

Towards a nuclear clock: half-life and decay-fraction measurements of the radiative decay of ^{229m}Th

ISOLDE workshop and users meeting 2024
Yens Killian Elskens



Context

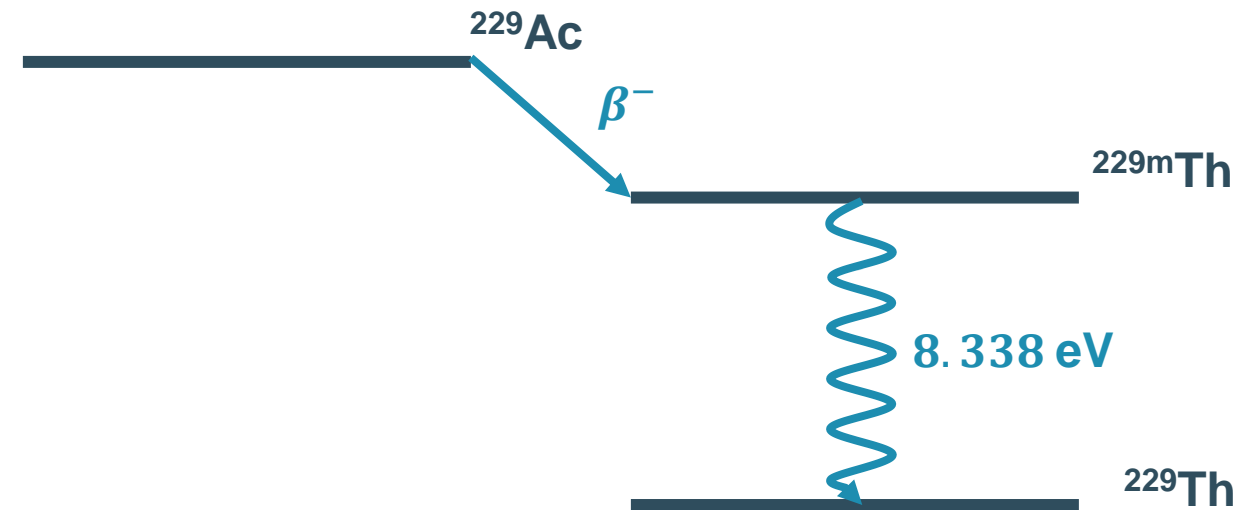
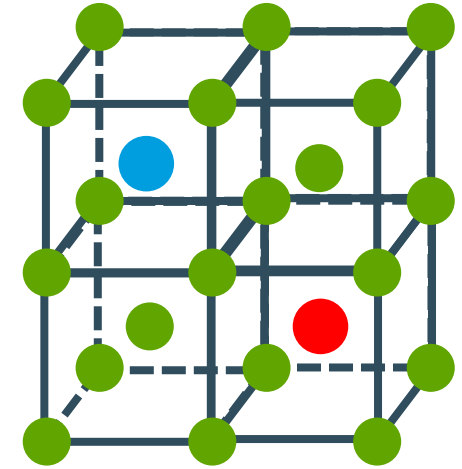
^{229}Th has an isomer that lies low enough to probe with a laser

Ideal two-level system for a **nuclear clock**

Challenge: dominating IC decay

Populating the isomer through the **β -decay of ^{229}Ac** within the context of a **large-bandgap crystal**

Results from 2021 ISOLDE beam time led to laser excitation in CaF_2 (PTB) and LiSrAlF_6 (UCLA), and even excitation with a frequency comb (JILA)



