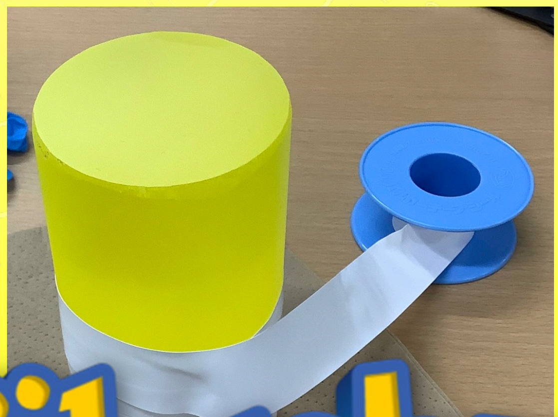
The PIKACHU experiment

– Challenge to discover $2\nu\beta\beta$ of ^{160}Gd –



2023 Dec. 21st (Thu)

Takashi Iida (U. of Tsukuba)

For the PIKACHU collaboration

NME2023 workshop @RCNP Osaka

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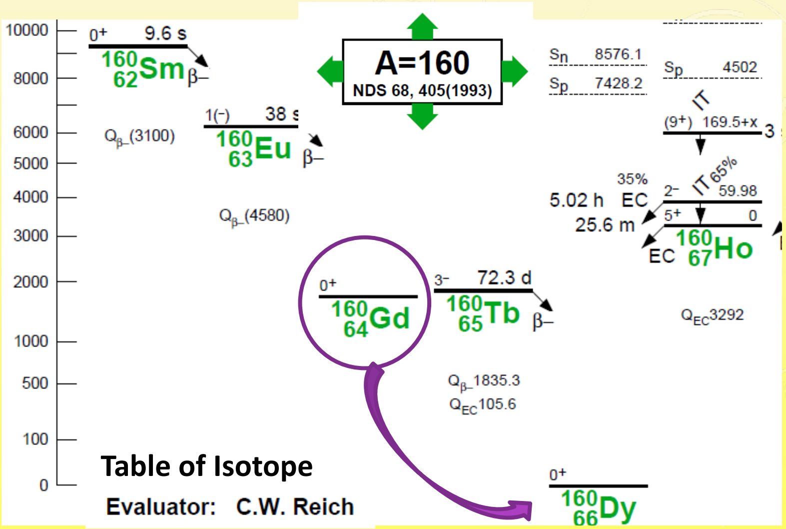
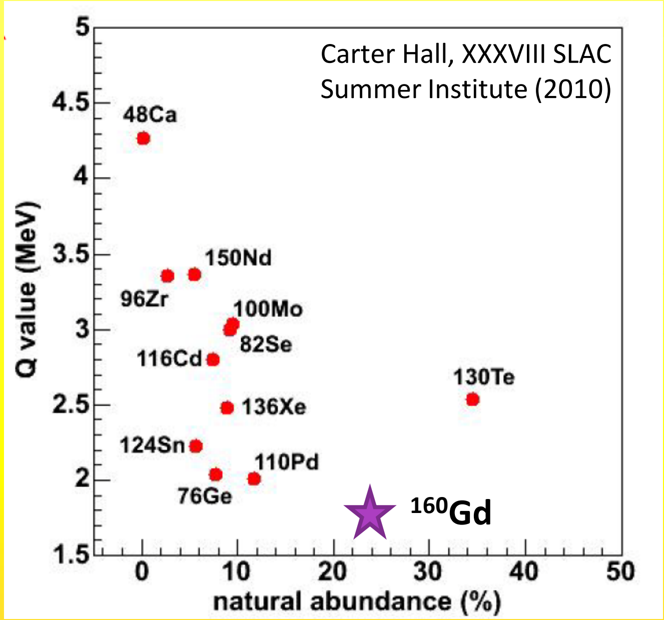
Double beta decay of ^{160}Gd

- Gadolinium (Gd) is a rare earth element lanthanide with atomic number 64 and atomic weight is 157.3.



- The ^{160}Gd isotope is one of the double beta-decay candidate nuclei.
  - ✓ Q-value : 1730 keV
  - ✓ Natural Abundance : 21.8%



Both 0νββ, 2νββ are undiscovered

NME of ^{160}Gd $2\nu\beta\beta$

- Two theoretical models predict $2\nu\beta\beta$ half-lives whose predictions differ by about an order of magnitude.

$$T_{1/2}^{2\nu} \sim 6 \times 10^{21}\ \text{yr} \quad [1] \ \ \text{(pseudo-SU (3) model)}$$

$$T_{1/2}^{2\nu} \sim 8 \times 10^{20}\ \text{yr} \quad [2] \ \ \text{(QRPA model)}$$

Theoretical description of the double beta decay of ^{160}Gd

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[1] J. G. Hirsch et al., Phys.Rev. C 66, 015502 (2002)

Global calculation of two-neutrino double-β decay within the finite amplitude method in nuclear density functional theory

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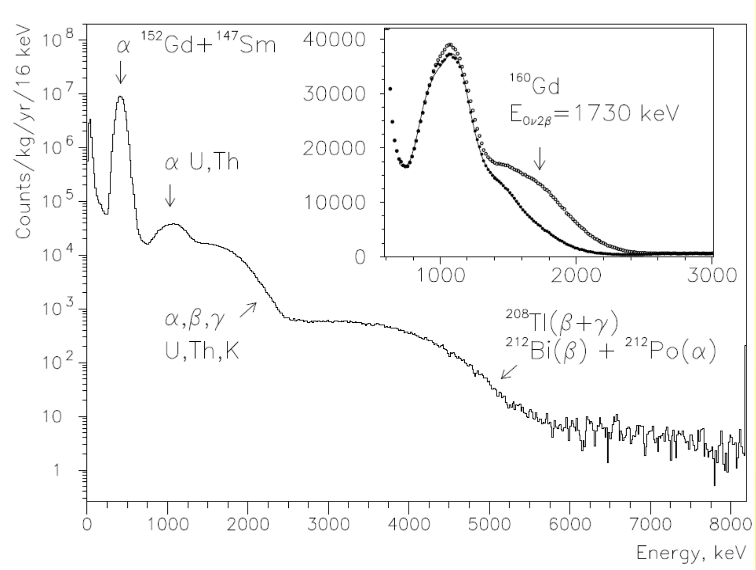
[2] N. Hinohara et al., Phys. Rev. C 105, 044314 (2022)

Previous research in Ukraine

- The world's best ββ search for ^{160}Gd is an experiment in Ukraine [3] using a 2-inch GSO scintillator.
- Alpha radiation from U/Th series impurities in the crystal appears to have been a serious background (BG).
- If the size and BG could be improved and the sensitivity increased by more than an order of magnitude, 2νββ could be found.



World best ^{160}Gd ββ search	
Detector	GSO scintillator
^{160}Gd mass	100g
Exp. period	~2 yr
Main BGs	Internal α/β of U/Th External γ from PMT
0νββ limit※	> 2.3×10^{21} yr
2νββ limit※	> 2.1×10^{19} yr

※ 90% C.L.

