Istituto Nazionale di Fisica Nucleare

Study for rare processes in naturally occurring Zr isotopes using Cs_2ZrCl_6 crystal scintillators

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Interest in studying the 2β decay

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

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

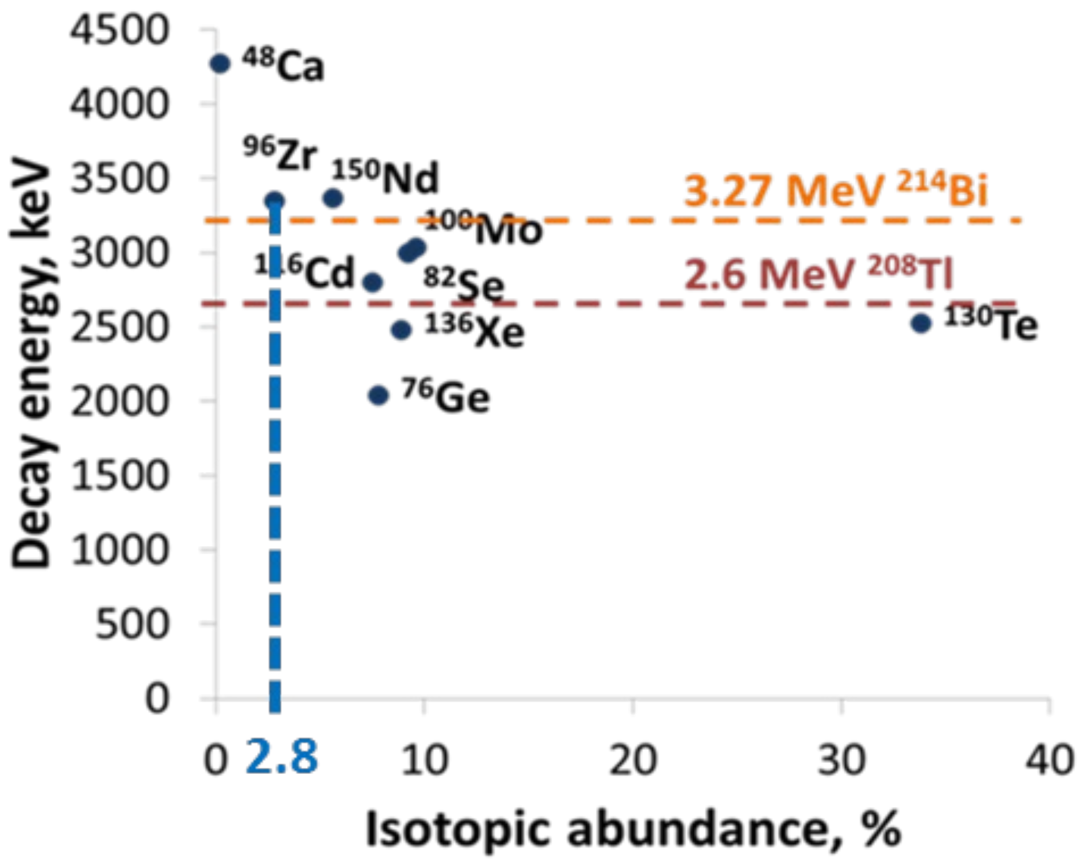
$$2\nu 2\beta^- :\ {}^{A}_{Z}X \rightarrow {}^{A}_{Z+2}Y + 2e^- + 2\bar{\nu}_e$$

L conserved

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

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

$0\nu2\beta$ 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.

Our proposal:

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

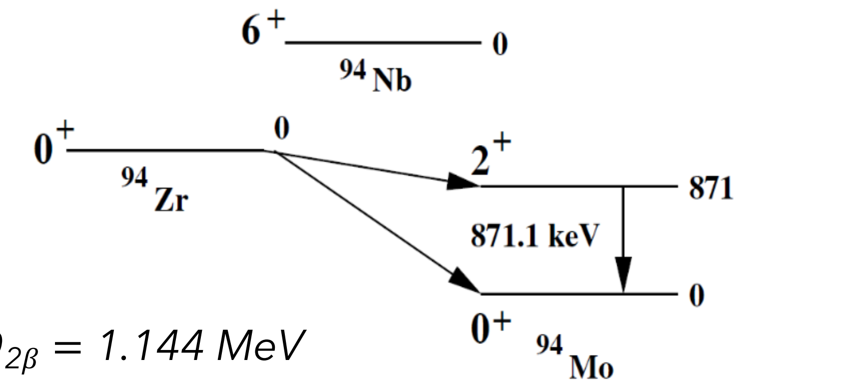
- *$Q_{2\beta}$ (^{96}Zr) = 3.35 MeV*
- *Favorable from a theoretical point of view $T_{1/2} \sim (Q_{2\beta})^5$*
- *Reasonable natural isotopic abundance*
- *About 15 g of enriched ^{96}Zr (55%) is available*
- *New advanced detector material (Cs_2ZrCl_6)*
- *Crystal production under full control*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

Investigation of 2β decay in $^{94,96}Zr$ and for ^{96}Zr's β decay

Experiment	Transition	$T_{1/2}$ 90% C.L. (y)	Ref.	Technique
ZICOS, (Kamioka Observatory, Japan)	$^{96}Zr\ 0^+ \rightarrow {}^{96}Mo\ 0^+_1$ (g.s.)	under construction (supported by Grant-in-Aid for Scientific Research on Innovative Areas 26105502)	[1]	Organic liquid scintillator (almost similar structure as KamLAND-Zen detector)
NEMO I, II, III, Frejus (France) (next: SuperNEMO)	$^{96}Zr\ 0^+ \rightarrow {}^{96}Mo\ 0^+_1$ (g.s.)	$>9.2\times10^{21}$ $>1.29\times10^{22}$	[2] [3]	Tracker detector
Kimballton Underground Research Facility, (USA)	$^{96}Zr\ 0^+ \rightarrow {}^{96}Mo\ 2^+_1$	$>3.1\times10^{20}$	[4]	HP-Ge
Collaboration at Fréjus Underground Laboratory	$^{96}Zr\ 0^+ \rightarrow {}^{96}Mo\ 2^+_1, 0^+_1, 2^+_2, 2^+_3$	$>(2.6 - 7.9)\times10^{19}$	[5]	HP-Ge
Collaboration at LNGS	$^{96}Zr\ 0^+ \rightarrow {}^{96}Mo\ 2^+_1$	$>3.8\times10^{19}$	[6]	HP-Ge
TILES (TIFR, Mumbai)	$^{94}Zr\ 0^+ \rightarrow {}^{94}Mo\ 2^+_1$	$>5.2\times10^{19}$	[7]	HP-Ge
Kimballton Underground Research Facility, (USA)	$^{96}Zr\ 0^+ \rightarrow {}^{96}Mo\ 6^+$	$>2.4\times10^{19}$	[8]	HP-Ge

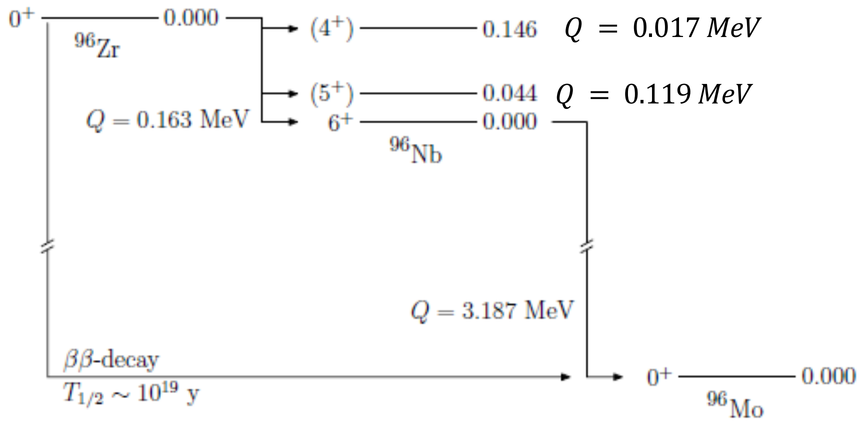
- Possibility to study $0\nu4\beta$ decay of $^{96}Zr \rightarrow {}^{96}Ru$ [9].

