^{229m}Th and its Application as a Nuclear Clock

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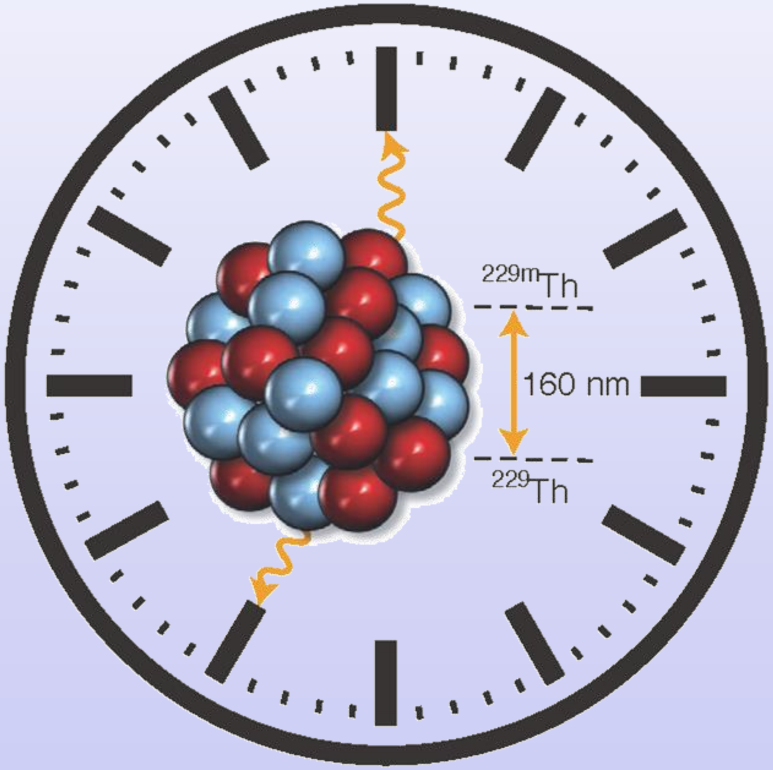
- ^{229m}Th properties and prospects
- Experimental approach & setup
- Measurements on ^{229m}Th:
  - first direct identification
  - halflife
  - hyperfine structure
  - ongoing: excitation energy
- Summary & Perspectives



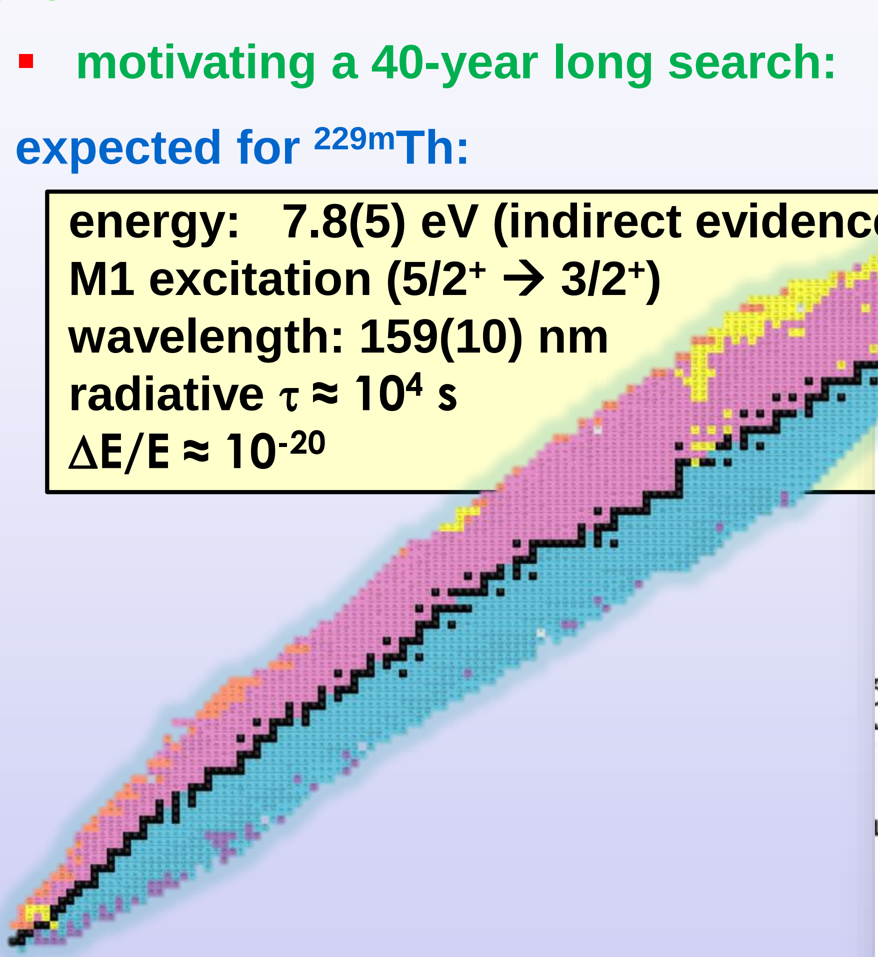
Unique properties of the 229-Thorium Isomer

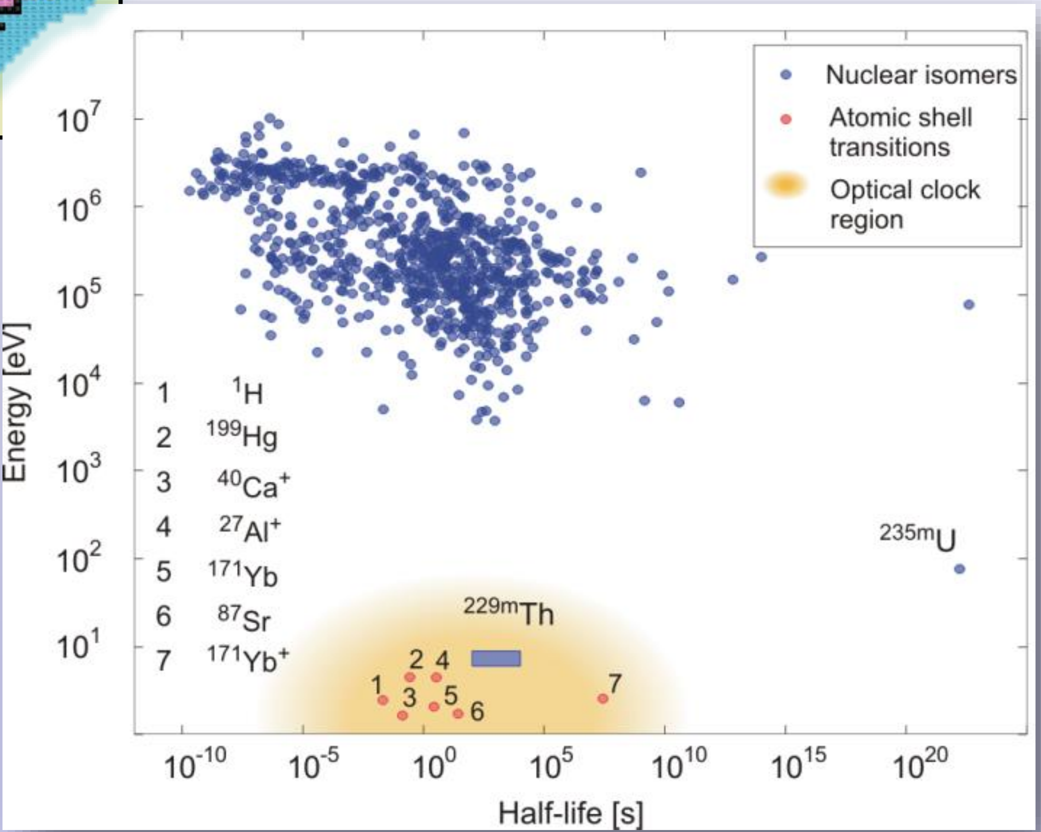
- motivating a 40-year long search:

expected for ^{229m}Th:

energy: 7.8(5) eV (indirect evidence)
M1 excitation $(5/2^+ \rightarrow 3/2^+)$
wavelength: 159(10) nm
radiative $\tau \approx 10^4$ s
$\Delta E/E \approx 10^{-20}$

→ extremely stable nuclear frequency standard: 'nuclear clock'





lowest E* of all ca. 176000 presently known nuclear excited states

L.A. Kroger and C.W. Reich, Nucl. Phys. A 259 (1976) 29
B.R. Beck et al., PRL 98 (2007) 142501

Potential Applications of Nuclear Clocks

- **satellite-based navigation** (GPS, Galileo..):

  presently: ‘thermal’ microwave atomic clocks (Rb, Cs)
  positioning precision: 1-10 m

  goal: improved positioning precision to cm – mm
  - autonomous driving
  - freight-/ component tracking …

- **time variation of fundamental constants**

  - theoretical suggestion: temporal (spatial) variations of fundamental “constants”
  - current limit: $\dot{\alpha}/\alpha = (-1.6 \pm 2.3) \cdot 10^{-17}\ \mathrm{yr}^{-1}$ Rosenband et al., Science 319, 1808 (2008)
  - temporal variation in transition energy of ^{229m}Th may provide enhanced sensitivity by (10^2 – 10^5) for fine structure constant $\dot{\alpha}/\alpha$ and strong interaction parameter (m_q/Λ_{QCD})

