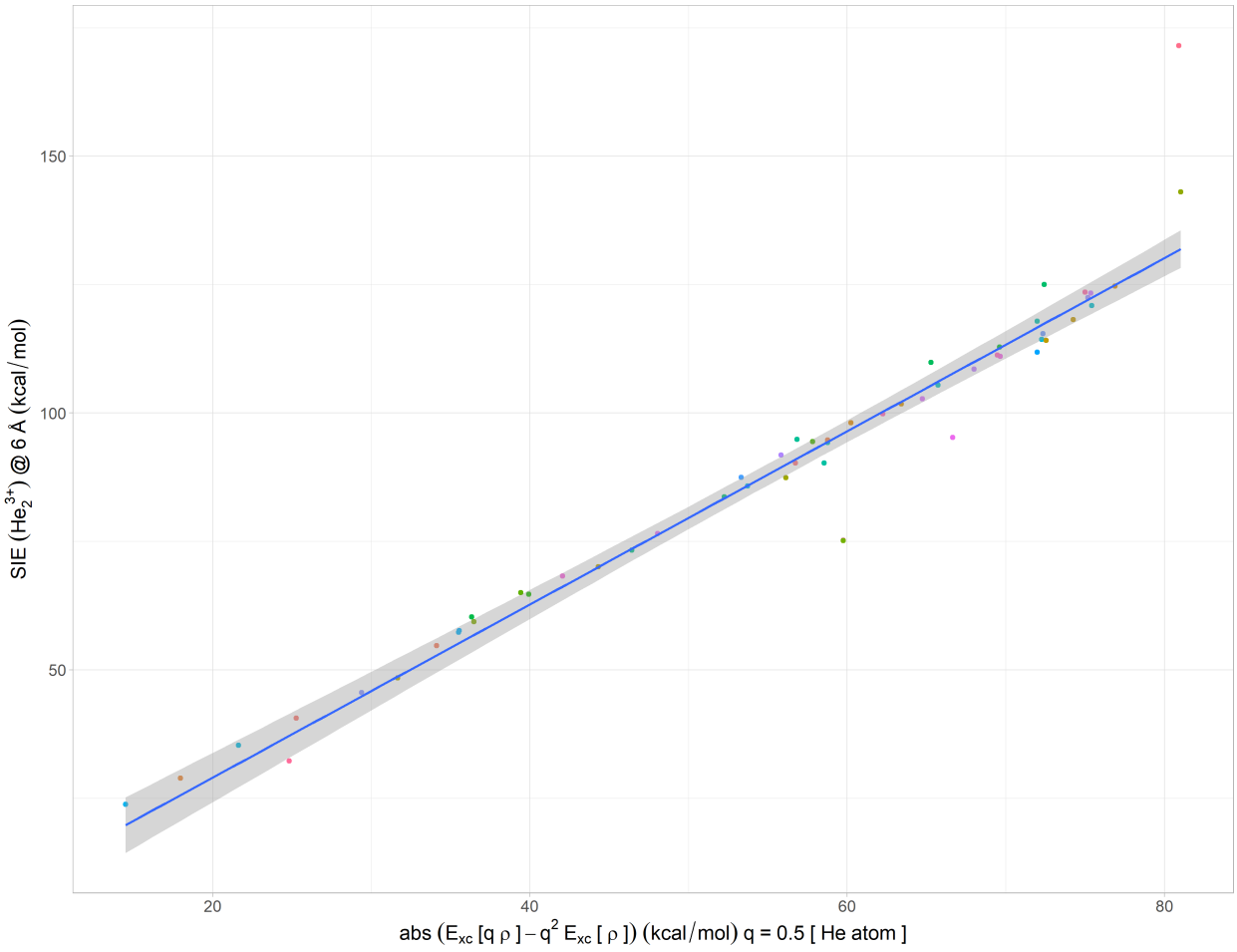

Figure S304: Self-interaction error of the He_2^{3+} diatomic at an internuclear separation of 6 Å vs the absolute value of eqn. 4 at a fractional electron number of q=0.5 for the helium atom across all tested density functional approximations.

