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

Assessing the accuracy of simplified Coupled Cluster methods for electronic excited states in f0 actinide compounds

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Supplementary Information

For ThO and ThS, we have used the all-electron atomic natural orbital relativistic correlation consistent (ANO-RCC) basis sets[1] available in the OpenMolcas program package version 17.0,[2] optimized specifically for the 2-nd order Douglas–Kroll–Hess (DKH2) Hamiltonian.[3–5] For all remaining molecules, we have employed the double-ζ correlation consistent basis sets of Peterson[6] for all heavy elements (cc-pVDZ-DK3), optimized specifically for the DKH3 Hamiltonian,[5,7,8] and Dunning's aug-cc-pVDZ basis set for all light elements.[9] Scalar relativistic effects were accounted for by the DKH2 Hamiltonian (ThO and ThS) and the DKH3 Hamiltonian (UO_2^{2+} and its isoelectronic series), respectively.

S1 UO_2^{2+}

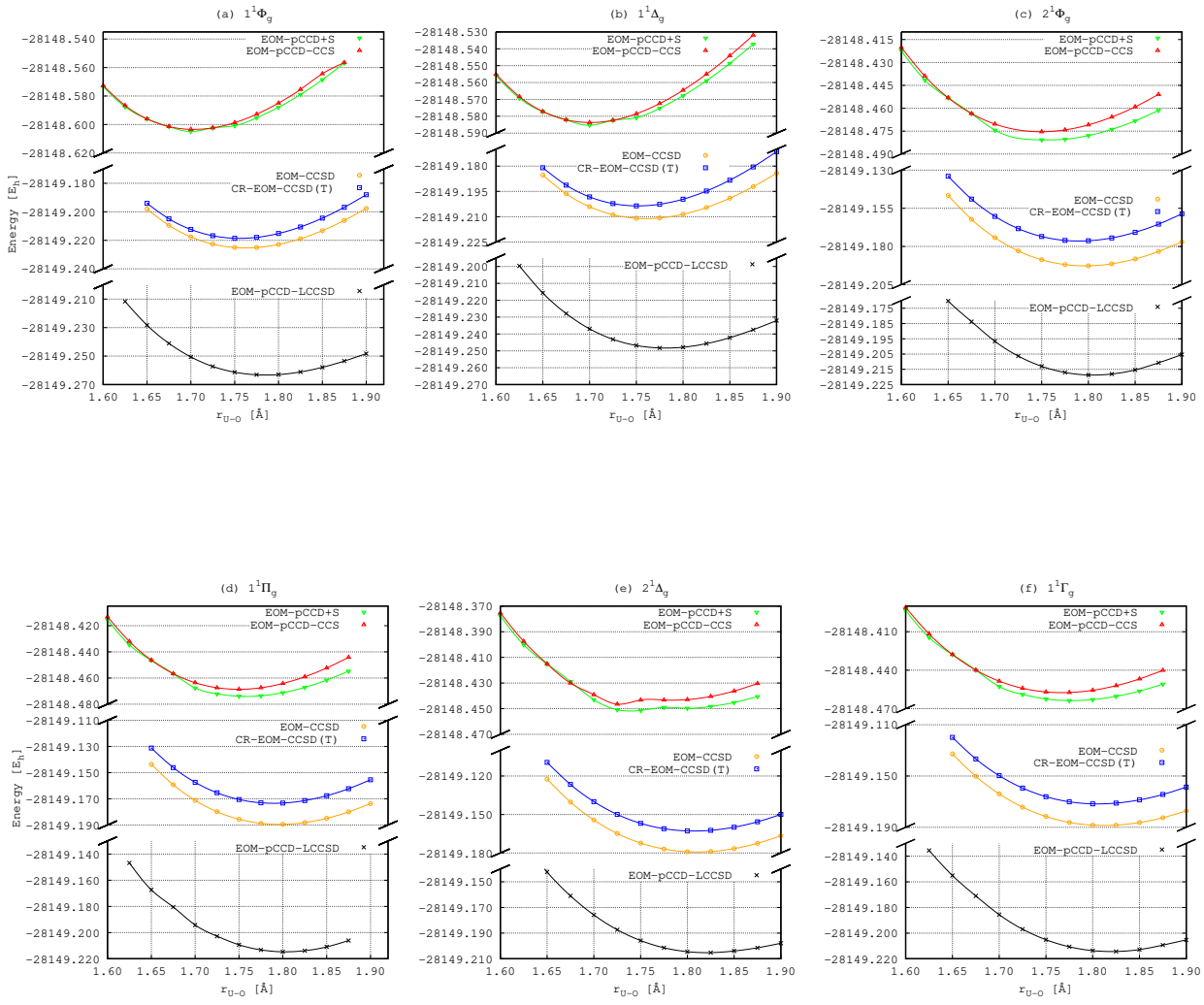

Figure S1: Potential energy surfaces of the lowest-lying excited states of UO_2^{2+} determined for various EOM-CC methods. In all pCCD-based methods, no symmetry constraints were imposed (C_1 symmetry), while in all conventional CC methods D_{2h} symmetry was imposed.

Table S1: Total electronic energies [E_h] of UO_2^{2+} for different states and various CC methods. In all pCCD-based methods, no symmetry constraints were imposed (C_1 symmetry), while in all conventional CC methods D_{2h} symmetry was imposed.

Distance[Å]	pCCD	pCCD-CCS	pCCD-LCCSD	CCSD	
			$X^1\Sigma^+$		
1.600	-28148.76552	-28148.76555			
1.625	-28148.77497	-28148.77500	-28149.37791		
1.650	-28148.77995	-28148.77999	-28149.38782	-28149.35231	
1.675	-28148.78106	-28148.78111	-28149.39399	-28149.35606	
1.700	-28148.77882	-28148.77888	-28149.39709	-28149.35641	
1.725	-28148.77371	-28148.77378	-28149.39755	-28149.35379	
1.750	-28148.76611	-28148.76621	-28149.39563	-28149.34858	
1.775	-28148.75639	-28148.75651	-28149.39175	-28149.34112	
1.800	-28148.74484	-28148.74499	-28149.38624	-28149.33170	
1.825	-28148.73173	-28148.73193	-28149.37930	-28149.32060	
1.850	-28148.71729	-28148.71754	-28149.37126	-28149.30804	
1.875	-28148.70172	-28148.70203	-28149.36231	-28149.29424	
1.900		-28148.68560	-28149.35333	-28149.27942	

Distance[Å]	EOM-pCCD+S	EOM-pCCD-CCS	EOM-pCCD-LCCSD	EOM-CCSD	CR-EOM-CCSD(T)
			$1^1\Phi_g$		
1.600	-28148.57269	-28148.57347			
1.625	-28148.58668	-28148.58759	-28149.21163		
1.650	-28148.59607	-28148.59611	-28149.22829	-28149.19797	-28149.19402
1.675	-28148.60147	-28148.60152	-28149.24107	-28149.20950	-28149.20492
1.700	-28148.60342	-28148.60492	-28149.25060	-28149.21760	-28149.21238
1.725	-28148.60237	-28148.60244	-28149.25726	-28149.22266	-28149.21680
1.750	-28148.59871	-28148.60082	-28149.26130	-28149.22503	-28149.21855
1.775	-28148.59280	-28148.59531	-28149.26311	-28149.22501	-28149.21791
1.800	-28148.58495	-28148.58792	-28149.26299	-28149.22288	-28149.21519
1.825	-28148.57541	-28148.57893	-28149.26114	-28149.21886	-28149.21061
1.850	-28148.56438	-28148.56853	-28149.25790	-28149.21318	-28149.20442
1.875	-28148.55657	-28148.55689	-28149.25346	-28149.20605	-28149.19682
1.900			-28149.24830	-28149.19764	-28149.18803
			$1^1\Delta_g$		
1.600	-28148.55511	-28148.55592			
1.625	-28148.56837	-28148.56934	-28149.19965		
1.650	-28148.57718	-28148.57722	-28149.21568	-28149.18534	-28149.18099
1.675	-28148.58212	-28148.58217	-28149.22791	-28149.19639	-28149.19131
1.700	-28148.58371	-28148.58535	-28149.23694	-28149.20409	-28149.19825
1.725	-28148.58239	-28148.58246	-28149.24317	-28149.20883	-28149.20224
1.750	-28148.57853	-28148.58086	-28149.24682	-28149.21095	-28149.20360
1.775	-28148.57249	-28148.57526	-28149.24828	-28149.21077	-28149.20264
1.800	-28148.56457	-28148.56786	-28149.24785	-28149.20855	-28149.19962
1.825	-28148.55504	-28148.55893	-28149.24571	-28149.20457	-28149.19479
1.850	-28148.54410	-28148.54868	-28149.24219	-28149.19904	-28149.18835
1.875	-28148.53192	-28148.53730	-28149.23748	-28149.19219	-28149.18051
1.900			-28149.23204	-28149.18424	-28149.17147
			$1^1\Gamma_g$		
1.600	-28148.39107	-28148.39344			
1.625	-28148.41170	-28148.41444	-28149.13551		
1.650	-28148.42780	-28148.42784	-28149.15510	-28149.13276	-28149.11986
1.675	-28148.43992	-28148.43997	-28149.17101	-28149.15023	-28149.13666
1.700	-28148.44856	-28148.45273	-28149.18560	-28149.16393	-28149.14970
1.725	-28148.45413	-28148.45420	-28149.19692	-28149.17426	-28149.15940
1.750	-28148.45697	-28148.46247	-28149.20515	-28149.18158	-28149.16609
1.775	-28148.45739	-28148.46372	-28149.21066	-28149.18617	-28149.17010
1.800	-28148.45569	-28148.46293	-28149.21367	-28149.18833	-28149.17169
1.825	-28148.45209	-28148.46039	-28149.21432	-28149.18829	-28149.17112
1.850	-28148.44683	-28148.45630	-28149.21285	-28149.18628	-28149.16862
1.875	-28148.44009	-28148.45088	-28149.20933	-28149.18252	-28149.16439
1.900			-28149.20532	-28149.17722	-28149.15863
			$2^1\Delta_g$		
1.600	-28148.37532	-28148.37767			
1.625	-28148.39748	-28148.40017	-28149.11951		
1.650	-28148.41511	-28148.41515	-28149.14239	-28149.12248	-28149.10936
1.675	-28148.43007	-28148.42885	-28149.16098	-28149.14030	-28149.12658
1.700	-28148.43911	-28148.44298	-28149.17586	-28149.15429	-28149.13998
1.725	-28148.44650	-28148.44657	-28149.18740	-28149.16483	-28149.14996
1.750	-28148.45134	-28148.45616	-28149.19581	-28149.17227	-28149.15689
1.775	-28148.44322	-28148.44908	-28149.20143	-28149.17692	-28149.16109
1.800	-28148.44291	-28148.44970	-28149.20451	-28149.17904	-28149.16282
1.825	-28148.44049	-28148.44831	-28149.20524	-28149.17886	-28149.16234
1.850	-28148.43624	-28148.44519	-28149.20394	-28149.17660	-28149.15989
1.875	-28148.43040	-28148.44058	-28149.20145	-28149.17245	-28149.15569
1.900			-28149.19791	-28149.16661	-28149.14995
			$2^1\Phi_g$		
1.600	-28148.42012	-28148.42237			
1.625	-28148.43889	-28148.44150	-28149.15261		
1.650	-28148.45316	-28148.45320	-28149.17019	-28149.14656	-28149.13402
1.675	-28148.46349	-28148.46354	-28149.18376	-28149.16223	-28149.14898
1.700	-28148.47037	-28148.47438	-28149.19664	-28149.17423	-28149.16029
1.725	-28148.47423	-28148.47431	-28149.20633	-28149.18294	-28149.16834
1.750	-28148.47540	-28148.48071	-28149.21302	-28149.18873	-28149.17349
1.775	-28148.47420	-28148.48031	-28149.21706	-28149.19188	-28149.17604
1.800	-28148.47091	-28148.47793	-28149.21870	-28149.19269	-28149.17628
1.825	-28148.46580	-28148.47385	-28149.21806	-28149.19138	-28149.17443
1.850	-28148.45907	-28148.46828	-28149.21538	-28149.18818	-28149.17074
1.875	-28148.45094	-28148.46144	-28149.21073	-28149.18331	-28149.16539
1.900			-28149.20535	-28149.17696	-28149.15860
			$1^1\Pi_g$		
1.600	-28148.41321	-28148.41547			
1.625	-28148.43202	-28148.43464	-28149.14676		
1.650	-28148.44632	-28148.44636	-28149.16733	-28149.14364	-28149.13121
1.675	-28148.45668	-28148.45672	-28149.18044	-28149.15927	-28149.14615
1.700	-28148.46358	-28148.46760	-28149.19423	-28149.17123	-28149.15742
1.725	-28148.46745	-28148.46752	-28149.20277	-28149.17990	-28149.16544
1.750	-28148.46862	-28148.47396	-28149.20933	-28149.18564	-28149.17055
1.775	-28148.46743	-28148.47357	-28149.21321	-28149.18876	-28149.17306
1.800	-28148.46415	-28148.47121	-28149.21468	-28149.18952	-28149.17326
1.825	-28148.45904	-28148.46713	-28149.21384	-28149.18817	-28149.17138
1.850	-28148.45232	-28148.46158	-28149.21094	-28149.18494	-28149.16765
1.875	-28148.44420	-28148.45475	-28149.20604	-28149.18004	-28149.16229
1.900			-28149.20029	-28149.17369	-28149.15548

Table S2: Excitation energies [eV] of UO_2^{2+} for different states and various CC methods. In all pCCD-based methods, no symmetry constraints were imposed (C_1 symmetry), while in all conventional CC methods D_{2h} symmetry was imposed.

