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Essential steps towards a nuclear clock: measurements of the radiative decay of ^{229m}Th in solid-state hosts

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EuNPC 2025

Context

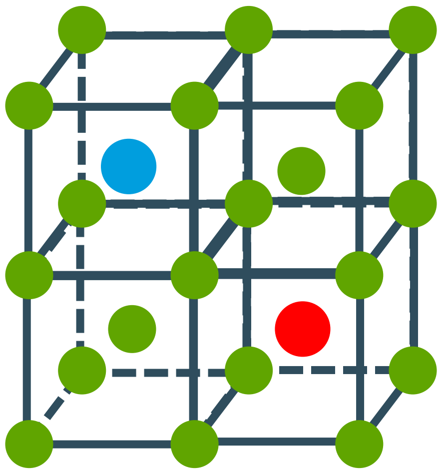
^{229}Th has an isomer that lies low enough to probe with a laser

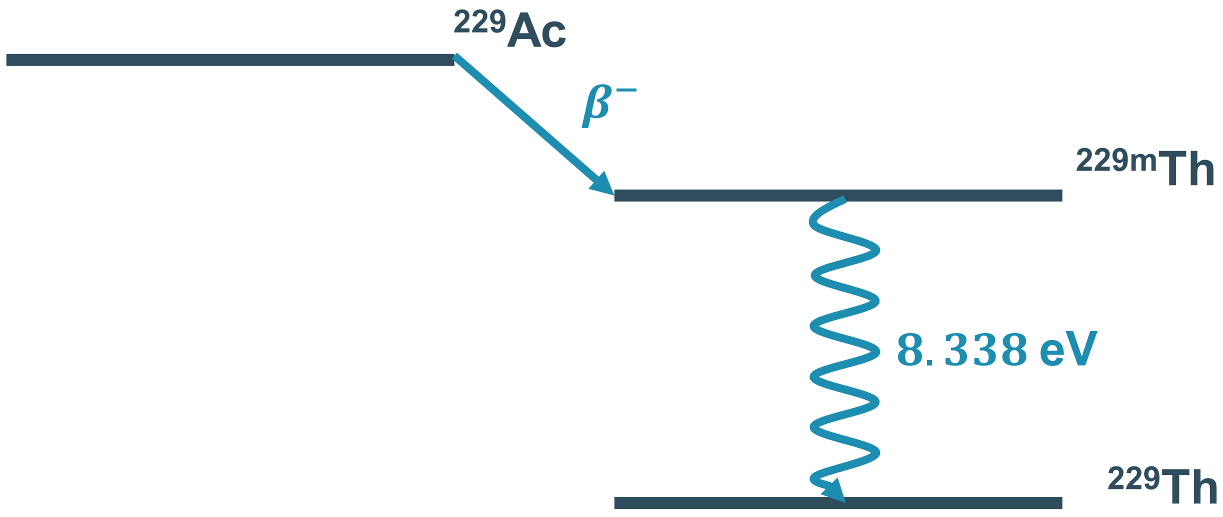
Ideal two-level system for a **nuclear clock**

Challenge: dominating IC decay

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

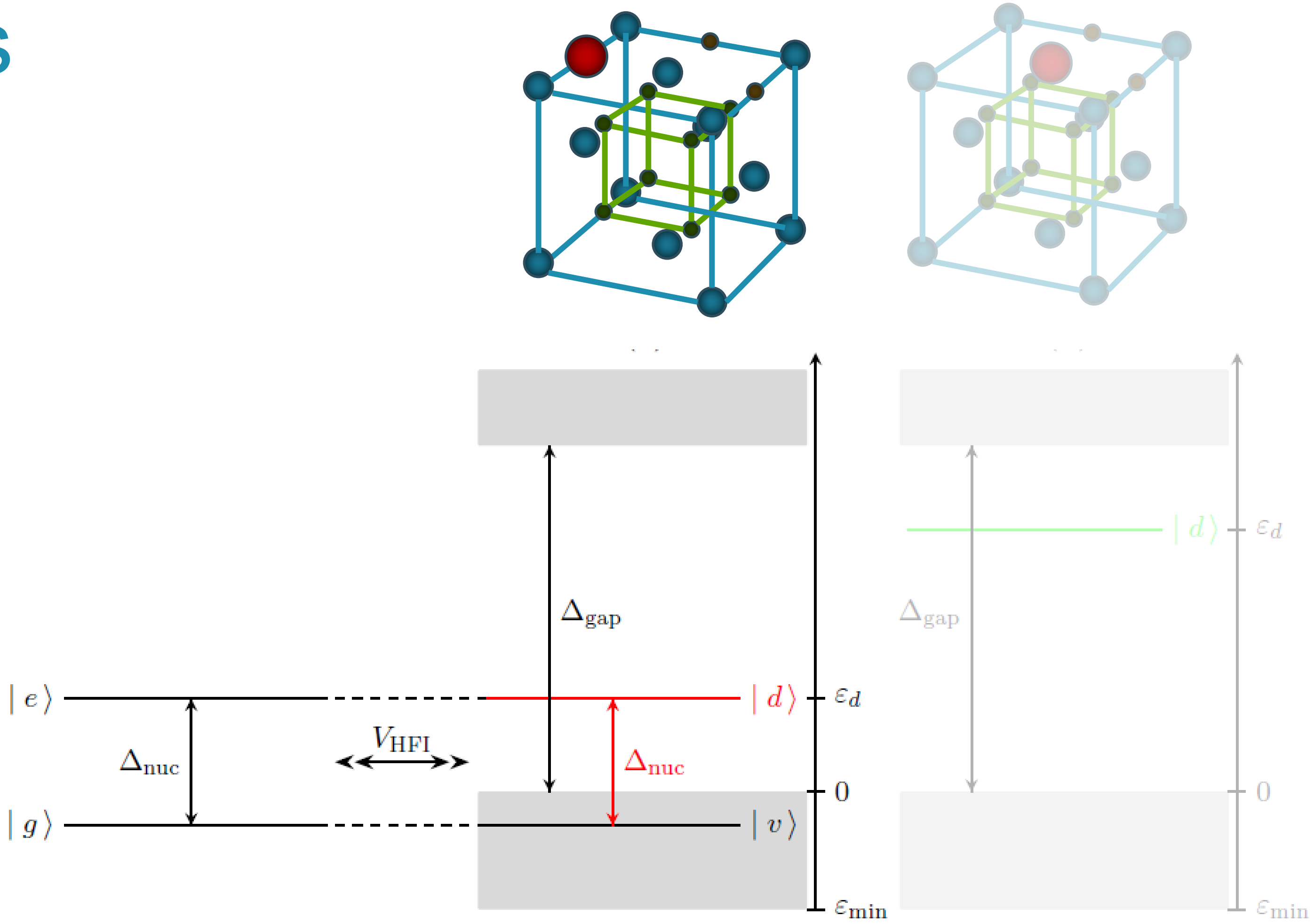
Results from 2021 ISOLDE beam time led to laser excitation in CaF_2 (PTB) and $LiSrAlF_6$ (UCLA), and even excitation with a frequency comb (JILA)





[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 ^{229m}Th nuclear isomeric transition and the ^{87}Sr atomic clock*, 2024

Forcing the radiative decay with large-bandgap crystals



Internal conversion allowed

Internal conversion forbidden

[5] H. W. T. Morgan et al. *Theory of internal conversion of the ^{229}Th nuclear clock isomer in solid-state hosts*

