Precision Calculation of Hyperfine Constants for Extracting Nuclear Moments of $^{229}$Th

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Determination of nuclear moments for many nuclei relies on the computation of hyperfine constants, with theoretical uncertainties directly affecting the resulting uncertainties of the nuclear moments. In this work, we improve the precision of such a method by including for the first time an iterative solution of equations for the core triple cluster amplitudes into the relativistic coupled-cluster method, with large-scale complete basis sets. We carried out calculations of the energies and magnetic dipole and electric quadrupole hyperfine structure constants for the low-lying states of $^{229}$Th$^{3+}$ in the framework of such a relativistic coupled-cluster single double triple method. We present a detailed study of various corrections to all calculated properties. Using the theory results and experimental data, we found the nuclear magnetic dipole and electric quadrupole moments to be $\mu = 0.366(6)\mu_N$ and $Q = 3.11(2)\text{ }\mu_b$, respectively, and reduce the uncertainty of the quadrupole moment by a factor of 3. The Bohr-Weisskopf effect of the finite nuclear magnetization is investigated, with bounds placed on the deviation of the magnetization distribution from the uniform one.

Laser spectroscopy in combination with atomic structure calculations can be used to directly determine nuclear moments in a nuclear-theory-independent way. Such an approach is limited by the ability to calculate hyperfine structure (HFS) constants $A$ and $B$ from first principles to high precision. This problem is exacerbated in heavy atoms, where electronic correlation corrections increase. A separate problem is a determination of the accuracy of the theoretical computations, as the theory uncertainty directly contributes to the uncertainty of the extracted nuclear magnetic-dipole and electric-quadrupole moments. While numerical uncertainties can be generally determined, estimating other uncertainties is difficult, because it requires a good understanding of the relative importance of various contributions and method accuracy. In addition, theoretical computations require modeling of magnetization distribution, which is generally not known.

In this work, we consider a solution to these problems for the $^{229}$Th nucleus, motivated by its unique features described below. The development of precision methods for extracting nuclear moments from laser spectroscopy measurements becomes of particular importance now, as more rare isotopes will become available with high yield at the Facility for Rare Isotope Beams [1] for exploring nuclear physics properties, especially of actinides.

As was established more than 40 years ago, the nuclear transition frequency between the ground and first excited states of $^{229}$Th is unusually small and amounts to only several eV [2]. Subsequent measurements of this quantity confirmed it; the current most precise value of 8.19(12) eV [3] is an average of two recent measurements [4,5]. Such a unique feature of this isotope opens up a number of theoretical and experimental research opportunities. Special interest in this nuclear transition is motivated by a possibility to build a superprecise nuclear clock [6] and very high sensitivity to the effects of possible temporal variation of the fundamental constants, including the fine structure constant $\alpha$, strong interaction, and quark mass [7,8].

The present uncertainty in the nuclear transition frequency 0.12 eV, corresponding to $\sim$30 THz, is many orders of magnitude greater than the natural linewidth, expected to be in the millihertz range. To determine the nuclear transition frequency with laser spectroscopic precision, as well as other properties of the ground and isomeric nuclear states, further experimental and theoretical investigations are required [3]. Using the experimentally measured and theoretically calculated HFS constants $A$ and $B$, the nuclear magnetic dipole and electric quadrupole moments were determined in Ref. [9] to be $\mu = 0.360(7)\mu_N$ and $Q = 3.11(6)\mu_b$ (where $\mu_N$ is the nuclear magneton and $e$ is the elementary charge). This value of the magnetic moment contradicts the result $\mu = 0.46(4)\mu_N$ found in Ref. [10] and the recent nuclear calculation value 0.43–0.48$\mu_N$ obtained in
Ref. [11]. Motivated by a necessity to confirm the results of Ref. [9] and by a need to better understand the Th$^{3+}$ properties for the development of the nuclear clock [3], we further developed the coupled-cluster single double triple (CCSDT) method, fully including into consideration both valence and core triple excitations, and applied it for the high-accuracy calculation of the $^{229}$Th$^{3+}$ properties. To the best of our knowledge, due to the exceptional complexity of the problem and very high computational demands, the core triples were never included in the computation of the properties of complicated atomic systems prior to this work.

