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, $\alpha$. 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 $\alpha$-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 $\alpha$-variation.

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

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

\[
h \delta f = \Delta E_C \frac{\delta \alpha}{\alpha}, \tag{1}
\]

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

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], $\Delta E_C$ 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

\[
\Delta E_C = -0.29 (43) \text{ MeV}, \tag{3}
\]

where the dominant source of error is the uncertainty in measured quadrupole moments of the ground and the exited states. Such a $\Delta E_C$ 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 $\Delta E_C$ 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

\[
a^2 c = R_0^3. \tag{4}
\]
where the mean-square radius \( \langle r^2 \rangle \) and the quadrupole moment \( Q_0 \) are

\[
\langle r^2 \rangle = \frac{1}{5} \left( 2a^2 + c^2 \right),
\]

\[
Q_0 = \frac{2}{5} \left( c^2 - a^2 \right).
\]

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]:

\[
E_C = E_C^0 B_C,
\]

where

\[
E_C^0 = \frac{3q_e^2Z^2}{5R_0},
\]

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

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 \( \Delta E_C \) involved derivatives of \( E_C \) both by \( Q_0 \) and by \( \langle r^2 \rangle \):

\[
\Delta E_C = \langle r^2 \rangle \frac{\partial E_C}{\partial \langle r^2 \rangle} \Delta \langle r^2 \rangle + Q_0 \frac{\partial E_C}{\partial Q_0} \Delta Q_0.
\]

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

\[
\Delta E_C = -485 \text{ MeV} \frac{\Delta \langle r^2 \rangle}{\langle r^2 \rangle} + 11.6 \text{ MeV} \frac{\Delta Q_0}{Q_0}.
\]

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

\[
\frac{dQ_0}{d\langle r^2 \rangle} = 1 + \frac{2\langle r^2 \rangle}{Q_0} = 7.8,
\]

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

\[
\Delta E_C = -180 \text{ MeV} \frac{\Delta \langle r^2 \rangle}{\langle r^2 \rangle}.
\]

The eccentricity \( e \) is defined by

\[
e^2 = 1 - \frac{a^2}{c^2},
\]

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

\[
\langle r^2 \rangle = \frac{1}{5} \left( 2a^2 + c^2 \right),
\]

\[
Q_0 = \frac{2}{5} \left( c^2 - a^2 \right).
\]

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 \( R_0 = 7.3615(16) \text{ fm} \) deduced from the measurements by which (15) is obtained. The red line corresponds to the 1σ experimental range of \( Q_0 \) [31].

| \( r_{\text{rms}} \) (fm) | \( n \) | \( p \) | \( n \) | \( p \) |
|-----------------|-----|-----|-----|-----|
| SkM*            | 5.8716 | 5.7078 | 5.8923 | 5.7769 |
| Q_0 (fm^2)     | 9.2608 | 9.3717 | 9.0711 | 9.1643 |
| \( \Delta Q_0 \) (fm^2) | 0.2647 | 0.2756 | -0.0516 | -0.0495 |
| \( \Delta r_{\text{rms}} \) (fm) | 0.0036 | 0.0039 | -0.0005 | -0.0005 |
| \( \Delta Q_0/\Delta \langle r^2 \rangle \) | 6.26 | 6.19 | 8.76 | 8.57 |
| \( \Delta E_C \) (MeV) | -0.307 | -0.307 | 0.001 | 0.001 |
| \( \Delta E_C \) (MeV) | -0.287 | -0.287 | 0.036 | 0.036 |

a From Ref. [18], Table II and Eq. (14) for \( \Delta E_C \).

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 $dQ_0/d(r^2)$ 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 $\Delta E_C$ 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 of Table I shows the result of application of this formula to the nuclear calculations of $\Delta \text{rms}$ 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

$$\Delta E_C = -0.076 (25) \text{ MeV}, \quad (15)$$

$$K = -0.9 (3) \times 10^4. \quad (16)$$

We observe that $|K|$ could be over 9000! [32] Since our model does not rely on the measured $\Delta Q_0$, 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. Pálfy 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]

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

where the coefficients $\beta_n$ 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

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

The parameter $\beta_3$ 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

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

Equation (23) is obtained by substituting (25), and is in good agreement with (13). We see that the sensitivity of the nuclear clock to $\alpha$-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 parameters $\beta_2$ and $\beta_3$ 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 (20) is a matter of definition [37], and fits with the special case of $Q_0$ in (6).

To determine $\beta_2$ 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 $\beta_2$ is fairly close to the theoretical prediction of [7], $\beta_2 = 0.24$, and is not particularly sensitive to the chosen value of $\beta_3$ (see Fig. 2).

In this model the anisotropy factor is [22]

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

Higher-order terms do not change our results within stated errors. With the aforementioned values for $\beta_2$ and $\beta_3$, we obtain for the constant-density ansatz (i.e. constant $E_C^0$),

$$\Delta E_C = -190 \text{ MeV} \frac{\Delta \langle r^2 \rangle}{\langle r^2 \rangle} - 0.42 \text{ MeV} \frac{\Delta \beta_3^2}{\beta_3^2}. \quad (23)$$
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 \(\Delta Q_0\) 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 \[\Delta \langle r^2 \rangle = \Delta \langle r^2 \rangle_{\text{sph}} + \Delta \langle r^2 \rangle_{\text{def}}. \tag{24}\]

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 \[\Delta \langle r^2 \rangle = \Delta \langle r^2 \rangle_{\text{sph}} + \frac{5}{4\pi} \langle r^2 \rangle_{\text{sph}} \Delta \beta^2 + \Delta \beta^2 + \ldots \tag{25}\]

where \(\langle r^2 \rangle_{\text{sph}}\) 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 \(^{229}\text{Th}\) isomers is less than a few parts per thousand, while the calculations of \[18\] imply a fractional volume change of about \(5 \times 10^{-4}\). 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 \(\alpha\), 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 \(\alpha\). Therefore, the six-orders-of-magnitude improvement in the sensitivity to \(\alpha\) 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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