

Correlation and relativistic effects in actinide ions

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Wavelengths, line strengths, and transition rates are calculated for the multipole ($E1$, $M1$, $E2$, $M2$, $E3$, and $M3$) transitions between the excited $6s^2 6p^5 nl$ and $6s 6p^6 nl$ states and the ground $6s^2 6p^6$ state in Ac^{3+} , Th^{4+} , and U^{6+} Rn-like ions. Relativistic many-body perturbation theory (RMBPT), including the Breit interaction, is used to evaluate energies and transition rates for multipole transitions in these hole-particle systems. The RMBPT method agrees with multiconfigurational Dirac-Fock (MCDF) calculations in lowest order, includes all second-order correlation corrections, and includes corrections from negative-energy states. The calculations start from a $[\text{Xe}]4f^{14}5d^{10}6s^2 6p^6$ Dirac-Fock potential. First-order perturbation theory is used to obtain intermediate-coupling coefficients, and second-order RMBPT is used to determine the matrix elements. Evaluated multipole matrix elements for transitions from excited states to the ground states are used to determine the line strengths, transition rates, and multipole polarizabilities. This work provides a number of yet unmeasured properties of these actinide ions for various applications and for benchmark tests of theory and experiment.

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I. INTRODUCTION

Very few studies of actinide atom and ion properties have been carried out to date owing to difficulties in obtaining accurate theoretical descriptions of these systems and in experimental methodologies. The atomic properties of the actinides are also needed for the actinide chemistry models. The radon-like Th^{4+} ion is the most common charge state of thorium in chemical compounds [1]. This work is motivated in part by recent experiments [2–5] that investigated the spectroscopy of high- L Rydberg levels of actinide ions and the dipole and quadrupole polarizabilities. These measurements present an excellent opportunity for benchmark tests of *a priori* theoretical descriptions of such highly relativistic ions.

In 2010, binding energies of high- L Rydberg levels of Th^{3+} were measured using the resonant excitation stark ionization spectroscopy (RESIS) technique by Hanni *et al.* [3]. Analysis of the data with the long-range polarization model was used to determine the dipole and quadrupole polarizabilities of the free Th^{4+} ion, $\alpha_d = 7.61(6)$ a.u. and $\alpha_Q = 47(11)$ a.u. [3]. To improve the precision of these properties, rf spectroscopy was used to directly measure intervals separating $n = 37$ levels of Th^{3+} with $9 \leq L \leq 15$ in [4]. The resulting measurement improved the precision of the dipole polarizability by an order of magnitude but indicated that the previous report of the quadrupole polarizability was an overestimate [4]. The dipole polarizability of Th^{4+} was determined to be $\alpha_d = 7.720(7)$ a.u., and the quadrupole polarizability was found to be $\alpha_Q = 21.5(3.9)$ a.u. In 2011, properties of Fr-like Th^{3+} were determined from spectroscopy of high- L Rydberg levels of Th^{2+} ions [5].

The study of the high- L Rydberg levels of U^{5+} using the RESIS technique [2] led to difficulties which were possibly caused by different numbers of metastable states in Th^{4+} and U^{6+} ions. Theoretical calculations of U^{6+} atomic properties may aid further experimental studies.

The importance of metastable states for determination of polarizability in Yb^{2+} ions was underlined in Ref. [6]. All states up to $E = 53\,736\text{ cm}^{-1}$ (counting from the ground state) that belong to either $[\text{Xe}]4f^{13}5d$ or $[\text{Xe}]4f^{13}6s$ odd configurations were considered [6]. Only three of these $[\text{Xe}]4f^{13}5d$ levels have total angular momentum $J = 1$ and, therefore, can decay to the ground state via strong electric-dipole transitions. As a result, all remaining 21 $[\text{Xe}]4f^{13}5d$ and $[\text{Xe}]4f^{13}6s$ levels with $J = 0, 2-6$ are metastable.

Very few studies of Rn-like ions have been carried out to date. Only the total binding energies and ionization potentials in Rn-like ions were presented in Refs. [7–12]. Dirac-Fock total energies, ionization energies, and orbital energies for uranium ions were evaluated by Rashid *et al.* [9]. The total and ionization energies in Rn-like U^{6+} were found to be equal to 761 540.14 and 88.45 eV, respectively. An *ab initio* pseudopotential and density-functional all-electron study of ionization and excitation energies of actinide atoms was performed by Liu *et al.* [10]. The ionization potential of Rn-like Ac^{3+} ions was found to be equal to 43.78 eV. Recently, a systematic study of atomic binding energies in the Dirac-Fock (DF) approximation was presented by Rodrigues *et al.* [12]. The total binding energy (DF) of the isoelectronic series of radon with $Z = 89, 90$, and 92 was found to be $-699\,502$, $-719\,769$, and $-761\,482$ eV, respectively.

In the present paper, we determine energies of the excited $[\text{Xe}]4f^{14}5d^{10}6s^2 6p^5 nl$ and $[\text{Xe}]4f^{14}5d^{10}6s 6p^6 n'l'$ states in Ac^{3+} , Th^{4+} , and U^{6+} ions with the nuclear charges $Z = 89, 90$, and 92. We omit $[\text{Xe}]4f^{14}5d^{10}$ from the level designation below for brevity. The calculations are carried out to second order in relativistic many-body perturbation theory (RMBPT). The RMBPT method is also used to determine line strengths and transition rates for all allowed and forbidden electric-multipole and magnetic-multipole ($E1$, $E2$, $E3$, $M1$, $M2$) transitions from the $6s^2 6p^5 nl$ and $6s 6p^6 n'l'$ excited states into

the ground state. These data are used to determine the ground state multipole polarizabilities for Rn-like ions with $Z = 89$, 90, and 92.

The second-order RMBPT calculations for Rn-like ions start from a $[\text{Xe}]4f^{14}5d^{10}6s^26p^6$ Dirac-Fock potential. We consider the $6s$ and $6p$ holes and the $5f$, $6d$, $7s$, and $7p$ particles leading to 30 even-parity $6s^26p^55f$, $6s^26p^57p$, $6s6p^66d$, and $6s6p^67s$ excited states and 25 odd-parity $6s^26p^56d$, $6s^26p^57s$, $6s6p^65f$, and $6s6p^67p$ excited states. In the present paper, we evaluate the corresponding energies, wavelengths, line strengths, and transition rates. We would like to underline that the RMBPT method is based on the relativistic many-body perturbation theory that agrees with MCDF calculations in lowest order, includes all second-order correlation corrections, and includes corrections from negative-energy states.

II. METHOD

Details of the RMBPT method for hole-particle states were presented in Ref. [13] for calculation of energies, in Refs. [6,13–19] for calculation of multipole matrix elements for transitions from excited states to the ground state, and in Refs. [6,20–22] for calculation of multipole matrix elements for transitions between excited states. The calculations are carried out using a basis set of Dirac-Fock (DF) orbitals. The orbitals used in the present calculation are obtained as linear combinations of B splines. These B -spline basis orbitals are determined using the method described in Ref. [23]. We use 50 B splines of order 10 for each single-particle angular momentum state, and we include all orbitals with orbital angular momentum $l \leq 9$ in our basis set.

For atoms with one hole in the closed shells and one electron above the closed shells, the model space is formed from hole-particle states of the type $a_i^\dagger a_a |0\rangle$ where a_i^\dagger and a_a are creation and annihilation operators, respectively, and $|0\rangle$ is the closed-shell $[\text{Xe}]4f^{14}5d^{10}6s^26p^6$ ground state. The single-particle indices v and a designate valence and core states, respectively. For our study of low-lying $6p^{-1}nl$ and $6s^{-1}nl$ states of Rn-like ion, the values of a are $6s_{1/2}$, $6p_{1/2}$, and $6p_{3/2}$ while the values of v are $5f_{5/2}$, $5f_{7/2}$, $6d_{3/2}$, $6d_{5/2}$, $7s_{1/2}$, $7p_{1/2}$, and $7p_{3/2}$.

To obtain orthonormal model states, we consider the coupled states $\Phi_{JM}(av)$ defined by

$$\Phi_{JM}(av) = \sqrt{(2J+1)} \sum_{m_a m_v} (-1)^{j_v - m_v} \times \begin{pmatrix} j_v & J & j_a \\ -m_v & M & m_a \end{pmatrix} a_{vm_v}^\dagger a_{am_a} |0\rangle. \quad (1)$$

Combining the $6p_j$ and $6s$ hole orbitals and the $5f_{5/2}$, $5f_{7/2}$, $6d_{3/2}$, $6d_{5/2}$, $7s_{5/2}$, $7p_{1/2}$, and $7p_{3/2}$ particle orbitals, we obtain 28 even-parity states consisting of three $J = 0$ states, seven $J = 1$ states, eight $J = 2$ states, six $J = 3$ states, three $J = 4$ states, and one $J = 5$ state. Additionally, there are 24 odd-parity states consisting of three $J = 0$ states, seven $J = 1$ states, seven $J = 2$ states, five $J = 3$ states, and two $J = 4$ states. The distribution of the 72 states in the model space is summarized in Table I. Instead of using the $6p_j^{-1}nl_{j'}$ and $6s^{-1}nl_j$ designations, we use the simpler designations $6p_j nl_{j'}$ and $6s nl_j$ in this table and in all the following tables and in the text below.

