



**TOR VERGATA**  
UNIVERSITÀ DEGLI STUDI DI ROMA

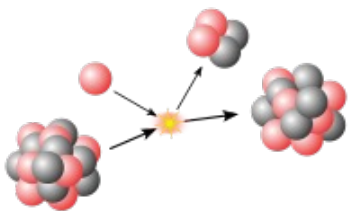


# **Radiopure crystal scintillators for rare-event searches: my PhD work recognized by the SIF “Ettore Pancini” Prize**



**Alice Leoncini**

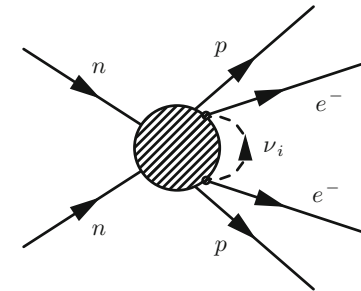




# The $2\beta$ decay

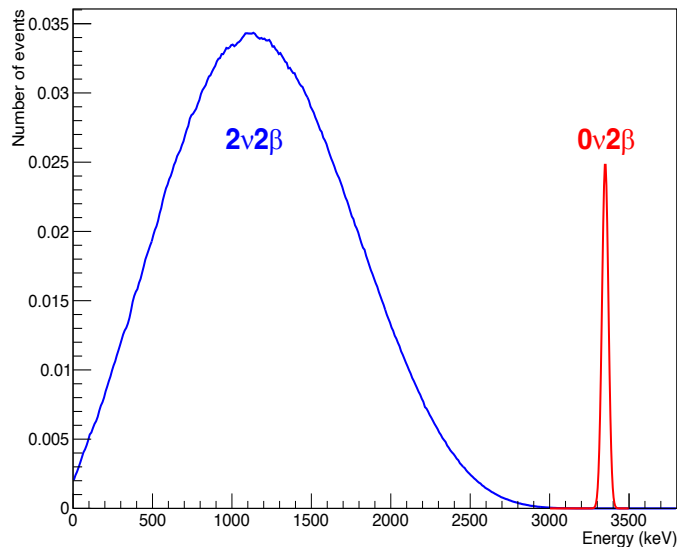
- $2\nu 2\beta$  decay is a rare process allowed in the SM → lepton number L conserved
- $0\nu 2\beta$ , if observed, could open a new window beyond the SM → L violated ( $\Delta L = 2$ ) → massive Majorana neutrino

(Neutrino oscillations)



$$\begin{aligned}
 2\nu 2\beta^- &: \quad {}^A_Z X \rightarrow {}^A_{Z+2} Y + 2e^- + 2\bar{\nu}_e \\
 2\nu 2\beta^+ &: \quad {}^A_Z X \rightarrow {}^A_{Z-2} Y + 2e^+ + 2\nu_e \\
 2\nu \varepsilon \beta^+ &: \quad e^- + {}^A_Z X \rightarrow {}^A_{Z-2} Y + e^+ + 2\nu_e \\
 2\nu 2\varepsilon &: \quad 2e^- + {}^A_Z X \rightarrow {}^A_{Z-2} Y + 2\nu_e
 \end{aligned}$$

$$\begin{aligned}
 0\nu 2\beta^- &: \quad {}^A_Z X \rightarrow {}^A_{Z+2} Y + 2e^- \\
 0\nu 2\beta^+ &: \quad {}^A_Z X \rightarrow {}^A_{Z-2} Y + 2e^+ \\
 0\nu \varepsilon \beta^+ &: \quad e^- + {}^A_Z X \rightarrow {}^A_{Z-2} Y + e^+ \\
 0\nu 2\varepsilon &: \quad 2e^- + {}^A_Z X \rightarrow {}^A_{Z-2} Y + \gamma
 \end{aligned}$$



Current sensitivity for  $2\nu 2\beta^-$  decay:  $T_{1/2} \sim 10^{18}$ - $10^{24}$  yr; for  $0\nu 2\beta^-$ :  $T_{1/2} \sim 10^{24}$ - $10^{26}$  yr

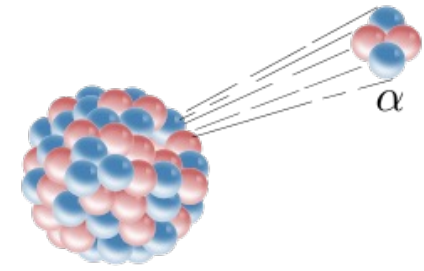
Positive channels: less studied but easier to identify. Current experimental sensitivities:  $T_{1/2} \sim 10^{16}$ - $10^{21}$  yr.

Complementary information to  $0\nu 2\beta^-$  + resonant effect

$$(T_{1/2}^{0\nu})^{-1} = g_A^4 G^{0\nu}(Q, Z) \left| M_{GT}^{0\nu} - \frac{g_V^2}{g_A^2} M_F^{0\nu} \right|^2 |m_{\beta\beta}|^2$$

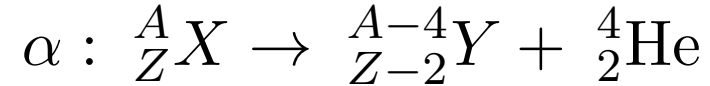
where  $m_{\beta\beta} = \sum_i U_{ei}^2 m_i$

Half-life of  $0\nu 2\beta \rightarrow$  measurement of  $m_{\beta\beta}$

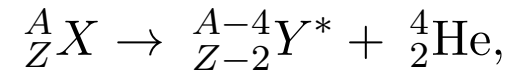


# The rare $\alpha$ decay

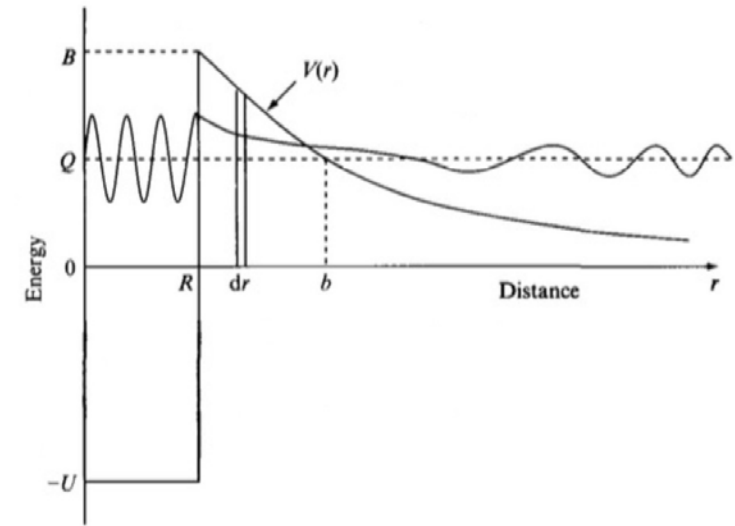
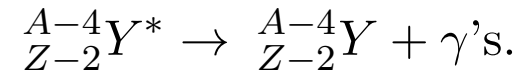
- $\alpha$  decay corresponds to a very asymmetric spontaneous fission:



- less-energetic  $\alpha$  decays are always accompanied by the emission of  $\gamma$  quanta:



with



$T_{1/2}$  ranges between  $10^{-8}$  s of  $^{217}\text{Ac}$  to  $10^{19}$  yr of  $^{209}\text{Bi}$

**Rare  $\alpha$  decay:**  $T_{1/2} > 10^{14}$  yr  
(e.g.  $^{184}\text{Os}$ ,  $^{180}\text{W}$ ,  $^{174}\text{Hf}$ ,  $^{152}\text{Gd}$ ,  
 $^{147}\text{Sm}$ ,  $^{143}\text{Nd}$ )



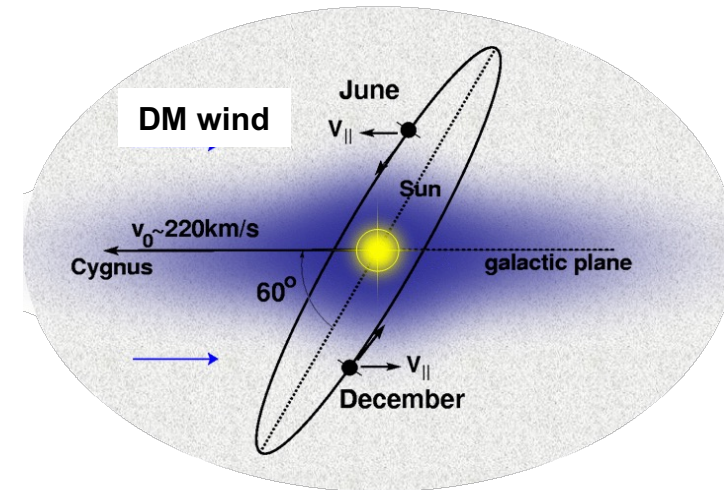
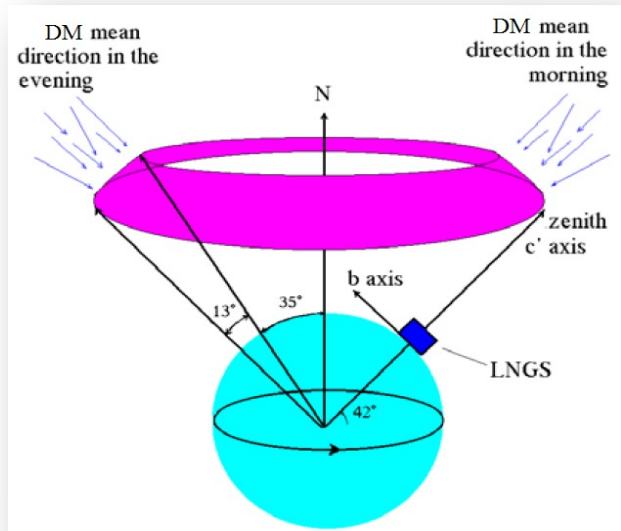
Extremely difficult to  
detect with conventional  
techniques

- **Crucial role in nuclear physics:** details on the nuclear structure, its levels and properties of nuclei.
- Essential also for **nuclear and particle astrophysics studies** ( $\alpha$ -capture reactions,  $\beta$ -delayed fission, nucleosynthesis).

# The directionality approach to study *Dark Matter* candidate particles

Based on the study of the correlation between the arrival direction of DM candidates able to induce a nuclear recoil and the Earth motion in the galactic frame.

Impinging direction of DM particle is (preferentially) opposite to the velocity of the Sun in the Galaxy...



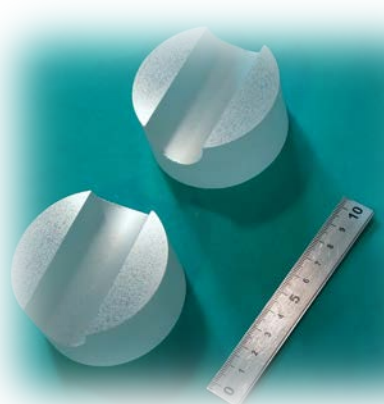
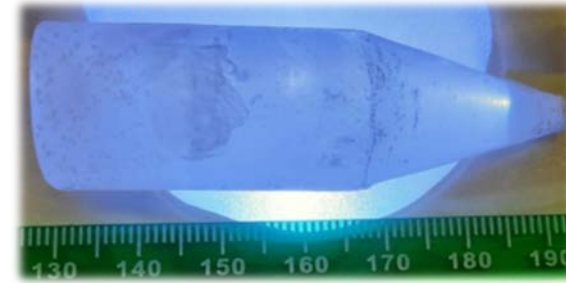
...and due to the Earth's rotation around its axis, the DM particles average direction with respect to an observer on the Earth changes with a period of a sidereal day

The direction of the induced nuclear recoil is expected to be strongly correlated with the direction of the impinging DM particle.

→ A direction-sensitive detector is needed

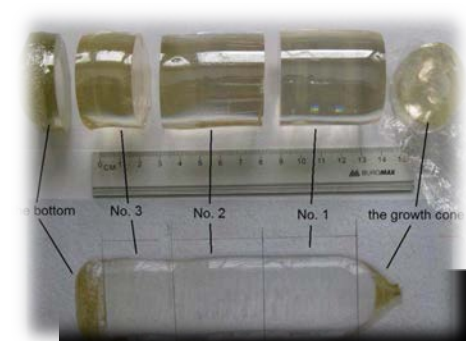


# Inorganic crystal scintillators for the search of rare processes



# DAMA set-ups

an observatory for rare processes @ LNGS



**DAMA/NaI**  
↓  
**DAMA/LIBRA-phase1**  
↓  
**DAMA/LIBRA-phase2**  
+ empowered

DAMA/CRYST

DAMA/R&D

low bckg DAMA/Ge  
for sampling meas.  
& activities in Stella  
facility

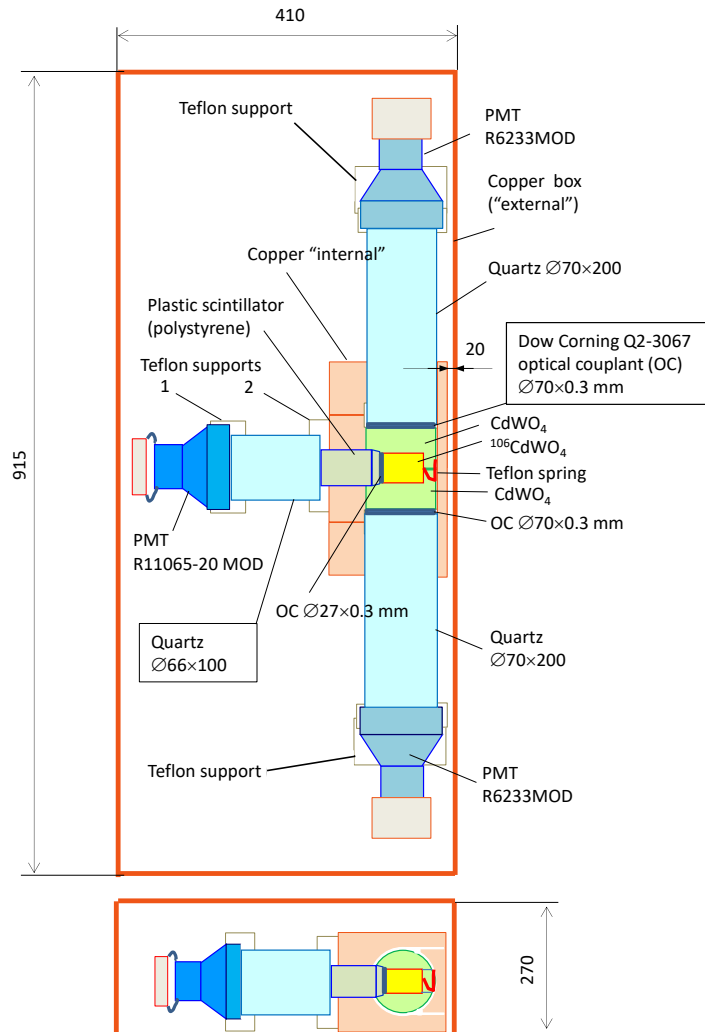


- Roma Tor Vergata, Roma La Sapienza, LNGS, IHEP/Beijing
- + by-products and small scale expts.: INR-Kiev, Queen's University + other institutions
  - + neutron meas.: ENEA-Frascati, ENEA-Casaccia
  - + in some studies on  $2\beta$  decays (DST-MAE and Inter-Universities project): IIT Kharagpur and Ropar, India

