Study of Cs_2MCl_6 family (M = Hf or Zr) crystal scintillators in the search for rare processes in Hf and Zr isotopes

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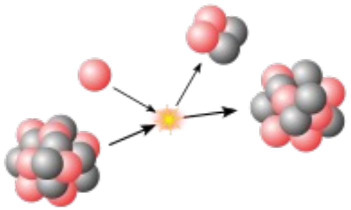
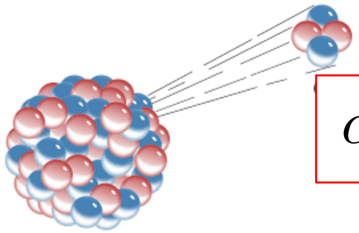
Interest in studying the 2β and rare α decay



$$0\nu2\beta^- : \ {}^{A}_{Z}X \rightarrow \ {}^{A}_{Z+2}Y + 2e^-$$



$$\alpha : \ {}^{A}_{Z}X \rightarrow {}^{A}_{Z-2}Y + {}^{4}_{2}He$$

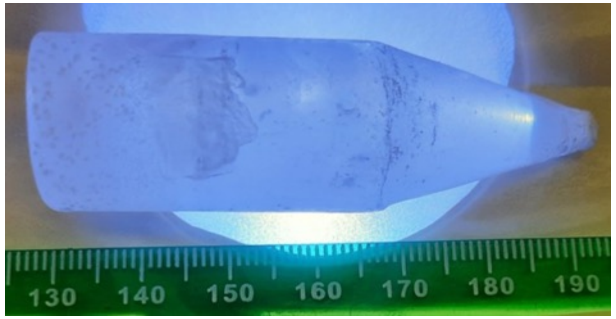
- ❖ **Neutrino physics**: Provides insights into the properties of neutrinos, including their mass and whether they are Majorana particles (particles that are their own antiparticles).
- ❖ **To test,** e.g., **calculations of different nuclear shapes** and the **decay modes** that involve the vector and **axial-vector g_A weak effective coupling constants**.
- ❖ **Beyond Standard Model**: Helps test theories beyond the Standard Model of particle physics, such as the presence of new particles or interactions.

- ❖ **Nuclear structure**: Provides detailed information about the structure and stability of atomic nuclei.
- ❖ **Nuclear Models**: Helps refine theoretical nuclear models by providing empirical data on rare decay processes.
- ❖ **Astrophysics**: essential also for nuclear and particle astrophysics studies (α–capture reactions, β–delayed fission, nucleosynthesis).

Some general properties	Cs_2HfCl_6	Cs_2ZrCl_6
Effective atomic number	58	46.6
Density (g/cm^3)	3.9	3.4
Melting point (°C)	820	850
Crystal structure	Cubic	Cubic
Emission maximum (nm)	400 - 430	450 - 470
Scintillation time constants (μs)	0.4; 5.1; 15.2 *	0.4; 2.7; 12.5*
Light Yield	up to 30000 photons/MeV**	up to 41000 photons/MeV**
Linearity of the energy response	Excellent, down to 100 keV	Excellent, down to 100 keV
Energy resolution (FWHM, %) @ 662 keV	3.2 - 3.7***	3.5 - 7.0***
Pulse-shape discrimination ability	Excellent	Excellent
Mass fraction of isotope of interest (%)	27	16

The Cs_2HfCl_6 (CHC) and Cs_2ZrCl_6 (CZC) crystal scintillators

Produced at Queen's University



* *for alpha events at room temperature (Dalton Trans. 2022, 51, 6944-6954)*

** *for gamma quanta at room temperature*

*** *depends on the crystal quality, surface treatment and readout system*

Low background measurements of the CHC and CZC crystals

measured with the ultra-low background **HP-Ge** γ spectrometers of the **STELLA** facility at LNGS.

Chain	Nuclide	Activity (mBq/kg)		
		CHC	CZC [2]	
			Cone	Cylinder
		16.87 g	10.63 g	23.95 g
^{238}U	^{226}Ra	<13	60(10)	< 8.7
	^{234}Th	<1200	< 180	< 260
	^{234m}Pa	<18	< 630	< 160
^{235}U	^{235}U	<18	< 16	< 12
^{232}Th	^{228}Ra	<13	< 16	< 23
	^{228}Th	<17	< 6.7	< 8.2
	^{137}Cs	<10	< 7.1	< 1.6
	^{134}Cs	37(4)	49(6)	42(5)
	^{132}Cs	-	< 8.2	< 11
	40K	<240	<120	<95

Maybe a cross-contamination happened during the sample preparation and installation.

Natural

Artificial

Cosmogenic activation

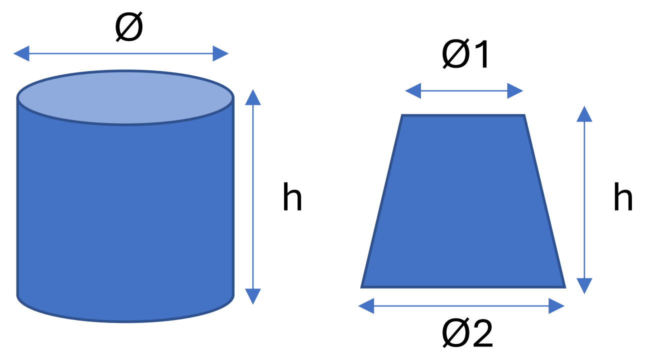
Only land transportation! $T_{1/2} \approx 2$ years

[2] P. Belli et al. Eur. Phys. J. A **59**, 176 (2023).

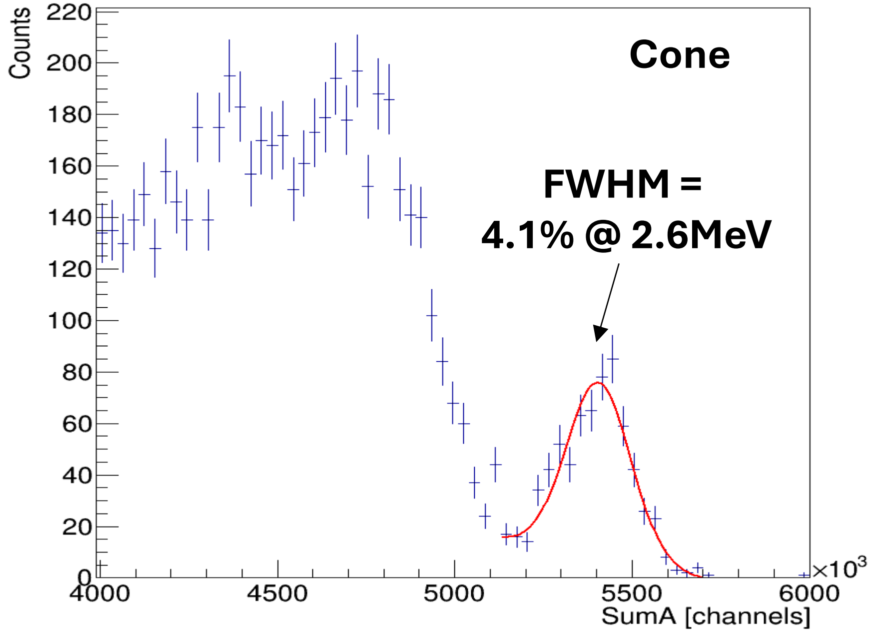
P. Belli et al. Eur. Phys. J. A **59**, 176 (2023).

