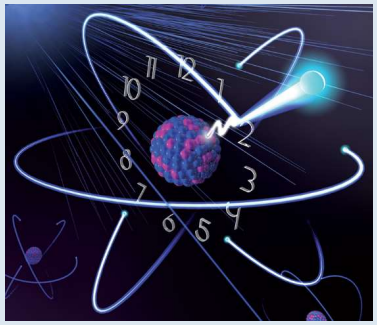
KU LEUVEN

19TH International Conference on Electromagnetic Isotope Separators and Related Topics
EMIS XIX

Ultraviolet Spectroscopy of the ^{229}Ac beta decay: The first observation of radiative decay of the ^{229}Th low-energy isomer

Piet Van Duppen
on behalf of the ISOLDE-IS658 collaboration

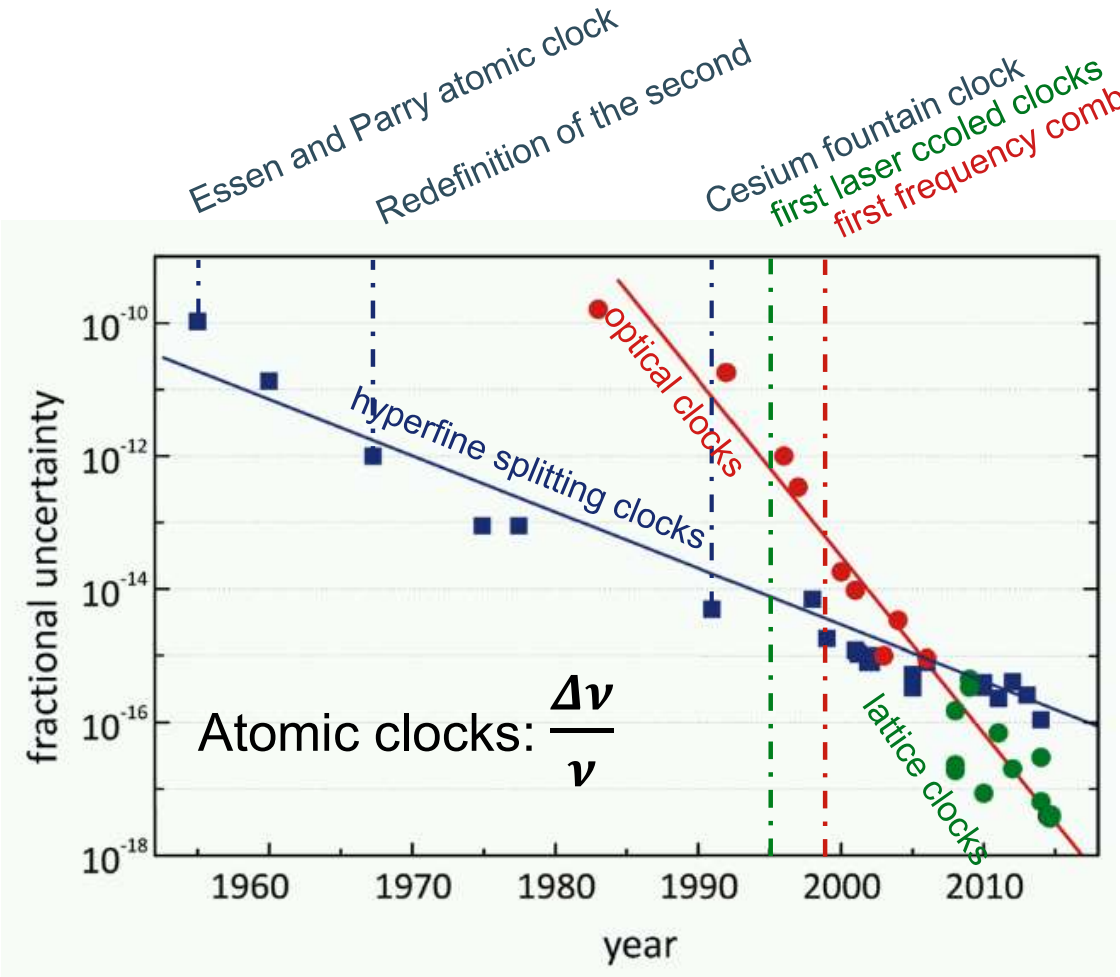


Outline:

- The road towards a nuclear clock
- VUV spectroscopy of ^{229m}Th at ISOLDE
- Results from experiment IS658
- Conclusion and Outlook

Thirolf et al., 2022, CERN Courier 62 - APS/Ann. Phys. 531 1800381

The road towards a nuclear clock



Limitations:

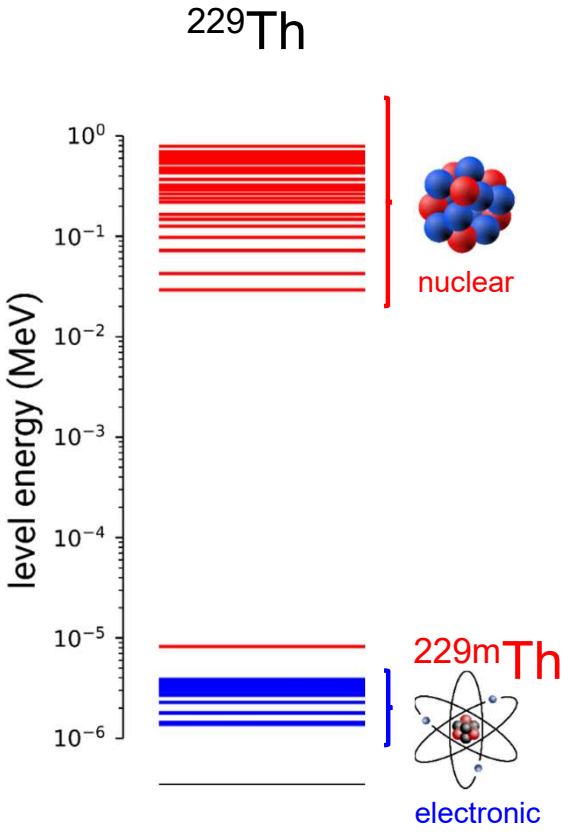
- Susceptibility to environment
- Transition properties (ΔE/E)

Modified from Riehle, 2016 *C.R.Physiques* **16** 506-515

The road towards a nuclear clock

➢ A nuclear clock based on ^{229m}Th

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



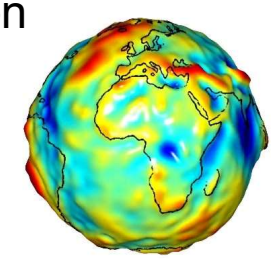
➢ Potential clock operation at 10^{-19} relative precision

