



LAL, Orsay, France, 10 March 2015

Scintillating bolometers for double-beta decay search D.V. Poda

CSNSM, CNRS-IN2P3, Univ. Paris-Sud, 91405 Orsay, France INR, NASU, 03680 Kyiv, Ukraine



About myself

2004–2007: Postgraduate studies at Institute for Nuclear Research, Lepton Physics Department (INR, Kyiv, Ukraine)

Development of CdWO₄, PbWO₄, ZnWO₄ scintillation detectors for DBD search

2009: Candidate of Science Degree (comparable to PhD), INR (Kyiv, Ukraine)

Thesis: "Search for double beta decay of ^{64,70}Zn and ^{180,186}W isotopes"

2008–present: Researcher at INR (Kyiv, Ukraine)

Development of CaMoO₄ and ¹⁰⁶CdWO₄, realization of several DBD experiments (observation of 2v DBD transition of ¹⁰⁰Mo to excited state of ¹⁰⁰Ru)

2010–2012: Postdoc at LNGS, DAMA group (Assergi, Italy)

Development of ${}^{116}CdWO_4$, realization of several small-scale DBD experiments (observation of 2v DBD of ${}^{116}Cd$)

2013–present: **Postdoc at CSNSM**, Solid Physics group, Cryogenic detectors team (Orsay, France)

Development of ZnMoO₄, Zn¹⁰⁰MoO₄ and Li₂MoO₄ scintillating bolometers for DBD search

In total: 11 years research devoted to DBD studies and R&D of detectors based on crystal scintillators for rare event search experiments

Outline

Double Beta Decay

Two neutrino and neutrinoless DBD Present experimental status Choice of DBD isotope and technology

Bolometric technology for DBD search

CUORE and its predecessors with TeO₂

Scintillating bolometers for DBD search

Čerenkov light from TeO_2 R&D of CdWO₄-based detector LUCIFER project with ZnSe AMoRE project with CaMoO₄ LUMINEU and LUCINEU programs (ZnMoO₄ / Li₂MoO₄) CUORE-IHE: beyond CUORE

Summary

Double Beta Decay (Two-neutrino)

SEPTEMBER 15, 1935

PHYSICAL REVIEW

VOLUME 48

Double Beta-Disintegration

M. GOEPPERT-MAYER, The Johns Hopkins University (Received May 20, 1935)

From the Fermi theory of β -disintegration the probability of simultaneous emission of two electrons (and two neutrinos) has been calculated. The result is that this process occurs sufficiently rarely to allow a half-life of over 10¹⁷ years for a nucleus, even if its isobar of atomic number different by 2 were more stable by 20 times the electron mass.



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

- Allowed in the Standard Model
- Second order process in weak interactions (very rare decay rate) $\left[T_{1/2}^{2\nu}\right]^{-1} = G^{2\nu}(Q_{\beta\beta}, Z) \cdot |M^{2\nu}(A, Z)|^2$



Investigation of 2νββ decay

1950: First geochemical evidence (¹³⁰Te, $T_{1/2} = 1.4 \times 10^{21}$ yr) [Phys. Rev. 78 (1950) 822]

1987: First observation in direct experiment (⁸²Se, T_{1/2} = 1.1×10²⁰ yr) [PRL 59 (1987) 1649]

Up to-date: Registered for 11 from 35 potentially $\beta\beta$ -active nuclides [ADNDT 80 (2002) 83] Measured $T_{1/2} \sim 10^{18} - 10^{21}$ yr (~ 10^{24} yr for ¹²⁸Te), see e.g. in [Nucl. Phys. A 935 (2015) 52] Observed $2\nu\beta\beta$ transition to first 0⁺ excited level (for ¹⁰⁰Mo and ¹⁵⁰Nd)



W. H. FURRY Physics Research Laboratory, Harvard University, Cambridge, Massachusetts Fest by using NLDBD

(Received October 16, 1939)

The phenomenon of double β -disintegration is one for which there is a marked difference between the results of Majorana's symmetrical theory of the neutrino and those of the original Dirac-Fermi theory. In the older theory double β -disintegration involves the emission of four particles, two electrons (or positrons) and two antineutrinos (or neutrinos), and the probability of disintegration is extremely small. In the Majorana theory only two particles-the electrons or positrons-have to be emitted, and the transition probability is much larger.



Neutrinoless Double Beta Decay



Schechter-Valle theorem (independent on mechanism) [PRD 25 (1982) 2951]:

• If a $0\nu\beta\beta$ occurs, there must be an effective Majorana mass term

Light Majorana neutrino exchange (standard mechanism) [1–6]

Neutrinos have non-zero masses and Majorana origin

$$\langle \mu \rangle^2 = \frac{\langle m_{\beta\beta} \rangle^2}{m_e^2} - \frac{\langle m_{\beta\beta} \rangle^2}{Effective Majorana neutrino mass} \langle m_{\beta\beta} \rangle^2 = \left| \sum_{i=1}^3 m_i \cdot U_{ei}^2 \right|^2$$

See recent reviews and references therein: [1] S.M. Bilenky, C. Giunti, Int. J. Mod. Phys. A 30 (2015) 1530001. [4] J.J. Gomez-Cadenas et al., Riv. Nuovo Cim. 35 (2012) 29. [2] O. Cremonesi and M. Pavan, AHEP 2014 (2014) 951432.