Distance[Å]	EOM-pCCD+S	EOM-pCCD-CCS	EOM-pCCD-LCCSD	EOM-CCSD	CR-EOM-CCSD(T)
			$1^1\Phi_g$		
1.600	5.25	5.23			
1.625	5.12	5.10	4.52		
1.650	5.00	5.00	4.34	4.20	4.31
1.675	4.89	4.89	4.16	3.99	4.11
1.700	4.77	4.73	3.99	3.78	3.92
1.725	4.66	4.66	3.82	3.57	3.73
1.750	4.56	4.50	3.66	3.36	3.54
1.775	4.45	4.39	3.50	3.16	3.35
1.800	4.35	4.27	3.35	2.96	3.17
1.825	4.25	4.16	3.22	2.77	2.99
1.850	4.16	4.05	3.08	2.58	2.82
1.875	3.95	3.95	2.96	2.40	2.65
1.900			2.86	2.23	2.49
			$1^1\Delta_g$		
1.600	5.73	5.70			
1.625	5.62	5.60	4.85		
1.650	5.52	5.52	4.68	4.54	4.66
1.675	5.41	5.41	4.52	4.34	4.48
1.700	5.31	5.27	4.36	4.14	4.30
1.725	5.21	5.21	4.20	3.94	4.12
1.750	5.10	5.04	4.05	3.75	3.95
1.775	5.00	4.93	3.90	3.55	3.77
1.800	4.91	4.82	3.77	3.35	3.59
1.825	4.81	4.71	3.64	3.16	3.42
1.850	4.71	4.59	3.51	2.97	3.26
1.875	4.62	4.48	3.40	2.78	3.09
1.900			3.30	2.59	2.94
			$1^1\Gamma_g$		
1.600	10.19	10.13			
1.625	9.89	9.81	6.60		
1.650	9.58	9.58	6.33	5.97	6.33
1.675	9.28	9.28	6.07	5.60	5.97
1.700	8.99	8.88	5.75	5.24	5.62
1.725	8.70	8.70	5.46	4.89	5.29
1.750	8.41	8.27	5.18	4.54	4.97
1.775	8.14	7.97	4.93	4.22	4.65
1.800	7.87	7.68	4.70	3.90	4.35
1.825	7.61	7.39	4.49	3.60	4.07
1.850	7.36	7.11	4.31	3.31	3.79
1.875	7.12	6.83	4.16	3.04	3.53
1.900			4.03	2.78	3.29
			$2^1\Delta_g$		
1.600	10.62	10.55			
1.625	10.27	10.20	7.03		
1.650	9.93	9.93	6.68	6.25	6.61
1.675	9.55	9.59	6.34	5.87	6.24
1.700	9.24	9.14	6.02	5.50	5.89
1.725	8.90	8.90	5.72	5.14	5.55
1.750	8.57	8.44	5.44	4.80	5.22
1.775	8.52	8.37	5.18	4.47	4.90
1.800	8.22	8.04	4.95	4.15	4.60
1.825	7.93	7.72	4.74	3.86	4.31
1.850	7.65	7.41	4.55	3.58	4.03
1.875	7.38	7.11	4.38	3.31	3.77
1.900			4.23	3.07	3.52
			$2^1\Phi_g$		
1.600	9.40	9.34			
1.625	9.15	9.08	6.13		
1.650	8.89	8.89	5.92	5.60	5.94
1.675	8.64	8.64	5.72	5.27	5.63
1.700	8.39	8.29	5.45	4.96	5.34
1.725	8.15	8.15	5.20	4.65	5.05
1.750	7.91	7.77	4.97	4.35	4.76
1.775	7.68	7.52	4.75	4.06	4.49
1.800	7.45	7.27	4.56	3.78	4.23
1.825	7.24	7.02	4.39	3.52	3.98
1.850	7.03	6.78	4.24	3.26	3.74
1.875	6.82	6.55	4.12	3.02	3.51
1.900			4.03	2.79	3.29
			$1^1\Pi_g$		
1.600	9.59	9.53			
1.625	9.33	9.26	6.29		
1.650	9.08	9.08	6.00	5.68	6.02
1.675	8.83	8.83	5.81	5.35	5.71
1.700	8.58	8.47	5.52	5.04	5.41
1.725	8.33	8.33	5.30	4.73	5.13
1.750	8.10	7.95	5.07	4.43	4.84
1.775	7.86	7.70	4.86	4.15	4.57
1.800	7.64	7.45	4.67	3.87	4.31
1.825	7.42	7.21	4.50	3.60	4.06
1.850	7.21	6.97	4.36	3.35	3.82
1.875	7.01	6.73	4.25	3.11	3.59
1.900			4.16	2.88	3.37

S2 NUN

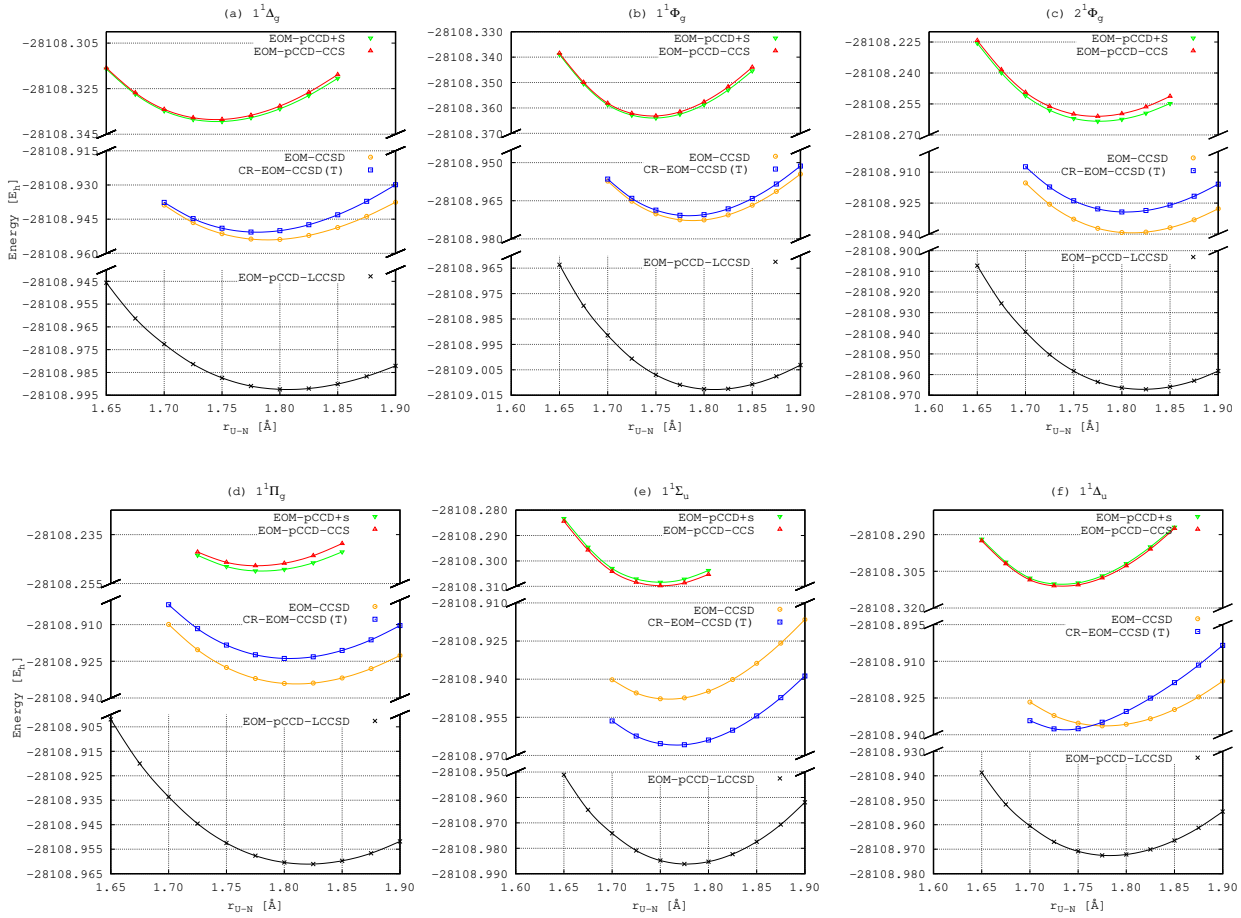

Figure S2: Potential energy surfaces of the lowest-lying excited states of NUN determined for various EOM-CC methods. In all pCCD-based methods, no symmetry constraints were imposed (C_1 symmetry), while in all conventional CC methods D_{2h} symmetry was imposed.

Table S3: Total electronic energies [E_h] of NUN for different states and various CC methods. In all pCCD-based methods, no symmetry constraints were imposed (C_1 symmetry), while in all conventional CC methods D_{2h} symmetry was imposed.

Distance[Å]	pCCD	pCCD-CCS	pCCD-LCCSD	CCSD	
			$X^1\Sigma^+$		
1.650	-28108.47864	-28108.47864	-28109.08992	-28109.05393	
1.675	-28108.48546	-28108.48546	-28109.10019	-28109.06409	
1.700	-28108.48865	-28108.48865	-28109.10594	-28109.06787	
1.725	-28108.48848	-28108.48849	-28109.10970	-28109.06964	
1.750	-28108.48541	-28108.48542	-28109.11075	-28109.06851	
1.775	-28108.47983	-28108.47984	-28109.10949	-28109.06483	
1.800	-28108.47208	-28108.47209	-28109.10622	-28109.05895	
1.825	-28108.46248	-28108.46248	-28109.10131	-28109.05113	
1.850	-28108.45131	-28108.45131	-28109.09496	-28109.04163	
1.875			-28109.08739	-28109.03068	
1.900			-28109.07869	-28109.01848	

Distance[Å]	EOM-pCCD+S	EOM-pCCD-CCS	EOM-pCCD-LCCSD	EOM-CCSD	CR-EOM-CCSD(T)
			$1^1\Delta_g$		
1.650	-28108.31639	-28108.31587	-28108.94563		
1.675	-28108.32738	-28108.32679	-28108.96126		
1.700	-28108.33474	-28108.33408	-28108.97256	-28108.93881	-28108.93763
1.725	-28108.33852	-28108.33775	-28108.98135	-28108.94652	-28108.94475
1.750	-28108.33941	-28108.33851	-28108.98737	-28108.95139	-28108.94898
1.775	-28108.33775	-28108.33670	-28108.99097	-28108.95374	-28108.95065
1.800	-28108.33386	-28108.33264	-28108.99241	-28108.95390	-28108.95005
1.825	-28108.32801	-28108.32660	-28108.99205	-28108.95213	-28108.94745
1.850	-28108.32046	-28108.31883	-28108.99007	-28108.94868	-28108.94307
1.875			-28108.98671	-28108.94376	-28108.93715
1.900			-28108.98211	-28108.93757	-28108.92990
			$1^1\Phi_g$		
1.650	-28108.33899	-28108.33849	-28108.96367		
1.675	-28108.35049	-28108.34993	-28108.97979		
1.700	-28108.35874	-28108.35812	-28108.99141	-28108.95727	-28108.95646
1.725	-28108.36284	-28108.36213	-28109.00060	-28108.96518	-28108.96404
1.750	-28108.36396	-28108.36315	-28109.00697	-28108.97014	-28108.96868
1.775	-28108.36248	-28108.36155	-28109.01088	-28108.97250	-28108.97073
1.800	-28108.35870	-28108.35763	-28109.01260	-28108.97256	-28108.97047
1.825	-28108.35292	-28108.35169	-28109.01247	-28108.97056	-28108.96818
1.850	-28108.34541	-28108.34399	-28109.01072	-28108.96675	-28108.96410
1.875			-28109.00755	-28108.96134	-28108.95842
1.900			-28109.00314	-28108.95450	-28108.95134
			$2^1\Phi_g$		
1.650	-28108.22557	-28108.22426	-28108.90726		
1.675	-28108.23990	-28108.23841	-28108.92554		
1.700	-28108.25100	-28108.24936	-28108.93926	-28108.91522	-28108.90738
1.725	-28108.25801	-28108.25616	-28108.95029	-28108.92554	-28108.91717
1.750	-28108.26202	-28108.25991	-28108.95826	-28108.93276	-28108.92386
1.775	-28108.26337	-28108.26098	-28108.96354	-28108.93722	-28108.92779
1.800	-28108.26242	-28108.25971	-28108.96638	-28108.93922	-28108.92927
1.825	-28108.25947	-28108.25640	-28108.96712	-28108.93904	-28108.92859
1.850	-28108.25476	-28108.25129	-28108.96593	-28108.93692	-28108.92597
1.875			-28108.96296	-28108.93308	-28108.92165
1.900			-28108.95827	-28108.92772	-28108.91581
			$1^1\Pi_g$		
1.650			-28108.90197		
1.675			-28108.92000		
1.700			-28108.93361	-28108.90993	-28108.90189
1.725	-28108.24344	-28108.24211	-28108.94456	-28108.92029	-28108.91168
1.750	-28108.24804	-28108.24632	-28108.95247	-28108.92754	-28108.91838
1.775	-28108.24984	-28108.24773	-28108.95766	-28108.93203	-28108.92233
1.800	-28108.24922	-28108.24671	-28108.96042	-28108.93406	-28108.92383
1.825	-28108.24654	-28108.24360	-28108.96106	-28108.93391	-28108.92316
1.850	-28108.24206	-28108.23865	-28108.95975	-28108.93182	-28108.92055
1.875			-28108.95665	-28108.92800	-28108.91623
1.900			-28108.95181	-28108.92265	-28108.91039
			$1^1\Sigma_u$		
1.650	-28108.28328	-28108.28436	-28108.95109		
1.675	-28108.29458	-28108.29572	-28108.96488		
1.700	-28108.30295	-28108.30415	-28108.97411	-28108.94022	-28108.95643
1.725	-28108.30710	-28108.30838	-28108.98090	-28108.94543	-28108.96237
1.750	-28108.30839	-28108.30976	-28108.98481	-28108.94769	-28108.96537
1.775	-28108.30717	-28108.30864	-28108.98620	-28108.94736	-28108.96579
1.800	-28108.30374	-28108.30531	-28108.98528	-28108.94475	-28108.96393
1.825			-28108.98231	-28108.94013	-28108.96008
1.850			-28108.97742	-28108.93376	-28108.95446
1.875			-28108.97064	-28108.92583	-28108.94728
1.900			-28108.96192	-28108.91656	-28108.93875
			$1^1\Delta_u$		
1.650	-28108.29186	-28108.29253	-28108.93871		
1.675	-28108.30123	-28108.30193	-28108.95170		
1.700	-28108.30778	-28108.30851	-28108.96047	-28108.92665	-28108.93427
1.725	-28108.31015	-28108.31092	-28108.96696	-28108.93216	-28108.93760
1.750	-28108.30973	-28108.31054	-28108.97086	-28108.93527	-28108.93755
1.775	-28108.30687	-28108.30771	-28108.97251	-28108.93638	-28108.93488
1.800	-28108.30187	-28108.30275	-28108.97216	-28108.93573	-28108.93050
1.825	-28108.29501	-28108.29592	-28108.97009	-28108.93346	-28108.92504
1.850	-28108.28695	-28108.28746	-28108.96642	-28108.92970	-28108.91872
1.875			-28108.96126	-28108.92455	-28108.91154
1.900			-28108.95462	-28108.91812	-28108.90349

Table S4: Excitation energies [eV] of NUN for different states and various CC methods. In all pCCD-based methods, no symmetry constraints were imposed (C_1 symmetry), while in all conventional CC methods D_{2h} symmetry was imposed.

