The ^{229}Thorium Isomer: Doorway to a Nuclear Clock

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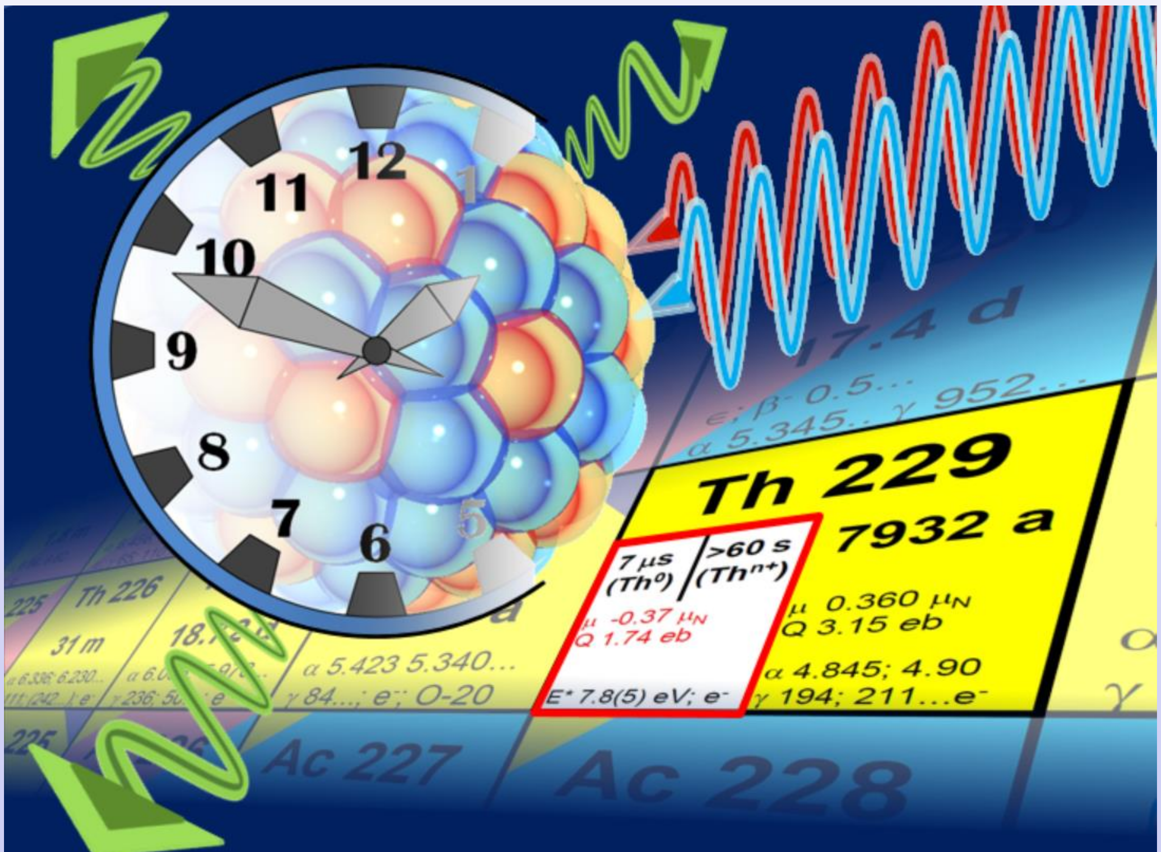
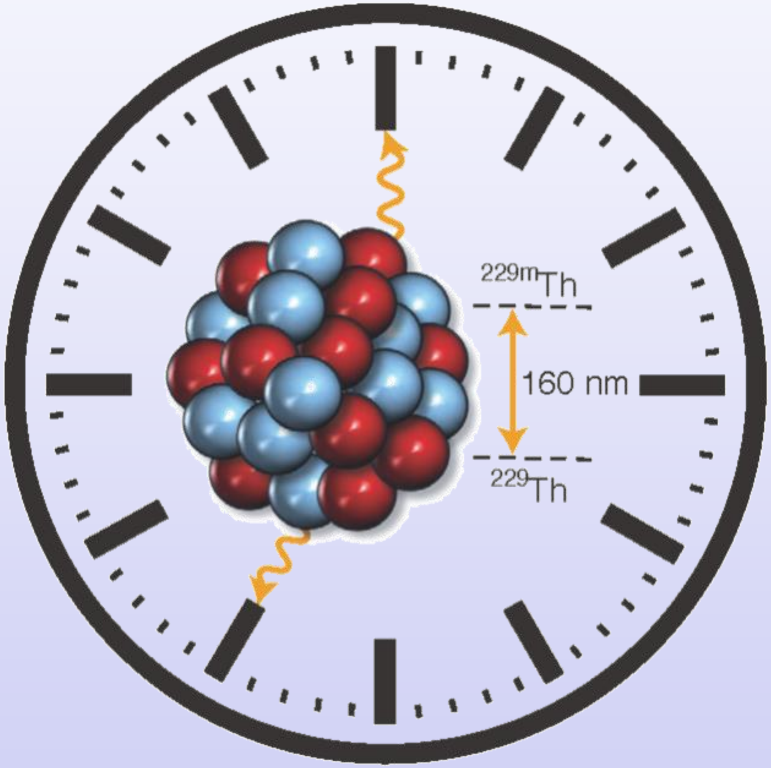


Outline

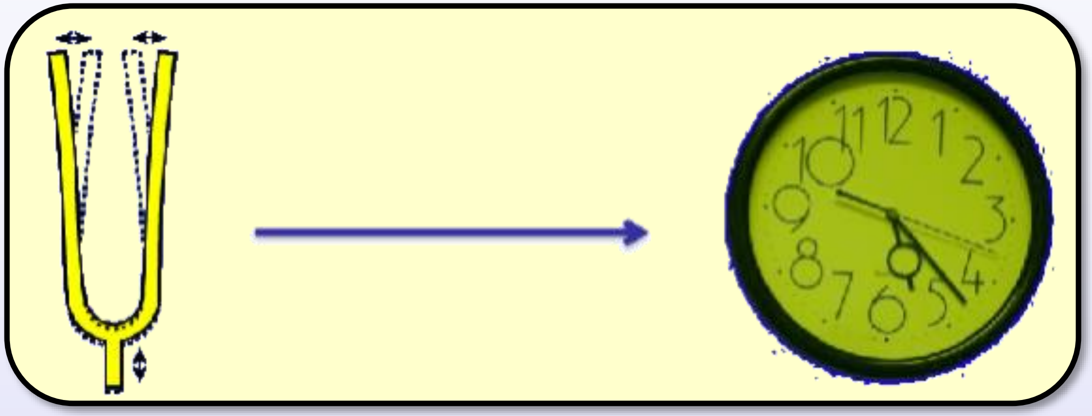
- Clock basics
- ^{229m}Th properties and prospects
- "Search & Characterization Phase" (nuclear physics driven)

  - experimental approach & setup
  - first identification
  - halflife
  - hyperfine structure
  - excitation energy
- "Consolidation & Realization Phase" (laser driven)

  - ongoing efforts and upcoming next steps
- Summay/Conclusion



What makes a Clock ?



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

Which Type of Clocks ?

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: 2.5×10^{-19}

nuclear clocks ??

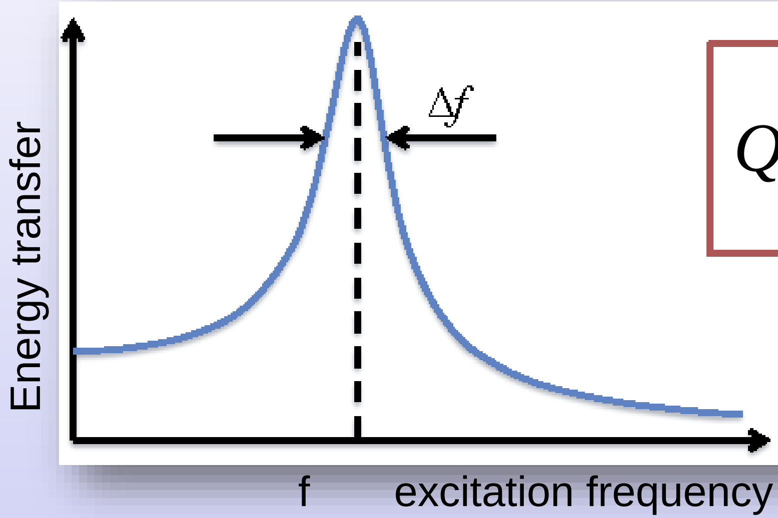
^{229m}Th

Clock Performance

- **continuous operation:**

  - feedback on resonance (e.g. of atomic transition)
  - 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: $Q > 10^{15}$??

Applications of Nuclear Clocks

- **Improved precision of satellite-based navigation** (GPS, Galileo..): m → cm (mm ?)

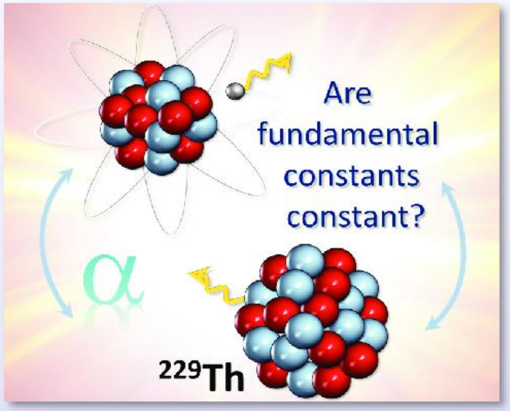
- **Temporal variation of fundamental constants**

  - theoretical suggestion: temporal (spatial) variations of fundamental "constants"

$$\dot{\alpha}/\alpha = (1.0. \pm 1.1) \cdot 10^{-18}\ \mathrm{yr}^{-1}$$

R. Lange et al., arXiv:2010.06620

  - enhanced sensitivity by (10^2 – 10^5) of ^{229m}Th expected



- **Search for Dark Matter**
  - topological dark matter: clumped to point-like monopoles, 1D strings, 2D 'domain walls'
  - use networks of ultra-precise synchronized clocks

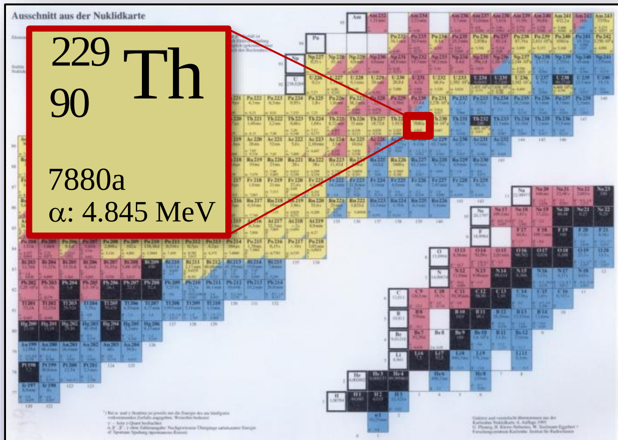
- **3D gravity sensor:** 'relativistic geodesy'
  - best present clocks: detect gravitational shifts of ± 1 cm
  - precise, fast measurements of nuclear clock network: monitor volcanic magma chambers, tectonic plate movements

