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$ZnWO_4$ scintillators for cryogenic dark matter experiments

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ABSTRACT

The scintillation properties of a zinc tungstate crystal, shaped as a hexagonal prism (height 40 mm, diagonal 40 mm) were determined. An energy resolution of 10.7% for the 662 keV γ-line of ^{137}Cs was measured with the scintillator placed in a light collection setup similar to that used by the CRESST dark matter search. The light output and decay kinetics of $ZnWO_4$ were examined over the temperature range 7–300 K and confirmed to be competitive with those of $CaWO_4$. The radioactive contaminations of the $ZnWO_4$ scintillator measured in the Solotvina Underground Laboratory do not exceed 0.1–10 mBq/kg (depending on radionuclide). Our study highlights the excellent feasibility of this $ZnWO_4$ scintillator for a cryogenic dark matter experiment.

1. Introduction

During the past decade there has been a continuous increase of research activity into new scintillation materials for use in crystalline cryogenic phonon-scintillation detectors (see e.g. Ref. [1] and references therein). These detectors combine excellent energy resolution and low threshold with the ability to discriminate between different types of interaction (electron, alpha or neutron interaction). This makes them very attractive tools in experimental searches for dark matter and neutrinoless double beta decay [2–4]. Different scintillation targets are needed to address different experimental objectives, and therefore the characterization and optimization of new scintillation materials, suitable for low-temperature application, is important; especially for EURECA,[1] where a multi-element target is planned for confirming a true dark matter signal.

Zinc tungstate ($ZnWO_4$) is in this regard a very promising scintillator material. It has been proposed for application in searches for double beta decay; dark matter; and for the study of rare alpha and beta decays [5–10]. There is a long history of research work on the scintillation properties of $ZnWO_4$, starting in the early 1980s, when it was first characterised as a radiation detector [11–16]. Recently, $ZnWO_4$ has been the subject of extensive research aiming to optimize its scintillation performance [17–19]. These recent efforts have resulted in the production of large-volume $ZnWO_4$ crystals of up to $\varnothing 50\,mm \times 100\,mm$ with improved scintillation properties [20]. These efforts made zinc tungstate scintillators readily available for experimental rare event searches.

The motivation of this work was to evaluate the performance characteristics of a large-volume $ZnWO_4$ crystal scintillator with emphasis on the requirements of the next generation cryogenic dark matter experiment EURECA [21]. EURECA aims to achieve sensitivity to WIMP-nucleon scattering cross sections down to 10^{-10} pb, which is at least two orders of magnitude better than that of present leading experiments. Critical for success will be the use of detectors with highest light yield and lowest intrinsic radioactivity possible, as these properties eventually determine the sensitivity and discrimination ability of the detectors. In this work we have measured the energy resolution, transmittance, radioactive contamination, temperature dependence of scintillation yield, and decay time constants down to a temperature of 7 K.

2. Measurements and results

2.1. Sample

The $ZnWO_4$ crystal, shaped as a hexagonal prism (height 40 mm, diagonal 40 mm), used in the present study was produced at the Institute of Scintillation Materials (Kharkiv, Ukraine). It was grown from a crystal ingot, using the Czochralski method, in a platinum crucible with high frequency heating [20]. The main axis of the hexagonal prism is perpendicular to the cleavage plane (0 1 0) of the crystal. The side surface of the prism was roughened,

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[1] European Underground Rare Event Calorimeter Array; www.eureca.ox.ac.uk

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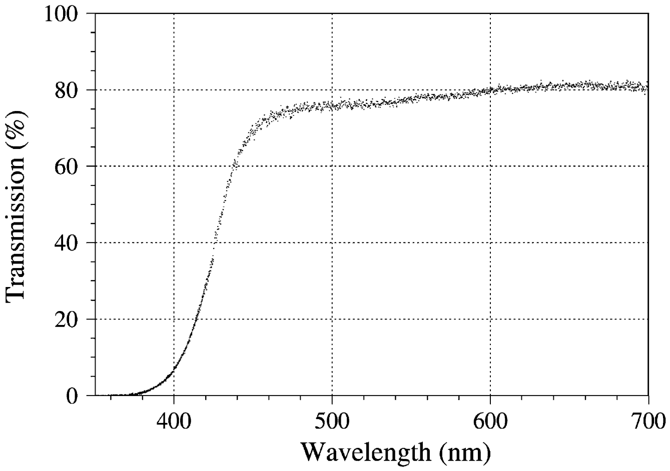

