The Thorium Isomer ^{229m}Th:

From the Atomic to the Nuclear Clock

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
  - recent developments
- Perspectives:
  - preparations for ionic lifetime measurement
  - development of VUV laser for direct spectroscopy
- Summary

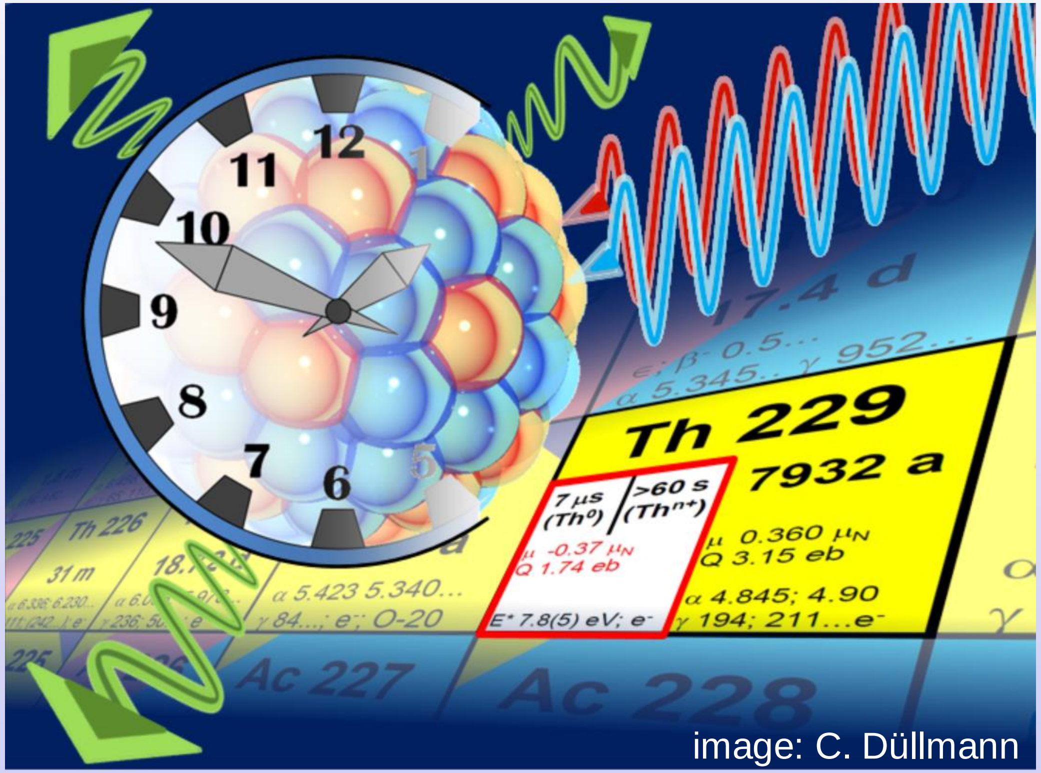
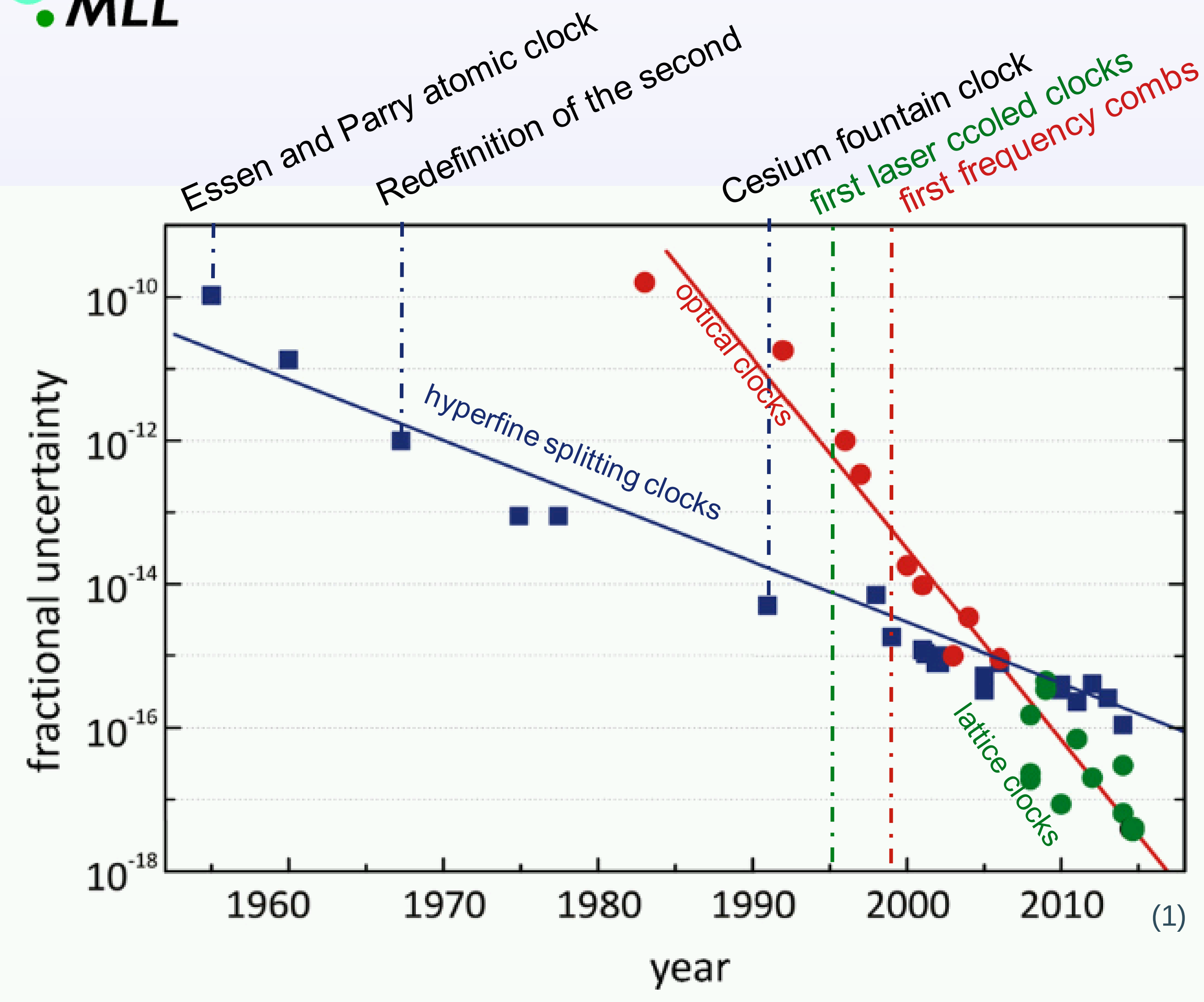


image: C. Düllmann

Clock Accuracy



(1) mod. from Riehle, *C.R. Physiques* 16, 506 (2016)

- **Limitations:**

external pertubations:
- Stark & Zeeman shifts
- second order Doppler
- black-body radiation

fundamental limit:

quantum projection noise

$$\boxed{\sigma_y(\tau) = \kappa \frac{\Delta\nu}{\nu} \sqrt{\frac{T_{int}}{N\,\tau}}}$$

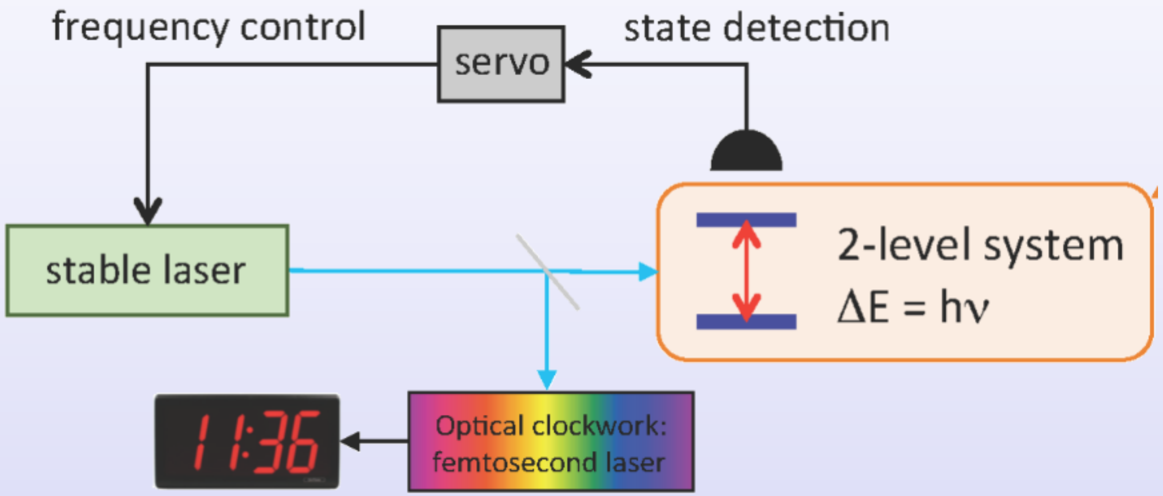
- **Best atomic clocks:**

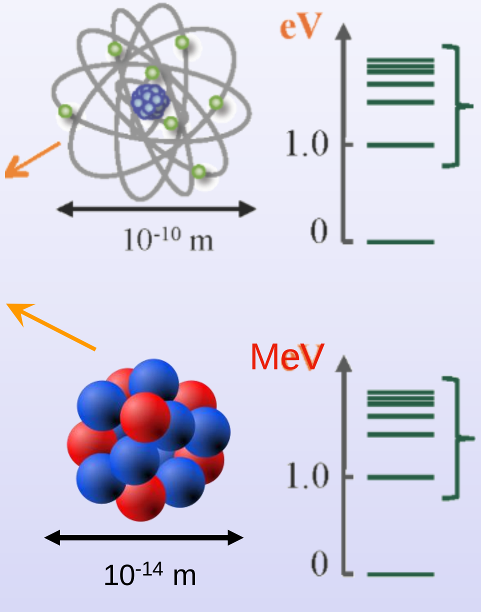
Ytterbium E3 single-ion clock :	$3.2 \cdot 10^{-18}$
N. Huntemann et al., PRL 116 (2016)	
Strontium optical lattice clock:	$2.1 \cdot 10^{-18}$
T.L. Nicholson et al., Nature Comm. (2015)	
Ytterbium optical lattice clock:	$1.4 \cdot 10^{-18}$
W.F. McGrew et al., Nature 5634 (2018)	
Aluminum single-ion clock:	$9.4 \cdot 10^{-19}$
S.M. Brewer et al., PRL 123 (2019)	

achieved accuracy: 1 s in 30 billion years

Thorium Nuclear Clock

scheme of an
atomic clock





electronic
excited
states

ground
state

nuclear
excited
states

ground
state

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)

Is there a suitable nuclear candidate ?

→ low energy (eV)
→ long-lived (isomer)

The ‘Thorium Isomer’ ^{229m}Th: Chasing a Phantom ?

(355 ± 20 nm)

(160 ± 10 nm)

1976: < 100 eV

1990: -1 ± 4 eV

1994: 3.5 ± 1 eV

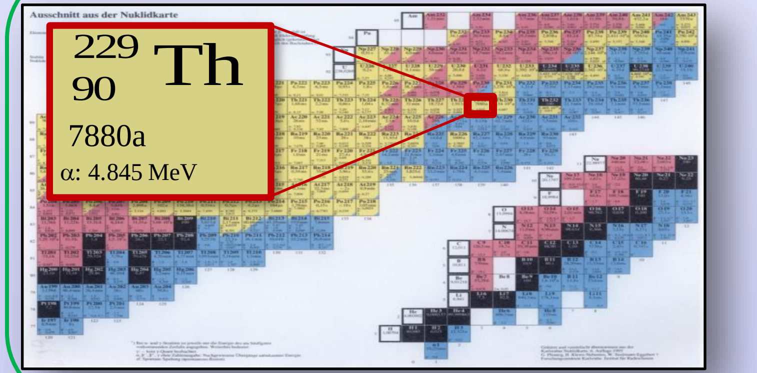
2005: 5.5 ± 1 eV

2007: 7.6 ± 0.5 eV

2009: 7.8 ± 0.5 eV

?

