Towards systematically improvable models for actinides in condensed phase: the electronic spectrum of uranyl in $Cs_2UO_2Cl_4$ as a test case †

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1 Supplementary Materials

1.1 Computational Details

Table 1 Approximate computational cost in terms of processor (in single-cpu elapsed time, in hours) and memory (in Gb per parallel process) usage for the calculations in this work.

		CAM-B3LYP		IHFSCC		
Model	Symmetry used	CPU	Memory	space	CPU	Memory
UO_2^{2+} (**a**)	D_{2h}	200	2			
UO_2^{2+} (**a**)	$D_{\infty h}$			Q_2	150	10
				Q_3	550	16
UO_2^{2+}(**b**)	D_{2h}	200	2	Q_2	700	26
UO_2^{2+}(**c**)	D_{2h}	200	2	Q_1	450	16
				Q_2	700	26
				Q_3	1600	64
UO_2^{2+}(**c'**), (**e**), (**f**)	C_{2h}	500	2	Q_1	600	30
$UO_2Cl_4^{2-}$ (**d**)	D_{2h}	2000	2			

1.2 Excited-state Compositon, DFT calculations

Below one finds the (occupied and virtual) CAM-B3LYP spinors for the different embedded models discussed in the text having the most significant contributions to the excited states as shown in tables 2, 3, 4 and 5. These spinors are labelled by their indexes, and we present their energies, classification in terms of the isolated uranyl species, and their composition in terms of its atomic components (a and b denote α and β components, respectively) from a Mulliken population analysis.

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uranyl embedded in point charges (model b), DFT (CAM-B3LYP) spinors, D2h symmetry
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occupied
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24: -0.5830888676094 (sigma)
   Total    B1uU  pz   B1uU  fxxz B1uU  fyyz B1uU  fzzz B1uO1 s    B1uO1 pz   B2uU  py   B3uU  px
a  0.9186   0.3044     0.0255     0.0255    -0.0229     0.4762     0.1064     0.0000     0.0000
b  0.0814   0.0000     0.0000     0.0000     0.0000     0.0000     0.0000     0.0372     0.0372

25: -0.2629209834884 (pi)
   Total    B1uU  fxxz B1uU  fyyz B1uU  fzzz B1uO1 pz   B2uU  fyyy B2uU  fyzz B2uO1 py   B3uU  fxxx B3uU  fxzz B3uO1 px
a  0.1480   0.0240     0.0240     0.0202     0.0716     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000
b  0.8520   0.0000     0.0000     0.0000     0.0000     0.0178     0.0842     0.3143     0.0178     0.0842     0.3143

27: -0.2265131707659 (sigma)
   Total    B1uU  pz   B1uU  fxxz B1uU  fyyz B1uU  fzzz B1uO1 pz   B2uO1 py   B3uO1 px
a  0.8485   0.0880     0.1782     0.1782     0.1042     0.3052     0.0000     0.0000
b  0.1515   0.0000     0.0000     0.0000     0.0000     0.0000     0.0580     0.0580


virtuals
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28:  0.0887695550 (f phi)
   Total    B1uU  fxxz B1uU  fyyz B2uU  fxxy B2uU  fyyy B3uU  fxxx B3uU  fxyy Au U  fxyz
a  0.1215   0.0208     0.0208     0.0000     0.0000     0.0000     0.0000     0.0790
b  0.8785   0.0000     0.0000     0.3310     0.1079     0.1079     0.3310     0.0000

29:  0.0991093761 (f delta)
   Total    B1uU  fxxz B1uU  fyyz B2uU  fyyy B2uU  fyzz B2uO1 py   B3uU  fxxx B3uU  fxzz B3uO1 px   Au U  fxyz
a  0.9528   0.2279     0.2279     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000     0.4925
b  0.0472   0.0000     0.0000     0.0038     0.0102     0.0091     0.0038     0.0102     0.0091     0.0000

30:  0.1216325649320 (f phi)
   Total    B2uU  fxxy B2uU  fyyy B3uU  fxxx B3uU  fxyy
a  0.0004   0.0000     0.0000     0.0000     0.0000
b  0.9996   0.3805     0.1183     0.1183     0.3805

31:  0.1252627134450 (f delta)
   Total    B1uU  fxxz B1uU  fyyz B2uU  fxxy B2uU  fyyy B3uU  fxxx B3uU  fxyy Au U  fxyz
a  0.8826   0.2370     0.2370     0.0000     0.0000     0.0000     0.0000     0.4037
b  0.1174   0.0000     0.0000     0.0447     0.0136     0.0136     0.0447     0.0000
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uranyl embedded in chlorides (model c), DFT-in-DFT (CAM-B3LYP) spinors, D2h symmetry
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occupied
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24: -0.5151039102219 (sigma)
   Total    B1uU  pz   B1uU  fxxz B1uU  fyyz B1uU  fzzz B1uO1 s    B1uO1 pz   B2uU  py   B3uU  px
a  0.9115   0.3075     0.0217     0.0217    -0.0176     0.4546     0.1200     0.0000     0.0000
b  0.0885   0.0000     0.0000     0.0000     0.0000     0.0000     0.0000     0.0390     0.0390

25: -0.1960531813424 (pi)
   Total    B1uU  fxxz B1uU  fyyz B1uU  fzzz B1uO1 pz   B2uU  fyyy B2uU  fyzz B2uO1 py   B3uU  fxxx B3uU  fxzz B3uO1 px
a  0.1384   0.0221     0.0221     0.0173     0.0692     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000
b  0.8616   0.0000     0.0000     0.0000     0.0000     0.0165     0.0803     0.3227     0.0165     0.0803     0.3227

27: -0.1558499625713 (sigma)
   Total    B1uU  pz   B1uU  fxxz B1uU  fyyz B1uU  fzzz B1uO1 pz   B2uO1 py   B3uO1 px
a  0.8574   0.0902     0.1823     0.1823     0.0946     0.3059     0.0000     0.0000
b  0.1426   0.0000     0.0000     0.0000     0.0000     0.0000     0.0535     0.0535


virtuals
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28:  0.1688544518396 (f phi)
   Total    B2uU  fxxy B2uU  fyyy B3uU  fxxx B3uU  fxyy Au U  fxyz
a  0.2077   0.0000     0.0000     0.0000     0.0000     0.1902
b  0.7923   0.3009     0.0922     0.0922     0.3009     0.0000

29:  0.1742104158004 (f delta)
   Total    B1uU  fxxz B1uU  fyyz B2uU  fxxy B2uU  fyyy B2uU  fyzz B3uU  fxxx B3uU  fxyy B3uU  fxzz Au U  fxyz
a  0.8989   0.2387     0.2387     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000     0.4181
b  0.1011   0.0000     0.0000     0.0145     0.0161     0.0110     0.0161     0.0145     0.0110     0.0000

30:  0.1934793569545 (f sigma)
   Total    B1uU  pz   B1uU  fxxz B1uU  fyyz B1uU  fzzz B1uO1 s    B1uO1 pz
a  0.9994   0.3101     0.0953     0.0953    -0.1715     0.9509    -0.2796
b  0.0006   0.0000     0.0000     0.0000     0.0000     0.0000     0.0000

