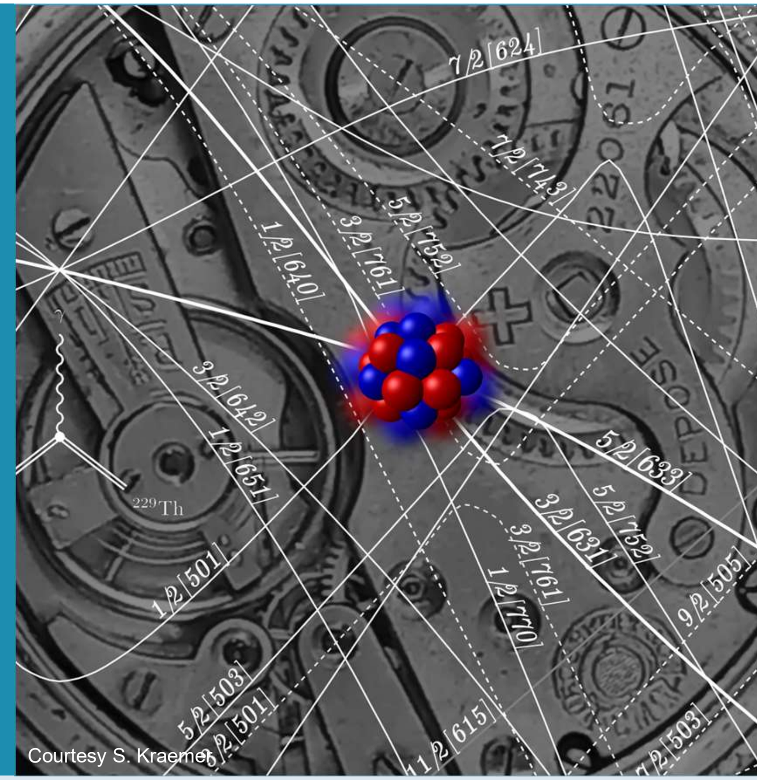
The Th-229 nuclear clock isomer: half life and energy determination in different crystals

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
- Conclusion

Thorium-229, its isomer and the nuclear clock

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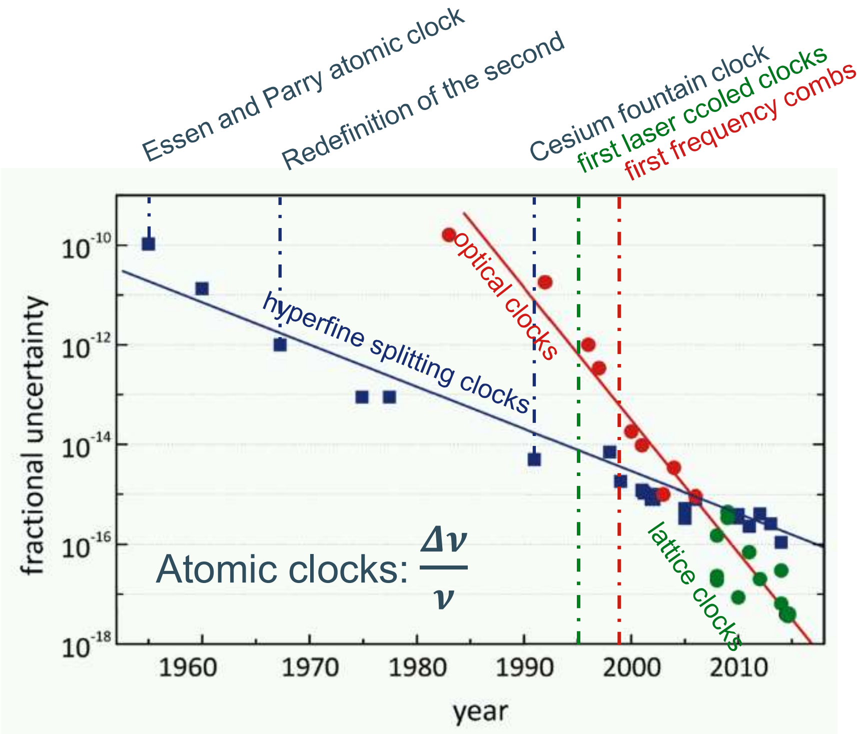
The road towards a nuclear clock



Fractional frequency uncertainty:

Environmental limit: external pertubations

e.g.

- Starkshift & Zeemanshift of external fields
- Blackbody : radiation

Best experimentally realized value: $\Delta\nu/\nu_0 = 7.6 \cdot 10^{-21}$
Al^{+} lattice clock, reached in 92 h

Clock stability:

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

Best experimentally realized value: Sr lattice clock $9.7 \cdot 10^{-18}\tau^{-1/2}$

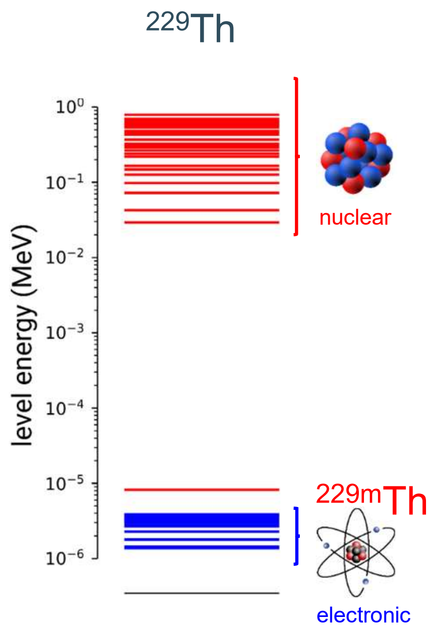
Modified from Riehle, 2016 *C.R.Physiques* **16** 506-515
Bothwell,- 2022, Nature 602 420-422
Zheng,- 2022, Nature 602 425-430
Brewer,- 2019, PRL 123 033201

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

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

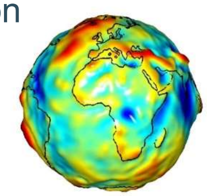
- Nuclear transition
  ⇒ less susceptible to perturbations
- Low-lying isomer
  ⇒ accessible with VUV lasers
- Long lifetime transition
  ⇒ favorable $\frac{\Delta E}{E}$ (≈ 10^{-20})
- Ion trap or solid-state* approach
  (*) Probe 10^{15} non-interacting oscillators

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



- ➢ Fundamental physics
- Temporal variation of the fine-structure constant
- Dark matter searches