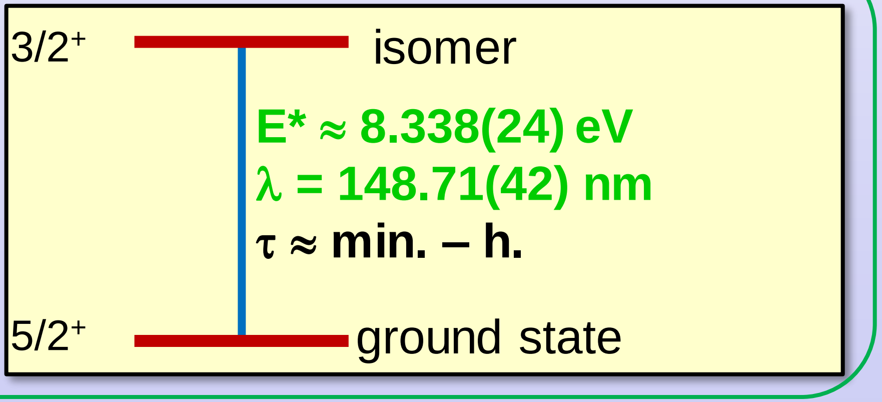
← Indirect Evidence →



^{229m}Th properties:

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

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



$E^* \approx 8.338(24)$ eV
$\lambda = 148.71(42)$ nm
$\tau \approx$ min. – h.

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:**

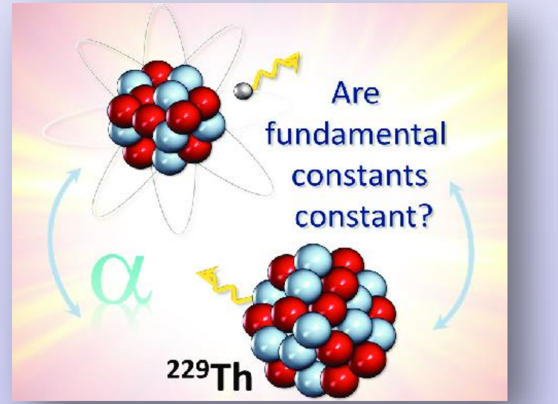
E. Peik, PT et al., Quant. Sci. Tech. 6, 034002 (2021)

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

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

$\dot{\alpha}/\alpha = (1.0 \pm 1.1) \cdot 10^{-18}\,\mathrm{yr}^{-1}$ R. Lange et al., PRL 126, 011102 (2021)

$\dot{\alpha}/\alpha = (1.8 \pm 2.5) \cdot 10^{-19}\,\mathrm{yr}^{-1}$ M. Filzinger et al., PRL 130, 253001 (2023)



- 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 …

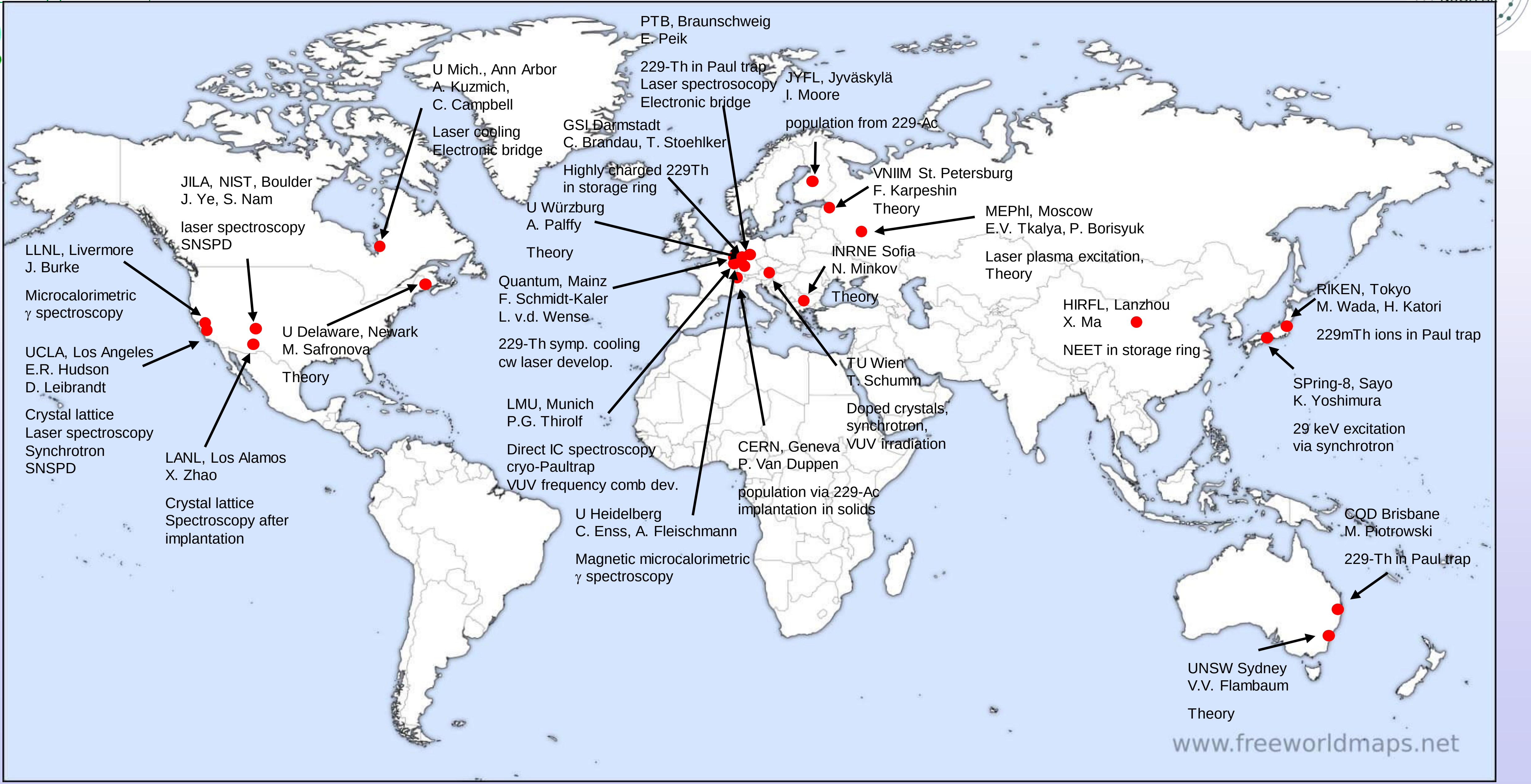
- **3D gravity sensor:** 'relativistic geodesy'
  - clock precision of 10^{-18}: detect gravitational shifts of ± 1 cm
  - precise, fast measurements of nuclear clock network:
    monitor volcanic magma chambers, tectonic plate movements

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

f: clock frequency
U: gravitat. potential

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

^{229m}Th research: worldwide activities



LLNL, Livermore
J. Burke

Microcalorimetric
γ spectroscopy

UCLA, Los Angeles
E.R. Hudson
D. Leibrandt

Crystal lattice
Laser spectroscopy
Synchrotron
SNSPD

JILA, NIST, Boulder
J. Ye, S. Nam

laser spectroscopy
SNSPD

LANL, Los Alamos
X. Zhao

Crystal lattice
Spectroscopy after
implantation

U Mich., Ann Arbor
A. Kuzmich,
C. Campbell

Laser cooling
Electronic bridge

U Delaware, Newark
M. Safronova

Theory

GSI Darmstadt
C. Brandau, T. Stoehlker

Highly charged 229Th
in storage ring

U Würzburg
A. Palffy

Theory

Quantum, Mainz
F. Schmidt-Kaler
L. v.d. Wense

229-Th symp. cooling
cw laser develop.

LMU, Munich
P.G. Thirolf

Direct IC spectroscopy
cryo-Paultrap
VUV frequency comb dev.

U Heidelberg
C. Enss, A. Fleischmann

Magnetic microcalorimetric
γ spectroscopy

PTB, Braunschweig
E. Peik

229-Th in Paul trap
Laser spectroscopy
Electronic bridge

JYFL, Jyväskylä
I. Moore

population from 229-Ac

VNIIM St. Petersburg
F. Karpeshin
Theory

MEPhI, Moscow
E.V. Tkalya, P. Borisyuk

Laser plasma excitation,
Theory

INRNE Sofia
N. Minkov

Theory

TU Wien
T. Schumm

Doped crystals,
synchrotron,
VUV irradiation

CERN, Geneva
P. Van Duppen

population via 229-Ac
implantation in solids

HIRFL, Lanzhou
X. Ma

NEET in storage ring

RIKEN, Tokyo
M. Wada, H. Katori

229mTh ions in Paul trap

SPring-8, Sayo
K. Yoshimura

29 keV excitation
via synchrotron

CQD Brisbane
M. Piotrowski

229-Th in Paul trap

UNSW Sydney
V.V. Flambaum

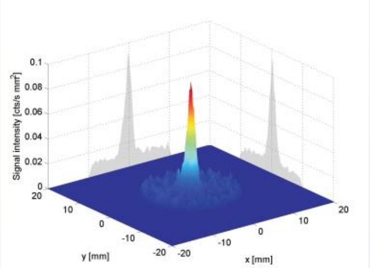
Theory

Characterization of ^{229m}Th 2016-2020

Internal Conversion decay:

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

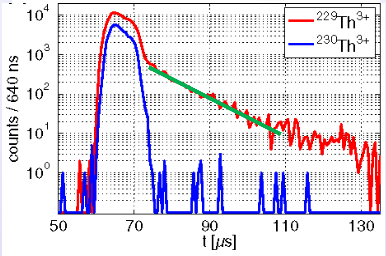
Nature 533, 2016



Isomer's Halflife:

neutral isomer: $t_{1/2} = 7 \pm 1\ \mu s$
conversion coefficient: $\alpha_{IC} \sim 10^9$ (in agreement with theory)

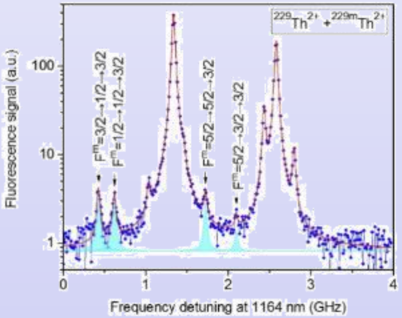
PRL 118, 2017



Hyperfine Structure:

collinear laser spectroscopy (LMU + PTB groups)
→ nuclear moments, charge radii

Nature 556, 2018



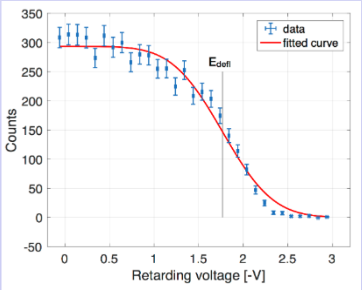
Isomeric excitation energy until 2022:

E*(iso) = 8.28 ± 0.17 eV (= 149.7 ± 3.1 nm)
E*(iso) = 8.10 ± 0.17 eV (= 153.1 ± 3.7 nm)

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

Nature 573, 2019

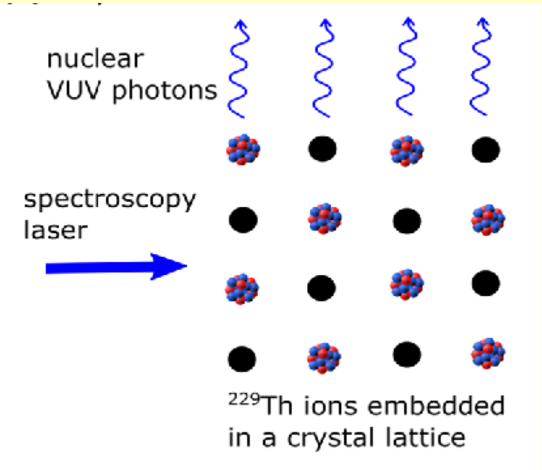
PRL 125, 2020



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

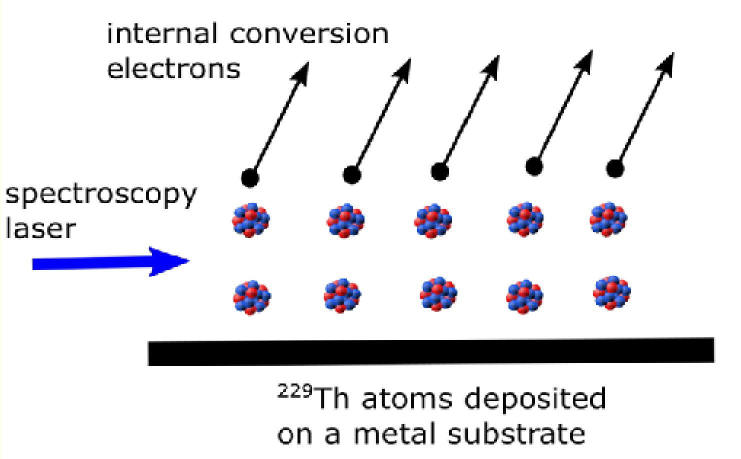


investigated by:

TU Vienna (Thorsten Schumm),
PTB (E. Peik), UCLA (Eric Hudson)

