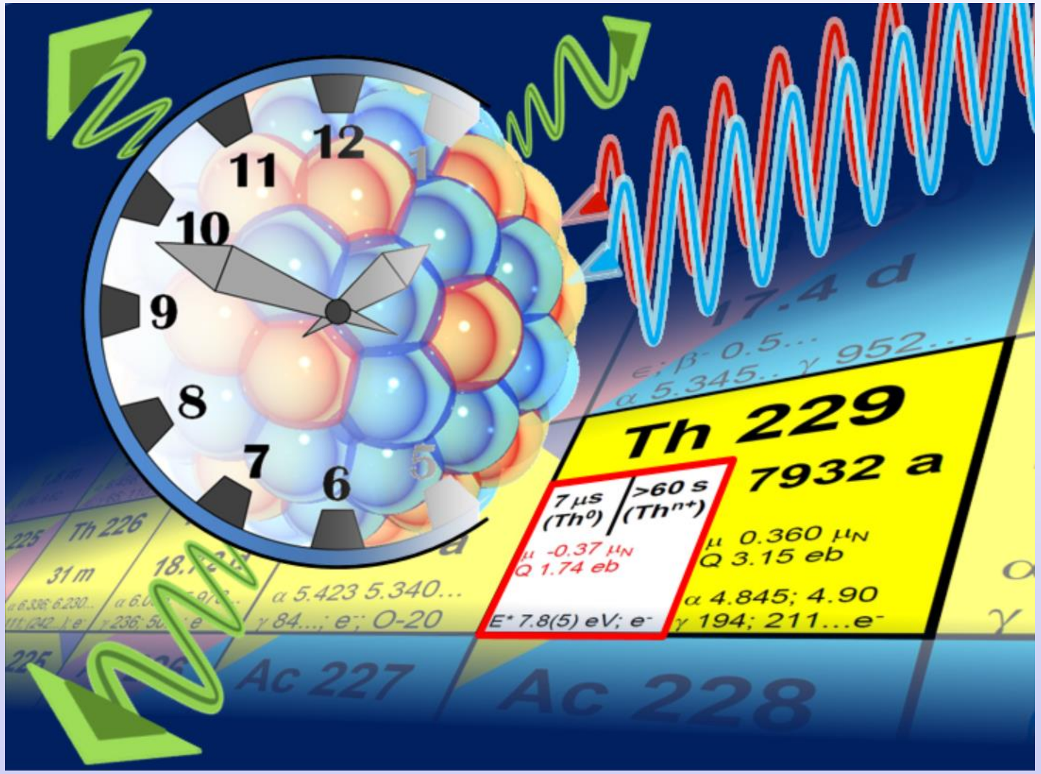
Towards a ^{229m}Th Nuclear Clock:

Status and Perspectives

Peter G. Thirolf, LMU München

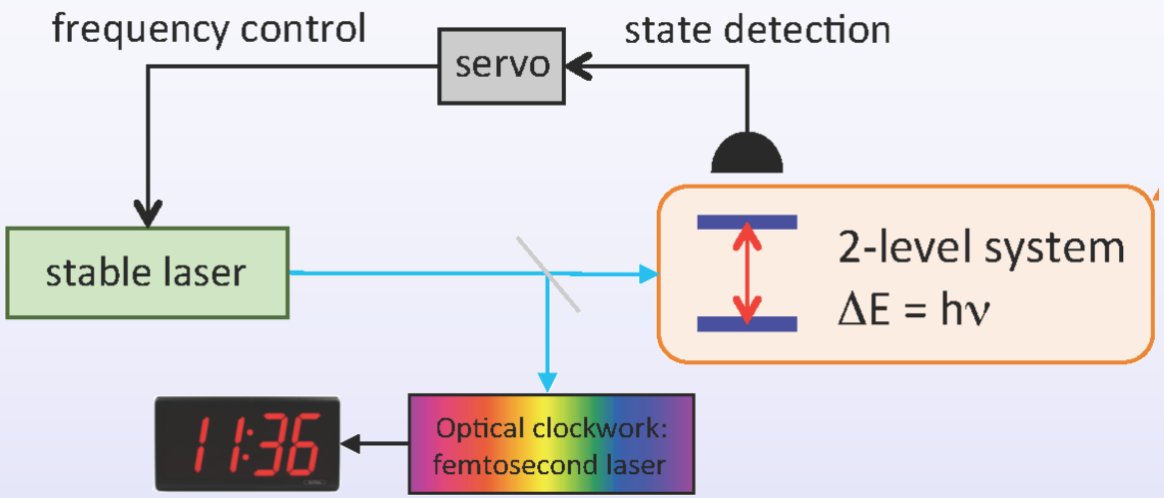
- Atomic Clock & Thorium Nuclear Clock
- Applications of a Nuclear Clock
- Knowledge on Thorium Isomer ^{229m}Th
  - direct (IC) decay, (neutral) $t_{1/2}$, HFS, E*
  - first observation of radiative decay
- Perspectives:
  - preparations for ionic lifetime measurement
  - development of VUV laser for direct spectroscopy
- Summary



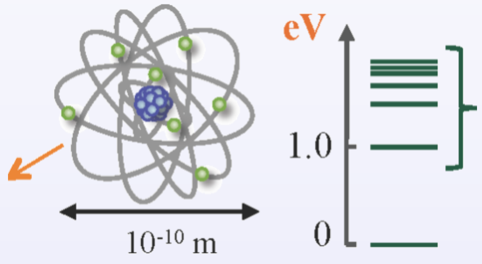
Thorium Nuclear Clock

scheme of an **atomic** clock

scheme of a **nuclear** clock

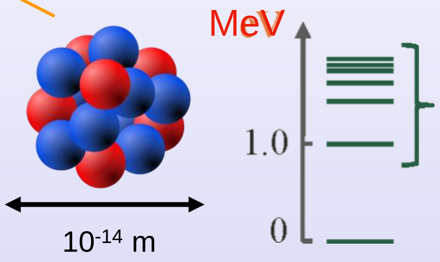


Nuclear clock proposal: E. Peik and Chr. Tamm, Europhys. Lett. 61, 181-186 (2003)
10^{-19} performance estimate of ^{229}Th ion clock: C. J. Campbell, et al., PRL 108, 120802 (2012)



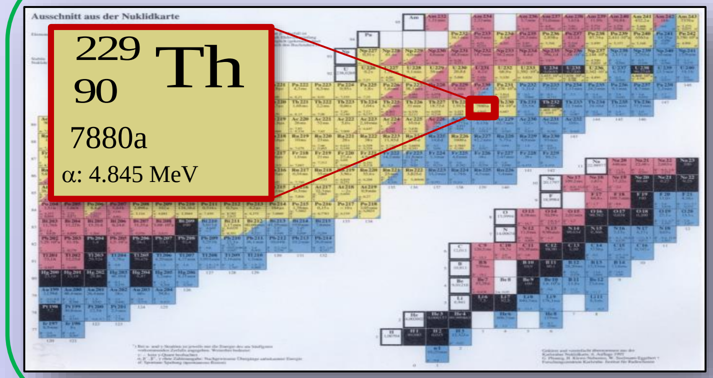
electronic excited states

ground state



nuclear excited states

ground state

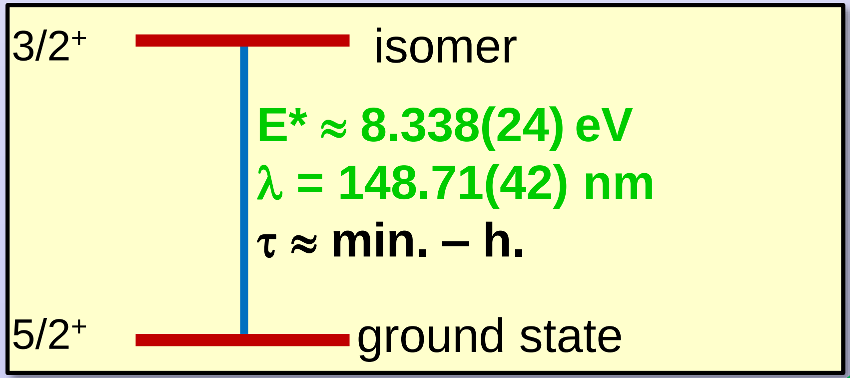


^{229m}Th properties:

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

ΔE/E ~ 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 + strong 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)

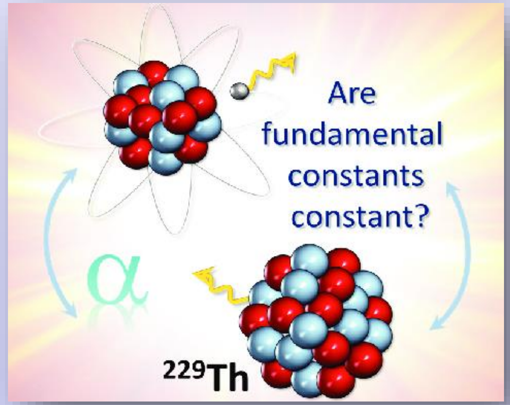
- **Temporal variation of fundamental constants**