➢ Fundamental physics

- Temporal variation of the fine-structure constant
- Dark matter searches

➢ Potential applications

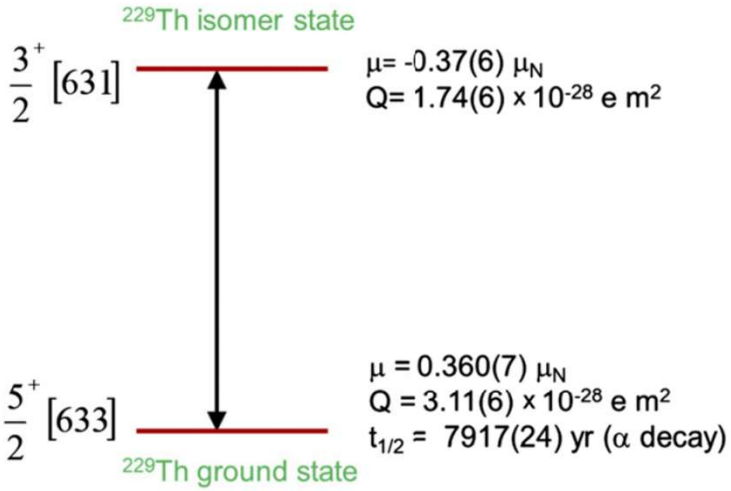
- Satellite-based navigation
- Geodesy

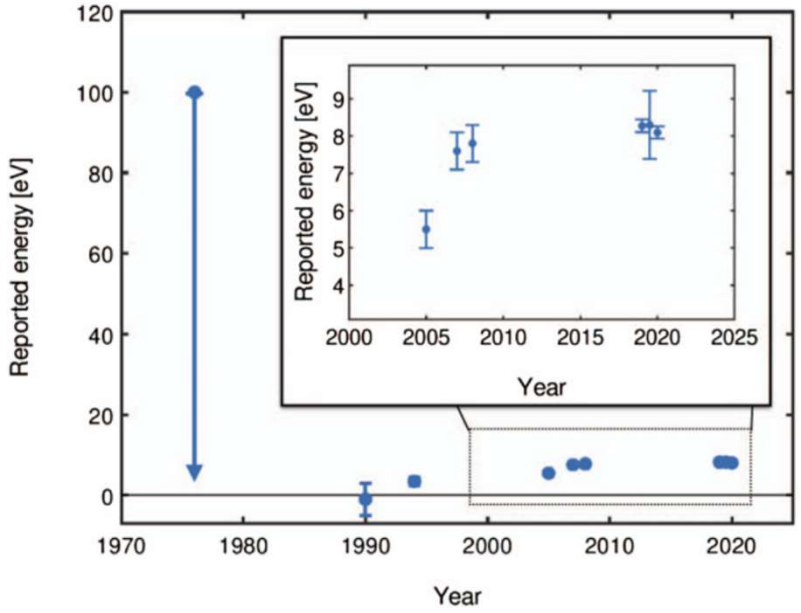


von der Wense et al, 2016 *Nature* **533** 47-51
Peik and Tamm 2014 *J. Phys. Condens. Matter* **26** 10
Campbell et.al. 2012 *PRL* **108** 12

Present knowledge

- Energy: 8.10 (17) eV (153.1 nm) and 8.28 (17) eV (149.7 nm) (> IP thorium = 6.3 eV)
- Competition between IC and Radiative decay (charge-state dependent): $\alpha \approx 10^9$
- 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





Open questions

- Radiative decay: not yet observed
- Energy: more precise value
- Lifetime of the radiative decay channel

Challenges

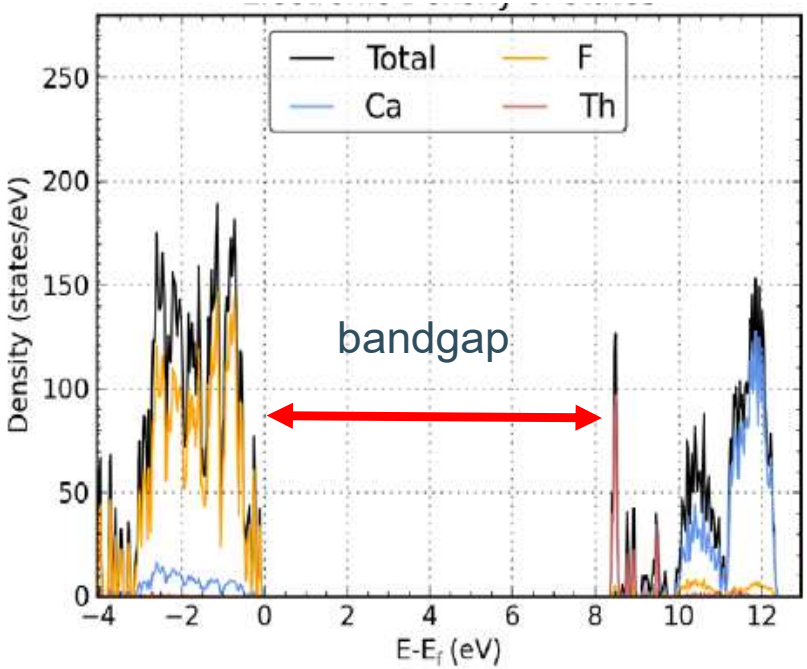
- Efficient population of the isomer
- Control over decay channel

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**

The road towards a nuclear clock

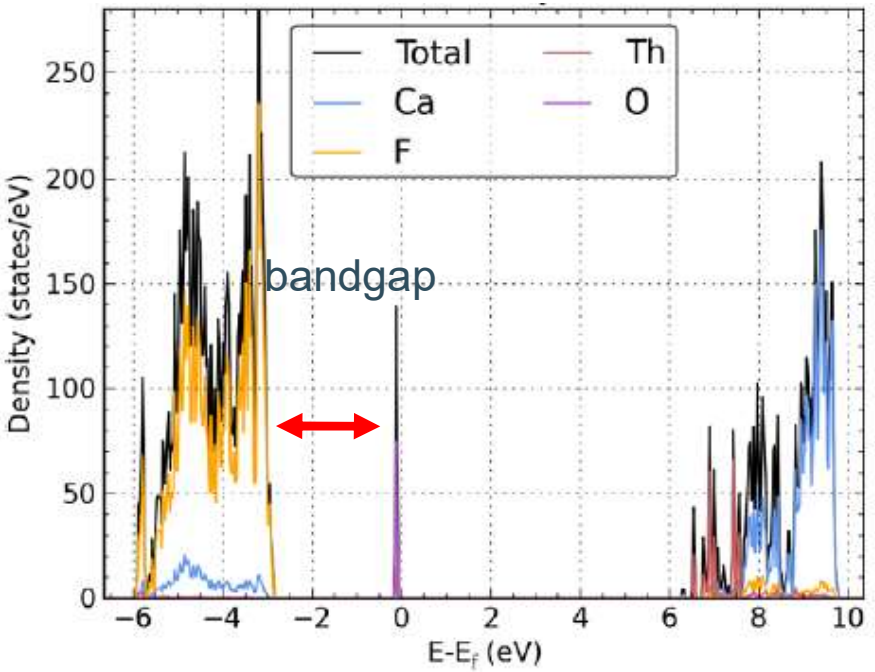
- Embedding in large-bandgap crystals (MgF_2, CaF_2,..) to achieve high charge state (Th^{3+}, Th^{4+})