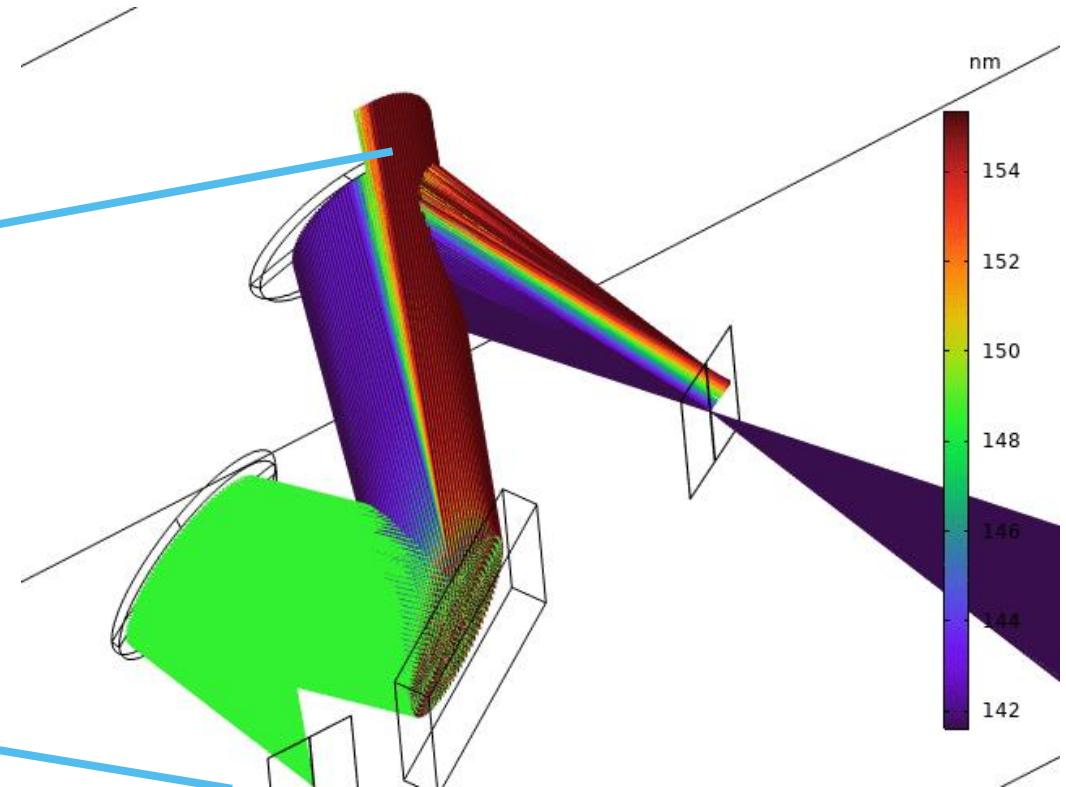
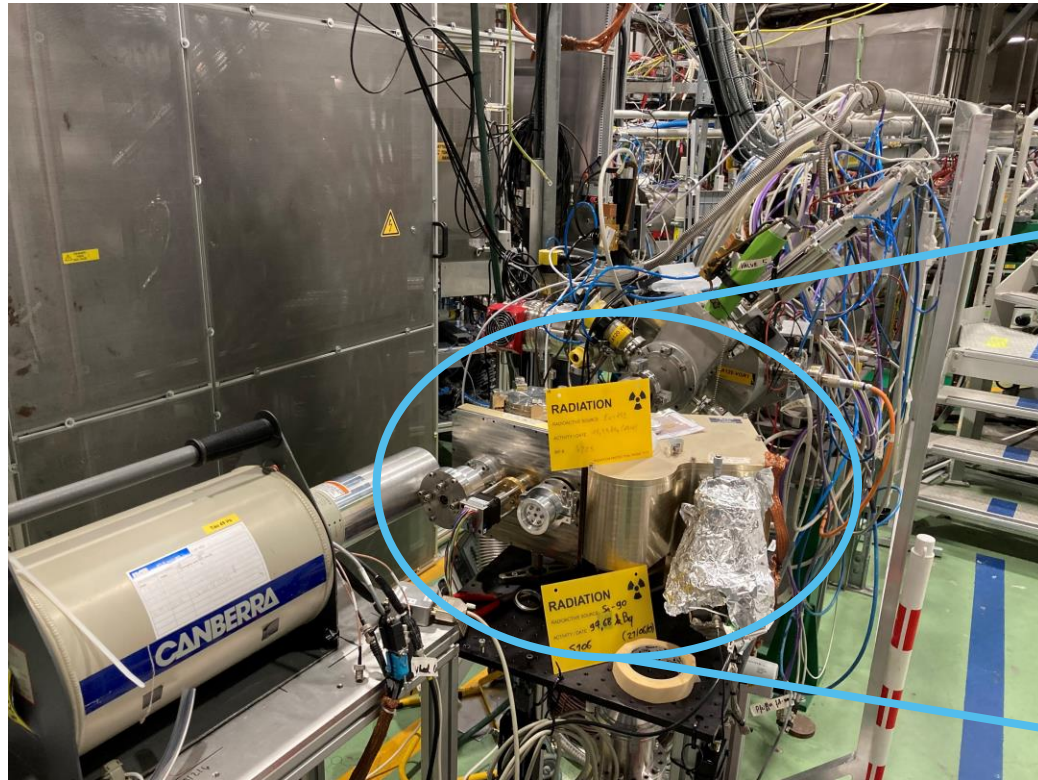
[1] S. Kraemer et al. *Observation of the radiative decay of the ^{229}Th nuclear clock isomer*, 2024