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[3] J.D. Vergados at al., Rep. Prog. Phys. 75 (2012) 106301. [5] A. Giuliani and A. Poves, AHEP 2012 (2012) 857016. [6] W. Rodejohann, Int. J. Mod. Phys. E 20 (2011) 1833.

Neutrino oscillations





Homestake exp.



The Nobel Prize in Physics 2002



Super-Kamiokande exp.



M. Koshiba

Discovery of neutrino oscillations



See e.g. [Particle Data Group, Chin. Phys. C 38 (2014) 090001]

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Status of $0\nu\beta\beta$ search



- Best current sensitivity: (m_{ββ}) ~ 0.15–0.9 eV, T_{1/2} ~ 10²⁴–10²⁵ yr KamLAND-ZeN (¹³⁶Xe), GERDA-I (⁷⁶Ge), CUORICINO (¹³⁰Te), NEMO-3 (¹⁰⁰Mo)
- Current generation experiments will start probe IH $\langle m_{BB} \rangle \sim 0.05-0.2 \text{ eV}, T_{1/2} \sim 10^{26} \text{ yr}$
- New / advanced technology is needed to cover IH $\langle m_{\beta\beta} \rangle \sim 0.02-0.05 \text{ eV}, T_{1/2} \sim 10^{27} \text{ yr}$

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

Choice of ββ isotope



Choice of experimental technology



High isotopic abundance

enrichment of $\beta\beta$ source

High registration efficiency

"detector = $\beta\beta$ source"

Large detector mass

scalability of a technology

Long-term measurements

operational stability



Extremely low background in ROI

underground conditions massive passive / active shields high radiopurity of a detector

High detector performance

high energy resolution tracking / topology capability background rejection capability fast time response

No technology can satisfy all requirements

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[see e.g. AHEP 2012 (2012) 857016] 10

Bolometric technique



[A. Fleischmann, talk at LUMINEU meeting, Orsay, France, 7-8 Dec. 2014] See also [Cryogenic Particle Detection, Topics Appl. Phys. 99 (2005) 501 pp.]

TeO₂-based bolometric $\beta\beta$ experiments





Radiopurity control protocol to limit bulk and surface contaminations in crystals [J. Cryst. Growth 312 (2010) 2999]

Dedicated runs @ LNGS [Astropart. Phys. 35 (2012) 839]

Nuclide	Allowed	Measured
²³⁸ U	\leq 0.3 ppt	≤ 0.05 ppt
²³² Th	≤ 0.3 ppt	≤ 0.2 ppt
²¹⁰ Pb	\leq 10 μ Bq/kg	\leq 3.3 μ Bq/kg
²¹⁰ Po	≤ 0.1 Bq/kg	≤ 0.05 Bq/kg

Background in CUORICINO and CUORE-0



Scintillating bolometer



ISSN 1063-7788, Physics of Atomic Nuclei, 2006, Vol. 69, No. 12, pp. 2109–2116. © Pleiades Publishing, Inc., 2006.

Proceedings of the 5th International Conference on NONACCELERATOR NEW PHYSICS Double-Beta Decay and Rare Processes

Scintillating Double-Beta-Decay Bolometers*

S. Pirro^{1)**}, J. W. Beeman²⁾, S. Capelli¹⁾, M. Pavan¹⁾, E. Previtali¹⁾, and P. Gorla³⁾

Received November 23, 2005

Abstract—We present the results obtained in the development of scintillating double-beta-decay bolometers. Several Mo and Cd based crystals were tested with the bolometric technique. The scintillation light was measured through a second independent bolometer. A 140-g CdWO₄ crystal was run in a 417-h live time measurement. Thanks to the scintillation light, the α background is easily discriminated, resulting in *zero* counts above the 2615-keV γ line of ²⁰⁸Tl. These results, combined with an extreme easy light detector operation, represent the first tangible proof demonstrating the feasibility of this kind of technique.

Čerenkov light emitted by TeO₂ bolometer



Cadmium tungstate (CdWO₄) as a ββ detector



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16

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CdWO₄ – excellent scintillating bolometer



Development of large enriched 116CdWO₄ crystal



116 , Ø45×14 5 6 7 8 9 10 Activity	CdWO ₄ 47 47 1 12 12 14 15	 Enriched cadmium (¹¹⁶Cd – 82.2%, but ¹¹³Cd ~ 2%) Deep purification of initial materials Low-thermal-gradient Czochralski (LTG Cz) method High crystal yield (87%) Low irrecoverable losses of ¹¹⁶Cd (< 2.3%) Excellent optical and scintillation properties High radiopurity (except ¹¹³Cd / ^{113m}Cd) DBD search in DAMA\R&D at LNGS (since 2011) 					
No.1	No.2	Radiopurity can be further improved by recrystallization					
0.031(3)*	0.054(2)*	A	Activity of	²²⁸ Th (mBc	/kg) in the	crystal boule	
0.5(2)	0.7(2)		10(2)	0.09(1)	0.04(1)	0.02(1)	
≤ 0.005	≤ 0.005						
0.6(2)	0.8(2)			4 3	2	1 U (3 þar	
≤ 0.9	≤ 0.9						
100(10)	100(10)	after the crystal					
460(20)	460(20)	grov	vth			And Hand	
$e \Rightarrow T_{1/2}$ (228	Th) ~ 1.9 yr	Bolometric test is expected					