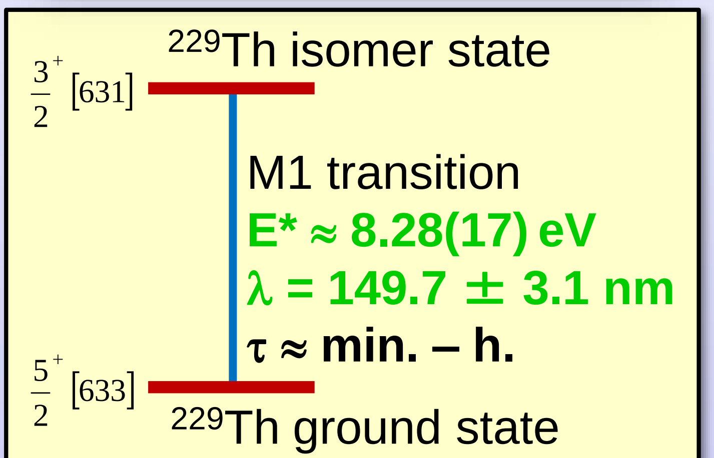
$$\Delta f / f = -\Delta U / c^2$$

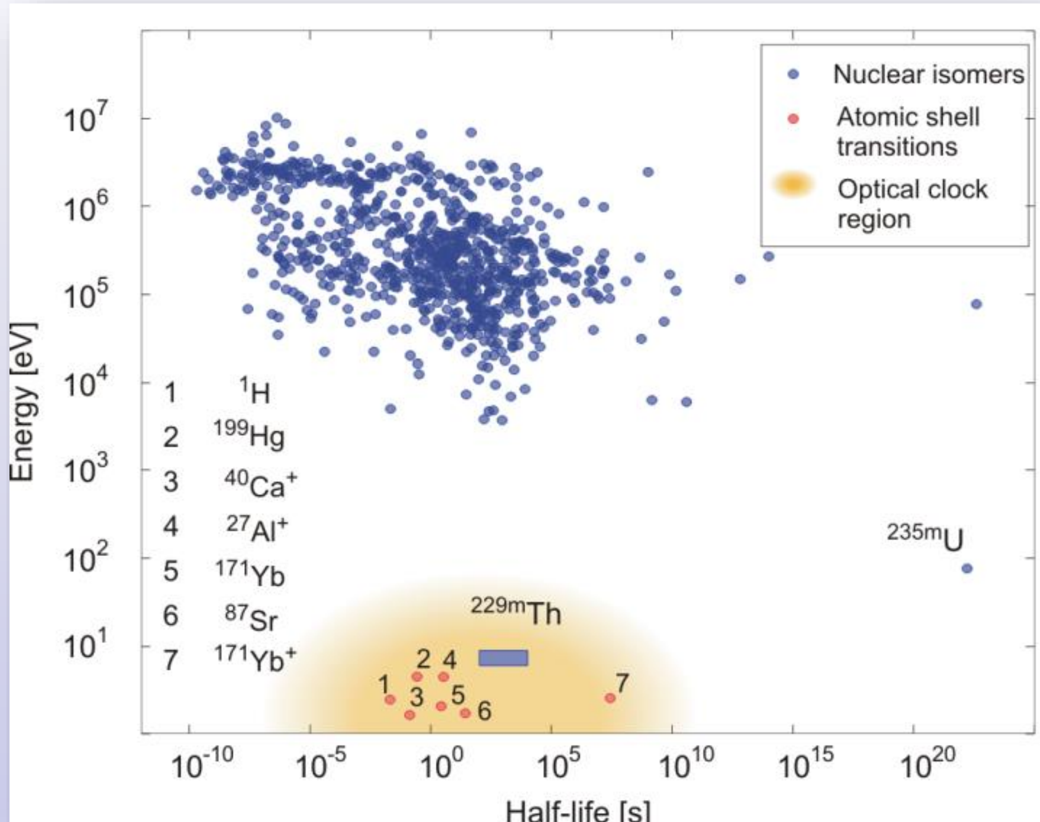
f: clock frequency
U: gravitat. potential

PT et al., Annalen d. Physik 531, 1800391 (2019)

Unique properties of the ^{229m}Th Isomer







lowest E* of all ca. 184000 presently known nuclear excited states

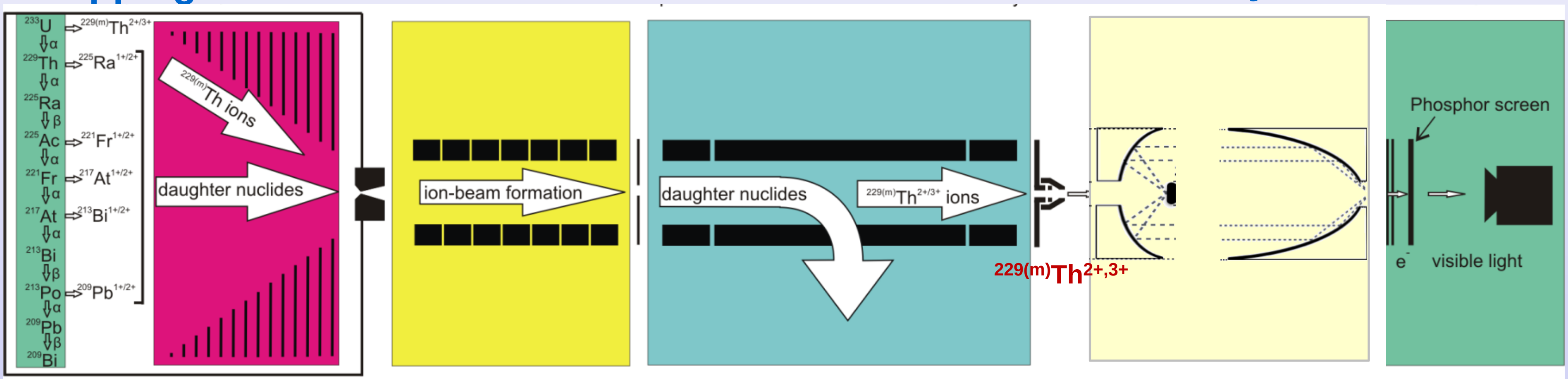
→ ΔE/E ~ 10^{-20}:

extremely stable nuclear frequency standard: ‘nuclear clock’

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

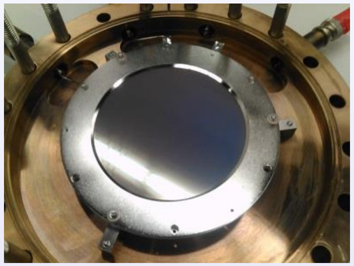
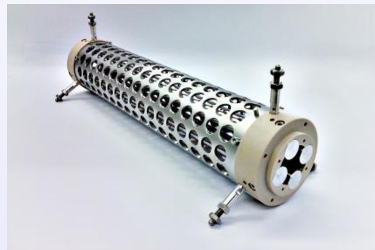
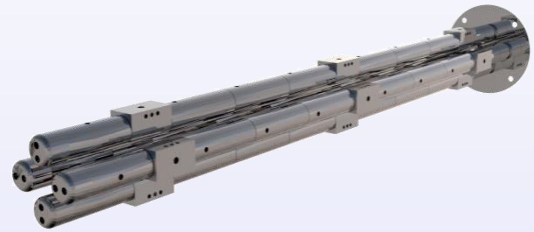
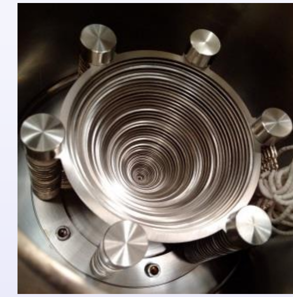


^{233}U source | RF + DC funnel | RF quadrupole ion guide | Quadrupole mass separator | UV detection | MCP | CCD

~30 mbar (He) | 10^{-2} mbar | 10^{-6} mbar

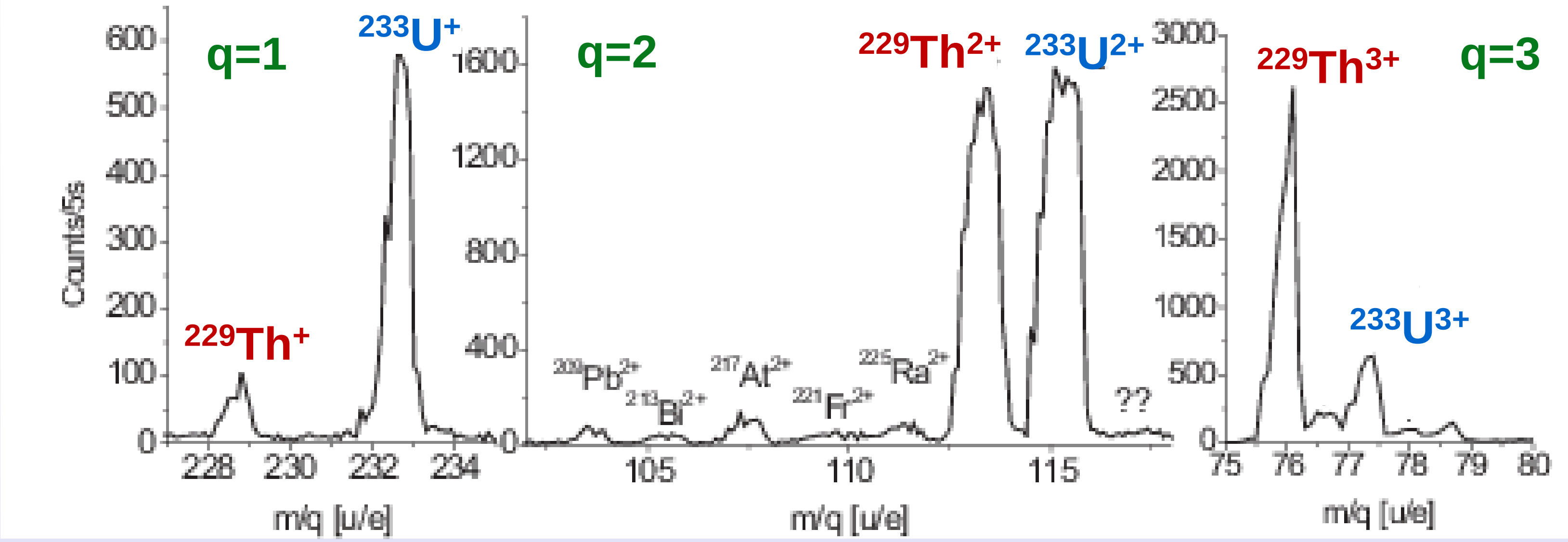
Experimental Setup

located at Maier-Leibnitz Laboratory, Garching:


Ion Extraction from Buffer Gas Cell

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



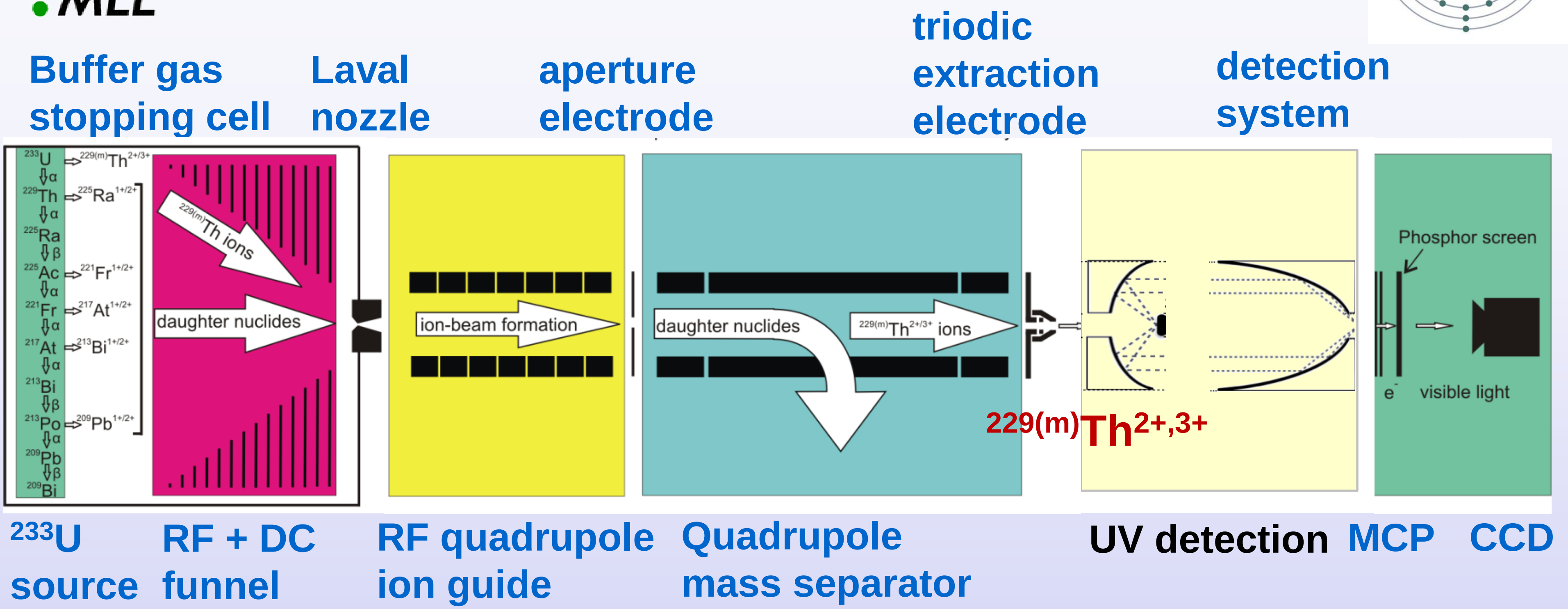
element	1⁺ [%]	2⁺ [%]	3⁺ [%]
Th	0.37(7)	5.5(11)	10(2)
Fr	21.0(42)	16.0(32)	≤ 1.5.10⁻³
Rn	5.8(12)	9.3(19)	0.053(11)
At	8.6(17)	13.0(26)	0.033(7)
Po	7.3(15)	8.1(16)	≤ 0.0021
Bi	4.3(9)	21.0(42)	0.083(16)
Pb	2.2(4)	11.0(22)	≤ 0.012

