Supporting Information:

Excitation energies of simple cyanine dyes

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(Dated: December 21, 2010)

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Experimental results:

Chain length	(R$_1$,R$_2$)		
	(H, H)	(H, Me)	(Me, Me)
3			5.54[d]
5	4.34[b]	4.20[b], 4.19[c]	3.97[a], 4.01[c], 3.96[d]
7	3.28[b]	3.15[b], 3.14[c]	3.01[e], 2.99[a], 3.02[c], 2.98[d]
9		2.53[b], 2.51[c]	2.40[a], 2.44[c], 2.39[d]
11			1.96[a], 2.03[c], 1.98[d]
3			224[d]
5	286[b]	295[b], 296[c]	312[a], 309[c], 313[d]
7	378[b]	394[b], 395[c]	412[e], 414[a], 411[c], 416[d]
9		491[b], 493[c]	516[a], 509[c], 519[d]
11			632[a], 611[c], 625[d]

(a) Ref. 1, measured in Methylendichloride (9.1, 20.0°C).

(b) Ref. 2, measured in H$_2$O (80.4, 20.0°C).

(c) Ref. 3, measured in Methanol (32.6, 25°C).

(d) Ref. 4, measured in Methylendichloride (9.1, 20.0°C).

(e) Ref. 5, measured in Ethanol (24.3, 25°C).

TABLE I. Experimental absorption maximum in eV (above) and nm (below) of the cyanine dye series for different solutions and substitutions at the nitrogen atoms (R$_1$,R$_2$). The dielectric constant and the corresponding experimental temperature are given in brackets.

Coupled-Cluster results:

	aug-cc-pVDZ	aug-cc-pVTZ	aug-cc-pVQZ	aug-cc-pV5Z
Carbon atom				
ANO-L-VDZP	0.0069	0.0030	0.0005	
ANO-L-VTZP		0.2879	0.0025	
ANO-L-VQZP			0.0119	0.0015
Nitrogen atom				
ANO-L-VDZP	0.0124	0.0032	0.0006	
ANO-L-VTZP		0.6557	0.0037	
ANO-L-VQZP			0.0229	0.0006
H_2				
ANO-L-VDZP	2.0923	0.0361	0.0007	
ANO-L-VTZP		5.1242	0.0425	
ANO-L-VQZP			0.0425	0.0035

TABLE II. Error in the correlation energy introduced by the RI-approximation when the ANO-L-VXZP basis sets are used in combination with aug-cc-pVXZ auxiliary basis sets. The quantity listed is $\alpha = \delta_{RI}/|\Delta E(MP2)|$ in ppm, where $\Delta E(MP2)$ is the MP2 energy correction to the Hartree-Fock energy and $\delta_{RI} = \frac{1}{4}| < ab||ij >_{\text{exact}} - < ab||ij >_{\text{RI}} |^2/(\epsilon_a - \epsilon_i + \epsilon_b - \epsilon_j)$. Calculations are performed on the C and N atoms, and H_2.

Molecule	Basis	CCS	CC2	CCSD	CC3	exCC3	ΔCC3
cn3	ANO-L-VDZP	8.71	7.36	7.32	7.27		-0.10
	ANO-L-VTZP	8.61	7.26	7.29	7.18	7.16	-0.08
	ANO-L-VQZP	8.61	7.26	7.30	7.18		-0.08
cn5	ANO-L-VDZP	6.07	5.02	4.98	4.89		-0.13
	ANO-L-VTZP	6.05	4.97	4.98	4.86	4.84	-0.11
	ANO-L-VQZP	6.05	4.96	4.99			
cn7	ANO-L-VDZP	4.77	3.83	3.79	3.69		-0.14
	ANO-L-VTZP	4.76	3.79	3.81		3.65	
cn9	ANO-L-VDZP	3.98	3.13	3.09	2.99		-0.14
	ANO-L-VTZP	3.97	3.10	3.11		2.96	
cn11	ANO-L-VDZP	3.45	2.66	2.62	2.52		-0.14
	ANO-L-VTZP					2.53	

TABLE III. Vertical excitation energies (eV) for the 1^1B_1 state of the cyanine dye series computed with CCS, CC2, and CC3, and the ANO-L-VXZP basis sets. ΔCC3 denotes the difference between the CC2 and CC3 excitation energies. The extrapolated CC3 values (exCC3) are obtained by adding the double-zeta ΔCC3 corrections to the triple-zeta CC2 results. Calculations are performed *without* the RI-approximation and the ground-state RI-MP2/cc-pVQZ structures are employed.

Molecule	2^1A_1	3^1A_1	1^1A_2	2^1A_2	$\mathbf{1^1B_1}$	2^1B_1	1^1B_2	2^1B_2
cn3	0.11778	0.00945	0.00000	0.00000	0.38783	0.00849	0.04360	0.00386
cn5	0.01641	0.02727	0.00000	0.00000	0.84739	0.06856	0.00004	0.00018
cn7	0.03147	0.00177	0.00000	0.00000	1.31952	0.00091	0.00129	0.00011
cn9	0.01095	0.02281	0.00000	0.00000	1.80588	0.00227	0.00005	0.00062
cn11	0.00572	0.03081	0.00000	0.00000	2.29771	0.00306	0.00098	0.00039

TABLE IV. Oscillator strengths of the two lowest excitation energies in each symmetry block calculated at the RI-CC2/ANO-L-VTZP level.

Molecule	Basis	2^1A_1	3^1A_1	1^1A_2	2^1A_2	$\mathbf{1^1B_1}$	2^1B_1	1^1B_2	2^1B_2
cn3	ANO-L-VDZP	11.20	13.55	8.50	9.55	7.36	13.03	9.75	10.94
	ANO-L-VTZP	11.04	13.25	8.40	9.22	7.26	10.81	9.43	10.72
	ANO-L-VQZP	11.01	12.23	8.45	9.27	7.26	10.68	9.48	10.70
cn5	ANO-L-VDZP	7.46	8.70	7.42	8.80	5.02	9.52	7.15	7.60
	ANO-L-VTZP	7.13	8.40	7.18	8.34	4.97	9.38	6.95	7.18
	ANO-L-VQZP	7.08	8.27	7.23	8.39	4.96	9.26	7.01	7.24
cn7	ANO-L-VDZP	6.38	6.60	6.40	6.74	3.83	7.07	6.61	7.86
	ANO-L-VTZP	6.30	6.42	6.11	6.26	3.79	6.66	6.34	7.09
	ANO-L-VQZP	6.28	6.40	6.17	6.32	3.79	6.55	6.39	7.05
cn9	ANO-L-VDZP	5.26	5.74	6.12	7.02	3.13	6.57	5.99	6.29
	ANO-L-VTZP	5.21	5.61	5.80	6.24	3.10	6.37	5.62	5.78
	ANO-L-VQZP	5.20	5.59	5.86	6.29	3.10	6.30	5.67	5.83
cn11	ANO-L-VDZP	4.49	5.08	5.73	6.00	2.66	5.81	5.81	6.52
	ANO-L-VTZP	4.45	4.99	5.31	5.47	2.64	5.74	5.46	5.76
	ANO-L-VQZP	4.44	4.97	5.35	5.52	2.63	5.71	5.50	5.81