* - Varies in time $\Rightarrow T_{1/2}$ (²²⁸Th) ~ 1.9 yr

Nuclide

²²⁸Th

238U

²²⁶Ra

²¹⁰Po

⁴⁰K

¹¹³Cd

^{113m}Cd

[JINST 6 (2011) P08011; Rad. Meas. 56 (2013) 66; EPJ WC 65 (2014) 01005]

LUCIFER project

Low-background Underground Cryogenics Installation For Elusive Rates





Co- Investigator: A.Giuliani

Coordinator: S.Pirro



erc



Double Beta Decay pilot project based on scintillating bolometers

The experimental basis for **LUCIFER** is the R&D performed by **S. Piro** at LNGS in the framework **BoLux** (INFN) and **ILIAS-IDEA** (EC WP2-P2)

Crystal	lsotope (Q _{ββ} , keV)	Useful material	LY _{γ(β)} , keV/MeV	
CdWO ₄	¹¹⁶ Cd (2814)	32%	~ 17	
ZnMoO ₄	¹⁰⁰ Mo (3034)	44%	~ 1	
ZnSe	⁸² Se (2996)	56%	~ 7	

Primary solution: ZnSe

- ✓ Higher % of useful material
- ✓ Lower price of enrichment
- ✓ Quite high light yield (essential for PSD)
- ZnSe is well known compound (used as an IR optical material and scintillation detector)

But: High melting point, volatility of Zn and Se, low crystal yield, no commercial use of large size

LUCIFER: performance of large mass ZnSe bolometer





LNGS, AmBe 160 17σ discrimination 0.000 17σ discrimination 0.000 17σ discrimination 0.000 1

LUCIFER: performance of large mass ZnSe bolometer



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21

Energy [keVee]

LUCIFER Schedule





Delay ~1 year due to crystallization issue

[J.W. Beeman et al., AHEP 2013 (2013) 237973]

CaMoO₄ as a detector of $0\nu\beta\beta$ of ¹⁰⁰Mo

2002: Idea, CaMoO₄ growth in Korea
2003: Russian group joined, R&D
2004: 1st talk at VIETNAM'2004, idea of CaMoO₄ cryogenic detector
2005–2007: 1st ISTC project for large CMO crystal growth in Russia
2006: Ukrainian group joined, CMO crystal growth in Ukraine
2007: start R&D of CMO cryogenic technique

2008: 2nd ISTC project for 1 kg enriched ⁴⁰Ca¹⁰⁰MoO₄ crystal growth

2009: AMoRE collaboration formed

with 5 countries

2010–2011: studies ⁴⁰Ca¹⁰⁰MoO₄ properties and radiopurity

2012: ⁴⁰Ca¹⁰⁰MoO₄ production line funded by Russia

2013: AMoRE project fully funded for 10 years by Korea (IBS CUNPA) D.V. Poda, LAL, 10 March 2015



Perspectives for a high sensitivity experiment to search for the $0v2\beta$ decay of ¹⁰⁰Mo are discussed. The energy resolution of 4–5% is enough to reach a sensitivity at the level of 10^{25} yr. The contamination of crystals by ²²⁶Ra and ²³²Th should not exceed the level of 0.1 mBq/kg. The two neutrino 2β decay of ⁴⁸Ca restricts the sensitivity of an experiment to search for the $0v2\beta$ decay of ¹⁰⁰Mo using CaMoO₄ crystal scintillators. A possible solution would be to produce <u>CaMoO₄ scintillators from Calcium depleted in ⁴⁸Ca</u>. A further improvement of sensitivity could be achieved by using CaMoO₄ crystals as scintillating bolometers.

AMoRE: Advanced Mo based Rare process Experiment

8 countries, 18 institutions, ~ 90 collaborators from 🛛 🍋 💳 💳 🚾 💳

~ 1.5 kg of ⁴⁰Ca¹⁰⁰MoO₄ crystals (¹⁰⁰Mo – 96.1%, ⁴⁰Ca – 99.964%, ⁴⁸Ca < 0.001%) produced by Czochralski method at the FOMOS-Materials plant (Moscow, Russia) [1–3]



Radiopurity tested at Yangyang underground laboratory (Korea)



 4π gamma veto system

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	Nuclide	Activity (mBq/kg)					
		SB28	NSB29	S35	SS68	SS81	
	²²⁸ Th	0.07	0.032	0.64	0.027		
C	²²⁷ Ac	-	0.67	1.6	0.24		
	²²⁶ Ra	0.08	0.23	4.5	0.062	1.6 *	

[1] H. Bhang et al., J. Phys. Conf. Ser. 375 (2012) 042023.

