New prospects in the search for 0ν2β decay of ^{96}Zr

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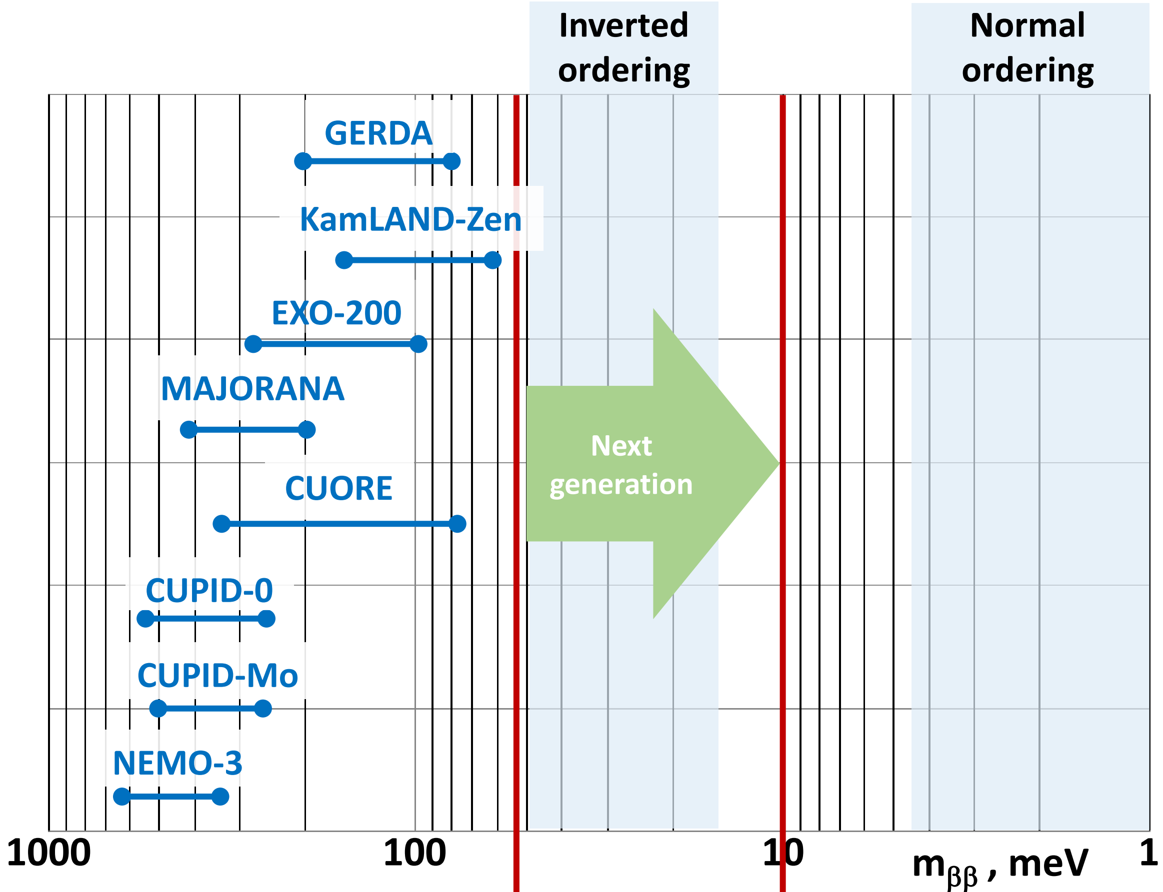
Walking through the forest of 2β giants…

Over the past 20 years, impressive progress has been achieved in the search for 0ν2β decay

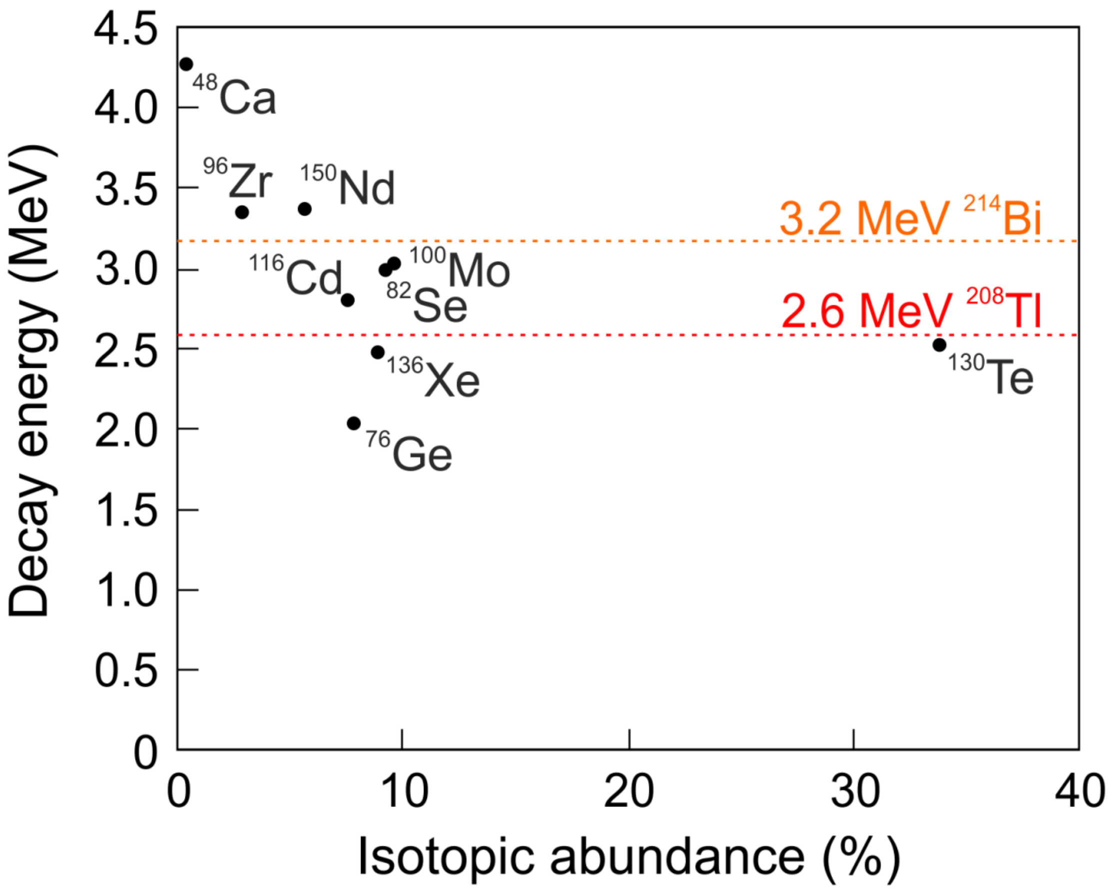
GERDA, ^{76}Ge *Phys. Rev. Lett. 125 (2020) 252502*	**$T_{1/2} > 1.8\times10^{26}$ yr**
KamLAND-Zen, ^{136}Xe *Phys. Rev. Lett. 117 (2016) 082503*	**$T_{1/2} > 1.1\times10^{26}$ yr**
EXO-200, ^{136}Xe *Phys. Rev. Lett. 123 (2019)161802*	**$T_{1/2} > 3.5\times10^{25}$ yr**
MAJORANA Dem., ^{76}Ge *Phys. Rev. C 100 (2019) 025501*	**$T_{1/2} > 2.7\times10^{25}$ yr**
CUORE, ^{130}Te *arXiv:2011.09295v2*	**$T_{1/2} > 3.2\times10^{25}$ yr**
CUPID-0, ^{82}Se *Phys. Rev. Lett. 129 (2022) 111801*	**$T_{1/2} > 4.6\times10^{24}$ yr**
CUPID-Mo, ^{100}Mo *Eur. Phys. J. C 82 (2022) 1033*	**$T_{1/2} > 1.8\times10^{24}$ yr**
NEMO-3, ^{100}Mo *Phys. Rev. D 92 (2015) 072011]*	**$T_{1/2} > 1.1\times10^{24}$ yr**



