TOWARDS AN OPTICAL NUCLEAR CLOCK WITH THORIUM-229

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Abstract

We report experimental results towards optical excitation of the low-lying isomer level in the ^{229}Th nucleus, a promising system for use in precise optical clocks and tests of fundamental physics. We have produced laser-cooled Wigner crystals of $^{229}Th^{3+}$ in a linear Paul trap, providing a convenient system to search for this nuclear transition and to utilize its potential with high precision spectroscopy. Furthermore, we have measured isotope shifts, magnetic dipole (A) and electric quadrupole (B) hyperfine constants for four low-lying electronic levels, deducing a new value of the spectroscopic nuclear electric quadrupole moment Q = 3.11(16) eb. These measurements provide a reference for evaluation of the sensitivity of the nuclear transition to the variation of the fundamental constants including the fine structure constant.

I. INTRODUCTION

Clock performance is determined by its oscillator quality factor and isolation from external perturbations. Nuclear isomer transitions offer supreme values of both of these factors, however keV to MeV excitation energies make them unavailable due to a dearth of the corresponding coherent radiation. A notable exception is ^{229}Th nucleus where the energy splitting of the ground state doublet is less than 10 eV **[1–4]**. This fortuitous coincidence offers prospects for a better clock with potential for 10^{-19} accuracy **[5]** and for a test of temporal variation of fundamental constants at an unprecedented level of precision **[6,7]**. This nuclear transition has not yet been directly observed.

Here, we report an important step towards the goals – producing laser-cooled Wigner crystals of $^{229}Th^{3+}$ ions in a linear Paul trap, where the ultra-cold ions can be interrogated on an hour-scale. The monovalent character of the $^{229}Th^{3+}$ ion is favorable for the isomer level search based on an electron-bridge process **[8]**. Once the isomer energy value is measured, either in our system or using one of other approaches **[9,10]**, a single trapped, cold $^{229}Th^{3+}$ ion is expected to be an ideal system to take advantage of this remarkable nucleus. The level structure of ^{229}Th is in Figure 1.

II. ENHANCED SENSITIVITY TO FUNDAMENTAL CONSTANT VARIATION

Importantly, the ground-isomer nuclear transition may be used to search for temporal variation of fundamental constants by monitoring the ratio of the frequencies of the nuclear transition and that of an atomic (electronic) clock transition, e.g., in Al+, Sr, or in the same $^{229}Th^{3+}$ ion to mitigate systematic frequency shifts. Flambaum et al. have predicted that such a frequency ratio is likely to be supremely sensitive to relative variation of the strong interaction parameter and the fine structure constant, with

enhancement factor K possibly reaching as high as 10^6 **[7,11,12]**. This enhancement is due to the fact that nuclear energy changes on MeV scale, compared to eV atomic energy scale. Although the complexity of this nuclear system makes a theoretical evaluation of K difficult **[13,14]**, a reliable empirical determination of the enhancement factor **[15]** can be obtained from spectroscopy. The sensitivity of the nuclear transition frequency to α variation is proportional to the change in the nuclear Coulomb energy (ΔV_C) between the ground (g) and isomer (m) states **[15]**:

$$\frac{\Delta V_C}{(\mathrm{MeV})} \approx -506\frac{\Delta\langle r^2\rangle}{\langle r^2\rangle} + 23\frac{\Delta Q_0}{Q_0}$$

where $\Delta\langle r^2\rangle = \langle r_m^2\rangle - \langle r_g^2\rangle$ is the change in the nucleus radius, $\Delta Q_0 = Q_m - Q_g$ is the change in the nuclear electric quadrupole moment. These terms can be evaluated with optical and microwave spectroscopy of the ground and isomer level manifolds by measuring the isomeric field shift and hyperfine constant coefficients, as demonstrated here for measurements of Q_g and ^{229}Th - ^{232}Th relative isotope shifts. These measurements should provide a reliable empirical determination of the enhancement factor with little or no dependence on the nuclear model assumed.