[2] J. Tiedau et al. *Laser excitation of the ^{229}Th nucleus*, 2024

[3] R. Elwell et al. *Laser excitation of the ^{229}Th nuclear isomeric transition in a solid-state host*, 2024

[4] C. Zhang et al. *Frequency ratio of the $^{229\text{m}}\text{Th}$ nuclear isomeric transition and the ^{87}Sr atomic clock*, 2024

On-line VUV spectroscopy of the isomer at LA1



2023 campaign goals



Study the **effect** of the implanted **crystal** on the **radiative decay fraction**



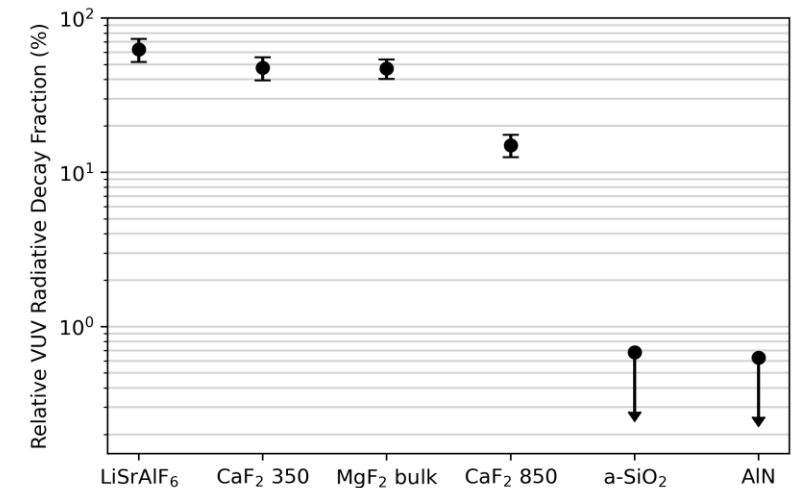
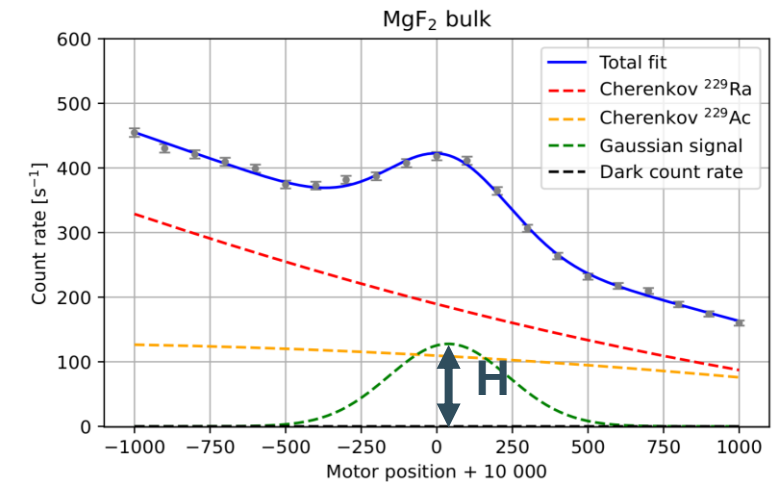
Study the **time behaviour** of the radiative decay in different crystalline environments

Decay fractions in different crystals

$$\epsilon_{\text{VUV}} = \frac{H}{A_{\text{iso}} \epsilon_{\text{I}}}$$

Compare the radiative decay fraction of different crystals relative to CaF_2 bulk (highest absolute efficiency)

