


Sensitivity of ^{229}Th nuclear clock transition to variation of the fine-structure constantPavel Fadeev,^{1,*} Julian C. Berengut², and Victor V. Flambaum^{1,2}¹*Helmholtz Institute Mainz, Johannes Gutenberg University, 55099 Mainz, Germany*²*School of Physics, University of New South Wales, Sydney, New South Wales 2052, Australia* (Received 1 July 2020; revised 2 November 2020; accepted 3 November 2020; published 30 November 2020; corrected 25 January 2021)

Peik and Tamm [*Europhys. Lett.* **61**, 181 (2003)] proposed a nuclear clock based on the isomeric transition between the ground state and the first excited state of thorium-229. This transition was recognized as a potentially sensitive probe of possible temporal variation of the fine-structure constant, α . The sensitivity to such a variation can be determined from measurements of the mean-square charge radius and quadrupole moment of the different isomers. However, current measurements of the quadrupole moment are yet to achieve an accuracy high enough to resolve nonzero sensitivity. Here we determine this sensitivity using existing measurements of the change in the mean-square charge radius, coupled with the ansatz of constant nuclear density. The enhancement factor for α variation is $K = -(0.82 \pm 0.25) \times 10^4$. For the current experimental limit, $\delta\alpha/\alpha \lesssim 10^{-17}$ per year, the corresponding frequency shift is ~ 200 Hz per year. This shift is six orders of magnitude larger than the projected accuracy of the nuclear clock, paving the way for increased accuracy of the determination of $\delta\alpha$ and interaction strength with low-mass scalar dark matter. We verify that the constant-nuclear-density ansatz is supported by nuclear theory and propose how to verify it experimentally. We also consider a possible effect of the octupole deformation on the sensitivity to α variation and calculate the effects of α variation in a number of Mössbauer transitions.

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I. INTRODUCTION

The first excited isomeric state of thorium-229, ^{229m}Th , is a candidate for the first nuclear optical clock [1]. This is due to the state's low excitation energy of several electronvolts [2–5] (the lowest of all known isomeric states) and long radiative lifetime of up to 10^4 s [6,7]. Several theoretical and experimental groups are making rapid progress toward using ^{229m}Th as a reference for a clock with unprecedented accuracy [8–16]. These papers proposed specific experimental schemes for nuclear clocks and performed detailed studies of systematic effects such as blackbody radiation shifts, effects of ion trapping fields in ion traps, and effects of stray fields. The advantage of the nuclear clock in comparison with atomic clocks is that, due to the very small size of the nucleus and its shielding by atomic electrons, it is insensitive to many systematic effects. For example, the nuclear polarizability and its contribution to the major systematic effect, the blackbody radiation shift, are 15 orders of magnitude smaller than in atomic transitions. Nuclear clocks may perform at a level of accuracy of 10^{-19} [9], 1–2 orders of magnitude higher than the accuracy of the best existing atomic clocks.

In a recent crucial step towards this goal, the transition was measured using spectroscopy of the internal conversion electrons emitted in flight during the decay of neutral ^{229m}Th atoms [17], yielding an excitation energy $E_{\text{is}} = 8.28(17)$ eV. Another approach, using γ -ray spectroscopy at 29.2 keV, obtained $E_{\text{is}} = 8.30(92)$ eV [18,19]. More recently, $E_{\text{is}} = 8.10(17)$ eV was reported [20].

The ^{229m}Th nuclear clock is expected to be a sensitive probe for time variation of the fundamental constants of nature [21]. To avoid dependence on units we consider the effect of variation of the dimensionless fine-structure constant, α , related to the electromagnetic interaction [21–26]. Another dimensionless parameter, m_q/Λ_{QCD} , where m_q is the quark mass and Λ_{QCD} is the QCD scale, is related to the strong interaction. The effect of m_q/Λ_{QCD} variation on the ^{229}Th transition has been estimated in Refs. [21,27,28]. The high sensitivity to α comes about because the change in Coulomb energy between the isomers, which depends linearly on α , is almost entirely canceled by the nuclear force contribution which has only weak α dependence. This cancellation makes the energy of the transition $E_{\text{is}} = 8$ eV low compared to typical nuclear transitions, so any change in α and the Coulomb energy leads to a relative change several orders of magnitude larger in the energy of the transition $\Delta E_{\text{is}}/E_{\text{is}}$.

We also should note that the measurement of the variation of the fundamental constants does not require absolute frequency measurement. All that is required is high stability of the ratio of two frequencies with a different dependence on the fundamental constants [29,30]. For example, it may be the ratio of the 8-eV nuclear transition frequency to that of an atomic clock transition in the Th ion, as considered in Ref. [31].

