Scintillating bolometers based on ZnMoO\(_4\) and Zn\(^{100}\)MoO\(_4\) crystals to search for 0\(\nu\)\(\beta\) decay of \(^{100}\)Mo (LUMINEU project): first tests at the Modane Underground Laboratory

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Abstract

The technology of scintillating bolometers based on zinc molybdate (ZnMoO\(_4\)) crystals is under development within the LUMINEU project to search for 0\(\nu\)\(\beta\) decay of \(^{100}\)Mo with the goal to set the basis for large scale experiments capable to explore the inverted hierarchy region of the neutrino mass pattern. Advanced ZnMoO\(_4\) crystal scintillators with mass of \(\sim 0.3\) kg were developed and enriched \(^{100}\)MoO\(_2\) crystal from enriched \(^{100}\)Mo was produced for the first time by using the low-thermal-gradient Czochralski technique. One ZnMoO\(_4\) scintillator and two samples (59 g and 63 g) cut from the enriched boule were tested aboveground at milli-Kelvin temperature as scintillating bolometers showing a high detection performance. The first results of the low background measurements with three ZnMoO\(_4\) and two enriched detectors installed in the EDELWEISS set-up at the Modane Underground Laboratory (France) are presented.

Keywords: Double beta decay, Scintillating bolometer, ZnMoO\(_4\) crystal scintillator, Low counting experiment
1. Introduction

Scintillating bolometers — cryogenic detectors with a heat-light double read-out — can play a crucial role in next-generation experiments to study neutrino properties and weak interaction via investigating neutrinoless double beta ($0\nu2\beta$) decay, as discussed in Refs. 1, 2. This technique is extensively developing now within the LUCIFER 3, 4, the AMORE 5, 6, and the LUMINEU 7 $0\nu2\beta$ projects. This paper describes the recent achievements in the framework of the LUMINEU programme (Luminescent Underground Molybdenum Investigation for NEUtrino mass and nature).

LUMINEU is devoted to the development of a technology based on zinc molybdate (ZnMoO$_4$) scintillating bolometers as a basis for the realization of a high-sensitivity $0\nu2\beta$ experiment. The good prospects of this material for the bolometric technique are clearly shown in recent investigations 1, 8–13. An important point in the realization of LUMINEU is concerned with the technology of growing high-quality radiopure large mass (0.3–0.5 kg) ZnMoO$_4$ single crystals with the aim to produce scintillators enriched in $^{100}$Mo (Zn$^{100}$MoO$_4$). Here we report a significant progress in the development of ZnMoO$_4$ crystal scintillators using deeply purified compounds (containing molybdenum with natural isotopic composition and enriched in $^{100}$Mo). We also present results of both aboveground and underground low temperature tests of new scintillating bolometers based on natural ZnMoO$_4$ and enriched Zn$^{100}$MoO$_4$ crystal scintillators in light of their possible application to next-generation $0\nu2\beta$ decay experiments.

2. Development of zinc molybdate based scintillating bolometers

A precursor of the LUMINEU programme, a slightly yellow colored 313 g ZnMoO$_4$ sample with irregular shape, was produced from the first large volume ZnMoO$_4$ crystal boule grown by the low-thermal-gradient Czochralski (LTG Cz) technique 14, 15 in the Nikolaev Institute of Inorganic Chemistry (NIIC, Novosibirsk, Russia). The second sample (with mass 329 g) produced from this boule was tested as a scintillating bolometer at the Gran Sasso National Laboratories (LNGS, Assergi, Italy) 16.

