Proposed Experimental Method to Determine α Sensitivity of Splitting between Ground and 7.6 eV Isomeric States in ^{229}Th

J. C. Berengut,[1] V. A. Dzuba,[1] V. V. Flambaum,[1] and S. G. Porsev[1,2]
[1]School of Physics, University of New South Wales, Sydney 2052, Australia
[2]Petersburg Nuclear Physics Institute, Gatchina, 188300, Russia
(Received 11 March 2009; published 29 May 2009)

The 7.6 eV electromagnetic transition between the nearly degenerate ground state and first excited state in the ^{229}Th nucleus may be very sensitive to potential changes in the fine-structure constant, $\alpha = e^2/\hbar c$. However, the sensitivity is not known, and nuclear calculations are currently unable to determine it. We propose measurements of the differences of atomic transition frequencies between thorium atoms (or ions) with the nucleus in the ground state and in the first excited (isomeric) state. This will enable extraction of the change in nuclear charge radius and electric-quadrupole moment between the isomers, and hence the α dependence of the isomeric transition frequency with reasonable accuracy.

DOI: 10.1103/PhysRevLett.102.210801 PACS numbers: 06.30.Ft, 21.10.Ft, 27.90.+b, 31.30.Gs

The isotope ^{229}Th has the lowest known excited state in nuclei; recent measurements show that the $3/2^+$ state lies just 7.6 eV above the $5/2^+$ ground state [1]. The width of this level is estimated to be about 10^{-4} Hz [2], which may explain why it is so hard to find the direct radiation of this very weak transition. Nevertheless, the frequency is within the range of lasers, and it has been proposed to use this narrow nuclear transition as a possible reference for an optical clock of very high accuracy [3]. Additionally, this transition could be a sensitive probe of possible variation of fundamental constants [4] because the near degeneracy of these isomers is a result of cancellation between very large energy contributions (order of MeV). Since these contributions would have different dependences on fundamental constants, any variation would be enhanced in the transition frequency. In Ref. [4], the relative effects of variation of α and the dimensionless strong interaction parameter $m_q/\Lambda_{\mathrm{QCD}}$ were estimated to be enhanced by 5 orders of magnitude.

An enhancement to the α sensitivity of this magnitude would have very important consequences for laboratory searches of α variation. Because the isomeric ^{229}Th resonance has a narrow linewidth and an extraordinary insensitivity to external perturbations, an optical clock utilizing this reference may have very high accuracy and high immunity from systematic frequency shifts [3]. By comparing this "nuclear clock" frequency with that of any other narrow optical or microwave transition (e.g., the Cs or Hg^+ frequency standards), one can test the variation of fundamental constants. Coupled with the enhancement in sensitivity, such a setup would be the most sensitive laboratory probe of α variation to date, possibly gaining several orders-of-magnitude improvement over the current limits of $\dot{\alpha}/\alpha = (-1.6 \pm 2.3) \times 10^{-17}$ yr^{-1} [5].

The sensitivity of the transition frequency to variation of α can be expressed as

$$\delta\omega = \Delta V_C \frac{\delta\alpha}{\alpha}, \qquad \frac{\delta\omega}{\omega} = K\frac{\delta\alpha}{\alpha}, \tag{1}$$

where ΔV_C is the difference in Coulomb energies between the two isomers, and K is the enhancement factor: $K = \Delta V_C/\omega$. Since the Coulomb energy of this nucleus is of order 10^9 eV, even a relatively small variation in V_C could produce a large enhancement. For $\Delta V_C = 100$ keV and $\delta\alpha/\alpha = 10^{-16}$, $\delta\omega = 10^{-11}$ eV $= 2.4 \times 10^3$ Hz, which is 4 orders of magnitude larger than the limits placed on shifts in atomic transitions in Ref. [5].