Search for 2β decay in $^{94,96}Zr$ using CZC crystal scintillators

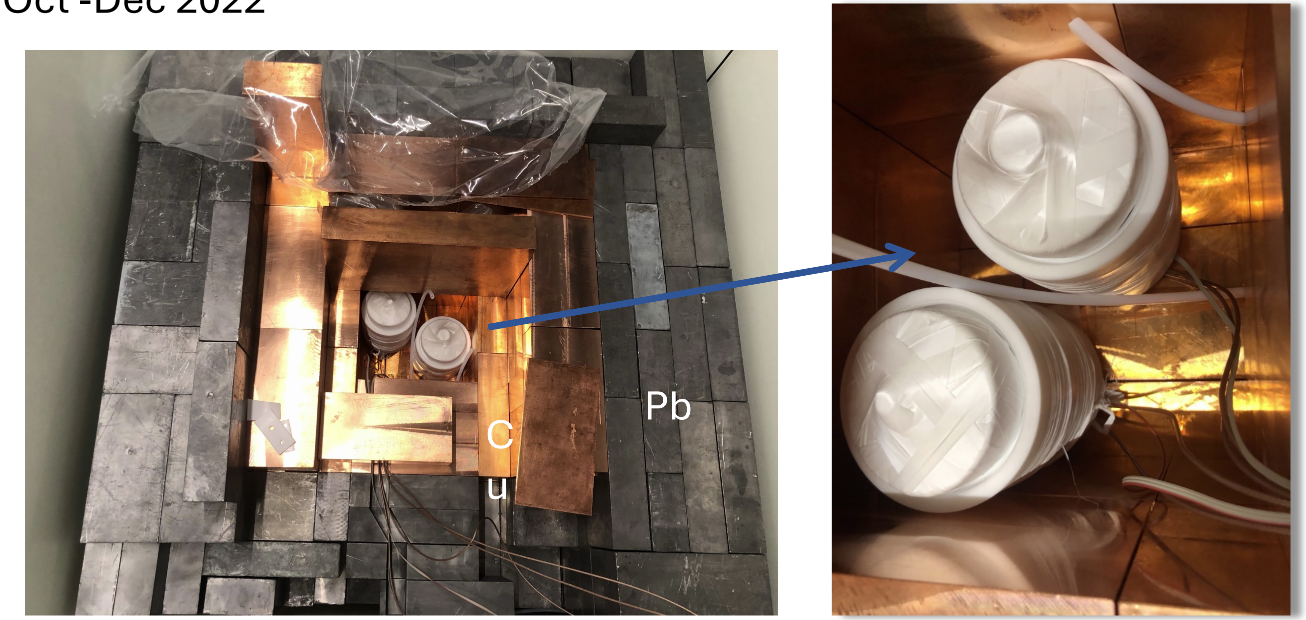
M=24,0(1) g
h =21,20(5) mm
Ø=21,00(5) mm

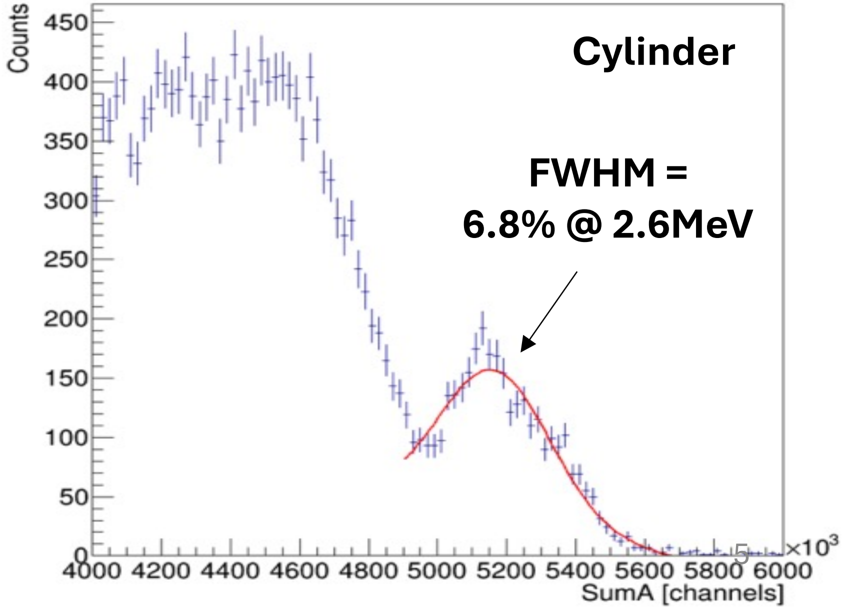


M=10,6(1) g
h =17,90(5) mm
Ø1=8,0(1) mm
Ø2=19,70(5) mm



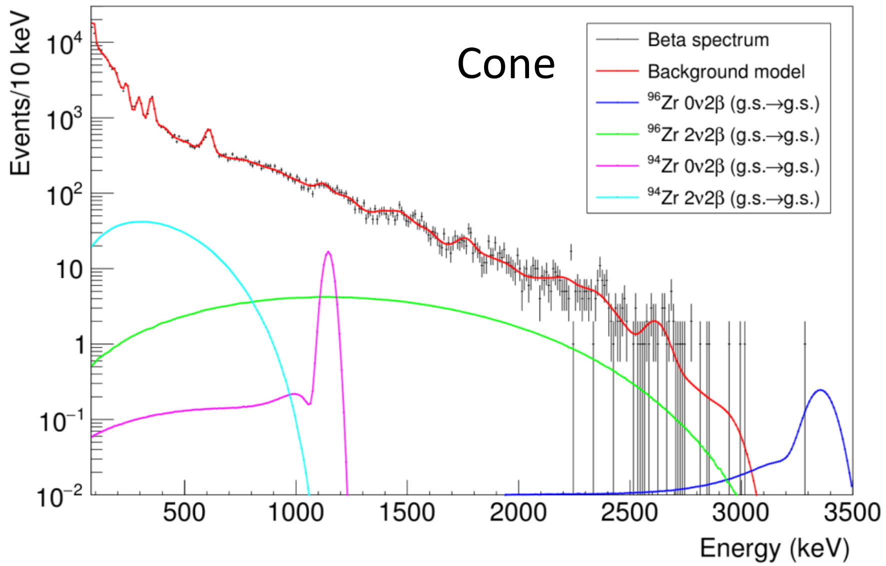
- ✓ Run 1: 456.5 days of data taking (time-window 80 μs), July 2021 - Oct 2022
- ✓ Run 2: 65 days of data taking (extended time-window for t-A analysis, 2 ms), Oct -Dec 2022

