Production and characterization of high-purity natural and enriched $ZnMoO_4$ crystals to search for neutrinoless double beta decay of ^{100}Mo

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M. Mancuso[1,2], D. M. Chernyak[1,3], F. A. Danevich[3], R. Decourt[4], A. Giuliani[1,2,5], I.M. Ivanov[6], E.P. Makarov[6], P. de Marcillac[7], S. Marnieros[1], S.G. Nasonov[6], C. Nones[8], E. Olivieri[1], G. Pessina[5], D.V. Poda[1], V.N. Shlegel[6], M. Tenconi[1], V.I. Tretyak[3], Ya.V. Vasiliev[6], M. Velazquez[4], V.N. Zhdankov[9]

[1] CSNSM/IN2P3/CNRS and Université Paris XI, Bâtiment 108, 91405 Orsay Campus (France)
[2] Università dell'Insubria and Sezione dell'INFN di Milano-Bicocca , 22100 Como (Italy)
[3] Institute for Nuclear Research, MSP 03680 Kyiv (Ukraine)
[4] ICMCB Bordeaux (France)
[5] Università di Milano-Bicocca e Sezione dell'INFN di Milano Bicocca, I-20126 Milano (Italy)
[6] Nikolaev Institute of Inorganic Chemistry, 630090 Novosibirsk (Russia)
[7] L'Institut d'Astrophysique Spatiale (IAS), Bâtiment 120, 91405 Orsay Campus (France)
[8] Service de Physique des Particules, CEA-Saclay, 91191 Gif sur Yvette (France)
[9] CML Ltd., 630090 Novosibirsk (Russia)

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Abstract

Scintillating bolometers are promising devices for the future experiments on neutrinoless double beta decay (0v2β). When the energy absorber in a bolometer scintillates at low temperatures, the simultaneous detection of scintillation light and heat provides a very powerful tool to identify the nature of the interacting particle and therefore to suppress background. A recently developed technique to grow large high quality radiopure zinc molybdate ($ZnMoO_4$) crystal scintillators makes this material advantageous for low temperature bolometric experiments. This is the case for LUMINEU program which aims to perform a pilot experiment on 0v2β using radiopure ZnMoO4 crystals operated as scintillating bolometers. Growing high quality radiopure crystals is a complex task, since there are no commercially available molybdenum compounds with the required levels of purity and radioactive contamination.

Here we present further progress in deep purification of molybdenum, growing natural and enriched of $ZnMoO_4$ crystals and new results about their optical, luminescent, thermal and bolometric properties.

Production of $ZnMoO_4$ crystals

Purification

Purification of MoO_3 by sublimation

Purification and cristallization were performed at Nikolaev Institute of Inorganic Chemistry (NIIC). To purify molybdenum by sublimation for $ZnMoO_4$ crystal growth we have added up to 1% of high purity zinc molybdate to the MoO_3 prepared for sublimation.

$$\mathbf{ZnMoO_4 + WO_3 \rightarrow ZnWO_4 + MoO_3\uparrow}$$

The sublimates were then annealed in the air atmosphere to obtain yellow color stoichiometric MoO_3.

Purification by recrystallization from aqueous solution

Molybdenum then was purified by double recrystallization of ammonium molybdate in aqueous solutions. We've added ZnO to initiate the precipitation in the basic ammonia solution. After several hours a precipitation of $ZnMoO_4$ occurs and sorbs impurities from the solution. After separation of the sediment the operation was repeated to bind the residuals Fe impurities.

Material	Concentration of impurities (ppm)							
	Na	Mg	Si	K	Ca	Fe	Zn	W
Initial MoO_3	60	1	60	50	60	8	10	200
Recrystallization from aqueous solutions	30	< 1	30	20	40	6	1000	220
Sublimation and recrystallization from aqueous solutions	-	<1	30	10	12	5	500	130
Double sublimation and recrystallization from aqueous solutions	-	< 1	-	< 10	<10	<5	70	<50