However, different nuclear calculations give wildly different values for ΔV_C. References [6,7] claim that both isomers have identical deformations and therefore the same Coulomb energies to within roughly 30 keV (corresponding to $K \lesssim 4000$). Reference [8] gives a value of 30 keV, while the calculations of [9] give values in the range -300 keV $< \Delta V_C < 450$ keV, depending on particulars of the model used. Lastly, Ref. [10] uses Nilsson wave functions to show that the value of ΔV_C as a function of deformation changes from 1.5 MeV at zero deformation down to -0.5 MeV at $\delta = 0.3$. Reference [10] concludes that a very small value of the Coulomb energy shift seems improbable.

In this Letter we propose a different method for extracting sensitivity to α variation using direct laboratory measurements of the change in nuclear mean-square charge radius, $\Delta\langle r^2\rangle$, and electric-quadrupole moment, ΔQ_0, between the isomer and the ground-state nucleus. We present a simple geometric model of the nucleus to relate the observable nuclear parameters to ΔV_C and hence K. We show that this model is self-consistent by comparing to the nuclear calculations of Ref. [9].

Once the change in Coulomb energy has been measured, the change in nuclear energy ΔE_N will be known also, since they almost cancel for this transition: $\Delta E_N + \Delta V_C = 7.6$ eV. This change in nuclear energy can be interpreted in

terms of variation of the dimensionless ratio $m_q/\Lambda_{\rm QCD}$, where m_q is the light quark mass and $\Lambda_{\rm QCD}$ is the pole in the running strong coupling constant. Variation of this ratio will also be enhanced in the ^{229}Th transition [4]; calculations may be found in [8,11].

The change in mean-square nuclear radius can be extracted using the isomeric field shift for an atomic transition. In principle, any transition in any ^{229}Th ion or the neutral atom can be used. There are two approaches. The first is entirely empirical: by combining the measurements of isomeric shifts and isotopic shifts for the same transition, one can extract the ratio of $\Delta\langle r^2\rangle$ for the isomer to the isotopic change in mean-square radius. The second approach does not require the additional measurement of isotope shift, but it does require high-precision atomic calculations.

To extract the change in nuclear quadrupole moment, the hyperfine structure may be used. The hyperfine-structure constant B, which can be determined experimentally, is proportional to the quadrupole moment Q. Therefore one must measure B for both the ground state and isomeric ^{229}Th. The value of Q for the ground state is known to within 20%; better accuracy can be obtained using the calculations presented here.

The radiative lifetime of the metastable ^{229m}Th nucleus is estimated to be a few hours [2]; however, this may be reduced if the energy of the excited state exceeds the ionization potential since an electron autoionization channel may open. The successive ionization energies of thorium ions are [12] 6.3 eV (Th I), 11.9 eV (Th II), 20.0 eV (Th III), 28.8 eV (Th IV). Therefore, the atomic experiments are likely to be easier for ionized thorium since the ionization energies exceed the excitation energy. In fact, Th IV may be the best choice since it is alkali-like and is amenable to laser cooling and trapping [3]. This ion has the additional advantage that calculations are likely to be more accurate, although we stress that we can obtain reasonable accuracy with any ion that experimentalists may find convenient.

We use a simple geometric model to relate the Coulomb energy of a nucleus to the experimentally observable mean-square charge radius and intrinsic electric-quadrupole moment, defined as

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

$$Q_0 = \int r^2[3\cos^2(\theta) - 1]\rho(r)d^3r, \tag{3}$$

where $\rho(r)$ is the electric charge density normalized to unity. The intrinsic quadrupole moment is related to the laboratory quadrupole moment of the ground rotational mode by (see, e.g., [13])

$$Q_{\rm lab} = ZQ_0\frac{I(2I-1)}{(I+1)(2I+3)}. \tag{4}$$

TABLE I. The values of $r_{\rm rms}$, Q_0, $\Delta r_{\rm rms}$, ΔQ_0, and V_C are reproduced from Ref. [9] and used to calculate the value of ΔV_C shown in the last line using our simple geometrical model. SkM* and SIII refer to two different energy functionals, while HF and HFB refer to Hartree-Fock and Hartree-Fock-Bogoliubov (the latter includes pairing correlations); for details see Ref. [9].