[3] F.A.Danevich et al., Nucl. Phys. A, Vol. 694, Iss. 1–2, 2001, Pages 375-391

The PIKACHU experiment

Pikachu ^{160}Gd

***P**ure **I**norganic scintillator experiment in **KA**mioka for **CH**allenging **U**nderground sciences*

◆ Double beta decay experiment ^{160}Gd using **$Ce{:}Gd_3Ga_2Al_3O_{12}$ (GAGG)**

GAGG crystal grown at IMR, Tohoku
6.5 cmφ × 14.5 cmL

- The aim is to increase the sensitivity by more than an order of magnitude over previous experiments using GSO and to discover $2\nu\beta\beta$.
- Large GAGG crystals can be fabricated by collaborators at the Institute for Materials Research, Tohoku University.

PIKACHU comprises six Japanese research institutes and 15 collaborators!

New collaborator is WELCOME!!

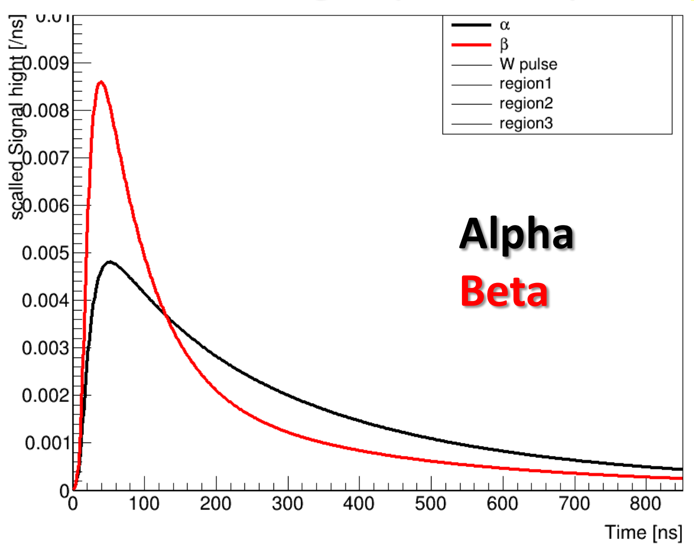
TOHOKU UNIVERSITY
筑波大学 University of Tsukuba
福井大学 UNIVERSITY OF FUKUI
JAXA
徳島大学 Tokushima University
東京大学

Our strategy

- **Large crystal**: One GAGG crystal includes 3-4 times more ^{160}Gd than GSO.
- **High LY**: Six times higher light yield enable us better energy resolution.
- **PSD**: α and β can be completely separated by PSD。
- **Low BG tech.**: Low radioactivity PMT for DM search etc.

◆ It is then necessary to remove radioactive impurities of the U/Th series inside the crystals.

GAGG averaged pulse（α/β）



	Ukraine	PIKACHU
Detector	GSO scintillator	GAGG scintillator
Amount of ^{160}Gd	100 g	700 g（2 crysltals）
LY	10,000 ph./MeV	60,000 ph./MeV
Exp. period	2 years	2 years?
BG level	Refer [3]	1/10 by PSD
$T_{1/2}$ limit	$T^{0\nu} > 2.3 \times 10^{21}$ y $T^{2\nu} > 2.1 \times 10^{19}$ y	$2\nu\beta\beta$ discovery

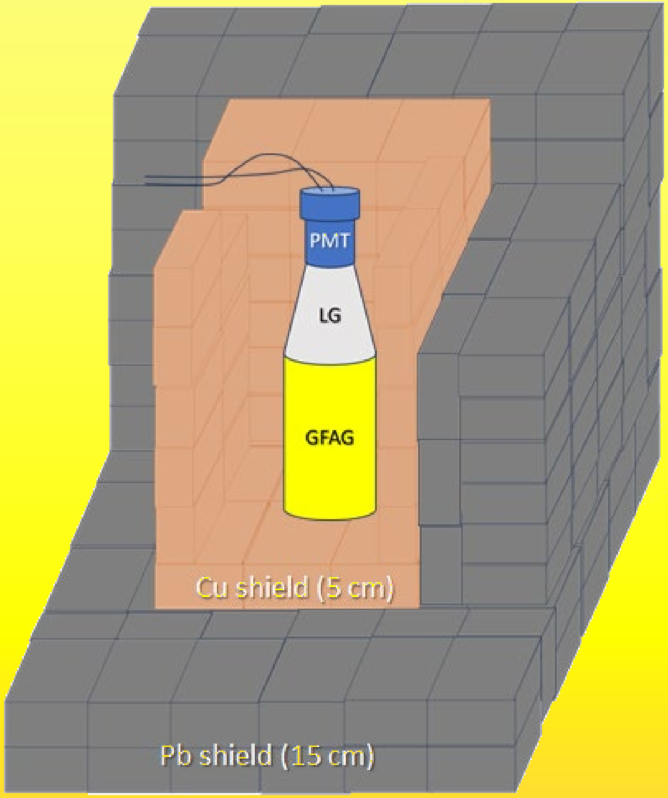
Measurement in Kamioka 2021

- The detector was made by combining GAGG, PMT and a light guide.
- It was installed in the Pb/Cu shield in the Kamioka underground laboratory of Tohoku University near KamLAND.
- BG survey was carried out in 2021.

Date： 2021 7/4～7/6

DAQ period ： 12 hours

- ✓ Shield : Pb 15cm ＆Cu 5cm thick
- ✓ GAGG ：6.5 cmϕ × 14.5 cmL (3.2 kg)
- ✓ Waveform digitizer（1.25ns/bin）
- ✓ Threshold： ~200 keV

