Radiopure $ZnMoO_4$ scintillating bolometers for the LUMINEU double-beta experiment

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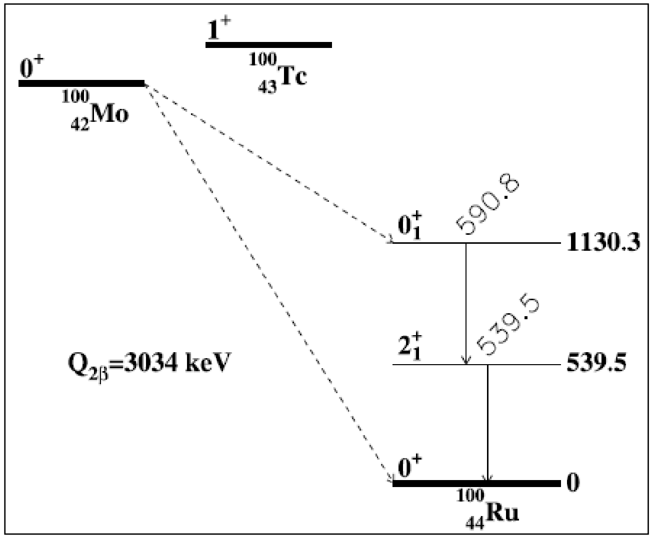
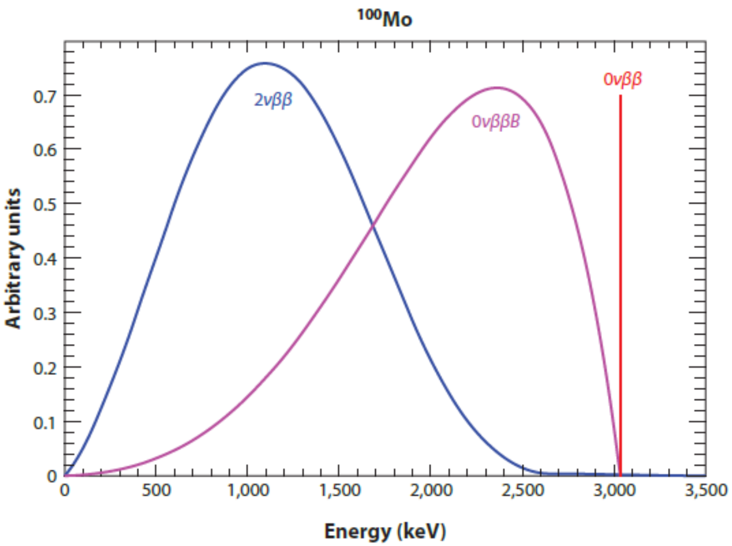
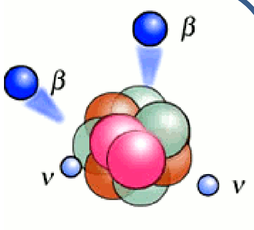
on behalf of
the LUMINEU and the EDELWEISS Collaborations

Double-beta decay





2ν2β decay

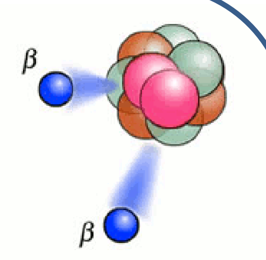


$$(A, Z) \rightarrow (A, Z + 2) + 2e^- + 2\bar{\nu}_e$$

- **Rarest nuclear transformation**
  ($T_{1/2} \sim 10^{18}$–10^{24} yr)
- **Experimental information about NME's**

$$\left[T_{1/2}^{2\nu}\right]^{-1} = G^{2\nu} \cdot |M^{2\nu}|^2$$

0ν2β decay

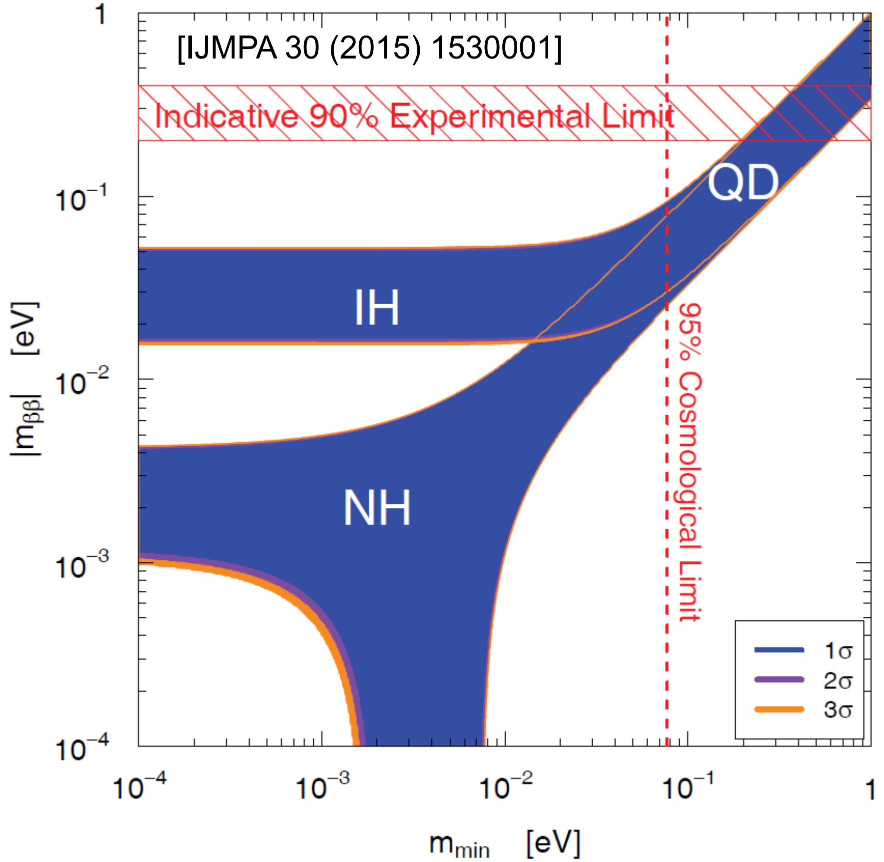


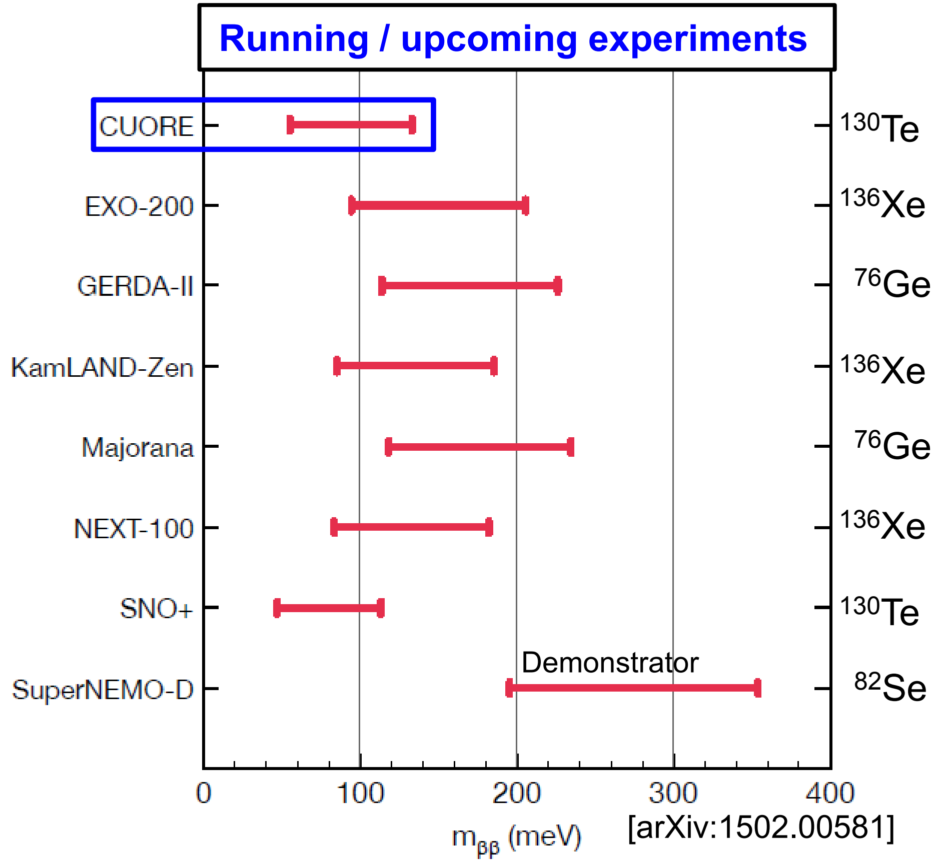
$$(A, Z) \rightarrow (A, Z + 2) + 2e^-$$

- **Lepton number non-conservation**
- **Neutrino properties**
  (Majorana nature of neutrino, the origin and absolute scale of neutrino masses, hierarchy of mass eigenstates, CP-violating phases)
- **Other effects beyond the Standard Model**
  (right-handed currents admixture in weak interactions, existence of Majoron,..)

$$\left[T_{1/2}^{0\nu}\right]^{-1} = G^{0\nu} \cdot |M^{0\nu}|^2 \cdot \langle\mu\rangle^2 \qquad \langle\mu\rangle^2 = \frac{\langle m_{\beta\beta}\rangle^2}{m_e^2}$$

Status of $0\nu2\beta$ search





- **Best sensitivity: $\langle m_{\beta\beta}\rangle$ ~ 0.15–0.9 eV, $T_{1/2}$ ~ 10^{24}–10^{25} yr**
  KamLAND-ZeN (^{136}Xe), GERDA-I (^{76}Ge), CUORICINO (^{130}Te), NEMO-3 (^{100}Mo)
- **Current generation experiments will start probe IH**
  $\langle m_{\beta\beta}\rangle$ ~ 0.05–0.2 eV, $T_{1/2}$ ~ 10^{26} yr (over next ~ 3–5 yrs)
- **New / advanced technology is needed to cover IH**
  $\langle m_{\beta\beta}\rangle$ ~ 0.02–0.05 eV, $T_{1/2}$ ~ 10^{27} yr

See e.g. [AHEP 2014 (2014) 951432] and refs. therein

Advanced 0ν2β detector: scintillating bolometer

Sensitivity to 0ν2β decay

$$\lim T_{1/2}^{0\nu} \propto \delta \cdot \varepsilon \cdot \sqrt{\frac{M \cdot t}{B \cdot \Delta E}}$$

Achieved for bolometers (CUORICINO / CUORE),
Expected for scintillating bolometers (e.g. LUMINEU)

δ – natural isotopic abundance of 2β nuclide ~ 35%

~ 99% (enriched 2β nuclide)

ε – detection efficiency > 80% (detector =2β source)

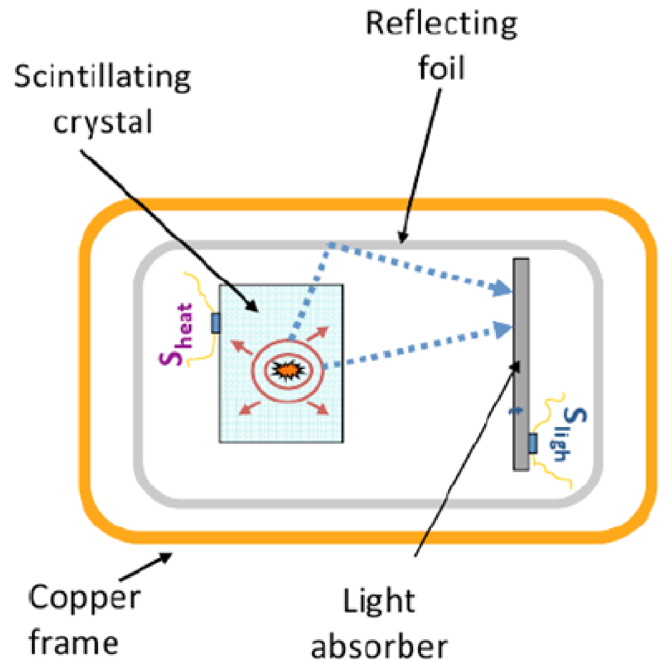
M – mass of ββ source ~ 100–1000 kg

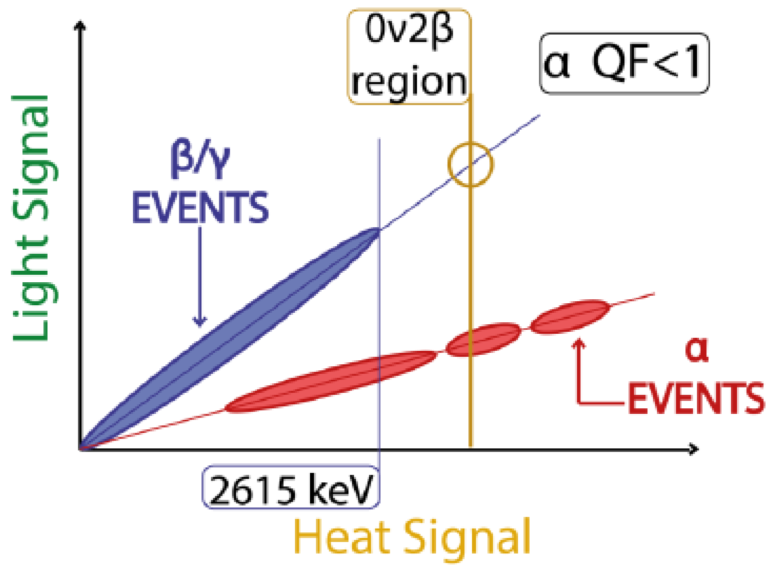
t – time of measurements ~ 5 yr

Δ***E*** – energy range of interest ~ 0.2% FWHM

B – background in ROI ~ 10^{-1}–10^{-2} c/kg/keV/yr
(nuclide with $Q_{2\beta}$ < 2.6 MeV, α's contribute 60% to *B*)

~ 10^{-4} c/kg/keV/yr (nuclide with $Q_{2\beta}$ > 2.6 MeV, α/γ discrimination > 5σ)





Next-generation $ZnMoO_4$-based $0\nu2\beta$ experiment

^{100}Mo as 2β nuclide:

- **$Q_{2\beta}$ = 3034 keV**
- **Reasonable price of enrichment** (up to 99%)
- **Promising theoretical predictions on $0\nu2\beta$ rate**

Scintillating bolometers:

- **$Zn^{100}MoO_4$ crystals** (~0.4 kg each; ^{100}Mo ~ 97%)
- **Cylindrical Cu holders**
- **Ge-based light detectors** (∅60×0.5 mm)

Performance:

- **FWHM = 6 keV @ 3 MeV**
- **Threshold > 20 keV**
- **Rejection of α's > 99.9%**
- **Anticoincidence cut**
- **5-yr data taking**

GEANT4-calculated background contributions to the ROI

Bkg source	Activity (μBq/kg)	Bkg (c/keV/kg/yr)	Bkg reduced (c/keV/kg/yr)
^{208}Tl in ZMO	**10**	**3×10^{-3}**	**~ 10^{-4} ***
^{228}Th in Cu	20	2×10^{-5}	2×10^{-5}
^{56}Co in ZMO	0.06	2×10^{-5}	2×10^{-5}
^{56}Co in Cu	0.02	2×10^{-5}	2×10^{-5}
$2\nu2\beta$ ^{100}Mo	**~9×10^{3}**	~3×10^{-4}	**~3×10^{-4} ****
Neutrons		< 10^{-4}	< 10^{-4}
Muons		~ 10^{-4}	< 10^{-4}
Total		**~3×10^{-3}**	**~4×10^{-4}**

Possible further reducing: * – longer vetoing of α-β's; ** – PSD

5-yr Sensitivity of $Zn^{100}MoO_4$-based $0\nu2\beta$ experiment

$Zn^{100}MoO_4$ crystals	^{100}Mo (kg)	$T_{1/2}$ (10^{25} yr)	$\langle m_{\beta\beta} \rangle$ (meV)
4	0.676	0.53	167–476
40	6.76	4.95	55–156
2000	**338**	**92.5**	**13–36**

[J.W. Beeman et al., Phys. Lett. B 710 (2012) 318]

Luminescent **U**nderground **M**olybdenum **I**nvestigation for **NEU**trino mass and nature

ANR funds (France); Start: October, 2012; Duration: 4 years

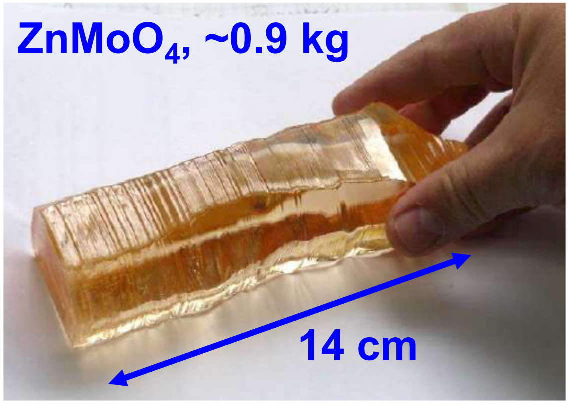
FRANCE
CSNSM Orsay (CNRS/IN2P3 + Paris Sud)
IAS Orsay (CNRS + Paris SuD)
ICMCB Bordeaux (CNRS + Bordeaux Univ.)
CEA Saclay

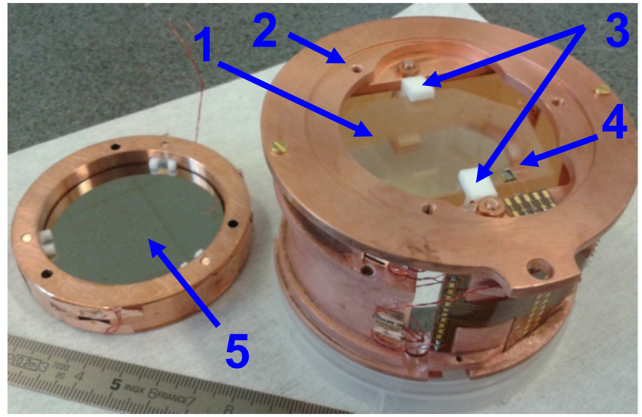
UKRAINE INR Kyiv
RUSSIA NIIC Novosibirsk
GERMANY KIP Heidelberg (Heidelberg Univ.)
ITALY INFN Milano Bicocca (Univ. Milano Bicocca)

Development of the technology based on scintillating bolometers for a next-generation $0\nu2\beta$ experiment

- **Development of zinc molybdate ($ZnMoO_4$) based scintillating bolometers**
  (R&D of Mo purification and ZMO growth conditions to produce large mass colorless samples from natural and enriched Mo; high detectors' performance: FWHMγ ~ 6 keV, α/γ separation > 5σ; high radiopurity: ^{228}Th/^{226}Ra ~ 0.01 mBq/kg, total α activity (except ^{210}Po) ~ 1 mBq/kg)
- **A pilot $0\nu2\beta$ experiment ~1 kg of ^{100}Mo: LUMINEU project**
- **Expansion to ~10 kg of ^{100}Mo: LUCINEU program to prove the technology in view of CUORE follow-up**
  (LUCIFER + LUMINEU; ^{100}Mo is available: MoU IN2P3-ITEP-INFN)

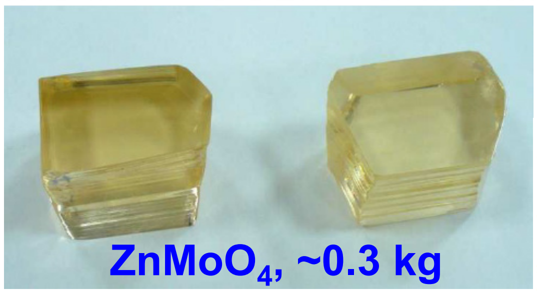
Large volume precursor of LUMINEU program



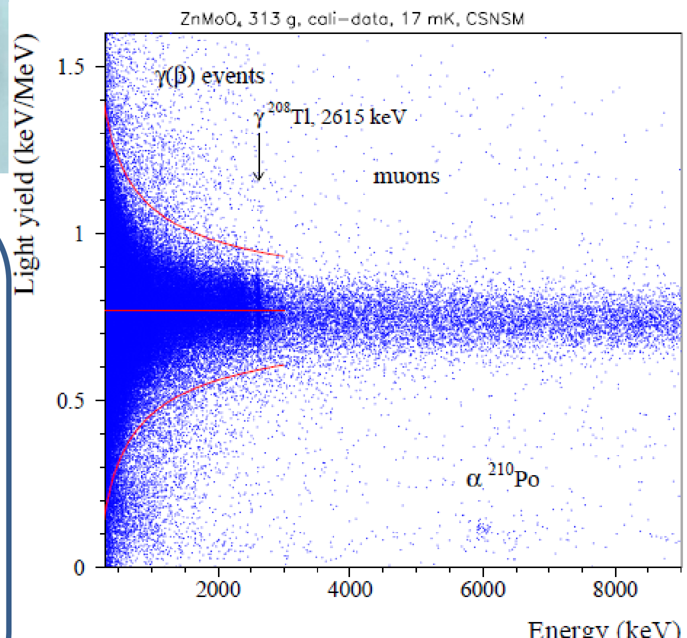


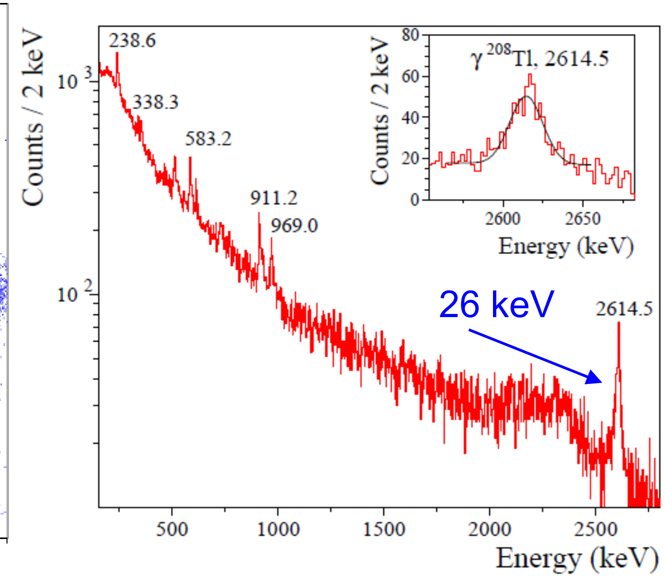
$ZnMoO_4$-based scintillating bolometer

(1) 313 g $ZnMoO_4$ crystal
(2) Cu holder of the detector
(3) PTFE supporting elements
(4) Two NTD thermistors
(5) Two Ge light detectors



Aboveground test at 17 mK at CSNSM





$ZnMoO_4$ development

- ZnO (99.995%) provided by Umicore (Belgium)
- MoO_3 (99.999%) purified at NIIC (Novosibirsk)
- Platinum crucible ∅8 cm
- LTG Cz crystal growth along the [001] axis

[Crystallogr. Rep. 59 (2014) 288]

Then, it was tested at Modane Underground Lab

LUMINEU: Purification of molybdenum

Two stage purification technique was developed at NIIC (Novosibirsk)

Sublimation in vacuum

with addition ~1% of $ZnMoO_4$

$ZnMoO_4 + WO_3 \rightarrow ZnWO_4 + MoO_3\uparrow$

Efficient removing of U/Th is expected

Material	Concentration of impurities (ppm)			
	Si	**K**	**Fe**	**W**
Initial MoO_3	600	100–500	6	200–500
Sublimation	100–500	10–50	2–6	100–200
Double sublimation	**70**	**1–8**	**< 1**	**30–40**

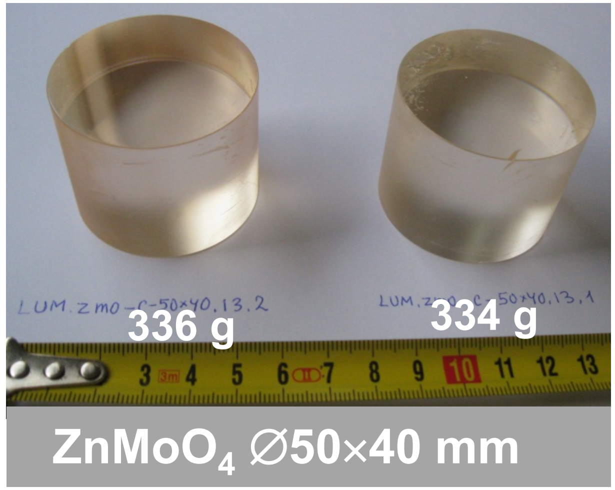
Recrystallization from aqueous solutions

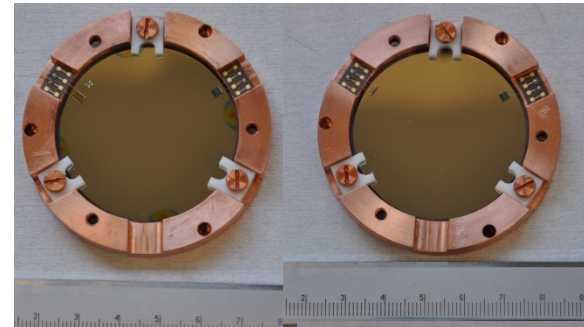
$MoO_3 + 2NH_4OH = (NH_4)_2MoO_4 + H_2O$ (using zinc molybdate as a collector)

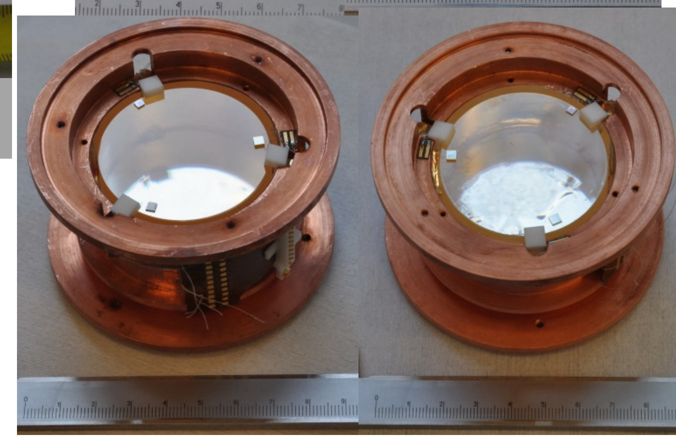
Material	Concentration of impurities (ppm)				
	Si	**K**	**Ca**	**Fe**	**W**
High purity MoO_3	60	50	60	8	200
Sublimation and recrystallization from aqueous solutions	30	10	12	5	130
Double sublimation and recrystallization from aqueous solutions	**-**	**< 10**	**< 10**	**< 5**	**< 50**

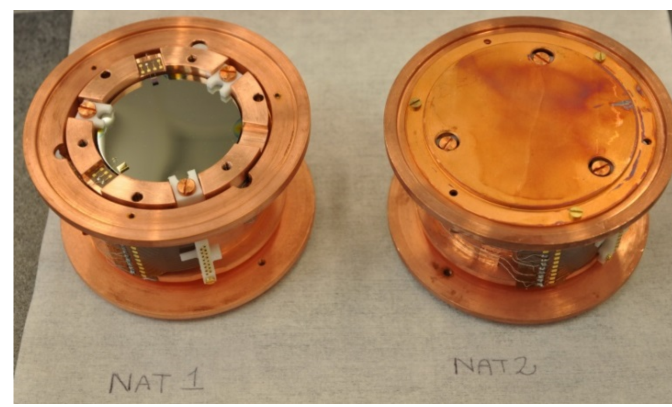
[L. Bergé et al., JINST 9 (2014) P06004]

LUMINEU: First massive $ZnMoO_4$ bolometers









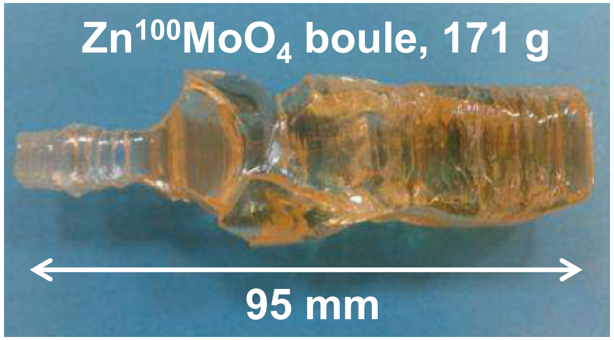
[E. Armengaud et al., submitted to JINST]

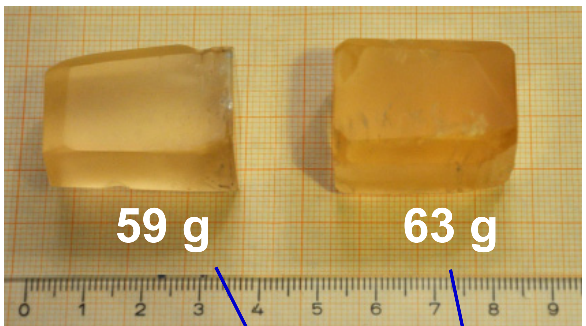
Technology of high quality large $ZnMoO_4$ crystal producing is developed

