LRT2026

Low Radioactivity Techniques

21–24 September 2026

The Guildhall, York

Rare alpha decays of natural Hf isotopes

A. Leoncini [1,2], P. Belli [1,2], R. Bernabei [1,2], F. Cappella [3,4], V. Caracciolo [1,2], R. Cerulli [1,2], A. Incicchitti [3,4], M. Laubenstein [5], V. Merlo [1,2], S. Nagorny [5,6], V. Nahorna [7], S. Nisi [5], P. Wang [7]

[1] Dipartimento di Fisica, Università di Roma 'Tor Vergata', I-00133 Rome, Italy
[2] INFN Sezione di Roma Tor Vergata, I-00133 Rome, Italy
[3] INFN Sezione Roma, I-00185 Rome, Italy
[4] Dipartimento di Fisica, Università di Roma 'La Sapienza', I-00185 Rome, Italy
[5] INFN Laboratori Nazionali del Gran Sasso, 67100 Assergi (AQ), Italy
[6] Gran Sasso Science Institute, L'Aquila I-67100, Italy
[7] Department of Chemistry, Queen's University, Kingston, ON K7L 3N6, Canada

Why search for rare α decays?

$$\alpha : {}^{A}_{Z}X \rightarrow {}^{A}_{Z-2}Y + {}^{4}_{2}He$$

General physics motivation

Rare α decays probe nuclear properties at extreme time scales

$$T_{1/2} \gtrsim 10^{14}\ yr$$

- **Nuclear structure**
  Stringent tests of α-decay models and nuclear-level properties.
- **Fusion–fission & superheavy nuclei**
  Input for α–nucleus interactions and superheavy-element stability.
- **Nuclear astrophysics**
  Relevant to α-capture reactions and nucleosynthesis studies.
- **Experimental challenge**
  Ultra-low background, radiopure materials and α-event identification.

Historical motivation

A long-standing, order-of-magnitude discrepancy between the early experimental ^{174}Hf half-life and theoretical predictions remained unresolved for decades.

Transition	Level of the daughter nucleus	Early experimental $T_{1/2}(yr)$	Theoretical predictions of $T_{1/2}(yr)$
^{174}Hf→^{170}Yb	0^+, 0 keV g.s. → g.s.	$2.0(4)\times10^{15}$ [*R.D. Macfarlane and T.P. Kohman, Phys. Rev. 121, 1758 (1961)*]	$(3.0 \text{ to } 7.0)\times10^{16}$

Cs_2HfCl_6 (CHC) crystal scintillators were first proposed as radiation detectors in 2015 showing promising scintillation properties.

Cs_2HfCl_6 (CHC) crystal scintillators

Some general properties	Cs_2HfCl_6
Effective atomic number	58
Density (g/cm^3)	3.9
Melting point (°C)	820
Crystal structure	Cubic
Emission maximum (nm)	400 - 430
Scintillation time constants (μs)	0.4; 5.1; 15.2 *
Light Yield	up to 30000 photons/MeV**
Linearity of the energy response	Excellent, down to 100 keV
Energy resolution (FWHM, %) @ 662 keV	3.2 - 3.7***
Pulse-shape discrimination ability	Excellent
Hf mass fraction (%)	27

* *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*

P. Belli et al. J. Phys. G: Nucl. Part. Phys. 53 (2026) 075112

Production and growth of CHC crystals

Produced at **Queen's University, Canada**

1. Starting materials

Low-purity (LP): CsCl (99.9%) + $HfCl_4$ (98%)
High-purity (HP): CsCl (99.999%) + $HfCl_4$ (99.9%)

2. $HfCl_4$ purification

Both $HfCl_4$ powders purified by a three-stage sublimation process

3. Synthesis in quartz ampoules

- Stoichiometric CsCl + purified $HfCl_4$ in a tapered 22 mm quartz ampoule;
- dried at 120 °C for 12 h in vacuum;
- reduction step at 300 °C for 1 h under H_2.