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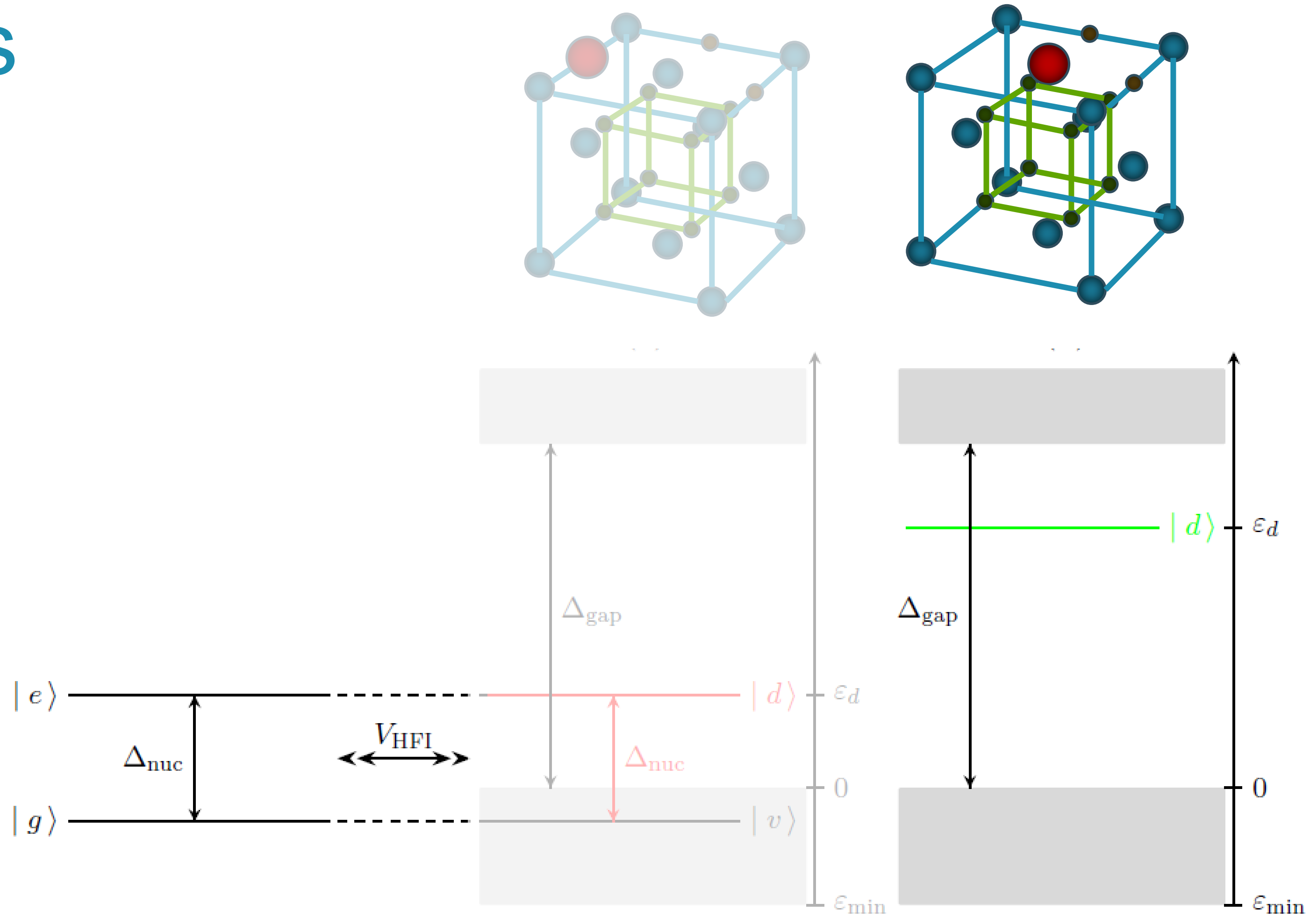

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Main objectives

 Performing **vacuum-ultraviolet spectroscopy** on ^{229m}Th to…

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Radiative decay of the ^{229m}Th nuclear clock isomer in different host materials

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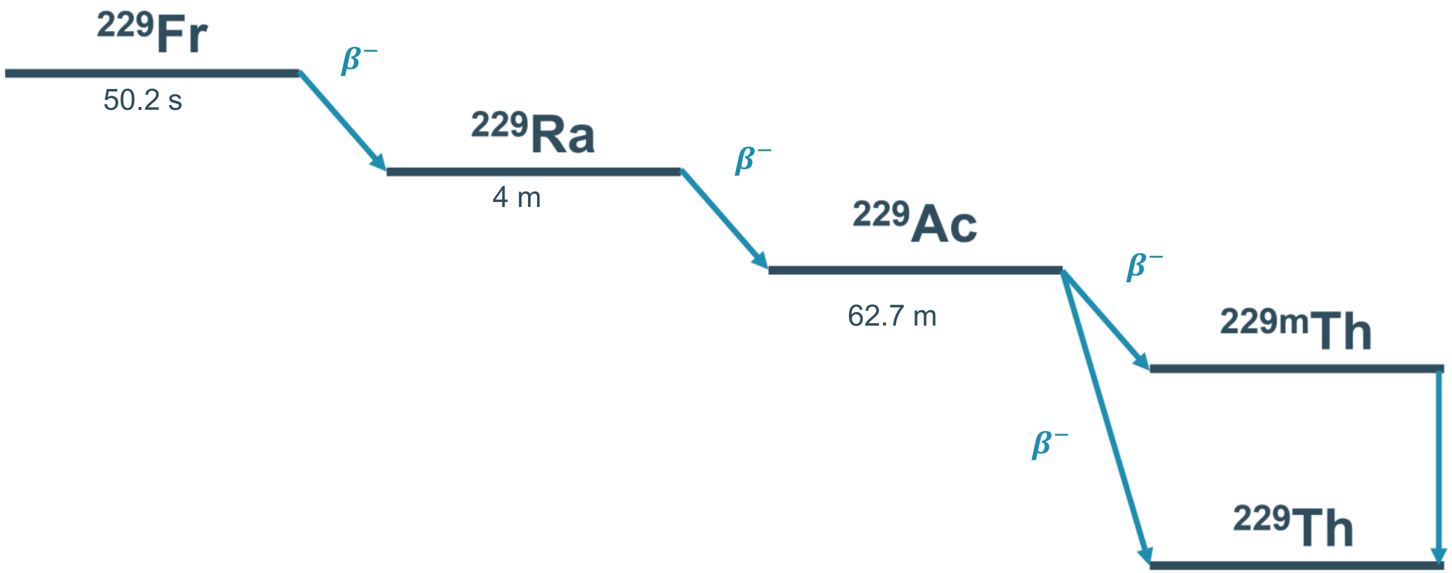
[6] S. Pineda et al. *Radiative decay of the ^{229m}Th nuclear clock isomer in different host materials, PRR* 2025