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

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

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

Isomer Detection Process

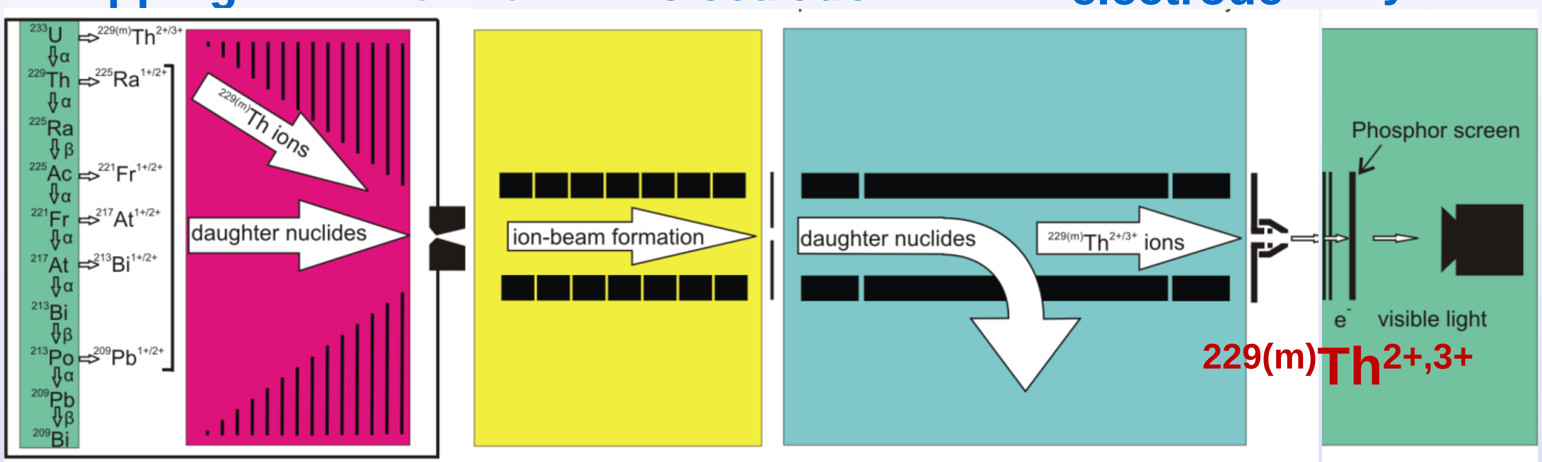


→ **VUV-optical detection system designed, built, commissioned, operated**

- **Expectation:** VUV photonic signal, well separated from background
- **But: no** UV photons observed from collection surface
- **Suspicion:** deexcitation occurs predominantly radiationless
  alternative decay branch ?
  Internal Conversion ? → search for electrons instead for photons

Isomer Detection Process

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



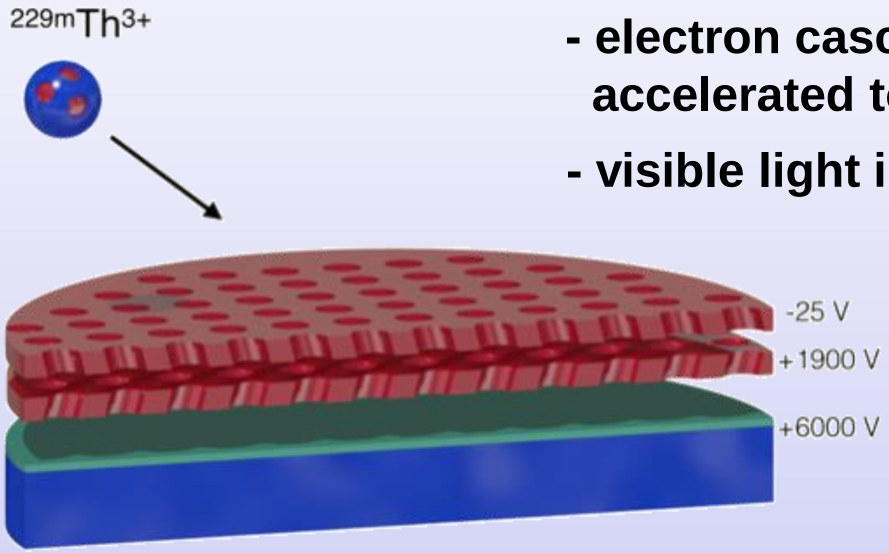
^{233}U source | RF + DC funnel | RF quadrupole ion guide | Quadrupole mass separator | MCP | CCD

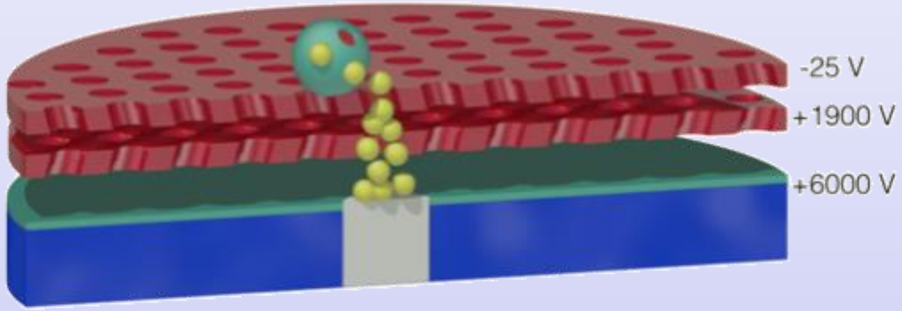
→ accumulate $^{229(m)}$Th ions directly onto MCP surface

Isomer Detection Process

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

- **impinging directly onto MCP surface behind triode exit**
- **'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**





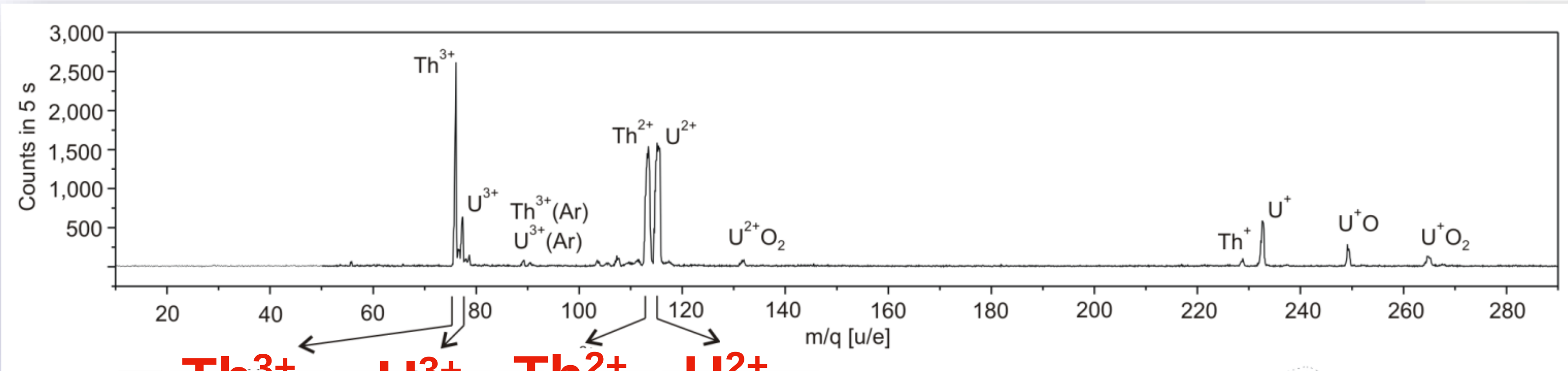
- internal conversion (IC) energetically allowed for neutral thorium:

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

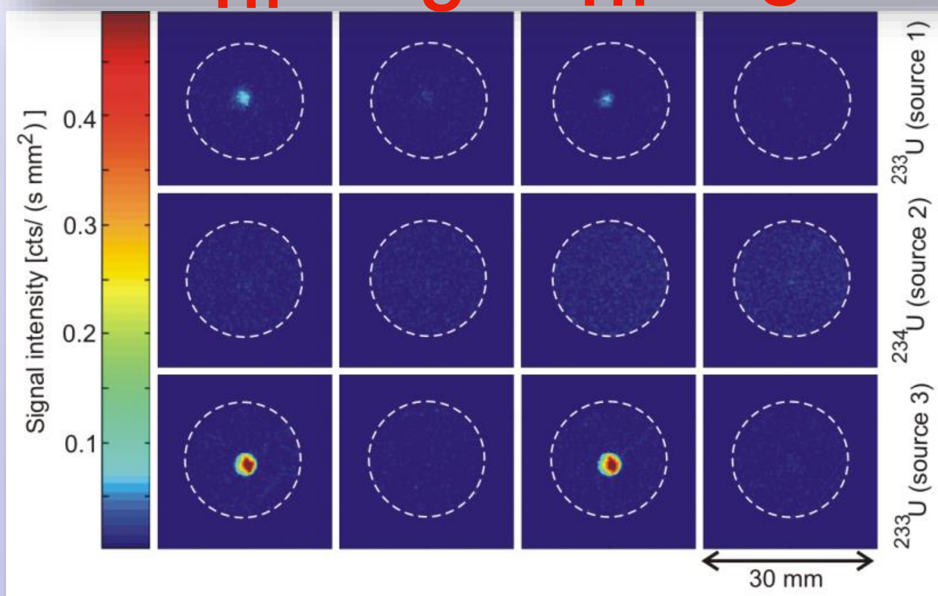
- isomer lifetime expected to be reduced by ca. 10^{-9} (from ~10^4 s → ~ 10 μs)
- Th^{q+} ions: IC is energetically forbidden, radiative decay branch may dominate

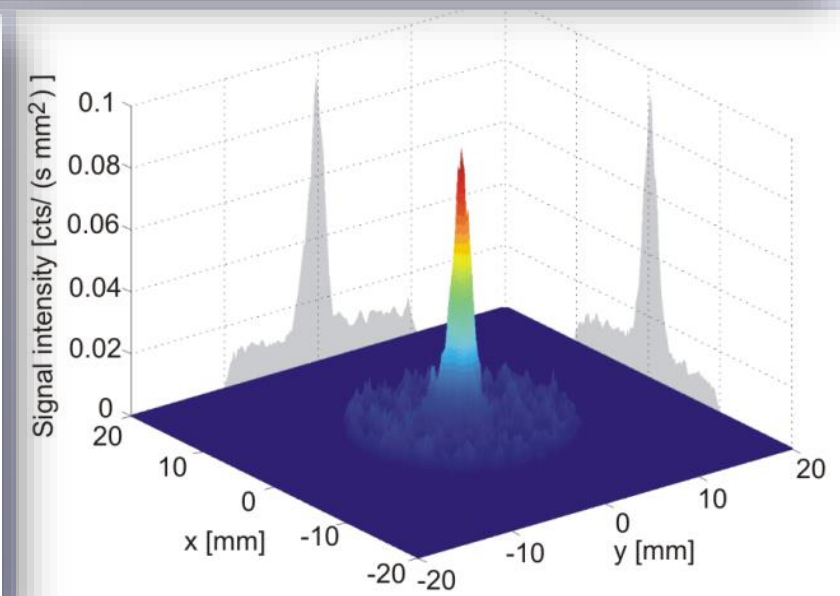
Direct Signal of IC Decay from ^{229m}Th