4. Compound formation

Sealed ampoules heated at 850 °C for 24 h to form Cs_2HfCl_6 and homogenize the melt

5. Vertical Bridgman–Stockbarger growth

Two-stage crystallization with the same gradient $\Delta T = 25$ °C cm^{-1}
Fast stage: 1.5 mm/h; Slow stage: 0.5 mm/h

6. Controlled cooldown

Boules cooled to room temperature at 0.1 °C min^{-1} to prevent cracking

Output of the growth

Two crystalline boules were cut by diamond wire

Boule	Samples	Mass (g)
HP	HP-1 cone HP-2 cylinder	9.8 19.6
LP	LP-1 cone LP-2 cylinder	15.4 17.0

P. Belli et al. J. Phys. G: Nucl. Part. Phys. 53 (2026) 075112

Radiopurity control: Sm contamination in CHC crystals

- ^{147}Sm is an α emitter with $\mathbf{E}_{\alpha} = \mathbf{2249.9}$ keV, close to the ^{174}Hf α decay ($\mathbf{E}_{\alpha} = \mathbf{2437.6}$ keV).
- Because of the limited energy resolution, a Sm contribution can partially overlap the ^{174}Hf region of interest.
- Therefore even a ppb-level Sm contamination must be carefully constrained.

Crystal sample	Sm Concentration (ppb)	
	Measured value	Conservative limit
LP-1	0.3 ± 0.2	< 0.5
LP-2	0.3 ± 0.2	< 0.5
HP-1	0.7 ± 0.4	< 1.1
HP-2	0.4 ± 0.2	< 0.6

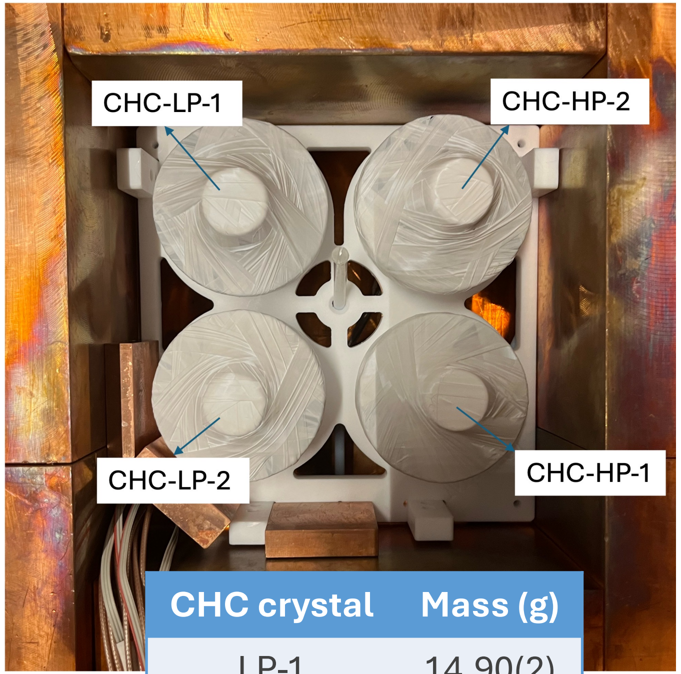
ICP-MS assay

- Dedicated double-focusing ICP-MS measurements in medium-resolution mode.
- ^{152}Sm was used for quantification because it provided the best signal-to-background ratio.
- The possible ^{152}Gd isobaric contribution was corrected by monitoring ^{156}Gd and ^{157}Gd.
- All four crystals show Sm concentrations at the sub-ppb level.

P. Belli et al. J. Phys. G: Nucl. Part. Phys. 53 (2026) 075112

The key experimental idea: “source = detector”

Hf is an intrinsic constituent of CHC (~27 wt%), enabling a high detection efficiency.



CHC crystal	Mass (g)
LP-1	14.90(2)
LP-2	14.25(2)
HP-1	9.50(2)
HP-2	18.87(3)

