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Corresponding Author:	Lars Wense GERMANY
Corresponding Author's Institution:	
Corresponding Author E-Mail:	L.Wense@physik.uni-muenchen.de
Order of Authors:	Lars von der Wense
	Benedict Seiferle
	Peter G. Thirolf
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TITLE:

Preparing an Isotopically Pure ^{229}Th Ion Beam for Studies of ^{229m}Th

AUTHORS AND AFFILIATIONS:

Lars von der Wense, Benedict Seiferle, Peter G. Thirolf

Faculty of Physics, Ludwig-Maximilians-Universität München, Germany

Corresponding Author:
Lars von der Wense
L.Wense@physik.uni-muenchen.de

Email Addresses of Co-authors:
Benedict Seiferle (Benedict.Seiferle@physik.uni-muenchen.de)
Peter G. Thirolf (Peter.Thirolf@physik.uni-muenchen.de)

KEYWORDS:

Nuclear clock, 229-Thorium isomer, buffer-gas cell, ion guide, Paul trap, mass separation, alpha decay, internal conversion

SUMMARY:

We present a protocol for the generation of an isotopically purified low-energy ^{229}Th ion beam from a ^{233}U source. This ion beam is used for the direct detection of the ^{229m}Th ground-state decay via the internal-conversion decay channel. We also measure the internal conversion lifetime of ^{229m}Th as well.

ABSTRACT:

A methodology is described to generate an isotopically pure ^{229}Th ion beam in the 2+ and 3+ charge states. This ion beam enables one to investigate the low-lying isomeric first excited state of ^{229}Th at an excitation energy of about 7.8(5) eV and a radiative lifetime of up to 10^4 seconds. The presented method allowed for a first direct identification of the decay of the thorium isomer, laying the foundations to study its decay properties as prerequisite for an optical control of this nuclear transition. High energy ^{229}Th ions are produced in the α decay of a radioactive ^{233}U source. The ions are thermalized in a buffer-gas stopping cell, extracted and subsequently an ion beam is formed. This ion beam is mass purified by a quadrupole-mass separator to generate a pure ion beam. In order to detect the isomeric decay, the ions are collected on the surface of a micro-channel plate detector, where electrons, as emitted in the internal conversion decay of the isomeric state, are observed.

INTRODUCTION:

The first excited metastable state in the thorium-229 nucleus, denoted as ^{229m}Th, exhibits a special position in the nuclear landscape, as it possesses the lowest nuclear excitation energy of all presently known ca. 176,000 nuclear excited states. While typical nuclear energies range from keV up to the MeV region, ^{229m}Th possesses an energy of below 10 eV above the nuclear

ground state[1,2,3]. The currently most accepted energy value for this state is 7.8(5) eV[4,5]. This low energy value has triggered interest from different physical communities and led to the proposal of several interesting applications. Among them are a nuclear laser[6], a highly stable qubit for quantum computing[7] and a nuclear clock[8,9].

The reason that ^{229m}Th is expected to offer a broad variety of applications is based on the fact that, due to its extraordinary low energy, it is the only nuclear state that could allow for direct nuclear laser excitation using currently available laser technology. So far, however, direct nuclear laser excitation of ^{229m}Th was prevented by insufficient knowledge of the metastable state's parameters like its precise energy and lifetime. Although the existence of a nuclear excited state of low energy in ^{229}Th was already conjectured in 1976[10], all knowledge about this state could only be inferred from indirect measurements, not allowing for a precise determination of its decay parameters. This situation has changed since 2016, when the first direct detection of the ^{229m}Th decay opened the door for a multitude of measurements aiming to pin down the excited state's parameters[11,12]. Here, a detailed protocol is provided, which describes the individual steps required for a direct detection of ^{229m}Th as achieved in the experiment of 2016. This direct detection provides the basis for a precise determination of the ^{229m}Th energy and lifetime and therefore for the development of a nuclear clock. In the following the concept of a nuclear clock as the most important application for ^{229m}Th will be discussed.

With a relative linewidth of $\Delta E/E \sim 10^{-20}$ the ground-state transition of the thorium isomer potentially qualifies as a nuclear frequency standard ('nuclear clock')[8,9]. Due to an atomic nucleus about 5 orders of magnitude smaller compared to the atomic shell, the nuclear moments (magnetic dipole and electric quadrupole) are accordingly smaller than the ones in atoms, rendering a nuclear clock largely immune against external perturbations (compared to the present state-of-the-art atomic clocks). Therefore, a nuclear frequency standard promises a highly stable and accurate clock operation. Although the accuracy achieved in the best present atomic clocks reaches about 2.1×10^{-18} [13], corresponding to a deviation of 1 second in a time period considerably longer than the age of the universe, nuclear clocks hold the potential of a further improvement which could become essential for a vast field of applications. Satellite-based navigational systems like the Global Positioning System (GPS), Global Navigation Satellite System (GLONASS) or Galileo presently operate with a positioning precision of a few meters. If this could be improved to the centimeter or even millimeter scale, a plethora of applications could be envisaged, from autonomous driving to freight or component tracking. Besides highly accurate clocks, such systems would require reliable uninterrupted operation, with long-term drift stability that secures long-resynchronization intervals. The use of nuclear clocks could turn out beneficial from this practical point of view. Further practical applications of (synchronized networks of) nuclear clocks could lie in the field of relativistic geodesy[14], where the clock acts as a 3D gravity sensor, relating local gravitational potential differences ΔU to measured (relative) clock frequency differences $\Delta f/f$ via the relation $\Delta f/f = -\Delta U/c^2$ (c denoting the speed of light). The best present clocks are capable of sensing gravitational shifts from height differences of about ±2 cm. Thus, ultra-precise measurements using a nuclear clock network could be used to monitor the dynamics of volcanic magma chambers or tectonic plate movements[15]. Moreover,

the use of such clock networks was proposed as a tool to search for the theoretically described class of topological dark matter[16]. Extensive discussion can be found in the literature on the application of a ^{229m}Th-based nuclear clock in the quest for the detection of potential temporal variations of fundamental constants like the fine structure constant α or the strong interaction parameter (m_q/Λ_{QCD}, with m_q representing the quark mass and Λ_{QCD} the scale parameter of the strong interaction), suggested in some theories unifying gravity with other interactions[17]. The detection of a temporal variation in the ground-state transition energy of ^{229m}Th may provide an enhanced sensitivity by about 2-5 orders of magnitude for temporal variations of the fine structure constant or the strong interaction parameter[18-26]. The current experimental limit for such a variation of α amounts to $(d\alpha/dt)/\alpha = -0.7(2.1)\cdot 10^{-17}$/yr [27]. In the following the experimental approach for the direct detection of the ^{229m}Th ground-state decay will be described.

Evidence for the existence of the 229-thorium isomer until recently could only be inferred from indirect measurements, suggesting an excitation energy of 7.8(5) eV (equivalent to a wavelength in the vacuum ultra-violet spectral range of 160(11) nm)[4,5]. Our experimental approach, aiming at a direct identification of the isomeric ground-state deexcitation of the ^{229m}Th isomer, builds on a spatial separation of the isomer population in a buffer-gas stopping cell, followed by an extraction, and mass-separated transport towards a suitable detection unit to register the deexcitation products[28,29]. Thus population and deexcitation of the isomer can be disentangled, resulting in a clean measurement environment, unaffected by prompt background contributions. Population of the isomer is achieved via the α decay from a radioactive ^{233}U source, where a 2% decay branch proceeds not directly to the ground state of ^{229}Th, but populates the isomeric first excited state instead. α-decay recoil nuclei are thermalized in an ultra-pure helium atmosphere of a buffer-gas stopping cell, before being guided by electric radiofrequency (RF) and direct current (DC) fields towards an extraction nozzle, where the emerging supersonic gas jet drags them into an adjacent vacuum chamber, housing a (segmented) radiofrequency quadrupole (RFQ) structure acting as ion guide, phase-space cooler and potentially also as linear Paul trap for bunching the extracted ions. For a detailed description of the buffer-gas stopping cell and extraction RFQ see Refs. [30,31,32]. Since up to that moment the extracted ion beam contains in addition to $^{229(m)}$Th also the chain of α decay daughter products, mass separation is performed using a quadrupole mass separator (QMS) in a subsequent vacuum chamber to finally generate an isotopically pure $^{229(m)}$Th beam in selectable charge states (q=1-3). A detailed description of the QMS can be found in Refs. [33,34]. Detection of the isomeric decay was achieved by impinging the Th ions directly on the surface of a microchannel-plate detector (MCP), where electrons are liberated, accelerated towards a phosphor screen and viewed by a charge-coupled device (CCD) camera. An overview of the experimental setup is shown in **Figure 1**. A detailed description is given in Ref. [35].

[place Figure 1 here]

The following protocol describes the underlying procedure to generate the $^{229(m)}$Th ion beam that enabled the first direct detection of the ground-state decay of the thorium isomer, thus laying the foundation for studying its decay properties as a prerequisite of the ultimately

envisaged all-optical control of this exotic nuclear state towards its application as an ultra-precise nuclear frequency standard. For better orientation a schematic overview of the setup used for direct detection of the isomeric decay[11] is provided in **Figure 2**, containing a numerical labelling of the components addressed in the following protocol. Also the components used for lifetime determination[12] are contained as an inset.

[place Figure 2 here]

PROTOCOL:

Note: Numbers given in the Protocol will reference to Figure 2.

1. **Direct Detection of Th-229 Isomeric Decay**

1.1. Mounting of the 233-uranium source

1.1.1. Mount the 233-uranium α source (1) through an access flange opening in the gas cell vacuum chamber to the upstream end of the funnel ring electrode system (2) inside the gas cell (3).

Note: The 290 kBq, 90 mm diameter ^{233}U source was produced via molecular plating onto a titanium-sputtered Si wafer[36]. In order to achieve optimum α recoil-efficiency of the source, its thickness should not exceed 16 nm, being the stopping range of 84 keV ^{229}Th in uranium.

1.1.2. Connect the cable to the source mount in order to allow for a DC offset of the source. Close and seal the access flange and connect the external wiring to the ^{233}U source.

1.2. Evacuation of the vacuum chamber and bake out

1.2.1. Start evacuation of the complete vacuum system by starting the roughing vacuum pump (4) if shut down (controlled via a computer-based user interface (5)) and open the three (hand-operated) valves (6) that connect the individual parts of the differential pumping stages to the roughing pump.

