

$J = 0$ metastable state of Th^{2+} for a hyperfine-free nuclear clock

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We present measurements on a metastable state in Th^{2+} with the electronic configuration $6d^2\ ^3P_0$ (5090 cm^{-1}). This is motivated by the prospect of using the state in laser excitation of the low-energy ^{229}Th nuclear resonance independent from the leading hyperfine interactions. The $6d^2\ ^3P_0$ state has no dipole-allowed radiative decay channel and is connected to the second ground state $6d^2\ ^3F_2$ (63 cm^{-1}) through an electric quadrupole transition only. We populate the state by laser excitation at 484 nm via a higher excited level and detect its population in laser-induced fluorescence. The isotope shift of the $J = 0$ level between $^{232}\text{Th}^{2+}$ and $^{229}\text{Th}^{2+}$ is determined as a measure of the interaction of electronic and nuclear charge distributions. The lifetime of the level in our ion trap with buffer gas is limited by collisional mixing with the nearby state $5f6d\ ^3G_3$ (5060 cm^{-1}). In ultrahigh vacuum, it could serve as a hyperfine-free nuclear clock that is largely immune to field-induced frequency shifts via the electron shell.

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I. INTRODUCTION

The exceptionally low-energy nuclear transition (8.4 eV) in ^{229}Th represents a unique opportunity for the realization of a nuclear optical clock [1,2]. Direct laser excitation of the thorium nucleus in a doped crystal [3,4] has resolved the long-standing uncertainty in the transition frequency and yielded a precise value. Furthermore, detailed studies with thorium-doped crystals, including measurements of the nuclear electric quadrupole structure [5] and investigations demonstrating frequency reproducibility [6], have significantly strengthened the foundation for precision nuclear frequency metrology.

In contrast to the solid-state approach, a nuclear clock based on a trapped ion in a radio-frequency trap could offer higher accuracy by isolating the ion from environmental perturbations. However, due to the hyperfine coupling between the nucleus and the valence electrons, the sensitivity of the transition frequency to external fields depends on the electron quantum numbers. Two classes of states have been identified that minimize the sensitivity: (i) states with electronic angular momentum $J = 0$ or $J = 1/2$ [1], and (ii) stretched states with the maximum value of the total angular momentum $F = |m_F| = J + I$ [2]. For both classes of states, the field-induced systematic frequency shifts can be suppressed so that they become negligible in comparison to the second-order Doppler effect which will dominate the uncertainty budget [1,2,7,8].

The electronic structures of thorium cations in charge states Th^+ to Th^{4+} show significantly different properties. The level

schemes of Th^+ and Th^{2+} are characterized by a high density of states and strong configuration mixing [9–12]. Th^{3+} , originally proposed for the nuclear clock [1], presents a simple level structure that is amenable to laser cooling [13,14]. The ground state possesses $J = 5/2$ and a metastable s state with $J = 1/2$ and an approximate lifetime of 600 ms exists [15,16]. Th^{4+} possesses a $J = 0$ ground state and a simple radonlike level structure with a first resonance line at about 61 nm [17,18]. This indicates that the nuclear excitation is well decoupled from electronic excitations, as is desired for an accurate nuclear clock.

Experimental studies of the electronic hyperfine structure of the ^{229}Th nuclear ground state have been performed in trapped ^{229}Th ions of different charge states. In particular, laser-cooled $^{229}\text{Th}^{3+}$ ions have been used for high-resolution spectroscopy [13,19], while buffer-gas-cooled $^{229}\text{Th}^+$ and $^{229}\text{Th}^{2+}$ ions have also been investigated [20,21]. Furthermore, hyperfine-resolved spectroscopy of both the nuclear ground state and the isomeric state of ^{229}Th , produced as recoil ions from the α decay of ^{233}U , has been demonstrated in trapped $^{229}\text{Th}^{2+}$ and $^{229}\text{Th}^{3+}$ ions under buffer-gas cooling conditions [21,22]. Experiments with $^{229}\text{Th}^{4+}$ have not yet been reported. As a recoil ion, Th^{4+} cannot be slowed down in a buffer gas because it would undergo charge exchange [23], and the lack of electronic resonance lines that are suitable for laser excitation makes state preparation and detection more difficult than in the lower charge states.

Observation of the isomeric state in laser-cooled or sympathetically cooled ^{229}Th ions has not yet been achieved. The successful realization of such measurements would enable more precise determinations of the isomer's nuclear moments and charge radius. Similarly, direct laser excitation of the nucleus in trapped ions remains experimentally challenging due to the limited available laser power at 148 nm and the achievable low excitation rate [24]. There is an increasing interest in Th^{2+} , which has favorable properties for nuclear excitation and as a potential candidate for a nuclear clock.

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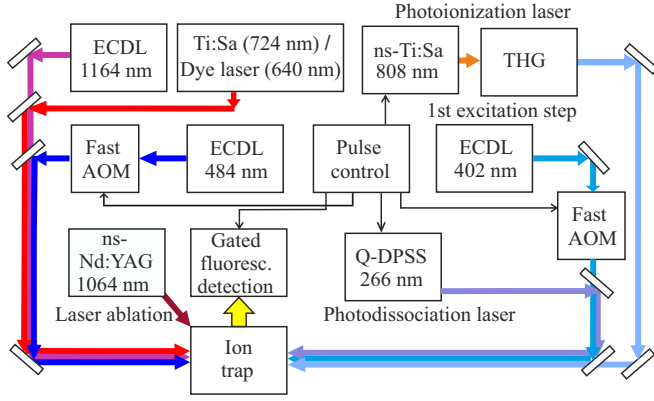


FIG. 1. Optical setup for the spectroscopy of Th^{2+} ions. An external cavity diode laser (ECDL) at 402 nm is used to monitor Th^+ ions and, in combination with photoionization laser, to produce Th^{2+} ions. A photodissociation laser is used to dissociate formed molecules of Th^+ . The lasers used for the spectroscopy of Th^{2+} are shown on the left side of the optical setup.

