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Magnetic dipole and electric quadrupole moments of the ^{229}Th nucleus

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We determine the magnetic dipole $\mu = 0.360(7)\mu_N$ and the electric quadrupole $Q = 3.11(6)e$b moments of the ^{229}Th nucleus by combining our high-precision calculations of the hyperfine constants with measurements reported in Campbell *et al.* [Phys. Rev. Lett. **106**, 223001 (2011)]. We find that the previous value $\mu = 0.46(4)\mu_N$ [Gerstenkorn *et al.*, J. Phys. (Paris) **35**, 483 (1974)] is incorrect by 25%. We report a method for determining the accuracy of theoretical hyperfine constants B/Q and demonstrate that it can be used to extract the electric quadrupole moment Q with a 1%–2% uncertainty for a large number of nuclei. This approach allowed us to identify 40% inconsistencies in measurements of Ra^+ hyperfine constants B.

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The electromagnetic nuclear moments are fundamental quantities that play an important role in many atomic, nuclear, and solid state processes. The determination of nuclear magnetic dipole and electric quadrupole moments is important to deepen our understanding of the nucleon-nucleon interaction [1–8]. Moreover, recent studies have shown that nuclear quadrupole couplings affect decoherence and relaxation of central spins in quantum dots [9] and can be used as a microscopic probe to study the motion of atomic tunneling systems in amorphous solids [10]. While nuclear magnetic moments are known well for many nuclei, many quadrupole moments are known poorly [11], due to the lack of either direct measurements or accurate theoretical values of hyperfine constants B/Q for atomic spectroscopy methods.

The ^{229}Th nucleus has an unusually low first excitation energy of only several eV [12,13], making the corresponding nuclear transition accessible with laser excitation. The transition is expected to be very narrow and well isolated from the effects of external fields and, therefore, presents a remarkable opportunity for the development of a nuclear clock [14–16]. It is essential to possess accurate values for both nuclear and electronic properties of $^{229}Th^{3+}$ for the implementation of the electronic bridge process for the ion-trap nuclear clock [14]. Moreover, the transition frequency is expected to be as many as five orders of magnitude more sensitive to temporal variation of the fine-structure constant α and the dimensionless strong interaction parameter $m_q/\Lambda_{\rm QCD}$ as compared to atomic transitions, making ^{229}Th one of the most attractive candidates for such studies [17]. Direct laboratory measurements of the change in the nuclear electric quadrupole moment between the isomer and the ground-state nucleus can be used to extract a variation of the fine-structure constant that requires accurate knowledge of Q for the ground-state nucleus [18]. Th^{3+} has been also proposed to study parity violation [19], and the hyperfine constants are the best parity-conserving benchmarks required for such studies.

In this Rapid Communication, we report (1) a development of a method to determine the accuracy of theoretical hyperfine constants B/Q that allows one to extract nuclear quadrupole moments for a number of nuclei with 1%–2% uncertainties, and (2) a determination of the magnetic dipole and electric quadrupole moments of the ^{229}Th nucleus with 2% uncertainties. We find a 25% discrepancy with a previous determination of μ [20]; our value of Q is in agreement with a previous determination [21] but is 2.7 times more precise. Hyperfine constants A and B are predicted for a number of states. We discuss the prospects for further accuracy improvements of the ^{229}Th nuclear moment and a possible extraction of the $^{229}Th^{3+}$ nuclear magnetic octupole moment. This work is relevant to all of the applications of ^{229}Th discussed above.

We also calculated Ba^+ and Ra^+ hyperfine constants as a part of our theoretical uncertainty study. We found a 40% discrepancy in the values of Q obtained from measurements of B in different isotopes [22,23]. Our results also imply a 2σ discrepancy in the measured values of Ra^+ $7s$ and $6d_{3/2}$ hyperfine constants A. Ra^+ is a subject of a present parity violation study in KVI [24] and the accurate determination of its hyperfine constants is a first step toward testing of the standard model of the electroweak interactions.