Determine limits for AlN and SiO_2



Studying the time behaviour of the VUV signal

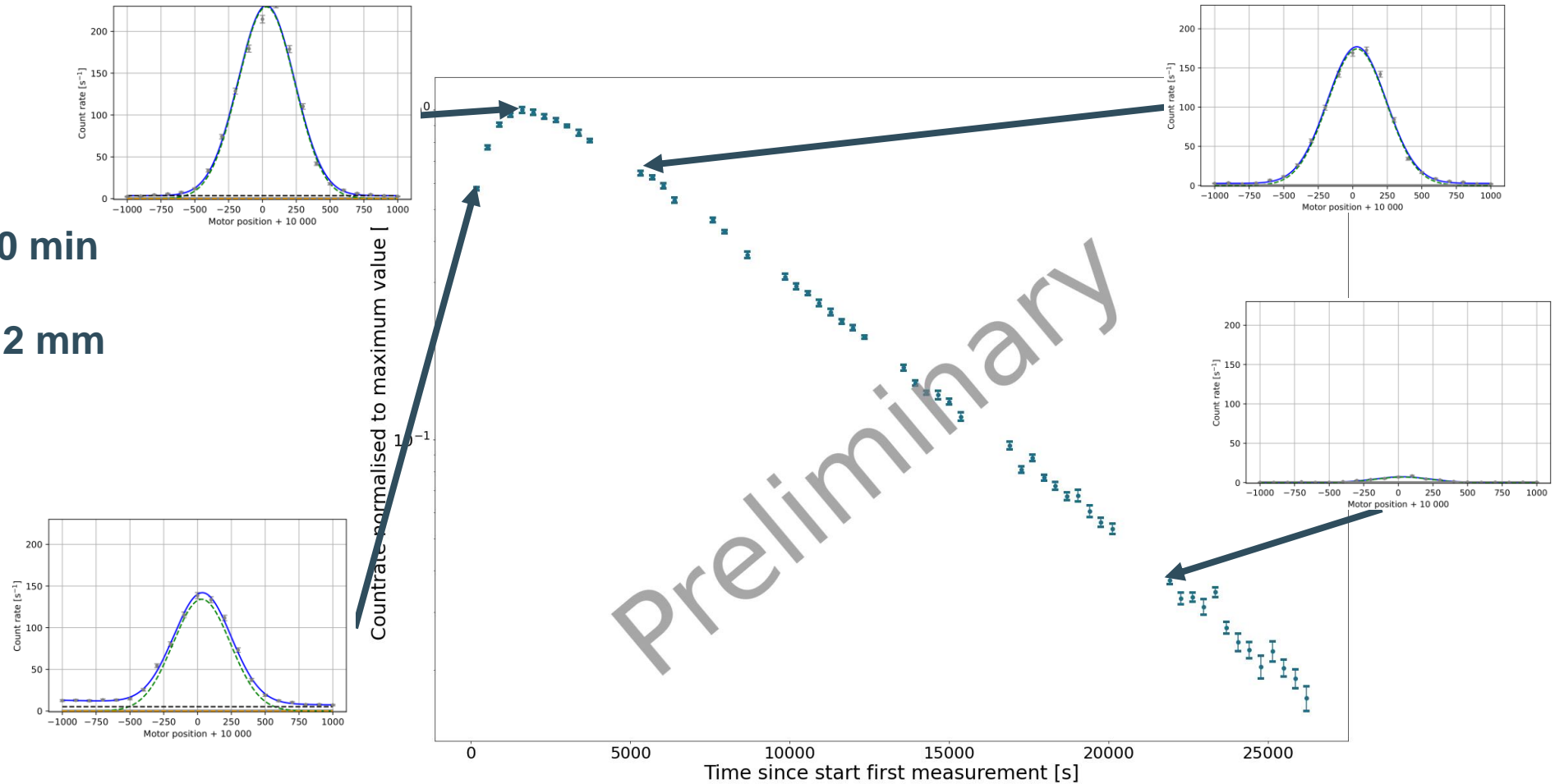
General method:

Implant crystal for **15-30 min**

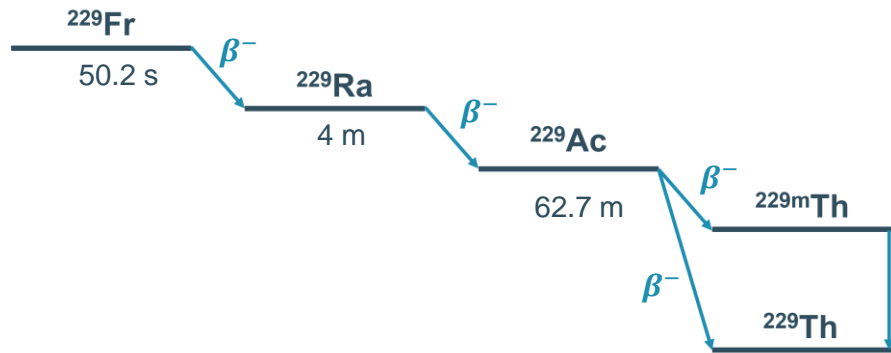
Move crystal in front of **2 mm entrance slit**

Repeatedly scan ROI

Determine height as a function of time

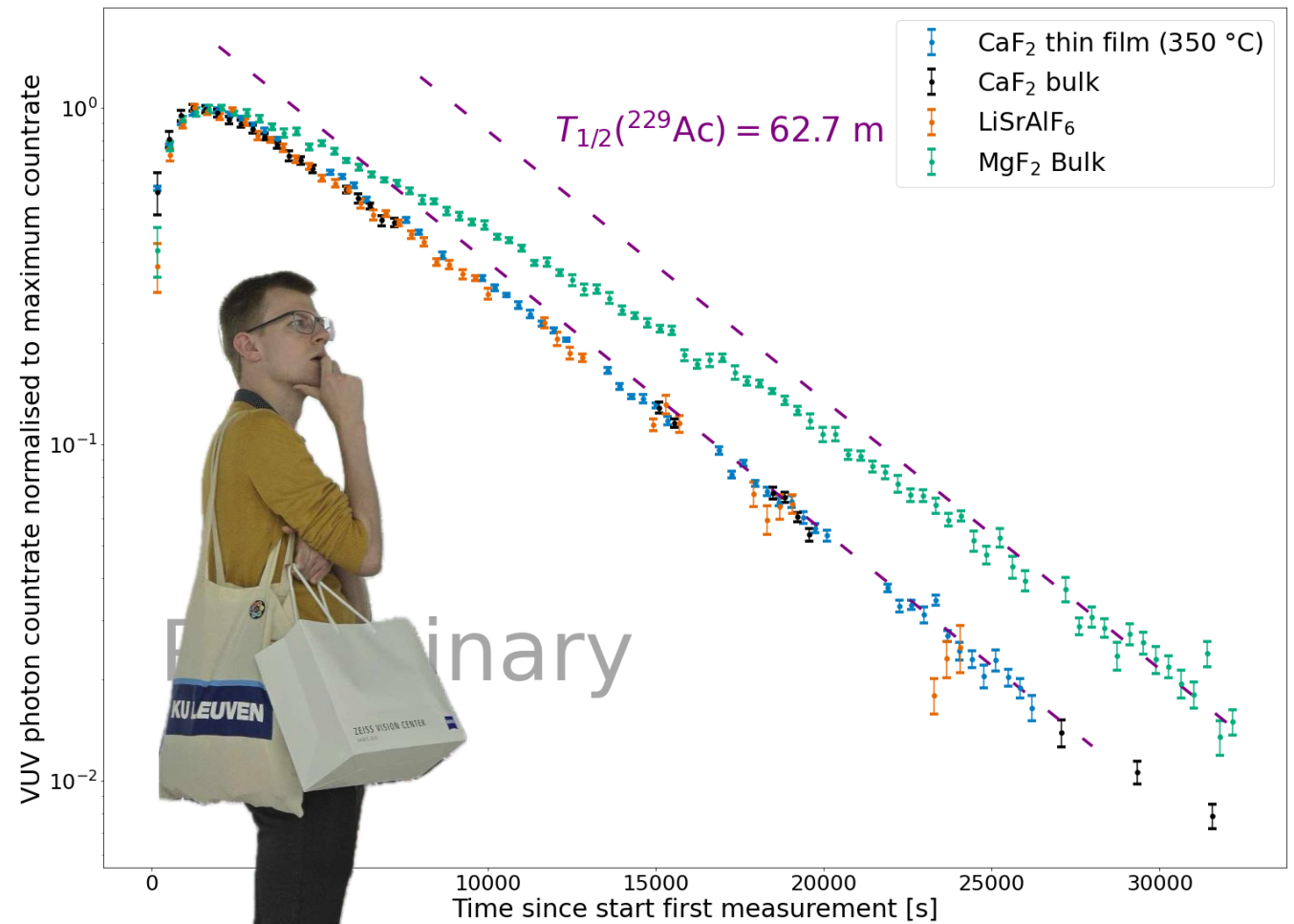


Studying the isomer's time behaviour



The isomer's radiative decay does not reach transient equilibrium with ^{229}Ac when expected