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

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

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 …

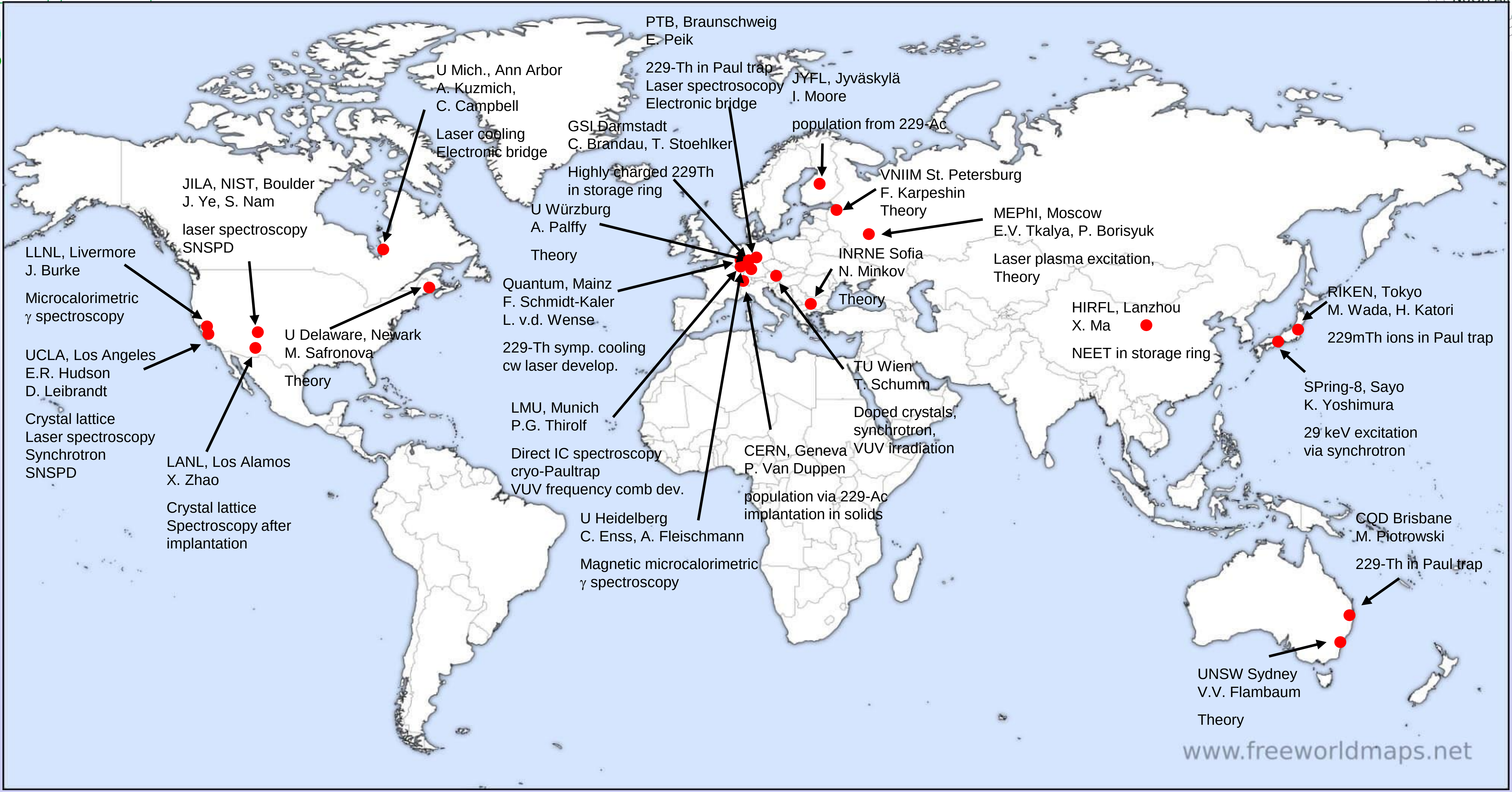
- **3D gravity sensor:** 'relativistic geodesy'
  - clock precision of 10^{-18}: detect gravitational shifts of ± 1 cm
  - precise, fast measurements of nuclear clock network:
    monitor volcanic magma chambers, tectonic plate movements

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

f: clock frequency
U: gravitat. potential

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

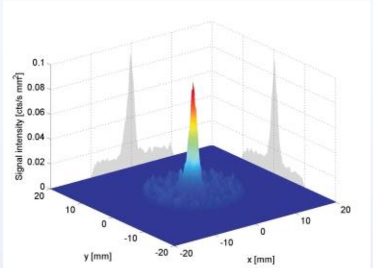
^{229m}Th research: worldwide activities



Characterization of ^{229m}Th since 2016

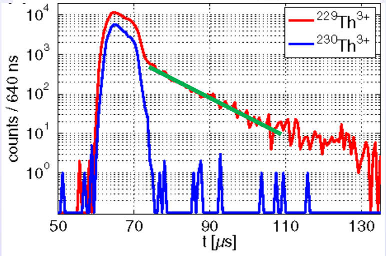
- **Internal Conversion decay:**

  First direct identification via Internal Conversion decay branch
  → electron detection (following α decay from ^{233}U) — Nature 533, 2016



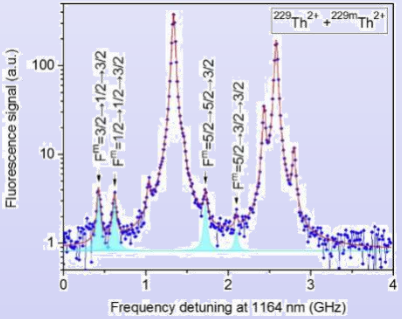
- **Isomer's Halflife:**

  neutral isomer: $t_{1/2}$ = 7±1 μs
  conversion coefficient: α_{IC} ~ 10^9 (in agreement with theory) — PRL 118, 2017



- **Hyperfine Structure:**

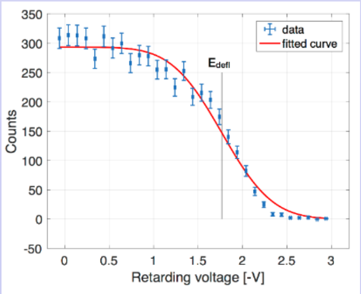
  collinear laser spectroscopy (LMU + PTB groups)
  → nuclear moments, charge radii — Nature 556, 2018



- **Isomeric excitation energy until 2022:**

  E*(iso) = 8.28 ± 0.17 eV (= 149.7 ± 3.1 nm) — Nature 573, 2019
  E*(iso) = 8.10 ± 0.17 eV (= 153.1 ± 3.7 nm) — PRL 125, 2020

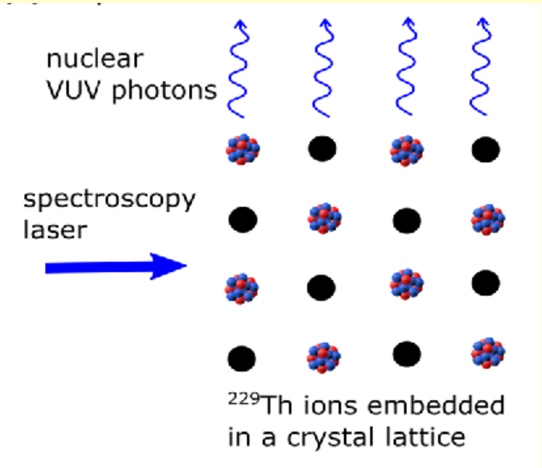
  → combined value 8.19 ± 0.12 eV (= 151.4 ± 2.2 nm)



Concepts for Direct Laser Spectroscopy of ^{229m}Th

Crystal lattice approach

- implant ^{229}Th nuclei in large-bandgap crystal
- IC forbidden if band gap > E^*_{iso}
- excite isomer with *VUV laser light*
- observe photons from isom. decay

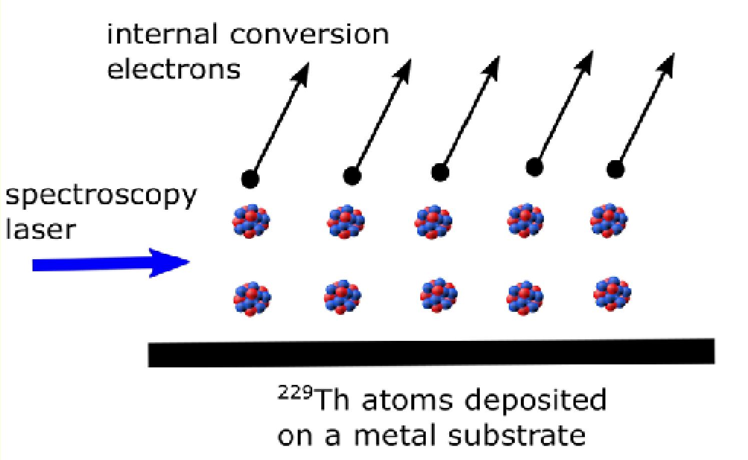


investigated by:

TU Vienna (Thorsten Schumm)
UCLA (Eric Hudson)

Solid surface approach

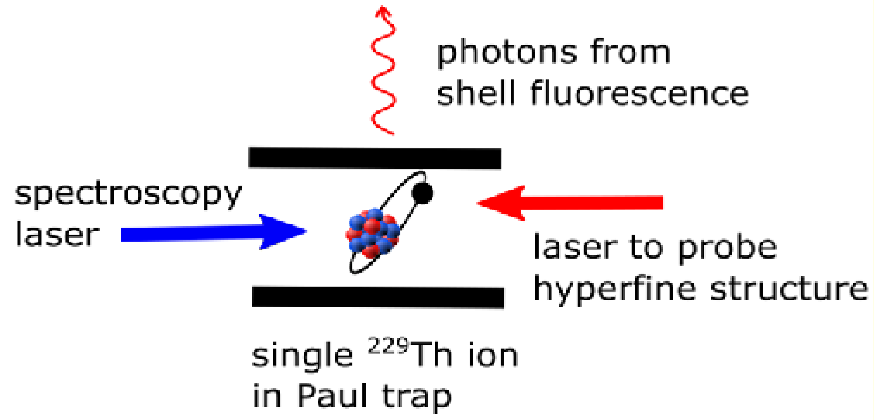
- deposit layer of ^{229}Th on surface
- IC allowed: band gap < E^*_{iso}
- excite isomer via *VUV laser light*
- observe electrons from isom. decay