VUV spectroscopy of the isomer at ISOLDE

Populate the isomer by implanting an A=229 beam in our crystals



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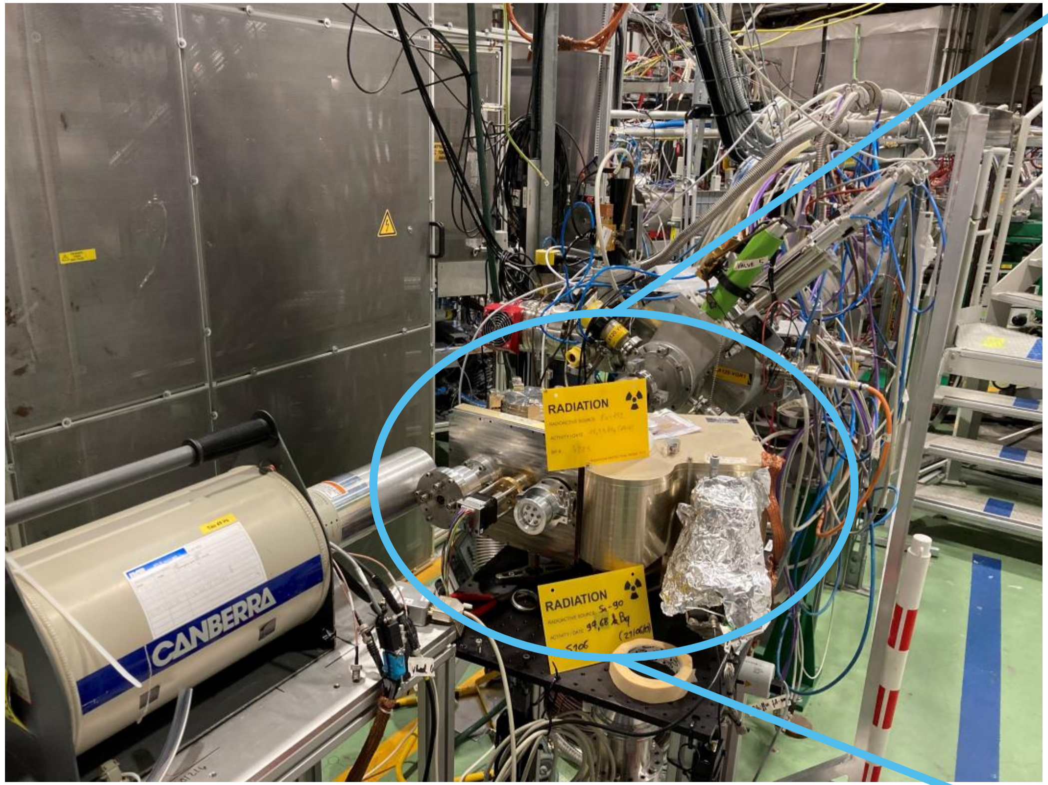
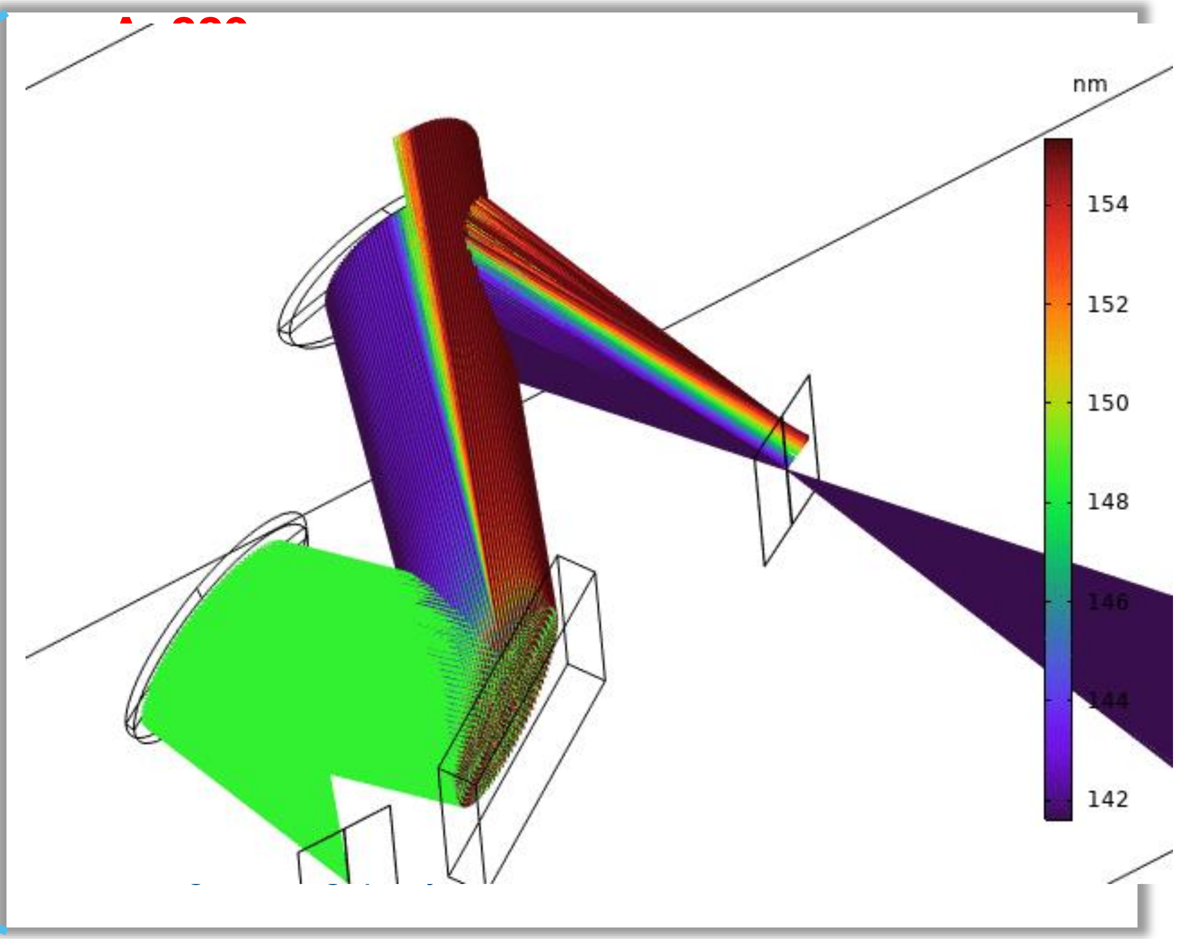




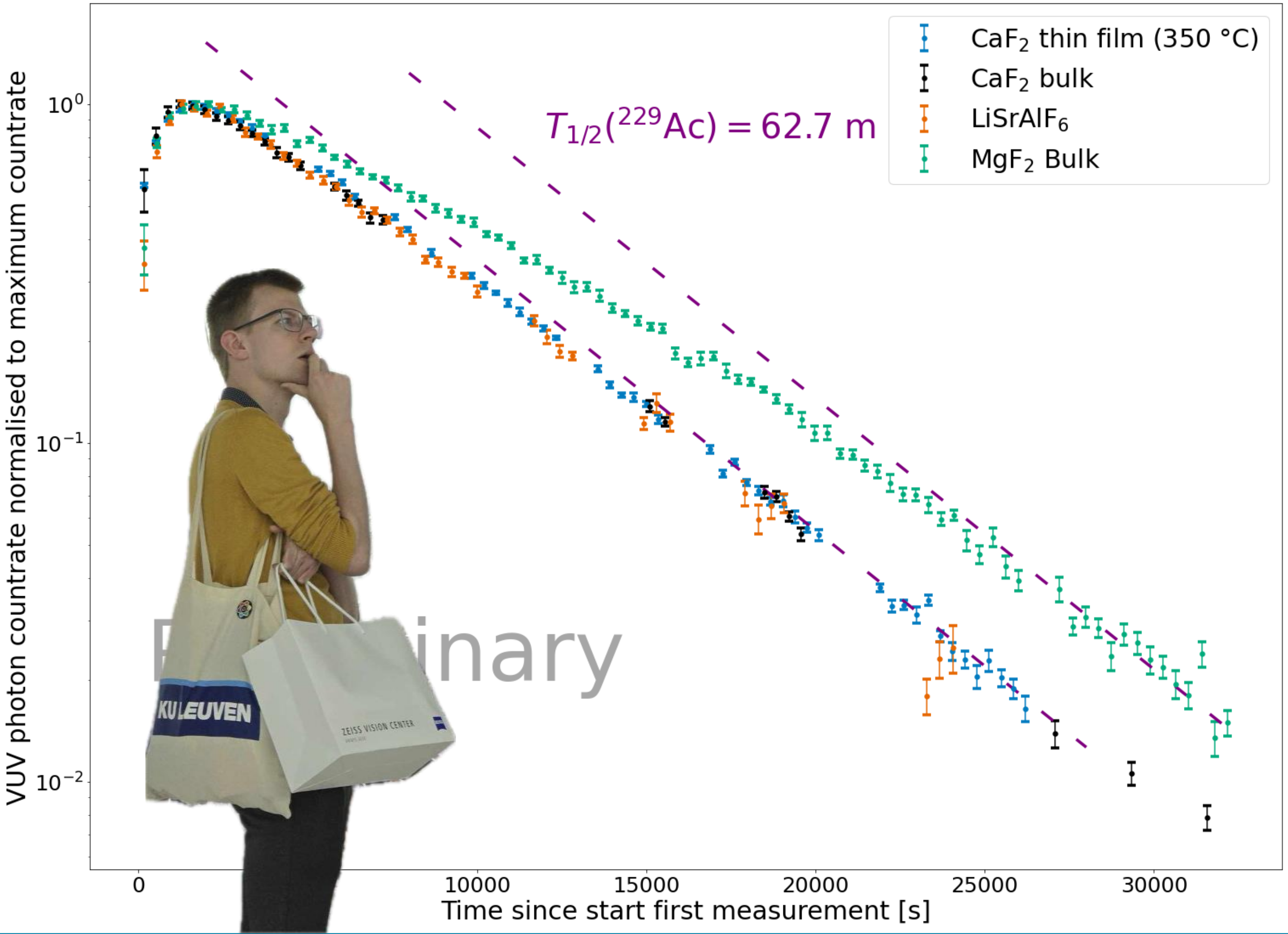
Studying the time behaviour of the VUV signal