When it reaches equilibrium depends on the crystal



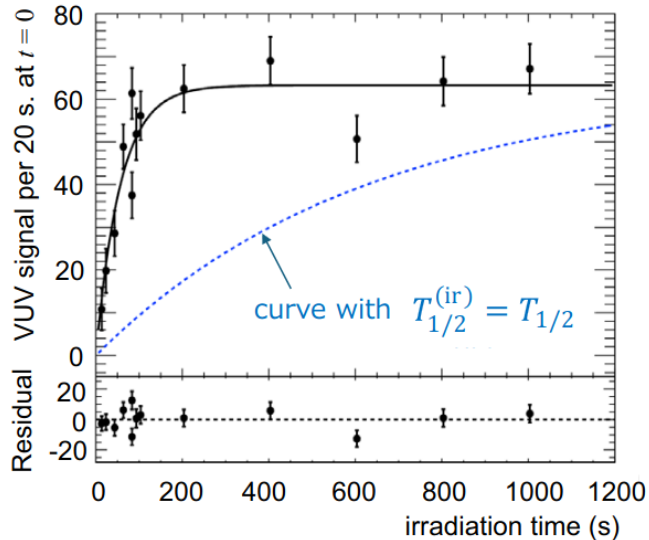
'Quenching' of the halflife

VUV spectroscopy at Spring-8 by X-ray pumping the isomer

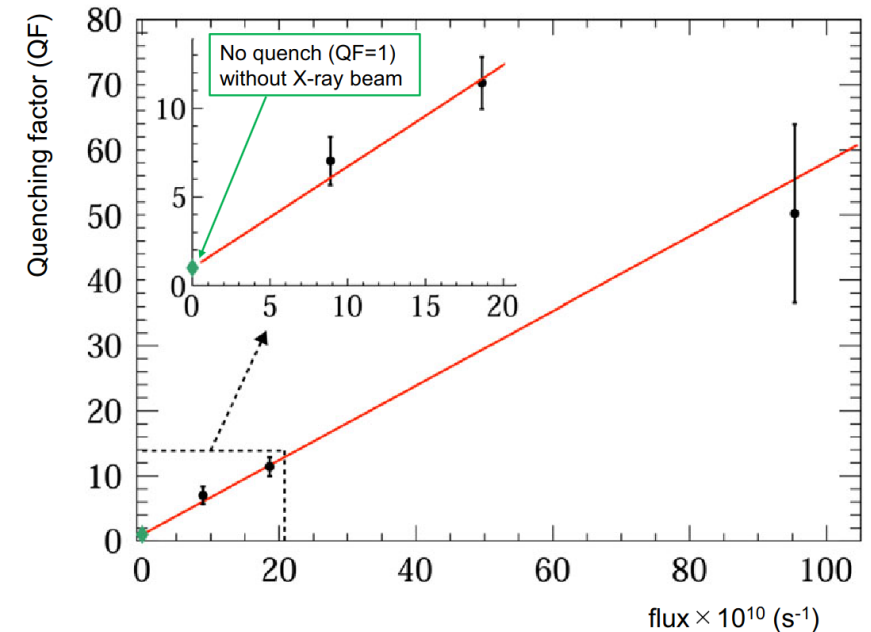
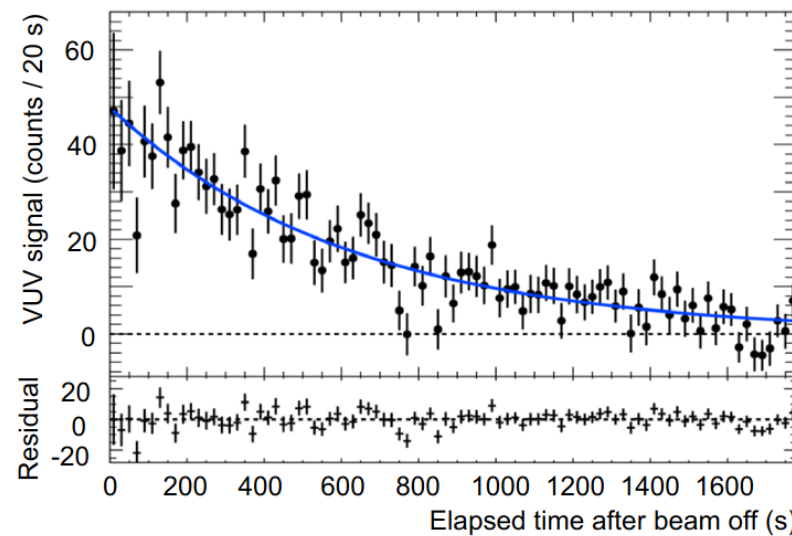
Flux-dependent 'quenching' of the observed halflife

