

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

Sandro Kraemer



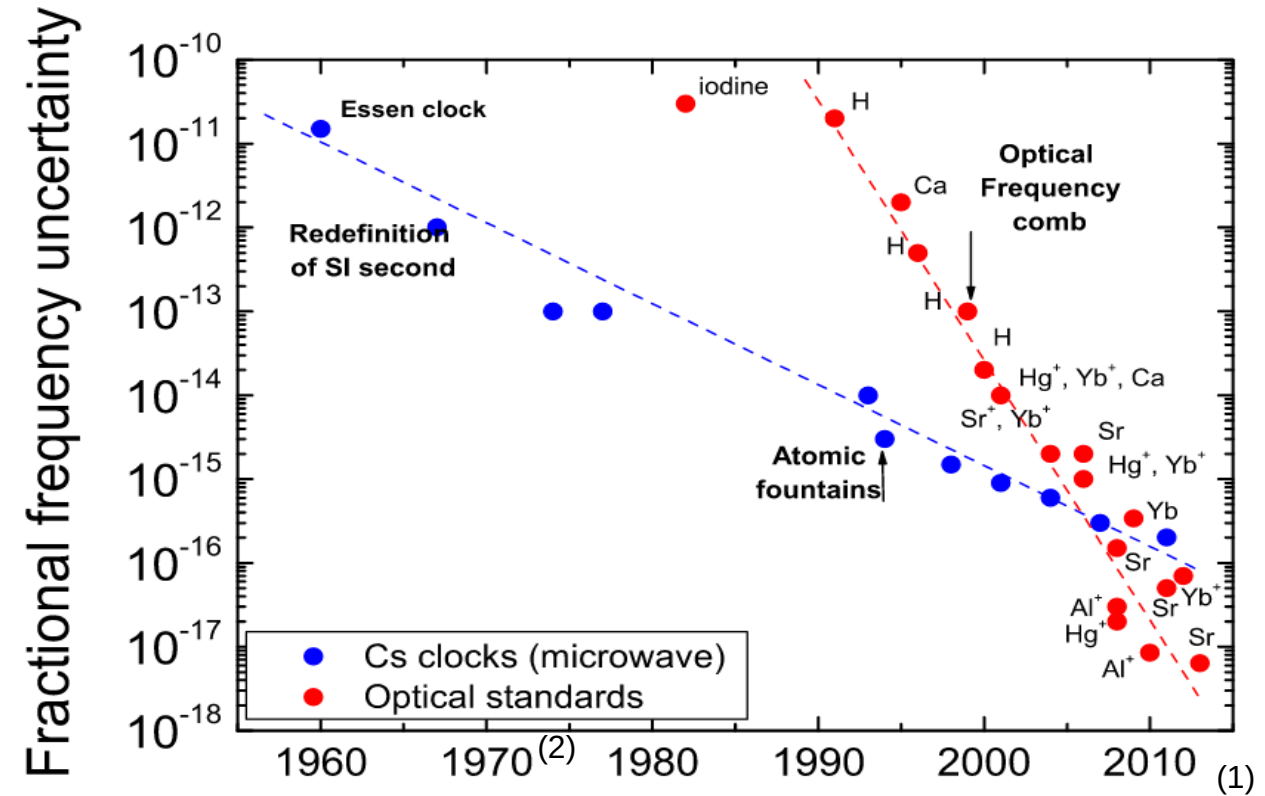
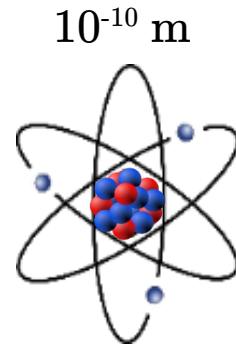
Outline:

- From atomic to nuclear clocks
- Vacuum-ultraviolet spectroscopy at ISOLDE
- Conclusion and Outlook

From atomic to nuclear clocks



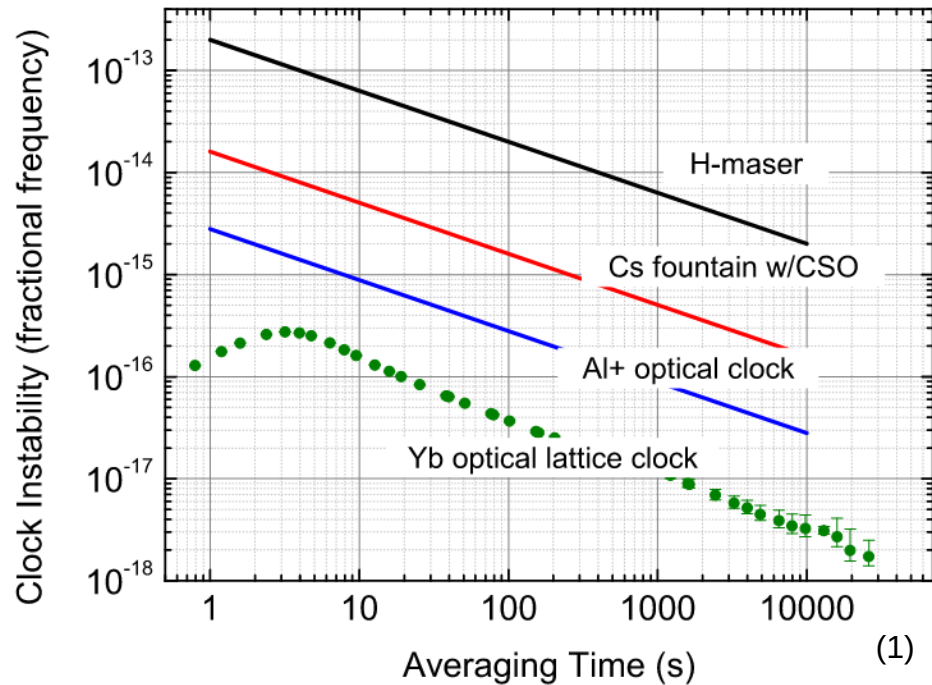
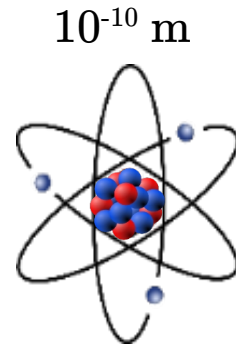
From atomic to nuclear clocks



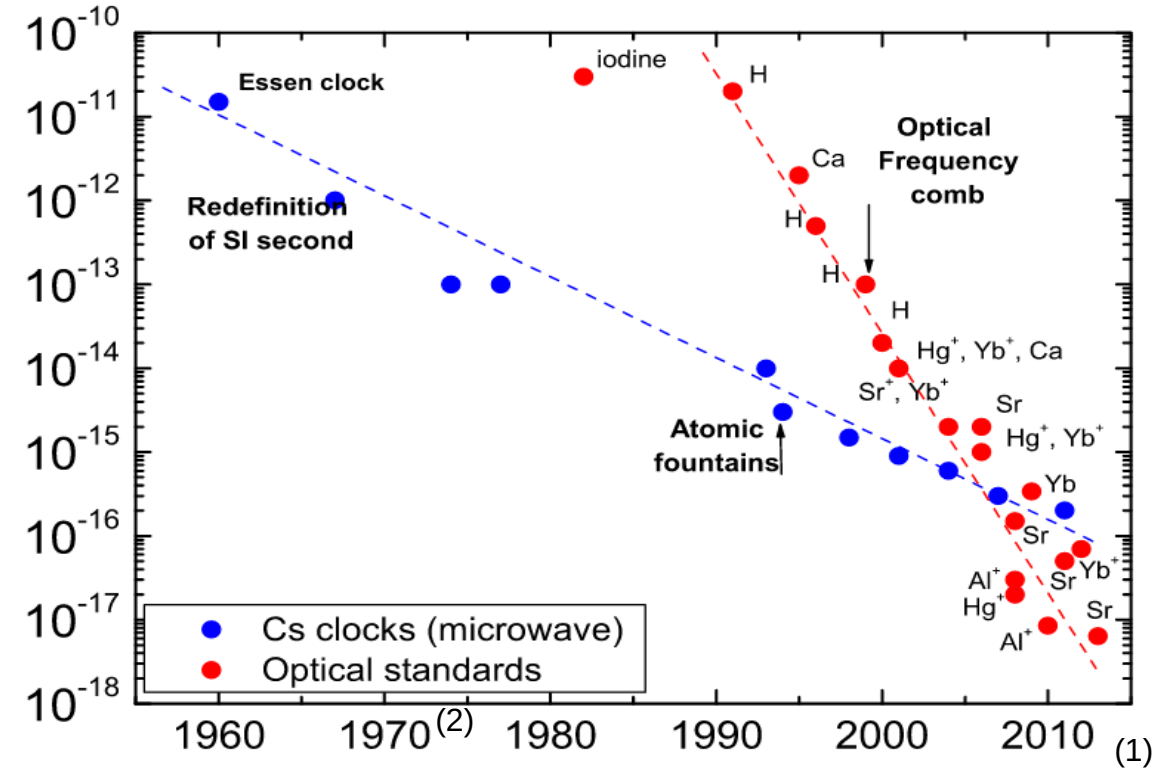
- Accuracy limited by uncertainty on environmental influences

(1)

From atomic to nuclear clocks



Fractional frequency uncertainty

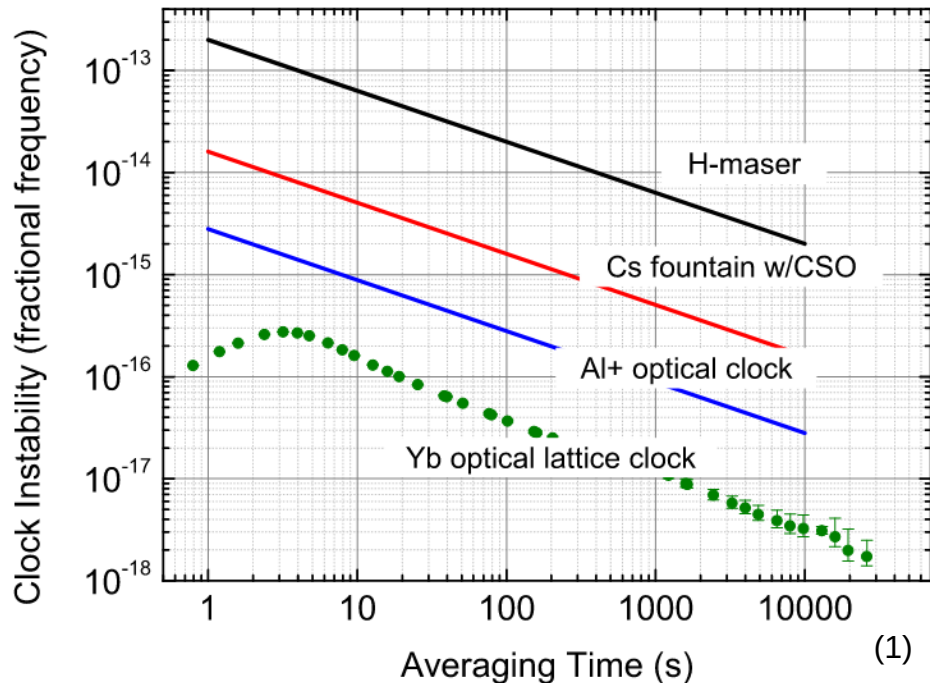
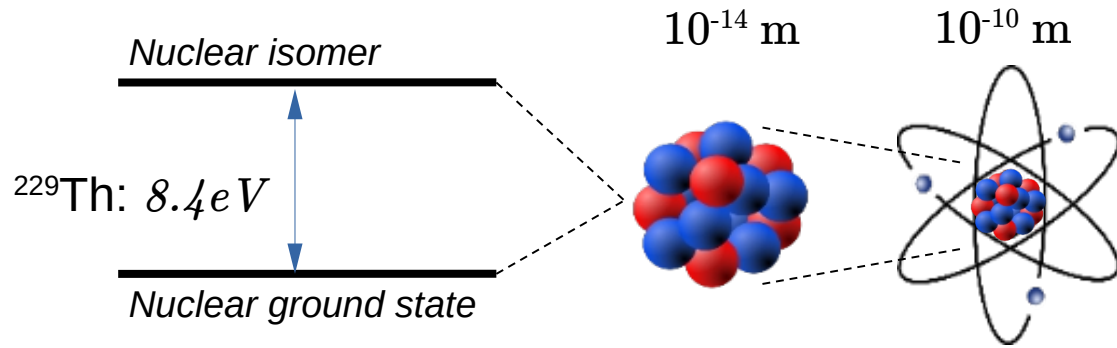