Solid surface approach

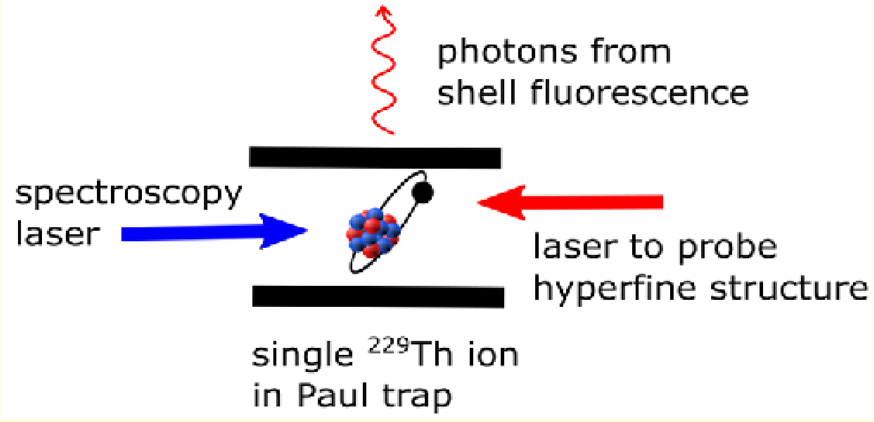
- deposit layer of ^{229}Th on surface
- IC allowed: band gap < E^*_{iso}
- excite isomer via *VUV laser light*
- observe electrons from isom. decay



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)

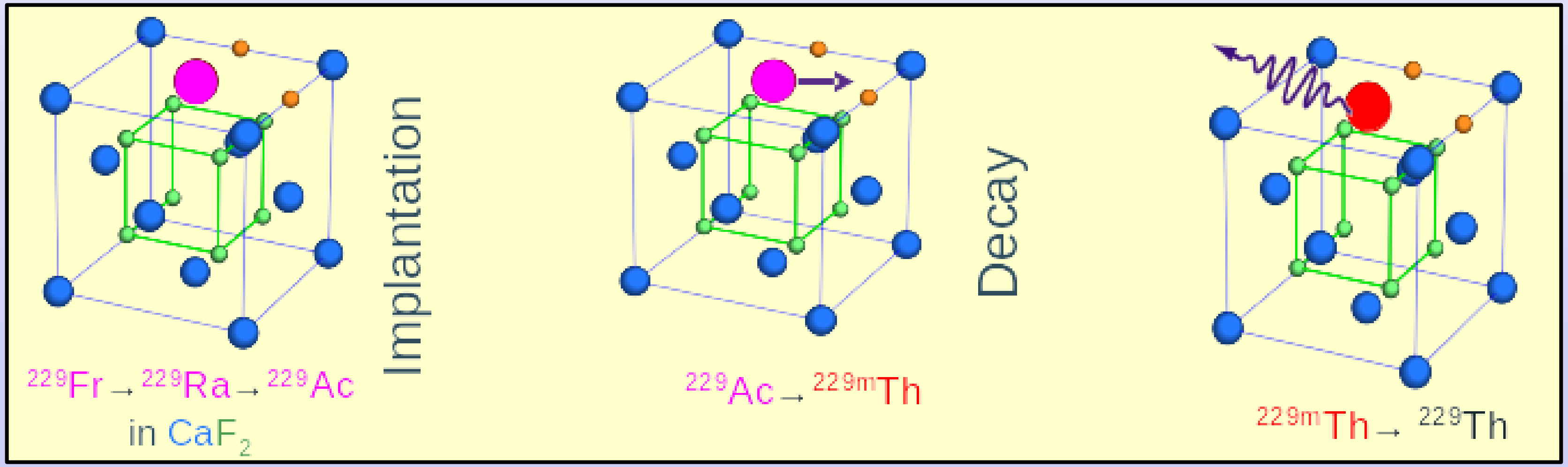
- **Photon spectroscopy of radioactive decay chains:**

  - 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

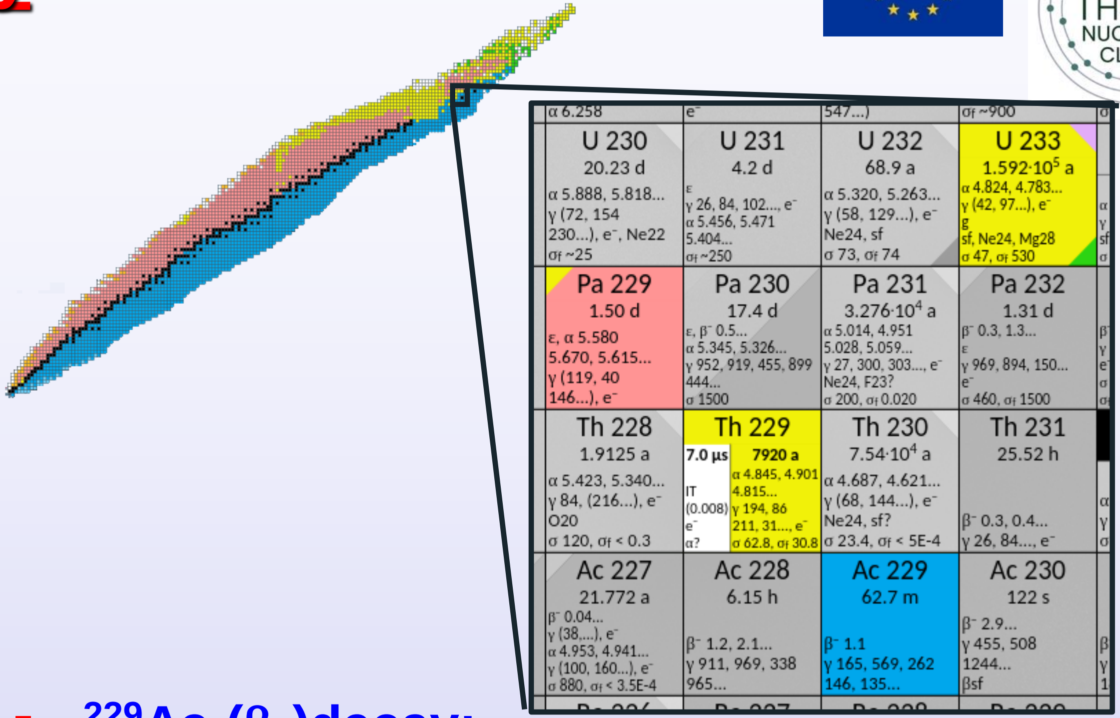
- **So far: experimental efforts using the alpha-decay of ^{233}U**

→ observation of radiative decay for decades unsuccessful

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



Exploit ^{229}Ac β decay

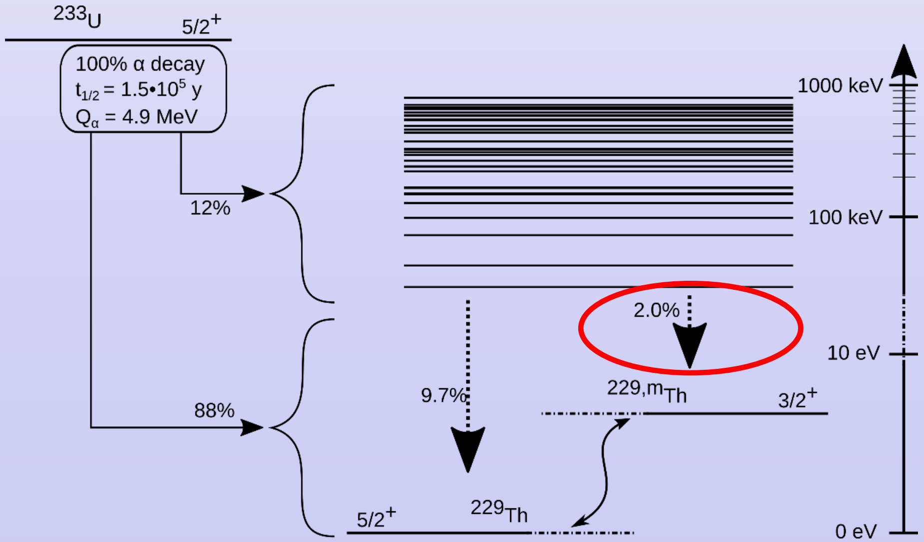


α 6.258	e⁻	547...)	σf ~900
U 230 20.23 d α 5.888, 5.818... γ (72, 154 230...), e⁻, Ne22 σf ~25	**U 231** 4.2 d ε γ 26, 84, 102..., e⁻ α 5.456, 5.471 5.404... σf ~250	**U 232** 68.9 a α 5.320, 5.263... γ (58, 129...), e⁻ Ne24, sf σ 73, σf 74	**U 233** $1.592 \cdot 10^5$ a α 4.824, 4.783... γ (42, 97...), e⁻ g sf, Ne24, Mg28 σ 47, σf 530
Pa 229 1.50 d ε, α 5.580 5.670, 5.615... γ (119, 40 146...), e⁻	**Pa 230** 17.4 d ε, β⁻ 0.5... α 5.345, 5.326... γ 952, 919, 455, 899 444... σ 1500	**Pa 231** $3.276 \cdot 10^4$ a α 5.014, 4.951 5.028, 5.059... γ 27, 300, 303..., e⁻ Ne24, F23? σ 200, σf 0.020	**Pa 232** 1.31 d β⁻ 0.3, 1.3... ε γ 969, 894, 150... e⁻ σ 460, σf 1500
Th 228 1.9125 a α 5.423, 5.340... γ 84, (216...), e⁻ O20 σ 120, σf < 0.3	**Th 229** **7.0 μs** **7920 a** IT (0.008) e⁻ α? α 4.845, 4.901 4.815... γ 194, 86 211, 31..., e⁻ σ 62.8, σf 30.8	**Th 230** $7.54 \cdot 10^4$ a α 4.687, 4.621... γ (68, 144...), e⁻ Ne24, sf? σ 23.4, σf < 5E-4	**Th 231** 25.52 h β⁻ 0.3, 0.4... γ 26, 84..., e⁻
Ac 227 21.772 a β⁻ 0.04... γ (38,...), e⁻ α 4.953, 4.941... γ (100, 160...), e⁻ σ 880, σf < 3.5E-4	**Ac 228** 6.15 h β⁻ 1.2, 2.1... γ 911, 969, 338 965...	**Ac 229** 62.7 m β⁻ 1.1 γ 165, 569, 262 146, 135...	**Ac 230** 122 s β⁻ 2.9... γ 455, 508 1244... βsf

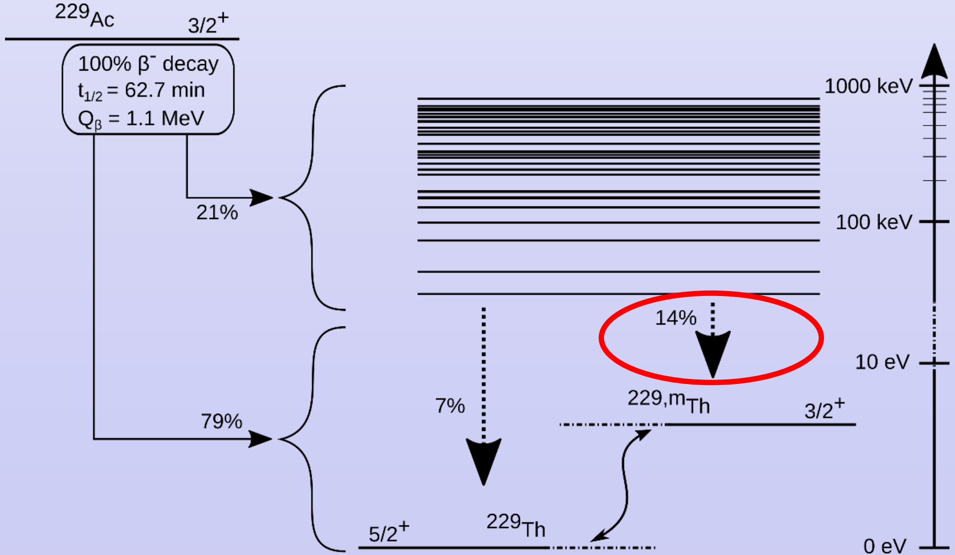
- **Efficient population of ^{229m}Th:**