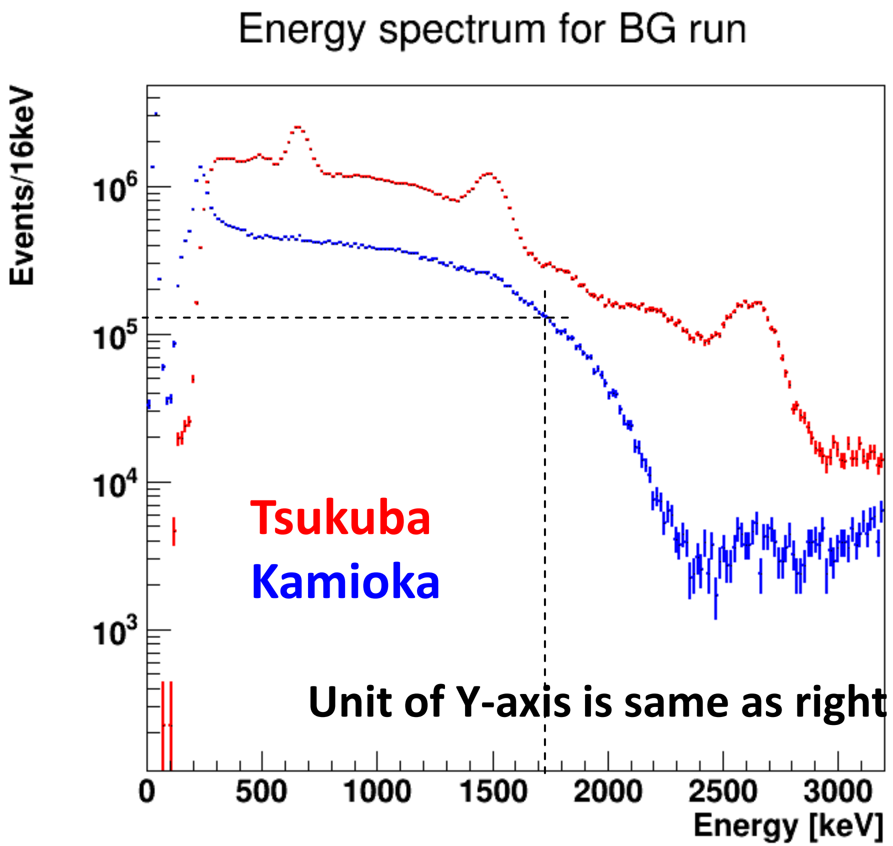


β-Spectrum comparison with GSO

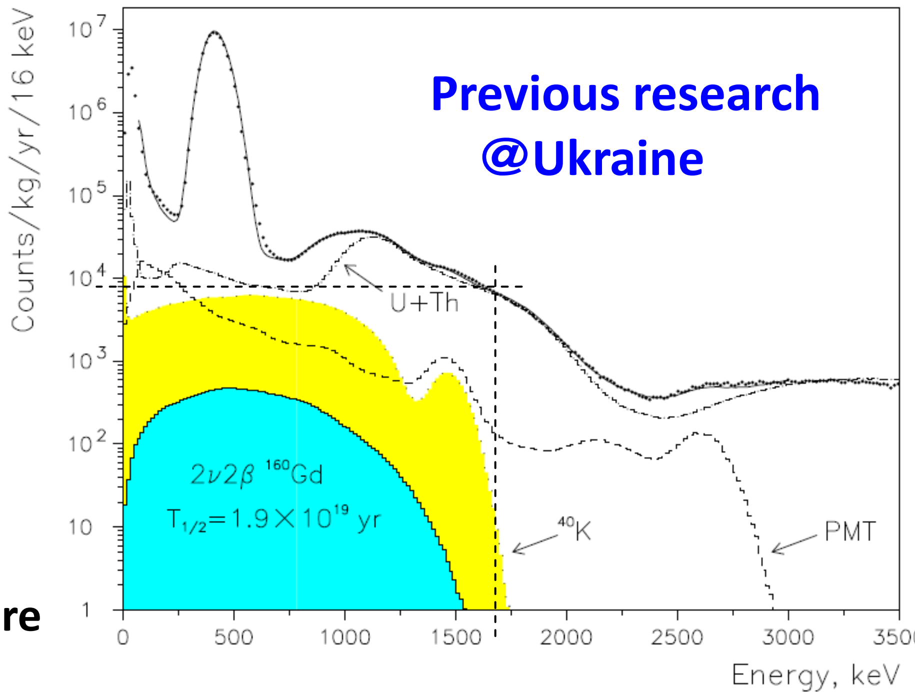
- Beta events are selected by PSD.
- BG reduced by moving to Underground.
- Our BG level is 20 time higher than in the previous research in Ukraine.

Summary of BG level

	VS Tsukuba	VS Ukraine
1000 keV	1/3	×10
1730 keV	1/2	×20
2615 keV	1/30	×10



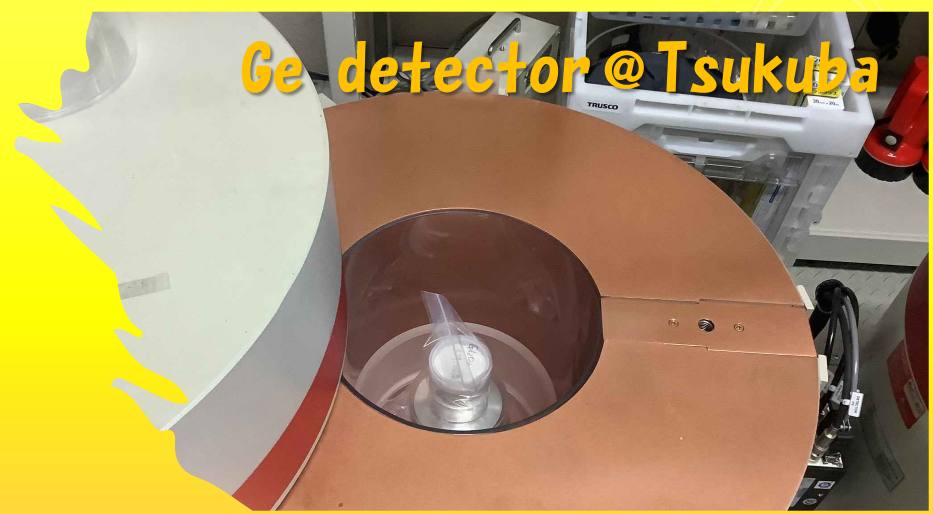
Unit of Y-axis is same as right figure



Impurity measurement of raw materials

- From the BG measurement in Kamioka, conventional GAGG crystals contain high U/Th-series radioactive impurities.

- The amounts of U/Th impurities in the main raw materials used to produce GAGG crystals were investigated using a Ge detector.
  1. Gadolinium oxide
  2. Gallium oxide
  3. Aluminium oxide
  4. Cerium oxide



Strategy to develop high purity GAGG

The following materials are used for growing large-sized GAGG crystals.

1.	**Gadolinium oxide (Gd_2O_3)**	**3.8 kg**
2.	**Gallium oxide (Ga_2O_3)**	**2.0 kg**
3.	**Aluminum oxide (Al_2O_3)**	**750 g**
4.	**Cerium oxide (CeO_2)**	**15 g**

- Pure Gd_2O_3 was made in cooperation with Nippon Yttrium (NYC), a company that makes Gd sulphate for SK-Gd. Purification by resin was carried out (¥50,000/kg).
- Al_2O_3 is difficult to purify because it is insoluble in acid; three types of high-purity Al_2O_3 are purchased; they are measured with a Ge detector and the one with the lowest impurity concentration is selected for use in the crystals.
- Ga_2O_3 and CeO_2 were also measured with a Ge detector, but these raw materials were sufficiently high purity, as described later.

Three types of Al_2O_3

Measurement of Gd_2O_3 powder

- Several raw materials were measured with a Ge detector to investigate the impurities.
- From left to right: original raw material, high-purity product from the same company, purified by NYC.

Unit is [mBq/kg]

	Gd_2O_3 (4N)	Gd_2O_3 (6N)	Gd_2O_3 (pure)
^{238}U-chain (upepr)	1750 ± 221	-	< 16.3
^{238}U-chain (midd.)	< 4.55	< 5.28	< 0.43
^{235}U-chain (lower)	130 ± 40	< 11.4	-
^{232}Th-chain	270 ± 12	10.3 ± 7.4	1.66 ± 0.41
^{40}K	84.8 ± 28.7	90.0 ± 43.8	< 0.27
Sample weight	419 g	120 g	2005 g
Meas. Time	6.0 days	20.5 days	26.9 days

✓ The purification process reduced the ^{238}U impurity by more than two orders of magnitude!!

Al_2O_3 Material investigation

[mBq/kg]

	Original	Cleaned	A	B
^{238}U-chain (upper)	476 ± 43.5	94.1 ± 22.1	52.5 ± 24.2	< 28.26
^{238}U-chain (mid.)	< 4.92	< 2.45	3.09 ± 1.67	< 5.49
^{235}U-chain (lower)	< 13.04	< 4.39	< 4.63	< 3.54
^{232}Th-chain	15.95 ± 24.37	< 9.54	16.4 ± 3.22	5.85 ± 2.80
^{40}K	< 96.5	< 40.7	123.6 ± 18.5	< 36.58
Sample weihgt	480 g	749 g	1 kg	1 kg
Meas. Time	6.4 days	10.7 days	7.5 days	6.7 days

- The original raw materials are highly impure in the ^{238}U series.
- The amount of ^{238}U impurities was reduced as a result of trying dilute nitric acid cleaning, but not enough.
- Al_2O_3 raw materials from two new companies were purchased and compared, and it was found that Company B had the lowest impurity content.

