KU LEUVEN

LMU LUDWIG-MAXIMILIANS-UNIVERSITÄT MÜNCHEN

Observation of the radiative decay of the low-energy thorium-229 isomer: *En route* towards a nuclear clock

Sandro Kraemer
on behalf of the ISOLDE-IS658 collaboration

Outline:

- The nuclear clock proposal
- Vacuum-ultraviolet spectroscopy at ISOLDE
- Results from the 2021 measurement campaign
- Conclusion and outlook

CGS-17
Grenoble, 20.07.23

The nuclear clock proposal

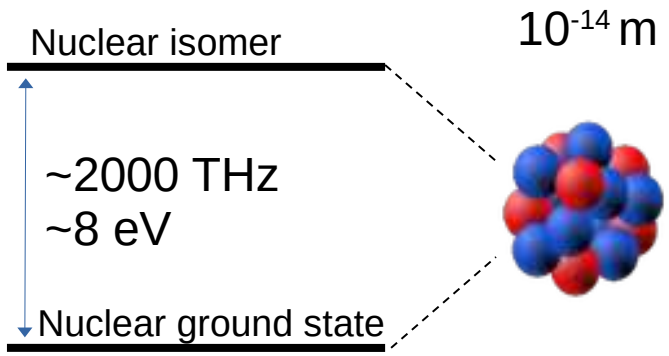
Metrology of time

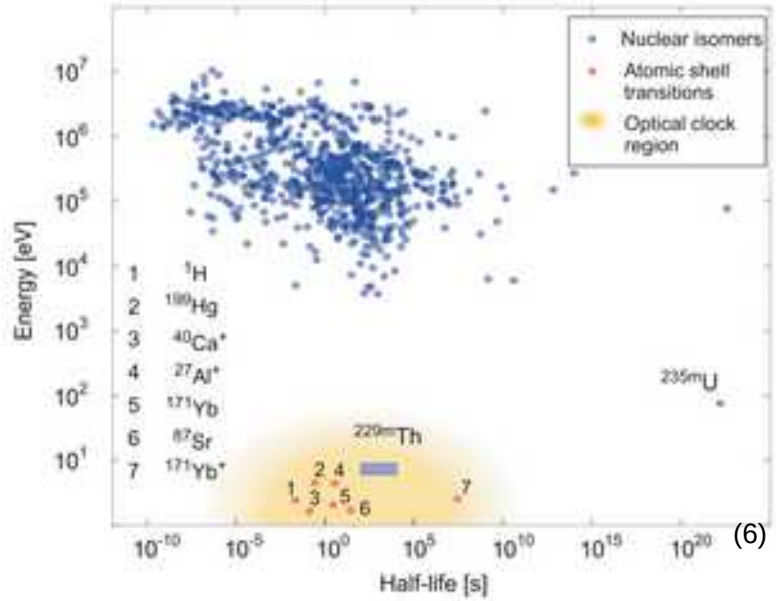
Chateau de Sassenage, uknown manufacturer, probably Paris, 17th century

Metrology of time

SI Definiton of time:
1 second = 9.192.631.770 oscillations of the ground state hyperfine splitting in Cs-133

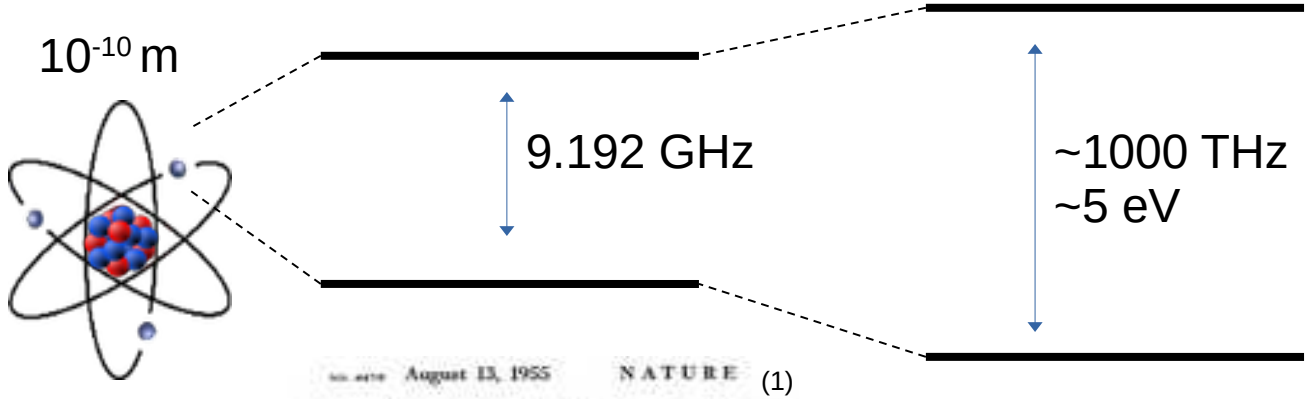
Peik and Tamm 2003:
Optical "nuclear" clock[5]

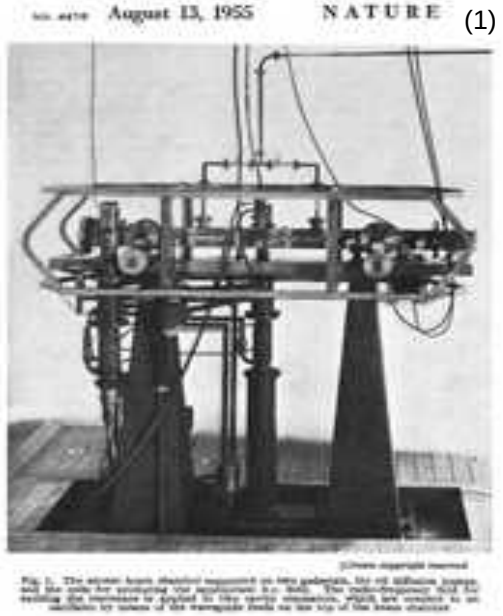


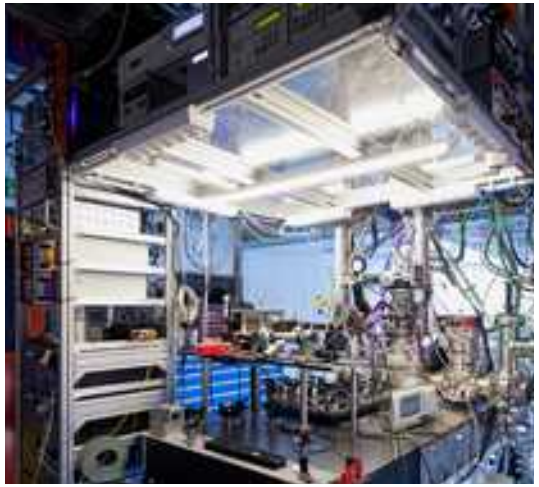


(6)

Hyperfine splitting clocks *Optical clocks*




August 13, 1955 NATURE
(1)



(4): Essen and Parry, *Nature* **4476**
(5) Peik and Tamm,2033, *EPL* **61** 2
(6) von der Wense et al., *Nature* **533** 7601
(7) Campbell et al., 2012, *PRL* **108** 120802

(1): Essen and Parry, *Nature* **4476**

Metrology of time

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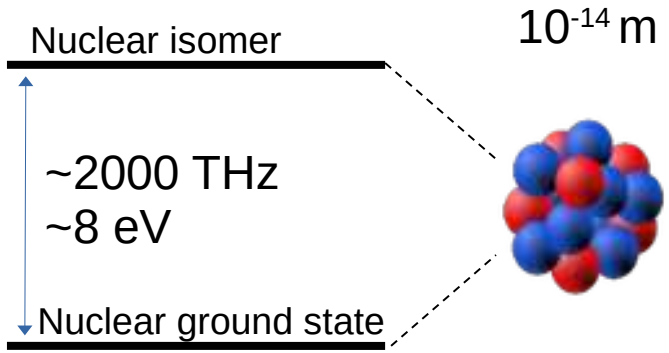
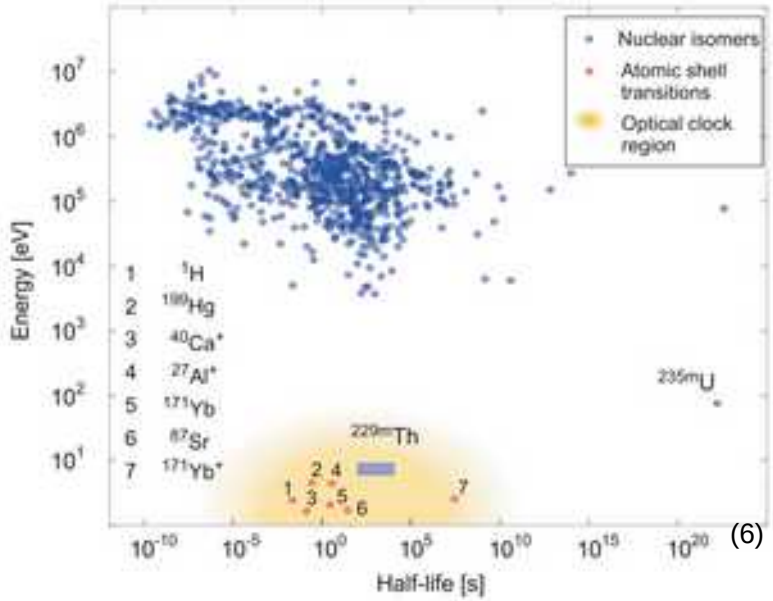


10^{-14} m



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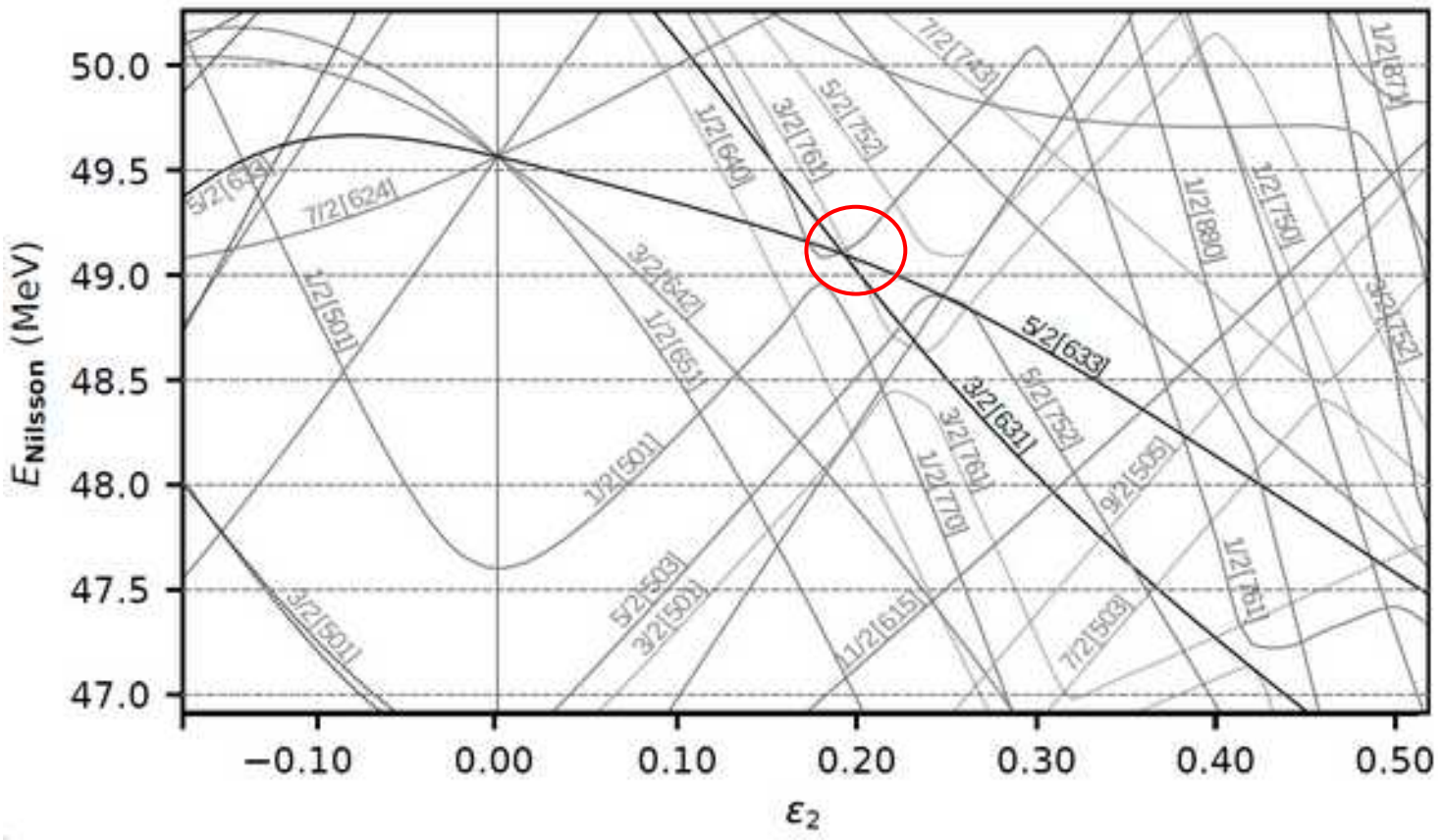