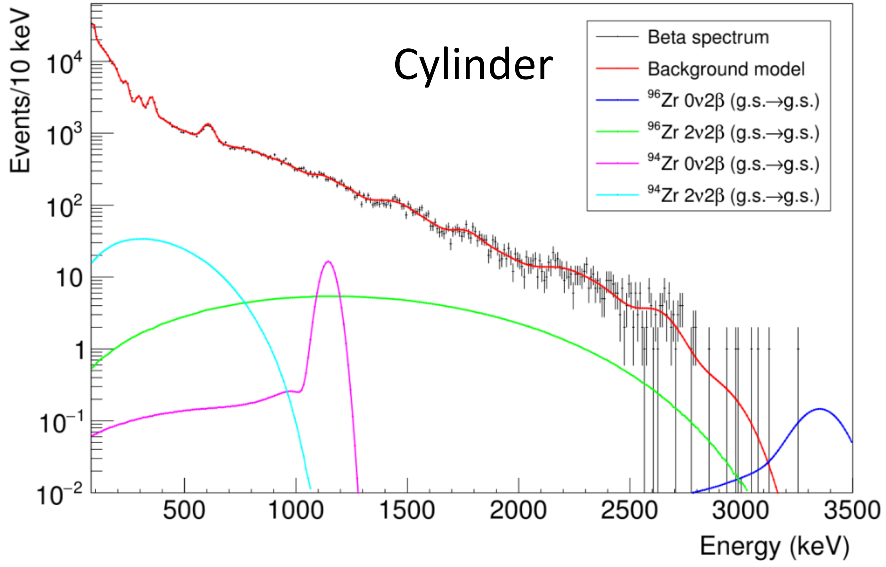




DAMA/CRYS setup at LNGS

Experimental limits on various decay modes in ^{94,96}Zr isotopes



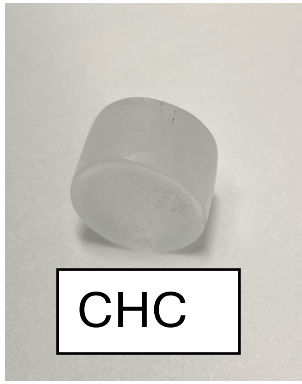


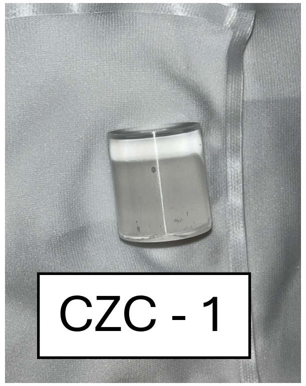
Transition	Decay mode	Final state of daughter nucleus, keV	Experimental limit on $T_{1/2}$ at 90%C.L., yr
^{96}Zr → ^{96}Mo	0ν2β	g.s.	**> 1.5×10^{20}**
		2_1^+, 778	> 1.5×10^{19}
	2ν2β	g.s.	> 7.4×10^{17}
		2_1^+, 778	> 3.8×10^{17}
	β	g.s.	> 1.0×10^{17}
^{94}Zr → ^{94}Mo	0ν2β	g.s.	**> 2.6×10^{19}**
		2_1^+, 871	> 3.8×10^{18}
	2ν2β	g.s.	**> 2.4×10^{18}**
		2_1^+, 871	> 1.9×10^{17}

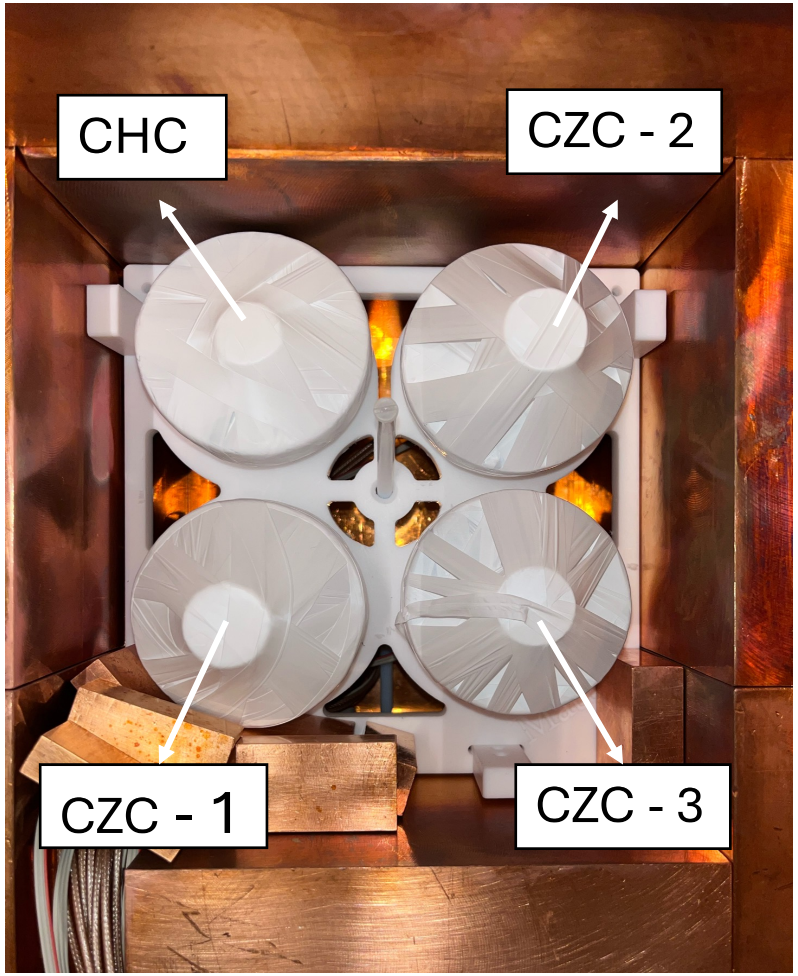
See more details in ***Eur. Phys. J. A 59 (2023) 176***
https://doi.org/10.1140/epja/s10050-023-01090-9

P. Belli *et al* 2024 *JINST* **19** P05037

New low-background measurements in DAMA/CRYS setup (LNGS)