- ➢ 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

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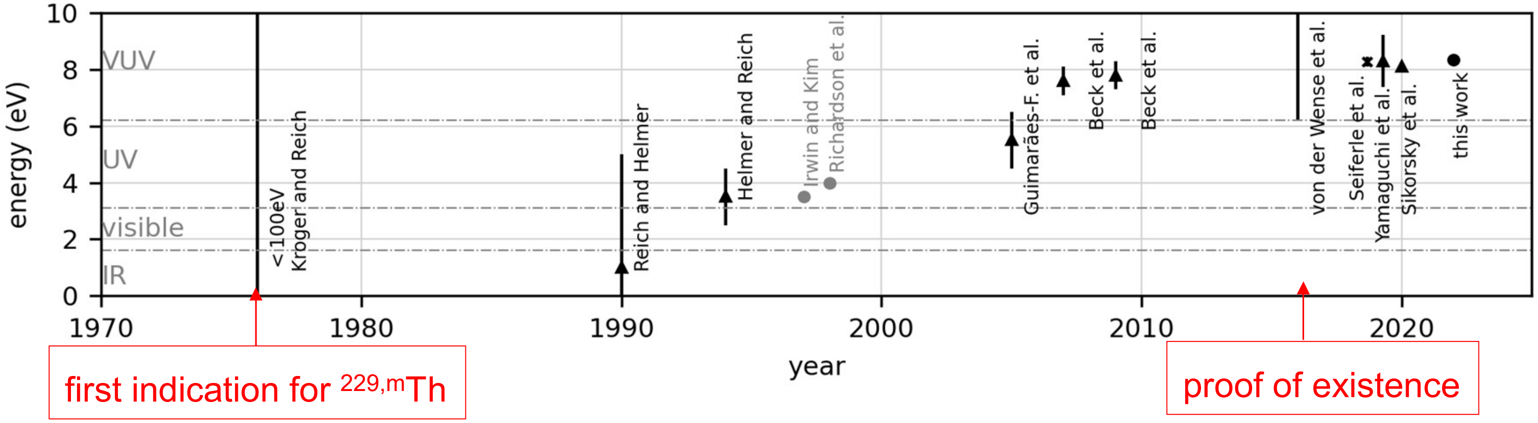

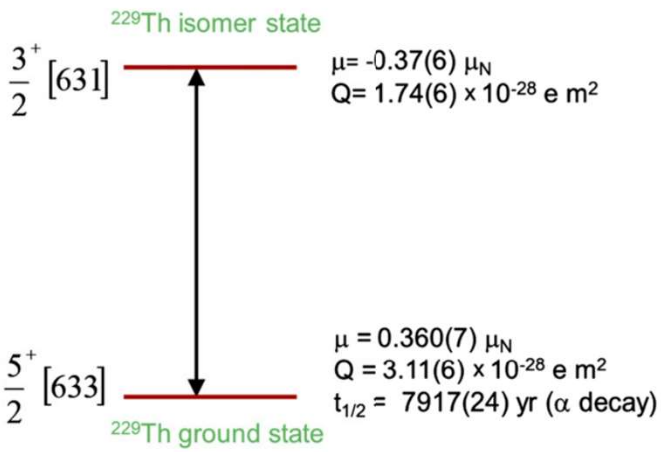

- Energy:
  - 8.10 (17) eV (153.1 nm) and 8.28 (17) eV (149.7 nm) > IP(Th) = 6.3 eV)
- Competition between IC and Radiative decay (charge-state dependent):
  - $\alpha \approx 10^9$ (neutral thorium atom)
- Lifetime of only IC in neutral charge state
  - $T_{1/2}$ (IC) = 7(1) μs
- $T_{1/2}$ (radiative decay, theory) = $10^3 - 10^4$ s
- Laser spectroscopy on isomer and ground state on $^{229m}Th^{+2}$
  - nuclear moments, charge radii

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**

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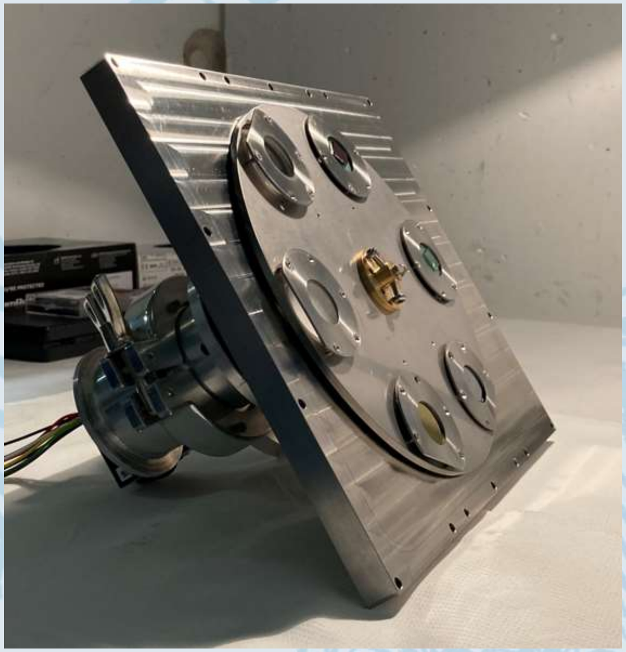
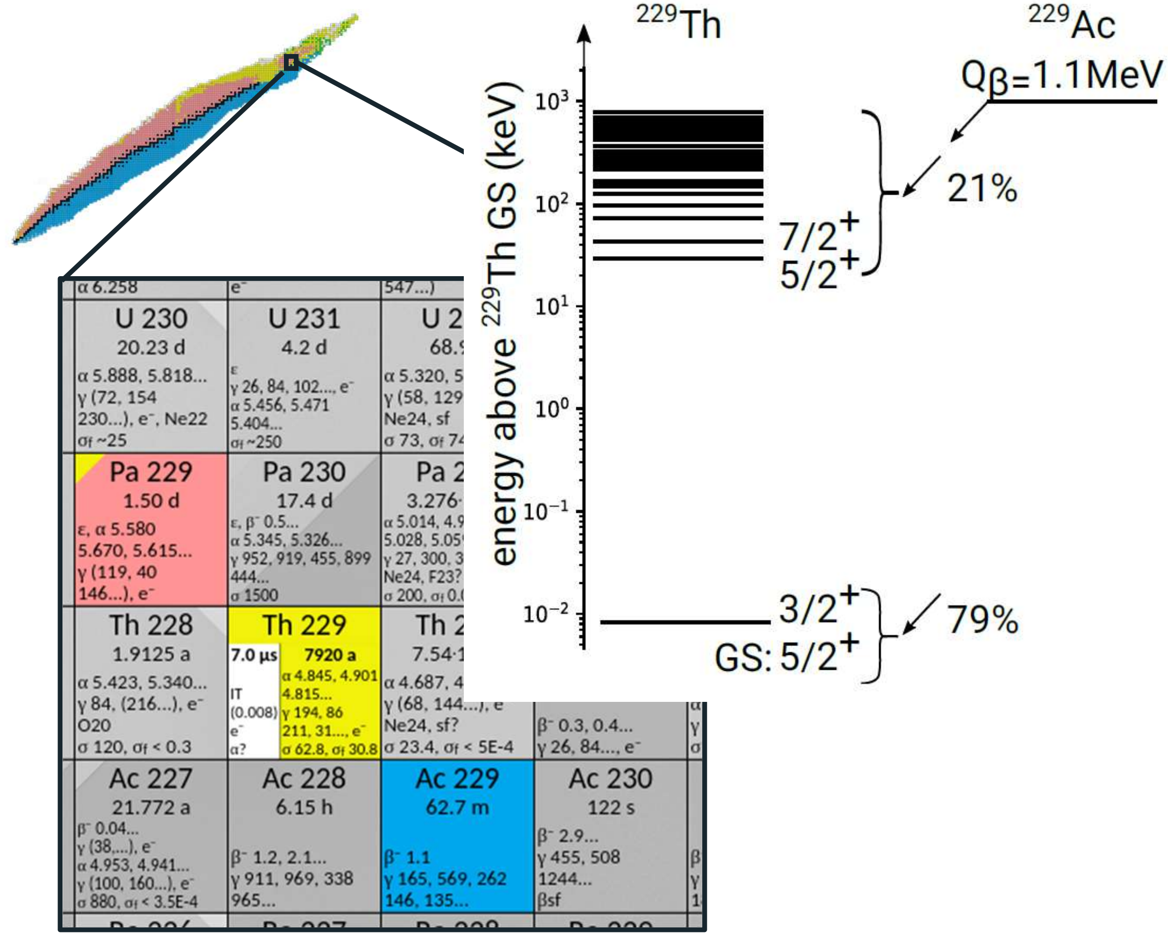
Improve the nuclear-structure information of ^{229,m}Th
→ Vacuum ultraviolet spectroscopy at ISOLDE



