Measurement of the Anisotropic Response of the $ZnWO_4$ Crystal for Developing the Direction-Sensitive Dark Matter Detector

Koichi Ichimura, Hiroyuki Sekiya, Juan W. Pedersen, Akihiro Yamaji, and Shunsuke Kurosawa

***Abstract*— We measured the scintillation light yield of $ZnWO_4$ crystals using a quasi-monochromatic neutron beam energy of 865 keV. Irradiating the neutron beam on two surfaces, which are almost perpendicular to the crystal axis, the light yield of oxygen recoil was measured. The obtained light yield ratio of a neutron recoil to gamma-ray-induced electron recoil for two surfaces was 0.235 ± 0.026 and 0.199 ± 0.020, respectively, which corresponds to 15.3% of anisotropy for ∼200-keV nuclear recoils of this crystal. This property can be applied to a direction-sensitive dark matter (DM) detector.**

***Index Terms*— Calibration measurement techniques, instrumentation and measurement, scintillators, solid scintillation detectors.**

I. Introduction

THE existence of dark matter (DM) is inferred from astrophysical and cosmological observation such as the lack of radial dependence of the rotation speed of distant stars around their host galaxy. One well-motivated DM candidate is weakly interacting massive particles (WIMPs). WIMPs are expected to interact with atoms via nuclear recoil. Since the event rate for the interaction between WIMPs and atoms is expected to be extremely small, the detectors require a large target mass, low energy threshold, and low background. As our galaxy rotates, our solar system passes through the "sea" of DM and receives a "wind" of WIMPs. One method to distinguish between background and signals from DM is to observe the annually modulated signal. The earth's velocity relative to the DM distribution in the galaxy changes, as the earth moves around the sun, thus producing a modulated signal with a maximum in June at the level of a few percent in a putative DM signal rate [1]. Such a modulated signal may indicate the direct detection of the DM. For example, the DAMA group using NaI(Tl) scintillation crystals reported an annual modulated signal with 12.9 σ C.L. [2]. This report became a great motivation for the recent DM search. In addition to this annual modulated signal, if the incident direction of the signal is measured, it will allow for even greater discrimination between DM and background signals.

For this purpose, many direction-sensitive detectors—mainly gaseous particle track detectors—are being proposed or developed [3]. On the other hand, solid direction-sensitive detectors have also been suggested to increase the target mass in the search for WIMPs [4]. For example, the scintillation emission efficiency of organic crystal scintillators for heavily charged particles is known to change depending on the incident direction of the particles. A WIMP detector using this property has been suggested [5] and investigated [6]. Such anisotropy allows us to extract a diurnal modulated signal associated with the earth's rotation due to the variation of the incident direction of WIMPs with respect to the crystal. However, such organic scintillators consist mainly of hydrogen and carbon and thus are not suitable targets to search for the prevailing DM candidates such as WIMPs, WINOs, and other heavy supersymmetry (SUSY) particles [7] which expect to interact with nuclei via elastic scattering. On the other hand, the inorganic scintillator can be made from materials that contain heavy nuclei. Such heavy nuclei are suitable for the WIMP and other heavy DM search. The $ZnWO_4$ scintillation crystal is one of the inorganic scintillators having anisotropic properties. Barabash *et al.* [8] and Belli *et al.* [9] reported the development of radiopure $ZnWO_4$ crystals. With respect to the anisotropy of $ZnWO_4$, the Anisotropic detectors for DArk Matter Observation (ADAMO) group, in 2013, reported the dependence of the scintillation yields in inorganic $ZnWO_4$ scintillation crystal on the incident direction of α particles [10]. Di Marco [11] reported the preliminary results on measurements of the anisotropic features of the $ZnWO_4$ crystal with alpha and neutrons. Neutrons do not have an electric charge and interact with nuclei via elastic scattering, and this property is the same as DM such as WIMPs. Thus, neutrons are widely used for the evaluation of the DM detector response. In these proceedings, we evaluated the anisotropic scintillation response of a $ZnWO_4$ crystal by nuclear recoils using neutrons to check its performance as a material for a direction-sensitive DM detector.