web site: <https://dama.web.roma2.infn.it/>

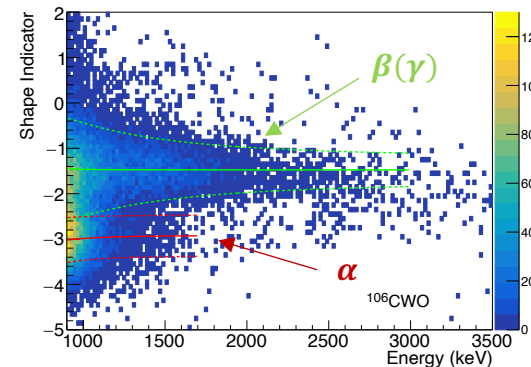
# Search for $2\beta^+$ decays in enriched $^{106}\text{CdWO}_4$ crystal scintillator

Detector system with the  $^{106}\text{CdWO}_4$  scintillator at the R&D set-up

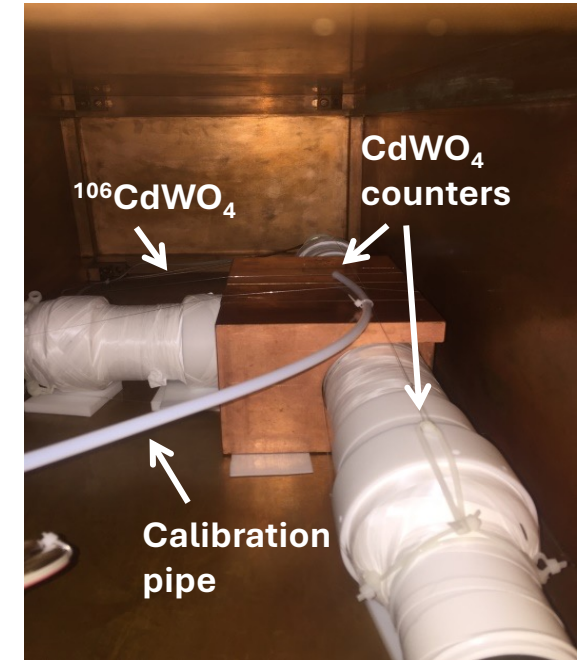


- **DAMA/R&D:** high-purity copper 10 cm thick, 15 cm of low-radioactive lead, a 1.5 mm layer of cadmium and from 4 to 10 cm of polyethylene/ paraffin.
- $^{106}\text{CdWO}_4$  is housed in a cylindrical cut-out of the two  $\text{CdWO}_4$  ( $\varnothing 70$  mm x 38 mm) scintillators which almost completely envelop the enriched crystal.
- An event-by-event DAQ records pulses in case of:
  - an event with  $E \gtrsim 500$  keV in  $^{106}\text{CdWO}_4$  detector;
  - $^{106}\text{CdWO}_4$  detector in coincidence with at least one of the  $\text{CdWO}_4$  counters.

PSD



DAMA/R&D setup at LNGS





# $\beta(\gamma)$ spectra in different modes

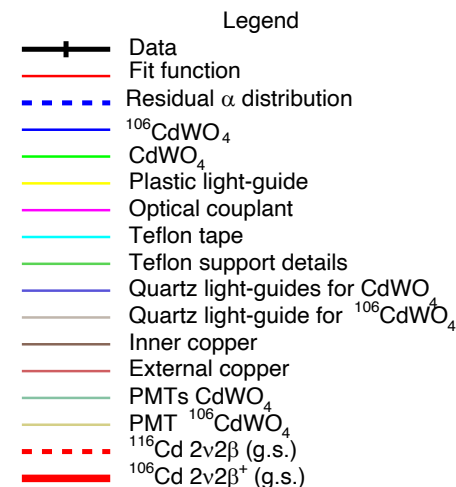
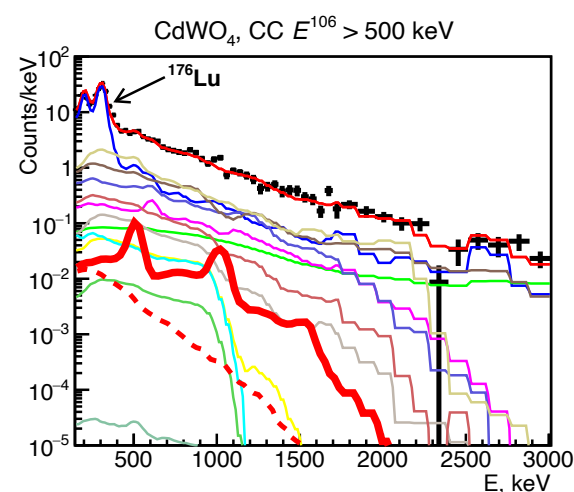
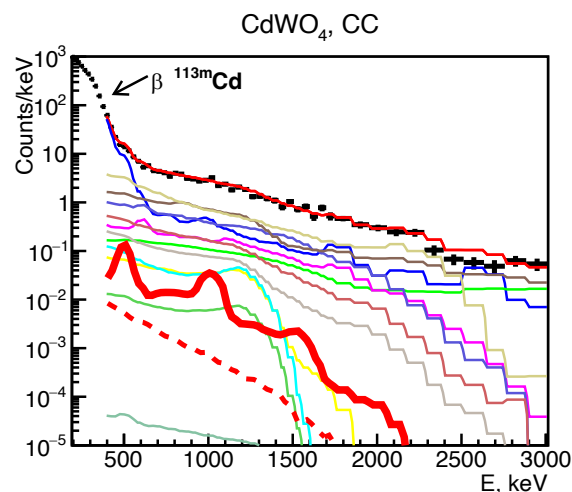
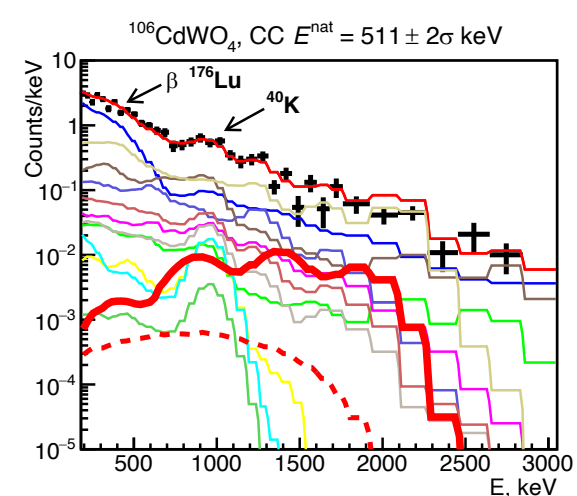
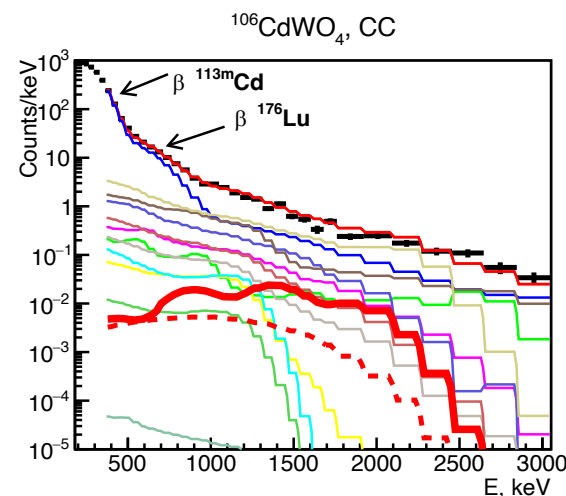
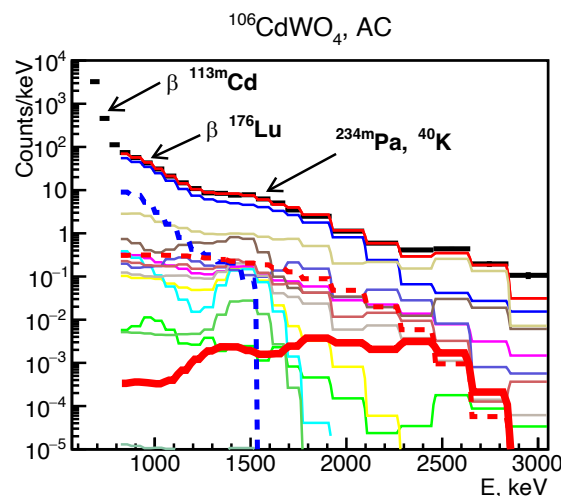
✓ 1075 days of data taking

## ❖ Anticoincidence mode (AC):

An event in the  $^{106}\text{CdWO}_4$  detector with an energy  $> 500$  keV.

## ❖ Coincidence mode (CC):

An event in the  $^{106}\text{CdWO}_4$  detector with an energy  $> 50$  keV in coincidence with at least one of the  $\text{CdWO}_4$  counters with  $E > 50$  keV.



- The main background contributions arise from the PMT, the light guide and the internal contamination of the  $^{106}\text{CdWO}_4$  crystal.

# Results of half-life limits on $2\beta$ decay processes in $^{106}\text{Cd}$

Decay	Level of $^{106}\text{Pd}$ , keV	Theoretical $T_{1/2}$ , Years	lim $T_{1/2}$ , Years	
			Previous Result	Present Work
$2\nu 2\beta^+$	g.s.	$(5.4 - 880) \times 10^{25}$ [47,48,50], $>2.4 \times 10^{27}$ [49]	$4.4 \times 10^{21}$ [54]	$1.7 \times 10^{22}$
	512	$(1.5 - 25) \times 10^{27}$ [47,55,56]	$4.1 \times 10^{21}$ [54]	$1.5 \times 10^{22}$
$0\nu 2\beta^+$	g.s.	$(1.4 - 32) \times 10^{27}$ [47,55–61]	$5.9 \times 10^{21}$ [62]	$2.2 \times 10^{22}$
	512		$4.1 \times 10^{21}$ [54]	$1.5 \times 10^{22}$
<u><math>2\nu\text{EC}\beta^+</math></u>	g.s.	$(1.4 - 240) \times 10^{21}$ [47,48,50–53], $>2.7 \times 10^{22}$ [49]	$2.1 \times 10^{21}$ [62]	$7.7 \times 10^{21}$
	512	$(5.3 - 24) \times 10^{25}$ [51,52], $>1.1 \times 10^{25}$ [49]	$3.3 \times 10^{21}$ [54]	$9.9 \times 10^{21}$
	1128	$3.7 \times 10^{30}$ [51]	$2.0 \times 10^{21}$ [54]	$1.2 \times 10^{22}$
	1134	$(1.3 - 13) \times 10^{26}$ [51,52], $>1.1 \times 10^{27}$ [49]	$2.5 \times 10^{21}$ [54]	$1.3 \times 10^{22}$
$0\nu\text{EC}\beta^+$	g.s.	$(1.0 - 17) \times 10^{26}$ [32,47,55,56]	$1.4 \times 10^{22}$ [62]	$1.5 \times 10^{22}$
	512		$9.7 \times 10^{21}$ [62]	$2.1 \times 10^{22}$
	1128		$1.0 \times 10^{22}$ [62]	$1.9 \times 10^{22}$
	1134	$(1.0 - 21) \times 10^{29}$ [32,55,57,58]	$2.7 \times 10^{21}$ [54]	$2.1 \times 10^{22}$

+ other channels

[32] Suhonen, J. Phys. Lett. B 2011, 701, 490–495.  
[47] Stoica, S.; Klapdor-Kleingrothaus, H.V. Eur. Phys. J. A 2003, 17, 529–536.  
[48] Shukla, A. et al. Eur. Phys. J. A 2005, 23, 235–242.  
[49] Domin, P. et al. Nucl. Phys. A 2005, 753, 337–363.  
[50] Raina, P.K. et al. Eur. Phys. J. A 2006, 28, 27–36.  
[51] Suhonen, J. AIP Conf. Proc. 2011, 1417, 115–119.  
[52] Pirinen, P.; Suhonen, J. Phys. Rev. C 2015, 91, 054309.

[53] Ejiri, H. J. Phys. G 2017, 44, 115201.  
[54] Leoncini, A. et al. Phys. Scr. 2022, 97, 064006.  
[55] Suhonen, J.; Aunola, M. Nucl. Phys. A 2003, 723, 271–288.  
[56] Rath, P.K. et al. Phys. Rev. C 2009, 80, 044303.  
[57] Suhonen, J. J. Phys. Conf. Ser. 2012, 338, 012030.  
[58] Suhonen, J. Phys. Scripta T 2012, 150, 014039.  
[62] Belli, P. et al. Universe 2020, 6, 182.