Population of ^{229m}Th

Efficient population in radioactive decay

	^{233}U	^{229}Ac
Total feeding fraction	2 %	14 – 93 %
Decay	α	β⁻
Recoil	84 keV	< 6 eV
production	stockpile	ISOL
technique	doping	implantation



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

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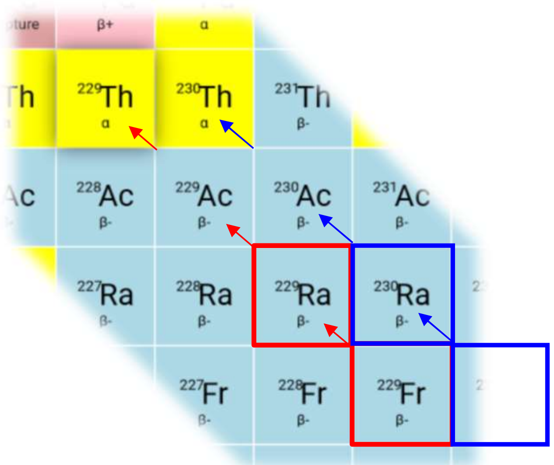
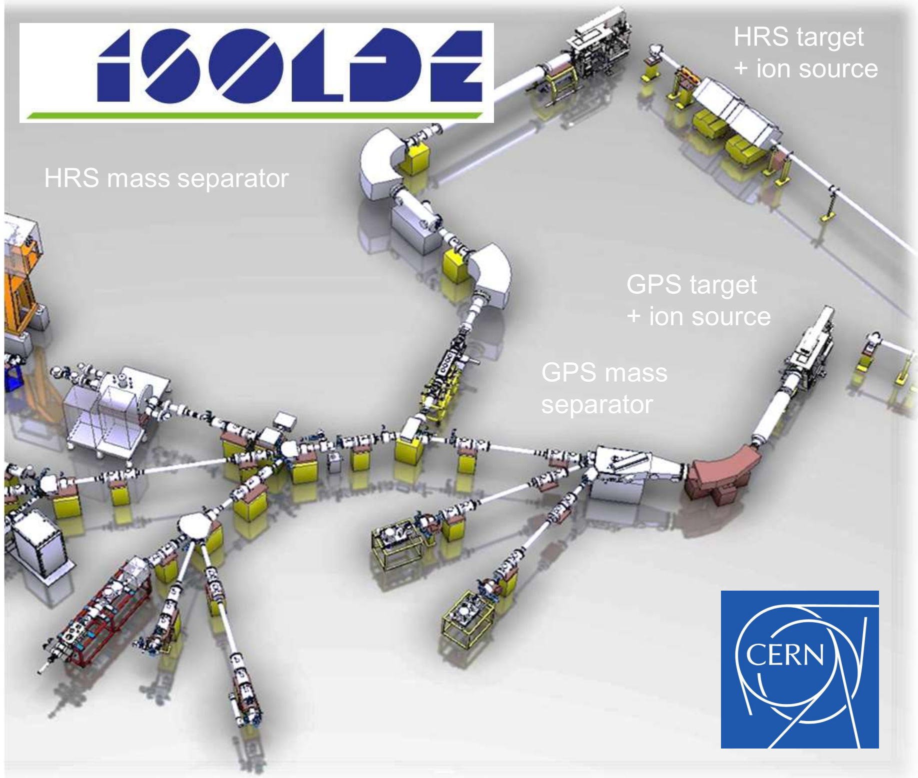
VUV spectroscopy at ISOLDE

- 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) → ^{229m}Th / ^{229}Th
- Implantation in large-bandgap crystals and VUV spectroscopy
- A = 230 and 231 beams used as proxy





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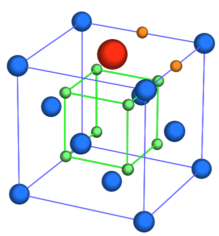
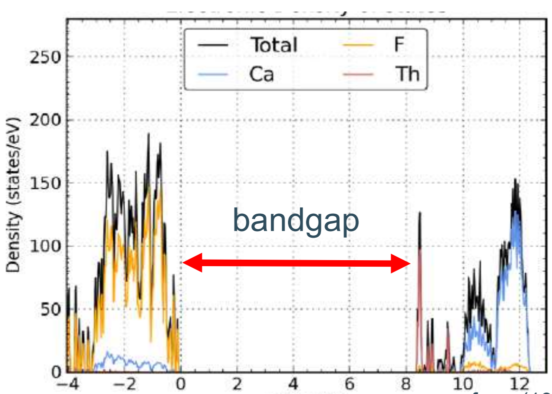
Solid state approach