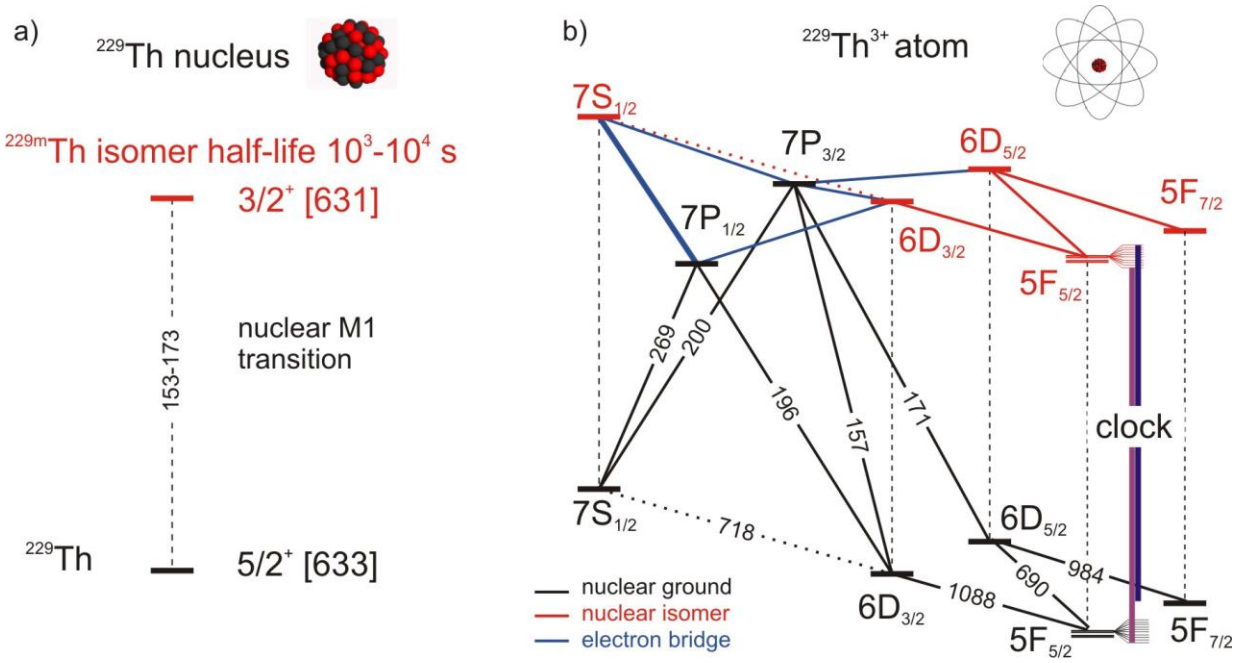

Figure 1. Level structures of ^{229}Th. a) nuclear ground and isomer energy levels (Nilsson classification quantum numbers). b) combined nuclear and electronic energy levels of $^{229}Th^{3+}$. Optical transition wavelengths are in nm.

III. LASER COOLING AND TRAPPING OF ^{229}Th

The first laser cooling and trapping of thorium, as well as first laser cooling and trapping of multiply charged ions, were done with $^{232}Th^{3+}$ **[8]**. This ^{232}Th isotope is naturally abundant, mildly radioactive and has zero nuclear spin, hence ^{232}Th atoms have no hyperfine structure, simplifying laser cooling. To the contrary, the isotope of interest, ^{229}Th, is highly radioactive, not naturally occurring, and has non-zero nuclear spins of 5/2 and 3/2 for the ground and isomer nuclear states, respectively. These factors dramatically complicate laser cooling and trapping, since only trace amounts of ^{229}Th are available and

ground hyperfine levels require a multitude of laser light fields to avoid dark states (Figure 2). Moreover, prior to the measurements performed here, the hyperfine structure was not known, prohibiting direct laser cooling.

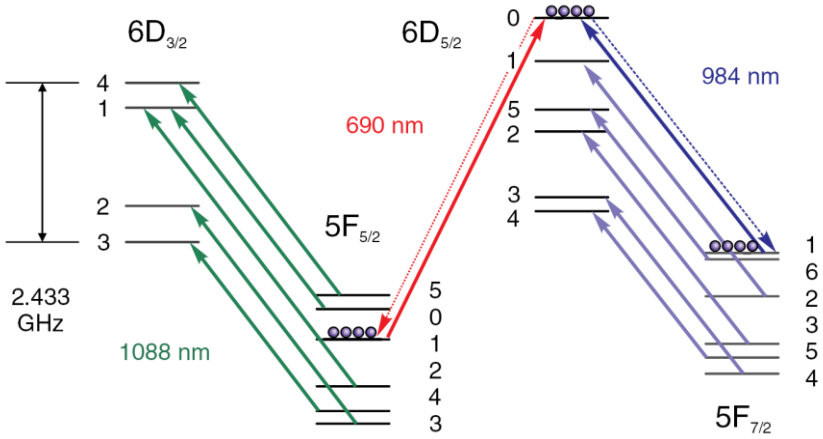

Figure 2. Level structure of four low-lying electronic levels of $^{229}Th^{3+}$ and light fields for laser cooling and imaging. Numbers near the levels represent total angular momentum quantum numbers.