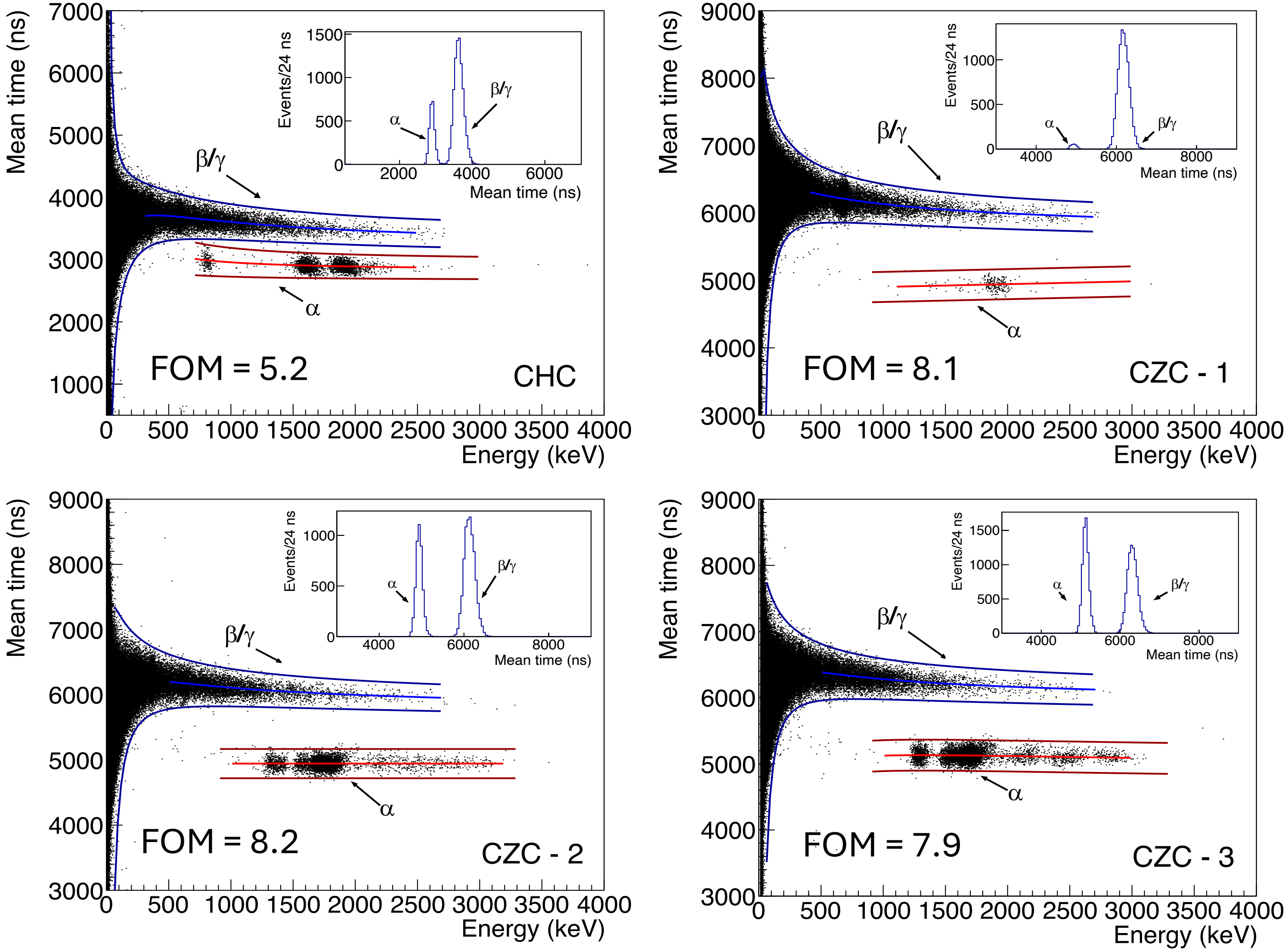




DAMA/CRYS setup at LNGS

- ✓ Three new Cs_2ZrCl_6 crystals + one Cs_2HfCl_6
- ✓ Total mass of 3 CZC = 59.5 g, mass of CHC= 16.87 g.
- ✓ FWHM = 6-8% @ 662keV
- ✓ Produced from high purity and purified raw materials (> 99.99%)
- ✓ CZC crystals are encapsulated in a silicon-base resin + quartz window
- ✓ Modified experimental setup
- ✓ Measurements started on June 30th, 2023, for a total of 97.7 days live time

Pulse Shape Discrimination (PSD) ability



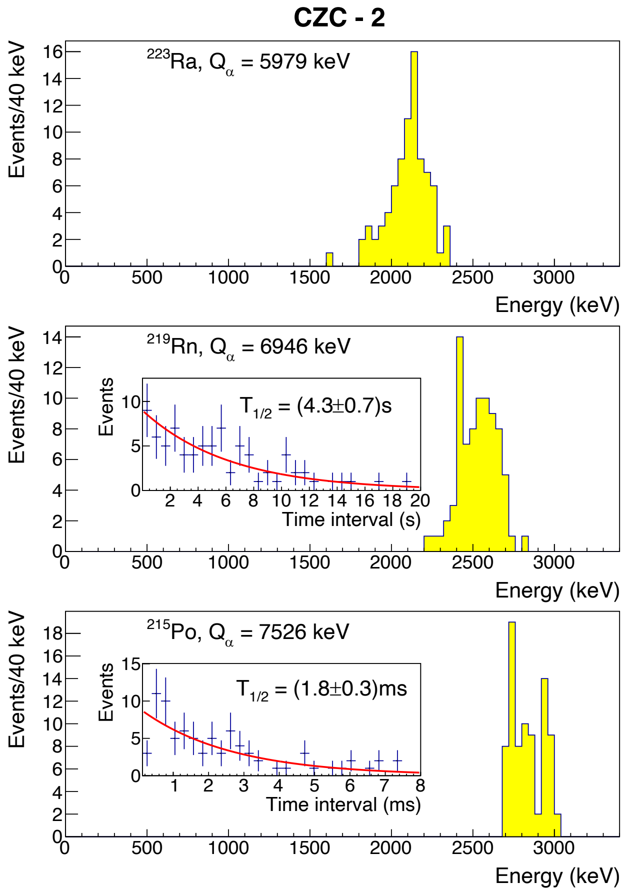
The difference in scintillation pulse time profile for different type of particles allows for an effective pulse-shape discrimination.

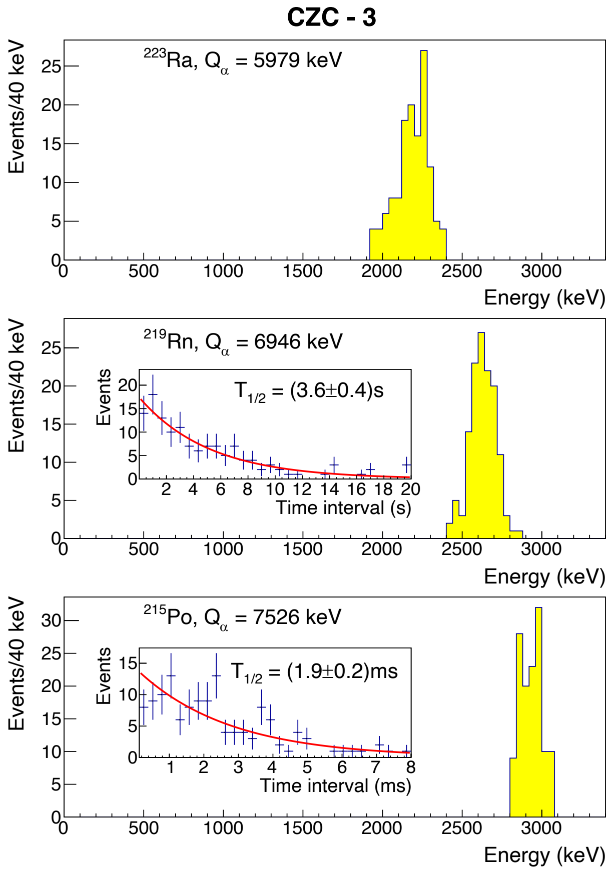
The "mean-time" ($\langle t \rangle$) method *[L.Bardelli et al., Nucl. Instr. Meth. A **584** (2008) 129]*. was used, and this parameter was determined according to:

$$\langle t \rangle = \sum f(t_k) t_k \Big/ \sum f(t_k)$$

where the sum is over the time channels (*k*), starting from the origin of pulse up to 24 μs, *f(t)* is the digitized amplitude (at the time *t*) of a given signal.