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

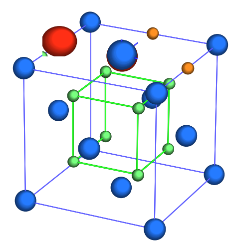


CaF_2: $^{229}Th^{4+}$ in Ca-substitutional position

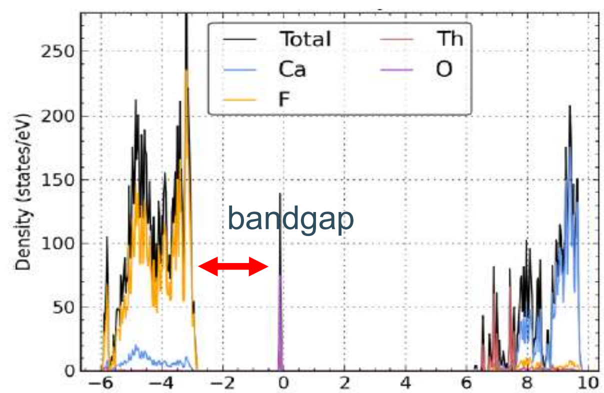


from (19)

radiative decay



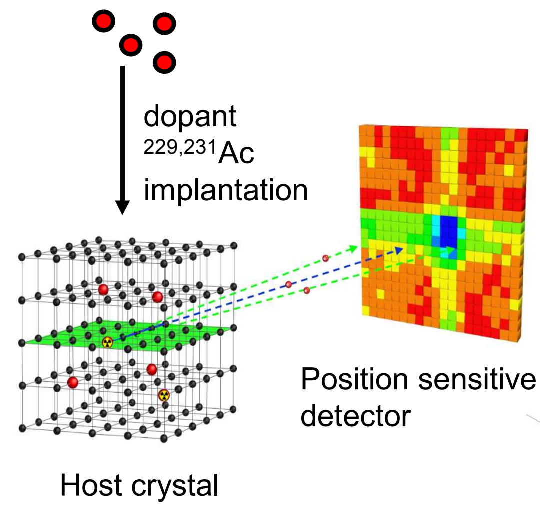
CaF_2: $^{229}Th^{4+}$ in interstitial position



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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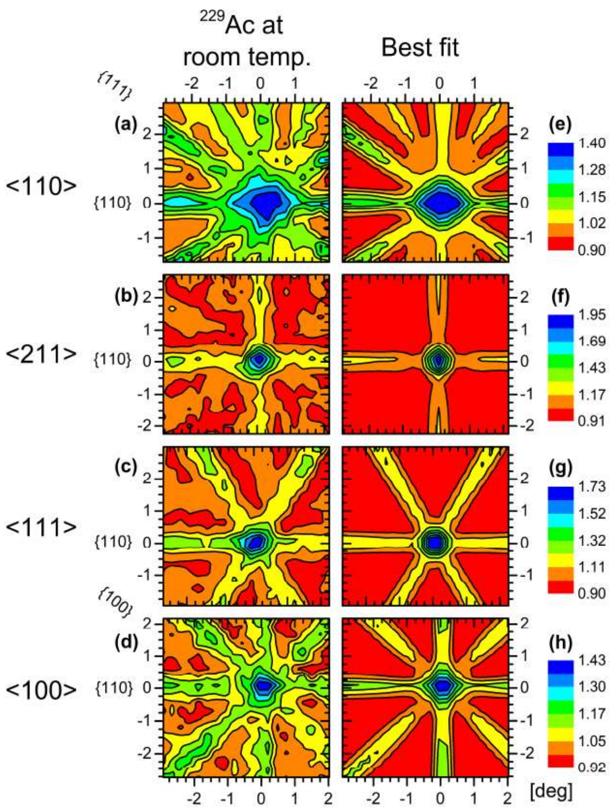
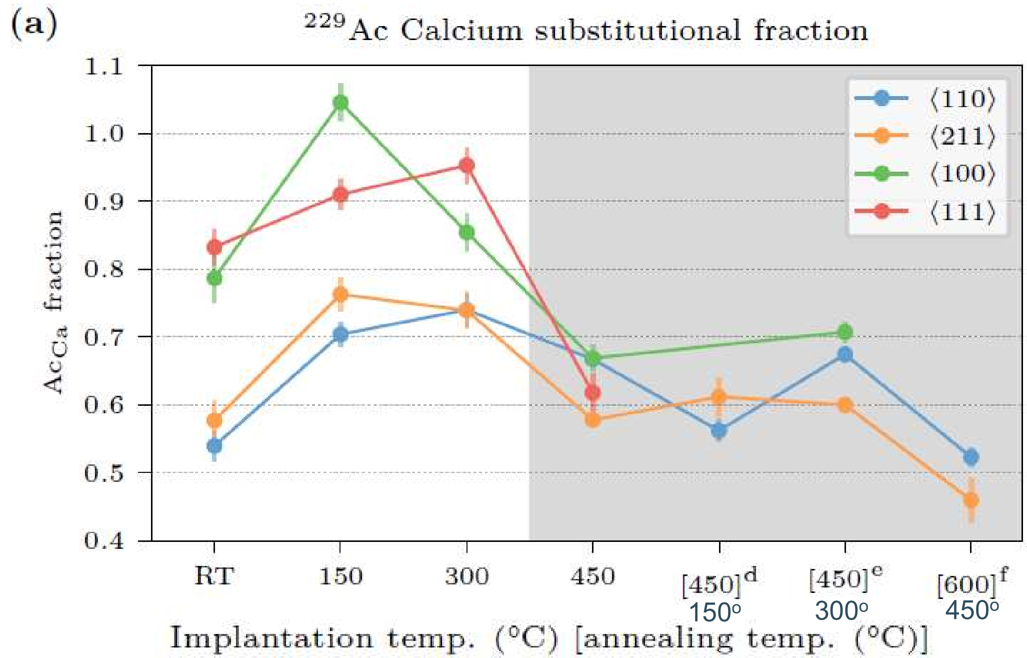
Emission Channeling at ISOLDE