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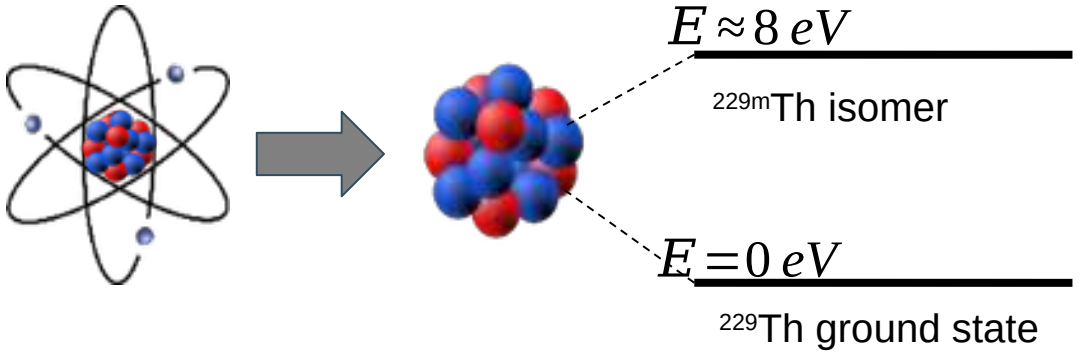
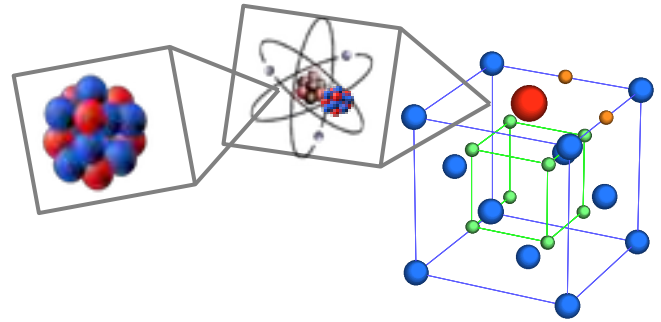
(5) Peik and Tamm,2033, *EPL* **61** 2
(6) von der Wense et al., *Nature* **533** 7601
(7) Campbell et al., 2012, *PRL* **108** 120802

(1): Essen and Parry, *Nature* **4476**

Applications(10)





Large number of nuclei in crystalline medium

→ **Ultrastable clock (-networks)**

Satellite-based navigation

Gravimetry

Sensitive to nuclear forces

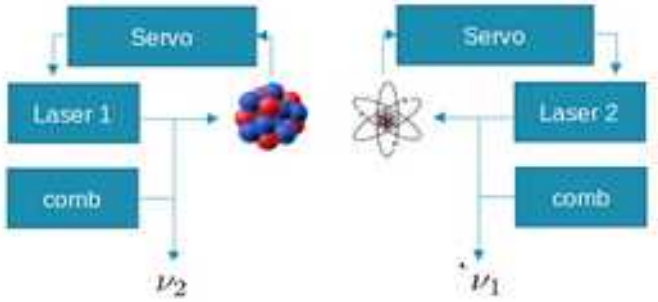
→ **Clock comparisons (-networks)**

oscillatory ultralight dark matter

→ time variations of fundamental constants

→ domain walls of topological dark matter

$$\frac{\partial}{\partial t}\ln\frac{\nu_2}{\nu_1} = (K_2 - K_1)\frac{1}{\alpha}\frac{\partial \alpha}{\partial t}$$



Topological dark matter:

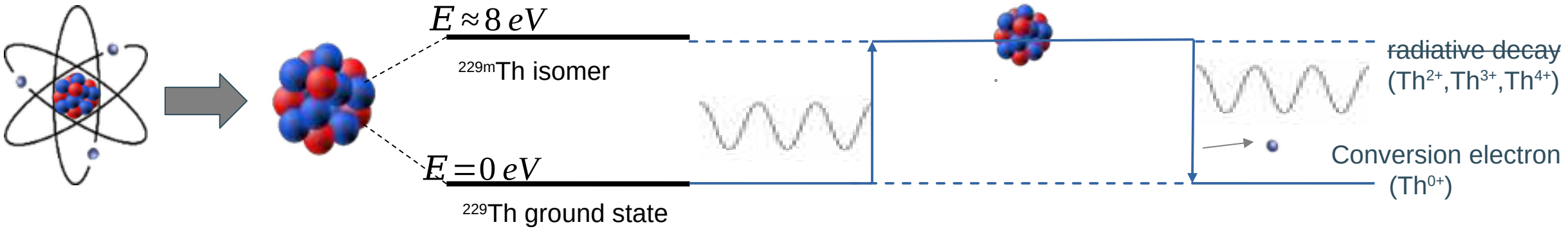
(11)

Network of synchronized clocks

(10) for a review: Thirolf et al., 2019 *Journ. Phys. B: At. Mol. Opt. Phys.* **52** 203001 and references therein

(11) Adhikari et al., 2014 *Nature* **10** 906

The nuclear clock proposal



Single ion NC[8]

LMU, PTB

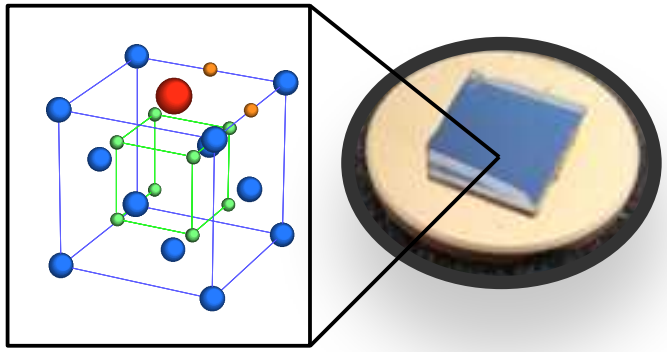
Hyperfine structure nuclear state detection

1 to 10 ions

→ Highest accuracy

Solid-state NC[8]

TU Vienna, UCL



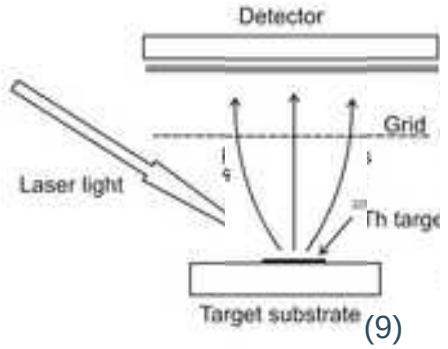
Radiative decay detection

>10^{15} nuclei

→ Highest stability

Conversion el. NC[9]

NIST

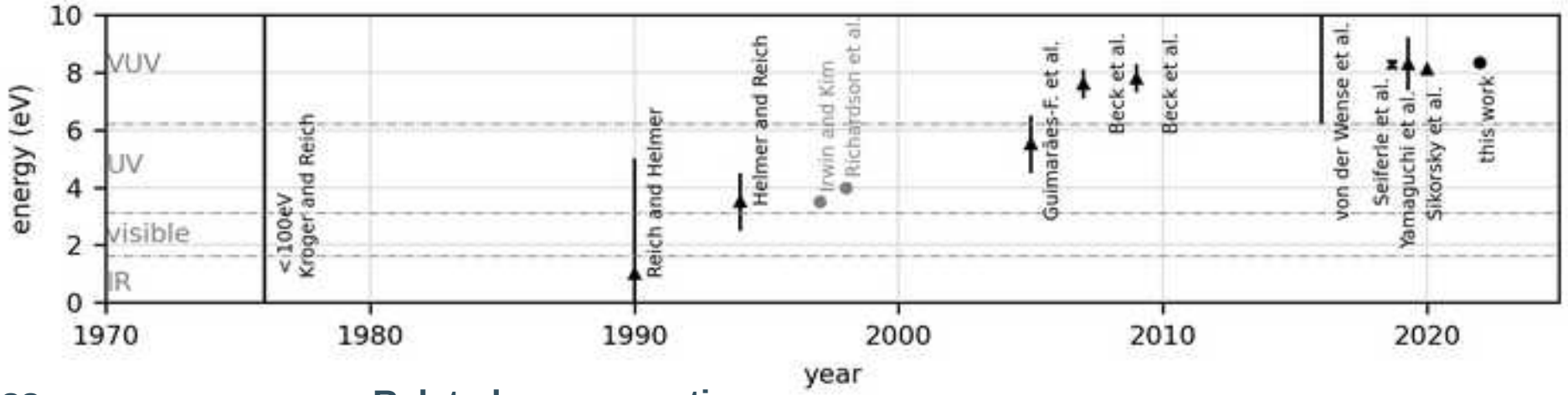


(9)

(8) Peik et al., 2021, *Quantum. Sci. Technol.* **6** 034003
(9) von der Wense et al., 2019, *Hyperf. Interact.* **240** 2

Thorium-229 and its isomer

Kröger and Reich 1976: first evidence for an isomer in ^{229}Th



Recent progress

- 2016: Proof of existence[12]
- 2017: Lifetime (IC decay) [13]
- 2018: Nuclear moments[14]
- 2019 and 2020: Energy [15,16]