# Results of half-life limits on $2\beta$ decay processes in $^{106}\text{Cd}$

Decay	Level of $^{106}\text{Pd}$	Theoretical $T_{1/2}$ , Years	lim $T_{1/2}$ , Years	
			Previous Result	Present Work
$2\nu 2\beta$			$4 \times 10^{21}$ [54]	$1.7 \times 10^{22}$
			$1 \times 10^{21}$ [54]	$1.5 \times 10^{22}$
$0\nu 2\beta$			$9 \times 10^{21}$ [62]	$2.2 \times 10^{22}$
			$1 \times 10^{21}$ [54]	$1.5 \times 10^{22}$
$2\nu\text{E}\beta^+$			$1 \times 10^{21}$ [54]	$7.7 \times 10^{21}$
			$3 \times 10^{21}$ [54]	$9.9 \times 10^{21}$
			$0 \times 10^{21}$ [54]	$1.2 \times 10^{22}$
	1134	$(1.3 - 13) \times 10^{20}$ [51,52], $>1.1 \times 10^{22}$ [49]	$2.5 \times 10^{21}$ [54]	$1.3 \times 10^{22}$
$0\nu\text{EC}\beta^+$	g.s.	$(1.0 - 17) \times 10^{26}$ [32,47,55,56]	$1.4 \times 10^{22}$ [62]	$1.5 \times 10^{22}$
	512		$9.7 \times 10^{21}$ [62]	$2.1 \times 10^{22}$
	1128		$1.0 \times 10^{22}$ [62]	$1.9 \times 10^{22}$
	1134	$(1.0 - 21) \times 10^{29}$ [32,55,57,58]	$2.7 \times 10^{21}$ [54]	$2.1 \times 10^{22}$

The sensitivity obtained on the  $T_{1/2}$  for the case  $2\nu\text{E}\beta^+$  is within the theoretical predictions:  
 $T_{1/2} \sim 10^{21} - 10^{22}$  yr.

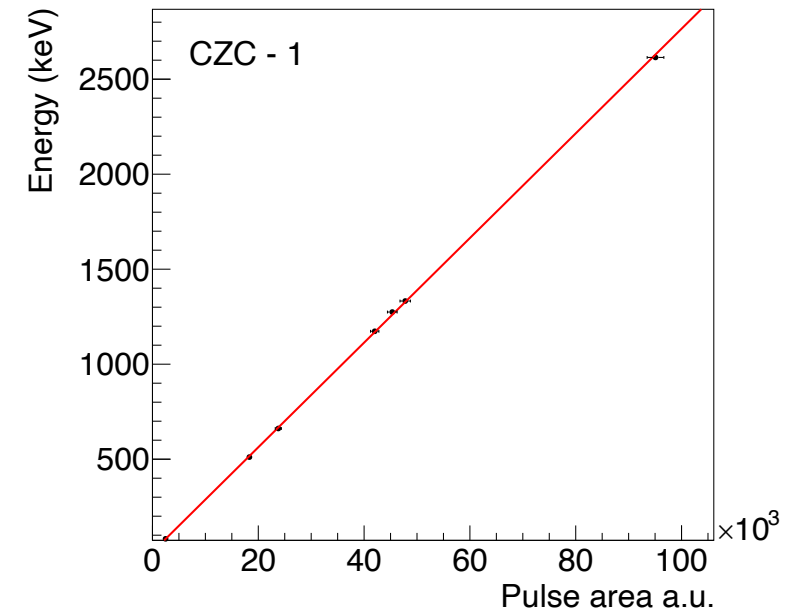
+ other channels

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[57] Suhonen, J. J. Phys. Conf. Ser. 2012, 338, 012030.  
[58] Suhonen, J. Phys. Scripta T 2012, 150, 014039.  
[62] Belli, P. et al. Universe 2020, 6, 182.

Some general properties	Cs <sub>2</sub> HfCl <sub>6</sub>	Cs <sub>2</sub> ZrCl <sub>6</sub>
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
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<sub>2</sub>HfCl<sub>6</sub> (**CHC**) and Cs<sub>2</sub>ZrCl<sub>6</sub> (**CZC**) crystal scintillators for the search of rare $\alpha$ and $2\beta$ decays



\* 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



# New measurement of the $\alpha$ decay of $^{174}\text{Hf}$ to the g.s. of $^{174}\text{Yb}$ using a **CHC** crystal scintillator

Mass = 16.87 g  
 $\varnothing 21.20(5) \times 12.8(1)$   
mm



**DAMA/CRYS  
setup at LNGS**

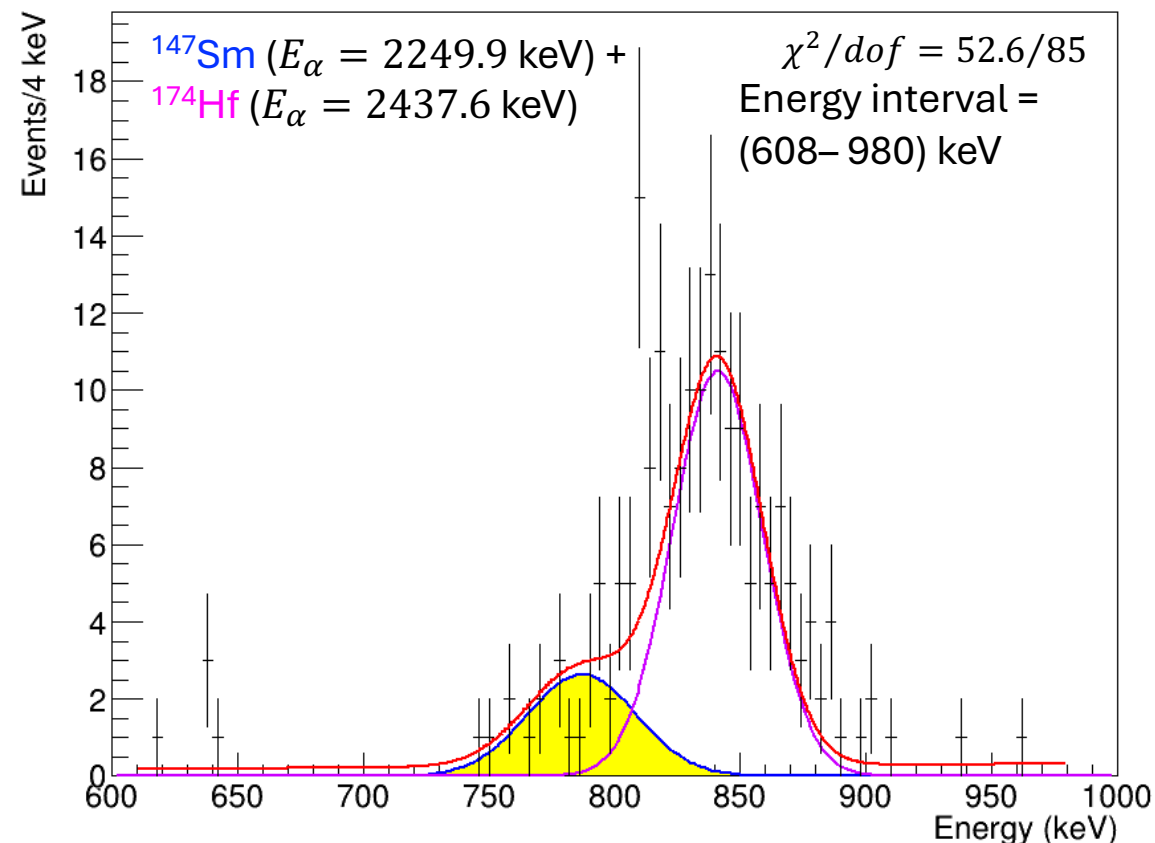
ICP-MS measurements

Element	Concentration (ppb)
U	0.73(22)
Th	0.16(5)
Pb	440(130)
Sm	2(1)
K	1900(570)

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

$T_{1/2} = 3.8_{-0.9}^{+1.7} \times 10^{16}$  yr of  $\alpha$  decay of  $^{174}\text{Hf}$   
to the g.s. of  $^{174}\text{Yb}$

➤ Data analysis on 4 CHC crystals ( $\varnothing 26 \times 20$  mm<sup>3</sup>)  
encapsulated in silicone-based sealant, **is ongoing** to  
improve sensitivity on  $\alpha$  decay of  $^{174}\text{Hf}$ .



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

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

# New measurement of the $\alpha$ decay of $^{174}\text{Hf}$ to the g.s. of $^{174}\text{Yb}$

Mass = 16.87 g  
 $\varnothing 21.20(5) \times 12.8(1)$   
 mm

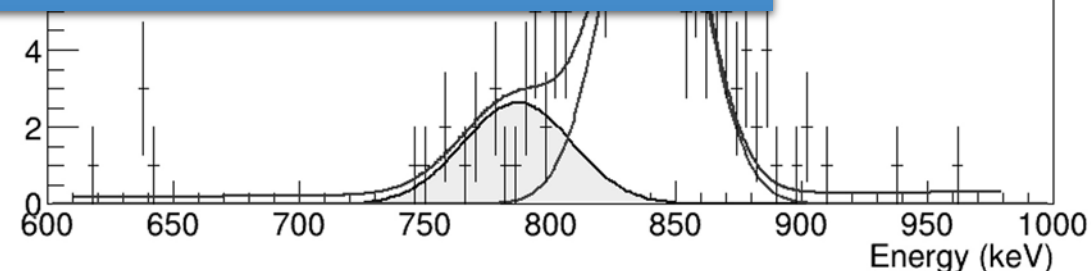
DAMA/CRY  
 setup at LNGS

Activity of  $^{147}\text{Sm}$   
 corresponding to a concentration of  $(2.0 \pm 0.3)$  ppb  
 of  $^{147}\text{Sm}$ , in agreement with ICP-MS measurements.

$T_{1/2} = 3.8_{-0.9}^{+1.7} \times 10^{16}$  yr of  $\alpha$  decay of  $^{174}\text{Hf}$   
 to the g.s. of  $^{174}\text{Yb}$

➤ Data analysis on 4 CHC crystals ( $\varnothing 26 \times 20$  mm<sup>3</sup>)  
 encapsulated in silicone-based sealant, is ongoing to  
 improve sensitivity on  $\alpha$  decay of  $^{174}\text{Hf}$ .

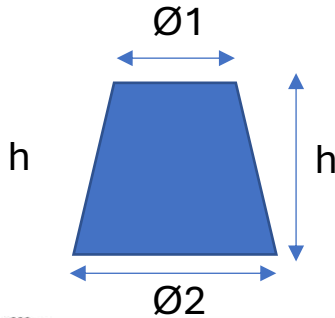
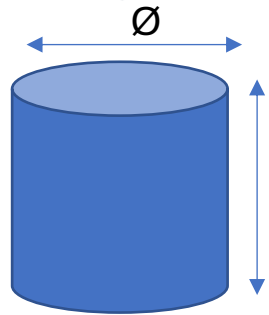
The sensitivity obtained on the  
 $T_{1/2}$  for the  $\alpha$  decay of  $^{174}\text{Hf}$  is  
 within the theoretical  
 predictions:  
 $T_{1/2} \sim (3.5 - 7.4) \times 10^{16}$  yr.



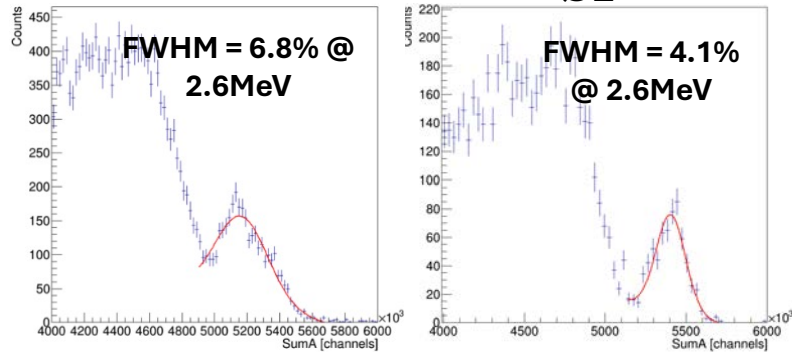
Q.F. =  $0.350 \pm 0.008$  for  $^{147}\text{Sm}$   
 Q.F. =  $0.345 \pm 0.001$  for  $^{174}\text{Hf}$

# First low-background measurements of **CZC** at LNGS

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



**M=10,6(1) g**  
 $h=17,90(5)$  mm  
 $\varnothing1=8,0(1)$  mm  
 $\varnothing2=19,70(5)$  mm

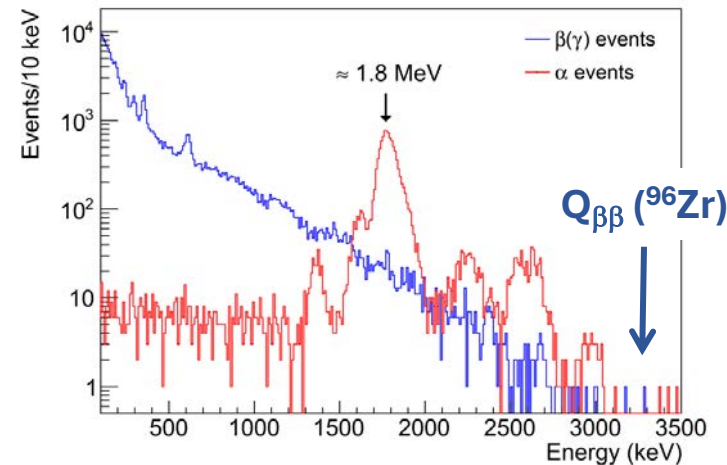
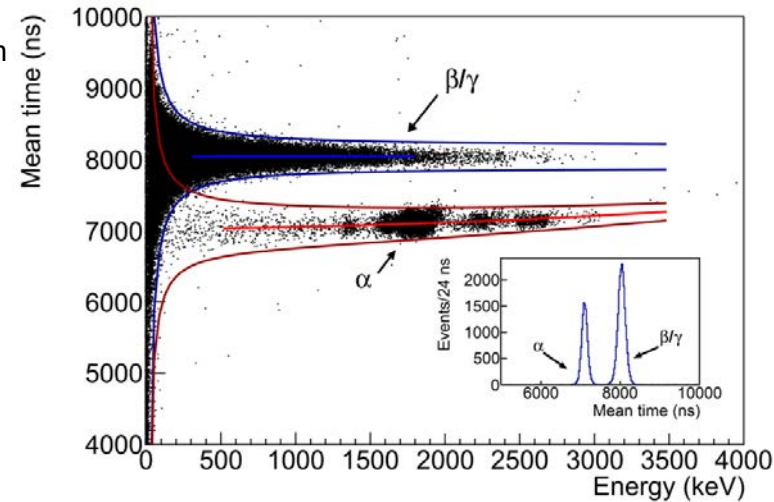


