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

Supporting Information for

Effects of the locality of a potential derived from hybrid density functionals on Kohn-Sham orbitals and excited states

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1. Atomization energies

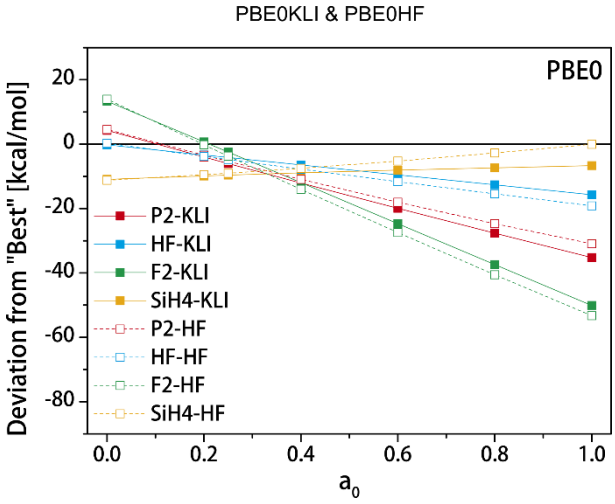

Figure S1. Atomization energies of P_2, HF, F_2, and SiH_4 as a function of the exact exchange ratio (a_0) withrespect to the ab initio calculations [R. Haunschild, W. Klopper, *J. Chem. Phys.* **2012**, *136*, 164102] which are denoted as “Best”.

2. Caricato set - Orbital energy gaps from PBE0HF-a_0

Table S1. The orbital energy gaps from PBE0HF-a_0 with various a_0 as an approximation to each excitation energy for the Caricato set. The third column indicates the type of excitations: V and R for valence and Rydberg excitations, respectively. The experimental values were obtained from [M. Caricato, G. W. Trucks, M. J. Frisch, K. B. Wiberg, *J. Chem. Theory Comput.* **2010**, *6* (2), 370–383]. Unit: eV

Molecule	State	Type	PBE0HF-a_0 (Orbital energy gap)						Expt.
			0.0	0.2	0.4	0.6	0.8	1.0	
formaldehyde	1^1A_2	V	3.52	5.81	8.08	10.27	11.84	13.24	4.00
	1^1B_2	R	5.78	7.35	8.83	10.26	11.66	13.07	7.08
	2^1B_2	R	6.36	7.69	9.07	10.47	11.87	13.27	7.97
	2^1A_1	R	6.34	7.68	9.02	10.38	11.76	13.16	8.14
	2^1A_2	R	6.43	7.75	9.10	10.49	12.31	13.84	8.37
	3^1B_2	R	6.41	7.76	9.12	10.50	11.91	13.33	8.88
	1^1B_1	V	8.27	10.78	13.25	15.63	17.39	18.96	9.00
	3^1A_2	R	6.88	14.49	9.65	11.05	12.50	14.06	9.22
	4^1B_2	R	6.71	8.10	9.50	10.94	12.38	13.81	9.26
	4^1A_1	R	6.81	8.18	9.57	10.97	12.37	13.79	9.58
	5^1B_2	R	6.76	13.79	9.61	11.01	12.41	13.83	9.63
acetaldehyde	$1^1A''$	V	3.87	6.12	8.37	10.58	12.52	15.14	4.28
	$2^1A'$	R	5.38	6.95	8.45	9.87	11.25	12.63	6.82
	$3^1A'$	R	5.89	7.31	8.65	10.00	11.35	12.72	7.46
	$4^1A'$	R	6.04	7.35	8.70	10.05	11.41	12.77	7.75
	$6^1A'$	R	6.13	7.49	8.87	10.21	11.57	12.93	8.43
	$7^1A'$	R	6.23	7.55	8.87	10.22	11.57	12.94	8.69
acetone	1^1A_2	V	3.97	6.18	8.31	10.39	12.53	14.99	4.43
	1^1B_2	R	4.94	2.99	8.11	9.55	10.93	12.29	6.36
	2^1A_2	R	5.73	7.06	8.42	9.70	11.04	12.38	7.36
	2^1A_1	R	5.63	7.02	8.36	9.69	11.03	12.38	7.41
	2^1B_2	R	5.75	7.06	8.40	9.73	11.07	12.42	7.49
	3^1A_1	R	5.85	7.19	8.50	9.82	11.16	12.50	7.80
	3^1B_2	R	5.78	7.09	8.41	9.78	11.17	12.55	8.09
	1^1B_1	R	5.89	7.18	8.49	9.81	11.14	12.48	8.17
ethylene	1^1B_{3u}	R	6.39	7.51	8.53	9.48	10.41	11.33	7.11
	1^1B_{1u}	V	5.63	7.33	8.87	9.80	10.71	11.63	7.65
	1^1B_{1g}	R	6.80	7.74	8.65	9.56	10.47	11.38	7.80
	1^1B_{2g}	R	6.78	7.73	8.65	9.56	10.47	11.38	7.90
	2^1A_g	R	6.92	7.83	8.76	9.68	10.60	11.52	8.28
	2^1B_{3u}	R	6.86	7.80	8.76	9.74	10.69	11.62	8.62
	3^1B_{3u}	R	7.08	7.99	8.89	9.80	10.72	11.65	8.90
	4^1B_{3u}	R	7.30	8.27	9.25	10.24	11.22	12.20	9.08
	3^1B_{1g}	R	6.97	7.97	8.92	9.86	10.79	11.72	9.20
	2^1B_{1u}	R	7.09	8.00	9.00	10.46	11.63	12.64	9.33
	5^1B_{3u}	R	7.81	8.81	9.80	10.77	11.73	12.68	9.51
isobutene	$^1B_1$	R	5.34	6.53	7.62	8.59	9.52	10.44	6.17
	$^1A_1$	R	5.23	6.75	7.80	8.70	9.60	10.51	6.7