Manuscript received February 8, 2020; accepted March 29, 2020. Date of publication April 2, 2020; date of current version June 19, 2020. This work was supported by JSPS KAKENHI under Grant JP15K13478, Grant JP17H02884, Grant JP17H05190, Grant JP19H02422, and Grant JP19H04684.

Koichi Ichimura is with the Research Center for Neutrino Science, Tohoku University, Sendai 980-8578, Japan (e-mail: ichimura@awa.tohoku.ac.jp).

Hiroyuki Sekiya is with the Kamioka Observatory, Institute for Cosmic Ray Research, University of Tokyo, Gifu 506-1205, Japan, and also with the Kavli Institute for the Physics and Mathematics of the Universe (WPI), University of Tokyo, Kashiwa 277-8582, Japan (e-mail: sekiya@icrr.u-tokyo.ac.jp).

Juan W. Pedersen is with the Graduate School of Arts and Sciences, University of Tokyo Komaba, Tokyo 153-8902, Japan (e-mail: pedersen@hep1.c.u-tokyo.ac.jp).

Akihiro Yamaji is with the Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan (e-mail: yamaji-a@imr.tohoku.ac.jp).

Shunsuke Kurosawa is with the Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan, and also with the Faculty of Science, Yamagata University, Yamagata 990-8560, Japan (e-mail: kurosawa@imr.tohoku.ac.jp).

Color versions of one or more of the figures in this article are available online at http://ieeexplore.ieee.org.

Digital Object Identifier 10.1109/TNS.2020.2985027

TABLE I
PROPERTIES OF $ZnWO_4$ CRYSTALS

Molar mass(g/mol)	313.22
Density (g/cm^3)	7.87
Reflective index	2.1∼2.2

TABLE II
ANGLES AND LENGTHS OF A UNIT CELL OF $ZnWO_4$ CRYSTALS

α[deg.]	β[deg.]	γ[deg.]
90.0000	90.6210	90.0000
a[Å]	b[Å]	c[Å]
4.96060	5.71820	4.92690

II. SETUP FOR THE MEASUREMENT

A. $ZnWO_4$ Crystals

The properties of the $ZnWO_4$ crystal and the information on the length and angle of the basic vector of the unit cell are summarized in Tables I and II, respectively. The $ZnWO_4$ crystal is a Wolframite-type tungstate scintillator similar to $MgWO_4$ crystal and $CdWO_4$ crystals. The light emission center of this type of scintillator is $(WO_6)^{6-}$. Two O–W bond distances (2.031 Å) are longer than the other four O–W bond distances (1.8938 and 1.8370 Å) [12]. On the other hand, other types of tungstate scintillators, such as $CaWO_4$, $BaWO_4$, and a $PbWO_4$, are regarded as Scheelite-type tungstate scintillators. The O–W bond distance of the Scheelite-type scintillator is the same. The Wolframite-type scintillator has anisotropy and the Scheelite-type scintillator does not, a difference in the bond distance may be a cause of the anisotropy [13].

One feature of the $ZnWO_4$ crystal is its long decay time. A typical scintillation light waveform with γ-ray irradiation lasts about 100 μs and shows a discrete waveform. The decay time constant is reported to be about 10–20 μs [14]–[16], although there are differences in each report. In this article, a cubic crystal with a size of 2 cm $\times$ 2 cm $\times$2 cm was used. This crystal was made by the Laboratory of Crystal Growth, Nikolaev Institute of Inorganic Chemistry, Russia, and cut by the Crystal Manufacturing Lab, Ltd., Russia. The crystal cut was made to match the shape of the unit cell as much as possible as follows: It has the same bottom surface as the unit cell and the side surfaces are parallel to the unit cell. As shown in Fig. 1, surfaces A, B, and C correspond to surfaces that are perpendicular to the *a*-, *b*-, and *c*-axes, respectively. Each backside is defined as A′, B′, and C′ surface. Surface B is perpendicular to the optical axis of the crystal (*b*-axis). In surfaces A and C, there is an angular deviation from the Miller index (100) and (001), respectively. However, the angular deviation is within 1° since the angle of β is 90.621° as mentioned in Table II. In addition, to check the structure of the crystal, pole figure measurements using the X-ray diffraction system (Bruker D8 Discover) from the Institute for Materials Research, Tohoku University, were performed. The crystal cut accuracy was confirmed within 1°.

