This work is supported by JSPS KAKENHI
Grant Number 15K13478, 17H02884

Studies of a (Zn, Mg)WO_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

Eur. Phys. J. C (2013) 73:2276
DOI 10.1140/epjc/s10052-013-2276-2

THE EUROPEAN PHYSICAL JOURNAL C

Regular Article - Experimental Physics

On the potentiality of the $ZnWO_4$ anisotropic detectors to measure the directionality of Dark Matter

F. Cappella[1], R. Bernabei[2,3,a], P. Belli[3], V. Caracciolo[4], R. Cerulli[4], F.A. Danevich[5], A. d'Angelo[1,6], A. Di Marco[2,3], A. Incicchitti[6], D.V. Poda[5], V.I. Tretyak[5]

Directional response with MeV alpha particles

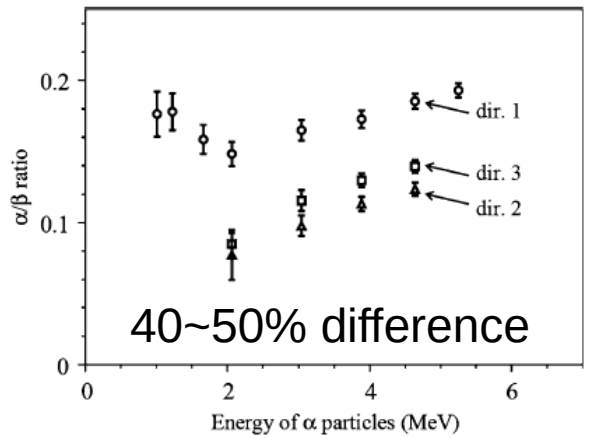


Fig. 3 Dependence of the α/β ratio on energy of α particles measured with $ZnWO_4$ scintillator. The crystal was irradiated in the directions perpendicular to (010), (001) and (100) crystal planes (directions 1, 2 and 3, respectively). The anisotropic behaviour of the crystal is evident [99]

"Estimated" quenching factor @ 5keV

Table 2 Quenching factors for O, Zn and W ions with energy 5 keV for different directions in $ZnWO_4$ crystal. Systematic uncertainties are estimated on the level of 20 % using data of [90]

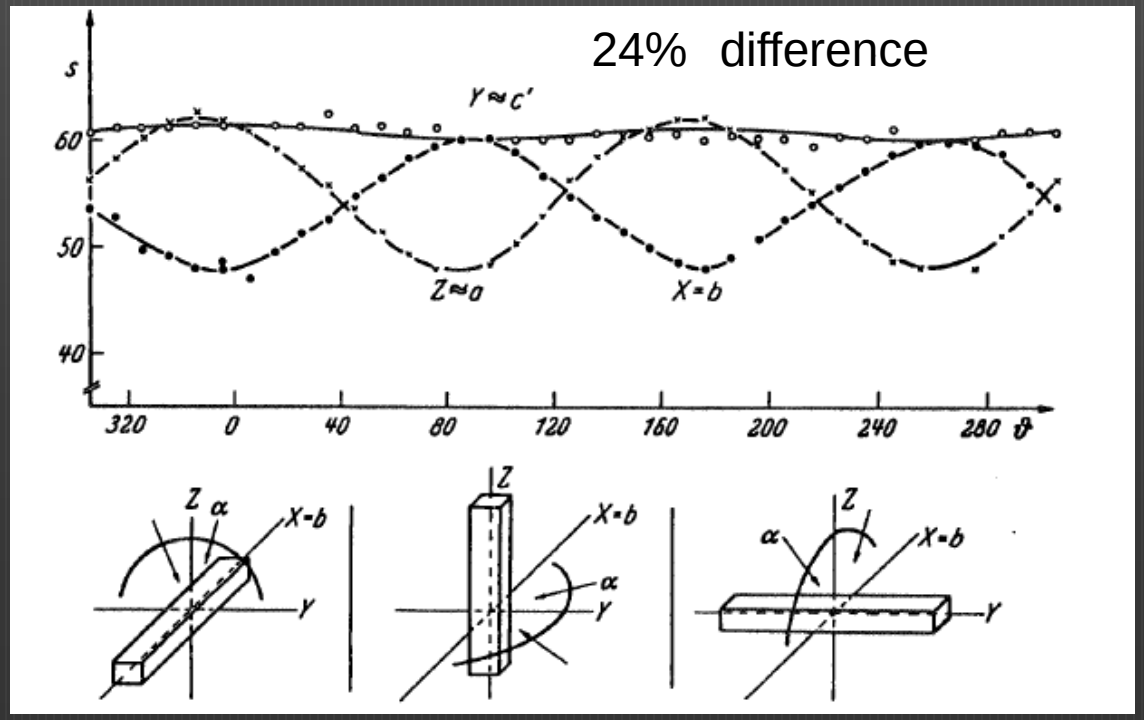
~30% difference

Ion	Quenching factor		
	dir. 1	dir. 2	dir. 3
O	0.235	0.159	0.176
Zn	0.084	0.054	0.060
W	0.058	0.037	0.041

In case of stilbene crystal

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

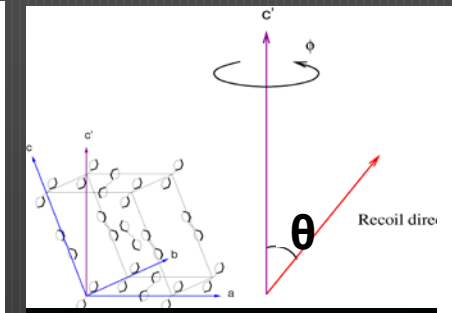
Directional response
with MeV alpha particles

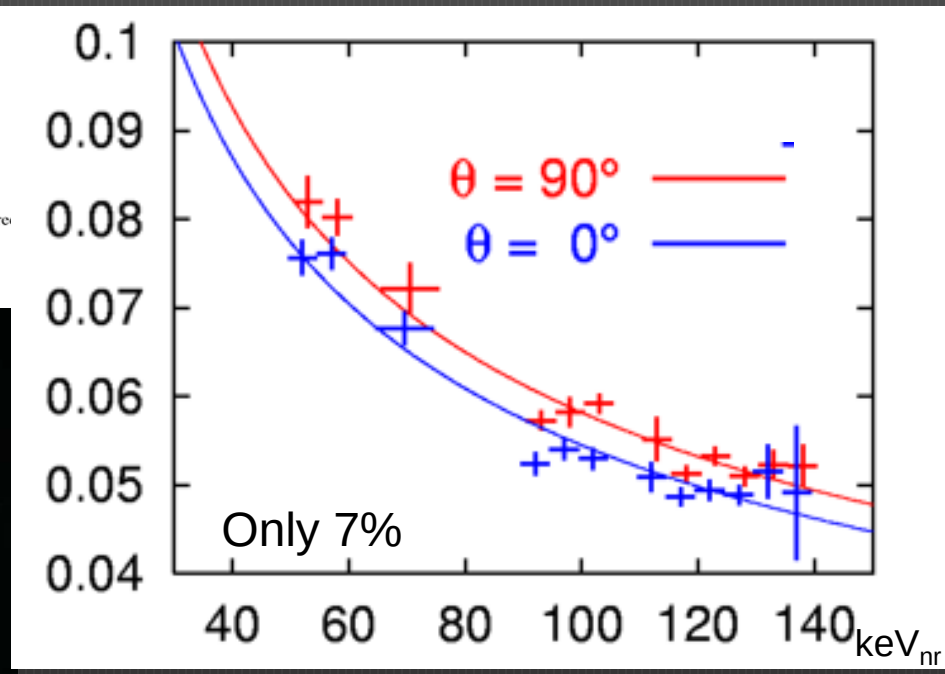


HS et al., Physics Letters B 571 (2003) 132+ IDM 2004

Measured quenching factor of C-recoils.

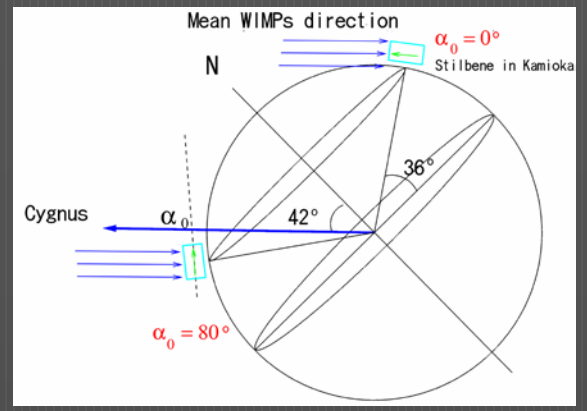
only 7 % difference

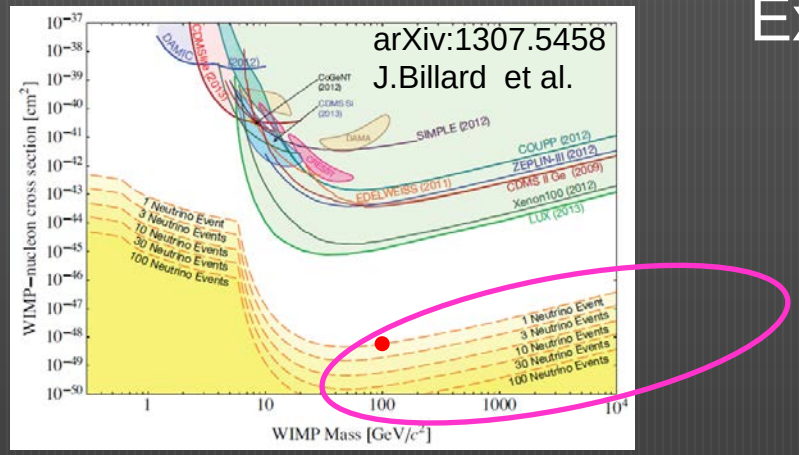




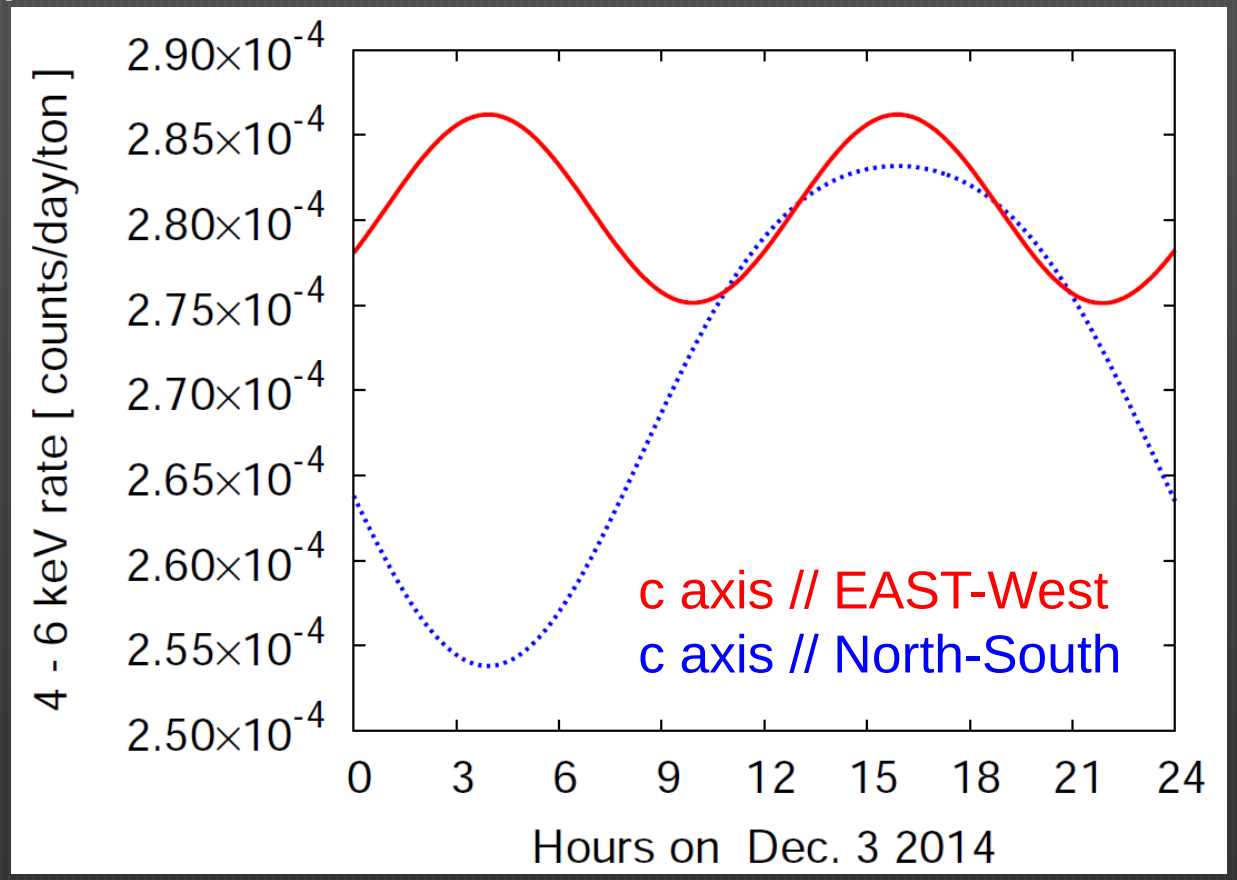
Requirement of exposure

- Assuming 7% anisotropy @5keVnr in $ZnWO_4$
- 100GeV, $10^{-48}cm^2$ signal + atmospheric ν (& DSNB) BG





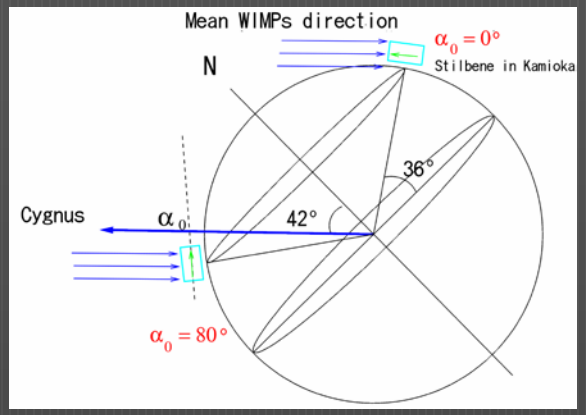
Expected "diurnal modulation"