## DAMA/CRYS setup (2021-2022)



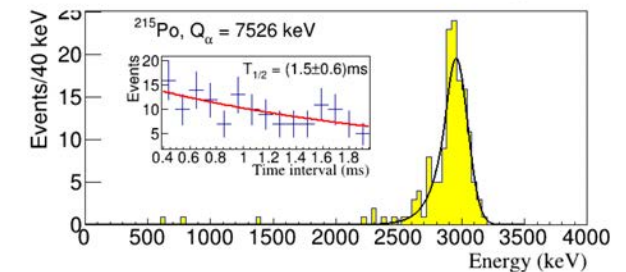
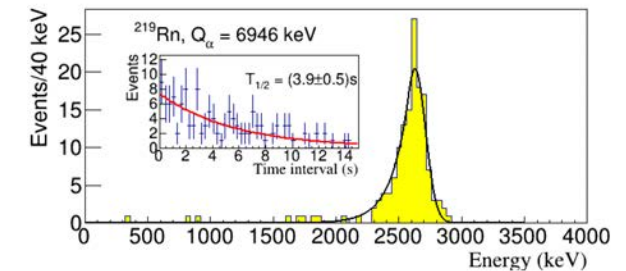
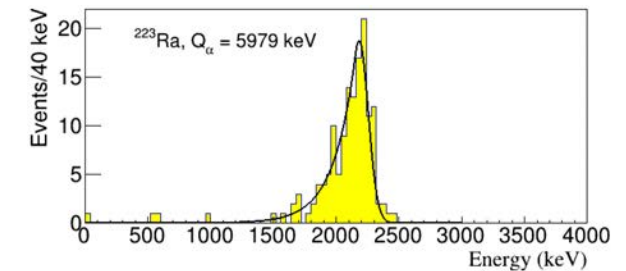
- OFHC Cu (15 cm)
- Low-activity Pb (20 cm)
- HDPE (5 cm)

Selection efficiency is  
**99.7% in [0.2–3.5] MeV**



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

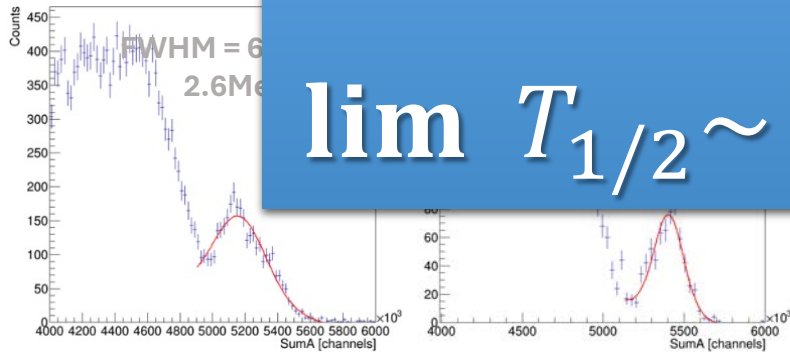
## Time-amplitude analysis



## First low-background measurements of C7C at LNGS

Experimental limits on the  $T_{1/2}$  for various decay modes in  $^{94,96}\text{Zr}$  at the level of:  
 $\lim T_{1/2} \sim 10^{17} - 10^{20} \text{ yr (90\% C.L.)}$

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



DAMA/CRY

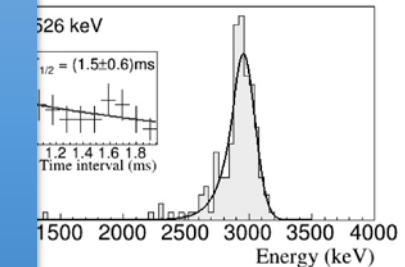
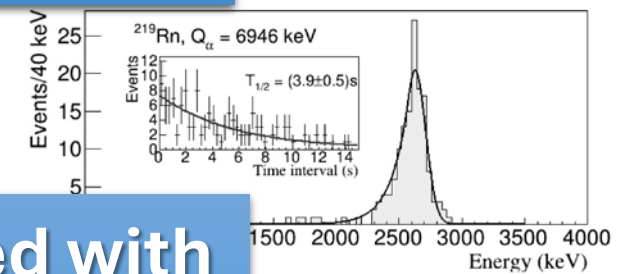
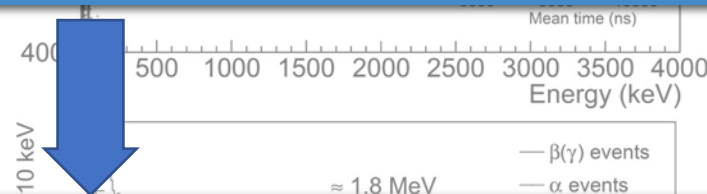
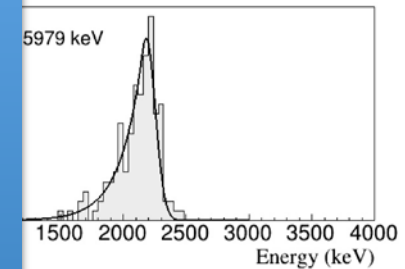
Limits on the  $T_{1/2}$  for  $^{94}\text{Zr}$  to the g.s. improved with respect to literature values:

$$T_{1/2}^{0\nu} > 2.6 \times 10^{19} \text{ yr (90\% C.L.)}$$

$$T_{1/2}^{2\nu} > 2.4 \times 10^{18} \text{ yr (90\% C.L.)}$$

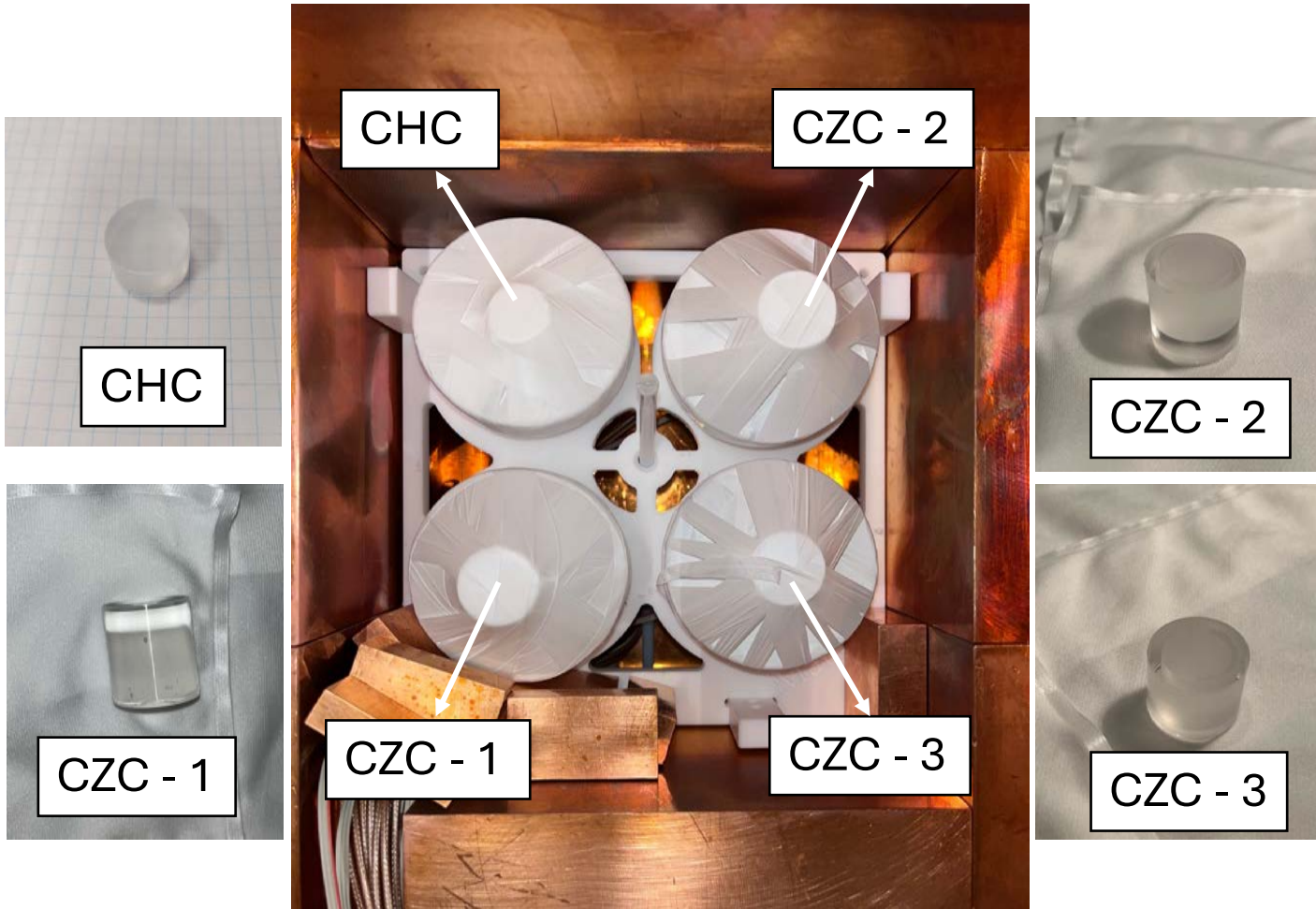
is 0.09 counts/(kg·keV·yr)

Amplitude analysis



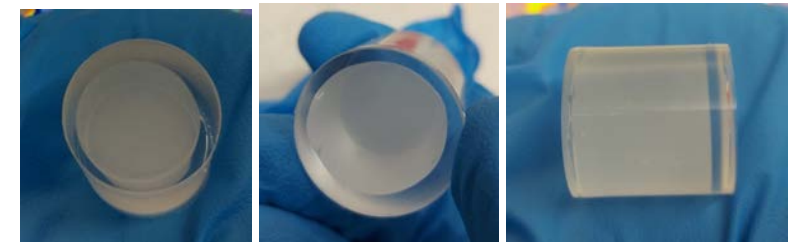


# New low-background measurements in DAMA/CRYSSetup (LNGS)



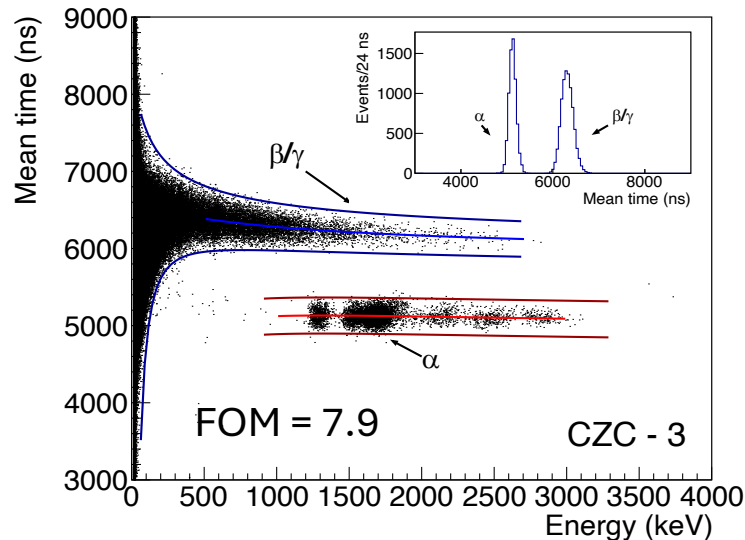
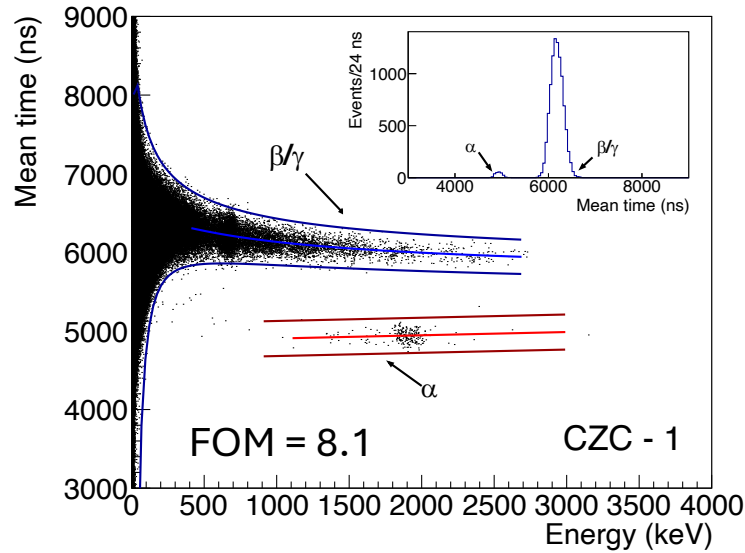
DAMA/CRYSSetup at LNGS

- ✓ Three new  $\text{Cs}_2\text{ZrCl}_6$  crystals + one  $\text{Cs}_2\text{HfCl}_6$
- ✓ 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-based resin + quartz window**
- ✓ Modified experimental setup
- ✓ Measurements started on June 30th, 2023, for a total of 97.7 days live time



