

**Relativistic all-order calculations of Th, Th<sup>+</sup>, and Th<sup>2+</sup> atomic properties**M. S. Safronova,<sup>1,2</sup> U. I. Safronova,<sup>3</sup> and Charles W. Clark<sup>2</sup><sup>1</sup>*Department of Physics and Astronomy, University of Delaware, Newark, Delaware 19716, USA*<sup>2</sup>*Joint Quantum Institute, National Institute of Standards and Technology and the University of Maryland, Gaithersburg, Maryland 20899, USA*<sup>3</sup>*Department of Physics, University of Nevada, Reno, Nevada 89557, USA*

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Excitation energies, term designations, and  $g$  factors of Th, Th<sup>+</sup>, and Th<sup>2+</sup> are determined using a relativistic hybrid configuration-interaction (CI) plus all-order approach that combines configuration-interaction and linearized coupled-cluster methods. The results are compared with other theoretical and experimental values where available. We find some vanishing  $g$  factors, similar to those known in lanthanide spectra. Reduced matrix elements, oscillator strengths, transition rates, and lifetimes are determined for Th<sup>2+</sup>. To estimate the uncertainties of our results, we compare our values with the available experimental lifetimes for higher  $5f7p\ ^3G_4$ ,  $7s7p\ ^3P_0$ ,  $7s7p\ ^3P_1$ , and  $6d7p\ ^3F_4$  levels of Th<sup>2+</sup>. These calculations provide a benchmark test of the CI plus all-order method for heavy systems with several valence electrons and yield recommended values for transition rates and lifetimes of Th<sup>2+</sup>.

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**I. INTRODUCTION**

The <sup>229</sup>Th nucleus provides a unique opportunity for the development of a nuclear clock [1] due to an unusually low first excitation energy of only several eV [2,3], making the corresponding nuclear transition accessible with laser excitation [4]. This very narrow,  $6h$  lifetime [4], nuclear transition is expected to be well isolated from effects of external fields leading to potentially very small ultimate uncertainty in the corresponding frequency standard.

The transition frequency is expected to be very sensitive to temporal variation of the fine-structure constant and the dimensionless strong interaction parameter  $m_q/\Lambda_{\text{QCD}}$  as compared to atomic transitions, making <sup>229</sup>Th one of the most attractive candidates for such studies [5]. The physical implementation of the frequency standard may employ, for example, the closed electronic shell of Th<sup>4+</sup> in a UV-transparent crystal doped with a macroscopic number of <sup>229</sup>Th nuclei [6] or the stretched states within the  $5f_{5/2}$  electronic ground level of both nuclear ground and isomer manifolds of a single trapped ion [7]. Laser-cooled Wigner crystals of <sup>229</sup>Th<sup>3+</sup> allow for high-precision spectroscopy [8,9]. Singly and doubly charged <sup>232</sup>Th and <sup>229</sup>Th ions have been produced by laser ablation of solid-state thorium compounds and by inductively coupled plasma techniques with mass-spectrometry analysis from liquid solutions of thorium [10]. The latter method was found to be more applicable for producing ions of radioactive <sup>229</sup>Th for laser experiments when searching for the energy value of the isomeric nuclear transition [10].

We note that a frequency standard based on the nuclear transition in <sup>229</sup>Th can be implemented with either neutral or ionized Th. The nuclear transition might be accessed using an electronic bridge process [11], which involves matching combined electronic and nuclear energy levels to drive the nuclear transition [11]. Implementing the electronic bridge will require knowledge of the complex electronic configurations of neutral or ionized Th.

Much of the recent work on the spectrum of thorium is motivated by its use as source of wavelength standards for high-resolution spectrographs [12]. The high density of Th I spectral lines—approximately 20 000 in the wavelength interval between 250 and 5500 nm—has made the thorium-argon hollow-cathode lamp a convenient tool for accurate wavelength calibration. In particular, such lamps are installed on many high-precision astronomical spectrographs. These include the High Accuracy Radial velocity Planet Searcher instrument, now the world's most precise astronomical spectrograph, which has a relative precision of 3 parts in 10<sup>9</sup> and can measure stellar velocities to within  $\sim 1$  m/s [13].

The best characterized Th ion today is Th<sup>3+</sup>, which has a monovalent Fr-like electronic structure and has been studied with the high-precision all-order method [14]. Recommended values for electric dipole matrix elements, oscillator strengths, transition rates, lifetimes, scalar and tensor polarizabilities, and hyperfine constants critically evaluated for their accuracy have been published for a large number states in Th<sup>3+</sup> [14]. The combination of the experimental measurements of hyperfine constants of Th<sup>3+</sup> with theoretical calculations has enabled accurate determination of magnetic dipole and electric quadrupole moments [15]. Analysis of resonant excitation Stark ionization spectroscopy spectra has led to the determination of the Th<sup>3+</sup> ground-state electric quadrupole moment, adiabatic scalar and tensor dipole polarizabilities, and the dipole matrix elements connecting the ground level to low-lying excited levels of Th<sup>3+</sup> [16].

Due to the more complicated atomic structure of Th<sup>2+</sup>, Th<sup>+</sup>, and Th, which have, respectively, two, three, and four valence electrons, their atomic properties are less precisely known than that of Th<sup>3+</sup>. The work presented in this paper demonstrates accurate calculation of the energies of these systems and provides transition matrix elements, oscillator strengths, and lifetimes for a large number of Th<sup>2+</sup> states. This work also serves as a benchmark test of the accuracy of the CI plus all-order method for systems with multiple valence electrons, including tetravalent neutral Th.

## II. REVIEW OF CURRENT KNOWLEDGE OF THE STRUCTURE OF THORIUM AND ITS IONS

Properties of thorium and its ions have received more extensive experimental investigation than theoretical, due to significant difficulties in accurate first-principles calculations of such heavy many-electron systems. We begin with a summary of the experimental work and conclude with a survey of theoretical approaches.

A list of about 9500 spectral lines of Th in the range 234.5–2966.2 nm was obtained and characterized by Zalubas [17]. This resulted in determination of 254 even-parity and 322 odd-parity levels. Their  $g$  factors were obtained by Zeeman spectroscopy and used as an aid in spectral classification, i.e., the assignment of angular momenta, parity, and electronic configurations.

Energy levels and classified lines in the second spectrum of thorium (Th II) were described by Zalubas and Corliss [18]. About 6500 lines were classified as transitions between 199 odd levels and 271 even levels; 188 levels result from the odd  $5f6d^2 + 5f7s^2$  and  $6d^27p + 7s^27p$  configurations [18]. The 235 levels of the even  $5f^27s + 5f7s7p + 5f6d7p + 5f^26d$  configurations were also determined [18]. Resonantly enhanced three-photon ionization of  $\text{Th}^+$  was used to determine that its ionization potential is between 11.9 and 12.3 eV [19].

The Th III spectrum was observed in the region 1000–3000 nm and ten lines were identified [20]. The ground level of  $\text{Th}^{2+}$  was determined to be  $5f6d^3H_4$  [20]. The first excited level  $6d^2^3F_2$  was determined to be only  $63.2\text{ cm}^{-1}$  above the ground level. Wyart and Kaufman [21] extended the analysis of  $\text{Th}^{2+}$ . They classified the 92 lines above 194 nm that were previously observed [21]. The  $5f6d$ ,  $5f^2$ ,  $5f7d$ , and  $5f8s$  configurations were completely identified [21].

Using time-resolved laser-induced fluorescence method, Biémont *et al.* measured the lifetimes of six levels belonging to the  $5f^2$ ,  $5f7p$ ,  $7s7p$ , and  $6d7p$  configurations of  $\text{Th}^{2+}$  [22]. These transitions provide the mechanism of a cosmochronometer for estimating the age of the galaxy.

An online database of published and unpublished actinide energy levels [23] lists 693 levels of Th and 507 of  $\text{Th}^+$  with uncertainties of  $0.001\text{ cm}^{-1}$ . Recently, Redman *et al.* [12] presented results of precise observations of a thorium—argon hollow-cathode lamp emission spectrum in the region between 3500 and 1175 nm using a high-resolution Fourier transform spectrometer. Their measurements were combined with results from previously published thorium line lists [24–29] to reoptimize the energy levels of neutral, singly ionized, and doubly ionized thorium (Th,  $\text{Th}^+$ , and  $\text{Th}^{2+}$ ). A systematic analysis of previous measurements in light of these results enabled Redman *et al.* [12] to identify and propose corrections for systematic errors and typographical errors and incorrect classifications in previous identifications. Redman *et al.* [12] present 787 levels of Th I and 516 of Th II in their tables.

Dzuba and Flambaum [30] presented analytical estimates and numerical calculations showing that the energy-level density in open-shell atoms increases exponentially with excitation energy. They used the relativistic Hartree-Fock and configuration-interaction methods to calculate the densities of states of Th and  $\text{Th}^+$ . Their results were used to estimate the

effect of electrons on the nuclear clock transition discussed in the previous section [30].

Porsev and Flambaum [31] used the CI plus many-body perturbation theory (MBPT) method to study the effect of atomic electrons on the nuclear clock transition due to the electronic bridge process. They calculated energies of several high-lying even-parity states that have yet to be seen by experiments. Roy *et al.* have performed relativistic two-component *ab initio* calculations for  $\text{Th}^+$  and  $\text{Th}^{2+}$  ions [32].