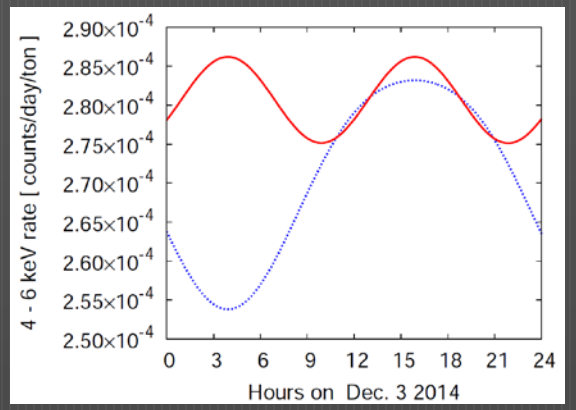


- **10^4 ton·days**
  =10 tons x 1000 days !
- **Needs 10t crystal**

Trying to get more information of WIMP direction

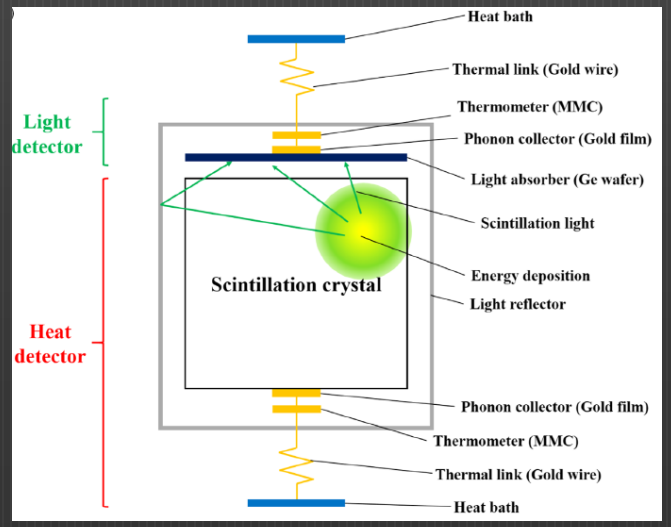
- So far, 10 tons are needed





- 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…

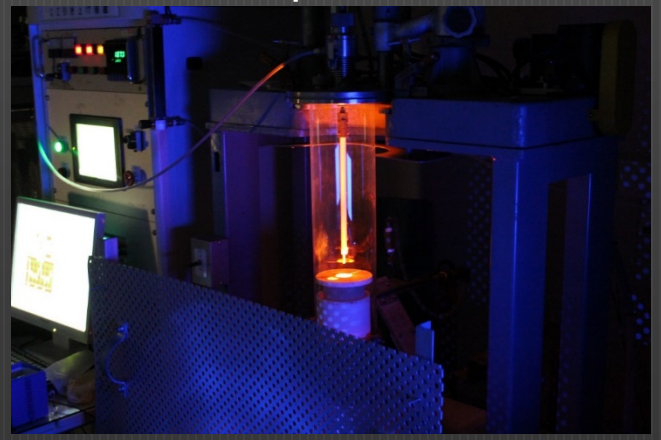
- 9mm x 9mm x 9mm crystal was made and tested.

a [Å]	b[Å]	c[Å]	α [deg]	β [deg]	γ [deg]
4.69060	5.71820	4.92690	90.0000	90.6210	90.0000

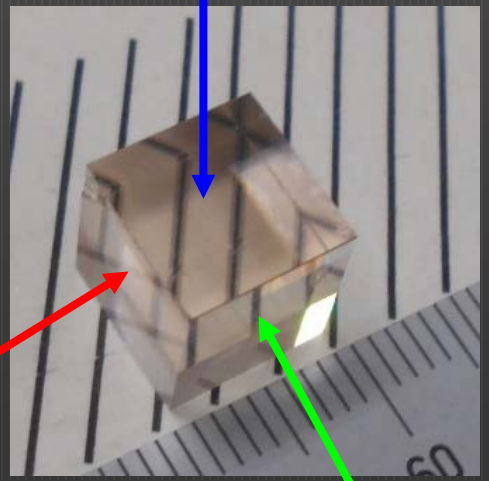
Monoclinic system

Schofield et al., Acta Crystallographica, Section B: Structural Science (1997) 53, p102-p112

Czochralski process

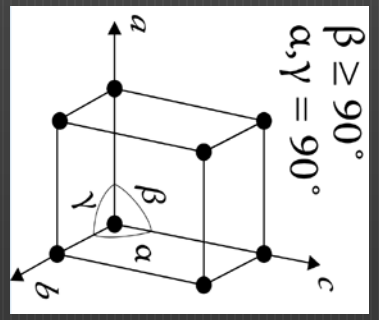


b (010)

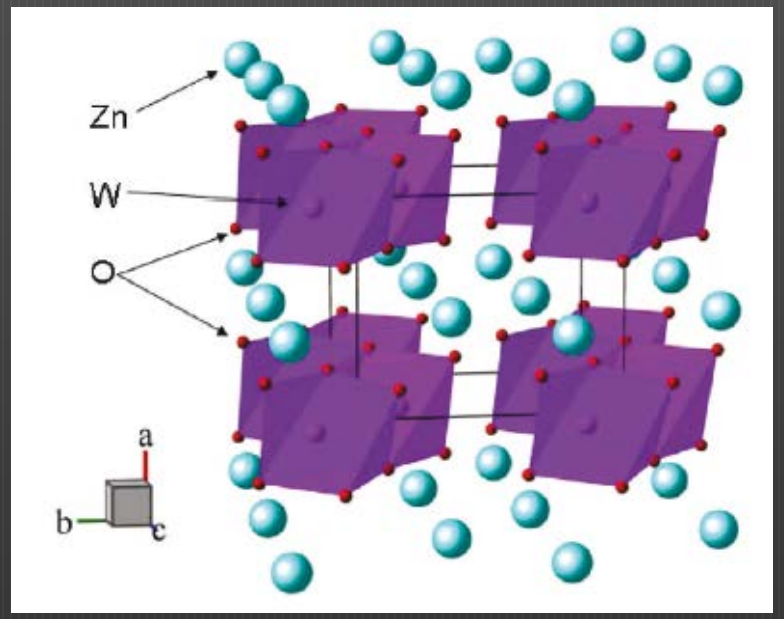


Approx. a (001)

Approx. c (100)

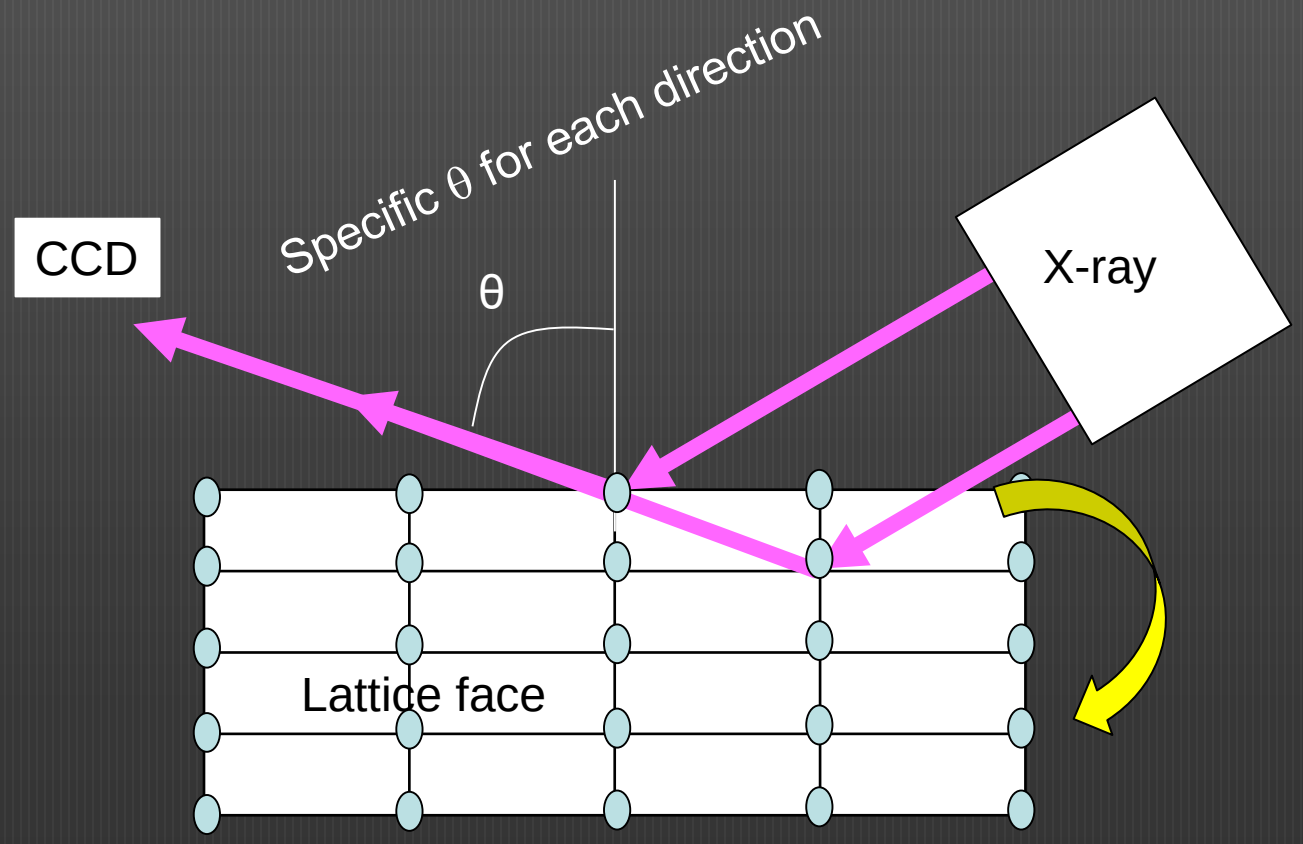


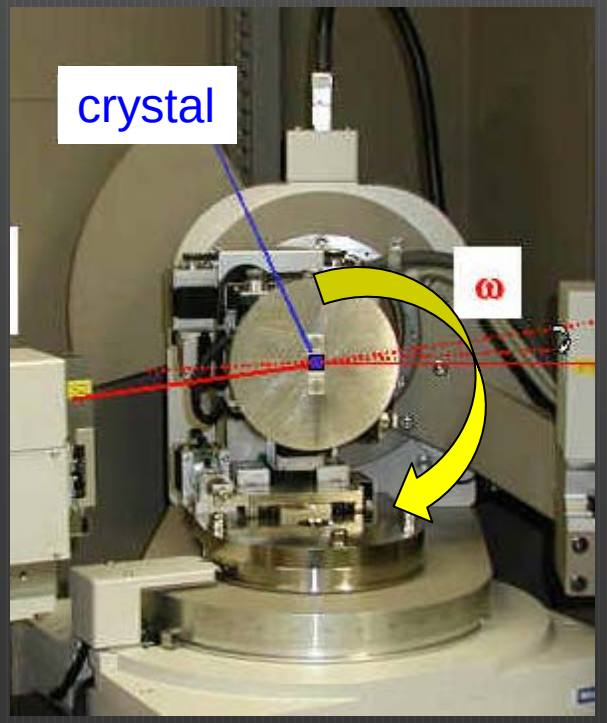
Atuchin + CGD 2011



ERRATA *b* and *c* axes (Laue patterns) were swapped…

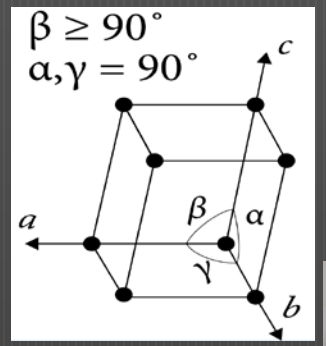
- Double checked with high precision X-ray diffraction devices
  - RIGAKU ATX-G 3D auto-scanning; This is the **surface check!**

