Tungstate and Molybdate Scintillators to Search for Dark Matter and Double Beta Decay

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***Abstract*—Results are presented on our latest research, aimed at the development and study of oxide scintillation crystals ($ZnWO_4$, $ZnMoO_4$, $PbWO_4$, $PbMoO_4$, and $MgWO_4$) with high scintillation yield and low intrinsic radioactivity. We report on the improvement of these properties for conventional scintillators, as well as on new promising crystals based on metal tungstates and molybdates. The results are discussed in view of applying these materials in cryogenic experiments searching for dark matter and/or neutrinoless double beta decay.**

***Index Terms*—Dark matter, double beta decay, low-temperature scintillators, oxide crystals, scintillation detectors.**

I. Introduction

OXIDE crystal scintillators play an important role in fundamental and applied research. They are widely used for detection of ionizing radiation in high-energy physics, space research, security applications, medical diagnostics, etc. Oxide crystal scintillators exhibit unique properties, such as high scintillation efficiency, thermal and radiation stability, and nonhygroscopicity. Further progress in many areas of modern science and technology relies on the development of a new generation of more efficient and sensitive scintillation detectors to address a variety of fundamental and applied objectives. Searches for rare events, such as double beta decay, dark-matter particle interactions, rare α- and β-decay processes, and hypothetical nuclear fissions are current important topics on the agenda of astroparticle physics [1]. The most important requirements for scintillators in these experiments are low intrinsic radioactivity and high scintillation light output. Studies of rare events using oxide scintillators containing isotopes of cadmium, tungsten, zinc, molybdenum, etc. are promising [2]–[4]. The success of these experiments depends upon many factors, the most important of which is the availability of a suitable detector that satisfies a number of requirements: maximum content of the nuclei of interest in the detector volume; high detection efficiency and selectivity; extremely low background level; and the possibility of prolonged measurements.

It is difficult with traditional scintillation techniques to collect enough light to obtain a low-energy threshold (a few thousand electron volts) needed for dark-matter searches, and energy resolution (as better as possible) required by future high-sensitivity double beta decay experiments. Therefore, the next generation of experiments will use also cryogenic detectors, capable of measuring very low energy deposition with very good energy resolution [5]. One of the latest advances in cryogenic detectors is the development of a technique for simultaneous measurement of the light and phonon response induced by a particle interaction in the scintillation target [6]. This allows efficient event type discrimination, offering a powerful tool for radioactive background identification. This proven technique has demonstrated its advantages in low-background physics experiments searching for rare events [7]–[9]. The successful operation of cryogenic phonon-scintillation detectors in these experiments has increased the research activity into materials that can be used as cryogenic scintillation targets (see [10] and references therein).

Among oxide scintillators, of great interest are crystals based on metal tungstates and molybdates. The possibility of producing scintillators of sufficiently large size, high resolution, and low intrinsic radioactivity underpins the prospect of these compounds in future experiments searching for rare events. Motivated by this, work has been started at the Institute for Scintillation Materials (Kharkov, Ukraine), aiming to develop and optimize scintillating oxide crystals to search for dark matter and double beta decay.

This paper reports our latest achievements in growing large-volume $ZnWO_4$ crystals with improved characteristics as well as the development of new scintillating crystals, i.e., $ZnMoO_4$ and $MgWO_4$. We shall discuss the performance characteristics of zinc tungstate scintillators and also first results on the characterization of zinc molybdate and magnesium tungstate crystals. We also included in this study relevant results on the tungstate

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Fig. 1. $ZnMoO_4$ single crystals.

Fig. 2. $MgWO_4$ single crystal.

and molybdate of lead; crystals that have been around for a long time and possibly are interesting targets for cryogenic rare-event searches.