The electronic level structure of the Fr-like Th^{3+} ion, with a single valence electron above the closed [Rn]=[Xe]$4f^{14}5d^{10}6s^26p^6$ core, is conducive to high-accuracy atomic calculations [18,25]. Laser-cooled Wigner crystals of $^{229}Th^{3+}$ allow for high-precision spectroscopy [21,26]. The atomic hyperfine constants A are proportional to the nuclear magnetic dipole moment μ. Therefore, we calculate $(A/\mu)^{\rm th}$ and determine μ as the ratio of the experimental value for the hyperfine constants $A^{\rm expt}$ and $(A/\mu)^{\rm th}$. Similarly, the nuclear electric quadrupole moment Q is determined as the ratio of the experimental value for the hyperfine constants $B^{\rm expt}$ and the computed quantity $(B/Q)^{\rm th}$. Therefore, the accuracy of the resulting μ and Q is limited by the estimated accuracy of the theoretical calculations. Since the hyperfine constants are very difficult to calculate accurately for heavier systems, it is even more difficult to estimate the accuracy of the resulting values. As a consequence, Q's are generally known quite poorly, to about 10% [11]. In this Rapid Communication, we resolve this problem by developing a

broadly applicable method to determine the uncertainty of the theoretical hyperfine constants calculated with the all-order method. Since the nuclear magnetic moments μ are known well for many nuclei from independent measurements, the comparison of the experimental and theoretical hyperfine constants calculated by the same all-order approach for a large number of systems and states allowed us to identify a stable and reliable pattern of how the uncertainty depends on the size of the correlations.

We start with a description of the general method to determine the uncertainties of A and B. Other secondary tests of the uncertainties will be discussed later on the example of Th^{3+}. We compare our hyperfine constant A with experiment for a large number of monovalent systems where nuclear magnetic moments are well known [27–30,30–32]. All calculations were carried out using a relativistic linearized coupled-cluster (all-order) method including single, double, and partial triple (SDpT) excitations [25,33,34]. A brief description of the method is given in the Supplemental Material [35]. The Fermi distribution is used for both change and magnetization distributions. The resulting values for Ca^+, Sr^+, Hg^+, Rb, Cs, Ba^+, Fr, and Ra^+ are compared with experimental values in Table I of the Supplemental Material [35]. The nuclear magnetic moments are taken from Ref. [11] unless noted otherwise. The results for the last four systems are given in Table I. Ba^+ and Ra^+ hyperfine constants are calculated in the present work. The lowest-order values are listed in the "DF" column of Table I. The relative correlation correction, evaluated as the difference of the *ab initio* final and the lowest-order values, is listed in column "CC." Our Fr result for the $A(9s)$ state has been used in Ref. [31] to extract the value of the ^{210}Fr nuclear magnetic dipole moment with 1% uncertainty.

We established the clear correlation between the size of the correlation corrections and the accuracy of the values by comparing the theory and experimental values for Ca^+, Sr^+, Hg^+, Rb, Cs, Ba^+, Fr, and Ra^+. All cases similar to the $5f$ and $6d$ states of Th^{3+} where the correlation correction is below 30% agree with experiment to better than 2%. We note that Hg^+ is not a true "monovalent" case owing to low-lying core excitations that are not present in Th^{3+}, so the agreement is slightly worse (−2.3%). We find that the average ratio of the last two columns of Table I of Ref. [35] (that give a correlation correction and difference with experiment) is 2.9%, and the maximum ratio is 6% (we excluded Hg^+ and the anomalous case of the $4d_{3/2}$ state in Rb, where the correlation is 65% for the average). If we average over only heavy systems listed in Table I (Cs, Ba^+, Fr, Ra^+), the average ratio is still 3.1%. Therefore, we can make a general conclusion that the uncertainty of the A and B calculations is expected to be on the order of 3% of the total correlation correction and should not exceed 6%. As long as the correlation does not exceed 50%, this finding does not depend on the size of the correlations, atomic system, or the electronic state. While the above analysis is done for A constants, it holds for a variety of states with completely different distributions of the correlation correction terms. Therefore, it is expected to be applicable to B constants as well.