Distance[Å]	EOM-pCCD+S	EOM-pCCD-CCS	EOM-pCCD-LCCSD	EOM-CCSD	CR-EOM-CCSD(T)
			$1^1\Delta_g$		
1.650	4.42	4.43	3.93		
1.675	4.30	4.32	3.78		
1.700	4.19	4.21	3.63	3.51	3.54
1.725	4.08	4.10	3.49	3.35	3.40
1.750	3.97	4.00	3.36	3.19	3.25
1.775	3.87	3.89	3.23	3.02	3.11
1.800	3.76	3.79	3.10	2.86	2.96
1.825	3.66	3.70	2.97	2.69	2.82
1.850	3.56	3.61	2.85	2.53	2.68
1.875			2.74	2.37	2.55
1.900			2.63	2.20	2.41
			$1^1\Phi_g$		
1.650	3.80	3.81	3.44		
1.675	3.67	3.69	3.28		
1.700	3.54	3.55	3.12	3.01	3.03
1.725	3.42	3.44	2.97	2.84	2.87
1.750	3.30	3.33	2.82	2.68	2.72
1.775	3.19	3.22	2.68	2.51	2.56
1.800	3.09	3.11	2.55	2.35	2.41
1.825	2.98	3.01	2.42	2.19	2.26
1.850	2.88	2.92	2.29	2.04	2.11
1.875			2.17	1.89	1.97
1.900			2.06	1.74	1.83
			$2^1\Phi_g$		
1.650	6.89	6.92	4.97		
1.675	6.68	6.72	4.75		
1.700	6.47	6.51	4.54	4.15	4.37
1.725	6.27	6.32	4.34	3.92	4.15
1.750	6.08	6.14	4.15	3.69	3.94
1.775	5.89	5.96	3.97	3.47	3.73
1.800	5.71	5.78	3.81	3.26	3.53
1.825	5.52	5.61	3.65	3.05	3.33
1.850	5.35	5.44	3.51	2.85	3.15
1.875			3.39	2.66	2.97
1.900			3.28	2.47	2.79
			$1^1\Pi_g$		
1.650			5.11		
1.675			4.90		
1.700			4.69	4.30	4.52
1.725	6.67	6.70	4.49	4.06	4.30
1.750	6.46	6.51	4.31	3.84	4.09
1.775	6.26	6.32	4.13	3.61	3.88
1.800	6.06	6.13	3.97	3.40	3.68
1.825	5.88	5.96	3.82	3.19	3.48
1.850	5.69	5.79	3.68	2.99	3.29
1.875			3.56	2.79	3.11
1.900			3.45	2.61	2.94
			$1^1\Sigma_u$		
1.650	5.32	5.29	3.78		
1.675	5.19	5.16	3.68		
1.700	5.05	5.02	3.59	3.47	3.03
1.725	4.94	4.90	3.50	3.38	2.92
1.750	4.82	4.78	3.43	3.29	2.81
1.775	4.70	4.66	3.36	3.20	2.70
1.800	4.58	4.54	3.29	3.11	2.59
1.825			3.24	3.02	2.48
1.850			3.20	2.94	2.37
1.875			3.18	2.85	2.27
1.900			3.18	2.77	2.17
			$1^1\Delta_u$		
1.650	5.08	5.06	4.11		
1.675	5.01	4.99	4.04		
1.700	4.92	4.90	3.96	3.84	3.64
1.725	4.85	4.83	3.88	3.74	3.59
1.750	4.78	4.76	3.81	3.63	3.56
1.775	4.71	4.68	3.73	3.50	3.54
1.800	4.63	4.61	3.65	3.35	3.50
1.825	4.56	4.53	3.57	3.20	3.43
1.850	4.47	4.46	3.50	3.05	3.34
1.875			3.43	2.89	3.24
1.900			3.38	2.73	3.13

S3 PaO_2^+

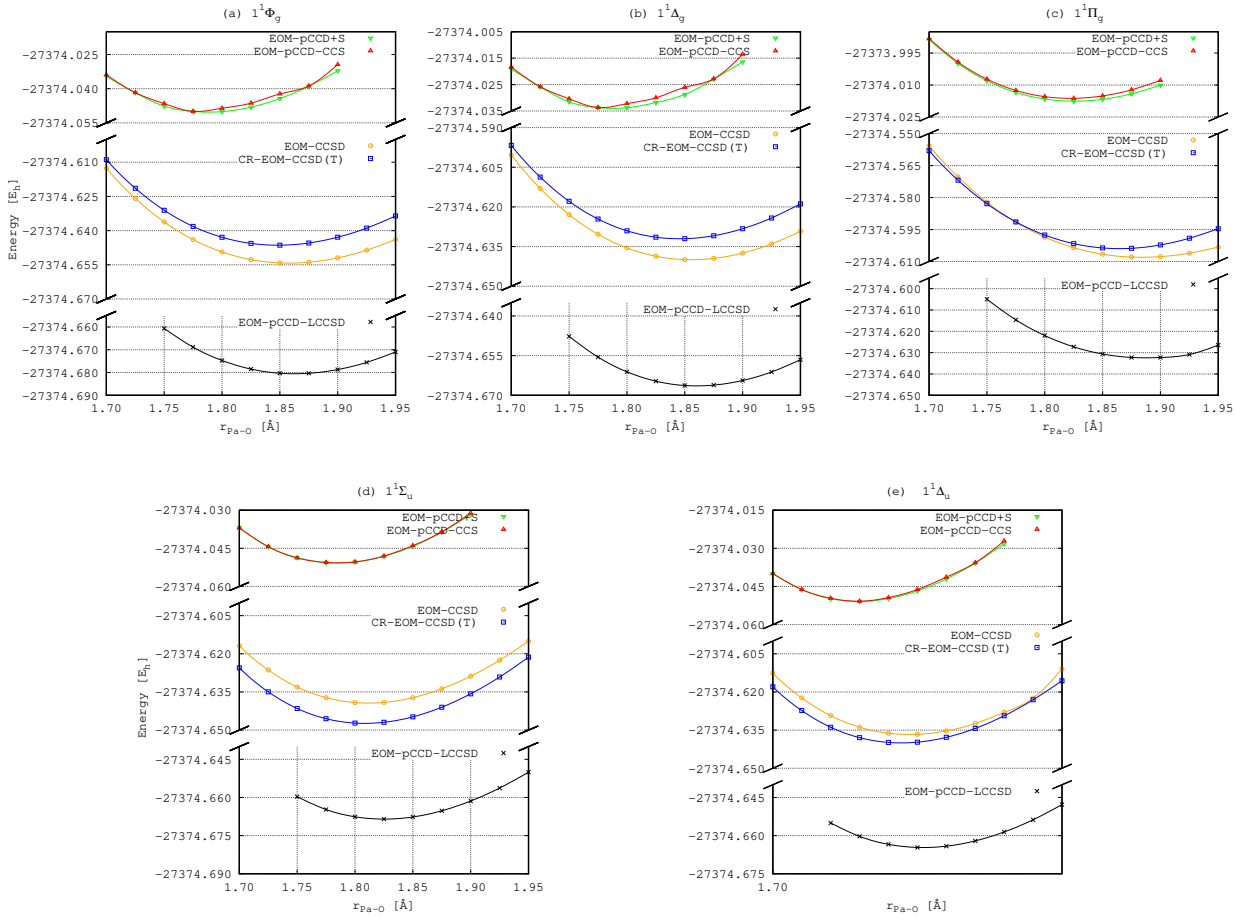

Figure S3: Potential energy surfaces of the lowest-lying excited states of PaO_2^+ determined for various EOM-CC methods. In all pCCD-based methods, no symmetry constraints were imposed (C_1 symmetry), while in all conventional CC methods D_{2h} symmetry was imposed.

Table S5: Total electronic energies [E_h] of PaO_2^+ for different states and various CC methods. In all pCCD-based methods, no symmetry constraints were imposed (C_1 symmetry), while in all conventional CC methods D_{2h} symmetry was imposed.

Distance[Å]	pCCD	pCCD-CCS	pCCD-LCCSD	CCSD
			$X^1\Sigma^+$	
1.650	-27374.24940	-27374.24938		
1.675	-27374.26221	-27374.26218	-27374.82507	-27374.79788
1.700	-27374.27120	-27374.27117	-27374.83615	-27374.80776
1.725	-27374.27684	-27374.27680	-27374.84410	-27374.81424
1.750	-27374.27951	-27374.27946	-27374.84899	-27374.81770
1.775	-27374.27954	-27374.27948	-27374.85139	-27374.81848
1.800	-27374.27724	-27374.27716	-27374.85152	-27374.81686
1.825	-27374.27286	-27374.27276	-27374.84966	-27374.81310
1.850	-27374.26665	-27374.26652	-27374.84603	-27374.80743
1.875	-27374.25879	-27374.25864	-27374.84078	-27374.80005
1.900	-27374.24947	-27374.24928	-27374.83413	-27374.79114
1.925	-27374.23883	-27374.23861	-27374.82626	-27374.78086
1.950	-27374.22704	-27374.22676	-27374.81732	-27374.76936
1.975	-27374.21422	-27374.21389		
2.000	-27374.20049	-27374.20009	-27374.79683	

Distance[Å]	EOM-pCCD+S	EOM-pCCD-CCS	EOM-pCCD-LCCSD	EOM-CCSD	CR-EOM-CCSD(T)
			$1^1\Phi_g$		
1.675				-27374.59629	-27374.59308
1.700	-27374.03469	-27374.03390		-27374.61268	-27374.60887
1.725	-27374.04178	-27374.04174		-27374.62586	-27374.62141
1.750	-27374.04764	-27374.04657	-27374.66066	-27374.63617	-27374.63107
1.775	-27374.05001	-27374.04995	-27374.66885	-27374.64393	-27374.63816
1.800	-27374.05006	-27374.04859	-27374.67472	-27374.64937	-27374.64293
1.825	-27374.04809	-27374.04636	-27374.67849	-27374.65272	-27374.64561
1.850	-27374.04432	-27374.04230	-27374.68032	-27374.65417	-27374.64638
1.875	-27374.03894	-27374.03879	-27374.68033	-27374.65387	-27374.64543
1.900	-27374.03215	-27374.02940	-27374.67870	-27374.65196	-27374.64289
1.925			-27374.67555	-27374.64858	-27374.63890
1.950			-27374.67097	-27374.64385	-27374.63359
			$1^1\Delta_g$		
1.675				-27374.58455	-27374.58163
1.700	-27374.01916	-27374.01836		-27374.60035	-27374.59673
1.725	-27374.02587	-27374.02583		-27374.61303	-27374.60870
1.750	-27374.03151	-27374.03039	-27374.64772	-27374.62293	-27374.61789
1.775	-27374.03374	-27374.03368	-27374.65556	-27374.63035	-27374.62458
1.800	-27374.03375	-27374.03221	-27374.66113	-27374.63552	-27374.62903
1.825	-27374.03182	-27374.03001	-27374.66467	-27374.63865	-27374.63145
1.850	-27374.02873	-27374.02605	-27374.66629	-27374.63991	-27374.63202
1.875	-27374.02296	-27374.02281	-27374.66615	-27374.63947	-27374.63090
1.900	-27374.01641	-27374.01352	-27374.66440	-27374.63746	-27374.62825
1.925			-27374.66117	-27374.63401	-27374.62419
1.950			-27374.65655	-27374.62923	-27374.61884
			$1^1\Sigma_u$		
1.675				-27374.60407	-27374.61282
1.700	-27374.03680	-27374.03704		-27374.61683	-27374.62552
1.725	-27374.04441	-27374.04437		-27374.62637	-27374.63497
1.750	-27374.04867	-27374.04881	-27374.65969	-27374.63307	-27374.64153
1.775	-27374.05064	-27374.05058	-27374.66475	-27374.63725	-27374.64553
1.800	-27374.05038	-27374.05037	-27374.66760	-27374.63919	-27374.64726
1.825	-27374.04816	-27374.04805	-27374.66851	-27374.63912	-27374.64694
1.850	-27374.04421	-27374.04397	-27374.66767	-27374.63725	-27374.64480
1.875	-27374.03867	-27374.03852	-27374.66523	-27374.63376	-27374.64101
1.900	-27374.03175	-27374.03116	-27374.66139	-27374.62881	-27374.63574
1.925			-27374.65629	-27374.62253	-27374.62913
1.950			-27374.65007	-27374.61506	-27374.62132
			$1^1\Delta_u$		
1.675				-27374.59985	-27374.60547
1.700	-27374.03998	-27374.03995		-27374.61265	-27374.61797
1.725	-27374.04624	-27374.04620		-27374.62235	-27374.62732
1.750	-27374.04982	-27374.04965	-27374.65502	-27374.62931	-27374.63388
1.775	-27374.05088	-27374.05082	-27374.66030	-27374.63385	-27374.63797
1.800	-27374.04977	-27374.04938	-27374.66344	-27374.63622	-27374.63984
1.825	-27374.04675	-27374.04622	-27374.66465	-27374.63665	-27374.63974
1.850	-27374.04205	-27374.04134	-27374.66417	-27374.63532	-27374.63784
1.875	-27374.03581	-27374.03566	-27374.66208	-27374.63241	-27374.63434
1.900	-27374.02824	-27374.02708	-27374.65856	-27374.62807	-27374.62937
1.925			-27374.65377	-27374.62242	-27374.62308
1.950			-27374.64781	-27374.61103	-27374.61559
			$1^1\Pi_g$		
1.675				-27374.53801	-27374.54113
1.700	-27373.98869	-27373.98823		-27374.55568	-27374.55802
1.725	-27373.99984	-27373.99925		-27374.57030	-27374.57185
1.750	-27374.00798	-27374.00724	-27374.60496	-27374.58216	-27374.58290
1.775	-27374.01346	-27374.01255	-27374.61462	-27374.59150	-27374.59141
1.800	-27374.01657	-27374.01546	-27374.62198	-27374.59855	-27374.59759
1.825	-27374.01756	-27374.01624	-27374.62727	-27374.60351	-27374.60162
1.850	-27374.01669	-27374.01511	-27374.63065	-27374.60658	-27374.60364
1.875	-27374.01412	-27374.01224	-27374.63225	-27374.60792	-27374.60381
1.900	-27374.01004	-27374.00784	-27374.63228	-27374.60771	-27374.60223
1.925			-27374.63088	-27374.60610	-27374.59909
1.950			-27374.62645	-27374.60323	-27374.59462

Table S6: Excitation energies [eV] of PaO_2^+ for different states and various CC methods. In all pCCD-based methods, no symmetry constraints were imposed (C_1 symmetry), while in all conventional CC methods D_{2h} symmetry was imposed.

Distance[Å]	EOM-pCCD+S	EOM-pCCD-CCS	EOM-pCCD-LCCSD	EOM-CCSD	CR-EOM-CCSD(T)
			$1^1\Phi_g$		
1.675				5.49	5.57
1.700	6.44	6.46		5.31	5.41
1.725	6.40	6.40		5.13	5.25
1.750	6.31	6.34	5.12	4.94	5.08
1.775	6.25	6.25	4.97	4.75	4.91
1.800	6.18	6.22	4.81	4.56	4.73
1.825	6.12	6.16	4.66	4.36	4.56
1.850	6.05	6.10	4.51	4.17	4.38
1.875	5.98	5.98	4.37	3.98	4.21
1.900	5.91	5.98	4.23	3.79	4.03
1.925			4.10	3.60	3.86
1.950			3.98	3.42	3.69
			$1^1\Delta_g$		
1.675				5.80	5.88
1.700	6.86	6.88		5.64	5.74
1.725	6.83	6.83		5.48	5.59
1.750	6.75	6.78	5.48	5.30	5.44
1.775	6.69	6.69	5.33	5.12	5.28
1.800	6.63	6.67	5.18	4.93	5.11
1.825	6.56	6.61	5.03	4.75	4.94
1.850	6.47	6.54	4.89	4.56	4.77
1.875	6.42	6.42	4.75	4.37	4.60
1.900	6.34	6.42	4.62	4.18	4.43
1.925			4.49	4.00	4.26
1.950			4.37	3.81	4.10
			$1^1\Sigma_u$		
1.675				5.27	5.04
1.700	6.38	6.37		5.20	4.96
1.725	6.32	6.32		5.11	4.88
1.750	6.28	6.28	5.15	5.02	4.79
1.775	6.23	6.23	5.08	4.93	4.71
1.800	6.17	6.17	5.00	4.83	4.62
1.825	6.11	6.11	4.93	4.73	4.52
1.850	6.05	6.06	4.85	4.63	4.43
1.875	5.99	5.99	4.78	4.52	4.33
1.900	5.92	5.94	4.70	4.42	4.23
1.925			4.63	4.31	4.13
1.950			4.55	4.20	4.03
			$1^1\Delta_u$		
1.675				5.39	5.24
1.700	6.29	6.29		5.31	5.16
1.725	6.28	6.28		5.22	5.09
1.750	6.25	6.25	5.28	5.13	5.00
1.775	6.22	6.22	5.20	5.02	4.91
1.800	6.19	6.20	5.12	4.92	4.82
1.825	6.15	6.16	5.03	4.80	4.72
1.850	6.11	6.13	4.95	4.68	4.61
1.875	6.07	6.07	4.86	4.56	4.51
1.900	6.02	6.05	4.78	4.44	4.40
1.925			4.69	4.31	4.29
1.950			4.61	4.31	4.18
			$1^1\Pi_g$		
1.675				7.07	6.99
1.700	7.69	7.70		6.86	6.80
1.725	7.54	7.55		6.64	6.60
1.750	7.39	7.41	6.64	6.41	6.39
1.775	7.24	7.26	6.44	6.18	6.18
1.800	7.09	7.12	6.25	5.94	5.97
1.825	6.95	6.98	6.05	5.70	5.75
1.850	6.80	6.84	5.86	5.47	5.55
1.875	6.66	6.70	5.67	5.23	5.34
1.900	6.52	6.57	5.49	4.99	5.14
1.925			5.32	4.76	4.95
1.950			5.19	4.52	4.75

