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An Improved $ZnMoO_4$ Scintillating Bolometer for the Search for Neutrinoless Double Beta Decay of ^{100}Mo

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Abstract We present a prototype scintillating bolometer for the search for neutrinoless double β decay of ^{100}Mo, consisting of a single $\approx$5 g $ZnMoO_4$ crystal operated aboveground in the 20–30 mK temperature range. The scintillation light is read out by two thin Ge bolometers. The phonon signals are collected by NTD Ge thermistors. The $ZnMoO_4$ crystal was grown with an advanced method (low-thermal-gradient Czochralski technique) and after purification of molybdenum. The results are very encouraging: the intrinsic energy resolution of the heat channel is $\approx$800 eV FWHM, the α/β rejection factor (crucial for background suppression) is better than 99.9% in

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the region of interest for double β decay ($\approx$3 MeV), and the radiopurity of $ZnMoO_4$ looks substantially improved with respect to previous devices.

Keywords Double beta decay · Neutrino mass · Low background · Bolometric technique · $ZnMoO_4$ crystals

1 Introduction and Motivations

Neutrinoless double β decay ($0\nu\beta\beta$) is a very rare nuclear transition which violates by two units the total lepton number conservation [1]. If observed, this process would shed light on crucial neutrino properties, enabling in principle the determination of its mass scale and the ordering of the three mass eigenstates. In addition, its detection would unquestionably prove that neutrinos are self-conjugate particles [2]. The experimental signature of $0\nu\beta\beta$ is a peak in the spectrum of the sum of the energy of the two β electrons emitted in the decay, located at the Q-value of the transition. Low temperature calorimeters were proposed more than 20 years ago as sensitive devices for the study of $0\nu\beta\beta$ [3]. Currently, experiments based on them are among the most sensitive in the world for the study of this phenomenon and among the most promising for the immediate future. Moving from simple low-temperature macrocalorimeters to hybrid detectors capable to detect heat and scintillation light would further increase the potential of the bolometric technique, providing a viable technology for next-generation searches [4]. The double read-out in fact enables an excellent discrimination of the α background, which is expected to be dominant in the region above 2615 keV (high energy limit of the natural γ radioactivity), where the Q-value of several interesting $0\nu\beta\beta$ candidates lays.

We present here the performance of a $ZnMoO_4$ scintillating bolometer prototype, realized in view of a frontier $0\nu\beta\beta$ experiment focused on the study of ^{100}Mo, characterized by a large transition energy ($Q_{2\beta} = 3034.40(17)$ keV). The same isotope is studied with a similar approach in the AMoRE experiment, which employs crystals of $CaMoO_4$ [5].

2 Description of the Detector Prototype

$ZnMoO_4$ crystals of advanced quality were developed in the Nikolaev Institute of Inorganic Chemistry (Novosibirsk, Russia). The crystal synthetization was preceded by a deep chemical purification of molybdenum (in the form of MoO_3), in order to get rid of metal contamination, which may compromise the scintillation yield of the crystals. After purification, the content of Na, Ca, Fe, Pb in the MoO_3 was at the level of <1 ppm. Contamination of other elements did not exceed the level of 0.1–0.001 ppm. Commercial zinc oxide with a content of impurities <1.5–0.1 ppm was used to obtain the $ZnMoO_4$ compound. $ZnMoO_4$ crystals up to 25 mm in diameter and 60 mm in length were grown by the low-thermal-gradient Czochralski technique in platinum crucible of size $\oslash$ 40 $\times$ 100 mm [6]. Clear crystals were obtained, while