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

When it reaches equilibrium depends on the crystal

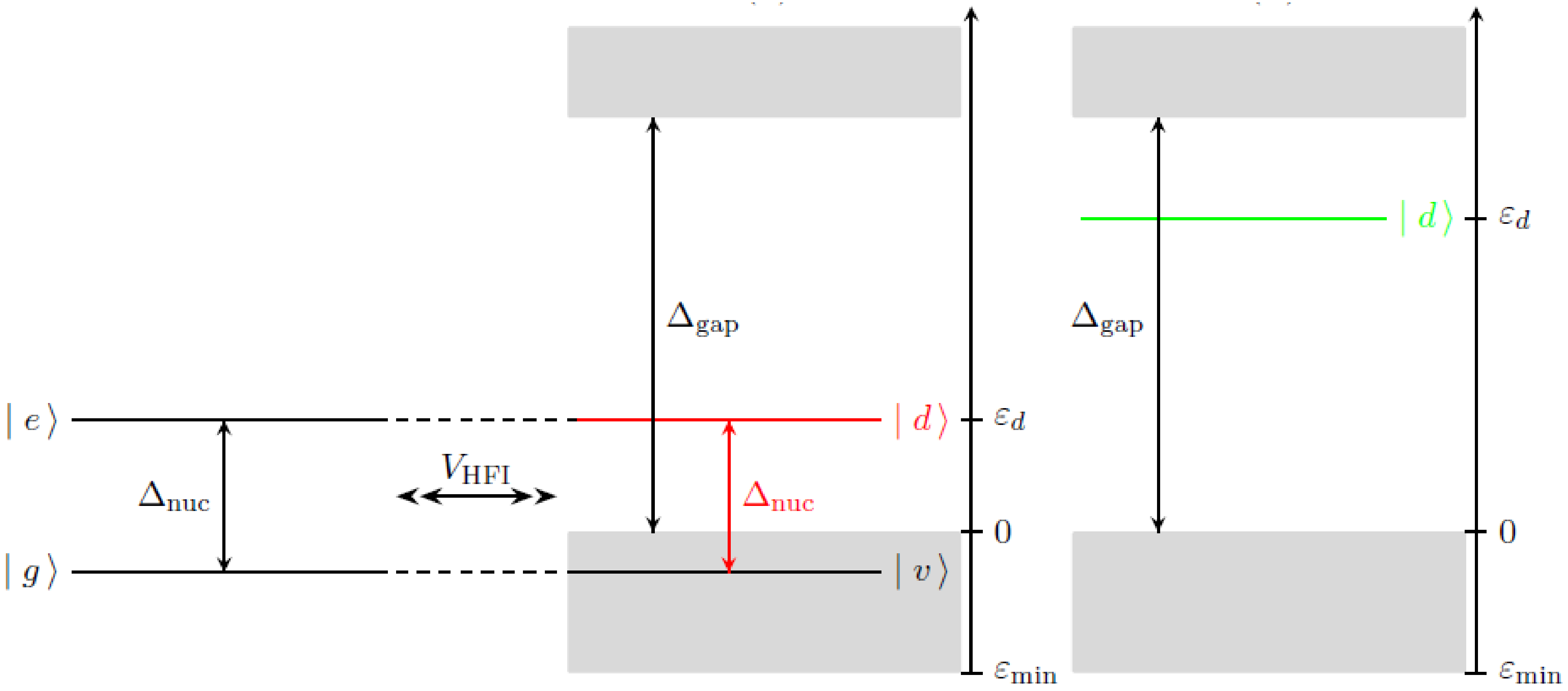
It looks like the activity of the precursors 'quenches' the VUV signal

Such quenching has been observed to be caused by X-rays (Spring-8) *and* by laser light (PTB)



[7] T. Hiraki et al. *Controlling ^{229}Th isomeric state population in a VUV transparent crystal,* Nature, 2024
[8] F. Shaden et al. *Laser-induced quenching of the Th-229 nuclear clock isomer in calcium fluoride,* 2024
[9] T. Hiraki et al. *laser Mössbauer spectroscopy of ^{229}Th,* 2025

Quenching mechanism: opening up the IC channel?



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[10] J. E. S. Terhune et al. *Photo-induced quenching of the ^{229}Th isomer in a solid-state host*, 2024