31:  0.2015400224341 (f delta)
   Total    B1uU  fxxz B1uU  fyyz B2uU  fxxy B2uU  fyyy B3uU  fxxx B3uU  fxyy Au U  fxyz
a  0.8398   0.2346     0.2346     0.0000     0.0000     0.0000     0.0000     0.3670
b  0.1602   0.0000     0.0000     0.0603     0.0172     0.0172     0.0603     0.0000

32:  0.2029131747762 (f phi)
   Total    B2uU  py   B3uU  px   B3uU  fxxx B3uU  fxyy B2uU  fxxy B2uU  fyyy
a  0.0010   0.0000     0.0000     0.0000     0.0000     0.0000     0.0000
b  0.9990   0.0155     0.0155     0.1019     0.3717     0.3717     0.1019
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uranyl chloride (model d), CAM-B3LYP supermolecular spinors, D2h symmetry
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occupied
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37: -0.1651021546052 (pi)
   Total    B1uU  fxxz B1uU  fyyz B1uU  fzzz B1uO1 s    B1uO1 pz   B2uU  fyyy B2uU  fyzz B2uO1 py  B3uU  fxxx B3uU  fxzz
a  0.1234   0.0186     0.0186     0.0135     0.0042     0.0596     0.0000     0.0000     0.0000     0.0000     0.0000
b  0.8766   0.0000     0.0000     0.0000     0.0000     0.0000     0.0167     0.0731     0.3448     0.0167     0.0731

            B3uO1 px
            0.0000
            0.3448

39: -0.1360767017907 (sigma)
   Total    B1uU  pz   B1uU  fxxz B1uU  fyyz B1uU  fzzz B1uO1 s    B1uO1 pz   B1uCl1pz   B1uCl3pz   B2uO1 py   B2uCl1py
a  0.8353   0.0519     0.1357     0.1357     0.0645     0.0122     0.2465     0.0928     0.0928     0.0000     0.0000
b  0.1647   0.0000     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000     0.0402     0.0263

            B2uO1 py   B2uCl1py
            0.0000     0.0000
            0.0402     0.0263

45: -0.0696732830   (sigma, mixed with ligands)
   Total    B1uU  pz   B1uU  fxxz B1uU  fyyz B1uU  fzzz B1uO1 pz   B1uCl1pz   B1uCl3pz   B2uCl1py   B2uCl3py   B3uCl1px
a  0.8906   0.0267     0.0448     0.0448     0.0206     0.0892     0.3330     0.3330     0.0000     0.0000     0.0000
b  0.1094   0.0000     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000     0.0318     0.0132     0.0132

            B3uCl3px
            0.0000
            0.0318

virtuals
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47:  0.1944409658987 (f phi)
   Total    B2uU  py   B2uU  fxxy B2uU  fyyy B2uU  fyzz B2uO1 py   B2uCl1s    B2uCl1py   B2uCl3py   B3uU  px   B3uU  fxxx
a  0.0016   0.0000     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000
b  0.9984   0.2253     0.0611    -0.0698     0.0436     0.0398     0.5680    -0.3384    -0.0338     0.2253    -0.0698

            B3uU  fxyy B3uU  fxzz B3uO1 px   B3uCl1px   B3uCl3s    B3uCl3px
            0.0000     0.0000     0.0000     0.0000     0.0000
            0.0611     0.0436     0.0398    -0.0338     0.5680    -0.3384

49:  0.2129379666929 (f phi)
   Total    B1uU  fxxz B1uU  fyyz B2uU  py   B2uU  fxxy B2uU  fyyy B2uCl1py   B3uU  fxxx B3uU  fxyy B3uU  fxzz B3uCl3px
a  0.1587   0.0141     0.0141     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000
b  0.8413   0.0000     0.0000    -0.0405     0.2919     0.0782     0.0604     0.0782     0.2919     0.0074     0.0604

            Au U  fxyz
            0.1281
            0.0000

50:  0.2193643484532 (f delta)
   Total    B1uU  fxxz B1uU  fyyz Au U  fxyz
a  0.9431   0.2459     0.2459     0.4261
b  0.0569   0.0000     0.0000     0.0000

51:  0.2447874928375 (f phi)
   Total    B2uU  py   B2uU  fxxy B2uU  fyyy B2uU  fyzz B2uCl1py   B2uCl3py   B3uU  px   B3uU  fxxx B3uU  fxyy B3uU  fxzz
a  0.0017   0.0000     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000
b  0.9983   0.0539     0.3716     0.0847     0.0102     0.0667     0.0127    -0.0539     0.0847     0.3716     0.0102

            B3uCl1px
            0.0000
            0.0127

52:  0.2448286388613 (f delta)
   Total    B1uU  fxxz B1uU  fyyz B2uU  fxxy B2uU  fyyy B2uCl1py  B3uU  fxxx B3uU  fxyy B3uCl3px   Au U  fxyz
a  0.8551   0.2072     0.2072     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000     0.4188
b  0.1449   0.0000     0.0000     0.0521     0.0136     0.0135     0.0136     0.0521     0.0135     0.0000
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uranyl embedded in chlorides (model c'), DFT-in-DFT (CAM-B3LYP) spinors, C2h symmetry
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occupied
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24: -0.5152636082084 (sigma)
   Total    Bu U  py   Bu U  pz   Bu U  fxxz Bu U  fyyz Bu U  fzzz Bu O1 s    Bu O1 pz   Au U  px
a  0.0887   0.0383     0.0002     0.0000     0.0000     0.0000     0.0001     0.0000     0.0396
b  0.9113   0.0002     0.3072     0.0214     0.0219    -0.0176     0.4545     0.1199     0.0000

25: -0.1962044767509 (pi)
   Total    Bu U  fxxz Bu U  fyyy Bu U  fyyz Bu U  fyzz Bu U  fzzz Bu O1 py   Bu O1 pz   Au U  fxxx Au U  fxzz Au O1 px
a  0.8616   0.0000     0.0181     0.0001     0.0832     0.0000     0.3344     0.0002     0.0165     0.0773     0.3109
b  0.1384   0.0218     0.0000     0.0223     0.0000     0.0173     0.0002     0.0691     0.0000     0.0000     0.0000

27: -0.1560123223629 (sigma)
   Total    Bu U  pz   Bu U  fxxz Bu U  fyyz Bu U  fzzz Bu O1 py   Bu O1 pz   Au O1 px
a  0.1429   0.0000     0.0001     0.0001     0.0001     0.0522     0.0000     0.0549
b  0.8571   0.0901     0.1798     0.1841     0.0944     0.0000     0.3057     0.0000


virtuals
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28:  0.1686603174558 (f phi)
   Total    Bu U  fxxy Bu U  fxxz Bu U  fyyy Bu U  fyyz Au U  fxxx Au U  fxyy Au U  fxyz
a  0.7929   0.2873     0.0002     0.1043     0.0003     0.1041     0.2902     0.0005
b  0.2071   0.0000     0.0945     0.0000     0.0946     0.0000     0.0000     0.0167

