Low Radioactivity Techniques (LRT2024)

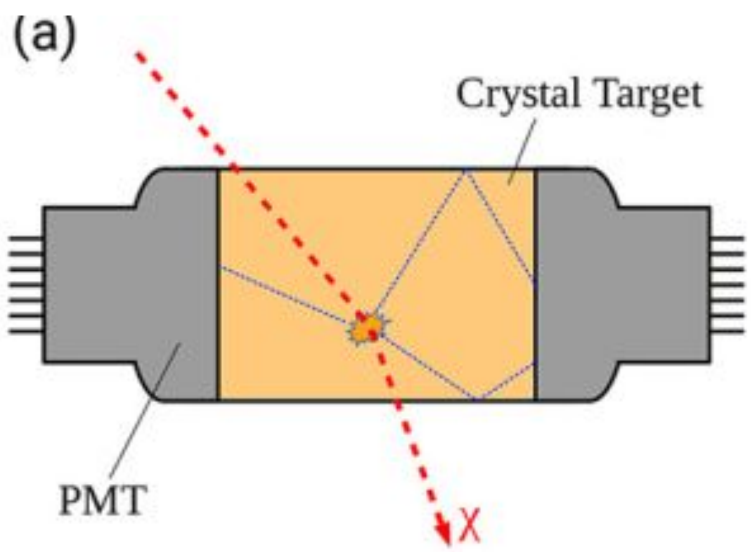
1–4 Oct 2024 - Kraków, Poland

Production of highly radio-pure NaI(Tl) crystals applied to dark matter search

Claudia Tomei - INFN Roma

on behalf of SABRE North and SABRE South Collaborations

The choice of NaI and NaI-based DM experiments



Experiment	Location	Target	Mass [kg]	Status
DAMA/LIBRA	LNGS	NaI(Tl)	250	running
ANAIS-112	LSC	NaI(Tl)	112.5	running
COSINE-100	Y2L	NaI(Tl)	106/61.3	upgrading
COSINE-200	Yemilab	NaI(Tl)	~200	in preparation
SABRE North / South	LNGS + SUPL	NaI(Tl)	~50 each	in preparation
COSINUS	LNGS	NaI	~1	in preparation
PICOLON	Kamioka	NaI(Tl)	~50	in preparation

credit: A. Ianni - IDM 2024

- well-known experimental technique, scalability
- possibility to grow large (~10 kg) crystals
- high duty cycle, high light output and good alpha/beta PSD
- possibility to carry on routine calibration in the keV range
- sensitivity to different DM scenarios and interactions

Disadvantages:

- hygroscopic crystals
- growing large crystals with the required radio purity has proven very challenging

So far DM-Ice, NaIAD, DAMA/LIBRA, ANAIS-112, and COSINE-100 have deployed arrays of NaI(Tl) detectors to search for DM.

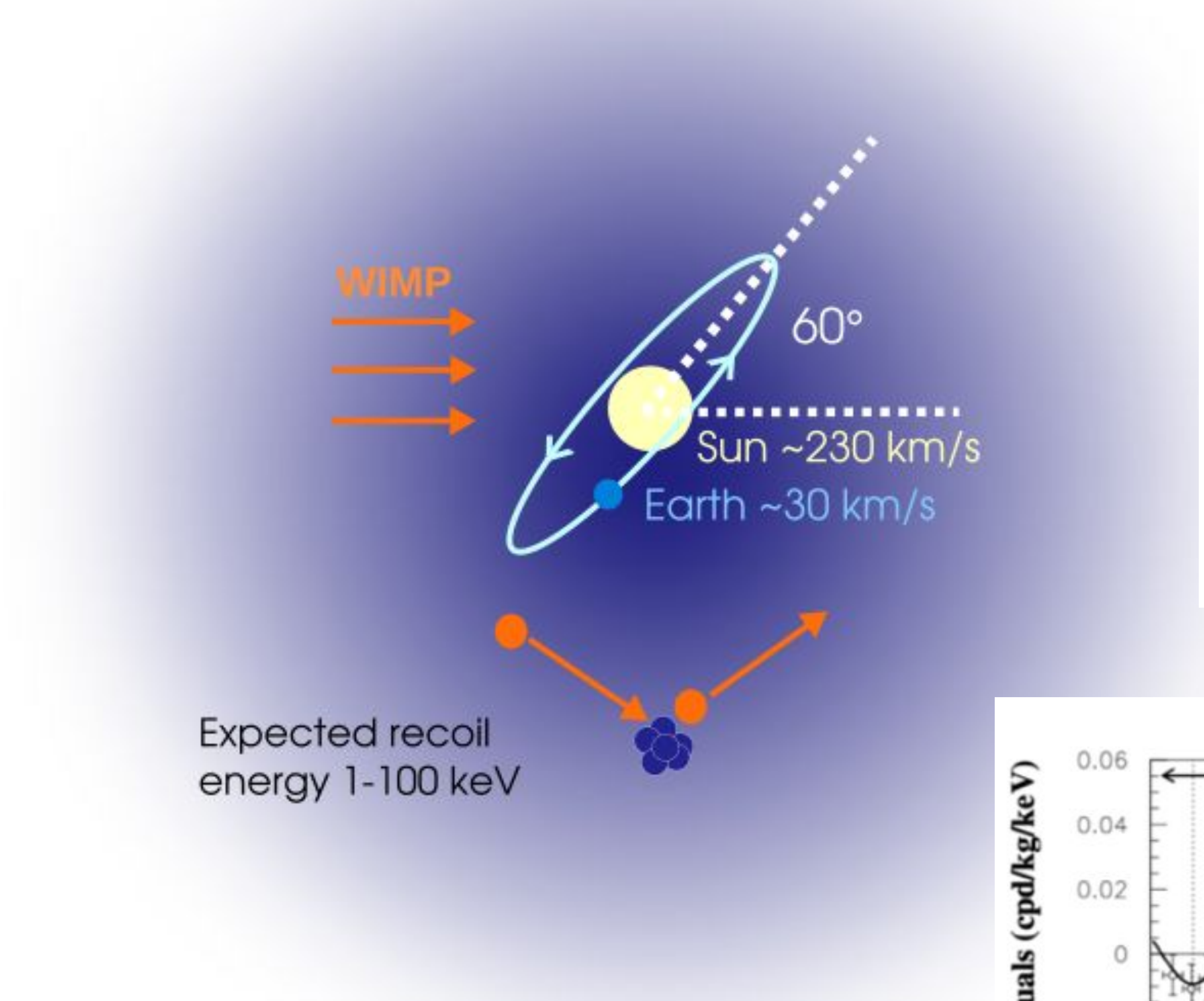
New programs are under development: COSINE-100+, COSINE-200, SABRE, COSINUS, and PICOLON.

R&D: ANAIS+, ASTAROTH.

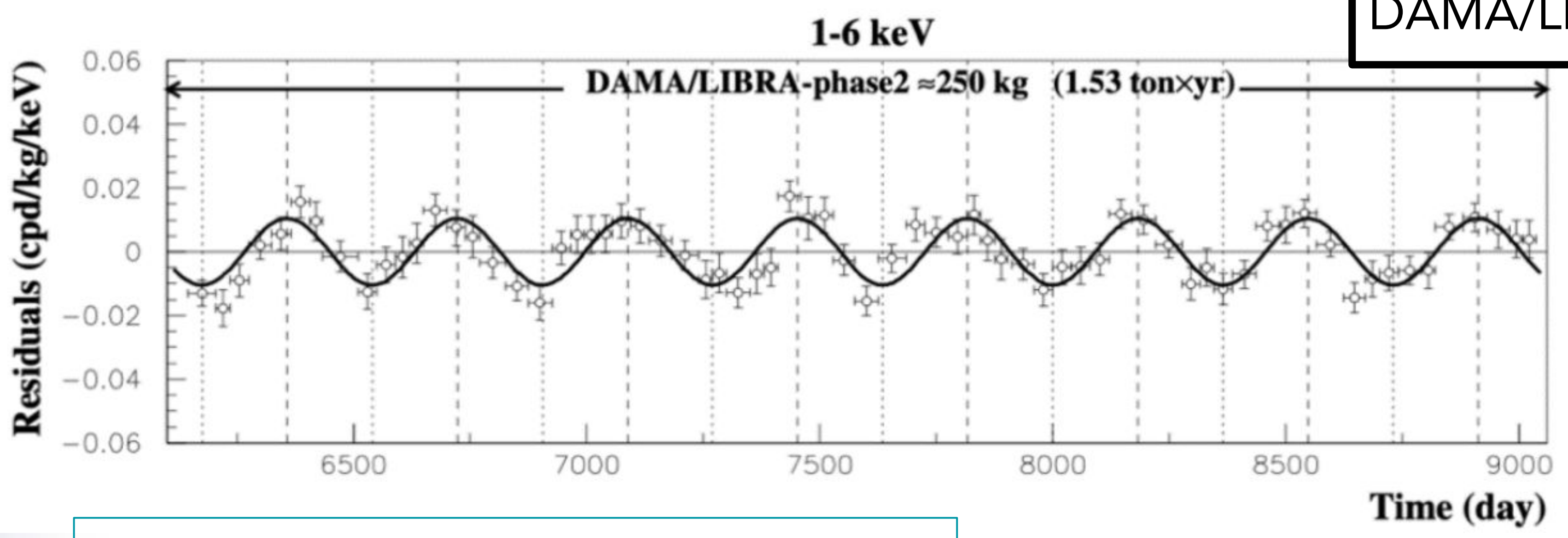
Dark Matter with annual modulation

$$R = S_0 + S_m \cos(\frac{2\pi}{T}(t - t_0))$$

- Expected rate in an Earth-based detector is modulated
- Small modulation fraction S_m/S_0 = O(~few %) on a background at the level of 1 cpd/kg/keV (1 dru)
- Region of interest [1-6] keV

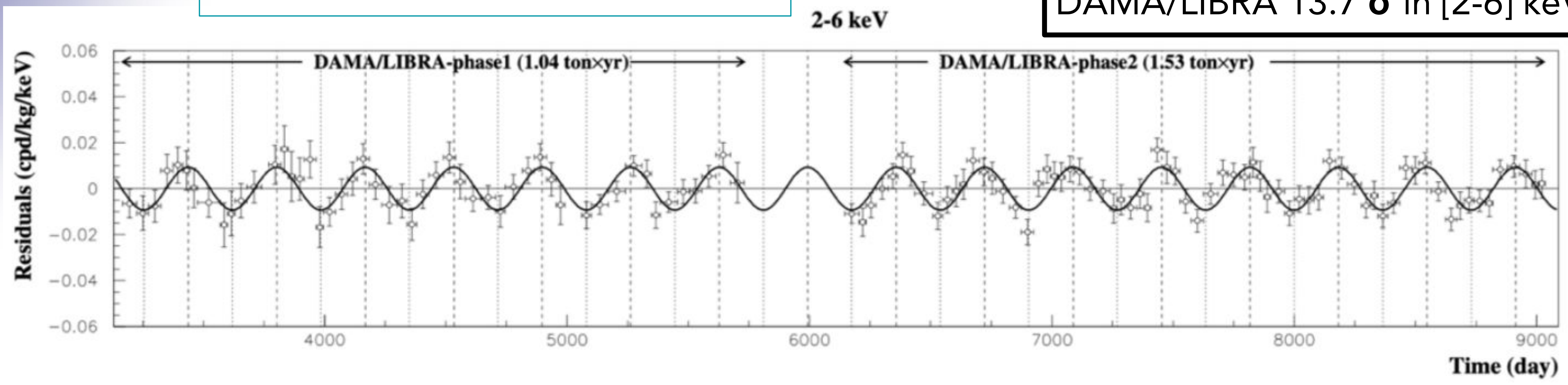


DAMA/LIBRA 11.6 **σ** in [1-6] keV



[Nucl. Phys. At. Energy 22 (2021) 329-342]

DAMA/LIBRA 13.7 **σ** in [2-6] keV



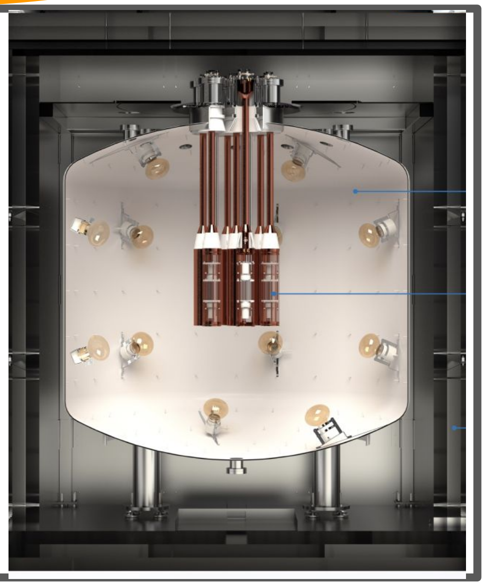
The modulation observed by DAMA/LIBRA satisfies the criteria expected of a DM-induced signal.

SABRE: Sodium-iodide with Active Background REjection

The goal of the SABRE experiment is to search for dark matter through **annual modulation** signature with **higher sensitivity** (= lower background) w.r.t. DAMA and other NaI(Tl) based experiments.

Ultra radio-pure crystals

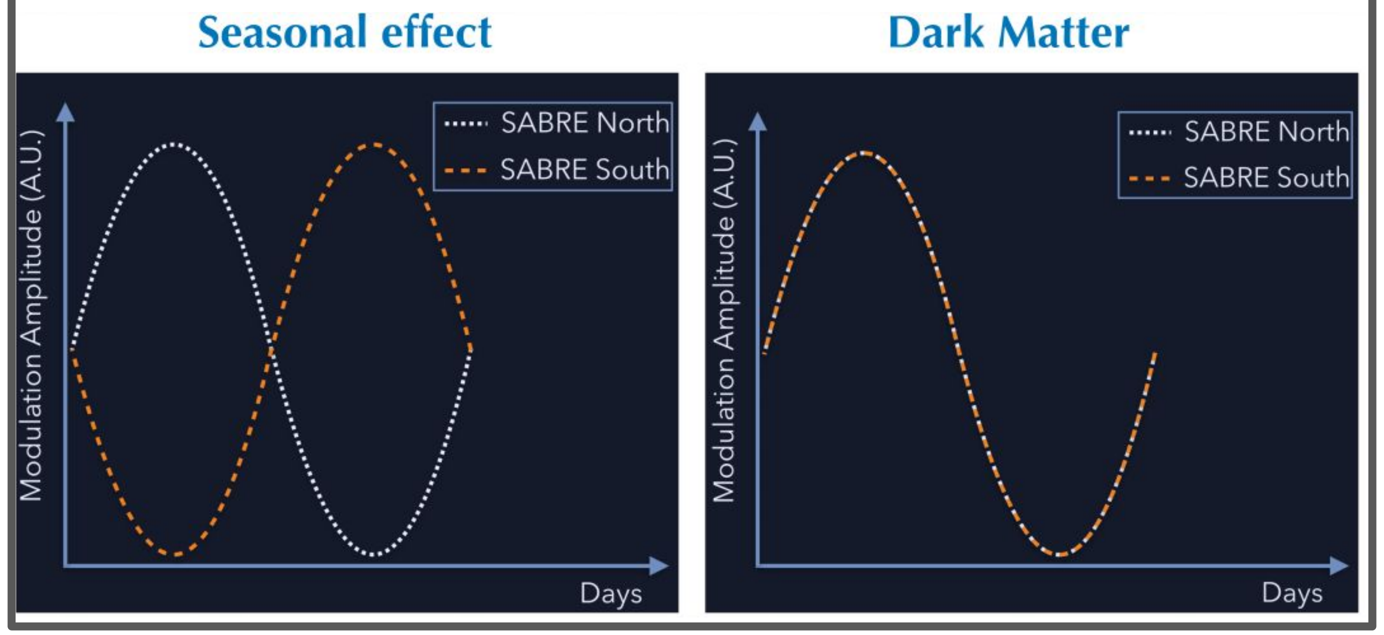
Active veto with liquid scintillator



@ SABRE South only

Double location:

Northern and Southern hemisphere

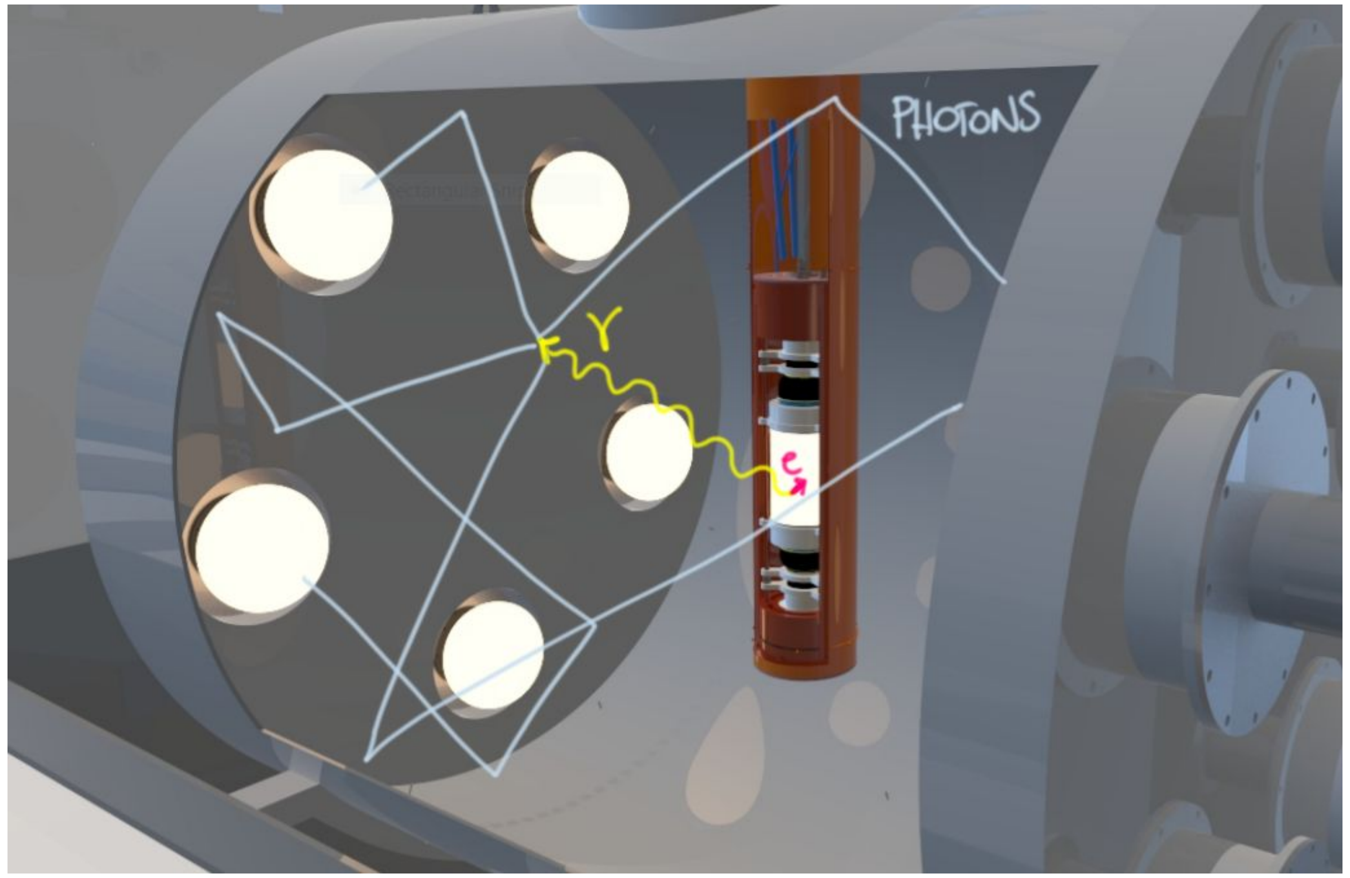


Internal backgrounds in NaI(Tl) crystals

Main contributions in ROI:

- ^{238}U, ^{232}Th
  - must be at the level of 10 ppt
- ^{40}K
  - must be at the level of a few ppb
- ^{87}Rb, ^{210}Pb, ^{3}H

no longer an option for SABRE North, due to the phase out of organic scintillators at LNGS



Internal contaminations:

Our initial effort focused on the reduction of Potassium content (clean powder, active veto).

Cosmogenic activation

Cosmogenic activation in the ROI mainly comes from ^{3}H, ^{113}Sn, ^{109}Cd, ^{22}Na.
Minimum order of 1 yr underground cooling from cosmogenic activity required (or underground growth).

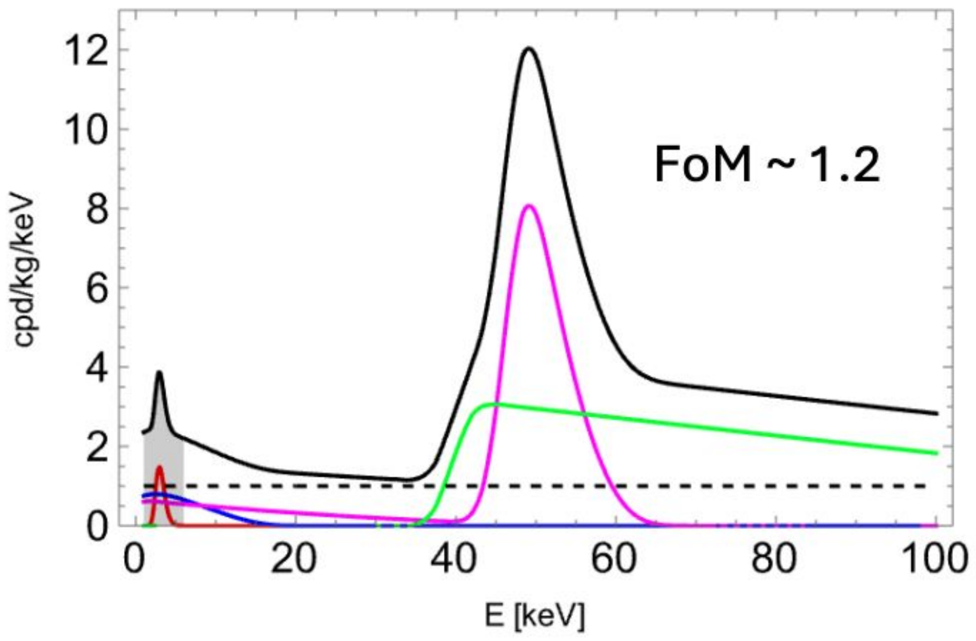
Internal backgrounds in NaI(Tl) crystals

The case of ^{210}Pb:

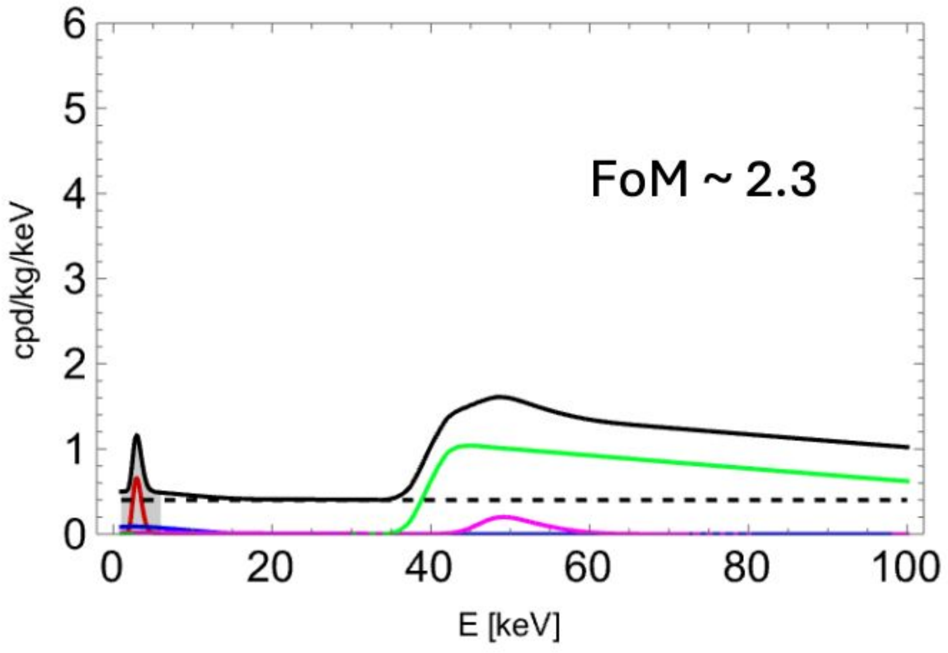
- it can originate from the crystal bulk or it can be implanted on the surface from the ^{222}Rn decay chain
- it can be present in the reflector around the crystal
- the contribution to the background in the ROI depends on the depth distribution

The case of ^{3}H:

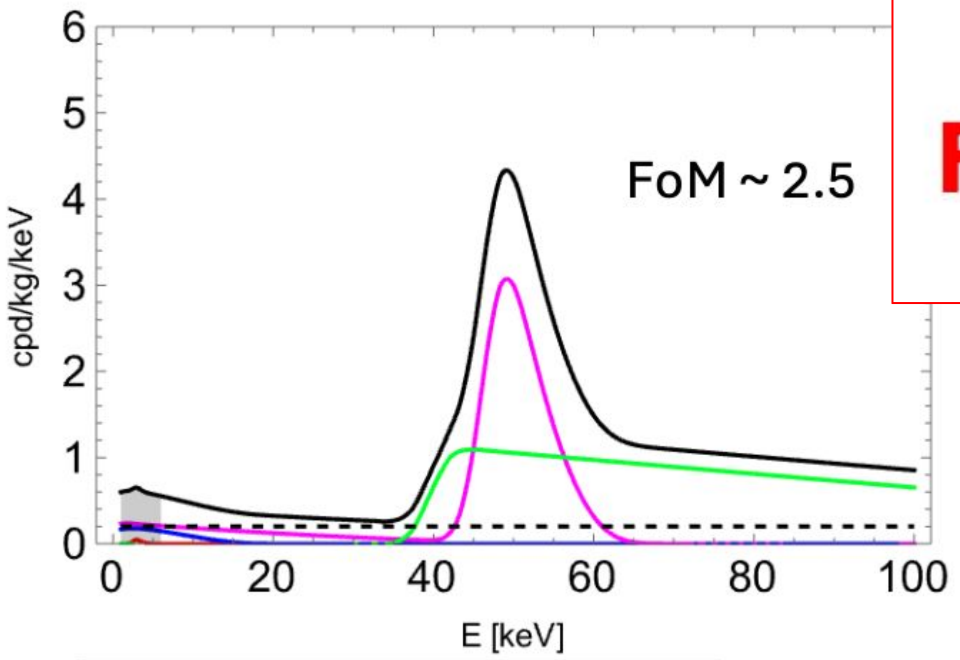
- relevant background source in the ROI
  - pure beta emitter with Q = 18.591 keV and $T_{1/2}$ ~ 12 years
- its activity in the crystal depends on the exposure on surface



^{210}Pb ~ 20% (1 mBq/kg)
K ~ 15% (32 ppb)
^{3}H ~ 28% (90 μBq/kg)



^{210}Pb ~ 4% (26 μBq /kg)
K ~ 26% (14 ppb)
^{3}H ~ 12% (10 μBq/kg)



^{210}Pb ~ 37% (0.4 mBq/kg)
K ~ 2% (1 ppb)
^{3}H ~ 28% (20 μBq/kg)

$$\text{FoM} = \frac{S_m}{\sqrt{2}} \sqrt{\frac{M\,t}{S_0 + B}}$$

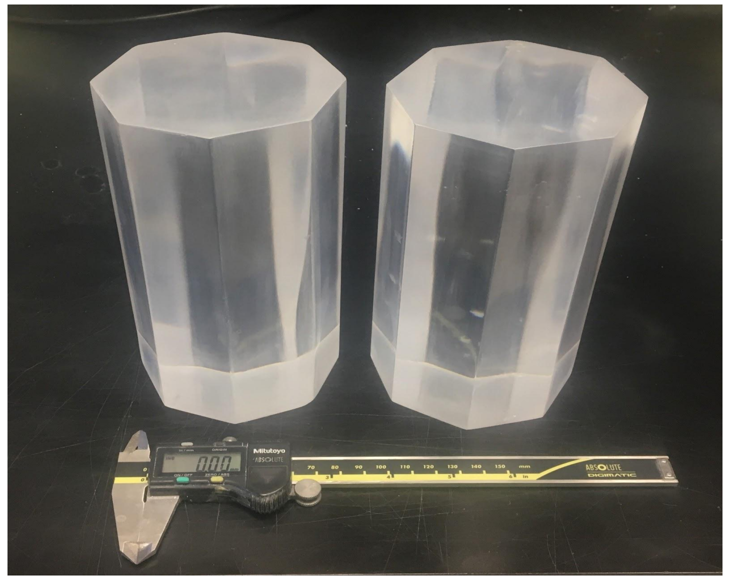
assume an exposure of 1000 kg x yr and Sm = 0.01 dru

credit: A. Ianni - IDM 2024

How to grow a radiopure NaI(Tl) crystal

Clean precursors: NaI powder, Tl dopant → Powder drying → Doping → Clean growth → Cutting & polishing



The collaboration between Princeton University and industrial partners led to:

- clean NaI powder Astrograde by Sigma Aldrich now Merck, Germany
- clean crystals grown by RMD - Radiation Monitoring Devices, MA (USA)
  - Vertical Bridgman technique: the powder is placed inside a sealed ampoule, reducing the possibility of contamination during the growth phase

Radiopurity of NaI powder

	DAMA-NaI (Saint Gobain)	DAMA/LIBRA (Saint Gobain)	SABRE - Astrograde (Merck)
^{238}U	0.56±0.04 ppb	0.02 ppb	<0.07 ppb
^{232}Th	0.21±0.01 ppb	0.02 ppb	<0.08 ppb
^{nat}K	<4.8 ppm	<0.1 ppm	~3-10 ppb
^{85}Rb			< 0.4 ppb
^{208}Pb			~1 ppb

