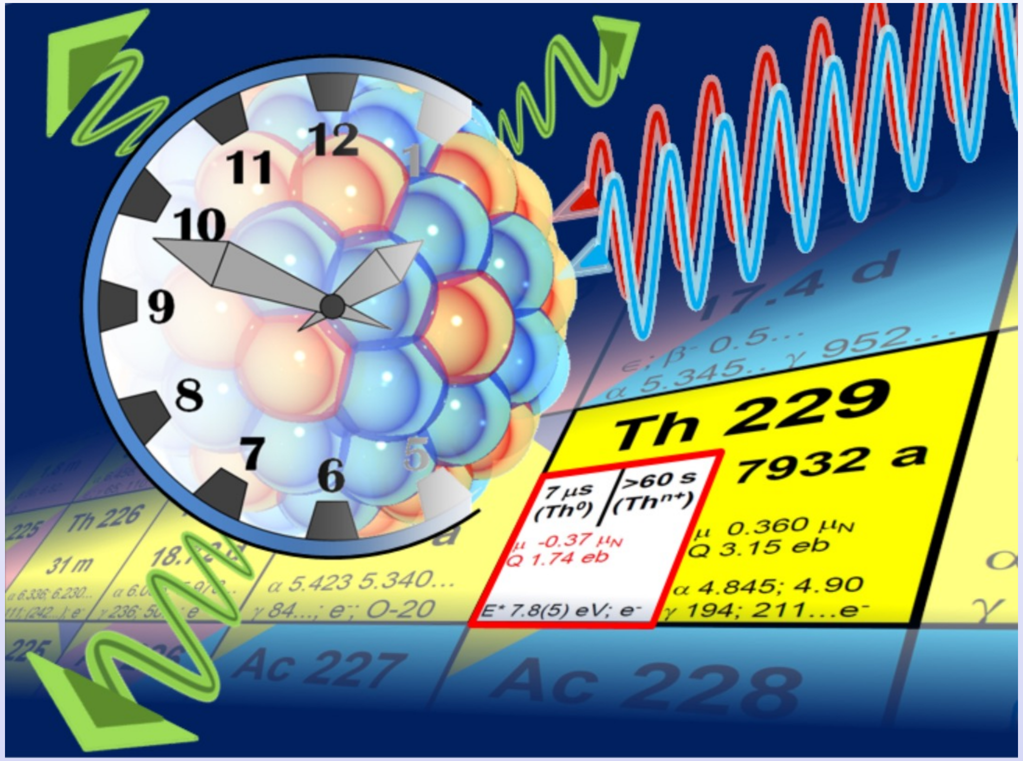
Unveiling the Thorium Isomer ^{229m}Th: Towards a Nuclear Clock as novel Quantum Sensor

Peter G. Thirolf, LMU München

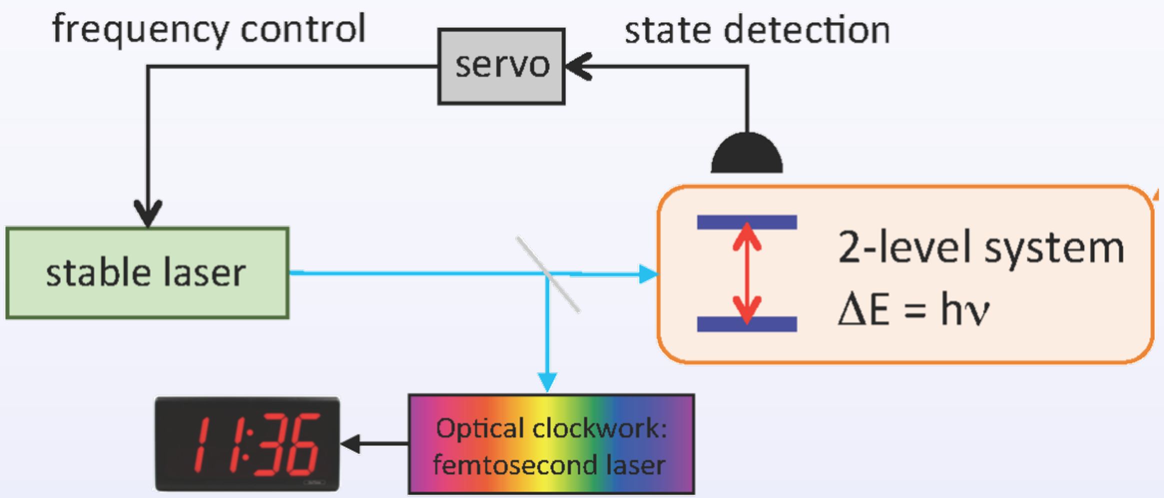
- Intro: Thorium Nuclear Clock & Applications
- Knowledge on ^{229m}Th
  - experimental approach & setup
  - IC decay, $t_{1/2}$, HFS, E*
  - first observation of radiative decay
- Perspectives

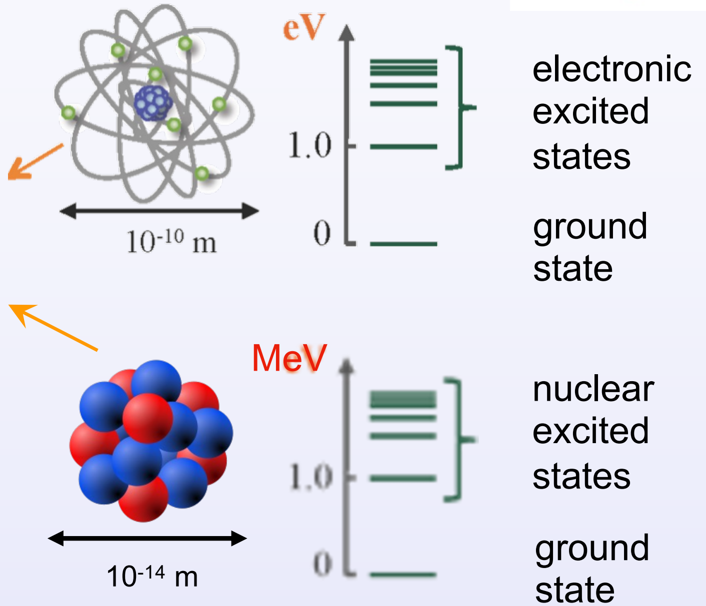


Thorium Nuclear Clock

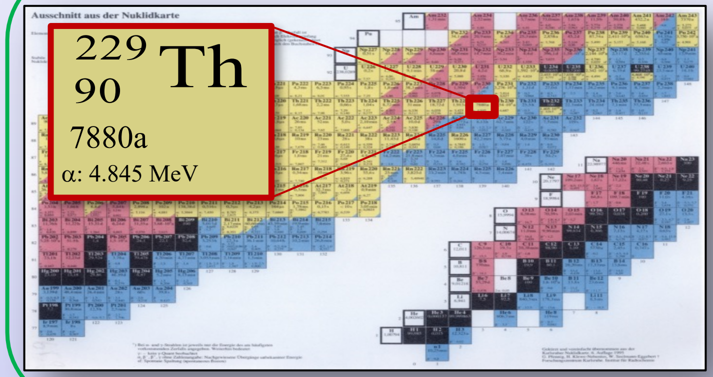
scheme of an **atomic** clock

scheme of a **nuclear** clock





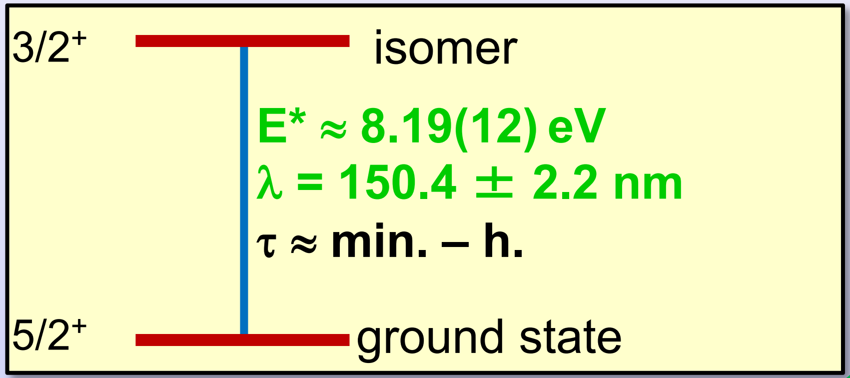
Nuclear clock proposal: E. Peik and Chr. Tamm, Europhys. Lett. 61, 181-186 (2003)
10^{-19} performance estimate of ^{229}Th ion clock: C. J. Campbell, et al., PRL 108, 120802 (2012)



^{229m}Th properties:

lowest E* of all ~186000 presently known nuclear excited states

$\Delta E/E \sim 10^{-20}$
~ 0.1 mHz nat. linewidth



Applications of Nuclear Clocks

- **Beyond Timekeeping: Quantum Sensor due to different operation principle compared to atomic clocks:**
  - **Coulomb + weak + <u>strong</u> interaction** contribute to clock frequency
  - **small nuclear moments:** less sensitivity to perturbations by external fields
  - **sensitivity** to new physics searches: **enhanced by 10^4-10^6** compared to present clocks

M.S. Safronova et al., Rev. Mod. Phys 90, 025008 (2018)

→ unique opportunity for new physics discoveries which cannot be accomplished with any other technology:

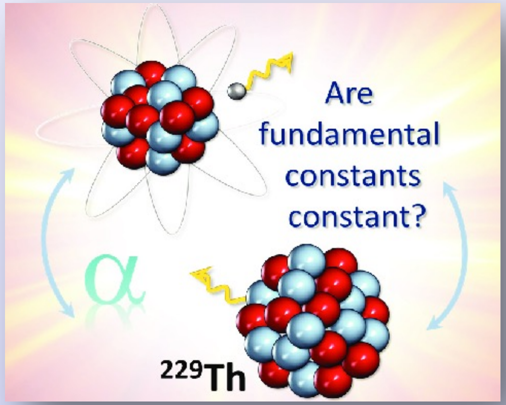
E. Peik, PT et al., Quant. Sci. Tech. 6, 034002 (2021)

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

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

J.P. Uzan, Living Rev. Relativ. 14, 2 (2011)

R. Lange et al., PRL 126, 011102 (2021)



- enhanced sensitivity by (10^5 – 10^6) of ^{229m}Th expected

V.V. Flambaum, PRL 97, 092502 (2006)

- measurements involve monitoring the ratio of nuclear/atomic clock over time

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

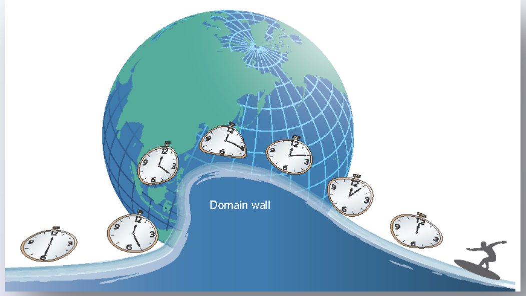
Applications of Nuclear Clocks

- **Test coupling of fundamental constants on changing gravitational potential**
  tests the local position invariance hypothesis and thus Einstein's Equivalence Principle

V. V. Flambaum,
PRL 117, 072501 (2016)

- **Search for Dark Matter**
  - *ultralight scalar fields:* searches for oscillatory variation of fundamental constants

Arvanitaki et al., PRD 91, 015015 (2015), Van Tilburg et al., PRL 115, 011802 (2015), Hees et al., PRL 117, 061301 (2016)



  - *topological dark matter*: monopoles, 1D strings, 2D 'domain walls'
  use networks of ultra-precise synchronized clocks

Derevianko & Pospelov,
Nat. Phys. 10, 933 (2014)

- **Improved precision of satellite-based navigation**
  (GPS, Galileo..): m → cm (mm ?)
  - autonomous driving
  - freight-/ component tracking …

- **3D gravity sensor:** 'relativistic geodesy'
  - clock precision of 10^{-18}: 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

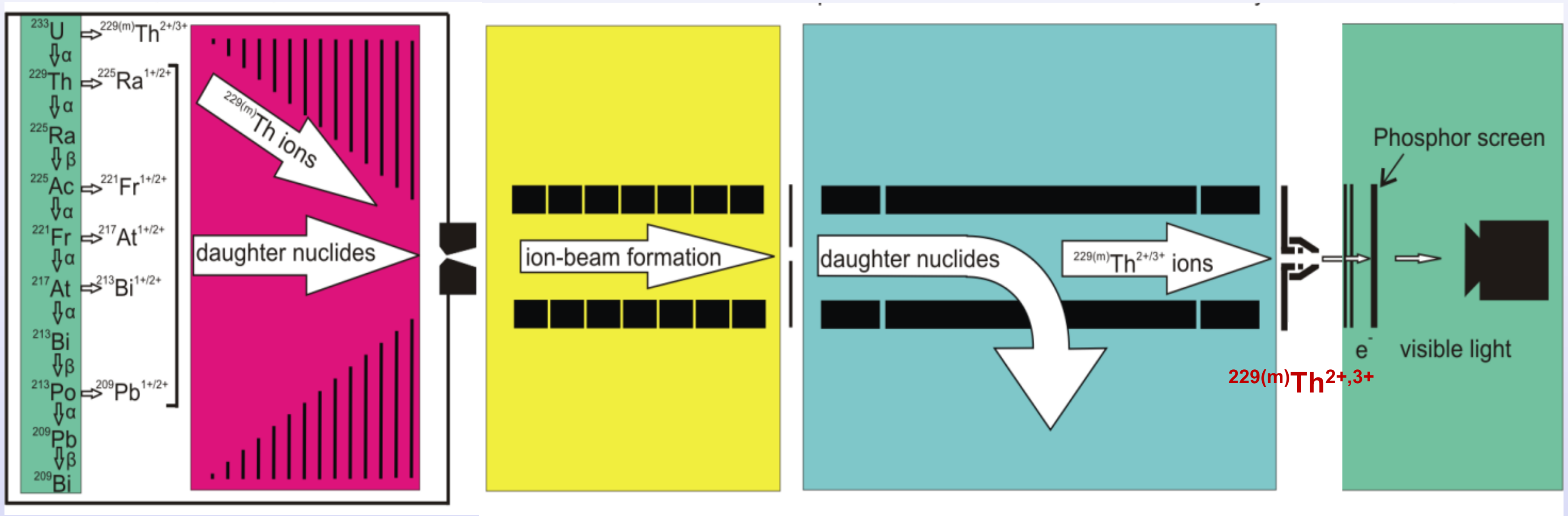
PT et al., Annalen d. Physik 531, 1800391 (2019)

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**



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

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

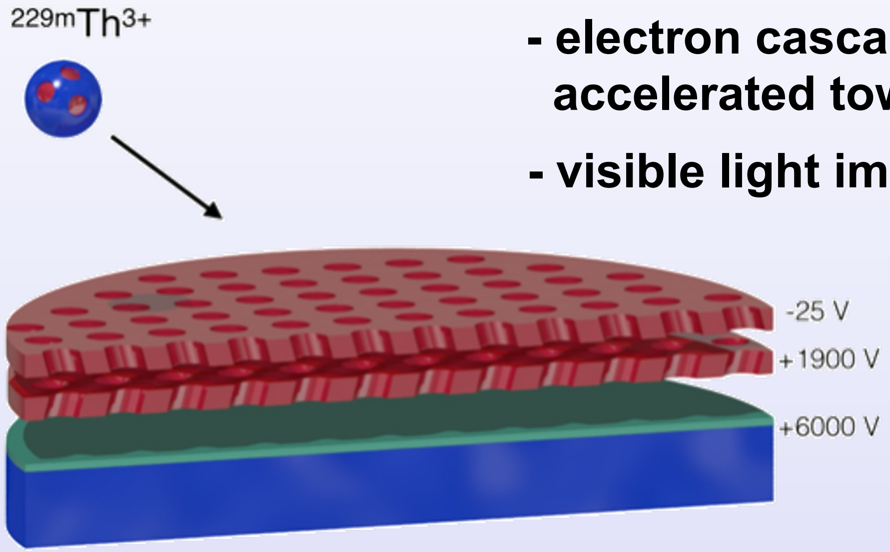
- initial fluorescence search unsuccessful