Note: Start opening the valves from chambers downstream of the gas cell with open gate valve (7) towards the turbo molecular pump of the gas cell to create a pressure gradient that prevents potential contaminations from downstream chambers to be sucked into the gas-cell chamber where highest cleanliness is demanded.

1.2.2. Once the pressures have reached a level in the sub-mbar range (read out via user interface (5)) start the turbo pumps of the gas cell (8), the extraction radio-frequency quadrupole (RFQ) (9) and the quadrupole mass separator (QMS) (10).

1.2.3. (optional) Open the bypass valve (11) to also allow for efficient evacuation of the gas-supply tubing.

1.2.4. Continue pumping for a few (4-5) hours until reaching saturation pressure, typically in the range of low 10^{-7} mbar.

1.2.5. Start the baking system (12) via the user interface (5) with an upramping (typically 20 °C - 40 °C per hour) heating curve to a maximum of 130 °C.

1.2.6. Keep baking the vacuum system at 130 °C for 1-2 days until the pressure readings start to decrease.

1.2.7. Start the cooling sequence of the baking system via the user interface (5) with a downramping sequence, typically 20 °C - 40 °C per hour.

Note: Cooldown of the system typically requires 8 hours and is performed overnight. Successful preparation of the vacuum system is achieved when the final cell-pressure after cool down ranges below $5x10^{-10}$ mbar. The pressures in the RFQ and QMS chamber will be in the 10^{-9} mbar and 10^{-8} mbar range, respectively.

1.2.8. Connect the external wiring to the RFQ vacuum chamber.

1.3. Preparation of the gas system and supply of ultra-pure He

1.3.1. Start the MonoTorr gas-purifier (13) and wait 20 minutes until it has reached its operating temperature.

1.3.2. Close the bypass valve (11) if open.

1.3.3. Open the He-gas cylinder (14) (He of 99.9999 % purity is used for operation).

1.3.4. Open the pressure reducer valve (15) until a pressure of about 0.5 bar is shown.

1.3.5. Open the valve that connects the pressure reducer to the gas tubing (16).

1.3.6. Open the gas-flow control (17) until a gas flow of about 1.1 (corresponding to about 5 mbar l/s) is shown.

1.3.7. Flush the gas tubing for about 10 minutes to remove residual gases from the tubing.

1.3.8. Close the valve that connects the pressure reducer to the gas tubing (16).

1.3.9. Wait a few minutes until the He is removed from the gas tubing.

1.3.10. (optional) For highest purity of the buffer gas, fill the cryo-trap (18) with liquid nitrogen.

1.3.11. Set the gate valve (7) between the buffer gas cell and its turbo molecular pump to automatic operation and close the valve via the user interface (5).

1.3.12. Open the valve that connects the pressure reducer to the gas tubing (16).

Note: The buffer-gas stopping cell is now filled with ca. 30 mbar of He gas. In this way the RFQ and QMS pressures are raised to 10^{-4} mbar and 10^{-5} mbar, respectively.

1.3.13. Adjust the rotary speed of the turbo-molecular pump of the extraction-RFQ vacuum chamber (9) to 50% in order to set an ambient pressure of about 10^{-2} mbar.

1.4. Apply the electric guiding fields for continuous ion extraction

1.4.1. Apply a DC potential to the 233-uranium α source (1) of 39 V in continuous mode via a customized DC voltage supply (19).

1.4.2. Apply a DC potential gradient of 4 V/cm (ranging from 35 V to 3 V) via a DC power supply (20) and a voltage offset of 3 V via a 24 channel DC offset supply (21) to the 50-fold segmented funnel ring-electrode system. All voltages are controlled with the computer-based user interface (5).

1.4.3. Apply a DC potential of typically 2 V to the extraction nozzle electrode (22) with the help of the same computer-based user interface (5).

1.4.4. Apply a DC potential gradient to the 12-fold segmented extraction-RFQ (27).

Note: The voltage of each segment can be applied individually with the help of the computer-based user interface (5) via the 24 channel DC offset supply (21). A voltage of 1.8 V is applied to the segment closest to the extraction nozzle. The voltages of the subsequent segments are then stepwise decreased by 0.2 V, resulting in a voltage of 0 V applied to the 10th RFQ segment. This corresponds to a DC gradient of 0.1 V/cm. In case of intended continuous transport of the extracted ions a voltage of 0 V is applied to the 11th and 12th RFQ segments. For this purpose the DC voltage supply of the 12^{th} RFQ segment (23) is left at 0 V and the customized trigger module (24) is set to continuous operational mode.

1.4.5. Apply RF frequency and amplitude to the funnel ring electrode system via a function generator (25) and linear RF amplifier (26).

Note: Typical values for frequency and amplitude are 850 kHz and 220 V_{pp}, respectively. The voltages can be controlled with a computer-based user interface (5). During funnel-RF voltage application, monitor the current of the funnel DC offset supply (21). In case of sparks, which can occur if buffer-gas purity is insufficient, this current will start to increase.

1.4.6. Apply RF frequency (typically 880 kHz) and amplitude (typically 120-250 V_{pp}) to the extraction radiofrequency quadrupole (27) (extraction-RFQ) via a frequency generator (28) and two RF amplifiers (29, 30), one for the RFQ and one for the individual bunching electrode. The voltage can be controlled with the computer-based user interface (5).

1.4.7. Apply a DC potential of -1 V to the exit electrode (31) of the extraction-RFQ via a Mesytec MHV-4 DC voltage supply (32).

1.4.8. Apply DC offset voltages to the quadrupole mass separator (33) (QMS). The offset voltage of the QMS (center electrode and Brubaker lenses) is chosen to be -2 V via customized DC offset modules (34,35).

1.4.9. Start the quadrupole mass separator (33) QMS by switching on the QMS function generator (36), the RF amplifier (37) and starting the QMS user-interface (38). In the QMS user interface the mass-over-charge ratio of the selected ion species is inserted (typically 76 u/e or 114.5 u/e, for the extraction of Th^{3+} or Th^{2+}, respectively). Also the QMS acceptance (typically 1 to 2 u/e) and the RF frequency (typically 825 kHz) is inserted.

Note: The Labview program will automatically apply and control the RF amplitude and the DC potentials required for ion selection. The required RF amplitudes range from 600 to 1500 V_{pp} and the DC potentials range from 50 V to 120 V. The DC potentials for mass separation are generated by a customized DC module (39). A feedback-loop is implemented for RF and DC voltage stabilization.

1.4.10. Apply DC potential to the focusing triodic electrode structure (40) behind the QMS (-2 V/- 62 V/-22 V) via the Mesytec 4 channel (MHV-4) voltage supply module (32).

1.5. Probe the ion extraction and tune the QMS

1.5.**1.** Apply an attractive surface potential of -1000 V to the front plate of the double-plate (chevron geometry) microchannel-plate detector (41) (MCP) via a high voltage (HV)-module (42).

1.5.2. Apply a potential of +900 V to the back side of the second MCP plate via a HV-module (43).

1.5.3. Apply a potential of +5000 V to the phosphor screen (44) placed behind the MCP detector via a HV-module (45).

1.5.4. Switch on the CCD camera (46) behind the phosphor screen and configure the exposure parameters of the CCD camera in the corresponding graphical user interface on the data acquisition PC (47).

Note: The CCD camera is placed in a light-tight housing (48) to cover the detection from ambient light. In case that the extraction is running properly and ions are passing through the QMS a strong signal should be visible on the phosphor screen caused by the ionic impact of the extracted ions. This signal is now monitored by the CCD camera.

1.5.5. Perform a mass scan to probe the signal shape and accordingly tune the QMS to extract the desired ion species.

Note: This is an iterative procedure carried out with the help of the QMS user interface (38). Select a desired mass-over-charge ratio (typically 114.5 u/e for $^{229}Th^{2+}$) and the QMS resolving power (typically 1 u/e), then probe the ionic impact signal via the CCD camera. Shift the selected mass in 0.5 u/e steps until a signal is observed. As soon as a signal is observed, probe if also the $^{233}U^{2+}$ signal is observable by shifting the mass-over-charge-ratio by 2 u/e to higher masses. If also this signal is observed, probe if the signals can be separated. If this is not the case, adapt the QMS resolving power until the $^{229}Th^{2+}$ and $^{233}U^{2+}$ signals can clearly be distinguished. Then set the QMS to extract only the $^{229}Th^{2+}$ ion species.

1.6. Detection of the isomeric decay

1.6.1. Switch off the QMS pressure sensor (49) via the pressure sensor control unit (50) in order to reduce background from ionized helium and light produced by the sensor.

1.6.2. Adjust the QMS parameters to extract the Th^{2+} or Th^{3+} ion species for isomeric decay detection.

1.6.3. Reduce the surface potential of the front plate of the MCP detector (41) to -25 V via (42) in order to avoid detecting the signal from electrons originating directly from the ionic impact of impinging ions. In this way a 'soft landing' of the $^{229(m)}Th$ ions on the MCP surface is achieved prior to the isomeric decay.

1.6.4. Apply an accelerating potential of typically +1900 V to the second MCP plate for optimum electron amplification via (43).

1.6.5. Apply an accelerating potential of typically +6000 V to the phosphor screen placed behind the MCP detector via (45).

Note: The actually applied voltages will depend on the MCP performance.

1.6.6. Start the acquisition sequence of CCD images and store the data on disk via the camera user-interface (47).

1.6.7. Use Matlab programs for image evaluation and post-processing.

Note: A description of the programs and how they are used can be found in Ref. [35] Appendix B.3. Raw data of image frames as well as the programs used for evaluation have been made available online at DOI 10.5281/zenodo.1037981.

2. Measurement of the ^{229m}Th Half-Life (Re-arrangement of the Setup)

2.1. Shut down and venting of the system.

2.1.1. Power off the high voltages of the MCP detection system (42,43,45), the QMS (37,38), the Funnel system (25,26) and extraction RFQ (28,29,30).

2.1.2. (Optional) Power off all remaining DC voltages.

2.1.3. Manually close the He supply system (valves 14 and 16) and wait until the pressure of the buffer-gas stopping cell is reduced to below 2 mbar.

2.1.4. Open the gate valve that connects the turbo pump to the buffer-gas stopping cell (7) via the user interface (5) and wait until the He is fully removed from the system.

2.1.5. Close valve (17) of the gas supply line and switch off the gas purifier (13).

2.1.6. Set the gate valve (7) to manual operation in order to hinder it from closing when the system is vented with dry nitrogen.

2.1.7. Close the three valves that connect the turbo pumps with the roughing pump (6) and power down the three turbo pumps (8,9,10).