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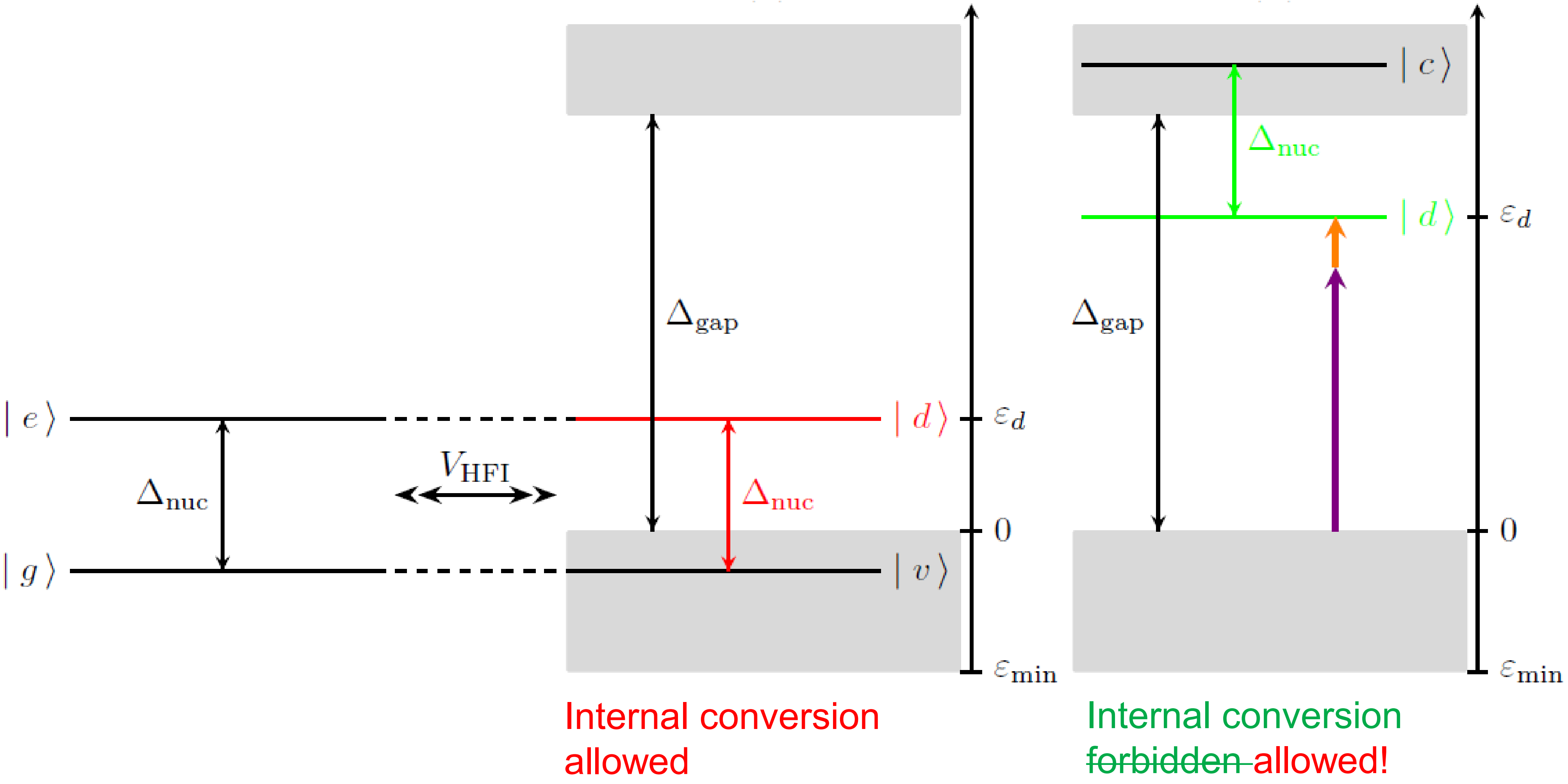
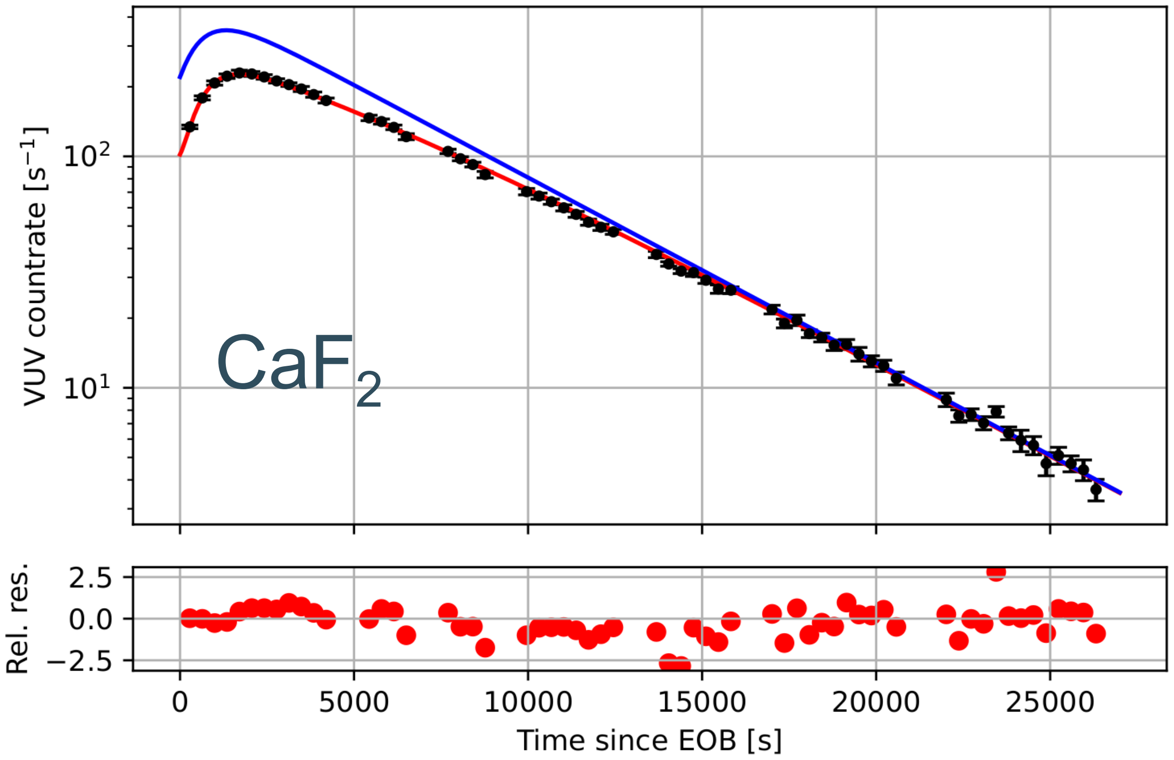


[10] J. E. S. Terhune et al. *Photo-induced quenching of the ^{229}Th isomer in a solid-state host*, 2024

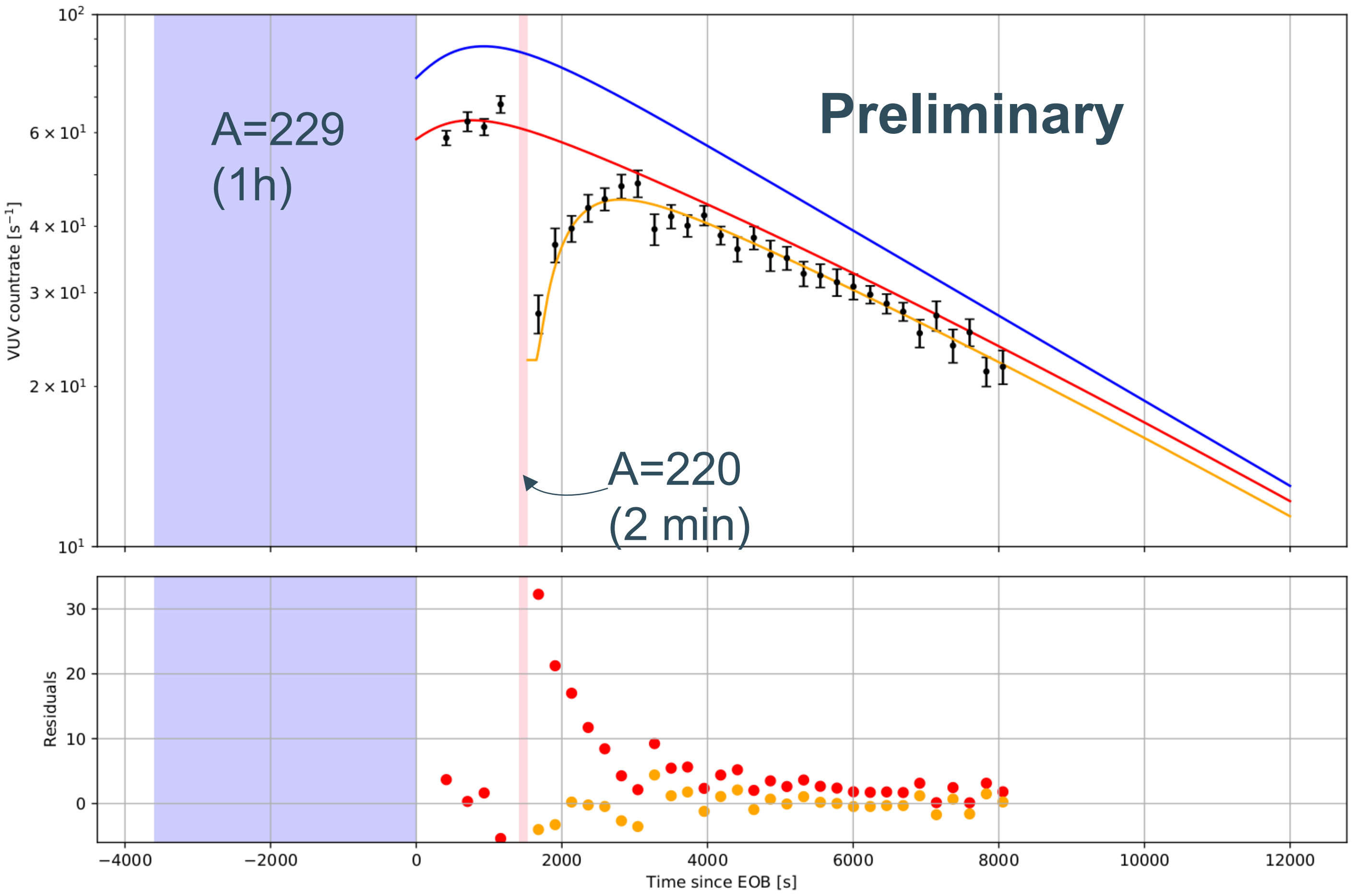
Describing the quenching mechanism in CaF_2