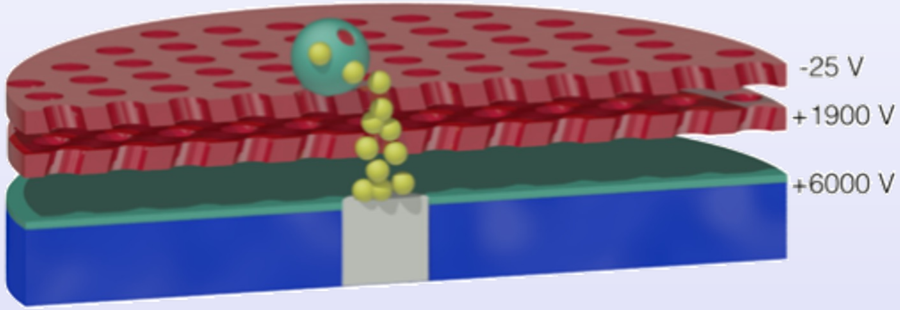
→ search in IC decay channel

→ 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





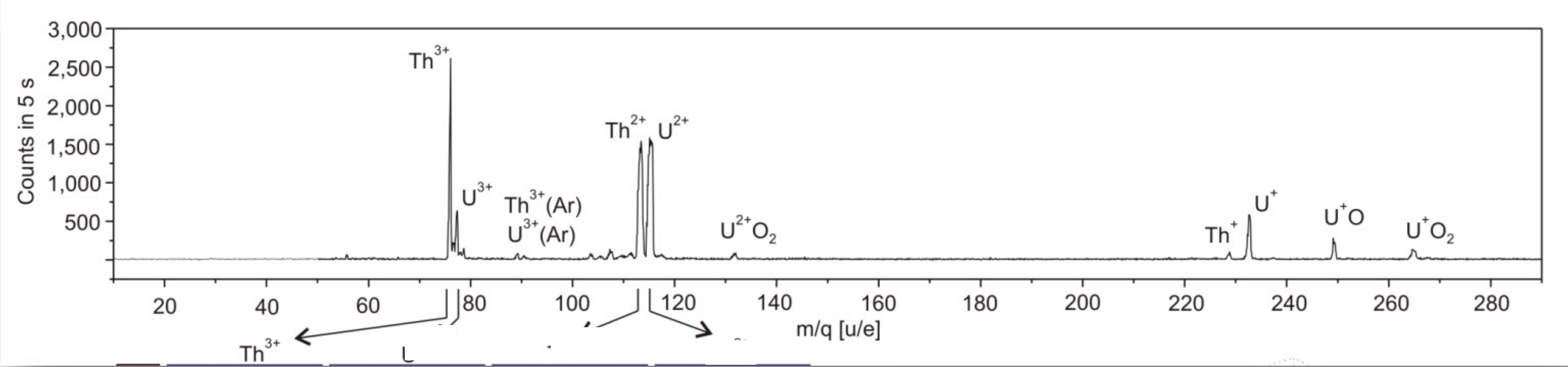
- internal conversion (IC) energetically allowed for neutral thorium:

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

- 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)

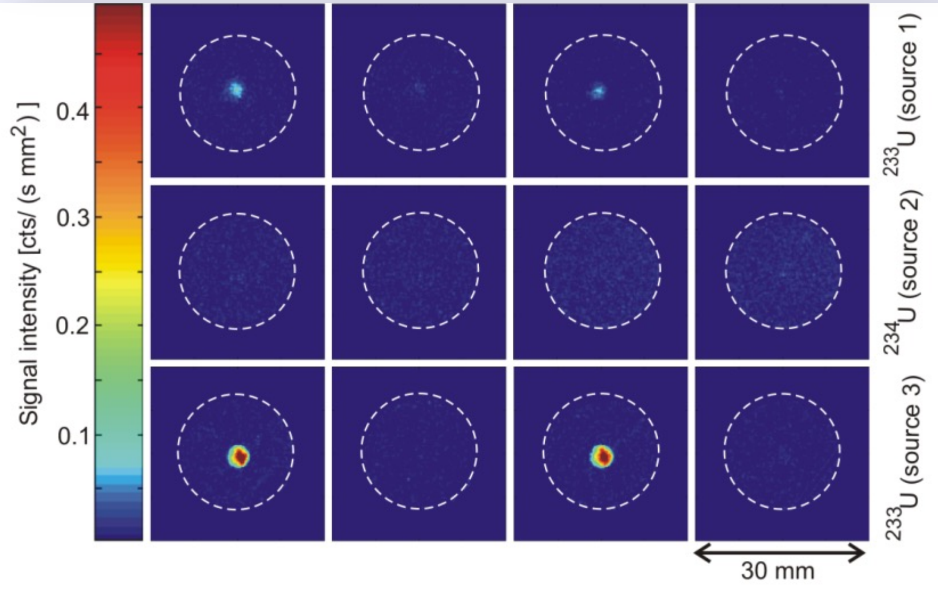


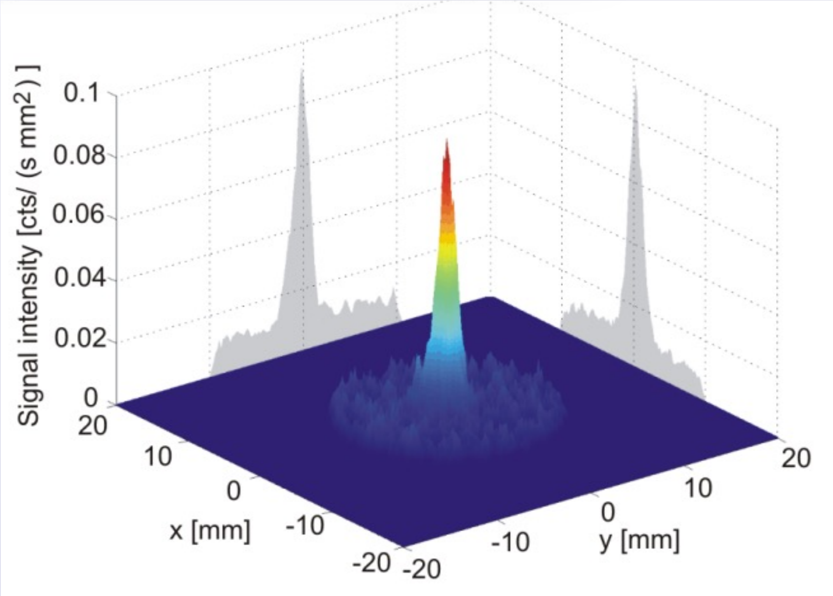
Th3+ U3+ Th2+ U2+

weak ^{233}U source

^{234} U source

strong ^{233}U source

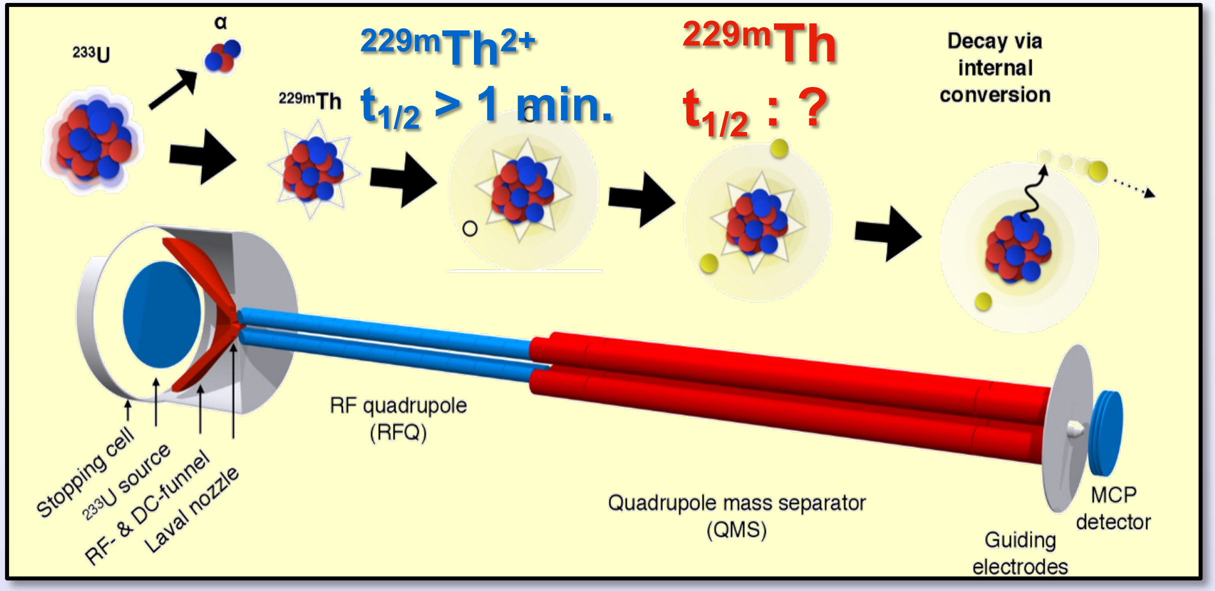


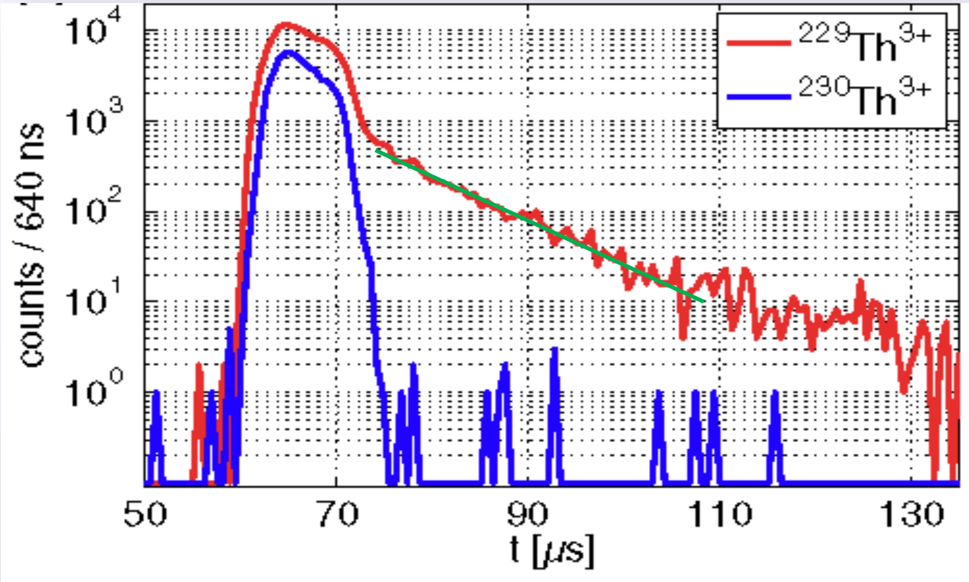


clear signal from Th^{3+}, Th^{2+} , no signal from U^{3+}, U^{2+}

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±1 μs

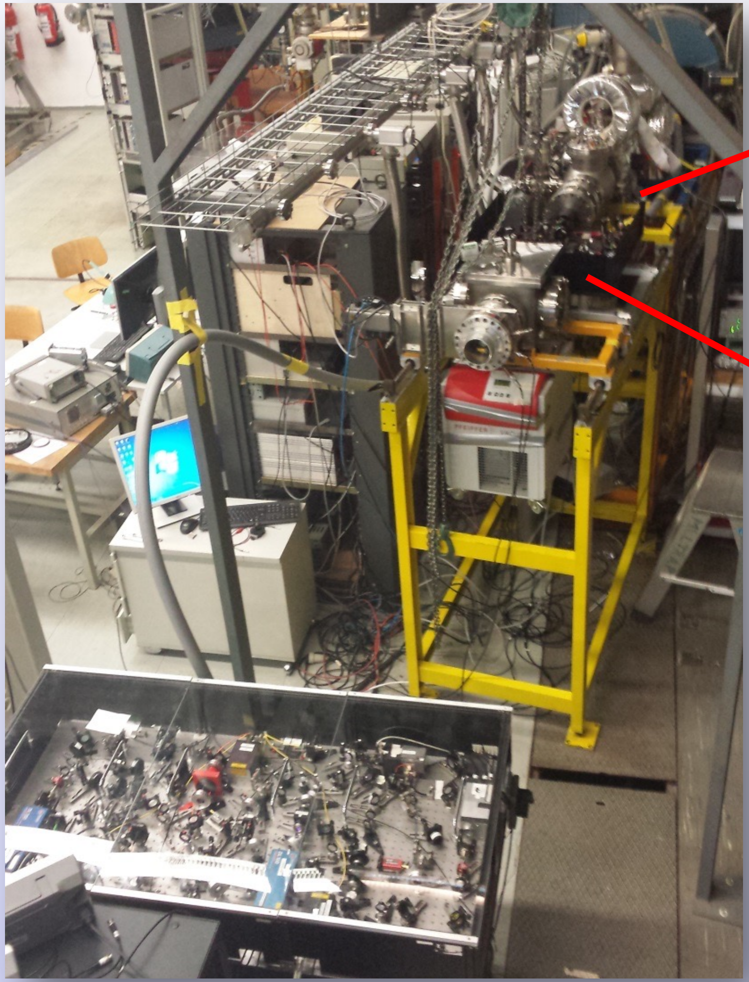
→ in agreement with expected $\alpha_{IC} = N_e/N_\gamma \sim 10^9$

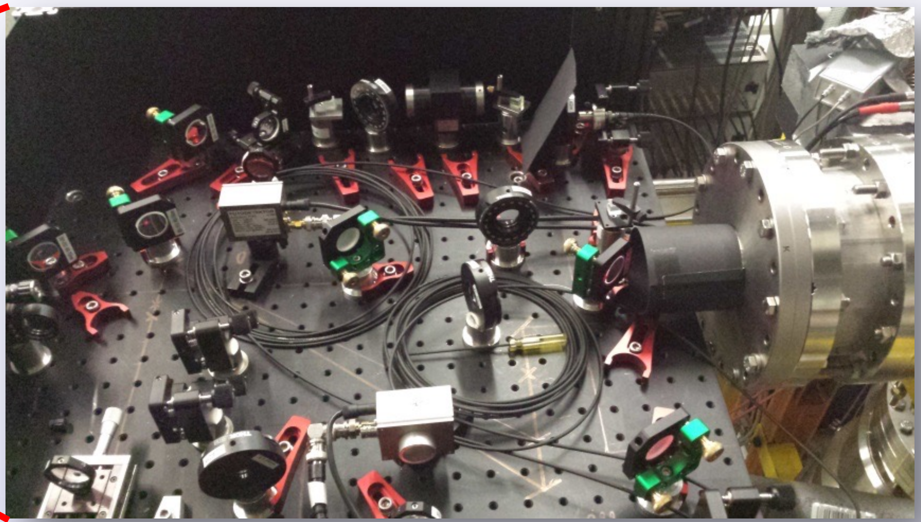
B. Seiferle, L. v.d. Wense, PT, PRL 118, 042501 (2017)

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 $^{229m}Th^{2+}$

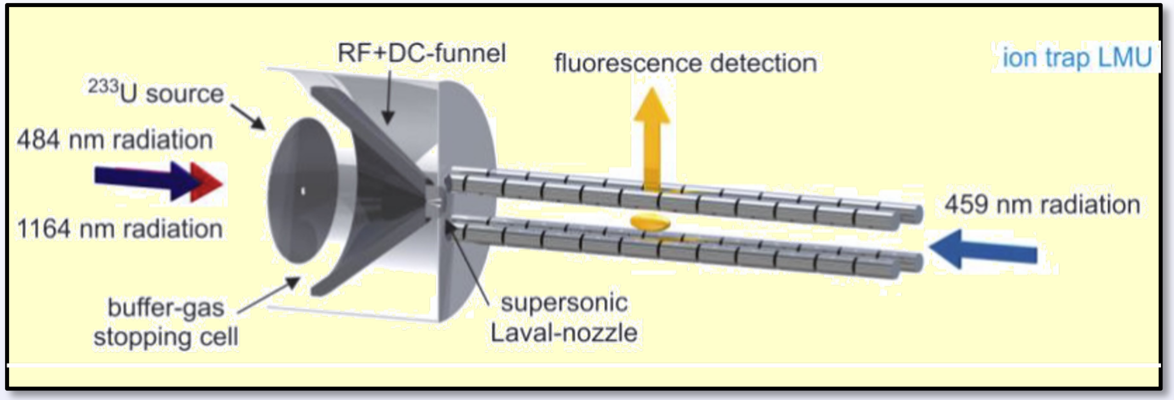




- laser excitation of $^{229(m)}Th^{2+}$ ions behind QMS:
  - → 3 external-cavity diode lasers
  - → co- and counter-propagating laser beams

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

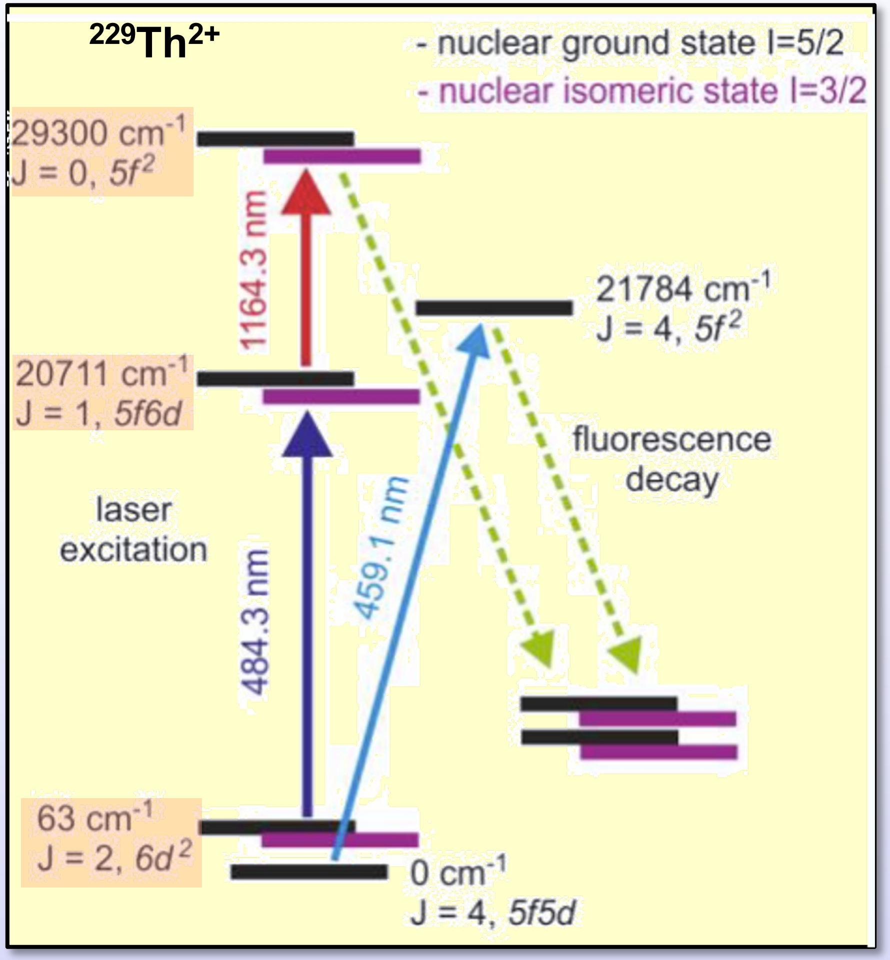
Colljnear Laser Spectroscopy on Thorium Isomer



