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# Feasibility study of a $ZnWO_4$ scintillator for exploiting materials signature in cryogenic WIMP dark matter searches

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**Abstract**

The mass number dependence of the WIMP–nucleus scattering offers a method for identifying a true WIMP signal over a neutron background. In this Letter we present a study on using a combination of $ZnWO_4$ and $CaWO_4$ absorbers to exploit this materials signature for WIMP detection. Using monochromatic X-ray radiation we examined the temperature variation of the luminescence properties for both materials and showed that at low temperature (8 K) $ZnWO_4$ exhibits $\sim 10\%$ higher light yield than $CaWO_4$. Analysis of relevant optical properties indicates that $ZnWO_4$ is a suitable cryogenic scintillator. We show that already modest exposure in the region of $\sim 5$ kg yr should allow the detection of WIMP interaction for cross sections at the level of current experimental sensitivities. The combination of these two tungstates could form the basis of the first multi-target detector capable of WIMP identification through materials signature.


## 1. Introduction

Most modern experiments aiming to detect WIMP dark matter are employing some form of active discrimination between nuclear recoil (the signal) and electron recoil (backgrounds). One of these discrimination techniques, used by CRESST and also ROSEBUD, is event by event discrimination through measuring a combination of phonon and scintillation signals from cryogenic detectors operating at temperatures in the milli-kelvin range [1,2]. The simultaneous detection of phonons and scintillation in dark matter searches, and in particular the use of calcium tungstate ($CaWO_4$) was pioneered by CRESST. A range of commonly available scintillators were tested for their feasibility as scintillating cryogenic detectors [1]. $CaWO_4$, having favourable properties overall, was shown to exhibit excellent discrimination [3].

A high scintillation yield in the milli-kelvin temperature range is a key criterion in the choice of a scintillating absorber in dark matter searches. Other criteria

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involve surface properties, radio purity and phonon propagation properties. $CaWO_4$ appears to be a rather satisfactory choice, providing high light yield at low temperature, and thereby offering good sensitivity for the detection of WIMP–nucleon elastic scattering. The higher the scintillation yield, the lower is the energy threshold for which discrimination between electron and nuclear recoil is possible for a given confidence level. Lower energy threshold translates into less exposure (mass × time) needed to reach certain levels of cross sections for WIMP–nucleon scattering. The characterization of scintillation materials at very low temperature has not been the mainstream of scintillation studies so far and although many scintillators have been examined at or near room temperature, little information is available at temperatures below that of liquid nitrogen.

$CaWO_4$ is an excellent target material for cryogenic dark matter searches, offering excellent discrimination between nuclear and electron recoil. Nuclear recoils are produced by WIMP scattering, but also by neutron interaction. The various types of events could be distinguished by the dependence of the scintillation yield on the type of recoiling nucleus. It is thus beneficial to have targets with a variety of nuclei, and also targets that are similar with only one nucleus different. This should allow extracting a WIMP signal via the material signature of WIMPs [4]. $ZnWO_4$ is a very interesting material in this regard. It differs from $CaWO_4$ only by having Ca replaced with Zn and the mass number of Zn ($A = 65.41$) is close to that of germanium ($A = 72.64$), thereby making cross calibration with detectors based on germanium easier [5,6].

In this Letter we report on the results of a comparative examination of the luminescence light yield of $ZnWO_4$ and $CaWO_4$ scintillators at low temperature, and present computer simulations on the sensitivity levels for WIMP–nucleon cross sections that could be accessed via a material signature.

## 2. Comparative characterization of $CaWO_4$ and $ZnWO_4$ scintillators

### 2.1. Scintillator characteristics at room temperature

An initial assessment of the feasibility of zinc tungstate as scintillating absorber for dark matter searches can be obtained from a comparison of the room-temperature scintillation characteristics of $ZnWO_4$ and $CaWO_4$. These materials have been known for decades as efficient phosphors and scintillators and their scintillation properties have been studied repeatedly [7–11]. Inspection of the data presented in Table 1 shows that $ZnWO_4$ has greater density than $CaWO_4$, meaning that $ZnWO_4$ possesses 19.3% more tungsten nuclei per volume than $CaWO_4$. With the strongest contribution to a signal from WIMP–nucleon scattering expected to involve tungsten nuclei, $ZnWO_4$ is advantageous as having a high density of tungsten nuclei. A further advantage arises from the scintillation emission spectrum, which centres near 480 nm for $ZnWO_4$ and 420 nm for $CaWO_4$. Although the exact position of the peak emission is not critical for the detection of scintillation light with a cryogenic detector (being a spectrally non-selective bolometer), there is an advantage of having the peak intensity at longer wavelength. This results from the fact that the reflectivity of most materials reduces for shorter wavelength. A peak emission of 480 nm is well positioned for many reflectors. $ZnWO_4$ has a longer scintillation time constant than $CaWO_4$ and this is likely to be the case also at low temperature. With photo-multipliers as detectors, this could be a disadvantage; however, the flexibility in design offered by cryo-detectors should allow a perfect match of time constants between scintillator and photon detector [12].