Bkg ~ 10^{-2}-10^{-3} counts/(keV·kg·yr)
M ~ 0.1 – 1 ton

Bkg ~ 10^{-4} counts/(keV·kg·yr)
M ~ 1 – 10 tons

$0\nu2\beta$ searches with non-trivial candidates



We propose to study
$0\nu2\beta$ of **^{96}Zr** with novel Cs_2ZrCl_6 scintillators
via "source = detector" experimental approach

^{76}Ge, ^{130}Te, ^{136}Xe are facing issues with an internal and environmental gamma background, while profiting from well-developed crystal production and material purification technologies

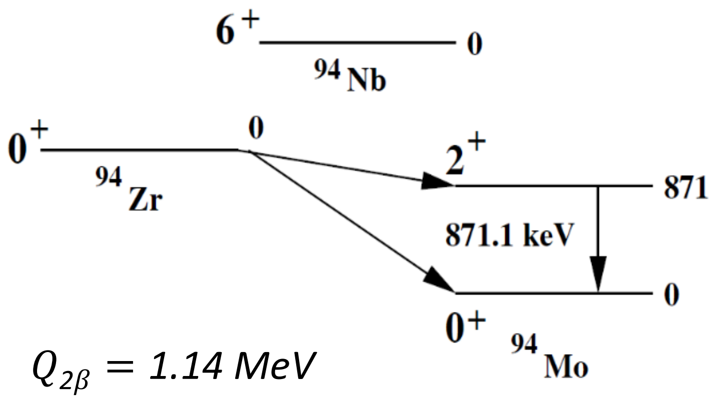
^{82}Se, ^{100}Mo, ^{116}Cd – only ^{100}Mo is under consideration due to a well-developed detector material and its high radiopurity

^{48}Ca, ^{96}Zr, ^{150}Nd are the less studied due to combination of unfavorable experimental conditions specific to each of them

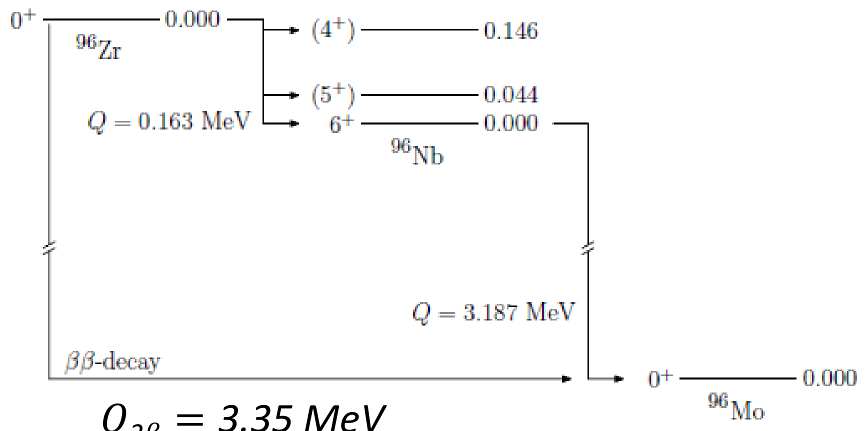
- *$Q_{\beta\beta}$ (^{96}Zr) = 3.35 MeV*
- *Favorable from a theoretical point of view $T_{1/2}$ ~$(Q_{\beta\beta})^5$*
- *Reasonable natural i.a. (2.8%)*
- *About 15 g of enriched ^{96}Zr (55%) could be available*
- *New advanced detector material (Cs_2ZrCl_6)*
- *Crystal production under full control*
- *Extensive studies of the detector properties*

Brief overview of rare decay search in $^{94,96}Zr$ isotopes

Experiment	Transition	$T_{1/2}$ @ 90% C.L. (yr)	Ref.	Technique
ZICOS (Kamioka Observatory, Japan)	$^{96}Zr \rightarrow {}^{96}Mo$ (g.s.)	under construction	[1]	Liquid scintillator
NEMO-3 (Frejus, France)	$^{96}Zr \rightarrow {}^{96}Mo$ (g.s.)	$> 9.2\times10^{21}$ $> 1.29\times10^{22}$	[2] [3]	Tracking detector
Kimballton Underground Research Facility, (USA)	$^{96}Zr \rightarrow {}^{96}Mo\ (2^+_1)$	$> 3.1\times10^{20}$	[4]	HPGe
Collaboration at Frejus, (France)	$^{96}Zr \rightarrow {}^{96}Mo\ (2^+_1, 0^+_1, 2^+_2, 2^+_3)$	$> (2.6 - 7.9)\times10^{19}$	[5]	HPGe
Collaboration at LNGS	$^{96}Zr \rightarrow {}^{96}Mo\ (2^+_1)$	$> 3.8\times10^{19}$	[6]	HPGe
Collaboration at LNGS	$^{94}Zr \rightarrow {}^{94}Mo\ (2^+_1)$	$> 2.1\times10^{20}$	[7]	HPGe
TILES (TIFR, Mumbai)	$^{94}Zr \rightarrow {}^{94}Mo\ (2^+_1)$	$> 5.2\times10^{19}$	[8]	HPGe
Kimballton Underground Research Facility (USA)	$^{96}Zr \rightarrow {}^{96}Mo\ (6^+)$	$> 2.4\times10^{19}$	[9]	HPGe



$Q_{2\beta} = 1.14\ MeV$



$Q_{2\beta} = 3.35\ MeV$

[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 (l) (1994) 29
[7] E.Celi et al., Eur. Phys. J. C 83 (2023) 396
[8] N. Dokania et al. J. Phys. G: Nucl. Part. Phys. 45 (2018) 075104
[9] S.W. Finch, W. Tornow, Nucl. Inst. Meth. A 806 (2016) 70
[10] J. Heeck and W. Rodejohann, EPL 103 (2013) 32001

→ Possibility to study $0\nu4\beta$ decay of $^{96}Zr \rightarrow {}^{96}Ru$

Cs_2ZrCl_6: a novel crystal scintillator

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

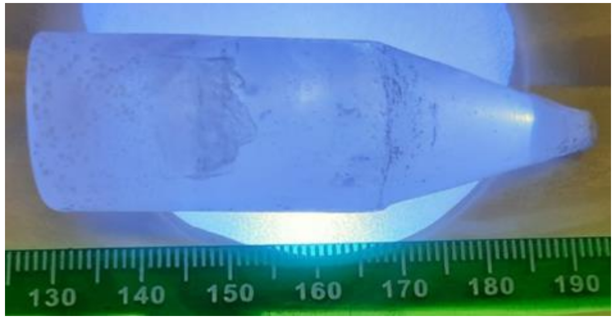
* *for alpha events at room temperature* (Dalton Trans. 2022, 51, 6944-6954)