In our present work we evaluate atomic properties of Th,  $\text{Th}^+$ , and  $\text{Th}^{2+}$  using the CI plus all-order approach. Excitation energies and  $g$  factors are compared with experimental [23] and other theoretical results [33]. We also evaluate multipole transition rates and lifetimes of low-lying levels for  $\text{Th}^{2+}$ .

## III. COMPUTATIONAL METHOD

Calculation of the properties of thorium and its first few ions requires an accurate all-order treatment of electron correlations. Low-order perturbation methods are ineffective in heavy systems with more than one valence electron, due to large effects of valence-valence electronic correlations. Moreover, the radonlike core of the thorium atom is sufficiently large that core-core and core-valence correlations have to be treated accurately as well. This can be accomplished within the framework of the CI plus all-order method that combines configuration-interaction and coupled-cluster approaches [34–38]. The CI plus all-order method was used to evaluate properties of systems with three valence electrons in Refs. [39–42]. Properties of systems with four valence electrons were calculated with the CI plus all-order method in Ref. [42] for Sn-like ions. The spectra of the superheavy elements No, Lr, and Rf with two, three, and four valence electrons was recently presented by Dzuba *et al.* [43].

In the CI plus all-order method, we start with a solution of the Dirac-Fock (DF) equations

$$H_0\psi_c = \varepsilon_c\psi_c, \quad (1)$$

where  $H_0$  is the relativistic DF Hamiltonian [34,44] and  $\psi_c$  and  $\varepsilon_c$  are single-electron wave functions and energies. The calculations are carried out in the  $V^{N-4}$ ,  $V^{N-3}$ , and  $V^{N-2}$  potentials for Th,  $\text{Th}^+$ , and  $\text{Th}^{2+}$ , respectively, where  $N$  is the total number of the electrons. Therefore, the calculations are carried out with the same radonlike  $\text{Th}^{4+}$  frozen-core Dirac-Fock potential of all three systems considered in this work.

The wave functions and the corresponding low-lying energy levels are determined by solving the many-electron relativistic equation for two, three, or four valence electrons [45]

$$H_{\text{eff}}(E_n)\Phi_n = E_n\Phi_n.$$

The effective Hamiltonian is defined as

$$H_{\text{eff}}(E) = H_{\text{FC}} + \Sigma(E), \quad (2)$$

where  $H_{\text{FC}}$  is the Hamiltonian in the frozen-core approximation. The energy-dependent effective Hamiltonian term  $\Sigma(E) = \Sigma_1 + \Sigma_2$  is calculated using a modified version of the all-order linearized coupled-cluster method with single and double excitations described in [46,47]. Therefore, the effective Hamiltonian contains dominant core and core-valence correlation corrections to all orders. The valence correlations

are treated by the CI method [45]. We refer the reader to Ref. [34] for the formulas and detailed description of the CI plus all-order method.

The CI plus all-order approach is based on the Brillouin-Wigner variant of the many-body perturbation theory, rather than the Rayleigh-Schrödinger variant, leading to dependence of  $\Sigma$  upon energy. This introduces some subtleties associated with appropriate treatment of energy denominators. This issue has been discussed in Ref. [34] and we adopt the technical procedure that was recommended there.

The configurations are strongly mixed in all three systems considered here. We present the results for the following configurations: (i) Th:  $6d^27s^2, 6d^37s, 6d^4, 6d^27s7p, 5f6d^27s, 5f6d7s^2, 6d7s^27p$ ; (ii) Th<sup>+</sup>:  $6d7s^2, 6d^27s, 6d^3, 5f^27s, 5f^26d, 5f6d7s, 5f7s^2, 5f6d^2$ ; and (iii) Th<sup>2+</sup>:  $5f6d, 5f7d, 5f7s, 5f8s, 6d6f, 6d7p, 6d^2, 5f^2, 7s^2, 5f7p, 5f6f, 6d7s$ . We also calculate  $g$  factors and compare them with experimental values given in Ref. [23]. For a single configuration that is described by pure  $LS$  coupling, the nonrelativistic  $g$  factor of the many-electron state is given by the Landè formula

$$g_{\text{nr}} = 1 + \frac{J(J+1) - L(L+1) + S(S+1)}{2J(J+1)}, \quad (3)$$

where  $J$  is the total angular momentum,  $L$  is the total orbital angular momentum, and  $S$  is the total spin angular momentum. As will be discussed in the next section, we find that the nonrelativistic  $g$  factors are often useful for spectral term classification.

#### IV. EXCITATION ENERGIES OF Th, Th<sup>+</sup>, and Th<sup>2+</sup>

Excitation energies of the 73 lowest states of neutral Th are listed in Table I. All energies are given relative to the  $6d^27s^2\ ^3F_2$  ground state. Theoretical results calculated with the CI plus all-order method are listed in columns labeled “Present.” The results are compared with experimental energies and  $g$  factors given in Ref. [23]. Some of the energy levels listed in [23] are only identified by the total angular momentum  $J$  and not by a complete  $LSJ$  term designation. Such designations are always approximate and sometimes ambiguous, as in cases of strong configuration mixing.

##### A. Term identification using $g$ factors

All of the states reported in Table I have some admixture of configurations, leading to ambiguities in term identification. Thus, we list two sets of term designations in Table I: the term listed in [23] and our term identification, which is based on comparing the experimental  $g$  factor with the Landè formula value given by Eq. (3) as described below. The corresponding columns are labeled “[23]” and “Present.” First, we group all levels by  $J$  and by parity. This leads to a relatively small number of possible terms for each level since there are only three configuration present for even levels and four configurations for odd levels for the energy range in Table I. We then identify the term appropriate for each level using the agreement of the experimental  $g$  factor with the Landè formula. When several configurations have the same  $J$  and parity, we also verify which configuration has the largest mixing coefficient. When no entry appears in a term column that

indicates that no term has been proposed. The  $g_{\text{nr}}$  calculated using Eq. (3) are given in columns “ $g_{\text{nr}}$ .” The “Present” column reports the actual  $g$  factor that we calculate for the state as a whole. In general, the calculated  $g$  factors are in good agreement with the Landè formula, but there are exceptions in cases of strong configuration mixing.

Two interesting entries in Table I are the levels  $6d^37s\ ^5F_1$  and  $6d^27s7p\ ^5F_1$ , for which the column “ $g_{\text{nr}}$ ” reports a value of zero for the theoretical  $g$  factor. That factor is the Landè  $g$  factor given by Eq. (3), which (but for a small correction due to the anomalous magnetic moment of the electron) describes the gyromagnetic ratio of a single-configuration quantum state with well-defined quantum numbers  $L$ ,  $S$ , and  $J$  (and orbital, spin, and total electronic angular momentum, respectively, in units of the reduced Planck constant  $\hbar$ ). A vanishing (or very small)  $g$  factor for a level implies that its energy is relatively insensitive to the presence of a magnetic field. Such an attribute is of interest in applications to atomic frequency standards and precision measurement, so we comment briefly on the use of Eq. (3) as a screen in the search for small  $g$  factors.

From Eq. (3) we find that  $g = 0$  when

$$3J(J+1) - L(L+1) + S(S+1) = 0, \quad (4)$$

where  $L \geq 0$  is an integer,  $J$  and  $S$  are both non-negative integers or half-integers, and  $|L - S| \leq J \leq L + S$ , the triangularity condition, is satisfied. Solution of the constrained equation (4) is an exercise in integer programming, for which even linear examples fall in the NP-hard class of computational complexity [48]. However, since only relatively small values of  $J$ ,  $L$ , and  $S$  are relevant to the Periodic Table, the practical solutions of Eq. (4) can be found by searching tables of computed values of its left-hand side.

Most of these solutions are of the type valid for any  $J$ :

$$L = 2J + 1, \quad S = J + 1, \quad (5)$$

i.e., those with term designations  $^{2J+3}(2J+1)_J$  for  $J = 0, \frac{1}{2}, 1, \dots$ . Some of these terms are listed in Tables 3 and 4 of Ref. [49].

A number of such levels are identified in the NIST Atomic Spectra Database [50], where they are usually found to have  $g$  factors of 0.01 or less. Examples include Nd I,  $4f^46s^2\ ^5F_1$ ; Pm II,  $4f^56s\ ^7H_2$ ; Pm II,  $4f^56s\ ^5F_2$ ; Tb I,  $4f^85d6s^2\ ^6G_{3/2}$ ; Dy II,  $4f^{10}(^5I)5d\ ^6G_{3/2}$ ; Sm I,  $4f^65d(^8H)6s\ ^7H_2$ ; and Sm I,  $4f^65d(^6H)6s\ ^7H_2$ .

There are other solutions of Eq. (4) for values of  $J$ ,  $L$ , and  $S$  that do not satisfy Eq. (5), such as  $J = 3/2$ ,  $L = 11$ , and  $S = 21/2$ . These all appear to be associated with highly excited states involving multiple open shells.

##### B. Hyperfine Landè $g$ factors

A similar method can be used to search for vanishing Landè  $g$  factors  $g_F$  for hyperfine levels. Ignoring contributions from the nuclear magneton and the electron anomaly, we have (see Ref. [51])

$$g_F = g_J \frac{F(F+1) + J(J+1) - I(I+1)}{2F(F+1)}, \quad (6)$$

where  $g_J$  is the  $g$  factor associated with the electrons,  $J$  the net electronic angular momentum,  $F$  the total angular

TABLE I. Levels (in  $\text{cm}^{-1}$ ) and  $g$  factors of the lowest states of thorium. Nonrelativistic values of  $g$  factors  $g_{\text{nr}}$  are given by Eq. (3). Calculations are compared with experiment [23]. Configuration and term labels are determined as described in Sec. IV A.