trans-butadiene	1^1B_u	V	3.91	5.42	6.93	8.36	9.22	10.05	5.91
	1^1B_g	R	5.48	6.47	7.37	8.21	9.04	9.87	6.22
	2^1A_u	R	5.74	6.65	7.48	8.29	9.10	9.93	6.66
	2^1B_u	R	5.99	6.80	7.60	8.49	9.49	10.32	7.07
	2^1B_g	R	5.95	6.74	7.57	8.37	9.18	10.00	7.36
	3^1A_g	R	6.18	6.95	7.74	8.54	9.36	10.18	7.62
	3^1B_u	R	6.30	7.08	7.88	8.68	9.85	10.92	8.00
pyridine	$^1B_1$	V	3.83	6.05	8.05	10.04	12.02	13.76	4.59
	$^1B_2$	V	4.80	6.31	7.80	9.27	10.71	11.90	4.99
	$^1A_2$	V	4.19	6.42	8.44	10.45	12.38	14.09	5.43
	$^1A_1$	V	5.16	6.69	8.20	9.68	11.07	12.23	6.38
pyrazine	$^1B_{3u}$	V	3.25	5.23	7.22	9.21	11.15	13.16	3.83
	$^1B_{2u}$	V	4.44	5.96	7.46	8.93	10.33	11.76	4.81
	$^1B_{2g}$	V	4.75	6.92	9.10	11.28	13.41	15.58	5.46
	$^1B_{1g}$	V	5.52	7.72	9.93	12.15	14.35	16.28	6.10
	$^1B_{1u}$	V	5.76	7.43	9.09	10.72	12.28	13.85	6.51
pyridazine	$^1B_1$	V	2.79	4.84	6.90	8.97	11.03	13.00	3.60
	$^1A_1$	V	4.89	6.45	8.00	9.53	11.02	12.41	5.00
	$^1A_2$	V	4.78	6.84	8.90	10.96	13.00	14.95	5.30
	$^1B_1$	V	5.34	7.44	9.53	11.61	13.66	15.41	6.00
	$^1B_2$	V	5.45	7.05	8.63	10.17	11.68	12.88	6.50
pyrimidine	$^1B_1$	V	3.58	5.59	7.60	9.62	11.75	13.55	3.85
	$^1A_2$	V	3.92	5.95	7.99	10.03	12.05	13.74	4.62
	$^1B_2$	V	5.01	6.58	8.13	9.66	11.30	12.60	5.12
	$^1A_2$	V	4.83	6.92	9.01	11.09	13.28	15.14	5.52
	$^1B_1$	V	5.17	7.28	9.39	11.50	13.58	15.33	5.90
	$^1A_1$	V	5.35	6.94	8.51	10.07	11.60	12.79	6.70
s-tetrazine	$^1B_{3u}$	V	1.53	3.60	5.68	7.78	9.89	12.02	2.25
	^1A_u	V	2.70	4.84	6.99	9.15	11.29	13.47	3.40
	^1A_u	V	4.35	6.44	8.53	10.63	12.73	14.84	5.00
	$^1B_{3u}$	V	5.52	7.68	9.84	12.00	14.13	16.29	6.34
Mean absolute error (total)			1.30	1.05	1.74	3.20	4.66	6.07	
Mean absolute error (valence)			0.75	1.27	3.14	5.04	6.85	8.55	
Mean absolute error (Rydberg)			1.73	0.89	0.67	1.80	2.98	4.15	

3. Caricato set – Orbital energy gaps from PBE0KLI-a_0

Table S2. The orbital energy gaps from PBE0KLI-a_0 with various a_0 as an approximation to each excitation energy for the Caricato set. The third column indicates the type of excitations: V and R for valence and Rydberg excitations, respectively. See **Table S1** for the experimental values. Unit: eV

Molecule	State	Type	PBE0KLI-a_0 (Orbital energy gap)						
			0.0	0.2	0.4	0.5	0.6	0.8	1.0
formaldehyde	1^1A_2	V	3.46	3.59	3.76	3.77	3.83	3.95	4.28
	1^1B_2	R	5.83	6.41	6.95	7.22	7.48	7.98	8.49
	2^1B_2	R	6.67	7.23	7.76	8.00	8.24	8.69	9.17
	2^1A_1	R	6.61	7.28	7.90	8.22	8.52	9.10	9.75
	2^1A_2	R	6.92	7.53	8.12	8.42	8.70	9.27	10.00
	3^1B_2	R	7.46	8.13	8.74	9.05	9.34	9.88	10.55
	1^1B_1	V	8.07	8.24	8.47	8.48	8.56	8.71	9.00
	3^1A_2	R	7.73	8.44	9.14	9.50	9.84	10.54	11.52
	4^1B_2	R	7.66	8.36	9.04	9.39	9.73	10.39	11.27
	4^1A_1	R	8.23	8.94	9.64	10.02	10.38	11.10	12.20
	5^1B_2	R	7.71	8.46	9.20	9.59	9.97	10.73	11.86
acetaldehyde	$1^1A''$	V	3.84	3.99	4.14	4.21	4.28	4.42	4.75
	$2^1A'$	R	5.38	5.92	6.42	6.66	6.90	7.34	7.78
	$3^1A'$	R	5.94	6.55	7.12	7.39	7.66	8.16	8.66
	$4^1A'$	R	6.33	6.96	7.55	7.82	8.09	8.60	9.10
	$6^1A'$	R	6.90	7.56	8.19	8.50	8.79	9.35	9.98
	$7^1A'$	R	7.07	7.78	8.47	8.80	9.13	9.76	10.51
acetone	1^1A_2	V	3.93	4.11	4.32	4.35	4.44	4.59	4.91
	1^1B_2	R	4.96	5.49	5.97	6.21	6.44	6.87	7.27
	2^1A_2	R	5.93	6.56	7.14	7.44	7.72	8.24	8.77
	2^1A_1	R	5.72	6.37	6.96	7.26	7.53	8.06	8.57
	2^1B_2	R	6.04	6.69	7.31	7.62	7.91	8.50	9.09
	3^1A_1	R	6.53	7.22	7.85	8.16	8.46	9.00	9.55
	3^1B_2	R	6.28	6.90	7.45	7.74	7.99	8.41	8.86
	1^1B_1	R	6.58	7.29	7.96	8.29	8.60	9.18	9.74
ethylene	1^1B_{3u}	R	6.46	6.85	7.21	7.40	7.58	7.91	8.04
	1^1B_{1u}	V	5.65	5.73	5.79	5.84	5.88	5.95	5.98
	1^1B_{1g}	R	7.01	7.47	7.89	8.12	8.32	8.70	8.91
	1^1B_{2g}	R	6.95	7.43	7.86	8.09	8.31	8.72	8.95
	2^1A_g	R	7.33	7.73	8.10	8.31	8.49	8.86	9.14
	2^1B_{3u}	R	7.79	8.28	8.68	8.89	9.07	9.39	9.59
	3^1B_{3u}	R	7.99	8.42	8.86	9.12	9.36	9.82	10.23
	4^1B_{3u}	R	8.26	8.75	9.23	9.50	9.74	10.23	10.78
	3^1B_{1g}	R	8.55	9.01	9.44	9.67	9.88	10.28	10.70
	2^1B_{1u}	R	8.12	8.60	9.06	9.31	9.55	10.01	10.54
	5^1B_{3u}	R	9.30	9.74	10.12	10.34	10.53	10.91	11.37
isobutene	$^1B_1$	R	5.37	5.74	6.08	6.24	6.40	6.70	6.84
	$^1A_1$	R	5.23	5.34	5.43	5.47	5.51	5.58	5.65
	1^1B_u	V	3.93	3.98	4.02	4.06	4.09	4.14	4.18

