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Study of the Radiative Decay of the Low-Energy Isomer in ^{229}Th

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64th INTC meeting

24/06/2020

The Road towards a Nuclear Clock

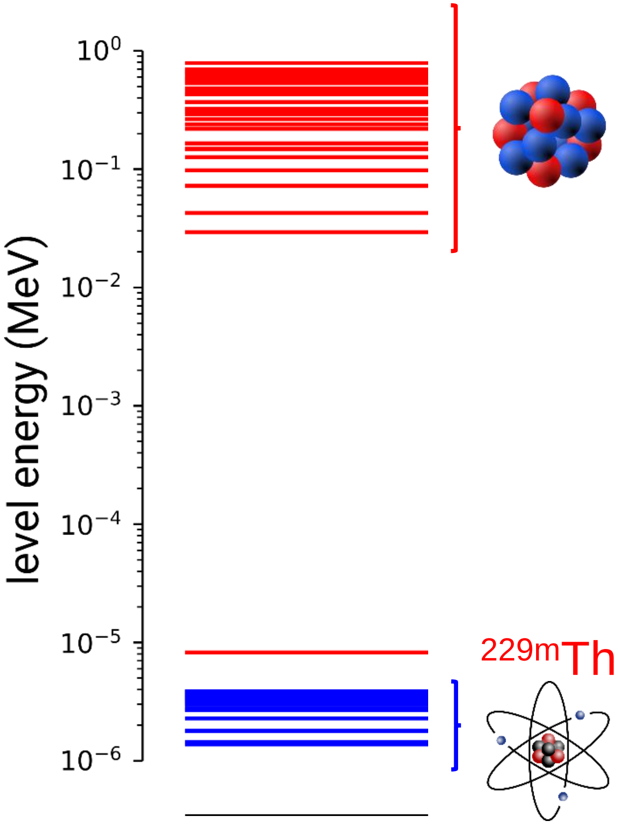
Atomic clock based on electronic shell

- Limitations:
  Susceptible to the environment, i.e. Stark, Zeeman
  Transition-intrinsic properties, i.e. $\nu_0/\Delta\nu$

Concept of a nuclear clock based on ^{229}Th isomer[1]

- Low-lying nuclear isomer $\sim 10eV$
  → still in range of today's laser technology
- Favorable lifetime → $\Delta E/E \sim 10^{-19}$
- Much less susceptible to environment→ expected clock acc. $\sim 10^{-19}$[2]
- new perspectives in ultra-high precision frequency quantum metrology:
  → applications
  → sensitivity to variations of fundamental constants [3]

(see also Th. Schumm (TU Vienna) – CERN Colloquium June 11, 2020)



(1) Peik and Tamm 2014 *J. Phys. Condens. Matter* **26** 10
(2) Campbell et.al. 2012 *PRL* **108** 12
(3) Flambaum 2006 *PRL* **97** 9

Nuclear Structure of the low-energy isomer ^{229m}Th

Time line of the measured properties:

1976: First evidence in γ-spectroscopy(4)

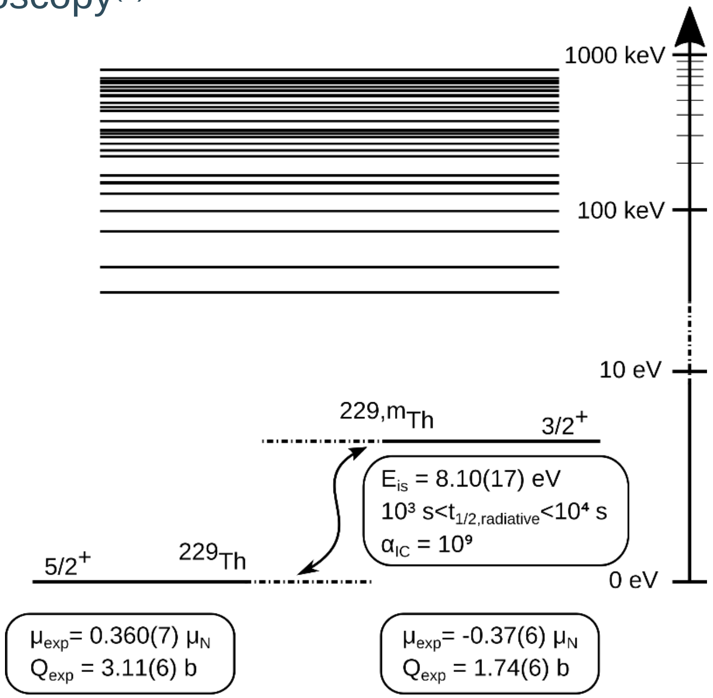
⋮

2016: Experimental proof of existence(5)

2018: Isomer's magnetic dipole and quadrupole moment(7)

2019: $E_{iso} = 8.28(17) eV$ from conversion electron spectroscopy(8)

2020: $E_{iso} = 8.10(17) eV$ from micro-calorimeter γ-spectroscopy(9)

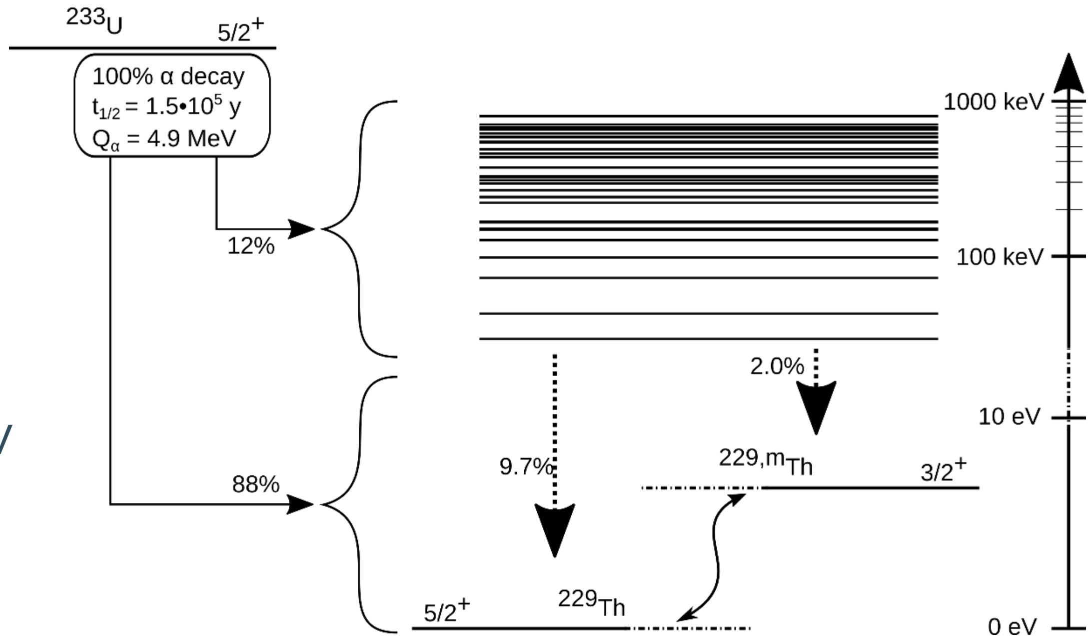


Current status – ^{229m}Th

- Energy is poorly-defined
- Radiative decay not yet observed
- Internal conversion ($^{229}Th^{0}$): $T_{1/2,IC} = 7(1)10^{-6}\ s$ (5,6)
- Radiative decay ([$^{229}Th^{1+}$], $^{229}Th^{2+}$,..): $T_{1/2,rad} \sim 10^{3} - 10^{4} s$

(4) Kroger and Reich 1976 *Nucl. Phys. A* **259** 1
(5) Von der Wense et.al. 2016 *Nature* **533** 7601
(6) Seiferle et.al. 2017 *PRL* **118** 4
(7) Thielking et.al. 2018 *Nature* **556** 7701
(8) Seiferle et.al. 2019 *Nature* **573** 7773
(9) Sikorsky et.al. Arxiv:2005.13340

Population of the isomer



Direct laser excitation of ^{229}Th

- Energy:
  $E_{iso} = 8.1(17)eV, \ \lambda = 153.1(37)nm,$
- Ionization potential of thorium:

Th^0: 6.1 eV – Th^{1+}: 11.5 eV – Th^{2+}: 20 eV

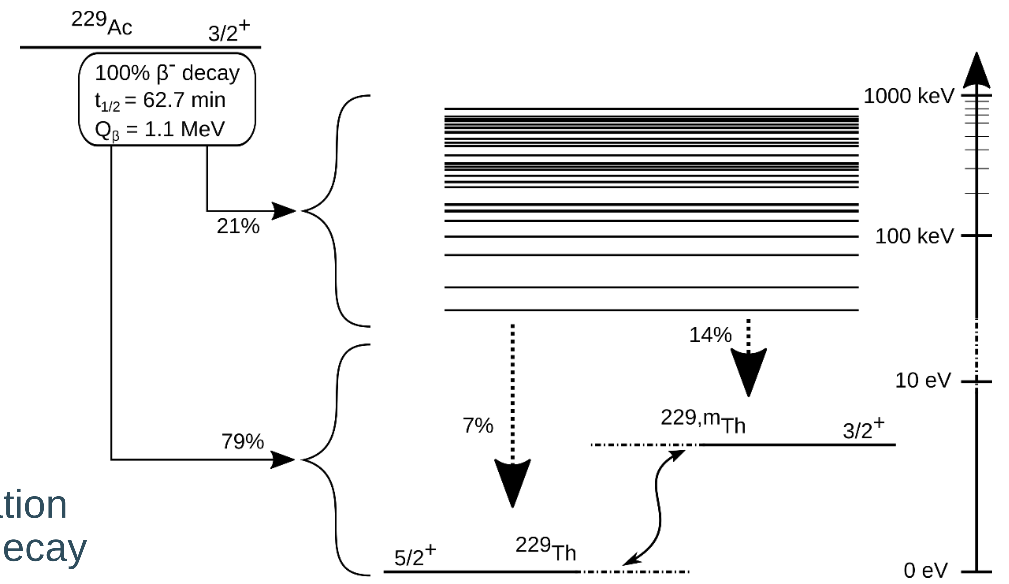
Population in α decay of ^{233}U

- ^{233}U feeds the isomer with a 2% branching ratio
- Embedded in a solid state matrix: ^{233}U-doped CaF_2 crystal (transparent at 150nm)
- Blocking of IC channel for a Th^{4+} charge state in a Ca^{2+} substitutional position(10)
- $85keV$ recoil of the ^{229m}Th/^{229}Th daughters – lattice position?
- Radio luminescence from α-radiation

→ Observation of radiative decay to-date not successful

(10) Dessovic *et al.* 2014 *J. Phys. Condens. Matter* **26** 10

Population of the isomer in the β-decay of ^{229}Ac



Alternative approach: β-decay of ^{229}Ac

- Feeding of the isomer: 14% indirect
  between 0% and 79% direct [11]
- $< 6eV$ recoil energy
  → preservation of lattice location
- $T_{1/2} = 62.7(5)min$ allows annealing
  →optimization of lattice position
- availability of a pure ^{229}Ac beam for implantation and ^{231}Ac for lattice position studies after β decay
- Laser-ionization and availability of other radioisotopes improve background characterization

Aims of this proposal

I. Quantification and optimization of the substitutional incorporation of Ac/Th in CaF_2

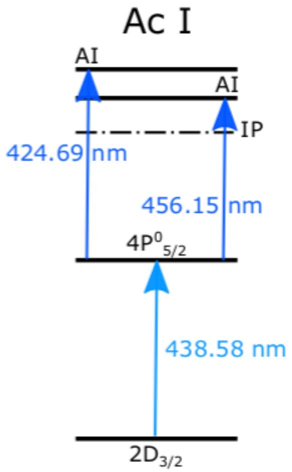
II. Detection of the radiative decay of the isomer

determination of the radiative half-life

determination of the energy: $< 0.1nm$ accuracy to bridge the gap to laser excitation

(11) E. Ruchowska et al., Physical Review C 73, 044326 (2006).

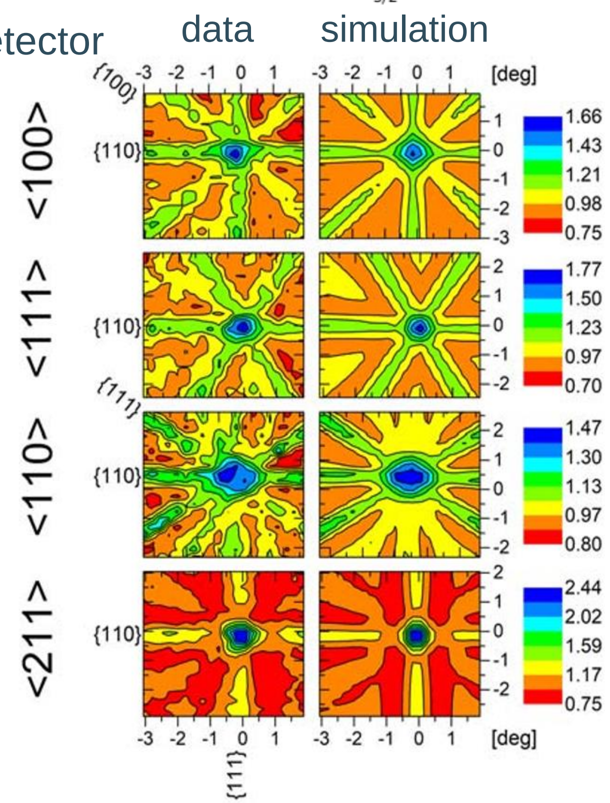
Preparatory study at ISOLDE: LoI198



- Implementation of laser ionization of Ac
  ^{229}Ac beam at LA1: $8.8(2) \cdot 10^5 pps$

- γ-CE-spectroscopy
  ^{229}Ac beam retarded to 2 keV and implanted in Nb and Au foils
  Low-energy electron detection system based on a Channeltron detector
  → no signature of isomer CE with $4\mu s < T_{1/2} < 80\mu s$
  → level scheme in agreement with literature

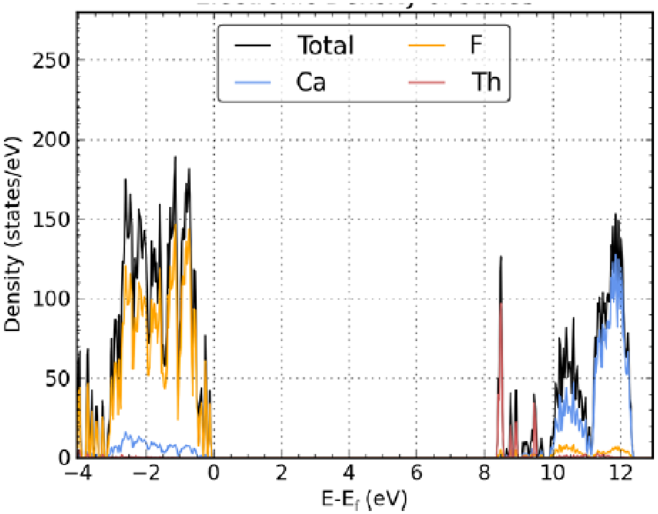
- Emission channeling of ^{229}Ac implanted in CaF_2
  → 90% of ^{229}Ac in substitutional position (preliminary)



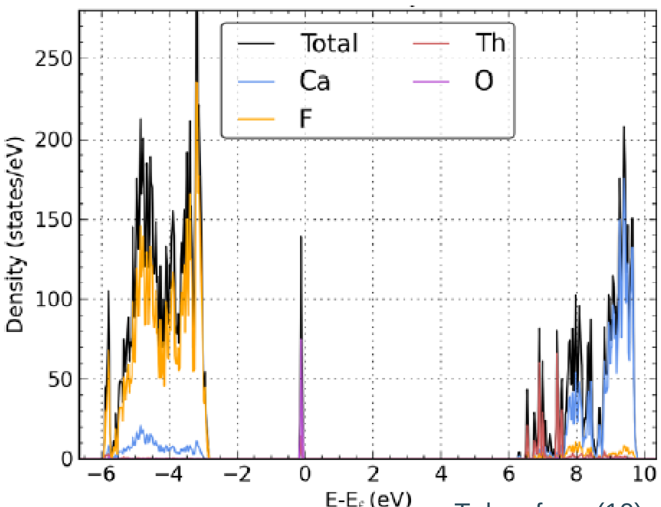
I. Characterisation of the Lattice Position

DFT calculation of Th:CaF_2 DOS

substitutional



interstitial



Taken from (10)

