Laser spectroscopic characterization of the nuclear–clock isomer $^{229}\text{mTh}$

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The isotope $^{229}\text{Th}$ is the only nucleus known to possess an excited state $^{229}\text{mTh}$ in the energy range of a few electronvolts—a transition energy typical for electrons in the valence shell of atoms, but about four orders of magnitude lower than typical nuclear excitation energies. Of the many applications that have been proposed for this nuclear system, which is accessible by optical methods, the most promising is a highly precise nuclear clock that outperforms existing atomic timekeepers. Here we present the laser spectroscopic investigation of the hyperfine structure of the doubly charged $^{229}\text{mTh}$ ion and the determination of the fundamental nuclear properties of the isomer, namely, its magnetic dipole and electric quadrupole moments, as well as its nuclear charge radius. Following the recent direct detection of this long-sought isomer, we provide detailed insight into its nuclear structure and present a method for its non-destructive optical detection.

$^{229}\text{Th}$ has a low-energy transition between the nuclear ground state and a long-lived isomer, $^{229}\text{mTh}$, at an excitation energy of about 7.8 eV. This enables the application of precision laser spectroscopy methods to excite and detect this nuclear transition.\textsuperscript{1-3} A nuclear clock—that is, an optical clock that uses this low-energy transition as the frequency reference—is expected to benefit from the smaller sensitivity of the nucleus to external perturbations, including frequency shifts from electromagnetic fields, compared to electronic transitions exploited in current atomic clocks.\textsuperscript{4-5} To achieve this application, we studied the nuclear properties of $^{229}\text{Th}$, as well as the excitation of the isomeric state and methods for its non-destructive observation.\textsuperscript{6}

The nuclear structure of $^{229}\text{Th}$ has been studied via $\gamma$-ray spectroscopy of the radiation emitted after the $\alpha$ decay\textsuperscript{9} of $^{233}\text{U}$, after the $3/2^+$ decay\textsuperscript{10.11} of $^{229}\text{Ac}$, after Coulomb excitation\textsuperscript{12}, and through a (d,t) transfer reaction\textsuperscript{13} with $^{230}\text{Th}$. The low-lying levels can be assigned to rotational bands described by the Nilsson model\textsuperscript{14}. The $^{229}\text{Th}$ ground state is the bandhead of a $5/2^+$ [633] rotational band and its nuclear moments have been determined experimentally\textsuperscript{12,15}. A second rotational band has been identified as $3/2^+$ [631]; its bandhead—the low-energy isomer $^{229}\text{mTh}$ that is investigated here—is still unresolved by $\gamma$-spectroscopy.

The transition energy between the ground state and the isomer has been determined\textsuperscript{16,17}, indirectly, as the difference between the $\gamma$-ray energies of intraband and interband transitions, to be $E_\gamma = 7.8(5)$ eV (all uncertainties represent a 68% confidence level). This corresponds to ultraviolet radiation of wavelength 160(10) nm, where the uncertainty is about 17 orders of magnitude larger than the expected natural linewidth. Depending on the electronic structure that surrounds the nucleus, the isomer may decay quickly via internal conversion\textsuperscript{2,3} or radiatively with an estimated natural (that is, unperturbed) lifetime of a few thousands of seconds.\textsuperscript{11,18-20} The nuclear moments of the isomer have been estimated from nuclear structure models\textsuperscript{18,21,22}. Many experimental attempts to induce and detect an optical excitation of this isomer have failed, impeded by the difficulty of producing widely tunable intense vacuum-ultraviolet radiation, by the background of ionizing radiation from the $^{229}\text{Th}$ samples and by competition with non-radiative relaxation processes\textsuperscript{6,23,24}. Apart from the spectroscopic determination of the nuclear spin and indirect measurements of the excitation energy\textsuperscript{16,17}, no experimental data on the nuclear properties of the isomer have been available until recently. Using recoil ions from the decay of $^{233}\text{U}$ as a source of $^{229}\text{mTh}$, electrons emitted from the internal-conversion decay of the isomer in neutral thorium were detected\textsuperscript{25} and the half-life for this process was measured\textsuperscript{26}.

The availability of the isomer through recoil ions provides a way to measure the unknown nuclear properties of $^{229}\text{mTh}$ via laser spectroscopy of electronic transitions. Here we report the optical detection of ions in the $^{229}\text{mTh}$ isomeric state and of the resolved hyperfine structure (HFS), which arises from the interaction of the isomer nucleus with the valence electrons (see Methods).

Laser spectroscopy of trapped $^{229}\text{Th}^{2+}$ ions

Of the charge states $^{229}\text{Th}^+$, $^{229}\text{Th}^{2+}$ and $^{229}\text{Th}^{3+}$ that are extracted from a $^{233}\text{U}$ source, $^{229}\text{Th}^{2+}$ was selected for the experiments because of its high production yield from the recoil source, the long lifetime of the isomer\textsuperscript{25} and its convenient electronic-level structure, which enables hyperfine spectroscopy with diode lasers with background-free fluorescence detection in the visible spectral range.

