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:

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

with

$$\Sigma_{pp}^{\text{SRG-}GW}(\omega) = \sum_{i\nu} \frac{(W_{pi}^{\nu})^2}{\omega - \epsilon_i + \Omega_\nu} e^{-2(\epsilon_p - \epsilon_i + \Omega_\nu)^2 s} + \sum_{a\nu} \frac{(W_{pa}^{\nu})^2}{\omega - \epsilon_a - \Omega_\nu} e^{-2(\epsilon_p - \epsilon_a - \Omega_\nu)^2 s}, \tag{S2}$$

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 ϵ_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.

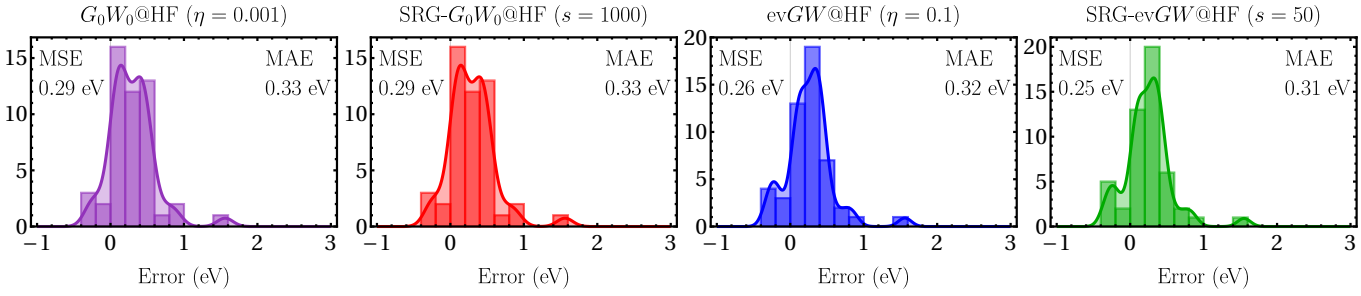

FIG. S1. Histogram of the errors [with respect to ΔCCSD(T)] for the principal IP of the GW50 test set calculated using G_0W_0@HF, SRG-G_0W_0@HF, evGW, and SRG-evGW. All calculations are performed with the aug-cc-pVTZ basis.

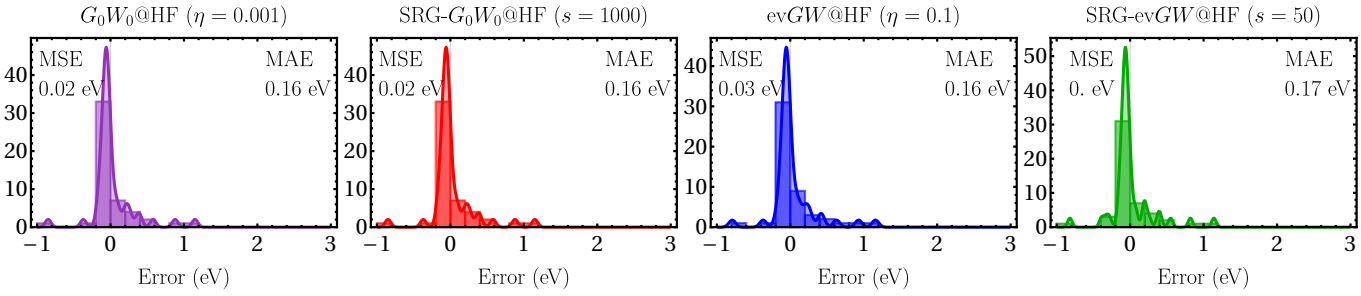

FIG. S2. Histogram of the errors [with respect to ΔCCSD(T)] for the principal EA of the GW50 test set calculated using G_0W_0@HF, SRG-G_0W_0@HF, evGW, and SRG-evGW. All calculations are performed with the aug-cc-pVTZ basis.

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TABLE S1. Principal IP and EA (in eV) of the $GW50$ test set calculated using ΔCCSD(T) (reference), G_0W_0@HF, SRG-G_0W_0@HF, evGW, and 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.

	Principal IP					Principal EA				
Mol.	ΔCCSD(T) (Ref.)	G_0W_0@HF ($\eta = 10^{-3}$)	SRG-G_0W_0@HF ($s = 10^3$)	evGW ($\eta = 10^{-1}$)	SRG-evGW ($s = 50$)	ΔCCSD(T) (Ref.)	G_0W_0@HF ($\eta = 10^{-3}$)	SRG-G_0W_0@HF ($s = 10^3$)	evGW ($\eta = 10^{-1}$)	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_2	16.40	16.49	16.49	16.52	16.51	−1.35	−1.28	−1.28	−1.28	−1.28
Li_2	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_2O	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_2	15.54	17.10	17.10	17.10	17.09	−2.37	−2.10	−2.10	−2.10	−2.10
CH_4	14.39	14.76	14.76	14.76	14.75	−0.65	−0.70	−0.70	−0.69	−0.69
BH_3	13.31	13.68	13.68	13.70	13.69	−0.09	−0.46	−0.46	−0.46	−0.45
NH_3	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_2	10.39	10.51	10.51	10.51	10.50	−0.52	−0.60	−0.60	−0.60	−0.60
F_2	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_3	12.85	13.56	13.56	13.53	13.51	1.82	2.19	2.19	2.25	2.25
C_2H_2	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_2H_6	12.27	12.81	12.81	12.81	12.80	−0.52	−0.56	−0.56	−0.56	−0.56
CH_2O	10.93	11.39	11.39	11.34	11.32	−0.60	−0.61	−0.61	−0.60	−0.60
C_2H_4	10.69	10.74	10.74	10.78	10.77	−1.90	−0.75	−0.75	−0.74	−0.74
SiH_4	12.79	13.22	13.22	13.23	13.22	−0.53	−0.59	−0.59	−0.59	−0.59
PH_3	10.60	10.79	10.79	10.82	10.81	−0.51	−0.58	−0.58	−0.58	−0.58
CH_4O	11.09	11.55	11.55	11.48	11.47	−0.59	−0.64	−0.64	−0.64	−0.64
H_2NNH_2	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_2	4.93	4.98	4.98	5.03	5.02	0.36	0.26	0.26	0.24	0.26
HN_3	10.77	11.12	11.12	11.11	11.10	−0.51	−0.6	−0.6	−0.59	−0.59
CO_2	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_2O_2	11.54	12.00	12.00	11.90	11.88	−0.63	−0.69	−0.69	−0.68	−0.68
C_4	11.43	11.77	11.77	11.77	11.76	2.38	2.24	2.24	2.34	2.35
C_3H_6	10.83	11.20	11.20	11.20	11.19	−0.94	−0.75	−0.75	−0.75	−0.75
C_2H_3F	10.63	10.84	10.84	10.85	10.84	−0.65	−0.69	−0.69	−0.68	−0.68
C_2H_4O	10.29	10.84	10.84	10.76	10.75	−0.54	−0.56	−0.56	−0.56	−0.56
C_2H_6O	10.82	11.37	11.37	11.31	11.30	−0.58	−0.65	−0.65	−0.64	−0.64
C_3H_8	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_2	10.72	10.49	10.49	10.52	10.51	0.43	0.47	0.47	0.53	0.54
MgF_2	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_2	10.48	11.38	11.38	11.34	11.33	2.24	2.82	2.82	2.87	2.88
C_2H_3Cl	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