β-emission channeling measurements allow to

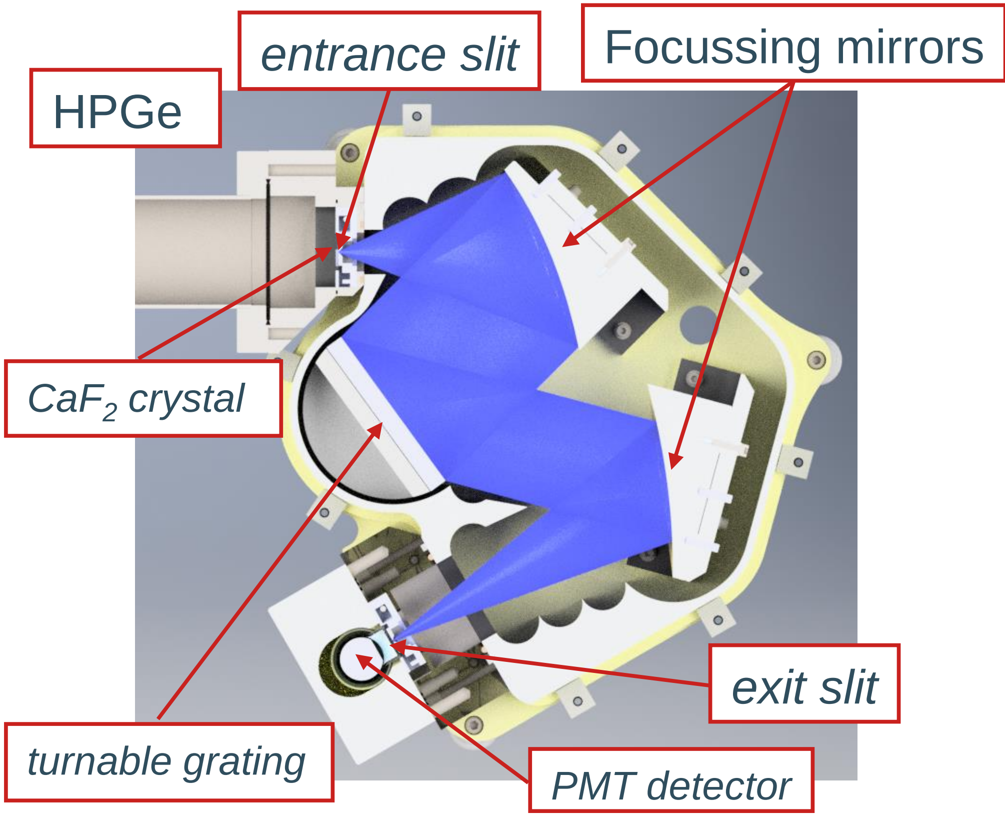
- develop thermal annealing procedure
  to improve substitutional lattice incorporation
  → first observation of radiative decay
  → future developments towards solid state clock
  Beam: $\geq 10^6 pps$ ^{229}Ac beam

- test the inheritance of lattice position in the
  β-decay of ^{229}Ac
  Beam: $\geq 10^5 pps$ ^{231}Ac beam
  Alternative (TAC comment): ^{231}RaF

(10) Dessovic *et al.* 2014 *J. Phys. Condens. Matter* **26** 10

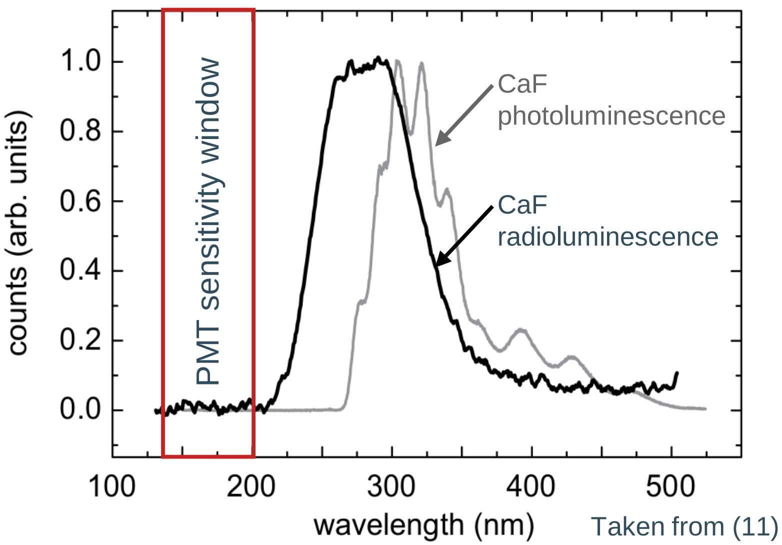
II. Spectroscopy of the Radiative Decay: Methodology

- Implantation into thin (50nm) CaF_2 crystal on Si backing (characterization at KU Leuven)
- Implantation time: 2 half-lives
- Transfer of crystal under vacuum to spectrometer
- Crystal positioned close to entrance slit of VUV spectrometer (design based on Resonance Ltd customized VM180)
- Activity monitoring using a Ge detector
- Simulation of signal strength and worst-case background contributions (see next slide)



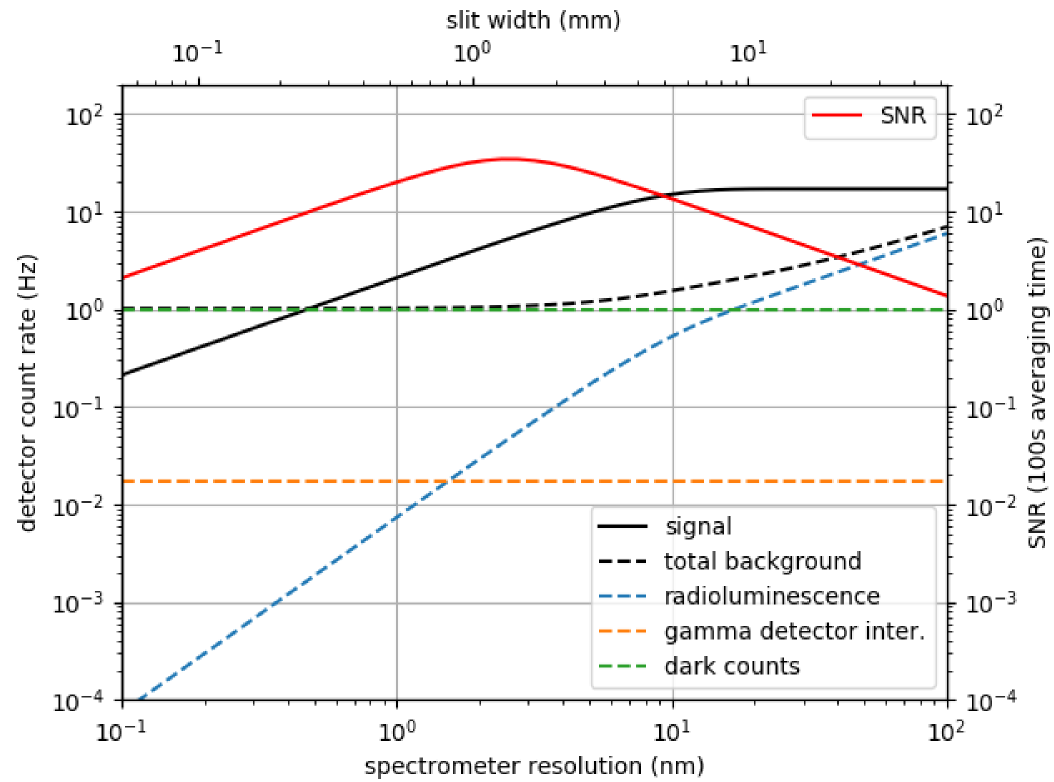
II. Spectroscopy of the Radiative Decay: Background

- Implantation of a 4 mm FWHM ion beam
- Scintillation properties in CaF_2
  α,β: from literature $\sim 1\%$ conversion
  γ: 100% conversion
- PMT sensitivity window
- Conservative estimates of
  - photon coll.+ det. efficiency: $> 0.01\%$
  - substitutional lattice position: 50%
  - isomer feeding: 14%



Taken from (11)

Signal (counts/sec.) and background contributions for $3h$ measurement at $10^6 pps$ implantation (2 h) and $2h$ isomer half-life:



(11) Stellmer *et al.* 2015 *Scient.Rep.* **5** 1

Summary

Study of the Radiative Decay of the Low-Energy Isomer in ^{229}Th

Advantages compared to previous attempts:

- > 14% feeding of the isomer in the β decay of ^{229}Ac (versus 2% in α decay of ^{233}U)
- Implantation into a thin CaF_2 crystal - substitutional position: low recoil energy after β decay (< 6 eV)
- $T_{1/2}$(^{229}Ac) = 62.7 min: annealing and manipulation
- Availability of a pure ^{229}Ac beam: less crystal damage
- Resonance ionization and availability of neighboring mass actinium: control of the background conditions
- Study of ^{231}Ac – ^{231}Th: assess the stability of substitutional incorporation against low-recoil β-decay

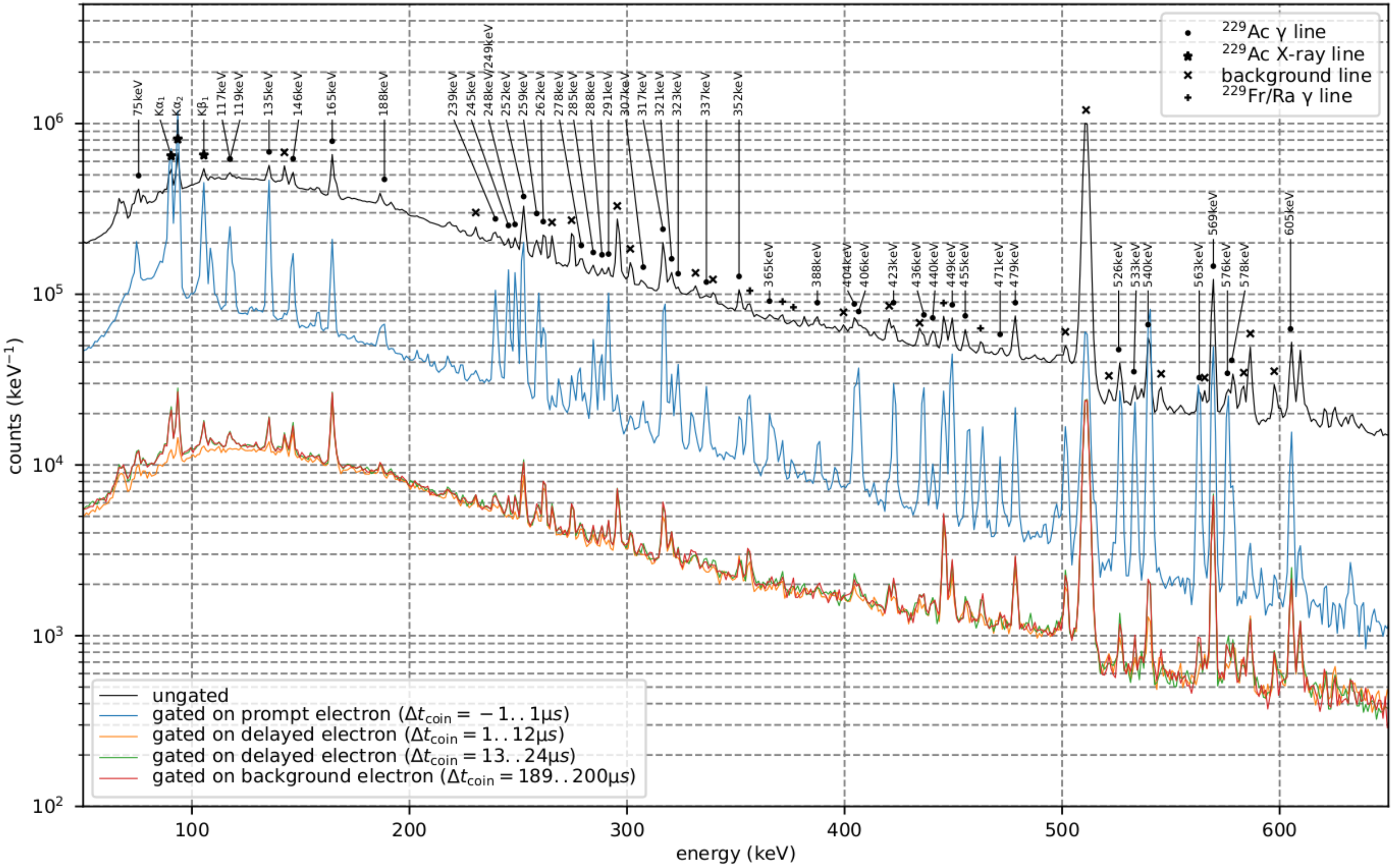
Beamtime Request

- Optimization of extraction of ^{231}Ac or ^{231}RaF
  3 shifts

- Emission channeling measurements
  Quantification and optimization of the substitutional incorporation of Ac/Th in CaF_2Study of the lattice position inheritance in the low-recoil β-decay (^{231}Ac or ^{231}RaF – comment from the TAC)
  2 shifts (multiple collections of 2h spread over several days)

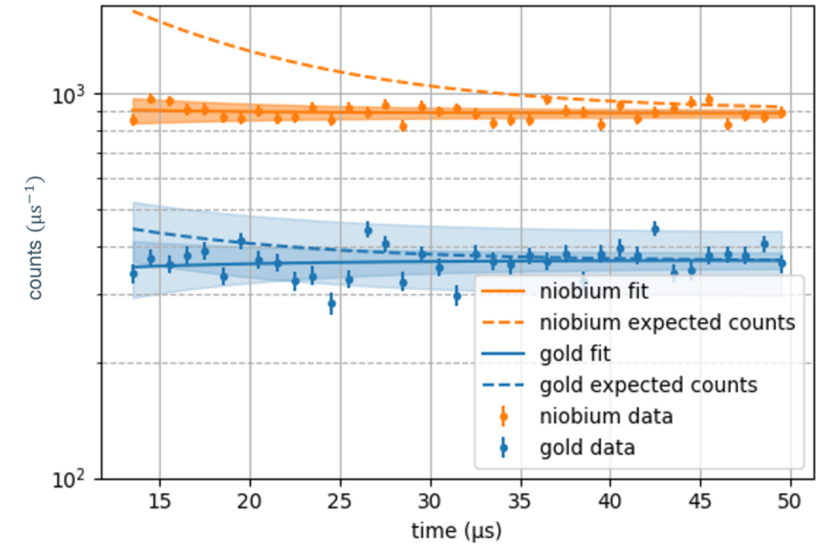
- VUV spectroscopy of the radiative decay
  6 shifts (>20 collections of 2h spread over several days)

LoI198: γ-Spectra



[Implantation into Nb at 2keV]