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

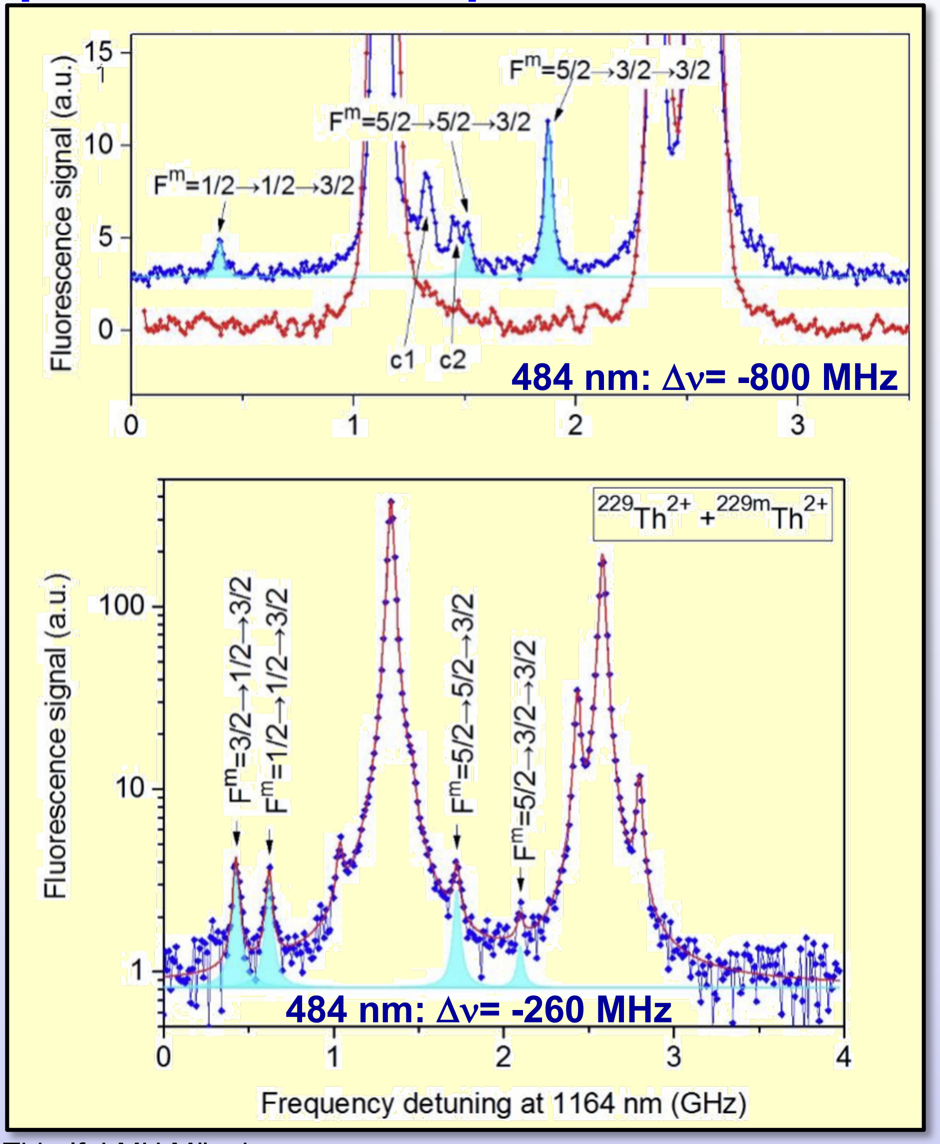


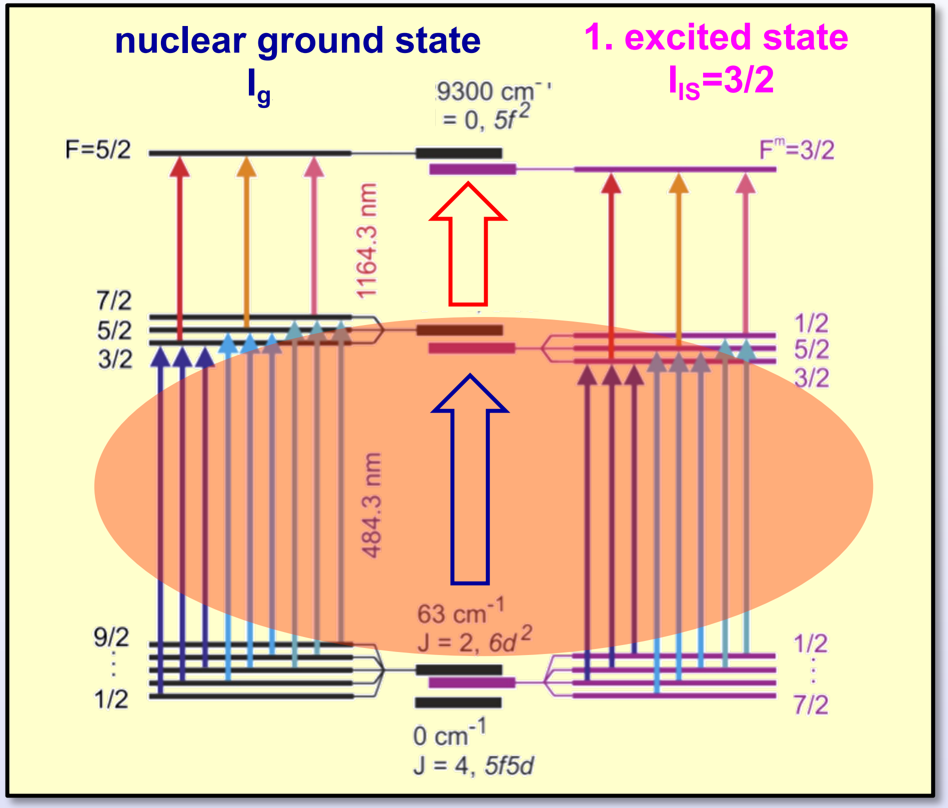
→ sensitive detection of deexcitation photons (fluorescence) , 3rd laser beam for normalization

Hyperfine Structure of ^{229m}Th

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

2 examples from ca. 70 spectra:



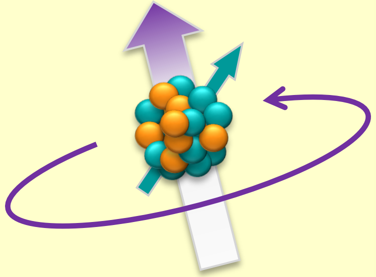


ground state: (I=5/2): 9 transitions

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

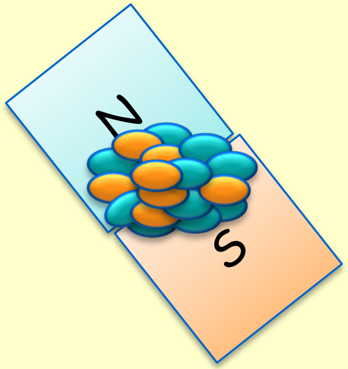
$$E_{HFS}(JIF) = \frac{1}{2} A K + B \frac{(3/4)K(K+1) - I(I+1)J(J+1)}{2I(2I-1)J(2J-1)}$$

Insights from HFS of ^{229m}Th

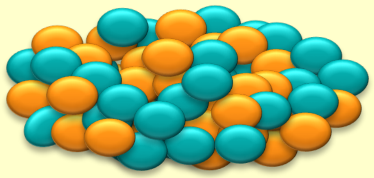


F=J+I

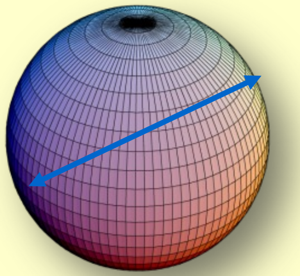
nuclear spin



magnetic
dipole moment



electrical
quadrupole moment



charge radius

I = 3/2

confirms level scheme

μ^m = - 0.37(6) μ_N

confirmed by theory

Q_0^m = 8.7(3) eb

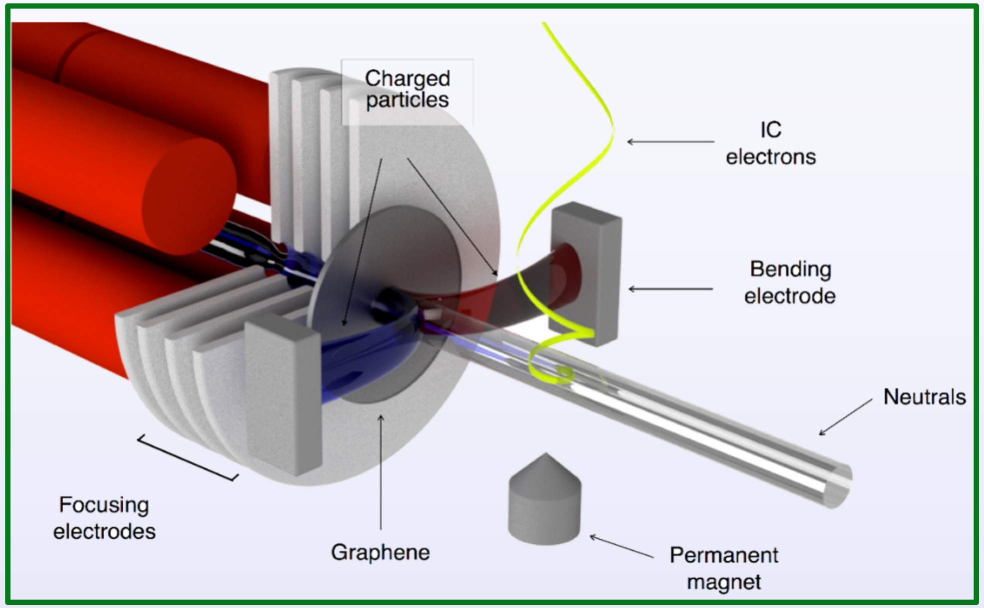
prolate deform.; $\dot{\alpha}$ sensitivity

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

sensitivity for $\dot{\alpha}$

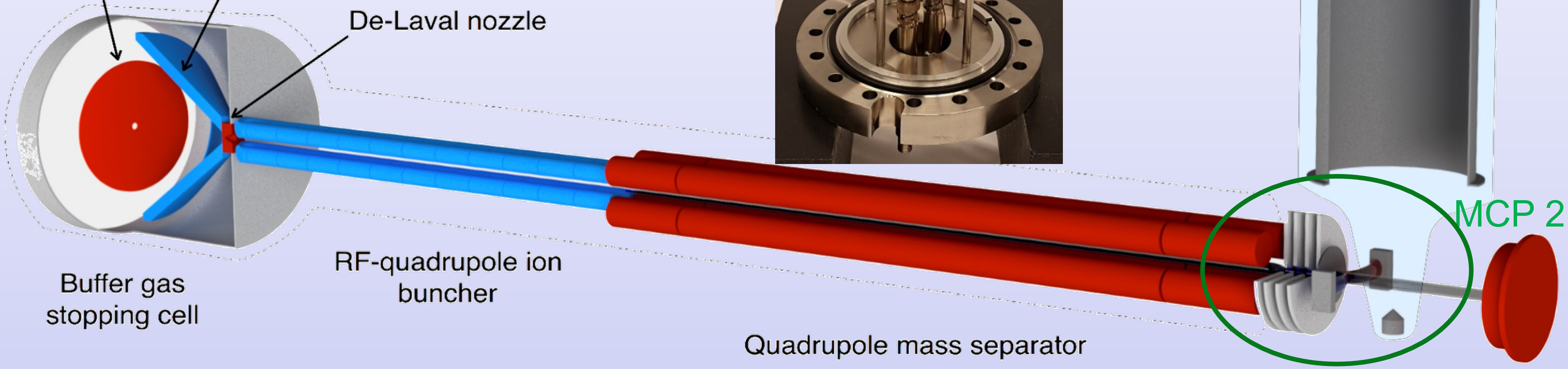
HFS: important for detection (tagging) of isomer excitation

Excitation Energy Measurement



neutralization of $^{229m}Th^{q+}$ in graphene foil:
→ contact-free IC decay
→ measure E_{kin} (e)
→ spectrometer resolution: 30 -50 meV

Magnetic bottle spectrometer



^{229m}Th Excitation Energy

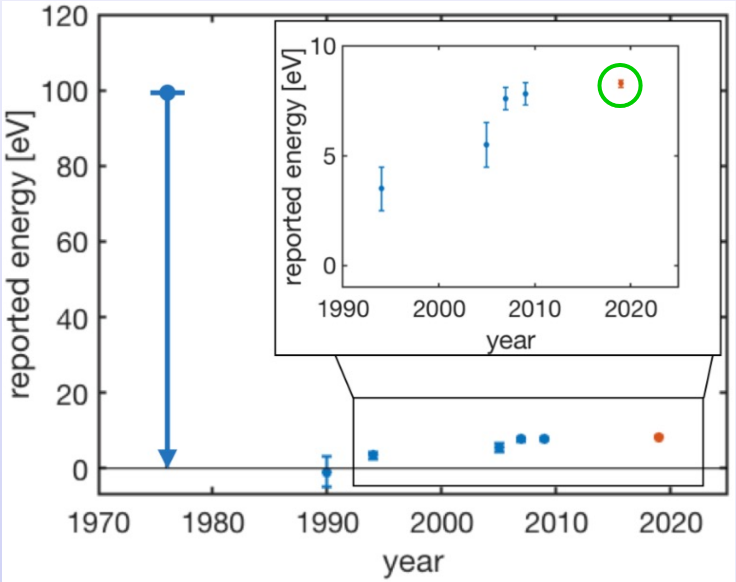
- via retarding field magnetic bottle spectrometer:

E* = 8.28 ± 0.17 eV
λ = 149.7 ± 3.1 nm

first direct measurement: Nature 575 (2019)

E*(iso) = 8.28 ± 0.17 eV (= 149.7 ± 3.1 nm)
B. Seiferle et al., Nature 575 (2019)
E*(iso) = 8.10 ± 0.17 eV (= 153.1 ± 3.7 nm)
T. Sikorsky et al., PRL 125 (2020)

E* ≈ 8.19(12) eV (= 150.4 ± 2.2 nm)



→ **clarifies regime of laser technology for optical control (presently excludes cw-laser crystal approaches)**

Missing: Radiative Decay

(prerequisite for solid-state nuclear clock)

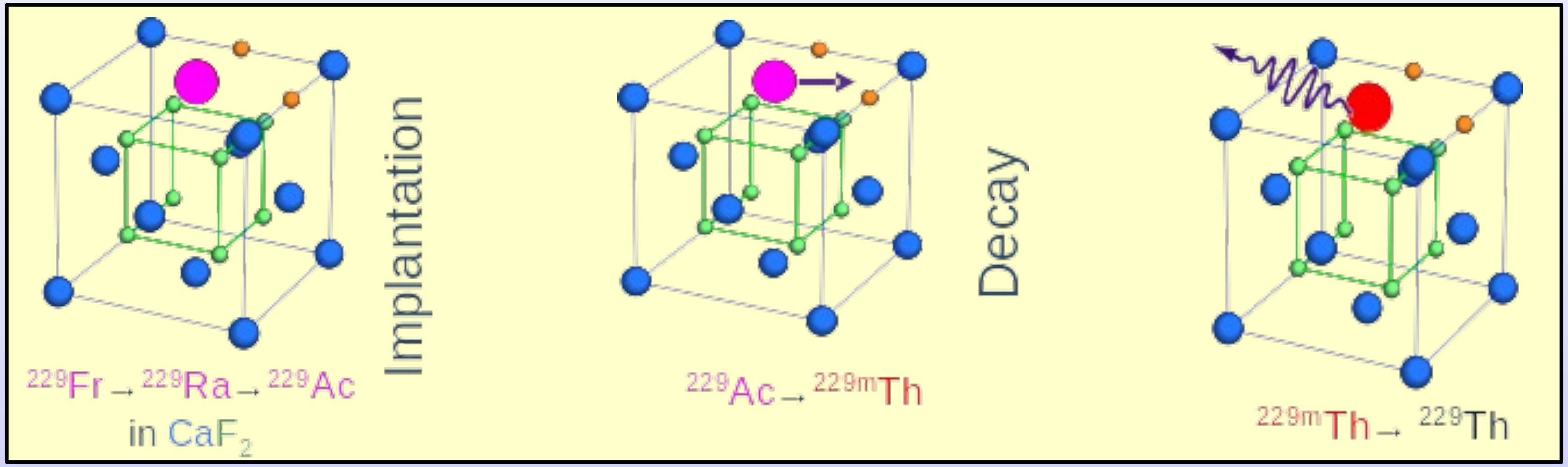
- **Photon spectroscopy of radioactive decay chains:**