trans-butadiene	1^1B_g	R	5.55	5.93	6.26	6.45	6.62	6.93	7.03
	2^1A_u	R	5.84	6.24	6.60	6.78	7.00	7.34	7.49
	2^1B_u	R	6.35	6.71	7.04	7.23	7.40	7.74	7.96
	2^1B_g	R	6.53	6.99	7.40	7.63	7.84	8.21	8.36
	3^1A_g	R	6.86	7.23	7.58	7.78	7.96	8.32	9.29
	3^1B_u	R	7.52	7.96	8.36	8.59	8.80	9.21	9.62
pyridine	$^1B_1$	V	3.99	4.08	4.18	4.22	4.27	4.35	4.62
	$^1B_2$	V	4.82	4.83	4.83	4.85	4.86	4.87	4.85
	$^1A_2$	V	4.36	4.48	4.58	4.65	4.71	4.81	5.11
	$^1A_1$	V	5.19	5.23	5.23	5.28	5.30	5.33	5.34
pyrazine	$^1B_{3u}$	V	3.19	3.25	3.32	3.33	3.36	3.41	3.66
	$^1B_{2u}$	V	4.46	4.45	4.43	4.44	4.43	4.42	4.38
	$^1B_{2g}$	V	4.67	4.76	4.89	4.89	4.93	5.01	5.22
	$^1B_{1g}$	V	5.45	5.59	5.74	5.79	5.85	5.98	6.25
	$^1B_{1u}$	V	5.71	5.75	5.82	5.81	5.83	5.87	5.86
pyridazine	$^1B_1$	V	2.69	2.80	2.87	2.94	2.99	3.08	3.37
	$^1A_1$	V	4.87	4.89	4.90	4.91	4.91	4.93	4.91
	$^1A_2$	V	4.68	4.76	4.88	4.88	4.92	4.98	5.22
	$^1B_1$	V	5.27	5.38	5.49	5.53	5.58	5.67	5.94
	$^1B_2$	V	5.46	5.50	5.52	5.56	5.58	5.61	5.63
pyrimidine	$^1B_1$	V	3.48	3.58	3.67	3.72	3.76	3.85	4.13
	$^1A_2$	V	3.84	3.96	4.06	4.13	4.18	4.29	4.60
	$^1B_2$	V	5.00	5.02	5.03	5.04	5.05	5.07	5.07
	$^1A_2$	V	4.73	4.82	4.91	4.95	5.00	5.08	5.34
	$^1B_1$	V	5.08	5.20	5.30	5.36	5.42	5.52	5.80
	$^1A_1$	V	5.36	5.40	5.42	5.45	5.47	5.51	5.53
s-tetrazine	$^1B_{3u}$	V	1.42	1.49	1.55	1.58	1.61	1.67	1.94
	^1A_u	V	2.64	2.76	2.83	2.93	2.98	3.09	3.43
	^1A_u	V	4.25	4.29	4.38	4.34	4.36	4.39	4.59
	$^1B_{3u}$	V	5.47	5.56	5.65	5.69	5.73	5.81	6.08
Mean absolute error (total)			1.01	0.67	0.40	0.38	0.48	0.72	0.95
Mean absolute error (valence)			0.78	0.70	0.62	0.58	0.54	0.49	0.40
Mean absolute error (Rydberg)			1.18	0.65	0.23	0.23	0.43	0.89	1.38

4. Caricato set – TDDFT results from PBE0HF-a_0

Table S3. The excitation energies of the Caricato set from TDDFT calculations performed on PBE0HF-a_0 orbitals with various a_0. The third column indicates the type of excitations: V and R for valence and Rydberg excitations, respectively. See **Table S1** for the experimental values. Unit: eV

Molecule	State	Type	PBE0HF-a_0 (TDDFT)					
			0.0	0.2	0.4	0.6	0.8	1.0
formaldehyde	1^1A_2	V	3.78	3.88	3.98	4.10	4.22	4.37
	1^1B_2	R	5.76	6.51	7.25	7.98	8.68	9.35
	2^1B_2	R	6.36	7.24	7.99	8.72	9.41	10.08
	2^1A_1	R	6.33	7.22	8.04	8.83	9.44	9.44
	2^1A_2	R	6.42	7.35	8.22	9.04	9.83	10.61
	3^1B_2	R	6.40	7.41	8.45	9.46	10.42	11.30
	1^1B_1	V	8.83	9.01	9.17	9.33	9.48	9.62
	3^1A_2	R	6.86	7.75	8.72	9.74	10.81	11.24
	4^1B_2	R	6.71	7.64	8.64	9.64	10.59	11.49
	4^1A_1	R	6.81	7.78	8.75	9.68	10.64	11.56
	5^1B_2	R	6.76	7.75	8.78	9.75	10.67	11.57
acetaldehyde	$1^1A''$	V	4.11	4.22	4.33	4.46	4.60	4.75
	$2^1A'$	R	5.43	6.25	7.04	7.80	8.53	9.22
	$3^1A'$	R	5.88	6.73	7.58	8.41	9.19	9.44
	$4^1A'$	R	6.05	6.96	7.79	8.59	9.27	9.99
	$6^1A'$	R	6.12	7.18	8.23	9.15	10.04	10.89
	$7^1A'$	R	6.22	7.23	8.26	9.23	10.11	10.97
acetone	1^1A_2	V	4.20	4.35	4.50	4.66	4.82	5.00
	1^1B_2	R	4.94	5.83	6.68	7.51	8.30	9.05
	2^1A_2	R	5.72	6.63	7.50	8.35	9.17	9.95
	2^1A_1	R	5.65	6.57	7.48	8.36	9.15	9.43
	2^1B_2	R	5.75	6.68	7.55	8.40	9.23	10.05
	3^1A_1	R	5.86	6.92	7.96	8.91	9.50	10.18
	3^1B_2	R	5.78	6.79	7.79	8.73	9.61	10.46
	1^1B_1	R	5.89	6.94	7.98	8.96	9.53	9.74
ethylene	1^1B_{3u}	R	6.38	6.72	7.04	7.35	7.65	7.93
	1^1B_{1u}	V	7.07	7.43	7.53	7.57	7.60	7.60
	1^1B_{1g}	R	6.79	7.23	7.59	7.93	8.26	8.56
	1^1B_{2g}	R	6.77	7.22	7.63	8.02	8.39	8.76
	2^1A_g	R	6.92	7.42	7.87	8.28	8.65	9.00
	2^1B_{3u}	R	6.86	7.50	8.10	8.64	9.10	9.50
	3^1B_{3u}	R	7.08	7.68	8.26	8.80	9.32	9.76
	4^1B_{3u}	R	7.29	7.85	8.40	8.93	9.40	9.86
	3^1B_{1g}	R	7.41	7.96	8.30	8.84	9.42	9.96
	2^1B_{1u}	R	7.33	7.73	8.32	8.89	9.43	9.92
	5^1B_{3u}	R	7.78	8.23	8.71	9.21	9.75	10.33
isobutene	$^1B_1$	R	5.33	5.79	6.22	6.63	7.01	7.37
	$^1A_1$	R	5.90	6.33	6.68	6.95	7.14	7.26
	1^1B_u	V	5.41	5.60	5.77	5.90	6.00	6.07