In particular, it has been proposed as a system for nuclear excitation through electronic bridge processes [25] and for two-photon nuclear excitation [26].

In this work, we investigate a metastable electronic level with total angular momentum $J = 0$ in Th^{2+} that is particularly attractive due to its symmetry and the isolation it provides from external perturbations to the nucleus through the hyperfine interaction. It may serve as a hyperfine-free nuclear clock candidate, analogous to a recent proposal for Th^{4+} clock schemes that exploit the closed-shell ground-state configuration with $J = 0$ [18]. The level, designated as $6d^2\ ^3P_0$ (5090 cm^{-1}), has no electric dipole-allowed transition to any lower level. Here, we show how the state can be populated via laser excitation to higher excited states. The population is detected in laser-induced fluorescence, and the isotope shift between $^{232}\text{Th}^{2+}$ and $^{229}\text{Th}^{2+}$ is measured. Furthermore, we also determine its lifetime as determined by collisions in different buffer gases under varying pressures. The radiative lifetime of this level has been calculated to be 95.5 s [11] for decay through the electric quadrupole transition ($E2$) to the $6d^2\ ^3F_2$ (63 cm^{-1}) state at a wavelength of 1989 nm .

Due to strong configuration mixing, designations of levels of Th^{2+} in LS coupling can only give an approximate description. In the following, we label the states by their energies (in cm^{-1}) and total angular momentum J as a subscript.

II. EXPERIMENTAL SETUP

The apparatus for trapping Th^{2+} ions used in this experiment is the same as that described in Refs. [21,27]. We first load Th^+ ions by laser ablation from ^{232}Th and ^{229}Th targets. The ions are cooled to near room temperature by collisions with buffer gas (Ar or He). The optical setup with all the lasers used is shown in Fig. 1. After loading Th^+ , we ionize it to Th^{2+} via a three-photon process, which includes one photon of 402 nm and two photons from the photoionization laser at 269 nm [28]. With a lifetime of Th^{2+} in the trap of approximately 400 s , we operate the system in a steady-state

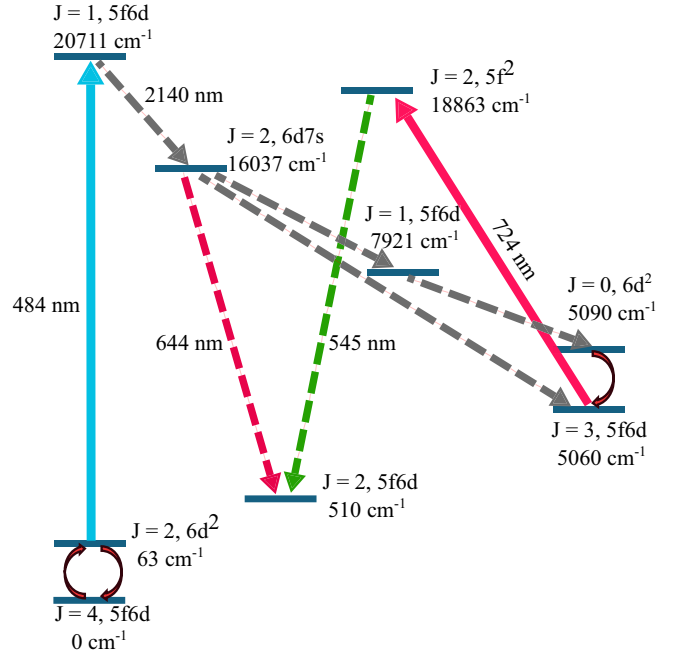


FIG. 2. Simplified level scheme of Th^{2+} showing the dominant transitions that are used to populate the $J = 0$ state. Solid arrows indicate applied lasers, dashed arrows indicate spontaneous decay from ions in different states, and curved arrows indicate collisional mixing.

regime of constant Th^{2+} number, where the loss of Th^{2+} through the formation of molecular ions is balanced by the ionization of Th^+ . Estimated ion numbers are $\approx 10^5$ for Th^+ and $\approx 10^3$ for Th^{2+} . To populate the 5090_0 state, we start with ions from the 63_2 state that is populated with about 40% of the ions by collisions with the buffer gas. The level scheme of the transitions used for excitation and fluorescence detection is shown in Fig. 2. Collisional mixing is illustrated by curved arrows. It generates the dominant decay mechanism for the metastable level in our experiment. The experimental sequence runs at a rate of 1 kHz . We apply a 484-nm laser pulse (5 mW of laser power) for $150\text{ }\mu\text{s}$ in duration that drives the transition $63_2 \rightarrow 20711_1$. The decay of the 20711_1 level populates a detectable number of ions in the 5090_0 state via the channel shown in Fig. 2. Increasing the pulse duration of the 484-nm laser does not improve the signal, as the lifetime of the metastable state in the buffer gas is of the same order. After $150\text{ }\mu\text{s}$, the 484-nm laser is turned off, and fluorescence from the metastable state is detected to verify the population there. The detailed scheme and the energy levels used are described in the next section. For the lifetime measurement, the experiment is performed using the ^{232}Th isotope because of easier loading of ions. We continuously monitor the fluorescence through the 5060_3 state for $700\text{ }\mu\text{s}$, similar to the optical shelving method [29,30]. This method allows us to continuously monitor the decay of ions from the metastable state through collisional mixing. After $700\text{ }\mu\text{s}$ of monitoring period, we apply a 402-nm laser pulse (500 ns) to monitor the number of Th^+ ions, along with an ionization pulse (20 ns) and a pulse at 266 nm (10 ns) to dissociate molecular ions that Th^+ may have formed in reactions with the background