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



-> radiative decay

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



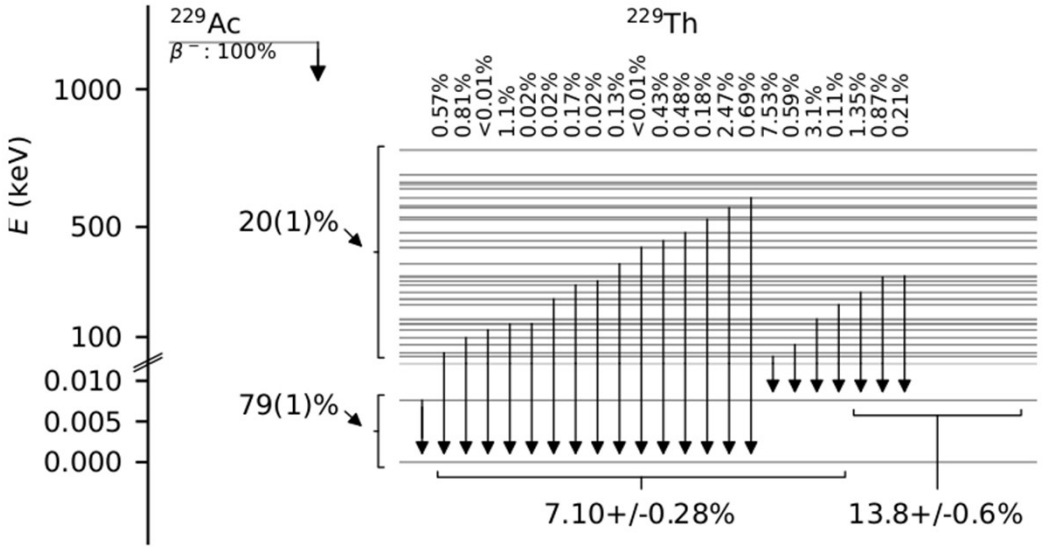
-> emission of a conversion electron

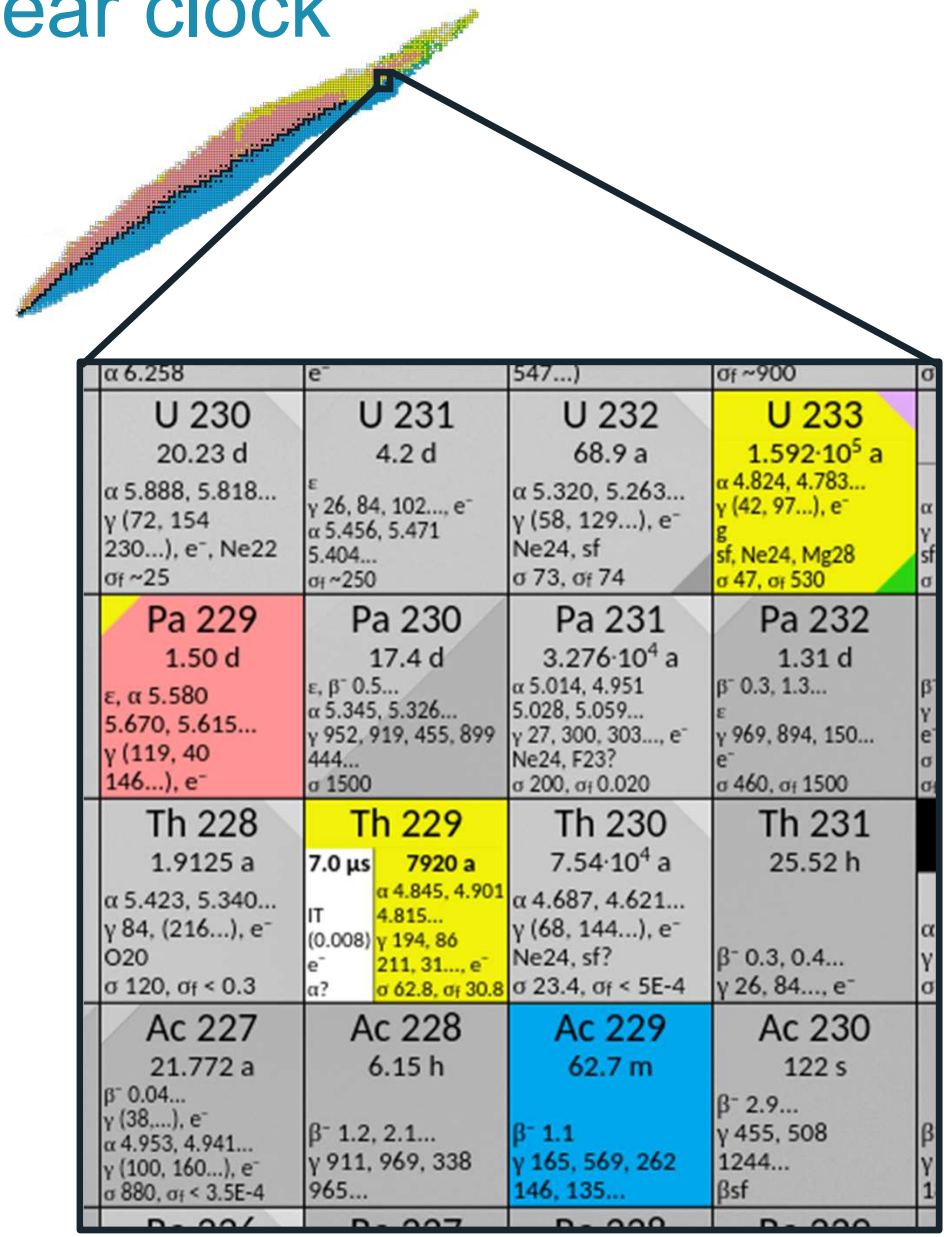
G A Kazakov et al 2012 *New J. Phys.* **14** 083019
Dessovic *et al.* 2014 *J. Phys. Condens. Matter* **26** 10

