Searching for Exotic Dark Matter Searches using Radioactive Source at Underground
Lecture-2 : GRANDE

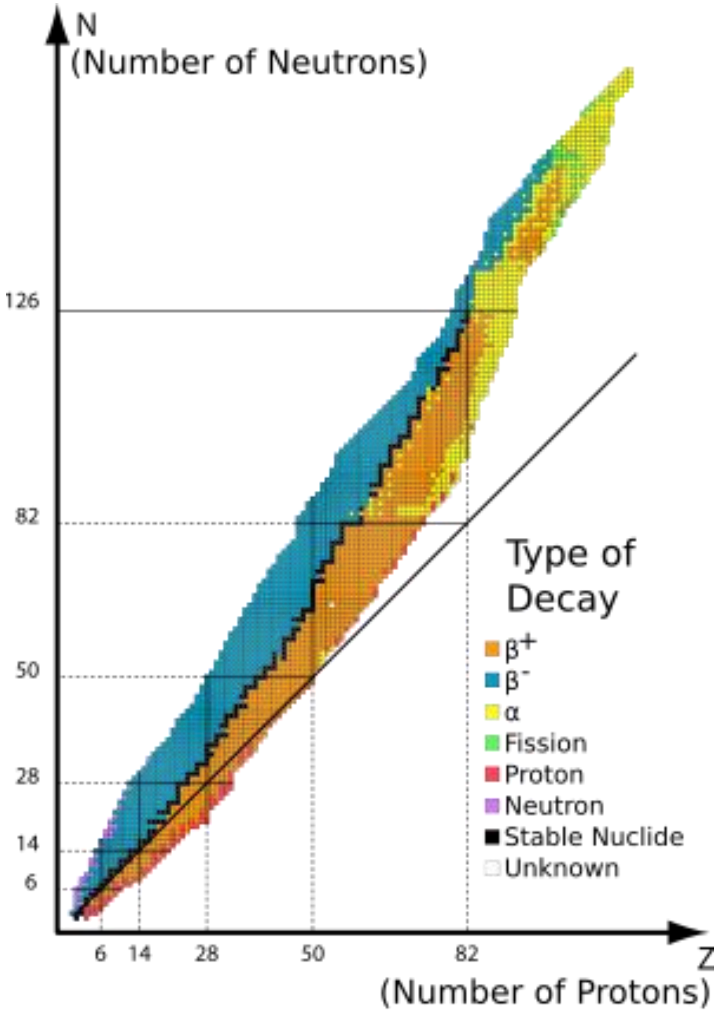
H.J.Kim (Kyungpook National Univ.)

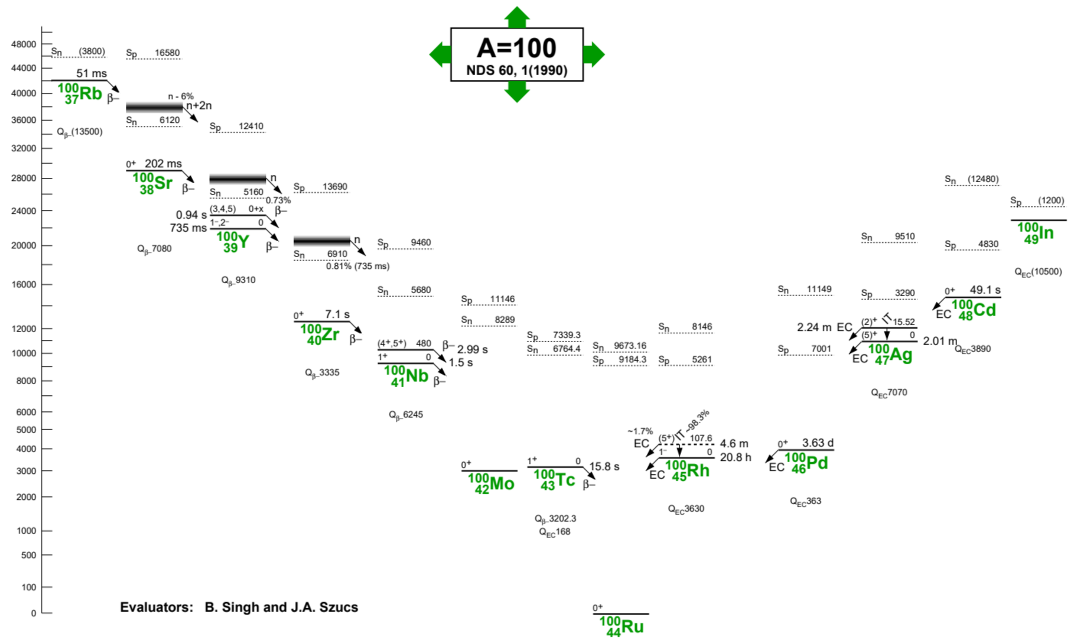
CNPR Workshop 2025: Neutrinos and Physics beyond the Standard Model

Oct 24-27, Chonnam National University

Nuclear isotopes

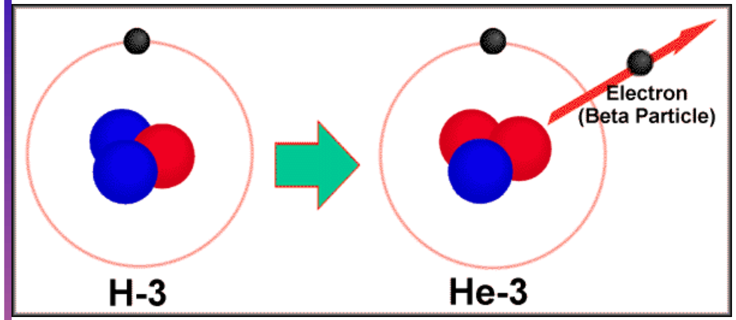
Known isotope: 1850
Stable isotope: 280





β decay - three types

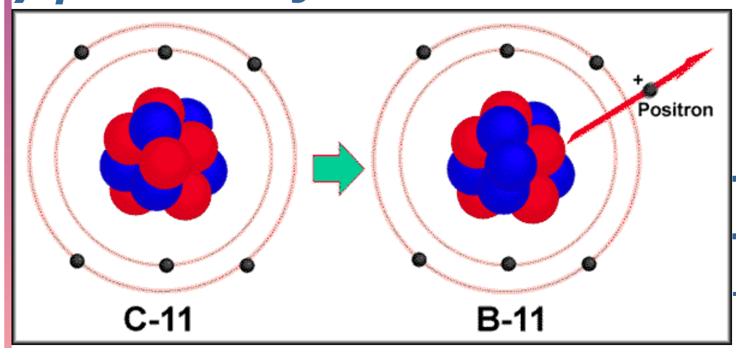
1) β- decay



$$^{3}_{1}H \xrightarrow{\beta} {}^{3}_{2}He + e^{-} + \overline{\upsilon}_{e}$$

- converts one neutron into a proton and electron
- no change of A, but different element
- release of anti-neutrino (no charge, no mass)

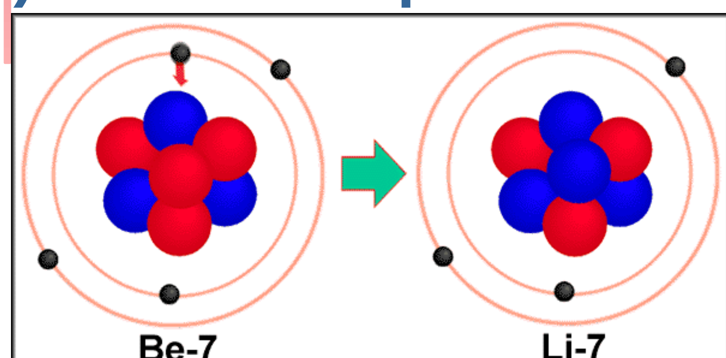
2) β+ decay



$$^{11}_{6}C \xrightarrow{\beta^{+}} {}^{11}_{5}B + e^{+} + \upsilon_{e}$$

- converts one proton into a neutron and electron
- no change of A, but different element
-release of neutrino

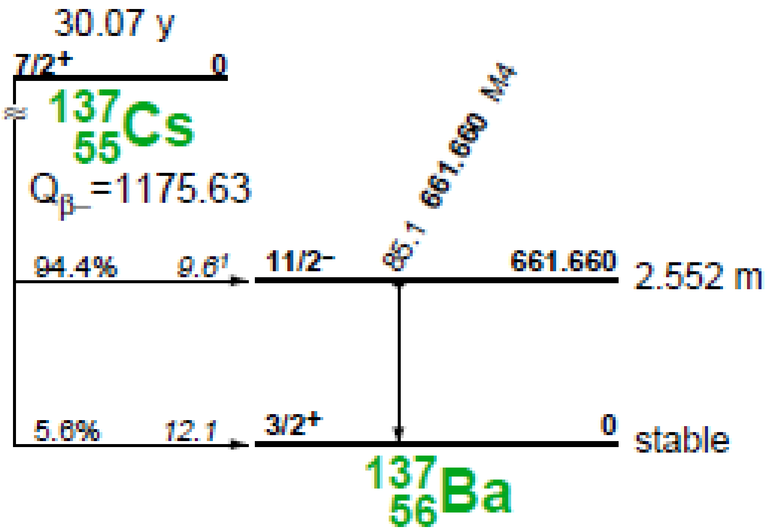
3) Electron capture



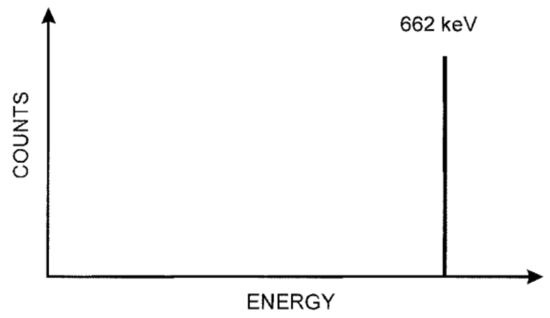
$$^{7}_{4}Be + e^{-} \xrightarrow{EC} {}^{7}_{3}B + \upsilon_{e}$$

- three body decay -> continuous spectrum

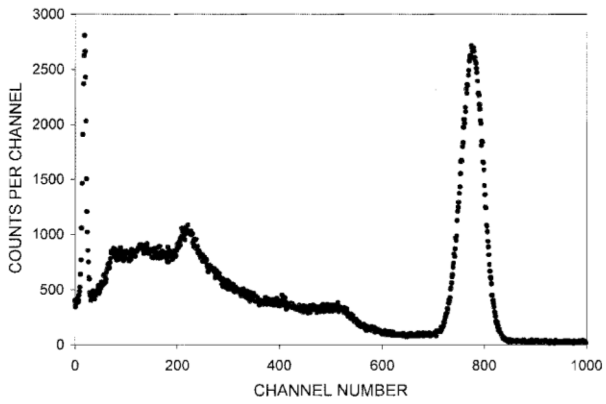
Lab. Sources : Decay scheme of ^{137}Cs



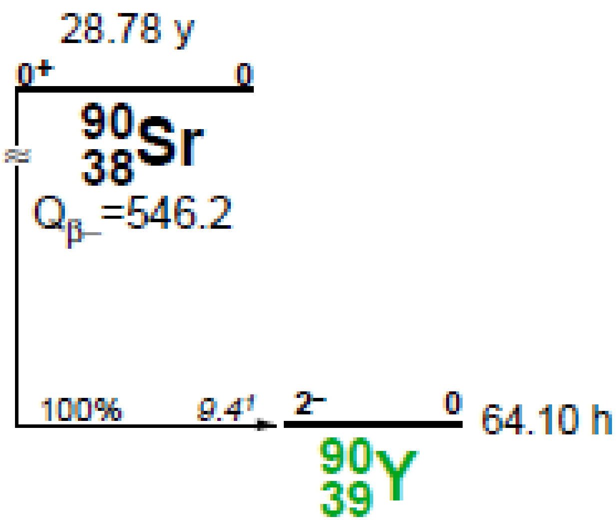
Based on the level diagram, what do we expect to see?

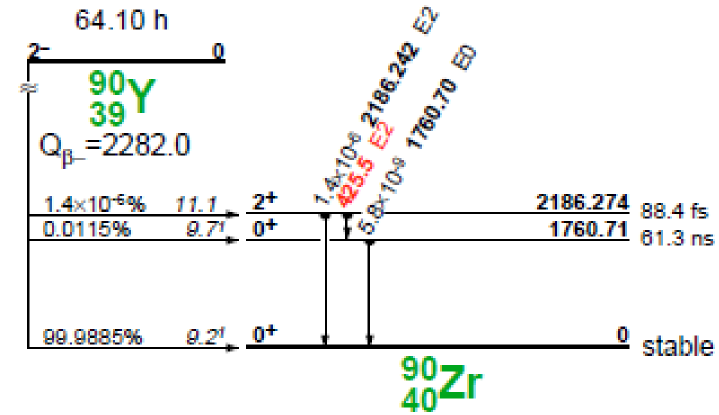


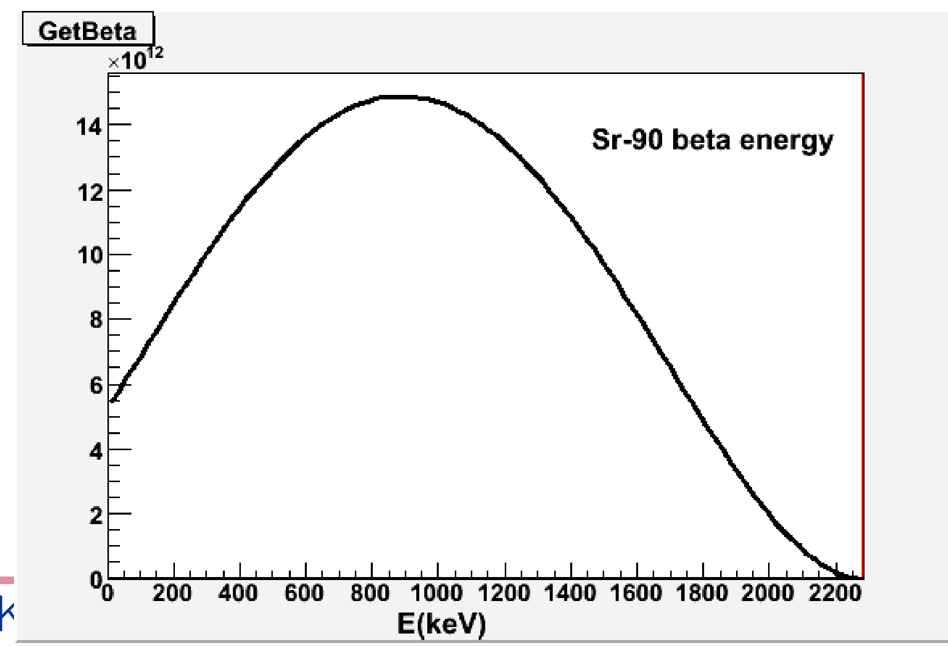
easured spectrum



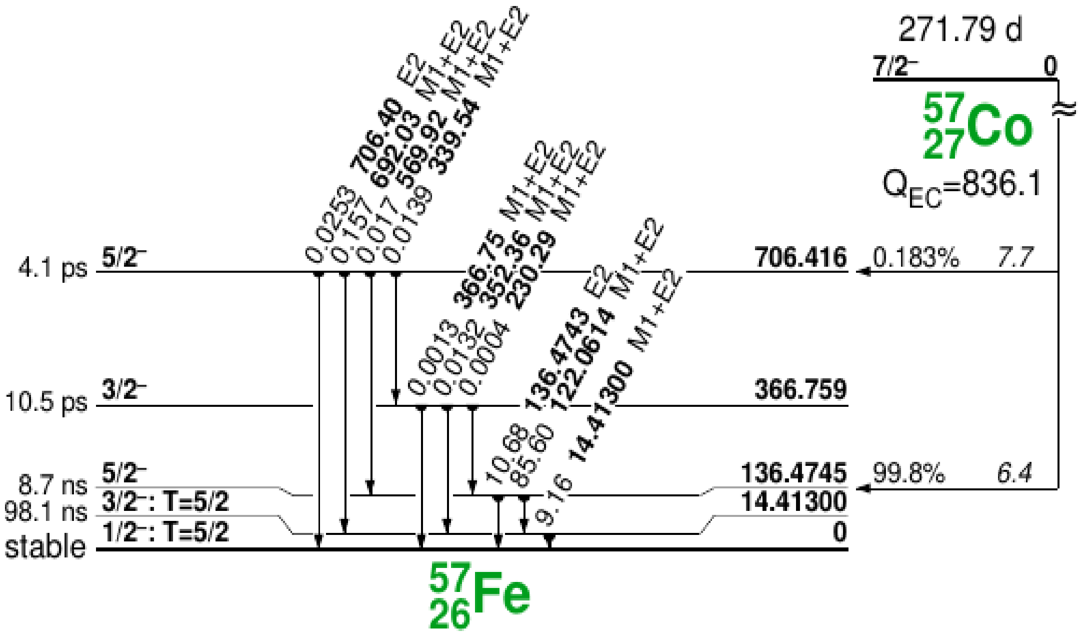
Decay Scheme for Sr-90







Decay Scheme for Co-57



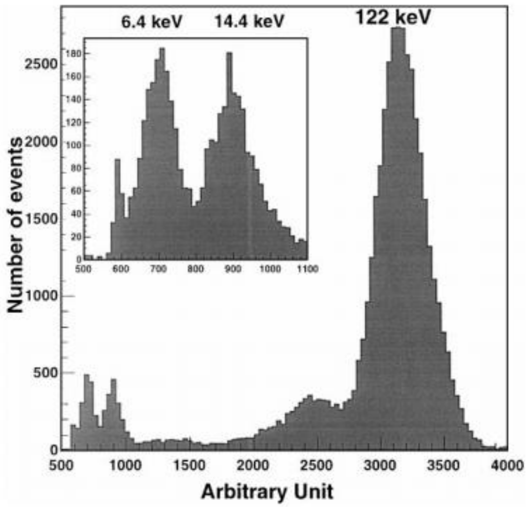


Fig. 2. Pulse height spectrum of CsI(Tℓ) for ^{57}Co source. The top left plot is a zoomed pulse height spectrum of the low energy X-ray.

