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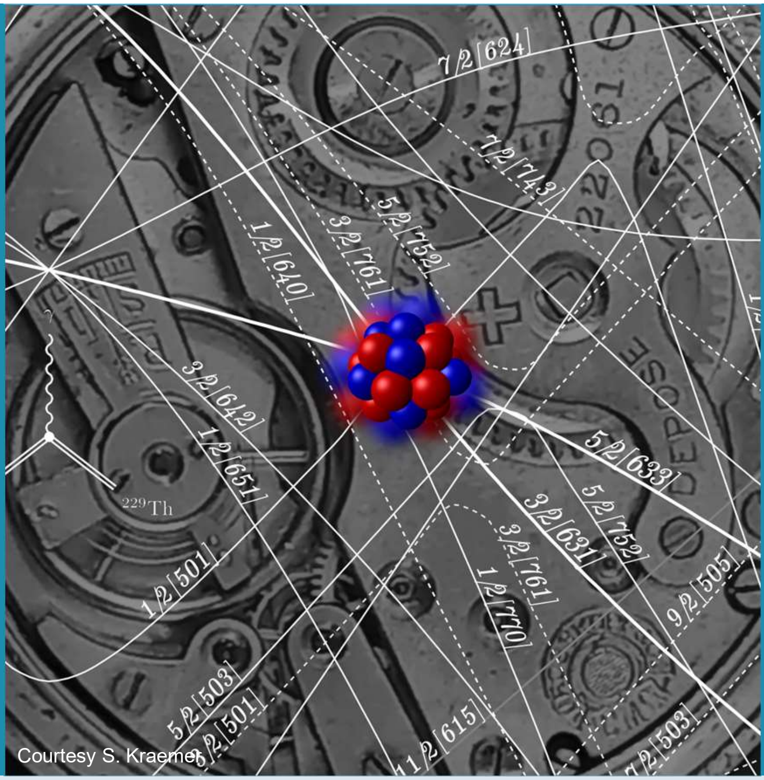
The Th-229 Nuclear Clock Isomer: VUV Spectroscopy Studies

Piet Van Duppen
on behalf of the ISOLDE-IS658 and IS-715 collaboration

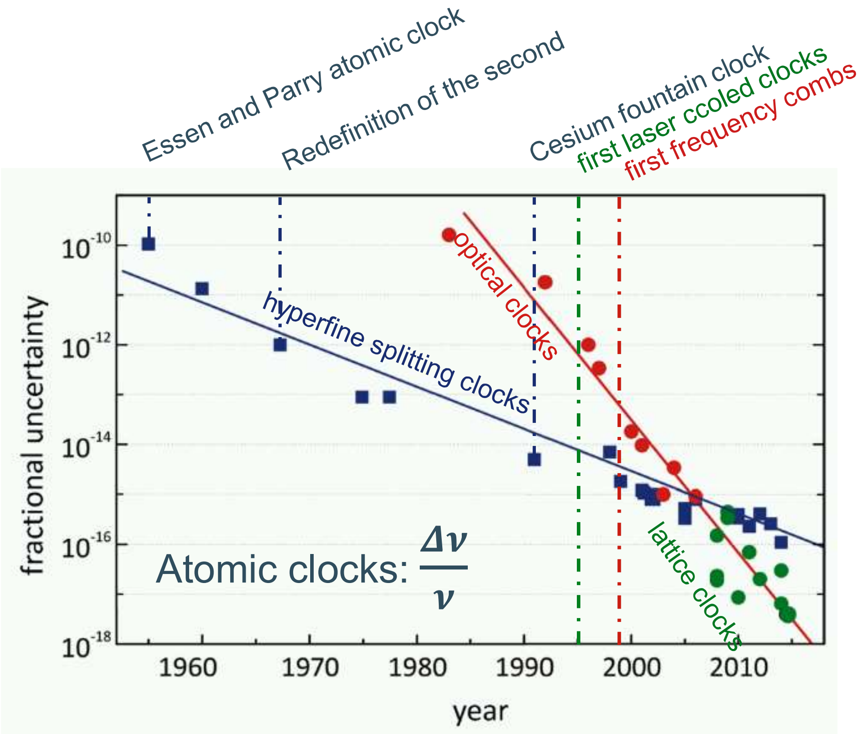


Outline:

- Thorium-229, its isomer and the nuclear clock
- Improve the nuclear-structure information of ^{229,m}Th
  - Vacuum-ultraviolet spectroscopy at ISOLDE
- Recent developments
- Conclusion and Outlook

Thorium-229, its isomer and the nuclear clock

The road towards a nuclear clock



Sr lattice clock: reached in 92 h (Bothwell 2022) $\Delta\nu/\nu_0 = 7.6 \cdot 10^{-21}$

Sr lattice clock: (Zheng 2022) $\Delta\nu/\nu_0 = 9.7 \cdot 10^{-18}\tau^{-1/2}$

Al quantum-logic clock: (Brewer 2019) $\Delta\nu/\nu_0 = 9.4 \cdot 10^{-19}$

Environmental limit: external pertubations

e.g.
- Starkshift & Zeemanshift of external fields
- Blackbody : radiation

Clock stability: $\sigma^2_{\text{Allan}}(\tau) = \frac{1}{2\sum_i 1}\sum_i\left(\frac{\nu_{i+1}}{\nu_0} - \frac{\nu_i}{\nu_0}\right)^2$

Fundamental limit: quantum projection noise

$$\sigma_{Allan} \approx \frac{1}{2\pi\nu}\sqrt{\frac{1}{N\tau T_{1/2}}}$$ (nuclear clock)

ν transition frequency
τ averaging time
N number of interrogated nuclei
$T_{1/2}$ coherence time ≈ half-life

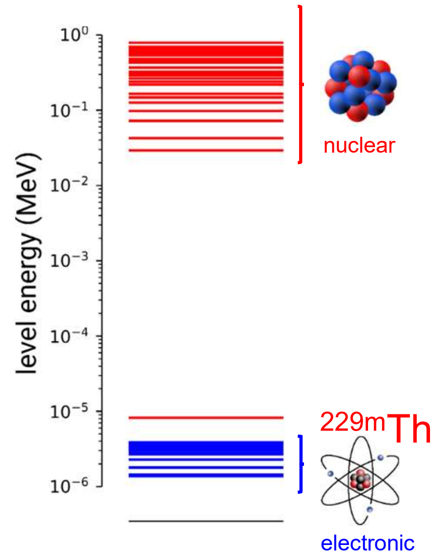
The road towards a nuclear clock (Peik and Tamm Europhys. Lett. 61 (2003) 181)

- A nuclear clock based on ^{229m}Th

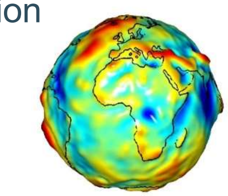
• Nuclear transition
  ⇒ less susceptible to perturbations
• Low-lying isomer
  ⇒ accessible with VUV lasers
• Suitable lifetime transition
  ⇒ favorable $\frac{\Delta E}{E}$ (≈ 10^{-20})
• Ion trap or solid-state* approach
  (*) Probe 10^{15} non-interacting oscillators

- Potential clock operation at 10^{-19} relative precision

- Fundamental physics
• Temporal variation of the fine-structure constant



- Potential applications
• Satellite-based navigation
• Geodesy



Dessovic 2014 *J. Phys. Condens. Matter* **26** 10
Campbell et.al. 2012 *PRL* **108** 12
Peik et al., Quantum Sci. Technol. 6 (2021) 034002
Thirolf et al., Ann. der Physik 531 (2019) 1800381
Flambaum V., Phys. Rev. Lett. 97 (2006) 092502