TABLE V. Basis-set dependence of the RI-CC2 vertical excitation energies (eV) computed with the ANO-L-VXZP basis sets. The ground-state RI-MP2/cc-pVQZ structures are employed.

Molecule	Basis	2^1A_1	3^1A_1	1^1A_2	2^1A_2	$\mathbf{1^1B_1}$	2^1B_1	1^1B_2	2^1B_2
cn3	cc-pVDZ	11.43	14.07	9.09	9.68	7.78	15.00	10.20	11.14
	cc-pVTZ	11.18	13.72	8.84	9.58	7.54	14.56	9.91	10.85
	cc-pVQZ	11.10	13.57	8.72	9.50	7.41	13.41	9.77	10.76
cn5	cc-pVDZ	7.99	8.85	8.02	8.90	5.16	9.64	7.81	8.21
	cc-pVTZ	7.61	8.69	7.72	8.69	5.06	9.47	7.66	7.83
	cc-pVQZ	7.41	8.63	7.57	8.63	5.01	9.42	7.53	7.62
cn7	cc-pVDZ	6.48	6.85	7.10	7.47	3.91	7.70	7.48	7.86
	cc-pVTZ	6.37	6.65	6.89	7.09	3.84	7.21	7.11	7.76
	cc-pVQZ	6.33	6.55	6.73	6.87	3.81	6.96	6.91	7.58
cn9	cc-pVDZ	5.32	5.87	7.11	7.32	3.18	6.70	6.75	7.09
	cc-pVTZ	5.24	5.74	6.71	7.05	3.13	6.57	6.50	6.70
	cc-pVQZ	5.21	5.68	6.48	6.87	3.11	6.50	6.31	6.47
cn11	cc-pVDZ	4.54	5.16	6.55	6.88	2.69	5.88	6.83	6.95
	cc-pVTZ	4.47	5.06	6.27	6.46	2.66	5.79	6.44	6.64
	cc-pVQZ	4.45	5.02	6.06	6.21	2.64	5.76	6.19	6.44

TABLE VI. Basis-set dependence of the RI-CC2 excitation energies (in eV) computed with the Dunning cc-pVXZ basis sets. The ground-state RI-MP2/cc-pVQZ structures are employed.

Molecule	Basis	2^1A_1	3^1A_1	1^1A_2	2^1A_2	$\mathbf{1^1B_1}$	2^1B_1	1^1B_2	2^1B_2
cn3	aug-cc-pVDZ	11.10	12.43	8.25	9.07	7.28	10.50	9.28	10.66
	aug-cc-pVTZ	11.02	12.29	8.42	9.24	7.26	10.64	9.45	10.70
	aug-cc-pVQZ	11.01	12.22	8.49	9.30	7.26	10.68	9.52	10.68
cn5	aug-cc-pVDZ	7.04	8.22	7.04	8.19	5.00	9.35	6.81	7.05
	aug-cc-pVTZ	7.07	8.28	7.21	8.36	4.97	9.30	6.98	7.22
	aug-cc-pVQZ	7.09	8.29	7.27	8.42	4.96	9.28	7.05	7.28
cn7	aug-cc-pVDZ	6.32	6.45	5.97	6.12	3.82	6.46	6.20	6.86
	aug-cc-pVTZ	6.29	6.41	6.14	6.30	3.80	6.53	6.37	7.03
	aug-cc-pVQZ	6.28	6.40	6.20	6.36	3.79	6.56	6.43	7.09
cn9	aug-cc-pVDZ	5.25	5.62	5.66	6.09	3.12	6.28	5.47	5.63
	aug-cc-pVTZ	5.20	5.60	5.83	6.26	3.10	6.29	5.64	5.80
	aug-cc-pVQZ	5.19	5.59	5.89	6.32	3.09	6.30	5.70	5.86
cn11	aug-cc-pVDZ	4.48	5.01	5.15	5.31	2.66	5.74	5.30	5.61
	aug-cc-pVTZ	4.44	4.98	5.31	5.48	2.64	5.72	5.47	5.78
	aug-cc-pVQZ	4.44	4.97	5.38	5.54	2.63	5.71	5.54	5.84

TABLE VII. Basis-set dependence of the RI-CC2 excitation energies (eV) computed with the Dunning aug-cc-pVXZ basis sets. The ground-state RI-MP2/cc-pVQZ structures are employed.

Molecule	Structure	2^1A_1	3^1A_1	1^1A_2	2^1A_2	$\mathbf{1^1B_1}$	2^1B_1	1^1B_2	2^1B_2
cn3	MP2	11.04	13.25	8.40	9.22	7.26	10.81	9.43	10.72
	PBE0	11.03	13.22	8.39	9.20	7.26	10.81	9.41	10.68
	PBE	10.82	13.15	8.32	9.13	7.13	10.77	9.31	10.56
	HF	11.19	13.31	8.50	9.31	7.37	10.86	9.53	10.82
cn5	MP2	7.13	8.40	7.19	8.34	4.97	9.38	6.95	7.18
	PBE0	7.13	8.39	7.16	8.32	4.94	9.34	6.94	7.16
	PBE	7.07	8.29	7.12	8.27	4.84	9.16	6.92	7.14
	HF	7.16	8.44	7.21	8.37	5.00	9.46	6.97	7.22
cn7	MP2	6.30	6.42	6.11	6.26	3.79	6.66	6.34	7.00
	PBE0	6.27	6.41	6.12	6.26	3.77	6.67	6.33	6.99
	PBE	6.16	6.31	6.12	6.24	3.69	6.63	6.31	6.96
	HF	6.34	6.47	6.13	6.30	3.82	6.69	6.37	7.03
cn9	MP2	5.21	5.61	5.80	6.24	3.10	6.37	5.62	5.78
	PBE0	5.17	5.60	5.81	6.23	3.08	6.37	5.63	5.78
	PBE	5.07	5.51	5.79	6.22	3.01	6.28	5.64	5.76
	HF	5.22	5.68	5.85	6.29	3.12	6.42	5.66	5.84
cn11	MP2	4.45	4.99	5.31	5.47	2.64	5.74	5.46	5.76
	PBE0	4.41	4.98	5.33	5.48	2.62	5.71	5.47	5.77
	PBE	4.33	4.89	5.34	5.46	2.56	5.61	5.46	5.75
	HF	4.45	5.07	5.36	5.56	2.66	5.78	5.53	5.85