LoI198: Delayed γ-electron coincidences and CE strength

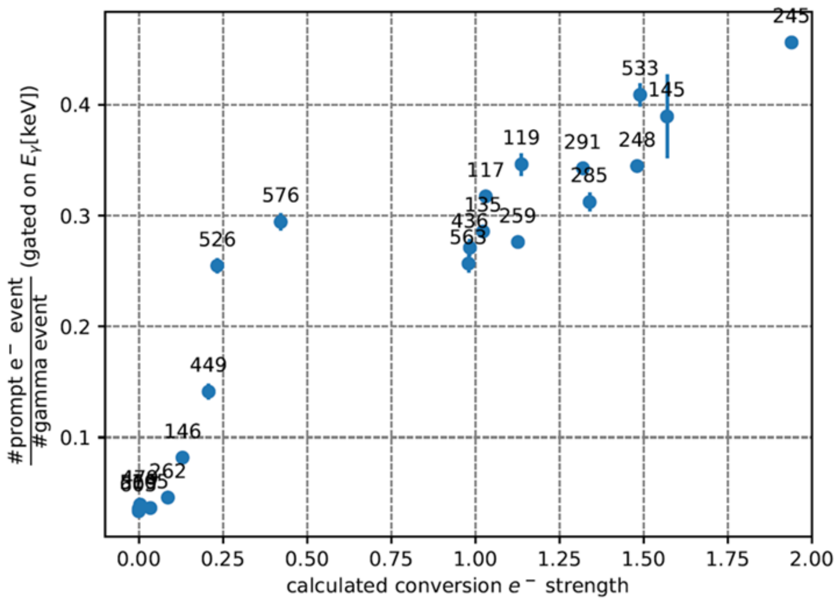


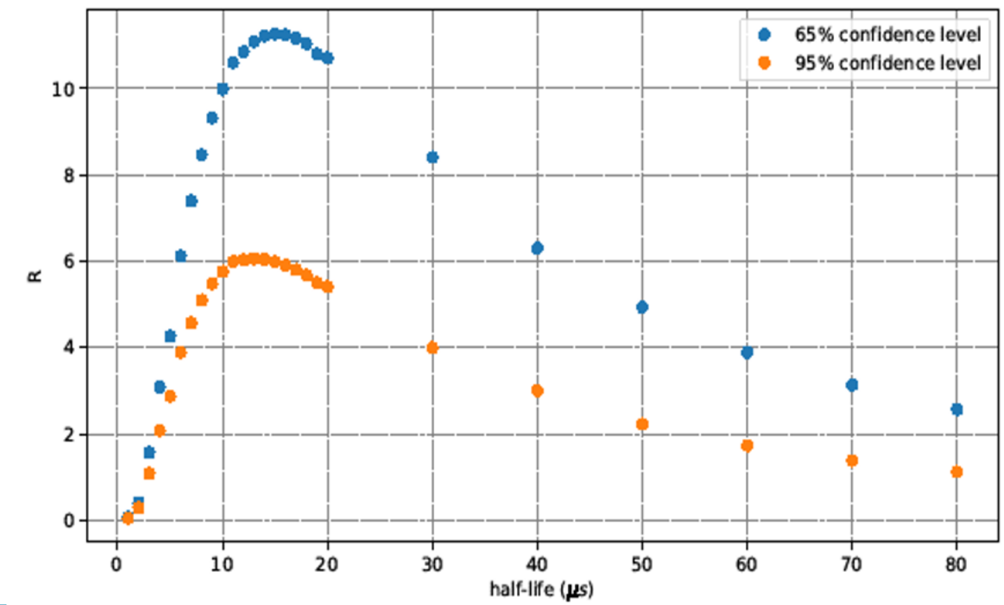
Sensitivity to presence/absence of isomer signal

$$R = \frac{\#observed\ counts(\tau_{IC}) + 2 \cdot uncertainty}{\#expected\ counts}$$