Related open questions

→ *Why has the radiative decay not been observed?*

→ *Laser excitation cross section*

→ *Solid state clock performance*

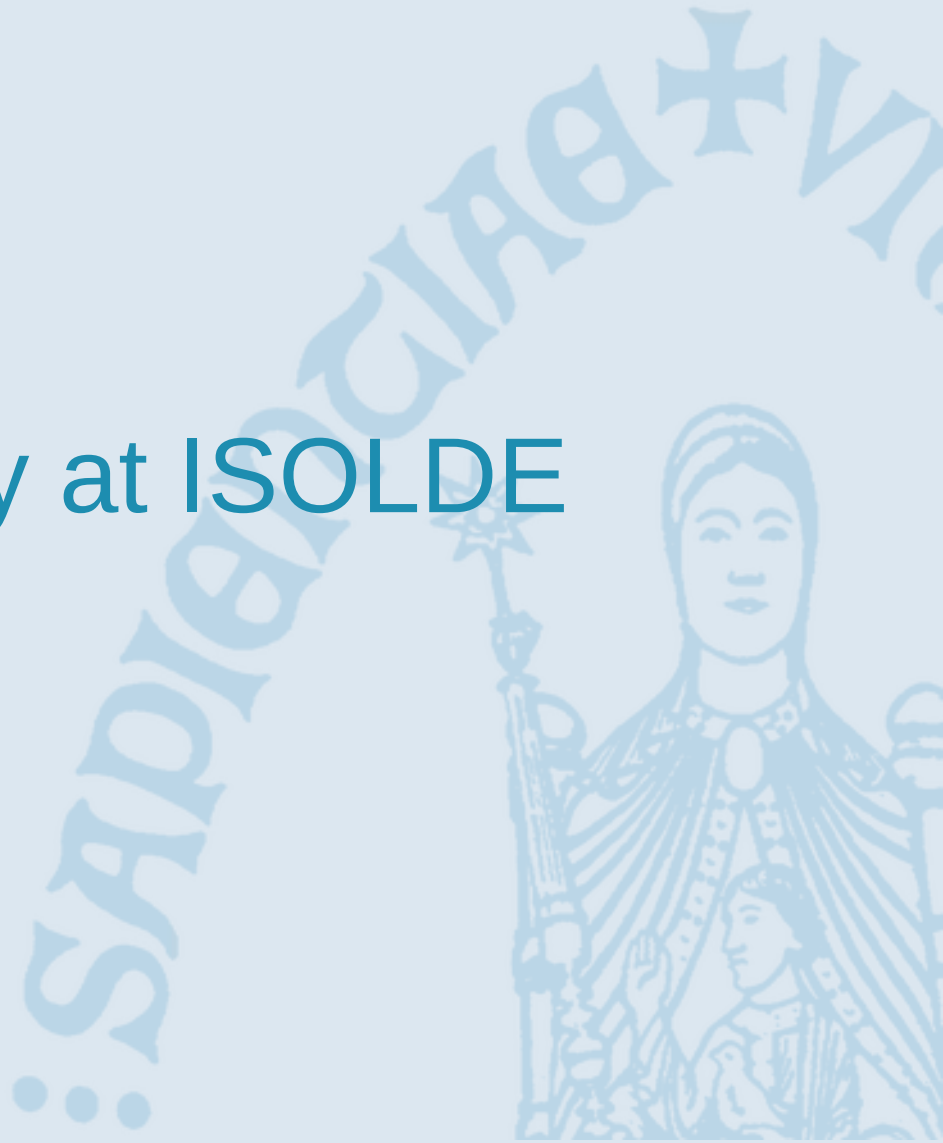
→ *Enhancement factor for fine structure constants*

→ *Needed for Vacuum-ultraviolet (VUV) laser development*
Uncertainty: 41THz

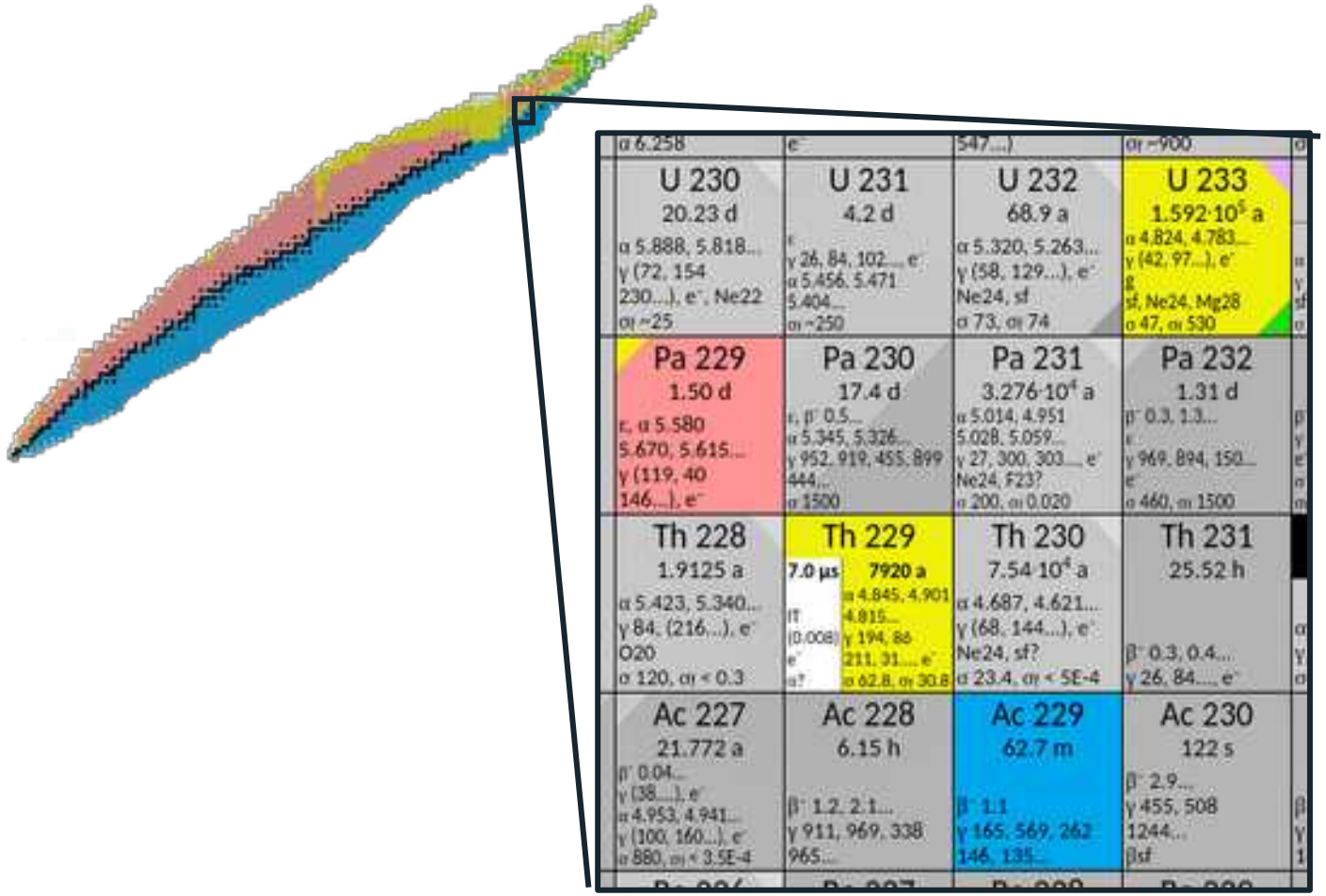
Population in radioactive decay + VUV spectroscopy as a bridge technology awaiting laser excitation

(12) von der Wense et al, 2016 *Nature* **533** 47-51
(13) Seiferle et al., 2017 *PRL* **118** 042501
(14) Thielking et al., 2018 *Nature* **556** 321-325
(15) Seiferle et al., 2019 *Nature* **573** 243-246
(16) Sikorsky et al., 2020 *PRL* **125** 142503

Vacuum ultraviolet spectroscopy at ISOLDE



Population in radioactive decay



(14) Verlinde, Kraemer, Moens *et al.*, 2019 *PRC* **100** 024315

Population in radioactive decay

standard approach:

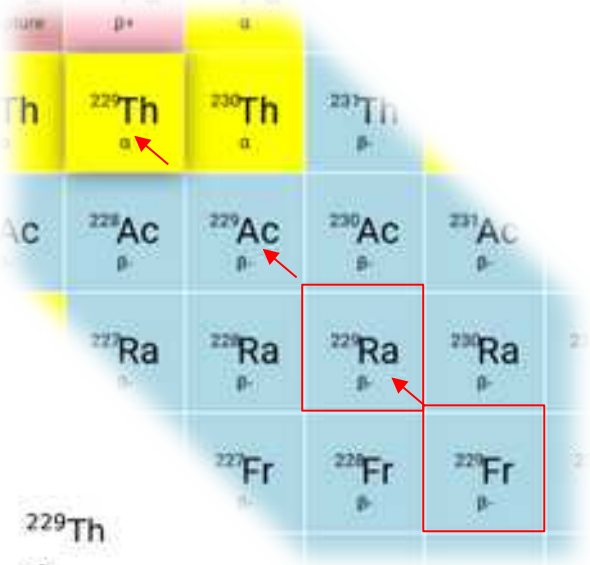
$^{233}U \rightarrow {}^{229}Th + \alpha$