Decay scheme of ^{94}Zr



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

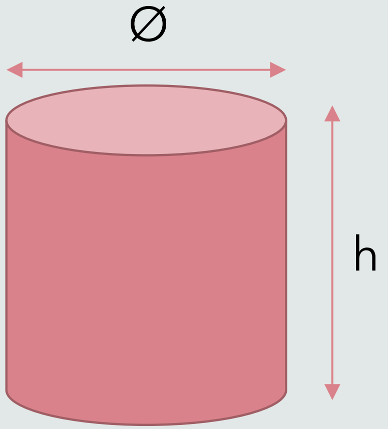
β and 2β decay of ^{96}Zr. The decay Q-values and excitation energies of the first three states of Nb are also indicated.

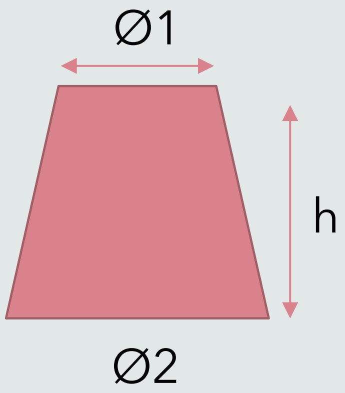


[1] EPS-HEP (2019) 437
[2] NPA 847 (2010) 168
[3] PhD U. Coll. London (2015)
[4] S.W. Finch et W. Tornow, Phys, Rev. C 92 (2015) 045501
[5] J. Phys. G: Nucl. Part. Phys. 22 (1996) 487
[6] C. Arpesella et al. Lett. 27 (I) (1994) pp. 29-34
[7] N. Dokania et al. J. Phys. G: Nucl. Part. Phys. 45 (2018) 075104
[8] S.W. Finch, W. Tornow, Nucl. Inst. Meth. A 806(2016)70-74
[9] J. Heeck and W. Rodejohann 2013 *EPL* 103 32001

The CZC crystal scintillator

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



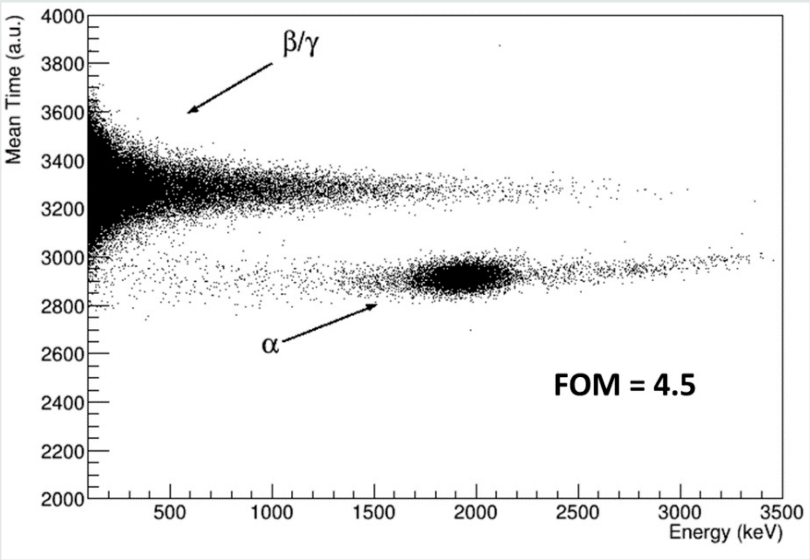


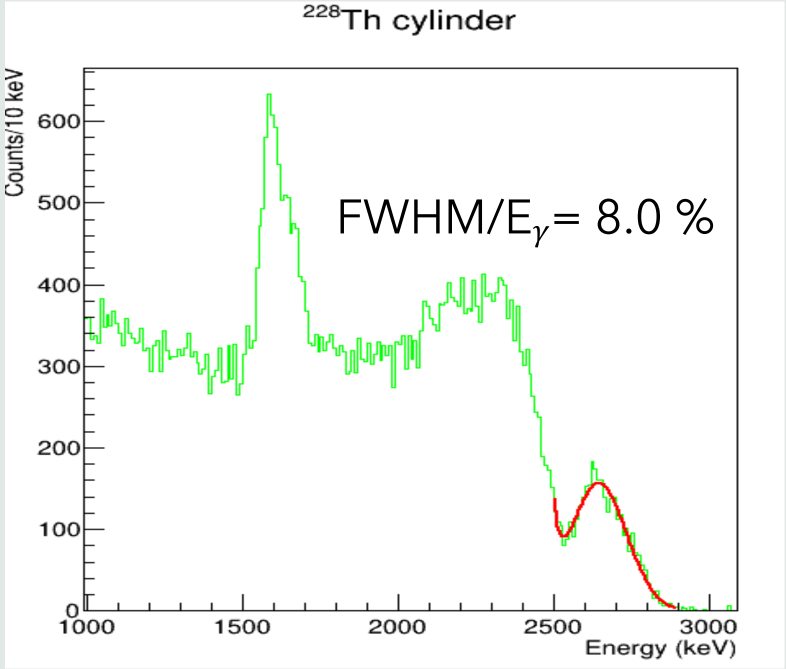
M=10,6(1) g
h =17,90(5) mm
Ø1=8,0(1) mm
Ø2=19,70(5) mm

It represents one of the promising new scintillating materials for γ spectroscopy:

- High light yield (up to 40000 photons/MeV);
- 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 $\boldsymbol{\beta(\gamma)}$ and $\boldsymbol{\alpha}$ particles;
- The Cs_2ZrCl_6 scintillating crystals are of particular interest as they are the first scintillating material containing 16% Zr by weight.

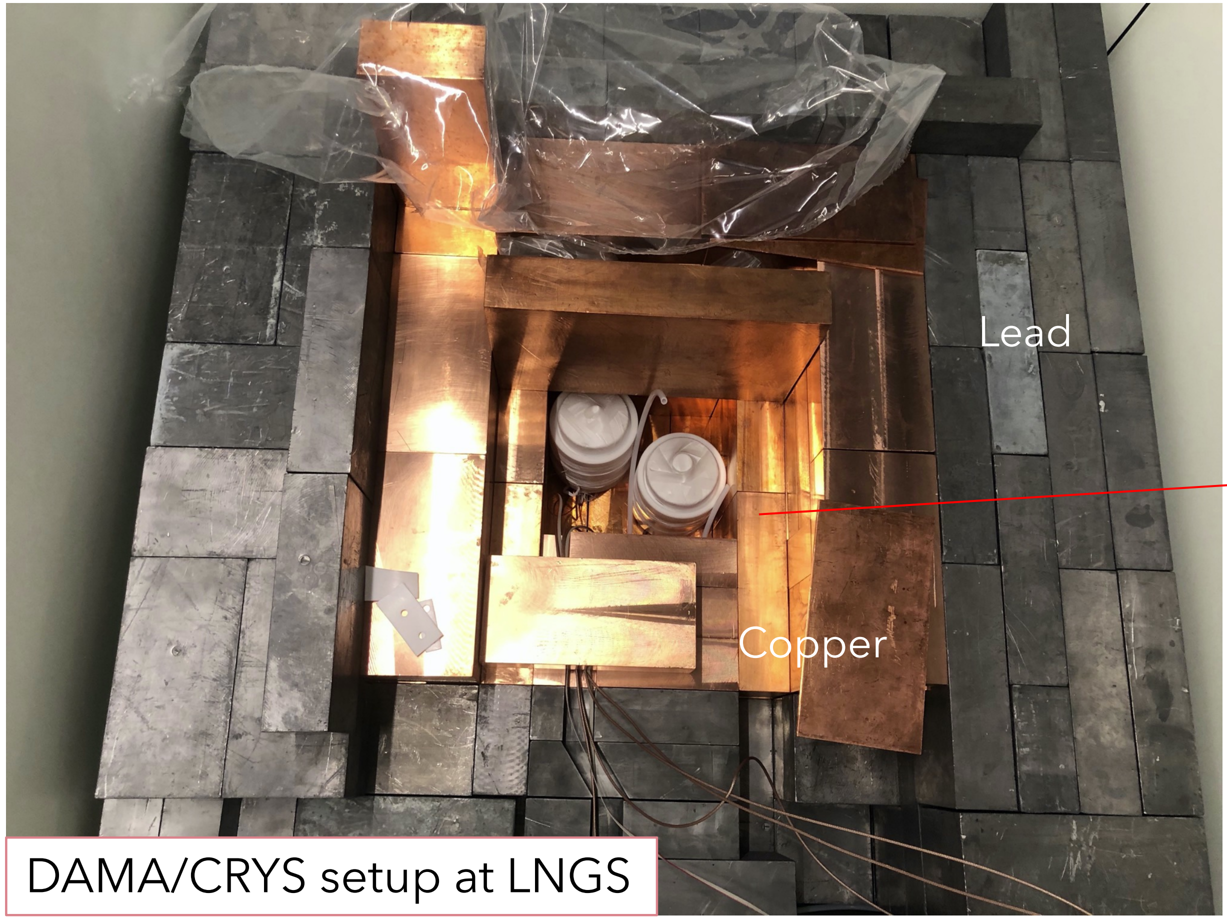
Excellent PSD



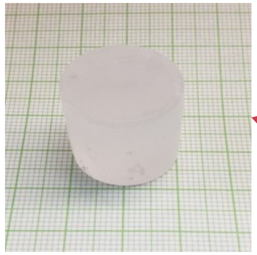


CZC low-background measurements at LNGS (Italy)