2.1.8. Switch on the QMS pressure sensor (49).

2.1.9. Wait until the rotation speed of the turbo pumps is reduced to significantly below 100 Hz as monitored on the user interface (5).

2.1.10. Fill the dewar (51) with liquid nitrogen and open the venting valve (52) slowly. Wait several minutes until the system is completely vented with dry nitrogen.

Note: Alternatively, dry nitrogen from a gas cylinder could be used. But in this case, care has to be taken that no overpressure would occur (*e.g.,* by inserting an overpressure valve or rupture disk). The use of air is also an alternative but will lead to slightly longer evacuation times due to the humidity.

2.1.11. Close the venting valve (52).

2.2. Replace the MCP with phosphor screen (41,44) by a small single-anode MCP detector (53)

2.2.1. Disconnect and remove the CCD camera (46) together with the light-tight housing (48).

2.2.2. Disconnect the MCP detector with phosphor screen (41,44).

2.2.3. Open the vacuum flange that connects the MCP and phosphor screen with the vacuum chamber.

2.2.4. Place the single-anode MCP (53) with a few mm distance behind the exit of the triode extraction system (40) and connect the three wires that connect the front plate (42), back plate (43) and the anode of the MCP (54) with the electric feedthroughs.

2.2.5. Close the vacuum chamber, the system is now ready for evacuation and bake out.

2.2.6. Provide the external wiring of the single anode MCP to the HV modules and the read-out system.

2.3. Evacuation of the system and bake out

2.3.1. Evacuate the vacuum system by following the steps 1.2.1 to 1.2.3.

2.3.2. Follow the bake-out procedure of steps 1.2.4 to 1.2.8.

2.4. Preparation of the gas-tubing and supply of ultra-pure He

2.4.1. Follow the steps 1.3.1 to 1.3.12.

Note: For bunched mode operation we typically operate the RFQ turbo-pump at 100 % rotation speed, resulting in a pressure in the 10^{-4} mbar range.

2.5. Apply the electric guidance fields for ion bunching

2.5.1. Apply a DC potential of 69 V to the 233-uranium α source (1) via the customized DC voltage supply (19).

2.5.2. Apply a DC potential gradient of 4 V/cm (ranging from 65 V to 33 V) via the DC power supply (20) and a voltage offset of 33 V via the 24 channel DC offset supply (21) to the 50-fold segmented funnel ring-electrode system. All voltages are controlled with the computer-based user interface (5).

2.5.3. Apply a DC potential of 32 V to the extraction nozzle electrode (22) with the help of the same computer-based user interface (5).

2.5.4. Apply a DC potential gradient to the 12-fold segmented extraction-RFQ.

Note: The voltage of each segment can be applied individually with the help of the computer-based user interface (5) via the 24 channel DC offset supply (21). A voltage of 31.8 V is applied to the segment closest to the extraction nozzle. The voltages of the subsequent segments are then stepwise decreased by 0.2 V, resulting in a voltage of 30 V applied to the 10th RFQ segment. This corresponds to a DC gradient of 0.1 V/cm. In case of creation of a bunched beam the ions are stored and cooled in the 11th electrode. Therefore, the 11th electrode is set to 25 V and the last RFQ segment is raised to 44 V via the DC voltage supply (23) to accumulate ions in the local potential bucket before releasing the ion bunch by lowering the last electrode segment to 0 V within a microsecond, triggered by a customized trigger module (24).

2.5.5. Set the trigger module (24) to bunch mode. The trigger module allows an adjustment of the trigger rate and timing. Typically, 10 Hz is chosen as the trigger rate.

2.5.6. Apply the remaining voltages to the system, following steps 1.4.5 to 1.4.10.

2.6. Probe the ion extraction and tune the QMS

2.6.1. Switch off the QMS pressure sensor (49) via the pressure sensor control unit (50) in order to reduce background from ionized helium and light produced by the sensor.

2.6.**2.** Apply an attractive surface potential of -2000 V to the front plate of the single anode MCP (52) via a HV-module (42).

2.6.3. Apply a potential of -100 V to the back side of the MCP. The MCP anode is set to ground.

2.6.4. Switch on the 12 V power supply module (55) for the MCP preamplifier (56).

Note: Single ions impinging on the MCP detector are now counted with the help of the combination of the preamplifier (56), an amplifier (57) and a constant fraction discriminator (CFD) (58). The CFD signal is send to a data acquisition (DAQ) card of the PC used for QMS control and can be monitored via the QMS user interface (38).

2.6.5. Perform a mass scan to probe the signal shape and accordingly tune the QMS to extract the desired ion species.

Note: This is done with the help of the QMS user Interface (38). For this purpose, an initial and a final mass-over-charge ratio is set (*e.g.,* 110 u/e to 120 u/e for the $^{229}Th^{2+}$ mass-range), as well as the resolving power (*e.g.,* 1 u/e) and the integration time (5 s) per scan step and the mass scan is started by pressing the scan button. In case that the extraction is running properly and ions are passing the QMS, strong signals of thorium and uranium will be visible caused by the ionic impact of the extracted ions.

2.7. Lifetime measurement

2.7.1. Adjust the QMS parameters to extract the Th^{2+} or Th^{3+} ion species for isomeric decay detection.

2.7.2. Reduce the surface potential of the front plate of the MCP detector (52) to -25 V via (42) in order to reduce the ionic impact signal.

2.7.3. Apply an accelerating potential of typically +1900 V to the second MCP plate for optimum electron amplification via (43).

2.7.4. Apply an accelerating potential of typically +2100 V to the MCP anode via (53).

2.7.5. Start the data acquisition via a microchannel scaler (59).

Note: The preamplifier (56) and the microchannel scaler (59) allow for time resolved read-out of the MCP detector. The ion bunches and the microchannel scaler are both triggered by the trigger module (24). The scaler signal is obtained via a Labview user interface (60). An exponential decay tail of about 10 microseconds lifetime becomes visible after the ion bunches, corresponding to the thorium isomeric decay.

REPRESENTATIVE RESULTS:

The method described before allowed for the extraction of α decay products from a ^{233}U source placed inside a buffer-gas stopping cell, operated at ca. 30 mbar ultra-pure helium gas at room temperature. For the first time up to triply charged ions could be extracted from such a device with high efficiency [29]. **Figure 3a** displays the mass spectrum of ions extracted from the buffer-gas cell, showing three groups of ^{233}U α-decay products (plus accompanying contaminant adducts) in singly, doubly and triply charged ionic states. Noteworthy is the dominance of $^{229}Th^{3+}$ extraction compared to $^{233}U^{3+}$, while both species are extracted with about equal intensity when being doubly charged. This fact was used for comparative measurements with ^{233}U ions, which allowed the exclusion of any ionic impact as signal origin.

[place Figure 3 here]

After transport, cooling and mass separation, the ion beam impinges onto the surface of a microchannel-plate detector, where a low attractive surface potential ensures the suppression of ionic impact signals and leaves only electrons arising from the Internal Conversion (IC) decay channel of the ^{229m}Th isomer to be multiplied in the strong electric field of the detector plate channels. The resulting MCP signals as obtained for three different uranium sources are displayed in **Figure 3b**. The ion species of doubly or triply charged ions which was selected with the help of the quadrupole mass separator in each individual measurement is indicated by the arrows from the upper panel. Shown are pictures acquired with the CCD camera behind the phosphor screen, onto which the electrons from the MCP were accelerated. The field of view of the CCD camera is indicated by the dashed circles for triply (first two columns) and doubly charged (last two columns) ^{229}Th and ^{233}U ions, respectively. The upper row represents the

result obtained for a small-area ^{233}U source (ca. 1000 extracted $^{229}Th^{3+}$ ions per second, source 1), while the bottom row shows the same for a stronger source with ca. 10,000 extracted $^{229}Th^{3+}$ ions per second (source 3). It is obvious that in both cases a clear signal is obtained for ^{229}Th, while no indication of an electron signal is observed for ^{233}U [11]. In order to prove that this signal indeed originates from a nuclear deexcitation and not from an atomic shell process, the middle row shows the resulting camera image when using a ^{234}U source, where the α decay populates the neighboring isotope ^{230}Th, with a comparable electronic, yet different nuclear structure. As expected for ^{230}Th, no indication of a conversion electron signal is found in any of the cases studied. So the strong signal, displayed in **Figure 3c** with excellent signal-to-background ratio, is clearly correlated with the decay of ^{229m}Th.

Additional verification measurements to support this interpretation are shown in **Figure 4**. They show two measurements to give further evidence that the registered electron signals indeed originate from the decay of the nuclear isomer: in **Figure 4a** it is shown that the attractive surface potential of the MCP detector was varied from -100 V (favoring the occurrence of electrons from ionic impact) down to 0 V, comparing the count rates registered with the MCP for extracted $^{229}Th^{2+}$ (red) and $^{233}U^{2+}$ ions (blue). Clearly the count rate drops down to zero for $^{233}U^{2+}$ when realizing a 'soft landing' of the incoming ions with a surface voltage below ca. -40 V, while a considerable count rate remains for $^{229}Th^{2+}$ until the threshold of 0 V. In **Figure 4b**, the blue curve shows the electron count rate registered for extracted ions after strong acceleration towards the MCP detector surface with -2000 V. Ionic impact of $^{233}U^{2+}$ and $^{229}Th^{2+}$ ions is observed with about equal intensity, as already shown for doubly charged ions in the extracted mass spectrum of **Figure 3a**. The red curve shows the same scenario, however now for a 'soft landing' of incoming ions with -25 V MCP surface potential. No indication of the ionic impact signal of $^{233}U^{2+}$ is visible any more, while for $^{229}Th^{2+}$ a signal remains, originating from the isomeric internal conversion decay[11].

[place Figure 4 here]

Thus, it can be unambiguously proven (together with additional arguments given in Ref. [11]) that the signal observed in **Figure 4** originates from the isomeric decay of ^{229m}Th and represents the first direct identification of the deexcitation of this elusive isomer.