Astrograde vs DAMA

For Dama-NaI
https://link.springer.com/article/10.1007/BF03035868

For DAMA/LIBRA:
https://doi.org/10.1016/j.nima.2008.04.082

SABRE CRYSTALS

Astrograde vs itself

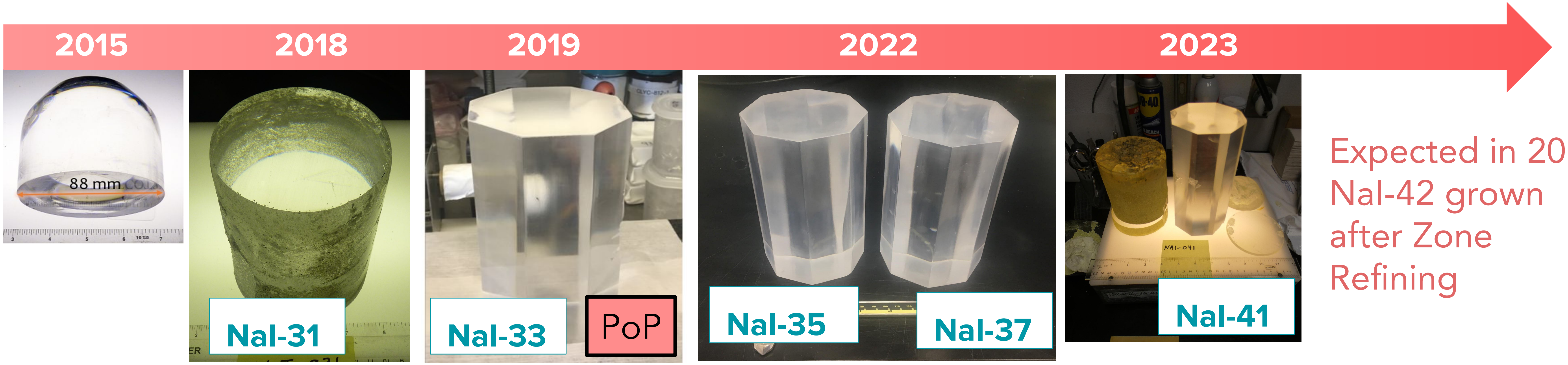
variability in the powder batch

	Powder Astro Grade batch	Mass [kg]	LY [phe/keV]	^{39}K [ppb] powder	^{39}K [ppb] crystal	^{210}Pb [mBq/kg]	Rate ROI [dru]	^{238}U [ppt]	^{232}Th [ppt]
NaI-31	MKBW4911V	3.0	9	8.0	18.5±0.7 14.6±3.0 (PoP)	1.02±0.07	2.74±0.03	–	–
NaI-33	MKCC0371	3.4	11	4.3	4.4±0.6 2.1±1.4 (PoP)	0.51±0.02	0.95±0.05	0.47±0.05	0.40±0.07
NaI-35	MKCC0371	4.36	9	4.3	8.2±0.6	0.53±0.01	1.26±0.03	0.18±0.03	–
NaI-37	113065	4.35	7.8	17.7	8.0±0.6	0.79±0.01	2.57±0.05	0.61±0.05	0.27±0.06
NaI-41*	76650	4.27	10	6.7	5.7±0.9	0.60±0.02	1.8±0.4	0.48±0.05	0.39±0.07

*NaI-41 still affected by cosmogenics

SABRE crystals R&D

R&D carried out by PU, INFN and ARC Centre of Excellence for DM
Clean NaI powder Astrograde by Sigma Aldrich now Merck, Germany
Crystals grown by RMD - Radiation Monitoring Devices, MA (USA)

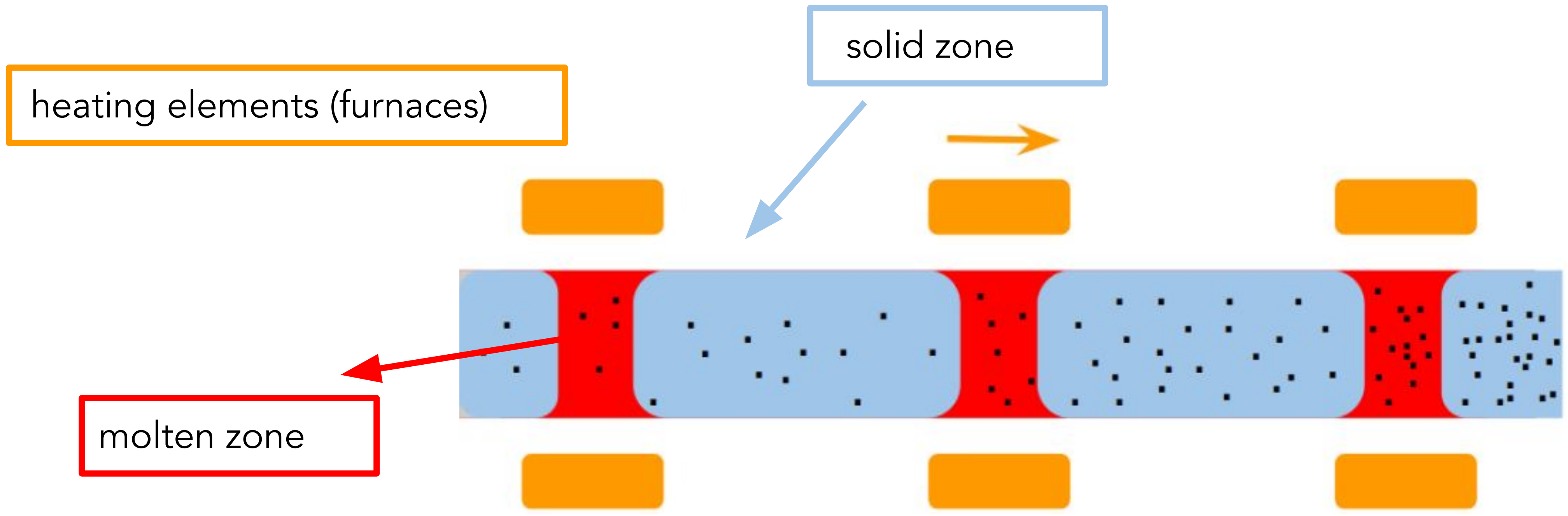


Expected in 2024:
NaI-42 grown
after Zone
Refining

- NaI-33: Background ~1 cpd/kg/keV → at the level of DAMA/LIBRA Phase 1
- NaI-35, NaI-37: Reproducibility of clean growth within factor 2
- NaI-41: Zone Refining R&D activity (see next slides)

Zone refining

- Technique successfully used in semiconductor industry.
- A narrow region of an ingot is melted, and the molten zone is moved along the crystal's axis.
  - The segregation coefficient of an impurity (ratio of the concentration in the solid phase with respect to the liquid phase) is usually less than one.
  - Impurities will diffuse into the liquid phase and will be segregated to one side of the ingot by the moving ovens.
- The process can be repeated until the desired level of purity is met.
- The refined crystal is obtained discarding the end part along with all the impurities.



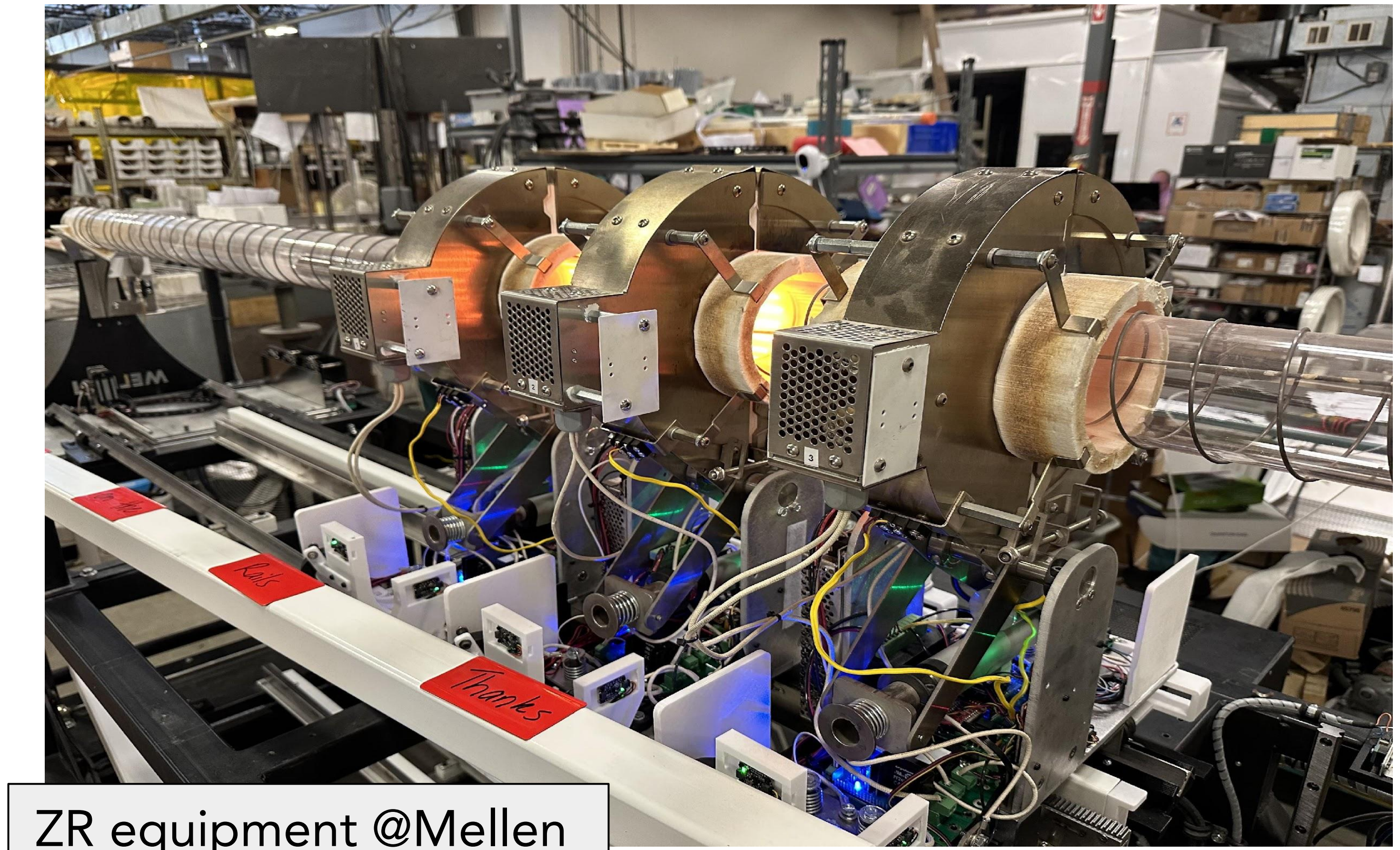
This same technique can be applied directly to an ampoule containing pre-melted NaI powder, to obtain a purified ingot out of which the final NaI(Tl) crystal will be later grown.

Zone refining

Tested on NaI Astro grade powder

Isotope	Impurity concentration (ppb)					
	Powder	S_1	S_2	S_3	S_4	S_5
^{39}K	7.5	< 0.8	< 0.8	1	16	460
^{85}Rb	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	0.7
^{208}Pb	1.0	0.4	0.4	< 0.4	0.5	0.5
^{24}Mg	14	10	8	6	7	140
^{133}Cs	44	0.3	0.2	0.5	3.3	760
^{138}Ba	9	0.1	0.2	1.4	19	330

Phys. Rev. Applied 16, 014060 (2021)



ZR equipment @Mellen

Refurbished and upgraded in 2023.

A zone refiner with three furnaces was designed by the SABRE Princeton group in collaboration with our industrial partner MELLEN, NH USA.

- Unique instrument: large enough for the production of kg size crystals.
- Continuous mode of operation:
  - The three annular furnaces move simultaneously on a motorised track along the ingot and perform multiple zone-refining steps (passes).
- Adjustable speed and number of passes.

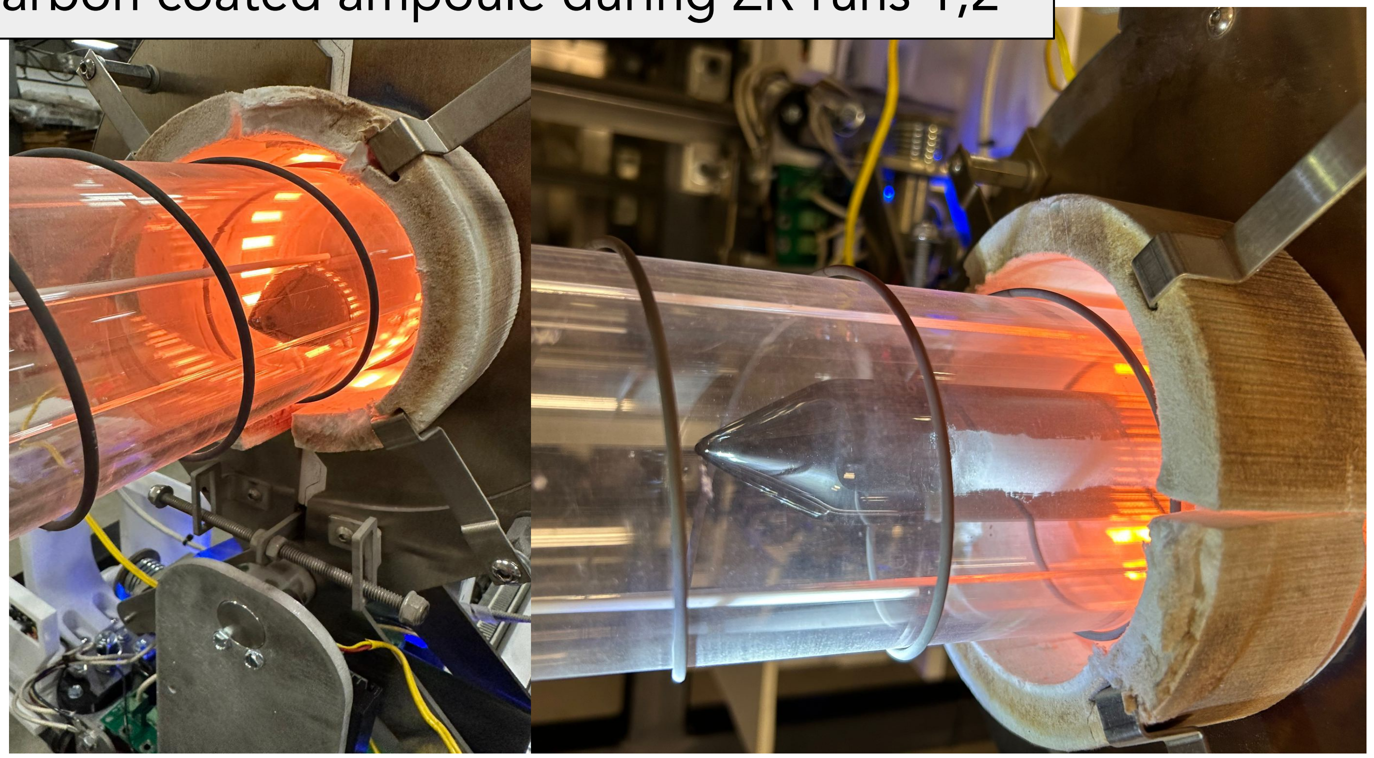
- PRIN 2022: Ultra purification of NaI with Zone Refining for dark matter detection - PI: Claudia Tomei
- PRIN 2022 PNRR: PUNTO (ProdUction of ultra-pure NaI detecTOrs by zone refining) - PI: Aldo Ianni

Zone refining (2023 - 2024)

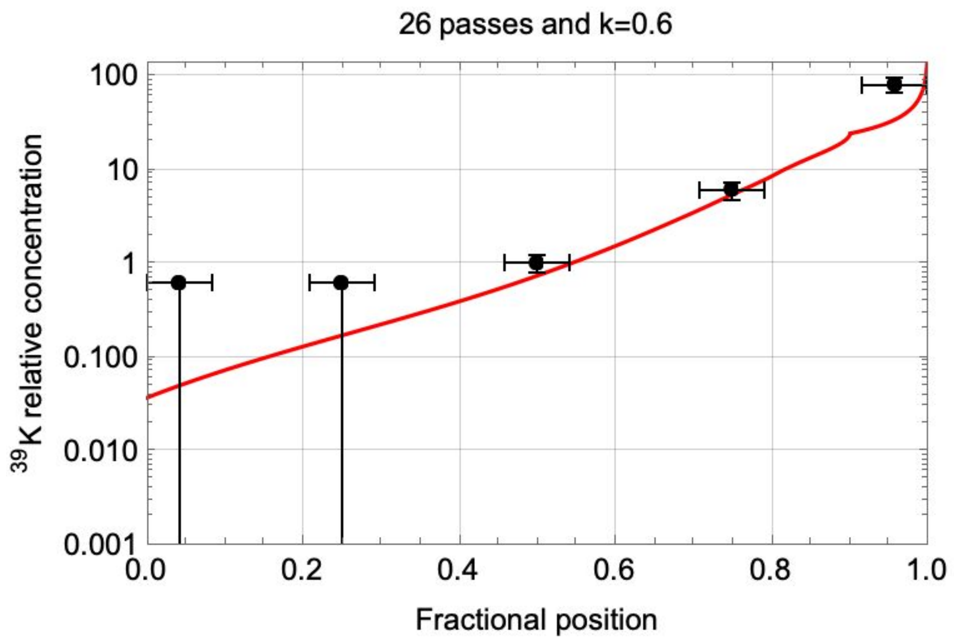
Four runs with 900 gr of Astro Grade NaI powder have been performed at MELLEN, NH, USA to determine efficiency and running conditions.