H.J. Kim et al. / Nuclear Instruments and Methods in Physics Research A 457 (2001) 471–475

Selection rules for γ transition

- The photon is a spin−1 vector boson
- For orbital angular momentum, we can have values $\ell = 0,1,2,3,\cdots$ that correspond to our multipolarity.
- Therefore, our selection rule is:

$$|J_i - J_f| \le \ell \le |J_i + J_f|$$

- Electromagnetic transitions:

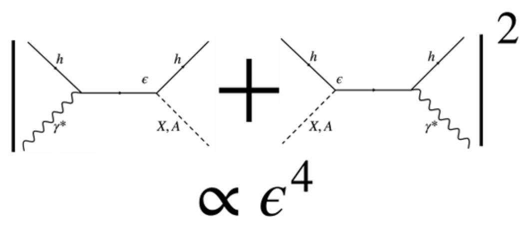
$$\Delta\pi\ (electric) = (-1)^{\ell}$$

$$\Delta\pi\ (magnetic) = (-1)^{\ell+1}$$

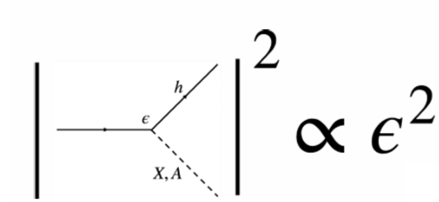
$\Delta\pi$	*yes*	E1	M2	E3	M4
	no	M1	E2	M3	E4

- Relation with E, M transition to particle type
  E0-> Scalar boson, E1 -> Vector boson, E2 -> Tensor boson
  M1-> Psudoscalar boson, M2 -> Axial Vector boson

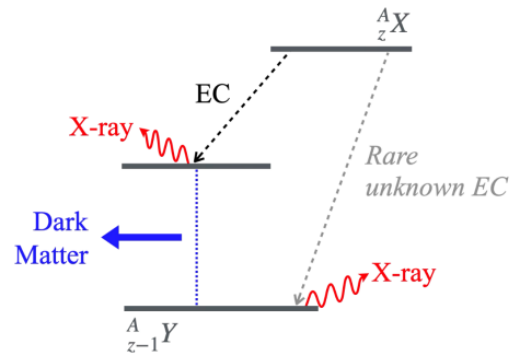
Radioactive sources for Dark Sector Search



Appearance Exp.
Accelerator, Reactor
~10^{24} POT
→ ε ~10^{-6}



Disappearance Exp.
Radioactive source
~$10^{12\text{-}14}$ Decay
→ ε ~$10^{-6\sim7}$



Axion-like particle or dark photon searches with M1 and E1, E2 transitions of nuclear decay

Advantage

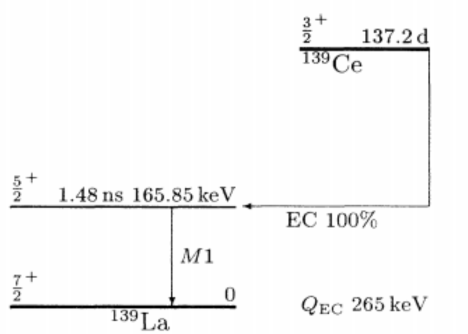
- ❑ Decay process: Coupling is proportional to ε^2 not ε^4 (Accelerator, Reactor)
- ❑ Tabletop-scale experiment (much lower cost)
- ❑ Source-detector technique (radioactive doping in fast scintillator)
- ❑ Time-delayed coincidence method to eliminate backgrounds in the case of isomeric states
- ❑ Underground experiment with Cu, Pb shielding

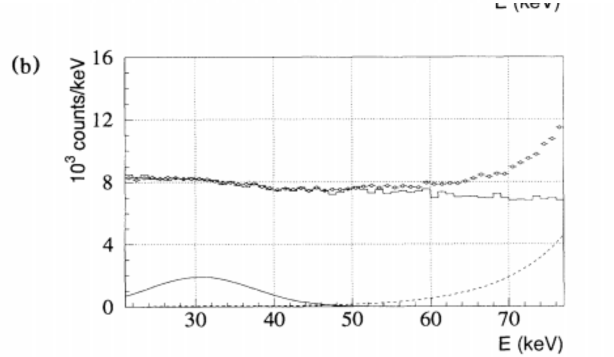
Limitation

- ❑ Low mass only (typically =< 1 MeV due to detector costs)
- ❑ Activity limitations

Tabletop experiments with radioactive source?

- Background is too high





$\Gamma_a/\Gamma_\gamma < 1.21 \times 10^{-6}$ at the 95%

Mass of hadronic axion > 26.7 keV by 95%CL
M.Minowa et al, Phys. Rev. Lett. 71, 4120

- Background is the serious issue
- Detector is too thin for ~ MeV gamma

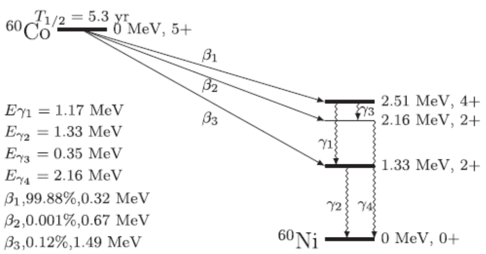
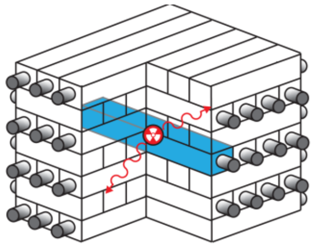
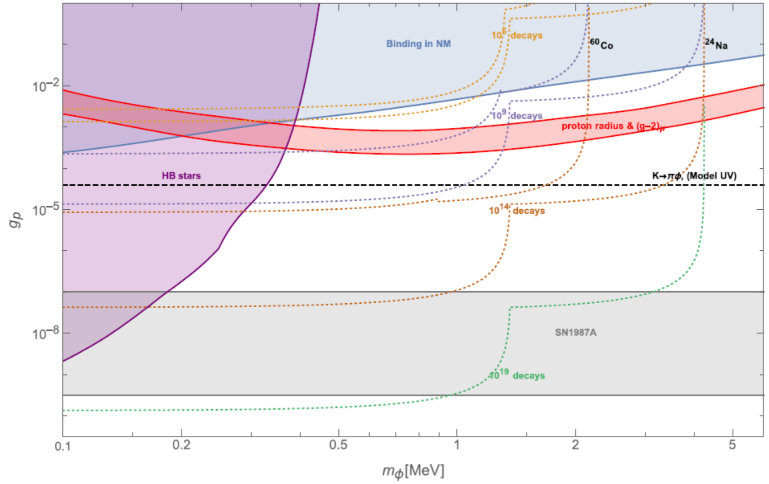


FIG. 2. Decay scheme of ^{60}Co.

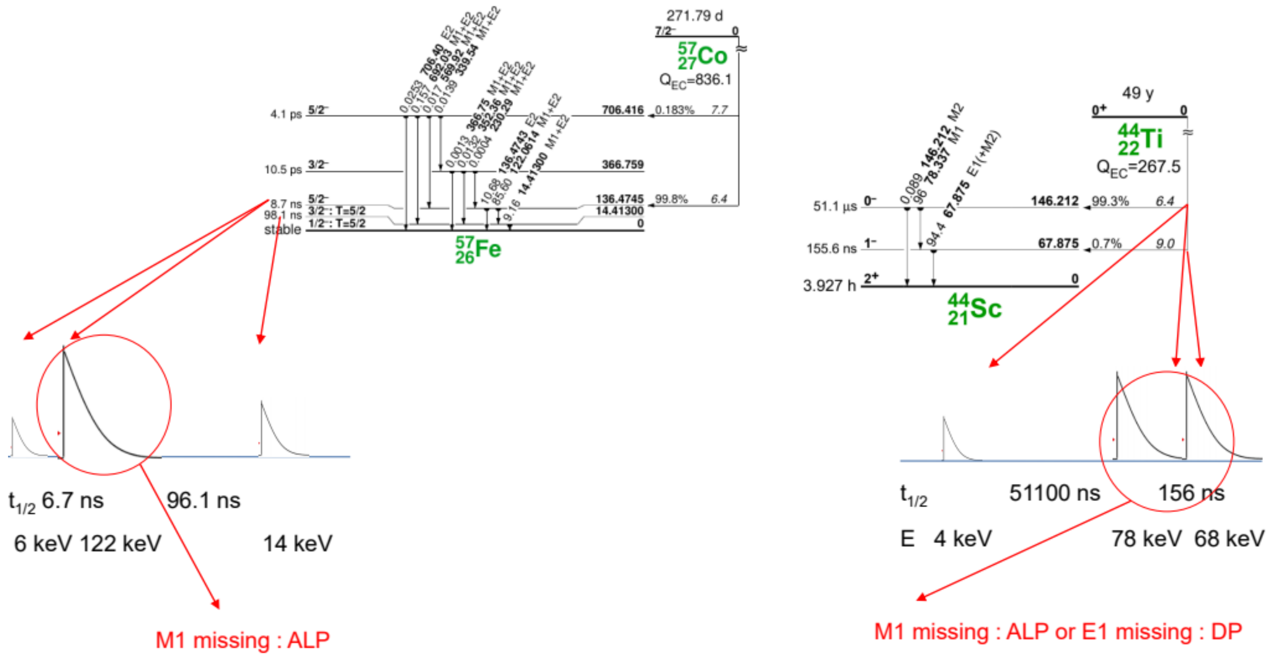


INVISIBLE DECAY MODES IN NUCLEAR GAMMA CASCADES PHYS. REV. D **99**, 035025 (2019)



Other table top experiments with radioactive source?

- ALP or DP search with M1 and E1 transition with KAPAE setup + fast scin.
- Time delay coincidence method to remove backgrounds.
- For the zero background experiment, we propose this experiment at Yemi Under ground lab. with branching fraction sensitivity of 10^{-12}



When experimentalist meets theorist

PHYSICAL REVIEW LETTERS **133,** 232501 (2024)

Constraints for Rare Electron-Capture Decays Mimicking Detection of Dark-Matter Particles in Nuclear Transitions

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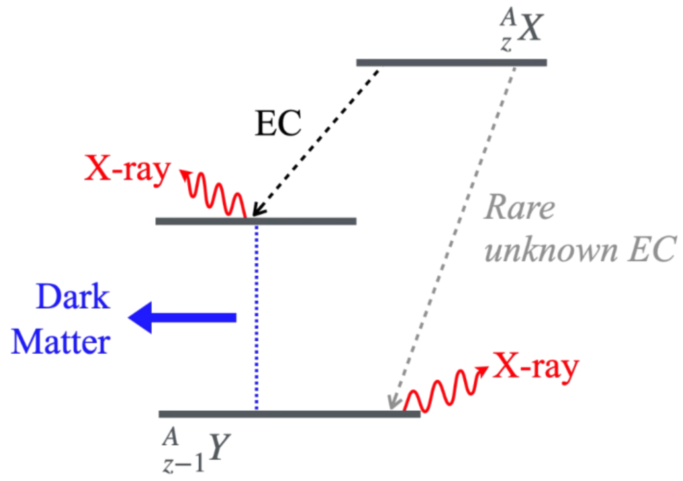
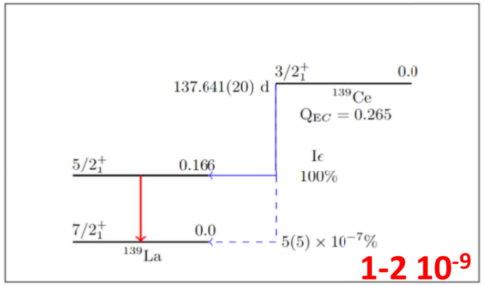


Major possibilities for dark matter particle searches through EC transitions



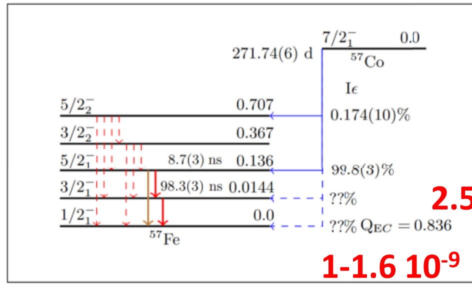
#1

M1 $5/2^+_1 \rightarrow 7/2^+_1$: 165.86 keV

→ K_α X-ray: ~33 keV

Easiest case

Search for single X-ray



2.5-3.3 10^{-8}

#1

M1 $5/2^-_1 \rightarrow 3/2^-_1$: 122.06 keV

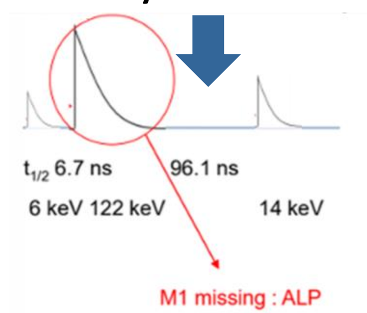
→ M1 $3/2^-_1 \rightarrow 1/2^-_1$: 14.4 keV

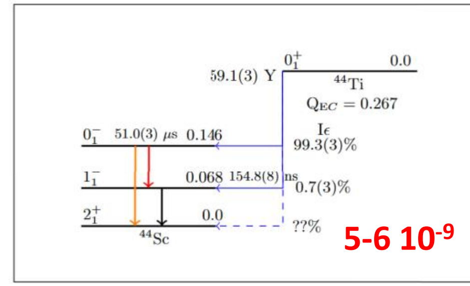
→ K X-ray: ~6 keV

#2

E2 $5/2^-_1 \rightarrow 1/2^-_1$: 136.47 keV

→ K X-ray: ~6 keV





#1

M1 $0^-_1 \rightarrow 1^-_1$: 78.33 keV

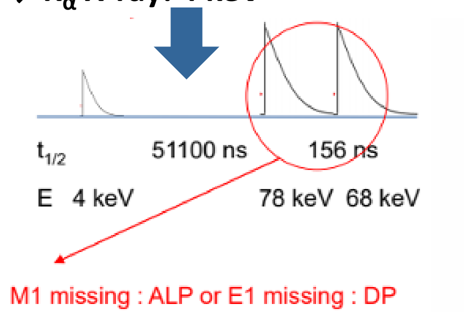
→ E1 $1^-_1 \rightarrow 2^+_1$: 67.87 keV

→ K_α X-ray: 4 keV

#2

M2 $0^-_1 \rightarrow 2^+_1$: 146.212 keV

→ K_α X-ray: 4 keV



GRANDE

(Global Rare Anomalous Nuclear Decay Experiment)