trans-butadiene	1^1B_g	R	5.46	5.79	6.10	6.41	6.71	7.00
	2^1A_u	R	5.73	6.11	6.37	6.82	7.17	7.50
	2^1B_u	R	6.03	6.44	6.82	7.17	7.49	7.81
	2^1B_g	R	5.95	6.47	6.97	7.40	7.79	8.16
	3^1A_g	R	6.17	6.65	7.12	7.57	8.00	8.40
	3^1B_u	R	6.31	6.79	7.28	7.76	8.25	8.74
pyridine	$^1B_1$	V	4.34	4.76	5.11	5.42	5.70	5.97
	$^1B_2$	V	5.31	5.49	5.64	5.76	5.86	5.94
	$^1A_2$	V	4.44	5.06	5.67	6.25	6.79	7.31
	$^1A_1$	V	6.17	6.28	6.38	6.39	6.34	6.25
pyrazine	$^1B_{3u}$	V	3.54	3.91	4.21	4.47	4.70	4.90
	$^1B_{2u}$	V	5.21	5.35	5.46	5.54	5.59	5.62
	$^1B_{2g}$	V	5.11	5.58	5.92	6.18	6.39	6.57
	$^1B_{1g}$	V	5.57	6.40	7.23	8.07	8.92	9.76
	$^1B_{1u}$	V	6.37	6.50	6.57	6.56	6.49	6.37
pyridazine	$^1B_1$	V	3.11	3.53	3.89	4.19	4.45	4.67
	$^1A_1$	V	5.44	5.62	5.78	5.90	6.00	6.07
	$^1A_2$	V	5.01	5.45	5.86	6.26	6.68	7.12
	$^1B_1$	V	5.43	6.07	6.66	7.24	7.79	8.33
	$^1B_2$	V	6.30	6.36	6.51	6.51	6.43	6.29
pyrimidine	$^1B_1$	V	3.77	4.22	4.63	5.01	5.36	5.70
	$^1A_2$	V	3.99	4.56	5.08	5.57	6.00	6.41
	$^1B_2$	V	5.57	5.75	5.91	6.05	6.17	6.27
	$^1A_2$	V	5.08	5.64	6.12	6.55	6.96	7.35
	$^1B_1$	V	5.31	5.94	6.53	7.10	7.65	8.17
	$^1A_1$	V	6.41	6.58	6.68	6.71	6.70	6.65
s-tetrazine	$^1B_{3u}$	V	1.82	2.19	2.51	2.78	3.01	3.22
	^1A_u	V	2.80	3.43	4.03	4.58	5.07	5.48
	^1A_u	V	4.61	5.07	5.45	5.80	6.12	6.44
	$^1B_{3u}$	V	5.63	6.28	6.90	7.49	8.05	8.60
Mean absolute error (total)			1.13	0.63	0.38	0.56	0.98	1.39
Mean absolute error (valence)			0.40	0.19	0.41	0.67	0.93	1.19
Mean absolute error (Rydberg)			1.70	0.98	0.35	0.48	1.01	1.55

Table S4. The excitation energies of the Caricato set from TDDFT calculations performed on PBE0HF-a_0 orbitals with various a_0. An LDA kernel was used. The third column indicates the type of excitations: V and R for valence and Rydberg excitations, respectively. See **Table S12** for the experimental values. Unit: eV

Molecule	State	Type	PBE0HF-a_0 (TDDFT w/ LDA ker.)					
			0.0	0.2	0.4	0.6	0.8	1.0
formaldehyde	1^1A_2	V	3.64	5.93	8.18	10.32	11.83	13.24
	1^1B_2	R	5.64	7.23	8.76	10.22	11.64	13.05
	2^1B_2	R	6.35	7.68	9.04	10.44	11.85	13.26
	2^1A_1	R	6.30	7.66	9.01	10.38	11.76	13.15
	2^1A_2	R	6.42	7.75	9.10	10.51	12.32	13.83
	3^1B_2	R	6.38	7.75	9.11	10.49	11.89	13.31
	1^1B_1	V	8.64	11.12	13.56	15.79	17.92	20.96
	3^1A_2	R	6.85	8.24	9.64	11.05	12.50	14.04
	4^1B_2	R	6.67	8.07	9.47	10.89	12.34	13.79
	4^1A_1	R	6.78	8.14	9.53	10.93	12.34	13.76
	5^1B_2	R	6.73	8.14	9.59	11.00	12.40	13.81
acetaldehyde	$1^1A''$	V	3.99	6.24	8.48	10.60	12.51	15.21
	$2^1A'$	R	5.36	6.93	8.43	9.86	11.24	12.63
	$3^1A'$	R	5.80	7.27	8.64	9.99	11.35	12.71
	$4^1A'$	R	6.04	7.35	8.70	10.05	11.41	12.77
	$6^1A'$	R	6.11	7.45	8.84	10.21	11.56	12.93
	$7^1A'$	R	6.21	7.54	8.87	10.21	11.57	12.93
acetone	1^1A_2	V	4.09	6.30	8.34	10.41	12.53	15.06
	1^1B_2	R	4.90	6.53	8.08	9.54	10.93	12.29
	2^1A_2	R	5.71	7.06	8.47	9.70	11.04	12.38
	2^1A_1	R	5.62	7.02	8.36	9.69	11.03	12.37
	2^1B_2	R	5.74	7.05	8.39	9.73	11.07	12.41
	3^1A_1	R	5.85	7.19	8.50	9.82	11.15	12.50
	3^1B_2	R	5.77	7.08	8.40	9.77	11.16	12.55
	1^1B_1	R	5.89	7.17	8.48	9.81	11.14	12.48
ethylene	1^1B_{3u}	R	6.28	7.43	8.48	9.46	10.39	11.32
	1^1B_{1u}	V	7.00	7.98	8.89	9.80	10.71	11.62
	1^1B_{1g}	R	6.77	7.73	8.65	9.56	10.47	11.38
	1^1B_{2g}	R	6.94	7.94	8.90	9.85	10.78	11.71
	2^1A_g	R	11.50	7.83	8.75	9.67	10.59	11.50
	2^1B_{3u}	R	6.86	7.78	8.73	9.70	10.66	11.61
	3^1B_{3u}	R	7.08	7.99	8.89	9.80	10.71	11.63
	4^1B_{3u}	R	7.28	8.25	9.23	10.21	11.20	12.17
	3^1B_{1g}	R	7.39	8.39	9.39	10.38	11.35	12.30
	2^1B_{1u}	R	7.15	8.48	9.63	10.66	11.64	12.61
	5^1B_{3u}	R	7.75	8.77	9.76	10.73	11.70	12.66
isobutene	$^1B_1$	R	5.27	6.47	7.58	8.57	9.51	10.44
	$^1A_1$	R	5.83	6.87	7.79	8.69	9.60	10.51
trans-butadiene	1^1B_u	V	5.25	6.59	8.68	10.02	9.97	12.77
	1^1B_g	R	5.40	6.41	7.34	8.20	9.03	9.86