Ga_2O_3 & CeO_2 material investigation

[mBq/kg]

Ga_2O_3	Original	NYC (pure)
^{238}U-chain (upper)	< 69.17	< 38.75
^{238}U-chain (mid.)	< 9.86	< 2.49
^{235}U-chain (lower)	< 8.54	< 9.57
^{232}Th-chain	< 10.76	< 7.56
^{137}Cs	24.62 ± 3.08	< 1.44
Sample weight	444 g	1 kg
Meas. Time	4.7 days	2.7 days

CeO_2	Original
^{238}U-chain (upper)	< 59.01
^{238}U-chain (mid.)	< 3.63
^{235}U-chain (upper)	< 4.62
^{232}Th-chain	< 4.35 ± 1.87
^{137}Cs	< 1.54

- ✓ Both Ga_2O_3 and CeO_2 were originally have low impurity concentrations.

Summary of material impurities

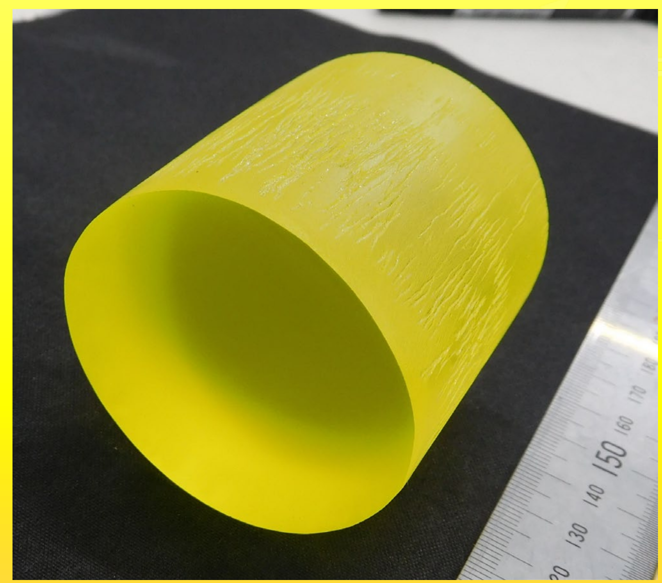
[mBq/kg]

Old	Gd_2O_3	Ga_2O_3	Al_2O_3
^{238}U-chain (upper)	1750 ± 221	< 69.17	476 ± 43.5
^{238}U-chain (mid.)	< 4.55	< 9.86	< 4.92
^{235}U-chain (lower)	130 ± 40	< 8.54	< 13.04
^{232}Th-chain	270 ± 12	< 10.76	15.95 ± 24.37
^{40}K	84.8 ± 28.7	< 76.7	< 96.5

New	Gd_2O_3	Ga_2O_3	Al_2O_3
^{238}U-chain (upper)	< 16.3	< 38.75	< 28.26
^{238}U-chain (mid.)	< 0.43	< 2.49	< 5.49
^{235}U-chain (lower)	-	< 9.57	< 3.54
^{232}Th-chain	1.66 ± 0.41	< 7.56	5.85 ± 2.80
^{40}K	< 0.27	< 27.6	< 36.58

High purity GAGG crystal for PIKACHU

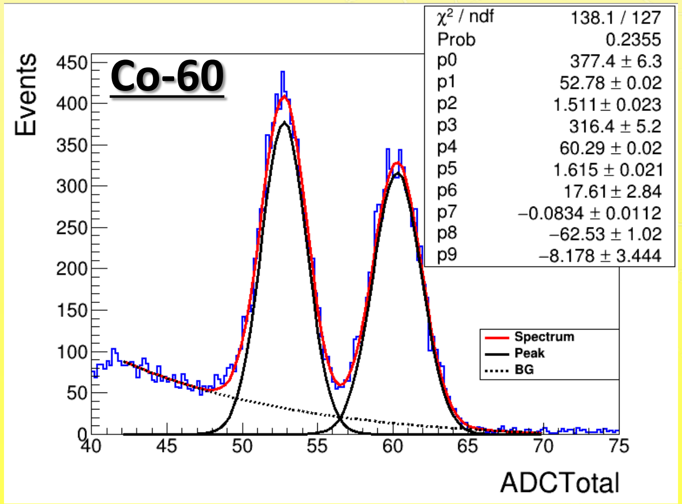
- Using high purity raw materials obtained by purification and selection, GAGG crystals of 2-inch size were grown at Tohoku University.
- The crystals were cut and polished, and the detector was fabricated by winding a reflective sheet and coupling it with a PMT and light guide.

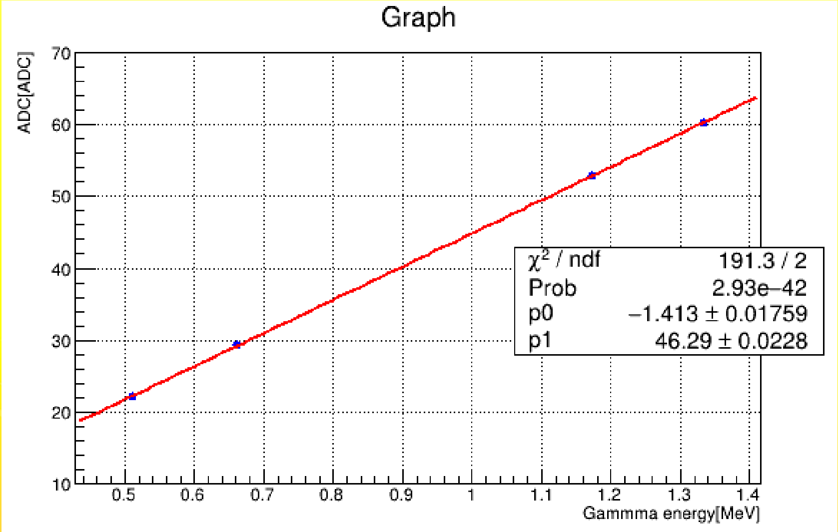


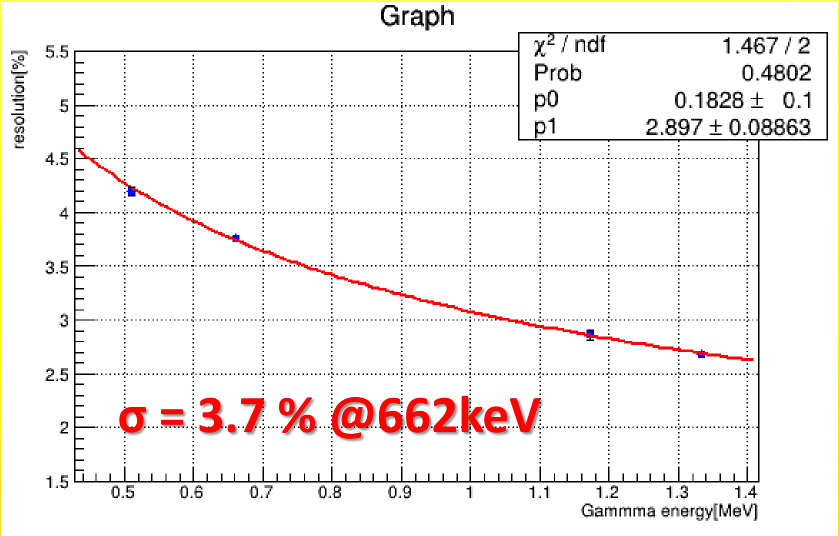
Energy calibration

- Data acquisition with three gamma-ray sources
- Fit γ-ray peak (exp + gaus)

