Optical Access to the Lowest Nuclear Transition in 229m Th \diamond

P.G. Thirolf, D. Habs, M. Bussmann, H.J. Maier, J.B. Neumayr, J. Schreiber, M. Sewtz, and

J. Szerypo

Amongst the almost 2800 nuclides presently known to exist, ²²⁹Th is outstanding because of its first excited state, which exhibits the lowest excitation energy known in nuclear physics, thus bearing the potential to bridge the gap between nuclear and laser physics. Since the first studies already more than 30 years ago [1] a precise determination of the transition energy to the ground state was attempted, resulting in the value of 3.5(10) eV adopted for the last 15 years [2] together with the assignment of the singleparticle quantum numbers for the ground state of ²²⁹Th as $J^{\pi} = 5/2^+$ [633] and $3/2^+$ [631] for the first excited state. In contrast to the ground state of ²²⁹Th, exhibiting a halflife of 7340 years, the first excited state happens to be a long-lived isomer with a halflife of 3-5 hours (which has not yet been experimentally observed).

Given the low energy and long lifetime of the 229m Th isomer its unique properties are evident: a natural line width of 10^{-4} Hz corresponds to an ultimately sharp ground state transition with $\Delta E/E \approx 10^{-20}$. Thus this transition qualifies as an intriguing candidate for the realization of an ultra-precise nuclear clock, by far surpassing present frequency standards based on atomic transitions with an accuracy of presently about 10^{-15} . Moreover, theoretical studies [3,4] have revealed that a measurement of a temporal variation of the transition frequency of the ground state transition of 229m Th will provide a drastically enhanced sensitivity for potential temporal variations of fundamental constants like the fine structure constant α or the strong interaction parameter $m_q/\Lambda_{\rm QCD}$ (with quark mass m_q) by 5-6 orders of magnitude. Therefore many attempts have been conducted in the past to directly populate the first excited state in ²²⁹Th by laser excitation, however none of them succeeded. An obvious reason for those failures was given by a recent γ spectroscopy experiment using a novel X-ray microcalorimeter [5]. A revised value of 7.6(5) eV for the ground transition energy of 229m Th was obtained, corresponding to a wavelength of 163(11) nm, now placing this transition energy into the deep UV range. In order to be able to exploit the fascinating perspectives offered by this rather unique isotope a much higher precision on the transition energy is mandatory before any promising attempt can be made for laser excitation of the 7.6 eV transition. This triggered our experimental effort at the MLL, where existing technology can be exploited to create an unrivaled clean environment for UV spectroscopy of 229m Th. While population of the Th isomer by direct laser excitation suffers from the extremely sharp linewidth, a 2% branch in the α decay from ²³³U leading to ^{229m}Th can be used instead. Our experimental approach at the MLL is based on using a ²³³U source placed inside our MLL-IonCatcher buffer gas stopping cell [6]. Recoiling $^{229(m)}$ Th ions from the α decay will be stopped within 10 mm in about 40 mbar helium gas and guided by electrical DC and RF fields to the nozzle exit, where they will be extracted by a supersonic jet into the vacuum regime of a subsequent RFQ phase space cooler. Extraction from the gas cell can be achieved within 1-2 ms. This ensures that all conversion processes and prompt excitations will occur inside the gas cell, while only a clean sample of $^{229(m)}$ Th ions will be extracted, thus avoiding background contributions to subsequent decay studies. Fig. 1 displays an inside view into the buffer gas stopping cell with the 233 U source mounted ca. 60 mm in front of the nozzle exit.



Fig. 1: View inside the buffer gas stopping cell, where the 233 U source is mounted and α -decay recoil ions will be stopped and extracted via an RFQ ion guide before collecting the ions on a steel needle tip.

Behind the RFQ the extracted ions are collected on a steel needle tip (diameter about 40μ m) held at a suitable potential in order to create a point-like source for the subsequent decay of the isomer. Fig. 2 illustrates the principle of our experiment, where the UV fluorescence light from the 229m Th decay will be focused by a UV lens onto an MCP detector, representing the first optical focusing of a nuclear transition. The electrons created by the MCP will be converted to visible light in a phosphorous screen monitored by a highly-sensitive CCD camera.