Crystal growth

Low thermal gradient Czochralski technique

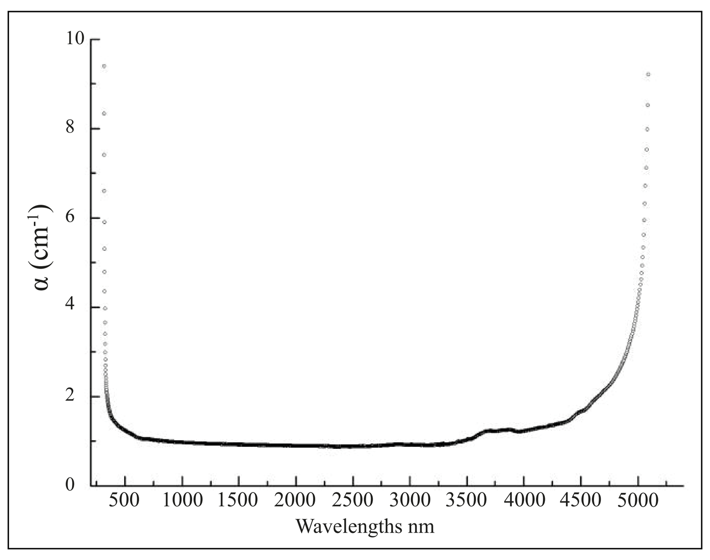
The ZnMoO4 crystals were grown by the Low Thermal Gradient Czochralski Technique (LTG Cz) developed at the NIIC.

In this method the temperature gradients in the melt were decreased to a level of about 1 K/cm and evaporation of melt components was strongly suppressed by a special design of the growth cell.

Characterization of $ZnMoO_4$ crystals

Optical absorption

The measurement was made on a 2.0 mm thick single crystal of $ZnMoO_4$ in a range of light spectra including visible and near infrared wavelengths. In the range of light emittance of $ZnMoO_4$ the thansmission coefficient, T, is about 0.8 and cut-off wavelengths are at 313 nm and 5.13 µm.



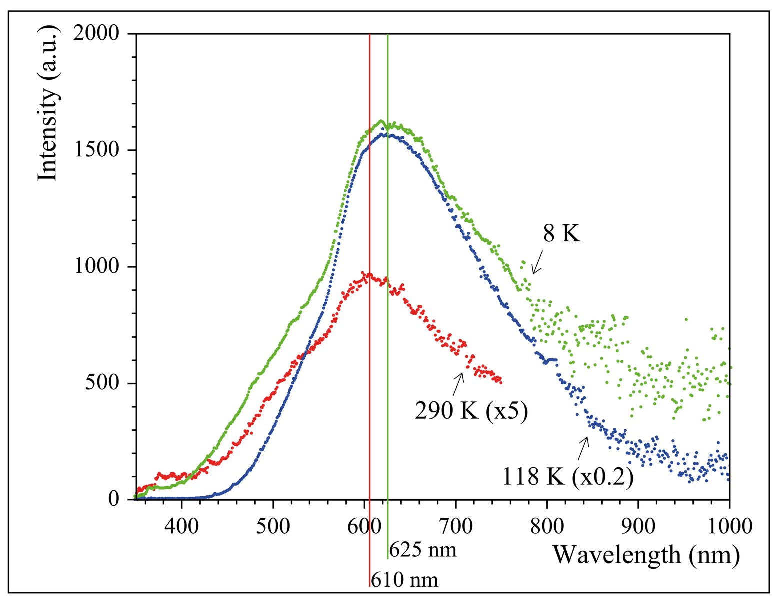
The assorbtion coefficient α is calculated as α = -(logT)x(ln10)/t, where t is the thickness of the crystal and α is in the range of 1.47 to 0.89 cm^{-1} in the wavelength region from 400 nm to 2 µm.

- **The absence of a broad assorbtion band around 440 nm shows a low contamination due to Fe^{+2}/Fe^{+3}.**
- **The refractive index at 650 nm is 1.96 in agreement with the literature.**

Luminescent under X-ray excitation

The luminescence of $ZnMoO_4$ crystal was investigated in the temperature interval 8–290 K under X-ray excitation.

A broad band in the visible region with a maximum at 610 nm was observed at room temperature. At 8 K luminescence exibits an emission band with a maximum at 625 nm.



The light output grows with decreasing temperature, reaches a maximum around 110–140 K and then drops with further cooling. This result is in agreement with the data of previous investigations.