Determining the scintillation yield of a material is crucial in assessing its suitability as a target material; however, there appears to be considerable spread among the experimental results presented by different

Table 1
Comparison of characteristics of the NaI-Tl, $CaWO_4$ and $ZnWO_4$ scintillators at 300 K

| Properties | NaI-Tl | $CaWO_4$ | $ZnWO_4$ |
|---|---|---|---|
| Density, g/cm$^3$ | 3.67 | 6.06 | 7.87 |
| Light yield* | 100 | 16[a] [9] | 29[a] [9] |
| | | 27[b] [10] | 18[b] [10] |
| | | | 14 [7] |
| | | | 18 [8] |
| | | | 28 [11] |
| Emission peak, nm | 415 | 420 | 480 |
| Decay time, µs | 0.2 | 9 | 21 |
| Refractive index | 1.85 | 1.93 | 2.1 |

* Relative to NaI-Tl; [a] crystal; [b] powder.

authors (see Table 1). This spread is most likely caused by a variation of quality between the scintillator samples that were investigated, but also systematic effects could be present in such data. Additionally, in regard of low-temperature application, the scintillation yield is expected to vary with temperature and thus, values given at room temperature can only give an initial indication for the feasibility of a material for cryogenic dark matter searches. In this Letter, we report on our comparative study of $ZnWO_4$ samples and a reference $CaWO_4$ crystal. The main result is the light yield of $ZnWO_4$ and $CaWO_4$ samples as function of temperature, under excitation by monochromatic X-ray photons from a synchrotron source.

## 2.2. Samples and experimental technique

We examined the off-cut of a $CaWO_4$ scintillator manufactured by SRC "Carat" (Ukraine) and a $ZnWO_4$ sample produced by Spectra-Physics Hilger Crystal (UK). The crystals where grown from the melt, both using the Czochralski technique, in iridium and platinum crucibles, respectively. According to the results of an ICP-MS (Inductively Coupled Plasma Mass Spectrometry) analysis the $CaWO_4$ crystal contains admixtures of Sr ($\sim$ 80 ppm), Ba ($\sim$ 30 ppm), Zn ($\sim$ 2 ppm), Ni, Cr, Cu, Gd, Pb ($\sim$ 1 ppm) and traces ($<$ 0.1 ppm) of a number of other impurities (Y, Ce, Nd, Sm). The $ZnWO_4$ sample is very clean in comparison; it contained $\sim$ 1 ppm of Cr and $\sim$ 0.1 ppm of Ba while the content of other metal impurities (35 in total) was below the detection limit of 0.02 ppm. The samples investigated in luminescence measurements were of size $5 \times 5 \times 1$ mm$^3$. The surfaces were mechanically polished to optical grade for the $CaWO_4$ sample and cleaved from an ingot in case of $ZnWO_4$.

Luminescence characterization of $CaWO_4$ and $ZnWO_4$ crystals was carried out at the MPW6.1 and 4.2 beamlines of the SRS Daresbury laboratory using the mobile luminescence end-station MoLES [13]. Using two different beamlines gave us access to X-ray photons of two excitation energies: 320 eV and 3.2 keV. These excitation energies were chosen to investigate in particular the response of the scintillators near the expected detection threshold for large cryogenic detectors used in dark matter searches. The samples where mounted in the holder of a closed-cycle cryostat, allowing measurement of luminescence spectra over the temperature range from 8 K to 300 K. The spectrometer for measuring the wavelength-resolved luminescence spectrum was a Triax-190 Jobin–Yvon monochromator and for photon detection, a GaAs Hamamatsu R2949 photomultiplier was used. The spectra presented in Fig. 1 are corrected for instrumental response of the detection system. All measurements were carried out in a fixed geometry and for equal size and shape for the two samples. This allowed direct comparison of the light output of the crystals. The flux of the incident radiation was monitored by measuring of the drain current signal from an aluminium foil or photodiode.