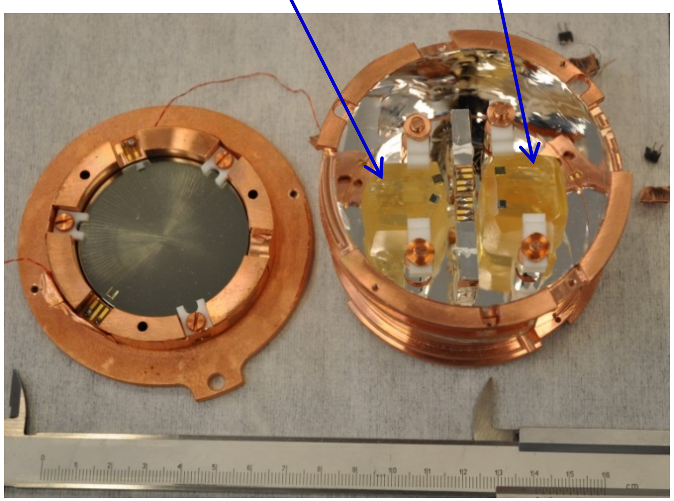
- Molybdenum was purified by using double recrystallization from aqueous solutions
- Advanced quality $ZnMoO_4$ boule was grown by directional solidification along [001] using LTG Cz (crystal yield ~ 80% of initial charge)
- $ZnMoO_4$ crystal boule was melted and then crystallized again (to test recrystallization)
- Produced $ZnMoO_4$ elements have the size expected for a pilot LUMINEU $0\nu2\beta$ experiment

LUMINEU: First enriched $Zn^{100}MoO_4$ detectors

[A.S. Barabash et al., EPJC 74 (2014) 3133]

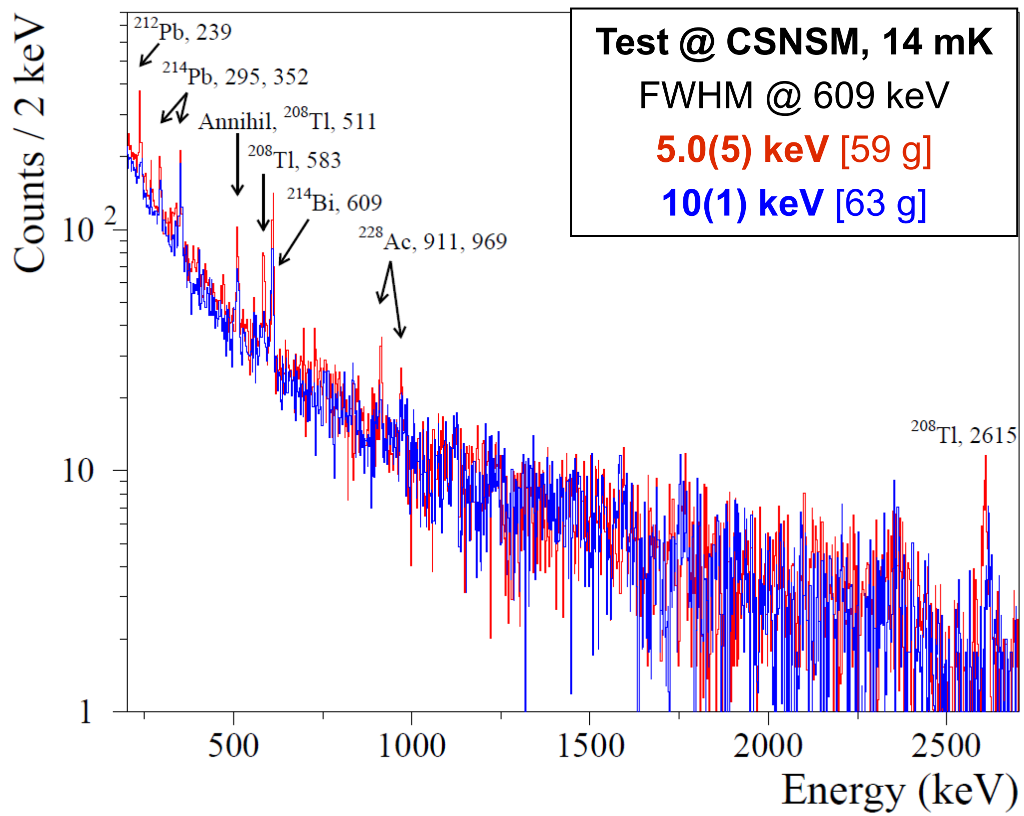






Development of first enriched $Zn^{100}MoO_4$ crystal

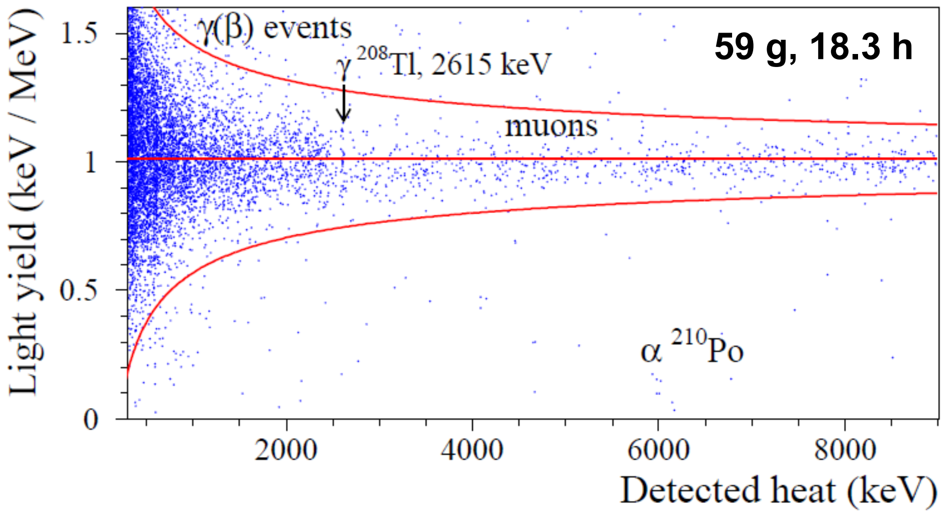
- $^{100}MoO_3$ (99.5% enrichment in ^{100}Mo) was purified by using sublimation in vacuum and recrystallization from aqueous solutions
- $Zn^{100}MoO_4$ boule was grown at NIIC by using LTG Cz (crystal yield ~ 84% of initial compound)
- Total irrecoverable losses of ^{100}Mo ~ 4%

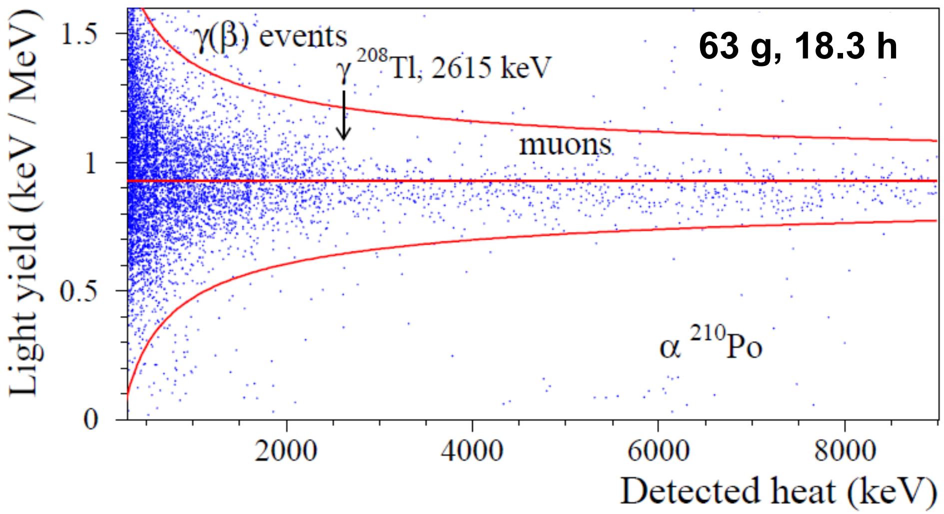


LUMINEU: Aboveground test of $Zn^{100}MoO_4$ array

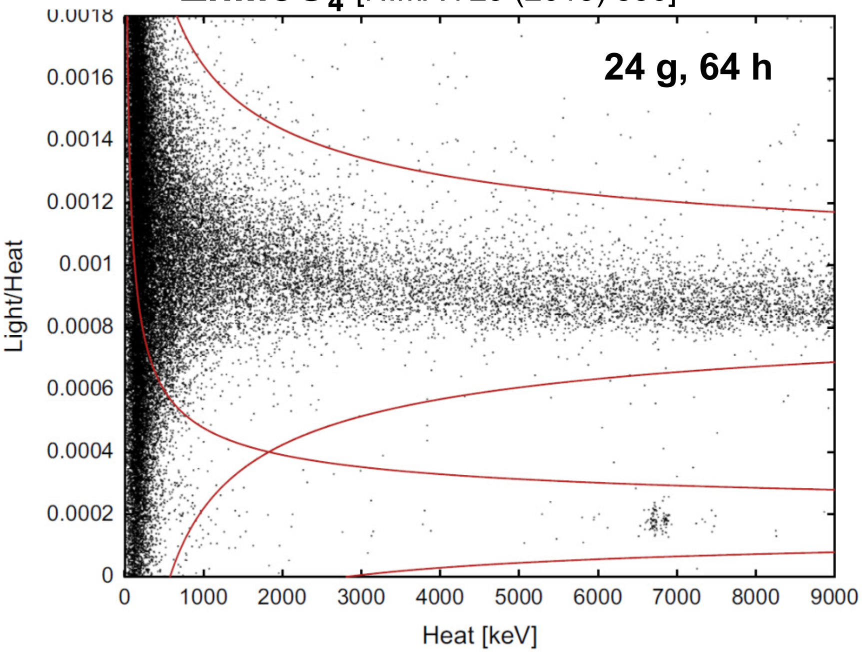
Encouraging radiopurity of $Zn^{100}MoO_4$ crystals

$Zn^{100}MoO_4$ [EPJC 74 (2014) 3133]





$ZnMoO_4$ [NIMA 729 (2013) 856]



Nuclide	Activity (mBq/kg)	
	$Zn^{100}MoO_4$	$ZnMoO_4$
^{228}Th	-	≤ 0.8
^{226}Ra	-	≤ 0.8
^{210}Po	~ 1	8(1)

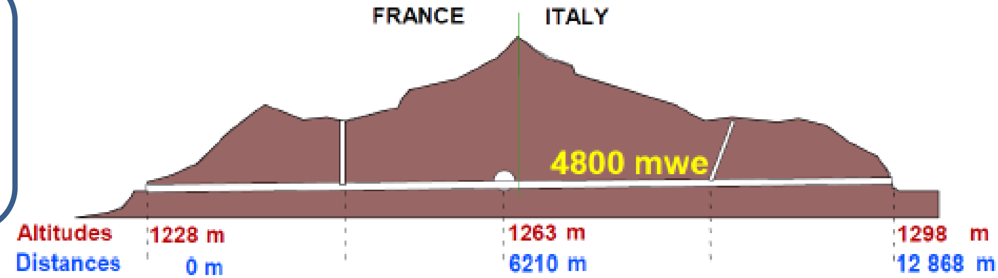
Underground cryogenic tests at Modane

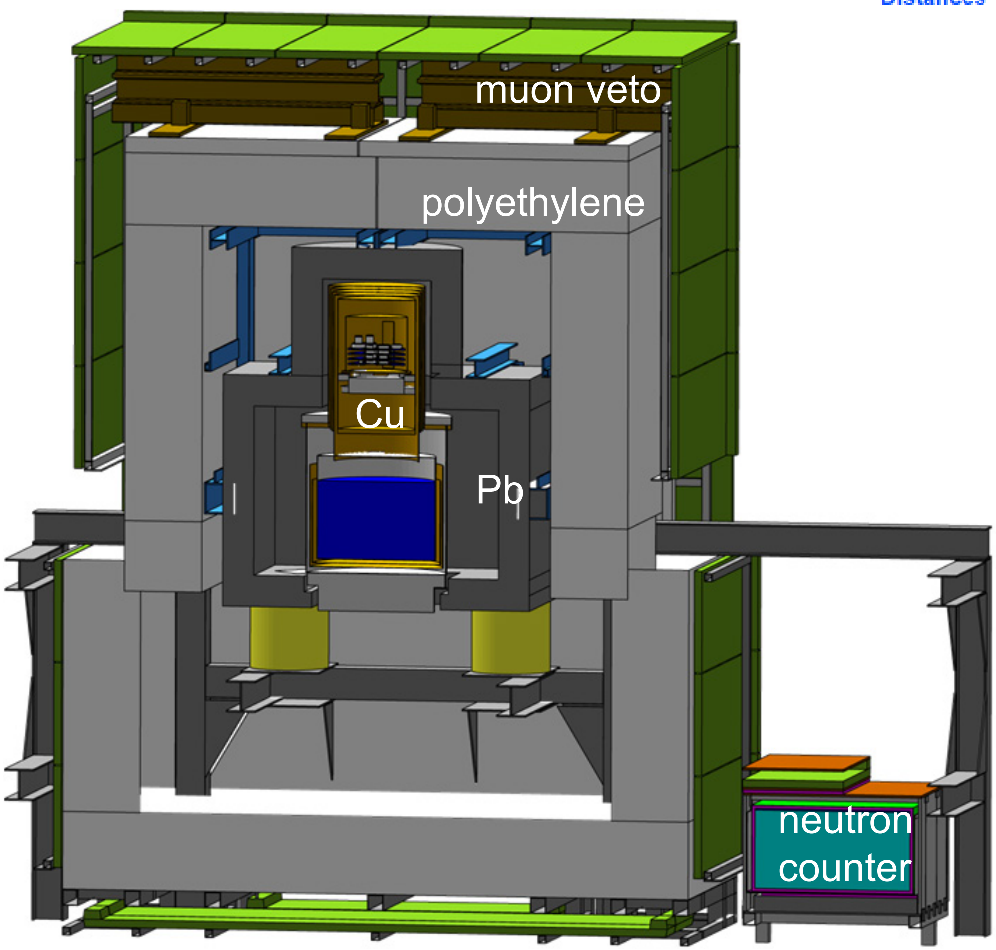
Laboratoire Souterrain de Modane

1.7 km rock overburden (~4.8 km w.e.)