For high-resolution spectroscopy of the HFS of $^{229}\text{Th}^{2+}$, we use two independent linear radio-frequency ion traps\textsuperscript{25,27} (see Fig. 1a and b and Methods). One of the traps (located at the Physikalisch-Technische Bundesanstalt, PTB) is loaded with $^{229}\text{Th}^+$ produced by laser ablation from a target containing $^{229}\text{Th}$ and $^{232}\text{Th}$. Three-photon ionization of trapped $^{229}\text{Th}^+$ is used to produce $^{229}\text{Th}^{2+}$. The second trap (located at Ludwig-Maximilians-Universität, LMU) is loaded with $^{229}\text{Th}$ recoil ions from the $\alpha$ decay of $^{233}\text{U}$, where the isomeric state is populated via a 2% decay branch\textsuperscript{8} (see Methods for details about the generation of the $^{229}\text{Th}^{1+}$ ion beam). Therefore, the trapped ion cloud consists of a mixture of ions in the ground and the isomeric nuclear states. Daughter products of the $^{235}\text{U}$ decay chain are also trapped, but do not disturb the spectroscopic measurement (see Methods for details). The combination of the measurements in both traps allows us to identify clearly the hyperfine components of $^{229}\text{mTh}$, which appear only with the trapped recoil ions, and to measure the isotope and isomer shifts. In both traps, the ions are cooled to near room temperature.

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The spectroscopy part of the experimental setup consists of three external-cavity diode lasers (ECDLs). The two-step excitation is provided by ECDLs at wavelengths of 484 nm and 1,164 nm, with overlapping beams aligned along the trap axis. The third diode laser, at 459 nm, is used for single-photon excitation of Th$^{2+}$ from the 0$^\text{th}$ ground state to the 21784 state to monitor the number of Th$^{2+}$ ions in the trap and thus normalize the fluorescence signals observed from the different HFS components. This is required because the ion number decreases with time owing to chemical reactions and charge exchange with impurities in the buffer gas. The laser setup is depicted in Extended Data Fig. 1.

By collisions with a high-purity buffer gas (argon at PTB, helium at LMU; see Methods). Recording an excitation spectrum by scanning a laser across the Doppler-broadened lines yields a resolution of about 700 MHz. This does not allow us to resolve the HFS of Th$^{2+}$ lines and to distinguish the resonances of Th$^{2+}$ from those of Th$^{2+}$. As an example, Fig. 2b shows a line of Th (nuclear spin $I = 0$, no HFS splitting) and an unresolved HFS lineshape of Th$^{2+}$. For higher resolution we use two-step laser excitation, which is free from Doppler broadening. In this approach, the first laser excites ions of a narrow velocity class out of the thermal distribution to an intermediate state, where they are probed by resonant excitation to a higher-lying level using a second tunable laser. These ions are detected by a sensitive fluorescence detection method using decay channels at other wavelengths, which are free from stray background laser light.

The two-step laser excitation we choose the transition from the 63$^\text{rd}$ electronic state to 29300 via the 20711 intermediate state (the 20711 transition). The two-step resonances are mapped for a system-atic search for the unknown frequencies of the isomer resonances and for the quantitative analysis of the HFS spectra. The frequency of the 484-nm laser is tuned within the Doppler-broadened HFS of the 63$^\text{rd}$ 20711 line in 35 discrete steps of about 120 MHz. At each frequency step, the 1,164-nm laser is scanned continuously over a frequency range of more than 4 GHz to detect the two-step HFS resonances. The frequency of the first-step laser is stabilized at each position of the map using a Fizeau wavemeter, resulting in absolute instability of ≤5 MHz. The full-width at half-maximum of the two-step resonances obtained in the PTB trap (which uses argon as a buffer gas) is 70 MHz and 40 MHz for those observed at LMU (helium buffer gas). The two-step excitation mapping is performed twice, with co- and counter-propagating laser beams, to confirm the identification of the HFS components.

**Fig. 1 | Experimental setup and 229Th$^{2+}$ level scheme.** a, Schematic of the configuration of the ion source, ion trap and laser beams at PTB. b, Corresponding configuration at LMU RF, radio frequency; DC, direct current. c, Transitions and electronic configurations of Th$^{2+}$ levels relevant to the experiment, labelled by their energy in cm$^{-1}$ and the electronic angular momentum $J$, which are the same for both nuclear states (see Methods for details). Solid arrows indicate laser excitation and dashed lines show fluorescence decay channels.
Comparison of excitation spectra measured in the PTB and LMU traps. a, Two-step excitation resonances of Th$^{2+}$, obtained with the first laser stabilized at $-800$ MHz detuning with respect to the $^{229}$Th HFS centre and with the second laser being scanned. Red and blue colours indicate data acquired in the PTB and the LMU trap, respectively. $2\%$ of the ions in the LMU trap are in the isomeric state. b, Magnified view of a, with the LMU signal up-shifted for easier inspection. The arrows indicate the total momenta $F$ and $F'$ of the transitions of the ground-state (a) and isomeric (b) resonances (Extended Data Fig. 1), respectively, and the isomeric-state peaks are shaded in cyan. Resonances c1 and c2 belong to the nuclear ground state and arise from collision-induced changes of the intermediate-state HFS (Extended Data Table 1); the transitions involved have total momenta $F$ of 7/2 $→$ 5/2 and 7/2 $→$ 5/2, respectively, where the two $F$ numbers in parentheses are mixed by collisions.