S4 ThO_2

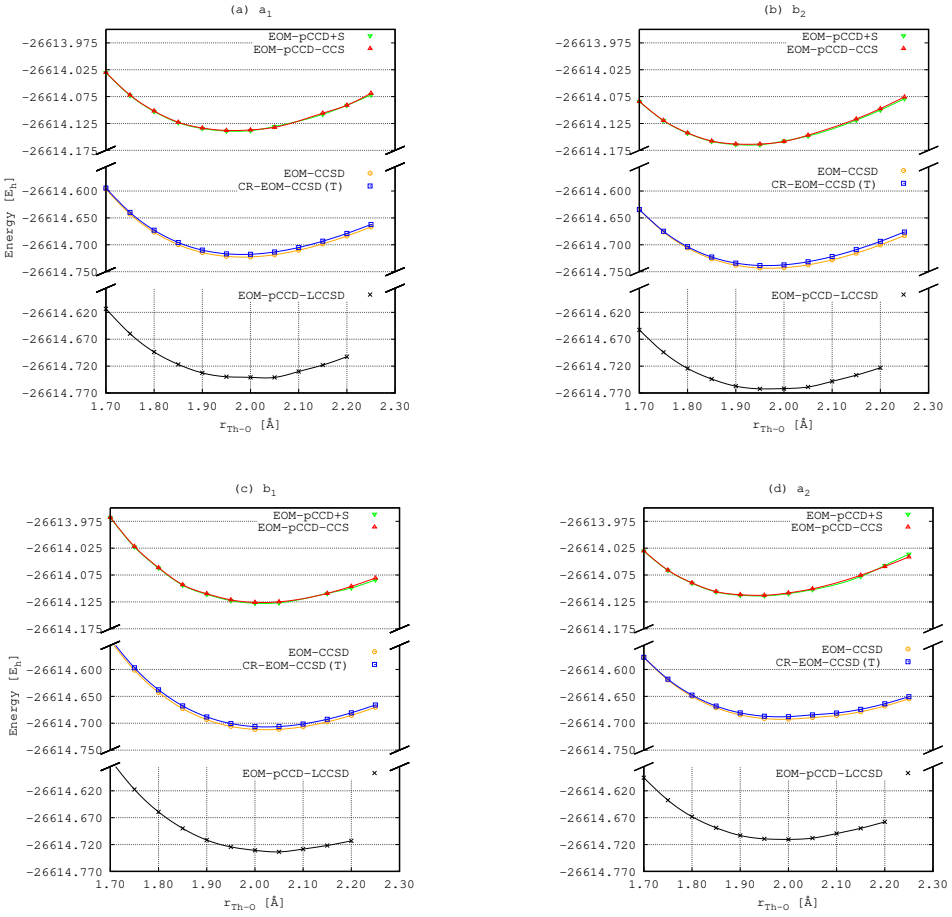

Figure S4: Potential energy surfaces of the lowest-lying excited states of ThO_2 determined for various EOM-CC methods. In all pCCD-based methods, no symmetry constraints were imposed (C_1 symmetry), while in all conventional CC methods D_{2h} symmetry was imposed.

Table S7: Total electronic energies [E_h] of ThO_2 for different states and various CC methods. In all pCCD-based methods, no symmetry constraints were imposed (C_1 symmetry), while in all conventional CC methods C_{2v} symmetry was imposed.

Distance[Å]	pCCD	pCCD-CCS	pCCD-LCCSD	CCSD	
			$X^1\Sigma^+$		
1.700	-26614.25295	-26614.25292	-26614.77198	-26614.74828	
1.750	-26614.29247	-26614.29244	-26614.81183	-26614.78697	
1.800	-26614.31837	-26614.31834	-26614.83911	-26614.81288	
1.850	-26614.33472	-26614.33469	-26614.85500	-26614.82817	
1.900	-26614.34039	-26614.34034	-26614.86430	-26614.83463	
1.950	-26614.33957	-26614.33951	-26614.86483	-26614.83378	
2.000	-26614.33274	-26614.33267	-26614.86013	-26614.82689	
2.050	-26614.32098	-26614.32090	-26614.85100	-26614.81505	
2.100			-26614.83818	-26614.79921	
2.150	-26614.28593	-26614.28581	-26614.82449	-26614.78017	
2.200	-26614.26433	-26614.26418	-26614.81099	-26614.75860	
2.250	-26614.24085	-26614.24066	-26614.79601	-26614.73509	

Distance[Å]	EOM-pCCD+S	EOM-pCCD-CCS	EOM-pCCD-LCCSD	EOM-CCSD	CR-EOM-CCSD(T)
			a_1		
1.700	-26614.03049	-26614.02994	-26614.61333	-26614.59677	-26614.59453
1.750	-26614.07277	-26614.07212	-26614.65957	-26614.64269	-26614.63972
1.800	-26614.10258	-26614.10181	-26614.69368	-26614.67638	-26614.67279
1.850	-26614.12351	-26614.12265	-26614.71679	-26614.69973	-26614.69564
1.900	-26614.13447	-26614.13344	-26614.73264	-26614.71433	-26614.70984
1.950	-26614.13917	-26614.13798	-26614.73978	-26614.72154	-26614.71676
2.000	-26614.13839	-26614.13703	-26614.74095	-26614.72256	-26614.71759
2.050	-26614.13067	-26614.13210	-26614.74105	-26614.71843	-26614.71336
2.100			-26614.73004	-26614.71006	-26614.70498
2.150	-26614.10801	-26614.10582	-26614.71801	-26614.69823	-26614.69323
2.200	-26614.09120	-26614.09105	-26614.70249	-26614.68361	-26614.67878
2.250	-26614.07187	-26614.06876		-26614.66675	-26614.66220
			b_2		
1.700	-26614.08433	-26614.08396	-26614.65264	-26614.63485	-26614.63453
1.750	-26614.11983	-26614.11935	-26614.69417	-26614.67610	-26614.67476
1.800	-26614.14319	-26614.14259	-26614.72431	-26614.70585	-26614.70354
1.850	-26614.15834	-26614.15762	-26614.74406	-26614.72597	-26614.72277
1.900	-26614.16430	-26614.16321	-26614.75733	-26614.73798	-26614.73398
1.950	-26614.16463	-26614.16332	-26614.76228	-26614.74317	-26614.73846
2.000	-26614.15780	-26614.15859	-26614.76174	-26614.74264	-26614.73731
2.050	-26614.14841	-26614.14682	-26614.75886	-26614.73732	-26614.73148
2.100			-26614.74838	-26614.72803	-26614.72180
2.150	-26614.11888	-26614.11661	-26614.73682	-26614.71548	-26614.70896
2.200	-26614.10006	-26614.09733	-26614.72314	-26614.70025	-26614.69358
2.250	-26614.07895	-26614.07565		-26614.68285	-26614.67617
			b_1		
1.700	-26613.96823	-26613.96757	-26614.56241	-26614.54684	-26614.54280
1.750	-26614.02261	-26614.02185	-26614.61739	-26614.60128	-26614.59677
1.800	-26614.06215	-26614.06126	-26614.65926	-26614.64262	-26614.63779
1.850	-26614.09393	-26614.09300	-26614.68990	-26614.67276	-26614.66770
1.900	-26614.11097	-26614.10978	-26614.71162	-26614.69335	-26614.68815
1.950	-26614.12259	-26614.12122	-26614.72440	-26614.70584	-26614.70057
2.000	-26614.12749	-26614.12591	-26614.73075	-26614.71148	-26614.70621
2.050	-26614.12668	-26614.12484	-26614.73360	-26614.71137	-26614.70614
2.100			-26614.72843	-26614.70644	-26614.70131
2.150	-26614.10922	-26614.10910	-26614.72176	-26614.69753	-26614.69256
2.200	-26614.09907	-26614.09610	-26614.71303	-26614.68533	-26614.68058
2.250	-26614.08411	-26614.08060		-26614.67046	-26614.66601
			a_2		
1.700	-26614.03049	-26614.02994	-26614.59561	-26614.57791	-26614.57731
1.750	-26614.06635	-26614.06585	-26614.63742	-26614.61954	-26614.61798
1.800	-26614.09018	-26614.08955	-26614.66813	-26614.65002	-26614.64759
1.850	-26614.10652	-26614.10577	-26614.68880	-26614.67119	-26614.66800
1.900	-26614.11256	-26614.11161	-26614.70310	-26614.68453	-26614.68069
1.950	-26614.11373	-26614.11258	-26614.70934	-26614.69127	-26614.68691
2.000	-26614.10968	-26614.10830	-26614.71036	-26614.69248	-26614.68772
2.050	-26614.10224	-26614.10058	-26614.70800	-26614.68938	-26614.68438
2.100			-26614.69951	-26614.68573	-26614.68092
2.150	-26614.07771	-26614.07537	-26614.68971	-26614.67863	-26614.67405
2.200	-26614.05723	-26614.05914	-26614.67779	-26614.66802	-26614.66374
2.250	-26614.03615	-26614.04128		-26614.65455	-26614.65062

Table S8: Excitation energies [eV] of ThO_2 for different states and various CC methods. In all pCCD-based methods, no symmetry constraints were imposed (C_1 symmetry), while in all conventional CC methods D_{2h} symmetry was imposed.

Distance[Å]	EOM-pCCD+S	EOM-pCCD-CCS	EOM-pCCD-LCCSD	EOM-CCSD	CR-EOM-CCSD(T)
			a_1		
1.700	6.05	6.07	4.32	4.12	4.18
1.750	5.98	6.00	4.14	3.93	4.01
1.800	5.87	5.89	3.96	3.71	3.81
1.850	5.75	5.77	3.76	3.49	3.61
1.900	5.60	5.63	3.58	3.27	3.40
1.950	5.45	5.48	3.40	3.05	3.18
2.000	5.29	5.32	3.24	2.84	2.97
2.050	5.18	5.14	2.99	2.63	2.77
2.100			2.94	2.43	2.56
2.150	4.84	4.90	2.90	2.23	2.37
2.200	4.71	4.71	2.95	2.04	2.17
2.250	4.60	4.68		1.86	1.98
			b_2		
1.700	4.59	4.60	3.25	3.09	3.10
1.750	4.70	4.71	3.20	3.02	3.05
1.800	4.77	4.78	3.12	2.91	2.98
1.850	4.80	4.82	3.02	2.78	2.87
1.900	4.79	4.82	2.91	2.63	2.74
1.950	4.76	4.79	2.79	2.47	2.59
2.000	4.76	4.74	2.68	2.29	2.44
2.050	4.70	4.74	2.51	2.12	2.27
2.100		0.00	2.44	1.94	2.11
2.150	4.55	4.60	2.39	1.76	1.94
2.200	4.47	4.54		1.59	1.77
2.250	4.41	4.49		1.42	1.60
			b_1		
1.700	7.75	7.76	5.70	5.48	5.59
1.750	7.34	7.36	5.29	5.05	5.18
1.800	6.97	7.00	4.89	4.63	4.76
1.850	6.55	6.58	4.49	4.23	4.37
1.900	6.24	6.27	4.15	3.84	3.99
1.950	5.90	5.94	3.82	3.48	3.62
2.000	5.59	5.63	3.52	3.14	3.28
2.050	5.29	5.34	3.19	2.82	2.96
2.100		0.00	2.99	2.52	2.66
2.150	4.81	4.81	2.80	2.25	2.38
2.200	4.50	4.57	2.67	1.99	2.12
2.250	4.27	4.36		1.76	1.88
			a_2		
1.700	6.05	6.07	4.80	4.64	4.65
1.750	6.15	6.17	4.75	4.56	4.60
1.800	6.21	6.23	4.65	4.43	4.50
1.850	6.21	6.23	4.52	4.27	4.36
1.900	6.20	6.22	4.39	4.08	4.19
1.950	6.15	6.18	4.23	3.88	4.00
2.000	6.07	6.11	4.08	3.66	3.79
2.050	5.95	6.00	3.89	3.42	3.56
2.100		0.00	3.77	3.09	3.22
2.150	5.67	5.73	3.67	2.76	2.89
2.200	5.64	5.58	3.62	2.46	2.58
2.250	5.57	5.43		2.19	2.30

S5 ThO

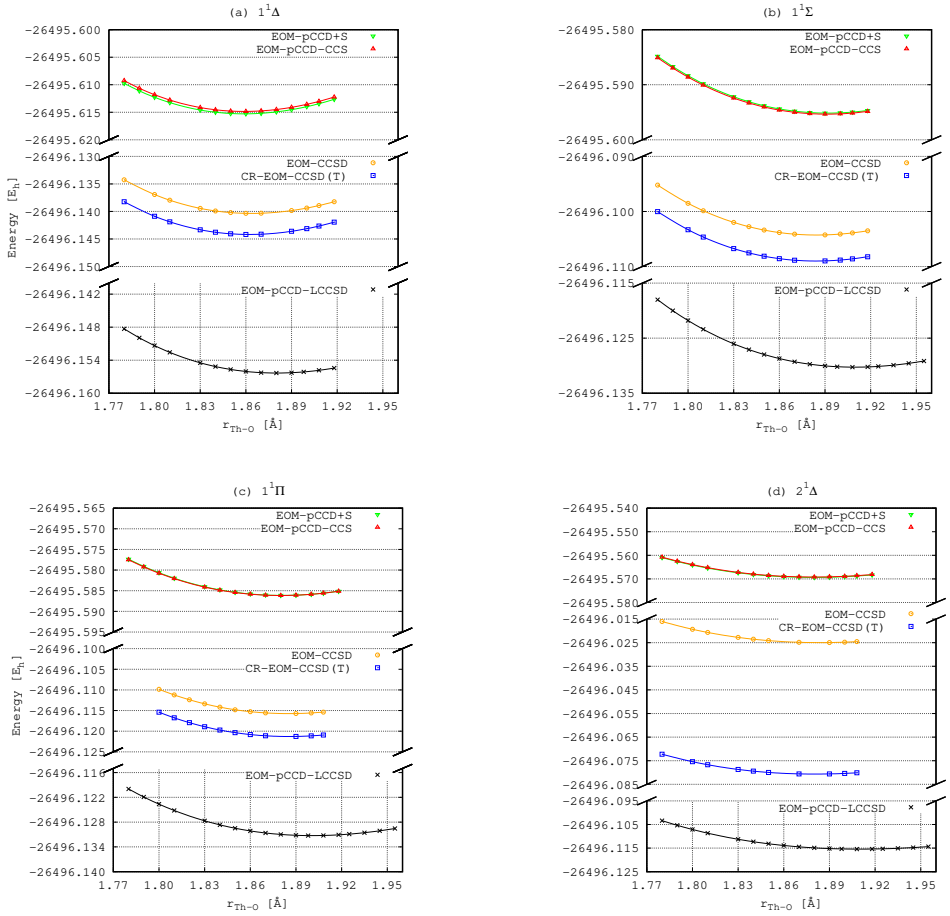

Figure S5: Potential energy surfaces of the lowest-lying excited states of ThO determined for various EOM-CC methods. In all pCCD-based methods, no symmetry constraints were imposed (C_1 symmetry), while in all conventional CC methods C_{2v} symmetry was imposed.

Table S9: Total electronic energies [E_h] of ThO for different states and various CC methods. In all pCCD-based methods, no symmetry constraints were imposed (C_1 symmetry), while in all conventional CC methods C_{2v} symmetry was imposed.