Source	γ Energy
Cs-137	662 keV
Na-22	511 keV
Co-60	1173 keV
	1333 keV

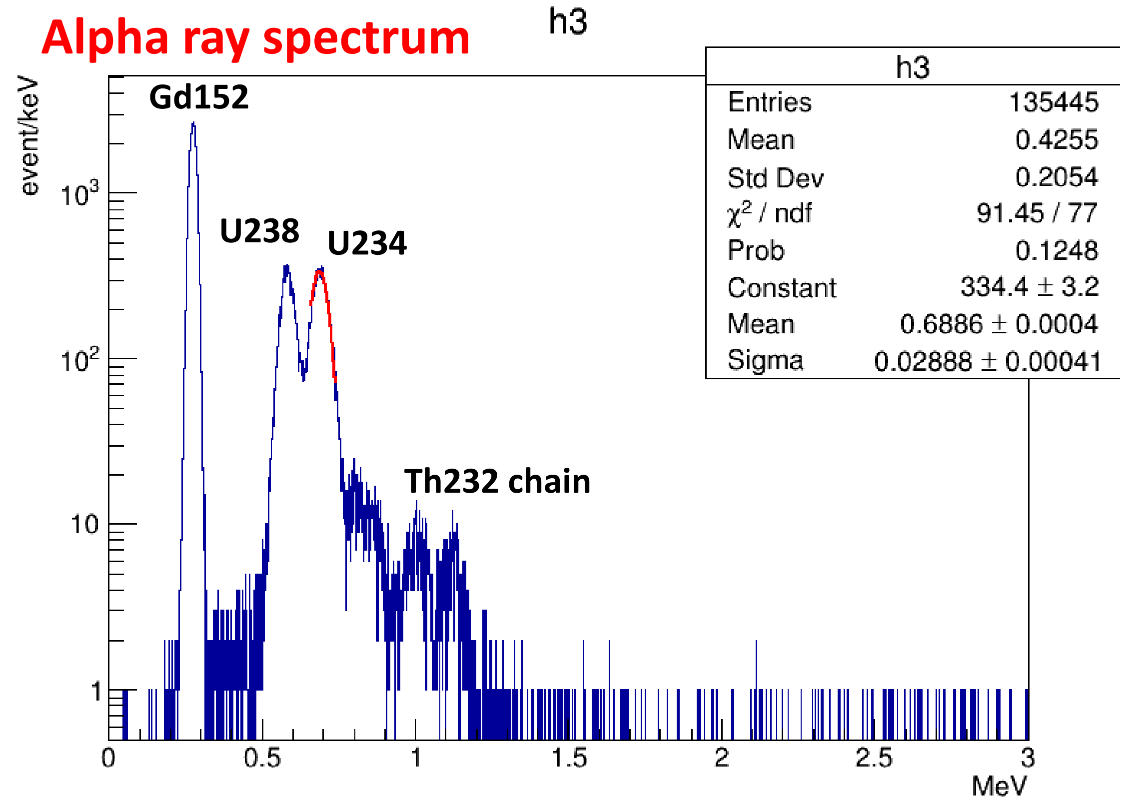


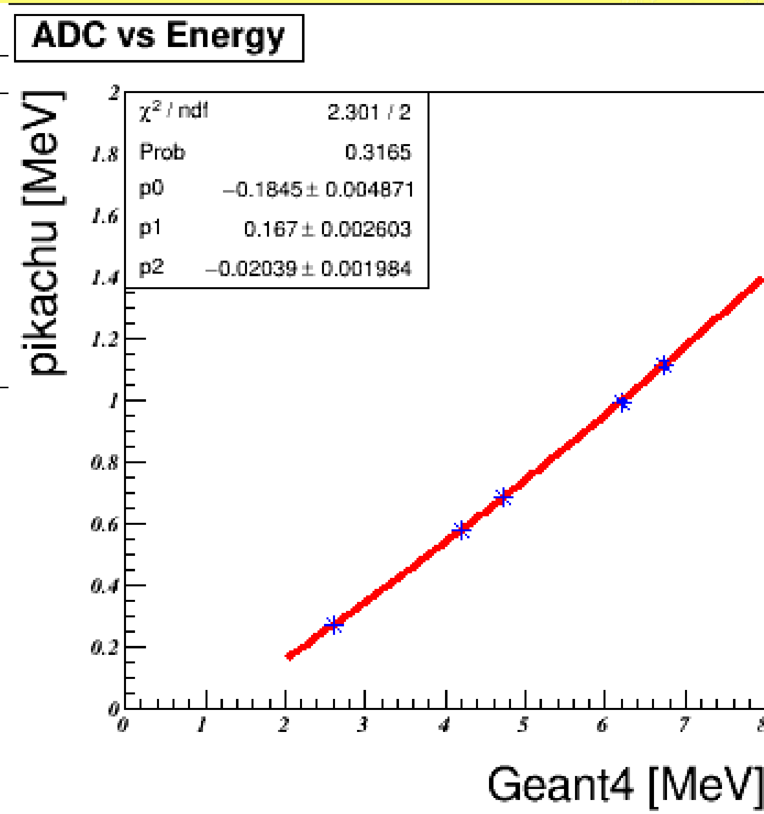




α quenching factor in GAGG

- Alpha rays emit less light per MeV (quenching) than beta rays.
- As the data are quenched, the energy dependence of the quenching coefficient was investigated for comparison with GEANT4 simulations.



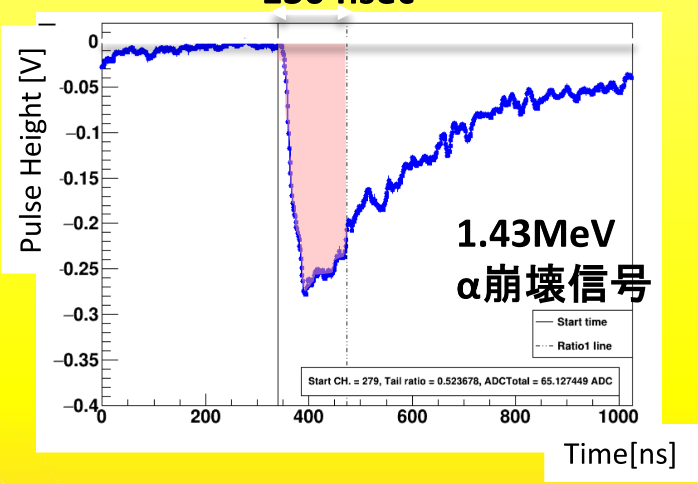


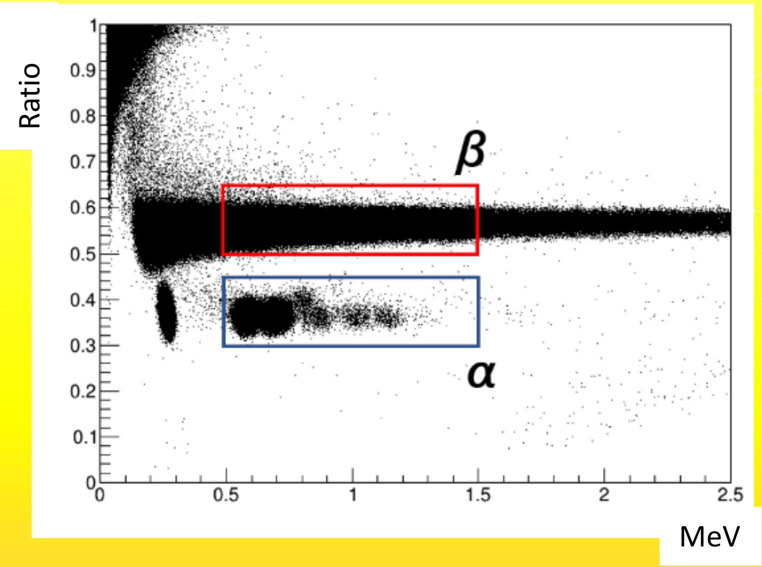
PSD capability of high purity GAGG

- Pulse shape discrimination method (PSD) is Important for removing BGs.