Next, we discuss the determination of the ^{229}Th nuclear magnetic moments and additional methods to determine the uncertainties. The determination of the ^{229}Th ($I = 5/2$) nuclear magnetic dipole moment is illustrated in Table II. The values of μ (in μ_N) are obtained as the ratio of the experimental A values and theoretical A/μ results for all four states. *Ab initio* theory values are listed for the $5f_{5/2}$ and $5f_{7/2}$ states. The values for the $6d_{3/2}$ and $6d_{5/2}$ states are obtained by scaling *ab initio* results by −0.75% and −13.7%, respectively, using a comparison of theoretical $5d$ Ba^+ values with experimental data. The scaling factors were calculated as the difference of the value for the corresponding $5d$ Ba^+ constant with experiment multiplied by the ratio of the correlation corrections for the $6d_j$ state of Th^{3+} and the $5d_j$ state of Ba^+. The value determined from the $6d_{3/2}$

TABLE I. Comparison of the hyperfine constants A (in MHz) with experiment in ^{133}Cs [36,37], $\mu = 2.582025(3)\mu_N$, $^{137}Ba^+$ [38–40], $\mu = 0.937365\mu_N$, ^{210}Fr [41,42], $\mu = 4.38(5)\mu_N$, $^{211}Ra^+$, $\mu = 0.878(4)\mu_N$, and $^{213}Ra^+$ [22,43,44], $\mu = 0.613(2)\mu_N$. Theory values for Cs and Fr are from Ref. [31]. Ba^+ and Ra^+ values are calculated in the present work. Lowest-order DF values are given in column "DF" and relative correlation corrections are given in column "CC" in percent.

System	Level	DF	Final	Expt.	CC (%)	Diff. (%)
^{133}Cs	$6s$	1424	2276	2298	37	1.0
	$7s$	391	540	546	28	1.1
	$8s$	163	216.8	219.3(1)	25	1.1
$^{137}Ba^+$	$6s$	2913	3998	4019	27	0.5
	$6p_{1/2}$	491	734.0	743.8	33	1.3
	$6p_{3/2}$	71.2	121.3	127.3	41	4.7
	$5d_{3/2}$	128	191.5	189.7	33	−1.0
	$5d_{5/2}$	51.6	−10.0	−12.0	616	17
^{210}Fr	$7s$	4740	7244	7195(1)	35	−0.7
	$8s$	1214	1577	1578(2)	23	0.1
$^{211}Ra^+$	$7s$	5099	6728	6625(1)	24	−1.6
	$7p_{1/2}$	860	1299.1	1299.7(8)	34	0.0
$^{213}Ra^+$	$7s$	17798	23488	22920(6)	24	−2.5
	$7p_{1/2}$	3001	4535	4542(7)	34	0.2
	$7p_{3/2}$	230	384[a]	384(5)[b]	38	0
	$6d_{3/2}$	360	538[a]	528(6)	34	−1.9

[a]Recommended value based on Ba^+ data.
[b]Rescaled from the measurement in ^{223}Ra.

TABLE II. Determination of the ^{229}Th nuclear magnetic dipole moment. The values of μ (in μ_N) are obtained as the ratios of the values in the expt. [21] and A/μ theory columns. Theoretical values of A (in MHz) with $\mu = 0.360\mu_N$ are listed in column "A, theory." Relative differences of the experimental hyperfine constants and theoretical values with $\mu = 0.360\mu_N$ are given in column "Diff." in percent.

