Search for neutrinoless double beta decay in ^{94,96}Zr isotopes using Cs_2ZrCl_6 crystal scintillators

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MEDEX'23
Matrix Elements for Double beta decay Experiments
Prague, 04 – 08 September 2023

LA PLATA ORSAY PRAGUE JYVÄSKYLÄ 14th MEDEX
September 4 – 8, 2023

Interest in studying the β and 2β decay

- **2β** decay without the presence of neutrinos, if observed, could open a new window beyond the Standard Model.

- The very accurate determination of the shape of the **β** spectrum at the end-point, can get information on the neutrino mass with direct and model-independent approach.

- The nuclear matrix elements for the **2ν** mode and for the **0ν** 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.

$$\beta^- : {}^A_Z X \rightarrow {}^{A}_{Z+1}Y + e^- + \bar{\nu}_e$$

$$2\nu2\beta^- : {}^A_Z X \rightarrow {}^{A}_{Z+2}Y + 2e^- + 2\bar{\nu}_e$$

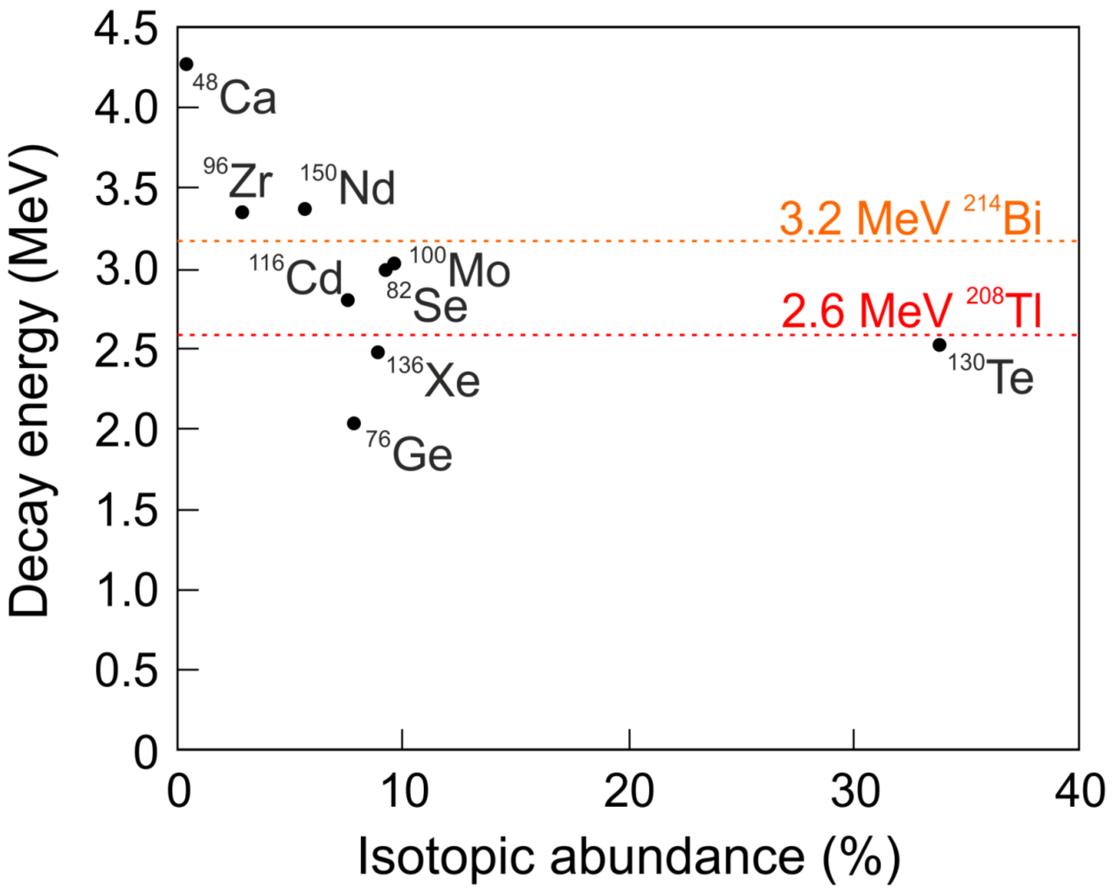
L conserved

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

L violated ($\Delta L = 2$) → massive Majorana neutrino

<u>2β and single β investigation with various nuclei would shed new light in constraining these and other important model-dependent parameters.</u>

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



^{76}Ge, ^{130}Te, ^{136}Xe are struggling 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 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.

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

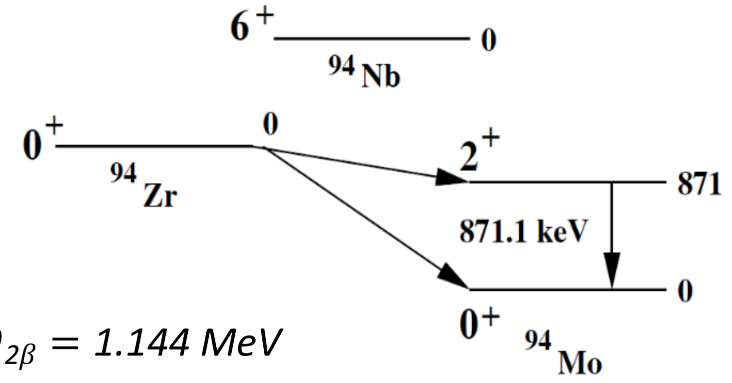
Our proposal:

$0\nu2\beta$ of ^{96}Zr with Cs_2ZrCl_6 scintillators at non-cryogenic temperature via "source = detector" experimental approach

