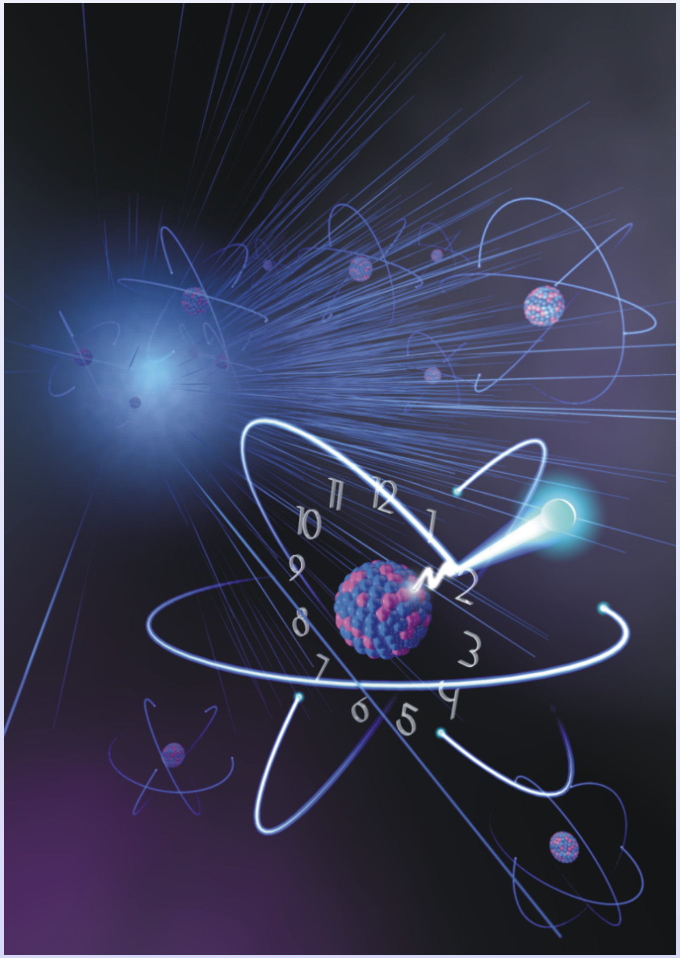
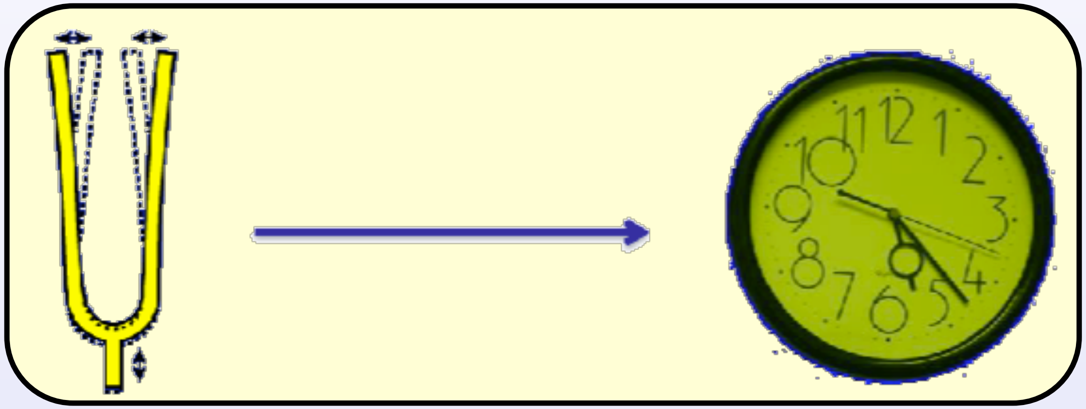
Direct Detection of the Elusive ^{229}Thorium Isomer: Milestone towards a Nuclear Clock

P.G. Thirolf, LMU München

- Clock basics
- ^{229m}Th properties and prospects
- Experimental approach & setup
- Isomer decay study results
- Verification studies
- Summary & Perspectives



Functional Principle of Clocks



periodic event (oscillation)

counting device

examples:

- rotation of earth, moon phases
- pendulum, spring
- quartz crystal
- atomic transitions
- nuclear transitions ?

examples:

- sun dial
- mechanical clocks
- ‘digital’ clocks
- microwave, laser

rising frequency

Clock Accuracies

Mechanical clocks

accuracy: 1s /day

world record (NIST): 1s / year

Quartz clocks

accuracy: 1s / month

world record: 10^{-12}

Atomic clocks

accuracy: 10^{-14}

world record: 10^{-18}

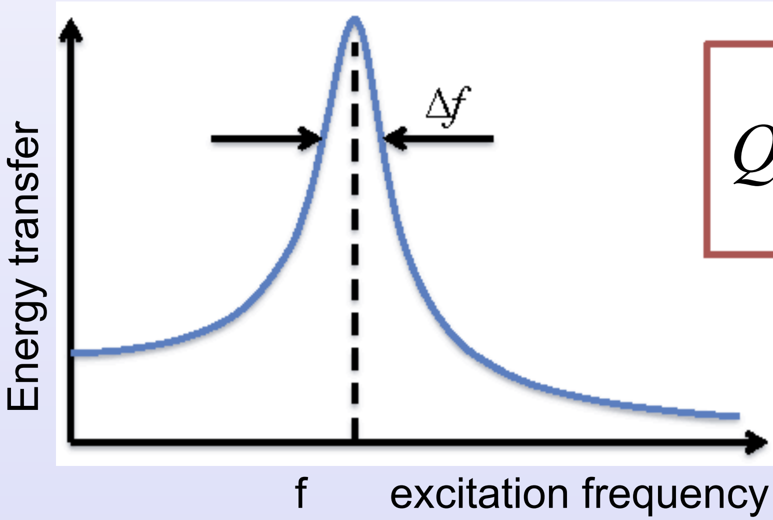
nuclear clocks ??

^{229m}Th

Clock properties

- continuous operation:
  - feedback on resonance
  - optimum energy transfer at specific excitation frequency f

- Quality factor Q:



$$Q = \frac{f}{\Delta f}$$

best clock:

- highest oscillation frequency (f large)
- as precise as possible (Δf small)

microwave atomic clocks: $Q \sim 10^{10}$

'optical' atomic clocks: $Q \sim 10^{14}$

nuclear clock: $Q > 10^{15}$??

What makes ^{229m}Th so unique ?

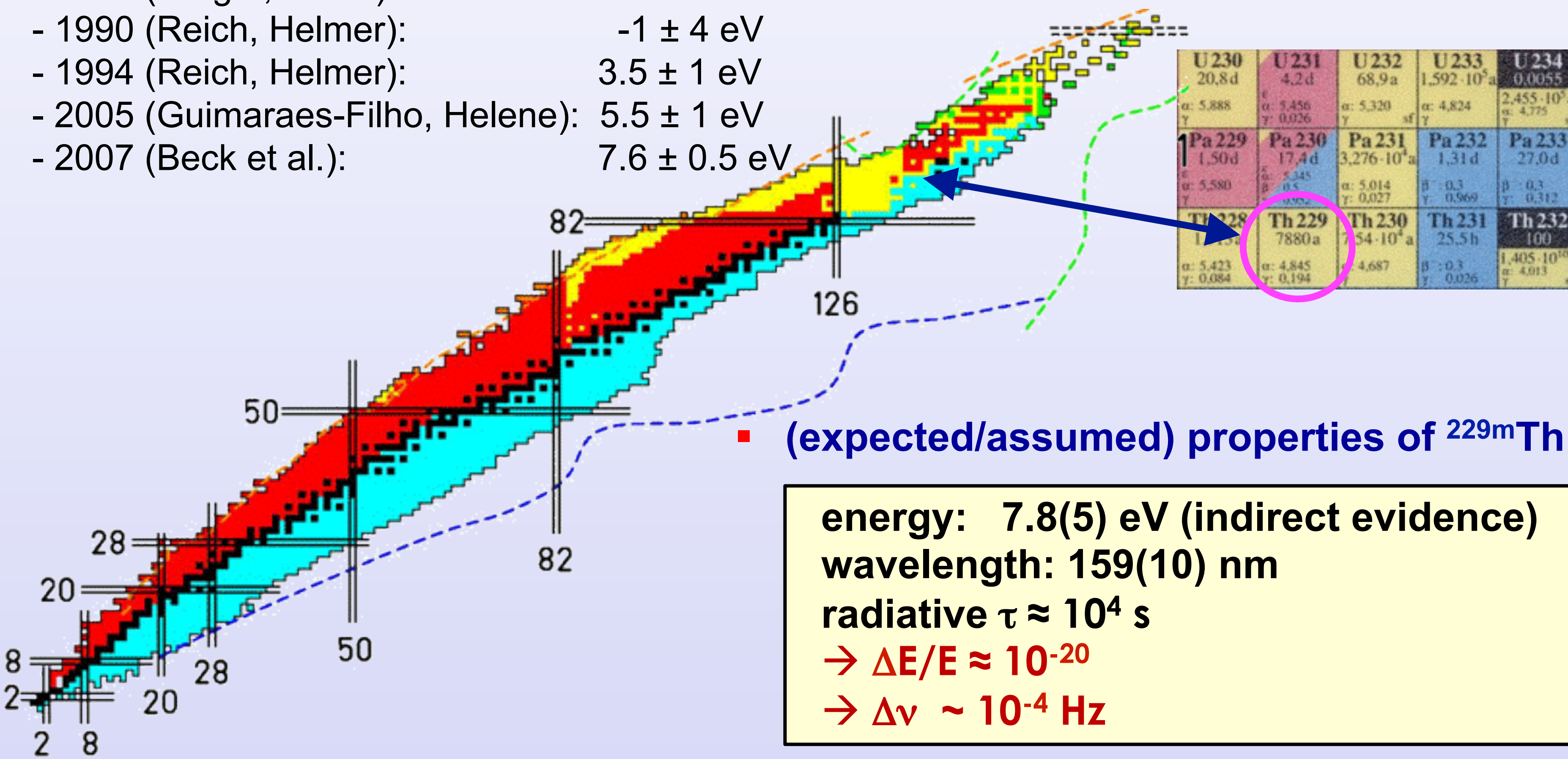
nuClock

MLL

- **since 40 years nuclear physicists seemed to chase a phantom:**

presumed low-lying isomer in ^{229}Th:

- 1976 (Kroger, Reich): < 100 eV
- 1990 (Reich, Helmer): -1 ± 4 eV
- 1994 (Reich, Helmer): 3.5 ± 1 eV
- 2005 (Guimaraes-Filho, Helene): 5.5 ± 1 eV
- 2007 (Beck et al.): 7.6 ± 0.5 eV



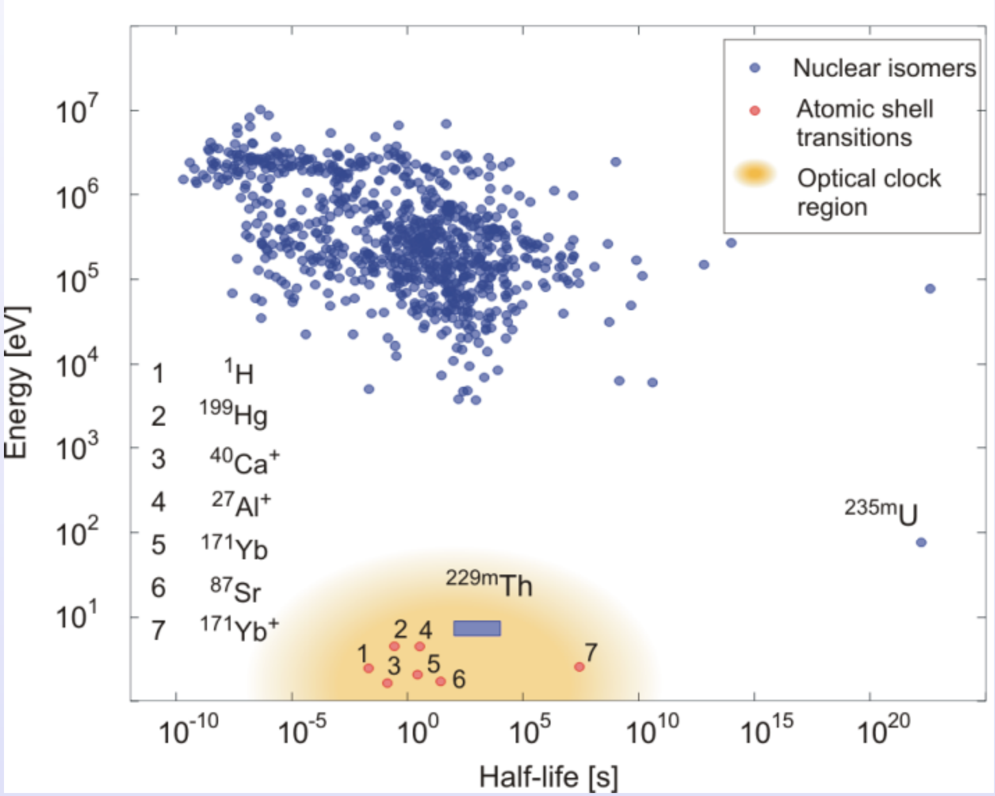
- **(expected/assumed) properties of ^{229m}Th:**

energy: 7.8(5) eV (indirect evidence)
wavelength: 159(10) nm
radiative $\tau \approx 10^4$ s
→ $\Delta E/E \approx 10^{-20}$
→ $\Delta\nu \sim 10^{-4}$ Hz

B.R. Beck et al., LLNL-PROC-415170 (2009)

^{229m}Th: Nuclear Clock ?

→ **lowest excitation energy of all ca. 176000 presently known excited nuclear states**



- insensitivity of ^{229m}Th nuclear transition frequency to external perturbations
  - → small nuclear moments
  - → largely immune to systematic frequency shifts:
  - → ultra-precise nuclear frequency standard ?