Fig. 1. Optical transmission curve of the $ZnWO_4$ crystal of 40 mm length.

while the top and bottom surfaces were polished. The results of the optical transmission measurements displayed in Fig. 1 demonstrate that the sample exhibits good transmission properties in the relevant wavelength range (>450 nm). This is a significant improvement compared with earlier $ZnWO_4$ samples exhibiting a pink coloration, caused by an absorption band spanning the visible range [11,18].

The improvement of the crystal transparency was achieved by optimization of the crystal production technique and, as we will show in Section 3, this has major implications for the performance characteristics.

2.2. Energy resolution

To measure its scintillation properties, the crystal was coupled to a 3 in. Philips XP2412 photomultiplier (PMT) using Dow Corning Q2-3067 optical grease. The crystal was shrouded by a reflecting cup ∅50 mm × 50 mm made of 3 M reflecting foil that had specular reflectivity 98% in the visible region. The PMT signals were amplified, using a home made spectrometric amplifier with 24 μs shaping time. The crystal was irradiated by γ-quanta from ^{60}Co, ^{137}Cs, ^{207}Bi, and ^{241}Am sources.

Fig. 2 shows the pulse amplitude spectra measured for the hexagonal $ZnWO_4$ scintillator. The energy resolution for the 662 keV γ-line of ^{137}Cs was found to be 10.7%. It is worthwhile noting that this is the first time such a good energy resolution has been obtained for a large volume $ZnWO_4$ (a few tens cm^3) crystal scintillator.

2.3. Non-proportionality of scintillation response

We studied the non-proportionality of the scintillation response of the $ZnWO_4$ scintillator using the same experimental set-up as was used for the measurements described in Section 2.2. Gamma and X-ray quanta from ^{241}Am (59.5 keV), ^{137}Cs (661.7 keV), and ^{133}Ba (30.9, 81.0, 295.3, and 356.0 keV) sources were used in these measurements. The measured light yield per deposited energy is compared with the light yield per energy at 661.7 keV (γ-line of ^{137}Cs). The dependence of the relative light yield of $ZnWO_4$ on the energy of X-ray and γ-quanta is displayed in Fig. 3. The scintillation response of $ZnWO_4$ exhibits a gradual enhancement with the excitation energy and a characteristic dip at the K-edge of tungsten. Dependences similar to this are also observed for $CdWO_4$ [22–24] and $CaWO_4$ [25] scintillators.