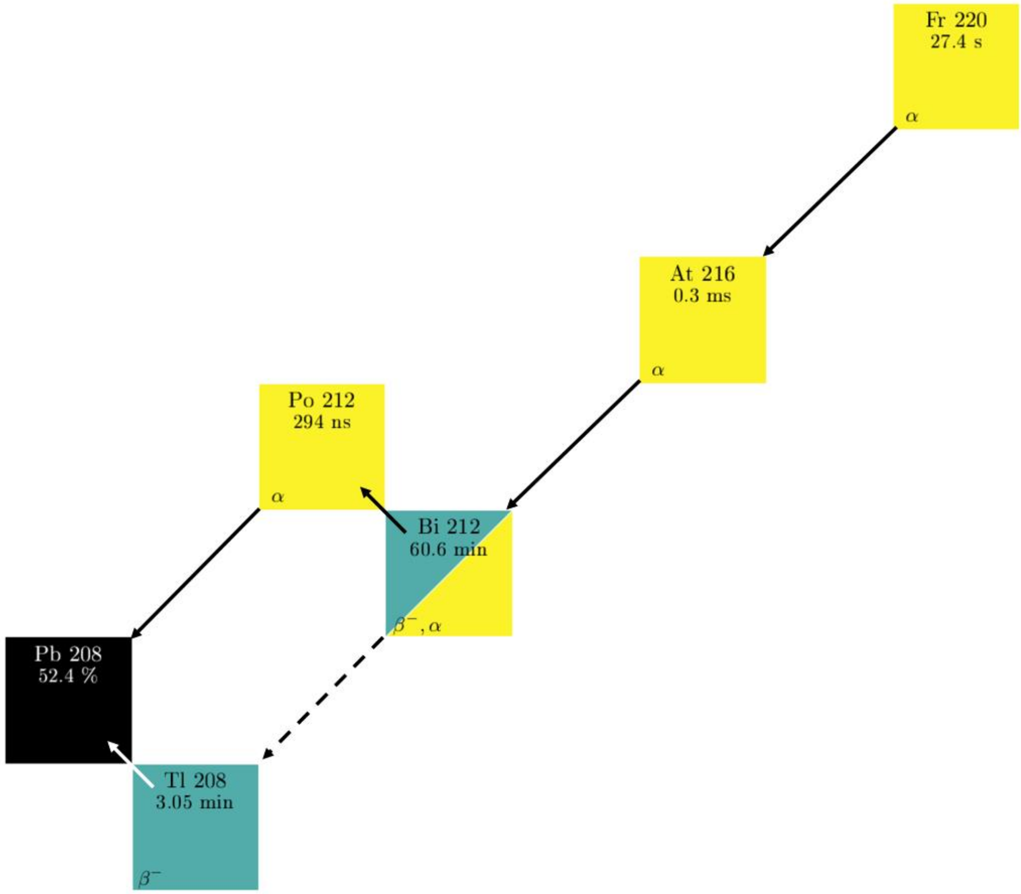
$$\frac{\mathrm{d}N_{\mathrm{rad}}}{\mathrm{d}t} = \lambda_{\mathrm{Ac}} N_{\mathrm{Ac}} - \left(1 + \frac{\sigma_{\mathrm{Ra}}}{\lambda_{\mathrm{rad}}} \lambda_{\mathrm{Ra}} N_{\mathrm{Ra}} + \frac{\sigma_{\mathrm{Ac}}}{\lambda_{\mathrm{rad}}} \lambda_{\mathrm{Ac}} N_{\mathrm{Ac}}\right) \lambda_{\mathrm{rad}} N_{\mathrm{rad}}$$

Assume **linear relationship** between quenched fraction and activity of the precursors present in the crystal.



2025: α emitters also quench the signal





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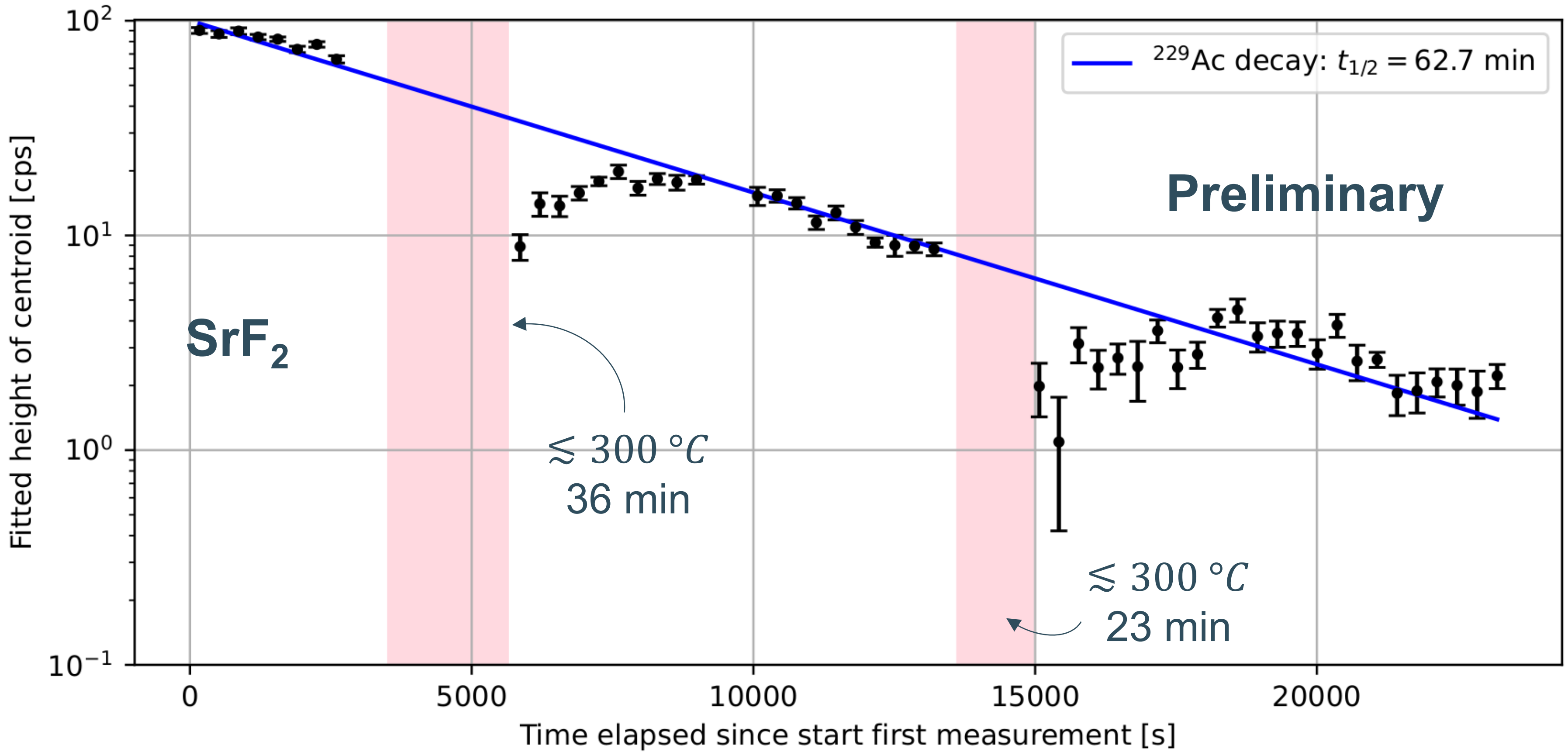
2025: effect annealing on quenching



Conclusion

We performed **vacuum-ultraviolet spectroscopy** on ^{229m}Th to measure the **time behaviour** of the radiative decay.