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

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





- 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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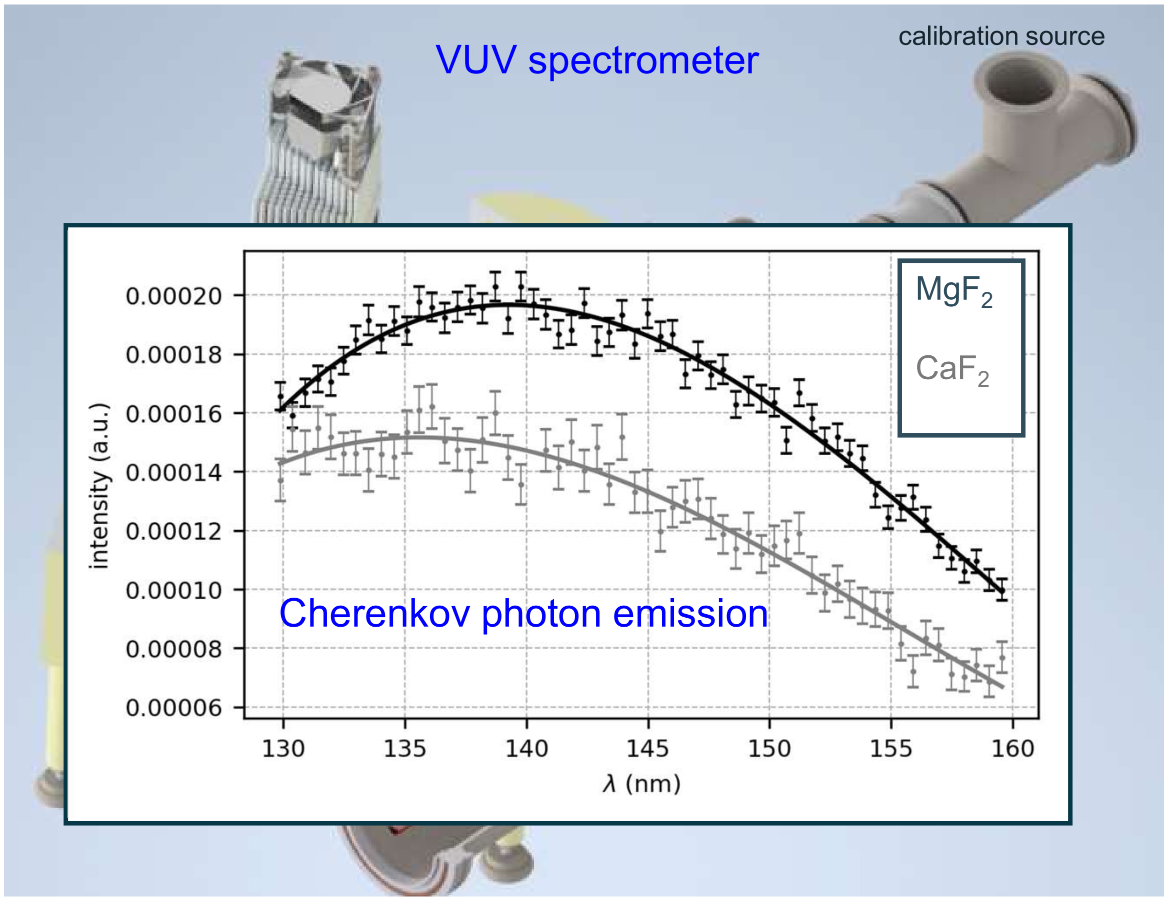
VUV spectroscopy at ISOLDE



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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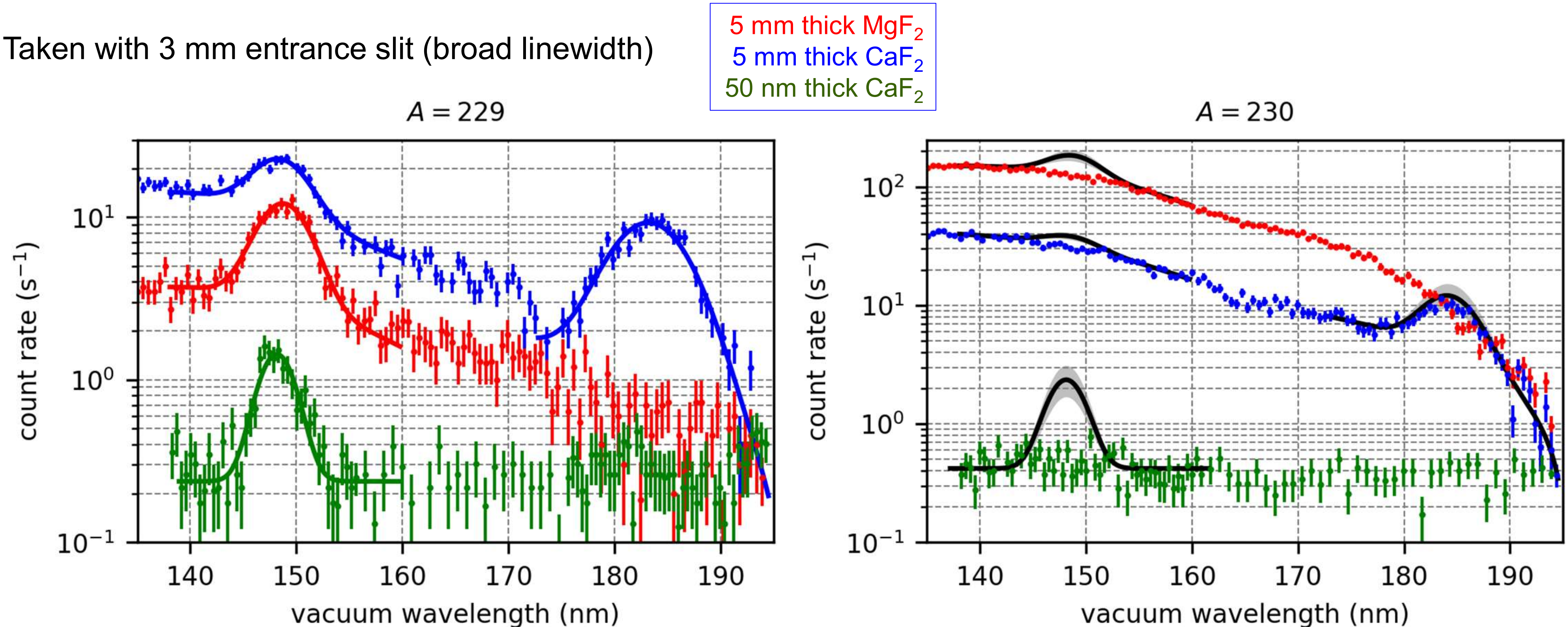
Identification

Taken with 3 mm entrance slit (broad linewidth)