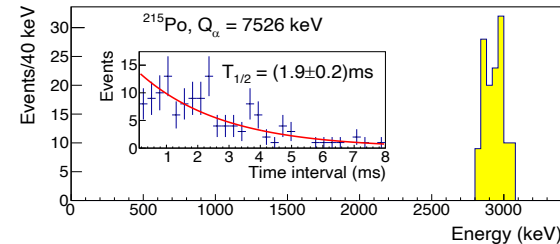
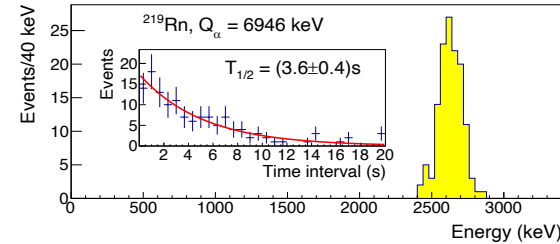
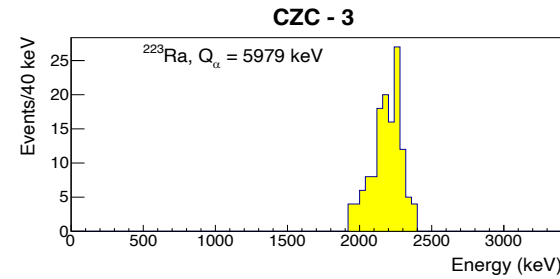
# Data analysis of the $\text{Cs}_2\text{ZrCl}_6$ crystals

Selection efficiency is  
99.7% in [0.5–3.5] MeV

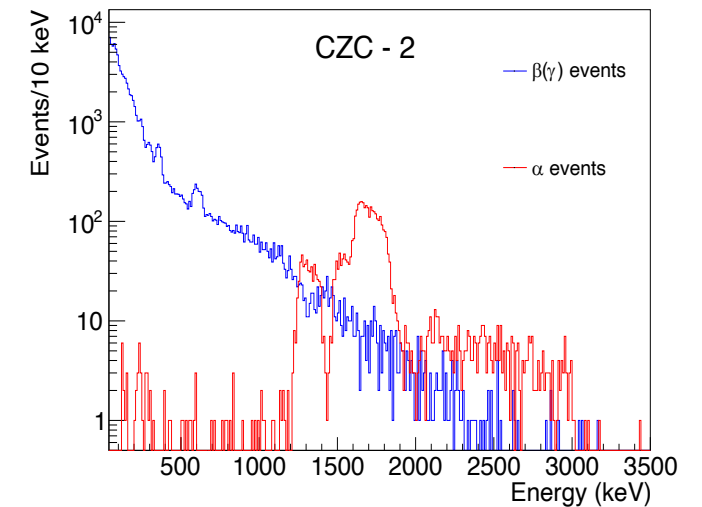
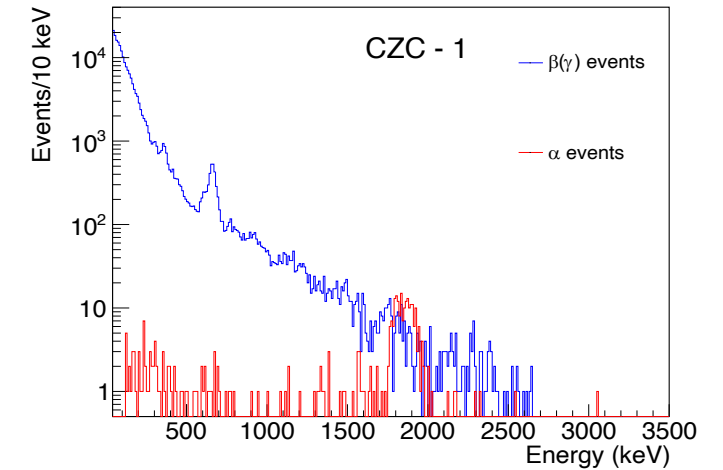


Time-amplitude analysis

$A(^{227}\text{Ac})$  < 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



Measured energy spectra over  
97.7 days of data taking



+ Background model considering  $^{238}\text{U}$ ,  $^{235}\text{U}$  and  $^{232}\text{Th}$  chains with their daughters.

# Study of *Dark Matter* with directionality approach using $\text{ZnWO}_4$ crystal scintillators

## Advantages in the use of $\text{ZnWO}_4$ crystal scintillators

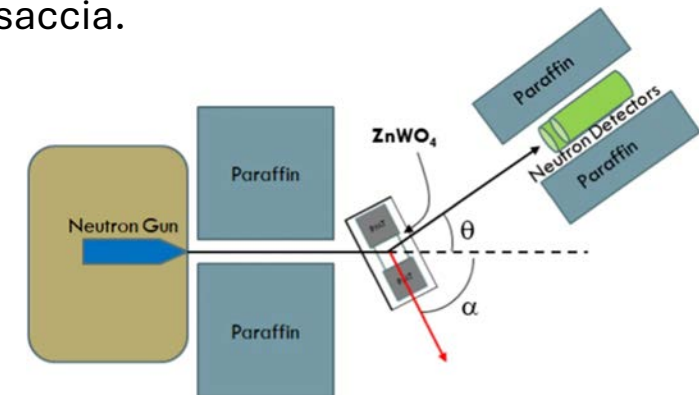
- Very good anisotropic features: for heavy particles ( $p$ ,  $\alpha$ , nuclear recoils), the light output and the pulse shape depends on the particle impinging direction with respect to the crystal axes.
- High level of radio-purity.
- High light output, that is low energy threshold feasible.
- High stability in the running conditions.
- Sensitivity to small and large mass DM candidate particles.
- Detectors with  $\sim \text{kg}$  masses.



## The experiment with $\text{ZnWO}_4$ crystal scintillator within ADAMO

In the framework of the ADAMO project, recent measurements [Eur. Phys. J. A 56 (2020) 83] were performed to verify the anisotropic response of a  $\text{ZnWO}_4$  crystal scintillator to:

1.  **$\alpha$  particles** : a small  $\text{ZnWO}_4$  crystal ( $10 \times 10 \times 10 \text{ mm}^3$ , with mass of 7.99 g), irradiated by a collimated beam of  $\alpha$  particles from an  $^{241}\text{Am}$  source in the directions along the crystal axes I, II and III.
2. **Oxygen nuclear recoils**: neutron beam of 14 MeV produced by a neutron generator at ENEA-Casaccia.





# Study of *Dark Matter* with directionality approach using $\text{ZnWO}_4$ crystal

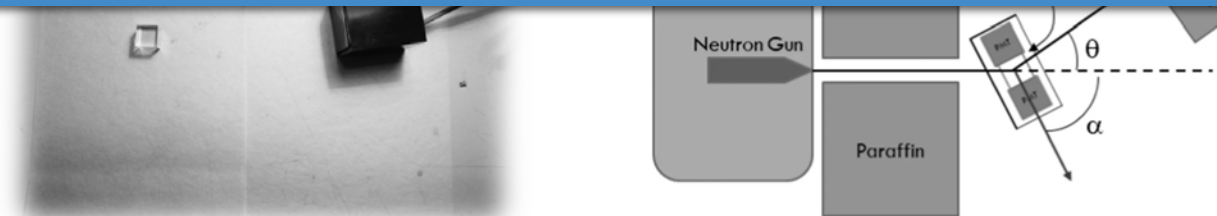
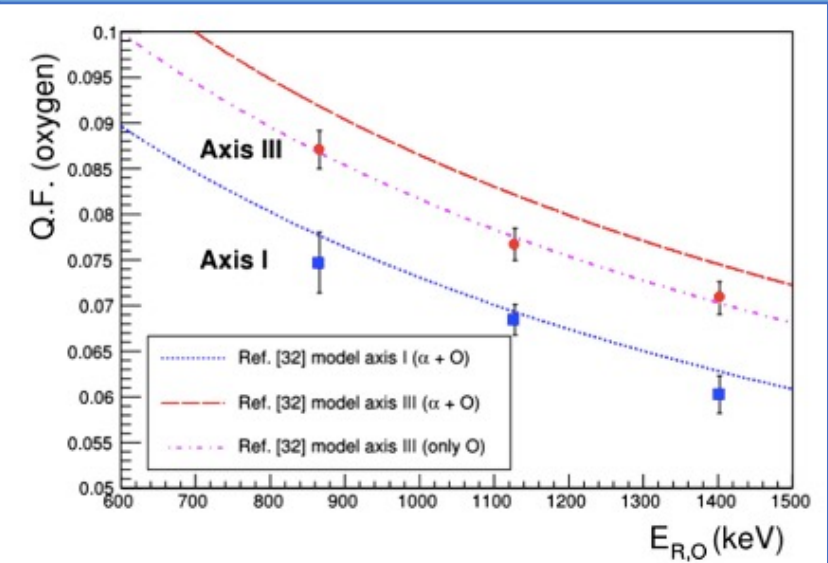
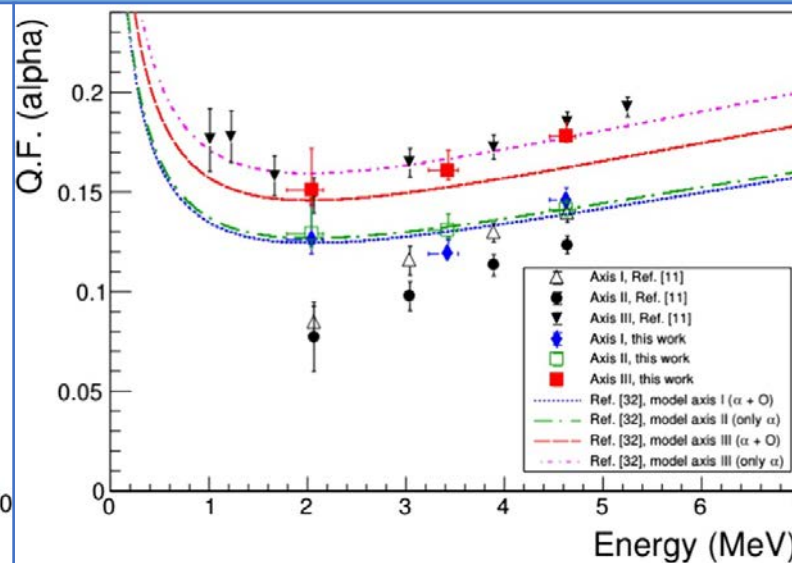
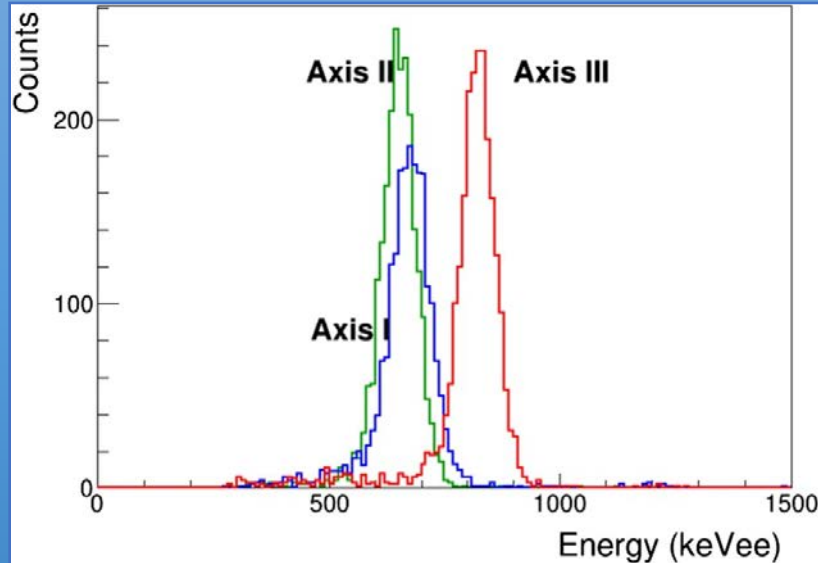
For the first time, the anisotropy for oxygen nuclear recoils in the energy region down to 100 keV was measured at  $5.4 \sigma$  C.L.

Advantages in the use of scintillating crystals

- Very good anisotropy for particles ( $p$ ,  $\alpha$ , nuclear recoils) and the nuclear charge

with  $\text{ZnWO}_4$  crystal in ADAMO

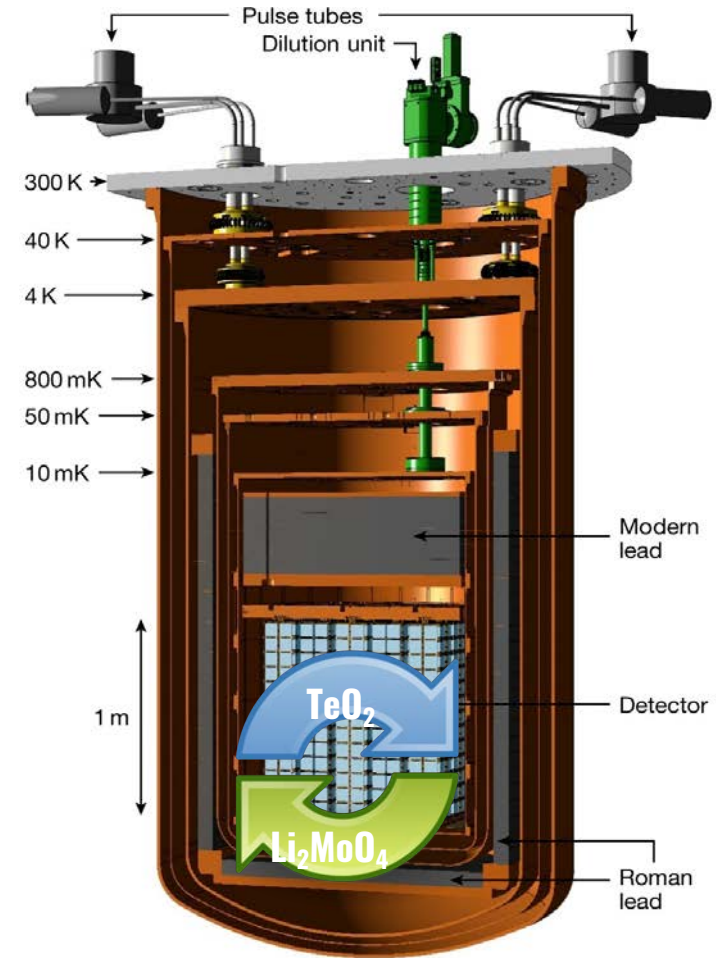
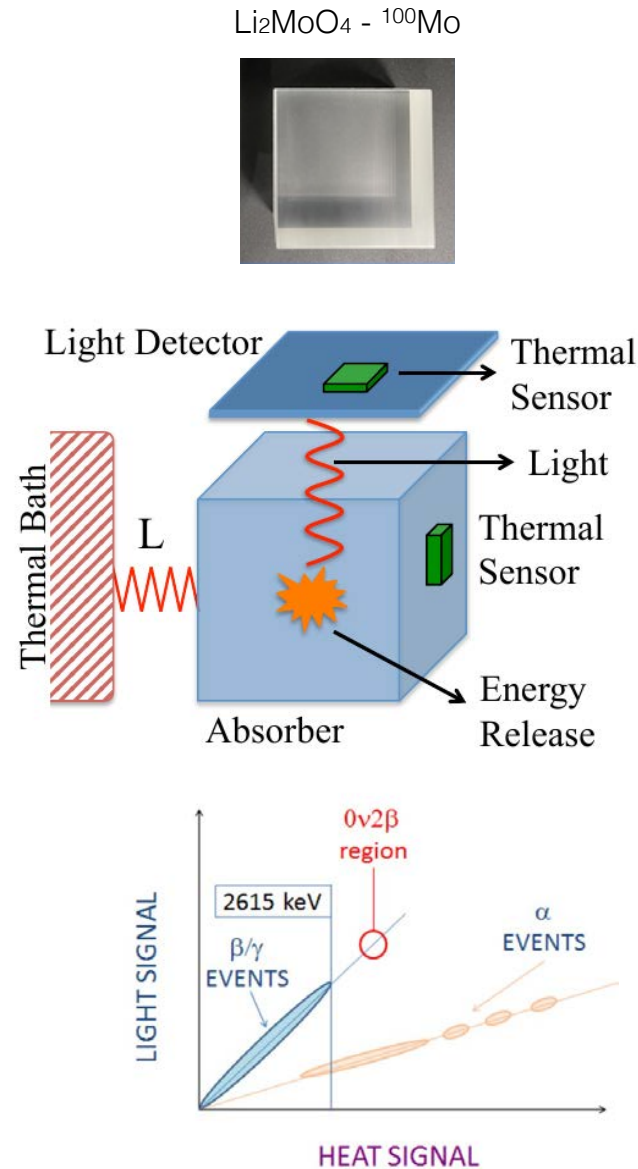
ADAMO project, recent J. A 56 (2020) 83] were the anisotropic response

