## New Limit for Neutrinoless Double-Beta Decay of <sup>100</sup>Mo from the CUPID-Mo Experiment

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The CUPID-Mo experiment at the Laboratoire Souterrain de Modane (France) is a demonstrator for CUPID, the next-generation ton-scale bolometric  $0\nu\beta\beta$  experiment. It consists of a 4.2 kg array of 20 enriched  $\text{Li}_2^{100}\text{MoO}_4$  scintillating bolometers to search for the lepton-number-violating process of  $0\nu\beta\beta$ decay in <sup>100</sup>Mo. With more than one year of operation (<sup>100</sup>Mo exposure of 1.17 kg  $\times$  yr for physics data), no event in the region of interest and, hence, no evidence for  $0\nu\beta\beta$  is observed. We report a new limit on the half-life of  $0\nu\beta\beta$  decay in <sup>100</sup>Mo of  $T_{1/2} > 1.5 \times 10^{24}$  yr at 90% C.I. The limit corresponds to an effective Majorana neutrino mass  $\langle m_{BB} \rangle < (0.31-0.54)$  eV, dependent on the nuclear matrix element in the light Majorana neutrino exchange interpretation.

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The discovery that neutrinos are massive particles through the evidence of neutrino flavor oscillations [1] opens the question of neutrino mass generation. Instead of having Dirac nature as charged leptons and quarks, the scale of neutrino masses could be well motivated by the Majorana theory [2,3]. In this scenario, neutrinos could coincide with their antimatter partner [4,5], which would have a tremendous impact on our vision of nature, implying the violation of the total lepton number L as well as for the matter-antimatter asymmetry in the Universe [6,7].

The distinction between Dirac and Majorana behavior is an extreme experimental challenge. Neutrinoless doublebeta  $(0\nu\beta\beta)$  decay is the traditional and the most sensitive tool to probe the Majorana nature of neutrinos. This process is a nuclear transition consisting in the transformation of an even-even nucleus into a lighter isobar containing two more protons and accompanied by the emission of two electrons and no other particles, with a change of the lepton number Lby two units [8–11]. An observation of this hypothetical process would establish that neutrinos are Majorana particles [12]. The current most stringent limits on  $0\nu\beta\beta$  decay halflives are at the level of  $10^{25}$ – $10^{26}$  yr in <sup>136</sup>Xe, <sup>76</sup>Ge, and <sup>130</sup>Te [13–17].  $0\nu\beta\beta$  decay can be induced by a variety of mechanisms [9,11,18,19]. Among them, the so-called mass mechanism-consisting in the exchange of a virtual light Majorana neutrino-represents a minimal extension of the standard model. In this mechanism, the  $0\nu\beta\beta$  decay rate is proportional to the square of the effective Majorana neutrino mass  $\langle m_{\beta\beta} \rangle$ , a linear combination of the three neutrino mass eigenvalues which fixes the absolute neutrino mass scale. Present limits on  $\langle m_{\beta\beta} \rangle$  are in the range of (0.06–0.6) eV [11], assuming that the axial charge  $g_A$  is not quenched and equal to the free nucleon value of  $\simeq 1.27$  [20–22].

The distinctive signal of  $0\nu\beta\beta$  decay is a peak in the sum energy spectrum of the two emitted electrons at the total available energy  $Q_{\beta\beta}$  of the  $0\nu\beta\beta$  transition. Among the 35 natural double-beta emitters ( $0\nu\beta\beta$  candidate isotopes) [23], only a few of them are experimentally relevant. These favorable candidates feature a high  $Q_{\beta\beta}$  (> 2 MeV), which leads to a high decay probability and to a low background level in the signal region. At the same time, these candidates exhibit a high natural abundance of the isotope of interest and/or a technically feasible isotopic enrichment at the tonne scale.