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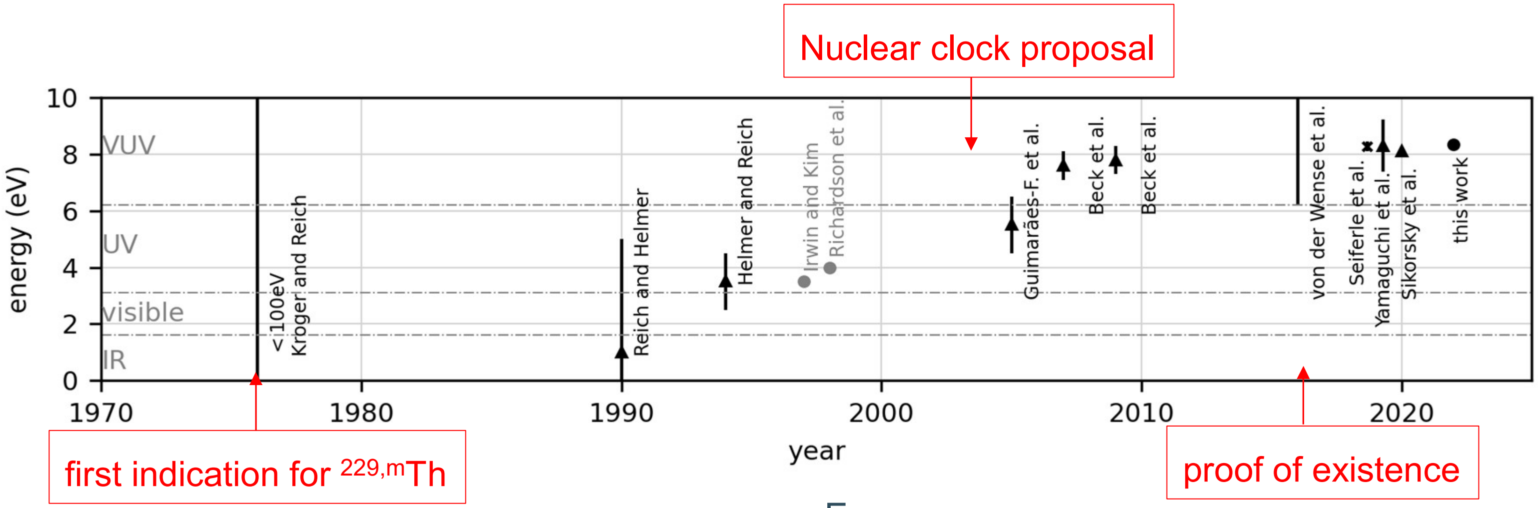
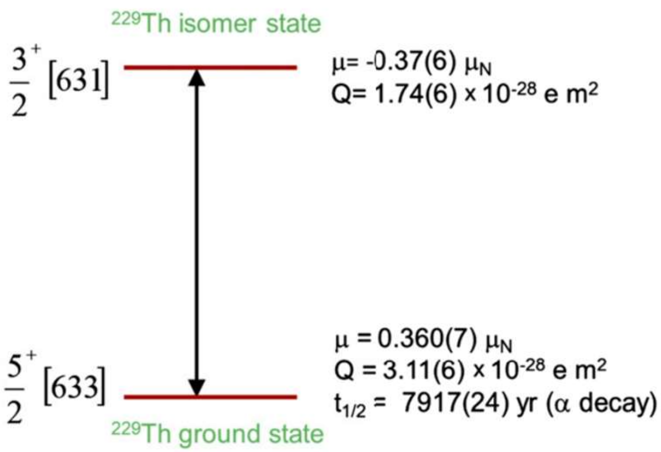




- Energy:

  8.10 (17) eV (153.1 nm) and 8.28 (17) eV (149.7 nm) > IP(Th) = 6.3 eV)

- Competition between Internal Conversion and Radiative decay (charge-state dependent):

  $\alpha \approx 10^9$ (neutral thorium atom)

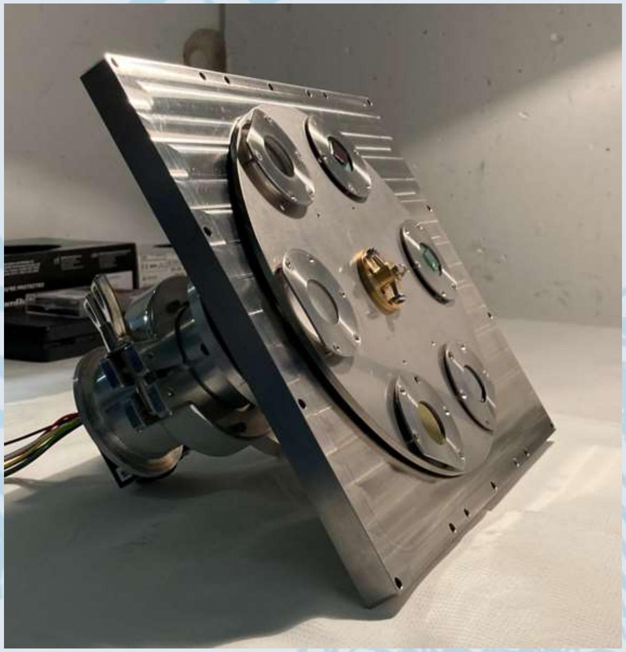
- Lifetime of only IC in neutral charge state

  $T_{1/2}$ (IC) = 7(1) μs

- $T_{1/2}$ (radiative decay, nuclear theory) = $10^3 - 10^4$ s

Korger and Reich, 1976 Nucl. Phys. A259 29
von der Wense et al, 2016 Nature 533 47-51
Seiferle et al., 2019 *Nature* **573** 243-246
Sikorsky et al., 2020 *PRL* **125** 142503
Seiferle et al., 2017 *PRL* **118** 042501
Thielking et al., 2018 *Nature* **556** 321-325
Tkalya et al., 2015 *PRC* **92**
Minkov & Palffy 2021 *PRC* **103**

Improve the nuclear-structure information of $^{229,m}Th$
→ Vacuum ultraviolet spectroscopy at ISOLDE



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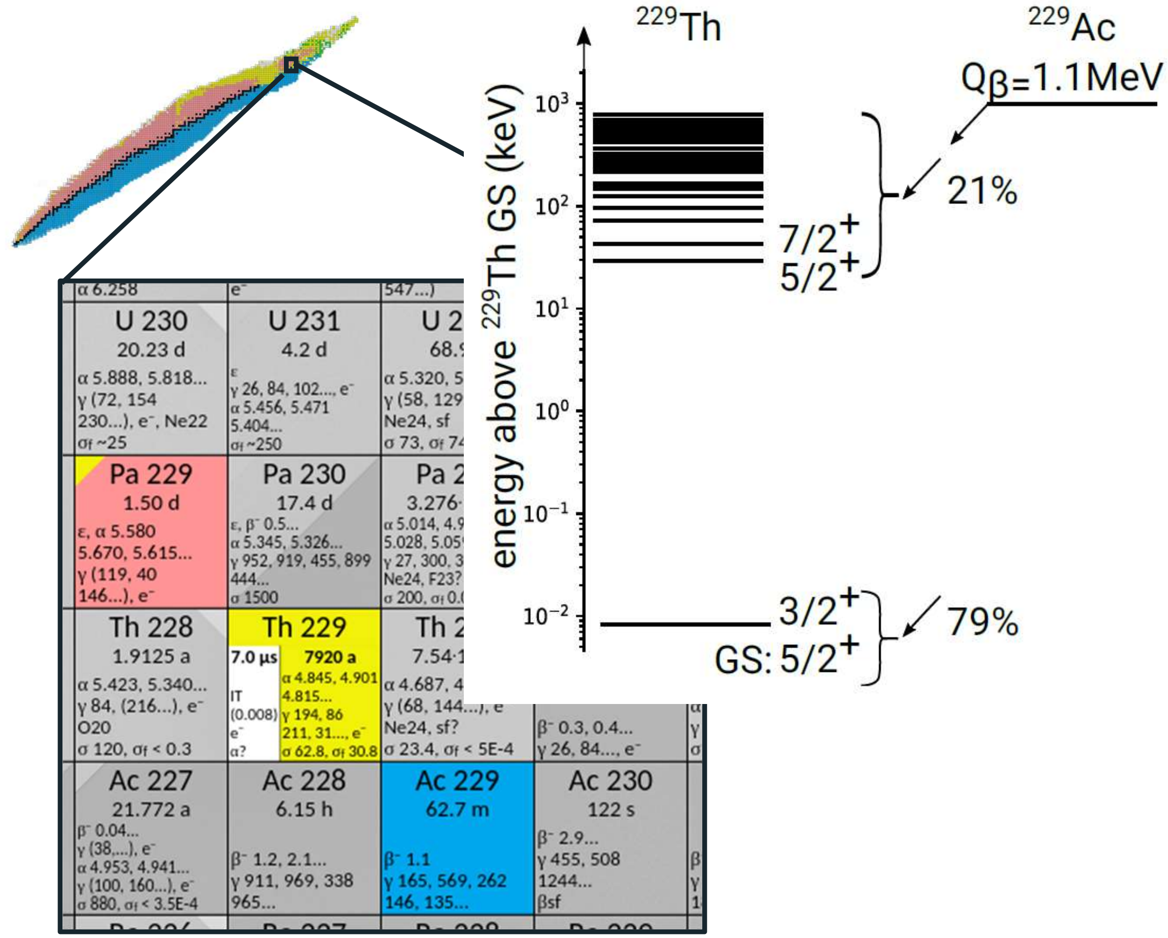
Population of ^{229m}Th

