

# Supporting Information for “A similarity renormalization group approach to Green’s function methods”

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## S1. SRG- $G_0W_0$ , evGW and SRG-evGW statistics

In this section, the values obtained with the two alternative SRG-based methods derived in the main manuscript, SRG- $G_0W_0$  and SRG-evGW, are reported along with their corresponding histogram plot of the errors. For the sake of completeness, the SRG-regularized self-energy and quasiparticle equation used for the SRG- $G_0W_0$  and SRG-evGW calculations are reported below:

$$\epsilon_p^{\text{HF}} + \Sigma_{pp}^{\text{SRG-GW}}(\omega) - \omega = 0, \quad (\text{S1})$$

with

$$\Sigma_{pp}^{\text{SRG-GW}}(\omega) = \sum_{iv} \frac{(W_{pi}^v)^2}{\omega - \epsilon_i + \Omega_v} e^{-2(\epsilon_p - \epsilon_i + \Omega_v)^2 s} + \sum_{av} \frac{(W_{pa}^v)^2}{\omega - \epsilon_a - \Omega_v} e^{-2(\epsilon_p - \epsilon_a - \Omega_v)^2 s}, \quad (\text{S2})$$

Therefore, the SRG- $G_0W_0$  values are obtained by solving once these equations (one-shot procedure) without linearization, while the SRG-evGW results correspond to solutions of these equations where self-consistency on the  $\epsilon_p$ ’s has been reached.

One observe in Table S1 that the  $G_0W_0$  and SRG- $G_0W_0$  values are the same for all systems (up to  $10^{-2}$  eV). Figure S2 shows that evGW provides a slight improvement over  $G_0W_0$ , while evGW and SRG-evGW perform similarly. One interesting fact is that the convergence of SRG-evGW deteriorates faster than for SRG-qsGW with respect to  $s$ . We suspect that it is due to the absence of the off-diagonal terms.

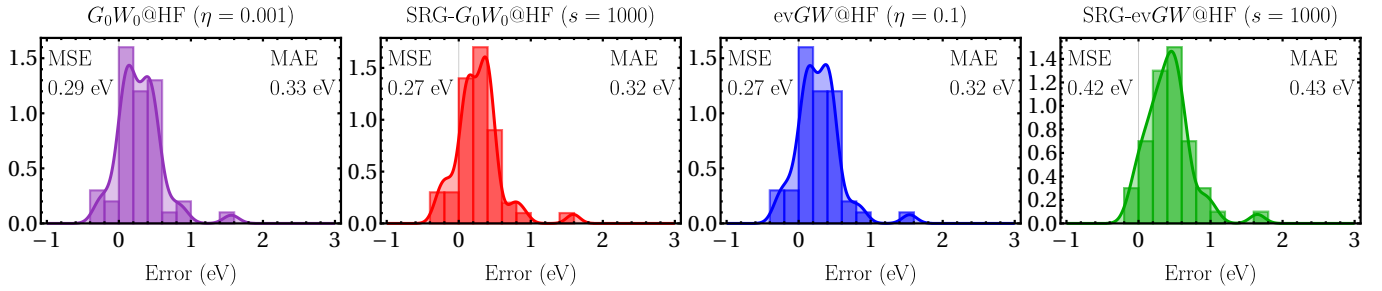


FIG. S1. Histogram of the errors [with respect to  $\Delta\text{CCSD(T)}$ ] for the principal IP of the GW50 test set calculated using  $G_0W_0$ @HF, SRG- $G_0W_0$ @HF, evGW, and SRG-evGW. All calculations are performed with the aug-cc-pVTZ basis.

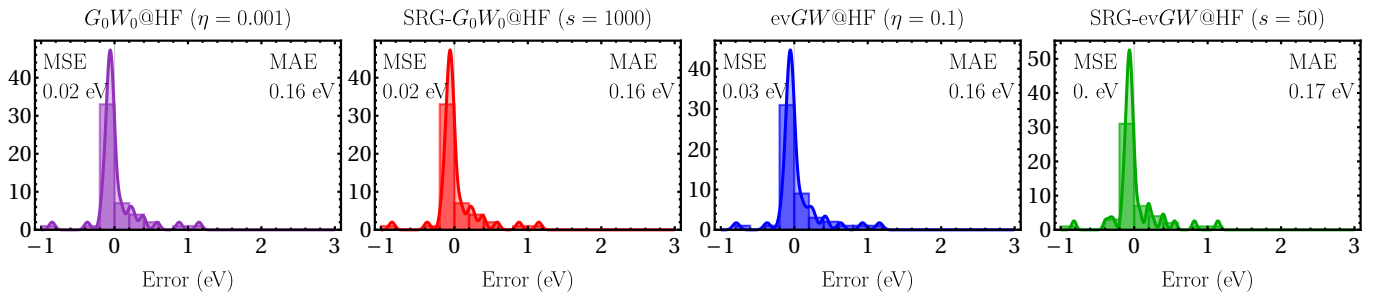


FIG. S2. Histogram of the errors [with respect to  $\Delta\text{CCSD(T)}$ ] for the principal EA of the GW50 test set calculated using  $G_0W_0$ @HF, SRG- $G_0W_0$ @HF, evGW, and SRG-evGW. All calculations are performed with the aug-cc-pVTZ basis.