Low-temperature calorimeters, often named bolometers, are the detectors of choice for several experimental efforts, including the one reported here. Featuring high energy resolution, high efficiency, and flexibility in detectormaterial choice [24–26], bolometers are perfectly tailored to  $0\nu\beta\beta$  search. These detectors consist in a single crystal that contains the  $0\nu\beta\beta$  source coupled to a temperature sensor. The signal is collected at very low temperatures  $\lesssim 20$  mK for large (0.1–1 kg) bolometers and consists of a thermal pulse registered by the sensor. A detector embedding a candidate with  $Q_{\beta\beta} > 2615$  keV is an optimal choice in terms of background control, as the bulk of the  $\gamma$  natural radioactivity ends at 2615 keV, corresponding to the energy of the <sup>208</sup>Tl line in the <sup>232</sup>Th decay chain. However, the energy region above ~2.6 MeV is dominated by events due to surface radioactive contamination, especially energy-degraded  $\alpha$  particles [17,27], as shown by the results of CUORE, the largest  $0\nu\beta\beta$  bolometric experiment currently under way.

A dual readout of light-scintillation or Cherenkovin addition to the thermal signal allows for the discrimination of  $\alpha$  events in various targets [25,26,28–33]. This technology has been developed for the scintillating  $Li_2^{100}MoO_4$  crystals used in CUPID-Mo by the LUMINEU Collaboration [34,35], and its effectiveness is described together with the experimental setup in Ref. [36]. The isotope of interest <sup>100</sup>Mo features a  $Q_{\beta\beta}$  of (3034.40  $\pm$ 0.17) keV [37] and a natural abundance of 9.7%, making large-scale enrichment viable by gas centrifuge isotopic separation [38]. In CUPID-Mo, it is embedded into  $Li_2^{100}MoO_4$  (LMO) crystals by a double low-thermalgradient Czochralski crystallization process [39] from enriched Mo previously used in the NEMO-3 experiment [40]. A total of 20 cylindrical ~210 g crystals are stacked into five towers, which results in a 100Mo mass of  $(2.258 \pm 0.005)$  kg with an average <sup>100</sup>Mo isotopic abundance of  $(96.6 \pm 0.2)$ %. Round Ge wafers, attached to the bottom of each LMO detector, are used as bolometric light detectors (LDs). Because of the stacking into the four-layer tower, most LMO detectors have a direct line of sight to a LD at both the top and bottom, except for the top crystal of each tower, which has a Cu lid on one side [36]. The LMO crystals as well as the LDs are instrumented with neutron-transmutation-doped-Ge sensors [41]. The towers are installed with a mechanical decoupling inside the EDELWEISS cryogenic infrastructure [42,43] at the Laboratoire Souterrain de Modane in France.

The data of the present analysis have been acquired over a 380 day period between March 2019 and April 2020 at operation temperatures of 20.7 and 22 mK. About 82% of the time was devoted to the  $0\nu\beta\beta$  search, split into 240 days of physics data and 73 days of calibration data. The physics data are grouped into a total of ten datasets with consistent operation conditions. In the following, we consider 213 out of the 240 days of physics data in seven (1–2-month-long) datasets and reject three (~1-week-long) datasets due to their small associated calibration statistics. From these seven datasets, we exclude periods of temperature instabilities, disturbances in the underground laboratory, and periods of excessive noise on the individual detectors, reducing the physics exposure by 6%. We reject one of the 20 LMO bolometers that shows an abnormal performance [36] and obtain a physics exposure of 2.16 kg × yr ( $\text{Li}_2^{100}\text{MoO}_4$ ).



FIG. 1. Physics spectrum (blue) for 2.16 kg × yr of data and calibration spectrum (red) scaled to match the 2615 keV counts from  $^{208}$ Tl. A ± 50 keV region around  $Q_{\beta\beta}$  has been blinded (gray).