Even-parity states ( $6d^27s^2 + 6d^37s$ )									Odd-parity states ( $6d^27s7p + 5f6d^27s + 5f6d7s^2 + 6d7s^27p$ )								
Conf.	Term		Energy		g factor			Conf.	Term		Energy		g factor				
	[23]	Present	$J$	Present	Expt.	Expt.	Present		$g_{\text{nr}}$	[23]	Present	$J$	Present	Expt.	Expt.	Present	$g_{\text{nr}}$
$6d^27s^2$	$^3F$	$^3F$	2	0	0	0.735	0.718	0.667	$5f6d7s^2$	$^3P$		0	13954	14247	0.0	0.000	0.0
$6d^27s^2$		$^3P$	2	3790	3688	1.255	1.287	1.500	$6d^27s7p$	$^5D$		0	18127	18382	0.0	0.000	0.0
$6d^37s$	$^5F$	$^5F$	2	6677	6362	1.010	1.001	1.000	$6d7s^27p$	$^3P$		0	20790	20543	0.0	0.000	0.0
$6d^27s^2$		$^3D$	2	7445	7280	1.185	1.152	1.167									
$6d^37s$	$^5P$	$^5P$	2	12449	11802	1.780	1.759	1.833	$6d7s^27p$	$^3D$	$^3D$	1	11455	11878	0.725	0.709	0.500
									$5f6d7s^2$	$^3P$	$^3P$	1	13989	14244	1.205	1.218	1.500
$6d^27s^2$	$^3P$		0	2708	2558	0.0	0.000	0.0	$6d^27s7p$	$^5F$	$^5F$	1	15805	15737	0.385	0.297	0.000
$6d^37s$			0	15243	14227	0.0	0.000	0.0	$5f6d7s^2$	$^3D$	$^1P$	1	17388	17357	0.505	1.024	1.000
$6d^27s^2$			0	17327	16351	0.0	0.000	0.0									
$6d^27s^2$	$^3P$	$^3P$	1	3948	3865	1.480	1.481	1.500	$5f6d7s^2$	$^3F$	$^3F$	2	7493	8244	0.775	0.796	0.667
$6d^37s$	$^5F$	$^5F$	1	5887	5563	0.065	0.048	0.000	$6d7s^27p$	$^3F$	$^3F$	2	10481	10783	0.725	0.730	0.667
$6d^37s$	$^5P$	$^5P$	1	12179	11601	2.400	2.430	2.500	$5f6d7s^2$	$^1D$	$^1D$	2	11223	12114	0.975	0.921	1.000
$6d^37s$		$^1P$	1	14754	13963	0.760	0.714	1.000	$6d7s^27p$	$^3D$	$^3D$	2	13641	14032	1.125	1.239	1.167
									$6d^27s7p$	$^5G$	$^3F$	2	14253	14465	0.810	0.759	0.667
									$6d7s^27p$	$^1D$	$^1D$	2	15879	16217	1.070	1.085	1.000
$6d^27s^2$	$^3F$	$^3F$	3	2815	2869	1.085	1.078	1.083	$6d^27s7p$	$^5F$	$^5F$	2	17187	17224	1.045	1.029	1.000
$6d^37s$	$^5F$	$^5F$	3	7818	7502	1.250	1.241	1.250	$5f6d7s^2$	$^3P$	$^3D$	2	17781	17847	1.165	1.122	1.167
$6d^37s$	$^5P$	$^5P$	3	13413	12848	1.390	1.629	1.667									
$6d^37s$	$^3G$	$^3G$	3	13613	13089	1.050	0.800	0.750	$5f6d7s^2$	$^3G$	$^5G$	3	10194	10527	0.870	0.940	0.917
$6d^37s$		$^3F$	3	16685	15970	1.205	1.166	1.083	$5f6d7s^2$	$^3F$	$^1F$	3	10762	11242	1.010	0.914	1.000
$6d^37s$		$^5F$	3	18138	17398	1.195	1.213	1.250	$6d7s^27p$	$^3F$	$^3F$	3	13664	13945	1.110	1.129	1.083
$6d^37s$	$^3F$	$^3F$	3	20527	19713	1.110	1.094	1.083	$6d^27s7p$	$^5G$	$^3G$	3	15054	15167	1.065	0.591	0.750
$6d^37s$	$^1F$	$^5F$	3	22222	21595	1.040	1.211	1.250									
									$5f6d7s^2$	$^3H$	$^3H$	4	7296	7795	0.865	0.872	0.800
$6d^27s^2$	$^3F$	$^3F$	4	4953	4962	1.210	1.219	1.250	$5f6d7s^2$	$^1G$	$^1G$	4	9858	10414	0.985	0.962	1.000
$6d^27s^2$	$^1G$	$^5G$	4	8156	8111	1.065	1.213	1.150	$5f6d7s^2$	$^3G$	$^3G$	4	12827	13175	1.095	1.111	1.050
$6d^37s$	$^5F$	$^5G$	4	9113	8800	1.310	1.138	1.150	$5f6d7s^2$	$^3F$	$^3F$	4	13683	14207	1.170	1.135	1.250
$6d^37s$	$^3G$	$^1G$	4	13782	13297	1.000	0.990	1.000	$5f6d^27s$	$^5H$	$^5H$	4	16092	16347	0.880	0.939	0.900
$6d^37s$	$^3H$	$^3H$	4	15939	15493	0.905	0.887	0.800	$5f6d^27s$	$^5I$	$^5I$	4	16840	16784	0.695	0.639	0.600
$6d^37s$	$^3F$	$^3F$	4	18692	17960	1.175	1.209	1.250	$6d7s^27p$	$^3F$	$^3F$	4	17883	18054	1.185	1.165	1.250
$6d^37s$	$^3F$	$^3F$	4	20347	19532	1.204	1.196	1.250	$6d^27s7p$	$^5G$	$^5G$	4	18503	18810	1.150	1.129	1.150
$6d^37s$		$^3G$	4	21725	21646	1.090	1.056	1.050									
									$5f6d7s^2$	$^3H$	$^3H$	5	10884	11197	1.040	1.028	1.033
$6d^37s$	$^5F$	$^5F$	5	10198	9805	1.365	1.360	1.400	$5f6d7s^2$	$^3G$	$^3G$	5	15255	15490	1.190	1.188	1.200
$6d^37s$		$^3G$	5	14723	14204	1.150	1.134	1.200	$5f6d^27s$	$^5I$	$^5H$	5	17311	17501	1.015	1.099	1.100
$6d^37s$	$^3H$	$^3H$	5	17614	17166	1.115	1.111	1.033	$5f6d^27s$	$^5H$	$^1H$	5	17974	18011	1.025	0.942	1.000
$6d^37s$	$^1H$	$^1H$	5	21775	21143	1.030	1.005	1.000	$6d^27s7p$	$^5G$	$^5H$	5	19320	19588	1.150	1.151	1.100
$5f6d7s7p$	$^5I$	$^1H$	5	22845	23277	1.010	1.007	1.000	$5f6d^27s$	$^1H$	$^3H$	5	20586	20322	1.060	1.040	1.033
$5f6d7s7p$	$^5H$	$^3H$	5	26060	26381	1.025	1.031	1.033	$6d^27s7p$	$^5F$	$^5G$	5	20932	21077	1.250	1.246	1.267
									$5f6d^27s$	$^3I$	$^5I$	5	22537	22399	0.930	0.893	0.900
$6d^37s$	$^3H$	$^3H$	6	17063	16554	1.165	1.161	1.167									
$5f6d7s7p$	$^5I$	$^5I$	6	26681	26997	1.110	1.109	1.071	$5f6d7s^2$	$^3H$	$^3H$	6	14188	14482	1.170	1.160	1.167
$6d^4$	$^3H$	$^3H$	6	28926	27972	1.125	1.164	1.167	$5f6d^27s$	$^5I$	$^5I$	6	19332	19227	1.085	1.069	1.071
$5f6d7s7p$	$^5H$	$^3H$	6	29335	29553	1.185	1.139	1.167	$5f6d^27s$	$^5H$	$^5H$	6	19940	19986	1.195	1.192	1.214
									$5f6d^27s$	$^3I$	$^3I$	6	23282	23307		1.029	1.024
									$6d^27s7p$	$^5G$	$^5G$	6	24038	24085	1.220	1.260	1.333
									$5f6d7s^2$	$^3I$	$^3H$	6	24861	24850		1.179	1.167

momentum, and  $I$  the angular momentum of the nucleus. Thus  $g_F = 0$  when

$$F(F+1) = I(I+1) - J(J+1) \quad (7)$$

subject to the triangularity condition between  $I$ ,  $J$ , and  $F$ .

The simple expression of Eq. (7) belies the diversity of its solutions, as suggested in Table II. A number of the values of  $I$  displayed there are associated with stable or long-lived nuclei.

### C. Energies of neutral Th

The calculated energy levels of thorium are in excellent agreement with experiment for such a heavy tetravalent neutral

TABLE II. Simplest nontrivial solutions of Eq. (7) for  $F \leq 10$ . The only solutions for  $F < 3/2$  are trivial. Multiple solutions are given for  $F = 4, 5$ , and  $8$  because they lie close together.

