Sensitivity of ^{229}Th nuclear clock transition to variation of the fine-structure constant

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A nuclear clock has been proposed 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 accuracy high enough to resolve non-zero sensitivity. Here we determine this sensitivity using existing measurements of the change in mean-square charge radius, coupled with the ansatz of constant nuclear density. The enhancement factor for α-variation is $K = -(0.9 \pm 0.3) \times 10^4$. For the current experimental limit $\delta\alpha/\alpha \lesssim 10^{-17}$ per year, the corresponding frequency shift is $\sim$200 Hz. This shift is six orders of magnitude larger than the projected accuracy of the nuclear clock, paving the way for increased accuracy for 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.

The first excited isomeric state of thorium-229, $^{229\text{m}}$Th, is a candidate for the first nuclear optical clock [1]. This is due to the state's low excitation energy of several electron-volts [2–5] (the lowest of all known isomeric states) and long radiative lifetime of up to 10^4 seconds [6, 7]. Several theoretical and experimental groups are making rapid progress to using $^{229\text{m}}$Th as a reference for a clock with unprecedented accuracy [8–10].

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 $^{229\text{m}}$Th atoms [11], yielding an excitation energy $E_{\text{is}} = 8.28\,(17)\,\text{eV}$. Another approach, using γ-ray spectroscopy at 29.2 keV, obtained $E_{\text{is}} = 8.30\,(92)\,\text{eV}$ [12, 13]. More recently, $E_{\text{is}} = 8.10\,(17)\,\text{eV}$ was reported [14].

The $^{229\text{m}}$Th nuclear clock is expected to be a sensitive probe for time variation in the fine-structure constant α [15–21]. This sensitivity comes about because the change in Coulomb energy between the isomers, which depends linearly on α, is almost entirely cancelled by the nuclear force contribution which has only weak α-dependence. 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 [15]

$$h\,\delta f = \Delta E_{\text{C}} \frac{\delta\alpha}{\alpha}\,, \tag{1}$$

where ΔE_{C} is the difference in Coulomb energy between the two isomers. The enhancement factor K is defined by

$$\frac{\delta f}{f} = K \frac{\delta\alpha}{\alpha}\,, \tag{2}$$

where $K = \Delta E_{\text{C}}/E_{\text{is}}$. Therefore, to find the sensitivity of $^{229\text{m}}$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 pointlike nucleus (r is the distance from the nucleus), nuclear systems can have deformed shapes as the potential originates from the nucleons themselves. Ref. [20] showed that, by modeling the nucleus as a prolate spheroid [22], ΔE_{C} can be deduced from measurements of the change in nuclear charge radius and quadrupole moment between the isomeric and ground states. Using this model with measurements of nuclear parameters, the authors in [23] give a value of

$$\Delta E_{\text{C}} = -0.29\,(43)\,\text{MeV}\,, \tag{3}$$

where the dominant source of error is the uncertainty in measured quadrupole moments of the ground and the exited states. Such a ΔE_{C} is consistent with a K value anywhere between zero and 10^5. This can be compared to a K of about 0.1–6 for current atomic clocks [24–29].

In this Letter 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 constant charge density between isomers. We verify that this assumption gives a relation that is consistent with previous results from nuclear theory [18]. 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.

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

$$a^2 c = R_0^3\,. \tag{4}$$

The eccentricity e is defined by

$$e^2 = 1 - \frac{a^2}{c^2}\,, \tag{5}$$

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

$$\langle r^2\rangle = \frac{1}{5}\left(2a^2 + c^2\right)\,, \tag{6}$$
$$Q_0 = \frac{2}{5}\left(c^2 - a^2\right)\,.$$

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 [30]:

$$E_C = E_C^0\, B_C\,, \tag{7}$$

where

$$E_C^0 = \frac{3}{5}\frac{q_e^2 Z^2}{R_0}\,, \tag{8}$$
$$B_C = \frac{(1-e^2)^{1/3}}{2e}\ln\left(\frac{1+e}{1-e}\right)\,. \tag{9}$$

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

In previous works [20], 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 $\langle r^2\rangle$:

$$\Delta E_C = \langle r^2\rangle \frac{\partial E_C}{\partial\langle r^2\rangle}\frac{\Delta\langle r^2\rangle}{\langle r^2\rangle} + Q_0\frac{\partial E_C}{\partial Q_0}\frac{\Delta Q_0}{Q_0}\,. \tag{10}$$

With current experimental values $\langle r^2\rangle = (5.76\text{ fm})^2$ and $Q_0 = 9.8(1)\text{ fm}^2$ [31], Eqs. (7) and (10) give

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

Substitution of measured changes in mean-square radius and quadrupole moment [23], $\Delta\langle r^2\rangle = 0.012\,(2)\text{ fm}^2$ 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 Figure 1, and we can express it as

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

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

$$\Delta E_C = -180\,\text{MeV}\,\frac{\Delta\langle r^2\rangle}{\langle r^2\rangle}\,. \tag{13}$$

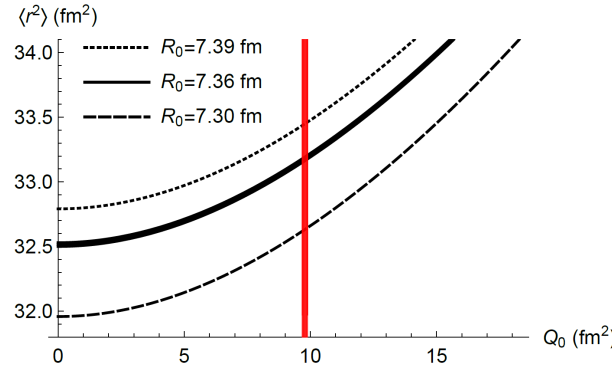

FIG. 1. Mean-square charge radius $\langle r^2\rangle$ as a function of intrinsic quadrupole moment Q_0 under the constant-volume ansatz for three different volumes. The dashed lower curve corresponds to R_0 deduced from Hartree-Fock-Bogoliubov calculations using the SkM* functional, while the upper dotted curve is based on 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 [31].

