Studies of a (Zn, Mg)WO\textsubscript{4} Crystal
Searching for the Origin of the Directional Dependence of the Scintillation Response

H. Sekiya
Kamioka Observatory, ICRR, University of Tokyo

S. Kurosawa*, A. Yoshikawa, T. Horiai, S. Kodama, R. Murakami, A. Yamaji
Research Lab. on Advanced Crystal Engineering, IMR, Tohoku University, *School of Science, Yamagata University

Y. H. Kim, Jin-A Jeon, H. L. Kim
Center for Underground Physics, Institute for Basic Science

June 14 2017 Xichang, Sichuan, China

CYGNUS 2017
ADAMO’s report

Directional response with MeV alpha particles

“Estimated” quenching factor @ 5keV

---

**Table 2** Quenching factors for O, Zn and W ions with energy 5 keV for different directions in ZnWO₄ crystal. Systematic uncertainties are estimated on the level of 20% using data of [90]

<table>
<thead>
<tr>
<th>Ion</th>
<th>Quenching factor</th>
<th>dir. 1</th>
<th>dir. 2</th>
<th>dir. 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>O</td>
<td>0.235</td>
<td>0.159</td>
<td>0.176</td>
<td></td>
</tr>
<tr>
<td>Zn</td>
<td>0.084</td>
<td>0.054</td>
<td>0.060</td>
<td></td>
</tr>
<tr>
<td>W</td>
<td>0.058</td>
<td>0.037</td>
<td>0.041</td>
<td></td>
</tr>
</tbody>
</table>
In case of stilbene crystal

P.H. Heckmann et al., Z. Phys. 162 (1961) 122

Directional response with MeV alpha particles

24% difference


Measured quenching factor of C-recoils.

only 7% difference
Requirement of exposure

- Assuming 7% anisotropy @5keVnr in ZnWO₄
- 100GeV, 10⁻⁴⁸cm² signal + atmospheric ν (& DSNB) BG

Expected "diurnal modulation"

- 10⁴ ton·days = 10 tons x 1000 days!
- Needs 10t crystal
Trying to get more information of WIMP direction

• So far, 10 tons are needed

• Phonon channel?
  – Total energy deposit vs direction-dependent light output makes the sensitivity better
  – Collaboration with AMoRE technologies!

  Jin-A’s talk
Review of the report in IDM2016 and a correction…

- 9mm x 9mm x 9mm crystal was made and tested.

<table>
<thead>
<tr>
<th>$a$ [Å]</th>
<th>$b$ [Å]</th>
<th>$c$ [Å]</th>
<th>$\alpha$ [deg]</th>
<th>$\beta$ [deg]</th>
<th>$\gamma$ [deg]</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.69060</td>
<td>5.71820</td>
<td>4.92690</td>
<td>90.0000</td>
<td>90.6210</td>
<td>90.0000</td>
</tr>
</tbody>
</table>

Monoclinic system


Czochralski process

Atuchin + CGD 2011

Approx. $a$ (001)

Approx. $c$ (100)
ERRATA $b$ and $c$ axes (Laue patterns) were swapped…

- Double checked with high precision X-ray diffraction devices
  - RIGAKU ATX-G 3D auto-scanning; This is the surface check!

![Diagram of X-ray diffraction setup]

Lattice face

Specific $\theta$ for each direction

CCD

X-ray

Crystal
The axes

- Cleavage plane ⊥ b

Approx. a (001)
Approx. c (100)

N.B. < 50 arcsec is good surface structure
No directional dependence of the transparency

- Transparency does not depend on the crystal orientation.

To measure:
- Crystal
- Reference

Spectrometer:
- V-550, JASCO

Halogen lamp:
- >340nm
- D2 lamp:
  - <340nm

Scintillation wavelength:
- ~a-axis
- b-axis
- ~c-axis

Scintillation intensity
- (a.u.)

Systematic error: <5%

Peak emission @480nm

Transmittance [%]

Wavelength [nm]

Hiroyuki Sekiya
CYGNUS 2017 Xichang, China June 14 2017
The result of the 9mm crystal @IDM2016

- Maximum along $b$-axis, $\sim a$ and $\sim c$ are same.
- 37% difference for 5.5MeV alpha, 32% for 59.5keV X-ray

![Graph showing counts vs MCA channel for 59.5keV X-ray and 5.5MeV alpha, with $\sim a$-axis, $\sim c$-axis, and b-axis marked.]