5 μ/day/m^2; 10^{-6} n/day/cm^2 (>1 MeV)

Deradonized air flow (~30 mBq/m^3)





EDELWEISS set-up

Installation at LSM

Clean room (ISO Class 4)

Copper cryostat

^{3}He/^{4}He table top dilution refrigerator

Large experimental volume (50 liters)

Passive shield

Low radioactivity lead (~ 20 cm)

Polyethylene (~ 50 cm)

Detection μ / n / Ra

Muon veto (98.5% covering)

Neutron counter

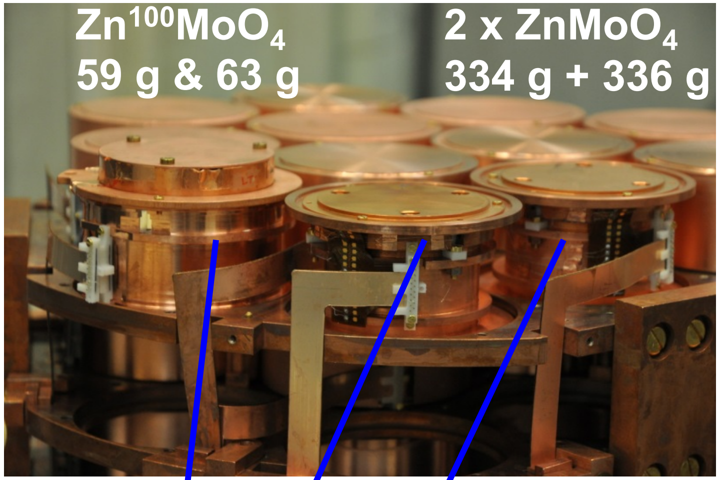
Radon counter

[E. Armengaud et al., PLB 702 (2011) 329]

LUMINEU detectors in the EDELWEISS set-up

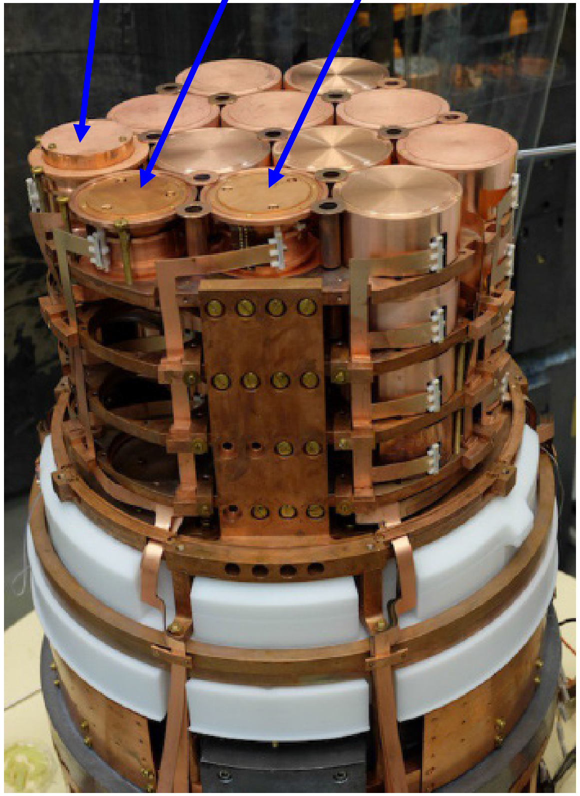
EDELWEISS-III commissioning run @ 19mK

- Improved cryogenic system
- Polyethylene at 1 K plate
- NOSV copper screens
- 15 germanium bolometers 800 g each
- Scintillating bolometer based on precursor $ZnMoO_4$ (313 g)
- Sept. 2013 – Feb. 2014



EDELWEISS-III physics run @ 18 mK

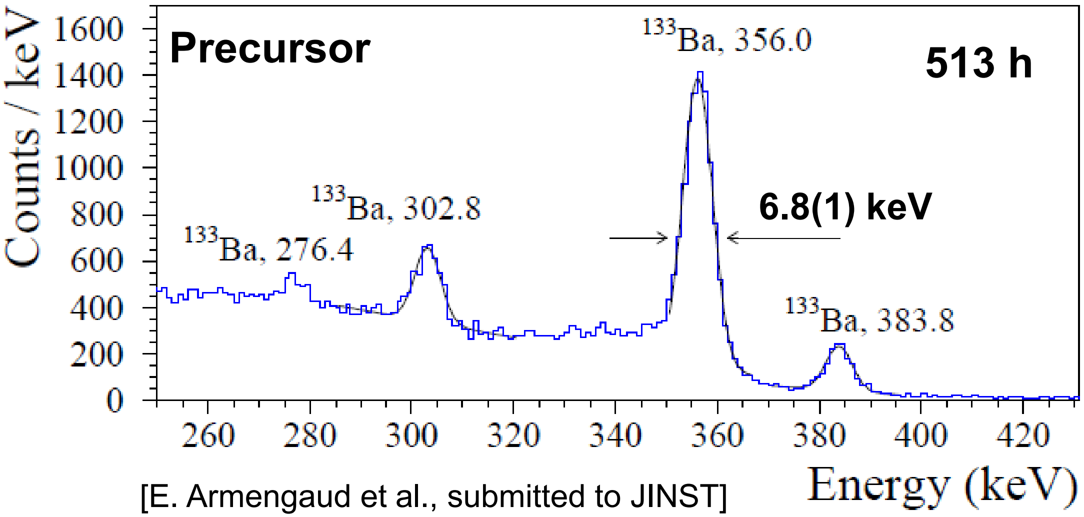
- Individual low bkg Kapton
- Implementation of device for thermal response control of LUMINEU detectors (since Feb. 2015)
- 36 germanium bolometers 800 g each
- $Zn^{100}MoO_4$ scintillating bolometers array
- 2 scintillating bolometers based on advanced quality $ZnMoO_4$ crystals
- June 2014 – present



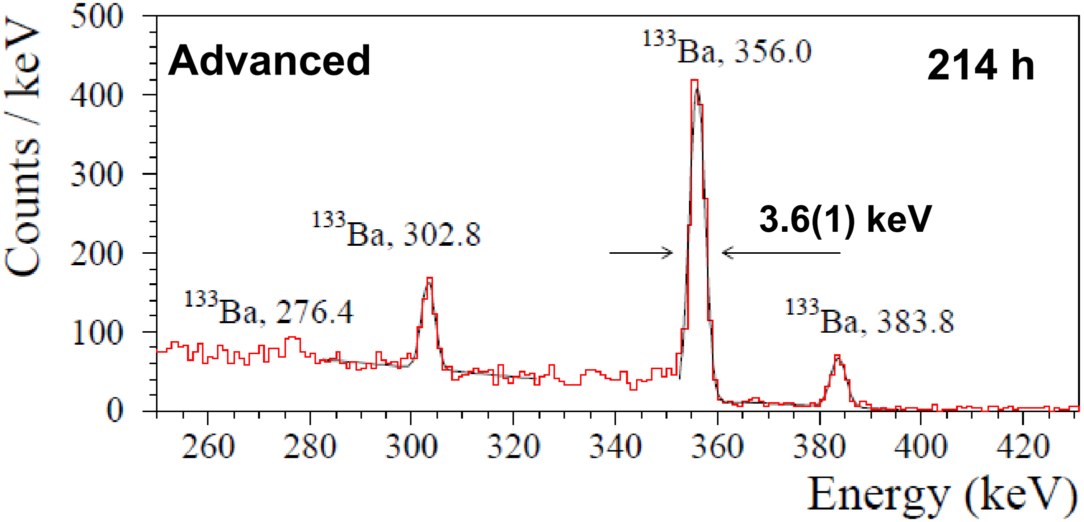
[E. Armengaud et al., submitted to JINST]

Precursor / Advanced $ZnMoO_4$: Calibration by ^{133}Ba

Excellent performance of $ZnMoO_4$ bolometers

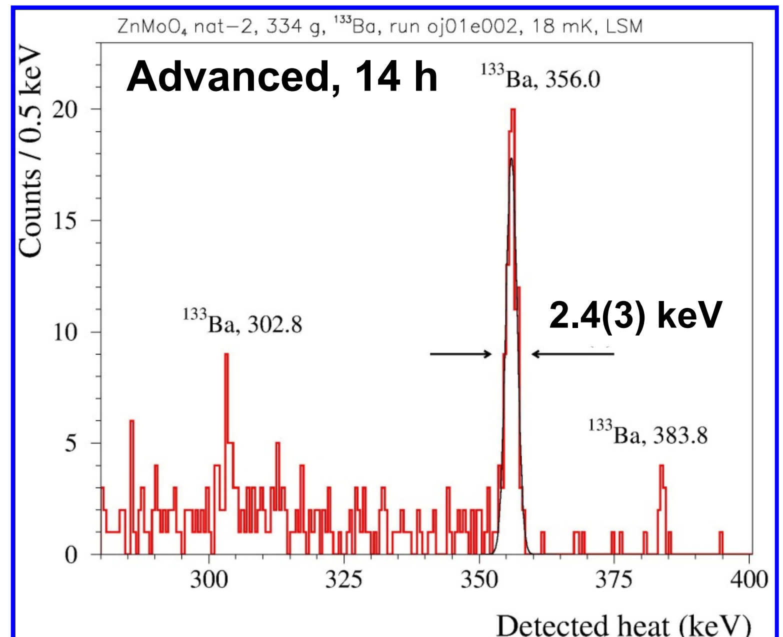


[E. Armengaud et al., submitted to JINST]



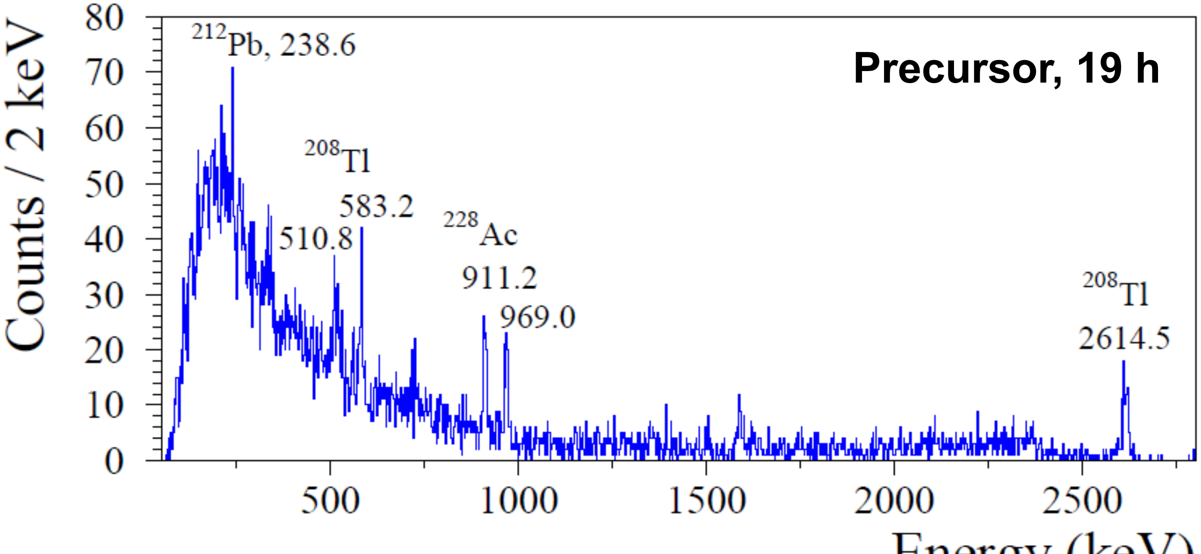
Performance in runs

$ZnMoO_4$ detector	FWHM (keV)	
	@ 356 keV	BsI
Precursor	6 ÷ 13	≈ 1.5
Advanced	3 ÷ 5	≈ 1.5

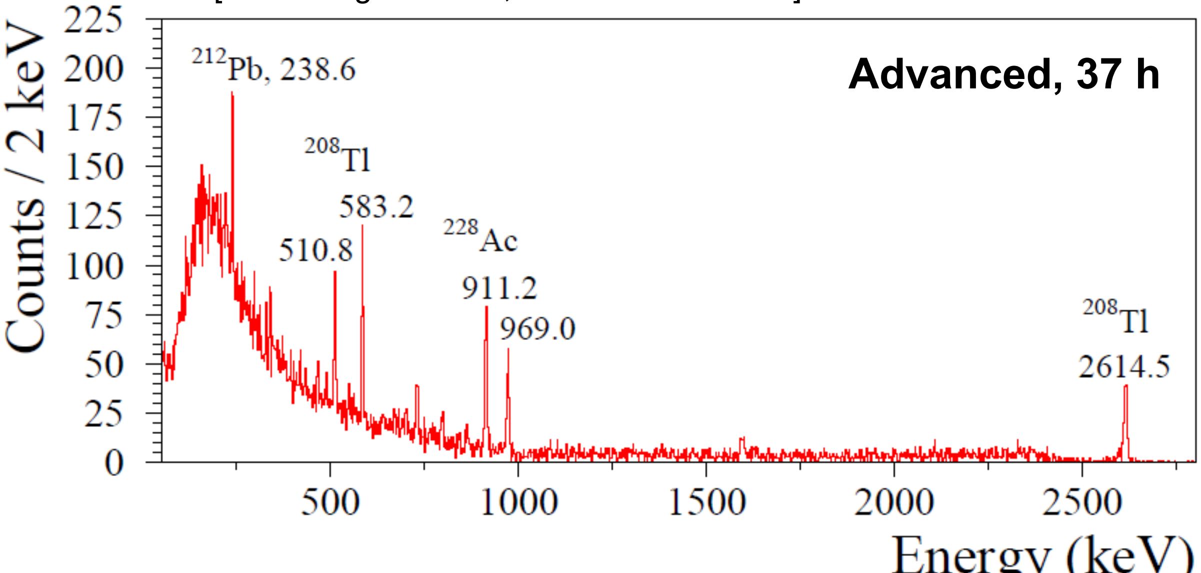


Precursor / Advanced $ZnMoO_4$: Calibration by ^{232}Th

Excellent performance of $ZnMoO_4$ bolometers



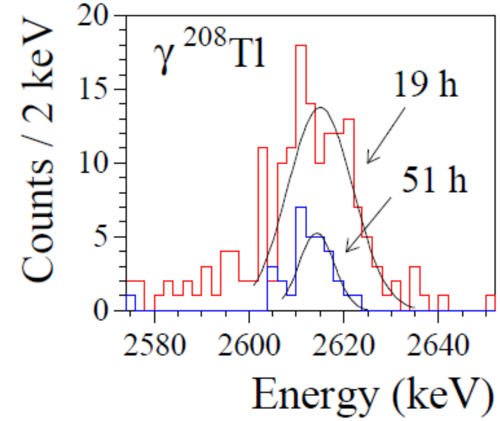
[E. Armengaud et al., submitted to JINST]



