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# Search for rare processes with $ZnWO_4$ crystal scintillators

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**Abstract.** Radiopure $ZnWO_4$ crystal scintillators with mass (0.1–0.7 kg) have been developed and put in measurement in the Gran Sasso National Laboratories of the INFN to search for rare processes. The radioactive contamination of the crystals have been estimated and the double beta decay of zinc and tungsten isotopes was searched for, reaching a sensitivity at the level of $10^{18}$ – $10^{21}$ yr; in addition a new half-life limit on alpha transition of $^{183}$W to the metastable excited level of $^{179}$Hf has also been obtained. The achieved radiopurity of the $ZnWO_4$ crystals make them very promising detectors for $\beta\beta$ decay investigations while their anisotropic features make them very interesting detectors to investigate dark matter particles directionality.

## 1 $ZnWO_4$ crystal scintillators

In recent years $ZnWO_4$ crystal scintillators were considered in the search for rare processes [1–5]. In particular four clear, slightly colored $ZnWO_4$ crystal scintillators have been produced: i) crystals ZWO-1 (117 g) and ZWO-2 (699 g) were produced in the Institute for Scintillation Materials (ISMA, Kharkiv, Ukraine) from crystal ingots grown in platinum crucibles by the Czochralski method; ii) the crystal ZWO-3 (141 g) was obtained by recrystallization from the sample ZWO-2 at the ISMA; iii) the ZWO-4 (239 g) was produced in the Nikolaev Institute of Inorganic Chemistry (Novosibirsk, Russia) by the low-thermal gradient Czochralski technique also in platinum crucible. The radioactive contamination of these $ZnWO_4$ crystal scintillators has been investigated deep underground in the Gran Sasso laboratory, by using the low background facility DAMA/R&D [6]. In the measurement the considered $ZnWO_4$ crystals were fixed inside a cavity of $\oslash 49 \times 59$ mm in the central part of a polystyrene light-guide 66 mm in diameter and 312 mm in length. The cavity was filled up with high purity silicone oil. The light-guide was optically connected, on the opposite sides, to two low radioactive EMI9265B53/FL 3 inches photomultipliers (PMT). The light-guide was wrapped by PTFE reflection tape. Some upgradings of the detector system have been performed in the different periods of measurements; in particular, in the final stages of the experiment two polished quartz light-guides ($\oslash 66 \times 100$ mm) were installed between the polystyrene light-guide and the PMTs to suppress $\gamma$ ray background from the PMTs. The detector was surrounded by Cu bricks and sealed in a low radioactive air-tight Cu box continuously flushed with high purity nitrogen gas. The Cu box was surrounded by a passive shield made of 10 cm of high purity Cu, 15 cm of low radioactive lead, 1.5 mm of cadmium and 4/10 cm polyethylene/paraffin to reduce the external background. The whole shield has been closed inside a Plexiglas box, also continuously flushed by high purity nitrogen gas. An event-by-event data acquisition system accumulates the amplitude, the arrival time, and the pulse shape of the events.

The energy scale and the energy resolution of the $ZnWO_4$ detectors have been measured with $\gamma$ sources $^{22}$Na, $^{60}$Co, $^{133}$Ba, $^{137}$Cs, $^{228}$Th, and $^{241}$Am. The energy resolution of the detectors (full width at half of maximum) was in the range of (8.8 – 14.6)% for 662 keV $\gamma$ line of $^{137}$Cs.