^{229m}Th and time dependence of fundamental constants

Theories unifying gravity with other interactions suggest temporal and spatial variations of the fundamental "constants" in an expanding universe

Uzan, Rev. Mod. Phys. 75, 403 (2003)

- near degeneracy of ^{229,229m}Th results from cancellation between large contributions from Coulomb energy (~10^9 eV)
- dependencies on fundamental constants
  → any variation would be enhanced in the transition frequency

temporal variation in transition energy may provide enhanced sensitivity by (10^2 – 10^5) for fine structure constant $\dot{\alpha}/\alpha$ and strong interaction parameter (m_q/Λ_{QCD})

- Flambaum, PRL 97 (2006)
- He, Ren, NP A 806 (2008)
- Litvinova, Feldmeier et al., PRC 79 (2009)
- Flambaum, Wiringa, PRC 79 (2009)
- Rellergert et al, PRL 104 (2010)
- Hayes, Friar, PL B 650 (2007)
- Berengut, Flambaum et al., PRL 102 (2009)
- Flambaum et al., Europhys. Lett. 85 (2009)
- Flambaum, Porsev, PRA 80 (2010)

▪ current (theor.) limit of potential time variation:

$$\dot{\alpha}/\alpha = (-1.6 \pm 2.3)\cdot 10^{-17}\ \mathbf{yr}^{-1}$$

Fujii et al., NP B573, 377 (2000)

Sensitivity to variations of α

- sensitivity of ^{229m}Th transition frequency ω to temporal variation of $\alpha = e^2/hc$:

$$\delta\omega = \Delta V_C \frac{\delta\alpha}{\alpha}, \qquad \frac{\delta\omega}{\omega} = K \frac{\delta\alpha}{\alpha}$$

ΔV_C: difference in Coulomb energy between ^{229}Th, ^{229m}Th (proton polarization by modified neutron distribution)

enhancement factor: $$K = \frac{\Delta V_C}{\omega}$$

- example: for ΔV_C = 100 keV, $\delta\alpha/\alpha = 10^{-16}$
  → $\delta\omega = 10^{-11}$ eV (= $2.4 \cdot 10^3$ Hz)
  → 10^4 larger than limits on shifts in atomic transitions

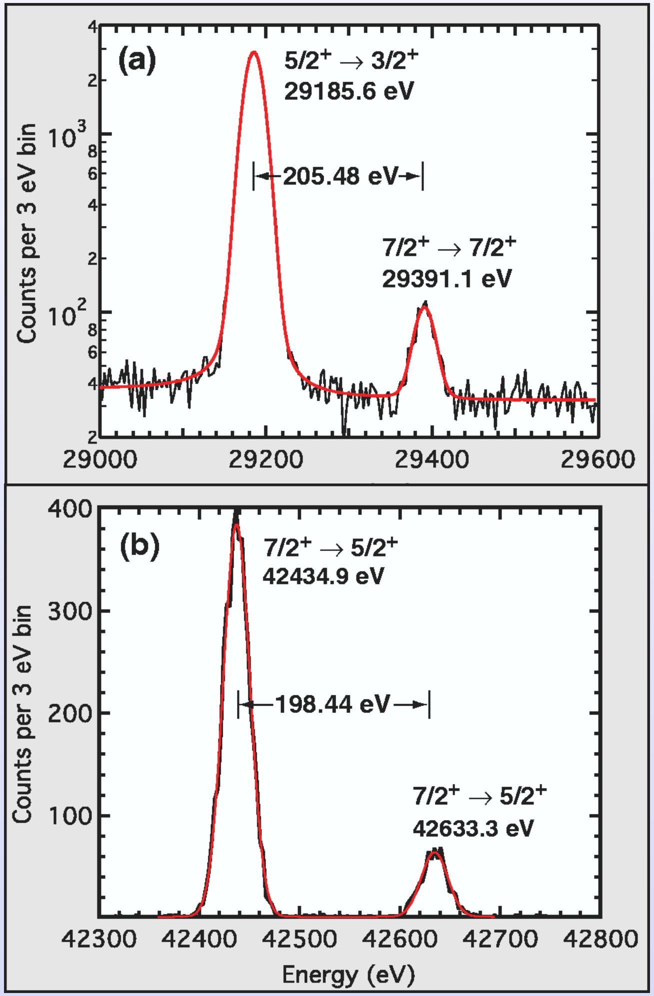
- problem: - Coulomb energy cannot be directly measured
  - V_C needs to be calculated in nuclear models
  - insufficient accuracy in present nuclear models (e.g. Hartree-Fock, Hartree-Fock-Bogoliubov: -300 keV < ΔV_C (^{229}Th) < 400 keV → $-4\cdot10^4 < K < +4\cdot10^4$)

system	K	λ [nm]
Sr	0.06	699
Yb^+ E2	0.91	436
Yb^+ E3	-6	467
Hg+	-2.9	281.5
Al^+	0.01	267
Ir^{17+} T1	-20.6	ca. 267
Ir^{17+} T2	32.2	ca. 470
Cf^{16+*} T1	75	ca. 520
Cf^{16+*} T2	-46	ca. 653
Th^* (nuclear)	8000	ca. 160

Schmidt & Crespo, Phys. Jour. Oct. 2016

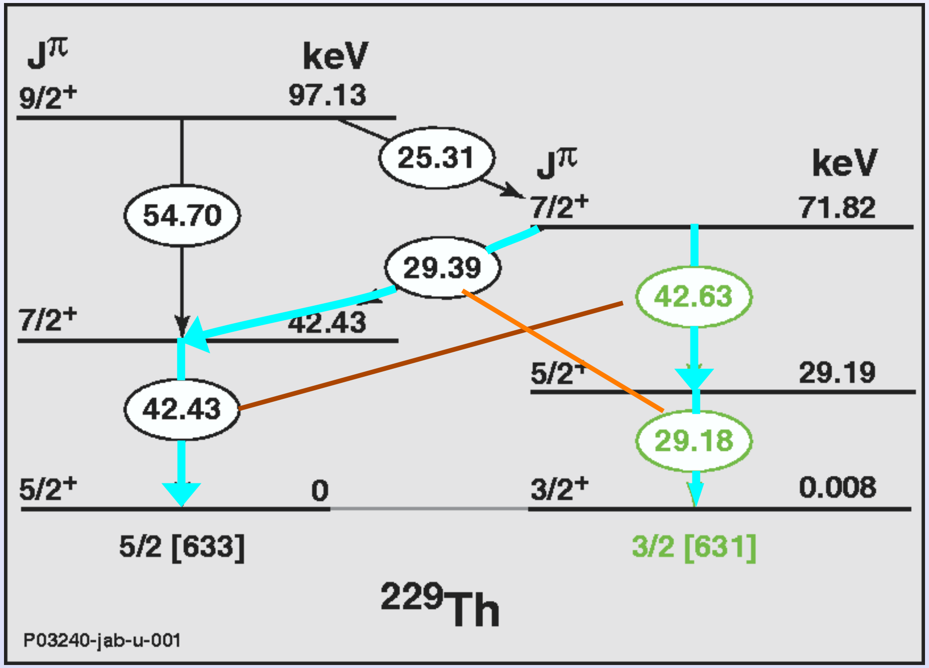
Indirect Evidence for ^{229m}Th

- data from X-ray micro-calorimeter



$E^* (^{229m}Th) = \Delta E_{29} - \Delta E_{42}$

$E^* (^{229m}Th) = (7.8 \pm 0.5)$ eV



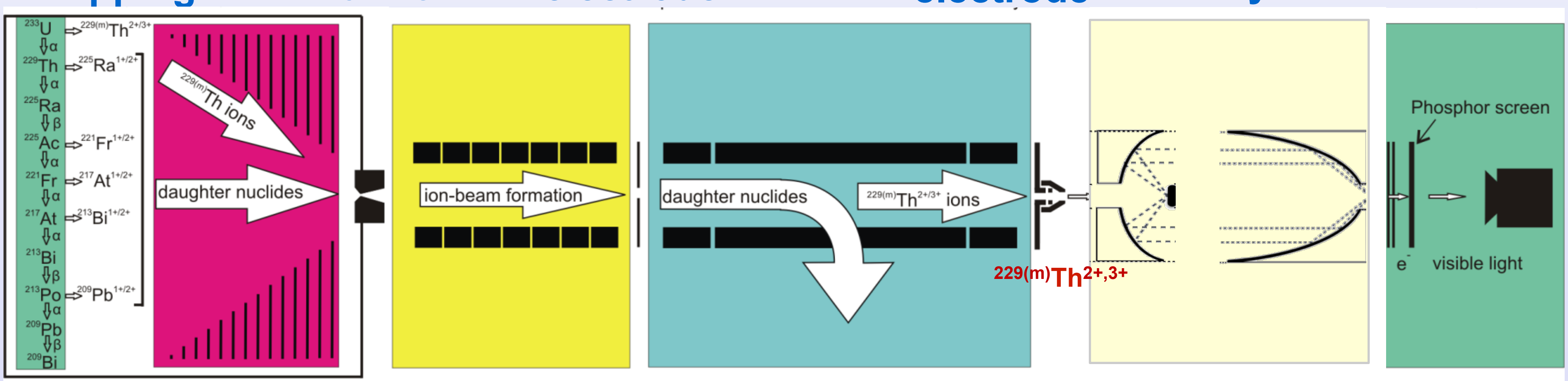
B.R. Beck et al., PRL **98** (2007) 142501
LLNL-PROC-415170 (2009)

from energy difference of high-energy transitions → low isomer excitation energy

Experimental Approach @ LMU

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
- detect the subsequently occurring isomeric decay

Buffer gas stopping cell | **Laval nozzle** | **aperture electrode** | **triodic extraction electrode** | **detection system**



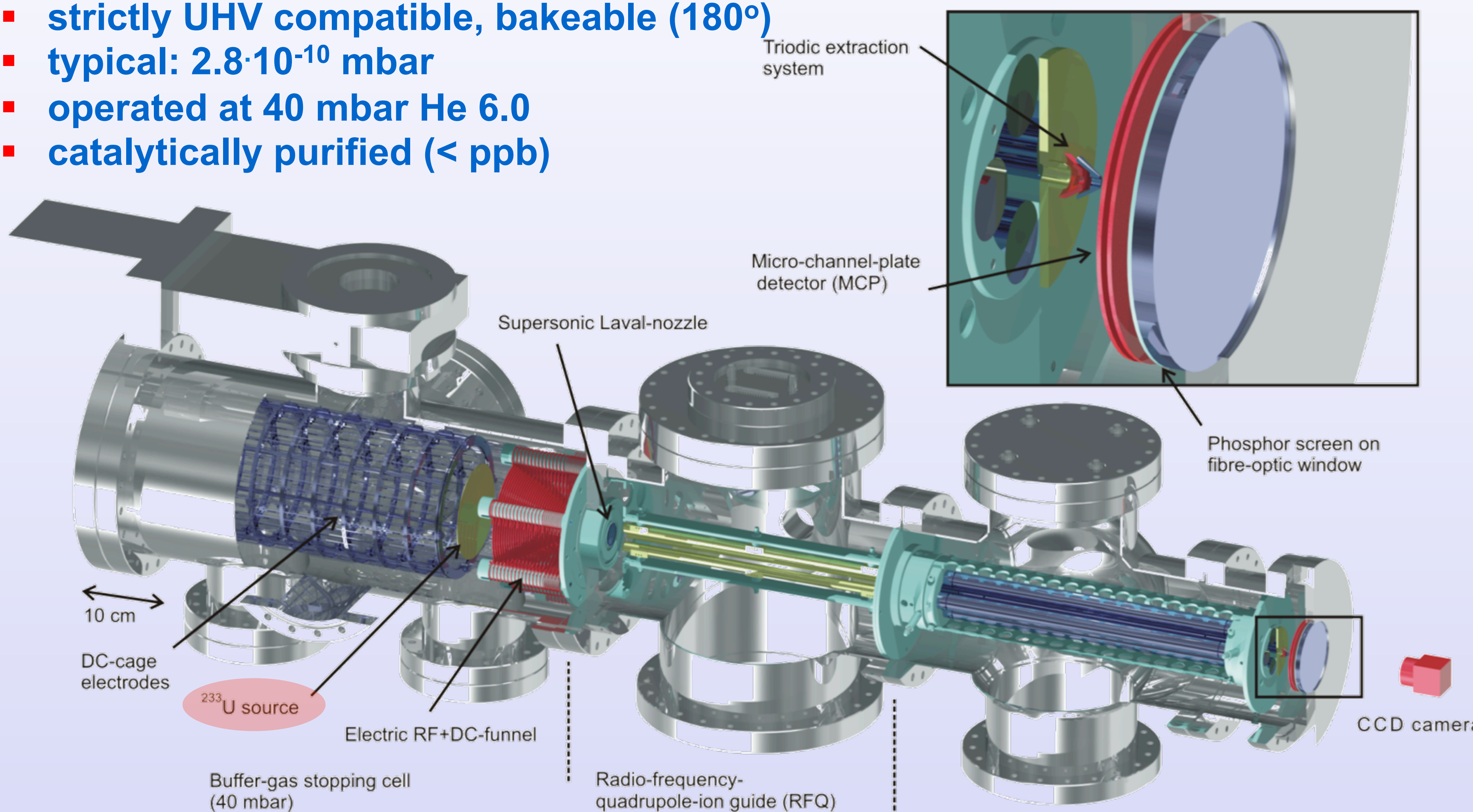
^{233}U source | **RF + DC funnel** | **RF quadrupole ion guide** | **Quadrupole mass separator** | **UV detection** | **MCP** | **CCD**