** *for gamma quanta at room temperature (article in press)*

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

Produced at Queen's University

CsCl (99.9%) +
$ZrCl_4$ (99.9%) double sublimed
Bridgman growth technique



∅21.5×60 mm, about 60 g

23.95 g **∅21.1×21.2 mm** **Cylindrical part**	**&**	**10.62 g** **∅20.5×14 mm** **Conical part**

were subjected to further studies

Chemical purity of reagents at the each production stage

HR-ICP-MS, concentrations are in ppb with 25% uncertainty

	CsCl initial	$ZrCl_4$ initial	$ZrCl_4$ 1st sublimation	$ZrCl_4$ 2nd sublimation	CZC 1st growth, tail	CZC 1st growth, nose	CZC 2st growth, middle
K	300	15000	700	700	2500	200	500
La	0.7	1.5	1	1	1	0.6	0.6
Ce	1.5	2	1	1	2.5	3	2
Pr	0.1	4	6	6	1.5	1	1
Nd	<1	30	25	30	5	3	3
Sm	0.5	1	4	1	1	0.6	0.6
Eu-Lu	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5
Hf	35	6400	5200	5600	1200	1800	1600
Ta, W, Re, Os, Ir	<2	<2	<2	<2	<2	<2	<2
Pt	<1	<100	<100	<100	<25	<25	<25
Tl	0.4	<0.2	<0.2	0.2	1	<0.2	<0.2
Pb	<1	30	20	30	150	1	13
Bi	<0.5	<0.5	1.5	2.6	1.5	<0.5	1.6
Th	<0.05	70	0.5	0.2	<0.05	<0.05	<0.05
U	<0.05	1000	7	0.36	0.35	0.13	<0.05

Cs_2ZrCl_6 crystal radiopurity

over 700 hours of low-background measurements on HPGe detector (STELLA facility, LNGS)

Chain	Nuclide	Activity, mBq/kg	
		Cone	Cylinder
		10.63 g	**23.95 g**
^{232}Th	^{228}Ra	< 16	< 23
	^{228}Th	< 6.7	< 8.2
^{238}U	^{226}Ra	60(10)	< 8.7
	^{234}Th	< 180	< 260
	^{234m}Pa	< 630	< 160
^{235}U	^{235}U	< 16	< 12
	^{40}K	< 120	< 95
	^{137}Cs	< 7.1	< 1.6
	^{134}Cs	49(6)	42(5)
	^{132}CS	< 8.2	< 11

Surface cross-contamination during the sample preparation

Natural

Artificial

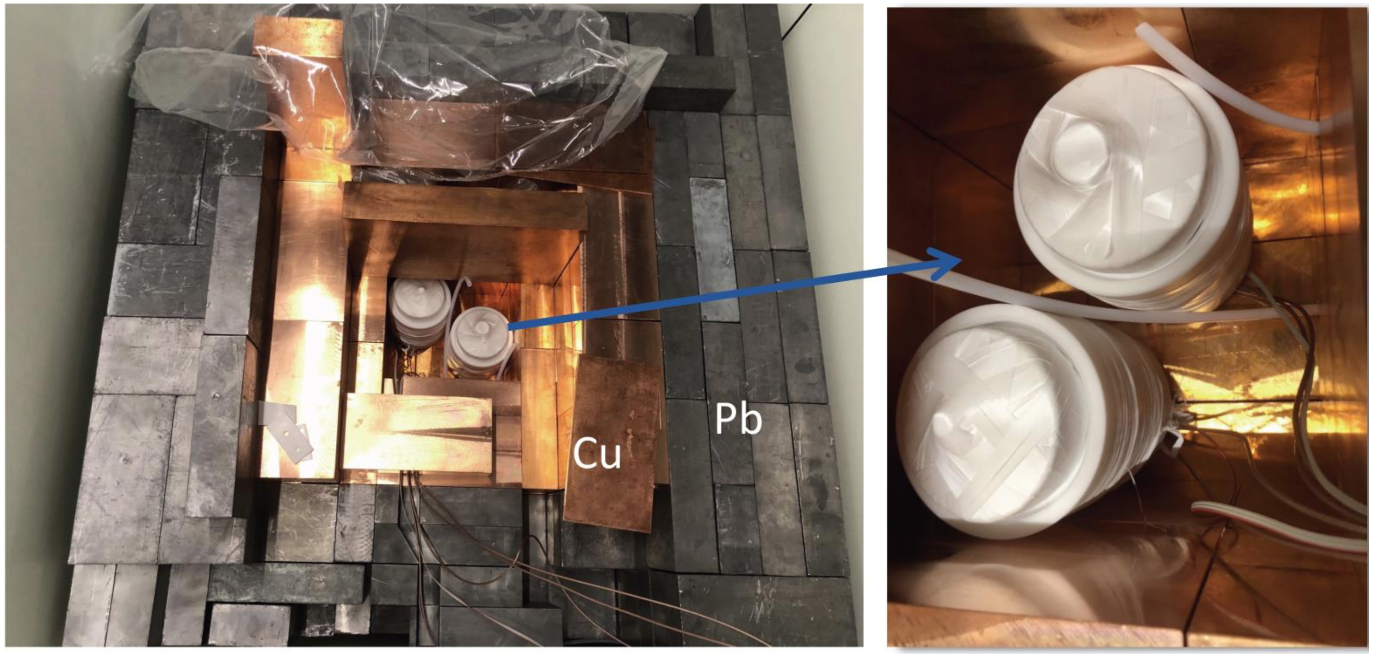
Cosmogenic activation

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

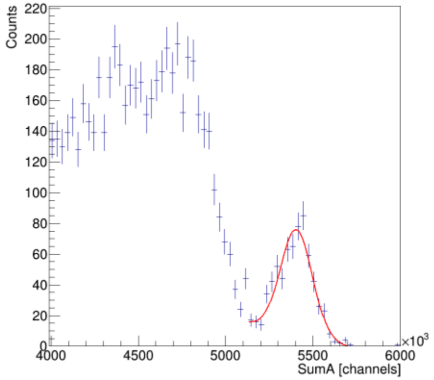
Our crystals are rather clean, even if they were grown from 99.9% purity grade raw materials

Low-background measurements at LNGS (Italy)

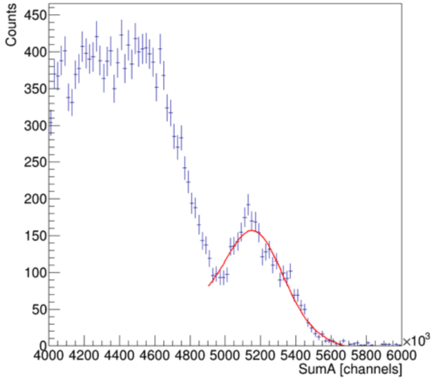
DAMA/CRYS low-background setup at LNGS



- OFHC Cu (15 cm)
- Low-activity Pb (20 cm)
- HDPE (5 cm)
- Borated HDPE (5cm)



