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Relativistic many-body calculation of energies, oscillator strengths, transition rates, lifetimes, polarizabilities, and quadrupole moment of a Fr-like Th IV ion

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Atomic properties of the 24 low-lying ns, np_j, nd_j, nf_j, and ng_j states in Th IV ion are calculated using the high-precision relativistic all-order method where all single, double, and partial triple excitations of the Dirac-Fock wave functions are included to all orders of perturbation theory. Recommended values are provided for a large number of electric-dipole matrix elements, oscillator strengths, transition rates, and lifetimes. Scalar polarizabilities of the ground and six excited states ($5f_j$, $6d_j$, $7p_j$, and $7s$), and tensor polarizabilities of the $5f_j$, $6d_j$, and $7p_{3/2}$ states of Th IV are evaluated. The uncertainties of the recommended values are estimated. These calculations provide recommended values critically evaluated for their accuracy for a number of Th IV atomic properties for use in theoretical modeling as well as planning and analysis of various experiments including development of an ultraprecise nuclear clock and resonant excitation Stark ionization spectroscopy studies of actinide ions.

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I. INTRODUCTION

The ^{229}Th nuclear excitation energy of a few eV [1,2] presents a remarkable opportunity to develop an ultraprecise clock based on this very narrow nuclear transition [3–5]. This transition was also proposed [6] for the laboratory search for variation of the fine-structure constant and the dimensionless strong interaction parameter $m_q/\Lambda_{\rm QCD}$ due to an estimated 5–6 orders of magnitude enhancement. The energy splittings of the ground and excited states of the nuclei are generally much larger and are not accessible with laser spectroscopy. In 2009, laser cooling of the ^{229}Th^{3+} was reported by Campbell *et al.* [4]. This was the first demonstration of laser cooling of a multiply charged ion. Laser-cooled Wigner crystals ^{229}Th^{3+} were produced in a linear Paul trap [4]. These experimental advances opened an avenue for excitation of the nuclear transition in a trapped, cold ^{229}Th^{3+} ion that may lead to new levels of metrological precision [4,5,7].

While the clock based on ultraviolet ^{229}Th nuclear transition can be designed with various Th ions, Th^{3+} is particularly attractive due to its simple electronic structure of one valence electron above the closed $[\mathrm{Rn}] = [\mathrm{Xe}]4f^{14}5d^{10}6s^2 6p^6$ core. The transition probability of the Th^{229} nucleus from its lowest-energy isomeric states to the ground state due to the electronic bridge process was evaluated in [8]. Implementation of the electronic bridge process will require good understanding of Th^{3+} atomic properties, including matrix elements of the electric-dipole and hyperfine operators. A single-ion nuclear clock based on the stretched states within the $5f_{5/2}$ electronic ground states of both nuclear isomeric and ground manifolds was recently proposed in [5].

The hyperfine A and B constants for the $5f_{5/2}$, $5f_{7/2}$, $6d_{3/2}$, and $6d_{5/2}$ states were recently measured allowing one to determine nuclear electric-quadrupole moment $Q = 3.11(16)$ eb [7]. Accurate measurement of the hyperfine constants, combined with precision theoretical calculations, may be used to produce more accurate determination of the ^{229}Th nuclear magnetic moment, which is presently known to be about 10% [9]. The relative isotope shifts with respect to ^{232}Th^{3+} were measured for three $5f$–$6d$ transitions [7]. The 717-nm electric quadrupole transition was observed in [10]; the $6d_{3/2}$–$7s$ transition frequency and the lifetime of the metastable $7s$ level were measured to be 417 845 964(30) MHz and 0.60(7) s, respectively.

In 2011, binding energies of high-L Rydberg states ($L \geqslant 7$) of Th^{2+} with $n = 27$–29 were studied using the resonant excitation Stark ionization spectroscopy (RESIS) method [11]. Analysis of the observed RESIS spectra led to determination of five properties of the Th^{3+} ion: its electric quadrupole moment, adiabatic scalar and tensor dipole polarizabilities, and the dipole matrix elements connecting the ground $5f_{5/2}$ level to the low-lying $6d_{3/2}$ and $6d_{5/2}$ levels.

The optical spectroscopy has been reported for Th^{3+} [12], determining the relative positions of the lowest 24 levels, but most Th^{3+} properties of even low-lying levels are not known experimentally.

Recently, oscillator strengths and transition rates were reported by Safronova *et al.* [13,14], Migdalek *et al.* [15], and Biémont *et al.* [16]. The pseudorelativistic Hartree-Fock (HFR + CP) method including core-polarization effects was used in Ref. [16] to evaluate oscillator strengths and transition rates for the 76 transitions in the Th^{3+} ion [16]. Dirac-Fock + core-polarization approximation, where core-valence electron correlations were treated in a semiclassical core-polarization picture, was used to evaluate properties of 20 $E1$ transitions in Th IV in Ref. [15]. Excitation energies, reduced matrix elements, oscillator strengths, transition rates, scalar and tensor ground-state polarizabilities, and lifetimes for a large number of levels were calculated in [14] using the third-order many-body perturbation theory (MBPT) and single-double (SD) all-order methods.

Accurate calculations of Th^{3+} atomic properties are difficult. While it is a Fr-like ion, its level structure is different from both Fr and Ra^+, which both have $7s$ ground state. Th^{3+} ground state is $5f_{5/2}$ causing further difficulties in the accurate calculation of its properties not present in either Fr or Ra^+. Moreover, Th^{3+} is sufficiently multicharged to make Breit contributions significant. Most of the theoretical and experimental high-precision studies involved ns, np, and nd levels resulting in lack of benchmarks for nf state properties in other systems that may be used to further improve *ab initio* calculations. The study of the electronic bridge process [8] noted rather poor agreement of theoretical and experimental energies.

Therefore, we calculate properties of the Th^{3+} ion by several different approaches to study the correlation contributions to various properties to evaluate accuracy of our calculations and to provide a pathway to further improvement in theoretical understanding of this ion. Due to the above noted interesting applications, Th^{3+} also represents an excellent benchmark for further development of high-precision methodologies of very heavy ions.

In the present work, we evaluated all properties using both SD and single-double partial triple (SDpT) all-order methods as well as carried out additional scaling to evaluate dominant missing correlation corrections and evaluate uncertainties of our calculations. The SD and SDpT methods and their application were discussed in a review [17] and references therein. Energies and lifetimes are calculated for the ns ($n = 7$–10), np ($n = 7$–8), nd ($n = 6$–8), nf ($n = 5$–7), and ng ($n = 5$–6) states. Reduced matrix elements, oscillator strengths, and transition rates are calculated for allowed electric-dipole transitions between these states. Scalar polarizabilities of the seven first $5f_j$, $6d_j$, $7p_j$, and $7s$ states, and tensor polarizabilities of the $5f_j$, $6d_j$, and $7p_{3/2}$ states of Th^{3+} are evaluated. Particular care was taken to accurately treat contributions from highly excited states. The present calculation of the transition rates, lifetimes, and polarizabilities required accurate representation of rather highly excited states, such as $7l_j$, leading to the use of the large $R = 100$ a.u. cavity for the generation of the finite B-spline basis set [18] and higher number of splines $N = 70$ to produce high-accuracy single-particle orbitals. The methods for evaluating the uncertainties of theoretical values calculated in the framework of the all-order approach are discussed. The calculation of uncertainties involved estimation of missing high-order effects and *ab initio* calculations in different approximations to establish the size of the higher-order corrections and to approximate missing contributions.

