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# Atomic properties of actinide ions with particle-hole configurations

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We study the effects of higher-order electronic correlations in the systems with particle-hole excited states using a relativistic hybrid method that combines configuration interaction and linearized coupled-cluster approaches. We find the configuration interaction part of the calculation sufficiently complete for eight electrons while maintaining good quality of the effective coupled-cluster potential for the core. Excellent agreement with experiment was demonstrated for a test case of  $\text{La}^{3+}$ . We apply our method for homologue actinide ions  $\text{Th}^{4+}$  and  $\text{U}^{6+}$  which are of experimental interest due to a puzzle associated with the resonant excitation Stark ionization spectroscopy (RESIS) method. These ions are also of interest to actinide chemistry and this is the first precision calculation of their atomic properties.

## I. INTRODUCTION

Last two decades of progress in atomic, molecular, and optical (AMO) physics brought forth a plethora of new AMO applications, ranging from quantum information [1] to dark matter searches [2–4]. These advances required further development of high-precision theory, first for alkali-metal and alkaline earth metal atoms, and then for more complicated systems with larger number of valence electrons. A hybrid method which combines configuration interaction and linearized coupled-cluster approaches was developed for these purposes [5–7] and applied for a wide range of problems in ultracold atoms [8–10] and search for physics beyond the Standard Model of particles and interactions [11–13]. This method has been tested and demonstrated to give accurate results up to four valence electrons [14, 15]. Recently, there was much interest in application of atoms and ions with even more complicated atomic structure, including lanthanides, actinides, various highly charged ions and negative ions [12, 16–18]. In particular, the ability to treat hole-particle states with good precisions is needed [19, 20]. The problem of applying CI+all-order method to predict properties of more complicated systems lies in the exponential scaling of the number of possible configurations with the number of valence electrons. However, if the most important sets of configuration are identified this method may still yield accurate values for larger number of electrons. In this work, we demonstrate a first accurate calculation of the systems with 8 valence electrons using the all-order effective Hamiltonian combined with a large-scale configuration interaction calculation in a valence sector.

We demonstrate the methodology on the example of  $\text{Th}^{4+}$  and  $\text{U}^{6+}$ . These ions are of particular interest to actinide chemistry, as U and Th usually occur in chemical compounds and solutions as multiply-charged cations, most commonly near the Rn-like ion with a closed shell

configuration [21]. No spectroscopy data exists for excited levels of the Rn-like U and Th ions, i.e. there is no experimental data for any of the energy levels. A very successful program was established to measure the dipole and quadrupole polarizabilities of Th ions with resonant excitation Stark ionization spectroscopy (RESIS) method [22–26]. In the RESIS method, a non-penetrating Rydberg electron is attached to the ion of interest to measure the binding energies of the resulting high-L Rydberg states [22]. The energy levels in the fine structure pattern are determined by the properties of the core ion, mainly by its dipole and quadrupole polarizabilities. Therefore, these properties can be extracted from the Rydberg high-L energy measurements. This method was successful for  $\text{Th}^{4+}$  and  $\text{Th}^{3+}$ , but failed completely for the  $\text{U}^{6+}$  ions - no resolved spectral features were observed [27, 28]. This is particularly puzzling since  $\text{Th}^{4+}$  and  $\text{U}^{6+}$  were predicted to have very similar energy levels structures [29]. However, the theory calculations were not of sufficient precision to definitively establish the order of the first two excited levels. Differences in the properties of the low-lying metastable states of  $\text{Th}^{4+}$  and  $\text{U}^{6+}$  may provide an explanation for the failure of the RESIS experiments in  $\text{U}^{6+}$  [27, 28]. In summary, reliable precision calculations are needed to resolve this puzzle.

The electronic configuration of the  $\text{Th}^{4+}$  and  $\text{U}^{6+}$  excited states makes accurate calculations difficult: the ground state configuration is a Rn-like closed shell system  $[\text{Hg}]6p^6$ , while the first two excited states have a hole in the  $6p$  shell, resulting in the  $6p^55f$  configuration. Since both of these configurations are of even parity, they have to be included in the calculations on the same footing, i.e. including the mixing of these configurations. In this work, we separate the treatments of the electronic correlations into two problems: (1) treatment of strong valence-valence correlations and (2) inclusion of core excitations from the entire core. We test the predictive ability of our method on the homologue case of Xe-like

TABLE I. Energies of Xe-like lanthanum,  $\text{La}^{3+}$ . The CI+all-order results obtained considering  $5s^2$  to be a core shell are listed in rows labeled “6-el”. The CI+all-order results obtained considering  $5s^2$  to be valence electrons are listed in rows labeled “8-el”. Results obtained with small, medium and large sets of configurations are given in the corresponding rows. The results are compared with experimental data compiled in NIST database [30].

