Physikalisch-Technische Bundesanstalt
Braunschweig and Berlin
National Metrology Institute

Towards laser excitation of the low-energy nuclear transition in ^{229}Th

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European Research Council
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TC FS

9th Symposium on Frequency Standards and Metrology,
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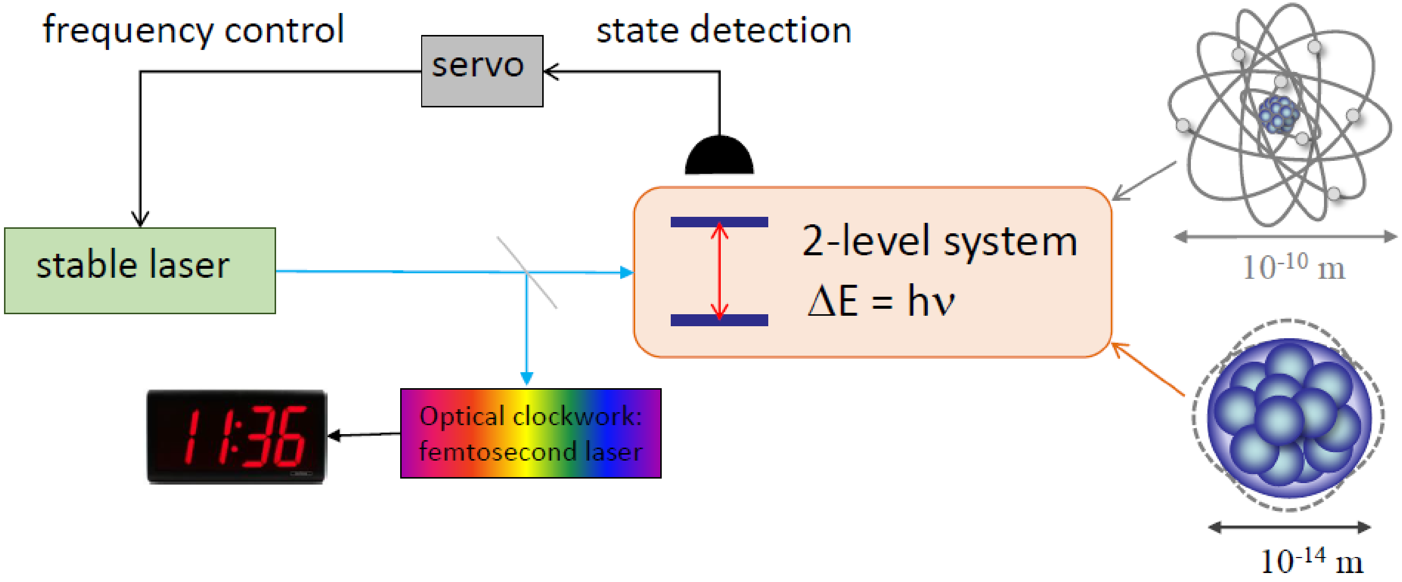
From the atomic to the nuclear clock



frequency control

servo

state detection

stable laser

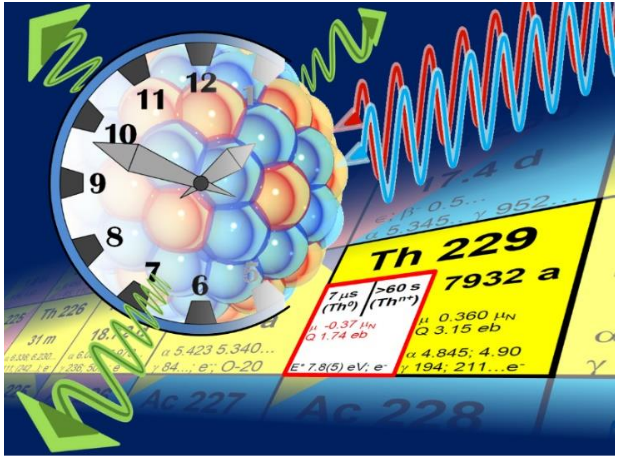
2-level system

$\Delta E = h\nu$

Optical clockwork: femtosecond laser

Nuclear Clock:

Oscillator that is frequency-stabilized
to a nuclear (γ-ray) transition



Motivation:

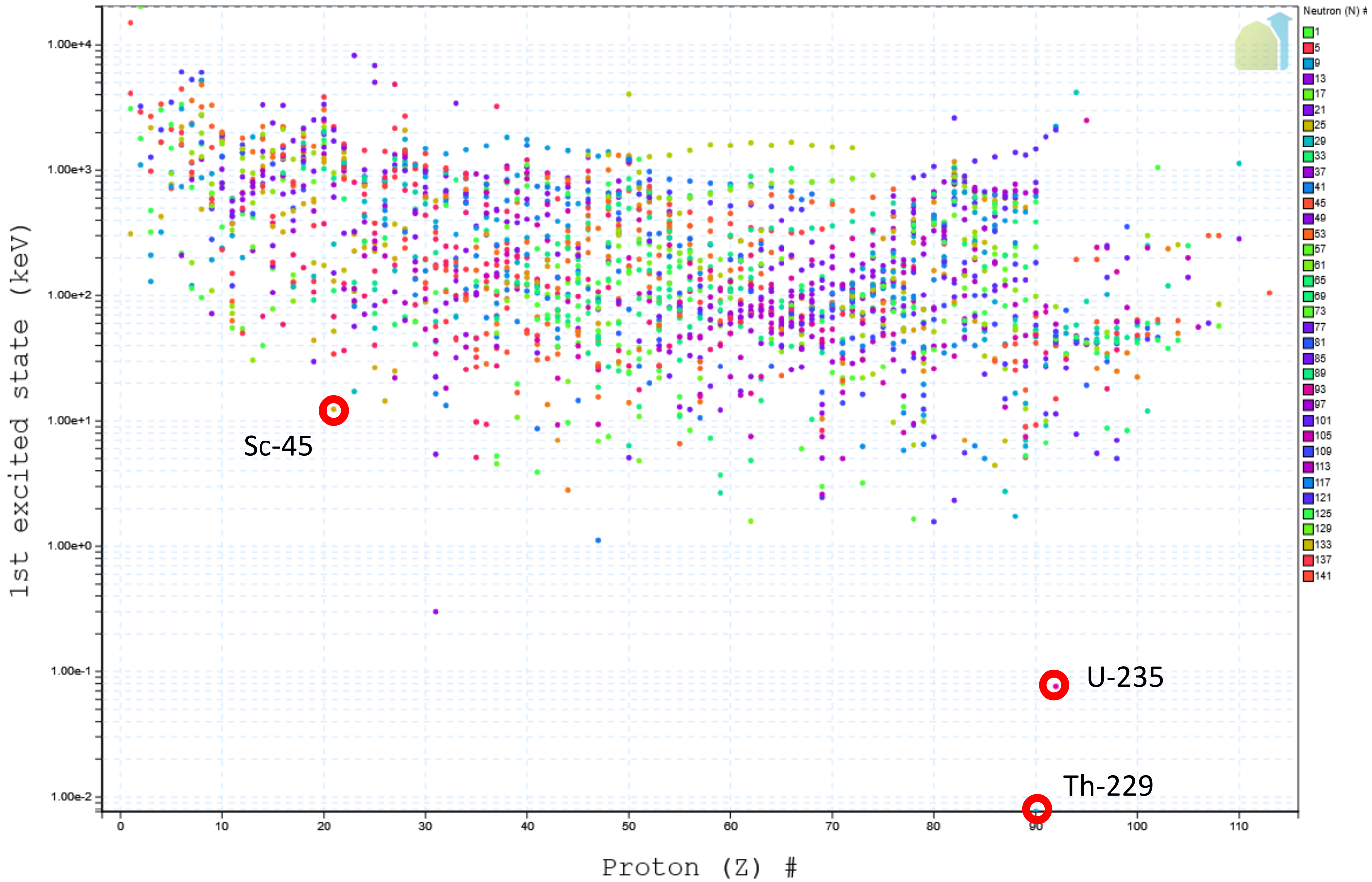
Higher precision: In many of the advanced optical clocks (trapped ion and optical lattice) field-induced shifts make a dominant contribution to the uncertainty budget. These can be reduced in a nuclear clock.