Efficient population in radioactive decay

	^{233}U
Total feeding fraction	2 %
Decay	α
Recoil	84 keV
production	stockpile
technique	doping



Verlinde *et al.*, 2019 *PRC* **100** 024315

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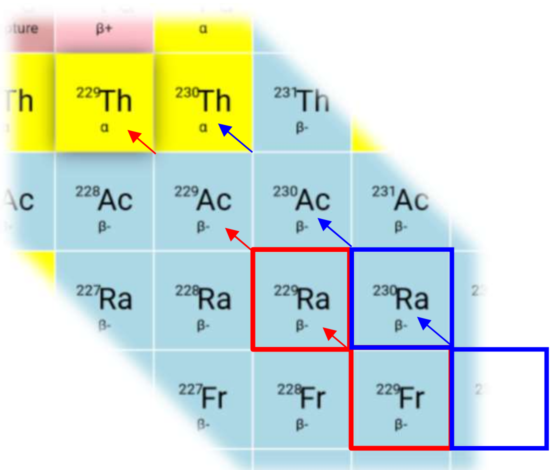
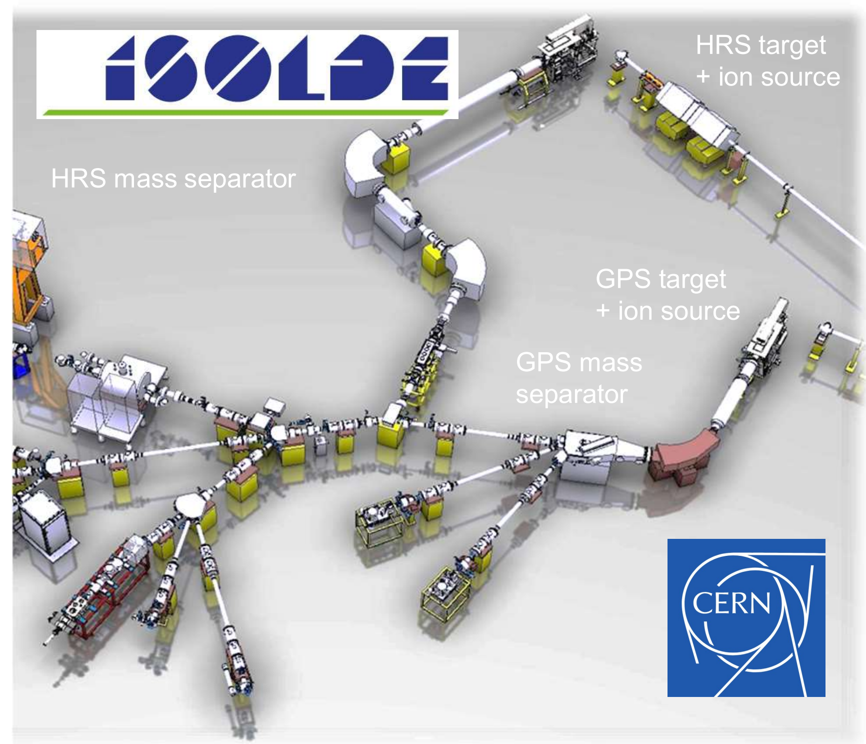
VUV spectroscopy at ISOLDE

- ISOLDE (CERN): 1.4 GeV protons on UCx – surface ionization – implantation at 30 keV
- Beam composition: ^{229}Fr ($T_{1/2}$ = 50.2 s, ~ 10^5 pps), ^{229}Ra (4.0 m, ~ 10^6 pps), ^{229}Ac (62.7 m, < 10^5 pps)
- ^{229}Ac ($T_{1/2}$ = 62.7 m) β^--decay → ^{229m}Th / ^{229}Th
- Implantation in large-bandgap crystals (CaF_2 and MgF_2)
- VUV spectroscopy
- A = 230 and 231 beams used as proxy





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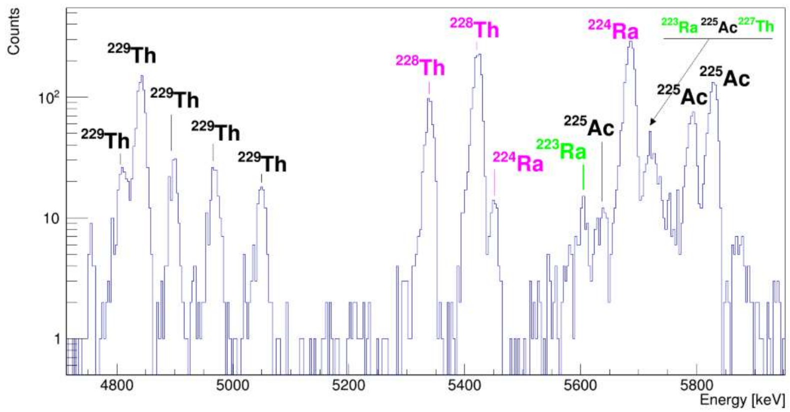
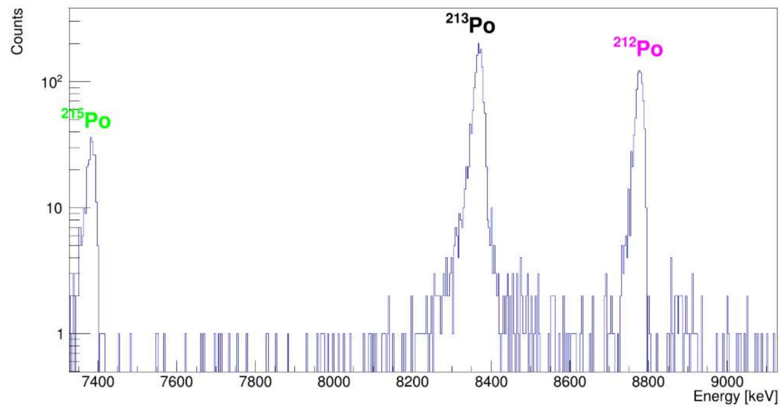
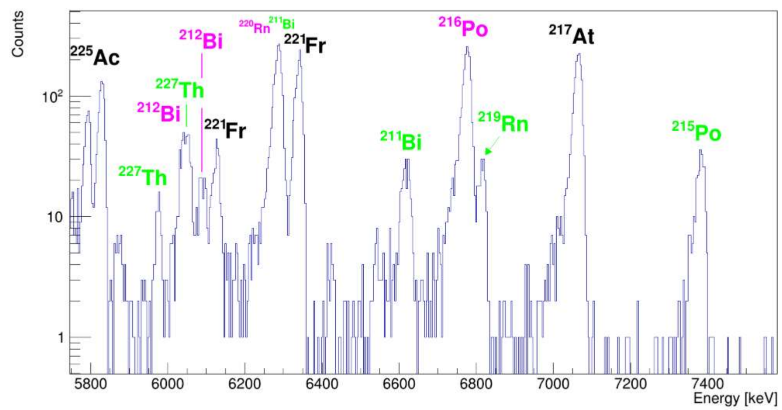
Radioactive beam purity

Black = ^{229}Ac chain, Pink = ^{228}Ac chain, Green = ^{227}Ac chain







- Alpha spectra from CaF_2 irradiated sample (6 months after beam time)
- Contamination in the A=229 beam:
  - A=228 - 0,49(4)%
  - A=227 - 0,05(1)%

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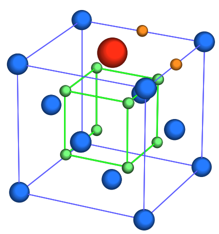
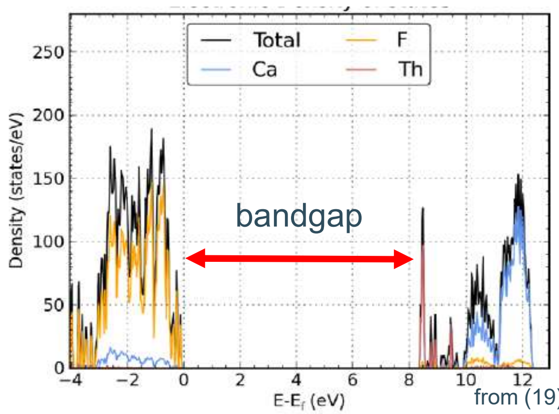
Solid state approach