Subsequently the segmented extraction-RFQ was operated as a linear Paul trap to create a bunched ion beam, thus allowing for lifetime measurements of the thorium isomer. Since our room-temperature high vacuum does not allow for sufficiently long storage times to investigate the expected radiative lifetime of up to 10^4 seconds, only a lower limit of $t_{1/2} > 1$ minute could be derived for charged ^{229m}Th ions, limited by the maximum achievable ion storage time in the linear Paul trap[11]. However, using the same detection strategy as applied before for the identification of the isomer decay after neutralization of the thorium ions on the surface of an MCP detector, the expected much shorter lifetime for neutral ^{229m}Th atoms undergoing internal conversion decay provides access to lifetime information[12]. **Figure 5a** shows the expected shape of the decay time spectrum as simulated for an ion bunch with a pulse width of 10 μs. While the red curve indicates the ionic impact signal and the signal from an exponential decay with 7 μs half-life is represented by the gray curve with a long decay tail, the expected signal

from the decay of the thorium isomer, comprised of both the ionic impact and the exponential isomeric decay, is illustrated by the blue curve. **Figure 5b** displays the outcome of the corresponding measurement for $^{233}U^{3+}$ (red) and $^{229}Th^{3+}$ (blue), respectively. While uranium ions only exhibit their ionic impact signal, for 229-thorium clearly the expected decay tail of the isomer decay can be observed[12].
[place Figure 5 here]

Fitting the decay tail with an exponential (corresponding to a linear fit to the logarithmic representation in **Figure 6**) finally results in a half-life of the neutral ^{229m}Th isomer of 7(1) μs [12]. This value nicely agrees with the theoretically expected lifetime reduction by nine orders of magnitude from the ca. 10^4 seconds in case of the charged isomer due to the large conversion coefficient of $\alpha_{IC} \sim 10^9$ [37].
[place Figure 6 here].

FIGURE AND TABLE LEGENDS:

Figure 1: Overview of the experimental setup. The thorium-229 isomer is populated via the 2% decay branch in the α decay of uranium-233. ^{229m}Th ions, leaving the ^{233}U source due to their kinetic recoil energy, are thermalized in a buffer-gas stopping cell filled with 30 mbar helium gas. The ions are extracted from the stopping volume with the help for RF and DC fields and a low-energy ion beam is formed with the help of a radio-frequency quadrupole (RFQ). The ion beam is mass-purified with the help of a quadrupole-mass-separator (QMS) and the ions are softly implanted into the surface of a micro-channel-plate (MCP) detector combined with a phosphor screen which allows for spatially resolved detection of any occurring signals. With kind permission of Springer Research, this figure has been modified from [11].

Figure 2: Schematic sketch of the experimental setup used for isomeric decay detection. The components used for lifetime measurement are shown as an inset. Individual components that will be referenced in the protocol section are numerically labelled.

Figure 3: Identification of the direct decay of the 229-thorium isomer. a) Complete mass scan performed with the ^{233}U source 1 [29]. Units are given as atomic mass (u) over electric charge (e). b) Comparison of MCP signals obtained during accumulation of thorium and uranium in the 2+ and 3+ charge states (as indicated by the arrows linking to the mass scan). ^{233}U and ^{234}U sources were used (the source number is given on the right-hand side of each row). Each image corresponds to an individual measurement of 2000 s integration time (20 mm diameter aperture indicated by the dashed circle). Measurements were performed at -25 V MCP surface voltage in order to guarantee soft landing of the ions. c) Signal of the ^{229}Th isomeric decay obtained during $^{229}Th^{3+}$ extraction with source 1. A signal area diameter of about 2 mm (FWHM) is achieved. The obtained maximum signal intensity is 0.08 counts/(s mm^2) at a background rate of about 0.01 counts /(s mm^2). With kind permission of Springer Research [11].

Figure 4: Isomer decay verification measurements. a) $^{229}Th^{2+}$ signal (red) compared to $^{233}U^{2+}$ (blue) as a function of the MCP surface voltage. Errors are indicated by shaded bands. b) Signal

of extracted ions as a function of the mass-to-charge ratio behind the QMS for MCP surface voltages of -25 V (isomer decay, red) and -2000 V (ion impact, blue). Note the different integration times and axis scales. In addition to the signal at 114.5 u/e (corresponding to $^{229}Th^{2+}$), a further signal at 117.5 u/e occurs, which originates from the isomeric decay of ^{235}U. With kind permission of Springer Research[11].

Figure 5: Simulated and measured temporal ion impact and decay characteristics. a) Simulation of the isomer decay time characteristics of ^{229}Th bunches. The simulation is based on a measured bunch shape and the assumption that 2 % of the ^{229}Th ions are in the isomeric state with a half-life of 7 μs after neutralization. The electron detection efficiency is assumed to be 25 times larger than the ion detection efficiency. b) Measurement of the isomeric decay with a bunched $^{229(m)}Th^{3+}$ ion beam (blue). A comparative measurement with $^{233}U^{3+}$ is shown in red. With kind permission of the American Physical Society[12].

Figure 6: Fit to ^{229m}Th decay curve. Logarithmic plot of the temporal decay characteristics for $^{229(m)}Th^{2+}$ ions (a) and $^{229(m)}Th^{3+}$ ions (b) together with a fit curve applied to extract the isomeric half-life of ^{229m}Th after charge recombination on the MCP detector surface. With kind permission of the American Physical Society[12].

DISCUSSION:

The range of recoiling α decay daughter nuclei in uranium amounts to only about 16 nm. In order to achieve a high efficiency of the source for α-recoil ions for a given source activity, it is mandatory to limit the source material thickness to this range. The α recoil extraction efficiency is strongly affected by the cleanliness of the buffer-gas cell. Contaminations of the stopping gas will lead to charge exchange or molecule formation. Therefore, the gas cell itself has to be built according to ultra-high vacuum standards, in particular to allow for a baking of the cell and avoiding any organic materials inside. The stopping gas has to be purified according to technical state-of-the-art, starting from highest-grade gas purity assisted by catalytic purification and delivery to the gas cell via an ultra-clean gas–supply line, partially surrounded by a cryogenic trap to freeze out impurities. In general, careful alignment of the central axis of the complete setup to the position of the gas cell extraction nozzle is essential for achieving a high transport and detection efficiency[29].

Step 1.4.5 is the most critical of the protocol. For efficient ion extraction a high RF amplitude has to be applied to the funnel ring electrode. However, if the amplitude is chosen too high, sparks in the gas cell will occur. The maximum achievable RF voltage amplitude depends critically on the purity of the buffer gas. A successful application of voltage is monitored via the current of the funnel offset voltage. This current will increase in the case of sparks. If sparks have occurred, the bake-out procedure has to be repeated in order to guarantee highest ion extraction efficiency.

A further critical point is the application of the high voltages to the MCP detector (steps 1.6.2-1.6.4). Field emissions can occur on the MCP, leading to the emission of electrons which can lead to artefactual signals.

Optimum ion extraction and (cooled and mass purified) transport towards the detection unit requires careful alignment of the central optical axis. The availability of an optical alignment system (alignment laser or theodolite) is essential. The efficient ion transport through the extraction RFQ and the QMS requires a continuous stabilization of the radio-frequency amplitudes for the two opposite phases applied to each opposite pair of rods[29]. Identification of extraction or transport problems can be facilitated by an ion diagnostic realized *e.g.,* via a multichannel-plate detector placed either consecutively at different positions along the ion path during the commissioning phase of the setup, or alternatively, *e.g.,* under 90^{o} behind the extraction RFQ with a high negative surface voltage (1-2 kV) to attract all extracted ions towards the detector.

During operation typically two problems can arise. Not all voltages are correctly applied. In this case usually no ions are extracted, and one has to find the place of not correctly applied voltage. Also, impurities are present in the helium buffer-gas. In this case the extraction efficiency for triply charged thorium ions will be drastically reduced and molecule formation occurs. In the worst case, even sparks will show up when the funnel voltage is applied. The reason for insufficient gas purity is typically a leakage in the gas supply line or a not properly closed flange of the buffer-gas stopping cell.

The described method to generate a clean beam of ions containing the energetically low-lying ^{229m}Th isomer can be applied to all comparable cases where the ion of interest can be extracted from the buffer-gas atmosphere in sizeable amounts. Cleanliness of the gas-cell and buffer gas is mandatory, thus the amount of remaining gas impurities is a limitation to the sensitivity of the method. While the employed microchannel-plate detector (MCP) is based on the detection of electrons, as exploited here for the registration of low-energy conversion electrons, this case already lies at the low-energy border of the efficiency curve for MCPs[38], while for higher energies the method would significantly gain in detection efficiency.

So far, the described method has provided the only reported direct and unambiguous identification of the de-excitation of the thorium isomer. Alternatively, vacuum ultra-violet (VUV)-transparent crystals (with large bandgaps, exceeding the assumed excitation energy of the isomer) are doped with ^{229}Th. The goal is to place ^{229}Th ions in high (4^{+}) charge state of crystal lattice positions, inhibit de-excitation by the large band gap and aim at an excitation of the isomer using X- rays from synchrotron light sources. Despite the elegant concept of this approach, so far no VUV fluorescence could be observed in a series of experiments reported by several groups worldwide[39-43]. The same holds for a class of experiments that aims to realize the nuclear excitation of the isomer via the electron shell of ^{229}Th, using a so-called electron-bridge transition. Here a resonant coupling between an electron shell transition and the nuclear isomer should allow for a more efficient isomer population[44,45]. Other experiments that aim for the investigation of the isomeric properties are based on microcalorimetry[46] or the observation of the hyperfine-shift in the atomic shell[47]. Very recently another method to excite the isomer in a laser-induced plasma was reported[48] and is subject to scientific discussion within the community.

The discovery of the internal conversion decay channel of the thorium isomer[11] and the determination of the corresponding half-life of neutral ^{229m}Th (7(1) μs)[12] can be exploited in the future to realize a first all-optical excitation with a pulsed, tunable VUV laser based on already existing technology. Thus the present paradigm that this would require much better knowledge of the excitation energy and a corresponding customized laser development can be circumvented. In contrast, exploiting the knowledge of internal conversion electron emission, gating the detection of conversion electrons with the laser pulse will provide a high signal-to-background ratio, while allowing for a scan of 1 eV of excitation energy in less than 3 days[49]. Moreover, a determination of the excitation energy of the isomer, still being work in progress, can be based on the described method of generating the ^{229m}Th beam by sending IC decay electrons into a magnetic-bottle electron spectrometer with retarding field electrode grids[50]. The same technique will also allow to determine the isomeric lifetime for different chemical environments (*e.g.,* on large band-gap materials like CaF_2 or frozen argon) or in $^{229}Th^+$ as well as in the free, neutral atom.

The described method of generating an isotopically pure thorium ion beam of 3+ charge state can be used as a tool to provide thorium ions for future laser-spectroscopy experiments. In this case the ion beam can be used to load a Paul trap in a stable and efficient way. So far, the only alternative method is to produce $^{229}Th^{3+}$ by laser ablation from a solid target. This, however, requires high laser intensities and a large quantity of ^{229}Th, which is an expensive radioactive material and leads to the contamination of used vacuum components. For this reason, the described method can be of significant advantage when it comes to nuclear laser spectroscopy experiments. A first application of this type has already been published[51].