Higher stability: In a Mößbauer solid state nuclear clock, many absorbers may be interrogated ($>10^{10}$ instead of $\approx 10^{0}$ (ion) or $\approx 10^{4}$ (lattice)).

Higher frequency: → higher stability. EUV or even X-ray transitions
may be used when suitable radiation sources become available.

High sensitivity in fundamental tests (strong and electromagnetic interactions)

Energy of the 1st excited nuclear state



Sc-45:
12389.59 eV, M2

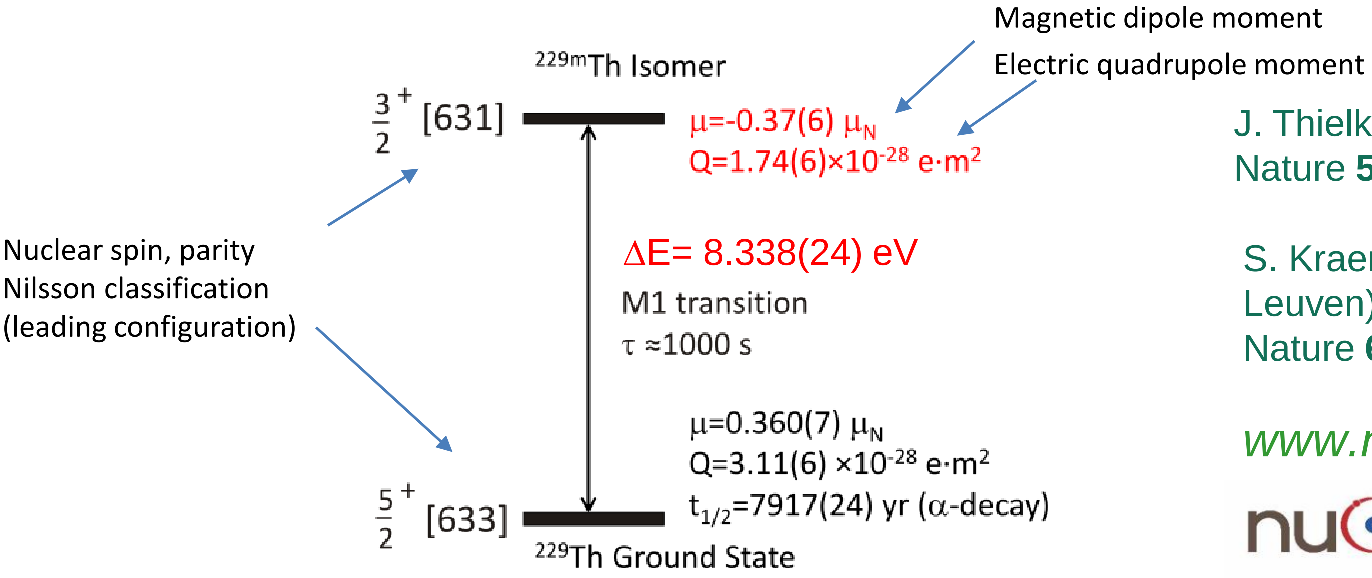
U-235:
76.737 eV, E3

Th-229:
8.338 eV, M1

https://www.nndc.bnl.gov/nudat3/

Low-energy transition in Th-229 as a reference for a nuclear clock

accessible for laser excitation at ≈ 150 nm



Magnetic dipole moment

Electric quadrupole moment

^{229m}Th Isomer

$\frac{3}{2}^{+}$ [631]

μ=-0.37(6) μ_N

Q=1.74(6)×10^{-28} e·m^2

Nuclear spin, parity
Nilsson classification
(leading configuration)

ΔE= 8.338(24) eV

M1 transition

τ ≈1000 s

μ=0.360(7) μ_N

Q=3.11(6) ×10^{-28} e·m^2

$t_{1/2}$=7917(24) yr (α-decay)

$\frac{5}{2}^{+}$ [633]

^{229}Th Ground State

J. Thielking et al. (PTB, LMU)
Nature **556**, 321 (2018)

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

www.nuclock.eu

nuclock

Advantage of the nuclear over the atomic clock: (nearly) free choice of a suitable electronic state for the interrogation of the nuclear resonance.

E. Peik, Chr. Tamm, Europhys. Lett. **61**, 181 (2003)

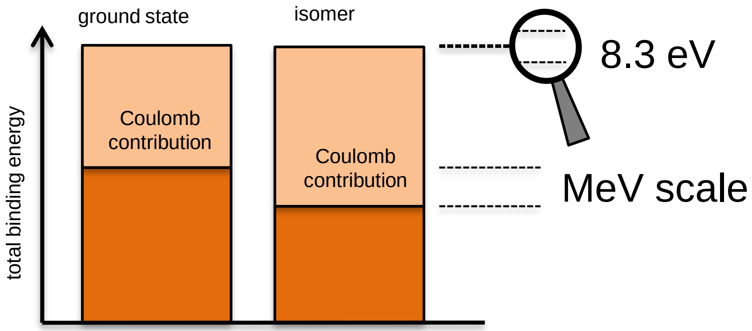
K. Beeks et al., Nat. Rev. Phys. **3**, 238 (2021)

High sensitivity of a Th-229 nuclear clock for violations of the equivalence principle

- Transition frequency is sensitive to the strong interaction (in addition to electromagnetism)

- Coulomb- and strong- contributions (MeV scale) cancel in the transition energy
  Enhanced sensitivity to variations of fundamental constants:
  V. Flambaum, Phys. Rev. Lett. 97, 092502 (2006)