- Accuracy limited by uncertainty on environmental influences
- Stability limited by amount of interrogable two-level systems

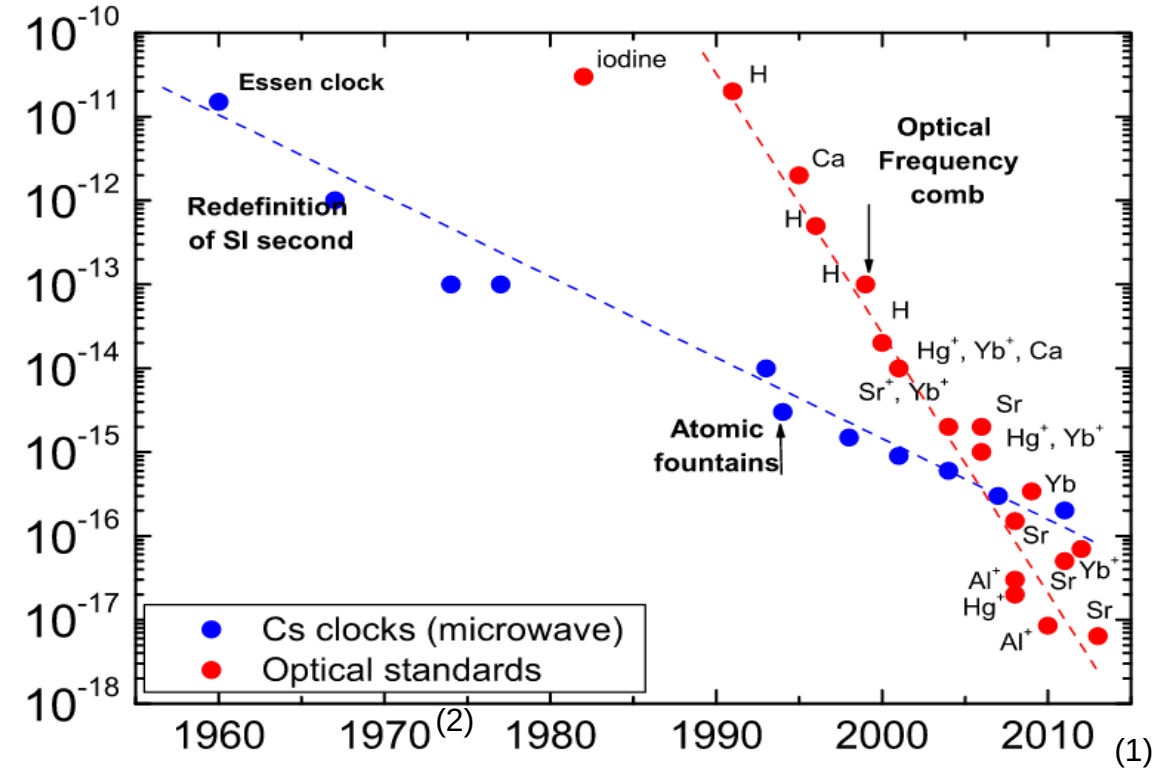
(1): Poli et al., 2013, *Rivista del Nuovo Cimento* **36** (12)

(2): Peik and Tamm, 2003, *Europhys. Lett.* **61** 2

From atomic to nuclear clocks



Fractional frequency uncertainty



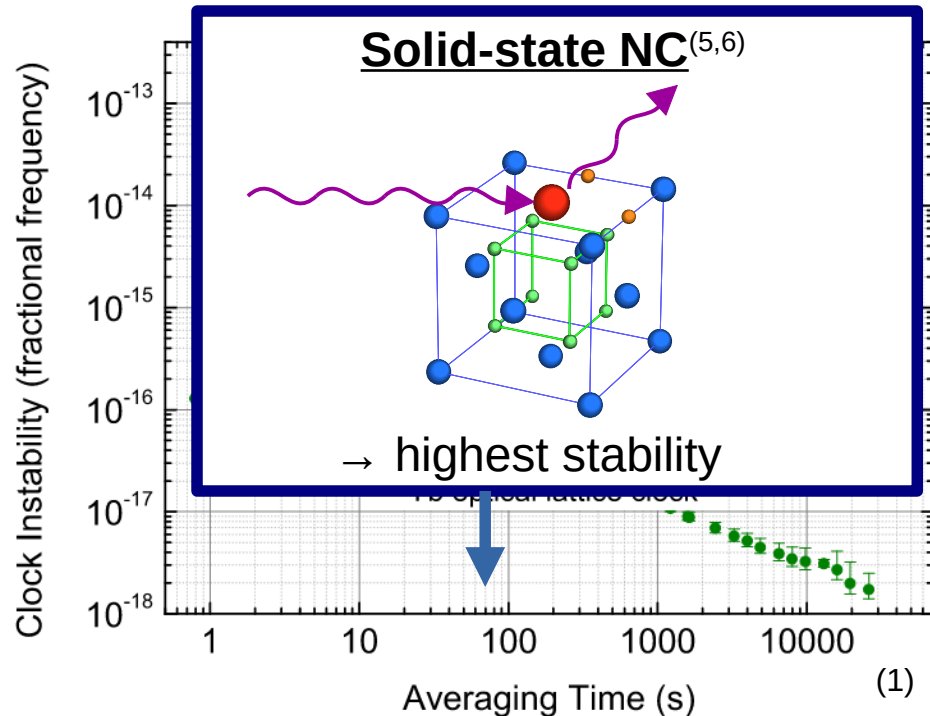
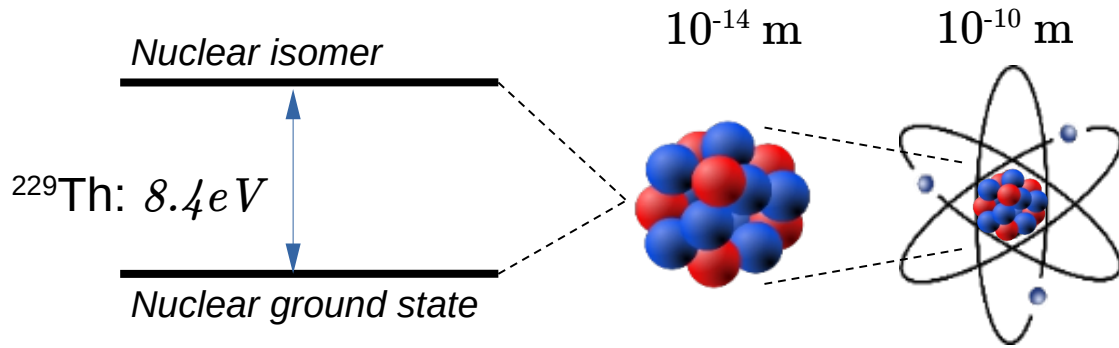
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→ From electronic to nuclear transition⁽²⁾

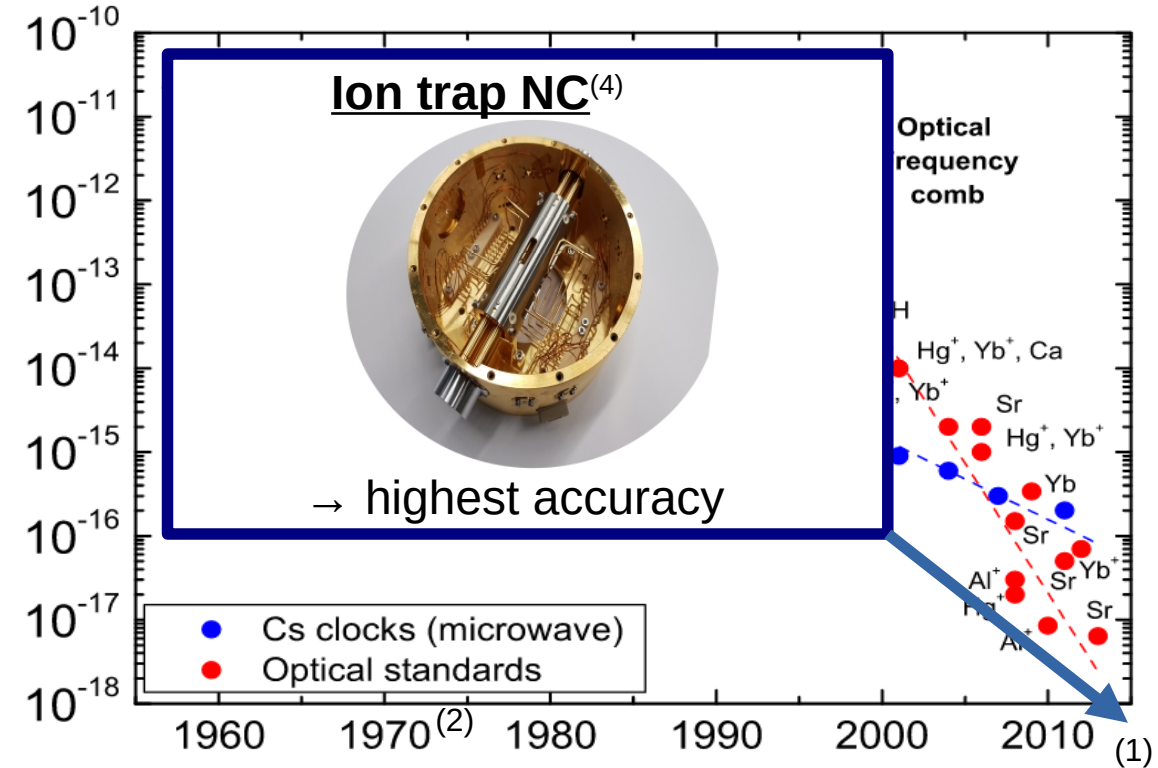
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From atomic to nuclear clocks



Fractional frequency uncertainty



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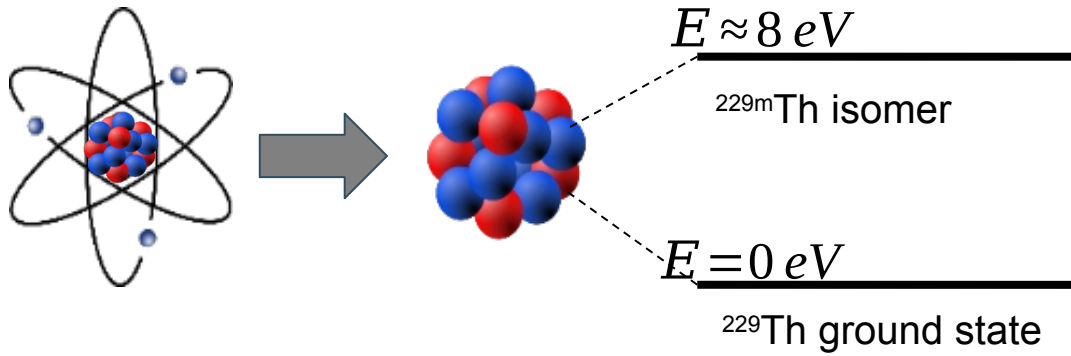
→ From electronic to nuclear transition⁽²⁾