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DISCLOSURES:
The authors have nothing to disclose.

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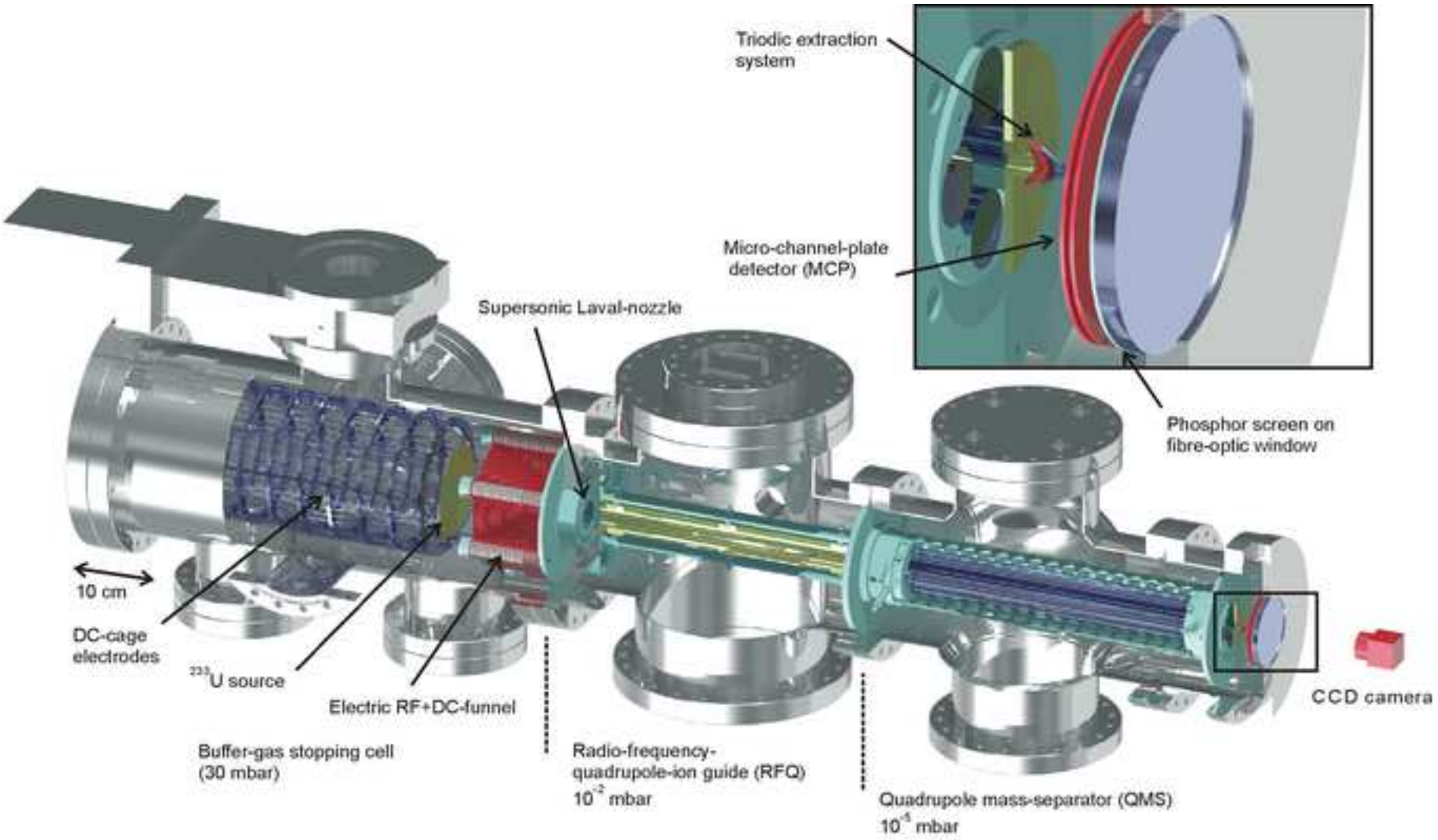
Figure



Figure

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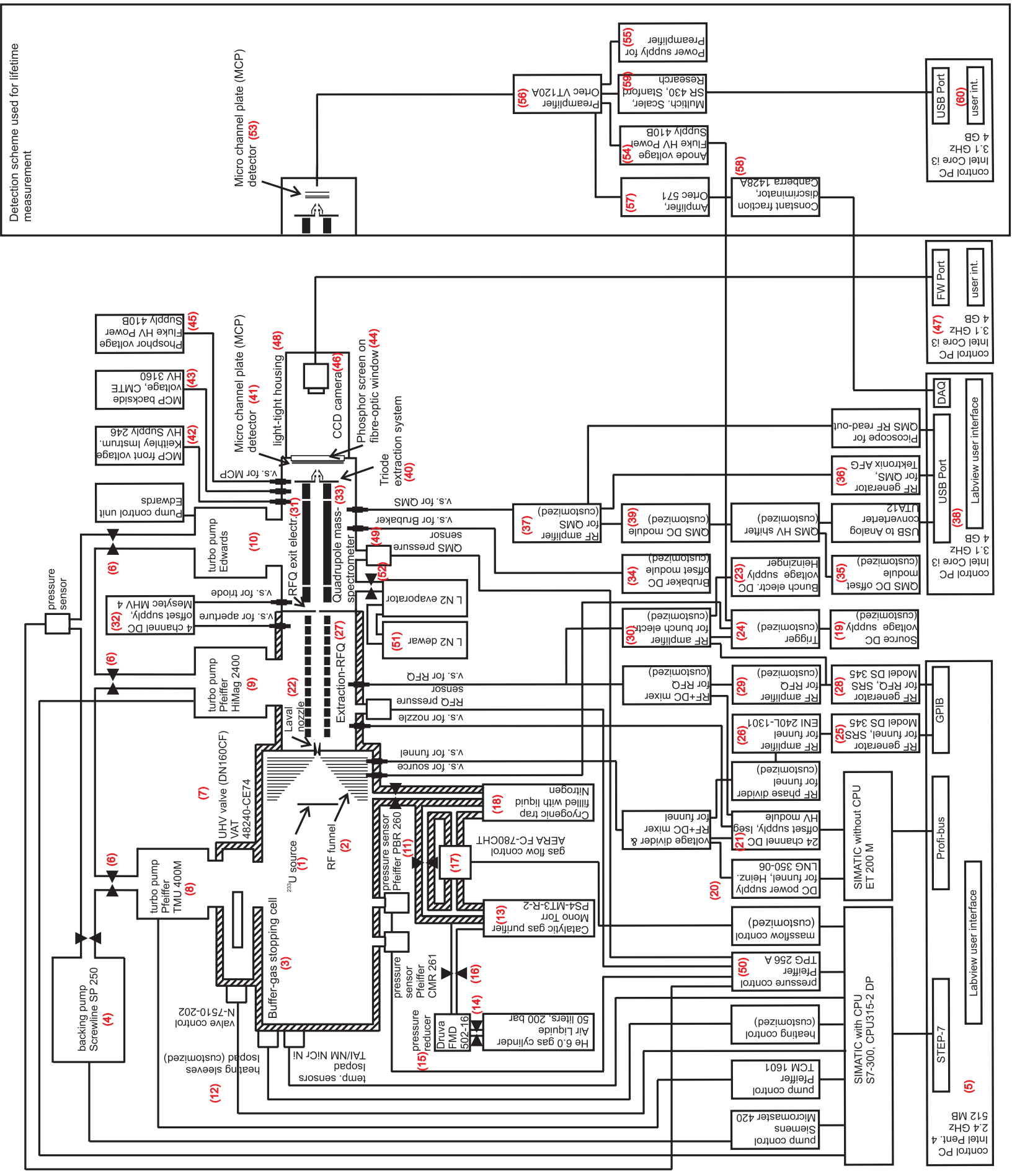

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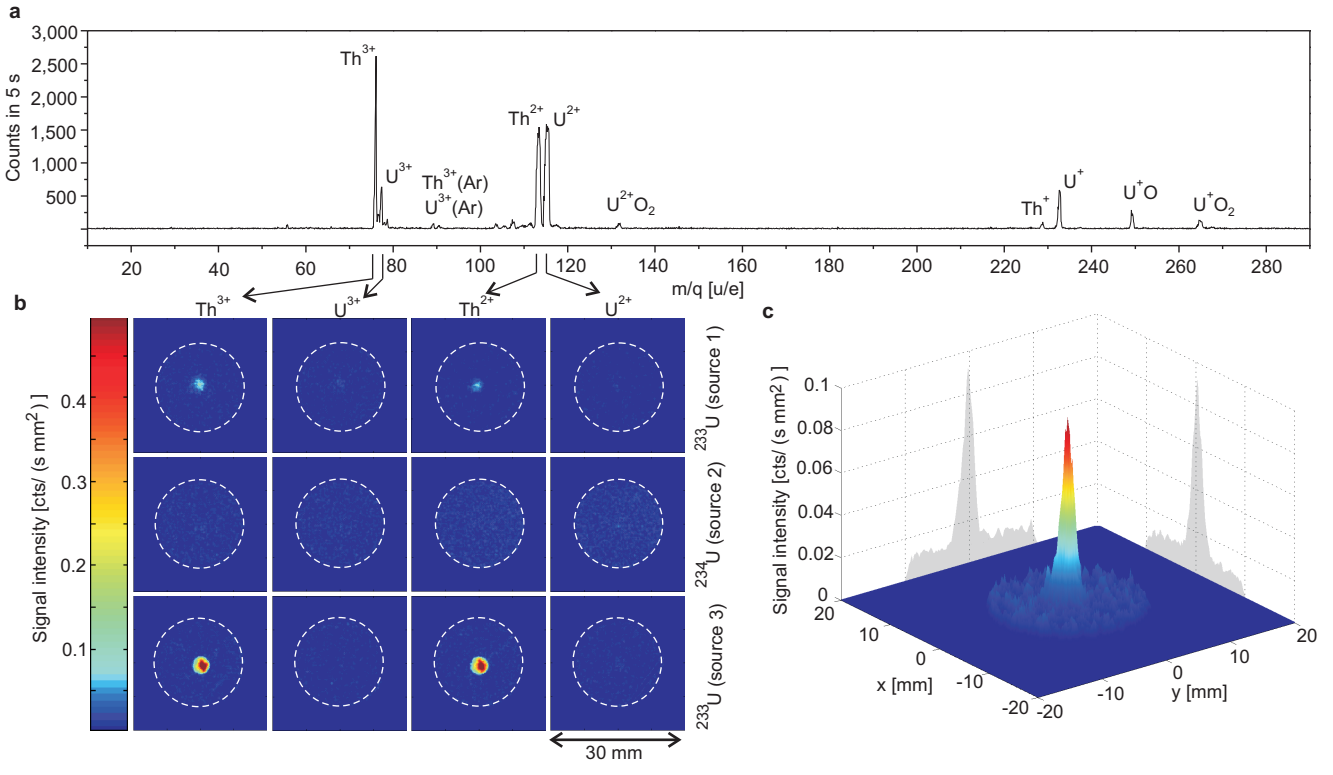