- Bound system of massive particles (n, p) at high energies
  Enhanced effect of LLI violation:
  V. Flambaum, Phys. Rev. Lett. 117, 072501 (2016)



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

News from other labs

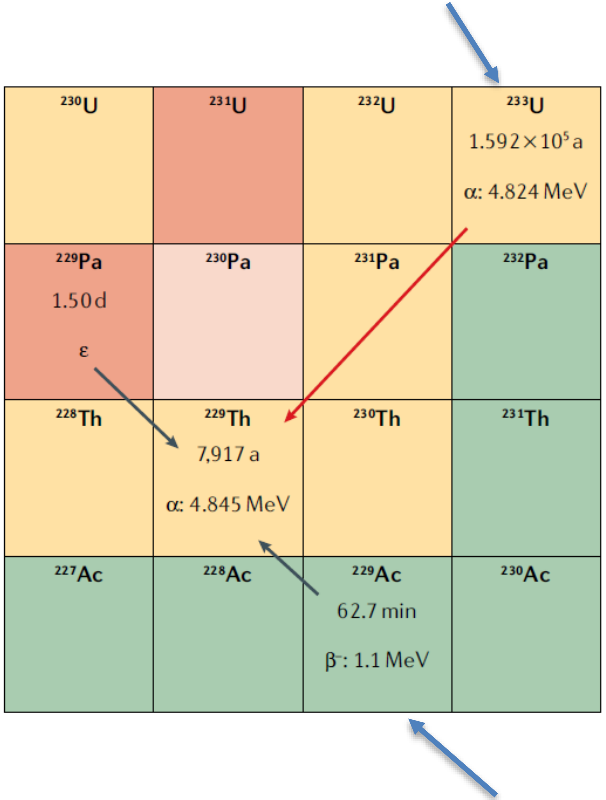
Production of ^{229m}Th through β-decay of ^{229}Ac

- First optical observation of the isomer decay
- First optical wavelength measurement
- First observation of Th-229 nuclear photon emission from solids (MgF_2, CaF_2)

Online experiment at CERN ISOLDE
Lead: KU Leuven, Piet van Duppen
S. Kraemer et al., arXiv:2209.1027
Nature **617**, 706 (2023)

Wavelength 148.71(42) nm
excitation energy 8.338(24) eV
The half-life of ^{229m}Th embedded in MgF_2 is determined to be 670(102) s.

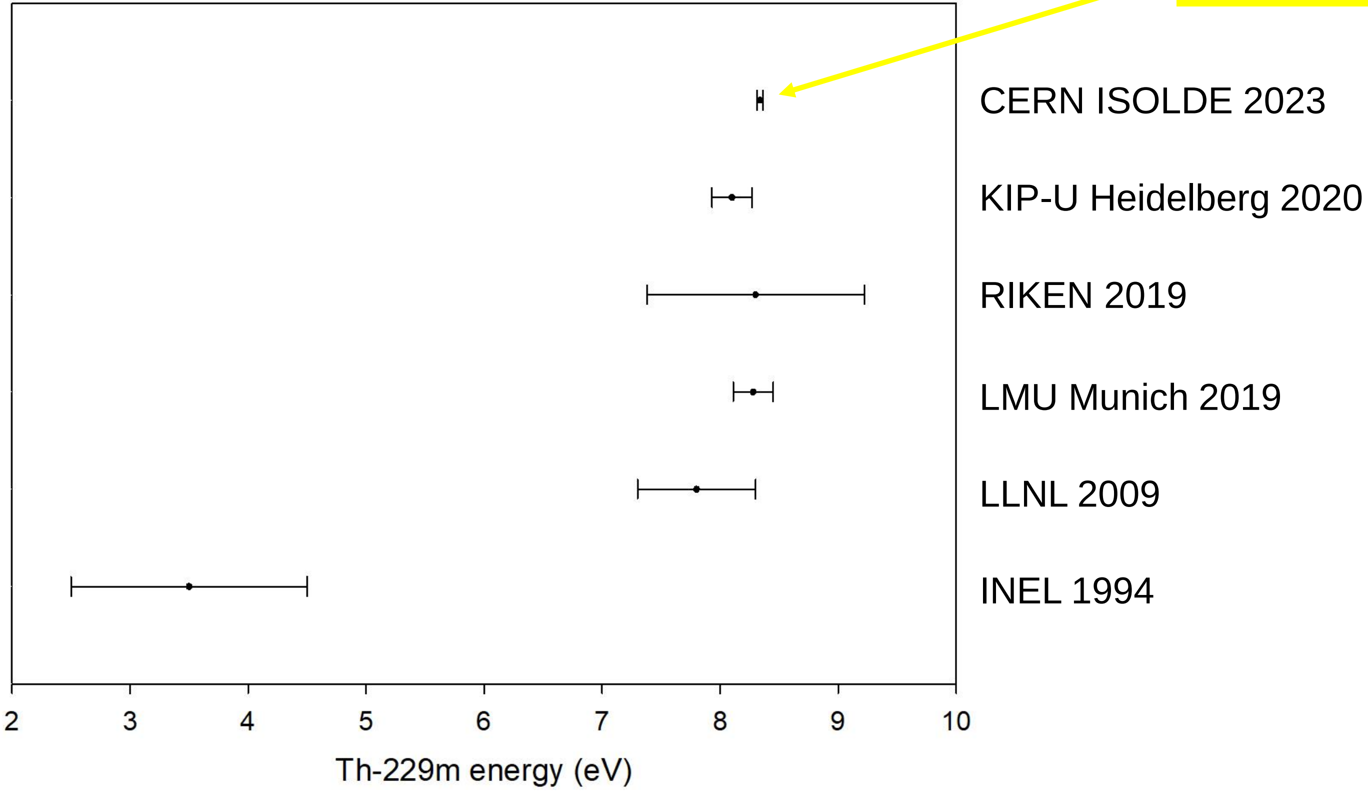
At LMU, PTB, LANL et al.



^{230}U	^{231}U	^{232}U	^{233}U 1.592×10^5 a α: 4.824 MeV
^{229}Pa 1.50 d ε	^{230}Pa	^{231}Pa	^{232}Pa
^{228}Th	^{229}Th 7,917 a α: 4.845 MeV	^{230}Th	^{231}Th
^{227}Ac	^{228}Ac	^{229}Ac 62.7 min β^-: 1.1 MeV	^{230}Ac