(4) Peik et al., 2021, *Quantum. Sci. Technol.* **6** 034003

(5) G A Kazakov et al., 2012 *New J. Phys.* **14** 083019

(6) von der Wense et al., 2019, *Hyperf. Interact.* **240** 2

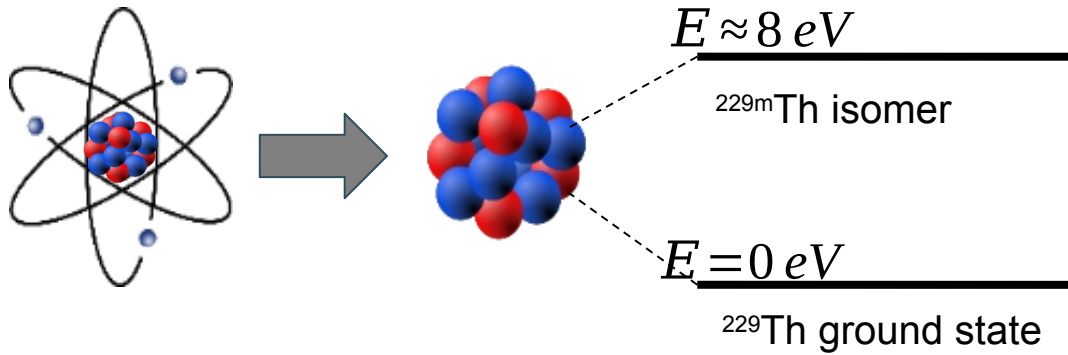
A novel quantum sensor



Atomic clock: electron
EM force

Nuclear clock: nucleon transition
EM force + nuclear forces

A novel quantum sensor

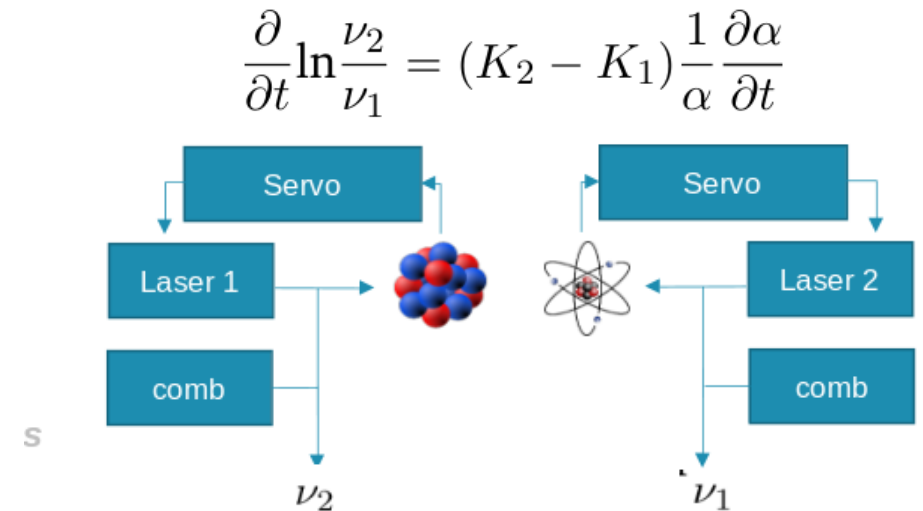


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 EM force

Nuclear clock: nucleon transition
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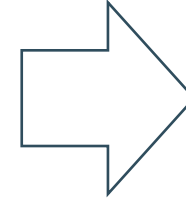
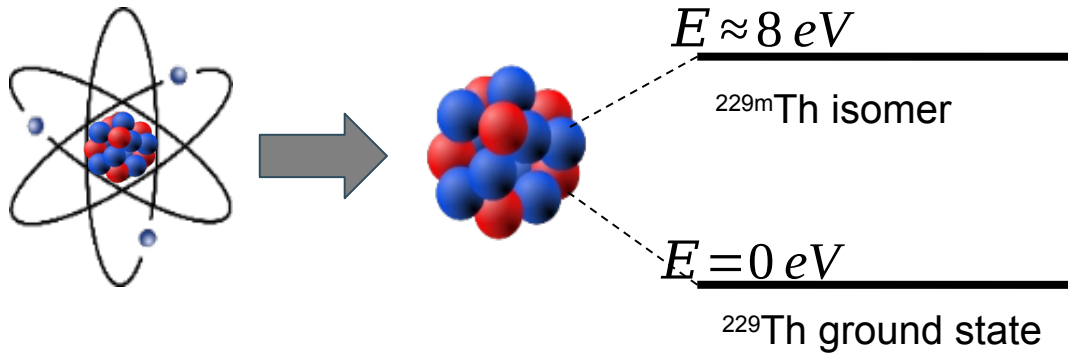
- **Lineshape analysis**⁽⁷⁾
- **Clock comparisons (-networks)**⁽⁸⁾
time variations of fundamental constants
 → up to 10^4 times more sensitive than optical clock comparisons
oscillatory ultralight dark matter (QCD coupling)
 → up to 10^8 more sensitive than atomic clock comparisons



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

(8) Fuchs et al. *ArXiv* 2407.15924

A novel quantum sensor

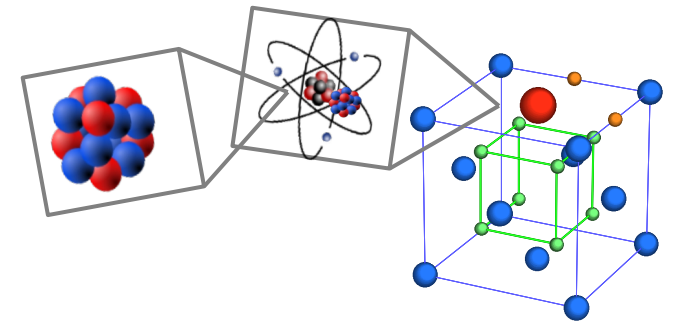


Atomic clock: electron
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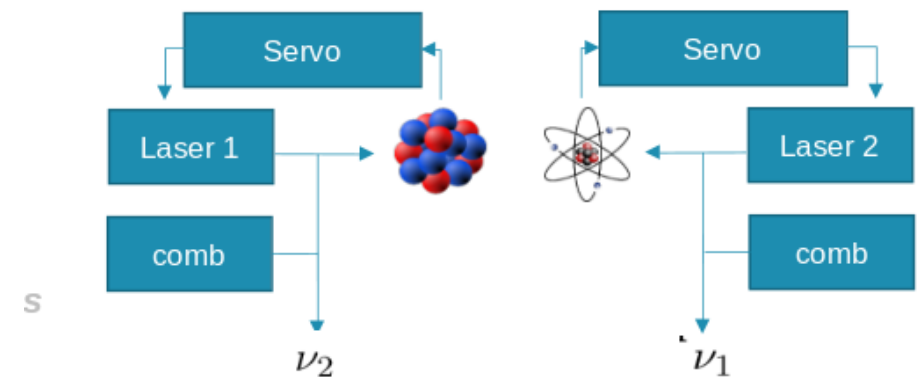
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Large number of nuclei in crystalline medium⁽⁸⁾

- **Ultrastable clock (-networks)**
gravimetry
- **Industrial applications**
telecom, satellite-based navigation

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

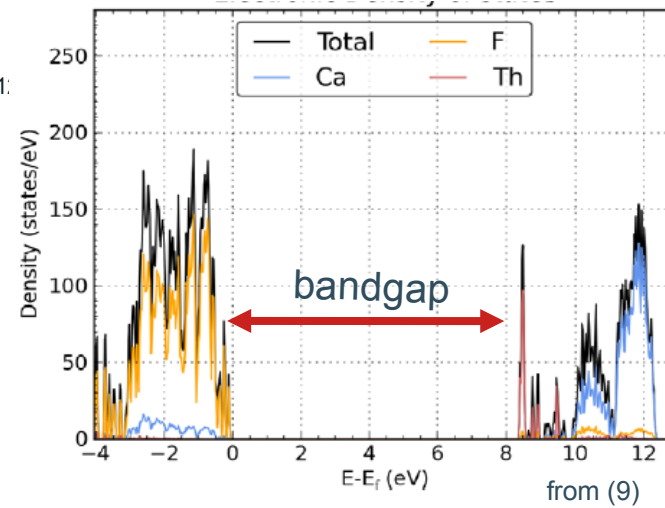
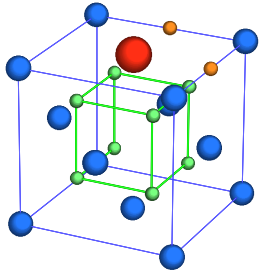


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The role of the chemical environment

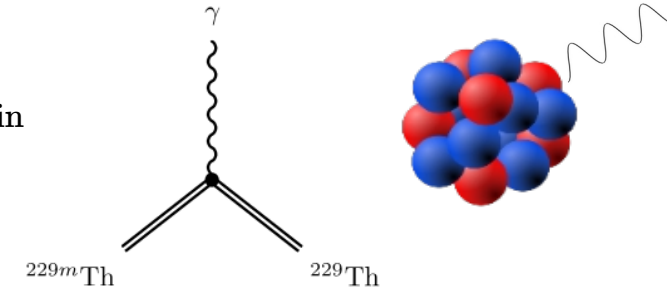
CaF_2 : $^{229}\text{Th}^{4+}$ in Ca-substitutional position⁽¹⁾



Decay mode:

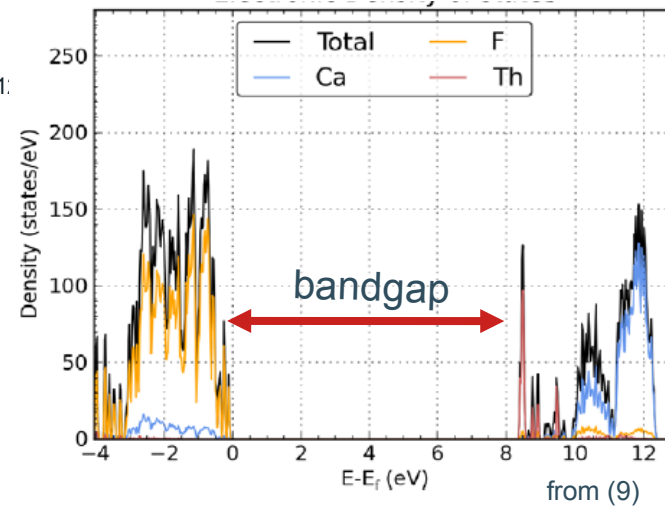
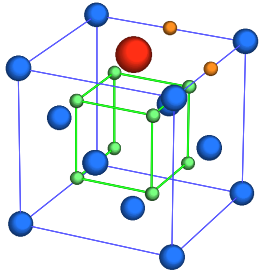
γ -decay

$$1/\Gamma_\gamma = T_{1/2} \approx 10 \text{ min}$$

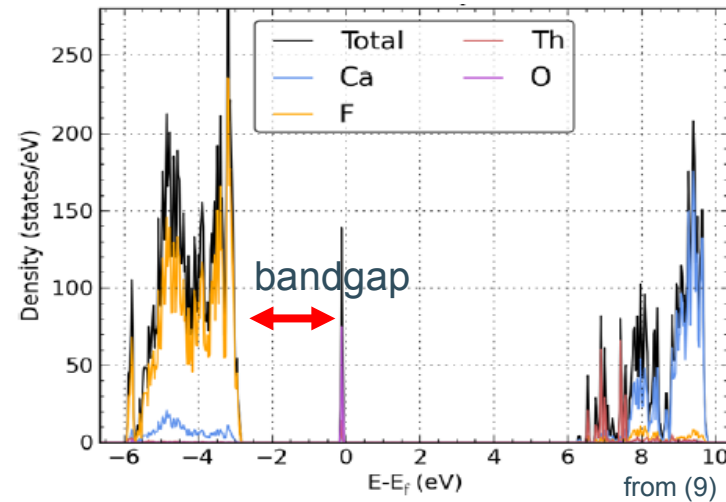
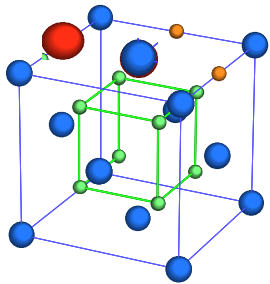