40 mbar (He) | 10^{-2} mbar | 10^{-6} mbar

→ 4 vacuum chambers , 3 differential pump sections, overall length ~ 2 m

Experimental Setup

- strictly UHV compatible, bakeable (180°)
- typical: $2.8 \cdot 10^{-10}$ mbar
- operated at 40 mbar He 6.0
- catalytically purified (< ppb)



m/Δm~ 150

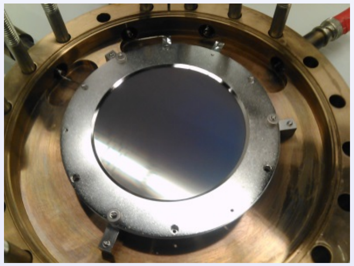
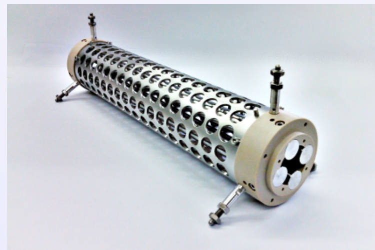
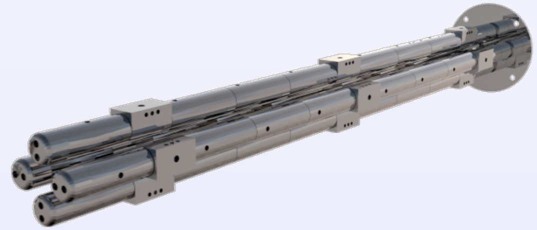
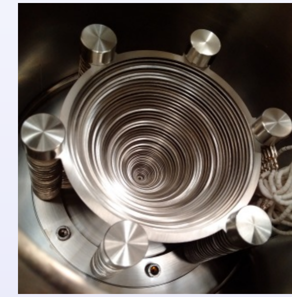
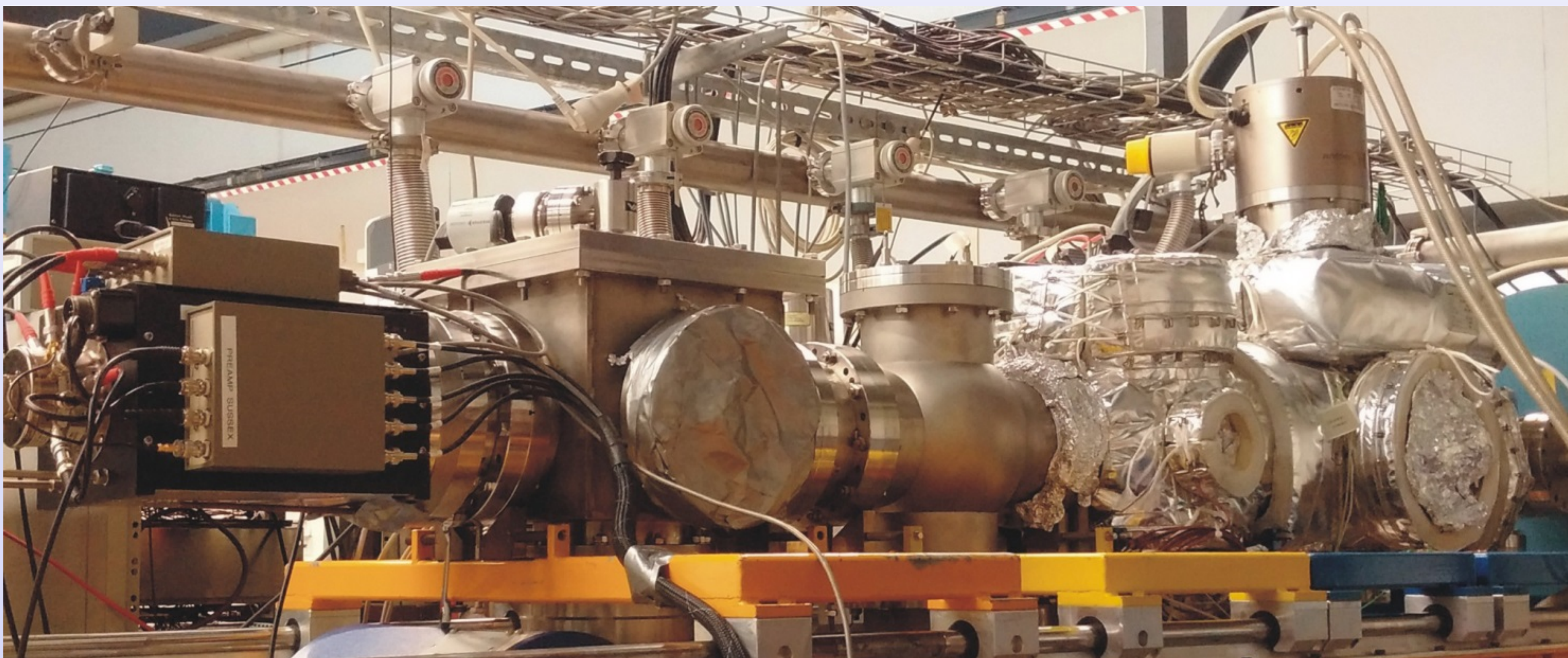
220 V_{pp}, 850 kHz
4 V/cm

200 V_{pp}, 880 kHz
0.1 V/cm

Th^{2+}: 925 kHz, 901.5 V_{pp}, 75.23 V
Th^{3+}: 925 kHz, 600.5 V_{pp}, 50.15 V

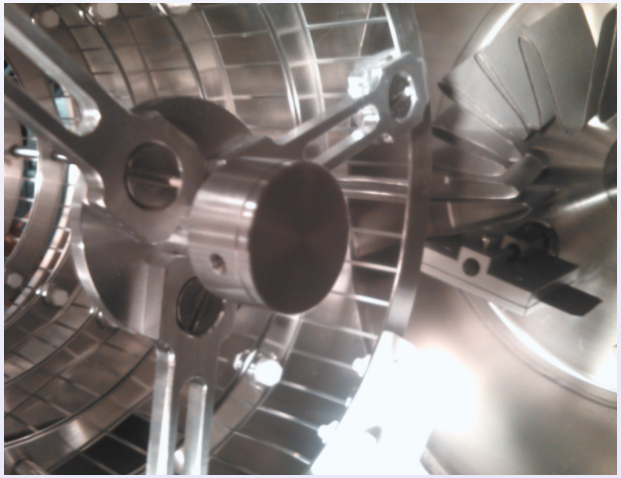
Experimental Setup

located at Maier-Leibnitz Laboratory, Garching:

^{233}U ion sources

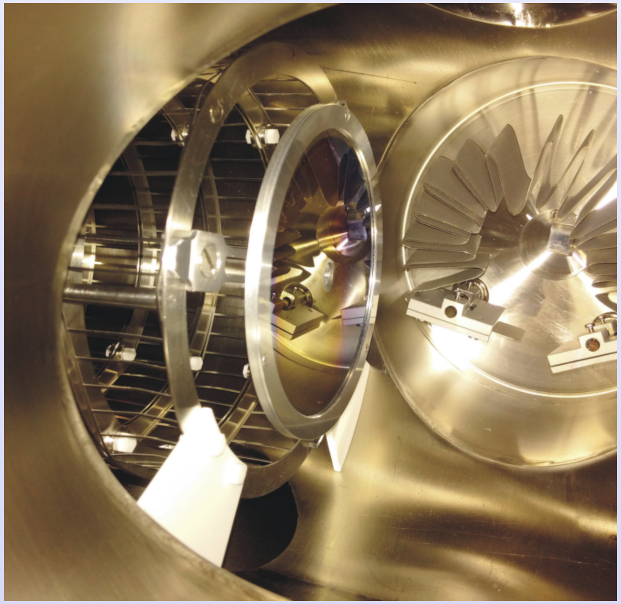
population of ^{229m}Th via 2% α decay branch of ^{233}U



2 different ^{233}U sources were used:

source 1:

- 200 kBq $^{233}UF_4$ evaporated onto 20 mm ø steel plate
- no radio-chemical purification before evaporation
- ^{232}U contamination ≤ 10^{-6}
- eff. $^{229(m)}$Th recoil activity: ca. 4000/s



source 2: (IRC Uni Mainz)

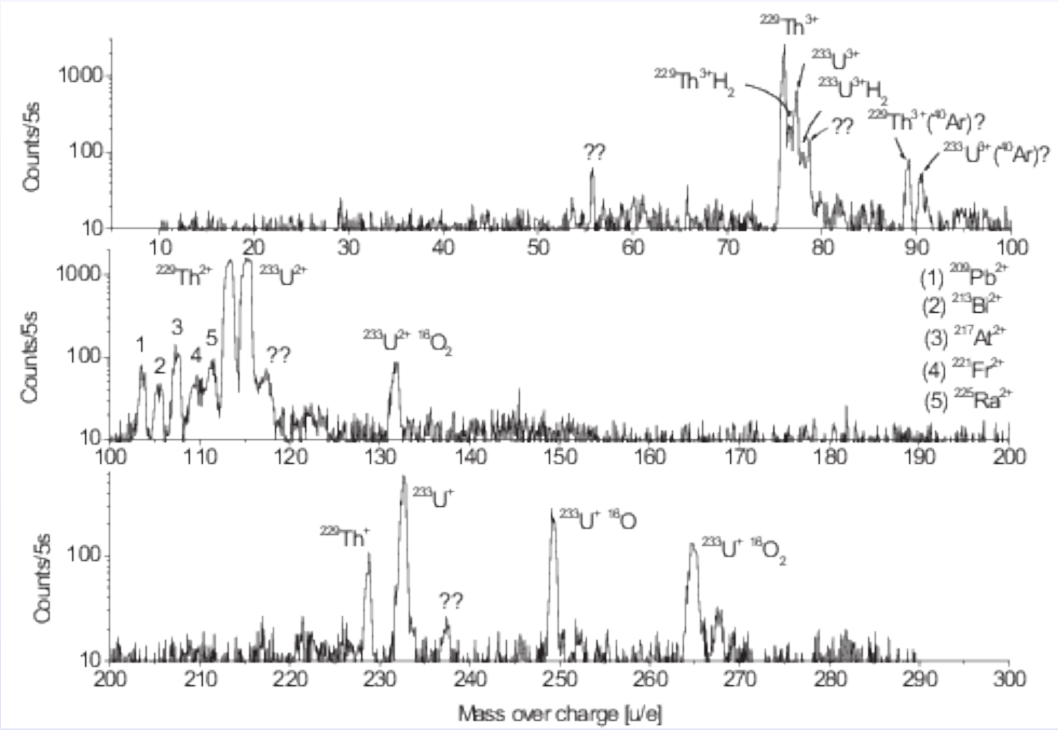
- 290 kBq electro-deposited onto 90 mm ø wafer
- radio-chemical purification before evaporation
- ^{232}U contamination ≤ 10^{-6}
- eff. $^{229(m)}$Th recoil activity: ca. 55000/s

Ion Extraction from Buffer Gas Cell

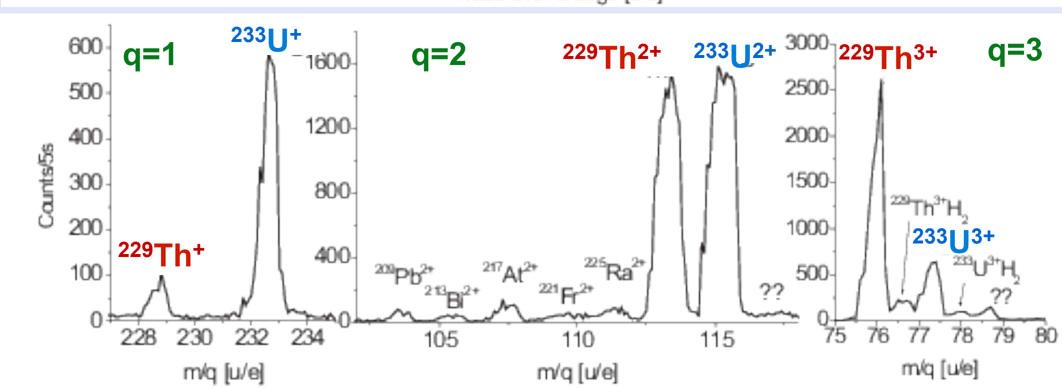
nuclock

MLL

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



element	1^+ [%]	2^+ [%]	3^+ [%]
Th	0.37(7)	5.5(11)	10(2)
Fr	21.0(42)	16.0(32)	≤ $1.5 \cdot 10^{-3}$
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

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

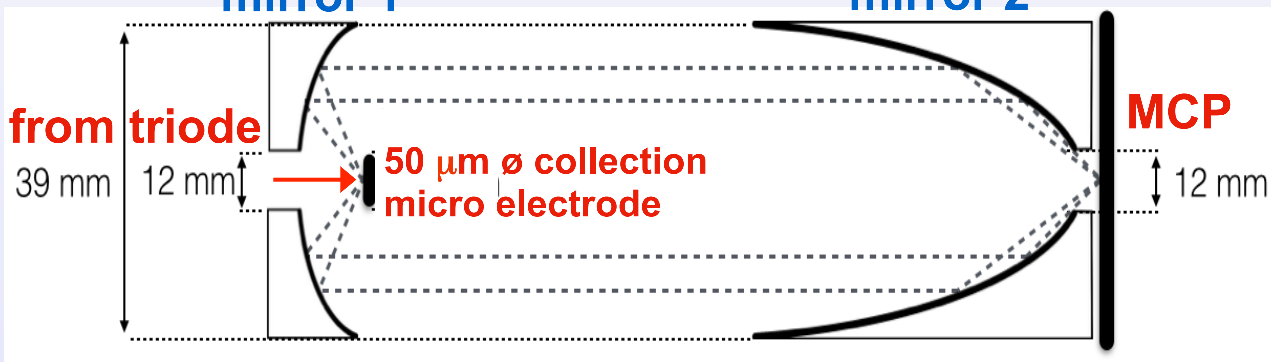
$I(He^+)$ = 24.6 eV

Isomer Decay: Search for VUV Fluorescence

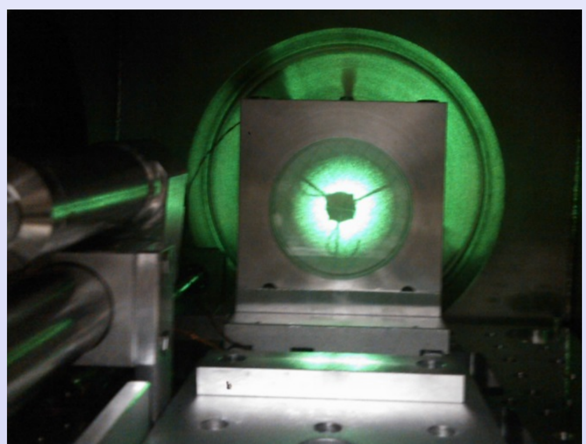
UV optical setup designed, built and commissioned (aligned):

→ 2 annular, parabolic mirrors (MgF_2 coated, from Fraunhofer IOF, Jena)