II. Experimental

$ZnWO_4$, $ZnMoO_4$, $PbWO_4$, and $PbMoO_4$ crystals were grown in platinum crucibles with high-frequency heating, using the Czochralski method. The raw charges of the required composition were prepared by solid-phase high-temperature synthesis. $ZnWO_4$, $PbWO_4$, and $PbMoO_4$ single crystals with dimension up to $\emptyset$ 50 mm $\times$ 100 mm were grown and scintillation elements of various sizes and shapes (cylinders, rectangular and hexahedron prisms) were produced. Technology for the production of new materials ($ZnMoO_4$ and $MgWO_4$), envisaged for application in rare-event searches has been developed. $ZnMoO_4$ crystals of diameter 20–30 mm and 30–40 mm height of cylindrical shape (see Fig. 1) were produced using the Czochralski technique. $MgWO_4$ crystals of ~1 cm^3 (see Fig. 2) were grown by pulling the seed from the melted flux solution.

The relative light output and the energy resolution were determined by means of γ-spectroscopy, using ^{241}Am (59.5 keV) and ^{137}Cs (662 keV) sources. The relative scintillation intensity of the crystals was studied over the temperature interval 7–300 K. A sample of the crystal with volume $5 \times 5 \times 1$ mm^3 was placed into an optical cryostat and excited with ^{241}Am alpha particles. Measurements were carried out using the multiple photon counting technique [11].

Measurements of the afterglow were carried out using a dedicated setup for studies of kinetics characteristics of scintillators. Samples were irradiated by X-ray pulses of duration 2 s, emitted by a RAPAN-200 X-ray source ($V = 120$ kV, $I = 3$ mA).

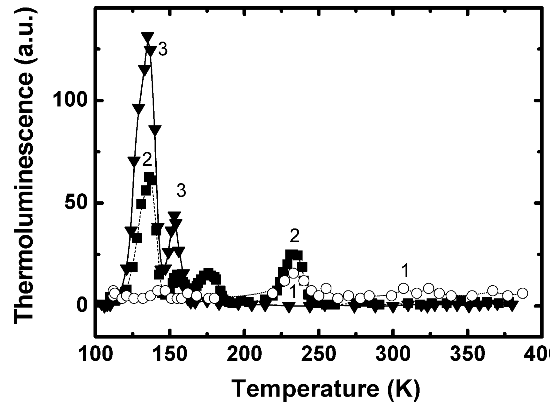

Fig. 3. Thermally stimulated luminescence of $ZnWO_4$ crystals: (1)—undoped, (2)—doped with fluorides, (3)—doped with ZnF_2 and Me_2O, where Me is a univalent metal.

Measurements of X-ray luminescence were carried out in helium or liquid nitrogen cryostats designed for these studies. An X-ray tube with Mo-anticathode, operating at 55 kV and 10 mA, was used as an excitation source. The luminescence spectra were recorded through an MDR-12 monochromator with a green-sensitive photomultiplier FEU-79, operating in the single-photon counting regime. The data presented here are not corrected for the spectral response of the detection system.

To measure thermally stimulated luminescence (TSL) of the crystal, a sample was first irradiated at the base temperature of the cryostat and then the integrated light signal was monitored while heating the crystal at a constant rate of 0.03 K/s.

The level of radioactive contamination of a $ZnWO_4$ crystal scintillator was evaluated using the low-background setup at the Solotvina Underground Laboratory described elsewhere [12].

III. Results and Discussion

A. $ZnWO_4$

Zinc tungstate scintillator is a very attractive material for the detection of rare decay processes [13]. Recently, it has also been suggested as a promising target for a cryogenic dark-matter search experiment [14]. This motivated us to initiate extensive research on optimization of the growth conditions, aiming to produce high-quality large-volume scintillating crystals of $ZnWO_4$ [12], [15]. Alongside optimization of the crystal production process, studies were carried out on the effects of stoichiometry and doping. It is found that co-doping with some univalent impurities and fluorides improves the scintillation properties of $ZnWO_4$ and significantly reduces the afterglow.

