

# 108° CONGRESSO NAZIONALE



Milano, 12-16 settembre 2022

## Studio di processi rari negli isotopi naturali di Hf e Zr utilizzando cristalli scintillatori di $\text{Cs}_2\text{HfCl}_6$ e $\text{Cs}_2\text{ZrCl}_6$



UNIVERSITÀ  
DEGLI STUDI  
DI MILANO  
DIPARTIMENTO DI FISICA



POLITECNICO  
MILANO 1863



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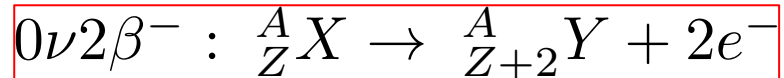
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# Interest in studying the $2\beta$ and rare $\alpha$ decay

- ❖  $2\beta$  decay without the presence of neutrinos could open a new window beyond the Standard Model.
- ❖ To test calculations of different nuclear shapes and the decay modes that involve the vector and axial-vector weak effective coupling constants; possible study of the “resonant effect” on the  $0\nu 2\epsilon$  mode.
- ❖ The nuclear matrix elements for the  $2\nu$  mode and for the  $0\nu$  mode can be related to each other through relevant parameters: in the free nucleon interaction, the  $g_A$  value is 1.2701, but, when considering a nuclear decay, there are indications that the phenomenological axial-vector coupling value is reduced at  $g_A < 1$ , more precisely:  $g_A \approx 1.269 A^{-0.18}$  or  $g_A \approx 1.269 A^{-0.12}$ , depending on the nuclear model adopted to infer the  $g_A$  value.
- ❖  $2\beta$  investigation with various nuclei would shed new light in constraining these and other important model-dependent parameters.
- ❖ The study of rare  $\alpha$  decay plays a crucial role in developing nuclear physics. It can offer details about the **nuclear structure, the nuclear levels and the properties of nuclei.**
- ❖ The phenomenon of  $\alpha$  decay can offer information about the fusion-fission reactions since the  $\alpha$  decay process involves sub-barrier penetration caused by the interaction between the  $\alpha$  particle and the nucleus. In particular the study of rare  $\alpha$  decay requires low background techniques and laboratories.
- ❖ Understanding the nuclear properties is essential also for nuclear and particle astrophysics studies, for example,  $\alpha$ -capture reactions (equivalent to the inverse  $\alpha$ -decay process) are important for nucleosynthesis and  $\beta$ -delayed fission, together with other fission modes, determine the so-called “fission recycling” in the r-process nucleosynthesis.



# The $\text{Cs}_2\text{HfCl}_6$ (CHC) and $\text{Cs}_2\text{ZrCl}_6$ (CZC) crystal scintillators

- High light yield (up to 30000 photons/MeV for CHC and up to 35000 photons/MeV for CZC);
- Excellent linearity in the energy response;
- Excellent energy resolution (<3.5% at 662 keV in the best configuration);
- Excellent ability for pulse shape discrimination (PSD) between  $\beta(\gamma)$  and  $\alpha$  particles;
- $\text{Cs}_2\text{ZrCl}_6$  crystal contains 16% Zr by weight.
- $\text{Cs}_2\text{HfCl}_6$  crystal contains a high fraction of Hf ( $\sim 27\%$  in mass).

Some general properties	CHC	CZC
Effective atomic number	58	46.6
Density (g/cm <sup>3</sup> )	3.9	3.4
Melting point (°C)	820	850
Crystal structure	cubic	cubic
Wavelength of emission (nm)	400-430	440-479
Decay time ( $\mu\text{s}$ )	0.4; 5.1; 15.2[1]	0.4; 2.7; 12.5[1]

[1] [Dalton Trans.](#),  
2022, **51**, 6944-  
6954

# Low background measurements of the CHC and CZC crystals

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

Chain		Nuclide	Activity (mBq/kg)		
			CHC [2]	CZC	
				Cone	Cylinder
			6.90 g	10.63 g	23.95 g
$^{232}\text{Th}$		$^{228}\text{Ra}$	<12	< 16	< 23
		$^{228}\text{Th}$	<3.6	< 6.7	< 8.2
$^{238}\text{U}$		$^{226}\text{Ra}$	<23	60(10)	< 8.7
		$^{234}\text{Th}$	<0.80	< 180	< 260
		$^{234\text{m}}\text{Pa}$	<0.48	< 630	< 160
$^{235}\text{U}$		$^{235}\text{U}$	<14	< 16	< 12
		$^{40}\text{K}$	$0.4(1)\times 10^3$	< 120	< 95
		$^{44}\text{Ti}$	10(4)	-	-
		$^{60}\text{Co}$	<25	-	-
		$^{137}\text{Cs}$	$0.78(8)\times 10^3$	< 7.1	< 1.6
		$^{134}\text{Cs}$	79(8)	49(6)	42(5)
		$^{132}\text{Cs}$	<15	< 8.2	< 11
		$^{181}\text{Hf}$	<11	-	-
		$^{190}\text{Pt}$	<20	-	-
		$^{202}\text{Pb}$	<9.1	-	-

Preliminary

Under investigation

Natural

Artificial

Cosmogenic activation

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

[2] NPA 1002 (2020) 121941

# Search for $2\beta$ and rare $\alpha$ decay in Hf isotopes using **passive** source approach

HPGe-detector

The isotope  $^{174}\text{Hf}$  is one of the potentially  $2\epsilon$ ,  $\epsilon\beta^+$  radioactive nuclides with the energy of decay  $Q_{2\beta}=1100.0(23)$  keV and the isotopic abundance  $\delta=0.156(6)\%$ .

Channel of the decay	Decay Mode	Level of daughter nucleus $J^\pi$ , energy (keV)	Experimental limit of $T_{1/2}$ (90% C.L.) (y)	
			[25]	[26]
$2K$	$2\nu$	g.s.	$\geq 7.1 \times 10^{16}$	$\geq 1.4 \times 10^{16}$
$KL$	$2\nu$	g.s.	$\geq 4.2 \times 10^{16}$	$\geq 1.4 \times 10^{16}$
$2K$	$2\nu$	$2^+$ , 76.5	$\geq 5.9 \times 10^{16}$	$\geq 7.9 \times 10^{16}$
$KL$	$2\nu$	$2^+$ , 76.5	$\geq 3.5 \times 10^{16}$	$\geq 7.9 \times 10^{16}$
$2L$	$2\nu$	$2^+$ , 76.5	$\geq 3.9 \times 10^{16}$	$\geq 7.9 \times 10^{16}$
$2K$	$0\nu$	g.s.	$\geq 5.8 \times 10^{17}$	$\geq 2.7 \times 10^{18}$
$KL$	$0\nu$	g.s.	$\geq 1.9 \times 10^{18}$	$\geq 4.2 \times 10^{17}$
$2L$	$0\nu$	g.s.	$\geq 7.8 \times 10^{17}$	$\geq 3.6 \times 10^{17}$
$2K$	$0\nu$	$2^+$ , 76.5	$\geq 7.1 \times 10^{17}$	$\geq 2.4 \times 10^{18}$
$KL$	$0\nu$	$2^+$ , 76.5	$\geq 6.2 \times 10^{17}$	$\geq 3.1 \times 10^{17}$
$2L$	$0\nu$	$2^+$ , 76.5	$\geq 7.2 \times 10^{17}$	$\geq 9.4 \times 10^{17}$
$K\beta^+$	$2\nu + 0\nu$	g.s.	$\geq 1.4 \times 10^{17}$	$\geq 5.6 \times 10^{16}$
$L\beta^+$	$2\nu + 0\nu$	g.s.	$\geq 1.4 \times 10^{17}$	$\geq 5.6 \times 10^{16}$