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

- **^{233}U (α-)decay:**

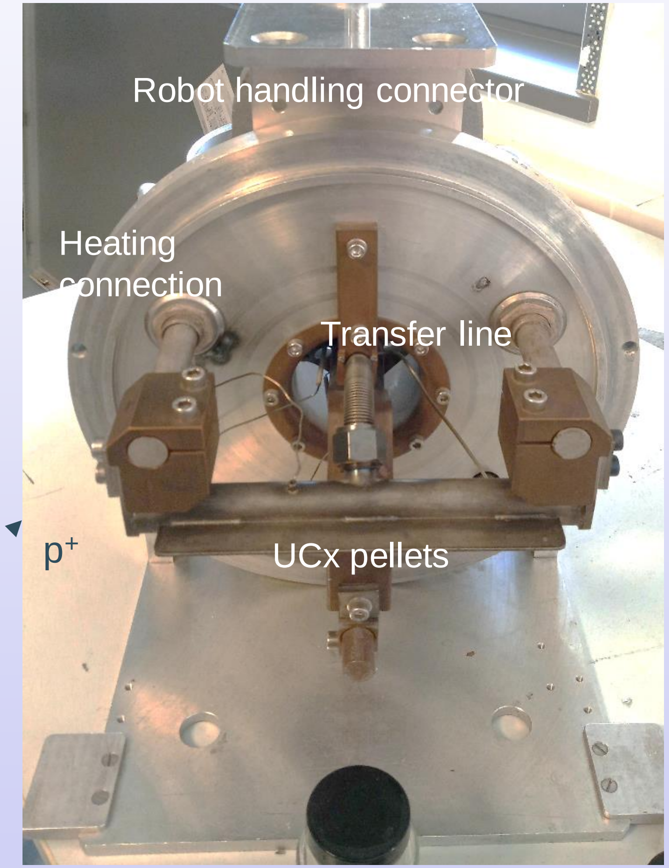


- **^{229}Ac (β-)decay:**



VUV spectroscopy at ISOLDE / CERN

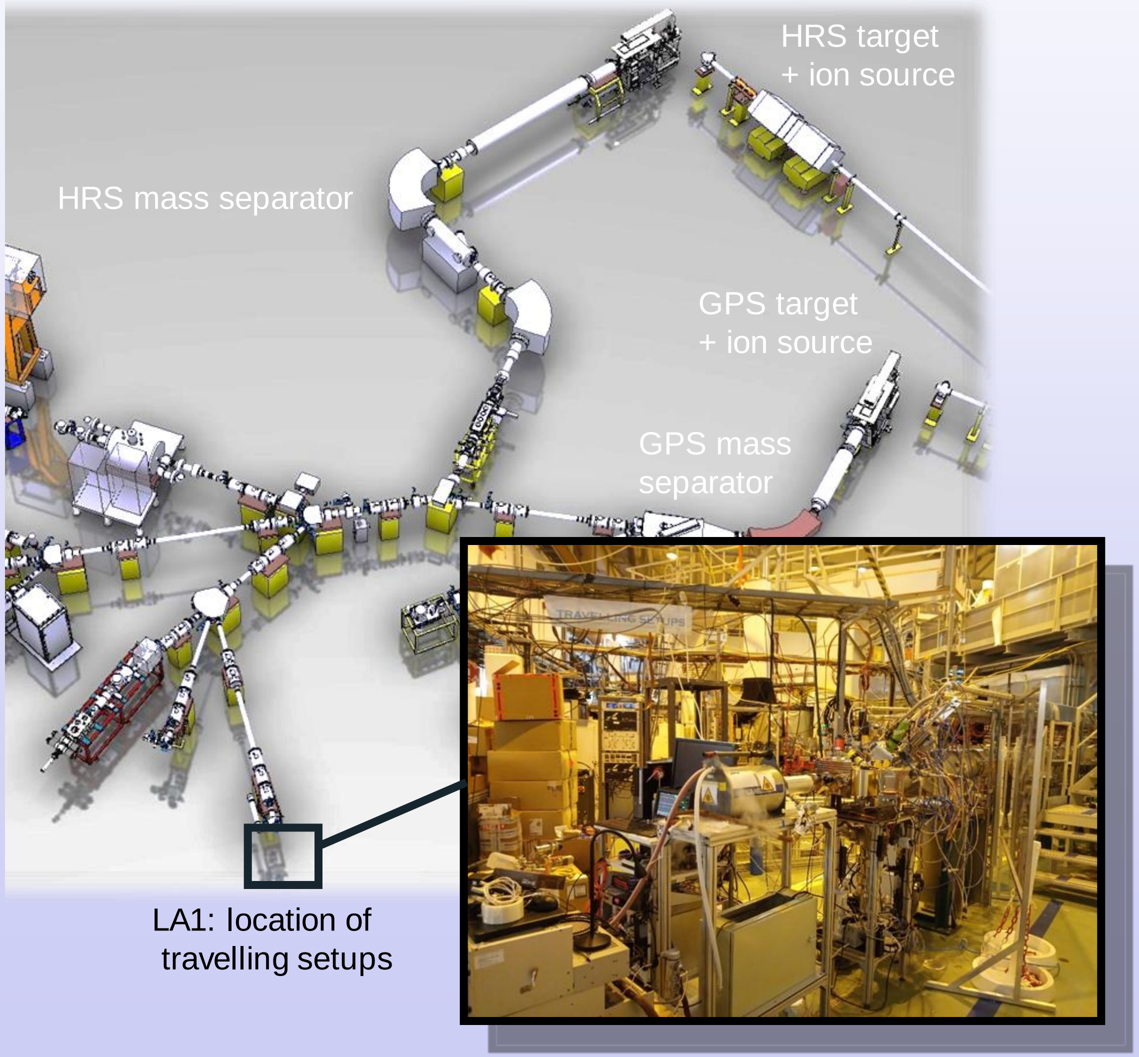
- **Production:** **1.4 GeV protons on UC$_x$**



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

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

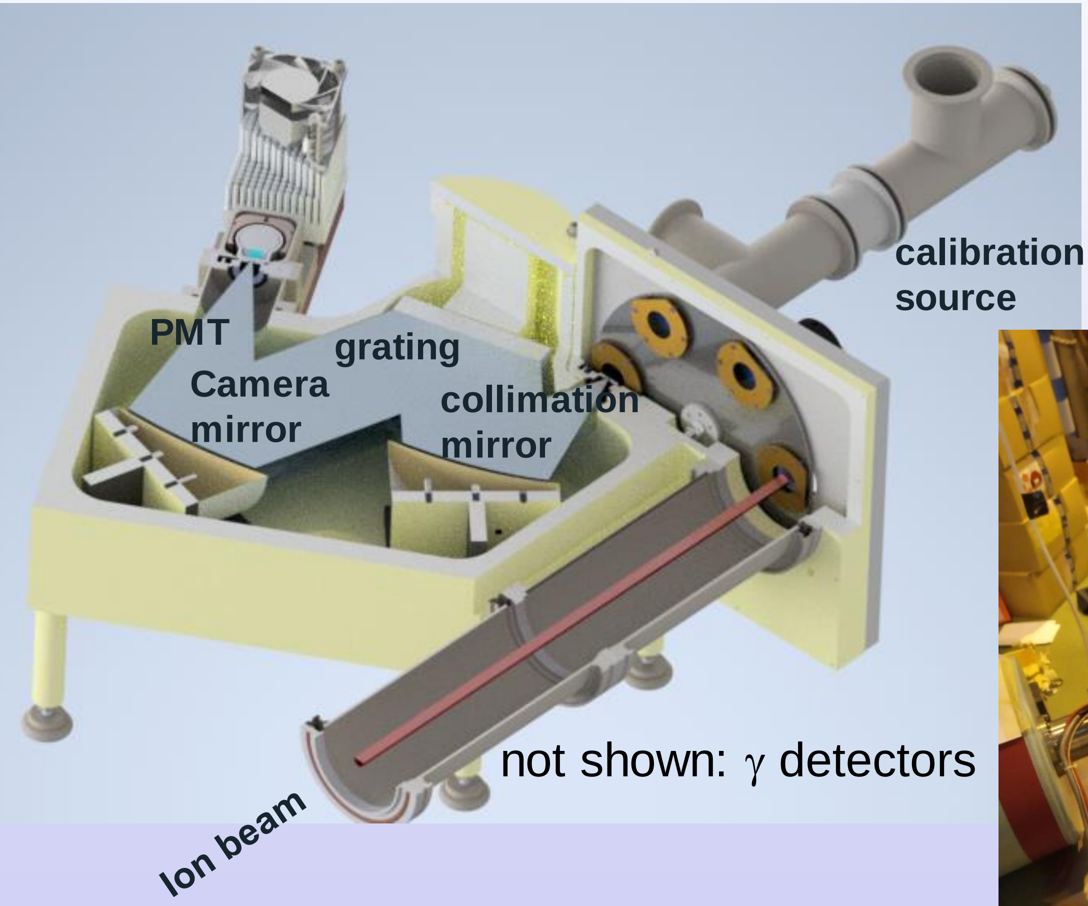
- **Beamline:** **ionization, mass separation, delivery**



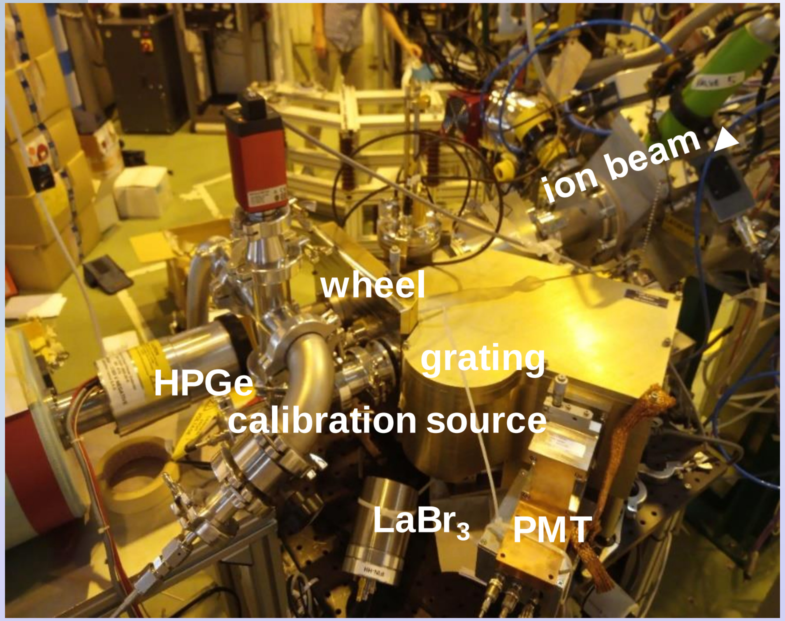
ISOLDE beamline sketch: CERN

VUV spectroscopy at ISOLDE / CERN

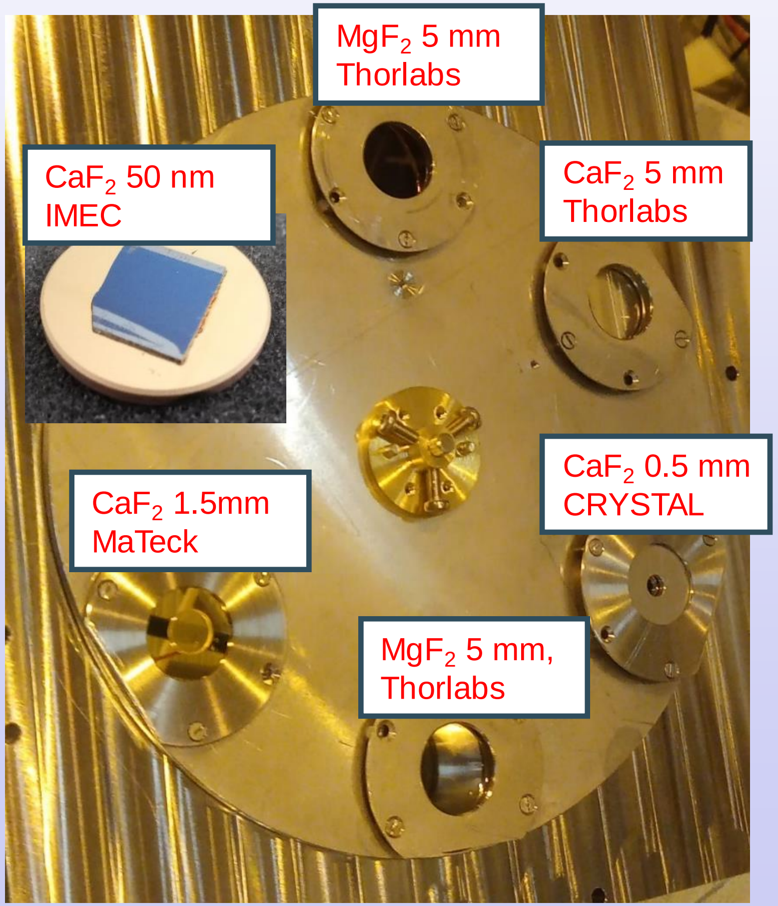
VUV spectrometer:



not shown: γ detectors



various crystals on wheel:



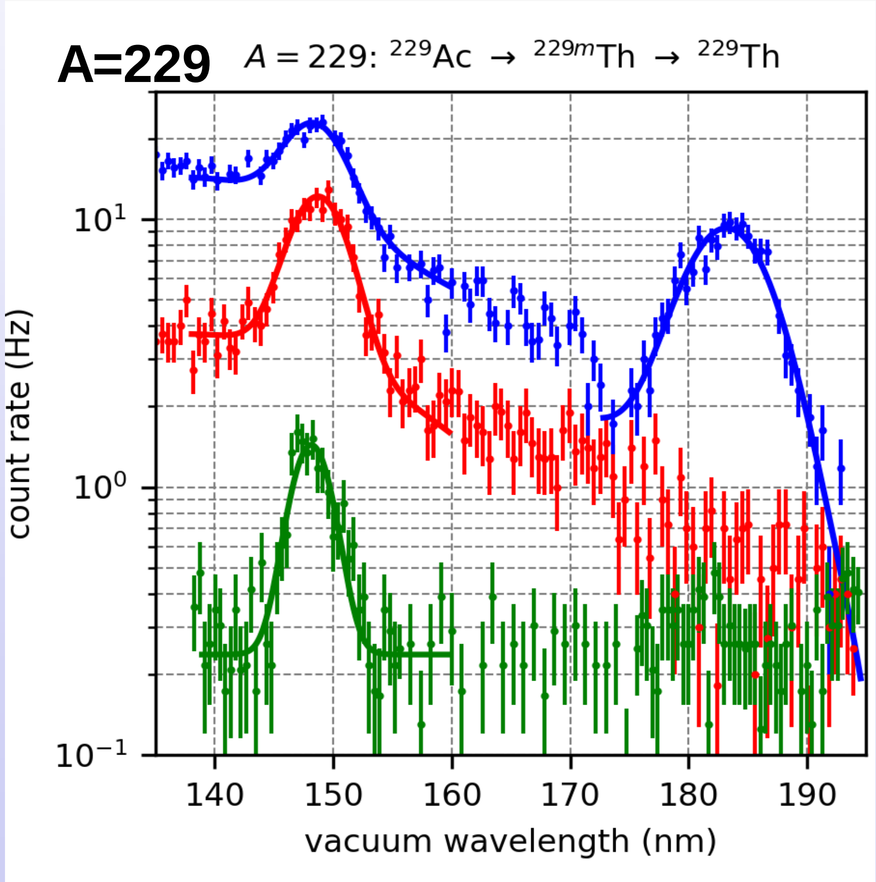
ISOLDE beam: ^{229}Fr, ^{229}Ra

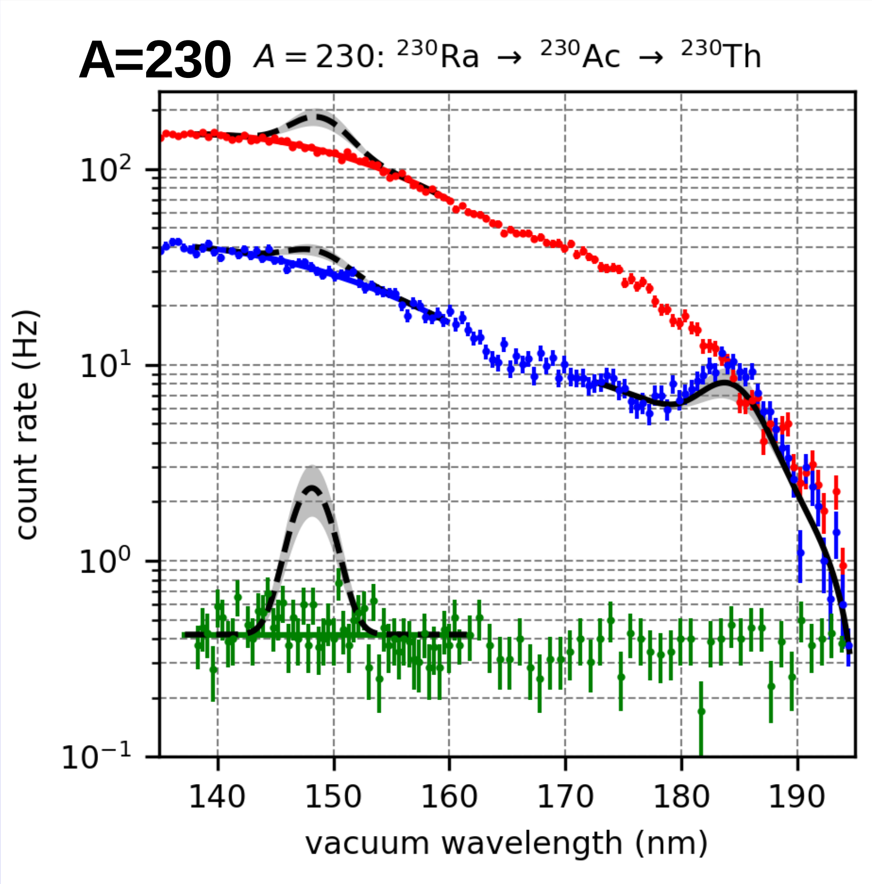
(collaboration led by KU Leuven group (P. Van Duppen et al.))

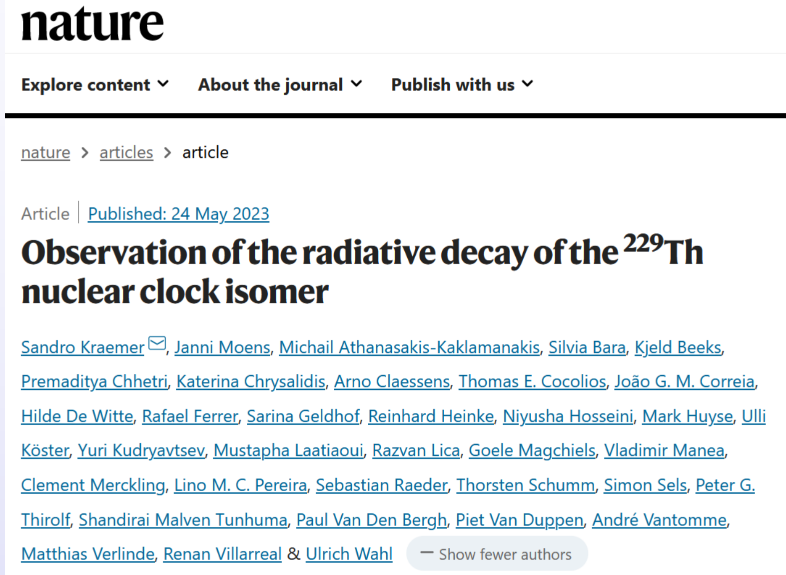
VUV Spectroscopy Results

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

MgF$_2$ (5 mm) CaF$_2$ (5 mm) CaF$_2$ (50 nm)



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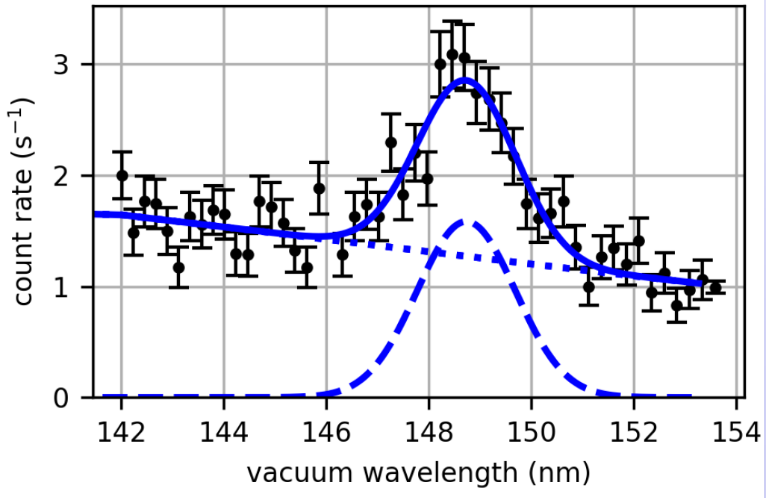
Article | Published: 24 May 2023

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

— Show fewer authors

CaF$_2$ (5 mm)

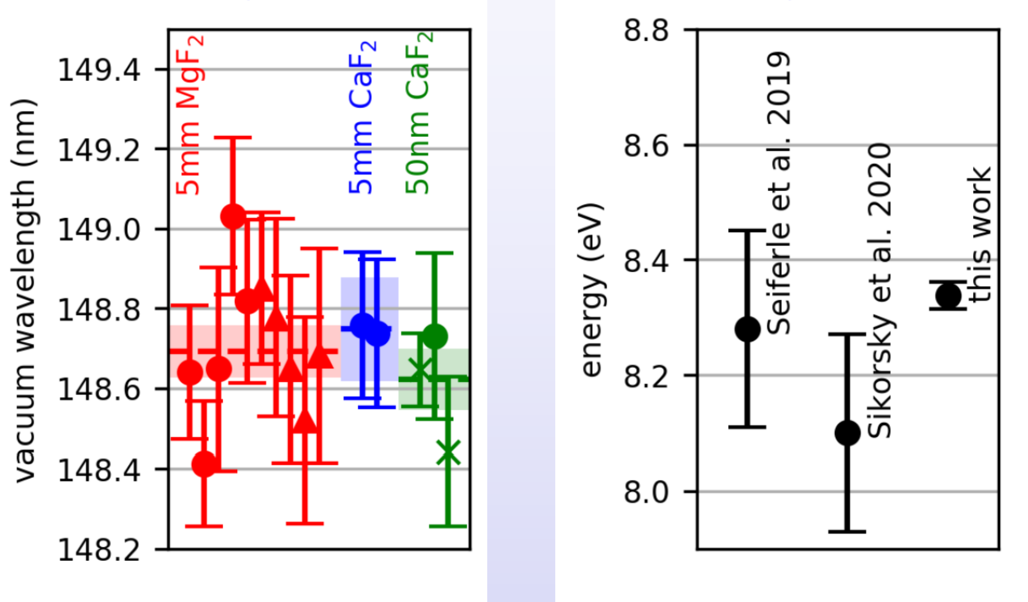


3 mm spectrometer entrance slit

0.5 mm entrance slit

VUV Spectroscopy Results

excitation energy/ emission wavelength:



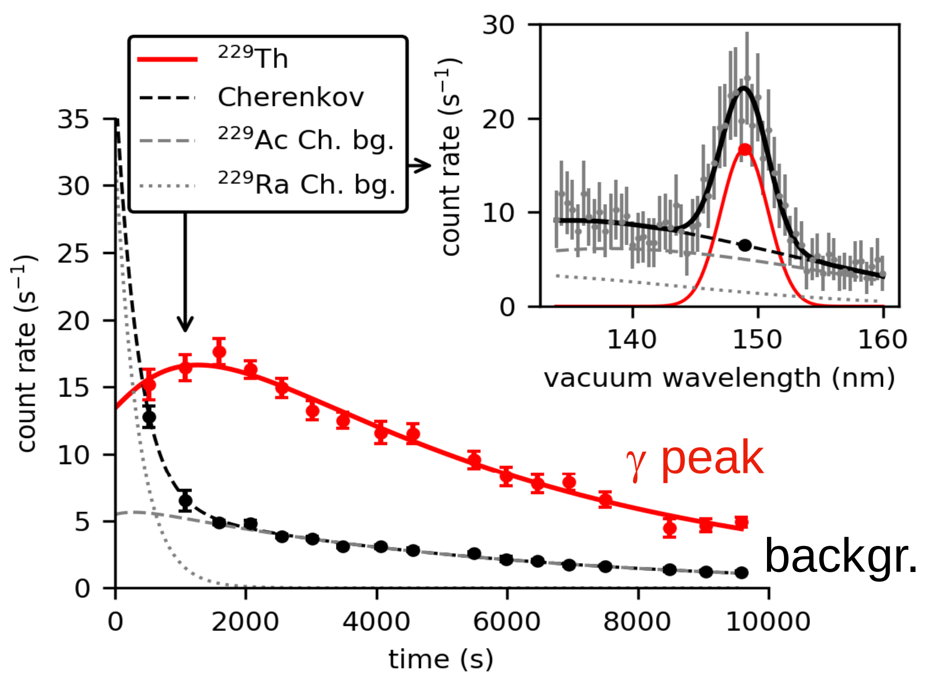
8.338 ± 0.003(stat.) ± 0.023(syst.) eV

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

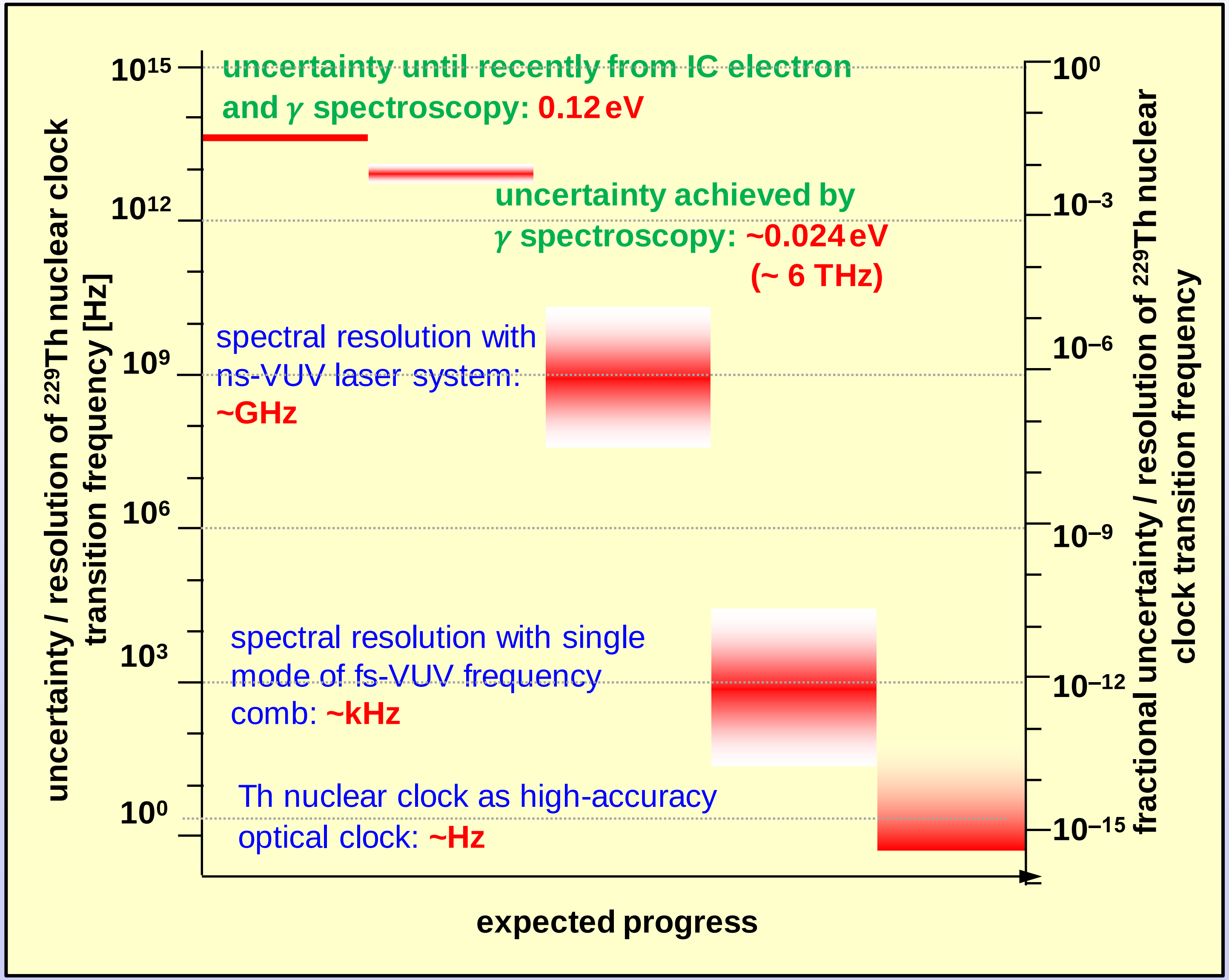
- **still to bridge: 10-12 orders of magnitude:**

heading from nuclear to optical domain:
“ from eV to (k)Hz”
nuclear methods → optical methods

- feasible with existing laser technology
- (4-wave mixing) laser setups:
  PTB (E. Peik et al.), UCLA (E. Hudson et al.)

- (VUV frequency comb) laser:
  JILA/NIST (J. Ye et al.)
  under development at LMU

- ultimate goal: narrow-band cw laser



uncertainty until recently from IC electron and γ spectroscopy: 0.12 eV

uncertainty achieved by γ spectroscopy: ~0.024 eV (~ 6 THz)

spectral resolution with ns-VUV laser system: ~GHz

spectral resolution with single mode of fs-VUV frequency comb: ~kHz

Th nuclear clock as high-accuracy optical clock: ~Hz

First laser excitation of the Thorium Isomer

Hot of the press (accepted, not yet published):

PHYSICAL REVIEW LETTERS

Laser excitation of the Th-229 nucleus

Phys. Rev. Lett.

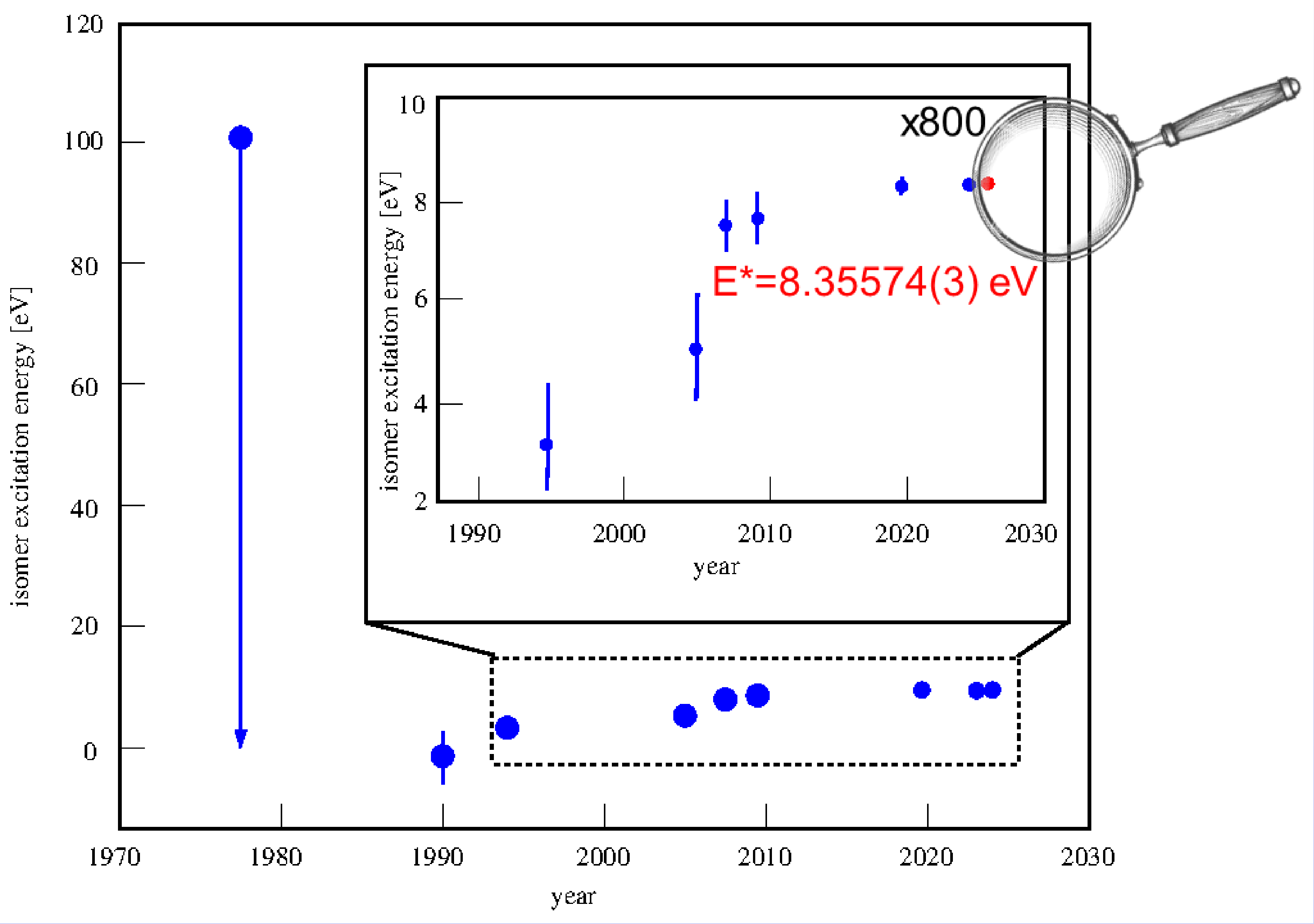
J. Tiedau, M. V. Okhapkin, K. Zhang, J. Thielking, G. Zitzer, E. Peik, F. Schaden, T. Pronebner, I. Morawetz, L. Toscani De Col, F. Schneider, A. Leitner, M. Pressler, G. A. Kazakov, K. Beeks, T. Sikorsky, and T. Schumm

Accepted 14 March 2024

“The nuclear resonance for the Th4+ ions in Th:CaF2 is measured at the wavelength 148.3821(5) nm, frequency 2020.409(7) THz, and the fluorescence lifetime in the crystal is 630(15) s, corresponding to an isomer half-life of 1740(50) s for a nucleus isolated in vacuum.”

Group of Ekkehard Peik at PTB using highly ^{229}Th-doped CaF$_2$ crystals from Thorsten Schumm’s group at TU Wien

Excitation Energy of the Thorium Isomer



E*=8.35574(3) eV

Perspectives for the Nuclear Clock

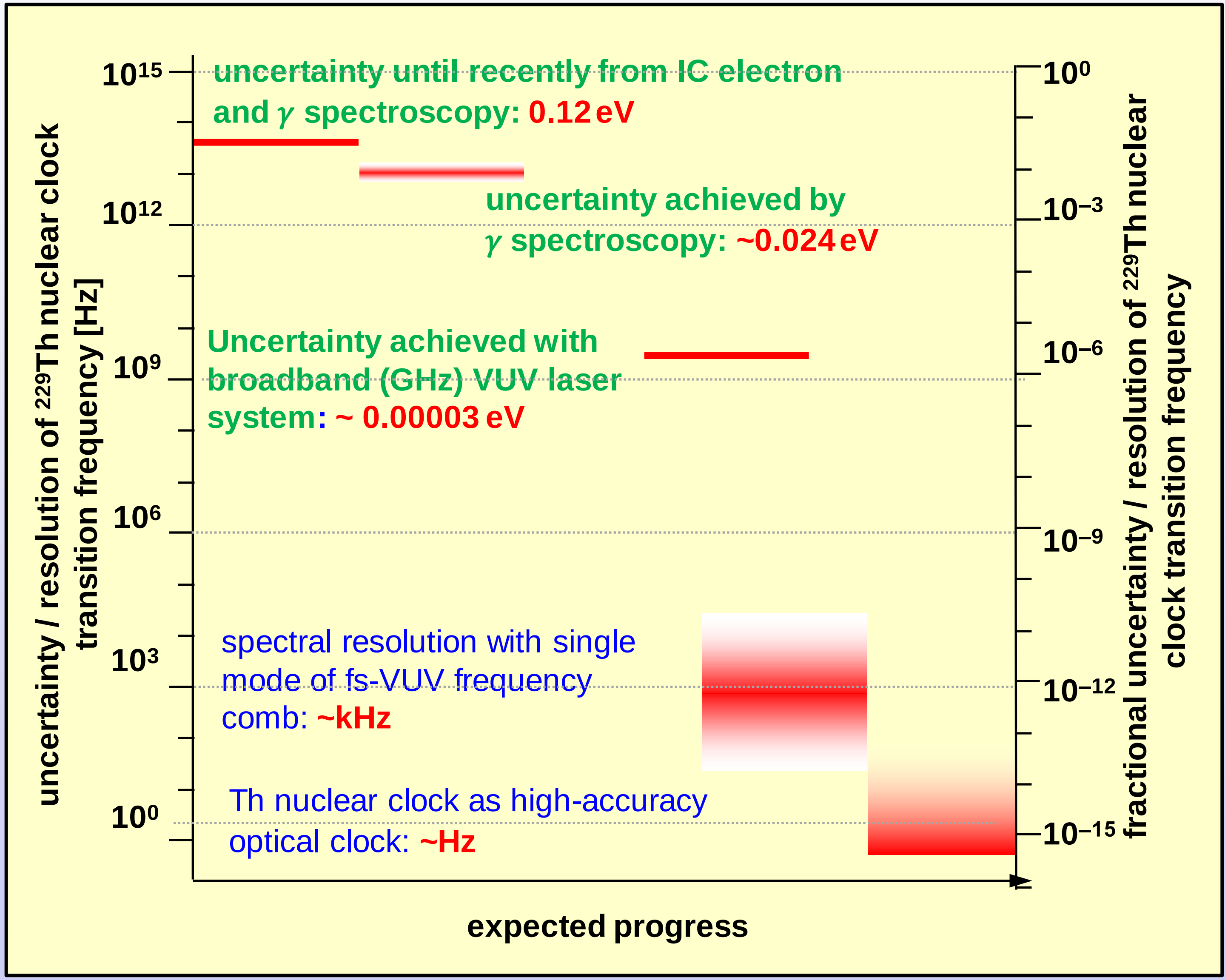
- **still to bridge: 10-12 orders of magnitude:**

heading from nuclear to optical domain:
“ from eV to (k)Hz”
nuclear methods → optical methods

- first optical excitation achieved:
  (4-wave mixing) laser setup at PTB (E. Peik et al.)