Advanced ZnMoO$_4$ crystal boules with mass of ~ 1 kg have been produced recently at the NIIC by using the LTG Cz growth technique and molybdenum purified by sublimation in vacuum and double recrystallization from aqueous solutions 13. The crystals were recrystallized to improve quality of the material, and two colorless ZnMoO$_4$ cylindrical samples (with size $\phi 50 \times 40$ mm and mass 336 and 334 g) were produced from them. Moreover, a zinc molybdate crystal boule (with mass 171 g) enriched in $^{100}$Mo to 99.5% was developed for the first time at the NIIC 16, 17, and two scintillation elements (with mass 59 and 63 g) were cut from the boule. The enriched molybdenum was purified by sublimation and recrystallization from aqueous solutions. It is worth noting the high yield of the Zn$^{100}$MoO$_4$ crystal boule from the initial charge (84%) and low level of total irrecoverable losses of enriched material (4%) achieved in the frame of this R&D 16. Some coloration of the crystal (in contradiction with the practically colorless samples produced from natural molybdenum) can be explained by remaining traces of iron in the enriched molybdenum and by crystallization procedure performed only one time 16.

In order to construct scintillating bolometers, all the above described samples were held inside Copper holders by using PTFE clamps. Both Zn$^{100}$MoO$_4$ crystals were mounted in one Copper holder. The crystal scintillators were surrounded by a reflector foil (3M VM2000/2002) to improve light collection. Thin ultrapure Ge wafers ($\phi 50 \times 0.25$ mm) were used for detecting scintillation light. The 313 g crystal was viewed by two light detectors fixed on the opposite sides. The ZnMoO$_4$ / Zn$^{100}$MoO$_4$ crystals and the Ge photodetectors were instrumented with Neutron Transmutation Doped (NTD) Ge thermistors used as temperature sensors. All the crystals were also assembled with an individual heating element based on a heavily-doped silicon meander. Such devices provide a stable resistance value and are used to inject periodically a certain amount of thermal energy with the aim to control and stabilize the thermal bolometric response. All the detector modules are shown in Fig. 1.

3. Aboveground low temperature tests

The 313 g ZnMoO$_4$ precursor and both Zn$^{100}$MoO$_4$ crystals were tested in aboveground cryogenic facilities of the Centre de Sciences Nucléaires et de Sciences de la Matière (CSNSM, Orsay, France) with “wet” and “dry” $^3$He/$^4$He dilution refrigerators, respectively.

Both cryostats are surrounded by passive shield made of low activity lead to minimize signals pile-up caused by environmental gamma rays due to a slow time response of the bolometers (hundreds millisecond). The
Figure 1: Photographs of scintillating bolometers based on ultra-pure Ge photodetectors and ZnMoO$_4$ precursor with mass of 313 g (a), advanced quality (see text) ZnMoO$_4$ crystals with masses of 336 and 334 g (b), and enriched Zn$^{100}$MoO$_4$ crystals with masses of 59 and 63 g (c).

Stream data were recorded by a 16 bit ADC with a sampling frequency of 30 kHz and 10 kHz for natural and enriched detectors, respectively. The ZnMoO$_4$ precursor was operated at 17 mK during the measurements (over 38 h), while the Zn$^{100}$MoO$_4$ array was tested at 13.7 mK (18 h), 15 mK (5 h), and 19 mK (24 h) base temperatures. Both detectors were irradiated by gamma quanta from a weak $^{232}$Th source, while the photodetectors were calibrated with the help of $^{55}$Fe sources fixed close to the Ge slabs.

The data treatment (here and below) was performed by using the optimum filtering [18]. The spectrometric performances of the precursor-based bolometer were deteriorated by the pile-ups effect due to considerably high counting rate $\approx 2.5$ Hz (e.g. see in Table 1 the energy resolution of the 2615 keV $\gamma$ peak). In spite of this, the test shows normal operability of the detector and allows us to estimate the scintillation light yield for the registered $\gamma(\beta)$ events and muons, as well as the possibility of particle discrimination between $\gamma(\beta)$ and $\alpha$ events due to the quenching of scintillation for $\alpha$ particles. All these data are reported in Table 1.