As an example of the energy distribution accumulated in the measurements, in Fig. 1 the spectra collected in some runs are reported [4]. Some peaks in the spectra can be ascribed to $\gamma$ lines of naturally occurring radionuclides such as $^{40}$K, $^{214}$Bi ($^{238}$U chain) and $^{208}$Tl ($^{232}$Th) from the materials of the set-up. A detailed investigation of the measured spectra has been performed by using various data analysis strategies and Monte Carlo simulations. In particular, the technique of the time-amplitude analysis (described in details in [7]) has been used in order to estimate the $^{228}$Th and $^{227}$Ac ($^{235}$U family) activities. The pulse shape discrimination (PDS) capability of the $ZnWO_4$ scintillators has been considered in order to study the measured $\alpha$ spectra and the so called BiPo events. The radioactive contamination of the $ZnWO_4$ crys-

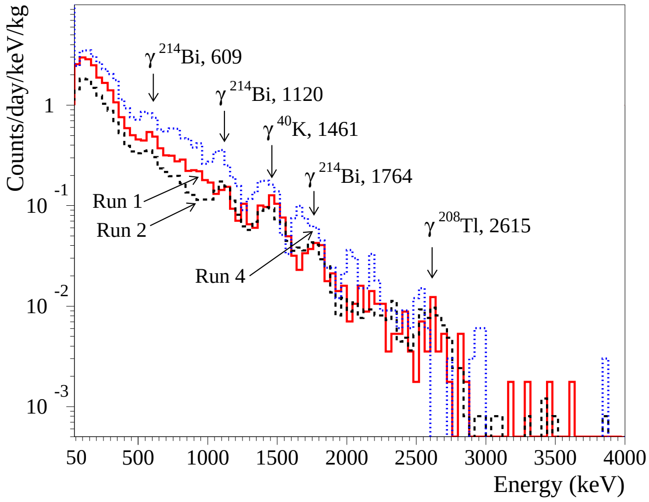

**Figure 1.** Energy distributions of the $ZnWO_4$ scintillators measured in the low background set-up during Runs 1 (with ZWO-1 in 2906 h) (solid red line), 2 (with ZWO-2 in 2130 h) (dashed black line), and 4 (with ZWO-4 in 834 h) (dotted blue line). Energies of $\gamma$ lines from residual contaminations are in keV.

tals is on the level of 0.002 – 0.025 mBq/kg; the total $\alpha$ activity is in the range 0.2 – 2 mBq/kg. Moreover, particular contaminations associated with the composition of $ZnWO_4$ detector were observed [4]: the *EC* active cosmogenic nuclide $^{65}$Zn – that can be also produced by neutrons – ($T_{1/2}$ = 244.26 d [8]) with activity 0.5 – 0.8 mBq/kg (depending on the $ZnWO_4$ sample) and the $\alpha$ active tungsten isotope $^{180}$W (with half-life: $T_{1/2} \approx 10^{18}$ yr [4, 9–11], and energy of the decay: $Q_\alpha$ = 2508(4) keV [12]) with activity 0.04 mBq/kg. In Fig. 2 it is shown the energy distribution of the $\beta(\gamma)$ events (identified by the PSD) accumulated in the low background set-up with the ZWO-4 crystal scintillator over 4305 h (Run 5) together with the model of the background. The main components of the background are shown: spectra of internal $^{65}$Zn, $^{90}$Sr-$^{90}$Y, daughters of $^{238}$U, and the contribution from the external $\gamma$ quanta from PMTs and Cu box in these experimental conditions.

A summary of the radioactive contamination of the $ZnWO_4$ crystal scintillators can be found in ref. [4].

### 1.1 Results of the search for $\beta\beta$ decay in Zn and W isotopes and on rare $\alpha$ decay of W isotopes

The collected data have also been considered to search for double beta decay processes [5]. Zinc tungstate scintillators contain four potentially $2\beta$ active isotopes: $^{64}$Zn, $^{70}$Zn, $^{180}$W and $^{186}$W. It is worthwhile mentioning that $^{64}$Zn and $^{186}$W have comparatively large natural abundance that allows to apply $ZnWO_4$ detectors without using high cost enriched isotopes. Moreover, the $2\nu2\beta^-$ decay of $^{186}$W is expected to be strongly suppressed which could provide favorable conditions to search for neutrinoless $2\beta^-$ decays, including processes with emission of majoron(s) which have broad energy spectra, somewhat similar to that of the two-neutrino mode. The $^{180}$W isotope is also an interesting $2\beta$ nuclide because in the case of the capture of two electrons from the K shell ($E_K$ = 65.4 keV), the decay energy is rather small (13 ±4) keV; such a coincidence could give a resonant enhancement of the $0\nu$ double electron capture to the corresponding level of the daughter nucleus.