Time-Amplitude analysis





To select the sequence of alpha events in ^{235}U sub-chain:

^{223}Ra (Q_α = 5979 keV, $T_{1/2}$ = 11.44 d)

↓

^{219}Rn (Q_α = 6946 keV, $T_{1/2}$ = 3.96 s)

↓

^{215}Po (Q_α = 7526 keV, $T_{1/2}$ = 1.782 ms)

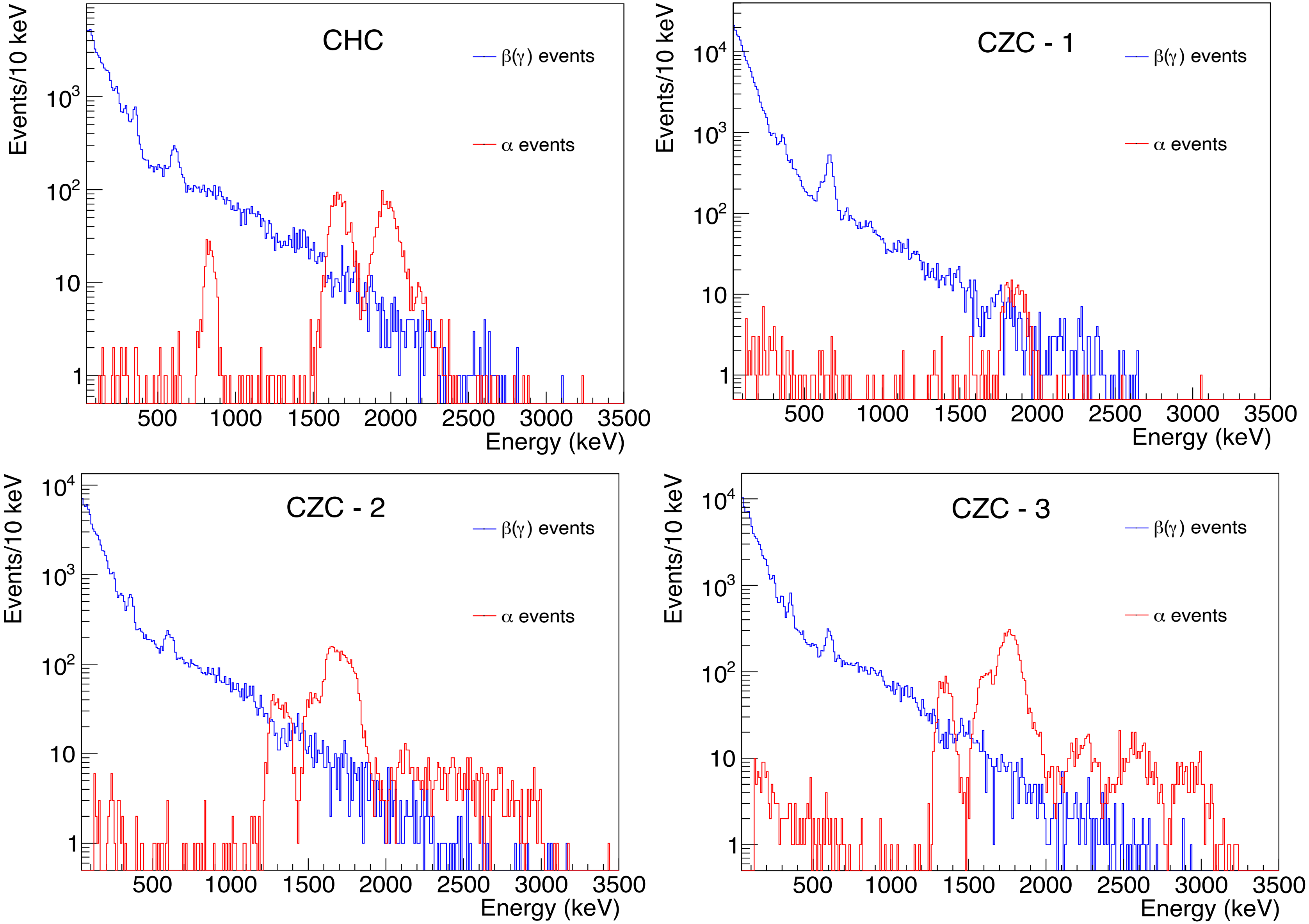
↓

^{211}Pb

A(^{227}Ac) < 0.020 mBq/kg in CHC
< 0.017 mBq/kg in CZC - 1
= 0.56(6) mBq/kg in CZC -2
= 0.88(8) mBq/kg in CZC - 3

+ *Confirmation of ^{235}U decay chain presence*
+ *Alpha peaks to precisely determine α/β ratio*

Measured energy spectra of the 4 crystals



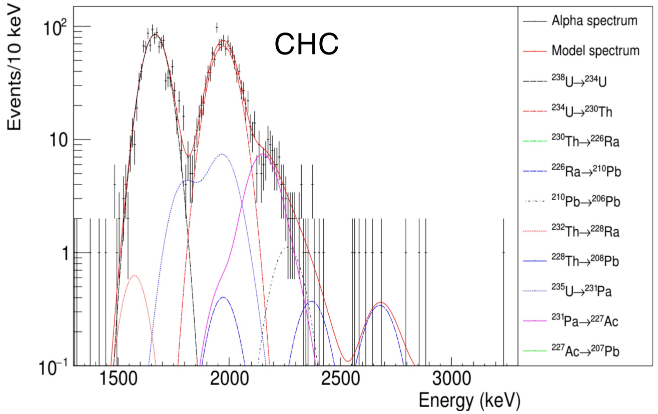
- ✓ Live time: 97.7 days
- ✓ Counting rates:

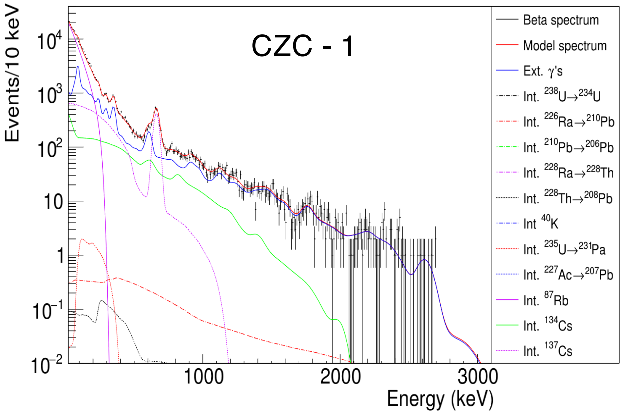
	CHC	CZC - 1	CZC - 2	CZC - 3
α/kg/day	1616	173	2302	3548
β/kg/day	46117	104303	43251	52292