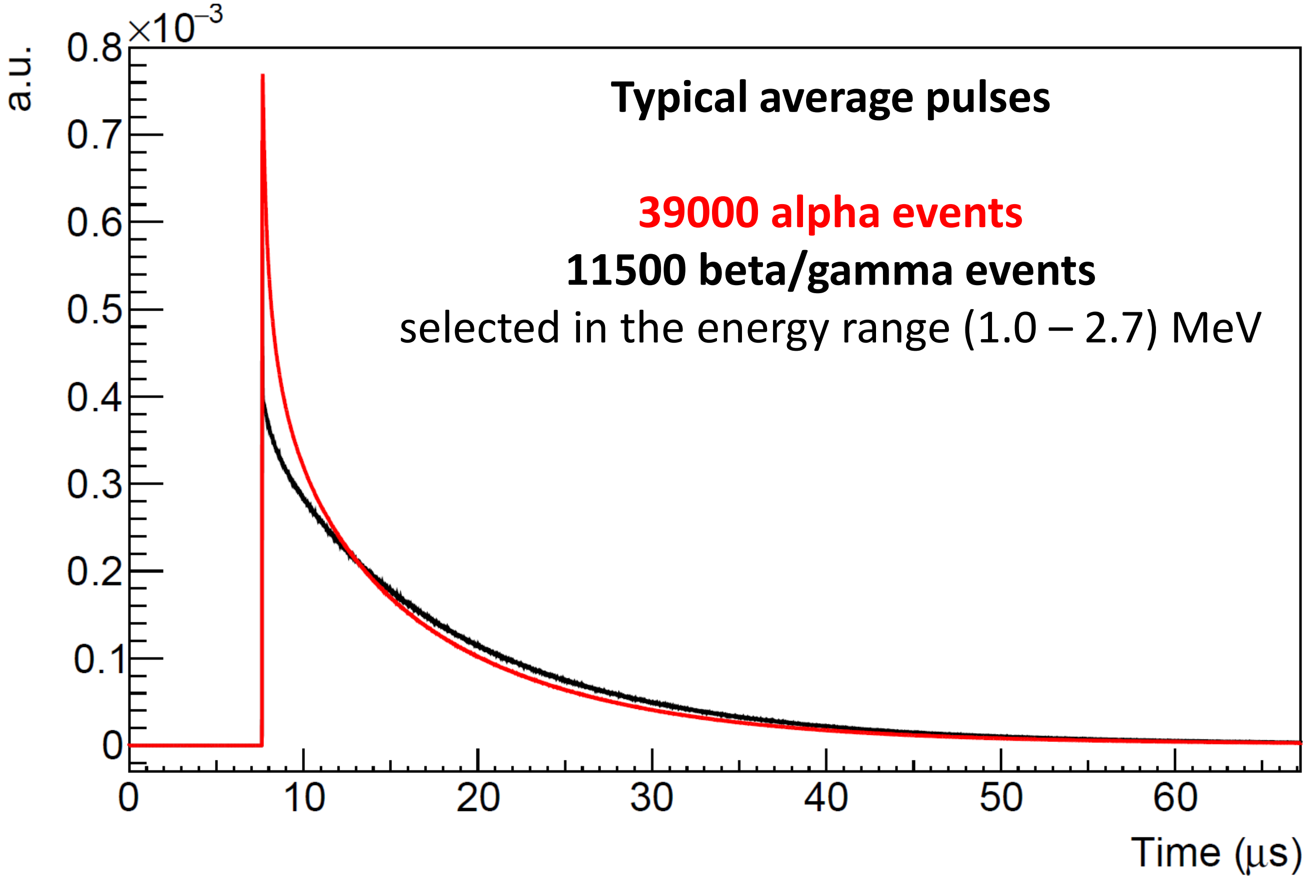
Cone
FWHM = 4.1% @ 2.6MeV



Cylinder
FWHM = 6.8% @ 2.6MeV

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

Pulse-shape discrimination ability



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

The "mean-time" (⟨*t*⟩) method [1] was used, and this parameter was determined according to:

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

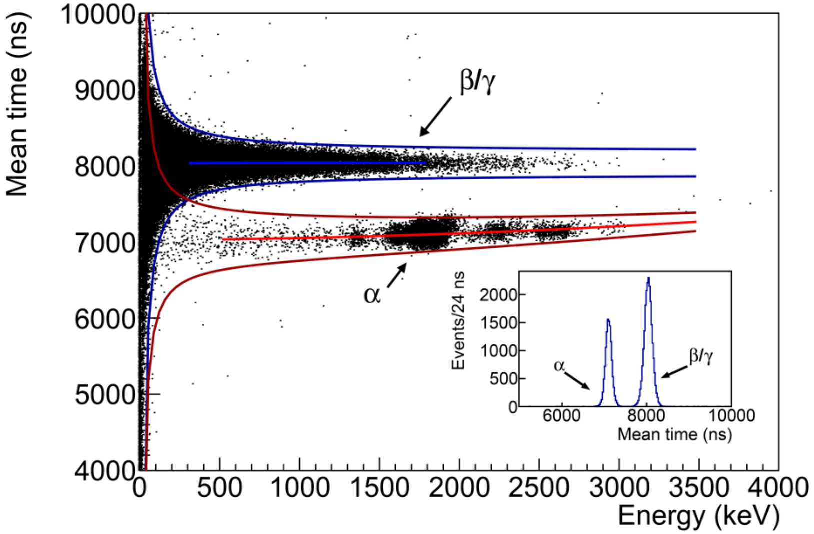
where the sum is over the time channels (*k*), starting from the origin of pulse up to 24 μs, *f(t)* is the digitized amplitude (at the time *t*) of a given signal

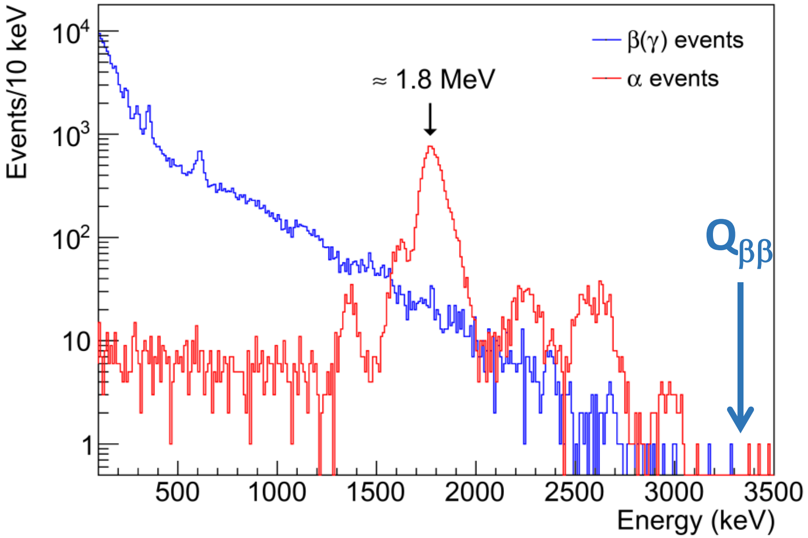
Mean-time for the presented pulses are:
⟨*t*⟩ = 7.07 and 8.00 μs, for alpha and beta/gamma events respectively