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TABLE S1. Principal IP and EA (in eV) of the GW50 test set calculated using  $\Delta\text{CCSD(T)}$  (reference),  $G_0W_0@HF$ ,  $\text{SRG-}G_0W_0@HF$ ,  $\text{evGW}$ , and  $\text{SRG-evGW}$ . The statistical descriptors associated with the errors with respect to the reference values are also reported. All calculations are performed with the aug-cc-pVTZ basis.

Mol.	Principal IP					Principal EA				
	$\Delta\text{CCSD(T)}$ (Ref.)	$G_0W_0@HF$ ( $\eta = 10^{-3}$ )	$\text{SRG-}G_0W_0@HF$ ( $s = 10^3$ )	$\text{evGW}$ ( $\eta = 10^{-1}$ )	$\text{SRG-evGW}$ ( $s = 50$ )	$\Delta\text{CCSD(T)}$ (Ref.)	$G_0W_0@HF$ ( $\eta = 10^{-3}$ )	$\text{SRG-}G_0W_0@HF$ ( $s = 10^3$ )	$\text{evGW}$ ( $\eta = 10^{-1}$ )	$\text{SRG-evGW}$ ( $s = 50$ )
He	24.54	24.59	24.59	24.58	24.57	-2.66	-2.66	-2.66	-2.66	-2.66
Ne	21.47	21.46	21.46	21.30	21.29	-5.09	-5.25	-5.25	-5.24	-5.24
H <sub>2</sub>	16.40	16.49	16.49	16.52	16.51	-1.35	-1.28	-1.28	-1.28	-1.28
Li <sub>2</sub>	5.25	5.38	5.38	5.44	5.42	0.34	0.17	0.17	0.16	0.17
LiH	8.02	8.22	8.22	8.26	8.23	-0.29	0.27	0.27	0.27	0.27
HF	16.15	16.25	16.25	16.10	16.09	-0.66	-0.71	-0.71	-0.71	-0.71
Ar	15.60	15.72	15.72	15.67	15.66	-2.55	-2.68	-2.68	-2.67	-2.67
H <sub>2</sub> O	12.69	12.90	12.90	12.80	12.79	-0.61	-0.68	-0.68	-0.68	-0.68
LiF	11.47	11.40	11.40	11.20	11.18	0.35	0.33	0.33	0.33	0.33
HCl	12.67	12.78	12.78	12.76	12.75	-0.57	-0.64	-0.64	-0.64	-0.64
BeO	9.95	9.74	9.74	9.64	9.61	2.17	2.28	2.28	2.30	2.31
CO	13.99	14.80	14.80	14.77	14.76	-1.57	-1.66	-1.66	-1.65	-1.65
N <sub>2</sub>	15.54	17.10	17.10	17.10	17.09	-2.37	-2.10	-2.10	-2.10	-2.10
CH <sub>4</sub>	14.39	14.76	14.76	14.76	14.75	-0.65	-0.70	-0.70	-0.69	-0.69
BH <sub>3</sub>	13.31	13.68	13.68	13.70	13.69	-0.09	-0.46	-0.46	-0.46	-0.45
NH <sub>3</sub>	10.91	11.22	11.22	11.19	11.17	-0.61	-0.68	-0.68	-0.68	-0.68
BF	11.15	11.34	11.34	11.37	11.36	-0.80	-0.90	-0.90	-0.90	-0.90
BN	12.05	11.76	11.76	11.78	11.76	3.02	3.90	3.90	3.95	3.95
SH <sub>2</sub>	10.39	10.51	10.51	10.51	10.50	-0.52	-0.60	-0.60	-0.60	-0.60
F <sub>2</sub>	15.81	16.35	16.35	16.15	16.14	0.32	-0.53	-0.53	-0.47	-0.47
MgO	7.97	8.40	8.40	8.34	8.28	1.54	1.64	1.64	1.65	1.66
O <sub>3</sub>	12.85	13.56	13.56	13.53	13.51	1.82	2.19	2.19	2.25	2.25
C <sub>2</sub> H <sub>2</sub>	11.45	11.57	11.57	11.60	11.59	-0.80	-0.71	-0.71	-0.71	-0.71