TABLE VIII. Dependence of the excitation energies (eV) on the method used to optimize the ground-state structure. The excitation energies are calculated at RI-CC2/ANO-L-VTZP level.

Molecule	2^1A_1	3^1A_1	1^1A_2	2^1A_2	$\mathbf{1^1B_1}$	2^1B_1	1^1B_2	2^1B_2
me2-cn3	9.04	9.33	6.72	7.71	6.07	8.25	7.98	8.90
me2-cn5	6.67	7.12	6.77	7.76	4.58	7.95	6.02	6.45
me2-cn7	5.85	6.22	5.46	5.68	3.53	6.22	5.97	6.68
me2-cn9	4.92	5.37	5.46	5.93	2.90	6.09	5.12	5.32
me2-cn11	4.25	4.75	4.90	5.10	2.47	5.49	5.14	5.47

TABLE IX. Excitation energies (in eV) of the methylated streptocyanine series at RI-CC2 level using ANO-L-VTZP basis sets. Structures from ground state RI-MP2/cc-pVQZ optimizations are employed.

Molecule	vertical	adiabatic
cn3	7.26	6.29
cn5	4.97	4.64
cn7	3.79	3.65
cn9	3.10	3.01
cn11	2.64	2.58

TABLE X. Vertical and Adiabatic excitation energies for the 1^1B_1-state obtained using RI-CC2 along with ANO-L-VTZP basis sets. Optimizations have been restricted to C_{2v}-symmetry.

CASSCF, CASPT2 and QMC results:

Molecule	CAS(n,m)		Basis	CASSCF	CASPT2	
	$n\ [a_2, b_2]$	$m\ [a_2, b_2]$			0-IPEA	S-IPEA
cn3	4 [2, 2]	6 [2, 4]	ANO-L-VDZP	7.59	7.07	7.26
			ANO-L-VTZP	7.56	6.99	7.19
			ANO-L-VQZP	7.56	6.99	7.20
cn5	6 [2, 4]	10 [4, 6]	ANO-L-VDZP	5.25	4.53	4.74
			ANO-L-VTZP	5.32	4.46	4.69
			ANO-L-VQZP	5.32	4.46	4.69
cn7	8 [4, 4]	14 [6, 8]	ANO-L-VDZP	3.85	3.35	3.55
			ANO-L-VTZP	3.91	3.30	3.52
			ANO-L-VQZP[a]	3.92	3.30	3.53
cn9	10 [4, 6]	18 [8, 10]	ANO-L-VDZP[a]	3.08	2.63	2.83
			ANO-L-VTZP[a]	3.13	2.59	2.81
			ANO-L-VQZP[a]	3.14	2.59	2.81
cn11	12 [6, 6]	11 [5, 6]	ANO-L-VDZP[a]	2.37	2.13	2.46
			ANO-L-VTZP[a]	2.39	2.10	2.46

[a] Obtained with the Cholesky decomposition with 10^{-8} threshold.

TABLE XI. CASSCF and CASPT2 vertical excitation energies (eV) for the 1^1B_1 state of the cyanine dye series computed with the ANO-L-VXZP basis sets and the optimal active space. A CAS(n,m) expansion is used to compute the ground-state (1^1A_1) and excited-state (1^1B_1) energies, where n and m denote the number of electrons and molecular orbitals, respectively. The ground-state RI-MP2/cc-pVQZ structures are employed and the calculations are done in C_{2v} symmetry.

| Molecule | CAS(n,m) | | Basis | CASSCF | CASPT2 | |
	n $[a_2, b_2]$	m $[a_2, b_2]$			0-IPEA	S-IPEA
cn3	4 $[2, 2]$	6 $[2, 4]$	ANO-L-VDZP	7.59	7.07	7.26
			ANO-L-VTZP	7.56	6.99	7.19
			ANO-L-VQZP	7.56	6.99	7.20
			cc-pVDZ	8.30	7.38	7.60
			cc-pVTZ	8.02	7.16	7.39
			cc-pVQZ	7.85	7.07	7.30
			aug-cc-pVDZ	7.55	7.01	7.21
			aug-cc-pVTZ	7.56	6.99	7.20
cn5	6 $[2, 4]$	10 $[4, 6]$	ANO-L-VDZP	5.25	4.53	4.74
			ANO-L-VTZP	5.32	4.46	4.69
			ANO-L-VQZP	5.32	4.46	4.69
			cc-pVDZ	5.52	4.63	4.85
			cc-pVTZ	5.45	4.53	4.76
			cc-pVQZ	5.40	4.50	4.73
			aug-cc-pVDZ	5.30	4.51	4.72
			aug-cc-pVTZ	5.32	4.47	4.70

TABLE XII. CASSCF and CASPT2 vertical excitation energies (eV) for the 1^1B_1 state of the cyanine dyes cn3 and cn5 computed with the ANO-L-VXZP and cc-pVXZ basis sets and the optimal active space. A CAS(n,m) expansion is used to compute the ground-state (1^1A_1) and excited-state (1^1B_1) energies, where n and m denote the number of electrons and molecular orbitals, respectively. The ground-state RI-MP2/cc-pVQZ structures are employed and the calculations are done in C_{2v} symmetry.