II. ENERGY LEVELS AND TRANSITION PROPERTIES

A. Energy levels

The calculation of energies in Th^{3+} was discussed in detail by Safronova *et al.* [14] where the third-order relativistic many-body perturbation theory (RMBPT) and all-order SD energies were presented. The third-order RMBPT approximation includes the second-order and third-order part of the correlation energies. The all-order SD approximation includes the second-order and the single-double part of the higher-order correlation energies. However, it is missing the part of the third-order contribution $E^{(3)}_{\mathrm{extra}}$. The additional third-order contribution to the energies was added in [14] using a separate calculation. The inclusion of the partial triple-excitations terms via the SDpT method described in [17] and implemented in the present work automatically includes the missing third-order energy. The data in [14] show extremely large contributions of the correlation corrections into the energy values. In fact, the lowest-order Dirac-Fock calculation gives a $6d_{3/2}$ ground state instead of the $5f_{5/2}$ state. We find that triple excitations beyond the third-order term $E^{(3)}_{\mathrm{extra}}$ are very large—3%–5% of the total correlation correction. For example, the difference of the correlation correction to the ionization potential calculated in the SD approximation $+\,E^{(3)}_{\mathrm{extra}}$ term and the SDpT value is 1200 cm^{-1}. Based on the size of all other corrections and experimental values, we estimate that the omitted triple and higher effects for the $5f$ and $6d$ state are on the order of already included triple excitations, which is a reasonable expectation of the accuracy in this case. The relative contribution of the correlations is substantially higher (by at least a factor of 2) for the $5f$ states than for all other states exacerbating the problem for the transition energies. Therefore, full inclusion of the triple excitations, and most likely an estimate of the higher excitations, would be required for accurate description of the energy-level differences with the ground state owing to significant imbalance of the correlation contribution between the ground state and all other states except $5f_{7/2}$. Our values for the fine-structure $5f_{5/2}$–$5f_{7/2}$ interval, 4165 cm^{-1}, is in good agreement with experiment, 4325 cm^{-1}. We included the Breit interaction on the same footing as the Coulomb interaction in the basis set, which incorporates high-order Breit effects. The Breit interaction was included to second order in [14], which significantly overestimates its correction. We also note that inclusion of the higher partial waves with $l > 6$ is very important for accurate description of the $5f$ states. The contribution of $l > 6$ is on the order of 1000 cm^{-1} for the $5f$ states and 250–300 cm^{-1} for the $6d$ states. We use experimental energy intervals in the calculation of all transitions properties and polarizabilities below, where available.

B. Electric-dipole matrix elements

In Table I, we list our recommended values for $E1$ $n'p$–ns, nd–$n'p$, nd–$n'f$, and ng–$n'f$ transitions. We note that we have calculated about 80 $E1$ matrix elements to consider all dipole transitions between ns, np, $n'd$, $n''f$, and $n''g$ states with $n = 7$–10, $n' = 6$–8, and $n'' = 5$–7. We refer to these values as the "best set" of the matrix elements. We list only the matrix elements that give significant contributions to the atomic properties calculated in the other sections. To evaluate the uncertainties of these values, we carried out several calculations in different approximations. To demonstrate the size of the second-, third-, and higher-order correlation corrections, we list the lowest-order Dirac-Fock (DF) Z^{DF}, second-order $Z^{(\mathrm{DF}+2)}$, and third-order $Z^{(\mathrm{DF}+2+3)}$ values in the first three numerical columns of Table I. The absolute values in atomic units (a_0e) are given in all cases. The third-order MBPT calculations are carried out following the method described in Ref. [19]. The $Z^{(\mathrm{DF}+2)}$ values are obtained as the sum of the second-order correlation correction

TABLE I. Recommended values of the reduced electric-dipole matrix elements in Th IV in atomic units. The first-order, second-order, third-order MBPT, and all-order SD and SDpT values are listed; the label "sc" indicates the scaled values. Absolute values are given. Final recommended values and their uncertainties are given in the Z^{final} column. The last column gives relative uncertainties of the final values in %.

Transition		Z^{DF}	$Z^{(\text{DF}+2)}$	$Z^{(\text{DF}+2+3)}$	Z^{SD}	$Z^{(\text{SD})}_{\text{sc}}$	Z^{SDpT}	$Z^{\text{SDpT}}_{\text{sc}}$	Z^{final}	Unc. (%)
$7p_{1/2}$	$6d_{3/2}$	2.5465	2.1960	2.0566	2.1220	2.1284	2.1312	2.1253	2.122(30)	1.4
$7p_{1/2}$	$7d_{3/2}$	3.8261	3.5613	3.4020	3.4490	3.4642	3.4635	3.4638	3.449(26)	0.8
$7p_{1/2}$	$7s_{1/2}$	2.8994	2.4738	2.3476	2.4197	2.4368	2.4323	2.4364	2.420(34)	1.4
$7p_{1/2}$	$8s_{1/2}$	1.5874	1.6297	1.5549	1.5492	1.5404	1.5542	1.5390	1.549(19)	1.3
$7p_{1/2}$	$9s_{1/2}$	0.4722	0.4910	0.4734	0.4657	0.4630	0.4667	0.4641	0.463(5)	1.1
$7p_{3/2}$	$6d_{3/2}$	0.9963	0.8823	0.8260	0.8488	0.8516	0.8533	0.8501	0.849(10)	1.2
$7p_{3/2}$	$6d_{5/2}$	3.1975	2.8762	2.6900	2.7550	2.7627	2.7665	2.7583	2.755(31)	1.1
$7p_{3/2}$	$7d_{3/2}$	2.0308	1.8920	1.8252	1.8445	1.8522	1.8503	1.8523	1.845(13)	0.7
$7p_{3/2}$	$7d_{5/2}$	5.9481	5.5572	5.3603	5.4192	5.4409	5.4377	5.4415	5.419(37)	0.7
$7p_{3/2}$	$7s_{1/2}$	3.9933	3.4515	3.2731	3.3677	3.3925	3.3866	3.3919	3.368(44)	1.3
$7p_{3/2}$	$8s_{1/2}$	3.0768	3.0702	2.9863	2.9756	2.9635	2.9804	2.9623	2.963(24)	0.8
$7p_{3/2}$	$9s_{1/2}$	0.7567	0.7439	0.7157	0.7123	0.7197	0.7141	0.7216	0.712(10)	1.5
$8p_{1/2}$	$7d_{3/2}$	5.4788	5.3607	5.1639	5.1791	5.1866	5.1907	5.1838	5.179(11)	0.2
$8p_{1/2}$	$8s_{1/2}$	5.0325	4.8413	4.6814	4.7280	4.7590	4.7405	4.7579	4.728(21)	0.5
$8p_{3/2}$	$7d_{3/2}$	2.1716	2.1485	2.0566	2.0630	2.0641	2.0690	2.0628	2.064(05)	0.2
$8p_{3/2}$	$7d_{5/2}$	6.8642	6.7805	6.5104	6.5180	6.5247	6.5343	6.5209	6.525(10)	0.2
$8p_{3/2}$	$8s_{1/2}$	6.7737	6.5493	6.3192	6.3881	6.4284	6.4068	6.4270	6.388(57)	0.9
$8p_{3/2}$	$9s_{1/2}$	5.4177	5.4364	5.3083	5.2854	5.2662	5.2961	5.2631	5.266(30)	0.6
$5f_{5/2}$	$6d_{3/2}$	2.4281	1.6597	1.3609	1.5296	1.5330	1.5423	1.5231	1.530(63)	4.1
$5f_{5/2}$	$6d_{5/2}$	0.6391	0.4586	0.3685	0.4116	0.4125	0.4154	0.4100	0.412(16)	3.9
$5f_{5/2}$	$5g_{7/2}$	1.1236	0.8404	0.6123	0.6895	0.6544	0.7034	0.6555	0.690(30)	4.4
$5f_{5/2}$	$7d_{3/2}$	0.0654	0.3417	0.2077	0.2588	0.2475	0.2710	0.2449	0.259(26)	9.9
$5f_{5/2}$	$7d_{5/2}$	0.0048	0.0671	0.0362	0.0521	0.0492	0.0554	0.0486	0.052(8)	15
$5f_{7/2}$	$6d_{5/2}$	2.9557	2.1257	1.7270	1.9191	1.9223	1.9371	1.9122	1.919(73)	3.8
$5f_{7/2}$	$5g_{7/2}$	0.2298	0.1760	0.1292	0.1478	0.1407	0.1506	0.1408	0.148(6)	3.9
$5f_{7/2}$	$5g_{9/2}$	1.3635	1.0667	0.7894	0.8855	0.8487	0.9022	0.8484	0.885(33)	3.8
$5f_{7/2}$	$7d_{5/2}$	0.0703	0.3889	0.2259	0.2961	0.2815	0.3114	0.2785	0.296(35)	12
$6f_{5/2}$	$6d_{3/2}$	2.6761	2.3276	2.3423	2.3443	2.3181	2.3372	2.3220	2.344(23)	1.0
$6f_{5/2}$	$6d_{5/2}$	0.7669	0.6780	0.6837	0.6800	0.6727	0.6774	0.6736	0.680(6)	0.9
$6f_{7/2}$	$6d_{5/2}$	3.3539	2.9806	3.0233	3.0008	2.9678	2.9886	2.9709	3.001(25)	0.8
$7f_{5/2}$	$8d_{3/2}$	13.4659	13.2664	12.6363	12.5835	12.7110	12.6315	12.6803	12.71(13)	1.0
$7f_{5/2}$	$8d_{5/2}$	3.5961	3.5454	3.3681	3.3554	3.3900	3.3553	3.3662	3.390(35)	1.0
$7f_{5/2}$	$7d_{5/2}$	1.0359	1.0451	1.1332	1.1178	1.0853	1.1014	1.0845	1.085(16)	1.5
$7f_{5/2}$	$6d_{3/2}$	1.2888	0.9357	0.8390	0.8131	0.8224	0.8260	0.8297	0.813(33)	4.1
$7f_{5/2}$	$6d_{5/2}$	0.3586	0.2713	0.2376	0.2286	0.2321	0.2323	0.2340	0.229(09)	4.0
$7f_{5/2}$	$5g_{7/2}$	8.5105	8.5592	8.6347	8.3981	8.4526	8.4008	8.4515	8.453(52)	0.6
$7f_{7/2}$	$5g_{9/2}$	9.5778	9.6340	9.7290	9.4935	9.4490	9.4944	9.4466	9.449(45)	0.5
$7f_{7/2}$	$5g_{7/2}$	1.6217	1.6312	1.6446	1.5971	1.5952	1.5988	1.5950	1.595(4)	0.2
$7f_{7/2}$	$6d_{5/2}$	1.5854	1.2543	1.1202	1.0734	1.0900	1.0872	1.0966	1.073(36)	3.3