The road towards the nuclear clock

Efficient population in radioactive decay

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





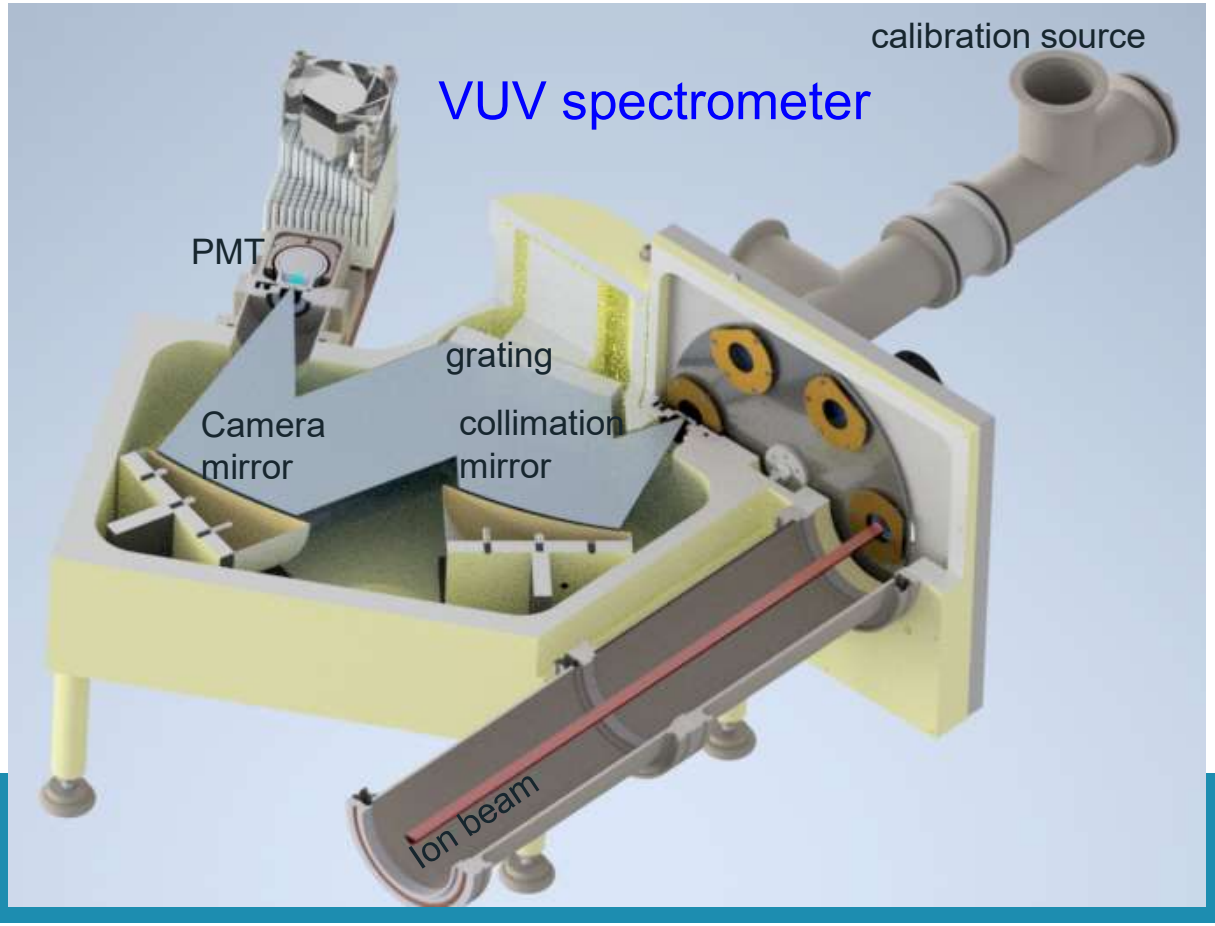
Verlinde, Kraemer, Moens *et al.* 2019 *PRC* **100** 024315

Vacuum-ultraviolet spectroscopy of ^{229m}Th at ISOLDE

VUV spectroscopy at ISOLDE

- ISOLDE (CERN): 1.4 GeV protons on UCx – surface ionization
- Beam composition: ^{229}Fr ($T_{1/2}$ = 50.2 s, ~ 10^5 pps), ^{229}Ra (4.0 m, ~ 10^6 pps)
- A = 230 and 231 beams used as proxy
- CaF_2 (E_{gap} = 11.8 eV) and MgF_2 (10.8 eV)

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



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)

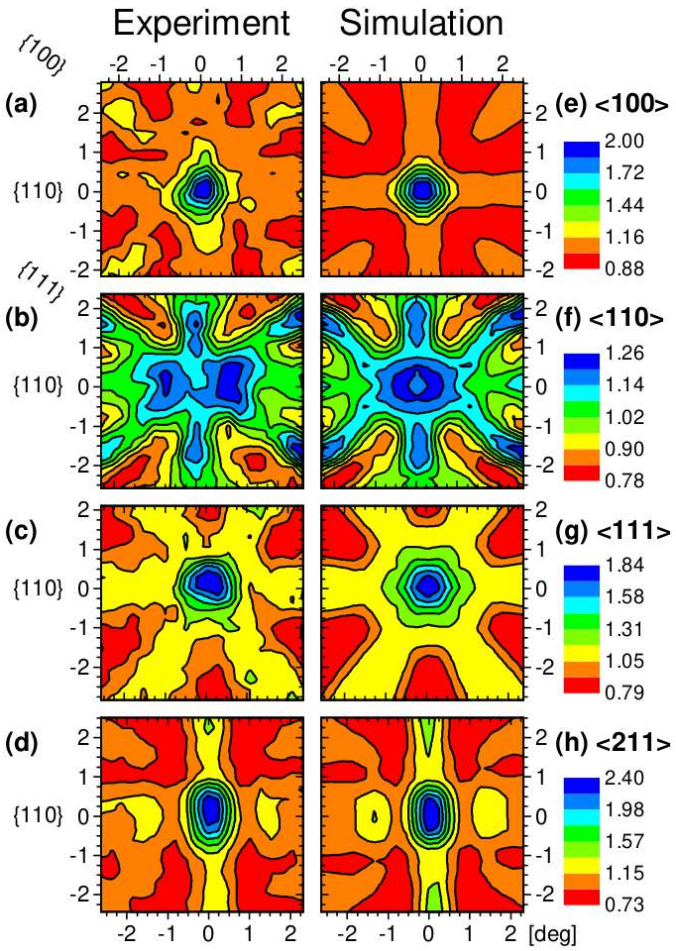
$$\varepsilon_{total} \simeq 10^{-3}\ at\ 149\ nm$$