The simplest version of this approach, the linearized coupled-cluster single double (LCCSD) method, was developed in Ref. [12]. In this version, only the linear terms involving the single ($S$) and double ($D$) excitations of the valence and core electrons were considered. A wide range of properties of univalent systems can be calculated with a very good accuracy using the LCCSD method (see, e.g., [13]). But the systematic prediction of the properties of complicated atomic systems prior to this work. We consider Th$^{3+}$ as a univalent ion with the ground state configuration [Ra]$5f_{5/2}$. The initial Dirac-Hartree-Fock (DHF) self-consistency procedure included the Breit interaction and was done for the core $[1s^2 \ldots 6p^6]$ electrons; the $5f$ and $6d$ orbitals were constructed in the frozen-core potential. The remaining virtual orbitals were formed using 40 basis set $B$-spline orbitals. The basis set included partial waves with the orbital quantum number up to $l = 6$.

The coupled-cluster equations were solved in a basis set consisting of single-particle states. In the equations for singles, doubles, and valence triples, the sums over excited states were carried out with 35 basis orbitals with the orbital quantum number $l \leq 6$. Because of high computational demands to the iterative solution of the equations for the core triple excitations, certain restrictions were applied. We solved these equations allowing the core excitations from the $[4d - 6p]$ core shells, the maximal orbital quantum number of all excited orbitals was equal to 5, and the largest principal quantum number of the virtual orbitals where excitations were allowed was 25. But, as our estimates showed, the omitted electron excitations can contribute to the removal energies of the valence states only at the level of several tens of cm$^{-1}$ and can change the HFS constants at the level of 0.1%. Thus, we took the triple excitations into consideration practically in full.

**Energies.**—Numerical results for the energies are presented in Table I. The lowest-order DHF contribution to the energies (with the inclusion of the Breit interaction) is labeled “BDHF.” At the next step, we carried out the calculation in the linearized coupled-cluster single double (LCCSD) approximation. Then, we subsequently added the NL terms, valence and core triples into consideration, designating these calculations as CCSD, CCSDvT, and CCSDT, respectively. Thus, each subsequent calculation includes all terms taken into account at the previous stage and the additional terms specific for the present approximation.

![FIG. 1. Illustration of single (S), double (D), and triple (T) excitations in the coupled-cluster approach.](253001-2)
TABLE I. The removal energies of the low-lying states for Th⁵⁺ (in cm⁻¹) in different approximations, discussed in the text, are presented. The theoretical total and experimental results are given in the rows \( E_{\text{total}} \) and \( E_{\text{expt}} \), respectively. The difference between the total and experimental values is presented (in percent) in the row labeled “Diff. (%).”

| \( E_{\text{total}} \) | \( E_{\text{expt}} \) |
|----------------------|----------------------|
| \( 5f_{5/2} \)       | \( 5f_{7/2} \)       | \( 6d_{5/2} \)    | \( 6d_{7/2} \)    |
| BDHF                 | CCSD                 | CCSDvT            | CCSDT             |
| 207 310              | 203 393              | 211 842           | 207 686           |
| 232 308              | 227 978              | 222 871           | 217 543           |
| 231 640              | 227 307              | 222 490           | 217 174           |
| 230 819              | 226 538              | 222 472           | 217 259           |
| 230 693              | 226 398              | 222 268           | 217 032           |
| Diff. (%)            | 0.30                 | 0.30              | 0.29              | 0.32              |

\( \Delta \equiv E_{\text{CCSD}} + \Delta E_{\text{extrap}} - E_{\text{expt}} \)

In this way, the most complete calculation is carried out in the CCSDT approximation.