  - Isomer population in radioactive decay
  - Implantation in (VUV transparent) large-bandgap crystals to ensure suitable chemical environment
  - Vacuum-ultraviolet spectroscopy of ~150 nm photons from radiative decay

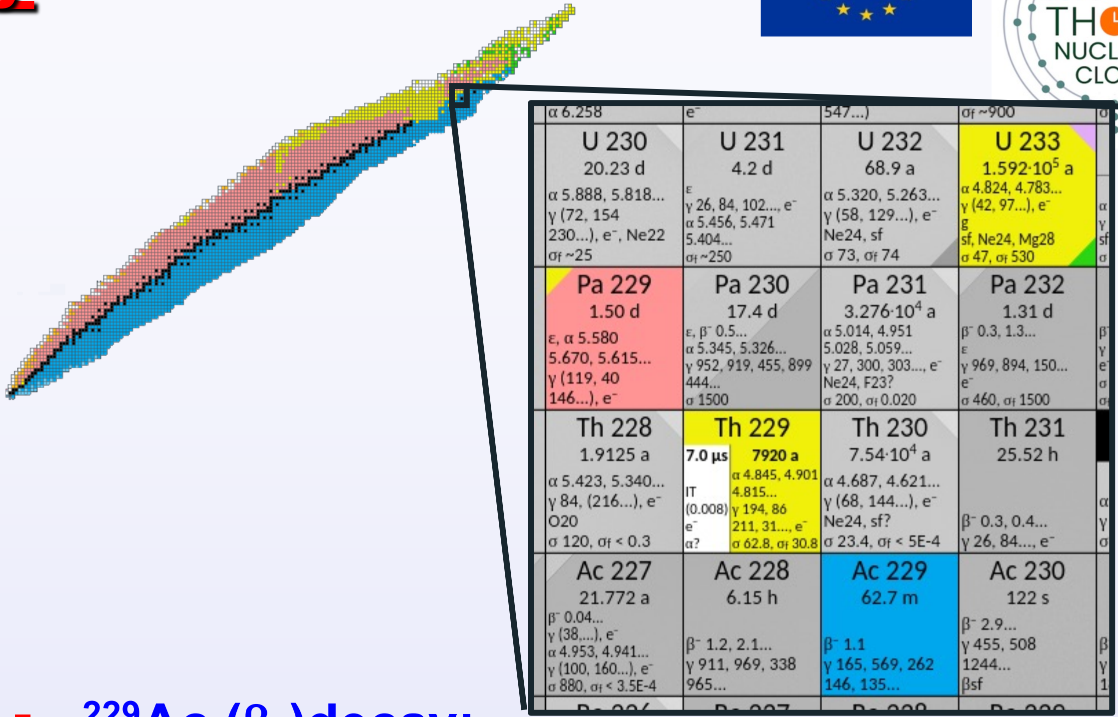
- **So far: experimental efforts using the alpha-decay of ^{233}U**

  → observation of radiative decay to-date unsuccessful

- **new approach:** using short-lived ^{229}Ac produced using ISOL technique (Isotope Production On-Line)



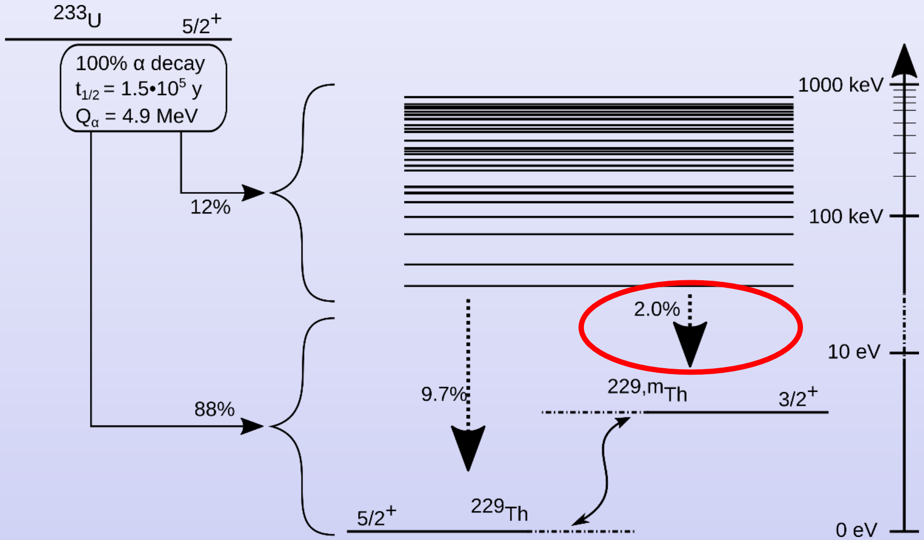
Exploit ^{229}Ac β decay



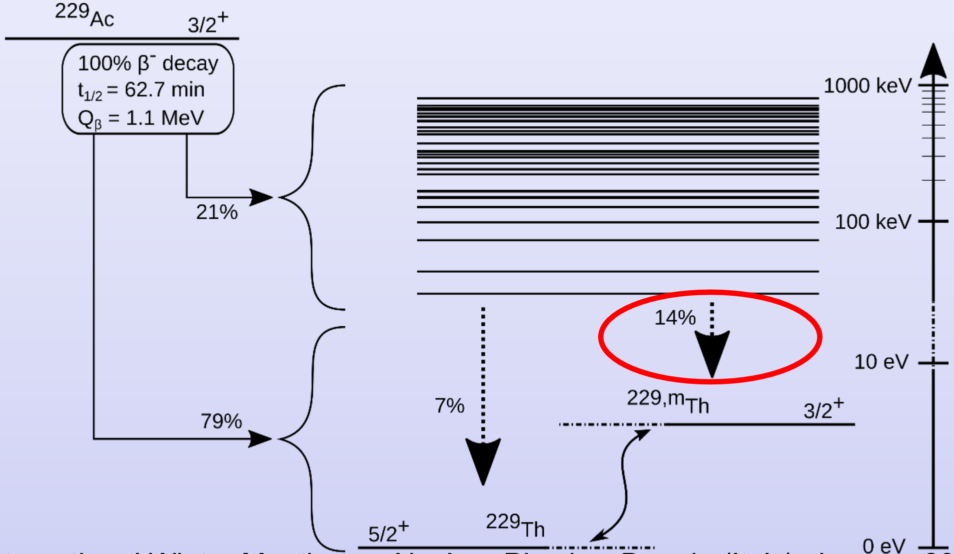
- **Efficient population of ^{229m}Th:**

	^{233}U	^{229}Ac
BR	2%	14%
Decay	α	β-
Recoil	84keV	<6eV
Production	stockpile	ISOL
Technique	doping	implantation

- **^{233}U (α-)decay:**



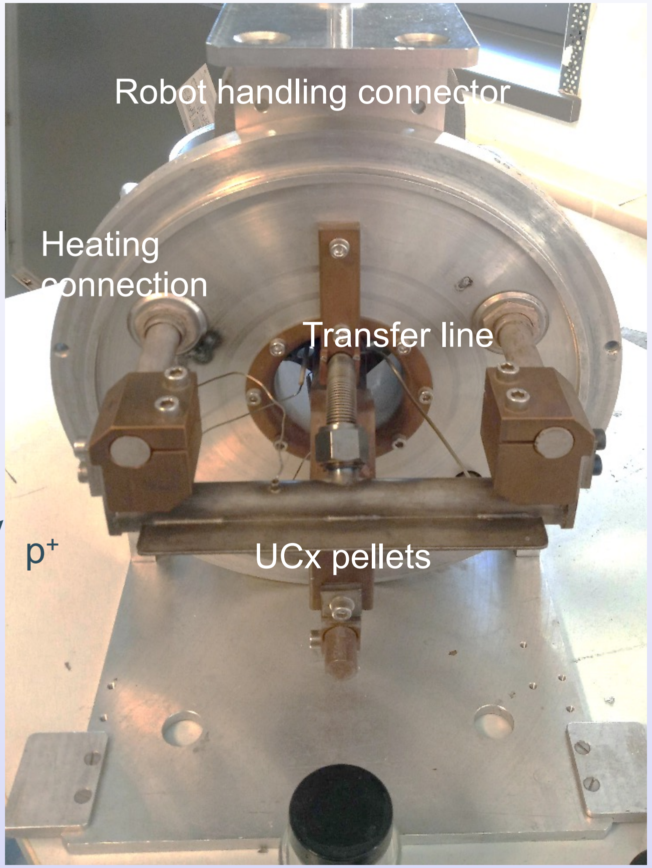
- **^{229}Ac (β-)decay:**



VUV spectroscopy at ISOLDE / CERN

Exp. collaboration led by KU Leuven group

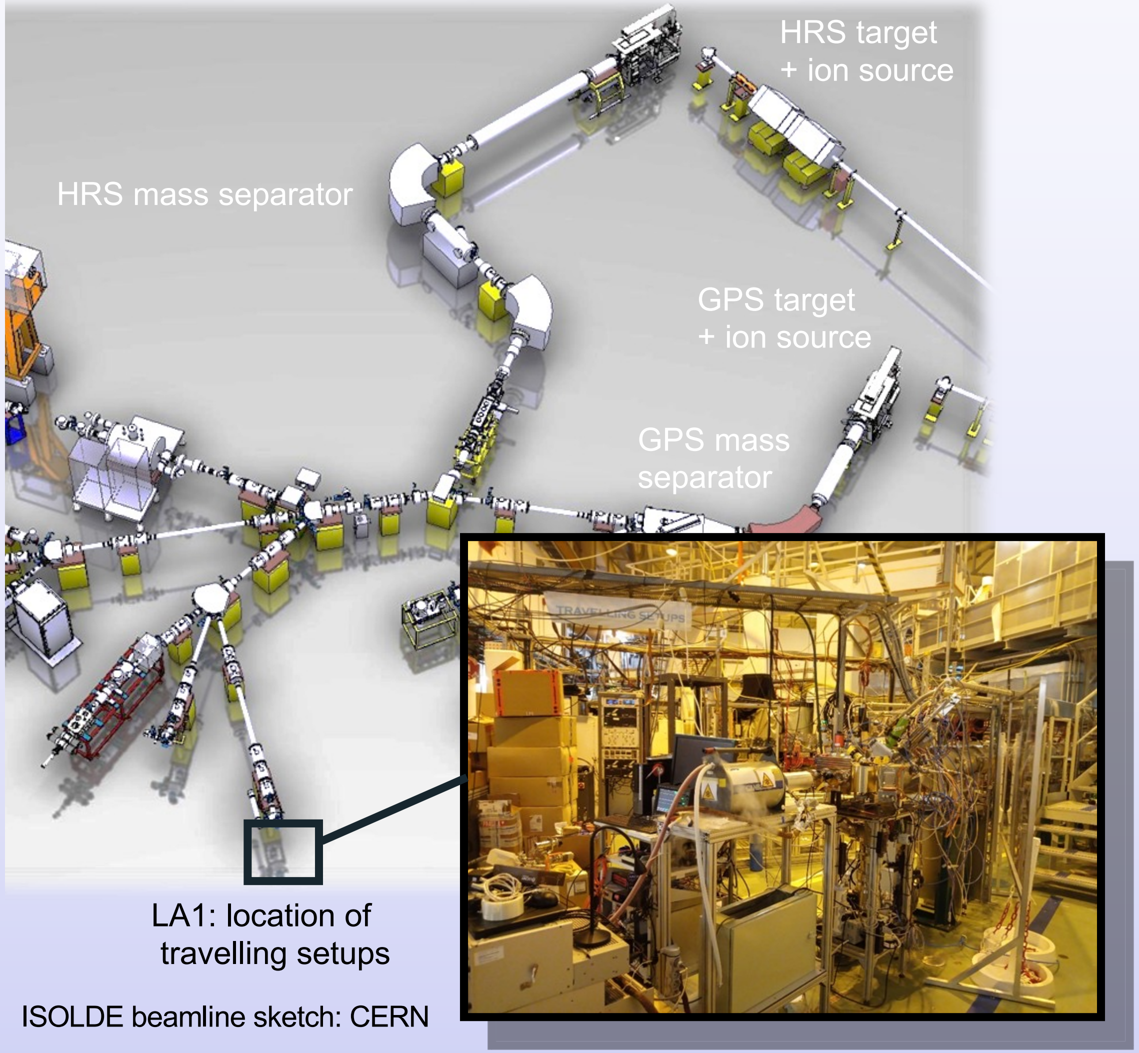
- **Production:** **1.4 GeV protons on UC_x**



Beam composition: ^{229}Fr, ^{229}Ra

^{229}Fr → ^{229}Ra → ^{229}Ac → ^{229m}Th/^{229}Th

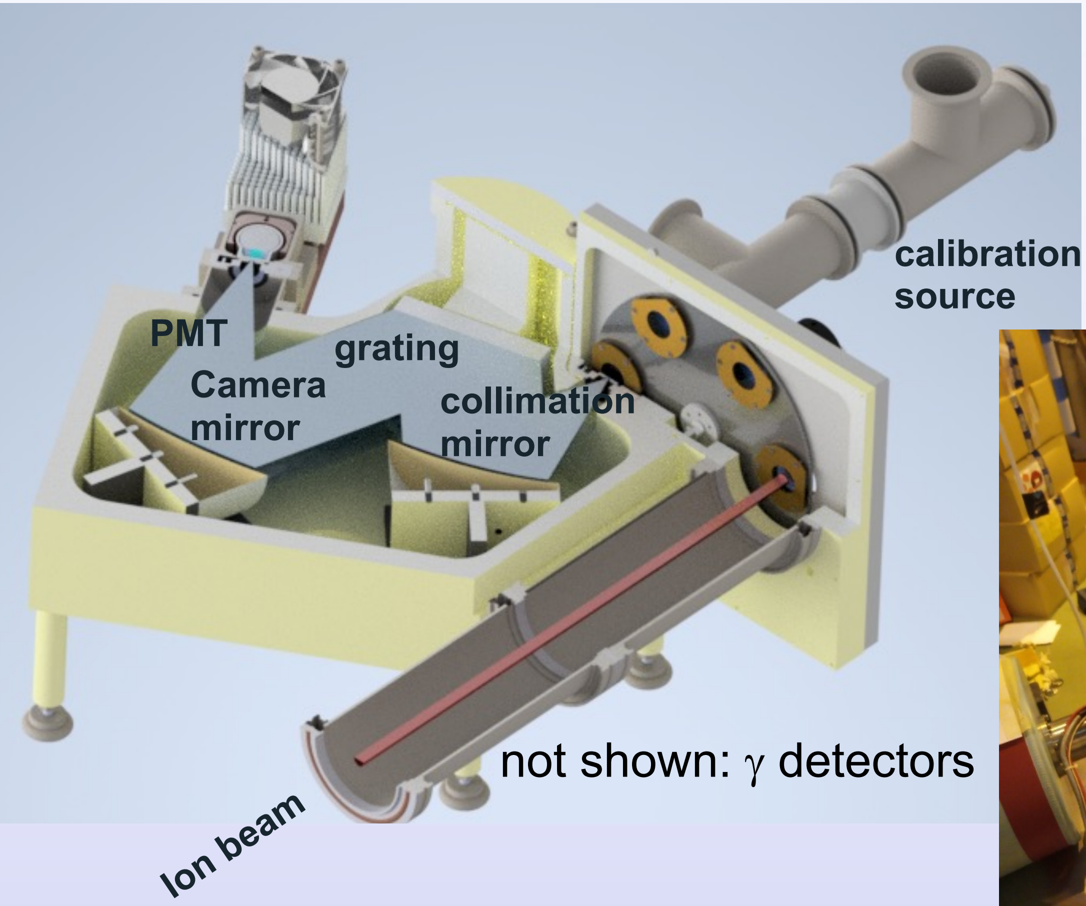
- **Beamline:** **ionization, mass separation, delivery**

