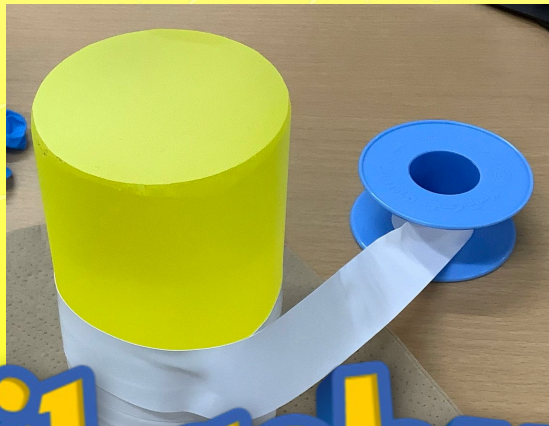


The PIKACHU experiment for the study of ^{160}Gd double beta decay



Pikachu

^{160}Gd

2024 Mar. 4th (Mon)

Takashi Iida (U. of Tsukuba)

For the PIKACHU collaboration

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UGAP2024 workshop @Tohoku Univ.

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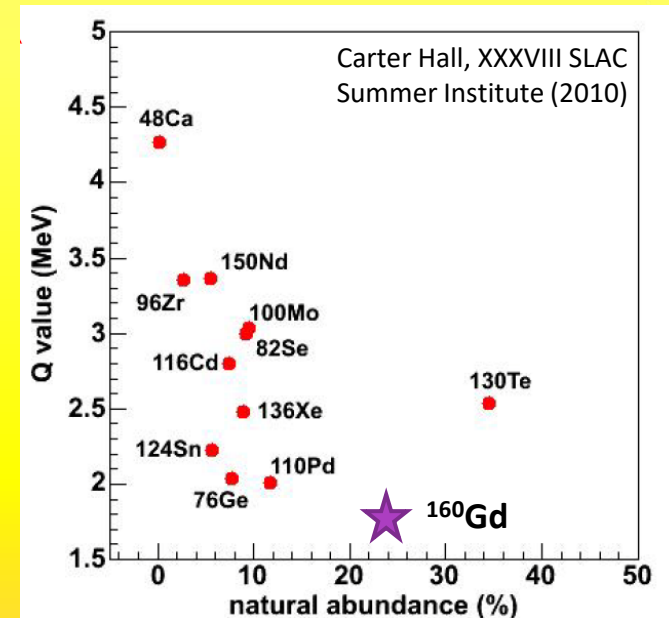
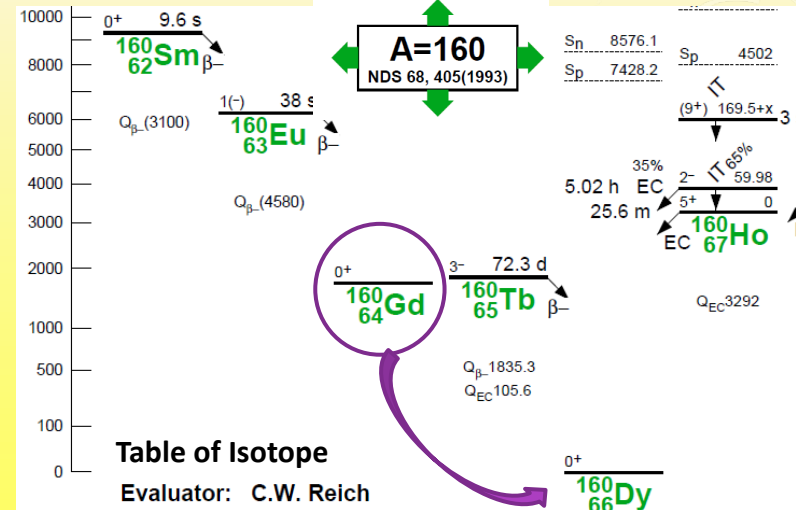
3. Summary

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 $0\nu\beta\beta$, $2\nu\beta\beta$ are undiscovered

Energy state of a nucleus with mass number 160



Nuclear matrix element (NME) of ^{160}Gd $2\nu\beta\beta$

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

$$T_{1/2}^{2\nu} \sim 6.02 \times 10^{21} \text{ yr} \quad [1] \quad (\text{pseudo-SU (3) model})$$

$$T_{1/2}^{2\nu} \sim 4.7 \times 10^{20} \text{ yr}^* \quad [2] \quad (\text{QRPA model})$$

* using same phase-space factor as ref. [1]

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)

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

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

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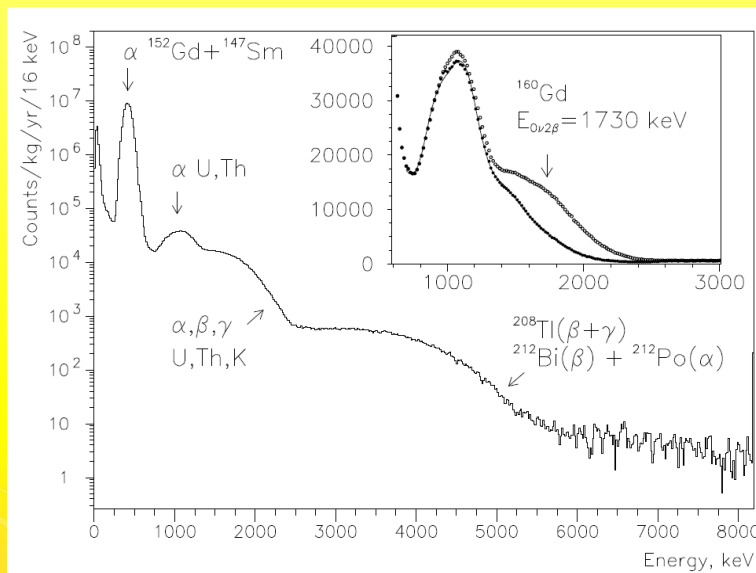
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³Department of Physics and Astronomy, University of North Carolina, Chapel Hill, North Carolina 27516-3255, USA

Previous research in Ukraine



- The world's best $\beta\beta$ search for ^{160}Gd is an experiment in Ukraine using a 2-inch GSO scintillator [3].
- α/β -rays from U/Th series impurities in the crystal was a serious background (BG).
- If the sensitivity increased by more than an order of magnitude, sensitivity approaches to predicted half-life of ^{160}Gd $2\nu\beta\beta$.



