



**HAL**  
open science

## Enriched $\text{Zn}^{100}\text{MoO}_4$ scintillating bolometers to search for $0\nu 2\beta$ decay of $^{100}\text{Mo}$ with the LUMINEU experiment

A. S. Barabash, D. M. Chernyak, F. A. Danevich, A. Giuliani, I. M. Ivanov, E. P. Makarov, M. Mancuso, S. Marnieros, S. G. Nasonov, C. Nones, et al.

### ► To cite this version:

A. S. Barabash, D. M. Chernyak, F. A. Danevich, A. Giuliani, I. M. Ivanov, et al.. Enriched  $\text{Zn}^{100}\text{MoO}_4$  scintillating bolometers to search for  $0\nu 2\beta$  decay of  $^{100}\text{Mo}$  with the LUMINEU experiment. European Physical Journal C: Particles and Fields, 2014, 74 (11), 3133 (7 p.). 10.1140/epjc/s10052-014-3133-7 . hal-01108483

**HAL Id: hal-01108483**

**<https://hal.science/hal-01108483>**

Submitted on 7 Aug 2024

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Distributed under a Creative Commons Attribution 4.0 International License

# Enriched $\text{Zn}^{100}\text{MoO}_4$ scintillating bolometers to search for $0\nu 2\beta$ decay of $^{100}\text{Mo}$ with the LUMINEU experiment

A. S. Barabash<sup>1</sup>, D. M. Chernyak<sup>2,3</sup>, F. A. Danevich<sup>2</sup>, A. Giuliani<sup>3,4,5,a</sup>, I. M. Ivanov<sup>6</sup>, E. P. Makarov<sup>6</sup>, M. Mancuso<sup>3,4</sup>, S. Marnieros<sup>3</sup>, S. G. Nasonov<sup>6</sup>, C. Nones<sup>7</sup>, E. Olivieri<sup>3</sup>, G. Pessina<sup>5</sup>, D. V. Poda<sup>2,3</sup>, V. N. Shlegel<sup>6</sup>, M. Tenconi<sup>3</sup>, V. I. Tretyak<sup>2,8</sup>, Ya. V. Vasiliev<sup>6</sup>, M. Velazquez<sup>9</sup>, V. N. Zhdankov<sup>10</sup>

- <sup>1</sup> Institute of Theoretical and Experimental Physics, 117218 Moscow, Russia  
<sup>2</sup> Institute for Nuclear Research, MSP, Kiev 03680, Ukraine  
<sup>3</sup> Centre de Sciences Nucléaires et de Sciences de la Matière, 91405 Orsay, France  
<sup>4</sup> Dipartimento di Scienza e Alta Tecnologia dell'Università dell'Insubria, 22100 Como, Italy  
<sup>5</sup> INFN, Sezione di Milano Bicocca, 20126 Milan, Italy  
<sup>6</sup> Nikolaev Institute of Inorganic Chemistry, 630090 Novosibirsk, Russia  
<sup>7</sup> Service de Physique des Particules, CEA-Saclay, 91191 Gif-sur-Yvette, France  
<sup>8</sup> INFN, Sezione di Roma "La Sapienza", 00185 Rome, Italy  
<sup>9</sup> CNRS, Université de Bordeaux, ICMCB, 33608 Pessac, France  
<sup>10</sup> CML Ltd., 630090 Novosibirsk, Russia

Received: 5 July 2014 / Accepted: 16 October 2014 / Published online: 29 October 2014  
© The Author(s) 2014. This article is published with open access at Springerlink.com

**Abstract** The LUMINEU project is a demonstrator experiment that will search for the neutrinoless double beta decay of the isotope  $^{100}\text{Mo}$  embedded in zinc molybdate ( $\text{ZnMoO}_4$ ) scintillating bolometers. In this context, a zinc molybdate crystal boule enriched in  $^{100}\text{Mo}$  to 99.5 % with a mass of 171 g was grown for the first time by the low-thermal-gradient Czochralski technique. The production cycle provided a high yield (the crystal boule mass was 84 % of the initial charge) and an acceptable level—around 4 %—of irrecoverable losses of the costly enriched material. Two crystals of 59 and 63 g, obtained from the enriched boule, were tested above ground at millikelvin temperatures as scintillating bolometers. They showed a good detection performance, equivalent to that of previously developed natural  $\text{ZnMoO}_4$  detectors. These results pave the way to future sensitive searches based on the LUMINEU technology, capable of approaching and exploring the inverted hierarchy region of the neutrino mass pattern.

## 1 Introduction

Neutrinoless double beta ( $0\nu 2\beta$ ) decay is a crucial process in particle physics since it provides the only experimentally viable method to ascertain the Majorana nature of the neutrino, performing in the meantime a sensitive test of the lep-

ton number conservation. In addition, it has the potential to establish the absolute neutrino mass scale and to give information as regards the hierarchy of the neutrino masses [1–9]. In particular, one of the main objectives of next-generation  $0\nu 2\beta$  decay experiments is to explore the inverted hierarchy region of the neutrino mass pattern. This implies reaching a lifetime sensitivity of the order of  $10^{26}$ – $10^{27}$  years, which requires a background level close to zero in the ton  $\times$  year exposure range.

