

Table S1: Excitation energies derived from all-electron numerical SCF calculations for the states averaged over nonrelativistic configurations of the  $\text{Th}^+$  cation with DCB Hamiltonian and accounting for the finite nuclear size and QED effects. The contributions from various effects are the differences from the results of the all-electron numerical SCF calculations without the QED effects, with the point nucleus, or without Breit/Gaunt interactions. Errors of the GRPPs are estimated as differences between the results of the all-electron calculations and the calculations with corresponding GRPP versions. All data are in  $\text{cm}^{-1}$ .

	Exc. energy	Contributions from					Errors of	
	DCB+QED	Finite					Full	Valence
$\dots 6d^1 7s^2 \rightarrow$	Finite nucl.	QED	nucl.	Breit	Gaunt	Retard.	GRPP	GRPP
$\dots 6d^2 7s^1$	1836	-190	-117	-141	-154	13	-1	-42
$\dots 6d^3$	10060	-357	-220	-268	-292	25	-1	-76
$\dots 6d^1 7s^1 7p^1$	25623	-124	-90	4	13	-9	3	-4
$\dots 6d^2 7p^1$	30182	-313	-205	-148	-154	6	2	-48
$\dots 5f^1 7s^2$	6231	-156	-69	-741	-869	128	-35	-556
$\dots 5f^1 6d^1 7s^1$	11266	-324	-173	-854	-991	137	-36	-577
$\dots 5f^1 6d^2$	22205	-471	-264	-951	-1096	145	-36	-592
$\dots 5f^1 7s^1 7p^1$	29767	-266	-149	-739	-859	120	-33	-563
$\dots 5f^1 6d^1 7p^1$	37469	-435	-252	-864	-995	131	-34	-587
$\dots 5f^2 7s^1$	37289	-429	-217	-1399	-1632	233	-65	-953
$\dots 5f^2 6d^1$	49949	-551	-295	-1447	-1680	233	-63	-935
$\dots 5f^2 7p^1$	61398	-529	-289	-1423	-1653	230	-64	-974

Table S2: SCF excitation energies for the  $U^{2+}$  cation. See the caption of Table S1

	Exc. energy	Contributions from					Errors of	
	DCB+QED Finite nucl.	QED	Finite nucl.	Breit	Gaunt	Retard.	Full GRPP	Valence GRPP
$\dots 5f^3 7s^1 \rightarrow$								
$\dots 5f^3 6d^1$	3623	-204	146	-120	-128	8	1	-7
$\dots 5f^3 7p^1$	35916	-149	125	-6	3	-9	2	-11
$\dots 5f^4$	11129	-319	197	-762	-884	122	-47	-439
$\dots 5f^2 6d^1 7s^1$	15741	158	-75	797	936	-139	55	578
$\dots 5f^2 6d^2$	15722	-68	86	652	780	-128	56	555
$\dots 5f^2 7s^2$	23143	408	-253	952	1103	-151	55	606
$\dots 5f^2 6d^1 7p^1$	54002	-6	61	793	942	-149	58	569
$\dots 5f^2 7s^1 7p^1$	58022	242	-113	959	1122	-163	57	598
$\dots 5f^1 6d^3$	49267	85	18	1534	1817	-283	117	1242
$\dots 5f^1 6d^2 7s^1$	53343	332	-157	1699	1995	-296	115	1277
$\dots 5f^1 6d^1 7s^2$	65121	603	-350	1875	2185	-310	114	1318
$\dots 5f^1 6d^2 7p^1$	93849	153	-9	1694	2001	-307	119	1269
$\dots 5f^1 6d^1 7s^1 7p^1$	102166	421	-197	1881	2204	-323	117	1311
$\dots 5f^1 7s^2 7p^1$	118117	712	-401	2078	2417	-339	115	1360