We investigate a **quenching mechanism** of the radiative decay, which can be …

α-induced
β-induced
Temperature enhanced

Acknowledgements

The IS715 collaboration

M. Athanasakis, S. Arasada, M. Au, S. Bara, M. Bartokos, K. Beeks, S. Casci, P. Chhetri, K. Chrysalidis, A. Claessens, J. G. Correia, Ch. Dullmann, Y. Elskens, R. Ferrer, R. Heinke, G. Holthoff, 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, S. Sabrieva, F. Schaden, K. Scharl, T. Schumm, S. Stegeman, J. Stricker, T. Teschler, P. Thirolf, P. Van Duppen, A. Vantomme, R. Villarreal, U. Wahl, Y. Wang, M. Wiesinger, F. Zacherl

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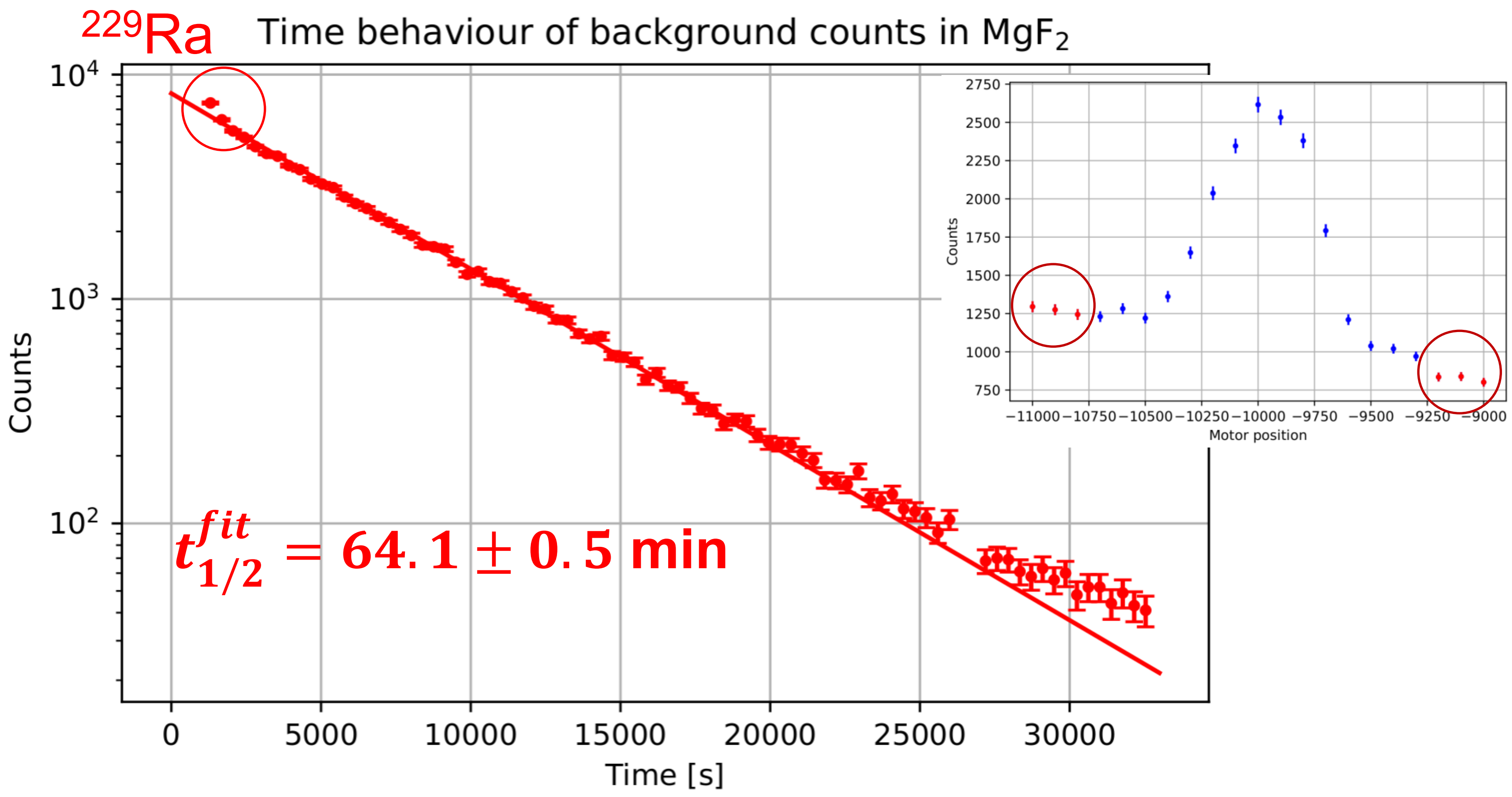
Back up

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

A: No

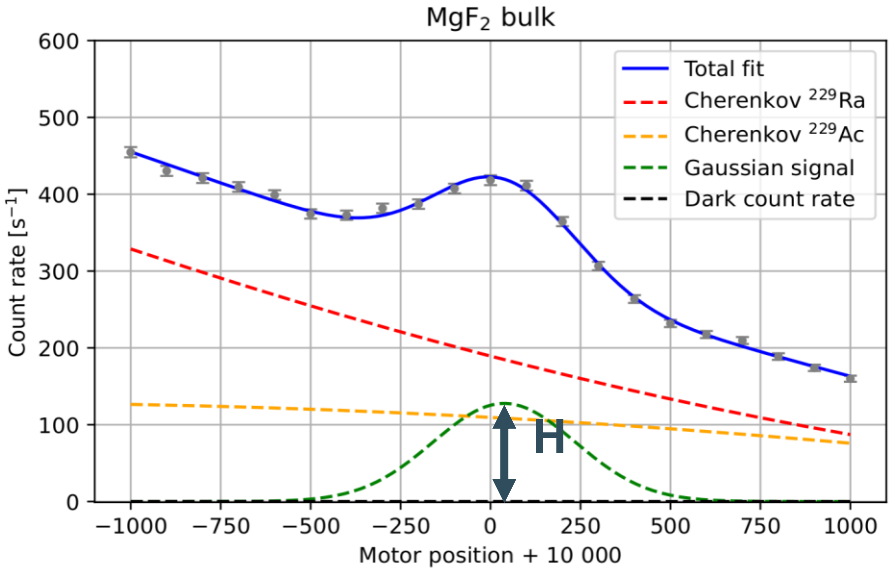


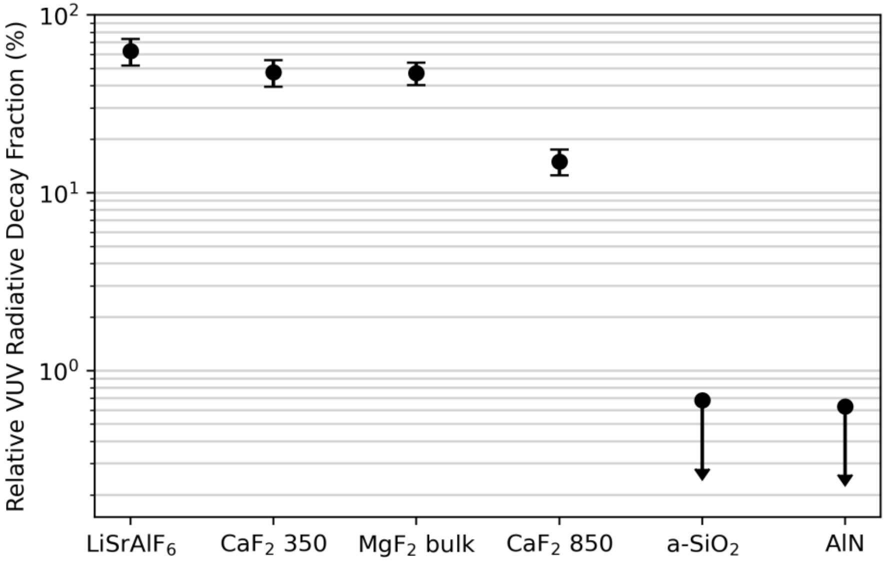
Which crystals have the highest radiative-decay fraction?