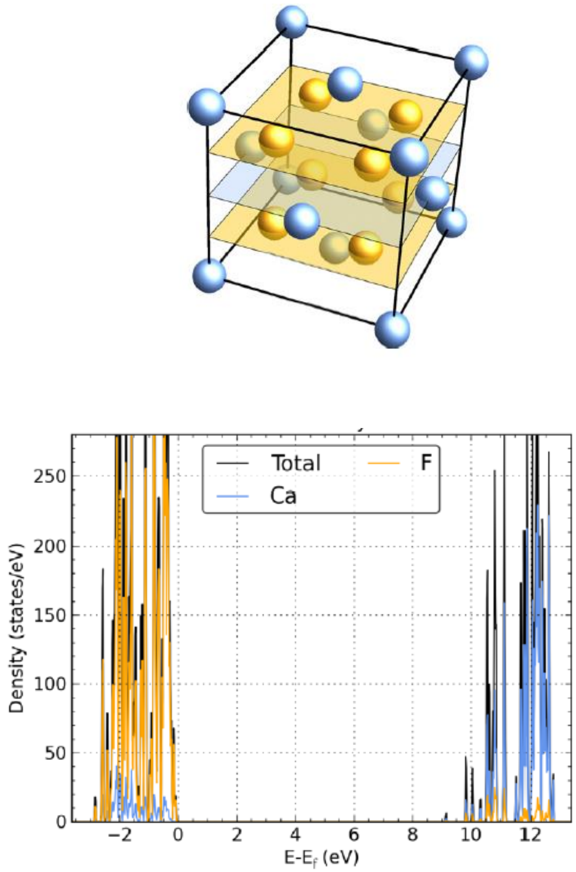
$R > 1$: No isomer signal detected

$R \leq 1$: No sensitivity

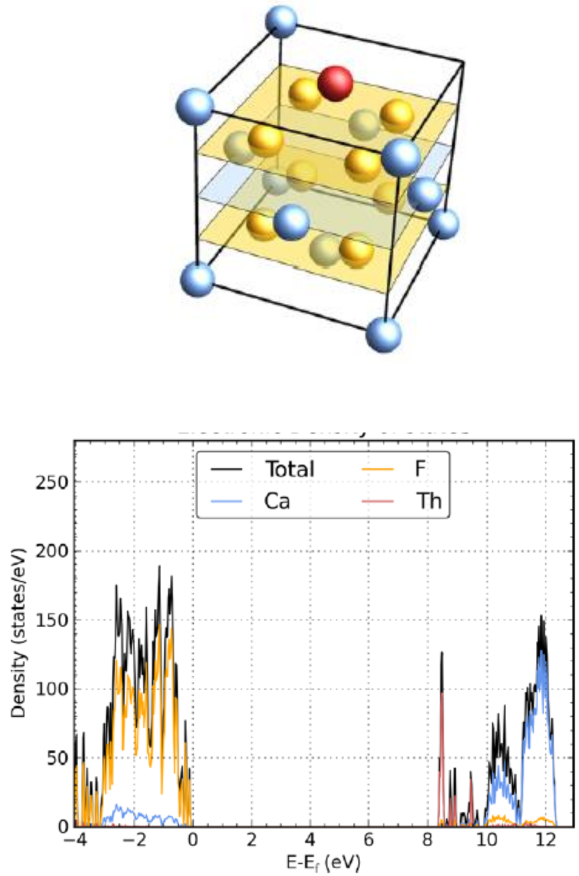




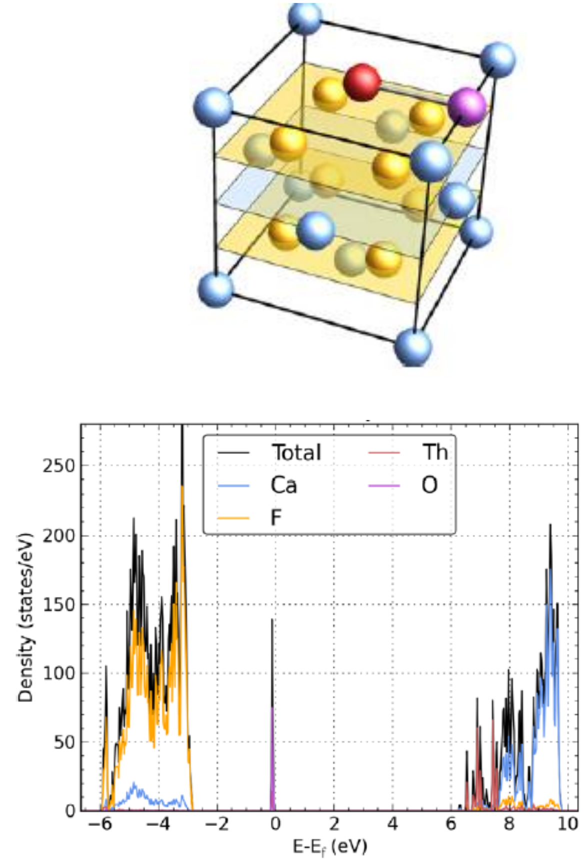
DFT calculation of Th:CaF



undoped

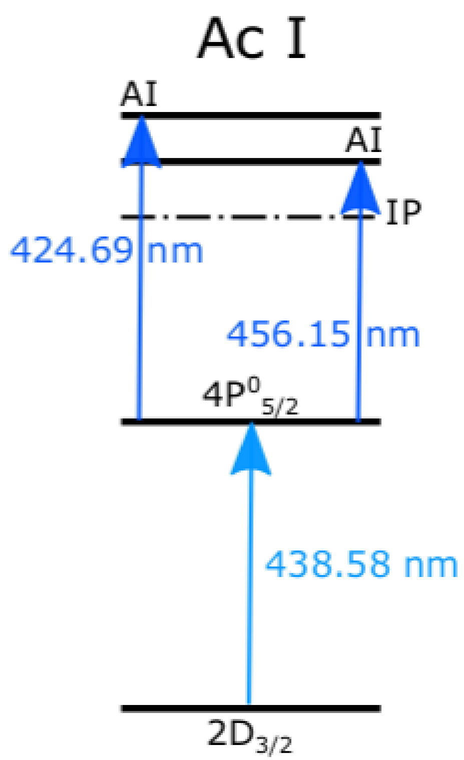


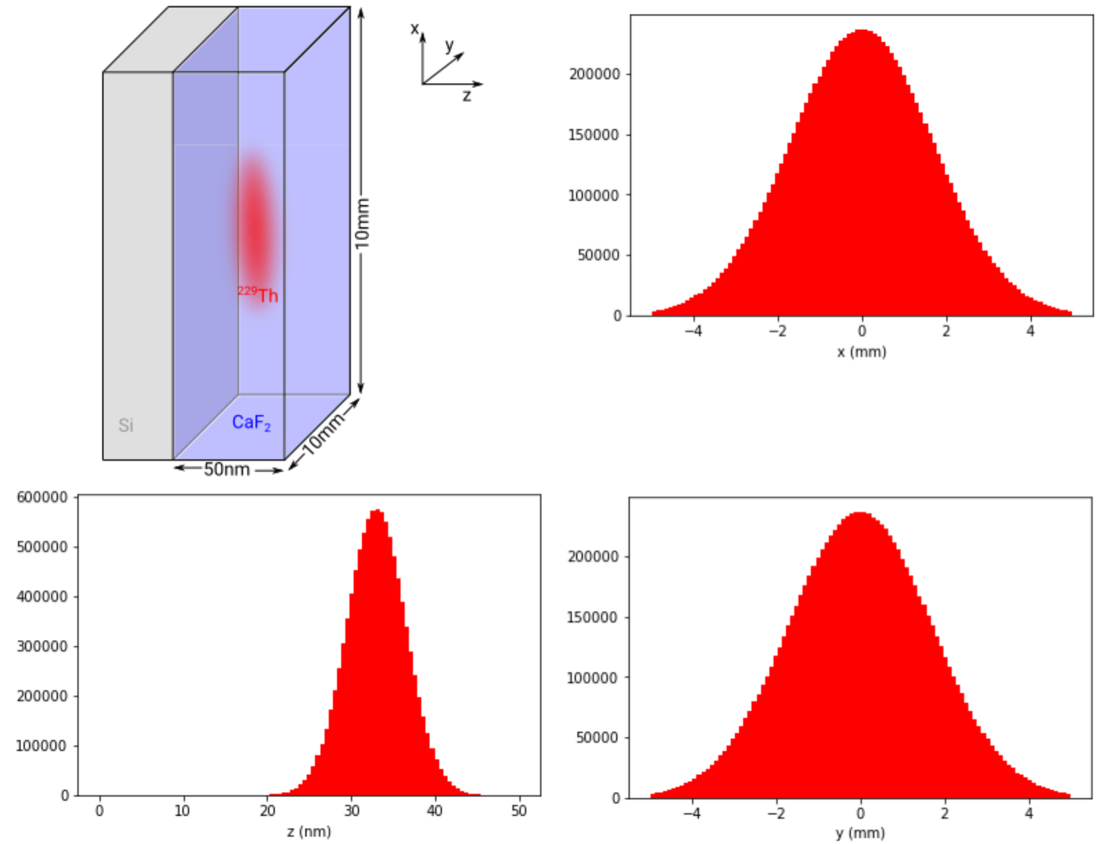
substitutional



Interstitial with O^- charge compensation

Implantation and Photon Source





VUV spectroscopy background

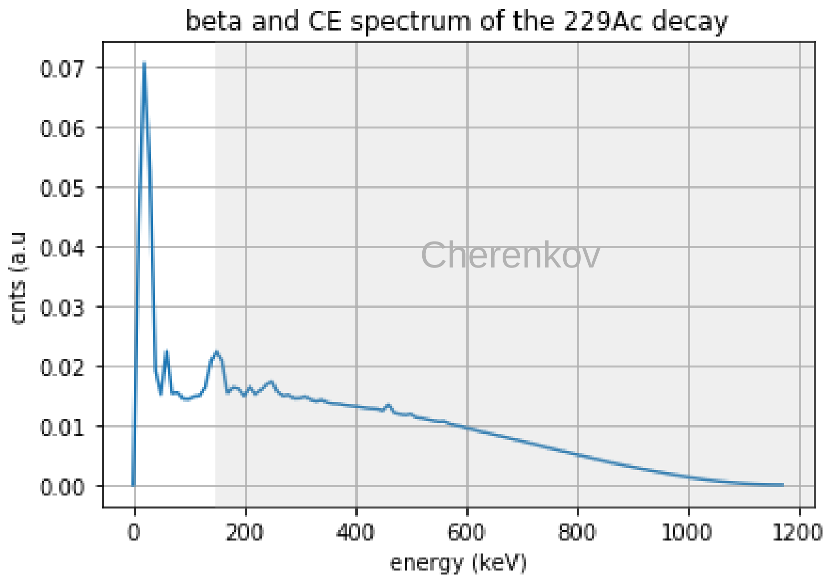
- Mean energy deposit in crystal based on
  - stopping power/linear attenuation
  - travelled path in crystal (for isotropic emission)

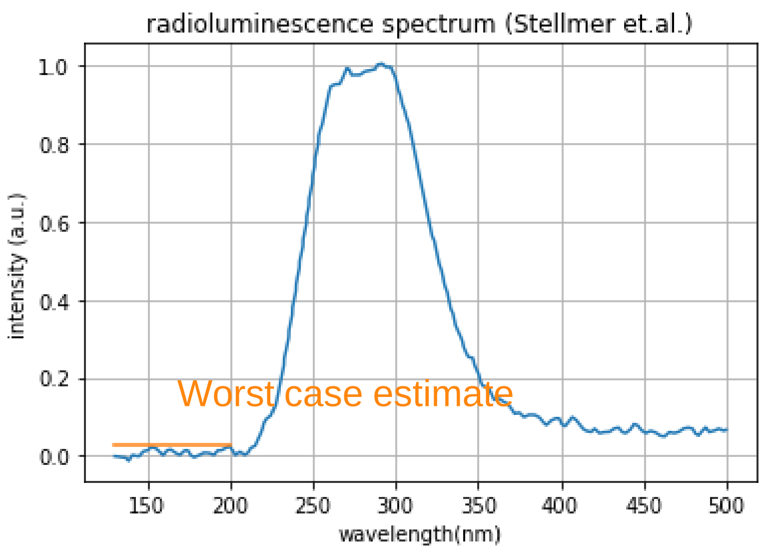
contribution	Energy deposit per β decay
γ	0.25eV
Xray	2.6eV
β	137eV
CE	71eV
α	31keV (per α)

(from center of 50nm x 1cm x 1cm crystal)

- Conversion deposited energy to photon:
  α,β: $\sim 1\%$
  γ: 100%

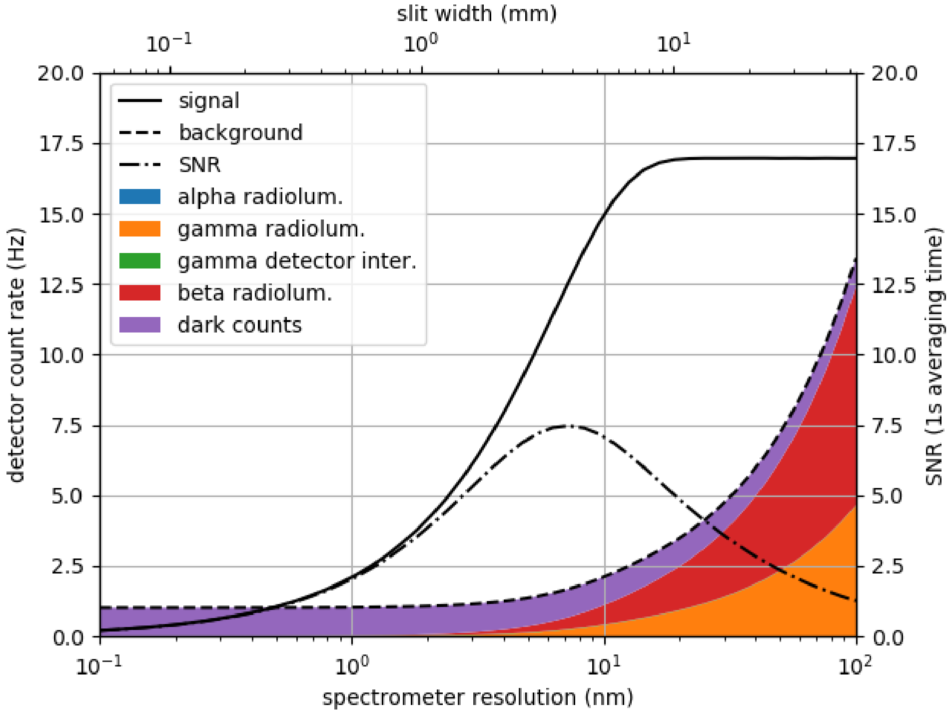
- Spectral contribution around 150nm:
  worst-case estimate from instrumental noise in measurement performed at TU Vienna