[2] J.H. So et al., IEEE Trans. Nucl. Sci. 59 (2012) 2214.

[3] Jungho So, 9th AMoRE Coll. meeting, SNU, Korea, 12-13.02. 2015]

AMoRE: ⁴⁰Ca¹⁰⁰MoO₄ scintillating bolometer



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AMoRE Schedule

	Pilot	Phase I	Phase II
⁴⁰ Ca ¹⁰⁰ MoO ₄ crystals	1.5 kg	10 kg	200 kg
Bkg in ROI (c/keV/kg/yr)	0.01	0.002	0.0002
Sensitivity to T _{1/2} (yr)	>1.1×10 ²⁴	~2×10 ²⁵	~4×10 ²⁶
Sensitivity to $m_{\beta\beta}$ (eV)	0.3–0.9	0.08–0.22	0.016-0.047
Location	Yangyang	Yangyang	Duta Mt. (new)
Schedule	Mar. 2015	Sept. 2016	2019–2023





35 CMO (Ø45×45 mm, 0.3 kg) 5 layers \times 7 columns 5 CMOs: ~ 1.5 kg $^{238}\text{U}/^{232}\text{Th} \sim 0.05 \text{ mBq/kg}$ AMoRE Pilot (2015)

AMoRE10 (2016)

390 CMO (Ø5×6 cm, 0.5 kg) 30 layers \times 13 columns (2.4 m height), ²³⁸U/²³²Th ~ 0.01 mBq/kg

AMoRE200 (2019)



LUM NEU Luminescent Underground Molybdenum

ANR funds; 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 $0\nu\beta\beta$ experiment

- ZnMoO₄ crystal production (R&D of Mo purification and ZMO growth conditions to produce large mass colorless samples from natural and enriched Mo; high radiopurity: ²²⁸Th and ²²⁶Ra ~ 0.01 mBq/kg, total α activity of U/Th ~ 1 mBq/kg)
- Temperature sensors production and optimization (NTD, TES, MMC)
- Light detectors development (based on HP Ge wafers)
- > **A pilot 0** $\nu\beta\beta$ experiment (~1 kg ¹⁰⁰Mo)

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5-yr sensitivity at 90% CL [PLB 710(2012)318] ZnMoO₄ nat. or enriched in ¹⁰⁰Mo to 97% Bkg $\approx 4 \times 10^{-4}$ cnts/keV/kg/yr in 6 keV window

	Number of $pprox 400~{ m g}$ crystals	Total isotope mass [kg]	Half-life sensitivity [10 ²⁵ y]	m _{ββ} sensitivity [meV]
Ľ	4	0.676	0.53	167-476
1	40	6.76	4.95	55-156
	2000 (nat.)	33.1	15.3	31-89
	2000	338	92.5	13-36

NEMO-3:

6.9 kg ¹⁰⁰Mo, 5 yr, 8% FWHM, 10⁻³ c/keV/kg/yr $T_{1/2} (0\nu\beta\beta) > 1.1 \times 10^{24} \text{ yr}, \langle m_{\beta\beta} \rangle < 0.3-0.9 \text{ eV}$ [R. Arnold et al., PRD 89(2014)11101(R)]

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)



Large volume precursor of LUMINEU program





ZnMoO₄ development

- ZnO (99.995%) provided by Umicore (Belgium)
- MoO₃ (99.999%) purified at NIIC (Novosibirsk)
- Platinum crucible \emptyset 8 cm
- LTG Cz crystal growth along the [001] axis [Crystallogr. Rep. 59 (2014) 288]



ZnMoO₄ scintillating bolometer

(1) 313 g ZnMoO₄ crystal grown in NIIC (Novosibirsk, Russia) (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



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	Si K		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	К	Са	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]

First LUMINEU samples: test at CSNSM

ZnMoO₄ development

- MoO₃ purified by double recrystallization from aqueous solutions
- HP ZnO (Umicore)
- LTG Cz crystal growth
- Crystal yield ~ 80%
- Four ZnMoO₄ samples produced (55 g and 160 g)







LUMINEU: first massive ZnMoO₄ bolometers







Technology of high quality large ZnMoO₄ crystal producing is developed

- Molybdenum was purified by using double recrystallization from aqueous solutions
- Advanced quality ZnMoO₄ boule was grown by directional solidification along [001] using LTG Cz (crystal yield ~ 80% of initial charge)
- ZnMoO₄ crystal boule was melted and then crystallized again (to test recrystallization)
- Produced ZnMoO₄ elements have the size expected for a pilot LUMINEU $0\nu\beta\beta$ experiment





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

LUMINEU: First enriched Zn¹⁰⁰MoO₄ detectors



First Zn¹⁰⁰MoO₄ crystal is developed from enriched molybdenum

- ¹⁰⁰MoO₃ (99.5% enrichment in ¹⁰⁰Mo) was purified by using sublimation in vacuum and recrystallization from aqueous solutions
- Zn¹⁰⁰MoO₄ boule was grown at NIIC by using low-thermal-gradient Czochralski process (crystal yield – 84% of initial compound)
- Total irrecoverable losses of $^{100}\text{Mo}~\sim4\%$