C.f. 2mm crystal @CYGNUS2015

~12% difference for 5.5MeV $\alpha$

~7% for 59.5keV X
Crystals from Institute for Single Crystal of National Academy of Sciences of Ukraine

• 1 x 1 x 1 cm$^3$
  – Black dots show the b-axis (cleavage) plane.
    » From Laue pattern
    » b-axis plane is clear,
    » Others were ambiguous

Hiroyuki Sekiya                                CYGNUS 2017    Xichang, China   June 14 2017
The response of the Ukraine crystal for $^{241}\text{Am}$ is shown in the diagram. There is a noticeable difference in the response when comparing the b-axis and c-axis for 5.5 MeV alpha particles, with a 38% difference. The small difference to X-rays is also indicated. The diagram includes points labeled A, B, C, D, E, and F, which may represent different measurements or data points.
Birefringence in Ukraine crystal

- The cut is based on optical axis.
  - i.e. not based on a-/b- axis.
ATX check

• The cut plane of the Ukraine crystal is 20° tilted to ours

No birefringence in our cut
The agenda in this time

- Response to electron recoil events
- The crystal structure and the anisotropic response
• In organic crystal case… two hypotheses
  – Anisotropic response exists, but non-straight path traveled by the electron washes out the effect.
  – Electrons deposit their energy with a much lower dE/dx than heavy charged particles, producing a lower excitation density. Changes in the excitation density due to directional transport may be on too small a scale compared to the overall density to affect the relative rates of kinetic processes for the electron recoil.
What is the origin of the anisotropic response?

- No anisotropy in muon events!
  - a high dE/dx is necessary for producing a directional dependence.
  - Physical mechanism is still unclear.
- In ZnWO$_4$, birefringence is observed (other than b-axis plane)
  - Should study the polarization and the scintillation.
  - Key to search for the best directional crystal.
More systematic measurements needed

• The way to check the systematics so far..
External gamma irradiation to see $\gamma/\alpha$, $\beta/\alpha$ ratio

- Calibration with $^{137}\text{Cs}(662\text{keV})$ $^{22}\text{Na}(511\text{keV},1275\text{keV})$
  - These cause electron multiple-scattering. Expected to be uniform response.
    - While many $^{241}\text{Am}$ 59.5keV events might be single photo-electric absorption.
    - $^{241}\text{Am}$ was also attached at the same time.

Ukraine crystal
Linarites for gammas looks OK

- In any directions.

![Graphs showing data for B face (b-axis) and E face (a'-axis).]
The spectra

$^{22}\text{Na}(511\text{keV, 1275keV})$

$^{241}\text{Am }\alpha$

$^{137}\text{Cs}(662\text{keV})$

$^{241}\text{Am }\alpha$
The ratio

<table>
<thead>
<tr>
<th></th>
<th>$\gamma(59.5\text{keV})/\alpha(5.5\text{MeV})$</th>
<th>$\beta\gamma(511\text{keV})/\alpha(5.5\text{MeV})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>B</td>
<td>0.0499 ± 0.0009</td>
<td>0.471 ± 0.001</td>
</tr>
<tr>
<td>~A 20° to c</td>
<td>0.0549 ± 0.0003</td>
<td>0.520 ± 0.001</td>
</tr>
<tr>
<td>~E 20° to a</td>
<td>0.0544 ± 0.0003</td>
<td>0.521 ± 0.001</td>
</tr>
</tbody>
</table>

- ~10% effects to both 59.5keV and to 511keV
- $^{241}\text{Am}$ only
The origin of the anisotropy... crystal structure itself

- The most asymmetric feature is the length of $b$

<table>
<thead>
<tr>
<th>$a$ [Å]</th>
<th>$b$ [Å]</th>
<th>$c$ [Å]</th>
<th>$\alpha$ [deg]</th>
<th>$\beta$ [deg]</th>
<th>$\gamma$ [deg]</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.69060</td>
<td>5.71820</td>
<td>4.92690</td>
<td>90.0000</td>
<td>90.6210</td>
<td>90.0000</td>
</tr>
</tbody>
</table>

- If $b$ is shorten, what can we see?