[25] HADES; NPA 996 (2020) 121703

[26] LNGS; NPA 1012 (2021) 122212

[28] HADES; Eur. Phys. J. A (2020) 56:5

[31] Phys. G (1991) 17 1223

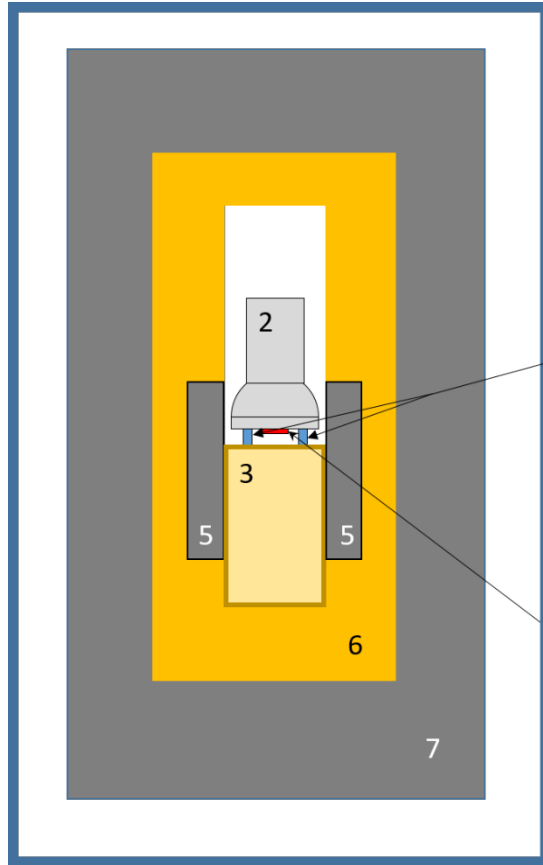
[32] J. Physique (1983) 44 791

[33] Rhys. Rev. C 92 (2015) 014602

Nuclide Transition	$J^\pi$ Parent $\rightarrow$ Daughter Nuclei and its level (keV)	$\delta$ (%) [29]	$Q_\alpha$ (keV) [30]	Decay isotope	Level of the daughter nucleus (keV)	$T_{1/2}$ (y)				
						Experimental		Theoretical		
						[28]	[26]	[31]	[32]	[33]
$^{174}\text{Hf} \rightarrow ^{170}\text{Yb}$	$0^+ \rightarrow 0^+$ , g.s. $0^+ \rightarrow 2^+$ , 84.2	0.156(6) [24]	2494.5(2.3)	$^{174}\text{Hf}$	$2^+$ , 84.2	$\geq 3.3 \cdot 10^{15}$	$\geq 2.8 \cdot 10^{16}$	$1.3 \cdot 10^{18}$	$3.0 \times 10^{18}$	$6.6 \times 10^{17}$
$^{176}\text{Hf} \rightarrow ^{172}\text{Yb}$	$0^+ \rightarrow 0^+$ , g.s. $0^+ \rightarrow 2^+$ , 78.7	5.26(70)	2254.2(1.5)	$^{176}\text{Hf}$	$2^+$ , 78.7	$\geq 3.0 \times 10^{17}$	$\geq 2.7 \times 10^{17}$	$1.3 \times 10^{22}$	$3.5 \times 10^{22}$	$4.9 \times 10^{21}$
$^{177}\text{Hf} \rightarrow ^{173}\text{Yb}$	$7/2^- \rightarrow 5/2^-$ , g.s. $7/2^- \rightarrow 7/2^-$ , 78.6	18.60(16)	2245.7(1.4)	$^{177}\text{Hf}$	$(7/2)^-$ , 78.6	$\geq 1.3 \times 10^{18}$	$\geq 1.1 \times 10^{18}$	$9.1 \times 10^{21}$	$1.2 \times 10^{24}$	$3.6 \times 10^{23}$
$^{178}\text{Hf} \rightarrow ^{174}\text{Yb}$	$0^+ \rightarrow 0^+$ , g.s. $0^+ \rightarrow 2^+$ , 76.5	27.28(28)	2084.4(1.4)	$^{178}\text{Hf}$	$2^+$ , 76.5	$\geq 2.0 \times 10^{17}$	$\geq 1.3 \times 10^{18}$	$2.4 \times 10^{25}$	$8.1 \times 10^{25}$	$7.1 \times 10^{24}$
$^{179}\text{Hf} \rightarrow ^{175}\text{Yb}$	$9/2^+ \rightarrow 7/2^+$ , g.s. $9/2^+ \rightarrow 9/2^+$ , 104.5	13.62(11)	1807.7(1.4)	$^{179}\text{Hf}$	$(7/2)^-$ , g.s. $(9/2)^+$ , 104.5	$\geq 2.2 \times 10^{18}$	-	$2.0 \times 10^{32}$	$4 \times 10^{32}$	$4.7 \times 10^{31}$
$^{180}\text{Hf} \rightarrow ^{176}\text{Yb}$	$0^+ \rightarrow 0^+$ , g.s. $0^+ \rightarrow 2^+$ , 82.1	35.08(33)	1287.1(1.4)	$^{180}\text{Hf}$	$2^+$ , 82.1	$\geq 2.2 \times 10^{18}$	$\geq 2.7 \times 10^{18}$	$2.0 \times 10^{32}$	$2.5 \times 10^{35}$	$2.2 \times 10^{34}$
						$\geq 1.0 \times 10^{18}$	$\geq 4.6 \times 10^{17}$	$4.0 \times 10^{49}$	$4.1 \times 10^{50}$	$2.1 \times 10^{48}$

# Search for rare $\alpha$ decay in Hf isotopes using **active** source approach

NPA 1002 (2020) 121941

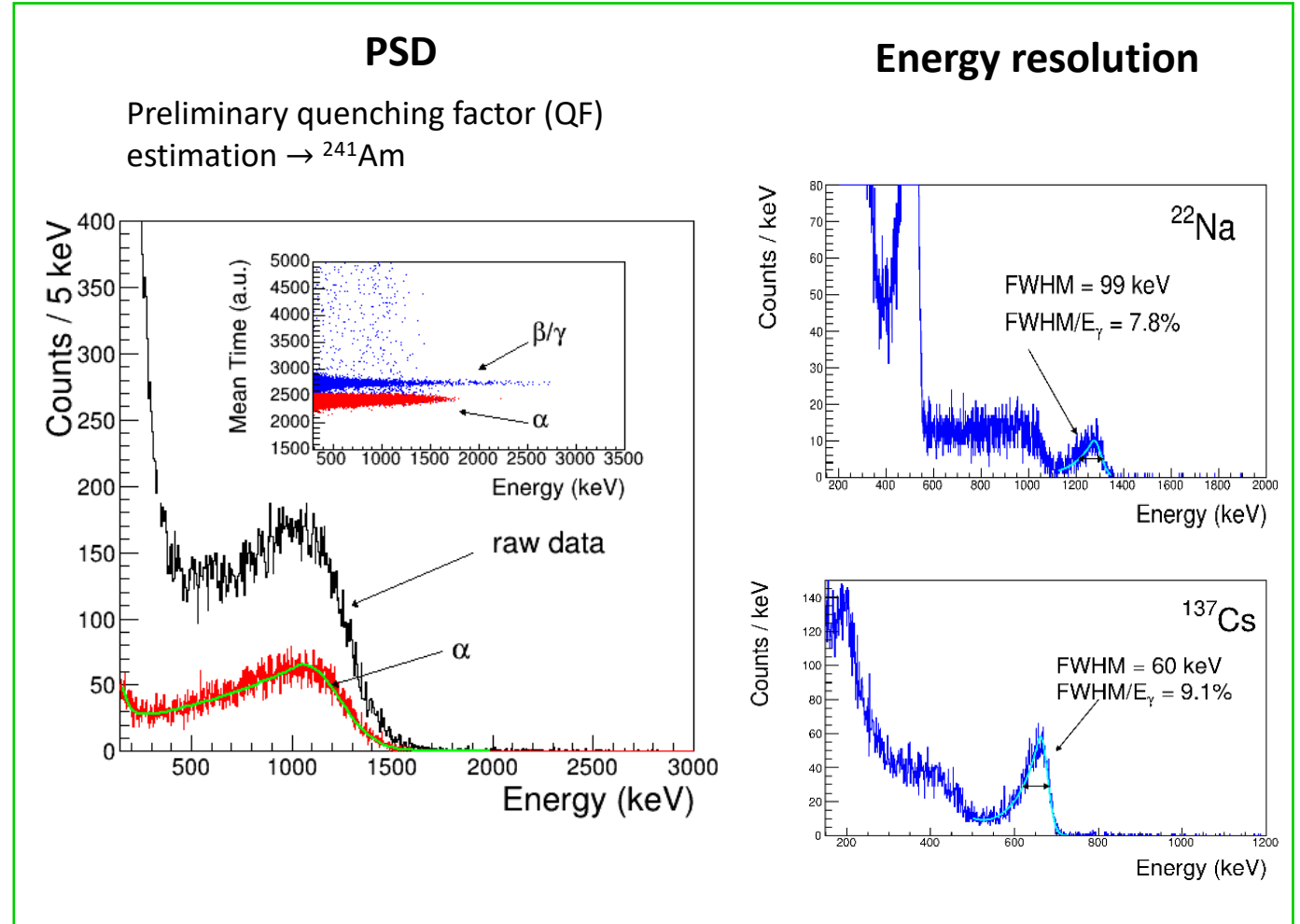


- ✓ CHC crystal (6.90(1) g) coupled low-radioactivity PMT (Hamamatsu R6233MOD) placed above the end-cap of the ultra-low background HP-Ge
- ✓ CAEN DT5720B digitizer 250 MSamples/s;
- ✓ 2848 h data taking