The change in the nuclear transition frequency, f , between the isomeric state and the ground state, δf , for a given change in the fine-structure constant, $\delta\alpha$, is [21]

$$h \delta f = \Delta E_C \frac{\delta\alpha}{\alpha}, \quad (1)$$

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where ΔE_C is the difference in Coulomb energy between the two isomers. The enhancement factor K is defined by

$$\frac{\delta f}{f} = K \frac{\delta \alpha}{\alpha}, \quad (2)$$

where $K = \Delta E_C / E_{is}$. Therefore, to find the sensitivity of ^{229m}Th transition to variation in α , one needs to know ΔE_C .

The Coulomb energy E_C depends on the shape of the nucleus. Unlike atomic systems, which are spherical due to the $1/r$ potential from the pointlike nucleus (r is the distance from the nucleus), nuclear systems can have deformed shapes as the potential originates from the nucleons themselves. Reference [25] showed that, by modeling the nucleus as a prolate spheroid [32], ΔE_C can be deduced from measurements of the change in nuclear charge radius and quadrupole moment between the isomeric and the ground states. Using this model with measurements of nuclear parameters, the authors of [33] give a value of

$$\Delta E_C = -0.29(43) \text{ MeV}, \quad (3)$$

where the dominant source of error is the uncertainty in measured quadrupole moments of the ground and the excited states. Such a ΔE_C is consistent with a K value anywhere between 0 and 10^5 . This can be compared to a K of about 0.1–6 for current atomic clocks [29,30,34–37].

In this paper we use the fact that the change in quadrupole moment is related to the change in charge radius to arrive at ΔE_C with errors consistent with a nonzero value, consequently giving a nonzero value for K . This relationship can be understood from the assumption of a constant charge density between isomers. We verify that this assumption gives a relation that is consistent with previous results from nuclear theory [23]. We also test this assumption in several Mössbauer transitions, which we find have much lower sensitivities to α variation than the ^{229}Th transition. Finally, following models that suggest the existence of an octupole deformation in ^{229}Th , we use a more general treatment of a deformed nuclei. The results of the two models coincide within uncertainties.

II. PROLATE SPHEROID MODEL

We start by modeling the nucleus as a prolate spheroid with semiminor and semimajor axes a and c . The volume $(4\pi/3)R_0^3$ depends on a and c by

$$a^2 c = R_0^3. \quad (4)$$

The eccentricity e is defined by

$$e^2 = 1 - \frac{a^2}{c^2}, \quad (5)$$

while the mean-square radius $\langle r^2 \rangle$ and the quadrupole moment Q_0 are

$$\begin{aligned} \langle r^2 \rangle &= \frac{1}{5}(2a^2 + c^2), \\ Q_0 &= \frac{2}{5}(c^2 - a^2). \end{aligned} \quad (6)$$

The Coulomb energy can be written as a product of E_C^0 , the Coulomb energy of an undeformed nucleus, and an anisotropy factor due to the deformation, B_C [38],

$$E_C = E_C^0 B_C, \quad (7)$$

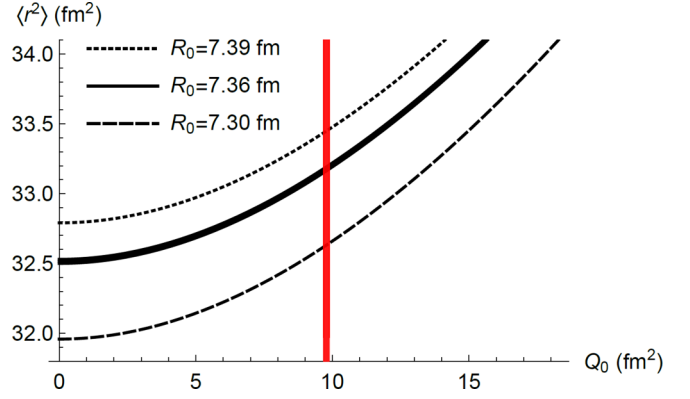


FIG. 1. Mean-square charge radius $\langle r^2 \rangle$ as a function of the intrinsic quadrupole moment Q_0 under the constant-volume ansatz for three volumes. The dashed lower curve corresponds to R_0 deduced from Hartree-Fock-Bogoliubov calculations using the SkM* functional, while the upper dotted curve is based on the SIII functional (see Table I). The middle curve, including errors, corresponds to $R_0 = 7.3615(16)$ fm deduced from the measurements by which (15) is obtained. The red line corresponds to the 1σ experimental range of Q_0 [39].

where

$$E_C^0 = \frac{3 q_e^2 Z^2}{5 R_0}, \quad (8)$$

$$B_C = \frac{(1 - e^2)^{1/3}}{2e} \ln \left(\frac{1 + e}{1 - e} \right). \quad (9)$$

Here q_e is the electron charge and Z is the number of protons.