The role of the chemical environment

CaF_2 : $^{229}\text{Th}^{4+}$ in Ca-substitutional position⁽¹⁾



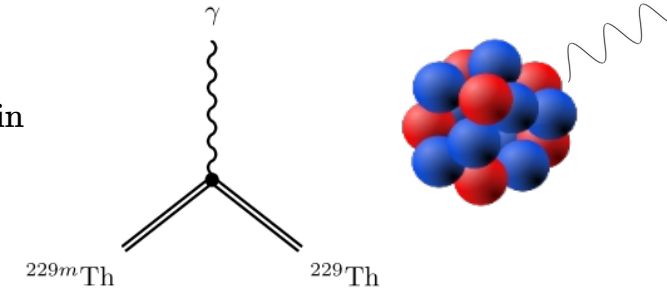
CaF_2 : $^{229}\text{Th}^{4+}$ in interstitial position⁽¹²⁾



Decay mode:

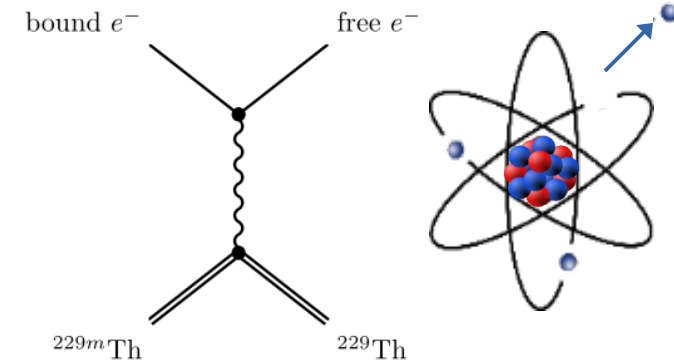
γ -decay

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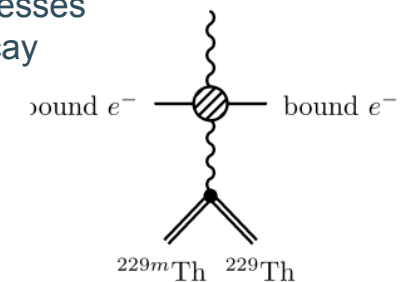


Internal conversion (IC) decay

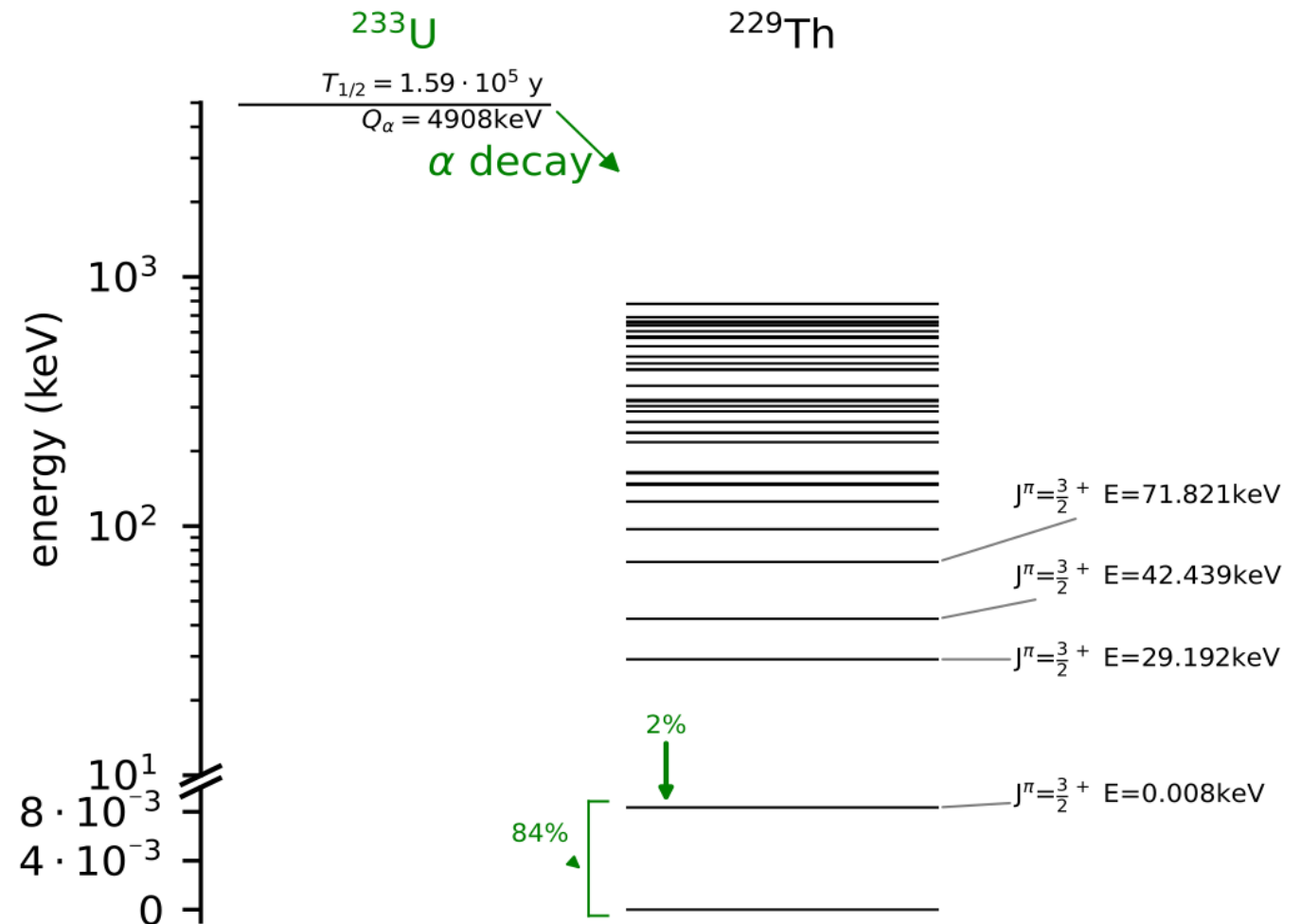
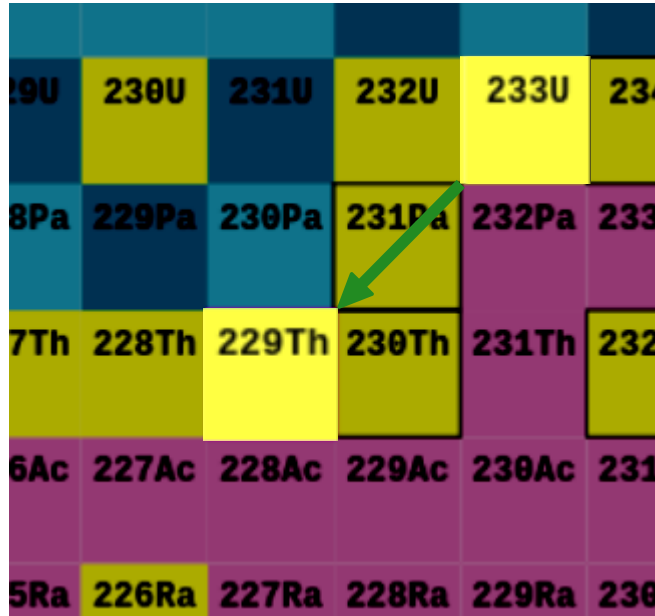
$$1/\Gamma_{\text{IC}} = T_{1/2} \approx 7 \mu\text{s}$$



Higher order processes
e.g. bound IC-decay



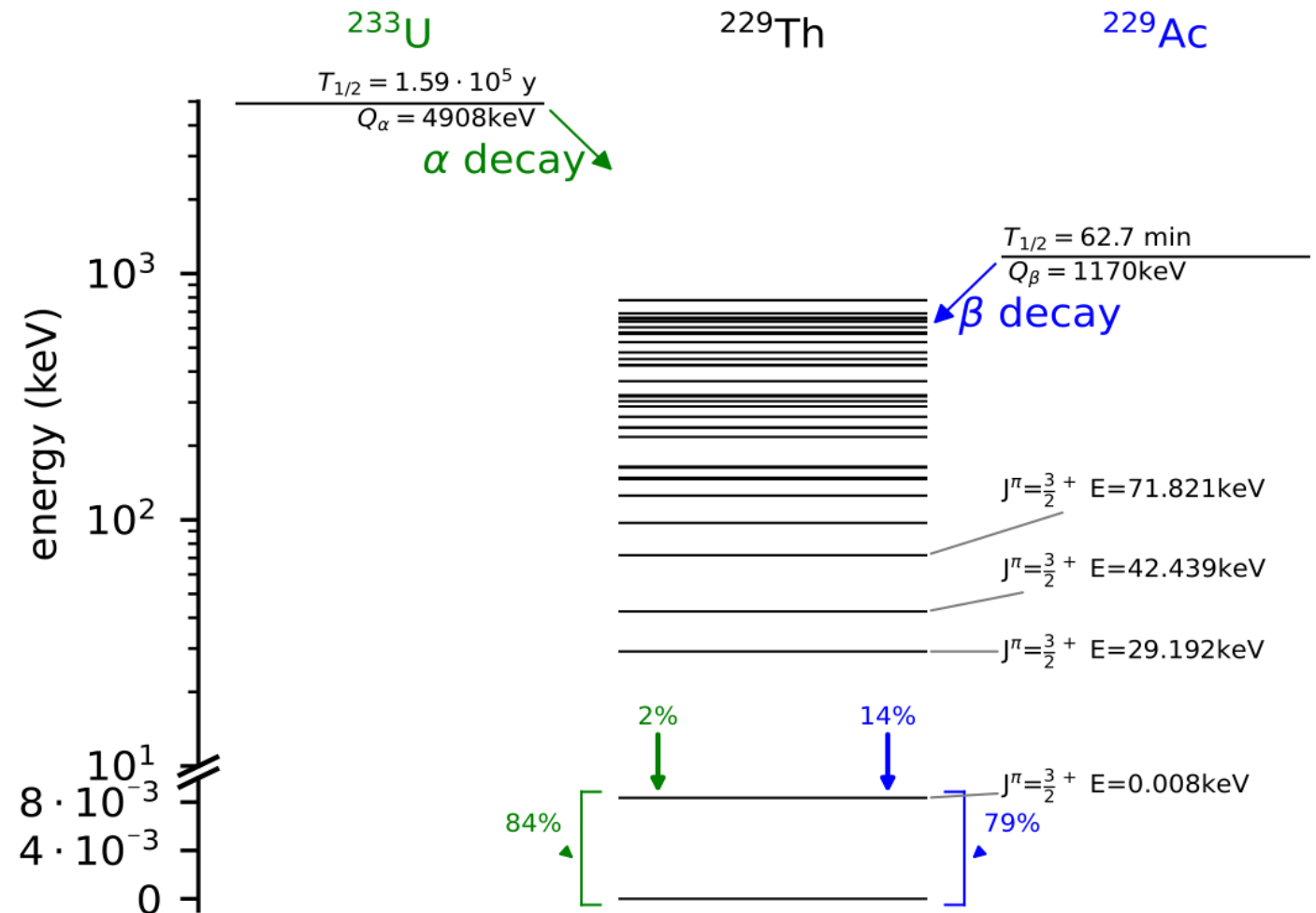
Populating the isomer



What about the radiative decay?