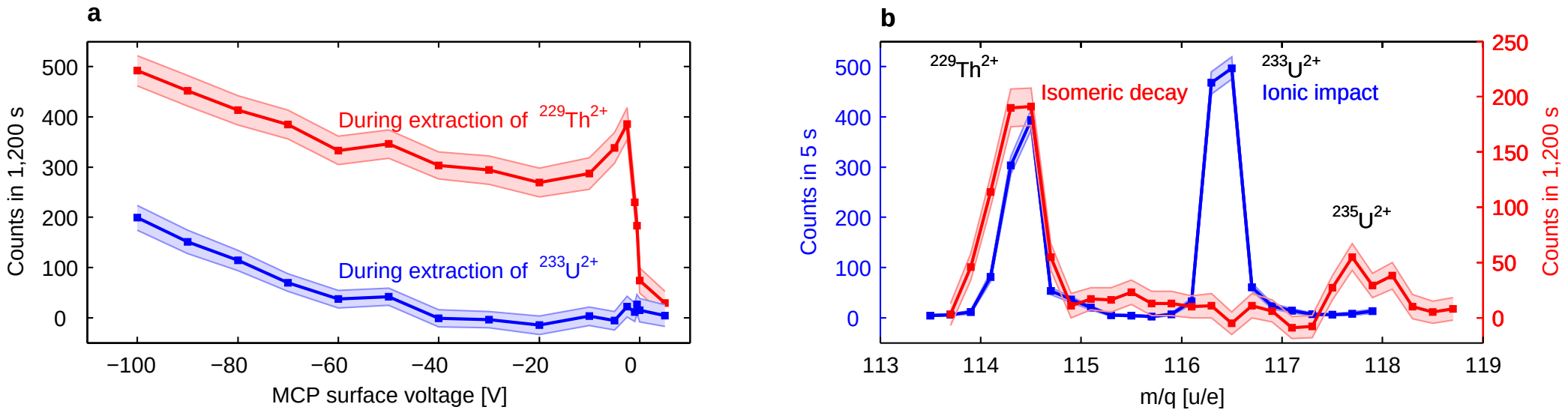

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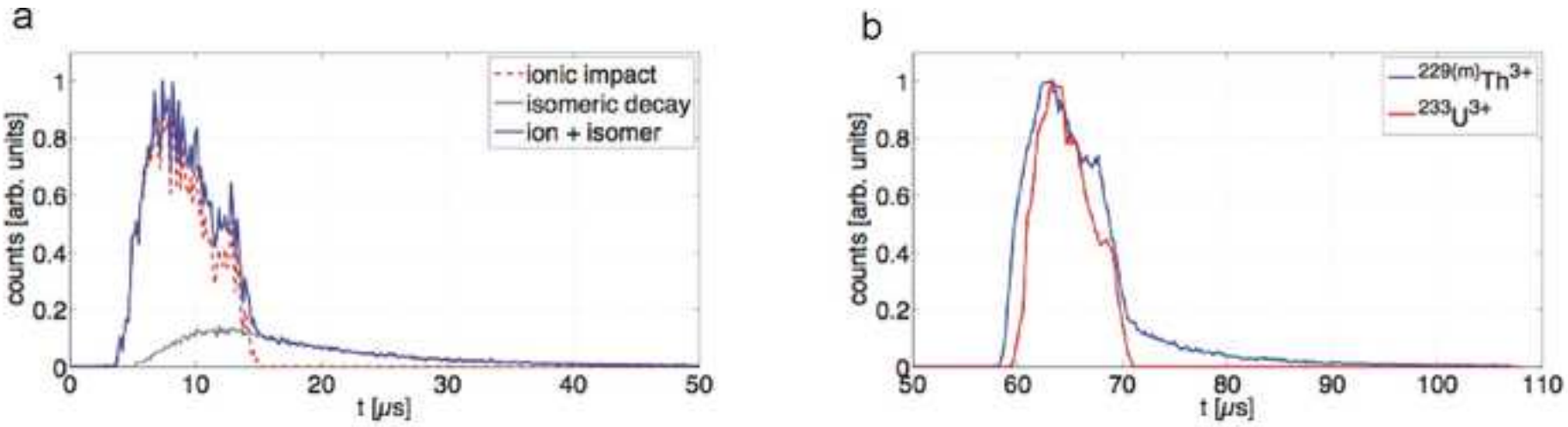

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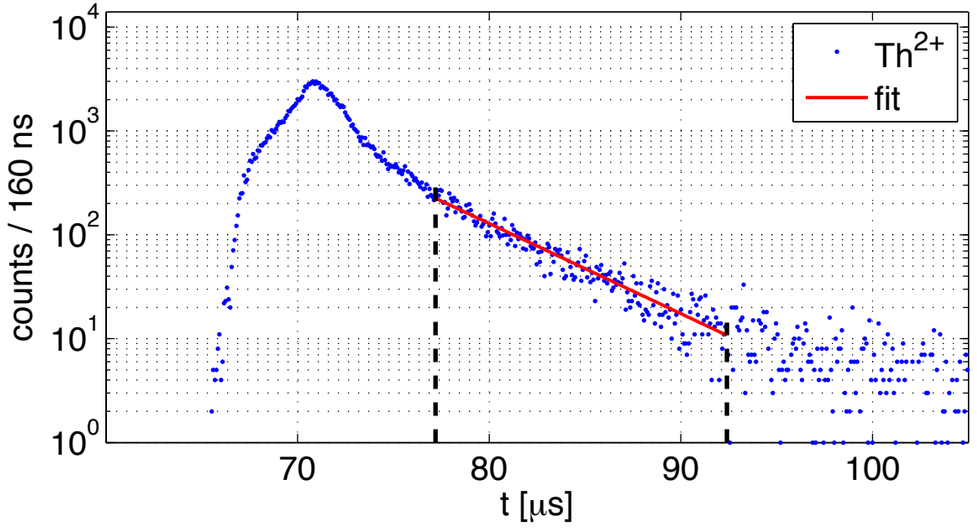

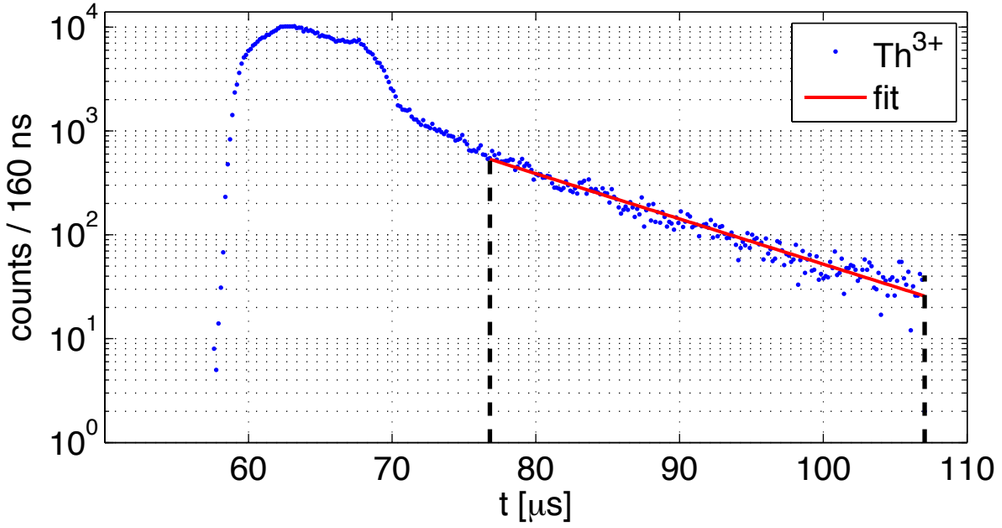

Name of Material/ Equipment	Company	Catalog Number	Comments/Description
Uranium-233 Source	Institut für Radiochemie Universität Mainz	customized	290 kBq U-233 deposited onto 90 mm diameter
RF funnel	Secamus Laserschneidtechnik GmbH	customized	50 ring electrodes, laser cut and electropolished
Buffer-gas stopping cell	Workshop of LMU Munich	customized	Vacuuchamber DN200 CF for buffer-gas stopping cell
Roughing pump	Leybold	Screwline SP 250	Roughing pump for entire system
Roughing pump control	Siemens	Micromaster 420	Control unit for Screwline SP 250
Vacuum gauge Prepressure	Pfeiffer	TPR 265	Pressure control for roughing pump
Vacuum gauge cell 1	Pfeiffer	CMR 261	Pressure control for cell (high-pressure range)
Vacuum gauge cell 2	Pfeiffer	PBR 260	Pressure control for cell (low-pressure range)
Vacuum gauge RFQ	Pfeiffer	PKR 261	Pressure control for RFQ pressure read-out
Pressure gauge QMS	Pfeiffer	PKR 261	Pressure control for QMS pressure read-out
Pressure control unit	Pfeiffer	TPG 256 A	Control unit for all pressure gauges
Control PC 1	Fujitsu	unknown	Control computer for buffer-gas stopping cell
Simatic with CPU	Siemens	S7-300	Simatic for automation and control
Simatic without CPU	Siemens	ET 200M	Simatic for automation and control
Vacuum valves	SMC	XLH-40	Vacuum valves for evacuation control
UHV gate valve	VAT	48240-CE74	Gate valve for cell closing during operation
Turbo-Molecular pump 1	Pfeiffer	TMU 400M	Turbo pump for cell
Control unit for TMP 1	Pfeiffer	TCM 1601	Control unit for TMP TMU 400M
Turbo-Molecular pump 2	Pfeiffer	HiMag 2400	Trubo pump for RFQ
Turbo-Molecular pump 3	Edwards	STP 603	Trubo pump for QMS
Control unit for TMP 3	Edwards	SCU-800	Control unit for TMP Edwards STP 603
Bypass valve of gas tubing	Swagelok	SS-6BG-MM	Valve to bypass the mass-flow controller
Heating sleeves	Isopad	customized	Heating sleeves for bake out of cell and RFQ
Temperature sensors	Isopad	TAI/NM NiCrNi	Temperature sensors for bake-out system
Heating control unit	Electronic workshop of LMU Munich	customized	Control unit for Isopad heating sleeves