JILA (Jun Ye)
UCLA (Eric Hudson)

Ion trap approach

- store ion(s) in Paul trap
- IC is forbidden (large IP)
- excite isomer via *VUV laser light*
- observe hyperfine shift of electron shell induced by nuclear spin change



PTB (Ekkehard Peik), LMU (Peter Thirolf)
U Mainz (F. Schmidt-Kaler)

L. v.d. Wense, B. Seiferle, EPJ-A 56, 277 (2020)

Missing: Radiative Decay

(prerequisite for solid-state nuclear clock)

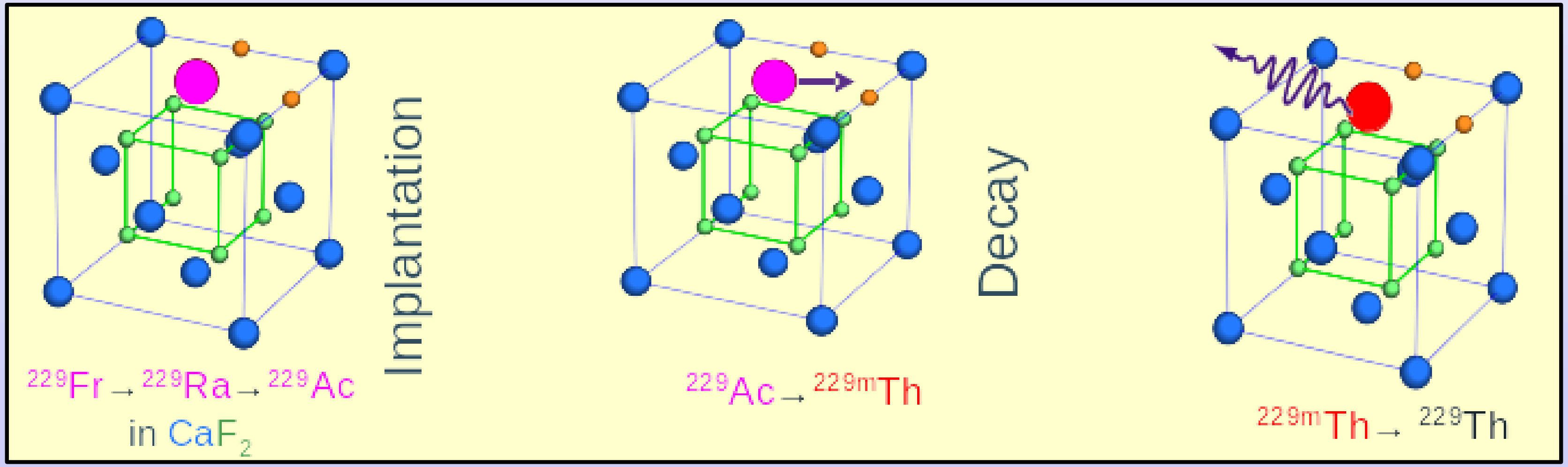
- **Photon spectroscopy of radioactive decay chains:**

  - 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

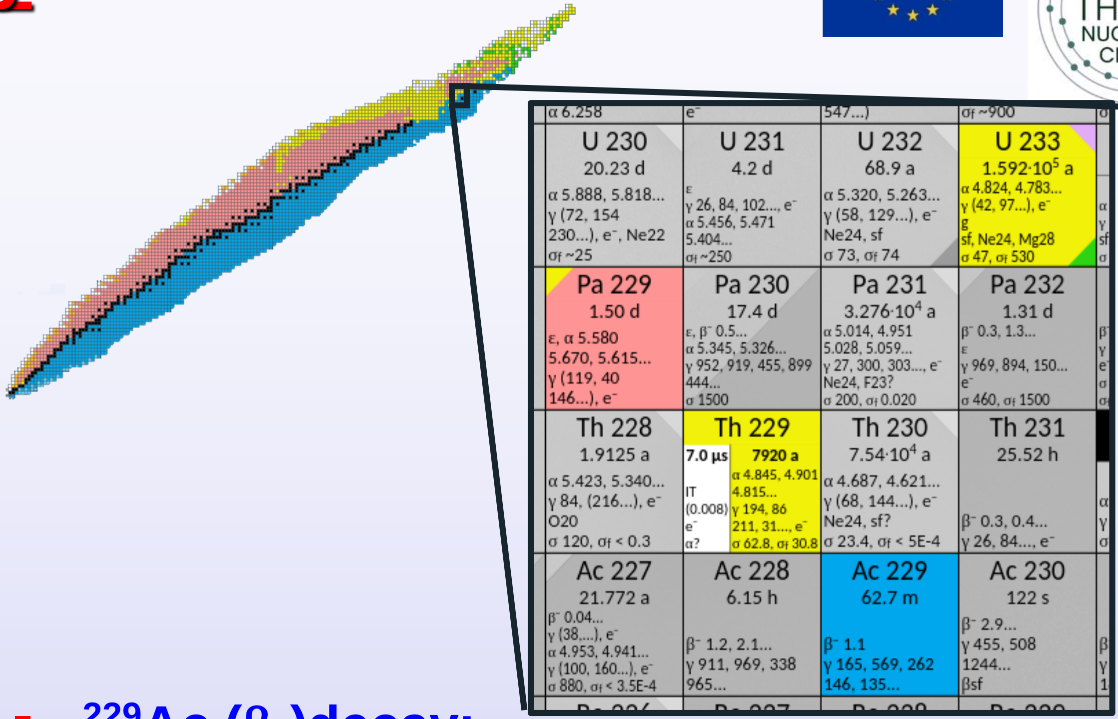
- **new approach:** using short-lived ^{229}Ac produced using ISOL technique (Isotope Production On-Line)



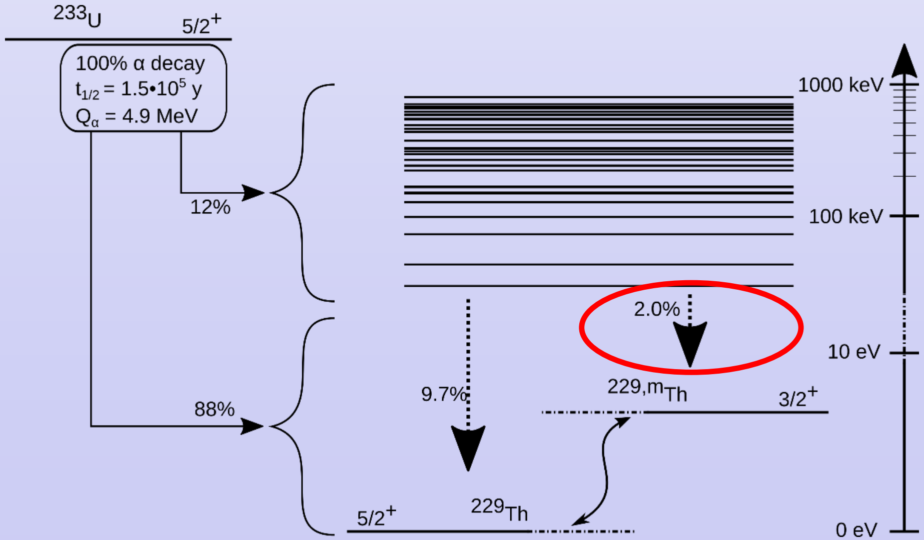
Exploit ^{229}Ac β decay

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

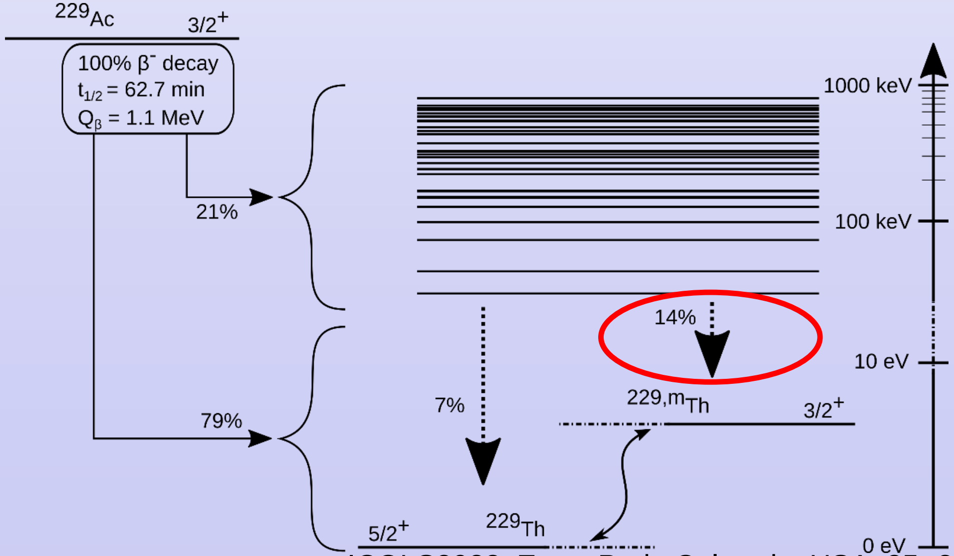
	^{233}U	^{229}Ac
BR	2%	14%
Decay	α	β^-
Recoil	84keV	<6eV
Production	stockpile	ISOL
Technique	doping	implantation