Level	COWAN	6el-small	6el-large	8el-small	8el-medium	8el-large	NIST
$5p^6\ ^1S_0$	0	0	0	0	0	0	0
$5p^54f\ ^3D_1$	150023	138453	137939	143780	142476	142168	143354.7
$5p^54f\ ^3D_2$	152843	141183	140609	146591	145219	144910	145949.0
$5p^54f\ ^3G_5$	155822	144530	143879	150541	149016	148620	
$5p^54f\ ^3G_4$	156371	145174	144421	151285	149622	149210	
$5p^54f\ ^3D_3$	157255	145363	144693	150949	149464	149153	149927.1
$5p^54f\ ^3G_3$	161005	149159	148330	155332	153519	153130	153339.1
$5p^54f\ ^3F_4$	164624	153238	152229	159590	157645	157252	
$5p^54f\ ^3F_2$	168059	157135	155922	163326	161024	160592	160486.4
$5p^54f\ ^3G_3$	177237	165352	164376	171959	169935	169562	
$5p^54f\ ^3F_3$	180229	168297	167235	174850	172780	172445	

$\text{La}^{3+}$  where the energies have been measured to high precision.

## II. METHOD

We use a hybrid approach developed in [5–7] that efficiently treats these two problems by combining configuration interaction (CI) and a linearized coupled-cluster methods, referred to as the CI+all-order method. The first problem is treated by a large-scale CI method in the valence space. The many-electron wave function is obtained as a linear combination of all distinct many-electron states of a given angular momentum  $J$  and parity:

$$\Psi_J = \sum_i c_i \Phi_i. \quad (1)$$

Usually, the energies and wave functions of the low-lying states are determined by diagonalizing the Hamiltonian in the CI method:

$$H = H_1 + H_2, \quad (2)$$

where  $H_1$  is the one-body part of the Hamiltonian, and  $H_2$  represents the two-body part, which contains Coulomb + Breit matrix elements. In the CI+all-order approach this bare Hamiltonian is replaced by the effective one,

$$H_1 \rightarrow H_1 + \Sigma_1, \quad (3)$$

$$H_2 \rightarrow H_2 + \Sigma_2, \quad (4)$$

where  $\Sigma_i$  corrections incorporate all single and double excitations from *all* core shells to all basis set orbitals (up to  $n_{\text{max}} = 35$  and  $l_{\text{max}} = 5$ , efficiently solving the second problem. The effective Hamiltonian  $H^{\text{eff}}$  is constructed

using a coupled cluster method [31]. The size of the core rather weakly affect the accuracy of the CI+all-order approach for  $Z \gtrsim 20$  and the method was used even for superheavy atoms with  $Z > 100$ .

## III. METHOD TESTS - $\text{La}^{3+}$ CALCULATION

To test the method, we carried out the calculation for a homologue system,  $\text{La}^{3+}$ , which has  $[\text{Pd}]5s^25p^6$  ground state and  $5s^25p^54f$  low-lying configurations. The experimental values for relevant  $\text{La}^{3+}$  states are available [30] for benchmark comparisons. We started with the assumption that  $5s^2$  shell may be kept closed. In this calculation, we used a  $V^{N-6}$  Dirac-Hartree-Fock (DHF) starting potential [32], where  $N$  is the number of electron, i.e. the potential of Cd-like ionic core of  $\text{La}^{9+}$ . Such calculation yielded poor results for the excited states of interest. Further tests showed that the  $5s6s5p^54f$  configuration gives the largest contribution to the low-lying states after the  $5s^25p^6$  and  $5s^25p^54f$  configurations. The next largest contributions come from the  $5s^25p^55f$  and  $5s^25p^56p$  configurations, as expected. Therefore, the  $\text{La}^{3+}$  calculations have to be carried out as a 8-valence electron computation, with both  $5s$  and  $5p$  shells open. We used a  $V^{N-8}$  starting potential, i.e. the potential of the  $\text{La}^{11+}$  ionic core. To construct the set of the most important even parity configurations, we started ionic core with the  $5s^25p^6$  and  $5s^25p^54f$  configurations and allowed to excite one or two electrons from these configurations to excited states up to  $7f$ . This produced the list of 3277 (relativistic) configurations resulting in 360 633 Slater determinants. Below we refer to this run as “small”. For the next run, we reordered the original set of configurations by their weight and allow further one-two excitations up to  $7f$  electrons from the 21 configurations with highest weights. This (medium) set has