In previous works [25], Q_0 and $\langle r^2 \rangle$ were treated as independent parameters. As such, calculation of ΔE_C involved derivatives of E_C both by Q_0 and by $\langle r^2 \rangle$:

$$\Delta E_C = \langle r^2 \rangle \frac{\partial E_C}{\partial \langle r^2 \rangle} \frac{\Delta \langle r^2 \rangle}{\langle r^2 \rangle} + Q_0 \frac{\partial E_C}{\partial Q_0} \frac{\Delta Q_0}{Q_0}. \quad (10)$$

With current experimental values $\langle r^2 \rangle = (5.76 \text{ fm})^2$ and $Q_0 = 9.8(1) \text{ fm}^2$ [39], Eqs. (7) and (10) give

$$\Delta E_C = -485 \text{ MeV} \frac{\Delta \langle r^2 \rangle}{\langle r^2 \rangle} + 11.6 \text{ MeV} \frac{\Delta Q_0}{Q_0}. \quad (11)$$

Substitution of measured changes in the mean-square radius and quadrupole moment [33], $\Delta \langle r^2 \rangle = 0.012(2) \text{ fm}^2$ and $\Delta Q_0 / Q_0 = -0.01(4)$, gives the limit, (3).

Let us now consider the ansatz of constant charge density between isomers, equivalent to the ansatz of constant volume. That is, R_0 and hence E_C^0 are kept constant in the isomeric transition. Therefore, changes in $\langle r^2 \rangle$ and Q_0 are coupled by (4) using (6). We show this dependence graphically in Fig. 1, and we can express it as

$$\frac{dQ_0}{d\langle r^2 \rangle} = 1 + \frac{2\langle r^2 \rangle}{Q_0} = 7.8, \quad (12)$$

where 7.8 corresponds to the experimental values. Substitution of (12) into (11) gives us the following result:

$$\Delta E_C = -180 \text{ MeV} \frac{\Delta \langle r^2 \rangle}{\langle r^2 \rangle}. \quad (13)$$

TABLE I. Theoretical values of the root-mean-square radius r_{rms} , Q_0 , ΔQ_0 , and Δr_{rms} calculated using the Hartree-Fock-Bogoliubov approach with two energy functionals, SkM* and SIII. In the fifth row we deduce the relationship between ΔQ_0 and $\Delta\langle r^2 \rangle$, which may be compared to the result of the constant-density ansatz, $dQ_0/d\langle r^2 \rangle = 7.8$. In the last two rows we show the change in Coulomb energy from direct calculation and using (14) with calculated values of charge radii.

	SkM*		SIII	
	n	p	n	p
r_{rms} (fm) ^a	5.8716	5.7078	5.8923	5.7769
Q_0 (fm ²) ^a	9.2608	9.3717	9.0711	9.1643
ΔQ_0 (fm ²) ^a	0.2647	0.2756	-60.051	-0.0495
Δr_{rms} (fm) ^a	0.0036	0.0039	-0.0005	-0.0005
$\Delta Q_0/\Delta\langle r^2 \rangle$	6.26	6.19	8.76	8.57
ΔE_C (MeV) ^b		-0.307		0.001
ΔE_C (MeV) ^a		-0.287		0.036

^aFrom Ref. [23], Table II and Eq. (14) for ΔE_C .

^bFrom Ref. [23], Table I.

The relation between changes in $\langle r^2 \rangle$ and Q_0 can also be obtained from nuclear calculations where the constant-density ansatz is not assumed. Results of the Hartree-Fock-Bogoliubov calculations of [23] are summarized in Table I. We extract $\Delta Q_0/\Delta\langle r^2 \rangle$ for two energy functionals, SkM* and SIII, and for both protons and neutrons (for details see [23]). In all cases the derivative is close to that predicted by the constant-density ansatz.

In addition to the results reproduced in Table I, Ref. [23] presents Hartree-Fock calculations (which do not include pairing) using the same functionals. For SkM*, the Hartree-Fock calculations give the wrong sign for $\langle r^2 \rangle$, while for SIII the change between isomers is very small and susceptible to numerical noise. Nevertheless, in both cases the Hartree-Fock calculations give reasonably close values for the derivative.