At CERN

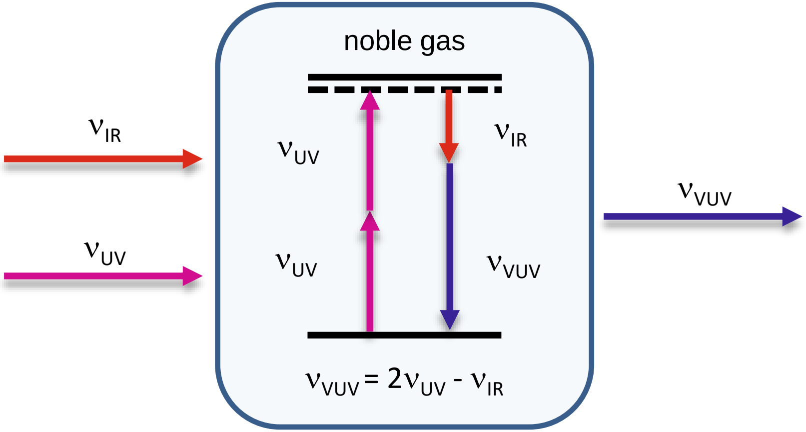
Measurements of the Th-229 isomer energy



≈11 THz ±1σ range

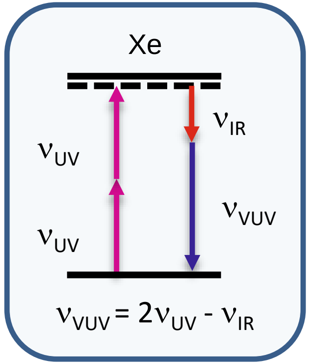
8-eV VUV generation: Four wave (difference) mixing principle

- Near-resonantly driving of 2-photon transition in noble gas.
- Supplying one photon with ν_{IR} yields fourth photon with $\nu_{VUV} = 2\nu_{UV} – \nu_{IR}$.
- Two-photon transition in Xe at 2 x 250 nm is suitable for VUV tunability from 167 nm to 148 nm, i.e. 7.42 eV to 8.38 eV.

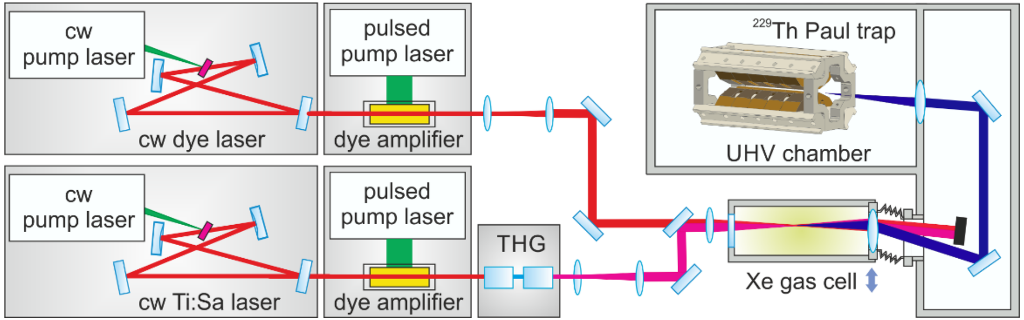


8-eV VUV generation: Four wave (difference) mixing principle

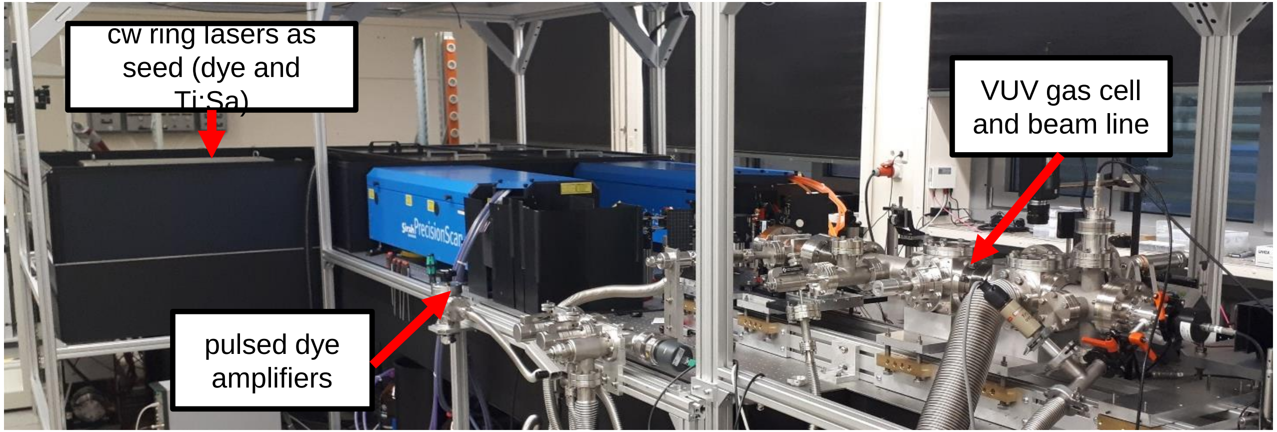
- Tuning over the range 7.9 eV to 8.3 eV requires laser beams at 250 nm and 610-759 nm.
- Third order process needs high intensity to achieve suitable efficiency.
- Pulsed lasers (~10 ns, 30 Hz repetition rate) best compromise between VUV pulse energy (>10^{13} photons/pulse) and Fourier transform limited bandwidth (<1 GHz).
- Our setup:
  - Two cw ring lasers as seed: 750 nm Ti:Sa laser, 610-759 nm tunable dye laser.
  - Pulsed dye amplifiers (~60 mJ/pulse, 30 Hz repetition rate).
  - Third harmonic generation to achieve 250 nm for two-photon transition.



$$\nu_{VUV} = 2\nu_{UV} - \nu_{IR}$$

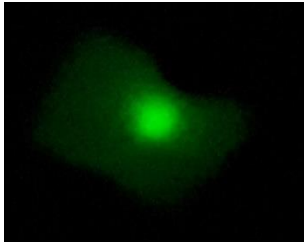


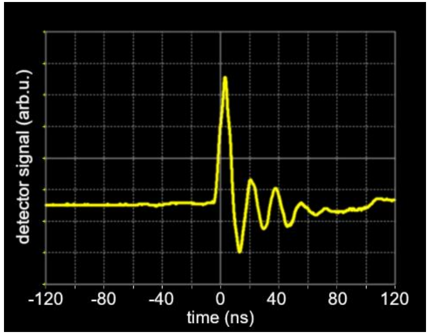
FWM laser system at PTB


cw ring lasers as seed (dye and Ti:Sa)