Molecule	CAS(n,m)		Basis	CASSCF	CASPT2	
	n $[a_2, b_2]$	m $[a_2, b_2]$			0-IPEA	S-IPEA
cn3	4 $[2,2]$	3 $[1,2]$	ANO-L-VTZP	8.12	6.55	6.90
		4 $[2,2]$		8.35	6.48	6.82
		5 $[2,3]$		7.57	6.98	7.19
		6 $[2,4]^b$		7.56	6.99	7.19
		8 $[3,5]$		7.64	6.98	7.16
		9 $[3,6]$		7.63	6.97	7.14
cn5	6 $[2,4]$	5 $[2,3]$	ANO-L-VTZP	5.46	4.23	4.62
		6 $[2,4]$		5.55	4.24	4.56
		7 $[3,4]$		5.21	4.48	4.74
		8 $[3,5]$		5.24	4.48	4.72
		10 $[4,6]^b$		5.32	4.46	4.69
		15 $[6,9]$		5.33	4.49	4.68
cn7	8 $[4,4]$	7 $[3,4]$	ANO-L-VTZP	3.92	3.17	3.56
		8 $[4,4]$		4.02	3.12	3.43
		14 $[6,8]^b$		3.91	3.30	3.52
		21 $[9,12]$		3.96	3.30	3.49
cn9	10 $[4,6]$	9 $[4,5]$	ANO-L-VTZP^a	2.99	2.55	2.92
		10 $[4,6]$		3.08	2.48	2.80
		18 $[8,10]^b$		3.13	2.59	2.81
cn11	12 $[6,6]$	11 $[5,6]^b$	ANO-L-VTZP^a	2.39	2.10	2.46
		12 $[6,6]$		2.45	2.07	2.39
		13 $[6,7]$		2.36	2.12	2.43
		15 $[7,8]$		2.40	2.14	2.42
		17 $[8,9]$		2.42	2.13	2.40

[a] Obtained with the Cholesky decomposition with 10^{-8} threshold.

[b] Optimal CAS active space.

TABLE XIII. CASSCF and CASPT2 vertical excitation energies (eV) for the 1^1B_1 state of the cyanine dye series computed with the ANO-L-VTZP basis sets and different active spaces. A CAS(n,m) expansion is used to compute the ground-state(1^1A_1) and excited-state (1^1B_1) energies, where n and m denote the number of electrons and molecular orbitals, respectively. The optimal active space is denoted with a ^{b}. The ground-state RI-MP2/cc-pVQZ structures are employed and the calculations are done in C_{2v} symmetry.

	CAS(n,m)		CASSCF	VMC	DMC	
	n $[a_2, b_2]$	m $[a_2, b_2]$				
cn3	4 $[2, 2]$	6 $[2, 4]$	7.62	7.48(1)	7.38(2)	(a),(b)
cn5	6 $[2, 4]$	10 $[4, 6]$	5.30	5.09(1)	5.03(2)	(a),(c)
cn7	8 $[4, 4]$	14 $[6, 8]$	3.89	3.90(1)	3.83(2)	(c)
cn9	10 $[4, 6]$	9 $[4, 5]$	2.98	3.18(1)	3.09(2)	(c)
cn11	12 $[6, 6]$	11 $[5, 6]$	2.37	2.68(2)	2.62(2)	(d)

(a) Orbitals optimized including all external orbitals;

(b) Thr. of 0.0; (c) Thr. of 0.02; (d) Thr. of 0.04.

TABLE XIV. VMC and DMC vertical excitation energies (eV) for the 1^1B_1 state of the cyanine dye series. For each molecule, we show the best available value from the QMC calculations obtained using the T$'$+ basis set. A CAS(n,m) expansion is used to compute the ground-state (1^1A_1) and excited-state (1^1B_1) energies, where n and m denote the number of electrons and molecular orbitals, respectively. The threshold on the expansion is also listed and the corresponding number of CSFs and determinants are listed in Table XV. Unless indicated, only the Jastrow and CI parameters are optimized and the Jastrow factor includes only electron-nuclear and electron-electron terms. The ground-state RI-MP2/cc-pVQZ structures are employed and the calculations are done in C_{2v} symmetry.

	CAS(n,m)		Basis	Thr.	CSF/Det.		CASSCF	VMC	DMC	
	$n\,[a_2,b_2]$	$m\,[a_2,b_2]$			1^1A_1	1^1B_1				
cn3	4 [2, 2]	6 [2, 4]	T'+	0.02	7/11	8/22	7.62	7.58(1)	7.58(2)	
			T'+	0.02	7/11	8/22	7.62	7.63(1)	7.58(2)	(a)
			T'+	0.02	7/11	8/22	7.62	7.47(1)	7.40(2)	(b)
			D+	0.00	57/113	48/144	7.63	7.61(1)	7.50(2)	
			T'+	0.00	57/113	48/144	7.62	7.52(1)	7.48(2)	
			T'+	0.00	57/113	48/144	7.55	7.52(1)	7.48(2)	(c)
			Q'+	0.00	57/113	48/144	7.58	7.51(1)	7.46(2)	
			T'+	0.00	57/113	48/144	7.62	7.56(1)	7.47(2)	(a)
			T'+	0.00	57/113	48/144	7.62	7.48(1)	7.38(2)	(b)
cn5	6 [2, 4]	10 [4, 6]	T'+	0.08	4/7	5/12	5.30	5.21(1)	5.11(2)	
			T'+	0.04	8/17	14/38	5.30	5.13(1)	5.05(2)	
			D+	0.02	20/51	27/102	5.29	5.13(1)	5.08(2)	
			T'+	0.02	22/59	28/106	5.30	5.15(1)	5.04(2)	
			T'+	0.02	22/59	28/106	5.30	5.09(1)	5.03(2)	(b)
cn7	8 [4, 4]	14 [6, 8]	T'+	0.02	40/111	42/156	3.89	3.90(1)	3.83(2)	
cn9	10 [4, 6]	9 [4, 5]	T'+	0.04	13/39	17/42	2.98	3.22(1)	3.11(2)	
			T'+	0.02	43/101	65/254	2.98	3.18(1)	3.09(2)	
cn11	12 [6, 6]	11 [5, 6]	T'+	0.04	17/54	21/98	2.37	2.68(2)	2.62(2)	

(a) Including Jastrow e-e-n term;

(b) Orbitals optimized including all external orbitals;

(c) T'+ basis set with f functions.

TABLE XV. VMC and DMC vertical excitation energies (eV) for the 1^1B_1 state of the cyanine dye series. A CAS(n,m) expansion is used to compute the ground-state (1^1A_1) and excited-state (1^1B_1) energies, where n and m denote the number of electrons and molecular orbitals, respectively. The threshold on the expansion and the corresponding number of CSFs and determinants are also listed. Unless indicated, only the Jastrow and CI parameters are optimized and the Jastrow factor includes only electron-nuclear and electron-electron terms. The ground-state RI-MP2/cc-pVQZ structures are employed and the calculations are done in C_{2v} symmetry.