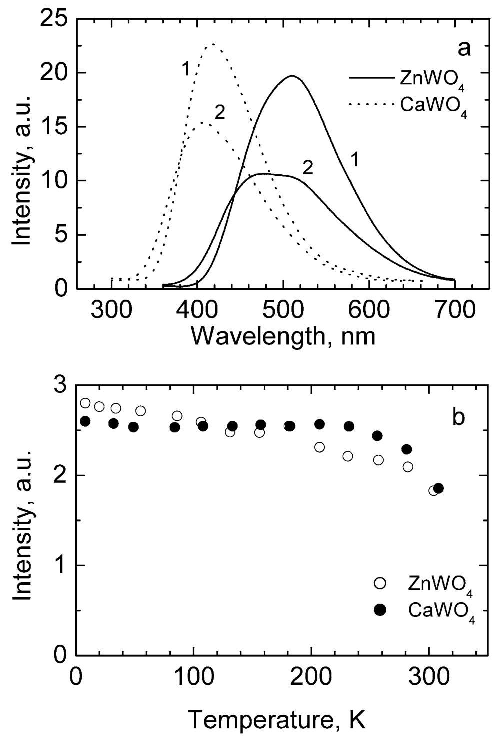


Fig. 1. (a) Emission spectra of $ZnWO_4$ and $CaWO_4$ crystals measured under excitation with 3.2 keV photons at $T = 8$ K (1) and 300 K (2). (b) Temperature dependence of the integrated luminescence yield. The integrated light yield of $ZnWO_4$ at a temperature of 8 K is $\sim$ 1.1 that of $CaWO_4$.

## 2.3. Experimental results

The luminescence spectra of $CaWO_4$ and $ZnWO_4$ scintillation crystals measured at a temperature of 8 K under excitation with 3.2 keV photons are presented in Fig. 1(a). The luminescence spectra of $CaWO_4$ exhibit the characteristic features expected for a crystal of sheelite structure; a structureless broad band emission centred at 420 nm wavelength with a pronounced extension towards longer wavelength. The prominent peak in the spectrum is the characteristic emission of calcium tungstate, interpreted as radiative transition of the charge transfer type between tungsten and oxygen within the $(WO_4)^{2-}$ molecular complex [14]. The extension on the long-wavelength side is attributed to the emission of defect centres. As the temperature increases, the emission maxima shift towards higher energy and the luminescence spectra exhibit broadening; such thermal changes are mostly due to interaction of the $(WO_4)^{2-}$ emission centre with lattice vibrations [15].

Zinc tungstate has wolframite crystal structure and the luminescence properties of this crystal are noticeably different from $CaWO_4$. At 8 K the luminescence spectrum spans the range from 400 nm to 700 nm showing a prominent emission peak at 510 nm (see Fig. 1(a)). This emission is associated with the radiative transitions between tungsten and oxygen within the $(WO_6)^{6-}$ molecular complex [16]. The position of the main emission peak of $ZnWO_4$ exhibits a significant shift towards shorter wavelength (from 510 nm to 480 nm) as the temperature increases to room temperature. This shift is a characteristic feature of all tungstates with wolframite crystal structure [17]. It is assumed that such temperature changes are due to the complex structure of the luminescence spectrum which is composed of several sub-bands [16,18]. Temperature variation of the relative intensity of these bands causes a corresponding change of the luminescence spectra.

Fig. 1(b) shows the variation of the integrated emission light output as function of temperature. For $CaWO_4$ this parameter shows very little variation over the temperature range from 8 K to 250 K; above that, thermal quenching causes a decrease of the light output. Zinc tungstate has a maximum light output at a temperature of 8 K, which decreases gradually with increasing temperature. At a temperature of 8 K and 3.2 keV excitation energy, the relative light output of $ZnWO_4$ is assessed to be by $\sim 10\%$ higher than that of $CaWO_4$. At room temperature the light yield of both samples is virtually identical. The same measurements of the light yield as function of temperature were carried out at 320 eV excitation energy. At this energy, the ratio for the light yield ($\sim 1.15$) obtained for the two crystals at 8 K is the same within the experimental accuracy ($\pm 10\%$). These results provide a strong indication that the intrinsic light yield of $ZnWO_4$ at low temperature is higher than that of $CaWO_4$.