Both enriched crystals demonstrate similar performance at all the temperatures [16]. The 2-dimensional histogram obtained from the heat-light double read-out of the 59 g Zn$^{100}$MoO$_4$ bolometer at 13.7 mK is shown in Fig. 2 (a). The light and the heat signals detected simultaneously allow to get a clear discrimination between $\alpha$ and $\gamma(\beta)$ particles. The absence in Fig. 2 (a) of peculiarities related with the detection of $\alpha$ events (except a small structure possibly caused by $^{210}$Po, as

Figure 2: (a) The scatter plot of the light-to-heat signal amplitude ratio as a function of the heat signal amplitude accumulated at 13.7 mK in aboveground test with the 59 g Zn$^{100}$MoO$_4$ scintillating bolometer during 18 h of calibration measurements with the $^{232}$Th source. The visible band is related to $\gamma(\beta)$ events (below 2.6 MeV) and cosmic muons. Three sigma intervals of the light yield for the $\gamma(\beta)$ band are shown by solid red curves together with the median value. (b) The energy spectrum built from the data presented in the upper plot. The peaks observed in the energy spectrum belong to the $^{232}$Th source and environmental gamma’s (daughters of $^{226}$Ra). The energy of marked peaks are given in keV.
often occurs in scintillators) indicates on encouraging radiopurity of the tested enriched crystals. Good spectrometric properties of the enriched detectors, even at aboveground conditions, are well visible from Fig. 2 (b), while some further information about their performances is presented in Table 1.

4. Underground cryogenic measurements

The 313 g detector was moved deep underground (∼4800 m w.e.) to the Modane Underground Laboratory (Laboratoire Souterrain de Modane, LSM, France) and tested during the EDELWEISS-III commissioning runs. The ZnMoO$_4$ bolometer together with fifteen ultra-pure Ge detectors (0.8 kg each) fully covered with interleaved electrodes (FID) were installed inside the $^3$He/$^4$He inverted dilution refrigerator with a large experimental volume (50 × 40 mm) and polyethylene (50 cm). The set-up is surrounded by a 5 cm thick plastic scintillator muon veto (95% coverage), and equipped by neutron and radon counters.

The triggered signals were recorded by a 14 bit ADC in 2 s window with 2 kHz sampling rate (the half of the window contains the baseline data). The base temperature was stabilized around 19 mK. One light detector was very sensitive to microphonic noise and could not be used for measurements. The energy scale of the ZnMoO$_4$ detector has been measured in calibration runs with $^{133}$Ba and $^{232}$Th $\gamma$ sources, performed over 546 h and 70 h, respectively. The background data were accumulated over 305 h.

The powerful discrimination capability achieved with the 313 g ZnMoO$_4$ scintillating bolometer is well illustrated in Fig. 3 (a), which shows a full separation of $\gamma$($\beta$)-induced events from populations of $\alpha$ particles caused by trace impurity by radionuclides from U/Th chains (mainly, $^{210}$Po, see below). The energy spectrum accumulated with the $^{232}$Th gamma source (see Fig. 3 (b)) demonstrates high spectrometric properties of the detector. An overview of the detector’s performances during underground measurements is given in Table 1.

After completing the EDELWEISS-III commissioning runs, other two ZnMoO$_4$-based scintillating bolometers ($\sim$50 × 40 mm) and the Zn$^{100}$MoO$_4$ array together with 36 FID Ge detectors were assembled. The

Table 1: List of achieved performances with ZnMoO$_4$ and Zn$^{100}$MoO$_4$ detectors tested in aboveground and underground measurements. We report the energy resolution for the heat channels (FWHM — Full Width at the Half of Maximum) estimated as filtered baseline and measured for $\gamma$ quanta and $\alpha$ particles of internal $^{210}$Po. We report also the light yield for $\gamma$($\beta$) events (LY$_{\gamma\beta}$) and quenching factor for $\alpha$ particles (QF$_{\alpha}$).

| Detector   | Crystal Mass (g) | FWHM (keV) | LY$_{\gamma\beta}$ (keV/MeV) | QF$_{\alpha}$  |
|------------|-----------------|------------|-----------------------------|---------------|
|            | ZnMoO$_4$       | 313        | 1.8(1)                      | 0.93(11)      |
|            | Zn$^{100}$MoO$_4$| 59         | 1.4(1)*                     | 0.15*         |

* — results based on the aboveground measurements

![Figure 3](image-url)
EDELWEISS set-up was also upgraded: a) a polyethylene shield at the 1 K plate was added; b) new ultra radiopure NOSV Copper \(220\) screens were installed; c) all detectors were provided with individual low background Copper-Kapton cables. In addition, a pulser system to assist to the calibration of the thermal response of the ZnMoO\(_4\) / Zn\(^{100}\)MoO\(_4\) detectors will be implemented soon.