- **^{233}U (α-)decay:**

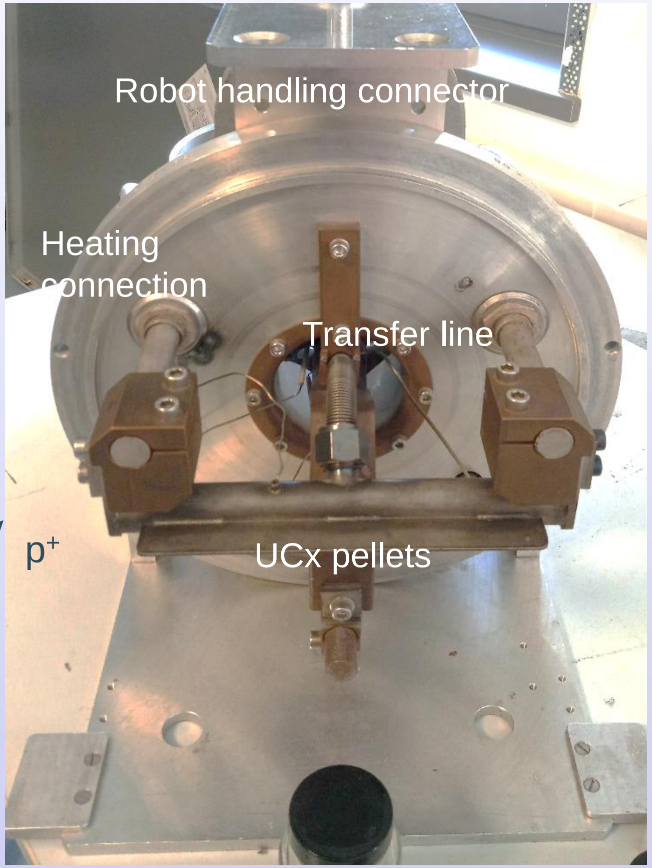


- **^{229}Ac (β-)decay:**



VUV spectroscopy at ISOLDE / CERN

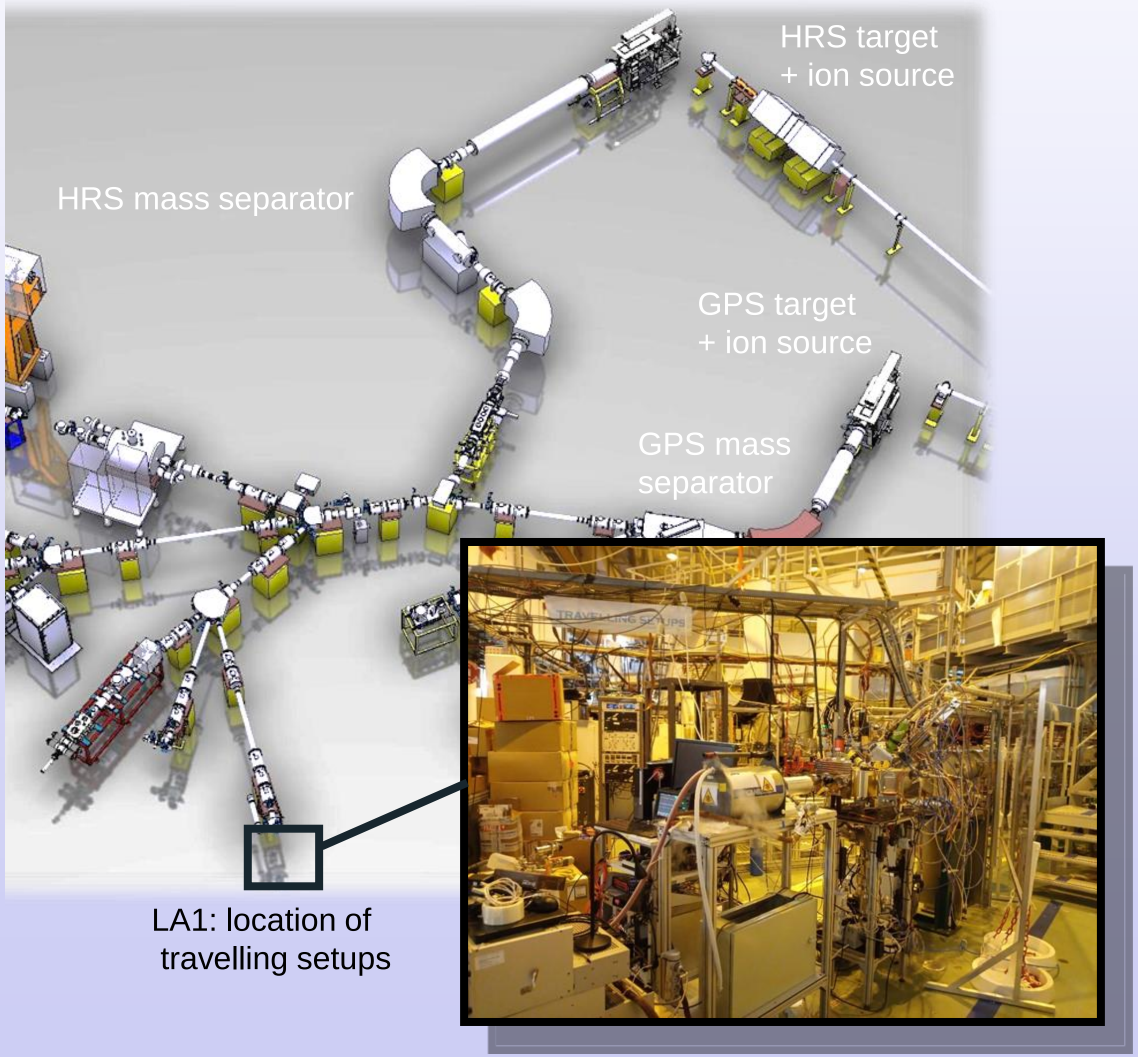
- **Production:** **1.4 GeV protons on UC_x**



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

$^{229}Fr \rightarrow$ $^{229}Ra \rightarrow$ $^{229}Ac \rightarrow$ $^{229m}Th/^{229}Th$

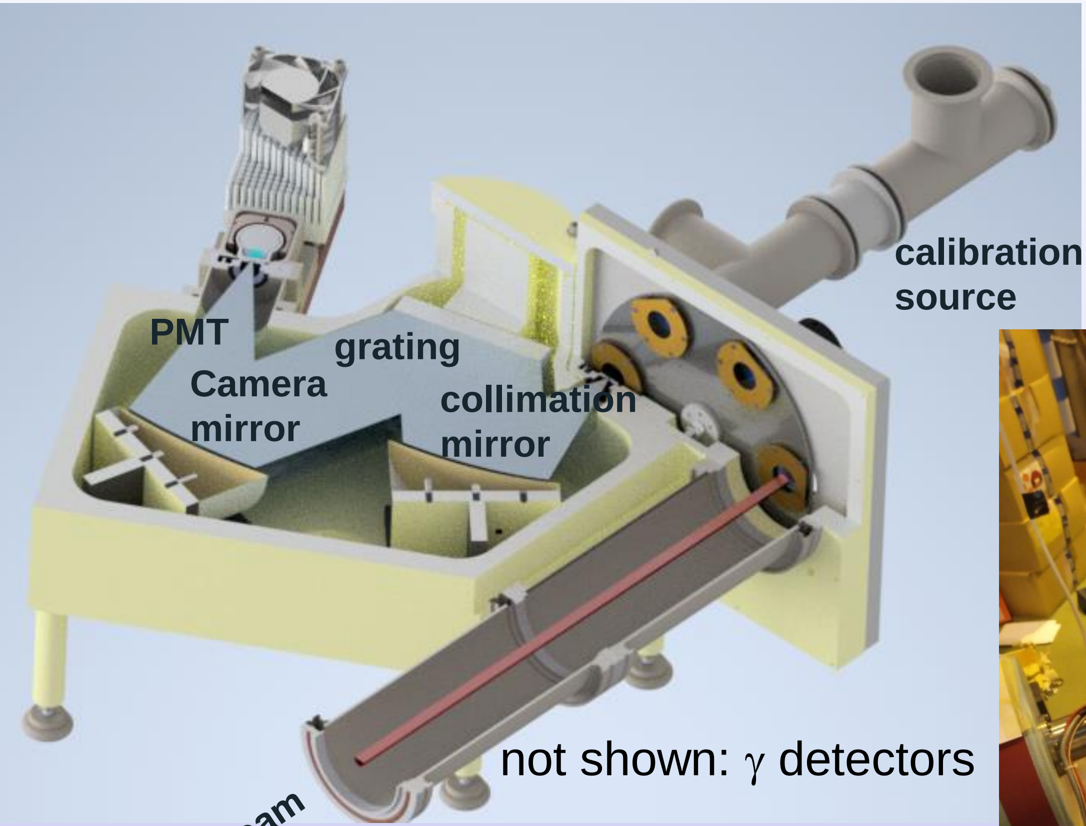
- **Beamline:** **ionization, mass separation, delivery**