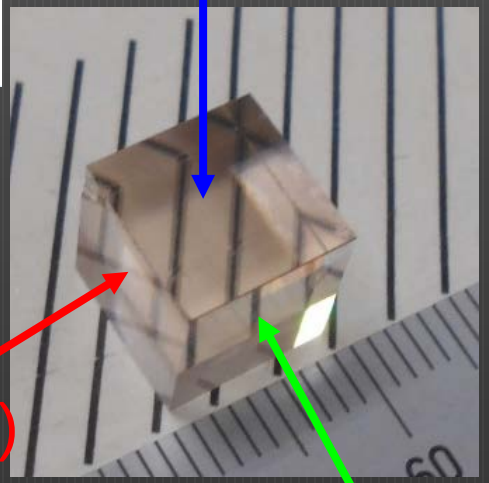


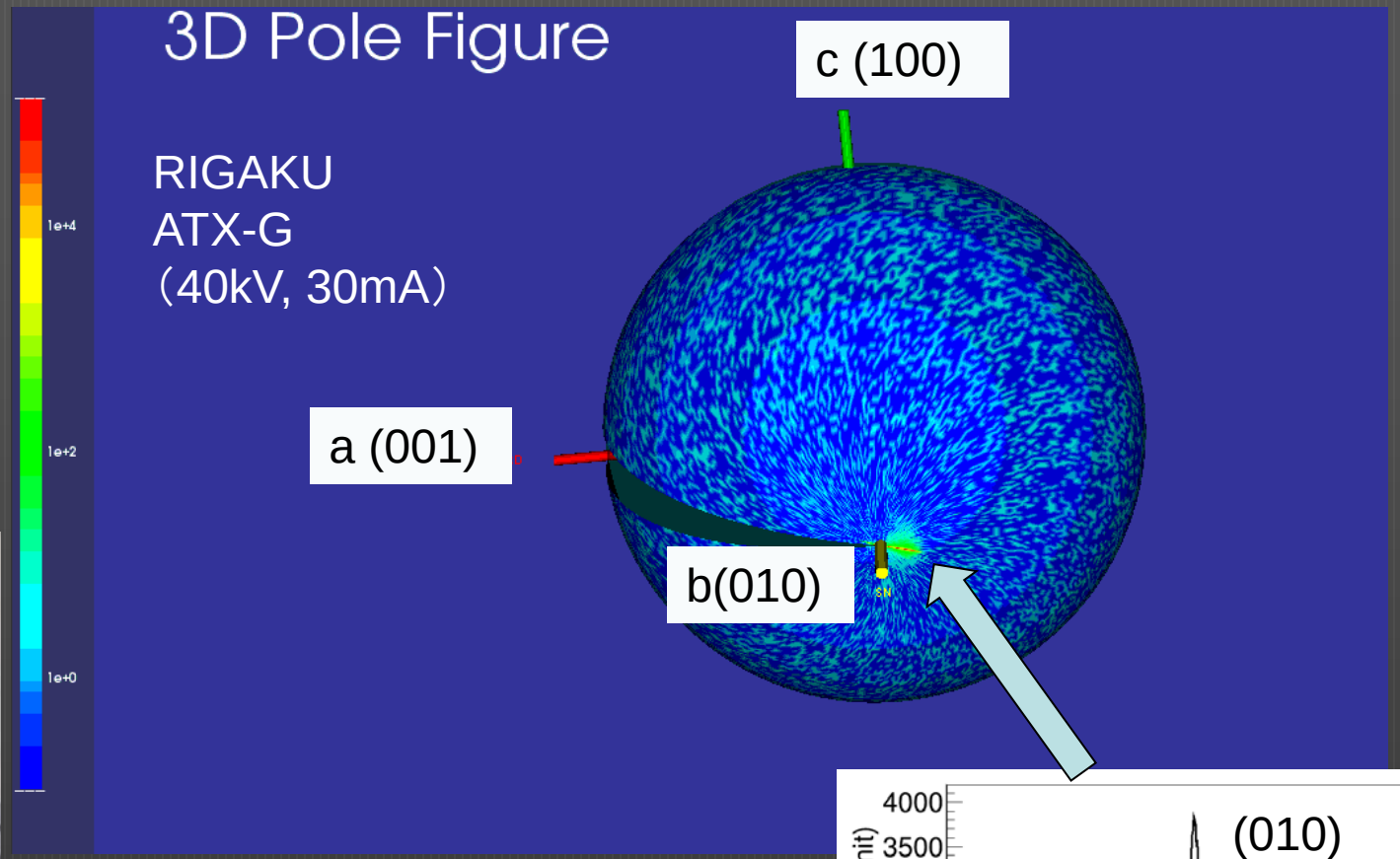


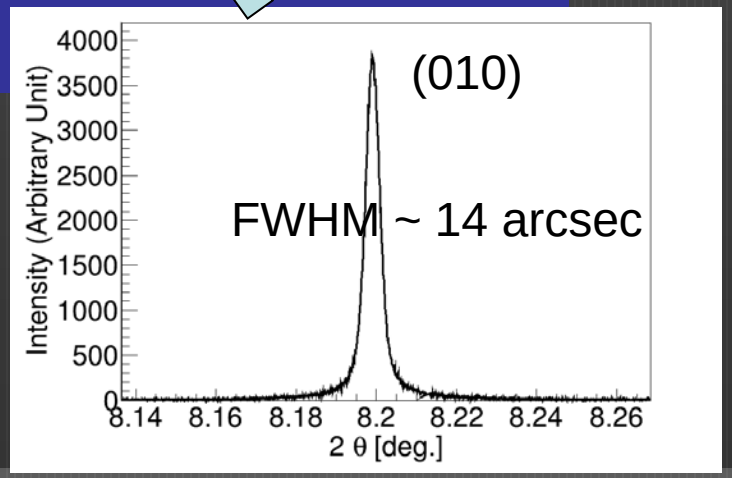
The axes

- Cleavage plane ⊥ b



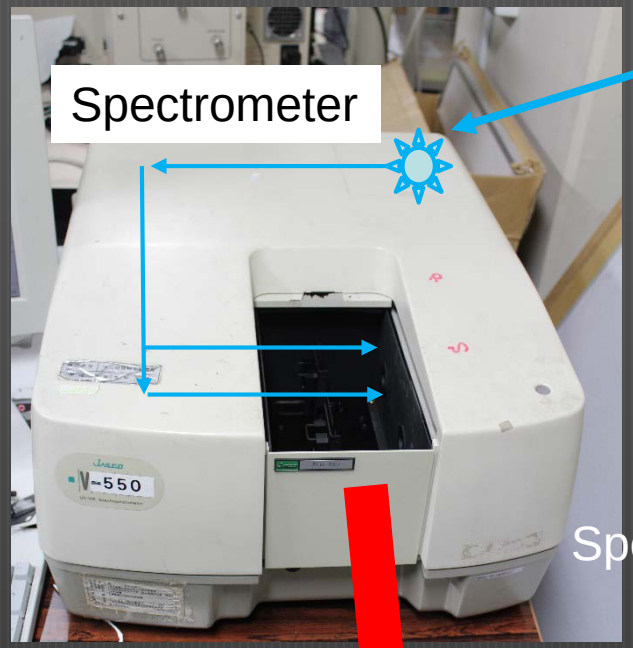






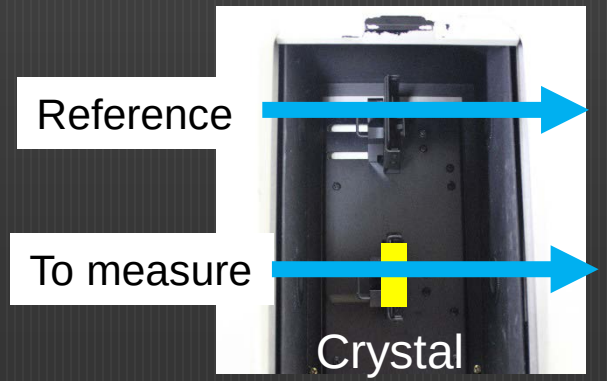
N.B. < 50 arcsec is good surface structure

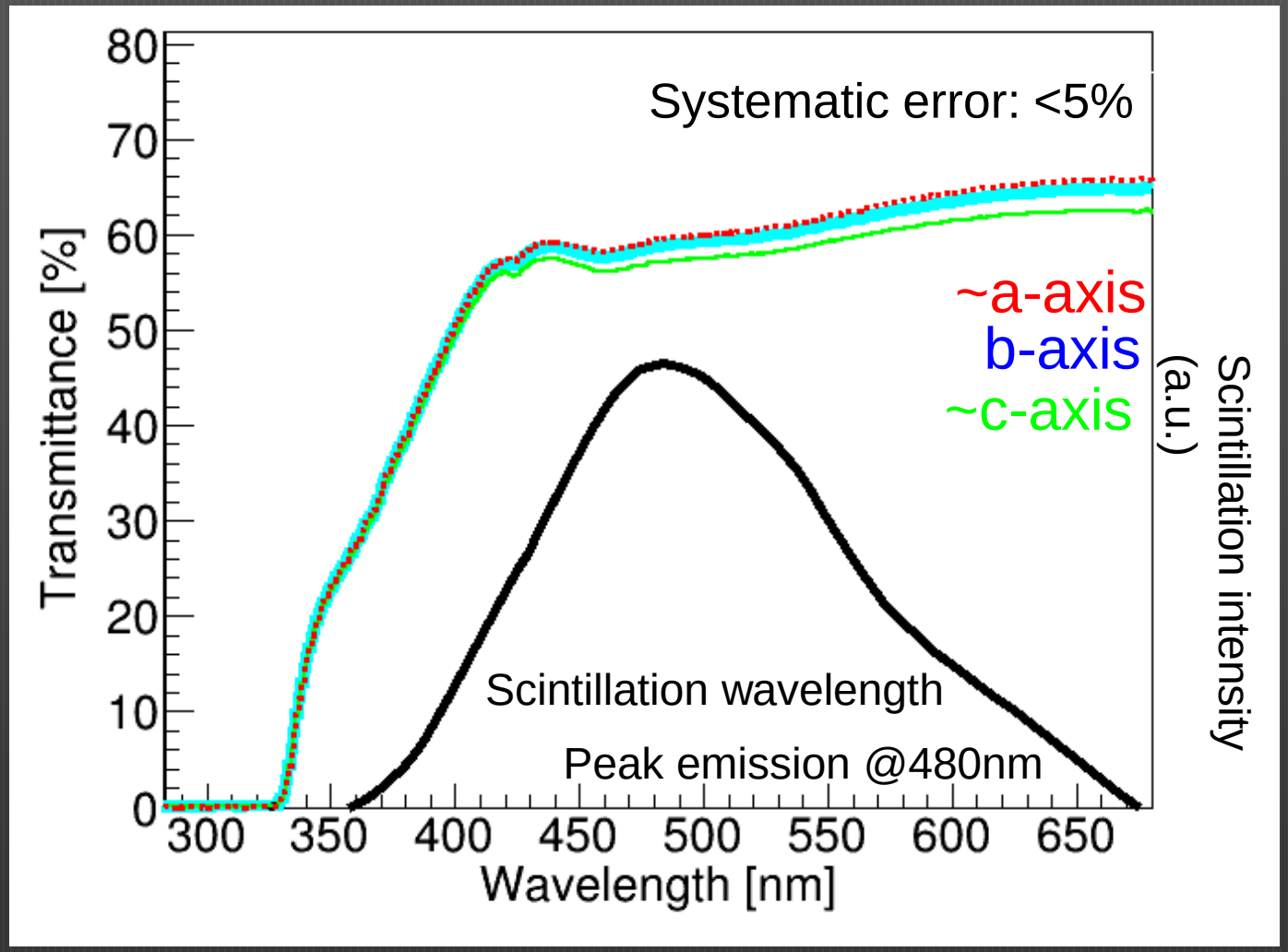
No directional dependence of the transparency



Halogen lamp
>340nm
D2 lamp
<340nm

Spectrometer
V-550, JASCO

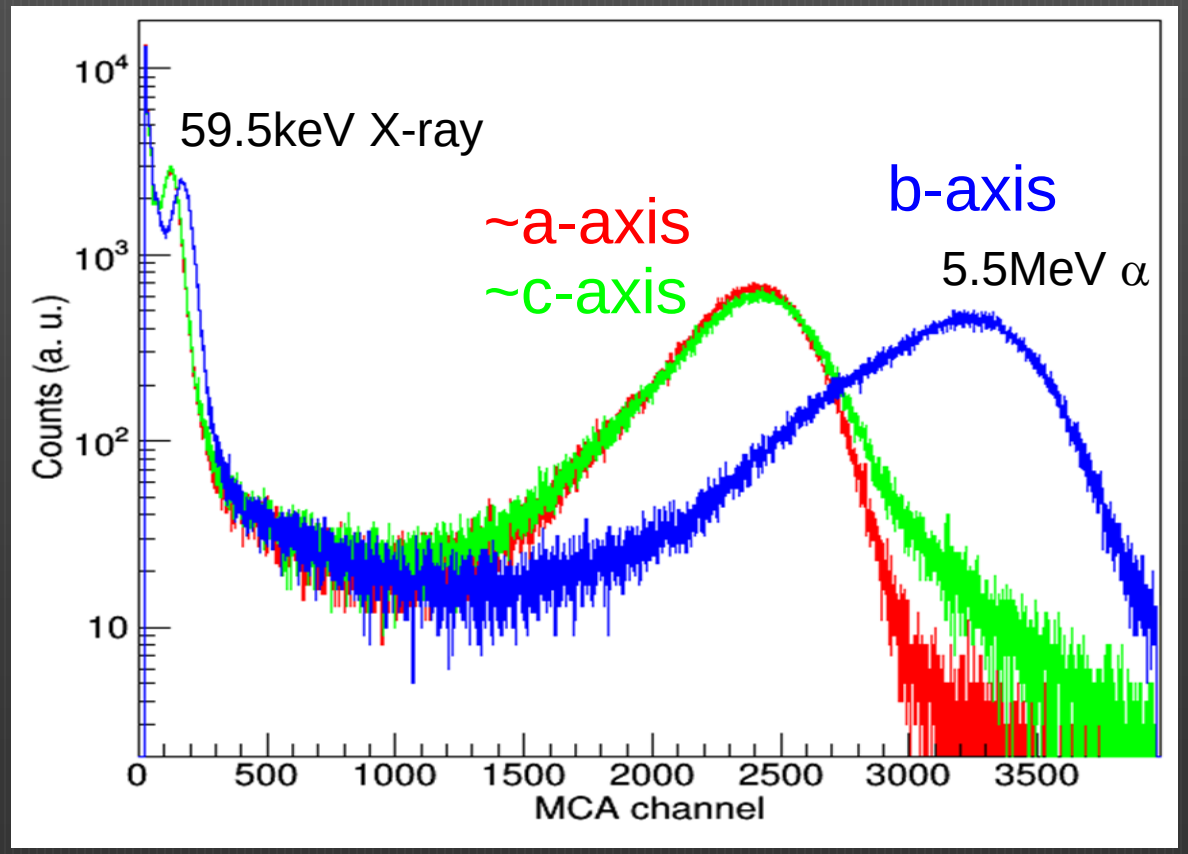




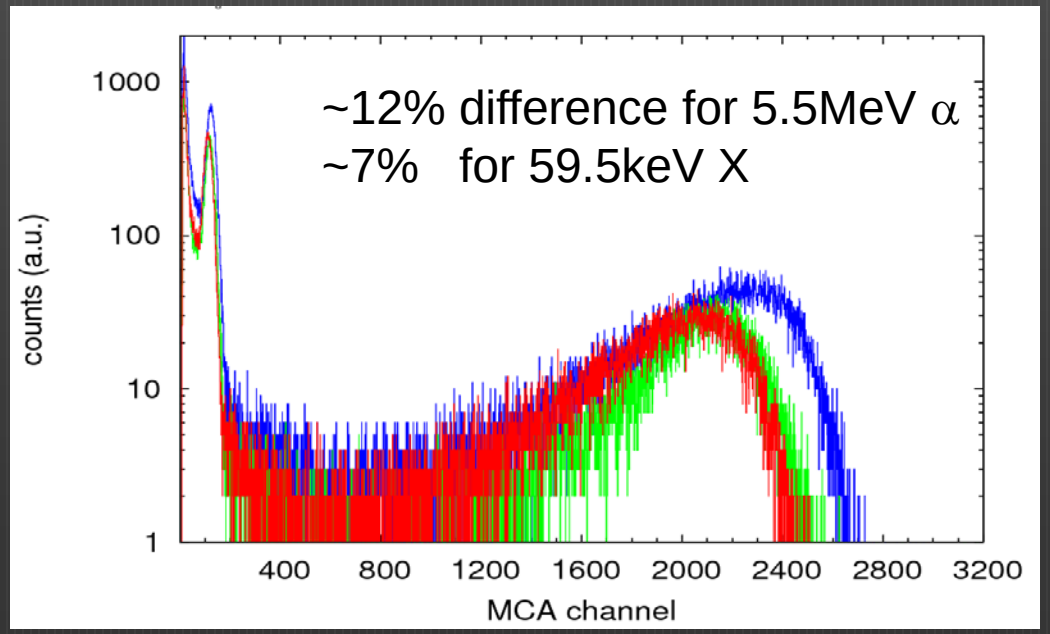
- Transparency does not depend on the crystal orientation.