III. EXCITATION ENERGIES

In Table II, we illustrate the relative size of various contributions before diagonalization by using the example of the even-parity states $6p_{j_1}5f_{j_2}$ with $J = 2$ and odd-parity states $6p_{j_1}6d_{j_2}$ with $J = 1$ in Rn-like Th^{4+} . The zeroth-, first-, and second-order Coulomb contributions $E^{(0)}$, $E^{(1)}$, and $E^{(2)}$ and the first- and second-order Breit-Coulomb corrections $B^{(1)}$ and $B^{(2)}$ are given.

The importance of correlation contributions is evident from this table; the ratio of the first and zeroth orders ($E^{(1)}/E^{(0)}$) is about 20–40%, and the ratio of the second and first ($E^{(2)}/E^{(1)}$) orders is even larger at 20–60%. It should be noted that corrections for the frequency-dependent Breit interaction [24] are included in the first order only. The difference between the first-order Breit corrections calculated with and without the frequency dependence is small: 1–2%. The Breit corrections are almost two orders of magnitude smaller than the corresponding Coulomb corrections of the same order: the ratio of the first-order Breit and Coulomb corrections is 1–2%.

The ratio of nondiagonal and diagonal matrix elements is larger for the second-order contributions than for the first-order contributions. The first-order nondiagonal matrix elements are

TABLE I. Possible hole-particle states in the $6p_j nl_{j'}(J)$ and $6s_{1/2} nl_j(J)$ complexes; jj coupling scheme.

Even-parity states			Odd-parity states		
$6p_{3/2}7p_{3/2}(0)$	$6p_{3/2}5f_{5/2}(2)$	$6p_{3/2}5f_{7/2}(3)$	$6p_{3/2}6d_{3/2}(0)$	$6p_{3/2}6d_{5/2}(2)$	$6p_{3/2}6d_{3/2}(3)$
$6p_{1/2}7p_{1/2}(0)$	$6p_{3/2}5f_{7/2}(2)$	$6p_{3/2}5f_{5/2}(3)$	$6p_{1/2}7s_{1/2}(0)$	$6p_{3/2}6d_{3/2}(2)$	$6p_{3/2}6d_{5/2}(3)$
$6s_{1/2}7s_{1/2}(0)$	$6p_{1/2}5f_{5/2}(2)$	$6p_{1/2}5f_{5/2}(3)$	$6s_{1/2}7p_{1/2}(0)$	$6p_{3/2}7s_{1/2}(2)$	$6p_{1/2}6d_{5/2}(3)$
	$6p_{3/2}7p_{1/2}(2)$	$6p_{1/2}5f_{7/2}(3)$		$6p_{1/2}6d_{3/2}(2)$	$6s_{1/2}5f_{5/2}(3)$
$6p_{3/2}5f_{5/2}(1)$	$6p_{3/2}7p_{3/2}(2)$	$6p_{3/2}7p_{3/2}(3)$	$6p_{3/2}6d_{3/2}(1)$	$6p_{1/2}6d_{5/2}(2)$	$6s_{1/2}5f_{7/2}(3)$
$6p_{3/2}7p_{1/2}(1)$	$6p_{1/2}7p_{3/2}(2)$	$6s_{1/2}6d_{5/2}(3)$	$6p_{3/2}6d_{5/2}(1)$	$6s_{1/2}5f_{5/2}(2)$	
$6p_{3/2}7p_{3/2}(1)$	$6s_{1/2}6d_{3/2}(2)$		$6p_{3/2}7s_{1/2}(1)$	$6s_{1/2}7p_{3/2}(2)$	$6p_{3/2}6d_{5/2}(4)$
$6p_{1/2}7p_{1/2}(1)$	$6s_{1/2}6d_{5/2}(2)$	$6p_{3/2}5f_{5/2}(4)$	$6p_{1/2}6d_{3/2}(1)$		$6s_{1/2}5f_{7/2}(4)$
$6p_{1/2}7p_{3/2}(1)$		$6p_{3/2}5f_{7/2}(4)$	$6p_{1/2}7s_{1/2}(1)$		
$6s_{1/2}6d_{3/2}(1)$		$6p_{1/2}5f_{7/2}(4)$	$6s_{1/2}7p_{1/2}(1)$		
$6s_{1/2}7s_{1/2}(1)$		$6p_{3/2}5f_{7/2}(5)$	$6s_{1/2}7p_{3/2}(1)$		

TABLE II. Contributions to the $E[6p_{j_1}5f_{j_2}, 6p_{j_3}5f_{j_4}, J = 2]$ and $E[6p_{j_1}6d_{j_2}, 6p_{j_3}6d_{j_4}, J = 1]$ energy matrices before diagonalization for Th^{4+} ion. Units are a.u.

$6p_{j_1}nl_{j_2}$	$6p_{j_3}nl_{j_4}$	$E^{(0)}$	$E^{(1)}$	$B^{(1)}$	$E^{(2)}$	$B^{(2)}$
$6p_{j_1}5f_{j_2}(2)-6p_{j_3}5f_{j_4}(2)$ energy matrix elements						
$6p_{1/2}5f_{5/2}$	$6p_{1/2}5f_{5/2}$	1.52956	-0.49040	-0.00600	-0.07886	-0.00185
$6p_{3/2}5f_{5/2}$	$6p_{3/2}5f_{5/2}$	1.19553	-0.49145	-0.00175	-0.06277	-0.00375
$6p_{3/2}5f_{7/2}$	$6p_{3/2}5f_{7/2}$	1.21113	-0.46358	-0.00258	-0.06599	-0.00332
$6p_{1/2}5f_{5/2}$	$6p_{3/2}5f_{5/2}$	0.00000	0.01756	-0.00001	-0.00176	0.00007
$6p_{3/2}5f_{5/2}$	$6p_{1/2}5f_{5/2}$	0.00000	0.01756	-0.00001	0.00188	0.00003
$6p_{1/2}5f_{5/2}$	$6p_{3/2}5f_{7/2}$	0.00000	0.04916	0.00000	-0.00837	-0.00010
$6p_{3/2}5f_{7/2}$	$6p_{1/2}5f_{5/2}$	0.00000	0.04916	0.00000	-0.00860	-0.00008
$6p_{3/2}5f_{5/2}$	$6p_{3/2}5f_{7/2}$	0.00000	-0.04483	0.00000	-0.00932	-0.00006
$6p_{3/2}5f_{7/2}$	$6p_{3/2}5f_{5/2}$	0.00000	-0.04483	0.00000	-0.00932	-0.00006
$6p_{j_1}6d_{j_2}(1)-6p_{j_3}6d_{j_4}(1)$ energy matrix elements						
$6p_{1/2}6d_{3/2}$	$6p_{1/2}6d_{3/2}$	1.50590	-0.32435	-0.00725	-0.06537	0.00610
$6p_{3/2}6d_{3/2}$	$6p_{3/2}6d_{3/2}$	1.17187	-0.38239	-0.00297	-0.01996	0.00440
$6p_{3/2}6d_{5/2}$	$6p_{3/2}6d_{5/2}$	1.19112	-0.25398	-0.00351	-0.07385	0.00443
$6p_{1/2}6d_{3/2}$	$6p_{3/2}6d_{3/2}$	0.00000	-0.00576	-0.00002	-0.01868	-0.00006
$6p_{3/2}6d_{3/2}$	$6p_{1/2}6d_{3/2}$	0.00000	-0.00576	-0.00002	-0.01110	-0.00003
$6p_{1/2}6d_{3/2}$	$6p_{3/2}6d_{5/2}$	0.00000	0.09682	-0.00002	-0.03915	-0.00032
$6p_{3/2}6d_{5/2}$	$6p_{1/2}6d_{3/2}$	0.00000	0.09682	-0.00002	-0.03937	-0.00010
$6p_{3/2}6d_{3/2}$	$6p_{3/2}6d_{5/2}$	0.00000	-0.07149	-0.00001	-0.01920	-0.00014
$6p_{3/2}6d_{5/2}$	$6p_{3/2}6d_{3/2}$	0.00000	-0.07149	-0.00001	-0.01785	-0.00015

symmetric, but the second-order nondiagonal matrix elements are not symmetric. The values of $E^{(2)}[a'v'(J), av(J)]$ and $E^{(2)}[av(J), a'v'(J)]$ matrix elements differ in some cases by a factor 2 to 3 and occasionally have opposite signs. We now discuss how the final energy levels are obtained from the above contributions. To determine the first-order energies, we diagonalize the symmetric first-order effective Hamiltonian, including both Coulomb and Breit interactions. The first-order expansion coefficient $C^N[av(J)]$ (often called a mixing coefficient) is the N th eigenvector of the first-order effective Hamiltonian, and $E^{(1)}[N]$ is the corresponding eigenvalue. The resulting eigenvectors are used to determine the second-order Coulomb correction $E^{(2)}[N]$, the second-order Breit-Coulomb correction $B^{(2)}[N]$, and the QED correction $E_{\text{LS}}[N]$.