For the Hartree-Fock-Bogoliubov calculations, the SkM* better reproduces the measured energy interval and change in the nuclear radius between the isomers. We take the average of the SkM* value $dQ_0/d\langle r^2 \rangle$ for protons and the experimental value from (12) as our estimate of the derivative and their difference as an estimate of the derivative's uncertainty, giving $dQ_0/d\langle r^2 \rangle = 7.0(1.6)$.¹ With this we write the change in Coulomb energy ΔE_C in terms of the change in mean-square radius at the physical point as

$$\Delta E_C = -210(60) \text{ MeV} \frac{\Delta\langle r^2 \rangle}{\langle r^2 \rangle}. \quad (14)$$

The last row in Table I lists the results of application of this formula to the nuclear calculations of Δr_{rms} from [23]. Filling in the measured $\Delta\langle r^2 \rangle = 0.0105(13) \text{ fm}^2$ [40] and

$\langle r^2 \rangle = (5.76 \text{ fm})^2$ [41], we obtain

$$\Delta E_C = -0.067(19) \text{ MeV}, \quad (15)$$

$$K = -0.82(25) \times 10^4. \quad (16)$$

Since our model does not rely on the measured ΔQ_0 , which gives the biggest error in Eq. (3), the result of (15) has a smaller error than (3). We also predict $\Delta Q_0/Q_0 = 0.0075(20) \text{ fm}^2$, which is within the experimental error of $\Delta Q_0/Q_0 = -0.01(4) \text{ fm}^2$ presented in Ref. [33].

III. EFFECT OF OCTUPOLE DEFORMATION

Nuclear calculations of N. Minkov and A. Pálffy suggest that the ^{229}Th nucleus has an octupole deformation [7,42] (see also a recent experiment [43]). They therefore describe the nucleus using a quadrupole-octupole model, obtaining a fair comparison to experimental results [7,42]. This prompts us to include an octupole deformation in addition to the quadrupole deformation.

To facilitate this we describe the nucleus shape by its radius vector in axially symmetric spherical harmonics [44,45],

$$r(\theta) = R_s \left[1 + \sum_{n=1}^N (\beta_n Y_{n0}(\theta)) \right], \quad (17)$$

where the coefficients β_n are called deformation parameters and $N = 3$ for the quadrupole-octupole model (pear shape). The length R_s is defined by normalization of the volume to that of the undeformed nucleus,

$$\frac{2\pi}{3} \int_0^\pi r^3(\theta) \sin \theta d\theta = \frac{4\pi R_0^3}{3}. \quad (18)$$

The parameter β_1 is set such that the center of mass of the shape is at the origin of the coordinate system.

The mean-square radius and the intrinsic quadrupole moment of the nucleus are related to the deformation parameters β_2 and β_3 through $r(\theta)$ by

$$\langle r^2 \rangle = \int r^2(\theta) \rho(r) d^3r, \quad (19)$$

$$Q_0 = 2 \int r^2(\theta) P_2(\cos \theta) \rho(r) d^3r, \quad (20)$$

where $\rho(r)$ is the charge density divided by the total charge. The factor 2 in Eq. (20) is a matter of definition [46] and fits with the special case of Q_0 in Eq. (6).

To determine β_2 for the pear shape, we solve (19) and (20) using the experimental values of Q_0 and $\langle r^2 \rangle$. As the octupole moment of ^{229}Th has not yet been measured, we take $\beta_3 = 0.115$ from nuclear calculations [7]. We arrive at $\beta_2 = 0.22$ and $R_s = 7.3 \text{ fm}$. This value of β_2 is fairly close to the theoretical prediction of [7], $\beta_2 = 0.24$, and is not particularly sensitive to the chosen value of β_3 (see Fig. 2).

In this model the anisotropy factor is [32]

$$B_C = 1 - \frac{5}{4\pi} \sum_{n=2}^{\infty} \frac{n-1}{2n+1} \beta_n^2 + O(\beta_n^3). \quad (21)$$

Higher-order terms do not change our results within stated errors. With the aforementioned values for β_2 and β_3 , we

¹Alternatively, we could use the average value between SkM* and SIII numbers for the derivatives $\Delta Q_0/\Delta r^2$ but the change in the result would be within the error bars.

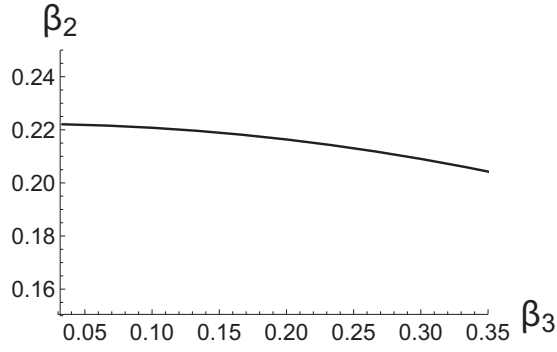


FIG. 2. Deformation parameter β_2 derived using (19) and (20) with experimental values of $Q_0 = 9.8 \text{ fm}^2$ and $\langle r^2 \rangle = (5.76 \text{ fm})^2$, as a function of β_3 .

obtain for the constant-density ansatz (i.e., constant E_C^0),

$$\Delta E_C = -76 \text{ MeV } \Delta \beta_2^2 - 108 \text{ MeV } \Delta \beta_3^2, \quad (22)$$

$$\approx -190 \text{ MeV } \frac{\Delta \langle r^2 \rangle}{\langle r^2 \rangle} - 0.42 \text{ MeV } \frac{\Delta \beta_3^2}{\beta_3^2}. \quad (23)$$

Equation (23) is obtained by substituting (25) and is in good agreement with (13). We see that the sensitivity of the nuclear clock to α variation does not depend strongly on the octupole moment.