Embedding in large-bandgap crystals (MgF_2, CaF_2,..) to achieve high charge state (Th^{3+}, Th^{4+})

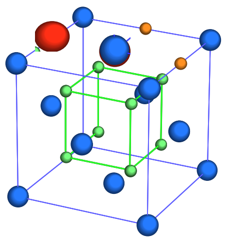


CaF_2: $^{229}Th^{4+}$ in Ca-substitutional position

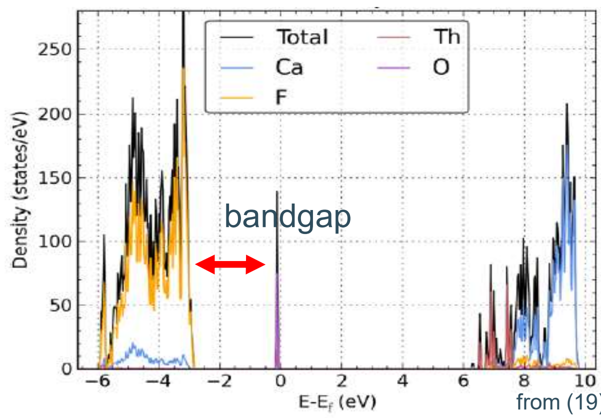


from (19)

radiative decay



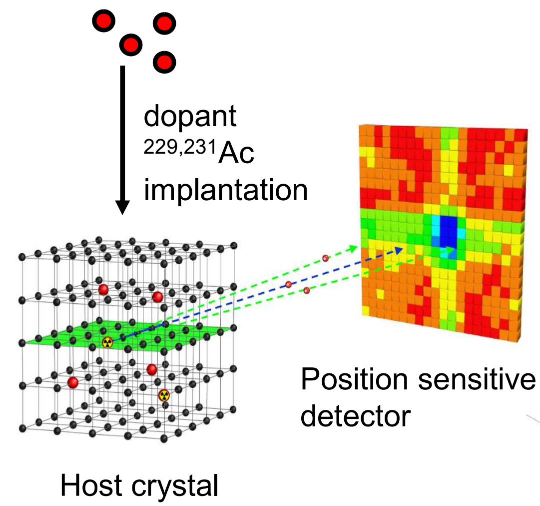
CaF_2: $^{229}Th^{4+}$ in interstitial position



from (19)

electron conversion decay

Emission channeling at ISOLDE (J. Moens et al.)



Kazakov et al., 2012 *New J. Phys.* **14** 083019
Dessovic. et al., 2014 *J. Phys. Condens. Matter* **26** 10

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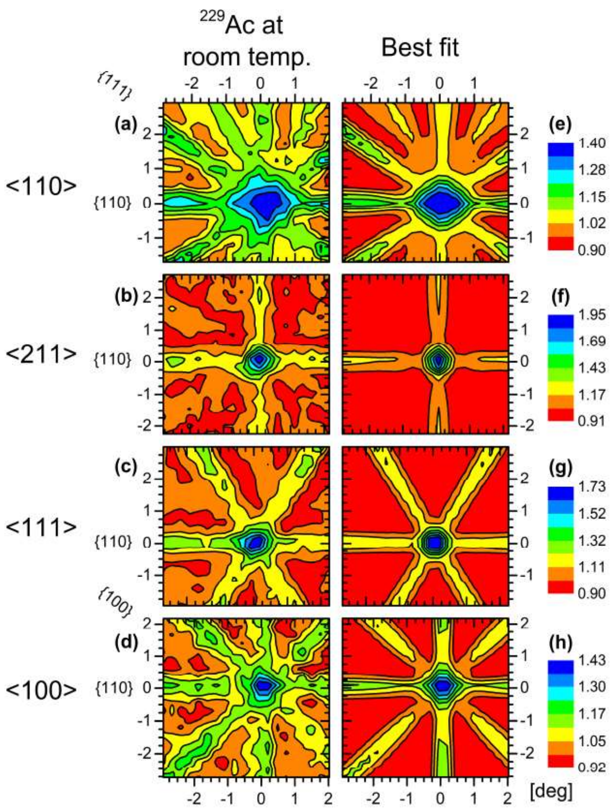
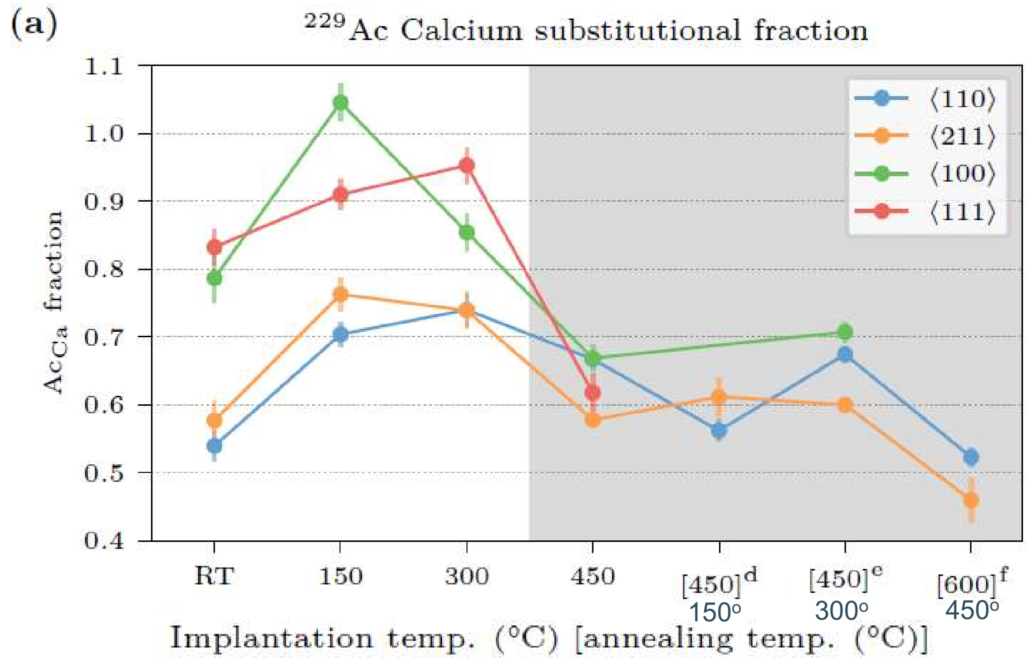
Emission Channeling at ISOLDE

^{229}Ra ($T_{1/2}$ = 4,9 m) → ^{229}Ac ($T_{1/2}$ = 62,7 m) → ^{229}Th

^{231}Ra ($T_{1/2}$ = 104 s) → ^{231}Ac ($T_{1/2}$ = 7.5 m) → ^{231}Th (25.5 h) → ^{231}Pa





- Th and Ac atoms occupy to a large extend Ca substitutional site

J.Moens, L. Perreira, A. Vantomme et al. to be published

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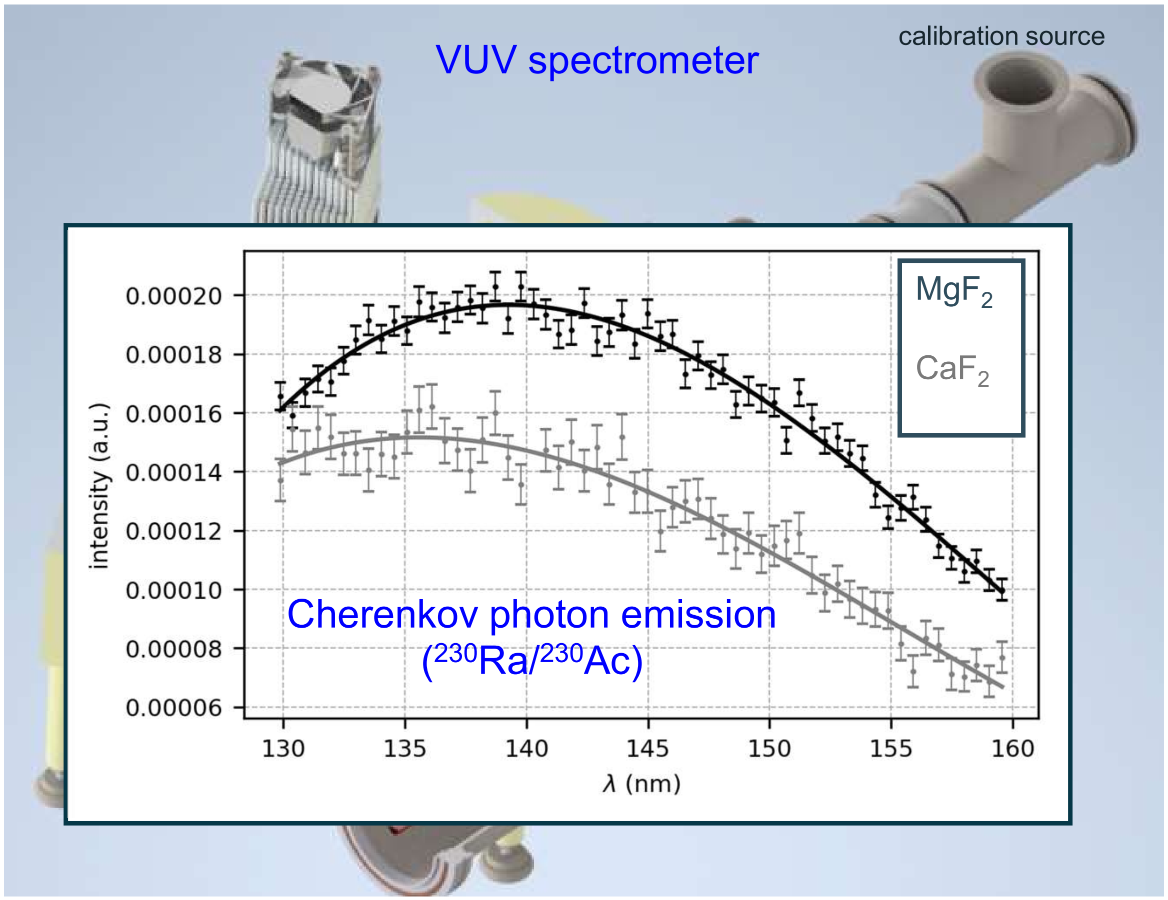
VUV spectroscopy at ISOLDE