In Table III, we list the following contributions to the energies of six excited states in Th^{4+} : the sum of the zeroth- and first-order energies $E^{(0+1)} = E^{(0)} + E^{(1)} + B^{(1)}$, the second-order Coulomb energy $E^{(2)}$, the second-order Breit-Coulomb correction $B^{(2)}$, the QED correction E_{LS} , and the sum of the

TABLE III. Energies of the Th^{4+} even-parity states with $J = 2$ and odd-parity states with $J = 1$ relative to the ground state. $E^{(0+1)} \equiv E^{(0)} + E^{(1)} + B^{(1)}$. Units are cm^{-1} .

jj Coupl.	$E^{(0+1)}$	$E^{(2)}$	$B^{(2)}$	E_{LS}	E_{tot}
$6p_{1/2}5f_{5/2}(2)$	146 877	-15682	-801	-36	130 358
$6p_{3/2}5f_{5/2}(2)$	168 907	-12053	-738	-34	156 082
$6p_{3/2}5f_{7/2}(2)$	228 605	-17832	-421	-4	210 349
$6p_{1/2}6d_{3/2}(1)$	165 691	-8367	951	36	158 309
$6p_{3/2}6d_{3/2}(1)$	203 897	-7154	1060	32	197 834
$6p_{3/2}6d_{5/2}(1)$	265 668	-19413	1267	4	247 525

above contributions E_{tot} . The Lamb shift E_{LS} is approximated as the sum of the one-electron self-energy and the first-order vacuum-polarization energy. The vacuum-polarization contribution is calculated from the Uehling potential using the results of Fullerton and Rinker [25]. The self-energy contribution is estimated for the s , $p_{1/2}$, and $p_{3/2}$ orbitals by interpolating among the values obtained by Mohr [26–28] using Coulomb wave functions. For this purpose, an effective nuclear charge Z_{eff} is obtained by finding the value of Z_{eff} required to give a Coulomb orbital with the same average $\langle r \rangle$ as the Dirac-Hartree-Fock (DHF) orbital.

When starting calculations from relativistic DF wave functions, it is natural to use jj designations for uncoupled energy matrix elements; however, neither jj nor LS coupling describes the *physical* states properly. We find that the mixing coefficients are 0.5–0.8. Therefore, we still use the jj designations in Table III. As discussed above, the correlation corrections are large and have to be included in order to obtain accurate energy values for Rn-like Th^{4+} ions. The second-order Coulomb contribution $E^{(2)}$ gives about 7–11% to the total values of the $6p_j5f_j(2)$ energies and about 5–7% in the case of the $6p_j6d_j(1)$ energies. Therefore, we expect energies to be accurate to few a 1000 cm^{-1} for the $6p_j5f_j$ and $6p_j6d_j$ states. Better accuracy is expected for higher states.

In Table IV, we compare our RMBPT results evaluated with different numbers of even and parity configurations to establish that the configuration space was chosen to be sufficiently large. We start our calculations with $6p5f$ even configurations and $6p6d$ odd configurations. This set of configurations is labeled set I. We note that, in RMBPT, energies are evaluated relative to the ground-state $6s^26p^6$ configuration, so all states

TABLE IV. Energies (in cm^{-1}) of Th^{4+} odd- and even-parity states relative to the ground state calculated in the first-order, second-order RMBPT (columns labeled RMBPT1 and RMBPT2, respectively) and Hartree-Fock relativistic methods implemented in COWAN code (column “COWAN”). The RMBPT values are evaluated with different configuration sets: I = $6p5f + 6p6d$, II = $6p5f + 6p7p + 6p6d + 6p7s$, and III = $6p5f + 6p7p + 6s6d + 6s7s$ and $6p6d + 6p7s + 6s5f + 6s7p$. Values listed in the COWAN column are obtained including the configuration set that is equivalent to set III. Values listed in the “COWAN2” column are obtained including larger configuration sets: IV = $6pn_1f + 6pn_2p + 6sn_3d + 6sn_2s$ and $6pn_3d + 6pn_2s + 6sn_1f + 6sn_2p$ with $n_1 = 5-9$, $n_2 = 7-9$, and $n_3 = 6-9$.

Level	I = $6p5f + 6p6d$		II = I + $6p7s + 6p7p$		III = II + $6snl$		III	IV	Level
	RMBPT1	RMBPT2	RMBPT1	RMBPT2	RMBPT1	RMBPT2	COWAN1	COWAN2	
$6p_{3/2}5f_{5/2}(1)$	141 555	129 246	141 547	105 289	140 482	103 961	135 015	135 013	$6s^26p^55f^3D_1$
$6p_{3/2}5f_{5/2}(2)$	146 877	130 394	146 869	105 388	145 992	103 880	140 455	140 469	$6s^26p^55f^3D_2$
$6p_{3/2}5f_{5/2}(4)$	150 413	136 694	150 413	112 300	150 413	111 611	143 711	143 819	$6s^26p^55f^3G_4$
$6p_{3/2}5f_{7/2}(5)$	150 707	138 834	150 707	115 935	150 707	115 151	145 498	145 606	$6s^26p^55f^3G_5$
$6p_{3/2}5f_{7/2}(3)$	154 183	138 917	154 169	114 801	153 648	114 426	147 639	147 698	$6s^26p^55f^3F_3$
$6p_{3/2}5f_{5/2}(3)$	157 390	143 301	157 387	118 076	157 314	117 597	150 675	150 759	$6s^26p^55f^1F_3$
$6p_{3/2}6d_{3/2}(0)$	161 473	160 273	161 270	143 308	161 118	142 323	158 046	158 019	$6s^26p^56d^3P_0$
$6p_{3/2}5f_{7/2}(4)$	162 859	148 681	162 859	122 502	162 859	123 001	156 273	156 377	$6s^26p^55f^3F_4$
$6p_{3/2}6d_{3/2}(1)$	165 690	158 274	165 329	141 809	165 218	142 612	161 938	161 937	$6s^26p^56d^3P_1$
$6p_{3/2}5f_{7/2}(2)$	168 907	156 116	168 895	132 377	168 847	131 630	160 896	160 980	$6s^26p^55f^1D_2$
$6p_{3/2}6d_{3/2}(3)$	172 385	170 385	172 385	152 598	171 752	151 508	167 627	167 594	$6s^26p^56d^3F_3$
$6p_{3/2}6d_{5/2}(2)$	172 998	170 417	172 477	152 808	172 385	152 205	168 668	168 716	$6s^26p^56d^3D_2$
$6p_{3/2}6d_{5/2}(4)$	173 587	173 071	173 587	157 264	172 707	156 599	170 023	169 952	$6s^26p^56d^3F_4$
$6p_{3/2}6d_{3/2}(2)$	177 952	175 189	177 770	158 302	177 529	157 399	173 883	173 943	$6s^26p^56d^1D_2$
$6p_{3/2}6d_{5/2}(3)$	184 028	182 885	184 028	163 728	183 988	163 203	180 423	180 490	$6s^26p^56d^3D_3$
$6p_{3/2}7s_{1/2}(2)$			200 043	181 307	199 837	176 394	197 339	197 368	$6s^26p^57s^3P_2$
$6p_{3/2}6d_{5/2}(1)$	203 897	197 802	200 224	193 055	200 125	181 998	196 638	196 142	$6s^26p^56d^3D_1$
$6p_{3/2}7s_{1/2}(1)$			206 648	204 574	206 336	184 100	201 455	201 094	$6s^26p^57s^1P_1$
$6p_{1/2}5f_{5/2}(3)$	216 016	200 234	216 016	175 538	216 008	174 712	209 762	209 865	$6s^26p^55f^3G_3$
$6p_{1/2}5f_{7/2}(3)$	221 128	205 968	221 087	181 138	220 528	179 353	215 134	215 174	$6s^26p^55f^3D_3$
$6p_{1/2}5f_{7/2}(4)$	223 552	208 384	223 552	182 604	223 552	181 736	217 423	217 527	$6s^26p^55f^3G_4$
$6p_{1/2}5f_{5/2}(2)$	228 605	210 352	228 536	183 656	228 343	182 566	218 808	218 875	$6s^26p^55f^1D_2$
$6p_{1/2}6d_{3/2}(2)$	240 564	234 779	240 570	218 010	239 469	214 502	234 454	234 409	$6s^26p^56d^3F_2$
$6p_{3/2}7p_{1/2}(1)$			241 862	225 806	241 203	225 123	239 426	239 426	$6s^26p^57p^3S_1$
$6p_{3/2}7p_{1/2}(2)$			243 260	227 234	243 127	227 154	241 353	241 220	$6s^26p^57p^3D_2$
$6p_{1/2}6d_{5/2}(2)$	247 155	243 237	247 348	225 029	247 313	224 226	242 599	242 601	$6s^26p^56d^3D_2$
$6p_{1/2}6d_{5/2}(3)$	249 440	245 446	249 440	226 857	248 809	225 564	244 843	244 774	$6s^26p^56d^3F_3$
$6p_{3/2}7p_{3/2}(3)$			257 423	243 556	257 358	243 547	254 321	254 057	$6s^26p^57p^3D_3$
$6p_{3/2}7p_{3/2}(1)$			257 837	244 127	257 565	243 751	254 497	254 490	$6s^26p^57p^1P_1$
$6p_{3/2}7p_{3/2}(2)$			260 638	245 352	260 604	245 298	257 628	257 660	$6s^26p^57p^3P_2$
$6p_{3/2}7p_{3/2}(0)$			265 082	249 010	265 006	247 409	262 805	262 436	$6s^26p^57p^1S_0$
$6p_{1/2}6d_{3/2}(1)$	265 668	247 521	265 725	238 692	265 207	236 355	260 581	257 646	$6s^26p^56d^1P_1$
$6p_{1/2}7s_{1/2}(0)$			271 083	248 586	270 620	242 959	267 274	267 242	$6s^26p^57s^3P_0$
$6p_{1/2}7s_{1/2}(1)$			272 272	255 027	271 974	244 433	269 390	268 988	$6s^26p^57s^3P_1$
$6p_{1/2}7p_{1/2}(1)$			314 337	294 877	314 067	294 316	311 834	308 721	$6s^26p^57p^3D_1$
$6p_{1/2}7p_{1/2}(0)$			319 975	295 120	319 824	294 038	317 680	317 230	$6s^26p^57p^3P_0$
$6p_{1/2}7p_{3/2}(1)$			329 978	312 026	329 480	311 070	325 794	325 548	$6s^26p^57p^3P_1$
$6p_{1/2}7p_{3/2}(2)$			330 479	312 400	330 351	312 142	326 655	327 359	$6s^26p^57p^3D_2$
$6s_{1/2}5f_{5/2}(2)$					379 141	268 796	373 775	373 234	$6s6p^65f^3F_2$
$6s_{1/2}5f_{5/2}(3)$					380 400	280 101	375 377	375 584	$6s6p^65f^3F_3$
$6s_{1/2}5f_{7/2}(4)$					383 146	290 684	378 968	379 915	$6s6p^65f^3F_4$
$6s_{1/2}5f_{7/2}(3)$					393 319	295 245	387 668	388 226	$6s6p^65f^1F_3$
$6s_{1/2}6d_{3/2}(1)$					408 353	348 770	404 219	406 496	$6s6p^66d^3D_1$
$6s_{1/2}6d_{3/2}(2)$					410 065	322 551	406 013	407 733	$6s6p^66d^3D_2$
$6s_{1/2}6d_{5/2}(3)$					414 519	340 751	411 416	412 197	$6s6p^66d^3D_3$
$6s_{1/2}6d_{5/2}(2)$					420 854	345 816	418 329	421 187	$6s6p^66d^1D_2$
$6s_{1/2}7s_{1/2}(1)$					440 017	363 232	438 376	439 210	$6s6p^67s^3S_1$
$6s_{1/2}7s_{1/2}(0)$					445 694	398 690	444 503	444 525	$6s6p^67s^1S_0$
$6s_{1/2}7p_{1/2}(0)$					482 789	434 148	481 499	482 141	$6s6p^67p^3P_0$
$6s_{1/2}7p_{1/2}(1)$					483 650	413 518	482 498	483 198	$6s6p^67p^3P_1$
$6s_{1/2}7p_{3/2}(2)$					498 021	405 747	495 728	496 608	$6s6p^67p^3P_2$
$6s_{1/2}7p_{3/2}(1)$					500 704	415 279	498 460	498 666	$6s6p^67p^1P_1$