$$Q = \frac{t_{1/2}}{t_{1/2}^{(irr)}}$$

(a) isomer production



(b) isomer decay



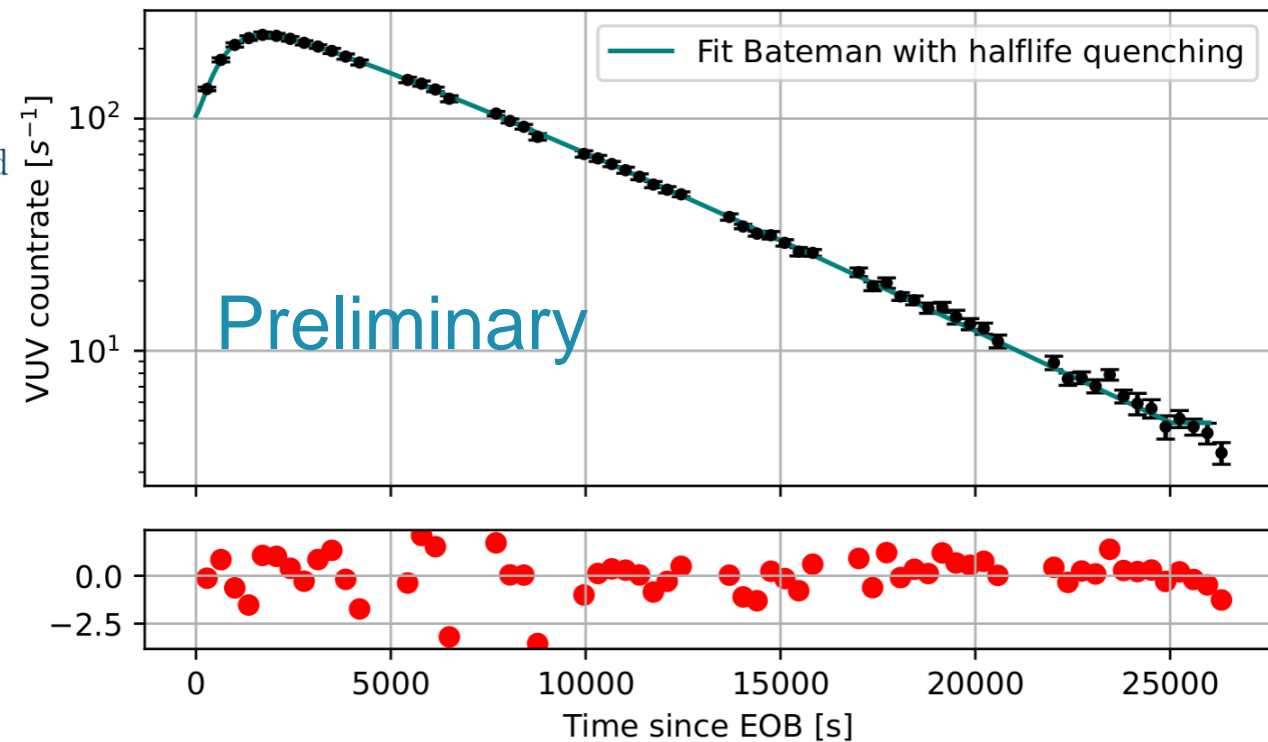
Quenching of the half-life: CaF₂ thin film