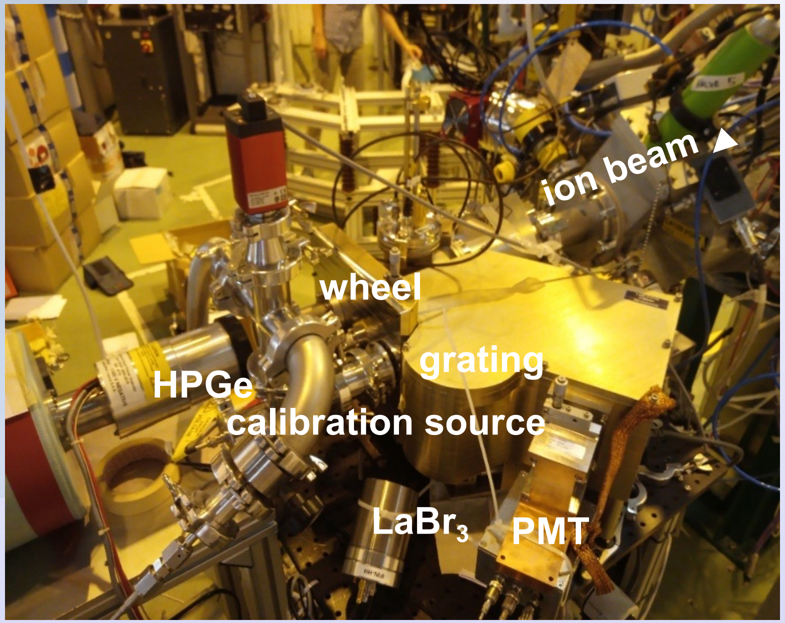


ISOLDE beamline sketch: CERN

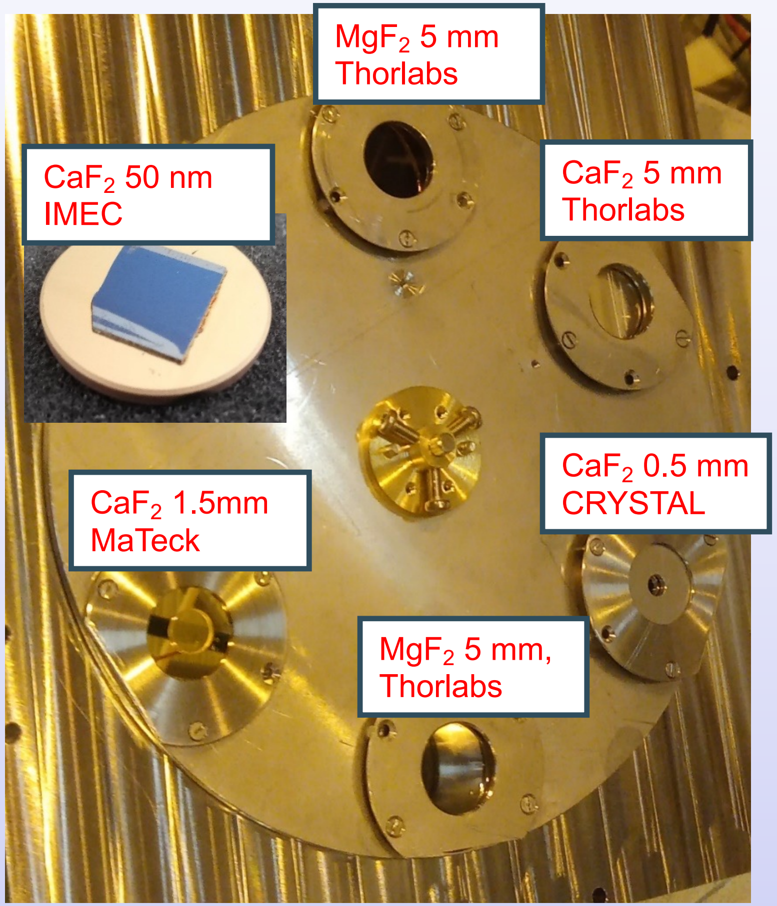
VUV spectroscopy at ISOLDE / CERN

VUV spectrometer:





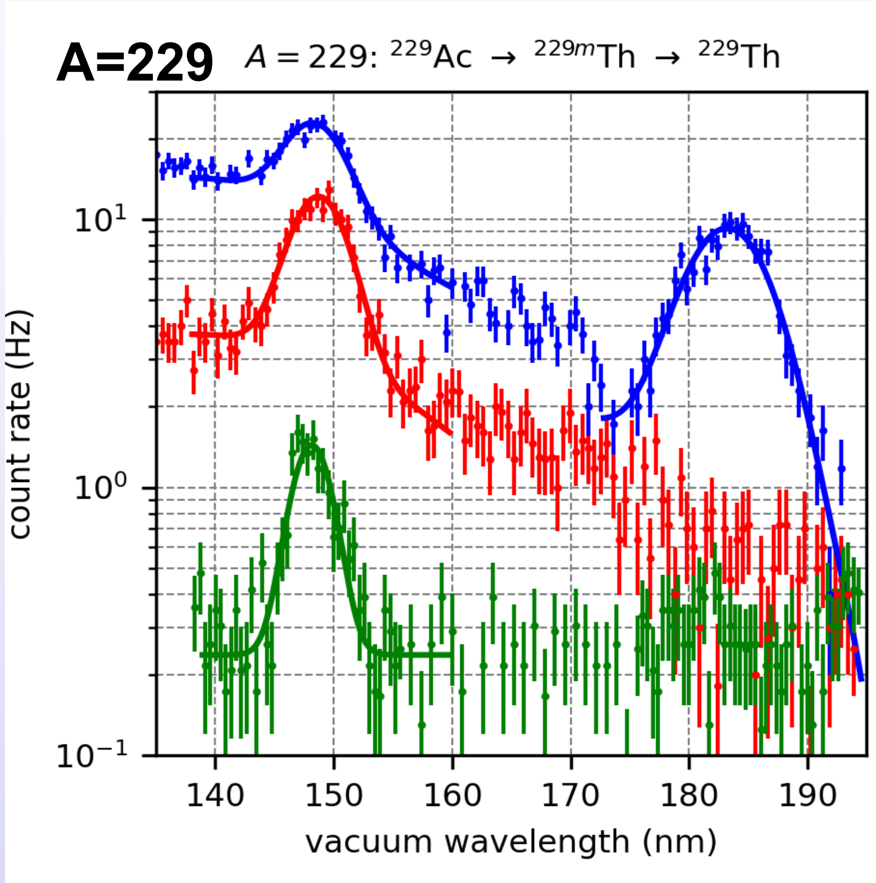
various crystals on wheel:

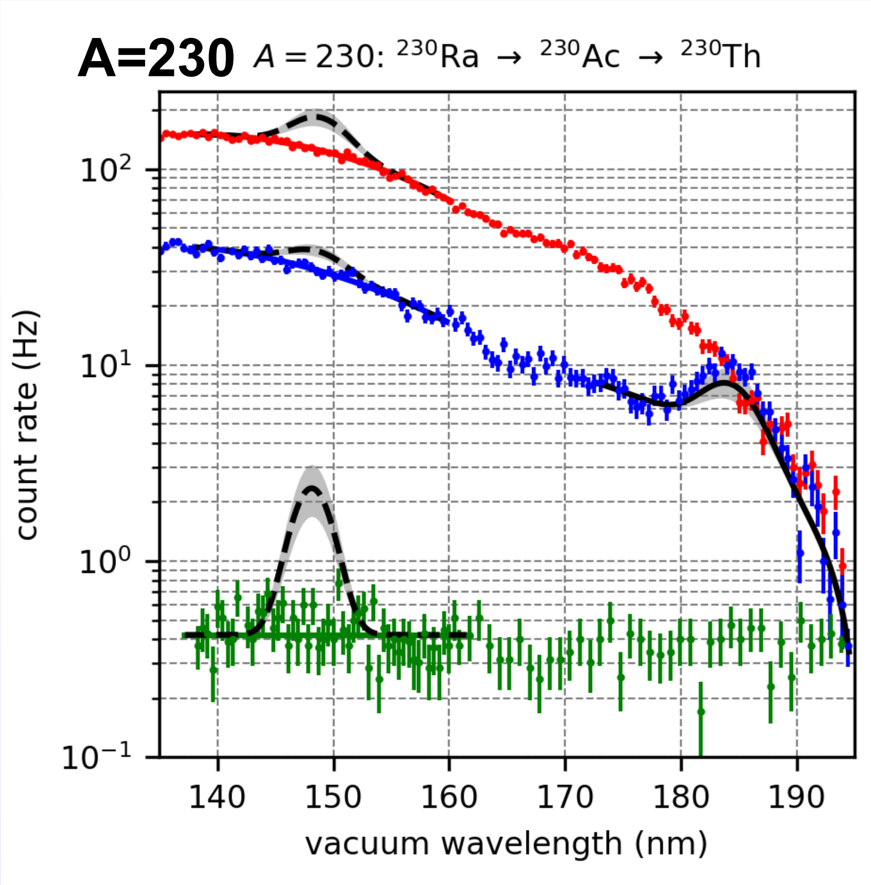


ISOLDE beam: ^{229}Fr, ^{229}Ra

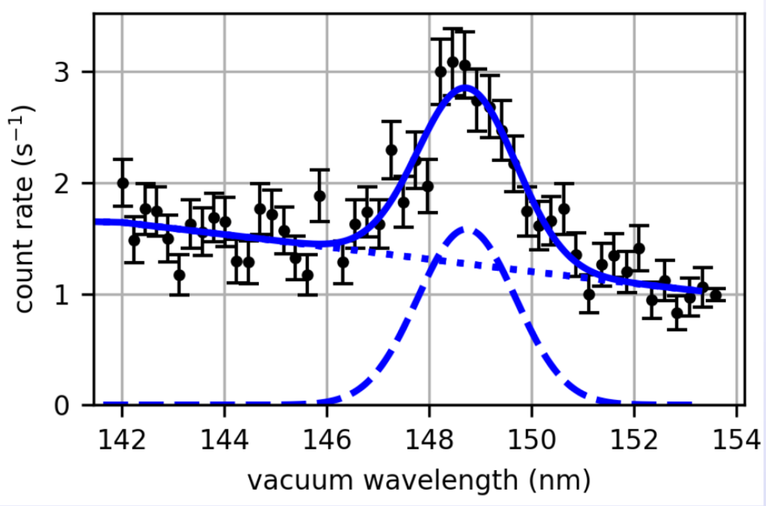
VUV Spectroscopy Results

MgF_2 (5 mm) CaF_2 (5 mm) CaF_2 (50 nm)





CaF_2 (5 mm)

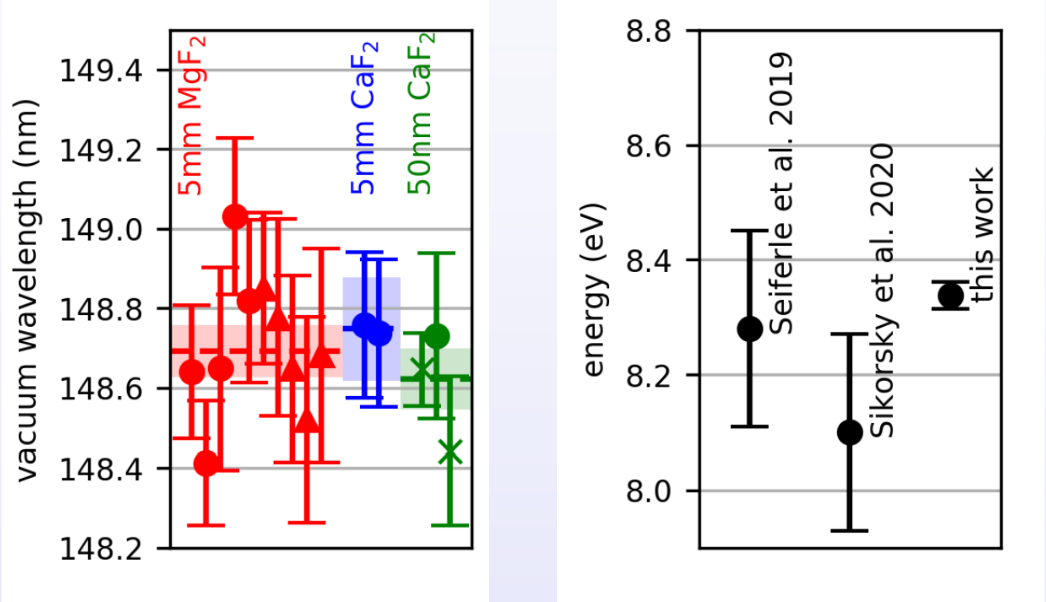


0.5 mm entrance slit

3 mm spectrometer entrance slit

VUV Spectroscopy Results

excitation energy/ emission wavelength:



8.338 ± 0.003(stat.) ± 0.023(syst.) eV

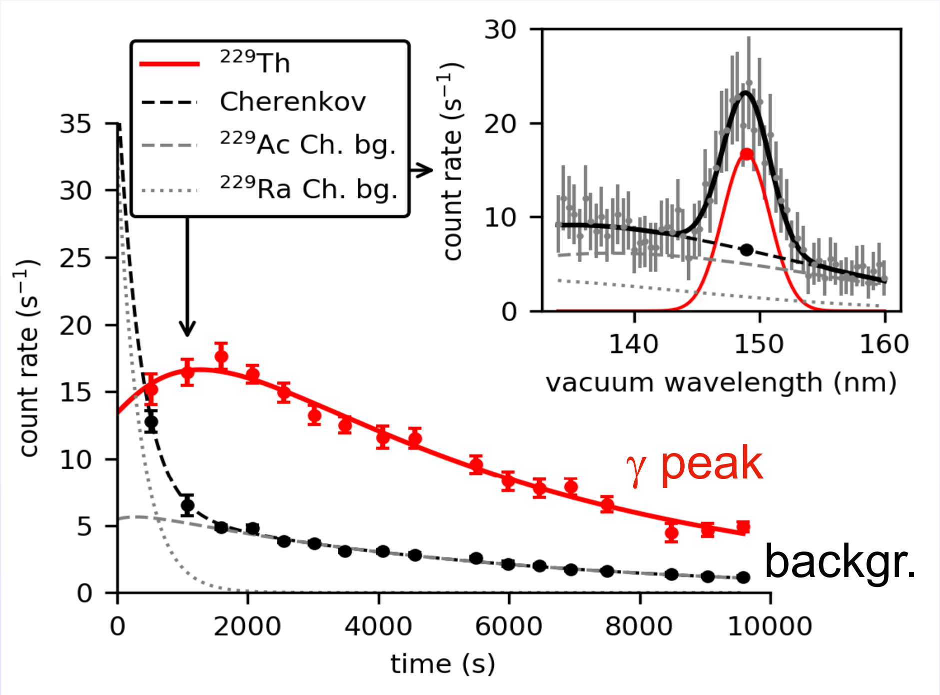
148.71 ± 0.06(stat.) ± 0.41(syst.) nm

$E^*(^{229m}Th)$ = 8.338(24) eV
λ = 148.71(42) nm

→ important for ongoing VUV laser developments

time evolution (after 1 hr. implantation):

MgF_2 (5 mm), 2 mm entrance slit, 5 s/grating pos.



→ **$t_{1/2}$ = 670(102) s**

- for decay of ^{229m}Th embedded in MgF_2 crystal
- direct $t_{1/2}$ measurement in cryo-Paultrap in preparation (LMU)

S. Kraemer et al., arXiv 2209.10276 (accepted in Nature)

Perspectives for the Nuclear Clock

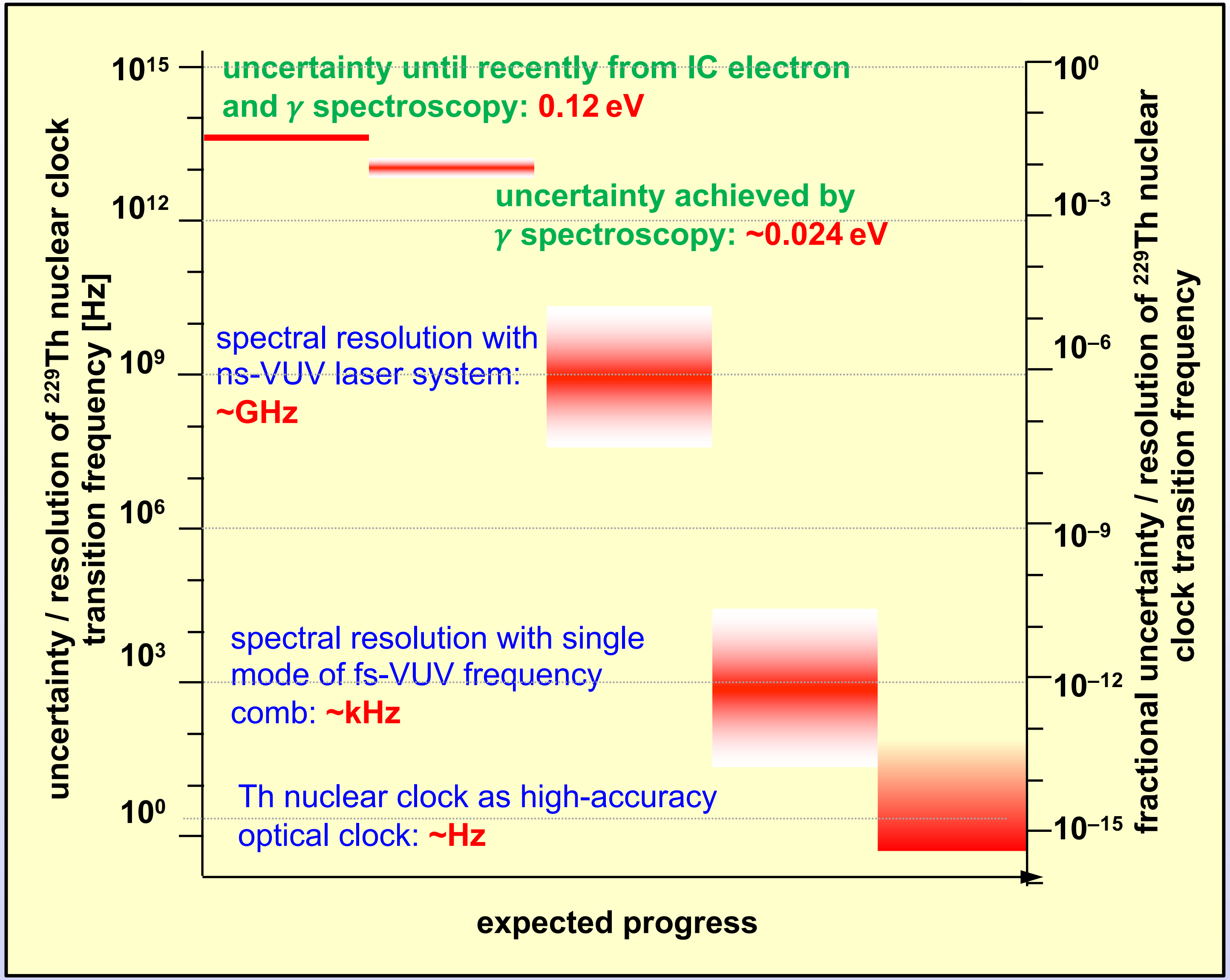
- **still to bridge: 10-11 orders of magnitude:**

" from eV to (k)Hz"

- already feasible with existing laser technology concept: L. v.d. Wense, PT et al, PRL 119 (2017)

- (4-wave mixing) laser setups at PTB (E. Peik et al.), UCLA (E. Hudson et al.), JILA/NIST (J. Ye et al.)