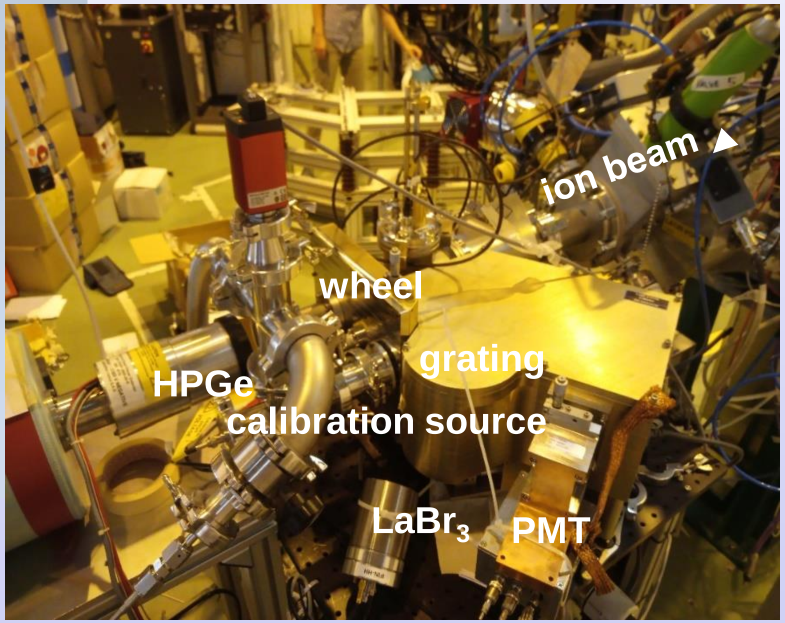


ISOLDE beamline sketch: CERN

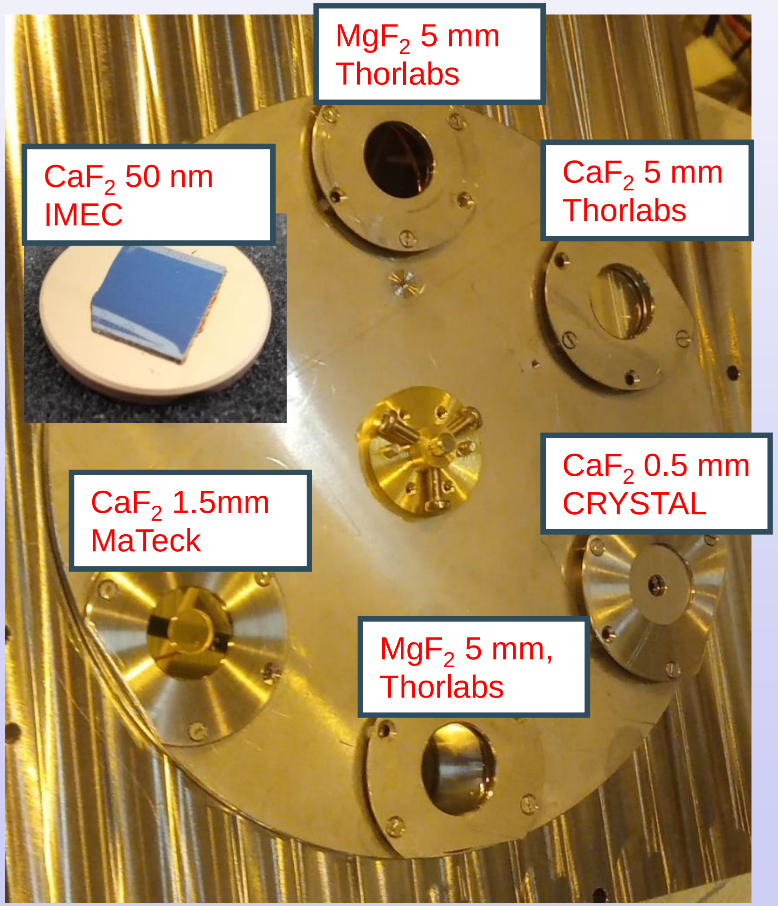
VUV spectroscopy at ISOLDE / CERN

VUV spectrometer:





various crystals on wheel:

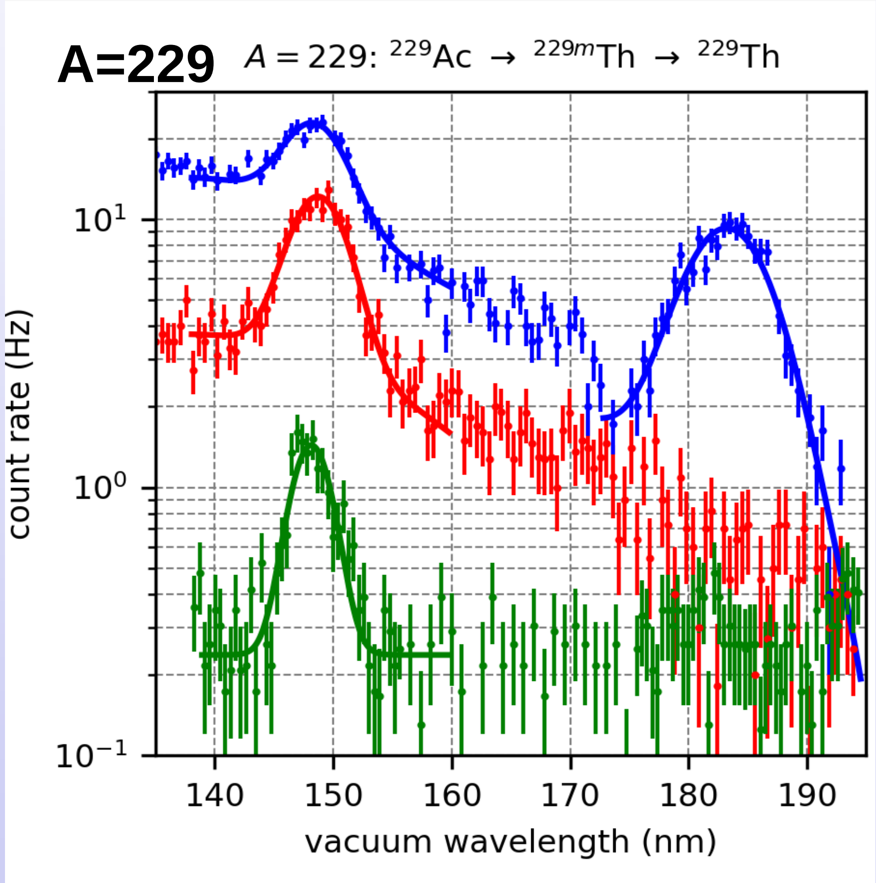


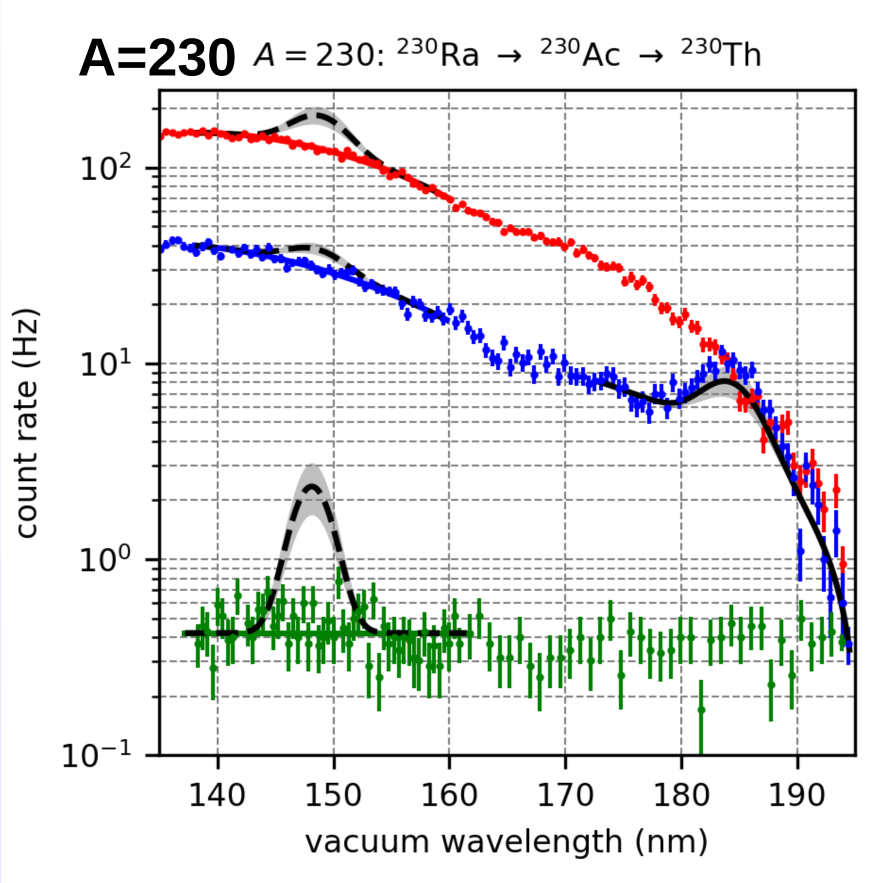
ISOLDE beam: ^{229}Fr, ^{229}Ra

VUV Spectroscopy Results

S. Kraemer et al., Nature 617, 706 (2023)

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





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Article | Published: 24 May 2023