29:  0.1740341979842 (f delta)
   Total    Bu U  fxxy Bu U  fxxz Bu U  fyyz Bu U  fyzz Au U  fxyy Au U  fxyz Au U  fxzz
a  0.1049   0.0283     0.0009     0.0012     0.0106     0.0295     0.0017     0.0112
b  0.8951   0.0000     0.2093     0.2066     0.0004     0.0001     0.4749     0.0001

30:  0.1934686730588 (f sigma)
   Total    Bu U  pz   Bu U  fxxz Bu U  fyyz Bu U  fzzz Bu O1 s    Bu O1 pz
a  0.0010   0.0002     0.0000     0.0000    -0.0001     0.0004    -0.0001
b  0.9990   0.3097     0.0947     0.0958    -0.1713     0.9507    -0.2795

31:  0.2013514193710 (f delta)
   Total    Bu U  py   Bu U  fxxy Bu U  fxxz Bu U  fyyy Bu U  fyyz Au U  fxxx Au U  fxyy Au U  fxyz
a  0.1707   0.0015     0.0512     0.0024     0.0205     0.0021     0.0223     0.0579     0.0091
b  0.8293   0.0001     0.0042     0.1819     0.0011     0.1798     0.0001     0.0000     0.4582

32:  0.2027237970337 (f phi)
   Total    Bu U  py     L Bu U  fxxy   L Bu U  fyyy   L Au U  fxxx   L Au U  fxyy
a  0.9981   0.0224         0.3354         0.1361         0.1338         0.3385
b  0.0019   0.0000         0.0006         0.0002         0.0000         0.0000
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uranyl embedded in crystal (model f), DFT-in-DFT (CAM-B3LYP) spinors, C2h symmetry
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occupied
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24: -0.7015613698127 (sigma)
   Total    Bu U  py   Bu U  pz   Bu U  fxxz Bu U  fyyz Bu U  fzzz Bu O1 s    Bu O1 pz   Au U  px
a  0.0877   0.0382     0.0002     0.0000     0.0000     0.0000     0.0001     0.0000     0.0390
b  0.9123   0.0002     0.3140     0.0222     0.0222    -0.0152     0.4452     0.1204     0.0000

25: -0.3821699764334 (pi)
   Total    Bu U  fxxz Bu U  fyyy Bu U  fyyz Bu U  fyzz Bu U  fzzz Bu O1 py   Bu O1 pz   Au U  fxxx Au U  fxzz Au O1 px
a  0.8580   0.0000     0.0152     0.0001     0.0739     0.0000     0.2826     0.0001     0.0194     0.0903     0.3541
b  0.1420   0.0227     0.0000     0.0223     0.0000     0.0180     0.0003     0.0718     0.0000     0.0000     0.0000

27: -0.3426637773925 (sigma)
   Total    Bu U  pz   Bu U  fxxz Bu U  fyyz Bu U  fzzz Bu O1 s    Bu O1 py   Bu O1 pz   Au O1 px
a  0.1469   0.0001     0.0001     0.0002     0.0001     0.0000     0.0576     0.0000     0.0518
b  0.8531   0.1048     0.1789     0.1785     0.0950    -0.0166     0.0000     0.3120     0.0000


virtuals
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28: -0.0186933227 (f phi)
   Total    Bu U  fxxy Bu U  fxxz Bu U  fyyy Bu U  fyyz Au U  fxxx Au U  fxyy Au U  fxyz
a  0.8021   0.2911     0.0000     0.1045     0.0000     0.1060     0.2947     0.0001
b  0.1979   0.0000     0.0887     0.0000     0.0896     0.0000     0.0000     0.0184

29: -0.0129180866 (f delta)
   Total    Bu U  fxxy Bu U  fxxz Bu U  fyyz Bu U  fyzz Au U  fxyy Au U  fxyz Au U  fxzz
a  0.0956   0.0256     0.0001     0.0002     0.0105     0.0266     0.0006     0.0114
b  0.9044   0.0000     0.2148     0.2148     0.0004     0.0000     0.4708     0.0001

30:  0.0142992863 (f delta)
   Total    Bu U  fxxy Bu U  fxxz Bu U  fyyy Bu U  fyyz Au U  fxxx Au U  fxyy Au U  fxyz
a  0.1742   0.0385     0.0058     0.0144     0.0059     0.0270     0.0697     0.0099
b  0.8258   0.0035     0.1754     0.0021     0.1771     0.0002     0.0000     0.4629

31:  0.0152942407 (f phi)
   Total    Bu U  fxxy Bu U  fyyy Au U  fxxxAu U  fxyy
a  0.9960   0.3680     0.1427     0.1331    0.3341
b  0.0040   0.0008     0.0003     0.0000    0.0000

32:  0.0303865554 (f sigma)
   Total    Bu U  py   Bu U  pz   Bu U  fxxz Bu U  fyyz Bu U  fyzz Bu U  fzzz Bu O1 s    Bu O1 pz   Bu O1 dzz  Au U  px
a  0.1770   0.0082     0.0032     0.0006     0.0002     0.0138    -0.0012    -0.0013     0.0053    -0.0001     0.0837
b  0.8230   0.0087     0.3625     0.0848     0.0538    -0.0006    -0.1683    -0.1554     0.6456    -0.0214     0.0000