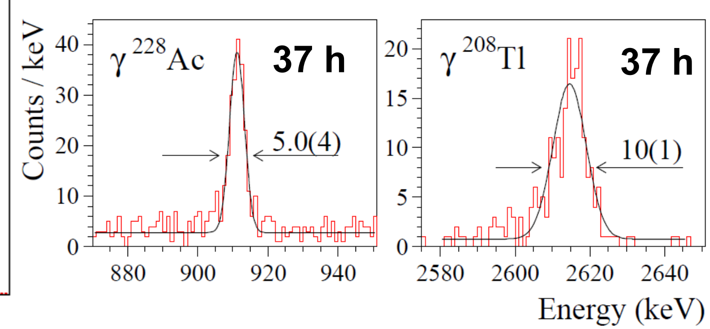
Strong / weak γ source

19 h: FWHM = 17(1) keV

51 h: FWHM = 9(2) keV

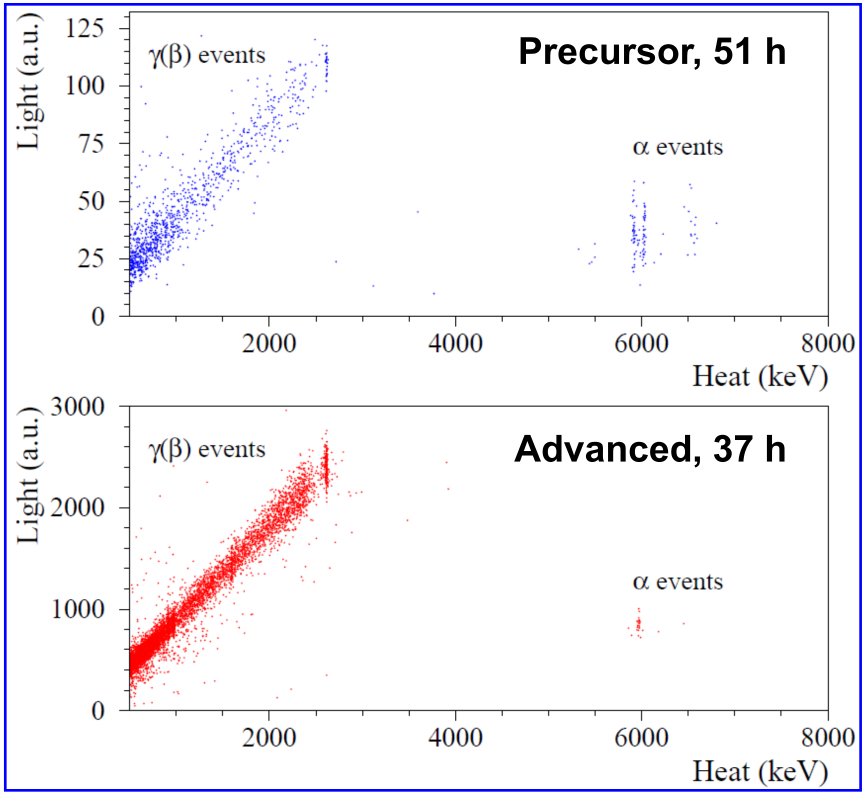


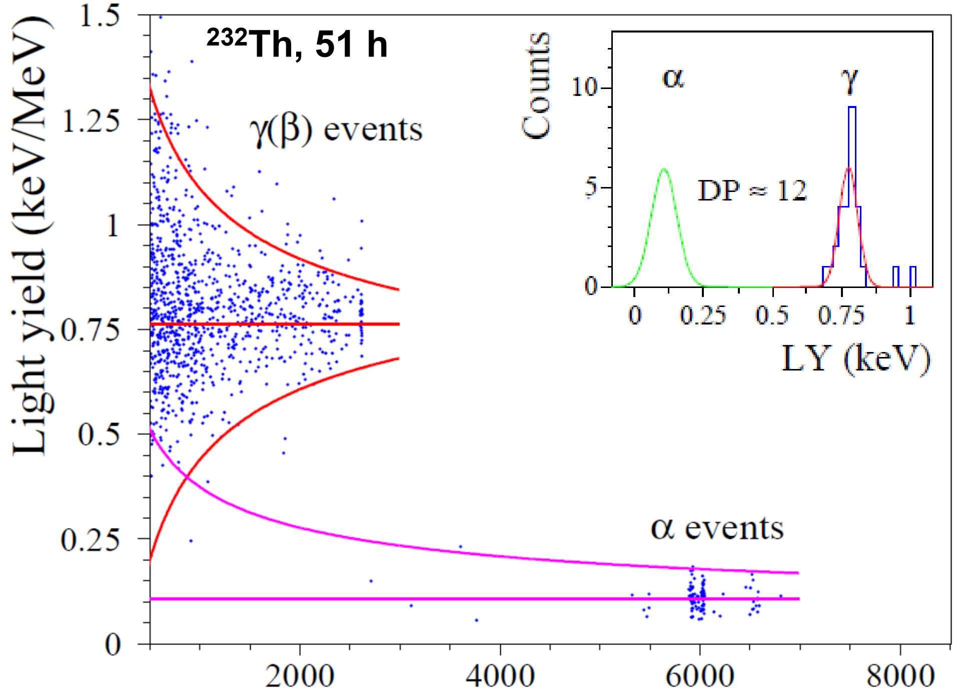
Strong γ source

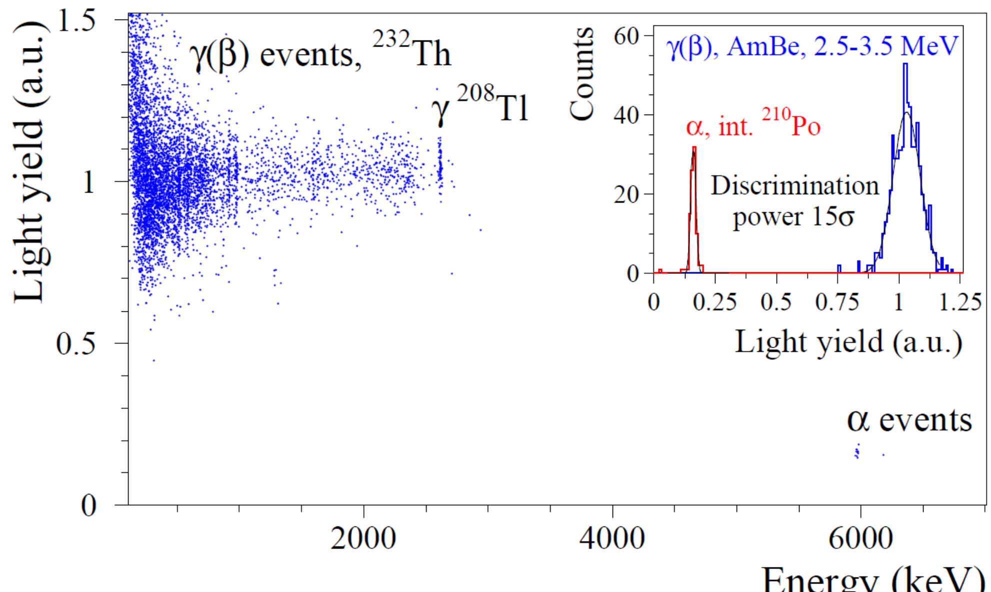


Precursor / Advanced $ZnMoO_4$: α/γ separation

Excellent α/γ separation







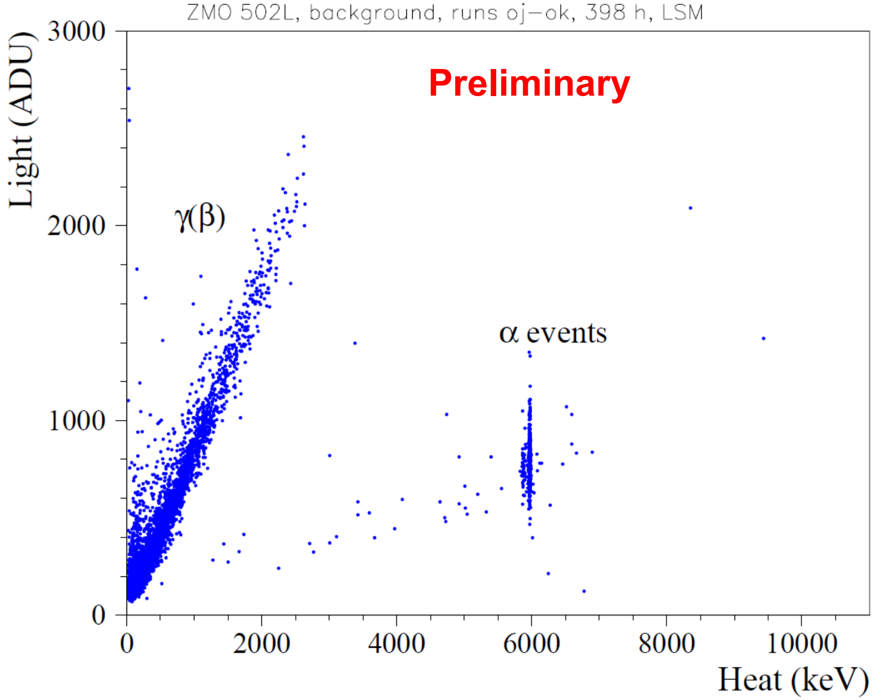
Discrimination power

$$DP(E) = \frac{\mu_{\gamma(\beta)}(E) - \mu_\alpha(E)}{\sqrt{\sigma^2_{\gamma(\beta)}(E) + \sigma^2_\alpha(E)}}$$

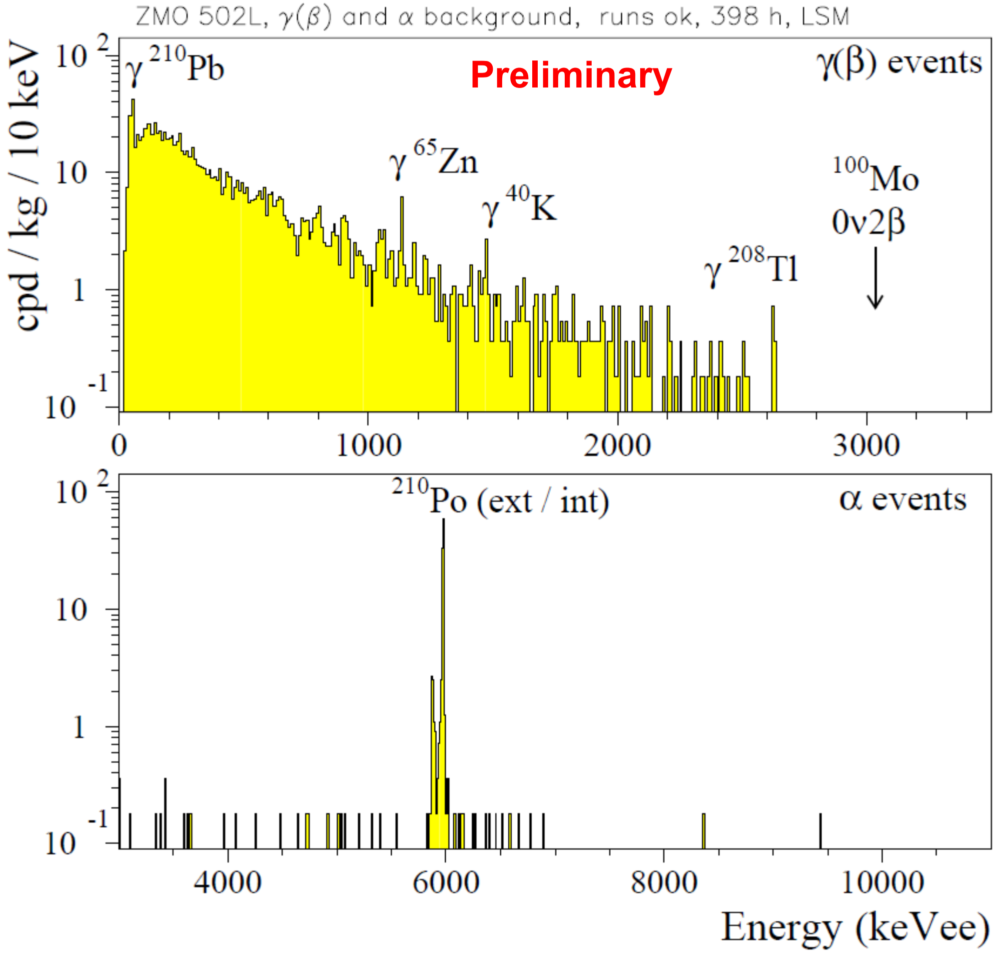
Background measurements with advanced $ZnMoO_4$