$F$	$J$	$I$	$F$	$J$	$I$
3/2	3	7/2	6	5	8
2	2	3	13/2	8	21/2
5/2	8	17/2	7	9/2	17/2
3	5	6	15/2	11	27/2
7/2	4	11/2	8	5/2	17/2
4	7/2	11/2	8	13/2	21/2
4	9	10	8	10	13
9/2	7	17/2	17/2	80	171/2
5	3	6	9	6	11
5	14	15	19/2	12	31/2
11/2	35	71/2	10	8	13

atom. We find 0.2%–2% differences between the theoretical and experimental energies for 35 out of 78 levels listed in Table I. Only 9 energies differ with experiment for more than 5%. We find that inclusion of a sufficient number of configurations is particularly important for achieving accurate results. To ensure that all dominant configurations are included we started with the preliminary calculation that included a few thousand configuration state functions (CSFs). The initial configuration space was constructed by allowing two excitations into the valence orbitals from the  $6d^27s^2$ ,  $6d^37s$ ,  $6d^27s7p$ ,  $6d7s^27p$ , and  $5f6d7s^2$  configurations. The results of that calculation allowed us to sort the configurations by their contributions to the energies of interest. Then we allowed two more excitations into the restricted valence space from the  $\sim 35$  most important configurations and one more excitation into a large valence space from the  $\sim 315$  most important configurations. The restricted valence space includes all orbitals with  $l < 5$  up to  $10g$  and the large valence space includes orbitals up to  $20d$ . The combined file that includes all of the configurations of the initial run is constructed and duplicate entries are removed. The resulting list includes 24 673 even and 28 651 odd CSFs. This algorithm for construction of the configuration space was tested previously on tetravalent Hf [43], where the results of this approach were compared with results of much larger calculations.

The differences between the preliminary and final runs are small only for a first few even states, ranging from 0 to  $300 \text{ cm}^{-1}$ . For most of the other states, the differences range between 900 and  $1500 \text{ cm}^{-1}$ , with the average difference being  $1250 \text{ cm}^{-1}$ . Moreover, the results of our preliminary runs show that lack of saturation of the CI space results in a  $1500\text{--}2000 \text{ cm}^{-1}$  shift of all  $5f6d7s^2$  and  $5f6d^27s$  configurations relative to the even  $7s^26d^2$  configuration. Selective expansion of the configuration space fixes this problem, resulting in the very good agreement between the energy levels of the  $5f6d7s^2$  and  $5f6d^27s$  configurations with experiment.

The shift of the even vs odd configurations involving an  $nf$  state is a well-known problem. For example, the energies of the  $4f^65d$  levels in Gd IV [52] were shifted by  $13\,500 \text{ cm}^{-1}$

relative to the  $4f^78S_{7/2}$  ground level to account for this problem. The procedure for correcting the shift of the  $4f^n5d$  energies relative to the ground state  $4f^{n+1}$  energies was used for Nd IV, Pm IV, Sm IV, and Eu IV ions [53].

#### D. Energies of Th ions

The energies and  $g$  factors of 38 even-parity and 31 odd-parity states of trivalent  $\text{Th}^+$  are listed in Table III. The table is structured in the same way as Table I. All values are counted from the  $6d^27s^2D_{3/2}$  ground-state energy. We note that Ref. [23] does not assign the ground-state  $LSJ$  term designation. The corresponding experimental and our calculated ground-state  $g$  factors listed in the first line of Table III,  $g_{\text{expt}} = 0.639$  and  $g_{\text{present}} = 0.662$ , are in between the nonrelativistic values 0.4 and 0.8 for the  $^4F_{3/2}$  and  $^2D_{3/2}$  terms, respectively. We assign the  $^2D_{3/2}$  term designation to the ground state of  $\text{Th}^+$ .

The 38 even-parity states listed in Table III belong to five configurations  $6d7s^2$ ,  $6d^27s$ ,  $6d^3$ ,  $5f^27s$ , and  $5f^26d$ , which are strongly mixed. Most odd-parity levels shown in Table III have a predominant composition of  $5f6d7s$ . The CI plus all-order results are in good agreement with experiment for most states, with the differences being less than 5% for the  $5f6d7s$  states. The larger discrepancies with experiment that are observed for even-parity levels with two  $5f$  electrons, such as  $5f^27s$  and  $5f^26d$ , are most likely due to the effects of correlations involving higher ( $l > 6$ ) partial waves. This problem is exacerbated when two  $5f$  electrons are present in the same configuration.

Energies and  $g$  factors for 95 levels of divalent  $\text{Th}^{2+}$  are listed in Table IV. The energies are counted from the ground state. The energies of the odd-parity  $5f6d$ ,  $5f7d5f7s$ ,  $5f8s$ ,  $6d6f$ , and  $6d7p$  configurations and even-parity  $6d^2$ ,  $5f^2$ ,  $7s^2$ ,  $5f7p$ ,  $5f6f$ , and  $6d7s$  configurations calculated with the CI plus all-order approach are compared with experimental energies [23] given in columns “Present” and “Expt.” of Table IV. We note that the ground state of  $\text{Th}^{2+}$  is  $5f6d^3H_4$  instead of the usual  $ns^2^1S_0$ , such as in isoelectronic  $\text{Ac}^+$  and  $\text{Ra}$ . The theoretical values agree well with experiment for most cases, with the exception of the  $5f6d^3F_2$  and  $6d^2^3F_2$  levels that are very close to the ground state. Since we calculate these energies as the differences of the large ground and excited divalent removal energies, the accuracy is reduced for such small energy intervals. Our values agree with experiment to 0.10%–1% for 38 levels.

In Table V we compare all results for levels for which theoretical calculations were performed both by us and by Berengut *et al.* [33], where a CI plus MBPT approach was used. Berengut *et al.* [33] noted that “while we believe the  $6d^2^3F_3$ ,  $6d^2^3F_4$ , and  $6d7s^3D_3$  transitions are accurate, the others are estimates only.” Comparison of the two theoretical results with experimental data [23] given in Table V shows that our results for  $\text{Th}^{2+}$  are in better agreement with experiment than the CI plus MBPT values except for the  $6d^2^3F_3$  and  $6d^2^3F_4$  levels. The case of  $\text{Th}^+$  is similar. The CI plus all-order method includes higher-order correlation beyond the CI plus MBPT approach. However, in some cases the higher-order terms may cancel with other contributions.

TABLE III. Levels (in  $\text{cm}^{-1}$ ) and  $g$  factors of the lowest states of once-ionized thorium. Nonrelativistic values of  $g$  factors  $g_{\text{nr}}$  are given by Eq. (3). Calculations are compared with experiment [12]. Configuration and term labels are determined as described in Sec. IV A.