$$\varepsilon_{\mathrm{VUV}} = \frac{H}{A_{\mathrm{iso}}\,\varepsilon_{\mathrm{I}}}$$

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

Determine limits for AlN and SiO_2





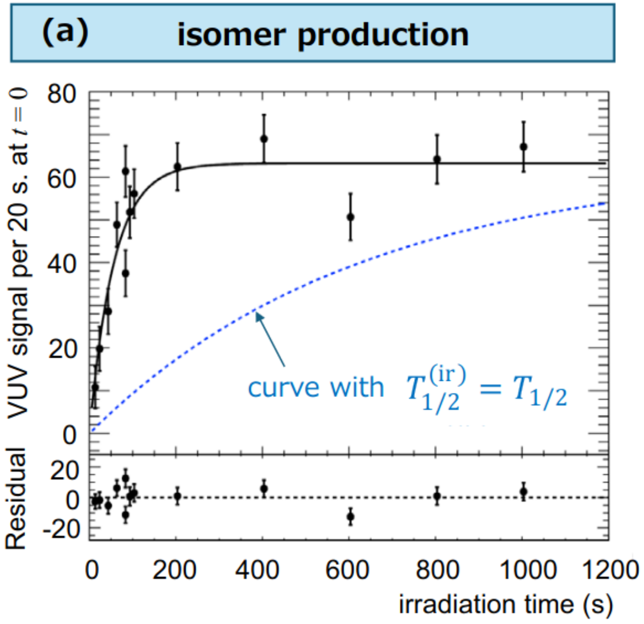
[6] S. Pineda et al. *Radiative decay of the ^{229m}Th nuclear clock isomer in different host materials, PRR* 2025

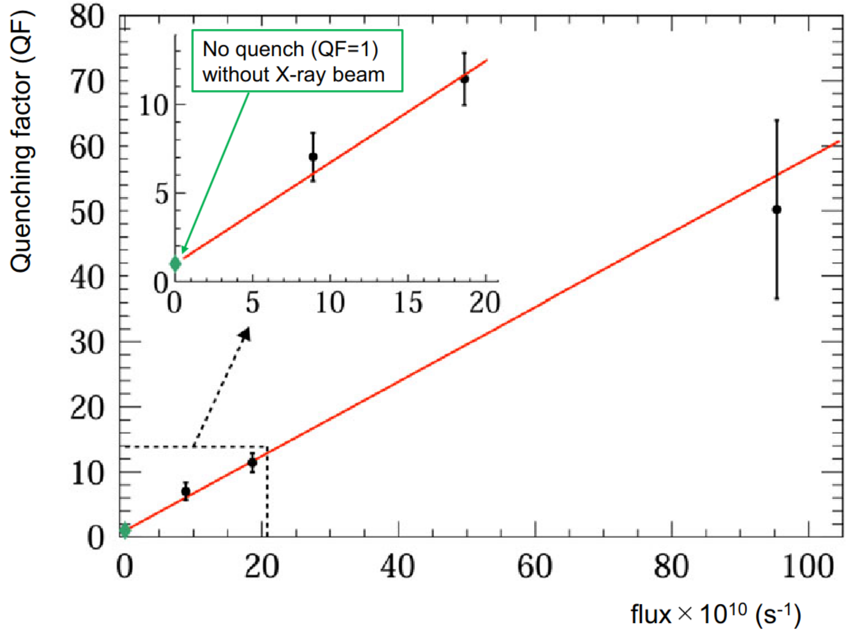
‘Quenching’ of the halflife

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

Flux-dependent ‘quenching’ of the observed halflife

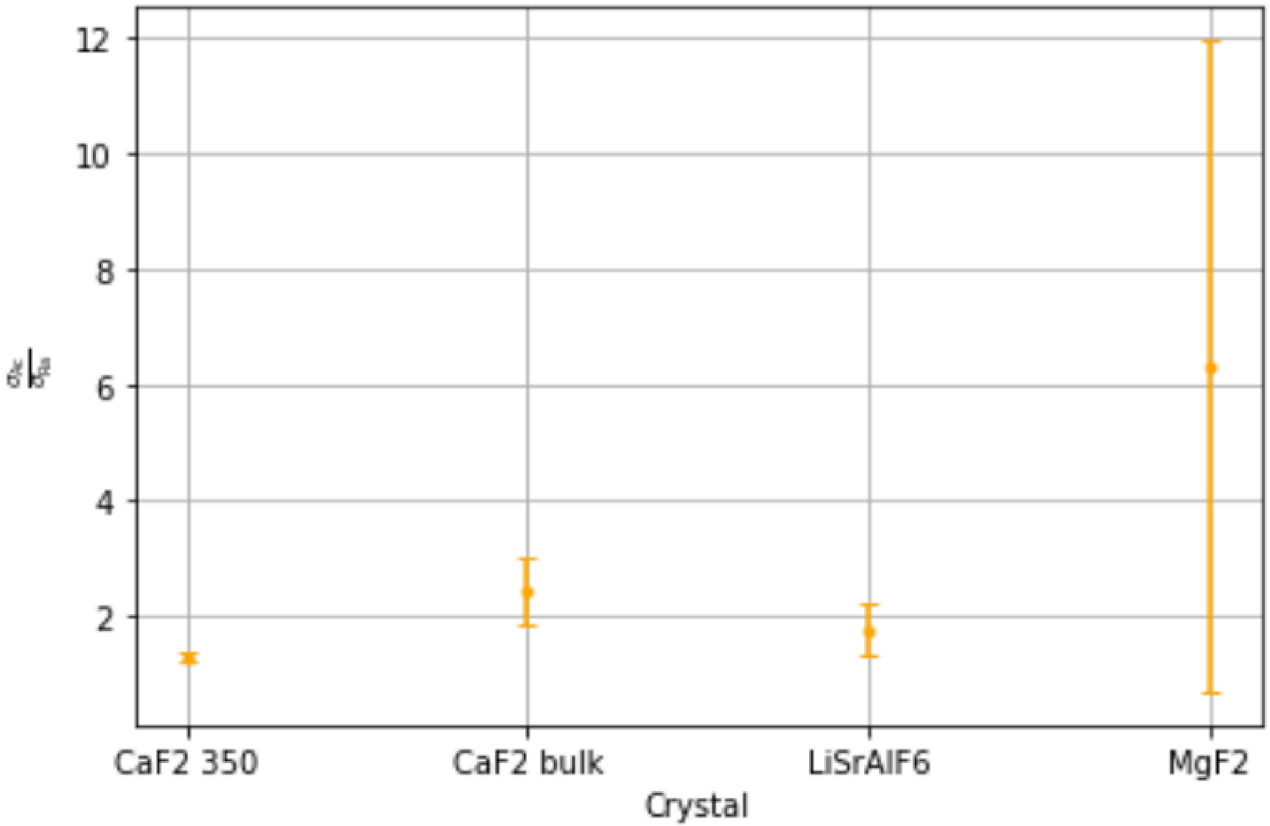
$$Q = \frac{t_{1/2}}{t_{1/2}^{(\mathrm{irr})}}$$





[7] T. Hiraki et al. *Controlling ^{229}Th isomeric state population in a VUV transparent crystal*, 2024

Overview: ratio of the cross sections



Consider relative contributions to Cherenkov background from Ac and Ra.in bulk crystals

Crystal	$\frac{\text{Cherenkov Ac}}{\text{Cherenkov Ra}}$
CaF_2	0.66
$LiSrAlF_6$	0.33
MgF_2	0.41

Totally different from fitted quenching cross sections ratios
→ **Suggests quenching is β-induced**