Material	Manufacturer	Thickness
MgF_2	Thorlabs Inc.	5 mm
CaF_2	Thorlabs Inc.	5 mm
CaF_2	MaTeck GmbH	0.7 mm
CaF_2	CRYSTAL GmbH	0.5 mm
CaF_2	Imec	50 nm

CaF_2 (E_{gap} = 11.8 eV) and MgF_2 (10.8 eV)

Efficient monochromator:

$$NA \approx F/1.2$$

$$\epsilon_{grating} \approx 40\ \%$$

Single photon counting PMT

$$\epsilon_{detector} \approx 19\ \%$$

Total detection efficiency (3 mm slit)

$$\varepsilon_{total} \simeq 10^{-3}\ at\ 149\ nm$$

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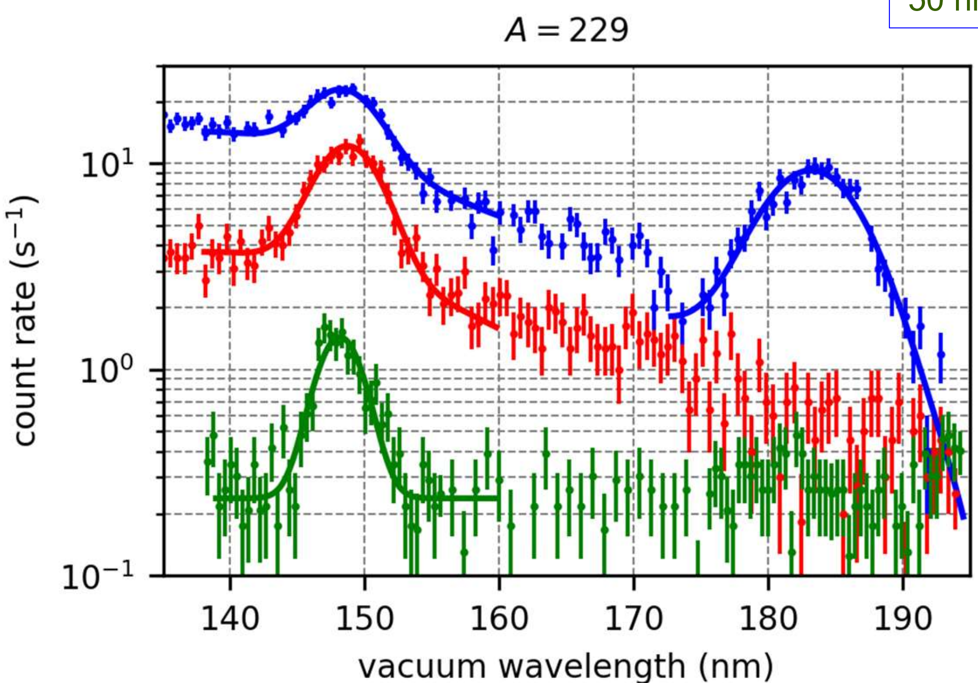
Identification

Taken with 3 mm entrance slit (broad linewidth)

5 mm thick MgF_2
5 mm thick CaF_2
50 nm thick CaF_2



^{229m}Th radiative decay

CaF_2 defect radioluminescence

CaF_2 defect radioluminescence

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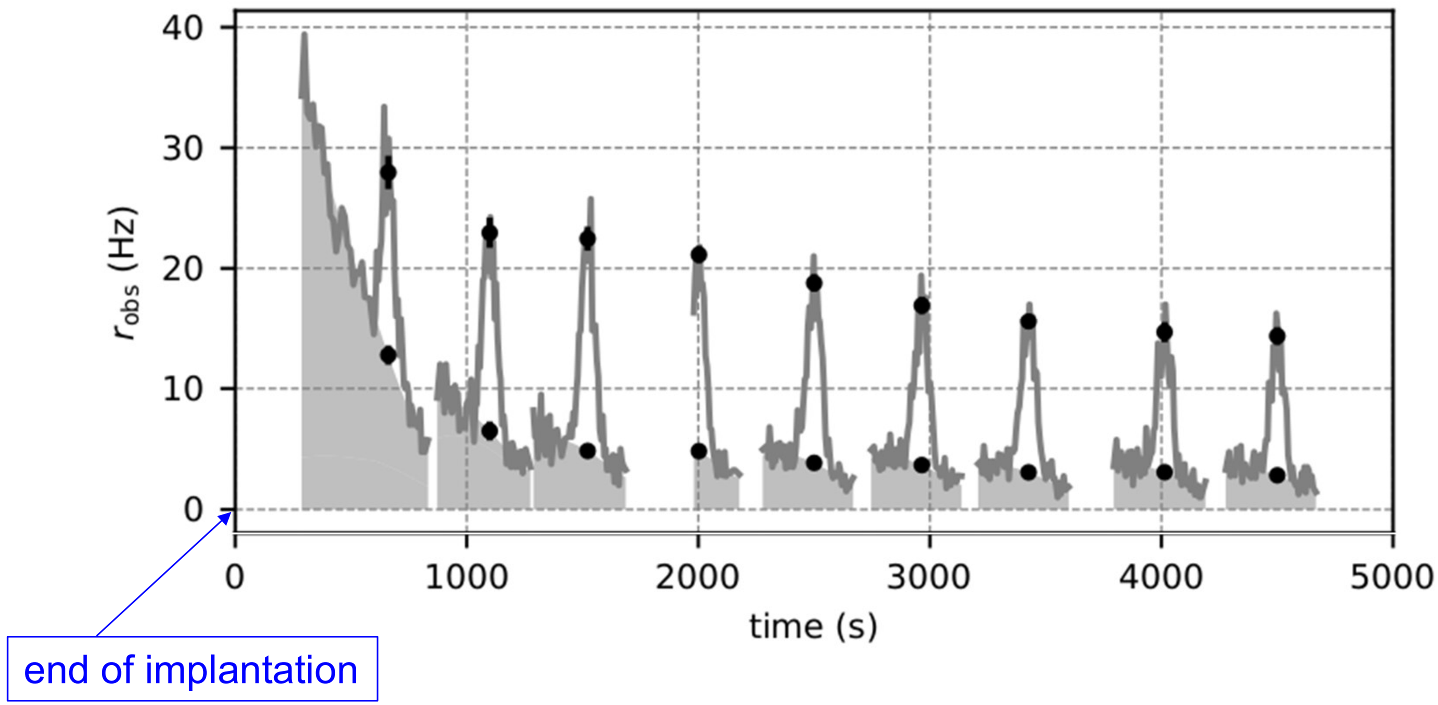
Time behaviour

A = 229 implantation for 3450 s in a MgF_2 crystal



(2 mm entrance slit (broad linewidth))