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

LUMINEU: Aboveground test of Zn¹⁰⁰MoO₄ array

Zn¹⁰⁰MoO₄ show bolometric properties similar to ZnMoO₄ detectors



LUMINEU: Aboveground test of Zn¹⁰⁰MoO₄ array

Zn¹⁰⁰MoO₄ crystals demonstrate encouraging radiopurity



LUMINEU: Effect of W doping on ZnMoO₄ properties



W-doped ZnMoO₄ grown by LTG Cz

- Admixture of (0.5–1) mol. % of WO₃ improves quality of ZnMoO₄ crystals (leads to the melt stability and reduces the mechanical stresses)
- Properties of W-doped ZnMoO₄ are similar to stoichiometric crystals (high transmission, no deterioration of bolometric performance by dopant)





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[D.M. Chernyak et al., to be submitted to Opt. Mat.] Heat (keV)
LUMINEU: Underground tests at Modane

Laboratoire Souterrain de Modane (LSM)

1.7 km rock overburden (~4.8 km w.e.) 5 μ /day/m²; 10⁻⁶ n/day/cm² (>1 MeV) Deradonized air flow (~30 mBq/m³)



Altitudes 1228 m 1263 m 1298 m Distances 0 m 6210 m 12 868 m

EDELWEISS set-up

Installation at LSM Clean room (ISO Class 4)

Copper cryostat

³He/⁴He table top dilution refrigerator Large experimental volume (50 liters)

Passive shield Low radioactivity lead (min. 20 cm) Polyethylene (min. 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 ZnMoO₄ (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
- Zn¹⁰⁰MoO₄ scintillating bolometers array
- 2 scintillating bolometers based on advanced $ZnMoO_4$ crystals
- June 2014 present







Precursor / Advanced ZnMoO₄: Calibration by ¹³³Ba

Excellent performance of ZnMoO₄ bolometers



Precursor / Advanced ZnMoO₄: Calibration by ²³²Th

Excellent performance of ZnMoO₄ bolometers



Precursor / Advanced ZnMoO₄: α / γ separation



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41

Background measurements with advanced ZnMoO₄

Perfect capability to get "zero" background in the ROI



Precursor / Advanced ZnMoO₄: α **Background**

High radiopurity of ZnMoO₄ crystals



Precursor / Advanced ZnMoO₄: Radiopurity

Radiopurity of ZnMoO₄ crystal satisfies the LUMINEU requirements



Advanced Li₂MoO₄ bolometer: development

Li_2MoO_4 as a perspective $\beta\beta$ detector

- ✓ High concentration of Mo (55% in mass!)
- ✓ Possible scintillating bolometer (but low LY)
- High (n,α) cross-section for ⁶Li (i.a. 8%): no neutron induced events in γ(β) band
- ✓ Comparatively easy crystal growth process (progress in growth 0.1–0.35 kg LMO by Cz)

Successful growth (0.1–0.37) kg LMO by LTG Cz method from deeply purified Mo and commercial Li_2CO_3 (99.99% purity grade)

Property	Value
Density (g/cm ³)	3.02 - 3.07
Melting point (K)	974 ± 2
Hygroscopicity	Weak
Index of refraction	1.44
Radioactive	
contamination (mBq/kg)	
40 K	170(80)
²³² Th	≤ 0.11
²³⁸ U	≤ 0.09



In \approx 4.6 larger than previously tested sample !

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45

Advanced Li₂MoO₄ bolometer: aboveground tests



Advanced Li₂MoO₄ bolometer: test at LNGS



Advanced Li₂MoO₄ bolometer: test at LNGS



LUMINEU schedule and extension

- > 1 or 2 Zn¹⁰⁰MoO₄ crystal(s) \emptyset 5×4 cm (March 2015)
- > 1 or 2 Li_2MoO_4 crystal(s) \emptyset 5×4 cm (March 2015)
- > 1 or 2 $\text{Li}_2^{100}\text{MoO}_4$ crystal(s) \emptyset 5×4 cm (May 2015)
- Tests of bolometric performance and radiopurity
- \succ Choice between ZnMoO₄ and Li₂MoO₄

LUCINEU (joint efforts of LUCIFER and LUMINEU groups)

- □ ~10 kg of ¹⁰⁰Mo previously used in NEMO-3 (MoU IN2P3-ITEP-INFN)
- □ Systematic ¹⁰⁰Mo purification, crystal growth in NIIC
- □ 2016: batch of 20 + 20 Mo-containing crystals
- Experiments at LSM (France) and LNGS (Italy) underground labs

LUCINEU project

LUCINEU goal

- ➤ ~10 kg of ¹⁰⁰Mo (97%)
- \succ 40 Zn¹⁰⁰MoO₄ or Li₂¹⁰⁰MoO₄ (Ø50×40 mm)
- FWHM at ROI: 5+9 keV
- **Bkg in ROI:** \triangleright ~4×10⁻³ counts/keV/kg/yr

> 5-yr sensitivity: $\langle m_{\scriptscriptstyle BB} \rangle \sim 0.05 \text{--} 0.15 \text{ eV}$





Beyond CUORE: CUORE-IHE

The Cryogenic Underground Observatory for Rare Events – Inverted Hierarchy Explorer

http://fsnutown.phy.ornl.gov/fsnufiles/positionpapers/PositionPaper_CUORE-IHE.pdf



Summary

NLDBD is one of the hottest subjects in Astroparticle physics

- Lepton number non-conservation (leptogenesis => matter-antimatter asymmetry)
- 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,..)