All spectra are available in the Supplementary Information (Supplementary Figs. 1–29 and 31–59). Because the expected isomer signal is only about $2\%$ of the signal from ions in the ground state, we choose averaging times of typically four hours per spectrum.

Detection of the isomeric HFS

In the PTB trap we detect all nine resonances of the HFS of the ground state. A typical spectrum of the nuclear ground-state HFS resonances for co-propagating beams, obtained at PTB with two-step laser excitation, is shown in Fig. 3 with red points. The second signal (blue points) shows the HFS signal obtained in the LMU trap, where a small fraction of the ions is in the isomeric state. The HFS resonances of the isomeric state are clearly observed when compared with the data acquired at PTB. Figure 4 shows four resonances of the HFS of the isomeric state in a logarithmic scale for a different frequency of the first-step 484-nm laser.

In total, we observe seven out of eight resonances of $^{229}$Th$^{2+}$ in both (co- and counter-propagating) beam configurations (see Extended Data Fig. 3). The amplitude of the eighth resonance is calculated to be small with respect to the signal-to-noise ratio achieved in the experiment. The fraction of the ions in the isomeric state, obtained from the ratio of integrated fluorescence signals of the isomeric and ground-state resonances, is determined to be 2.1(5)\% (see Methods and Extended Data Fig. 4). This confirms the previously assumed branching to the isomeric state, which was inferred from $\gamma$ spectroscopy.

We observe a collision-induced change of the intermediate-state HFS population due to interactions with the buffer gases, such as the resonances c1 and c2 in Fig. 3. This effect is more substantial for He in the LMU trap than for Ar in the PTB trap. The positions of these resonances can be calculated from the measured hyperfine splitting of the intermediate state, enabling us to identify them in the spectra. This prevents the wrong assignment of resonances originating from collisions as isomeric-state HFS peaks. The width of these resonances is approximately 1.5 times larger than those of the nine main HFS resonances.

Furthermore, the amplitude ratio between these resonances and the direct two-step resonances drops considerably with the reduction of the He buffer-gas pressure, indicating that these resonances are indeed caused by collisions (see Extended Data Fig. 5).

Isomer properties

The observation of the isomeric-state HFS allows us to determine the magnetic dipole and quadrupole moments and the nuclear charge radius. To derive these properties, we determine the hyperfine constants $A$ (magnetic dipole) and $B$ (electric quadrupole) of the electronic structure for both the ground and isomeric nuclear states. We measure the frequency intervals between the resonances relative to the transmission peaks of a reference cavity and use a least-squares fit to determine the hyperfine constants for the $63$ and the $207$ $\text{Th}$ electronic states (see Methods). The upper electronic state of the two-step excitation has $J = 0$ and therefore no hyperfine splitting. The results are shown in Table 1.

From the measured ratio $A/m/A = -1.73(25)$, where $A$ and $A'$ are the hyperfine magnetic dipole constants of the isomeric state and the ground state, respectively, we determine the magnetic dipole moment $\mu$ of the isomer according to the relation $\mu = \mu A/m A A'$, where $\mu$ indicates the magnetic moment of the ground state and $\mu$ and $A$ are the spin values of the isomeric and ground states, respectively. Here we neglect the HFS anomalies, which are small for the $d^2$, $f^2$ and $g^2$ electronic configurations of the $\text{Th}^{2+}$ levels (see Fig. 1), and derive $\mu = -1.04(15)$. The nuclear magnetic moment $\mu$ of the ground state has been measured in two experiments$^{33,34}$ and the most precise value, $\mu = 0.360(7) \mu_B$ (where $\mu_B$ is the nuclear magneton), was obtained from high-precision calculations$^{33,34}$ and measurements$^{34}$ of the HFS of $^{229}$Th$^{2+}$. On the basis of this value, we derive the magnetic dipole moment of the isomeric state as $\mu = -0.37(6) \mu_B$. The discrepancy between these two results indicates that the simplified Nilsson approach is insufficient to quantitatively characterize the isomer because it neglects factors such as the expected collective quadrupole–octupole coupling of the nuclear deformation.