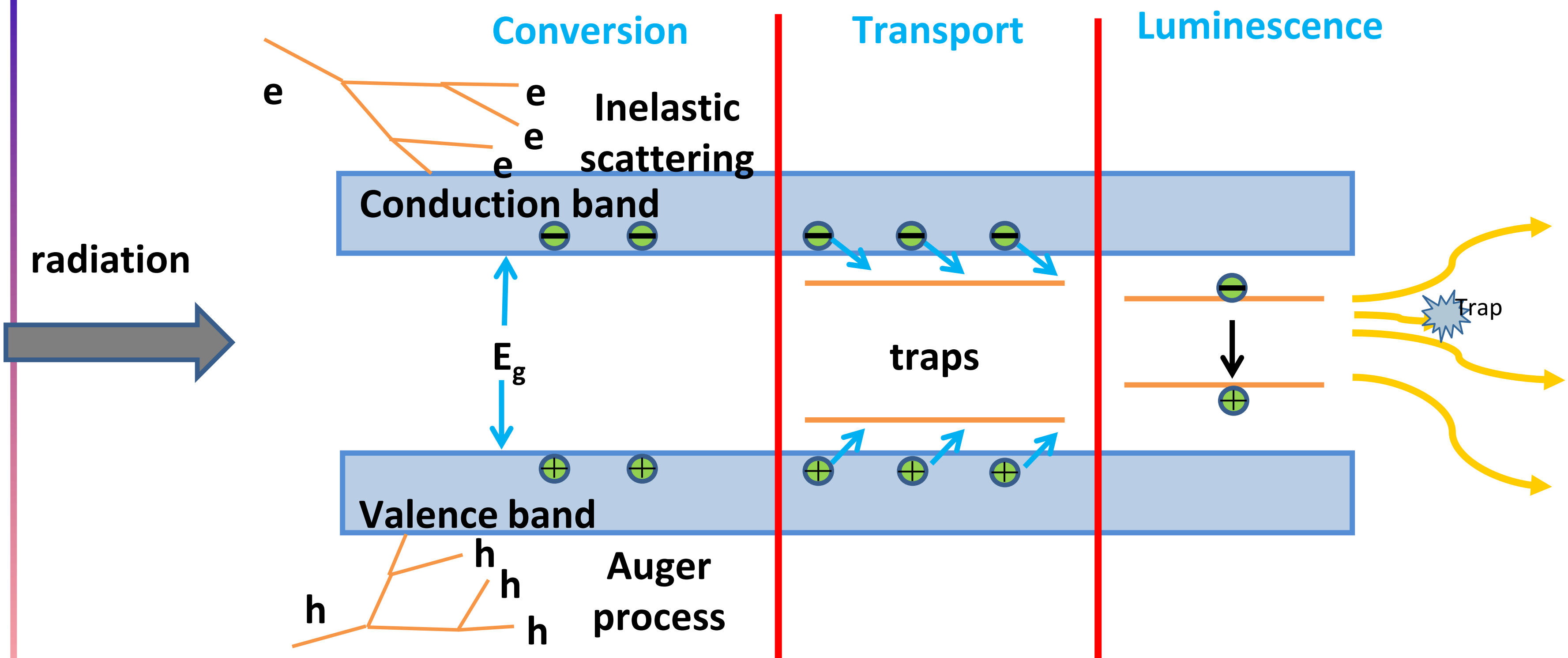
GRANDE Collaboration (5 countries, 8 institution, 14 members)

- H.J. Kim, N.T. Luan, D.W. Jeong, J.Y. Lee, N.D. Ton (Kyungpook National University, KOREA)
- M.H. Lee, E.K. Lee (Center for Underground Physics, IBS, KOREA)
- J.H. So (Yemi operation center, IBS, KOREA)
- J. Suhonen, A. Agnihotri (University of Jyväskylä, Finland)
- I. Gkialas (University of the Aegean, Greece)
- J. Kaewkhao (Nakhon Pathom Rajabhat University, Thailand)
- S. Kothan (Chiangmai University, Thailand)
- T. D. Trong (Institute of Physics, Vietnam)

Scope of the GRANDE

- ❑ Experiment setup at the Yemi Underground Lab with low-background Cu+Pb shielding
- ❑ Radioactive source embedded in a crystal scintillator
- ❑ 4π VETO with BGO
- ❑ Aiming for extremely low background condition
- ❑ Measurement of Rare EC process, rare beta, and alpha decay and searching for new particles.

Scintillation light yield fundamentals



Incident E	e-h eff.	Transfer eff.	Quantum eff.	Transmission eff.
E	1/β	S	Q	T

$$N_{ph} = S \times Q \times T \times N_{eh} = S \times Q \times T \times E/\beta E_g \, (N_{eh} = E/\beta E_g)$$

The best case S, Q, T=1 and β=2.5 : $N_{ph} = E/(2.5 \times E_g)$

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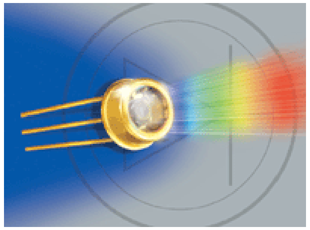
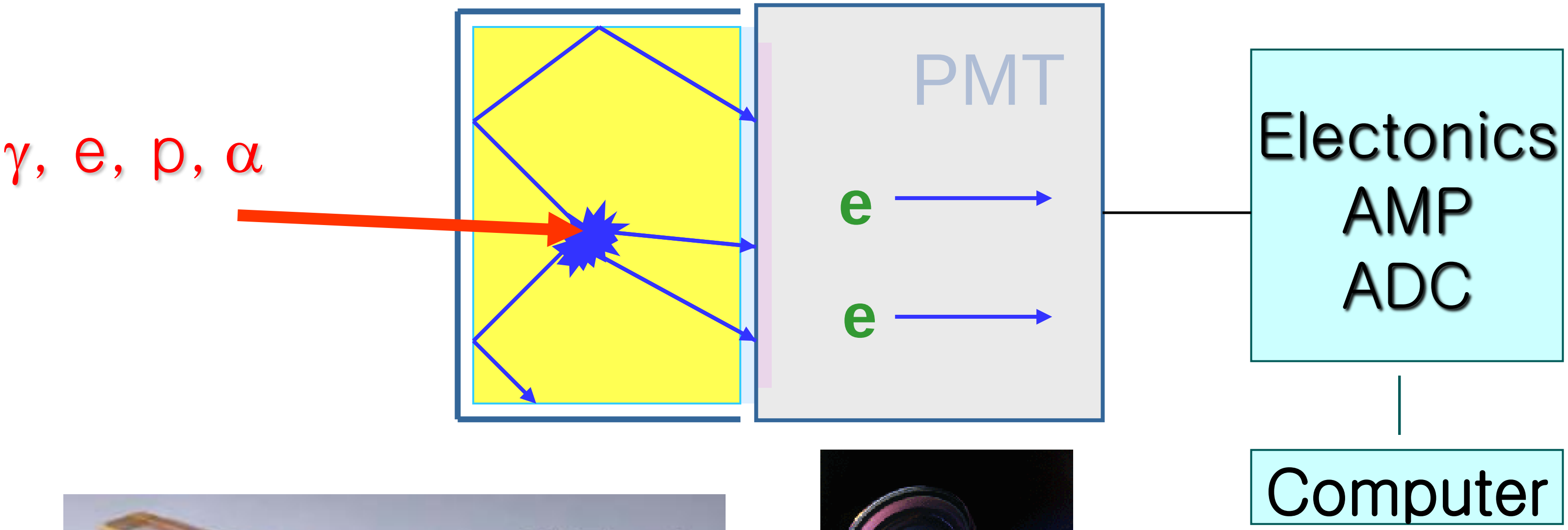
Principle of Radiation detection with Scintillator

Scintillator



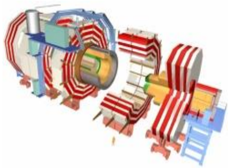
Photosensor

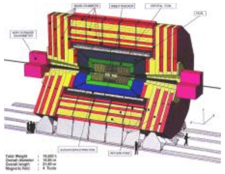


Application of Scintillators

Medical application

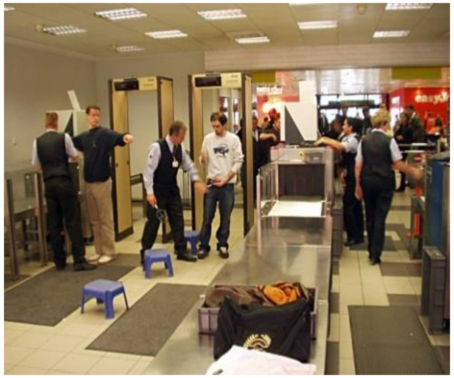
High energy physics





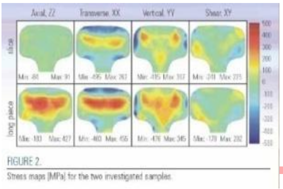
Astro-particle physics, ...

Security check

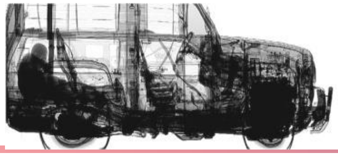


X-ray scanning

Nondestructive analysis



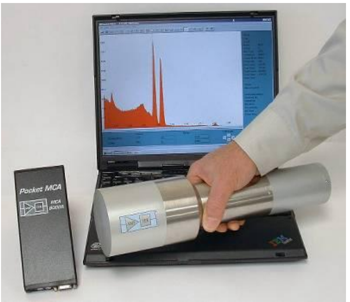
Board inspection



Radiation monitoring

Scintillation Detectors

NaI(Tl) Spectrometer



Scintillation Surveymeter

Scintillator Development Direction

Invited Article

The quest for high resolution γ-ray scintillators

Pieter Dorenbos

Optical Materials: X 1 (2019) 100021

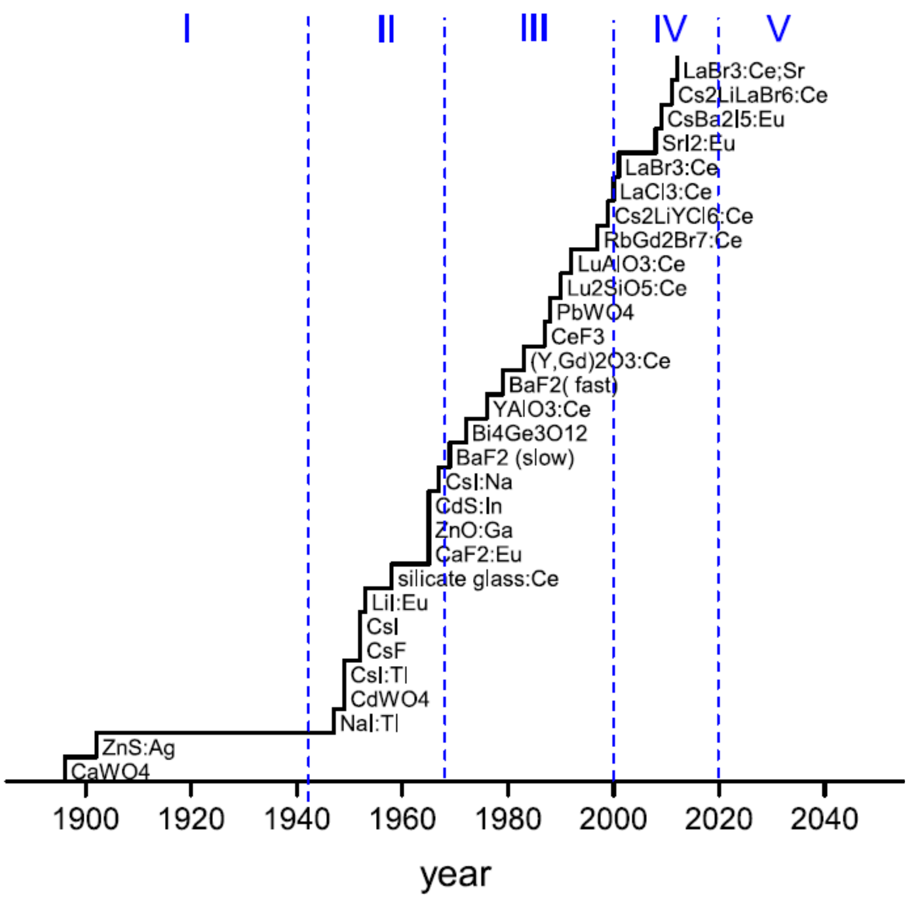


Fig. 2. History of scintillator discovery that distinguishes phases I to IV and phase V for future discoveries.

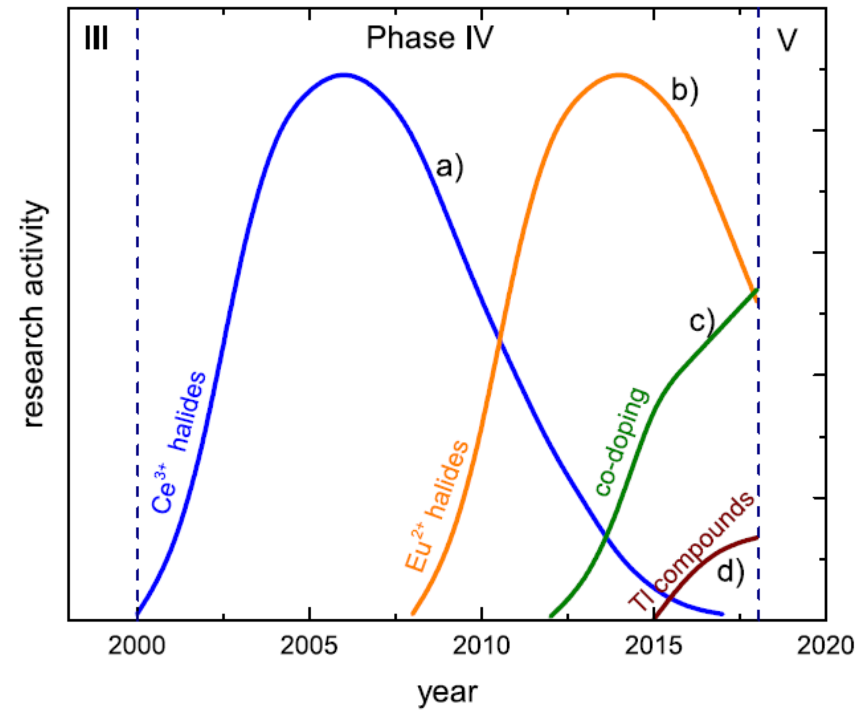
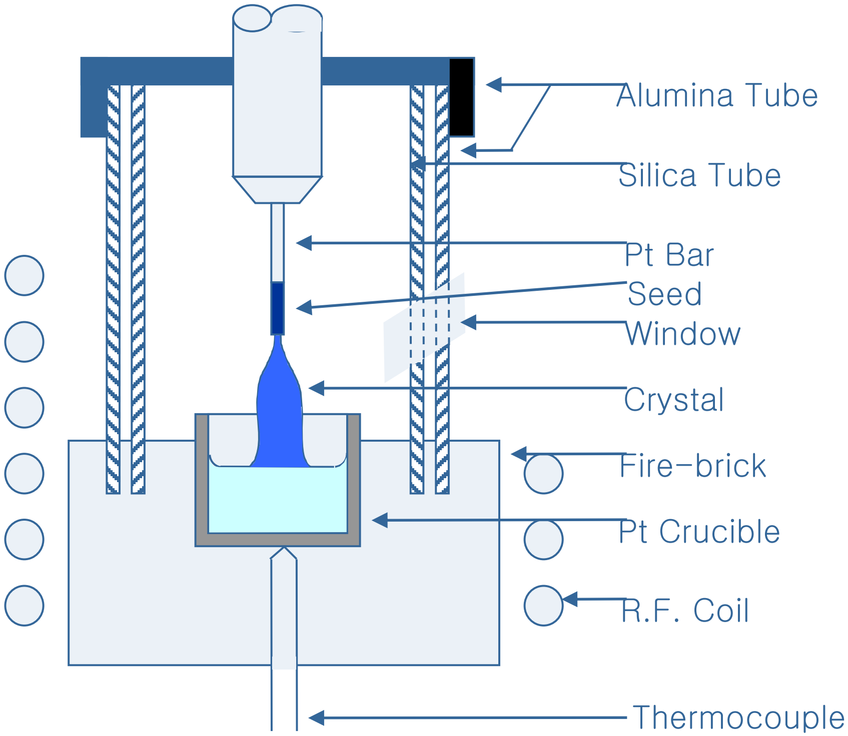
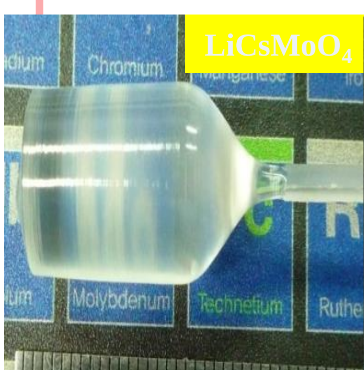


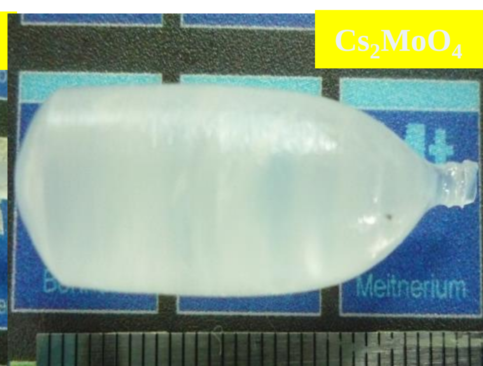
Fig. 4. Main lines of research during phase IV of scintillator discovery. a) The research on Ce^{3+} activated halide compounds inspired by the discovery of $LaBr_3$:Ce^{3+}, b) the research on Eu^{2+} doped halide compounds initiated by the re-discovery of SrI_2;Eu^{2+}, c) the increased research on co-doped scintillators stimulated by discovery of Sr^{2+} co-doped $LaBr_3$:Ce^{3+}, d) the research on Ce^{3+} activated Tl-based compounds initiated by $Tl_2LiGdCl_6$.