Distance[Å]	pCCD	pCCD-CCS	pCCD-LCCSD	CCSD	
			$X^1\Sigma^+$		
1.780	-26495.68519	-26495.68518	-26496.20682	-26496.18616	
1.790	-26495.68646	-26495.68645	-26496.20835		
1.800	-26495.68751	-26495.68750	-26496.20967	-26496.18854	
1.810	-26495.68836	-26495.68835	-26496.21079	-26496.18942	
1.820				-26496.19009	
1.830	-26495.68946	-26495.68945	-26496.21247	-26496.19058	
1.840	-26495.68973	-26495.68972	-26496.21304	-26496.19088	
1.850	-26495.68982	-26495.68981	-26496.21345	-26496.19101	
1.860	-26495.68973	-26495.68973	-26496.21370	-26496.19097	
1.870	-26495.68949	-26495.68948	-26496.21380	-26496.19077	
1.880	-26495.68908	-26495.68907	-26496.21376		
1.890	-26495.68852	-26495.68851	-26496.21358	-26496.18992	
1.900	-26495.68781	-26495.68780	-26496.21334	-26496.18928	
1.908	-26495.68714	-26495.68713	-26496.21294	-26496.18867	
1.918	-26495.68618	-26495.68617	-26496.21241	-26496.18779	

Distance[Å]	EOM-pCCD+S	EOM-pCCD-CCS	EOM-pCCD-LCCSD	EOM-CCSD	CR-EOM-CCSD(T)
			$1^1\Delta$		
1.780	-26495.60974	-26495.60926	-26496.14831	-26496.13425	-26496.13824
1.790	-26495.61112	-26495.61063	-26496.14993		
1.800	-26495.61228	-26495.61180	-26496.15135	-26496.13693	-26496.14087
1.810	-26495.61324	-26495.61277	-26496.15258	-26496.13795	-26496.14188
1.820					
1.830	-26495.61459	-26495.61413	-26496.15447	-26496.13944	-26496.14332
1.840	-26495.61499	-26495.61454	-26496.15516	-26496.13991	-26496.14378
1.850	-26495.61522	-26495.61477	-26496.15568	-26496.14021	-26496.14405
1.860	-26495.61528	-26495.61484	-26496.15604	-26496.14034	-26496.14417
1.870	-26495.61518	-26495.61475	-26496.15626	-26496.14032	-26496.14413
1.880	-26495.61493	-26495.61450	-26496.15633		
1.890	-26495.61453	-26495.61410	-26496.15627	-26496.13984	-26496.14360
1.900	-26495.61398	-26495.61356	-26496.15613	-26496.13938	-26496.14313
1.908	-26495.61344	-26495.61302	-26496.15584	-26496.13893	-26496.14265
1.918	-26495.61265	-26495.61223	-26496.15543	-26496.13824	-26496.14194
			$1^1\Sigma$		
1.780	-26495.58483	-26495.58509	-26496.11801	-26496.09521	-26496.10003
1.790	-26495.58670	-26495.58696	-26496.12001		
1.800	-26495.58836	-26495.58861	-26496.12179	-26496.09852	-26496.10331
1.810	-26495.58982	-26495.59007	-26496.12339	-26496.09987	-26496.10465
1.820					
1.830	-26495.59218	-26495.59241	-26496.12602	-26496.10198	-26496.10674
1.840	-26495.59309	-26495.59331	-26496.12707	-26496.10277	-26496.10752
1.850	-26495.59382	-26495.59404	-26496.12797	-26496.10338	-26496.10813
1.860	-26495.59439	-26495.59460	-26496.12870	-26496.10383	-26496.10857
1.870	-26495.59479	-26495.59500	-26496.12928	-26496.10412	-26496.10885
1.880	-26495.59504	-26495.59524	-26496.12973		
1.890	-26495.59515	-26495.59534	-26496.13003	-26496.10426	-26496.10898
1.900	-26495.59510	-26495.59528	-26496.13018	-26496.10411	-26496.10882
1.908	-26495.59497	-26495.59514	-26496.13026	-26496.10390	-26496.10861
1.918	-26495.59468	-26495.59484	-26496.13022	-26496.10352	-26496.10822
			$2^1\Delta$		
1.780	-26495.56094	-26495.56075	-26496.10338	-26496.01603	-26496.07224
1.790	-26495.56263	-26495.56245	-26496.10535		
1.800	-26495.56412	-26495.56394	-26496.10711	-26496.01932	-26496.07542
1.810	-26495.56541	-26495.56522	-26496.10868	-26496.02065	-26496.07670
1.820					
1.830	-26495.56740	-26495.56723	-26496.11126	-26496.02275	-26496.07869
1.840	-26495.56813	-26495.56795	-26496.11230	-26496.02352	-26496.07942
1.850	-26495.56868	-26495.56851	-26496.11317	-26496.02412	-26496.07997
1.860	-26495.56907	-26495.56890	-26496.11389		
1.870	-26495.56929	-26495.56912	-26496.11447	-26496.02483	-26496.08058
1.880	-26495.56936	-26495.56919	-26496.11490		
1.890	-26495.56928	-26495.56911	-26496.11520	-26496.02494	-26496.08059
1.900	-26495.56906	-26495.56889	-26496.11535	-26496.02477	-26496.08038
1.908	-26495.56878	-26495.56862	-26496.11543	-26496.02455	-26496.08012
1.918	-26495.56831	-26495.56815	-26496.11540		
			$1^1\Pi$		
1.780	-26495.57737	-26495.57750	-26496.11997		
1.790	-26495.57911	-26495.57923	-26496.12191		
1.800	-26495.58064	-26495.58075	-26496.12365	-26496.10985	-26496.11539
1.810	-26495.58197	-26495.58207	-26496.12518	-26496.11122	-26496.11676
1.820				-26496.11239	-26496.11793
1.830	-26495.58404	-26495.58412	-26496.12768	-26496.11337	-26496.11892
1.840	-26495.58479	-26495.58486	-26496.12866	-26496.11417	-26496.11972
1.850	-26495.58538	-26495.58543	-26496.12948	-26496.11480	-26496.12035
1.860	-26495.58579	-26495.58583	-26496.13013	-26496.11526	-26496.12081
1.870	-26495.58604	-26495.58607	-26496.13063	-26496.11557	-26496.12111
1.880	-26495.58613	-26495.58615	-26496.13098		
1.890	-26495.58607	-26495.58608	-26496.13119	-26496.11572	-26496.12127
1.900	-26495.58586	-26495.58586	-26496.13126	-26496.11558	-26496.12113
1.908	-26495.58560	-26495.58559	-26496.13123	-26496.11537	-26496.12092
1.918	-26495.58514	-26495.58512	-26496.13108	-26496.11500	-26496.12054

Table S10: Excitation energies [eV] of ThO for different states and various CC methods. In all pCCD-based methods, no symmetry constraints were imposed (C_1 symmetry), while in all conventional CC methods C_{2v} symmetry was imposed.

Distance[Å]	EOM-pCCD+S	EOM-pCCD-CCS	EOM-pCCD-LCCSD	EOM-CCSD	CR-EOM-CCSD(T)
			$1^1\Delta$		
1.780	2.05	2.07	1.59	1.41	1.30
1.790	2.05	2.06	1.59		
1.800	2.05	2.06	1.59	1.40	1.30
1.810	2.04	2.06	1.58	1.40	1.29
1.820					
1.830	2.04	2.05	1.58	1.39	1.29
1.840	2.03	2.05	1.58	1.39	1.28
1.850	2.03	2.04	1.57	1.38	1.28
1.860	2.03	2.04	1.57	1.38	1.27
1.870	2.02	2.03	1.57	1.37	1.27
1.880	2.02	2.03	1.56		
1.890	2.01	2.02	1.56	1.36	1.26
1.900	2.01	2.02	1.56	1.36	1.26
1.908	2.01	2.02	1.55	1.35	1.25
1.918	2.00	2.01	1.55	1.35	1.25
			$1^1\Sigma$		
1.780	2.73	2.72	2.42	2.47	2.34
1.790	2.71	2.71	2.40		
1.800	2.70	2.69	2.39	2.45	2.32
1.810	2.68	2.67	2.38	2.44	2.31
1.820					
1.830	2.65	2.64	2.35	2.41	2.28
1.840	2.63	2.62	2.34	2.40	2.27
1.850	2.61	2.61	2.33	2.38	2.26
1.860	2.59	2.59	2.31	2.37	2.24
1.870	2.58	2.57	2.30	2.36	2.23
1.880	2.56	2.55	2.29		
1.890	2.54	2.54	2.27	2.33	2.20
1.900	2.52	2.52	2.26	2.32	2.19
1.908	2.51	2.50	2.25	2.31	2.18
1.918	2.49	2.49	2.24	2.29	2.17
			$2^1\Delta$		
1.780	3.38	3.39	2.81	4.63	3.10
1.790	3.37	3.37	2.80		
1.800	3.36	3.36	2.79	4.60	3.08
1.810	3.35	3.35	2.78	4.59	3.07
1.820					
1.830	3.32	3.33	2.75	4.57	3.04
1.840	3.31	3.31	2.74	4.55	3.03
1.850	3.30	3.30	2.73	4.54	3.02
1.860	3.28	3.29	2.72		
1.870	3.27	3.28	2.70	4.52	3.00
1.880	3.26	3.26	2.69		
1.890	3.24	3.25	2.68	4.49	2.98
1.900	3.23	3.24	2.67	4.48	2.96
1.908	3.22	3.23	2.65	4.47	2.95
1.918	3.21	3.21	2.64		
			$1^1\Pi$		
1.780	2.93	2.93	2.36		
1.790	2.92	2.92	2.35		
1.800	2.91	2.90	2.34	2.14	1.99
1.810	2.90	2.89	2.33	2.13	1.98
1.820	0.00		0.00	2.11	
1.830	2.87	2.87	2.31	2.10	1.95
1.840	2.86	2.85	2.30	2.09	1.94
1.850	2.84	2.84	2.29	2.07	1.92
1.860	2.83	2.83	2.27	2.06	1.91
1.870	2.82	2.81	2.26	2.05	1.90
1.880	2.80	2.80	2.25		
1.890	2.79	2.79	2.24	2.02	1.87
1.900	2.77	2.77	2.23	2.01	1.85
1.908	2.76	2.76	2.22	1.99	1.84
1.918	2.75	2.75	2.21	1.98	1.83

S6 ThS

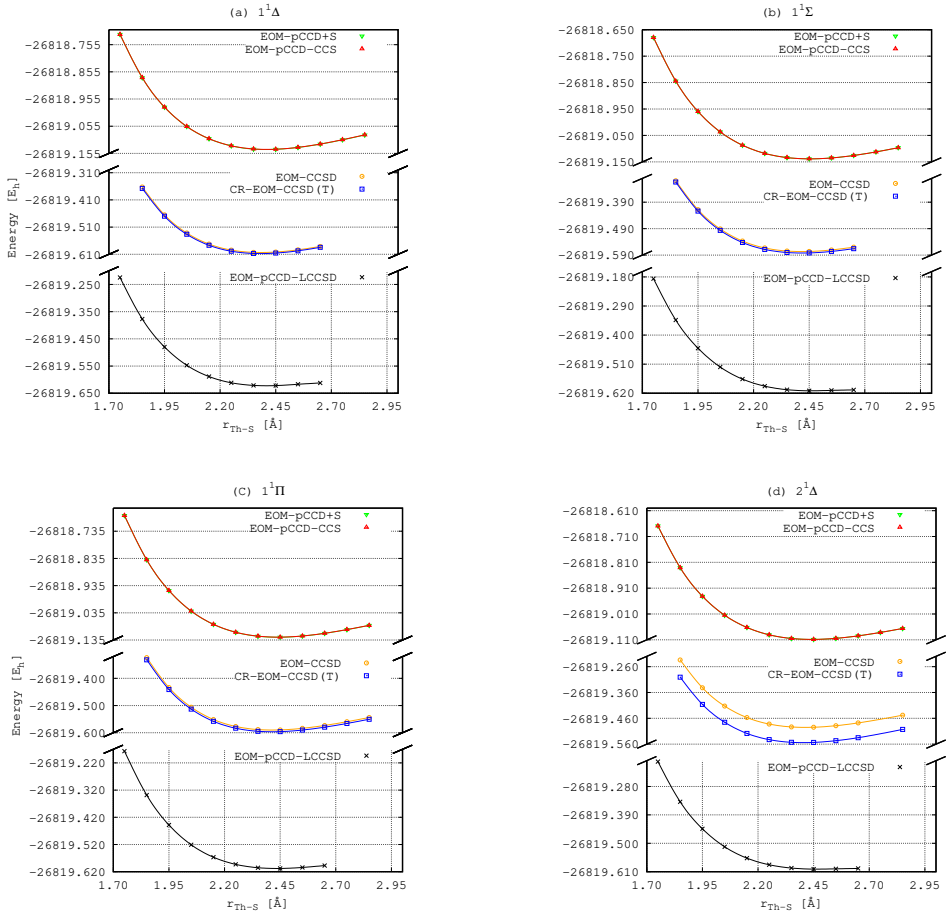

Figure S6: Potential energy surfaces of the lowest-lying excited states of ThS determined for various EOM-CC methods. In all pCCD-based methods, no symmetry constraints were imposed (C_1 symmetry), while in all conventional CC methods C_{2v} symmetry was imposed.

Table S11: Total electronic energies [E_h] of ThS for different states and various CC methods. In all pCCD-based methods, no symmetry constraints were imposed (C_1 symmetry), while in all conventional CC methods C_{2v} symmetry was imposed.

Distance[Å]	pCCD	pCCD-CCS	pCCD-LCCSD	CCSD
			$X^1\Sigma^+$	
1.75	-26818.79109	-26818.79109	-26819.28533	
1.85	-26818.95340	-26818.95340	-26819.44122	-26819.41836
1.95	-26819.06278	-26819.06278	-26819.54446	-26819.52127
2.05	-26819.13299	-26819.13299	-26819.61056	-26819.58672
2.15	-26819.17619	-26819.17619	-26819.65022	-26819.62540
2.25	-26819.20012	-26819.20012	-26819.67115	-26819.64495
2.35	-26819.20946	-26819.20945	-26819.67912	-26819.65089
2.45	-26819.20819	-26819.20818	-26819.67849	-26819.64726
2.55	-26819.19924	-26819.19924	-26819.67293	-26819.63707
2.65	-26819.18493	-26819.18492	-26819.66727	-26819.62253
2.75	-26819.16717	-26819.16716		-26819.60525
2.85	-26819.14756	-26819.14755		-26819.58645