A powerful technology to achieve this outstanding performance uses scintillating bolometers, to study isotopes with a  $0\nu 2\beta$   $Q$ -value greater than 2.6 MeV, corresponding to the end-point of the bulk of natural  $\gamma$  radioactivity. They would therefore benefit from a significantly reduced  $\gamma$  background in the  $0\nu 2\beta$  signal region. Close-by sources or internal contamination can still produce  $\gamma$  or  $\beta/\gamma$  events above 2.6 MeV, but this tiny component can be kept effectively under control with a proper selection of radiopure materials. The dominant background is expected to be given by energy-degraded  $\alpha$  particles, which, however, can be rejected thanks to the different scintillation-to-heat ratio between  $\alpha$  and  $\beta$  particles [10–12].

The LUMINEU project [13] (Luminescent Underground Molybdenum Investigation for NEUtrino mass and nature) aims at developing scintillating bolometers based on zinc molybdate ( $\text{ZnMoO}_4$ ) crystals to study the isotope  $^{100}\text{Mo}$  ( $Q$ -value = 3034 keV [14]). The project will be capable of reaching the performance required to explore the inverted

<sup>a</sup> e-mail: andrea.giuliani@csnsm.in2p3.fr

hierarchy region, as shown for the first time in Ref. [15] and subsequently confirmed in Ref. [16]. A critical step in the LUMINEU path is the growth of high-quality radiopure large-mass (300–500 g)  $\text{ZnMoO}_4$  monocrystals. As far as non-isotopically enriched material is concerned, the required crystallization technology for a sensitive  $0\nu 2\beta$  decay experiment has been established, and individual detectors—prefiguring the single modules of future large arrays—have already shown to be able to reach the desired bolometric performance and intrinsic radiopurity levels [15, 17–21].

However, the crucial step of reproducing these excellent preliminary results with molybdenum enriched in  $^{100}\text{Mo}$  above the natural 9.7 % [22] is not trivial for two main reasons. On one hand, the initial chemical and radioactive purity levels of enriched samples, normally poorer than those characterizing natural material, may conflict with the construction of well performing bolometric detectors. This problem was observed for instance in  $\text{TeO}_2$  bolometers [23]. On the other hand, enriched material is very expensive and its procurement represents the highest cost factor in future searches. Therefore, the purification/crystallization chain must imply negligible irrecoverable losses of the  $0\nu 2\beta$  decay candidate isotope. This strong requirement is not a priori compatible with all the purification and crystal-growth technologies. In this letter, we show for the first time that both potential obstacles are actually overcome in the  $\text{ZnMoO}_4$  case, adding a further essential element in favor of the use of the LUMINEU technology for future  $0\nu 2\beta$  decay searches.

## 2 Production of a $\text{Zn}^{100}\text{MoO}_4$ crystal boule

Molybdenum enriched in the isotope  $^{100}\text{Mo}$  up to 99.5 % was used to develop zinc molybdate crystals. The enriched molybdenum was produced at the Kurchatov Institute in the 1980s of the last century (former Soviet Union). Approximately 1 kg of the material in form of metal, belonging to the Institute for Nuclear Research (Kyiv, Ukraine) and now available for the LUMINEU program, was utilized in an experiment at the Modane Underground Laboratory (France) to search for double beta decay of  $^{100}\text{Mo}$  to excited states of  $^{100}\text{Ru}$  [24]. Afterwards, in order to improve its purity level, the metallic sample of enriched  $^{100}\text{Mo}$  was dissolved in 20 % ultrapure nitric acid and transformed into molybdenum acid ( $^{100}\text{MoO}_3 \cdot n\text{H}_2\text{O}$ ). After rinsing the compound by nitric acid solution and annealing, a sample of 1199 g of purified molybdenum oxide ( $^{100}\text{MoO}_3$ ) was obtained and used in the ARMONIA experiment [25]. The purification procedure has effectively removed the pollution in  $^{40}\text{K}$  and  $^{137}\text{Cs}$  by one order of magnitude. The concentrations of thorium and radium were also decreased by factors 2 and 4, respectively. The following radioactive contamination of the  $^{100}\text{MoO}_3$  sample can be derived from the data of the experi-

**Table 1** Contamination of  $^{100}\text{MoO}_3$  measured by inductively coupled plasma mass-spectrometry (ICP-MS) and atomic absorption spectroscopy (AAS) methods

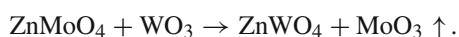
Element	Concentration of element in $^{100}\text{MoO}_3$ (ppm)	
	ICP-MS	AAS
Na	–	<60
Mg	<0.5	<4
Al	2.4	–
Si	–	<500
K	<15	<10
Ca	–	<10
V	0.05	–
Cr	0.2	<5
Mn	0.1	–
Fe	8	<5
Ni	0.01	–
Cu	0.1	–
Zn	0.1	<4
Ag	0.3	–
W	1700	550
Pb	0.008	–
Th	<0.0005	–
U	0.001	–

ment (in mBq/kg): 2.0(2) for  $^{226}\text{Ra}$  ( $^{238}\text{U}$  chain), 0.8(1) for  $^{228}\text{Th}$  ( $^{232}\text{Th}$  chain), 5.9(1) for  $^{137}\text{Cs}$ , and 36(2) for  $^{40}\text{K}$  [25].

The level of impurities in the  $^{100}\text{MoO}_3$  was also measured by inductively coupled plasma mass-spectrometry (ICP-MS) and atomic absorption spectroscopy (AAS) methods. The results of these measurements, presented in Table 1, as well as radioactive contamination of the sample, suggest that the material should be additionally purified to be used for crystal scintillator production. For example, the concentration of the isotope  $^{228}\text{Th}$  in  $\text{ZnMoO}_4$  crystals to be used in future  $0\nu 2\beta$  decay searches must not be higher than  $10 \mu\text{Bq/kg}$  in order to approach the required zero background conditions mentioned in the Sect. 1 [15].