            Au U  fxzz Au O1 px
            0.0287     0.0144
            0.0000     0.0000
```

Table 2 CAM-B3LYP excited state composition in terms of the most significant determinants (in %) for model (**b**).

		orbital pairs (left: occupied, right: virtual) defining the excited determiants.											
E(cm^{-1})	State	(24,28)	(24,29)	(24,30)	(24,31)	(25,28)	(25,29)	(25,30)	(25,31)	(27,28)	(27,29)	(27,30)	(27,31)
17265	I,II	0.1	0.9			0.5	4.0			13.7	81.0	0.6	0.2
16341	III	1.0				3.6	0.8			89.5	6.0		0.1
16239	IV	1.0				3.9	0.6			91.4	3.9		0.2
17681	V,VI	0.7	0.2	0.1		2.8	0.7	0.2	0.2	74.5	14.3	5.1	1.9
19394	VII		0.7		0.3	0.1	2.1		1.2	4.1	74.1		18.5
19660	VIII		0.6		0.4	0.1	1.9		1.3	5.6	68.2		22.8
23424	IX,X			0.1	1.0			0.6	3.5	0.5		8.5	86.3
23875	XI			1.0				4.2				94.8	
23876	XII			1.0				4.2				94.7	

Table 3 CAM-B3LYP excited state composition in terms of the most significant determinants (in %) for model (**c**).

		orbital pairs (left: occupied, right: virtual) defining the excited determiants.											
E(cm^{-1})	State	(24,28)	(24,29)	(24,31)	(24,32)	(25,28)	(25,29)	(25,31)	(25,32)	(27,28)	(27,29)	(27,31)	(27,32)
18115	I,II	0.2	0.9			0.6	3.5			15.3	80.4		0.1
18321	III	0.8	0.2			2.2	1.7			71.7	23.9	0.3	
17981	IV	1.0				3.5	0.6	0.1		89.9	5.0	0.9	
19565	V,VI	0.6	0.1		0.1	2.5	0.5	0.3	0.2	73.3	14.8	3.4	4.4
20539	VII		0.7	0.3		0.2	1.6	0.9		6.0	74.2	16.9	
20829	VIII	0.1	0.5	0.4		0.4	1.1	1.2		20.9	52.4	23.6	
24747	IX,X			1.0	0.1			3.0	0.5	1.3	0.2	86.4	7.3
26137	XI				0.9				3.4				91.3
26140	XII				0.9				3.5				91.3

Table 4 CAM-B3LYP excited state composition in terms of the most significant determinants (in %) for model (**d**).

		orbital pairs (left: occupied, right: virtual) defining the excited determiants.											
E(cm^{-1})	State	(37,49)	(37,50)	(37,52)	(39,49)	(39,50)	(39,51)	(39,52)	(45,47)	(45,49)	(45,50)	(45,51)	(45,52)
19018	I,II	0.3	2.4		4.1	34.8		0.3		6.1	48.8	0.1	0.3
19934	III	1.4	1.0	0.1	24.6	9.9		0.8	1.4	41.8	13.8		0.9
19288	IV	1.8	0.8	0.2	26.9	8.3		2.1	0.7	42.7	10.7		2.5
20970	V,VI	1.6	0.3	0.3	26.7	4.0	1.5	2.8	1.4	45.9	6.3	2.1	3.0
21745	VII	0.4	0.8	0.5	8.0	21.0		6.4	1.0	14.3	35.2		8.1
21592	VIII	0.3	0.9	0.5	9.1	20.6		6.7	0.2	15.4	34.0		8.6
25531	IX,X			2.0	0.9		2.8	33.1	0.2	2.5		3.7	50.1
27058	XI		2.2		34.7				57.1				
27112	XII		2.2		35.0				57.2				

Table 5 CAM-B3LYP excited state composition in terms of the most significant determinants (in %) for models (**c'**) and (**f**).

		orbital pairs (left: occupied, right: virtual) defining the excited determiants.														
E(cm^{-1})	State	(24,28)	(24,29)	(24,30)	(24,31)	(24,32)	(25,28)	(25,29)	(25,30)	(25,31)	(25,32)	(27,28)	(27,29)	(27,30)	(27,31)	(27,32)
							model (**c'**)									
18114	I	0.2	0.9				0.6	3.4				15.7	8		0.1	0.1
18112	II	0.2	0.9				0.6	3.4				16.0	79.7		0.1	0.1
17975	III	0.9					3.5	0.6		0.1		89.4	5.5		0.9	
18317	IV	0.8	0.2				2.2	1.7				71.0	24.5		0.3	
19568	V	0.6	0.1			0.1	2.5	0.5		0.3	0.2	73.4	14.6		3.4	4.4
19552	VI	0.6	0.1			0.1	2.5	0.5		0.3	0.2	73.5	14.7		3.3	4.4
20536	VII		0.7		0.3		0.2	1.5		0.9		6.0	74.2		16.8	0.1
20825	VIII	0.1	0.5		0.4		0.4	1.1		1.1		20.8	52.5		23.4	0.1
24749	IX				1.0	0.1				3.0	0.5	1.3	0.2		86.0	7.5
24738	X				1.0	0.1				3.0	0.5	1.2	0.2		86.3	7.7
26131	XI					0.9					3.4				0.4	91.0
26134	XII					0.9					3.4				0.4	90.9
							model (**f**)									
18119	I	0.1	0.9				0.6	3.6				14.9	80.6	0.1	0.1	
18120	II	0.1	0.9				0.6	3.6				14.9	80.5		0.1	
17914	III	1.0					3.6	0.6	0.1			89.9	4.8	0.8		
18236	IV	0.8	0.2				2.4	1.6				74.2	21.3	0.3		
19494	V	0.6	0.1		0.1		2.5	0.6	0.3	0.1		73.6	14.4	3.6	4.3	
19475	VI	0.6	0.1		0.1		2.6	0.5	0.3	0.2		73.7	14.5	2.8	5.0	
20494	VII		0.7	0.3			0.2	1.6	1.0			5.8	74.5	16.7	0.1	
20808	VIII	0.1	0.5	0.4			0.3	1.2	1.2			18.6	54.5	23.7	0.1	
24711	IX			1.0	0.1				3.0	0.7		1.2	0.1	83.7	10.4	
24698	X			1.0					3.2	0.4		1.2	0.1	88.8	5.3	0.1
26014	XI				0.9					3.7				0.3	94.2	0.2
26017	XII				0.9					3.7				0.4	94.0	0.2

1.3 Excited-state Compositon, WFT calculations

Below one finds the (occupied and virtual) Hartree-Fock spinors for the different embedded models discussed in the text and which make up the most significant determinants in the model (P_m) spaces in tables 6, 7, and 8. These spinors are labelled by their indexes in the model space, and we present their energies, classification in terms of the isolated uranyl species, and their omposition in terms of its atomic components (a and b denote α and β components, respectively) from a Mulliken population analysis.

```
-------------------------------------------------------------------------------
uranyl embedded in point charges (model b), WFT (Hartree-Fock) spinors, D2h symmetry
-------------------------------------------------------------------------------

occupied
--------

 4: -0.748372901167   (sigma)
   Total     B1uU  pz    B1uU  fxxz   B1uU  fyyz   B1uU  fzzz   B1uO1 s     B1uO1 pz    B2uU  py    B3uU  px
a  0.9068    0.3466      0.0320       0.0320      -0.0319       0.4038       0.1204       0.0000       0.0000
b  0.0932    0.0000      0.0000       0.0000       0.0000       0.0000       0.0000       0.0427       0.0427

 5: -0.346866056773   (pi)
   Total     B1uU  pz    B1uU  fxxz   B1uU  fyyz   B1uU  fzzz   B1uO1 pz    B2uU  fyzz   B2uO1 py    B3uU  fxzz   B3uO1 px
a  0.4553    0.0218      0.0856       0.0856       0.0615       0.1930       0.0000       0.0000       0.0000       0.0000
b  0.5447    0.0000      0.0000       0.0000       0.0000       0.0000       0.0385       0.2214       0.0385       0.2214

 7: -0.321860829713   (sigma)
   Total     B1uU  pz    B1uU  fxxz   B1uU  fyyz   B1uU  fzzz   B1uO1 pz    B2uU  fyzz   B2uO1 py    B3uU  fxzz   B3uO1 px
a  0.5427    0.0542      0.1217       0.1217       0.0683       0.1769       0.0000       0.0000       0.0000       0.0000
b  0.4573    0.0000      0.0000       0.0000       0.0000       0.0000       0.0209       0.1924       0.0209       0.1924


virtuals
--------

 4:  0.283667326604    (f delta)
   Total     B1uU  fxxz B1uU  fyyz   B2uU  fxxy   B2uU  fyzz   B3uU  fxyy   B3uU  fxzz   Au U  fxyz
a  0.9017    0.1289      0.1289       0.0000       0.0000       0.0000       0.0000       0.6367
b  0.0983    0.0000      0.0000       0.0258       0.0157       0.0258       0.0157       0.0000