	CAS(n,m)		Basis	Thr.	VMC		
	$n\ [a_2, b_2]$	$m\ [a_2, b_2]$			1^1A_1	1^1B_1	
cn3	4 [2, 2]	6 [2, 4]	T′+	0.02	-28.2964(4)	-28.0170(4)	
			T′+	0.02	-28.3174(3)	-28.0370(3)	(a)
			T′+	0.02	-28.3018(4)	-28.0275(4)	(b)
			D+	0.00	-28.2924(4)	-28.0129(4)	
			T′+	0.00	-28.2974(4)	-28.0210(4)	
			T′+	0.00	-28.3022(4)	-28.0257(4)	(c)
			Q′+	0.00	-28.2979(4)	-28.0220(4)	
			T′+	0.00	-28.3190(3)	-28.0412(3)	(a)
			T′+	0.00	-28.3024(4)	-28.0275(4)	(b)
cn5	6 [2, 4]	10 [4, 6]	T′+	0.08	-40.8322(4)	-40.6407(4)	
			T′+	0.04	-40.8344(4)	-40.6460(4)	
			D+	0.02	-40.8266(4)	-40.6379(4)	
			T′+	0.02	-40.8371(4)	-40.6479(4)	
			T′+	0.02	-40.8456(4)	-40.6586(4)	(b)
cn7	8 [4, 4]	14 [6, 8]	T′+	0.02	-53.3703(4)	-53.2270(4)	
cn9	10 [4, 6]	9 [4, 5]	T′+	0.04	-65.8942(4)	-65.7758(4)	
			T′+	0.02	-65.9006(4)	-65.7838(4)	
cn11	12 [6, 6]	11 [5, 6]	T′+	0.04	-78.4218(4)	-78.3232(4)	

(a) Including Jastrow e-e-n term;

(b) Orbitals optimized including all external orbitals;

(c) T'+ basis set with f functions.

TABLE XVI. Absolute VMC energies (a.u.) for the ground-state (1^1A_1) and excited-state (1^1B_1) of the cyanine dye series. A CAS(n,m) expansion is used to compute the ground-state (1^1A_1) and excited-state (1^1B_1) energies, where n and m denote the number of electrons and molecular orbitals, respectively. The threshold on the expansion is also listed and the corresponding number of CSFs and determinants are listed in Table XV. Unless indicated, only the Jastrow and CI parameters are optimized and the Jastrow factor includes only electron-nuclear and electron-electron terms. The ground-state RI-MP2/cc-pVQZ structures are employed and the calculations are done in C_{2v} symmetry.

	CAS(n,m)		Basis	Thr.	DMC		
	$n\ [a_2, b_2]$	$m\ [a_2, b_2]$			1^1A_1	1^1B_1	
cn3	$4\ [2, 2]$	$6\ [2, 4]$	T′+	0.02	-28.3605(5)	-28.0820(5)	
			T′+	0.02	-28.3588(4)	-28.0802(4)	(a)
			T′+	0.02	-28.3607(5)	-28.0888(5)	(b)
			D+	0.00	-28.3581(5)	-28.0826(5)	
			T′+	0.00	-28.3597(5)	-28.0849(5)	
			T′+	0.00	-28.3612(5)	-28.0863(5)	(c)
			Q′+	0.00	-28.3611(5)	-28.0868(5)	
			T′+	0.00	-28.3587(5)	-28.0843(5)	(a)
			T′+	0.00	-28.3601(5)	-28.0889(5)	(b)
cn5	$6\ [2, 4]$	$10\ [4, 6]$	T′+	0.08	-40.9283(5)	-40.7407(5)	
			T′+	0.04	-40.9295(5)	-40.7441(5)	
			D+	0.02	-40.9270(5)	-40.7402(5)	
			T′+	0.02	-40.9307(5)	-40.7453(5)	
			T′+	0.02	-40.9325(5)	-40.7477(5)	(b)
cn7	$8\ [4, 4]$	$14\ [6, 8]$	T′+	0.02	-53.4956(6)	-53.3550(6)	
cn9	$10\ [4, 6]$	$9\ [4, 5]$	T′+	0.04	-65.0553(6)	-65.9410(6)	
			T′+	0.02	-65.0576(6)	-65.9440(6)	
cn11	$12\ [6, 6]$	$11\ [5, 6]$	T′+	0.04	-78.6150(6)	-78.5186(6)	

(a) Including Jastrow e-e-n term;

(b) Orbitals optimized including all external orbitals;

(c) T'+ basis set with f functions.

TABLE XVII. Absolute DMC energies (a.u.) for the ground-state (1^1A_1) and excited-state (1^1B_1) of the cyanine dye series. A CAS(n,m) expansion is used to compute the ground-state (1^1A_1) and excited-state (1^1B_1) energies, where n and m denote the number of electrons and molecular orbitals, respectively. The threshold on the expansion is also listed and the corresponding number of CSFs and determinants are listed in Table XV. Unless indicated, only the Jastrow and CI parameters are optimized and the Jastrow factor includes only electron-nuclear and electron-electron terms. The ground-state RI-MP2/cc-pVQZ structures are employed and the calculations are done in C_{2v} symmetry.

TDDFT results:

Molecule	Basis	2^1A_1	3^1A_1	1^1A_2	2^1A_2	$\mathbf{1^1B_1}$	2^1B_1	1^1B_2	2^1B_2
cn3	ANO-L-VDZP	11.08	12.79	8.51	9.29	7.73	12.71	9.58	10.14
	ANO-L-VTZP	10.99	12.53	8.30	8.96	7.62	10.76	9.20	10.08
	ANO-L-VQZP	10.99	12.00	8.31	8.97	7.63	10.60	9.22	10.08
cn5	ANO-L-VDZP	7.48	8.60	7.26	7.84	5.37	9.28	7.10	7.39
	ANO-L-VTZP	7.20	8.39	6.98	7.82	5.33	9.24	6.86	7.02
	ANO-L-VQZP	7.16	8.28	7.00	7.83	5.34	9.18	6.88	7.04
cn7	ANO-L-VDZP	6.07	6.74	6.33	6.56	4.21	7.01	6.45	6.86
	ANO-L-VTZP	6.04	6.62	6.05	6.16	4.18	6.67	6.17	6.83
	ANO-L-VQZP	6.04	6.61	6.06	6.17	4.19	6.58	6.18	6.84
cn9	ANO-L-VDZP	4.95	5.85	5.95	6.39	3.52	6.41	5.89	6.11
	ANO-L-VTZP	4.95	5.76	5.64	6.10	3.50	6.27	5.57	5.67
	ANO-L-VQZP	4.95	5.76	5.66	6.12	3.50	6.22	5.59	5.69
cn11	ANO-L-VDZP	4.18	5.18	5.60	5.83	3.05	5.59	5.63	6.15
	ANO-L-VTZP	4.18	5.13	5.27	5.36	3.03	5.57	5.30	5.65
	ANO-L-VQZP	4.19	5.13	5.28	5.37	3.04	5.57	5.31	5.67