Crystal growing system with Czochalski method

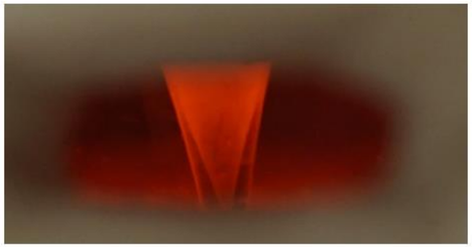


At KNU

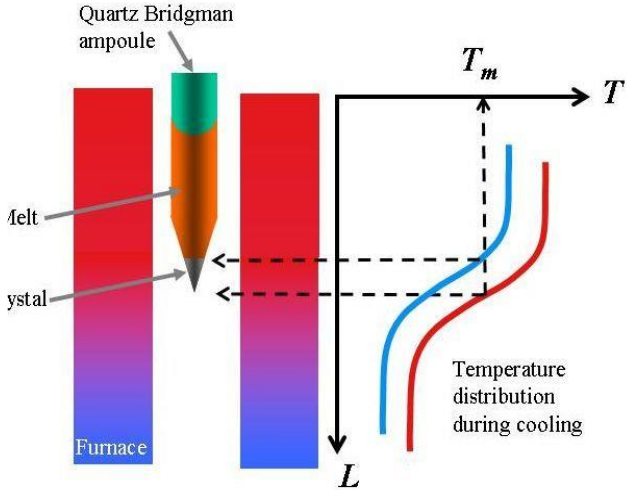




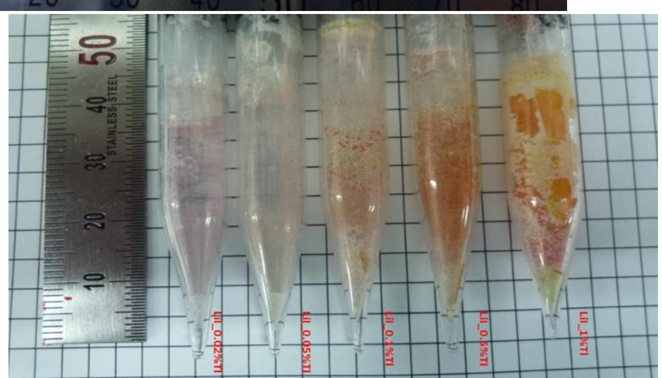
Bridgman Crystal Growing Methods



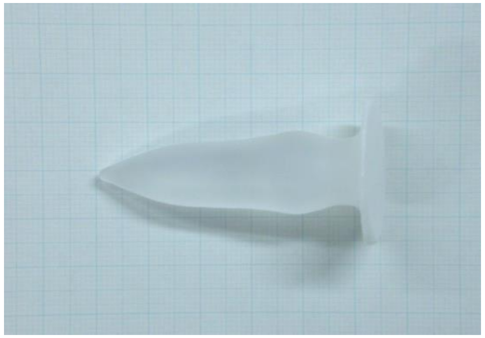
Crystal is growing in Bridgman furnace



At KNU



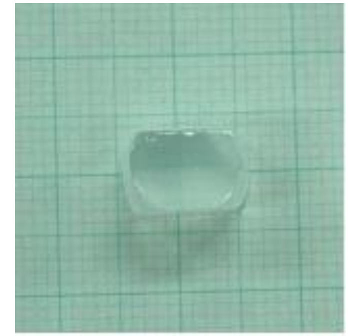
Crystals growing research at KNU over 25 years



$SrCl_2(Eu)$

$CeCl_3$

$LaCl_3(Ce)$

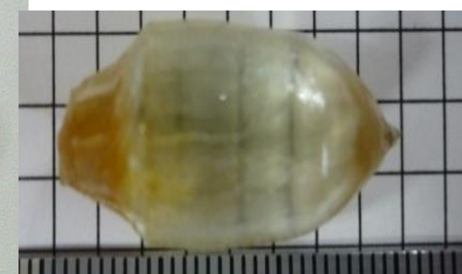


$CeBr_3$

GAGG:Ce

$Ba_{0.2}Sr_{0.8}Cl_2$

$PbCl_2:Eu^{2+}$



BGSO

- NaI:Tl, CsI(Tl, Co3, Na), BGO, BSO, BGSO, SrWO4, CaMoO4, SrMoO4 et al.
- New material : BaSrCl2, CsCe2Cl7, Cs(Rb)2Li(Na)CeCl6, Cs2LiGd(Lu)Cl(Br)6:Ce, Li6Lu(Gd,Y)(Bo3)3, NaGd(Wo4)3, LiBaF3, ZnMnTe, Tl-based scintillators et al.

H.J.Kim, KNU

AMoRE-II: Mo crystals grown and tested

ORIGINAL PAPER

CRYSTAL Research&Technology

www.crt-journal.org

Search for New Molybdenum-Based Crystal Scintillators for the Neutrino-Less Double Beta Decay Search Experiment

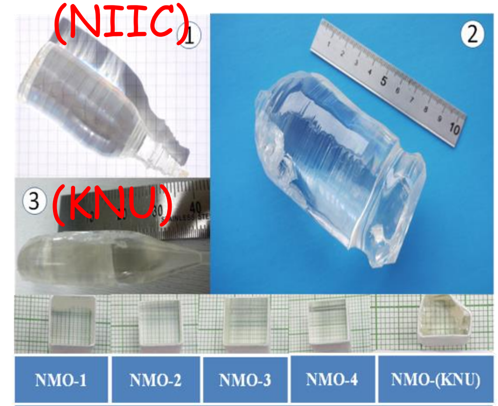
HongJoo Kim, Indra Raj Pandey, Arshad Khan, Ju-Kyung Son, Moo Hyun Lee,* and Yeongduk Kim*

O (CUP)

NMO (KNU)

Ø40 mm X 100 mm

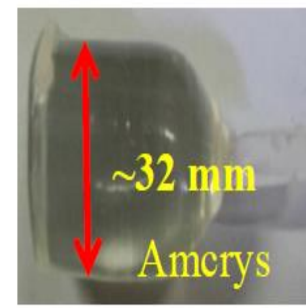
(NIIC)



PMO (NIIC)

LMO (KNU)

AMCRYS



CMO (NIIC)

CMO (CARAT)

LMO (CUP)

NaI:Tl growing with Bridgeman at KNU

Growth crystals for COSINE 200

Motivation

Bridgman

Czochralski

Method

Sticky to the ampoule's wall

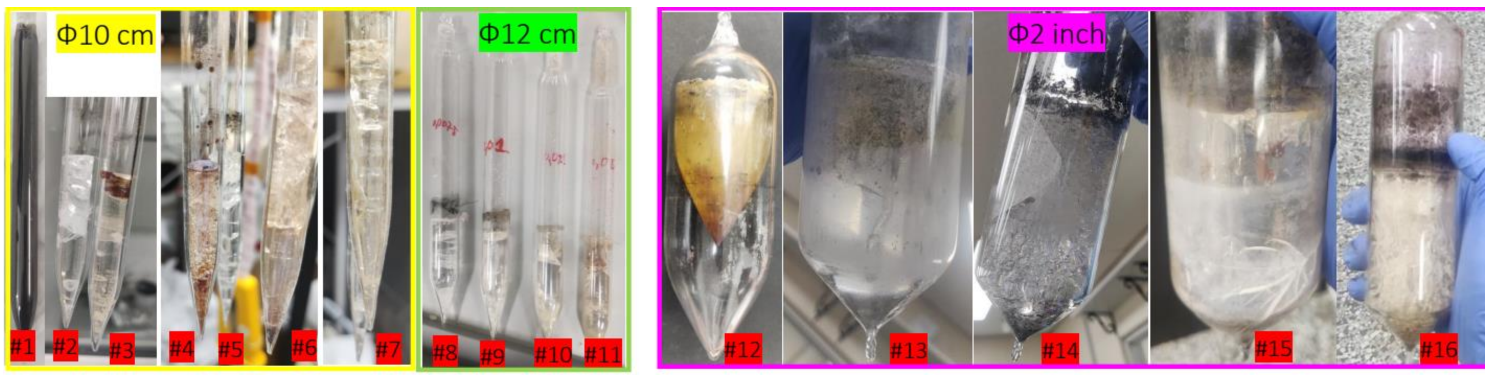
Bubbles

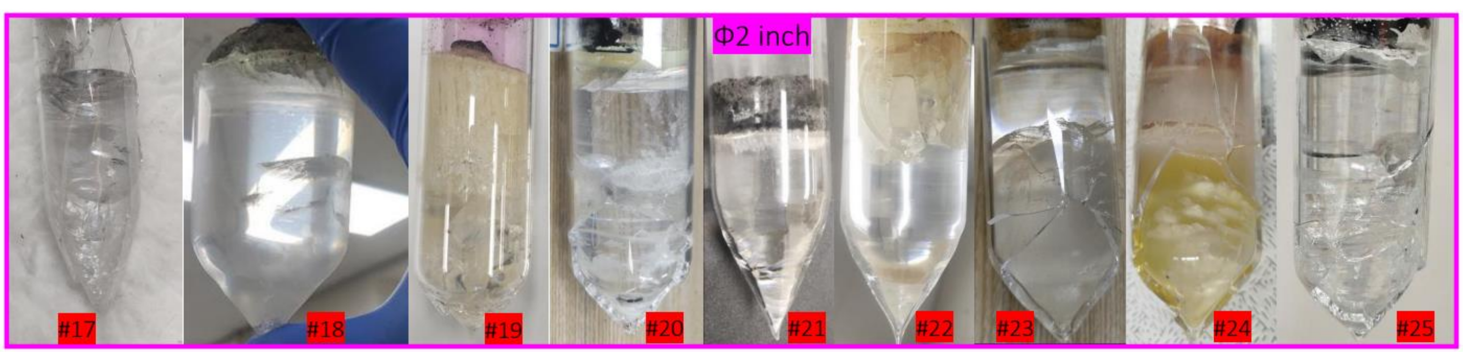
Problems

C coating, $\mathbf{NH_4}\boldsymbol{I}$, $SiCl_4$, $\mathrm{SiI_4}$, …

…

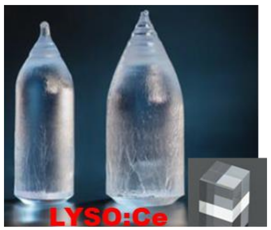
Treatments





Intrinsic radioactivity in crystal scintillators : Con

Medical Imaging : PET



^{176}Lu (2.4%) : 300 Bq/cm^3

Space mission : γ-spectroscopy

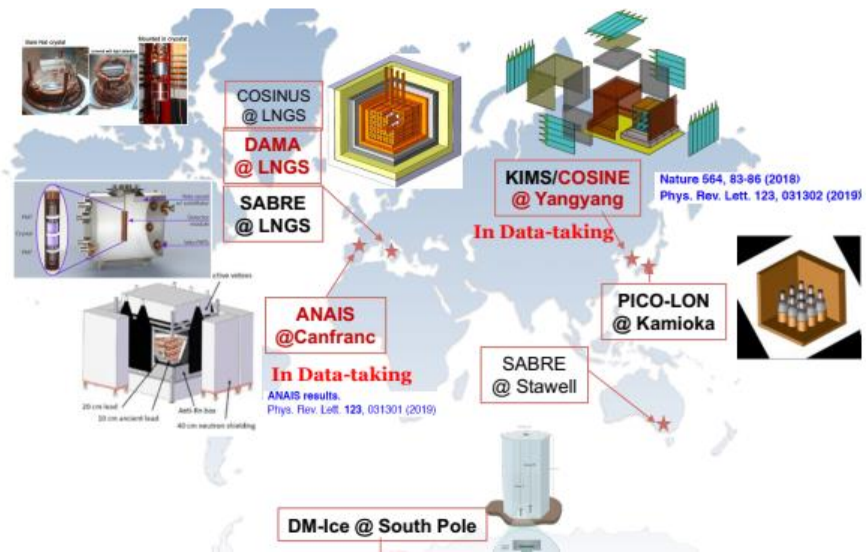
-> $CeBr_3$@ESA

^{138}La(0.09%) : 1.5 Bq/cm^3

Rare event searches

Race to reduce radioactive background

1) Dark matter search
2) 0ν ββ decay search



Internal radioactivity in crystal scintillators : Pro

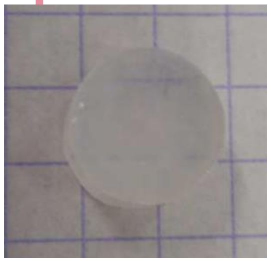
1. γ–Calibration source by coincidence method (γ–β or γ–γ)

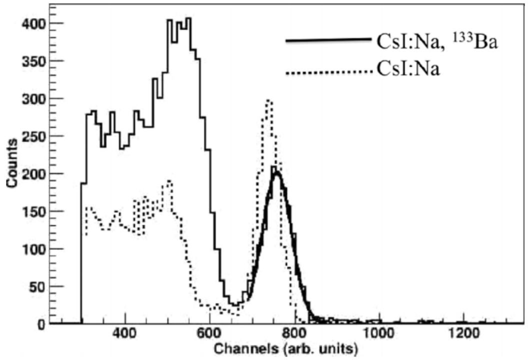
- One can use intrinsic radioactive scintillator or radioactivity loaded scintillator

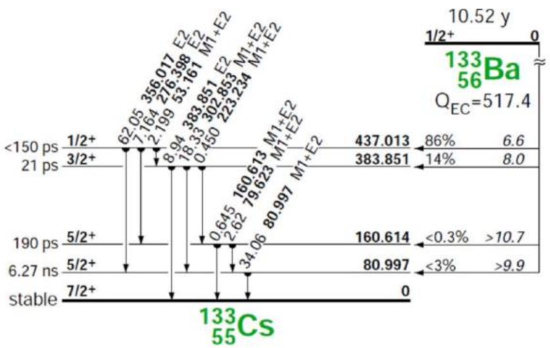
IEEE TRANSACTIONS ON NUCLEAR SCIENCE, VOL. 60, NO. 2, APRIL 2013

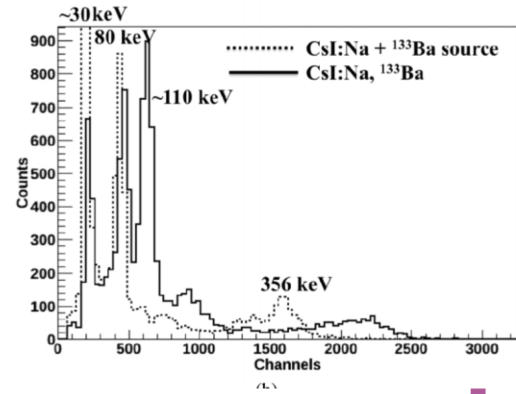
Scintillation Properties of CsI:Na, ^{133}Ba Crystal

M. J. Kim, Hong Jooknu Kim, H. Park, K. Tanida, Sunghwan Kim, J. K. Cheon, and K. B. Lee





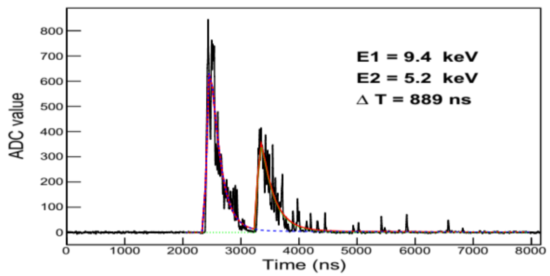




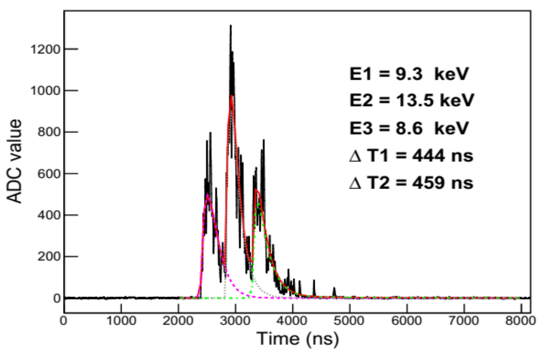
Internal radioactivity in crystal scintillators : Pro

