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Fine-structure constant sensitivity of the Th-229 nuclear clock transition

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Nuclear laser spectroscopy at the 10^{-12} precision level (Nature 633.8028 (2024): 63–70) determined the fractional change in the nuclear quadrupole moment of ^{229}Th upon excitation, $\Delta Q_0/Q_0 = 1.791(2)\%$. Such high-accuracy nuclear parameters enable stringent tests and refinement of ^{229}Th nuclear models. Using a semi-classical prolate-spheroid model, we quantify the transition frequency's sensitivity to fine-structure constant variations as $K = 5900(2300)$, with uncertainty dominated by the measured charge-radius change $\Delta\langle r^2\rangle$. This supports the predicted higher α-sensitivity of nuclear clocks over atomic clocks, important for new-physics searches. We find ΔQ_0 strongly dependent on nuclear volume, challenging the constant-volume approximation. The deviation between measured and predicted $\Delta Q_0/Q_0$ underlines the need for improved modeling and measurement of additional nuclear parameters. We explicitly assess the octupole contribution to α-sensitivity.

The ^{229}Th nucleus features a first excited metastable state, ^{229m}Th, with an unusually low excitation energy of 8.4 eV. This nucleus represents a unique opportunity to build a high-precision nuclear clock (see review[1] and references therein), as the transition is narrow, insensitive to external perturbations, and within the range of tabletop narrow line-width lasers.

A nuclear clock based on trapped Th^{3+} ions promises low systematic shifts, with an estimated total fractional inaccuracy of 10^{-19} [2,3]. Another unique opportunity enabled by the Th isomer is the development of a solid-state clock based on ^{229}Th doped into a VUV-transparent crystal, where atoms are confined to a lattice in a space that is much smaller than the excitation wavelength (Lamb-Dicke regime)[4,5].

Resonant laser excitation of the isomer was first demonstrated in Th-doped CaF_2[6] and then in Th-doped $LiSrAlF_6$ crystals[7] using tabletop broadband tunable laser systems. Soon after, a frequency-stabilized VUV frequency comb was used to improve frequency resolution by ~10^6 and directly resolve the nuclear quadrupole splitting in Th-doped CaF_2[8]. The nuclear transition frequency was determined to be 2 020 407 384 335(2) kHz.

A variation of fundamental constants is predicted by many theories beyond the standard model of particle physics[9,10]. Moreover, ultralight dark matter may lead to oscillations of fundamental constants[11–15], and searches for such variation present a direct dark matter detection opportunity. If fundamental constants of nature change with space or time, so will atomic and nuclear energy levels, and consequently the associated clock frequencies. The amplitude of such changes, however, strongly depends on the particular clock transition under investigation[16–21]. Any spacetime-dependent variation of clock frequency ratios would unambiguously point to new physics[22,23].

Highly enhanced sensitivity of the ^{229}Th nuclear clock transition frequency to variation of the fine-structure constant α and the strong interaction coupling constant α_S was predicted by Flambaum[24]. Such an enhancement leads to increased discovery potential of any new physics that manifests as a variation of the fundamental constants[14,19].

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However, the nuclear clock enhancement factor K for the variation of α defined as $\frac{\delta\nu}{\nu} = K\frac{\delta\alpha}{\alpha}$ has yet to be precisely determined, where ν is the clock transition frequency.

To determine $K = \Delta E_C/E$, where $E = h\nu$ is the isomer excitation energy, we need to know the difference in the Coulomb energy of the isomer and the ground state ΔE_C[24]. Since the Coulomb energies of both nuclear states are on the order of GeV, and ΔE_C is expected to be on the order of MeV, direct nuclear computation of the isomer and ground state energy is not sufficiently accurate for the determination of K (see, for example[25]). Therefore, a method to determine ΔE_C from measured nuclear properties of the ground and isomeric state using a geometric model, the liquid drop model, was proposed[26]. The validity of this approach stems from the fact that the change in fine-structure constant modifies the total coulomb energy of the nucleus slightly. The change in coulomb energy is not enough to affect nuclear structure and to compute the coulomb energy of the nucleus on the MeV level precision a liquid drop model suffices[27]. Currently, it is the only existing model relating experimental measurements and model parameters, as other microscopic models rely on intense numerical calculations to relate model parameters to experimental observables which complicates direct comparison. The geometrical model hinges on a sufficiently precise measurement of the ratio of quadrupole moments of the two states. Hartree-Fock-Bogolubov nuclear many-body calculations[25] were used for validating the geometric model[26]. More elaborate collective quadrupole-octupole models were recently developed[28,29], as there is indication for ^{229}Th being on the edge of the octupole deformation region of nuclei and possibly displaying octupole vibrational character[30]. However, the new models could not yet provide an accurate determination of ΔE_C.