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

Pulse-shape discrimination and background α event selection

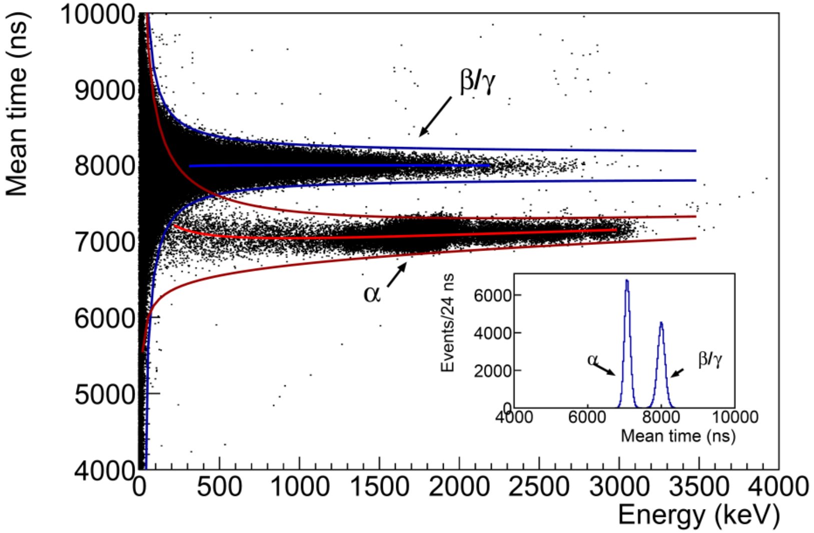
Cone
FoM = 7.8

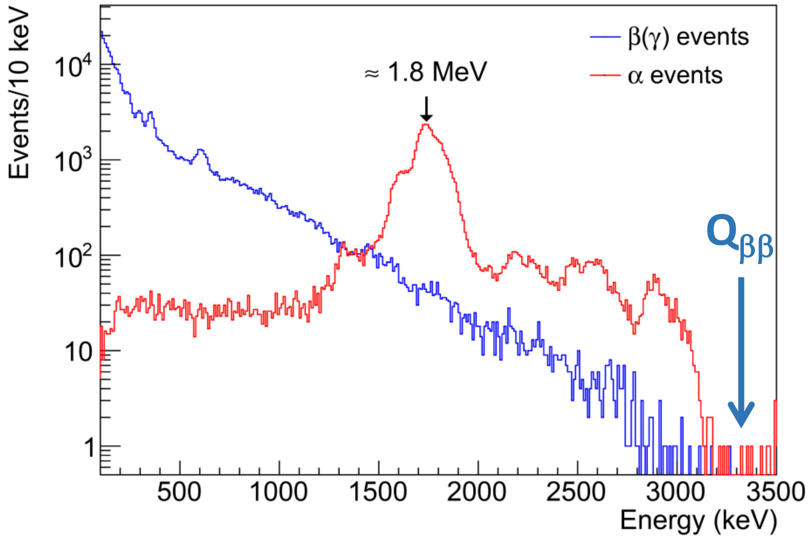




Selection efficiency is 99.7% in [0.1–3.5] MeV

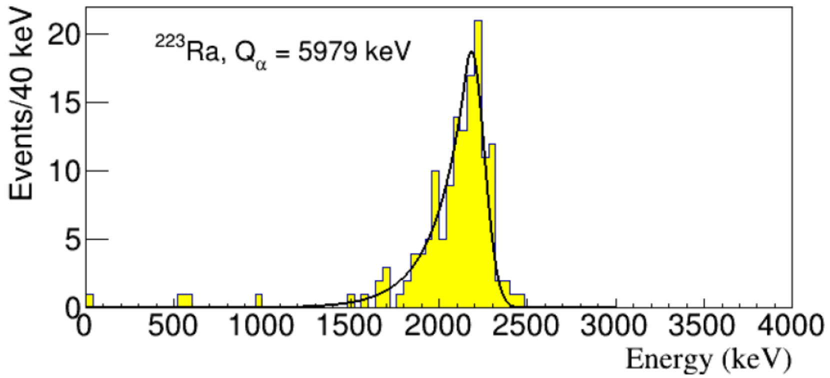
Cylinder
FoM = 7.2

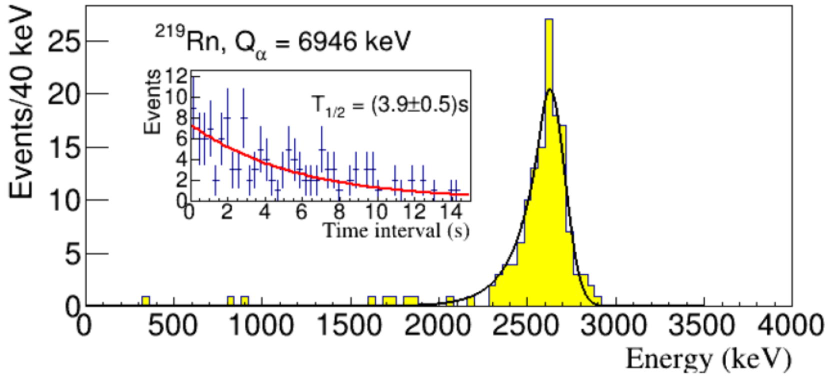


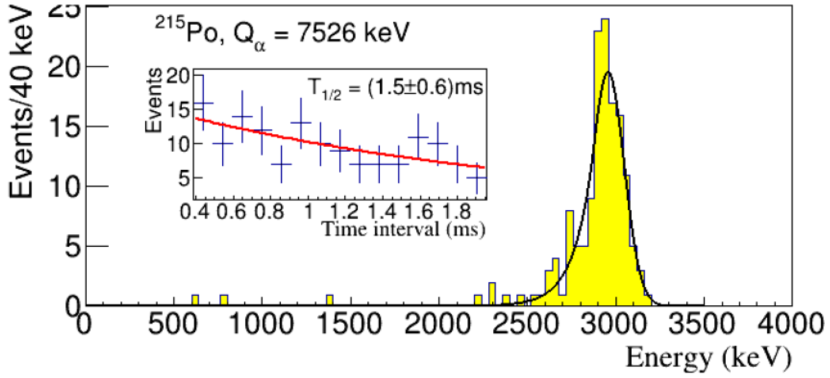


Counting rate at ROI is 0.09 counts/(kg·keV·yr)

Time-Amplitude analysis







<u>Run 2:</u> 65 days of data taking within Oct-Dec 2022 (extended time-window 2 ms)

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

^{223}Ra (Q_α = 5979 keV, $T_{1/2}$ = 11.44 d) **39.28% selection eff.**

↓

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

↓

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

↓

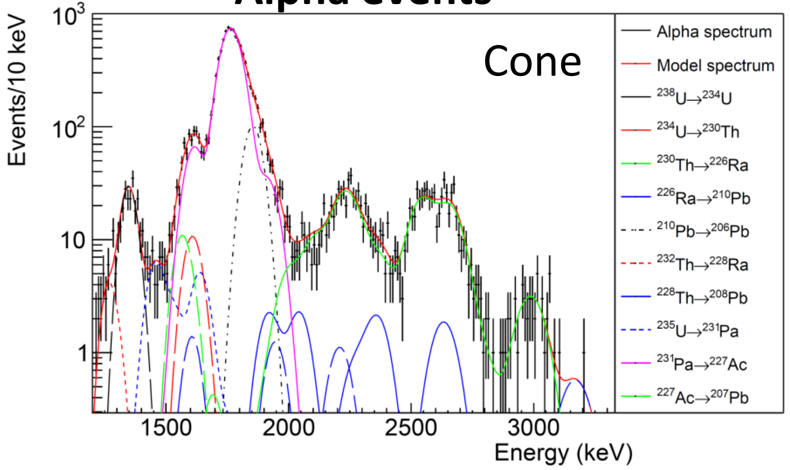
^{211}Pb

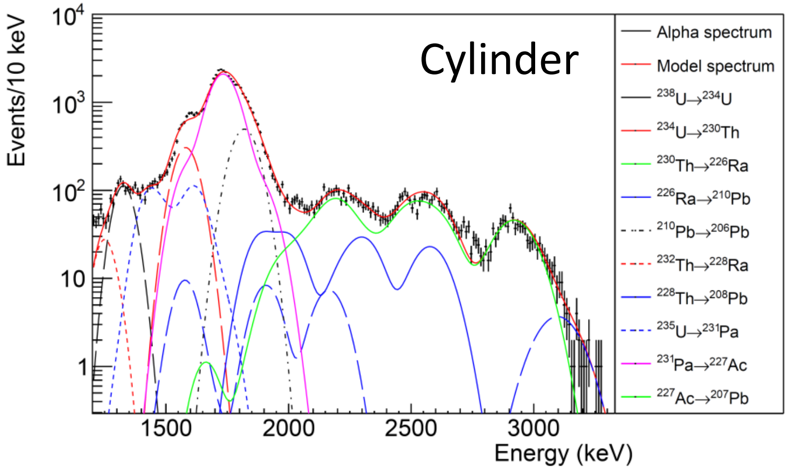
A(^{227}Ac) = 1.4(2) mBq/kg in cone
= 2.7(2) mBq/kg in cylinder

