$^{100}$Mo-enriched Li$_2$MoO$_4$ scintillating bolometers for 0ν2β decay search: from LUMINEU to CUPID-0/Mo projects

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Abstract. A scintillating bolometer technology based on $^{100}$Mo-enriched lithium molybdate (Li$_2^{100}$MoO$_4$) crystals has been developed by LUMINEU to search for neutrinoless double-beta (0ν2β) decay of $^{100}$Mo. The results of several low temperature tests at underground environments have proved the reproducibility of high detector performance and crystal radiopurity: in particular ~5–6 keV FWHM energy resolution and at least 9σ rejection of α’s in the vicinity of the 0ν2β decay of $^{100}$Mo (3034 keV) and below 10 μBq/kg bulk activity of $^{228}$Th and $^{226}$Ra. A modest acquired exposure (0.1 kg•yr) is a limiting factor of the LUMINEU experiment sensitivity to the 0ν2β decay half-life of $^{100}$Mo ($T_{1/2} ≥ 0.7×10^{23}$ yr at 90% C.L.), however the two-neutrino 2β decay has been measured with the best up-to-date accuracy, $T_{1/2} = [6.92 ± 0.06(\text{stat.}) ± 0.36(\text{syst.})] × 10^{20}$ yr. The applicability of the LUMINEU technology for a ton-scale 0ν2β decay bolometric project CUPID is going to be demonstrated by the CUPID-0/Mo experiment with ~5 kg of $^{100}$Mo embedded in forty 0.2 kg Li$_2^{100}$MoO$_4$ scintillating bolometers. A first phase of the experiment with twenty Li$_2^{100}$MoO$_4$ detectors is in preparation at the Modane underground laboratory (France) to start by the end of 2017.

INTRODUCTION

Searches for neutrinoless double-beta decay (0ν2β) — a lepton number violating spontaneous nuclear transition which requires a Majorana nature of neutrinos (e.g. see the recent review [1] and references herein) — are among the hottest worldwide experimental efforts in Astroparticle physics. The sensitivity to the 0ν2β half-lives already / to be achieved by the present generation leading 0ν2β experiments (lim$T_{1/2}^{0ν2β} ~ 10^{24} − 10^{26}$ yr [1, 2]) has to be improved by two orders of magnitude to make a further significant progress in the field [3]. Such enhancement would be possible with a new / advanced technology, which can provide near zero-background conditions in the region of interest (around the Q-value of the 0ν2β transition, Q$_{ββ}$) for a ton-scale detector over years of exposure. LUMINEU (Luminescent Underground Molybdenum Investigation for NEUtrino mass and nature) is a French-funded project (2012–2017) aiming at the development of such challenging technology based on $^{100}$Mo-enriched zinc and lithium molybdate (ZnMoO$_4$ and Li$_2$MoO$_4$) scintillating bolometers. Since recently, LUMINEU is part of an R&D activity towards CUPID (CUORE Upgrade with Particle ID) [4, 5], a next-generation 0ν2β project aiming at using as much as possible the infrastructure of the present ton-scale bolometric experiment CUORE. In this paper we will overview the main LUMINEU achievements concerning the R&D of Li$_2$MoO$_4$ scintillating bolometers which led to the preparation of a 0ν2β experiment CUPID-0/Mo aiming at a demonstration of the suitability of the LUMINEU technology for CUPID.

LUMINEU R&D OF Li$_2^{100}$MoO$_4$ SCINTILLATING BOLOMETERS

An R&D of $^{100}$Mo-enriched Li$_2$MoO$_4$ scintillating bolometers has been accomplished by LUMINEU as follows:

- Development of molybdenum purification methods [6].
- Screening selection of a commercial ultra-pure Li$_2$CO$_3$ powder [7]. An R&D of Li$_2$CO$_3$ powder purification is still ongoing to ensure its high purity.
- Optimization of the crystal growth with the help of the low-temperature-gradient Czochralski technique [8]. Investigation of a double crystallization to further improve crystal’s quality and radiopurity [7, 8].
- Underground tests of Li$_2$MoO$_4$ and Li$_2^{100}$MoO$_4$ ($^{100}$Mo enrichment is ~97%) scintillating bolometers [7].
• A pilot $2\beta$ experiment with four 0.2-kg Li$_2^{100}$MoO$_4$ cryogenic detectors.

The results of the LUMINEU R&D demonstrated that the crystallization technology is mature for a mass production of large (up to $\varnothing 4.5 \times 15$ cm or $\varnothing 6 \times 10$ cm), perfect optical quality, highly radiopure Li$_2^{100}$MoO$_4$ scintillators and that the bolometric technology is well reproducible in terms of high detector’s performance (e.g. see in Table 1 and Figure 1), which altogether completely satisfies the LUMINEU specifications.