TABLE XVIII. Basis-set dependence of the TDDFT/PBE0 excitation energies (eV) computed with the ANO-L-VXZP basis sets. The ground-state PBE0/cc-pVQZ structures are employed.

Molecule	Basis	2^1A_1	3^1A_1	1^1A_2	2^1A_2	$\mathbf{1^1B_1}$	2^1B_1	1^1B_2	2^1B_2
cn3	cc-pVDZ	11.21	13.07	8.79	9.28	8.02	14.03	9.80	10.22
	cc-pVTZ	11.13	12.87	8.63	9.21	7.87	13.75	9.58	10.17
	cc-pVQZ	11.10	12.73	8.49	9.12	7.78	13.33	9.43	10.13
cn5	cc-pVDZ	7.83	8.73	7.61	7.87	5.44	9.30	7.44	7.81
	cc-pVTZ	7.60	8.66	7.39	7.86	5.42	9.31	7.33	7.52
	cc-pVQZ	7.43	8.61	7.23	7.85	5.39	9.30	7.21	7.32
cn7	cc-pVDZ	6.10	6.88	6.76	7.00	4.24	7.49	6.87	7.00
	cc-pVTZ	6.12	6.81	6.59	6.72	4.24	7.14	6.73	6.87
	cc-pVQZ	6.09	6.74	6.43	6.52	4.22	6.92	6.54	6.86
cn9	cc-pVDZ	4.96	5.91	6.38	6.59	3.53	6.47	6.41	6.59
	cc-pVTZ	4.98	5.88	6.29	6.39	3.53	6.47	6.20	6.29
	cc-pVQZ	4.97	5.84	6.07	6.38	3.52	6.41	6.02	6.09
cn11	cc-pVDZ	4.18	5.21	6.19	6.34	3.05	5.59	6.14	6.30
	cc-pVTZ	4.20	5.20	5.95	6.03	3.06	5.62	5.99	6.15
	cc-pVQZ	4.20	5.18	5.75	5.82	3.05	5.61	5.77	6.09

TABLE XIX. Basis-set dependence of the TDDFT/PBE0 excitation energies (eV) computed with the Dunning cc-pVXZ basis sets. The ground-state PBE0/cc-pVQZ structures are employed.

Molecule	Basis	2^1A_1	3^1A_1	1^1A_2	2^1A_2	$\mathbf{1^1B_1}$	2^1B_1	1^1B_2	2^1B_2
cn3	aug-cc-pVDZ	10.91	12.34	8.23	8.89	7.57	10.62	9.12	10.04
	aug-cc-pVTZ	10.99	12.11	8.30	8.95	7.63	10.60	9.20	10.08
	aug-cc-pVQZ	10.99	11.97	8.30	8.96	7.63	10.57	9.21	10.08
cn5	aug-cc-pVDZ	7.12	8.33	6.93	7.79	5.28	9.15	6.82	6.97
	aug-cc-pVTZ	7.16	8.32	6.99	7.83	5.34	9.21	6.87	7.03
	aug-cc-pVQZ	7.15	8.27	6.99	7.83	5.34	9.19	6.88	7.03
cn7	aug-cc-pVDZ	5.98	6.55	6.02	6.12	4.14	6.54	6.13	6.79
	aug-cc-pVTZ	6.04	6.60	6.05	6.17	4.19	6.57	6.18	6.84
	aug-cc-pVQZ	6.04	6.60	6.06	6.17	4.19	6.57	6.18	6.84
cn9	aug-cc-pVDZ	4.90	5.70	5.61	6.07	3.47	6.18	5.55	5.65
	aug-cc-pVTZ	4.95	5.75	5.65	6.11	3.50	6.22	5.58	5.68
	aug-cc-pVQZ	4.95	5.75	5.65	6.11	3.50	6.21	5.59	5.69
cn11	aug-cc-pVDZ	4.14	5.08	5.24	5.34	3.00	5.51	5.27	5.63
	aug-cc-pVTZ	4.19	5.13	5.27	5.37	3.04	5.57	5.31	5.66
	aug-cc-pVQZ	4.19	5.13	5.27	5.37	3.03	5.57	5.31	5.66

TABLE XX. Basis-set dependence of the TDDFT/PBE0 excitation energies (eV) computed with the Dunning aug-cc-pVXZ basis sets. The ground-state PBE0/cc-pVQZ structures are employed.

Molecule	2^1A_1	3^1A_1	1^1A_2	2^1A_2	$\mathbf{1^1B_1}$	2^1B_1	1^1B_2	2^1B_2
cn3	10.63	11.54	7.71	8.33	7.40	10.18	8.55	9.73
cn5	6.71	7.85	6.41	7.24	5.22	8.33	6.33	6.53
cn7	5.23	6.43	5.52	5.70	4.11	6.16	5.60	6.19
cn9	4.18	5.57	5.08	5.61	3.44	5.50	5.05	5.24
cn11	3.47	4.97	4.72	4.94	2.98	4.73	4.73	5.18

TABLE XXI. TDDFT/PBE excitation energies (eV) computed with the ANO-L-VTZP basis sets. The ground-state PBE0/cc-pVQZ structures are employed.

Molecule	1^1A_1	2^1A_1	1^1A_2	2^1A_2	$\mathbf{1^1B_1}$	2^1B_1	1^1B_2	2^1B_2
me2-cn3	8.67	8.96	6.74	7.54	6.00	8.38	7.83	8.68
me2-cn5	6.69	7.00	6.63	7.58	4.75	7.91	6.09	6.40
me2-cn7	5.61	6.23	5.57	5.71	3.81	6.31	5.88	6.55
me2-cn9	4.69	5.46	5.41	5.86	3.23	5.97	5.23	5.35
me2-cn11	4.02	4.87	5.00	5.11	2.82	5.28	5.11	5.43

TABLE XXII. Excitation energies (in eV) of the methylated streptocyanine series at PBE0 level using ANO-L-VTZP basis sets. Structures from ground state PBE0 cc-pVQZ optimizations are employed.