Even-parity states ( $6d^27s + 6d^3 + 5f^27s + 5f^26d$ )									Odd-parity states ( $5f6d7s + 5f7s^2 + 5f6d^2$ )								
Conf.	Term		Energy		g factor			Conf.	Term		Energy		g factor				
	[23]	Present	$J$	Present	Expt.	Expt.	Present		$g_{\text{nr}}$	[23]	Present	$J$	Present	Expt.	Expt.	Present	$g_{\text{nr}}$
$6d^27s$		$^2D$	3/2	0	0	0.639	0.662	0.800	$5f6d7s$	$^4D$	$^4D$	1/2	11550	11725	0.255	0.239	0.000
$6d^27s$	$^4F$	$^4F$	3/2	1948	1859	0.586	0.554	0.400	$5f6d7s$		$^2P$	1/2	13965	14102	0.523	0.552	0.667
$6d^3$		$^2D$	3/2	7779	7001	0.800	0.806	0.800	$5f6d7s$	$^4P$	$^4P$	1/2	15401	15324	2.565	2.567	2.667
$6d^27s$	$^4P$	$^4P$	3/2	8622	8018	1.608	1.608	1.733	$5f6d7s$		$^2S$	1/2	17538	17838	1.08	1.582	2.000
$6d^3$		$^2D$	3/2	9209	8460	0.968	0.945	0.800									
$6d^27s$	$^2D$	$^4D$	3/2	13270	12220	0.977	0.946	1.200	$5f6d7s$	$^4F$	$^4F$	3/2	6020	6691	0.492	0.510	0.400
$6d^3$	$^4P$	$^4P$	3/2	16503	15237	1.592	1.612	1.733	$5f6d7s$	$^2D$	$^2D$	3/2	10695	11576	0.832	0.754	0.800
$6d^3$		$^2D$	3/2	19973	18119	0.93	0.862	0.800	$5f6d7s$		$^4D$	3/2	12657	12902	1.167	1.184	1.200
									$5f6d7s$		$^2P$	3/2	15198	15145	1.366	1.278	1.333
$6d^27s$	$^4P$	$^4P$	1/2	6972	6244	2.112	2.144	2.667	$5f6d7s$	$^2D$	$^4D$	3/2	15676	15711	1.06	1.162	1.200
$6d^27s$		$^2S$	1/2	8509	7828	1.254	1.201	2.000									
$6d^3$	$^4P$	$^4P$	1/2	15632	14349	2.555	2.564	2.667	$5f7s^2$	$^2F$	$^2F$	5/2	3882	4490	0.856	0.853	0.857
									$5f6d7s$	$^4F$	$^4F$	5/2	6650	7331	1.061	1.070	1.029
$6d^27s$	$^4F$	$^4F$	5/2	1722	1522	1.076	1.070	1.029	$5f6d7s$	$^4G$	$^4G$	5/2	9299	9585	0.601	0.606	0.571
$6d^27s$		$^2D$	5/2	4185	4113	1.163	1.150	1.200	$5f6d7s$		$^4F$	5/2	9978	10673	1.088	1.089	1.029
$6d^27s$	$^2F$	$^2F$	5/2	9198	8606	0.986	0.982	0.857	$5f6d7s$	$^2F$	$^4F$	5/2	12045	12472	0.982	0.935	1.029
$6d^27s$	$^4P$	$^4P$	5/2	9680	9061	1.419	1.408	1.600	$5f6d7s$	$^4D$	$^4D$	5/2	14310	14546	1.339	1.346	1.371
$6d^3$		$^4F$	5/2	10145	9401	1.034	1.035	1.029									
$6d^27s$		$^2D$	5/2	14220	13251	1.245	1.235	1.200	$5f6d7s$	$^4H$	$^4H$	7/2	5743	6168	0.718	0.729	0.667
$6d^3$	$^4P$	$^4P$	5/2	17100	15787	1.571	1.566	1.600	$5f7s^2$	$^2F$	$^2F$	7/2	7794	8379	1.132	1.127	1.143
$6d^3$	$^2D$	$^4D$	5/2	22144	20159	1.19	1.189	1.371	$5f6d7s$	$^2G$	$^2G$	7/2	8681	9202	0.911	0.899	0.889
									$5f6d7s$	$^4F$	$^4F$	7/2	9304	9720	1.173	1.167	1.238
$6d^27s$	$^4F$	$^4F$	7/2	4374	4147	1.232	1.227	1.238	$5f6d7s$	$^4G$	$^4G$	7/2	10751	11117	0.983	0.977	0.984
$6d^27s$	$^2G$	$^4G$	7/2	10502	9712	0.953	0.947	0.984	$5f6d^2$	$^2F$	$^2F$	7/2	13270	12486	0.855	1.036	1.143
$6d^3$		$^2F$	7/2	11621	10855	1.166	1.171	1.143									
$6d^3$		$^2F$	7/2	13297	12570	1.131	1.122	1.143	$5f6d7s$	$^4H$	$^4H$	9/2	6265	6700	1.018	1.025	0.970
$6d^3$	$^2G$	$^2G$	7/2	18247	16818	0.916	0.906	0.889	$5f6d7s$	$^2G$	$^2G$	9/2	8818	9238	1.086	1.068	1.111
$6d^3$	$^2F$	$^2F$	7/2	25165	22834	1.12	1.132	1.143	$5f6d7s$	$^4H$	$^4H$	9/2	10435	10572	0.931	0.927	0.970
									$5f6d7s$	$^4F$	$^4F$	9/2	12076	12488	1.245	1.253	1.333
$6d^27s$	$^4F$	$^4F$	9/2	6528	6213	1.312	1.309	1.333	$5f6d7s$	$^4G$	$^4G$	9/2	13156	13469	1.185	1.178	1.172
$6d^27s$		$^2G$	9/2	11158	10379	1.153	1.145	1.111	$5f6d^2$	$^4H$	$^2H$	9/2	14421	15243	1.00	0.839	0.909
$6d^3$	$^4F$	$^4F$	9/2	14217	13249	1.242	1.256	1.333									
$6d^3$	$^2H$	$^4H$	9/2	16585	15305	1.006	0.983	0.970	$5f6d7s$	$^4H$	$^4H$	11/2	9953	10189	1.128	1.121	1.133
$6d^3$	$^2G$	$^2G$	9/2	21328	19880	1.08	1.075	1.111	$5f6d7s$	$^4G$	$^2H$	11/2	14390	15350	1.267	1.085	1.091
$5f^27s$		$^2H$	9/2	30481	25246	0.96	0.939	0.909	$5f6d^2$	$^4I$	$^4G$	11/2	15111	16565	0.98	1.261	1.273
									$5f6d^2$	$^4H$	$^4I$	11/2	16717	17771	1.10	0.966	0.965
$6d^3$	$^2H$	$^4H$	11/2	19039	17727	1.09	1.086	1.091									
$5f^27s$	$^4H$	$^2H$	11/2	33216	27937	1.12	1.118	1.133									
$5f^27s$	$^2H$	$^4H$	11/2	35702	30485	1.08	1.085	1.091									
$5f^27s$	$^4K$	$^2I$	11/2	37981	32621	0.826	0.885	0.923									
$5f^27s$	$^4H$	$^4H$	13/2	35827	30549	1.23	1.221	1.231									
$5f^26d$	$^4K$	$^4K$	13/2	41108	35401	0.98	0.973	0.964									
$5f^27s$	$^2I$	$^4I$	13/2	42291	37575	1.088	1.120	1.108									

The CI plus MBPT approach was used by Porsev and Flambaum [31] to evaluate energies and  $g$  factors in  $\text{Th}^+$ . Tabulated results were given for even-parity states with  $J = 3/2$  and  $5/2$  in the range from 18 119 to 40 644  $\text{cm}^{-1}$ . Since our calculation was carried out for lower levels, we can only compare results for two states  $6d^3 \ ^2D_{3/2}$  and  $6d^3 \ ^4D_{5/2}$ . The CI plus MBPT results of [31] differ from the experimental values by 18% and our CI plus all-order values by 10%.

## V. MULTIPOLE TRANSITION AMPLITUDES AND LIFETIMES IN Ra-LIKE $\text{Th}^{2+}$

We now discuss some multipole transition amplitudes in  $\text{Th}^{2+}$  that are representative of the calculations that may have to be done for the investigation of the electronic bridge process [11]. Our CI plus all-order results for the multipole matrix elements, transition rates, and lifetimes in Ra-like  $\text{Th}^{2+}$  are given in Table VI.

TABLE IV. Levels (in  $\text{cm}^{-1}$ ) and  $g$  factors of the lowest states of twice-ionized thorium. Nonrelativistic values of  $g$  factors  $g_{\text{nr}}$  are given by Eq. (3). Calculations are compared with experiment [12]. Configuration and term labels are determined as described in Sec. IV A.