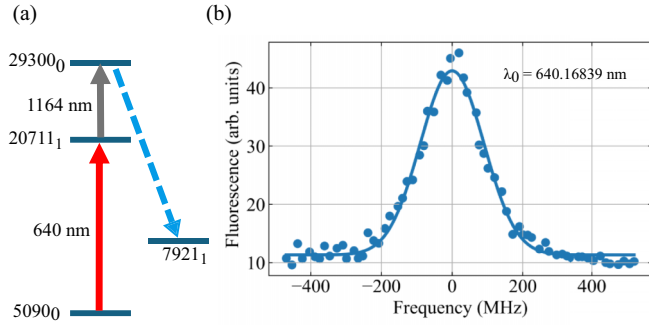


FIG. 3. Fluorescence detection after two-step excitation from the metastable state of $^{232}\text{Th}^{2+}$. Solid markers represent experimental data, and the line corresponds to the fitted curve.

gas. We provide a delay of 150 μs to allow for the decay of a luminescence background induced by the pulsed lasers in the viewports. The entire cycle runs at a repetition rate of 1 kHz.

III. DETECTION OF POPULATION IN THE METASTABLE STATE

To detect ions present in the 5090₀ state, we use a two-step excitation scheme, coupling 5090₀ $\rightarrow$ 20711₁ $\rightarrow$ 29300₀ states. This scheme is shown in Fig. 3(a) with the corresponding energy levels and their J values. The previously used two-step scheme from 63₂ $\rightarrow$ 20711₁ $\rightarrow$ 29300₀ enabled us to identify the wavelength for the transition 20711₁ $\rightarrow$ 29300₀, which corresponds to a wavelength of 1164.3 nm [21]. We stabilize the frequency of the 1164-nm laser (15 mW) and scan the 640-nm laser (10 mW) to couple 5090₀ $\rightarrow$ 20711₁. In Fig. 3(b), we show the detected fluorescence signal that is proportional to the population of the metastable state for $^{232}\text{Th}^{2+}$. The measured full width at half maximum (FWHM) is 210 MHz, determined by the frequency selectivity of the second step. The value for the transition wavelength reported in Refs. [10,31] for the 640-nm transition lies within this range, indicating good agreement for the position of the 5090₀ level for $^{232}\text{Th}^{2+}$.

For ^{229}Th , with a ground-state nuclear spin of $I = 5/2$, the hyperfine structure gives rise to three F levels in the intermediate state ($J = 1$). The scheme and energy level splitting are shown in Fig. 4(a). The 1164-nm laser is first locked to the $F = 5/2 \rightarrow 5/2$ transition, while the 640-nm laser is scanned to determine the exact wavelength for $^{229}\text{Th}^{2+}$. A fluorescence signal is observed as shown in Fig. 4(b), with the yellow color corresponding to the $F = 5/2 \rightarrow 5/2 \rightarrow 5/2$ transition. The FWHM of this transition is approximately 175 MHz.

To observe the other two intermediate levels which lie within the Doppler broadening of the 640-nm transition, the 1164-nm laser is locked to the $7/2 \rightarrow 5/2$ transition, while the 640-nm laser is scanned. The strong observed peak [shown in blue in Fig. 4(b)] corresponds to the $5/2 \rightarrow 7/2 \rightarrow 5/2$ transition, whereas the weak peak corresponds to the $5/2 \rightarrow 3/2 \rightarrow 5/2$ transition. The measured frequency separation for 640 nm between the $5/2 \rightarrow 5/2$ and $5/2 \rightarrow 7/2$ transitions is approximately equal to the applied shift of the 1164-nm laser, that is, 1250 MHz. The frequency difference between the $5/2 \rightarrow 7/2$ and $5/2 \rightarrow 3/2$ transitions is influenced by

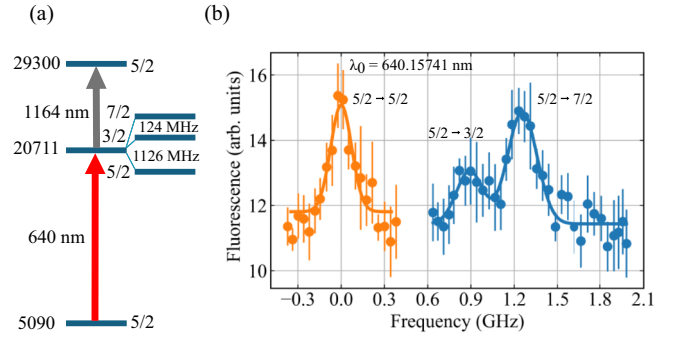


FIG. 4. Fluorescence detection after two-step excitation from the metastable state of $^{229}\text{Th}^{2+}$. The wavelength is given for the $5/2 \rightarrow 5/2$ transition. $F \rightarrow F$ labels are shown for the 640-nm transition. Solid markers represent experimental data with error bars, and the lines correspond to the fitted curve.

the choice of the 1164-nm laser frequency, as both transitions lie within the Doppler broadening. When the 1164-nm laser is locked to the $7/2 \rightarrow 5/2$ transition, the $5/2 \rightarrow 3/2$ transition experiences an extra frequency shift given by $\Delta f_1 = \Delta f_2(1 + f_1/f_2)$, where f_1 and f_2 denote the transition frequencies, in the copropagating geometry [32,33], which corresponds to 350 MHz for Δf_2 of 124 MHz. Here, we also observe a peak shift of approximately 350 MHz between $5/2 \rightarrow 3/2$ and $5/2 \rightarrow 7/2$.