Catalytic gas purifier	SAES MonoTorr	PS4-MT3-R-2	Gas purifier for ultra-pure helium supply
He gas cylinder	Air Liquide	He 6.0, 50 liters	Helium of 99.9999 % purity
Pressure reducer	Druva	FMD 502-16	Pressure reducer for He gas cylinder
Valve of gas supply	Swagelok	SS-6BG-MM	Valve to open or close the gas supply
Mass flow control	AERA	FC-780CHT	Mass flow control valve for He supply
control unit for mass flow valve	Electronic workshop of LMU Munich	customized	Control unit for AERA mass flow control
Gas tubing	Dockweiler	Ultron	electropolished gas tubing for He supply
Cryogenic trap	Isotherm	unknown	cryogenic trap for He purification (optional)
DC voltage supply for source	Electronic workshop of LMU Munich	customized	DC offset voltage supply for U-233 source
DC voltage supply for funnel	Heinzinger	LNG 350-6	Power supply for DC gradient of funnel
DC voltage supply for RFQ	Iseg	unknown	DC voltage supply for funnel offset, nozzle and RFQ
Laval nozzle	Friatec AG	customized	Laval nozzle for He and ion extraction
DC voltage supply for buncher	Heinzinger	LNG 350-6	DC supply for bunching electrode
Trigger module	Electronic workshop of LMU Munich	customized	Trigger module for bunched operation
RF generator for funnel	Stanford Research Systems	SRS DS 345	RF generator for funnel
RF amplifier for funnel	Electronic Navigation Industries	ENI 240L-1301	Rf amplifier for funnel
RF phase divider for funnel	Electronic workshop of LMU Munich	customized	RF phase divider for funnel
RF+DC mixer for funnel	Electronic workshop of LMU Munich	customized	Voltage divider and RF+DC mixer for funnel voltage
Extraction RFQ	Workshop of LMU Munich	customized	Extraction RFQ for ion-beam formation or storage
RF generator for RFQ	Stanford Research Systems	SRS DS 345	RF generator for RFQ
RF amplifier for RFQ	Electronic workshop of LMU Munich	customized	RF amplifier for RFQ
RF amplifier for bunch electrode	Electronic workshop of LMU Munich	customized	RF amplifier for bunch electrode

RF+DC mixer for RFQ	Electronic workshop of LMU Munich	customized	Mixes the RF and DC potentials for RFQ voltage
RFQ exit electrode	Workshop of LMU Munich	customized	2-mm diameter exit aperture for differential pumping
4 Channel DC supply	Mesytec	MHV 4	DC offset for aperture and triode
QMS	Workshop of LMU Munich	customized	Quadrupole mass separator for m/q selection
Brubaker DC offset module	Electronic workshop of LMU Munich	customized	DC offset supply for Brubaker lenses of QMS
QMS DC offset module	Electronic workshop of LMU Munich	customized	DC offset supply for QMS
USB-to-Analog converter	EA Elektro-Automatik	UTA12	to generate signal for QMS HV shifter
QMS HV shifter	Electronic workshop of LMU Munich	customized	to shift the voltage of the QMS DC module
QMS DC module	Electronic workshop of LMU Munich	customized	Module to provide DC voltages for QMS
RF generator for QMS	Tektronix	AFG 3022B	RF generator for QMS
RF amplifier for QMS	Electronic workshop of LMU Munich	customized	RF amplifier for QMS
Picoscope	Pico Technology	Picoscope 4227	Oscilloscope for QMS RF control
Control PC 2	Fujitsu	Esprimo P900	Control computer for QMS
Triode extraction system	Workshop of LMU Munich	customized	Set of three ring electrodes to guide ions
MCP detector	Beam-Imaging-Solutions	BOS-75-FO	MCP detector with phosphor sreen
DC voltage supply for MCP	Keithley Instruments	HV Supply 246	Voltage supply for MCP front side
DC voltage supply for MCP	CMTE (NIM module)	HV 3160	Voltage supply for MCP back side
DC voltage supply for MCP	Fluke	HV Supply 410B	Voltage supply for phosphor sreen
CCD camera	PointGrey	FL2-14S3M-C	CCD camera for image recording
Control PC 3	Fujitsu	Esprimo P910	Control computer for CCD camera
Light-tight housing	Workshop of LMU Munich	customized	Light tight wooden box for CCD camera
Dewar for LN2 supply	Isotherm	unknown	Dewar to provide dry nitrogen for venting

Evaporator for LN2	Workshop of LMU Munich	customized	Evaporator to provide dry nitrogen
Single anode MCP detector	Hamamatsu	F2223	Single anode MCP for lilfetime measurement
DC voltage supply for MCP	Fluke	HV supply 410B	Voltage supply for MCP anode
Power supply for preamplifier	Delta Elektronika	E 030-1	Power supply for preamplifier
Preamplifier for MCP signals	Ortec	VT120A	Preamplifier for MCP signals
Amplifier for MCP signals	Ortec (NIM module)	Ortec 571	Amplifier for MCP signals
CFD	Canberra	1428A	Constant-fraction-discriminator for MCP signals
Multichannel Scaler	Stanford Research	SR 430	Multichannel scaler for signal read-out
Control PC 4	Fujitsu	Esprimo P920	Control computer for scaler read-out
Labview	National Instruments	various versions	Program used for measurement control
Matlab	Mathworks Inc.	version 7.0	Program used for data analysis

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Author(s): Lars von der Wense, Benedict Seiferle, Peter G. Thirolf

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CORRESPONDING AUTHOR:

Name: Lars van der Wense

Department: Physics

Institution: Ludwig-Maximilians-University munich

Article Title: Preparation of an isotopically pure ^{229}Th ion beam for studies of ^{229m}Th

Signature: L. v. d. Wense Date: 25.5.2018

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Letter of Reply to the Editors and Referees.

We would like to thank the Editors and Referees for their careful reading of the manuscript, the many helpful comments and their positive decisions regarding the publication process.

In the following each point is shortly addressed directly in the comment letter:

Editorial comments:

Changes to be made by the Author(s):

1. Please take this opportunity to thoroughly proofread the manuscript to ensure that there are no spelling or grammar issues.

We have revised and proofread the manuscript.

2. Please revise lines 44-50 to avoid previously published text; see attached iThenticate report for details.

The lines have been completely revised in order to avoid previously published text.

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4. Please rephrase the Summary to clearly describe the protocol and its applications in complete sentences between 10-50 words: “Here, we present a protocol to …”

The summary has been rephrased.

5. Please remove the headers in the Introduction.

The headers have been removed.

6. Please define all abbreviations before use.

Abbreviations have been defined where required.

7. Please include a space between all numbers and their corresponding units: 15 mL, 37 °C, 60 s; etc.

The spaces have been introduced.

8. Please adjust the numbering of the Protocol to follow the JoVE Instructions for Authors. For example, 1 should be followed by 1.1 and then 1.1.1 and 1.1.2 if necessary. Please refrain from using bullets, dashes, or indentations.

The numbering of the Protocol has been completely revised and adapted to the Jove requirements.

9. Please reference figures showing experimental set-up in the Protocol.

As all numbers in the protocol refer to Fig. 2 of the manuscript, we have referenced to this figure once in the beginning of the protocol.

10. Please revise the protocol to contain only action items that direct the reader to do something. The actions should be described in the imperative tense in complete sentences wherever possible. Avoid usage of phrases such as “could be,” “should be,” and “would be” throughout the Protocol. Any text that cannot be written in the imperative tense may be added as a “Note.” Please revise A.4.4, A.5.6, A.5.7, A.6.1, B.5.4, B.6.5, B.6.6, B.6.7, B.7.1, B.7.5, etc., accordingly.

The mentioned protocol steps have been accordingly revised.

11. Please add more details to your protocol steps. Please ensure you answer the “how” question, i.e., how is the step performed? Alternatively, add references to published material specifying how to perform the protocol action.

The following references have been added to provide further background information: [30,31,32,33,34,36].

12. A.5.7: Please describe how to perform a mass scan with the QMS user Interface.

The required information has been added.

13. A.6.6, B.6.5, B.6.7: Please provide more details here.

More details have been provided.

14. Please include single-line spaces between all paragraphs, headings, steps, etc., and align all text to the left margin. After that, please highlight 2.75 pages or less of the Protocol (including headings and spacing) that identifies the essential steps of the protocol for the video, i.e., the steps that should be visualized to tell the most cohesive story of the Protocol.

The text has been accordingly edited. About 2.75 pages have been marked in yellow.

15. Please include volume and issue numbers for all references.

All required information is provided for all references.

16. Although the majority of this does involve a custom-built system, please include any other materials, equipment, and/or programs necessary to carry out this protocol in the Table of Materials.

A complete table of materials has been added.

Reviewers' comments:

Reviewer #1:

Manuscript Summary:

The manuscript describes in detail a setup and the operation procedure to realize a beam of Thorium ions, which a 2% fraction in the low-energy isomeric state. This state is of highest interest for a continuously growing community in atomic and nuclear physics as well as metrology.

Major Concerns:
None, the manuscript is clear and VERY detailed. The setup and protocol described here has lead to a multitude of breakthrough results and is currently the defining system in the field. Even if it is unlikely that someone will reproduce this setup to follow the described procedures, the individual technical and physical solutions found here are of great importance.

Minor Concerns: None

We thank Reviewer 1 for the report.

Reviewer #2:

Manuscript Summary:
The authors describe the methods to obtain isotopically pure ion beam of 229Th, which is important for the search of candidate nuclei of nuclear clock. The methods are used to and can be extended to get long-lived isotopically pure ion beams. The steps of the experiment are described in detail and clearly, and can be followed by the readers. The authors highlighted the critical steps in the provided protocol. The results are useful to the researchers who are concerning the nuclear clocks and isomers. However, the abstract is more like for scientific paper, it would be better if the authors rewrite it and provide more key experimental points. In the materials, the following improvements can be made:

We thank Reviewer 2 for the report. The abstract has been revised as proposed above. Each point will be discussed individually in the following.