B. Neutron Beam

To measure the anisotropic response to neutrons, we use a quasi-monochromatic neutron beam source whose central energy is 885 keV. This neutron beam was provided by a 4-MV Pelletron accelerator in the National Institute of Advanced Industrial Science and Technology (AIST) on March 26 and 27, 2019. The pulsed proton beam accelerated at 1.7 MV interacts with a tritium (T) target and generates neutrons in all directions via the T (p, n) reaction. Although monochromatic neutrons are generated by this reaction, the energy of the neutron becomes broaden with an energy width of about 885^{+10}_{-20} keV due to the thickness of the tritium target. The beam flux is ∼310 neutrons/cm^2/s at a distance of 100 cm. In this setup, a 2-cm cubic crystal was placed at a distance of 60 cm from the beam source, corresponding to ∼2644 neutrons/(2 cm)^2/s.

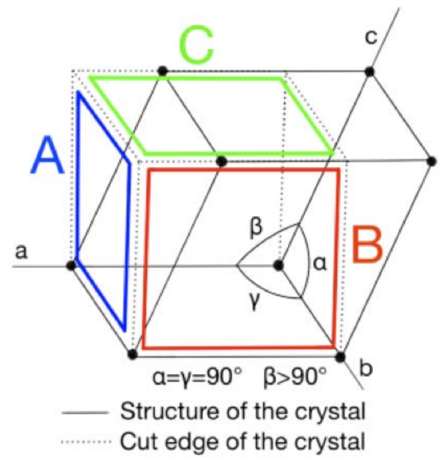

Fig. 1. Structure of a unit cell and the cut edge of $ZnWO_4$ crystals. Source: [17].

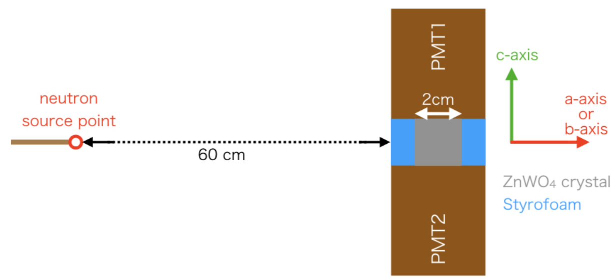

Fig. 2. Schematic view of the experimental setup.

C. Experimental Setup

Fig. 2 shows the schematic view of the experiment. Surfaces C and C′ were attached to two Hamamatsu H6411 photomultipliers (PMTs) with optical grease to focus on the anisotropic measurement of surfaces A and B. Teflon tape was wrapped around A, A′, B, and B′ surfaces to collect the scintillation photons effectively. In addition, styrofoam was used to hold the crystal. For events in which both PMTs had a signal exceeding a threshold, output signals from both PMTs were fed into a preamp with the charge integration time constant of 66 μs. Then, the preamplified signals were sent to the shaping amplifier and recorded by peak hold analog to digital converters (ADCs). The distance between the neutron beam source and the center of the crystal was 60 cm. In this measurement, neutron beam data were acquired in the order

TABLE III
GAMMA AND X-RAY CALIBRATION SOURCES AND NUMBERS OF DETECTED PHOTOELECTRONS

γ (X) source	Peak energy (keV)	Number of photoelectrons
^{109}Cd	22.1	21.0 ± 2.25
^{137}Cs	32.2	30.2 ± 3.41
^{241}Am	59.5	55.5 ± 5.46
^{133}Ba	81.0	74.6 ± 6.98