 5:  0.288715419916    (f phi)
   Total     B1uU  fxxz B1uU  fyyz   B2uU  fxxy   B2uU  fyyy   B3uU  fxxx   B3uU  fxyy   Au U  fxyz
a  0.3538    0.1624      0.1624       0.0000       0.0000       0.0000       0.0000       0.0258
b  0.6462    0.0000      0.0000       0.2366       0.0855       0.0855       0.2366       0.0000

 6:  0.307673432326    (f delta)
   Total     B1uU  fxxz B1uU  fyyz   B1uO1 dyy    B2uU  fxxy   B2uU  fyyy   B3uU  fxxx   B3uU  fxyy   Au U  fxyz
a  0.6892    0.1871      0.1871       0.0016       0.0000       0.0000       0.0000       0.0000       0.3085
b  0.3108    0.0000      0.0000       0.0000       0.1208       0.0323       0.0323       0.1208       0.0000

 7:  0.314127154931    (f phi)
   Total     B1uU  pz    B1uO1 s      B1uO1 pz     B2uU  fxxy   B2uU  fyyy   B2uU  fyzz   B3uU  fxxx   B3uU  fxyy   B3uU  fxzz
a  0.0149    0.0231     -0.0444       0.0294       0.0000       0.0000       0.0000       0.0000       0.0000       0.0000
b  0.9851    0.0000      0.0000       0.0000       0.3890       0.0672       0.0223       0.0672       0.3890       0.0223

 8:  0.323358044101    (f phi)
   Total     B1uU  pz    B1uU  fxxz   B1uU  fyyz   B1uU  fzzz   B1uO1 s     B1uO1 pz    B2uU  fyyy   B2uU  fyzz   B2uO1 py
a  0.4863    1.9904     -0.1335      -0.1335       0.3714      -2.6872       1.0779       0.0000       0.0000       0.0000
b  0.5137    0.0000      0.0000       0.0000       0.0000       0.0000       0.0000       0.0729       0.1268       0.0550

             B3uU  fxxx  B3uU  fxzz
             0.0000      0.0000
             0.0729      0.1268
```

```
-------------------------------------------------------------------------------
uranyl embedded in chlorides (model c), WFT-in-DFT (Hartree-Fock) spinors, D2h symmetry
-------------------------------------------------------------------------------

occupied
--------

 4: -0.6781437160390  (sigma)
   Total    B1uU  pz   B1uU  fxxz B1uU  fyyz B1uU  fzzz B1uO1 s    B1uO1 pz    B2uU  py   B3uU  px
a  0.8987   0.3489     0.0283     0.0283    -0.0270     0.3840     0.1322     0.0000     0.0000
b  0.1013   0.0000     0.0000     0.0000     0.0000     0.0000     0.0000     0.0452     0.0452

 5: -0.2784351582524  (pi)
   Total    B1uU  pz   B1uU  fxxz B1uU  fyyz B1uU  fzzz B1uO1 pz   B2uU  fyzz B2uO1 py   B3uU  fxzz   B3uO1 px
a  0.3342   0.0134     0.0604     0.0604     0.0408     0.1507     0.0000     0.0000     0.0000       0.0000
b  0.6658   0.0000     0.0000     0.0000     0.0000     0.0000     0.0425     0.2755     0.0425       0.2755

 7: -0.2497842845315  (sigma)
   Total    B1uU  pz   B1uU  fxxz B1uU  fyyz B1uU  fzzz B1uO1 pz  B2uU  fyzz  B2uO1 py    B3uU  fxzz   B3uO1 px
a  0.6634   0.0633     0.1473     0.1473     0.0751     0.2239     0.0000     0.0000     0.0000       0.0000
b  0.3366   0.0000     0.0000     0.0000     0.0000     0.0000     0.0142     0.1402     0.0142       0.1402


virtuals
--------

 6:  0.3543316232608  (f phi)
   Total    B1uU  fxxz B1uU  fyyz B2uU  py   B2uU  fyzz B2uO1 py  B3uU  px   B3uU  fxxx B3uU  fxzz  B3uO1 px    Au U  fxyz
a  0.1431   0.0216     0.0216     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000      0.0000      0.0990
b  0.8569   0.0000     0.0000     0.7739     0.0966    -0.3823     0.7739     0.0400     0.0966     -0.3823      0.0000

 7:  0.3626915741719  (f delta)
   Total    B1uU  fxxz B1uU  fyyz B2uU  py   B2uU  fyyy B2uU  fyzz B2uO1 py   B3uU  px   B3uU  fxxx B3uU  fxzz  B3uO1 px
a  0.7840   0.0903     0.0903     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000      0.0000      0.0000
b  0.2160   0.0000     0.0000     0.2255    -0.0180     0.0217    -0.1262     0.2255    -0.0180      0.0217     -0.1262

            Au U  fxyz
            0.5990
            0.0000

 8:  0.3705754209442  (f phi)
   Total    B1uU  fxxz B1uU  fyyz B2uU  fxxy B2uU  fyyy B3uU  fxxx B3uU  fxyy Au U  fxyz
a  0.4763   0.2082     0.2082     0.0000     0.0000     0.0000     0.0000     0.0566
b  0.5237   0.0000     0.0000     0.1891     0.0716     0.0716     0.1891     0.0000

 9:  0.3876430862045  (f delta)
   Total    B1uU  fxxz B1uU  fyyz B2uU  fxxy B2uU  fyyy B3uU  fxxxB3uU  fxyy Au U  fxyz
a  0.5427   0.1582     0.1582     0.0000     0.0000     0.0000     0.0000     0.2226
b  0.4573   0.0000     0.0000     0.1724     0.0500     0.0500     0.1724     0.0000

10:  0.3925403714328  (f sigma)
   Total    B1uU  pz   B1uU  fxxz B1uU  fyyz B1uU  fzzz B1uO1 s    B1uO1 pz   B1uO1 dxx  B1uO1 dyy  B1uO1 dzz B2uU  py
a  0.9008  -0.0342    -0.2622    -0.2622    -0.1669     0.1111     1.6233    -0.0209    -0.0209    -0.0666    0.0000
b  0.0992   0.0000     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000    0.0409

            B2uU  fyyy B2uU  fyzz B3uU  px   B3uU  fxxx B3uU  fxzz
            0.0000     0.0000     0.0000     0.0000     0.0000
           -0.0108     0.0109     0.0409    -0.0108     0.0109

11:  0.3981739494585  (f phi)
   Total    B1uU  pz   B1uU  fxxz B1uU  fyyz B1uO1 pz   B2uU  py   B2uU  fxxy B2uU  fyyy B2uO1 py   B3uU  px L B3uU  fxxx
a  0.0470  -0.0113    -0.0122    -0.0122     0.0901     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000
b  0.9530   0.0000     0.0000     0.0000     0.0000     0.0143     0.3597     0.0806     0.0113     0.0143     0.0806