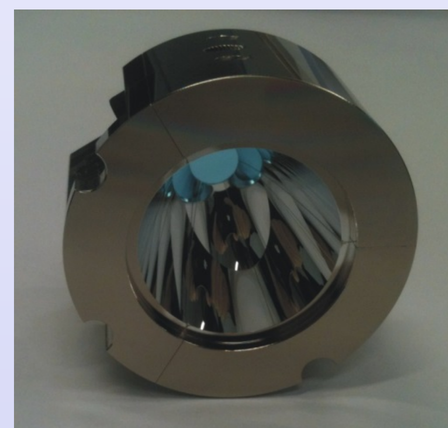
mirror 1 mirror 2



mirror 1:



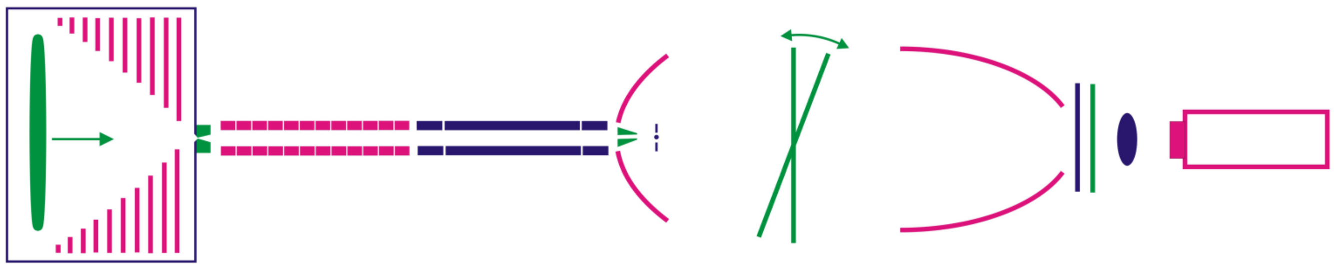
mirror 2:



B. Seiferle, L. v.d. Wense, M. Laatiaoui, PT, EPJ D70, 58 (2016)

VUV Optics Setup

Step 1: Stopping & Extraction | Step 2: Mass Filtering | Step 3: Collection | Step 4: VUV Optics & | Step 5: Detection



efficiency:

Isomer:	Extraction:	Charge:	Mass Filter:	Collection:	Mirror acceptance:	Mirror reflectivity:	Focusing:	Detection:	Total efficiency:
2%	16%	65%	80%	40%	42%	49%	76%	10%	$1.3 \cdot 10^{-5}$

L. v.d. Wense, PT, D. Kalb, M. Laatiaoui, Journal of Instrumentation 8, P03005 (2013)

- **signal-to-noise ratio:**

- **VUV fluorescence ?**

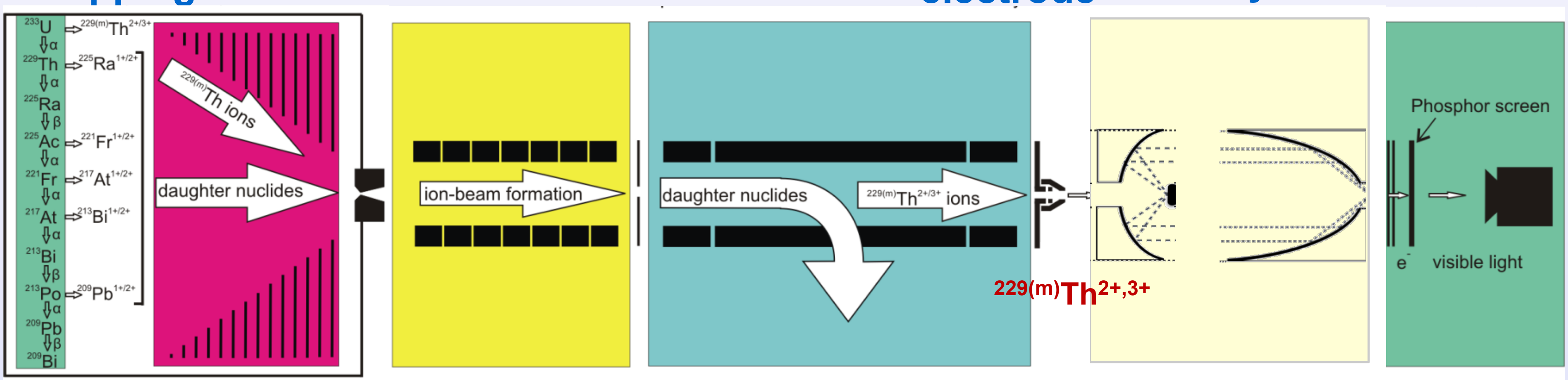
ion collection on micro-electrode: no fluorescence signal was observed

→ **radiative decay branch too weak ?**
→ **search for dominant IC decay branch**
→ **measure conversion electrons**

^{233}U source 2	
eff. ^{229}Th recoil activity	55000 / s
detectable VUV photons	0.72 / s
count rate/area	81.82 / mm^{2}s
dark count rate	0.01 / mm^{2}s
signal/noise ratio	8182:1

Isomer Detection Process

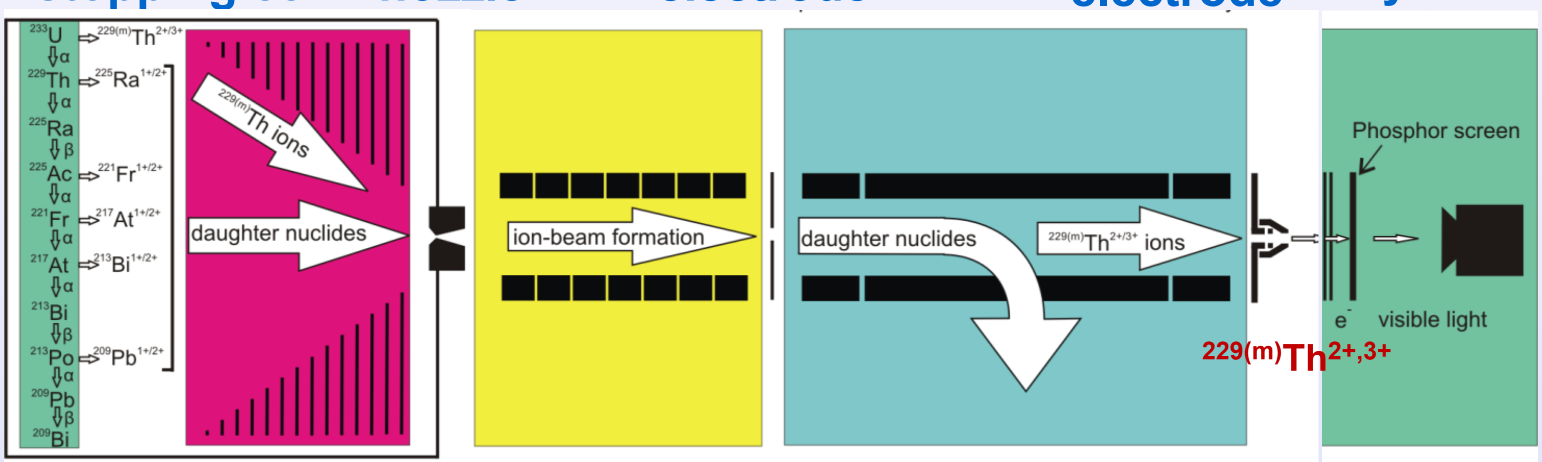
Buffer gas stopping cell | Laval nozzle | aperture electrode | triodic extraction electrode | detection system



^{233}U source | RF + DC funnel | RF quadrupole ion guide | Quadrupole mass separator | UV detection | MCP | CCD

Isomer Detection Process

Buffer gas stopping cell | Laval nozzle | aperture electrode | triodic extraction electrode | detection system

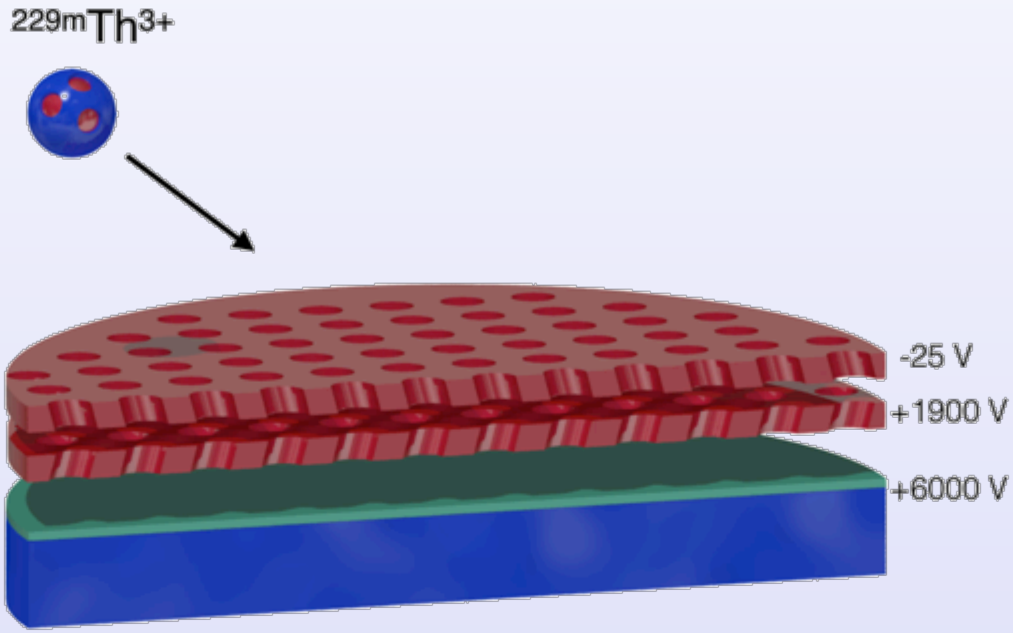


^{233}U source | RF + DC funnel | RF quadrupole ion guide | Quadrupole mass separator | MCP | CCD

→ accumulate $^{229(m)}$Th ions directly onto MCP surface

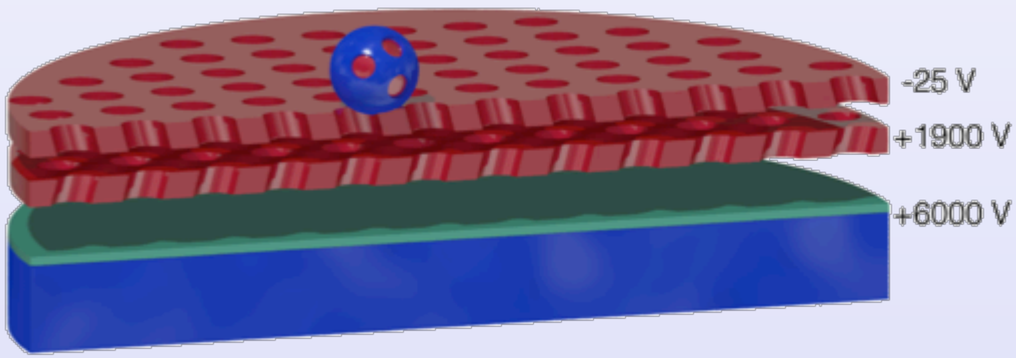
Isomer Detection Process

extracted $^{229(m)}Th^{3+}$ ions: **impinging directly onto MCP surface behind triode exit**



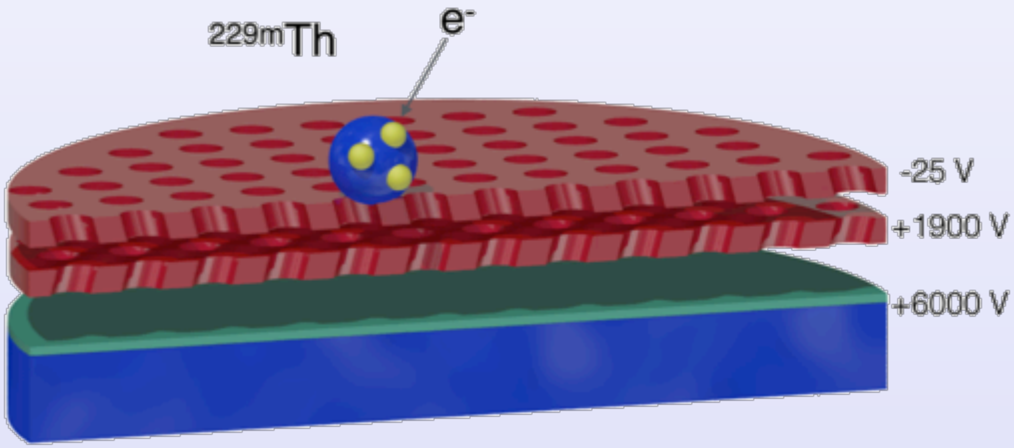
Isomer Detection Process

extracted $^{229(m)}Th^{3+}$ ions: **'soft landing' on MCP surface**



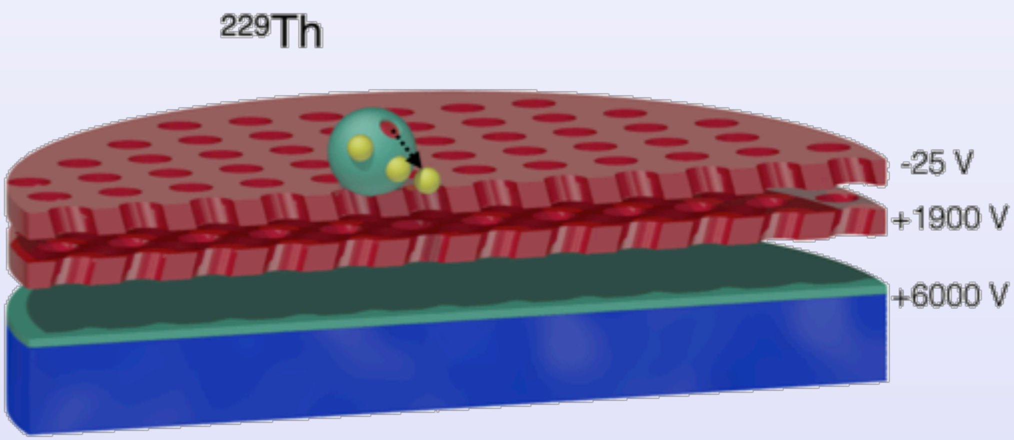
Isomer Detection Process

electron capture on MCP surface: **neutralization of Th ions**



Isomer Detection Process

isomer decay by Internal Conversion: electron emission

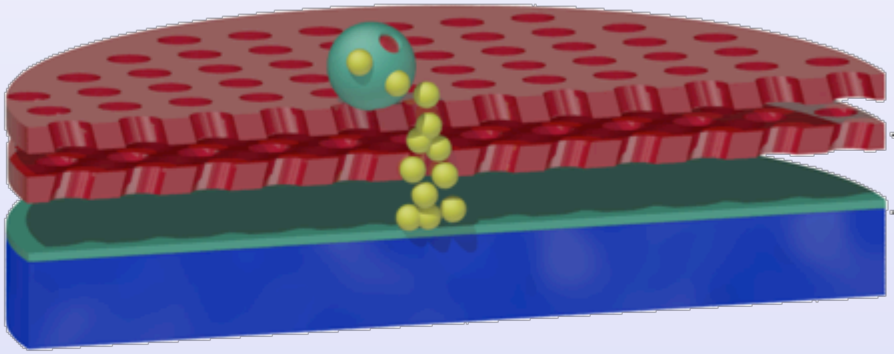


- internal conversion (IC) energetically allowed for neutral thorium:

  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 ions: IC is energetically forbidden, radiative decay branch may dominate

Isomer Detection Process

amplification of IC electron: **electron cascade generated, accelerated towards phosphor screen**



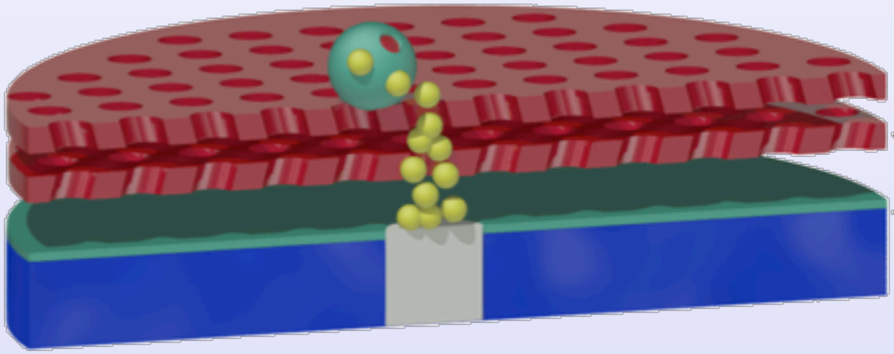
- internal conversion (IC) energetically allowed for neutral thorium:

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

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

Isomer Detection Process

electrons hit phosphor screen: **visible light imaged by CCD camera**



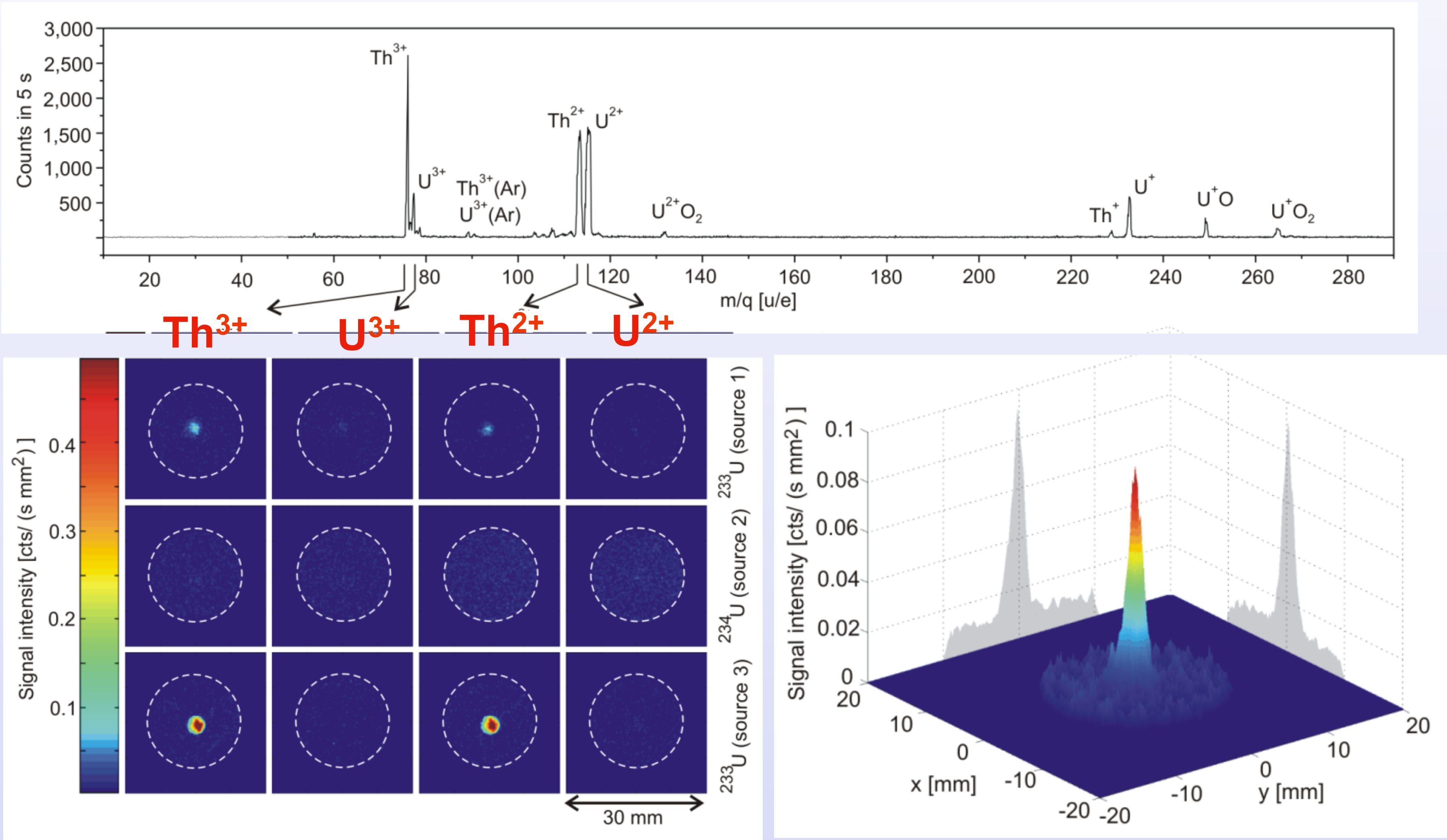
- internal conversion (IC) energetically allowed for neutral thorium:

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

- isomer lifetime expected to be reduced by ca. 10^9 (from ~10^4 s → ~ 10 μs)
- Th 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, 47-53 (2016)



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

Verification Measurements

LMU LUDWIG-MAXIMILIANS-UNIVERSITÄT MÜNCHEN

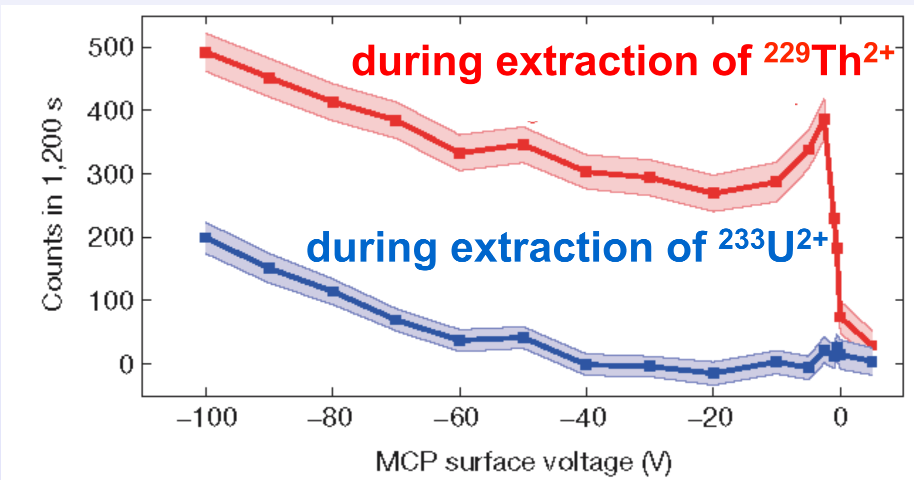
MLL

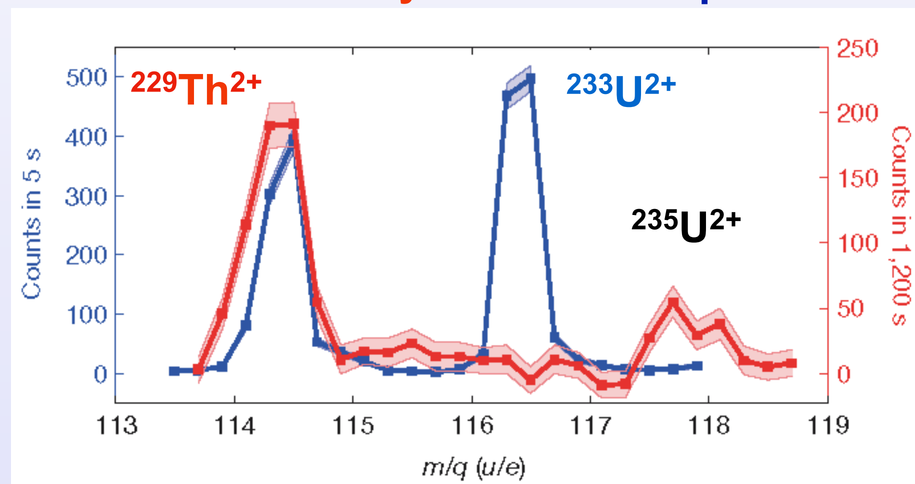
nuClock

Background-corrected isomeric decay signals:

U_{MCP}= -25 V → isomeric decay

U_{MCP}= -900 V → ionic impact





- ionic impact signal decreases with U_{MCP}
- $^{233}U^{2+}$ signal drops to zero
- $^{229}Th^{2+}$ signal remains, cutoff at E_{kin}=0

  (rise: IC electrons back-attracted to MCP surface by repelling triode)

- comparable mass peak amplitudes for $^{229}Th^{2+}$, $^{233}U^{2+}$ ion impact signals
- for U_{MCP}= -25 V $^{233}U^{2+}$ signal vanishes
- $^{229}Th^{2+}$ signal remains

Potential Background Contributions

A) **Background due to kinetic energy or charge state of impinging ions**

B) **Background signals from setup components**
(source, gas stopping cell, extraction system, QMS , MCP system)

C) **Signals from thorium atomic shell**
(e.g. long-lived excited states, chemical reactions on MCP surface)

D) **Signals from short-lived daughter nuclides or non- ^{229}Th isomers**

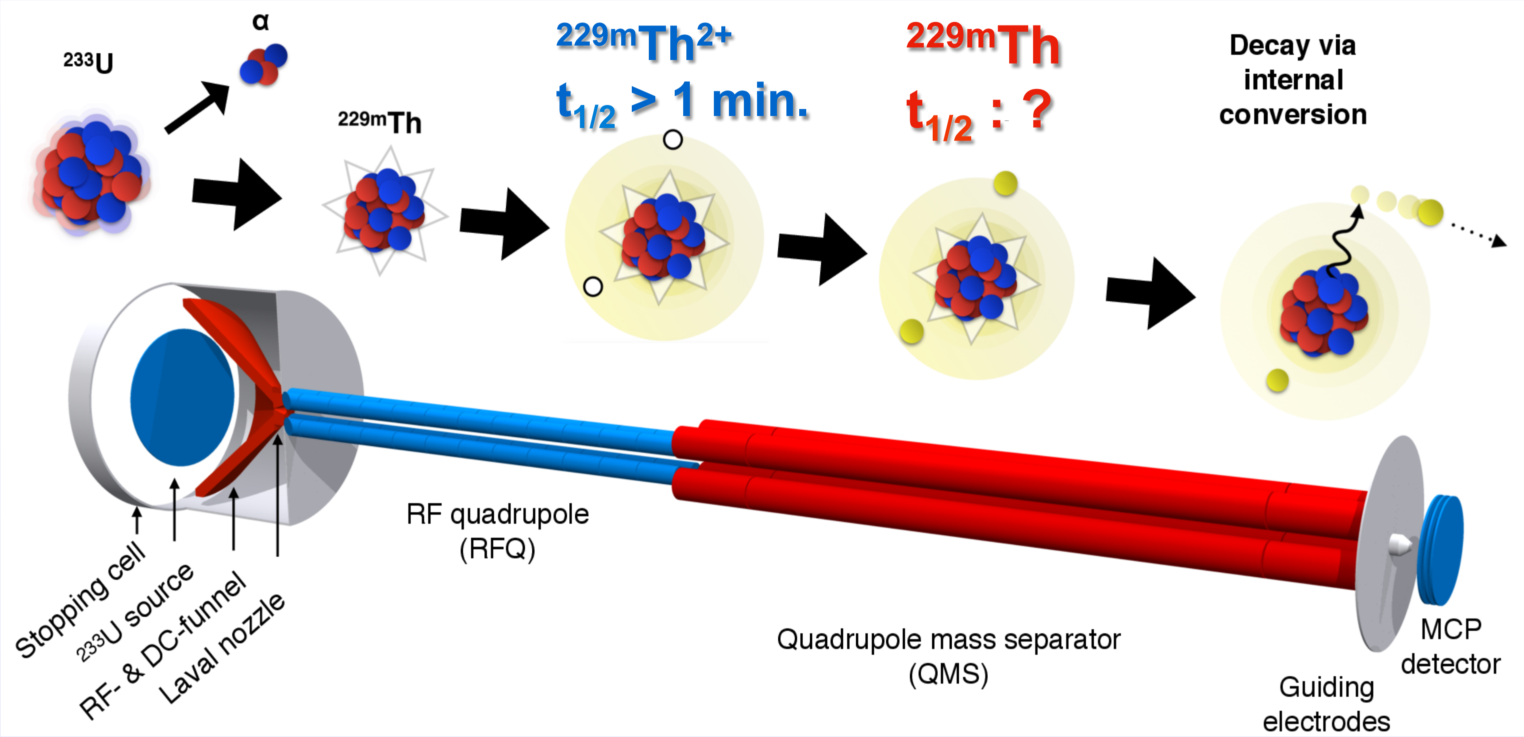
could all be excluded, mostly by several ways