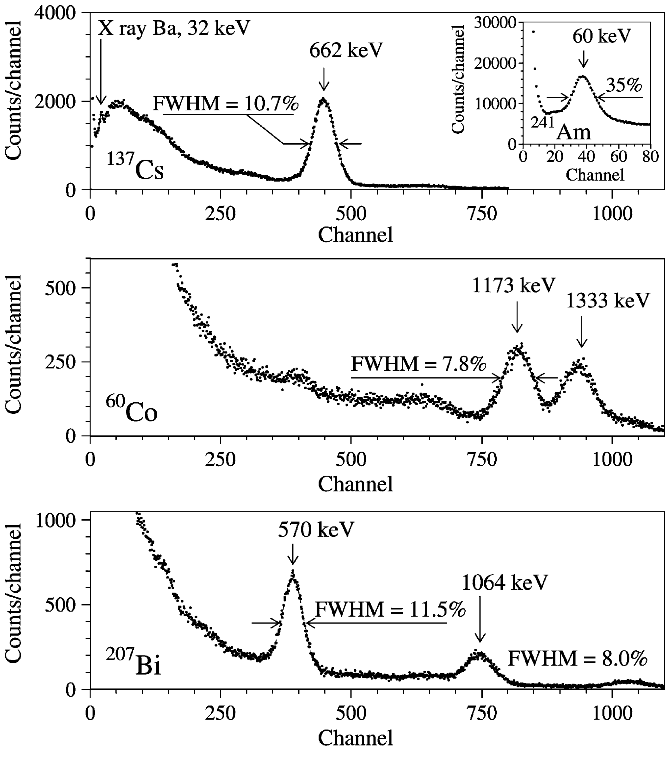

Fig. 2. Energy spectra of ^{137}Cs, ^{241}Am (inset), ^{60}Co, and ^{207}Bi γ-rays measured for the $ZnWO_4$ crystal of hexagonal shape (H=40 mm and D=40 mm).

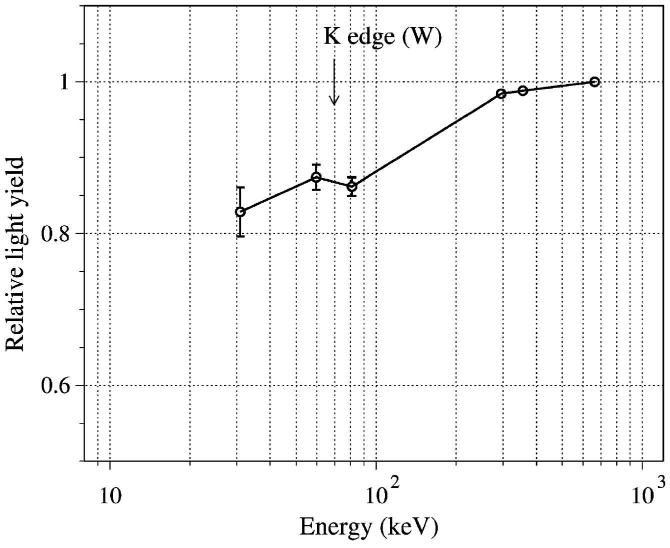

Fig. 3. Non-proportionality of the scintillation response for $ZnWO_4$.

2.4. Low temperature measurements of light output and decay time

The relative scintillation intensity and decay kinetics of $ZnWO_4$ were studied over the temperature range 7–300 K. A 5 × 5 × 1 mm^3 sample of the crystal was placed into an optical cryostat and excited with ^{241}Am α-particles and ^{60}Co γ-quanta. The measurements where carried out using the multiple photon counting technique and a green sensitive photomultiplier (Electron Tubes 9124A); for further details see Ref. [26].

The variation with temperature of the light output of $ZnWO_4$ in the temperature interval 7–300 K is shown in Fig. 4. The relative scintillation efficiency of $ZnWO_4$ prior to spectral correction was found to be 77% that of $CaWO_4$ at T=7 K. The correction factor of

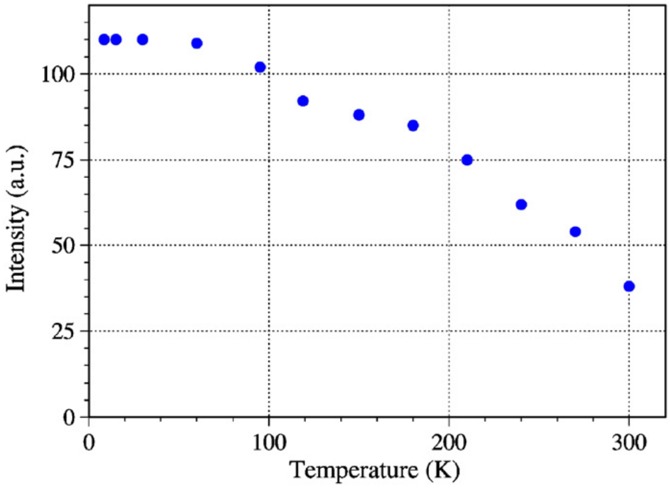

Fig. 4. Temperature dependence of the light output of the $ZnWO_4$ crystal scintillator for excitation with ^{241}Am α-particles.