Search for NLDBD is a challenging task due to extremely rare rate

Continuous enormous efforts to increase sensitivity (fighting with background, increasing of source's mass towards to the ton-scale, improving of detector's performance, using costly enriched materials, long-term experiments)

Scintillating bolometer is an advanced technology for a high sensitivity NLDBD experiment capable to explore the inverted hierarchy

- ➢ Prospects of "zero"-background NLDBD experiment (isotopes with Q_{ββ} above 2.6 MeV, high energy resolution FWHM=0.1÷0.5%, excellent α/γ discrimination, high radiopurity e.g. ²²⁸Th and ²²⁶Ra < 5 µBq/kg in ZnMoO₄)
- Several ~10 kg demonstrators are developing simultaneously with different isotopes to validate the technology (LUCIFER, AMORE, LUMINEU => LUCINEU)
- > AMoRE is funded to 200-kg scale (the projected sensitivity is in the IH region)
- CUORE-IHE is a possible 1-ton scale experiment with this technology (with the aim to cover the IH neutrino mass pattern)

Backup slides

Next-generation ZnMoO₄-based $\mathbf{0}\nu\beta\beta$ experiment

[Phys. Lett. B 710 (2012) 318]

Monte Carlo basis

Detectors:

2000 Zn¹⁰⁰MoO₄ crystals (~0.4 kg each; ¹⁰⁰Mo ~ 97%) PTFE clamps (6x for ZMO) Cylindrical Cu holders Ge-based light detectors (\emptyset 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

Software:

COSMO (3 months activation aboveground and 1 yr cooling underground) DECAY0 (event generator) GEANT4 package

Source of backgro	ound Activity	[µBq/kg] E	SKG [counts/(keV k	gy)]
²⁰⁸ Tl in ZMO	10	3	8.2×10^{-3}	
²¹⁴ Bi in ZMO	10	3	10^{-8}	
²²⁸ Th in Cu	20	1	$.6 \times 10^{-5}$	
²²⁶ Ra in Cu	70	1	$.3 \times 10^{-7}$	
²²⁸ Th in PTFE	100	2	2×10^{-7}	
²²⁶ Ra in PTFE	60	<	< 10 ⁻⁹	
⁵⁶ Co in ZMO	0.06	1	$.8 \times 10^{-5}$	
⁵⁶ Co in Cu	0.02	8	10^{-6}	
⁸⁸ Y in ZMO	0.3	7	1×10^{-7}	
Total		3	3.2×10^{-3}	
 Neutron's < 10⁻⁴ Muon's ~ 10⁻⁴ (+ can be reduced by muon veto) Pile-ups of 2vββ of ¹⁰⁰Mo ~ 3×10⁻⁴ (+ possible reduction by PSD) Total Background in ROI: ~ 4×10⁻⁴ counts/keV/kg/yr 				
Number of	Total	Half_life	maa	-
$\sim 400 \text{ g}$	isotope	concitivity	sonsitivity	,
$\sim 400 \text{ g}$	isotope	10^{25} m		′
crystals	mass [kg]	[10 ²⁰ y]	[mev]	_
4	0.676	0.53	167-476	
40	6.76	4.95	55-156	
2000 (nat.)	33.1	15.3	31-89	
2000	338	92.5	13-36)

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



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

Specific background of bolometric DBD experiment

Eur. Phys. J. C (2012) 72:1989 DOI 10.1140/epjc/s10052-012-1989-y THE EUROPEAN Physical Journal C

Letter

Random coincidence of $2\nu 2\beta$ decay events as a background source in bolometric $0\nu 2\beta$ decay experiments \Im_{40}^{st}

D.M. Chernyak^{1,2}, F.A. Danevich¹, A. Giuliani^{2,a}, E. Olivieri², M. Tenconi², V.I. Tretyak¹

¹Institute for Nuclear Research, MSP, 03680 Kyiv, Ukraine ²Centre de Spectrométrie Nucléaire et de Spectrométrie de Masse, 91405 Orsay, France