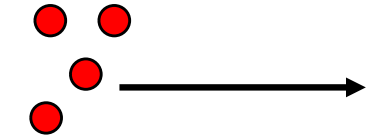
Results
from experiment IS658

VUV spectroscopy at ISOLDE

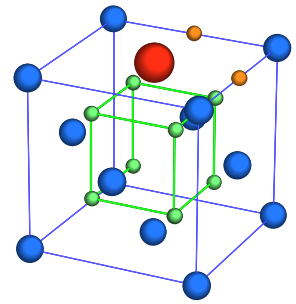
Emission Channeling:

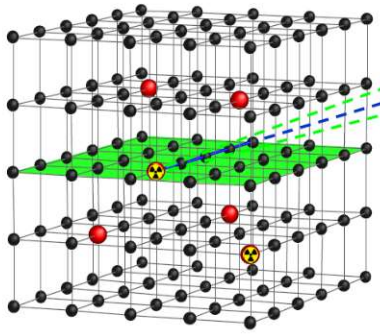
^{231}Ra ($T_{1/2}$ = 104 s) → ^{231}Ac ($T_{1/2}$ = 7.5 m) → ^{231}Th (25.5 h) → ^{231}Pa decay



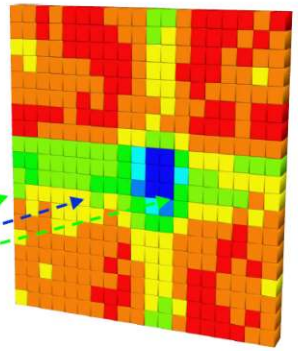


dopant
^{231}Ac





Host crystal

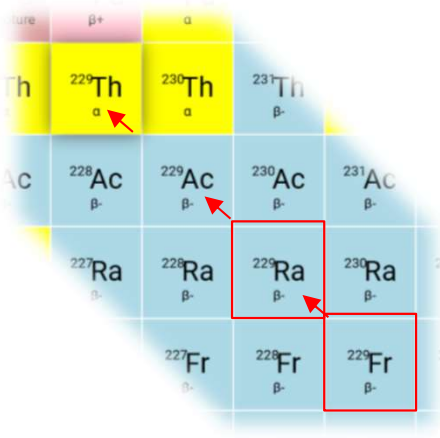


Position sensitive detector

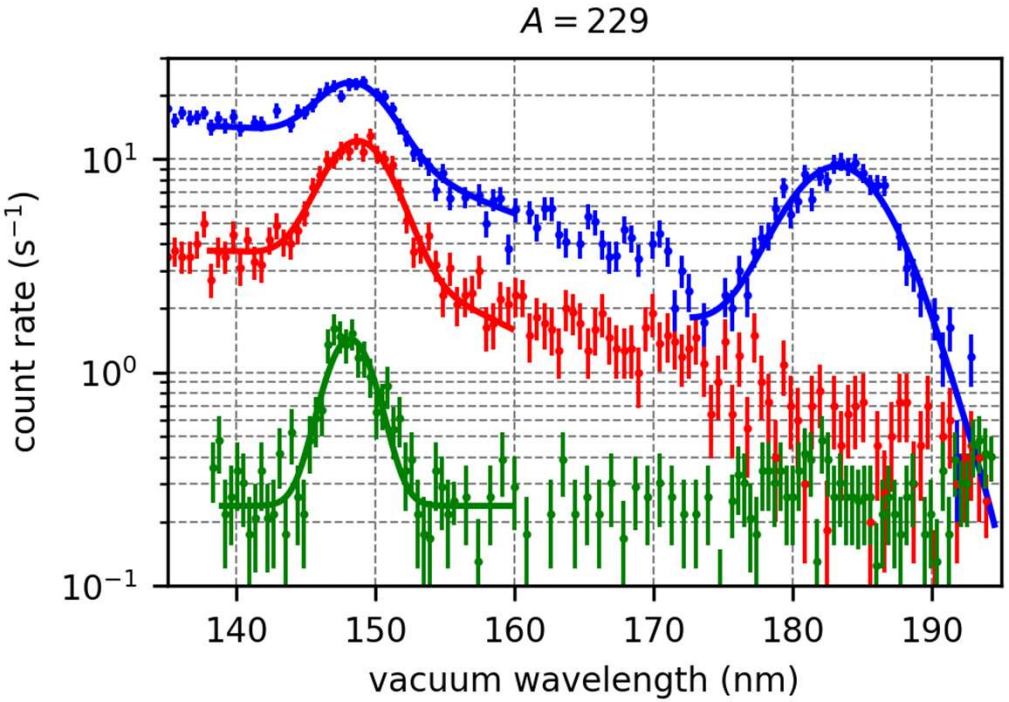
- Using the implantation technique: majority (70% to 100%) in substitutional position