Measurement of the isotope shift of the metastable state in $^{232}\text{Th}^{2+}$ and $^{229}\text{Th}^{2+}$

As a measure of the interaction with the nucleus, we determine the isotope shift ($\nu_{229} - \nu_{232}$) for the forbidden transition 63₂ $\rightarrow$ 5090₀. We start from the known isotope shift for the 63₂ $\rightarrow$ 20711₁ transition, which is 8.2(2) GHz [34]. The hyperfine constants A and B for the level 20711₁ are taken from Ref. [35]. Using these constants, we calculated the position of the centroid frequency relative to the peak $5/2 \rightarrow 5/2$ for the 5090₀ $\rightarrow$ 20711₁ transition for $^{229}\text{Th}^{2+}$. The centroid is found to be shifted by approximately 800 MHz relative to the $5/2 \rightarrow 5/2$ peak shown in Fig. 4(b). From this, the total isotope shift for the 5090₀ $\rightarrow$ 20711₁ transition between $^{232}\text{Th}^{2+}$ and $^{229}\text{Th}^{2+}$ is 8.8(2) GHz at 640 nm. Knowing that the isotope shift for the 63₂ $\rightarrow$ 20711₁ transition is 8.2(2) GHz, the resulting isotope shift for the 63₂ $\rightarrow$ 5090₀ transition is therefore 0.6(3) GHz. This low value of isotope shift arises from the nearly identical electronic configurations of the $6d^2\ ^3F_2$ (63 cm^{-1}) and $6d^2\ ^3P_0$ (5090 cm^{-1}) states and from the small field shift associated with the d -electrons' orbitals in Th^{2+} [34,35].

IV. LIFETIME MEASUREMENTS

In our experiment, the lifetime of the 5090₀ state is determined by collisional mixing with the nearby 5060₃ level. The 5060₃ state decays to the 63₂ level through an electric dipole transition. We monitor the population of the 5060₃ state in fluorescence at 545 nm by exciting a transition with a wavelength of 724 nm to the 18863₂ level as shown in Fig. 2. A pulse of 150 μs in duration of the 484-nm laser drives the

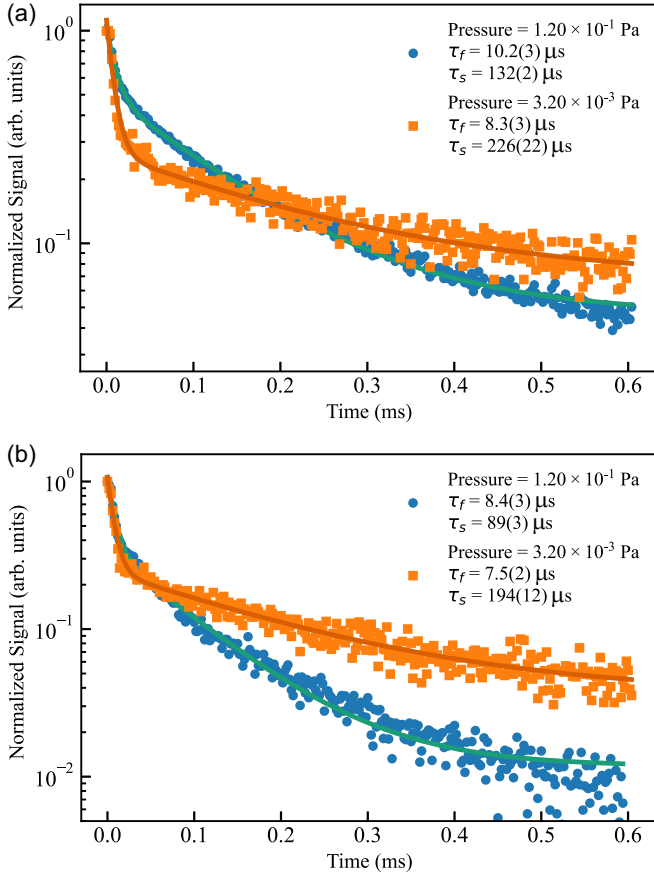


FIG. 5. (a) Decay of the $J = 0$ state in the presence of Ar as buffer gas at two different pressures. The data are fitted with a biexponential function. Here, τ_f and τ_s correspond to the time constants of the fast- and slow-decay components. (b) Decay in the presence of He as buffer gas at two different pressures.