Perfect capability to get "zero" background in the ROI

$ZnMoO_4$, 334 g, Bkg, 398 h, LSM
Heat-vs-Light scatter-plot

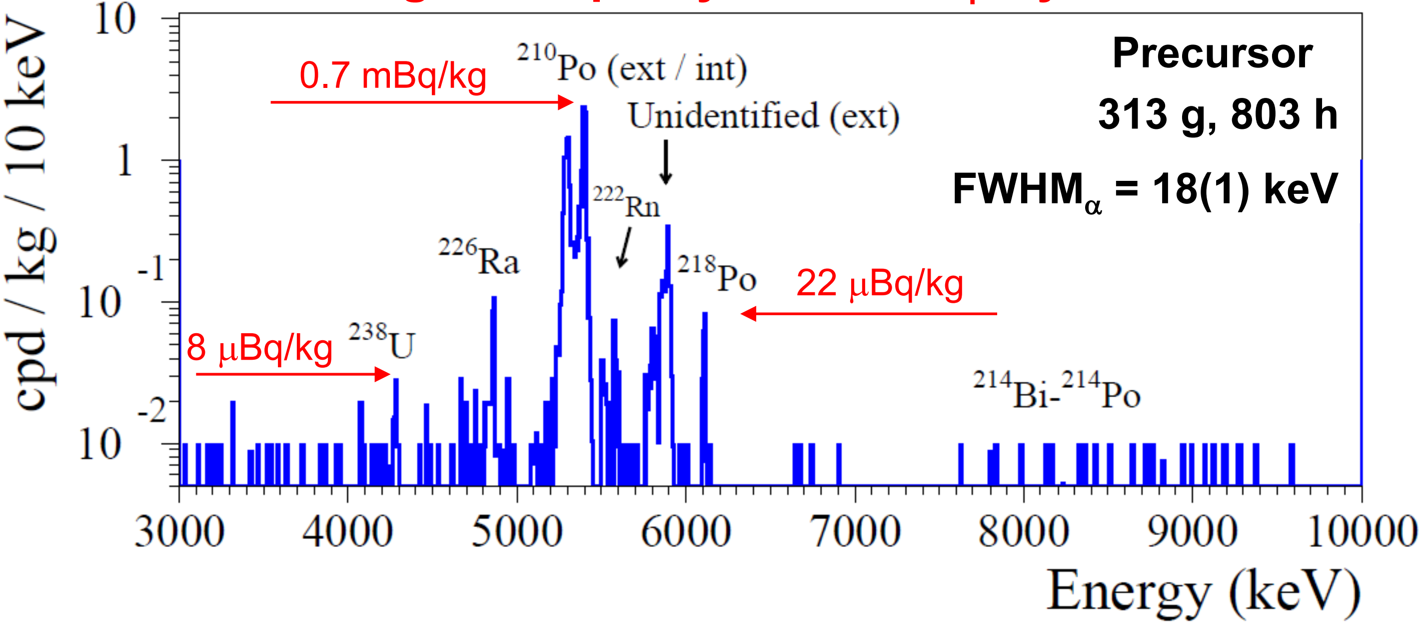


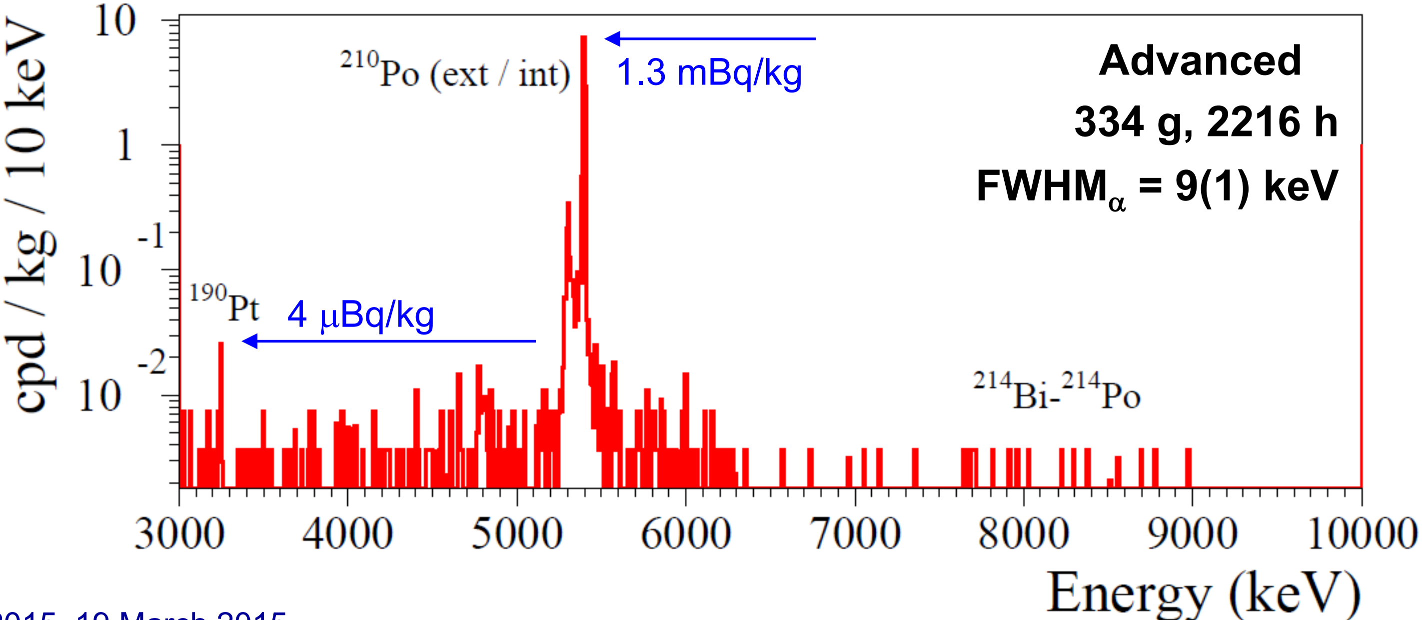
Selected γ(β) and α background



Precursor / Advanced $ZnMoO_4$: α Background

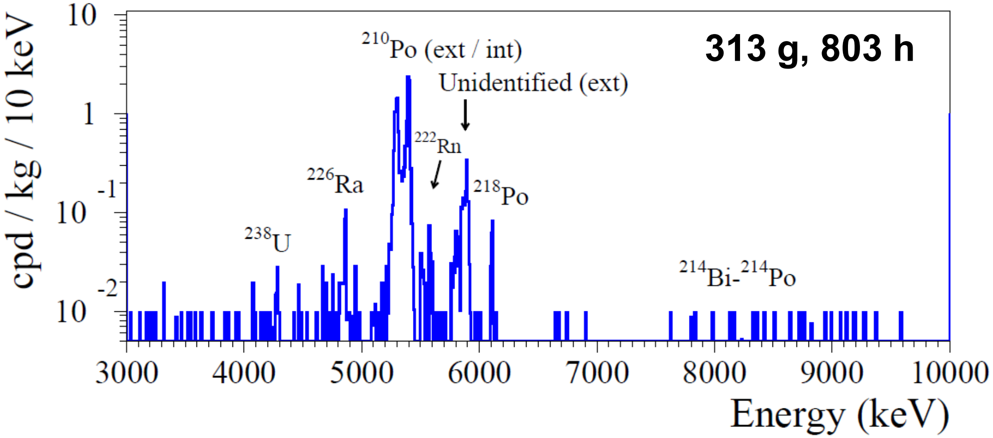
High radiopurity of $ZnMoO_4$ crystals

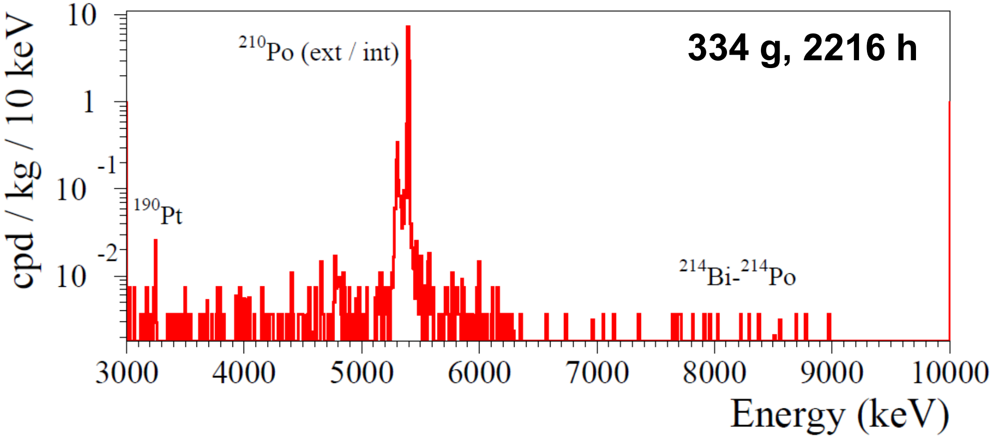




Precursor / Advanced $ZnMoO_4$: Radiopurity

Radiopurity of $ZnMoO_4$ crystal satisfies the LUMINEU requirements





$ZnMoO_4$ radiopurity is even higher than one discussed in [PLB 710(2012)318] for a next-generation $0\nu2\beta$ experiment (^{228}Th / ^{226}Ra ~ 10 μBq/kg)

	Activity (μBq/kg)		
	Advanced	Precursor	
	LSM [1]	LSM [1]	LNGS [2]
	334 g	313 g	329 g
	2216 h	803 h	524 h
^{232}Th	≤ 2	≤ 6	≤ 8
^{228}Th	≤ 5	12(4)	≤ 6
^{238}U	≤ 2	8(3)	≤ 6
^{234}U	≤ 3	≤ 8	≤ 11
^{230}Th	≤ 2	≤ 8	≤ 6
^{226}Ra	≤ 5	22(5)	27(6)
^{210}Po	1271(22)	703(28)	700(30)
^{235}U	≤ 3	≤ 7	–
^{190}Pt	4(1)	≤ 7	–

[1] E. Armengaud et al., submitted to JINST.
[2] J. Beeman et al., EPJC 72 (2012) 2142.

Summary and perspectives

LUMINEU developed technology for producing of high quality large volume $ZnMoO_4$ crystals

- **Two-stage Mo purification is developed** (sublimation, recrystallization from solutions)
- **Large volume colorless $ZnMoO_4$ is produced** (~1 kg, ~80% of initial charge)

R&D of high quality enriched $Zn^{100}MoO_4$ crystals is in progress

- **First enriched $Zn^{100}MoO_4$ is developed** (99.5% of ^{100}Mo, 0.17 kg crystal boule, 84% of initial charge, irrecoverable losses ~ 4%)
- **R&D of high quality large $Zn^{100}MoO_4$ is ongoing** (from ~1 kg of ^{100}Mo)

The technology for the LUMINEU single module is well established

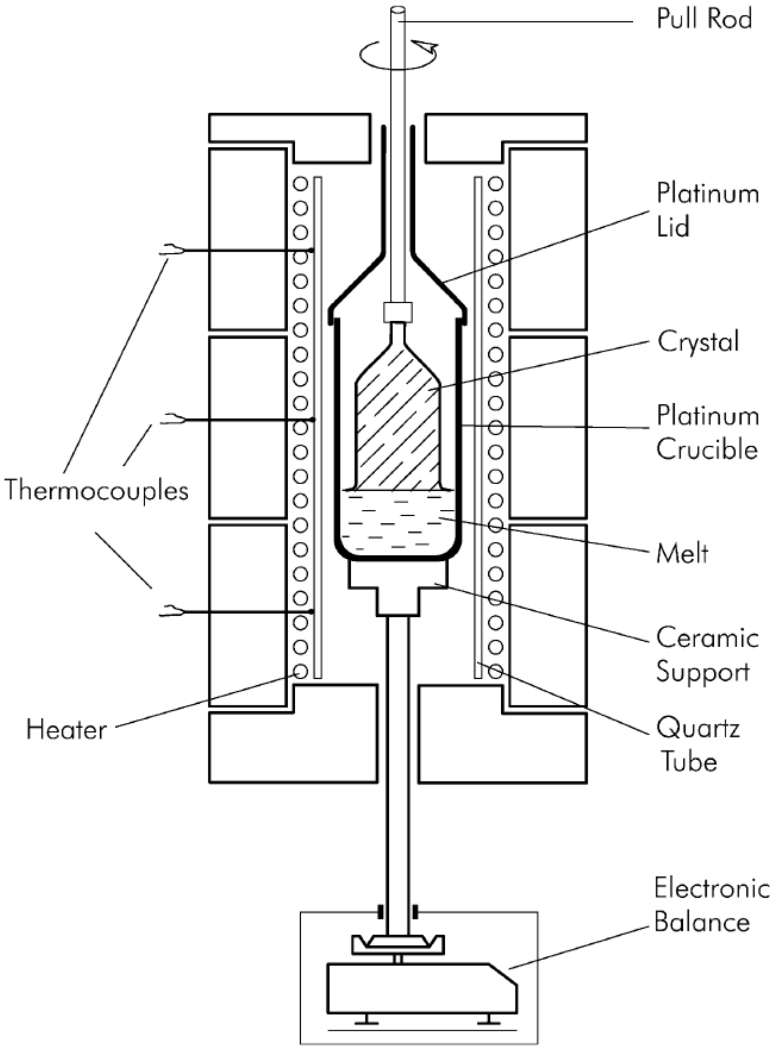
- **Massive $ZnMoO_4$ scintillating bolometers were successfully tested** (~0.3 kg $ZnMoO_4$, ~2500 h in the EDELWEISS set-up at LSM)
- **High performance and full α/γ separation has been achieved** ($FWHM_\gamma$ ~ 4 keV at 356 keV, discrimination power ~ 15σ in 2.5–3.5 MeV)
- **Approved purification procedure allows to reach high radiopurity of $ZnMoO_4$** (e.g. activity of ^{228}Th and ^{226}Ra < 5 μBq/kg)
- **Radiopurity and performance of $ZnMoO_4$ scintillating bolometers satisfy demands of a next-generation cryogenic $0\nu2\beta$ experiment**

LUMINEU extension: LUCINEU $0\nu2\beta$ experiment with ~ 10 kg of ^{100}Mo

- **To prove the technology for a CUORE follow-up next-generation $0\nu2\beta$ project**

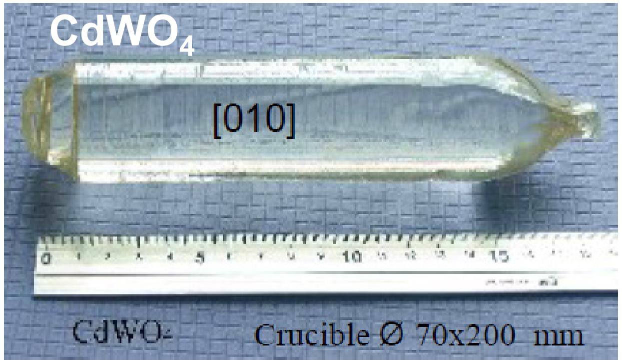
Backup slides

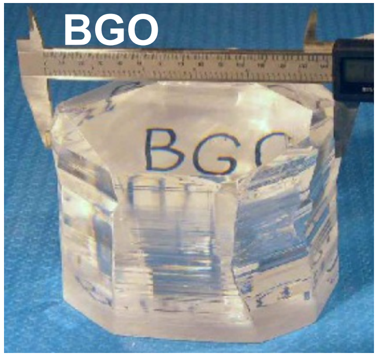
Advanced growth technique: LTG Czochralski

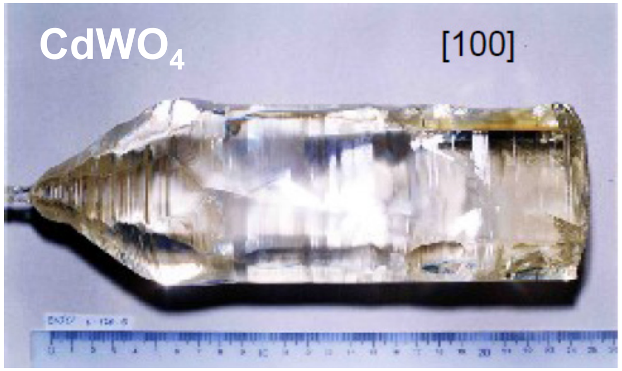


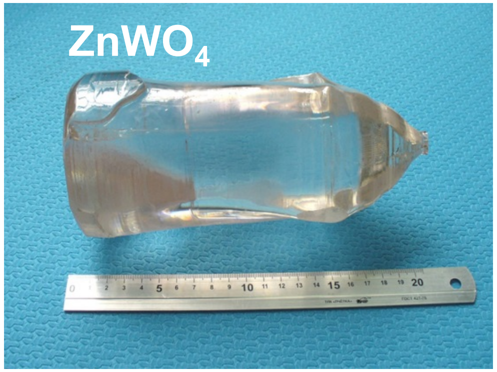
Low-Thermal-Gradient Czochralski
(NIIC, Novosibirsk, Russia)
[JCG 229 (2001) 305]