- **3D gravity sensor:** ‘relativistic geodesy’

  - best present clocks: detect gravitational shifts of ± 2 cm

    → allows long-distance potential difference measurements
  - precise, fast measurements of nuclear clock network:
    e.g. monitor volcanic magma chambers, tectonic plate movements

$$\boxed{\Delta f \big/ f = -\Delta U \big/ c^2}$$

f: clock frequency
U: gravitat. potential

Experimental Approach

concept:

- populate the isomeric state via 2% decay branch in the α decay of ^{233}U
- spatially decouple $^{229(m)}$Th recoils from the ^{233}U source: avoid background
- detect the subsequently occurring isomeric decay

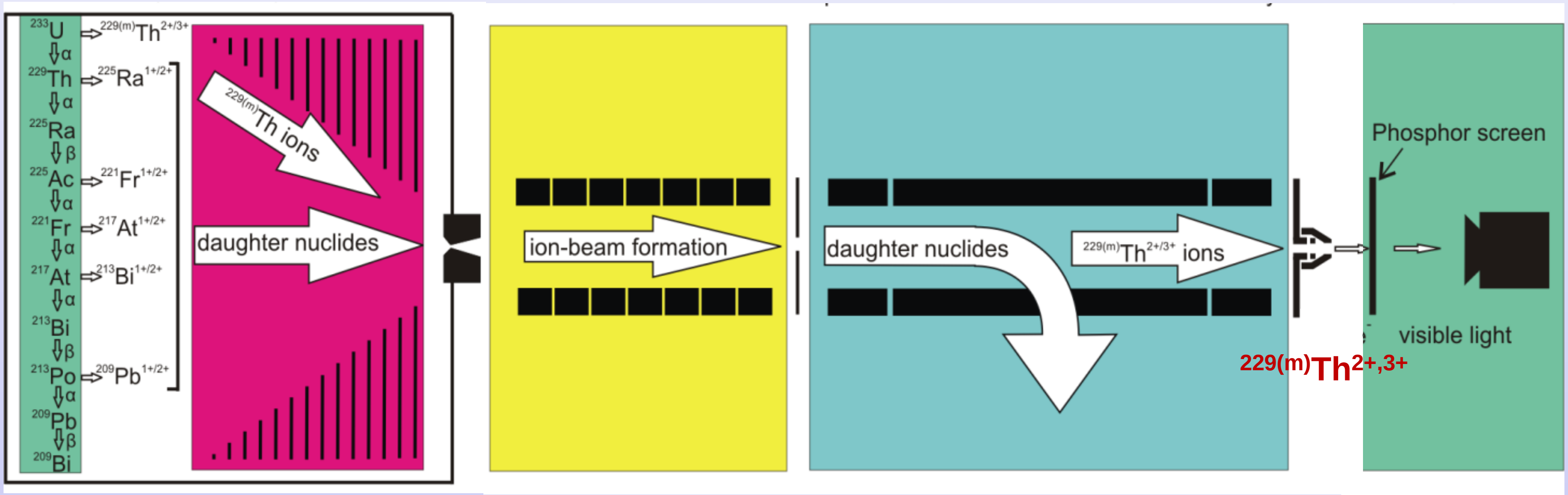
Buffer gas stopping cell

Laval nozzle

aperture electrode

triodic extraction electrode

detection system



^{233}U source

RF + DC funnel

RF quadrupole ion guide

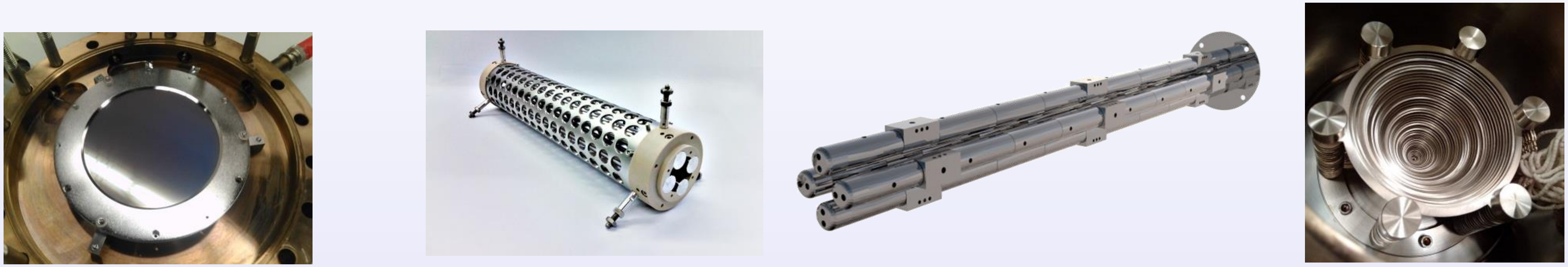
Quadrupole mass separator

MCP

CCD

Experimental Setup

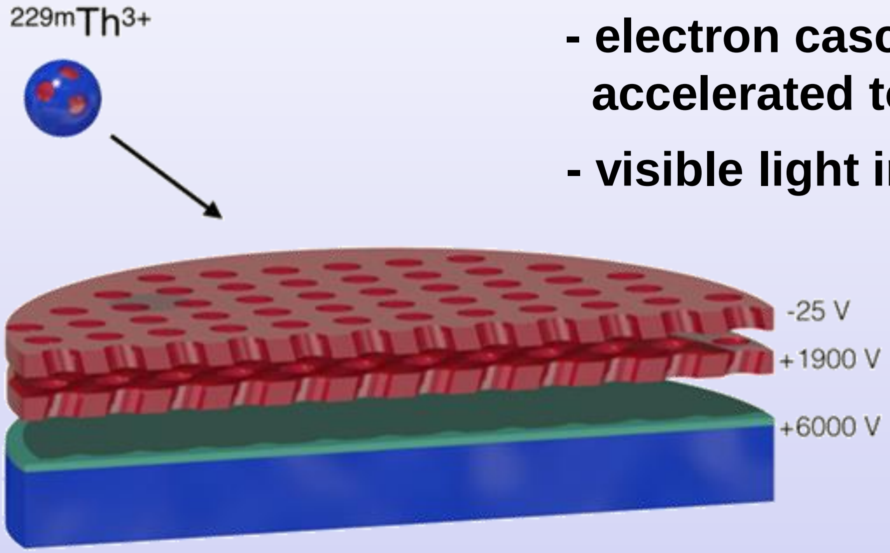
located at Maier-Leibnitz Laboratory, Garching:

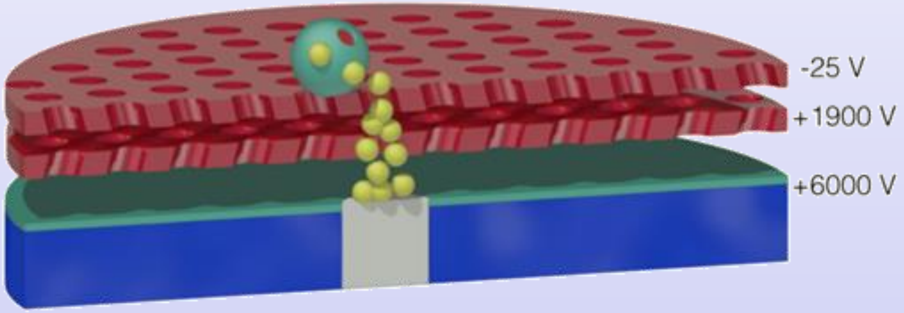


Isomer Detection Process

extracted $^{229(m)}Th^{3+}$ ions:

- impinging directly onto MCP surface behind triode exit
- ‘soft landing’ on MCP surface: avoid ionic impact signal
- neutralization of Th ions
- isomer decay by Internal Conversion: electron emission
- electron cascade generated, accelerated towards phosphor screen
- visible light imaged by CCD camera





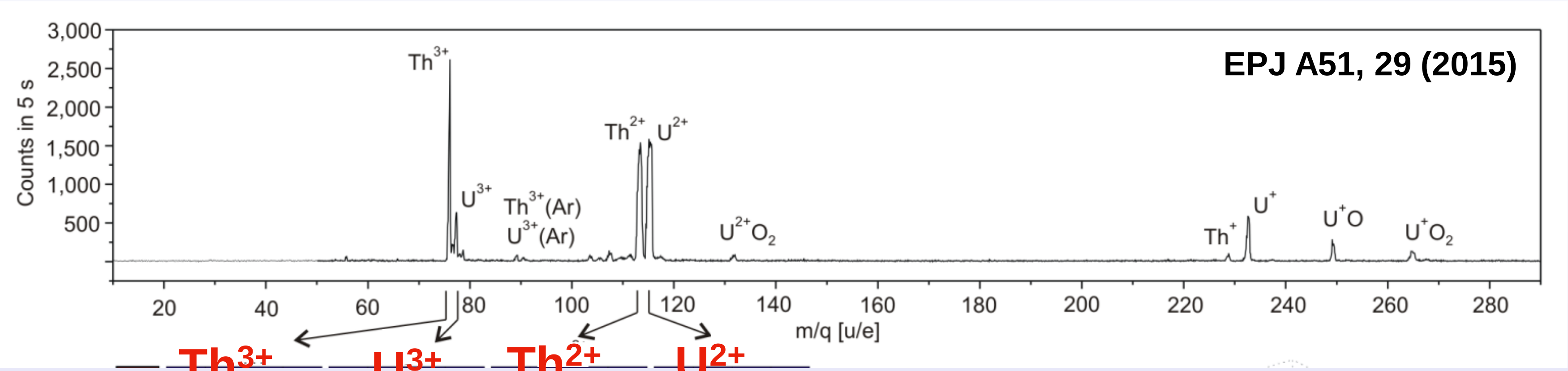
- internal conversion (IC) energetically allowed for neutral thorium:

  $I(Th^{+}, 6.31\ eV) < E^{*}(^{229m}Th, 7.8\ eV)$

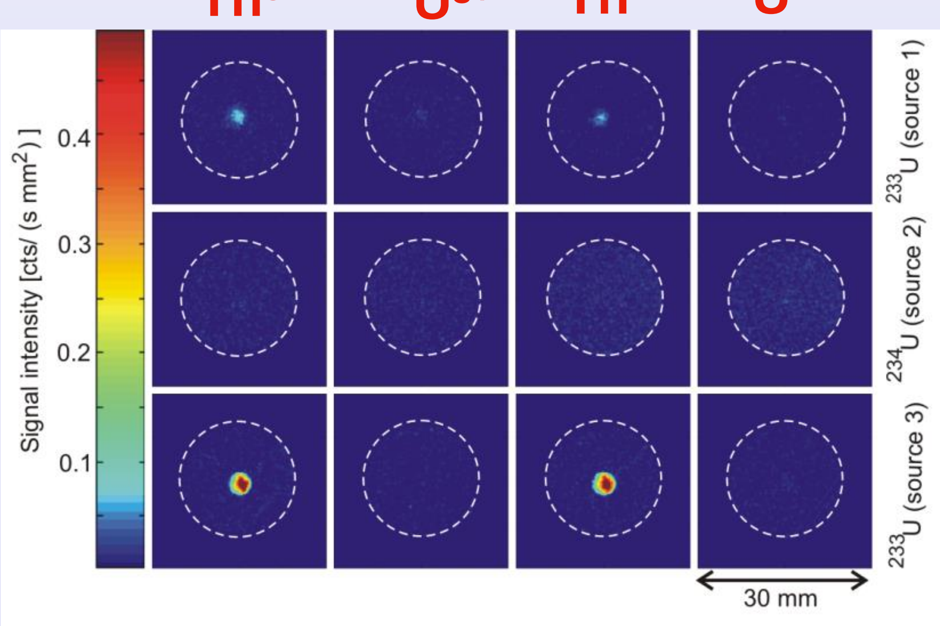
- isomer lifetime expected to be reduced by ca. 10^{-9} (from ~10^{4} s → ~ 10 μs)
- Th^{q+} ions: IC is energetically forbidden, radiative decay branch may dominate

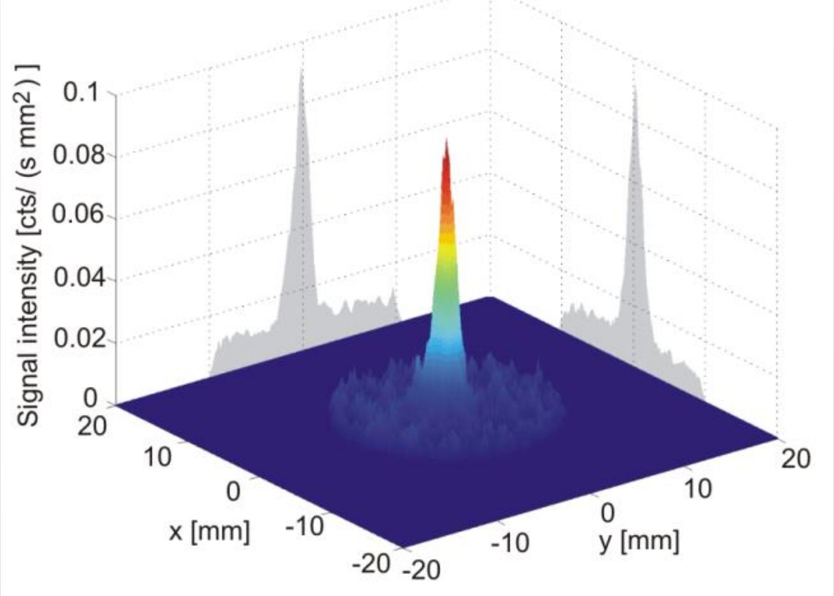
Direct Signal of IC Decay from ^{229m}Th

L. v.d. Wense, PT et al., Nature 533, 47-51 (2016)



EPJ A51, 29 (2015)





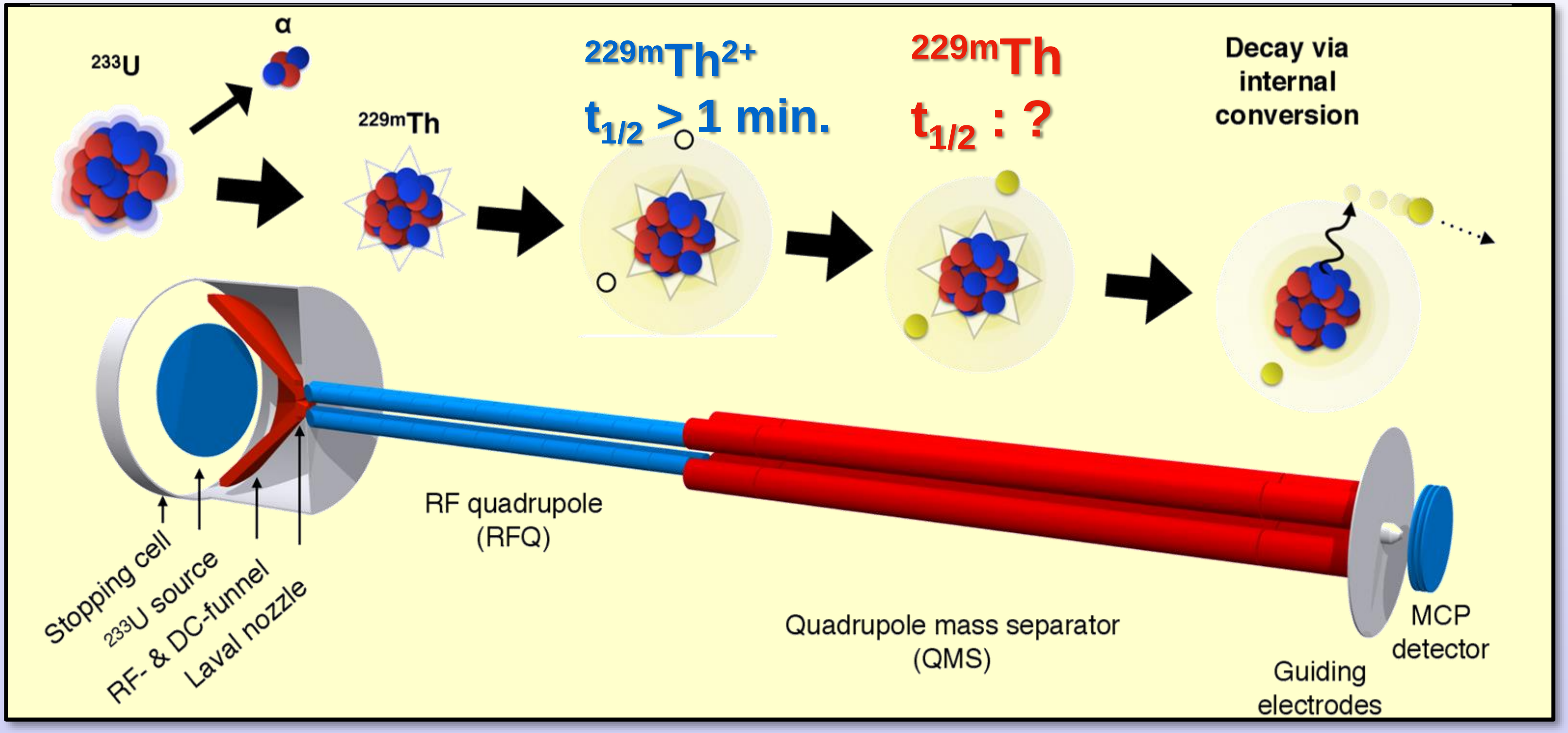
clear signal from Th^{3+}, Th^{2+}
no signal from U^{3+}, U^{2+}

all potential background contributions could be excluded

Next step: Halflife determination

use pulsed extraction from RFQ

- charged $^{229m}Th^{2+}$: $t_{1/2}$ > 1 min. (limited by RFQ storage time)
- neutral ^{229m}Th:

