'PHASE TRANSITION'
IN THE 'THORIUM-ISOMER STORY'*

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Given the drastic progress achieved during recent years in our knowledge on the decay and nuclear properties of the thorium isomer ^{229m}Th, the focus of research on this potential nuclear clock transition will turn in the near future from the nuclear physics driven 'search and characterization phase' towards a laser physics driven 'consolidation and realization phase'. This prepares the path towards the ultimate goal of the realization of a nuclear frequency standard, the 'nuclear clock'. This article briefly summarizes our present knowledge, focusing on recent achievements, and points to the next steps envisaged on the way towards the nuclear clock.

1. Introduction

The 'thorium isomer' in ^{229}Th denotes the first excited state in ^{229}Th, an exotic singularity in the landscape of presently more than 3 300 known isotopes with their currently more than 184 000 excited nuclear states. The thorium isomer ^{229m}Th exhibits the lowest nuclear excitation in all presently known nuclides, 4 to 5 orders of magnitude lower than typical nuclear excitation energies. Figure 1 summarizes our present knowledge of the nuclear properties of the ^{229}Th ground and isomeric first excited state.

Early on, the wide application potential arising from the unique properties of the thorium isomer for various physics disciplines such as "optics, solid state physics, lasers, plasma physics, and others" [1] was realized. Amongst

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* Presented at the XXXVI Mazurian Lakes Conference on Physics, Piaski, Poland, September 1–7, 2019.
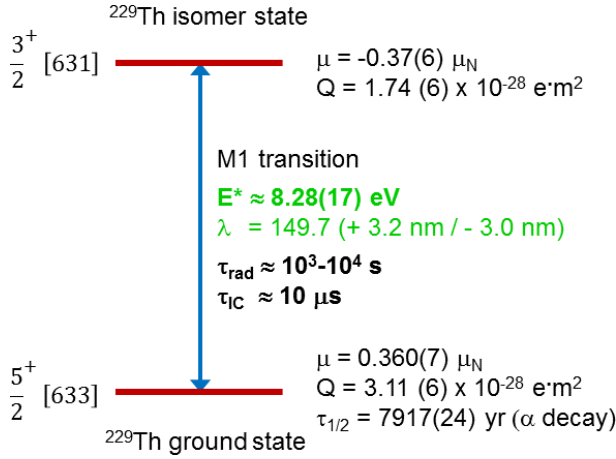
^{229}Th isomer state

$\frac{3}{2}^{+}$ [631]

$\mu = -0.37(6)\ \mu_N$
$Q = 1.74\ (6) \times 10^{-28}\ e{\cdot}m^2$

M1 transition

$E^* \approx 8.28(17)$ eV
$\lambda = 149.7$ (+ 3.2 nm / - 3.0 nm)
$\tau_{rad} \approx 10^3\text{-}10^4$ s
$\tau_{IC} \approx 10\ \mu s$

$\frac{5}{2}^{+}$ [633]

$\mu = 0.360(7)\ \mu_N$
$Q = 3.11\ (6) \times 10^{-28}\ e{\cdot}m^2$
$\tau_{1/2} = 7917(24)$ yr (α decay)

^{229}Th ground state

Fig. 1. Properties of the ^{229}Th ground and isomeric first excited state. Nuclear levels are given with their spin, parity and the Nilsson classification. μ: magnetic moment, Q: electric quadrupole moment.

them frequency metrology plays a central role, as the thorium isomer is placed in the energy and half-life region of atomic transitions exploited for high-precision optical atomic clocks. Its small relative natural linewidth of $\Delta E/E \approx 10^{-20}$ (derived from the expected long half-life of a few 10^3 seconds [2]) and the about 5 orders of magnitude smaller nuclear electromagnetic moments compared to those of atoms render the thorium isomer a natural candidate for a highly precise nuclear frequency standard, a nuclear clock, which was first proposed by Peik and Tamm in 2003 [3].

Starting from its first indirect conjecture from γ spectroscopic data in 1976 [4], intensive experimental efforts were dedicated towards a direct detection of the thorium isomer and improved knowledge of its properties, in order to be able to exploit its unique potential for applications as a nuclear frequency standard, in geodesy or for fundamental studies like the identification of potential variations of fundamental constants like the fine structure constant. Details can be found in the review articles [5–7].