are of the hole-particle type. In our designations, the first nl indicates the hole state and the second nl indicates the particle state. In set II, $6p7p$ and $6p7s$ configurations are added and in set III $6snl$ configurations are added. Therefore, our largest set (set III) contains $6p5f + 6p7p + 6s6d + 6s7s$ even and $6p6d + 6p7s + 6s5f + 6s7p$ odd configurations. To show the size of the correlation contribution, we also included the data evaluated in the first-order approximation in the columns labeled “RMBPT1” in Table IV. These data are obtained as a sum of the $E^{(0)}$, $E^{(1)}$, and $B^{(1)}$ values (see caption of Table III).

Our final second-order values are listed in the columns labeled “RMBPT2.” The ratios of values in the RMBPT1 and RMBPT2 columns range from 1.2 to 1.6. We also include results obtained from the relativistic Hartree-Fock approximation (COWAN code) [29] in the same table. Values listed in the “COWAN” column are obtained including the configurations $6s^26p^6 + 6s^26p^55f + 6s^26p^57p + 6s6p^66d + 6s6p^67s$ and $6s^26p^56d + 6s^26p^57s + 6s6p^65f + 6s6p^67p$. Therefore, the set of configurations is equivalent to our final set III. We also carried out additional calculations of the energies of Th^{4+} odd- and even-parity states relative to the ground state evaluated by COWAN code using the large set of configurations: $IV = 6pn_1f + 6pn_2p + 6sn_3d + 6sn_2s$, $6pn_3d + 6pn_2s + 6sn_1f + 6sn_2p$ with $n_1 = 5-9$, $n_2 = 7-9$, and $n_3 = 6-9$. Results of our calculations are incorporated into Table IV in the additional column “COWAN2.” We find very small differences, 0.003–0.5%, between results displayed in columns “COWAN1” and “COWAN2.” Slightly larger differences, about 1%, are observed for the energies of the $6s^26p^56d^1P_1$ and $6s^26p^57p^3D_1$ levels. The scaling of electrostatic integrals in the COWAN code allows us to correct for correlation effects. In many systems, it leads to good agreement with experimental energies. We used the same scaling factor (0.85) for all electrostatic integrals. The 0.85 scaling factor was introduced for the first time by Fawcett *et al.* [30]. The authors explained that the 0.85 factor was found empirically to obtain results in good agreement with experiment. Later, the 0.85 factor was used in other publications. We note that this factor may not produce accurate results in all systems.

The differences between the RMBPT1 values for different numbers of configurations (columns 2, 4, and 6 in Table IV) are very small (about 0.1–0.8%). This means that configuration interaction (CI) does not significantly contribute in first-order RMBPT. The differences between the RMBPT2 values for different numbers of configurations (columns 3, 5, and 7 in Table IV) are much larger (about 7–20%), indicating that including CI contribution leads to additional correlation correction.

As expected, the second-order correction (the difference between the RMBPT2 and RMBPT1 values) depends on the number of considered configurations. In the first case ($I = 6p5f, 6p6d$) the differences in the results given in columns 2 and 3 are about 1–11%, while the differences in the results given in columns 4 and 5, and 6 and 7 are about 9–28%. The differences between RMBPT2 $6p5f$ and $6p6d$ values calculated with sets I and II are large for most of the states, indicating that set I, which includes mixing only within these configurations, is insufficient. The differences between sets II

and III for the $6p5f$ and $6p6d$ configurations are small for most of the states.

The COWAN results are in better agreement with RMBPT1 values (about 1–5%). The differences of the COWAN results with our final RMBPT2 values are about 5–25%. We note that second-order RMBPT has a general tendency to overestimate the correlation correction. Full all-order treatment, which may be carried out within the framework of the coupled-cluster approach, is needed to improve the accuracy. It may be possible in the future to implement the hybrid configuration interaction + the linearized coupled-cluster method [31] for hole-particle states. This work provides a starting point for further development of theoretical methods for such highly correlated and relativistic systems.

Our RMBPT values presented in Table IV are *ab initio* values for the energy levels in Th^{4+} . To the best of our knowledge, there are no experimental energy values for this ion.

In Table V, we list energies of odd- and even-parity configurations in Rn-like Ac^{3+} and U^{6+} calculated in first-order and second-order RMBPT (RMBPT1 and RMBPT2, respectively). All values are given relative to the ground-state energy. The RMBPT values are evaluated including the following configurations: $6p5f + 6p7p + 6s6d + 6s7s$ and $6p6d + 6p7s + 6s5f + 6s7p$. This set of states was used to evaluate energies in Rn-like Th^{4+} given in columns 6 and 7 of Table IV (set III). The major difference of the level distribution in Rn-like Ac^{3+} and U^{6+} is in the placement of the metastable states relative to the ground state. In Rn-like U^{6+} , the first eleven levels are metastable, while in Rn-like Ac^{3+} , there is only one metastable level before the odd-parity level with $J = 1$. It should be noted that there are eight first metastable states in the case of Rn-like Th^{4+} . The importance of the existence of the low-lying metastable states was discussed in the recent experimental work by Hanni *et al.* [2].