No.	Way of background exclusion	Type of background			
		A	B	C	D
1	Signal comparison between $^{229}Th^{2+}$ and $^{233}U^{2+}$	x	x		
2	Comparative $^{229}Th^{2+}$ and $^{233}U^{2+}$ signal behaviour as a function of MCP surface voltage	x	x		
3	Signal comparison between ^{229}Th and ^{230}Th	x	x	x	
4	Signal comparison between $^{229}Th^{2+}$ and $^{229}Th^{3+}$				x
5	Signal comparison between ^{229}Th originating from chemically purified and unpurified ^{233}U sources				x
6	Exclusion based on signal appearance and the ^{229m}Th half-life limit				x
7	Search for α and β decays using Si and LN_2 cooled Si(Li) detectors				x

Lively Echo to Publication

Next steps: Halflife determination

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



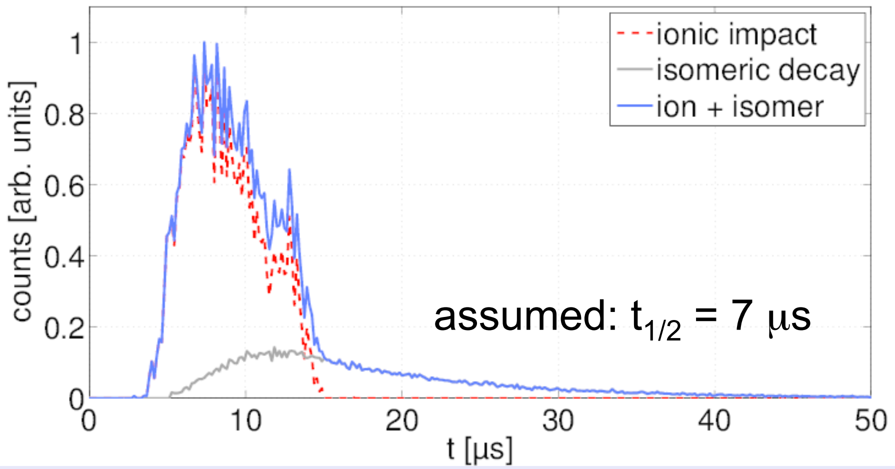
- expected conversion coefficient: ca. 10^9

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

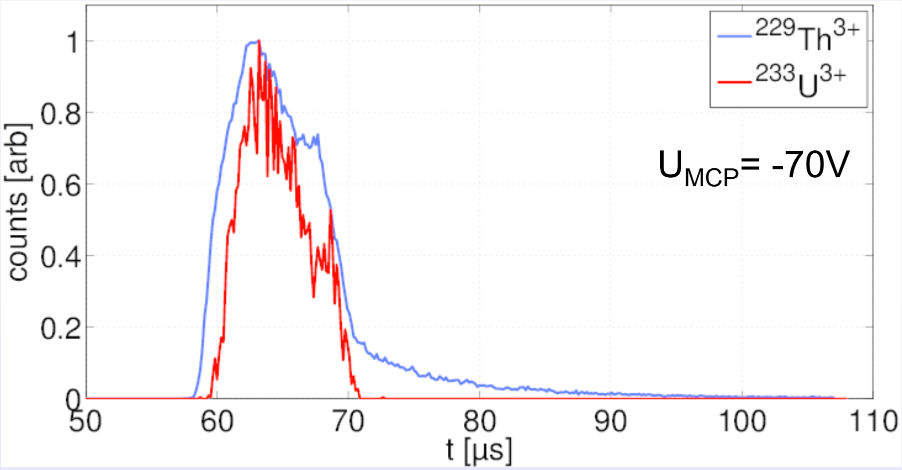
B. Seiferle et al., to be published

Halflife: Neutral ^{229m}Th

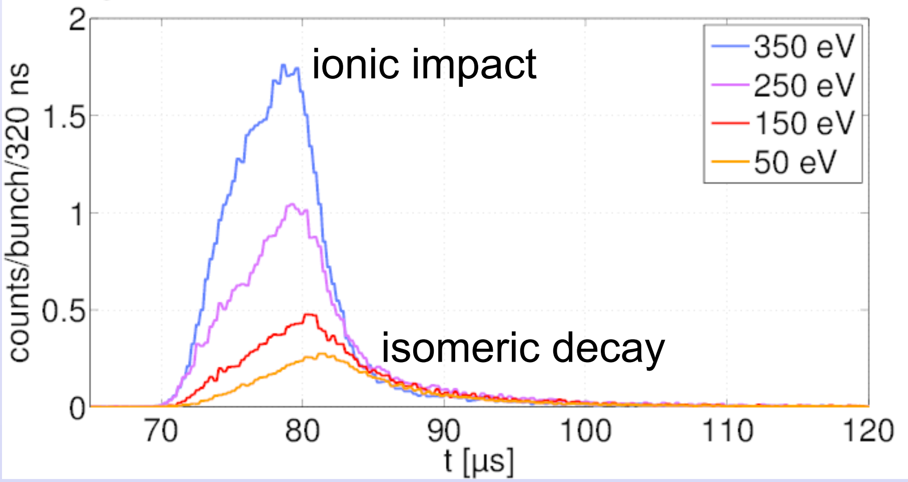
simulation:



measurement:



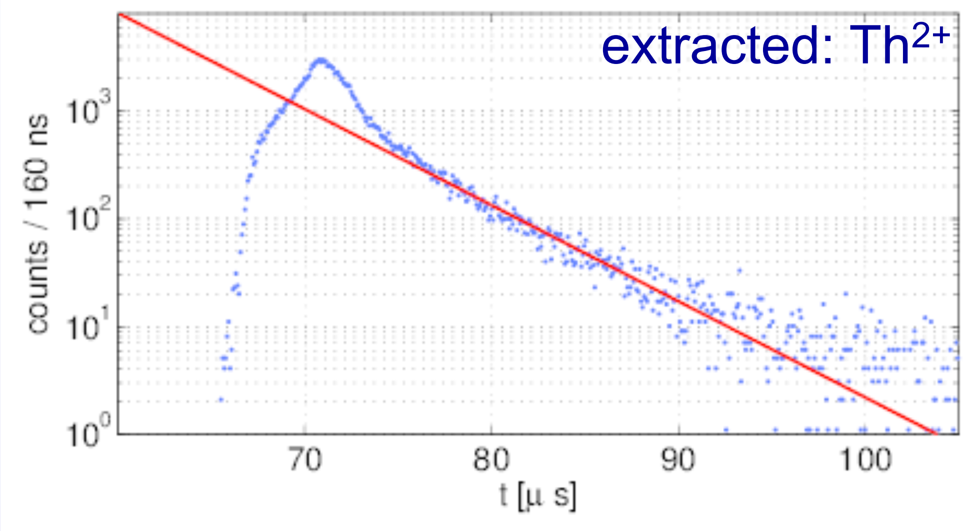
MCP signal of $^{229(m)}Th^{2+}$ ion bunches with different kinetic energies:

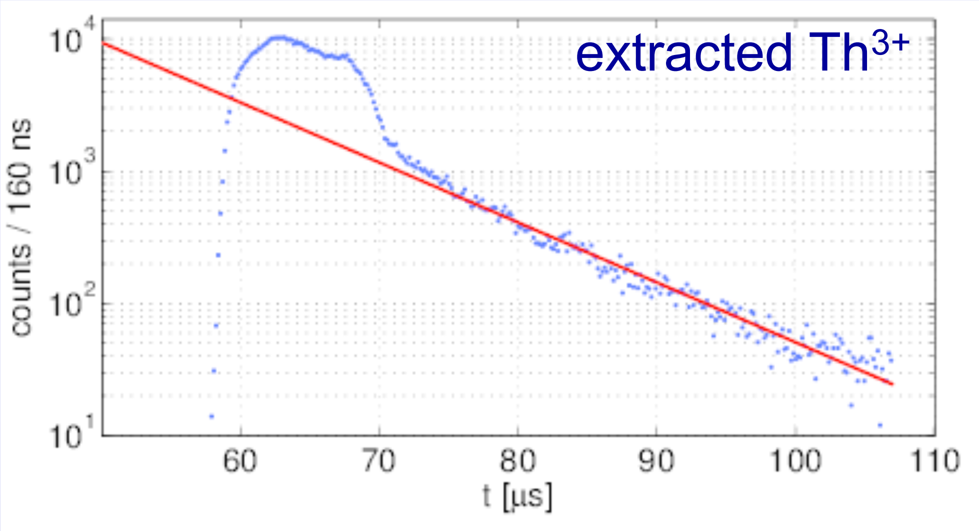


- bunch width: ca. 10 μs
- 600- 800 $^{229(m)}Th^{2+,3+}$ ions/bunch

B. Seiferle et al., to be published

Halflife: Neutral ^{229m}Th





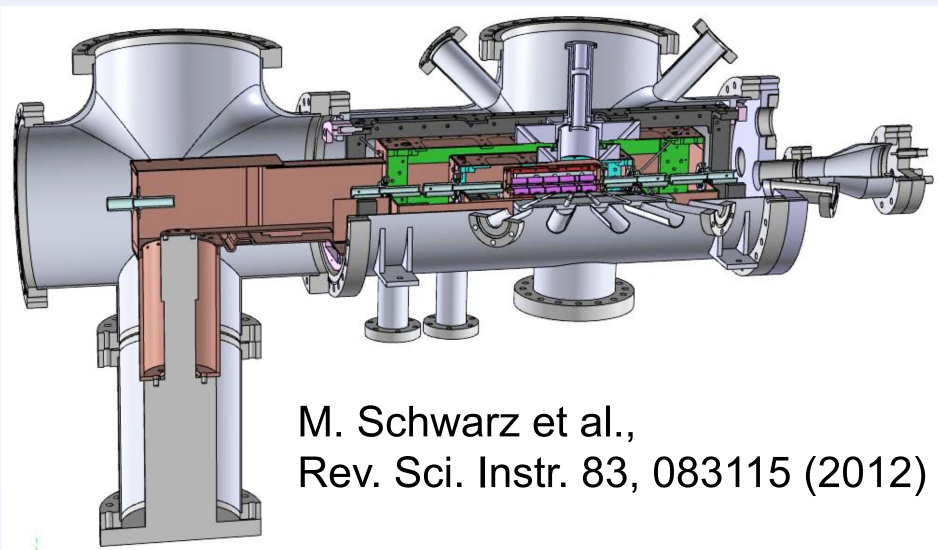
based on expected charged isomeric half-life of a few 10^3 s:
confirms expected conversion coefficient of ca. 10^9 → photonic decay branch ??

Halflife: Charged ^{229m}Th

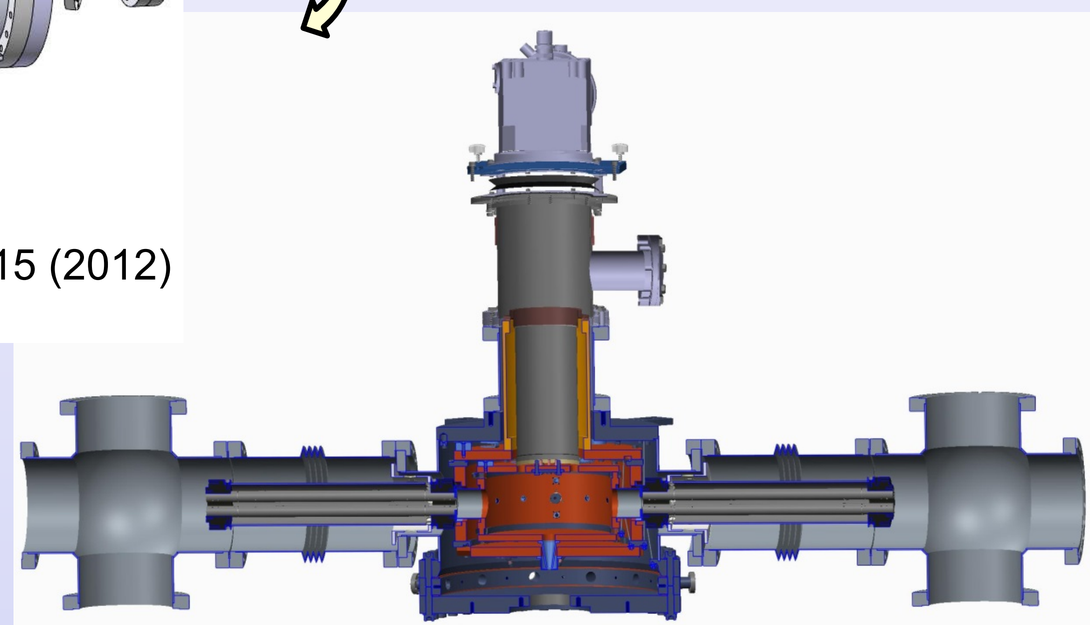
- $^{229m}Th^{q+}$: longer ion storage time needed (→ UHV)
  → cryogenic linear Paul trap:

- in operation at MPI-K (HD): CryPTEx

→ operating at 4 K (p(H_2) ~ 10^{-15} mbar)
→ ion storage time ≤ 30 h
→ laser access ports