started June 21st, 2021



Cs_2ZrCl_6 crystals



CZC crystal radiopurity

over 700 hours of low-background measurements on HPGe detector

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

Under investigation (arrow to ^{226}Ra, Cone: 60±10)

Natural (^{228}Ra through ^{40}K)

Artificial (^{137}Cs)

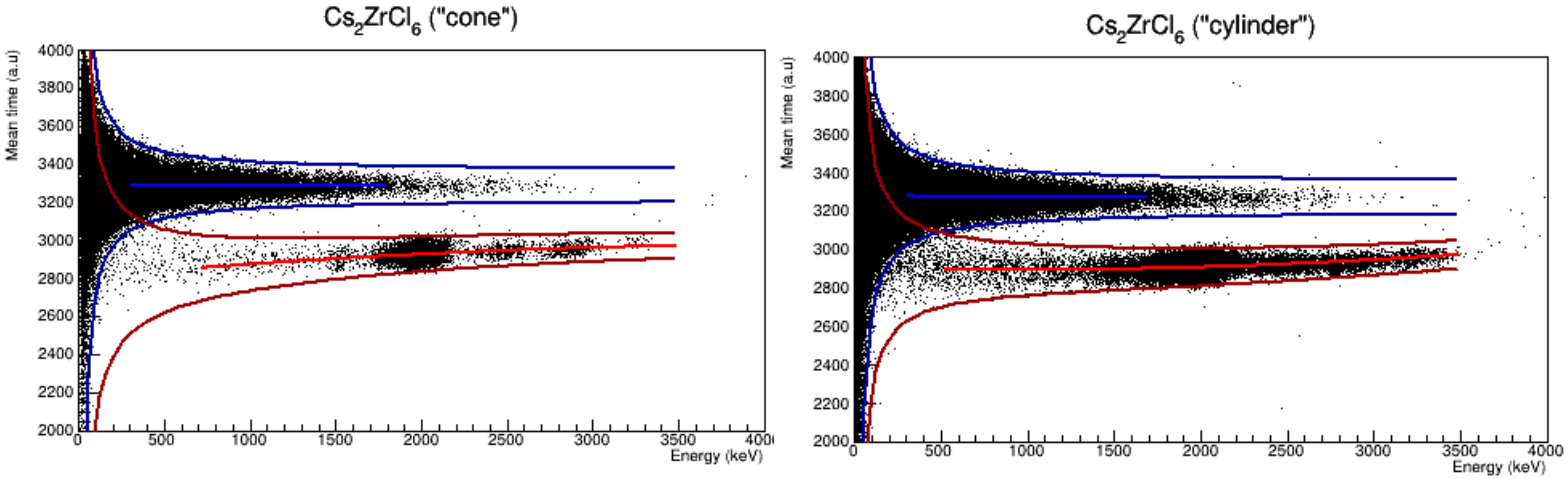
Cosmogenic activation (^{134}Cs, ^{132}Cs)

Only land transportation! $T_{1/2} \approx 2$ years (arrow to ^{134}Cs)

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

Data analysis

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



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

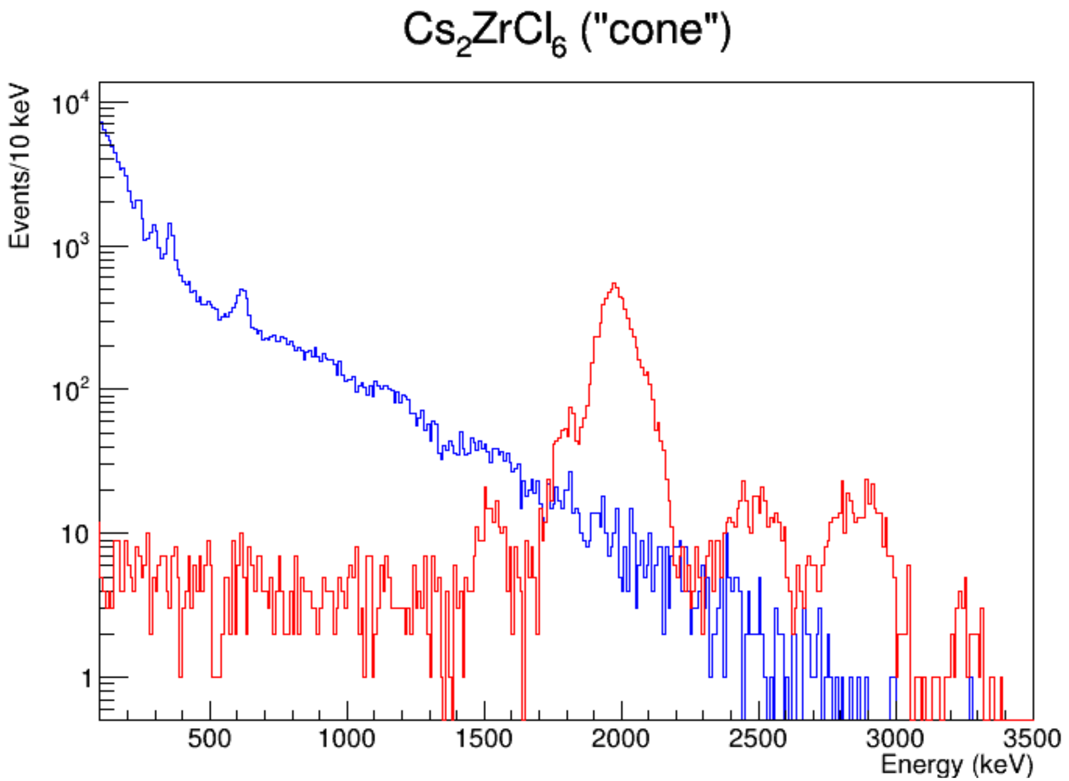
[10] L. Bardelli *et al.*, Nucl. Instr. Meth. A **584**, 129 (2008).