The result of the 9mm crystal @IDM2016

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

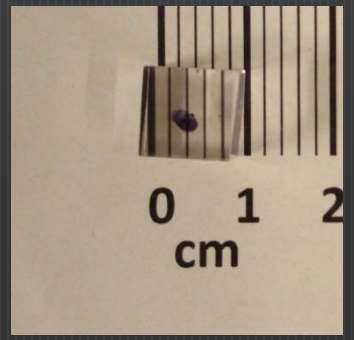


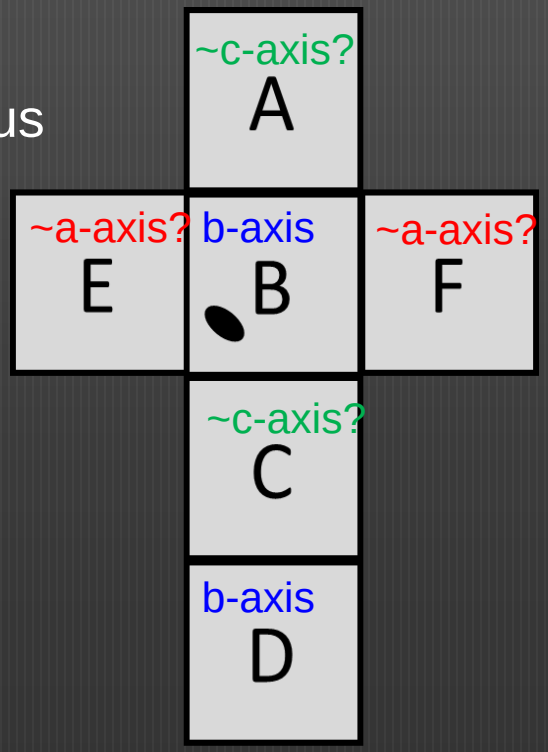
c.f. 2mm crystal @CYGNUS2015

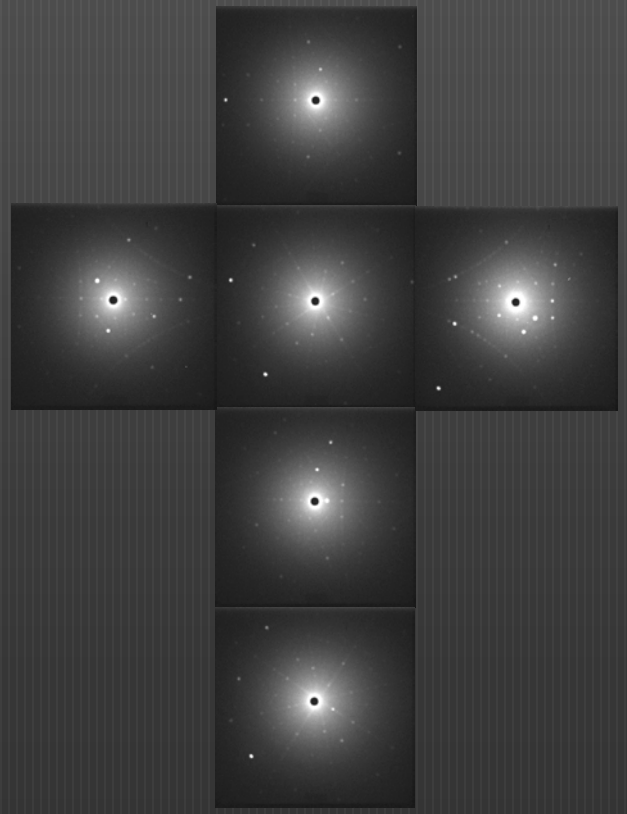


Crystals from Institute for Single Crystal of National Academy of Sciences of Ukraine

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

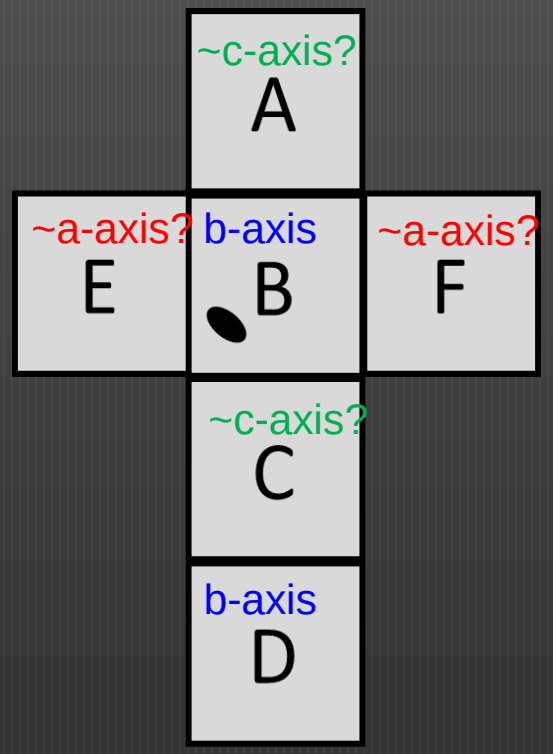


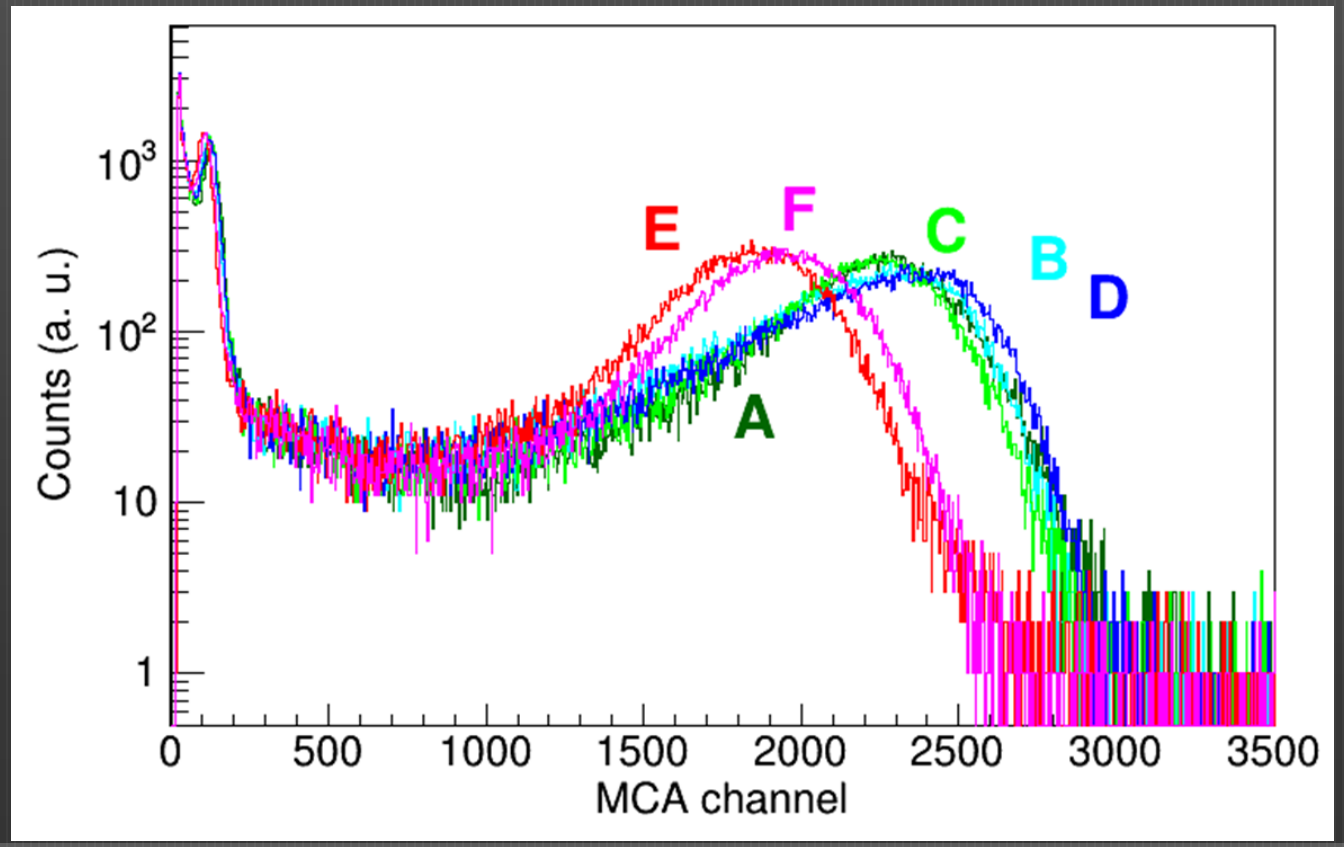




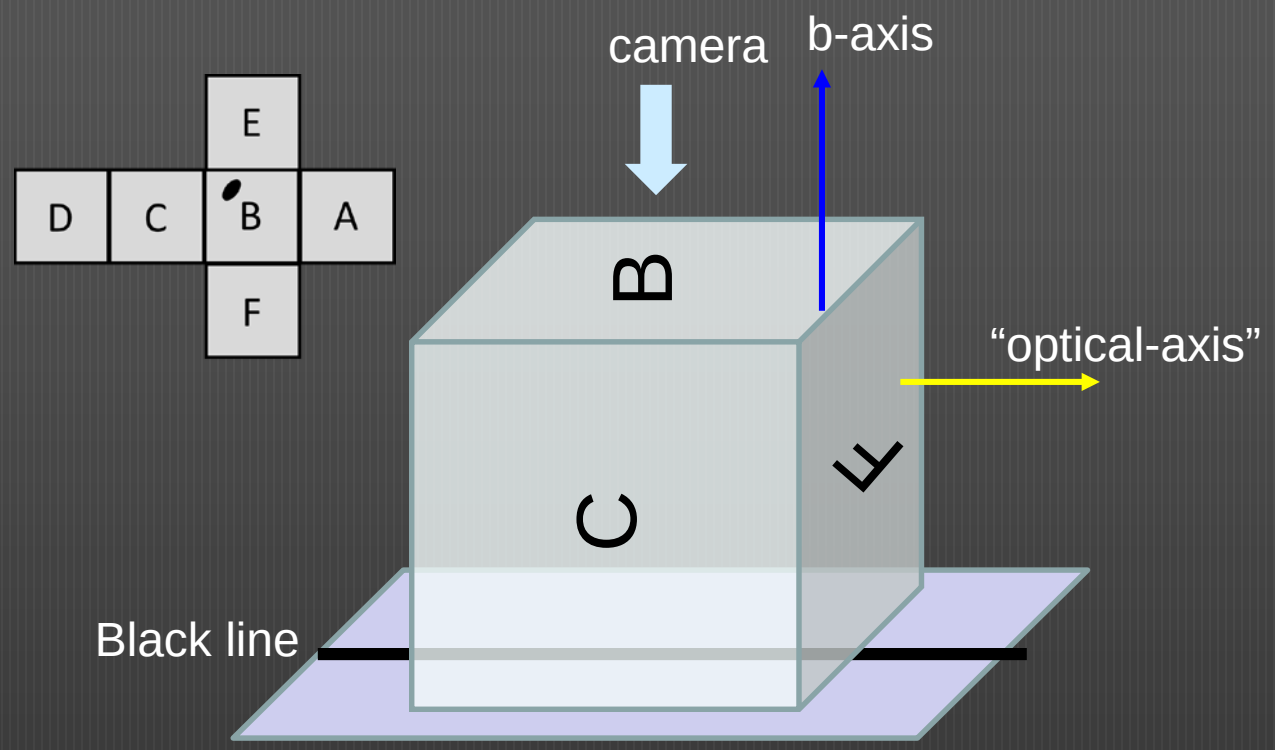
^{241}Am response of the Ukraine crystal @IDM2016

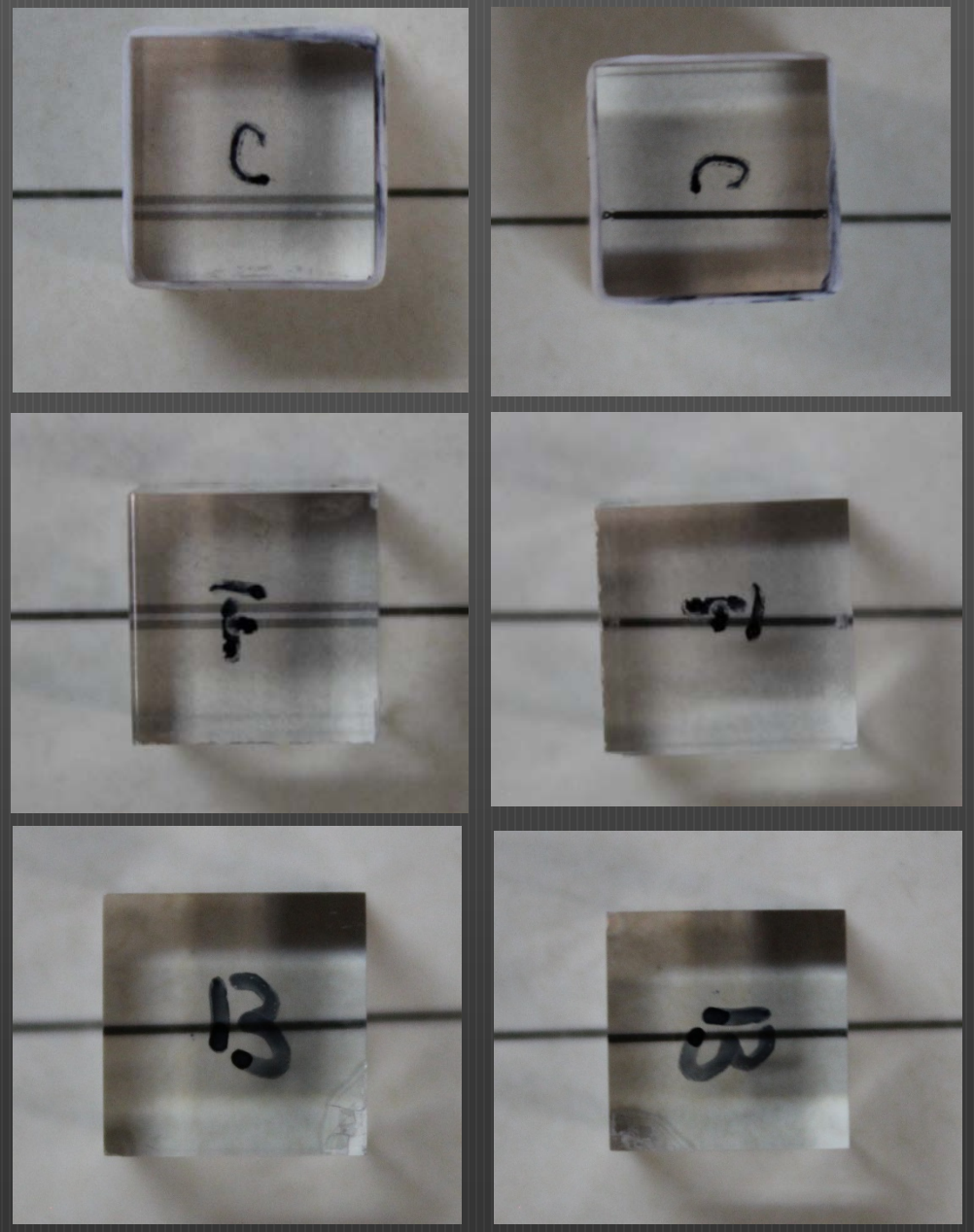
- b-zxis : ~c-axis? 38% difference for 5.5MeV alpha.
- Small difference to X-rays.