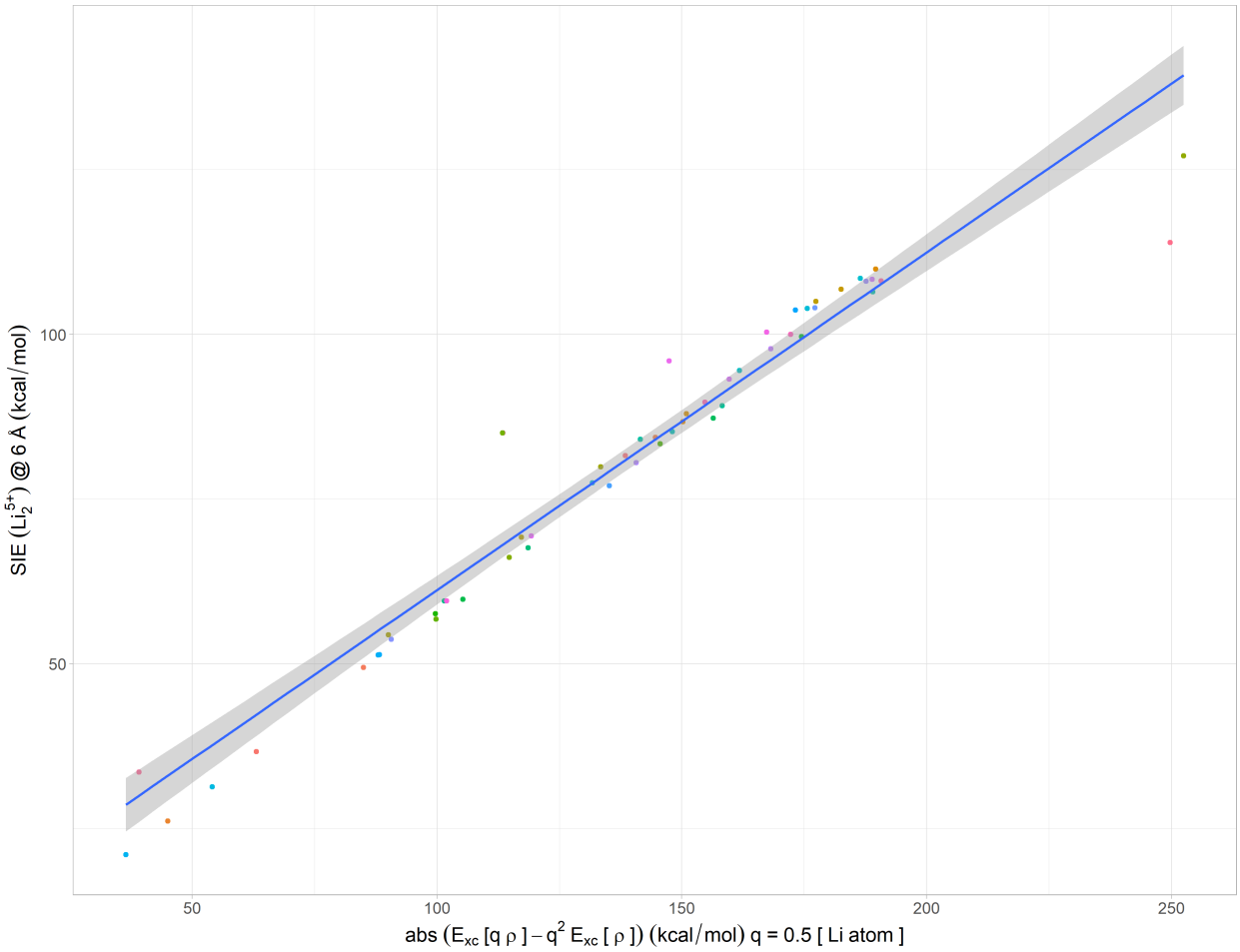

Figure S305: Self-interaction error of the Li_2^{5+} diatomic at an internuclear separation of 6 Å vs the absolute value of eqn. 4 at a fractional electron number of q=0.5 for the lithium atom across all tested density functional approximations.