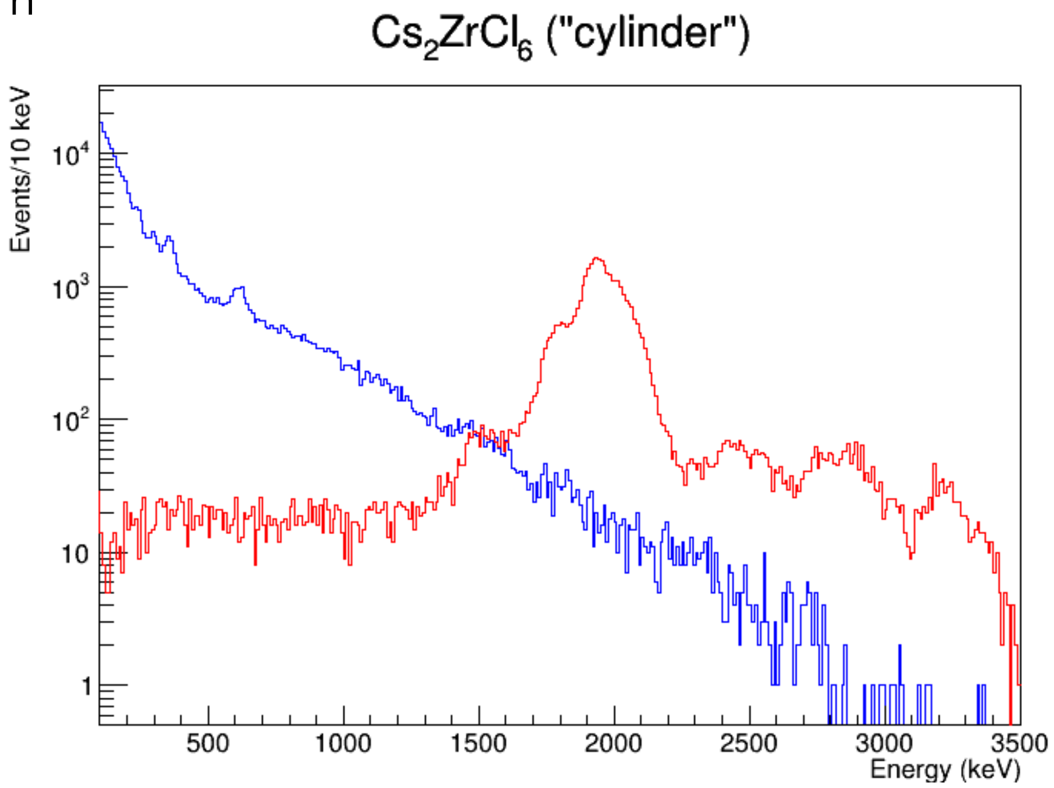
Red: alpha
Blue: β/γ

Measured spectra

With the selection on α given by the PSD

T = 8736 h





Cone crystal:

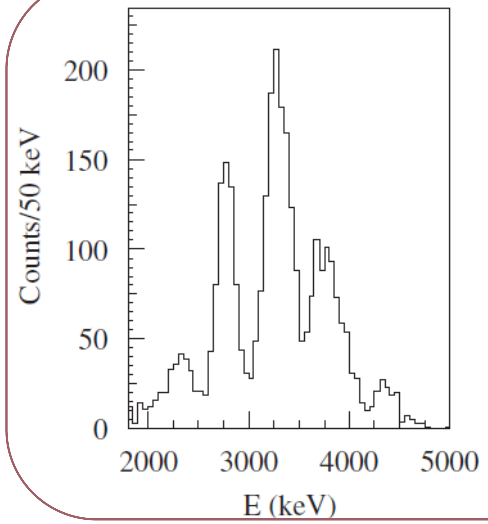
- 9581 α events in [0.1, 4.0] MeV ⇒ 2480 α/kg/day
- Degraded α events in [0.1, 1.4] MeV = 568 (5.9%)

Cylinder crystal:

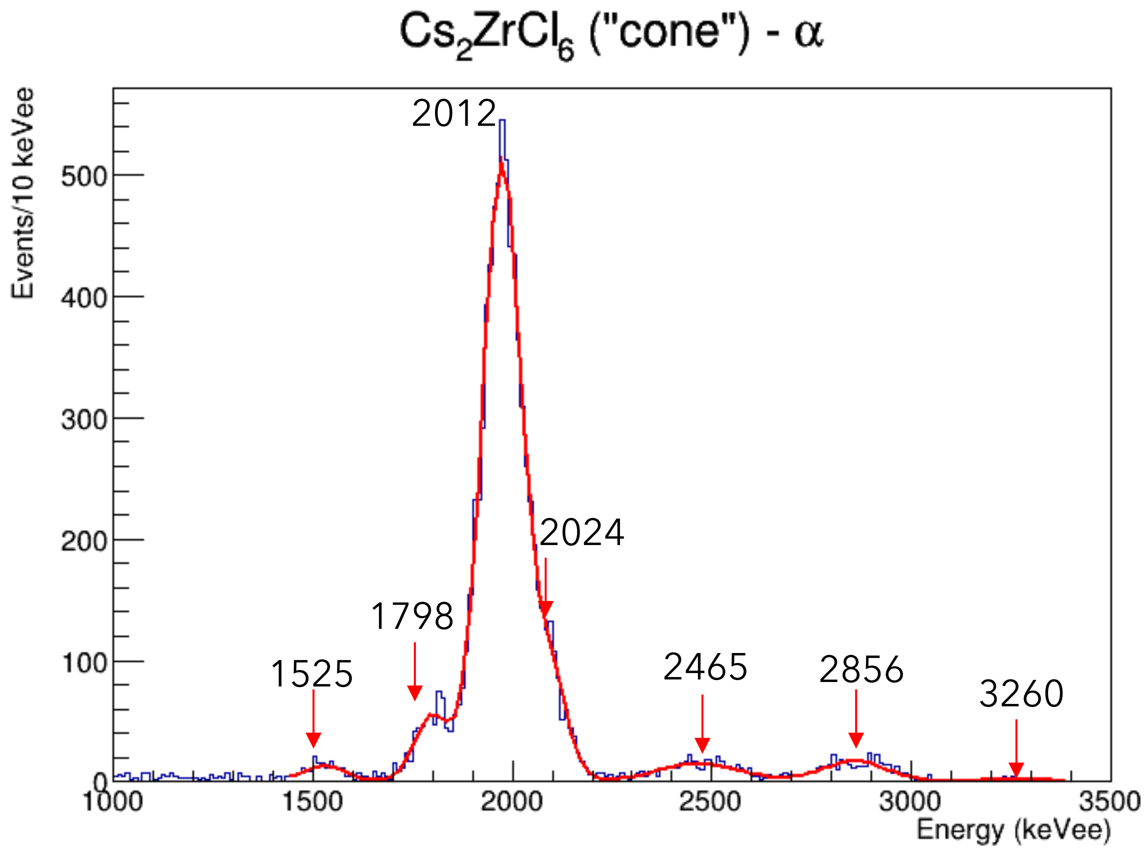
- 43820 α events in [0.1, 4.0] MeV ⇒ 5020 α/kg/day
- Degraded α events in [0.1, 1.4] MeV = 2428 (5.5%)

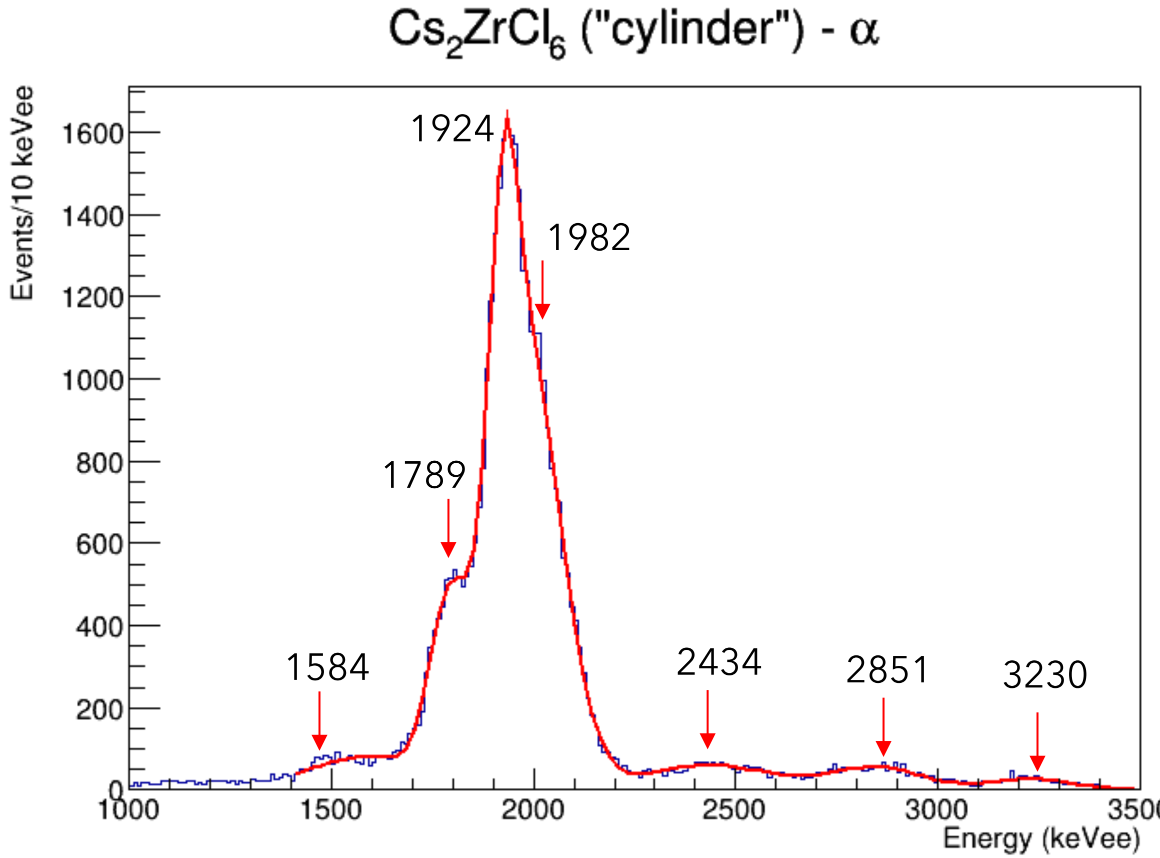
About the α spectra

- The alpha spectra measured in the 2 crystals are very similar in shape
- The spectra seem to have 7 α peaks
- But the counting rate of peak at ≈ 2 MeVee is much higher than the others