All data are acquired as a continuous stream with 500 Hz sampling frequency and analyzed with a software package developed by the CUORE [44] and CUPID-0 [45] Collaborations, first used in CUPID-Mo in Refs. [36,46]. We estimate pulse amplitudes with an optimum filter [47], designed to maximize the signal to noise ratio for a known signal and noise spectrum with 3-s-long pulse traces for both the LMO and LD channels. The data were triggered offline using the optimum filter [17,48], obtaining 90% trigger efficiency at typical (median) energies of 9.4 (0.5) keV for the LMOs (LDs). The LMO analysis and coincidence thresholds have been set at 45 keV, well above this efficiency turn-on. For each signal on an LMO detector, we evaluate the resolution-weighted average light signal of the two (one) adjacent LDs to discriminate  $\alpha$  events, exhibiting ~20% of the light yield of  $\gamma$  and  $\beta$  events of the same energy [36]. We calibrate the response of the LMO detectors with a second-order polynomial using the four labeled peaks from the U/Th calibration data shown in Fig. 1 in red (see [46]) and cross-calibrate the LD against the LMO signals. We confirm the LMO's energy scale in background data fitting a second-order polynomial in reconstructed-to-expected peak position of the 352, 583, 609, 1461, and 2615 keV peaks and observe no systematic deviation. The extrapolation for the position of  $Q_{\beta\beta}$  agrees to within  $E_{\text{bias}}^{Q_{\beta\beta}} = (-0.2 \pm 0.4)$  keV.

We adopt a blinding strategy, removing all events in a  $\pm 50$  keV window around  $Q_{\beta\beta}$  to avoid any bias in the optimization of our analysis procedures and consider the following event selections. For events (i) to be contained in a single crystal and in anticoincidence with a triple-module trigger and energy deposit in the muon-veto system [49] based on a  $\pm 100$  ms time window; (ii) to have a single trigger in each 3-s pulse window; (iii) to have a flat pretrace with a slope of less than 15 median absolute deviations;

(iv) to have a pulse shape compatible with the principal components (PCs) established by a newly developed PC analysis described in Ref. [50]—this cut is optimized using calibration data by maximizing a hypothetical discovery sensitivity for a  $0\nu\beta\beta$  process equal in half-life to the previous best limit [40]; (v) to have the expected light yield for  $\gamma$  and  $\beta$  events and no difference in top and bottom LDs. Both of these cuts are set to obtain close to full coverage at  $\pm 3\sigma$  each, based on a Gaussian fit of the light yield in calibration data. The energy dependence of the cut was modeled with a phenomenological linear function, after we observed an excess broadening of the recorded light yield with respect to the photon statistics model discussed in Ref. [36]. The median excess width of 32 eV (~40%) at  $Q_{\beta\beta}$  is associated with an undersampling of the faster LD pulses and is presently under further investigation. A modified photon statistics model is also considered as a systematic in the limit setting.

The resulting physics spectrum summed over 19 LMO detectors and the entire data-taking period is shown in Fig. 1 in blue. Because of the short  $2\nu\beta\beta$  half-life (high rate) of <sup>100</sup>Mo [51,52], a smooth  $2\nu\beta\beta$  component dominates the spectrum from 0.5 to 3 MeV. A limited set of  $\gamma$  peaks remains visible, most notably <sup>208</sup>Tl, <sup>40</sup>K, <sup>60</sup>Co, and an activation  $\gamma$  peak from <sup>99</sup>Mo, present for a short time after a neutron irradiation of the detectors [53]. For more details, we refer to a prior characterization of the backgrounds in the EDELWEISS facility [42].

We optimize the  $0\nu\beta\beta$  search for a Poisson counting process in the low-background regime. We consider detector- and dataset-based (19 × 7) energy resolutions, a preliminary estimate of our background index, and an exposure of 2.8 kg × yr, as we intend to replicate the present analysis for the full exposure of the now-completed CUPID-Mo experiment.

The most representative  $\gamma$  peak for the <sup>100</sup>Mo region of interest (ROI) with sufficient statistics to extract detectorand dataset-based resolutions is the 2615 keV line from  $^{208}$ Tl in calibration data. We perform a simultaneous unbinned extended maximum likelihood fit of this peak with individual parameters for the detector resolutions, peak amplitudes, and position and with common parameters for the peak-background ratio [46]. We then project these resolutions with a global scaling factor  $s = \sigma_{\text{phys}}(3034 \text{ keV})/$  $\sigma_{\rm cal}(2615 \text{ keV})$  (common to all datasets and detectors) to  $Q_{\beta\beta}$ . In addition to the method described in Ref. [46], we extract s from a polynomial fit of the global  $\gamma$  peaks in background and calibration data [17,27]. We adopt the scaling factor from this latter method as a conservative choice, predicting a 0.2% worse resolution of  $[7.6 \pm$  $0.7(\text{stat}) \pm 0.2(\text{syst})$  keV FWHM at  $Q_{\beta\beta}$  for the overall data taking. The noted systematic uncertainty of 2% is due to pile-up-related non-Gaussian tails in calibration data that affect the calibration resolution estimates through the PC analysis cut.