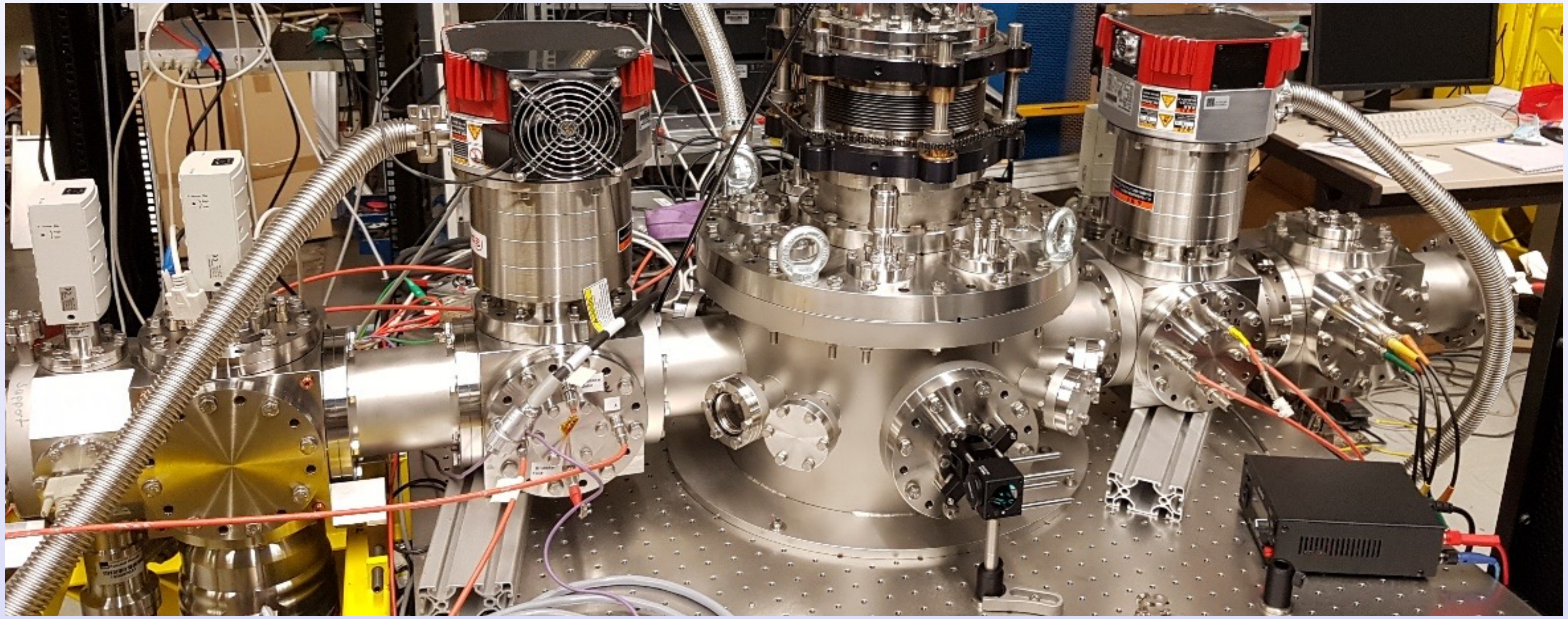
- (VUV frequency comb) laser under development (ILT Aachen + LMU)

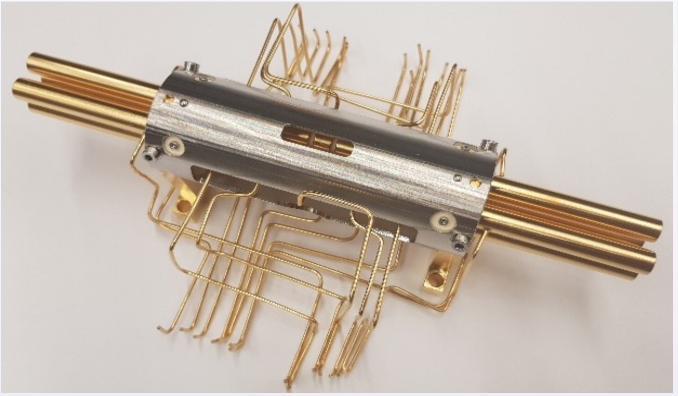


Ionic Lifetime Measurement

needs longer storage time (= better vacuum)

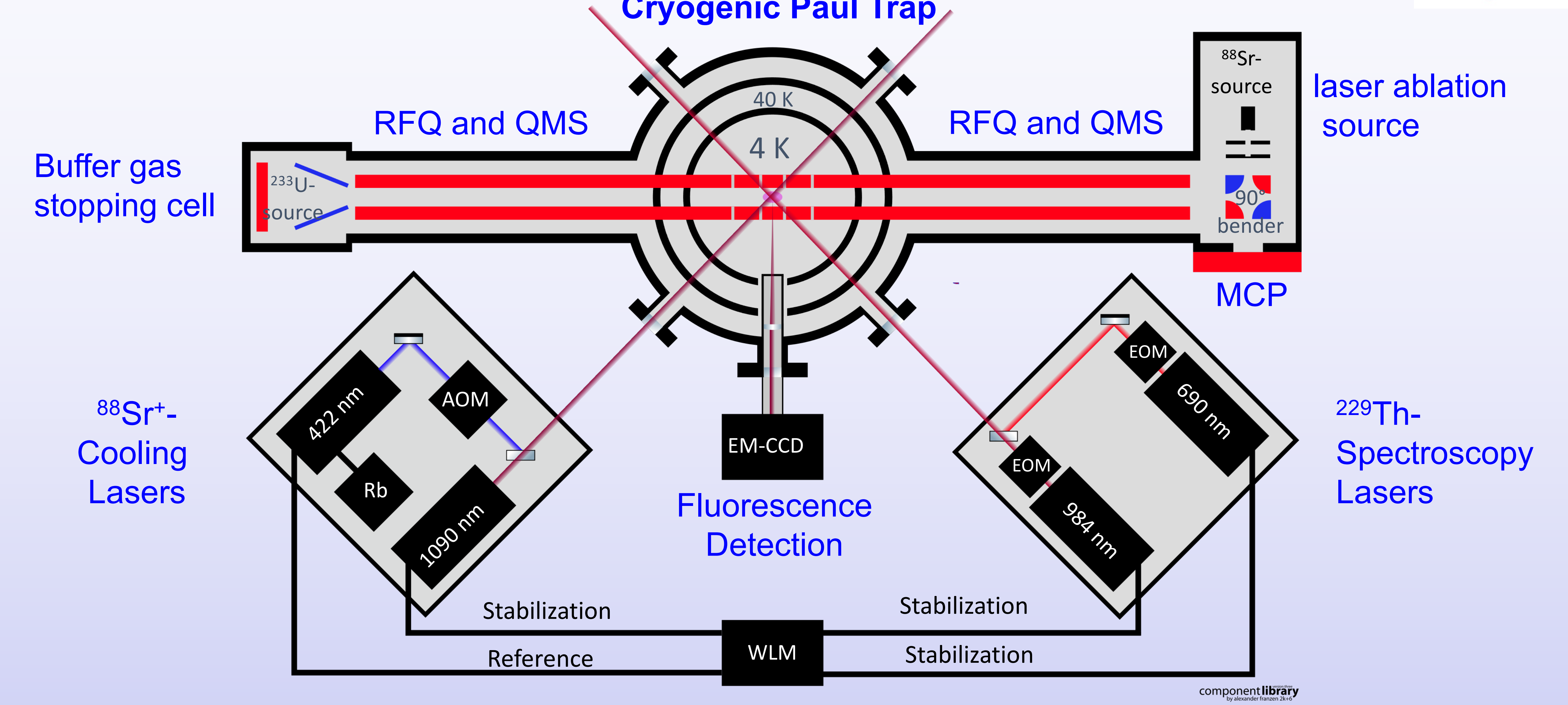
- setup of a cryogenic Paul trap
- platform for laser manipulation
- ionic lifetime measurement: via HFS spectroscopy of $^{229m}Th^{3+}$





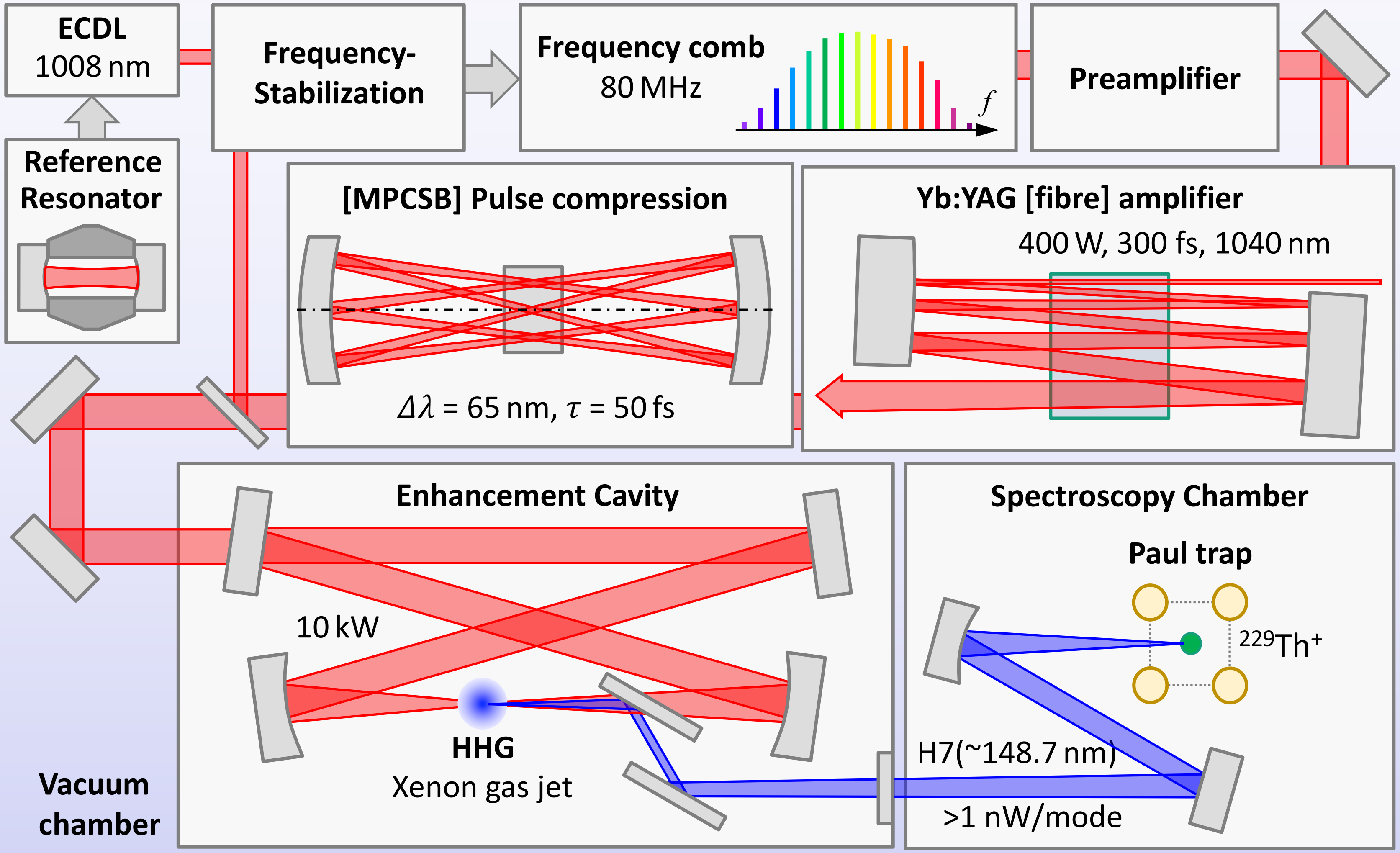
- sympathetic laser cooling with $^{88}Sr^{+}$ set up and ready
- ready for commissioning of fluorescence detection

Cryo Trap & Laser Setup



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

- VUV frequency comb: use 7th harmonic of amplified IR frequency comb:



laser under development at Fraunhofer ILT (Aachen)

operational: early 2024

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

Summary

Exp. achievements in last 5 years:

- identification & characterization of the thorium isomer:
  direct IC decay, neutral $t_{1/2}$, hyperfine structure, E*
- first observation of radiative decay mode:
  E* (^{229m}Th) = 8.338 (24) eV, λ = 148.71(42) nm , $t_{1/2}$ (in MgF_2) = 670(102) s

Ongoing activities & next steps

- directly determine ^{229m}Th ionic lifetime:
  cryogenic Paul trap, sympathetic (Sr^+) laser cooling, HFS spectroscopy
  → commissioning ongoing at LMU
- identify nuclear resonance with laser spectroscopic precision:
  → broadband (4-wave-mixing) lasers operational
  → narrowband laser (VUV frequency comb) under development
- determine sensitivity enhancement for $\dot{\alpha}$
- doped-crystal approach: radiative, IC branches

Ambitious goals lie ahead:

- excite for the first time a nuclear transition by laser
- drastically improve sensitivity to new physics ($\dot{\alpha}$, DM candidates)

Thanks to ….

LMU Munich: L. v.d. Wense, B. Seiferle, K. Scharl, D. Moritz, S. Ding, L. Löbell, I. Hussain, F. Zacherl

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

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

TU Wien: ***T. Schumm***, S. Stellmer, K. Beeks, C. Lemell, F. Libisch

MPQ: ***J. Weitenberg***

MPIK-HD: ***A. Pàlffy***, P. Bilous, N. Minkov, J. Crespo

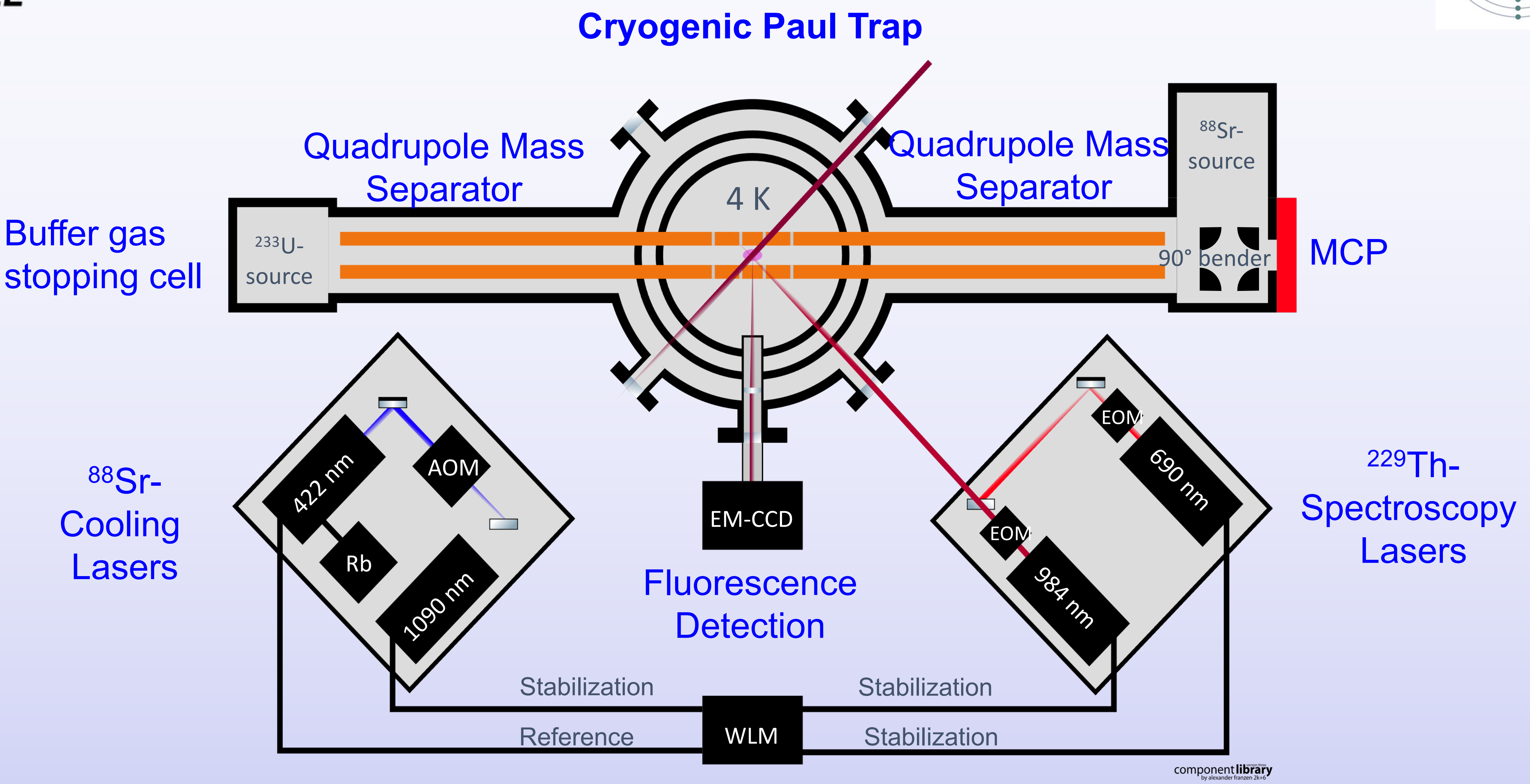
NIST: S. Nam, G. O'Neil

UCLA: E. Hudson, C. Schneider, J. Jeet

U Delaware: ***M. Safronova***

Thank you for your attention !