IV. MULTIPOLE TRANSITIONS FROM EXCITED STATES TO GROUND STATE

The first 52 excited states in Rn-like Ac, Th, and U ions are $6s^26p^5nl$ and $6s6p^6n'l'$ states. There are 45 metastable levels with $J = 0, 2-5$, but only seven $J = 1$ levels that can decay to the ground state via electric-dipole transitions. Below, we consider all possible multipole ($E1$, $E2$, $E3$, $M1$, $M2$, and $M3$) transitions from the $6s^26p^5nl$ and $6s6p^6n'l'$ states to the $6s^26p^6$ ground state in Rn-like Ac, Th, and U ions.

We calculate the electric-dipole ($E1$) matrix elements for the transitions between the seven odd-parity $6p_j6d_j(1)$, $6p_j7s(1)$, and $6s7p_j(1)$ excited states and the ground state, magnetic-quadrupole ($M2$) matrix elements between the seven odd-parity $6p_j6d_j(2)$, $6p_j7s(2)$, $6s5f_j(2)$, and $6s7p_j(2)$ excited states and the ground state, and electric-octupole ($E3$) matrix elements between the five odd-parity $6p_j6d_j(3)$ and $6s5f_j(3)$ excited states and the ground state.

The magnetic-dipole ($M1$) matrix elements are calculated for the transitions between the seven even-parity $6p_j5f_j(1)$, $6p_j7p_j(1)$, $6s6d_j(1)$, and $6s7s(1)$ excited states and the ground state, electric-quadrupole ($E2$) matrix elements between the eight even-parity $6p_j5f_j(2)$, $6p_j7p_j(2)$, and

TABLE V. Energies (in cm^{-1}) of odd- and even-parity states in Ac^{3+} and U^{6+} calculated in first-order and second-order RMBPT (RMBPT1 and RMBPT2, respectively). All values are given relative to the ground-state energy.

Level	RMBPT1	RMBPT2	Level	RMBPT1	RMBPT2	Level	RMBPT1	RMBPT2
Rn-like Ac^{3+}								
$6p_{3/2}6d_{3/2}(0)$	142 038	124 368	$6p_{3/2}7p_{1/2}(1)$	196 806	181 316	$6p_{1/2}7p_{3/2}(1)$	269 663	251 835
$6p_{3/2}6d_{3/2}(1)$	145 180	126 163	$6p_{1/2}5f_{5/2}(2)$	198 324	182 388	$6p_{1/2}7p_{3/2}(2)$	270 351	250 787
$6p_{3/2}6d_{5/2}(2)$	150 536	131 374	$6p_{1/2}5f_{5/2}(3)$	208 424	195 064	$6s_{1/2}6d_{3/2}(1)$	365 044	282 364
$6p_{3/2}6d_{5/2}(4)$	150 919	135 347	$6p_{1/2}6d_{3/2}(2)$	208 664	184 565	$6s_{1/2}6d_{3/2}(2)$	366 332	263 154
$6p_{3/2}6d_{3/2}(3)$	150 969	131 709	$6p_{3/2}7p_{3/2}(1)$	208 726	195 394	$6s_{1/2}6d_{5/2}(3)$	369 412	289 445
$6p_{3/2}6d_{3/2}(2)$	155 359	136 135	$6p_{3/2}7p_{1/2}(2)$	210 971	194 549	$6s_{1/2}6d_{5/2}(2)$	373 809	294 984
$6p_{3/2}5f_{5/2}(1)$	158 406	127 266	$6p_{1/2}6d_{5/2}(2)$	214 246	191 942	$6s_{1/2}5f_{5/2}(2)$	375 275	257 234
$6p_{3/2}6d_{5/2}(3)$	160 333	140 529	$6p_{3/2}7p_{3/2}(0)$	214 361	200 320	$6s_{1/2}5f_{5/2}(3)$	376 279	266 253
$6p_{3/2}5f_{5/2}(2)$	163 021	125 988	$6p_{1/2}6d_{5/2}(3)$	215 371	192 305	$6s_{1/2}5f_{7/2}(4)$	378 368	258 153
$6p_{3/2}7s_{1/2}(2)$	163 262	140 653	$6p_{1/2}7s_{1/2}(0)$	222 529	195 812	$6s_{1/2}7s_{1/2}(1)$	378 646	299 676
$6p_{3/2}6d_{5/2}(1)$	164 608	138 801	$6p_{1/2}6d_{3/2}(1)$	223 611	195 756	$6s_{1/2}7s_{1/2}(0)$	383 436	328 361
$6p_{3/2}5f_{7/2}(5)$	166 952	136 050	$6p_{1/2}5f_{7/2}(3)$	223 637	186 587	$6s_{1/2}5f_{7/2}(3)$	386 293	271 861
$6p_{3/2}5f_{5/2}(4)$	167 323	133 173	$6p_{3/2}7p_{3/2}(3)$	226 680	190 132	$6s_{1/2}7p_{1/2}(0)$	414 440	321 913
$6p_{3/2}5f_{7/2}(3)$	169 386	135 646	$6p_{1/2}7s_{1/2}(1)$	228 866	204 119	$6s_{1/2}7p_{1/2}(1)$	415 115	322 430
$6p_{3/2}5f_{5/2}(3)$	172 871	138 259	$6p_{1/2}5f_{7/2}(4)$	229 410	192 294	$6s_{1/2}7p_{3/2}(2)$	425 336	334 106
$6p_{3/2}5f_{7/2}(4)$	177 118	142 705	$6p_{3/2}7p_{3/2}(2)$	234 014	193 320	$6s_{1/2}7p_{3/2}(1)$	427 437	335 361
$6p_{3/2}7s_{1/2}(1)$	177 403	158 475	$6p_{1/2}7p_{1/2}(1)$	258 590	239 611			
$6p_{3/2}5f_{7/2}(2)$	183 095	150 944	$6p_{1/2}7p_{1/2}(0)$	262 964	244 151			
Rn-like U^{6+}								
$6p_{3/2}5f_{5/2}(1)$	949 98	550 84	$6p_{3/2}6d_{5/2}(3)$	230 270	207 864	$6s_{1/2}5f_{7/2}(4)$	389 533	289 901
$6p_{3/2}5f_{5/2}(2)$	101 988	572 70	$6p_{3/2}6d_{5/2}(1)$	254 864	233 974	$6s_{1/2}5f_{7/2}(3)$	400 241	300 071
$6p_{3/2}5f_{5/2}(4)$	105 362	644 83	$6p_{3/2}7s_{1/2}(2)$	278 438	252 308	$6p_{1/2}7p_{1/2}(1)$	432 078	410 751
$6p_{3/2}5f_{7/2}(5)$	108 529	706 64	$6p_{3/2}7s_{1/2}(1)$	283 002	257 632	$6p_{1/2}7p_{1/2}(0)$	440 258	414 253
$6p_{3/2}5f_{7/2}(3)$	111 012	683 32	$6p_{1/2}6d_{3/2}(2)$	302 991	276 027	$6p_{1/2}7p_{3/2}(1)$	457 683	437 407
$6p_{3/2}5f_{5/2}(3)$	114 963	731 17	$6p_{1/2}6d_{5/2}(2)$	316 064	291 979	$6p_{1/2}7p_{3/2}(2)$	458 964	438 152
$6p_{3/2}5f_{7/2}(4)$	122 022	794 33	$6p_{1/2}6d_{5/2}(3)$	317 833	292 780	$6s_{1/2}6d_{3/2}(1)$	497 589	420 550
$6p_{3/2}5f_{7/2}(2)$	125 875	860 91	$6p_{3/2}7p_{1/2}(1)$	334 373	316 388	$6s_{1/2}6d_{3/2}(2)$	500 122	406 883
$6p_{1/2}5f_{5/2}(3)$	194 364	151 076	$6p_{3/2}7p_{1/2}(2)$	336 877	318 700	$6s_{1/2}6d_{5/2}(3)$	507 805	426 632
$6p_{3/2}6d_{3/2}(0)$	199 190	179 669	$6p_{1/2}6d_{3/2}(1)$	336 883	307 969	$6s_{1/2}6d_{5/2}(2)$	517 101	431 761
$6p_{1/2}5f_{7/2}(3)$	202 138	158 903	$6p_{3/2}7p_{3/2}(3)$	360 871	345 607	$6s_{1/2}7s_{1/2}(1)$	569 975	490 373
$6p_{3/2}6d_{3/2}(1)$	204 827	180 757	$6p_{3/2}7p_{3/2}(1)$	360 906	345 845	$6s_{1/2}7s_{1/2}(0)$	577 075	426 676
$6p_{1/2}5f_{7/2}(4)$	205 279	161 277	$6p_{3/2}7p_{3/2}(2)$	365 284	348 152	$6s_{1/2}7p_{1/2}(0)$	626 479	540 259
$6p_{1/2}5f_{5/2}(2)$	206 954	159 384	$6p_{3/2}7p_{3/2}(0)$	371 487	351 760	$6s_{1/2}7p_{1/2}(1)$	627 635	540 631
$6p_{3/2}6d_{3/2}(3)$	212 406	189 846	$6p_{1/2}7s_{1/2}(0)$	374 193	344 198	$6s_{1/2}7p_{3/2}(2)$	651 916	569 289
$6p_{3/2}6d_{5/2}(2)$	214 136	191 900	$6p_{1/2}7s_{1/2}(1)$	375 981	345 896	$6s_{1/2}7p_{3/2}(1)$	655 499	570 646
$6p_{3/2}6d_{5/2}(4)$	216 216	198 106	$6s_{1/2}5f_{5/2}(2)$	383 261	284 422			
$6p_{3/2}6d_{3/2}(2)$	221 462	200 736	$6s_{1/2}5f_{5/2}(3)$	384 876	285 957			

$6s6d_j(2)$ excited states and the ground state, and magnetic-octupole ($M3$) matrix elements between the six even-parity $6p_j5f_j(3)$, $6p_j7p_j(3)$, and $6s6d_j(3)$ excited states and the ground state.