**STELLA facility of the LNGS**

- (1) CHC crystal scintillator
- (2) PMT
- (3) HP-Ge detector
- (4) Teflon ring
- (5) Pb, 2.5 cm
- (6) HP Cu
- (7) Pb, 25 cm
- (8) Plexiglas box

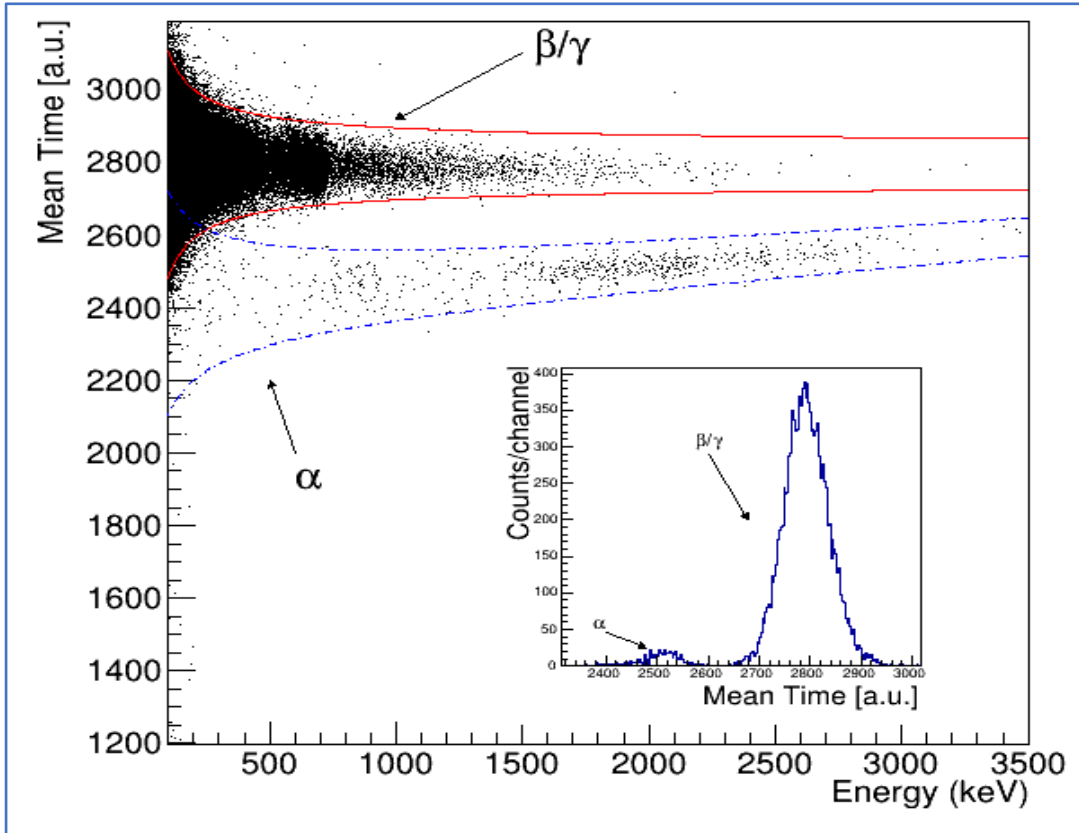
**Cs<sub>2</sub>HfCl<sub>6</sub> crystal (CHC)**



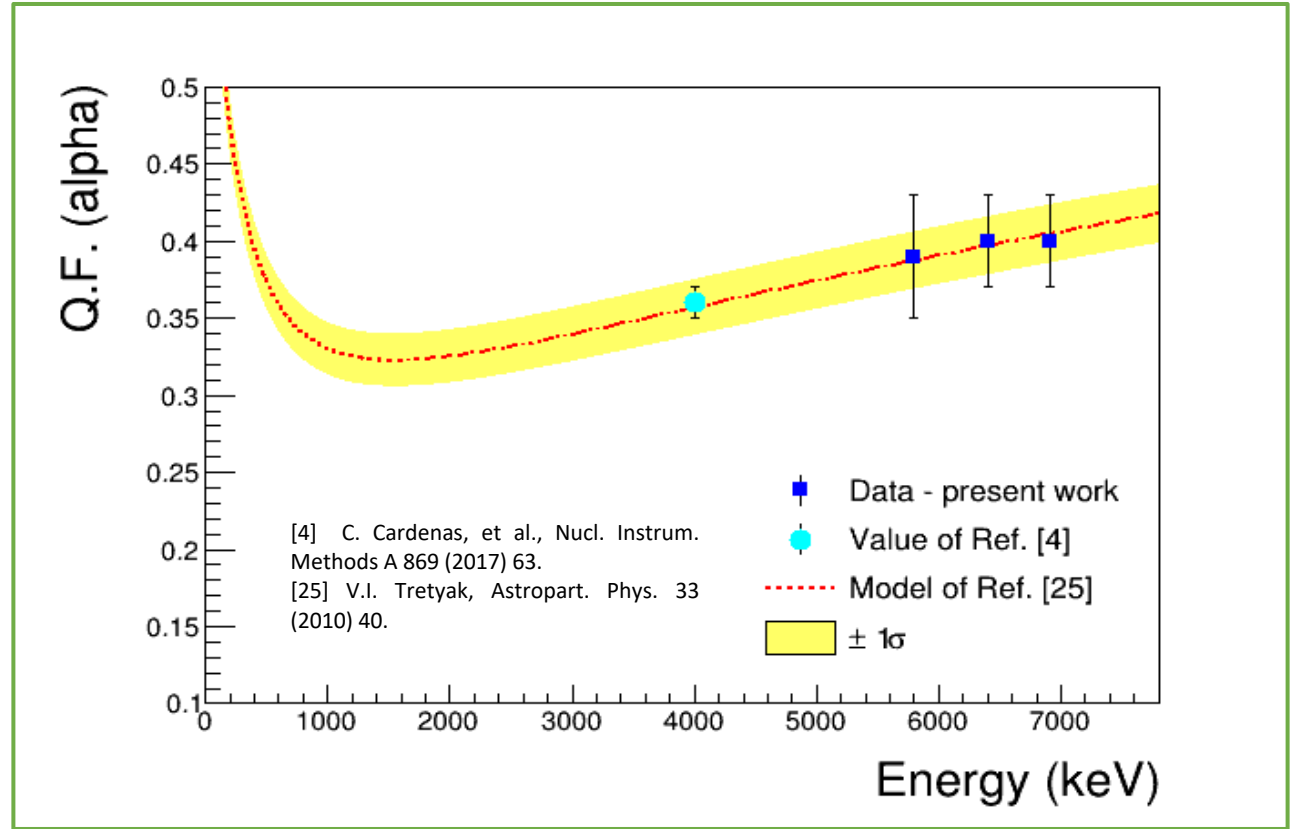
# Data analysis for the CHC crystal

Time-amplitude analysis of  $^{228}\text{Th}$  sub-chain and the derived Q.F.

Pulse Shape Discrimination (PSD) based on the pulse mean-time



The time-amplitude analysis was used to select the events of the following decay sub-chain of the  $^{232}\text{Th}$  family:  
 $^{224}\text{Ra}$  ( $Q = 5789$  keV;  $T_{1/2} = 3.66$  d)  $\rightarrow$   $^{220}\text{Rn}$  ( $Q = 6405$  keV;  $T_{1/2} = 55.6$  s)  $\rightarrow$   $^{216}\text{Po}$  ( $Q = 6906$  keV;  $T_{1/2} = 0.145$  s)  $\rightarrow$   $^{212}\text{Pb}$ .