Detector preparation

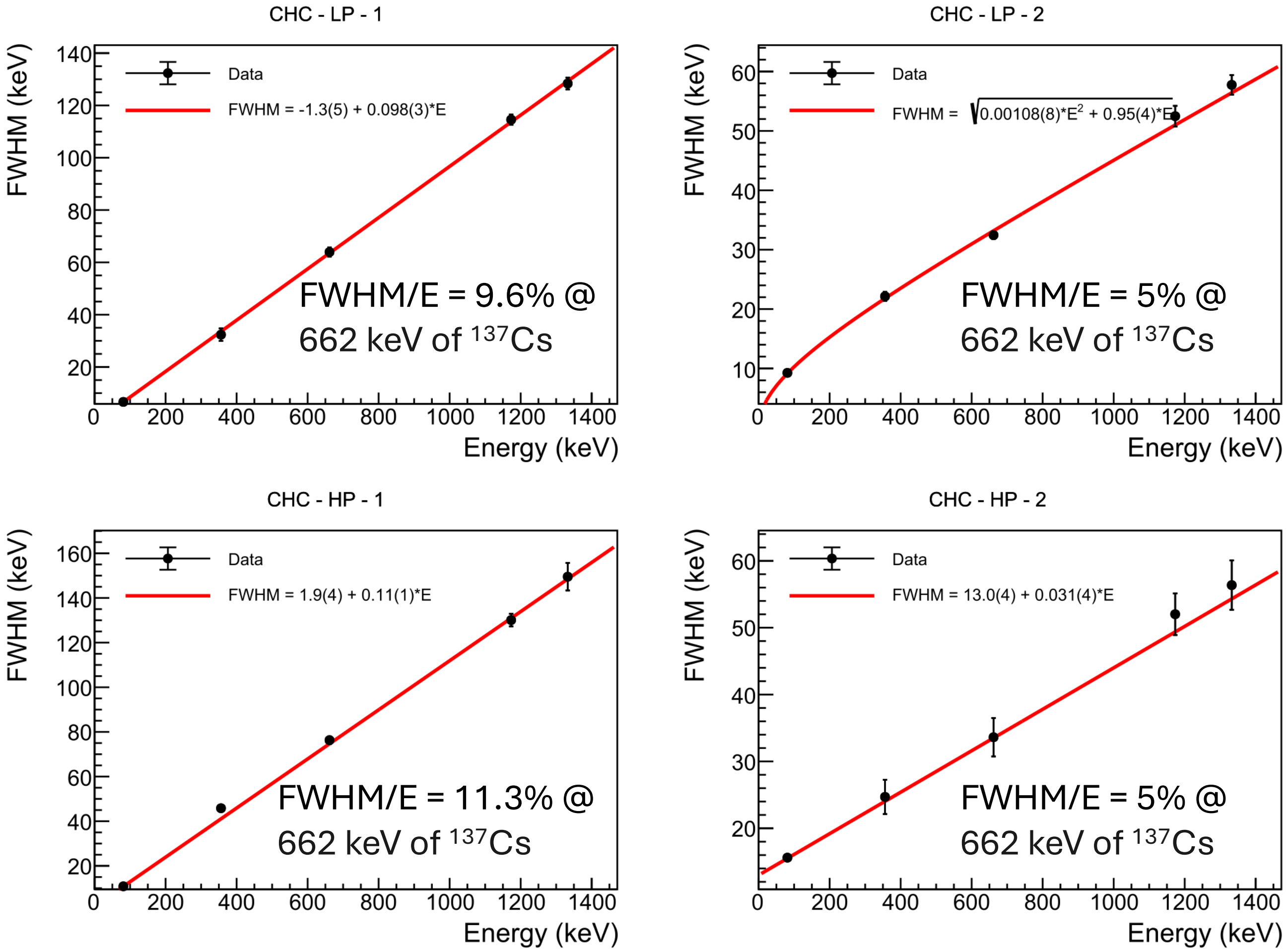
- Cleaned with toluene
- Encapsulated using the SYLGARD 184™ Silicone Elastomer Kit
- A fused quartz window at the bottom of the mould served as optical window and mechanical support
- After curing (>24 h at room temperature), each detector was wrapped in Teflon tape

Low-background setup

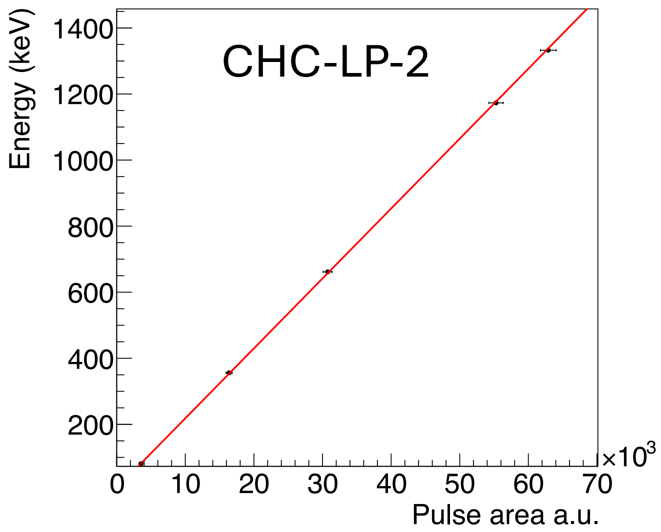
- Each CHC detector was optically coupled to a Hamamatsu PMT
- Installed deep underground in the DAMA/CRYS setup at LNGS
- Passive shield: 15 cm OFHC Cu + 20 cm low-radioactivity Pb + 5 cm HDPE
- The apparatus was enclosed in a polyethylene box continuously flushed with HP-N_2
- Measurement conditions: $T = (20.0 \pm 0.5)$ °C, live time = 85.23 d