Populating the isomer

229U	230U	231U	232U	233U	234U
228Pa	229Pa	230Pa	231Pa	232Pa	233Pa
227Th	228Th	229Th	230Th	231Th	232Th
226Ac	227Ac	228Ac	229Ac	230Ac	231Ac
225Ra	226Ra	227Ra	228Ra	229Ra	230Ra

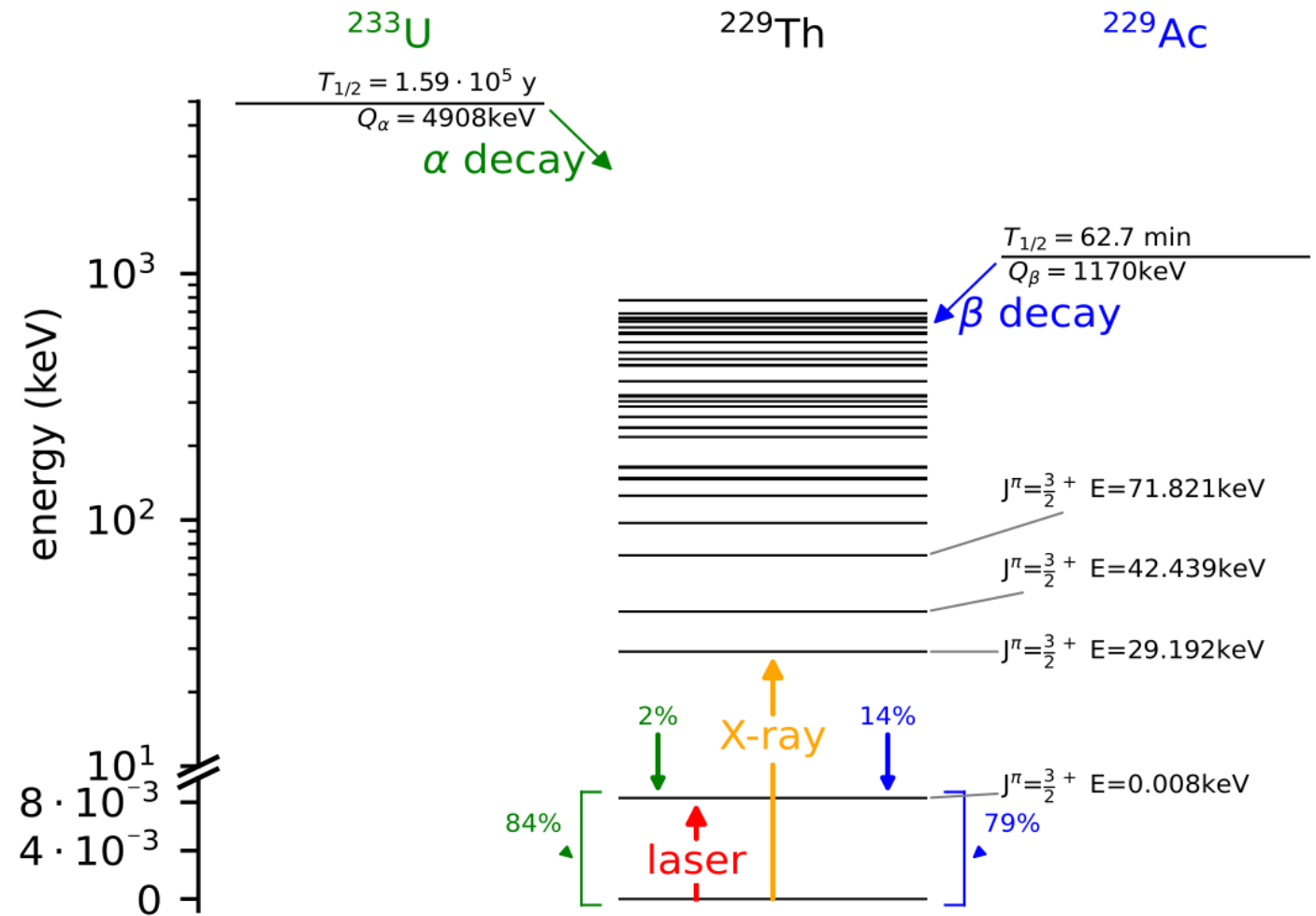


What about the radiative decay?

→ ^{229}Ac β -decay + solid-state host⁽¹⁰⁾

Populating the isomer

229U	230U	231U	232U	233U	234U
228Pa	229Pa	230Pa	231Pa	232Pa	233Pa
227Th	228Th	229Th	230Th	231Th	232Th
226Ac	227Ac	228Ac	229Ac	230Ac	231Ac
225Ra	226Ra	227Ra	228Ra	229Ra	230Ra



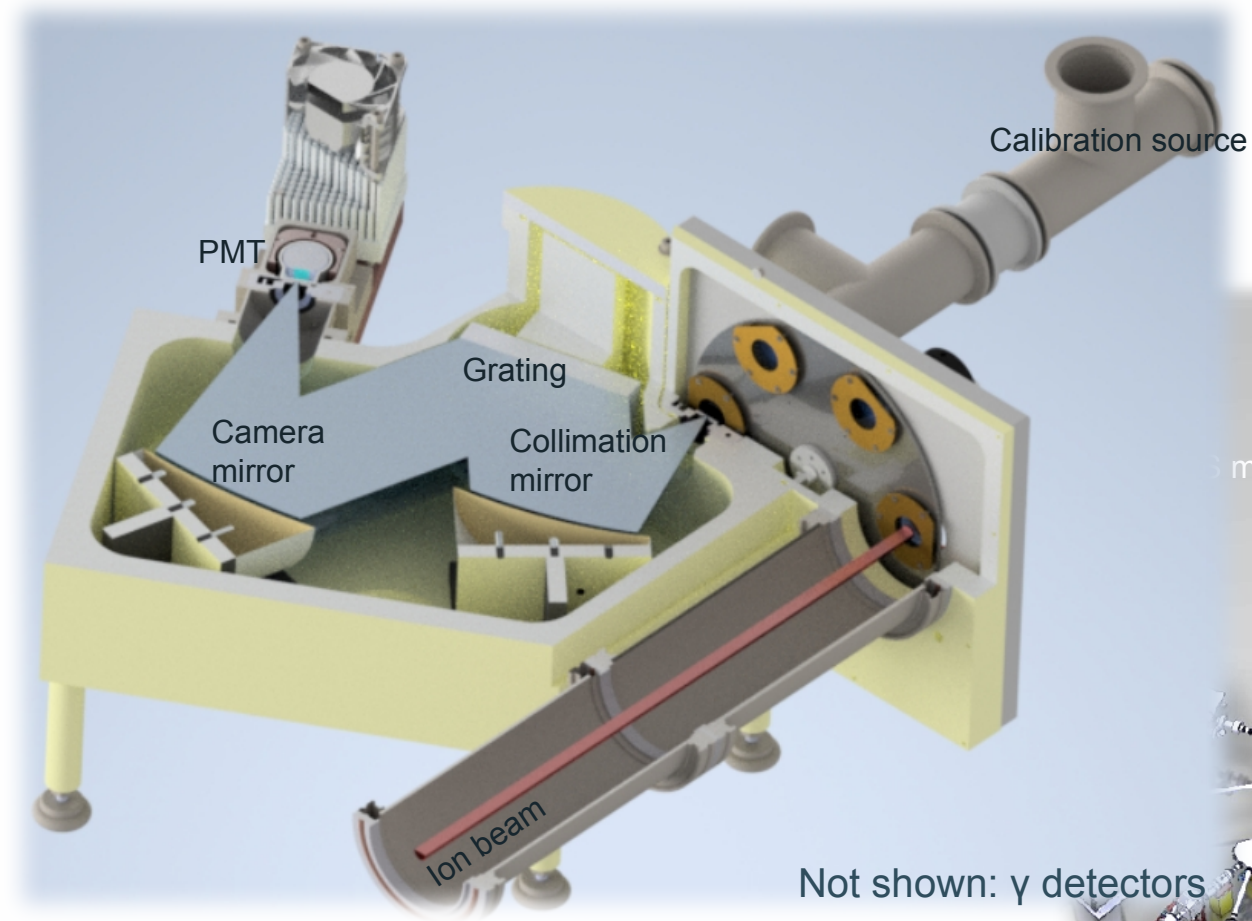
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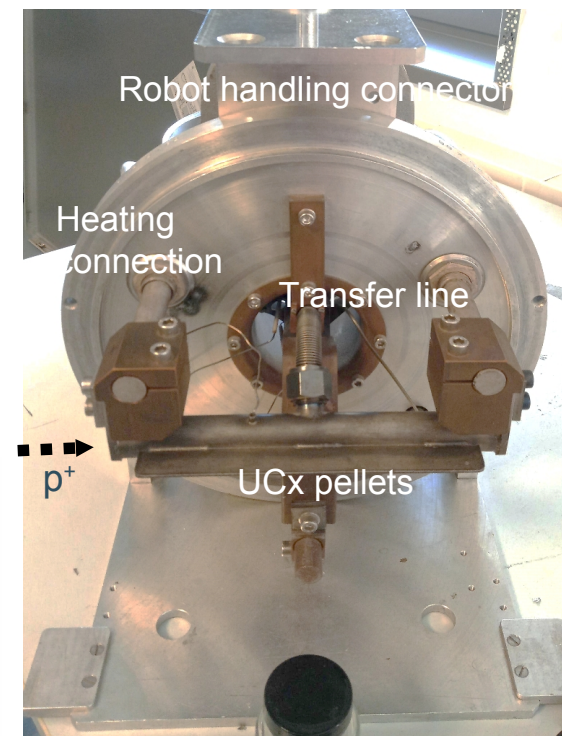
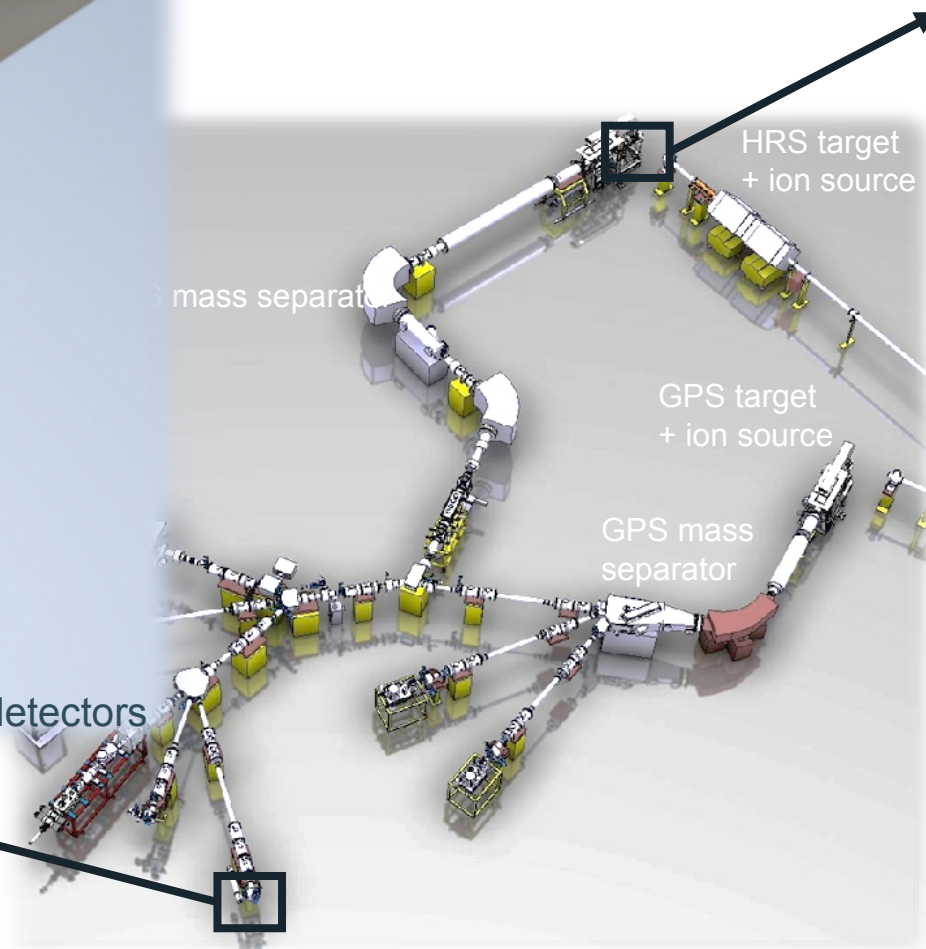
Vacuum-ultraviolet spectroscopy at ISOLDE