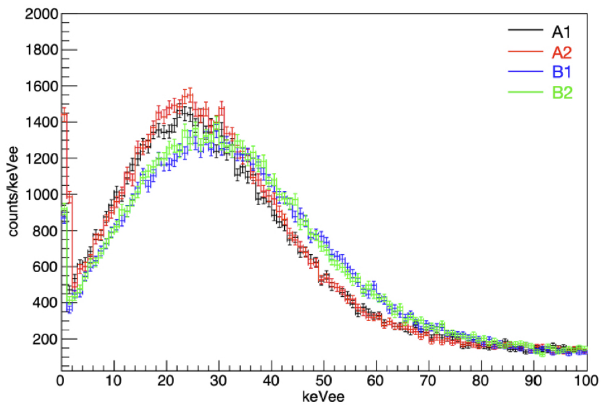

Fig. 3. Energy spectrum of each measurement. Black and red histograms are results for surface A irradiation, and blue and green histograms are for surface B irradiation. Source: [17].

of surfaces B, A, B, and A for the systematic check. Hereafter, each measurement is named as B1, A1, B2, and A2. Before starting the B1 measurement and between each measurement, energy calibration using the gamma and X-ray source of ^{109}Cd, ^{137}Cs, ^{241}Am, and ^{133}Ba was performed to check the scintillation response to gammas and to convert the value of the peak hold ADCs to electron equivalent energy (keVee). Table III summarizes the calibration source and numbers of detected photoelectrons. From this table, we confirmed that the scintillation response is linear to gammas, whose slope is about 0.94 pe/keV, in the 0–80 keV energy range. As for the neutron beam source, we took 150 000 events for each measurement. The response to the nuclear recoil is discussed in Section III.

D. Monte Carlo Simulation of Nuclear Recoils

The energy spectra obtained from the neutron beam run are superpositions of events scattered at any scattering angle in the crystal. To evaluate the quenching factor of the nuclear recoil, a Toy Monte Carlo (MC) simulation based on Geant4 [18] was performed. A toy MC consists of a 2-cm cubic $ZnWO_4$ Styrofoam mentioned in Section II-C. Neutrons that have the energy of 885^{+10}_{-20} keV are emitted isotopically 60 cm from $ZnWO_4$. From the deposited energy in a $ZnWO_4$ crystal, we confirmed the maximum energy deposit of oxygen, zinc, and tungsten nucleus recoil for 885 keV neutron, corresponding to a scattering angle of 180°, is 197, 59, and 22 keV, respectively.

III. RESULTS

Fig. 3 shows the energy spectrum obtained by four neutron beam measurements. The difference in light output between the irradiation to surface A and surface B is clearly observed. The quenching factor, the light yield ratio of nuclear recoils to electron recoils, is calculated by comparing the visible energy spectrum and the nuclear recoil spectrum obtained from the MC mentioned in Section II-D. Since the recoil energy dependence of the quenching factor is not known here, the quenching factor is derived from the ratio of the "edge" positions of the spectra of the MC simulation to that of the measurement result, by fitting their spectra with "an error function plus a linear function." From the fitting of the MC simulation, the edge position of the spectra is evaluated to be 200.9 ± 9.9 keV. Table IV summarizes the edge position of the spectra of each measurement with the obtained quenching factor. With respect to the systematic uncertainty for the quenching factor, the systematic uncertainty on the energy scale from the energy calibration shown in Table III, the uncertainty on the neutron energy from the experimental configurations corresponds to the angle error between the surface of the crystal and the beam (~2°) is considered in a quadratic way. As seen from Table IV, the obtained quenching factors of the ~200 keV oxygen nuclear recoil for the first (A1 and B1) and second beams (A2 and B2) are consistent. By averaging the result, the quenching factors of surface A (QF_A) and surface B (QF_B) are 0.199 ± 0.020 and 0.235 ± 0.026, respectively.