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

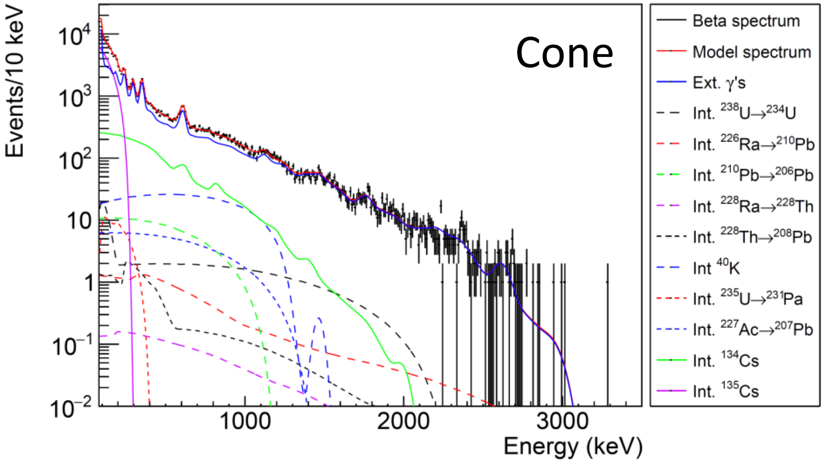
Background model

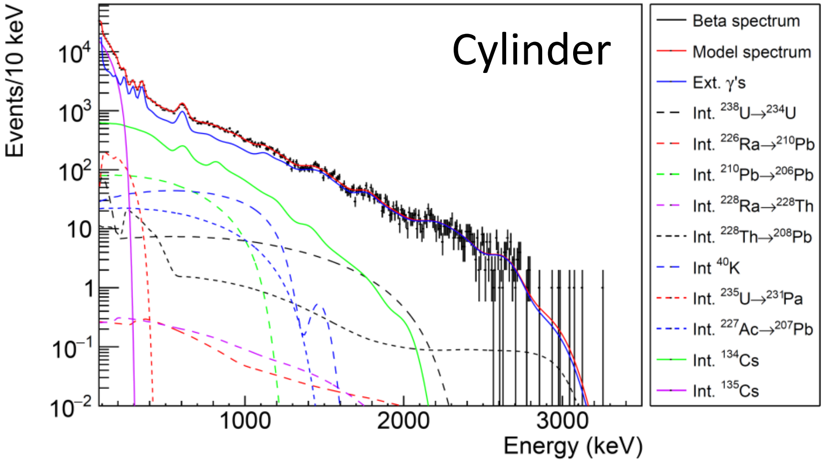
Alpha events





Beta/gamma events





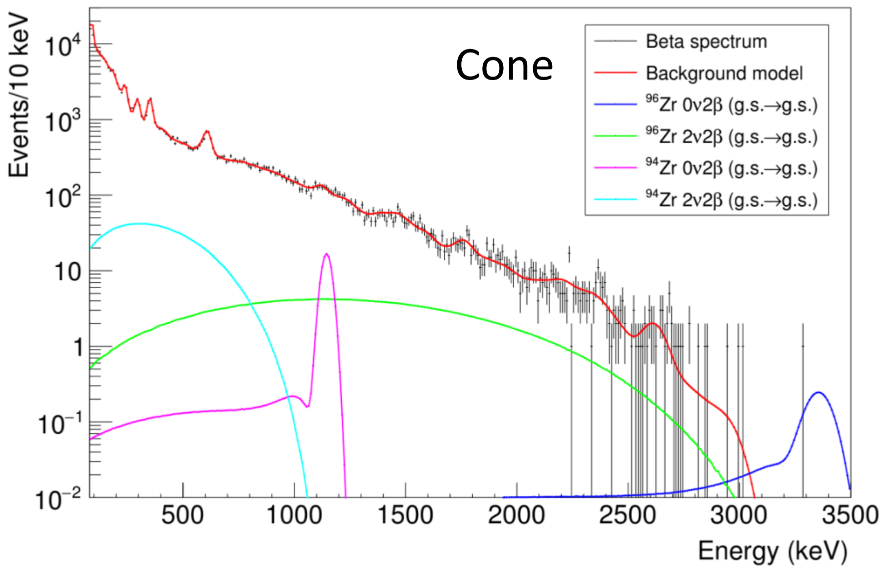
α/β ratio:
Cone: 0.2113(14) + 0.02607(27) × E_α [MeV]
Cylinder: 0.2109(19) + 0.02491(20) × E_α [MeV]

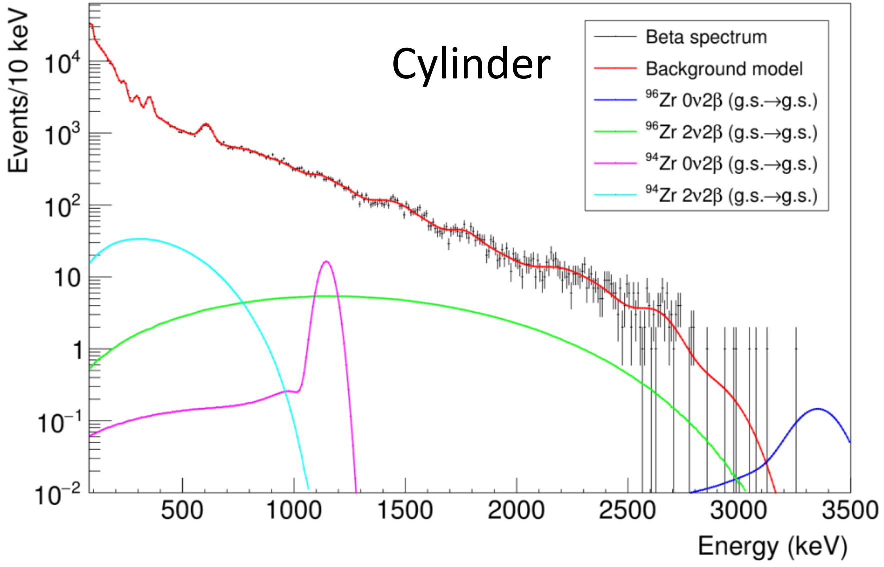
Contribution of external gammas from PMT's is dominant

Chain	Nuclide	Internal contamination, mBq/kg	
		Cone	Cylinder
^{232}Th	^{232}Th	0.07(2)	0.28(7)
	^{228}Th	0.05(2)	0.44(4)
^{235}U	^{235}U	0.29(4)	3.0(1)
	^{231}Pa	**21.0(3)**	**33.9(3)**
	^{227}Ac	0.70(3)	1.08(3)
^{238}U	^{238}U	0.53(4)	1.17(5)
	^{234}U	0.2(1)	3.8(1)
	^{230}Th	0.23(7)	< 0.02
	^{226}Ra	0.03(3)	0.12(3)
	^{210}Pb	2.2(2)	6.7(3)
	^{40}K	6(1)	5(1)
	^{134}Cs	36(4)	42(2)
	^{135}Cs	**267(4)**	**289(2)**

- Comply with measurements on HPGe
- High contamination by ^{235}U daughters
- Segregation of impurities is observed