We use the geometric model[26] in combination with the new precision laser spectroscopy performed in[8] to extract the nuclear quadrupole splitting (see "methods"), and then determine the sensitivity of the nuclear clock to α-variation. Within this model, the spectroscopy data quantitatively confirms a non-zero K. The constant volume assumption, used in previous work[20], is found to be unreliable in extracting the mean-square charge radius. We also estimate the effect of including octupole deformation into the geometric model and find that it may have a significant effect on K, depending on the yet unknown octupole moment of ^{229}Th in ground and isomeric state. Therefore we emphasize the need for the development of advanced nuclear models relating experimental measurements to model parameters.

Results

Nuclear theory

Following[20,26,31], we describe the nucleus as a prolate spheroid with uniform charge density. Although ground and excited state have different charge density functions, we assume the uniformity is not affected[32,33]. If a generalized Fermi function or for example a folded Yukawa-plus-exponential potential is used for the charge density profile, the Coulomb energy is corrected with a factor $\propto \left(1+(\frac{z}{R})^2\right)$ for a spherical distribution where z is the skin thickness[34]. Although this factor might seem small, it needs to be tested for a prolate spheroid as well. This further refinement was tested for in[35] and found that for typical values ($z \approx 0.5$ fm), K is between $10^3 - 10^5$, in line with the following calculations. The nuclear quadrupole moment Q_0 and the mean-square charge radius $\langle r^2\rangle$ are expressed via semi-minor and semi-major axes a and c as

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

Very coarsely, upon nuclear excitation, the distribution of a valence neutron is modified and polarizes the proton distribution via the strong interaction, altering $\langle r^2\rangle$ as well as the nuclear deformation and in consequence Q_0. These two quantities, therefore, are essential for the determination of the Coulomb energies of the nuclear ground and isomeric state. The Coulomb energy (E_C)[36] is given by Eq. (5.11) in ref. 37,

$$E_C = \frac{3q_e^2Z^2}{5R_0}\frac{(1-e^2)^{1/3}}{2e}\ln\frac{1+e}{1-e}, \tag{2}$$

where $e^2 = 1 - (a^2)/(c^2)$ is the eccentricity, Z the proton number, and $R_0^3 = a^2c$ is the equivalent sharp spherical radius, see Eq. (6). The difference of the Coulomb energy (ΔE_C) of the isomer and the ground state is computed as

$$\Delta E_C = \langle r^2\rangle\frac{\partial E_C}{\partial\langle r^2\rangle}\frac{\Delta\langle r^2\rangle}{\langle r^2\rangle} + Q_0\frac{\partial E_C}{\partial Q_0}\frac{\Delta Q_0}{Q_0} \tag{3}$$

$$= -485\,\text{MeV}\frac{\Delta\langle r^2\rangle}{\langle r^2\rangle} + 11.3\,\text{MeV}\left(\frac{Q_0^{\text{m}}}{Q_0} - 1\right), \tag{4}$$

where we used $\langle r^2\rangle = 5.756(14)$ fm^2 [38], the spectroscopic quadrupole moment $Q_{\text{lab}} = 3.11(2)$ eb[39] for the ground state and $\Delta Q_0 = Q_0^{\text{m}} - Q_0$. The spectroscopic quadrupole moment in the laboratory frame (Q_{lab}), can be related to the quadrupole moment in the nuclear frame (see supplemental). Q_{lab} was recently extracted by modeling experimental measurements of the electronic hyperfine-structure[40] using the coupled cluster method with single, double, and triple excitations for both valence and core electrons, improving the accuracy[39]. This newer value accounts for a small difference in the second coefficient 11.3 MeV compared to ref. 20,31. We express Eq. (4) in terms of the difference of $\langle r^2\rangle$ and the ratio of quadrupole moments as these are the quantities which are determined from experiments.