HCN	13.76	13.86	13.86	13.87	13.86	-0.53	-0.52	-0.52	-0.52	-0.52
B <sub>2</sub> H <sub>6</sub>	12.27	12.81	12.81	12.81	12.80	-0.52	-0.56	-0.56	-0.56	-0.56
CH <sub>2</sub> O	10.93	11.39	11.39	11.34	11.32	-0.60	-0.61	-0.61	-0.60	-0.60
C <sub>2</sub> H <sub>4</sub>	10.69	10.74	10.74	10.78	10.77	-1.90	-0.75	-0.75	-0.74	-0.74
SiH <sub>4</sub>	12.79	13.22	13.22	13.23	13.22	-0.53	-0.59	-0.59	-0.59	-0.59
PH <sub>3</sub>	10.60	10.79	10.79	10.82	10.81	-0.51	-0.58	-0.58	-0.58	-0.58
CH <sub>4</sub> O	11.09	11.55	11.55	11.48	11.47	-0.59	-0.64	-0.64	-0.64	-0.64
H <sub>2</sub> NNH <sub>2</sub>	9.49	9.84	9.84	9.80	9.79	-0.60	-0.69	-0.69	-0.68	-0.68
HOOH	11.51	11.96	11.96	11.85	11.83	-0.96	-0.75	-0.75	-0.75	-0.75
KH	6.32	6.44	6.44	6.48	6.42	0.30	0.28	0.28	0.28	0.28
Na <sub>2</sub>	4.93	4.98	4.98	5.03	5.02	0.36	0.26	0.26	0.24	0.26
HN <sub>3</sub>	10.77	11.12	11.12	11.11	11.10	-0.51	-0.6	-0.6	-0.59	-0.59
CO <sub>2</sub>	13.80	14.24	14.24	14.16	14.15	-0.88	-0.98	-0.98	-0.97	-0.97
PN	11.90	12.33	12.33	12.34	12.33	-0.02	-0.03	-0.03	0.01	0.01
CH <sub>2</sub> O <sub>2</sub>	11.54	12.00	12.00	11.90	11.88	-0.63	-0.69	-0.69	-0.68	-0.68
C <sub>4</sub>	11.43	11.77	11.77	11.77	11.76	2.38	2.24	2.24	2.34	2.35
C <sub>3</sub> H <sub>6</sub>	10.83	11.20	11.20	11.20	11.19	-0.94	-0.75	-0.75	-0.75	-0.75
C <sub>2</sub> H <sub>3</sub> F	10.63	10.84	10.84	10.85	10.84	-0.65	-0.69	-0.69	-0.68	-0.68
C <sub>2</sub> H <sub>4</sub> O	10.29	10.84	10.84	10.76	10.75	-0.54	-0.56	-0.56	-0.56	-0.56
C <sub>2</sub> H <sub>6</sub> O	10.82	11.37	11.37	11.31	11.30	-0.58	-0.65	-0.65	-0.64	-0.64
C <sub>3</sub> H <sub>8</sub>	12.13	12.61	12.61	12.60	12.59	-0.63	-0.70	-0.70	-0.70	-0.70
NaCl	9.10	9.20	9.20	9.16	9.13	0.67	0.64	0.64	0.64	0.64
P <sub>2</sub>	10.72	10.49	10.49	10.52	10.51	0.43	0.47	0.47	0.53	0.54
MgF <sub>2</sub>	13.93	13.94	13.94	13.74	13.72	0.29	0.15	0.15	0.16	0.16
OCS	11.23	11.52	11.52	11.50	11.49	-1.43	-1.03	-1.03	-1.02	-1.01
SO <sub>2</sub>	10.48	11.38	11.38	11.34	11.33	2.24	2.82	2.82	2.87	2.88
C <sub>2</sub> H <sub>3</sub> Cl	10.17	10.39	10.39	10.39	10.38	-0.61	-0.66	-0.66	-0.65	-0.65
MSE		0.29	0.29	0.26	0.25		0.02	0.02	0.03	0.00
MAE		0.33	0.33	0.32	0.31		0.16	0.16	0.16	0.17
RMSE		0.43	0.43	0.41	0.40		0.28	0.28	0.29	0.28
SDE		0.31	0.31	0.31	0.32		0.29	0.29	0.29	0.29
Min		-0.29	-0.29	-0.31	-0.34		-0.85	-0.85	-0.79	-0.82
Max		1.56	1.56	1.56	1.55		1.15	1.15	1.16	1.14