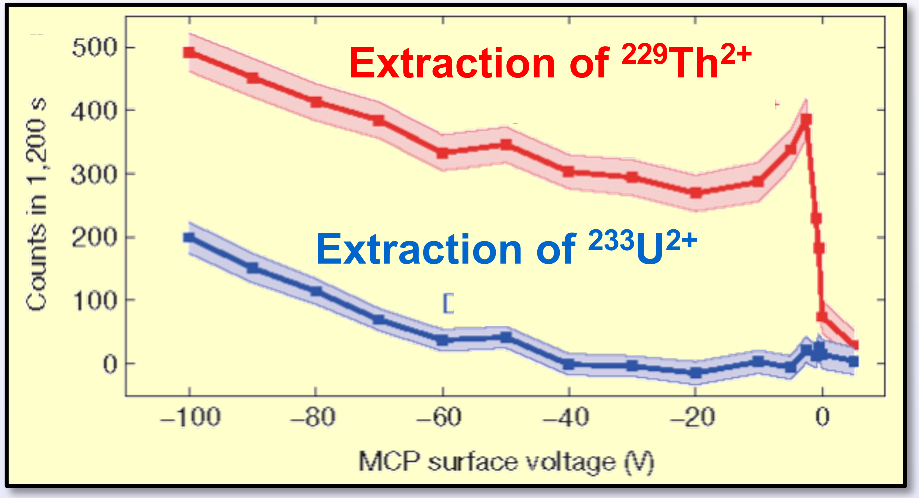
Cryotrap Setup

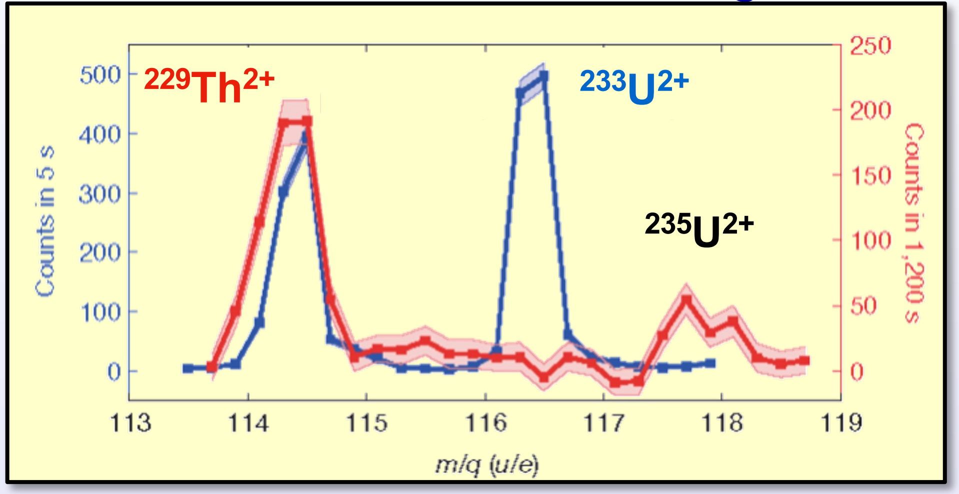


Verification Measurements

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**

^{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)

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})

- 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)

- almost degenerate doublet ^{229,229m}Th:
  → from cancellation between large contributions from Coulomb energy V_C ~10^9 eV
- dependency of V_C on fundamental constants:
  → any variation would be enhanced in the ^{229m}Th transition frequency

- **current (theor.) limit of potential time variation:**

$$\dot{\alpha}/\alpha = (-0.7 \pm 2.1) \cdot 10^{-17}\ \text{yr}^{-1}$$

R. Godun et al., PRL 113, 210801 (2014)

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: Coulomb energy difference between ^{229}Th, ^{229m}Th

enhancement factor:

$$K = \frac{\Delta V_C}{\omega}$$

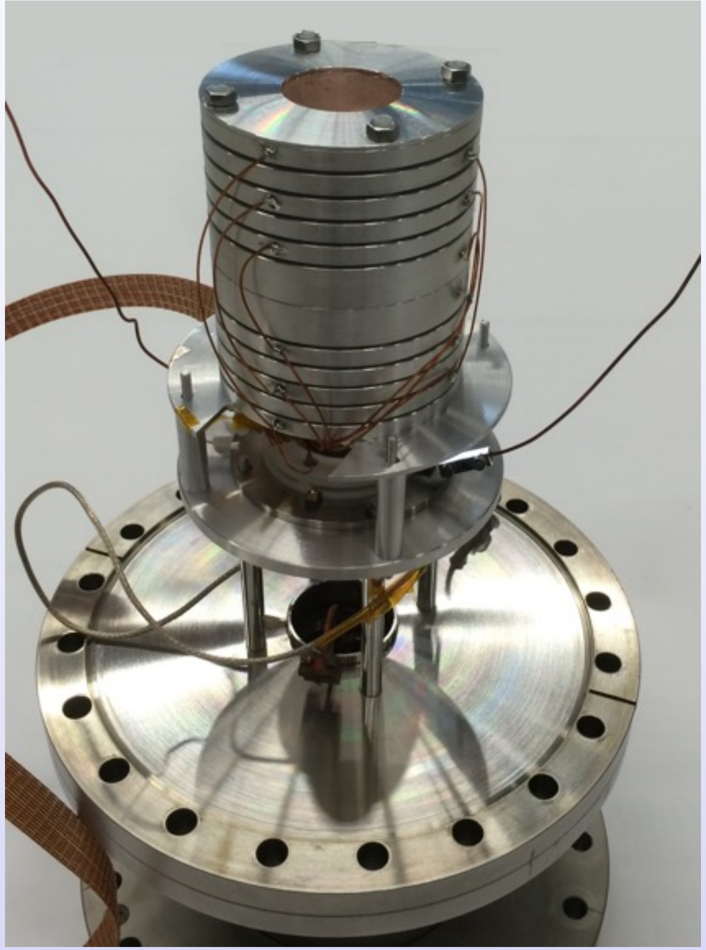
- **problem:** - Coulomb energy cannot be directly measured
  - V_C needs to be calculated in nuclear models
  - insufficient accuracy in present nuclear models

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

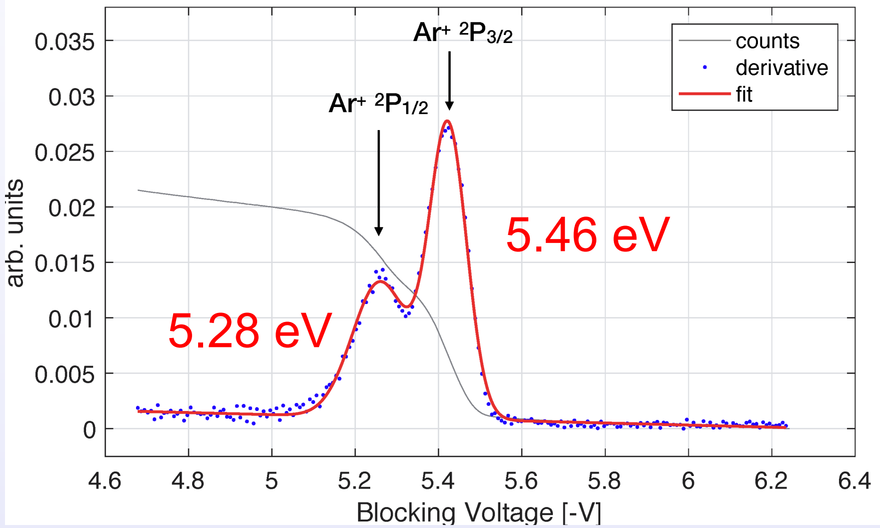
Electron Spectrometer

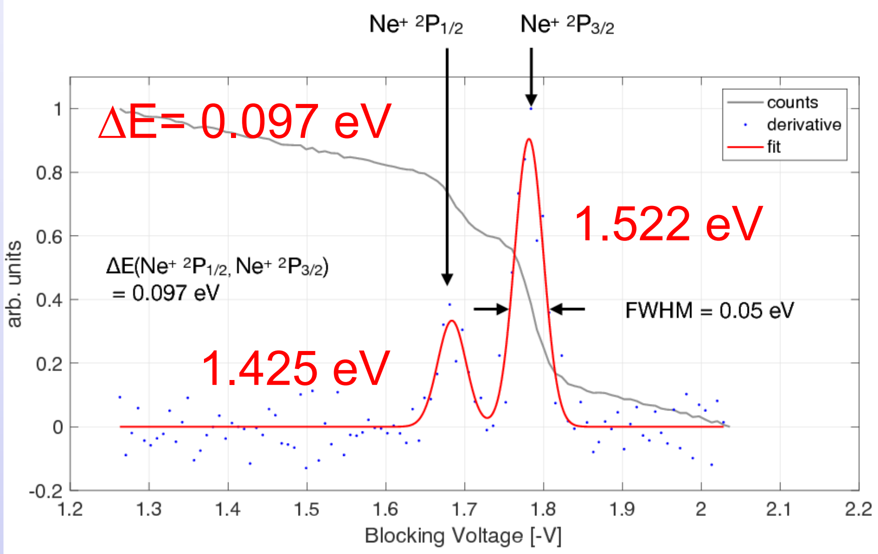
- Magnetic-bottle spectrometer:



spectrometer: ΔE ~ 3% (0.03-0.05 eV)

- Calibration: gas discharge excitation:





^{229m}Th: Testbench for variation of α ?

model: - small isomeric transition energy results from cancellation of change in Coulomb energy of ^{229}Th, ^{229m}Th:

$$\Delta E_C = E_C^m - E_C$$

- by strong force via proton polarization by modified neutron distribution

→ cancellation: sensitive to (changes of) coupling constants of electromagnetic and strong interaction

assumption: nucleus = uniform, hard-edged, prolate ellipsoid

Berengut et al., PRL 102, 210801 (2009) (with updated Q_0):

$$\Delta E_C = (-485\ \text{MeV})\ [\ (\langle r_{229m}^2\rangle / \langle r_{229}^2\rangle) - 1]$$
$$+ (11.6\ \text{MeV})\ [(Q_0^m / Q_0^g) - 1\ ]$$

→ $\Delta E_C = -0.29(43)$ MeV

$$K = \frac{\Delta E_C}{\omega}$$

→ still not sufficiently precise to prove that $|\Delta E_C| \gg E_{IS}$ but still leaves room that ^{229}Th nuclear clock largely exceeds α sensitivity of atomic clocks

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)

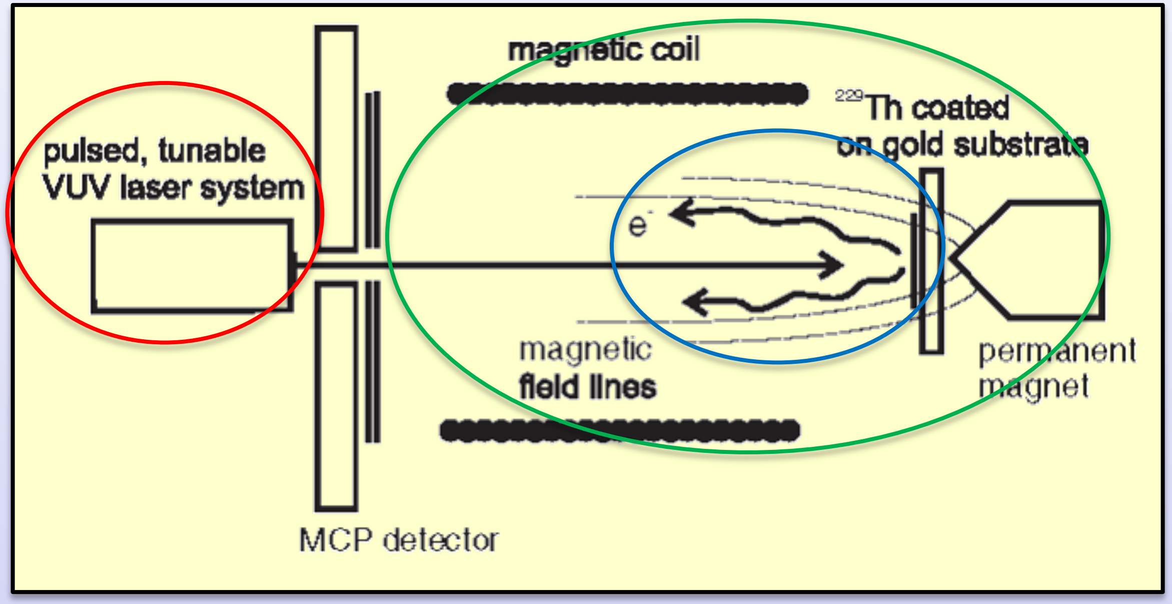
Laser excitation scheme for ^{229m}Th

- **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%



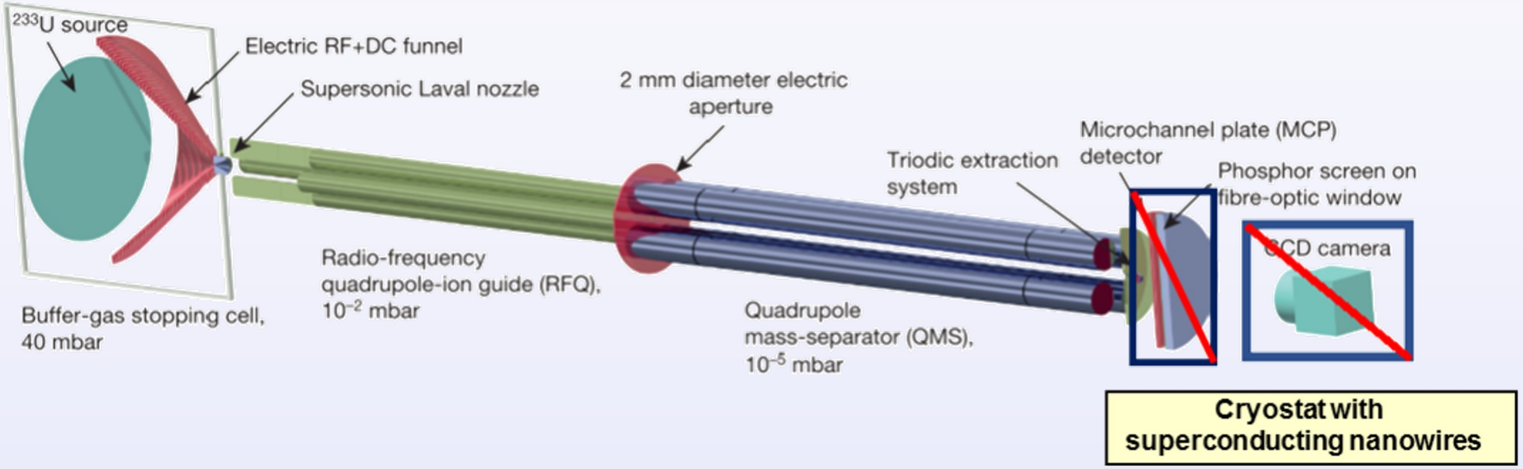
IC electron detection:
- ^{229}Th coated sample in magnetic bottle (could be curved)
- detection time: 10 μs

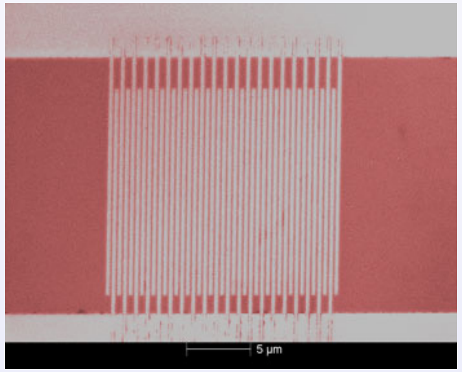
High signal-background:
S/B ~ 7 x 10^4
Short scan time:
< 3 days for 1 eV