            B3uU  fxyy B3uO1 px
            0.0000     0.0000
            0.3597     0.0113

12:  0.4056360289236  (f phi)
   Total    B1uU  pz   B1uO1 s    B1uO1 pz   B2uU  py   B2uU  fxxy B2uU  fyyy B2uU  fyzz B2uO1 py   B3uU  px L B3uU  fxxx
a  0.0628  -0.0407     0.0136     0.1158     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000
b  0.9372   0.0000     0.0000     0.0000     0.0123    -0.0134     0.0373    -0.0105     0.4454     0.0123     0.0373

            B3uU  fxyy B3uU  fxzz   B3uO1 px
            0.0000     0.0000       0.0000
           -0.0134    -0.0105       0.4454
```

```
--------------------------------------------------------------------------------
uranyl embedded in chorides (model c'), WFT-in-DFT (Hartree-Fock) spinors, C2h symmetry
--------------------------------------------------------------------------------

occupied
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 4: -0.6783078719105     (sigma)
     Total     Bu U  py   Bu U  pz   Bu U  fxxz Bu U  fyyz Bu U  fzzz Bu O1 s    Bu O1 pz   Au U  px
a    0.1015    0.0443     0.0002     0.0000     0.0000     0.0000     0.0001     0.0000     0.0460
b    0.8985    0.0002     0.3486     0.0279     0.0286    -0.0269     0.3839     0.1321     0.0000

 5: -0.2785841248702     (pi)
     Total     Bu U  pz   Bu U  fxxz Bu U  fyyz Bu U  fyzz Bu U  fzzz Bu O1 py   Bu O1 pz   Au U  fxzz Au O1 px
a    0.6655    0.0000     0.0000     0.0000     0.0431     0.0000     0.2780     0.0002     0.0419     0.2724
b    0.3345    0.0134     0.0598     0.0610     0.0001     0.0408     0.0002     0.1506     0.0000     0.0000

 7: -0.2499471161837     (sigma)
     Total     Bu U  py   Bu U  pz   Bu U  fxxz Bu U  fyyz Bu U  fyzz Bu U  fzzz Bu O1 py   Bu O1 pz   Au U  fxzz Au O1 px
a    0.3372    0.0052     0.0000     0.0000     0.0001     0.0136     0.0001     0.1357     0.0000     0.0147     0.1452
b    0.6628    0.0000     0.0632     0.1453     0.1486     0.0005     0.0748     0.0000     0.2237     0.0000     0.0000


virtuals
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 6:  0.3548293534542     (f phi)
     Total     Bu U  py   Bu U  pz   Bu U  fxxy Bu U  fxxz Bu U  fyyy Bu U  fyyz Bu U  fyzz Bu O1 py   Au U  px   Au U  fxxx
a    0.8549    0.2040    -0.0001    -0.0054     0.0000    -0.0094     0.0000     0.0288    -0.0989     1.3112    -0.0500
b    0.1451    0.0001     0.0106     0.0000     0.0496     0.0000     0.0455     0.0000     0.0000     0.0000     0.0000

              Au U  fxyy Au U  fxyz Au U  fxzz Au O1 px
             -0.0142     0.0000     0.1589    -0.6433
              0.0000     0.0440     0.0000     0.0000

 7:  0.3625555445221     (f delta)
     Total     Bu U  py   Bu U  fxxz Bu U  fyyz Bu O1 py   Au U  px   Au U  fxxx Au U  fxyy Au U  fxyz Au U  fxzz Au O1 px
a    0.2274    0.2033     0.0001     0.0001    -0.1132     0.2722    -0.0265     0.0033     0.0000     0.0243    -0.1524
b    0.7726    0.0002     0.2930     0.2973    -0.0001     0.0000     0.0000     0.0000     0.1769     0.0001     0.0000

 8:  0.3703815860768     (f phi)
     Total     Bu U  fxxy Bu U  fxxz Bu U  fyyy Bu U  fyyz Au U  fxxx Au U  fxyy Au U  fxyz
a    0.5244    0.2018     0.0001     0.0569     0.0001     0.0593     0.1999     0.0030
b    0.4756    0.0013     0.0278     0.0005     0.0284     0.0006     0.0008     0.4128

 9:  0.3874449321876     (f delta)
     Total     Bu U  fxxy Bu U  fxxz Bu U  fyyy Bu U  fyyz Au U  fxxx Au U  fxyy Au U  fxyz
a    0.4569    0.1629     0.0001     0.0604     0.0002     0.0604     0.1604     0.0000
b    0.5431    0.0001     0.1086     0.0001     0.1129     0.0000     0.0000     0.3160

10:  0.3924998627149     (f sigma)
     Total     Bu U  pz   Bu U  fxxz Bu U  fyyz Bu U  fzzz Bu O1 s   Bu O1 pz   Au U  px   Au U  fxzz
a    0.1029    0.0002    -0.0003    -0.0004    -0.0002     0.0001     0.0015     0.0429     0.0112
b    0.8971   -0.0335    -0.2566    -0.2639    -0.1657     0.1106     1.6145     0.0000     0.0000

11:  0.3979825488601     (f phi)
     Total     Bu U  py   Bu U  pz   Bu U  fxxy Bu U  fxxz Bu U  fyyy Bu U  fyyz Bu O1 py   Bu O1 pz  Au U  px   Au U  fxxx
a    0.9512    0.0182    -0.0001     0.2967     0.0000     0.1338     0.0001     0.0127     0.0001    0.0103     0.1369
b    0.0488    0.0000    -0.0115     0.0000    -0.0127     0.0000    -0.0127     0.0000     0.0931    0.0000     0.0000

              Au U  fxyy
              0.3131
              0.0000

12:  0.4054929413480     (f phi)
     Total     Bu U  py   Bu U  pz   Bu U  fxxy Bu U  fyyy Bu U  fyzz Bu O1 s    Bu O1 py   Bu O1 pz   Au U  px   Au U  fxxx
a    0.9361   -0.0108    -0.0002     0.0789    -0.0478    -0.0212     0.0000     0.5316     0.0000     0.0347    -0.0441
b    0.0639   -0.0001    -0.0411     0.0000     0.0000     0.0000     0.0138     0.0001     0.1174     0.0000     0.0000