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

The PIKACHU experiment

*Pure Inorganic scintillator experiment in
KAmioka for CHallenging UUnderground sciences*



- Double beta decay experiment ^{160}Gd using **Ce:Gd₃Ga₂Al₃O₁₂ (GAGG)**

♣ Phase 1 (2024~)

Update $0\nu\beta\beta$ search sensitivity of ^{160}Gd using large crystals of similar purity as GSO in previous studies.



♠ Phase 2 (2026~?)

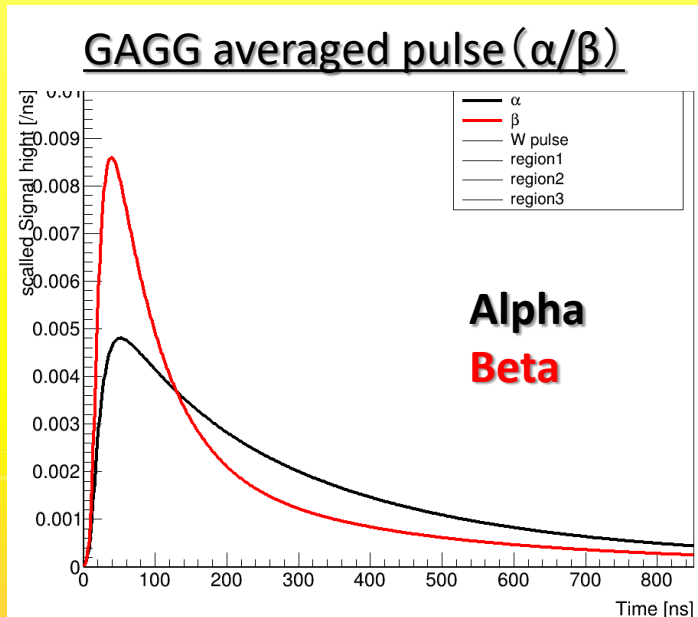
Discovery of ^{160}Gd $2\nu\beta\beta$ using ultra-high purity crystals with an order of magnitude higher purity.



6 Japanese institutes and 12 collaborators!

Our strategy to increase sensitivity

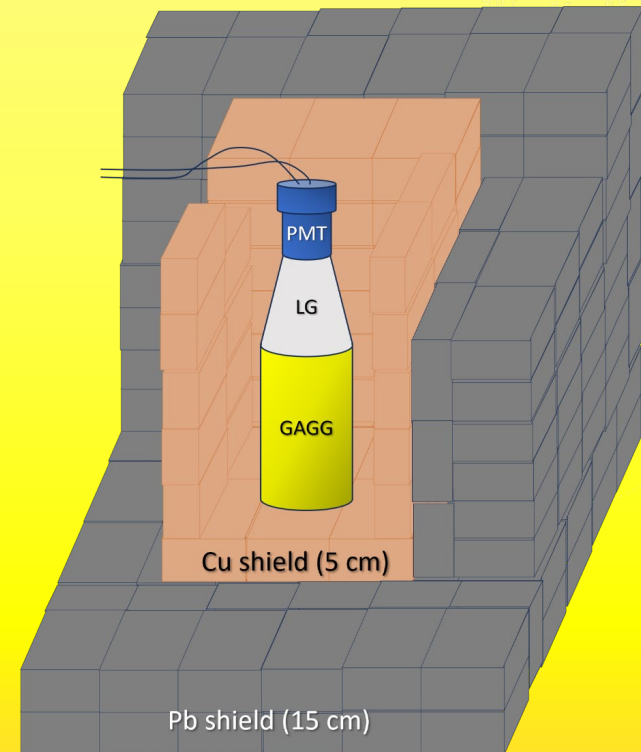
- **Large crystal:** One GAGG crystal includes 3-4 times more ^{160}Gd than GSO.
 - **High LY:** Six times higher light yield enable us better energy resolution.
 - **PSD:** α and β can be completely separated by PSD.
 - **Low BG tech.:** Low radioactivity PMT for DM search etc.
- ◆ It is then necessary to remove **radioactive impurities of the U/Th series** inside the crystals.



	Ukraine	PIKACHU
Detector	GSO scintillator	GAGG scintillator
Amount of ^{160}Gd	100 g	700 g (2 crystals)
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	$0\nu\beta\beta$ search ph.1 $2\nu\beta\beta$ discovery ph.2

BG study in Kamioka @2021

- BG survey using conventional GAGG was carried out in Kamioka in Jul. 2021.
- The detector module was installed in the Pb/Cu shield in the KamLAND area. Thanks to Inoue-san, Koga-san and Ikeda-san for the cooperation!
 - Shield : Pb 15cm & Cu 5cm thick
 - GAGG : 6.5 cm ϕ \times 14.5 cmL (3.2 kg)



✓ **As a result, we found that conventional GAGG crystal contains 10 times more U/Th compared to GSO in Ukraine** ☹️☹️☹️