$Z^{(2)}$ and the DF matrix elements Z^{DF}. The third-order matrix elements $Z^{(\text{DF}+2+3)}$ include the DF values, the second-order $Z^{(2)}$ results, and the third-order $Z^{(3)}$ correlation correction. $Z^{(3)}$ includes random-phase-approximation (RPA) terms iterated to all orders, Brueckner orbital corrections, the structural radiation, and normalization terms (see [19] for definition of these terms). The next four columns give the results of four different all-order calculations. *Ab initio* electric-dipole matrix elements evaluated in the all-order SD (single-double) and SDpT approximations (single-double all-order method including partial triple excitations [20]) are given in columns labeled Z^{SD} and Z^{SDpT} of Table I. The SD and SDpT matrix elements Z^{SD} include $Z^{(3)}$ completely, along with important fourth- and higher-order corrections. The difference between the Z^{SD} and Z^{SDpT} values is about 0.2%–2%, i.e., the effect of the triple excitations on the values of matrix elements is significantly smaller than for the energies.

The last column of Table I gives relative uncertainties of the final values Z^{final} in %. We use two different methods for the estimation of the uncertainties based on the type of the dominant correlation corrections for a specific transition. If the correlation terms containing valence single-excitation coefficients are dominant, the omitted correlation corrections can be estimated by a scaling procedure described, for example, in Ref. [21]. In this case, we use a well-defined and rather accurate procedure for the evaluation of the uncertainty of the matrix elements described in detail in [21–23]. It is based on four different all-order calculations that included two *ab initio* all-order calculations with (SDpT) and without (SD) the inclusion of the partial triple excitations and two calculations that included a semiempirical estimate of high-order correlation corrections starting from both *ab initio* runs, $\mathrm{SD}_{\mathrm{sc}}$ and $\mathrm{SDpT}_{\mathrm{sc}}$. The differences of these four values were used to estimate uncertainty in the final result for each transition and the SD scaled values are taken as final for these cases. We note that the scaling may be less reliable in Th^{3+} than in other systems due to large uncertainty in the experiment ionization potential 231 065(200) cm^{-1} [24] as scaling relies on the experimental values of the removal energies. However, *ab initio* SDpT results are generally already close to the final scaled values.

Unfortunately, a different type of the correlation terms is dominant for a large fraction of the transitions of interest for this work (including all of the transitions containing the $5f$ states). In these cases, the above strategy for evaluating uncertainties is expected to underestimate the uncertainties. We have developed a different approach for these cases using the study of uncertainties in a similar reference ion, Rb-like Y, where the above (scaling) procedure is expected to work well [23]. We have compared the estimated uncertainties for 60 nd–$n'f$ transitions [23] with the size of the correlation corrections for the same transitions. We find that, *on average*, the estimate uncertainty was about 7% of the correlation correction, which was calculated as the difference of the all-order and the lowest-order results. Therefore, we use 7% of the correlation correction to estimate the uncertainties as the second method for evaluating the uncertainties and list these uncertainties for transitions where the first method is not expected to produce reliable results. We note that the second method is less precise than the first one and provides a rough estimate of the accuracy. The *ab initio* SD data are listed as final for these transitions. An accurate benchmark reference measurement is needed to improve the accuracy estimates.

We find three cases in Table I where neither of the two methods is expected to provide accurate estimates of the uncertainties. For all three $5f$–$7d$ transitions, the lowest-order values are less than 0.1 a.u. and almost entire values come from the correlation correction. In these cases, we took 50% of the entire higher-order correction (calculated as the difference of the all-order and the third-order values) as the uncertainty. The 50% was chosen based on the comparison of the higher-order effects for other transitions with the corresponding estimates of their uncertainties carried out by the other methods.

We find that the uncertainties are 0.2%–2% for most of the transitions. Larger uncertainties occur for some of the transitions with large correlation contributions such as $5f$–ng. Our final results and their uncertainties are used to calculate the recommended values of the transition rates, oscillator strengths, lifetimes, and the polarizabilities, as well as to evaluate the uncertainties of these results.

C. Transition rates and oscillator strengths

We combine experimental energies [12] and our final values of the best set matrix elements to calculate transition rates A and oscillator strengths f. The transition rates are calculated using

$$A_{ab} = \frac{2.026\,13 \times 10^{18}}{\lambda^3} \frac{S}{2j_a + 1} \mathrm{s}^{-1}, \tag{1}$$

where the wavelength λ is in Å and the line strength $S = d^2$ is in atomic units.

Transition rates A (s^{-1}) for the 60 ns–np, np–nd, nd–nf, and nf–ng transitions are given in Table II. Vacuum wavelengths obtained from experimental energies [12] are also listed for reference. The relative uncertainties of the transition rates listed in the column labeled "Unc."are twice the corresponding matrix element uncertainties since the transition rates are proportional to the squares of the matrix elements. The smallest uncertainties are for the $5g$–$7f$ transitions, while the largest ones are for the $6d$–$8p$ and $5f$–$8d$ transitions owing to large corresponding uncertainties in the $E1$ transition matrix elements. We already discussed the importance of the size of the correlation effects for the dipole matrix element uncertainties. For example, the DF value for the $5f_{5/2}$–$7d_{3/2}$ transition (see Table I) is smaller than the all-order SD value by a factor of 4. The SDpT value obtained with including partial triple excitations is larger than the SD value by 4.7% for this transition. The scaling procedure decreases both SD and SDpT values by 4.3% and 9.6%, respectively. The contributions affected by scaling are related to the correlation potential and, therefore, the values of the correlation energies for the specific state. The scaling coefficients are obtained as a ratio of the "experimental" correlation energy (obtained as the difference of the experimental values and the lowest-order results) and the theoretical SD or SDpT correlation energies. Lower accuracy of the theoretical correlation energy leads to a larger scaling effect owing to a larger omitted correlation contribution to the matrix elements of the certain class, in particular for weaker transition with small DF values.