Table 2 Counting rate of two randomly coincident $2\nu 2\beta$ events in cryogenic Zn⁸²Se, ⁴⁰Ca¹⁰⁰MoO₄, Zn¹⁰⁰MoO₄, ¹¹⁶CdWO₄, and TeO₂ detectors of 100 cm³ volume. Enrichment of ⁸²Se, ¹⁰⁰Mo, and ¹¹⁶Cd is assumed to be 100 %, while for Te the natural isotopic abundance (34.08 %) is taken. *C* is the mass concentration of the isotope of inter-

est, ρ is the density of the material (g/cm³), N is the number of 2β candidate nuclei in one detector, and $B_{\rm rc}$ is the counting rate at $Q_{2\beta}$ (counts/(keV·kg·yr)) under the assumption of 1 ms time resolution of the detector

Isotope	$T_{1/2}^{2\nu 2\beta}$ (yr) [27]	Detector (ρ)	С	Ν	$B_{\rm rc}$
⁸² Se	9.2×10^{19}	Zn ⁸² Se (5.65)	55.6 %	2.31×10^{24}	5.9×10^{-6}
¹⁰⁰ Mo	7.1×10^{18}	$^{40}Ca^{100}MoO_4$ (4.35)	49.0 %	1.28×10^{24}	3.8×10^{-4}
		$Zn^{100}MoO_4$ (4.3)	43.6 %	1.13×10^{24}	2.9×10^{-4}
¹¹⁶ Cd	2.8×10^{19}	¹¹⁶ CdWO ₄ (8.0)	31.9 %	1.32×10^{24}	1.4×10^{-5}
¹³⁰ Te	6.8×10^{20}	TeO ₂ (5.9)	27.2 %	0.76×10^{24}	1.1×10^{-8}

Rejection of pile-ups by ZnMoO₄ bolometer



Sublimation purification



Purification by sublimation

- Sublimation of molybdenum oxide is widely used in the industry of molybdenum
- Nevertheless the concentration of impurities, particularly of W (up to 0.5wt% even in the high purity grade materials) still exceeds the ZnMoO₄ crystal growth requirements
- ➢ We have developed a technique of molybdenum purification by sublimation of MoO₃ in vacuum (with addition of zinc molybdate ZnMoO₄ + WO₃ → ZnWO₄ + MoO₃↑)
- The technique is expected to be efficient to remove Th and U

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



Sublimation at NIIC (Novosibirsk, Russia)

- The temperature up to 700°C
- Vacuum pump
- Initial material MoO₃ (impurities 10-100 ppm)
- Single entry sublimation reduces the content of W, Fe, Cr more than one order magnitude
- Productivity the laboratory setup 1–2 kg/week

Recrystallization from solutions

Molybdenum in aqueous solutions $MoO_3 + 2NH_4OH = (NH_4)_2MoO_4 + H_2O$





[V.N. Slegel, talk at RPSCINT 2013]

Development of Zn¹⁰⁰MoO₄ crystal boule

[A.S. Barabash et al., EPJC 74 (2014) 195]

- Purification of ¹⁰⁰MoO₃ in two stages sublimation in vacuum: recrystallization from aqueous solutions:
- ¹⁰⁰MoO₃ (132 g, ¹⁰⁰Mo is 99.5%)
- ZnO (72 g, 99.995% purity, UMICORE)
- Zn¹⁰⁰MoO₄ powder (204 g) was obtained by solid-phase synthesis
- Zn¹⁰⁰MoO₄ boule (171 g) was grown in Pt crucible Ø40×100 mm by using lowthermal-gradient Czochralski technique 20 rot/min (beginning) → 4 rot/min (end) temperature gradient ≤ 1 °C/cm
- The yield of crystal boule is 84%
- Irrecoverable loses of ¹⁰⁰Mo at all stages is 4% (compatible with results of ^{106,116}CdWO₄ crystals developing [1,2])

[1] P. Belli et al., NIMA 615 (2010) 301.

[2] A.S. Barabash et al., JINST 06 (2011) P08011.

D.V. Poda, LAL, 10 March 2015

Table 1 Contamination of ${}^{100}MoO_3$ measured by inductively coupled plasma mass-spectrometry (ICP-MS) and atomic absorption spectroscopy (AAS) methods.

Element	Concentra in ¹⁰⁰	Concentration of element in ¹⁰⁰ MoO ₂ (ppm)		
	ICP-MS	AAS		
Na	_	< 60		
Mg	< 0.5	< 4		
Al	2.4	_		
Si	_	< 500		
Κ	< 15	< 10		
\mathbf{Ca}	_	< 10		
V	0.05	_		
Cr	0.2	< 5		
Mn	0.1	_		
\mathbf{Fe}	8	< 5		
Ni	0.01	_		
Cu	0.1	_		
Zn	0.1	< 4		
Ag	0.3	_		
W	1700	550		
Pb	0.008	_		
Th	< 0.0005	_		
U	0.001	_		

Table 2 Irrecoverable losses of enriched molybdenum in all the stages of $\text{Zn}^{100}\text{MoO}_4$ crystal scintillator production.