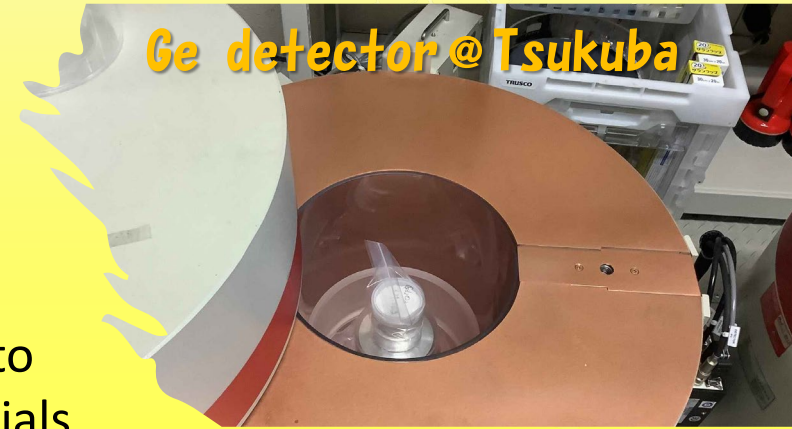
Development of high purity GAGG



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 |

Ge detector in Tsukuba and Kamioka were used to investigate radio impurities inside the raw materials.



- ✓ Pure Gd_2O_3 was made in cooperation with Nippon Yttrium (NYC), a company that makes $\text{Gd}_2(\text{SO}_4)_3$ 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. Several samples were measured with a Ge detector and **the lowest impurity one is selected for use.**
- ✓ Ga_2O_3 and CeO_2 were also measured with a Ge detector, but these raw materials were sufficiently high purity.

Summary of raw material purification

[mBq/kg]

	Gd_2O_3		Al_2O_3	
	High-purity	Original	High-purity	Original
^{238}U	< 16.3	1750 ± 221	< 28.3	476 ± 44
^{235}U	< 10.0	130 ± 40	< 7.82	< 21.1
^{232}Th	1.66 ± 0.41	270 ± 12	5.85 ± 2.80	16.0 ± 6.6
^{40}K	< 2.70	84.8 ± 28.7	< 36.58	< 96.48

	Ga_2O_3	CeO_2
^{238}U	< 69.2	< 59.0
^{235}U	< 8.54	< 15.5
^{232}Th	< 10.8	4.4 ± 1.9
^{40}K	< 35.8	< 23.5

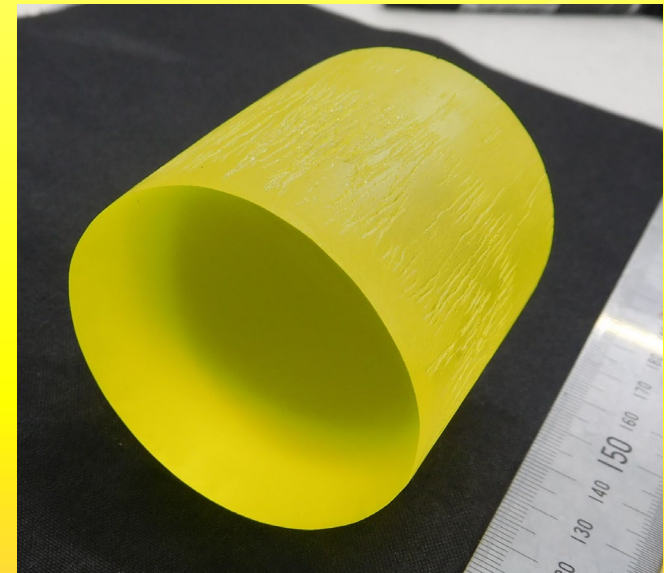
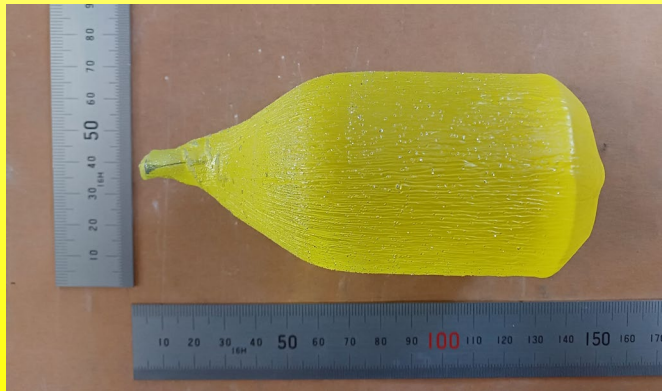
- ^{238}U , most important BG in PIKACHU, was significantly reduced by purification of Gd_2O_3 and selection of Al_2O_3 .
- For Ga_2O_3 and CeO_2 , original materials were enough pure.