The background index has been evaluated from the stillblinded data with a phenomenological fit model that contains an exponential to approximate both the highenergy part of the  $2\nu\beta\beta$  spectrum as well as tails from U/Th contaminants in the setup and a constant as a conservative estimate for the coincident detection of two  $2\nu\beta\beta$  events in the same crystal, remaining unvetoed muon events and close contamination from the high-energy beta decays in the natural U/Th chains. The result of an unbinned extended maximum likelihood fit is strongly dependent on the low-energy and high-energy limit of the fit range. For a fit with the low-energy limit varied from 2.65 to 2.9 MeV and the high-energy limit from the upper end of the blinded region to 4 MeV, we obtain a background index of  $2 \times 10^{-3}$  counts/(keV × kg × yr) to  $6 \times 10^{-3}$  counts/(keV × kg × yr) in a 10 keV window around  $Q_{\beta\beta}$ . Considering the large remaining uncertainty, we round the background index for the ROI optimization to  $b = 5 \times 10^{-3}$  counts/(keV × kg × yr). We model the background as locally flat, consider detector- and dataset-based resolutions, and simulate the  $0\nu\beta\beta$  peak containment in our GEANT4 Monte Carlo model. As this background index is both poorly constrained and indicative of a most probably background-free  $0\nu\beta\beta$  search, we select the ROI maximizing the mean-limit-setting sensitivity for a Poisson process with zero background:

$$\overline{S_{90}} = \sum_{i=0}^{\infty} P(i, b, \Delta E_{\text{ROI}}) \cdot S_{90}(i)$$

with the sum running over the product of the Poisson probability  $P(i, b, \Delta E_{ROI})$  of obtaining *i* events for an ROI with width  $\Delta E_{\text{ROI}}$  and a background index b times the expected classical 90% confidence exclusion limit  $S_{90}(i)$ . We transfer this maximization from the optimization of the energy range for a peak search in 19 (detectors) times 7 (datasets) to the optimization of a single parameter by splitting the simulated smeared  $0\nu\beta\beta$  peaks into 0.1 keV bins and ranking each bin associated with a triplet (detector, dataset, energy) in signal-background (B/S) likelihood space. The optimal cutoff parameter (B/S)<sub>cutoff</sub> results in an ROI that is on average (exposure weighted) 17.9 keV wide. It has a mean signal containment of 75.8% with a spread of  $\pm 1.0\%$ . The ROI width corresponds to an average 2.7 $\sigma$  Gaussian coverage with the loss of  $0\nu\beta\beta$  decay events in the full energy peak dominated by events with energy loss from bremsstrahlung and electron escape close to the surface of the crystals. The optimization exhibits only a mild dependence on the background index or the knowledge of the resolution, with the overall containment changing by  $\pm 0.7\%$  for a 50% change in b (2.2 keV wider, 1.5 keV narrower ROI). We truncate the computation of the mean-limit-setting sensitivity after the first three terms, as the probability of three or more background events is negligible for the considered ROI.

As the discussed Poisson sensitivity is by construction applicable only for limit setting, we implemented a binned likelihood analysis instead to extract either the final limit or a potential signal on the rate of  $0\nu\beta\beta$  events. This analysis is built on the Bayesian analysis toolkit [54] and considers both the signal region as well as the sidebands of our 100-keV-wide blinded region. The likelihood function