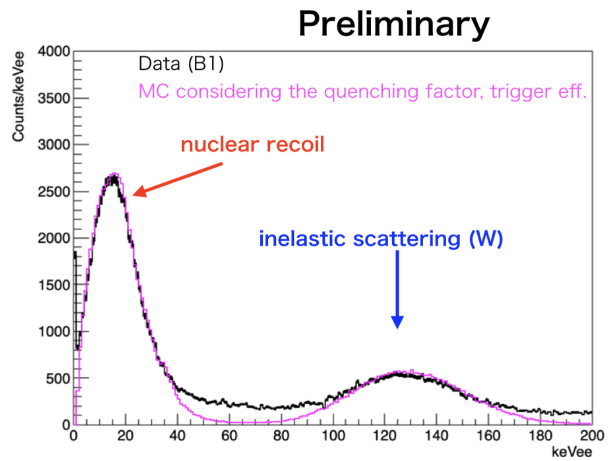

Fig. 4. Preliminary result of the comparison of the energy spectrum between data and MC simulation. As for data, neutron beams are irradiated to surface B. For MC simulation, the quenching factor of 0.235, the energy resolution, and the trigger efficiency evaluated by gamma calibration sources are considered. The lower energy peak corresponds to the elastic scattering with oxygen, and higher energy peak corresponds to the inelastic scattering with tungsten nuclei.

TABLE IV
EDGE POSITION OF THE MEASUREMENT RESULT OBTAINED BY THE FITTING WITH AN ERROR FUNCTION PLUS A LINEAR FUNCTION AND THE OBTAINED QUENCHING FACTOR FOR EACH BEAM MEASUREMENT

Beam	edge position (keVee)	quenching factor
A1	39.8 ± 3.4	0.198 ±0.020
A2	40.2 ± 3.2	0.200 ± 0.019
B1	47.6 ± 4.6	0.237 ± 0.026
B2	46.8 ± 4.7	0.233 ± 0.026

The anisotropy value between these two surfaces defined as $\chi = |QF_B - QF_A|/QF_B$ is evaluated to be 0.153. Fig. 4 shows the comparison between B1 data and MC simulation with the consideration of the quenching factor and trigger efficiency. This is the first observation of the anisotropic response of the $ZnWO_4$ crystal to nuclear recoil using the neutron beam.

IV. Conclusion

The measurement of the anisotropy of the $ZnWO_4$ crystal was conducted with a quasi-monochromatic neutron beam source whose energy is 885^{+10}_{-20} keV. Depending on the surface, which is perpendicular to the *a*- and *b*-axes of the crystal, the obtained quenching factor of the surface for about 200 keV oxygen nuclear recoil was 0.199 ± 0.020 and 0.235 ± 0.026, respectively. Accordingly, the scintillation yield of the crystal shows at least 15.3% of anisotropy depending on the incident direction of the neutrons, although this is a superposition of all scattering angle events similar to actual DM experiments. Cappella *et al.* [10] reported about 30% anisotropy of the scintillation light yield with 2–5-MeV alpha particles. The difference in the value of the anisotropy can be explained by the difference of the energy region and the difference of the incident particles. While the bulk effect of the crystal is observed with neutron sources, the surface effect of the crystal is observed with an alpha source; therefore, it is inferred that another effect was observed in Cappella *et al.* [10]. This is the first measurement of the anisotropic scintillation response of the $ZnWO_4$ crystal to neutrons, and the possibility of the $ZnWO_4$ crystal as a direction-sensitive DM detector was verified. In the assumption of this anisotropy, WIMP mass of 100 GeV/c^2, and 10^{-48} cm^2 WIMP-nucleon cross section, we can observe about 10% level of diurnal count rate variation in 4–6 keV energy range with 10^4 ton·days exposure [19]. The article including the detailed analysis for this measurement can be found in [17].

Acknowledgment

The authors would like to thank Dr. Tetsuro Matsumoto and Dr. Akihiko Masuda from the Radioactivity and Neutron Standards Group, Research Institute for Measurement and Analytical Instrumentation, National Meteorology Institute of Japan, National Institute of Advanced Industrial Science and Technology, for the accelerator operation.