- **The crystals are transparent to their emitted light**

Specific heat measurement

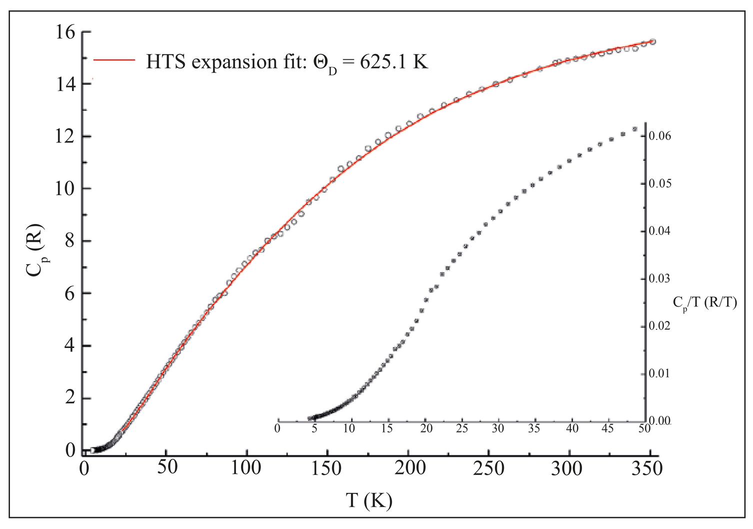
Specific heat measurements were made on a 3x3x2 mm^3 single crystal. The specific heat could be approximated for temperatures higher than ~23 K using high-temperature series expansion:

$$C_p \propto 1 + \sum_{i=1}^{4} B_i \left[1 + \left(2\pi \frac{T}{\Theta_D}\right)^2\right]^{-i}$$

Debye temperature: Θ_D = 625. 1 K

Bernoullii numbers: B1 = 1.9091 , B2 = 1.86714

B3 = -0.96009 , B4 = -0.00907



The inset shows Cp/T as a function of T at low temperatures to evidence the absence of any low range order down to 4 K.

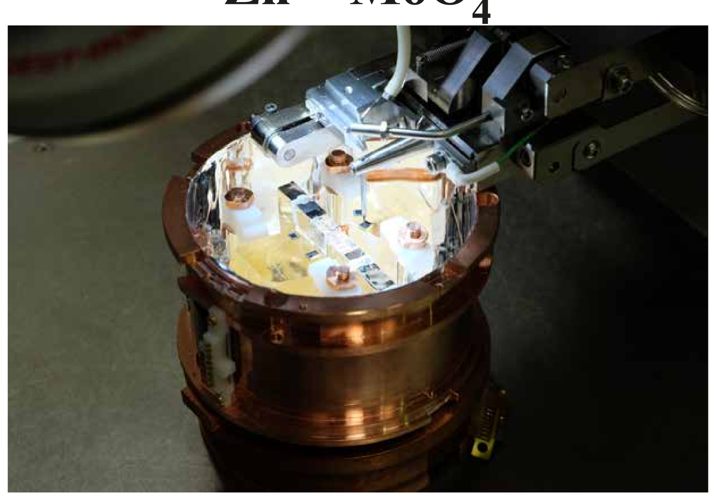
- **Debye temperature measurement was performed for the first time for $ZnMoO_4$ and it's favorable for bolometric application**

Bolometric Test

The detectors

$ZnMoO_4$

$Zn^{100}MoO_4$



$ZnMoO_4$ detectors:

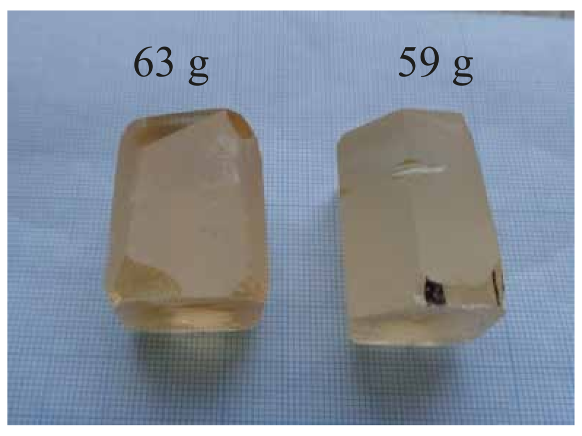
- 2 natural $ZnMoO_4$ about 330g each
- 2 enriched $Zn^{100}MoO_4$ about 60g each
- PTFE provide mechanical coupling with the copper holder
- The temperature read-out is provide by NTD Ge thermistor
- A heater element is used to stabilize the detectors response

Light detectors:

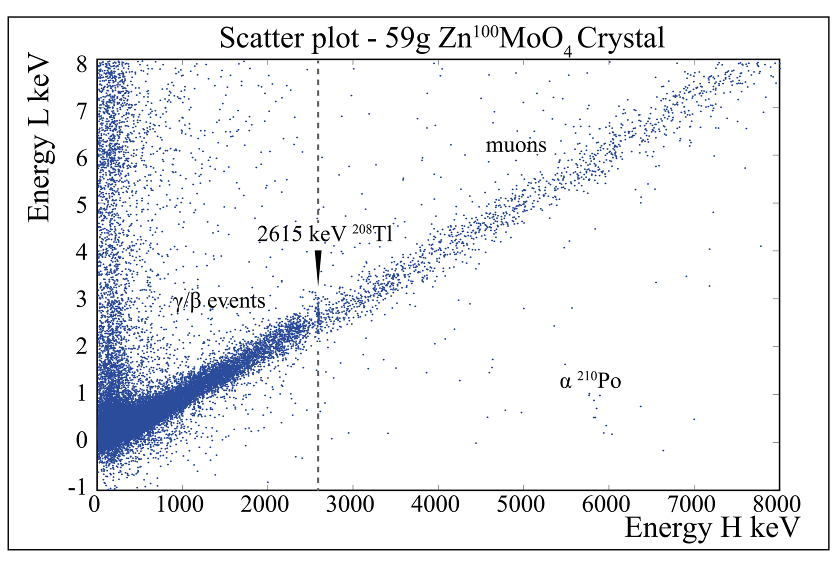
- Hyper-pure Ge slab
- Three PTFE clamps provide mechanical coupling and a weak thermal link towards the copper holder
- The temperature read-out is provide by NTD Ge thermistor
- A heater element is used to stabilize the detectors response
- One side is coated with SiO to increase the collected light

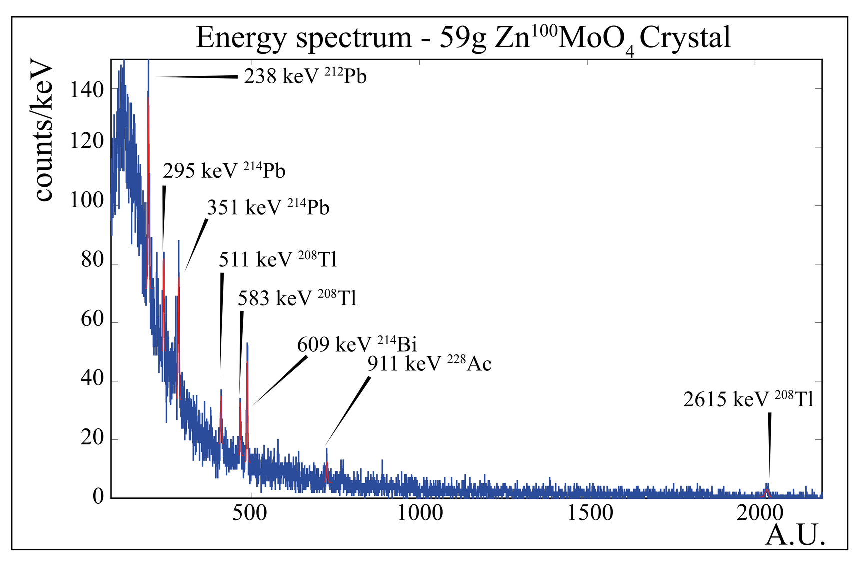
- **The detectors are installed in the cryogenic setup in the underground lab of Modane (France) and are ready for data taking.**
- **The mass content in ^{100}Mo of these detectors is 81.1 g.**

Above-ground performances of enriched crystals at CSNSM



$Zn^{100}MoO_4$	59 g	63 g
Working Temperature	13.7 mK	13.7 mK
Signal	87 µ V/MeV	96 µ V/MeV
$FWHM_{Baseline}$	1.4(1) keV	1.8(1) keV
Light $Yield_{\gamma/\beta}$	1.01(11)	0.93(11)
τ_{rise} in 0.55-2.65 MeV	9 ms	5.5 ms
τ_{decay} in 0.55-2.65 MeV	46.3 ms	26.2 ms





- **^{100}Mo enriched crystals show very good performances as bolometers. In the undroground setup the energy resolution is close to the baseline FWHM.**

Conclusions and Perspectives

We present here further improvement in the production and characterization of $ZnMoO_4$ crystals for LUMINEU program.

- **We developed a technique for the production of high quality large mass $ZnMoO_4$ crystals**
- **We proved the possibility to use enriched material in large experiments for the search for 0v2β of ^{100}Mo**
- **A new bunch of enriched $Zn^{100}MoO_4$ crystals with masses of the order of 300-400g is in production at NIIC**
- **The crystal properties are fully characterized and they are favorable for bolometric experiments**
- **2 large natural and enriched $ZnMoO_4$ scintillating bolometers are now under investigation in the underground laboratory of Modane (France)**

References:
[] L. Bergé et al., Purification of molybdenum, growth and characterization of medium volume ZnMoO_4 crystals for the LUMINEU program, JINST, in the press
[] A.S. Barabash et al., Enriched Zn100MoO 4 scintillating bolometers to search for 0v2β decay of 100Mo with the LUMINEU experiment