Example of α spectrum in LIBRA (only 5 peaks from ^{238}U and ^{232}Th)

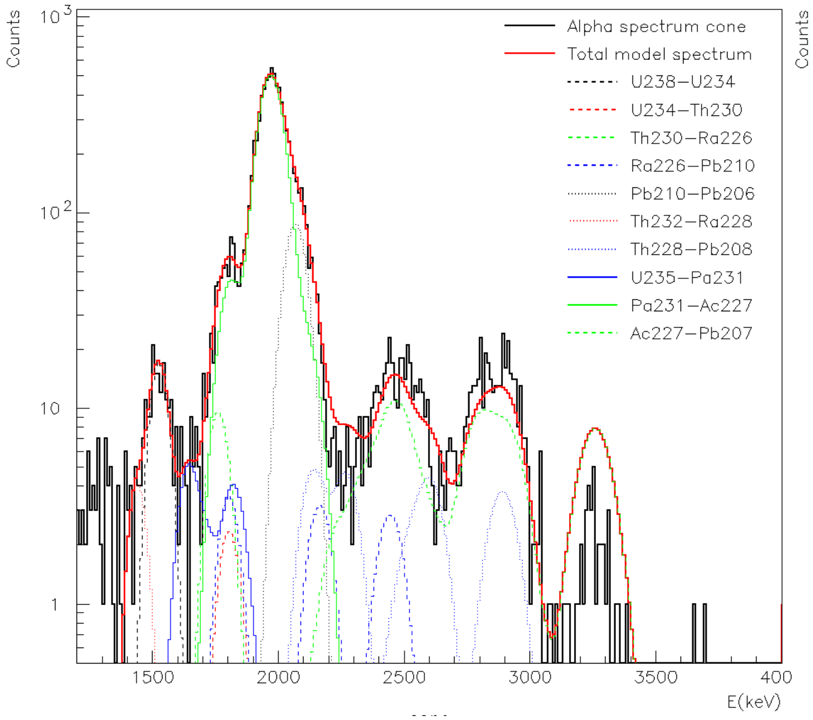




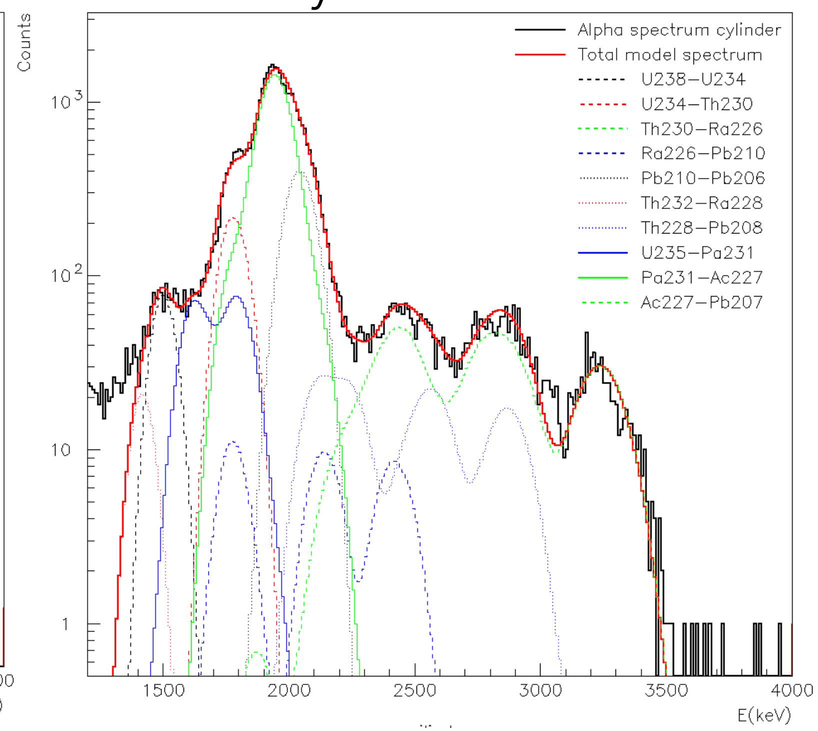
Fit of the α spectra with α decays from ^{238}U, ^{232}Th and ^{235}U chains

Fit window: [1430, 3400] keV

Cone case



Cylinder case



Radioactive contamination

Chain	Nuclide	Activity (mBq/kg)	
		Cone	Cylinder
		10.63 g	**23.95 g**
^{232}Th	^{232}Th	0.10 ± 0.03	0.29 ± 0.08
	^{228}Th	0.16 ± 0.03	0.47 ± 0.03
^{238}U	^{238}U	0.43 ± 0.04	1.14 ± 0.06
	^{234}U	0.07 ± 0.11	3.73 ± 0.16
	^{230}Th	0.28 ± 0.02	0.00 ± 0.02
	^{226}Ra	0.10 ± 0.04	0.20 ± 0.05
	^{210}Pb	2.82 ± 0.20	7.78 ± 0.30
^{235}U	^{235}U	0.32 ± 0.05	2.75 ± 0.12
	^{231}Pa	20.00 ± 0.32	32.00 ± 0.35
	^{227}Ac	0.40 ± 0.02	0.91 ± 0.02

α/β light ratio:

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

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

Fit of the α spectra with α decays from ^{238}U, ^{232}Th and ^{235}U chains

Summary

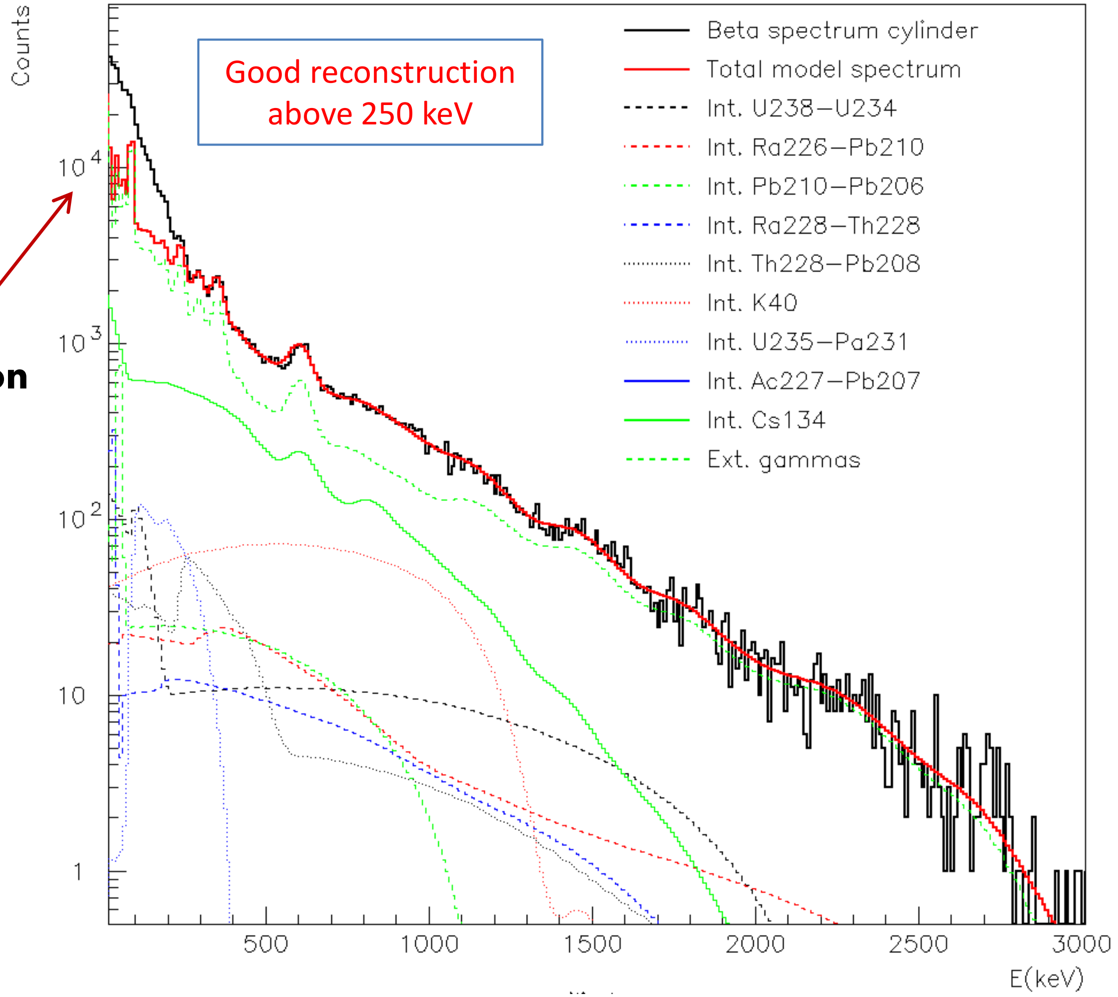
- Fits including also α events from ^{235}U chain reproduce well the measured spectra
- For the cone crystal seems that some events are missing in the higher E peak with respect to the model
- The peak with highest counting rate at ≈ 2 MeVee seems to be due to ^{231}Pa decay from ^{235}U chain for both the crystals
- **But**, the origin of the 3 α peaks at higher energy (i.e. E = 2.45, 2.85 and 3.25 MeVee), mostly due to ^{227}Ac→…→^{207}Pb sub-chain n. 3 from ^{235}U chain, is still under study

Fit of the $\beta(\gamma)$ spectrum of the CZC- cylinder crystal

Preliminary

- Radioactive contaminants from ^{238}U, ^{232}Th and ^{235}U chains have been simulated in the two CZC crystals and in the various materials of the setup (PMTs, Teflon, Copper).
- The PMTs contribution turns out to be dominant when taking into account the measured activities in the different materials.

Under investigation

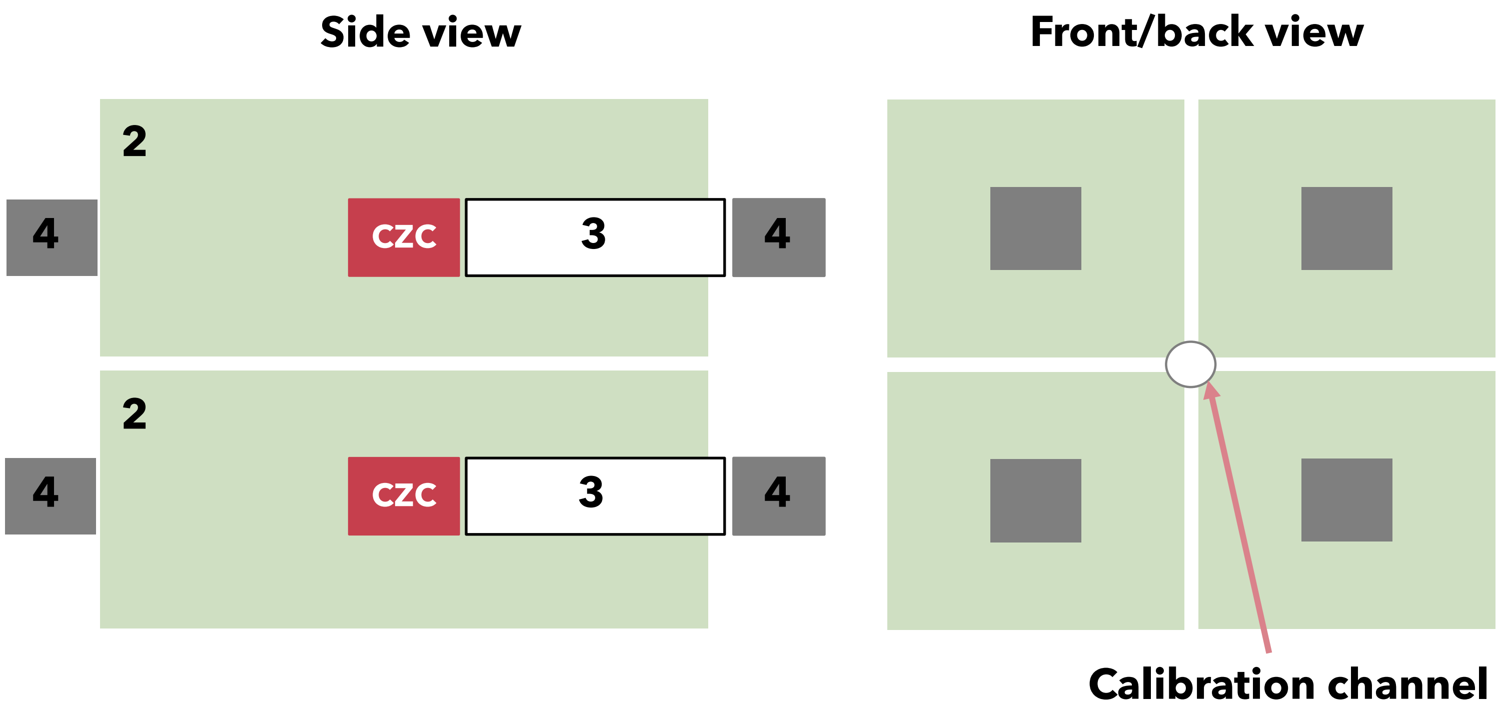


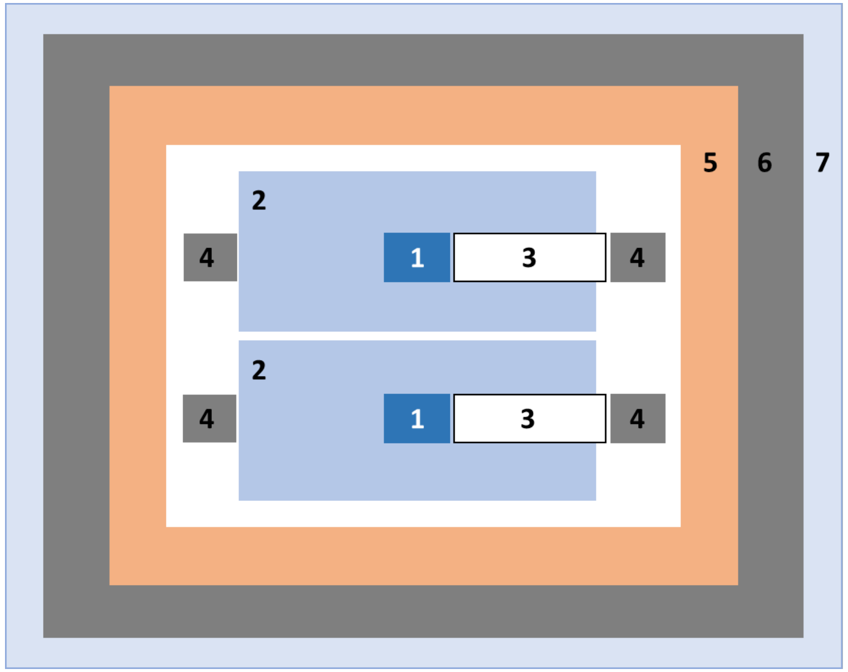
Fit window: [290, 2800] keV, χ^2=371, d.o.f.=231

Fit output:

Int. u238-1:	2.0000 mBq/kg
Int. u238-4:	1.2500 mBq/kg
Int. u238-5:	2.2313 mBq/kg
Int. t232-2:	1.2500 mBq/kg
Int. t232-3:	2.0000 mBq/kg
Int. k40:	9.9018 mBq/kg
Int. u235-1:	2.5207 mBq/kg
Int. u235-3:	0.0000 mBq/kg
Int. cs134:	52.7343 mBq/kg
u238-1-pmt:	142.5848 mBq/kg
u238-2-pmt:	539.7463 mBq/kg
u238-3-pmt:	1290.0000 mBq/kg
u238-4-pmt:	250.3394 mBq/kg
u238-5-pmt:	765.9943 mBq/kg
t232-1-pmt:	104.9851 mBq/kg
t232-2-pmt:	0.0000 mBq/kg
t232-3-pmt:	59.3276 mBq/kg
k40-pmt:	322.1013 mBq/kg

BREEZE detector array schematic (1st phase @ Queen's)





NEWS-G3 low-background setup

Four separate detector's modules, each consist of:

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

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

10^{21}-10^{22} y sensitivity level in one year of data taking

Perspectives and conclusions

No collaboration has ever involved a crystal-scintillator based experiment, except for the present one, that guarantees several well-known advantages as very high duty cycle, good energy resolution, high stability during the running condition, high detection efficiency, safety environmental impact, etc.