The experimental results used to compute the ΔE_C from Eq. (4), can be derived as follows. The ratio, ζ, of the isomer and isotope shifts

$$\zeta = \frac{\langle r^2_{229m}\rangle - \langle r^2_{229}\rangle}{\langle r^2_{232}\rangle - \langle r^2_{229}\rangle} = 0.035(4) \tag{5}$$

was measured in ref. 31 for two transitions in Th^{2+}. Using this ratio, we can derive $\Delta\langle r^2\rangle$. New isotope shift measurements and more advanced theoretical calculations[41] yielded an improved value $\langle r^2_{232}\rangle - \langle r^2_{229}\rangle = 0.299(15)$ fm^2, reducing the uncertainty in this value from 15% to 5%. As a result, the uncertainty in the $\Delta\langle r^2_{229}\rangle = \langle r^2_{229m}\rangle - \langle r^2_{229}\rangle = 0.0105(13)$ fm^2 was reduced from 17% to 12%[41]. We note that this improvement in the accuracy is important for the determination of the α-sensitivity K due to a strong cancellation of the $\langle r^2\rangle$ and Q_0 contribution, as discussed below.

Using the spectroscopic quadrupole moment (in the lab frame, see "methods") for the ground state $Q_{\text{lab}} = 3.11(2)$ eb[39] and the ratio of the intrinsic quadrupole moments $Q_0^{\text{m}}/Q_0 = 1.01791(2)$, we find $Q_0^{\text{m}} = 9.85(6)$ fm^2 and $Q_{\text{lab}}^{\text{m}} = 1.77(1)$ eb. Note that we follow the designations of [20] for Q_0, which differs from[31], where Q_0 was defined without the factor q_eZ. Using designations and units of[31], we get $Q_0^{\text{m}} = 8.86(6)\,e$b. The measured value[8] of $\Delta Q_0/Q_0 = Q_0^m/Q_0 - 1 = 0.01791(2)$ (see "methods") differs by a factor of 2.4 from the prediction of $\Delta Q_0/Q_0 = 0.0075(20)$ obtained assuming a constant nuclear volume for the ground and the isomeric state[20]. Using the predicted value in ref. 20, the authors arrived at a negative $K = -8200(2500)$[20]. To test the constant volume approximation, we computed the respective volumes using

$$V = \frac{4\pi}{3}R_0^3 = \frac{4\pi}{3}a^2c \tag{6}$$

and find a small but non-zero −0.055% difference, with the isomer volume being smaller. This change in the volume is well within the 0.8%

experimental uncertainty. However, it is interesting to note that volume changes due to the change in the rms radius and Q_0 contribute with opposite signs, +0.05% and −0.105%, respectively, leading to some cancellation of the overall effect. We tested the dependence of $\Delta Q_0/Q_0$ on the volume change and find that even a small −0.055% change leads to a difference by over a factor of two in the resulting $\Delta Q_0/Q_0$, making the constant volume approximation unreliable for extracting this quantity from the difference in the $\langle r^2\rangle$.

Substituting the experimental values into the expression for the Coulomb energy difference given by Eq. (4) gives

$$\begin{aligned}\Delta E_C &= -0.154(19)\,\mathrm{MeV} + 0.203(4)\,\mathrm{MeV}\\ &= 0.049(19)\,\mathrm{MeV}\end{aligned} \tag{7}$$

demonstrating strong cancellation between $\langle r^2\rangle$ and Q_0 terms, making the ΔE_C and K values extremely sensitive to $\Delta Q_0/Q_0$. The enhancement factor is

$$K = -18400(2300) + 24300(400) = 5900(2300), \tag{8}$$

where the uncertainty is overwhelmingly due to the uncertainty in $\Delta\langle r^2\rangle$. An improved measurement of ζ is needed for further improvement in the accuracy of K.

Such a high value of K drastically increases the sensitivity of a nuclear clock to dark matter and other related new physics searches in comparison with current optical atomic clocks. The highest sensitivity in atomic clocks is $K = -6$ (for ^{171}Yb E3 transitions[42]). Using a nuclear clock thus gives three orders of magnitude improvement for the same clock accuracy. This improvement translates into being able to probe dark matter with three orders of magnitude smaller couplings.