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

Identification

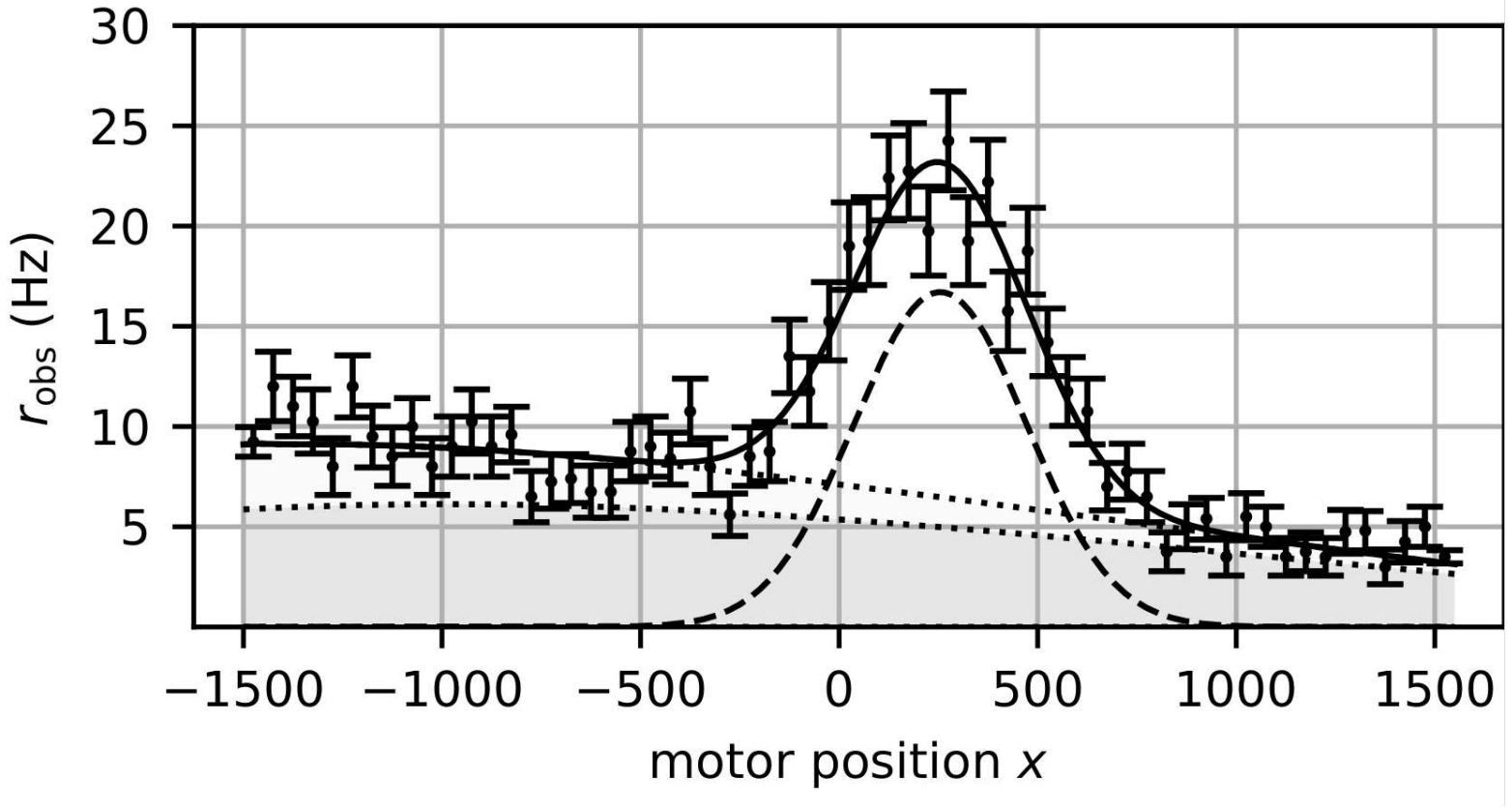


5mm thick MgF_2
5mm thick CaF_2
50nm thick CaF_2



(3 mm entrance slit (broad linewidth))

Wavelength spectra



PMT dark counts
0.25-1 Hz

Gaussian peak:
Monochromatic emission line

$$r(x(t)) = r_{\mathrm{dark}} + b_0 \exp\left(-\frac{t\ln(2)}{62.7\,\mathrm{min}}\right)\exp\left(-\frac{(x-x_0)^2}{2\sigma^2}\right)$$

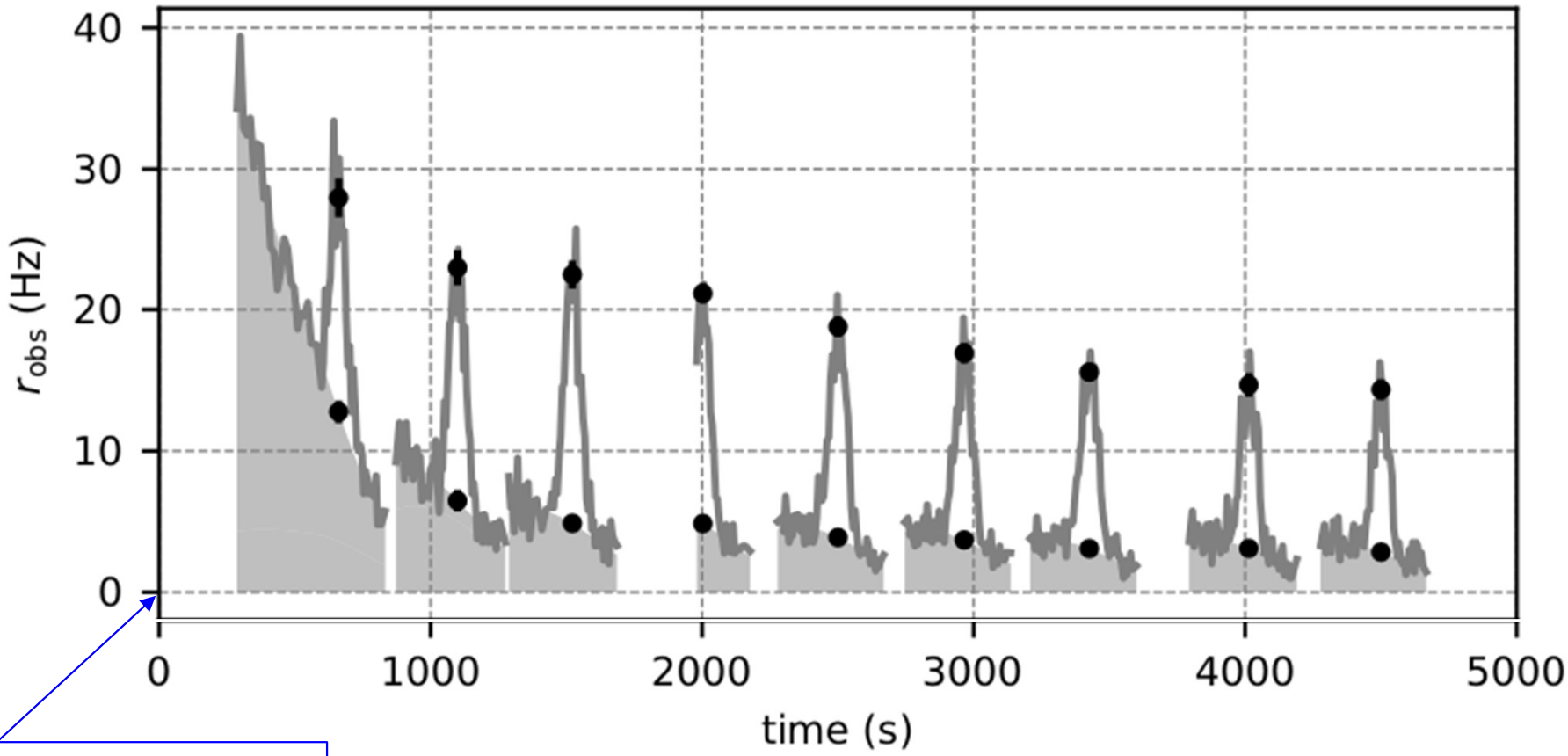
^{229}Ra Cherenkov background

^{229}Ac Cherenkov background

$$+b_1 \exp\left(-\frac{t\ln(2)}{62.7\,\mathrm{min}}\right)\sum_{j=0}^{3} a_j x^j + b_2 \exp\left(-\frac{t\ln(2)}{4\,\mathrm{min}}\right)\sum_{j=0}^{3} a_j x^j$$