Table S3: Basis set for Th (adapted for all-electron calculations)

s	p	d	f		
5.7372472E+07	7.2974232E+07	5.3945587E+05	1.9495163E+03		
1.5274857E+07	2.6222425E+07	1.2481597E+05	6.5321253E+02		
5.2287218E+06	9.6396368E+06	3.9067151E+04	2.7657880E+02		
1.9911461E+06	3.7180139E+06	1.4490467E+04	1.3221675E+02		
8.3294306E+05	1.4914786E+06	6.0646913E+03	6.7322922E+01		
3.7024359E+05	6.1958412E+05	2.7754714E+03	3.5748720E+01		
1.7401271E+05	2.6538254E+05	1.3603469E+03	1.9282595E+01		
8.5351033E+04	1.1694933E+05	7.0308463E+02	1.0420910E+01		
4.3522358E+04	5.2967927E+04	3.7817822E+02	5.5494732E+00		
2.2897565E+04	2.4647085E+04	2.1008995E+02	2.8546293E+00		
1.2365131E+04	1.1793294E+04	1.1944362E+02	1.3616238E+00		
6.8123949E+03	5.8112196E+03	6.9167371E+01	6.0962858E-01		
3.8216976E+03	2.9520515E+03	4.0526574E+01	2.5283879E-01		
2.1876513E+03	1.5459858E+03	2.3754294E+01	9.1321054E-02		
1.2845154E+03	8.3271616E+02	1.3812695E+01	0.050404710		
7.7514556E+02	4.5979539E+02	8.0361154E+00			
4.7794411E+02	2.5964223E+02	4.6268519E+00			
3.0108104E+02	1.4905823E+02	2.5969052E+00			
1.9472481E+02	8.6644660E+01	1.4346916E+00			
1.2576401E+02	5.1449971E+01	7.6657941E-01			
7.9566905E+01	3.0555992E+01	3.7837468E-01			
4.9492239E+01	1.7980889E+01	1.8023811E-01			
3.0215779E+01	1.0714043E+01	8.2104366E-02			
1.8217418E+01	6.3449373E+00	3.5428853E-02			
1.1330243E+01	3.6913390E+00				
7.0264506E+00	2.1349318E+00				
4.3179378E+00	1.2173577E+00				
2.5703476E+00	6.7609571E-01				
1.5336087E+00	3.6904041E-01				
9.2576591E-01	1.9816814E-01				
5.3273696E-01	9.8623375E-02				
3.0732299E-01	4.9499235E-02				
1.7263001E-01	2.4523811E-02				
1.1178547E-01	1.2048163E-02				
6.2052232E-02					
3.3255867E-02					
1.7363052E-02					
g					
4.051400	-0.39050416	-0.61954067	-0.96399632	1.62489561	1.83082198
2.626800	-0.14497714	-0.12104339	0.53736468	-1.90617971	-3.03581936
1.253372	-0.43643468	0.06754934	0.60776808	0.17506986	2.32246997
0.598043	-0.22671258	0.57762149	-0.03877817	0.78677621	-1.23439570
0.285355	-0.11743458	0.18337172	-0.56850726	-0.43180841	-0.04663287
0.125406	-0.01366788	0.10101326	-0.20312263	-0.38775773	0.49713826
0.070000	-0.00788183	-0.00473646	-0.05147319	-0.07013222	0.11908926
h					
3.355101	0.60387854	0.86268421	0.97574163	0.68874430	
1.717548	0.20048876	-0.31035087	-1.43221801	-1.56791538	
0.829135	0.40376963	-0.41481115	0.37786594	1.73039803	
0.400260	0.04311932	-0.35643139	0.38817295	-0.86490159	
0.193223	0.05181809	-0.01801598	0.24106505	-0.24693901	
0.093277	-0.00896431	-0.02460562	0.01017355	-0.16069223	
i					
4.170400	-0.60702568	-0.78168583	-0.88597888		
2.085200	-0.27775539	0.25844373	1.33775876		
1.042600	-0.32522282	0.48122880	-0.40385773		
0.521300	-0.05639583	0.31526697	-0.40950817		
0.260650	-0.02799659	0.03433980	-0.18926618		