Analytical expressions in the first- and the second-order RMBPT are given by Eqs. (2.12)–(2.17) of Ref. [13] for the $E1$ matrix elements and in Refs. [15,17,18] for the $M1$, $M2$, $M3$, $E2$, and $E3$ matrix elements.

We refer to the first- and second-order Coulomb corrections and second-order Breit-Coulomb corrections to reduced multipole matrix elements as $Z^{(1)}$, $Z^{(2)}$, and $B^{(2)}$, respectively, throughout the text. In Table VI, we list values of *uncoupled* first- and second-order $E1$, $E2$, $E3$, $M1$, $M2$, and $M3$ matrix elements $Z^{(1)}$, $Z^{(2)}$, and $B^{(2)}$ together with derivative terms $P^{(\text{deriv})}$ for Th^{4+} (see Refs. [15,17,18] for detail). The

importance of the correlation contribution is evident from this table; the ratio of the second and first orders, $Z^{(2)}/Z^{(1)}$, is very large for $E1$ transitions [25–35% for the $6p_j6d_j(1)$ states and a factor of 2 to 3 for the $6s7p_j(1)$ states]. However, the ratio of the second and first orders, $Z^{(2)}/Z^{(1)}$, is much smaller for the $E2$ and $E3$ transitions (10 and 5%, respectively). The $Z^{(2)}/Z^{(1)}$ ratio for the $M1$ magnetic dipole transitions is about 15–20% for the two $6p_j7p_j(1)$ states. We note that we do not list $M1$ transitions with almost zero value of the $Z^{(1)}$ matrix elements for the $6p_{3/2}5f_{5/2}(1)$, $6s6d_{3/2}(1)$, $6s7s(1)$ states. The second-order $Z^{(2)}$ contributions for these states are comparable with the $Z^{(2)}$ contributions for the $6p_j7p_j(1)$ states listed in Table VI. The ratio of the second and first orders, $Z^{(2)}/Z^{(1)}$, for the $M2$ and $M3$ transitions is about 10–20%.

TABLE VI. $E1$, $E2$, $E3$, $M1$, $M2$, and $M3$ uncoupled reduced matrix elements in length L form for transitions from $av(J)$ states with $J = 1, 2$, and 3 into the ground state in Th^{4+} .

$av(J)$	$Z^{(1)}$	$Z^{(2)}$	$B^{(2)}$	$p^{(\text{deriv})}$
<i>E1</i> uncoupled reduced matrix elements				
$6p_{3/2}6d_{3/2}(1)$	-0.9643	0.2442	-0.0011	-0.9643
$6p_{3/2}6d_{5/2}(1)$	-2.8448	0.6978	-0.0040	-2.8444
$6p_{1/2}6d_{3/2}(1)$	-1.8154	0.6314	-0.0040	-1.8152
$6s_{1/2}7p_{1/2}(1)$	-0.0928	0.1739	0.0010	-0.0926
$6s_{1/2}7p_{3/2}(1)$	0.0611	-0.2119	0.0004	0.0612
<i>E2</i> uncoupled reduced matrix elements				
$6p_{3/2}5f_{5/2}(2)$	-1.9189	0.1946	-0.0089	-3.8378
$6p_{3/2}5f_{7/2}(2)$	4.8084	-0.5097	0.0185	9.6164
$6p_{1/2}5f_{5/2}(2)$	-3.2324	0.3907	-0.0159	-6.4644
<i>E3</i> uncoupled reduced matrix elements				
$6p_{3/2}6d_{3/2}(3)$	12.1130	-0.4005	0.0542	36.4897
$6p_{3/2}6d_{5/2}(3)$	10.0607	-0.0287	0.0428	30.0992
$6p_{1/2}6d_{5/2}(3)$	-8.5284	-0.5985	-0.0507	-25.4016
<i>M1</i> uncoupled reduced matrix elements				
$6p_{3/2}7p_{1/2}(1)$	-0.1333	-0.0200	0.0009	-0.1332
$6p_{1/2}7p_{3/2}(1)$	-0.1001	-0.0229	0.0005	-0.1001
<i>M2</i> uncoupled reduced matrix elements				
$6p_{3/2}6d_{5/2}(2)$	-12.5437	-0.9870	-0.0177	-25.0863
$6p_{3/2}7s_{1/2}(2)$	-4.3993	-0.4262	-0.0061	-8.7979
$6p_{1/2}6d_{3/2}(2)$	-1.4060	0.0526	-0.0031	-2.8122
$6p_{1/2}6d_{5/2}(2)$	-4.4641	-0.8952	-0.0116	-8.9274
<i>M3</i> uncoupled reduced matrix elements				
$6p_{3/2}5f_{7/2}(3)$	27.9342	-2.6561	0.1081	83.8009
$6p_{1/2}5f_{7/2}(3)$	14.4537	-1.7104	0.0625	43.3598
$6p_{3/2}7p_{3/2}(3)$	-13.6545	-2.6745	-0.0329	-40.9599
$6s_{1/2}6d_{5/2}(3)$	18.0124	-4.3662	0.0450	54.0327

The $E1$, $E2$, $E3$, $M2$, $M3$, and $M3$ transition probabilities A_r (s^{-1}) for the transitions between the ground state and $6p_j n l_j(J)$ and $6s n l_j(J)$ states are obtained in terms of line strengths S (a.u.) and energies E (a.u.) as

$$A(Ek) = \frac{C^{(k)} [E]^{2k+1}}{(2J+1)} S(Ek),$$

$$\begin{aligned} C^{(1)} &= 2.14200 \times 10^{10}, \\ C^{(2)} &= 5.70322 \times 10^4, \\ C^{(3)} &= 7.71311 \times 10^{-2}, \end{aligned} \quad (2)$$

$$A(Mk) = \frac{D^{(k)} [E]^{2k+1}}{(2J+1)} S(Mk),$$

$$\begin{aligned} D^{(1)} &= 2.85161 \times 10^5, \\ D^{(2)} &= 7.59260 \times 10^{-1}, \\ D^{(3)} &= 1.02683 \times 10^{-6}. \end{aligned} \quad (3)$$

The line strengths $S(E1)$, $S(E2)$, $S(E3)$, $S(M1)$, $S(M2)$, and $S(M3)$ are obtained as squares of the corresponding *coupled* $E1$, $E2$, $E3$, $M2$, $M3$, and $M3$ matrix elements. The $E1$, $E2$, $E3$, $M2$, $M3$, $M2$, and $M3$ *coupled* matrix elements are evaluated using an intermediate-coupling scheme (see Ref. [13] for details). Results of our calculations are given in Tables VII and VIII, where we present wavelengths

(in Å), line strengths (S), and radiative rates (A_r) for the electric-multipole ($E1$, $E2$, and $E3$) and magnetic-multipole ($M1$, $M2$, and $M3$) transitions from $6pnl$ and $6snl$ states to the ground state in Rn-like Th^{4+} , Ac^{3+} , and U^{6+} ions. Wavelengths, line strengths and radiative rates are calculated in first- and second-order RMBPT. As expected, we find substantial differences between first- and second-order MBPT results, which are given in the columns labeled “a” and “b.” These differences give the value of the second-order correlation correction. We discuss results for Th^{4+} ion first. The ratios of transition rates, A_r^{E1}/A_r^{E2} and A_r^{E2}/A_r^{M1} , are equal to 10^3 to 10^6 and 10 to 10^2 , respectively. $M3$ transitions are the weakest transitions, as expected (10^{-6} to 10^{-3} s^{-1}). The values of the transition rates change inside the of complex states with fixed J by three to four orders of magnitude. As a result, the values of transition rates given in Table VIII vary from 10^{-6} s^{-1} for the $M3$ transitions up to 10^{10} s^{-1} for the $E1$ transitions.

The difference between results in Rn-like Ac^{3+} (left columns) and U^{6+} ions (right columns) is not only in the values of wavelengths, line strengths, and radiative rates but in the order of transitions. The study of the order of the first five energy levels given in Table V shows that the order of possible strongest decay channels is $E1$, $M2$, $E3$ for the Ac^{3+} ion and $M1$, $E2$, and $M3$ for the U^{6+} ion (the other two levels cannot decay to the ground state via any of the six transitions up to $k = 3$).