Dark matter search experiments require large absorber crystals, e.g., in CRESST, cylinders of 40 mm diameter and 40 mm height are used. While scintillation efficiency is an important issue, other parameters, such as light absorption within the crystal are equally important. An indication of light absorption can be obtained from measurements of the transmittance; however, there are several experimental complications, such as scattering and multiple reflections that need to be taken into account. Especially multiple reflections tend to become very complex in the case of optically anisotropic materials [19]. In order to avoid these complications, an approximation that considers the crystal as an isotropic material has been adopted. Transmittance spectra were measured with unpolarized light and the results were corrected for reflectivity losses using reflection coefficients with a spectral dependence based on the averaged dispersions taken from [20] and [21]. This procedure introduces an error on the absorption coefficient of less than 3% for $CaWO_4$ and less than 8% for $ZnWO_4$ owing to the anisotropy of the refractive index.

Fig. 2 shows the optical absorption spectra of $CaWO_4$ and $ZnWO_4$ crystals measured at temperatures of 300 K and 8 K. Reducing temperature results in a shift of the absorption edge to lower wavelength, associated with an increase in the energy gap. Light pink coloration is inherent for thick $ZnWO_4$ crystals and the absorption band visible at 360 nm is the cause of this. Such coloration is usually attributed to trace impurities of iron and/or chromium ions [18]. The fundamental absorption of $CaWO_4$ sets in below 280 nm. At lower temperature, where the absorption edge is shifted towards shorter wavelength, an additional feature, identified by a “shoulder”, becomes apparent. This feature is attributed to the presence of defects. Fig. 2 further shows that $CaWO_4$ is more transparent than $ZnWO_4$ below a wavelength of 400 nm. This, however, is of little effect for the luminescence yield as most important is the absorption in the wavelength region of the emitted luminescence band. Taking into account the positions of the emission bands for the

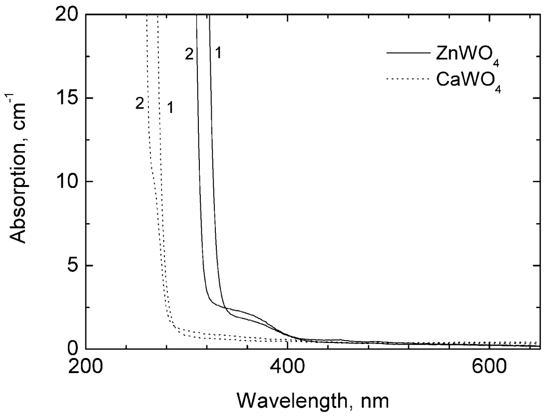


Fig. 2. Optical absorption spectra of $ZnWO_4$ and $CaWO_4$ crystals measured at temperatures of 300 K (1) and 8 K (2), respectively. The spectra are corrected for reflection losses.

two materials, the absorption is effectively the same for both materials at low temperatures.

Summarizing the results of experimental studies we can draw the conclusion that because of the high light yield of $ZnWO_4$, a low-temperature scintillation detector made from this material should perform at least at the same level as a detector with a $CaWO_4$ absorber.

## 3. WIMP material signature sensitivity

The operation of two different target materials for particle dark matter detection offers the exciting possibility of exploiting the target material dependence of the scalar WIMP–nucleus scattering cross section as a signature [4]. We carried out Monte Carlo simulations on the material signature for $CaWO_4$ and $ZnWO_4$ detectors operating together in the same experimental setup.

### 3.1. Background estimate

The key issue in calculating sensitivity levels for discrimination by material composition is knowledge of the spectral shape of possible backgrounds. Discrimination via phonon and scintillation measurement removes backgrounds resulting in electron recoils, which leaves nuclear recoils caused by neutron interaction as the dominant source of background. The energies to consider for a WIMP signal range from the discrimination threshold (typically 10 keV) up to 50 keV. Above $\sim 20$ keV one can approximate the spectrum of neutron energies as being inversely proportional to energy; below $\sim 20$ keV an approximation as exponential in energy is more appropriate.

In the following we assume the background as being caused by neutron elastic scattering in the target material. Monte Carlo simulations of each resulting recoil spectrum are obtained using the GEANT4 simulation package [22]. These recoil energy spectra are normalised and used as probability density functions in Monte Carlo simulations of the background energy spectra.