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

High purity GAGG crystal for PIKACHU



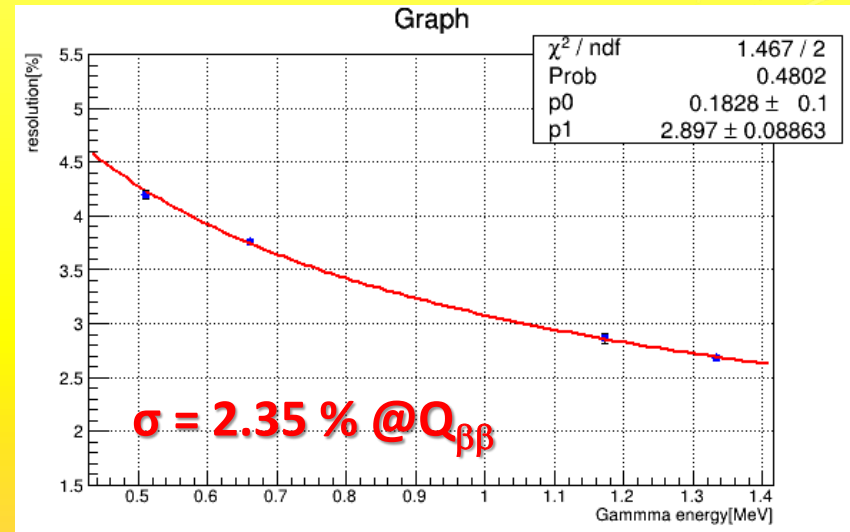
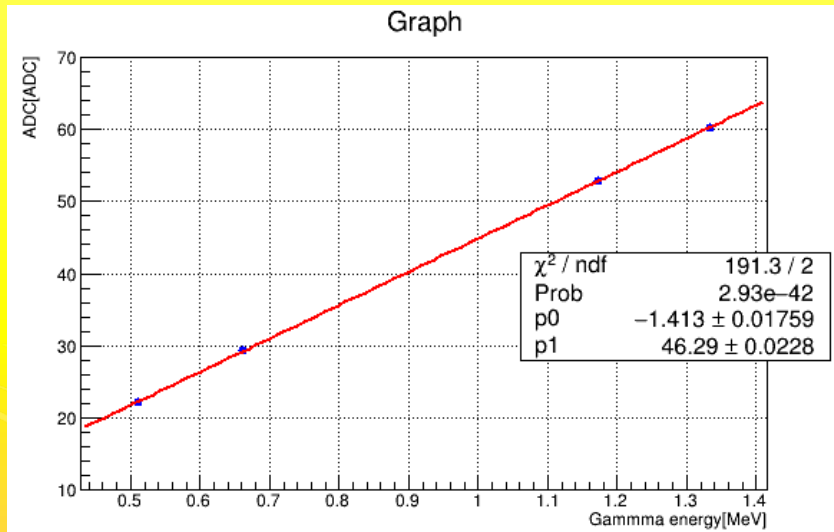
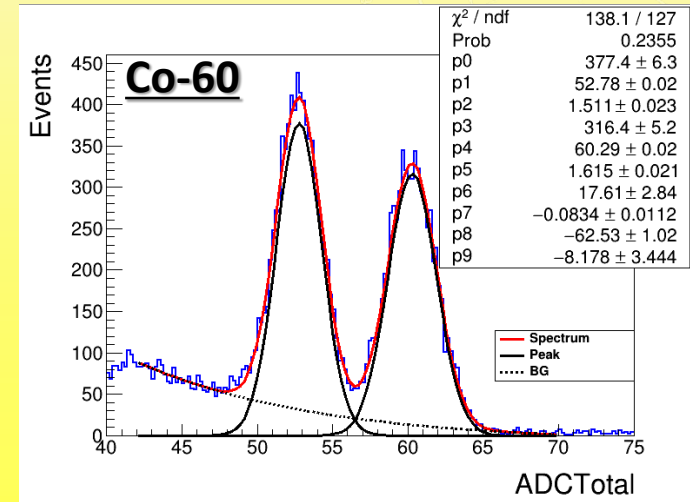
- Using high purity raw materials obtained by purification and selection, **GAGG crystals of 2-inch size were grown** at Tohoku University.
- The crystals were cut and polished, and the detector was fabricated by winding a reflective sheet and coupling it with a PMT and light guide.



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



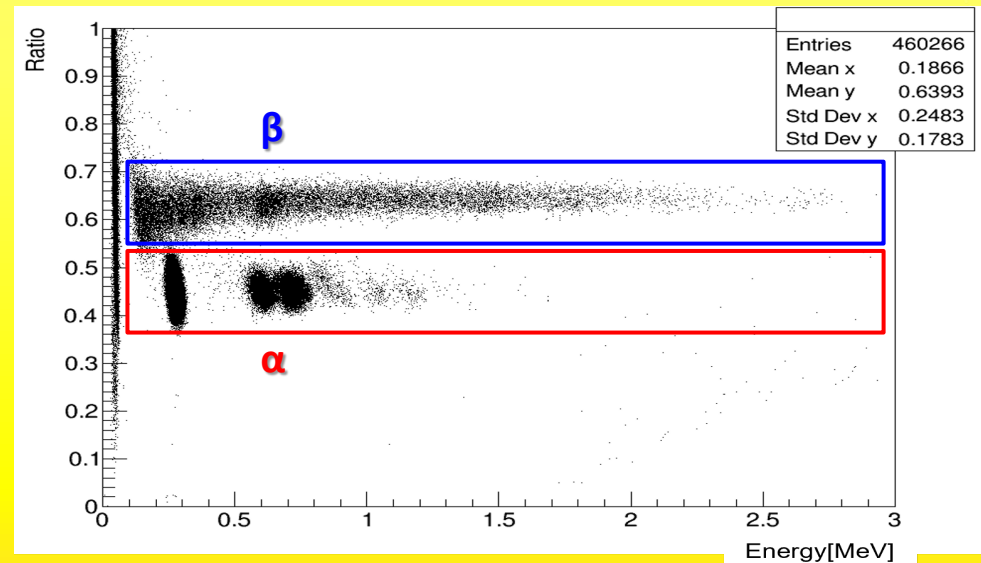
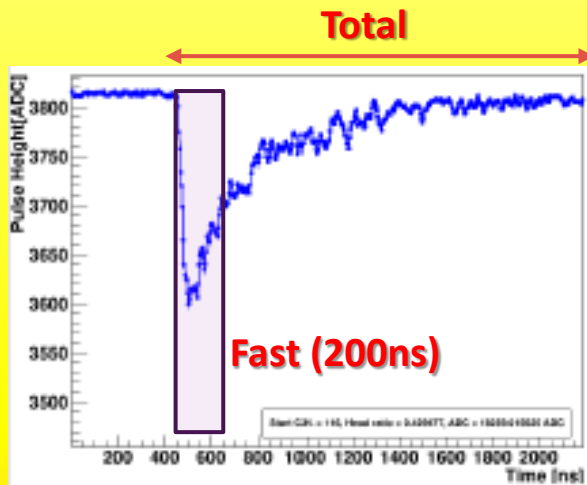
PSD capability of high-purity GAGG

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



- BG measurements carried out in Tsukuba and evaluate the PSD performance of high purity GAGG.