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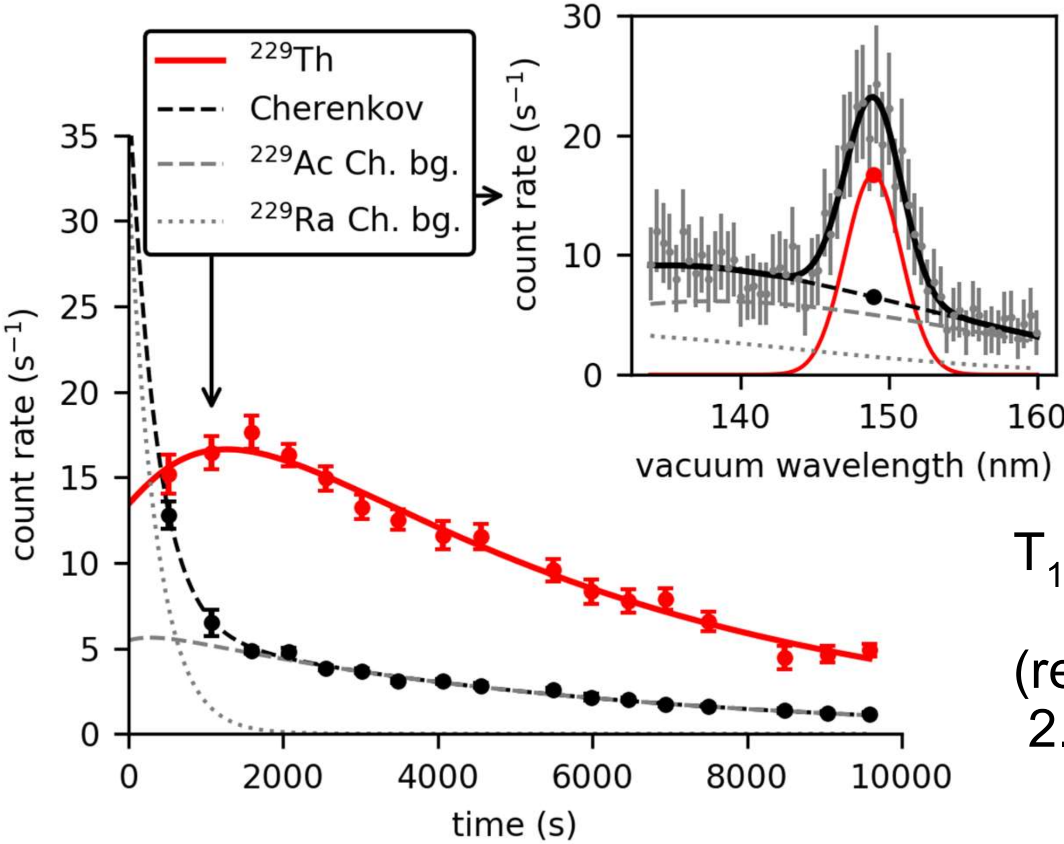
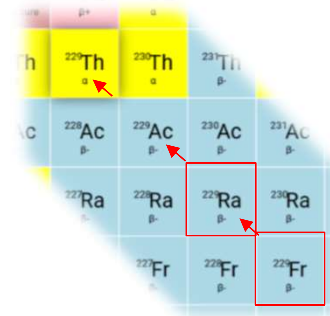
Time behaviour

^{229}Fr ($T_{1/2}$ = 50.2 s) → ^{229}Ra (4.0 m) → ^{229}Ac (62.7 m) → ^{229,m}Th(?)





$T_{1/2}$ = 670 (102) s in MgF_2

(refractive index - n^3 dependence:
2.21(34) 10^3 s)

Tkalya et al., PRC61 (2000)

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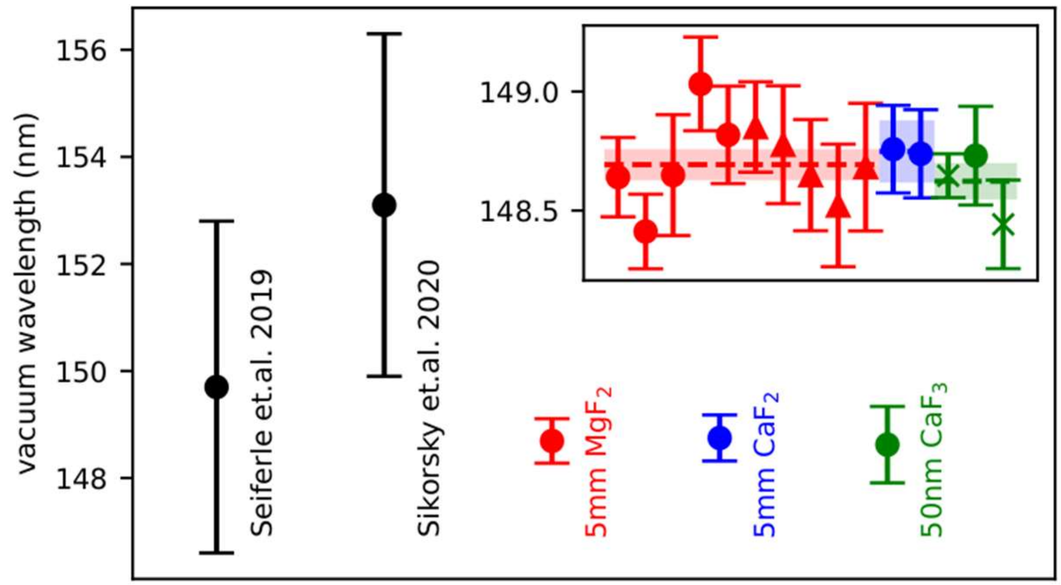
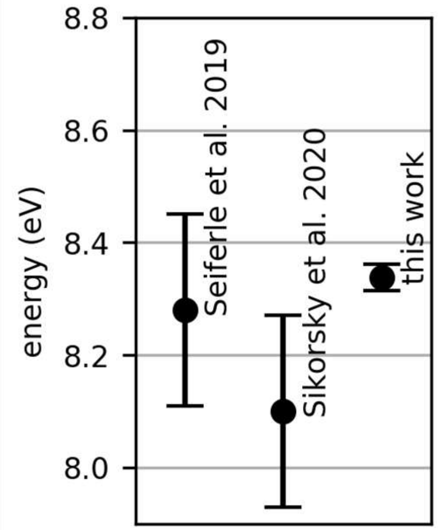
Energy

1 (x), 0.5 (•) and 0.25 (Δ) mm entrance slit





New energy value:
148.71 ± 0.06 (stat.) ± 0.41 (syst.) nm
8.338 ± 0.003(stat.) ± 0.023(syst.) eV

→Uncertainty reduced from 41 THz to 5.8 THz

Article

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

https://doi.org/10.1038/s41586-023-05894-z
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Accepted: 28 February 2023
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Check for updates

Sandro Kraemer[1,2], Janni Moens[3], Michail Athanasakis-Kaklamanakis[1,4], Silvia Bara[1], Kjeld Beeks[5], Premaditya Chhetri[1], Katerina Chrysalidis[4], Arno Claessens[1], Thomas E. Cocolios[1], João G. M. Correia[6], Hilde De Witte[1], Rafael Ferrer[1], Sarina Geldhof[1], Reinhard Heinke[4], Niyusha Hosseini[5], Mark Huyse[1], Ulli Köster[7], Yuri Kudryavtsev[1], Mustapha Laatiaoui[8,9,10], Razvan Lica[4,11], Goele Magchiels[3], Vladimir Manea[1], Clement Merckling[12], Lino M. C. Pereira[3], Sebastian Raeder[9,10], Thorsten Schumm[5], Simon Sels[1], Peter G. Thirolf[2], Shandirai Malven Tunhuma[3], Paul Van Den Bergh[1], Piet Van Duppen[1], André Vantomme[3], Matthias Verlinde[1], Renan Villarreal[3] & Ulrich Wahl[6]