Make quenching depend linearly on activities of Ra and Ac

$$\frac{dN_{\text{rad}}}{dt} = \lambda_{\text{Ac}}N_{\text{Ac}} - \underbrace{(1 + \alpha\lambda_{\text{Ra}}N_{\text{Ra}} + \beta\lambda_{\text{Ac}}N_{\text{Ac}})}_{Q^{-1}} \lambda_{\text{rad}}N_{\text{rad}}$$

Half-life of 488 ± 48 (stat.) s (very preliminary) seems to correspond with results from PTB (436 ± 10 s), Spring-8 (447 ± 25 s) and JILA (444 ± 3 s)

Doesn't describe MgF₂ well. Probably needs a 'population quenching' on top of the half-life quenching.



Conclusions and outlook



Determined relative radiative decay fractions in different crystals

CaF_2 , MgF_2 , LiSrAlF_6 , AlN , SiO_2



Environmental-dependent 'quenching' mechanism observed.

Describes time behaviour in CaF_2 , not in MgF_2

Beam time 2025

More crystals:
 LiCaAlF_6 , SrF_2 ,
 ThF_4 , Al_2O_3 , BaLiF
...

Understanding
'quenching':
 Φ - and T-
dependence

Acknowledgements

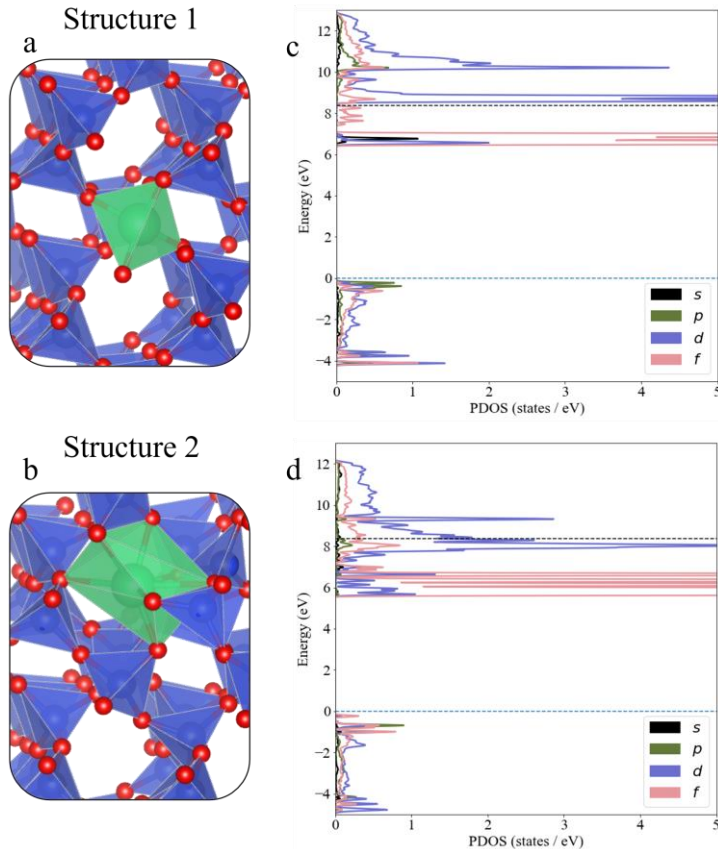
The IS715 collaboration

M. Athanasakis, M. Au, S. Bara, M. Bartokos, K. Beeks, S. Casci, P. Chhetri, K. Chrysalidis, A. Claessens, J. G. Correia, Y. Elskens, R. Ferrer, R. Heinke, E. Hudson, F. Ivandikov, U. Köster, S. Kraemer, M. Laatiaoui, R. Lica, G. Magchiels, J. Moens, H. Morgan, D. Moritz, I. Morawetz, L. Pereira, S. V. Pineda, S. Raeder, S. Rother, A. de Roubin, F. Schaden, K. Scharl, T. Schumm, S. Stegeman, P. Thirolf, P. Van Duppen, A. Vantomme, R. Villarreal, U. Wahl



Back up

DFT calculations for SiO₂ by UCLA



DFT calculations for crystalline SiO₂ (experiment = amorphous)

2 structures modeled:

1. Th substitutes Si
2. Th and two O's are added as interstitial defect

Empty Th states emerge within the band gap for both structures

Q: Is the 'quenching' actually a dead-time issue?

A: No