The spectroscopic quadrupole moment of the isomeric state $Q^m$ is determined by $Q^m = Q_B/m A A'$, where $Q_B$ is the spectroscopic quadrupole moment of the ground state. We use only the constants obtained for the $207\text{Th}$ electronic state to derive $B/m A A'$ (see Table 1).
The measured ratio of the spectroscopic quadrupole moments is $Q_m^\alpha/Q_s = 0.555(19)$. $Q_s$ has been measured in two independent experiments\cite{3.0,3.1} to be $3.15(3)$ eb and $3.11(6)$ eb (where eb stands for electron-barn, 1 eb = 1.6022 $\times 10^{-47}$ C m$^2$). Using the weighted mean value of these measurements, the quadrupole moment of the isomer is $Q_m^\alpha = 1.74(6)$ eb. The spectroscopic quadrupole moment is related to the intrinsic quadrupole moment $Q_0$ through\cite{3.2,3.3} $Q_m^\alpha = Q_0(3K_z^2-I(I+1))/(I(I+1)/(2I+3))$, resulting in $Q_m^\alpha = 8.7(3)$ eb for the isomeric state and $Q_0 = 8.8(1)$ eb for the ground state. Both states are the band-heads of their rotational bands and therefore the projection of the nuclear spin on the symmetry axis $K$ is equal to $I$. The intrinsic quadrupole moments of the two states are the same ($Q_0^\alpha/Q_0 = 0.994(1)$) within the uncertainty. Therefore, the nuclear charge distribution has a similar prolate shape in both states. This is in good agreement with theoretical predictions\cite{3.4,3.5}.

To investigate the difference of the charge radius between the ground state and the isomeric nucleus, the isomeric shifts of the first and second excitation steps are derived from the centres of the HFSs, calculated by setting $A = B = 0$. The isomeric HFS is shifted to a frequency 0.29(3) GHz lower than that of $^{229}$Th$^2$ for the transition $63^2 \rightarrow 207^{11}$. The isotope shift of this line between $^{229}$Th and $^{229}$mTh is 0.035(4). The isotopic isomer and isotope shifts for both transitions is $0.035(4)$. The uncertainty in $\Delta = \Delta E/C$ is dominated by the contribution from the $\pm$4% uncertainty in $Q_m^\alpha/Q_s$. Although this result is not sufficient to prove that $|\Delta E| \gg E_m$, it is possible that the $\alpha$-sensitivity of the $^{229}$Th nuclear clock exceeds those of existing atomic clocks by several orders of magnitude. Combined with the expected high accuracy of the nuclear clock, this high sensitivity to $\alpha$ will enable us to test predictions of temporal variations of coupling constants and to experimentally assess theories unifying gravity with other interactions.

| Table 1 | Hyperfine constants of $^{229}$Th$^2$ and $^{229m}$Th$^2$ for the electronic levels $63^2$ and $207^{11}$. |
|-----------------|-----------------|-----------------|-----------------|
| Level (cm$^{-1}$) | Nuclear ground state | Nuclear isomeric state |
| A (MHz) | B (MHz) | $A^\alpha$ (MHz) | $B^\alpha$ (MHz) |
| 63 | 151(8) | 73(27) | $-263(29)$ | 53(65) |
| 20,711 | 68(4) | 897(14) | $-151(22)$ | 498(15) |

The systematic uncertainty constitutes about 50% of the total uncertainty of the measurements. The main contributions to the systematic uncertainty are from the uncertainty in the reference cavity length, the instability of the frequency of the first-excitation-step laser and the nonlinearity of the frequency tuning of the second-step laser.

This is based on a model in which the small transition energy $E_m \approx 7.8$ eV appears as the result of the nearly perfect cancellation of a change in the Coulomb energy $\Delta E_C = E_m - E_0 \approx -1$ MeV by opposite and nearly equal changes of the nuclear energy through the strong interaction. Such a cancellation would be very sensitive to the values of the coupling constants of the electromagnetic and strong forces. This model has been criticized because the transition is performed by an unpaired neutron, leaving the Coulomb energies of $^{229}$Th and $^{229m}$Th essentially equal\cite{3.6}. By treating the nucleus as a uniform, hard-edged, prolate ellipsoid, the change in Coulomb energy can be expressed in terms of quantities that have been measured here:

$$\Delta E_C = (-485 \text{MeV}) \frac{(r_0^2/229)}{(r_0^2/229)} - 1 + (11.6 \text{MeV}) (Q_m^\alpha/Q_s) - 1 = -0.29(43) \text{MeV}$$

The uncertainty in $\Delta E_C$ is dominated by the contribution from the $\pm$4% uncertainty in $Q_m^\alpha/Q_s$. Although this result is not sufficient to prove that $|\Delta E| \gg E_m$, it is possible that the $\alpha$-sensitivity of the $^{229}$Th nuclear clock exceeds those of existing atomic clocks by several orders of magnitude. Combined with the expected high accuracy of the nuclear clock, this high sensitivity to $\alpha$ will enable us to test predictions of temporal variations of coupling constants and to experimentally assess theories unifying gravity with other interactions.

**Online content**

Any Methods, including any statements of data availability and Nature Research reporting summaries, along with any additional references and Source Data files, are available in the online version of the paper at https://doi.org/10.1038/s41586-018-0011-8.