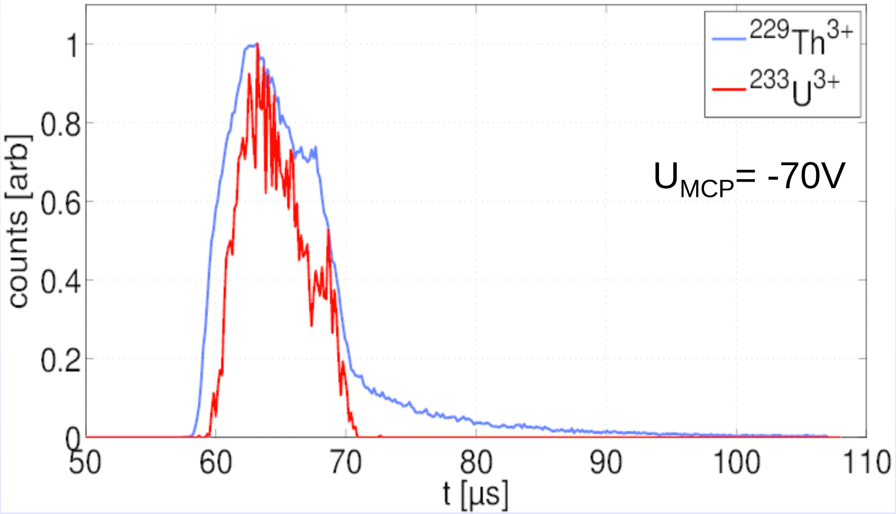


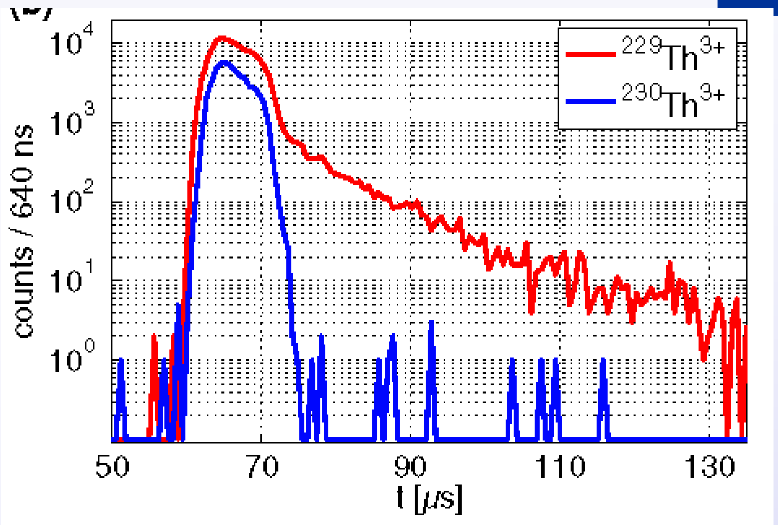
- expected conversion coefficient: $\alpha = N_e/N_\gamma \sim 10^9$

→ provides constraint for strength of photonic decay branch
(if IC cannot be suppressed, e.g. by suitable crystal lattice implantation)

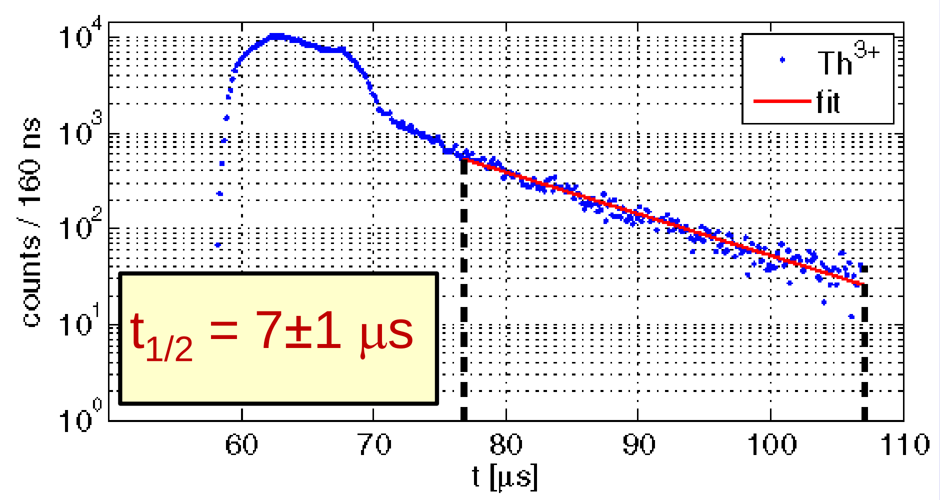
Halflife: Neutral ^{229m}Th

B. Seiferle, L. v.d. Wense, PT, PRL 118, 042501 (2017)





- bunch width: ca. 10 µs
- ca. 400 $^{229(m)}$Th$^{2+,3+}$ ions/bunch



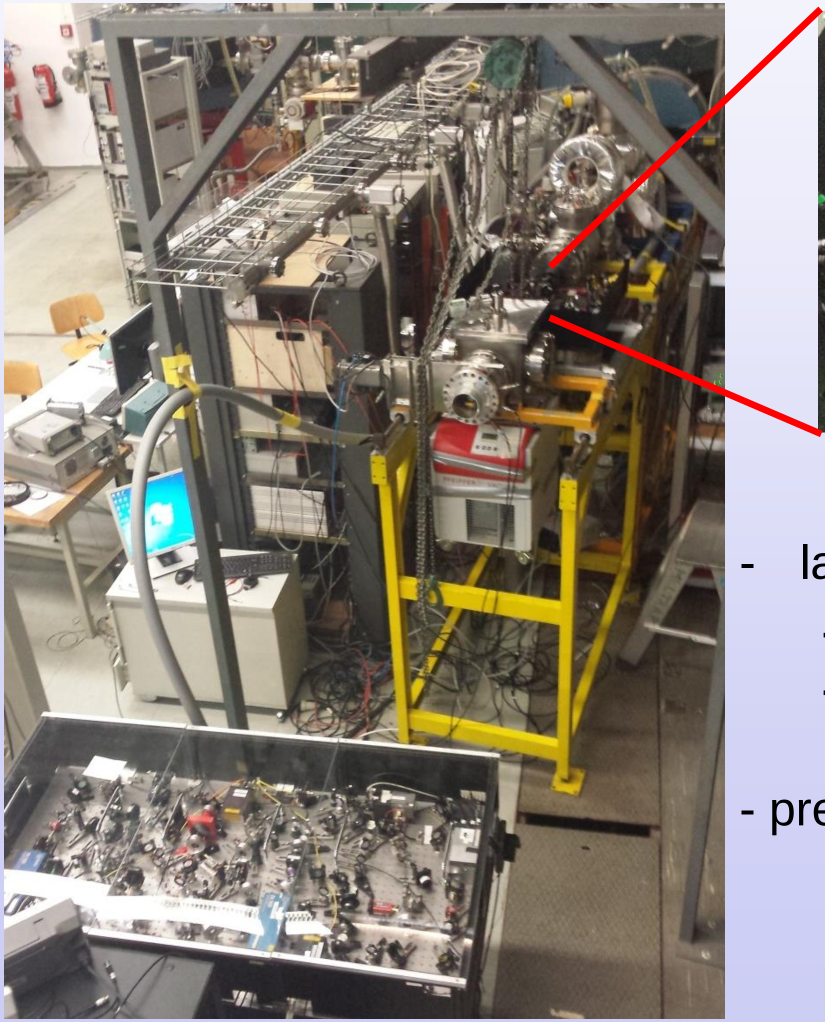
$t_{1/2}$ = 7±1 µs

→ confirms expected conversion coefficient: ~10^9

Collinear Laser Spectroscopy of ^{229m}Th

- Collaboration with PTB Braunschweig: (E. Peik, M. Okhapkin et al.):

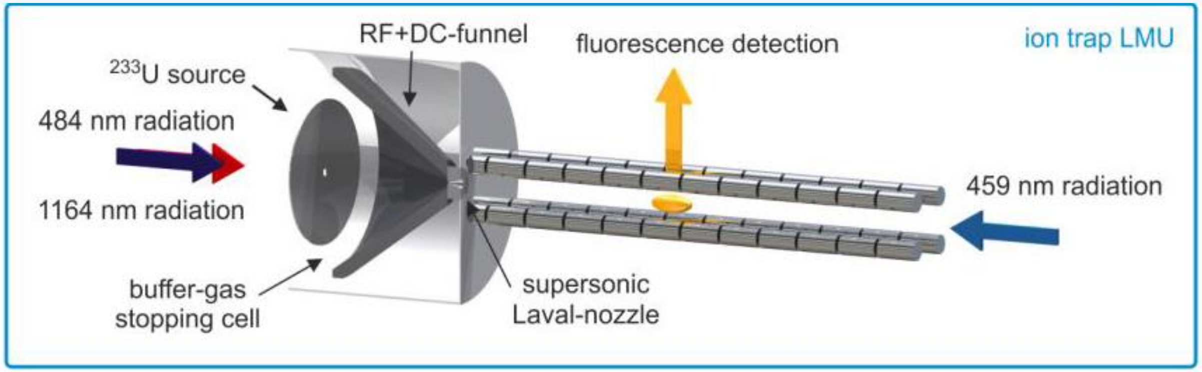
Goal: resolve hyperfine structure of $^{229m}Th^{2+}$: derive signature for nuclear excitation

