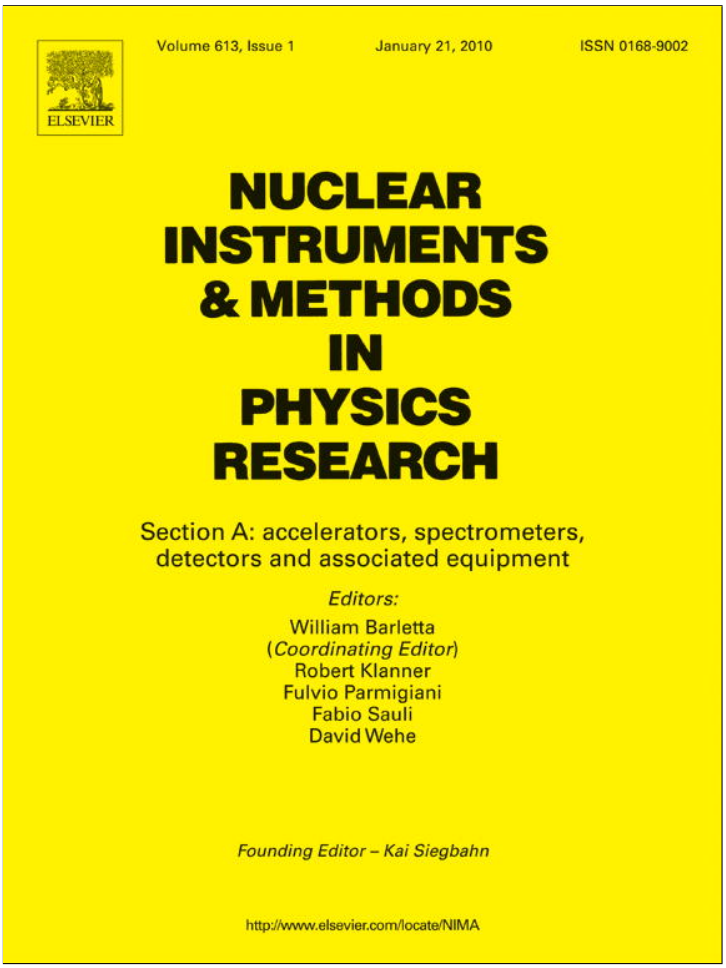

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First test of Li_2MoO_4 crystal as a cryogenic scintillating bolometer

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ABSTRACT

Luminescence properties (emission spectra, thermally stimulated luminescence, dependence of intensity on temperature) of lithium molybdate (Li_2MoO_4) single crystal have been studied under X-ray excitation from the liquid nitrogen to room temperature. The applicability of Li_2MoO_4 single crystal as a cryogenic scintillating bolometer was checked for the first time. Both scintillation and heat signals were observed making Li_2MoO_4 material an interesting candidate for searches for neutrinoless double beta decay of ^{100}Mo.

1. Introduction

Neutrinoless (0ν) double beta (2β) decay of atomic nuclei is a unique nuclear process able to provide very important information about the neutrino and weak interactions: the nature of the neutrino (Majorana or Dirac particle), the hierarchy and absolute scale of neutrino mass, lepton number conservation, the presence of right-handed current in weak interactions, and the existence of Majorons [1]. Recent observations of neutrino oscillations (see f.e. summary [2]) give clear evidence that the neutrino is a massive particle. The present situation calls for high sensitivity experiments to search for 2β processes in different nuclei using different experimental approaches.

^{100}Mo is one of the most promising 2β isotopes due to its high transition energy ($Q_{2\beta} = 3035$ keV [3]) and relatively high natural isotopic abundance (9.67% [4]). The high $Q_{2\beta}$ value leads to a large phase-space integral and thus to a comparatively high probability of decay. From the experimental point of view, the high $Q_{2\beta}$ value simplifies the problem of background caused by natural radioactivity.