	SkM*		SIII	
	HF	HFB	HF	HFB
$r_{\rm rms}$ (fm)[a]	5.7180	5.7078	5.7817	5.7769
Q_0 (fm^2)[a]	9.5461	9.3717	9.3542	9.1643
$\Delta r_{\rm rms}$ (fm)[a]	−0.0038	0.0039	0.0000	−0.0005
ΔQ_0 (fm^2)[a]	−0.1824	0.2756	−0.0339	−0.0495
V_C (MeV)[b]	924	925	912	912
ΔV_C (MeV)[b]	0.4510	−0.3070	−0.0980	0.0010
ΔV_C (MeV)	0.4190	−0.3270	−0.0360	0.0290

[a]From Ref. [9], Table II.
[b]From Ref. [9], Table I.

We wish to express the Coulomb energy in terms of these measurable quantities. Assume that both the ground-state nucleus and the lowest-energy isomer are uniform, hard-edged, prolate ellipsoids. Using formulas presented in [14] we find

$$V_C = \left(\frac{3}{5}\right)^{3/2}\frac{(Ze)^2}{\langle r^2\rangle^{1/2}}\left(1 + \frac{3}{40}\frac{Q_0^2}{\langle r^2\rangle^2} - \frac{1}{56}\frac{Q_0^3}{\langle r^2\rangle^3} + \ldots\right). \tag{5}$$

With this equation we can extract ΔV_C if we know $\Delta\langle r^2\rangle$ and ΔQ_0 between the ^{229}Th isomers. Note that V_C and ΔV_C are vastly more sensitive to changes in $\langle r^2\rangle$ than Q_0.

To estimate the effect of skin thickness, we use a spherical Fermi distribution model $\rho(r) = \rho_0(1 + \exp[(r - C)/z])^{-1}$. The Coulomb energy is

$$V_C = \left(\frac{3}{5}\right)^{3/2}\frac{(Ze)^2}{\langle r^2\rangle^{1/2}}\left(1 + 8.379\frac{z^3}{\langle r^2\rangle^{3/2}} + \ldots\right), \tag{6}$$

and one sees that V_C is not sensitive to the skin thickness parameter z.

With the current data for ^{229}Th, $r_{\rm rms} = 5.6807 \pm 0.0509$ fm [15] and $Q_{\rm lab} = 4.3 \pm 0.9$ eb [16], we obtain $Q_0 = 13.4$ and Coulomb energy $V_C = 967$ MeV. We es-

TABLE II. Calculated energies and field-shift constants of transitions in Th IV. The last column shows expected "order of magnitude" isomeric shifts in ^{229}Th, assuming $|\Delta r_{\rm rms}| = 0.004$ fm; however, the actual shift could differ by an order of magnitude. All transitions are to the $5f_{5/2}$ ground state.

	ω (cm^{-1})			
Level	Experimental	Calculated	F (GHz/fm^2)	$\lvert\delta\nu^m\rvert$(GHz)
$5f_{7/2}$	4325	4899	2(2)	0.09
$6d_{3/2}$	9193	11 721	33(8)	1.40
$6d_{5/2}$	14 486	17 534	35(8)	1.50
$7s_{1/2}$	23 131	24 740	146(4)	6.30
$7p_{1/2}$	60 239	63 051	57(3)	2.50
$7p_{3/2}$	73 056	76 319	49(2)	2.10

TABLE III. Calculated energies, ω (cm^{-1}), field-shift constants, F ($\mathrm{GHz/fm}^2$), and isotope shifts, $\delta\nu^{232,230}$ (10^{-3} cm^{-1}), of some transitions in Th III. All transitions are to the $5f6d\,^3\mathrm{H}_4^0$ ground state. Note that, while we believe the $6d^2\,^3\mathrm{F}_3$, $6d^2\,^3\mathrm{F}_4$, and $6d7s\,^3\mathrm{D}_3$ transitions are accurate, the others are estimates only.