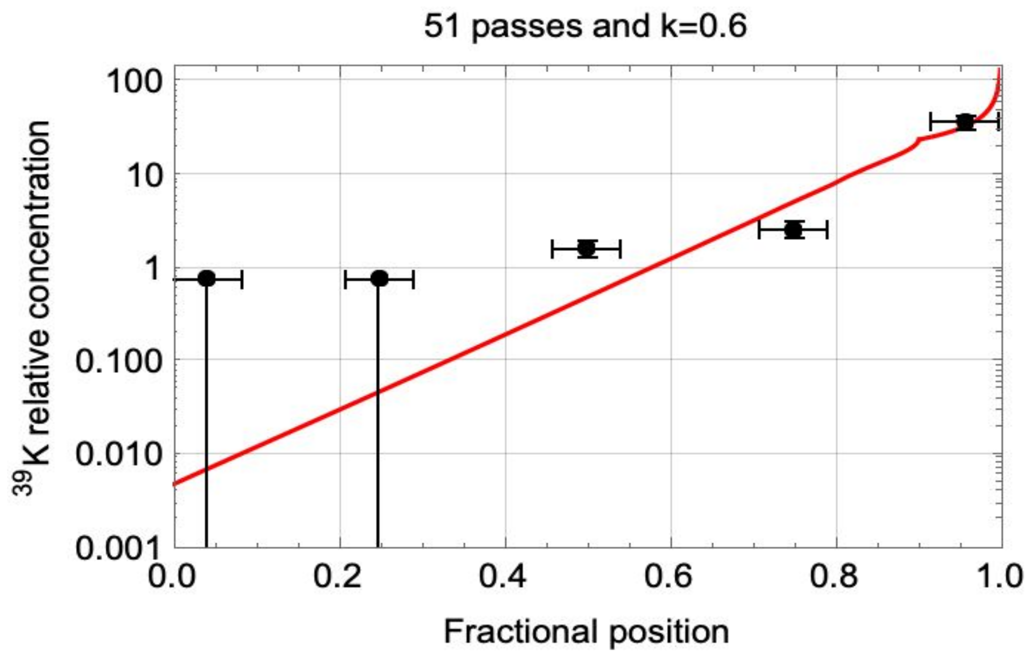
- RUN1: Carbon coated ampoule
- RUN2: Carbon coated ampoule with increased number of passes

Carbon coated ampoule during ZR runs 1,2



Comparison with the model





For each run we took 5 samples from the ingot and shipped them to Canfranc Laboratory and Seastar for ICPMS measurements.

- 3±1 cm from tip
- 15±1 cm from tip
- 30±1 cm from tip
- 15±1 cm from tail
- 3±1 cm from tail

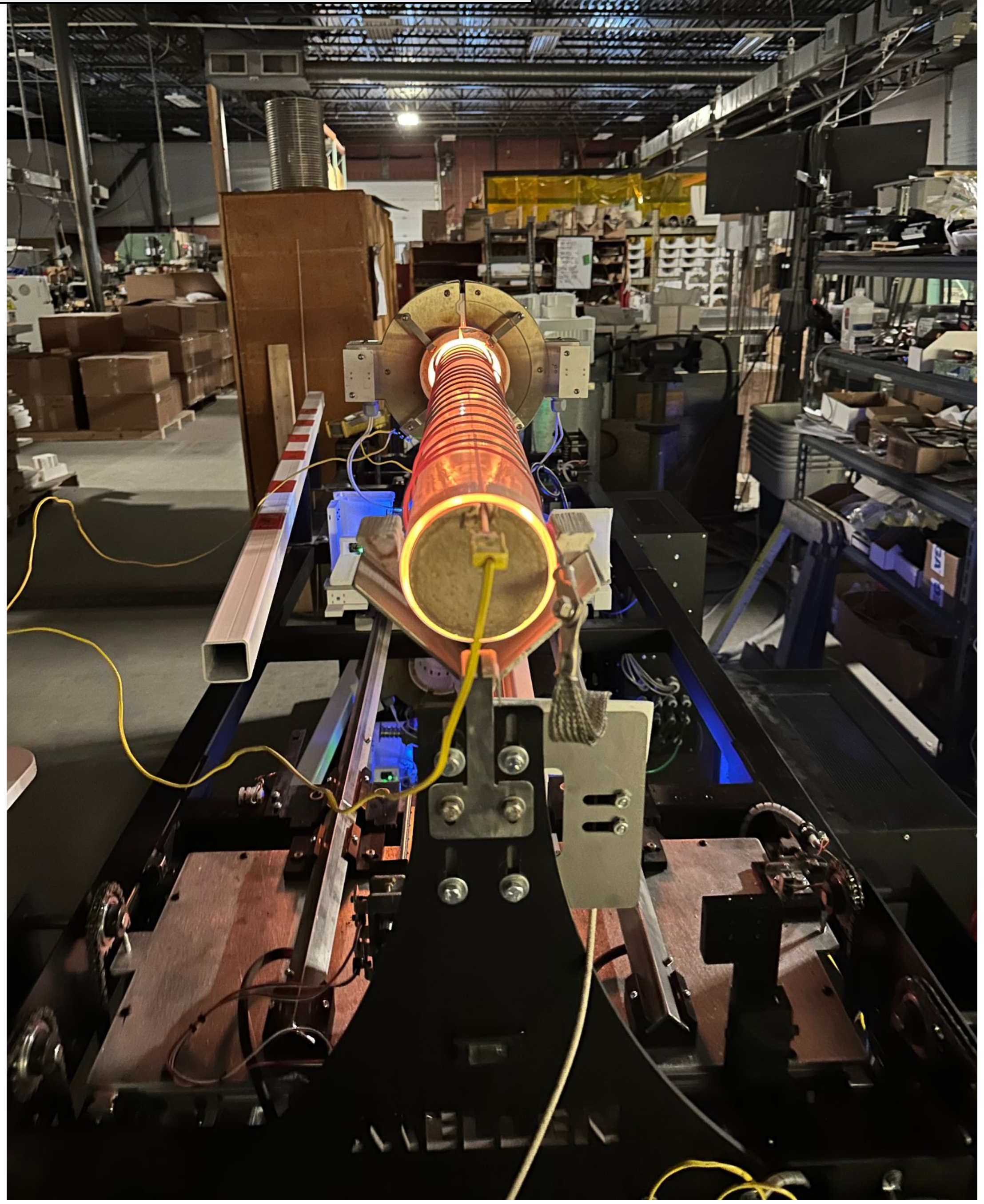
These preliminary results confirm our prediction that going from 26 to 51 passes does not significantly improve the average purification.

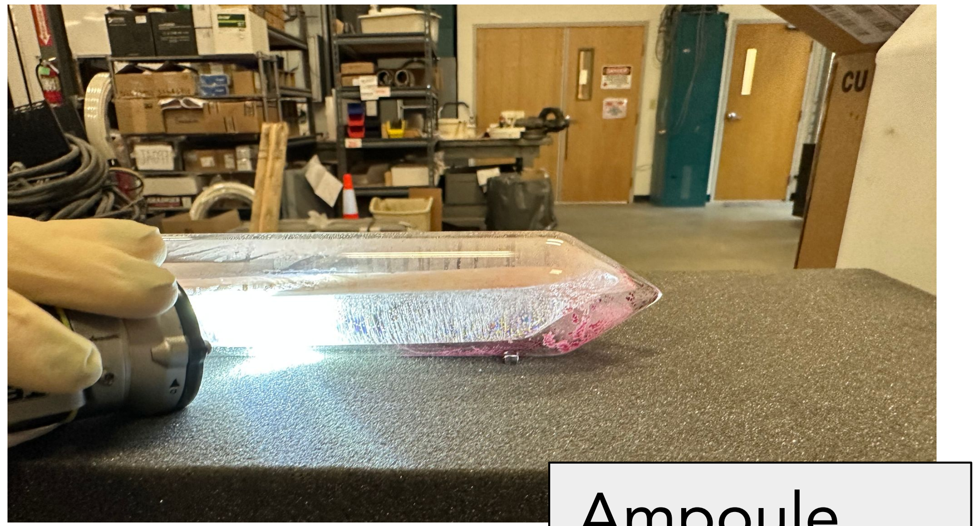
Zone refining (2023 - 2024)

Four runs with 900 gr of Astro Grade NaI powder have been performed at MELLEN, NH, USA

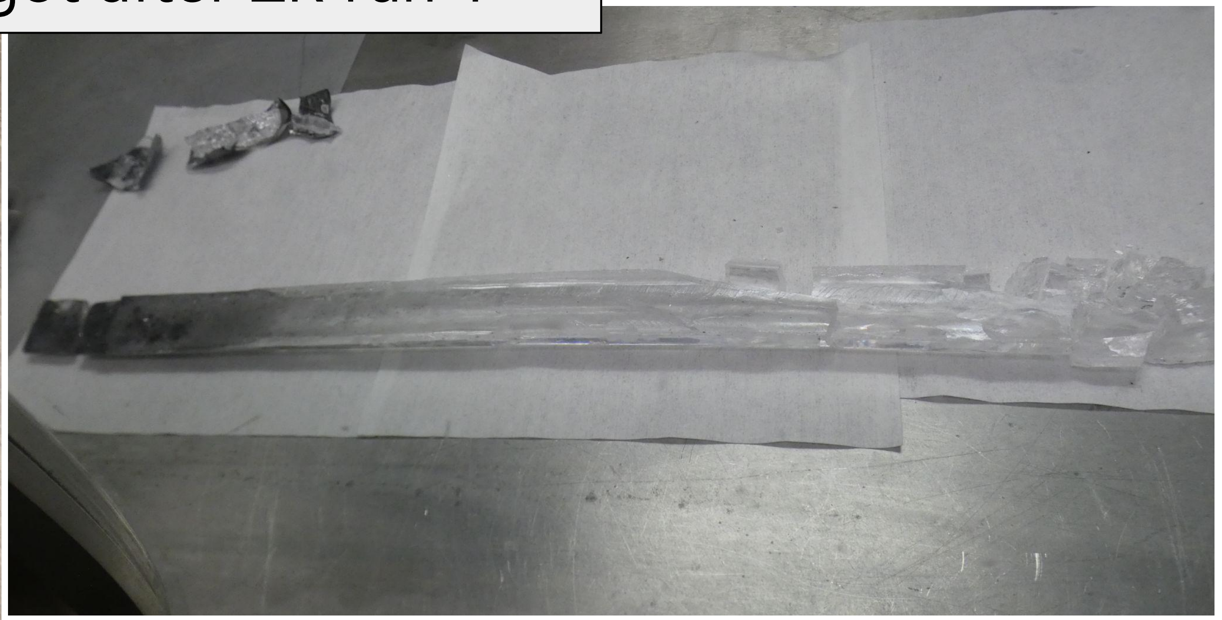
- RUN3: No coating + use of $SiCl_4$ to avoid sticking
- RUN4: No coating + use of $SiCl_4$
  - Ampoule sealed without gas inside
  - Selected option for SABRE

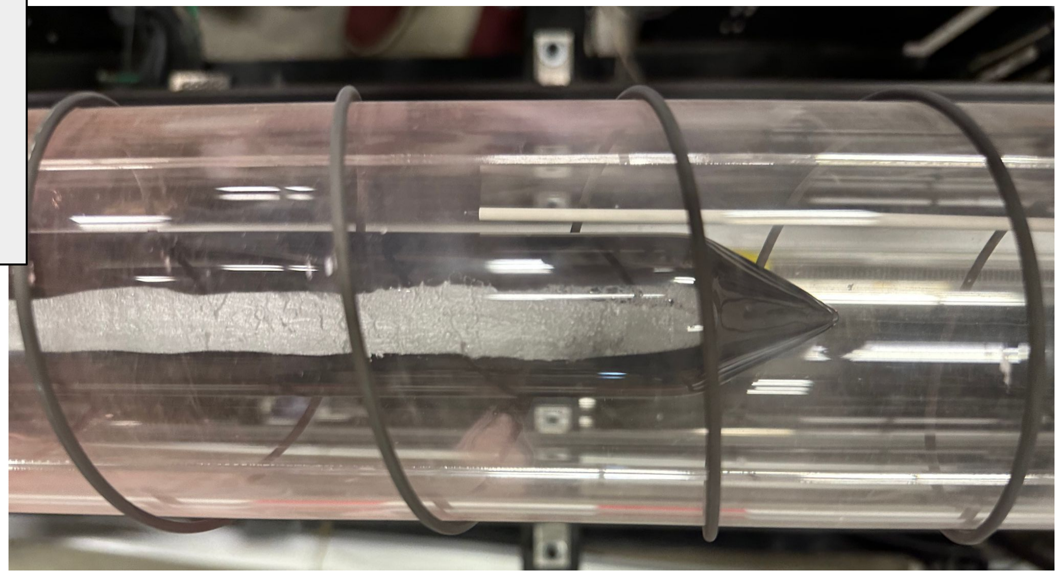
ZR equipment @Mellen





Ingot after ZR run 1





Ampoule treated with $SiCl_4$ during and after ZR runs 3,4

Zone refining preliminary results

Sample Run4	^{39}K [ppb]	^{65}Cu [ppb]	^{85}Rb [ppb]	^{133}Cs [ppb]	^{138}Ba [ppb]	^{208}Pb [ppb]
powder	7	5	<0.2	1	3.6	1.1
Zone 1	<4	<4	<0.8	<0.3	<0.3	2.0±0.3
Zone 2	<4	<4	<0.8	<0.3	1.2±0.3	1.6±0.2
Zone 3	10.1±0.6	<4	<0.8	<0.3	2.7±0.2	1.6±0.3
Zone 4	21.5±0.7	<4	<0.8	1.1±0.1	8.1±0.5	1.9±0.3
Zone 5	68±2	10±1	<0.8	203±6	17±0.9	1.2±0.3

Clear segregation visible for Potassium and Rubidium (both contributing to background in the DM ROI).