- **Low temperature gradients** (~ 1.0 K/cm)
- **Crystal inside a crucible during the process**
- **Weighing control at all the stages**
- **Pipe socket as a diffusion barrier** (evaporation and decomposition processes are suppressed ⇒ losses < 1% of charge)
- **Layered growth mechanism** (dominant) and the **faceted crystallization front**
- **Diameter of crystal up to 0.8∅ of crucible** (up to 90% crystal yield from initial compound)





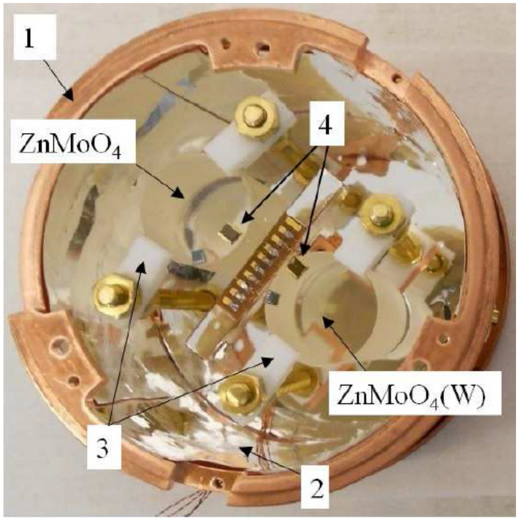




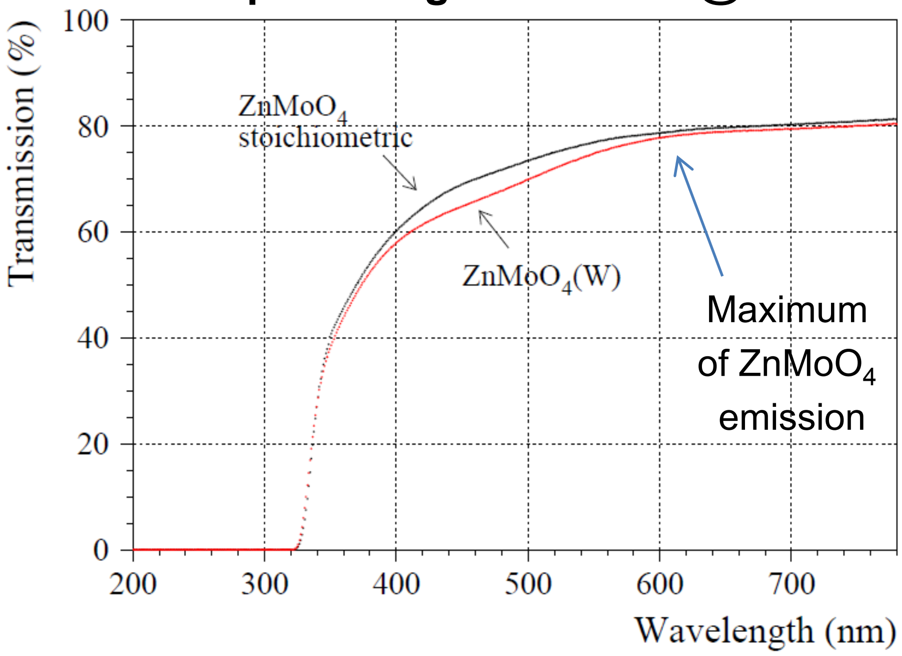
LUMINEU: Effect of W doping on $ZnMoO_4$ properties

W-doped $ZnMoO_4$ grown by LTG Cz

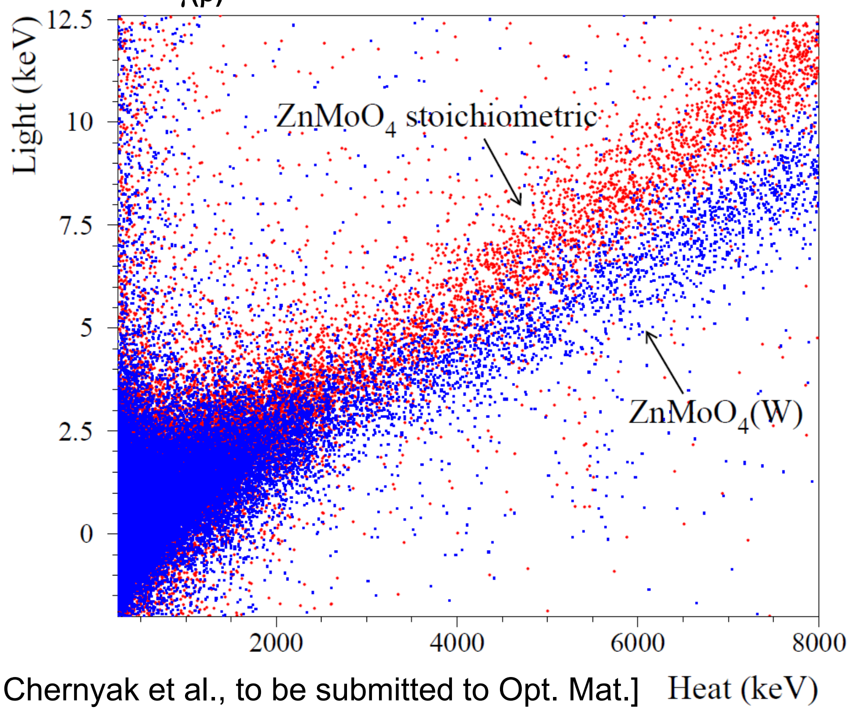
- **Admixture of (0.5–1) mol. % of WO_3 improves quality of $ZnMoO_4$ crystals** (leads to the melt stability and reduces the mechanical stresses)
- **Properties of W-doped $ZnMoO_4$ are similar to stoichiometric crystals** (high transmission, no deterioration of bolometric performance by dopant)



Absorption length = 55 cm^{-1} @ 600 nm

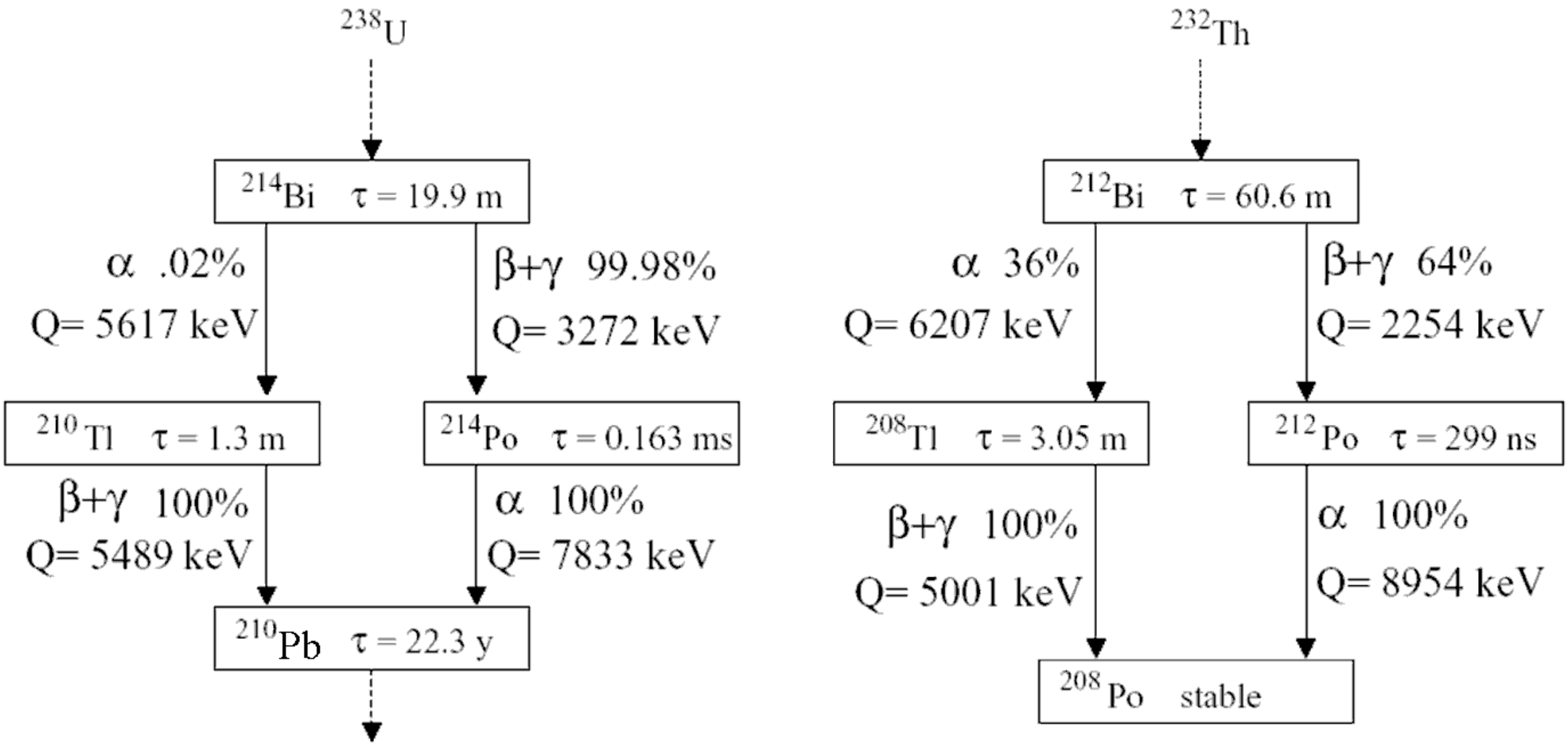


$LY_{\gamma(\beta)}$(W-doped) = 1.08(1) keV/MeV



[D.M. Chernyak et al., to be submitted to Opt. Mat.]

Scintillating bolometer: ^{214,212}Bi-induced Bkg



- **BiPo's totally rejected** (overlapped (β+α)-events)
- **Contribution from ^{210,208}Tl can be suppressed** (identification of α's of ^{214,212}Bi and 10 half-lives vetoing: e.g. 30 m for ^{208}Tl gives suppression 2^{10}=1024 times and negligible dead time, < 1%, if activity of ^{228}Th ~ 0.01 mBq/kg)

Rejection of pile-ups by $ZnMoO_4$ bolometer

Eur. Phys. J. C (2014) 74:2913
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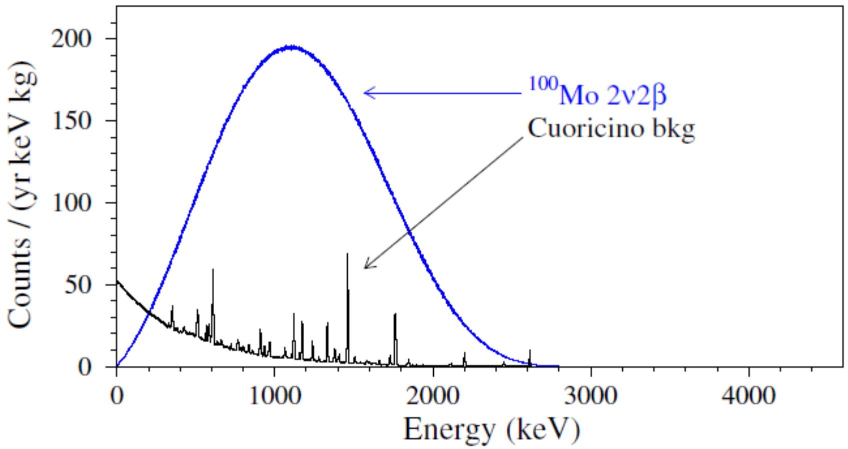
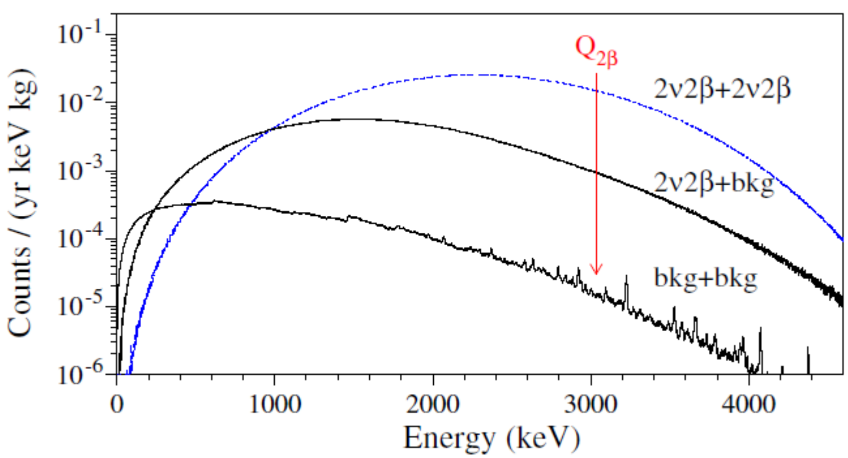
THE EUROPEAN PHYSICAL JOURNAL C

Regular Article - Experimental Physics

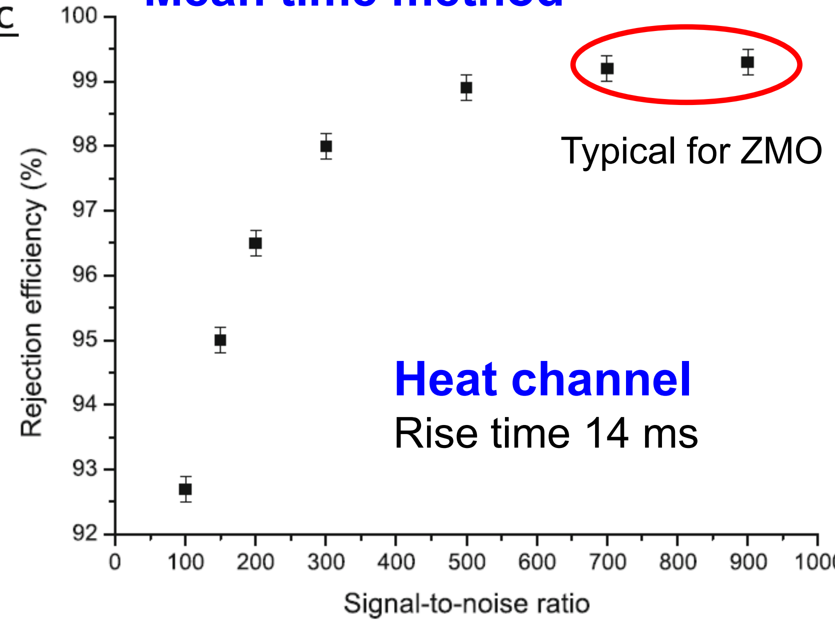
Rejection of randomly coinciding events in $ZnMoO_4$ scintillating bolometers