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

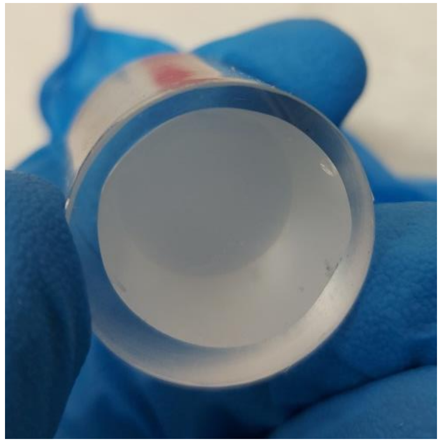


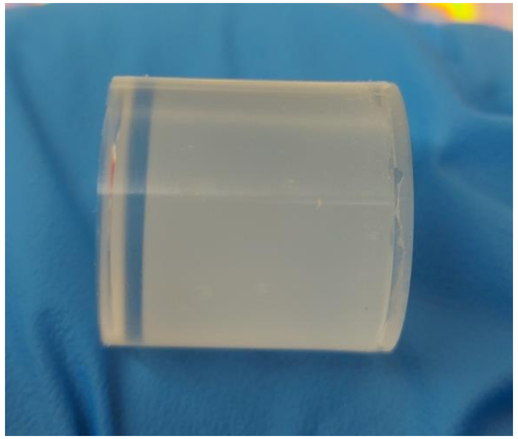


Transition	Decay mode	Final state of daughter nucleus, keV	Experimental limit on $T_{1/2}$ at 90%C.L., yr
^{96}Zr → ^{96}Mo	2β0ν	g.s.	**> 1.5×10^{20}**
		2_1^+, 778	> 1.5×10^{19}
	2β2ν	g.s.	> 7.4×10^{17}
		2_1^+, 778	> 3.8×10^{17}
	β	g.s.	> 1.0×10^{17}
^{94}Zr → ^{94}Mo	2β0ν	g.s.	> 2.6×10^{19}
		2_1^+, 871	> 3.8×10^{18}
	2β2ν	g.s.	> 2.4×10^{18}
		2_1^+, 871	> 1.9×10^{17}

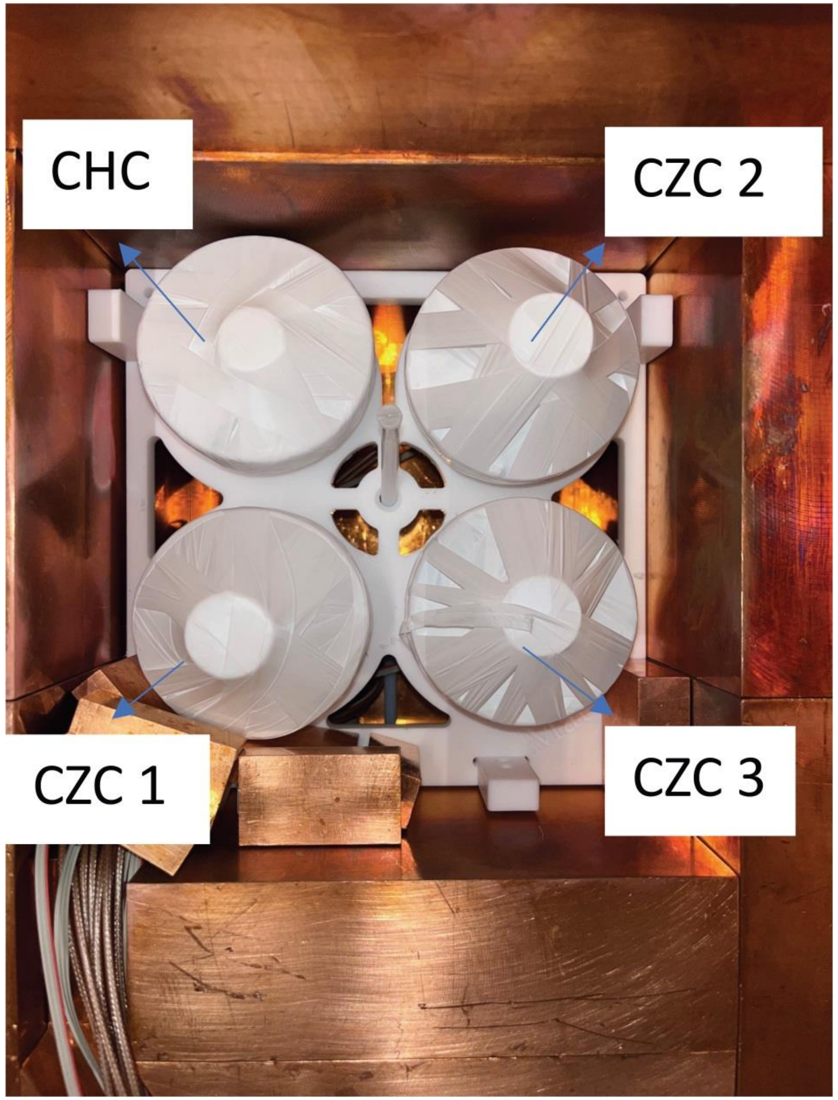
See more details in ***Eur. Phys. J. A 59 (2023) 176***
https://doi.org/10.1140/epja/s10050-023-01090-9