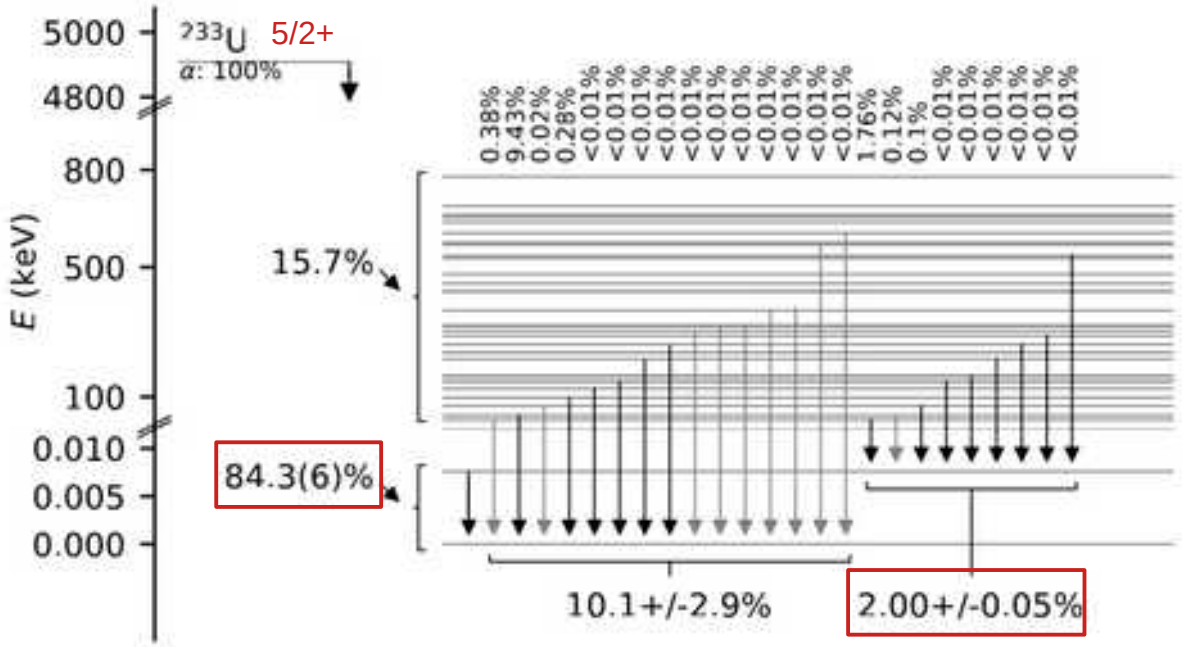
recoil: 84keV

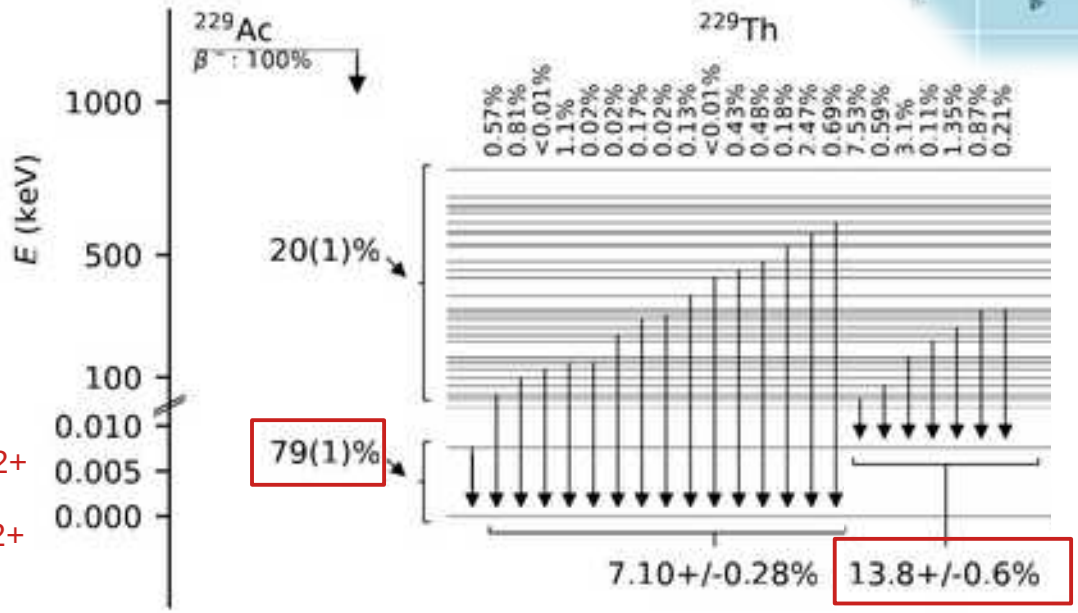
ISOLDE approach

Online production of ^{229}Ra and ^{229}Fr

Recoil: < 6 eV



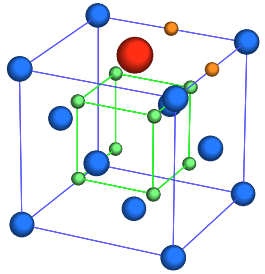




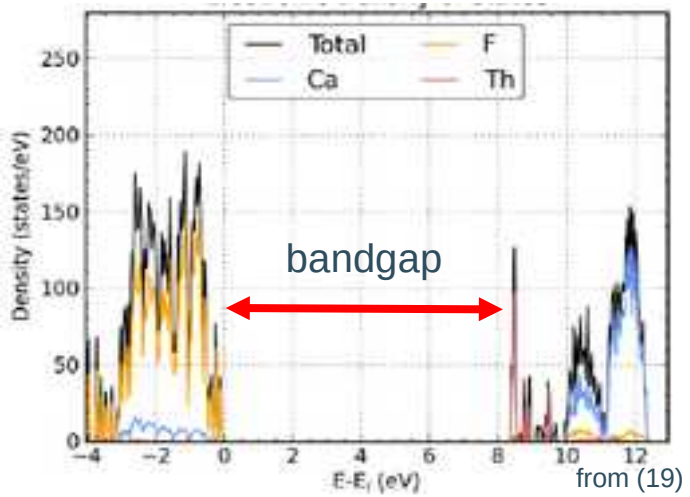
(14) Verlinde, Kraemer, Moens *et al.*, 2019 *PRC* **100** 024315

Solid state approach

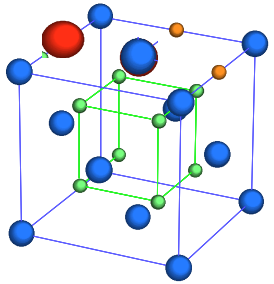
Embedding in large-badgap crystals (MgF_2, CaF_2,..) to achieve high charge state (Th^{3+}, Th^{4+})(15)



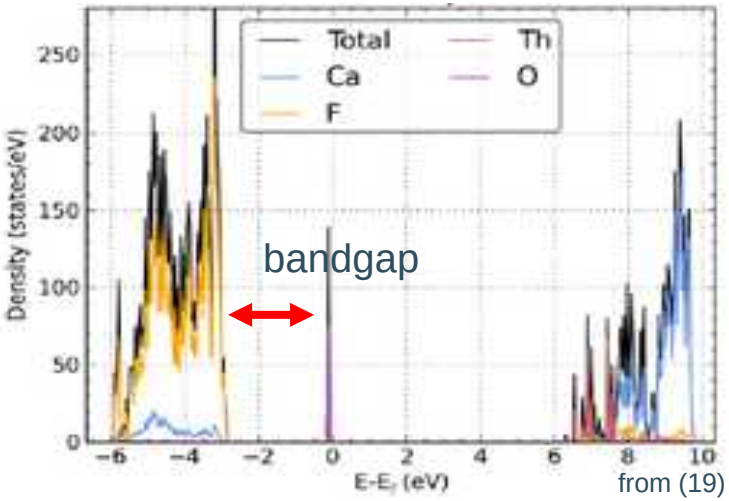
CaF_2: $^{229}Th^{4+}$ in Ca-substitutional position(16)



No electrons available for e⁻ conversion decay
→ radiative decay

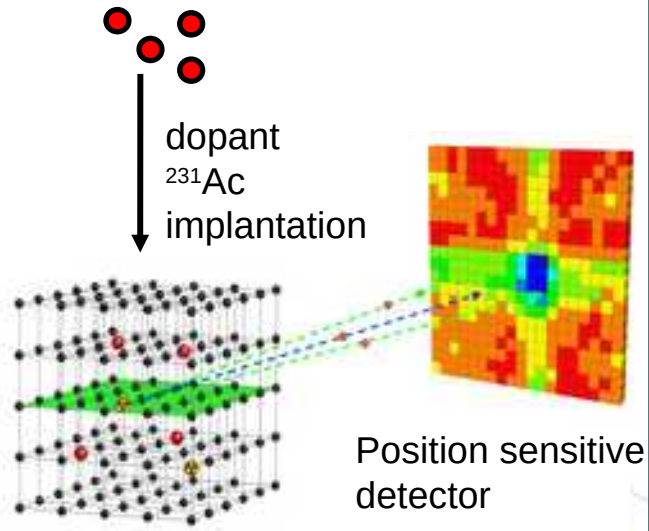


CaF_2: $^{229}Th^{4+}$ in interstitial position(16)