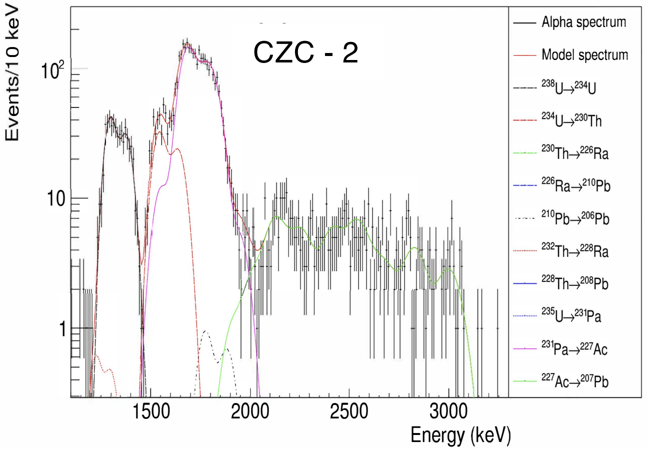
Background model

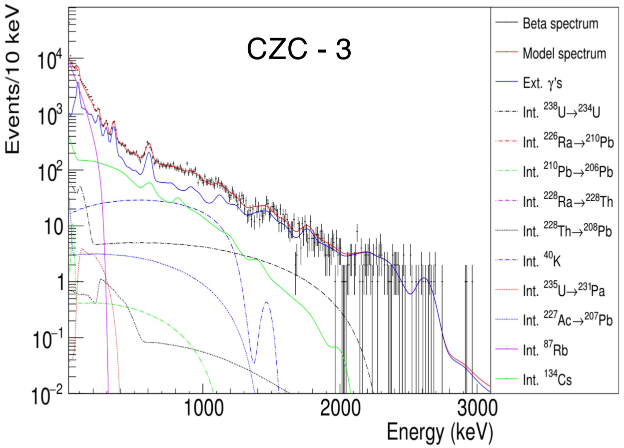
Alpha events

Beta/gamma events









Chain	Nuclide	Internal contamination, mBq/kg			
		CHC	CZC - 1	CZC -2	CZC – 3
^{238}U	^{238}U	7.6(3)	< 0.08	3.16(14)	4.58(18)
	^{234}U	6.7(5)	< 0.12	2.86(22)	4.20(33)
	^{230}Th	< 0.50	< 0.12	< 0.28	< 0.6
	^{226}Ra	0.04(2)	< 0.05	< 0.06	< 0.17
	^{210}Pb	0.12(7)	< 1.3	< 0.6	1.32(38)
^{235}U	^{235}U	1.3(5)	< 0.14	< 0.16	< 0.37
	^{231}Pa	0.92(13)	< 1.3	16.95(48)	24.69(56)
	^{227}Ac	< 0.005	< 0.013	0.62(3)	0.94(6)
^{232}Th	^{232}Th	< 0.22	< 0.10	< 0.12	< 0.12
	^{228}Th	< 0.02	< 0.011	< 0.04	< 0.16
	^{147}Sm	0.25(10)	-	-	-
	^{137}Cs	-	100(3)	-	-
	^{134}Cs	44(8)	58(6)	42(7)	55(7)
	87Rb	< 400	1067(5)	318(14)	441(9)
	^{40}K	< 2.3	< 1.1	11(2)	17(3)

Contribution of external gammas from PMT's is dominant

Search for the α decay of ^{174}Hf to the g.s. of ^{174}Yb

^{147}Sm ($E_\alpha = 2249.9$ keV) + ^{174}Hf ($E_\alpha = 2437.6$ keV)

Fit with 2 gaussians + pol1

Parameters of the fit:

First peak

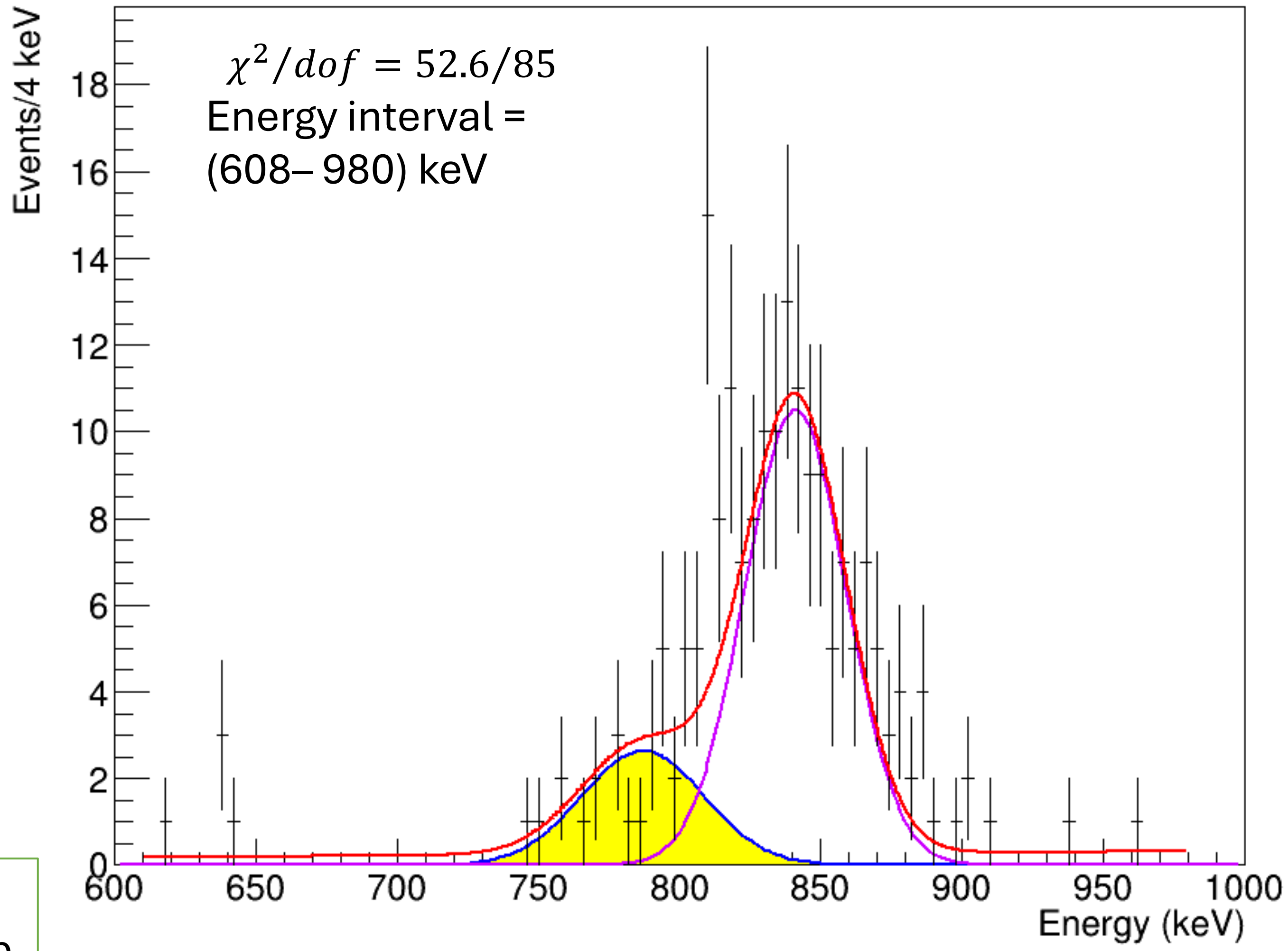
Area = (36 ± 15) counts

Peak position = (787 ± 18) keV

Second peak

Area = (118 ± 37) counts

Peak position = (841 ± 3) keV



Q.F. $= 0.350 \pm 0.008$ for ^{147}Sm

Q.F. $= 0.345 \pm 0.001$ for ^{174}Hf

Activity of ^{147}Sm = (0.25 ± 0.10) mBq/kg, corresponding to a concentration of (2.0 ± 0.8) ppb of ^{nat}Sm, in agreement with ICP-MS measurements.