We present weighted oscillator strengths gf calculated using our recommended values of reduced electric-dipole matrix elements gf^{final} and their uncertainties in Table III. The relative uncertainties are listed in column "Unc." in %. In columns "DF," we list gf values calculated in the DF approximation. In column "Expt.," we list λ recommended by compilation in Ref. [12]. In column "HFR + CP," we list gf values calculated by the HFR + CP method [16]. In the left column of Table III, we list the 32 transitions when the "HFR + CP" values are in better agrement with our "Final" result. Disagreement between the $gf^{\mathrm{HFR+CP}}$ and gf^{final} values is about 2%–20%. The 15 transitions given in the right column of Table III are transitions when the "HFR + CP" values are in better agreement with our gf values obtained in the DF approximation than with our "Final" result. The last 16 transitions given in the right column of Table III present transitions when the "HFR + CP" values disagree with the DF values as well as with the "Final" values by a factor of 2–10 for most of the transitions. It should be noted that among these 16 transitions there are at least 10 transitions with very small gf values (10^{-2}). The uncertainties for such transitions are significantly larger than the ones for the other transitions shown in Table III.

D. Lifetimes

We list the lifetimes of the (7–9)s, (7–8)p, (6–8)d, (5–7)f, and (5–6)g states in Table IV. These values are obtained using the transition rates listed in Table II. The uncertainties in the lifetime values given in parentheses are

TABLE II. Wavelengths λ (Å) and transition rates A_r (s^{-1}) for transitions in Th IV calculated using our recommended values of reduced electric-dipole matrix elements A_r^{final} and their uncertainties. The relative uncertainties are listed in column "Unc." in %. In columns λ, we list experimental data [12]. Numbers in brackets represent powers of 10.

Transition		λ	A_r	Unc.	Transition		λ	A_r	Unc.	Transition		λ	A_r	Unc.
Lower	Upper	(Å)	(s^{-1})	(%)	Lower	Upper	(Å)	(s^{-1})	(%)	Lower	Upper	(Å)	(s^{-1})	(%)
$5f_{5/2}$	$8d_{5/2}$	615.28	3.08[6]	12	$7p_{3/2}$	$9s_{1/2}$	1140.61	3.46[8]	3.0	$8p_{3/2}$	$8d_{3/2}$	4528.35	5.21[7]	0.5
$5f_{5/2}$	$8d_{3/2}$	617.46	9.47[7]	14	$6d_{3/2}$	$7p_{3/2}$	1565.86	9.51[7]	2.4	$8p_{3/2}$	$9s_{1/2}$	4794.51	2.55[8]	1.1
$5f_{5/2}$	$5g_{7/2}$	627.39	4.88[8]	8.8	$7p_{1/2}$	$7d_{3/2}$	1682.21	1.27[9]	1.5	$8s_{1/2}$	$8p_{3/2}$	4938.44	1.72[8]	1.8
$5f_{7/2}$	$8d_{5/2}$	632.10	8.49[7]	14	$7p_{1/2}$	$8s_{1/2}$	1684.00	5.09[8]	2.5	$7d_{3/2}$	$8p_{3/2}$	4953.85	1.78[7]	0.5
$6d_{3/2}$	$7f_{5/2}$	643.66	8.37[8]	8.2	$6d_{5/2}$	$7p_{3/2}$	1707.37	7.72[8]	2.2	$7d_{5/2}$	$8p_{3/2}$	5421.88	1.35[8]	0.3
$5f_{7/2}$	$5g_{7/2}$	644.89	2.06[7]	7.8	$6d_{3/2}$	$7p_{1/2}$	1959.02	6.07[8]	2.8	$7f_{5/2}$	$6g_{7/2}$	5841.02	2.22[8]	5.3
$5f_{7/2}$	$5g_{9/2}$	644.97	5.92[8]	7.6	$7s_{1/2}$	$7p_{3/2}$	2003.00	7.15[8]	2.6	$7f_{7/2}$	$6g_{7/2}$	6018.30	7.90[6]	3.0
$6d_{5/2}$	$7f_{7/2}$	664.13	9.96[8]	6.7	$7p_{3/2}$	$7d_{5/2}$	2067.35	1.12[9]	1.4	$7f_{7/2}$	$6g_{9/2}$	6018.66	2.21[8]	2.6
$6d_{5/2}$	$7f_{5/2}$	666.36	5.96[7]	8.0	$8p_{1/2}$	$10s_{1/2}$	2086.62	6.81[7]	2.7	$8s_{1/2}$	$8p_{1/2}$	6713.71	7.48[7]	0.9
$6d_{3/2}$	$8p_{3/2}$	765.24	2.29[7]	7.4	$7p_{3/2}$	$7d_{3/2}$	2144.60	1.75[8]	1.4	$7d_{3/2}$	$8p_{1/2}$	6742.22	8.87[7]	0.4
$6d_{5/2}$	$8p_{3/2}$	797.55	1.84[8]	7.3	$7p_{3/2}$	$8s_{1/2}$	2147.50	8.98[8]	1.6	$5f_{5/2}$	$6d_{5/2}$	6903.05	1.74[5]	7.7
$6d_{3/2}$	$8p_{1/2}$	797.94	7.24[7]	16	$7d_{3/2}$	$7f_{5/2}$	2228.66	4.22[8]	3.4	$5f_{7/2}$	$6d_{5/2}$	9841.58	1.30[6]	7.6
$7p_{1/2}$	$10s_{1/2}$	818.32	1.14[8]	0.7	$7d_{5/2}$	$7f_{7/2}$	2291.90	4.51[8]	7.2	$5f_{5/2}$	$6d_{3/2}$	10 877.60	9.21[5]	8.2
$6d_{3/2}$	$6f_{5/2}$	846.91	3.06[9]	2.0	$7d_{5/2}$	$7f_{5/2}$	2318.70	3.19[7]	3.0	$7d_{3/2}$	$6f_{5/2}$	13 184.61	7.34[6]	3.0
$6d_{5/2}$	$6f_{7/2}$	882.39	3.32[9]	1.6	$8p_{3/2}$	$10s_{1/2}$	2349.07	1.08[8]	1.4	$7d_{5/2}$	$6f_{7/2}$	15 653.86	4.76[6]	3.0
$6d_{5/2}$	$6f_{5/2}$	886.66	2.24[8]	1.8	$7s_{1/2}$	$7p_{1/2}$	2694.81	3.03[8]	2.8	$7d_{5/2}$	$6f_{5/2}$	17 117.13	2.38[5]	3.1
$7s_{1/2}$	$8p_{1/2}$	897.78	7.71[7]	11	$6f_{5/2}$	$5g_{7/2}$	3113.25	6.75[8]	3.0	$5g_{9/2}$	$7f_{7/2}$	17 580.87	4.16[6]	1.0
$7p_{3/2}$	$10s_{1/2}$	914.20	1.71[8]	1.6	$6f_{7/2}$	$5g_{7/2}$	3167.09	2.42[7]	2.9	$5g_{7/2}$	$7f_{7/2}$	17 639.80	1.17[5]	0.5
$7p_{1/2}$	$8d_{3/2}$	983.14	2.93[8]	4.1	$6f_{7/2}$	$5g_{9/2}$	3169.00	6.76[8]	2.9	$5g_{7/2}$	$7f_{5/2}$	19 362.21	3.32[6]	1.2
$7p_{1/2}$	$9s_{1/2}$	995.13	2.20[8]	2.3	$8p_{1/2}$	$8d_{3/2}$	3644.65	3.43[8]	9.4	$8d_{3/2}$	$7f_{5/2}$	38 451.19	9.60[5]	2.0
$7p_{3/2}$	$8d_{5/2}$	1117.67	1.81[8]	5.0	$8p_{1/2}$	$9s_{1/2}$	3815.10	1.42[8]	2.5	$8d_{5/2}$	$7f_{7/2}$	39 506.95	9.65[5]	3.1
$7p_{3/2}$	$8d_{3/2}$	1124.88	2.27[7]	6.0	$8p_{3/2}$	$8d_{5/2}$	4413.67	3.18[8]	0.5	$8d_{5/2}$	$7f_{5/2}$	49 336.43	3.23[4]	2.1

obtained from the uncertainties in the matrix elements. The column "Unc." gives relative uncertainties of the final values in %. We also list the lifetime values calculated in DF approximation in Table IV to show the size of the correlation correction for each case. The energies recommended by Blase and Wyart [12] are given in column "Energy." Our values are compared with the SD calculation of Ref. [14]. The difference with the present results is due to more complete inclusion of the correlation correction in the present work.