	2^1A_u	R	5.69	6.63	7.48	8.29	9.10	9.92
	2^1B_u	R	6.02	6.88	7.56	8.39	9.21	10.04
	2^1B_g	R	5.94	6.73	7.55	8.37	9.18	10.00
	3^1A_g	R	6.21	7.42	7.74	8.54	9.35	10.18
	3^1B_u	R	6.30	7.09	7.85	8.67	9.49	10.31
pyridine	$^1B_1$	V	4.25	6.24	8.23	10.20	12.16	13.77
	$^1B_2$	V	5.24	6.79	8.29	9.80	11.05	11.90
	$^1A_2$	V	4.41	6.42	8.44	10.45	12.38	14.08
	$^1A_1$	V	5.99	7.53	9.02	10.41	11.08	12.88
pyrazine	$^1B_{3u}$	V	3.45	5.43	7.41	9.40	11.16	13.35
	$^1B_{2u}$	V	5.13	6.70	8.22	9.71	10.34	12.22
	$^1B_{2g}$	V	5.01	7.17	9.35	11.52	13.43	15.85
	$^1B_{1g}$	V	5.55	7.75	9.98	12.16	14.36	16.36
	$^1B_{1u}$	V	6.19	7.75	9.30	10.74	11.78	13.82
pyridazine	$^1B_1$	V	3.03	5.07	7.12	9.18	11.06	13.01
	$^1A_1$	V	5.38	6.97	8.55	10.09	11.05	12.42
	$^1A_2$	V	4.92	6.98	9.03	11.08	13.03	15.15
	$^1B_1$	V	5.39	7.48	9.57	11.65	13.70	15.94
	$^1B_2$	V	6.11	7.73	9.29	10.81	11.59	13.00
pyrimidine	$^1B_1$	V	3.70	5.71	7.72	9.73	11.72	13.61
	$^1A_2$	V	3.95	5.98	8.02	10.05	12.08	13.89
	$^1B_2$	V	5.50	7.11	8.69	10.25	11.72	12.78
	$^1A_2$	V	5.01	7.09	9.17	11.24	13.29	15.21
	$^1B_1$	V	5.26	7.37	9.47	11.57	13.64	15.48
	$^1A_1$	V	6.23	7.82	9.40	10.90	12.23	13.99
s-tetrazine	$^1B_{3u}$	V	1.74	3.81	5.89	7.99	10.10	12.22
	^1A_u	V	2.76	4.90	7.05	9.20	11.33	13.24
	^1A_u	V	4.53	6.61	8.70	10.79	12.89	13.51
	$^1B_{3u}$	V	5.58	7.74	9.90	12.06	14.17	16.34
Mean absolute error (total)			1.21	1.00	1.87	3.33	4.70	6.17
Mean absolute error (valence)			0.47	1.53	3.45	5.31	6.95	8.81
Mean absolute error (Rydberg)			1.77	0.59	0.66	1.81	2.98	4.14

5. Caricato set – TDDFT results from PBE0KLI-a_0

Table S5. The excitation energies of the Caricato set from TDDFT calculations performed on PBE0KLI-a_0 orbitals with various a_0. An LDA kernel was used. The third column indicates the type of excitations: V and R for valence and Rydberg excitations, respectively. See **Table S12** for the experimental values. Unit: eV

Molecule	State	Type	PBE0KLI-a_0 (TDDFT w/ LDA ker.)						
			0.0	0.2	0.4	0.5	0.6	0.8	1.0
formaldehyde	1^1A_2	V	3.78	3.91	4.07	4.09	4.15	4.26	4.59
	1^1B_2	R	5.81	6.40	6.95	7.23	7.50	8.01	8.54
	2^1B_2	R	6.66	7.25	7.79	8.05	8.31	8.78	9.31
	2^1A_1	R	6.59	7.27	7.90	8.23	8.54	9.13	9.81
	2^1A_2	R	6.88	7.50	8.08	8.38	8.67	9.23	9.96
	3^1B_2	R	7.48	8.17	8.80	9.12	9.42	9.99	10.68
	1^1B_1	V	8.76	8.92	8.96	9.14	9.22	9.36	9.63
	3^1A_2	R	7.70	8.41	9.11	9.47	9.81	9.89	10.13
	4^1B_2	R	7.63	8.33	9.01	9.36	9.70	10.36	11.24
	4^1A_1	R	8.27	8.98	9.68	10.03	10.36	10.92	11.78
	5^1B_2	R	7.72	8.45	9.20	9.59	9.97	10.72	11.85
acetaldehyde	$1^1A''$	V	4.14	4.29	4.44	4.51	4.58	4.72	5.04
	$2^1A'$	R	5.46	6.00	6.50	6.74	6.97	7.42	7.86
	$3^1A'$	R	5.94	6.56	7.15	7.43	7.71	8.23	8.76
	$4^1A'$	R	6.31	6.95	7.55	7.84	8.11	8.63	9.09
	$6^1A'$	R	6.89	7.55	8.18	8.48	8.78	9.11	9.53
	$7^1A'$	R	7.04	7.75	8.44	8.78	9.03	9.37	10.01
acetone	1^1A_2	V	4.23	4.40	4.61	4.64	4.72	4.87	5.19
	1^1B_2	R	4.96	5.50	5.99	6.24	6.47	6.91	7.31
	2^1A_2	R	5.92	6.55	7.14	7.44	7.62	7.66	7.89
	2^1A_1	R	5.78	6.43	7.02	7.31	7.59	8.10	8.59
	2^1B_2	R	6.03	6.69	7.31	7.62	7.91	8.43	8.90
	3^1A_1	R	6.51	7.20	7.85	8.17	8.46	9.01	9.47
	3^1B_2	R	6.27	6.91	7.48	7.77	8.03	8.55	8.97
	1^1B_1	R	6.56	7.27	7.95	8.26	8.33	8.40	8.63
ethylene	1^1B_{3u}	R	6.44	6.84	7.20	7.40	7.57	7.91	8.05
	1^1B_{1u}	V	7.64	7.96	8.18	8.31	8.40	8.53	8.53
	1^1B_{1g}	R	6.98	7.44	7.78	7.81	7.84	7.89	8.17
	1^1B_{2g}	R	6.95	7.43	7.87	8.11	8.32	8.74	8.99
	2^1A_g	R	7.39	7.79	8.16	8.37	8.56	8.93	9.24
	2^1B_{3u}	R	7.77	8.27	8.68	8.90	9.09	9.43	9.67
	3^1B_{3u}	R	7.96	8.40	8.85	9.10	9.34	9.81	10.21
	4^1B_{3u}	R	8.26	8.75	9.23	9.50	9.75	10.24	10.80
	3^1B_{1g}	R	8.55	9.02	9.45	9.68	9.89	10.30	10.72
	2^1B_{1u}	R	8.64	9.00	9.36	9.58	9.78	10.19	10.71
	5^1B_{3u}	R	9.28	9.74	10.15	10.38	10.59	10.99	11.45
isobutene	$^1B_1$	R	5.36	5.73	6.08	6.25	6.40	6.71	6.85
	$^1A_1$	R	6.07	6.47	6.82	6.98	7.12	7.37	7.53
	1^1B_u	V	5.74	5.87	5.96	6.02	6.06	6.13	6.12