A two-stage technique of molybdenum purification, consisting of sublimation of molybdenum oxide in vacuum and recrystallization from aqueous solutions by co-precipitation of impurities on zinc molybdate sediment, was applied to purify the enriched molybdenum. The purification procedure is reported in detail in Ref. [21].

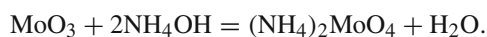
Sublimation was carried out with the addition of zinc oxide to reduce the concentration of tungsten:



The residuals after the sublimation process were analyzed at the analytical laboratory of the Nikolaev Institute of Inor-

ganic Chemistry with the help of atomic emission spectrometry. They contain Fe (0.05 wt%), Si (0.6–0.8 wt%), Mo (11–12 wt%), W (22–33 wt%) and Zn (12–16 wt%). It should be stressed that the analysis confirmed a contamination by iron in the initial  $\text{MoO}_3$ . The sublimates were then annealed in air atmosphere to obtain yellow color stoichiometric  $\text{MoO}_3$ . Losses of enriched molybdenum at this stage of purification did not exceed 1.4 %.

After the sublimation process, the molybdenum oxide contained needlelike granules up to a few millimeters long. The presence of these granules (which cause difficulties to synthesize zinc molybdenum compound) is one more argument in favor of applying an additional stage of purification, consisting of recrystallization of the obtained molybdenum oxide in aqueous solution. For this purpose, the molybdenum oxide was dissolved in ammonia solution at room temperature using zinc molybdate as a collector:



The molybdenum losses during recrystallization from aqueous solutions are expected to be recovered in conditions of mass production. Therefore, we have estimated the losses at this stage taking into account a purification process of about 20 kg of natural molybdenum performed in the framework of the LUMINEU program and its follow-up. The losses do not exceed 2 %.

Powdered  $\text{Zn}^{100}\text{MoO}_4$  (203.98 g) was obtained by solid-phase synthesis of high-purity zinc oxide (72.23 g, produced by Umicore) and purified enriched molybdenum oxide  $^{100}\text{MoO}_3$  (131.75 g). The mixture of the two compounds was kept at a temperature of  $\approx 680^\circ\text{C}$  in a platinum cup over 12 h.

A zinc molybdate crystal boule from the enriched  $^{100}\text{Mo}$  compound was grown by the low-thermal-gradient Czochralski technique [26–28] in a platinum crucible of size  $\varnothing 40 \times 100$  mm. The crystal was grown at a rotation speed of 20 rotations per minute at the beginning of the process, decreasing to 4 rotations per minute at the end. The temperature gradient did not exceed  $1^\circ\text{C}/\text{cm}$ .

The mass of the crystal boule, shown in Fig. 1, is of 170.7 g. Some coloration of the crystal (in contradiction with the practically colorless samples produced from natural molybdenum [20]) can be explained by remaining traces of iron in the powder used for the growth. It should be noted that the initial  $^{100}\text{MoO}_3$  was contaminated by iron at the level of 8 ppm. Furthermore, the special set of lab-ware used for purification of the small amount of enriched material was not perfectly clean. We hope to improve substantially the optical properties of the enriched crystals in the course of the R&D now in progress. It is expected that the increase of the purification-cycle scale should improve the quality of the  $\text{Zn}^{100}\text{MoO}_4$  compound. In any case, as will be demonstrated in the next section, even the present optical quality of the enriched crystal scintillators is high enough to use the mate-



**Fig. 1** Boule of  $\text{Zn}^{100}\text{MoO}_4$  single crystal with mass of 170.7 g and length of 95 mm grown by the low-thermal-gradient Czochralski process

**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 $^{100}\text{MoO}_3$	1.4
Recrystallization from aqueous solutions	2
Crystal growth	0.6
Total	4

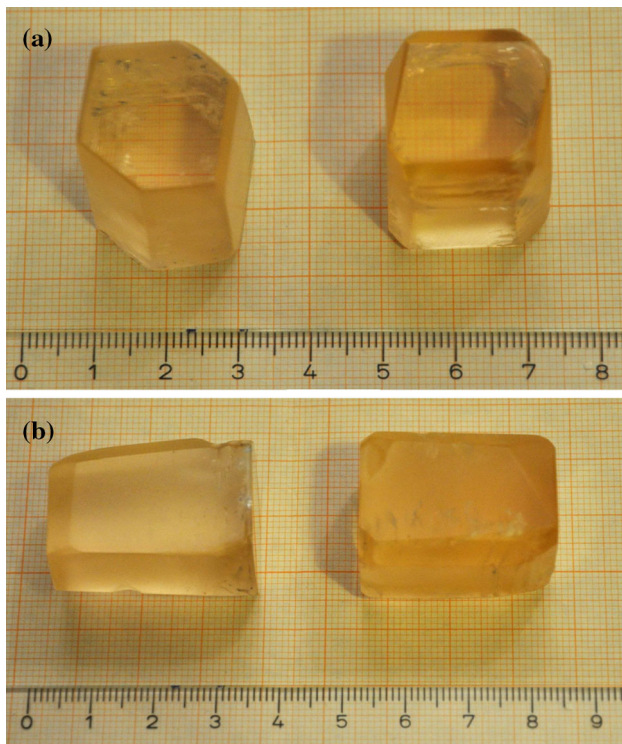
rial for the development of high-performance scintillating bolometers.