We found that the shape of the recoil energy spectrum exhibits only negligible dependence on variations in the spectral shape of the incoming neutron flux. We used existing neutron flux measurements [23] and varied them within the uncertainties given for each energy bin and observed that no significant variation is detectable in the resulting recoil spectrum shape in the relevant energy interval between five and hundred keV. To further demonstrate the negligible dependence of the recoil spectrum on the spectral shape of the neutron flux, we used the simulation of the recoil energy spectrum resulting from the neutron flux which has been transported through the existing CRESST lead and copper shield [24,25]. Again, no significant deviations can be observed in the relevant energy interval. All results presented below have been obtained by using the latter set of neutron fluxes as it is considered to be a realistic model for the neutron flux reaching the detectors.

Another observation in connection with background neutron events on $CaWO_4$ and $ZnWO_4$ is that the shapes of the nuclear recoil spectra are very similar. For small recoil energies (below $\sim 10$ keV), scattering by tungsten nuclei gives the dominant contribution to the WIMP signal. Above $\sim 50$ keV, oxygen dominates and in the range between, recoil of zinc or calcium nuclei contribute to the WIMP signal, differing by only $\sim 10\%$. For the following data analysis method we used the simulated background spectra for each individual detector material in order to keep systematic uncertainties as small as possible.

### 3.2. Data analysis method for the material signature

The data analysis algorithm for calculating minimum sensitivities for WIMP signature detection is based on a Monte Carlo technique, for which the assumptions are:

- the exposure (mass × time) for $CaWO_4$ and $ZnWO_4$ detectors is the same,
- both detectors share the same background, operating in the same set-up concurrently,
- signal-to-background ratio is 10 (see comments below on this ratio),
- energy threshold is 10 keV,
- the WIMP signal for each detector material is calculated following [26],
- only spin-independent (scalar) interactions of WIMPs are considered,
- WIMP–nucleon scattering scalar cross sections of $10^{-6}$ pbarn and $10^{-7}$ pbarn are assumed.

The data analysis algorithm includes the following steps:

- calculate the WIMP signals in both materials for a range of WIMP masses,
- produce a WIMP spectrum (Poisson-distributed with mean being the expected rate per energy bin) with total count rate $s_m$ for each material M,
- produce Poisson-distributed background spectra containing 10% of the total simulated signals ($b_m = 0.1 s_m$) and add these to the signals $s_m$ in order to obtain recoil spectra of signal-plus-background, $S_m$,
- produce another background spectrum with total count rate $B_m$ equal to $S_m$,
- fit the expected WIMP signal on each material to the simulated $S_m$ (see description below) and to the simulated background spectra and store the fit results,
- repeat the simulation for 1000 random measurements and analyse the results.

We use a maximum likelihood fit procedure such that the model must be optimised for both materials simultaneously[1] once for the two random signal-plus-background samples and then for the two pure background samples. The model is defined as containing the WIMP signal and background. The WIMP signal contains two fit parameters, WIMP–nucleon scalar cross section and WIMP mass. The background has one free parameter. These three parameters are calculated for each set of $S_m$ and $B_m$ and their distribution is used for the subsequent hypothesis test, which in the end determines the minimum exposure for WIMP detection.

Our hypothesis testing works as follows: the distribution of fit parameters is approximated as Gaussian in the three-dimensional parameter space which might seem presumptuous for 1000 data points. However, we checked the validity of the assumption subsequently with selected high statistic (and computationally intensive) Monte Carlo simulations for $10^5$ events. We require the three-dimensional distribution to contain the three known parameter values each in a parameter interval $\pm 2.5$ (2 s.f.) times the standard deviations of the single distributions around their respective means. This requirement results from quantiles of the $\chi^2$-distribution for three fitted parameters [27] and defines our acceptance to 90% C.L. for WIMP parameter reconstruction from $S_m$.

For the power of the test, i.e., how many pure background samples would be identified as successful WIMP fits in this Monte Carlo simulation, we also require a 90% C.L. which means less than 100 neutron samples shall be identified as WIMP signals.

In contrast to WIMP signatures based on a stimulus with known time-structure, i.e., annual or diurnal modulation, we found that the material signature requires a signal-to-background ratio of at least one for medium and high WIMP masses. Low-mass WIMPs exhibit a spectral shape sufficiently different to background spectral shapes to be still detectable even with a signal to background ratio of one. Nevertheless, for a realistic WIMP search one should aim for an experimental setup which offers a large signal to background ratio for as large a range of WIMP cross sections as possible. Due to the WIMP mass dependence of this signature, we recommend an ‘on/off-approach’ to verify a possible detection. A measurement utilising the best possible signal to background ratio should be compared to a previous and/or subsequent measurement with increased background. In our simulations we found that spectra containing neutron background only give rise to a goodness of fit nearly indepen-

[1] We determine the logarithmic likelihood function for each detector separately first and then add them to obtain the full likelihood function to be maximised.