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METHODS
Ion trapping. PTB trap. Using laser ablation with a neodymium-doped yttrium–aluminium garnet (Nd:YAG) laser emitting 5-ns pulses with an energy of $\sim 1$ mJ at 1.064 nm, the radio-frequency linear Paul trap located at PTB is loaded with about $10^7$ $^{233}$Th$^+$ ions from a Th(NO$_3$)$_2$ solution (containing approximately equal amounts of $^{229}$Th and $^{233}$Th) doped with about 90-ns-long pulses by an acousto-optical modulator, to use the pumped 2847 nm $^{233}$Th$^+$ state. The second- and third- ionization-step photons are provided by third-harmonic generation of a pulsed titanium sapphire (Ti:Sa) laser (pulse length of 20 ns, third-harmonic generation peak power of about 1 kW) via the 63,258 nm $^{233}$Th$^+$ state. Both lasers operate at a repetition rate of 1 kHz, and the pulses of the Ti:Sa laser and the ECDL are overlapped in time. The continuous resonant ionization produces a cloud of about $10^6$ $^{233}$Th$^{2+}$ ions and compensates the loss of $^{233}$Th$^{2+}$ ions due to the formation of molecules with impurities of the buffer gas. By using argon as a buffer gas at a pressure of 0.1 Pa, we cool the ions to room temperature and depopulate metastable states by collisional quenching. Argon is used because it achieves a higher trap-loading efficiency than He. For the photo- dissociation of $^{233}$Th$^+$ compounds formed in the trap, we use the fourth-harmonic radiation of a Q-switched Nd:YAG laser operating at 266 nm with a pulse energy of about 10 $\mu$J (see Extended Data Fig. 2 for the scheme of the optical setup). Owing to the presence of $^{229}$Th- and $^{233}$Th$^+$ isotopes in the solution on the target, we trap these ions (together with the $^{233}$Th$^{2+}$ ions) with a buffer-gas stopping cell filled with $3.2 \times 10^5$ Pa of ultra-pure He 6.0, which is further purified by catalytic purification and a cryotrap.$^{44,45}$ During the stopping process, charge exchange occurs between the thorium recoils and the buffer gas, producing pre-dominantly thorium ions in the 2+ and 3+ charge states. After thermalization, the ions are separated by charge-state-dependent decay in a magnetic field. The ions are guided by an electric radio-frequency and direct-current funnel system consisting of 50 ring electrodes towards a Laval extraction nozzle with a 0.6-mm-diameter nozzle throat. The extraction nozzle forms a supersonic gas jet and directs the ions into the subsequent (12-fold segmented) radio-frequency quadrupole, operated in this experiment as an ion trap. The extraction efficiency of the buffer gas stopping cell is about 5% and 10% in the 2+ and 3+ charge states, respectively, and thus more than $10^6$ $^{229}$Th$^+$ ions are actually trapped because of the trap’s limited loading capacity. Besides $^{229}$Th, other isotopes originating from the $\alpha$ decay of nuclides other than $^{229}$U (for example, from $^{232}$U and its decay chain) are discussed in the sections ‘Exclusion of $^{230}$Th isotope’ and ‘Exclusion of coin- compounds formed in the trap$^{42}$ we use the fourth-harmonic channel of the 484-nm or 459-nm excitations by using a filter that transmits at 643 ± 10 nm or 540 ± 8 nm, respectively, which blocks the laser stray light. Counters are used to register the photomultiplier signals. In the PTB trap, photon counting is terminated during the ionization and dissociation laser pulses to prevent them influencing the spectroscopic signal.

HFS. If a nucleus has spin $I > 1/2$, it may have a magnetic dipole moment and an electric quadrupole moment. The interaction of the valence electrons with these moments cause hyperfine splitting of the electronic levels, where the energy shift of an individual level is determined by $E_{\text{HFS}}(I, F, F') = A K + B (3/4)(K(K + 1) - I(I + 1))(F' + 1)/2(2I(I + 1)(2I - 1))$ with $K = F(F + 1) - I(I + 1) - I(I + 1)$. The hyperfine constants $A$ and $B$ are determined from the magnetic dipole and electric quadrupole interactions$^{46,47}$. For $^{232}$Th, the ground state has spin $I = 5/2$ and the isomer has $P^+ = 3/2$. This leads to a splitting of the 63; electronic level into five sub-levels for the ground state and four sub-levels for the isomeric state. The 20711 level consists of three hyperfine levels in both nuclear states. Following the selection rules for electric dipole transitions ($\Delta F = 0, \pm 1$), the spectrum of the two-step excitation with angular momentum $2 \rightarrow 1$ corresponds to the 63; electronic level into five sub-levels for the ground state and four sub-levels for the isomeric state. The 20711 level consists of three hyperfine levels in both nuclear states. Following the selection rules for electric dipole transitions ($\Delta F = 0, \pm 1$), the spectrum of the two-step excitation with angular momentum $2 \rightarrow 1$ to 0 for the ground and isomeric states consists of nine and eight resonances, respectively (see Extended Data Fig. 1).

Two-step excitation resonances. Assume laser 1 has a frequency detuning $\Delta f_1$ with respect to the centre of the first-step HFS. $\Delta f_2$ and $\Delta f_3$ are the frequency shifts of the second and third laser excitation steps. The amplitude of the resonance depends on the fraction of ions within the velocity group of the 20711 level, which is determined by $\Omega = \Delta f_1 + \Delta f_2 + \Delta f_3$.