trans-butadiene	1^1B_g	R	5.52	5.90	6.23	6.42	6.59	6.91	7.01
	2^1A_u	R	5.83	6.24	6.61	6.77	6.96	7.30	7.44
	2^1B_u	R	6.67	6.95	7.23	7.41	7.57	7.88	8.13
	2^1B_g	R	6.51	6.97	7.38	7.61	7.72	7.76	8.02
	3^1A_g	R	6.85	7.23	7.57	7.78	7.96	8.30	8.49
	3^1B_u	R	7.50	7.93	8.33	8.56	8.77	9.17	9.57
pyridine	$^1B_1$	V	4.34	4.43	4.52	4.57	4.61	4.69	4.96
	$^1B_2$	V	5.46	5.49	5.51	5.55	5.56	5.59	5.58
	$^1A_2$	V	4.40	4.52	4.63	4.70	4.75	4.86	5.15
	$^1A_1$	V	6.72	6.73	6.69	6.73	6.74	6.76	6.73
pyrazine	$^1B_{3u}$	V	3.55	3.61	3.68	3.69	3.71	3.76	4.01
	$^1B_{2u}$	V	5.43	5.45	5.46	5.47	5.48	5.48	5.45
	$^1B_{2g}$	V	5.09	5.18	5.31	5.31	5.35	5.43	5.64
	$^1B_{1g}$	V	5.53	5.66	5.81	5.86	5.93	6.05	6.32
	$^1B_{1u}$	V	6.96	6.98	6.95	6.98	7.00	7.02	7.00
pyridazine	$^1B_1$	V	3.09	3.19	3.26	3.33	3.38	3.47	3.76
	$^1A_1$	V	5.56	5.60	5.63	5.65	5.67	5.70	5.69
	$^1A_2$	V	4.97	5.05	5.16	5.16	5.20	5.26	5.50
	$^1B_1$	V	5.37	5.48	5.59	5.63	5.69	5.78	6.05
	$^1B_2$	V	6.77	6.75	6.83	6.73	6.78	6.80	6.73
pyrimidine	$^1B_1$	V	3.71	3.80	3.89	3.94	3.98	4.07	4.35
	$^1A_2$	V	3.93	4.05	4.15	4.22	4.28	4.38	4.69
	$^1B_2$	V	5.69	5.74	5.77	5.80	5.81	5.85	5.86
	$^1A_2$	V	5.05	5.14	5.22	5.26	5.30	5.38	5.63
	$^1B_1$	V	5.28	5.39	5.49	5.55	5.60	5.70	5.98
	$^1A_1$	V	6.96	6.95	6.94	6.99	7.01	7.03	7.01
s-tetrazine	$^1B_{3u}$	V	1.77	1.84	1.90	1.93	1.96	2.02	2.29
	^1A_u	V	2.77	2.89	2.95	3.06	3.11	3.21	3.55
	^1A_u	V	4.58	4.61	4.70	4.66	4.67	4.70	4.90
	$^1B_{3u}$	V	5.60	5.69	5.79	5.82	5.87	5.95	6.22
Mean absolute error (total)			0.83	0.50	0.25	0.26	0.37	0.60	0.85
Mean absolute error (valence)			0.42	0.36	0.33	0.33	0.33	0.33	0.36
Mean absolute error (Rydberg)			1.14	0.61	0.19	0.21	0.40	0.80	1.23

6. Caricato set – TDDFT results from PBE0KLI-0.4 and PBE0HF-0.4

Table S6. The excitation energies of the Caricato set calculated using PBE0KLI-0.4. The third column indicates excitation types: V and R for vertical and Rydberg excitations, respectively. Experimental values were obtained from [M. Caricato, G. W. Trucks, M. J. Frisch, K. B. Wiberg, *J. Chem. Theory Comput.* **2010**, *6* (2), 370–383]. Unit: eV

Molecule	State	Type	PBE0KLI-0.4 (Orbital energy gaps)	PBE0KLI-0.4 w/ LDA kernel (TDDFT)	the portion of the major configuration in TDDFT calculation ($\|c\|^2$)	Expt.
formaldehyde	1^1A_2	V	3.76	4.07	1.00	4.00
	1^1B_2	R	6.95	6.95	1.00	7.08
	2^1B_2	R	7.76	7.79	0.99	7.97
	2^1A_1	R	7.90	7.90	0.99	8.14
	2^1A_2	R	8.12	8.08	1.00	8.37
	3^1B_2	R	8.74	8.80	0.97	8.88
	1^1B_1	V	8.47	8.96	0.94	9.00
	3^1A_2	R	9.14	9.11	1.00	9.22
	4^1B_2	R	9.04	9.01	1.00	9.26
	4^1A_1	R	9.64	9.68	0.96	9.58
	5^1B_2	R	9.20	9.20	0.98	9.63
acetaldehyde	$1^1A''$	V	4.14	4.44	1.00	4.28
	$2^1A'$	R	6.42	6.50	0.99	6.82
	$3^1A'$	R	7.12	7.15	0.99	7.46
	$4^1A'$	R	7.55	7.55	0.98	7.75
	$6^1A'$	R	8.19	8.18	0.98	8.43
	$7^1A'$	R	8.47	8.44	0.99	8.69
acetone	1^1A_2	V	4.32	4.61	1.00	4.43
	1^1B_2	R	5.97	5.99	1.00	6.36
	2^1A_2	R	7.14	7.14	1.00	7.36
	2^1A_1	R	6.96	7.02	0.98	7.41
	2^1B_2	R	7.31	7.31	1.00	7.49
	3^1A_1	R	7.85	7.85	1.00	7.80
	3^1B_2	R	7.45	7.48	0.99	8.09
	1^1B_1	R	7.96	7.95	0.99	8.17
ethylene	1^1B_{3u}	R	7.21	7.20	1.00	7.11
	1^1B_{1u}	V	5.79	8.18	0.69	7.65
	1^1B_{1g}	R	7.89	7.78	0.74	7.80
	1^1B_{2g}	R	7.86	7.87	1.00	7.90
	2^1A_g	R	8.10	8.16	0.99	8.28
	2^1B_{3u}	R	8.68	8.68	0.99	8.62
	3^1B_{3u}	R	8.86	8.85	0.99	8.90
	4^1B_{3u}	R	9.23	9.23	1.00	9.08
	3^1B_{1g}	R	9.44	9.45	1.00	9.20
	2^1B_{1u}	R	9.06	9.36	0.69	9.33
	5^1B_{3u}	R	10.12	10.15	0.92	9.51