2. Rare or Isomer decay study

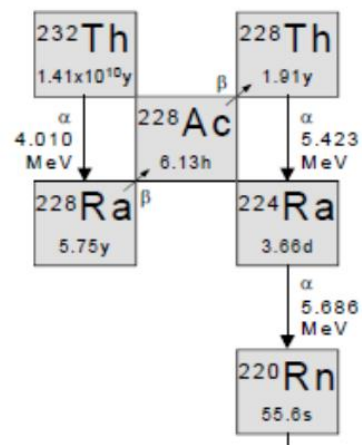
- ^{228}Ac Isomer background at COSINE dark matter search (100kg NaI:Tl)



Crystal 3 (Oct. 21, 2016)



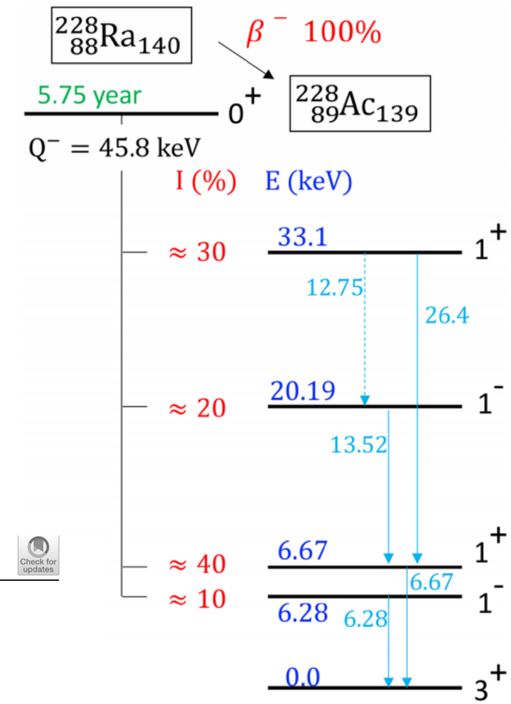
Crystal 7 (July 12, 2018)



Eur. Phys. J. C (2021) 81:746
https://doi.org/10.1140/epjc/s10052-021-09473-2
THE EUROPEAN PHYSICAL JOURNAL C
Regular Article - Experimental Physics

Identification of new isomers in ^{228}Ac: impact on dark matter searches

COSINE-100 Collaboration



250 ns decay time is too long

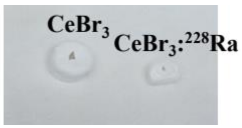
Internal radioactivity in crystal scintillators : Pro

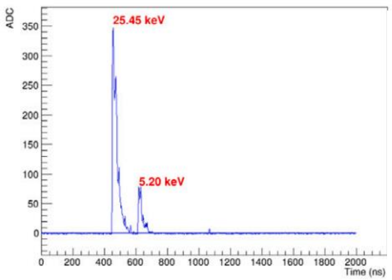
Further ^{228}Ac Isomer study, $CeBr_3$:^{228}Ra was developed.

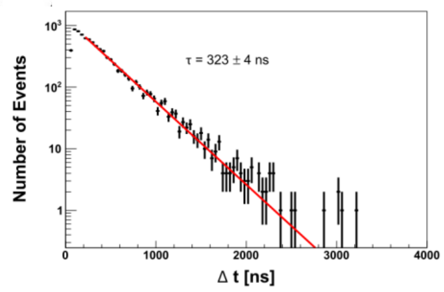
Sejin Na et al, IEEE TNS,VOL. 55,NO.3,JUNE,2008

$CeBr_3$ and NaI(Tl) properties.

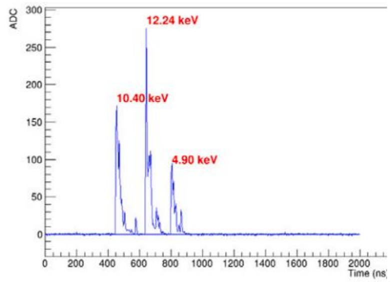
Property	$CeBr_3$	NaI(Tl)
Density (g/cm^3)	5.10	3.67
Effective Z	45.9	51
Melting point (°C)	722	651
Light yield (photons/MeV)	60,000–70,000	38 000
Energy resolution @662 keV (%)	~4–5	~6–7
Decay time (ns)	~20	~250
Wavelength of emission max (nm)	380	415

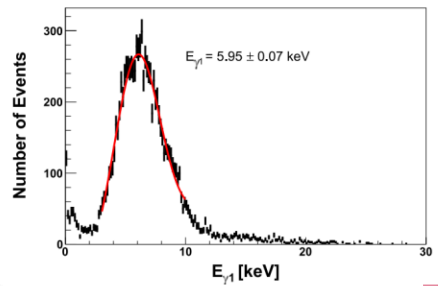






(a)





Radiation Physics and Chemistry 238 (2026) 113148

Measurement of new ^{228}Ac Isomers using a ^{228}Ra doped $CeBr_3$ scintillation crystal

D.H. Lee [a], E.J. Choi [a], H.S. Lee [b], N.T. Luan [c], J.H. So [b], H.J. Kim [a,c],*

Internal radioactivity in crystal scintillators : Pro

2. Application of radioactivity doped crystal scintillator : Source-Detector technique

- Short decay time (>10 ns) and low energy gamma Isomer study
- Low energy β or alpha with γ emitting isotope study for new state
- β spectral shape measurement
- Extremely rare nuclear transition research
- Half life should be longer (> 1 month)

3. New particle search

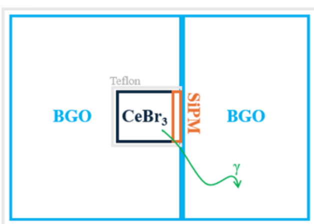
- Dark photon, Dark Z
- Axion like particle (ALP)
- Others?

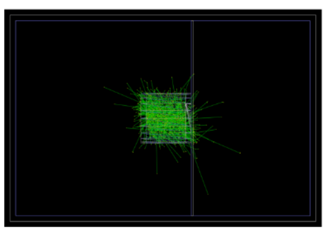
Source-as-Detector Experiment

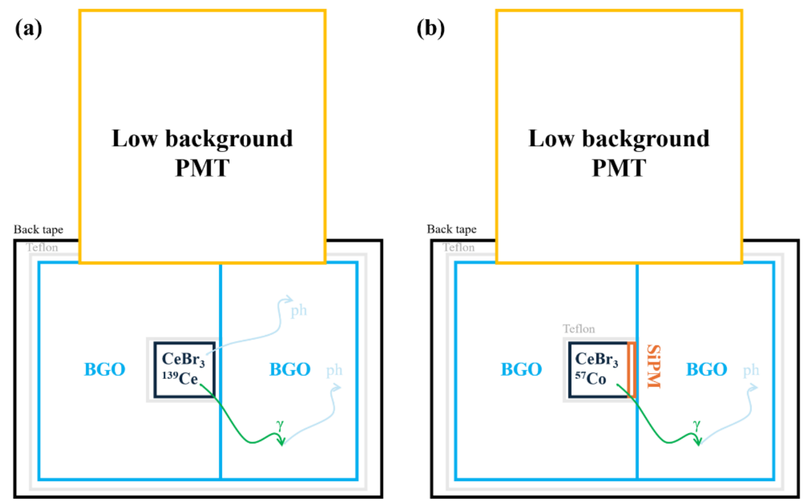
$CeBr_3$ advantages :
Fast decay time(~20 ns), high light yield (~60,000 ph/MeV), good energy resolution (4% FWHM @661 keV)
Disadvantages : very hygroscopic, internal radioactive background?

Target Isotopes : $CeBr_3$:^{139}Ce, $CeBr_3$:^{57}Co, $CeBr_3$:^{44}Ti

Detector schematic

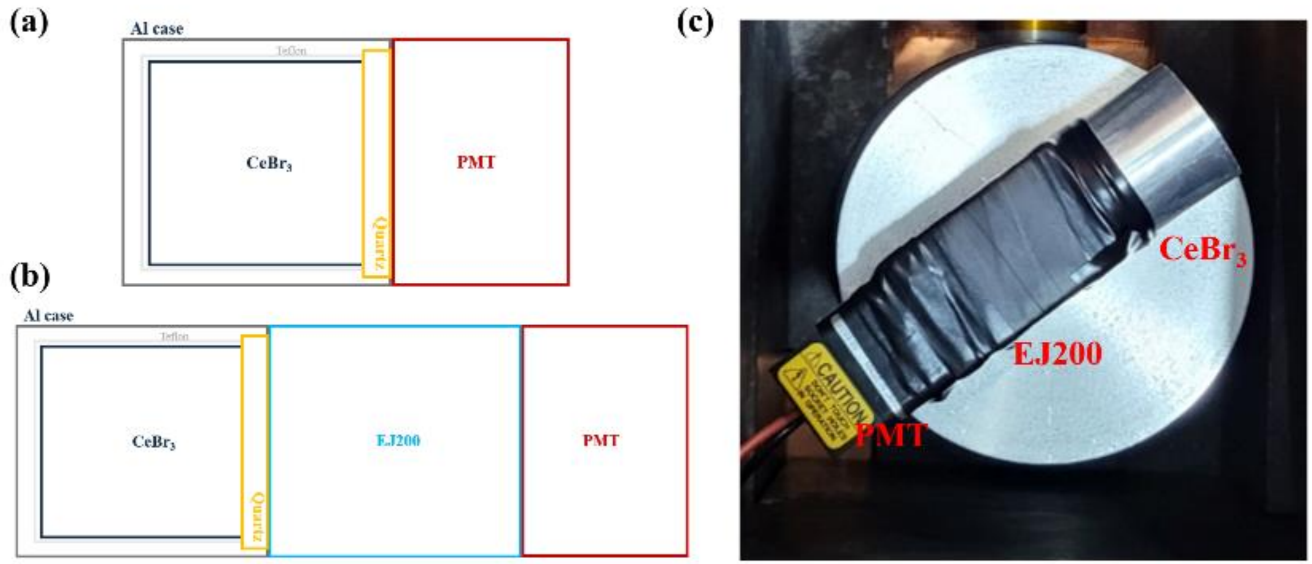


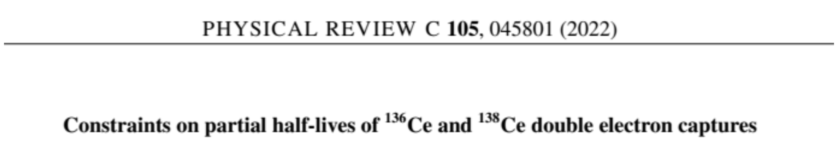




Phoswich detector

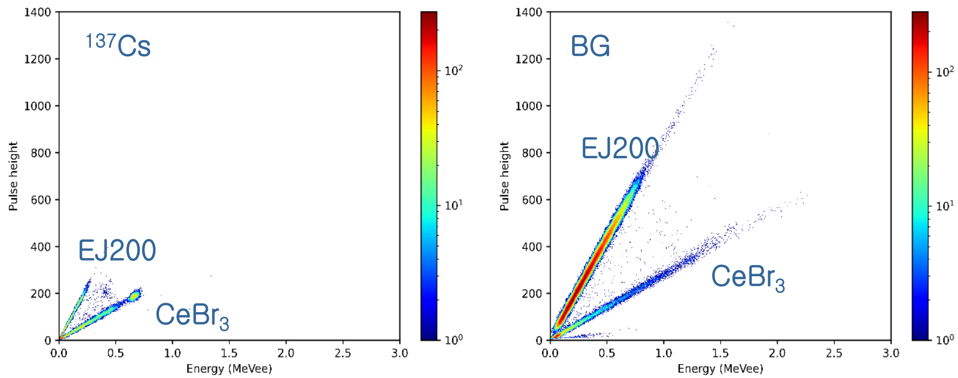
1″ $CeBr_3$ internal background study @Yemi

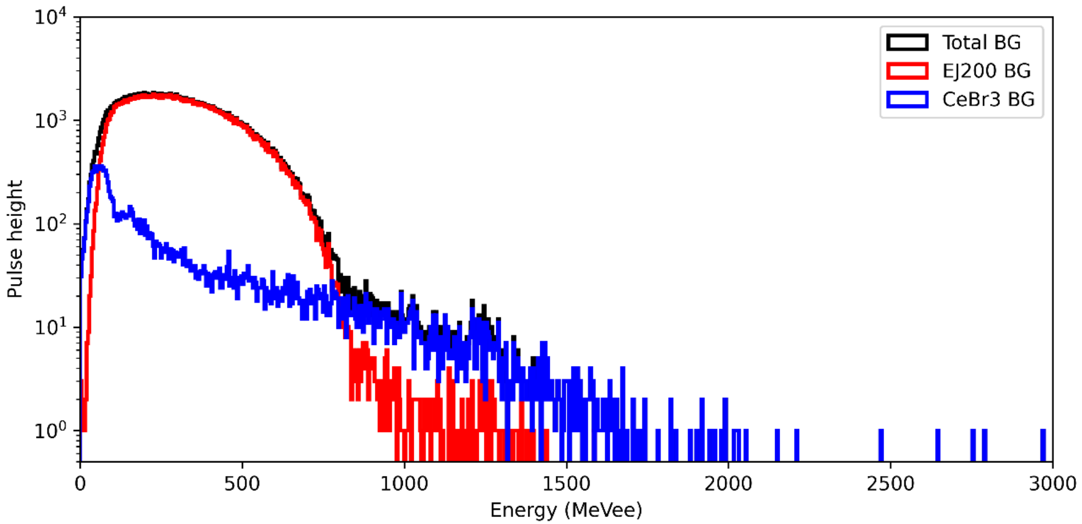



PHYSICAL REVIEW C **105**, 045801 (2022)

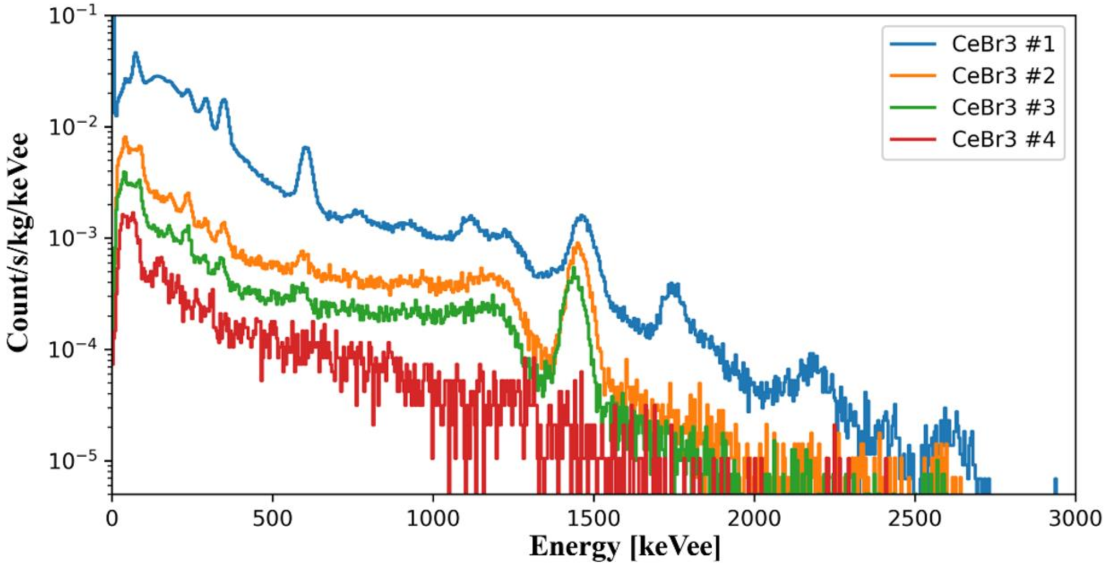
Constraints on partial half-lives of ^{136}Ce and ^{138}Ce double electron captures