The removal energies of the valence states obtained on each stage are presented in the table. We also found complementary correction due to the basis extrapolation (\( \Delta E_{\text{extrap}} \)), determined as the contribution of the higher \((l > 6)\) partial waves. Based on an empirical rule obtained for Ag-like ions in Ref. [27] and used in Ref. [24], we estimate this contribution as the difference of two calculations carried out with \( l_{\text{max}} = 6 \) and \( l_{\text{max}} = 5 \). We note that this contribution is comparable with that of the triples. The total values, presented in the row labeled “\( E_{\text{total}} \)”, are found as \( E_{\text{CCSDT}} + \Delta E_{\text{extrap}} \).

For the \( 5f \) and \( 6d \) states, the quantum-electrodynamic corrections to the energies are small, and we did not include them in the full-scale calculation. An estimate of this effect, made following Ref. [28], shows that it can potentially change the removal energies of the considered states at the level of 100–200 cm⁻¹.

The experimental removal energy for the ground state is 231 065(200) cm⁻¹ [25]; i.e., its uncertainty is comparable to the difference between our total value and the experimental result. The experimental values for the excited states were taken from Ref. [26].

To illustrate a consistent improvement in the results when we successively add different coupled-cluster terms, we present the differences between the theoretical and experimental values obtained at each stage in the lower panel in Table I. Comparing \( \Delta_{\text{CCSDT}} \) and \( \Delta_{\text{CCSD}} \), we see that the difference between the theory and experiment decreased by almost 4 times for the \( 5f \) states and 2 times for the \( 6d \) states when we included the NL terms and triples.

TABLE II. The theoretical and experimental [26] transition energies (in cm⁻¹) of the excited states counted from the ground state.

| \( f \) | Theory | Experiment | Diff. (cm⁻¹) | Diff. (%) |
|--------|--------|------------|-------------|----------|
| \( 5f_{7/2} \) | 4318 | 4325 | 7 | 0.16 |
| \( 6d_{3/2} \) | 9223 | 9193 | –30 | –0.33 |
| \( 6d_{5/2} \) | 14 475 | 14 486 | 11 | 0.08 |

For completeness, using the total values of the removal energies, we present in Table II the theoretical transition energies counted from the ground state and compare them with the experimental data [26].

**Hyperfine structure constants.**—The magnetic dipole and electric quadrupole HFS constants \( A \) and \( B \) were calculated for the low-lying states of \(^{229}\text{Th}^{5+}\) in Refs. [9,29]. In Ref. [9], the authors also used for the calculation the coupled-cluster method but a significantly less sophisticated version. In this work, we carry out a more complete calculation, including the NL terms and the valence and core triple excitations. In addition, our calculation is purely \textit{ab initio}; no semiempirical methods are applied.

The results for the magnetic-dipole HFS constants \( A_f \equiv A / g \) [where \( g = \mu / \mu_N \) is the \( g \) factor and \( I \) is the nuclear spin, \( I = 5/2 \)] are presented in Table III.

The LCCSD and BDHF values and the difference between them, \( \Delta(\text{SD}) \), are given in the upper panel of the table. Rows 4–6 give the corrections due to the NL terms, \( \Delta(\text{NL}) \), and the valence and core triples, \( \Delta(\nu\text{T}) \) and \( \Delta(\nu\text{C}) \), respectively. The CCSDT values, obtained as the sum of the LCCSD values and the NL, \( \nu\text{T} \), and \( \nu\text{C} \) contributions, are more accurate.