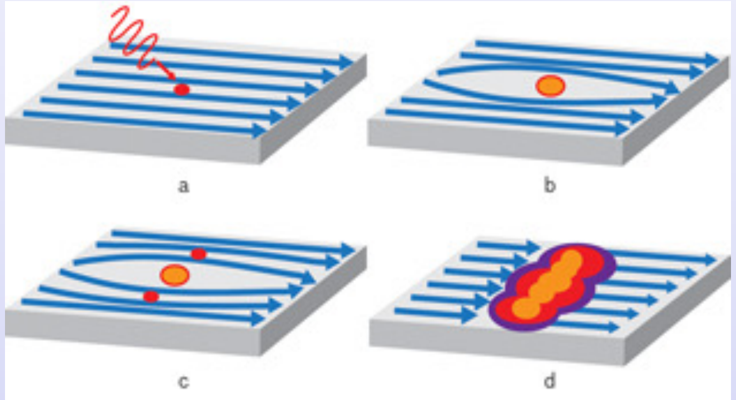
* S.J. Hanna et al., Int. J. Mass Spectr. 279, 134 (2009)

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

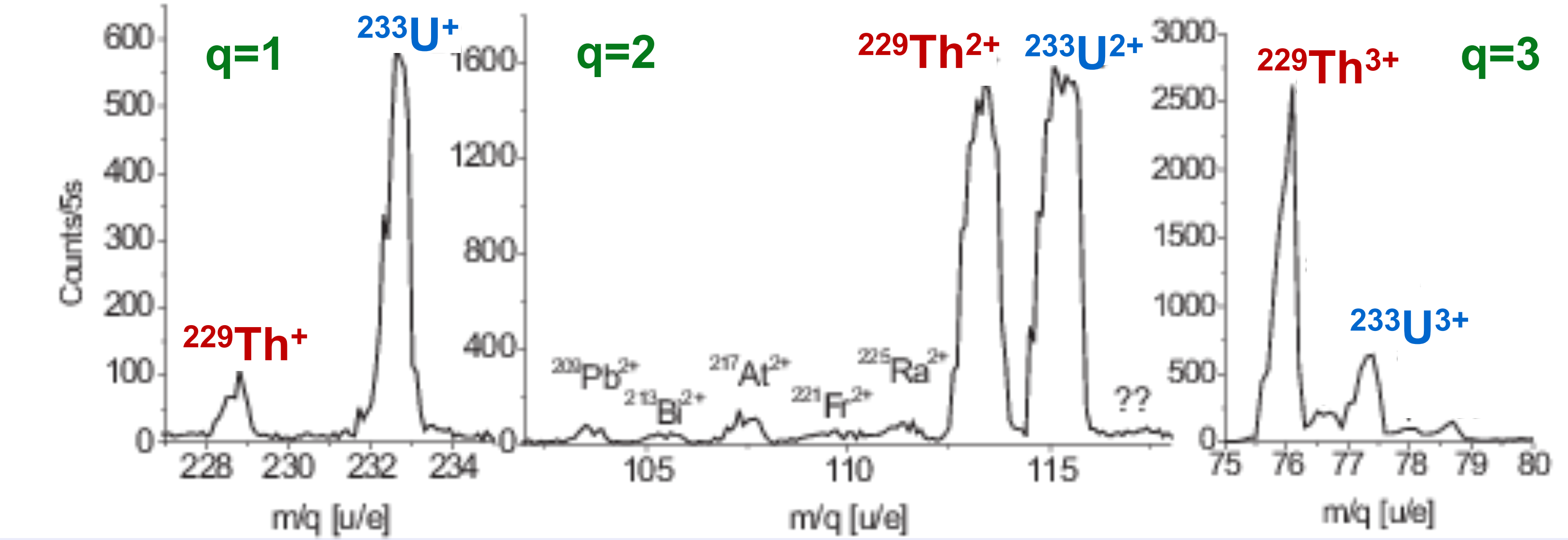
measurements are ongoing with first promising results

collaboration with UCLA, NIST/Boulder, TU Wien

Ion Extraction from Buffer Gas Cell

efficient $^{229(m)}Th^{3+}$ extraction

mass scan of extracted ion species:



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)

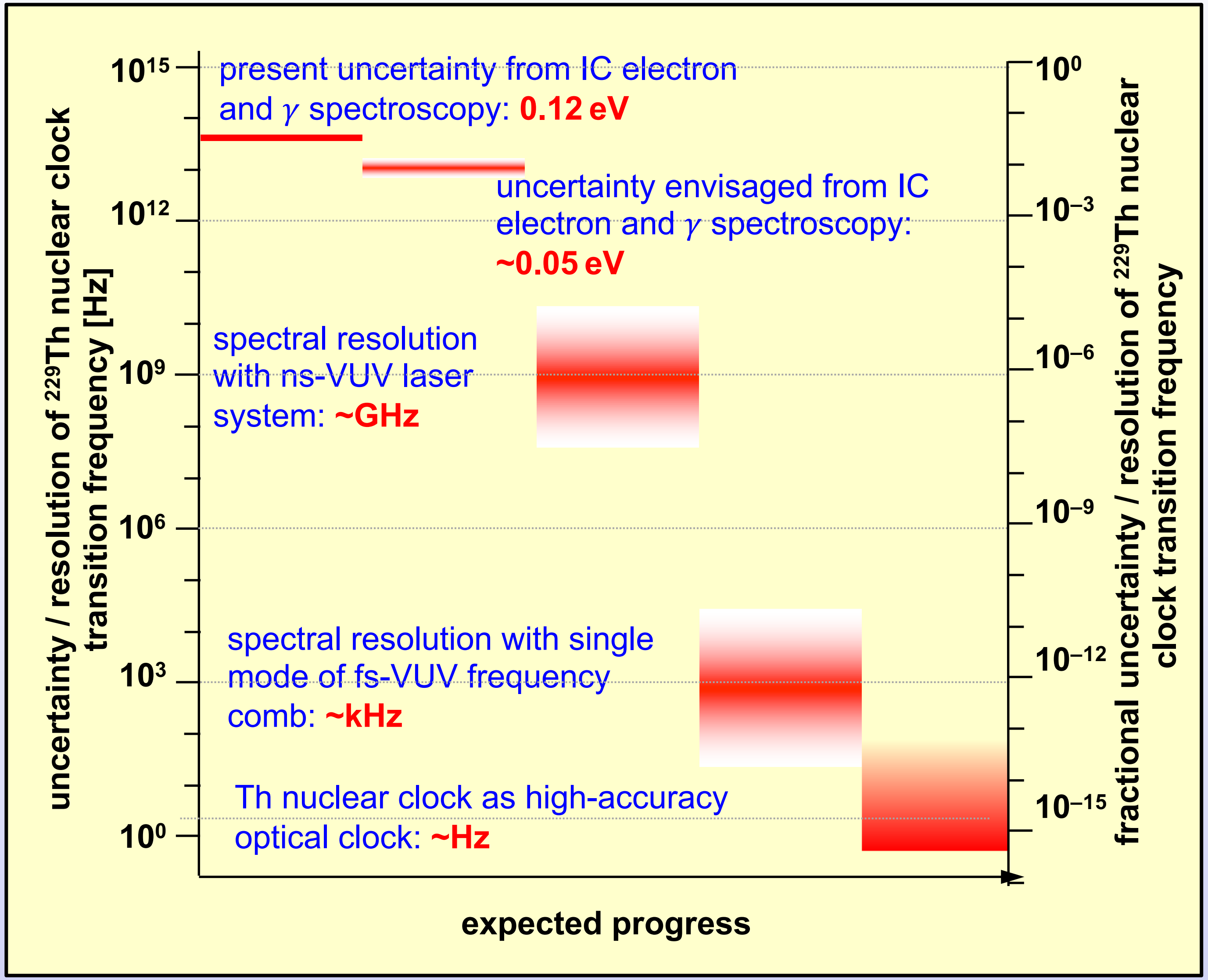
Towards the Nuclear Clock

still to bridge: 11-12 orders of magnitude: " from eV to (k)Hz"

- already feasible with existing laser technology
  L. v.d. Wense, PT et al, PRL 119 (2017)

- (4-wave mixing) laser set up at PTB (E. Peik et al.)

VUV frequency comb laser under development (Fraunhofer + LMU)



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)
  first direct measurement: Nature 575 (2019)

"ThoriumNuclearClock"

E. Peik, T. Schumm, M.S. Safronova, A. Pálffy, J. Weitenberg, P.G. Thirolf,
Quantum Sci. Technol. 6, 034002 (2021)

- **ERC Synergy project: 2020-2026** https://thoriumclock.eu
- **Team:** PI: PTB (E. Peik et al.), TU Wien (T. Schumm et al.), LMU Munich (P. Thirolf et al.), U Delaware (M. Safronova et al.) + A. Pálffy (U Würzburg), J. Weitenberg (Fraunhofer ILT/RWTH Aachen)

Fundamental and technology goals

- Perform the first laser excitation of a nuclear transition
- Precisely determine ^{229}Th nuclear structure parameters
- Development of a < kHz-level linewidth VUV laser
- Quantify the sensitivity to fundamental constants
- Measure isomer energy to > 12 digits
- Demonstrate nuclear clock with Th^{3+} ions
- Demonstrate completely new solid-state clock scheme
- Test fundamental concepts of physics
- Search for dark matter

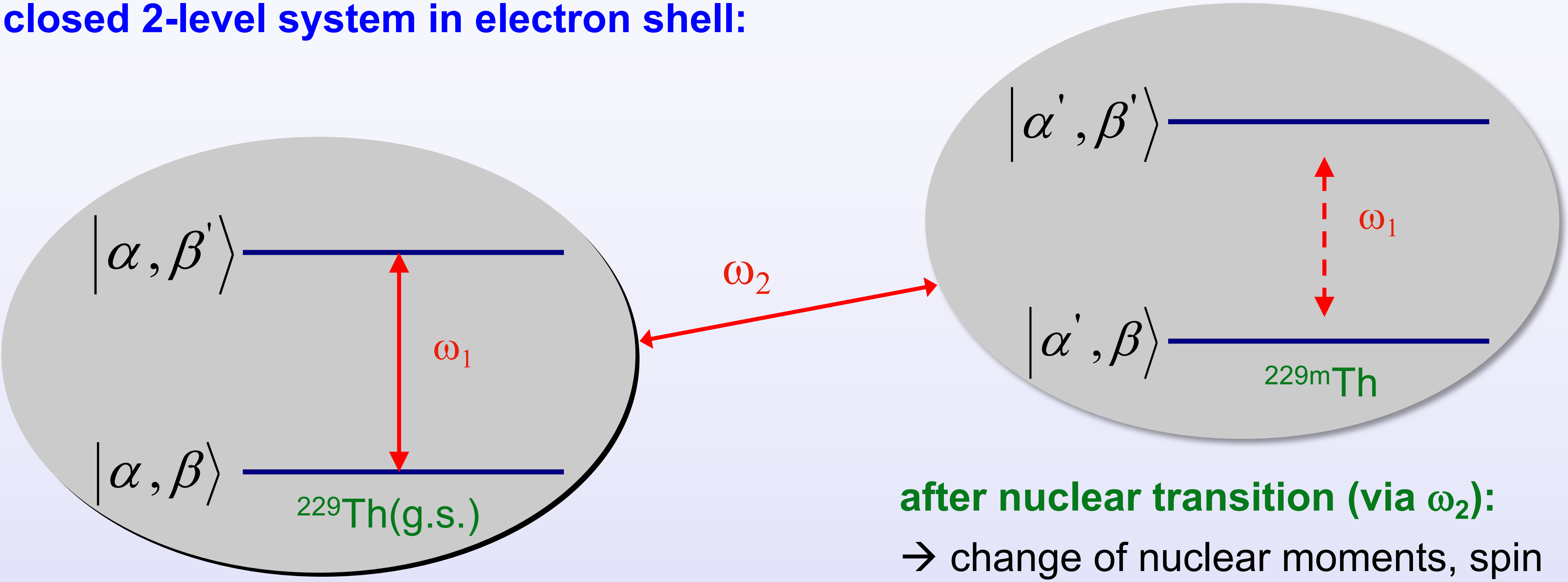
THORIUM NUCLEAR CLOCK

Scientific advances in many fields

- Precision metrology (MAX-PLANCK-GESELLSCHAFT, PTB)
- Nuclear physics (PTB, MAX-PLANCK-GESELLSCHAFT, University of Delaware)
- Atomic physics (LMU, PTB, University of Delaware)
- Laser physics (LMU, TU Wien, ILT, MAX-PLANCK-GESELLSCHAFT)
- Particle physics (University of Delaware, MAX-PLANCK-GESELLSCHAFT)
- Solid-state physics (LMU, TU Wien, MAX-PLANCK-GESELLSCHAFT)
- Radiochemistry (TU Wien, PTB)

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

Summary

look back: huge progress in last 5 years:

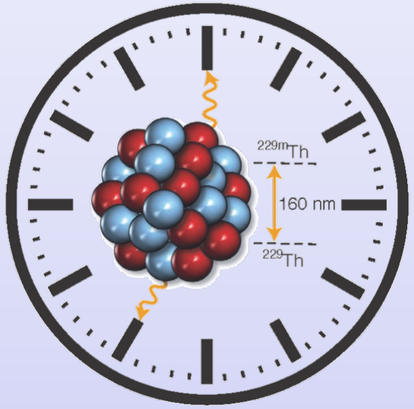
- identification & characterization of the thorium isomer

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

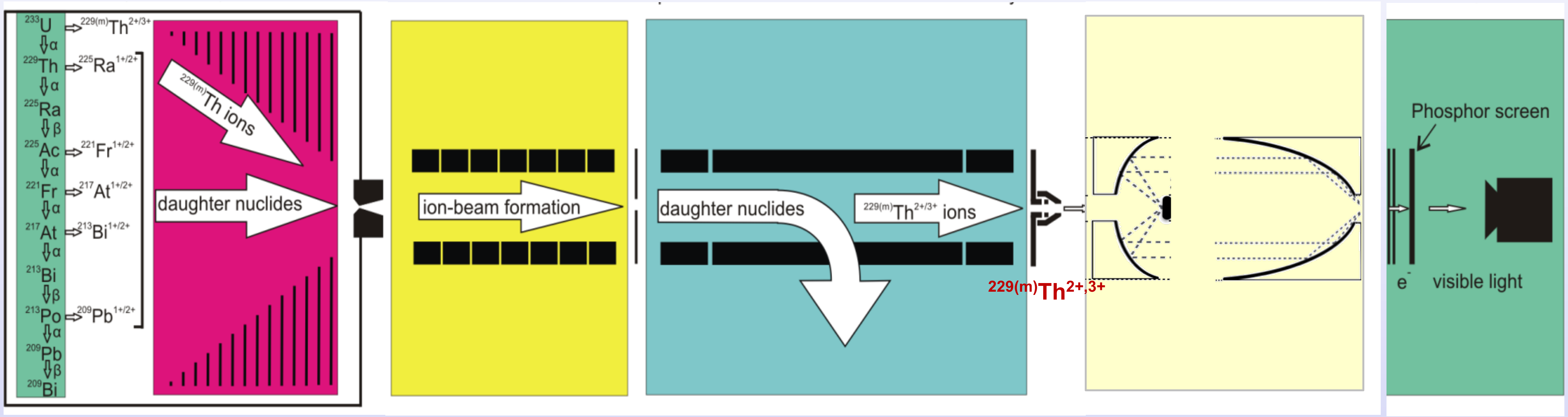


Experimental Approach @ LMU

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**

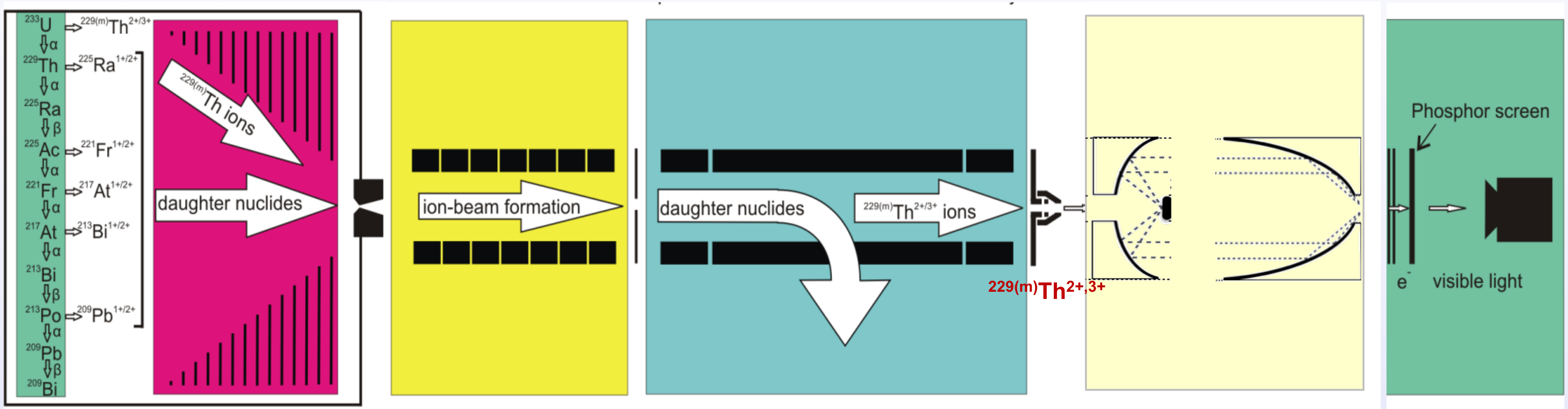


^{233}U source | **RF + DC funnel** | **RF quadrupole ion guide** | **Quadrupole mass separator**

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

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

→ 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

Experimental Setup

located at Maier-Leibnitz Laboratory, Garching: (recently moved to dedicated (laser-)laboratory)