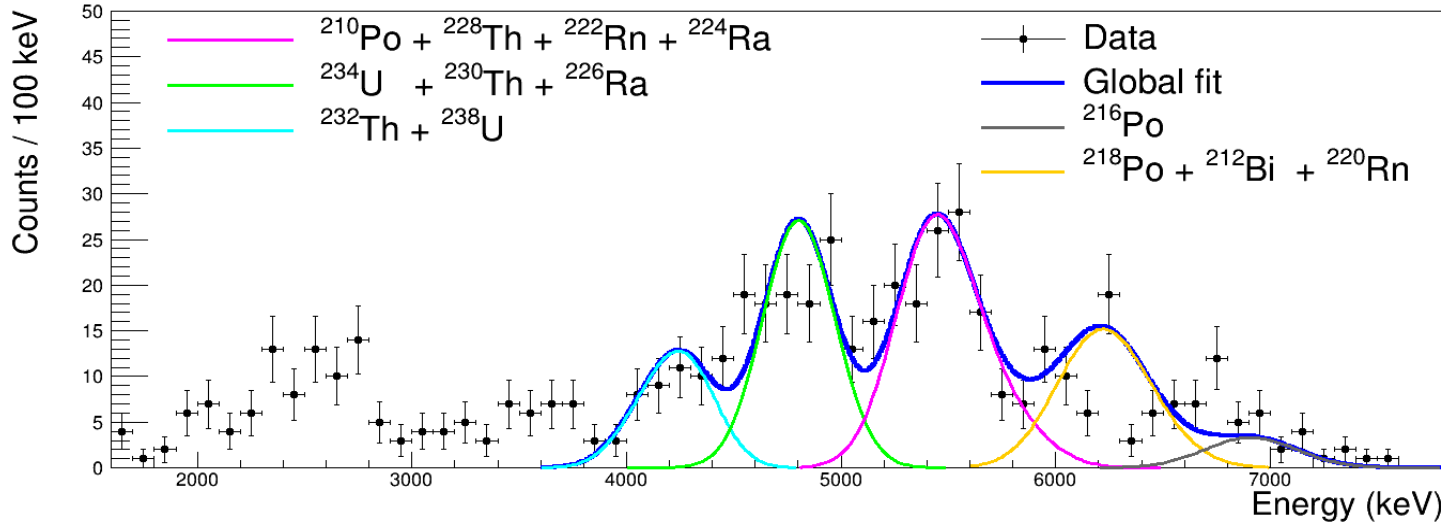


The energies of the peaks of  $^{224}\text{Ra}$ ,  $^{220}\text{Rn}$  and  $^{216}\text{Po}$ , selected by the described time-amplitude analysis, are **2260(200) keV**, **2540(200) keV**, **2780(240) keV** ( $\gamma$  scale), respectively.

# Fit of the $\alpha$ spectrum of CHC crystal and results on $\alpha$ decays of naturally occurring Hf isotopes

[24] NPA 1002 (2020) 121941

Energy spectrum of the  $\alpha$  events selected by PSD from the data of the low-background measurements with the CHC crystal scintillator over 2848 h.



- In the hypothesis of the **half-life of 1961** ([38] T.P. Kohman, Phys. Rev. 121, 1758)  **$2.0(4) \times 10^{15} \text{ y}$**  for the  $^{174}\text{Hf}$  decay, the expected number of events (**2848 h** of data taking) is about **1100 counts**.
- The **measured  $\alpha$  events are 553(23)** in total: even ascribing all of them to  $^{174}\text{Hf}$  decay, one **can safely rule out the old result for the  $T_{1/2}$**

$$T_{1/2} = (7.0 \pm 1.2) \times 10^{16} \text{ yr}$$

Chain	Sub-Chain	Activity (mBq/kg)
$^{232}\text{Th}$	$^{232}\text{Th}$	0.2(1)
	$^{228}\text{Th}$	0.2(1)
$^{238}\text{U}$	$^{238}\text{U}$	0.6(1)
	$^{234}\text{U} + ^{230}\text{Th}$	1.4(2)
	$^{226}\text{Ra}$	0.2(1)
	$^{210}\text{Po}$	1.4(2)

The total internal  $\alpha$  activity is **7.8(3) mBq/kg**.

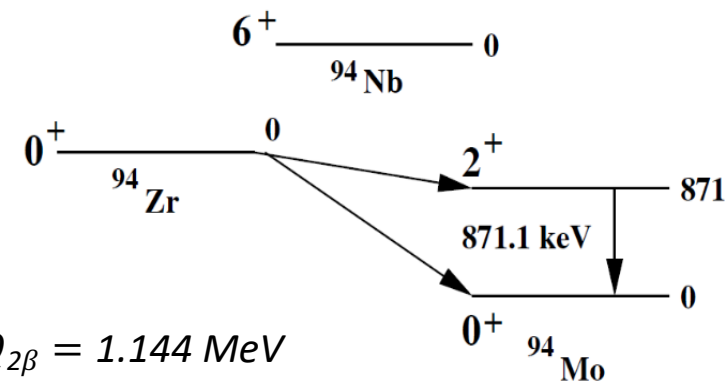
Nuclide Transition	Parent, Daughter Nuclei and its Energy Level (keV)	$T_{1/2}$ (y)				
		Experimental		Theoretical		
		[24]	[28]	[31]	[32]	[33]
$^{174}\text{Hf} \rightarrow ^{170}\text{Yb}$	$0^+ \rightarrow 0^+$ , g.s.	$7.0 \pm 1.2 \times 10^{16}$	$2.0 \pm 0.4 \times 10^{15}$ [38]	$3.5 \cdot 10^{16}$	$7.4 \times 10^{16}$	$3.5 \times 10^{16}$
	$0^+ \rightarrow 2^+$ , 84.3	$\geq 1.1 \times 10^{15}$	$\geq 3.3 \times 10^{15}$	$1.3 \cdot 10^{18}$	$3.0 \times 10^{18}$	$6.6 \times 10^{17}$
$^{176}\text{Hf} \rightarrow ^{172}\text{Yb}$	$0^+ \rightarrow 0^+$ , g.s.	$\geq 9.3 \times 10^{19}$	–	$2.5 \times 10^{20}$	$6.6 \times 10^{20}$	$2.0 \times 10^{20}$
	$0^+ \rightarrow 2^+$ , 78.7	$\geq 1.8 \times 10^{16}$	$\geq 3.0 \times 10^{17}$	$1.3 \times 10^{22}$	$3.5 \times 10^{22}$	$4.9 \times 10^{21}$
$^{177}\text{Hf} \rightarrow ^{173}\text{Yb}$	$7/2^- \rightarrow 5/2^-$ , g.s.	$\geq 3.2 \times 10^{20}$	–	$4.5 \times 10^{20}$	$5.2 \times 10^{22}$	$4.4 \times 10^{22}$
	$7/2^- \rightarrow 7/2^-$ , 78.6	$\geq 7.5 \times 10^{16}$	$\geq 1.3 \times 10^{18}$	$9.1 \times 10^{21}$	$1.2 \times 10^{24}$	$3.6 \times 10^{23}$
$^{178}\text{Hf} \rightarrow ^{174}\text{Yb}$	$0^+ \rightarrow 0^+$ , g.s.	$\geq 5.8 \times 10^{19}$	–	$3.4 \times 10^{23}$	$1.1 \times 10^{24}$	$2.2 \times 10^{23}$
	$0^+ \rightarrow 2^+$ , 76.5	$\geq 6.9 \times 10^{16}$	$\geq 2.0 \times 10^{17}$	$2.4 \times 10^{25}$	$8.1 \times 10^{25}$	$7.1 \times 10^{24}$
$^{179}\text{Hf} \rightarrow ^{175}\text{Yb}$	$9/2^+ \rightarrow 7/2^+$ , g.s.	$\geq 2.5 \times 10^{20}$	$\geq 2.2 \times 10^{18}$	$4.5 \times 10^{29}$	$4.0 \times 10^{32}$	$4.7 \times 10^{31}$
	$9/2^+ \rightarrow 9/2^+$ , 104.5	$\geq 5.5 \times 10^{17}$	$\geq 2.2 \times 10^{18}$	$2.0 \times 10^{32}$	$2.5 \times 10^{35}$	$2.2 \times 10^{34}$
$^{180}\text{Hf} \rightarrow ^{176}\text{Yb}$	$9/2^+ \rightarrow 7/2^+$ , g.s.	–	–	$6.4 \times 10^{45}$	$5.7 \times 10^{46}$	$9.2 \times 10^{44}$
	$9/2^+ \rightarrow 9/2^+$ , 82.1	–	$\geq 1.0 \times 10^{18}$	$4.0 \times 10^{49}$	$4.1 \times 10^{50}$	$2.1 \times 10^{48}$



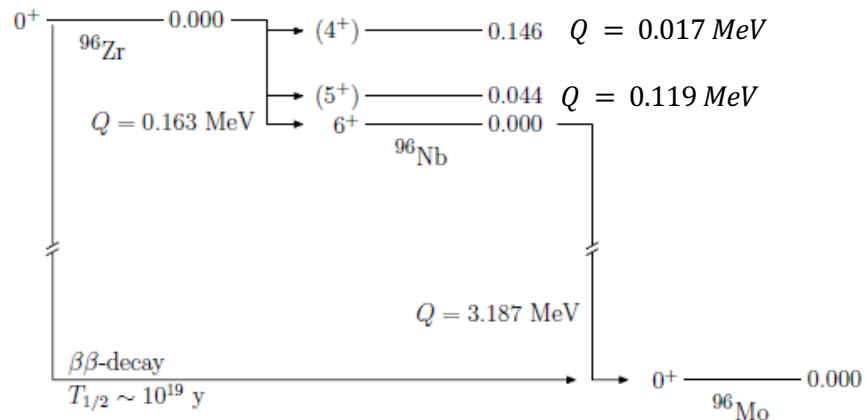
# Search for $2\beta$ decay in $^{94,96}\text{Zr}$ and for $^{96}\text{Zr}$ 's $\beta$ decay