P. Belli et al. J. Phys. G: Nucl. Part. Phys. 53 (2026) 075112

Detector performance: energy calibration and resolution



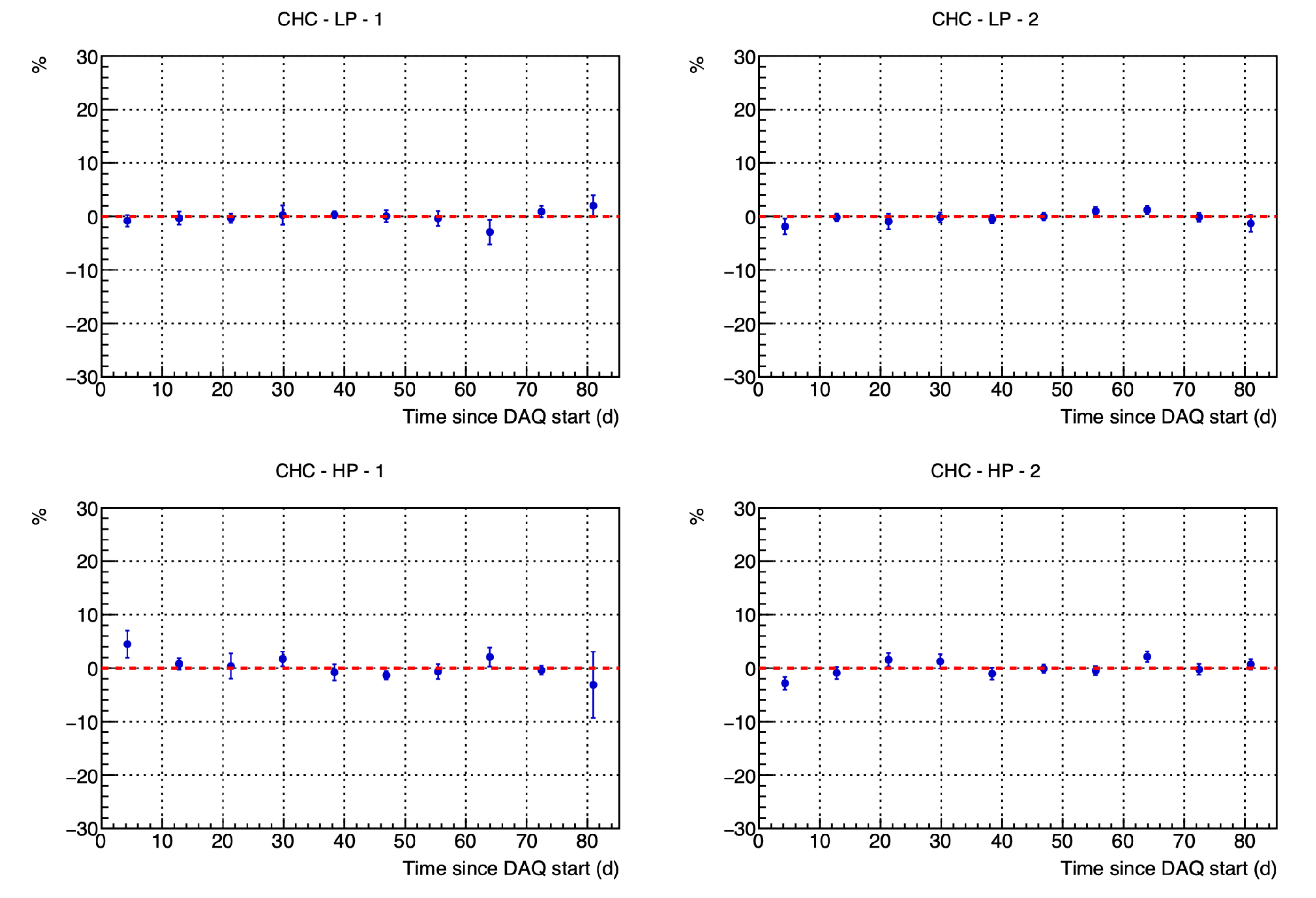
- Periodic energy calibration with ^{60}Co, ^{133}Ba, ^{137}Cs and ^{228}Th sources.
- Linear energy response from 81 keV to 2615 keV over the full data-taking period.