IV. DISCUSSION

The constant-volume ansatz used in the present work may be tested in experiments. This ansatz allows one to relate the change in nuclear quadrupole moment to the change in nuclear charge radius. Therefore, determination of $\Delta \langle r^2 \rangle$ by measuring the field isotope shift of atomic transitions and extraction of ΔQ_0 from the hyperfine structure or nuclear rotational bands gives a measure of the change in the nuclear charge density.

TABLE II. Sensitivity of Mössbauer transitions to variation of the fine-structure constant. Coulomb energy shifts ΔE_C and enhancement factors K are calculated using data from quadrupole moments [58] and isomeric shift measurements [59]. In columns 2 and 4 we use the constant-density ansatz, where we have assumed a 25% error from the ansatz, Eq. (12), and 50% error in the values of $\Delta \langle r^2 \rangle$ from Mössbauer isomer shifts [59]. The Eu isomers have more accurate values of $\Delta \langle r^2 \rangle$ taken from muonic x-ray and Mössbauer data [60–62]. Columns 3 and 5 use experimental values of ΔQ_0 from [58] in the general formula, (10). The ground-state $\langle r^2 \rangle$ values are taken from [41]. ^{229}Th results are shown for comparison (discussed in the text).

	ΔE_C (MeV)		$ K $	
	Constant density	General	Constant density	General
^{151}Eu , 22 keV	−0.099 (51)	−0.099 (85)	4.6 (2.4)	4.6 (4.0)
^{153}Eu , 103 keV	0.32 (18)	0.02 (15)	3.1 (1.8)	0.2 (1.5)
^{155}Gd , 105 keV	0.030 (22)	0.08 (32)	0.28 (21)	0.8 (3.1)
^{157}Gd , 64 keV	−0.055 (41)	−0.06 (21)	0.86 (63)	0.9 (3.3)
^{161}Dy , 75 keV	−0.031 (23)	0.29 (55)	0.42 (31)	3.8 (7.4)
^{181}Ta , 6 keV	0.19 (13)	0.20 (26)	30 (21)	32 (41)
^{243}Am , 84 keV	0.23 (17)	0.45 (75)	2.8 (2.0)	5.4 (9.0)
^{229}Th , 8 eV	−0.067 (19)	−0.26 (39)	0.82 (25) 10^4	3.1 (4.8) 10^4

A specific procedure can be encoded in the change in mean-square radius [47,48]

$$\Delta \langle r^2 \rangle = \Delta \langle r^2 \rangle_{\text{sph}} + \Delta \langle r^2 \rangle_{\text{def}}. \quad (24)$$

Here the spherical part $\Delta \langle r^2 \rangle_{\text{sph}}$ describes the change in nuclear volume, i.e., volume contribution, and $\Delta \langle r^2 \rangle_{\text{def}}$ describes the deformation part assuming a constant volume, i.e., shape contribution. The latter can be expressed by deformation parameters [47–50]

$$\Delta \langle r^2 \rangle = \Delta \langle r^2 \rangle_{\text{sph}} + \frac{5}{4\pi} \langle r^2 \rangle_{\text{sph}} (\Delta \beta_2^2 + \Delta \beta_3^2 + \dots), \quad (25)$$

where $\langle r^2 \rangle_{\text{sph}}$ is the mean-square charge radius of the nucleus assuming a spherical distribution. Equation (25) can be used in the future to test the volume-conservation hypothesis in isomers, once the $\Delta \beta$ is determined to higher accuracy.

Using existing experimental data [33] we may conclude that the relative change in volume between ^{229}Th isomers is less than a few parts per thousand, while the calculations in [23] imply a fractional volume change of about 5×10^{-4} . This gives a quantitative evaluation of the constant-volume ansatz, which at times is used in the literature (see, e.g., [51–53]).

The sensitivity to potential variation of α , i.e., the enhancement factor K , is three orders of magnitude larger than that of the most sensitive atomic clocks. For the present experimental bound, $\delta\alpha/\alpha \lesssim 10^{-17}$ per year, the frequency shift is up to ~ 200 Hz per year. Since such a frequency shift is six orders of magnitude larger than the projected accuracy of the nuclear clock [9], an unexplored range of $\delta\alpha$ may be tested. As discussed in Refs. [54–56], the interaction between low-mass scalar dark matter and the electromagnetic field leads to oscillatory variation of α . Therefore, the improvement in the sensitivity to α variation by six orders of magnitude afforded by such a clock should also lead to improved sensitivity in the search for low-mass scalar dark matter.