Effect of the octupole deformation

We point out that the discussion above does not include uncertainties connected with the present theoretical model, but only uncertainties of the experimentally determined quantities. In particular, a contribution of the currently unknown nuclear octupole deformation to the Coulomb energy E_C is not accounted for. In order to gauge the effect, we approximate the vibrational quadrupole-octupole character of the ^{229}Th nucleus[28,29] by a static, axially symmetric octupole deformation. This approach is purely demonstrative to qualitatively study the effect of the octupole deformation on the coulomb energy and therefore the fine-structure constant sensitivity. Combined with experimental observations of β_3, these models can be used to estimate K as the validity of using macroscopic geometric nuclear models to relate macroscopic characteristics (β_n, Q, r, ΔE_C) does not change. Following[20], the surface of the nucleus $r(\theta)$ is expanded as

$$r(\theta) = R_s\left[1 + \sum_{n=1}^{N}\left(\beta_n Y_n^0(\theta)\right)\right], \tag{9}$$

where β_n are deformation parameters, $Y_n^0(\theta)$ are spherical harmonics, and R_s is defined by normalizing the volume to that of the spherical nucleus with equivalent sharp spherical radius R_0. The normalization condition can be expressed as $\int\rho_q(\boldsymbol{r})d^3\boldsymbol{r} = 1$, where in the present approximation of constant charge density, $\rho_q = 1/V$ is the charge density divided by the total charge $q_e Z$ and $V = (4\pi/3)R_0^3$:

$$\frac{2\pi}{3}\int_0^{\pi} r^3(\theta)\,\sin(\theta)\,d\theta = V. \tag{10}$$

For a pear-shaped, axially symmetric nucleus with quadrupole and octupole deformation, $N = 3$. The coefficient β_1 is determined from the condition that the center of mass of the shape is at the origin of the coordinate system, $\int_V \boldsymbol{r} d^3\boldsymbol{r} = 0$[43]. For a pure quadrupole deformation (i.e., prolate spheroid), $N = 2$ and $\beta_1 = 0$. The nuclear properties are related to the β coefficients via

$$\langle r^2\rangle = \int r^2\rho_q(\boldsymbol{r})d^3\boldsymbol{r} \tag{11}$$

$$Q_0 = 2\int r^2 P_2(\cos\theta)\rho_q(\boldsymbol{r})d^3\boldsymbol{r}, \tag{12}$$

$$Q_{30} = 2\int r^3 P_3(\cos\theta)\rho_q(\boldsymbol{r})d^3\boldsymbol{r}, \tag{13}$$

where P_2 and P_3 are the Legendre polynomials, and Q_{30} is the intrinsic charge octupole moment[43].

The expression for the Coulomb energy was given in ref. 37, Eqs. (6.47),

$$E_C = \frac{3q_e^2Z^2}{5R_0}\left(1 - \frac{1}{4\pi}\beta_2^2 - \frac{5}{14\pi}\beta_3^2\right), \tag{14}$$

β_2 and β_3 are the quadrupole and octupole deformations, respectively. We keep the first in two terms in Eq. (6.47) to omit $O(\beta_n^3)$ terms, negligible at the present level of accuracy. We use a substitution of Eq. (6.51) to account for a representation of $R(\theta)$ with axially symmetric spherical harmonics instead of Legendre polynomials.

The change in the Coulomb energy was described in ref. 20 using the expression

$$\begin{aligned}\Delta E_C &= \frac{\partial E_C}{\partial\beta_2^2}\Delta\beta_2^2 + \frac{\partial E_C}{\partial\beta_3^2}\Delta\beta_3^2\\ &= \frac{3q_e^2Z^2}{5R_0}\left(-\frac{1}{4\pi}\Delta\beta_2^2 - \frac{5}{14\pi}\Delta\beta_3^2\right).\end{aligned} \tag{15}$$

However, this expression utilizes the constant volume approximation and assumes intrinsic axial symmetry, while R_0 depends on β_2 and β_3 via the volume normalization above. As in the ellipsoid model above, this leads to a wrong result for ΔE_C, when using the experimental values for $\langle r^2\rangle$ and Q_0.

To evaluate the effect of a possible octupole deformation described by the change in β_3 upon nuclear excitation, we calculate ΔE_C using experimentally available numbers and the geometric model described above. We use the definitions for $\langle r^2\rangle$ and Q_0 and the normalization condition above, the experimental values for $\langle r^2\rangle$ and Q_0, to calculate R_s, β_2, and R_0 for both ground and isomeric state.

First, we take $\beta_3 = 0$ for both the ground and isomeric state. The resulting values of $\beta_2 = 0.220$ and $\beta_2^{\mathrm{m}} = 0.223$ are consistent with ^{228}Th value from ref. 44. We compute $\Delta E_C = 0.052\,\mathrm{MeV}$ giving $K = 6300(2300)$ using Eq. (14), in agreement with the result obtained for the ellipsoid model (we note that Eq. (14) is approximate). Then, we repeat the same computation but use $\beta_3 = \beta_3^{\mathrm{m}} = 0.115$ from ref. 28, which yields essentially the same result for K, as expected. However, when we vary β_3 differentially between the ground and excited state, while keeping all other parameters constant, we find that 1%, 3%, and 5% differential change in β_3 lead to ΔK = 2850, ΔK = 8600, and ΔK = 14500, respectively. A larger β_3 of the isomer yields a positive change in K and inversely, a smaller β_3 yields a negative change in K, possibly jeopardizing the fine-structure constant sensitivity. However, it was concluded in[35] that this is unlikely.