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

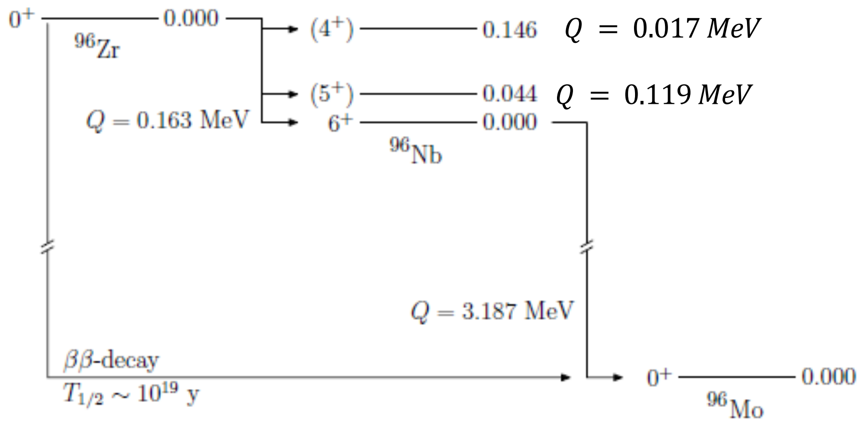
Experiment	Transition	$T_{1/2}$ @ 90% C.L. (yr)	Ref.	Technique
ZICOS (Kamioka Observatory, Japan)	$^{96}Zr \to {}^{96}Mo$ (g.s.)	under construction	[1]	Liquid scintillator
NEMO-3 (Frejus, France)	$^{96}Zr \to {}^{96}Mo$ (g.s.)	$> 9.2\times10^{21}$ $> 1.29\times10^{22}$	[2] [3]	Tracking detector
Kimballton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

Decay scheme of ^{94}Zr $\delta \sim 17\%$



$Q_{2\beta} = 1.144\ MeV$

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
[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

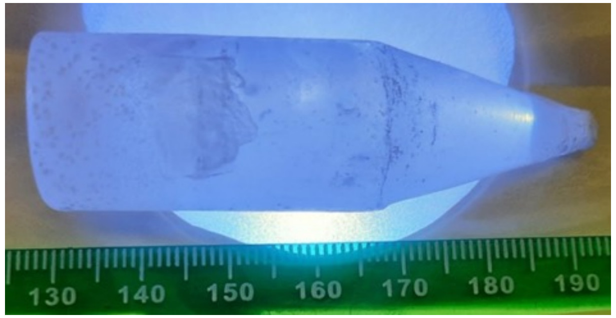
→ Possibility to study $0\nu4\beta$ decay of $^{96}Zr \to {}^{96}Ru$

The novel Cs_2ZrCl_6 (CZC) 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

Produced at Queen's University

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



∅21.5×60 mm, about 60 g

* *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*

CZC crystal radiopurity

measured with the ultra-low background **HP-Ge** γ spectrometers of the **STELLA** facility at LNGS over 700 hours

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 and installation

Natural

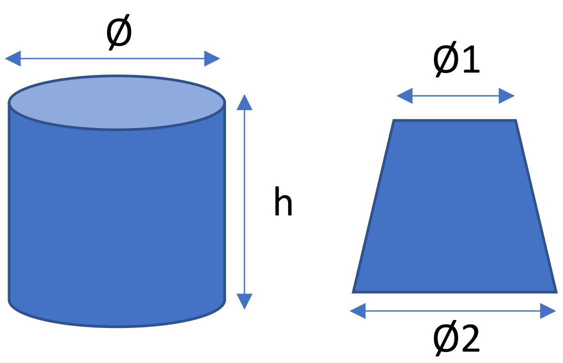
Artificial

Cosmogenic activation

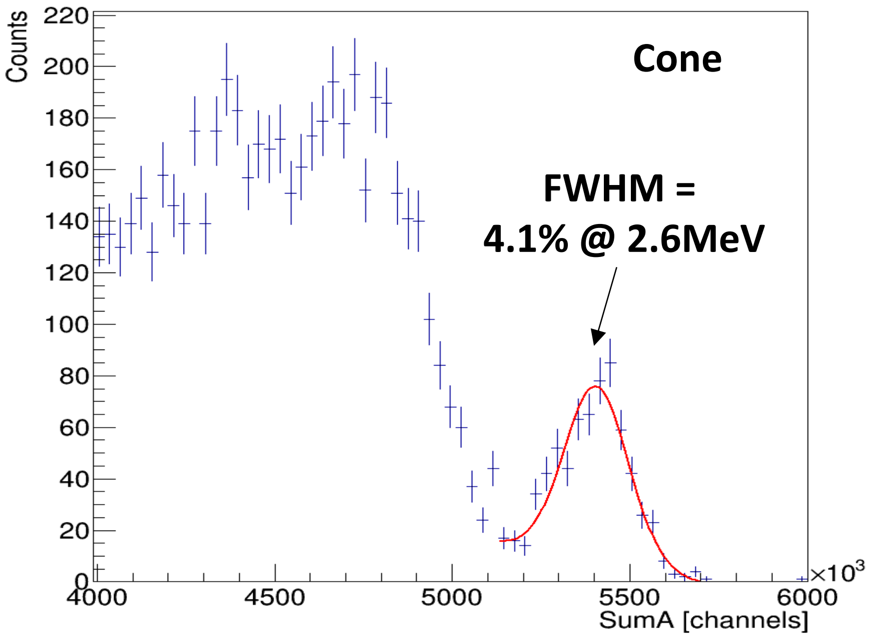
Only land transportation! $T_{1/2} \approx 2$ years

Low-background measurements at LNGS (Italy)

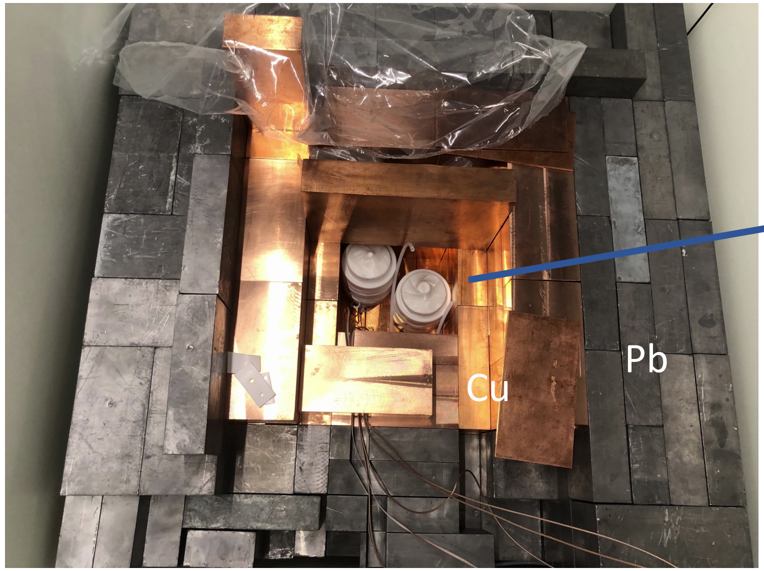
M=24,0(1) g
h =21,20(5) mm
Ø=21,00(5) mm