isobutene	$^1B_1$	R	6.08	6.08	1.00	6.17
	$^1A_1$	R	5.43	6.82	0.52	6.70
trans-butadiene	1^1B_u	V	4.02	5.96	0.82	5.91
	1^1B_g	R	6.26	6.23	1.00	6.22
	2^1A_u	R	6.60	6.61	0.99	6.66
	2^1B_u	R	7.04	7.23	0.87	7.07
	2^1B_g	R	7.40	7.38	0.99	7.36
	3^1A_g	R	7.58	7.57	0.96	7.62
	3^1B_u	R	8.36	8.33	0.99	8.00
pyridine	$^1B_1$	V	4.18	4.52	0.99	4.59
	$^1B_2$	V	4.83	5.51	0.67	4.99
	$^1A_2$	V	4.58	4.63	1.00	5.43
	$^1A_1$	V	5.23	6.69	0.55	6.38
pyrazine	$^1B_{3u}$	V	3.32	3.68	1.00	3.83
	$^1B_{2u}$	V	4.43	5.46	0.80	4.81
	$^1B_{2g}$	V	4.89	5.31	0.98	5.46
	$^1B_{1g}$	V	5.74	5.81	1.00	6.10
	$^1B_{1u}$	V	5.82	6.95	0.61	6.51
pyridazine	$^1B_1$	V	2.87	3.26	0.99	3.60
	$^1A_1$	V	4.90	5.63	0.64	5.00
	$^1A_2$	V	4.88	5.16	0.99	5.30
	$^1B_1$	V	5.49	5.59	0.99	6.00
	$^1B_2$	V	5.52	6.83	0.37	6.50
pyrimidine	$^1B_1$	V	3.67	3.89	0.99	3.85
	$^1A_2$	V	4.06	4.15	0.98	4.62
	$^1B_2$	V	5.03	5.77	0.66	5.12
	$^1A_2$	V	4.91	5.22	0.97	5.52
	$^1B_1$	V	5.30	5.49	0.99	5.90
	$^1A_1$	V	5.42	6.94	0.54	6.70
s-tetrazine	$^1B_{3u}$	V	1.55	1.90	1.00	2.25
	^1A_u	V	2.83	2.95	0.99	3.40
	^1A_u	V	4.38	4.70	0.99	5.00
	$^1B_{3u}$	V	5.65	5.79	0.99	6.34
Mean absolute error (total)			0.399	0.253	Average of $\|c\|^2$: 0.92	
Mean absolute error (valence)			0.617	0.334		
Mean absolute error (Rydberg)			0.231	0.191		

Table S7. The excitation energies of the Caricato set calculated using PBE0HF-0.4. The third column indicates excitation types: V and R for vertical and Rydberg excitations, respectively. Experimental values were obtained from [M. Caricato, G. W. Trucks, M. J. Frisch, K. B. Wiberg, *J. Chem. Theory Comput.* **2010**, *6* (2), 370–383]. Unit: eV

Molecule	State	Type	PBE0HF-0.4 (Orbital energy gaps)	PBE0HF-0.4 (TDDFT)	the portion of the major configuration in TDDFT calculation ($\|c\|^2$)	Expt.
formaldehyde	1^1A_2	V	8.08	3.98	0.96	4.00
	1^1B_2	R	8.83	7.25	0.61	7.08
	2^1B_2	R	9.07	7.99	0.34	7.97
	2^1A_1	R	9.02	8.04	0.39	8.14
	2^1A_2	R	9.10	8.22	0.53	8.37
	3^1B_2	R	9.12	8.45	0.38	8.88
	1^1B_1	V	13.25	9.17	0.96	9.00
	3^1A_2	R	9.65	8.72	0.46	9.22
	4^1B_2	R	9.50	8.64	0.67	9.26
	4^1A_1	R	9.57	8.75	0.62	9.58
	5^1B_2	R	9.61	8.78	0.41	9.63
acetaldehyde	$1^1A''$	V	8.37	4.33	0.97	4.28
	$2^1A'$	R	8.45	7.04	0.63	6.82
	$3^1A'$	R	8.65	7.58	0.23	7.46
	$4^1A'$	R	8.70	7.79	0.20	7.75
	$6^1A'$	R	8.87	8.23	0.41	8.43
	$7^1A'$	R	8.87	8.26	0.29	8.69
acetone	1^1A_2	V	8.31	4.50	0.49	4.43
	1^1B_2	R	8.11	6.68	0.73	6.36
	2^1A_2	R	8.42	7.50	0.35	7.36
	2^1A_1	R	8.36	7.48	0.44	7.41
	2^1B_2	R	8.40	7.55	0.34	7.49
	3^1A_1	R	8.50	7.96	0.54	7.80
	3^1B_2	R	8.41	7.79	0.25	8.09
	1^1B_1	R	8.49	7.98	0.67	8.17
ethylene	1^1B_{3u}	R	8.53	7.04	0.49	7.11
	1^1B_{1u}	V	8.87	7.53	0.70	7.65
	1^1B_{1g}	R	8.65	7.59	0.44	7.80
	1^1B_{2g}	R	8.65	7.63	0.47	7.90
	2^1A_g	R	8.76	7.87	0.67	8.28
	2^1B_{3u}	R	8.76	8.10	0.50	8.62
	3^1B_{3u}	R	8.89	8.26	0.78	8.90
	4^1B_{3u}	R	9.25	8.40	0.46	9.08
	3^1B_{1g}	R	8.92	8.30	0.66	9.20
	2^1B_{1u}	R	9.00	8.32	0.69	9.33
	5^1B_{3u}	R	9.80	8.71	0.72	9.51
isobutene	$^1B_1$	R	7.62	6.22	0.63	6.17
	$^1A_1$	R	7.80	6.68	0.29	6.70