State	A expt.	Unc. expt.	A/μ theory	μ	A theory	Diff.
$5f_{5/2}$	82.2(6)	0.7%	229.2	0.359	82.5	−0.4%
$5f_{7/2}$	31.4(7)	2.2%	86.1	0.365	31.0	1.3%
$6d_{3/2}$	155.3(12)	0.8%	431.5	0.360	155.3	0
$6d_{5/2}$	−12.6(7)	5.6%	−36.7	0.343	−13.2	−4.8
Final				0.360(7)		

TABLE III. Determination of the ^{229}Th nuclear electric quadrupole moment from theory values of Ref. [18] and present results. The "Present work" B/Q column contains final *ab initio* theory results in MHz/eb. Theoretical values of B with $Q = 3.11e$b are listed in columns "B." The relative difference of the experimental hyperfine constants [21] and theoretical values with $Q = 3.11e$b are given in column "Diff." in percent.

		Reference [18]				Present work			
State	Expt. [21] B	B/Q	Q	B	Diff.	B/Q	Q	B	Diff.
$5f_{5/2}$	2269(6)	740	3.07	2300	−1.4%	725	3.13	2254	0.7%
$5f_{7/2}$	2550(12)	860	2.97	2680	−4.9%	809	3.15	2515	1.4%
$6d_{3/2}$	2265(9)	690	3.28	2150	5.3%	738	3.07	2295	−1.3%
$6d_{5/2}$	2694(7)	860	3.13	2680	0.7%	873	3.09	2716	−0.8%
Final			3.11(16)				3.11(6)		

hyperfine constant is taken as the final result because of the Ba^+ benchmark comparison that allowed for additional improvement of the value. We note that the value extracted from the $5f_{5/2}$ state differs by only 0.38% from the final value. The $5f_{5/2}$ *ab initio* result is expected to be the most accurate since it has the smallest correlation correction, only 11%, while the correlation correction for the $6d_{3/2}$ state is 24%. To illustrate the consistency of the extracted μ values with all four experimental results, we multiply the theory values for A/μ by our final value of $\mu = 0.360\mu_N$ and compare them with experimental data ($6d_{3/2}$ will be in exact agrement since it was used to determine the final value of μ). The relative differences of theory and experiment are listed in the last column "Diff." in percent. We find that all four of the theory values are in agreement with experiment within experimental uncertainties listed in percent in column "Unc." for convenience. Our final value of $\mu = 0.360(7)\mu_N$ differs by 25% from the previous result $\mu = 0.46(4)\mu_N$ [20] that was also derived from the hyperfine constant measurements.

The determination of the ^{229}Th nuclear electric quadrupole moment is illustrated in Table III. The table structure is similar to that of Table II, but also includes a prior determination of Q carried out in Ref. [21] using theory data from Ref. [18] for comparison. Columns 3–6 show the determination of Q using theory values from Ref. [18], and columns 7–10 show the determination of Q using our present *ab initio* theory results. We take the average of the results obtained from four states as the final Q value since the experimental and theoretical accuracy is similar for all four states and no relevant benchmarks exist for any of the four states. Remarkably, the average value of Q determined using our data and Ref. [18] is identical to three significant figures, despite 2%–6% differences in the values of B/Q. However, the differences of the Q values determined from all four $5f_j$ and $6d_j$ states and the final value are lower in the present work, 0.7%–1.3% (compare the two "Diff." columns). Our final value, $Q = 3.11(6)e$b (1 b = 10^{-28} m^2), is significantly lower than the previous value of $4.3(9)e$b inferred from hyperfine measurements [20], but is in agreement with the result deduced from the Coulomb excitation of the nucleus, $Q = 3.15(3)e$b [45].

To evaluate the uncertainties in the values of μ and Q, we first apply the method described above, which gives an average uncertainty of 3% of the correlation correction and a maximum uncertainty to be 6% of the correlation correction.