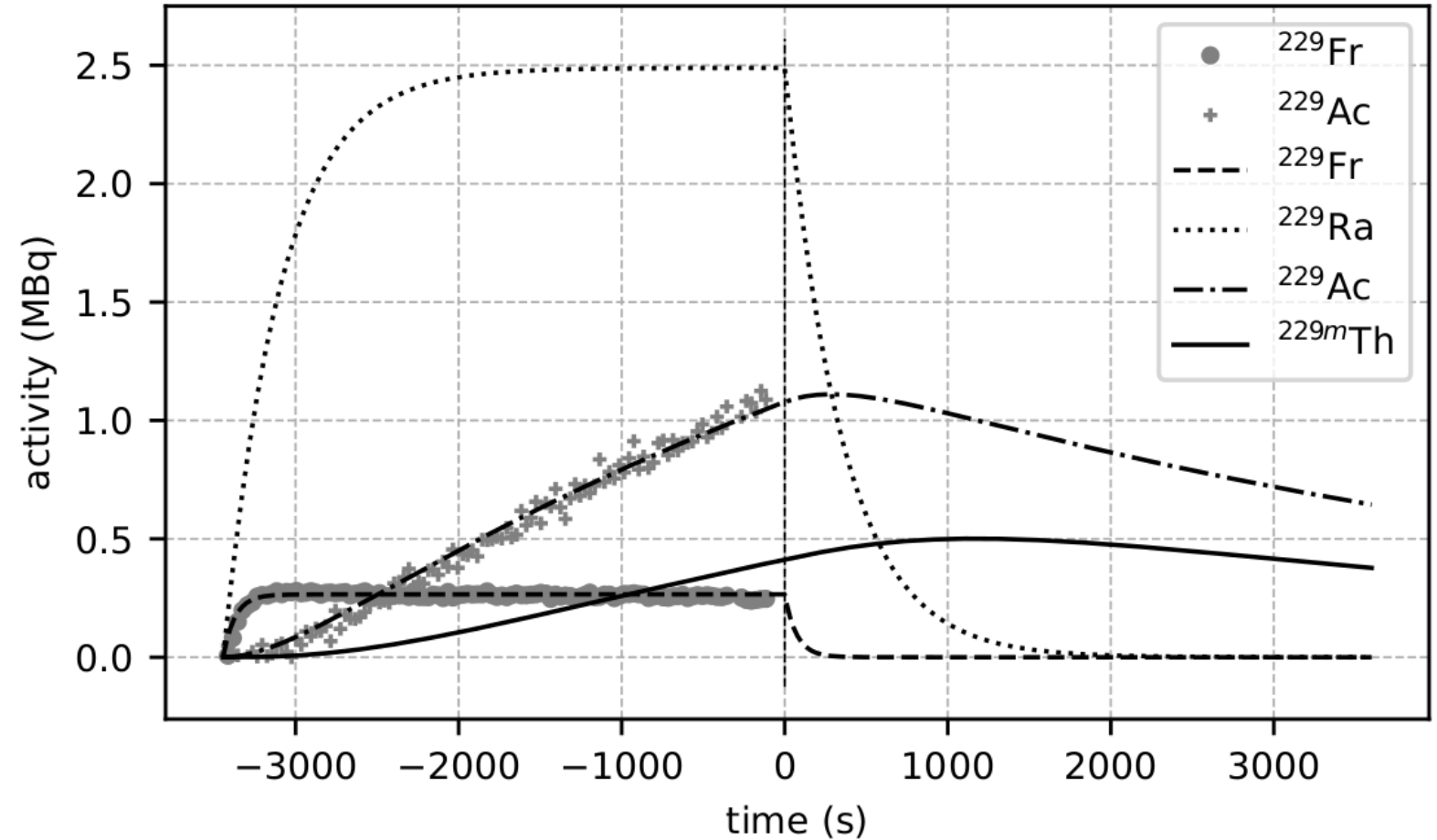
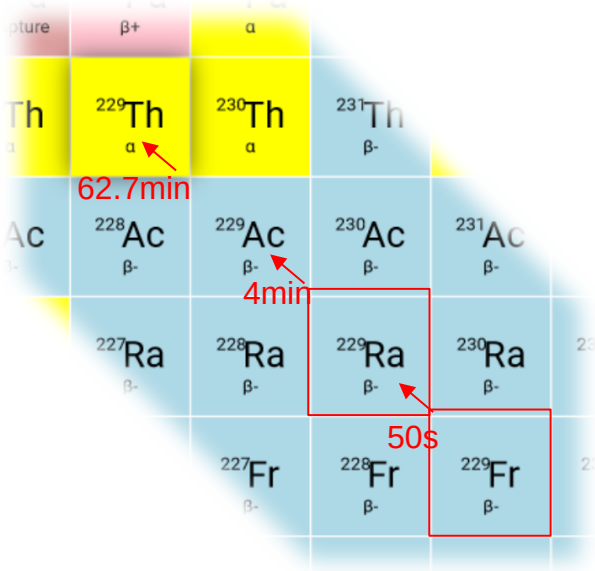
Vacuum-ultraviolet spectroscopy at ISOLDE



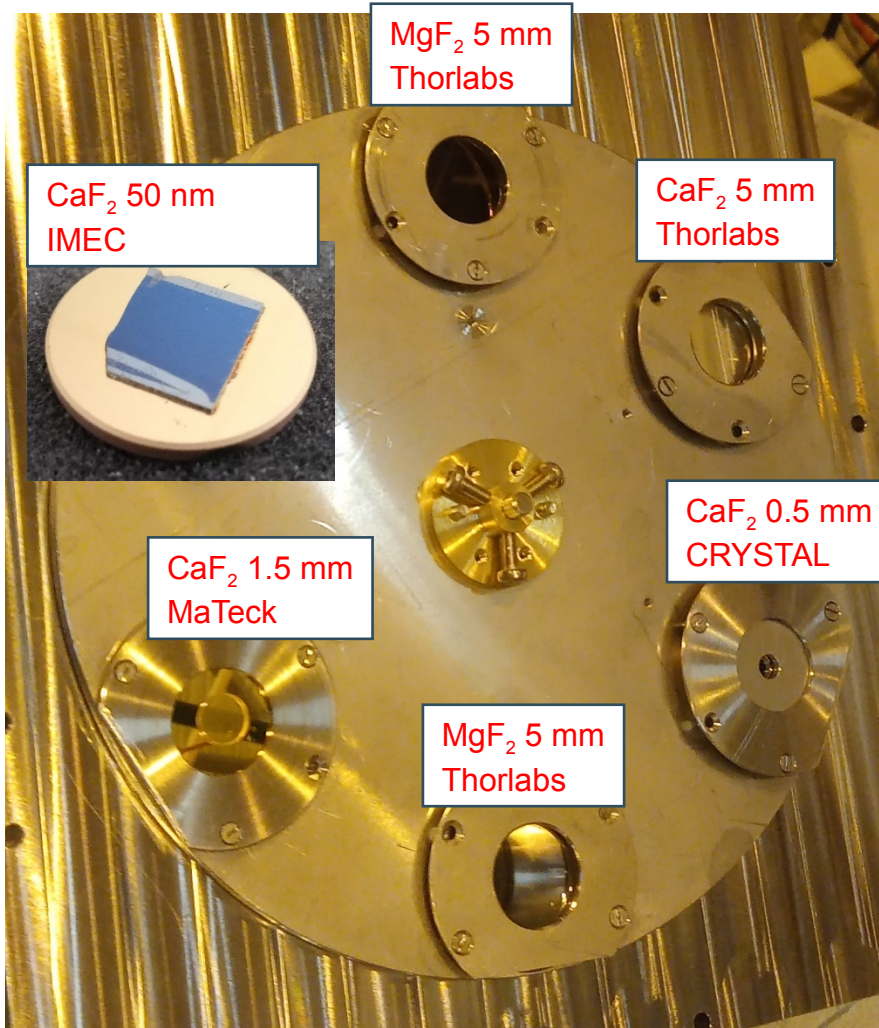
Not shown: γ detectors



Vacuum-ultraviolet spectroscopy at ISOLDE



Vacuum-ultraviolet spectroscopy at ISOLDE

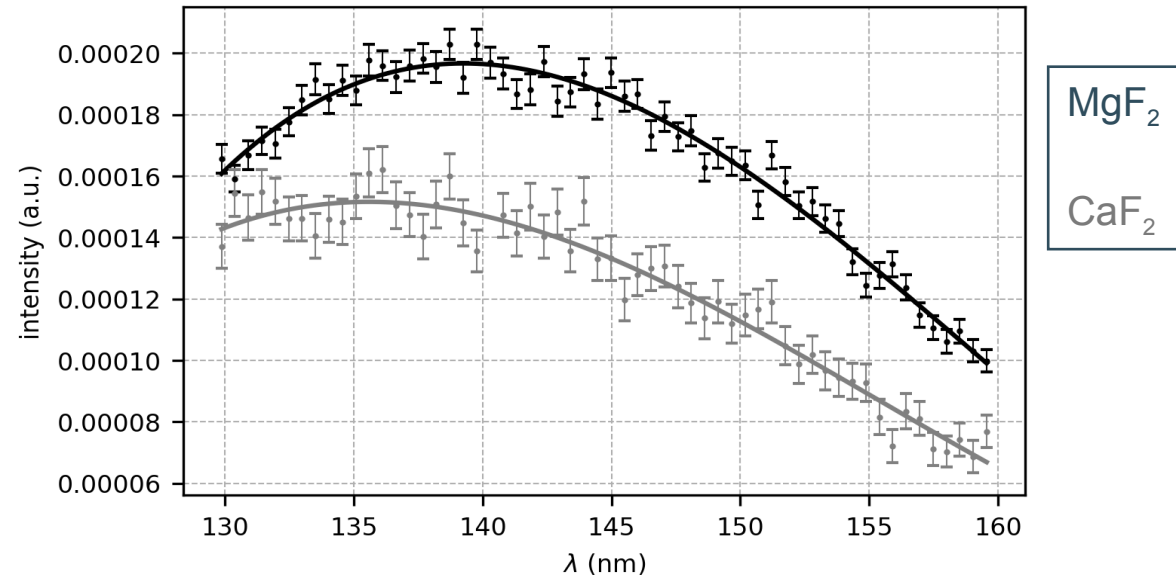


Background induced by radiation:

Cherenkov photon emission
→ *Continuous spectrum*

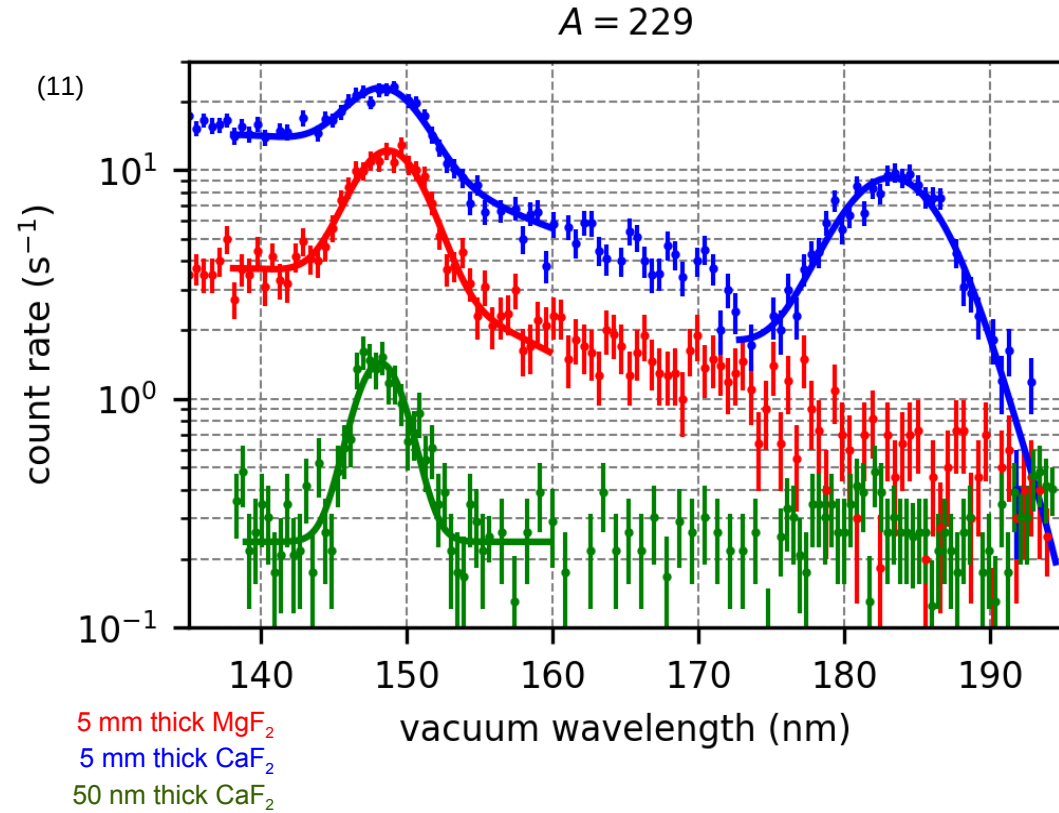
Observed spectral shape:

Cherenkov spectrum + instruments and crystal bandpass

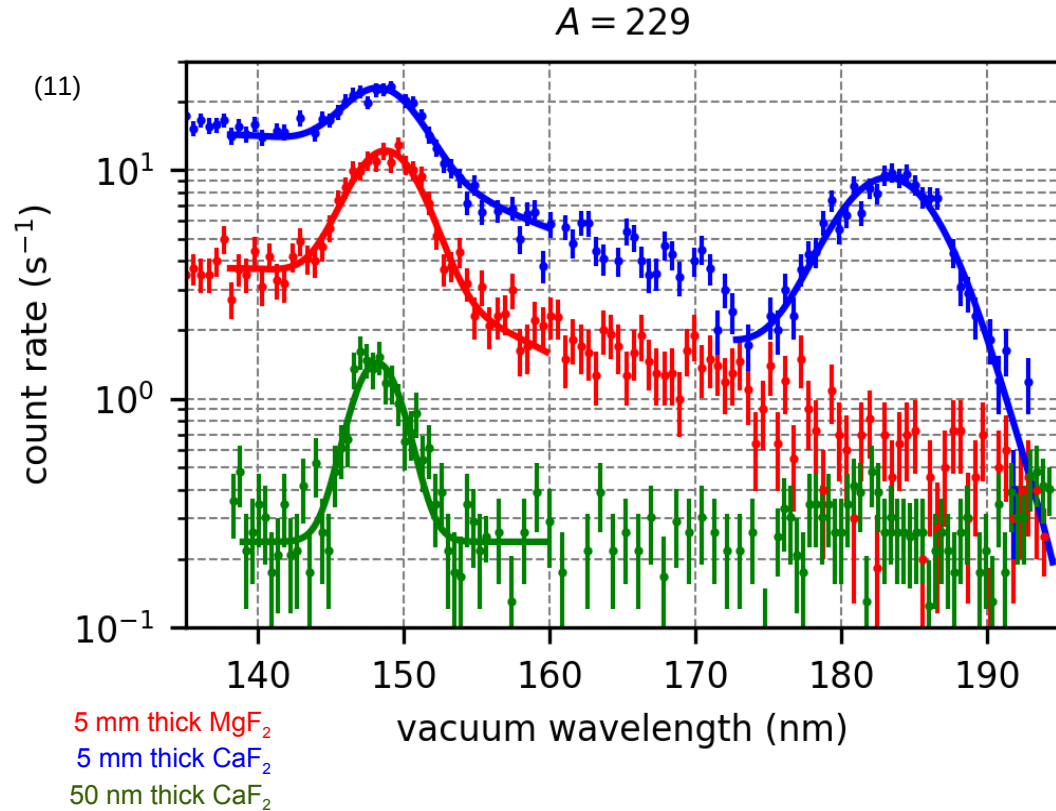


Radioluminescence of crystal defects
→ *Limited spectral width*

Identification

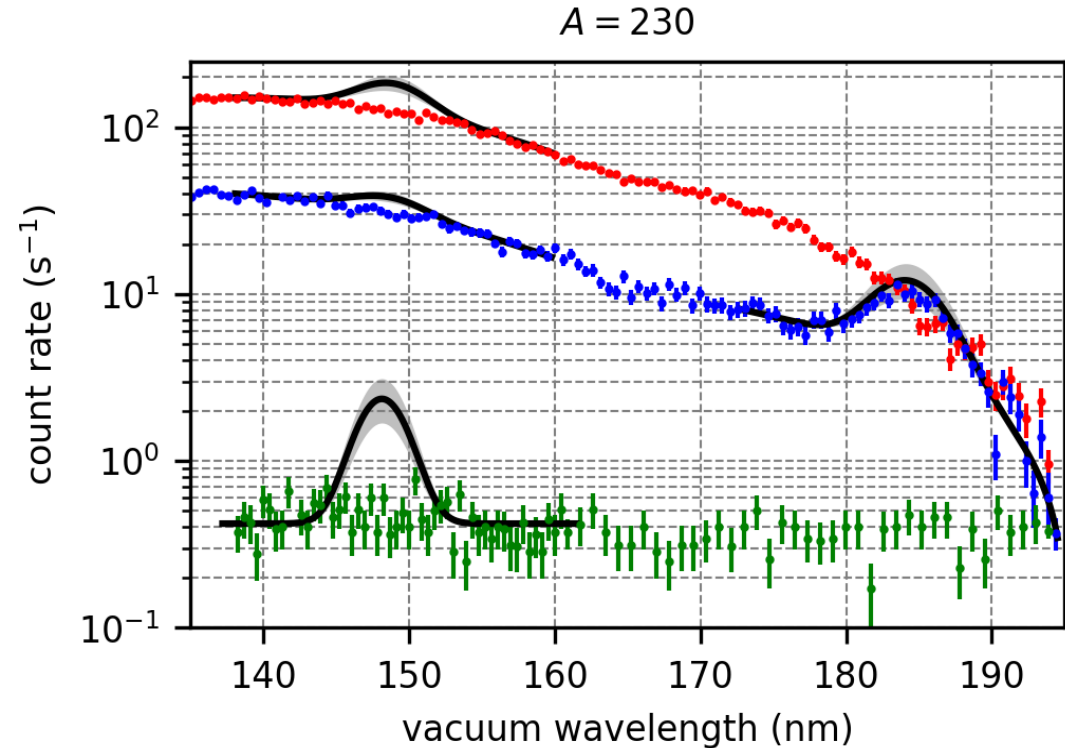


Identification



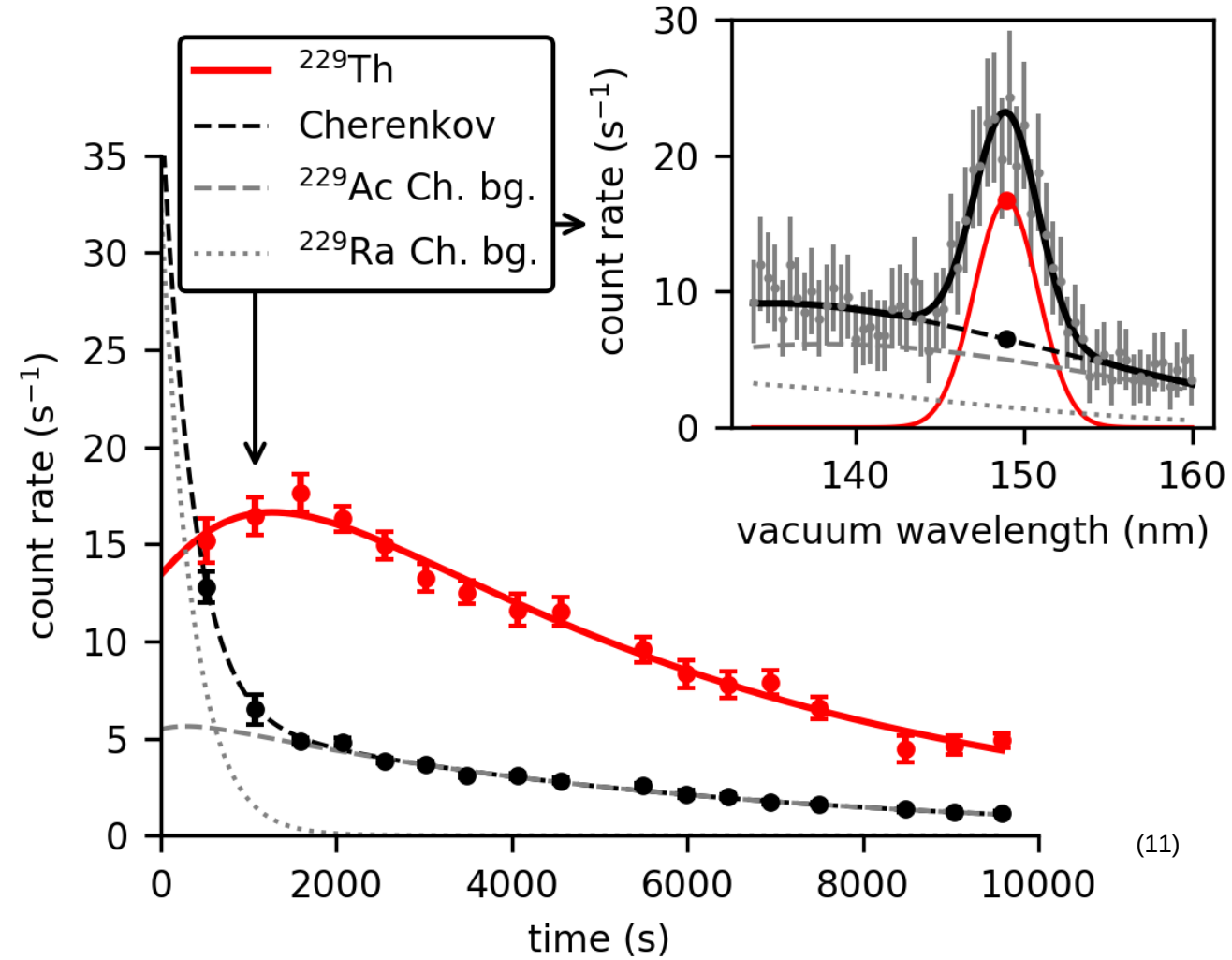
↑
 ^{229m}Th radiative decay

↑
 CaF_2 defect radioluminescence



↑
 CaF_2 defect radioluminescence

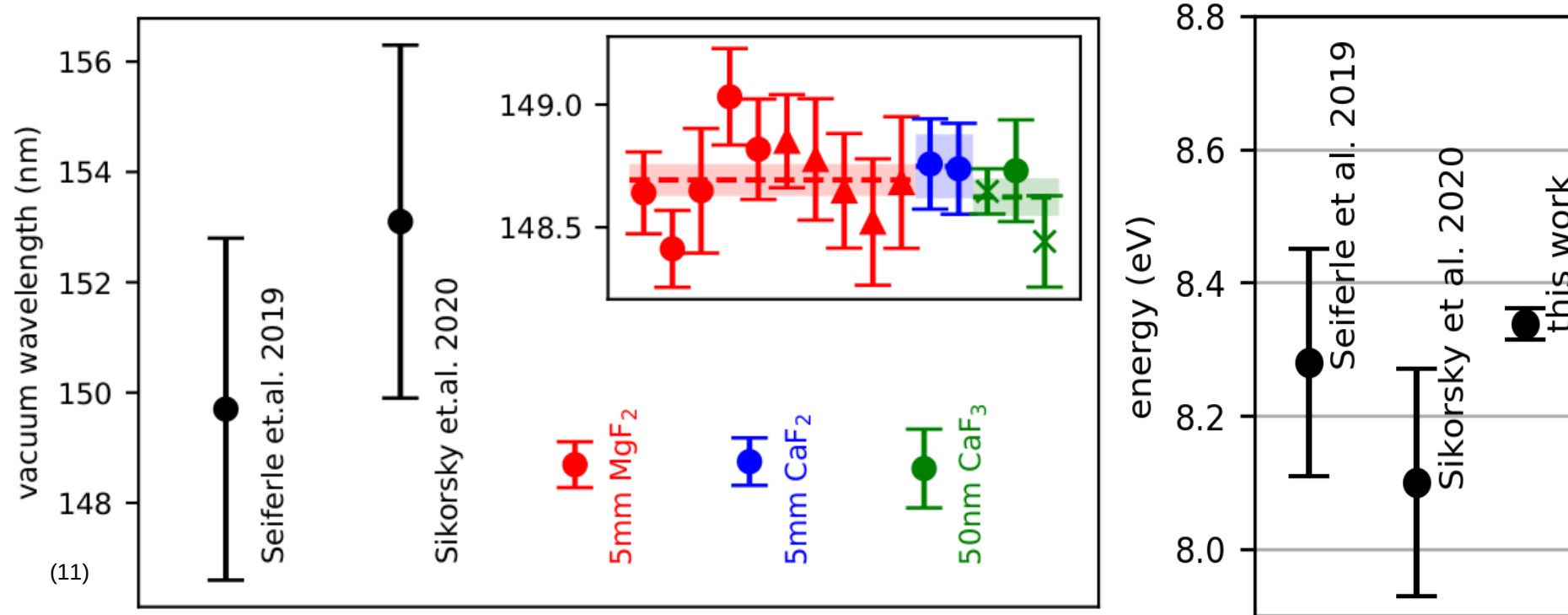
Half-life



670(102) s in MgF_2

Crystal: 5 mm MgF_2
Entrance slit: 2 mm

Excitation energy



New energy value:
8.338(24) eV

→ Uncertainty reduced from 41 THz to 5.8 THz

Conclusion and Outlook



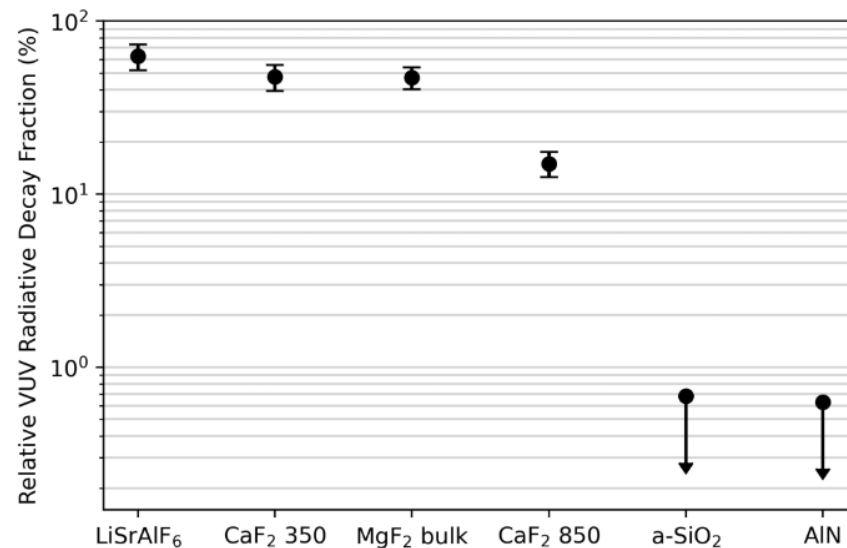
Conclusion

This work⁽¹¹⁾:

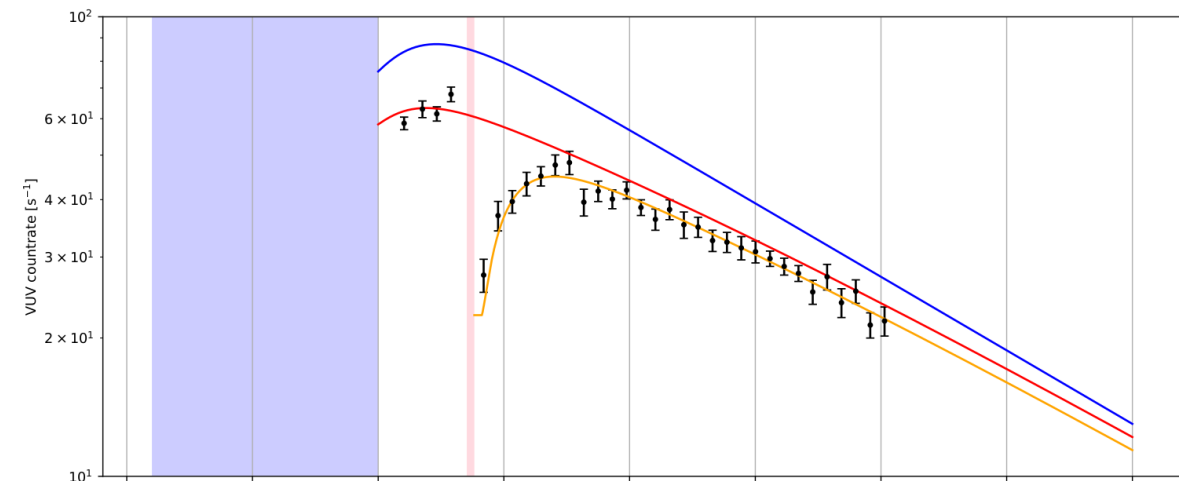
- Demonstration of the feasibility of the solid-state approach
- New energy value to facilitate laser excitation