              Au O1 px
              0.3588
              0.0000
```

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--------------------------------------------------------------------------------
uranyl embedded in crystal (model f), WFT-in-DFT (Hartree-Fock) spinors, C2h symmetry
--------------------------------------------------------------------------------

occupied
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 4: -0.8645808346533    (sigma)
     Total    Bu U  py   Bu U  pz   Bu U  fxxz Bu U  fyyz Bu U  fzzz Bu O1 s    Bu O1 pz   Au U  px
a    0.1003   0.0441     0.0002     0.0000     0.0000     0.0000     0.0001     0.0000     0.0452
b    0.8997   0.0002     0.3539     0.0288     0.0289    -0.0251     0.3777     0.1317     0.0000

 5: -0.4643740574441    (pi)
     Total    Bu U  pz   Bu U  fxxz Bu U  fyyz Bu U  fyzz Bu U  fzzz Bu O1 py   Bu O1 pz   Au U  fxzz Au O1 px
a    0.6486   0.0000     0.0000     0.0000     0.0400     0.0000     0.2450     0.0002     0.0463     0.2870
b    0.3514   0.0189     0.0632     0.0634     0.0001     0.0436     0.0004     0.1593     0.0000     0.0000

 7: -0.4362900466429   (sigma)
     Total    Bu U  pz   Bu U  fxxz Bu U  fyyz Bu U  fyzz Bu U  fzzz Bu O1 py   Bu O1 pz  Au O1 px
a    0.3564   0.0001     0.0001     0.0002     0.0184     0.0001     0.1685     0.0000    0.1265
b    0.6436   0.0703     0.1413     0.1413     0.0008     0.0737     0.0002     0.2202    0.0000


virtuals
--------

 6:  0.1613277193544    (f sigma)
     Total    Bu U  py   Bu U  pz   Bu U  fxxy Bu U  fxxz Bu U  fyyy Bu U  fyyz Bu U  fyzz Bu U  fzzz Bu O1 s    Bu O1 py
a    0.9866   0.8734     3.6978    -0.0460    -0.4621    -0.0759    -0.3792     0.0935     0.5160    -2.9704    -0.4339
b    0.0134   0.0040     0.0049    -0.0002     0.0033     0.0002     0.0029     0.0012     0.0004    -0.0053    -0.0016

              Bu O1 pz  Bu O1 dxx  Bu O1 dyy  Au U  px   Au O1 px
              0.1572   -0.0154    -0.0124     0.0587    -0.0292
              0.0015    0.0000     0.0001     0.0000     0.0000

 7:  0.1707077642101    (f delta)
     Total    Bu U  py   Bu U  pz   Bu U  fxxy Bu U  fxxz Bu U  fyyy Bu U  fyyz Bu U  fyzz Bu U  fzzz Bu O1 s    Bu O1 py
a    0.1609   0.0229     0.0414     0.0001     0.0443    -0.0012     0.0386     0.0032     0.0044    -0.0371    -0.0094
b    0.8391   0.5152     0.9802    -0.0157    -0.0568     0.0118     0.0591     0.0493     0.1573    -0.5765    -0.2643

              Bu O1 pz L Bu O1 dxx Au U  px   Au U  fxyz Au O1 px
              0.0047     0.0000    0.0289     0.0283    -0.0145
             -0.0540    -0.0212    0.0006     0.0472     0.0005

 8:  0.1776401699581    (f phi)
     Total    Bu U  py   Bu U  pz   Bu U  fxxy Bu U  fxxz Bu U  fyyy Bu U  fyyz Bu U  fyzz Bu U  fzzz Bu O1 s    Bu O1 py
a    0.3735   0.3754     0.5646    -0.0109    -0.0705    -0.0197    -0.0687     0.0179     0.0871    -0.3362    -0.2044
b    0.6265   0.1277     0.6102     0.0006     0.0634     0.0070     0.0084     0.0083     0.1009    -0.3537    -0.0571

              Bu O1 pz L Au U  px  Au U  fxyz Au O1 px
             -0.0266     0.0349    0.0389    -0.0130
             -0.0387     0.0002    0.1534     0.0006

 9:  0.1832445278344    (f phi)
     Total    Bu U  pz   Bu U  fxxy Bu U  fxxz Bu U  fyyy Bu U  fyyz Bu O1 s    Au U  fxxx Au U  fxyy Au U  fxyz
a    0.5495   0.0016     0.2122    -0.0002     0.0558    -0.0002    -0.0010     0.0582     0.2111     0.0057
b    0.4505   0.0489     0.0016     0.0336     0.0015     0.0248    -0.0291     0.0003     0.0034     0.3521

10:  0.2003995454197    (f delta)
     Total    Bu U  pz   Bu U  fxxy Bu U  fxxz Bu U  fyyy Bu U  fyyz Au U  fxxx Au U  fxyy Au U  fxyz
a    0.4395   0.0005     0.1565     0.0002     0.0555     0.0000     0.0559     0.1527     0.0018
b    0.5605   0.0322     0.0007     0.1147     0.0009     0.0990     0.0001     0.0019     0.3216

11:  0.2103894057439    (f phi)
     Total    Bu U  py   Bu U  fxxy Bu U  fyyy Bu U  fyzz Au U  px   Au U  fxxx Au U  fxyy Au O1 px
a    0.9965   0.0301     0.2933     0.1373     0.0161     0.0288     0.1453     0.2815     0.0402
b    0.0035   0.0000     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000     0.0000

12:  0.2167306363990    (f phi)
     Total    Bu U  py   Bu U  pz   Bu U  fxxy Bu U  fyyy Bu U  fyzz Bu O1 py   Au U  px   Au U  fxxx Au U  fxyy Au U  fxzz
a    0.9742  -0.0215     0.0033     0.0810    -0.0156     0.0547     0.1842    -0.1922    -0.0271     0.1193    -0.0285
b    0.0258  -0.0002     0.0195     0.0002     0.0000    -0.0001     0.0003     0.0000     0.0000     0.0001     0.0000
```

Table 6 IHFSCCSD/Q_2 excited state composition in terms of the most significant determinants (in %) in the model space (P_m) for model (**b**).

		orbital pairs (left: occupied, right: virtual) defining the excited determiants.														
E(cm^{-1})	State	(4, 4)	(4, 5)	(4, 6)	(4, 7)	(4, 8)	(5, 4)	(5, 5)	(5, 6)	(5, 7)	(5, 8)	(7, 4)	(7, 5)	(7, 6)	(7, 7)	(7, 8)
14757	I,II	5.2	0.2				24.9	1.2				62.9	2.8			
15132	III	3.0	2.6				16.7	8.9	0.2			39.8	26.8	0.2		
15421	IV	0.1	5.7				1.8	23.9	0.2			2.8	64.3	0.2		
16620	V,VI	0.2	4.9	0.1	0.4		1.0	22.9	0.8	0.7	0.1	2.8	61.0	1.2	2.8	0.2
17600	VII	1.5	2.3	1.7			5.9	10.6	7.4			21.2	31.3	16.9		
17687	VIII	3.2	0.1	2.1			13.8	0.9	8.8			46.1	2.0	21.1		
21061	IX,X			5.2	0.3			0.1	22.5	2.7	0.2		0.4	62.1	4.4	0.3
22829	XI				5.2	0.4				23.3	1.8				61.5	4.8
22829	XII				5.2	0.4				23.3	1.8				61.4	4.8

Table 7 IHFSCCSD and IHFSCC-112 excited state composition in terms of the most significant determinants (in %) in the model space (P_m) for model (**c**), for different correlation spaces (Q).