Birefringence in Ukraine crystal



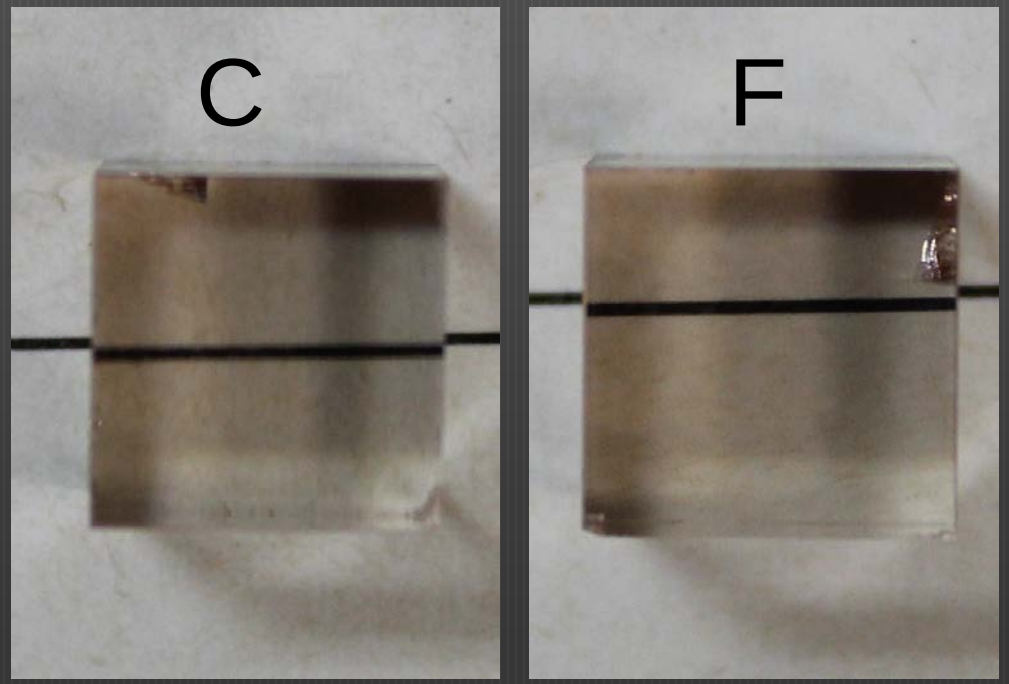


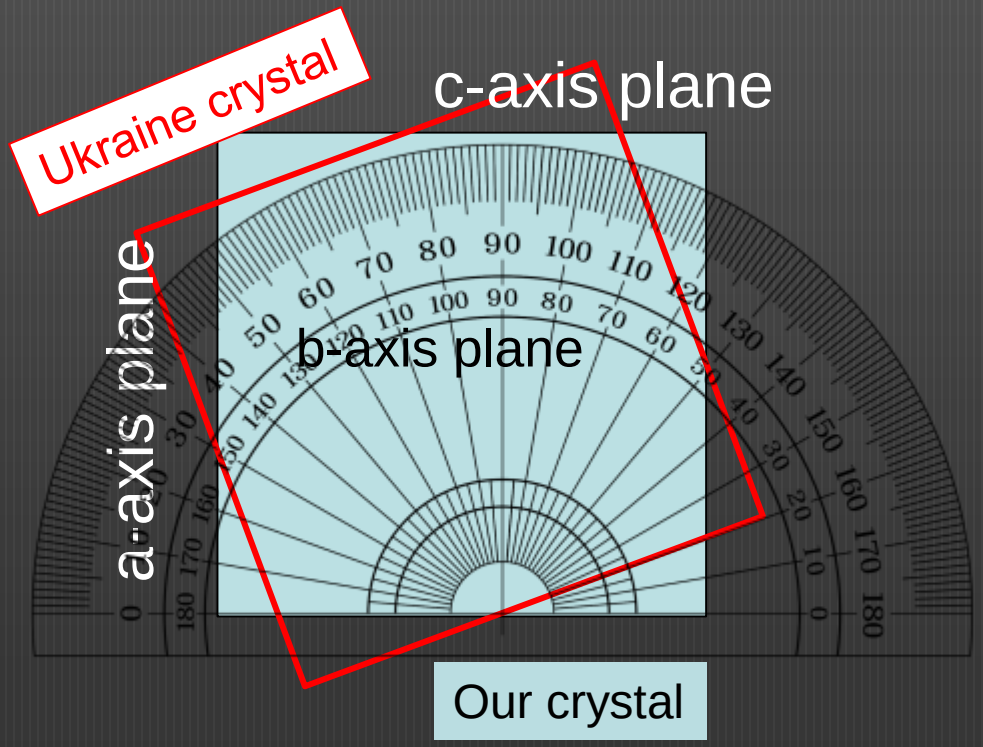
- The cut is based on optical axis.
  - i.e. not based on a-/b- axis.

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

No directional dependence in electron recoil events

NUCLEAR INSTRUMENTS AND METHODS 121 (1974) 69–76; © NORTH-HOLLAND PUBLISHING CO.

DIRECTIONAL ANISOTROPY IN ORGANIC SCINTILLATION CRYSTALS

F. D. BROOKS and D. T. L. JONES*

Physics Department, University of Cape Town, Rondebosch, C.P., South Africa

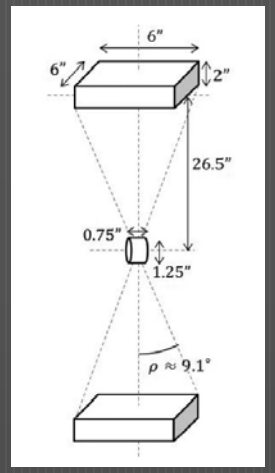
- In organic crystal case… two hypotheses
  - Anisotropic response exists, but non-straight path traveled by the electron washes out the effect.
  - Electrons deposit their energy with a much lower dE/dx than heavy charged particles, producing a lower excitation density. Changes in the excitation density 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?

1942 IEEE TRANSACTIONS ON NUCLEAR SCIENCE, VOL. 63, NO. 3, JUNE 2016

Investigating the Anisotropic Scintillation Response in Anthracene through Neutron, Gamma-Ray, and Muon Measurements

Patricia Schuster, *Member, IEEE*, and Erik Brubaker, *Member, IEEE*

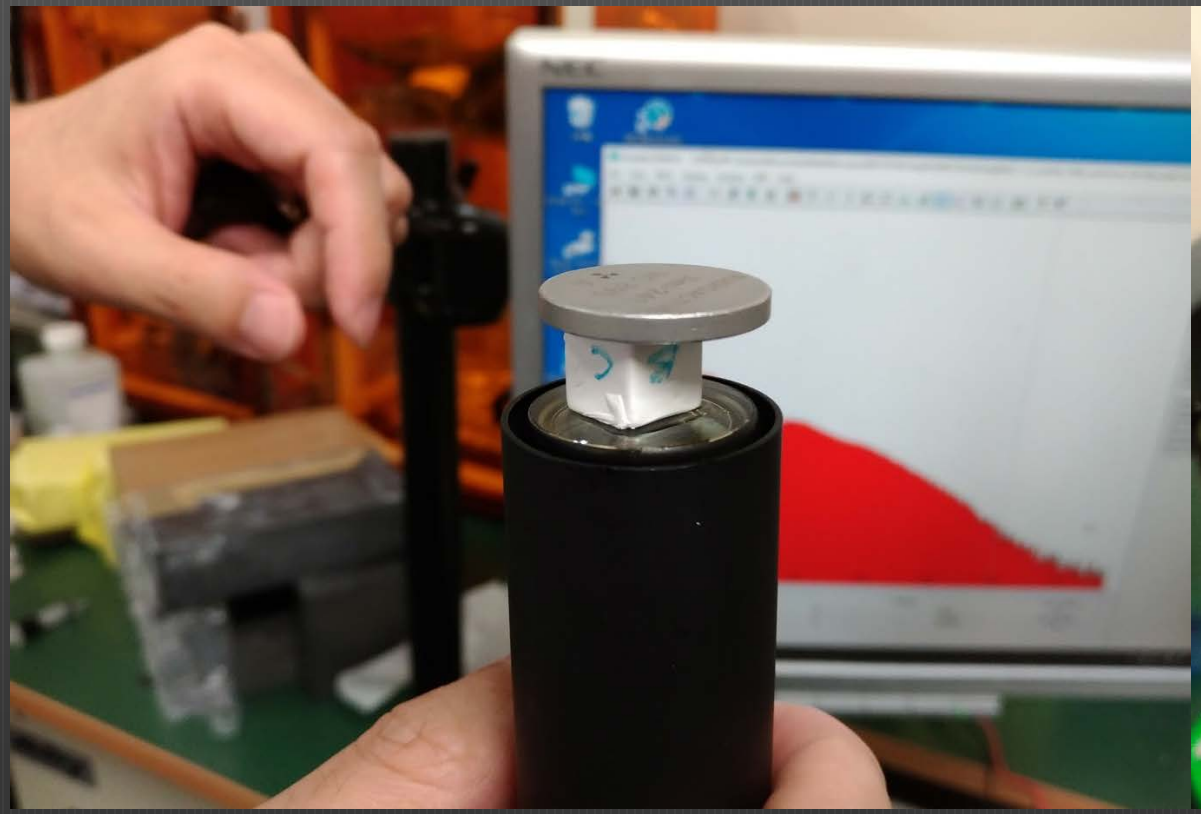


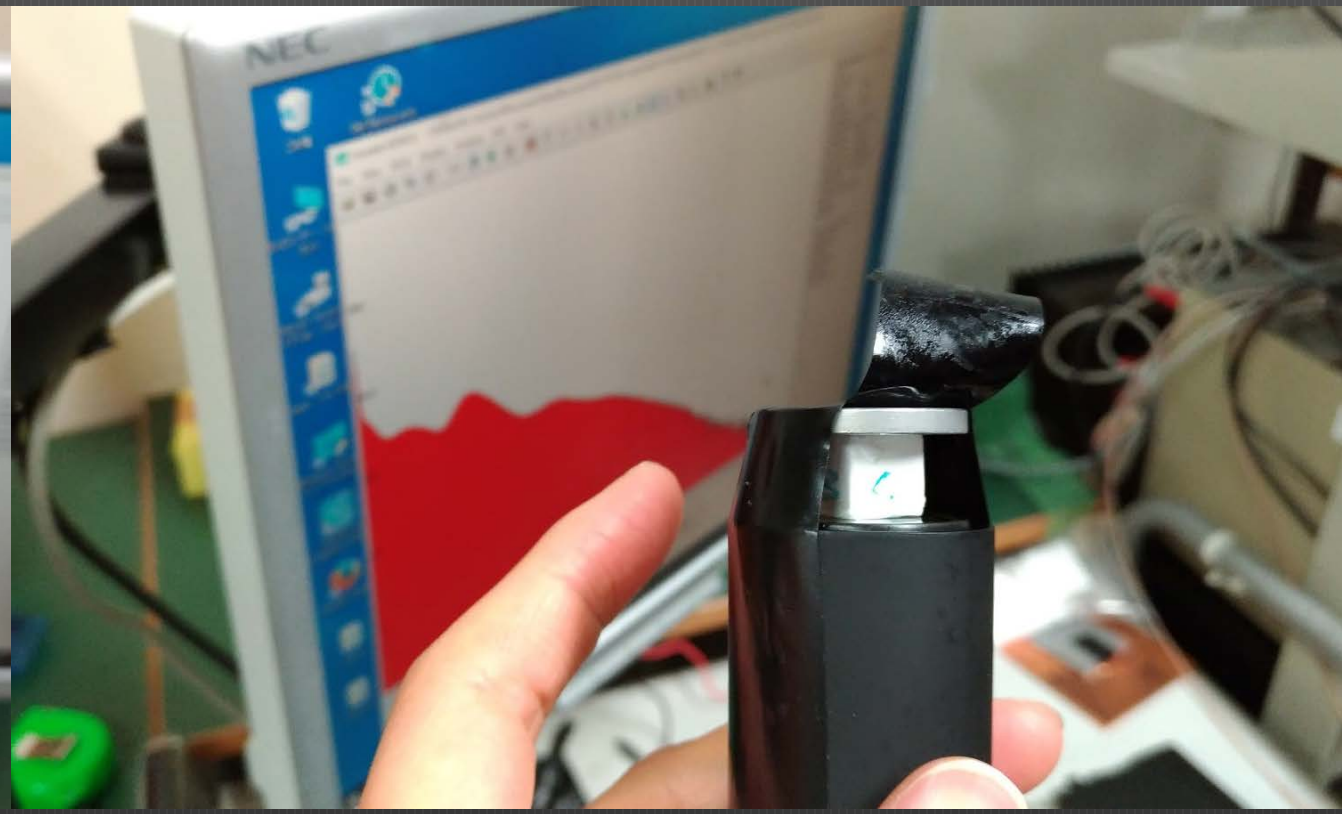
Source Particle	Neutron	Neutron	Gamma ray	Muon
Recoil Particle	Proton	Proton	Electron	–
Path	Straight	Straight	Non-straight	Straight
Energy (MeV)	14	2.5	0.478	4000
dE/dx (MeV/cm)	42.9	166.7	2.5	2.4
$\hat{L}$ mag. change	1.060±0.005	1.108±0.012	1.005±0.004	1.005±0.009
$\hat{S}$ mag. change	1.062±0.002	1.022±0.007	1.006±0.017	1.004±0.026
Anisotropy Observed	Yes	Yes	No	No

- 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

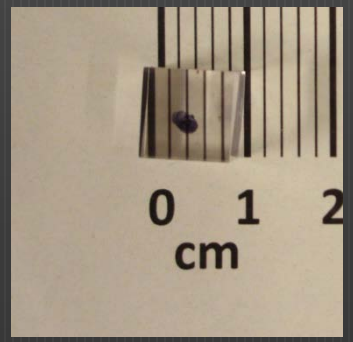
- The way to check the systematics so far..