Figure 4: (a) Scatter plot of the light versus the heat signals measured by the 334 g ZnMoO\(_4\) scintillating bolometer in a 15 h calibration run with the \(^{133}\)Ba gamma source in the EDELWEISS set-up. A cluster of events located far from the \(\gamma / \beta\) population corresponds to \(\alpha\) particles of \(^{210}\)Po. The data for the light channel are presented in ADU (Analogue-to-Digital Unit). (Insert) Part of the scatter plot corresponding to the energy range of the used source. (b) The energy spectrum of the \(^{133}\)Ba source measured over 15 h by the 334 g ZnMoO\(_4\) scintillating bolometer.

After the upgrade of the set-up the data are recorded by a 16 bit ADC with 1 kHz sampling rate (the length of pulse profile is 2 s with the half of the window for the baseline data). The working temperature is stabilized at 18 mK. The energy scale of the detectors was measured with the \(^{133}\)Ba gamma source (the measurements with the \(^{232}\)Th source are foreseen).

The set-up is still under optimization, especially as far as the control of the vibration-induced noise is concerned. Therefore, we discuss here, as an illustrative example, only the results achieved with the 334 g natural ZnMoO\(_4\) scintillating bolometer. This detector exhibits full \(\alpha / \gamma (\beta)\) separation, as shown in Fig. 4(a), as well as excellent spectrometric properties, as demonstrated in Fig. 4(b). Other relevant information about performances of \(\varnothing 50 \times 40\) mm ZnMoO\(_4\) detectors are reported in Table I.

5. Radiopurity of ZnMoO\(_4\) and Zn\(^{100}\)MoO\(_4\) crystals

The radiopurity level of the ZnMoO\(_4\) crystals was estimated by analysis of the \(\alpha\) events selected from the underground runs, while the data of the aboveground measurements were used in case of the Zn\(^{100}\)MoO\(_4\) samples. The position of the 5.4 MeV \(\alpha\) peak of the internal \(^{210}\)Po, clearly visible in the data for the natural crystals, was used to stabilize the thermal response of the detectors. For instance, the spectra of the \(\alpha\) events registered by the detectors based on 313 g (a) and 334 g (b) ZnMoO\(_4\) crystals over 851 h and 527 h, respectively, are shown in Fig. 5.

![Figure 5](image-url)

Figure 5: The \(\alpha\) spectra collected in the low background measurements in the EDELWEISS set-up with the ZnMoO\(_4\) scintillating bolometers based on the 313 g precursor (a) and the 334 g advanced sample (b) operated over 851 h and 527 h, respectively. The origin of the \(\alpha\) events providing the highest rate are indicated.

The crystals are slightly polluted by \(^{210}\)Po detected through 5.4 MeV \(\alpha\) peak confirming a broken equilibrium in the radioactive chain. \(^{226}\)Ra (and its daughters \(^{222}\)Rn, \(^{218}\)Po, and \(^{214}\)Bi,\(^{214}\)Po events), and \(^{228}\)Th (with daughter \(^{224}\)Rn) were detected in the 313 g crystal, while the ZnMoO\(_4\) scintillators produced by recrystallization have shown a much better level of radiopurity, particularly in \(^{226}\)Ra. It is also evident a higher surface contamination by \(^{210}\)Po of the 313 g crystal or/and of the bolometer components close to it (a peak at 5.3 MeV

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\(^{3}\)Taking into account a short half-life of \(^{216}\)Po (145 ms), which is comparable with the time response of the 313 g detector (hundreds ms), subsequent \(\alpha\) decays of \(^{220}\)Rn,\(^{216}\)Po give pile-ups and therefore were discarded from the data by the pulse-shape analysis.
corresponds to $E_\alpha$ of $^{210}$Po. In addition, excess counts around 5.8 MeV also indicate a possible surface contamination but its origin has not been identified.