2. Current knowledge on the properties of the nuclear clock isomer ^{229m}Th

The last few years have seen several major experimental breakthroughs in our understanding of the exotic thorium isomer, starting with the first direct detection of the isomer's ground-state decay in the internal conversion (IC) decay channel published in 2016 [8], the first determination of the lifetime of the neutral thorium isomer in 2017 [9], the first determination of the

hyperfine structure of ^{229m}Th via collinear laser spectroscopy [10] to the most recent milestone of the first direct measurement of the thorium isomer's excitation energy, again via the IC decay channel [11]. All of these results were based on the production of an isotopically pure beam of $^{229(m)}$Th ions (from the α decay of a ^{233}U source), extracted from a buffer gas cell and purified by a quadrupole mass separator [12].

Here, the most recent achievement of a direct decay energy measurement will be described in more detail. The basic idea is to measure the kinetic energy of the electrons emitted during the IC decay and to derive the excitation energy of ^{229m}Th from there. In order to avoid the influence of surface effects, neutralization of the ^{229m}Th ions followed by the IC decay (occurring within few microseconds by emitting an electron) was initiated by sending the ions through a bi-layer of thin graphene foils (set at -300 V). For these measurements, the 'standard' setup used to generate a ^{229m}Th ion beam was complemented by a magnetic-bottle-type retarding field electron spectrometer. The experimental setup can be seen in Fig. 2, in addition to the schematic overview, a photograph of the home-built retarding field electron spectrometer can be seen in the inset.
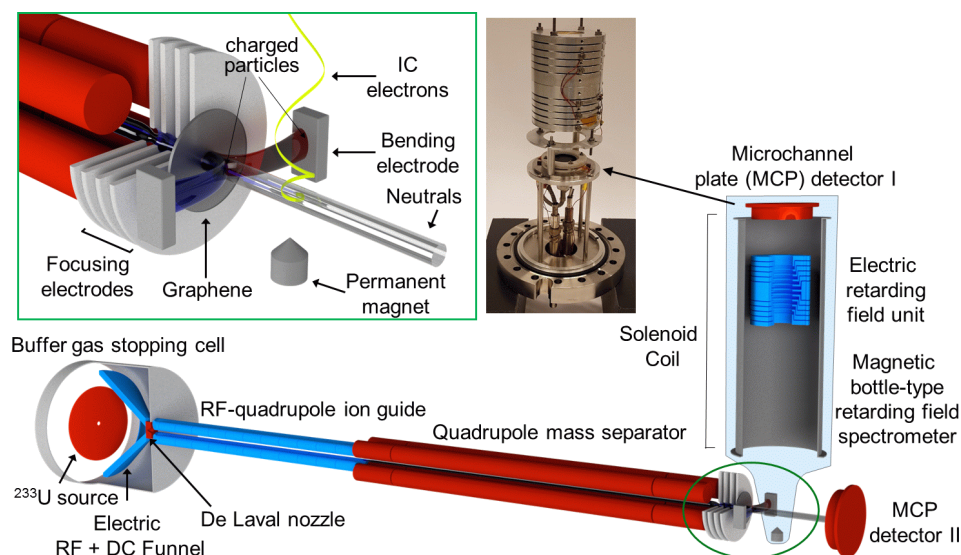

Fig. 2. Experimental setup for the determination of the thorium isomer's excitation energy, complementing the buffer-gas cell based 'isomer generator' with a magnetic bottle electron spectrometer (retarding field unit shown as photograph in the inset). Adapted from Ref. [11].

The spectrometer [13] consisted of a strong permanent magnet that generated an inhomogeneous magnetic field (about 200 mT in the area above the magnet) and a solenoid coil creating a weak homogeneous transport field (typically 2 mT). Internal conversion electrons emitted in a spherical volume with a radius of about 1 mm above the permanent magnet were collected by the magnetic field and directed into the solenoidal transport field and guided towards a retarding-field unit placed in the solenoid coil. The kinetic energy of the IC electrons can be analyzed by applying a retarding voltage to a gold grid (surrounded by ring electrodes to ensure a smooth gradient and terminated by additional gold grids) and by counting the electrons that reach the multichannel-plate electron detector (MCP I in Fig. 2). Only electrons with kinetic energies sufficient to overcome the applied retarding voltage were registered with MCP I. This resulted in an integrated spectrum that decreased monotonically with increasing retarding voltage. The spectrometer reached an excellent relative energy resolution $\Delta E/E$ (FWHM) of about 3% (*ca.* 30 meV for 1 eV electrons).