The best limit to-date on the 0ν2β decay of ^{100}Mo comes from the NEMO-3 experiment, where $\simeq 7$ kg of enriched ^{100}Mo are used; after 389 days of data taking the limit $T^{0\nu}_{1/2} > 4.6 \times 10^{23}$ yr at 90% CL was reached [5]. However, some disadvantages of the NEMO-3 experiment are low energy resolution (about 10% at the energy of the ^{100}Mo double β transition), and a low detection efficiency of 0ν2β events (about 14%). Therefore detectors which contain the ^{100}Mo nuclei, with a sufficiently large detection efficiency of the 0ν2β signal,[1] are of considerable interest: in fact 1.2 kg of ^{100}Mo in a crystal scintillator will be equivalent to 7 kg of ^{100}Mo in the NEMO-3 experiment. Cryogenic scintillation bolometers (see f.e. [8]) with a typical energy resolution of a few keV in a wide energy region are the most promising technique for future 0ν2β decay experiments exploring the inverted hierarchy of neutrino mass (half-life sensitivity on the level of $10^{26}-10^{27}$ years). Furthermore the energy resolution (a few keV) needed to go towards the normal hierarchy of the neutrino mass (half-life sensitivity on the level of $10^{28}-10^{30}$ years) could be achieved only by bolometers and semiconductors [9]. Scintillating bolometers also have an excellent particle discrimination ability, allowing the effective suppression of the α particles background.

There exist a few crystals containing molybdenum. The most promising of them are $CaMoO_4$ [10–12], $CdMoO_4$ [13,14], $PbMoO_4$ [14–16], $Li_2Zn_2(MoO_4)_3$ [17], $ZnMoO_4$ [18–20]. However, all the listed crystal scintillators have some disadvantages:

[1] For instance the detection efficiency is ≈86% in the CUORICINO experiment with 0.75 kg tellurium oxide crystals operating as low temperature bolometers [6], and ≈83% in the Solotvina experiment utilizing four cadmium tungstate crystal scintillators with the total mass of ≈0.3 kg [7].

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$CaMoO_4$ contains $2\nu2\beta$ active isotope ^{48}Ca (present in natural Ca with abundance of $\delta = 0.187\%$ [4]) which creates a background at the $Q_{2\beta}$ energy of ^{100}Mo [12]; $CdMoO_4$ contains β active ^{113}Cd ($T_{1/2} = 8.04 \times 10^{15}$ yr [21], $\delta = 12.22\%$ [4]); $PbMoO_4$ has a high effective Z value which causes stronger interactions with background γ quanta, and contains radioactive ^{210}Pb; $Li_2Zn_2(MoO_4)_3$ has a low light yield; and recently developed $ZnMoO_4$ has some coloration, which leads to substantial self-absorption of scintillation light. An important advantage of Li_2MoO_4 is the absence of heavy elements and a high concentration of Mo (55% in weight).

The main purpose of our work was to investigate the applicability of Li_2MoO_4 crystal as a possible cryogenic scintillator to search for the double beta decay of molybdenum. We have also measured the luminescence of the material under X-ray excitation. The possibility to search for solar axions by using Li_2MoO_4 crystals will also be briefly discussed. Some preliminary results of this work were reported in Ref. [22].

2. Li_2MoO_4 crystals

The lithium molybdate (Li_2MoO_4) was obtained by a solid-state synthesis technique from MoO_3 and Li_2CO_3 powders (both of 99.5% purity) with subsequent recrystallization from water solutions. Transparent single crystals of up to $\varnothing 25 \times 35$ mm were grown by the Czochralski technique with a drawing speed of 4 mm/h. The room temperature X-ray powder diffraction (XRD) pattern was indexed according to the lattice parameters of the trigonal system (space group $R\bar{3}$). The structure is isotypic with the phenacite Be_2SiO_4. The density calculated from the XRD pattern (3.07 g/cm^3) is slightly different from the hydrostatic density (3.02 g/cm^3) what could be explained by the presence of some defects in the crystal lattice. The material is soluble in water and weakly hygroscopic; the melting point is at $(701 \pm 2)^\circ$C [23].

3. Measurements, results and discussion

3.1. Luminescence under X-ray excitation

The luminescence characteristics of crystal Li_2MoO_4 were investigated at temperatures of 10 and 300 K under synchrotron radiation excitation [24]. While no luminescence was detected at 300 K, it was observed at 10 K with a maximum of emission spectrum at 580 nm.

We studied the luminescence and thermally stimulated luminescence (TSL) of a small sample of Li_2MoO_4 crystal ($\varnothing 25 \times 0.9$ mm) under irradiation from a BHV7 X-ray tube with a rhenium anode (20 kV, 20 mA), at room (295 K) and liquid nitrogen (85 K) temperatures. Light from the crystal was detected in the visible region by a FEU-106 photomultiplier (sensitive in the wide wavelength region of 300–800 nm). Spectral measurements were carried out using a high-aperture MDP-2 monochromator.