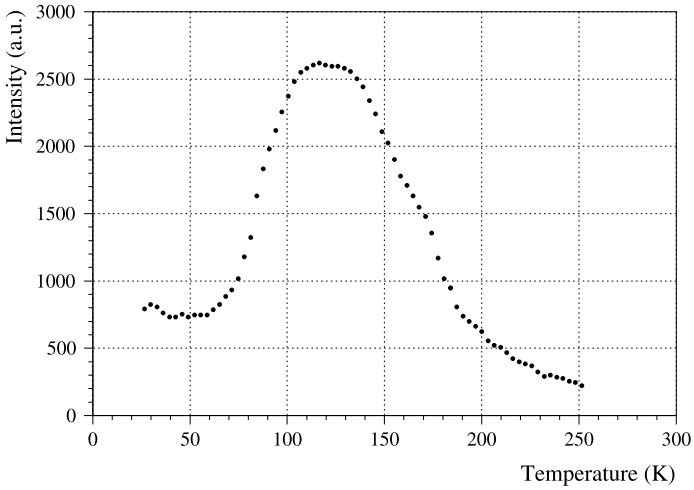

Fig. 1 Relative integral intensity of X-ray luminescence as a function of the temperature

in previous tests the $ZnMoO_4$ samples exhibited a pinkish or yellowish hue, with a broad absorption band with a maximum at ≈430–450 nm, which is now no longer detectable in transmittance measurement.

X-ray luminescence was also investigated at 8, 118 and 290 K. The emission spectrum exhibits a broad band in the visible region with a maximum at 610 nm at room temperature, which shifts to 625 nm at 8 K. The integral luminescence is strongly quenched at room temperature, reaches a maximum around 110–140 K and then drops with cooling, tending to stabilize below 60 K at a factor ≈4 lower than the maximum (see Fig. 1). Thermostimulated luminescence at ≈75 K and phosphorescence indicates the presence of defects and impurities in the crystal, showing that there is still a considerable potential to improve the quality of the material.

After these preliminary investigations, described in much more detail in a work in preparation, a prototype was designed, fabricated and cooled down, with the aim to investigate the quality of $ZnMoO_4$ as bolometric material, its light yield at low temperatures, the α/β rejection factor and the radiopurity of a sample crystal grown with an advanced approach after a special procedure of molybdenum purification. The tests have been performed aboveground in the cryogenic laboratory of the University of Insubria (Como, Italy) and in the Centre de Spectrométrie Nucléaire et de Spectrométrie de Masse (Orsay, France).

The energy absorber of the assembled detector (see Fig. 2) was a rectangular single $ZnMoO_4$ crystal, with size $15 \times 15 \times 5$ mm. It was contained in a cylindrical copper holder, acting as a heat sink for the detector. The mechanical coupling to the holder was supplied by four PTFE elements. On the two opposite 15×15 mm sides, the $ZnMoO_4$ crystal was faced by two light-detecting square ultrapure Ge slabs, 15 mm side and 0.5 mm thickness. The light detectors were kept in position by two PTFE pieces, clamping the slabs on two opposite edges.

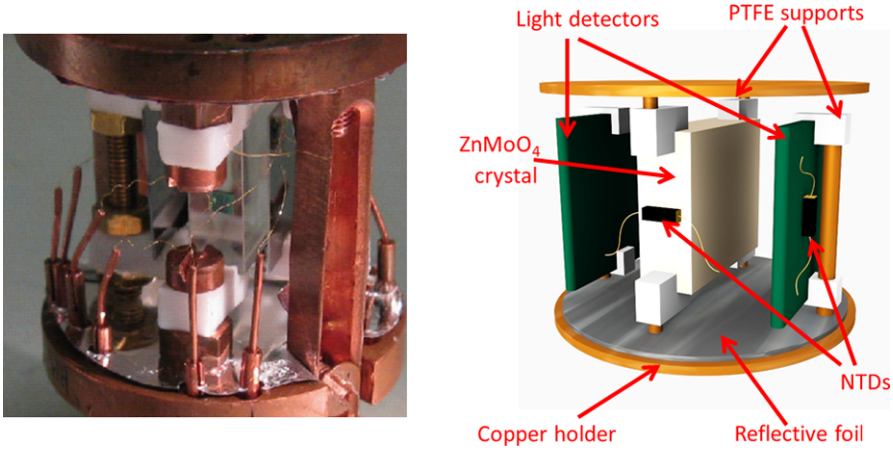

Fig. 2 (Color online) A photo and a scheme of the detector prototype described in the text