VUV gas cell and beam line

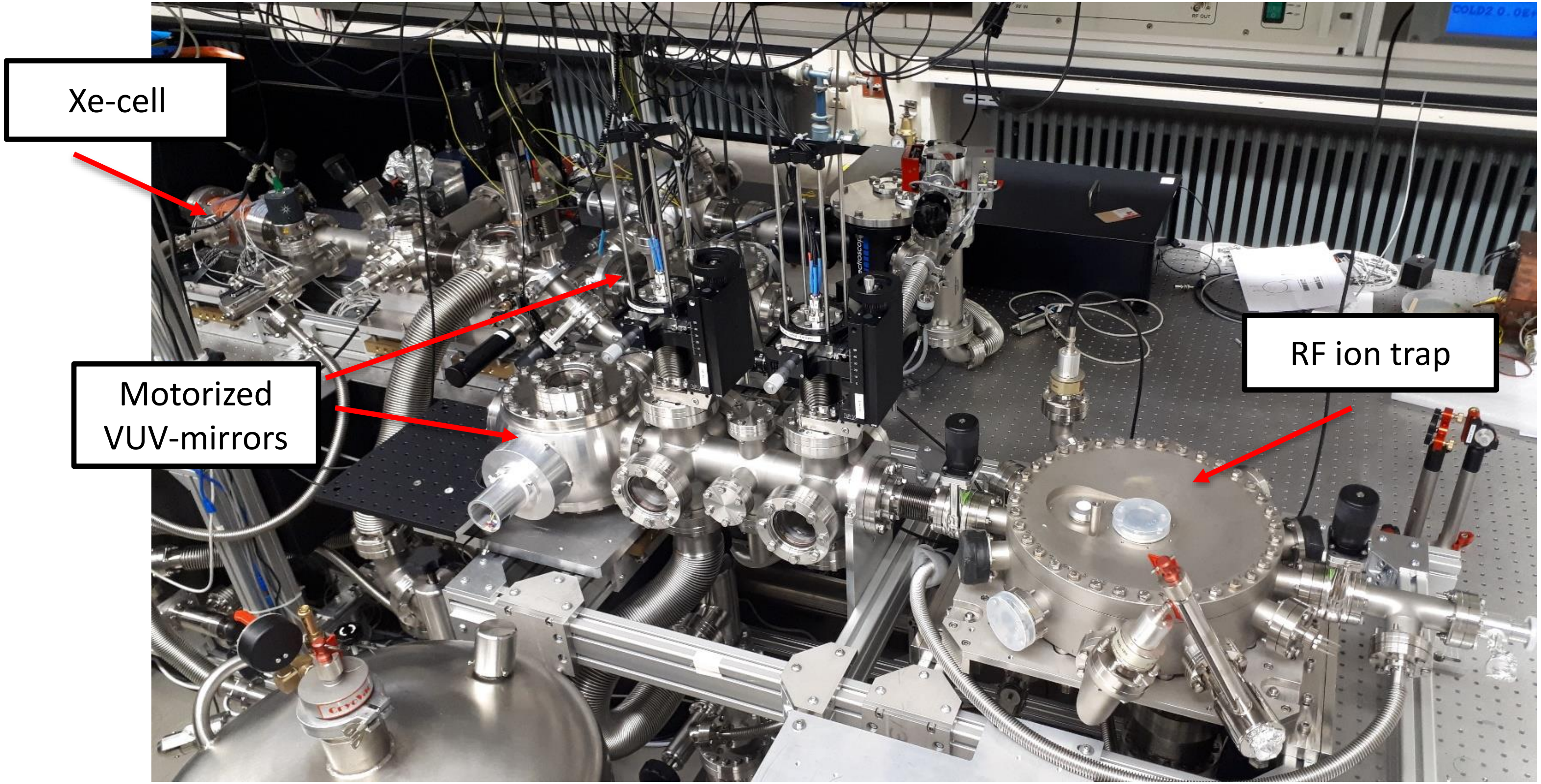
pulsed dye amplifiers


VUV beam visualization on Ce:YAG phosphor



- Relative power measurements with Cu-based photo-electron detector.
- Absolute measurements with pyro-electric power meter show $E_{pulse} > 5$ µJ.

VUV Beam Line

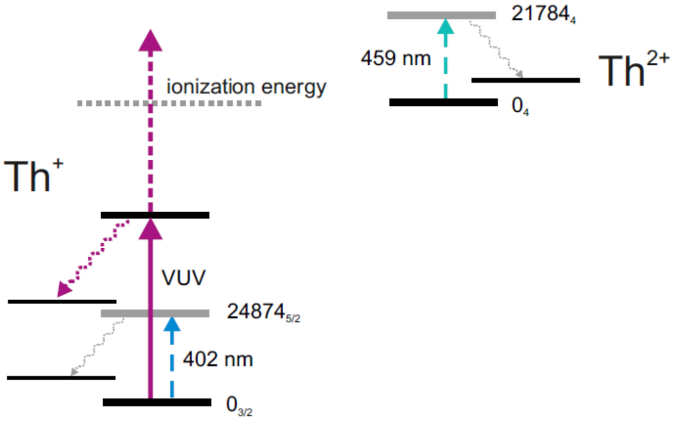


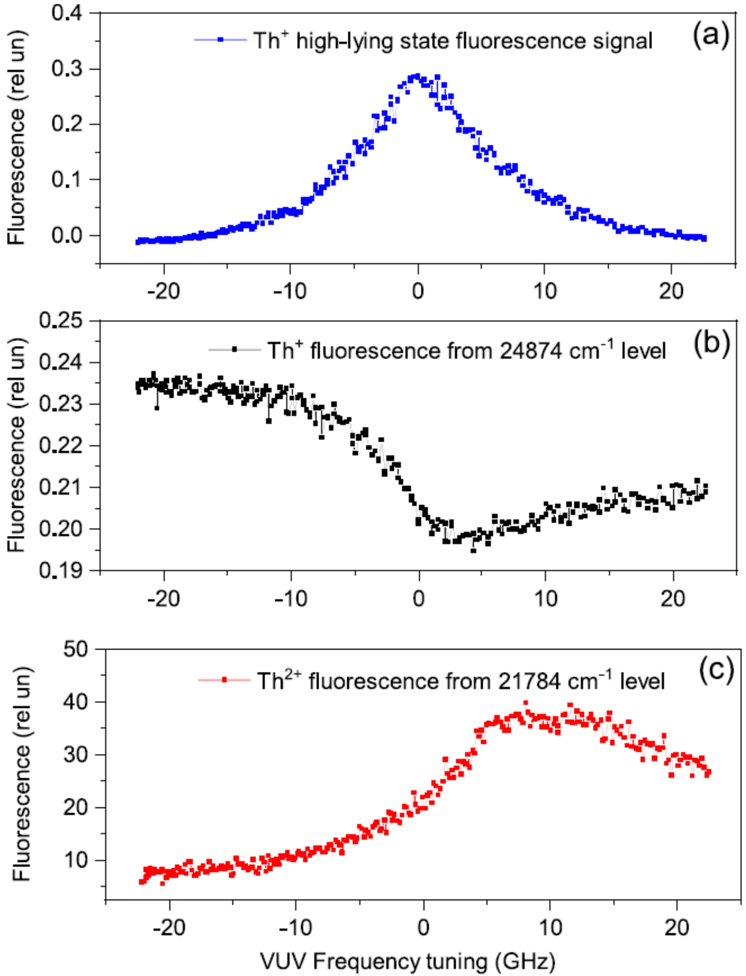
Xe-cell

Motorized VUV-mirrors

RF ion trap

First VUV laser excitation of trapped Th$^+$ ions (electronic resonance lines)





Detection:
in Th$^+$: provides laser spectral profile
in Th^{2+} : sensitive to the integrated excitation rate