P. Belli et al. J. Phys. G: Nucl. Part. Phys. 53 (2026) 075112

Stability of the energy-scale during data taking

Monitoring of the 609 keV γ line from ^{214}Bi over the full 85.23 d run



Stability check

- Data divided into 10 subsets of ~8 d
- Centroids fitted with a constant value for each crystal
- Linear trends tested to search for gain drift

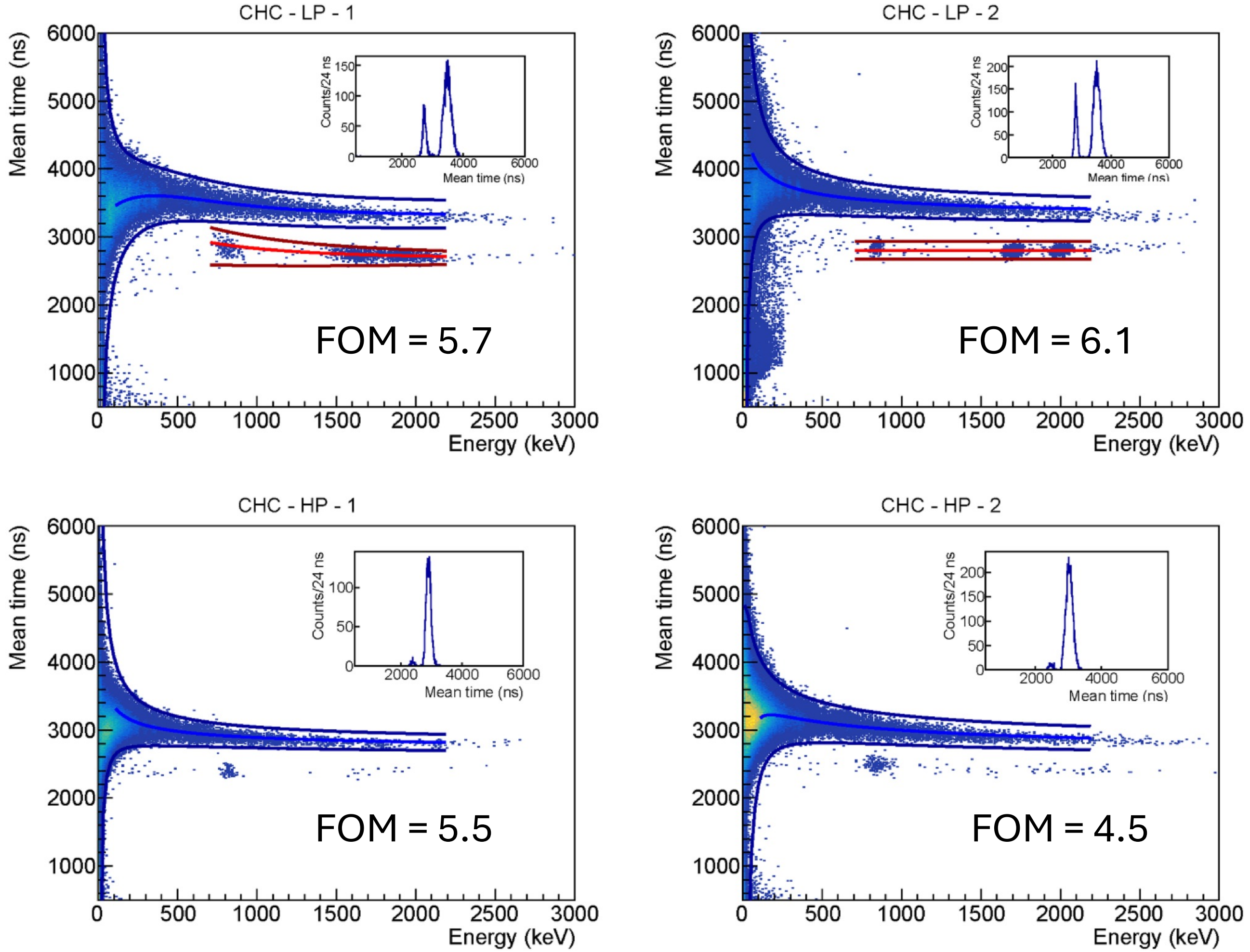
Relative spread of the peak position

CHC-LP-1: **0.78%**
CHC-LP-2: **0.79%**
CHC-HP-1: **1.4%**
CHC-HP-2: **1.3%**

- ➢ No significant gain drift observed
- ➢ Slopes consistent with zero within $\sim 2\sigma$