M. Schwarz et al.,
Rev. Sci. Instr. 83, 083115 (2012)



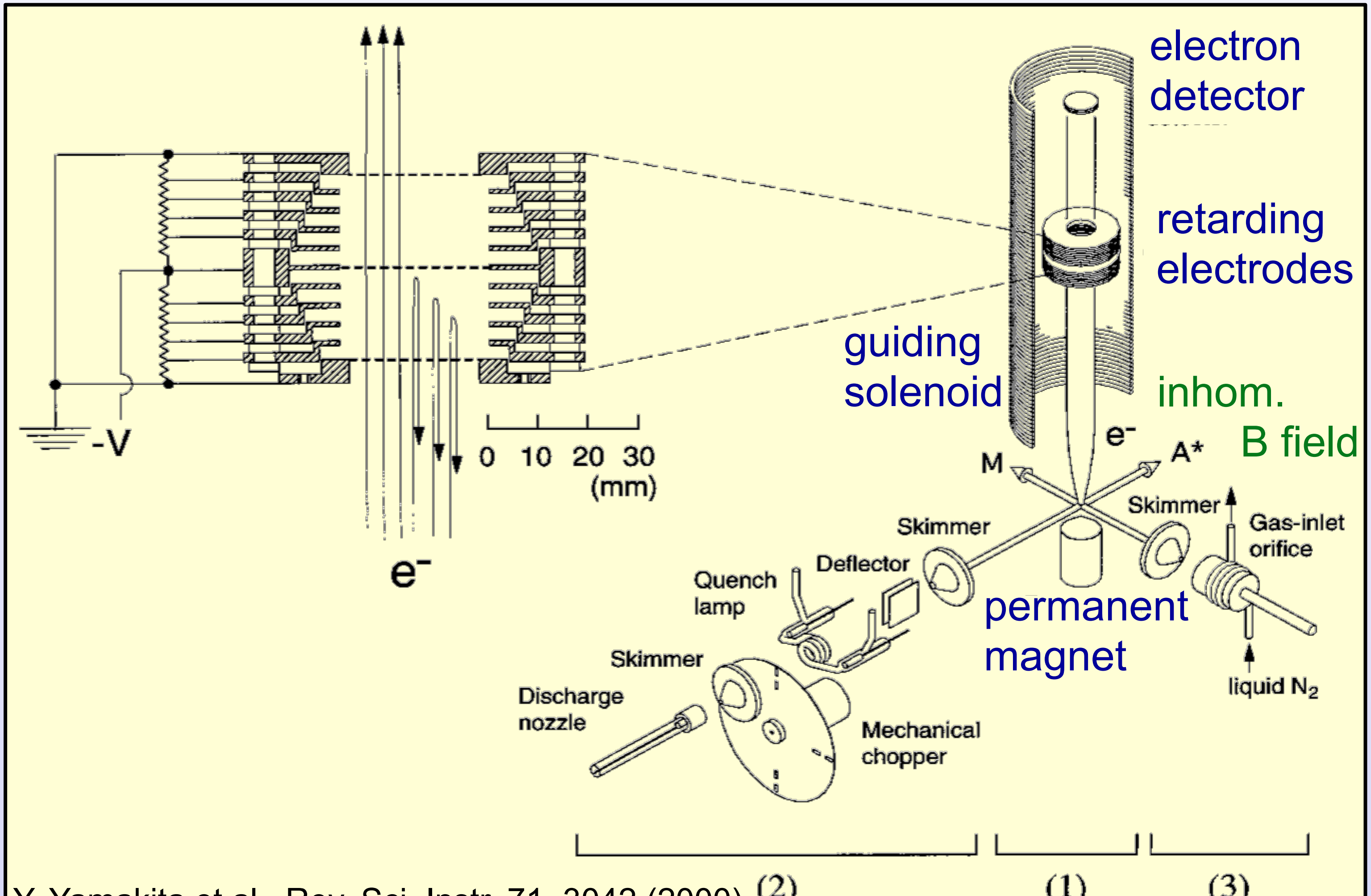
→ prerequisite for laser manipulation of $^{229m}Th^{3+}$

thanks to: MPIK (J. Crespo et al.), PTB (P. Schmidt)

"Holy Grail": Excitation Energy

our present constraint: 6.31 eV < E(^{229m}Th) < 18.3 eV

- Magnetic bottle spectrometer: large acceptance needed (~ 4π)
  10000 ^{229}Th ions extracted/sec. → 200 e$^-$/s



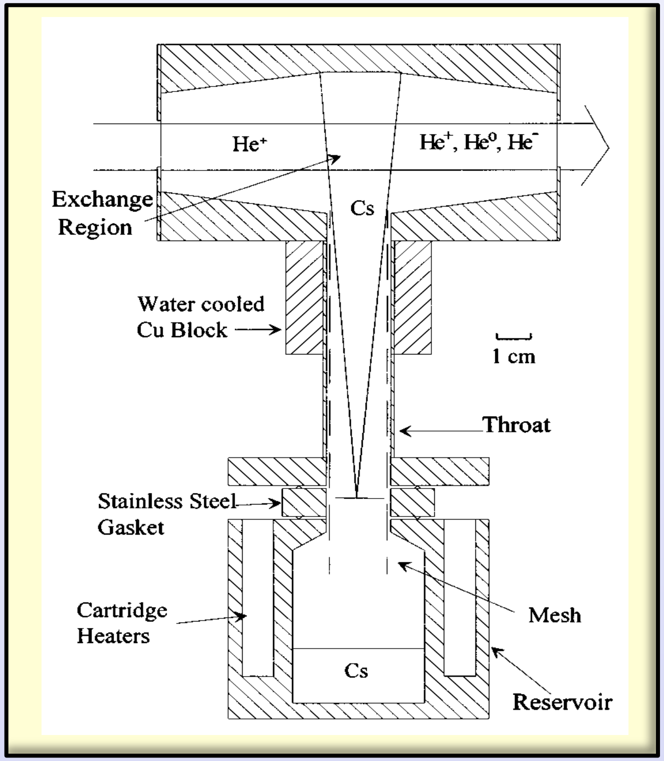
Y. Yamakita et al., Rev. Sci. Instr. 71, 3042 (2000)

Excitation Energy

- $^{229m}Th^{2+,3+}$ neutralization with subsequent IC:

  - metallic catcher electrode
  - but: how to control/avoid surface effects (work function) ?

  → contact-free neutralization: charge-transfer cell

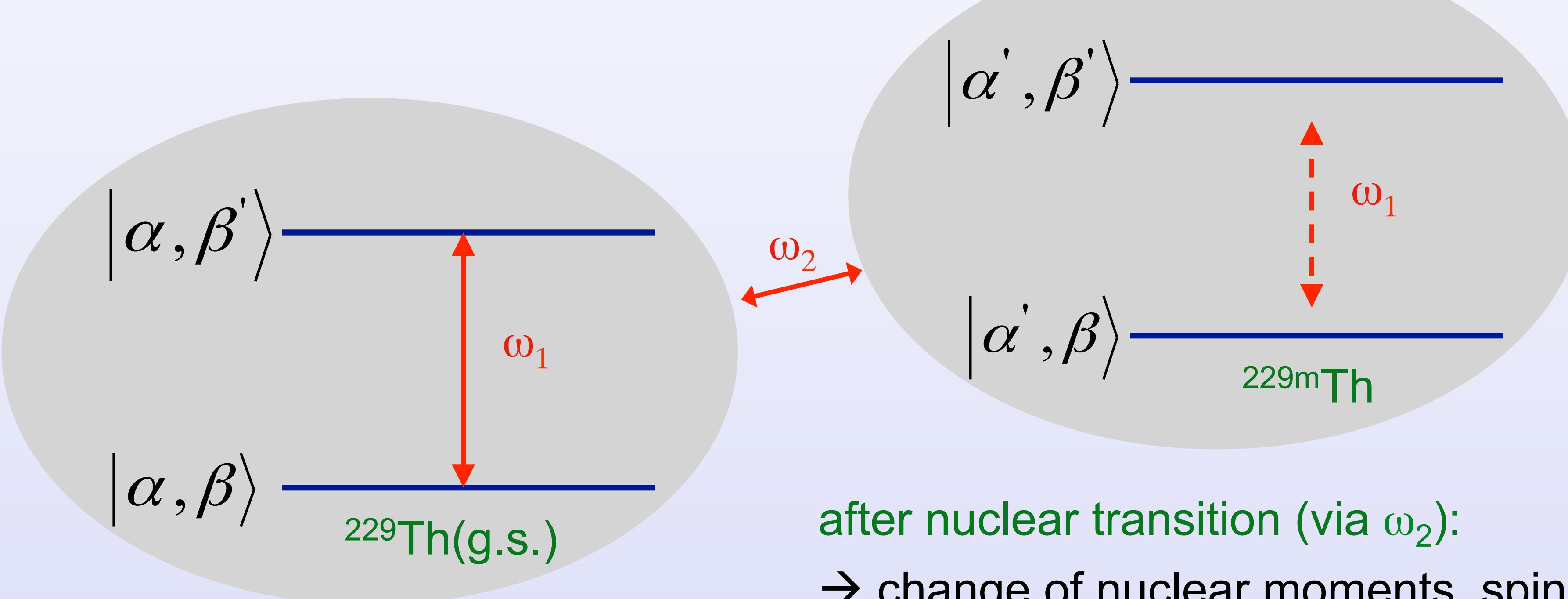


finally expected:
$\Delta E < 50$ meV

M.G. Helander et al., Applied Surface Science 256, 2602 (2010)

Laser diagnostics of isomer excitation

- use closed 2-level system in electron shell:



after nuclear transition (via ω_2):

→ change of nuclear moments, spin
→ change of hyperfine splitting, total angular momenta
→ ω_1 out of resonance (~GHz)
→ drop in resonance fluorescence

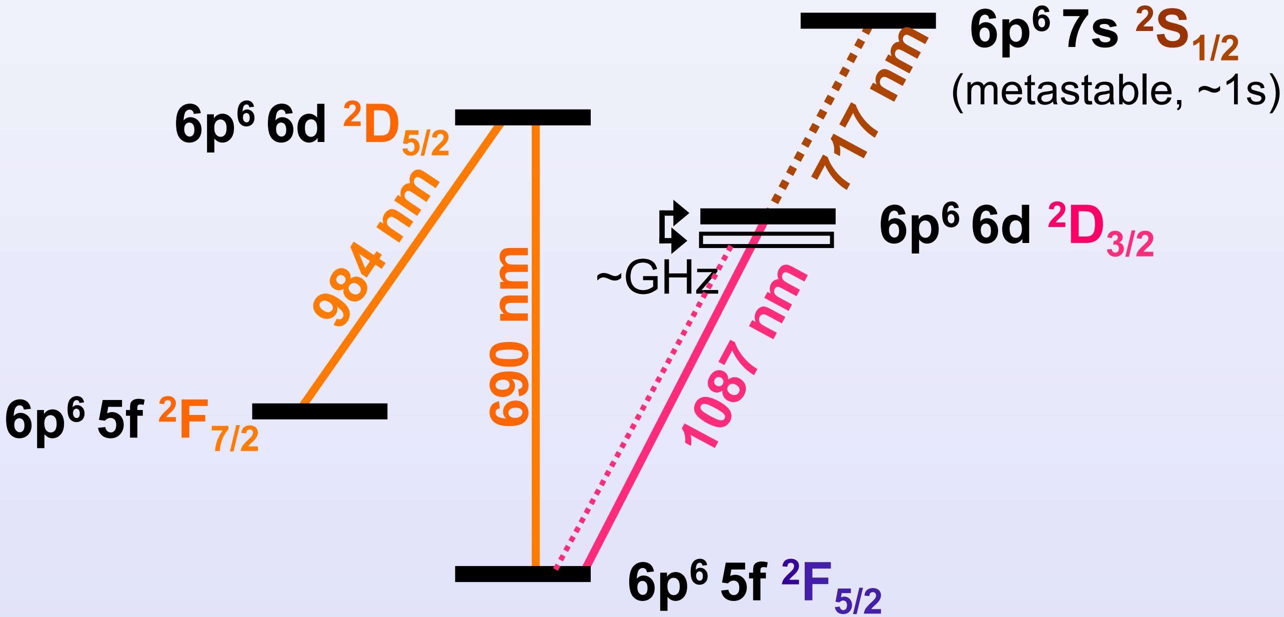
α: nuclear, β: electronic

⟶ double resonance method Dehmelt's 'electron shelving'

Peik, Tamm, Eur. Phys. Lett. 61 (2003) 181

$^{229}Th^{3+}$: suitable level scheme

- simple structure: alkali-like (Rn core + single valence electron)



→ closed 3-level Λ system and closed 2-level system available for laser cooling and fluorescence detection

- constant resonance fluorescence scattering with closed optical transition
- after nuclear excitation: out of resonance

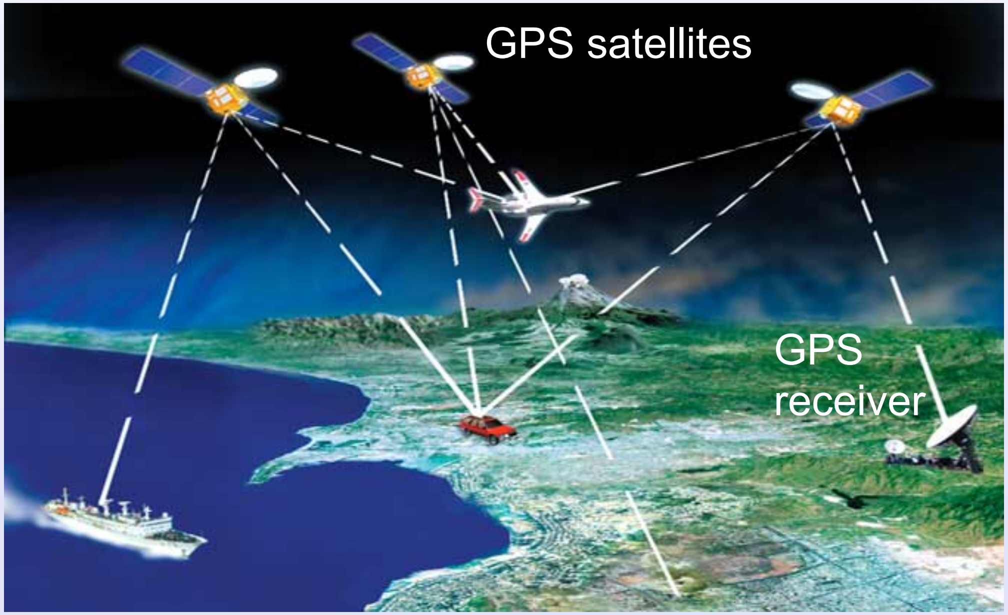
→ spectroscopy of laser-cooled $^{229}Th^{3+}$ (e.g. in linear Paul trap)

Why better clocks at all ?

every time that clock precision has improved, new and frequently unexpected applications have emerged

- example: **GPS navigation**
- present technology:
  - ‘thermal’ microwave atomic clocks (Rb, Cs)
  - GPS, GLONASS: active
  - Galileo, Beidou, IRNSS: under construction