For reference, the measured change in Q_0, and correspondingly β_2, is 1.8%. This computation shows that even 1% variation of the octupole deformation changes K by more than 1σ. Therefore, it is important to estimate the sign and constrain the magnitude of the octupole deformation change between the ground and isomeric state and model the effect on the change in intrinsic quadrupole moment when including the octupole deformation, for which more sophisticated nuclear models are needed.

Table 1 | Observed lines and their interpretations according to[8], together with calculated relative intensities W

j	ν_j [THz]	m_g	m_e	$W(E_2 - E_1)$
1	2020.407283847(4)	± 3/2	± 1/2	0.184
2	2020.407298727(4)	± 5/2	± 3/2	0.328
3	2020.407446895(4)	± 1/2	± 1/2	0.310
4	2020.407530918(4)	± 3/2	± 3/2	0.149
5	2020.40769398(2)	± 1/2	± 3/2	0.0233
6	2020.407051655	± 5/2	± 1/2	0.0058

Line 6 is asymptotically forbidden; it becomes accessible through the $\eta \neq 0$ EFG asymmetry but is not experimentally observed due to the weak intensity. The observed line intensities qualitatively agree with theoretical predictions and more precise measurements will be carried out in the future.

We can estimate the approximate size of the intrinsic charge octupole moment in ^{229}Th ground state and isomer. Using the β_2 value obtained above and β_3 within the range used in[28], 0.11 to 0.145, we get $Q_{30} = 35 - 44$ fm^3 (Eq. (13)). This is consistent with estimates[44] for the octupole moment of ^{228}Th. Note that[44] gives the values of Zq_eQ. E3 matrix elements of nuclear Coulomb excitation should be measured to determine the octupole moment, as has been demonstrated on other isotopes[45,46]. We also note that even higher moments might contribute to the nuclear Coulomb energies. Octupole deformation in ^{229}Th allows for the search of permanent electric-dipole moments[44,47].

Discussion

The precise measurement of the change in nuclear quadrupole moment $\Delta Q_0/Q_0$[8] between the ^{229}Th ground and isomeric state together with previous measurements of the mean-square charge radius $\langle r^2 \rangle$ allow us to extract the change in Coulomb contribution ΔE_C to the nuclear energies. We quantify the sensitivity of the nuclear transition to changes of the fine-structure constant α, within the presented model, with the factor $K = 5900(2300)$ which is for the first time inconsistent with zero by 2σ. This result shows the potential for using the nuclear clock for measurements in fundamental physics. Future theoretical and experimental work will focus on developing and verifying a model that bridges experimental observables and the intrinsic quadrupole moment in a microscopic description of the ^{229}Th isomeric excitation, taking into account the quadrupole-octupole deformed shell, as we have shown that small octupole deformations strongly affect the fine-structure sensitivity. In addition, an improved measurement of the ratio ζ of the isomer and isotope shifts is required. The measured quantitative deviation from the constant volume approximation illustrates the power of the emerging precision laser spectroscopy on ^{229}Th for testing fundamental models in nuclear physics. Recently we learned about a new work that uses a theoretical d-wave halo model to estimate $K \approx 10^4$[35].

Methods

Quadrupole structure

In the nuclear laser spectroscopy performed in[8], samples containing ^{229}Th nuclei doped in a CaF_2 matrix were used[48–50]. The interaction of the nuclear quadrupole moment (Q_0) in the lab frame (Q_{lab}) with the crystal electric field gradient (EFG), $V_{ij} = \partial^2 V/(\partial x_i \partial x_j)$, leads to the emergence of a nuclear quadrupole splitting[5]. We note that any octupole moment interaction with the EFG that would show a measurable influence on the quadrupole level structure would need to be a *CP* violating effect[51].