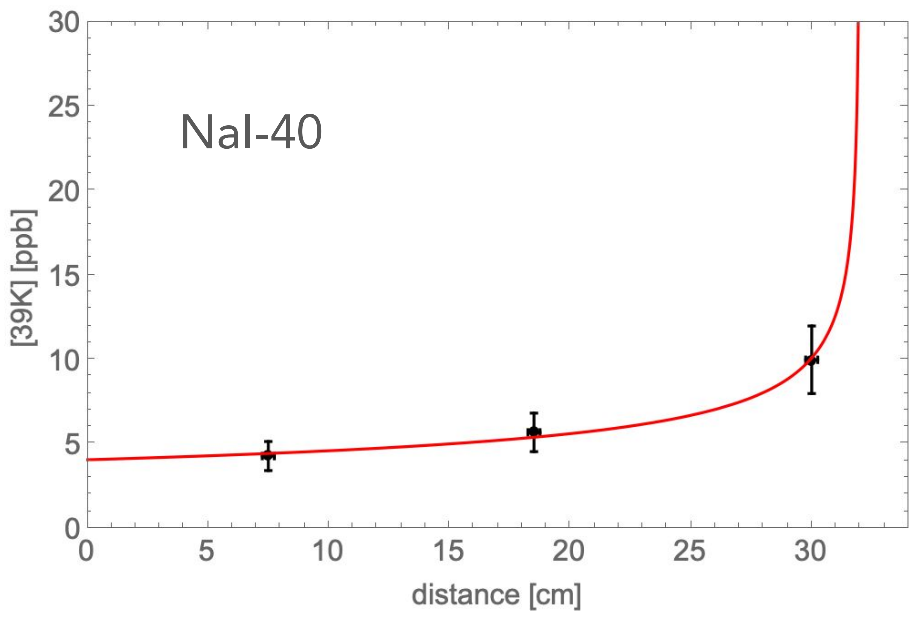
The results on Pb may seem to indicate a contamination during the handling of the material.

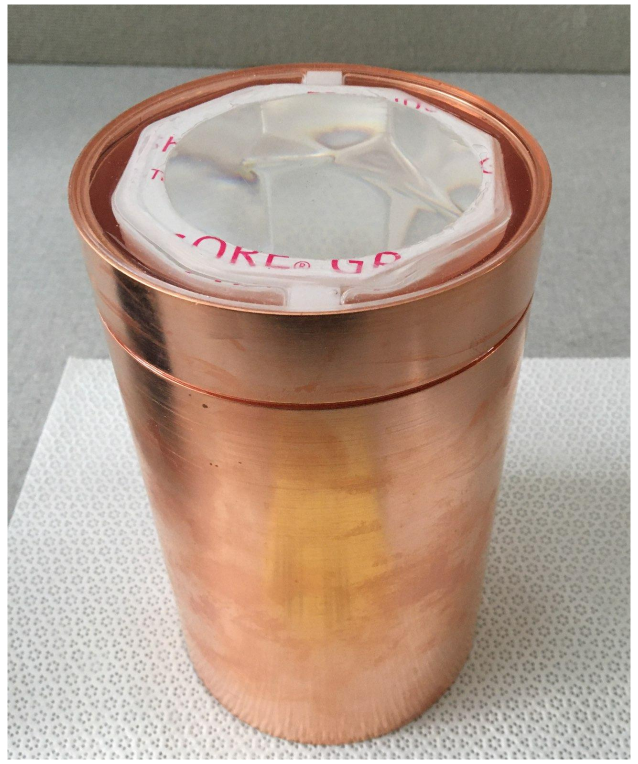
Credits to Laura Cid-Barrio and the Canfranc ICP-MS lab for the extensive measurement campaign!.

NaI-41 and growth from chunks

- ZR as a purification method for the powder: subsequent crystal growth done from chunks of the purified ingot.
- Successful growth from chunks of an optically sound crystal with good scintillation properties had to be tested.
- Crystal NaI-41 was grown @ RMD starting from chunks of a previous undoped crystal, named NaI-40.
- Growth of NaI-41 started on Sept 2023: encapsulated at RMD for easier and faster characterization at LNGS.
- NaI-41 shipped to LNGS by plane soon after production. Underground at LNGS since 15 Dec 2023.

	^{39}K [ppb]	^{65}Cu [ppb]	^{85}Rb [ppb]	^{138}Ba [ppb]	^{133}Cs [ppb]	^{208}Tl [ppm]
Tip	<4	<5	<0.8	<0.2	<0.3	520±4
Tail	13.4±0.3	<5	<0.8	1.1±0.2	1.7±0.1	1343±10
Far Tail	20.2±0.4	10.8±0.8	<0.8	2.8±0.3	4.2±0.3	1607±12





Average potassium contamination in the body of crystal NaI-40: 5.8±0.7 ppb

Compatible with that of NaI-41: the growth process does not introduce potassium contamination.
Successful growth of crystal NaI-41 from chunks and its good optical properties marks a critical step in our strategy to grow highly radiopure crystals.

NaI-42: upcoming full test of the ZR-based growth of SABRE crystals

NaI-42 will be the first crystal entirely grown out of ZR NaI powder (Astrograde).

It will be produced from 12 kg of ZR powder in 4 runs of 3 kg each.

Zone refining will smear out differences in powder batches purity.

Zone refining started @Mellen, crystal expected by the end of 2024.

Zone refining at MELLEN, ampoule loading and pre-melting handled at RMD.
Crystal growth by RMD (2 ovens dedicated to SABRE).

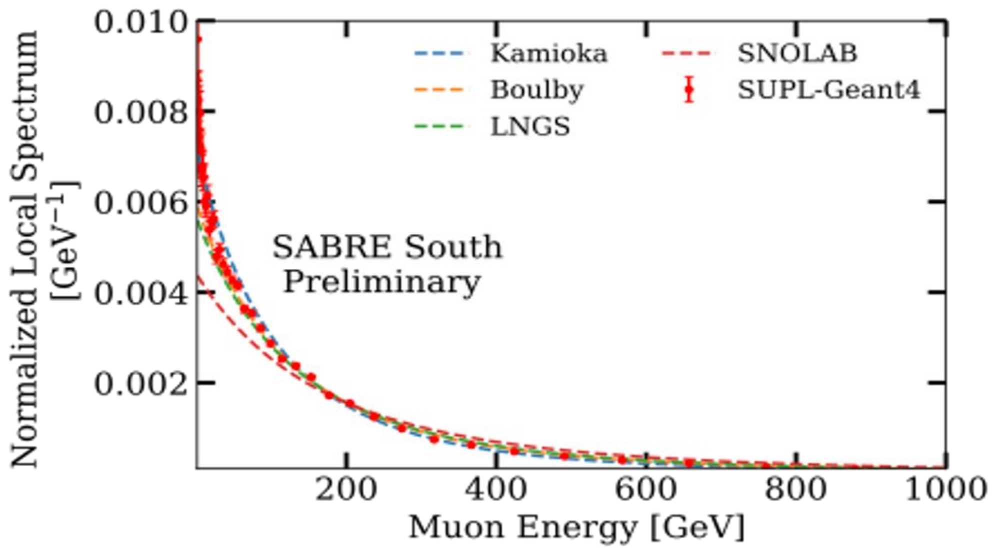
Glove box @ RMD
recently upgraded with recirculation system, dehumidifier, and sensors (T, P)

SABRE North and South

- SABRE North at Laboratori Nazionali del Gran Sasso (LNGS) in Italy
- SABRE South at Stawell Underground Physics Laboratory (SUPL) in Australia

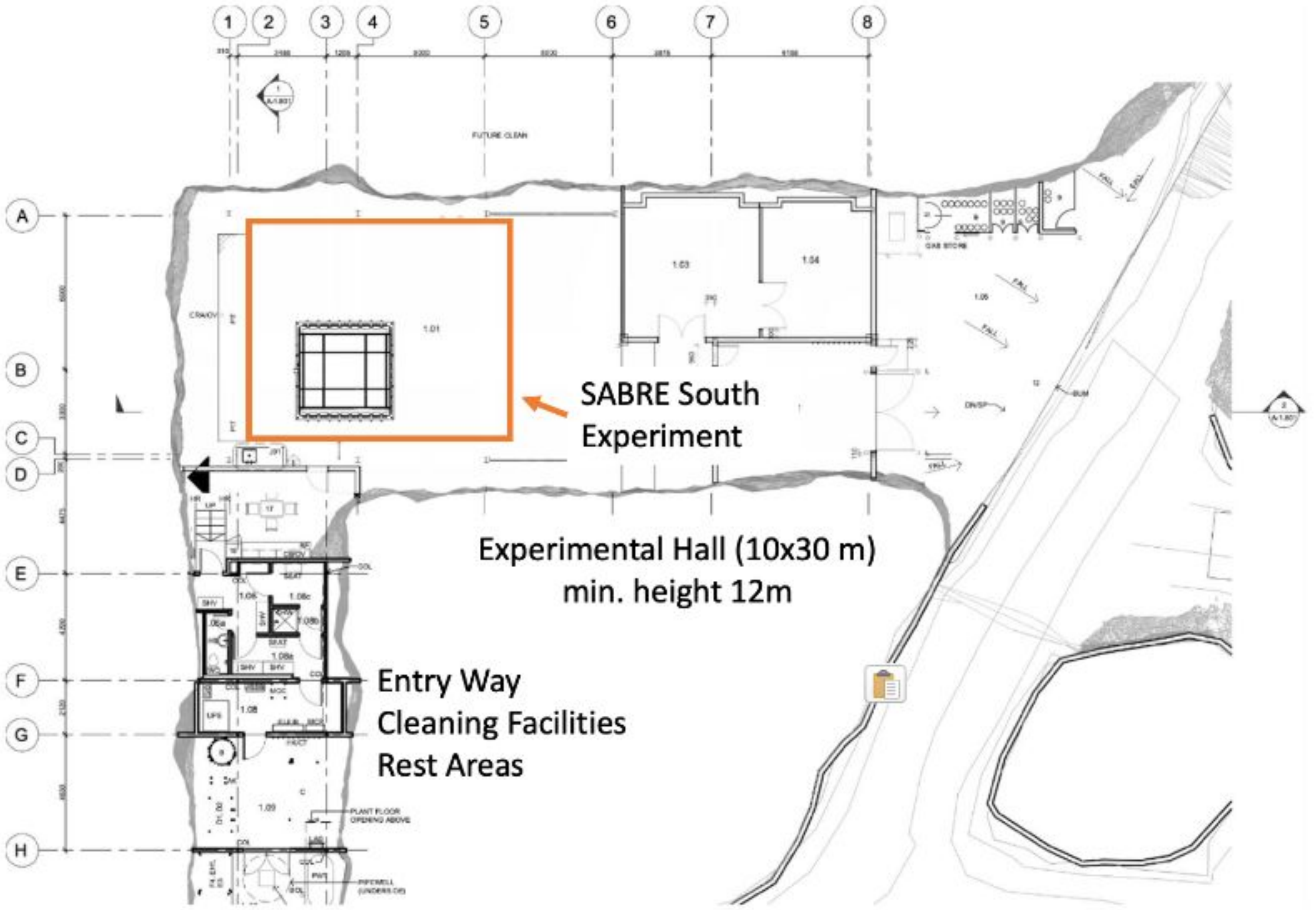
Stawell Underground Physics Lab (SUPL)

- First deep underground laboratory in the Southern Hemisphere, completed in 2022/2023:
  - 1025 m deep (2900 m water equivalent) with flat overburden, helical drive access.
- Located in the Stawell Gold Mine, 240 km west of Melbourne, Victoria, Australia.



Muon detectors have been **installed** for muon flux measurements at SUPL and currently collecting data.
The first detectors set-up at SUPL:

1. measure of muon flux and angular distributions;
2. provide the first test of the remote data acquisition system (DAQ) and processing pipelines.



https://www.supl.org.au

The SABRE strategy

- SABRE Proof-of-principle (PoP) and PoP-dry achieved a background of ~1 cpd/kg/keV

- Strategy to lower the background
  - For internal backgrounds

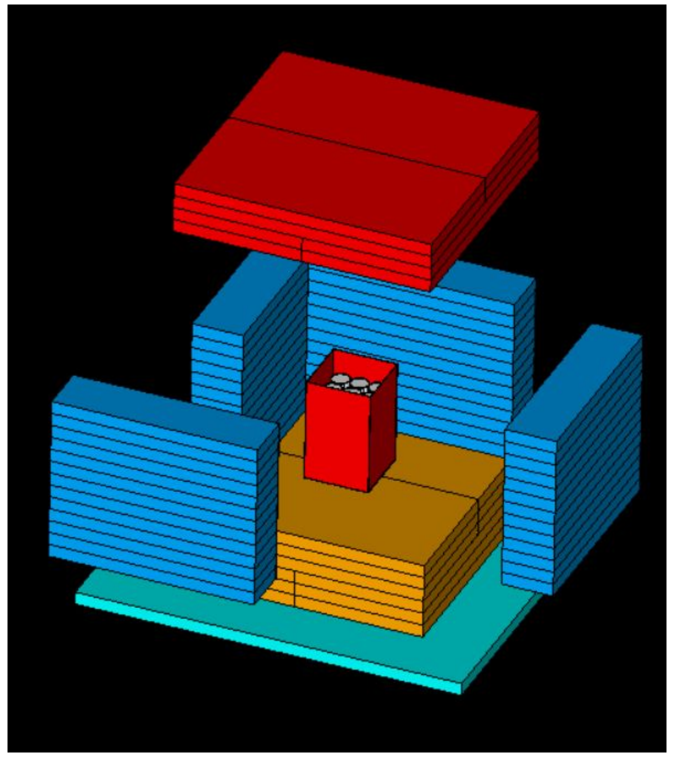
    → SABRE North & South: Zone Refining
  - For external background:

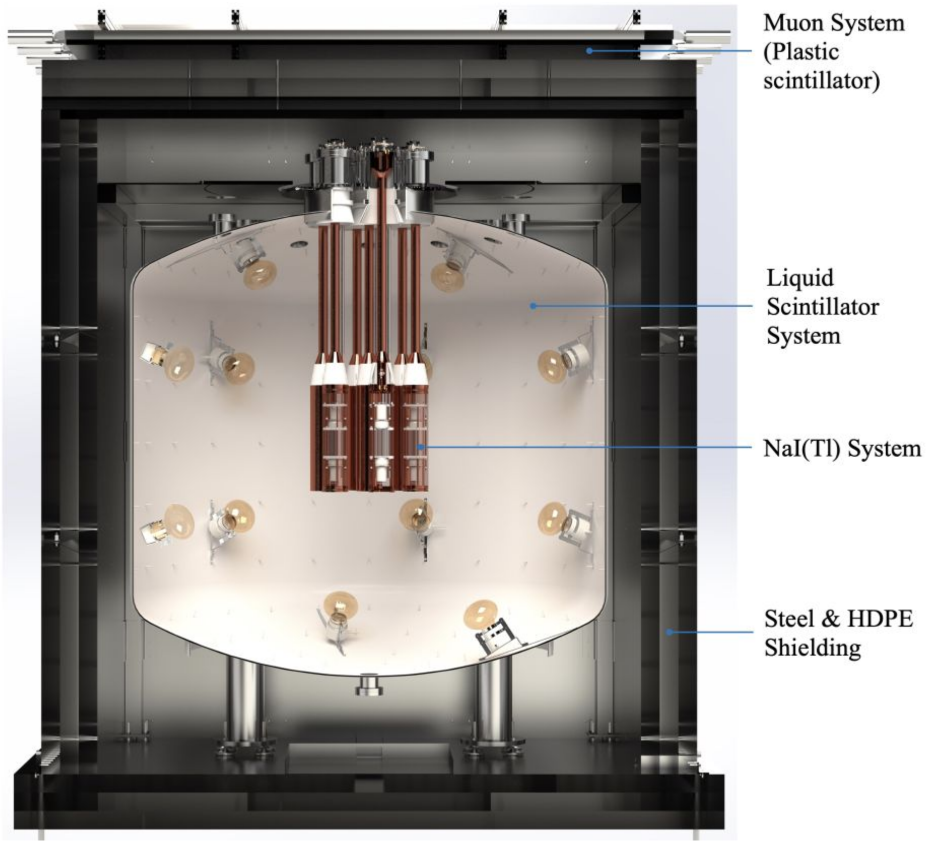
    → SABRE North: improve passive shielding