We should note a certain similarity between the research on ^{229}Th isomeric transition and the very extensive experimental and theoretical studies of isomeric (chemical) shifts in Möss-

bauer spectroscopy, which also involve effects of the change in the nuclear charge radius and electric quadrupole moment between the ground and the excited nuclear states connected by a γ transition. X-ray studies of muonic atoms are also able to deduce these nuclear properties. Using the same technique as in ^{229}Th we calculated the Coulomb energy difference ΔE_C and the relative sensitivity to α variation K for nuclei where we have found sufficient experimental data. The results are presented in Table II. The enhancement factors K for Mössbauer transitions ($K = \Delta E_C/E_{\text{is}} \sim 1\text{--}30$) are much smaller than K for ^{229}Th since the energy of Mössbauer transitions is much higher, $E \sim 5\text{--}100$ keV. However, they are comparable or even larger than $K \sim 0.1\text{--}6$ in atomic clocks. The energy resolution in Mössbauer γ transitions may be as good as 10^{-18} ; see, e.g., the measurement of the gravitational red shift in Ref. [57], where such a resolution was achieved after 5 days of measurements. This is even higher than that achieved recently in optical transitions, 10^{-17} to 10^{-18} . However, the

authors of Ref. [57] noted a problem with solid-state effects, which are difficult to control.

The results in Table II serve as a test of the constant density ansatz. The predictions for ΔE_C using the constant-density model and using the more general formula, (10), with experimental data for both $\Delta\langle r^2 \rangle$ and ΔQ_0 agree within the error bars. In these examples, using one of the values of $\Delta\langle r^2 \rangle$ or ΔQ_0 , the constant-density ansatz reproduces the other value within error bars. This provides a check on the validity of the ansatz.