$63_2 \rightarrow 20711_1$ transition, and decay of the 20711_1 state populates the 5090_0 and 5060_3 levels. A 724-nm laser with a power of 10 mW is applied continuously. After turning off the 484-nm pumping laser, the fluorescence signal as shown in Fig. 5 exhibits an initial fast decay followed by a slower decay. The fast decay corresponds to the loss of initial population from 5060_3 , while the slow decay is directly connected to the lifetime of the 5090_0 level which populates the 5060_3 level through collisional mixing. In Fig. 5(a), the data are taken with argon as a buffer gas at two different pressures. We fit the data with a biexponential fit to extract the time constants for fast and slow decay. The time constant for slow decay is determined by the rate of collisional mixing and limits the lifetime in our setup. A detailed study of multiexponential decay is presented in Refs. [36–38]. Detecting the lifetime through the population of 5060_3 allows us to continuously record the fluorescence signal using a scalar card with a time resolution of 100 ns and an averaging rate of 1 kHz. This method is not prone to fluctuations in the initial ion number over longer timescales, which occur in our setup due to molecule formation of Th^{2+} . Similarly, we also performed the measurement with helium as the buffer gas. The result is shown in Fig. 5(b). To understand the behavior of the mixing, we performed measurements at different pressures.

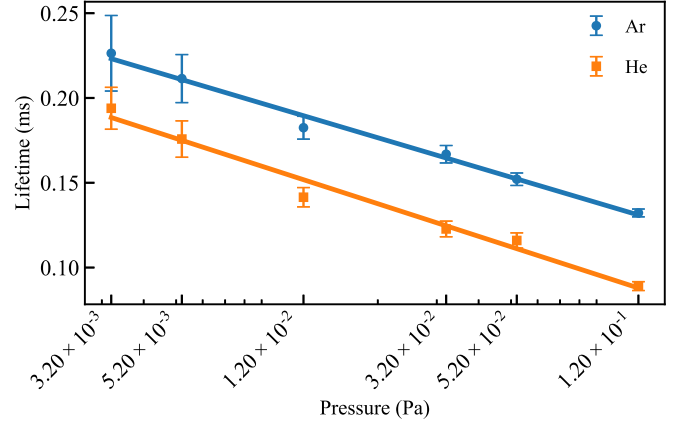


FIG. 6. Lifetime measurement of the $J = 0$ state at different pressures for the buffer gases Ar and He. The lifetime corresponds to the time constant for slow decay (τ_s) as seen in Fig. 5.

A linear decrease in lifetime with $\log(P)$ is observed as shown in Fig. 6. For argon as the buffer gas, collisional mixing is slightly slower than that for helium, resulting in a longer lifetime. A similar difference in collision dynamics has been observed in Ref. [21]. The longest observed lifetime of the 5090_0 state is significantly shorter than the calculated radiative lifetime of the level of approximately 95 s [11]. Our measurement cannot be directly extrapolated to ultrahigh vacuum conditions although the limit arises from collisional mixing [39,40]. Reducing the buffer gas pressure in our setup has the side effect of increasing the temperature of the ions to above room temperature because of radio-frequency heating in the ion trap. From the time constant determined for the ions at room temperature and 0.12 Pa, we may conclude that for laser-cooled ions in ultrahigh-vacuum (10^{-8} Pa), collisional mixing from rare gases is not likely to considerably shorten the lifetime below 95 s as determined from radiative decay.

V. CONCLUSIONS

In this experiment, we presented studies on the metastable state with $J = 0$ in Th^{2+} , demonstrating schemes to efficiently populate the state by laser excitation at 484 nm and to detect it in fluorescence after excitation at 724 nm. We determined an isotope shift of 0.6(3) GHz between $^{232}\text{Th}^{2+}$ and $^{229}\text{Th}^{2+}$, smaller by an order of magnitude than previously reported isotope shifts for other transitions in Th^{2+} . The small isotope shift suggests that the 5090_0 state has minimal interaction with the nuclear charge distribution and identical field shift coefficient as 63_2 [20,34]. We also measured the lifetime at different pressures and observed a maximum lifetime of 225 μs at 3.2×10^{-3} Pa of argon buffer gas. In the present setup, the lifetime is limited by collisional mixing, which would be strongly suppressed for cooled ions in an ultrahigh vacuum.

As an outlook, we propose a hyperfine-free nuclear optical clock by preparing $^{229}\text{Th}^{2+}$ in the long-lived 5090_0 state as shown in Fig. 7. The long lifetime (≈ 95 s) of the 5090_0 state allows long interrogation times of the nuclear transition, whereas the s state with $J = 1/2$ in Th^{3+} is limited to a lifetime of only ~ 600 ms [1,15]. A minor drawback of using

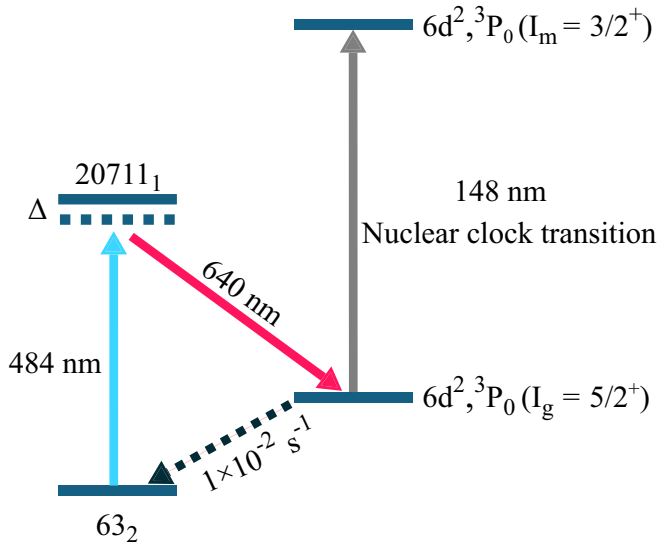


FIG. 7. Simplified level scheme for a hyperfine-free nuclear clock with Th^{2+} . Here 484- and 640-nm lasers drive a coherent Raman transition for state preparation. Δ is the detuning used to suppress off-resonant scattering.