TABLE II. Energies of the  $\text{Th}^{4+}$  and  $\text{U}^{6+}$  even states calculated using the CI+all-order method. The results obtained considering  $6s^2$  to be valence electrons are listed in rows labelled “8-el”. Results obtained with small and large sets of configuration are given in the corresponding rows.

$\text{Th}^{4+}$				$\text{U}^{6+}$			
Level	COWAN	8el-small	8el-large	Level	COWAN	8el-small	8el-large
$6p^6\ ^1S_0$	0	0	0	$6p^6\ ^1S_0$	0	0	0
$6p^55f\ ^3D_1$	135013	137121	134995	$6p^55f\ ^3D_1$	87975	92458	90850
$6p^55f\ ^3D_2$	140469	142088	139842	$6p^55f\ ^3D_2$	94775	98554	96863
$6p^55f\ ^3G_4$	143819	145579	143160	$6p^55f\ ^3G_4$	97064	101288	99539
$6p^55f\ ^3G_5$	145606	147001	144714	$6p^55f\ ^3F_3$	102529	105492	103627
$6p^55f\ ^3F_3$	147698	148752	146314	$6p^55f\ ^3G_5$	101946	105732	104079
$6p^55f\ ^1F_3$	150769	150615	148081	$6p^55f\ ^1F_3$	107300	109051	107240
$6p^55f\ ^3F_4$	156377	157221	154552	$6p^55f\ ^3F_4$	113656	116417	114499
$6p^55f\ ^1D_2$	160980	162300	159248	$6p^55f\ ^1D_2$	116277	119989	117725
$6p^55f\ ^3G_3$	209865	208535	205597	$6p^55f\ ^3G_3$	188185	186470	183699
$6p^55f\ ^3D_3$	215174	213424	210417	$6p^55f\ ^3D_3$	196853	193827	190953
$6p^55f\ ^3G_4$	217527	217054	214015	$6p^55f\ ^3F_2$	196653	197749	193197

11785 configurations and 3 453 220 determinants, making it ten times larger than the small run. Note that it is the number of Slater determinants that defines the computational time. Finally, we also allow a single excitation from the 59 highest weight configurations to all electrons up *20spd16f12g*. This (large) run has 18187 configurations and 4 187 914 determinants.

The results for the energies of even states of xenon-like lanthanum ( $\text{La}^{3+}$ ) are summarized in Table I. The CI+all-order results obtained considering  $5s^2$  to be a core shell are listed in rows labeled “6-el”. The CI+all-order results obtained considering  $5s^2$  to be valence electrons are listed in rows labeled “8-el”. Results obtained with small, medium and large sets of configuration are given in the corresponding rows. The results are compared with experimental data from NIST database [30]. The COWAN code [33] data are given for reference. The table clearly demonstrates problems of the 6-el approach. The differences between medium and large runs all relatively small. The results of the small run are larger than the experimental values and the results of both larger runs are smaller than the experimental values. Therefore, the accuracy of the inclusion of the core-valence correlations via the effective Hamiltonian is comparable with the contribution of the remaining configurations. Since the inclusion of further configurations can only lower the values, inclusion of the further configuration will not improve the accuracy of the theory.