Level		ω (cm^{-1})			$\delta\nu^{232,230}$
Term	J	Experimental	Calculated	F	Calculated[a]
$6d^2\,^3\mathrm{F}$	3	4056	4023	24	165
$6d^2\,^3\mathrm{F}$	4	6538	6795	22	147
$6d7s\,^3\mathrm{D}$	3	9954	9204	118	804
$6d^2\,^1\mathrm{G}$	4	10 543	11 051	8	56
$5f^2\,^3\mathrm{H}$	4	15 149	13 358	−11	−77
$5f^2\,^3\mathrm{H}$	5	17 887	16 068	−20	−136
$5f^2\,^3\mathrm{F}$	3	20 840	19 080	−18	−122
$5f^2\,^3\mathrm{F}$	4	21 784	20 366	−15	−101
$5f^2\,^1\mathrm{G}$	4	25 972	25 269	10	−66
$5f7p$ $(\frac{5}{2},\frac{1}{2})$	3	33 562	33 402	13	92
$5f7p$ $(\frac{7}{2},\frac{1}{2})$	3	38 432	38 617	15	101

[a]$\Delta\langle r^2\rangle = 0.205$ fm^2, from Ref. [15]

timate $z = 0.5$ fm. In this case the change in Coulomb energy can be expressed

$$\frac{\Delta V_C}{(\mathrm{MeV})} = -506\frac{\Delta\langle r^2\rangle}{\langle r^2\rangle} + 23\frac{\Delta Q_0}{Q_0} + 17\frac{\Delta z}{z} \qquad (7)$$

from which the sensitivity of the transition to α variation is easily deduced. Note that the contribution of Δz is small.

As a consistency check, we have recalculated ΔV_C using the values of r_{rms}, Δr_{rms}, Q_0, and ΔQ_0 calculated in Ref. [9]; this is shown in Table I. That we are able to reproduce the results of Ref. [9] shows the validity of the geometrical model. The differences seen in the SIII entries of Table I (last two columns) are probably due to lack of numerical precision. If measurements of Δr_{rms} and ΔQ_0 are made accurately, the model should suffice even when the measurable nuclear parameters are small.

Mean-square radius.—We have shown that $\Delta\langle r^2\rangle$ is the most important quantity for determining ΔV_C and hence sensitivity to α variation. $\Delta\langle r^2\rangle$ can be extracted from the isomeric shift of any atomic transition, obtained by comparing ^{229}Th and ^{229m}Th. This is similar to the usual isotope shift.

The shift in energy of any transition in an isotope with mass number A' with respect to an isotope with mass number A can be expressed as

$$\Delta\nu^{A',A} = (k_{\mathrm{NMS}} + k_{\mathrm{SMS}})\left(\frac{1}{A'} - \frac{1}{A}\right) + F\Delta\langle r^2\rangle^{A',A}. \qquad (8)$$

Here the first term is the "mass shift" due to the finite mass of the nucleus and the second term is the "volume" or "field" shift due to the finite size of the nuclear charge distribution (see, e.g., [17]). In the case of the isomeric shift that we are interested in, the mass shift vanishes since isomers have equal mass. Thus in order to extract $\Delta\langle r^2\rangle$ from a measurement of isomeric shift $\Delta\nu^m$ for an atomic transition, we need simply divide by the field-shift constant F:

$$\Delta\nu^m = F\Delta\langle r^2\rangle. \qquad (9)$$

These may be calculated or extracted from known isotope shifts.

In Tables II, III, and IV, we present calculated field-shift constants for transitions in several ions of Th. In Table II we have included an estimated size of the isomeric shift, $\delta\nu^m$, assuming that $\Delta r_{\mathrm{rms}} = 0.004$ fm, which is the magnitude of the largest shifts in [9] (from the SkM* nuclear energy functionals).

We calculate the field-shift constants F using methods developed in previous works [17]. Briefly, we perform an energy calculation several times, modifying the nuclear radius in our codes. F is extracted from the gradient: $F = dE/d\langle r^2\rangle$ at $r_{\mathrm{rms}} = 5.6807$ fm. The energy calculations are performed using a combination of second-order many-body perturbation theory and configuration interaction

TABLE IV. Calculated energies, ω (cm^{-1}), field-shift constants, F ($\mathrm{GHz/fm}^2$), and isotope shifts, $\delta\nu^{232,230}$ (10^{-3} cm^{-1}), of some transitions in Th II. All transitions are to the $6d^27s$ $J = 3/2$ ground state.