The results from thermally stimulated luminescence, displayed in Fig. 3, demonstrate that there is correlation between the afterglow and the magnitude of the peaks at $T > 233$ K. It is assumed that the traps associated with these TSL peaks are responsible for the accumulation of charge carriers at room temperature, and this accounts for the observed slow decay process. Doping creates shallow trapping centers with activation energy so low that storing carriers at room temperature is not possible. This causes a noticeable improvement in the afterglow characteristics of the crystals: it reduces drastically from 0.79% in the undoped crystal to 0.005% in the co-doped

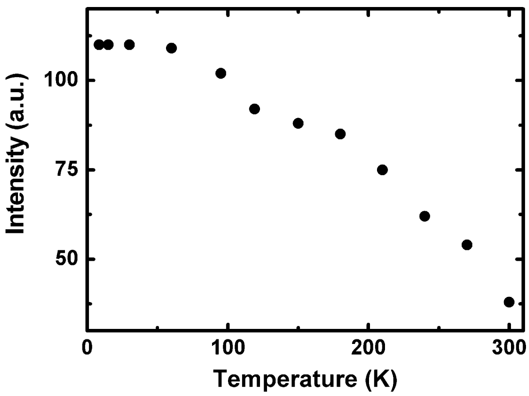

Fig. 4. Light output of $ZnWO_4$ crystal scintillator as function of temperature. Excitation with ^{241}Am α particles.

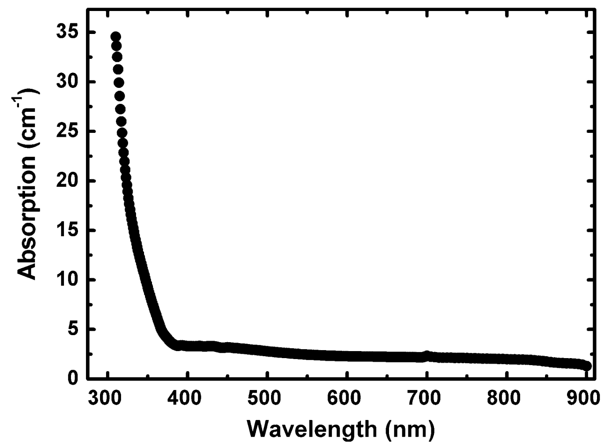

Fig. 5. Absorption spectrum of $ZnMoO_4$ crystal at room temperature.

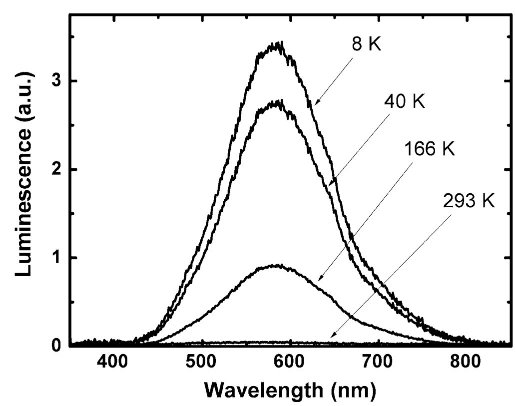

Fig. 6. X-ray luminescence of $ZnMoO_4$ crystal at different temperatures.

with some univalent impurities and fluorides sample. The results of these studies allowed us to optimize the process of crystal growth. Large-volume $ZnWO_4$ crystals with improved scintillation properties [15] were produced this way.

The energy resolution of $ZnWO_4$ scintillation elements is 8.5% for a 1 cm^3 sample and 10.7% for a large scintillation element (hexagonal prism with main diameter 40 mm and height of 40 mm) at excitation with the 662 keV γ quanta of ^{137}Cs [12]. The light output of the $ZnWO_4$ samples is 50% and 27% respectively, relative to that of a $CdWO_4$ sample with dimensions $10 \times 10 \times 10$ mm^3 and light yield $\sim$30 000 photons/MeV. The afterglow of the best samples, measured 20 ms after termination of irradiation, is 0.002%.

Given the strong interest in the application of the $ZnWO_4$ as a cryogenic scintillation detector, we studied the light output and decay time constants of the crystal as a function of temperature in the temperature interval 7–300 K. It is shown that the light output of the crystal increases with cooling by $\sim$ 60% (Fig. 4). The relative light output of $ZnWO_4$ at 10 K is ca. 110–115% that of $CaWO_4$. This is consistent with earlier estimates [16].

Intrinsic radioactivity of the crystal is an issue of particular concern for rare-event search experiments and this characteristic of the $ZnWO_4$ scintillator was the subject of a detailed investigation. Measurements carried out at the low-background facility at the Gran Sasso Underground Laboratory demonstrate that zinc tungstate is currently one of the purest scintillation materials in terms of intrinsic radioactivity. The total internal α activity of the best sample studied is less than 0.2 mBq/kg [17].