We first describe the interaction Hamiltonian (see supplementary information) between the EFG V_{ij} and the spectroscopic nuclear quadrupole moment Q_{lab} and derive the energy levels and eigenstates. Here we define axes of the EFG such that V_{ij} is diagonal, $|V_{zz}|$ is the maximum value of the EFG and $\eta = (V_{xx} - V_{yy})/(V_{zz})$ is the asymmetry of the EFG. We then calculate the energies of the quadrupole states and excitation probabilities $W(E_2 - E_1)$ by calculating the transition matrix elements of the interaction Hamiltonian between the eigenstates and the driving laser field. The results of these calculations can be found in Table 1. A detailed derivation can be found in the supplementary information.

Fitted data

In ref. 8, an absolute frequency measurement of 5 lines was performed, which were identified as transitions between certain quadrupole sublevels of the ground and the isomeric state of the ^{229}Th nucleus, see Table 1. The transition energies were fit to analytical solutions of the Hamiltonian to yield η, $Q_{\text{lab}}V_{zz}$ and $Q^{\text{m}}_{\text{lab}}V_{zz}$ thus obtaining the ratio of $Q^{\text{m}}_{\text{lab}}/Q_{\text{lab}}$. The frequency of the EFG-free isomer transition ν_{Th} was determined by a weighted averaging of the line frequencies.

Here, we independently verify the data fitting using the numerical solutions of the interaction Hamiltonian with $Q_{\text{lab}}V_{zz}$, η, Q^{m}_0/Q_0 and ν_{Th} as free parameters. Details of the employed statistical models and in particular the estimation of uncertainties can be found in the supplementary information, see figures 1 and 2 in there. We obtain $\nu_{\text{Th}} = 2020\,407\,384\,335(2)$ kHz, $Q^{\text{m}}_0/Q_0 = 1.01791(2)$, $\eta = 0.59164(5)$ and $Q_{\text{lab}}V_{zz} = 339.263(7)$ eb V/Å^2. The fit results are fully consistent with the results reported in ref. 8.

The experimental line intensities reported in ref. 8 qualitatively agree with the theoretical expectations. The transition frequencies are arithmetically consistent and stable over the measurement campaign; we therefore assume fluctuations in V_{ij}, i.e., due to changes in temperature or pressure, to be negligible. The temperature of the crystal in the measurement was 150(1) K. A detailed study of the above parameters with crystal temperature is presented in ref. 52.

Using the value $Q_{\text{lab}} = 3.11(2)$ eb for the ^{229}Th ground state quadrupole moment derived in ref. 39 yields $V_{zz} = 109.1(7)$ V/Å^2. The rather high asymmetry parameter η indicates a non-rotationally symmetric microscopic charge compensation configuration involving multiple atoms, such as two fluoride interstitials[53]. The microscopic atomic and electronic structure of the ^{229}Th defect in the CaF_2 crystal will be subject of an upcoming publication.

Data availability

The simple calculations, code, or data, generated during the current study are available from the corresponding author on request. However, we encourage using the equations derived in the manuscript.

References

1. Peik, E. et al. Nuclear clocks for testing fundamental physics. *Quantum Sci. Technol.* **6**, 034002 (2021).
2. Campbell, C. J. et al. Single-ion nuclear clock for metrology at the 19th decimal place. *Phys. Rev. Lett.* **108**, 120802 (2012).
3. Yamaguchi, A. et al. Laser spectroscopy of triply charged ^{229}th isomer for a nuclear clock. *Nature* **629**, 62–66 (2024).
4. Rellergert, W. G. et al. Constraining the evolution of the fundamental constants with a solid-state optical frequency reference based on the ^{229}Th nucleus. *Phys. Rev. Lett.* **104**, 200802 (2010).
5. Kazakov, G. A. et al. Performance of a ^{229}thorium solid-state nuclear clock. *N. J. Phys.* **14**, 083019 (2012).
6. Tiedau, J. et al. Laser excitation of the th-229 nucleus. *Phys. Rev. Lett.* **132**, 182501 (2024).
7. Elwell, R. et al. Laser excitation of the ^{229}Th nuclear isomeric transition in a solid-state host. *Phys. Rev. Lett.* **133**, 013201 (2024).
8. Zhang, C. et al. Frequency ratio of the ^{229m}th nuclear isomeric transition and the ^{87}sr atomic clock. *Nature* **633**, 63–70 (2024).
9. Safronova, M. S. et al. Search for New Physics with Atoms and Molecules. *Rev. Mod. Phys.* **90**, 025008 (2018).
10. Stadnik, Y. V. New bounds on macroscopic scalar-field topological defects from nontransient signatures due to environmental dependence and spatial variations of the fundamental constants. *Phys. Rev. D.* **102**, 115016 (2020).