Level			ω (cm^{-1})			$\delta\nu^{232,230}$		
Configuration		J	Experimental	Calculated	F	Experimental	Calculated[a]	Calculated[b]
$5f7s^2$	$^2\mathrm{F}^0$	5/2	4490	4856	4	54	47	43
$5f6d7s$	$^4\mathrm{F}^0$	3/2	6691	7487	−53	−362	−401	−362
$5f6d7s$	$^4\mathrm{F}^0$	5/2	7331	8325	−53	−365	−405	−365
$5f6d7s$	$^4\mathrm{G}^0$	5/2	9585	10 045	−55	−375	−406	−366
$5f6d7s$	$^4\mathrm{H}^0$	5/2	10 673	12 168	−53	−361	−406	−367
$5f6d7s$	$^2\mathrm{D}^0$	3/2	11 576	13 054	−54	−367	−408	−368
$5f6d7s$	$^4\mathrm{D}^0$	1/2	11 725	12 897	−67	−456	−460	−415
$5f6d7s$	$^2\mathrm{F}^0$	5/2	12 472	14 564	−58	−399	−463	−418
$5f6d7s$	$^4\mathrm{F}^0$	3/2	12 902	14 233	−58	−395	−444	−400
$5f6d7s$	$^4\mathrm{G}^0$	1/2	14 102	15 853	−79	−539	−610	−550

[a]$\Delta\langle r^2\rangle = 0.205$ fm^2, from Ref. [15]
[b]$\Delta\langle r^2\rangle = 0.185$ fm^2, best fit value.

(CI + MBPT, Refs. [18,19]) which takes into account dominating relativistic and correlation effects. In the single-valence-electron case, Th IV, the technique reduces to the correlation-potential method [20] in which Brueckner orbitals are calculated for states of the valence electron. The accuracy of these calculations is high, as illustrated in Tables II, III, and IV.

For Th II there are experimental isotope shifts available [21], and we compare them with our calculations in Table IV. The second-last column is a calculation with $\Delta\langle r^2\rangle^{232,230} = 0.205(30)$ fm^2 [15], while the last column gives values of the isotope shift calculated with $\Delta\langle r^2\rangle^{232,230} = 0.185$; this is the value that gives the best agreement between our calculated isotope shifts and the experimental data. Note that we have ignored the mass shift; it is negligible in such a heavy element.

The field-shift constant is generally larger for transitions involving a change in the s-wave configuration, e.g., $5f_{5/2} \rightarrow 7s_{1/2}$ transition in Th IV and the $5f6d\,^3\mathrm{H}_4^o \rightarrow 6d7s\,^3\mathrm{D}_3$ transition in Th III. Measurement of the isomeric shift may be easier for these cases. However, if there are good reasons to use transitions with smaller shifts (e.g., the higher-energy transitions in Th III), then we recommend the experimentalists contact us for more precise values of the constants. Again we stress that these constants may be extracted from measured isotope shifts with accuracy only limited by knowledge of the isotopic change in mean-square radius, $\Delta\langle r^2\rangle^{A',A}$.

Electric quadrupole.—Although we have shown (7) that the change in Coulomb energy of the 7.6 eV transition in the ^{229}Th nucleus is far more sensitive to $\langle r^2\rangle$ than Q_0, ΔQ_0 could still be important if $\Delta\langle r^2\rangle$ is found to be very small. Fortunately ΔQ_0 can be extracted from measurements of the hyperfine structure of the isomers by using (4) and noting that Q_{lab} is proportional to the electric-quadrupole hyperfine-structure constants B. Since the electric-quadrupole moment of the ground-state ^{229}Th nucleus is known to about 20% accuracy [$Q_{\mathrm{lab}} = 4.3(9)$ eb [16]], ΔQ_0 can be extracted to this level of accuracy, and no atomic calculations are needed for interpretation of the results.