Background

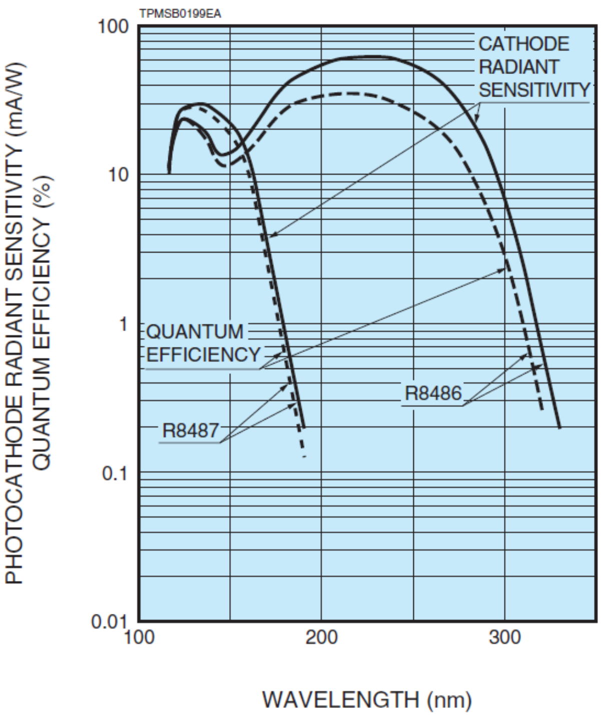
Contribution	Mean activity/rate
β activity	318 kBq
isomer activity	21 kBq
collected photons at detector	10.5 Hz
detected isomer VUV photons	**2.1 Hz**
gamma interactions in PMT detector	17 mHz
detected crystal γ radioluminescence	<5.7 mHz
detected crystal β radioluminescence	1.3 mHz
detected crystal α radioluminescence	30 nHz
radiation induced counts at 150 ± 0.5 nm	**0.024 Hz**
detector dark counts at 300 K	**1 Hz**



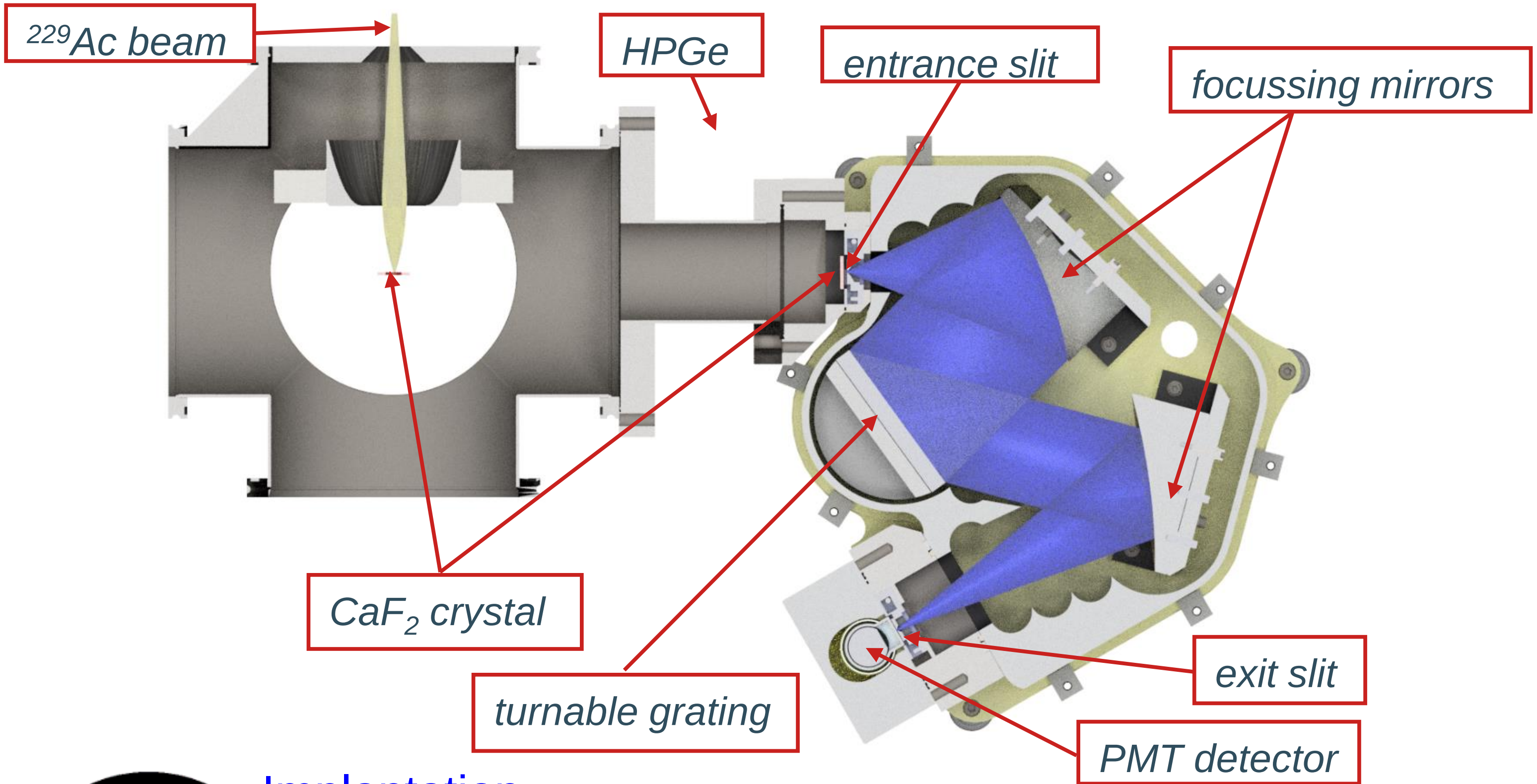
10^6 implantation rate (pps), 2 hrs isomer half-life, 3 h measurement

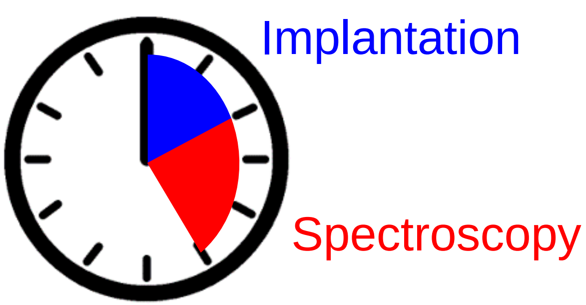
VUV spectrometer

contribution	note	Efficiency
Transmission through entrance slit	Beam: 4mm FWHM Gaussian	12.3%
Solid angle spectrometer	Corresponding to F/1.2	3.85%
Reflectivity grating+mirrors	into 1st order	38%
Quantum efficiency PMT detector	@ 150nm	20%
Total:		0.035%
Proposal:	(Conservative estimate)	0.01%



Spectroscopy of the Radiative Decay: Methodology





- Implantation into thin (50nm) CaF_2 crystal on Si backing
- Implantation time: 2 half-lifes
- Transfer of crystal under vacuum to spectrometer
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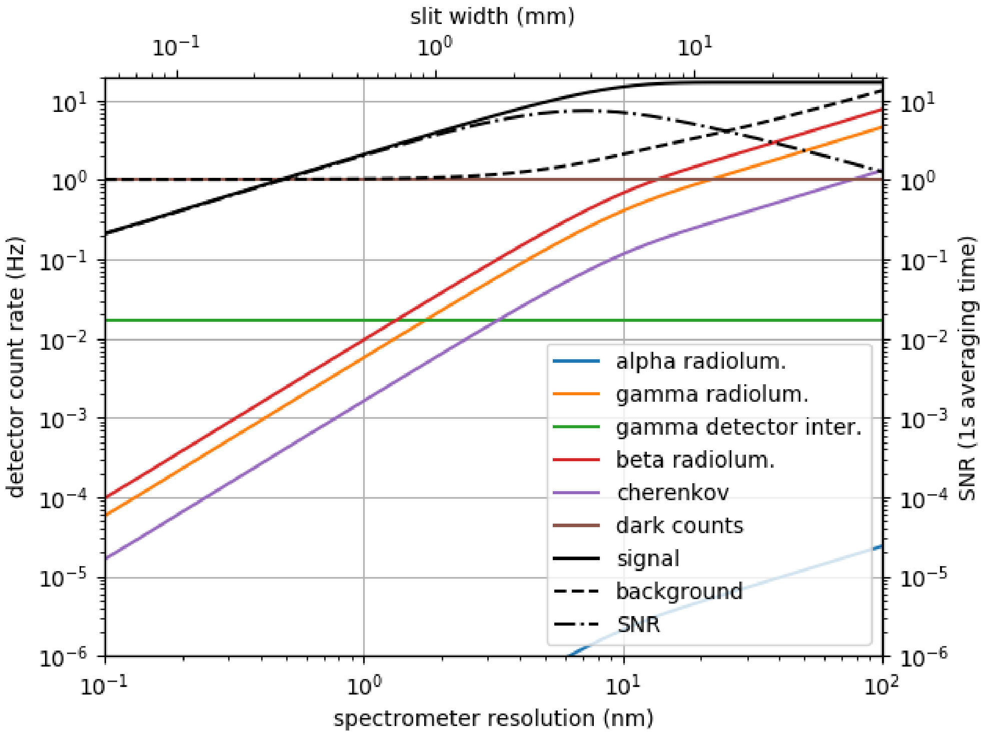
(11) Stellmer *et al.* 2015 *Scient.Rep.* **5** 1