Experiment	Transition	$T_{1/2}$ 90% C.L. (y)	Technique	Ref.
ZICOS, (Kamioka Observatory, Japan)	$^{96}\text{Zr } 0^+ \rightarrow ^{96}\text{Mo } 0^+_1$ (g.s.)	under construction	Organic liquid scintillator	[1]
NEMO I, II, III, Frejus (France) (next: SuperNEMO)	$^{96}\text{Zr } 0^+ \rightarrow ^{96}\text{Mo } 0^+_1$ (g.s.)	$>9.2 \times 10^{21}$	Tracker detector	[2]
		$>1.29 \times 10^{22}$		[3]
Kimballton Underground Research Facility, (USA)	$^{96}\text{Zr } 0^+ \rightarrow ^{96}\text{Mo } 2^+_1$	$>3.1 \times 10^{20}$	HP-Ge	[4]
Collaboration at Fréjus Underground Laboratory	$^{96}\text{Zr } 0^+ \rightarrow ^{96}\text{Mo } 2^+_1, 0^+_1, 2^+_2, 2^+_3$	$>(2.6 - 7.9) \times 10^{19}$	HP-Ge	[5]
Collaboration at LNGS	$^{96}\text{Zr } 0^+ \rightarrow ^{96}\text{Mo } 2^+_1$	$>3.8 \times 10^{19}$	HP-Ge	[6]
TILES (TIFR, Mumbai)	$^{94}\text{Zr } 0^+ \rightarrow ^{94}\text{Mo } 2^+_1$	$>5.2 \times 10^{19}$	HP-Ge	[7]
Kimballton Underground Research Facility, (USA)	$^{96}\text{Zr } 0^+ \rightarrow ^{96}\text{Mo } 6^+$	$>2.4 \times 10^{19}$	HP-Ge	[8]

Decay scheme of  $^{94}\text{Zr}$



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



[1] EPS-HEP (2019) 437

[2] NPA 847 (2010) 168

[3] PhD U. Coll. London (2015)

[4] S.W. Finch et W. Tornow, Phys. Rev. C 92 (2015) 045501

[5] J. Phys. G: Nucl. Part. Phys. 22 (1996) 487

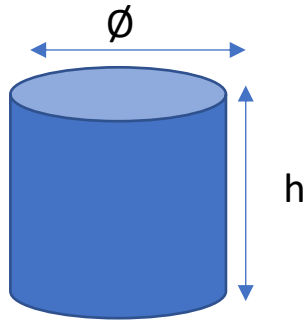
[6] C. Arpesella et al. Lett. 27 (I) (1994) pp. 29–34

[7] N. Dokania et al. J. Phys. G: Nucl. Part. Phys. 45 (2018) 075104

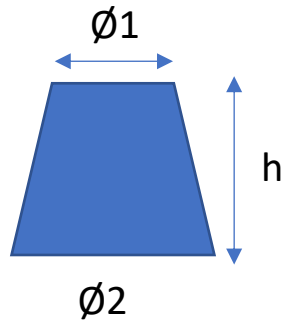
[8] S.W. Finch, W. Tornow, Nucl. Inst. Meth. A 806(2016)70–74

[9] J. Heeck and W. Rodejohann 2013 EPL 103 32001

# Search for $2\beta$ decay in Zr isotopes using **active** source approach



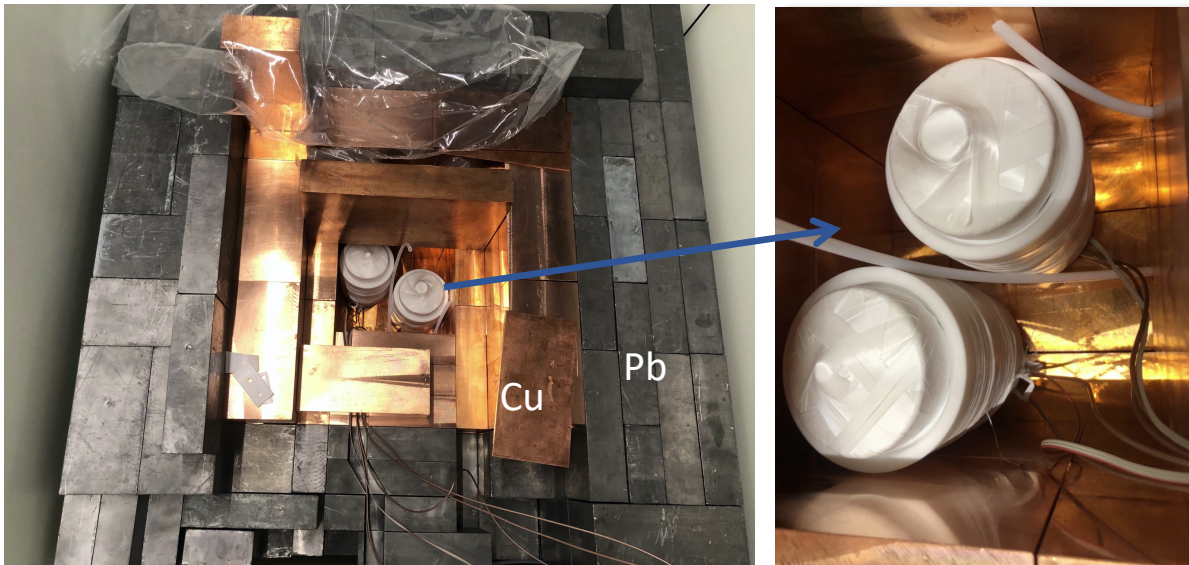
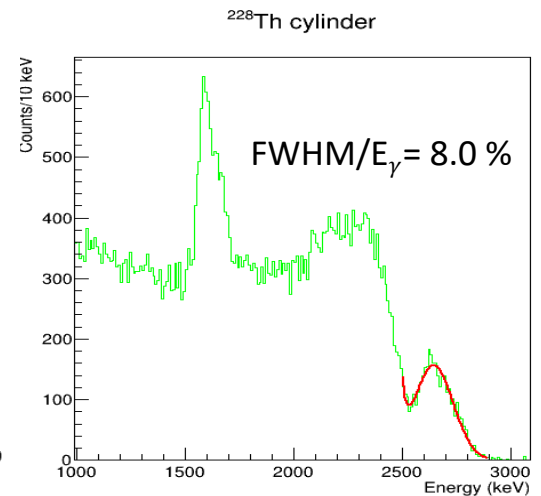
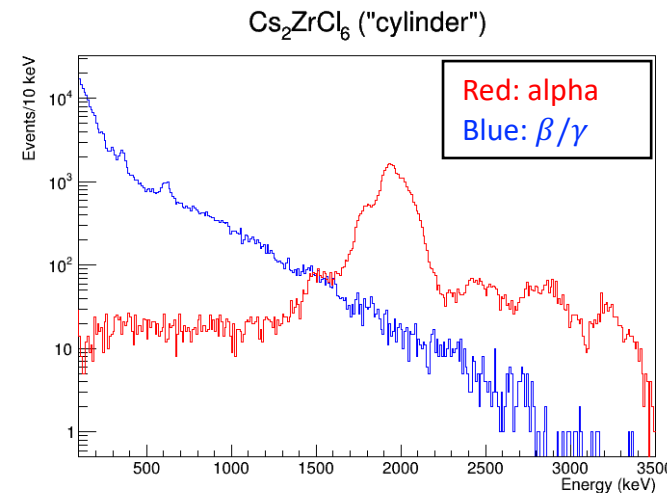
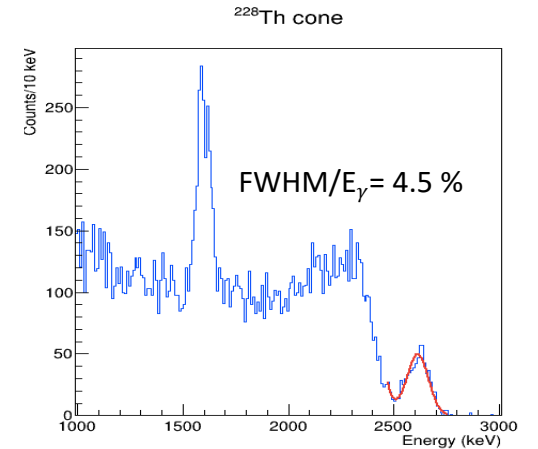
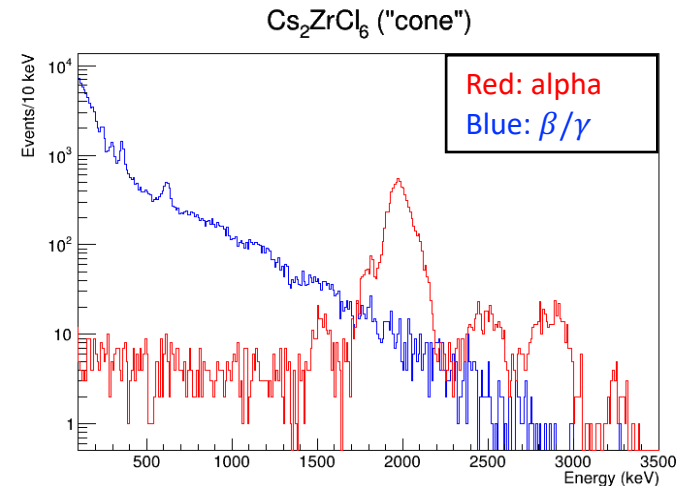
M=24,0(1) g  
h=21,20(5) mm  
 $\phi=21,00(5)$  mm