$J = 0$ instead of $J = 1/2$ is that the nuclear transition will be split into Zeeman components with first-order magnetic field sensitivities between ± 0.4 and $\pm 6 \text{ Hz}/\mu\text{T}$. A potential systematic frequency offset can be eliminated by averaging over two Zeeman components with opposite signs of the shift.

A viable experimental setup will consist of sympathetically cooled Th^{2+} with cotrapped laser-cooled ions as it is done for Th^{3+} [41,42]. Sr^+ or In^+ would be suitable species because their charge-to-mass ratio is similar to that of Th^{2+} . To prepare ions in 5090_0 , a stimulated Raman scheme can be used to transfer the ions from 63_2 to 5090_0 via 20711_1 as an intermediate state, also shown in Fig. 7. The lasers at 484 and 640 nm, as used in this work, can enable coherent population transfer through the Raman process with a few milliwatts of optical power [43–45]. Since no electronic levels of Th^{2+} are known

in the vicinity of 9.0-eV excitation energy, applying laser radiation at 148 nm to drive the nuclear transition from the 5090_0 level is not expected to lead to parasitic electronic excitation. The detection of nuclear excitation can be performed with quantum logic spectroscopy via the same cotrapped ions as those used for sympathetic cooling [46,47].

Studying the 8.4-eV nuclear resonance of ^{229}Th in different electronic configurations of atomic ions will provide an opportunity to compare nuclear-electronic hyperfine interactions for a variety of cases. Examples of strong coupling include electronic bridge processes in Th^+ [48] and in Th^{35+} [49,50] and hyperfine-mixing in hydrogenlike Th^{89+} [51,52]. In contrast, the $J = 0$ states in Th^{4+} [18] and Th^{2+} as discussed here are cases where the leading hyperfine interactions of the nucleus with the electron shell are minimized.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [53].

- [1] E. Peik and C. Tamm, Nuclear laser spectroscopy of the 3.5 eV transition in Th -229, *Europhys. Lett.* **61**, 181 (2003).
- [2] C. J. Campbell, A. G. Radnaev, A. Kuzmich, V. A. Dzuba, V. V. Flambaum, and A. Derevianko, Single-ion nuclear clock for metrology at the 19th decimal place, *Phys. Rev. Lett.* **108**, 120802 (2012).
- [3] J. Tiedau, M. V. Okhapkin, K. Zhang, J. Thielking, G. Zitzer, E. Peik, F. Schaden, T. Pronebner, I. Morawetz, L. T. De Col, F. Schneider, A. Leitner, M. Pressler, G. A. Kazakov, K. Beeks, T. Sikorsky, and T. Schumm, Laser excitation of the Th -229 nucleus, *Phys. Rev. Lett.* **132**, 182501 (2024).
- [4] R. Elwell, C. Schneider, J. Jeet, J. E. S. Terhune, H. W. T. Morgan, A. N. Alexandrova, H. B. Tran Tan, A. Derevianko, and E. R. Hudson, Laser excitation of the ^{229}Th nuclear isomeric transition in a solid-state host, *Phys. Rev. Lett.* **133**, 013201 (2024).
- [5] C. Zhang, T. Ooi, J. S. Higgins, J. F. Doyle, L. von der Wense, K. Beeks, A. Leitner, G. A. Kazakov, P. Li, P. G. Thirolf, T. Schumm, and J. Ye, Frequency ratio of the ^{229m}Th nuclear isomeric transition and the ^{87}Sr atomic clock, *Nature (London)* **633**, 63 (2024).
- [6] T. Ooi, J. F. Doyle, C. Zhang, J. S. Higgins, J. Ye, K. Beeks, T. Sikorsky, and T. Schumm, Frequency reproducibility of solid-state thorium-229 nuclear clocks, *Nature (London)* **650**, 72 (2026).
- [7] K. Beloy, Trap-induced ac Zeeman shift of the thorium-229 nuclear clock frequency, *Phys. Rev. Lett.* **130**, 103201 (2023).
- [8] A. Chakraborty and B. K. Sahoo, Comprehensive assessment of Th^{3+} properties for nuclear clock and fundamental physics applications, *Phys. Rev. A* **113**, L020801 (2026).
- [9] S. L. Redman, G. Nave, and C. J. Sansonetti, The spectrum of thorium from 250 nm to 5500 nm: Ritz wavelengths and optimized energy levels, *Astrophys. J. Suppl. Ser.* **211**, 4 (2014).