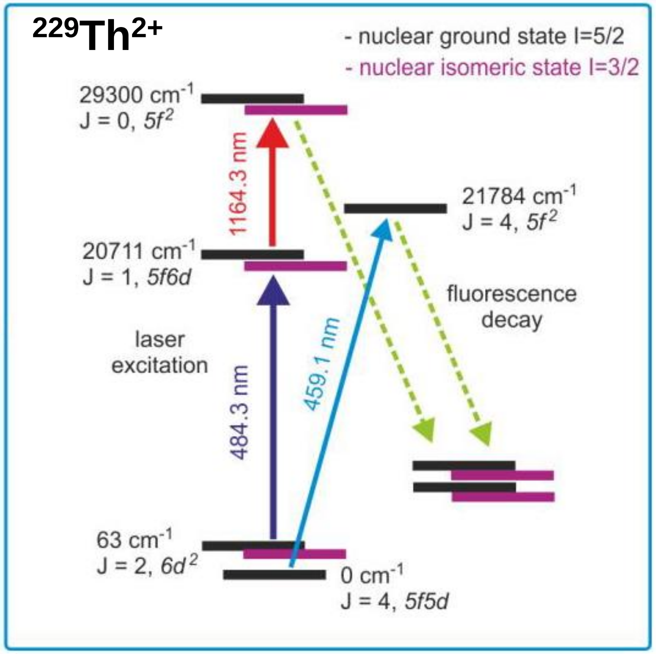


- laser excitation of $^{229(m)}Th^{2+}$ ions behind QMS:
  - → 3 external-cavity diode lasers
  - → co- and counter-propagating laser beams

- preparatory experiments on ^{229}Th at PTB Paul trap

Hyperfine Structure of ^{229m}Th





- Doppler-free two-step laser excitation (J=2→1→0):

  i) 484.3 nm: excites narrow velocity class of ions from thermal distribution to intermediate state
  - tuned in 35 steps of 120 MHz within Doppler profile

  ii) 1164.3 nm: intermediate state probed by tunable resonant excitation to final state
  - at each step of i): continuous scan over ≥ 4 GHz

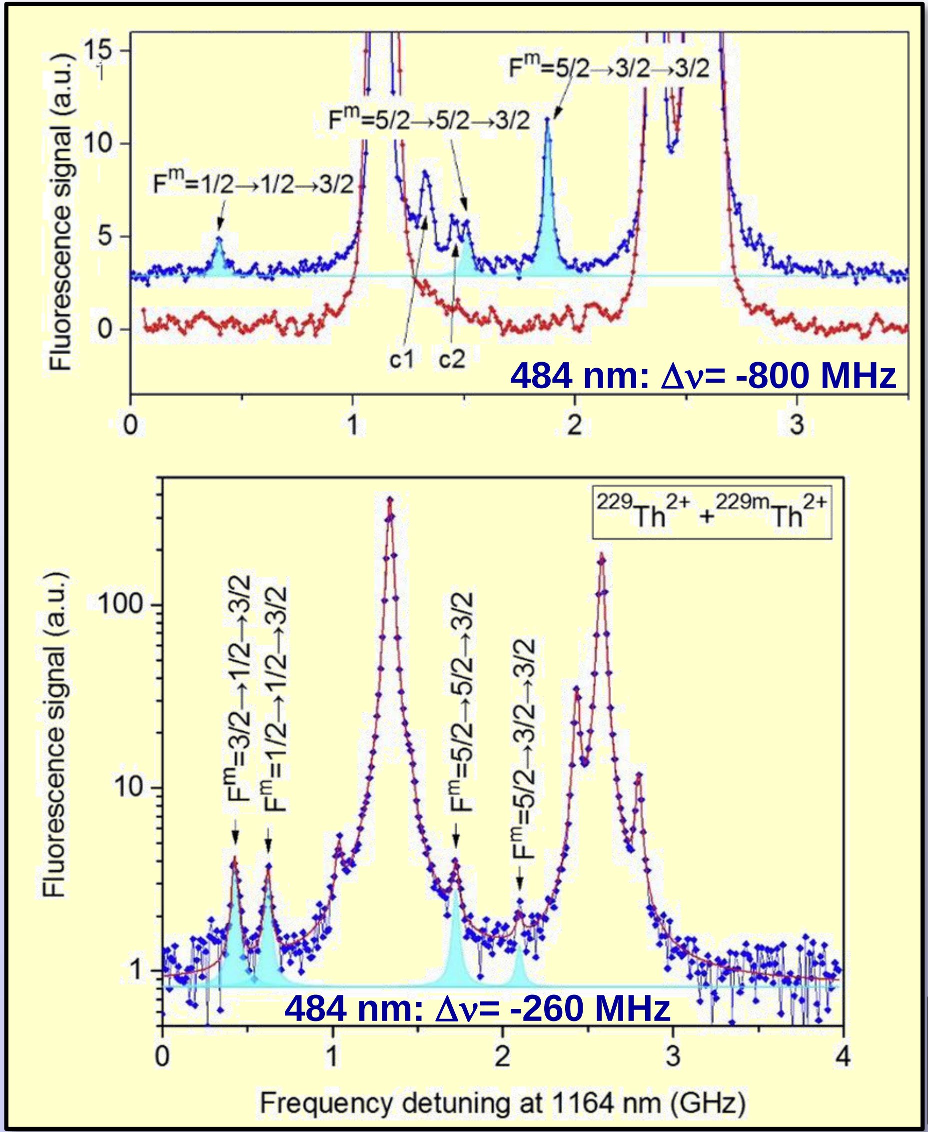
- Sensitive fluorescence detection:
  - decay channels at other wavelengths, free from laser stray light

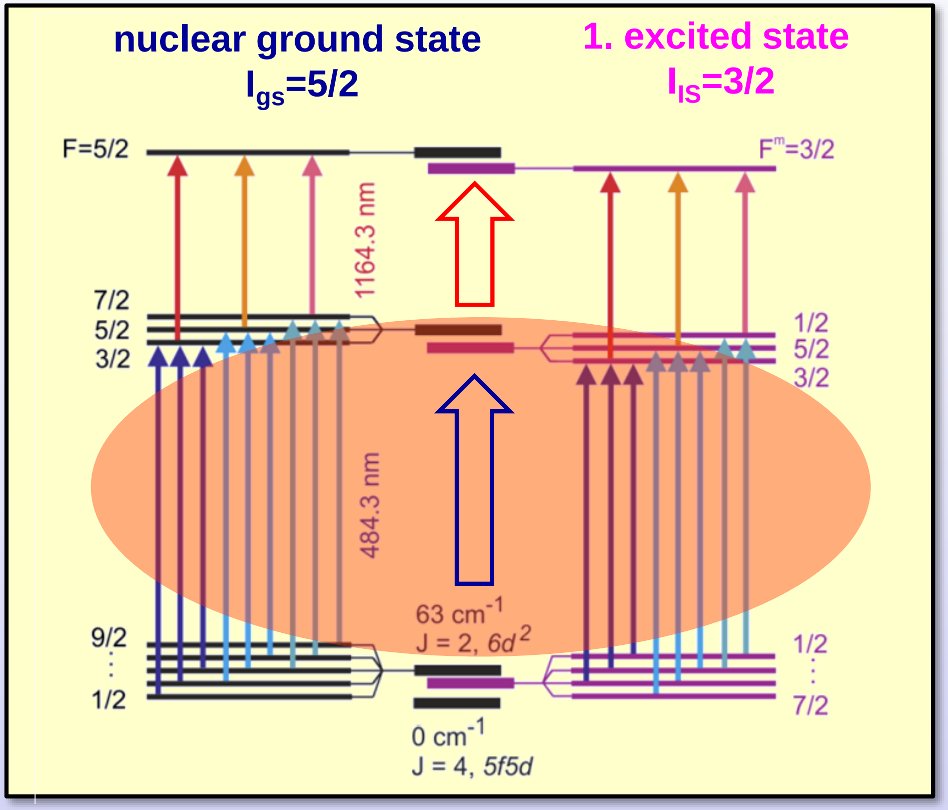
- 3rd laser at 459.1 nm: single-photon excitation to monitor amount of ^{229m}Th^{2+} in trap
  → normalization of fluorescence signals from HFS components

Hyperfine Structure of ^{229m}Th

J. Thielking,.., PT et al., Nature 556, 321-325 (2018)

2 examples from ca. 70 spectra:





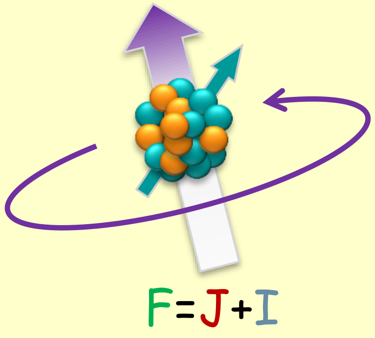
ground state: (I=5/2): 9 transitions

isomeric state: (I=3/2): 8 transitions

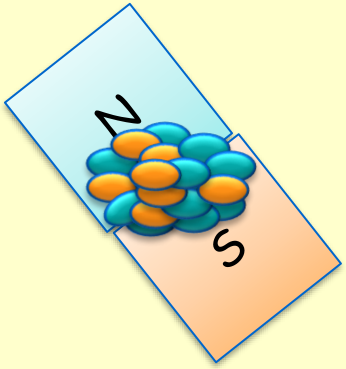
$$E_{HFS}(JIF)=\frac{1}{2}\boxed{A}K+\boxed{B}\frac{(3/4)K(K+1)-I(I+1)J(J+1)}{2I(2I-1)J(2J-1)}$$

What does HFS teach about ^{229m}Th ?