Odd-parity states ( $5f6d + 5f7d + 5f7s + 5f8s + 6d6f + 6d7p$ )									Even-parity states ( $6d^2 + 5f^2 + 7s^2 + 5f7p + 5f6f + 6d7s$ )								
Conf.	[23]	Term		Energy		g factors			Conf.	[23]	Term		Energy		g factors		
		Present	$J$	Present	Expt.	Expt.	Present	$g_{\text{nr}}$			Present	$J$	Present	Expt.	Expt.	Present	$g_{\text{nr}}$
$5f6d$	$^3H$	$^3H$	4	0	0.0	0.885	0.894	0.800	$6d^2$	$^3P$		0	6151	5090	0.00	0.000	0.000
$5f6d$	$^3F$	$^1G$	4	3207	3188	0.976	0.949	1.000	$7s^2$	$^1S$		0	12428	11961	0.00	0.000	0.000
$5f7s$	$^3F$	$^3F$	4	6237	6311	1.22	1.205	1.250	$6d^2$	$^1S$		0	22008	18993	0.00	0.000	0.000
$5f6d$	$^3G$	$^3G$	4	8197	8142	1.103	1.096	1.050	$5f^2$	$^3P$		0	29579	29299	0.00	0.000	0.000
$5f6d$	$^3H$	$^3F$	4	9063	8981	1.188	1.178	1.250	$5f^2$	$^1S$		0	55356	51161	0.00	0.000	0.000
$6d7p$		$^3F$	4	54333	53052	1.27	1.243	1.250				0	84509		0.00	0.000	0.000
$5f7d$		$^3H$	4	79062	78417		0.891	0.800	$5f6f$			0	89234	88313	0.00	0.000	0.000
$5f8s$		$^3F$	4	79519	78930		1.210	1.250				0	94112		0.00	0.000	0.000
$5f6d$	$^3P$		0	11766	11233	0.0	0.000	0.000	$6d7s$	$^3D$	$^3D$	1	6137	5524	0.50	0.499	0.500
$7s7p$	$^3P$		0	43188	42260	0.0	0.000	0.000	$6d^2$	$^3P$	$^3P$	1	8905	7876	1.50	1.491	1.500
$6d7p$			0	52776	51745	0.0	0.000	0.000	$5f^2$	$^3P$	$^3P$	1	30636	30402	1.494	1.492	1.500
$5f7d$			0	81644	80906	0.0	0.000	0.000	$5f7p$		$^3D$	1	44946	44603	0.495	0.497	0.500
$5f6d$	$^3D$	$^3D$	1	8260	7921	0.621	0.599	0.500	$6d^2$	$^3F$	$^3F$	2	895	63	0.744	0.756	0.667
$5f6d$	$^3P$	$^3P$	1	11564	11123	1.352	1.376	1.500	$6d^2$		$^1D$	2	5426	4676	1.020	1.003	1.000
$5f6d$	$^1P$	$^1P$	1	22733	20711		1.006	1.000	$6d7s$	$^3D$	$^3D$	2	7943	7176	1.180	1.155	1.167
$6d7p$		$^1P$	1	40282	39281	0.911	0.902	1.000	$6d^2$	$^3P$	$^3P$	2	11417	10440	1.36	1.370	1.500
$7s7p$	$^3P$	$^3P$	1	45938	45064	1.120	1.109	1.500	$6d7s$	$^1D$	$^1D$	2	16438	16037	1.00	0.961	1.000
$6d7p$		$^3P$	1	51875	50993	1.22	1.202	1.500	$5f^2$	$^3F$	$^3F$	2	18616	18864	0.694	0.738	0.667
$6d7p$		$^3P$	1	55035	53939	1.25	1.248	1.500	$5f^2$	$^1D$	$^3D$	2	28971	28233	1.12	1.164	1.167
$7s7p$	$^1P$	$^1P$	1	69930	69001		1.012	1.000	$5f^2$	$^3P$	$^3P$	2	33488	32867	1.344	1.300	1.500
$5f6d$	$^3F$	$^3F$	2	189	511	0.711	0.765	0.667	$6d^2$	$^3F$	$^3F$	3	4938	4056	1.083	1.078	1.083
$5f6d$	$^3F$	$^3F$	2	2958	3181	0.725	0.732	0.667	$6d7s$	$^3D$	$^3D$	3	10641	9954	1.339	1.325	1.333
$5f6d$	$J$	$^1D$	2	5797	6288	0.908	0.858	1.000	$5f^2$	$^3F$	$^3F$	3	20378	20840	1.096	1.078	1.083
$5f6d$	$^3D$	$^3D$	2	10458	10181	1.19	1.181	1.167	$5f7p$		$^3G$	3	33715	33562	0.849	0.837	0.750
$5f6d$	$^3P$	$^3P$	2	13513	13208	1.432	1.436	1.500	$5f7p$		$^3F$	3	38736	38432	1.170	1.149	1.083
$6d7p$		$^3F$	2	38322	37280	0.795	0.782	0.667	$5f7p$		$^1F$	3	42544	42313	0.971	0.960	1.000
$6d7p$		$^3D$	2	45159	44088	1.200	1.187	1.167	$5f7p$		$^3F$	3	47876	47472	1.202	1.199	1.083
$6d7p$		$^1D$	2	48723	47680	1.02	1.002	1.000	$6d7d$		$^1F$	3	84838	83702		0.906	1.000
$5f7s$		$^3F$	3	2436	2527	1.071	1.052	1.083	$6d^2$	$^3F$	$^3F$	4	7264	6538	1.20	1.161	1.250
$5f6d$		$^1F$	3	4853	4827	1.003	0.988	1.000	$6d^2$	$^1G$	$^3G$	4	10822	10543	1.05	1.064	1.050
$5f6d$	$^3G$	$^3G$	3	5085	5061	0.869	0.858	0.750	$5f^2$	$^3H$	$^3H$	4	14514	15149	0.81	0.814	0.800
$5f7s$		$^1F$	3	7609	7501	1.027	1.015	1.000	$5f^2$	$^3F$	$^3F$	4	21782	21784	1.18	1.201	1.250
$5f6d$	$^3D$	$^3D$	3	11236	10741	1.22	1.244	1.333	$5d2$	$^1G$	$^1G$	4	27045	25972	1.072	1.033	1.000
$5f6d$	$^1F$	$^3F$	3	16506	15453	1.07	1.060	1.083	$5f7p$		$^3G$	4	38980	38581	1.105	1.100	1.050
$6d7p$		$^3F$	3	45686	44465	1.125	1.121	1.083	$5f7p$		$^3G$	4	44034	43702	1.069	1.068	1.050
$6d7p$		$^3D$	3	51059	49981	1.19	1.188	1.333	$5f7p$		$^3F$	4	47745	47261	1.14	1.114	1.250
$5f6d$	$^3H$	$^3H$	5	4802	4490	1.04	1.028	1.033	$5f^2$	$^3H$	$^3H$	5	17131	17888	1.01	1.028	1.033
$5f6d$	$^3G$	$^3G$	5	11456	11277	1.186	1.187	1.200	$5f7p$		$^3G$	5	47781	47422	1.207	1.194	1.200
$5f6d$	$^1H$	$^1H$	5	20144	19009	1.001	1.001	1.000	$5f6f$		$^3I$	5	87443	86934		0.887	0.833
$5f7d$		$^1H$	5	80841	80137		1.033	1.000	$5f6f$		$^1H$	5	88176	87667		1.006	1.000
$5f7d$		$^3G$	5	83847	83023		1.137	1.200	$6d7d$		$^3G$	5	90979	90085		1.141	1.200
$5f7d$		$^3H$	5	85158	84239		1.047	1.033	$5f6f$		$^3H$	5	92840	92103		1.020	1.033
$6d6f$		$^3H$	5	97802	96317		1.049	1.033	$5f8p$		$^3G$	5	94046	94144		1.186	1.200
$5f6d$	$^3H$	$^3H$	6	8810	8437	1.17	1.160	1.167	$5f^2$	$^3H$	$^3H$	6	20123	20771	1.16	1.157	1.167
$5f7d$		$^3H$	6	84721	83963		1.161	1.167	$5f^2$	$^1I$	$^1I$	6	28635	28350		0.999	1.000
		$^3H$	6	102352			1.160	1.167	$5f6f$		$^3I$	6	88845	88387		1.021	1.024
		$^3I$	6	108394			1.032	1.024	$5f6f$		$^3H$	6	93648	93045		1.101	1.167
		$^3K$	6	108614			0.868	0.857	$5f6f$		$^3I$	6	94584	94018		1.052	1.024

TABLE V. Levels (in  $\text{cm}^{-1}$ ) of the lowest states of once- and twice-ionized thorium. All energy values are given relative to the respective ground state. Calculations are compared with experiment [12] and other theory [33].

Th <sup>+</sup>	Term	Energies			Th <sup>2+</sup>	Term	Energies		
		Present	Expt. [12]	Theory [33]			Present	Expt. [12]	Theory [33]
$5f7s^2$	$^2F_{5/2}$	3882	4490	4856	$6d^2$	$^3F_3$	4938	4056	4023
$5f6d7s$	$^4F_{3/2}$	6020	6691	7487	$6d^2$	$^3F_4$	7264	6538	6795
$5f6d7s$	$^4F_{5/2}$	6651	7331	8325	$6d7s$	$^3D_3$	10641	9954	9204
$5f6d7s$	$^4G_{5/2}$	9229	9585	10045	$6d^2$	$^1G_4$	10822	10543	11051
$5f6d7s$	$^4H_{5/2}$	9978	10673	12168	$5f^2$	$^3H_4$	14514	15149	13358
$5f6d7s$	$^2D_{3/2}$	10695	11576	13054	$5f^2$	$^3H_5$	17131	17887	16068
$5f6d7s$	$^4D_{1/2}$	11575	11725	12897	$5f^2$	$^3F_3$	20378	20840	19080
$5f6d7s$	$^2F_{5/2}$	12045	12472	14564	$5f^2$	$^3F_4$	21782	21784	20366
$5f6d7s$	$^4F_{3/2}$	12657	12902	14233	$5f^2$	$^1G_4$	27045	25972	25269
$5f6d7s$	$^4G_{1/2}$	13965	14102	15853	$5f7p$	$^3G_3$	33715	33562	33402
					$5f7p$	$^3F_3$	38736	38432	38617

We evaluate multipole matrix elements between 12 odd-parity states with energies in the 0–8142  $\text{cm}^{-1}$  range with 12 even-parity states with energies in the 63–11 961  $\text{cm}^{-1}$  range. This results in 45  $E1$ , 66  $M2$ , and 82  $E3$  transitions between the odd-even and even-odd states. We evaluate also the 83  $M1$ ,  $E2$ , and  $M3$  transitions inside the even-parity complex, as well as the 110  $M1$ ,  $E2$ , and  $M3$  transitions inside the odd-parity complex. That gives us 386 multipole matrix elements for transitions between lowest-lying levels in the  $\text{Th}^{2+}$  ion.

In Table VI we include results for 45 selected electric multipole ( $E1$  and  $E2$ ) and magnetic multipole ( $M1$  and  $M2$ ) transitions that are most important for the evaluation of the corresponding lifetimes. The octupole ( $E3$  and  $M3$ ) transitions make negligible contributions to the lifetimes and are omitted.

We use atomic units to express all transition matrix elements throughout this section: The numerical values of the elementary charge  $e$ , the reduced Planck constant  $\hbar = h/2\pi$ , and the electron mass  $m_e$  are set equal to 1. The atomic unit for the electric dipole matrix element is  $ea_0$ , where  $a_0$  is the Bohr radius.

To show the importance of using effective transition operators (for example, electric dipole  $D^{\text{eff}}$ ), which include random-phase-approximation (RPA) corrections, instead of the bare operators we give the matrix elements with and without the RPA correction in columns labeled  $Z^{\text{CI+all-order}}$  and  $Z^{\text{noRPA}}$ , respectively. We find that the RPA correction is significant (20%–50%) for most transitions, so the  $Z^{\text{CI+all-order}}$  final values are used in calculating transition rates and lifetimes. The RPA correction is small for  $M1$  matrix elements  $5f7s^3F_3-5f7s^3F_2$  and  $6d^2^3P_1-7s^2^1S_0$ .

The  $E1$ ,  $E2$ ,  $E3$ ,  $M2$ ,  $M3$ , and  $M3$  transition probabilities  $A_r$  ( $\text{s}^{-1}$ ) are obtained in terms of line strengths  $S$  (a.u.) and energies  $\mathcal{E}$  (a.u.) as

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

$$C^{(1)} = 2.14200 \times 10^{10},$$

$$C^{(2)} = 5.70322 \times 10^4,$$

$$C^{(3)} = 7.71311 \times 10^{-2},$$
(8)

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

$$D^{(1)} = 2.85161 \times 10^5,$$

$$D^{(2)} = 7.59260 \times 10^{-1},$$

$$D^{(3)} = 1.02683 \times 10^{-6}.$$
(9)

The line strengths  $S(E1)$ ,  $S(E2)$ ,  $S(E1)$ ,  $S(E3)$ ,  $S(M1)$ ,  $S(M2)$ , and  $S(M3)$  are obtained as squares of the corresponding  $E1$ ,  $E2$ ,  $E3$ ,  $M2$ ,  $M3$ , and  $M3$  matrix elements listed in column  $Z^{\text{CI+all}}$  of Table VI. Energies are from the experimental compilation of Ref. [23]. We list the experimental energies for upper and lower states as well as the corresponding transition wavelengths  $\lambda$  in Table VI for reference. Our results for the transition rates are given in column  $A_r^{\text{CI+all}}$  of Table VI.