Observation of the radiative decay of the ^{229}Th nuclear clock isomer

Sandro Kraemer, Janni Moens, Michail Athanasakis-Kaklamanakis, Silvia Bara, Kjeld Beeks, Premaditya Chhetri, Katerina Chrysalidis, Arno Claessens, Thomas E. Cocolios, João G. M. Correia, Hilde De Witte, Rafael Ferrer, Sarina Geldhof, Reinhard Heinke, Niyusha Hosseini, Mark Huyse, Ulli Köster, Yuri Kudryavtsev, Mustapha Laatiaoui, Razvan Lica, Goele Magchiels, Vladimir Manea, Clement Merckling, Lino M. C. Pereira, Sebastian Raeder, Thorsten Schumm, Simon Sels, Peter G. Thirolf, Shandirai Malven Tunhuma, Paul Van Den Bergh, Piet Van Duppen, André Vantomme, Matthias Verlinde, Renan Villarreal & Ulrich Wahl

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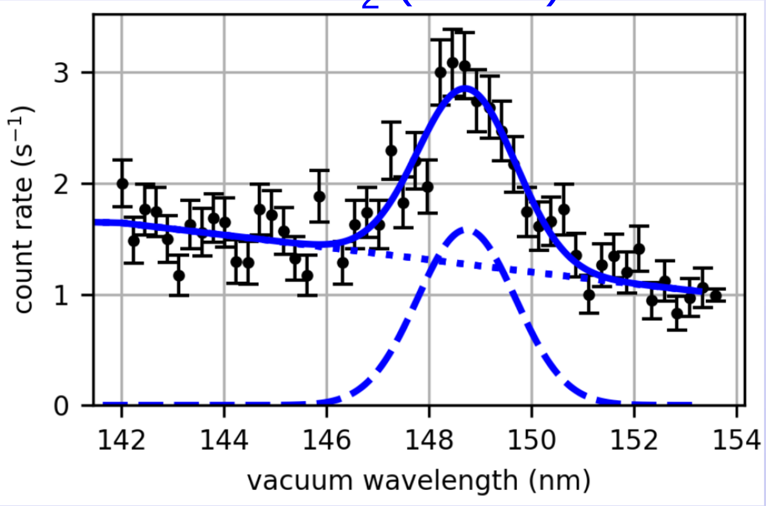
CaF_2 (5 mm)

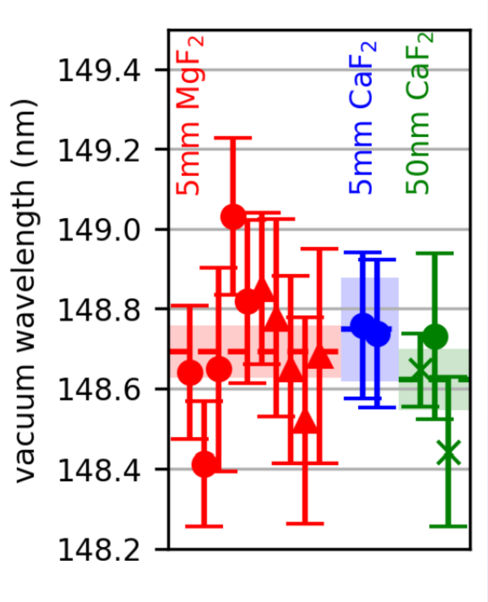


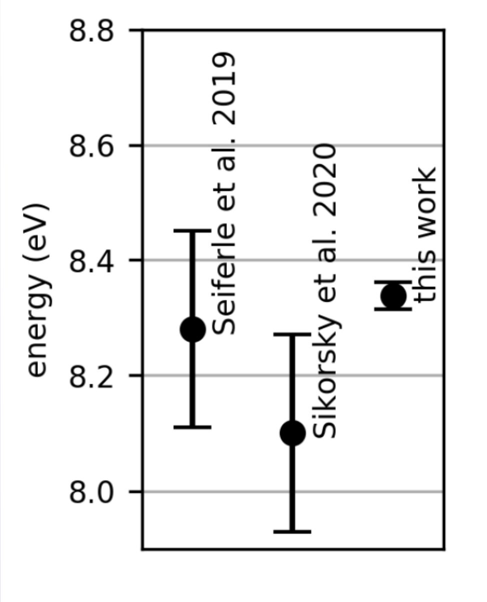
3 mm spectrometer entrance slit

0.5 mm entrance slit

VUV Spectroscopy Results

excitation energy/ emission wavelength:





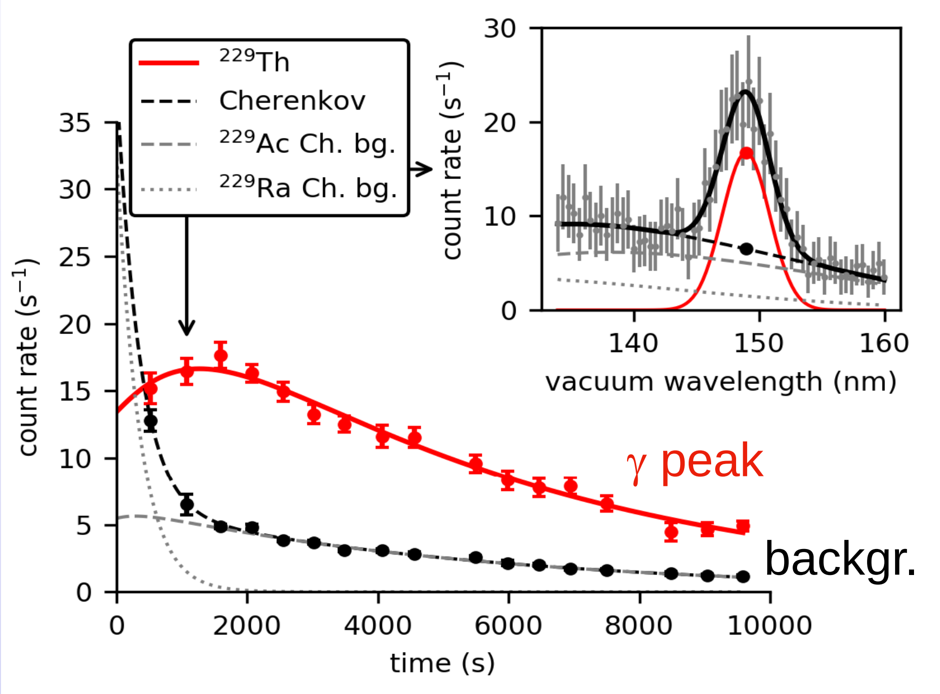
8.338 ± 0.003(stat.) ± 0.023(syst.) eV

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 with n^3 scaling: n ~1.55 (@ 148 nm): $t_{1/2}$~2500 s
- direct $t_{1/2}$ measurement in cryo-Paultrap in preparation (LMU)

S. Kraemer et al., Nature 617, 706 (2023)

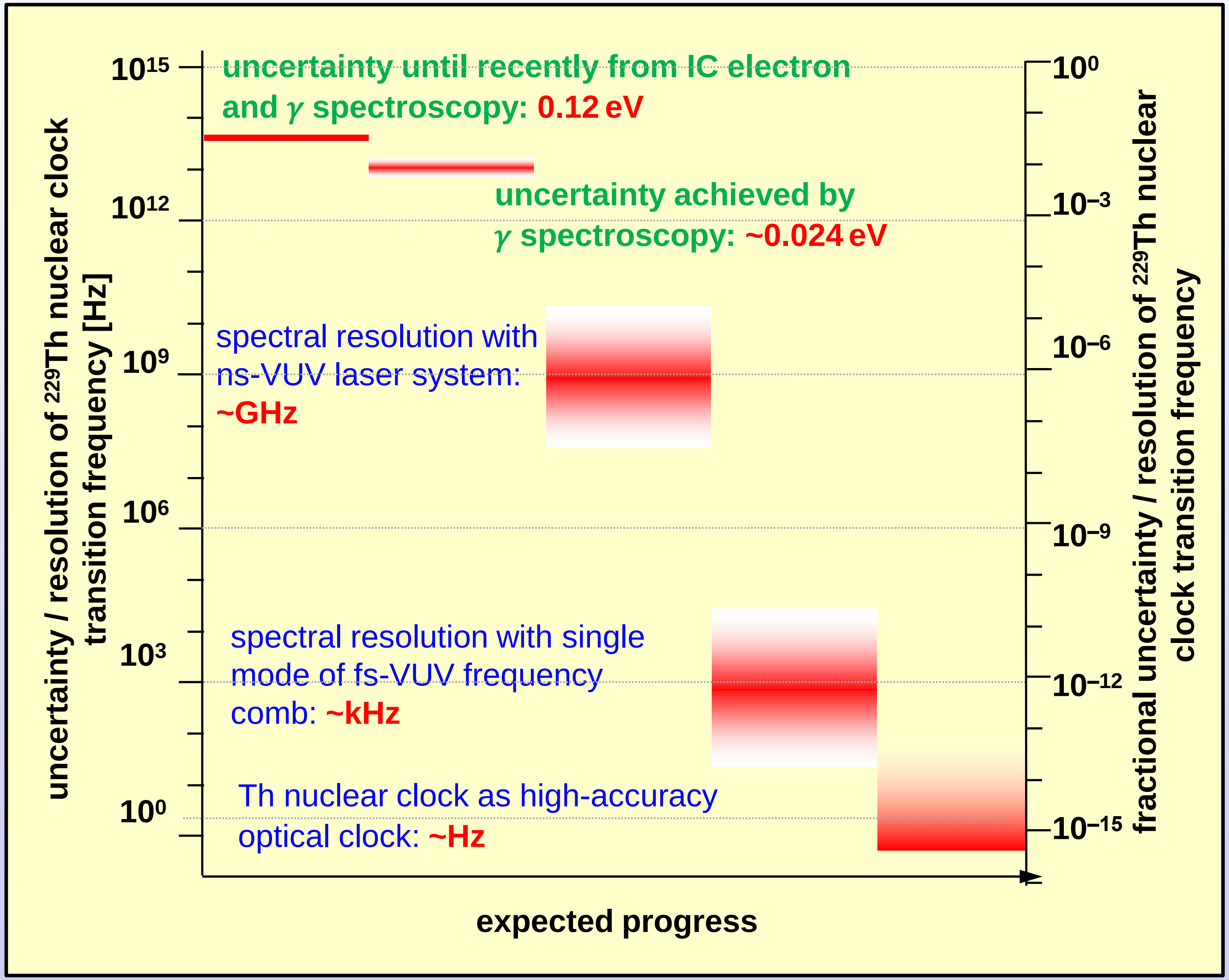
Perspectives for the Nuclear Clock

- **still to bridge: 10-12 orders of magnitude:**

heading from nuclear to optical domain:
" 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.)