B. $ZnMoO_4$

$ZnMoO_4$ was identified a couple of years ago as a suitable scintillation material for cryogenic rare-event searches [18]. Recently, for the first time, comparatively large $ZnMoO_4$ single crystals were grown [19]. Possible applications of this crystal include studies of double beta decay of ^{100}Mo and a search for dark matter using a multitarget detector comprising of complementary AMO_4 scintillators (A=Ca, Zn; M=Mo, W). This motivated our development of the technological process for preparation of the crystal, and we produced several ingots of diameter 20–30 mm and height 30–40 mm. As is seen in Fig. 1, the crystal exhibits strong yellow coloration; its absorption spectrum has an abrupt rise at $\sim$ 380 nm and a band at 460 nm (see Fig. 5). This might be an indication of a high level of impurities in line with what is known of the nature of coloration of $ZnWO_4$ crystals [20]. The material needs further R&D to be used of enough large ($\approx$50–100 cm^3) volume. As a next step we intend to grow $ZnMoO_4$ crystal scintillators using high-purity grade (5N) raw materials.

Fig. 6 shows the X-ray luminescence spectra of the $ZnMoO_4$ crystal measured at several temperatures. The emission of the crystal at room temperature is very faint. As the temperature decreases, the luminescence intensity progressively increases, and at $T = 8$ K the spectrum exhibits a broad emission band with a maximum at 585 nm. The luminescence of $ZnMoO_4$ is attributed to the electronic transition of the charge transfer type within the oxyanion complex $[MoO_4]^{2-}$ [18].

Thermally stimulated luminescence of the crystals, measured over the temperature range 6–300 K is displayed in Fig. 7. The glow curves show two intense peaks at 60 and 140 K and a weak structure in the 150–250 K region. The main TSL peaks are associated with the relatively shallow traps that can not give rise to the slow decay processes at room temperature. This is consistent with the low-level of afterglow (0.02%), that has been detected for the $ZnMoO_4$ crystal.

C. $MgWO_4$

Cryogenic scintillation experiments to search for dark-matter particles require several crystal scintillators constituting of dif-

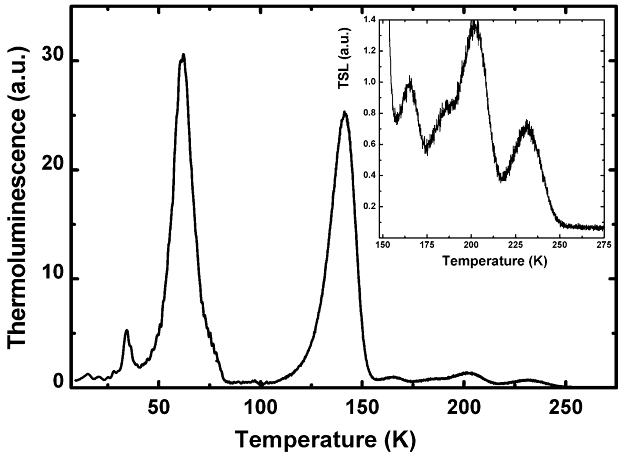

Fig. 7. Thermally stimulated luminescence (TSL) of $ZnMoO_4$ crystal. (Inset) TSL in the temperature interval 160–275 K.

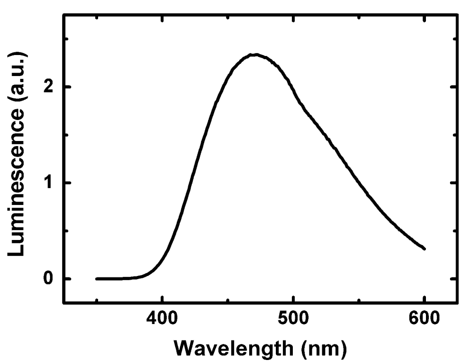

Fig. 8. X-ray luminescence of $MgWO_4$ crystal at room temperature.