J. Thielking, K. Zhang, J. Tiedau, J. Zander, G. Zitzer, M. V. Okhapkin, E. Peik, New J. Phys. **25**, 083026 (2023)

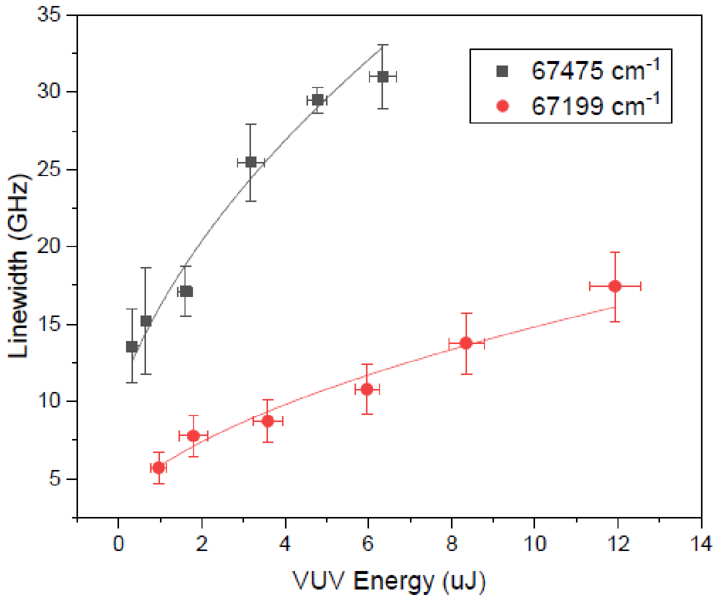


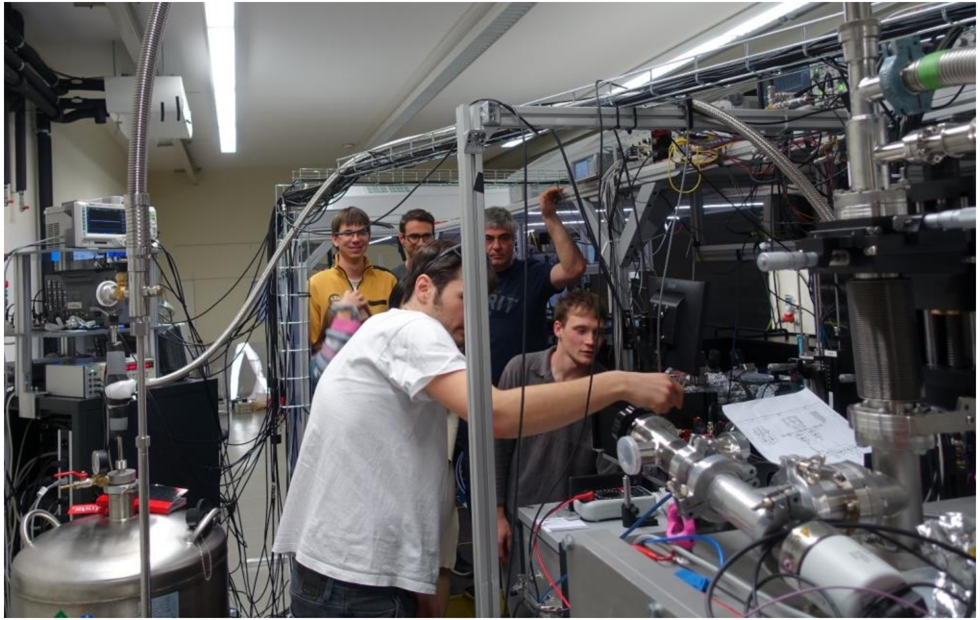
Figure 7. Width of the resonances vs. VUV pulse energy for two transitions. A contribution from Doppler broadening of 2.7 GHz is subtracted from the observed resonance widths.

Linewidth of the VUV source: ≤6 GHz

(phase noise from the amplifiers gets upconverted in THG and FWM processes)

J. Thielking, K. Zhang, J. Tiedau, J. Zander, G. Zitzer, M. V. Okhapkin, E. Peik, New J. Phys. **25**, 083026 (2023)

In cooperation with TU Vienna, group of Thorsten Schumm:
Study of ^{229}Th-doped calcium fluoride crystals



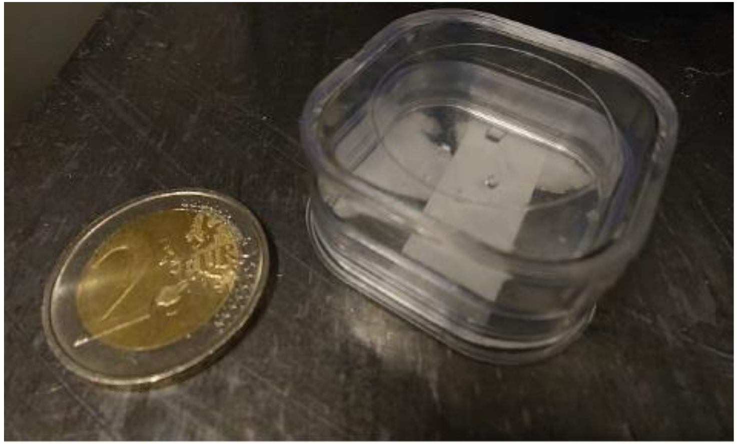
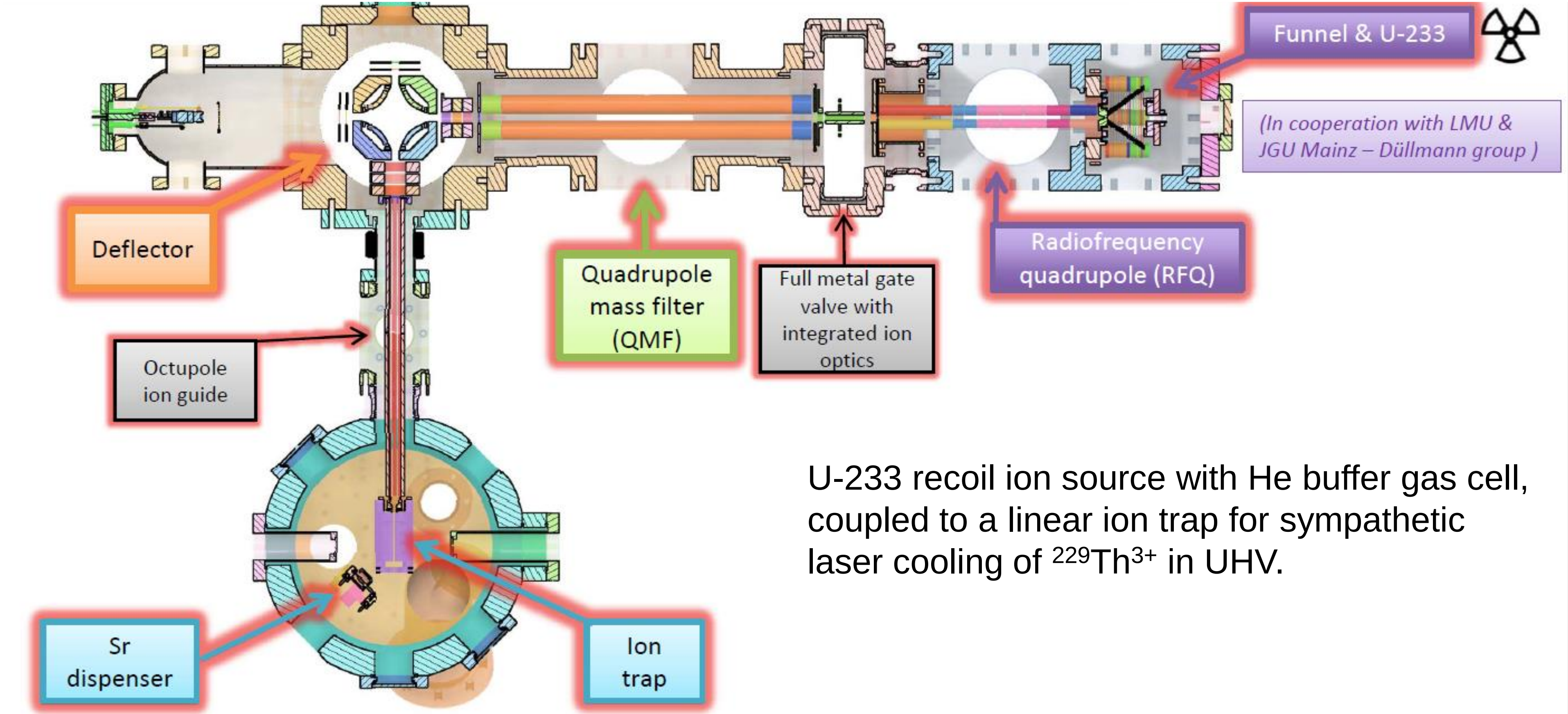