L. v.d. Wense, PT et al., Nature 533, 47-53 (2016)



Th^{3+} U^{3+} Th^{2+} U^{2+}

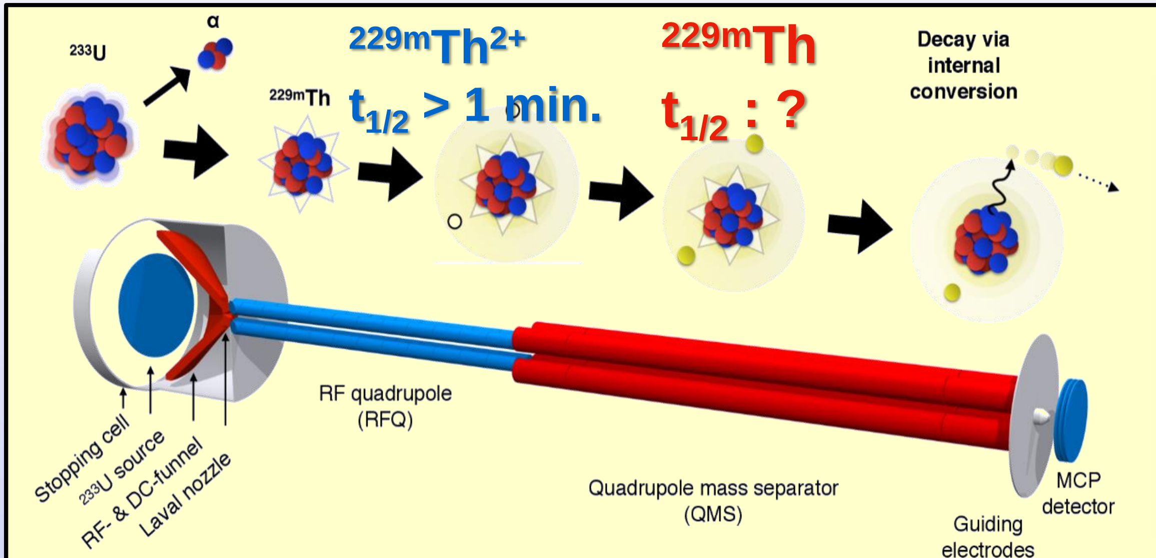


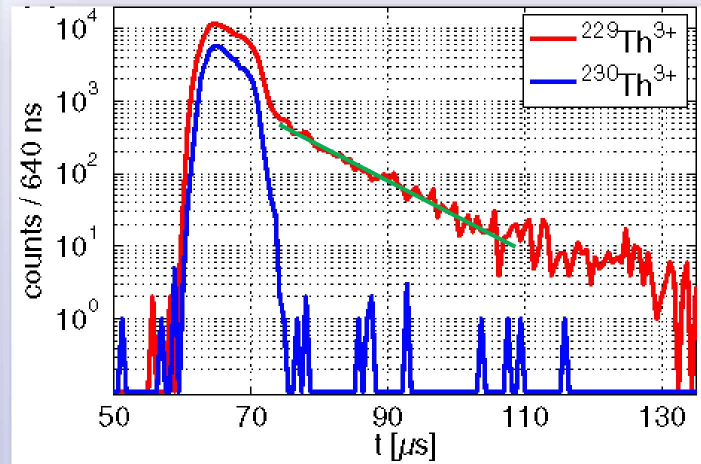


clear signal from Th^{3+}, Th^{2+}
no signal from U^{3+}, U^{2+}

Halflife of (neutral) ^{229m}Th

- operate segmented RFQ as linear Paul trap: pulsed ion extraction
- ion bunches: width ca. 10 μs, ~ 400 $^{229(m)}Th^{2+,3+}$ ions/bunch





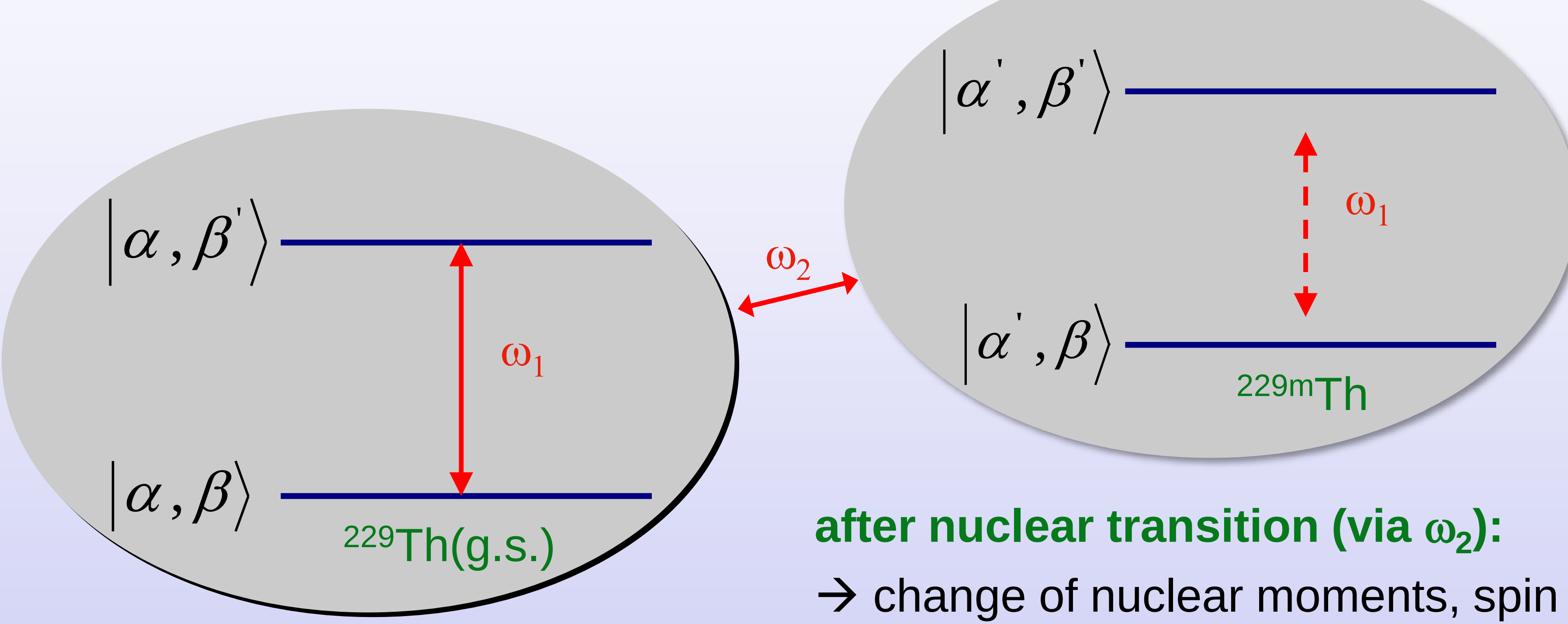
- charged $^{229m}Th^{2+}$: $t_{1/2}$ > 1 min. (limited by ion storage time in RFQ, i.e vacuum quality)
- after neutralization on MCP surface: $t_{1/2} = 7 \pm 1\ \mu s$

→ in agreement with expected $\alpha_{IC} = N_e/N_\gamma \sim 10^9$

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

Laser-based diagnostics of isomer excitation

- use closed 2-level system in electron shell:



α: nuclear, β: electronic

⟶ double resonance method
Dehmelt's 'electron shelving'

after nuclear transition (via ω_2):

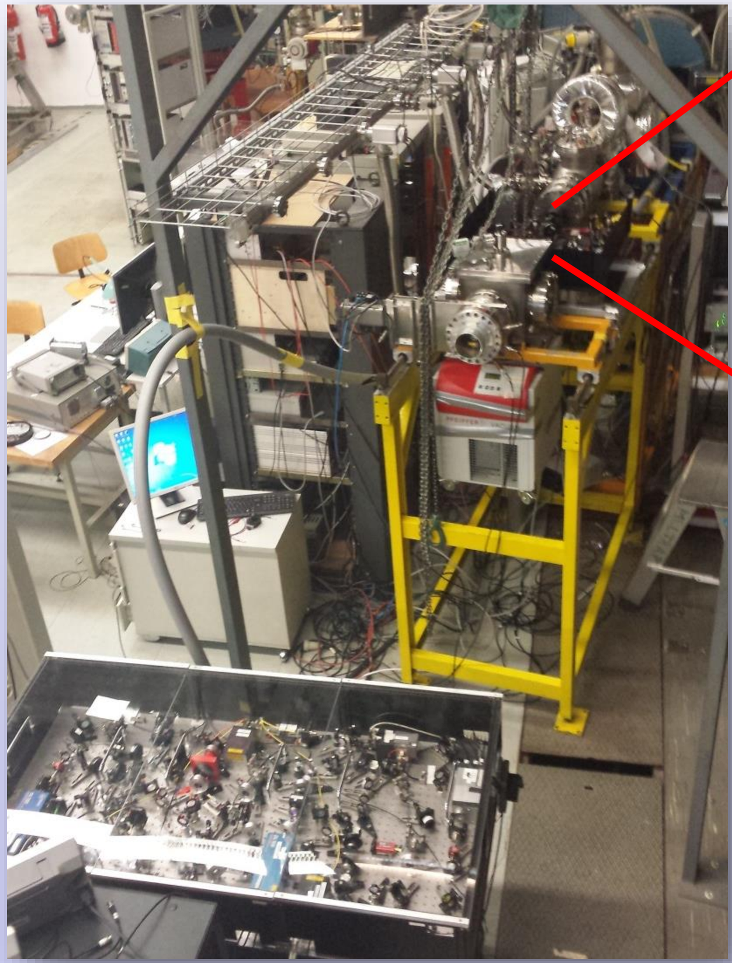
→ change of nuclear moments, spin
→ change of hyperfine splitting, total angular momenta
→ ω_1 out of resonance (~GHz)
→ drop in resonance fluorescence

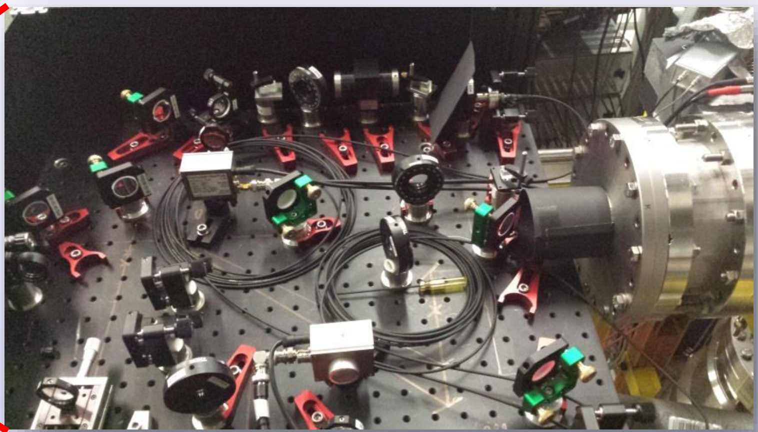
Peik, Tamm, Eur. Phys. Lett. 61 (2003) 181

Collinear Laser Spectroscopy of ^{229m}Th

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

isomer beam (LMU) + laser system (PTB) → resolve hyperfine structure of $^{229m}Th^{2+}$

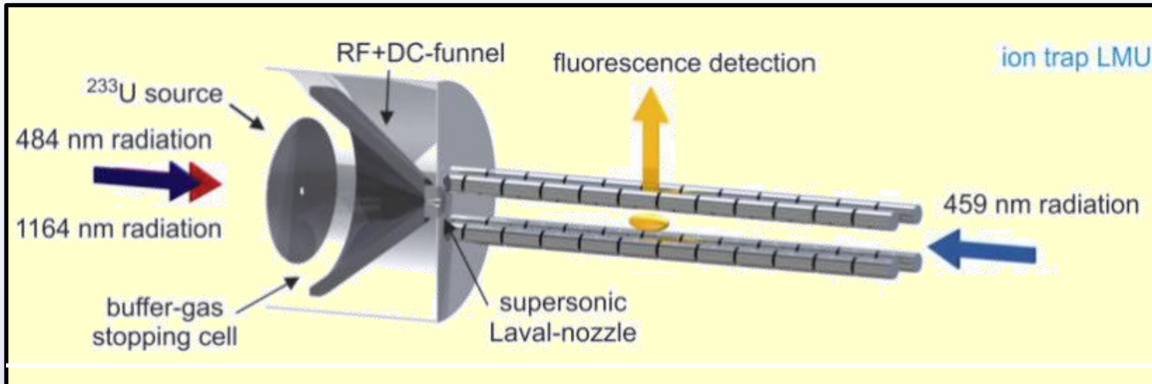




- laser excitation of $^{229(m)}Th^{2+}$ ions behind QMS:
  - → 3 external-cavity diode lasers
  - → co- and counter-propagating laser beams