Half-life of α decay of ^{174}Hf to the g.s. of ^{174}Yb

Area of 2nd peak = $118 \pm 11(\mathrm{stat}) \pm 35\ (sys) = 118 \pm 37$

Half-life:

$$T_{1/2} = \ln 2 \cdot N \cdot \epsilon \cdot t/S$$

M = 16.87 g;
W(Cs_2HfCl_6) = 657 g/mole;
δ(^{174}Hf) = 0.156(6) %

- N (number of nuclides) = $\frac{\mathrm{M}}{\mathrm{W}} \cdot \delta \cdot \mathrm{N_A} = 2.412 \times 10^{19}$
- ϵ is the PSD efficiency which corresponds to 99%;
- t is the measurement time (= 2344.8 h = 0.26767 yr);

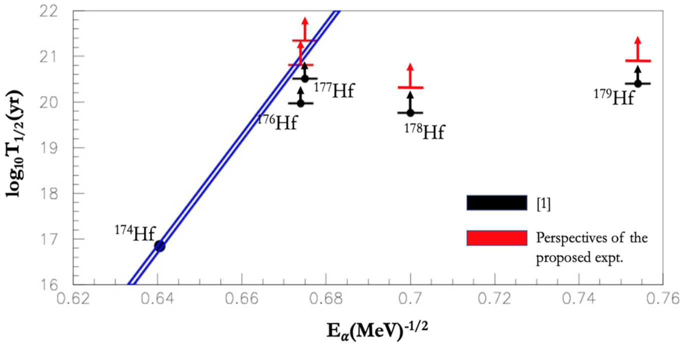
$\Longrightarrow T_{1/2} = [3.8^{+0.4}_{-0.3}(stat)^{+1.6}_{-0.9}(sys)] \times 10^{16} = 3.8^{+1.7}_{-0.9} \times 10^{16}$ yr of α decay of ^{174}Hf

Comparing with result in [*NPA 1002 (2020) 121941*]: $\frac{|3.8-7.0|}{\sqrt{(1.7)^2 + (1.2)^2}} = 1.5$

Theoretical predictions: $(3.5 - 7.4) \times 10^{16}\ yr$.

Perspectives and conclusions

Diagram $T_{1/2}$ vs the inverse of the square root of α energy in MeV.



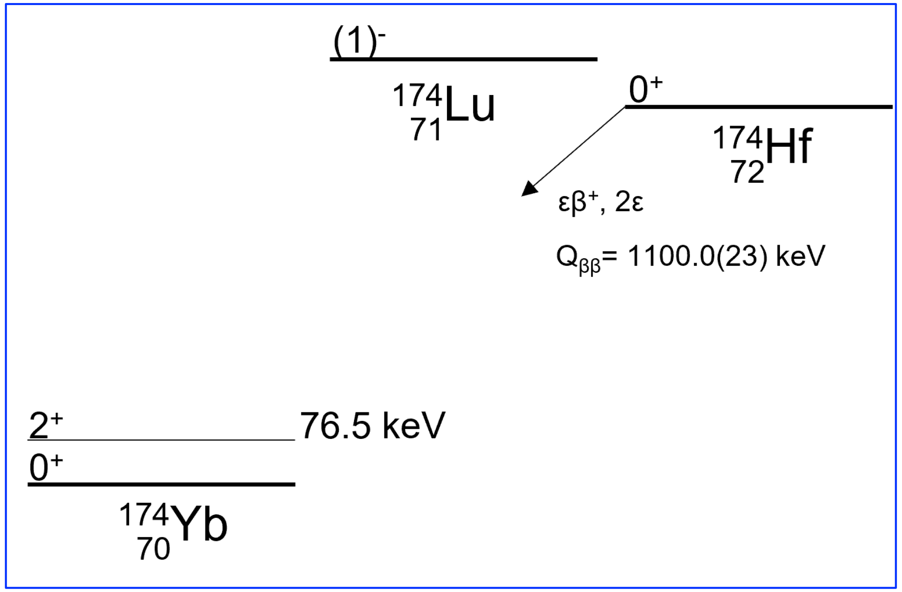
- The blue band is the extrapolation of the predictions on $T_{1/2}$ for all the Hf isotopes using the Geiger-Nuttall scaling law considering the data point observed in Ref. **[1]NPA 1002 (2020) 121941.**
- The red symbols represent the sensitivity that the measurement can reach using CHC crystal scintillators with 43.83 kg × day of total exposure.

- First two Cs_2ZrCl_6 scintillating crystals have been grown in Queen's University and studied at the LNGS, Italy to search for 2β decay of $^{94,96}Zr$ isotopes [P. Belli et al. Eur. Phys. J. A **59**, 176 (2023)].
- An experiment using a CHC crystal scintillator and three CZC crystal scintillators has been performed in the DAMA/CRYS setup at LNGS [P. Belli *et al* 2024 *JINST* **19** P05037].
- It has observed α decay of ^{174}Hf to the ground state with a **$T_{1/2}$ = $3.8^{+1.7}_{-0.9} \times 10^{16}$ yr.** This value is in good agreement with the theoretical predictions.
- A new experiment is ongoing with 4 CHC crystals (∅ 26 ×20 mm^3) encapsulated in silicone-base sealeant.
- We are hoping in 1 year of data taking to improve limits also for **2ε** and **$\varepsilon\beta^+$** decay of ^{174}Hf and α decay in the naturally occurring Hf isotopes.