The thermal signals from the $ZnMoO_4$ crystal and the two Ge slabs were provided by three nominally identical $3 \times 1 \times 0.6$ mm neutron transmutation doped Ge thermistors. The thermistor contacts coincide with two opposite 1×0.6 mm sides. The Efros-Shklovskii Variable Range Hopping parameters T_0 and ρ_0 [7] are respectively 3.83 K and 1.09 Ω mm. The thermistors were thermally connected to the energy absorber through a two component epoxy (*Araldite rapide*). In order to well define the contact area with the energy absorbers, the thermistors presented a 50 μm thick stand-off, with 1×1 mm area, placed at the center of one of the 3×1 face. The epoxy was deposited as a small dot on this stand-off, and then pressed in order to form a thin veil not overflowing from the 1×1 mm region. The three thermistors were electrically connected by means of golden wires with ⊘ 50 μm and about 15 mm long. The wires are ball-bonded at the thermistor pads at one end and crimped in a copper tube at the other end, providing the main thermal link to the heat sink. The internal wall of the holder was covered with a highly reflective polymeric multilayer foil (VM2000/VM2002, 3 M) to improve light collection.

In some runs, radioactive sources were placed in the vicinity of the detector for calibration purposes. In particular, a collimated ^{241}Am source, characterized by a main α line at 5.48 MeV and an intense γ line at 59 keV, illuminated the $ZnMoO_4$ crystal at the center of a 15×5 mm face. Two weak ^{55}Fe sources, providing X-rays at 5.9 keV and 6.4 keV, irradiated the external side of each Ge slab.

3 Experimental Results

During the cold tests, the copper holder was placed in the experimental vacuum of two dilution refrigerators (Como and Orsay), in thermal contact with the mixing chamber. The detector was operated at various base temperatures, between 25 and 32 mK. The typical NTD thermistor resistances at the operation points which provided the

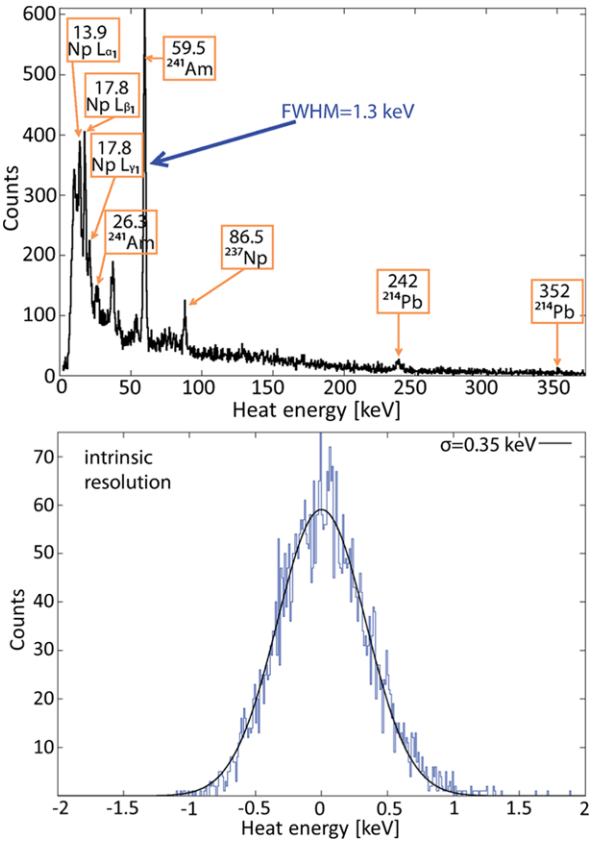

Fig. 3 (Color online) The energy resolution of the heat channel. (*Top*) Low energy γ spectrum. (*Bottom*) Baseline width, corresponding to the intrinsic energy resolution

best performance were of the order of 1 MΩ, with bias currents ranging between 2 and 5 nA. For the electronic readout, the NTD Ge thermistors were connected to low noise voltage amplifiers, which presented a cold stage inside the cryostat in Orsay only. The bias current was injected through two room-temperature load resistors and a DC voltage source in the Como set-up. In Orsay, the detector was AC-biased with a 2 kHz square-wave excitation. Both set-ups provided similar performance in terms of energy resolution and α/β rejection factor. The full waveforms of the signal were acquired and registered, and the method of the optimum filter was applied off-line in order to optimize the energy resolution.