The yield of the crystal boule is 83.7 %. This efficiency is unachievable in the ordinary Czochralski method (maximum 30–45 %). It should be noted that some amount of  $\text{Zn}^{100}\text{MoO}_4$  crystal ( $\approx 1.3$  g) remained in the seed. Taking into account the mass of the residual of the melt after the growth process (30.83 g) one can estimate that the losses in the crystal-growth process are about 0.6 %.

The data on losses of enriched molybdenum in all the stages of crystal scintillator production are summarized in Table 2. The amount of losses is comparable to that of enriched cadmium in the production of cadmium tungstate crystal scintillators from  $^{106}\text{Cd}$  (2.3 %) [29] and  $^{116}\text{Cd}$  ( $\approx 2$  %) [30]. An R&D program to decrease the losses of enriched molybdenum by a factor 1.5–2 by optimization of all the stages of the production process is in progress.

### 3 Fabrication and operation of two enriched $\text{Zn}^{100}\text{MoO}_4$ scintillating bolometers

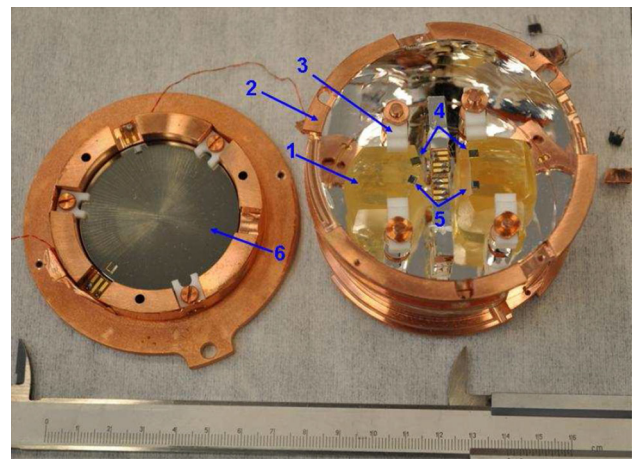
Two samples with a similar size ( $\text{Zn}^{100}\text{MoO}_4$ , top, and  $\text{Zn}^{100}\text{MoO}_4$ , bottom, with masses of 59.2 and 62.9 g, respectively) were produced from the  $\text{Zn}^{100}\text{MoO}_4$  boule shown in



**Fig. 2** Top (a) and side (b) views of the zinc molybdate single crystals obtained from material enriched in  $^{100}\text{Mo}$  to 99.5 %. The  $\text{Zn}^{100}\text{MoO}_4$ , top, sample (see text), at the left, has a mass of 59.2 g and the  $\text{Zn}^{100}\text{MoO}_4$ , bottom, sample, at the right, of 62.9 g

Fig. 1. The sample  $\text{Zn}^{100}\text{MoO}_4$ , top, corresponds to the top part of the boule, close to its ingot, and characterized by a less intense orange color (corresponding probably to fewer chemical impurities and a lower defect concentration). A photograph of the produced crystal elements is shown in Fig. 2. The two samples are only approximately rectangular. Their shape is irregular as it was decided to keep their mass as high as possible in order to make the bolometric tests more significant. We just took the precaution to get two flat parallel bases in order to facilitate holding the crystals by polytetrafluoroethylene (PTFE) elements in the bolometer construction.

The two  $\text{Zn}^{100}\text{MoO}_4$  crystals were used to assemble an array of scintillating bolometers. Each sample was equipped with a neutron transmutation doped (NTD) Ge thermistor for the read-out of the thermal signals. The thermistors have a mass of about 50 mg, a resistance at 20 mK of  $\sim 500$  k $\Omega$  and a logarithmic sensitivity  $A = -d \log(R)/d \log(T) \simeq 6.5$ . They were attached at the crystal surface by using six epoxy glue spots and a 25  $\mu\text{m}$  thick Mylar spacer (which was removed after the gluing procedure). In addition, each crystal was provided with a heating element glued by means of one epoxy spot, consisting of a resistive meander of heavily doped silicon with a low-mobility metallic behavior down to very low temperatures. The purpose of this heater is to provide periodically a fixed amount of thermal energy in order to



**Fig. 3** Photograph of the assembled  $\text{Zn}^{100}\text{MoO}_4$  bolometer array together with the photodetector: (1)  $\text{Zn}^{100}\text{MoO}_4$  crystals with mass of 59.2 g (left) and 62.9 g (right); (2) copper holder of the detectors with the light reflecting foil fixed inside; (3) PTFE supporting elements; (4) NTD Ge thermistors; (5) heating devices; (6) light detector

control and stabilize the thermal response of the  $\text{Zn}^{100}\text{MoO}_4$  bolometers.

The samples were assembled inside a copper holder by using PTFE elements. A light detector (LD) [31], made of a  $\varnothing 50 \times 0.25$  mm high-purity Ge disk and instrumented with an NTD Ge thermistor was mounted  $\approx 2$  mm from the top face plane of the  $\text{Zn}^{100}\text{MoO}_4$  crystals to collect the emitted scintillation light. The inner surface of the copper holder was covered by a reflecting foil (VM2000, VM2002 by 3M) to improve light collection. The  $\text{Zn}^{100}\text{MoO}_4$  scintillating bolometer array, before mounting of the LD, is shown in Fig. 3.