**TABLE 1.** Performance and radiopurity of four Li$_2^{100}$MoO$_4$ scintillating bolometers tested at 17 mK in the EDELWEISS set-up at the Modane underground laboratory (LSM, France). The energy resolution (FWHM) is measured at 2615 keV $\gamma$ quanta of $^{208}$Tl during a $^{232}$Th calibration. The light yield for $\gamma$'s (LY) and the $\alpha$/$\gamma$ separation efficiency are extracted from an AmBe calibration data (the 2.5–3.5 MeV $\gamma$'s and $\alpha$+t events with $\sim$5.1 MeV electron-equivalent energy caused by $^4$Li(n,$\alpha$) reaction were used). The radioactive contamination is estimated from the analysis of the energy spectra of $\alpha$ events accumulated in the background measurements. The performed analysis is similar to the one described in detail in [7].

| Detector’s ID | Crystal’s mass (g) | FWHM (keV) at 2615 keV | LY $\gamma$/$\beta$ (keV/MeV) | $\alpha$/$\gamma$ Separation above 2.5 MeV | Activity ($\mu$Bq/kg) |
|---------------|--------------------|------------------------|-------------------------------|------------------------------------------|----------------------|
| enrLMO-1      | 186                | 5.8(6)                 | 0.41                          | 9$\sigma$                                | $\leq$ 4 450(30)     |
| enrLMO-2      | 204                | 5.7(6)                 | 0.38                          | 9$\sigma$                                | $\leq$ 6 200(20)     |
| enrLMO-3      | 213                | 5.5(5)                 | 0.73                          | 14$\sigma$                               | $\leq$ 3 76(10)      |
| enrLMO-4      | 207                | 5.7(6)                 | 0.74                          | 14$\sigma$                               | $\leq$ 5 20(6)       |

**FIGURE 1.** Left panel: The energy spectra of the $^{232}$Th source and the dependence of FWHM energy resolution measured by a single 0.2 kg Li$_2^{100}$MoO$_4$ module and four-bolometer array operated at LNGS (12 mK; [7]) and LSM (17 mK), respectively. The expected resolution at $Q_{\beta\beta}$ of $^{100}$Mo is $\sim$5–6 keV FWHM shown as an open circle and an open square with error bars according to the fits to the LNGS and LSM data respectively. Right panel: The scatter-plots of light yield vs. heat energy of the AmBe data (290 h) of the enrLMO-2 and enrLMO-4 detectors operated at LSM without and with a reflecting film, respectively.

**INVESTIGATION OF 2$\beta$ DECAY OF $^{100}$Mo**

The LUMINEU pilot $2\beta$ experiment was able to perform a precise investigation of the two neutrino double-beta decay of $^{100}$Mo, as it is illustrated in Figure 2 (left). The analysis is similar to the one described in [7]. The half-life value is derived with the best up to-date accuracy (see Table 2). Figure 2 (right) shows a few events registered above the 2615 keV peak with an average rate 1.1(2) cpd/kg, but no events are observed in the 200-keV-wide energy interval centered at $Q_{\beta\beta}$ of $^{100}$Mo. Thus, we set a lower half-life limit $T_{1/2}^{\text{2$\beta$}} \geq 0.7 \times 10^{23}$ yr at 90% C.L. which is about one order of magnitude weaker than the NEMO-3 result ($T_{1/2}^{\text{2$\beta$}} \geq 1.1 \times 10^{24}$ yr at 90% C.L. [9]), but it is achieved over an exposure of $^{100}$Mo shorter by a factor 600 (0.06 vs. 34.3 kg·yr).
is going to be dedicated to the EDEL WEISS low mass WIMPs search program [12].

Left panel: The background energy spectrum of FIGURE 2. leading ones in the field.

scintillating bolometers over 0.04 kg\times\muon-induced events [7]. Therefore, we are going to improve the present background by removing identified Th contaminated elements and using an available muon veto with a 98% coverage.

Taking into account the achievements of the LUMINEU project, an extension of the LUMINEU pilot experiment to the availability of \sim 7 kg of \textsuperscript{100}Mo-enriched molybdenum and the cryogenic set-up(s) able to host the LUMINEU-like modules, the CUPID-0/Mo project is planned to be realized in two phases:

- Twenty Li\textsubscript{2}\textsuperscript{100}MoO\textsubscript{4} crystals (\circ \times 45 mm, \sim 0.2 kg each; 2.34 kg of \textsuperscript{100}Mo) to be operated as scintillating bolometers in five towers inside the EDELWEISS set-up (LSM, France) by the end of 2017.
- Additional twenty similar-size Li\textsubscript{2}\textsuperscript{100}MoO\textsubscript{4}-based detectors ready to be operated in a complementary set-up\textsuperscript{1}, e.g. CUPID-0 (LNGS, Italy), in the middle of 2018.