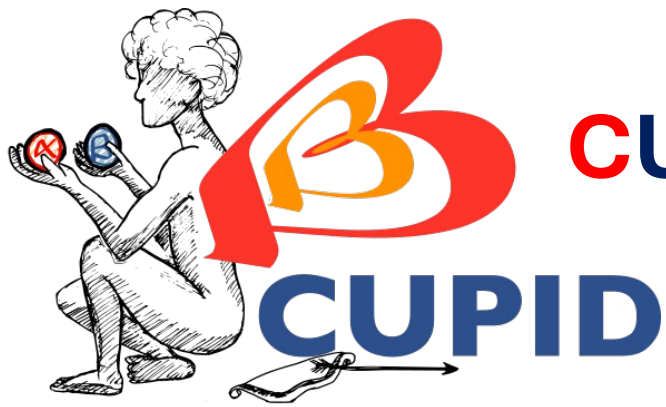




# CUORE Upgrade with Particle Identification

- ❖ Near future bolometric  $0\nu 2\beta$  experiment in existing CUORE infrastructure at LNGS.
- ❖ Replace CUORE  $\text{TeO}_2$  detector with an array of  $\text{Li}_2^{100}\text{MoO}_4$  scintillating crystals bolometers, to exploit  $^{100}\text{Mo}$  ( $Q_{2\beta} = 3034 \text{ keV}$ ) as  $2\beta$  candidate.
- ❖ New detector array:
  - 1596  $\text{Li}_2\text{MoO}_4$  scintillating crystals (280 g each)
  - 240 kg  $^{100}\text{Mo}$  (95% enrichment)
  - 1710 light detectors (Ge wafer + NTD Ge thermistor)  $\rightarrow$  scintillation signal read-out
- ❖ Sensitivity goal:  $T_{1/2}^{0\nu} > 1 \times 10^{27} \text{ yr}$ ,  $m_{\beta\beta} = 12 - 20 \text{ meV}$ .





# CUORE Upgrade with Particle Identification



## VSTT (Vertical Slice Test Tower):

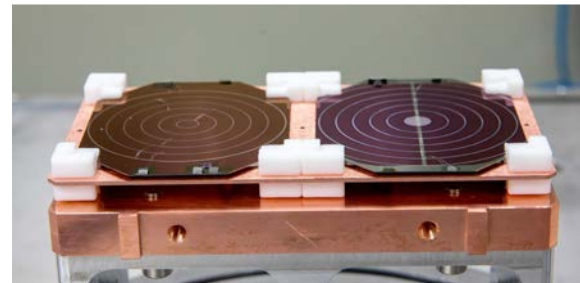
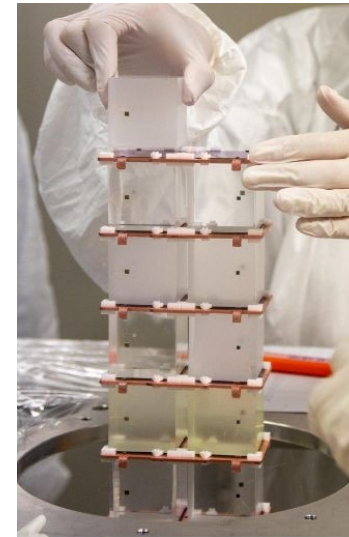
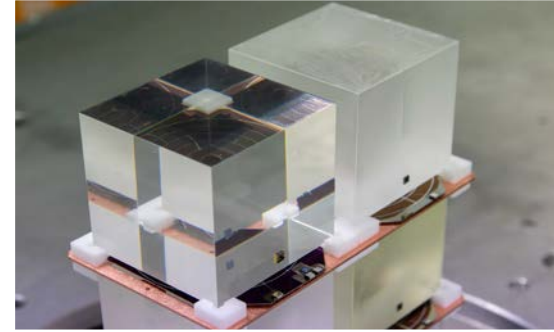
full integrated test in CUPID HallA facility @LNGS (2025)

- Mechanical assembly with upgraded tower design
- Test of the assembly-line (glueing, mounting, bonding,...)
- NTL light detectors
- Optical fibers along the tower
- New electronics and DAQ

+ test on new enriched LMO crystals (SICCAS) in HallC

VSTT cooled down in **early August 2025**, at  $T \sim 7\text{mK}$ .  
Currently taking data for characterizing the detectors response!

VSTT will prove the overall readiness to  
CUORE detector construction







# GAIAS

GAxIal Analysis with Scintillators

## GAxIal Analysis with Scintillators

- ❖ The effective value of  $g_A$  in finite nuclei is uncertain:  $g_A^{free} \simeq 1.2723$ , but inside a nucleus  $g_A \sim A^{-\alpha}$ , with  $\alpha = 0.15 - 0.25$ , depending on the nuclear model adopted to infer the  $g_A$  value.

→ Key parameter in modeling  $\beta$  and  $2\beta$  decays.

→ Directly impacts predictions for  $0\nu 2\beta$  decay sensitivity.

→ The quenching of  $g_A$  can depend on the process type and momentum transfer.

- ❖ **Forbidden non-unique  $\beta$  decays** are especially informative: study electron spectral shapes of  $\beta$  decays can be used to extract the value of  $g_A$ .

Transition type	$\Delta J^{\Delta\pi}$	$\Delta\pi$	Forbiddenness
Allowed	$0^+, 1^+$		
Forbidden <b>non-unique</b>	$0^-, 1^-, 2^+, 3^-, 4^+, \dots$	$(-1)^{\Delta J}$	$\Delta J$
Forbidden <b>unique</b>	$2^-, 3^+, 4^-, \dots$	$(-1)^{\Delta J-1}$	$\Delta J - 1$

The half-life of the a  **$\beta$  transition** can be obtained from:

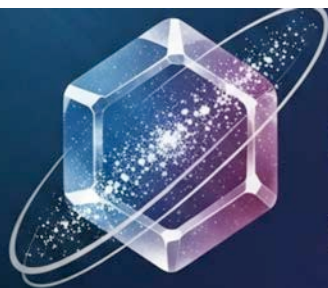
$$T_{1/2} = \kappa / \tilde{C}$$

where  $\kappa$  is a constant and  $\tilde{C}$  is the integrated shape function  $C$ , containing phase-space factors and NMEs.

The complexity of the shape function  $C$  can, however, be condensed to a simple dependence on the weak couplings by writing:

$$C(w_e) = g_V^2 C_V(w_e) + g_A^2 C_A(w_e) + g_V g_A C_{VA}(w_e)$$

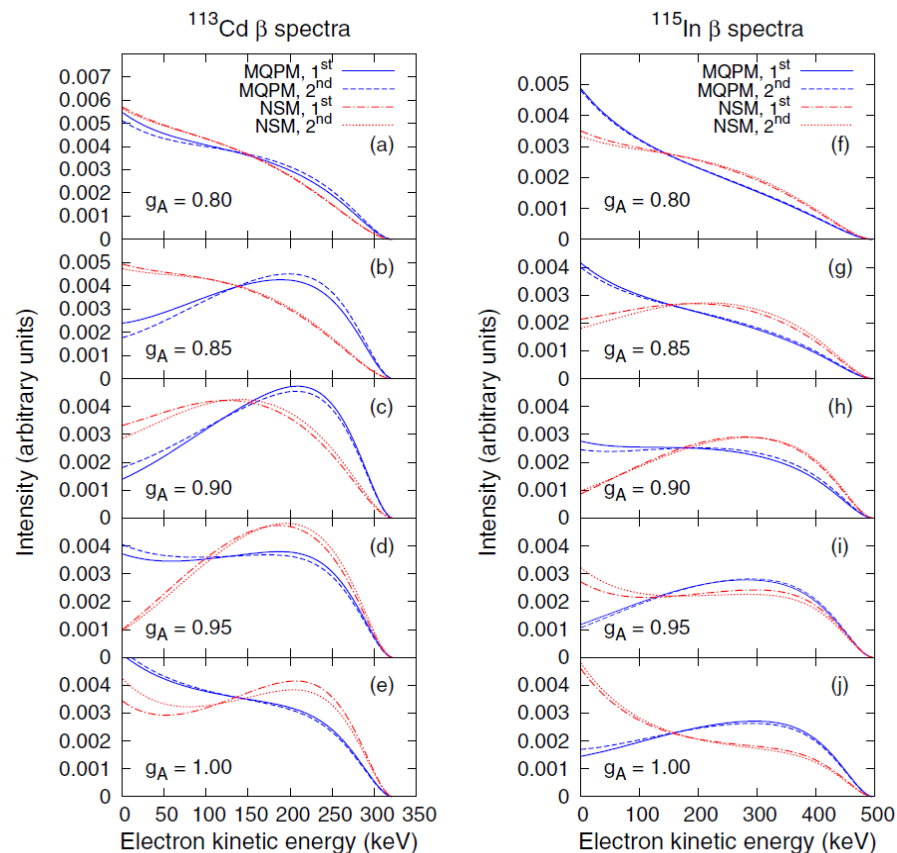
where  $w_e$  is the total (rest-mass plus kinetic) energy of the emitted electron.



# GAIAS

GAXial Analysis with Scintillators

Example of  $\beta$  spectra of the two fourth-forbidden decays for  $^{113}\text{Cd}$  and  $^{115}\text{In}$  for  $g_A = 0.8 - 1.0$ . The spectra show the theoretical expectation of the spectral shape for different values of  $g_A$  calculated with the MQPM and NSM nuclear models.