Ratio = Fast (200ns) / Total



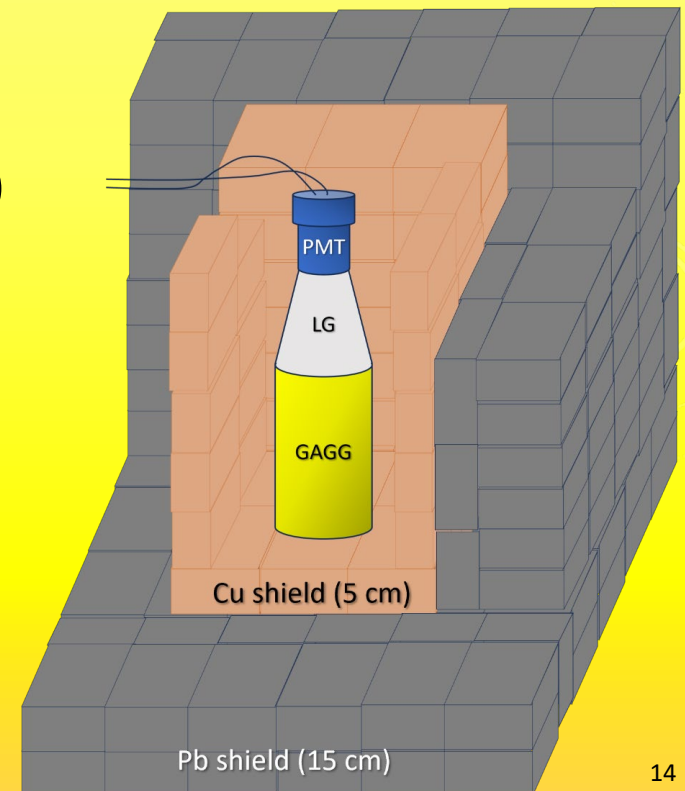
✓ β/α are completely distinguishable above 300 keV !!

BG study 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 estimation and sensitivity study for PIKACHU phase 1.

Date: 2023 6/12 ~ 6/14

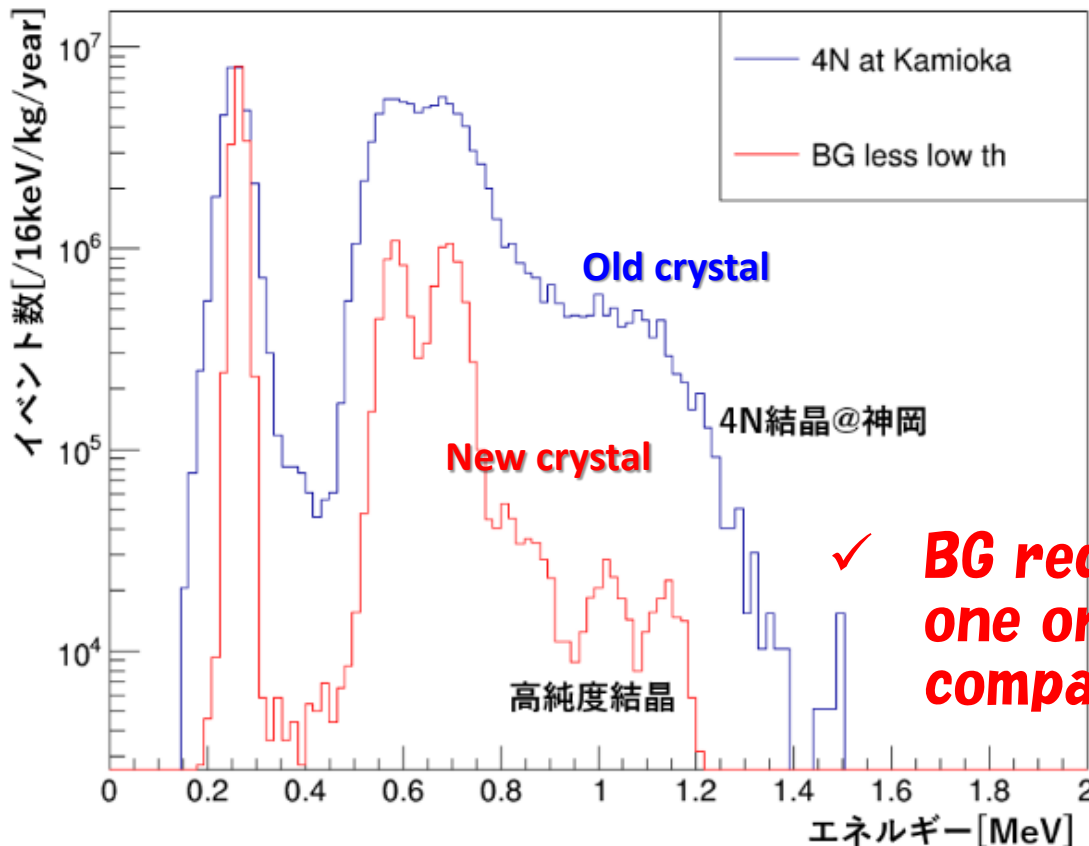
- ✓ Shield : Pb 15cm & Cu 5cm thick
- ✓ HP-GAGG : 5.4 cm ϕ \times 5.2 cmL (0.8 kg)



BG level compared with old crystal

- The radioactive BG levels inside the crystals were estimated from the α -ray rates.