External gamma irradiation to see γ/α, β/α ratio

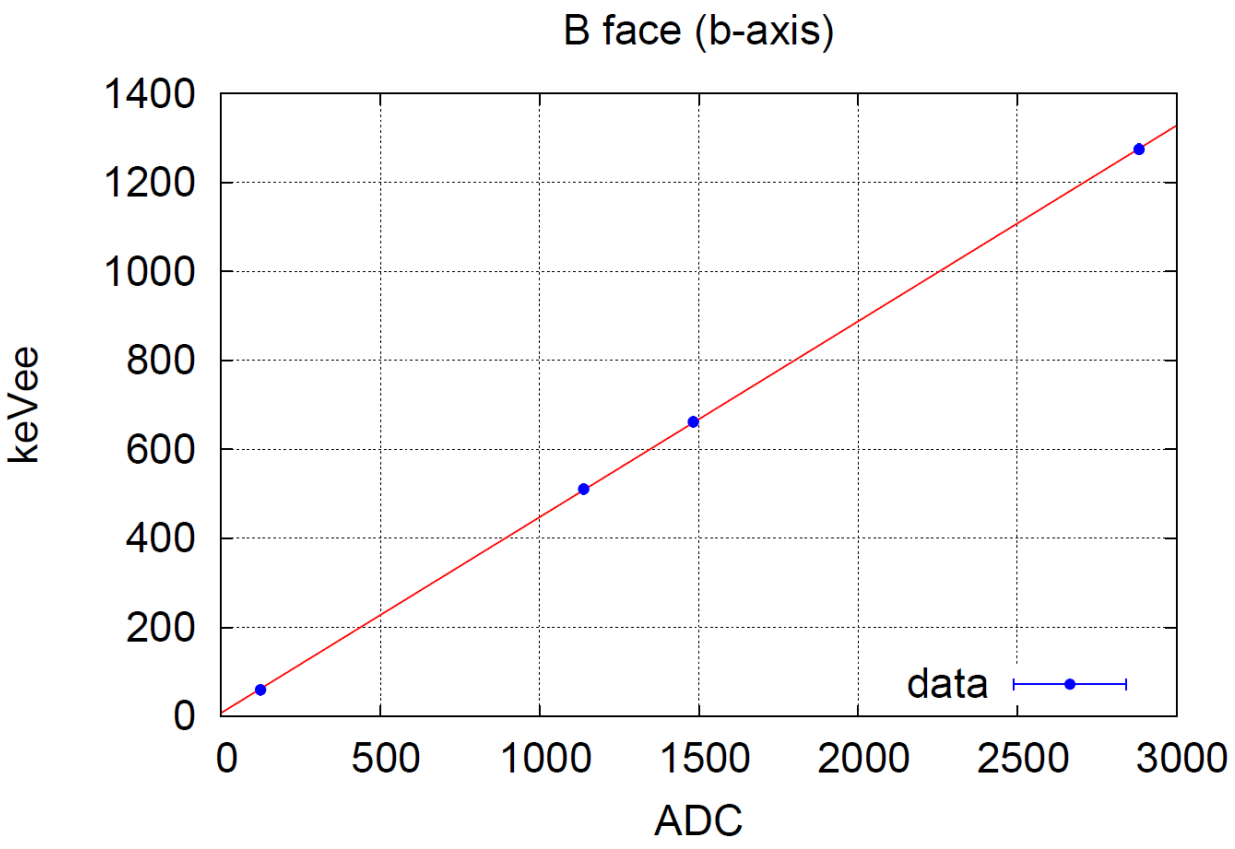
- Calibration with ^{137}Cs(662keV) ^{22}Na(511keV,1275keV)
  - These cause electron multiple-scattering. Expected to be uniform response.
    - While many ^{241}Am 59.5keV events might be single photo-electric absorption.
    - ^{241}Am was also attached at the same time.

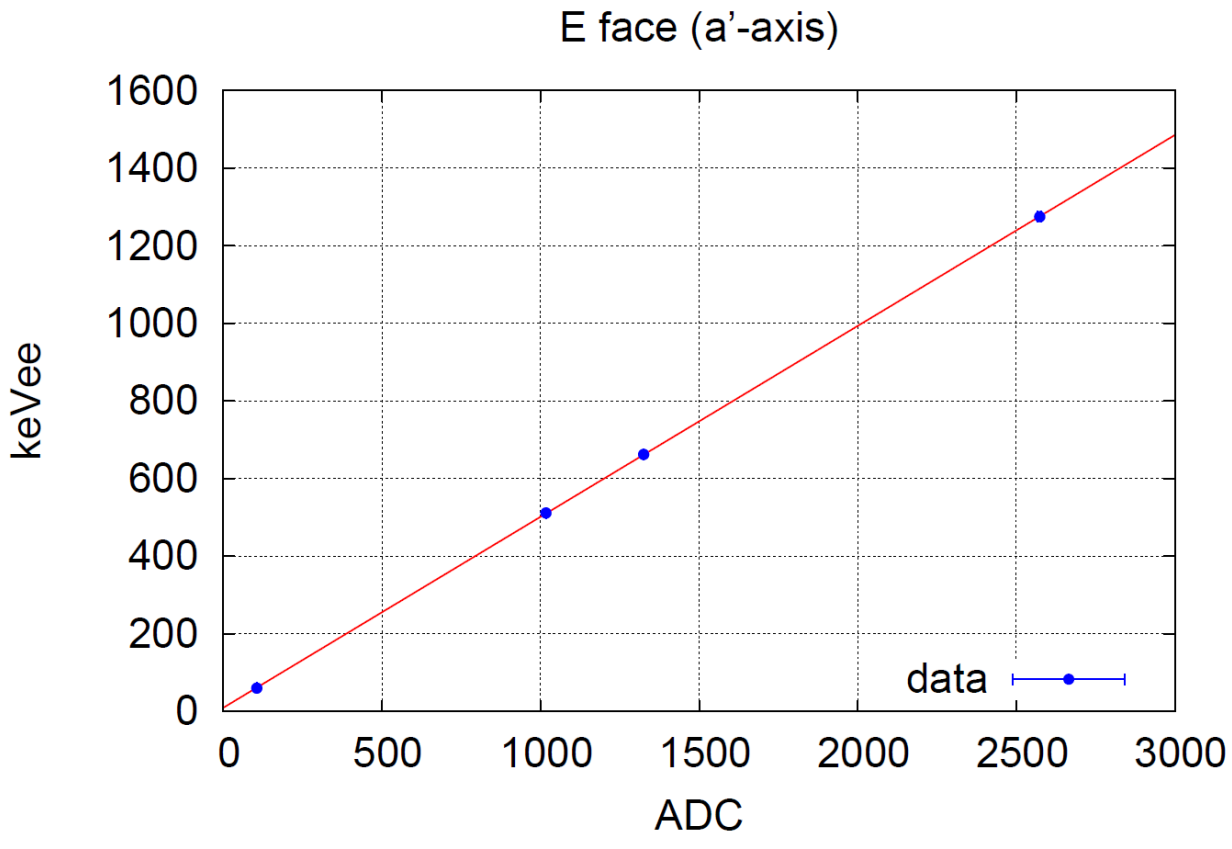


Ukraine crystal

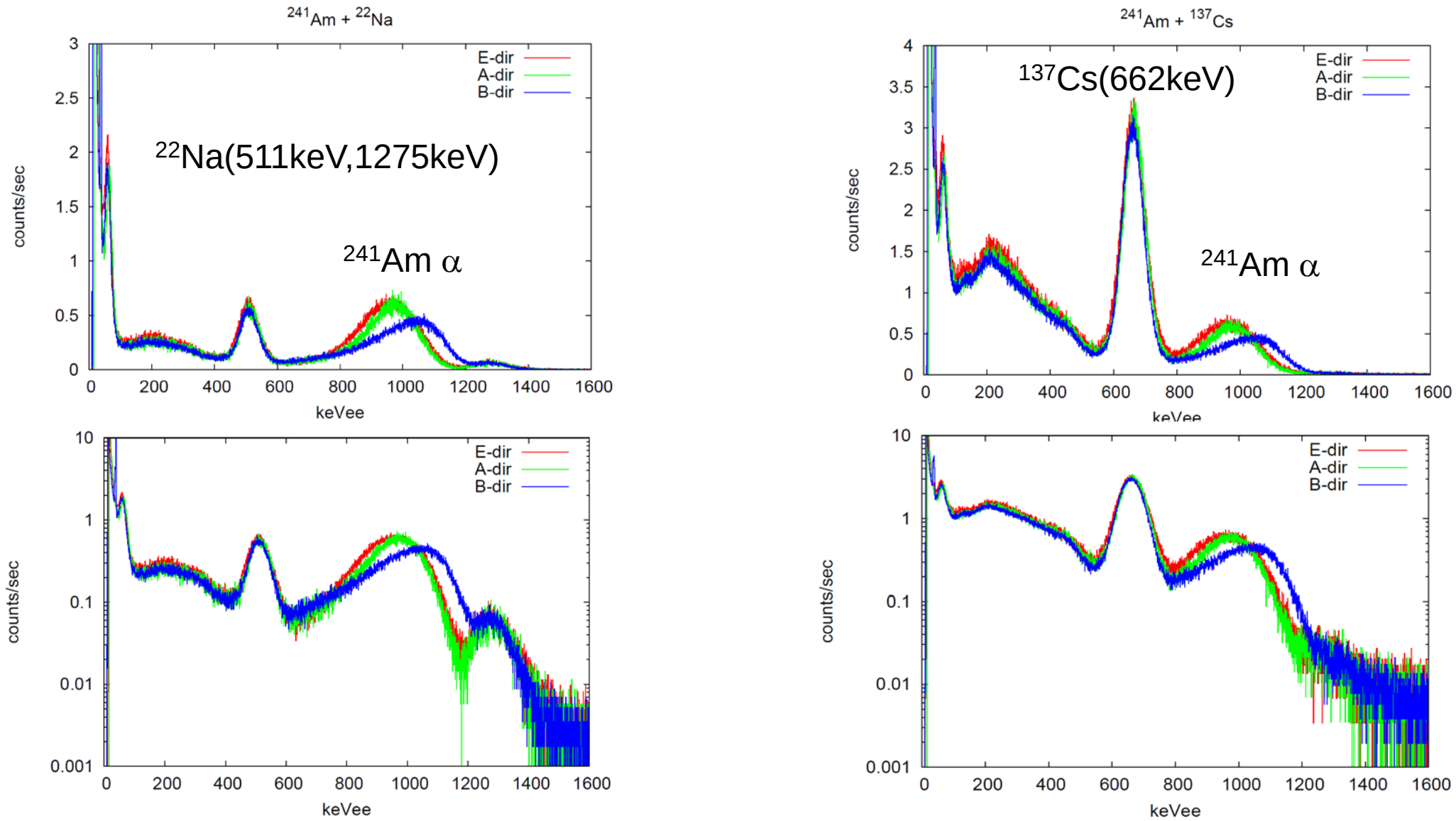
Linarites for gammas looks OK

- In any directions.





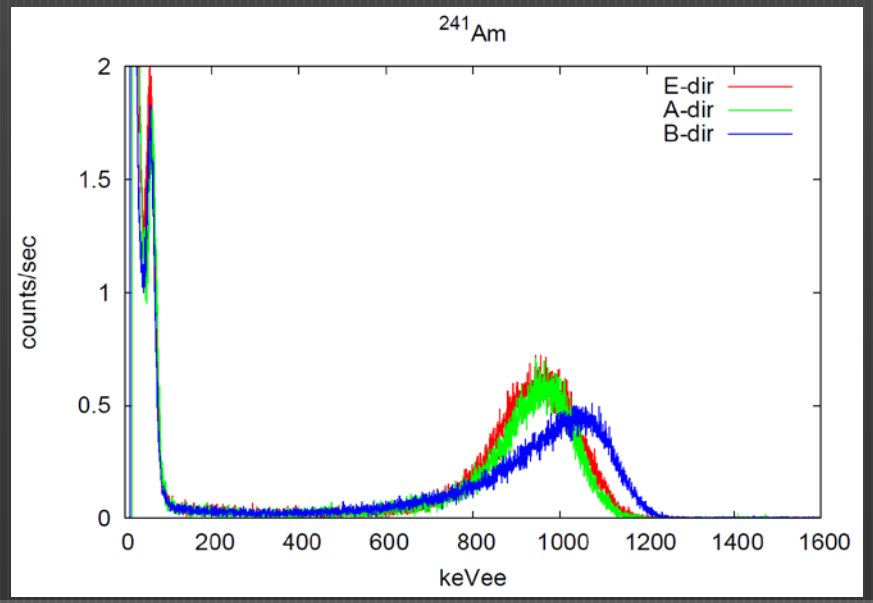
The spectra

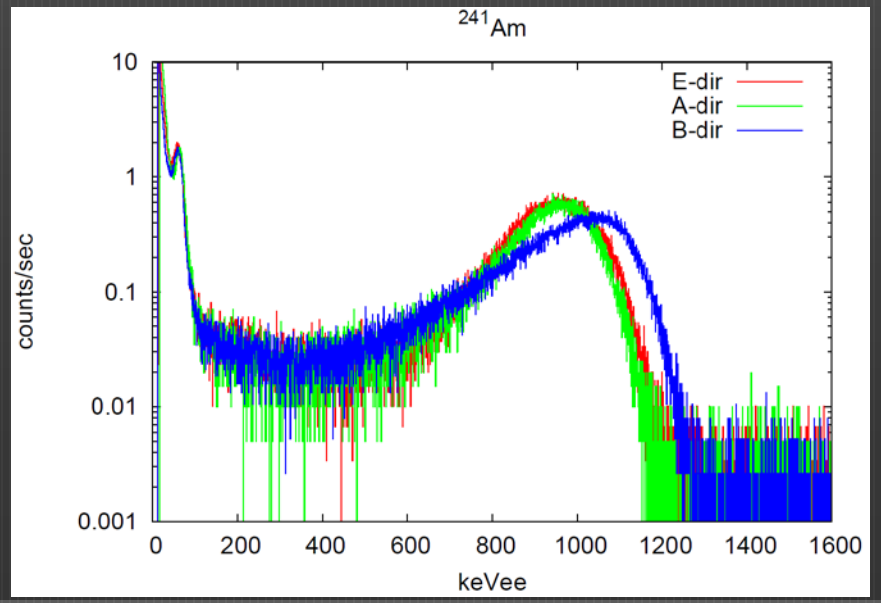

241Am + 22Na
22Na(511keV,1275keV)
241Am α
E-dir
A-dir
B-dir
counts/sec
keVee
241Am + 137Cs
137Cs(662keV)
241Am α
E-dir
A-dir
B-dir
counts/sec
keVee
E-dir
A-dir
B-dir
counts/sec
keVee
E-dir
A-dir
B-dir
counts/sec
keVee

The ratio

	γ(59.5keV)/α(5.5MeV)	βγ(511keV)/α(5.5MeV)
B	0.0499 ± 0.0009	0.471 ± 0.001
~A 20° to *c*	0.0549 ± 0.0003	0.520 ± 0.001
~E 20° to *a*	0.0544 ± 0.0003	0.521 ± 0.001