Unpopulated states can be populated
→ electron conversion decay

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

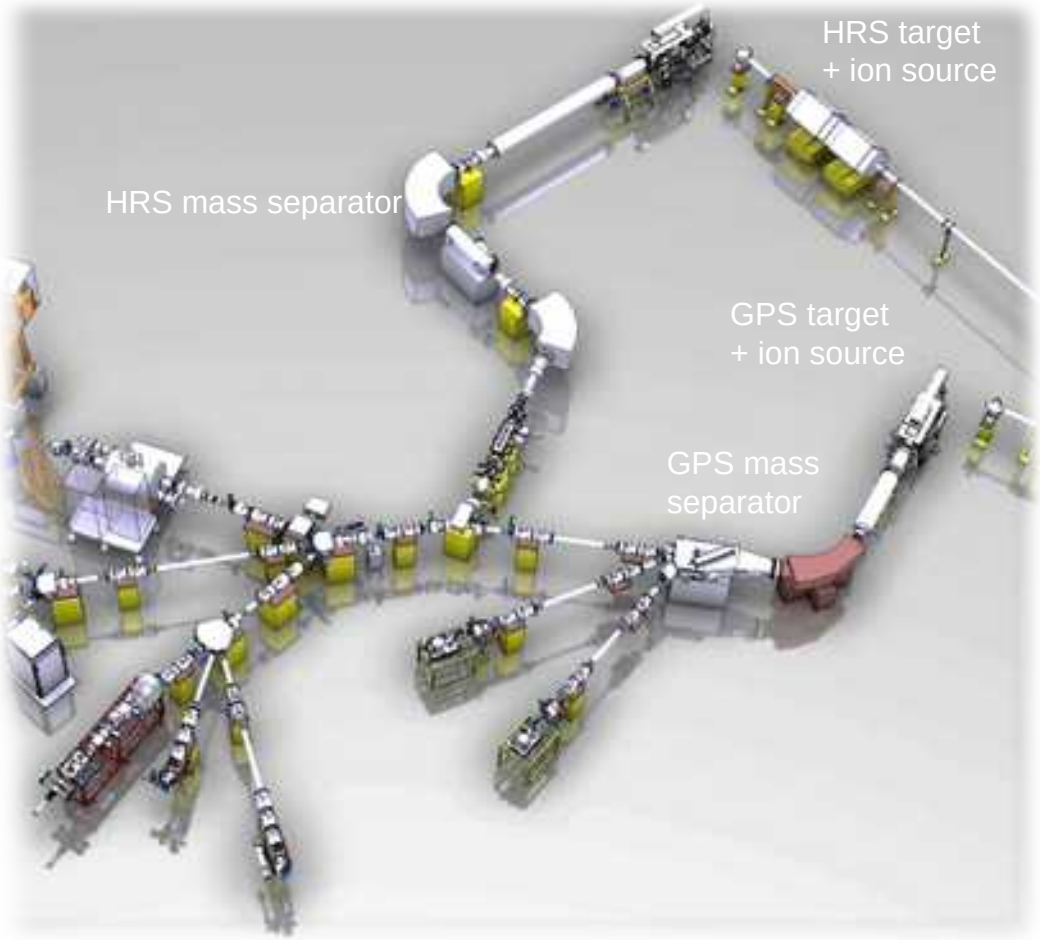


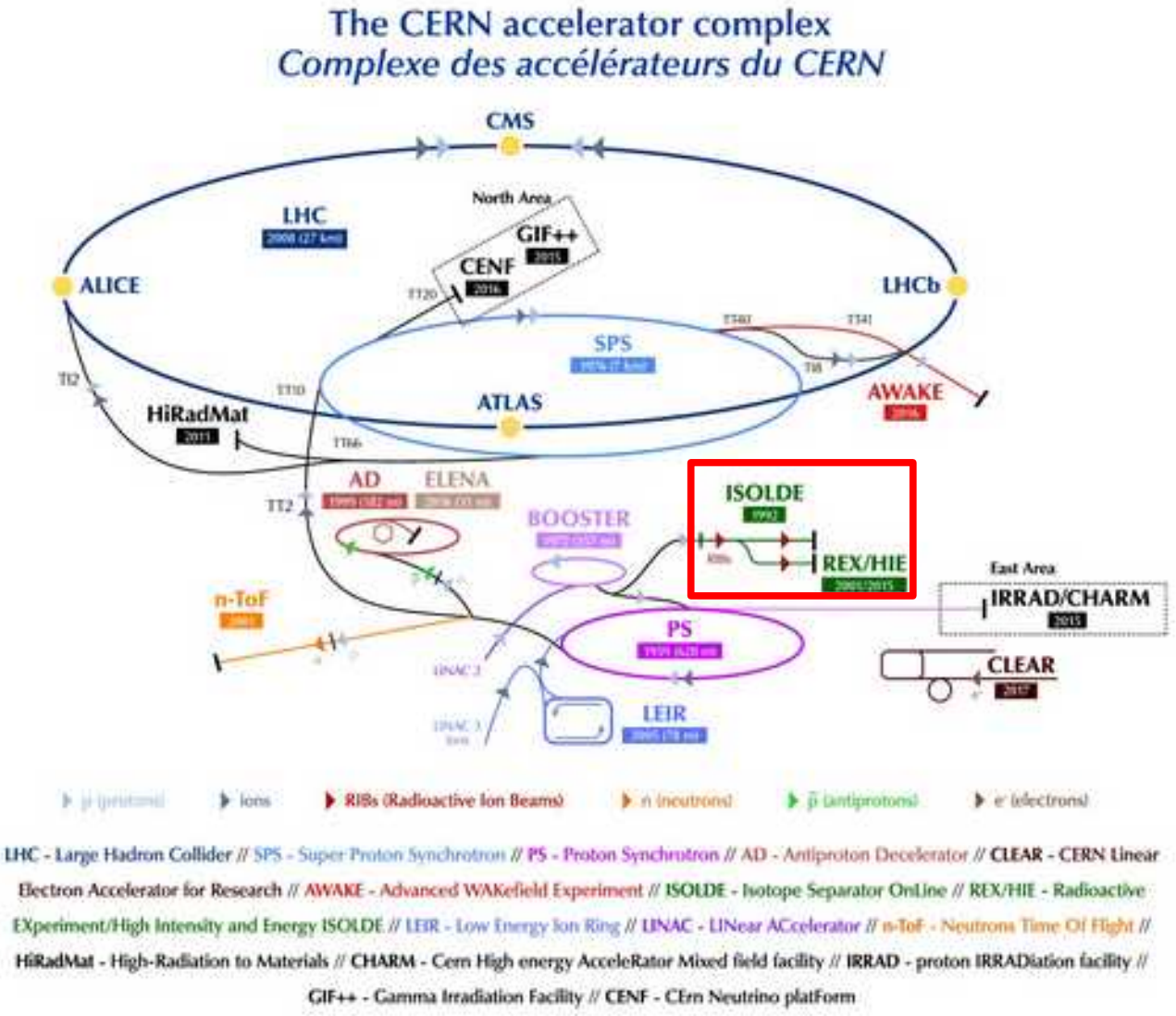
Host crystal

→ 77% to 100%
in substitutional position

(15) G A Kazakov et al., 2012 *New J. Phys*. **14** 083019
(16) Dessovic. et al., 2014 *J. Phys. Condens. Matter* **26** 10

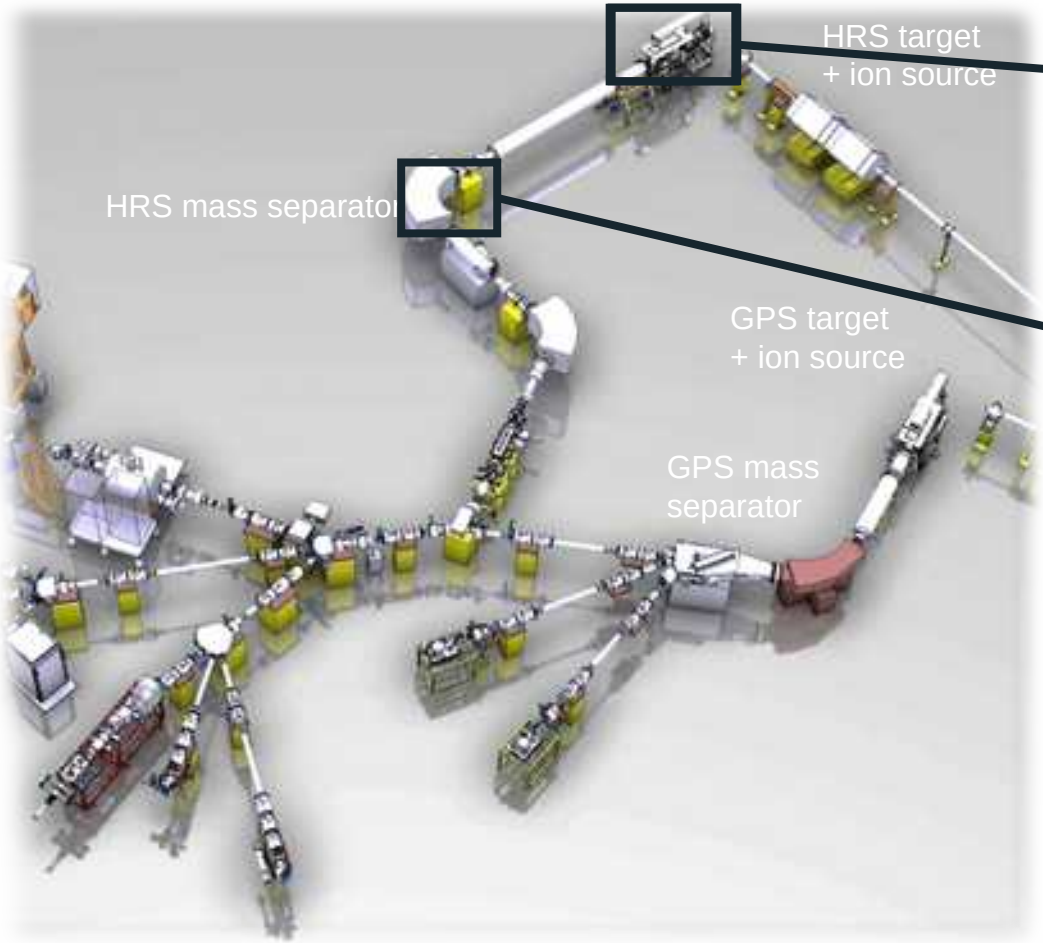
VUV spectroscopy at ISOLDE





ISOLDE beamline and accelerator complex sketches: CERN

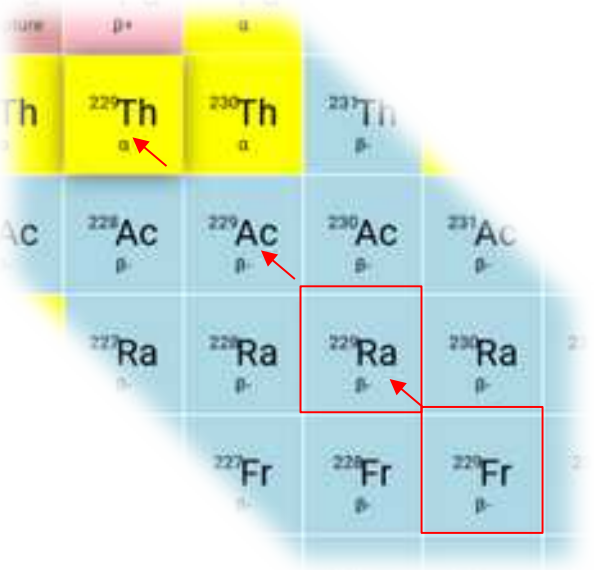
VUV spectroscopy at ISOLDE



UCx ISOL target *(meanwhile ThCx available)*

- Surface ionization + RILIS on Ac
- Acceleration to 30keV

- A=229 beams of ~10^6 pps
- Composition: 90% ^{229}Ra (4 min) , 10% ^{229}Fr (38 s)



ISOLDE beamline and accelerator complex sketches: CERN

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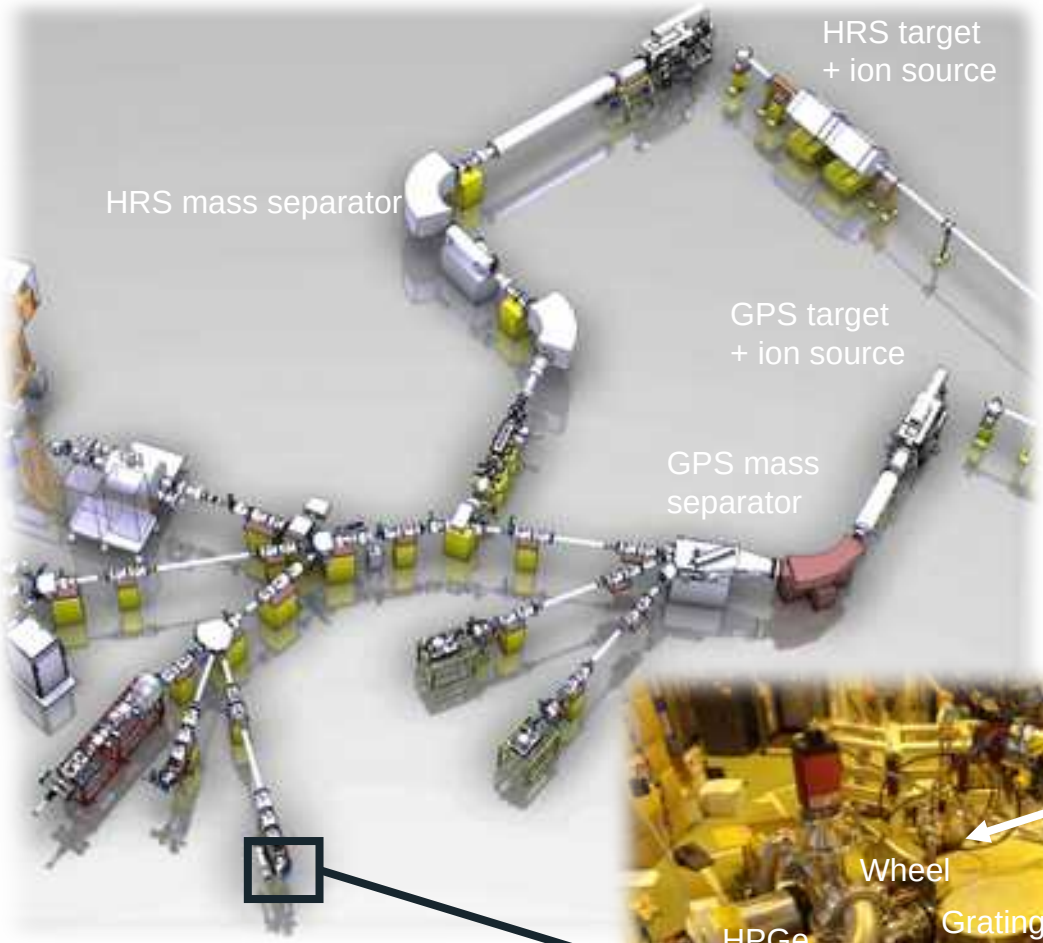
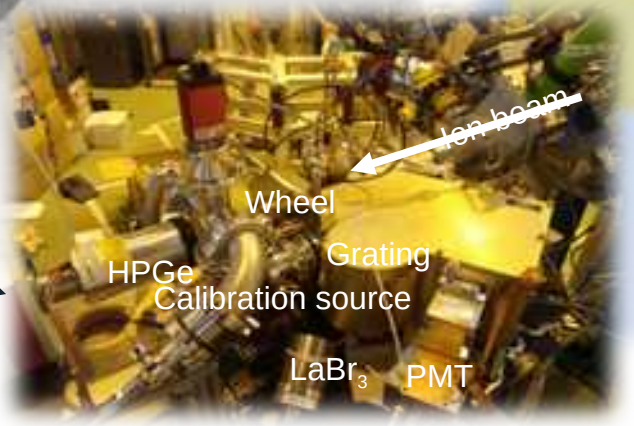
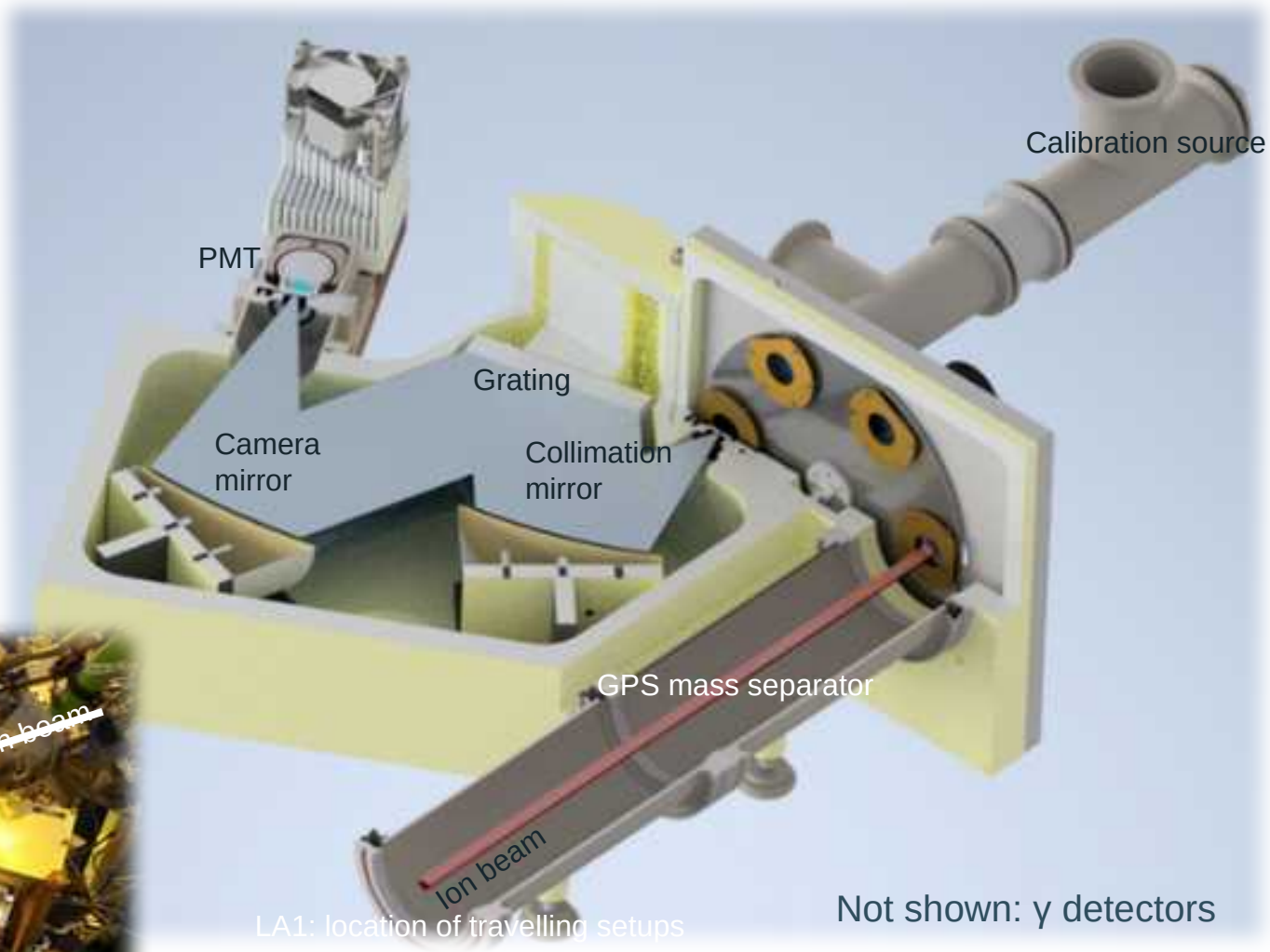