Fig. 2: Schematics of the experimental approach to study the ground state transition in 229m Th at the MLL using the MLL-IonCatcher buffer gas stopping cell. Exploiting the population of 229m Th via α decay of 233 U, the recoiling α -decay product ions are extracted from the gas cell and collected on a needle tip at a suitable potential. Subsequently the UV fluorescence light from the 229m Th decay will be focussed onto an MCP monitored by a CCD camera behind a phosphorous screen.

The collection of Th ions on the needle tip behind the RFQ was optimized using ion optical trajectory simulations using the SIMION code. In order to enable a soft

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landing of the ions on the needle tip (operated at -20 V), they are reflected by a repelling potential of + 3.5 V onto the needle. A conducting cap covering the ceramic needle holder (+3.2 V) and an aperture (diameter 5 mm, 4.2 V) are included to shape the electrostatic field. The resulting ion trajectories are shown in Fig. 3. The probability of electron bridge processes [7] in the atomic shell of thorium or with the conducting surface of the needle (potentially able to quench the isomeric transition) could be reduced by coating the needle tip with a thin layer of a diamond-like carbon foil (DLC).



Fig. 3: Optimization of the ion optical trajectories behind the extraction RFQ by suitable potentials in order to allow for a soft landing of the ²²⁹Th ions on a steel needle tip.

In a first step the extraction efficiency of the setup was characterized using a $^{223}\mathrm{Ra}$ recoil source. Detecting the α decay of the ²¹⁵Po (grand-)daughter nuclei a rather high extraction efficiency of 48% could be determined. Subsequently a ²³³U source with an activity of 4 kBq (limited by the legal activity limits for the experimental area) was placed inside the gas cell and the recoil ions were extracted and collected onto the needle tip as described before. The α decay energy spectrum registered in a Si monitor detector close to the needle shown in Fig. 4 reveals the characteristic α peaks from the ²²⁹Th decay chain, thus proving the feasibility of the above described isomer preparation scheme.



Fig. 4: α energy spectrum from the decay of recoil ions extracted from the gas stopping cell following α decay of $^{233}\mathrm{U}.$ Prior to their decay the ions have been collected on a steel needle tip behind the extraction RFQ.

In order to detect the decay from the 7.6 eV transition, a CaF_2 UV lens was positioned behind the needle, allowing to focus the fluorescence light onto an MCP detector (diameter 75 mm, chevron geometry) followed by a phosphorous screen. A highly-sensitive low-noise 14 bit CCD camera monitors the phosphorous screen, allowing for exposure times between μ s up to 49 days without being limited by dark current or readout noise. Fig. 5 shows the setup as installed at the MLL with the MLL ionCatcher in the back, the lens and MCP detector in the middle section and the CCD camera in the front part.



Fig. 5: Photograph of the experimental setup at the MLL designed to search for the ground state decay of 229m Th at 7.6(5) eV. In the back the MLL-IonCatcher gas cell device is visible, while in the front the low-noise CCD camera for the detection of the UV fluorescence light around 160 nm can be seen, which has been converted to electrons using an MCP detector.

As soon as a fluorescence signal from the 229m Th decay will be detected, its transition energy can be determined to a precision of below 1 nm by inserting already available customized UV filters with very sharp absorption edges between 160 nm and 170 nm (allowing for a variation of the absorption edge by changing the angle of incidence). Thus it will be possible to improve the present knowledge on the transition energy by more than a factor of ten, which is a prerequisite for further improvements e.g. by using a grazing-incidence spectrometer. The final goal is to improve the accuracy of the 7.6 eV transition energy to a level, where laser excitation will be realistically feasible. A possible laser excitation scheme is given by the electronic level scheme of ²²⁹Th³⁺, allowing to apply a double-resonance method on a closed 3-level Λ system for laser cooling and fluorescence detection [8] e.g. in a linear Paul trap.

While there is certainly still a long way to go until the unique frequency accuracy offered by 229m Th can be practically exploited, nevertheless the perspective to improve the sensitivity to temporal changes of fundamental constants or the accuracy of frequency standards by orders of magnitude justifies any experimental efforts towards these ambitious goals.

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