- Data from BG measurements carried out in Tsukuba, were used to evaluate the PSD performance of high purity GAGG.

Ratio = (integral of first 130 ns)/(integral of all region)





✓Completely distinguishable above 300 keV !!

Measurement in Kamioka 2023

- A BG survey of high-purity GAGG crystals was carried out in a low-BG environment 1000 m underground in Kamioka.
- BG comparison with previous studies. Feasibility study for sensitivity update.

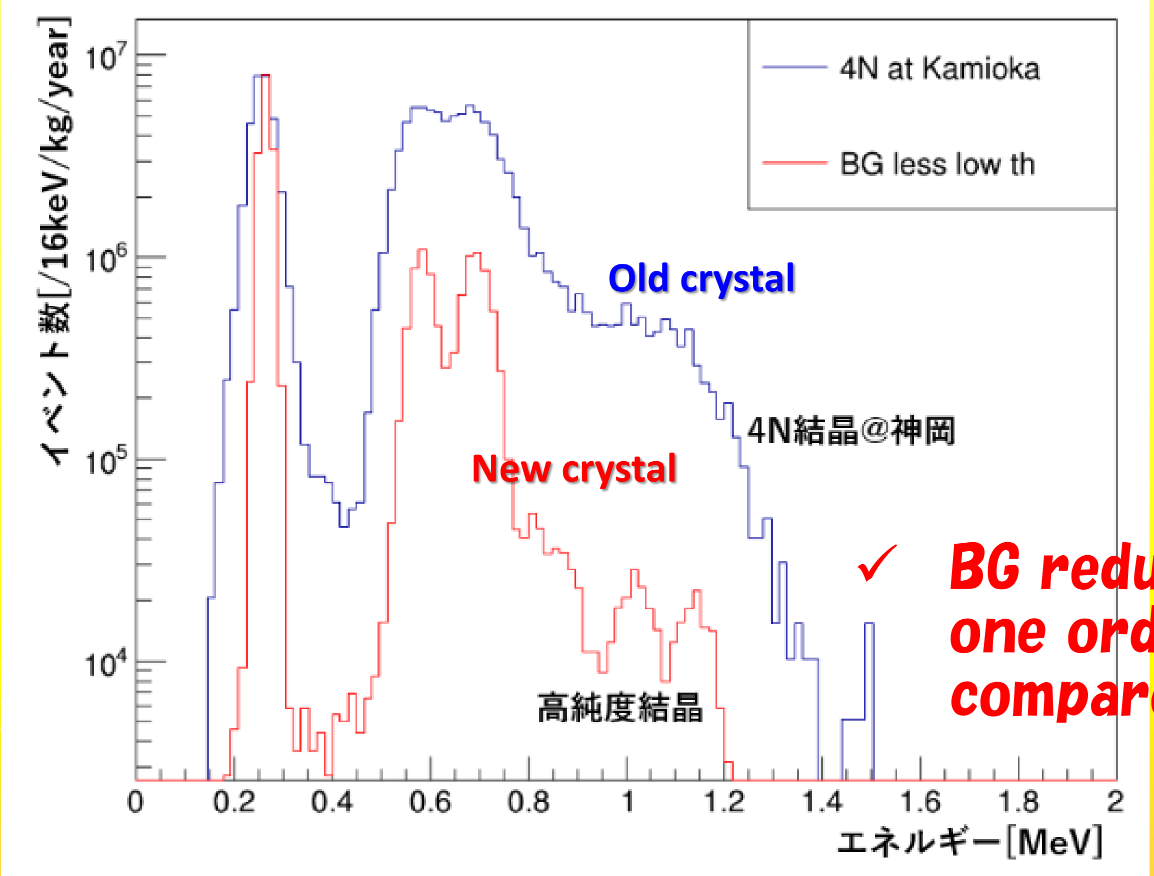
Date： 2023 6/12～6/14

DAQ period ： 18 hours

- ✓ Shield : Pb 15cm ＆Cu 5cm thick
- ✓ GAGG ：5.4 cmϕ × 5.2 cmL (0.8 kg)
- ✓ Waveform digitizer（4 ns/bin）
- ✓ Threshold： ~150 keV

BG level compared with old crystal

- The radioactive BG levels inside the crystals were estimated from the α-ray rates.
- Assumed that alpha rays are not externally caused.



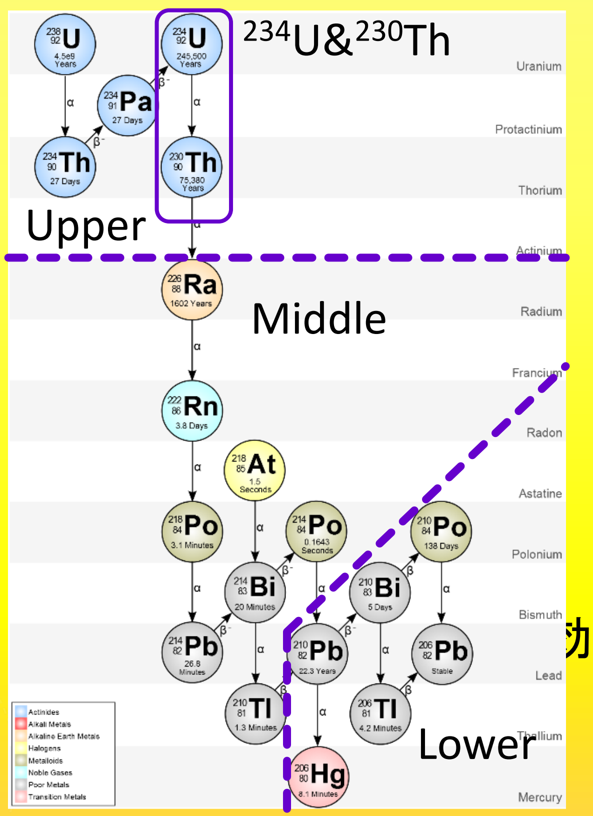
The energy resolution is also improved by changing the crystal composition.