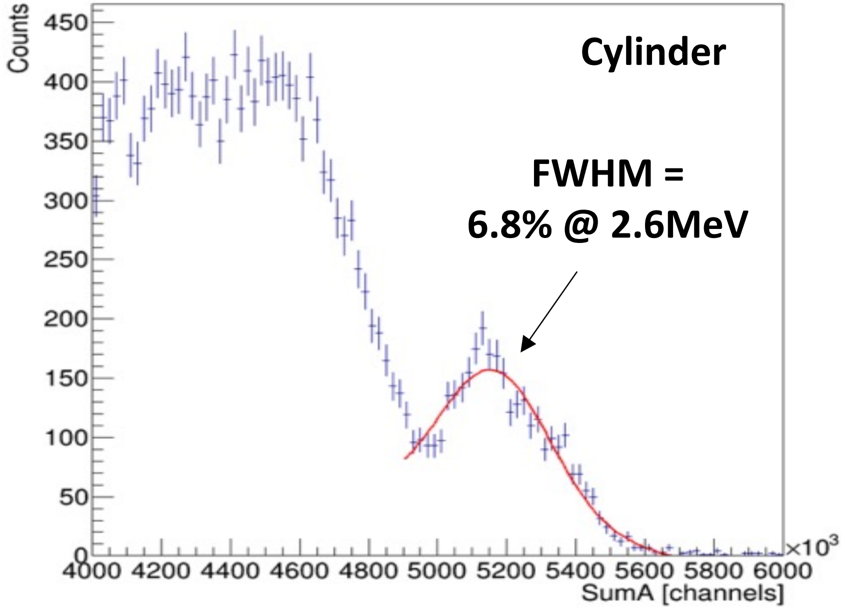
M=10,6(1) g
h =17,90(5) mm
Ø1=8,0(1) mm
Ø2=19,70(5) mm



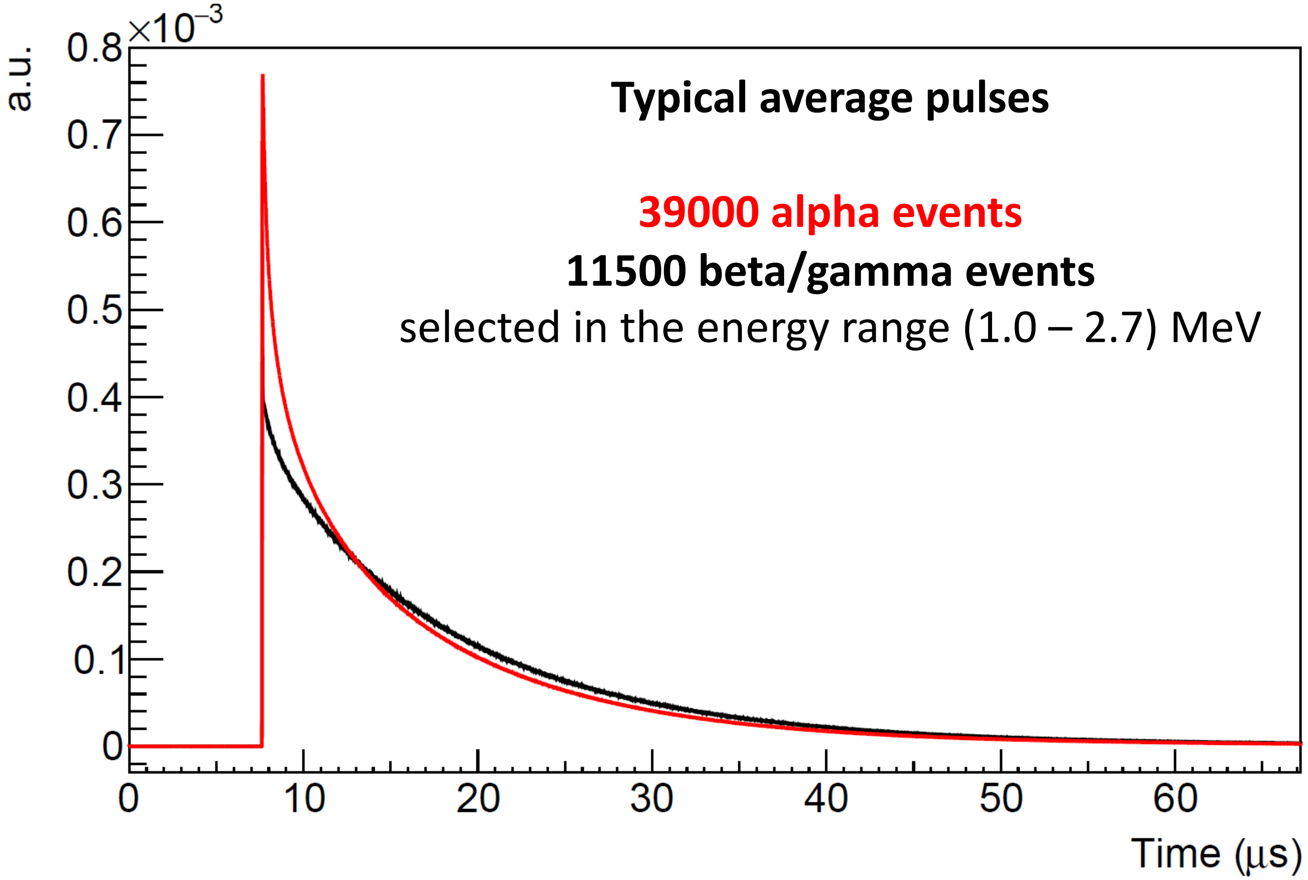
- ✓ Run 1: 456.5 days of data taking (time-window 80 μs), July 2021 - Oct 2022
- ✓ Run 2: 65 days of data taking (extended time-window for t-A analysis, 2 ms), Oct -Dec 2022



DAMA/CRYS setup at LNGS



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" ($\langle t \rangle$) method [2] was used, and this parameter was determined according to:

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

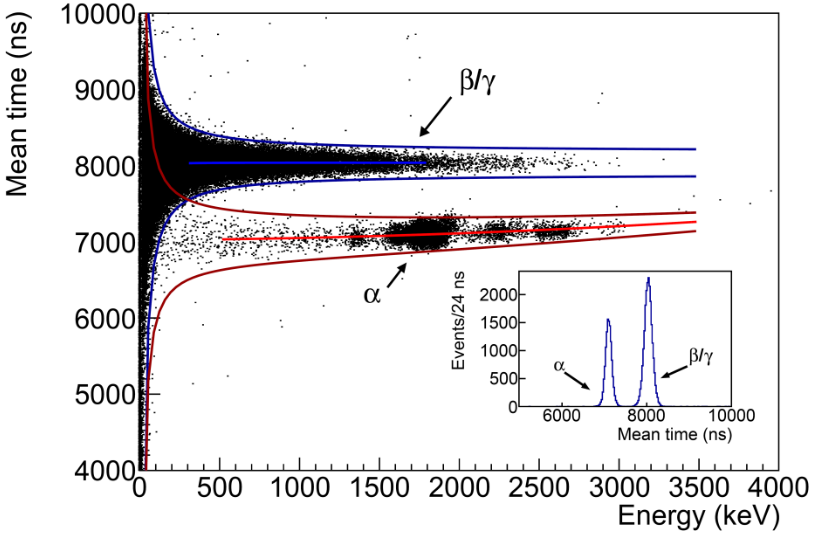
where the sum is over the time channels (k), starting from the origin of pulse up to 24 μs, $f(t)$ is the digitized amplitude (at the time t) of a given signal.

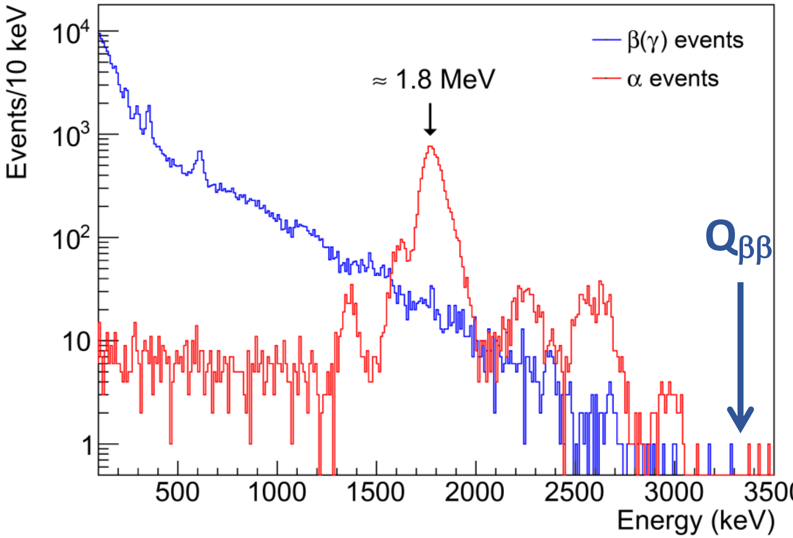
Mean-time for the presented pulses are:
$\langle t \rangle$ = 7.07 and 8.00 μs, for alpha and beta/gamma events respectively