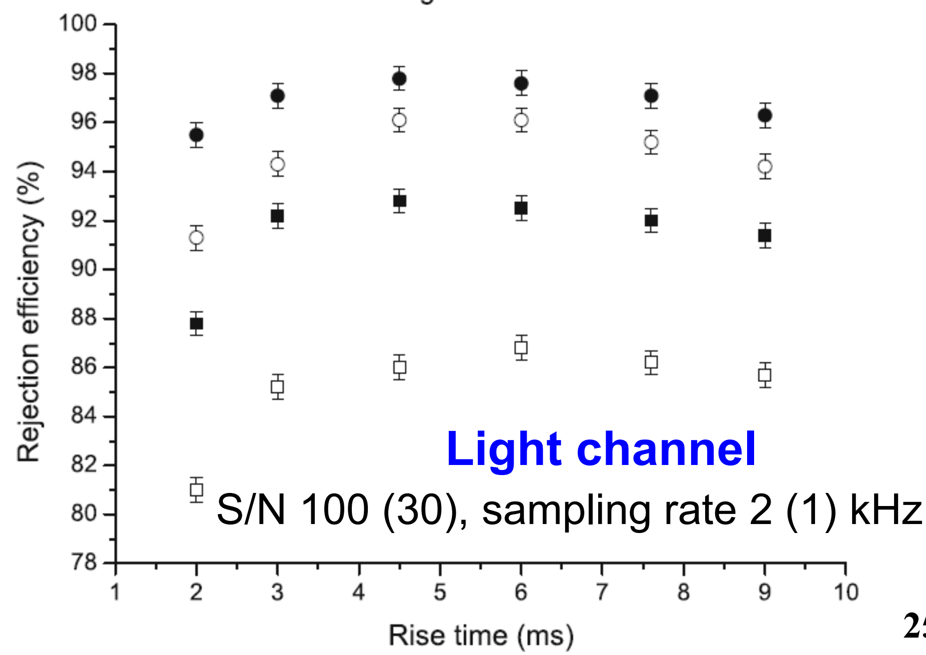
D. M. Chernyak[1,2], F. A. Danevich[1], A. Giuliani[2,3,a], M. Mancuso[2,3], C. Nones[4], E. Olivieri[2], M. Tenconi[2], V. I. Tretyak[1]





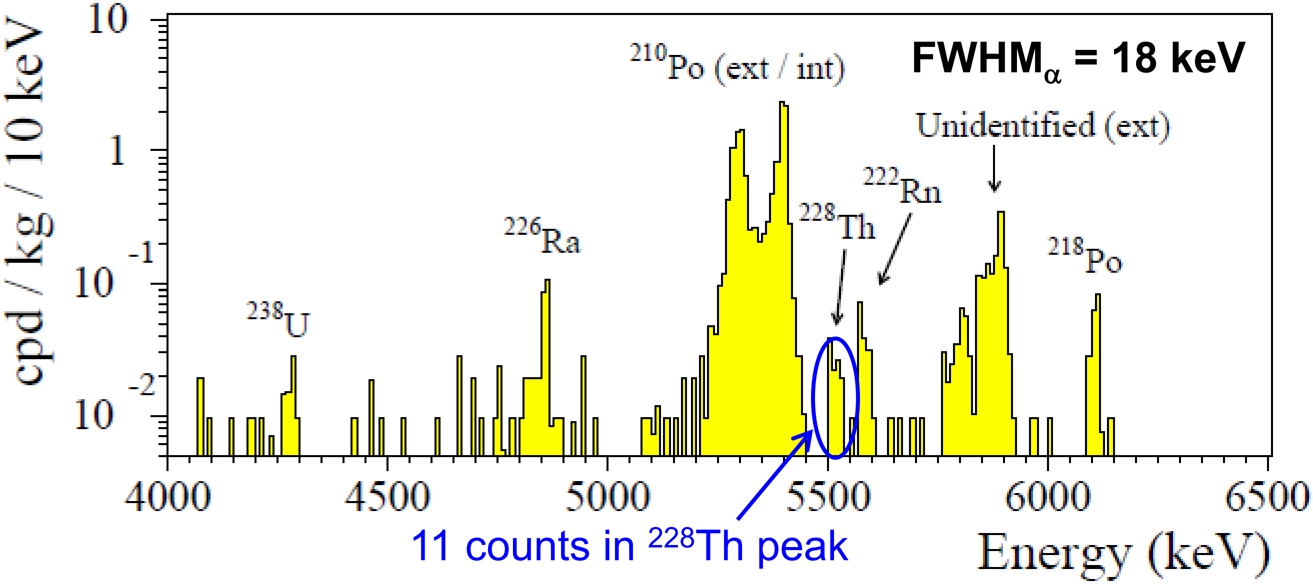
Mean time method

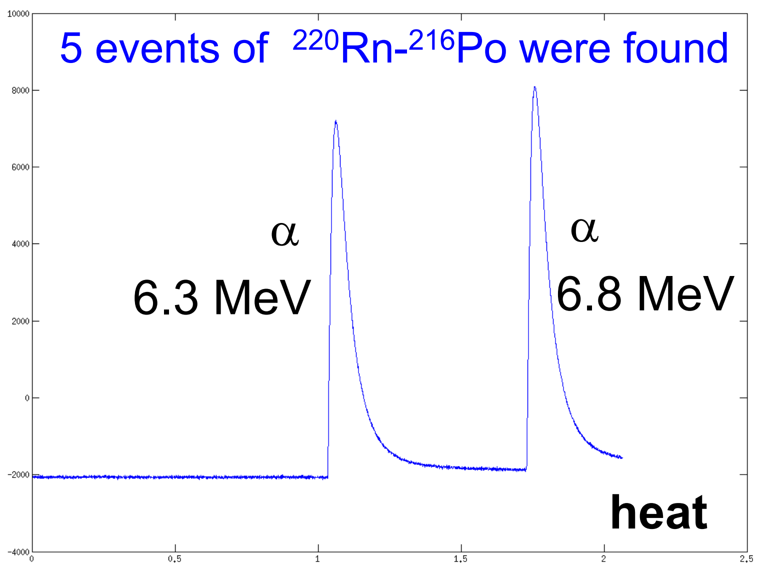


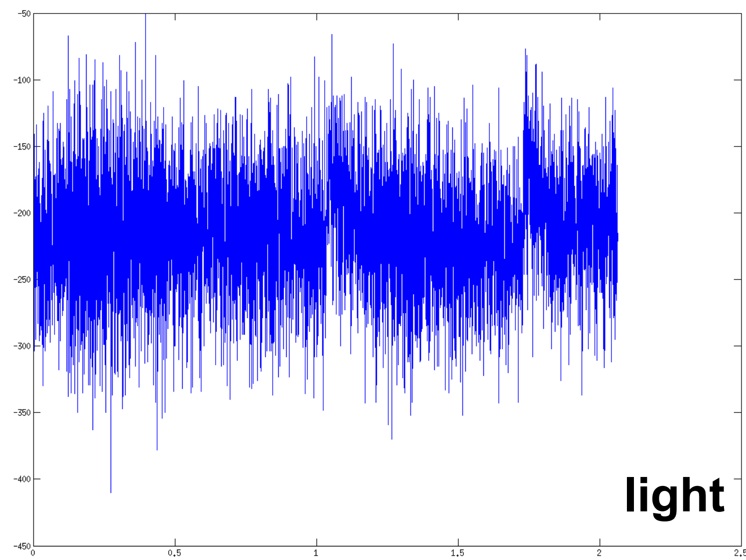


Precursor $ZnMoO_4$: internal ^{228}Th

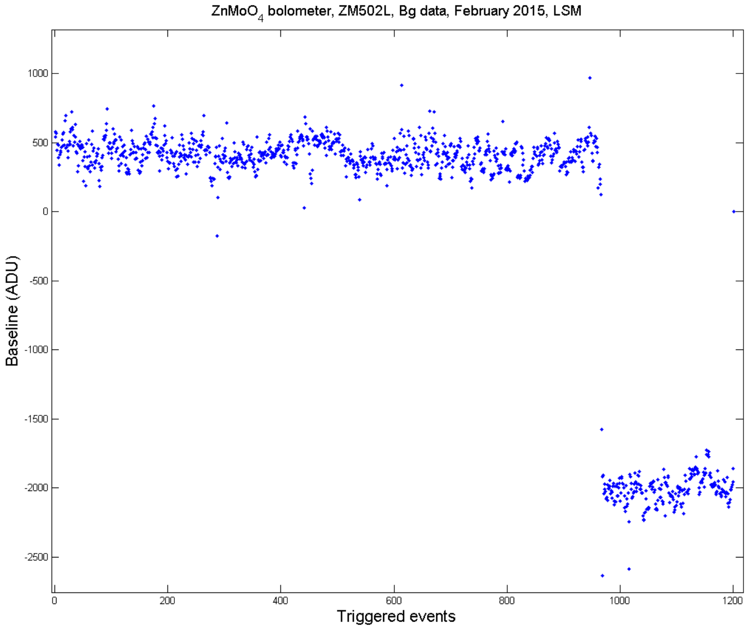
$ZnMoO_4$ 313 g, α spectrum, 803 h, LSM

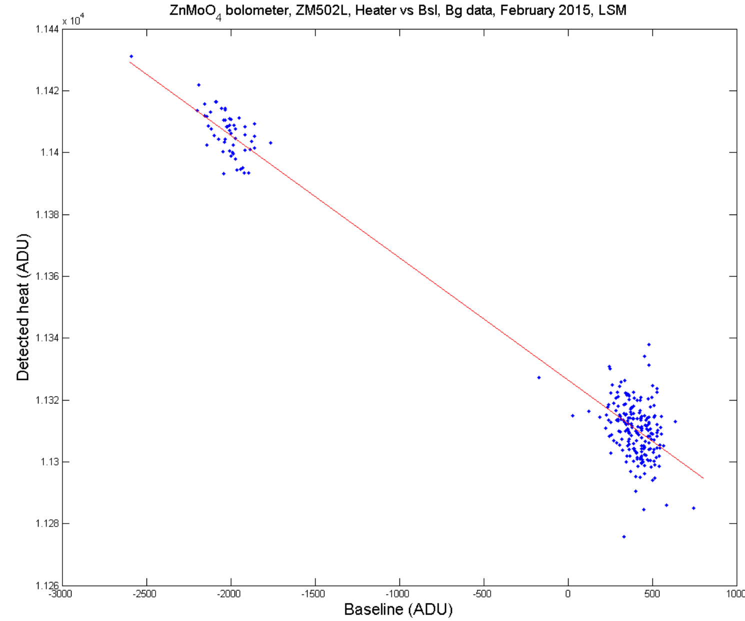


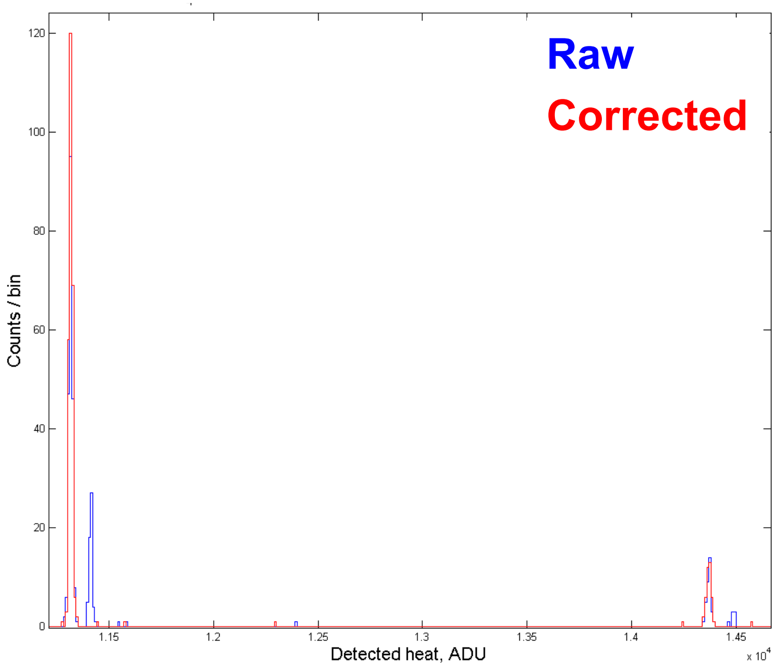


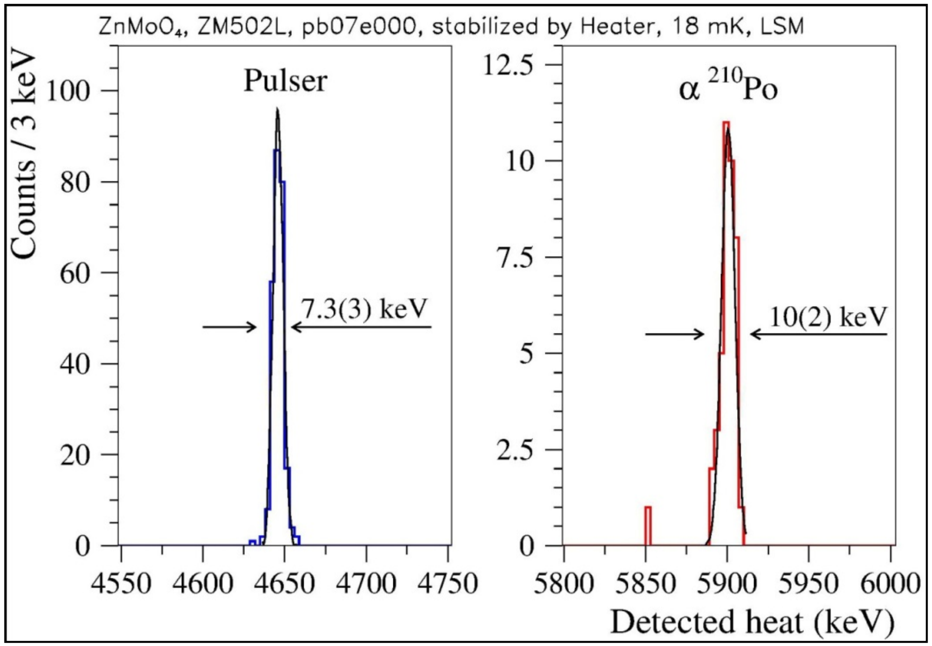


Advanced $ZnMoO_4$: Pulser's performance









Calibration of $Zn^{100}MoO_4$ by ^{232}Th and AmBe