Time behaviour

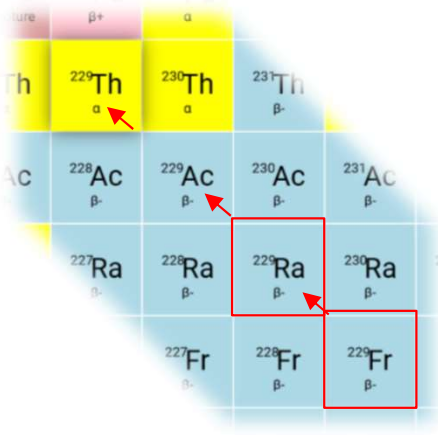
A = 229 implantation for 3442 s in a MgF_2 crystal

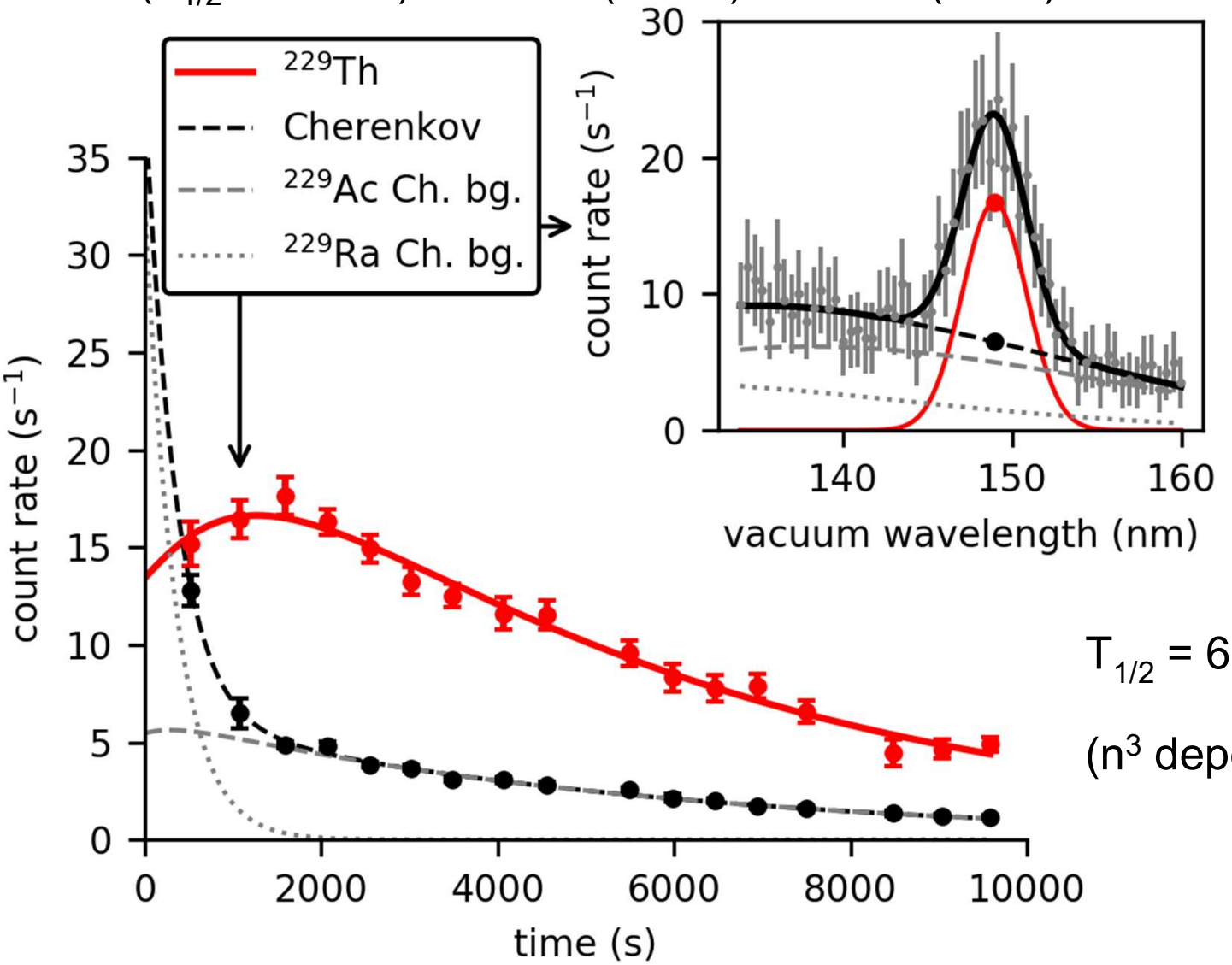


(2 mm entrance slit (broad linewidth))