In order to determine the lifetimes listed in the last column of Table VI, we sum over all possible radiative transitions. The number of contributing transitions increases significantly for higher levels. For example, 18 transitions contribute to the lifetime of the relatively low-lying  $5f7s^3F_4$  state  $E(5f7s^3F_4) = 6310.81 \text{ cm}^{-1}$ . However, only one transition  $6d^2^3F_3-5f7s^3F_4$  contributes significantly and the total contribution of other 17 transitions to the  $5f7s^3F_4$  lifetime is equal to 0.1%. The final values of  $\tau^{\text{CI+all}}$  for the 23 lowest-lying levels are listed in the last column of Table VI. The term designation for those levels are in the first column of Table VI. In Table VII we present results for other  $E1$  transitions for low-lying levels with smaller transition rates.

Unfortunately, we did not find any theoretical or experimental results to compare with our  $A_r$  and  $\tau$  values for the low-lying states listed in Table VI. Experimental measurements of lifetimes for six higher levels of  $\text{Th}^{2+}$  were performed by Biémont *et al.* [22] and are summarized in Table VIII. In order to calculate these lifetimes with our CI plus all-order method, we carried out extensive additional calculations to obtain the wave functions and corresponding electric dipole matrix elements for 112 levels with  $0 \leq J \leq 6$ . Using the calculated CI plus all-order electric dipole matrix elements and experimental energies [23], we obtain values for 1152



TABLE VI. Lifetimes  $\tau^{\text{CI+all}}$  (in s), transition rates  $A_r$  (in  $\text{s}^{-1}$ ), and reduced matrix elements  $Z^{\text{CI+all}}$  (in a.u.) for electric multipole ( $E1$  and  $E2$ ) and magnetic multipole ( $M1$  and  $M2$ ) transitions in the Ra-like  $\text{Th}^{2+}$  ion evaluated in the CI plus all approximation. Energies (in  $\text{cm}^{-1}$ ) are from Ref. [23]. The numbers in square brackets represent powers of 10.

Transition		Energies ( $\text{cm}^{-1}$ )		$\lambda$	$Z^{\text{no RPA}}$	$Z^{\text{CI+all}}$	$A_r^{\text{CI+all}}$	$\tau^{\text{CI+all}}$	
Upper	Lower	Lower	Upper	( $\text{\AA}$ )	(a.u.)	(a.u.)	( $\text{s}^{-1}$ )	(s)	
$6d^2^3F_2$	$5f6d^3H_4$	$M2$	0.00	63.27	1580528	2.1134	1.4359	6.224[-19]	1.607[18]
$5f6d^3F_2$	$5f6d^3H_4$	$E2$	0.00	510.76	195787	3.6338	3.1838	7.901[-9]	1.266[8]
$5f7s^3F_3$	$6d^2^3F_2$	$E1$	63.27	2527.09	40587	1.1061	0.7668	2.546[3]	3.928[-4]
$5f7s^3F_2$	$5f6d^3H_4$	$E2$	0.00	3181.50	31432	11.1410	10.6310	8.251[-4]	3.038[2]
	$5f7s^3F_3$	$M1$	2527.09	3181.50	152809	1.4806	1.4766	3.292[-3]	
$5f6d^1G_4$	$5f6d^3H_4$	$E2$	0.00	3188.30	31365	5.4527	5.0061	1.028[-4]	1.740[3]
	$5f6d^3F_2$	$E2$	510.87	3188.30	37349	3.5556	3.4795	2.073[-5]	
	$5f7s^3F_3$	$M1$	2527.09	3188.50	151192	0.7239	0.7220	4.513[-4]	
$6d^2^3F_3$	$5f6d^3H_4$	$E1$	0.00	4056.02	24655	0.0662	0.0214	8.841[0]	1.235[-3]
	$5f6d^3F_2$	$E1$	510.76	4056.02	28207	0.3400	0.2483	7.953[2]	
$5f6d^3H_5$	$5f6d^3H_4$	$M1$	0.00	4489.64	22274	2.4360	2.4438	1.325[0]	7.410[-1]
	$5f6d^1G_4$	$M1$	3188.30	4489.64	76844	2.1292	2.1303	2.453[-2]	
$5f6d^3F_3$	$6d^2^3F_2$	$E1$	63.27	4826.83	20993	1.1113	0.7438	1.731[4]	5.777[-5]
$6d^2^3F_2$	$5f7s^3F_3$	$E1$	2327.09	4676.43	42565	0.9413	0.5569	1.629[3]	6.139[-4]
$5f6d^3G_3$	$6d^2^3F_2$	$E1$	63.27	5061.54	20007	2.1735	1.4623	7.766[4]	1.288[-5]
$6d^2^3P_0$	$6d^2^3F_2$	$E2$	63.27	5090.06	19893	5.8665	5.3985	1.047[-2]	9.555[1]
$6d7s^3D_1$	$5f6d^3F_2$	$E1$	510.76	5523.88	19948	0.7040	0.4896	2.040[4]	4.159[-5]
	$5f7s^3F_2$	$E1$	3181.50	5523.88	42692	1.1268	0.6477	3.642[3]	
$5f6d^1D_2$	$6d^2^3F_3$	$E1$	4056.02	6288.42	44795	0.1510	0.0727	2.379[1]	4.187[-2]
$5f7s^3F_4$	$6d^2^3F_3$	$E1$	4056.02	6310.81	44350	0.7621	0.5514	7.849[2]	1.273[-3]
$6d^2^3F_4$	$5f7s^3F_3$	$E1$	2527.09	6537.78	24933	0.2457	0.2126	6.568[2]	1.131[-3]
	$5f6d^3F_3$	$E1$	4826.83	6537.81	58446	0.3792	0.3273	1.208[2]	
	$5f6d^3G_3$	$E1$	5060.94	6537.81	67711	0.5535	0.3609	9.279[1]	
$6d7s^3D_2$	$5f7s^3F_3$	$E1$	2527.09	7176.11	21510	0.9399	0.5919	1.426[4]	6.720[-5]
	$5f6d^3F_3$	$E1$	4826.83	7176.11	42566	0.2150	0.3040	6.203[2]	
$5f7s^3F_3$	$6d^2^3F_2$	$E1$	63.27	7500.61	13446	0.1114	0.0762	6.907[2]	3.182[-4]
	$6d^2^3F_2$	$E1$	4676.43	7500.61	35409	0.8405	0.6070	2.403[3]	
	$6d^2^3F_4$	$E1$	6537.81	7500.61	103864	0.4824	0.4054	4.249[1]	
$6d^2^3P_1$	$5f6d^3F_2$	$E1$	510.76	7875.83	13578	0.1028	0.0840	1.902[3]	4.898[-4]
	$5f7s^3F_2$	$E1$	3181.50	7875.83	21302	0.0712	0.0446	1.392[2]	
$5f6d^3G_4$	$6d^2^3F_3$	$E1$	4056.02	8141.75	24475	2.7360	1.8865	5.465[4]	1.830[-5]
$6d7s^3D_3$	$5f6d^3H_4$	$E1$	0.00	9953.58	10047	0.2032	0.1630	7.579[3]	2.666[-5]
	$5f6d^1G_4$	$E1$	3188.50	9953.58	14782	0.4667	0.3538	1.122[4]	
	$5f7s^3F_4$	$E1$	6310.81	9953.58	27452	1.6388	1.0983	1.687[4]	
$6d^2^3P_2$	$5f6d^3F_3$	$E1$	4826.83	10440.24	17814	0.1880	0.1106	8.772[2]	1.982[-4]
	$5f6d^3G_3$	$E1$	5060.54	10440.24	18588	0.2160	0.1468	1.353[3]	
	$5f7s^3F_3$	$E1$	7500.61	10440.24	34018	0.7666	0.5082	2.658[3]	
$6d^2^1G_4$	$5f6d^3F_3$	$E1$	4826.83	10542.90	17495	1.0369	0.7953	2.949[4]	2.366[-5]
	$5f6d^3G_3$	$E1$	5060.54	10542.90	18240	0.7825	0.5208	1.001[4]	
$7s^2^1S_0$	$6d^2^3F_2$	$E2$	4676.43	11961.13	13727	7.0555	6.7483	1.046[-1]	1.418[0]
	$6d^2^3P_1$	$M1$	7875.83	11961.13	24478	0.5484	0.5504	5.572[-1]	

transition rates  $A_r$ . These are summed over all transitions to determine the lifetimes for about 100 levels.

In Table VIII we present lifetimes  $\tau^{\text{CI+all}}$ , transition rates  $A_r$ , and branching ratios of electric dipole transitions relevant to the comparison with the lifetime measurements of [22]. Energies in both tables are from the compilation of Ref. [23]. Only the dominant transitions are listed, but all transitions are included in the lifetime calculation.