Spectroscopic lasers. The spectroscopy of the HFS of the thorium isomer is performed using continuous-wave ECDLs with a typical linewidth of 100 kHz. The ECDL at 1.164 nm has an output power of 30 mW and a tuning range greater than about 4 $\mu$Hz near 459 nm to 20 ns long, beginning at about 15 mW and a tuning range greater than 15 GHz. The radiation of the lasers is delivered to the trap by single-mode polarization-maintaining fibres. The power of all lasers in both traps is about 4 mW owing to losses in fibre coupling and clipping by the supersonic nozzle, which corresponds to an intensity of 1.5 W cm$^{-2}$ for each beam. The scheme of the optical setup is shown in Extended Data Fig. 2.

Laser frequency measurement and stabilization. To avoid long-term frequency drifts of the 459-nm laser and to provide controlled frequency steps of the 484-nm laser with an accuracy of a few megahertz, their wavelengths are stabilized to a Fabry–Pérot wavemeter (HighFinesse WS7) by a computer-based locking system. A RB-stabilized ECDL at 780 nm is used to calibrate the wavemeter in intervals of 1,000 s. The 780-nm ECDL is stabilized to the $^{230}$Yb$^+(F = 3) \rightarrow \text{P}_{1/2}(F = 4)^{3/2}$ line, where $F$ is the total angular momentum, by the modulation-transfer spectroscopy technique$^{46}$.

Fluorescence detection. The fluorescence of the excited Th$^{2+}$ ions is detected using a photomultiplier tube with a bandpass interference filter that transmits in the range of 445 ± 23 nm, which corresponds to one of the decay channels of the excited state and provides a background-free detection for the two-step excitation. A second photomultiplier tube is used for the measurements of the isotopic shift or, alternatively, to provide measurements of the number of thorium ions in the trap during the two-step excitation. This photomultiplier is set to detect the decay channels of the 484-nm or 459-nm excitations by using a filter that transmits at 643 ± 10 nm or 540 ± 8 nm, respectively, which blocks the laser stray light. Counters are used to register the photomultiplier signals. In the PTB trap, photon counting is terminated during the ionization and dissociation laser pulses to prevent them influencing the spectroscopic signal.

Map of the HFS resonances. Extended Data Fig. 3 shows selected spectra demonstrating the evolution of the HFS peaks of the ground and isomeric states for different frequency positions of the first excitation step. The isomeric-state HFS resonances are marked with red labels and all resonances are described in Extended Data Table 1. The evaluation of the positions and the amplitudes of the seven additional peaks that are only observed in the LMU trap over the mapping indicates that those resonances fit to the HFS pattern of an $I = 3/2$ isomer. We tested for the appearance of spurious resonances from the collision-induced intermediate-state
changes, possible laser multimode operation and back-reflection of the spectroscopic laser beams (mixing of co- and counter-propagating beam geometries) and confirmed that the observed resonances are genuine spectroscopic features.

Extended Data Fig. 4 was generated by tracking the positions and amplitudes of the second-step resonances for all acquired spectra in both nuclear states. The figure shows the relative amplitudes of the resonances and was used to calculate the fraction of ions in the isomeric state.

Exclusion of $^{230}$Th isotope. Estimations for daughter products from the $^{231}$U source (Extended Data Table 2) show upper limits for the presence of the $^{228}$Th and $^{230}$Th isotopes. For all transitions, the resonance of $^{230}$Th ($I = 0$, no HFS) should appear between the lines of $^{232}$Th and $^{229}$Th, like those of the HFS of $^{229m}$Th. The isotopic shift of $^{230}$Th$^{1+}$ with respect to $^{229}$Th$^{1+}$ for the 484-nm transition is 3.2 GHz, as calculated from previous measurements at 459 nm using the LMU trap with a $^{234}$U source and determined in an earlier work. This shift is outside the range of the observed isomeric HFS. In the experiment with the $^{231}$U source, the $^{230}$Th$^{1+}$ resonance is not observed, indicating a considerably smaller flux than that listed in Extended Data Table 2.

Exclusion of coincident absorption lines. Because the $^{231}$U source emits a variety of ions of different elements ($\alpha$-decay daughter products of the $^{233}$U decay chain), which are loaded into the trap simultaneously with the thorium ions (see Extended Data Table 2), spectral lines of those elements might be detected in parallel to the isomer signal. Only two elements (U and Pu) have a flux high enough to be detected. For the first excitation step, the uranium and plutonium lines that are closest to the $^{229}$Th$^{2+}$ resonances and that originate from low-lying levels are detuned by about 200 GHz ($^{234}$U, $\nu = 20,640.51$ cm$^{-1}$, transition from 91592 to 2115$^{(5/2)}$, and about 60 GHz ($^{239}$Pu, $\nu = 20,645.62$ cm$^{-1}$, transition from 397052 to 2461$^{(5/2)}$). We exclude also the influence of Th$^{2+}$-He complexes, which can be formed by interaction with the buffer gas: owing to their low binding energy, estimated to be less than 7,500 cm$^{-1}$, these complexes would be dissociated by laser excitation at 484 nm (20.648 cm$^{-1}$). Moreover, because the signal that we detect requires coincidence of two resonance conditions, we can rule out that the recorded thorium spectra are affected by other elements.