Table S4: Basis set for Th (adapted for GRPP calculations)

s	p	d	f		
160.0719800	82.16746000	522.5137000	1748.457720		
87.56533000	65.73397000	134.0478700	613.2081400		
67.66944000	52.58717000	46.44729000	264.9269100		
45.23225000	30.55599200	37.15783000	129.3075800		
30.21577900	17.98088900	23.75429400	67.04702000		
18.21741800	10.71404300	13.81269500	35.74872000		
11.33024300	6.344937300	8.036115400	19.28259500		
7.026450600	3.691339000	4.626851900	10.42091000		
4.317937800	2.134931800	2.596905200	5.549473200		
2.570347600	1.217357700	1.434691600	2.854629300		
1.533608700	0.676095710	0.766579410	1.361623800		
0.925765910	0.369040410	0.378374680	0.609628580		
0.532736960	0.198168140	0.180238110	0.252838790		
0.307322990	0.098623375	0.082104366	0.091321054		
0.172630010	0.049499235	0.035428853	0.050404710		
0.111785470	0.024523811				
0.062052232	0.012048163				
0.033255867					
0.017363052					
g					
4.051400	-0.39050416	-0.61954067	-0.96399632	1.62489561	1.83082198
2.626800	-0.14497714	-0.12104339	0.53736468	-1.90617971	-3.03581936
1.253372	-0.43643468	0.06754934	0.60776808	0.17506986	2.32246997
0.598043	-0.22671258	0.57762149	-0.03877817	0.78677621	-1.23439570
0.285355	-0.11743458	0.18337172	-0.56850726	-0.43180841	-0.04663287
0.125406	-0.01366788	0.10101326	-0.20312263	-0.38775773	0.49713826
0.070000	-0.00788183	-0.00473646	-0.05147319	-0.07013222	0.11908926
h					
3.355101	0.60387854	0.86268421	0.97574163	0.68874430	
1.717548	0.20048876	-0.31035087	-1.43221801	-1.56791538	
0.829135	0.40376963	-0.41481115	0.37786594	1.73039803	
0.400260	0.04311932	-0.35643139	0.38817295	-0.86490159	
0.193223	0.05181809	-0.01801598	0.24106505	-0.24693901	
0.093277	-0.00896431	-0.02460562	0.01017355	-0.16069223	
i					
4.170400	-0.60702568	-0.78168583	-0.88597888		
2.085200	-0.27775539	0.25844373	1.33775876		
1.042600	-0.32522282	0.48122880	-0.40385773		
0.521300	-0.05639583	0.31526697	-0.40950817		
0.260650	-0.02799659	0.03433980	-0.18926618		

Table S5: Basis set for U (adapted for all-electron calculations)

s	p	d	f		
56697466.00000000	78339499.00000000	533180.91000000	1163.36020000		
15093688.00000000	29808990.00000000	123237.63000000	573.50499000		
5164101.30000000	11443420.00000000	38642.36400000	285.71163000		
1964638.80000000	4581199.80000000	14357.18700000	136.47329000		
820774.81000000	1897246.00000000	6012.79360000	69.43427500		
364106.97000000	809797.42000000	2754.36670000	36.84465700		
170668.08000000	355067.41000000	1351.96790000	19.84804900		
83455.39000000	159603.26000000	699.79805000	10.72168500		
42423.37700000	73450.10100000	377.01141000	5.70450120		
22228.77400000	34594.86700000	209.73941000	2.92433440		
11931.71300000	16686.68700000	119.30191000	1.43960400		
6539.04200000	8256.41640000	69.11171400	0.67385930		
3678.23270000	4196.77840000	40.43241500	0.29434970		
2139.54940000	2191.89420000	23.65423900	0.11323985		
1288.45330000	1174.82830000	13.80639900			
792.07718000	644.61478000	8.06231670			
484.69932000	361.41388000	4.66375400			
298.00127000	206.27034000	2.63565040			
186.73959000	119.33670000	1.45796000			
120.33238000	70.14308600	0.77481216			
75.85031400	41.70407000	0.37248472			
49.22312100	24.70288000	0.17195534			
30.97937000	14.49639800	0.07613361			
19.33898600	8.57587000				
11.90917900	4.98086080				
7.28108690	2.84040830				
4.52381560	1.59903720				
2.69147870	0.86476597				
1.62933420	0.45724205				
1.02810670	0.23893352				
0.60588439	0.11879300				
0.35564698	0.05700662				
0.20180019					
0.12481356					
0.06383616					
g					
28.78919130	0.00068974	-0.00118120	-0.00737416	0.00494647	-0.10867784
14.01320380	-0.00898795	-0.05421964	0.17284128	-0.36594951	-0.33163329
6.82095857	0.07975524	0.18134441	-0.50920321	1.20716202	1.79274020
3.32011698	0.29993353	0.65729862	-0.50617914	-0.67702022	-2.65141994
1.61607443	0.39302861	0.04007231	1.01049051	-0.71780643	2.66362756
0.78662787	0.34151054	-0.44799971	0.05077223	1.59628888	-2.01035588
0.38289289	0.18576214	-0.38383279	-0.74551549	-1.12509753	0.90492997
h					
14.01320380	-0.02449353	-0.07337288	-0.15906456	-0.23132505	
6.82095857	0.10594999	0.30442614	0.71623431	1.22980099	
3.32011698	0.35765256	0.65468802	0.09614021	-1.24473080	
1.61607443	0.43019757	-0.18764249	-0.95514471	0.32540556	
0.78662787	0.31957043	-0.48353526	0.41946211	0.73545263	
0.38289289	0.10197395	-0.22548628	0.48672026	-0.90679010	
i					
6.82095857	0.12524957	-0.37984959	0.81416792		
3.32011698	0.42961205	-0.61109425	-0.35518331		
1.61607443	0.42313291	0.38916709	-0.73501069		
0.78662787	0.31396902	0.50454838	0.88347742		