    → SABRE South: Liquid Scintillator (LAB)

    + Muon Veto

SABRE North





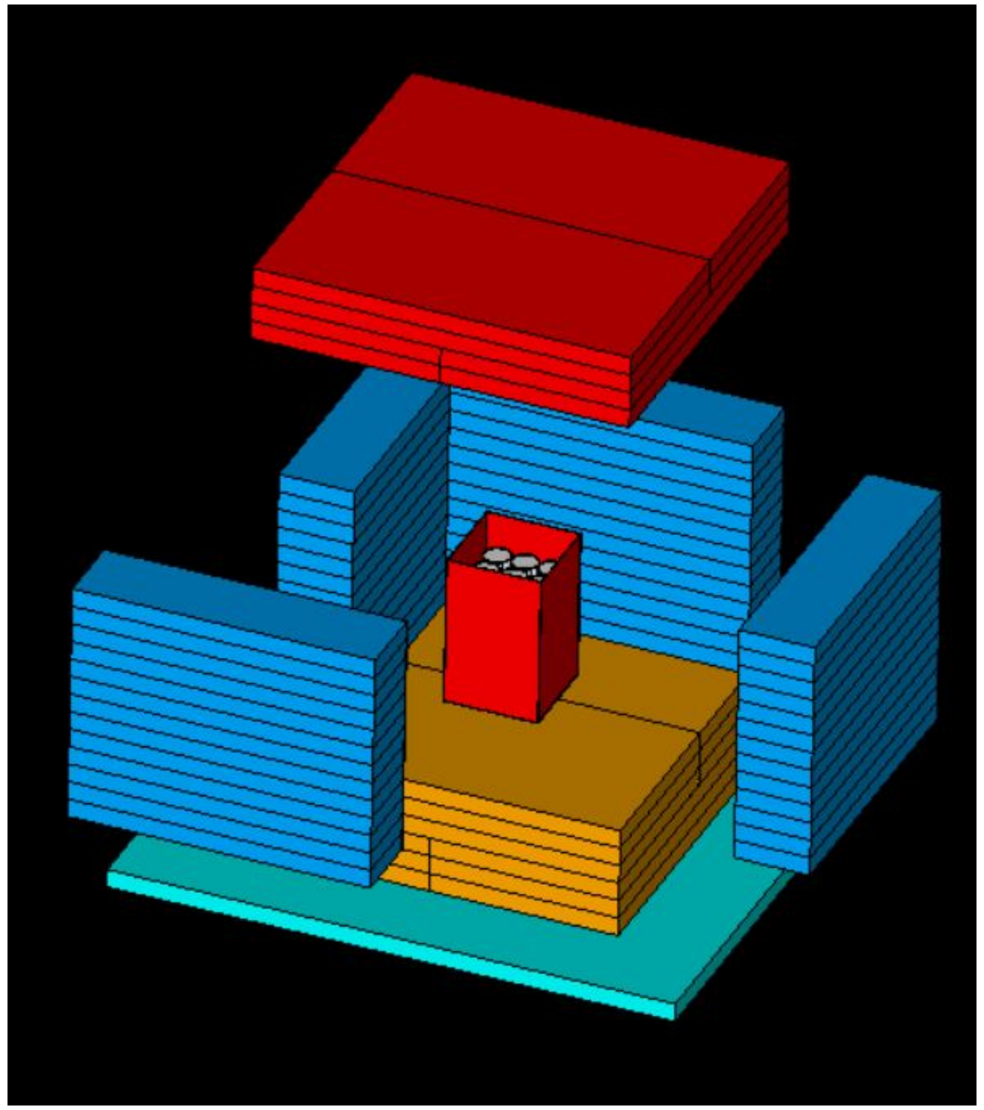
SABRE South

SABRE North status

- Conceptual design report presented in July 2021
- TDR presented in summer 2024
- TDR approved by INFN CSN2

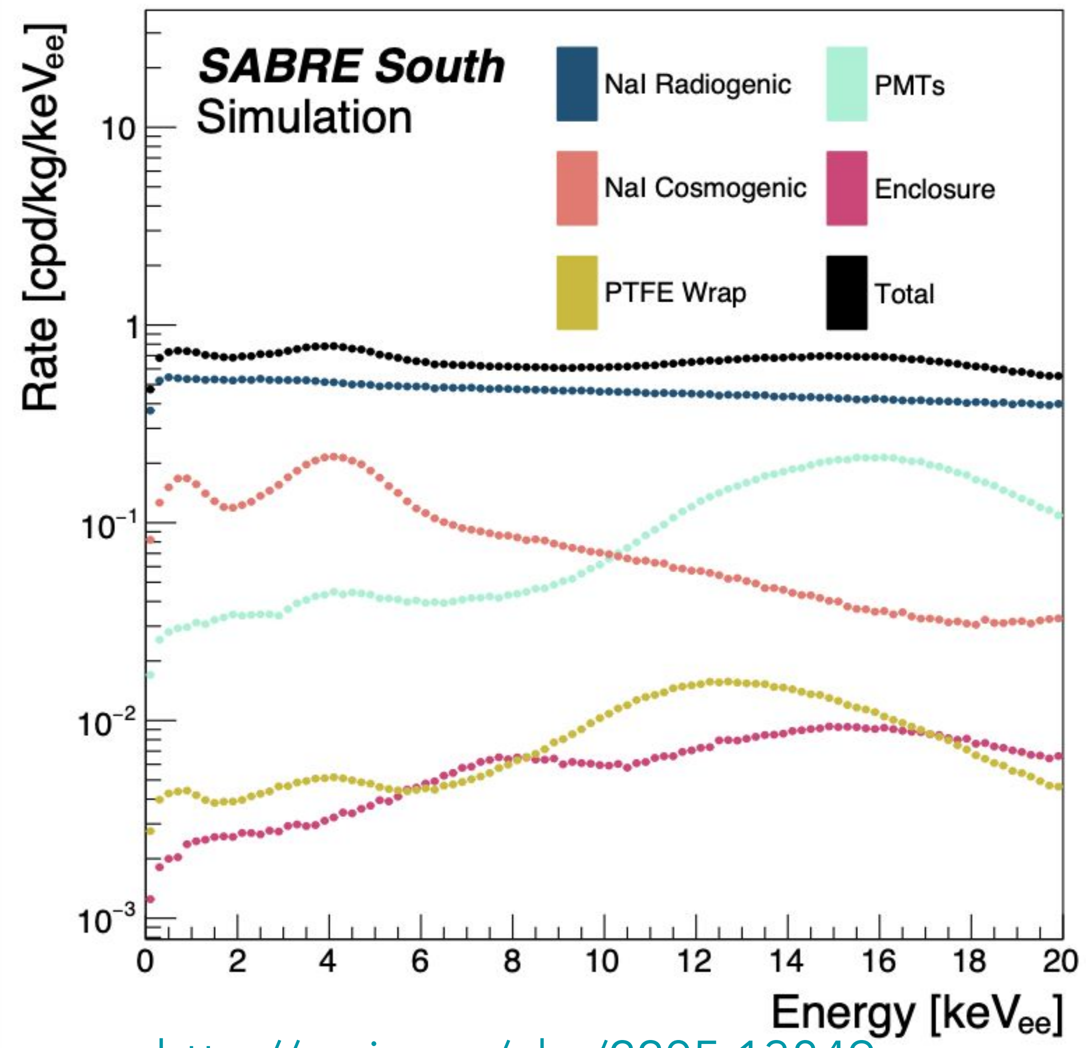
3x3 NaI matrix with 15 cm copper shielding + 40 (60) cm polyethylene lateral and top (bottom)

- 3 x 3 matrix of crystals of ~5 kg mass each
- Fully passive shielding design: 15 cm copper + 80 cm PE → enough shielding power and negligible contribution to the total background
- Expected background:
  - 0.5 cpd/kg/keV (with ZR)
  - 1 cpd/kg/keV (w/o ZR).



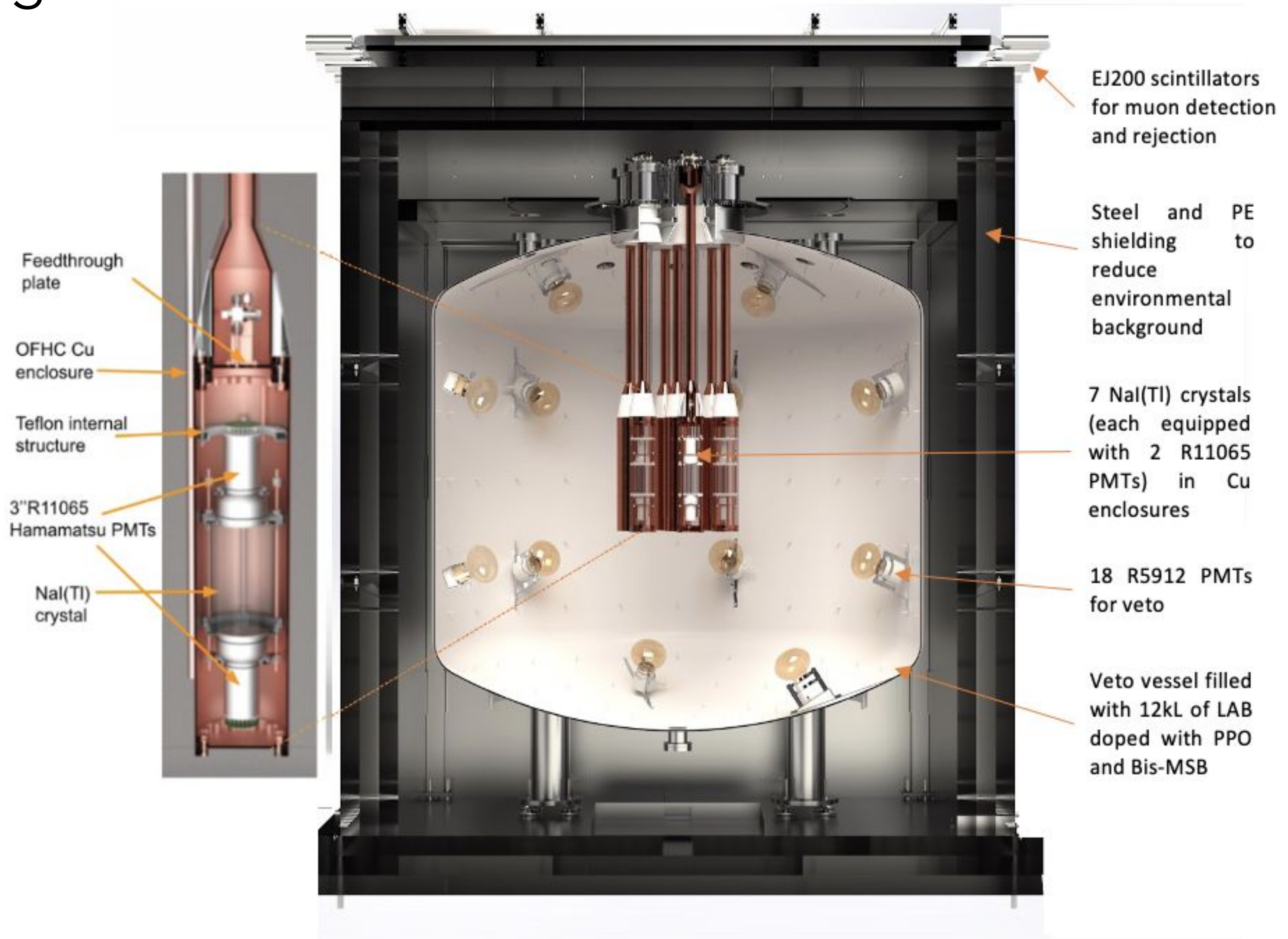
SABRE South status

- Design: 7 crystals array of ~5-7 kg mass (SABRE South TDR available online)
- Vessel + LAB, PMTs, muon detector, DAQ electronics, Crystal insertion system ... all ready.
- Crystal procurement in synergy with SABRE North
- Highest purity crystals and largest active veto: 0.72 cpd/kg/keV.



http://arxiv.org/abs/2205.13849
(accepted to EPJC)

- SABRE South commissioning has started. Muon detector is already taking data underground.
- SABRE South detector deployment to be completed in 2025.



Conclusions

- Growing large NaI(Tl) crystals with radiopurity at the level of the ones from the DAMA/LIBRA experiment is a challenge not yet fully completed;
- Reproducibility (which is crucial for a detector array) is also an issue;
- We are confident we found the right path within SABRE (Astrograde + Zone Refining + partnership with reliable companies);
- Full test of the production chain with crystal NaI-42 coming soon;
- SABRE North TDR just approved by INFN CSN2, and SABRE South commissioning started: exciting times ahead!

THANK YOU FOR LISTENING

Extra slides

SABRE North and South synergy

SABRE North and South detectors have **common core features:**

- Same crystal production and R&D.
- Same detector module concept (Ultra-pure crystals and HPK R11065 PMTs)
- Common simulation, DAQ and data processing frameworks
- Exchange of engineering know-how with official collaboration agreements between the ARC Centre of Excellence for Dark Matter and the INFN

SABRE North and South detectors **have different shielding designs:**

- SABRE North has opted for a fully passive shielding due to the phase out of organic scintillators at LNGS. Direct counting and simulations demonstrate that this is compliant with the background goal of SABRE North at LNGS.
- SABRE South will be the first experiment in SUPL, the liquid scintillator will be used for in-situ evaluation and validation of the background in addition to background rejection and particle identification.

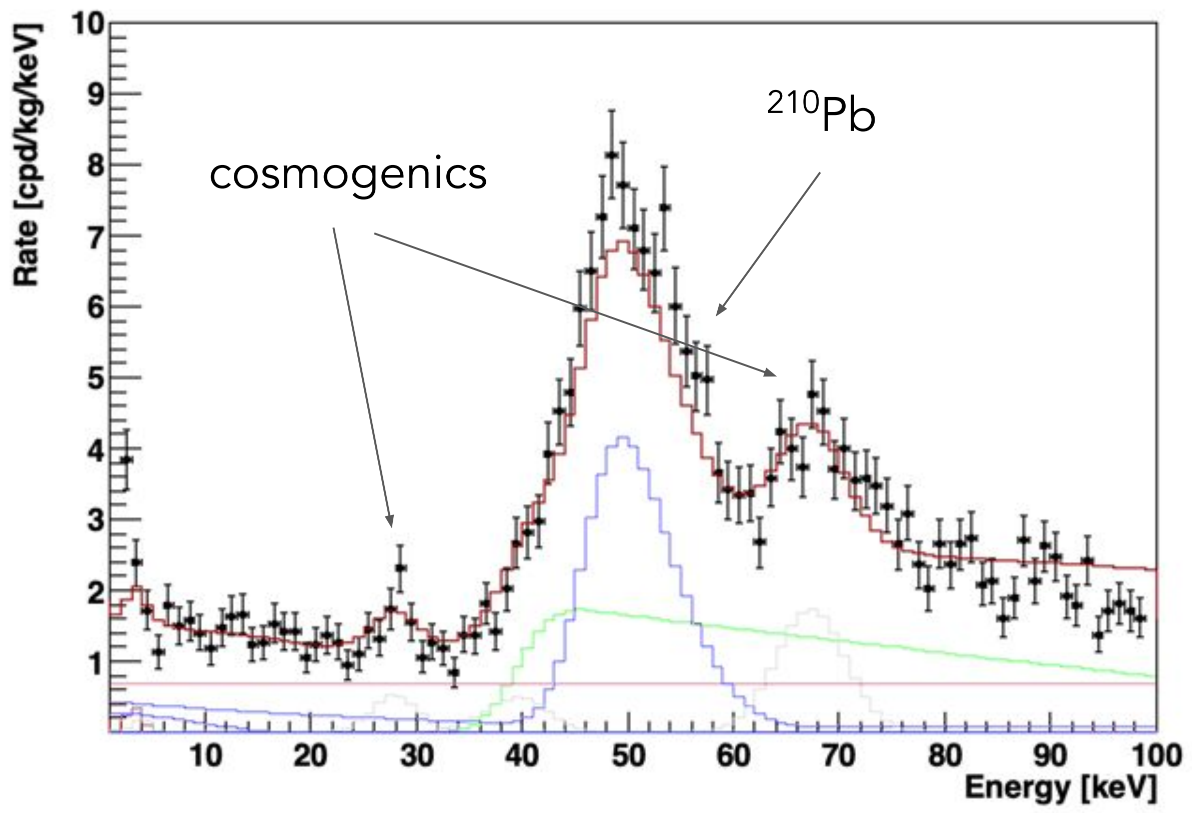
SABRE North facilities @LNGS (2024)

New SABRE experimental area in the corridor between Hall B and Hall A.