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

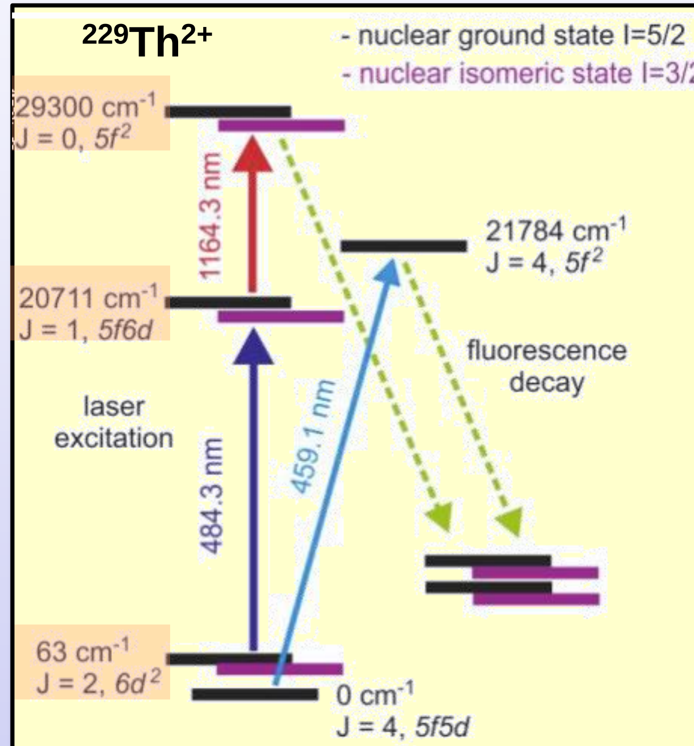
Colljnear Laser Spectroscopy on Thorium Isomer



2-photon laser excitation (J=2 → 1 → 0):

i) 484.3 nm: excitation of ions from thermal distribution into intermediate state
- 35 steps across frequency profile

ii) 1164.3 nm: excitation from intermediate state with variable excitation into final state
- for each step of i): continuous frequency scan

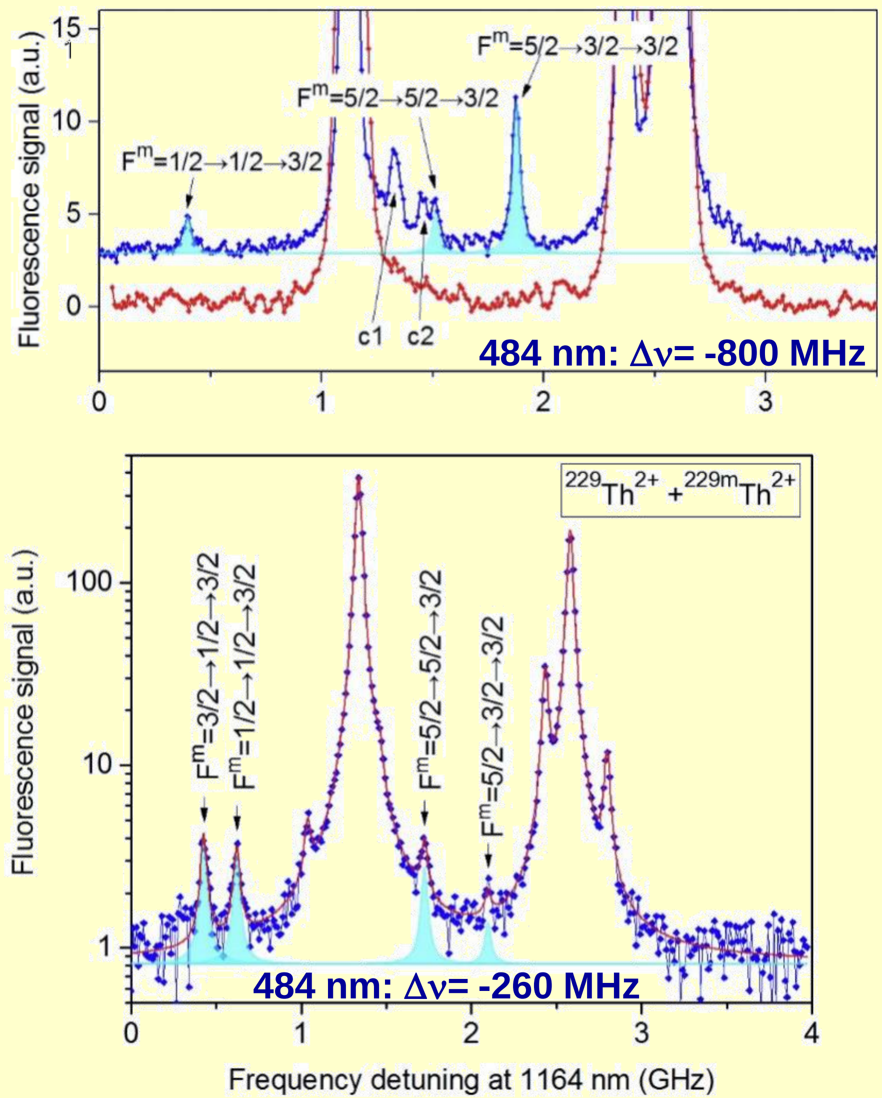


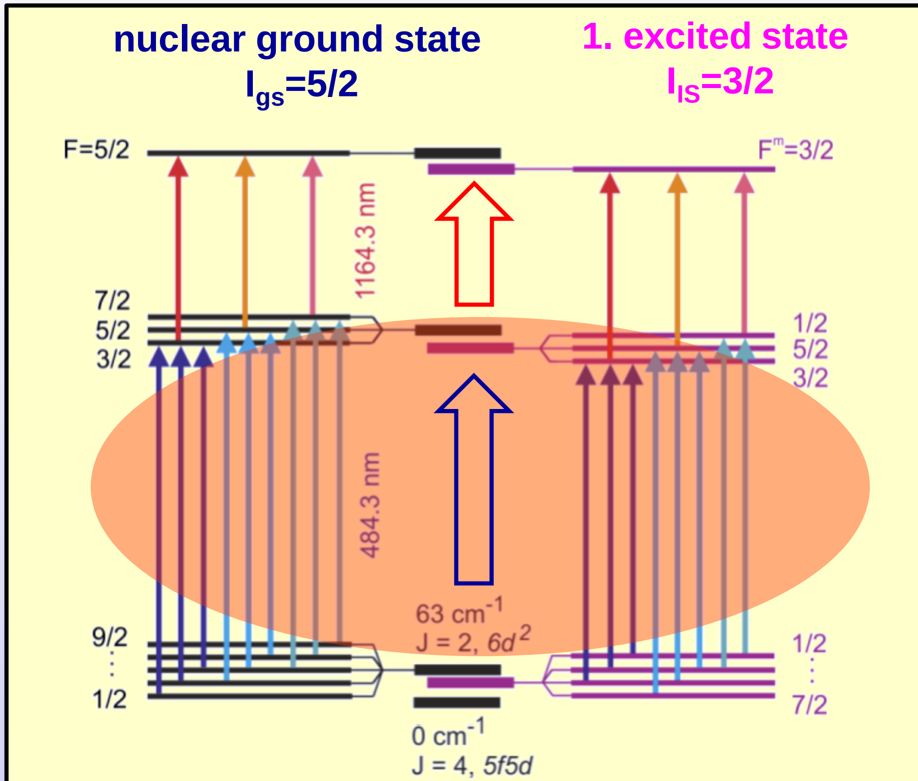
→ sensitive detection of deexcitation photons (fluorescence)
3. laser beam for normalization

Hyperfine Structure of ^{229m}Th

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

2 examples from ca. 70 spectra:



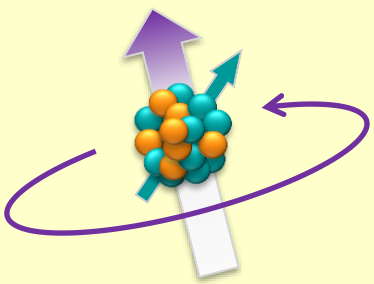


ground state: (I=5/2): 9 transitions

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

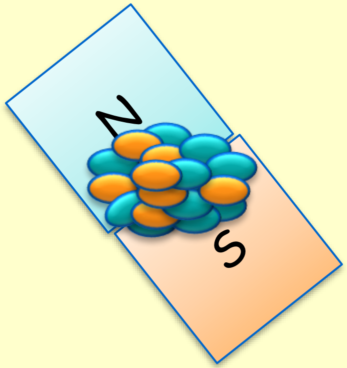
$$E_{HFS}(JIF) = \frac{1}{2} A K + B \frac{(3/4)K(K+1) - I(I+1)J(J+1)}{2I(2I-1)J(2J-1)}$$

Insights from HFS of ^{229m}Th

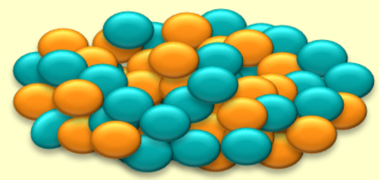


$F = J + I$

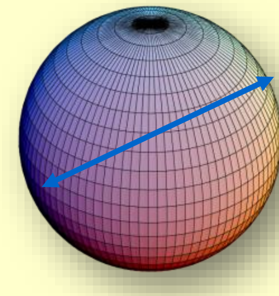
nuclear spin



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

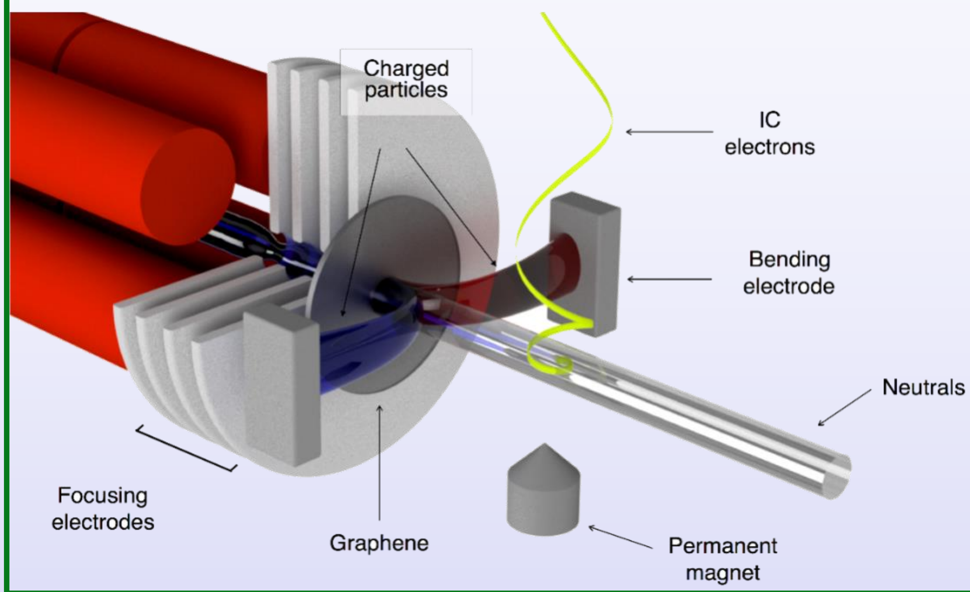
prolate deform.; $\dot{\alpha}$ sensitivity

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

sensitivity for $\dot{\alpha}$

HFS: important for detection (tagging) of isomer excitation

Excitation Energy Measurement

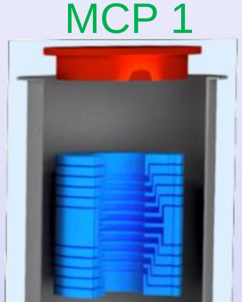


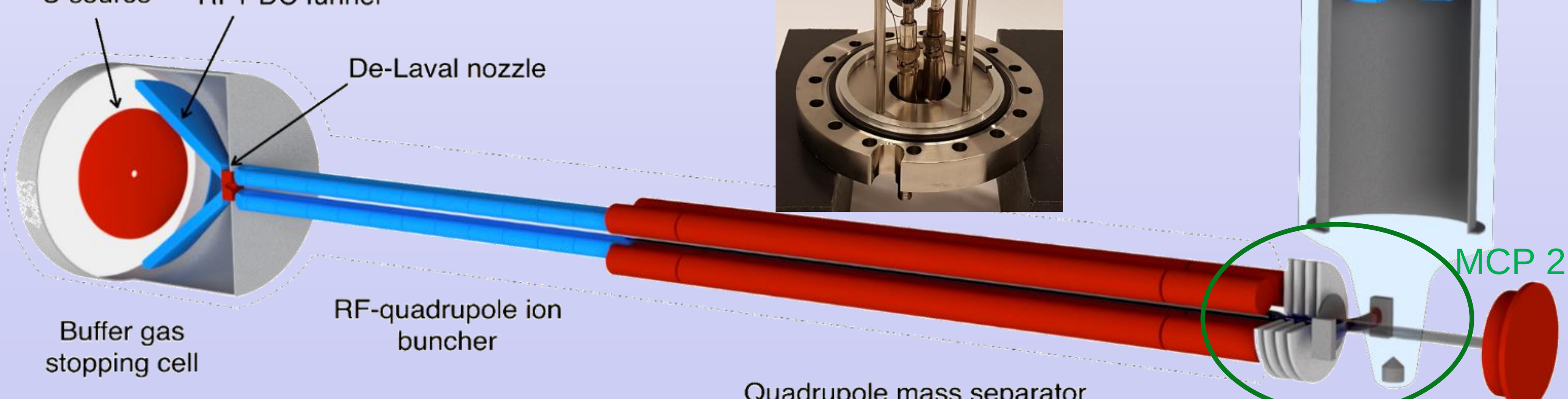
neutralization of $^{229m}Th^{q+}$ in graphene foil:

→ contact-free IC decay

→ measure E_{kin} (e)

→ spectrometer resolution: 30 -50 meV

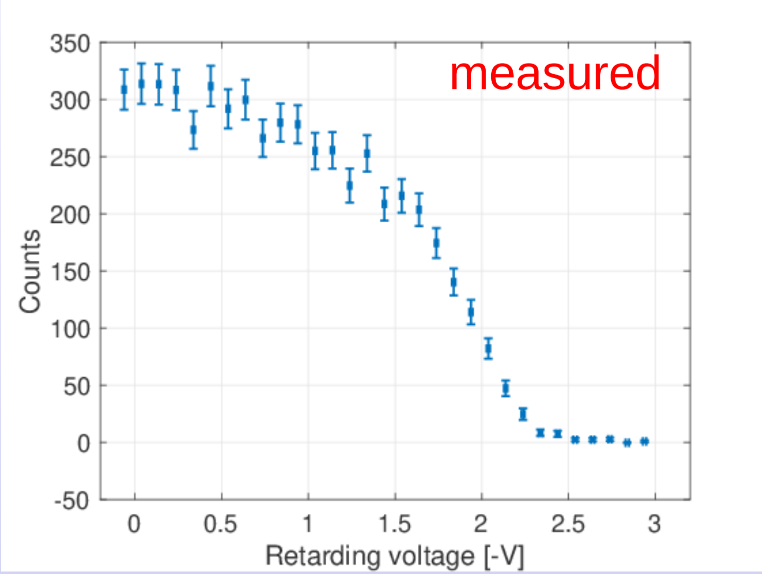


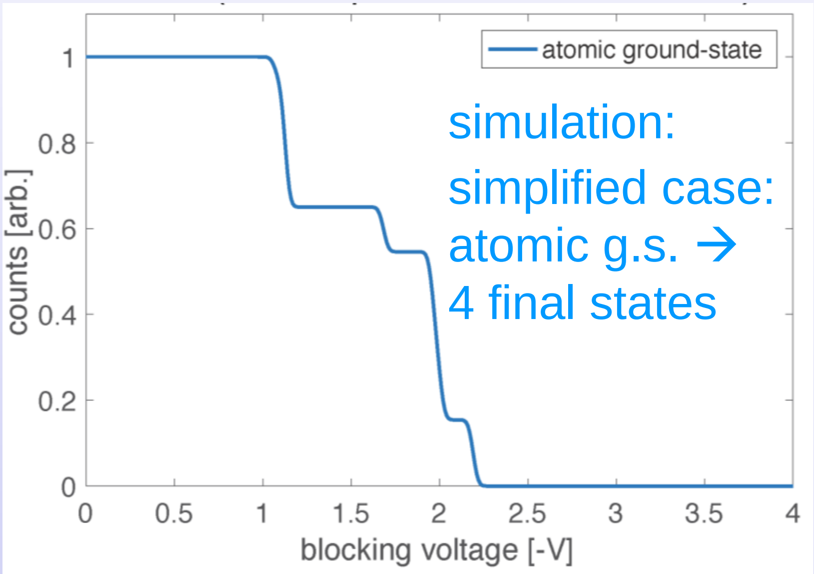


Excitation Energy: Analysis

- **Experimental challenge:**
  - resonant neutralization of $^{229m}Th^{q+}$ ends in excited atomic state and IC decay leads to excited electronic states

$E_{kin}(e) = E^*(iso) - IP - E_{ion,final} + E_{atom,initial}$ (IP(Th) = 6.308(3) eV)





- IC transitions from ≤ 4 excited atomic states could be resolved
- measurement: no steps clearly identified: ≥ 5 initial states must contribute
- 82 states can contribute in relevant energy range (below 20000 cm^{-1}, ≈ 2.5 eV)
- individual population unknown

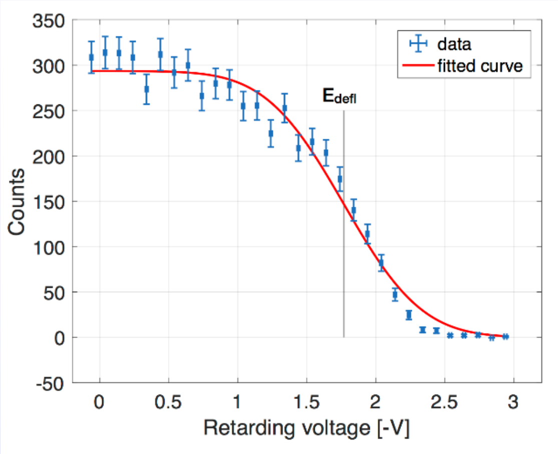
atomic calculations:
P. Bilous, A. Palffy (MPIK Heidelberg)
F. Libisch, C. Lemell (TU Wien)

Excitation Energy: Analysis

- **fit error function to measured data:** $f(u) = a\,(1 - \mathrm{erf}\,[\,(U - E_{defl}) / b\,])$
  → deflection point E_{defl} **= 1.77(3) eV**

  → $E^*(iso) = E_{defl} + E_0$

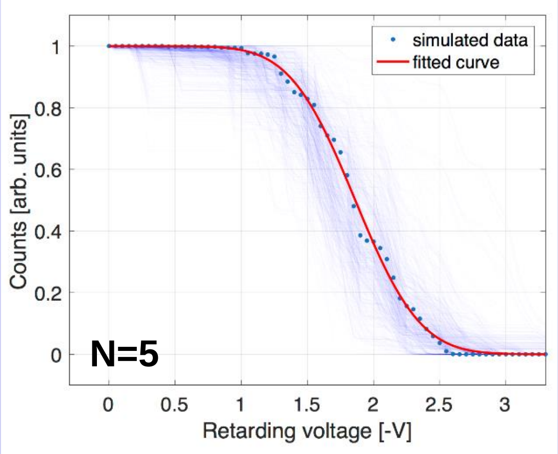
  → predict E_0 from simulated spectra

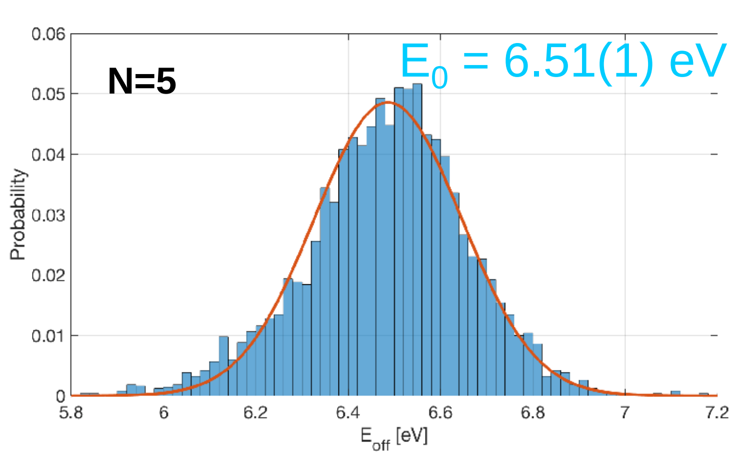


→ create simulated data from combinations of (N=5) initial atomic states:

Expected IC electron energy spectra

20000 population distributions: any 5 (of 82) E_i to all possible final E_f

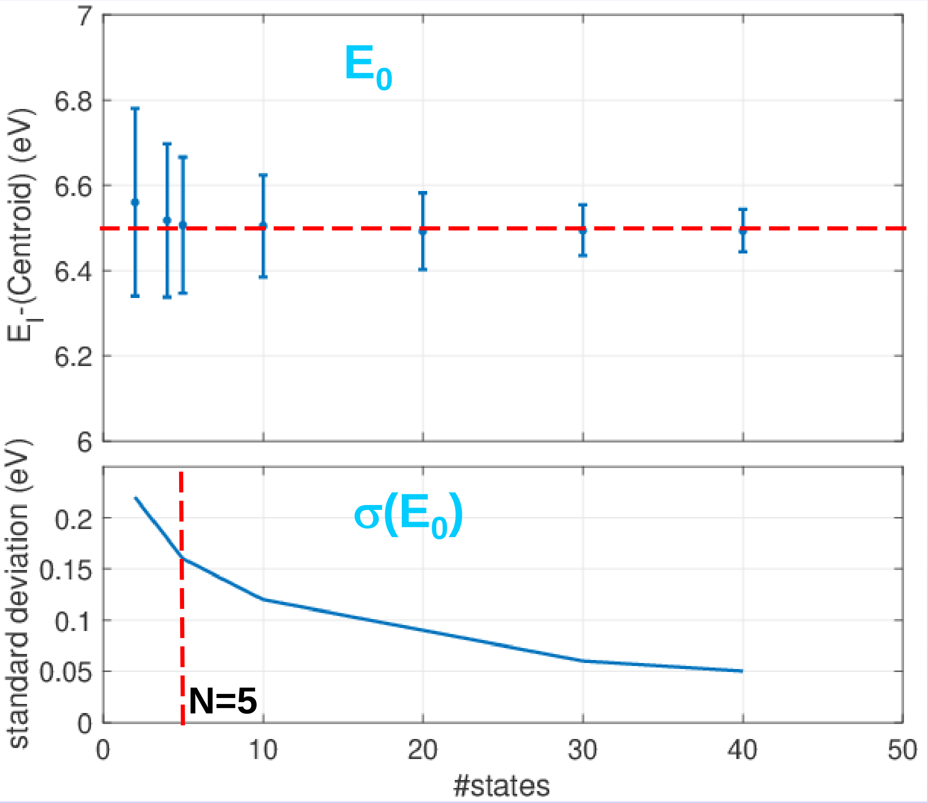




Excitation Energy: Analysis

- **Findings from simulated spectra:**

robust position of E_0 → E_0 = 6.51(1) eV



larger N : smaller uncertainty of E_0

→ N=5: conservative estimate of experimental uncertainty

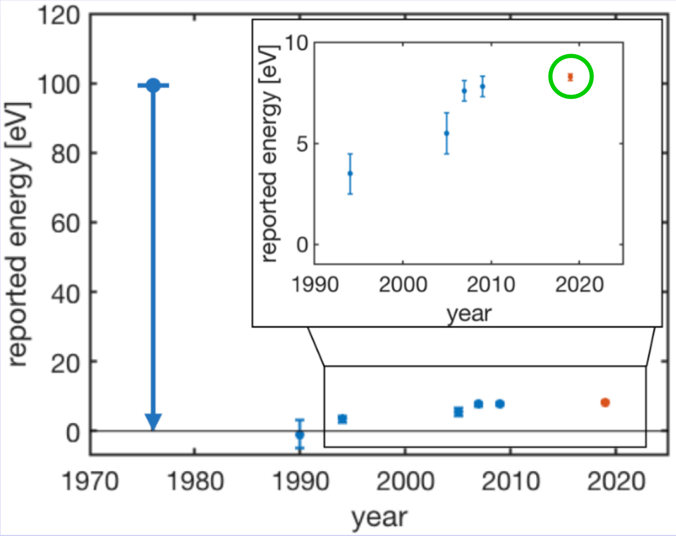
→ **E_0 = 6.51 ± 0.16 eV**

- **First direct measurement:** **E*(iso) = 8.28 ± 0.17 eV (= 149.7 ± 3.1 nm)**

B. Seiferle, PT et al., Nature 575 (2019)

Achievements in "Search & Characterization Phase"

- **Existence of ^{229m}Th: first direct detection via IC decay** — Nature 533 (2016)
- **Half-life of neutral ^{229m}Th: $t_{1/2}$ = 7 μs → $\alpha_{IC} \sim 10^9$** — PRL 118 (2017)
- **Hyperfine structure of ^{229m}Th**
  → via collinear laser spectroscopy — Nature 556 (2018)
  → nuclear moments, charge radius
- **isomeric excitation energy:** — method: EPJ A53 (2017)
  → via retarding field magnetic bottle electron spectrometer
  first direct measurement: Nature 575 (2019)



E* = 8.28 ± 0.17 eV

λ = 149.7 ± 3.1 nm

→ clarifies regime of laser technology for optical control (excludes laser crystal approaches)