In 2012, the lifetime of the metastable $7s$ level has been measured to be 0.60(7) s, which is in excellent agreement with our earlier prediction of 0.59 s [14]. The $7s$–$6d_{3/2}$ and $7s$–$6d_{5/2}$ $E2$ transitions give the only significant contributions to the $7s$ lifetime. In this work, we have carried out additional SDpT and scaled calculations of these values and obtained 7.110(47) a.u. and 9.211(59) a.u. for the $7s$–$6d_{3/2}$ and $7s$–$6d_{5/2}$ $E2$ reduced matrix elements, respectively. Our final value of the $7s$ lifetime is 0.570(8) s.

III. SCALAR AND TENSOR EXCITED-STATE POLARIZABILITIES

The valence scalar $\alpha_0(v)$ and tensor $\alpha_2(v)$ polarizabilities of an excited state v of Th IV are given by

$$\alpha_0(v) = \frac{2}{3(2j_v+1)} \sum_{nlj} \frac{|\langle v||rC_1||nlj\rangle|^2}{E_{nlj}-E_v}, \qquad (2)$$

$$\alpha_2(v) = (-1)^{j_v} \sqrt{\frac{40 j_v(2j_v-1)}{3(j_v+1)(2j_v+1)(2j_v+3)}} \times \sum_{nlj} (-1)^j \begin{Bmatrix} j_v & 1 & j \\ 1 & j_v & 2 \end{Bmatrix} \frac{|\langle v||rC_1||nlj\rangle|^2}{E_{nlj}-E_v}, \qquad (3)$$

where $C_1(\hat{r})$ is a normalized spherical harmonic and the sum over nlj runs over all states with allowed electric-dipole transitions to a state v [25]. The reduced matrix elements in the dominant contributions to the above sum are evaluated using our final values of the dipole matrix elements and available experimental energies [12]. The uncertainties in the polarizability contributions are obtained from the uncertainties in the matrix elements. We use theoretical SD energies and SD wave functions to evaluate terms with $n < 26$ in Eqs. (2) and (3). The remaining contributions to α_0 and α_2 from orbitals with $27 \leqslant n \leqslant 70$ are evaluated in the RPA approximation since the contributions from these terms are smaller than 0.01% in all cases. These terms are grouped together as "Tail." Their uncertainty is estimated as the difference of the corresponding DF and RPA values. We note that the ng states with $n > 18$, nf states with $n > 19$, nd states with $n > 20$, and np, ns states with $n > 21$ have positive energies and provide a discrete representation of the continuum in our basis.

We list the contributions to the scalar polarizabilities of the $5f_j$, $6d_j$, $7p_j$, and $7s$ states and tensor polarizabilities of the $5f_j$, $6d_j$, and $7p_{3/2}$ states in Table V. The dominant contributions are listed separately. The remaining contributions are grouped together. For example, "$nd_{3/2}$" contribution includes

TABLE III. Wavelengths λ (Å) and weighted oscillator strengths gf for transitions in Th IV calculated using our recommended values of reduced electric-dipole matrix elements and their uncertainties. The relative uncertainties are listed in column "Unc." in %. In columns "DF," we list f values calculated in DF approximation. In columns "Expt.," we list experimental λ values [12]. In column "HFR + CP," we list f values calculated by the HFR + CP method [16]. Numbers in brackets represent powers of 10.

Transition		λ	Oscillator strengths			Unc.	Transition		λ	Oscillator strengths			Unc.
Low	Upper	Expt.	DF	HFR + CP	Final	(%)	Low	Upper	Expt.	DF	HFR + CP	Final	(%)
$6d_{5/2}$	$7f_{7/2}$	664.13	1.15[0]	5.4[−1]	5.27[−1]	6.7	$5f_{5/2}$	$5g_{7/2}$	627.39	6.11[−1]	4.8[−1]	2.30[−1]	8.8
$6d_{5/2}$	$7f_{5/2}$	666.36	5.86[−2]	2.7[−2]	2.38[−2]	8.0	$7p_{3/2}$	$10s_{1/2}$	914.20	5.35[−2]	5.6[−2]	4.30[−2]	1.6
$6d_{3/2}$	$6f_{5/2}$	846.91	2.57[0]	1.9[0]	1.97[0]	2.0	$7p_{3/2}$	$9s_{1/2}$	1140.61	1.52[−1]	1.5[−1]	1.35[−1]	3.0
$6d_{5/2}$	$6f_{7/2}$	882.39	3.87[0]	2.6[0]	3.10[0]	1.6	$7p_{1/2}$	$7d_{3/2}$	1682.21	2.64[0]	2.7[0]	2.15[0]	1.5
$6d_{5/2}$	$6f_{5/2}$	886.66	2.02[−1]	1.3[−1]	1.58[−1]	1.8	$8p_{1/2}$	$10s_{1/2}$	2086.62	9.16[−2]	1.1[−1]	8.89[−2]	2.7
$7p_{1/2}$	$8d_{3/2}$	983.14	2.85[−1]	1.4[−1]	1.70[−1]	4.1	$7d_{5/2}$	$7f_{7/2}$	2291.90	2.69[0]	2.6[0]	2.84[0]	7.2
$6d_{3/2}$	$7p_{3/2}$	1565.86	1.93[−1]	1.5[−1]	1.40[−1]	2.4	$7d_{5/2}$	$7f_{5/2}$	2318.70	1.41[−1]	1.3[−1]	1.54[−1]	3.0
$6d_{5/2}$	$7p_{3/2}$	1707.37	1.82[0]	1.2[0]	1.35[0]	2.2	$8p_{3/2}$	$10s_{1/2}$	2349.07	1.87[−1]	1.9[−1]	1.79[−1]	1.4
$6d_{3/2}$	$7p_{1/2}$	1959.02	1.00[0]	6.0[−1]	6.98[−1]	2.8	$8p_{1/2}$	$8d_{3/2}$	3644.65	2.96[0]	3.5[0]	2.73[0]	9.4
$7s_{1/2}$	$7p_{3/2}$	2003.00	2.42[0]	1.6[0]	1.72[0]	2.6	$7d_{3/2}$	$8p_{3/2}$	4953.85	2.89[−1]	3.0[−1]	2.61[−1]	0.5
$7p_{3/2}$	$7d_{5/2}$	2067.35	5.20[0]	4.0[0]	4.32[0]	1.4	$7d_{5/2}$	$8p_{3/2}$	5421.88	2.64[0]	2.5[0]	2.39[0]	0.3
$7p_{3/2}$	$7d_{3/2}$	2144.60	5.84[−1]	4.3[−1]	4.82[−1]	1.4	$7f_{5/2}$	$6g_{7/2}$	5841.02	1.11[1]	1.0[1]	9.07[0]	5.3
$7p_{3/2}$	$8s_{1/2}$	2147.50	1.34[0]	1.1[0]	1.24[0]	1.6	$5f_{5/2}$	$6d_{5/2}$	6903.05	1.80[−2]	1.6[−2]	7.45[−3]	7.7
$7d_{3/2}$	$7f_{5/2}$	2228.66	1.68[0]	1.9[0]	1.88[0]	3.4	$5f_{7/2}$	$6d_{5/2}$	9841.54	2.70[−1]	2.2[−1]	1.14[−1]	7.6
$7s_{1/2}$	$7p_{1/2}$	2694.81	9.48[−1]	6.0[−1]	6.60[−1]	2.8	$5f_{5/2}$	$6d_{3/2}$	10 877.55	1.65[−1]	1.4[−1]	6.53[−2]	8.2
$6f_{5/2}$	$5g_{7/2}$	3113.25	9.64[0]	7.9[0]	7.85[0]	3.0							
$6f_{7/2}$	$5g_{7/2}$	3167.09	3.55[−1]	2.9[−1]	2.91[−1]	2.9	$5f_{5/2}$	$8d_{3/2}$	617.46	4.24[−6]	1.8[−3]	2.17[−2]	14
$6f_{7/2}$	$5g_{9/2}$	3169.00	1.24[1]	1.0[1]	1.02[1]	2.9	$5f_{7/2}$	$8d_{5/2}$	632.10	3.23[−5]	2.6[−3]	3.05[−2]	14
$8p_{3/2}$	$8d_{5/2}$	4413.67	6.00[0]	5.2[0]	5.57[0]	0.5	$6d_{3/2}$	$7f_{5/2}$	643.66	7.84[−1]	3.9[−1]	3.12[−1]	8.2
$8p_{3/2}$	$8d_{3/2}$	4528.35	6.91[−1]	5.6[−1]	6.41[−1]	0.5	$5f_{7/2}$	$5g_{7/2}$	644.89	2.49[−2]	1.7[−2]	1.03[−2]	7.8
$8p_{3/2}$	$9s_{1/2}$	4794.51	1.86[0]	1.5[0]	1.76[0]	1.1	$5f_{7/2}$	$5g_{9/2}$	644.97	8.75[−1]	6.0[−1]	3.69[−1]	7.6
$8s_{1/2}$	$8p_{3/2}$	4938.44	2.82[0]	2.6[0]	2.51[0]	1.8	$6d_{3/2}$	$8p_{3/2}$	765.24	1.87[−2]	5.9[−3]	8.04[−3]	7.4
$7f_{7/2}$	$6g_{7/2}$	6018.30	4.07[−1]	3.7[−1]	3.43[−1]	3.0	$6d_{5/2}$	$8p_{3/2}$	797.55	1.62[−1]	5.0[−2]	7.00[−2]	7.3
$7f_{7/2}$	$6g_{9/2}$	6018.66	1.43[1]	1.3[1]	1.20[1]	2.6	$6d_{3/2}$	$8p_{1/2}$	797.94	6.32[−2]	2.8[−2]	1.38[−2]	16
$8s_{1/2}$	$8p_{1/2}$	6713.71	1.15[0]	9.5[−1]	1.01[0]	0.9	$7p_{1/2}$	$10s_{1/2}$	818.32	2.52[−2]	3.1[−2]	2.29[−2]	0.7
$7d_{3/2}$	$8p_{1/2}$	6742.22	1.35[0]	1.1[0]	1.21[0]	0.4	$5f_{5/2}$	$7d_{5/2}$	823.54	8.49[−6]	1.7[−4]	1.00[−3]	31
$7p_{3/2}$	$8d_{5/2}$	1117.67	3.75[−1]	2.3[−1]	2.03[−1]	5.0	$5f_{5/2}$	$7d_{3/2}$	835.53	1.56[−3]	2.3[−3]	2.43[−2]	20
$7d_{3/2}$	$6f_{5/2}$	13 184.61	1.41[0]	1.2[0]	1.15[0]	3.0	$5f_{7/2}$	$7d_{5/2}$	853.96	1.76[−3]	3.2[−3]	3.12[−2]	24
$7d_{5/2}$	$6f_{7/2}$	15 653.86	1.71[0]	1.4[0]	1.40[0]	3.0	$7s_{1/2}$	$8p_{1/2}$	897.78	1.08[−3]	2.1[−3]	1.86[−2]	11
$7d_{5/2}$	$6f_{5/2}$	17 117.13	7.72[−2]	6.5[−2]	6.28[−2]	3.1	$7p_{1/2}$	$9s_{1/2}$	995.13	6.81[−2]	8.7[−2]	6.54[−2]	2.3
$5g_{9/2}$	$7f_{7/2}$	17 580.87	1.58[0]	1.5[0]	1.54[0]	1.0	$7p_{3/2}$	$8d_{3/2}$	1124.88	3.51[−2]	2.5[−2]	1.72[−2]	6.0
$5g_{7/2}$	$7f_{7/2}$	17 639.80	4.53[−2]	4.4[−2]	4.38[−2]	0.5	$7p_{1/2}$	$8s_{1/2}$	1684.00	4.55[−1]	6.9[−1]	4.33[−1]	2.5
$5g_{7/2}$	$7f_{5/2}$	19 362.21	1.14[0]	1.1[0]	1.12[0]	1.2	$8p_{1/2}$	$9s_{1/2}$	3815.10	6.63[−1]	9.5[−1]	6.21[−1]	2.5