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

Pulse-shape discrimination and background α event selection

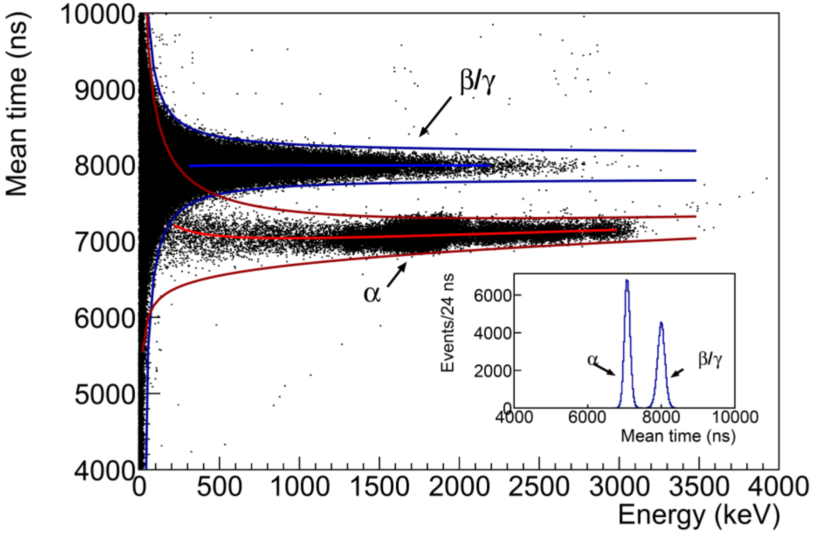
Cone
FoM = 7.8

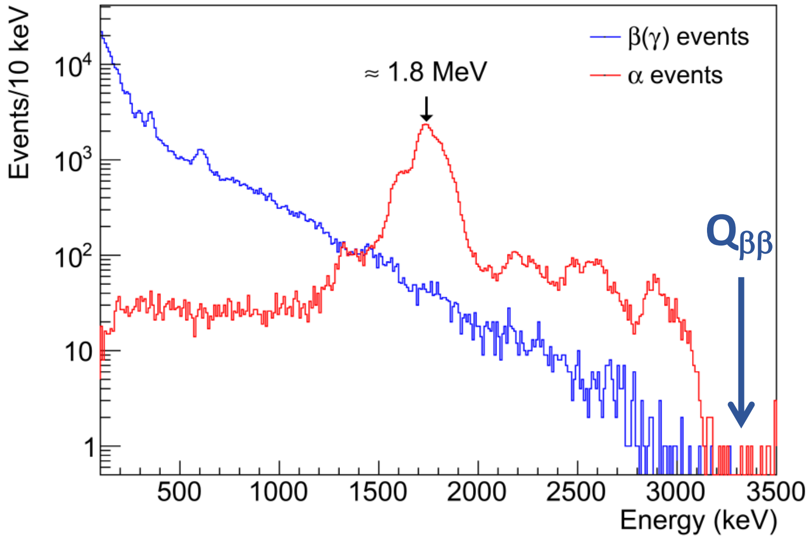




Selection efficiency is 99.7% in [0.1–3.5] MeV

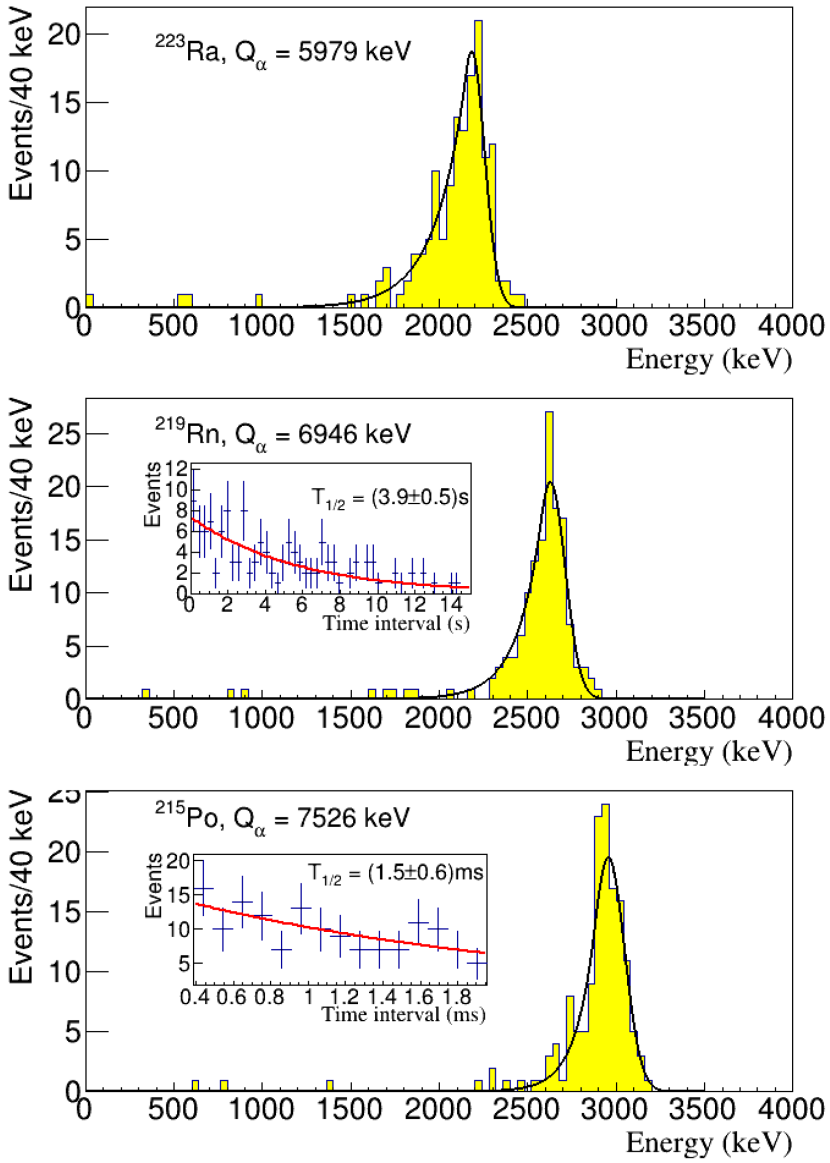
Cylinder
FoM = 7.2





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)