		orbital pairs (left: occupied, right: virtual) defining the excited determiants.																	
E(cm^{-1})	State	(4, 7)	(4, 8)	(4, 9)	(4, 10)	(4, 11)	(5, 6)	(5, 7)	(5, 8)	(5, 9)	(5, 10)	(5, 11)	(7, 6)	(7, 7)	(7, 8)	(7, 9)	(7, 10)	(7, 11)	(7, 12)
IHFSCCSD/Q_3																			
15680	I,II	4.2	0.2				3.1	12.8	0.5				14.1	58.8	2.2				
16365	III	3.1	1.6				2.4	10.6	3.1	0.3			10.8	47.2	17.7	0.2			
17043	IV	0.4	5.0				0.7	2.2	13.1	0.3			2.6	8.2	64.7	0.6			
18318	V,VI		4.9			0.3		0.4	14.9	0.5		0.2	0.2	2.0	71.0	1.1		2.2	0.2
19323	VII	2.0	0.5	2.4			1.3	4.7	1.7	6.0			9.7	33.1	7.9	28.1			
19370	VIII	0.7	2.9	1.9			0.2	1.3	7.5	5.1			2.5	11.0	42.8	22.1			
22746	IX,X			5.1		0.4				13.4		1.9			0.4	69.7		5.5	0.4
25155	XI					5.0					0.4	14.4					1.9	69.1	4.2
25157	XII					5.0					0.4	14.4					1.9	69.0	4.3
IHFSCCSD/Q_2																			
15746	I,II	4.2	0.2				3.1	12.8	0.5				14.1	58.8	2.1				
16432	III	3.1	1.6				2.4	10.6	3.1	0.3			10.8	47.3	17.5	0.3			
17116	IV	0.4	5.0				0.7	2.3	13.1	0.3			2.6	8.3	64.5	0.6			
18389	V,VI		4.9			0.3		0.4	14.9	0.5		0.2	0.2	1.9	70.9	1.2		2.2	0.2
19400	VII	2.0	0.5	2.3			1.3	4.6	1.7	6.0			9.6	32.9	8.1	28.1			
19448	VIII	0.7	2.9	1.9			0.2	1.2	7.6	5.2			2.4	10.8	43.0	22.1			
22805	IX,X			5.1		0.4				13.4		1.9			0.4	69.6		5.6	0.4
25218	XI					5.0					0.4	14.4					1.9	69.1	4.2
25220	XII					5.0					0.4	14.4					1.9	69.0	4.3
IHFSCCSD/Q_1																			
16896	I,II	4.0					3.2	13.2	0.5	0.2			14.1	58.7	2.0				
17624	III	3.1	1.3				2.5	11.2	2.7	0.5			11.2	48.9	14.8	0.6			
18400	IV	0.5	4.6				0.8	2.7	12.6	0.6			3.3	10.4	60.6	1.5			
19696	V,VI		4.7			0.3		0.4	15.2	0.8		0.2	0.2	1.8	69.8	2.3		1.9	0.2
20834	VII	1.8	0.7	2.2			1.3	4.4	2.5	5.9			9.0	30.6	11.6	27.3			
20915	VIII	0.6	3.0	1.8			0.2	0.9	8.3	5.2			1.9	8.4	45.8	21.8			
24108	IX,X			4.8		0.4			0.2	13.5		2.0			1.1	68.2	0.2	5.9	0.4
26626	XI					4.8					0.4	14.8					1.9	69.0	3.9
26628	XII					4.8					0.4	14.8					1.9	68.8	4.0
IHFSCC-112/Q_1																			
17998	I,II	4.1	0.2				3.1	12.8	0.5	0.2			14.2	58.7	2.2				
18705	III	3.1	1.5				2.4	10.6	2.9	0.4			11.2	48.1	16.4	0.4			
19409	IV	0.5	4.8				0.7	2.4	12.7	0.4			3.0	9.2	62.9	1.0			
20689	V,VI		4.8			0.3		0.4	14.9	0.6		0.2	0.2	2.0	70.8	1.6		2.0	0.2
21797	VII	1.9	0.6	2.3			1.3	4.6	2.1	5.8			9.5	31.8	1	27.3			
21855	VIII	0.6	2.9	1.8			0.2	1.1	7.9	5.0			2.2	9.6	44.7	21.6			
25131	IX,X			5.0		0.3				13.5		1.8			0.7	69.5		5.2	0.4
27602	XI					4.9					0.4	14.5					1.9	69.1	4.0
27603	XII					4.9					0.4	14.5					1.9	69.0	4.2

Table 8 IHFSCC-112/Q_1 excited state composition in terms of the most significant determinants (in %) in the model space (P_m) for models (**c'**) and (**f**).

		orbital pairs (left: occupied, right: virtual) defining the excited determiants.																
		(4, 7)	(4, 8)	(4, 9)	(4, 10)	(4, 11)	(5, 6)	(5, 7)	(5, 8)	(5, 9)	(5, 10)	(5, 11)	(7, 6)	(7, 7)	(7, 8)	(7, 9)	(7, 10)	(7, 11)	(7, 12)
model (**c'**)																			
18151	I	4.0	0.2				3.1	12.6	0.5				13.7	57.7	2.3				
18154	II	4.0	0.2				3.1	12.6	0.5				14.1	57.6	2.2				
18874	III	3.1	1.5				2.3	10.5	3.0	0.4			10.6	47.2	16.5	0.4			
19552	VI	0.5	4.8				0.7	2.3	12.7	0.4			3.1	8.9	63.0	1.0			
20836	V		4.8			0.3		0.5	14.9	0.5		0.2	0.2	2.1	70.6	1.5		1.9	0.2
20843	VI		4.8			0.3		0.5	14.9	0.5		0.2	0.2	2.1	70.6	1.5		2.0	0.2
21944	VII	1.9	0.6	2.3			1.2	4.5	2.1	5.8			8.8	31.4	1	27.3			
22005	VIII	0.6	2.9	1.8			0.3	1.2	7.9	5.0			2.5	9.6	44.5	21.5			
25307	IX			5.0		0.3				13.6		1.6			0.7	69.7	0.3	4.6	0.4
25297	X			5.0		0.4				13.6		1.9			0.7	69.4		5.6	0.2
27779	XI					4.9					0.4	14.5					2.0	69.1	3.9
27781	XII					4.9					0.4	14.6					2.0	69.0	4.0
model (**f**)																			
18128	I	2.3	2.7				0.4	7.9	8.8	0.4			1.6	33.5	36.6	1.0			
18124	II	2.3	2.7				0.4	7.9	8.8	0.4			1.5	32.9	36.9	1.1			
18816	III	2.0	1.4	1.8			0.4	7.1	5.4	4.1	0.2		1.6	29.8	21.9	19.6	0.4		
19492	VI	0.2	0.7	4.6				1.0	3.1	12.7	0.5	0.2		3.7	11.8	58.1	1.0		
20760	V			4.8		0.3		0.5		15.9	0.6	0.3	0.2	2.0		69.1	1.5	2.1	0.2
20768	VI			4.8		0.3		0.5		15.8	0.6	0.3	0.2	2.3		68.8	1.5	1.9	0.3
21848	VII	1.2	1.1	0.7	2.2		0.2	3.1	2.7	2.5	6.2		1.4	19.5	17.4	11.5	26.4		
21905	VIII	0.3	0.7	2.8	1.8			0.7	1.5	8.1	5.2			5.2	1	41.1	20.3		
25201	IX				5.0	0.3				0.2	14.4	2.0		0.4	0.3	0.8	67.5	5.1	0.3
25185	X				5.0	0.4				0.2	14.4	1.9		0.3	0.2	0.7	67.4	5.1	0.7
27634	XI					5.0						15.5						67.8	5.4
27637	XII					5.0						15.6						67.6	5.6