11. Arvanitaki, A., Huang, J. & Van Tilburg, K. Searching for dilaton dark matter with atomic clocks. *Phys. Rev. D.* **91**, 015015 (2015).
12. Stadnik, Y. V. & Flambaum, V. V. Can dark matter induce cosmological evolution of the fundamental constants of nature? *Phys. Rev. Lett.* **115**, 201301 (2015).
13. Stadnik, Y. V. & Flambaum, V. V. Improved limits on interactions of low-mass spin-0 dark matter from atomic clock spectroscopy. *Phys. Rev. A* **94**, 022111 (2016).
14. Antypas, D. et al. New horizons: Scalar and vector ultralight dark matter. https://arxiv.org/abs/2203.14915. (2022).
15. Kennedy, C. J. et al. Precision metrology meets cosmology: improved constraints on ultralight dark matter from atom-cavity frequency comparisons. *Phys. Rev. Lett.* **125**, 201302 (2020).
16. Dzuba, V. A., Flambaum, V. V. & Webb, J. K. Space-time variation of physical constants and relativistic corrections in atoms. *Phys. Rev. Lett.* **82**, 888–891 (1999).
17. Dzuba, V. A., Flambaum, V. V. & Webb, J. K. Calculations of the relativistic effects in many-electron atoms and space-time variation of fundamental constants. *Phys. Rev. A* **59**, 230–237 (1999).
18. Kozlov, M., Safronova, M., Crespo López-Urrutia, J. & Schmidt, P. Highly charged ions: optical clocks and applications in fundamental physics. *Rev. Mod. Phys.* **90**, 045005 (2018).
19. Safronova, M. S. The search for variation of fundamental constants with clocks. *Ann. Phys.* **531**, 1800364 (2019).
20. Fadeev, P., Berengut, J. C. & Flambaum, V. V. Sensitivity of ^{229}Th nuclear clock transition to variation of the fine-structure constant. *Phys. Rev. A* **102**, 052833 (2020).
21. Barontini, G. et al. Measuring the stability of fundamental constants with a network of clocks. *EPJ Quantum Technol.* **9**, 12 (2022).
22. Beloy, K. et al. Frequency ratio measurements at 18-digit accuracy using an optical clock network. *Nature* **591**, 564–569 (2021).
23. Filzinger, M. et al. Improved limits on the coupling of ultralight bosonic dark matter to photons from optical atomic clock comparisons. *Phys. Rev. Lett.* **130**, 253001 (2023).
24. Flambaum, V. V. Enhanced effect of temporal variation of the fine structure constant and the strong interaction in ^{229}Th. *Phys. Rev. Lett.* **97**, 092502 (2006).
25. Litvinova, E., Feldmeier, H., Dobaczewski, J. & Flambaum, V. Nuclear structure of lowest ^{229}Th states and time-dependent fundamental constants. *Phys. Rev. C.* **79**, 064303 (2009).
26. Berengut, J. C., Dzuba, V. A., Flambaum, V. V. & Porsev, S. G. Proposed experimental method to determine α sensitivity of splitting between ground and 7.6 eV isomeric states in ^{229}Th. *Phys. Rev. Lett.* **102**, 210801 (2009).
27. Möller, P., Sierk, A. J., Ichikawa, T. & Sagawa, H. Nuclear ground-state masses and deformations: Frdm (2012). *Data Nucl. Data Tables* **109**, 1–204 (2016).
28. Minkov, N. & Pálffy, A. ^{229m}Th isomer from a nuclear model perspective. *Phys. Rev. C.* **103**, 014313 (2021).
29. Minkov, N., Pálffy, A., Quentin, P. & Bonneau, L. Skyrme-hartree-fock-bcs approach to th-229m and neighboring nuclei. *Phys. Rev. C.* **110**, 034327 (2024).
30. Ruchowska, E. et al. Nuclear structure of th-229. *Phys. Rev. C.* **73**, 044326 (2006).
31. Thielking, J. et al. Laser spectroscopic characterization of the nuclear-clock isomer ^{229m}th. *Nature* **556**, 321 (2018).
32. Krane, K. S. et al. *Introductory nuclear physics* (John Wiley & Sons, 1991).
33. Barrett, R. C. & Jackson, D. F. *Nuclear Sizes And Structure* (United Kingdom: Clarendon Press, 1977).
34. Hasse, R. W. & Myers, W. D.*Geometrical Relationships Of Macroscopic Nuclear Physics* (Springer Science & Business Media, 2012).