Distance[Å]	EOM-pCCD+S	EOM-pCCD-CCS	EOM-pCCD-LCCSD	EOM-CCSD	CR-EOM-CCSD(T)
			$1^1\Delta$		
1.75	-26818.71747	-26818.71737	-26819.22449		
1.85	-26818.87673	-26818.87649	-26819.37707	-26819.36253	-26819.36774
1.95	-26818.98506	-26818.98484	-26819.47994	-26819.46501	-26819.47000
2.05	-26819.05612	-26819.05590	-26819.54720	-26819.53172	-26819.53648
2.15	-26819.10110	-26819.10088	-26819.58866	-26819.57251	-26819.57702
2.25	-26819.12722	-26819.12699	-26819.61154	-26819.59450	-26819.59877
2.35	-26819.13934	-26819.13859	-26819.62118	-26819.60292	-26819.60696
2.45	-26819.13990	-26819.13957	-26819.62183	-26819.60168	-26819.60549
2.55	-26819.13317	-26819.13278	-26819.61701	-26819.59370	-26819.59727
2.65	-26819.12094	-26819.12049	-26819.61218	-26819.58115	-26819.58451
2.75	-26819.10514	-26819.10458			
2.85	-26819.08736	-26819.08666		-26819.54849	-26819.55147
			$1^1\Sigma$		
1.75	-26818.67960	-26818.67952	-26819.18675		
1.85	-26818.84451	-26818.84447	-26819.34316	-26819.30889	-26819.31384
1.95	-26818.95947	-26818.95946	-26819.44960	-26819.41821	-26819.42355
2.05	-26819.03659	-26819.03657	-26819.52094	-26819.49099	-26819.49653
2.15	-26819.08697	-26819.08694	-26819.56651	-26819.53678	-26819.54239
2.25	-26819.11783	-26819.11779	-26819.59320	-26819.56284	-26819.56845
2.35	-26819.13374	-26819.13368	-26819.60656	-26819.57460	-26819.58020
2.45	-26819.13857	-26819.13849	-26819.61067	-26819.57613	-26819.58174
2.55	-26819.13515	-26819.13508	-26819.60917	-26819.57047	-26819.57611
2.65	-26819.12575	-26819.12571	-26819.60727	-26819.55991	-26819.56563
2.75	-26819.11228	-26819.11231			
2.85	-26819.09633	-26819.09646		-26819.53041	-26819.53656
			$2^1\Delta$		
1.75	-26818.66899	-26818.66887	-26819.18369		
1.85	-26818.83089	-26818.83070	-26819.33931	-26819.23398	-26819.30085
1.95	-26818.94150	-26818.94134	-26819.44410	-26819.34166	-26819.40609
2.05	-26819.01489	-26819.01473	-26819.51365	-26819.41266	-26819.47532
2.15	-26819.06218	-26819.06204	-26819.55761	-26819.45709	-26819.51845
2.25	-26819.09068	-26819.09054	-26819.58317	-26819.48222	-26819.54258
2.35	-26819.10485	-26819.10472	-26819.59592	-26819.49338	-26819.55291
2.45	-26819.10850	-26819.10836	-26819.60002	-26819.49452	-26819.55330
2.55	-26819.10440	-26819.10425	-26819.59898	-26819.48860	-26819.54662
2.65	-26819.09478	-26819.09463	-26819.59745	-26819.47781	-26819.53501
2.75	-26819.08155	-26819.08138			
2.85	-26819.06630	-26819.06608		-26819.44784	-26819.50292
			$1^1\Pi$		
1.75	-26818.67710	-26818.67686	-26819.17673		
1.85	-26818.84018	-26818.83997	-26819.33835	-26819.32377	-26819.33185
1.95	-26818.95328	-26818.95305	-26819.44840	-26819.43372	-26819.44113
2.05	-26819.02880	-26819.02855	-26819.52112	-26819.50611	-26819.51312
2.15	-26819.07746	-26819.07720	-26819.56683	-26819.55134	-26819.55808
2.25	-26819.10663	-26819.10636	-26819.59306	-26819.57691	-26819.58347
2.35	-26819.12110	-26819.12080	-26819.60542	-26819.58831	-26819.59474
2.45	-26819.12475	-26819.12444	-26819.60812	-26819.58959	-26819.59596
2.55	-26819.12046	-26819.12014	-26819.60442	-26819.58379	-26819.59014
2.65	-26819.11053	-26819.11022	-26819.59744	-26819.57319	-26819.57956
2.75	-26819.09693	-26819.09663		-26819.55947	-26819.56591
2.85	-26819.08133	-26819.08103		-26819.54393	-26819.55047

Table S12: Excitation energies [eV] of ThS for different states and various CC methods. In all pCCD-based methods, no symmetry constraints were imposed (C_1 symmetry), while in all conventional CC methods C_{2v} symmetry was imposed.

Distance[Å]	EOM-pCCD+S	EOM-pCCD-CCS	EOM-pCCD-LCCSD	EOM-CCSD	CR-EOM-CCSD(T)
			$1^1\Delta$		
1.75	2.00	2.01	1.66		
1.85	2.09	2.09	1.75	1.52	1.38
1.95	2.11	2.12	1.76	1.53	1.40
2.05	2.09	2.10	1.72	1.50	1.37
2.15	2.04	2.05	1.68	1.44	1.32
2.25	1.98	1.99	1.62	1.37	1.26
2.35	1.91	1.93	1.58	1.31	1.20
2.45	1.86	1.87	1.54	1.24	1.14
2.55	1.80	1.81	1.52	1.18	1.08
2.65	1.74	1.75	1.50	1.13	1.03
2.75	1.69	1.70			
2.85	1.64	1.66		1.03	0.95
			$1^1\Sigma$		
1.75	3.03	3.04	2.68		
1.85	2.96	2.96	2.67	2.98	2.84
1.95	2.81	2.81	2.58	2.80	2.66
2.05	2.62	2.62	2.44	2.60	2.45
2.15	2.43	2.43	2.28	2.41	2.26
2.25	2.24	2.24	2.12	2.23	2.08
2.35	2.06	2.06	1.97	2.08	1.92
2.45	1.89	1.90	1.85	1.94	1.78
2.55	1.74	1.75	1.74	1.81	1.66
2.65	1.61	1.61	1.63	1.70	1.55
2.75	1.49	1.49			
2.85	1.39	1.39		1.52	1.36
			$2^1\Delta$		
1.75	3.32	3.33	2.77		
1.85	3.33	3.34	2.77	5.02	3.20
1.95	3.30	3.30	2.73	4.89	3.13
2.05	3.21	3.22	2.64	4.74	3.03
2.15	3.10	3.11	2.52	4.58	2.91
2.25	2.98	2.98	2.39	4.43	2.79
2.35	2.85	2.85	2.26	4.29	2.67
2.45	2.71	2.72	2.14	4.16	2.56
2.55	2.58	2.58	2.01	4.04	2.46
2.65	2.45	2.46	1.90	3.94	2.38
2.75	2.33	2.33			
2.85	2.21	2.22		3.77	2.27
			$1^1\Pi$		
1.75	3.10	3.11	2.96		
1.85	3.08	3.09	2.80	2.57	2.35
1.95	2.98	2.99	2.61	2.38	2.18
2.05	2.84	2.84	2.43	2.19	2.00
2.15	2.69	2.69	2.27	2.02	1.83
2.25	2.54	2.55	2.12	1.85	1.67
2.35	2.40	2.41	2.01	1.70	1.53
2.45	2.27	2.28	1.91	1.57	1.40
2.55	2.14	2.15	1.86	1.45	1.28
2.65	2.02	2.03	1.90	1.34	1.17
2.75	1.91	1.92		1.25	1.07
2.85	1.80	1.81		1.16	0.98

S7 Error measures for total electronic energies and excitation energies

Table S13: Error measures [eV] for excitation energies along the potential energy surfaces of the lowest-lying excited states of ThO, ThS, UO_2^{2+}, NUN, PaO_2^{+}, and ThO_2 determined for various EOM-CC methods with respect to CR-EOM-CCSD(T).

		EOM-pCCD+S				EOM-pCCD-CCS				EOM-pCCD-LCCSD				EOM-CCSD			
	State	NPE	MAE	ME	RME	NPE	MAE	ME	RME	NPE	MAE	ME	RME	NPE	MAE	ME	RME
ThS	$1^1\Delta$	0.041	0.727	0.715	0.715	0.028	0.733	0.725	0.725	0.107	0.465	0.389	0.391	0.061	0.142	0.113	0.114
	$1^1\Pi$	0.150	0.876	0.838	0.839	0.152	0.884	0.846	0.847	0.300	0.731	0.501	0.510	0.047	0.220	0.202	0.184
	$1^1\Sigma$	0.133	0.169	0.120	0.128	0.137	0.170	0.120	0.128	0.160	0.176	0.067	0.081	0.033	0.167	0.152	0.152
	$2^1\Delta$	0.131	0.192	0.146	0.153	0.140	0.196	0.149	0.156	0.091	0.481	0.418	0.419	0.321	1.820	1.644	1.647
ThO	$1^1\Delta$	0.004	0.753	0.752	0.752	0.002	0.765	0.764	0.764	0.015	0.303	0.295	0.296	0.008	0.108	0.104	0.104
	$1^1\Pi$	0.003	0.920	0.919	0.919	0.007	0.920	0.917	0.917	0.037	0.383	0.365	0.365	0.000	0.151	0.150	0.150
	$1^1\Sigma$	0.062	0.387	0.354	0.355	0.060	0.380	0.348	0.349	0.003	0.074	0.072	0.072	0.003	0.131	0.129	0.129
	$2^1\Delta$	0.014	0.281	0.274	0.274	0.015	0.286	0.279	0.279	0.015	0.301	−0.293	0.293	0.017	1.530	1.520	1.520
UO_2^{2+}	$1^1\Phi_g$	0.406	1.341	1.046	1.068	0.602	1.299	1.002	1.021	0.337	0.371	0.169	0.200	0.154	0.262	−0.190	0.196
	$1^1\Delta_g$	0.670	1.526	1.195	1.214	0.532	1.388	1.133	1.146	0.341	0.363	0.158	0.191	0.229	0.348	−0.226	0.237
	$1^1\Gamma_g$	0.328	3.586	3.448	3.449	0.156	3.407	3.310	3.310	0.733	0.741	0.322	0.391	0.155	0.506	−0.434	0.436
	$2^1\Delta_g$	0.316	3.623	3.478	3.481	0.246	3.466	3.353	3.354	0.639	0.706	0.326	0.386	0.099	0.456	−0.421	0.422
	$2^1\Phi_g$	0.365	3.318	3.154	3.157	0.154	3.103	3.021	3.021	0.721	0.739	0.310	0.384	0.158	0.500	−0.427	0.430
	$1^1\Pi_g$	0.355	3.417	3.258	3.260	0.153	3.209	3.124	3.125	0.775	0.792	0.333	0.412	0.157	0.495	−0.423	0.426
NUN	$1^1\Delta_g$	0.235	0.879	0.760	0.764	0.261	0.923	0.790	0.795	0.132	0.217	0.141	0.148	0.177	0.209	−0.112	0.125
	$1^1\Phi_g$	0.269	0.772	0.635	0.641	0.290	0.811	0.661	0.668	0.144	0.229	0.148	0.155	0.064	0.086	−0.056	0.059
	$2^1\Phi_g$	1.901	2.201	2.156	2.156	0.152	2.296	2.223	2.224	0.314	0.483	0.297	0.314	0.111	0.324	−0.270	0.272
	$1^1\Pi_g$	0.030	2.399	2.384	2.384	0.086	2.492	2.448	2.448	0.338	0.511	0.312	0.331	0.115	0.334	−0.278	0.280
	$1^1\Sigma_u$	0.025	2.021	2.009	2.009	0.035	1.988	1.972	1.972	0.453	1.008	0.736	0.750	0.163	0.604	0.522	0.525
	$1^1\Delta_u$	0.160	1.286	1.189	1.191	0.165	1.267	1.168	1.384	0.183	0.323	0.215	0.223	0.358	0.398	−0.116	0.240
PaO_2^{+}	$1^1\Phi_g$	0.751	1.775	1.452	1.478	0.905	1.949	1.481	1.508	0.241	0.288	0.143	0.163	0.192	0.279	−0.184	0.194
	$1^1\Delta_g$	0.793	1.909	1.515	1.536	0.846	1.983	1.546	1.569	0.239	0.279	0.135	0.156	0.203	0.283	−0.184	0.195
	$1^1\Sigma_u$	0.276	1.696	1.557	1.560	0.295	1.707	1.557	1.560	0.166	0.523	0.433	0.196	0.068	0.238	0.211	0.213
	$1^1\Delta_u$	0.490	1.618	1.373	1.382	0.517	1.644	1.380	1.390	0.152	0.428	0.342	0.345	0.135	0.153	0.096	0.105
	$1^1\Pi_g$	0.483	1.375	1.129	1.140	0.526	1.429	1.159	1.172	0.188	0.439	0.323	0.327	0.232	0.234	−0.053	0.112
ThO_2	1^1A_1	0.745	2.615	2.261	2.273	0.810	2.694	2.285	2.297	0.647	0.781	0.287	0.347	0.078	0.138	−0.117	0.120
	1^1B_2	1.309	2.802	2.176	2.215	1.384	2.887	2.208	2.251	0.473	0.621	0.259	0.297	0.173	0.182	−0.121	0.133
	1^1B_1	0.268	2.424	2.278	2.280	0.302	2.476	2.316	2.318	0.432	0.543	0.236	0.270	0.033	0.144	−0.133	0.134
	1^1A_2	1.871	3.271	2.224	2.298	1.712	3.127	2.229	2.296	0.896	1.043	0.368	0.466	0.120	0.136	−0.133	0.105

Table S14: Error measures [eV] for adiabatic excitations energies of ThO and ThS determined for all excited states within a given method with respect to CR-EOM-CCSD(T).

	ThO				ThS			
Method	NPE	MAE	ME	RME	NPE	MAE	ME	RME
EOM-pCCD+S	0.640	0.920	0.575	0.636	0.770	0.840	0.435	0.552
EOM-pCCD-CCS	0.640	0.920	0.578	0.639	0.780	0.850	0.443	0.562
EOM-pCCD-LCCSD	0.300	0.340	0.088	0.276	0.460	0.470	0.100	0.380
EOM-CCSD	1.410	1.510	0.473	0.763	1.500	1.610	0.510	0.815

Table S15: Error measures [eV] for adiabatic excitations energies of UO_2^{2+}, NUN, PaO_2^{+}, and ThO_2 determined for all excited states within a given method with respect to CR-EOM-CCSD(T).

	UO_2^{2+}				NUN				PaO_2^{+}				ThO_2			
State	NPE	MAE	ME	RME	NPE	MAE	ME	RME	NPE	MAE	ME	RME	NPE	MAE	ME	RME
EOM-pCCD+S	2.640	3.680	2.688	2.914	1.830	2.540	1.628	1.781	0.320	1.620	1.484	1.489	0.160	2.320	2.238	2.239
EOM-pCCD-CCS	2.740	3.810	2.800	3.039	1.860	2.590	1.643	1.799	0.320	1.650	1.502	1.507	0.180	2.360	2.260	2.261
EOM-pCCD-LCCSD	0.090	0.100	−0.050	0.060	0.550	0.570	0.082	0.274	0.310	0.330	0.128	0.190	0.070	0.190	0.155	0.157
EOM-CCSD	0.270	0.460	−0.362	0.380	0.440	0.490	−0.030	0.264	0.220	0.310	−0.026	0.202	0.020	0.140	−0.133	0.133

Table S16: Error measures [eV] for total electronic energies along the potential energy surfaces for different states of $UO_2{}^{2+}$, NUN, $PaO_2{}^{+}$, ThO_2, ThO, and ThS determined for various EOM-CC methods with respect to CR-EOM-CCSD(T). pCCD/EOM-pCCD+S indicates that pCCD has been employed for the ground state calculation, while EOM-pCCD+S was used to target excited states.