1.46 ± 0.23 was calculated using the quantum sensitivity curve of the PMT and emission spectra of two crystals at 8 K [7] as explained in Ref. [26]. This results in a relative light yield for $ZnWO_4$ of $115\pm18\%$ that of $CaWO_4$. This is in good agreement with the ratio of the luminescence light output of these two crystals (1.10) measured at 8 K for monochromatic synchrotron X-ray excitation [7].

The temperature dependence of the decay time constants of $ZnWO_4$, measured using a γ-source, is displayed in Fig. 5. The pulse shape of the $ZnWO_4$ scintillation signal can be fitted using a sum of three exponential functions with decay time constants: $\tau_1\approx1\,\mu s$, $\tau_2\approx4\,\mu s$ and $\tau_3\approx25\,\mu s$ (T=295 K), respectively. The decay time constants increase slightly when the temperature is reduced from room temperature down to ~20 K. Below ~20 K, the values for the decay time constants increase significantly upon further cooling. This behaviour is characteristic of the decay process in tungstates [26]. It is controlled by radiative and non-radiative transitions within the emission centre, constituting a metastable level just below the emitting level [27].

2.5. Intrinsic radioactivity

The intrinsic radioactivity of scintillating crystals is a very important factor in experimental rare event searches. We studied the radioactive contamination of a $26\times24\times24\,mm^3$ $ZnWO_4$ crystal produced in the same way as the hexagonal prism scintillator. The measurements were carried out in the Solotvina Underground Laboratory, built in a salt mine 430 m underground (~1000 m w.e.). The scintillator was viewed by a special low-radioactive 5″ PMT (EMI D724KFLB) through a high-purity quartz light guide of 10 cm diameter and 33 cm length. The detector was surrounded by a passive shield made of Teflon (3–5 cm), Plexiglas (6–13 cm), high purity copper (3–6 cm) and lead (15 cm). For each event in the detector the signal amplitude and its arrival time were recorded; the scintillation pulses were digitized with a 20 MS/s sampling frequency.

The energy spectrum of the crystal, accumulated over 44.7 h in the low background set-up (see Fig. 6), shows no features one could assign to specific radioactive isotopes. This allows us to set limits on radioactive contaminations of the crystal by nuclides from the U/Th families as well as ^{40}K, ^{90}Sr–^{90}Y, ^{137}Cs, and ^{147}Sm. The measured spectrum was fitted in the energy interval 120–3990 keV (χ^2/n.d.f.=80/66=1.21) with a fit function composed of the appropriate background components. The components

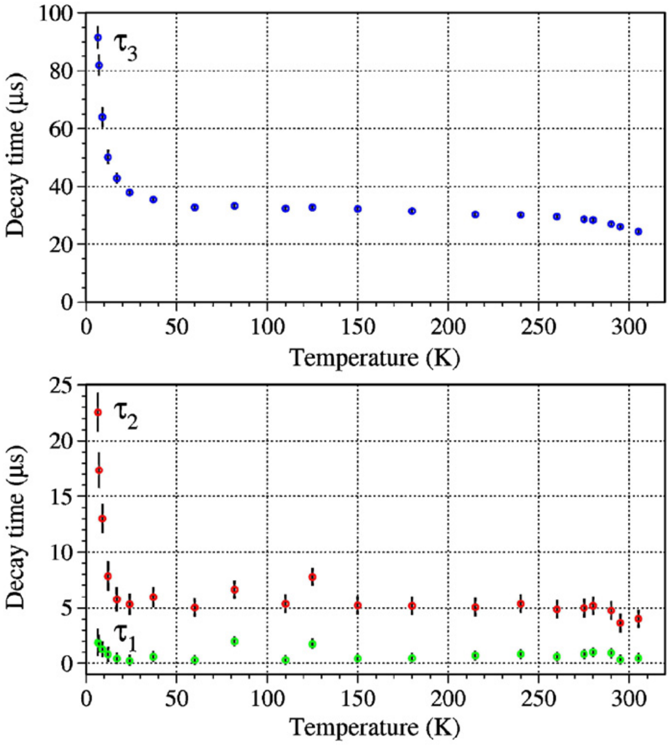

Fig. 5. Temperature dependence of the decay time constants measured for irradiation with ^{60}Co γ-quanta.