TABLE IV. Lifetimes τ in Fr-like Th IV in ns. Uncertainties are given in parentheses. The last column gives relative uncertainties of the final values in %. Experimental energies [12] are given in cm^{-1}. The values of lifetimes evaluated in DF approximation are given to illustrate the correlation contribution. Lifetime of the metastable 7*s* level is given in text.

Level	Energy	$\tau^{(\mathrm{DF})}$	$\tau^{(\mathrm{final})}$	Unc. (%)	Ref. [14]
$6d_{3/2}$	9193.245	431	1086(89)	8.2	1090
$6d_{5/2}$	14 486.34	285	678(46)	6.8	676
$7p_{1/2}$	60 239.1	0.764	1.099(23)	2.1	1.099
$7p_{3/2}$	73 055.9	0.459	0.632(10)	1.6	0.632
$8s_{1/2}$	119 621.6	0.665	0.711(10)	1.4	0.707
$7d_{3/2}$	119 684.6	0.563	0.665(10)	1.5	0.667
$7d_{5/2}$	121 427.1	0.739	0.855(14)	1.6	0.854
$6f_{5/2}$	127 269.2	0.234	0.304(6)	1.8	0.300
$6f_{7/2}$	127 815.3	0.241	0.301(5)	1.6	0.297
$8p_{1/2}$	134 516.5	1.925	3.19(14)	4.5	3.194
$8p_{3/2}$	139 870.9	1.077	1.87(5)	2.6	1.871
$5g_{9/2}$	159 371	0.449	0.79(3)	3.9	0.780
$5g_{7/2}$	159 390	0.455	0.83(3)	3.9	0.815
$9s_{1/2}$	160 728.1	0.960	1.038(13)	1.3	1.031
$8d_{3/2}$	161 954	0.980	1.19(6)	4.9	1.176
$8d_{5/2}$	162 527.8	1.385	1.62(5)	3.1	1.600
$7f_{5/2}$	164 554.7	0.376	0.738(38)	5.2	0.684
$7f_{7/2}$	165 059.0	0.384	0.689(35)	5.1	0.639
$6g_{9/2}$	181 674	0.641	1.85(21)	11	1.567
$6g_{7/2}$	181 675	0.649	2.21(31)	14	1.768

all of the $nd_{3/2}$ terms with $n \leqslant 26$, excluding only the terms that were already listed separately.

We evaluate the contribution from the ionic core α_{core} in the RPA and find $\alpha_{\mathrm{core}} = 7.75(7)$ a.u. We estimate uncertainty in this term to be on the order of 1% based on the comparison of the RPA values for heavier noble gases (Kr and Xe) with experiment and comparison of the ionic core RPA values for heavy ions (such as Ba^{2+}) with coupled-cluster results (see Table 4 of Ref. [26]). Our result is in excellent agreement with the recent RESIS measurement of Th^{4+} polarizability of 7.702(6) a.u. [27]. A counterterm α_{vc} compensating for excitation from the core to the valence shell which violates the Pauli principle is also evaluated in the RPA and is given in rows labeled "vc" in Table V. A difference of the DF and RPA values is taken to be its uncertainty. The core polarizability gives a very large contribution to all scalar polarizabilities, ranging from nearly 100% for the $7p_{1/2}$, where valence terms cancel out each other, to 20% for the 7*s* state. Its contribution to the ground-state $5f_{5/2}$ polarizability is 53%. For comparison, the core polarizability contributes only 6% to the total 7*s* ground-state polarizability of Fr.