The array was tested at very low temperatures by using a pulse-tube cryostat housing a high-power dilution refrigerator [32] installed at the CSNSM (Orsay, France). The cryostat is surrounded by a massive shield made out of low-activity lead to minimize pile-up effects, which are particularly disturbing in aboveground measurements with slow detectors such as bolometers based on NTD Ge thermistors.

Room-temperature low-noise electronics, consisting of DC-coupled voltage-sensitive amplifiers [33] and located inside a Faraday cage, was used for the read-out of the NTD Ge thermistors. Data streams were recorded by a 16 bit commercial ADC with 10 kHz sampling frequency. The test was performed at three base temperatures: 13.7 mK (over 18.3 h), 15 mK (over 4.8 h), and 19 mK (over 24.2 h). The last working point was chosen to simulate the typical temperature conditions expected in the EDELWEISS set-up [34], an apparatus to search for dark matter which will be available for our next test deep underground in the Modane underground laboratory (France).

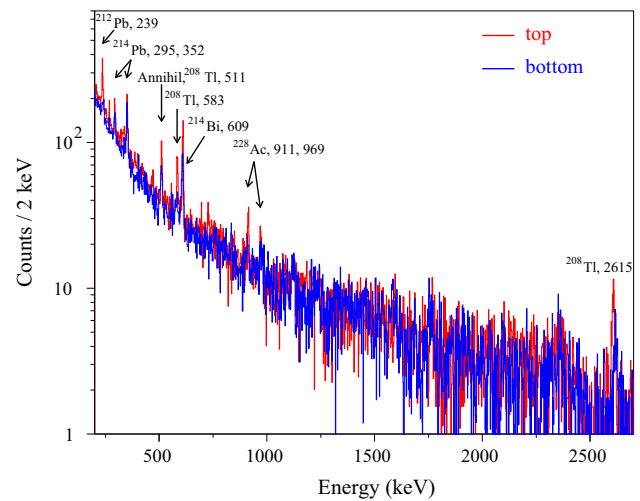
Several experimental parameters were evaluated for each set of measurements in order to extract information as regards

**Table 3** Experimental parameters (see text) for the  $Zn^{100}MoO_4$  scintillating bolometers array registered in aboveground measurements at 13.7 mK (first row) and 19 mK (second row). An event distribution within energy ranges of 500–3000 keV and 10–30 keV for the  $Zn^{100}MoO_4$  bolometers and for the LD, respectively, were used to evaluate the parameters  $\tau_R$  and  $\tau_D$

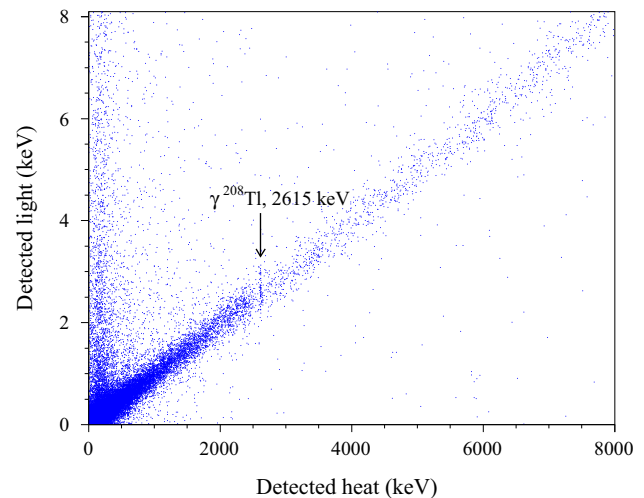
Detector	$R_{bol}$ (M $\Omega$ )	$A_{signal}$ ( $\mu$ V/MeV)	FWHM $_{bsl}$ (keV)	$\tau_R$ (ms)	$\tau_D$ (ms)
Top	1.54	86.8	1.4(1)	9.0	46.3
	1.17	65.0	1.8(1)	8.9	48.4
Bottom	1.82	95.8	1.8(1)	5.5	26.2
	1.35	84.2	2.4(1)	5.8	30.7
LD	0.97	409	0.28(1)	2.5	14.8
	0.81	336	0.37(2)	2.5	15.5

the bolometric performance of the  $Zn^{100}MoO_4$  array. In particular, we have registered the thermistor resistance ( $R_{bol}$ ) at the working temperature, the signal amplitude ( $A_{signal}$ ) for a unitary deposited energy, the full width at half maximum baseline width (FWHM $_{bsl}$ ), and pulse rise ( $\tau_R$ ) and decay ( $\tau_D$ ) times. These last two parameters were computed from 10 to 90 % and from 90 to 30 % of the signal maximum amplitude, respectively. An overview of the detector performance at 13.7 and 19 mK in terms of the mentioned parameters is provided in Table 3. Similar values were obtained with the  $Zn^{100}MoO_4$  detectors cooled down to 15 mK, but these results are omitted here due to the very short duration of this measurement.