- [10] E. Biémont, P. Palmeri, P. Quinet, Z. G. Zhang, and S. Svanberg, Doubly ionized thorium: Laser lifetime measurements and transition probability determination of interest in cosmochemistry, *Astrophys. J.* **567**, 1276 (2002).
- [11] M. S. Safronova, U. I. Safronova, and C. W. Clark, Relativistic all-order calculations of Th, Th⁺, and Th²⁺ atomic properties, *Phys. Rev. A* **90**, 032512 (2014).
- [12] J.-F. Wyart and V. Kaufman, Extended analysis of doubly ionized thorium (Th III), *Phys. Scr.* **24**, 941 (1981).
- [13] C. J. Campbell, A. G. Radnaev, and A. Kuzmich, Wigner crystals of ²²⁹Th for optical excitation of the nuclear isomer, *Phys. Rev. Lett.* **106**, 223001 (2011).
- [14] C. J. Campbell, A. V. Steele, L. R. Churchill, M. V. DePalatis, D. E. Naylor, D. N. Matsukevich, A. Kuzmich, and M. S. Chapman, Multiply charged thorium crystals for nuclear laser spectroscopy, *Phys. Rev. Lett.* **102**, 233004 (2009).
- [15] A. G. Radnaev, C. J. Campbell, and A. Kuzmich, Observation of the 717-nm electric quadrupole transition in triply charged thorium, *Phys. Rev. A* **86**, 060501(R) (2012).
- [16] U. I. Safronova, W. R. Johnson, and M. S. Safronova, Excitation energies, polarizabilities, multipole transition rates, and lifetimes in Th IV, *Phys. Rev. A* **74**, 042511 (2006).
- [17] M. S. Safronova, U. I. Safronova, and M. G. Kozlov, Atomic properties of actinide ions with particle-hole configurations, *Phys. Rev. A* **97**, 012511 (2018).
- [18] V. V. Flambaum, V. A. Dzuba, and E. Peik, Nuclear clock based on the Th v ion, *Phys. Rev. A* **112**, 023103 (2025).
- [19] G. Zitzer, J. Tiedau, C. E. Düllmann, M. V. Okhapkin, and E. Peik, Laser spectroscopy on the hyperfine structure and isotope shift of sympathetically cooled ²²⁹Th³⁺ ions, *Phys. Rev. A* **111**, L050802 (2025).
- [20] M. V. Okhapkin, D. M. Meier, E. Peik, M. S. Safronova, M. G. Kozlov, and S. G. Porsev, Observation of an unexpected negative isotope shift in ²²⁹Th⁺ and its theoretical explanation, *Phys. Rev. A* **92**, 020503(R) (2015).
- [21] J. Thielking, M. V. Okhapkin, P. Glowacki, D. M. Meier, L. von der Wense, B. Seiferle, C. E. Düllmann, P. G. Thirolf, and E. Peik, Laser spectroscopic characterization of the nuclear-clock isomer ^{229m}Th, *Nature (London)* **556**, 321 (2018).
- [22] A. Yamaguchi, Y. Shigekawa, H. Haba, H. Kikunaga, K. Shirasaki, M. Wada, and H. Katori, Laser spectroscopy of triply charged ²²⁹Th isomer for a nuclear clock, *Nature (London)* **629**, 62 (2024).
- [23] L. v. d. Wense, B. Seiferle, M. Laatiaoui, and P. G. Thirolf, Determination of the extraction efficiency for ²³³U source α -recoil ions from the MLL buffer-gas stopping cell, *Eur. Phys. J. A* **51**, 29 (2015).
- [24] K. Beeks, T. Sikorsky, T. Schumm, J. Thielking, M. V. Okhapkin, and E. Peik, The thorium-229 low-energy isomer and the nuclear clock, *Nat. Rev. Phys.* **3**, 238 (2021).
- [25] V. A. Dzuba and V. V. Flambaum, Using the Th iii ion for a nuclear clock and searches for new physics, *Phys. Rev. A* **111**, 053109 (2025).
- [26] V. I. Yudin, A. V. Taichenachev, O. N. Prudnikov, M. Y. Basalae, A. N. Goncharov, S. V. Chepurov, and V. G. Pal'chikov, Optical clock based on two-photon spectroscopy of the nuclear transition in a ²²⁹Th ion in a monochromatic field, *JETP Lett.* **121**, 345 (2025).
- [27] O. A. Herrera-Sancho, M. V. Okhapkin, K. Zimmermann, C. Tamm, E. Peik, A. V. Taichenachev, V. I. Yudin, and P. Glowacki, Two-photon laser excitation of trapped ²³²Th⁺ ions via the 402-nm resonance line, *Phys. Rev. A* **85**, 033402 (2012).
- [28] O. A. Herrera-Sancho, N. Nemitz, M. V. Okhapkin, and E. Peik, Energy levels of Th⁺ between 7.3 and 8.3 eV, *Phys. Rev. A* **88**, 012512 (2013).
- [29] P. Staunum, I. S. Jensen, R. G. Martinussen, D. Voigt, and M. Drewsen, Lifetime measurement of the metastable $3d\ ^2D_{5/2}$ state in the ⁴⁰Ca⁺ ion using the shelving technique on a few-ion string, *Phys. Rev. A* **69**, 032503 (2004).
- [30] M. Fan, C. A. Holliman, A. Contractor, C. Zhang, S. F. Gebretsadkan, and A. M. Jayich, Measurement of the Ra⁺ $7p\ ^2P_{3/2}$ state lifetime, *Phys. Rev. A* **105**, 042801 (2022).
- [31] B. A. Palmer and R. J. Engleman, Atlas of the thorium spectrum, Technical Report No. LA-9615, Los Alamos National Laboratory (LANL), Los Alamos, NM, 1983.
- [32] J. E. Bjorkholm and P. F. Liao, Line shape and strength of two-photon absorption in an atomic vapor with a resonant or nearly resonant intermediate state, *Phys. Rev. A* **14**, 751 (1976).
- [33] E. C. Cook, A. D. Vira, and W. D. Williams, Resonant two-photon spectroscopy of the $2s3d\ ^1D_2$ level of neutral ⁹Be, *Phys. Rev. A* **101**, 042503 (2020).
- [34] M. S. Safronova, S. G. Porsev, M. G. Kozlov, J. Thielking, M. V. Okhapkin, P. Glowacki, D. M. Meier, and E. Peik, Nuclear charge radii of ²²⁹Th from isotope and isomer shifts, *Phys. Rev. Lett.* **121**, 213001 (2018).
- [35] R. A. Müller, A. V. Maiorova, S. Fritzsche, A. V. Volotka, R. Beerwerth, P. Glowacki, J. Thielking, D.-M. Meier, M. Okhapkin, E. Peik, and A. Surzhykov, Hyperfine interaction with the ²²⁹Th nucleus and its low-lying isomeric state, *Phys. Rev. A* **98**, 020503(R) (2018).
- [36] D. P. Moehs and D. A. Church, Magnetic dipole transition rates from measured lifetimes of levels of Be-like and B-like argon ions, *Phys. Rev. A* **58**, 1111 (1998).
- [37] E. Träbert, Atomic lifetime measurements with ion traps of many sizes, *Phys. Scr.* **61**, 257 (2000).
- [38] E. Träbert, Limits to the precision of atomic lifetime measurements, *Atoms* **12**, 73 (2024).
- [39] M. A. Gearba, J. H. Wells, P. H. Rich, J. M. Wesemann, L. A. Zimmerman, B. M. Patterson, R. J. Knize, and J. F. Sell, Collisional excitation transfer and quenching in Rb (⁵P)-methane mixtures, *Phys. Rev. A* **99**, 022706 (2019).
- [40] M. Endo, H. Nagaoka, and F. Wani, Collisional mixing and quenching cross sections of Cs 6^2P levels with methane, ethane, and propane, *Opt. Express* **29**, 42887 (2021).
- [41] G. Zitzer, J. Tiedau, M. V. Okhapkin, K. Zhang, C. Mokry, J. Runke, C. E. Düllmann, and E. Peik, Sympathetic cooling of trapped Th³⁺ alpha-recoil ions for laser spectroscopy, *Phys. Rev. A* **109**, 033116 (2024).
- [42] D. Moritz, K. Scharl, M. Wiesinger, G. Holthoff, T. Teschler, M. I. Hussain, J. R. Crespo López-Urrutia, T. Dickel, S. Ding, C. E. Düllmann, E. R. Hudson, S. Kraemer, L. Löbell, C. Mokry, J. Runke, B. Seiferle, L. von der Wense, F. Zacherl, and P. G. Thirolf, A cryogenic Paul trap for probing the nuclear isomeric excited state ^{229m}Th³⁺, *Eur. Phys. J. D* **79**, 127 (2025).