P. Belli et al. J. Phys. G: Nucl. Part. Phys. 53 (2026) 075112

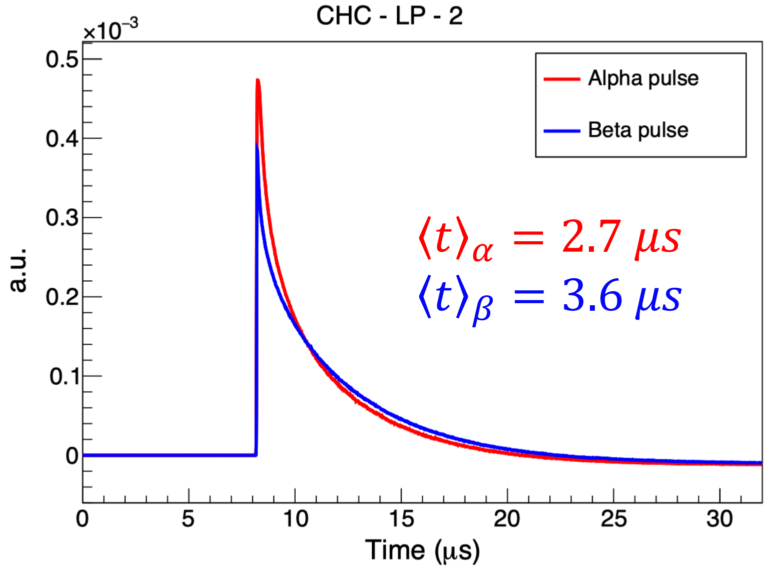
PSD between α and $\beta(\gamma)$



- Quality of discrimination:

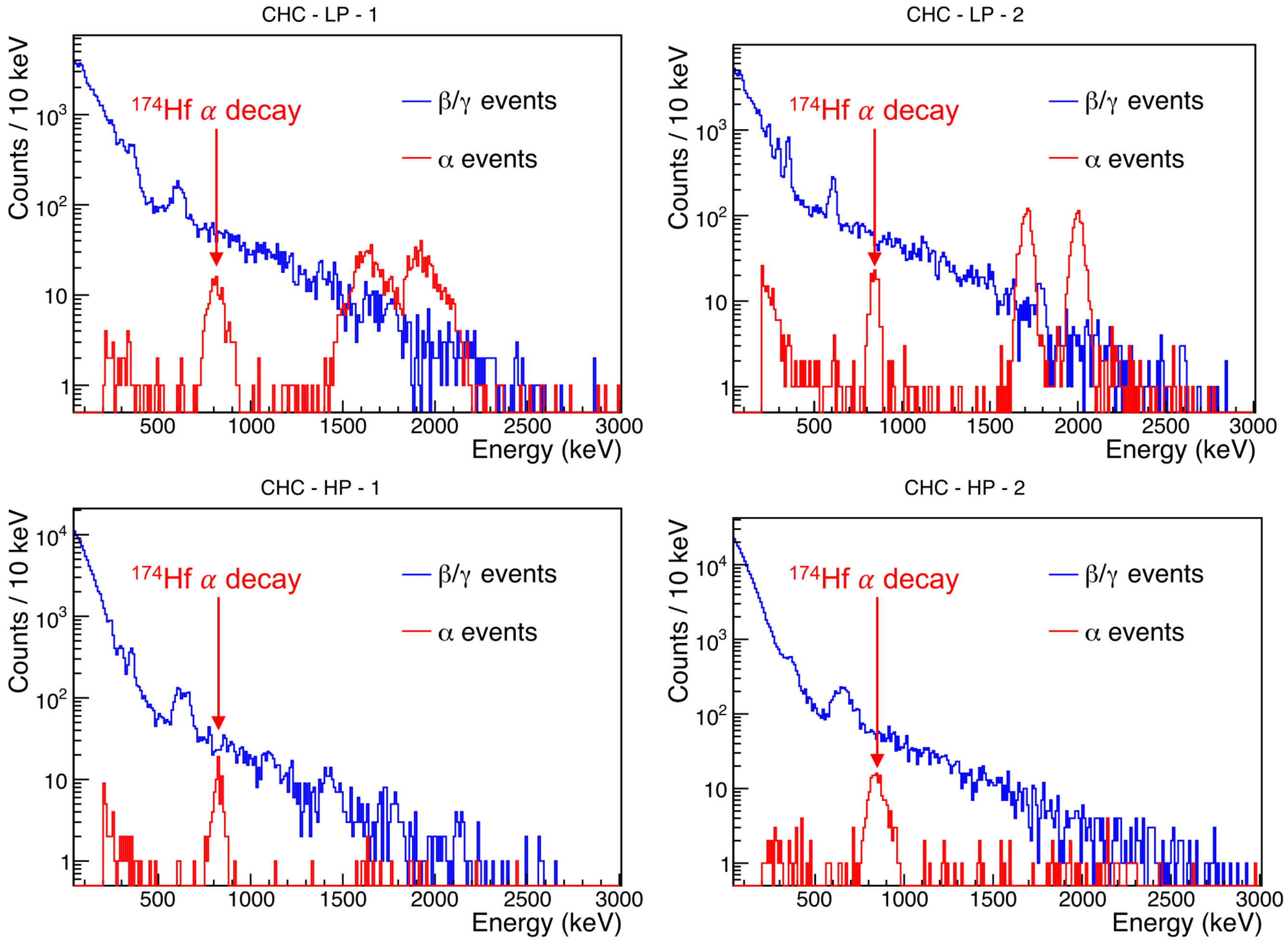
$$\text{FOM} = \frac{|\mu_\alpha - \mu_\beta|}{\sqrt{\sigma_\alpha^2 + \sigma_\beta^2}}$$