The optimum filter procedure [35], typically used for the analysis of bolometric data, was applied to extract the amplitudes of each recorded signal. The calibration of the LD was performed by using a weak  $^{55}Fe$  source. The energy resolution (FWHM) of the LD at 5.9 keV of  $^{55}Fe$  was 0.42(2) keV at 13.7 mK and 0.57(4) keV at 19 mK. The energy scale of the  $Zn^{100}MoO_4$  bolometers was determined by means of a low-activity  $^{232}Th$  source and  $\gamma$  quanta from natural radioactivity (mainly  $^{226}Ra$  daughters). The energy spectra accumulated by the  $Zn^{100}MoO_4$  detectors operated at 13.7 mK are shown in Fig. 4. The energy resolution for the detectors at 2614.5 keV of  $^{208}Tl$  was FWHM = 11(3) keV for the  $Zn^{100}MoO_4$ , top, and FWHM = 15(3) keV for the  $Zn^{100}MoO_4$ , bottom, in the measurements at 13.7 mK. As one can see from Fig. 4, the 2614.5 keV peaks have quite low statistics due to the small mass of the crystals and the short duration of the measurements. Therefore, it is reasonable to estimate the resolution of the detectors for the more intensive  $\gamma$  lines presented in the spectra below 1 MeV. For example, the FWHM at 609.3 keV peak of  $^{214}Bi$  was 5.0(5) and 10(1) keV for the  $Zn^{100}MoO_4$  top and bottom, respectively. Experience with large-mass slow bolometric detectors shows that the energy resolution on the  $\gamma$  lines is significantly worsened by pulse pile-up. We expect therefore that the energy



**Fig. 4** The energy spectrum accumulated in aboveground measurements over 18.3 h by two  $Zn^{100}MoO_4$  scintillating bolometers (*top* and *bottom*) mounted in one holder. The detector was operated at 13.7 mK and irradiated by low-active  $^{232}Th$  source and environmental  $\gamma$ s. The energy of  $\gamma$  peaks are in keV

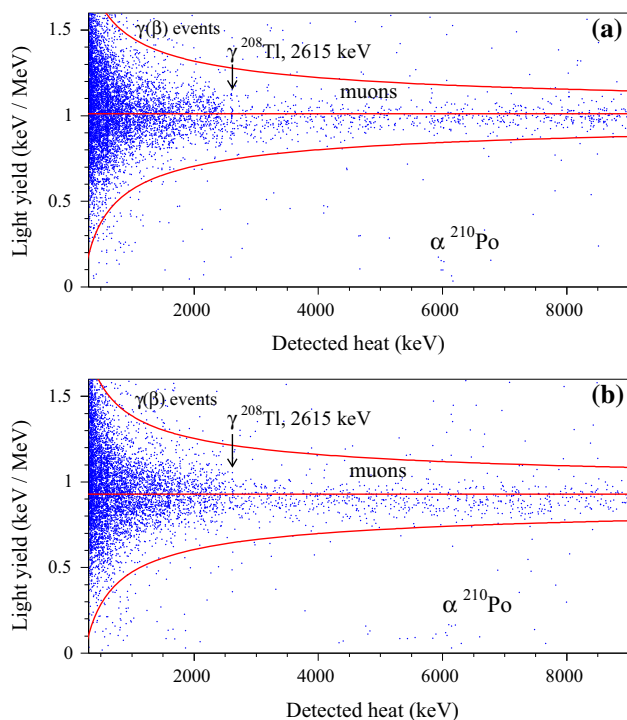


**Fig. 5** The scatter plot of light versus detected heat based on the 13.7 mK data accumulated with the scintillating bolometer  $Zn^{100}MoO_4$ , *top*, during 18.3 h of calibration measurements. The band populated by  $\gamma(\beta)$  events (below 2.6 MeV) and cosmic muons is clearly visible

resolution in an underground set-up is much closer to the FWHM baseline width and definitely better than 10 keV, as already observed in underground-operated natural  $ZnMoO_4$  bolometers [17, 19].

Plots reporting the light-to-heat signal amplitude ratio as a function of the heat signal amplitude for the data accumulated over 18.3 h in the aboveground set-up with the enriched  $Zn^{100}MoO_4$  detectors are presented in Fig. 5.

The data allow us to estimate the light yield (the amount of detected light energy per particle energy measured by the deposited heat) related to  $\gamma(\beta)$  events ( $LY_{\gamma(\beta)}$ ). For instance, the distribution of the  $LY_{\gamma(\beta)}$  versus the detected heat accu-



**Fig. 6** The light yield as a function of the detected heat in aboveground measurements on scintillating bolometers based on  $\text{Zn}^{100}\text{MoO}_4$ , *top* (a) and  $\text{Zn}^{100}\text{MoO}_4$ , *bottom* (b) crystals. The data were accumulated at 13.7 mK. The events above 2.6 MeV in the  $\gamma(\beta)$  region are caused by cosmic muons. The position of the  $\alpha$  events related to  $^{210}\text{Po}$  is slightly shifted from the nominal value of  $Q_\alpha$  ( $\approx 5.4$  MeV) due to a different heat response for  $\alpha$  particles, already observed in this type of detectors [15, 17–19]. The mean value and three  $\sigma$  intervals for  $\text{LY}_{\gamma(\beta)}$  are shown by solid red lines.

culated by the  $\text{Zn}^{100}\text{MoO}_4$ , *top*, scintillating bolometer is depicted in Fig. 6. The  $\text{LY}_{\gamma(\beta)}$  was estimated by fitting the data in a 600–2700 keV interval. The fit gives similar values for all working temperatures: e.g. 1.01(11) keV/MeV (at 13.7 mK) and 1.02(11) keV/MeV (at 19 mK) for the crystal  $\text{Zn}^{100}\text{MoO}_4$ , *top*, and 0.93(11) keV/MeV (at 13.7 mK) and 0.99(12) keV/MeV (at 19 mK) for the crystal  $\text{Zn}^{100}\text{MoO}_4$ , *bottom*.