#### IV. $\text{Th}^{4+}$ AND $\text{U}^{6+}$ CI+ALL-ORDER CALCULATIONS

We use the results of  $\text{La}^{3+}$  tests to construct the  $\text{Th}^{4+}$  and  $\text{U}^{6+}$  configuration sets for 8 valence electrons with about 3 800 000 determinants. We used a  $V^{N-8}$  start-

ing potentials, i.e. the potentials of the  $\text{Th}^{12+}$  and  $\text{U}^{14+}$  ionic cores. The resulting energies are given in Table II. Results obtained with small and large sets of configurations are given in the corresponding rows. The small set is equivalent to the  $\text{La}^{3+}$  small set. The results for the reduced  $M1$  (in  $\mu_0$ ) and  $E2$  (in  $ea_0^2$ ) matrix elements between first three states are given in Table III. Corresponding transition energies in  $\text{cm}^{-1}$ , transition rates in  $\text{s}^{-1}$ , branching ratios, and radiative lifetimes of the  $6p^55f\ ^3D_{1,2}$  states in seconds are also listed. The transition rates (in  $\text{s}^{-1}$ ) are obtained as

$$A(M1) = \frac{2.69735 \times 10^{13}}{(2J+1)\lambda^3} \left( \frac{\langle M1 \rangle}{\mu_0} \right)^2, \quad (5)$$

$$A(E2) = \frac{1.11995 \times 10^{18}}{(2J+1)\lambda^5} \left( \frac{\langle E2 \rangle}{ea_0^2} \right)^2, \quad (6)$$

where  $\langle M1 \rangle$  and  $\langle E2 \rangle$  are reduced matrix elements of the magnetic dipole and electric quadrupole operators,  $\lambda$  is the transition wavelength in Å, and  $J$  is the total angular momentum of the upper state. The branching ratios are the ratios of the rate of the given transition to the total rate, and the lifetime is the inverse of the total transition rate.

All of the values are obtained from the “large” runs. We found only a weak dependence of the matrix elements on the size of the configuration space with the exception of the  $6p^55f\ ^3D_2 - 6p^55f\ ^3D_1$   $E2$  matrix element in  $\text{U}^{6+}$ . The values of this  $E2$  matrix element in  $\text{Th}^{4+}$  and  $\text{U}^{6+}$  calculated with small, medium, and large number of configurations are listed in Table IV. The final results in Table III all include the random-phase approximation (RPA) correction to the  $M1$  and  $E2$  operators. In Table IV we listed the values without the RPA corrections as well. It is clear that while the value of this matrix element are similar for all runs in

TABLE III. Transition energies (in  $\text{cm}^{-1}$ ), matrix elements M1 (in  $\mu_0$ ) and E2 (in  $ea_0^2$ ), transition rates (in  $1/s$ ), and radiative lifetimes (in  $s$ ) of the  $6p^5 5f \ ^3D_{1,2}$  levels in  $\text{Th}^{4+}$  and  $\text{U}^{6+}$ .

Ion	Upper level	Lower level	Transition	Transition energy	Matrix element	Transition rate	Branching ratio	Lifetime
$\text{U}^{6+}$	$6p^5 5f \ ^3D_1$	$6s^2 \ ^1S_0$	M1	90850	5.8E-04	0.0022	1	450
$\text{Th}^{4+}$	$6p^5 5f \ ^3D_1$	$6s^2 \ ^1S_0$	M1	134994	5.6E-04	0.0070	1	140
$\text{U}^{6+}$	$6p^5 5f \ ^3D_2$	$6s^2 \ ^1S_0$	E2	96863	0.183	6.38	0.610	
	$6p^5 5f \ ^3D_2$	$6p^5 5f \ ^3D_1$	M1	6013	1.87	4.09	0.390	
	$6p^5 5f \ ^3D_2$	$6p^5 5f \ ^3D_1$	E2	6013	-0.039	2.7E-07	0.000	0.0955
$\text{Th}^{4+}$	$6p^5 5f \ ^3D_2$	$6s^2 \ ^1S_0$	E2	139842	0.479	274	0.992	
	$6p^5 5f \ ^3D_2$	$6p^5 5f \ ^3D_1$	M1	4848	1.913	2.3	0.008	
	$6p^5 5f \ ^3D_2$	$6p^5 5f \ ^3D_1$	E2	4848	-0.320	6.2E-06	0.000	0.00361

TABLE IV.  $E2 \ 6p^5 5f \ ^3D_2 - 6p^5 5f \ ^3D_1$  reduced matrix element in  $ea_0^2$ .