✓ ***BG reduction of approximately one order of magnitude compared to the old crystals!***

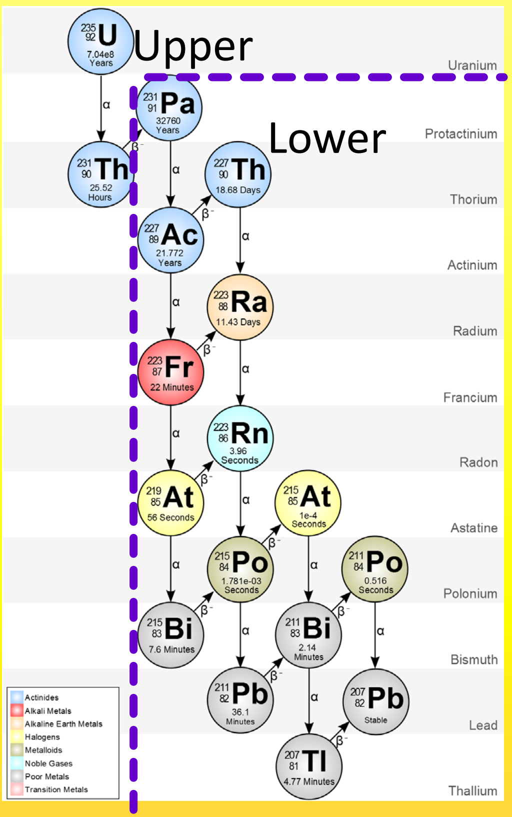
BG modelling with GEANT4

- Still remaining BGs should be quantitatively understood.
- So, an evaluation of BG in the data was attempted using GEANT4.
- Assuming radiative equilibrium below the long-lived nucleus, a U/Th-derived BG model was developed.

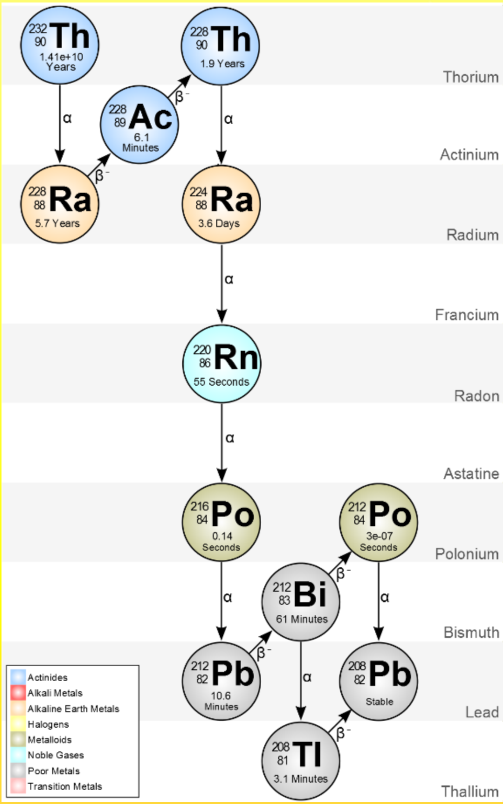
^{238}U-chain



^{235}U-chain



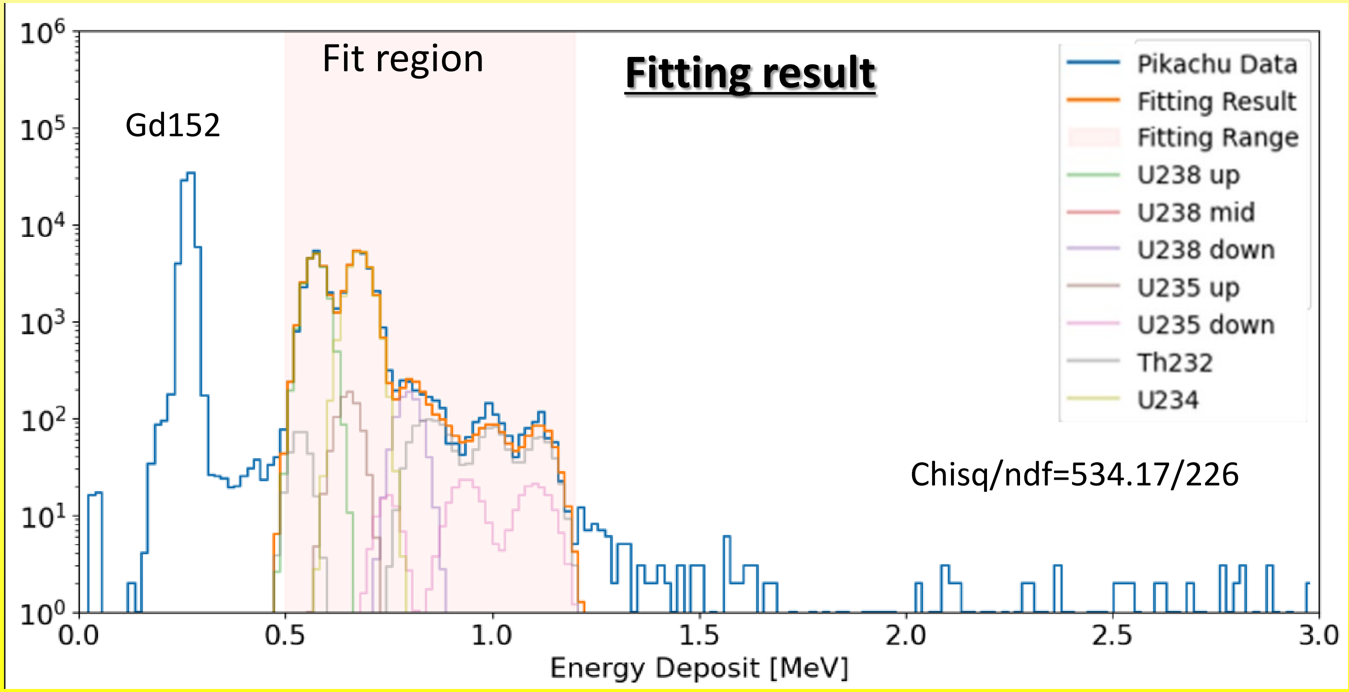
^{232}Th-chain



Understanding α-ray BG by fitting

- Fitting α spectrum with the simulated BG a spectrum.

→ Successful identification of impurity nuclides!



Unit : mBq/kg

	^{238}U upper	^{238}U middle	^{235}U lower	^{232}Th
Old GAGG	911±10	16.5±3.5	73.5±15.3	64.3±3.0
HP GAGG	**125±2**	< 0.3	3.2±0.7	2.2±0.2
Gd_2O_3 mater.	**< 16.3**	< 0..43	-	1.7 ±0.4