The evaluations of the α_0 and α_2 polarizabilities differ only in the angular part. Both scalar and tensor ground-state valence polarizabilities are dominated by a single transition, $5f_{5/2}$–$6d_{3/2}$. It contributes 89% of the scalar valence polarizability. Its contribution (-6.21 a.u.) to the tensor polarizability is larger than the total value, since the $5f_{5/2}$–$6d_{5/2}$ contributes 0.33 a.u. with the opposite sign. The continuous part of spectra is responsible for 1% of α_0 and α_2 for the $5f_{5/2}$ state. We discuss comparison of the ground-state polarizability values with RESIS experiments [11] in Sec. V.

The dominant contribution, 98.5%, to the $\alpha_2(5f_{7/2})$ value comes from the $nd_{5/2}$ states, particularly from the $6d_{5/2}$ state. The contributions to the $\alpha_2(5f_{7/2})$ value from the $ng_{7/2}$ and $ng_{9/2}$ states are 0.2% and 1.7%, respectively, and have a different sign. The dominant contributions to the $\alpha_2(6d_{3/2})$ value are from the $7p_{1/2}$ and $5f_{5/2}$ states and they partly cancel each other. All other states contribute to the $\alpha_2(6d_{3/2})$ value only 15%. The dominant contributions to the $\alpha_2(6d_{5/2})$ value are from the $7p_{3/2}$ and $5f_{7/2}$ states and they nearly exactly cancel each other (to 0.1%). As a result, the $\alpha_2(6d_{5/2})$ polarizability value mainly comes from the $5f_{5/2}$ and $6f_{7/2}$ states. The contributions to the $\alpha_2(7p_{3/2})$ value are distributed between the $7s_{1/2}$, 8*s*, $7d_{3/2}$, $6d_{5/2}$, and $7d_{5/2}$ states. There are very large cancellations among these five terms.

The uncertainties in the values of the 5*f* and 6*d* polarizabilities are overwhelmingly dominated by the uncertainties in the 5*f*–6*d* transitions. Accurate measurement of the 6*d* lifetimes would allow one to significantly reduce all of the uncertainties.

IV. QUADRUPOLE MOMENT

The electric-quadrupole moment $\Theta(\gamma J)$ of an atom in electronic state $|\gamma J\rangle$ is defined as the diagonal matrix element of the $q = 0$ component of the electric-quadrupole operator Q in a spherical basis,

$$\Theta(\gamma J) = \langle \Psi(\gamma J M_J)|Q_0|\Psi(\gamma J M_J)\rangle, \tag{4}$$

with the magnetic quantum number M_J taken to be equal to its maximum value, $M_J = J$ [28]. The quadrupole moment is expressed via the reduced matrix element of the quadrupole operator as

$$\Theta(\gamma J) = \frac{(2J)!}{\sqrt{(2J-2)!(2J+3)!}}\langle \Psi(\gamma J)\|Q\|\Psi(\gamma J)\rangle. \tag{5}$$

Therefore, the calculation of the quadrupole moment of the ground state of Th IV reduces to the calculation of the diagonal matrix element of the electric-quadrupole operator.

The summary of the calculations is given in Table VI, where we list the results of the lowest-order DF, third-order many-body perturbation theory MBPT3, and all-order SD, SDpT, and SD$_{\mathrm{sc}}$ calculations. The MBPT3(6) and MBPT3(10) columns give the third-order values calculated with $l_{\max} = 6$ and $l_{\max} = 10$, respectively. The difference of these values gives an estimate of the higher-partial wave contributions, and is added to the all-order values which were obtained with $l_{\max} = 6$. It contributes 1% lowering the quadrupole moment value. We cannot use the difference of the *ab initio* and scaled all-order results to accurately estimate the uncertainty of the final value, since the correction terms affected by the scaling account for only 1/3 of the total correlation. Instead, we use the calculation of the quadrupole moments in *nd* states of Ca$^+$, Sr$^+$, and Ba$^+$ [29], where the uncertainties could be accurately estimated. The Ca$^+$ theoretical ground-state quadrupole moment [29] is in excellent agreement with a precision experiment [30]. We find that the uncertainties of theoretical values reported in [29] were about 3% of the correlation correction for Ca$^+$ and 2.3% for Ba$^+$, while the correlation corrections contributed 25% and 17% for these ions. In the present Th IV case, the correlation contributed 35% to the ground-state quadrupole moment, so

TABLE V. Contributions to the scalar (α_0) and tensor (α_2) polarizabilities of Th IV ion in a_0^3. Uncertainties are given in parentheses.

$5f_{5/2}$			$5f_{7/2}$			$6d_{3/2}$		
Contr.	α_0	α_2	Contr.	α_0	α_2	Contr.	α_0	α_2
$6d_{3/2}$	6.21(51)	−6.21(51)	$6d_{5/2}$	6.63(50)	−6.63(50)	$7p_{1/2}$	3.23(9)	−3.23(9)
$7d_{3/2}$	0.014(3)	−0.014(3)	$7d_{5/2}$	0.014(3)	−0.014(3)	$8p_{1/2}$	0.011(2)	−0.011(2)
$8d_{3/2}$	0.007(7)	−0.007(7)	$8d_{5/2}$	0.007(7)	−0.007(7)	$9p_{1/2}$	0.000	0.000
$nd_{3/2}$	0.03(3)	−0.03(3)	$nd_{5/2}$	0.03(3)	−0.03(3)	$np_{1/2}$	0.02(2)	−0.02(2)
$6d_{5/2}$	0.29(2)	0.33(3)	$5g_{7/2}$	0.003(1)	0.003(1)	$7p_{3/2}$	0.413(10)	0.330(8)
$7d_{5/2}$	0.001(1)	0.001(1)	$6g_{7/2}$	0.001(1)	0.001(1)	$8p_{3/2}$	0.006(1)	0.005(1)
$nd_{5/2}$	0.002(2)	0.001(1)	$ng_{7/2}$	0.006(6)	0.007(7)	$np_{3/2}$	0.008(8)	0.006(6)
$5g_{7/2}$	0.07(1)	−0.026(2)	$5g_{9/2}$	0.09(1)	−0.043(3)	$5f_{5/2}$	−9.31(76)	1.86(5)
$6g_{7/2}$	0.02(2)	−0.005(1)	$6g_{9/2}$	0.03(1)	−0.012(2)	$6f_{5/2}$	1.70(3)	−0.341(7)
$7g_{7/2}$	0.02(1)	−0.007(1)	$7g_{9/2}$	0.03(1)	−0.012(1)	$7f_{5/2}$	0.156(13)	−0.031(3)
$ng_{7/2}$	0.13(13)	−0.05(5)	$ng_{9/2}$	0.10(10)	−0.05(5)	$nf_{5/2}$	0.22(11)	−0.19(8)
Tail	0.15(20)	−0.05(7)	Tail	0.17(20)	−0.07(9)	Tail	0.02(5)	−0.005(11)
Core	7.75(7)		Core	7.75(7)		Core	7.75(7)	
vc	−0.02(1)		vc	−0.02(1)		vc	−0.43(7)	
Total	14.67(60)	−6.07(53)	Total	14.84(59)	−6.95(52)	Total	4.53(81)	−1.62(21)