Isomeric lifetime in Th$^{2+}$. It is in principle possible to determine the isomeric lifetime in Th$^{2+}$ by measuring the time evolution of the amplitudes of the resonances for both nuclear states. The isomeric signal will show an additional exponential decay due to its finite lifetime. This experiment is at present limited by the storage time of the ions in the trap (about 60 s), which is defined by chemical reactions and charge exchange with impurities in the buffer gas. Therefore, this value can only be given as a lower limit for the isomer lifetime. By improving the storage time by two orders of magnitude, measuring the isomeric lifetime will become feasible and provide an important parameter of the clock.

Sensitivity to the fine-structure constant. The $^{229}$Th nuclear clock has been proposed as a particularly sensitive system to search for temporal variations of the fine structure constant $\alpha$, but this proposal has been met with scepticism. The sensitivity ($\delta \alpha/\delta f_0$) of the nuclear transition frequency $f_0$ to the value of $\alpha$ is equal to the ratio $\Delta E_C/E_C$ of the change in Coulomb energy ($\Delta E_C$) and the total transition energy ($E_C \approx 7.8$ eV) between the ground and isomeric states. Theoretical estimations of $\Delta E_C$ from nuclear structure calculations vary from a few kiloelectronvolts to a few meaelectronvolts. It has been proposed that $\Delta E_C$ can be calculated via the change of the nuclear charge radius and electric quadrupole moment from measured isomer shift and HFS data. Applied to our data, using updated values of $Q_0$ and $(r_2^{2+})$, this results in $\Delta E_C = -0.29(43)$ MeV. Because the change in the charge radius $\Delta (r^2)/(r^2) \approx 4 \times 10^{-7}$ is small, the uncertainty in the change of the quadrupole moments, which is known from $Q_0^2/Q_0 = 0.99(4)$, yields the dominant uncertainty contribution to $\Delta E_C$. Although smaller values cannot be excluded with certainty, the most probable modulus for the $\alpha$-sensitivity of a $^{229}$Th nuclear clock is about $4 \times 10^{-5}$.

Data availability. Source Data for Figs. 2–4 and Extended Data Figs. 3–5 are provided in the online version of the paper. Further data are available at https://zenodo.org/communities/nuclock/ and from the corresponding author upon reasonable request.

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Extended Data Fig. 1 | Detailed level scheme of the two-step excitation. Transitions and electronic configurations of the initial (g), intermediate (i) and excited (e) states relevant to the experiment are shown, labelled by their energy in cm$^{-1}$ and the electronic angular momentum $J$. Hyperfine sub-levels are indicated by their total angular momentum $F$ and $F^m$. Transitions belonging to the same intermediate hyperfine level are depicted with the same colour. The hyperfine intervals are calculated from the hyperfine constants $A$ and $B$ presented in Table 1.
Extended Data Fig. 2 | Scheme of the optical setup. The spectroscopy laser of the first step excitation (484 nm) is locked to the wavemeter, which is calibrated by a Rb-stabilized ECDL at 780 nm. The second-step (1,164 nm) laser tuning is monitored with the confocal cavity. The ECDL at 459 nm is used to detect the number of ions in the traps. The loading of Th$^{2+}$ in the PTB trap is provided by ablation (nanosecond Nd:YAG laser at 1,064 nm) and further three-photon ionization. The first step uses a 402-nm ECDL, pulsed via an acousto-optical modulator (AOM), and the second and third steps involve third-harmonic generation (THG) of a nanosecond Ti:Sa laser. Molecular compounds of Th$^+$ are photodissociated by pulses from a Q-switched diode-pumped solid-state laser (Q-DPSS).
Extended Data Fig. 3 | Selected spectra obtained by two-step excitation. The resonances recorded for different positions of the 484-nm ECDL show the observed isomeric peaks for the case of co-propagating beams (labelled ‘i’). The resonances that originate from collisions of ions in the intermediate state are labelled ‘c’. The description of the peaks and their total angular momenta are given in Extended Data Table 1. Black lines show the recorded data and blue lines represent a multi-Lorentz fit with fixed width, which is used to extract the line centres and frequency intervals.
Extended Data Fig. 4 | Mapping of the second excitation step. The experimental points represent amplitudes and positions of the two-step resonances obtained by setting the 484-nm laser at certain frequencies and tuning the 1,164-nm laser. The frequency of the 484-nm laser is changed in steps of about 120 MHz. The resonance groups shown with the same colour correspond to transitions from the same intermediate state with total angular momentum $F$, which is populated from different ground-state hyperfine components. The graphs show the HFS transitions of $^{229}$Th$^{2+}$ in the ground state (a) and the isomer (b).
Extended Data Fig. 5 | Pressure dependence of collision-induced changes in the intermediate-state HFS. The two-step excitation resonances of Th$^{2+}$ were obtained with the first laser stabilized at $-800$ MHz detuning with respect to the $^{232}$Th HFS centre and the second laser scanned. The measurement is performed for two different He buffer-gas pressures and shows a decrease in the relative amplitude of the collisional resonances for the reduction of the buffer-gas pressure. We note that the isomeric resonance is not affected by the change in He pressure.
Extended Data Table 1 | Systematics of the observed resonances