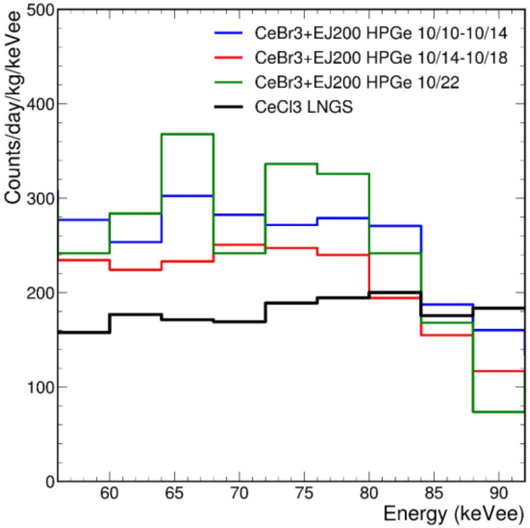
Background measurement using $CeBr_3$+EJ200 detector





Background measurement using $CeBr_3$ detector





The Gran Sasso National Laboratory (LNGS) of the INFN (Italy)
P Belli et al., J. Phys. G: Nucl. Part. Phys. 38 (2011) 015103 (15pp)

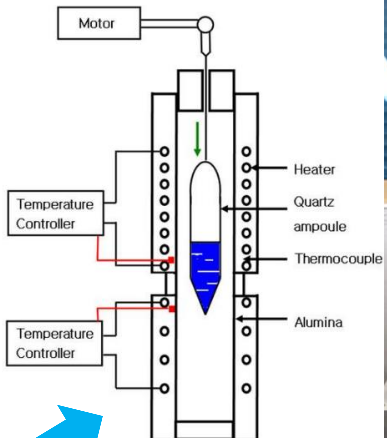
$CeBr_3$ #1: Measure at High Rn level using Lead shielding in air
$CeBr_3$ #2: Measure at HPGe shielding in Argon-filled condition
$CeBr_3$ #3: Measure at HPGe shielding in Argon-filled condition after 4 days
$CeBr_3$ #4: Measure at HPGe shielding in Argon-filled condition with EJ200 light guide

Radioactive source doped $CeBr_3$ crystal growth

Rad. Source in Solution

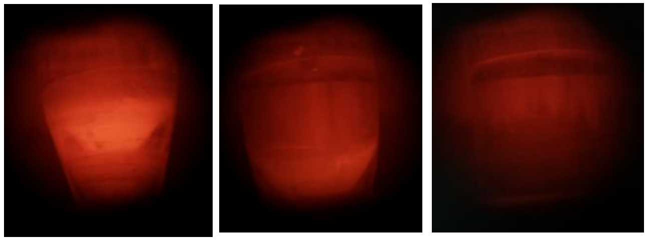
Rad. ampoule coating (under vacuum)

"home-made" Bridgman furnace



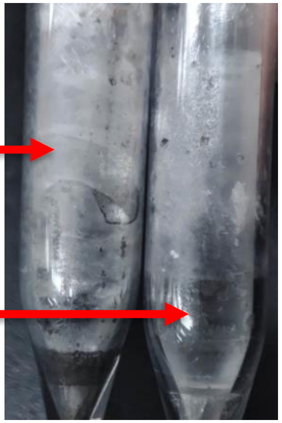
Fail process (dehydrate) (solved)

Realtime camera assistant interface monitor monitoring and changing growth rate



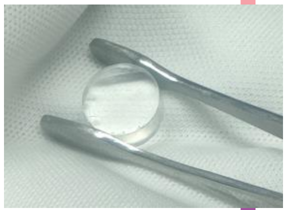
#Early
Dirty ampoule
Bad cracks

#NOW
#Optimized
Clean
No cracks
Long



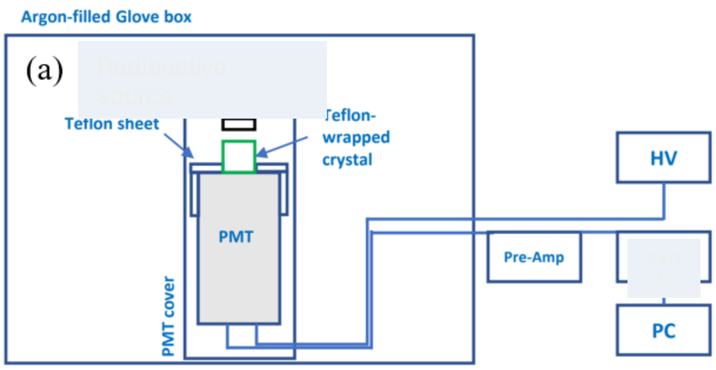
Cutting

Crystal

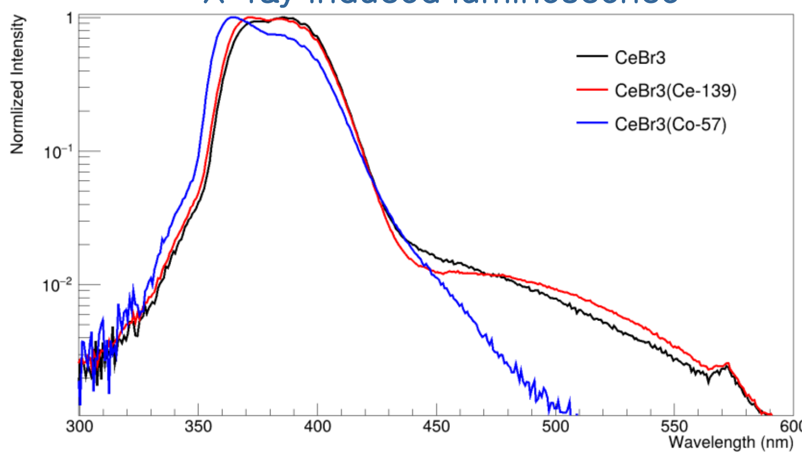


Characterization of $CeBr_3$:^{139}Ce and $CeBr_3$:^{57}Co

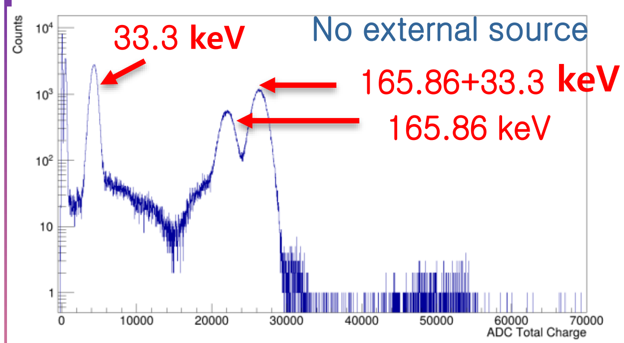
- Super bi-alkali PMT 12669SEL (High quantum efficiency, low background)
- Korea Notice 500-MHz flash Analog to Digital Converter

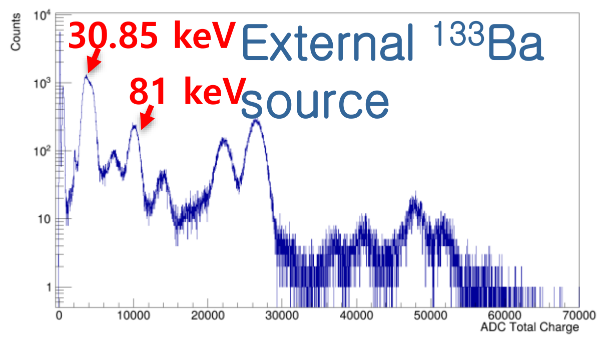


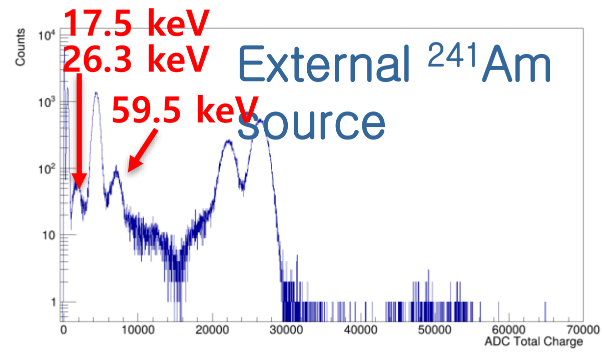
X-ray induced luminescence

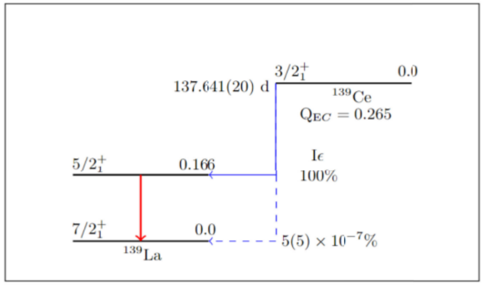


$CeBr_3$:^{139}Ce









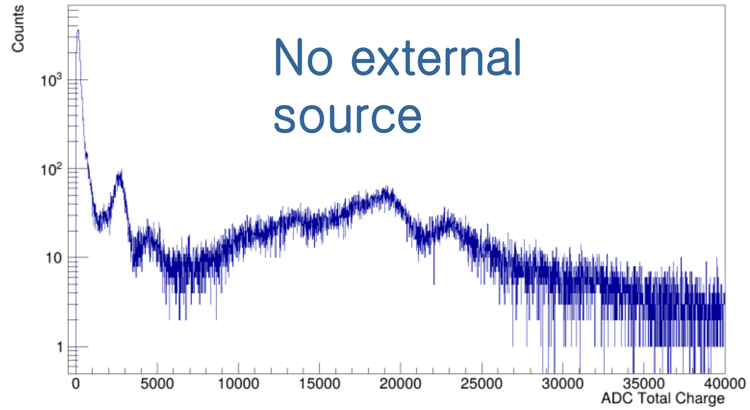
#1

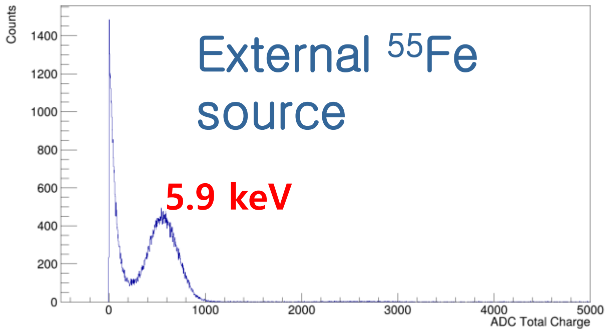
M1 $5/2^+_1 \rightarrow 7/2^+_1$:

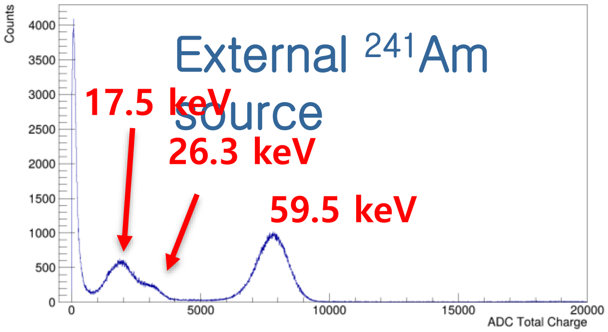
165.86 keV

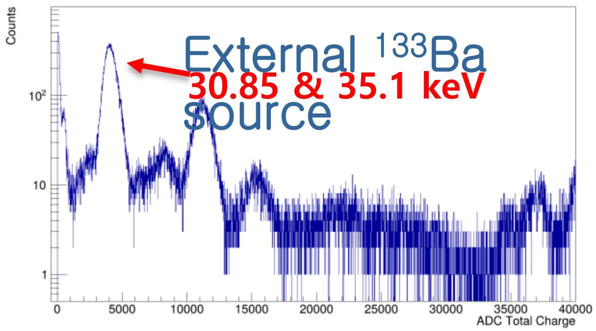
→ K_α X-ray: ~33 keV

$CeBr_3$:^{57}Co

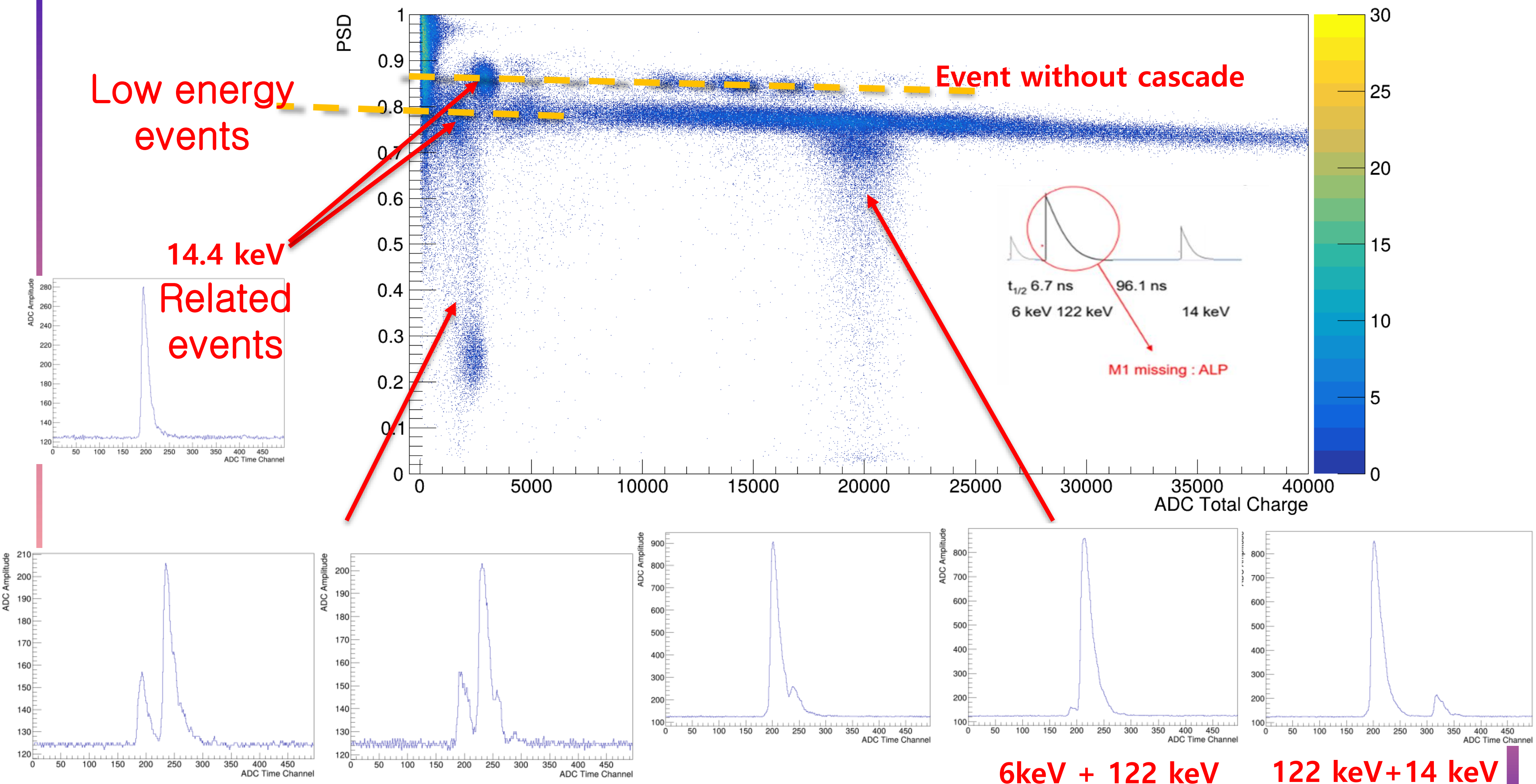

No external source
Counts
ADC Total Charge


External 55Fe source
5.9 keV
Counts
ADC Total Charge

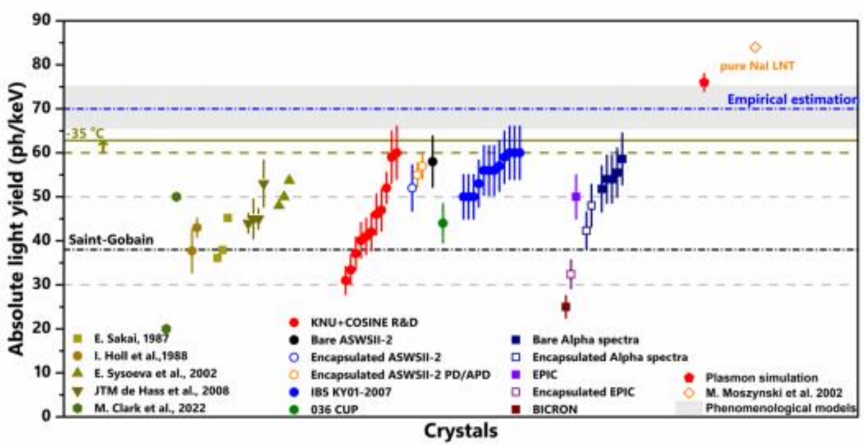

External 241Am source
17.5 keV
26.3 keV
59.5 keV
Counts
ADC Total Charge

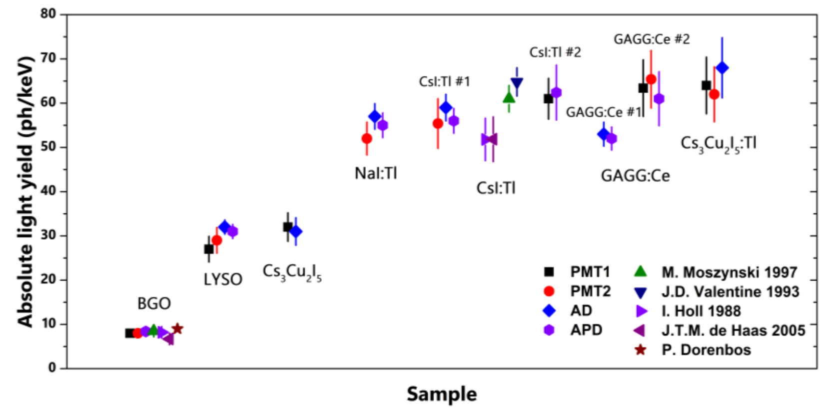

External 133Ba source
30.85 & 35.1 keV
Counts
ADC Total Charge

CeBr$_3$:^{57}Co cascade decay



Absolute light yield measurement



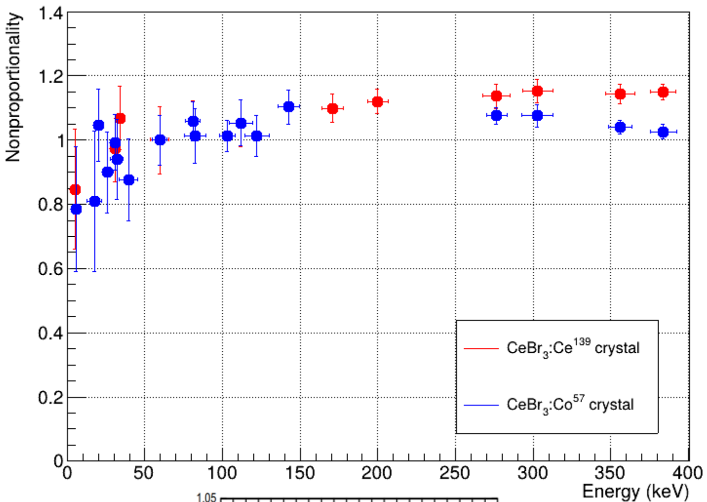


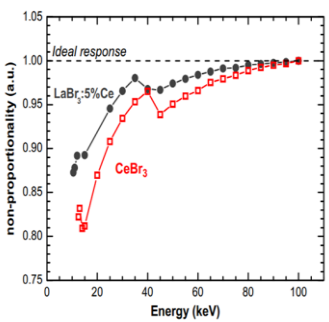
- Nguyen Thanh Luan H.J. Kim et al 2025 JINST 20 P06024

$CeBr_3$:^{139}Ce	$CeBr_3$:^{57}Co
60.0±6.0 ph/keV @ ~5 keV 74.0±7.4 ph/keV @ ~34 keV	47.0±4.7 ph/keV @ 5.9 keV 51.0±5.1 ph/keV @ 14.4 keV 56.0±5.6 ph/keV @ 32 keV

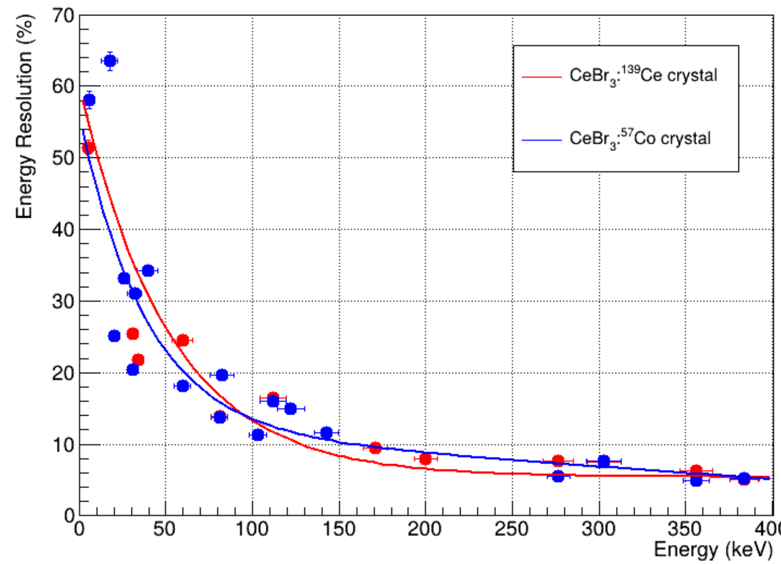
Nonproportionality

Light output

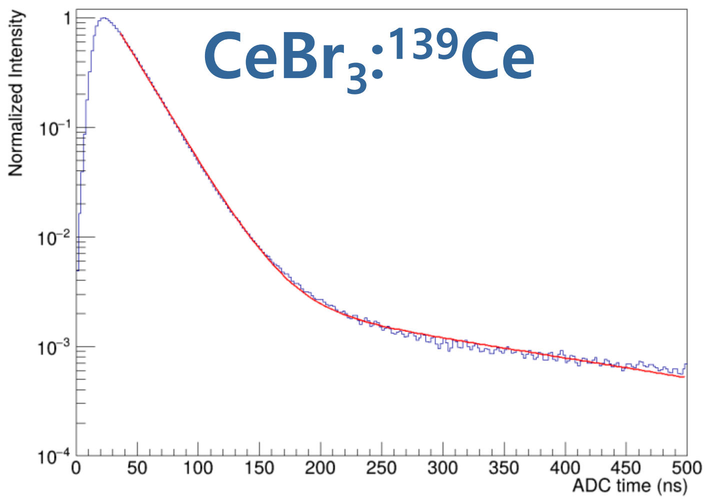


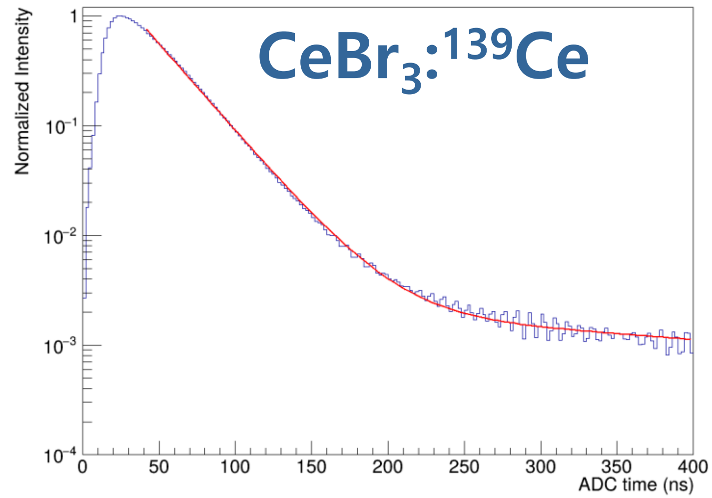


Energy resolution



Decay time





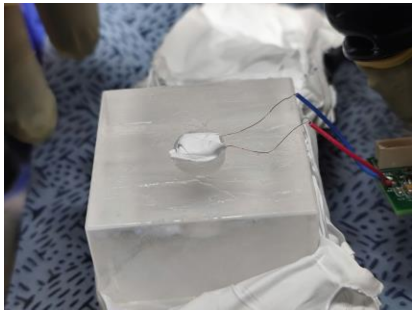
$CeBr_3$:^{139}Ce	$CeBr_3$:^{57}Co
23±2 ns (97%)	27±2 ns (98%)
240±35 ns (3%)	430±30 ns (2%)

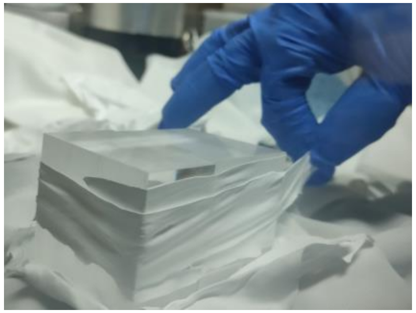
❑ Super bi-alkali PMT 12669SEL (High quantum efficiency, low background)

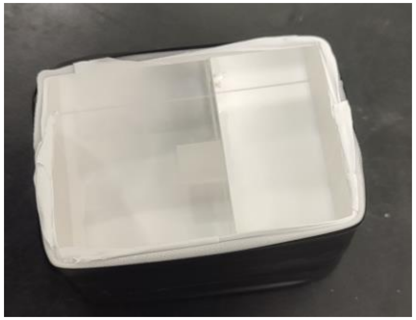
BGO veto

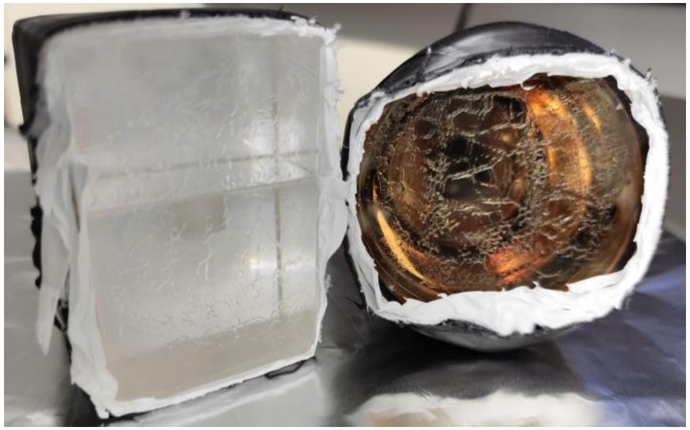
Cutting & drilling

Detector preparation
(Argon environment)

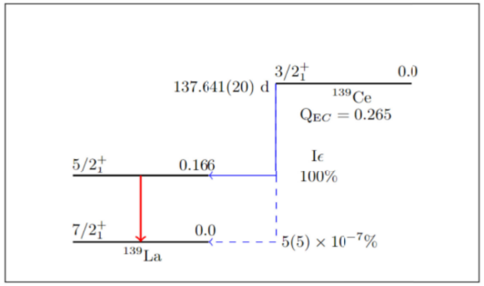








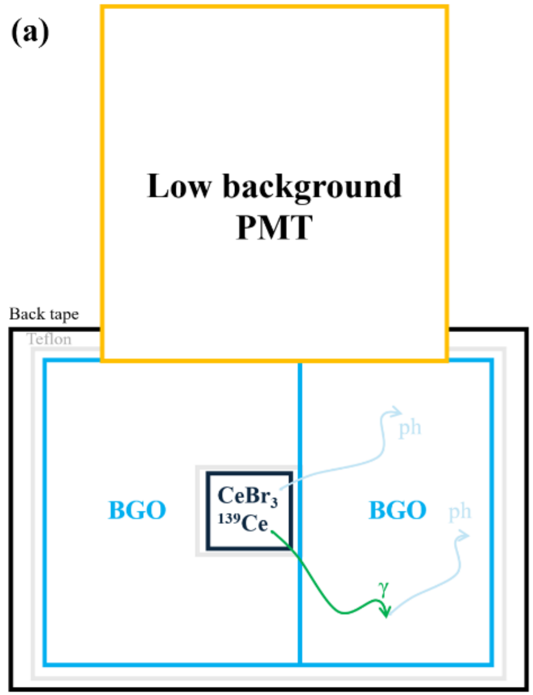
1st Experiment using ^{139}Ce Source



#1
M1 $5/2^+_1 \rightarrow 7/2^+_1$: 165.86 keV
→ K_α X-ray: ~33 keV

Easiest case

Search for single X-ray



Phoswich detector

Background shielding @ YemiLab

GRANDE Room

Lead shielding

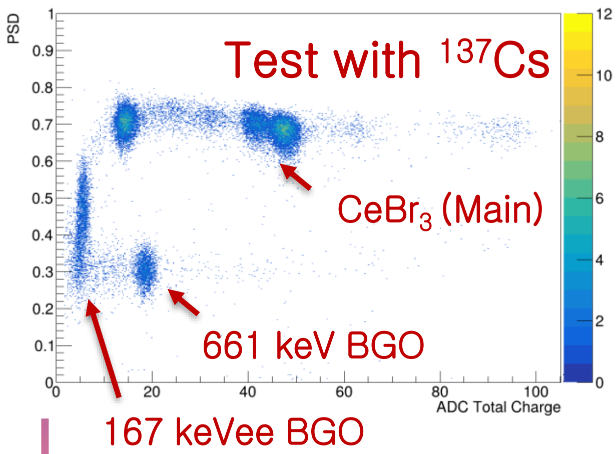
Copper shielding

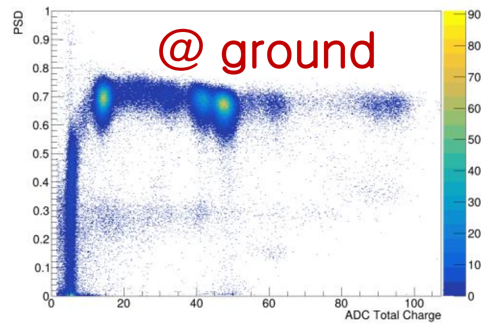
Experiment using ^{139}Ce Source

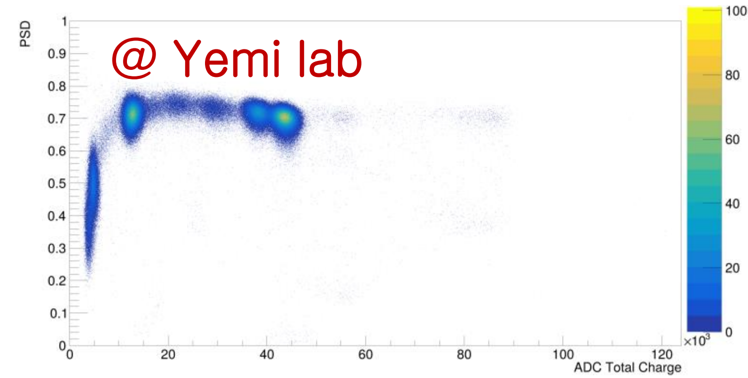
- ❑ Super bi-alkali PMT 12669SEL (High quantum efficiency, low background)
- ❑ Korea Notice 500-MHz flash Analog to Digital Converter
- ❑ Caen High Voltage Supplier
- ❑ PC with 18Tb storage
- ❑ Data rate ~20 kHz