For the determination of the thorium isomer's excitation energy, it is most important to notice that the neutralization process will typically end in excited electronic states E_i of the neutral ^{229}Th atom, while the subsequent IC decay leads to excited electronic states E_f of the resulting thorium ion. Thus, the kinetic energy of an IC electron, E_{kin}, is connected to the energy of the isomer, E_{is}, via

$$E_{kin} = E_{is} - IP + E_i - E_f, \qquad (2.1)$$

where IP is the thorium ionization potential (6.308 ± 0.003 eV [14]). The transition rate for specific (E_i, E_f) pairs can be precisely predicted [15].

Density functional theory (DFT) calculations allow to identify the relevant energy range for E_i (wavenumbers below $20\,000$ cm^{-1}), where atomic theory identified 82 excited electronic states, E_i, in thorium, each of them contributing (with unknown statistical population probability) to the deexcitation via IC to about four final states E_f [11]. Given the non-observation of clear transition lines in the integrated electron energy spectrum and the instrumental energy resolution of about 30 meV, it could be concluded that at least 5 initial states must have contributed to the measured electron spectrum. This number was taken as input for a statistical analysis based on simulated energy spectra using random distributions of initial electronic states. The details of the method that was used is comprehensively described in Ref. [11], the analysis resulted in the presently most precise value for the excitation energy of the thorium isomer of 8.28 ± 0.17 eV (corresponding to a wavelength of 149.7 ± 3.1 nm).

Figure 3 gives an overview on the temporal development of our knowledge of the ^{229m}Th excitation energy from its conjecture in 1976 to the recently determined most precise value (highlighted by circle).
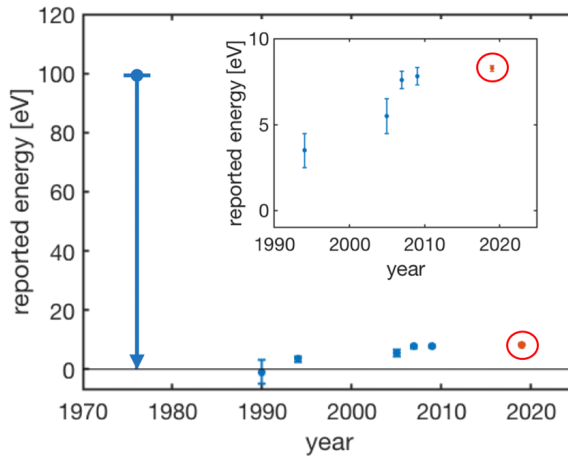

Fig. 3. Improvement of our knowledge on the excitation energy of the thorium isomer ^{229m}Th from its conjecture in 1976 to the recently determined most precise value from Ref. [11] (encircled).

Having achieved a first direct determination of the thorium isomer's ground-state transition energy with 3-fold improved precision compared to the value adopted during the last decade only marks an intermediate step towards the ultimate goal of realizing a nuclear clock, as will be further outlined in the next section.

3. Perspectives towards a nuclear clock

Figure 4 illustrates the challenges lying still ahead on the way towards a high-precision nuclear clock: having recently improved the precision of the ^{229m}Th isomeric excitation energy still leaves us about 14 orders of magnitude away from the ultimate goal of an ultra-high precision nuclear frequency standard on the Hertz level. However, a clear roadmap can be depicted that puts this seemingly tremendous difference into a realistic scenario for realization within the next 5–6 years, staged in 3 phases.