(I) We list $^{229}Th^{3+}$ hyperfine constants A and B in Table IV in MHz; our final values, $\mu = 0.360\mu_N$ and $Q = 3.11e$b, are used. The relative size of the correlation corrections defined as the ratio of *ab initio* (SDpT-DF)/SDpT, where DF are the lowest-order results, is listed in columns "CC" in %. We find that the correlation corrections to the $6d$ and $5f$ A and B values are relatively small, 11%–29%, with the only exception of $A(6d_{5/2})$. The uncertainty of both theory and experiment is about 5% for the $6d_{5/2}$ state, so it was not used in the determination of μ. For the $6d_{3/2}$ and $5f_j$ A constants, 3%/6% of the correlation gives a (0.3%–0.7%)/(0.6%–1.4%) uncertainty, depending on the state, with $5f_{5/2}$ being the most accurate. For all $6d_j$ and $5f_j$ B constants, 3%/6% of the correlation gives a (0.5%–0.9%)/(1%–1.8%) uncertainty, depending on the state, with $6d_{3/2}$ being the most accurate. This analysis gives a 1.5% upper bound on the theory uncertainty for μ and 2% for Q. The experimental uncertainty is 0.7% for $A(6d_{3/2})$ and $A(5f_{5/2})$, giving a 2% combined upper bound on the uncertainty of μ. The experimental uncertainty for all B constants is 0.26%–0.47%, which can be considered negligible in comparison with the theory uncertainty.

(II) Next, we carry out an additional analysis of the uncertainties by using a specific benchmark system. We find that Ba^+ and Ra^+ represent the best benchmark testing cases for Th^{3+} owing to a very similar level structure and size of the correlation correction, and the magnetic moment of ^{137}Ba is known with less than 0.01% uncertainty [11]. To ascertain that these cases are indeed very similar, we compare the entire breakdown of the 20 correlation correction terms and normalization contributions to the hyperfine constants. The comparative breakdown of correlation terms for various

TABLE IV. Hyperfine constants A and B in $^{229}Th^{3+}$ ($I = 5/2$, $\mu = 0.360\mu_N$, $Q = 3.11e$b) in MHz. Relative correlation corrections are listed in columns "CC" in percent.

Level	A	CC	B	CC	Level	A	CC	B	CC
$5f_{5/2}$	82.5	11%	2254	26%	$7s$	5806	17%		
$5f_{7/2}$	31.0	−23%	2515	29%	$7p_{1/2}$	1419	23%		
$6d_{3/2}$	155.3[a]	24%	2295	17%	$8s$	2114	15%		
$6d_{5/2}$	−13.2[a]	485%	2716	26%	$8p_{1/2}$	582	19%		
$7d_{3/2}$	43.0	28%	824	40%	$7p_{3/2}$	119.4[a]	31%	5310	30%
$7d_{5/2}$	8.59	−36%	977	45%	$8p_{3/2}$	51.3	30%	2238	26%

[a]These three values are obtained by scaling *ab initio* 156.5, −11.4, and 115.4 MHz results using Ba^+ data.

states of Ba^+, Ra^+, and Th^{3+} is given in Table II of the Supplemental Material [35]. We find that the Ba^+ and Ra^+ $nd_{3/2}$ cases are nearly identical, while the Th^{3+} $6d_{3/2}$ case is very similar, but has a smaller overall correlation correction, 24%, instead of 33%–34%. This is expected as the correlation effects decrease with increasing degree of ionization. As we noted above, we use a -1% difference of our Ba^+ value with experiment and a 24/34 correlation correction ratio to adjust our $6d_{3/2}$ value by -0.75%. As a result, we expect that our $6d_{3/2}$ theory value is accurate to better than 1%. However, the difference for the similarly adjusted $6d_{3/2}$ hyperfine constant of $^{213}Ra^+$ with experiment is -1.9%. The Ra^+ measurement uncertainty is 1.1% for this state, and the uncertainty of the $^{213}Ra^+$ magnetic moment (from a direct measurement) is 0.3%. We note that there is a 1% inconsistency in the values of measured $7s$ hyperfine constants in $^{211}Ra^+$ and $^{213}Ra^+$ isotopes and our corrected (using Ba^+ data) value of the $7p_{3/2}$ constant is in perfect agreement with the experiment. Therefore, it is likely that the discrepancy is due to uncertainty in the Ra^+ measurement. Nevertheless, we keep our original 2% estimated uncertainty of μ. We also find that the measurement of the $B(6d_{3/2})$ in ^{211}Ra [23] is inconsistent with the earlier measurement of the $B(7p_{3/2})$ in ^{223}Ra [22] by 40%. Using our theoretical values of B/Q yields $Q(^{211}\mathrm{Ra}) = 0.34(2)e$b while the rescaled value for this isotope listed in Ref. [22] is $0.48(2)e$b. Using a more precise measurement of $B(7p_{3/2})$ in $^{223}Ra^+$ and our theoretical calculation of B/Q, we obtain $Q(^{223}\mathrm{Ra}) = 1.24(3)e$b.