$$L = \prod_{i=1}^{3} \frac{e^{\lambda_i} \lambda_i^{n_i}}{n_i!}$$

is the product over three Poisson terms for the two sidebands and the signal ROI with observed events  $n_i$ and expected events  $\lambda_i$ . The mean number of expected events  $\lambda_i$  is computed considering the phenomenological background model described above and a Gaussian signal contribution, in which we leave the strength of the signal and flat background component free by using uninformative flat priors. After defining all analysis steps, we unblind and obtain the spectrum in Fig. 2. We observe no event in the signal region and a single event (cyan) in the right-hand side region. The corresponding marginalized posterior distribution for the number of signal events has a most probable value of zero with an upper limit of 2.4 events at 90% C.I., resulting in a half-life limit for  $0\nu\beta\beta$ decay in <sup>100</sup>Mo of  $T_{1/2}^{0\nu} > 1.4 \times 10^{24}$  yr (90% C.I.). The posterior for the flat background is nonzero with a  $1\sigma$ interval of  $3^{+7}_{-3} \times 10^{-3}$  counts/(keV × kg × yr), and the posterior distributions for the parameters of the exponential are compatible with priors from a fit of the  $2\nu\beta\beta$  spectrum in the 2650–2980 keV interval. We repeat the same fit for



FIG. 2. Physics spectrum for 2.16 kg × yr of data after unblinding. No event is observed in the detector- and dataset-based ROI. A single event, highlighted in cyan, has been observed in the analysis region. In a further refinement of the analysis, it was identified as a  $\beta$  candidate out of the <sup>212</sup>Bi  $\rightarrow$  <sup>208</sup>Tl  $\rightarrow$  <sup>208</sup>Pb part of the natural decay chain (see the text). For visualization, the exposure-weighted mean ROI for  $0\nu\beta\beta$  decay (17.9 keV wide) has been indicated with solid black lines.

TABLE I. Nuisance parameters included in the analysis and their implementation with flat or Gaussian prior in the Bayesian fit. Parameters 2 and 4 are multiplicative scaling factors instead of absolute uncertainties; see the text for details.

Systematic	Index	Value	Prior
$0\nu\beta\beta$ detector response	1	0.95-1.00	Flat
$0\nu\beta\beta$ containment MC	2	$1.000\pm0.015$	Gauss
Analysis efficiency <sup>a</sup>	3	$0.906\pm0.004$	Gauss
Light yield selection <sup>a</sup>	4	0.998-1.008	Flat
Isotopic enrichment	5	$0.966\pm0.002$	Gauss

<sup>a</sup>Dataset-dependent; exposure-weighted mean value presented.

the approximation of a Gaussian signal with a locally flat background over the 100 keV analysis region. The limit on  $0\nu\beta\beta$  decay of <sup>100</sup>Mo is unaffected.

The nuisance parameters considered in this limit are summarized in Table I. Uncertainties on the detector response, in particular, the energy scale and resolutions, are included in the simulation of  $0\nu\beta\beta$  events. They are, hence, covered in the resulting containment in the optimized central ROI on a detector and dataset basis and not considered independently. The only remaining uncertainty for the detector response (index 1, Table I) is based on a potential non-Gaussianity of the  $0\nu\beta\beta$  peak. In this analysis, we estimate this contribution based on the shape of the 2615 keV calibration peak. We observe evidence for non-Gaussian tails, which are dominated by unrejected pile-up events caused by the high trigger rate in calibration data. We set a conservative systematic on the containment reduction of up to 5%. The second nuisance parameter on the containment (index 2) accounts for the GEANT4 modeling uncertainty of bremsstrahlung events. Reported accuracies for the GEANT4 bremsstrahlung production of a few MeV electrons in thick targets [55,56] of ~10% result in a systematic uncertainty in the overall containment of the  $0\nu\beta\beta$  signal in the optimized ROI of  $(75.8 \pm 1.1)\%$  for our crystal geometry, which is applied as a single common multiplicative factor of  $1.000 \pm 0.015$  in the limit setting. The inclusion of the analysis efficiency

$$\epsilon = [90.6 \pm 0.4(\text{stat})^{+0.8}_{-0.2}(\text{syst})]\%$$

is split into two parts. For the evaluation of the mean value and its statistical uncertainty (index 3), we make use of the two independent signals in the LDs and LMOs to evaluate cut efficiencies on a clean sample of signal events in the 1.3–2 MeV  $2\nu\beta\beta$  spectrum or from the <sup>210</sup>Po peak [46]. Energy-independent cuts are evaluated directly from the ratio between passed and total events with binomial uncertainty. The pulse shape analysis efficiency is extracted from a linear fit extrapolated to  $Q_{\beta\beta}$  in order to account for the energy dependence in the reconstruction error. The systematic uncertainty associated with the excess broadening of the light yield cut has been evaluated with a set of pseudo-experiments considering the linear and modified photon statistics model introduced before. It is reflected in our limit setting as a multiplicative factor with uniform prior in 0.998–1.008 (index 4). Lastly, we include a subdominant uncertainty in the enrichment and number of <sup>100</sup>Mo atoms of 0.2% (index 5).