1. the figures 1 and 2 are almost duplicate, it could be better to give one figure describing the experimental setups, pointing out the detection part for decay detection and lifetime measurement with inset, the other figure with all the diagrams/sketches. Maybe this is more appropriate for the JoVE.

We have followed this proposal and condensed figures 1 and 2 into a single figure (now Fig. 2). Further, a figure showing the experimental setup was added (now Fig. 1).

2. in figure 3, the upper three panels are not necessary, because the whole spectrum is presented in the upper panel of figure 4, which is a much nicer presentation. it would be

better to rescale the spectrum from 60 to 280. Only the lowest panel, the enlarged plots are useful. This means the authors have to reconsider how to arrange the figures.

We agree that some information given in previous Fig. 3 was redundant. We now reference to Fig. 4 (now Fig. 3) instead und have revised the text accordingly.

3. in line 436 and 441, Figure 6 (top panel)... and the middle panel of fig 6 ... where top and middle panels are not existing in the figure.

The corrections were implemented.

The method deserves for publication after the modifications suggested above.

Reviewer #3:

Manuscript Summary:

The submitted article presents a detailed overview and protocol that must be followed in order to successfully extract 229Th recoil ions and subsequent daughter isotopes from an ultra-pure buffer gas cell. As illuminated in the abstract and introduction, 229Th is a current hot topic, hosting the lowest energy isomeric state of any nucleus, indirectly deduced to be a few eV above the nuclear ground state. This unique state offers a rich playground for precision spectroscopy and applications, branching fields of nuclear physics, atomic physics as well as time and frequency standards. It is this latter application to which the authors pay particular attention as the relative linewidth of the ground-state transition is such that the isomer potentially qualifies as a nuclear frequency standard of unprecedented precision which, if realized, could be essential for a vast field of applications and fundamental tests. Until recently however the isomeric state itself had only been inferred from indirect measurements and much controversy arose over the years as to whether the radioative decay of the state had been seen. It is now known that this was not the case as a number of background signals can mimic the decay. The Munich team, with a number of outstanding recent results, conclusively proved the existence of the isomeric state through a detection of the internal conversion electrons emitted from the decay of the state in a neutral (atomic) form. In this current manuscript the procedure to repeat such measurements is presented in a clear and step-by-step manner such that other groups, if they had access to the sources and a similar setup would be able to identify the isomeric decay.

The isomer is populated via a 2% branch from the alpha decay of 233U. In order to spatially separate the resulting population from other contaminants including daughters in the radioactive decay chain of 233U, the authors choose to stop the recoil products in an ultra-pure buffer-gas stopping cell, guide them using an electrode system to the exit nozzle, inject

into a radiofrequency quadrupole and mass separate using a quadrupole mass spectrometer (QMS). Depending on whether direct detection of the isomeric state is the goal, or whether a lifetime measurement of the state is required, the ions may be gently impinged onto the surface of a multichannel plate detector coupled to a phosphor screen and CCD detector. A number of verification measurements have been supported to prove that registered electron signals indeed originate from the decay of the nuclear isomer.

Following an appropriate abstract and introduction, with an extensive reference list which suitably covers the current status in the field as well as recognizing the earlier (now historical) efforts, the protocol is presented. Mounting of the source, initial evacuation of the vacuum chamber and baking out of impurities in the system, preparation of the buffer gas and supply of ultra-pure helium, application of suitable electric guiding fields (and later the needed modifications required for ion bunching and lifetime measurements), tuning of the ion extraction and the mass separation and final detection of the state are detailed. In order to follow the steps two very detailed figures accompany this aspect of the article, with all components numbered accordingly. A set of representative results (already published prior to this manuscript) are given which present the reader with what one might expect to see, from a mass spectrum of ions extracted from the gas cell, to the signal obtained using the CCD camera which indicates direct detection of the isomeric state. This is useful as the results also indicate what one might expect to detect should a different isotope of uranium be used as a source - again, verifying the existence of the isomer populated in the decay of 233U. Finally, the critical steps are summarized, troubleshooting options are presented, and the method presented here is placed in context with other worldwide efforts which aim at the study of the isomeric state using vastly different methodologies. It should be noted that currently these other methods have either not worked or are currently the subject of significant discussion in the scientific community.

In the following I have some minor comments. The article as it is currently presented is suitable for publication and is thorough enough to be a useful resource for other researchers aiming to learn about this technique.

We thank the reviewer for this comprehensive report. In the following the comments will be considered individually.

1. Abstract, line 28-29, "enabling the investigation of…."

The abstract has been completely rephrased.

2. Line 48, "For the past 40 years…."

The first part of the introduction has been rephrased.

3. Line 107, it might be better to use the word "viewed" rather than "inspected" when referring to the CCD camera.

This has been corrected.

4. Line 108, "…the underlying procedure to generate…"

This has been corrected.

5. Line 108, "foundation for studying"

This has been corrected.

6. In the protocol of part A 1.1, the authors state that the source thickness should not exceed 16 nm, however surely this could be exceeded? The only drawback is that for a given source strength, the overall recoil efficiency would be reduced due to the limited range of 229Th recoils in the uranium. But this would presumably be a matter of handling stronger sources with redundant source material.

This is of course correct and the sentence has been accordingly rephrased.

7. In protocol 2 and 3 the evacuation of the chamber and bake out procedure is presented, followed by the preparation of the gas supply. In this procedure I do not see any explicit mention of monitoring the pressure in the subsequent vacuum chambers. This could be noted, especially after the filling of 30 mbar of helium into the buffer gas cell, so that one might understand the baseline pressures to be expected in the QMS chamber.

The information has been added.

8. Line 191, protocol 4.4. After this first line I suggest to add (27) as this is where the extraction RFQ is mentioned for the first time.

The information has been added.

9. Line 249, protocol 5.6. "…passing through the QMS…"

This has been corrected.

10. Protocol 6.4. Here an accelerating potential of typically +6000 V is applied to the phosphor screen, however in protocol 5.3 only +5000 V is applied. Does this additional voltage make much difference?

For electron attraction the voltage difference between the second MCP plate and the phosphor screen is set to +4000 V. The second MCP plate is changed from +900 V (Step 5.2) to +1900 V (Step 6.4). The phosphor screen is accordingly adapted. In case that this adaptation is not performed, the kinetic electron energy at impact will be reduced by 1/4. This may slightly reduce the brightness of the spot produced on the phosphor screen during electron impact, but would most likely not affect the concept.

11. Line 290, protocol 1.10. Would it also be an option to vent the system with dry nitrogen from a bottle for example, or is it preferable to use the boiled off nitrogen which seems to be suggested here?

Indeed this would be an alternative and has been added as a note. However, when venting with nitrogen from a bottle, care has to be taken (e.g. by adding an overpressure valve to the vacuum system) not to exceed atmospheric pressure and enter an overpressure regime.

Thus the vacuum system would become (unnecessarily) slightly more complex.

12. Line 339, "The trigger module allows an adjustment of…"

This has been corrected.

13. Protocol 7.5. Can you verify whether the trigger module which is labeled (30) here should be (24)?

Indeed that was a mistake in labeling.

14. Representative results. In the first paragraph it is stated that the 229Th3+ and 233U3+ are extracted with about equal intensities. This is also the same for the doubly-charged state according to Fig. 3, however not the case for the singly-charged state. It is claimed to be noteworthy, however no explanation of why this is noteworthy is given. I suggest to expand a little here on why this may be important.

An explanation has been added.

15. Line 481. "Besides" should be worded, "In addition to…."

This has been corrected.

16. Line 500, "…for a given source activity"

This has been corrected.

17. Line 515-516 is a little confusing. Perhaps, "A successful application of voltage is monitored via the current… This current will increase in the case of sparks. If sparks have occurred…"

The sentence has been rephrased in accordance with the proposal.

18. Line 576-577, "Thus the present paradigm that this would require….can be circumvented".

This has been corrected.

19. Please note that both figures 1 and 2 are currently very poor quality which needs addressed. It was very hard to read the numbering and text written in these figures - which are the basis of much of the discussion in the protocol list.

We provide the figure now as a vector graphic and hope that this will solve the problem of readability.

20. I noticed that the valve immediately after the backing pump was not labeled. Should this also be (6)? Is it worth an additional mention or is this valve always open?

This valve is a safety valve which remains open during normal operation, for this reason we think that no labelling is required at this point.

Reviewer #4:

Manuscript Summary:
The authors propose to visualize the preparation of a pure 229Th ion beam for studies of 229mTh. The isomer of this isotope has an energy of 7.8 (5) eV. At neutral thorium it decays via internal conversion with a half-life of 7 µs. In charged states this channel is energetically not allowed and a radiative decay is the only possible decay channel which has a lifetime, according to calculations, of up to 10.000 s resulting in a very narrow line width. As pointed out in the Introduction of the manuscript there are many potential and rather important applications relying on such an isomer.
No doubt, the presented material is valid. It has been presented prior to this paper in highly rated refereed journals.

We thank Reviewer 4 for the report. In the following, the points will be discussed individually.

Major Concerns:
The authors focus in the yellow highlighted part on the preparation procedure of the isomeric beam and the detection of the isomeric decay. It describes in detail the rather involved figure 1 which indicates the essential parts of the protocol to be filmed. I strongly support the production of such a film although I have no idea how this can be done on the basis of such an involved diagram. However, this is not my task as has been pointed out in the criteria for reviewing the article. Anyway, I have the feeling that the film must show the experimental setup and the signal generation, e.g., on the basis of figure 2 and 3 of the Nature Vol. 533 article. Such figures are missing in the manuscript. I do not know whether this is of importance for the film production, or not.

We partly follow the proposal and have added a figure showing a drawing of the experimental setup (new Fig. 1). We consider the physical details of the detection process to be of minor importance in this rather technical journal format. This, however, can be subject to further discussion also with the editors.

Minor Concerns:
Line 62: Add explanations of the acronyms GPS, GLONASS or GALILEO in words.

The explanation of the acronyms has been added as far as possible. As it turned out “Galileo” is not an acronym but rather the name of the program.
Line 414: The quoted charge state for thorium 3+ should be 2+.

This has been corrected.

Line 419: The voltage -900 V is inconsistent with regard to the nature article and the figure caption.

This has been corrected.

Line 436: Figure 6 (top panel) is not in accord with the shown figure where it is a left panel.

This has been corrected.

Line 441: The middle panel of Figure 6 does not exist as well in the shown figure.

This has been corrected.

The table JoVE_Materials is empty.

A complete Material list has been added.

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Name:	Benedict Seiferle
Affiliation:	Individual
Email Id:	benedict.seiferle@physik.uni-muenchen.de
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