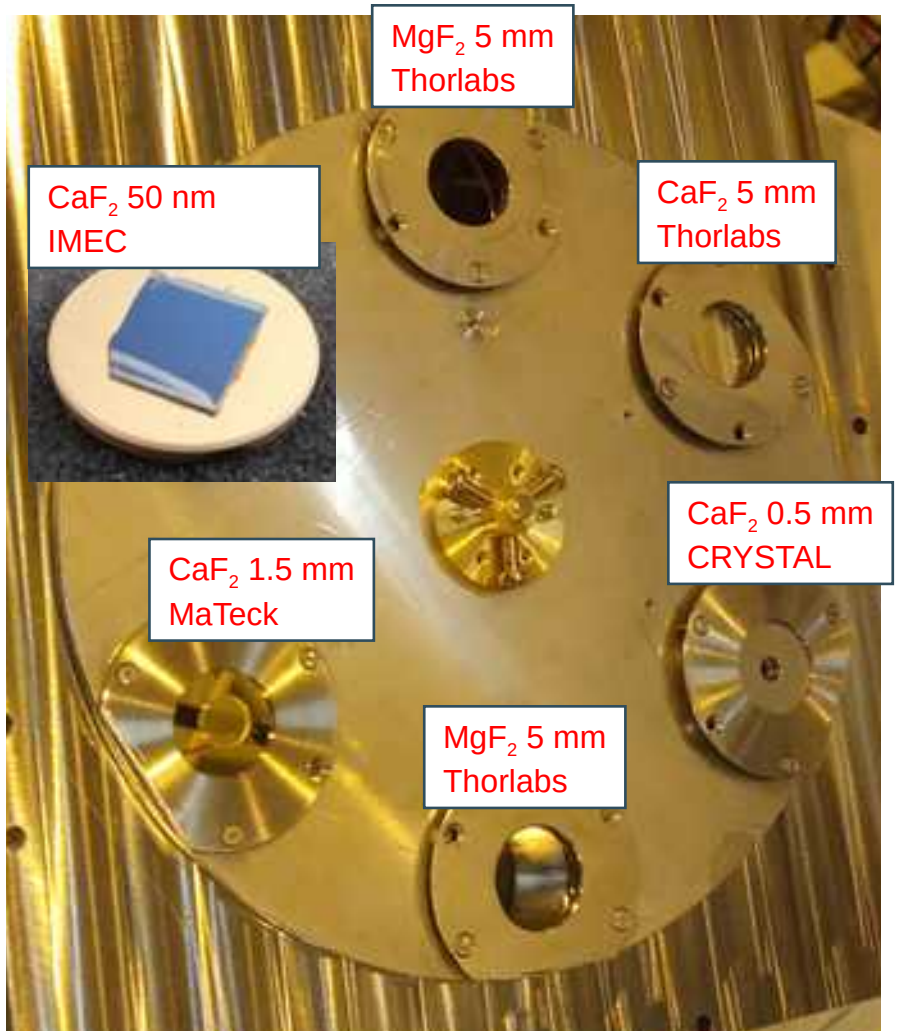


LA1: location of travelling setups

Not shown: γ detectors

ISOLDE beamline and accelerator complex sketches: CERN

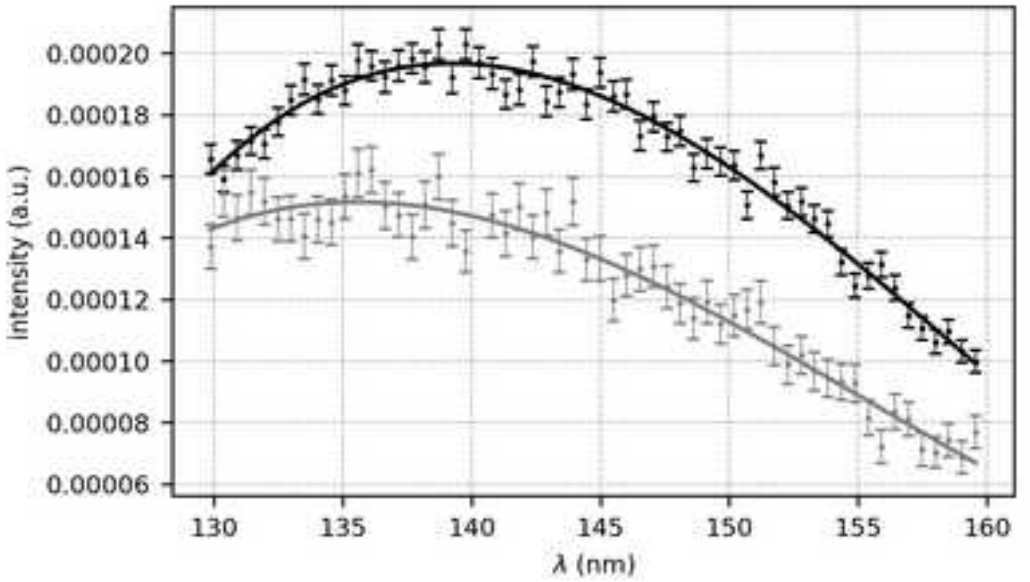
Crystals



Background induced by radiation:

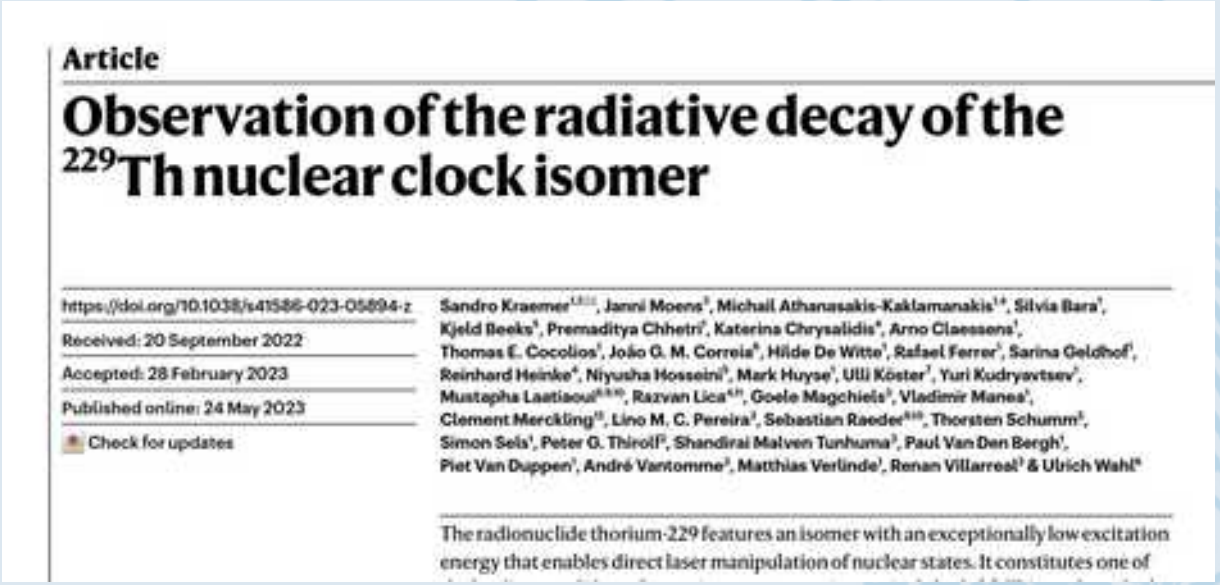
Cherenkov photon emission
→ *Continuous spectrum*