TABLE I. Theoretical values of root-mean-square radius $r_{\rm rms}$, Q_0, ΔQ_0, and $\Delta r_{\rm rms}$ calculated using 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
$r_{\rm rms}$ (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	−0.0516	−0.0495
$\Delta r_{\rm rms}$ (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

[a] From Ref. [18], Table II and Eq. (14) for ΔE_C.
[b] From Ref. [18], 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 [18] are summarized in Table I. We extract $\Delta Q_0/\Delta\langle r^2\rangle$ for two different energy functionals, SkM* and SIII, and for both protons and neutrons (for details see [18]). 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. [18] 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 changes in nuclear parameters 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. With this we write the change in Coulomb energy $\Delta E_{\rm C}$ in terms of the change in mean-square radius at the physical point as

$$\Delta E_{\rm C} = -210\,(60)\,\text{MeV}\ \frac{\Delta\langle r^2\rangle}{\langle r^2\rangle}\,. \tag{14}$$

The last row of Table I shows the result of application of this formula to the nuclear calculations of $\Delta r_{\rm rms}$ from [18]. Filling in the measured $\Delta\langle r^2\rangle = 0.012\,(2)\ \text{fm}^2$ and $\langle r^2\rangle = (5.76\ \text{fm})^2$ [23], we obtain

$$\Delta E_{\rm C} = -0.076\,(25)\,\text{MeV}\,, \tag{15}$$

$$K = -0.9\,(3)\times 10^4\,. \tag{16}$$

We observe that $|K|$ could be over 9000! [32] Since our model does not rely on the measured ΔQ_0, which gives the biggest error in (3), the result in (15) has smaller error than (3). Under the constant-volume ansatz we predict $\Delta Q_0 = 0.084\,(24)\,\text{fm}^2$, which is within the experimental error presented in [23].

Theoretical nuclear calculations of A. Pálffy and N. Minkov suggest that the ^{229}Th nucleus has an octupole deformation [7, 33] (see also the recent experiment [34]). They therefore describe the nucleus using a quadrupole-octupole model, obtaining a fair comparison to experimental results [7, 33]. 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 [35, 36]

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

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

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

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

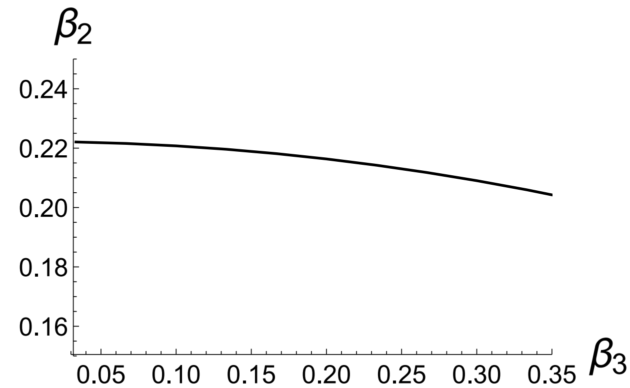

FIG. 2. Deformation parameter β_2 derived using (19) and (20) with experimental values of $Q_0 = 9.8\,\text{fm}^2$ and $\langle r^2\rangle = (5.76\,\text{fm})^2$, as a function of β_3.

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

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

where $\rho(r)$ is the charge density divided by the total charge. The factor 2 in (20) is a matter of definition [37], and fits with the special case of Q_0 in (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 $R_s = 7.3$ 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 [22]

$$B_{\rm C} = 1 - \frac{5}{4\pi}\sum_{n=2}^{\infty}\frac{n-1}{2n+1}\beta_n^2 + \mathcal{O}(\beta_n^3)\ . \tag{21}$$

Higher-order terms do not change our results within stated errors. With the aforementioned values for β_2 and β_3, we obtain for the constant-density ansatz (i.e. constant E_C^0),

$$\Delta E_{\rm C} = -76\,\text{MeV}\ \Delta\beta_2^2 - 108\,\text{MeV}\ \Delta\beta_3^2 \tag{22}$$

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

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.

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.

A specific procedure can be encoded in the change of mean-square radius [38, 39]

$$\Delta\langle r^2\rangle = \Delta\langle r^2\rangle_{\rm sph} + \Delta\langle r^2\rangle_{\rm def}\,. \qquad (24)$$

Here the spherical part $\Delta\langle r^2\rangle_{\rm sph}$ describes the change in nuclear volume, i.e. volume contribution, and $\Delta\langle r^2\rangle_{\rm def}$ describes the deformation part assuming a constant volume, i.e. shape contribution. The latter can be expressed by deformation parameters [38–41]

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

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

Using existing experimental data [23] we may conclude that the relative change in volume between ^{229}Th isomers is less than a few parts per thousand, while the calculations of [18] 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. [42–44].

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 $\sim$200 Hz. Since such a frequency shift is six orders of magnitude larger than the projected accuracy of the nuclear clock [8], an unexplored range of $\delta\alpha$ may be tested. As discussed in Refs. [45–47], the interaction between low-mass scalar dark matter and electromagnetic field leads to oscillatory variation of α. Therefore, the six orders-of-magnitude improvement in the sensitivity to α variation afforded by such a clock should also lead to improved sensitivity in the search for low-mass scalar dark matter.

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