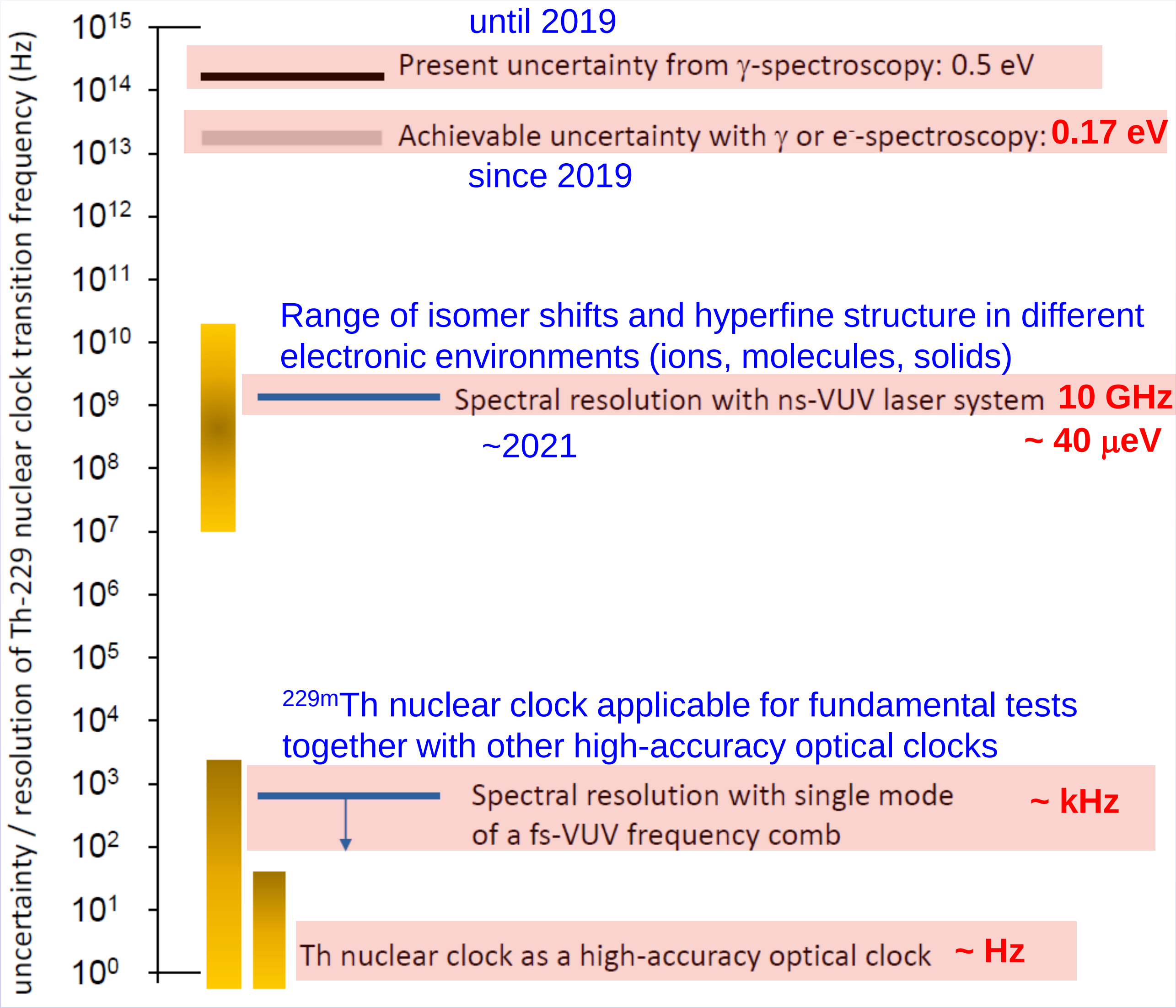
The long way towards the Nuclear Clock

- **still to bridge: 14 orders of magnitude:**

already feasible with existing laser technology

concept:

L. v.d. Wense, PT et al, PRL 119 (2017)



Laser excitation scheme for ^{229m}Th

- **Paradigm:**

  - direct laser excitation of ^{229m}Th needs improved knowledge on E* and dedicated laser
  - since: i) 8.28(17) eV requires at least 0.34 eV to be scanned
    ii) long radiative isomeric lifetime (hours) → long detection times

- **But:**

  - probing the laser excitation by exploiting the (fast) Internal Conversion decay channel (τ~10 μs) allows for using existing (VUV) laser technology

  → direct nuclear laser spectroscopy by optical excitation of ^{229m}Th is in reach

- **Experimental approach:**

  - trigger the decay electron detection with the laser pulse
  - achieve a high signal-to-background ratio

  → corresponding experiment is in preparation
  (in collaboration with UCLA (USA) & Univ./Laserzentrum Hannover)

L. v.d. Wense et al., PRL 119, 132503 (2017)

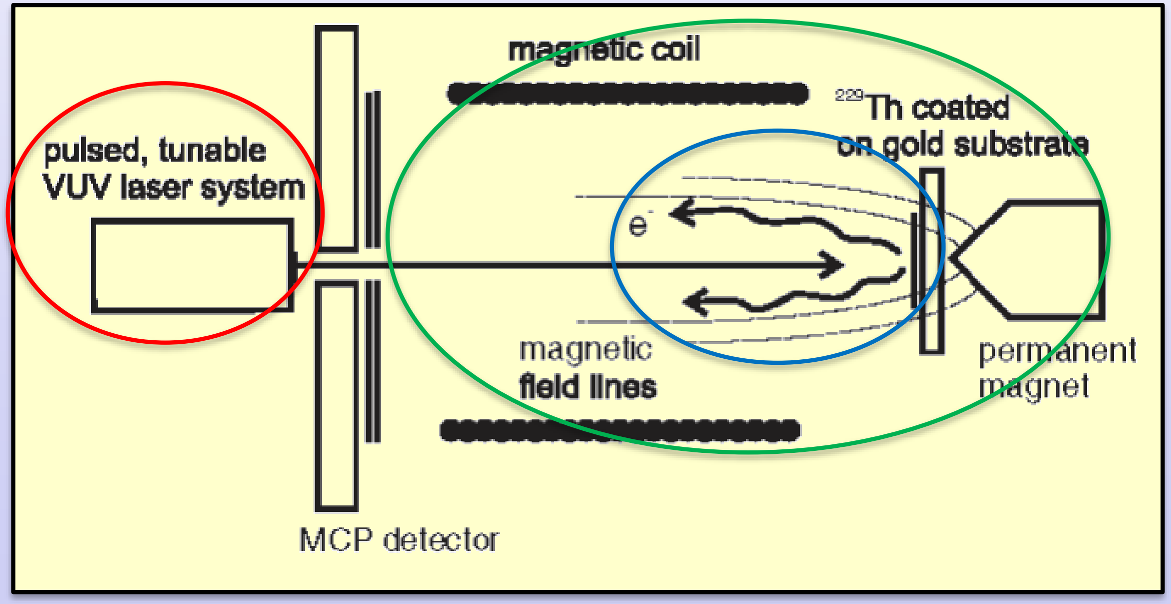
Laser excitation scheme for ^{229m}Th

- **Proposed experimental setup and procedure:**

assumed (tunable and pulsed VUV) laser source*:

pulse energy: E_L = 10 μJ @ ca. 160 nm
bandwidth: $\Delta\nu_L$ = 10 GHz
pulse length: T_L = 5 ns
repetition rate R_L = 10 Hz

^{229}Th layer:
thickness 2.5 nm, area 1 mm^2
→ ca. 4200 ^{229}Th excited/pulse,
total electron detection efficiency at MCP ~ 12.5%



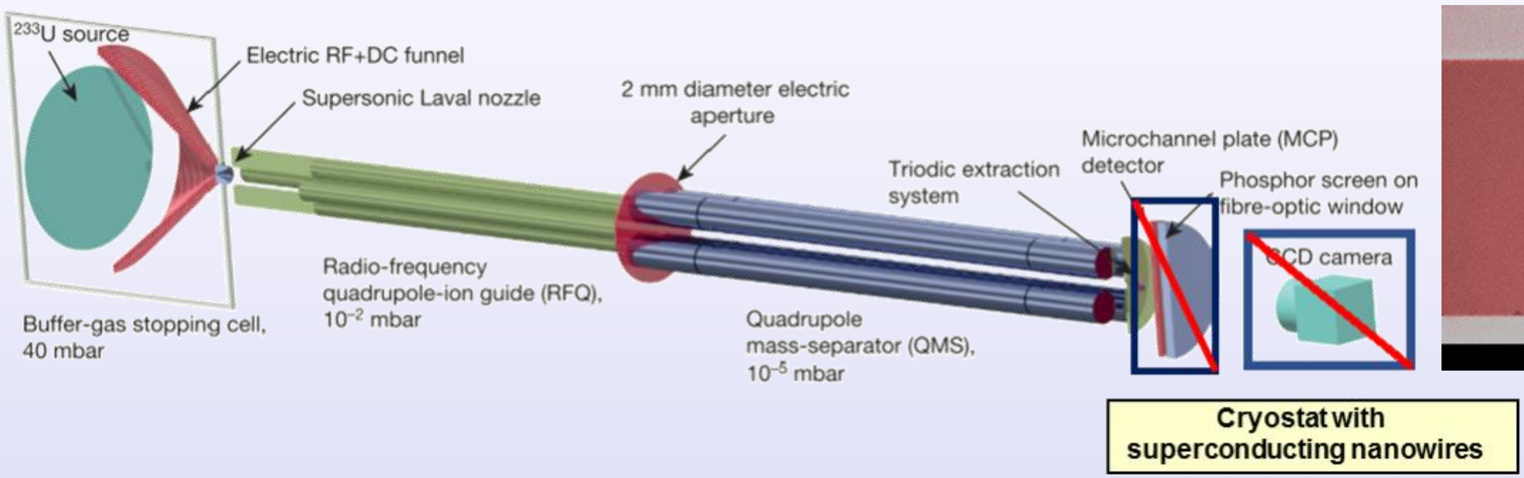
IC electron detection:
- ^{229}Th coated sample in magnetic bottle (could be curved)
- detection time: 10 μs

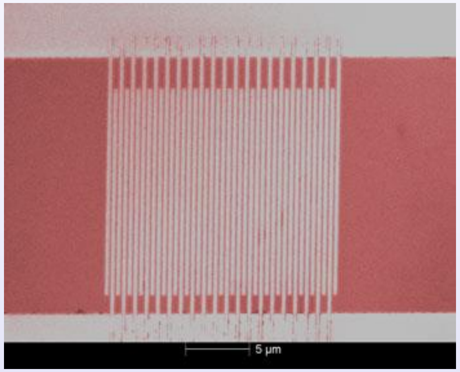
High signal-background:
S/B ~ 7 x 10^4
Short scan time:
< 3 days for 1 eV

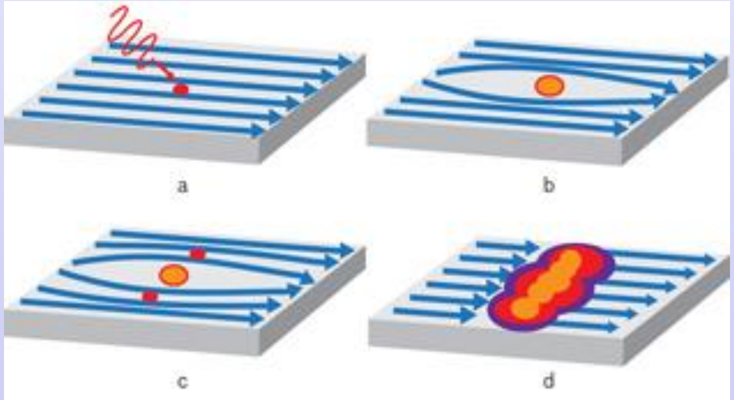
* S.J. Hanna et al., Int. J. Mass Spectr. 279, 134 (2009)