- [43] J. Spahn, H. Bodnar, K. König, and W. Nörtershäuser, Demonstration of a Raman velocity filter in collinear laser spectroscopy: Towards applications for sub-ppm high-voltage measurements, *New J. Phys.* **28**, 053202 (2026).
- [44] S. P. Carman, J. Rudolph, B. E. Garber, M. J. Van de Graaff, H. Swan, Y. Jiang, M. Nantel, M. Abe, R. L. Barclay, and J. M. Hogan, Collinear three-photon excitation of a strongly forbidden optical clock transition, *Phys. Rev. X* **15**, 031051 (2025).
- [45] H. Levine, D. Bluvstein, A. Keesling, T. T. Wang, S. Ebadi, G. Semeghini, A. Omran, M. Greiner, V. Vuletić, and M. D. Lukin, Dispersive optical systems for scalable Raman driving of hyperfine qubits, *Phys. Rev. A* **105**, 032618 (2022).
- [46] P. O. Schmidt, T. Rosenband, C. Langer, W. M. Itano, J. C. Bergquist, and D. J. Wineland, Spectroscopy using quantum logic, *Science* **309**, 749 (2005).
- [47] P. Mücke, T. Leopold, S. A. King, E. Benkler, L. J. Spieß, L. Schmöger, M. Schwarz, J. R. Crespo López-Urrutia, and P. O. Schmidt, Coherent laser spectroscopy of highly charged ions using quantum logic, *Nature (London)* **578**, 60 (2020).
- [48] Y. Shigekawa, A. Yamaguchi, K. Tokoi, N. Sato, H. Kikunaga, K. Shirasaki, Y. Kasamatsu, M. Wada, and H. Haba, Lifetime of the singly charged ^{229}Th nuclear isomer, *Nat. Phys.* **22**, 832 (2026).
- [49] P. V. Bilous, H. Bekker, J. C. Berengut, B. Seiferle, L. von der Wense, P. G. Thirolf, T. Pfeifer, J. R. C. López-Urrutia, and A. Pálffy, Electronic bridge excitation in highly charged ^{229}Th ions, *Phys. Rev. Lett.* **124**, 192502 (2020).
- [50] S. G. Porsev, C. Cheung, and M. S. Safronova, Low-lying energy levels of $^{229}\text{Th}^{35+}$ and the electronic bridge process, *Quantum Sci. Technol.* **6**, 034014 (2021).
- [51] F. F. Karpeshin, S. Wycech, I. M. Band, M. B. Trzhaskovskaya, M. Pfützner, and J. Żylicz, Rates of transitions between the hyperfine-splitting components of the ground-state and the 3.5 eV isomer in $^{229}\text{Th}^{89+}$, *Phys. Rev. C* **57**, 3085 (1998).
- [52] K. Pachucki, S. Wycech, J. Żylicz, and M. Pfützner, Nuclear-spin mixing oscillations in $^{229}\text{Th}^{89+}$, *Phys. Rev. C* **64**, 064301 (2001).
- [53] S. S. Maurya, V. Lal, J. Tiedau, M. Okhapkin, and E. Peik, J_0_clock_Th(2p)_Maurya_2026, Zenodo, 2026, <https://doi.org/10.5281/zenodo.21266298>.