| Label | \( F_g \rightarrow F_i \rightarrow F_e \) | Label | \( F_g \rightarrow F_i; F_i \rightarrow F_e \) | Label | \( F_{g}^{n} \rightarrow F_{i}^{n} \rightarrow F_{e}^{n} \) |
|-------|--------------------------------|-------|--------------------------------|-------|--------------------------------|
| 1     | \( 9/2 \rightarrow 7/2 \rightarrow 5/2 \) | c1    | \( 7/2 \rightarrow 5/2; 7/2 \rightarrow 5/2 \) | i1    | \( 7/2 \rightarrow 5/2 \rightarrow 3/2 \) |
| 2     | \( 7/2 \rightarrow 7/2 \rightarrow 5/2 \) | c2    | \( 7/2 \rightarrow 5/2; 3/2 \rightarrow 5/2 \) | i2    | \( 5/2 \rightarrow 5/2 \rightarrow 3/2 \) |
| 3     | \( 5/2 \rightarrow 7/2 \rightarrow 5/2 \) | c3    | \( 9/2 \rightarrow 7/2; 3/2 \rightarrow 5/2 \) | i3*   | \( 3/2 \rightarrow 5/2 \rightarrow 3/2 \) |
| 4     | \( 7/2 \rightarrow 5/2 \rightarrow 5/2 \) | c4    | \( 5/2 \rightarrow 5/2; 3/2 \rightarrow 5/2 \) | i4    | \( 5/2 \rightarrow 3/2 \rightarrow 3/2 \) |
| 5     | \( 5/2 \rightarrow 5/2 \rightarrow 5/2 \) | c5    | \( 7/2 \rightarrow 7/2; 3/2 \rightarrow 5/2 \) | i5    | \( 3/2 \rightarrow 3/2 \rightarrow 3/2 \) |
| 6     | \( 3/2 \rightarrow 5/2 \rightarrow 5/2 \) | c6    | \( 7/2 \rightarrow 7/2; 5/2 \rightarrow 5/2 \) | i6    | \( 1/2 \rightarrow 3/2 \rightarrow 3/2 \) |
| 7     | \( 5/2 \rightarrow 3/2 \rightarrow 5/2 \) | c7    | \( 5/2 \rightarrow 3/2; 5/2 \rightarrow 5/2 \) | i7    | \( 3/2 \rightarrow 1/2 \rightarrow 3/2 \) |
| 8     | \( 3/2 \rightarrow 3/2 \rightarrow 5/2 \) | c8    | \( 3/2 \rightarrow 3/2; 5/2 \rightarrow 5/2 \) | i8    | \( 1/2 \rightarrow 1/2 \rightarrow 3/2 \) |
| 9     | \( 1/2 \rightarrow 3/2 \rightarrow 5/2 \) | | | |

The detected resonances are listed with the total angular momenta of the electronic states involved in the excitation. The resonances of the nuclear ground state are labelled with numbers. The resonances that arise from collisional changes of the intermediate state population are described by both quantum numbers, \( F_i \) (before the collision) and \( F_i' \) (after the collision), and are labelled 'c'. Isomeric resonances are marked with 'i'. The resonance i3 (marked with an asterisk) is not observed in the experiment.
Extended Data Table 2 | Extraction of isotopes from the $^{233}$U source

| Isotope          | Extraction (rel. to Th-229) | Isotope          | Extraction (rel. to Th-229) |
|------------------|-----------------------------|------------------|-----------------------------|
| Th-229           | 1                           | Pu-238           | $4 \times 10^{-6}$          |
| U-233            | $\approx 1$                 | Pu-239           | $2 \times 10^{-3}$          |
| Th-229 decay chain | $2 \times 10^{-4}$           | U-235            | $1 \times 10^{-2}$          |
| U-232            | $6 \times 10^{-7}$           | Pu-240           | $4 \times 10^{-4}$          |
| Th-228           | $1 \times 10^{-3}$           | U-236            | $1 \times 10^{-2}$          |
| Th-228 decay chain | $7 \times 10^{-4}$           | Pa-231           | $2 \times 10^{-5}$          |
| U-234            | $2 \times 10^{-2}$           | Ac-227           | $1 \times 10^{-4}$          |
| Th-230           | $7 \times 10^{-3}$           | Ac-227 decay chain | $6 \times 10^{-6}$          |

The numbers of recoils leaving the $^{233}$U source via α decay and sputtering are listed. The extraction ratios show the upper limit on the number of recoils relative to the number of $^{233}$Th recoils.