trans-butadiene	1^1B_u	V	6.93	5.77	0.98	5.91
	1^1B_g	R	7.37	6.10	0.54	6.22
	2^1A_u	R	7.48	6.37	0.42	6.66
	2^1B_u	R	7.60	6.82	0.71	7.07
	2^1B_g	R	7.57	6.97	0.33	7.36
	3^1A_g	R	7.74	7.12	0.75	7.62
	3^1B_u	R	7.88	7.28	0.44	8.00
pyridine	$^1B_1$	V	8.05	5.11	0.97	4.59
	$^1B_2$	V	7.80	5.64	0.77	4.99
	$^1A_2$	V	8.44	5.67	0.99	5.43
	$^1A_1$	V	8.20	6.38	0.64	6.38
pyrazine	$^1B_{3u}$	V	7.22	4.21	0.99	3.83
	$^1B_{2u}$	V	7.46	5.46	0.90	4.81
	$^1B_{2g}$	V	9.10	5.92	0.95	5.46
	$^1B_{1g}$	V	9.93	7.23	0.96	6.10
	$^1B_{1u}$	V	9.09	6.57	0.77	6.51
pyridazine	$^1B_1$	V	6.90	3.89	0.98	3.60
	$^1A_1$	V	8.00	5.78	0.74	5.00
	$^1A_2$	V	8.90	5.86	0.97	5.30
	$^1B_1$	V	9.53	6.66	0.97	6.00
	$^1B_2$	V	8.63	6.51	0.54	6.50
pyrimidine	$^1B_1$	V	7.60	4.63	0.99	3.85
	$^1A_2$	V	7.99	5.08	0.98	4.62
	$^1B_2$	V	8.13	5.91	0.76	5.12
	$^1A_2$	V	9.01	6.12	0.97	5.52
	$^1B_1$	V	9.39	6.53	0.98	5.90
	$^1A_1$	V	8.51	6.68	0.73	6.70
s-tetrazine	$^1B_{3u}$	V	5.68	2.51	1.00	2.25
	^1A_u	V	6.99	4.03	0.99	3.40
	^1A_u	V	8.53	5.45	0.99	5.00
	$^1B_{3u}$	V	9.84	6.90	0.99	6.34
Mean absolute error (total)			1.743	0.376	Average of $\|c\|^2$: 0.67	
Mean absolute error (valence)			3.142	0.405		
Mean absolute error (Rydberg)			0.667	0.353		

7. Caricato set – Orbital energy gaps and TDDFT results from BH&H (KLI)

Table S8. The excitation energies of the Caricato set calculated using BH&H (KLI). The third column indicates excitation types: V and R for vertical and Rydberg excitations, respectively. Experimental values were obtained from [M. Caricato, G. W. Trucks, M. J. Frisch, K. B. Wiberg, *J. Chem. Theory Comput.* **2010**, *6* (2), 370–383]. Unit: eV

Molecule	State	Type	BH&H (KLI) (Orbital energy gaps)	BH&H (KLI) w/ LDA kernel (TDDFT)	Expt.
formaldehyde	1^1A_2	V	3.73	4.05	4.00
	1^1B_2	R	6.89	6.90	7.08
	2^1B_2	R	7.66	7.71	7.97
	2^1A_1	R	7.91	7.92	8.14
	2^1A_2	R	8.13	8.09	8.37
	3^1B_2	R	8.74	8.82	8.88
	1^1B_1	V	8.49	9.00	9.00
	3^1A_2	R	9.22	9.19	9.22
	4^1B_2	R	9.09	9.06	9.26
	4^1A_1	R	9.71	9.75	9.58
	5^1B_2	R	9.29	9.28	9.63
acetaldehyde	$1^1A''$	V	4.18	4.47	4.28
	$2^1A'$	R	6.34	6.43	6.82
	$3^1A'$	R	7.10	7.14	7.46
	$4^1A'$	R	7.51	7.53	7.75
	$6^1A'$	R	8.20	8.19	8.43
	$7^1A'$	R	8.53	8.51	8.69
acetone	1^1A_2	V	4.39	4.67	4.43
	1^1B_2	R	5.85	5.88	6.36
	2^1A_2	R	7.15	7.15	7.36
	2^1A_1	R	6.96	7.04	7.41
	2^1B_2	R	7.30	7.31	7.49
	3^1A_1	R	7.89	7.90	7.80
	3^1B_2	R	7.45	7.48	8.09
	1^1B_1	R	8.03	8.02	8.17
ethylene	1^1B_{3u}	R	7.08	7.08	7.11
	1^1B_{1u}	V	5.75	8.05	7.65
	1^1B_{1g}	R	7.84	7.66	7.80
	1^1B_{2g}	R	7.80	7.81	7.90
	2^1A_g	R	8.04	8.12	8.28
	2^1B_{3u}	R	8.60	8.60	8.62
	3^1B_{3u}	R	8.84	8.83	8.90
	4^1B_{3u}	R	9.22	9.23	9.08
	3^1B_{1g}	R	9.42	9.43	9.20
	2^1B_{1u}	R	9.07	9.33	9.33
	5^1B_{3u}	R	10.04	10.07	9.51
isobutene	$^1B_1$	R	5.88	5.89	6.17
	$^1A_1$	R	5.39	6.71	6.70

trans-butadiene	1^1B_u	V	4.01	5.87	5.91
	1^1B_g	R	6.12	6.09	6.22
	2^1A_u	R	6.49	6.50	6.66
	2^1B_u	R	6.95	7.17	7.07
	2^1B_g	R	7.32	7.31	7.36
	3^1A_g	R	7.50	7.51	7.62
	3^1B_u	R	8.30	8.27	8.00
pyridine	$^1B_1$	V	4.19	4.53	4.59
	$^1B_2$	V	4.81	5.49	4.99
	$^1A_2$	V	4.60	4.65	5.43
	$^1A_1$	V	5.22	6.63	6.38
pyrazine	$^1B_{3u}$	V	3.31	3.66	3.83
	$^1B_{2u}$	V	4.41	5.43	4.81
	$^1B_{2g}$	V	4.91	5.33	5.46
	$^1B_{1g}$	V	5.78	5.85	6.10
	$^1B_{1u}$	V	5.83	6.89	6.51
pyridazine	$^1B_1$	V	2.91	3.29	3.60
	$^1A_1$	V	4.90	5.63	5.00
	$^1A_2$	V	4.90	5.18	5.30
	$^1B_1$	V	5.51	5.62	6.00
	$^1B_2$	V	5.51	6.79	6.50
pyrimidine	$^1B_1$	V	3.71	3.93	3.85
	$^1A_2$	V	4.10	4.19	4.62
	$^1B_2$	V	5.02	5.77	5.12
	$^1A_2$	V	4.92	5.22	5.52
	$^1B_1$	V	5.31	5.50	5.90
	$^1A_1$	V	5.41	6.89	6.70
s-tetrazine	$^1B_{3u}$	V	1.59	1.93	2.25
	^1A_u	V	2.86	2.98	3.40
	^1A_u	V	4.37	4.68	5.00
	$^1B_{3u}$	V	5.64	5.78	6.34
Mean absolute error (total)			0.402	0.250	
Mean absolute error (valence)			0.607	0.316	
Mean absolute error (Rydberg)			0.244	0.199	