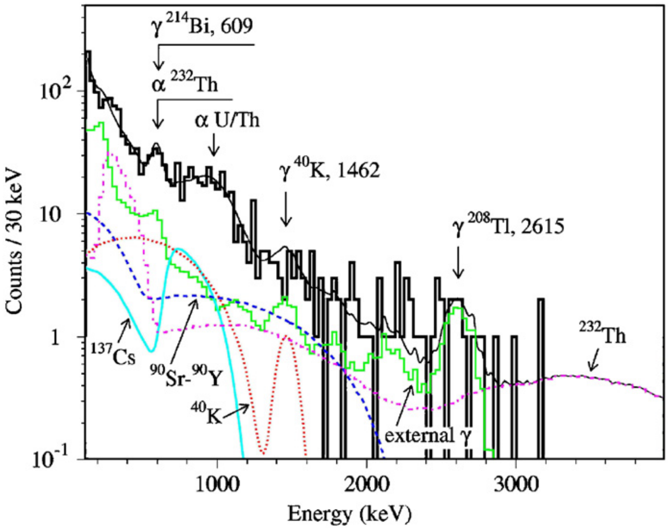

Fig. 6. (Color online) Energy spectrum accumulated in a low-background set-up with the $ZnWO_4$ crystal scintillator $26\times24\times24\,mm^3$ over 44.7 h together with the model of the background. The main components of the background are shown: spectra of ^{40}K, ^{90}Sr–^{90}Y, ^{137}Cs, beta active daughters of ^{232}Th and the contribution from external γ-quanta. Peculiarities in the spectrum at the energies ≈0.6 and ≈1 MeV can be explained by the presence of α active U/Th daughters in the crystal. Energies of γ-lines are in keV.

were simulated using the GEANT4 package [28,29]. Since the equilibrium of the U/Th families in the crystals is expected to be broken, different parts of the U/Th families (^{228}Ac, ^{208}Tl, ^{210}Bi, ^{214}Bi, ^{234m}Pa) were considered separately. Pulse-shape discrimination between γ/β events and α particles [6] was used to estimate the total α activity of U/Th as ≈3 mBq/kg in the energy region 0.5–1.2 MeV. The fast chains, $^{214}Bi\rightarrow{}^{214}Po\rightarrow{}^{210}Pb$ (giving the activity of ^{226}Ra within the ^{238}U family) and $^{220}Rn\rightarrow$

Table 1
Radioactive contamination of $ZnWO_4$ crystal.

Chain	Source	Activity (mBq/kg)
^{232}Th	^{232}Th	$\leqslant 4$
	^{228}Th	$\leqslant 0.1$
^{238}U	^{226}Ra	$\leqslant 0.2$
Total α activity	U/Th	≈ 3
	^{40}K	$\leqslant 14$
	^{90}Sr–^{90}Y	$\leqslant 15$
	^{137}Cs	$\leqslant 3$
	^{147}Sm	$\leqslant 5$

$^{216}Po \rightarrow {}^{212}Pb$ (^{228}Th within ^{232}Th) were identified using time-amplitude analysis [30]. A summary of the measured radioactive contaminations of the $ZnWO_4$ scintillator is given in Table 1.