^{229m}Th radiative decay

CaF_2 defect radioluminescence

CaF_2 defect radioluminescence

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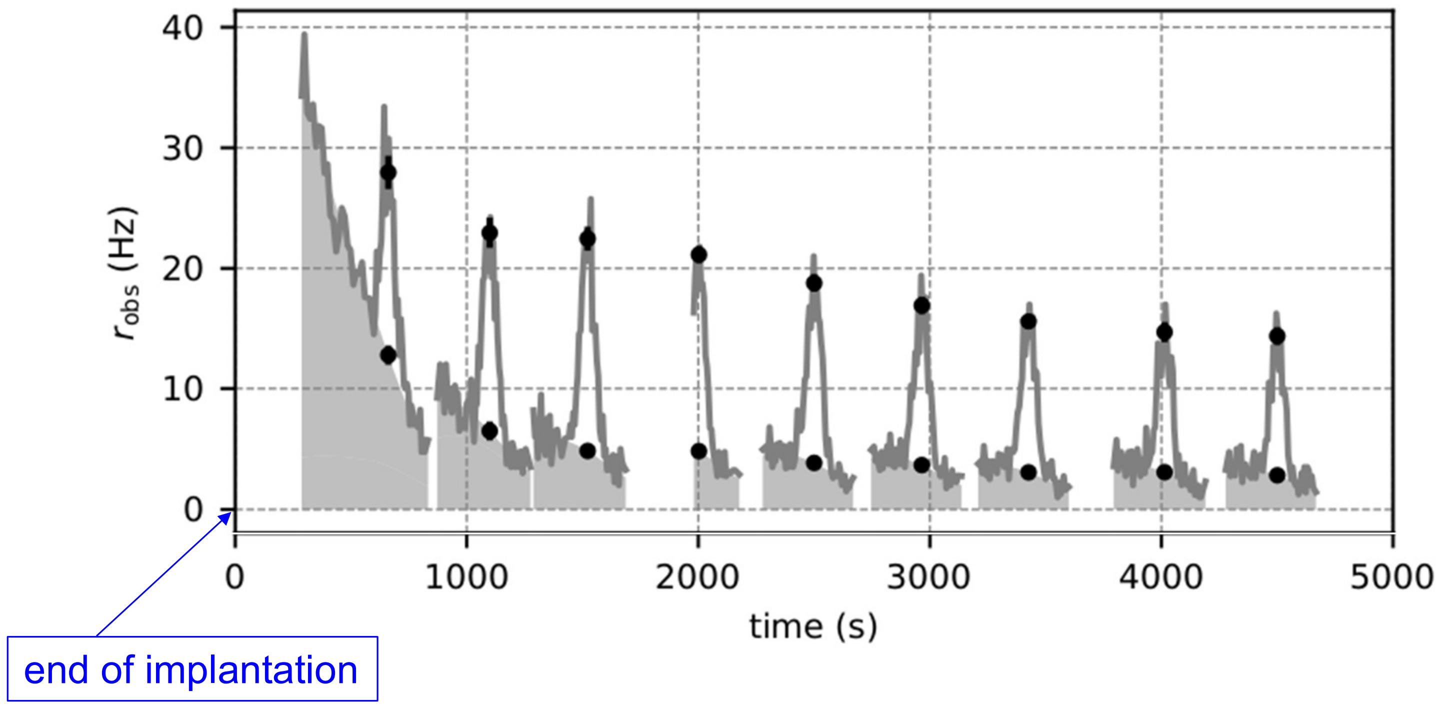
Time behaviour

A = 229 implantation for 3450 s in a MgF_2 crystal



(2 mm entrance slit (broad linewidth))

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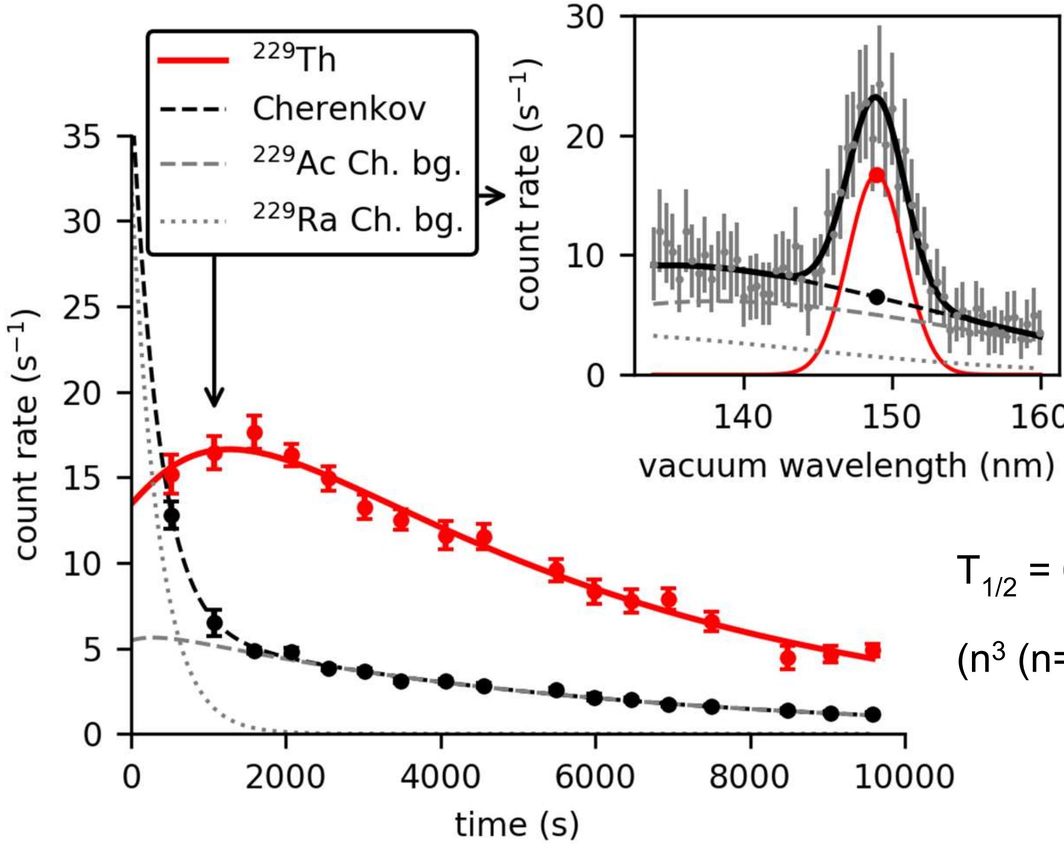
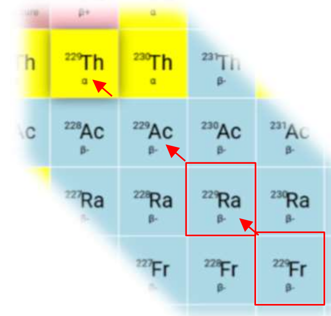
Time behaviour

^{229}Fr ($T_{1/2}$ = 50.2 s) → ^{229}Ra (4.0 m) → ^{229}Ac (62 m) → ^{229,m}Th





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

(n^3 (n=1,488) dependence: 2.21(32) 10^3 s)