- Establishing a robust system for material testing using implanted radioactive ion beams

New measurement campaigns^(12,13)

- Testing of the radiative decay fraction in candidate materials
- Better understanding of quenching effects



Radioactive quenching (c.f. contribution by Y.Elskens)

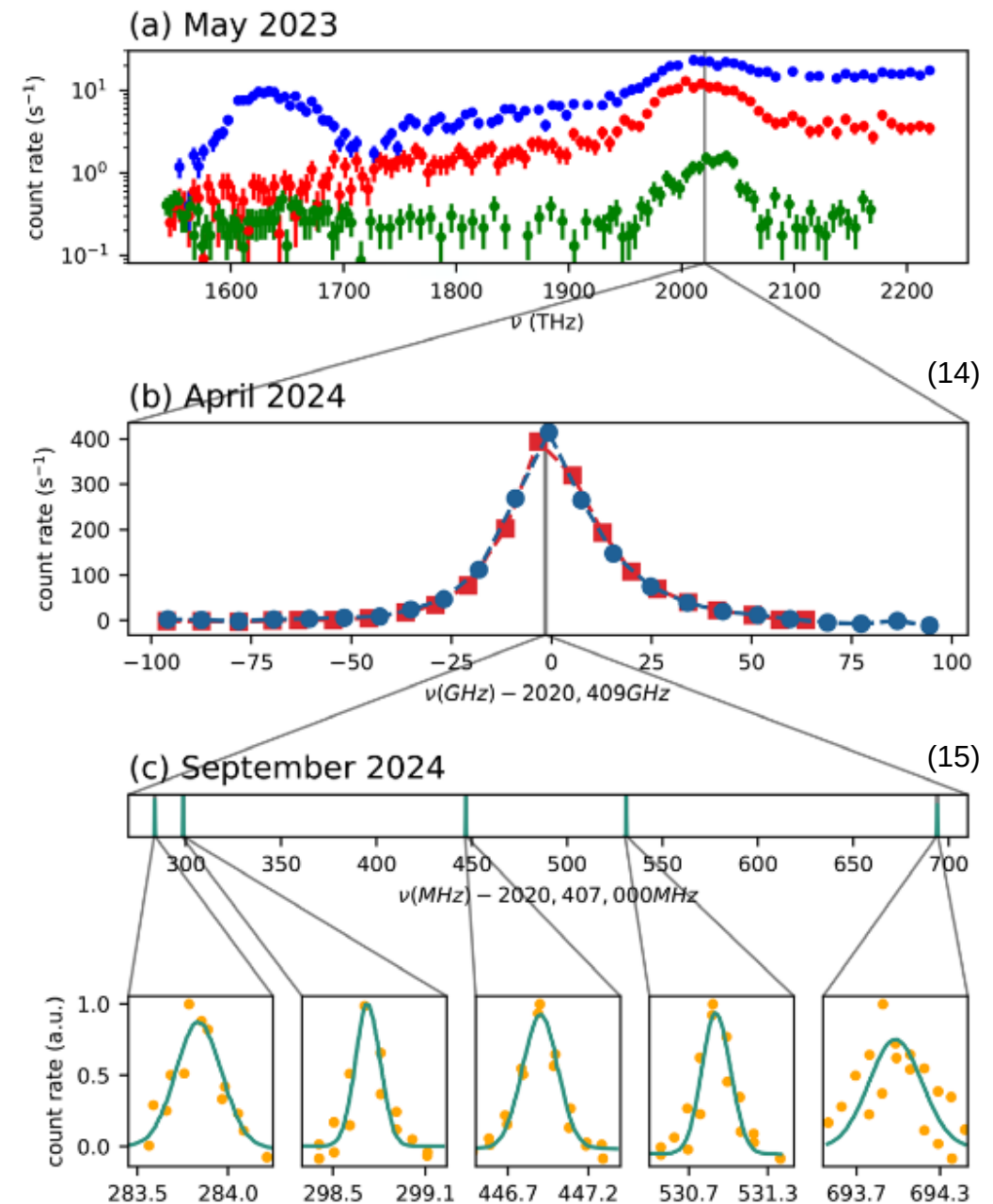
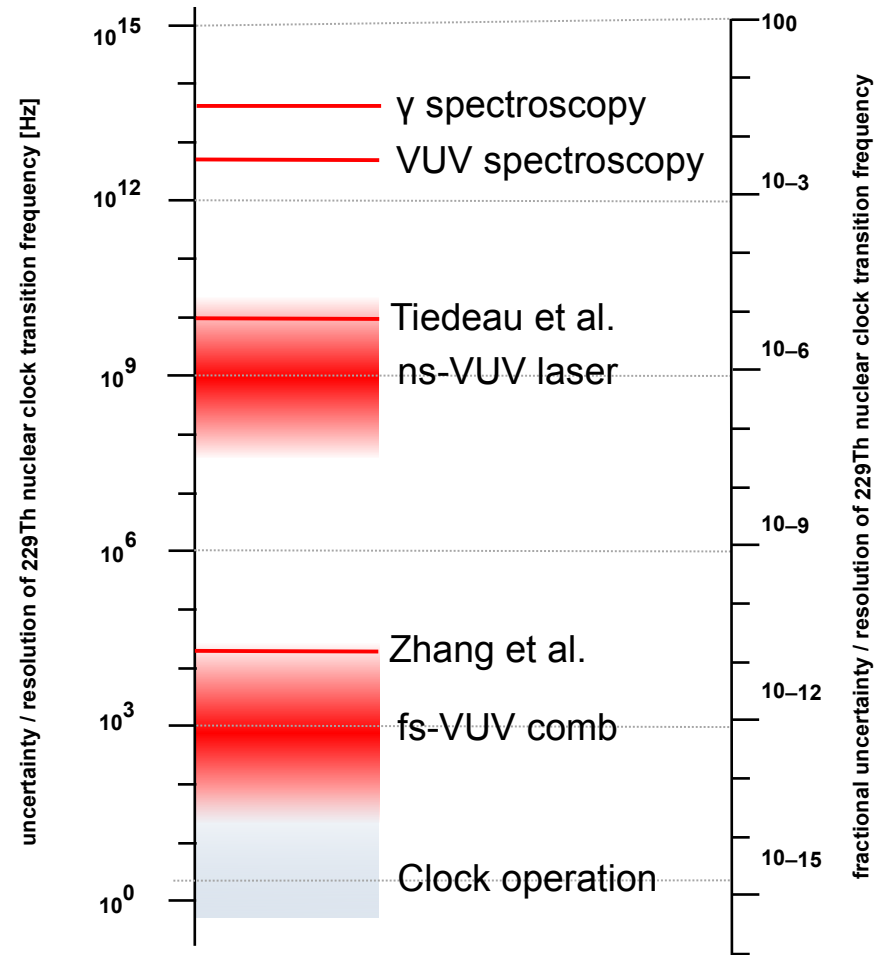


(11) Kraemer et al., *Nature* **617** 7962

(12) Pineda, S. V. et al. *Phys. Rev. Res.* **7**, 013052 (2025).

(13) Y.Elskens et al., in preparation

Outlook:Recent developments



(14) Tiedau et al. *Phys. Rev. Lett.* **132**, 182501 (2024).

(15) Zhang, C. et al. *Nature* **633**, 63–70 (2024).

Thank you very much!

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



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