The signals from the $ZnMoO_4$ crystal (the heat channel) corresponded to voltage pulses with an amplitude of ≈200 μV for 1 MeV energy deposited by βs or γs. The signal time structure was characterized by ≈1 ms rise time (from 10% to 90% of the signal amplitude) and ≈10 ms decay time (from 90% to 30% of the signal maximum amplitude). The long decay time with respect to a conventional nuclear detector forced us to perform always low rate calibration, not exceeding ≈0.1 Hz. The detector time behavior is consistent with the thermal network representing the detector, and corresponds to what commonly observed in dielectric macrobolometers operated in similar conditions. The position of the 5.48 MeV α peak of the ^{241}Am

source on an energy scale calibrated with γ lines shows that the phonon yield of an α particle is larger than that of a γ or a β by a factor ≈ 1.1, as observed in other scintillating bolometers [8]. This effect prevents from evaluating the detector non-linearity over its full operational range by comparing γ and α line positions.

The intrinsic energy resolution of the heat channel (corresponding to the FWHM baseline fluctuations) is of the order of 800 eV (see Fig. 3-bottom). The energy resolutions on the lines is worse (this is commonly observed in macro-bolometers), but it is always excellent, of the order of 1.3 keV FWHM at the 59 keV ^{241}Am line (see Fig. 3-top) and, at higher energies, of 3.8 keV FWHM at 2615 keV ^{208}Tl line. This performance shows that $ZnMoO_4$ is a very promising material for high energy resolution bolometers: behaviors similar to those observed in large mass Ge or TeO_2 bolometers are expected in much larger crystals.

The energy calibration of the light detectors was performed using the ^{55}Fe sources. The signals presented pulse amplitudes in the range $\approx 1-2$ μV for 1 keV deposited energy. The baseline width of the best light detector was $\sigma = 31$ eV. However, the light detectors were hardly able to resolve the 5.9 keV and 6.4 keV doublet. This excess line spread is not understood at the moment.

Observing the time coincidences between signals collected with the Ge slabs and the $ZnMoO_4$ crystal, and thanks to the energy calibration of the heat and light channels, it was possible to deduce the light yield for α and β-like particles (β, γ and cosmic muons). It resulted respectively to be 2.08 keV/MeV for β and 0.42 keV/MeV for α particles (corresponding to 1048 and 212 photons/MeV respectively assuming an average photon energy of 1.98 eV), consistent with previous measurements for this material [9]. These values take into account the light collected by both light detectors (which is the same in each of them for the same event in the $ZnMoO_4$ crystal within the light detector energy resolution), but not the light collection efficiency. Therefore, they are inferior limits. Even considering this, it is clear that $ZnMoO_4$ is a very poor intrinsic scintillator. However, the light output is more than enough to perform efficiently α/β discrimination. A rejection factor better than 99.9% has been determined at the energy of interest for ^{100}Mo $0\nu2\beta$ (see Fig. 4-top). As already observed in previous works [10], in scintillating bolometers α and β particles can be distinguished using the heat channel only, on the basis of a different temporal structure of the phonon signal. This is the case for our prototype too. The main difference in the pulse time behavior was observed both in the rise time and decay time, which is some % shorter for α particles. This behavior can be used for rejection purposes. We have constructed the sum over the sampling points of the squares of the differences between each pulse and the average pulse (once normalized). The distribution of this shape parameter (see Fig. 4-bottom) allows to achieve easily the same information provided by the simultaneous detection of heat and light, with a rejection factor exceeding 99.9% in this case as well. Even if in principle this would allow to get rid of the light channels in a future $0\nu2\beta$ experiment, we are persuaded that a redundancy of the methods for the rejection is welcome.