Table S6: Basis set for U (adapted for GRPP calculations)

s	p	d	f		
11931.71300000	8256.41640000	6012.79360000	1163.36020000		
6539.04200000	4196.77840000	2754.36670000	573.50499000		
3678.23270000	2191.89420000	1351.96790000	285.71163000		
2139.54940000	1174.82830000	699.79805000	136.47329000		
1288.45330000	644.61478000	377.01141000	69.43427500		
792.07718000	361.41388000	209.73941000	36.84465700		
484.69932000	206.27034000	119.30191000	19.84804900		
298.00127000	119.33670000	69.11171400	10.72168500		
186.73959000	70.14308600	40.43241500	5.70450120		
120.33238000	41.70407000	23.65423900	2.92433440		
75.85031400	24.70288000	13.80639900	1.43960400		
49.22312100	14.49639800	8.06231670	0.67385930		
30.97937000	8.57587000	4.66375400	0.29434970		
19.33898600	4.98086080	2.63565040	0.11323985		
11.90917900	2.84040830	1.45796000			
7.28108690	1.59903720	0.77481216			
4.52381560	0.86476597	0.37248472			
2.69147870	0.45724205	0.17195534			
1.62933420	0.23893352	0.07613361			
1.02810670	0.11879300				
0.60588439	0.05700662				
0.35564698					
0.20180019					
0.12481356					
0.06383616					
g					
28.78919130	0.00068974	-0.00118120	-0.00737416	0.00494647	-0.10867784
14.01320380	-0.00898795	-0.05421964	0.17284128	-0.36594951	-0.33163329
6.82095857	0.07975524	0.18134441	-0.50920321	1.20716202	1.79274020
3.32011698	0.29993353	0.65729862	-0.50617914	-0.67702022	-2.65141994
1.61607443	0.39302861	0.04007231	1.01049051	-0.71780643	2.66362756
0.78662787	0.34151054	-0.44799971	0.05077223	1.59628888	-2.01035588
0.38289289	0.18576214	-0.38383279	-0.74551549	-1.12509753	0.90492997
h					
14.01320380	-0.02449353	-0.07337288	-0.15906456	-0.23132505	
6.82095857	0.10594999	0.30442614	0.71623431	1.22980099	
3.32011698	0.35765256	0.65468802	0.09614021	-1.24473080	
1.61607443	0.43019757	-0.18764249	-0.95514471	0.32540556	
0.78662787	0.31957043	-0.48353526	0.41946211	0.73545263	
0.38289289	0.10197395	-0.22548628	0.48672026	-0.90679010	
i					
6.82095857	0.12524957	-0.37984959	0.81416792		
3.32011698	0.42961205	-0.61109425	-0.35518331		
1.61607443	0.42313291	0.38916709	-0.73501069		
0.78662787	0.31396902	0.50454838	0.88347742		