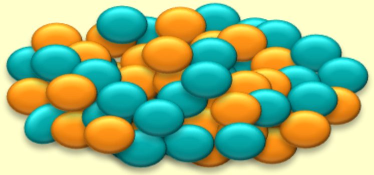
after 40 years of uncertainty about its existence:
deep insight in nuclear structure of the thorium isomer



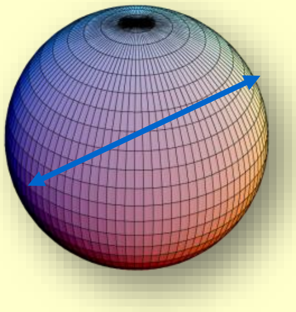
nuclear spin



magnetic
dipole moment



electrical
quadrupole moment



charge radius

$I = 3/2$

confirms level scheme

$\mu^m = - 0.37(6)\ \mu_N$

confirmed by recent theory

$Q_0{}^m = 8.7(3)$ eb

prolate deform.; $\dot{\alpha}$ sensitivity

$\langle r^2\rangle^{229m} - \langle r^2\rangle^{229} = 0.012(2)\ \mathrm{fm}^2$

sensitivity for $\dot{\alpha}$

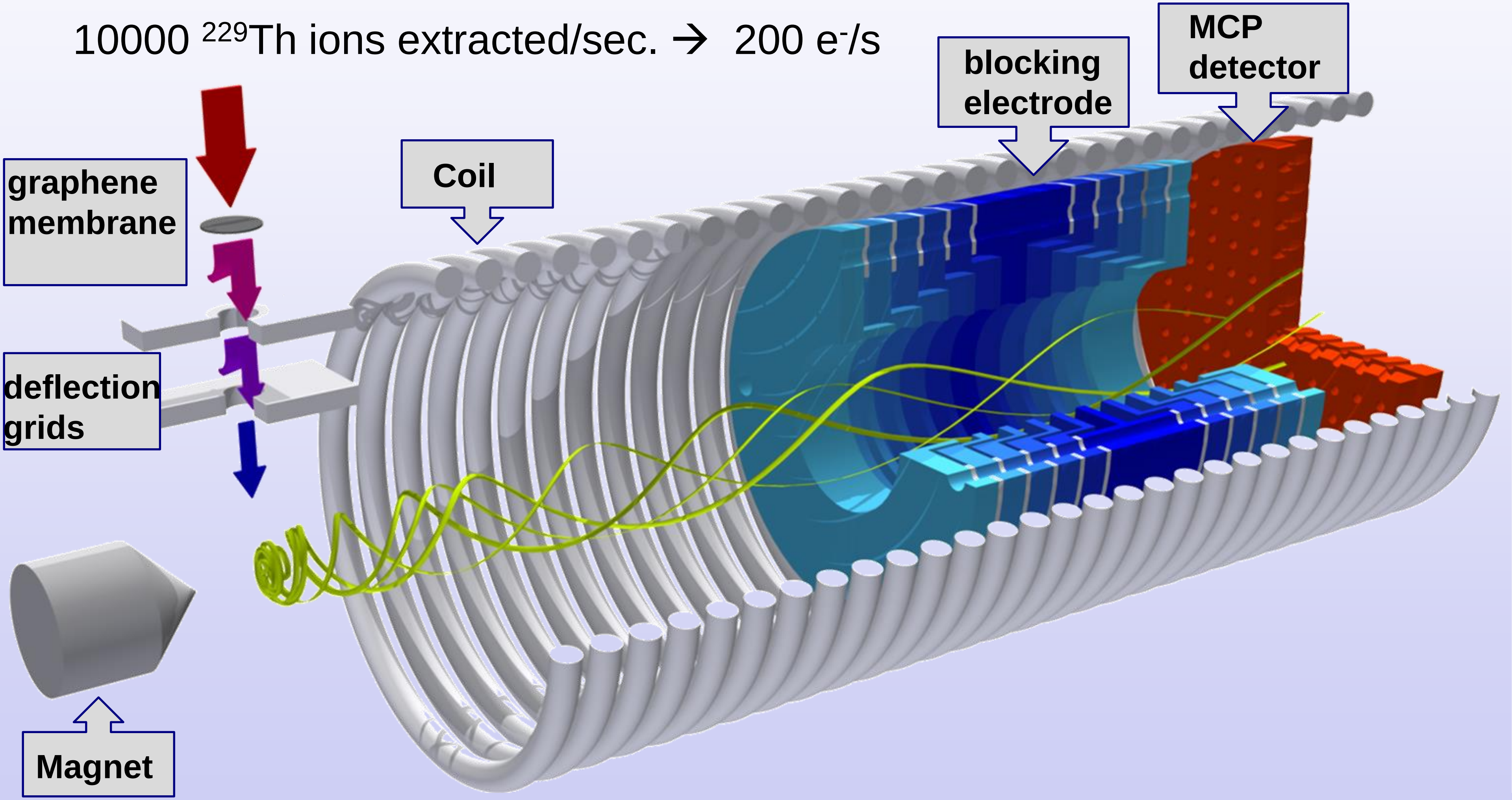
HFS: important for detection (tagging) of isomer excitation

“Holy Grail”: Excitation Energy

presently: 6.31 eV < E(^{229m}Th) < 18.3 eV

- Magnetic-bottle spectrometer: large angular acceptance required

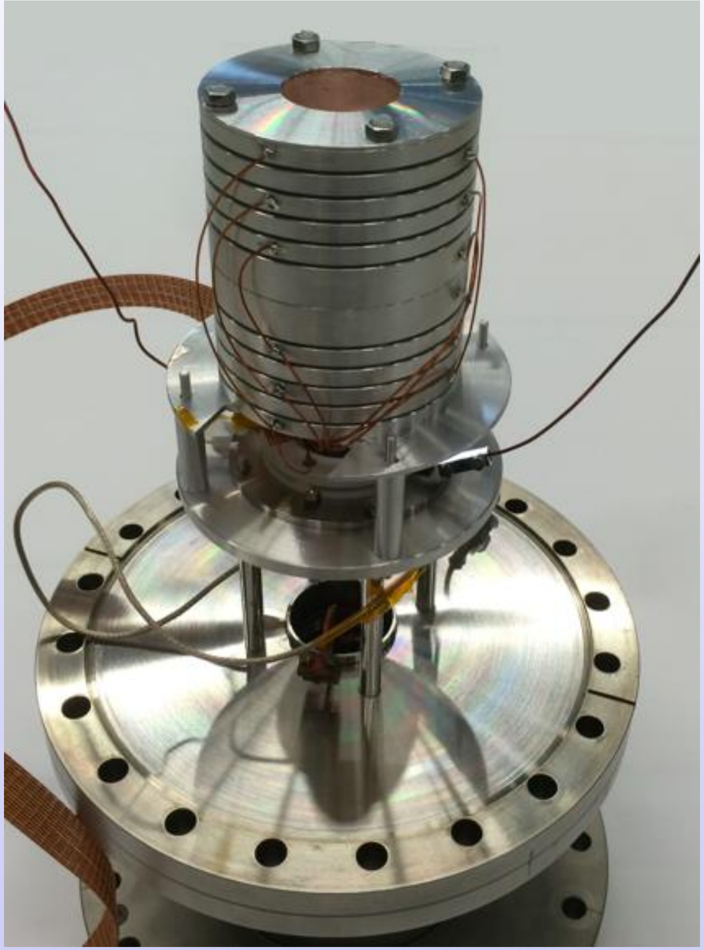
10000 ^{229}Th ions extracted/sec. → 200 e$^-$/s



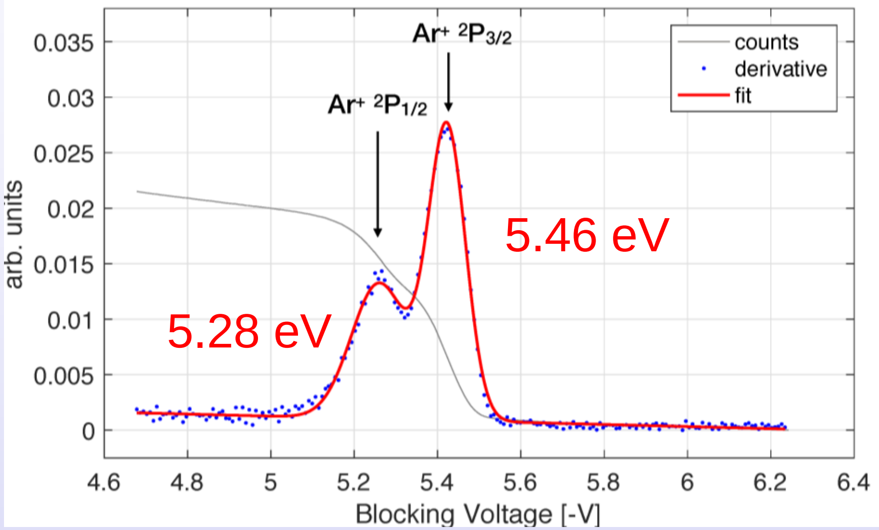
contact-free decay from neutral ^{229m}Th