We further refined our analysis after unblinding, implementing a cut designed to reject high-energy  $\beta$  events from the <sup>212</sup>Bi<sup> $\vec{\alpha}$  208</sup>Tl<sup> $\vec{\beta}$ </sup> <sup>208</sup>Pb branch in the thorium chain  $(T_{1/2}^{208\text{Tl}} = 183 \text{ s}, 5 \text{ MeV } Q \text{ value})$ . Similar to previous analyses with scintillating bolometers [28,31,45], we tag <sup>212</sup>Bi  $\alpha$  candidates with energies in the 6.0–6.3 MeV range and veto any decay in the same crystal in a 10-half-life period (1832 s). This cut has a negligible impact on the lifetime (0.02%) accidentally rejecting 2:10 000 events, but it does reject the event close to the ROI in cyan in Fig. 2. The energy of the preceding  $\alpha$  candidate is consistent with the Q value of <sup>212</sup>Bi within 10 keV, and the time difference between the events is 113 s. We report a final  $0\nu\beta\beta$  limit that is 1.3% stronger and rounds to

$$T_{1/2}^{0\nu} > 1.5 \times 10^{24} \text{ yr}(90\% \text{ C.I.}).$$

The posterior for the flat background of the Bayesian fit in this case is peaked at zero with a 90% C.I. of  $1.1 \times 10^{-2}$  counts/(keV × kg × yr).

We interpret the obtained half-life limit in the framework of light Majorana neutrino exchange using  $g_A = 1.27$ , phase space factors from Refs. [57,58], and nuclear matrix element calculations from Refs. [59-66]. The resulting limit on the effective Majorana neutrino mass of  $\langle m_{\beta\beta} \rangle < (0.31-0.54)$  eV is the fourth most stringent limit worldwide, obtained with a modest <sup>100</sup>Mo exposure of 1.17 kg  $\times$  yr. It is the leading constraint for <sup>100</sup>Mo, exceeding the previous best limit from NEMO-3 [40] by 30% with almost 30 times lower <sup>100</sup>Mo exposure. The technology of CUPID-Mo has proven that it can be operated reliably and reaches high efficiency for a  $0\nu\beta\beta$  search of 68.6% (containment  $\times$  analysis efficiency) and a resolution of  $0.11\%(1\sigma)$  at  $Q_{\beta\beta}$ . The present analysis strengthens the projection of the CUPID sensitivity [38], by demonstrating a detailed understanding of the  $0\nu\beta\beta$  ROI and confirming key assumptions like the efficiency of Li2<sup>100</sup>MoO<sub>4</sub>-based cryogenic scintillating bolometers. Extremely low U/Th contamination levels in the LMO crystals reported in Ref. [46] surpass the requirements for CUPID [38], and an efficient alpha separation has been demonstrated both in cylindrical [34,36] and recently also in cubic LMO detectors [67,68]. The preliminary estimate of the background in the ROI at the few  $10^{-3}$  counts/(keV × kg × yr) level in CUPID-Mo, obtained in an experimental setup that was not designed for a  $0\nu\beta\beta$  search, is encouraging and supports our believe that a  $10^{-4}$  counts/(keV × kg × yr) background level for CUPID [38] seems feasible.

Further analyses from CUPID-Mo will be focused on precisely reconstructing remaining backgrounds, comparing to the best reported background index for a bolometric  $0\nu\beta\beta$  search  $[3.5^{+1}_{-0.9} \times 10^{-3} \text{ counts}/(\text{keV} \times \text{kg} \times \text{yr})]$  in CUPID-0 [31,32] and to optimally design and use the technology of the CUPID-Mo experiment in CUPID [38].

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