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Study of Dark Matter with directionality approach using $ZnWO_4$ crystal scintillators

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Abstract. Low-background anisotropic scintillators represents an innovative approach to study the presence, in the galactic halo, of those Dark Matter (DM) candidate particles able to induce just nuclear recoils, by exploiting the directionality approach. $ZnWO_4$ crystal scintillators are particularly well-suited for such investigations, since the light output and scintillation pulse shape vary depending on the angle of incidence of heavy particles (e.g., α particles and nuclear recoils) relative to the crystal axes. Due to this anisotropic behavior, a signal induced by those DM candidates can be investigated in two independent modes: studying the directionality variation both of the signal rate and of the pulse shape discrimination from the γ/β radiation (that does not give rise to any anisotropic effects). Additionally, the detector's sensitivity spans a wide range of DM masses, attributed to the differing atomic masses of its target nuclei (Zn, W, and O). Building on these characteristics, the ADAMO project carried out new studies to examine the anisotropic response of $ZnWO_4$ scintillators to α particles and nuclear recoils induced by neutron scattering. A summary of these investigations are presented in this paper.

1 Introduction

The existence of DM has been confirmed through extensive cosmological and astrophysical evidence, suggesting that a significant fraction of it could consist of relic particles. Among the direct detection methods developed to explore DM particles in the galactic halo, the directionality approach provides a distinctive signature to separate DM signals from background

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noise [1–4]. This method aims to identify DM by correlating nuclear recoil directions with the Earth's galactic motion. Detecting anisotropy in the directions of nuclear recoils could offer deeper insights into those DM candidate particles able to induce nuclear recoils and refine our understanding of related astrophysical scenarios. Moreover, other signatures for direct detection experiments, offering different sensitivities to various classes of DM candidates, are: a diurnal modulation caused by the Earth's rotation around its axis [5], a daily variation of the interaction rate caused by the DM particles' different depths [6] and the annual modulation of the interaction rate, successfully explored by the DAMA collaboration [7–13].
Anisotropic scintillators are suitable detectors to study DM candidate particles by employing the directionality approach, since their response to particles capable of inducing nuclear recoils, in terms of quenching factor (Q.F.) and pulse shape, depends on the orientation of the crystallographic axes relative to the particle trajectory. Consequently, the orientation of the axes with respect to the direction of DM particles changes throughout the sidereal day, causing variations in the measured low-energy spectrum for nuclear recoils. As a result, the counting rate within a specific low-energy range exhibits a distinct pattern over the sidereal day, enabling the discrimination of DM events from the electromagnetic background (see, e.g., Refs. [3, 4, 14, 15]). Originally proposed in Ref. [3] and refined in Ref. [4] for the study of the directionality signature, this detection approach first considered anthracene scintillators. However, operational challenges limited their practicality. To address these issues, $ZnWO_4$ crystal scintillators were later proposed as a viable alternative [14].

2 Development of new advanced $ZnWO_4$ crystals

With the purpose to obtain high quality $ZnWO_4$ crystal scintillators in terms of optical and scintillation performance, a comprehensive R&D was done using the Czochralski crystal growth method. This process involved adjusting the stoichiometric composition of the starting WO_3 material sourced from different suppliers. Additional steps included single or double crystallization, with or without annealing of the resulting boules. The luminescence characteristics of the synthesized $ZnWO_4$ crystals were examined across a temperature range from 85 K to room temperature [16]. This included studying their emission spectra, luminescence intensity as a function of temperature and dose, phosphorescence, and thermally induced luminescence. Particular focus was placed on thermally stimulated luminescence up to 350 K. The scintillation properties were assessed using γ-ray sources such as ^{60}Co, ^{137}Cs, ^{207}Bi, ^{232}Th, and ^{241}Am. For example, Fig. 1 shows the energy spectrum of γ-ray quanta from ^{137}Cs and ^{241}Am sources, measured using the $ZnWO_4$ crystal sample with the highest light yield (see Ref. [16]). It is noteworthy that such excellent energy resolution (R, full-width at half-maximum over peak position) has not been previously reported for $ZnWO_4$ crystal scintillators. Optical transmission spectra were also measured within the 300-700 nm wavelength range. The highest optical and scintillation performance was observed in $ZnWO_4$ crystals produced via single crystallization from a stoichiometric $ZnWO_4$ compound derived from highly purified WO_3 and annealed in an air atmosphere (see Ref. [16] for additional details). Additionally, since both phosphorescence and dose dependence of XRL intensity are negligible in scintillation measurements, no observable correlation was found between the scintillation light output and the luminescence intensity of the samples. This lack of correlation suggests that improvements in $ZnWO_4$ production methods are still possible. In particular, the quality of the scintillators—especially those produced through double crystallization—has yet to reach its full potential. Further RD efforts are underway, focusing on producing larger volume crystals for low-background experiments.

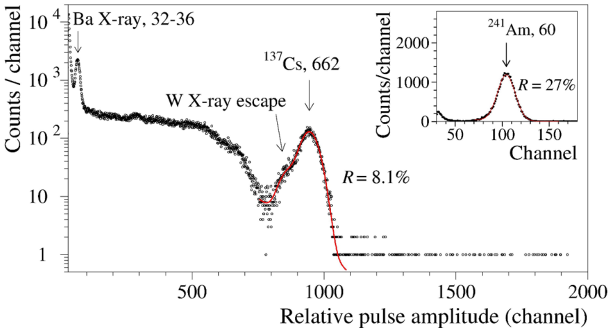

Figure 1. The energy spectra of γ-ray quanta from ^{137}Cs measured with a scintillation detector employing the $ZnWO_4$ crystal sample No. 84, as detailed in Ref. [16]. The inset displays the energy spectrum of γ-ray quanta from ^{241}Am. The energies of X-ray and γ-ray quanta are reported in keV.