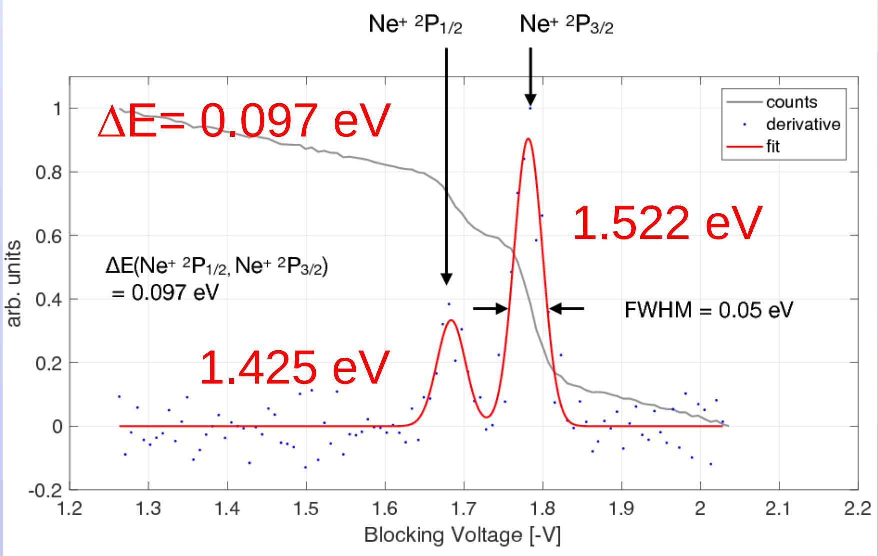
Electron Spectrometer

- Magnetic-bottle spectrometer:



- Calibration: gas discharge excitation:

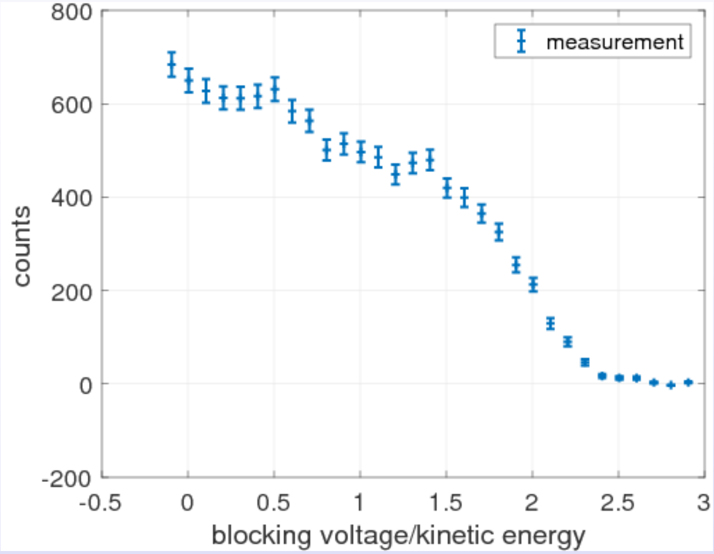


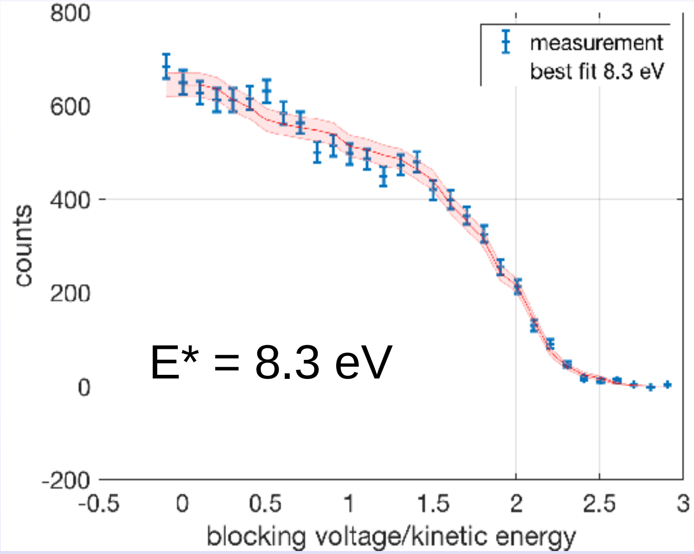


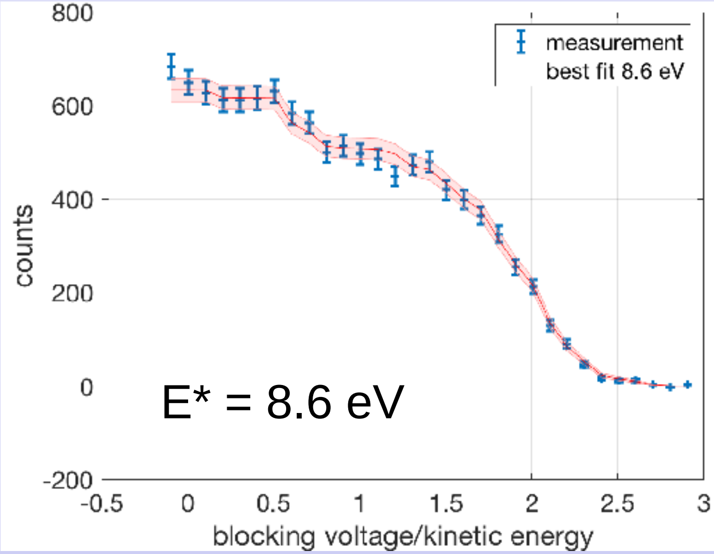
finally expected: ΔE ~ 0.05 - 0.1eV

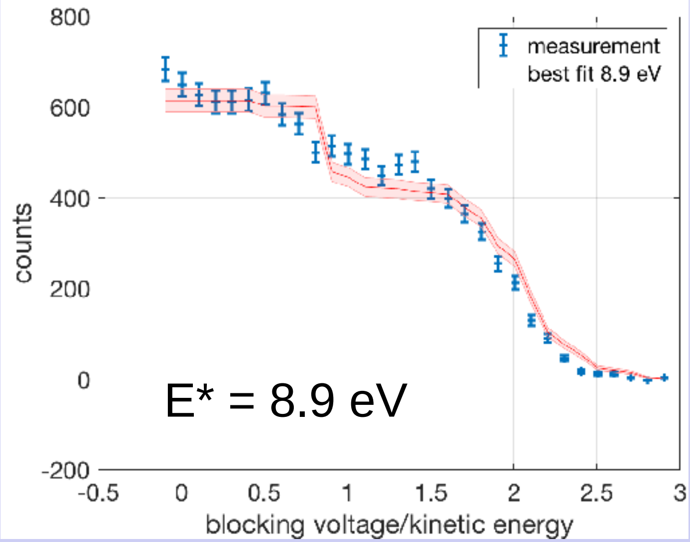
Measurements of E*(^{229m}Th)

- neutralization can end in excited atomic states
- measurement to be compared to theory (P. Bilous, A. Palffy, MPI-K HD)









Summary & Perspectives

- **229-Thorium isomer exists: first direct detection via IC decay channel**
- **constraints of ^{229m}Th properties:** **$6.3\ \text{eV} \leq E^* \leq 18.3\ \text{eV}$** Nature 533 (2016)
  $\tau > 60\ \text{s}$
- **Half-life of neutral ^{229m}Th:** **$t_{1/2} = 7\ \mu\text{s} \rightarrow \alpha_{IC} \sim 10^9$** PRL 118 (2017)
- **Hyperfine structure of ^{229m}Th measured via collinear laser spectroscopy:**
  →nuclear moments, charge radius, (prolate) deformation Nature 556 (2018)
- **isomeric excitation energy:** method: EPJ A53 (2017)
  measurements with (retarding field) magnetic bottle electron spectrometer in progress
- **contrary to general paradigm: laser excitation of ^{229m}Th feasible with existing laser technology** method: PRL 119 (2017)
- **charged ^{229m}Th: needs longer storage time**
  → setup of a cryogenic Paul trap in progress

Thanks to ….

LMU Munich: **L. v.d. Wense, B. Seiferle**, N. Arlt, B. Kotulski, I. Amersdorffer

PTB Braunschweig: **J. Thielking, P. Glowacki**, D.M. Meier M. Okhapkin, E. Peik

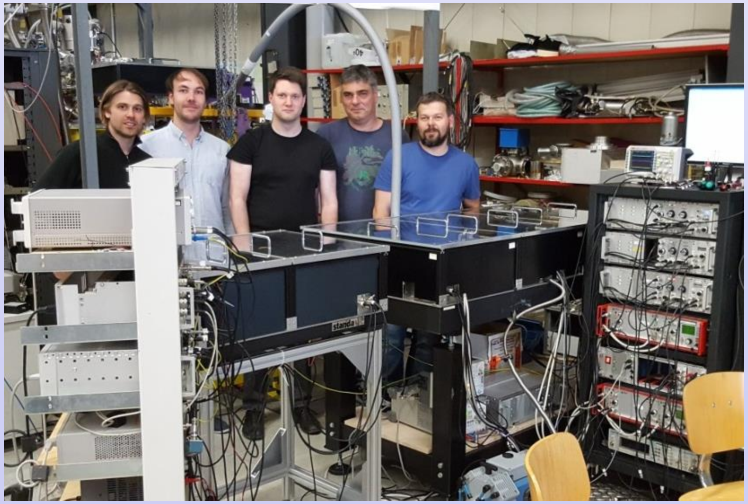
GSI Darmstadt & Helmholtz-Institut Mainz: M. Laatiaoui