Signal and background contributions for $3h$ measurment at $10^6 pps$ implantation and $2h$ isomer half-life:



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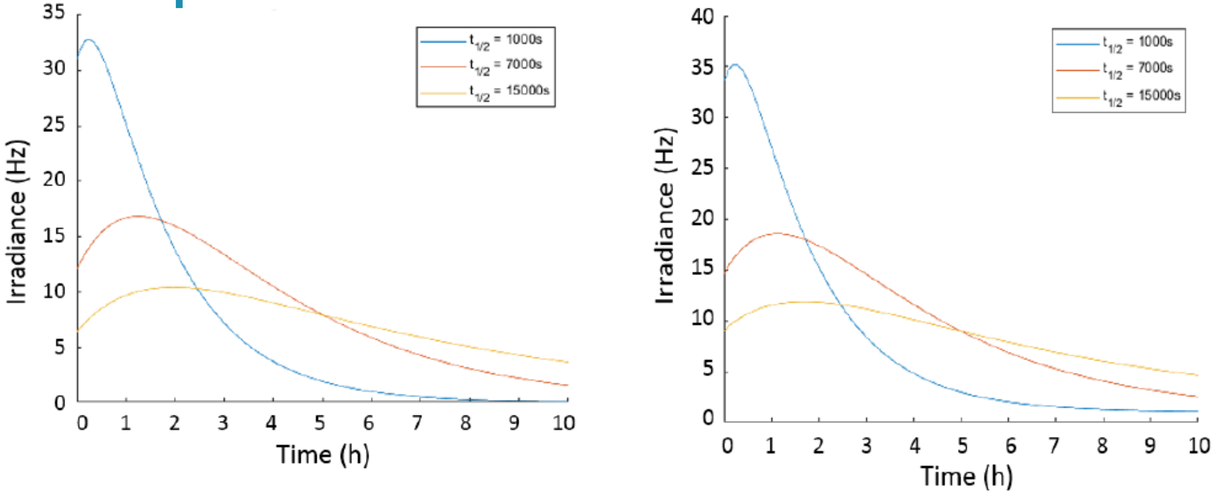
Dependance on the isomer’s half life



(a) VUV-signal strength (b) Total signal (VUV + background)

Figure 5.9: Influence of the ^{229}Th isomeric half-life on the VUV-signal strength and total signal (VUV + background). For a 125 min implantation time with a beam intensity of 10^6 pps.

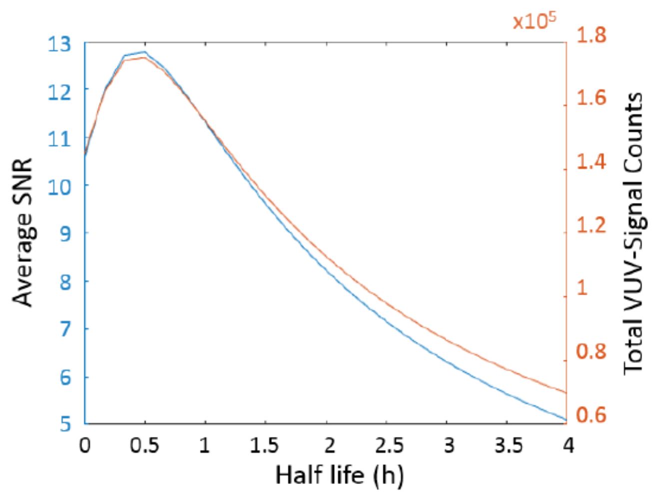
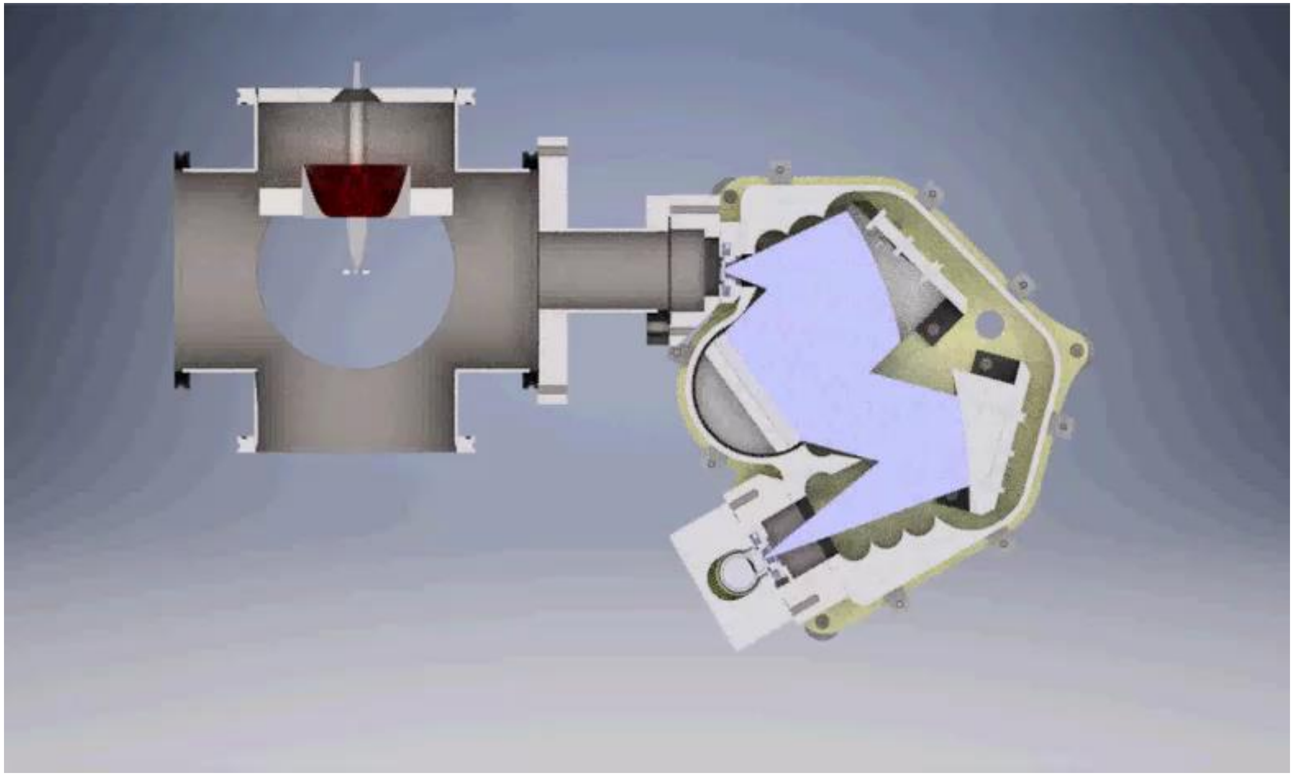
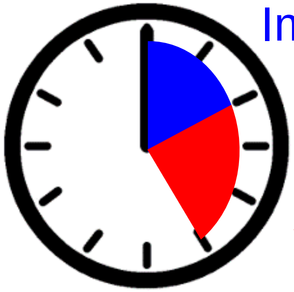


Figure 5.10: (left) Influence of the half-life on the average SNR using a 2hr measurement time. (right) Total VUV-signal counts during a 2hr measurement. For a 125 min implantation time with a beam intensity of 10^6 pps.

Spectroscopy of the Radiative Decay: Methodology





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Preparatory study at ISOLDE: LoI198

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  ^{229}Ac beam at LA1: $8.8(2) \cdot 10^5 pps$

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  ^{229}Ac beam retarded to 2keV and implanted in Nb and Au foils
  Low-energy electron detection based on a Channeltron detector
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- Emission channeling of ^{229}Ac implanted in CaF_2
  → 90% of ^{229}Ac in substitutional position (preliminary)