3 Investigation of the anisotropic properties of $ZnWO_4$ crystal scintillators

3.1 Studies with α particles

The initial studies of the anisotropic behavior of $ZnWO_4$ and their quenching factors (Q.F.s) were conducted using α particles, as reported in Ref. [17]. Subsequent measurements were performed in Ref. [18] with a small $ZnWO_4$ crystal and a ^{241}Am source, employing various layers of thin mylar films to reduce the energy of the α particles. For each measurement, the energy scale of the crystal was calibrated using ^{137}Cs and ^{22}Na γ sources. The energy spectra of α particles interacting along the three crystallographic axes of the $ZnWO_4$ crystal are shown in Fig. 2-Left. The orientations of the crystal were adjusted so that the α beam was directed perpendicular to the (100), (001), and (010) crystallographic planes, corresponding to axes I, II, and III, respectively. Fig. 2-Right displays the dependence of the Q.F.s on

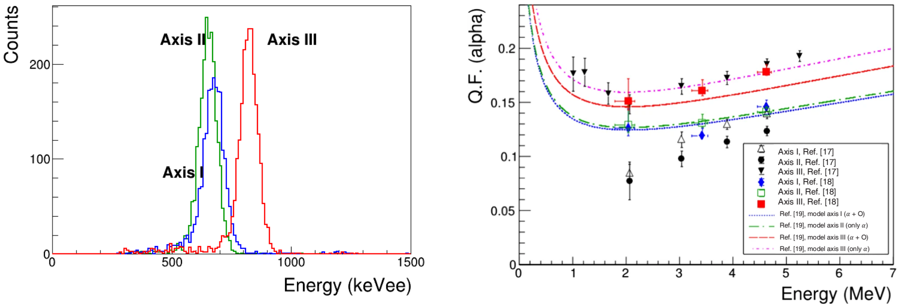

Figure 2. Left: Energy spectra of 4.63 MeV α particles (expressed in keVee) for incident directions along the three crystallographic axes of the $ZnWO_4$ crystal. Right: The α/β ratio as a function of α particle energy, measured with a $ZnWO_4$ scintillator in Ref. [17] (black points) and compared to results from Ref. [18] (colored points). The crystal's anisotropic response is clearly visible. Models corresponding to each crystallographic axis, obtained from global fits of all recoil and α particle data (see text), are shown based on Ref. [18] and aligned with Ref. [19].

energy for the three orientations of the α beam relative to the crystallographic axes. The Q.F. measured along axis III is approximately 1.2 times greater than the values recorded along axes I and II, which are closely matched to each other. The Q.F. values and anisotropic effects reported in Ref. [18] are consistent with the data in Ref. [17], as shown in Fig. 2-Right. The same figure also includes predictions for Q.F. behavior along each crystallographic axis based on the model from Ref. [19]. These findings reinforce the evidence for the anisotropic properties of $ZnWO_4$ scintillators when exposed to α particles with energies up to several MeV.

3.2 Studies with neutron generator

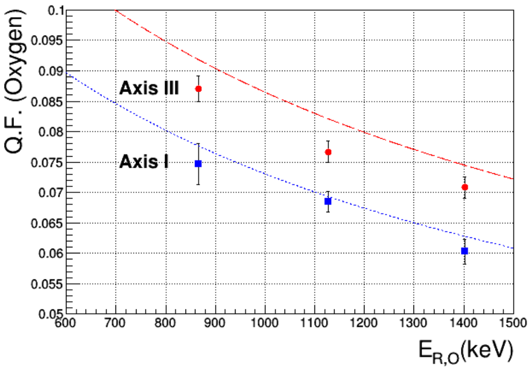

Figure 3. The Q.F.s values for oxygen nuclear recoils in ZnWO4 are shown as a function of the expected recoil energies ER, O for crystallographic axes I and III. The plot also includes the predicted Q.F. behavior for these axes, derived from global fits of α particle and oxygen recoil data, following the model in Ref. [19] (see Ref. [18] for further details).

The recoil energies of oxygen nuclei were studied using the same $ZnWO_4$ crystal and a monochromatic neutron generator. A detailed description of the experimental configuration and data analysis procedure is provided in Ref. [18], specifically in Fig. 4 of that work. The resulting Q.F.s are displayed in Fig. 3, alongside theoretical models for the corresponding crystallographic axes based on the framework outlined in Ref. [19] (further information is available in Ref. [18]). The study in Ref. [18] also reports clear evidence of anisotropy in oxygen nuclear recoils, observable down to recoil energies of around 100 keV, with a statistical significance of 5.4 σ (refer to Table 1 in Ref. [18]).

4 Conclusions

In summary, directionality studies offer a novel approach to investigate DM candidates that induce nuclear recoils in the galactic halo. This method could provide additional evidence for the existence of DM and shed light on the nature and interaction mechanisms of DM particles. Anisotropic $ZnWO_4$ detectors exhibit promising characteristics for exploring the directional properties of DM-induced nuclear recoils. Extensive research and development have been carried out on $ZnWO_4$ crystal scintillators to optimize their performance. First evidence of anisotropic behavior in the response of $ZnWO_4$ scintillators to nuclear recoils has been reported in Ref. [18], demonstrating sensitivity down to a few hundred keV with a statistical significance of 5.4 σ.

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