M=10,6(1) g  
h=17,90(5) mm  
 $\phi_1=8,0(1)$  mm  
 $\phi_2=19,70(5)$  mm

✓ 8736 h data taking

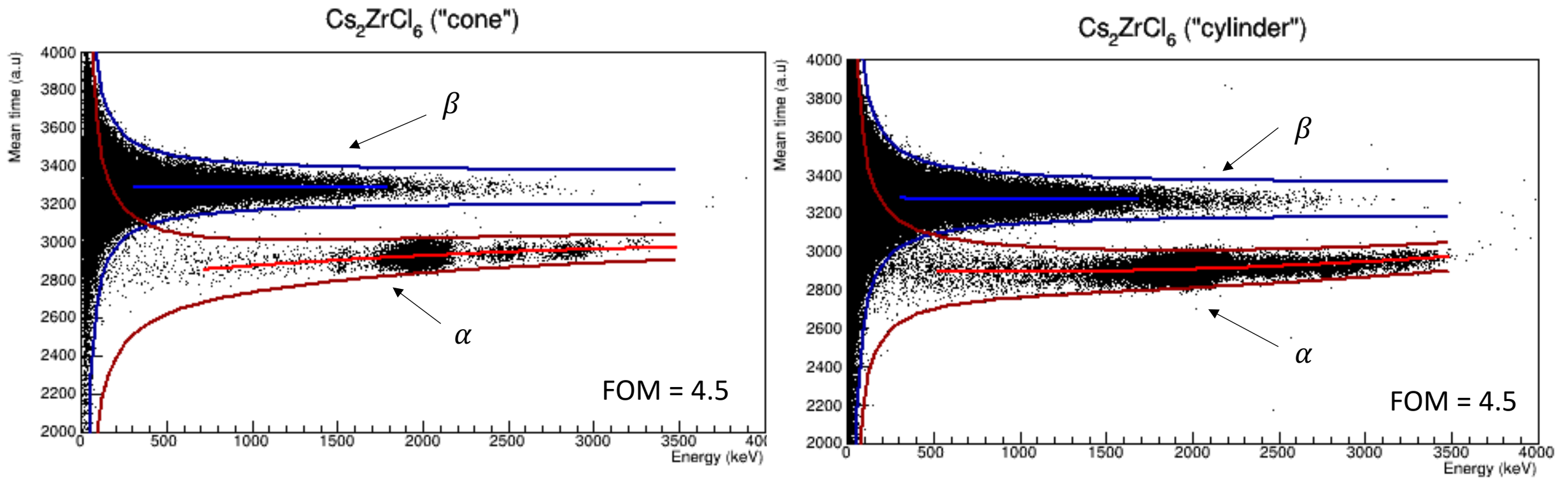
**Cs<sub>2</sub>ZrCl<sub>6</sub> crystals (CZC)**



DAMA/CRYS setup at LNGS

# Data analysis for the CZC crystals

The mean-time pulse-shape discrimination (PSD) method [10] was used to discriminate  $\beta(\gamma)$  events from  $\alpha$  events caused by  $\alpha$  radioactive contamination of the detectors by  $^{232}\text{Th}$  and  $^{238}\text{U}$  with their daughters.



The mean value of the mean time vs energy is represented together with  $3\sigma$  intervals for the two CZC crystals.

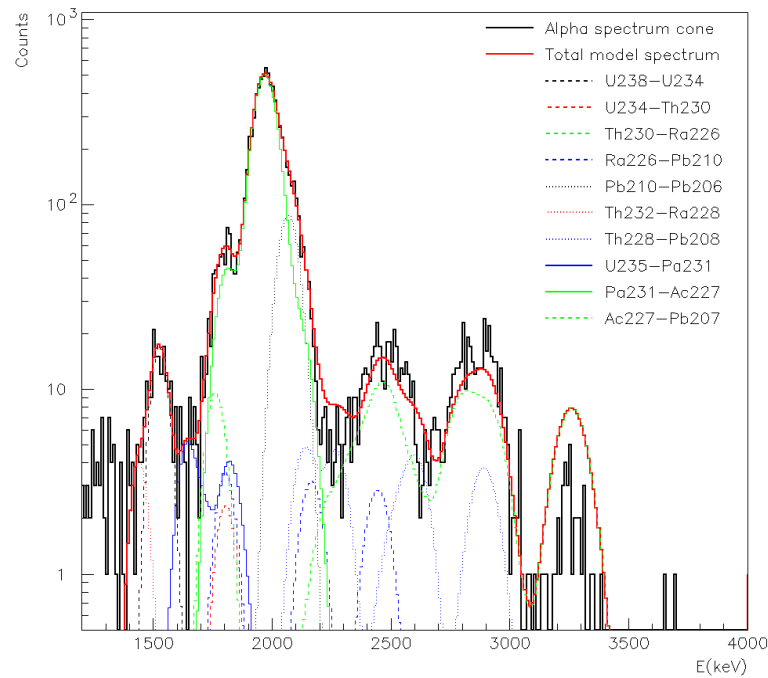
[10] L. Bardelli *et al.*, Nucl. Instr. Meth. A **584**, 129 (2008).

# Fit of the $\alpha$ spectra of CZC crystals with $\alpha$ decays from $^{238}\text{U}$ , $^{232}\text{Th}$ and $^{235}\text{U}$ chains

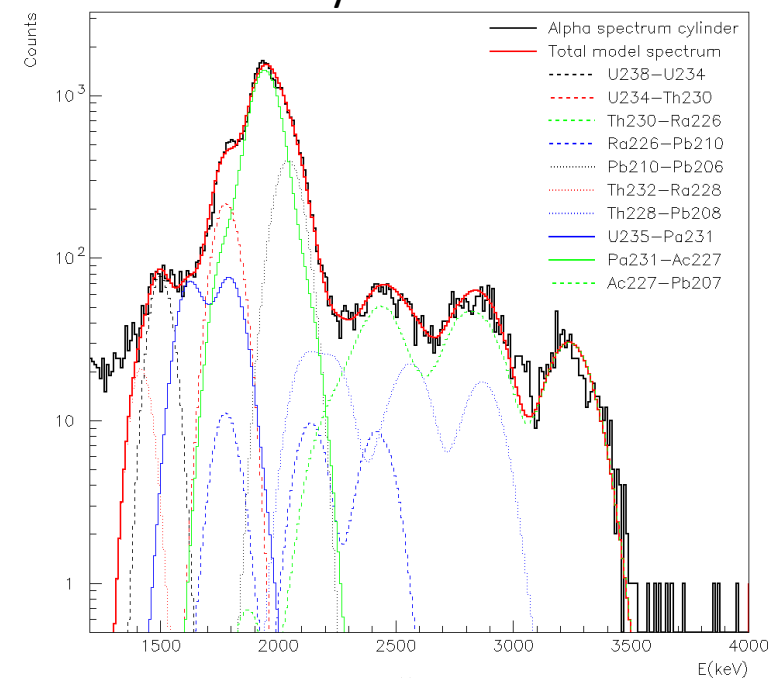
Preliminary

Fit window: [1430, 3400] keV

Cone case



Cylinder case



Radioactive contamination

Chain	Nuclide	Activity, mBq/kg	
		Cone	Cylinder
		<b>10.63 g</b>	<b>23.95 g</b>
$^{232}\text{Th}$	$^{232}\text{Th}$	0.10(3)	0.30(8)
	$^{228}\text{Th}$	0.16(3)	0.47(3)
$^{238}\text{U}$	$^{238}\text{U}$	0.43(4)	1.14(6)
	$^{234}\text{U}$	0.07(11)	3.73(16)
	$^{230}\text{Th}$	0.28(2)	< 0.02
	$^{226}\text{Ra}$	0.10(4)	0.20(5)
	$^{210}\text{Pb}$	2.8(2)	7.8(3)
$^{235}\text{U}$	$^{235}\text{U}$	0.32(5)	2.8(1)
	$^{231}\text{Pa}$	<b>20.0(3)</b>	<b>32.0(4)</b>
	$^{227}\text{Ac}$	0.40(2)	0.91(2)

- Preliminary fits including also  $\alpha$  events from  $^{235}\text{U}$  chain reproduce well the measured spectra.
- The peak with highest counting rate at  $\approx 2$  MeVee may be attributed to  $^{231}\text{Pa}$  decay from  $^{235}\text{U}$  chain for both the crystals.
- **But**, the origin of the 3  $\alpha$  peaks at higher energy (i.e.  $E = 2.45, 2.85$  and  $3.25$  MeVee), mostly due to  $^{227}\text{Ac} \rightarrow \dots \rightarrow ^{207}\text{Pb}$  sub-chain from  $^{235}\text{U}$  chain, is still under study.