The activity of internal $^{210}$Po was derived from the fit of the 5.4 MeV peak, while 3$\sigma$ intervals (according to the energy resolution of the internal $^{210}$Po — see Table 1 centered at the $Q_\alpha$ value were used for the calculation of the area of the peaks of other radionuclides from U/Th chains. The background contribution was evaluated in two energy regions (3.3–4 and 4.35–4.7 MeV) with a flat $\alpha$ continuum in which no peaks are expected. The number of counts excluded with 90% C.L. were calculated by using the Feldman-Cousins procedure [21].

Table 2: Radioactive contamination of the ZnMoO$_4$ and Zn$^{100}$MoO$_4$ crystals tested as scintillating bolometers in aboveground and underground conditions. The mass of the crystals and the total time of the accumulated data are also presented. The results for the large mass ZnMoO$_4$ crystal which was operated as the scintillating bolometer at the LNGS (Italy) [11] are given for comparison. The uncertainties are given with 68% C.L., while all the limits are at 90% C.L.

| Nuclide | Activity (mBq/kg) Zn$^{100}$MoO$_4$ | | Activity (mBq/kg) Zn$^{100}$MoO$_4$ | |
|---------|-------------------------------| --- |-------------------------------|---|
| $^{212}$Po | 5.9 g | 0.2 g | 216 g | 316 g | 334 g | 313 g | 336 g |
| $^{226}$Ra | 0.27 | 0.25 | 213 g | 214 g | 215 g | 216 g | 217 g |
| $^{228}$Ra | 0.31 | 0.26 | 214 g | 215 g | 216 g | 217 g | 218 g |

Data (or limits) on radioactive contamination of the ZnMoO$_4$ and Zn$^{100}$MoO$_4$ scintillators are summarized in Table 2 where the results for another ZnMoO$_4$ sample, produced from the same boule as the 313 g crystal was, are presented for comparison. As it is seen from Table 2 the improved purification and crystallization procedure adopted for the LUMINEU crystals of 334 and 336 g has lead to a significant reduction of the internal contamination, especially for $^{228}$Ra which is not detectable now while it was clearly present in both precursor crystals (313 and 329 g). In particular, the radiopurity levels ($\leq 0.01$ mBq/kg) achieved for $^{228}$Th and $^{226}$Ra are fully compatible with next-generation $0\nu$2$\beta$ experiments capable to explore the inverted hierarchy region of the neutrino mass pattern [1, 2].

6. Conclusions

A significant progress is achieved in development of ZnMoO$_4$ crystal scintillators for the LUMINEU project. Large volume crystal boules (~ 1 kg each) were grown by the low-thermal-gradient Czochralski technique from deeply purified molybdenum. A Zn$^{100}$MoO$_4$ crystal boule with a mass of 0.17 kg was produced from enriched $^{100}$Mo (to 99.5%) for the first time. Three natural (~ 0.3 kg) and two enriched (~ 0.06 kg) scintillation elements were produced for low temperature studies. Production of large volume Zn$^{100}$MoO$_4$ crystal scintillators from enriched $^{100}$Mo is in progress.

The cryogenic scintillating bolometric tests of the natural and enriched crystals showed a high performance of the detectors. The deep purification of molybdenum and recrystallization significantly improve the radioactive contamination of ZnMoO$_4$ crystals by $^{228}$Th and $^{226}$Ra to the level of $\leq 0.01$ mBq/kg requested by the LUMINEU project.

The results of this study clarify the excellent prospects of ZnMoO$_4$ scintillating bolometers for the next generation $0\nu$2$\beta$ experiments aiming to approach the inverted hierarchy region of the neutrino mass pattern.

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