The experimental apparatus is shown in Figure 3. A typical protocol starts with ablation of the nitrate salt of ^{229}Th (Figure 3a) with a 10 ns long 100 µJ laser pulse at 355 nm. The generated plume contains $^{229}Th^{3+}$ (70%) and $^{232}Th^{3+}$ (30%) ions which propagate at about 10 km/s axially into a linear Paul trap, where they are trapped and laser cooled. Details are provided in Reference **[16]**. Ions are illuminated with light fields as shown in Figure 3b. The ion imaging system collects fluorescence at 984 nm with ~7% efficiency to a low-noise CCD camera. Images of various Wigner crystals of laser-cooled $^{229}Th^{3+}$ and $^{232}Th^{3+}$ ions are shown in Figure 4.

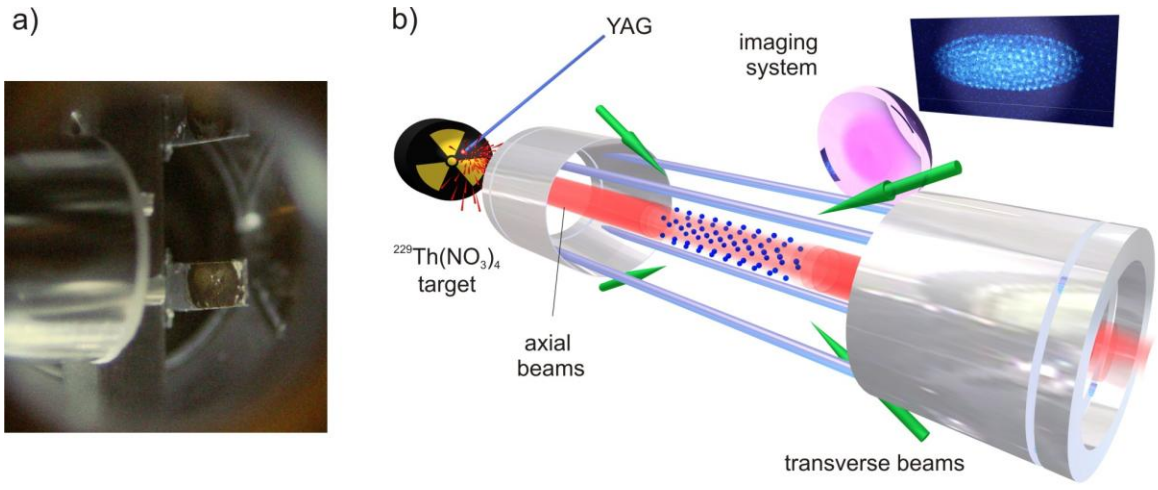

Figure 3. Experimental setup. a) Photograph of the ^{229}Th nitrate salt (brown patch) at the ion trap entrance. The white spots in the center are ablation craters. The radioactivity level of the 20 µg sample of ^{229}Th is 150 kBq and its half-life is 7340 years. b) Essential elements of the experiment (see text for details).

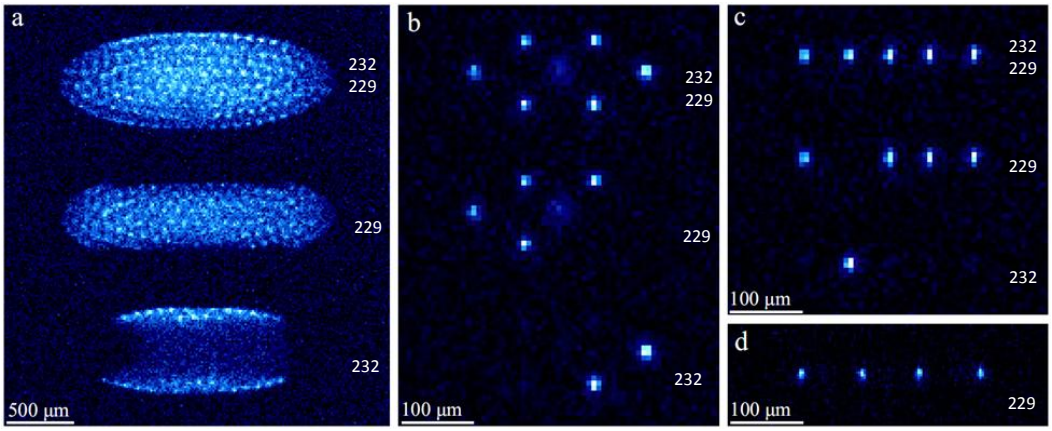

Figure 4. Trapped $^{229}Th^{3+}$ and $^{232}Th^{3+}$ ions, laser-cooled to crystallization. The larger mass-to-charge ratio of $^{232}Th^{3+}$ causes its radial accumulation in the outer shells of the crystal. The short focal depth of the imaging system is apparent as most of the $^{232}Th^{3+}$ shell is out of focus.