Stage	Loss
Sublimation of $^{100}MoO_3$	1.4%
Recrystallization from aqueous solutions	2%
Crystal growth	0.6%
Total	4%

60

Raw data: Heat and Light signals

313 g ZnMoO₄ bolometer at LSM



61

Data treatment by Optimum Filter technique $H(\omega) = k \times S^{*}(\omega) / N(\omega) \times exp\{i \ \omega \ t_{max}\}$



Zn¹⁰⁰MoO₄: Calibration by ²³²Th and AmBe



Precursor / Advanced ZnMoO₄: α **Background**



Precursor ZnMoO₄: internal ²²⁸Th

ZnMoO₄ 313 g, α spectrum, 803 h, LSM



Advanced ZnMoO₄: pulser's performance



Advanced ZnMoO₄: pulser's performance



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LUMINEU: development of light detectors

Optimisation of the way to held the detector



Nomex wires



Kevlar wires

+ Optimisation of the NTD thermal coupling





12 thin \emptyset 6 μ m wires Sapphire sphere \emptyset 1.5 mm

Performance of optical bolometers tested at CSNSM

	Optical bolometer	Size (mm)	Sensitivity (µV/keV)	FWHM noise (keV)
Developed within < the LUCIFER project	>LT8	Ø 50 × 0.250	0.3	0.56
	→LT9	Ø 50 × 0.250	0.4	0.28
Developed @ CSNSM <	→ M1	Ø44 × 0.250	1.0	0.40
	→ M3	Ø44 × 0.250	2.5	0.65
Developed @ IAS (new sapphire pat held)	[→] M4	Ø44 × 0.250	0.9	0.27
	\rightarrow IAS	Ø40 × 0.043	6.6	0.07
Developed @ CSNSM —	→ Luke	Ø44 × 0.250	0.7	0.50 (0 ∨)
D.V. Poda, LAL, 10 March 2015			17.5	0.02(05 V)

LUMINEU: Neganov-Luke effect assisted LD



Photons

- \rightarrow creates e-h pairs
- \rightarrow e-h pairs are drifted by electric field
- \rightarrow Phonon emission while e-h pairs drift

$$\rightarrow$$
 Amplification $E_{heat} = H$

$$E(1+\frac{qV}{\varepsilon})$$

Luke LD, LED pulses at 0 V and 53.17 V, 16.7 mK, CSNSM





- HP Ge LD (Ø50×0.2 mm) with annual Al electrodes
- NTD Ge (3×1.5×0.6 mm)
- Hamamatsu LED

High impedance Nb_xSi_{1-x} TES for LUMINEU



Background in ROI: Effect of α rejection



7000

6000

²³²Th

LUCIFER: ZnMoO₄-based bolometers


ZnSe crystal production



Deeply purified materials

- 15 kg of ⁸²Se (95.5%) by URENCO (Netherland) 6 kg of ⁸²Se grains are stored @ LNGS
- 15 kg of HP Zn grains by NSC KIPT (Ukraine) all amount is stored @ LNGS

	Activity, mBq/kg				
	²²⁸ Th	²²⁶ Ra	⁷⁵ Se	⁶⁵ Zn	
⁸² Se	< 0.27	< 0.3	0.19(6)	-	
HP Zn	< 0.04	< 0.07	-	5.2(6)	

Zn⁸²Se crystal production

- Crystal production in ISC (Kharkiv, Ukraine) (by Bridgman growth technique)
- R&D for natural ZnSe crystal production (should be finished in Jan. 2015)
- First enriched Zn⁸²Se crystal growth (planned to be in Feb. 2015)
- A few days for 1 kg Zn⁸²Se synthesis (3 synthesis/day + annealing in H₂ within 2 days)
- > Almost one year for whole amount of $Zn^{82}Se$ (1 furnace × 3 crystals/month \Rightarrow 36 crystals/yr)

[F. Orio, talk at 4th ISOTTA meeting, Orsay, France, 1-2.12.2014]

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Czochralski and Bridgman growth techniques



AMoRE: CaMoO₄-based prototype



AMoRE: Geant4 Monte Carlo simulations

Background source	Activity [µBq/kg]	Bg [10 ⁻⁴ cnt/keV/kg/yr]	Bg reduced by PSD [10 ⁻⁴ cnt/keV/kg/yr]
TI-208, internal	10 (²³² Th)	0.36	0.36
TI-208, in Cu	16 (²³² Th)	0.22	0.22
BiPo-214, internal	10	0.11 ¹⁾	≤ 0.01
BiPo-214, in Cu	60	1.8 ^{1) 2)}	≤ 0.18
BiPo-212, internal	10 (²³² Th)	0.08 1)	≤ 0.01
BiPo-212, in Cu	16 (²³² Th)	0.36 ^(1) 2)	≤ 0.04
Y-88, internal	20	0.19	0.19
Random 2v2β	8.7×10 ³⁾	3 .1 ³⁾	1.2
Total		6.2	≤ 2.2

1) Can be reduced x0.1 by alpha/beta PSD

2) Can be reduced by Teflon coating of Cu (to remove surface α)

3) Can be reduced by pulse-shape discrimination

Muon background @Y2L : ~1.4e⁻⁴ cnt/keV/kg/yr

[F.A. Danevich, 4th ISOTTA meeting]

CRESST-II: Radiopurity of CaWO₄ scint. bolometer



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77 recoil energy [keV]