S. Kraemer,- Nature (2023) 617 706-710
S. Kraemer, PhD thesis (2022) KU Leuven

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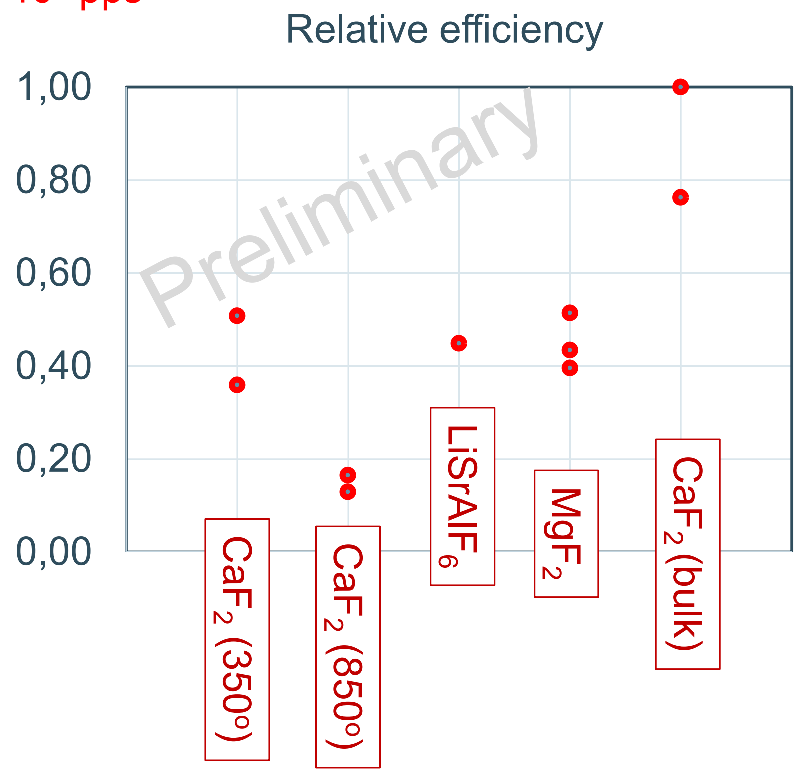
Preliminary results from IS-715 (July 2023)

- Th-C production target at ISOLDE/CERN: ^{229}Ra about 2 10^8 pps
- Different crystals (bandgap energy)

CaF_2 (11.8 eV)	✓
MgF_2 (10.8 eV)	✓
$LiSrAlF_6$ (10.7 eV)(*)	✓
SiO_2 (8.9 eV)	< 2 10^{-3}
AlN (6.0 eV)	< 2 10^{-3}



Absolute efficiency (CaF_2 bulk) > 1.9 – 13.1 % (depending on γ intensity)

(*) E. Hudson - UCLA

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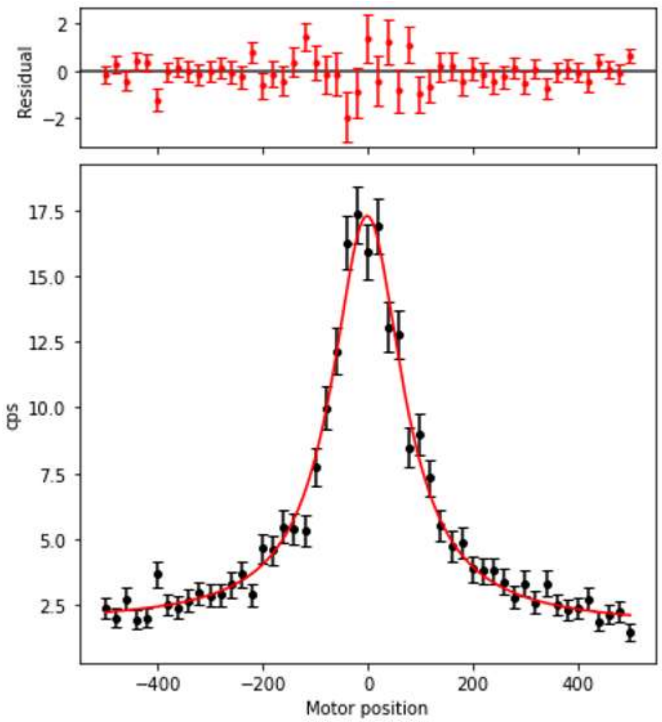
Preliminary results from IS-715 (July 2023)

- Th-C production target at ISOLDE/CERN: ^{229}Ra about 2 10^8 pps
- Different crystals (bandgap energy)
- Energy measurements: slit width 250 μm / CaF_2(thin)



Preliminary

N_{imp}	λ_{229m} (nm)	
7	148.479 ± 0.033	148,491 ± 0,019 (stat.) ± 0,1 (syst.) nm (N = 7, 15, 21, 23)
15	148.503 ± 0.041	
21	148.510 ± 0.037	
23	148.476 ± 0.040	
27	148.402 ± 0.086	$LiSrAlF_6$

<u>Literature:</u>

148.71 ± 0.06 (stat.) ± 0.41 (syst.) nm (Kraemer et al., Nature 2023)
148,18 ± 0.38 (stat.) ± 0.19 (syst.) nm (Hiraki et al., arXiv May 4, 2024)

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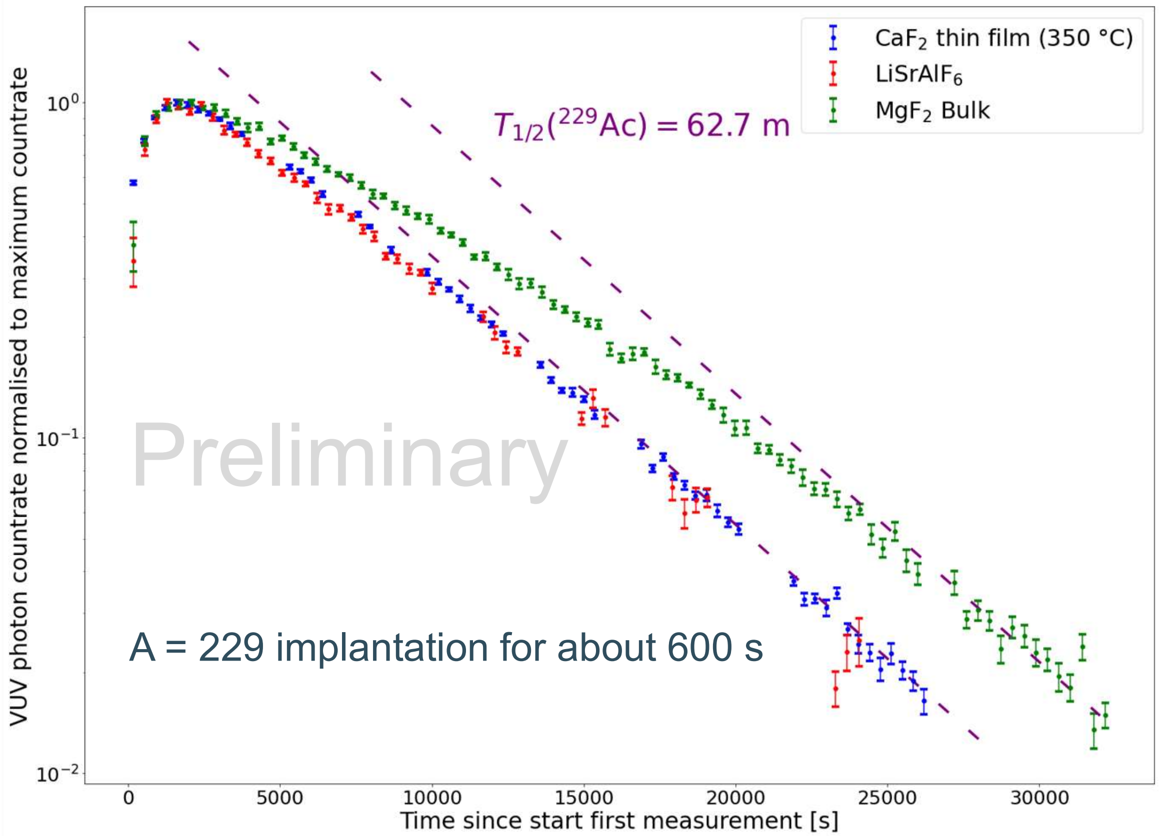
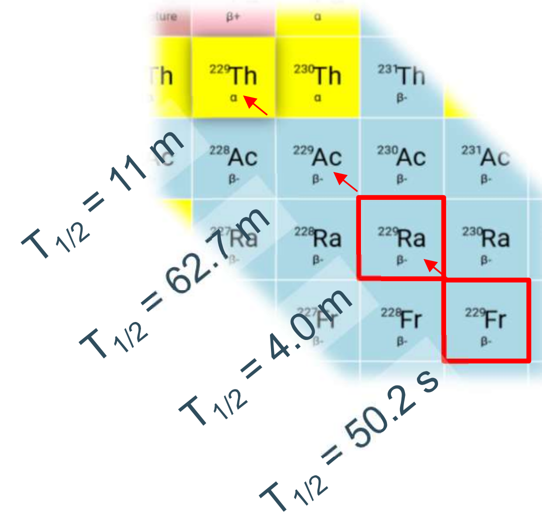
Preliminary results from IS-715 (July 2023)





- Sites with different half-life?
- Defect annealing ($\tau_{annealing} \approx$ hours)?
- Quenching of isomer due to radioactivity (cf. X-ray quenching: Hiraki et al. ArXiv 2204)?