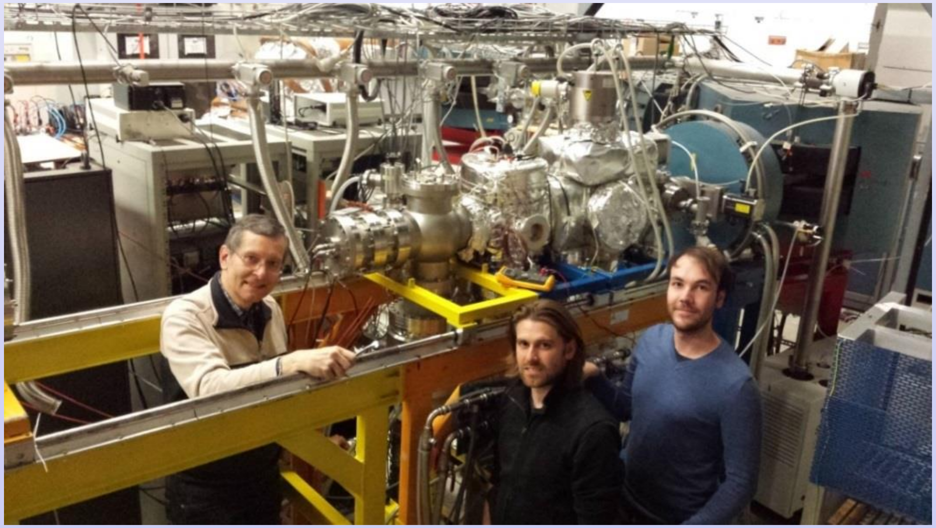
Helmholtz-Institut Mainz & Johannes Gutenberg-Universität Mainz:
C. Mokry, J. Runke, K. Eberhardt, N.G. Trautmann, C.E. Düllmann

TU Wien: T. Schumm, S. Stellmer

MPQ: J. Weitenberg, T. Udem

MPI-HD: A. Palffy, P. Bilous, N. Minkov,
J. Crespo

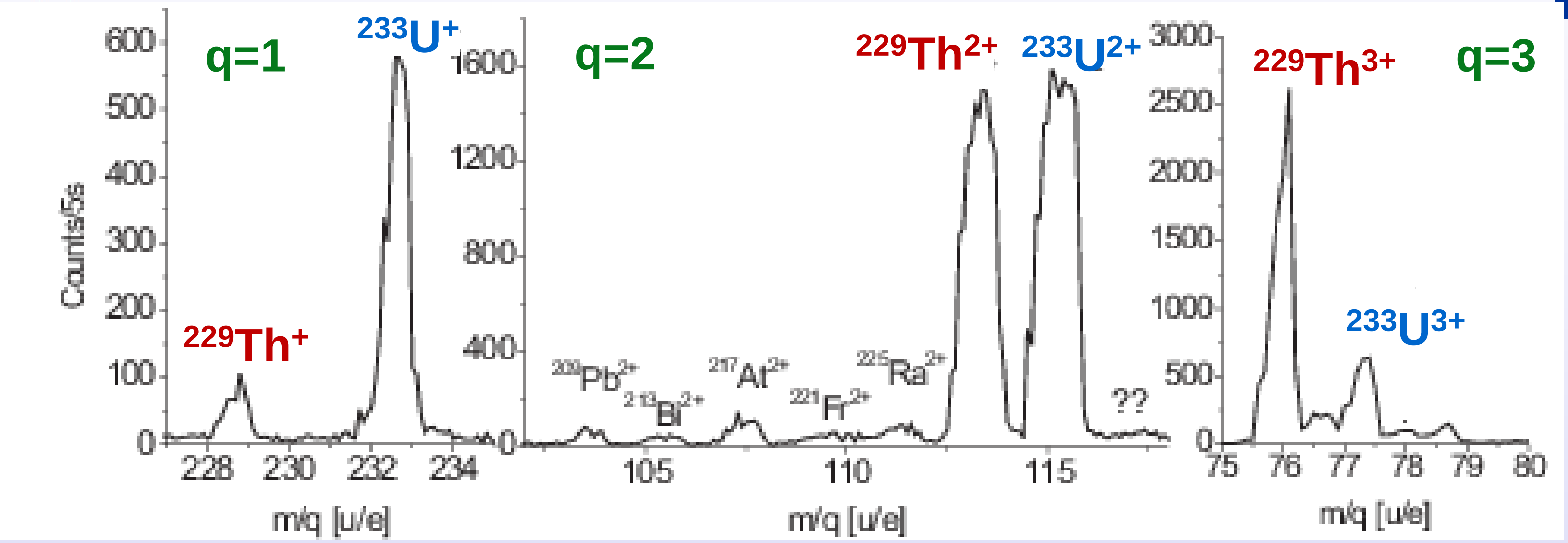




Thank you for your attention !

Ion Extraction from Buffer Gas Cell

mass scan of extracted ion species: efficient $^{229(m)}\mathrm{Th}^{3+}$ extraction



element	1^+ [%]	2^+ [%]	3^+ [%]
Th	0.37(7)	5.5(11)	10(2)
Fr	21.0(42)	16.0(32)	≤ 1.5.10^{-3}
Rn	5.8(12)	9.3(19)	0.053(11)
At	8.6(17)	13.0(26)	0.033(7)
Po	7.3(15)	8.1(16)	≤ 0.0021
Bi	4.3(9)	21.0(42)	0.083(16)
Pb	2.2(4)	11.0(22)	≤ 0.012

element	1^+ [eV]	2^+ [eV]	3^+ [eV]
U	6.1	11.6	19.8
Th	6.3	11.9	18.3
Ra	5.3	10.1	31.0
Fr	4.1	22.4	33.5
Rn	10.7	21.4	29.4
At	9.3	17.9	26.6
Po	8.4	19.3	27.3
Bi	7.3	16.7	25.6

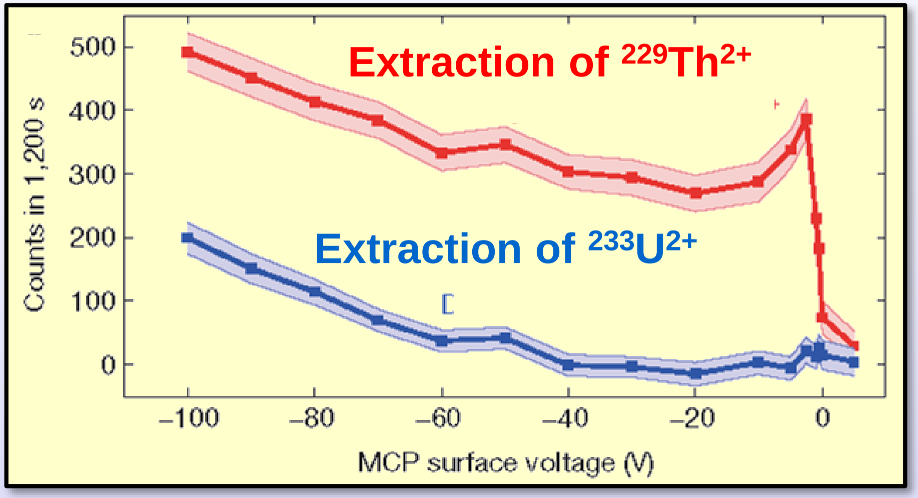
$I(\mathrm{He}^{+}) = 24.6$ eV

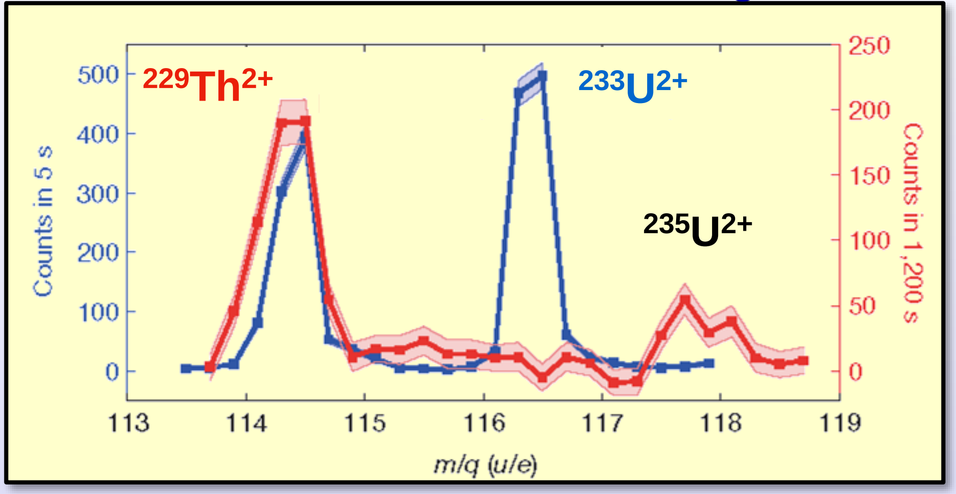
L. v.d. Wense, B. Seiferle, M. Laatiaoui, PT, EPJ A51, 29 (2015)

Verification Measurements

U_{MCP}= -25 V
→ isomeric decay

U_{MCP}= -900 V
→ Ionic signal





- ionic impact signal decreases with lower acceleration towards MCP

- $^{233}U^{2+}$ signal drops to zero

- $^{229}Th^{2+}$ signal remains, cutoff at E_{kin}=0

- for strong acceleration towards MCP: comparable signals for $^{229}Th^{2+}$, $^{233}U^{2+}$

- for 'soft landing' $^{233}U^{2+}$ signal vanishes

- $^{229}Th^{2+}$ signal remains

all potential background contributions could be excluded, mostly by multiple ways