After a period of more than 10 years where no improvement on the isomeric excitation energy 7.8(5) eV [16, 17] could be achieved, as presented before, a first direct measurement with a precision improvement by about a factor of three resulted in the currently best value of 8.28(17) eV [11]. In view of the dynamic activities presently dedicated to research on ^{229m}Th in many groups worldwide, it can be expected that a further improvement via γ or
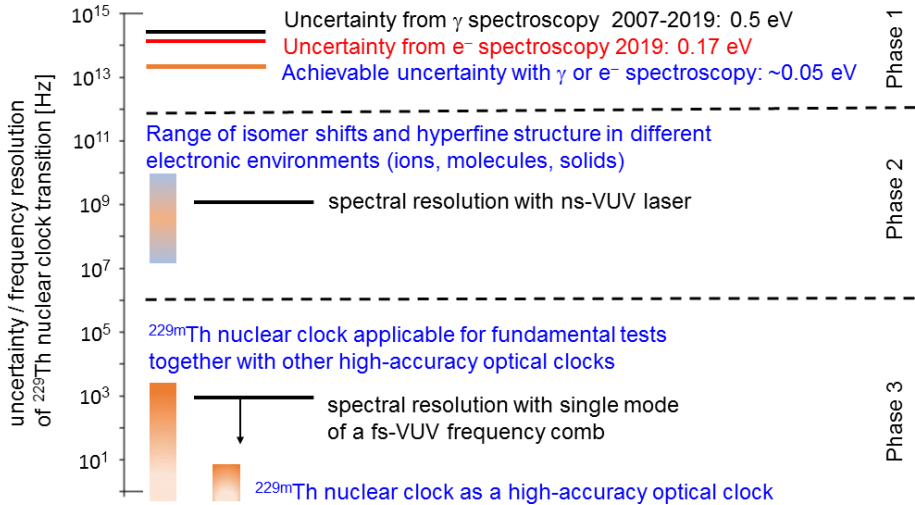
Fig. 4. Frequency scale of uncertainty and spectral resolution of the nuclear ground-state transition frequency from the thorium isomer ^{229m}Th.

electron spectroscopy can further reduce the uncertainty to about 0.05 eV as the endpoint of what can be regarded as the still nuclear-physics dominated 'phase 1' of experimental efforts. This phase will also see extensive efforts to detect VUV photons following non-resonant nuclear excitation. Moreover, the still unknown lifetime of the ionic isomer can be experimentally targeted in a cryogenic Paul trap, which has been set up and is presently under commissioning in Garching. Conceptually based on the design of the CryPTex trap built at Max-Planck-Institute for Nuclear Physics in Heidelberg [18], long storage times in excess of 10 hours will be realized, sufficiently long to cover the full expected range of 10^3–10^4 s. Figure 5 displays photographs documenting the present status of the Garching Paul trap setup. The top row shows the four-rod trap electrode system and its electrical contact rods (left), while the right panel shows the electrode system already mounted in the surrounding inner Cu vessel, together forming the cold mass of the cryogenic trap finally operating at liquid helium temperature. All parts have been gold plated to achieve optimum surface quality. The bottom picture shows the full assembly of the cryotrap, which is mounted to an active vibration-compensated optical table. The interaction chamber sits on top of a circular hole in the optical table, such that the main turbomolecular vacuum pump of the setup can be mounted from underneath the table. On top of the main vacuum chamber with the cold mass and 12 in-plane and 4 out-of-plane access ports for ion injection and extraction, diagnostics and optical manipulation a pulse tube cold head is mounted, vibrationally decoupled by an ultra-low vibration interface.
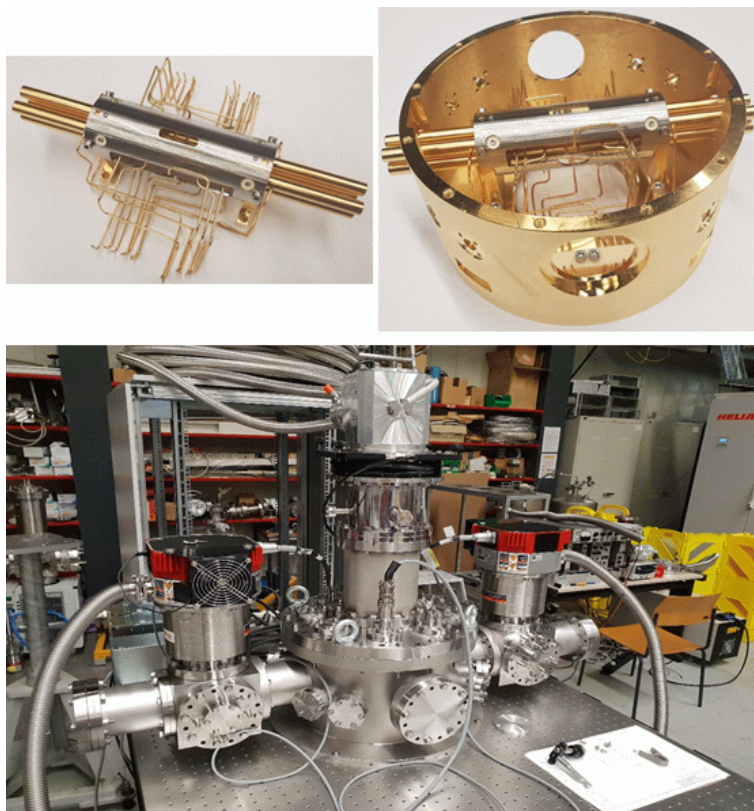
Fig. 5. Photographs of the cryogenic Paul trap under commissioning as the backbone for future laser manipulation (cooling, excitation) of ^{229m}Th ions aiming at establishing a nuclear frequency standard based on the thorium isomer. Top panels: cold mass of the cryotrap consisting of the four-rod trap electrode system and its electrical contact rod (left) and its assembly together with the surrounding Cu vessel (right). All parts have been gold plated for surface quality improvement. Bottom: assembly of the cryotrap mounted to an optical table, with the cold head on top, coupled to the cold mass via an ultra-low vibration interface. An optimized ^{229m}Th injection system with a new gas cell, extraction RFQ and quadrupol mass separator (QMS) is shown on the left, while the extraction side on the right features another QMS and a detection section.