Only one band in visible region, with the maximum at 538 nm, was observed at room temperature (Fig. 1). The radioluminescence (RL) spectrum, measured at 85 K, consists of two bands with maxima at 543 and 458 nm. In addition, the spectrum measured at 85 K is also distributed in the wavelength region above 650 nm.

The temperature dependences of RL in a wide interval of wavelength (integral) and in the narrow region near the emission maximum (535 nm) are presented in Fig. 2. The RL intensity slowly falls, but it still remains observable at 450 K temperature. We assume that the observation of luminescence even above room temperature (in contrast to Ref. [24]) can be explained by higher sensitivity of the set-up used in the present study.

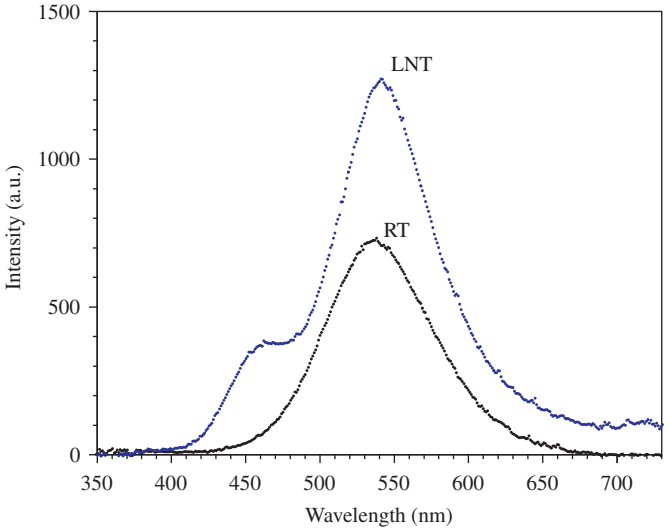

Fig. 1. (Color online) Emission spectrum of Li_2MoO_4 crystal under X-ray excitation at room (295 K, RT) and liquid nitrogen (85 K, LNT) temperatures.

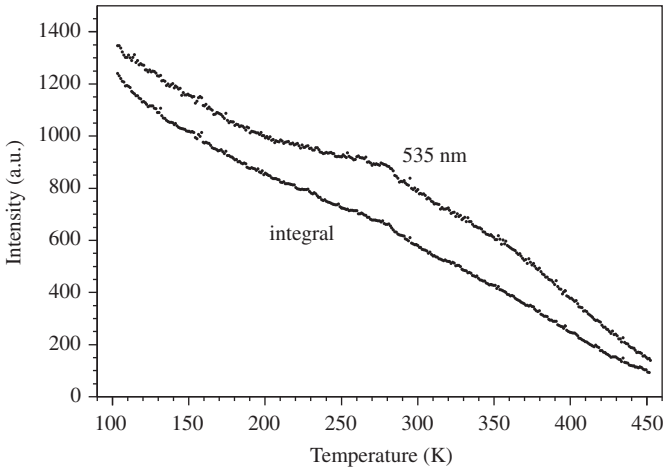

Fig. 2. Temperature dependence of luminescence intensity under X-ray excitation for the wavelengths near the emission maximum (535 nm) and in integral spectrum.

TSL was observed over a wide temperature interval (see Fig. 3), however, the majority of light is released in the range 120–270 K, with a maximum at 170 K. The TSL observed in our measurements indicates the presence of traps in the Li_2MoO_4 sample. We assume it is due to defects and impurities in the crystal, which was grown from raw materials of rather low quality. An additional purification of initial materials is necessary to improve the quality of Li_2MoO_4 crystals.

3.2. Test at milli-Kelvin temperature

The sample of Li_2MoO_4 crystal was measured over 36.9 h as a scintillating bolometer in the CUORE R&D experimental set-up in the Gran Sasso National Laboratory. In the set-up (described in

detail in Ref. [8]) the sample was glued on a copper support. Heat signals from the crystal were measured with a neutron transmutation doped (NTD) germanium thermistor thermally coupled to the sample. In addition to the heat signal, the scintillation light was detected simultaneously by a germanium crystal $\varnothing 66 \times 1$ mm (facing the Li_2MoO_4 sample) with another NTD Ge thermistor glued on its surface. The germanium crystal was coated by thin layer of silicone oxide to improve the absorption of scintillation light. The set-up was cooled to ≈ 10 mK. The front-end electronics are described in Refs. [25,26].