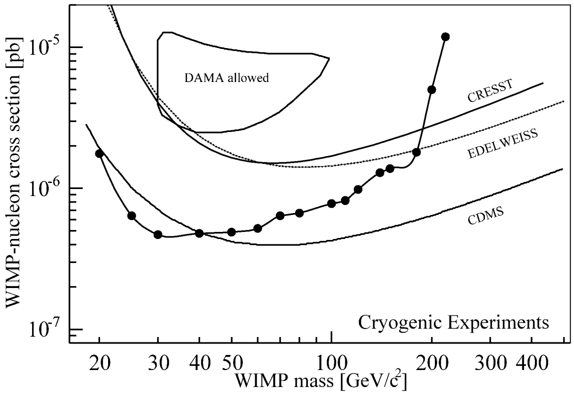


Fig. 3. Cross section sensitivity limit (•) for material signature using $CaWO_4$ and $ZnWO_4$ targets calculated for WIMP–nucleon scalar interaction. Calculations were carried out for WIMP masses corresponding to the solid marker symbols; the smooth curve through the calculated points is an interpolation shown as guide to the eye. An exposure of 5 kg yr for each of $CaWO_4$ and $ZnWO_4$ was used in the calculations. Discrimination via material signature is possible for interaction cross sections above the curve. Further details on the calculations resulting in this plot are given in the text. For comparison, recent upper limits for cross sections from leading cryogenic dark matter searches are shown; and the evidence region reported by DAMA [29–32].

dent of WIMP masses above $\sim 50$ GeV. Attempts to fit such a neutron data sample effectively return the start value for the WIMP mass as best fit. If, however, the recoil spectrum contains a contribution from WIMP interaction, the fit procedure behaves in a robust way to variations of the initial parameter values, provided the signal is statistically significant and the signal-to-background ratio is large enough (we so far checked for a ratio of 10 only). Recoil energy spectra with a lower signal to background ratio should not exhibit a robust solution with regard to WIMP signal identification. In that case the goodness of fit is nearly independent of WIMP mass, indicating neutron background as discussed above.

The minimum WIMP–nucleon scalar interaction cross sections as a function of WIMP mass for detecting a WIMP signature with the above material combination are shown in Fig. 3. The most remarkable feature is the absolute scale of cross sections. For most WIMP masses, assuming an exposure of 5 kg yr, WIMPs with cross sections of $10^{-6}$ pbarn or less can be detected at 90% C.L. This should be compared with results from a time-dependent WIMP modulation signature [28], which is generally higher by at least an order of magnitude. An exposure of 5 kg yr (meaning 5 kg of $CaWO_4$ and 5 kg $ZnWO_4$ for one year) is sufficient to detect WIMPs with cross sections at the level of current sensitivities of cryogenic WIMP direct detection experiments [29–31]. Results for the experimentally more challenging cross section of $10^{-7}$ pbarn can be obtained by scaling the minimum exposures up by a factor of 10. This represents the near or mid-term future sensitivity of experiments. We used separate calculations for exposures of 50 kg yr as a numerical cross-check to confirm the expected factor 10 improvement in sensitivity. The steep rise towards higher WIMP masses (greater than about 180 GeV) indicates the limit of applicability of material signature detection for this particular combination of materials. For WIMP masses much greater than that of Zn and Ca the contribution to the recoil energy spectra from scattering by these nuclei tends to be small, making differences in the overall recoil spectra difficult to detect.

## 4. Conclusion

We have shown that $ZnWO_4$ is a suitable and attractive target material for a cryogenic dark matter search experiment, especially when combined with $CaWO_4$ in order to exploit the materials signature for WIMP–nucleus interaction. Operating both target materials in close proximity, within the same experimental set-up, should allow detection of WIMP interaction via a material signature already at rather modest exposures in the region of $\sim 5$ kg yr. This needs to be contrasted with the need for much higher exposures required for detecting an annual modulation signature.

$ZnWO_4$ exhibits an improvement of scintillation efficiency when cooled to low temperatures. Overall, $ZnWO_4$ appears to be an excellent complement to $CaWO_4$ in cryogenic dark matter searches based on the detection of phonon and scintillation signals. The combination of these two tungstates could form the basis for the first multi-target detector capable of WIMP identification through a material signature.

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