➢ Determining $T_{1/2}$ in different crystals

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(Very) recent progress (I): energy

PHYSICAL REVIEW LETTERS **132,** 182501 (2024)

Laser Excitation of the Th-229 Nucleus

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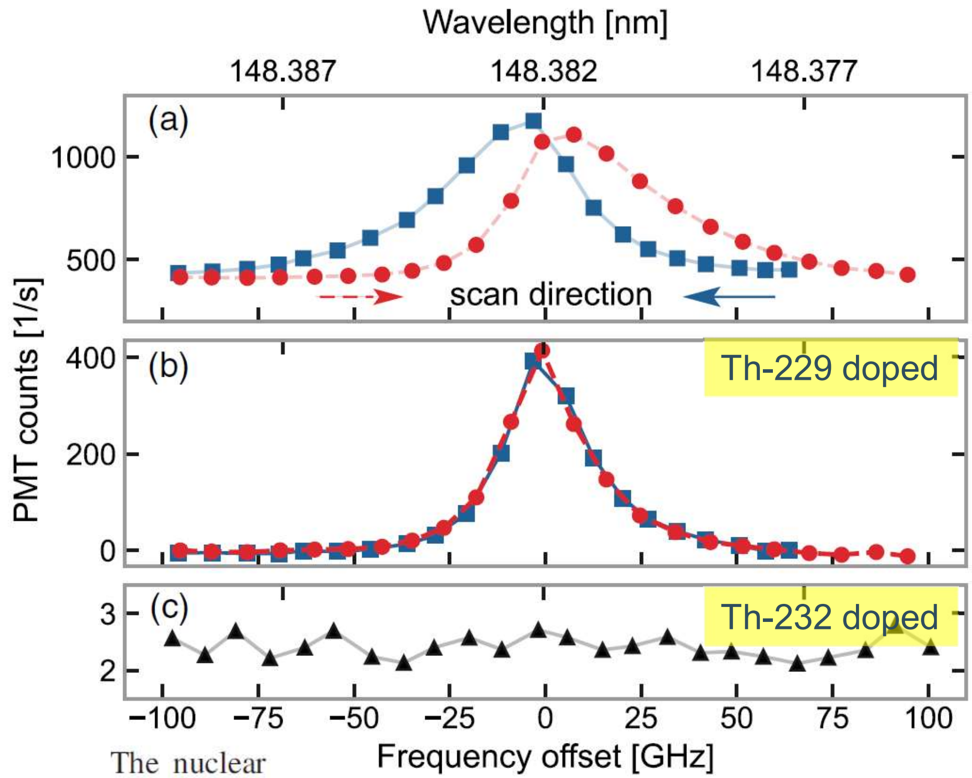
Physikalisch-Technische Bundesanstalt, 38116 Braunschweig, Germany

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‡
Vienna Center for Quantum Science and Technology, Atominstitut, TU Wien, 1020 Vienna, Austria

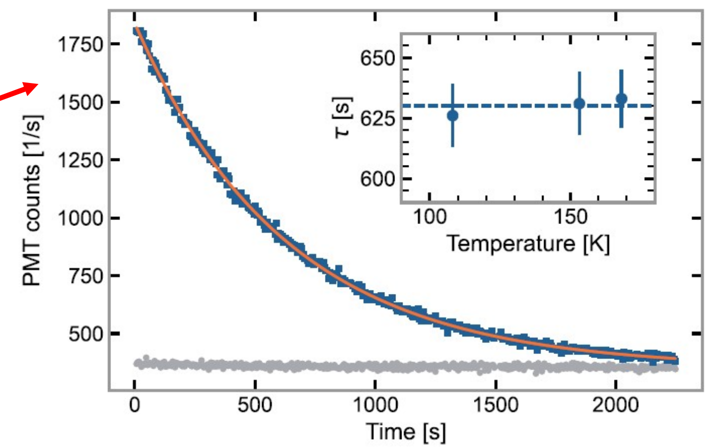


The nuclear resonance for the Th^{4+} ions in $Th:CaF_2$ is measured at the wavelength 148.3821(5) nm, frequency 2020.409(7) THz, and the fluorescence lifetime in the crystal is 630(15) s, corresponding to an isomer half-life of 1740(50) s for a nucleus isolated in vacuum. These results pave the way toward Th-229 nuclear laser spectroscopy and realizing optical nuclear clocks.

- Wavelength confirmed R. Elwell,- arXiv 2024
- Uncertainty reduced from 41 THz to 5.8 THz to 0,007 THz

(Very) recent progress (II): radiative decay half life

Host	$T_{1/2}$ (s)	n	Ref.
Th: MgF_2	2.210(340)	1,488	Kraemer,- Nature (2023)
Th: CaF_2	1.740(50)	1,586	Tiedau,- PRL (2024)
Th^{+3}	1.400 (+600/-300)	-	Yamaguchi,- Nature (2024)
Th:$LiSrAlF_6$	2.684 $(43)_{stat}$ $(66)_{sys}$	1,485	Elwell,- arXiv (2024)
Th:CaF_2	1,790 $(64)_{stat}$ $(80)_{sys}$	1,588	Hiraki,- arXiv (2024)



Laser spectroscopy of triply charged ^{229}Th isomer for a nuclear clock

Nature 629 (2024) 62–66

https://doi.org/10.1038/s41586-024-07296-1

Received: 24 September 2023

Atsushi Yamaguchi[1,2,3], Yudai Shigekawa[4], Hiromitsu Haba[4], Hidetoshi Kikunaga[5], Kenji Shirasaki[6], Michiharu Wada[7] & Hidetoshi Katori[1,2,8]

Sympathetic cooling of trapped Th^{3+} alpha-recoil ions for laser spectroscopy

G. Zitzer,[1] J. Tiedau,[1] M. V. Okhapkin,[1] K. Zhang,[1] C. Mokry,[2,3] J. Runke,[2,4] Ch. E. Düllmann,[2,3,4] and E. Peik[1]

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PHYSICAL REVIEW A **109**, 033116 (2024)

Conclusion and outlook

Conclusion and Outlook

- Observed the radiative decay of ^{229m}Th populated via the beta decay of ^{229}Ac at ISOLDE
  - Isomer's energy = 8,388(24) eV - $T_{1/2}$ = 670(102) s (in MgF_2)
- Implantation technique: good performance for large band-gap and thin-film crystals
- Photon emission observed in MgF_2, CaF_2 and $LiSrAlF_6$ crystals (consequences for a solid-state-based ^{229m}Th clock)
- Time behaviour of VUV signal influenced by defect annealing, different implantation sites or radiation induced quenching?

  → Developing models to describe the data – extract the radiative-decay half life in different crystals

  → Annealing and implantation studies in different crystals ($BaLiF_3$, $LiCaAlF_6$, …)

  Gong et al., Phys. Rev. A109 (2024) 033109

  Pimon et al., Adv. Theory Simul. 5 (2022) 2200185

- Laser excitation of the isomer in a CaF_2 crystal and symphetatic cooling of Th^{+3} ions has been reported (Tiedau,- PRL 2024 – Zitzer,- PRA 2024)

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Outlook: Development towards a nuclear clock

Characterization of nuclear properties → Hyperfine interaction *with crystalline environment* → Laser excitation of the isomer *at MHz to GHz level* → Nuclear spectroscopy *at Hz to kHz level* → Clock development

<u>*VUV-spectroscopy at ISOLDE:*</u>

- *Half-life in additional large bandgap crystals, energy determination at 0.1 nm uncertainty*
- *Study of suitable material for solid-state clocks*
- *Study of the decay-rate-dependence on the material*

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ISOLDE: IS658 – IS715

M. Athanasakis, M. Au, S. Bara, K. Beeks, P. Chhetri, A. Claessens, T. Cocolios, Y. Elskens, J.G. Correia, S.Cottenier, H. De Witte, R. Ferrer, S. Geldhof, N. Hosseini, S. Kraemer, U Köster, M. Laatiaoui, R. Lica, G. Magchiels, V. Manea, J. Moens, I. Moore, L. M. Pereira, S. Pineda, S. Raeder, S. Rothe, T. Schumm, S.Sels, S. Stegeman, P.G. Thirolf, M. Tunhuma, P. Van Den Bergh, P. Van Duppen, A. Vantomme, R. Villareal, M. Verlinde, U. Wahl