E^{iso}: Complementary approach

- **Superconducting Single Photon Nanowire Detectors (SNSPDs):**







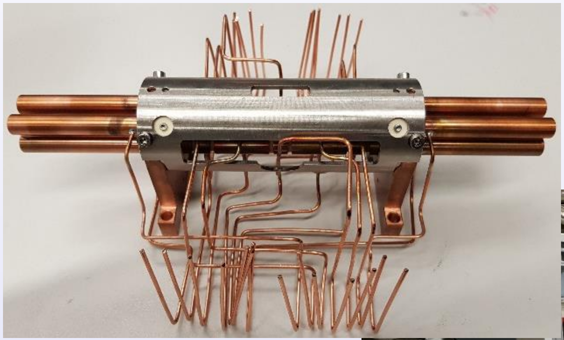
- **SNSPD:** meander-shaped sc wire with bias current
- implant ^{229m}Th on the sc nanowire
- deposited decay energy breaks superconductivity
- measure current
- decay energy spectrum via scanning of bias current
- expected resolution ~ 0.1 eV

measurements are ongoing with first promising results

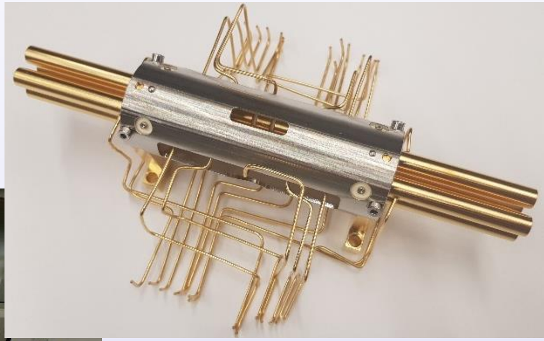
collaboration with UCLA, NIST/Boulder, TU Wien

Ionic Lifetime Measurement

needs longer storage time (= better vacuum)

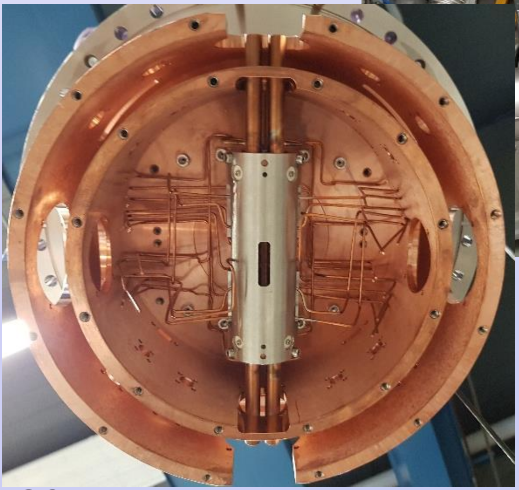


- setup of a cryogenic Paul trap
- platform for laser manipulation



Cu basis

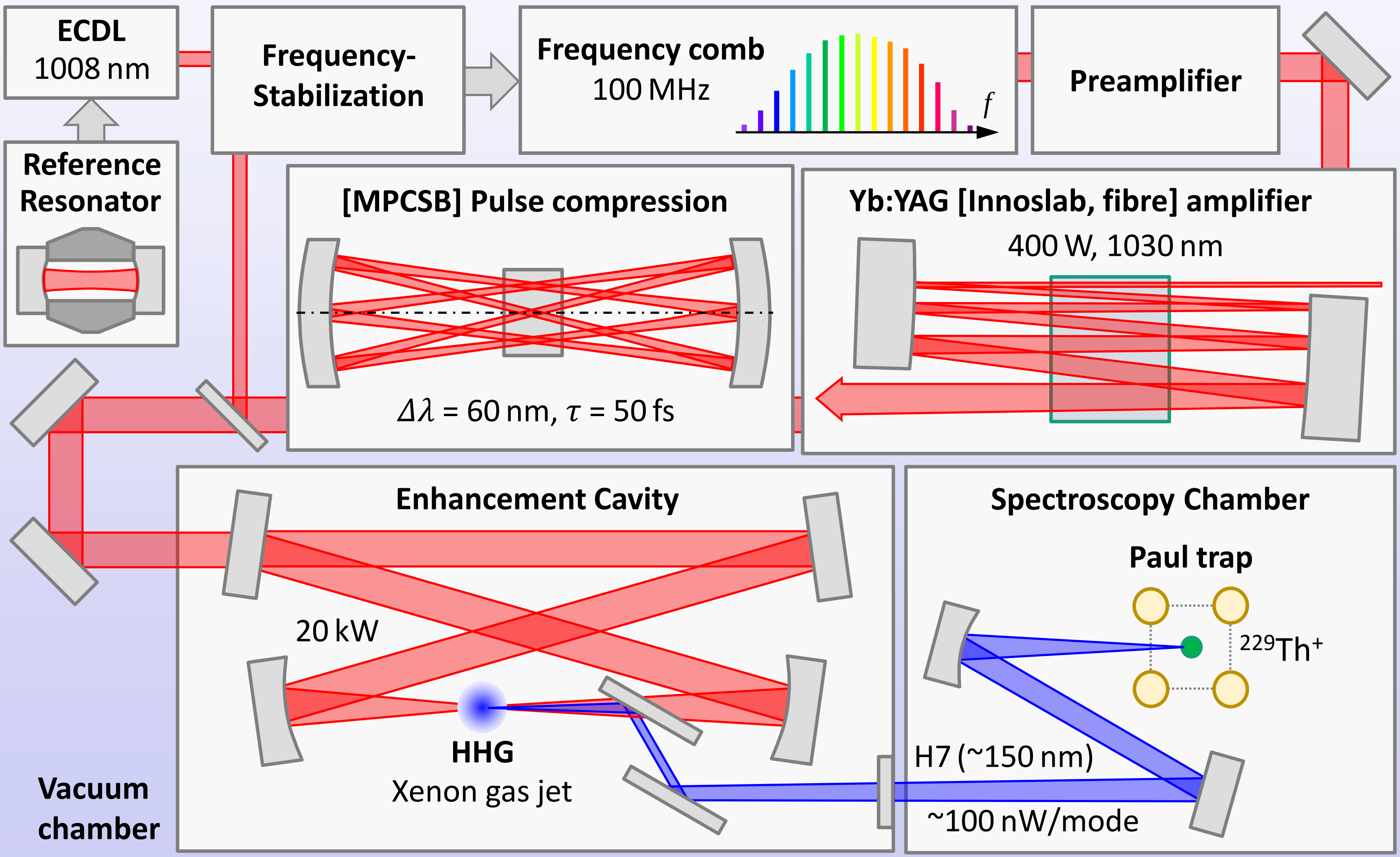
Au plated



- ready for commissioning
- sympathetic laser cooling with $^{88}Sr^{+}$ set up and ready

Concept for a VUV laser source for the ^{229m}Thorium - Nuclear Clock transition

7th harmonic of VUV frequency comb:



J. Weitenberg, ILT Fraunhofer/ RWTH Aachen & MPQ Garching

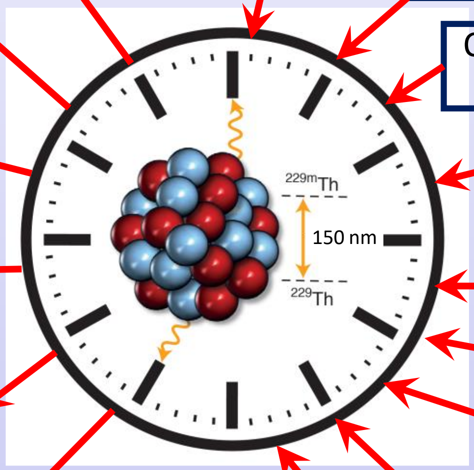
New ERC-Synergy Project: ThoriumNuclearClock

Scientific advances in many fields

- Precision metrology
- Nuclear physics
- Atomic physics
- Laser physics
- Solid-state physics
- Particle physics

Projects goals

- Perform the first laser excitation of a nuclear transition
- Precisely determine the 229Th nuclear structure parameters
- Quantify the sensitivity of the nuclear clock to variation of fundamental constants
- Improve the precision of the nuclear transition frequency by 9 orders of magnitude
- Development of a Hz-level linewidth VUV laser
- Demonstrate nuclear clock with Th3+ ions
- Demonstrate completely new solid-state clock scheme
- Test fundamental concepts of physics
- Search for dark matter



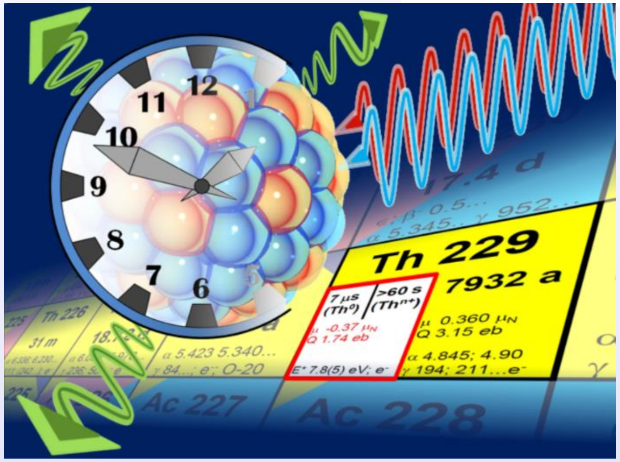
Build three nuclear clocks

https://thoriumclock.eu

Summary

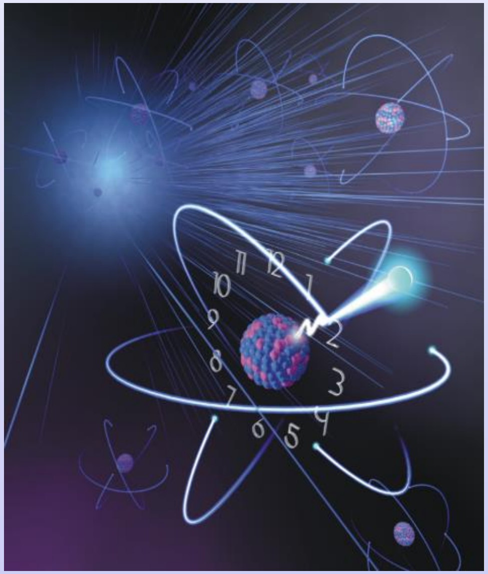
look back: huge progress in last 4 years:

- identification & characterization of the thorium isomer



look ahead: ongoing consolidation & next steps

- excitation energy from complementary techniques
- cryogenic Paul trap, sympathetic (Sr^+) laser cooling
- ^{229m}Th ionic lifetime
- determine sensitivity enhancement for $\dot{\alpha}$
- doped-crystal approach: radiative, IC branches
- laser spectroscopy: resonance search



ambitious, exciting, important research topic:

- excite for the first time ever the nuclear transition by laser
- build clocks based on completely new principles
- ability to drastically improve sensitivity to new physics
- ability to search for dark matter candidates not accessible by any other means

the door is open for the realization of a nuclear clock …

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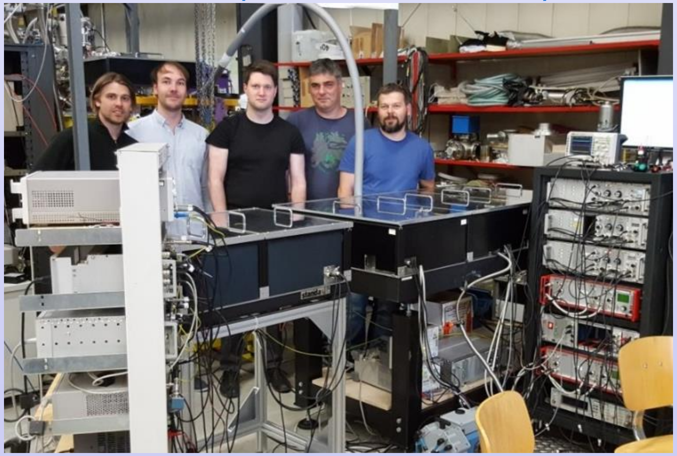
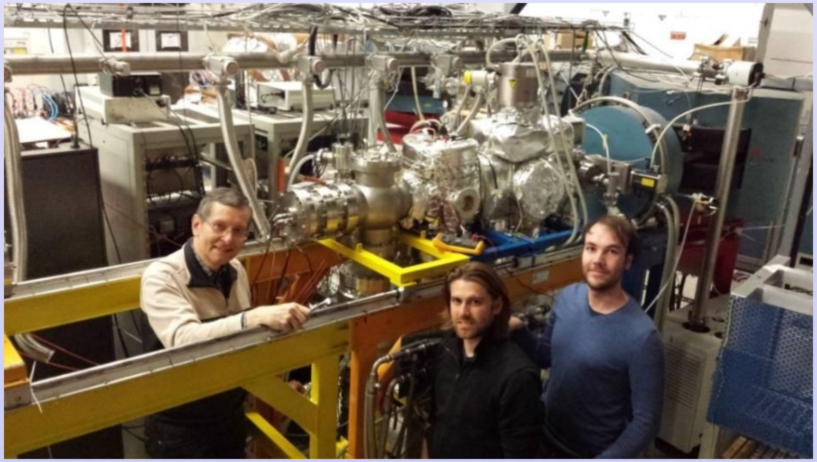
DFG Deutsche Forschungsgemeinschaft





Thank you for your attention !