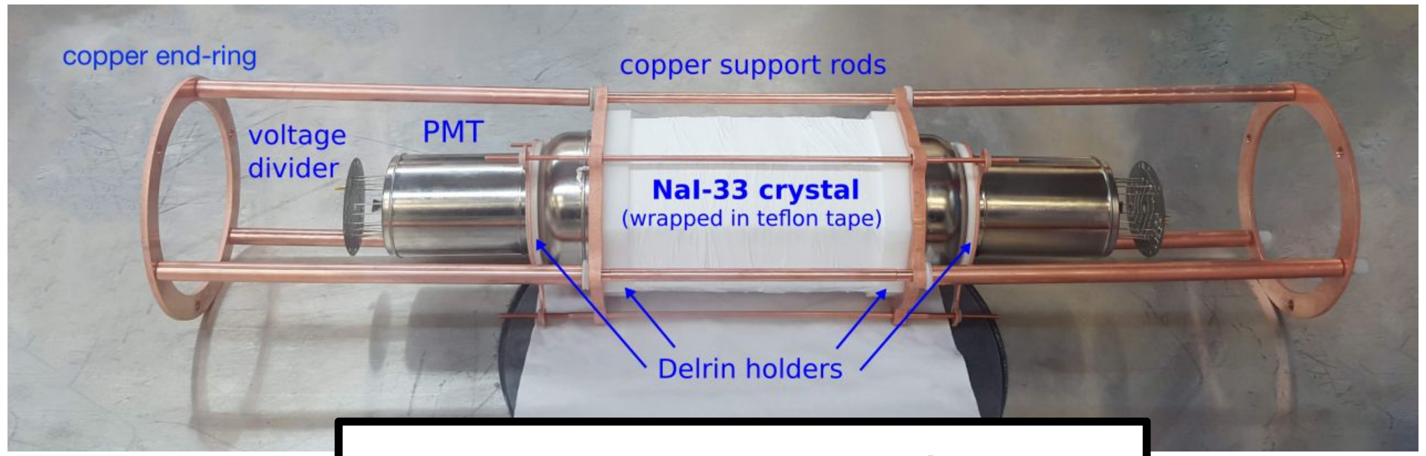
Consists of a two storeys building:

1. Ground floor (PT): set-up SABRE NORTH
2. First floor (P1). DAQ & counting room

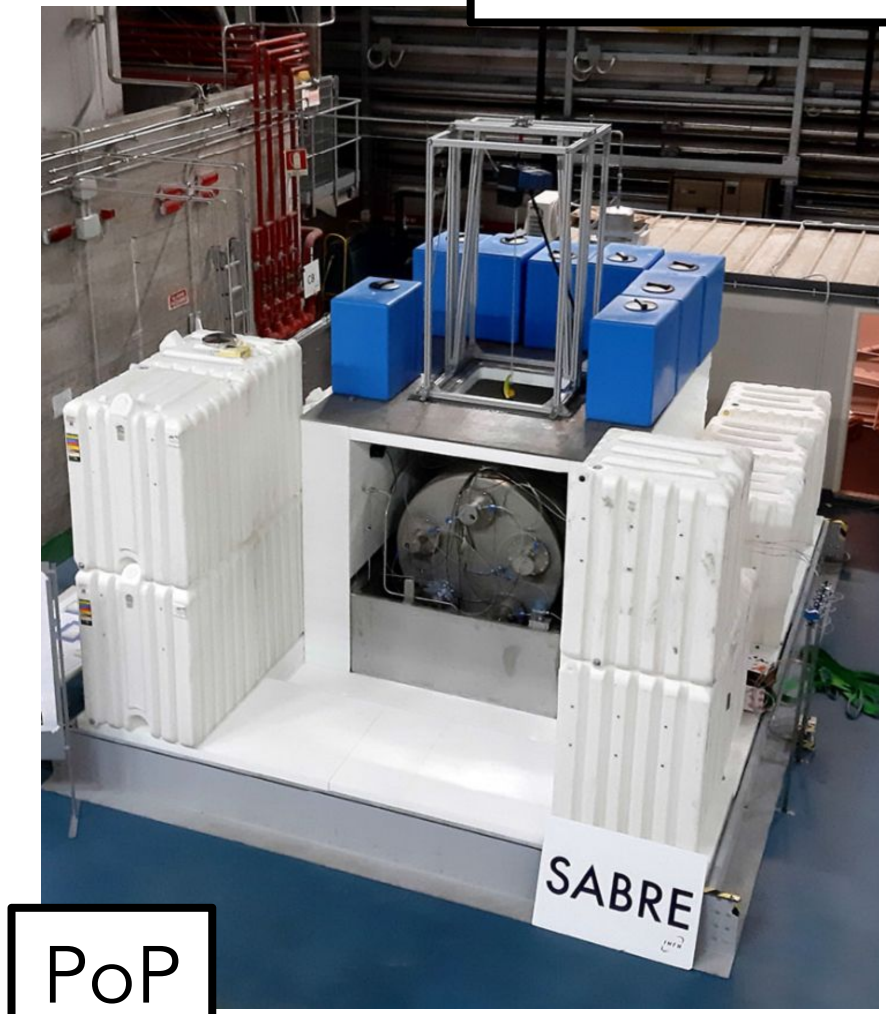
Preliminary energy spectrum from NaI-41



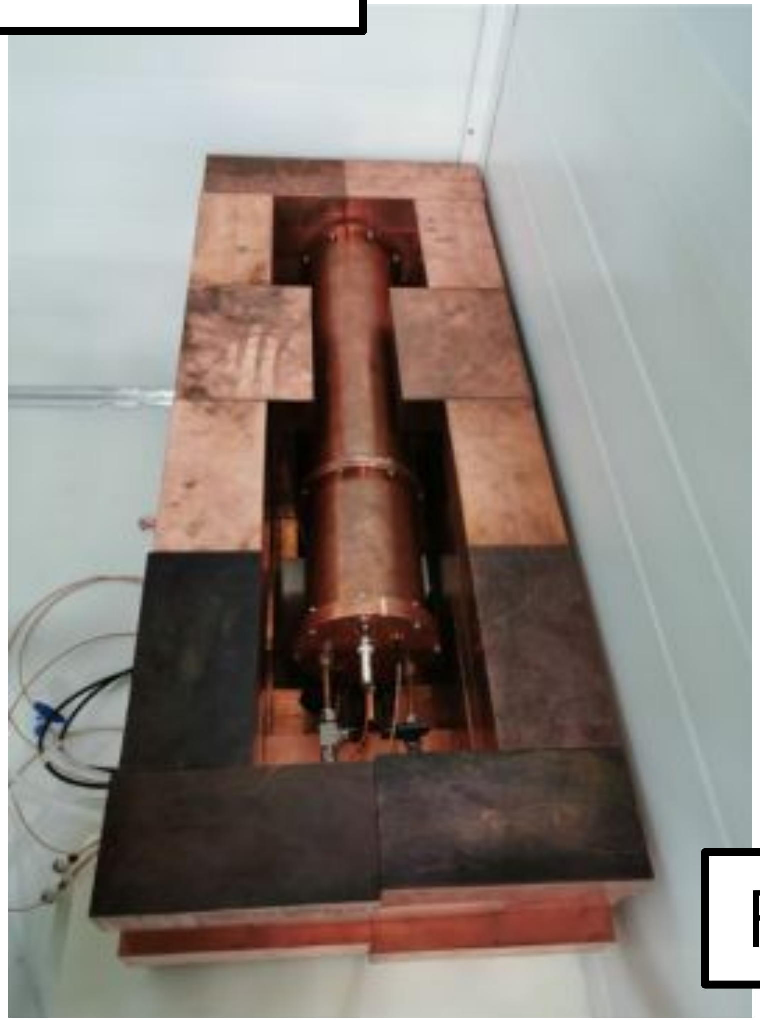
The SABRE Proof-of-Principle @LNGS (2018-2022)



Detector module



PoP



PoP-dry

- Run in 2020 with Borexino liquid scintillator and NaI-33
  - 2 tons active veto with 10 8-inch PMTs + H_2O shielding
- Exploited successfully ^{40}K tagging with sensitivity at the level of 1 ppb
- Demonstration by direct counting of first crystal production after DAMA/LIBRA with background in [1,6] keV of order 1 cpd/kg/keV
- PoP-dry run in 2021: passive shielding with additional layer of copper
  - confirmed background level

SABRE North facilities @LNGS (2022-2023)

- Two passive shielding setups for crystal characterization
- A clean room with SABRE glovebox for crystal assembling

- 10 cm Cu + 15 cm of Pb
- Host 1 detector module
- Lexan box flushed with N2

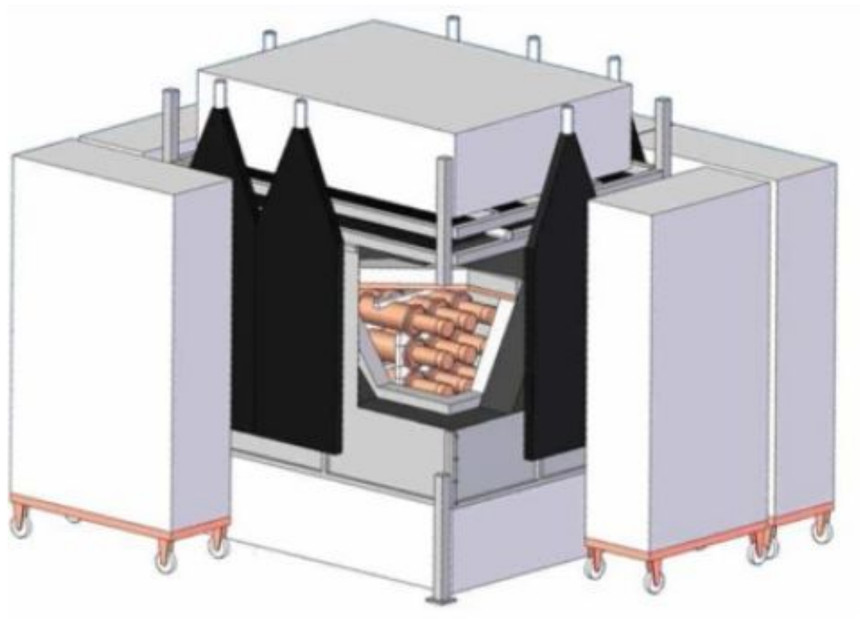
- 30 cm Cu shielding
- Host 1-3 detector modules
- Flushed with N2

SABRE glovebox
for the handling of
hygroscopic NaI(Tl) crystals.

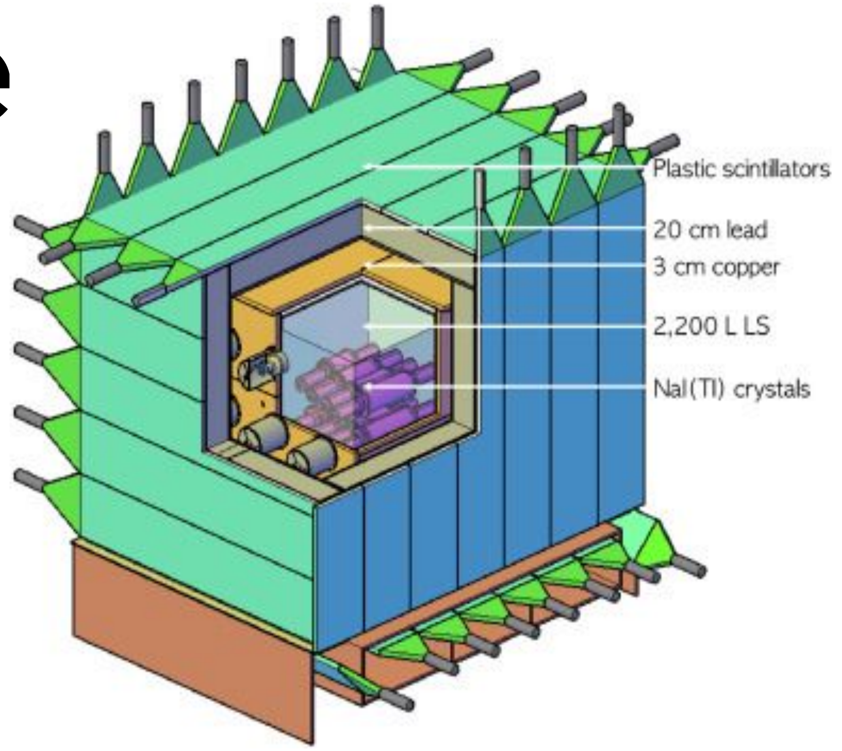
- A new site for SABRE North has been identified at LNGS.
- Moving of the equipment in progress.