References

[1] A. K. Drukier, "Freese and DN Spergel," *Phys. Rev. D, Part. Fields*, vol. 33, p. 3495, Jun. 1986.
[2] R. Bernabei *et al.*, "Blois 2019: Highlights and outlook," *Nucl. Phys. Atomic Energy*, vol. 19, no. 11, p. 116, 2018.
[3] J. B. R. Battat *et al.*, "Readout technologies for directional WIMP dark matter detection," *Phys. Rep.*, vol. 662, pp. 1–46, Nov. 2016.
[4] P. Belli, R. Bernabei, C. Bacci, A. Incicchitti and D. Prosperi, "Identifying a "dark matter" signal by nonisotropic scintillation detector," *Nuovo Cimento C*, vol. 15, no. 4, pp. 475–479, 1992.
[5] R. Bernabei, P. Belli, F. Nozzoli and A. Incicchitti, "New results from DAMA/LIBRA," *Eur. Phys. J.*, vol. 28, pp. 39–49, Mar. 2010.
[6] H. Sekiya, M. Minowa, Y. Shimizu, Y. Inoue and W. Suganuma, "Measurements of anisotropic scintillation efficiency for carbon recoils in a stilbene crystal for dark matter detection," *Phys. Lett. B*, vol. 571, pp. 132–138, Oct. 2003.
[7] J. Hisano, K. Ishiwata and N. Nagata, "A complete calculation for direct detection of Wino dark matter," *Phys. Lett. B*, vol. 690, pp. 311–315, Jun. 2010.
[8] A. S. Barabash *et al.*, "Improvement of radiopurity level of enriched $^{116}CdWO_4$ and $ZnWO_4$ crystal scintillators by recrystallization," *Nucl. Instrum. Methods Phys. Res. A, Accel. Spectrom. Detect. Assoc. Equip.*, vol. 833, p. 77, Jun. 2016.
[9] P. Belli *et al.*, "New development of radiopure $ZnWO_4$ crystal scintillators," *Nucl. Instrum. Methods Phys. Res. A, Accel. Spectrom. Detect. Assoc. Equip.*, vol. 935, p. 89, Jun. 2019.
[10] F. Cappella *et al.*, "On the potentiality if the $ZnWO_4$ anisotropic detectors to measure the directionality of Dark Matter," *Eur. Phys. J. C*, vol. 73, no. 1, p. 2276, 2013.
[11] D. Marco. (2019). *Talks at CYGNUS 2019 Workshop in Directional DM Detection*. [Online]. Available: http://people.roma2.infn.it/~dama/pdf/cygnus_2019.pdf
[12] P. Yadav, S. K. Rout, and E. Sinha, "Correlation between optical properties and environmental parameter of $ZnWO_4$ ceramic using complex chemical bond theory," *J. Alloys Compounds*, vol. 726, pp. 1014–1023, Dec. 2017.
[13] H. Sekiya *et al. Talks at CYGNUS 2019 Workshop in Directional DM Detection*. Accessed: Feb. 5, 2019. [Online]. Available: agenda.infn.it/event/18542/contributions/96204/attachments/65200/79265/Cygnus2019-Sekiya.pdf
[14] B. C. Grabmaier, "Crystal scintillators," *IEEE Trans. Nucl. Sci.*, vol. NS-31, no. 1, pp. 372–376, Feb. 1984.
[15] I. Holl, E. Lorenz, and G. Mageras, "A measurement of the light yield of common inorganic scintillators," *IEEE Trans. Nucl. Sci.*, vol. NS-35, no. 1, pp. 105–109, Feb. 1988.
[16] L. L. Nagornaya *et al.*, "Large volume $ZnWO_4$ crystal scintillators with excellent energy resolution and low background," *IEEE Trans. Nucl. Sci.*, vol. 56, no. 3, pp. 994–997, Jun. 2009.
[17] S. Ito *et al.*, "Development of a method for measuring rare Earth elements in the environment for future experiments with gadolinium loaded detectors," 2019, *arXiv:1905.10962*. [Online]. Available: http://arxiv.org/abs/1905.10962
[18] S. Agostinelli *et al.*, "GEANT4—A simulation toolkit," *Nucl. Instrum. Meth.*, vol. 506, p. 250, Jul. 2003.
[19] H. Sekiya. *Talks at CYGNUS 2015 Workshop in Directional DM Detection*. Accessed: Feb. 5, 2019. [Online]. Available: http://www.cygnus2015.com/wp-content/uploads/2015/06/Cygnus2015-Sekiya.pdf