Time behaviour

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





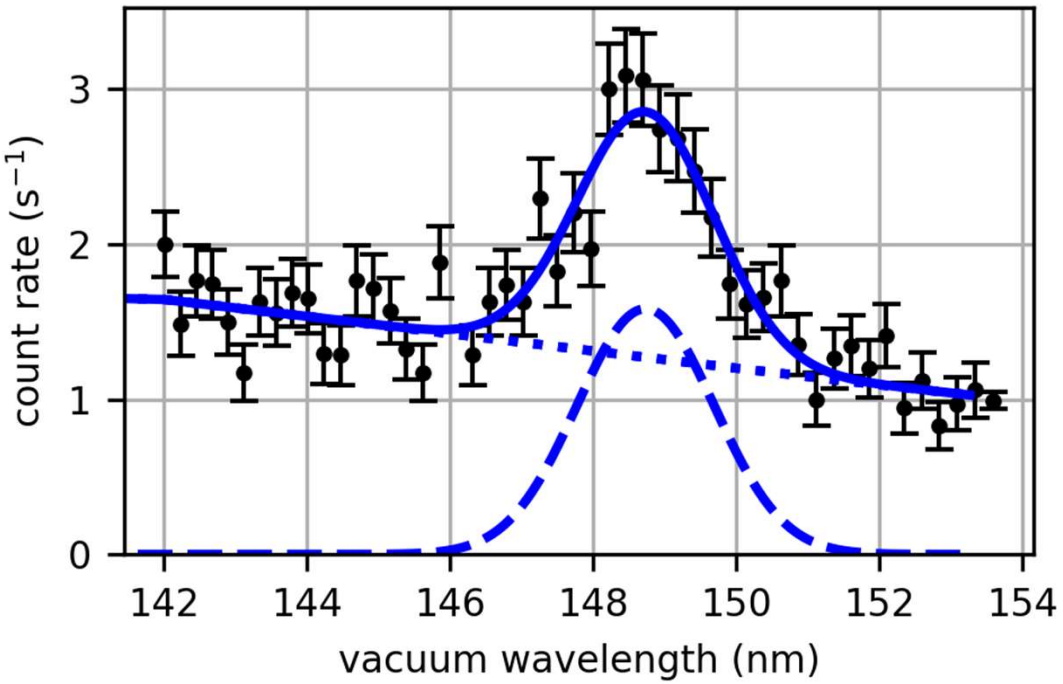
$T_{1/2}$ = 670 (102) s in MgF_2

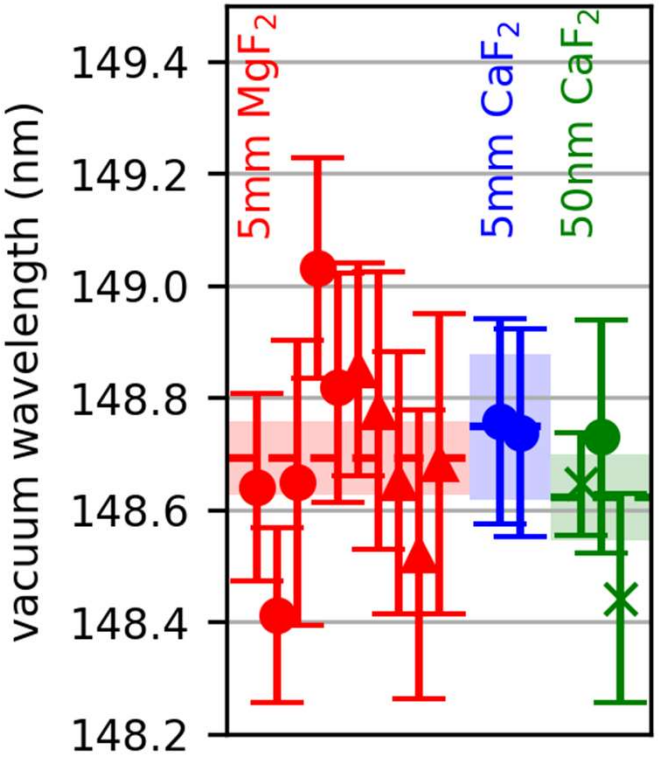
(n^3 dependence: 2.21(32) 10^3 s)

Tkalya et al., PRC61 (2000)

Energy

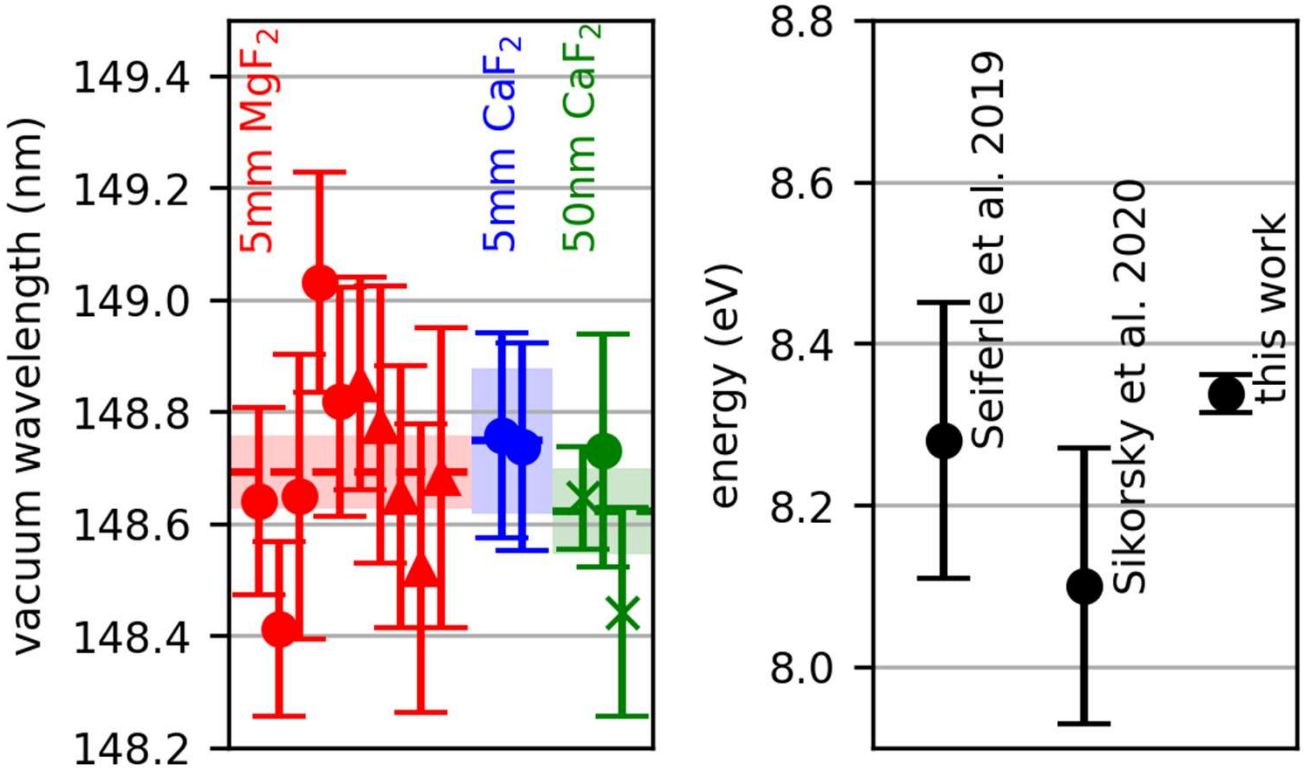
Entrance slit: 0.5 mm
Resolution: 2.25 (21) nm FWHM





taken with 1 (x), 0.5 (•) and 0.25 (Δ) mm entrance slit (narrow linewidth)

Energy



$\lambda = 148.71 \pm 0.06(\text{stat.}) \pm 0.41(\text{syst.})$ nm

$E = 8.338 \pm 0.003(\text{stat.}) \pm 0.023(\text{syst.})$ eV

Seiferle et al., 2019 *Nature* **573** 243-246
Sikorsky et al., 2020 *PRL* **125** 142503

Conclusion and Outlook

Conclusion and Outlook

Conclusions

- Robust system populating the ^{229m}Th isomer using a ^{229}Ra and ^{229}Fr beam at ISOLDE
- 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 ^{229}Th isomer

→ new energy value: $E = 8.388\ (24)$ eV

→ fraction of VUV photon emitting ^{229,m}Th: lower limit of 1 - 7 %

→ isomer detection in crystal matrix: consequences for a solid-state-based ^{229m}Th clock

Outlook:

- New measurement campaign in preparation:
  half-life measurement in CaF_2, energy at 0.1nm uncertainty, additional crystal materials
- Study of the decay-rate-dependence on the material

ISOLDE-IS658 collaboration:

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