First two Cs_2ZrCl_6 scintillating crystals have been grown in Queen's University and studied at the National Laboratory of Gran Sasso (LNGS, Italy).

CZC have very good scintillating performance and radiopurity levels.

We are planning a new experiment with larger mass and better quality crystals.

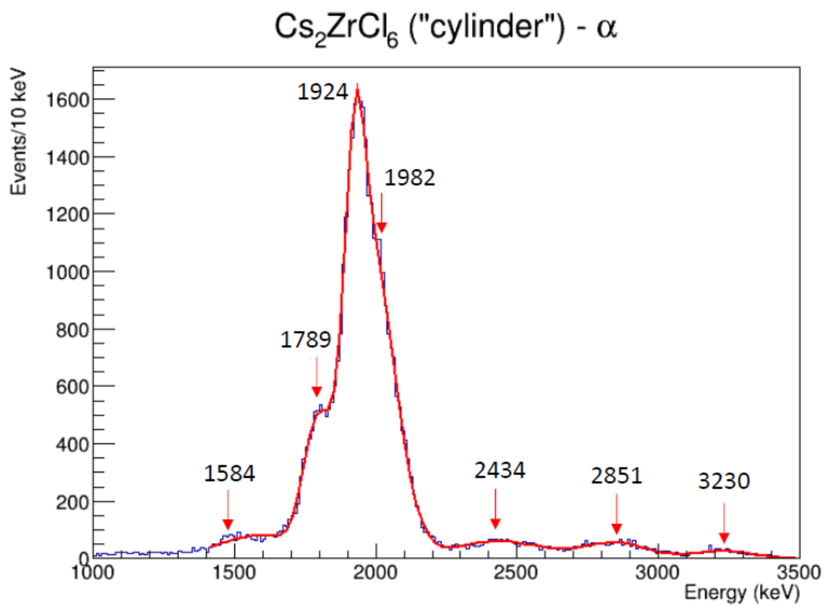
Backup slides

Contamination measured in R6233MOD PMTs

Time (s)	Mass (kg)	^{226}Ra (Bq/kg)	^{234m}Pa (Bq/kg)	^{235}U (mBq/kg)	^{228}Ra (Bq/kg)	^{228}Th (mBq/kg)	^{40}K (Bq/kg)	^{137}Cs (mBq/kg)	^{60}Co (mBq/kg)
233164	0.1599	0.46±0.02	1.3±0.7	48±18	0.13±0.02	91±16	0.61±0.12	< 12	< 10
252817	0.1429	0.42±0.02	< 1.6	47±18	0.097±0.023	75±15	0.45±0.10	< 27	< 7
179043	0.1493	0.42±0.03	< 2.2	< 61	0.11±0.03	83±17	0.53±0.13	15±9	< 13
253541	0.1431	0.49±0.02	2.6±0.9	69±20	0.12±0.03	100±20	0.65±0.12	< 10	< 14
171680	0.1513	0.45±0.03	< 2.9	35±21	0.12±0.03	72±17	0.66±0.15	< 24	< 5
147685	0.1461	0.38±0.03	< 3.2	< 51	0.12±0.03	62±18	0.45±0.13	< 17	< 6
173967	0.1547	0.54±0.03	2.1±0.9	45±19	0.14±0.03	120±20	0.91±0.16	< 20	< 6
86402	0.1550	0.39±0.04	< 2.4	57±25	0.15±0.04	78±23	0.38±0.17	< 18	< 15
86333	0.1597	0.34±0.03	< 2.3	< 59	0.12±0.04	64±19	0.57±0.18	< 33	< 25
252918	0.1548	0.43±0.02	< 2.1	37±17	0.12±0.02	100±20	0.46±0.10	< 17	< 12
190066	0.1458	0.42±0.03	< 1.7	47±20	0.16±0.03	66±14	0.49±0.12	< 21	< 14
167544	0.1462	0.51±0.03	< 1.8	59±23	0.12±0.03	100±20	0.73±0.16	< 14	< 8
165333	0.1480	0.39±0.03	< 2.9	38±19	0.13±0.03	100±20	0.29±0.11	< 13	< 7
257147	0.1474	0.36±0.02	< 2.0	< 44	0.097±0.023	73±13	0.52±0.11	15±7	< 10
160374	0.1531	0.48±0.03	1.7±0.9	42±22	0.11±0.03	67±18	0.32±0.12	< 17	< 8
163032	0.1442	0.35±0.03	< 2.8	36±19	0.14±0.03	83±19	0.60±0.15	< 17	< 6
Average		0.43	—	47	0.12	83	0.54	—	—
Standard deviation		0.06	—	10	0.02	17	0.16	—	—

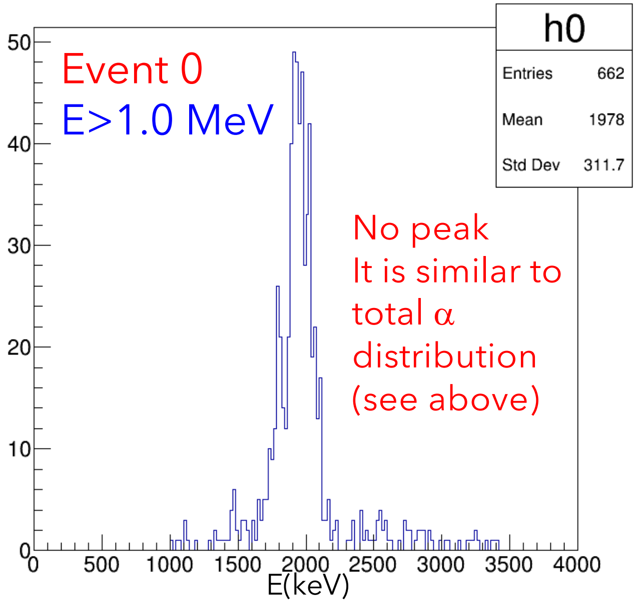
Analysis of time correlation between α events