## GAXial Analysis with Scintillators

Transition	Forbiddness	$Q_\beta$ (keV) $\delta$ (%)	Half-life	Activity <sup>1</sup>	Possible scintillator, Note
$^{113}\text{Cd} \rightarrow ^{113}\text{In}$	$1/2^+ \rightarrow 9/2^+$ (100%, 4 FNU)	323.84(27) 12.2227(7)	$8.04(5) \times 10^{15}$ yr	1.8 mBq/g of Cd	$\text{CdWO}_4$
$^{113\text{m}}\text{Cd} \rightarrow ^{113}\text{In}$	$11/2^- \rightarrow 9/2^+$ (~100%, 1 FNU)	587.38(27) Synthetic	13.58(32) yr	65-130 mBq/g of $^{106}\text{CdWO}_4$	$^{106}\text{CdWO}_4$
$^{87}\text{Rb} \rightarrow ^{87}\text{Sr}$	$3/2^- \rightarrow 9/2^+$ (100%, 3 FNU)	282.275(6) 27.83(2)	$4.97(3) \times 10^{10}$ yr	867 Bq/g of Rb	$\text{CsI}(\text{Na}, \text{Rb})$ , $\text{NaI}(\text{Tl}, \text{Rb})$
$^{99}\text{Tc} \rightarrow ^{99}\text{Ru}$	$9/2^+ \rightarrow 5/2^+$ (100%, 2 FNU)	297.5(9) Synthetic	$2.111(12) \times 10^5$ yr	$6.33 \times 10^8$ Bq/g of $^{99}\text{Tc}$	$\text{NaI}(\text{Tl}, ^{99}\text{Tc})$ , $\text{CsI}(\text{Na}, ^{99}\text{Tc})$
$^{115}\text{In} \rightarrow ^{115}\text{Sn}$	$9/2^+ \rightarrow 1/2^+$ (100%, 4 FNU)	497.489(10) 95.719(52)	$4.41(25) \times 10^{14}$ yr	0.25 Bq/g of In	$\text{CsI}(\text{In})$ , $\text{LiInSe}_2$ , InI
$^{210}\text{Bi} \rightarrow ^{210}\text{Po}$	$1^- \rightarrow 0^+$ (100%, 1 FNU)	1162.2(8) daughter of $^{210}\text{Pb}$ ( $^{238}\text{U}$ chain)	5.012(5) d	$4.6 \times 10^{15}$ Bq/g of $^{210}\text{Bi}$	$\text{CaWO}_4$ , $\text{PbI}_2$
$^{135}\text{Cs} \rightarrow ^{135}\text{Ba}$	$7/2^+ \rightarrow 3/2^+$ (100%, 2 FNU)	268.70(29) Synthetic	$2.3(3) \times 10^6$ yr	$4.3 \times 10^7$ Bq/g of $^{135}\text{Cs}$	$\text{CsI}(\text{Tl})$
$^{137}\text{Cs} \rightarrow ^{135}\text{Ba}$	$7/2^+ \rightarrow 3/2^+$ (5.6%, 2 FNU)	1175.63(17) Synthetic	30.04(4) y	$3.2 \times 10^{12}$ Bq/g of $^{137}\text{Cs}$	$\text{CsI}(\text{Tl})$
$^{129}\text{I} \rightarrow ^{129}\text{Xe}^*$	$7/2^+ \rightarrow 3/2^+$ (100%, 2 FNU)	189(3) ( $10^{-13}$ - $10^{-12}$ )	$1.57(4) \times 10^7$ yr	0.7-7 $\mu\text{Bq/g}$ of I	$\text{CsI}$
$^{36}\text{Cl} \rightarrow ^{36}\text{Ar}$	$2^+ \rightarrow 0^+$ (98.1%, 2 FNU)	709.53(4) ( $\sim 7 \times 10^{-13}$ )	$3.013(15) \times 10^5$ yr	$\approx 1$ mBq/g of Cl	$\text{CeCl}_3$
$^{214}\text{Bi} \rightarrow ^{214}\text{Po}$	$1^- \rightarrow 0^+$ (19.2%, 1 FNU)	3269(11) daughter of $^{226}\text{Ra}$ ( $^{238}\text{U}$ chain)	19.71(2) min	$1.6 \times 10^{18}$ Bq/g of $^{214}\text{Bi}$	$\text{CaWO}_4$ , $\text{NaI}(\text{Tl})$ , $\text{CsI}(\text{Tl})$ , $\text{SrI}_2(\text{Eu})$ , $\text{LaCl}_3$ , ...
$^{74}\text{As} \rightarrow ^{74}\text{Se}$	$2^- \rightarrow 0^+$ (18.6%, 1 FU)	1352.8(18) Synthetic	17.77(2) d	$3.7 \times 10^{15}$ Bq/g of $^{74}\text{As}$	Highly effective organic scintillation material with an external synthetic source
$^{94}\text{Nb} \rightarrow ^{94}\text{Mo}$	$6^+ \rightarrow 4^+$ (100%, 2 FNU)	2045.0(15) Synthetic	$2.03(16) \times 10^4$ yr	$6.9 \times 10^9$ Bq/g of $^{94}\text{Nb}$	Highly effective organic scintillation material with an external synthetic source

# Conclusions

- ❖ **Novel crystal scintillators:** thanks to their high radiopurity and low-background measurements possible, they represent one of **the best solution to search for rare processes**.
- ❖ Assembling, data taking and analysis of a  $^{106}\text{CdWO}_4$  detector enriched in  $^{106}\text{Cd}$  (66%) to search for  $2\beta^+$  in  $^{106}\text{Cd}$ . The sensitivity obtained on the  $T_{1/2}$  for the case  $2\nu\epsilon\beta^+$  is well inside the theoretical predictions:  $T_{1/2} \sim 10^{21} - 10^{22}$  yr.
- ❖ Assembling, data taking and analysis of two next-generation  $\text{Cs}_2\text{ZrCl}_6$  crystal scintillators for the study of  $2\beta$  decay in Zr isotopes. Set limits  $T_{1/2} \sim 10^{18} - 10^{20}$  yr for  $^{94}\text{Zr}$  and  $^{96}\text{Zr}$  isotopes.
- ❖ Assembling, data taking and analysis of three  $\text{Cs}_2\text{ZrCl}_6$  and one  $\text{Cs}_2\text{HfCl}_6$  to search for rare  $\alpha$  and  $2\beta$  decays in Hf and Zr isotopes. New measurement of a decay of  $^{174}\text{Hf}$  to the g.s.:  $T_{1/2} = 3.8^{+1.7}_{-0.9} \times 10^{16}$  yr.
- ❖ Measurements of  $\text{ZnWO}_4$  anisotropic response to  $\alpha$  particles and nuclear recoils to study Dark Matter with the directionality approach. Study of optical and scintillation properties to improve the performances of such inorganic crystal scintillators.
- ❖ Thanks to the experience I gained during my PhD, I am now working in **CUPID**, where I perform data analysis and tests on new LMO scintillating bolometers, and in **GAIAS**, focusing on the study of the axial coupling constant  $g_A$ .



+ **PRIN PNRR Project A-DREAM** to investigate whether the half-life of the  **$\alpha$ -decay of  $^{226}\text{Ra}$**  can be significantly reduced when the isotope is embedded in a **metallic matrix** and cooled to **cryogenic temperatures ( $\sim 2.2$  K)**.



attualmente assegnista presso l'Università di Roma "Tor Vergata"

*Per i suoi contributi in fisica nucleare sperimentale nello sviluppo e caratterizzazione di cristalli scintillatori altamente radiopacchi, cui sono stati ottenuti nuovi risultati sul doppio beta nel  $^{106}\text{Cd}$  e  $^{96,98}\text{Zr}$ .*

**Thanks for attention!**

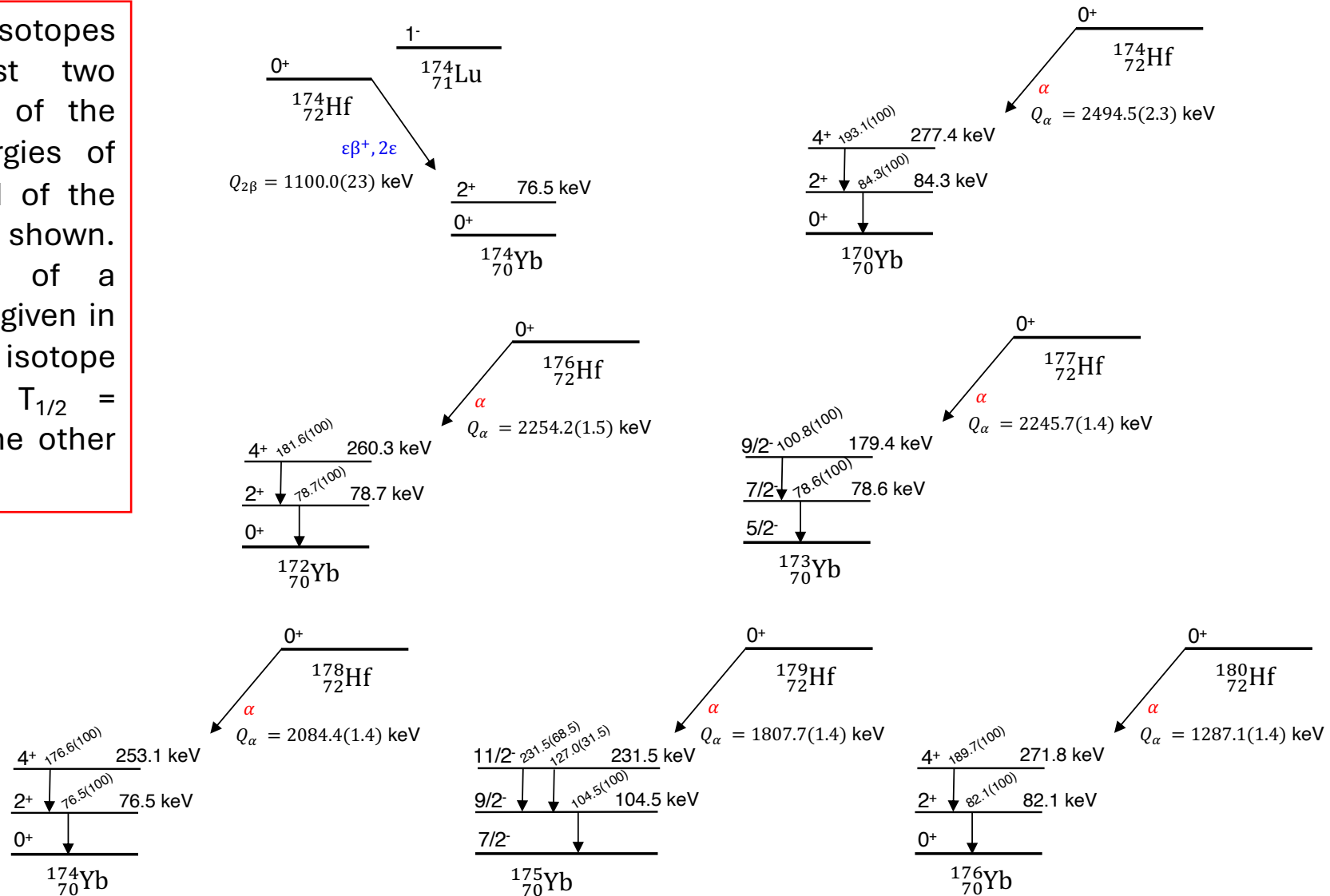
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# **BACKUP SLIDES**

# Simplified decay schemes of naturally occurring Hf isotopes

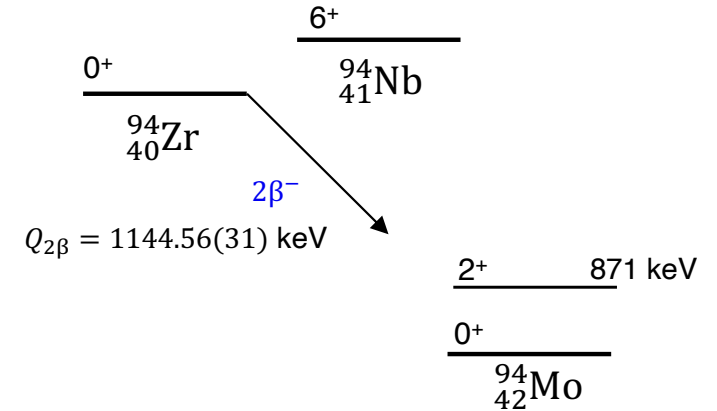
$\alpha$  decays of Hf isotopes considering the first two excited energy levels of the daughter nuclei. Energies of the excited levels and of the emitted  $\gamma$  quanta are shown. Relative probabilities of a single energy level are given in parentheses. The  $^{175}\text{Yb}$  isotope decays via 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



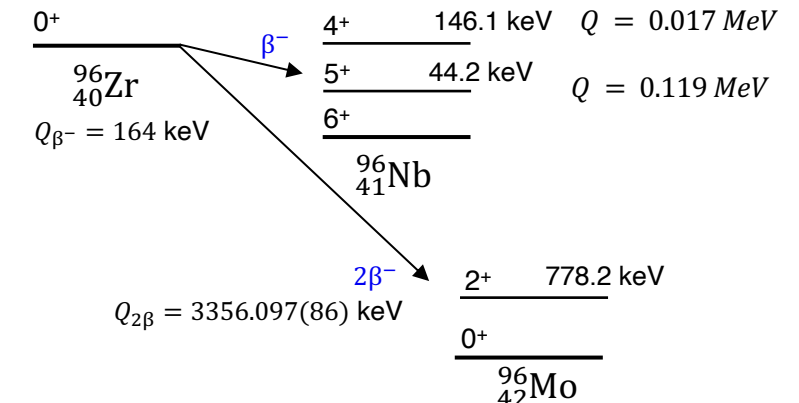
# Search for $2\beta$ decay in $^{94,96}\text{Zr}$ and for $^{96}\text{Zr}$ 's $\beta$ decay

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, Fréjus (France) (next: SuperNEMO)	$^{96}\text{Zr } 0^+ \rightarrow ^{96}\text{Mo } 0^+_1$ (g.s.)	$>9.2 \times 10^{21}$ $>1.29 \times 10^{22}$	Tracker detector	[2] [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

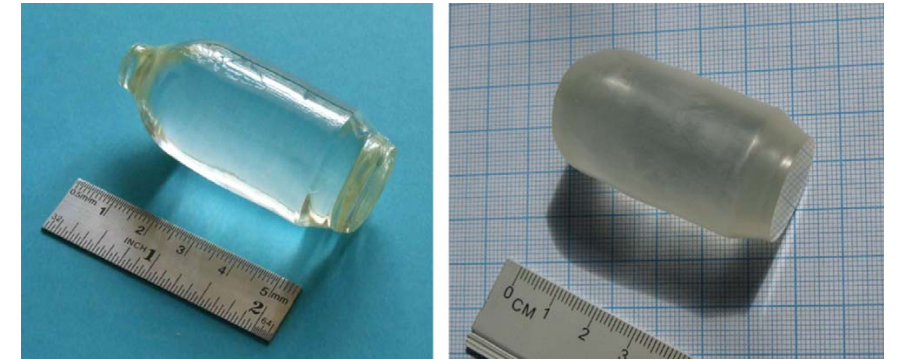
[8] S.W. Finch, W. Tornow, Nucl. Inst. Meth. A 806(2016)70–74

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

# CdWO<sub>4</sub> crystal scintillators

Some general properties	CdWO <sub>4</sub>
Effective atomic number	66
Density (g/cm <sup>3</sup> )	7.9
Melting point (°C)	1325
Refractive index	2.2 – 2.3
Emission maximum (nm)	475 – 490
Scintillation time constants (μs)	~ <b>15</b> (89%), ~4.6 (9%), ~0.8 (2%), ~0.15 (0.5%) [1]
Light Yield	≈ 15000 photons/MeV
Linearity of the energy response	Excellent, down to ~100 keV
Energy resolution (FWHM, %) @ 662 keV	~ 7.0 – 10
Pulse-shape discrimination ability	Excellent

Boule of <sup>106</sup>CdWO<sub>4</sub> single crystal grown by low-thermal-gradient Czochralski technique and crystal scintillator obtained by cutting a boule and grinding its surface.