  → positioning precision: 1-10 m

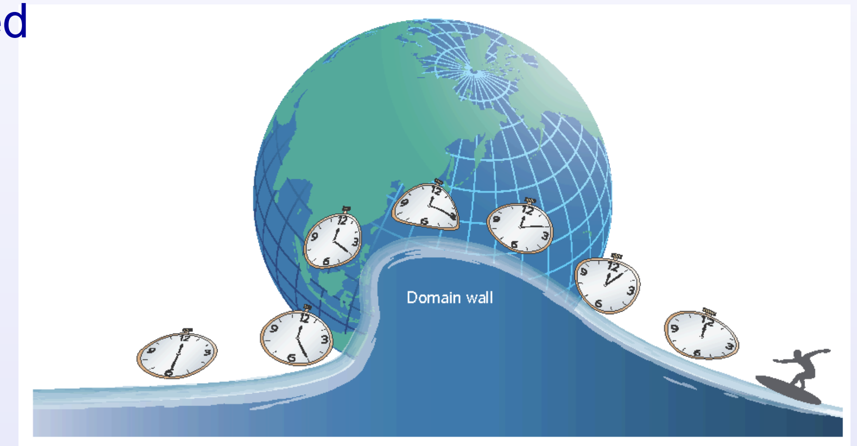


- goal: positioning to cm – mm
  - autonomous driving
  - freight-/ component tracking
  - …..
- requirements:
  - better clocks (10^{-16} – 10^{-18})
  - robust, small, low power consumption …

Further potential applications

- (networks of) ultra-precise clocks could be used to search for dark matter

  → Derevianko & Pospelov, Nature Physics 10, 933–936 (2014)
  - topological dark matter: clumped to point-like monopoles, 1D strings, 2D 'domain walls'



- networks of clocks can be used as 3D gravity sensors

  → Ludlow, Boyd, Ye, Peik, Schmidt, Rev. Mod. Phys. 87, 637–701 (2015)

$$\Delta f / f = -\Delta U / c^2$$

Δf: clock frequency difference
ΔU: gravitational potential difference

- best present clocks: detect gravitational shifts of ± 2 cm
- "relativistic geodesy" or "chronometric leveling":
  → allows long-distance potential difference measurements
- precise, fast measurements of nuclear clock network:
  e.g. monitor volcanic magma chambers, tectonic plate movements

The nuClock Consortium

- FET-OPEN project within EU Horizon 2020:

  → experiment, theory, industrial R&D

TU Wien (T. Schumm et al., Coord.)

Univ. Jyväskylä (I. Moore et al.)

Univ. Heidelberg (C. Enss et al.)

MPI-K, HD (A. Pálffy et al.)

PTB Braunschweig (E. Peik et al.)

MPQ Garching (T. Udem et al.)

TOPTICA Photonics AG, Munich

LMU Munich (PT et al.)

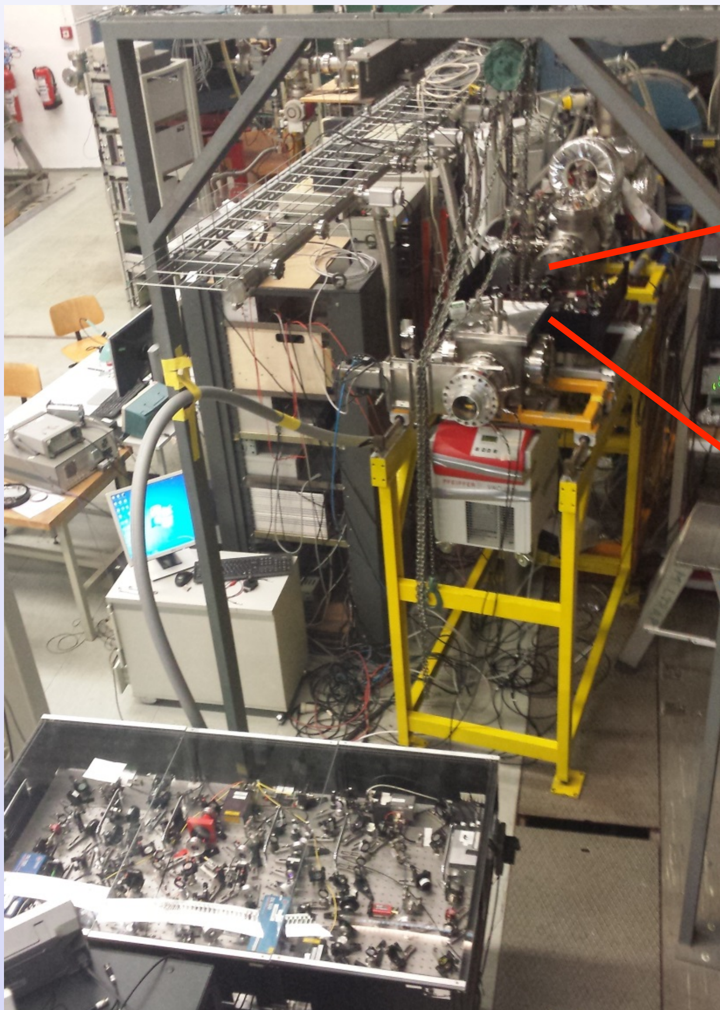
associated:

- Okayama Univ., Japan (SPRING8)
- Metrology Light Source (MLS), Berlin-Adlershof
- U Mainz/ HIM (C. Düllmann et al.)
- Kerstin Ergenzinger (Berlin): artist, via FEAT (future emerging art and technology)

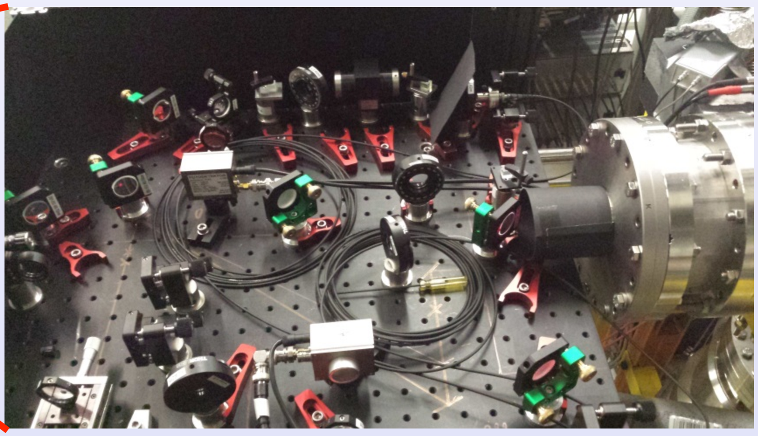
http://www.nuclock.eu/

Collaboration with PTB Braunschweig

- E. Peik, M. Okhapkin et al.: targeting hyperfine structure of $^{229m}Th^{1+,2+}$



collinear laser spectroscopy:

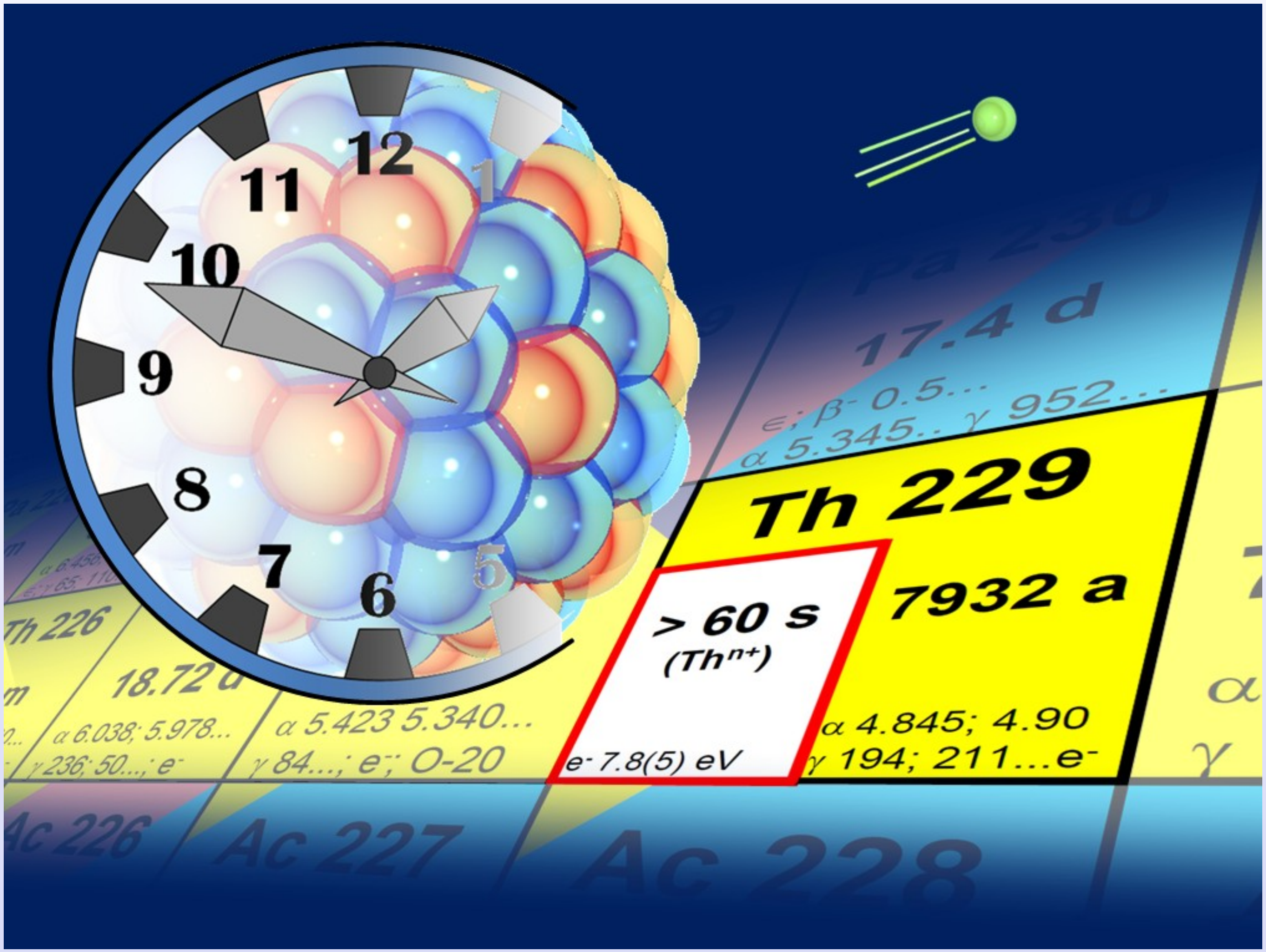


- laser interaction with ^{229}Th ions extracted from QMS
- measurement campaign started

Summary

- successful experimental approach based on spatial decoupling of population and deexcitation via buffer-gas stopping cell
- efficient extraction of pure $^{229(m)}Th^{2,3+}$ ions achieved
- VUV optical detection system was designed and commissioned
- radiative decay channel in neutral ^{229m}Th drastically reduced (in favor of IC)
- IC decay channel provided first direct (and unambiguous) identification of isomeric ground-state transition in ^{229m}Th
- constraints of ^{229m}Th properties: $6.3 \text{ eV} \leq E^* \leq 18.3 \text{ eV}$, $\tau > 60$ s
- next steps: determination of halflife for neutral ^{229}Th
  towards determination of isomeric excitation energy (magnetic bottle spectrometer)
- started: setup of cryogenic Paul trap
  → to achieve longer storage times for halflife measurement in charged ^{229m}Th
  → prerequisite also for laser manipulation of $^{229m}Th^{3+}$

we made a big step towards the ultimate goal of a Nuclear Clock ….



but many more are yet to come ….

Thanks to ….

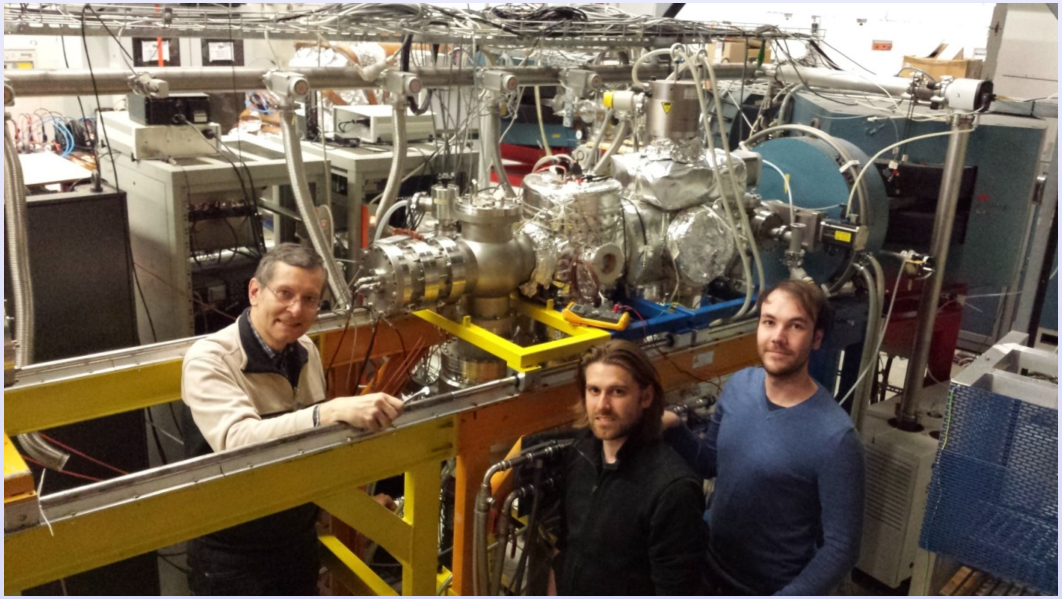
LMU Munich: **L. v.d. Wense, B. Seiferle**, J.B. Neumayr, H.-J. Maier, H.-F. Wirth, D. Habs

GSI Helmholtzzentrum f. Schwerionenforschung, Darmstadt & Helmholtz-Institut Mainz: M. Laatiaoui

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

THE FRAMEWORK PROGRAMME FOR RESEARCH AND INNOVATION
HORIZON 2020

DFG Deutsche Forschungsgemeinschaft



Thank you for your attention !