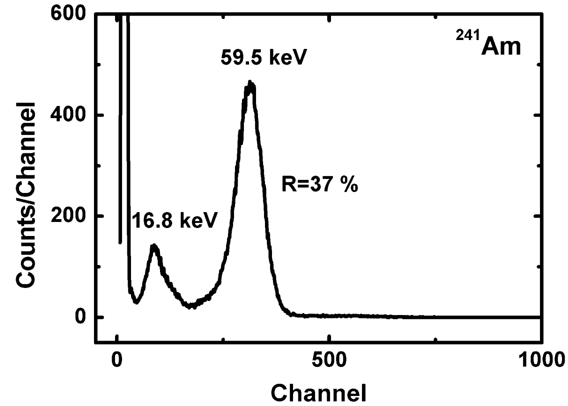

Fig. 9. Pulse amplitude spectra of $MgWO_4$ scintillation element irradiated with 59.5 keV (^{241}Am).

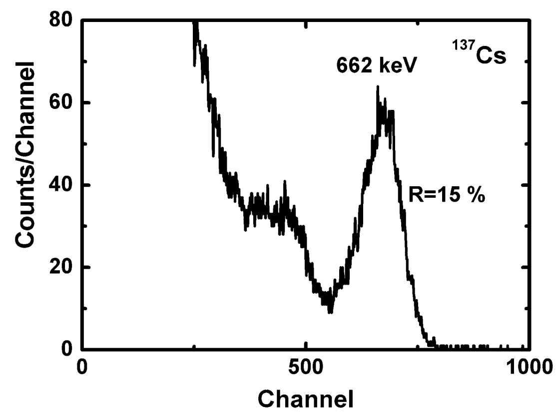

Fig. 10. Pulse amplitude spectra of $MgWO_4$ scintillation element irradiated with 662 keV γ-quanta (^{137}Cs).

ferent elements for confirming a true dark-matter signal. Recent studies of powder $MgWO_4$ samples demonstrated that this compound is an attractive scintillation material for cryogenic applications because of its high scintillation light output which is comparable to $ZnWO_4$ [21]. Due to high-temperature phase transition, $MgWO_4$ can not be grown using the conventional Czochralski method. Therefore, we developed the technology for flux growth and produced a single crystalline sample of $MgWO_4$ (see Fig. 2). The material is not brittle and not hygroscopic. The crystal shows luminescence under X-ray excitation at room temperature (see Fig. 8). The broad emission band exhibits a maximum at 475 nm. This agrees well with the room temperature data obtained for a powder sample [21]. For the first time, the single crystalline sample of $MgWO_4$ was characterized as a scintillation detector. Fig. 9 shows the pulse amplitude spectrum of $MgWO_4$ excited by γ quanta and X ray of ^{241}Am. The energy resolution of the sample for the γ line 59.5 keV is found to be 37%, very close to the value of 35% observed for a large $ZnWO_4$ scintillator [14]. The energy spectrum for 662 keV γ-quanta of ^{137}Cs is shown in Fig. 10. The energy resolution 15% is obtained for the 662 keV γ-line of ^{137}Cs. The results of comprehensive characterization of this material are discussed in the forthcoming publication [22]. Altogether these results confirm the good prospect of magnesium tungstate for scintillation applications. Therefore, furthering the production technology of large $MgWO_4$ scintillators is currently on its way.

D. *$PbWO_4$ and $PbMoO_4$*

Crystal $PbWO_4$ had been earlier developed in the Institute for Scintillation Materials as a scintillation detector for high-energy physics [23]. Thanks to these studies we have advanced the production technology of this material and currently we can grow large, high-quality crystals. Furthermore, there are ample possibilities for the modification of the material properties that makes this crystal an excellent choice for testing and implementing new ideas on material development. To a lesser extent, this is also applicable to $PbMoO_4$, which has been around for decades as optoelectronic material.