The light output of the sample was estimated as 7% relatively to a $CdMoO_4$ crystal $10 \times 10 \times 2$ mm. Taking into account the relative light outputs of $CdMoO_4$ and $CaMoO_4$ at 9 K [13], this corresponds to $\approx 20\%$ relative to $CaMoO_4$. Light output for α particles is $\approx 30\%$ relative to scintillation signals from γ quanta. It allows us to discriminate α and $\gamma(\beta)$ events (see Fig. 4).

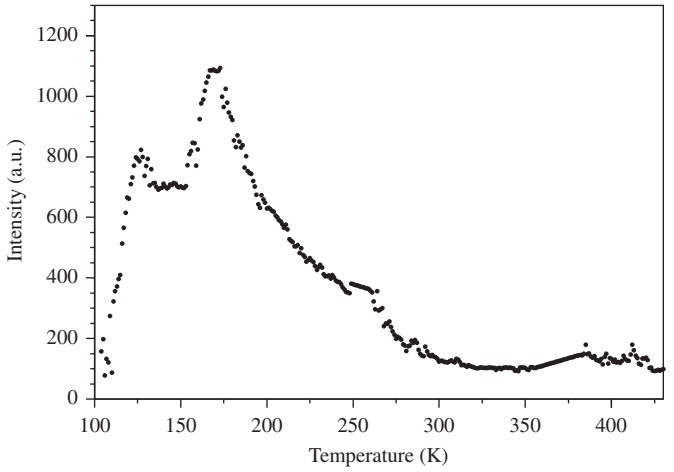

Fig. 3. Thermoluminescence of Li_2MoO_4 crystal after X-ray excitation at liquid nitrogen temperature.

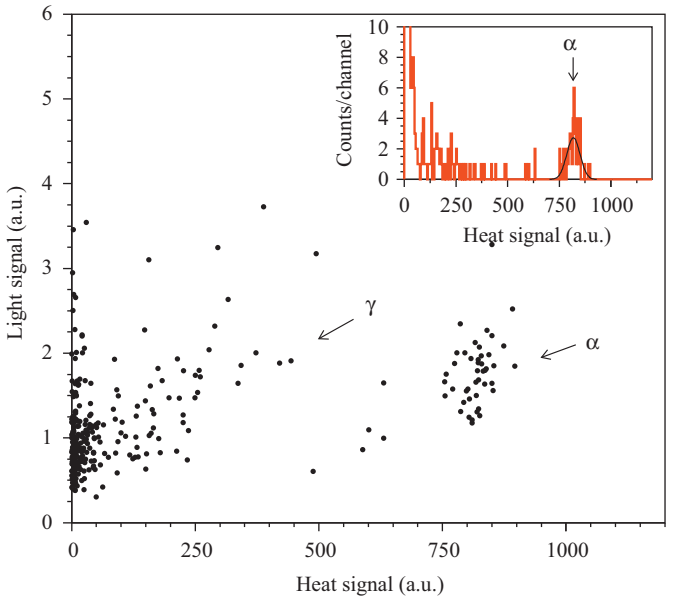

Fig. 4. (Color online) Scatter plot of the light signal versus heat signal for 36.9 h background exposition with Li_2MoO_4 crystal $\varnothing 25 \times 0.9$ mm. (Inset) Distribution of heat signals. Fit of α peak by Gaussian function with the energy resolution FWHM = 9.3% is shown by solid line.

The peak observed in the heat signal spectrum is probably due to contamination of the crystal by ^{210}Pb (or ^{210}Po if the equilibrium is broken). However, it could also be due to contamination of the set-up, which was not particularly clean. Therefore we can only give a limit on the activity of ^{210}Po in the sample. Assuming that all the peak area (44 counts) is due to internal contamination of the sample by ^{210}Po, and taking into account the time of measurement (36.9 h) and mass of the sample (1.3 g), we estimate internal activity of ^{210}Po in Li_2MoO_4 as ≤ 0.3 Bq/kg. It should be noted that, because this peak is much wider than usual for bolometers, it also could be sum of a few peaks (probably with long tails at lower energies due to surface contamination).