As one can see in Table 3, the sensitivity of the considered CUPID-0/Mo configurations would be comparable with the most stringent constraints on the effective Majorana neutrino mass (0.06–0.6 eV) derived from the results of the most sensitive 0\nu2\beta experiments [1, 2], for which a typical exposure is tens–hundreds kg\times yr of isotope of interest. It should be noted that the sensitivity of the CUPID-0/Mo Phase I remains substantially unaffected even with an order of magnitude worse projected background (10^{-2} counts/yr/kg/keV), which is similar to the 0.06\pm 0.03 counts/yr/kg measured in the 2.8–3.6 MeV energy interval by the LUMINEU pilot 2\beta experiment\textsuperscript{2}. So, in spite of the considerably small scale of the CUPID-0/Mo experiment, this search would be among the leading ones in the field.

\textsuperscript{1}In principle, the experimental volume of the EDELWEISS set-up is able to host also these additional 20 detectors, however a part of the cryostat is going to be dedicated to the EDELWEISS low mass WIMPs search program [12].

\textsuperscript{2}The observed events above 2.65 MeV (see Figure 2) could be explained by pile-ups of \gamma cascade from \textsuperscript{208}Tl decays nearby the detectors and/or muon-induced events [7]. Therefore, we are going to improve the present background by removing identified Th contaminated elements and using an available muon veto with a 98% coverage.

CUPID-0/Mo 2\beta EXPERIMENT

| $T_{2\nu2\beta}^{1/2}$ (10^{18} yr) | S/B | Experiment | $\beta\beta$ Source | \textsuperscript{100}Mo exposure | Year [Ref.] |
|-----------------|-----|-------------|------------------|---------------------|------------|
| 7.11\pm 0.02(stat)\pm 0.54(syst) | 40  | NEMO-3      | \textsuperscript{100}Mo foils | 7.37 kg\times yr | 2005 [10] |
| 6.92\pm 0.06(stat)\pm 0.36(syst) | 10  | LUMINEU Li\textsubscript{2}\textsuperscript{100}MoO\textsubscript{4} bol. | 0.04 kg\times yr | 2017 [This work] |

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### Table 3

The CUPID-0/Mo sensitivity (90% C.L.) to the 0ν2β decay half-life of 100Mo for different configurations of the experiment. Assumed background is 10⁻³ counts/yr/kg/keV in 10 keV window centered at Qββ of 100Mo (73% of decays); the efficiency of the pulse shape discrimination is set to be 95% [7]. The recent calculations of a phase-space factor [13, 14], the nuclear matrix elements [15, 16], and an axial-vector coupling constant equal to 1.269 are used to estimate the sensitivity to the effective Majorana neutrino mass ⟨mββ⟩.

| CUPID-0/Mo configuration | Exposure (kg×yr of 100Mo) | limT_{1/2}^{0ν2β} (yr) | lim⟨mββ⟩ (eV) |
|---------------------------|---------------------------|-------------------------|-----------------|
| (1) 20×0.5 crystal×yr     | 1.2                       | 1.3×10⁻²⁴               | 0.33–0.56       |
| (2) 20×1.5 crystal×yr     | 3.5                       | 4.0×10⁻²⁴               | 0.19–0.32       |
| (3) 40×3.0 crystal×yr     | 14                        | 1.5×10⁻²⁵               | 0.10–0.17       |

### Conclusions

A production line of large, optical quality, radiopure 100Mo-enriched Li₂100MoO₄ crystal scintillators and their high performance as scintillating bolometers have been established within the LUMINEU project. A reasonably high sensitivity to the 0ν2β and the most precise half-life value for the 2ν2β decay of 100Mo (g.s. to g.s. transitions) have been achieved in a pilot LUMINEU 2β experiment over only ≈0.1 kg×yr exposure of four 0.2 kg Li₂100MoO₄ detectors array operated inside the EDELWEISS set-up in the Modane underground laboratory (France). A successful accomplishment of the LUMINEU project triggered its extension to CUPID-0/Mo 2β experiment aiming at operating forty 0.2-kg Li₂100MoO₄ scintillating bolometers. The start of data taking with 20 detectors is foreseen in the EDELWEISS set-up by the end of 2017. The main CUPID-0/Mo goal is to demonstrate zero-background conditions in the vicinity of the expected 100Mo 0ν2β decay peak and therefore to prove the viability of the LUMINEU technology for CUPID project, a ton-scale bolometric 0ν2β experiment.

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