		pCCD/EOM-pCCD+S				EOM-pCCD-CCS				EOM-pCCD-LCCSD				EOM-CCSD			
	State	NPE	MAE	ME	RME	NPE	MAE	ME	RME	NPE	MAE	ME	RME	NPE	MAE	ME	RME
$UO_2{}^{2+}$	$X\,^1\Sigma$	0.549	16.124	15.868	15.869	0.541	16.12	15.864	15.865	0.886	2.011	-1.420	1.458	–	–	–	–
	$1\,^1\Phi_g$	0.702	17.422	16.913	16.918	1.143	17.414	16.866	16.870	0.708	1.640	-1.251	1.271	0.154	0.262	−0.190	0.196
	$1\,^1\Delta_g$	1.219	17.649	17.062	17.067	1.074	17.503	16.997	17.000	0.704	1.648	-1.262	1.281	0.229	0.348	−0.226	0.237
	$1\,^1\Gamma_g$	0.877	19.709	19.315	19.317	0.585	19.416	19.174	19.175	0.336	1.271	-1.098	1.103	0.155	0.506	−0.434	0.436
	$2\,^1\Delta_g$	0.845	19.736	19.345	19.348	0.569	19.459	19.217	19.218	0.406	1.305	-1.094	1.101	0.099	0.456	−0.421	0.422
	$2\,^1\Phi_g$	0.914	19.441	19.022	19.024	0.630	19.156	18.885	18.886	0.326	1.272	-0.931	1.115	0.158	0.500	−0.427	0.430
	$1\,^1\Pi_g$	0.904	19.540	19.126	19.128	0.618	19.253	18.988	18.989	0.286	1.220	-0.908	1.090	0.157	0.495	−0.423	0.426
NUN	$X\,^1\Sigma$	0.409	0.590	0.583	0.583	0.409	0.590	0.583	0.583	0.659	1.639	-1.248	1.267	–	–	–	–
	$1\,^1\Delta_g$	0.537	16.942	16.676	16.677	0.563	16.987	16.706	16.707	0.470	1.421	-1.167	1.177	0.177	0.209	−0.112	0.125
	$1\,^1\Phi_g$	0.571	16.835	16.551	16.552	0.593	16.874	16.577	16.578	0.458	1.409	-1.161	1.170	0.064	0.086	−0.056	0.059
	$2\,^1\Phi_g$	0.403	18.265	18.072	18.073	0.454	18.359	18.139	18.140	0.288	1.156	-1.011	1.016	0.111	0.324	−0.270	0.272
	$1\,^1\Pi_g$	0.279	18.463	18.326	18.326	0.335	18.556	18.390	18.390	0.264	1.127	-0.996	1.000	0.115	0.334	−0.278	0.280
	$1\,^1\Sigma_u$	0.183	17.965	17.875	17.876	0.173	17.922	17.838	17.838	0.154	0.636	-0.572	0.574	0.163	0.604	0.522	0.525
	$1\,^1\Delta_u$	0.144	17.191	17.105	17.105	0.150	17.178	17.084	17.084	0.679	1.391	-1.094	1.118	0.358	0.398	−0.116	0.240
$PaO_2{}^{+}$	$X\,^1\Sigma$	0.181	14.740	14.68	14.678	0.188	15.915	15.604	15.571	0.454	1.305	-1.062	1.072	–	–	–	–
	$1\,^1\Phi_g$	0.879	16.503	16.131	16.134	1.048	16.694	16.161	16.165	0.212	1.017	-0.918	0.921	0.192	0.279	−0.184	0.194
	$1\,^1\Delta_g$	0.932	16.649	16.193	16.196	0.989	16.728	16.226	16.229	0.215	1.026	-0.927	0.929	0.203	0.283	−0.184	0.195
	$1\,^1\Sigma_u$	0.416	16.436	16.235	16.236	0.438	16.452	16.238	16.238	0.288	0.782	-0.629	0.636	0.068	0.238	0.211	0.213
	$1\,^1\Delta_u$	0.630	16.358	16.051	16.052	0.660	16.389	16.061	16.062	0.301	0.877	-0.720	0.727	0.135	0.153	0.096	0.105
	$1\,^1\Pi_g$	0.622	16.114	15.807	15.809	0.669	16.174	15.840	15.841	0.266	0.866	-0.739	0.745	0.232	0.234	−0.053	0.112
ThO_2	$X\,^1A_1$	0.051	13.479	13.450	13.450	0.051	13.479	13.453	13.453	1.013	1.658	-0.971	1.017	–	–	–	–
	$1A_1$	0.715	16.064	15.712	15.713	0.785	16.148	15.738	15.739	0.242	0.753	-0.621	0.625	0.078	0.138	−0.117	0.120
	$1B_2$	1.279	16.251	15.626	15.632	1.359	16.341	15.660	15.666	0.311	0.804	-0.650	0.657	0.173	0.182	−0.121	0.133
	$1B_1$	0.260	15.873	15.729	15.729	0.291	15.930	15.768	15.768	0.349	0.883	-0.673	0.680	0.033	0.144	−0.133	0.134
	$1A_2$	1.841	16.721	15.674	15.685	1.687	16.581	15.681	15.691	0.260	0.643	-0.540	0.546	0.120	0.136	−0.133	0.105
ThO	$X\,^1\Sigma^+$	0.018	13.650	13.640	13.640	0.017	13.650	13.640	13.640	0.108	0.670	-0.617	0.618	–	–	–	–
	$1\,^1\Delta$	0.022	14.403	14.392	14.392	0.020	14.414	14.404	14.404	0.357	0.367	-0.321	0.323	0.008	0.108	0.104	0.104
	$1\,^1\Pi$	0.020	14.569	14.559	14.559	0.024	14.570	14.557	14.557	0.279	0.287	-0.252	0.253	0.000	0.151	0.150	0.150
	$1\,^1\Sigma$	0.045	14.019	13.994	13.994	0.042	14.012	13.988	13.988	0.581	0.599	-0.545	0.546	0.003	0.131	0.129	0.129
	$2\,^1\Delta$	0.001	13.914	13.913	13.913	0.001	13.919	13.918	13.918	0.930	0.961	-0.904	0.905	0.017	1.530	1.520	1.520
ThS	$X\,^1\Sigma^+$	0.744	12.652	12.132	12.674	0.744	12.652	12.132	12.674	0.595	1.217	0.789	0.811	–	–	–	–
	$1\,^1\Delta$	0.747	13.361	12.868	12.870	0.741	13.368	12.878	12.880	0.499	0.753	-0.400	0.428	0.061	0.142	0.113	0.114
	$1\,^1\Pi$	0.617	13.379	12.970	12.972	0.615	13.385	12.978	12.979	0.309	0.486	-0.288	0.303	0.047	0.220	0.202	0.184
	$1\,^1\Sigma$	0.801	12.771	12.273	12.276	0.802	12.772	12.273	12.277	0.477	1.133	-0.782	0.795	0.033	0.167	0.152	0.152
	$2\,^1\Delta$	0.907	12.789	12.286	12.290	0.907	12.794	12.291	12.294	0.665	1.699	-1.207	1.225	0.321	1.820	1.644	1.647

S8 Results for TZ-quality basis sets

We have employed the triple-ζ correlation consistent basis sets of Peterson[6] for all heavy elements (cc-pVTZ-DK3), optimized specifically for the DKH3 Hamiltonian,[5,7,8] and Dunning's aug-cc-pVTZ basis set for all light elements.[9]

Table S17: Vertical excitation energies of the lowest-lying states of NUN determined for a basis set of double-ζ ($D\zeta$) and triple-ζ ($T\zeta$) quality.

	$D\zeta$	$T\zeta$	
Method	T_v [eV]	T_v[eV]	ΔT_v [eV]
		$1^1\Delta_g$	
EOM-pCCD+S	3.97	4.03	−0.06
EOM-pCCD-CCS	4.00	4.05	−0.05
EOM-pCCD-LCCSD	3.36	3.35	+0.01
		$1^1\Phi_g$	
EOM-pCCD+S	3.30	3.38	−0.08
EOM-pCCD-CCS	3.33	3.40	−0.07
EOM-pCCD-LCCSD	2.82	2.82	±0.00
		$2^1\Phi_g$	
EOM-pCCD+S	6.08	6.10	−0.02
EOM-pCCD-CCS	6.14	6.16	−0.02
EOM-pCCD-LCCSD	4.15	4.29	−0.14
		$1^1\Pi_g$	
EOM-pCCD+S	6.46	6.48	−0.02
EOM-pCCD-CCS	6.51	6.53	−0.02
EOM-pCCD-LCCSD	4.31	4.38	−0.07
		$1^1\Sigma_u$	
EOM-pCCD+S	4.82	4.85	−0.03
EOM-pCCD-CCS	4.78	4.81	−0.03
EOM-pCCD-LCCSD	3.43	3.56	−0.13
		$1^1\Delta_u$	
EOM-pCCD-CCS	4.76	4.79	−0.03
EOM-pCCD-LCCSD	3.81	3.85	−0.04

Table S18: Vertical excitation energies of the lowest-lying states of UO_2^{2+} determined for a basis set of double-ζ ($D\zeta$) and triple-ζ ($T\zeta$) quality.

	$D\zeta$	$T\zeta$	
Method	T_v [eV]	T_v[eV]	ΔT_v [eV]
		$1^1\Phi_g$	
EOM-pCCD+S	4.62	4.63	−0.01
EOM-pCCD-CCS	4.66	4.67	−0.01
EOM-pCCD-LCCSD	3.82	3.76	+0.06
EOM-CCSD	3.57	3.49	+0.08
CR-EOM-CCSD(T)	3.73	3.59	+0.14
		$1^1\Delta_g$	
EOM-pCCD+S	5.15	5.16	−0.01
EOM-pCCD-CCS	5.21	5.20	+0.01
EOM-pCCD-LCCSD	4.20	4.15	+0.05
EOM-CCSD	3.94	3.87	+0.07
CR-EOM-CCSD(T)	4.12	3.99	+0.13
		$1^1\Gamma_g$	
EOM-pCCD+S	8.57	8.59	−0.02
EOM-pCCD-CCS	8.70	8.72	−0.02
EOM-pCCD-LCCSD	5.46	5.43	+0.03
EOM-CCSD	4.89	4.86	+0.03
CR-EOM-CCSD(T)	5.29	5.17	+0.12
		$2^1\Delta_g$	
EOM-pCCD+S	8.79	8.82	−0.03
EOM-pCCD-CCS	8.90	8.94	−0.04
EOM-pCCD-LCCSD	5.72	5.67	+0.05
EOM-CCSD	5.14	5.11	+0.03
CR-EOM-CCSD(T)	5.55	5.41	+0.14
		$2^1\Phi_g$	
EOM-pCCD+S	8.03	8.06	−0.03
EOM-pCCD-CCS	8.15	8.18	−0.03
EOM-pCCD-LCCSD	5.20	5.19	+0.01
EOM-CCSD	4.65	4.63	+0.02
CR-EOM-CCSD(T)	5.05	4.94	+0.11
		$1^1\Pi_g$	
EOM-pCCD+S	8.21	8.25	−0.04
EOM-pCCD-CCS	8.33	8.37	−0.04
EOM-pCCD-LCCSD	5.30	5.31	+0.01
EOM-CCSD	4.73	4.72	+0.01
CR-EOM-CCSD(T)	5.13	5.02	+0.11

Table S19: Vertical excitation energies of the lowest-lying states of ThO_2 determined for a basis set of double-ζ ($D\zeta$) and triple-ζ ($T\zeta$) quality.

	$D\zeta$	$T\zeta$	
Method	T_v [eV]	T_v[eV]	ΔT_v [eV]
		a_1	
EOM-pCCD+S	5.45	5.50	−0.05
EOM-pCCD-CCS	5.48	5.53	−0.05
EOM-pCCD-LCCSD	3.40	3.55	−0.15
EOM-CCSD	3.05	3.28	−0.23
CR-EOM-CCSD(T)	3.18	3.36	−0.18
		b_2	
EOM-pCCD+S	4.76	4.79	−0.03
EOM-pCCD-CCS	4.79	4.83	−0.04
EOM-pCCD-LCCSD	2.79	2.94	−0.15
EOM-CCSD	2.47	2.74	−0.27
CR-EOM-CCSD(T)	2.59	2.74	−0.12
		b_1	
EOM-pCCD+S	5.90	5.90	±0.00
EOM-pCCD-CCS	5.94	5.93	+0.01
EOM-pCCD-LCCSD	3.82	3.96	−0.14
EOM-CCSD	3.48	3.96	−0.48
CR-EOM-CCSD(T)	3.62	3.99	−0.31
		a_2	
EOM-pCCD+S	6.15	6.15	±0.00
EOM-pCCD-CCS	6.18	6.18	±0.00
EOM-pCCD-LCCSD	4.23	4.31	−0.10
EOM-CCSD	3.88	4.14	−0.26
CR-EOM-CCSD(T)	4.00	4.13	−0.13

S9 Example results for all tested EOM-CC methods with imposed D_{2h} symmetry

S9.1 UO_2^{2+}

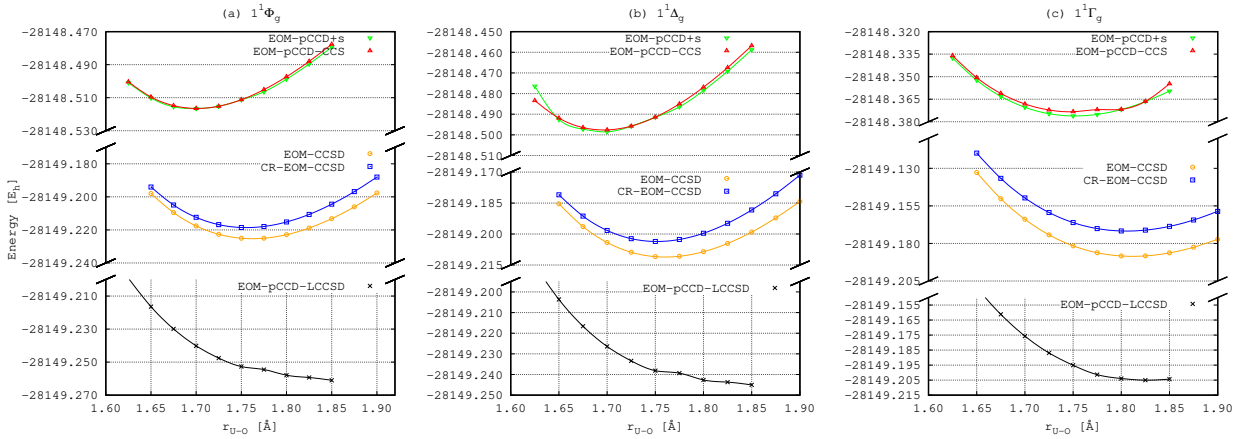

Figure S7: Potential energy surfaces of the lowest-lying excited states of UO_2^{2+} determined for various EOM-CC methods with imposed D_{2h} symmetry.

Table S20: Total electronic energies $[E_h]$ of UO_2^{2+} for different states and various CC methods. All EOM-CC methods are calculated with imposed D_{2h} symmetry.