TABLE VII. Wavelengths (λ in Å), line strengths (S in a.u), and radiative rates (A_r in s^{-1}) for the electric-multipole ($E1$, $E2$, and $E3$) and magnetic-multipole ($M1$, $M2$, and $M3$) transitions from $6pnl$ and $6snl$ states to the ground state in Rn-like Th^{4+} . The wavelengths, line strengths, and radiative rates calculated in the first-order RMBPT are listed in columns labeled “(a).” The second-order RMBPT results are listed in columns labeled “(b).” Numbers in brackets represent powers of ten.

$6ln'l'$	Wavelength		Line strength		Transition rate		$6ln'l'$	Wavelength		Line strength		Transition rate			
	(a)	(b)	(a)	(b)	(a)	(b)		(a)	(b)	(a)	(b)	(a)	(b)		
$M3$	$6p_{3/2}5f_{7/2}$	650.8	873.9	6.1[2]	1.9[3]	7.3[−6]	2.9[−6]	$E3$	$6p_{1/2}6d_{5/2}$	401.9	443.3	8.7[1]	2.9[2]	2.3[0]	3.8[0]
$M3$	$6p_{3/2}5f_{5/2}$	635.7	850.4	3.0[2]	8.6[2]	4.3[−6]	1.6[−6]	$M3$	$6p_{3/2}7p_{3/2}$	388.6	410.6	2.1[2]	3.7[2]	9.5[−5]	1.1[−4]
$E1$	$6p_{3/2}6d_{3/2}$	605.3	701.2	5.8[−3]	6.7[−3]	1.8[7]	1.3[7]	$M1$	$6p_{3/2}7p_{3/2}$	388.3	410.3	1.1[−3]	1.9[−3]	1.6[2]	2.4[2]
$E3$	$6p_{3/2}6d_{5/2}$	543.5	612.7	1.3[2]	1.6[2]	4.3[−1]	2.2[−1]	$E2$	$6p_{3/2}7p_{3/2}$	383.7	407.7	2.3[0]	1.4[0]	6.2[4]	2.9[4]
$M2$	$6p_{3/2}7s_{1/2}$	500.4	566.9	5.9[0]	3.6[0]	5.6[−1]	1.8[−1]	$E1$	$6p_{1/2}6d_{3/2}$	377.1	423.1	7.6[0]	3.2[0]	9.6[10]	2.8[10]
$E1$	$6p_{3/2}7s_{1/2}$	484.6	543.2	5.5[0]	4.0[0]	3.3[10]	1.7[10]	$E1$	$6p_{1/2}7s_{1/2}$	367.7	409.1	2.7[−1]	3.4[−1]	3.6[9]	3.4[9]
$M3$	$6p_{1/2}5f_{5/2}$	462.9	572.4	2.0[1]	3.7[1]	2.6[−6]	1.1[−6]	$M1$	$6p_{1/2}7p_{3/2}$	303.5	321.5	1.1[−2]	2.0[−2]	3.6[3]	5.4[3]
$M2$	$6p_{1/2}6d_{3/2}$	417.6	466.2	5.2[0]	2.8[0]	1.2[0]	3.8[−1]	$E2$	$6p_{1/2}7p_{3/2}$	302.7	320.4	9.3[−1]	6.2[−1]	8.2[4]	4.1[4]
$M1$	$6p_{3/2}7p_{1/2}$	414.6	444.2	1.6[−2]	1.9[−2]	2.0[3]	1.9[3]	$M2$	$6s_{1/2}5f_{5/2}$	263.8	372.0	3.5[−3]	9.0[−3]	8.2[−3]	3.8[−3]
$E2$	$6p_{3/2}7p_{1/2}$	411.3	440.2	5.8[0]	3.1[0]	1.1[5]	4.3[4]	$E2$	$6s_{1/2}6d_{3/2}$	243.9	310.0	1.0[0]	7.5[−1]	2.6[5]	5.8[4]
$M2$	$6p_{1/2}6d_{5/2}$	404.3	446.0	8.5[0]	6.6[0]	2.4[0]	1.1[0]	$M2$	$6s_{1/2}7p_{3/2}$	200.8	246.5	2.3[−1]	1.9[−1]	2.0[0]	6.3[−1]

V. GROUND STATE MULTIPOLE POLARIZABILITIES IN RADON-LIKE ACTINIUM, THORIUM, AND URANIUM IONS

The ground-state multipole polarizabilities are evaluated here in the DHF and random-phase approximation (RPA) [32] using the sum-over-states approach:

$$\alpha^{Ek} = \frac{2}{2k+1} \sum_{n,a} \frac{|\langle nl_j || r^k C_{kq} || al'_{j'} \rangle|^2}{E_{nl_j} - E_{al'_{j'}}}, \quad (4)$$

where $C_{kq}(\hat{r})$ is a normalized spherical harmonic and where $al'_{j'}$ include all core states in Rn-like ions from the $1s_{1/2}$ up to $6p_j$ states, nl_j includes all valence states above the core,

and $k = 1, 2$, and 3 , for dipole, quadrupole, and octupole polarizabilities, respectively [33]. The quantities α^{E1} and α^{E2} are dipole and quadrupole polarizabilities labeled α_D and α_Q in [4]. We also calculated parameters β_D and γ_D defined in [4] as

$$\beta_D = \frac{1}{3} \sum_{n,a} \frac{|\langle nl_j || D || al'_{j'} \rangle|^2}{(E_{nl_j} - E_{al'_{j'}})^2}, \quad (5)$$

$$\gamma_D = \frac{1}{6} \sum_{n,a} \frac{|\langle nl_j || D || al'_{j'} \rangle|^2}{(E_{nl_j} - E_{al'_{j'}})^3}, \quad (6)$$

where D is the dipole operator. All sums are carried out with the finite basis set described above, making the sums finite

TABLE VIII. Wavelengths (λ in Å), line strengths (S in a.u), and radiative rates (A_r in s^{-1}) for the electric-multipole ($E1$, $E2$, and $E3$) and magnetic-multipole ($M1$, $M2$, and $M3$) transitions from $6pnl$ and $6snl$ states to the ground state in Rn-like Ac^{3+} and U^{6+} ions. The wavelengths, line strengths, and radiative rates calculated in the first-order RMBPT are listed in columns labeled “(a).” The second-order RMBPT results are listed in columns labeled “(b).” Numbers in brackets represent powers of ten.