Recently, the intrinsic radioactivity of $ZnWO_4$ crystal scintillators was measured with higher sensitivity in the experimental search for the 2β decays of zinc and tungsten using $ZnWO_4$ crystal scintillators [8,9]. This experiment was carried out in the DAMA R&D set-up at the Gran Sasso underground laboratory at a depth of ~3600 m w.e. It was found that the $ZnWO_4$ crystal scintillator exhibits exceptionally low radioactive contamination: ^{228}Th~2–5 μBq/kg, ^{226}Ra~2–6 μBq/kg, $^{210}Po \leqslant 0.2$ mBq/kg, total α activity (U/Th) 0.2–0.4 mBq/kg, and $\leqslant 1$ mBq/kg (^{90}Y–^{90}Sr) [9,31]. This moves $ZnWO_4$ crystals much closer to satisfying the stringent radiopurity requirements of the EURECA dark matter search experiment.

3. $ZnWO_4$ and mass production of cryogenic scintillation detectors

The discrimination quality of phonon-scintillation detectors is governed mainly by the energy resolution of the light channel, but also by that of the phonon channel. The energy resolution already achieved for the phonon channels is perfectly adequate for dark matter searches. CRESST, for example, achieves an energy resolution of 1 keV (FWHM) at 47 keV through using transition edge sensors (TES) made of tungsten [32]. Improving the energy resolution of the light channel is the current focus of research, carried out in regard of phonon-scintillation detectors. Increasing the scintillation yield, a physical property of the scintillator, and producing large crystals that exhibit long optical absorption lengths, are important areas being worked on.

Another aspect, especially in light of mass production for EURECA, is the reliability and high production yield of the detector fabrication technology. The technology developed for direct deposition of phonon sensors on $CaWO_4$ [32] is unsuitable for $ZnWO_4$ targets due to the need of heating up the scintillator to several hundred °C during TES film deposition. This heating results in oxygen depletion with detrimental consequences for the scintillation light yield of the crystal [10]. $ZnWO_4$ targets need thermometers that are being glued onto their surface, which can be done without heating the $ZnWO_4$ crystal. In addition to preserving a high light yield of $ZnWO_4$, by not heating up, a further advantage of gluing thermometers is that they can be prefabricated in bulk, and tested, before attaching them to the scintillating target.

Neutron transmutation doped germanium (NTD-Germanium), glued onto the surface of targets, has been established successfully by cryogenic experiments [4,33,34]. Recently, gluing of tungsten TES phonon sensors onto dark matter targets was investigated and shown to give results competitive with tungsten TES sensors vacuum-deposited directly onto the targets' surface [35]. Encouraging results from recent tests of $ZnWO_4$ detectors with glued phonon sensors [36], and also progress in the technology of producing large zinc tungstate crystals of high quality, pave the way for future use of this material in the cryogenic dark matter experiment EURECA.

4. Conclusions

The study of a large $ZnWO_4$ scintillator in the shape of a hexagonal prism (height 40 mm, diagonal 40 mm) showed an energy resolution of 10.7% for 662 keV γ-quanta of ^{137}Cs. It is found that $ZnWO_4$ exhibits the non-proportionality of the scintillation response that is typical for tungstate scintillators. The relative intensity and scintillation decay kinetics were investigated over the temperature range 7–300 K. The light output of $ZnWO_4$ exhibits an about two-fold increase with cooling and exceeds that of $CaWO_4$ by 15% at 7 K.

Measurements of the intrinsic radioactivity of the $ZnWO_4$ crystal in the low-background set-up at the Solotvina Underground Laboratory showed that this $ZnWO_4$ crystal is one of the cleanest inorganic scintillators. Only upper limits at the level of ~0.1 mBq/kg could be set for the contamination by nuclides from the U/Th families, while radioactive contamination by ^{40}K, ^{90}Sr–^{90}Y, ^{137}Cs, and ^{147}Sm was found to be less than a few mBq/kg.

Preliminary calculations indicate that a scintillation element of hexagonal shape is preferable over one of cylindrical shape for maximizing the light collection from a $ZnWO_4$ scintillator. To verify this experimentally, we are preparing both, Monte Carlo simulations and measurements, of two $ZnWO_4$ samples, one of cylindrical and the other of hexagonal shape, but both produced from the same crystalline boule.

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