The α -ray spectra selected by PSD

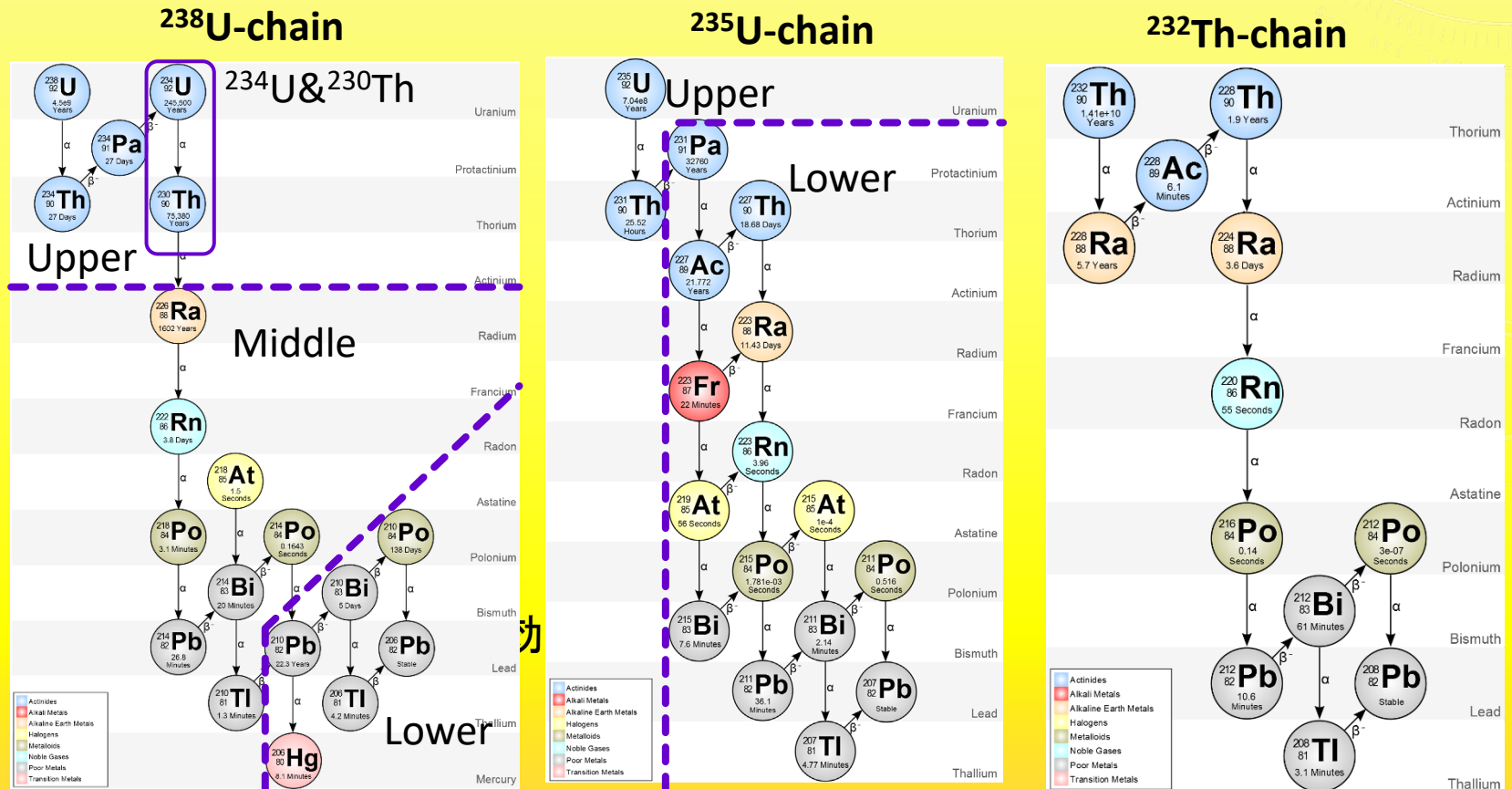


The energy resolution is also improved by changing the crystal composition.