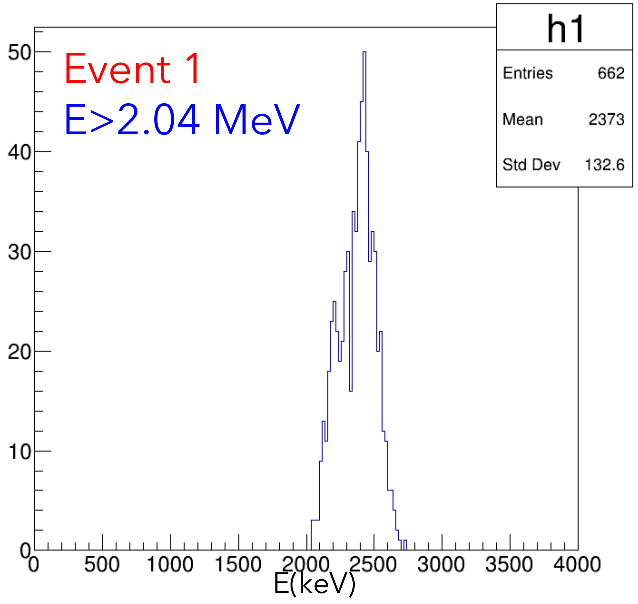
Preliminary

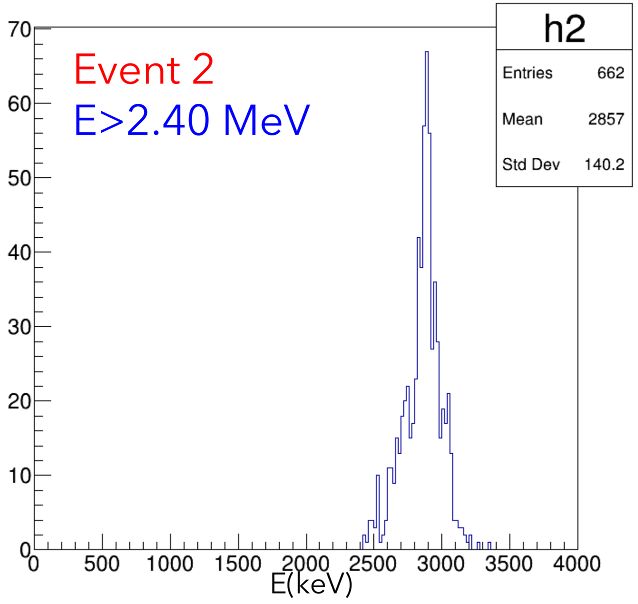


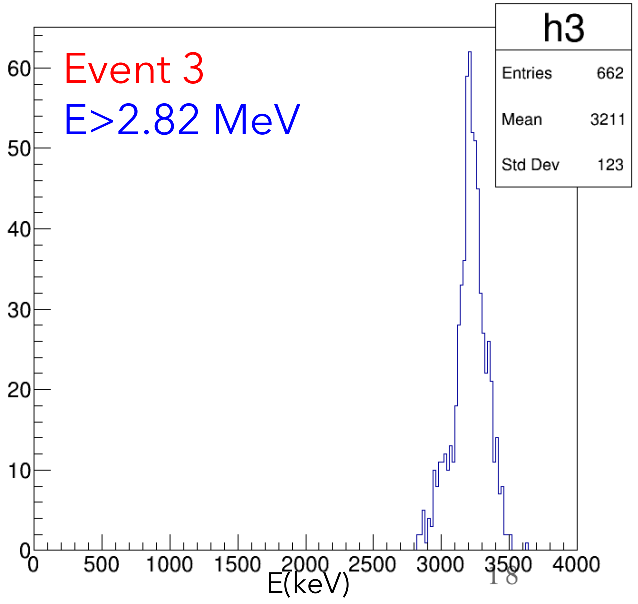
- Case of cylinder (higher statistics for α events)
- Starting point: the highest E peak at ≈ 3.2 MeVee (because in both ^{232}Th and ^{235}U chains it is the last peak of a fast sequence of α decays)
- Remind: if independent events the average delay is expected to be 720 s

1) Search for the α decay that precedes an α event in the E=3.2 MeVee peak;
2) Study of its E distribution and delay DT between events
3) (is E distribution a peak?).AND.(is average delay << 720 s?)
   If yes, continue the search for a further previous α decay → goto 1)





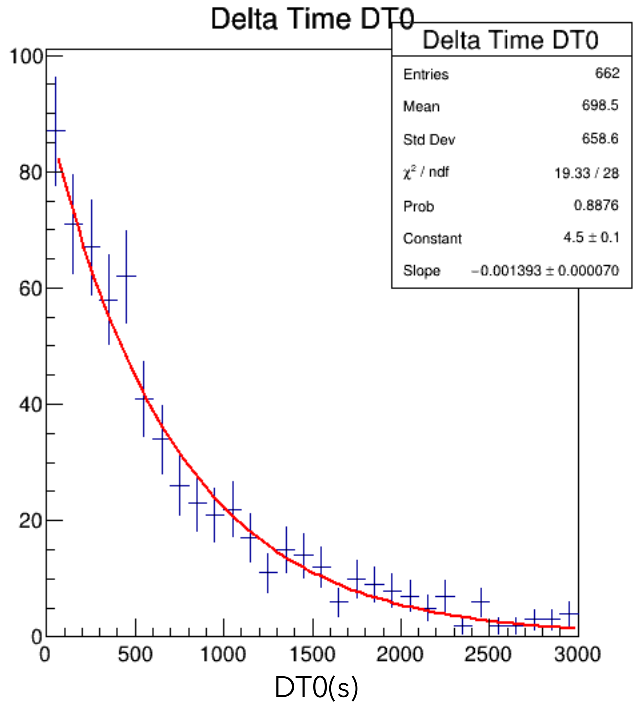


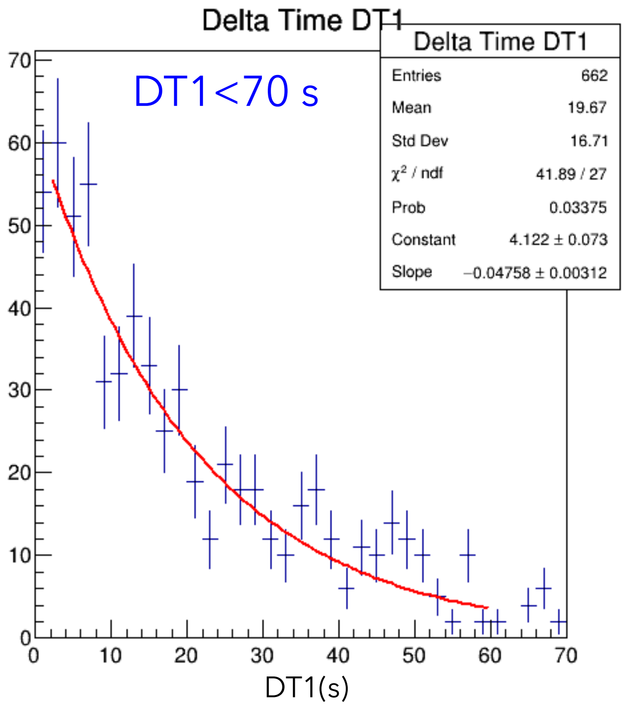


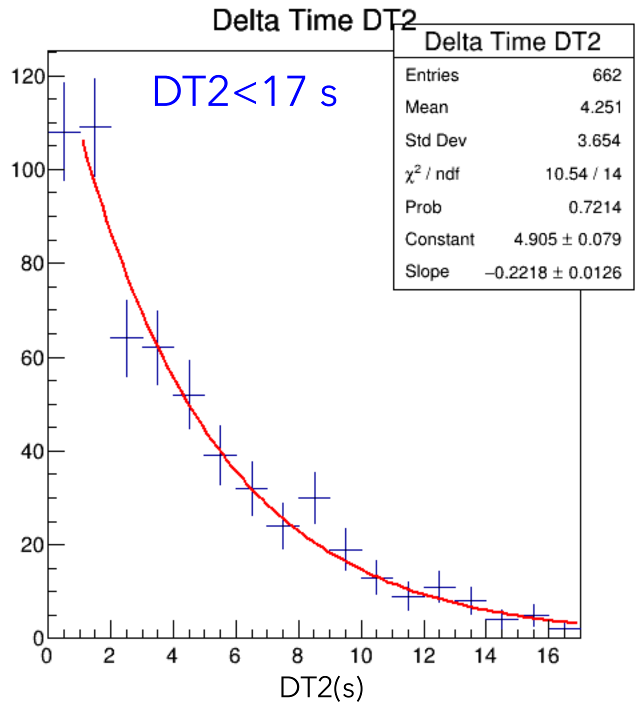
Analysis of time correlation between α events

Preliminary

Distributions of the delays between the 4 subsequent α events







Measured $\tau = (718\pm36)$ s, in perfect agreement with the expected delay for uncorrelated event (720 s)
$\Rightarrow$ Event 0 is uncorrelated

Measured $\tau = (21.0\pm1.4)$ s
$\Rightarrow$ Event 1 and 2 are correlated
$\Rightarrow$ $T_{1/2} = (14.6\pm1.0)$ s

Measured $\tau = (4.51\pm0.26)$ s,
$\Rightarrow$ Event 2 and 3 are correlated
$\Rightarrow$ $T_{1/2} = (3.13\pm0.18)$ s