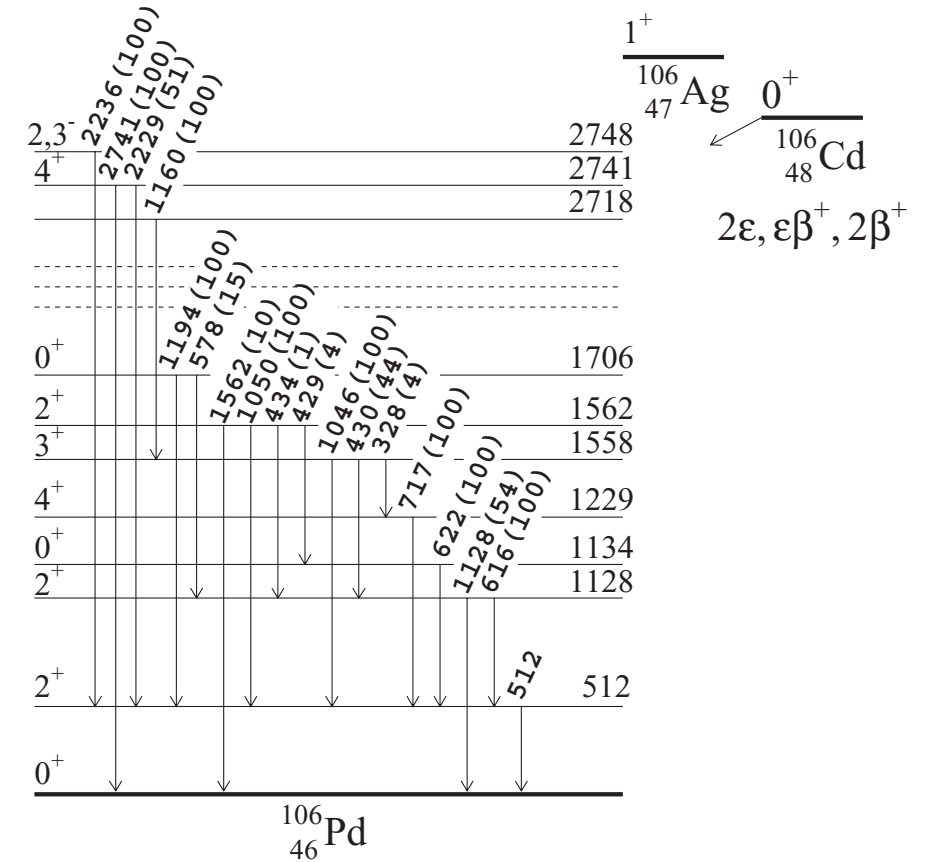
[2] Belli, P. et al., NIMA 2010, 615, 301-306.

[1] Bardelli, L. et al. Nucl. Instrum. Meth. A 2006, 569, 743–753.

# $2\beta$ decay in $^{106}\text{Cd}$

## Advantages in the use of $^{106}\text{Cd}$ :

- One of the biggest  $2\beta^+$  decay energy :  $Q_{2\beta} = (2775.39 \pm 0.10)$  keV;
- Relatively high isotopic abundance:  $\delta = (1.245 \pm 0.022) \%$ ;
- Possibility of enrichment by gas centrifugation;
- Favorable theoretical predictions for half-lives for some  $2\nu$  modes ( $T_{1/2} \sim 10^{21} - 10^{22}$  yr) that could be reached by modern low-counting techniques;
- Possibility of «near resonant»  $0\nu 2\varepsilon$  to excited levels of  $^{106}\text{Pd}$ ;
- Existing technologies of cadmium purification and availability of Cd-containing detectors to realize calorimetric experiments with a high detection efficiency.



# Estimation of half-lives limits

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

- $N$  is the number of nuclei of interest in the crystal;
- $\epsilon$  is the detection efficiency for the process of decay (calculated as a ratio of the events number in the signal model which satisfies the investigated experimental condition, to the number of generated events);
- $t$  is the time of measurements;
- $\lim S$  is the number of events of the effect searched for, which can be excluded at a given confidence level (C.L.; in the present study all limits are given at the 90% C.L.).

## Half-life of $\alpha$ decay of $^{174}\text{Hf}$ to the g.s. of $^{174}\text{Yb}$

Area of 2<sup>nd</sup> peak =  $118 \pm 11(\text{stat}) \pm 35(\text{sys}) = 118 \pm 37$

Half-life:

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

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

$M = 16.87 \text{ g};$

$W(\text{Cs}_2\text{HfCl}_6) = 657 \text{ g/mole};$

$\delta(^{174}\text{Hf}) = 0.156(6) \%$

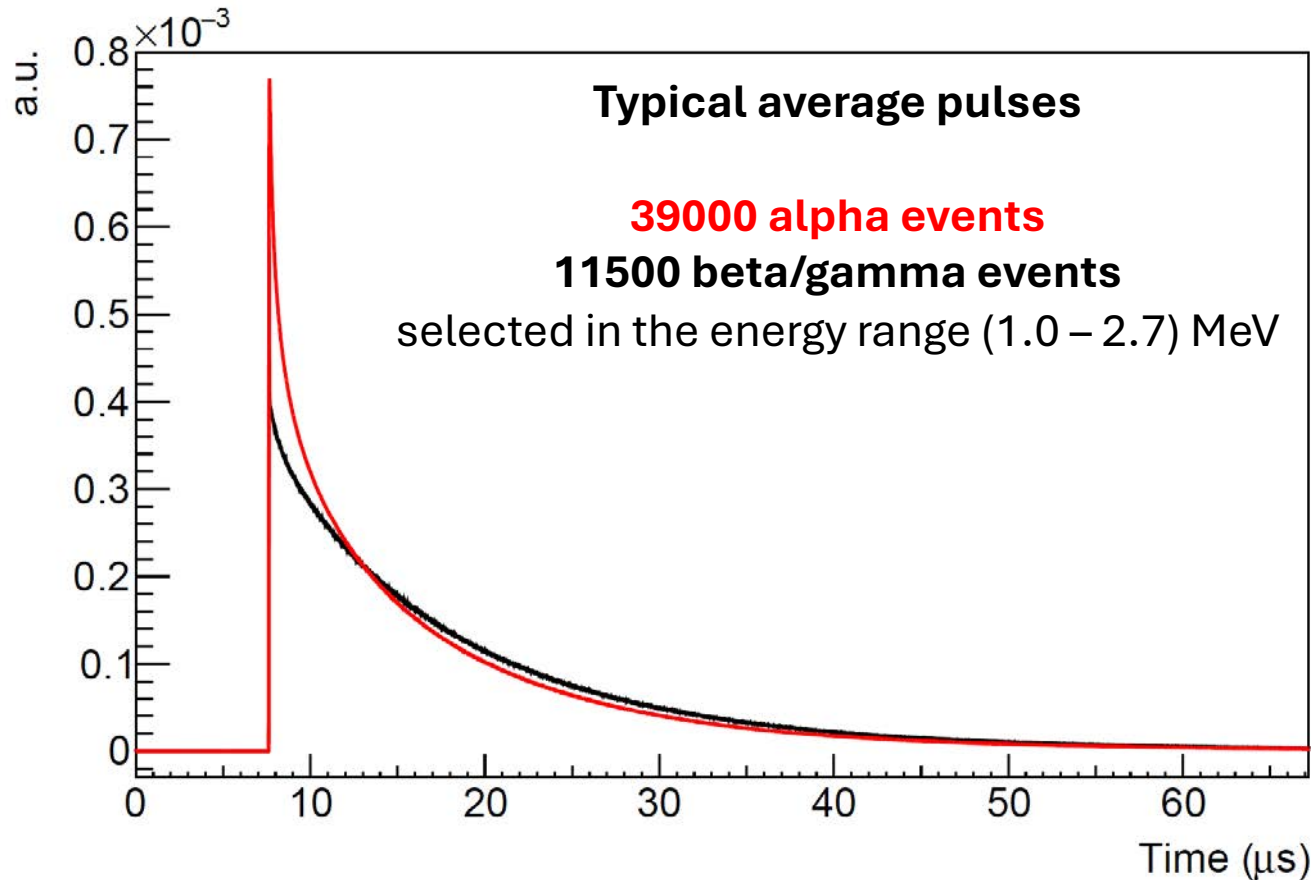
$$\Rightarrow T_{1/2} = [3.8_{-0.3}^{+0.4}(\text{stat})_{-0.9}^{+1.6}(\text{sys})] \times 10^{16} = 3.8_{-0.9}^{+1.7} \times 10^{16} \text{ yr of } \alpha \text{ decay of } ^{174}\text{Hf}$$

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

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



# 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:

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

where the sum is over the time channels ( $k$ ), starting from the origin of pulse up to 24  $\mu\text{s}$ ,  $f(t)$  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  $8.00 \mu\text{s}$ , for alpha and beta/gamma events respectively

# Cs<sub>2</sub>HfCl<sub>6</sub> and Cs<sub>2</sub>ZrCl<sub>6</sub> crystal radiopurity

measured with the ultra-low background **HP-Ge**  $\gamma$  spectrometers of the **STELLA** facility at LNGS over  $\sim 700$  hours.

[1] P. Belli et al. *Nucl. Phys. A* **1053**, 122976 (2025).  
[2] P. Belli et al. *Eur. Phys. J. A* **59**, 176 (2023).

Chain	Nuclide	Activity (mBq/kg)		
		CHC	CZC	
			Cone	Cylinder
		<b>16.87 g [1]</b>	<b>10.63 g [2]</b>	<b>23.95 g [2]</b>
<sup>238</sup> U	<sup>226</sup> Ra	<13	60(10)	< 8.7
	<sup>234</sup> Th	<1200	< 180	< 260
	<sup>234m</sup> Pa	<18	< 630	< 160
<sup>235</sup> U	<sup>235</sup> U	<18	< 16	< 12
<sup>232</sup> Th	<sup>228</sup> Ra	<13	< 16	< 23
	<sup>228</sup> Th	<17	< 6.7	< 8.2
	<sup>137</sup> Cs	<10	< 7.1	< 1.6
	<sup>134</sup> Cs	37(4)	49(6)	42(5)
	<sup>132</sup> Cs	-	< 8.2	< 11
	<sup>40</sup> K	<240	<120	<95

Surface cross-contamination happened during the sample preparation and installation.

**Natural**

**Artificial**

**Cosmogenic activation**

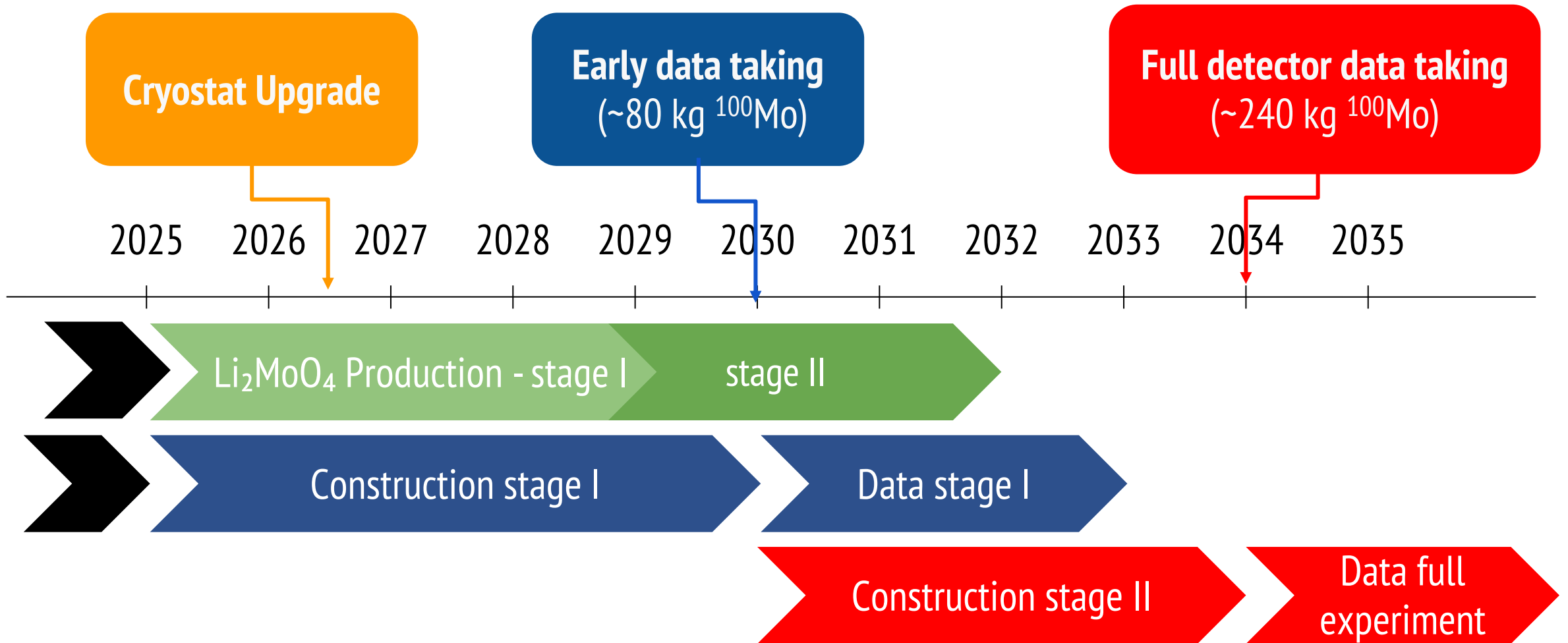
**Natural**

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***

# CUPID timeline

## CUPID staged deployment





# Accelerated Depletion of Radioactive Waste in Metals (A-DREAM)

- Aim to investigate whether the  $\alpha$ -decay of  $^{226}\text{Ra}$  can be significantly reduced when the isotope is embedded in a **metallic matrix** and cooled to **cryogenic temperatures** ( $\sim 2.4\text{ K}$ ).
- If confirmed, this effect could revolutionize the management of **radioactive waste**, drastically shortening its storage time and mitigating environmental impact.



Measurements with a **metallic Ga-Hg alloy samples** incorporating trace amounts of  $^{226}\text{Ra}$ , prepared through an electrolytic reduction process, are ongoing in our laboratory in Tor Vergata.