TABLE III. Different contributions to \( A_f \) (in megahertz) and obtaining the recommended value of \( g \) are explained in the text. The experimental values of the HFS constants \( A_f \) [30] are given in the row labeled “\( A \) (exper.)”. The uncertainties are given in parentheses.

| \( f \) | \( 5f_{5/2} \) | \( 5f_{7/2} \) | \( 6d_{3/2} \) | \( 6d_{5/2} \) |
|--------|-------------|-------------|-------------|-------------|
| BDHF   | 507         | 263         | 831         | 304         |
| \( \Delta(\text{SD}) \) | 72 | –45 | 268 | –386 |
| LCCSD  | 579         | 218         | 1099        | –81         |
| \( \Delta(\text{NL}) \) | –3.3 | –4.6 | –17 | 18 |
| \( \Delta(\nu\text{T}) \) | –12 | –5.1 | –21 | –46 |
| \( \Delta(\nu\text{C}) \) | –1.5 | –2.3 | 5.8 | –1.8 |
| Total  | 562(3)      | 209(3)      | 1063(12)    | –104(22)    |
| Ref. [9] | 573 | 215 | 1079 | –92 |

\( A \) (exper.) [30] 82.2(6) 31.4(7) 155.3(1.2) –12.6(7)

The values, listed in Ref. [9], are multiplied by \( I = 5/2 \).
corrections, are presented in the row labeled “CCSDT.” The basis extrapolation corrections are given in the row labeled “Basis extrapol.” The total values, listed in the row “Total,” are found as the sum of the CCSDT value and the basis extrapolation correction.

Based on a comparison of the theoretical and experimental HFS constants for a number of univalent elements, the authors of Ref. [9] suggested a method to estimate the uncertainties of these constants. The uncertainty of the A and B calculations is expected to be on the order of 3%–6% of the total correlation correction (found as the difference between the final and LCCSD value), if this correction does not exceed 50%. Following this approach, we estimate the uncertainties of A, for the 5f_{5/2}, 5f_{7/2}, and 6d_{3/2} states as 5% of their total correlation corrections. For the 6d_{5/2} state, the correlation correction is very large, and the validity of this method is questionable. Applying it, we roughly estimate the uncertainty of A, for this state at the level of 20%–25%.

We note that all values presented in Table III were obtained for the nucleus considered as the charged ball with uniform magnetization. But, as was shown in Ref. [31], the nucleus of ^{239}Th has a complex structure and different collective effects, such as quadruple-octupole vibration-rotation motion of the nucleus, the single-particle motion of the unpaired nucleon, and the Coriolis interaction between this nucleon and the nuclear core, are important. Thus, the real nuclear magnetization can differ from the uniform magnetization. To investigate this problem, we follow the approach developed in Ref. [32]. We can express the HFS constant A as

\[ A = g A_0 (1 - d_{\text{nuc}} y), \]  

where A_0 is the theoretical value calculated at the pointlike magnetization of the nucleus and d_{\text{nuc}} and y are the parameters depending on the nuclear and electronic structure, respectively. The quantities g and d_{\text{nuc}} are assumed to be unknown. The experimental value of A can be used on the left-hand side of Eq. (1), and y can be found from the calculation.

Indeed, taking into account that d_{\text{nuc}} = 0 and d_{\text{nuc}} = 1 correspond to the pointlike and uniform magnetization, respectively, we can find y from Eq. (1) as

\[ y = 1 - A_I / A_0, \]  

where A_I are given in Table III for different states. We note that the ratios A_I / A_0 are very stable and insensitive to different corrections, and we determine the uncertainty of y at the level of 0.02%.

To find g and d_{\text{nuc}}, we use the HFS constants for the 5f_{5/2} and 6d_{3/2} states, which are known most precisely both experimentally and theoretically. Using for each of them Eq. (1) and solving the system of two equations in two unknowns, we arrive at

\[ g \approx 0.1465(24), \]  

\[ d_{\text{nuc}} \approx 1.7(2.1). \]  

Thus, the g factor is determined with the accuracy \( \sim 1.5\%. \) Using this value, we find the nuclear magnetic moment: \( \mu_I = g I \approx 0.366(6) \). This result is in good agreement with that reported in Ref. [9]: \( \mu_I \approx 0.360(7) \). We note that the uncertainty estimate in Ref. [9] did not include uncertainty due to the magnetization distribution.