FIGURE 3.18: Four 1 mm^3 and one 4 mm^3 ^{229}Th:CaF$_2$ crystals cut and polished using above techniques.

Kjeld Beeks, PhD thesis TU Wien, 2022

- Radiation damage to crystals (loss of F)
- VUV-absorbing „ices“ at cryogenic temperature

Trapping and cooling of $^{229}\mathrm{Th}^{3+}$

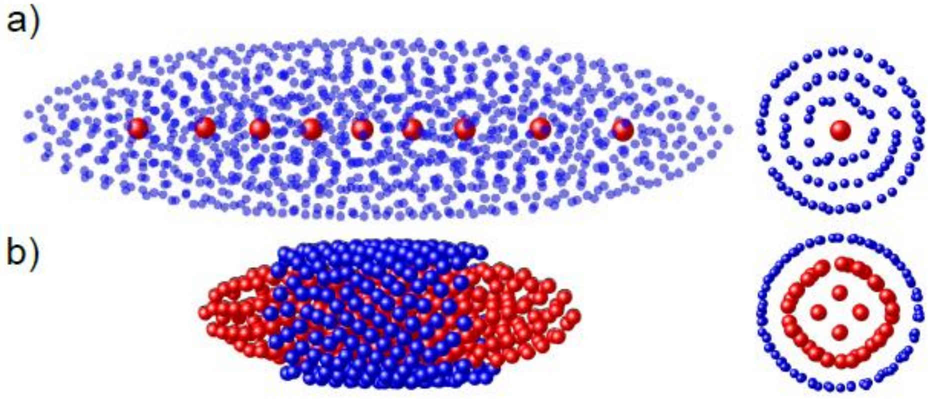


U-233 recoil ion source with He buffer gas cell, coupled to a linear ion trap for sympathetic laser cooling of $^{229}\mathrm{Th}^{3+}$ in UHV.

Sympathetic laser cooling of $^{229}\text{Th}^{3+}$ with $^{88}\text{Sr}^{+}$

Both species have similar q/m and form closely coupled two-species Coulomb crystals in a linear RF ion trap.

Simulations



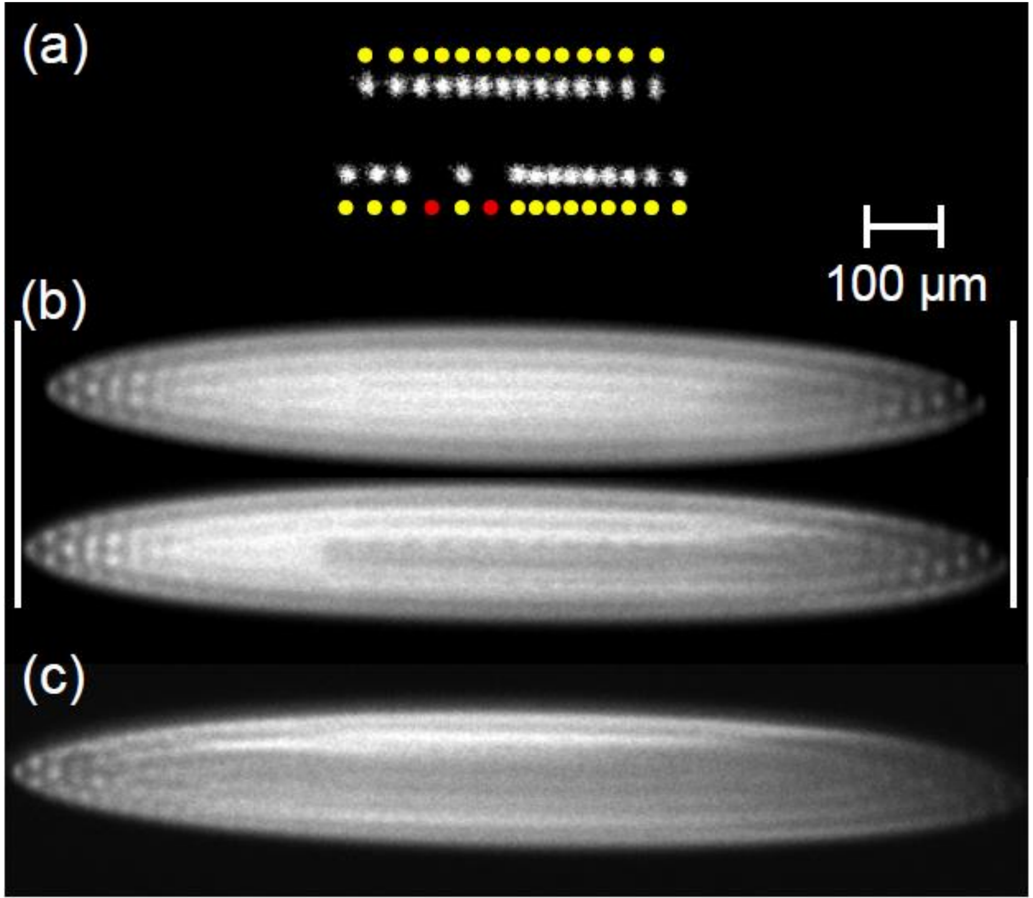
1000 Sr^{+} (blue) and 9 Th^{3+} (red)

500 Sr^{+} (blue) and 500 Th^{3+} (red)

First laser cooling of $^{229}\text{Th}^{3+}$: Georgia Tech
C. J. Campbell, A. G. Radnaev, and A. Kuzmich, Phys. Rev. Lett. 106, 223001 (2011).