↓

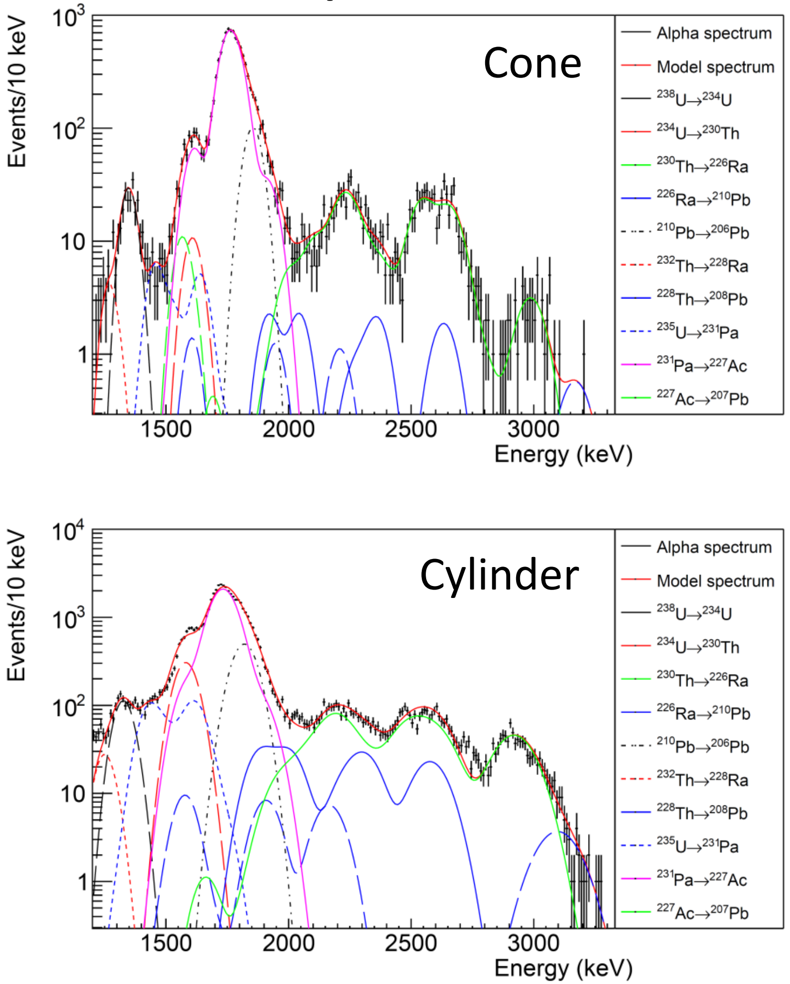
^{211}Pb

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

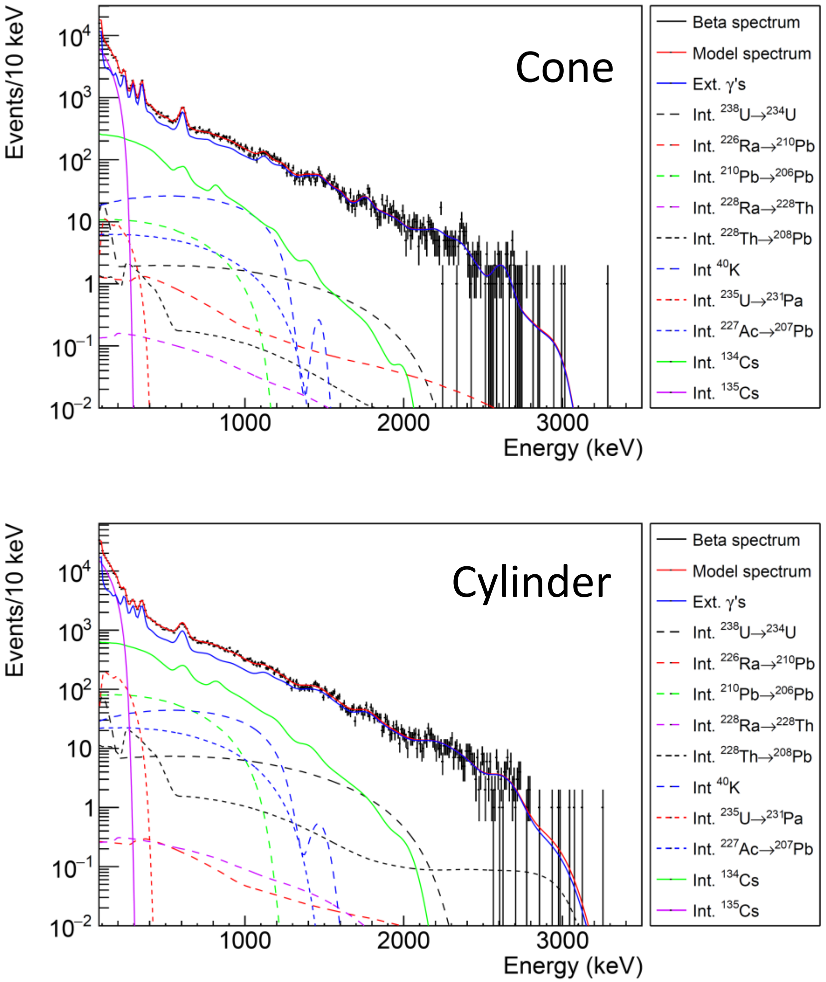
+ *Confirmation of ^{235}U decay chain presence*
+ *Alpha peaks to precisely determine α/β ratio*

Background model

Alpha events



Beta/gamma events



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)**

α/β light ratio:

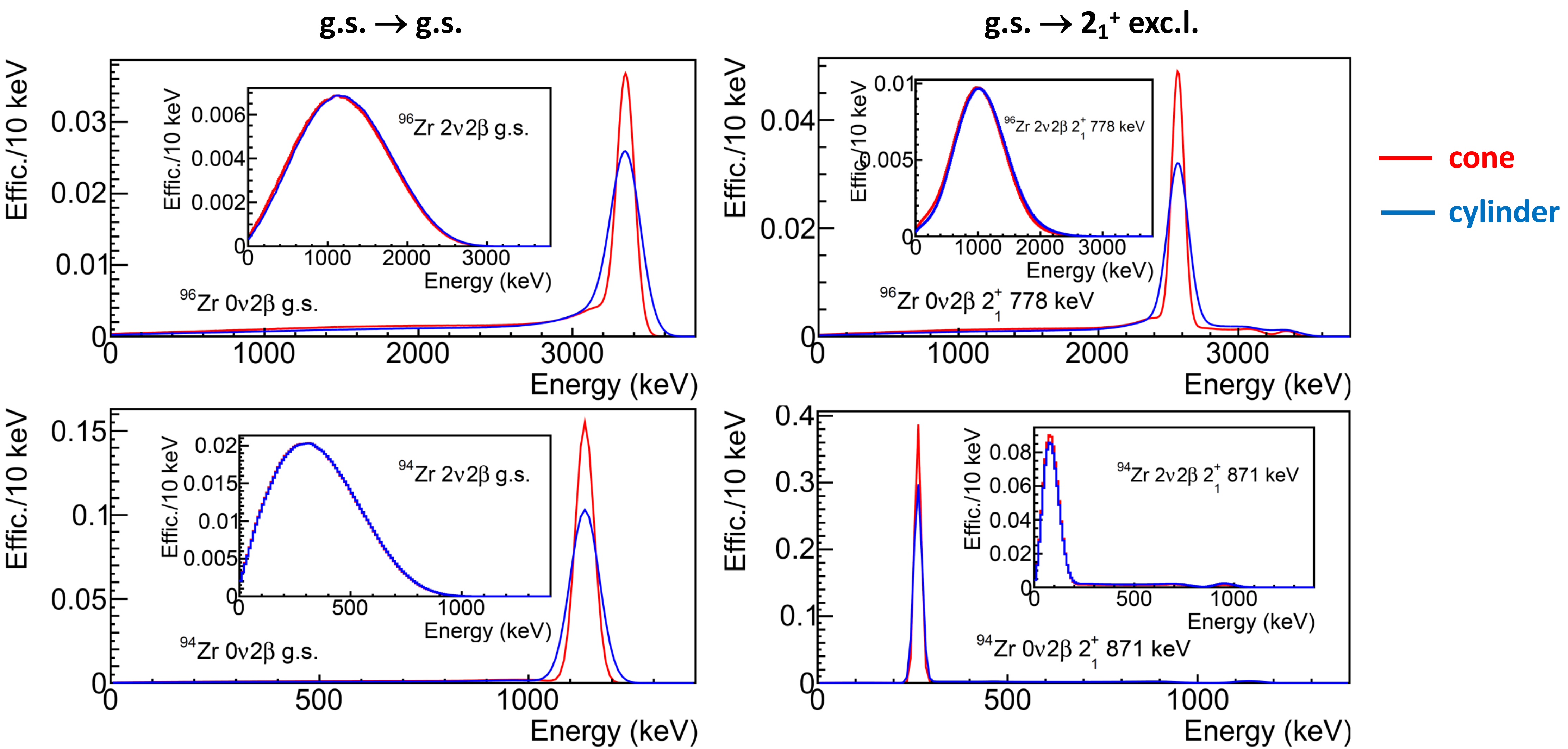
Cone: α/β = 0.2113(14) +0.02607(27)·E_{α}[MeV]

Cylinder: α/β = 0.2109(19)+ 0.02491(20)·E_{α}[MeV]