Molecule	Basis	2^1A_1	3^1A_1	1^1A_2	2^1A_2	$\mathbf{1^1B_1}$	2^1B_1	1^1B_2	2^1B_2
cn3	ANO-L-VDZP	11.11	12.93	8.52	9.32	7.68	12.63	9.65	10.16
	ANO-L-VTZP	11.02	12.66	8.28	8.99	7.55	10.74	9.25	10.10
	ANO-L-VQZP	11.03	12.08	8.29	8.99	7.57	10.59	9.26	10.11
cn5	ANO-L-VDZP	7.59	8.91	7.40	8.00	5.31	9.64	7.19	7.43
	ANO-L-VTZP	7.22	8.48	7.10	7.99	5.26	9.55	6.93	7.06
cn7	ANO-L-VDZP	6.65	6.73	6.46	6.63	4.15	7.22	6.63	7.11
	ANO-L-VTZP	6.54	6.64	6.16	6.23	4.12	6.76	6.33	6.96
cn9	ANO-L-VDZP	5.55	5.89	6.17	6.69	3.46	6.95	6.06	6.21
	ANO-L-VTZP	5.53	5.79	5.85	6.24	3.44	6.60	5.70	5.79
cn11	ANO-L-VDZP	4.79	5.23	5.81	5.95	3.00	6.22	5.87	6.40
	ANO-L-VTZP								

TABLE XXIII. TDDFT/CAM-B3LYP excitation energies (eV) of the streptocyanine series computed with the ANO-L-VXZP basis sets. The ground-state PBE0/cc-pVQZ structures are employed.

Optimized ground state structures:

cn3	$N_1 - C_2$	$C_2 - N_3$
MP2	130.29	130.29
PBE0	130.34	130.34
PBE	131.46	131.46
HF	129.48	129.48

cn5	$N_1 - C_2$	$C_2 - C_3$	$C_3 - C_4$	$C_4 - N_5$
MP2	131.10	137.85	137.85	131.10
PBE0	131.19	138.10	138.10	131.19
PBE	132.42	138.94	138.94	132.42
HF	130.27	137.88	137.88	130.27

cn7	$N_1 - C_2$	$C_2 - C_3$	$C_3 - C_4$	$C_4 - C_5$	$C_5 - C_6$	$C_6 - N_7$
MP2	131.66	137.76	138.24	138.24	137.76	131.66
PBE0	131.78	137.91	138.57	138.57	137.91	131.78
PBE	133.00	138.80	139.43	139.43	138.80	133.00
HF	130.92	137.45	138.44	138.44	137.45	130.92

cn9	$N_1 - C_2$	$C_2 - C_3$	$C_3 - C_4$	$C_4 - C_5$	$C_5 - C_6$	$C_6 - C_7$	$C_7 - C_8$	$C_8 - N_9$
MP2	132.11	137.50	138.60	138.19	138.19	138.60	137.50	132.11
PBE0	132.20	137.58	138.98	138.38	138.38	138.98	137.58	132.20
PBE	133.42	138.53	139.78	139.30	139.30	139.78	138.53	133.42
HF	131.49	136.86	139.13	138.00	138.00	139.13	136.86	131.49

cn11	$N_1 - C_2$	$C_2 - C_3$	$C_3 - C_4$	$C_4 - C_5$	$C_5 - C_6$	$C_6 - C_7$	$C_7 - C_8$	$C_8 - C_9$	$C_9 - C_{10}$	$C_{10} - N_{11}$
MP2	132.48	137.25	138.90	137.98	138.48	138.48	137.98	138.90	137.25	132.48
PBE0	132.54	137.31	139.32	138.08	138.77	138.77	138.08	139.32	137.31	132.54
PBE	133.73	138.32	140.05	139.08	139.61	139.61	139.08	140.05	138.32	133.73
HF	131.97	136.34	139.79	137.34	138.69	138.69	137.34	139.79	136.34	131.97

TABLE XXIV. Ground-state bond lengths (pm) computed with the HF, MP2, DFT/PBE and PBE0 approaches. The cc-pVQZ basis is employed and the optimization is restricted to C_{2v} symmetry.

cn3

	$N_1 - C_2$	$C_2 - N_3$
D	131.56	131.56
T	130.41	130.41
Q	130.29	130.29

cn5

	$N_1 - C_2$	$C_2 - C_3$	$C_3 - C_4$	$C_4 - N_5$
D	132.28	139.62	139.62	132.28
T	131.16	137.85	137.85	131.16
Q	131.10	137.85	137.85	131.10

cn7

	$N_1 - C_2$	$C_2 - C_3$	$C_3 - C_4$	$C_4 - C_5$	$C_5 - C_6$	$C_6 - N_7$
D	132.80	139.59	139.93	139.93	139.59	132.80
T	131.70	137.78	138.20	138.20	137.78	131.70
Q	131.66	137.76	138.24	138.24	137.76	131.66

cn9

	$N_1 - C_2$	$C_2 - C_3$	$C_3 - C_4$	$C_4 - C_5$	$C_5 - C_6$	$C_6 - C_7$	$C_7 - C_8$	$C_8 - N_9$
D	133.21	139.35	140.23	139.91	139.91	140.23	139.35	133.21
T	132.14	137.52	138.54	138.15	138.15	138.54	137.52	132.14
Q	132.11	137.50	138.60	138.19	138.19	138.60	137.50	132.11

cn11

	$N_1 - C_2$	$C_2 - C_3$	$C_3 - C_4$	$C_4 - C_5$	$C_5 - C_6$	$C_6 - C_7$	$C_7 - C_8$	$C_8 - C_9$	$C_9 - C_{10}$	$C_{10} - N_{11}$
D	133.55	139.12	140.52	139.73	140.17	140.17	139.73	140.52	139.12	133.55
T	132.49	137.28	138.84	137.95	138.44	138.44	137.95	138.84	137.28	132.49
Q	132.48	137.25	138.90	137.98	138.48	138.48	137.98	138.90	137.25	132.48