$\left( \text{Zn}_{0.95} \text{Mg}_{0.05} \right)_4 \text{WO}_4$

5mol% of Zn in ZnWO$_4$ replaced with Mg

Ion radius: 
- Mg$^{2+}$: 0.57 Å
- Zn$^{2+}$: 0.60 Å

Czochralski process

<table>
<thead>
<tr>
<th>Ingredients</th>
<th>MgCO$_3$, ZnO, WO$_3$ (purity 99.99%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pull-up speed</td>
<td>0.5mm/h</td>
</tr>
<tr>
<td>Direction of the seed-crystal</td>
<td>c-axis</td>
</tr>
<tr>
<td>Purge gas</td>
<td>Ar+O$_2$ (2%)</td>
</tr>
<tr>
<td>Rotation</td>
<td>12rpm</td>
</tr>
</tbody>
</table>
Bulk structure check with powder X-ray diffraction

- Monoclinic structure was confirmed.
- Slight right shift corresponds to the smaller $b$

Device: BURKER D8 DISCOVER
X-ray: Cu-Kα
40 kV, 40 mA
Lattice constants

<table>
<thead>
<tr>
<th></th>
<th>ZnWO$_4$ Schofield et al</th>
<th>ZnWO$_4$</th>
<th>(Zn, Mg)WO$_4$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$a$</td>
<td>4.69060</td>
<td>4.6937(15)</td>
<td>4.6929(10)</td>
</tr>
<tr>
<td>$b$</td>
<td>5.71820</td>
<td>5.7197(18)</td>
<td>5.7155(12)</td>
</tr>
<tr>
<td>$c$</td>
<td>4.92690</td>
<td>4.9275(16)</td>
<td>4.9303(11)</td>
</tr>
</tbody>
</table>

- Smaller $b$: smaller distortion of the lattice.

Laue pattern

$\beta \geq 90^\circ$

$\alpha, \gamma = 90^\circ$

$0.1\%$ shrinkage.. Significant?
γ/α irradiation check

LY is about 70% of ZnWO₄
The ratio

<table>
<thead>
<tr>
<th></th>
<th>$\gamma$(59.5keV)/$\alpha$(5.5MeV)</th>
<th>$\beta\gamma$(511keV)/$\alpha$(5.5MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>B (b-axis)</td>
<td>0.0502 ± 0.0011</td>
<td>0.522 ± 0.002</td>
</tr>
<tr>
<td>A (c-axis)</td>
<td>0.0482 ± 0.0039</td>
<td>0.534 ± 0.001</td>
</tr>
<tr>
<td>E (a-axis)</td>
<td>0.0492 ± 0.0069</td>
<td>0.535 ± 0.001</td>
</tr>
</tbody>
</table>

- 3~4% difference or agree within systematic error
- $^{241}$Am only
The ratio of ZnWO₄

<table>
<thead>
<tr>
<th></th>
<th>$\gamma(59.5\text{keV})/\alpha(5.5\text{MeV})$</th>
<th>$\beta\gamma(511\text{keV})/\alpha(5.5\text{MeV})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>B</td>
<td>0.0499 ± 0.0009</td>
<td>0.471 ± 0.001</td>
</tr>
<tr>
<td>~A 20° to c</td>
<td>0.0549 ± 0.0003</td>
<td>0.520 ± 0.001</td>
</tr>
<tr>
<td>~E 20° to a</td>
<td>0.0544 ± 0.0003</td>
<td>0.521 ± 0.001</td>
</tr>
</tbody>
</table>

- ~10% effects to both 59.5keV and to 511keV
- $^{241}\text{Am}$ only
Summary

• >10% anisotropic response to $\alpha$ particles was confirmed in ZnWO$_4$ crystal.
  – Along $b$-axis is the maximum, consistent with ADAMO’s report
  – Little or no anisotropic response to X-ray

• Zn$_{0.95}$Mg$_{0.5}$WO$_4$ crystal shows smaller anisotropic response for $\alpha$ particles.
  – The monoclinic structure and $b$-axis length (lattice distortion?)

• It is really bulk effect.
  – Neutron calibration is needed!
Plan

- Novosibirsk Crystals same as ADAMO are in preparation.
- $^{252}\text{Cf}$ run is in preparation.
- Response in 40mK will be reported in the next talk.

- 1inch crystal is also under preparation.
- Sensitivities in room temperature and in lower temperature with light and phonon signals will be investigated.
backups