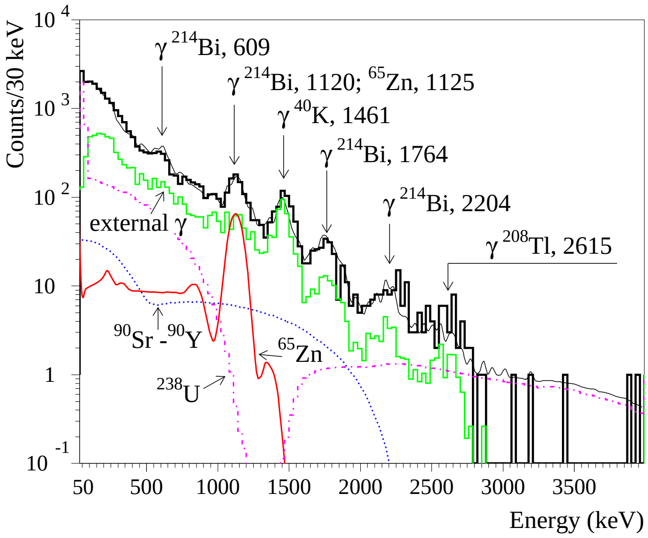

**Figure 2.** Energy distribution of the $\beta(\gamma)$ events (identified by the PSD) accumulated in the low background set-up with the ZWO-4 crystal scintillator over 4305 h (Run 5) together with the model of the background. The main components of the background are shown: spectra of internal $^{65}$Zn, $^{90}$Sr-$^{90}$Y, daughters of $^{238}$U, and the contribution from the external $\gamma$ quanta from PMTs and Cu box in these experimental conditions.

The response functions of the $ZnWO_4$ detectors for the $2\beta$ processes in Zn and W isotopes were simulated with the help of the GEANT4 package [13] with the Low Energy Electromagnetic extensions. The initial kinematics of the particles emitted in the decays was generated with the DECAY0 event generator [14]. The background models included the internal contamination of the $ZnWO_4$ scintillators ($^{40}$K, $^{60}$Co, $^{65}$Zn, $^{87}$Rb, $^{90}$Sr-$^{90}$Y, $^{137}$Cs), and the external $\gamma$ rays from radioactive contamination of the PMTs and the copper box ($^{40}$K, $^{232}$Th, $^{238}$U). Comparing the simulated response functions with the measured energy spectra of the $ZnWO_4$ detectors, no clear peculiarities, which can be evidently attributed to double beta decay of zinc or tungsten isotopes, have been found. Therefore only lower half-life limits have been set.

As an example, in Fig.3 the energy spectrum of the $ZnWO_4$ crystal scintillator ⌀41 × 27 mm measured over 2798 h, corrected for the energy dependence of detection efficiency is reported, together with the $2\nu2K$ peak of $^{64}$Zn with $T_{1/2}^{2\nu2K} = 1.1 \times 10^{19}$ yr excluded at 90% C.L.

Several $\beta\beta$ decay modes have been studied and competitive limits have been set. The main results are shortly summarized.