$6d_{5/2}$			$7p_{3/2}$			$7p_{1/2}$		$7s_{1/2}$	
Contr.	α_0	α_2	Contr.	α_0	α_2	Contr.	α_0	Contr.	α_0
$7p_{3/2}$	3.16(7)	−3.16(7)	$7s_{1/2}$	−8.31(22)	8.31(22)	$7s_{1/2}$	−11.54(32)	$7p_{1/2}$	11.54(32)
$8p_{3/2}$	0.036(3)	−0.036(3)	$8s_{1/2}$	6.90(11)	−6.90(11)	$8s_{1/2}$	2.96(7)	$8p_{1/2}$	0.036(4)
$9p_{3/2}$	0.003	−0.003	$9s_{1/2}$	0.212(6)	−0.212(6)	$9s_{1/2}$	0.16	$np_{1/2}$	0.08(8)
$np_{3/2}$	0.05(5)	−0.05(5)	$ns_{1/2}$	0.10(1)	−0.10(1)	$ns_{1/2}$	0.09(2)	$7p_{3/2}$	16.62(43)
$5f_{5/2}$	−0.29(2)	−0.33(3)	$6d_{3/2}$	−0.413(10)	−0.330(8)	$6d_{3/2}$	−6.45(18)	$8p_{3/2}$	0.003(2)
$6f_{5/2}$	0.100(2)	0.114(2)	$7d_{3/2}$	2.67(4)	2.14(3)	$7d_{3/2}$	14.64(22)	$np_{3/2}$	0.05(5)
$nf_{5/2}$	0.05(3)	−0.06(3)	$nd_{3/2}$	0.03(2)	0.03(2)	$8d_{3/2}$	0.40(7)		
$5f_{7/2}$	−8.84(67)	3.16(24)	$6d_{5/2}$	−4.74(11)	0.95(2)	$nd_{3/2}$	0.12(6)		
$6f_{7/2}$	1.94(3)	−0.69(1)	$7d_{5/2}$	22.21(30)	−4.44(6)				
$7f_{7/2}$	0.19(1)	−0.067(4)	$8d_{5/2}$	0.31(2)	−0.06(1)				
$nf_{7/2}$	0.21(11)	−0.08(4)	$nd_{5/2}$	0.07(7)	−0.01(1)				
Tail	0.02(4)	−0.01(1)	Tail	0.01(2)	−0.008(6)	Tail	0.01(3)	Tail	0.003(1)
Core	7.75(7)		Core	7.75(7)		Core	7.75(7)	Core	7.75(7)
vc	−0.72(4)		vc	0.001		vc	−0.004(1)	vc	−0.50(10)
Total	3.67(70)	−1.09(27)	Total	26.88(42)	−0.64(26)	Total	8.13(41)	Total	35.58(55)

we estimate that it is accurate to about 4%, yielding the final value of 0.624(14) a.u.

V. COMPARISON WITH RESIS VALUES

Binding energies of high-L Rydberg states ($L \geqslant 7$) of Th^{2+} with $n = 27$–29 were studied using the resonant excitation Stark ionization spectroscopy (RESIS) method in [11]. Analysis of the observed RESIS spectra led to determination of five properties of the Th^{3+} ion: its electric-quadrupole moment Θ in the ground state, adiabatic scalar and tensor ground-state dipole polarizabilities, and the dipole matrix elements connecting the ground $5f_{5/2}$ level to the low-lying $6d_{3/2}$ and $6d_{5/2}$ levels. The frequencies of the 14 well-resolved single lines were fit to determine the best values of the following parameters [11]: $\langle 5f_{5/2}||D||6d_j\rangle$ matrix elements, Θ, and scalar and tensor polarizabilities $\alpha_{0,2}^{\mathrm{mod}}$ with the contribution of the $5f_{5/2}$–$6d$ terms subtracted out. The results of the fit were used to determine full adiabatic polarizabilities α_0 and α_2. The core polarizability α_{core}, i.e., the polarizability of Th^{4+}, was determined in Ref. [27]. We list comparison of our results with RESIS data for all of these quantities in Table VII. We have already discussed the calculations of these properties and their uncertainties in the previous section, so we discuss only comparison of the results here. The present values and the RESIS fit results for the quadrupole moment Θ and $\langle 5f_{5/2}||D||6d_{3/2}\rangle$ agree to 2σ and 1.5σ, respectively. The central values for the $\langle 5f_{5/2}||D||6d_{5/2}\rangle$ are nearly identical. This leads to a difference in ratio R of the $5f_{5/2}$–$6d_{3/2}$ and $5f_{5/2}$–$6d_{5/2}$ matrix elements. The theoretical prediction for this ratio is by far more accurate (0.6%) than the theory values of the matrix elements (4%), since the correlation corrections are very similar for the transitions involving states of the same fine-structure multiplet. We took the difference of the ratios

TABLE VI. Quadrupole moment Θ of Th IV in the ground $5f_{5/2}$ state in a.u. See text for designations.

DF	MBPT3(6)	MBPT3(10)	SD	SDpT	SD$_{\mathrm{sc}}$
0.916	0.555	0.550	0.620	0.628	0.624(14)

TABLE VII. Comparison of the electric-dipole $5f_{5/2}$–$6d_j$ matrix elements D, ground-state quadrupole moment Θ, scalar α_0 and tensor α_2 ground-state Th^{3+} polarizabilities, and Th^{4+} ground-state polarizability with the RESIS experimental results [11,27]. R is the ratio of the $5f_{5/2}$–$6d_{3/2}$ and $5f_{5/2}$–$6d_{5/2}$ matrix elements. α^{mod} is the polarizability with the contribution of the $5f_{5/2}$–$6d$ terms subtracted out. α_0^{mod1} has core polarizability α_{core} subtracted out as well. All values are in atomic units.

Property	Present	Refs. [11,27]
Θ	0.624(14)	0.54(4)
$\lvert\langle 5f_{5/2}\vert\vert D\vert\vert 6d_{3/2}\rangle\rvert$	1.530(63)	1.435(10)
$\lvert\langle 5f_{5/2}\vert\vert D\vert\vert 6d_{5/2}\rangle\rvert$	0.412(16)	0.414(24)
R	3.716(23)	3.47(20)
α_0	14.67(60)	15.42(17)
α_0^{mod}	8.18(34)	9.67(15)
α_{core} [Th^{4+}]	7.75(7)	7.702(6)
α_0^{mod1}	0.43(33)	1.97(15)
α_2	−6.07(53)	−3.6(1.3)
α_2^{mod}	−0.19(13)	1.5(1.3)

calculated using third-order MBPT and all-order methods as the uncertainty, which is rather conservative. Using our value of the ratio and RESIS $5f_{5/2}$–$6d_{3/2}$ matrix element yields 0.386(4) a.u. for the $5f_{5/2}$–$6d_{5/2}$ matrix element, which is shifted by 1σ from the RESIS fit value of 0.414(24) a.u.

The α_0^{mod} value is dominated by the core polarizability; therefore, we separated it out for comparison purposes:

$$\alpha_0^{\text{mod}} = \alpha_{\text{core}} + \alpha_0^{\text{mod1}}.$$

The theoretical value for the core polarizability is in excellent agreement with the experimental value [27]. The remainder α_0^{mod1} disagrees significantly with the RESIS fit. A large fraction (65%) of this remainder contribution and essentially all of its uncertainty comes from the $ng_{7/2}$ terms with $n > 7$. Even if we add DF value for $(n > 7)g_{7/2}$ terms, 0.70 a.u. (which is an upper bound for this property, since DF systematically and significantly overestimates the polarizability contributions), to the remaining contributions, we get $\alpha_0^{\text{mod1}} = 0.86$ a.u. Therefore, it is difficult to come up with a scenario in which α_0^{mod1} is as high as 1.97 a.u. The total theoretical α_0 and α_2 values are in agreement with RESIS values to about 2σ. It would be very interesting to see if RESIS line data can be reproduced by using only one free parameter, $\langle 5f_{5/2}\vert\vert D\vert\vert 6d_{3/2}\rangle$, and allowing $\Theta = 0.624(14)$ a.u., $R = 3.716(23)$ a.u., $\alpha_{\text{core}} = 7.702(6)$ a.u., $\alpha_0^{\text{mod1}} = 0.43(33)$ a.u., and $\alpha_2^{\text{mod}} = -0.19(12)$ a.u. to vary within the 1–2σ uncertainties.

VI. CONCLUSION

In summary, we carried out a systematic study of Fr-like Th IV atomic properties for the $7s$, $8s$, $9s$, $10s$, $7p$, $8p$, $6d$, $7d$, $5f$, $6f$, $7f$, $5g$, and $6g$ states using a high-precision relativistic all-order approach. Reduced matrix elements, oscillator strengths, transition rates, and lifetimes for the 24 first low-lying levels, ground-state quadrupole moment, scalar polarizabilities of the seven first states, and tensor polarizabilities of the $5f$, $6d$, and $7p_{3/2}$ states are calculated. We evaluate the uncertainties of our calculations for all of the values listed in this work. Detailed comparison of the present values with RESIS experimental results [11,27] is carried out. These calculations provide recommended values critically evaluated for their accuracy for the development of ultraprecise nuclear clock, RESIS experiments with actinide ions, and other studies.

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