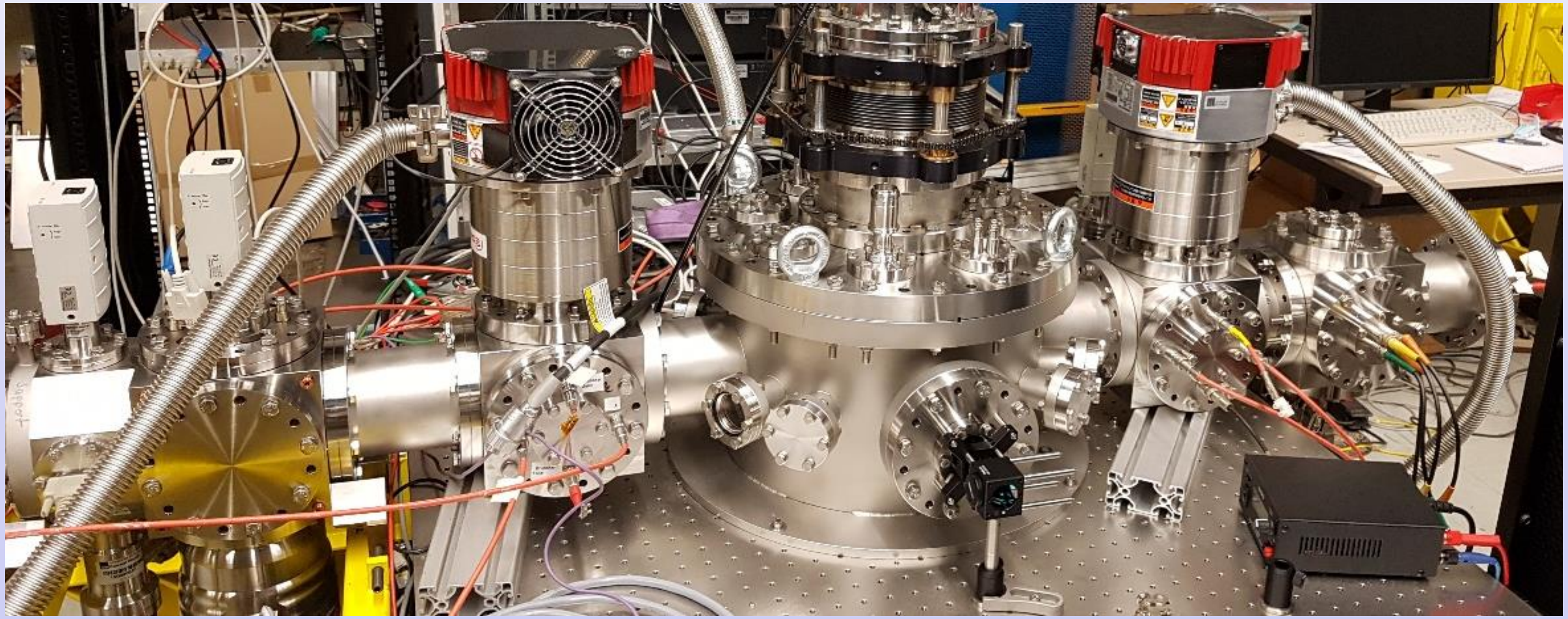
- (VUV frequency comb) laser under development
- ultimate goal: narrow-band cw laser

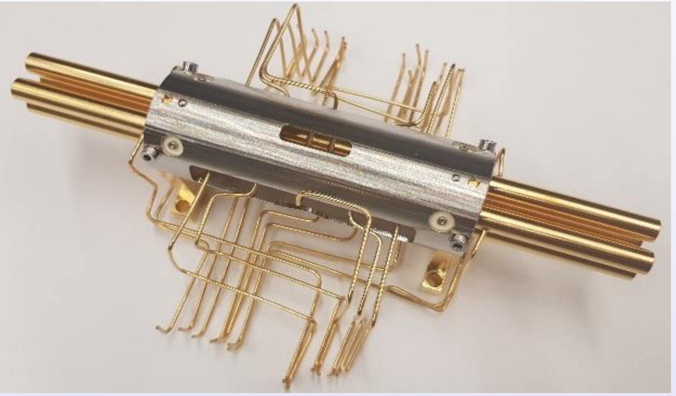


Ionic Lifetime Measurement

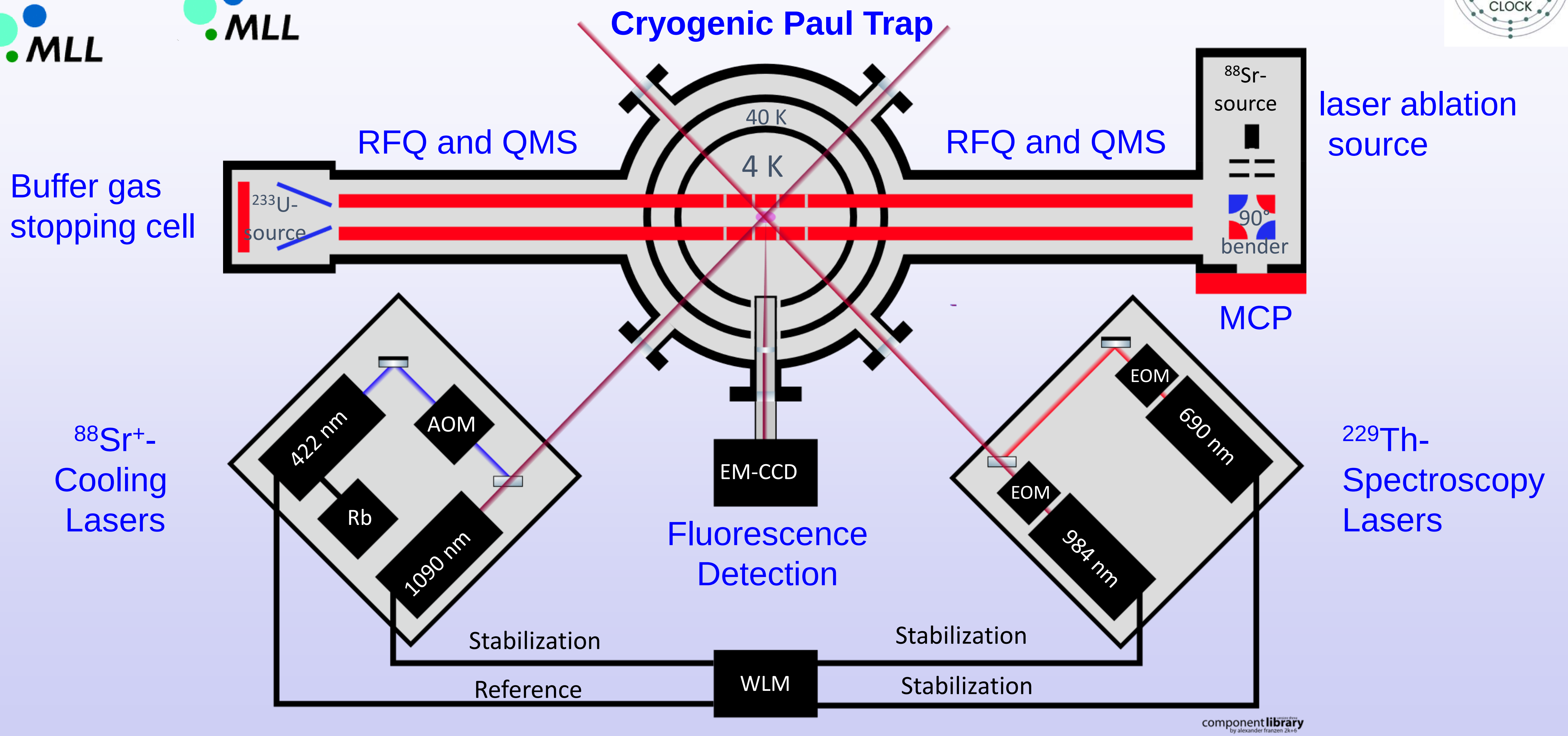
needs longer storage time (= better vacuum)