On the left-hand side of the main chamber, the injection section will be mounted, comprised of a new buffer gas stopping cell (housing the ^{233}U recoil source, an RF/DC funnel and a de-Laval extraction nozzle) followed by an extraction radiofrequency quadrupole (RFQ) and a subsequent quadrupole mass separator (QMS). On the extraction side, another QMS and a detection section is mounted. Pumping is provided by turbomolecular pumps at

the injection and extraction side and underneath the main chamber. The cryotrap setup is presently being finalized and commissioning tests will start in late 2019. Subsequently, sympathetic laser cooling of Th^{3+} ions with Sr^+ will be added to the trap setup (featuring a favorable mass-to-charge ratio and offering a suitable electronic level scheme for laser cooling down to mK temperatures). First measurements on stored and cooled ^{229m}Th ions (1–10 ions simultaneously trapped) will target the ionic lifetime of the isomer.

From then on laser spectroscopy will dominate the methodology towards improved insight into the properties of the thorium isomer. The biggest challenge and ultimate doorway to the realization of a nuclear clock will be given by achieving a resonant laser excitation of the nuclear clock transition in a second phase of experimental campaigns, based on already existing laser technology [19]. Nanosecond VUV laser systems with GHz frequency bandwidth will allow for a systematic search for the nuclear resonance, while giving also the access to the spectral range of isomer shifts and hyperfine structure in different electronic environments of ions, molecules and solids.

A third phase will start upon realization of resonant laser excitation of the thorium isomer. Customized laser systems in the wavelength range around 150 nm will have to be developed, with a spectral bandwidth of 1 kHz and better, in order to allow for establishing a nuclear clock already able to contribute competitively to fundamental physics tests like variations of fundamental constants. With a laser system operating around 150 nm with a spectral bandwidth of 10^2–10^3 Hz (corresponding to $\frac{\Delta f}{f} = 5 \times 10^{-13}$–$5 \times 10^{-14}$), together with an expected sensitivity enhancement of about 10^4 provided by ^{229m}Th for studies of temporal variations of the fine structure constant α, better limits than the presently best value of $\frac{\dot{\alpha}}{\alpha} = (-0.7 \pm 2.1) \times 10^{-17}$ yr^{-1} [20] can be achieved.