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

BG modelling with GEANT4

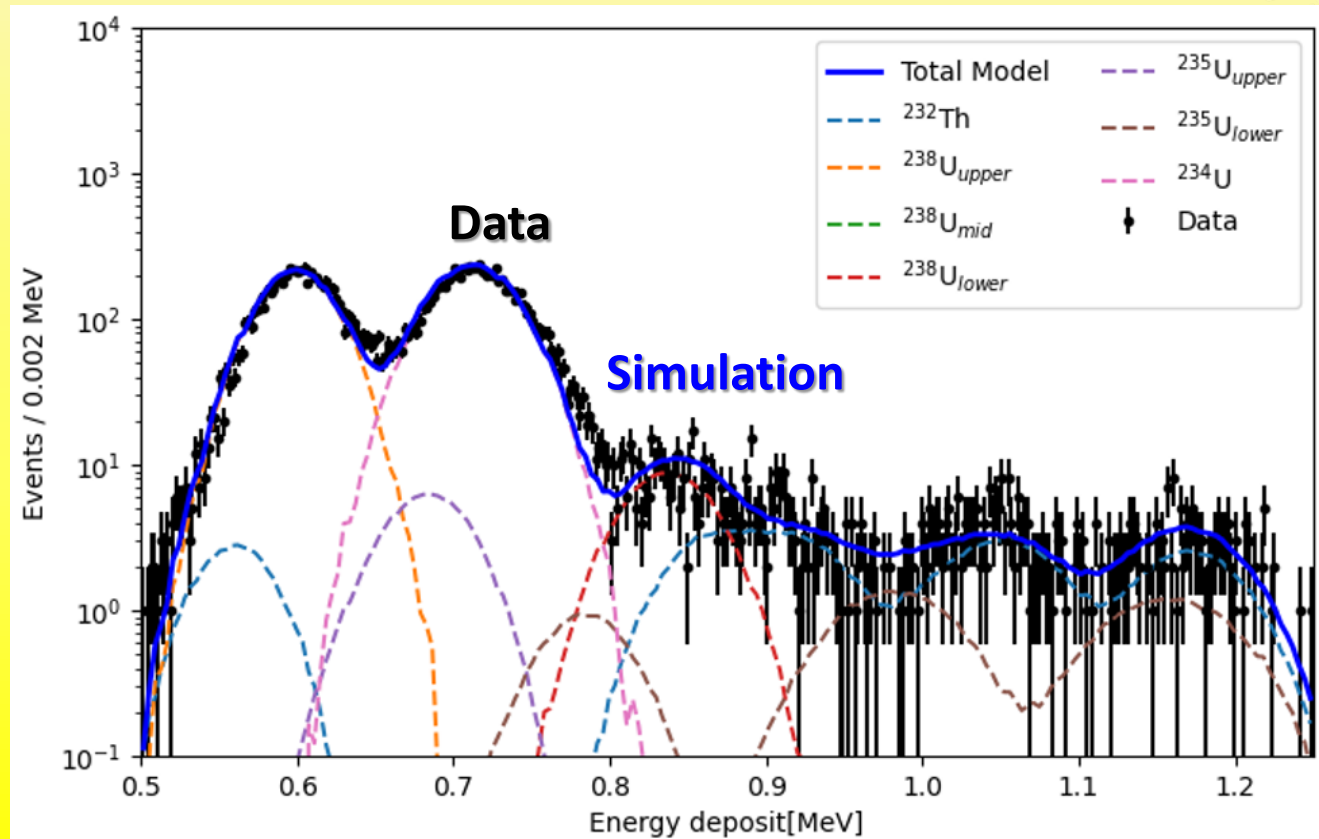
- In order to understand BGs quantitatively, GEANT4 simulation was used.
- Assuming radiative equilibrium below the long-lived ($T_{1/2} > 10$ yr) nucleus, BG energy spectra for ^{238}U , ^{235}U and ^{232}Th were simulated.



Understanding α -ray BG by fitting



- Fitting α spectrum (selected by PSD) with the simulated BG spectra.



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

β -ray BG in high-purity GAGG

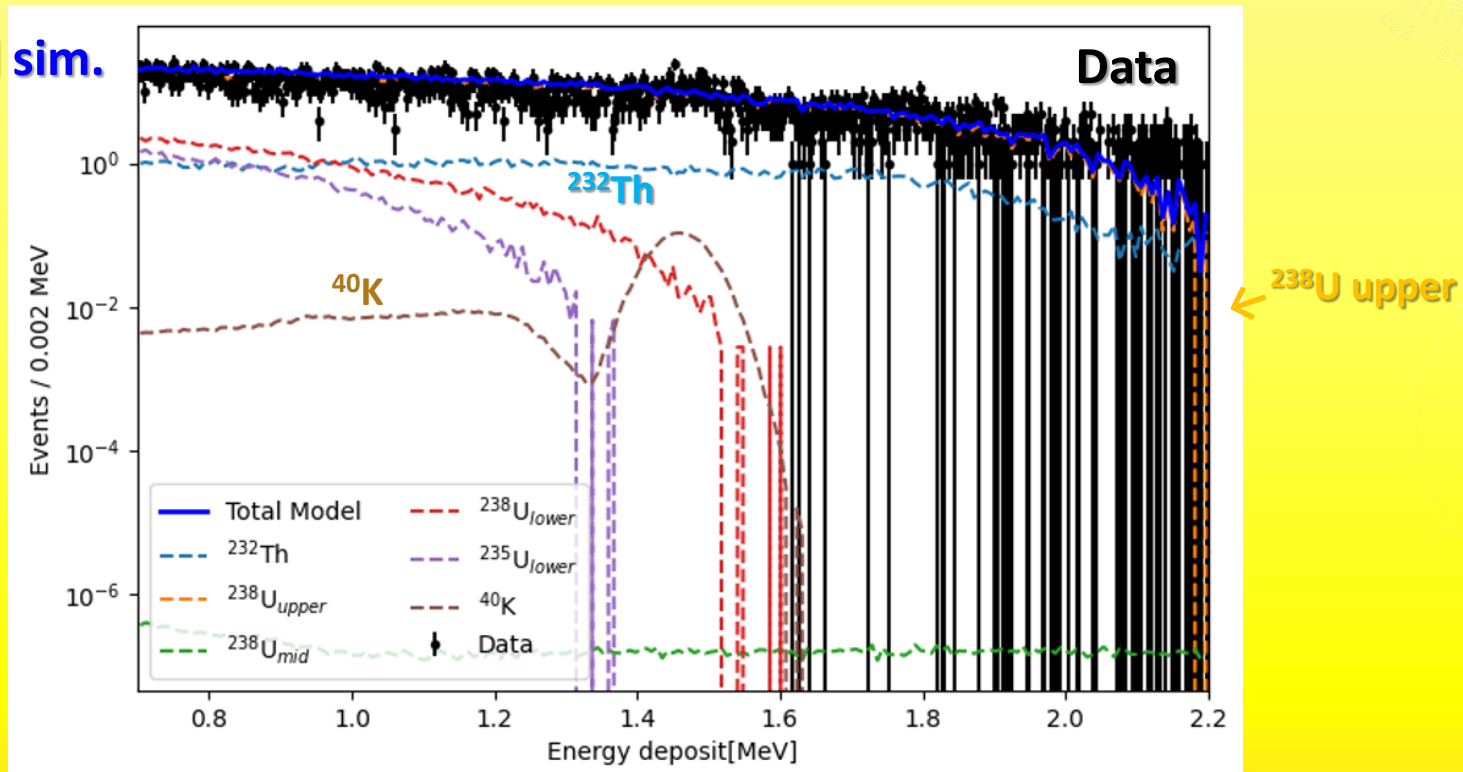
✓ **Data:** β -ray events were extracted from Kamioka data using PSD.