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- [1] E. Peik and C. Tamm, *Europhys. Lett.* **61**, 181 (2003).
 [2] C. W. Reich and R. G. Helmer, *Phys. Rev. Lett.* **64**, 271 (1990).
 [3] R. G. Helmer and C. W. Reich, *Phys. Rev. C* **49**, 1845 (1994).
 [4] B. R. Beck, J. A. Becker, P. Beiersdorfer, G. V. Brown, K. J. Moody, J. B. Wilhelmy, F. S. Porter, C. A. Kilbourne, and R. L. Kelley, *Phys. Rev. Lett.* **98**, 142501 (2007).
 [5] B. R. Beck, J. A. Becker, P. Beiersdorfer, G. V. Brown, K. J. Moody, C. Y. Wu, J. B. Wilhelmy, F. S. Porter, C. A. Kilbourne, and R. L. Kelley, Report No. LLNL-PROC-415170 (Los Alamos National Laboratory, Los Alamos, NM, 2009).
 [6] E. V. Tkalya, C. Schneider, J. Jeet, and E. R. Hudson, *Phys. Rev. C* **92**, 054324 (2015).
 [7] N. Minkov and A. Pálffy, *Phys. Rev. Lett.* **118**, 212501 (2017).
 [8] S. G. Porsev, V. V. Flambaum, E. Peik, and Chr. Tamm, *Phys. Rev. Lett.* **105**, 182501 (2010).
 [9] C. J. Campbell, A. G. Radnaev, A. Kuzmich, V. A. Dzuba, V. V. Flambaum, and A. Derevianko, *Phys. Rev. Lett.* **108**, 120802 (2012).
 [10] P. G. Thirolf, B. Seiferle, and L. von der Wense, *J. Phys. B: At. Mol. Opt. Phys.* **52**, 203001 (2019).
 [11] L. von der Wense, B. Seiferle, and P. G. Thirolf, *Meas. Tech.* **60**, 1178 (2018).
 [12] E. V. Tkalya, *Phys. Rev. Lett.* **124**, 242501 (2020).
 [13] B. S. Nickerson, M. Pimon, P. V. Bilous, J. Gugler, K. Beeks, T. Sikorsky, P. Mohn, T. Schumm, and A. Pálffy, *Phys. Rev. Lett.* **125**, 032501 (2020).
 [14] S. G. Porsev and V. V. Flambaum, *Phys. Rev. A* **81**, 032504 (2010).
 [15] S. G. Porsev, V. V. Flambaum, *Phys. Rev. A* **81**, 042516 (2010).
 [16] A. Y. Dzublik, *Phys. Rev. C* **102**, 024604 (2020).
 [17] B. Seiferle, L. von der Wense, P. V. Bilous, I. Amersdorfer, C. Lemell, F. Libisch, S. Stellmer, T. Schumm, C. E. Düllmann, A. Pálffy, and P. G. Thirolf, *Nature* **573**, 243 (2019).
 [18] T. Masuda, A. Yoshimi, A. Fujieda, H. Fujimoto, H. Haba, H. Hara, T. Hiraki, H. Kaino, Y. Kasamatsu, S. Kitao, K. Konashi, Y. Miyamoto, K. Okai, S. Okubo, N. Sasao, M. Seto, T. Schumm, Y. Shigekawa, K. Suzuki, S. Stellmer, K. Tamasaku, S. Uetake, M. Watanabe, T. Watanabe, Y. Yasuda, A. Yamaguchi, Y. Yoda, T. Yokokita, M. Yoshimura, and K. Yoshimura, *Nature* **573**, 238 (2019).
 [19] A. Yamaguchi, H. Muramatsu, T. Hayashi, N. Yuasa, K. Nakamura, M. Takimoto, H. Haba, K. Konashi, M. Watanabe, H. Kikunaga, K. Maehata, N. Y. Yamasaki, and K. Mitsuda, *Phys. Rev. Lett.* **123**, 222501 (2019).
 [20] T. Sikorsky, J. Geist, D. Hengstler, S. Kempf, L. Gastaldo, C. Enss, C. Mokry, J. Runke, C. E. Düllmann, P. Wobrauschek, K. Beeks, V. Rosecker, J. H. Sterba, G. Kazakov, T. Schumm, and A. Fleischmann, *Phys. Rev. Lett.* **125**, 142503 (2020).
 [21] V. V. Flambaum, *Phys. Rev. Lett.* **97**, 092502 (2006).
 [22] V. V. Flambaum, N. Auerbach, and V. F. Dmitriev, *Europhys. Lett.* **85**, 50005 (2009).
 [23] E. Litvinova, H. Feldmeier, J. Dobaczewski, and V. V. Flambaum, *Phys. Rev. C* **79**, 064303 (2009).
 [24] P. G. Thirolf, B. Seiferle, and L. von der Wense, *Ann. Phys. (Berlin)* **531**, 1800381 (2019).
 [25] J. C. Berengut, V. A. Dzuba, V. V. Flambaum, and S. G. Porsev, *Phys. Rev. Lett.* **102**, 210801 (2009).
 [26] J. C. Berengut and V. V. Flambaum, *Nuclear Phys. News* **20**, 19 (2010).
 [27] X.-T. He and Z.-Z. Ren, *J. Phys. G: Nucl. Part. Phys.* **34**, 1611 (2007).
 [28] V. V. Flambaum and R. B. Wiringa, *Phys. Rev. C* **79**, 034302 (2009).
 [29] V. A. Dzuba, V. V. Flambaum, and J. K. Webb, *Phys. Rev. Lett.* **82**, 888 (1999).
 [30] V. A. Dzuba, V. V. Flambaum, and J. K. Webb, *Phys. Rev. A* **59**, 230 (1999).
 [31] V. V. Flambaum and S. G. Porsev, *Phys. Rev. A* **80**, 064502 (2009).
 [32] R. W. Hasse and W. D. Myers, *Geometrical Relationships of Macroscopic Nuclear Physics* (Springer-Verlag, Heidelberg, 1988).
 [33] J. Thielking, M. V. Okhapkin, P. Glowacki, D. M. Meier, L. von der Wense, B. Seiferle, C. E. Düllmann, P. G. Thirolf, and E. Peik, *Nature* **556**, 321 (2018).