- Efficiency of the pulse shape discrimination (PSD) cut is 99.7%.



P. Belli et al. J. Phys. G: Nucl. Part. Phys. 53 (2026) 075112

PSD-selected energy spectra



- A simultaneous model of the ^{174}Hf ($E_\alpha = 2437.6$ keV) and ^{147}Sm ($E_\alpha = 2249.9$ keV) α peaks is required because the two α energies are close after quenching in the scintillator.
- $QF_\alpha = 0.331 - 0.346$

P. Belli et al. J. Phys. G: Nucl. Part. Phys. 53 (2026) 075112

Search for α decay of ^{174}Hf

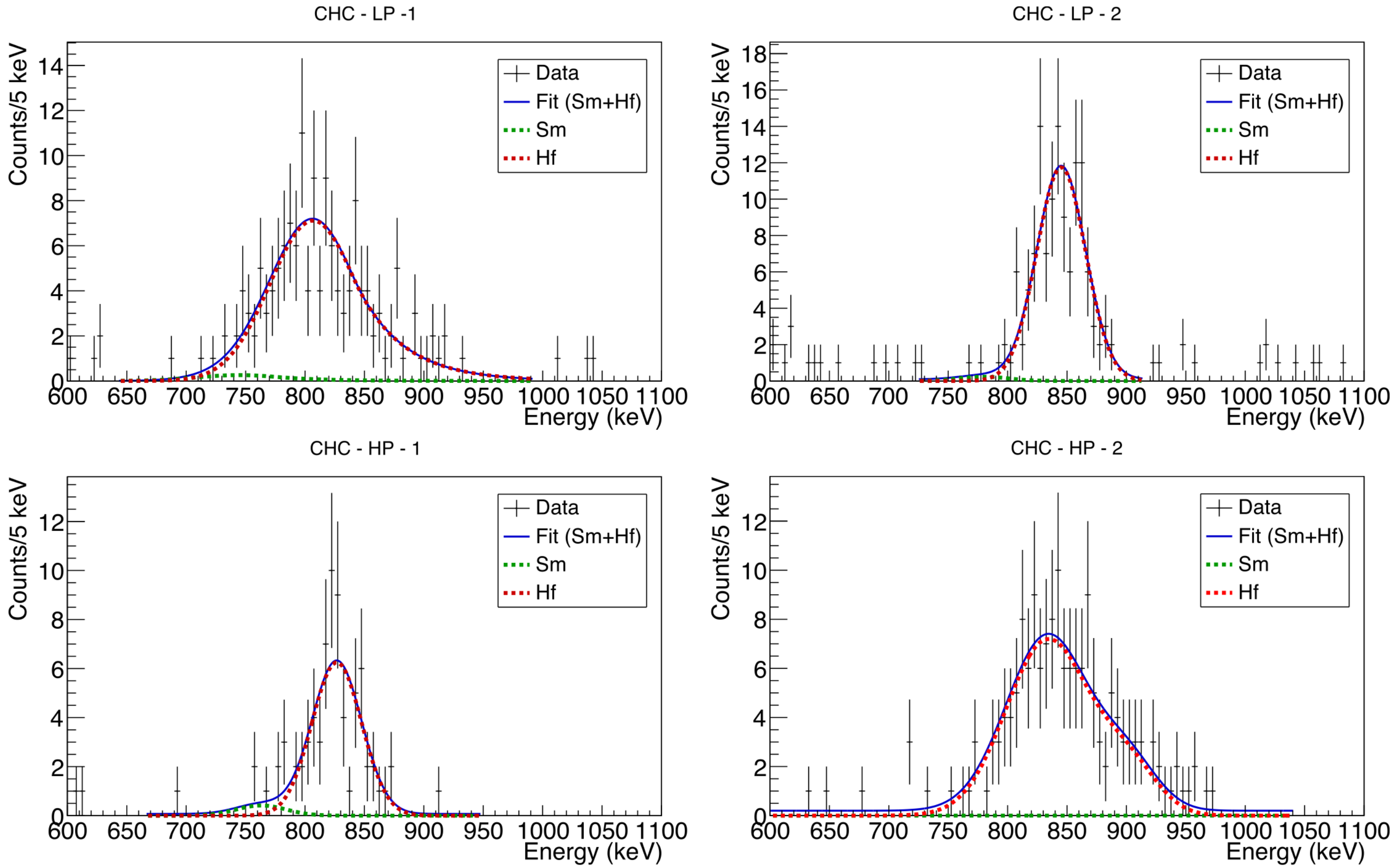
Fit model: ^{174}Hf + ^{147}Sm + const background