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

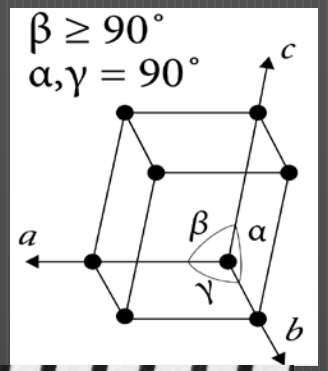




The origin of the anisotropy... crystal structure itself

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

a [Å]	*b[Å]*	*c[Å]*	α [deg]	β [deg]	γ [deg]
4.69060	5.71820	4.92690	90.0000	90.6210	90.0000



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

$(Zn_{0.95}Mg_{0.05})WO_4$

5mol% of Zn in $ZnWO_4$ replaced with Mg

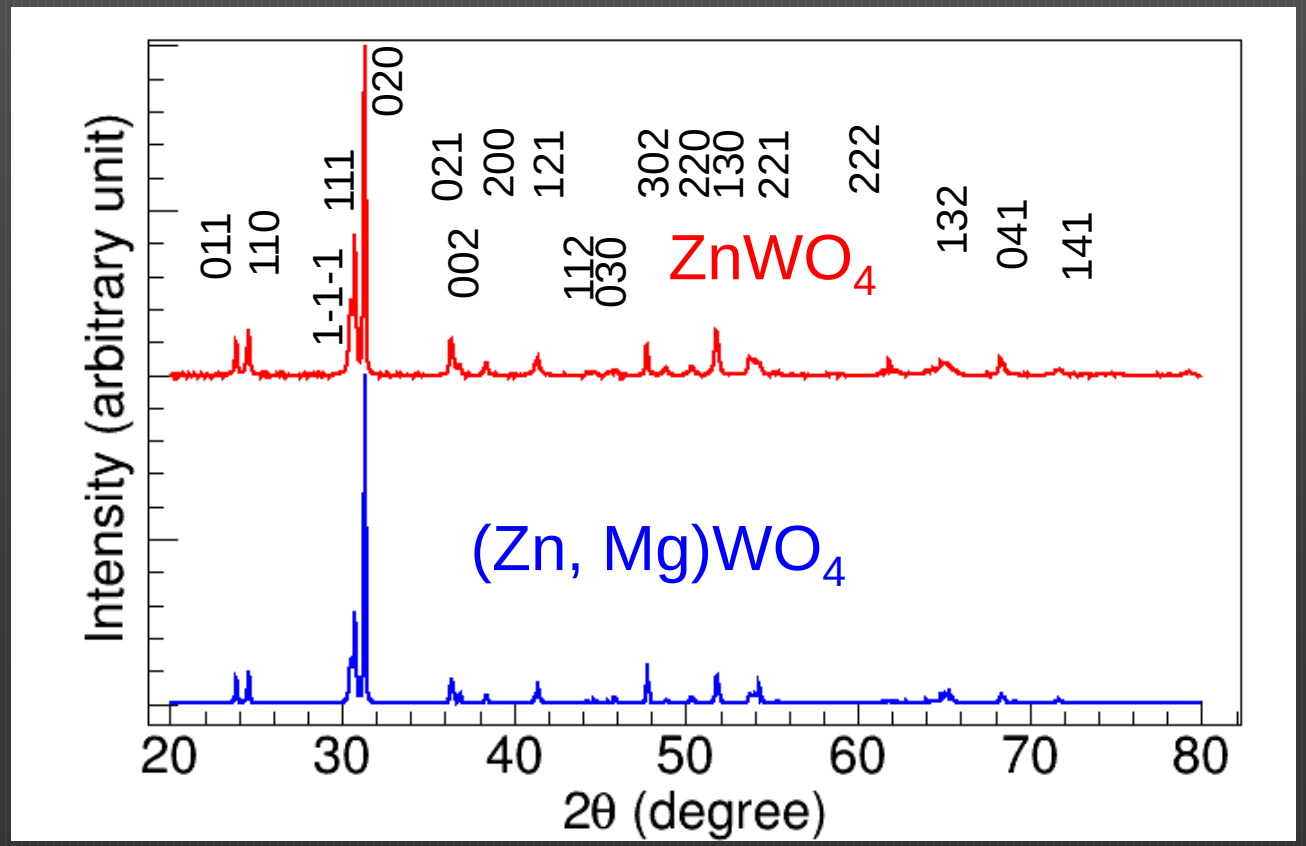
Ion radius:		
	Mg^{2+}	0.57 Å
	Zn^{2+}	0.60 Å

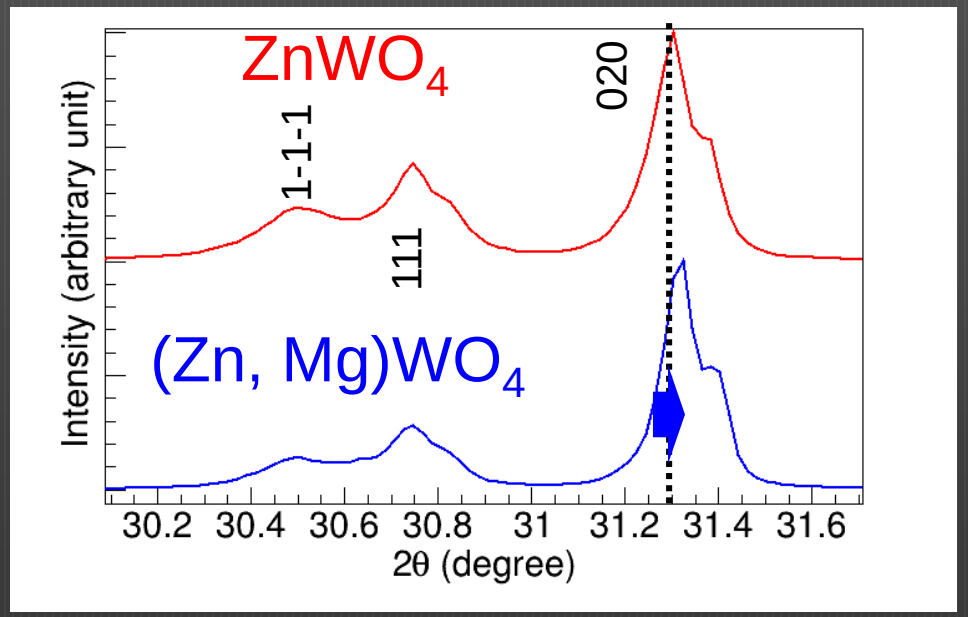
Czochralski process

Ingredients	$MgCO_3$,ZnO, WO_3 (purity 99.99%)
Pull-up speed	0.5mm/h
Direction of the seed-crystal	c-axis
Purge gas	Ar+O2 (2%)
Rotation	12rpm

Bulk structure check with powder X-ray diffraction

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

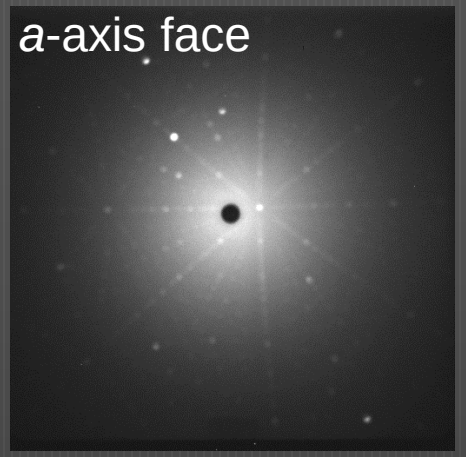


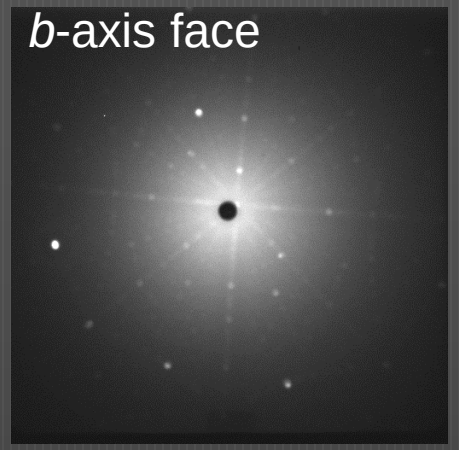


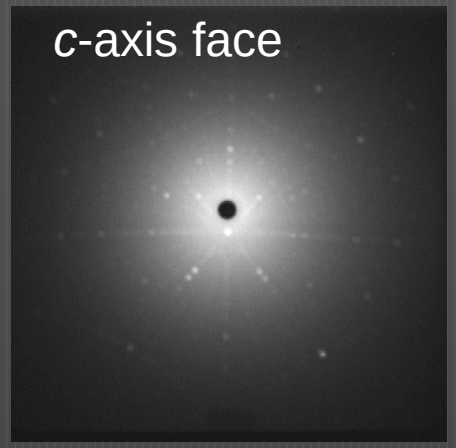
Device：BURKER D8 DISCOVER
X-ray： Cu-Kα
40 kV, 40 mA

Lattice constants

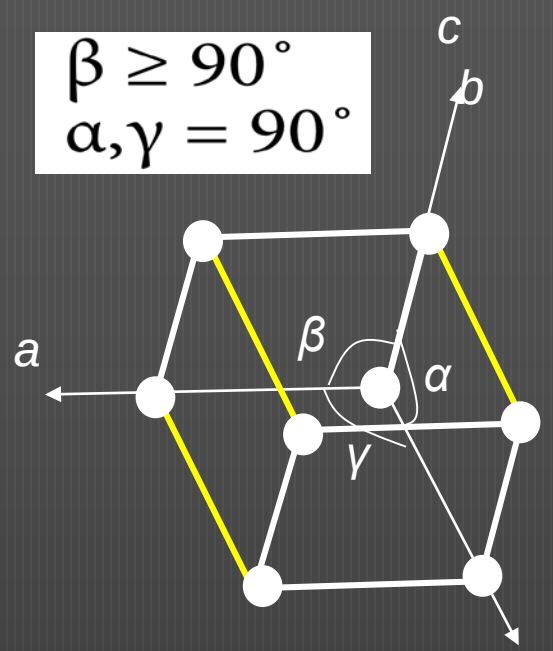
Laue pattern







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



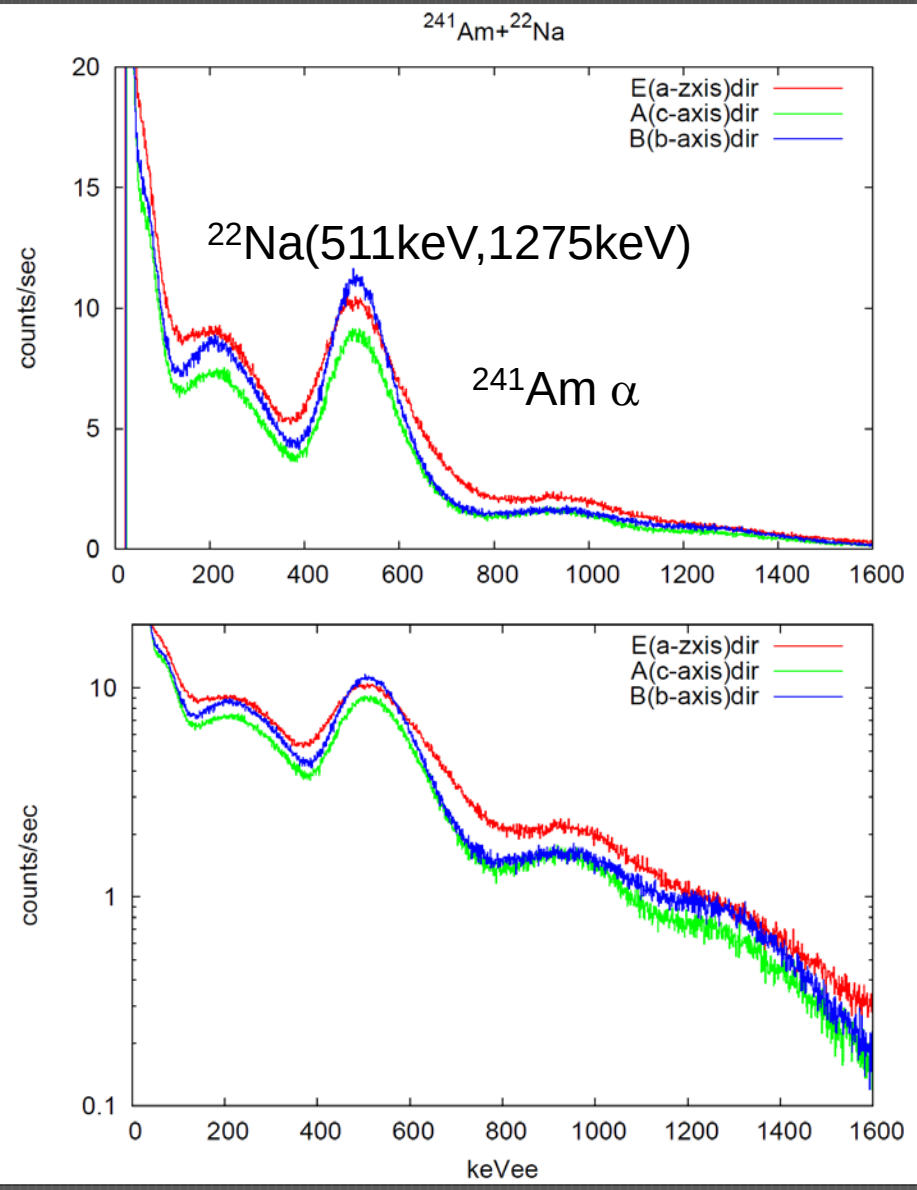
	$ZnWO_4$ Schofield et al	$ZnWO_4$	$(Zn, Mg)WO_4$
a	4.69060	4.6937(15)	4.6929(10)
b	5.71820	5.7197(18)	5.7155(12)
c	4.92690	4.9275(16)	4.9303(11)