- [34] N. Huntemann, B. Lipphardt, C. Tamm, V. Gerginov, S. Weyers, and E. Peik, *Phys. Rev. Lett.* **113**, 210802 (2014).
- [35] V. V. Flambaum and V. A. Dzuba, *Can. J. Phys.* **87**, 25 (2009).
- [36] V. A. Dzuba, V. V. Flambaum, and S. Schiller, *Phys. Rev. A* **98**, 022501 (2018).
- [37] M. S. Safronova, *Ann. Phys. (Berlin)* **531**, 1800364 (2019).
- [38] B. C. Carlson, *J. Math. Phys.* **2**, 441 (1961).
- [39] Note that the expressions for the laboratory and spectroscopic quadrupole moments differ by a factor of Z between [25,33]. That is because the factor Z was absorbed into the definition of the intrinsic quadrupole moment Q_0 in Ref. [33]. In this paper Q_0 is defined without the factor Z , so the quoted number for Q_0 in Ref. [33] equals Q_0Z in our notation. For the units of Q_0 we use fm^2 ; another common choice of units is eb (where eb stands for electron-barn, $1 \text{ eb} = 1.6022 \times 10^{-47} \text{ C m}^2$), such that $Q \text{ (eb)} = 10^2 eQ \text{ (fm}^2\text{)}$.
- [40] M. S. Safronova, S. G. Porsev, M. G. Kozlov, J. Thielking, M. V. Okhapkin, P. Głowacki, D. M. Meier, and E. Peik, *Phys. Rev. Lett.* **121**, 213001 (2018).
- [41] I. Angeli and K. P. Marinova, *At. Data Nucl. Data Tables* **99**, 69 (2013).
- [42] N. Minkov and A. Pálffy, *Phys. Rev. Lett.* **122**, 162502 (2019).
- [43] M. M. R. Chishti, D. O'Donnell, G. Battaglia, M. Bowry, D. A. Jaroszynski, B. S. Nara Singh, M. Scheck, P. Spagnoletti, and J. F. Smith, *Nat. Phys.* **16**, 853 (2020).
- [44] P. Möller, J. R. Nix, W. D. Myers, and W. J. Swiatecki, *At. Data Nucl. Data Tables* **59**, 185 (1995).
- [45] P. A. Butler and W. Nazarewicz, *Rev. Mod. Phys.* **68**, 349 (1996).
- [46] J. D. Jackson, *Classical Electrodynamics* (Wiley, New York, 1962), sec. 4.2 [3rd ed. (1999).]
- [47] S. Ahmad, W. Klempt, R. Neugart, E. Otten, P.-G. Reinhard, G. Ulm, and K. Wendt, *Nucl. Phys. A* **483**, 244 (1988).
- [48] G. Ulm, S. K. Bhattacharjee, P. Dabkiewicz, G. Huber, H. J. Kluge, T. Köhl, H. Lochmann, E. W. Otten, K. Wendt, S. A. Ahmad, W. Klempt, R. Neugart, and the ISOLDE Collaboration, *Z. Phys. A* **325**, 247 (1986).
- [49] A. Bohr and B. R. Mottelson, *Nuclear Structure Volume II: Nuclear Deformations* (World Scientific, London, 1998).
- [50] W. Kälber, J. Rink, K. Bekk, W. Faubel, S. Göring, G. Meisel, H. Rebel, and R. C. Thompson, *Z. Phys. A* **334**, 103 (1989).
- [51] D. T. Yordanov, D. L. Balabanski, M. L. Bissell, K. Blaum, I. Budinčević, B. Cheal, K. Flanagan, N. Frömmgen, G. Georgiev, Ch. Geppert, M. Hammen, M. Kowalska, K. Kreim, A. Krieger, J. Meng, R. Neugart, G. Neyens, W. Nörtershäuser, M. M. Rajabali, J. Papuga, S. Schmidt, and P. W. Zhao, *Phys. Rev. Lett.* **116**, 032501 (2016).
- [52] A. Boucenna, S. Madjber, and A. Bouketir, [arXiv:nucl-th/0305026](https://arxiv.org/abs/nucl-th/0305026).
- [53] M. Avgoulea, Y. P. Gangrsky, K. P. Marinova, S. G. Zemlyanoi, S. Fritzsche, D. Iablonskyi, C. Barbieri, E. C. Simpson, P. D. Stevenson, J. Billowes, P. Campbell, B. Cheal, B. Tordoff, M. L. Bissell, D. H. Forest, M. D. Gardner, G. Tungate, J. Huikari, A. Nieminen, H. Penttilä, and J. Äystö, *J. Phys. G* **38**, 025104 (2011).
- [54] A. Arvanitaki, J. Huang, and K. Van Tilburg, *Phys. Rev. D* **91**, 015015 (2015).
- [55] K. Van Tilburg, N. Leefler, L. Bougas, and D. Budker, *Phys. Rev. Lett.* **115**, 011802 (2015).
- [56] Y. V. Stadnik and V. V. Flambaum, *Phys. Rev. Lett.* **115**, 201301 (2015).
- [57] W. Potzel, C. Schafer, M. Steiner, H. Karzel, W. Schiessl, M. Peter, G. M. Kalvius, T. Katila, E. Ikonen, P. Helisto and J. Hietaniemi, *Hyperfine Interact.* **72**, 197 (1992).
- [58] N. J. Stone, *At. Data Nucl. Data Tables* **111–112**, 1 (2016).
- [59] G. M. Kalvius and G. K. Shenoy, *At. Data Nucl. Data Tables* **14**, 639 (1974).
- [60] Y. Tanaka, R. M. Steffen, E. B. Shera, W. Reuter, M. V. Hoehn, and J. D. Zumbro, *Phys. Rev. C* **29**, 1897 (1984).
- [61] J. Hess, *Nucl. Phys. A* **142**, 273 (1970).
- [62] E. Steichele, S. Hübner, and P. Kienle, *Phys. Lett.* **14**, 321 (1965).

Correction: The works cited in Refs. [1] and [2] were erroneously changed by the production group during the proof cycle and have been restored.