This work:

$$T_{1/2}(^{174}\mathrm{Hf} \rightarrow ^{170}\mathrm{Yb\ g.s}) = [2.95^{+0.15}_{-0.14}(stat) \pm 0.09\ (\mathrm{sys})] \times 10^{16}\ \mathrm{yr}$$

Final combined value with CHC – 2025 *[P. Belli et al. Nucl. Phys. A 1053 (2025) 122976]* **:**

$$T_{1/2} = (2.95 \pm 0.16) \times 10^{16}\ \mathrm{yr}$$



P. Belli et al. J. Phys. G: Nucl. Part. Phys. 53 (2026) 075112

Fit-derived constraint on ^{147}Sm contamination

CHC crystal	Concentration of ^{147}Sm (ppb), in brackets the upper limits (90 % C.L.)
LP-1	0.33 ± 0.47 (< 1.1)
LP-2	0.18 ± 2.00 (< 3.5)
HP-1	0.50 ± 0.80 (< 1.8)
HP-2	0.00 ± 1.18 (< 1.9)

Compatible with zero; consistent with independent ICP-MS assay.

^{147}Sm is not significantly observed, but it is explicitly constrained in the ^{174}Hf analysis.

P. Belli et al. J. Phys. G: Nucl. Part. Phys. 53 (2026) 075112

Search for α decay of the other naturally occurring Hf isotopes

New limits have been set on the α decays of naturally occurring Hf isotopes.

Isotope	E_α (keV)	I.a.(%)*	$T_{1/2}$(yr) at 90% C.L.
^{176}Hf	2203.3(1.5)	5.26(7)	$> 2.3\times10^{19}$
^{177}Hf	2195.3(1.4)	18.60(9)	$> 8.2\times10^{19}$
^{178}Hf	2037.9(1.4)	27.28(7)	$> 2.5\times10^{20}$
^{179}Hf	1767.6(1.4)	13.62(2)	$> 7.5\times10^{19}$
^{180}Hf	1258.7(1.4)	35.08(16)	$> 1.5\times10^{20}$

First experimental limit!

* Natural isotopic abundances were adopted and confirmed by ICP-MS.

Conclusions and outlook

- ✓ A precise measurement of the ^{174}Hf α decay half-life was obtained with 4 CHC, yielding a final combined value of:

$$\mathbf{T_{1/2} = (2.95 \pm 0.16) \times 10^{16}\ yr}$$

- ✓ The result improves the precision of ^{174}Hf α decay half-life and contributes to resolving the long-standing discrepancy between earlier experimental results and theoretical predictions
- ✓ New experimental limits on naturally occurring Hf isotopes were set.
- ✓ The first limit on the ^{180}Hf α decay was obtained: $\mathbf{T_{1/2} > 1.5{\times}10^{20} yr}$
- ✓ CHC are promising detectors for rare $\boldsymbol{\alpha}$ decay searches thanks to their high Hf mass fraction, good scintillating performance and strong PSD
- ✓ **Next steps**: increase crystal mass, enhance optical quality and radiopurity, further constrain ultra-trace Sm contamination.