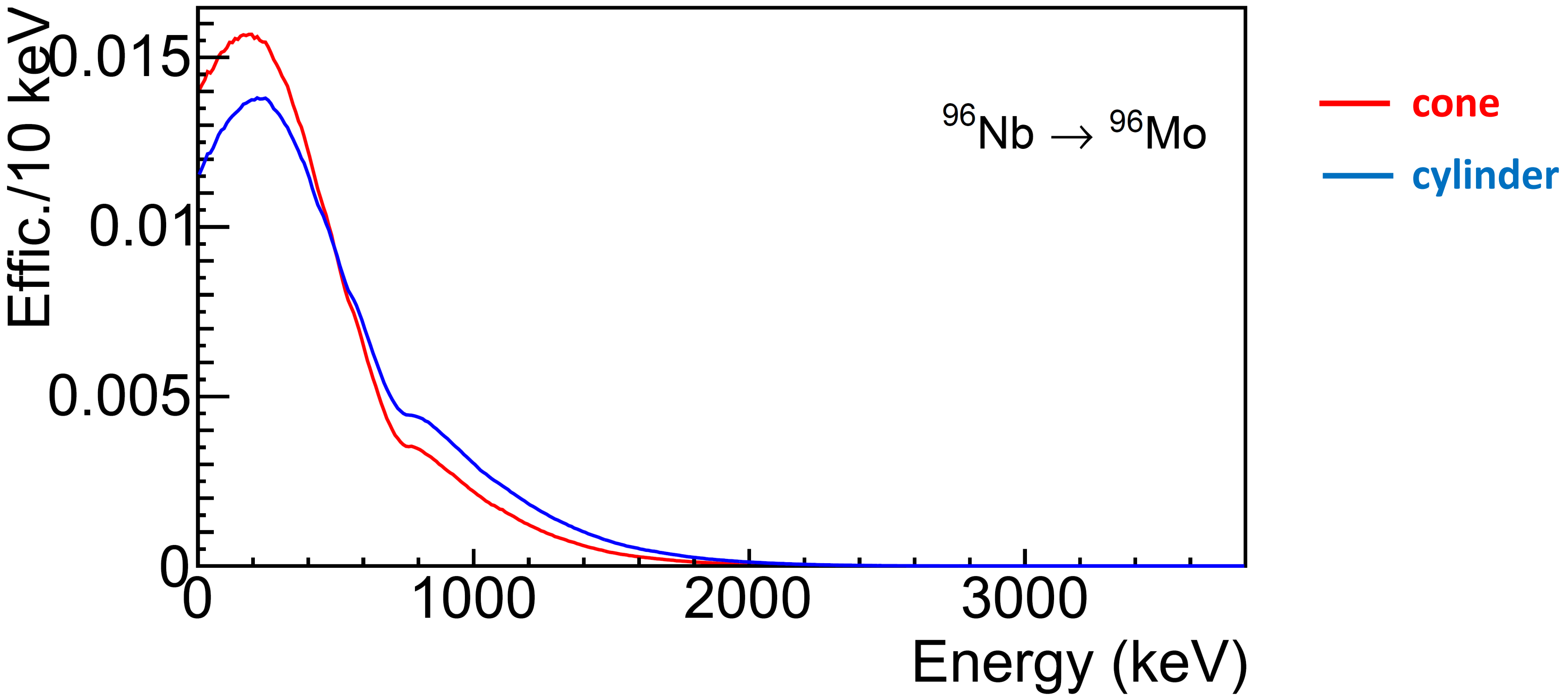
Contribution of external gammas from PMT's is dominant

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

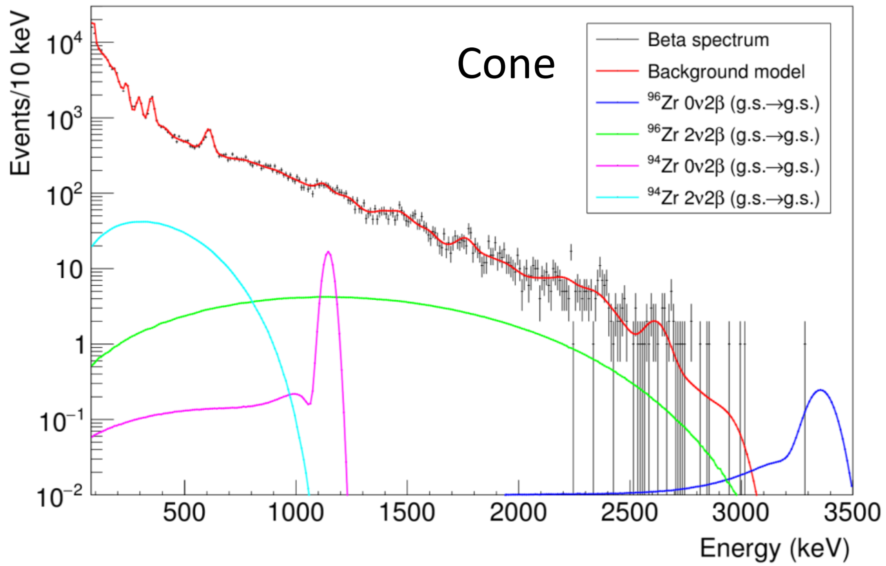
Simulated response functions of CZC crystals to 2β processes