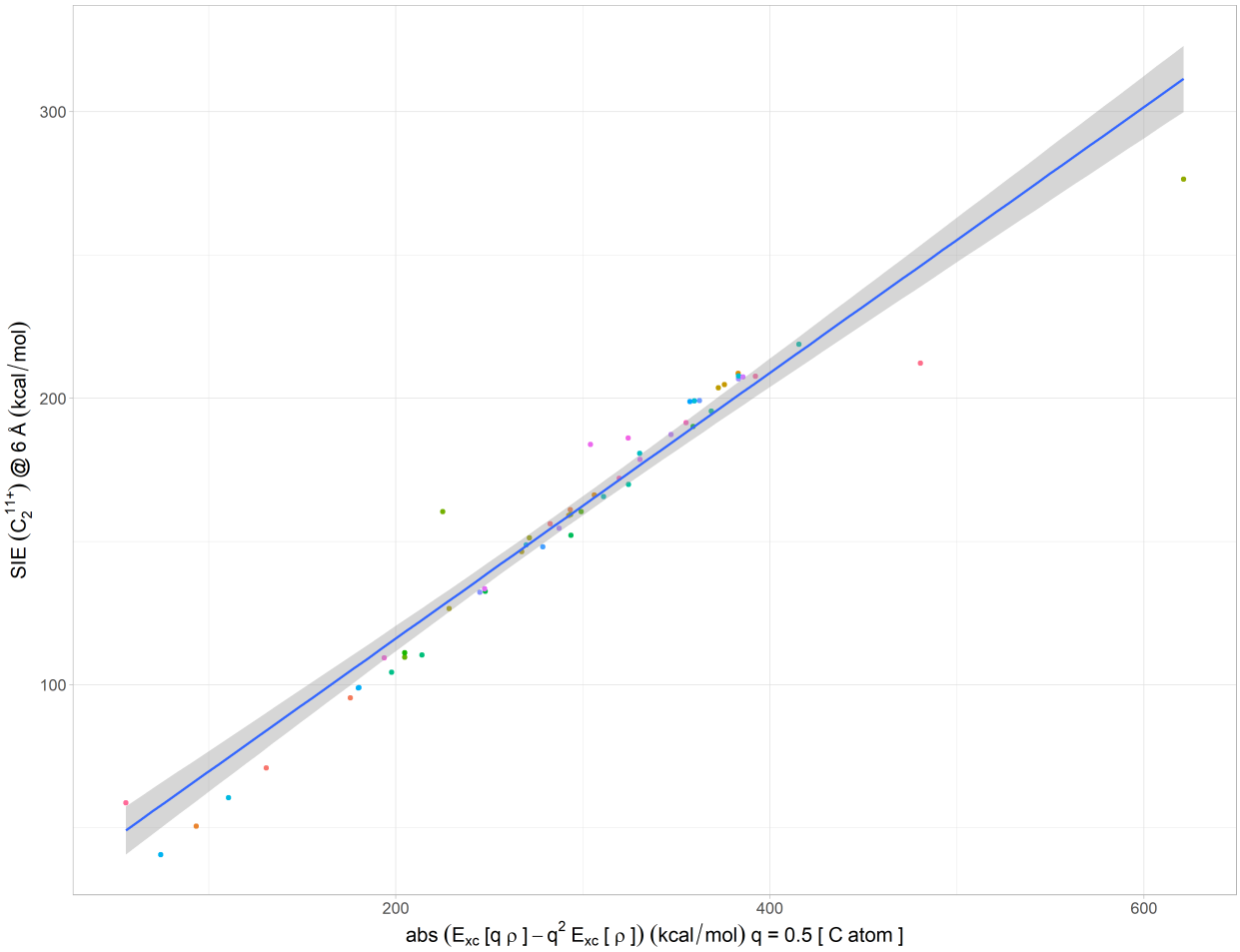

Figure S306: Self-interaction error of the C_2^{11+} diatomic at an internuclear separation of 6 Å vs the absolute value of eqn. 4 at a fractional electron number of q=0.5 for the carbon atom across all tested density functional approximations.