$6ln'l'(J)$	Wavelength		Line strength		Transition rate		$6ln'l'(J)$	Wavelength		Line strength		Transition rate			
	(a)	(b)	(a)	(b)	(a)	(b)		(a)	(b)	(a)	(b)	(a)	(b)		
Radon-like Actinium						Radon-like Uranium									
$E1$	$6p_{3/2}6d_{3/2}(1)$	689	793	3.1[−3]	4.3[−2]	6.4[6]	5.8[7]	$M1$	$6p_{3/2}5f_{5/2}(1)$	1053	815	1.8[−25]	8.2[−15]	3.2[−9]	1.4[−4]
$M2$	$6p_{3/2}6d_{5/2}(2)$	664	761	1.6[4]	5.1[3]	2.9[0]	8.3[−1]	$E2$	$6p_{3/2}5f_{5/2}(2)$	981	746	1.6[−1]	3.9[−2]	4.0[1]	5.3[−1]
$E3$	$6p_{3/2}6d_{3/2}(3)$	662	759	1.8[2]	5.9[3]	1.4[−1]	1.8[0]	$M3$	$6p_{3/2}5f_{7/2}(3)$	901	464	1.3[4]	4.9[4]	1.4[−7]	9.2[−9]
$M2$	$6p_{3/2}6d_{3/2}(2)$	644	735	7.2[1]	3.8[1]	1.9[0]	5.3[−1]	$E3$	$6p_{3/2}6d_{5/2}(3)$	434	481	6.1[1]	5.9[1]	9.4[−1]	4.5[−1]
$E1$	$6p_{3/2}6d_{5/2}(1)$	608	721	4.1[−1]	6.5[−1]	1.2[9]	1.2[9]	$E1$	$6p_{3/2}6d_{5/2}(1)$	392	427	3.5[0]	1.8[0]	3.9[10]	1.6[10]
$E1$	$6p_{3/2}7s_{1/2}(1)$	564	631	6.4[0]	6.6[0]	2.4[10]	1.8[10]	$M2$	$6p_{3/2}7s_{1/2}(2)$	359	396	1.1[1]	6.3[0]	5.4[0]	1.9[0]
$M1$	$6p_{3/2}7p_{1/2}(1)$	508	552	1.3[−2]	2.1[−2]	9.0[2]	1.1[3]	$E1$	$6p_{3/2}7s_{1/2}(1)$	353	388	1.4[0]	1.4[0]	2.1[10]	1.6[10]
$E2$	$6p_{1/2}5f_{5/2}(2)$	504	548	7.2[0]	3.5[0]	5.0[4]	1.6[4]	$M2$	$6p_{1/2}6d_{3/2}(2)$	330	362	4.4[0]	3.4[0]	3.4[0]	1.6[0]
$M3$	$6p_{1/2}5f_{5/2}(3)$	480	513	2.4[2]	4.7[2]	2.4[−5]	3.0[−5]	$E3$	$6p_{1/2}6d_{5/2}(3)$	315	342	4.2[1]	8.8[1]	6.2[0]	7.3[0]
$M2$	$6p_{1/2}6d_{3/2}(2)$	480	542	5.4[0]	2.9[0]	6.4[−1]	1.9[−1]	$M1$	$6p_{3/2}7p_{1/2}(1)$	299	316	2.0[−2]	2.5[−2]	6.8[3]	7.1[3]
$M1$	$6p_{3/2}7p_{3/2}(1)$	479	512	1.0[−3]	1.6[−3]	8.3[1]	1.0[2]	$E2$	$6p_{3/2}7p_{1/2}(2)$	297	314	4.0[0]	2.4[0]	3.9[5]	1.8[5]
$E2$	$6p_{3/2}7p_{1/2}(2)$	474	514	3.4[0]	1.7[0]	3.2[4]	1.1[4]	$E1$	$6p_{1/2}6d_{3/2}(1)$	297	325	6.6[0]	3.4[0]	1.7[11]	6.7[10]
$E1$	$6p_{1/2}6d_{3/2}(1)$	447	511	1.4[−1]	9.9[−2]	1.1[9]	5.0[8]	$M3$	$6p_{3/2}7p_{3/2}(3)$	277	289	1.4[2]	2.4[2]	6.6[−4]	8.5[−4]
$M3$	$6p_{1/2}5f_{7/2}(3)$	447	536	2.8[1]	6.9[1]	4.7[−6]	3.2[−6]	$M1$	$6p_{3/2}7p_{3/2}(1)$	277	289	9.8[−4]	1.1[−3]	4.2[2]	4.0[2]
$M3$	$6p_{3/2}7p_{3/2}(3)$	441	526	2.0[2]	3.7[2]	3.6[−5]	2.0[−5]	$E2$	$6p_{3/2}7p_{3/2}(2)$	274	287	1.9[0]	1.3[0]	2.8[5]	1.5[5]
$M1$	$6p_{1/2}7p_{1/2}(1)$	387	417	3.8[−7]	8.1[−7]	5.9[−2]	1.0[−1]	$E1$	$6p_{1/2}7s_{1/2}(1)$	266	289	1.8[−1]	2.6[−1]	6.6[9]	7.3[9]
$M1$	$6p_{1/2}7p_{3/2}(1)$	371	397	9.3[−3]	2.3[−2]	1.6[3]	3.4[3]	$M1$	$6p_{1/2}7p_{1/2}(1)$	231	244	8.9[−7]	1.8[−6]	6.5[−1]	1.1[0]
$E2$	$6p_{1/2}7p_{3/2}(2)$	370	399	6.4[−1]	4.3[−1]	2.1[4]	9.5[3]	$M1$	$6p_{1/2}7p_{3/2}(1)$	219	229	1.4[−2]	8.8[−3]	1.2[4]	6.6[3]

TABLE IX. Multipole ground state polarizabilities α^{Ek} and parameters β_D and γ_D calculated using the DF and RPA approximation (a.u.) in Rn-like ions with $Z = 89, 90$, and 92 . Experimental values from [4] are given in the row labeled “Expt.”

Z		α^{E1}	α^{E2}	α^{E3}	β_D	γ_D
89	RPA	10.08	42.39	344.1	4.419	1.999
89	DF	11.42	35.70	261.1	4.668	2.056
90	RPA	7.750	28.82	192.5	2.971	1.177
90	Expt.	7.720 (7)	21.5 (3.9)			
90	DF	8.957	24.54	148.5	3.209	1.240
92	RPA	5.029	15.97	74.06	1.554	0.498
92	DF	5.977	13.44	58.68	1.729	0.541

(with $n_{\max} = 50$). The reduced matrix elements in the above sum are evaluated using both the DF and RPA approximations.

In Table IX, we give ground-state multipole polarizabilities α^{Ek} and parameters β_D and γ_D calculated using the DF and RPA approximation in Rn-like ions with $Z = 89, 90$, and 92 . The correlation contributions (difference between the RPA and DF values) are about 15%. RPA is expected to give results for the dipole ground-state polarizability that are accurate to about 5% based on the comparison of the RPA values with more accurate coupled-cluster calculations [34]. We discuss this analysis in the next section. Unfortunately, such an analysis could not be carried out for the quadrupole polarizabilities. Our RPA $E1$ polarizability value for Th^{4+} is in excellent agreement with experimental result of [4]. Our DF value for the quadrupole polarizability is consistent with the experimental results, while the RPA value is somewhat larger.

VI. UNCERTAINTY ESTIMATES AND CONCLUSION

Comparison of the RMBPT1 and RMBPT2 energies and transition rates presented in Tables IV, V, VII, and VIII gives us the first rough estimates of the uncertainties of our results because it gives the second-order correlation correction. The differences in the RMBPT1 and RMBPT2 values are about 8% for energies, about 50% for the largest values of transition rates A_r , and a factor of 2 for the smallest values of A_r . The third-order corrections for the energies and transition rates were evaluated for monovalent atomic systems (see, for example, Refs. [38–40]). The differences between the RMBPT3 and RMBPT2 energies were about 2–3% for Ba II, Sr II, and Hg II ions. In addition, we evaluated the energies and wavelengths in Rn-like Ac, Th, and U ions using the COWAN codes. These results are between our RMBPT1 and RMBPT2 values (see Table IV).

To check the accuracy of our RPA results for the electric-dipole polarizability, we compare in Table X the RPA values for other relevant heavy systems with theoretical results obtained by more sophisticated coupled-cluster approaches. The calculated static polarizabilities of the doubly charged Ba and Ra at the nonrelativistic, relativistic Douglas-Kroll and Dirac-Coulomb level of theory were presented by

TABLE X. Comparisons of RPA and coupled-cluster (CC) calculations for the ground-state dipole polarizabilities of closed-shell systems. The coupled-cluster data are from Refs. [35–37]. The last three rows give values from the present work.

Ion	DF	RPA	CC	Dif.% RPA vs DF	Dif.% RPA vs CC
Xe	26.87	26.97	27.06 [37]	0.4%	−0.3%
Ba ²⁺	11.70	10.61	10.491 [35]	−10.3%	1.1%
Rn	34.42	34.99	33.18 [37]	1.6%	5.2%
Fr ⁺	21.56	20.40	20.4 (2) [36]	−5.7%	consistent
Ra ²⁺	15.19	13.79	13.361 [35]	−10.1%	3.1%
Ac ³⁺	11.42	10.08		−13.3%	
Th ⁴⁺	8.957	7.750		−15.6%	
U ⁶⁺	5.977	5.029		−18.9%	

Lim *et al.* in Ref. [35]. Recommended dipole polarizabilities of the positively charged Fr, calculated from the relativistic coupled-cluster method with singles, doubles, and perturbative triples [CCSD(T)] including spin-orbit contributions were given by Lim *et al.* in Ref. [36]. The relativistic effects for static polarizabilities α of rare gas atoms (Xe and Rn) were investigated theoretically with the third-order Douglas-Kroll method [37]. In columns “RPA” and “CC” of Table X we present results from Refs. [35–37]. In columns “RPA vs DF” and “RPA vs CC,” we give the difference of the corresponding values in percent, relative to the RPA values. From this comparison, we can expect the RPA value for the dipole polarizability to be accurate to about 5%. The difference between DF and RPA for Rn-like Ac³⁺, Th⁴⁺, and U⁶⁺ increases from 13.3% for Ac³⁺ up to 18.9% for U⁶⁺, demonstrating the importance of relativistic effects in the calculation of ground-state polarizability.

We have presented a systematic second-order relativistic MBPT study of excitation energies, reduced matrix elements, and transition rates for multipole transitions in Rn-like Ac³⁺, Th⁴⁺, and U⁶⁺ ions. Our multipole matrix elements include correlation corrections from Coulomb and Breit interactions. We determine the energies of the $6p_j5f_j(J)$, $6p_j6d_j(J)$, $6p_j7s(J)$, $6p_j7p_j(J)$, $6s5f_j(J)$, $6s6d_j(J)$, $6s7s(J)$, and $6s7p_j(J)$ excited states. Wavelengths, line strengths, and transition rates were evaluated for the 40 multipole matrix elements for transitions from excited states to the ground state. Ground-state multipole polarizabilities in Rn-like actinium, thorium, and uranium ions were calculated in the DF and RPA approximation. Our RMBPT results presented in this paper are *ab initio* calculations of energies and transition rates in Ac³⁺, Th⁴⁺, and U⁶⁺ ions. This work provides a starting point for further development of theoretical methods for such highly correlated and relativistic system.

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