Tkalya et al., PRC61 (2000)

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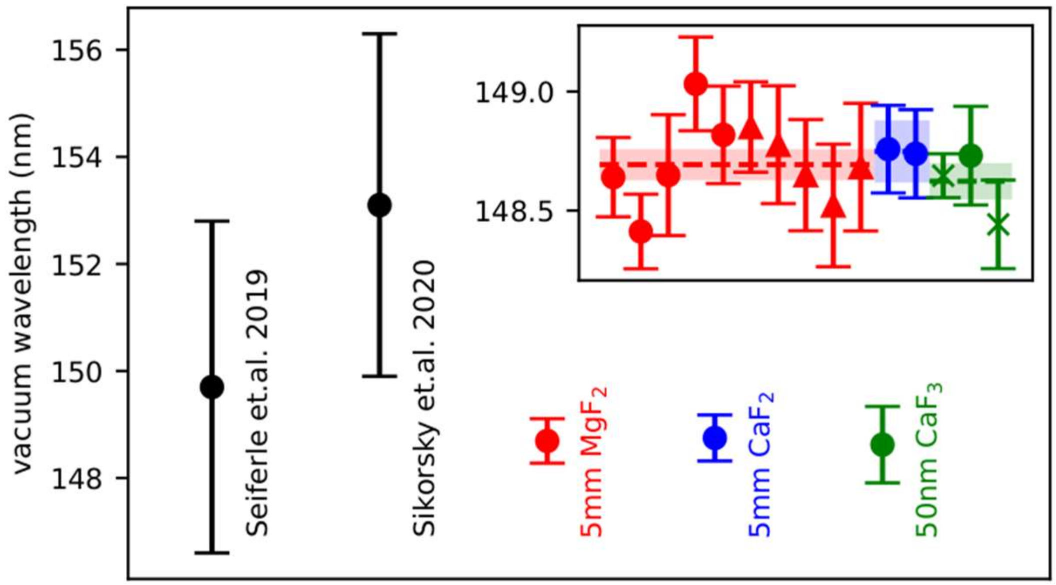
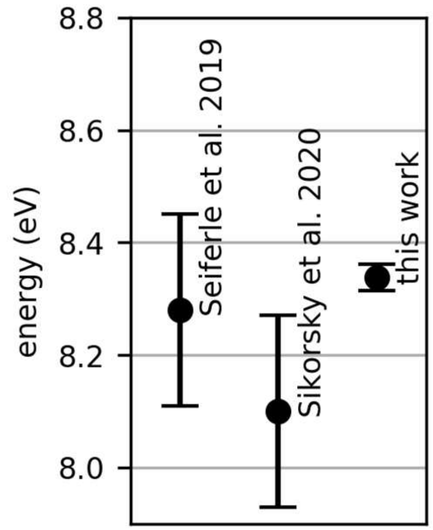
Energy

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





New energy value:
148.71 ± 0.06 (stat.) ± 0.41 (syst.) nm

- ➢ Uncertainty reduced from 41 THz to 5.8 THz

Article

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

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

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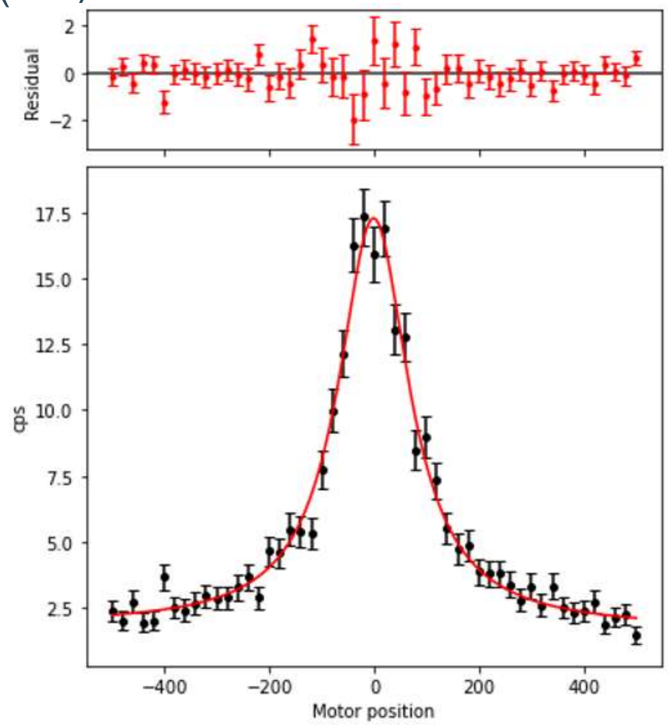
Preliminary results from IS-715 (July 2023)

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

CaF_2 (11.8 eV)	✓
MgF_2 (10.8 eV)	✓
$LiSrAlF_6$ (10.7 eV)	✓
SiO_2 (8.9 eV)	No
AlN (6.0 eV)	No



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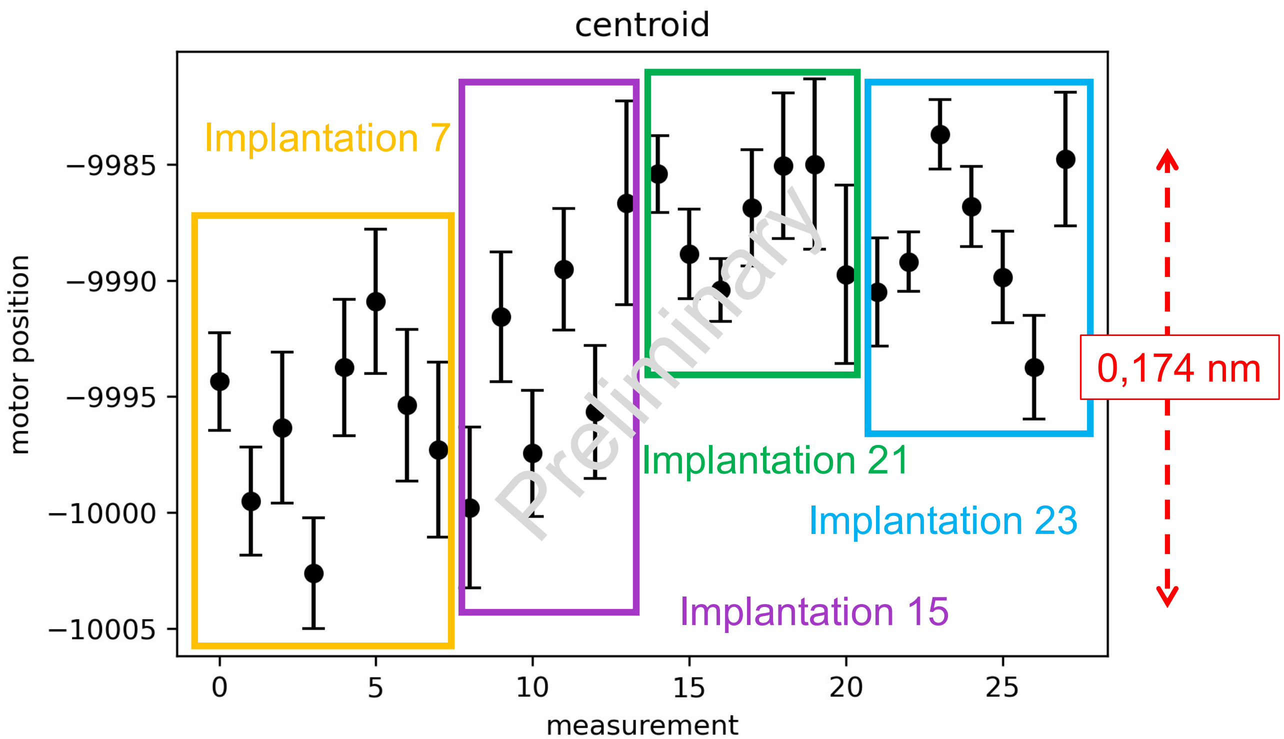