In the two last columns of Table VIII we compare our CI plus all-order lifetimes with measurements [22].

We find excellent agreement between the CI plus all-order lifetimes and experimental values for the  $5f7p^3G_4$ ,  $7s7p^3P_0$ ,  $7s7p^3P_1$ , and  $6d7p^3F_4$  levels. There is a 15% difference for the lifetime of the  $5f^2^3P_2$  level. Several channels contribute significantly to this lifetime. The largest branching ratio for this level is 0.31 for the  $5f6d^3P_2-5f^2^3P_2$  transition. The branching ratios for the other five transitions shown in Table VIII add 56%. An additional 13% comes from eight transitions that are not shown in Table VIII.

TABLE VII. Oscillator strengths  $f$  and transition rates  $A_r$  (in  $\text{s}^{-1}$ ) for electric dipole transitions in the Ra-like  $\text{Th}^{2+}$  ion. Wavelengths (in  $\text{\AA}$ ) from the compilation of Ref. [23] are listed for reference. The numbers in square brackets represent powers of 10.

Transition		Wavelength ( $\text{\AA}$ )	$f^{\text{CI+all}}$	$A_r^{\text{CI+all}}$ ( $\text{s}^{-1}$ )
Lower	Upper			
$5f6d^3H_4$	$6d^2^3G_4$	9485.6	3.733[-2]	3.417[4]
$6d^2^3F_2$	$5f6d^3D_1$	12726.7	1.651[-2]	4.532[4]
$5f7s^3F_3$	$6d7s^3D_3$	13465.8	1.664[-2]	1.250[4]
$5f6d^3F_2$	$6d7s^3D_2$	15003.5	9.591[-3]	1.137[4]
$5f6d^3H_4$	$6d^2^3F_4$	15296.1	6.768[-3]	2.383[3]
$6d^2^1D_2$	$5f6d^3P_1$	15512.2	6.619[-3]	1.223[4]
$6d^2^3F_2$	$5f6d^1D_2$	16064.4	1.080[-3]	1.117[3]
$6d^2^3F_3$	$5f6d^3D_2$	16327.7	3.118[-2]	2.229[4]
$6d^2^1D_2$	$5f6d^3D_3$	16489.3	6.448[-2]	4.520[4]
$6d7s^3D_1$	$5f6d^3P_0$	17515.8	4.814[-3]	3.488[4]
$6d7s^3D_1$	$5f6d^3P_1$	17859.9	4.407[-3]	1.024[4]
$6d^2^3F_2$	$5f6d^3G_3$	20011.4	1.625[-1]	7.737[4]
$6d^2^3F_3$	$5f6d^3F_4$	20307.0	9.844[-2]	2.528[4]
$6d^2^3F_4$	$5f6d^3G_5$	21102.0	5.705[-1]	8.633[4]
$6d^2^3F_4$	$5f6d^3D_3$	23791.1	1.139[-1]	2.131[4]
$5f6d^3F_2$	$6d^2^1D_2$	24006.2	8.657[-3]	4.007[3]
$5f6d^3D_1$	$7s^2^1S_0$	24752.7	7.869[-4]	2.857[3]
$5f6d^3F_2$	$6d^2^3F_3$	28207.2	3.317[-3]	7.947[2]
$5f6d^1G_4$	$6d^2^3F_4$	29855.6	2.675[-2]	2.473[3]
$6d^2^3F_2$	$5f6d^3F_2$	32070.0	1.603[-3]	4.159[2]
$6d^2^1D_2$	$5f7s^1F_3$	35396.5	1.569[-2]	2.386[3]
$5f7s^3F_3$	$6d^2^1D_2$	46526.4	1.439[-2]	1.266[3]
$6d^2^1D_2$	$5f6d^1D_2$	62035.6	1.691[-3]	1.172[2]
$5f7s^3F_3$	$6d^2^3F_3$	65405.7	1.286[-2]	4.094[2]
$6d^2^3F_2$	$5f6d^3F_2$	223469.2	3.053[-3]	1.637[1]

TABLE VIII. Lifetimes (in ns), transition rates  $A_r$  (in  $\text{s}^{-1}$ ), and branching ratios of electric dipole transitions in Ra-like  $\text{Th}^{2+}$ . Experimental lifetimes are taken from Ref. [22]. Levels (in  $\text{cm}^{-1}$ ) are from the experimental compilation of Ref. [23]. The numbers in square brackets represent powers of 10.

Transition		Levels ( $\text{cm}^{-1}$ )		$\lambda$ ( $\text{\AA}$ )	$Z^{\text{CI+all}}$ (a.u.)	$A_r^{\text{CI+all}}$ ( $\text{s}^{-1}$ )	Branching ratio	$\tau^{\text{CI+all}}$ (ns)	$\tau^{\text{Expt}}$ (ns)
Upper	Lower	Upper	Lower						
$5f^2^3P_2$	$5f6d^3F_2$	32867.27	510.76	3090.6	0.8137	9.09[6]	0.19	21.2	$25.8 \pm 1.5$
$5f^2^3P_2$	$5f6d^1D_2$	32867.27	6288.42	3762.4	0.7188	3.93[6]	0.08		
$5f^2^3P_2$	$5f6d^3D_3$	32867.27	10741.15	4519.5	1.0101	4.48[6]	0.10		
$5f^2^3P_2$	$5f6d^3P_1$	32867.27	11123.18	4599.0	0.8850	3.26[6]	0.07		
$5f^2^3P_2$	$5f6d^3P_2$	32867.27	13208.21	5086.7	2.1928	1.48[7]	0.31		
$5f^2^3P_2$	$5f6d^3F_3$	32867.27	15453.41	5742.6	1.6403	5.76[6]	0.12		
$5f7p^3G_4$	$5f6d^1G_4$	38580.60	3188.30	2825.5	2.0580	4.23[7]	0.10	2.41	$2.7 \pm 0.2$
$5f7p^3G_4$	$5f6d^3H_5$	38580.60	4489.64	2933.3	3.1200	8.68[7]	0.21		
$5f7p^3G_4$	$5f7s^3F_4$	38580.60	6310.81	3098.9	3.8614	1.13[8]	0.27		
$5f7p^3G_4$	$5f7s^1F_3$	38580.60	7500.61	3217.5	3.2880	7.31[7]	0.18		
$5f7p^3G_4$	$5f6d^3F_4$	38580.60	8980.56	3378.4	2.5555	3.81[7]	0.09		
$7s7p^3P_0$	$6d7s^3D_1$	42259.71	5523.88	2722.1	1.2104	1.47[8]	0.91	6.19	$6.6 \pm 0.4$
$7s7p^3P_0$	$6d^2^3P_1$	42259.71	7875.83	2908.3	0.4150	1.42[7]	0.09		
$7s7p^3P_1$	$6d^2^3F_2$	45063.97	63.27	2222.2	1.0592	6.91[7]	0.15	2.22	$2.4 \pm 0.2$
$7s7p^3P_1$	$6d^2^1D_2$	45063.97	4676.43	2476.0	2.2857	2.33[8]	0.52		
$7s7p^3P_1$	$6d7s^3D_1$	45063.97	5523.88	2529.1	0.7118	2.12[7]	0.05		
$7s7p^3P_1$	$6d7s^3D_2$	45063.97	7176.11	2639.4	1.4716	7.96[7]	0.18		
$7s7p^3P_1$	$7s^2^1S_0$	45063.97	11961.13	3020.9	0.8471	1.76[7]	0.04		
$6d7p^3F_4$	$6d^2^3F_3$	53052.47	4056.02	2041.0	0.7944	1.67[7]	0.02	1.41	$1.3 \pm 0.2$
$6d7p^3F_4$	$6d^2^3F_4$	53052.47	6537.81	2149.9	2.3177	1.22[8]	0.17		
$6d7p^3F_4$	$6d7s^3D_3$	53052.47	9953.58	2320.2	5.3100	5.08[8]	0.72		
$6d7p^3F_4$	$6d^2^3G_4$	53052.47	10542.90	2352.4	1.7656	5.39[7]	0.08		

## VI. CONCLUSION

In summary, a systematic study of the Th,  $\text{Th}^+$ , and  $\text{Th}^{2+}$  energies was carried out using the CI plus all-order approach. Excitation energies are compared with experimental [23] and available theoretical results [33]. Good agreement with experiment was found even for neutral Th owing to the all-order treatment of the dominant correlation corrections and sufficient saturation of the configuration space. The differences between the theoretical and experimental energies of neutral thorium did not exceed 2% for 35 out of 78 levels listed in Table I and only 9 energies differed with experiment for more than 5%. These results show the success of the algorithm that we have developed for efficient selection of important configurations for tetravalent systems.

We explored the issue of accidentally vanishing  $g$  factors, where the Landè formula gives  $g = 0$ , such as for  $^3F_1$  terms. We identified a number of cases in which hyperfine  $g$  factors may also vanish.

The recommended values are provided for multipole transition rates and lifetimes of low-lying levels in Ra-like  $\text{Th}^{2+}$ . We expect these values to be accurate to a few percent for strong transitions based on our calculations in divalent alkaline-earth atoms [54]. To further verify the accuracy of our calculations, we compared our results with the only available experimental lifetimes [22] for higher  $5f7p^3G_4$ ,  $7s7p^3P_0$ ,  $7s7p^3P_1$ , and  $6d7p^3F_4$  levels of  $\text{Th}^{2+}$  ion. This work demonstrates the ability to perform accurate calculations for Th and its ions for Th nuclear clock development and other applications.

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