Distance[Å]	pCCD	pCCD-CCS	pCCD-LCCSD	CCSD	
			$X^1\Sigma^+$		
1.600	-28148.67865	-28148.67861			
1.625	-28148.68626	-28148.68623	-28149.36515		
1.650	-28148.68938	-28148.68934	-28149.37486	-28149.35230585	
1.675	-28148.68862	-28148.68858	-28149.38108	-28149.35605907	
1.700	-28148.68454	-28148.68448	-28149.38437	-28149.35640687	
1.725	-28148.67758	-28148.67751	-28149.38519	-28149.35378603	
1.750	-28148.66819	-28148.66811	-28149.38399	-28149.34857922	
1.775	-28148.65668	-28148.65658	-28149.37962	-28149.34111895	
1.800	-28148.64333	-28148.64322	-28149.37726	-28149.33170396	
1.825	-28148.62840	-28148.62828	-28149.37293	-28149.32059737	
1.850	-28148.61215	-28148.61200	-28149.36891	-28149.3080368	
1.875				-28149.2942417	
1.900				-28149.2794163	

Distance[Å]	EOM-pCCD+S	EOM-pCCD-CCS	EOM-pCCD-LCCSD	EOM-CCSD	CR-EOM-CCSD(T)
			$1^1\Phi_g$		
1.600					
1.625	-28148.50097	-28148.50042	-28149.19921		
1.650	-28148.51019	-28148.50957	-28149.21639	-28149.19797	-28149.19402
1.675	-28148.51552	-28148.51480	-28149.22989	-28149.20950	-28149.20492
1.700	-28148.51657	-28148.51651	-28149.24014	-28149.21760	-28149.21238
1.725	-28148.51523	-28148.51516	-28149.24757	-28149.22266	-28149.21680
1.750	-28148.51132	-28148.51124	-28149.25268	-28149.22503	-28149.21855
1.775	-28148.50644	-28148.50514	-28149.25450	-28149.22501	-28149.21791
1.800	-28148.49876	-28148.49729	-28149.25791	-28149.22288	-28149.21519
1.825	-28148.48963	-28148.48801	-28149.25932	-28149.21886	-28149.21061
1.850	-28148.47940	-28148.47763	-28149.26099	-28149.21318	-28149.20442
1.875				-28149.20605	-28149.19682
1.900				-28149.19764	-28149.18803
			$1^1\Delta_g$		
1.600					
1.625	-28148.47640	-28148.48343	-28149.18705	-28149.18534	-28149.18099
1.650	-28148.49255	-28148.49188	-28149.20365	-28149.19639	-28149.19131
1.675	-28148.49724	-28148.49646	-28149.21663	-28149.20409	-28149.19825
1.700	-28148.49851	-28148.49759	-28149.22641	-28149.20883	-28149.20224
1.725	-28148.49582	-28148.49576	-28149.23340	-28149.21095	-28149.20360
1.750	-28148.49150	-28148.49142	-28149.23813	-28149.21077	-28149.20264
1.775	-28148.48645	-28148.48501	-28149.23933	-28149.20855	-28149.19962
1.800	-28148.47854	-28148.47692	-28149.24263	-28149.20457	-28149.19479
1.825	-28148.46922	-28148.46742	-28149.24370	-28149.19904	-28149.18835
1.850	-28148.45879	-28148.45682	-28149.24504	-28149.19219	-28149.18051
1.875				-28149.18424	-28149.17147
1.900					
			$1^1\Gamma_g$		
1.600					
1.625	-28148.33757	-28148.33603	-28149.12087		
1.650	-28148.35233	-28148.35061	-28149.14308	-28149.13276	-28149.11986
1.675	-28148.36310	-28148.36116	-28149.16118	-28149.15023	-28149.13666
1.700	-28148.37038	-28148.36819	-28149.17567	-28149.16393	-28149.14970
1.725	-28148.37467	-28148.37218	-28149.18688	-28149.17426	-28149.15940
1.750	-28148.37615	-28148.37392	-28149.19510	-28149.18158	-28149.16609
1.775	-28148.37513	-28148.37198	-28149.20134	-28149.18617	-28149.17010
1.800	-28148.37187	-28148.36860	-28149.20390	-28149.18833	-28149.17169
1.825	-28148.36659	-28148.36285	-28149.20503	-28149.18829	-28149.17112
1.850	-28148.35960	-28148.35474	-28149.20433	-28149.18628	-28149.16862
1.875				-28149.18252	-28149.16439
1.900				-28149.17722	-28149.15863
			$2^1\Delta_g$		
1.600					
1.625	-28148.32666	-28148.32526	-28149.11021		
1.650	-28148.34395	-28148.34244	-28149.13284	-28149.12248	-28149.10936
1.675	-28148.35743	-28148.35553	-28149.15131	-28149.14030	-28149.12658
1.700	-28148.36528	-28148.36313	-28149.16609	-28149.15429	-28149.13998
1.725	-28148.37009	-28148.36765	-28149.17756	-28149.16483	-28149.14996
1.750	-28148.37207	-28148.36932	-28149.18598	-28149.17227	-28149.15689
1.775	-28148.36628	-28148.36368	-28149.19296	-28149.17692	-28149.16109
1.800	-28148.36501	-28148.36213	-28149.19516	-28149.17904	-28149.16282
1.825	-28148.36152	-28148.35836	-28149.19643	-28149.17886	-28149.16234
1.850	-28148.35740	-28148.35239	-28149.19588	-28149.17660	-28149.15989
1.875				-28149.17245	-28149.15569
1.900				-28149.16661	-28149.14995
			$2^1\Phi_g$		
1.600					
1.625	-28148.36327	-28148.36180	-28149.13612		
1.650	-28148.37639	-28148.37474	-28149.15661	-28149.14656	-28149.13402
1.675	-28148.38559	-28148.38373	-28149.17309	-28149.16223	-28149.14898
1.700	-28148.39131	-28148.38921	-28149.18600	-28149.17423	-28149.16029
1.725	-28148.39405	-28148.39165	-28149.19569	-28149.18294	-28149.16834
1.750	-28148.39134	-28148.39126	-28149.20244	-28149.18873	-28149.17349
1.775	-28148.38844	-28148.38834	-28149.20610	-28149.19188	-28149.17604
1.800	-28148.38657	-28148.38318	-28149.20843	-28149.19269	-28149.17628
1.825	-28148.37976	-28148.37604	-28149.20830	-28149.19138	-28149.17443
1.850	-28148.36722	-28148.36708	-28149.20643	-28149.18818	-28149.17074
1.875				-28149.18331	-28149.16539
1.900				-28149.17696	-28149.15860
			$1^1\Pi_g$		
1.600					
1.625	-28148.35626	-28148.35478	-28149.13304		
1.650	-28148.36945	-28148.36780	-28149.15346	-28149.14364	-28149.13121
1.675	-28148.37873	-28148.37686	-28149.16986	-28149.15927	-28149.14615
1.700	-28148.38454	-28148.38242	-28149.18268	-28149.17123	-28149.15742
1.725	-28148.38735	-28148.38494	-28149.19225	-28149.17990	-28149.16544
1.750	-28148.38470	-28148.38462	-28149.19887	-28149.18564	-28149.17055
1.775	-28148.38189	-28148.38179	-28149.20113	-28149.18876	-28149.17306
1.800	-28148.38015	-28148.37673	-28149.20455	-28149.18952	-28149.17326
1.825	-28148.37345	-28148.36969	-28149.20421	-28149.18817	-28149.17138
1.850	-28148.36099	-28148.36085	-28149.20210	-28149.18494	-28149.16765
1.875				-28149.18004	-28149.16229
1.900				-28149.17369	-28149.15548

S10 Example results for all tested EOM-CC methods with imposed C_{2v} symmetry

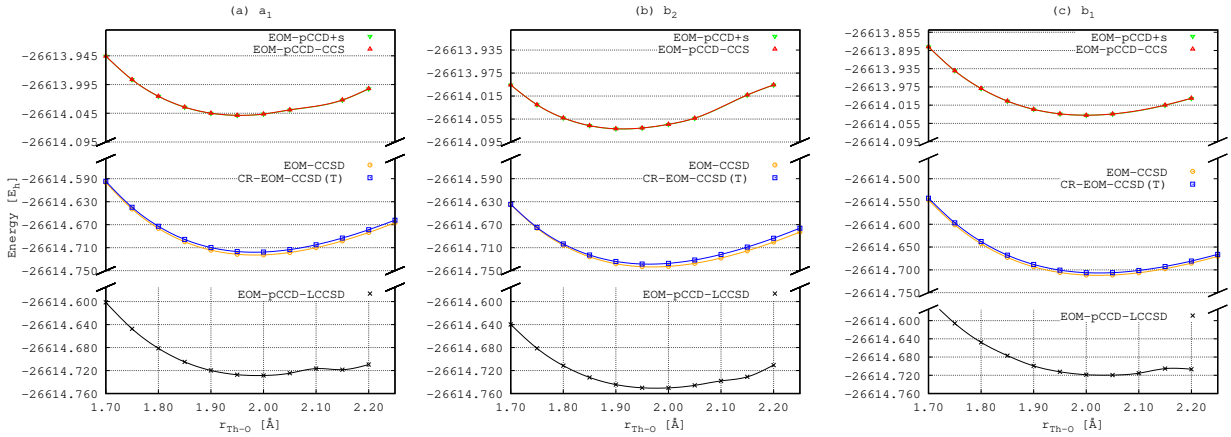

Figure S8: Potential energy surfaces of the lowest-lying excited states of ThO_2 determined for various EOM-CC methods with imposed C_{2v} symmetry.

Table S21: Total electronic energies $[E_h]$ of ThO_2 for different states and various CC methods. All EOM-CC methods are calculated with imposed C_{2v} symmetry.

Distance[Å]	pCCD	pCCD-CCS	pCCD-LCCSD	CCSD	
			$X^1\Sigma^+$		
1.700	-26614.15693	-26614.15693	-26614.75762	-26614.74828	
1.750	-26614.19402	-26614.19401	-26614.79719	-26614.78697	
1.800	-26614.21821	-26614.21821	-26614.82420	-26614.81288	
1.850	-26614.23164	-26614.23163	-26614.84090	-26614.82817	
1.900	-26614.23606	-26614.23606	-26614.84900	-26614.83463	
1.950	-26614.23303	-26614.23302	-26614.85042	-26614.83378	
2.000	-26614.22384	-26614.22383	-26614.84663	-26614.82689	
2.050	-26614.20947	-26614.20947	-26614.83949	-26614.81505	
2.100			-26614.83394	-26614.79921	
2.150	-26614.16884	-26614.16883	-26614.83187	-26614.78017	
2.200	-26614.14410	-26614.14409	-26614.76551	-26614.75860	
2.250				-26614.73509	
Distance[Å]	EOM-pCCD+S	EOM-pCCD-CCS	EOM-pCCD-LCCSD	EOM-CCSD	CR-EOM-CCSD(T)
			a_1		
1.700	-26613.94573	-26613.94555	-26614.60135	-26614.59677	-26614.59453
1.750	-26613.98642	-26613.98642	-26614.64742	-26614.64269	-26614.63972
1.800	-26614.01553	-26614.01533	-26614.68133	-26614.67638	-26614.67279
1.850	-26614.03441	-26614.03420	-26614.70500	-26614.69973	-26614.69564
1.900	-26614.04501	-26614.04478	-26614.71987	-26614.71433	-26614.70984
1.950	-26614.04875	-26614.04851	-26614.72723	-26614.72154	-26614.71676
2.000	-26614.04650	-26614.04624	-26614.72863	-26614.72256	-26614.71759
2.050	-26614.03933	-26614.03903	-26614.72460	-26614.71843	-26614.71336
2.100			-26614.71661	-26614.71006	-26614.70498
2.150	-26614.02219	-26614.02180	-26614.71847	-26614.69823	-26614.69323
2.200	-26614.00257	-26614.00211	-26614.70954	-26614.68361	-26614.67878
2.250				-26614.66675	-26614.66220
			b_2		
1.700	-26613.99584	-26613.99571	-26614.63981	-26614.63485	-26614.63453
1.750	-26614.03016	-26614.03015	-26614.68137	-26614.67610	-26614.67476
1.800	-26614.05323	-26614.05306	-26614.71146	-26614.70585	-26614.70354
1.850	-26614.06664	-26614.06646	-26614.73202	-26614.72597	-26614.72277
1.900	-26614.07217	-26614.07196	-26614.74447	-26614.73798	-26614.73398
1.950	-26614.07076	-26614.07053	-26614.75003	-26614.74317	-26614.73846
2.000	-26614.06442	-26614.06416	-26614.75018	-26614.74264	-26614.73731
2.050	-26614.05374	-26614.05344	-26614.74578	-26614.73732	-26614.73148
2.100			-26614.73808	-26614.72803	-26614.72180
2.150	-26614.01341	-26614.01304	-26614.73097	-26614.71548	-26614.70896
2.200	-26613.99594	-26613.99551	-26614.71045	-26614.70025	-26614.69358
2.250				-26614.68285	-26614.67617
			b_1		
1.700	-26613.88716	-26613.88696	-26614.55140	-26614.54684	-26614.54280
1.750	-26613.93955	-26613.93955	-26614.60606	-26614.60128	-26614.59677
1.800	-26613.97856	-26613.97833	-26614.64771	-26614.64262	-26614.63779
1.850	-26614.00618	-26614.00594	-26614.67681	-26614.67276	-26614.66770
1.900	-26614.02427	-26614.02401	-26614.69934	-26614.69335	-26614.68815
1.950	-26614.03421	-26614.03393	-26614.71233	-26614.70584	-26614.70057
2.000	-26614.03702	-26614.03702	-26614.71875	-26614.71148	-26614.70621
2.050	-26614.03459	-26614.03424	-26614.71958	-26614.71137	-26614.70614
2.100			-26614.71538	-26614.70644	-26614.70131
2.150	-26614.01525	-26614.01480	-26614.70534	-26614.69753	-26614.69256
2.200	-26614.00025	-26613.99973	-26614.70666	-26614.68533	-26614.68058
2.250				-26614.67046	-26614.66601
			a_2		
1.700	-26613.94188	-26613.94175	-26614.58280	-26614.57791	-26614.57731
1.750	-26613.97658	-26613.97658	-26614.62467	-26614.61954	-26614.61798
1.800	-26614.00035	-26614.00018	-26614.65537	-26614.65002	-26614.64759
1.850	-26614.01474	-26614.01456	-26614.67829	-26614.67119	-26614.66800
1.900	-26614.02156	-26614.02135	-26614.69028	-26614.68453	-26614.68069
1.950	-26614.02166	-26614.02142	-26614.69721	-26614.69127	-26614.68691
2.000	-26614.01714	-26614.01686	-26614.69865	-26614.69248	-26614.68772
2.050	-26614.00845	-26614.00814	-26614.69940	-26614.68938	-26614.68438
2.100			-26614.69943	-26614.68573	-26614.68092
2.150	-26613.97251	-26613.97211	-26614.69484	-26614.67863	-26614.67405
2.200	-26613.95817	-26613.95769	-26614.67439	-26614.66802	-26614.66374
2.250				-26614.65455	-26614.65062

S11 Total CPU timing

Table S22: Total timings (in seconds [s]) for different EOM-CC methods. Please note that all the excited state calculations were performed on differenct computing nodes and using different software packages and optimization parameters (like, C_1 or D_{2h} point group symmetry). Thus, these timings are very approximate and should be considered with caution*.

	PIERNIK			NWChem* (version 6.8)	Molpro2012
Molecule	EOM-pCCD+S	EOM-pCCD-CCS	EOM-pCCD-LCCSD	EOM-CCSD + CR-EOM-CCSD(T)	FCIDUMP
ThO	173	262	9'459	24'501	188
ThS	174	254	7'669	30'936	342
ThO_2	233	221	36'836	8'349	74
NUN	91	166	4'510	6'576	35
$UO_2{}^{2+}$	85	146	6'642	10'121	35
$PaO_2{}^{+}$	89	192	6'985	6'633	35
CUO	66	140	3'426	6'785	45
NUO^{+}	66	137	5'252	10'029	47

*Specifically, the EOM-CCSD and CR-EOM-CCSD(T) calculations in NWChem benefit from the D_{2h} and C_{2v} point group symmetries, while all the calculations in Piernik do either not fully exploit the symmetry or have been done relaxing to C_1 point group symmetry. The NWChem code is written in modern CC+/Fortran languages and its fully optimized. The Piernik code is written mainly in Python, where only some few bottleneck operations have been exported to C++, and is not fully optimized, yet. Note that the Davidson diagonalization in Piernik is fully implemented in Python. Finally, the results for NWChem correspond to 5-7 excited states in one irreducible representation, while in Piernik up to 15-20 lowest-lying excited states have been targeted in total.

S12 Experimental reference data

TableS23: Experimental spectroscopic parameters of the ground and some low-lying singlet excited states of ThO,[10,11] ThS,[12] NUN,[13] and ThO_2.[14] r_e is the equilibrium bond length, ω_e the harmonic vibrational frequency, and T_v the excitation energy, respectively.

ThO				ThS			
State	r_e [Å]	ω_e [cm^{-1}]	T_v [eV]	State	r_e [Å]	ω_e [cm^{-1}]	T_v [eV]
$X^1\Sigma$	1.840	896		$X^1\Sigma$		475	
$1^1\Delta$	1.881	816	2.23				
$1^1\Sigma$	1.867	846	1.31				
$1^1\Pi$	1.864	843	1.38				
NUN				ThO_2			
State	r_e [Å]	ω_e [cm^{-1}]	T_v [eV]	State	r_e [Å]	ω_e [cm^{-1}]	T_v [eV]
$X^1\Sigma_g$		1008		$X^1 1A_1$		808	
				B_2		757	

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