The feasibility of using lead tungstate and molybdate in rare-event searches has been first highlighted in [24] and [25]. It has been shown that $PbMoO_4$ is an interesting material to search for the double beta decay of ^{100}Mo [24], [26], while $PbWO_4$ can be used in a high-sensitivity experiment to search for double beta decay of ^{116}Cd as a shield (anticoincidence detector and/or active light-guide) to suppress a radioactive background [25]. Furthermore radiopure $PbWO_4$ and $PbMoO_4$ crystal scintillators potentially can be used as target materials for cryogenic dark-matter experiments. Nonetheless, the high intrinsic radioactivity ($\sim$ 100 Bq/kg) due to ^{210}Pb is the main obstacle, limiting the usefulness of these materials in low-background experiments. To address this issue we have started work on the purification of archaeological lead for growing lead tungstate and molybdate

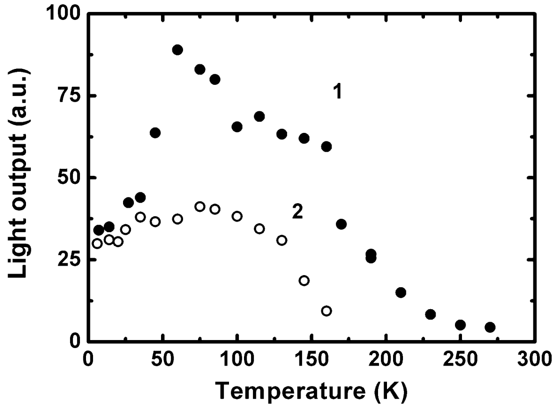

Fig. 11. Light output of $PbWO_4$ (1) and $PbMoO_4$ (2) crystals scintillator as function of temperature measured with α-particles of ^{241}Am source.

crystals with low radioactive background. It has been shown that due to natural decay of radioactive ^{210}Pb in archeological lead, its contribution to the intrinsic radioactivity of a sample can be as low as a few mBq/kg [27]. Using ancient lead (discovered in Ukraine [28]) for crystal growth should permit the production of lead tungstate and molybdate with substantially reduced intrinsic radioactivity. This can pave the way for their application in rare-event search experiments.

In order to assess the feasibility of $PbWO_4$ and $PbMoO_4$ as cryogenic scintillators we measured the temperature dependence of the light output of the crystals down to 7 K (Fig. 11). Due to thermal quenching, both crystals are very poor scintillators at room temperature; only $PbWO_4$ exhibits measurable light output at $T = 295$ K [29] while the light response of $PbMoO_4$ can be detected only at $T < 170$ K. The light output increases with decreasing temperature and reaches a maximum value at 60 K in $PbWO_4$ and 70 K in $PbMoO_4$. At these temperatures the general trend reverses, leading to a decrease of the light yield with further cooling. It should be noted that the scintillation decay time constant of both crystals increases with cooling. At temperatures below 50 K the character of the decay kinetics in both crystals changes: a recombination component with very long (~ 1 ms) time constant appears in the scintillation pulse in addition to the fast exponential components. This long-lasting component points to charge trapping processes occurring in the crystals at these temperatures [30] that leads to a decrease in the number of carriers contributing to the scintillation event. Below 10 K the light output of $PbWO_4$ and $PbMoO_4$ changes insignificantly, suggesting that these scintillators are suitable for cryogenic application.

IV. Conclusion

In this paper we reviewed the results of our efforts directed at the development of oxide scintillators for rare-event search experiments. We already succeeded in producing a large volume $ZnWO_4$ scintillator with improved performance characteristics. Good scintillation properties at low temperatures and exceptionally low intrinsic radioactivity make zinc tungstate an excellent material for cryogenic double beta decay and dark-matter experiments.

Single crystal samples of $ZnMoO_4$ were produced using the Czochralski technique. We investigated the feasibility of this material for cryogenic rare-event search experiments and identified ways to improve the scintillation properties of the crystal.

We investigated methods of producing $MgWO_4$ crystals and demonstrated the feasibility of the flux growth technique. For the first time the single crystals $MgWO_4$ were produced and their luminescence and scintillation characteristics were measured.

Given the good prospects for reducing the intrinsic radioactivity of lead-based crystals we studied the temperature dependence of scintillation characteristics of $PbWO_4$ and $PbMoO_4$. We have shown that these crystals can also be used as cryogenic scintillators. We are planning to produce lead tungstate and molybdate with substantially lower intrinsic radioactivity using ancient lead.

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