**$\alpha/\beta$  light ratio:**

Cone:  $\alpha/\beta = 0.257(2) + 0.0247(4) \cdot E_\alpha [\text{MeV}]$

Cylinder:  $\alpha/\beta = 0.246(1) + 0.0258(2) \cdot E_\alpha [\text{MeV}]$

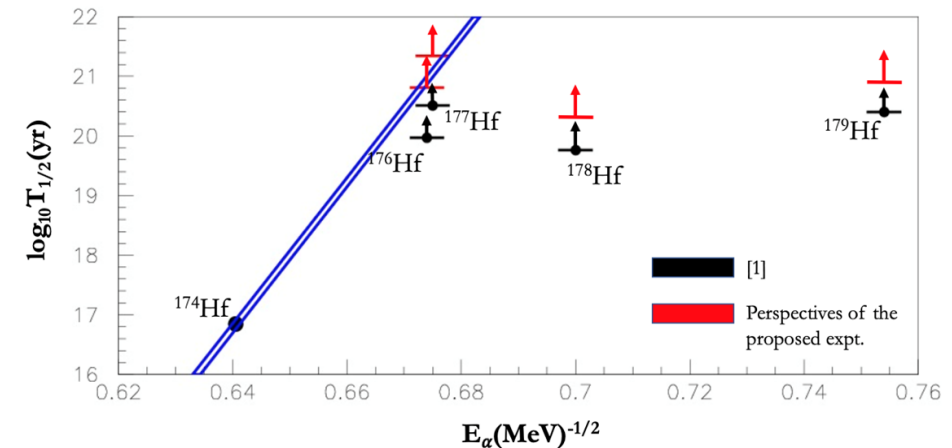
# Perspectives and conclusions

Great interest in the development of scintillating crystals from the metal hexachlorides  $\text{Cs}_2\text{MCl}_6$  (M = Hf or Zr) family

- An experiment using a CHC crystal scintillator in coincidence with a HP-Ge detector has observed  $\alpha$  decay of  $^{174}\text{Hf}$  to the ground state with a  $T_{1/2} = 7.0(1.2) \times 10^{16}$  y. This value is in good agreement with the theoretical predictions.
- New lower limits of the half-life for  $2\varepsilon$  and  $\varepsilon\beta^+$  decay of  $^{174}\text{Hf}$  ( $10^{16}$ - $10^{18}$  y) have been set.
- New lower limits of the half-life for  $\alpha$  decay of  $^{174}\text{Hf}$  to the first excited state and for  $\alpha$  decays of  $^{176}\text{Hf}$ ,  $^{177}\text{Hf}$ ,  $^{178}\text{Hf}$ ,  $^{179}\text{Hf}$  either to the ground states or to the first excited levels of daughter nuclides ( $10^{16}$ - $10^{20}$  y) have been set.
- Four CHC detectors will be fully characterized in the incoming months: the expected results after 1 year of data taking of the four detectors will allow higher accuracy for the half-life of  $^{174}\text{Hf}$ ,  $\alpha$  decay, of the order of 2.5%.
- Sensitivity for the discovery of the  $\alpha$  decay of the  $^{176}\text{Hf}$ ,  $^{177}\text{Hf}$  isotopes will reach  $T_{1/2} \sim 6.5 \times 10^{20}$  yr and  $2.2 \times 10^{21}$  yr.

- First two  $\text{Cs}_2\text{ZrCl}_6$  scintillating crystals have been grown in Queen's University and studied at the National Laboratory of Gran Sasso (LNGS, Italy).
- We are planning a new experiment with 4 CZC crystals ( $\varnothing 21 \times 21$  mm<sup>3</sup>) in optimized geometry, with light guides to reduce background from PMT, and active veto to further reduce background and mitigate cosmic muons.
- We are hoping in 1 year of data taking to reach  $10^{21}$ - $10^{22}$  y experimental sensitivity for  $T_{1/2}$  of  $0\nu 2\beta$  of  $^{96}\text{Zr}$ .

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

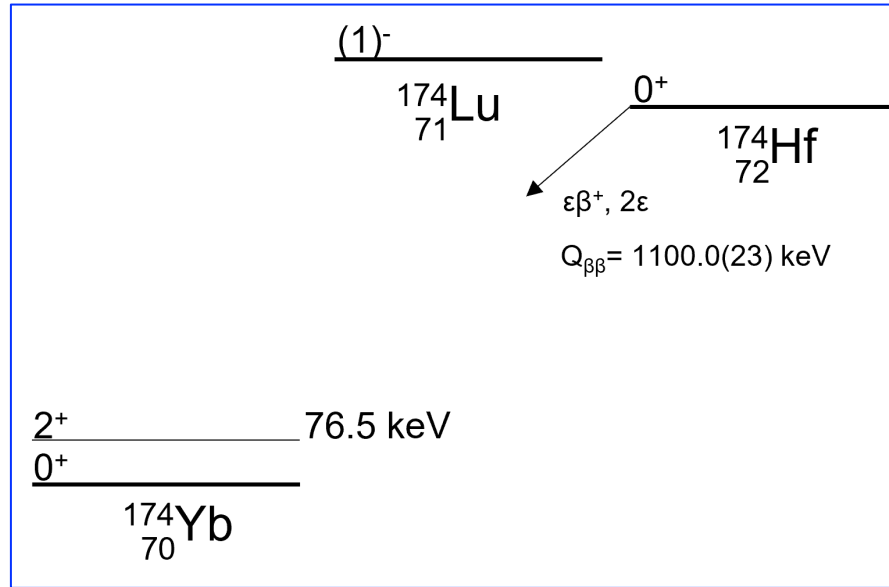


- The blue band is the extrapolation of the predictions on  $T_{1/2}$  for all the Hf isotopes using the Geiger-Nuttall scaling law considering the data point observed in Ref. [1]NPA 1002 (2020) 121941.
- The red symbols represent the sensitivity that the measurement can reach using CHC crystal scintillators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