IV. MEASUREMENT OF $^{229}Th^{3+}$ HYPERFINE STRUCTURE AND ^{229}Th NUCLEAR ELECTRIC QUADRUPOLE MOMENT

Theoretical calculations **[15]** and previous experimental data **[8, 18]** provided $^{229}Th^{3+}$ electronic transition frequencies with 2 GHz accuracy, insufficient for laser cooling. Therefore, helium buffer gas is used to cool the ions close to room temperature, allowing for several transitions to be identified with 100 MHz accuracy. Utilizing laser cooling at these transitions with sympathetic cooling provided by laser-cooled $^{232}Th^{3+}$, the ultra-cold regime is reached and most of the remaining $^{229}Th^{3+}$ lines are measured with 5 MHz accuracy. Furthermore, the magnetic dipole (A) and electric quadrupole (B) hyperfine coefficients of all four levels and the relative isotope shifts of all three transitions were directly extracted from the recorded data via a global least-squares fit. Results of the fit are presented in Table 1.

Table 1. Measured $^{229}Th^{3+}$ hyperfine constants A and B and relative isotope shifts from $^{232}Th^{3+}$. All units are MHz and all uncertainties are one sigma.

Valence Orbital	A	B	Transition	Isotope Shift
$5F_{5/2}$	82.2(6)	2269(6)	$5F_{5/2} \leftrightarrow 6D_{3/2}$	−9856(10)
$5F_{7/2}$	31.4(7)	2550(12)	$5F_{5/2} \leftrightarrow 6D_{5/2}$	−10 509(7)
$6D_{3/2}$	155.3(12)	2265(9)	$5F_{7/2} \leftrightarrow 6D_{5/2}$	−9890(9)
$6D_{5/2}$	−12.6(7)	2694(7)		

Overall, about 20% of the uncertainty is statistical for each extracted value, while the remaining 80% is due to uncertainties in Zeeman splittings combined with optical pumping effects, laser cooling dynamics, and light shifts. By taking the unweighted average ratio of the four measured B's to the corresponding calculated electronic hyperfine matrix elements of Reference **[14]**, a value of the spectroscopic nuclear electric quadrupole moment Q_g = 3.11(16) eb is obtained. Here, the uncertainty is determined by the few percent accuracy of the atomic structure calculations. No accurate calculations are currently available to extract the value of the nuclear magnetic dipole moment, although they are possible **[17]**. Our value for Q_g may be compared with the commonly cited value of 4.3 eb ± 20% measured in Th^+ **[18]**, and the post-

factum discovered uncommonly cited value of 3.149(32) measured with Coulomb excitation of the ^{229}Th **[19]**.

V. SEARCH FOR THE NUCLEAR TRANSITION WITH ELECTRON-BRIDGE

The most recent and precise published measurement of the ^{229}Th isomer energy is 7.6(5) eV **[4]**. In order to span ±3 σ in the search for this nuclear level, direct optical excitation of the isomer in trapped cold ions may not be a viable method, given available UV sources and the large energy uncertainty. Instead, the electron-bridge (EB) process may be utilized **[8]**, as shown in Figure 5. In this case, hyperfine-induced mixing of the ground and the isomer nuclear manifolds opens electric-dipole transitions between the two. The mixing is expected to be the strongest for the S-electronic states, as the electron probability density at the nucleus is highest. For example, considering only the $7S_{1/2}$ and $8S_{1/2}$ electronic orbitals in first-order perturbation theory,

$$\overline{|7S_{1/2}, m\rangle} \approx |7S_{1/2}, m\rangle + \frac{\langle 8S_{1/2}, g|H_{\text{int}}|7S_{1/2}, m\rangle}{E_{7S,m} - E_{8S,g}}|8S_{1/2}, g\rangle,$$

where H_{int} is the electron-nucleus interaction Hamiltonian and $g(m)$ indicates the nuclear ground (isomer) level. The $|8S_{1/2}, g\rangle$ admixture, with expected amplitude of order $\eta \sim 10^{-5}$ **[20]**, couples to the $|7P_{1/2}, g\rangle$ level via electric-dipole radiation of frequency $(E_{7S,\,m} - E_{7P_{1/2},\,g})/\hbar$ (see Figure 1). This shifts the spectral interrogation region from the challenging 130–200 nm range to the manageable 250–800 nm range where high-power coherent light sources are available. It should be noted that this electron-bridge transition is predicted to be substantially stronger than the nuclear M1 transition **[20]**.