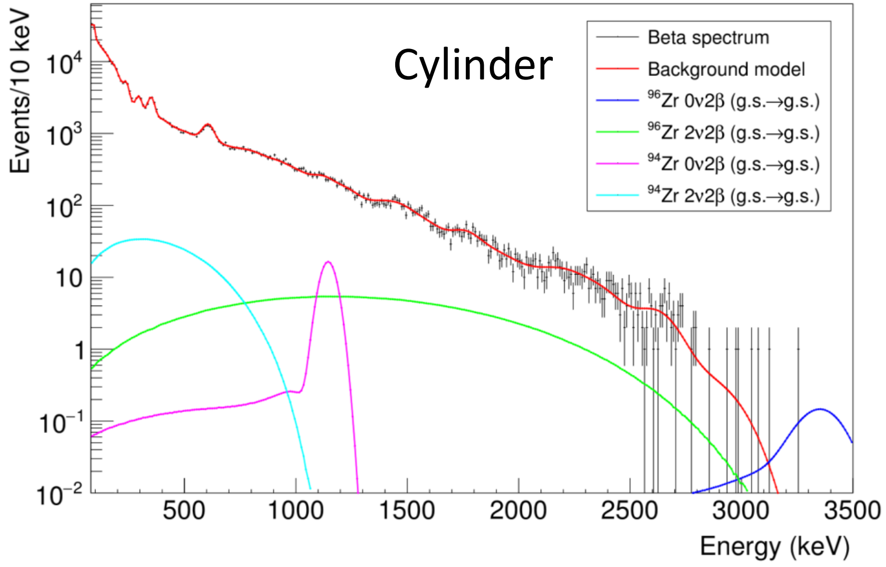


Simulated response functions of CZC crystals to single β decay of ^{96}Zr



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

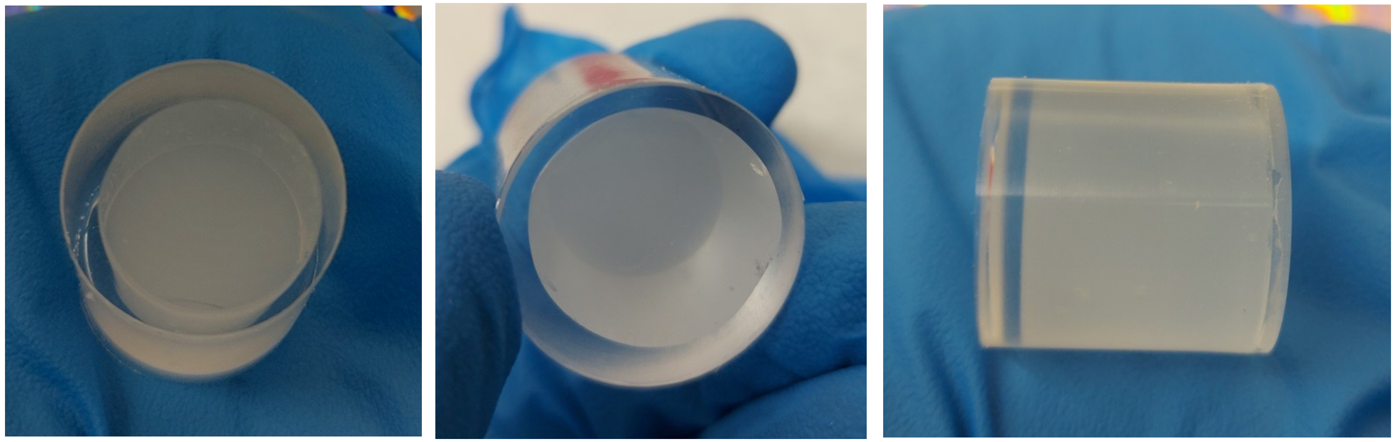


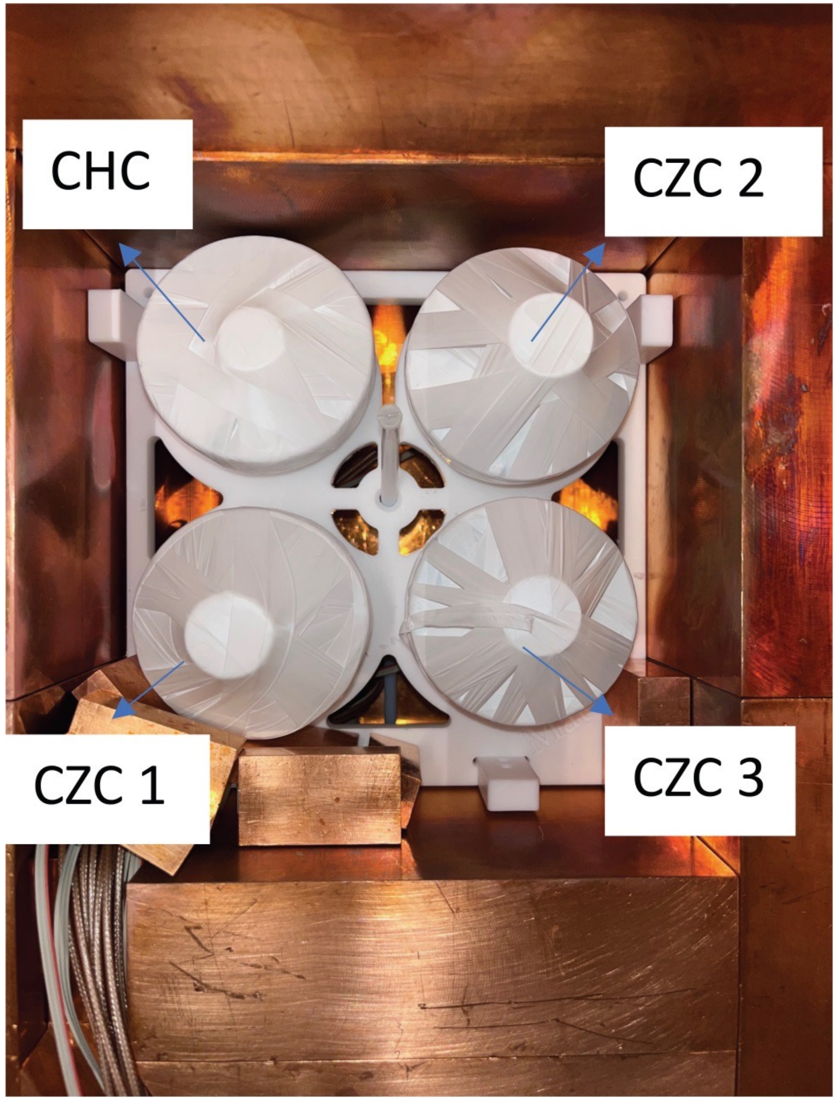


Transition	Decay mode	Final state of daughter nucleus, keV	Experimental limit on $T_{1/2}$ at 90%C.L., yr
$^{96}Zr \rightarrow ^{96}Mo$	$0\nu2\beta$	g.s.	**$> 1.5\times10^{20}$**
		2_1^+, 778	$> 1.5\times10^{19}$
	$2\nu2\beta$	g.s.	$> 7.4\times10^{17}$
		2_1^+, 778	$> 3.8\times10^{17}$
	β	g.s.	$> 1.0\times10^{17}$
$^{94}Zr \rightarrow ^{94}Mo$	$0\nu2\beta$	g.s.	**$> 2.6\times10^{19}$**
		2_1^+, 871	$> 3.8\times10^{18}$
	$2\nu2\beta$	g.s.	**$> 2.4\times10^{18}$**
		2_1^+, 871	$> 1.9\times10^{17}$

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 (LNGS)





- 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

Conclusions

- 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 a unique opportunity to study rare decays of Zr isotopes and retrieve important information as g_A, shape of β and 2β energy spectrum, matrix elements, etc..