Sympathetic laser cooling of $^{229}Th^{3+}$ with $^{88}Sr^{+}$

Experiment: fluorescence image from Sr^{+}, Th^{3+} appear dark



Sr^{+} only

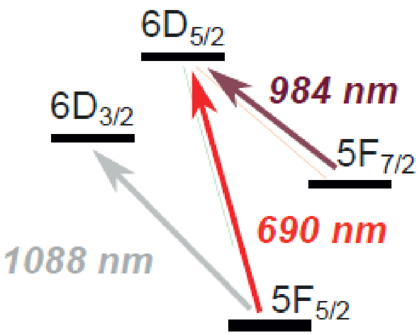
with 2 Th^{3+}

Sr^{+} only

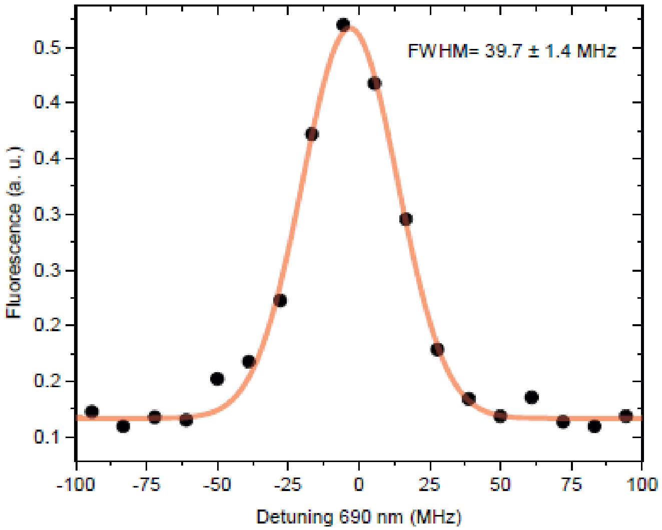
with ≈12 Th^{3+}

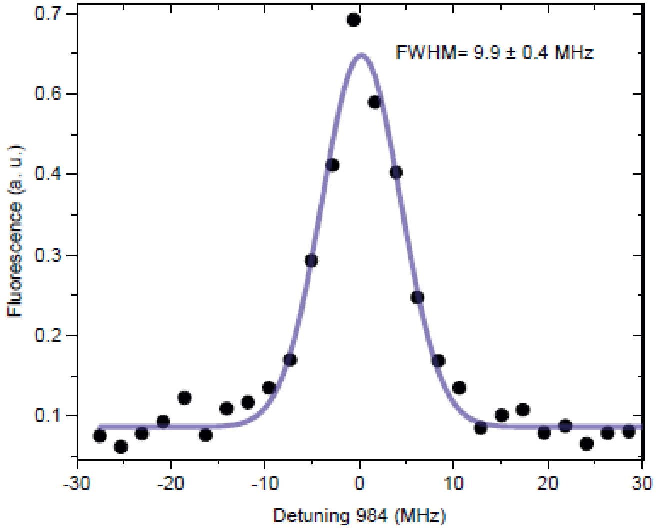
with >20 Th^{3+}

Laser spectroscopy of sympathetically cooled $^{230}Th^{3+}$ ions
(without hyperfine structure, produced as recoil ions from ^{234}U)



Level structure of Th^{3+}

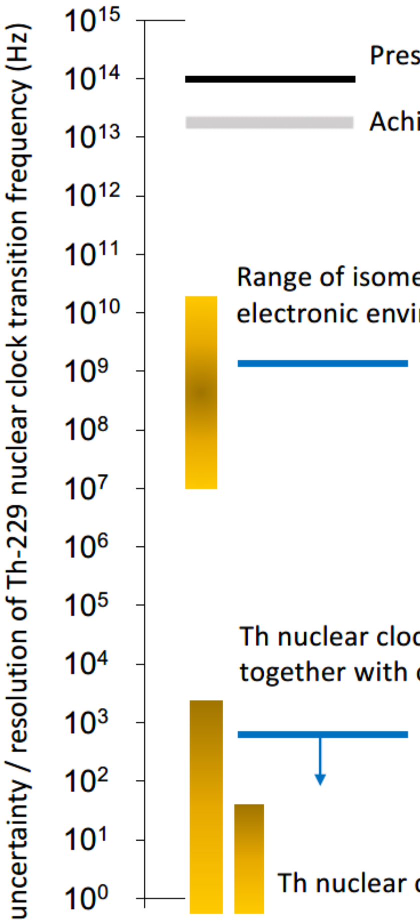




Prospects: precision HFS spectroscopy of ^{229}Th and ^{229m}Th:
→ more precise nuclear moments, determining the α-sensitivity from the Coulomb energy

G. Zitzer, J. Tiedau, M. V. Okhapkin, K. Zhang, C. Mokry, J. Runke, C. E. Düllmann, E. Peik, to be published

„Roadmap“ in frequency uncertainty for the Th-229 nuclear transition



Present uncertainty from γ or e^--spectroscopy: 0.2 eV

Achievable uncertainty with γ or e^--spectroscopy: 0.05 eV

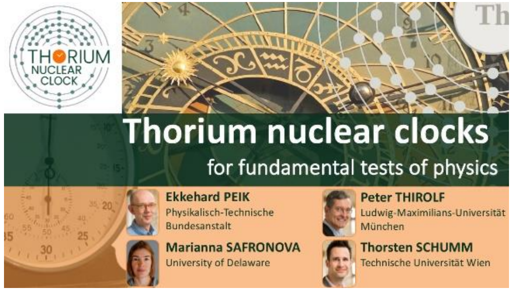
Range of isomer shifts and hyperfine structure in different electronic environments (ions, molecules, solids)

Spectral resolution with ns-VUV laser system

Th nuclear clock applicable for fundamental tests together with other high-accuracy optical clocks

Spectral resolution with single mode of a fs-VUV frequency comb

Th nuclear clock as a high-accuracy optical clock



Thorium nuclear clocks

for fundamental tests of physics

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Marianna SAFRONOVA University of Delaware

Thorsten SCHUMM Technische Universität Wien

ERC Synergy Project 2020

European Research Council
Established by the European Commission

PTB Working Group
Laser Nuclear Spectroscopy

J. Tiedau
J. Thielking
M. Okhapkin
Ke Zhang
G. Zitzer

Positions available !

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