If better than 20% accuracy is required, the values of Q_{lab} can be found by comparison of the calculated and measured B. Calculations with this level of accuracy for many-valence-electron ions are difficult, but can be performed if required. In this Letter we present calculations of B for the single-valence-electron ion Th IV. The constant B for a particular valence state v is found as a matrix element

$$B_v = A\langle\psi_v^{\mathrm{Br}}\|\hat{F} + \delta V\|\psi_v^{\mathrm{Br}}\rangle, \tag{10}$$

where A is a numerical constant, ψ_v^{Br} is the Brueckner orbital for the valence state v, $\hat{F}$ is the operator of the nuclear electric-quadrupole moment, and δV is the correction to the atomic self-consistent potential due to the effect of nuclear quadrupole electric field on atomic electrons. The same Brueckner orbitals are used as in the previous section. The results are presented in Table V: accuracy is expected to be at the level of a few percent.

TABLE V. Calculated electric-quadrupole hyperfine-structure constants B for some low energy states of Th IV. In the last column, the nuclear electric-quadrupole moment Q is taken to be 4.3 b.

Level	B (MHz)	
$5f_{5/2}$	$740Q$	3180
$5f_{7/2}$	$860Q$	3700
$6d_{3/2}$	$690Q$	2970
$6d_{5/2}$	$860Q$	3700
$7p_{3/2}$	$1810Q$	7790

This work is supported by the Australian Research Council, Marsden Grant, and the NCI National Facility.

[1] 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).
[2] E. V. Tkalya, A. N. Zherikhin, and V. I. Zhudov, Phys. Rev. C **61**, 064308 (2000).
[3] E. Peik and Chr. Tamm, Europhys. Lett. **61**, 181 (2003).
[4] V. V. Flambaum, Phys. Rev. Lett. **97**, 092502 (2006).
[5] T. Rosenband *et al.*, Science **319**, 1808 (2008).
[6] A. C. Hayes and J. L. Friar, Phys. Lett. B **650**, 229 (2007).
[7] A. C. Hayes, J. L. Friar, and P. Möller, Phys. Rev. C **78**, 024311 (2008).
[8] X.-t. He and Z.-z. Ren, J. Phys. G **35**, 035106 (2008).
[9] E. Litvinova, H. Feldmeier, J. Dobaczewski, and V. V. Flambaum, arXiv:0901.1240 [Phys. Rev. C (to be published)].
[10] V. V. Flambaum, N. Auerbach, and V. F. Dmitriev, Europhys. Lett. **85**, 50 005 (2009).
[11] V. V. Flambaum and R. B. Wiringa, Phys. Rev. C **79**, 034302 (2009).
[12] *CRC Handbook of Chemistry and Physics (Internet Version 2009)*, edited by D. R. Lide (CRC Press/Taylor and Francis, Boca Raton, FL, 2009), 89th ed.
[13] E. Segrè, *Nuclei and Particles* (Benjamin-Cummings, Reading, MA, 1977).
[14] R. W. Hasse and W. D. Myers, *Geometrical Relationships of Macroscopic Nuclear Physics* (Springer-Verlag, Heidelberg, 1988).
[15] I. Angeli, At. Data Nucl. Data Tables **87**, 185 (2004).
[16] N. J. Stone, At. Data Nucl. Data Tables **90**, 75 (2005).
[17] J. C. Berengut, V. A. Dzuba, and V. V. Flambaum, Phys. Rev. A **68**, 022502 (2003).
[18] V. A. Dzuba, V. V. Flambaum, and M. G. Kozlov, Phys. Rev. A **54**, 3948 (1996).
[19] V. A. Dzuba and W. R. Johnson, Phys. Rev. A **57**, 2459 (1998).
[20] V. A. Dzuba, V. V. Flambaum, P. G. Silvestrov, and O. Sushkov, J. Phys. B **20**, 1399 (1987).
[21] J. Blaise and J.-F. Wyart, *Selected Constants: Energy Levels and Atomic Spectra of Actinides* (Tables Internationales de Constantes, Paris, 1992).