In spite of the short duration of the measurements and the aboveground conditions, the data presented in Fig. 6 demonstrate an encouraging internal radiopurity level of the tested  $\text{Zn}^{100}\text{MoO}_4$  crystals. There are no remarkable accumulations of counts in the region where events caused by  $\alpha$  radionuclides from the U/Th chains are expected.<sup>1</sup> A weak event cluster probably related to  $^{210}\text{Po}$  is used to determine the activity of this radionuclide, considering a 200 keV interval

<sup>1</sup> Taking into account the published values of quenching factors for  $\alpha$  particles ( $\approx 0.12$ – $0.18$ ) derived in measurements with  $\text{ZnMoO}_4$ -based bolometers [15, 17–19], we consider all the events with energy in the range 4–9 MeV and light yield below 0.2 keV/MeV to be related to potential  $\alpha$  contaminants.

around its centroid. The analysis of the data accumulated at 13.7 and 19 mK with the  $\text{Zn}^{100}\text{MoO}_4$ , *top*, and  $\text{Zn}^{100}\text{MoO}_4$ , *bottom*, bolometers gives an activity of  $^{210}\text{Po}$  in the crystals at the level of 1.1(3) and 1.2(3) mBq/kg, respectively. These activities are similar to those measured precisely in natural  $\text{ZnMoO}_4$  bolometers [19]. It should be noted that the current statistics of the  $^{210}\text{Po}$  counts (4–6 events depending on the set of measurements) is not enough to get precise values of the activities and to distinguish a bulk contamination of  $^{210}\text{Po}$  (or of the progenitor  $^{210}\text{Pb}$ ) from surface pollution. The radiopurity of the  $\text{Zn}^{100}\text{MoO}_4$  crystals will be precisely determined in next measurements in underground conditions, but the absence of significant  $\alpha$  peaks at this level indicates that the contamination of the harmful nuclides  $^{228}\text{Th}$  and  $^{214}\text{Bi}$  [15] does not exceed the level of a few mBq/kg.

In Figs. 5 and 6 a few points appear below the  $\gamma/\beta$ /muon band, at energies both higher and lower than  $\sim 3$  MeV. Due to the low statistics it is difficult to fully understand the origin of these events. They could be due partly to energy-degraded  $\alpha$  particles coming from surface contamination and partly to pile-up effects. In fact, the different time structures of heat pulses and light pulses (which have shorter rise and decay times, as reported in Table 3) can produce piled-up events with a fake heat-to-light ratio, higher than that expected for a  $\beta$ -like interaction. This effect vanishes underground, where random coincidences are extremely improbable.

Another important consideration is the good reproducibility of the behavior of the two detectors. The slight differences in the operational parameters of the two devices are well within the typical spread observed in this type of detector. Both detectors, despite some difference in the optical quality, have shown practically identical bolometric and scintillation characteristics.

## 4 Conclusions and prospects

In the framework of the LUMINEU program, a zinc molybdate crystal with a mass of 171 g was produced from molybdenum enriched in  $^{100}\text{Mo}$  to 99.5 %, after a complex and effective purification method. The output of the crystal boule is 84 %, which demonstrates an important advantage of the low-thermal-gradient Czochralski technique for crystal growing. The irrecoverable losses of enriched molybdenum were found to be a few %. The results on the scintillating bolometers fabricated with two enriched crystals obtained from the boule show that the response of these devices meets the requirements of a high-sensitivity double beta decay search based on this technology and discussed in Ref. [15]. In particular, the baseline width is compatible with energy resolutions of few keV FWHM in the region of interest and the measured light yield can provide an  $\alpha$  rejection factor far higher than 99.9 %. Last but not least, the response is not dis-

tinguishable from the one observed in recent measurements performed with non-enriched detectors [15, 17–21]. Encouraging, although preliminary, results were also obtained in terms of radiopurity. One can therefore expect a successful operation of scintillating bolometers based on enriched  $\text{Zn}^{100}\text{MoO}_4$  crystals in an underground environment. This measurement is presently in preparation.

Recently, significant improvements in the growth technology developed at NIIC (Novosibirsk, Russia) have enabled the synthesis of large regular-shape cylindrical ( $\varnothing 5 \times 4$  cm) natural  $\text{ZnMoO}_4$  crystals. By using an amount of enriched material larger than that employed in the experiment here described,<sup>2</sup> we foresee to develop  $\text{Zn}^{100}\text{MoO}_4$  single detectors with masses in the 300–500 g range in the near future. The size of these devices corresponds to the one envisaged for the single module of large arrays of  $\text{Zn}^{100}\text{MoO}_4$  detectors to search for  $0\nu 2\beta$  decay of  $^{100}\text{Mo}$  [15]. The currently available enriched material will allow the development of an array with a total mass of  $\approx 15$ –20 kg, corresponding to a sensitivity to the effective Majorana mass in the range 0.05–0.15 eV [15] and capable therefore to approach the inverted hierarchy region of the neutrino mass pattern.