4. Possible applications in searches for 2β processes in Mo and in searches for solar axions

Lithium molybdate contains three potentially 2β decaying isotopes of molybdenum. They are listed in Table 1.

It should be stressed that 2β processes in ^{92}Mo and ^{98}Mo have been investigated to a much lesser extent than those for ^{100}Mo, with limits on the level of $10^{18}-10^{20}$ yr for 2ε and $\varepsilon\beta^+$ decays of ^{92}Mo (see summary [29] for exact values for different modes and further references) and a very poor limit of 10^{14} yr for ^{98}Mo derived in Ref. [29] from the old data of Ref. [30]. These values could be improved in a possible experiment with low background Li_2MoO_4 cryogenic scintillator in the "source = detector" approach.

Radioactive contamination plays an important role for high sensitivity 2β experiments. Radiopurity of a Li_2MoO_4 crystal with mass of 34 g was measured deep underground at the Gran Sasso National Laboratory (Italy) with an ultra-low background HP Ge γ spectrometer. These measurements have shown a comparatively low level of radioactive contamination of the material. The following limits were set as <20 mBq/kg for ^{226}Ra, <30 mBq/kg for ^{228}Th, <4 mBq/kg for ^{137}Cs and <8 mBq/kg for ^{60}Co. Only ^{40}K was detected in the sample with the activity 170 ± 80 mBq/kg [31].

We intend to test this Li_2MoO_4 crystal in the CUORE R&D set-up. As a next step we are going to grow a Li_2MoO_4 crystal from raw compounds selected for low chemical impurities and radioactive contamination.

It should be mentioned that the crystal contains also ^{7}Li with a high natural abundance of 92.41% [4]. Therefore the material can be used as a target to search for hypothetical axions emitted in the solar core in de-excitation of the 478 keV level, which is being populated in one of branches of the pp chain (see f.e. recent paper [32] for details). In Ref. [32], 478 keV γ quanta were searched for with an external HP Ge detector; thus efficiency was quite low (near 4%). Evidently, the experimental sensitivity could be

Table 1
2β unstable nuclides present in Li_2MoO_4 crystals.

Transition	$Q_{2\beta}$ (keV)		δ (%)		Decay channel
^{92}Mo → ^{92}Zr	1649(4)	[3]	14.84(35)	[4]	$2\varepsilon, \varepsilon\beta^+$
			14.5246(15)	[27]	
^{98}Mo → ^{98}Ru	113(6)	[3]	24.13(31)	[4]	$2\beta^-$
			24.391(18)	[27]	
^{100}Mo → ^{100}Ru	3035(6)	[3]	9.63(23)	[4]	$2\beta^-$
	3034.40(17)	[28]	9.824(50)	[27]	

For isotopic abundance (δ) the recommended values [4] and the most precise recently measured values [27] are given.

improved using the "source=detector" approach if a Li_2MoO_4 crystal will be used as a cryogenic bolometer.

5. Conclusions

Luminescence in a Li_2MoO_4 single crystal was studied under X-ray excitation in the temperature interval 85–450 K. Weak luminescence was observed at room temperature in the visible region $\lambda \approx 500-580$ nm with a maximum at 538 nm. The second band with a maximum at 458 nm appears at 85 K, while the maximum of the main band shifts to 543 nm. The intensity of luminescence increases as the temperature falls and is very low at $T > 450$ K. Thermally stimulated luminescence was observed in a wide temperature interval with the main part of light released at $T = 120-270$ K with a maximum at 170 K.

Applicability of Li_2MoO_4 crystals as cryogenic phonon-scintillation detectors was demonstrated for the first time. Light output of Li_2MoO_4 was estimated as 20% compared to $CaMoO_4$ with quenching factor ≈ 3 for alpha particles. A limit on the activity of ^{210}Po at the level of ≤ 0.3 Bq/kg was set from the data of the cryogenic test. Limits on activities of other radioactive impurities were obtained in previous measurements with a HP Ge detector [31].

This crystal scintillator could be used as a possible detector to search for some rare nuclear processes: double beta decay of Mo isotopes, and resonant capture of hypothetical solar axions on ^{7}Li.

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