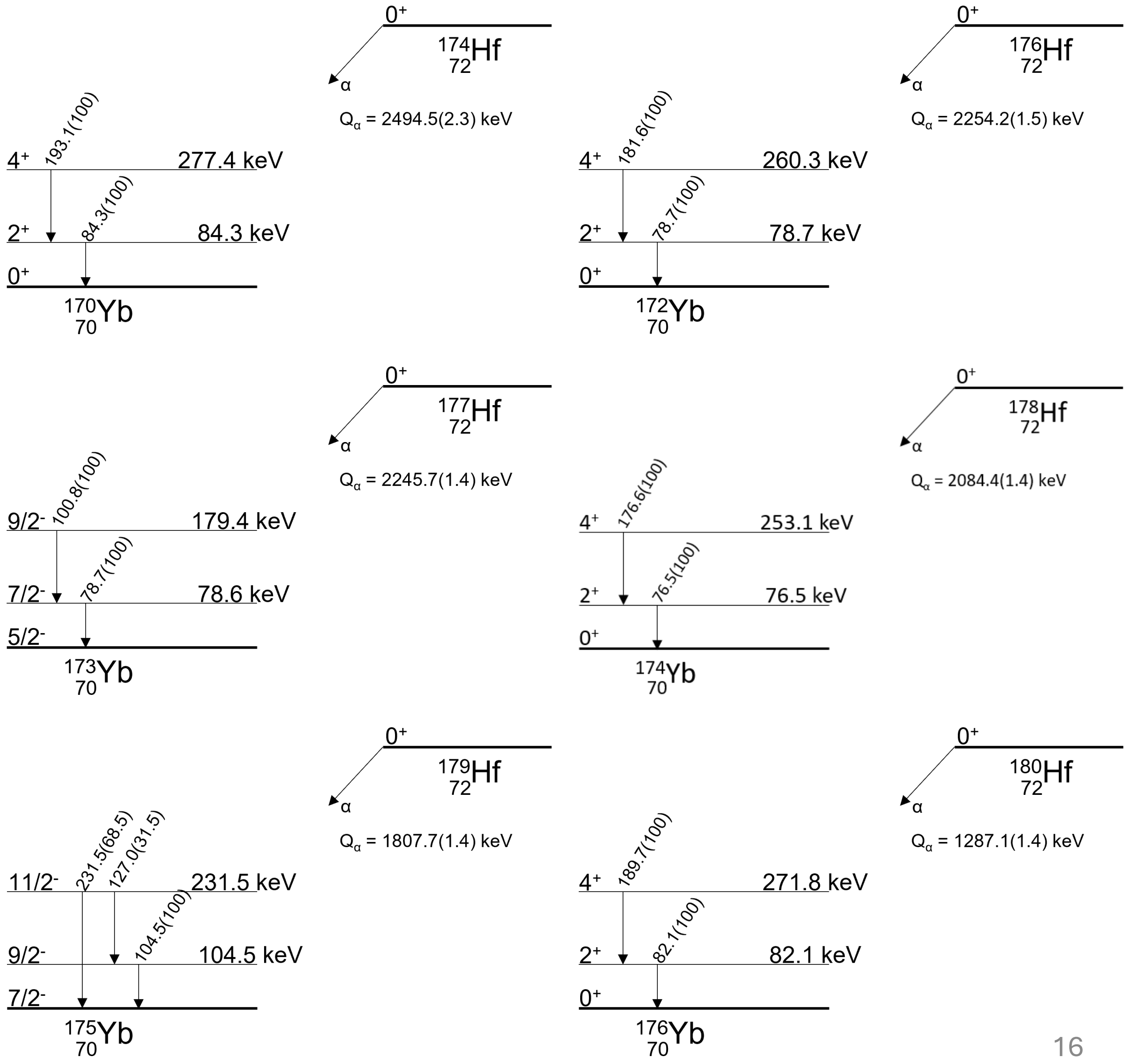
BACKUP SLIDES

Simplified decay schemes of naturally occurring Hf isotopes

2ε, εβ$^+$ decay of ^{174}Hf



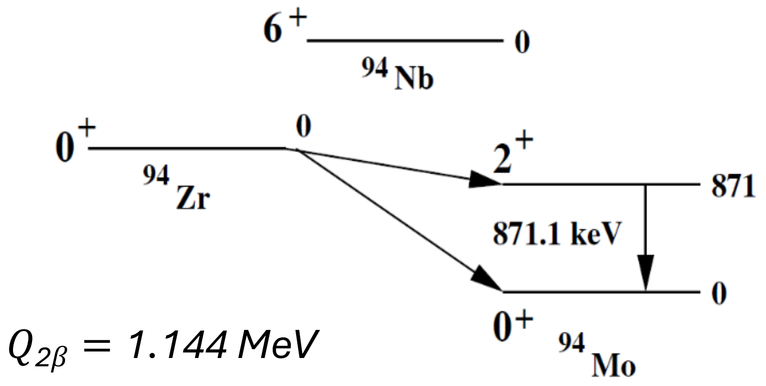
α decays of Hf isotopes considering the first two excited energy levels of the daughter nuclei. Energies of the excited levels and of the emitted γ quanta are shown. Relative probabilities of a single energy level are given in parentheses. The ^{175}Yb isotope decays via β$^-$ with $T_{1/2}$ = 4.185(1) d, while all the other Yb nuclei are stable.



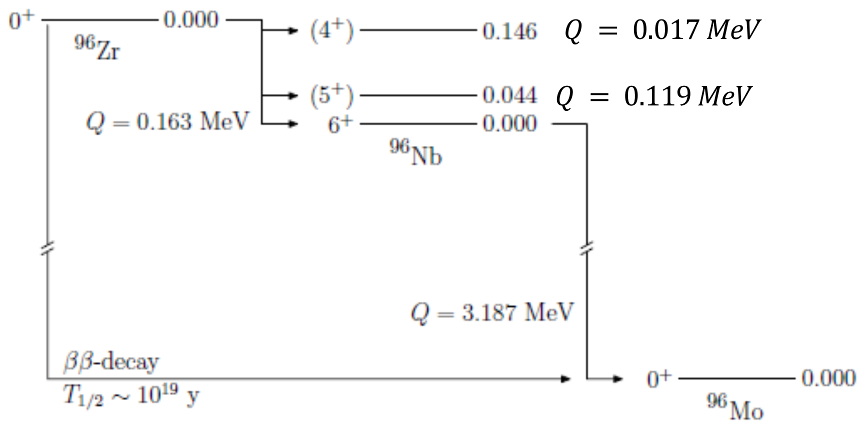
Search for 2β decay in ^{94,96}Zr and for ^{96}Zr's β decay

Experiment	Transition	$T_{1/2}$ 90% C.L. (y)	Technique	Ref.
ZICOS, (Kamioka Observatory, Japan)	$^{96}Zr\ 0^+ \rightarrow {}^{96}Mo\ 0^+_1$ (g.s.)	under construction	Organic liquid scintillator	[1]
NEMO I, II, III, Frejus (France) **(next: SuperNEMO)**	$^{96}Zr\ 0^+ \rightarrow {}^{96}Mo\ 0^+_1$ (g.s.)	$>9.2\times10^{21}$ $>1.29\times10^{22}$	Tracker detector	[2] [3]
Kimballton Underground Research Facility, (USA)	$^{96}Zr\ 0^+ \rightarrow {}^{96}Mo\ 2^+_1$	$>3.1\times10^{20}$	HP-Ge	[4]
Collaboration at Fréjus Underground Laboratory	$^{96}Zr\ 0^+ \rightarrow {}^{96}Mo\ 2^+_1$, 0^+_1, 2^+_2, 2^+_3	$>(2.6-7.9)\times10^{19}$	HP-Ge	[5]
Collaboration at LNGS	$^{96}Zr\ 0^+ \rightarrow {}^{96}Mo\ 2^+_1$	$>3.8\times10^{19}$	HP-Ge	[6]
TILES (TIFR, Mumbai)	$^{94}Zr\ 0^+ \rightarrow {}^{94}Mo\ 2^+_1$	$>5.2\times10^{19}$	HP-Ge	[7]
Kimballton Underground Research Facility, (USA)	$^{96}Zr\ 0^+ \rightarrow {}^{96}Mo\ 6^+$	$>2.4\times10^{19}$	HP-Ge	[8]

Decay scheme of ^{94}Zr



β and 2β decay of ^{96}Zr. The decay Q-values and excitation energies of the first three states of Nb are also indicated.



[1] EPS-HEP (2019) 437
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