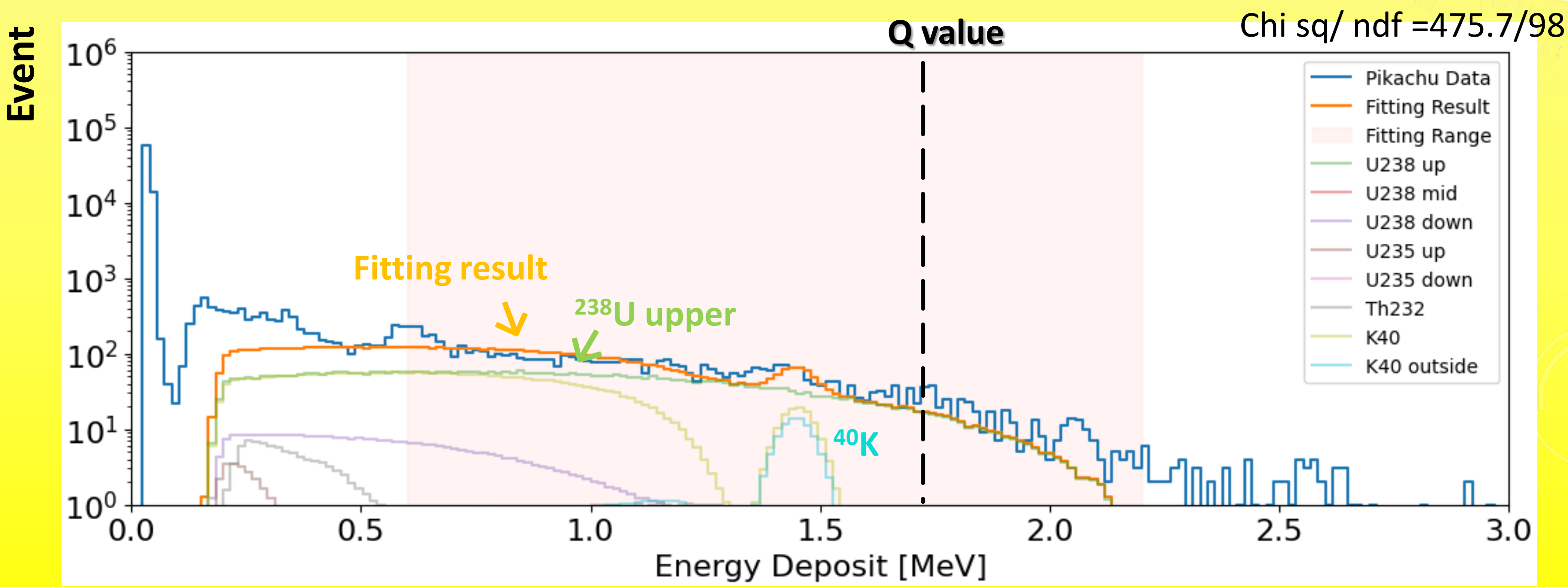
β-ray BG in high purity GAGG

✓ **Data:** Only beta events were extracted from Kamioka data using PSD.

✓ **Simulation:** two β-ray BG models

U/Th decay series in crystal ⇒ Fixed impurity content from α-ray fitting results

^{40}K of PMT origins ⇒ Spectra generated by Geant4 and fitted



✓ The β-ray spectrum was successfully reproduced by Geant4!!

⇒ The dominant BG is upstream of ^{238}U around the Q value (1.73 MeV)

✓ ^{40}K (1.46 MeV) is beginning to appear in the spectrum.

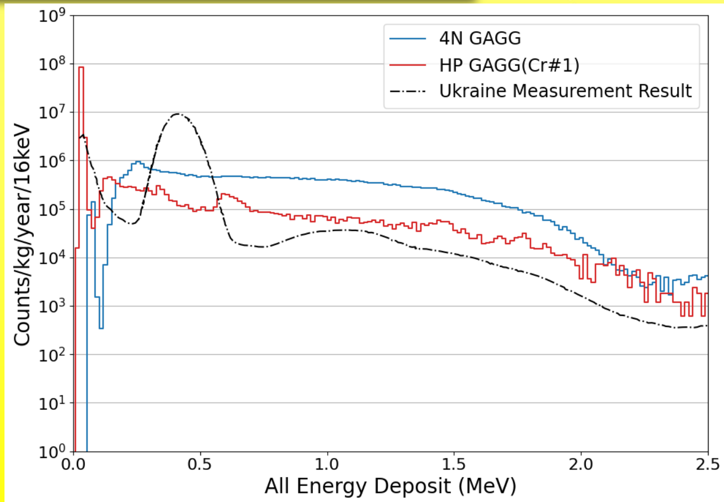
⇒ Consideration required if purification progresses

Sensitivity comparison with previous study

	GSO	Large GAGG ×2
Crystal mass	635 g	6.40 kg
^{160}Gd included	103.6 g	709.9 g
BG in Q±1.5σ	73,673 eve/kg/yr	176,669 eve/kg/yr

Assuming the same measurement time, $\boldsymbol{Sig/\sqrt{BG}}$ was compared with previous studies.

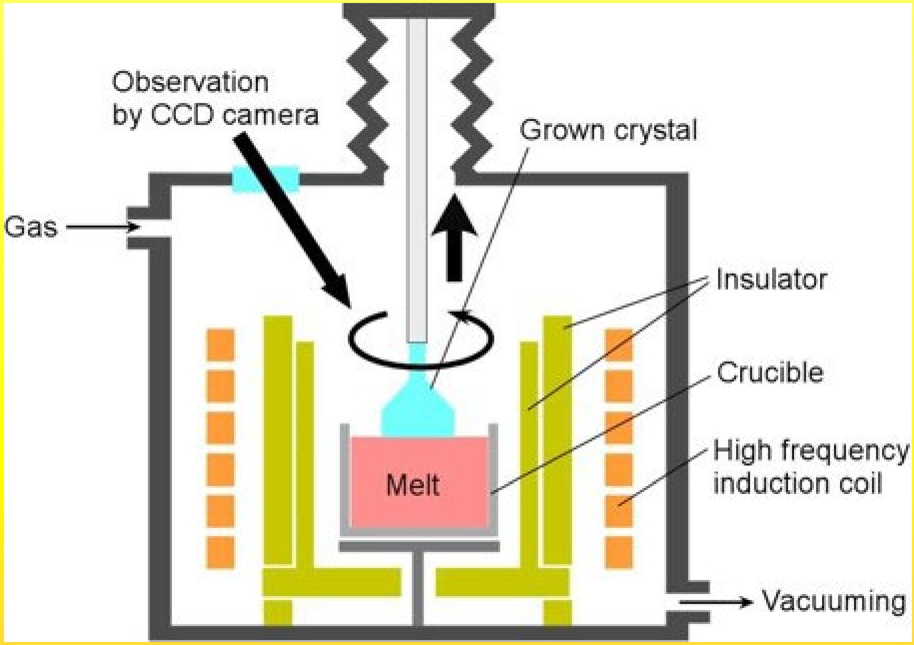
Sensitivity improve approximately 1.4 times that of the previous study is expected



- ✓ Experiment will start next year with large, high-purity GAGG crystals!
- ✓ A further reduction of BG by more than one order of magnitude is needed to search for $2\nu\beta\beta$ of ^{160}Gd.
- ✓ In parallel with the experiment, further purification will be explored.

Further purification of GAGG

- The crystal raw materials are of higher purity than the crystals, which suggests that U/Th impurities were introduced during the crystal growing process.
- The insulator made from ZrO_2 is contaminated by U/Th impurities.
- New crystals are currently being grown, while devising ways to prevent contamination from insulation.



Future prospect

- Suppose that one more order of magnitude of crystal purification is successfully achieved in the next crystal.
- Two large crystals will be grown and experiment will start in Kamioka in 2024.
- After about two years of measurements, a 4~5 times increase in sensitivity will be achieved.
- In parallel, further purification and an increase in the number of crystals are aimed at making the sensitivity 10 times higher than in the Ukrainian experiments.

I want to find a ^{160}Gd $2\nu\beta\beta$ in about 5 years!

Summary

- Double beta decay search (PIKACHU) experiment for ^{160}Gd.
- Aiming to increase sensitivity by one order of magnitude over previous studies, to discover $2\nu\beta\beta$.

  - ✓ Development of high-purity GAGG crystals!! BG~1/10 is achieved.
  - ✓ Sensitivity is equivalent to or higher than that of the previous study .

New collaborator is always welcome!!

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