Observed spectral shape:
Cherenkov spectrum + instruments and crystal bandpass



Radioluminescence of crystal defects
→ *Limited spectral width*

Results from the 2021 measurement campaign (IS 658)



Article

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

https://doi.org/10.1038/s41586-023-05894-z
Received: 20 September 2022
Accepted: 28 February 2023
Published online: 24 May 2023
Check for updates

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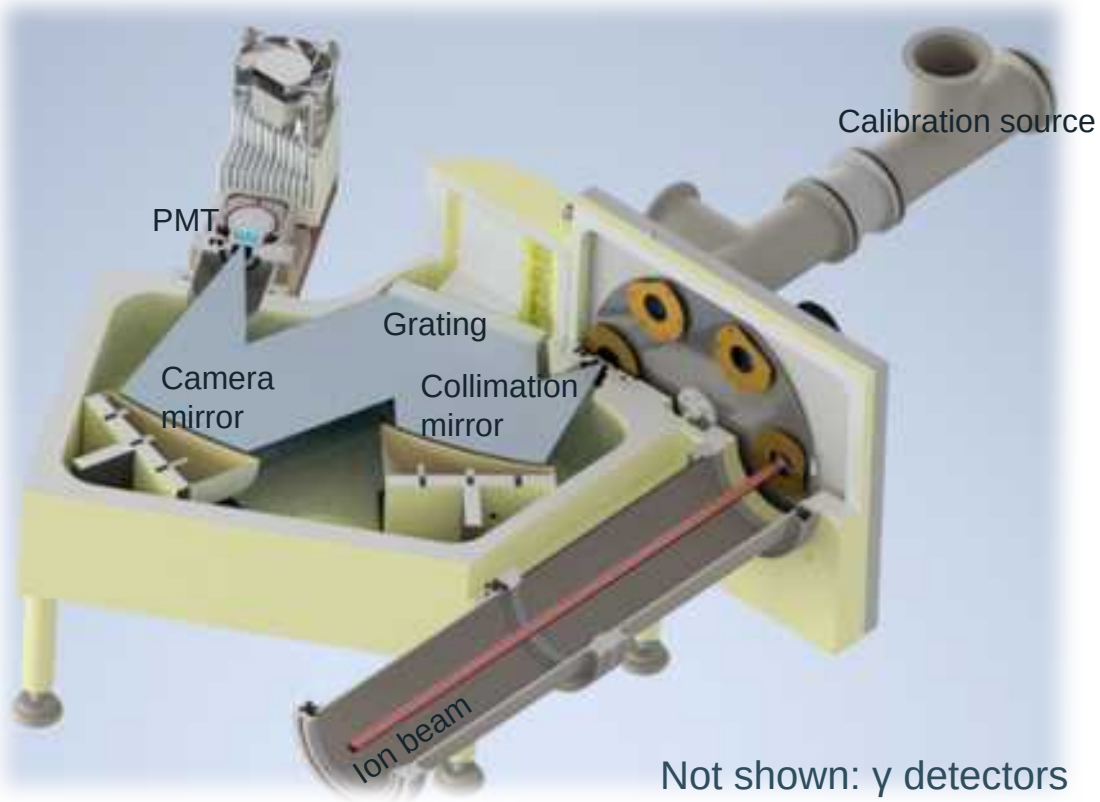
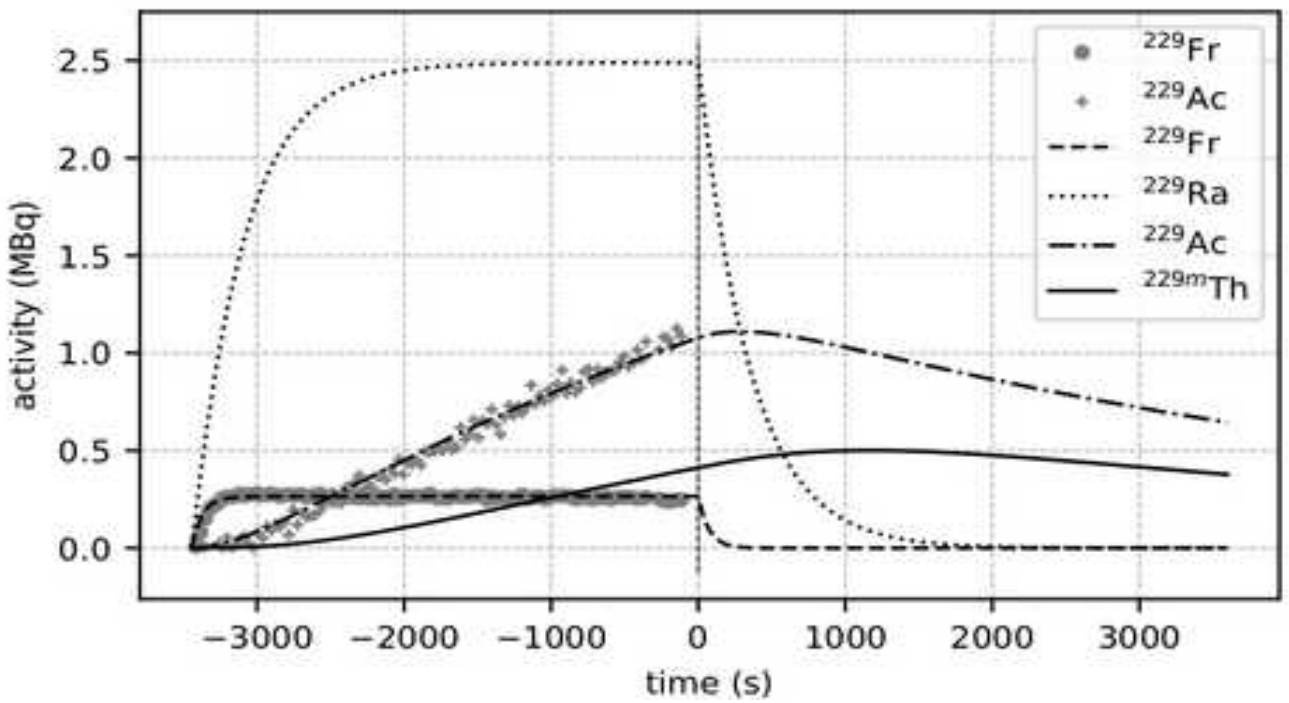
The radionuclide thorium-229 features an isomer with an exceptionally low excitation energy that enables direct laser manipulation of nuclear states. It constitutes one of