In general, the optical search time required to find the isomer transition energy can be estimated as $T \sim \Delta\omega/\Omega^2$, where Ω is the resonant optical excitation Rabi frequency and $\hbar\Delta\omega$ is the energy range over which the search is conducted. Considering the $|7P_{1/2}, g\rangle \leftrightarrow |7S_{1/2}, m\rangle$ electron bridge, an effective electric-dipole moment d_{EB} is predicted to be ~ 2 x $10^{-5} ea_0$ **[20]**. For an ion in the $|7P_{1/2}, g\rangle$ level, placed at the center of a Gaussian light beam of power P and waist r_0, $\Omega^2 = (4Pd_{\text{EB}}^2)/(\pi\varepsilon_0 c r_0^2)$ so that $T \sim (\pi\varepsilon_0 c r_0^2 \hbar\Delta\omega)/(4Pd_{\text{EB}}^2)$. The spectral range in which this transition is likely to exist (250–800 nm) is completely accessible with a mode-locked Ti:sapphire oscillator at the fundamental (1 W), second (150 mW), and third (20 mW) harmonics, and with an optical parametric oscillator in the visible spectrum (200 mW). By focusing ultrafast excitation pulses from these or similar light sources to a waist $r_0 \sim 5$ μm and onto a single crystallized $^{229}Th^{3+}$ ion, we estimate that the isomer level can be excited from the $|7P_{1/2}, g\rangle$ level in tens of hours of ion illumination. Alternatively, $|7S_{1/2}, g\rangle \leftrightarrow |7S_{1/2}, m\rangle$ two-photon excitation through the $|7P_{1/2}, g\rangle$ level may be a suitable approach for the search. It should be noted that the ionization potential of Th^{3+} is 29 eV, making multiphoton ionization with these protocols unlikely.

Once the isomer level is found, a single ion within a linear crystallized chain [Figures 4(c) and 4(d)] can been excited to the isomer manifold and its hyperfine structure and isomeric level shifts may be accurately measured. This knowledge would allow both for unambiguous identification of the isomer level and for an empirical determination of the isomer transition sensitivity to α variation **[14]**.

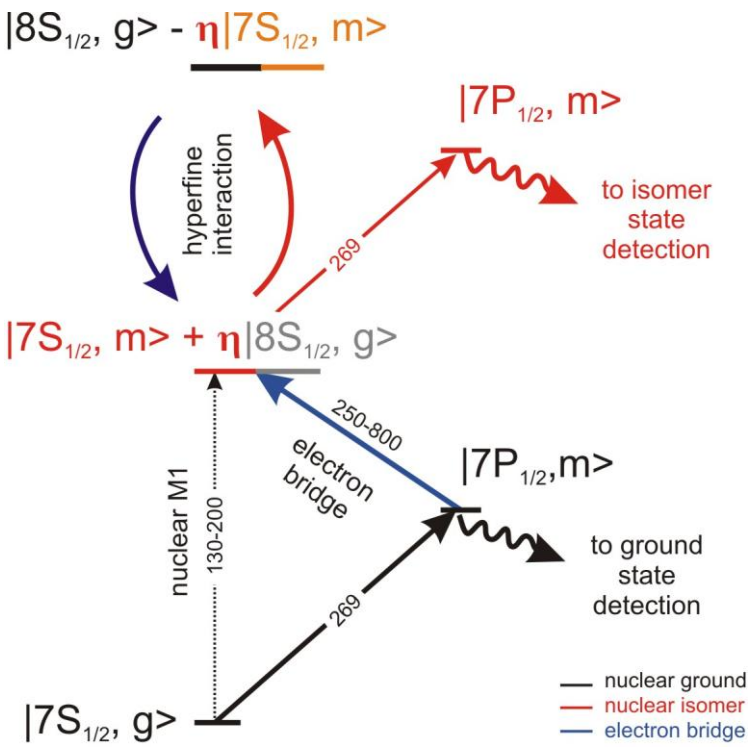

Figure 5. Direct nuclear M1 and electron-bridge isomer level excitation schemes (details in text).

VI. CONCLUSION

In summary, we have produced laser-cooled crystals of triply charged ^{229}Th in a linear Paul trap. Our work opens an avenue toward location and precise measurement of the nuclear transition energy in a single trapped, cold ion. Laser excitation of this nuclear transition may lead to new levels of precision in timekeeping and tests of temporal variation of fundamental constants.

VII. REFERENCES

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