New low-background measurements in DAMA/CRYS setup

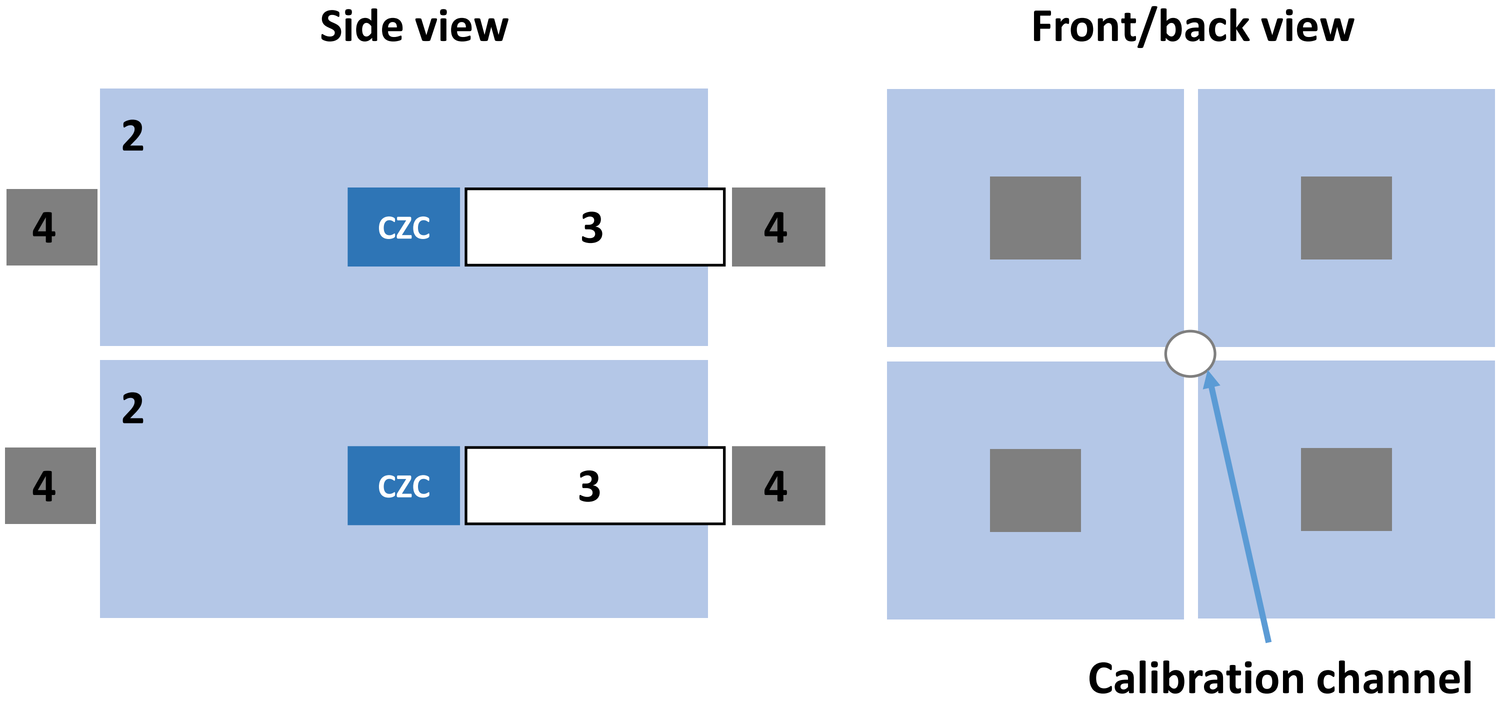


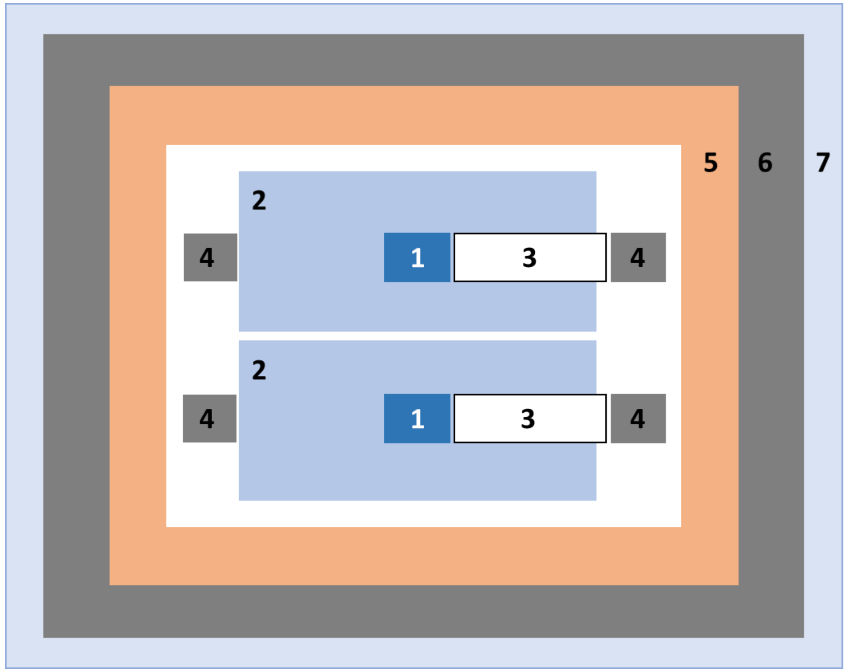


- Three new Cs_2ZrCl_6 crystals (more crystals are under production)
- Total mass = 59.5 g
- FWHM = 6-8% @ 662keV
- Produced from high purity and purified raw materials (> 99.99%)
- Crystals are encapsulated in a silicon-base resin + quartz window
- Modified experimental setup
- Measurements started in June 30th, 2023



Final detector array schematic ($T_{1/2} > 10^{22}$ yr)





Low-background setup, schematic view

<u>**Four separate detector's modules, each consist of:**</u>

(1) CZC ∅ 21×21 mm³
(2) Plastic scintillator block roughly 200×200×300 mm³
(3) Quartz light guide ∅ 25×(100-150) mm³
(4) 2" ultra-low-background PMTs

(5) OFHC Cu, 15 cm
(6) Pb, 20 cm
(7) HDPE, 10 cm
(8) 4π muon veto

Summary

- First experiment based on Cs_2ZrCl_6 scintillating crystals aiming to study 2β decay processes of $^{94,96}Zr$ isotopes within the "source = detector" approach was successfully realized

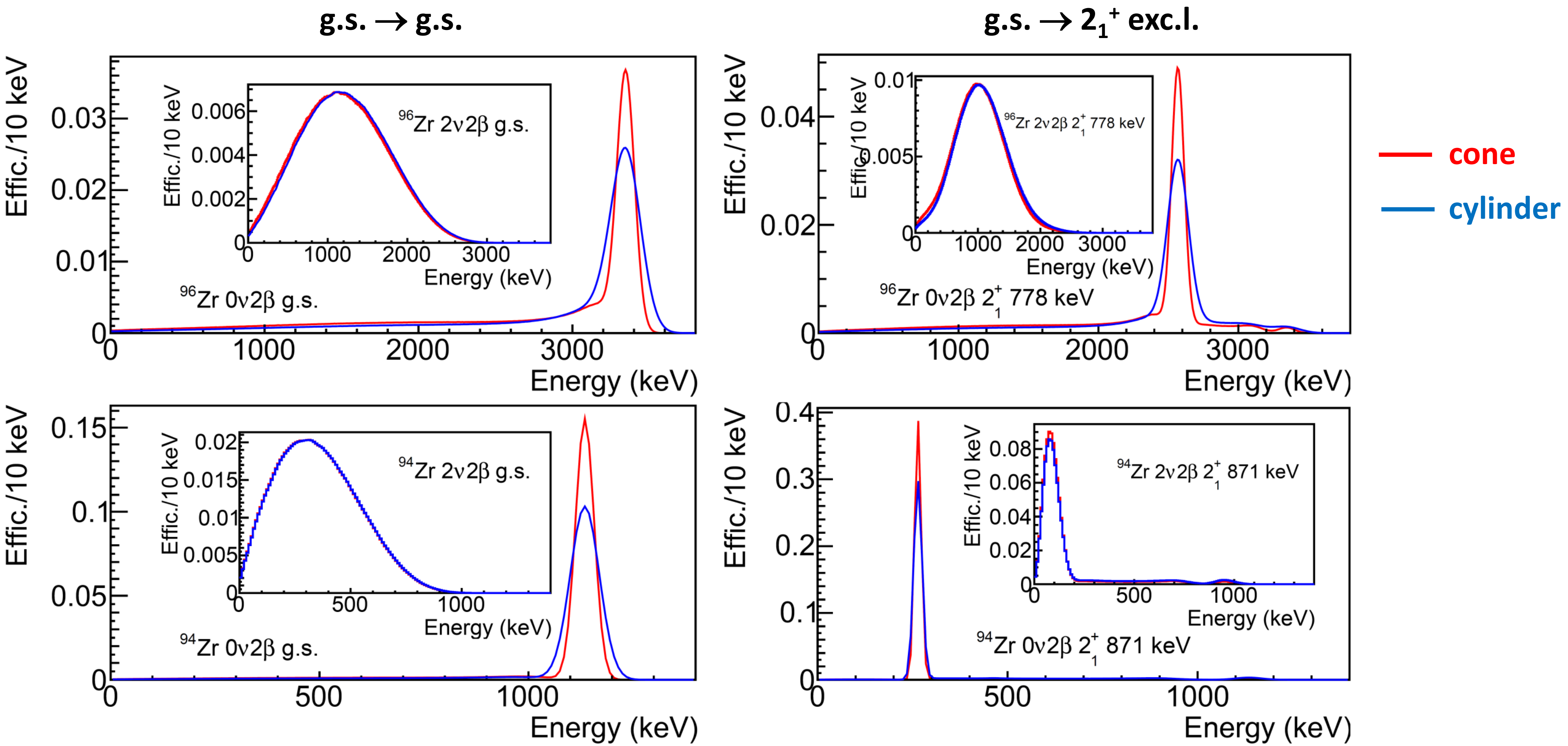
- Despite a very limited mass of the Cs_2ZrCl_6 detector (about 35 g) the experimental limits were established at the level of 10^{17}–10^{20} yr, depending on the decay mode

- A new experiment is ongoing with new Cs_2ZrCl_6 crystals (59.5 g) in an optimized geometry aiming to reach experimental sensitivity more than 10^{21} yr

- Extensive studies of Cs_2ZrCl_6 crystal scintillating performance, non-proportionality, internal and cosmogenically induced background, crystal lattice characteristics and phonon propagation properties, material handling and machining are on-going

- Cs_2ZrCl_6 crystal scintillators provide an unique opportunity to study rare decays of Zr isotopes with an ultimate experimental sensitivity

Simulated response functions of CZC crystals to DBD processes



Simulated response functions of CZC crystals to single β decay of ^{96}Zr