S. Kraemer, J. Moens, et.al. (2023) *Nature* 617 707-710

Implantation



Not shown: γ detectors



Length: 0.5 to 2 h
Implantation depth: ~ 20 nm

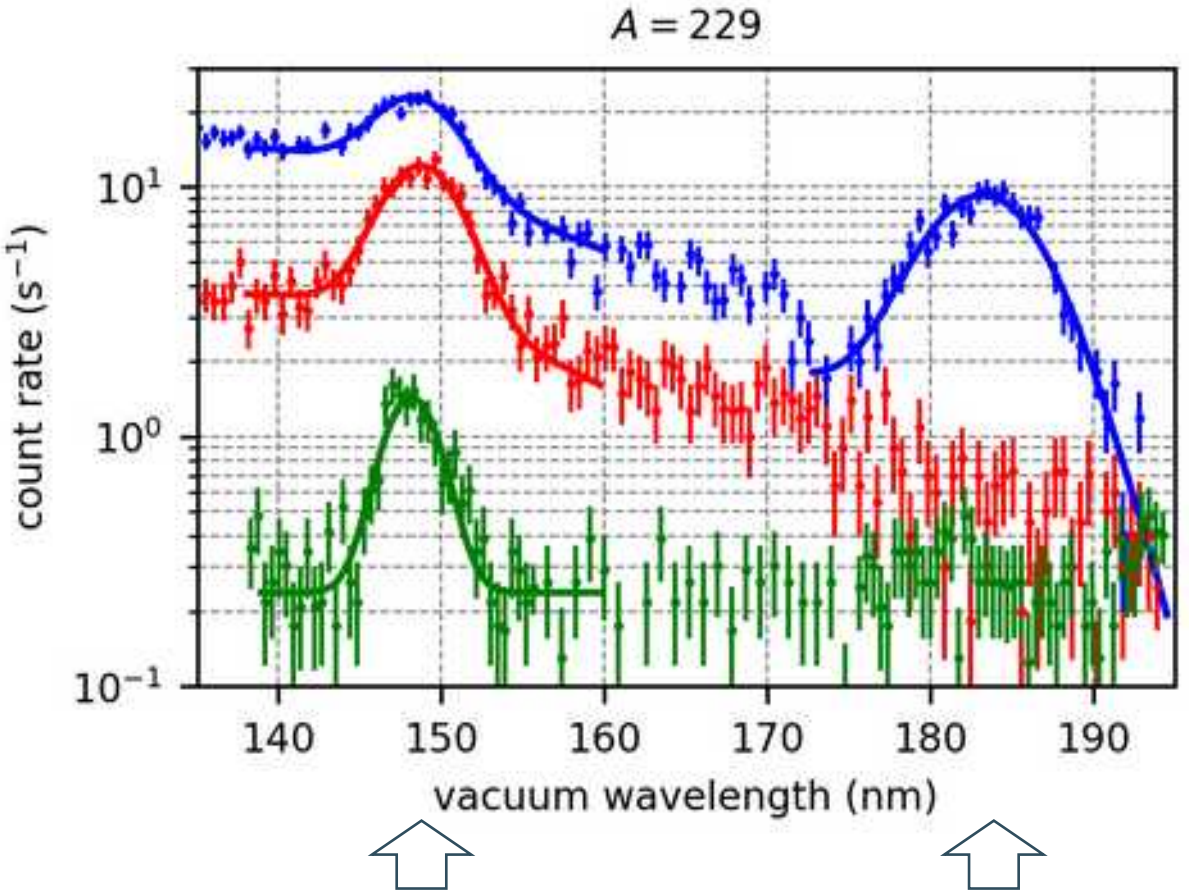
Identification

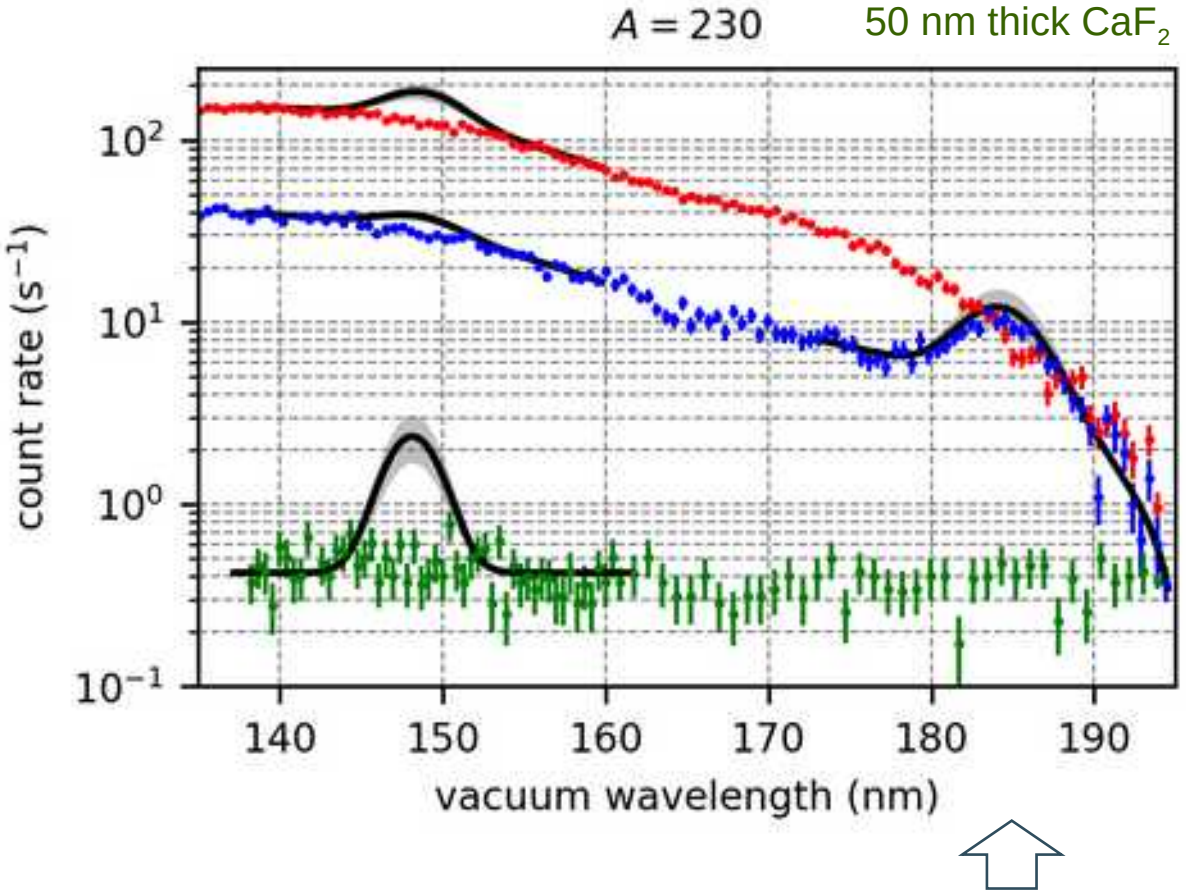
Taken with 3 mm entrance slit (broad linewidth)

5 mm thick MgF_2

5 mm thick CaF_2

50 nm thick CaF_2



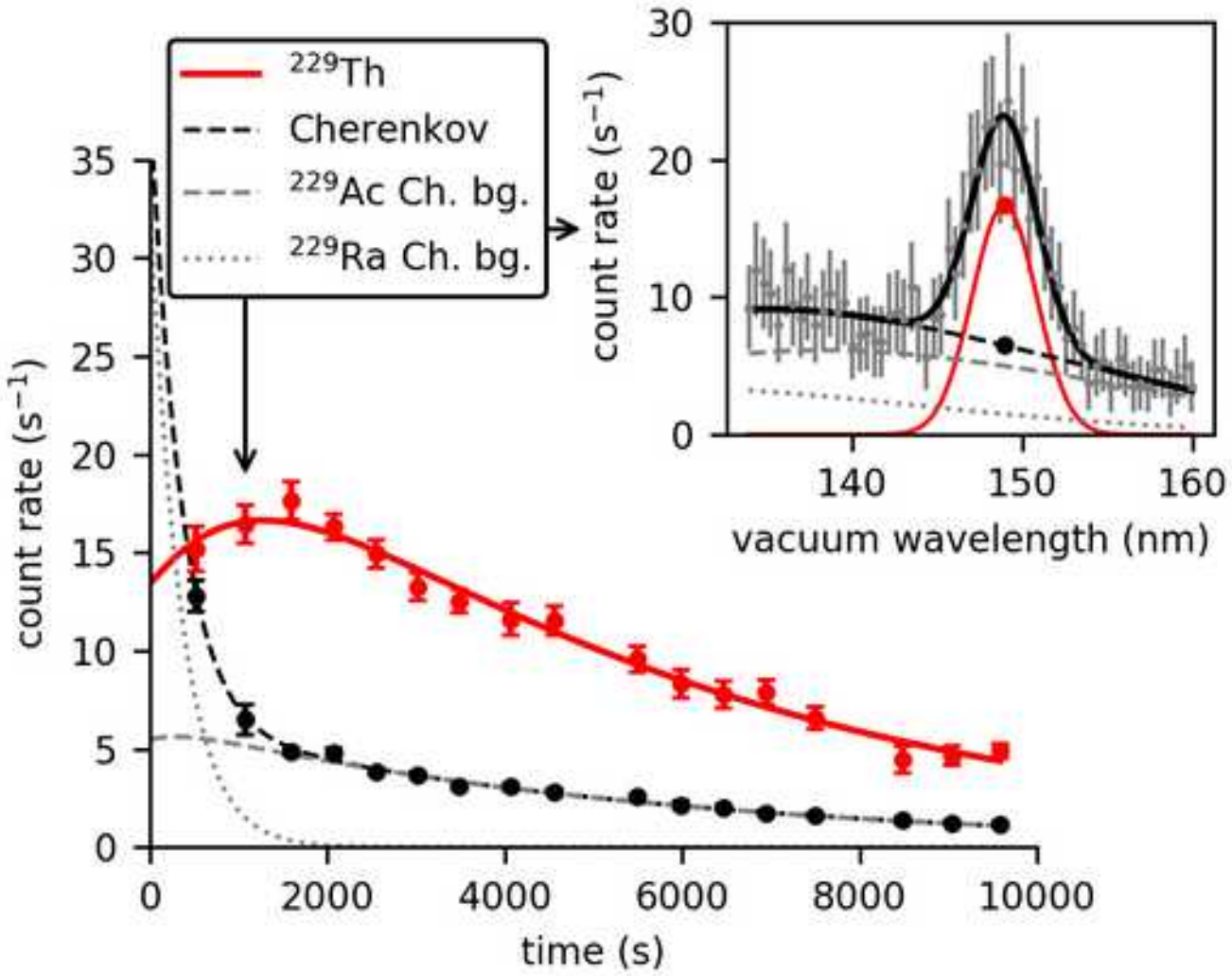


^{229m}Th radiative decay

CaF_2 defect radioluminescence

CaF_2 defect radioluminescence

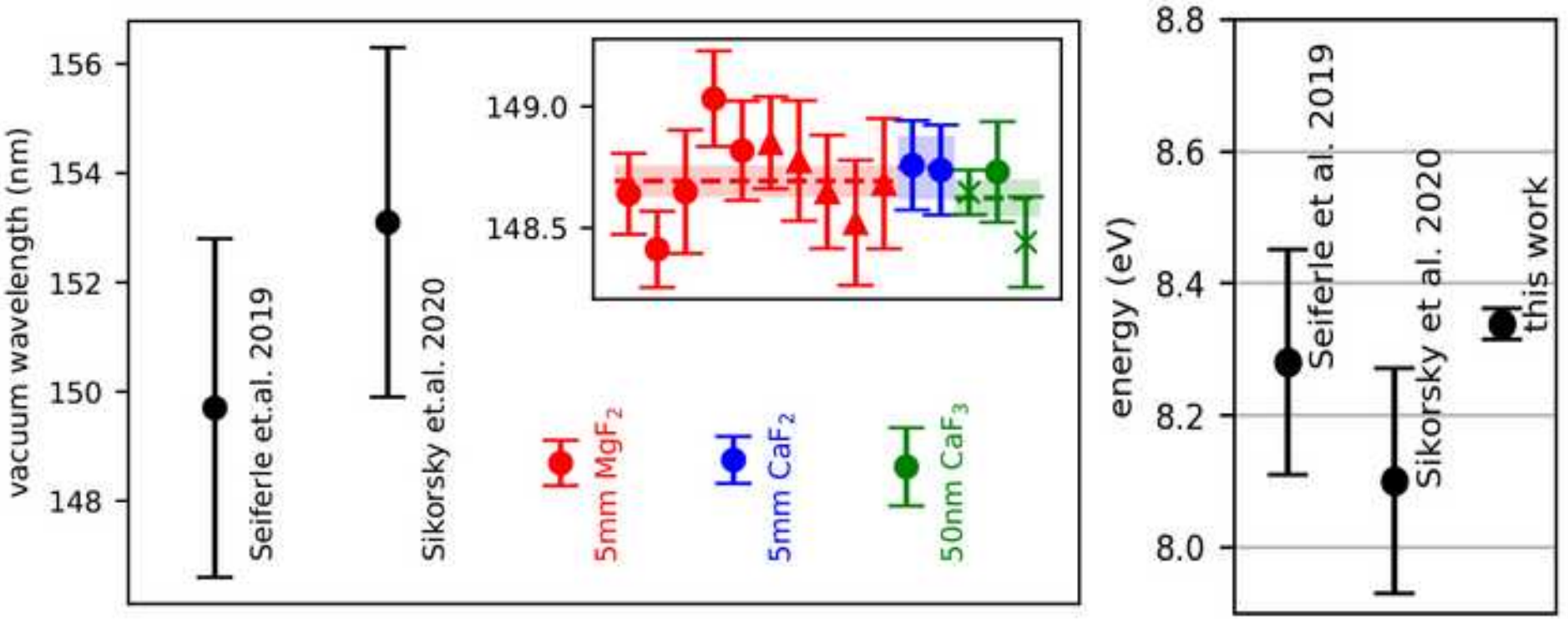
Half-life



$670(102)$ s in MgF_2

Crystal: 5 mm MgF_2
Entrance slit: 2 mm

Energy



New energy value:
8.338(24) eV

→ Uncertainty reduced from 41 THz to 5.8 THz

Conclusion and outlook

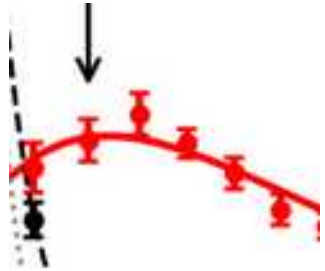
Conclusion

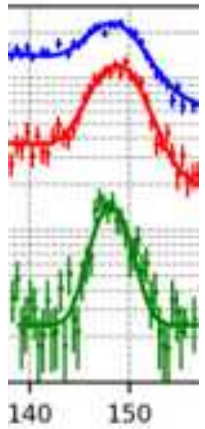
Method:

- Robust system populating the ^{229m}Th isomer using a ^{229}Ra and ^{229}Fr beam at ISOLDE
- Implantation technique: good performance of off-the-shelf large band-gap and thin-film crystals

Conclusions:

- Photon emission from a narrow linewidth emission found
  - In MgF_2 and CaF_2
  - Observed in $A=229$ and not present in $A=230$ decay chain
  - Signature of a mother-daughter decay with $T_{1/2} = 670(102)$ s (in MgF_2)
  - ➢ *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*





- New energy value: $E = 8.388(24)$ eV

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

VUV-spectroscopy at ISOLDE:

New measurement campaign:

IS715 beamtime took place last week

- *Half-life measurements*
- *Additional large-bandgap materials*

Thank you very much!

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Funding acknowledgements:
Fonds Wetenschappelijk Onderzoek: GOA/2015/010 BOF KU Leuven
Fonds Wetenschappelijk Onderzoek and Fonds de la Recherche Scientifique – FNRS Excellence of Science nr. 40007501,
Fundação para a Ciência e Tecnologia CERN/FIS-TEC/0003/2019),
Fonds zur Förderung der wissenschaftlichen Forschung I5971 (REThorIC),
European Union's Horizon 2020 ENSAR2 nr. 654002, MSCA nr. 101026762
European Research Council Thorium Nuclear Clock nr. 856415, LRC nr. 819957