The previous limits on the $T_{1/2}$ of the $\beta\beta$ decay modes of $^{64}$Zn, $^{70}$Zn, $^{180}$W and $^{186}$W have been improved up to 2 orders of magnitude. In particular, new improved half-life limits on double electron capture and electron capture with positron emission in $^{64}$Zn have been set in the range:

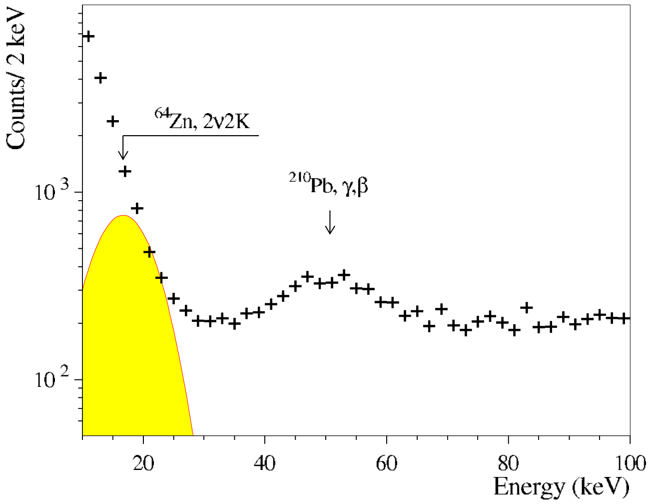

**Figure 3.** The energy spectrum of the $ZnWO_4$ crystal scintillator ⌀41 × 27 mm measured over 2798 h, corrected for the energy dependence of detection efficiency, together with the $2\nu 2K$ peak of $^{64}$Zn with $T_{1/2}^{2\nu 2K} = 1.1 \times 10^{19}$ yr excluded at 90% C.L.

$10^{19}$ yr to $10^{21}$ yr depending on the mode. Similar sensitivity have been also reached for the $2\beta$ processes in $^{70}$Zn, $^{180}$W, and $^{186}$W ($10^{18} - 10^{21}$ yr ); note that the $0\nu 2\varepsilon$ capture in $^{180}$W is of particular interest due to the possibility of resonant process [5]. The possible preliminary indication on the $(2\nu + 0\nu)\varepsilon\beta^+$ decay of $^{64}$Zn with $T_{1/2} = (1.1 \pm 0.9) \times 10^{19}$ yr suggested in [15] is disproved by the results of these measurements. It is worth noting that to date only six nuclides ($^{40}$Ca, $^{78}$Kr, $^{96}$Ru, $^{106}$Cd, $^{112}$Sn, and $^{120}$Te) among 34 candidates to $2\varepsilon$, $\varepsilon\beta^+$, and $2\beta^+$ processes were studied at $10^{21}$ yr level of sensitivity in direct experiments. However, the limits are still far from theoretical predictions.

Rare $\alpha$ decay of $^{180}$W has also been investigated in the same data by obtaining a new measurement of the half-life $T_{1/2} = 1.3^{+0.6}_{-0.5} \times 10^{18}$ yr. Also a new half-life limit on $\alpha$ transition of $^{183}$W to the $1/2^-$ 375 keV metastable level of $^{179}$Hf has been set as $T_{1/2} \geq 6.7 \times 10^{20}$ yr [5].

## 2 $ZnWO_4$ scintillator to investigate Dark Matter particle directionality

The above mentioned measurements and the correlated R&D works have shown that the $ZnWO_4$ scintillators can offer suitable features to investigate some Dark Matter (DM) particle candidates by exploiting the directionality technique. The use of anisotropic scintillators to study the directionality signature was proposed for the first time in ref. [16] and revisited in [17]. The directionality technique is effective only for those DM candidate particles able to induce just nuclear recoils. This approach studies the correlation between the arrival direction of the DM particles and the Earth motion in the galactic rest frame. In fact, the dynamics of the rotation of the Milky Way galactic disc through the halo of DM causes the Earth to experience a wind of DM particles apparently flowing along a direction opposite to that of the solar motion relative to the DM halo. However, because of the Earth's rotation around its axis, their average direction with respect to an observer fixed on the Earth changes during the sidereal day. The possible nuclear recoils induced by the DM particles are expected to be strongly correlated with their impinging direction, while the background events are not; therefore, the study of the nuclear recoils direction can offer a way for pointing out the presence of the considered DM candidate particles.