✓ **Simulation:** two β -ray BG models

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

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

Total sim.



✓ **The BG spectrum was successfully reproduced by Geant4!!**

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

Sensitivity study for PIKACHU Phase 1

We estimate **the sensitivity for $0\nu\beta\beta$ search in PIKACHU Phase 1.**

$$T_{1/2}^{0\nu} = (\ln 2) N_a \frac{a}{A} \epsilon \sqrt{\frac{M \cdot t}{BG \cdot \Delta E}}$$

Na: Avogadro number, A: Atomic mass, ϵ : Efficiency,
a: Natural abundance, M: Mass of target nuclei,
t: Live time, BG \cdot ΔE : Background rate in $Q_{\beta\beta}$ region

- ✓ Assuming 2 large crystals (6.4kg), HP-GAGG BG rate and 100% eff.

PIKACHU sensitivity = 4.4×10^{21} years

cf. $T^{0\nu} > 1.3 \times 10^{21}$ years in GSO experiment


- ✓ Experiment will start this 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 Phase 2.

First paper of PIKACHU



JOURNAL ARTICLE ACCEPTED MANUSCRIPT

First Study of the PIKACHU Project: Development and Evaluation of High-Purity $\text{Gd}_3\text{Ga}_3\text{Al}_2\text{O}_{12}:\text{Ce}$ Crystals for ^{160}Gd Double Beta Decay Search

Takumi Omori, Takashi Iida , Azusa Gando, Keishi Hosokawa, Kei Kamada, Keita Mizukoshi, Yasuhiro Shoji, Masao Yoshino, Ken-Ichi Fushimi, Hisanori Suzuki ...

Show more

Progress of Theoretical and Experimental Physics, ptae026,
<https://doi.org/10.1093/ptep/ptae026>

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Latest Issue

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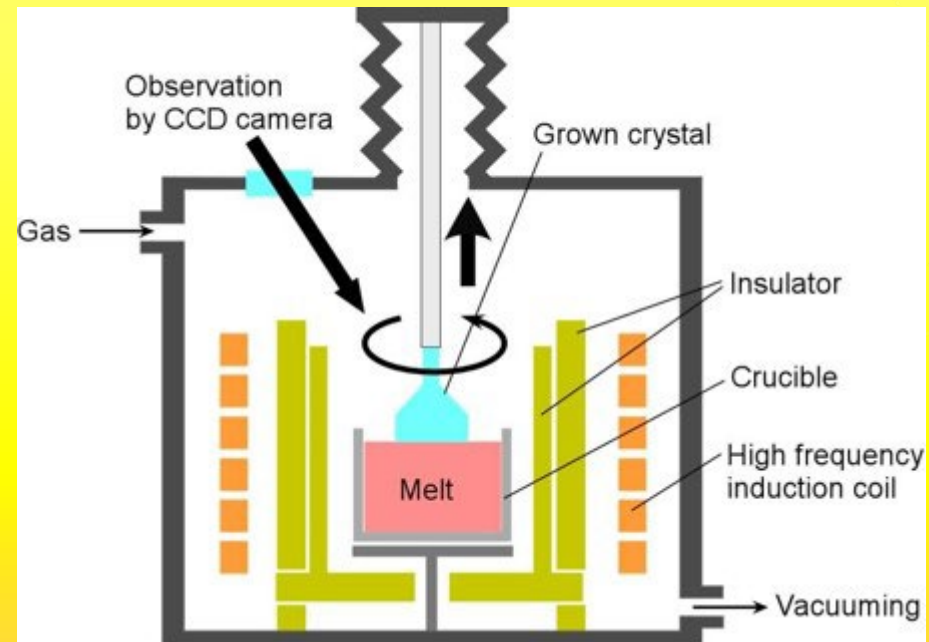
Further purification of GAGG for Phase 2

- The crystal raw materials are of higher purity than the crystals, which suggests that **U/Th impurities were contaminated 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.**

Comparison of U/Th impurities

	^{238}U	^{232}Th
Original GAGG	911 ± 10	289 ± 20
HP-GAGG	125 ± 2	10.2 ± 0.4
Gd₂O₃ raw material	< 16.3	1.66 ± 0.41

[mBq/kg]



Future prospect

- **PIKACHU experiment Phase 1 will start in 2024.**
- ^{160}Gd $0\nu\beta\beta$ search sensitivity will be updated in two years.
- In parallel with Phase 1 PIKACHU, we develop **ultra-high-purity GAGG crystal**, whose purity is one more order of magnitude better than current HP-GAGG.
- Start Phase 2 PIKACHU after 2026 with a few tens of UHP-GAGG for the first detection of ^{160}Gd $2\nu\beta\beta$.

We aim to detect ^{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!! $BG \sim 1/10$ is achieved.
 - ✓ Sensitivity of PIKACHU Phase 1 will be higher than that of the previous study .

New collaborator is always welcome!!

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