		$\text{Th}^{4+}$	$\text{U}^{6+}$
no RPA	Small	0.348	0.0399
RPA	Small	0.301	0.0026
RPA	Medium	0.303	0.0003
no RPA	Large		0.0389
RPA	Large	0.320	0.0034

$\text{Th}^{4+}$ , this is not the case in  $\text{U}^{6+}$ . This is explained as follows. The dominant one-electron contributions to the  $6p^5 5f \ ^3D_2 - 6p^5 5f \ ^3D_1$   $E2$  matrix element come from the  $6p_{3/2} - 6p_{3/2}$  and  $5f_{5/2} - 5f_{5/2}$ , and  $5f_{5/2} - 5f_{7/2}$  matrix elements. These contributions strongly cancel, leading to a small final value. In  $\text{Th}^{4+}$  there is noticeable addition from the configurations containing  $7p$  and  $6f$  orbitals, which are mixed with the  $6p^5 5f$  configuration.

In  $\text{U}^{6+}$ , the  $5f$  electron becomes stronger bound and closer to the ground configuration. This is due to a level crossing mechanism, which is responsible for the presence of optical transitions in highly charged ions [34]. As a result, the configuration mixing with higher orbitals, such as  $7p$  and  $6f$ , is suppressed. For example, the admixture of the  $6f$  orbital to the  $6p^5 5f$  configuration is almost two times larger in  $\text{Th}^{4+}$ , than in  $\text{U}^{6+}$ . This weakens the cancellation between  $np - np$  and  $nf - nf$  contributions. As a result, the  $E2$  matrix element  $6p^5 5f \ ^3D_2 - 6p^5 5f \ ^3D_1$  in  $\text{U}^{6+}$  is highly dependent on the details of the calculations, but is more stable for  $\text{Th}^{4+}$ . Because of that for uranium we can not predict this amplitude reliably. It is certainly significantly smaller than in  $\text{Th}^{4+}$ , most likely by an order of magnitude.

## V. SUMMARY OF THE DIFFERENCES BETWEEN THE $\text{Th}^{4+}$ AND $\text{U}^{6+}$ RESULTS FOR LOW-LYING LEVELS

Below, we outline the resulting differences between the low-lying metastable levels of the two ions.

- The  $6p^5 5f$  levels lie *closer* to the ground state in  $\text{U}^{6+}$  than in  $\text{Th}^{4+}$ . This is expected, as in hydrogenic ions the  $5f$  shell lies below the  $6p$  shell. Therefore, the level crossing must take place along the isoelectronic sequence.
- The lifetime of the first,  $^3D_1$ , excited state in  $\text{U}^{6+}$  is more than 3 times longer (450 s). This is purely due to smaller transition energy in  $\text{U}^{6+}$  as the M1 matrix element is practically the same. Note that M1 transition rates scale as  $\lambda^{-3}$ .
- The lifetime of the second,  $^3D_2$ , excited state in  $\text{U}^{6+}$  is 26 times longer. This is both due to  $\lambda^{-5}$  scaling of the  $E2$  transition rate and smaller  $E2$  matrix element, compared to  $\text{Th}^{4+}$ .
- The branching ratio of the  $^3D_2$  level to the ground state is over 99% in Th ion, but only 61% in U ion. As a result, the large fraction of the U ions from  $^3D_2$  state ends up in a highly metastable  $^3D_1$  level, but very few Th ions do.
- As described above, the  $^3D_2 - ^3D_1$   $E2$  matrix element is much smaller in U ion. We note that this  $E2$  transition is extremely weak in both cases and M1 decay is orders of magnitude stronger. The M1 matrix elements  $^3D_2 - ^3D_1$  are similar in both ions and M1 transition rate is factor of 2 larger in U ion.

## VI. CONCLUSION

We have established that the  $ns$  electrons have to be considered as valence for an accurate determination of the properties of particle-hole states with a hole in the respective  $np$  shell. We find that the CI+all-order method works well with the  $V^{N-8}$  starting potential which extends the applicability of this approach. We have developed an algorithm for the efficient construction of the large-scale CI configuration sets. The methodology is tested on the  $\text{La}^{3+}$  ion and excellent agreement with experiment is obtained. These results suggest that the un-

certainities of our predictions for the energy levels in  $\text{Th}^{4+}$  and  $\text{U}^{6+}$  ions are expected to be less than 1%. Matrix elements, branching ratios and lifetimes of the  $\text{Th}^{4+}$  and  $\text{U}^{6+}$  low-lying states were calculated and analyzed.

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