In order to determine the intrinsic contamination of the crystal, we have performed a run without the ^{241}Am α source. Over an exposure of 25.6 g $\times$ days, we observed hints for a surface contamination only, probably in ^{210}Po or ^{210}Pb. Since there is no

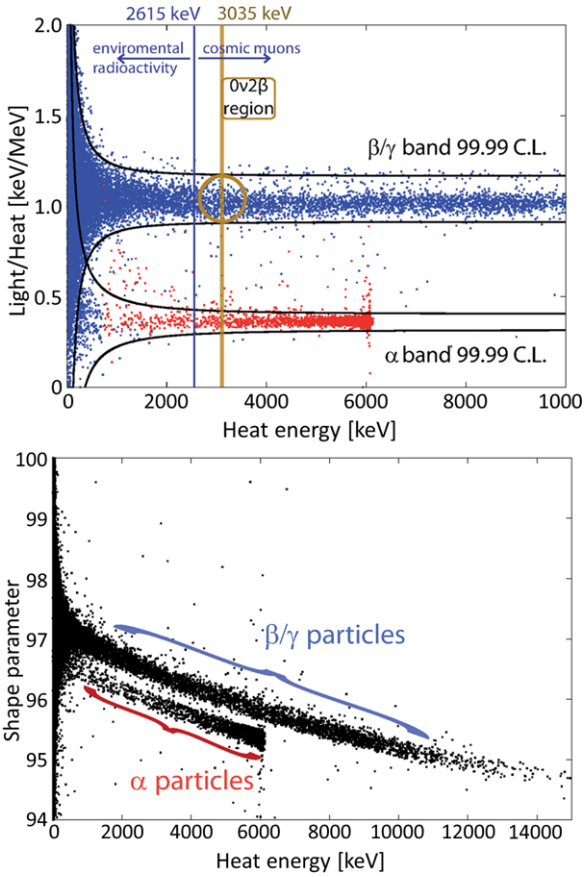

Fig. 4 (Color online) The α/β discrimination capability of the detector prototype. (*Top*) The light-to-heat pulse amplitude ratio (for one light detector) plotted versus heat energy (γ calibration). (*Bottom*) Pulse shape parameter (defined in the text) versus the heat energy (γ calibration). The *red* (*blue*) points in the top graph correspond to α (β-like) particles selected via pulse shape as shown in the bottom graph

evidence of internal α peaks, it is possible to set limits on the contamination of several nuclides belonging to the natural radioactivity chains looking at their first α decay. When the expected α peak is above 5.5 MeV, we have used both background and calibration spectrum in order to increase the exposure up to 43.1 g $\times$ days. For the isotopes ^{232}Th, ^{228}Th, ^{227}Ac, ^{238}U, ^{234}U, ^{230}Th, ^{226}Ra and ^{212}Bi an upper limit (at 90% c.l.) of respectively 3.3 mBq/kg, 1.1 mBq/kg, 1.1 mBq/kg, 2.7 mBq/kg, 1.1 mBq/kg, 1.7 mBq/kg, 1.1 mBq/kg and 0.64 mBq/kg can be set. A substantial improvement was observed with respect to a $ZnMoO_4$ crystal tested in a previous work [9], in particular for the isotope ^{226}Ra, which exhibited an internal activity—now absent—of 8.1 ± 0.3 mBq/kg. Above 5.5 MeV and therefore out of the region affected by the common contaminants ^{210}Po and/or ^{210}Pb, the α background spectrum is empty. In particular, the result on ^{212}Bi (expected peak at 6208 keV) allows to set a limit on the internal activity of its daughter ^{208}Tl, one of the most dangerous contaminant, at 0.23 mBq/kg. In fact, ^{208}Tl is a beta active nucleus with a Q-value of 4.999 MeV, producing therefore γs and βs that are energetic enough to give events around 3 MeV on the whole if they are partially absorbed together, as it may happen when the contamination is inside the $ZnMoO_4$ crystal itself.

4 Conclusions

The results here reported show that arrays of $ZnMoO_4$ scintillating bolometers are viable candidates for a next-generation $0\nu\beta\beta$ experiment with high energy resolution and efficient α background rejection, capable to explore the inverted hierarchy region of the neutrino mass pattern. This point is discussed elsewhere [11], where an extensive evaluation of the expected background, supported by Monte Carlo simulations and reasonable assumptions on the radioactive contaminations, is reported.

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