(III) Finally, we use the consistency of μ and Q obtained from different states as an independent uncertainty estimate. Table II of the Supplemental Material [35] illustrates that the contributions of the various correlation terms are quite different for the $6d_{3/2}$, $5f_{5/2}$, and $5f_{7/2}$ states. In fact, the correlation correction is negative for the $5f_{7/2}$ level while it is positive for the other two levels. Therefore, these calculations are sufficiently different that the spread of the μ values obtained from three levels (0.8%–1.3%) provides another independent estimate of the accuracy. The 0.7%–1.4% difference of the Q values obtained for four levels with the average final value confirms a 2% uncertainty estimate for Q.

Improved measurements of the $6d_{3/2}$ and $5f_j$ hyperfine intervals are critical for further improvements of the nuclear moment values. Using sub-kHz linewidth lasers and an ultracold sample sympathetically cooled with, e.g. $^{232}Th^{3+}$, the ∼100-kHz-wide optical transitions of interest could all be measured with inaccuracies of <10 kHz. In extracting the hyperfine intervals, this would reduce the current uncertainties in the A constants from $\sim 10^{-2}$ to $\sim 10^{-5}$, a level well below that of the companion calculations. Such measurements would also reduce the current uncertainties in the measured B constants by three orders of magnitude.

The structure of the $^{229}Th^{3+}$ nucleus may also be studied beyond the first two electromagnetic moments. The $5f_{5/2}$ electronic ground level is well suited for such studies, as it has a large total angular momentum. Because J is greater than 1, the valence electron has nonzero coupling to the nuclear magnetic octupole moment Ω. This effect, parametrized by the magnetic octupole hyperfine constant C, is expected to shift hyperfine intervals at a level of 10^1–10^2 Hz. Observing such an effect is straightforward in this ground-state hyperfine manifold, using microwave spectroscopy, as the individual states are effectively infinitely narrow and each hyperfine level contains a $m_F = 0$ clock state, allowing for the removal of first-order Zeeman shifts from the measurements. All hyperfine intervals should be measurable with inaccuracies of ∼0.01 Hz. This level of error is $<1\%$ of an expected minimum value for C, indicating that percent-level inaccuracies in the atomic structure calculations and nuclear magnetic dipole and electric quadrupole moment values would lead to an extraction of Ω with an uncertainty of at most a few percent, on par with the most accurately determined nuclear octupole moment to date [46].

In conclusion, we determined the nuclear magnetic dipole and electric quadrupole moments of ^{229}Th with 2% uncertainty by combining high-precision theoretical and experimental values of the hyperfine constants. The present work also presents a systematic method for the determination of electric quadrupole nuclear moments in a large number of nuclei with 1%–2% uncertainty by establishing that the uncertainty of the all-order hyperfine constants is 3%–6% of the correlation correction. We explored additional methods to determine the uncertainties in the Th^{3+} nuclear magnetic moments by using the reference benchmark systems and consistency checks between results obtained using different states.

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