Having learned from the recently refined value of the isomeric excitation energy that at least currently no CW laser will be able to reach into the energy region of the thorium isomer's ground-state transition around 150 nm, VUV frequency combs will be the present technology of choice to realize a laser system that will allow for operating a thorium nuclear clock. Identification of a resonant excitation of ^{229m}Th in the Paul trap will be achieved via a double-resonance technique as outlined in Ref. [3]. Figure 6 features a conceptual overview of such a high mode power VUV frequency comb system, based on the 7th harmonic of 1030 nm generated in a Xe gas jet.

In this preliminary layout and specification, the laser is based on an ultra-low noise IR frequency comb (mode spacing 100 MHz or 250 MHz). IR frequency combs are available with high longterm stability, 5×10^{-19} in 1000 s and 5×10^{-18} in 1 s could already be demonstrated [21]. The comb will be equipped with a near-IR (NIR) high-power output port at 1030 nm.
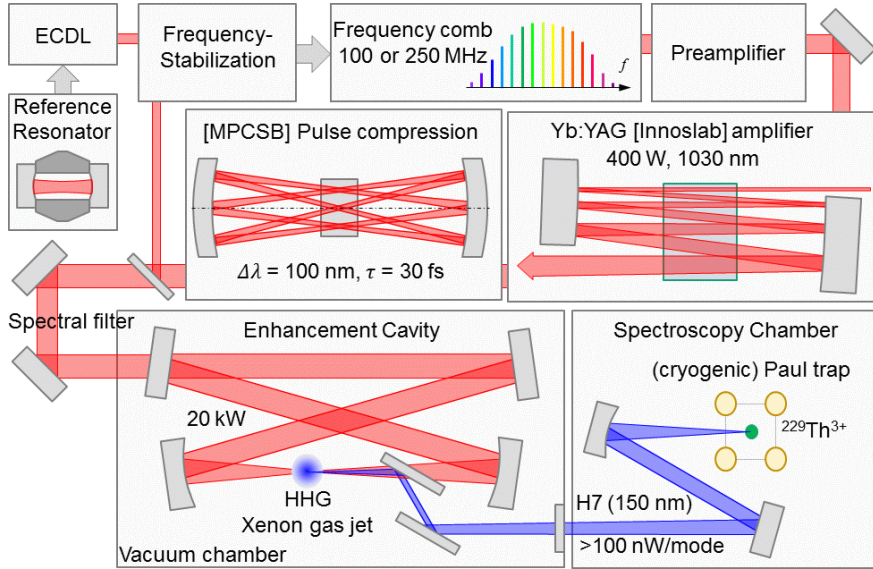
Fig. 6. Schematical outline of the VUV frequency-comb-based laser designed to drive the nuclear clock transition in ^{229m}Th.

The system will be stabilized to an external cavity laser (1542.14 nm), locked to an ultrastable high-finesse optical reference cavity. The comb output will subsequently be amplified to 400 W via a preamplifier and an Yb amplifier on the basis of the InnoSlab amplification technology [22], providing a large amplification and high average power at high efficiency, while no chirped pulse amplification (CPA) is needed at multi 10 MHz repetition rate. The output pulse exhibits a pulse length of about 700 fs with a pulse repetition rate of about 50 MHz. The amplification stage is followed by a 2-stage pulse compression, resulting in an output bandwidth of about 100 nm and a pulse duration of 30 fs. Finally, the compressed laser pulse will be injected into an enhancement cavity, where the 7th harmonic of the driving IR (comb) radiation in a Xe gas jet will be generated. A laser power of ≥ 1 nW per mode is conservatively targeted, while ultimately aiming for $(\geq)$ 100 nW/mode. The bandwidth will be ≤ 500 Hz, ultimately aiming at the few Hz regime.

Such a laser system could open the door to the operation of a nuclear clock and to experimental campaigns addressing the unique applications specifically envisaged for fundamental tests of physics beyond the Standard Model like time variations of fundamental constants.

With the presently ongoing dynamic experimental as well as theoretical efforts, stimulated by the recent experimental breakthroughs, the 'phase transition' from the nuclear-physics driven identification and characterization phase to a laser-physics driven consolidation, realization and application

phase in the more than 40 year old story around the thorium isomer and its potential as a nuclear frequency standard is already ongoing. Further intriguing results originating from this exotic solitaire in the nuclear physics landscape can be expected in the coming years.

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