- next milestone:
  narrowband resonant excitation via
  VUV frequency comb laser:
    JILA/NIST (J. Ye et al.)
    under development at LMU

- ultimate goal: narrow-band cw laser



uncertainty until recently from IC electron and γ spectroscopy: 0.12 eV

uncertainty achieved by γ spectroscopy: ~0.024 eV

Uncertainty achieved with broadband (GHz) VUV laser system: ~ 0.00003 eV

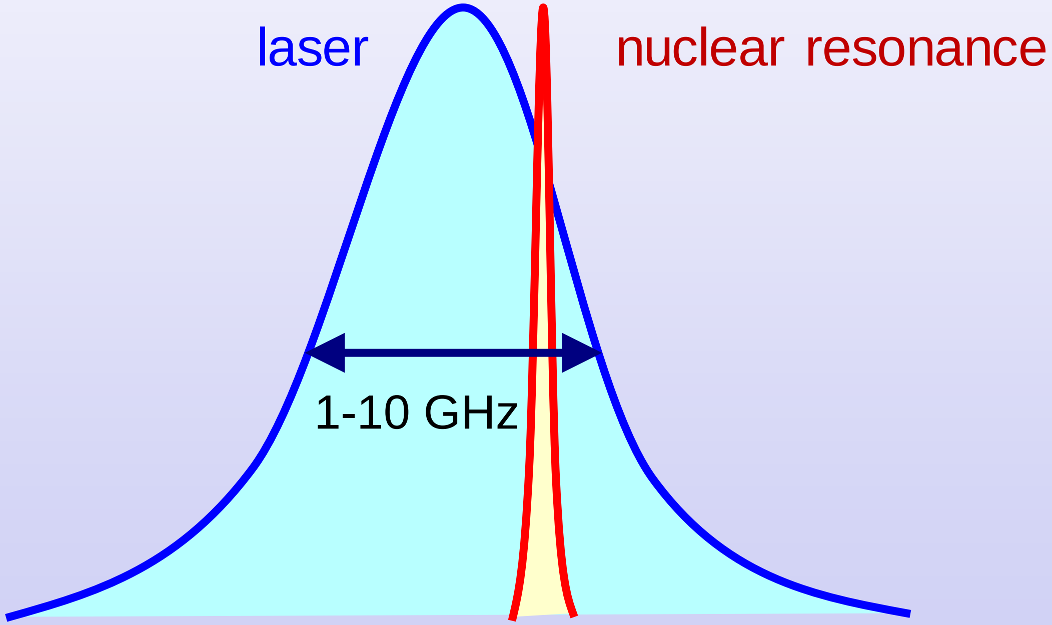
spectral resolution with single mode of fs-VUV frequency comb: ~kHz

Th nuclear clock as high-accuracy optical clock: ~Hz

VUV Lasers for ^{229m}Th Nuclear Excitation

high power, broad bandwidth

- initial search
- shorter scan times
- compromise on frequency resolution

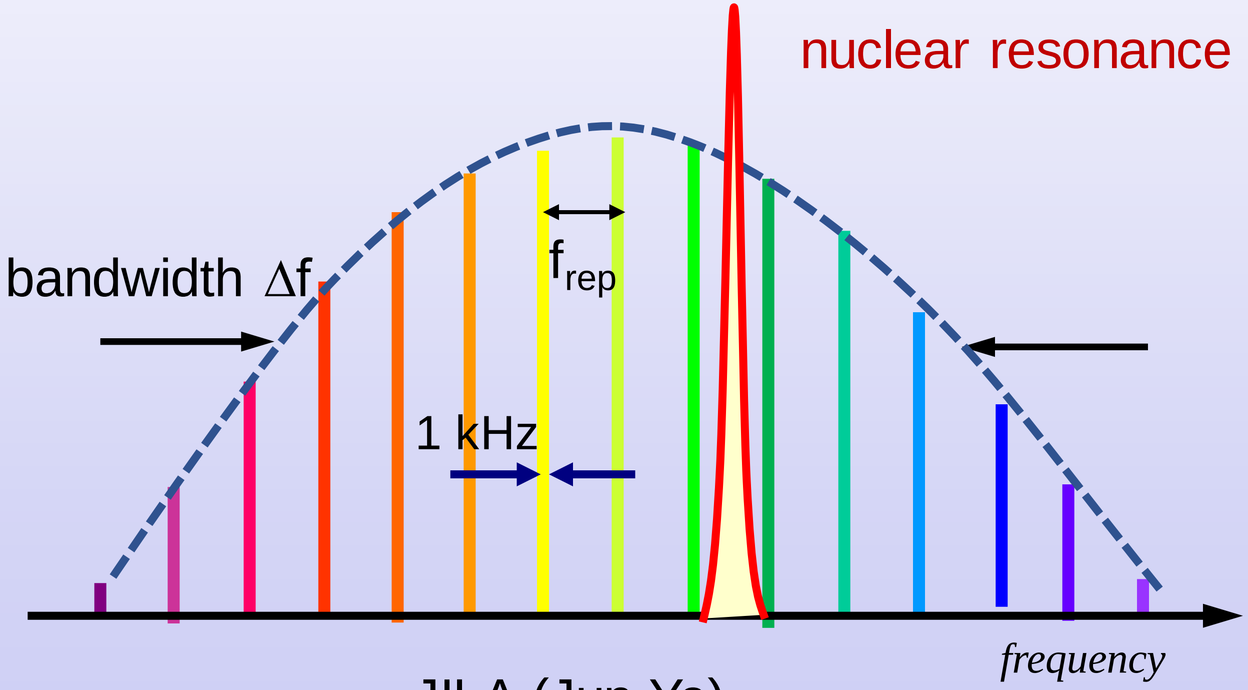


groups:

UCLA (Eric Hudson)
PTB (Ekkehard Peik)

frequency comb

- precision frequency determination
- longer scan times

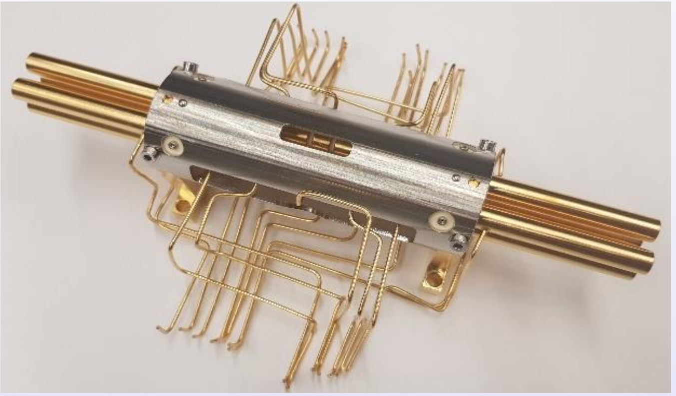


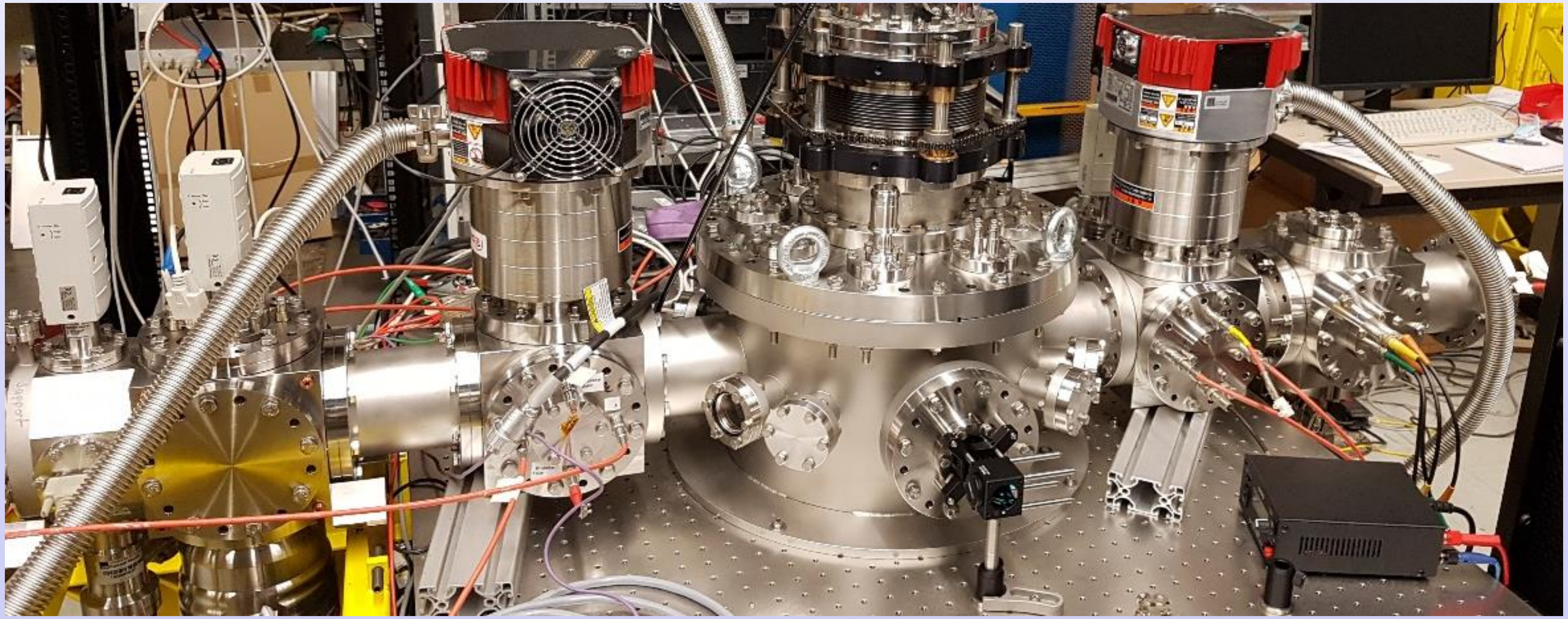
JILA (Jun Ye)
TU Wien (Thorsten Schumm)
LMU (Peter Thirolf)

Ionic Lifetime Measurement

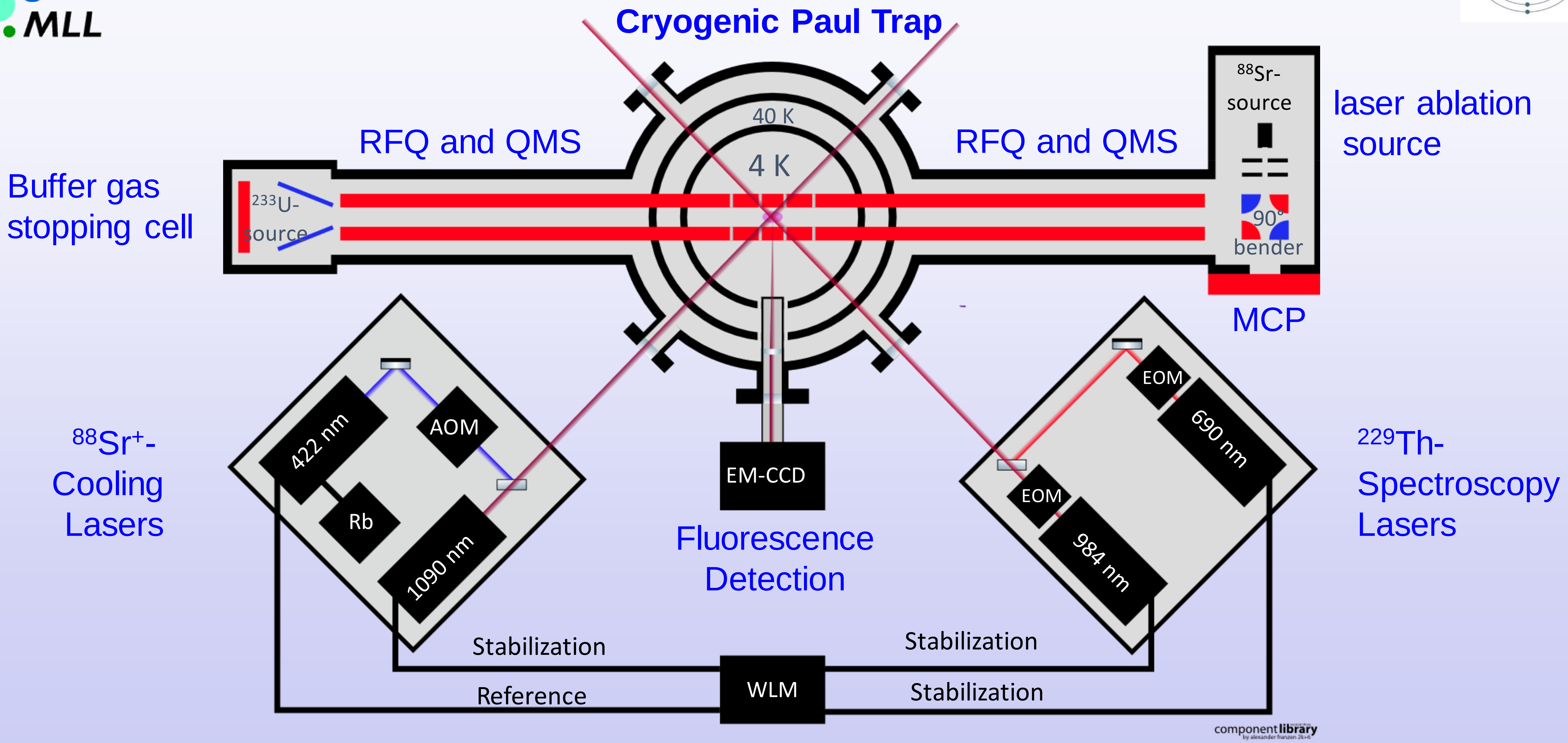
needs longer storage time (= better vacuum)