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





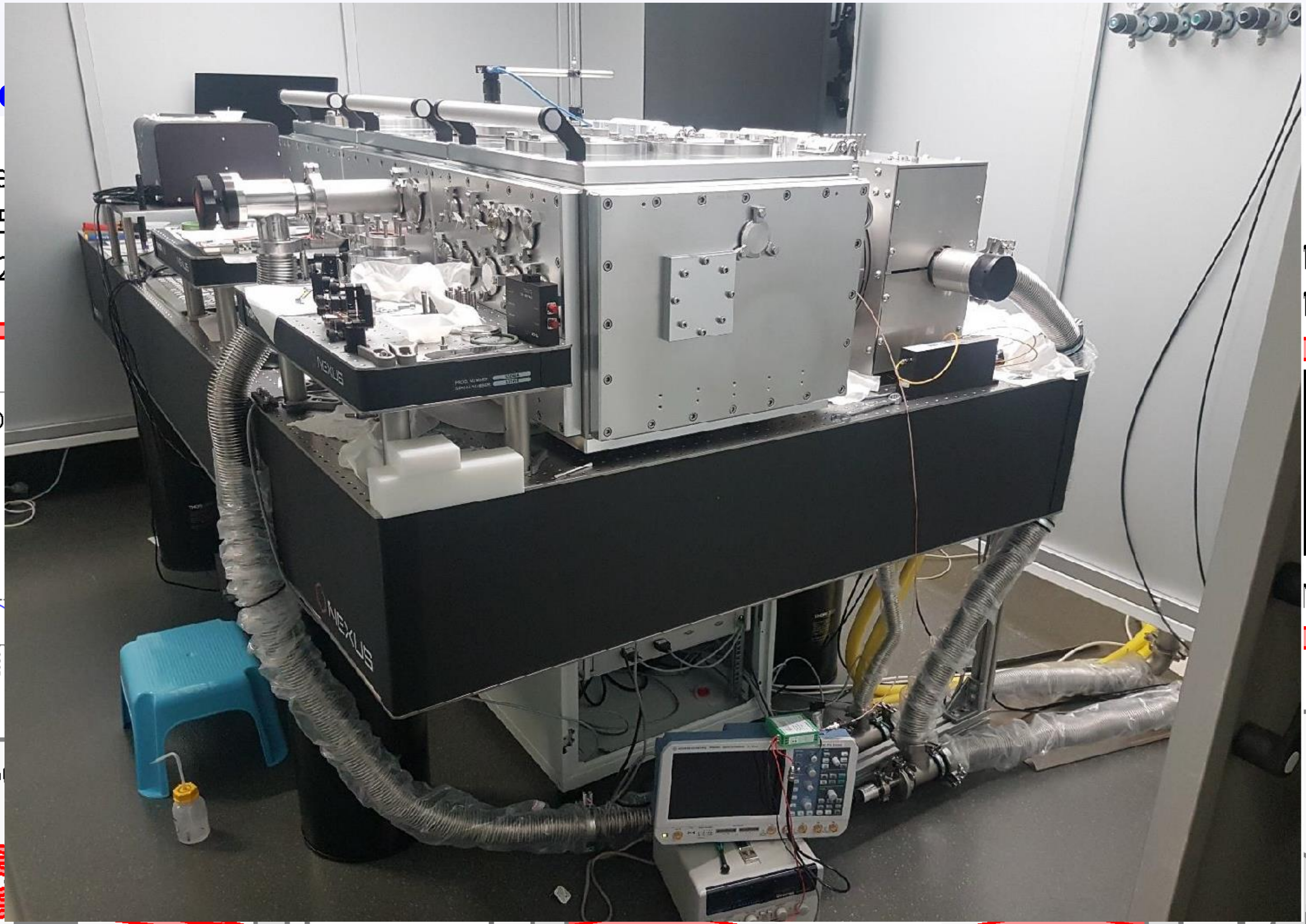
Cryo Trap & Laser Setup



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

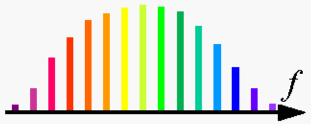
VUV laser source for ^{229m}Thorium

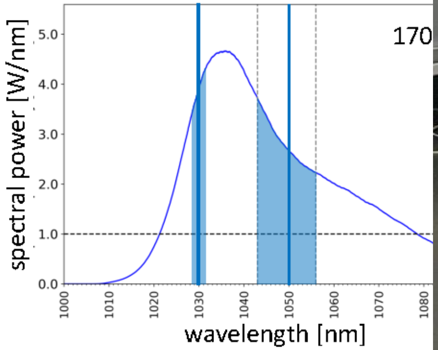
- VUV frequency co



frequency comb

$\nu_{rep} = 80$ MHz





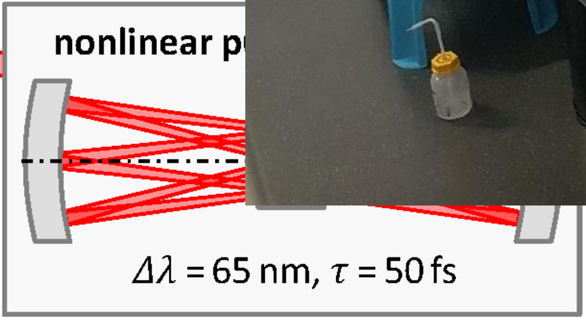
se
35
12

IW @40 MHz
IW @80 MHz

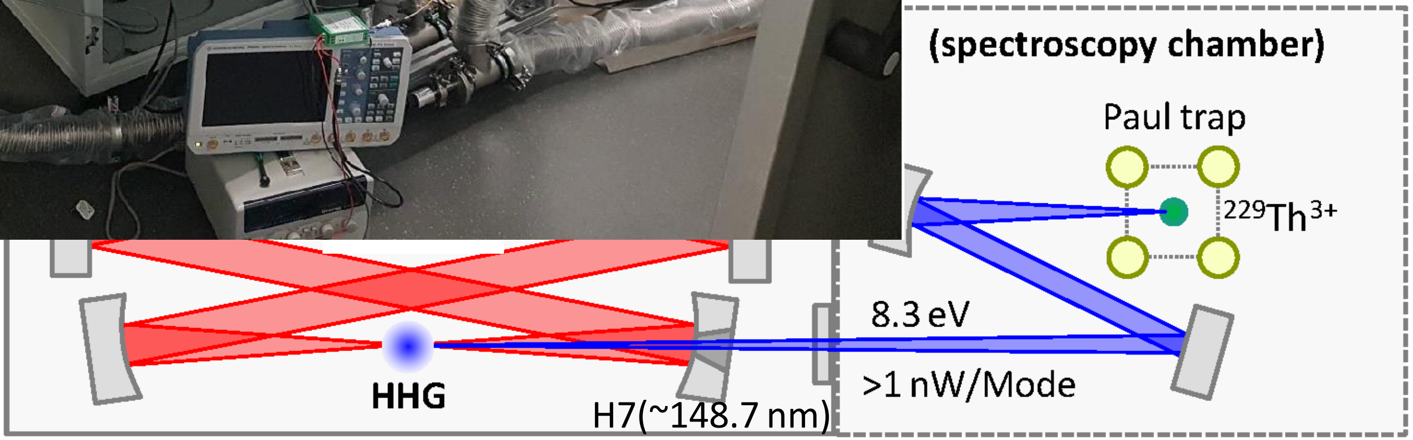
laser under development at Fraunhofer ILT (Aachen)
operational: early 2024

MW @40 MHz

peak power 9-16 MW



compression factor 5-15 (2 stages)

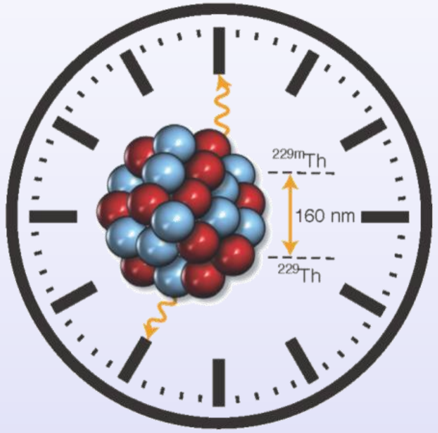


J. Weitenberg, ILT Fraunhofer/ RWTH Aachen & MPQ Garching

Summary

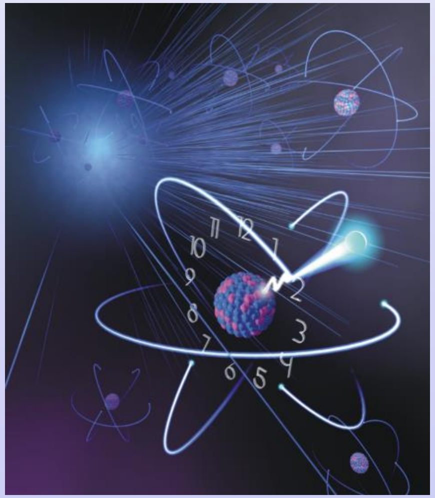
Exp. achievements in recent years:

- 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 ($\dot{\alpha}$)
- search for dark matter candidates not accessible by any other means

Thanks to ….

LMU: **K. Scharl, D. Moritz, S. Kraemer, I. Hussain, T. Rozibakieva, L. Löbell,**
F. Zacherl, L. v.d. Wense, B. Seiferle, **G. Holthoff**

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

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

MPQ/ILT Aachen: ***J. Weitenberg***, S. Wissemberg

MPI-HD: P. Bilous, N. Minkov, J. Crespo
U Würzburg: ***A. Pàlffy***
NIST: S. Nam, G. O'Neil
UCLA: E. Hudson, C. Schneider, J. Jeet
U Delaware: ***M. Safronova***

Thank you for your attention !

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