**Acknowledgments** The development of  $\text{Zn}^{100}\text{MoO}_4$  scintillating bolometers is an essential part of the LUMINEU program, receiving funds from the French National Research Agency (ANR). The work was supported partly by the project “Cryogenic detectors to search for neutrinoless double beta decay of molybdenum” in the framework of the Programme “Dnipro” based on Ukraine-France Agreement on Cultural, Scientific and Technological Cooperation. We are pleased to acknowledge funding from the P2IO LabEx (ANR-10-LABX-0038) in the framework “Investissements d’Avenir” (ANR-11-IDEX-0003-01) managed by the ANR. The aboveground facility for the test of the scintillating bolometers was realised with the indispensable support of ISOTTA, a project funded by the ASPERA 2nd Common Call dedicated to R&D activities. The group from the Institute for Nuclear Research (Kyiv, Ukraine) was supported in part by the Space Research Program of the National Academy of Sciences of Ukraine. It is a pleasure to thank Stefano Nisi from the Gran Sasso Laboratory (Italy) for his kind help in the measurements of enriched molybdenum oxide by inductively coupled plasma mass-spectrometry.

**Open Access** This article is distributed under the terms of the Creative Commons Attribution License which permits any use, distribution, and reproduction in any medium, provided the original author(s) and the source are credited.

Funded by SCOAP<sup>3</sup> / License Version CC BY 4.0.

## References

- O. Cremonesi, M. Pavan, *Adv. High Energy Phys.* **2014**, 951432 (2014)
- B. Schwingenheuer, *Ann. Phys.* **525**, 269 (2013)
- S.R. Elliott, *Mod. Phys. Lett. A* **27**, 1230009 (2012)
- J.D. Vergados, H. Ejiri, F. Simkovic, *Rep. Prog. Phys.* **75**, 106301 (2012)
- A. Giuliani, A. Poves, *Adv. High Energy Phys.* **2012**, 857016 (2012)
- J.J. Gomez-Cadenas et al., *Riv. Nuovo Cim.* **35**, 29 (2012)
- W. Rodejohann, *Int. J. Mod. Phys. E* **20**, 1833 (2011)
- V.I. Tretyak, Yu.G. Zdesenko, *At. Data Nucl. Data Tables* **61**, 43 (1995)
- V.I. Tretyak, Yu.G. Zdesenko, *At. Data Nucl. Data Tables* **80**, 83 (2002)
- A. Giuliani, S. Sanguinetti, *Mater. Sci. Eng. R* **11**, 1 (1993)
- A. Alessandrello et al., *Phys. Lett. B* **420**, 109 (1998)
- S. Pirro et al., *Phys. Atom. Nucl.* **69**, 2109 (2006)
- M. Tenconi et al., LUMINEU: a pilote scintillating bolometer experiment for neutrinoless double beta decay search. Presented at TAUP 2013 and to be published in the proceedings
- S. Rahaman et al., *Phys. Lett. B* **662**, 111 (2008)
- J.W. Beeman et al., *Phys. Lett. B* **710**, 318 (2012)
- D.R. Artusa et al., *Eur. Phys. J. C* **74**, 3096 (2014). [arXiv:1404.4469](https://arxiv.org/abs/1404.4469) [nucl-ex]
- J.W. Beeman et al., *Astropart. Phys.* **35**, 813 (2012)
- J.W. Beeman et al., *J. Low Temp. Phys.* **167**, 1021 (2012)
- J.W. Beeman et al., *Eur. Phys. J. C* **72**, 2142 (2012)
- D.M. Chernyak et al., *Nucl. Instrum. Methods A* **729**, 856 (2013)
- L. Bergé et al., *JINST* **9**, P06004 (2014)
- M. Berglund, M.E. Wieser, *Pure Appl. Chem.* **83**, 397 (2011)
- C. Arnaboldi et al., *Phys. Rev. C* **78**, 035502 (2008)
- D. Blum et al., *Phys. Lett. B* **275**, 506 (1992)
- P. Belli et al., *Nucl. Phys. A* **846**, 143 (2010)
- A.A. Pavlyuk, Ya.V. Vasiliev, L.Yu. Kharchenko, F.A. Kuznetsov, in *Proceedings of the APSAM-92, Asia Pacific Society for Advanced Materials*, Shanghai, 26–29 April 1992, Institute of Materials Research, Tohoku University, Sendai, Japan (1993), p. 164
- A. Borovlev et al., *J. Cryst. Growth* **229**, 305 (2001)
- E.N. Galashov, V.A. Gusev, V.N. Shlegel, Ya.V. Vasiliev, *Crystallogr. Rep.* **54**, 689 (2009)
- P. Belli et al., *Nucl. Instrum. Methods A* **615**, 301 (2010)
- A.S. Barabash et al., *JINST* **6**, P08011 (2011)
- M. Tenconi et al., Bolometric light detectors for neutrinoless double beta decay search. Presented at international workshop on new photon-detectors 2012, LAL Orsay, France, 13–15 June 2012. *PoS (PhotoDet 2012)* 072
- M. Mancuso et al., *J. Low Temp. Phys.* **571**, 571 (2014)
- C. Arnaboldi et al., *IEEE Trans. Nucl. Sci.* **49**, 2440 (2002)
- E. Armengaud et al., *Phys. Lett. B* **702**, 329 (2011)
- E. Gatti, P.F. Manfredi, *Riv. Nuovo Cim.* **9**, 1 (1986)

<sup>2</sup> Several kilograms of enriched molybdenum, belonging partly to the Institute of Theoretical and Experimental Physics (Moscow, Russia) and partly to the Institute for Nuclear Research (Kyiv, Ukraine) are available for the development of  $\text{Zn}^{100}\text{MoO}_4$  scintillating bolometers.