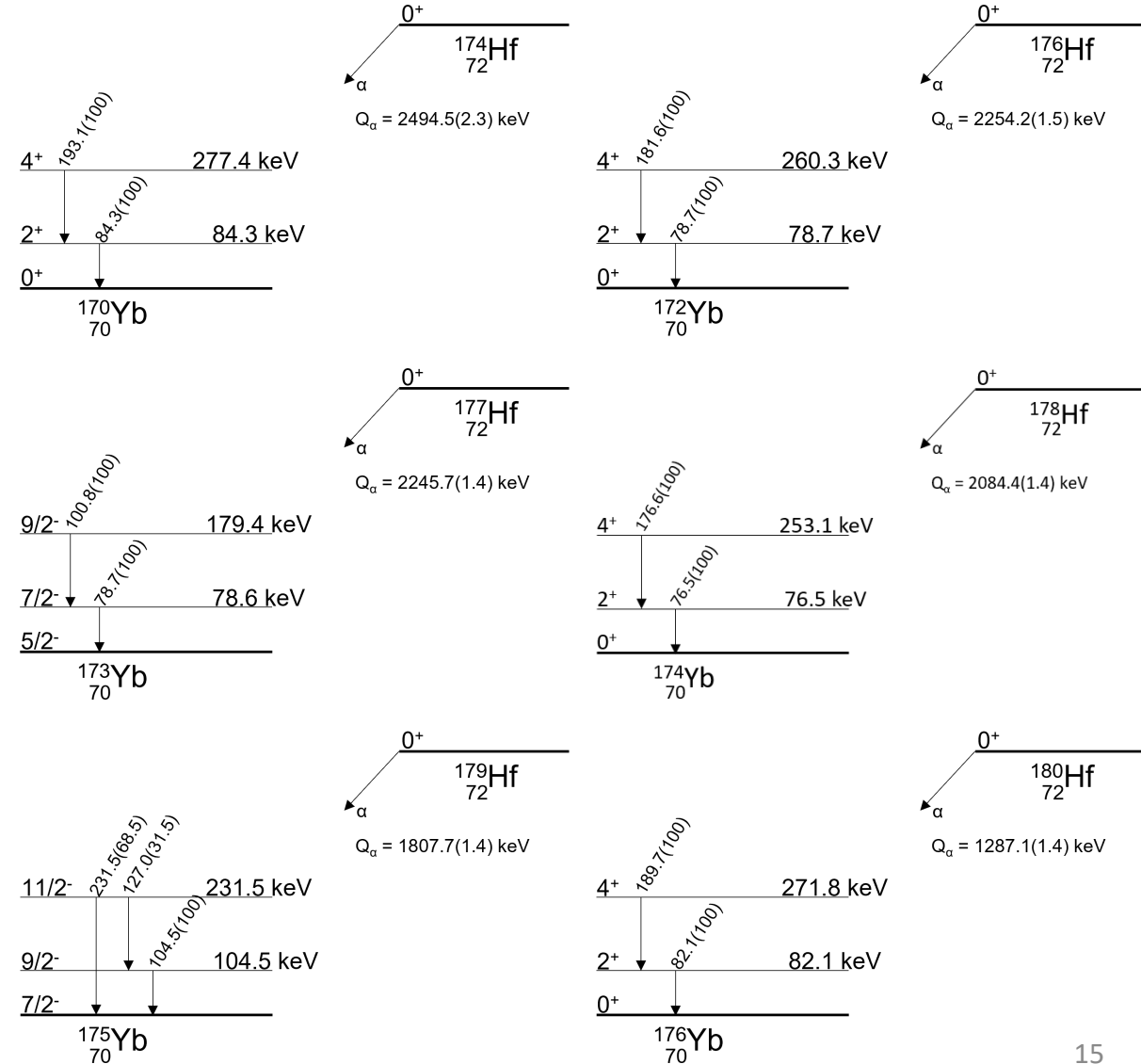
# BACKUP SLIDES

# Simplified decay schemes of naturally occurring Hf isotopes

$2\varepsilon, \varepsilon\beta^+$  decay of  $^{174}\text{Hf}$



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



# Pulse shape discrimination (PSD) based on pulse mean time

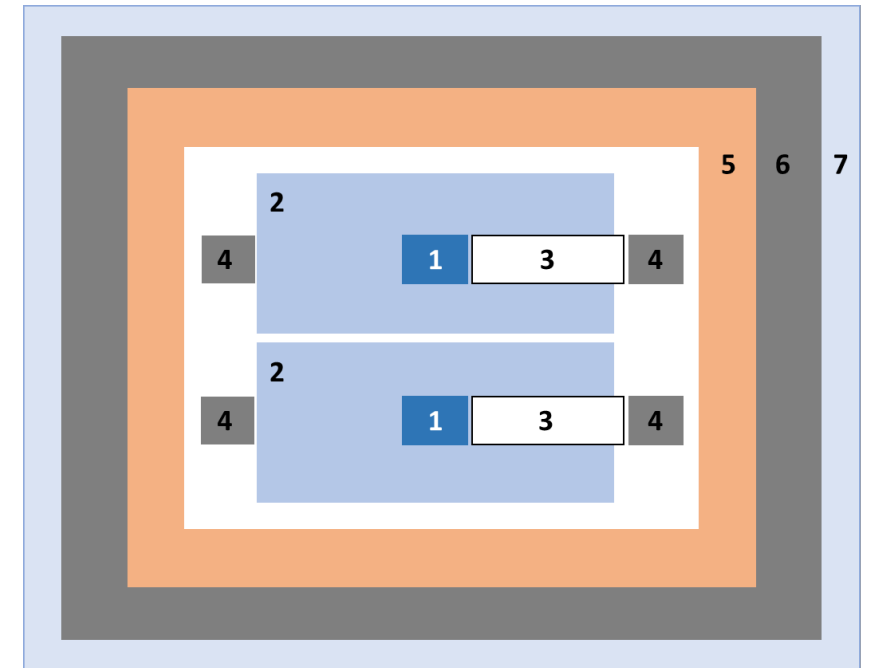
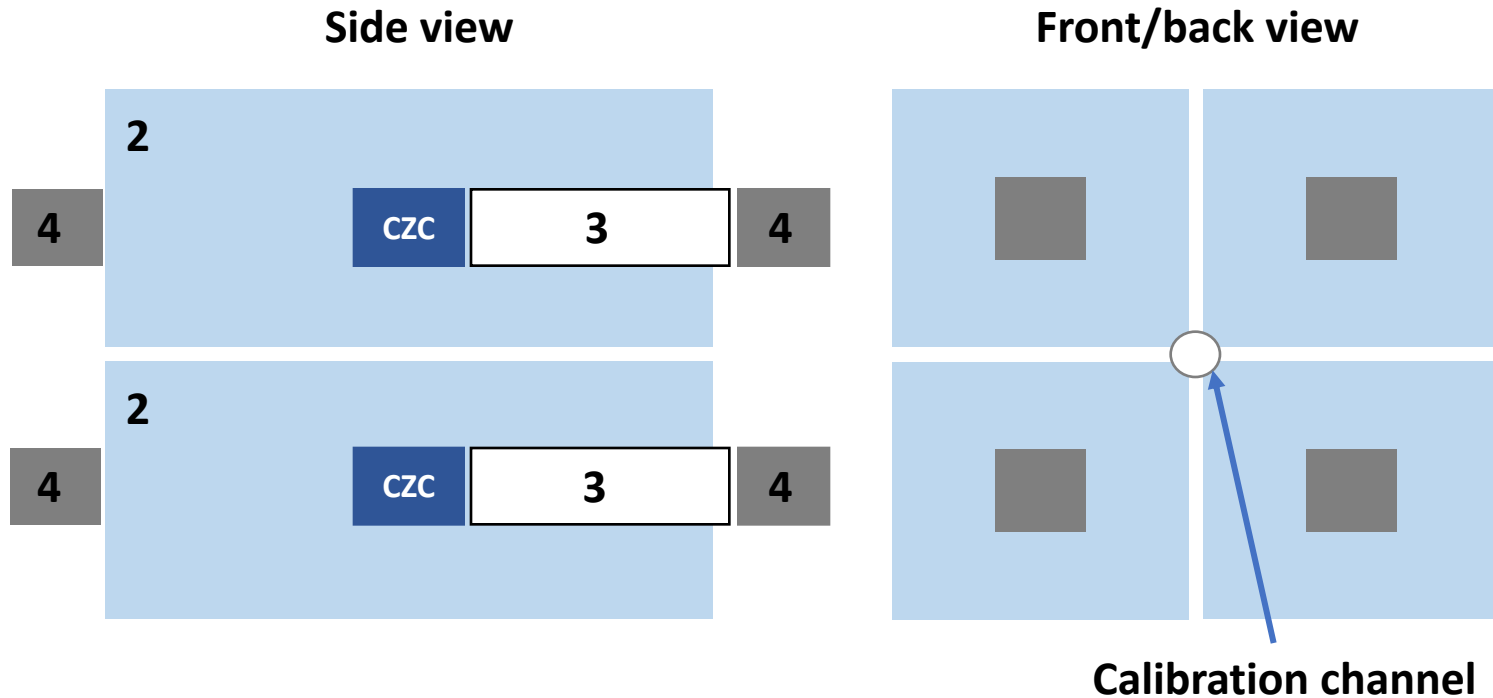
The difference in scintillation time profile of each event was used in order to determine its “mean time” ( $\langle t \rangle$ ) according to:

$$\langle t \rangle = \frac{\sum f(t_k)t_k}{\sum f(t_k)}$$

where the sum is over the time channels,  $k$ , starting from the origin of the pulse up to 8  $\mu\text{s}$ . Moreover,  $f(t)$  is the digitized amplitude (at the time  $t$ ) of a given signal.



# BREEZE detector array schematic (1<sup>st</sup> phase planned @ Queen's)



*NEWS-G3 low-background setup*

## Four separate detector's modules, each consist of:

- (1) CZC  $\varnothing$  21×21 mm<sup>3</sup>
- (2) Plastic scintillator block roughly 200×200×300 mm<sup>3</sup>
- (3) Quartz light guide  $\varnothing$  25×(100-150) mm<sup>3</sup>
- (4) 2 low-background PMTs

- (5) OFHC Cu, 10 cm
- (6) Pb, 20 cm
- (7) HDPE, 10 cm
- (8) 4 $\pi$  muon veto