For heavy nuclei, the parameter d_{\text{nuc}} can vary over a wide range. For example, for the gold isotopes ^{197,199,191}Au with the nuclear spin of 3/2, d_{\text{nuc}} = \pm 5.5(6) \) [32]. Our result (4) suggests that for ^{239}Th the absolute value of d_{\text{nuc}} is smaller and it is most likely positive. As a result, the correction to the g factor, due to inhomogeneity of the nuclear magnetization, is small but not negligible.

In Table IV, we present the results obtained for the electric-quadrupole HFS constants B/Q. All designations in the upper panel of the table are the same as in Table III. In the lower panel, we present the experimental results for the HFS constants B [30]. The values of B (in e\( b \)) are found as the ratios of B (experim.) and values listed in the row labeled “Total.”

The values of B/Q obtained in this work turned out to be between the results of Refs. [9,29] but are somewhat closer to the former. The experimental uncertainty for

| \( 5f_{5/2} \) | \( 5f_{7/2} \) | \( 6d_{3/2} \) | \( 6d_{5/2} \) |
|---|---|---|---|
| BDHF | 535 | 572 | 611 | 648 |
| \( \Delta(\text{SD}) \) | 202 | 251 | 132 | 228 |
| LCCSD | 737 | 822 | 743 | 877 |
| \( \Delta(\text{NL}) \) | 38 | 45 | 9 | 9 |
| \( \Delta(\text{VT}) \) | \( -55 \) | \( -57 \) | \( -34 \) | \( -27 \) |
| \( \Delta(\text{cT}) \) | 3 | 3 | 7 | 7 |
| CCSDT | 723 | 814 | 725 | 866 |
| Basis extrapol. | 6 | 8 | 3 | 4 |

| Total | 729(10) | 822(13) | 728(6) | 869(11) |
| Ref. [9] | 725 | 809 | 738 | 873 |
| Ref. [29] | 740 | 860 | 690 | 860 |
| \( B (\text{experim.}) [30] \) | 2269(2) | 2550(12) | 2265(9) | 2694(7) |
| \( Q \) | 3.11(4) | 3.10(5) | 3.11(3) | 3.10(4) |
| \( Q(\text{recommended}) \) | 3.11(2) |
| Ref. [9] | 3.11(6) |
| Refs. [29,30] | 3.11(16)\(^{a}\) |

\(^{a}\)This result was obtained using the measurements of Ref. [30] and calculations of Ref. [29].
the constants $B$ does not exceed 0.5%, while the theoretical uncertainties are at the level of 0.8%–1.5%. The uncertainties were determined in the same manner as was done for the HFS constants $A$. The constants $B$ are large for all four considered states, and their fractional uncertainties are comparable. For this reason, the recommended value was obtained as the weighted average over four values of $Q$ given in Table IV. We note the perfect agreement of our recommended value with the results obtained in Refs. [9,29,30], but our uncertainty is a few times smaller.

**Conclusion.**—We have developed the relativistic CCSDT method with the full inclusion of valence and core linear triple excitations. Using the theoretical values of the HFS constants obtained in this work and the experimental results [9], we determined the values of the nuclear magnetic dipole and electric quadrupole moments. Analyzing the results obtained at the different stages, we determined the uncertainties of the recommended values to be 1.5% for $\mu_I$ and 0.6% for $Q$. We investigated the effect of the inhomogeneity of the nuclear magnetization and found it to be small but not negligible. We conclude that it should be taken into account to determine the magnetic dipole nuclear moment $\mu_I$ with accuracy better than 1%. Further experimental work to measure the HFS (especially magnetic dipole) constants more precisely is needed to improve precision and better understand the effect of nuclear magnetization. The method developed in this work can be used for any monovalent system and, when combined with configuration interaction, can be extended to systems with 2–6 valence electrons.

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