35. Caputo, A. et al. Sensitivity of nuclear clocks to new physics. *Phys. Rev. C* **112**, L031302 (2025)
36. Beringer, R. & Knox, W. J. Liquid-drop nuclear model with high angular momentum. *Phys. Rev.* **121**, 1195–1200 (1961).
37. Hasse, R. & Myers, W. D.*Geometrical Relationships of Macroscopic Nuclear Physics* (Springer-Verlag, Heidelberg, 1988).
38. Angeli, I. & Marinova, K. Table of experimental nuclear ground state charge radii: an update. *Data Nucl. Data Tables* **99**, 69 (2013).
39. Porsev, S. G., Safronova, M. S. & Kozlov, M. G. Precision calculation of hyperfine constants for extracting nuclear moments of ^{229}Th. *Phys. Rev. Lett.* **127**, 253001 (2021).
40. Campbell, C. J., Radnaev, A. G. & Kuzmich, A. Wigner crystals of ^{229}Th for optical excitation of the nuclear isomer. *Phys. Rev. Lett.* **106**, 223001 (2011).
41. Safronova, M. S. et al. Nuclear charge radii of ^{229}Th from isotope and isomer shifts. *Phys. Rev. Lett.* **121**, 213001 (2018).
42. Flambaum, V. & Dzuba, V. Search for variation of the fundamental constants in atomic, molecular, and nuclear spectra. *Can. J. Phys.* **87**, 25–33 (2009).
43. Butler, P. A. & Nazarewicz, W. Intrinsic reflection asymmetry in atomic nuclei. *Rev. Mod. Phys.* **68**, 349–421 (1996).
44. Chishti, M. M. R. et al. Direct measurement of the intrinsic electric dipole moment in pear-shaped thorium-228. *Nat. Phys.* **16**, 853 (2020).
45. Gaffney, L. P. et al. Studies of pear-shaped nuclei using accelerated radioactive beams. *Nature* **497**, 199–204 (2013).
46. Butler, P. A. et al. Evolution of octupole deformation in radium nuclei from coulomb excitation of radioactive ra-222 and ra-228 beams. *Phys. Rev. Lett.* **124**, 042503 (2020).
47. Flambaum, V. V. Enhanced nuclear Schiff moment and time-reversal violation in ^{229}Th-containing molecules. *Phys. Rev. C.* **99**, 035501 (2019).
48. Beeks, K. et al. *The nuclear excitation of thorium-229 in the CaF_2environment: Development of a crystalline nuclear clock*. Ph.D. thesis, Technische Universität Wien (2022).
49. Beeks, K. et al. Growth and characterization of thorium-doped calcium fluoride single crystals. *Sci. Rep.* **13**, 3897 (2023).
50. Beeks, K. et al. Optical transmission enhancement of ionic crystals via superionic fluoride transfer: Growing VUV-transparent radioactive crystals. *Phys. Rev. B* **109**, 094111 (2024).
51. Scheck, M. et al. A new avenue in the search for CP violation: Mössbauer spectroscopy of ^{227}Ac. *Eur. Phys. J. A* **59**, 116 (2023).
52. Higgins, J. S. et al. Temperature sensitivity of a thorium-229 solid-state nuclear clock. *Phys. Rev. Lett.* **134**, 113801 (2025).
53. Pimon, M., Grüneis, A., Mohn, P. & Schumm, T. Ab-initio study of calcium fluoride doped with heavy isotopes. *Crystals* **12**, 1128 (2022).

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Author contributions

K.B., Th.S., J.Y., and M.S.S. conceived the research. K.B., F.S., I.M., L.T.C., T.R., M.B., and To.S. prepared the thorium-doped crystal samples, and C.Z., T.O., J.S.H., J.F.D., and J.Y. performed the nuclear laser spectroscopy measurements. C.Z., T.O., J.S.H., and J.F.D. developed and operated the frequency-stabilized VUV frequency comb and contributed to the precision spectroscopy experiments. K.B., G.A.K., and M.S.S. developed the theoretical framework, carried out the modeling, and analysed the sensitivity to fundamental constants. K.B., G.A.K., Th.S. and M.S.S. wrote the initial draft manuscript. M.S.S. coordinated and supervised the project. All authors discussed the results, contributed to the writing of the manuscript, and approved the final version.

Competing interests

The authors declare no competing interests.

Additional information

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