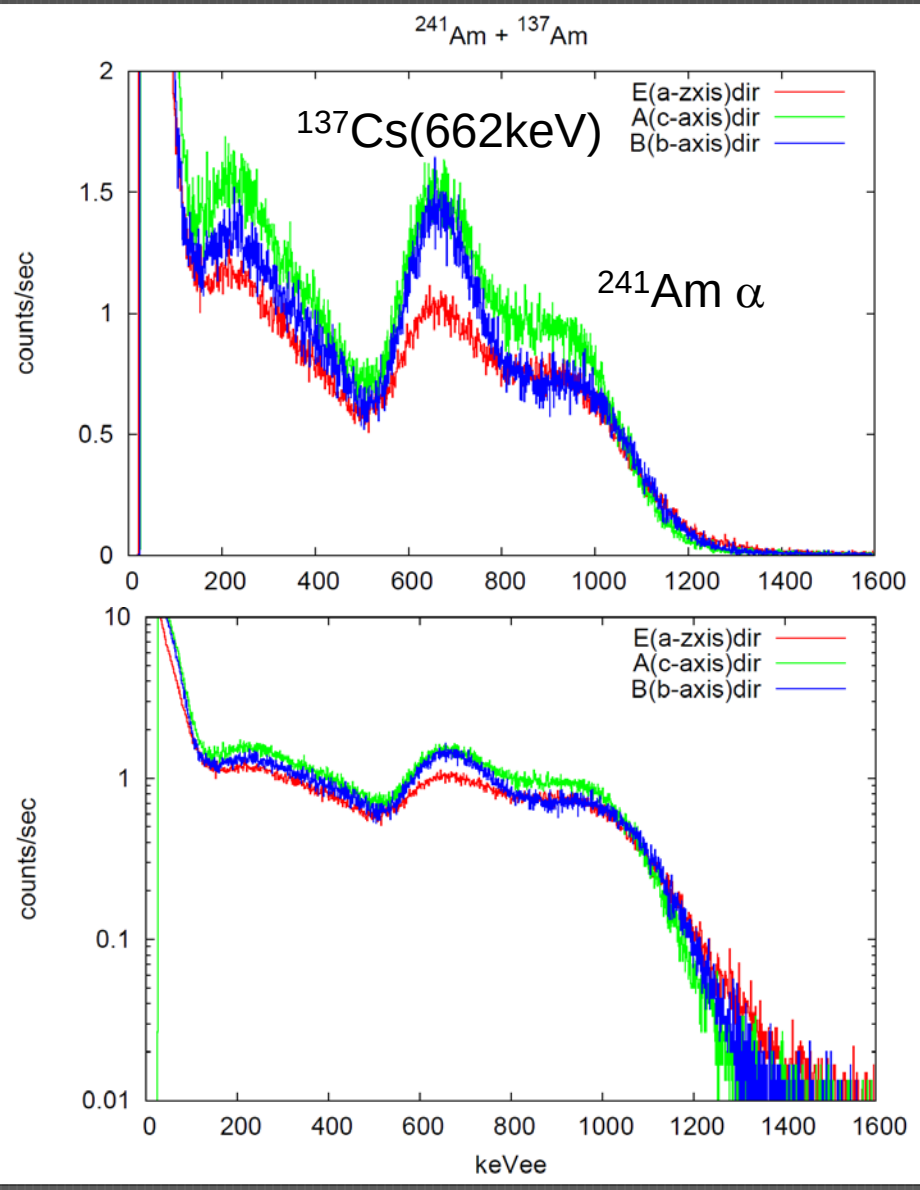
➡ 0.1% shrinkage.. Significant?

- Smaller b: smaller distortion of the lattice.

γ/α irradiation check

LY is about 70% of $ZnWO_4$

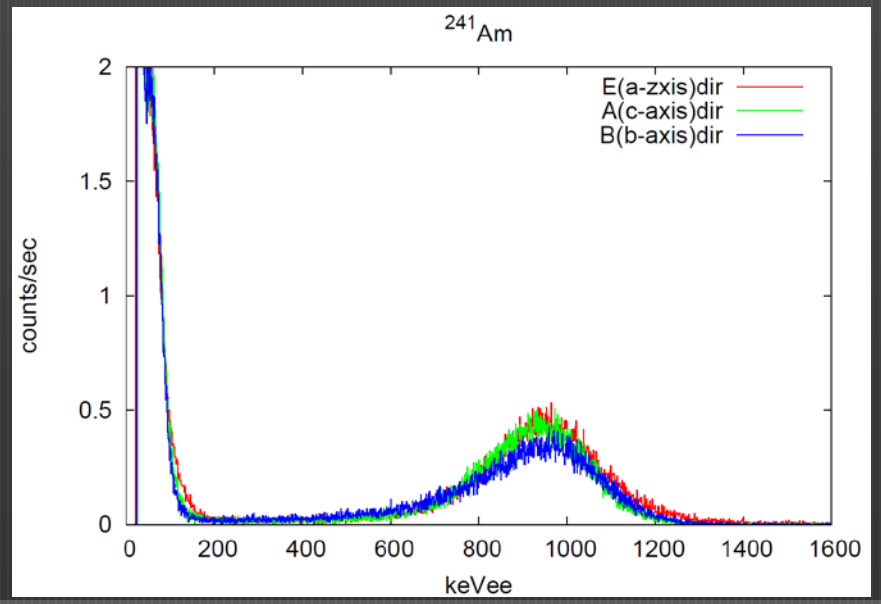


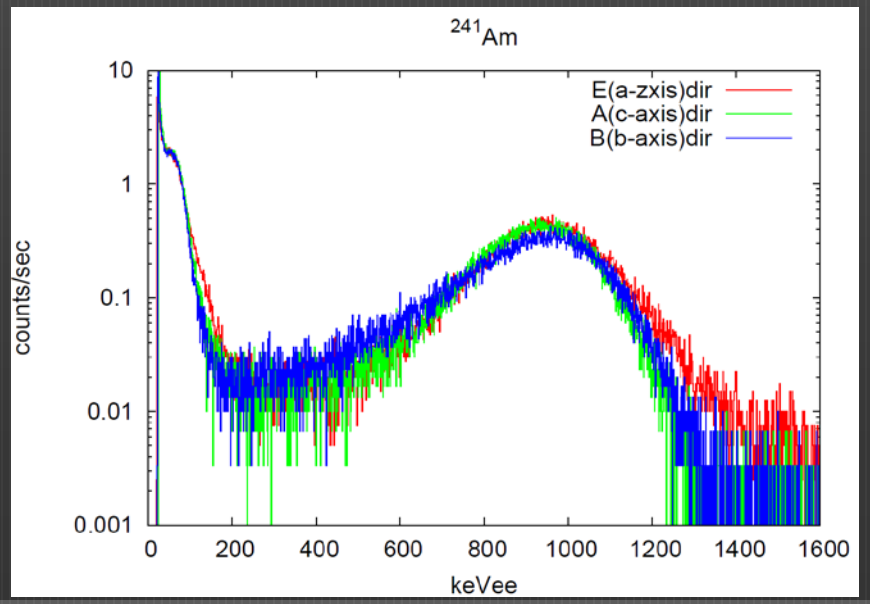


The ratio

	γ(59.5keV)/α(5.5MeV)	βγ(511keV)/α(5.5MeV)
B (*b*-axis)	0.0502 ± 0.0011	0.522 ± 0.002
A (*c*-axis)	0.0482 ± 0.0039	0.534 ± 0.001
E (*a*-axis)	0.0492 ± 0.0069	0.535 ± 0.001

- 3~4% difference or agree within systematic error
- ^{241}Am only

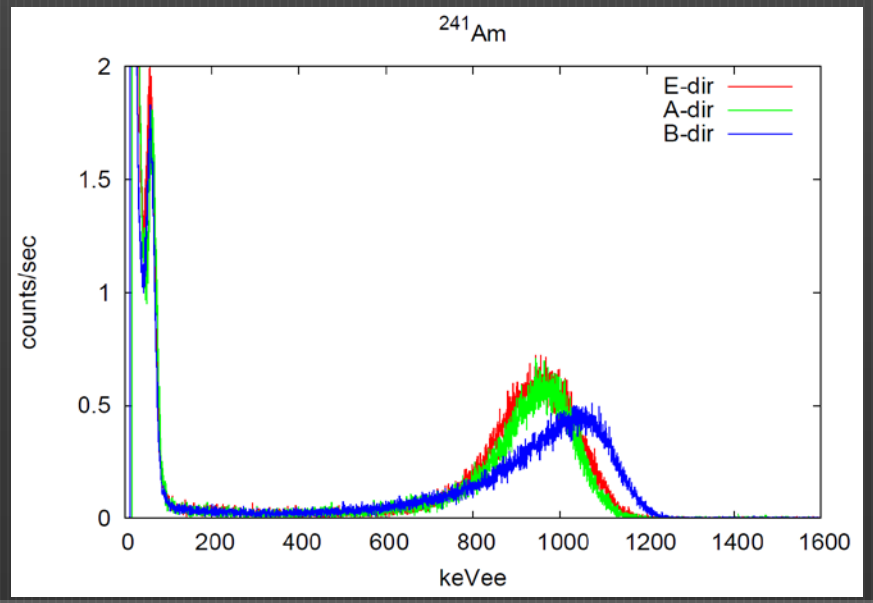


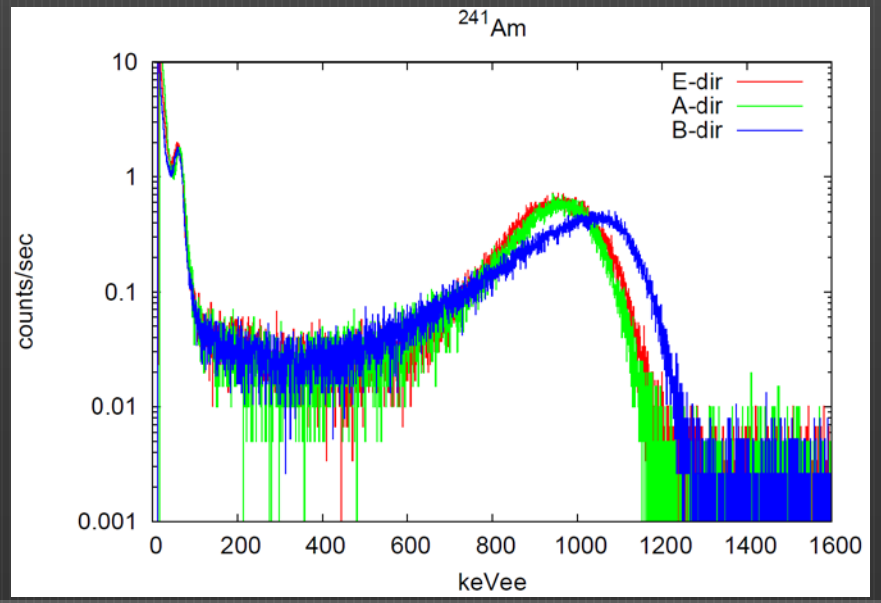


The ratio of $ZnWO_4$

	γ(59.5keV)/α(5.5MeV)	βγ(511keV)/α(5.5MeV)
B	0.0499 ± 0.0009	0.471 ± 0.001
~A 20° to *c*	0.0549 ± 0.0003	0.520 ± 0.001
~E 20° to *a*	0.0544 ± 0.0003	0.521 ± 0.001

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





Summary

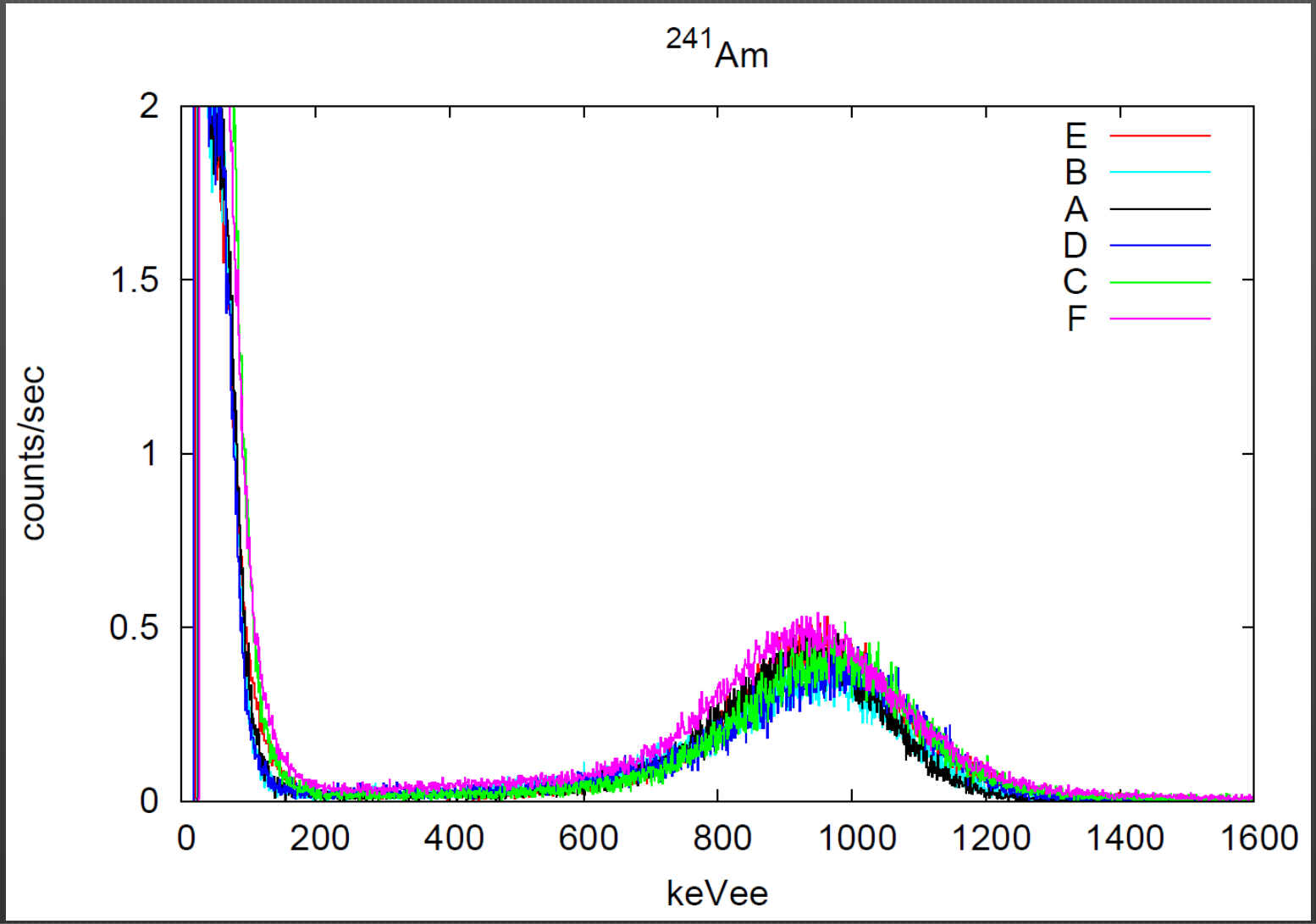
- >10% anisotropic response to α particles was confirmed in $ZnWO_4$ crystal.
  - Along *b*-axis is the maximum, consistent with ADAMO's report
  - Little or no anisotropic response to X-ray
- $Zn_{0.95}Mg_{0.5}WO_4$ crystal shows smaller anisotropic response for α particles.
  - The monoclinic structure 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}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


241Am
counts/sec
keVee
0
0.5
1
1.5
2
0
200
400
600
800
1000
1200
1400
1600
E
B
A
D
C
F