The main advantages of $ZnWO_4$ scintillators to study the directionality are: i) high level of radio-purity reachable in future development, considering the very good results already achieved; ii) an energy threshold at level of few keV reachable (room for further improvement is possible); iii) light output of heavy particles (p, $\alpha$, nuclear recoils) depending on the impinging direction of the particles with respect to the crystal axes, while the response to $\gamma$ radiation being isotropic; iv) the scintillation decay time showing the same property.

The anisotropic effect has been ascribed to preferred directions of the excitons' propagation in the crystal lattice affecting the dynamics of the scintillation mechanism [18]. The anisotropic features of the $ZnWO_4$ detectors can provide two independent ways to exploit the directionality approach. In particular, the presence of heavy ionizing particles with a preferred direction (like recoil nuclei induced by the DM candidates considered here) could be discriminated from the electromagnetic background by comparing the low energy distributions measured by using different orientations of the crystal axes along the day. Moreover, the directionality technique can also be explored at some extent studying the time behaviour of the induced nuclear recoil pulses.

Thus, in the case of the $ZnWO_4$ detector, the anisotropy of the light output for recoiling nuclei induced by DM candidates could be discriminated from the electromagnetic events because of the expected variation of their detected energy distribution during the day [17]. In fact because of the Earth's daily rotation around its axis the preferential impinging direction of DM particle change during the sideral time and the expected counting rate in a defined energy interval is expected to have a diurnal variation. This peculiarity of the rate can be considered to investigate the presence DM particle candidate inducing just nuclear recoils in the Galactic halo. Detailed discussion can be found in ref. [19].

## 3 Conclusion

In the last years radiopure $ZnWO_4$ crystal scintillator have been realized and have been measured in the DAMA/R&D facility at the Gran Sasso National Laboratory over more than 19 thousands hours. The measurements allowed us to study in details the radioactive residual contaminations of these crystals. The reached very good level of radiopurity and the study of the procedures followed to grow these crystals give us confidence for future developments of new $ZnWO_4$ crystal scintillators with higher level of radiopurity.

A low background experiment to search for $2\beta$ processes in $^{64}$Zn, $^{70}$Zn, $^{180}$W, and $^{186}$W was also carried out with a total exposure of 0.5295 kg × yr. New improved half-life limits on double beta decay modes in Zn and W isotopes in the range $10^{19}$ yr to $10^{21}$ yr have been set. The indication on the $(2\nu + 0\nu)\varepsilon\beta^+$ decay of $^{64}$Zn with $T_{1/2} = (1.1 \pm 0.9) \times 10^{19}$ yr suggested in [15] is not confirmed. Note that to date only four nuclides ($^{40}$Ca, $^{78}$Kr, $^{112}$Sn, and $^{120}$Te) among 34 candidates to $2\varepsilon$, $\varepsilon\beta^+$, and $2\beta^+$ processes were studied at $10^{21}$ level of sensitivity in direct experiments. However, it is worth noting that the limits are still far from theoretical predictions.

A search for rare $\alpha$ decay in W isotopes have also been performed. The rare $\alpha$ decay of $^{180}$W with a half-life $T_{1/2} = 1.3^{+0.6}_{-0.5} \times 10^{18}$ yr has been observed and new half-life limit $T_{1/2} \geq 6.7 \times 10^{20}$ yr on $\alpha$ transition of $^{183}$W to the $1/2^-$ 375 keV metastable level of $^{179}$Hf has been set.

The anisotropic features of $ZnWO_4$ crystals make them very interesting target to investigate dark matter particles directionality.

Thus the $ZnWO_4$ crystal scintillators can be very good detectors for future developments and experiments either to study $\beta\beta$ decay modes and to investigate the directionality for some dark matter candidate particles.

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