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



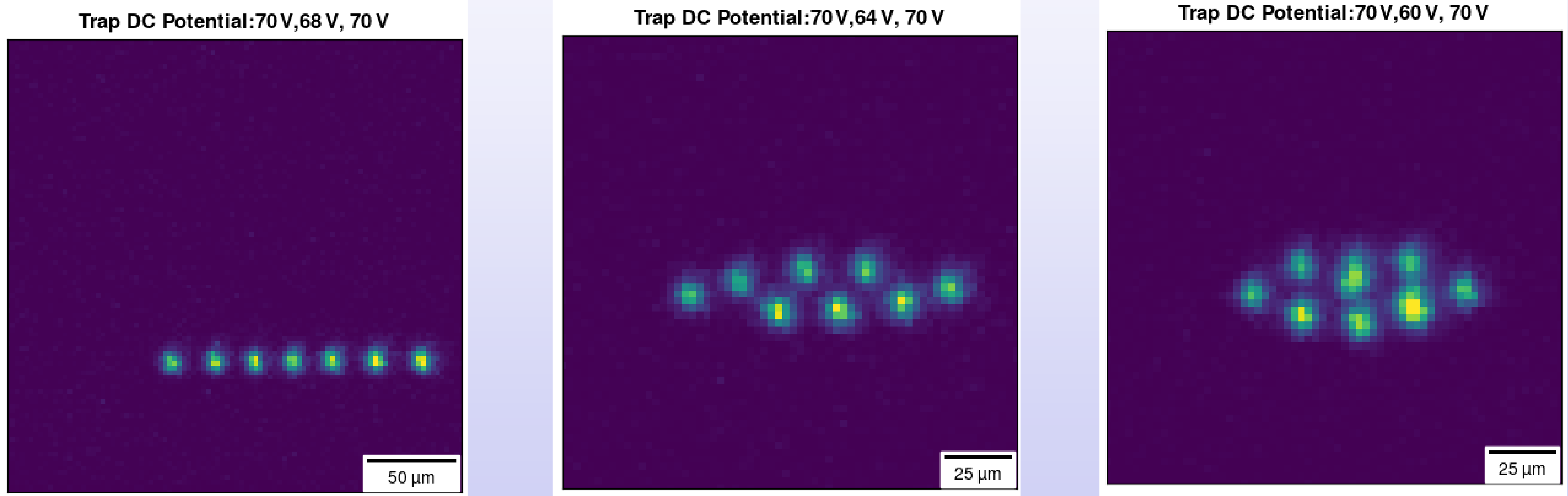


Cryo Trap & Laser Setup



Cryo Trap & Laser Setup

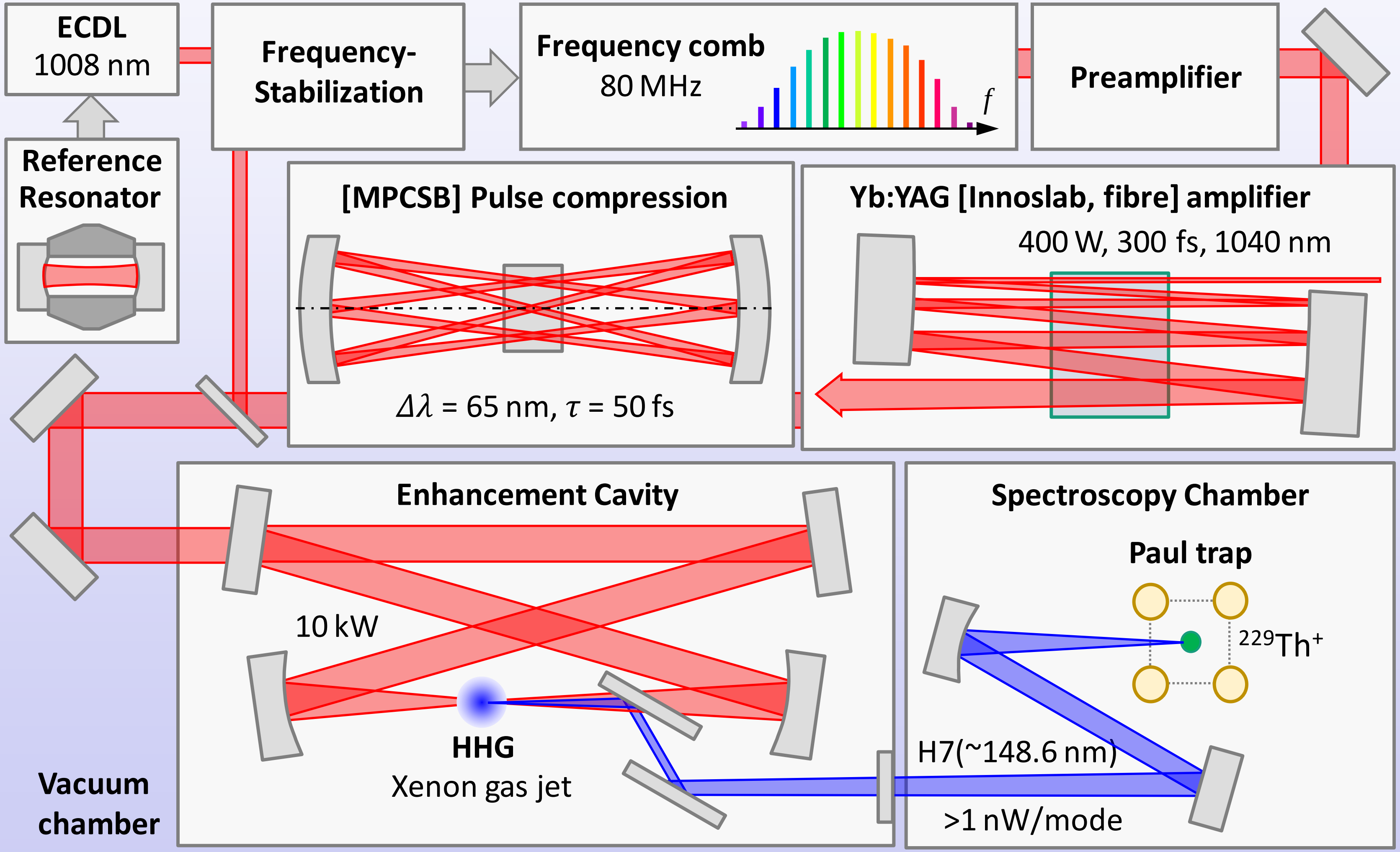
- laser cooling of $^{88}Sr^{+}$: Coulomb crystals



- commissioning of ^{229}Th injection, cooling, detection: work in progress

VUV laser source for ^{229m}Thorium

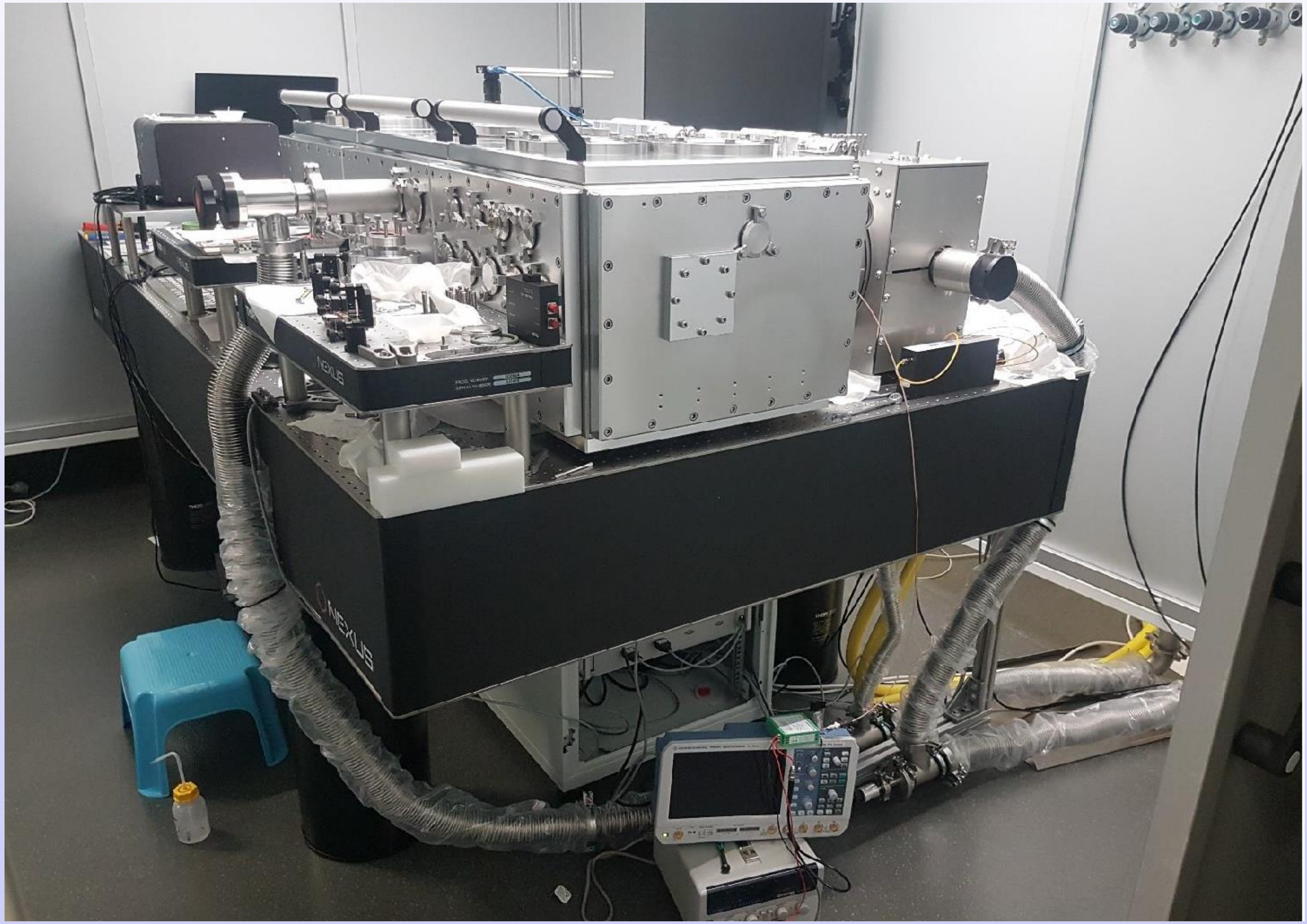
- **VUV frequency comb: use 7th harmonic of amplified IR frequency comb:**



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

VUV laser source for ^{229m}Thorium

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



laser under development at Fraunhofer ILT (Aachen) together with LMU.

operational: end of 2024

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

ERC Synergy Project: “ThoriumNuclearClock”

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

Scientific advances in many fields:

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

thorium nuclear clock

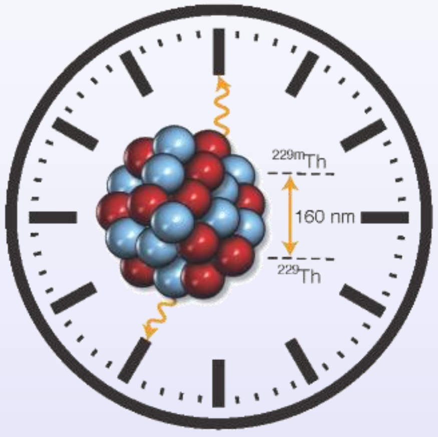
Projects goals:

- Perform the first laser excitation of the ^{229m}Th nuclear transition
- Precisely determine the ^{229}Th 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 Th^{3+} ions
- Demonstrate completely new solid-state clock scheme
- Test fundamental concepts of physics
- Search for dark matter

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
- identification/excitation of nuclear resonance with (broadband) laser:
  E*= 8.35574(3) eV, λ = 148.3821(5) nm, τ (in CaF_2): 630(15) s → $t_{1/2}$ (vacuum) = 1740(50) 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 narrowband laser precision:
  → VUV frequency comb under development
- determine sensitivity enhancement for $\dot{\alpha}$

Ambitious goals lie ahead:

- 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,**
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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 !