TABLE XXV. Basis-set dependence of the ground-state RI-MP2 structures computed with the cc-pVXZ basis sets. The optimization was restricted to C_{2v}-symmetry. Bond lengths are given in pm.

cn3	$N_1 - C_2$	$C_2 - N_3$
D	131.90	131.90
T	130.46	130.46
Q	130.31	130.31

cn5	$N_1 - C_2$	$C_2 - C_3$	$C_3 - C_4$	$C_4 - N_5$
D	132.71	139.67	139.67	132.71
T	131.23	137.88	137.88	131.23
Q	131.15	137.84	137.84	131.15

cn7	$N_1 - C_2$	$C_2 - C_3$	$C_3 - C_4$	$C_4 - C_5$	$C_5 - C_6$	$C_6 - N_7$
D	133.27	139.60	140.07	140.07	139.60	133.27
T	131.81	137.77	138.26	138.26	137.77	131.81
Q	131.72	137.75	138.24	138.24	137.75	131.72

cn9	$N_1 - C_2$	$C_2 - C_3$	$C_3 - C_4$	$C_4 - C_5$	$C_5 - C_6$	$C_6 - C_7$	$C_7 - C_8$	$C_8 - N_9$
D	133.72	139.35	140.40	140.03	140.03	140.40	139.35	133.72
T	132.27	137.51	138.61	138.20	138.20	138.61	137.51	132.27
Q	132.18	137.48	138.60	138.19	138.19	138.60	137.48	132.18

cn11	$N_1 - C_2$	$C_2 - C_3$	$C_3 - C_4$	$C_4 - C_5$	$C_5 - C_6$	$C_6 - C_7$	$C_7 - C_8$	$C_8 - C_9$	$C_9 - C_{10}$	$C_{10} - N_{11}$
D	134.10	139.12	140.70	139.85	140.31	140.31	139.85	140.70	139.12	134.10
T	132.63	137.27	138.92	138.01	138.51	138.51	138.01	138.92	137.27	132.63
Q	132.55	137.23	138.91	137.98	138.48	138.48	137.98	138.91	137.23	132.55

TABLE XXVI. Basis-set dependence of the ground-state RI-MP2 structures computed with the aug-cc-pVXZ basis sets. The optimization was restricted to C_{2v}-symmetry. Bond lengths are given in pm.

cn3	$N_1 - C_2$	$C_2 - N_3$								
D	130.88	130.88
T	130.42	130.42
Q	130.34	130.34

cn5	$N_1 - C_2$	$C_2 - C_3$	$C_3 - C_4$	$C_4 - N_5$
D	131.69	138.85	138.85	131.69
T	131.26	138.15	138.15	131.26
Q	131.19	138.10	138.10	131.19

cn7	$N_1 - C_2$	$C_2 - C_3$	$C_3 - C_4$	$C_4 - C_5$	$C_5 - C_6$	$C_6 - N_7$
D	132.24	138.69	139.27	139.27	138.69	132.24
T	131.83	137.97	138.60	138.60	137.97	131.83
Q	131.78	137.91	138.57	138.57	137.91	131.78

cn9	$N_1 - C_2$	$C_2 - C_3$	$C_3 - C_4$	$C_4 - C_5$	$C_5 - C_6$	$C_6 - C_7$	$C_7 - C_8$	$C_8 - N_9$
D	132.65	138.40	139.65	139.12	139.12	139.65	138.40	132.65
T	132.27	137.65	139.01	138.43	138.43	139.01	137.65	132.27
Q	132.20	137.58	138.98	138.38	138.38	138.98	137.58	132.20

cn11	$N_1 - C_2$	$C_2 - C_3$	$C_3 - C_4$	$C_4 - C_5$	$C_5 - C_6$	$C_6 - C_7$	$C_7 - C_8$	$C_8 - C_9$	$C_9 - C_{10}$	$C_{10} - N_{11}$
D	132.95	138.15	139.97	138.83	139.47	139.47	138.83	139.97	138.15	132.95
T	132.60	137.38	139.35	138.14	138.80	138.80	138.14	139.35	137.38	132.60
Q	132.54	137.31	139.32	138.08	138.77	138.77	138.08	139.32	137.31	132.54

TABLE XXVII. Basis-set dependence of the ground-state PBE0 structure computed with the cc-pVXZ basis sets. The optimization was restricted to C_{2v}-symmetry. Bond lengths are given in pm.

cn3	$N_1 - C_2$	$C_2 - N_3$
D	131.01	131.01
T	130.43	130.43
Q	130.36	130.36

cn5	$N_1 - C_2$	$C_2 - C_3$	$C_3 - C_4$	$C_4 - N_5$
D	131.89	138.83	138.83	131.89
T	131.29	138.14	138.14	131.29
Q	131.22	138.11	138.11	131.22

cn7	$N_1 - C_2$	$C_2 - C_3$	$C_3 - C_4$	$C_4 - C_5$	$C_5 - C_6$	$C_6 - N_7$
D	132.47	138.65	139.31	139.31	138.65	132.47
T	131.88	137.94	138.63	138.63	137.94	131.88
Q	131.82	137.92	138.59	138.59	137.92	131.82

cn9	$N_1 - C_2$	$C_2 - C_3$	$C_3 - C_4$	$C_4 - C_5$	$C_5 - C_6$	$C_6 - C_7$	$C_7 - C_8$	$C_8 - N_9$
D	132.91	138.34	139.73	139.17	139.17	139.73	138.34	132.91
T	132.32	137.62	139.03	138.43	138.43	139.03	137.62	132.32
Q	132.26	137.59	139.02	138.40	138.40	139.02	137.59	132.26

cn11	$N_1 - C_2$	$C_2 - C_3$	$C_3 - C_4$	$C_4 - C_5$	$C_5 - C_6$	$C_6 - C_7$	$C_7 - C_8$	$C_8 - C_9$	$C_9 - C_{10}$	$C_{10} - N_{11}$
D	133.24	138.07	140.06	138.85	139.52	139.52	138.85	140.06	138.07	133.24
T	132.66	137.33	139.38	138.13	138.81	138.81	138.13	139.38	137.33	132.66
Q	132.59	137.31	139.35	138.10	138.80	138.80	138.10	139.35	137.31	132.59

TABLE XXVIII. Basis-set dependence of the ground-state PBE0 structure computed with the aug-cc-pVXZ basis sets. The optimization was restricted to C_{2v}-symmetry. Bond lengths are given in pm.

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