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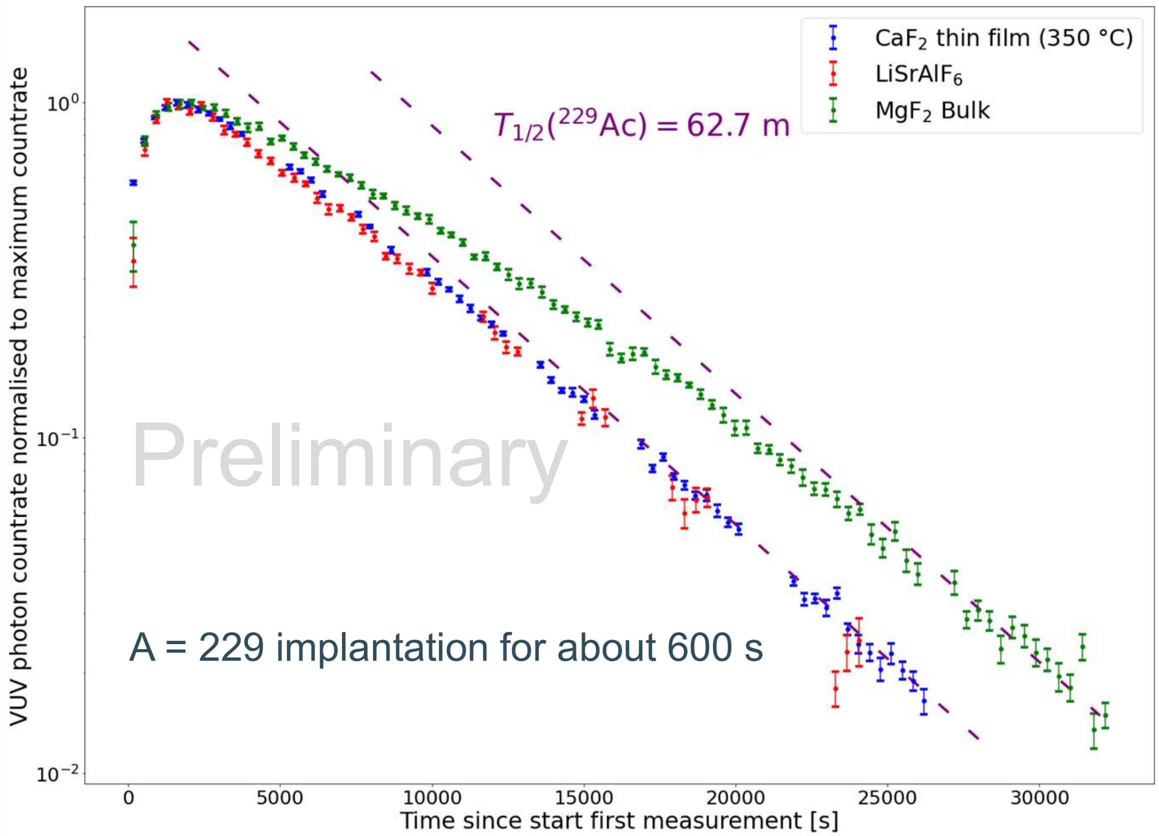
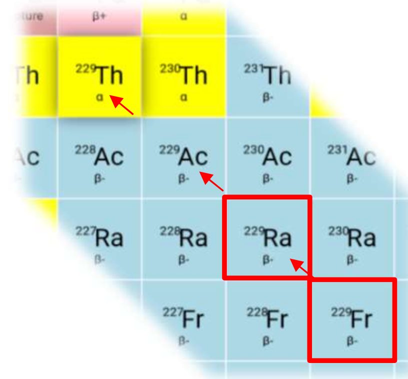
Preliminary results from IS-715 (July 2023)





- Different fractions at implantation sites with different half-life?
- Diffusion?
- ??

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Conclusion

Conclusion

- Populating the ^{229m}Th isomer via the beta decay of ^{229}Ac at ISOLDE
- Implantation technique: good performance for large band-gap, thick and thin-film crystals

  (solid-state based nuclear clock)
- Photon emission from a narrow linewidth emission found in large-band gap crystals
  - In MgF_2, CaF_2 and $LiSrAlF_6$ crystals
  - Signature of a mother-daughter decay with $T_{1/2}$ = 670(102) s (in MgF_2) and new energy value E = 8,388(24) eV

    → *First observation of the radiative decay of the ^{229m}Th isomer*

    → *Isomer detection in crystal matrix: consequences for a solid-state-based ^{229m}Th clock*
  - More precise energy value will become available (about 0,006/0,01 eV uncertainty)
  - Time behaviour of VUV signal influenced by diffusion, different implantation sites,…?

    → Annealing studies in different crystals
- Upgrade VUV spectrometer – position sensitive MCP

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Thank you very much!

ISOLDE: IS658 – IS715

M. Athanasakis, M. Au, S. Bara, K. Beeks, P. Chhetri, A. Claessens, T. Cocolios, J.G. Correia, S.Cottenier, H. De Witte, Y. Elskens, R. Ferrer, S. Geldhof, N. Hosseini, S. Kraemer, U Köster, M. Laatiaoui, R. Lica, G. Magchiels, V. Manea, J. Moens, I. Moore, S. Pineda, L. M. Pereira, 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