NaI experimental landscape

ANAIS
@Canfranc,
Spain



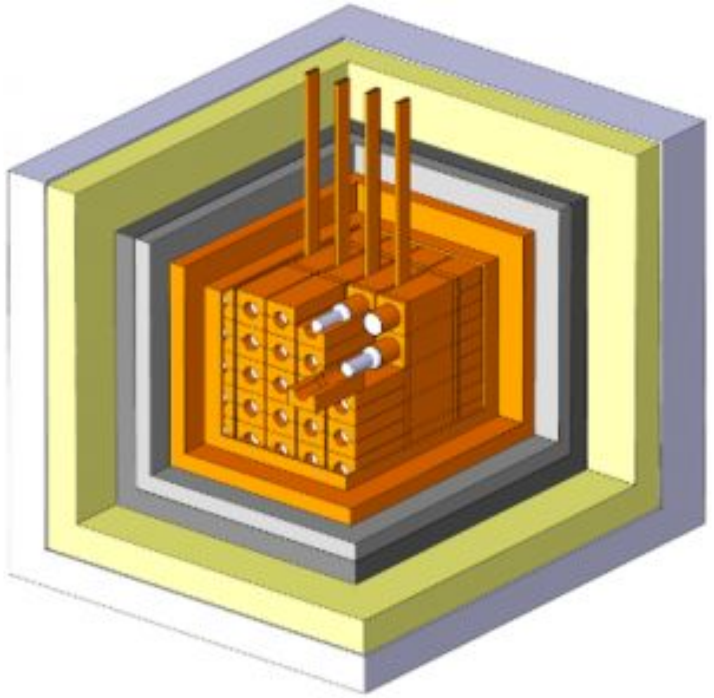
ANAIS, TAUP 2023



COSINE-100
@Yang Yang,
South Korea

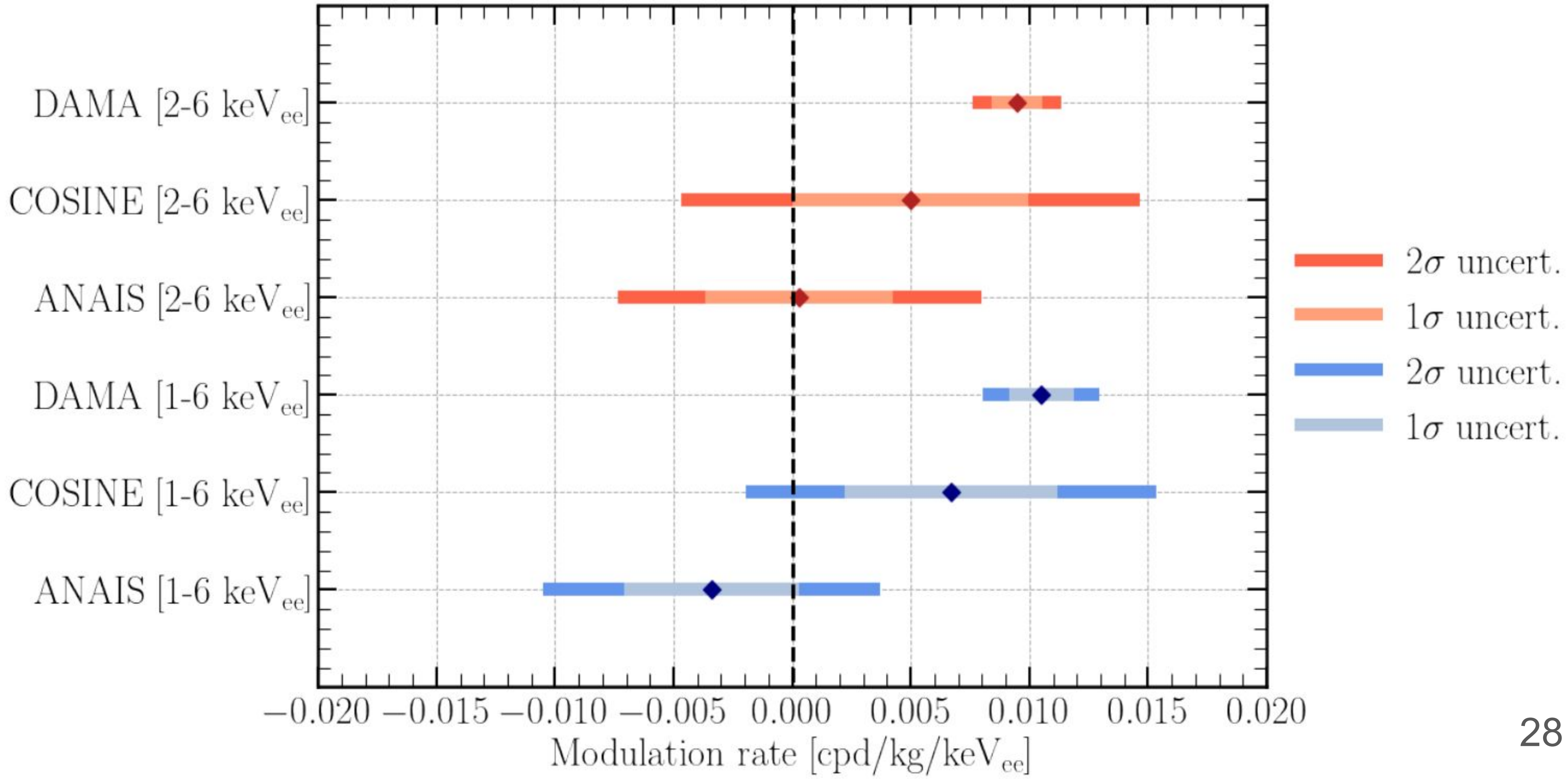
Phys. Rev. D 106, 052005 (2022)

DAMA/LIBRA @LNGS, Italy



arXiv: 2211.15861

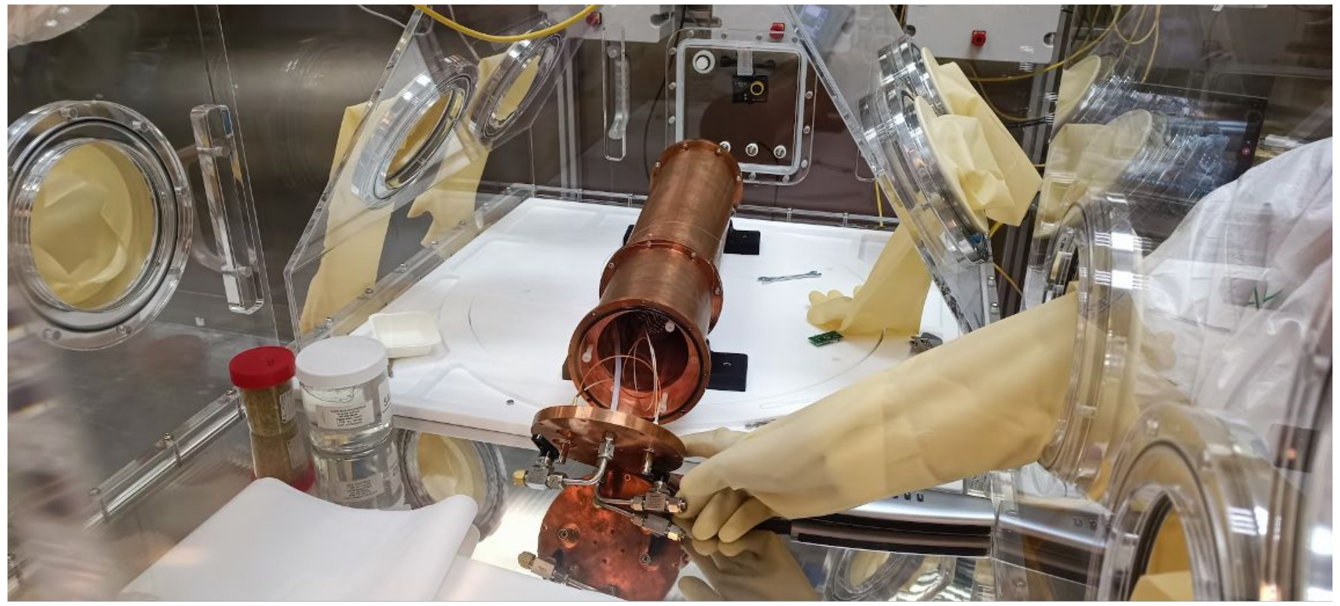
Nucl. Phys. At. Energy 22 (2021) 329-342

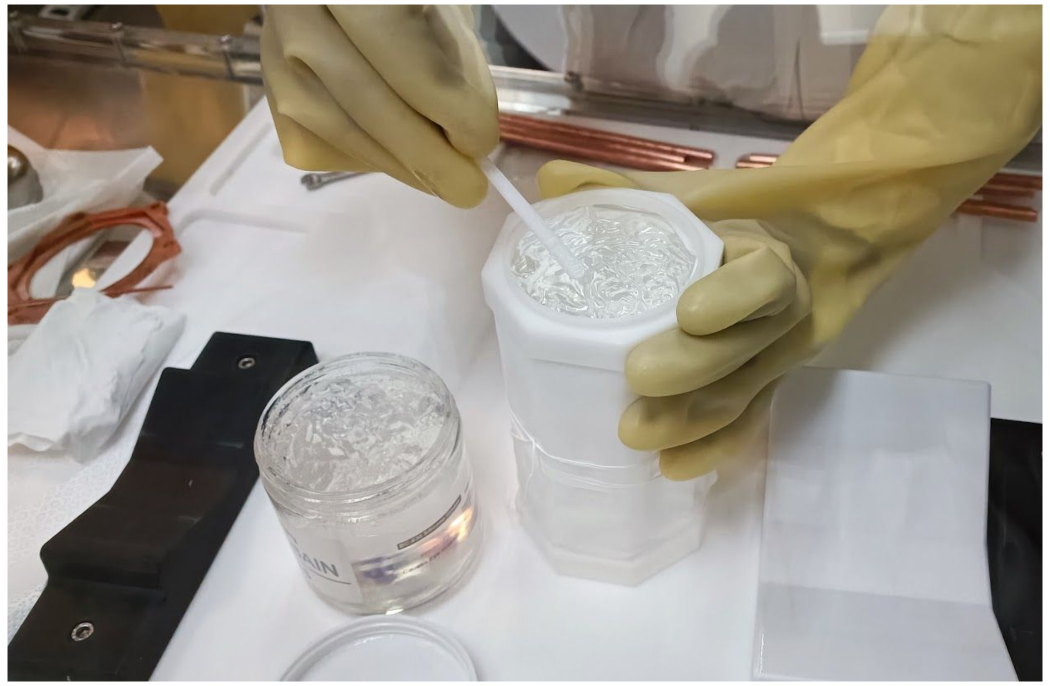


Crystal operations in glovebox 2022-24

- 27/09/2022 change of teflon reflector in NaI-33
- 29/11/2022 change of tefon reflector in NaI-33
- 7/12/2022 first assembly of NaI-37
- 24/01/2023 second assembly of NaI-37

All operations successful and moisture level in the glove-box kept always below 5% RH

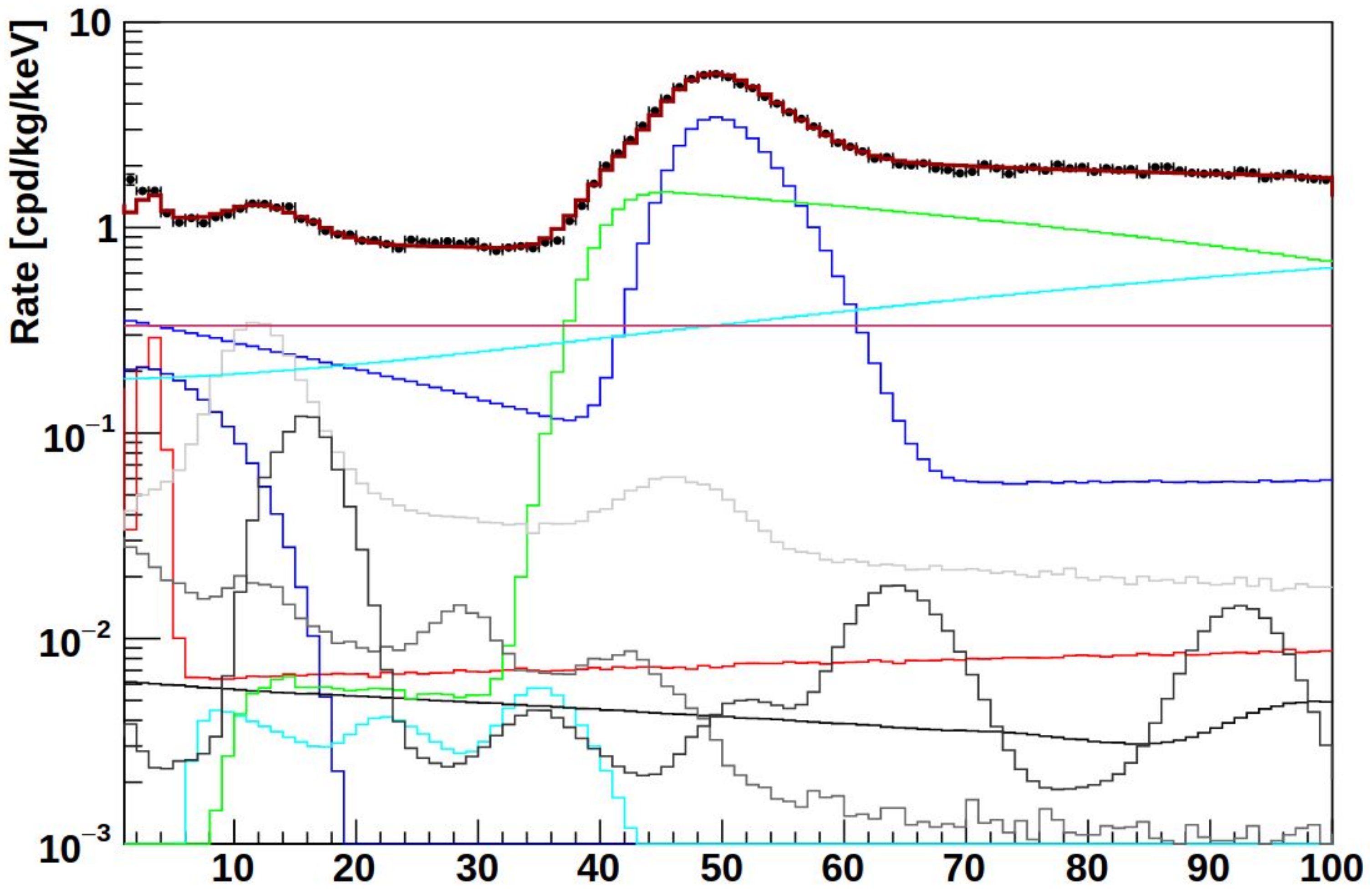




SABRE background model (NaI-33)

- Background model updated since Eur. Phys. J. C (2022) 82:1158
- Background from reflector is not dominant (now constrained from direct measurements)
- Dominant backgrounds: ^{210}Pb in crystal bulk and external background

Source	Rate in ROI [1,6] keV [cpd/kg/keV]	Activity from fit
40K	0.125	0.16±0.01 mBq/kg
210Pb bulk	0.333	0.49±0.05 mBq/kg
210Pb reflector bulk	0.054	11±1 mBq/kgPTFE
210Pb reflector surface	0.023	<0.6 mBq/m2
3H	0.198	24±2 mBq/kg
129I	0.0003	1.03±0.05 mBq/kg
238U	0.006	5.9±0.6 mBq/kg
232Th	0.0003	1.6±0.3 mBq/kg
PMT	0.003	1.9±0.4 mBq/PMT
External	0.185	0.89±0.05 relative unit to reference spectrum
Other b's	0.333	297±15 counts
TOTAL	1.26±0.27	



NaI crystals background comparison

Crystal	^{nat}K (ppb)	^{238}U (ppt)	^{210}Pb (mBq/kg)	^{232}Th (ppt)	Active mass (kg)
DAMA [1]	13	0.7-10	(5-30)x10^{-3}	0.5-7.5	250
ANAIS [2]	31	<0.81	1.5	0.36	112
COSINE [3]	35.1	<0.12	1-1.7	<2.4	~60
SABRE [4]	4.3	0.4	0.49	0.2	~35+40=75 (total goal)
PICOLON [5]	<20	-	<5.7x10^{-3}	-	~20 (goal)

[1] Nucl.Instrum.Meth.A 592 (2008) 297-31
[2] Eur.Phys.J.C 79 (2019) 5, 412
[3] Eur.Phys.J.C 78 (2018) 490
[4] Eur.Phys.J.C 81 (2021) 4, 299, Phys.Rev.D 104 (2021) 2, L021302, Eur.Phys.J.C 82 (2022) 12, 1158
[5] PTEP 2021 (2021) 4, 043F01