Running 5 month without problem!
No degradation of CeBr3 crystal

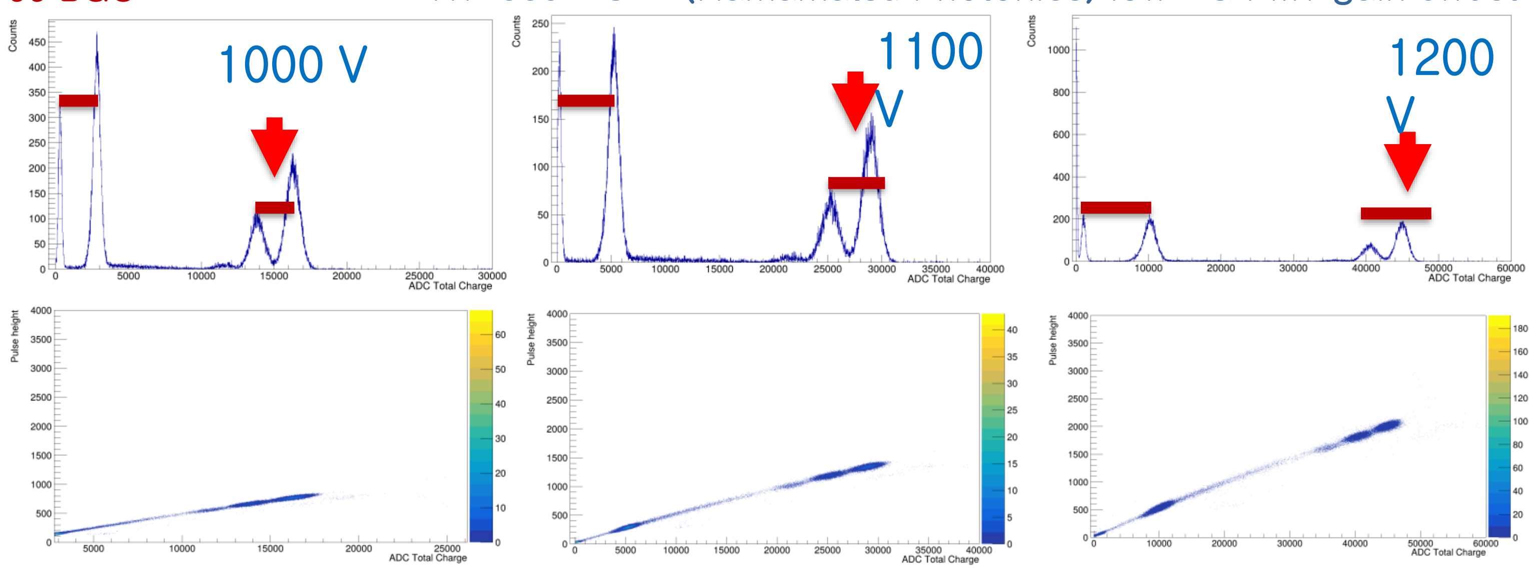
$CeBr_3$:^{139}Ce + BGO veto



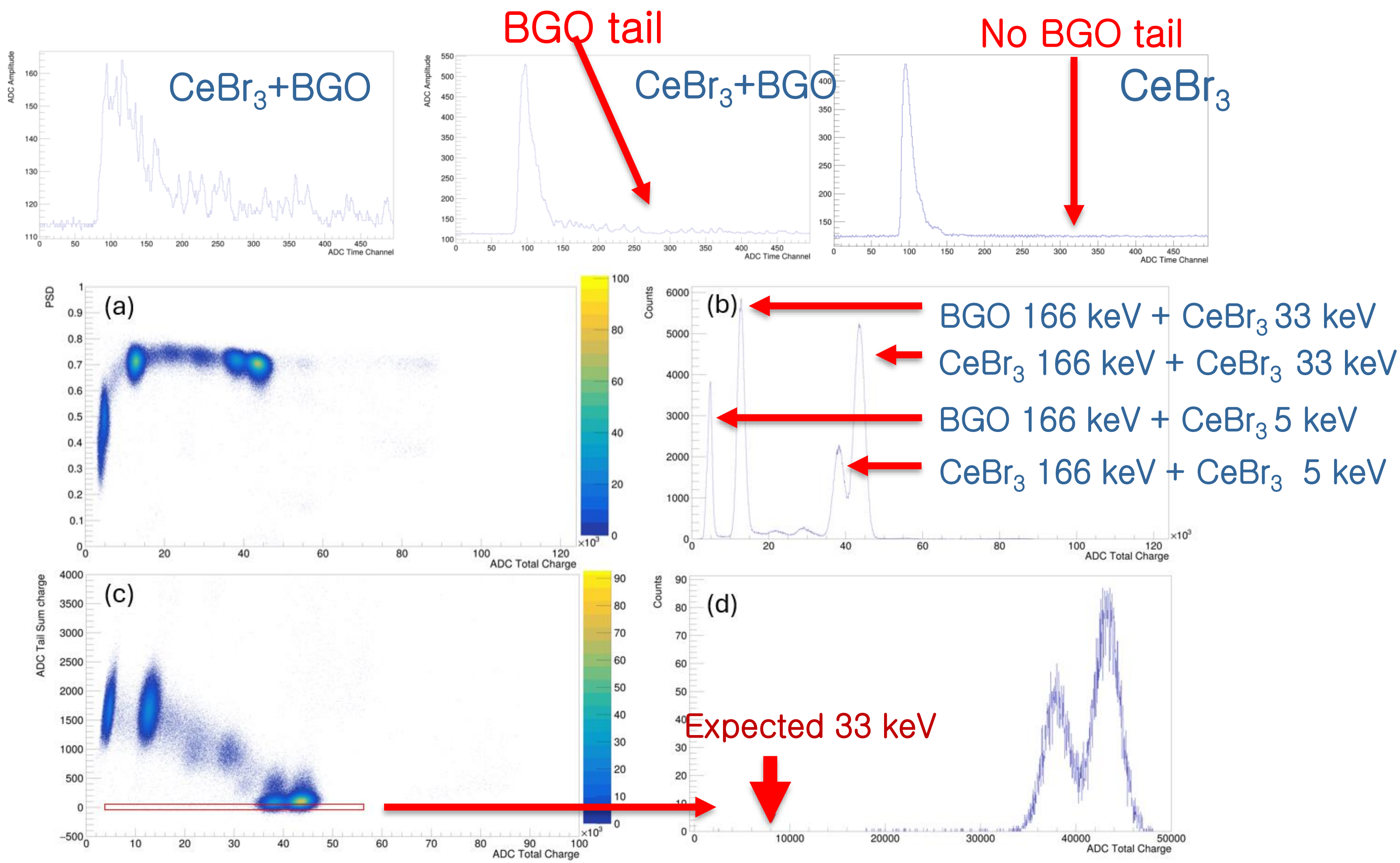




R12669 – SEL (Hamamatsu Photonics) low BG PMT gain effect

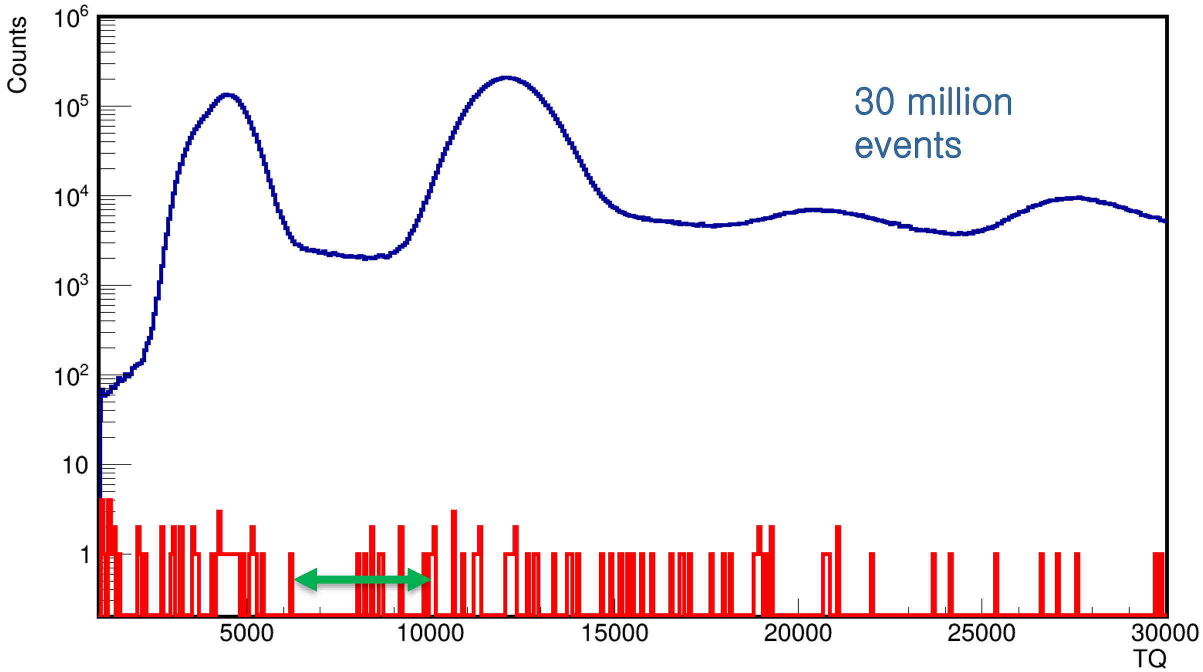


$CeBr_3$:^{139}Ce + BGO veto



$CeBr_3$:^{139}Ce + BGO veto

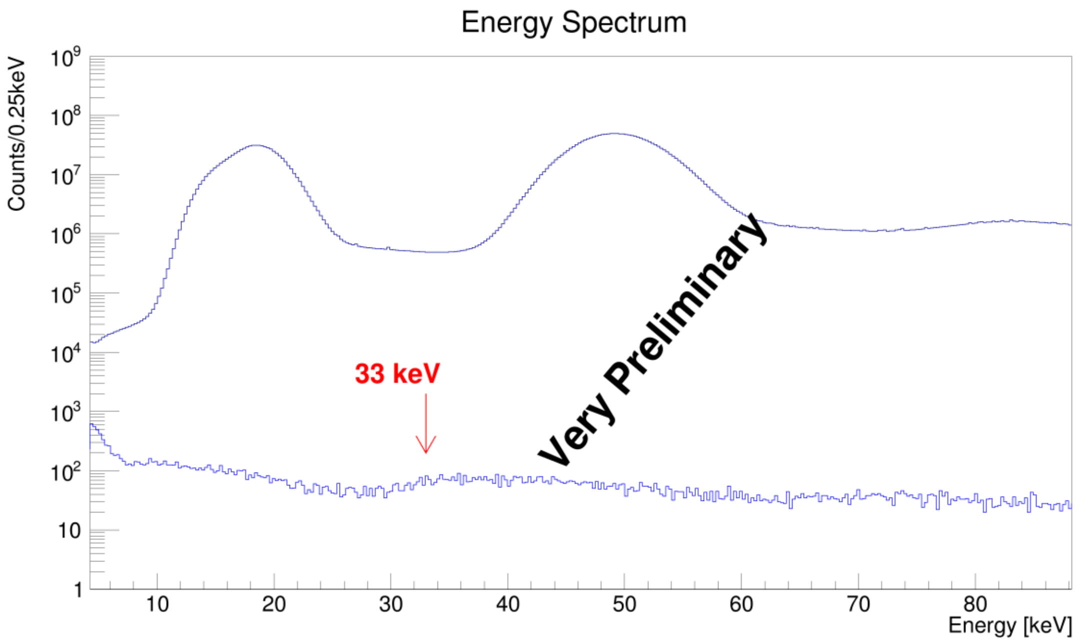
- 2 million events → reach 1 x 10^{-6}
- 30 million events → reach 4 x 10^{-7}

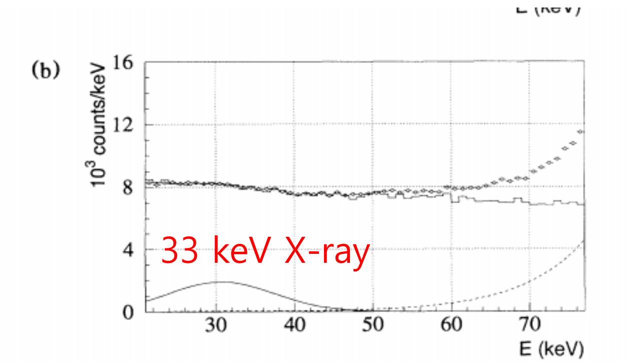


8000 -> 33 keV (20% FWHM energy resolution)

$CeBr_3$:^{139}Ce + BGO veto

- Very preliminary results with 10 billion events (<1 month data taking)
- 200 times lower background than M. Minowa's experiment

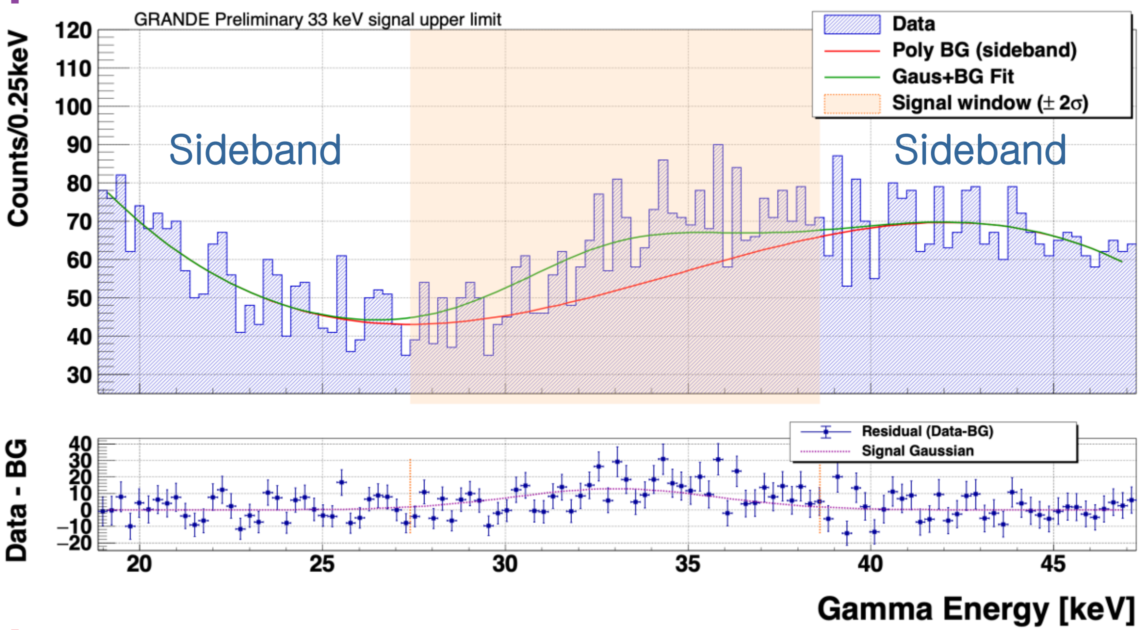




$$\Gamma_a/\Gamma_\gamma < 1.21 \times 10^{-6} \text{ at the } 95\%$$

Mass of hadronic axion > 26.7 keV by 95%CL
M.Minowa et al, Phys. Rev. Lett. 71, 4120

Very preliminary ALP upper limit of ^{139}Ce



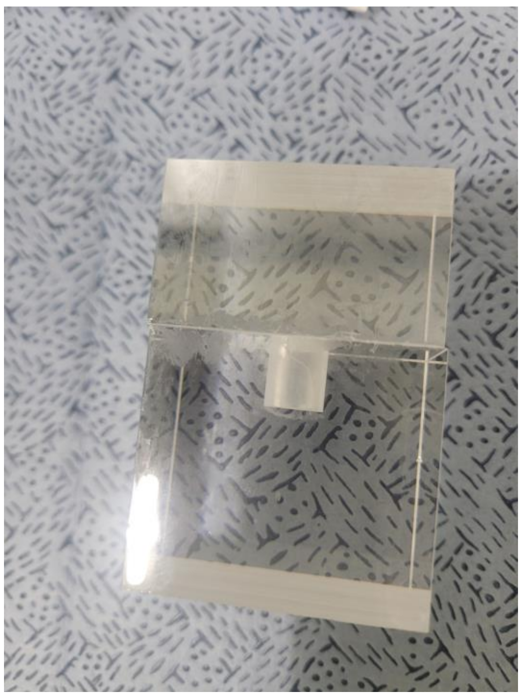
- **Signal window:**
  33 keV, ±2σ region
- **Sideband:** 33 keV, ±5σ region, excluding the signal window (±2σ) with 3rd-order polynomial
- **Branching ratio upper limit:** 1.1×10^{-8} **(90% C.L.)**
  Preliminary
- **Previous experiment**
  $\Gamma_a/\Gamma_\gamma < 1.21 \times 10^{-6}$ at the 95%

Upgrade of $CeBr_3$:^{139}Ce experiment with KAPAE-II

Phoswitch to KAPAE-II

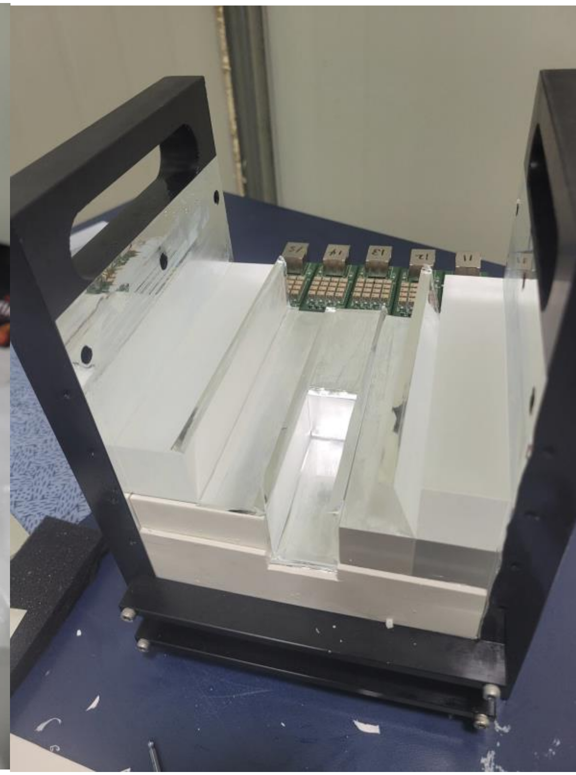
1. BGO thickness is only 2.0cm (166 keV γ leak) -> 6cm for KAPAE-II setup
2. Phoswitch does not give a perfect separation between CeBr3 and BGO signal

Phoswitch setup



KAPAE-II Setup

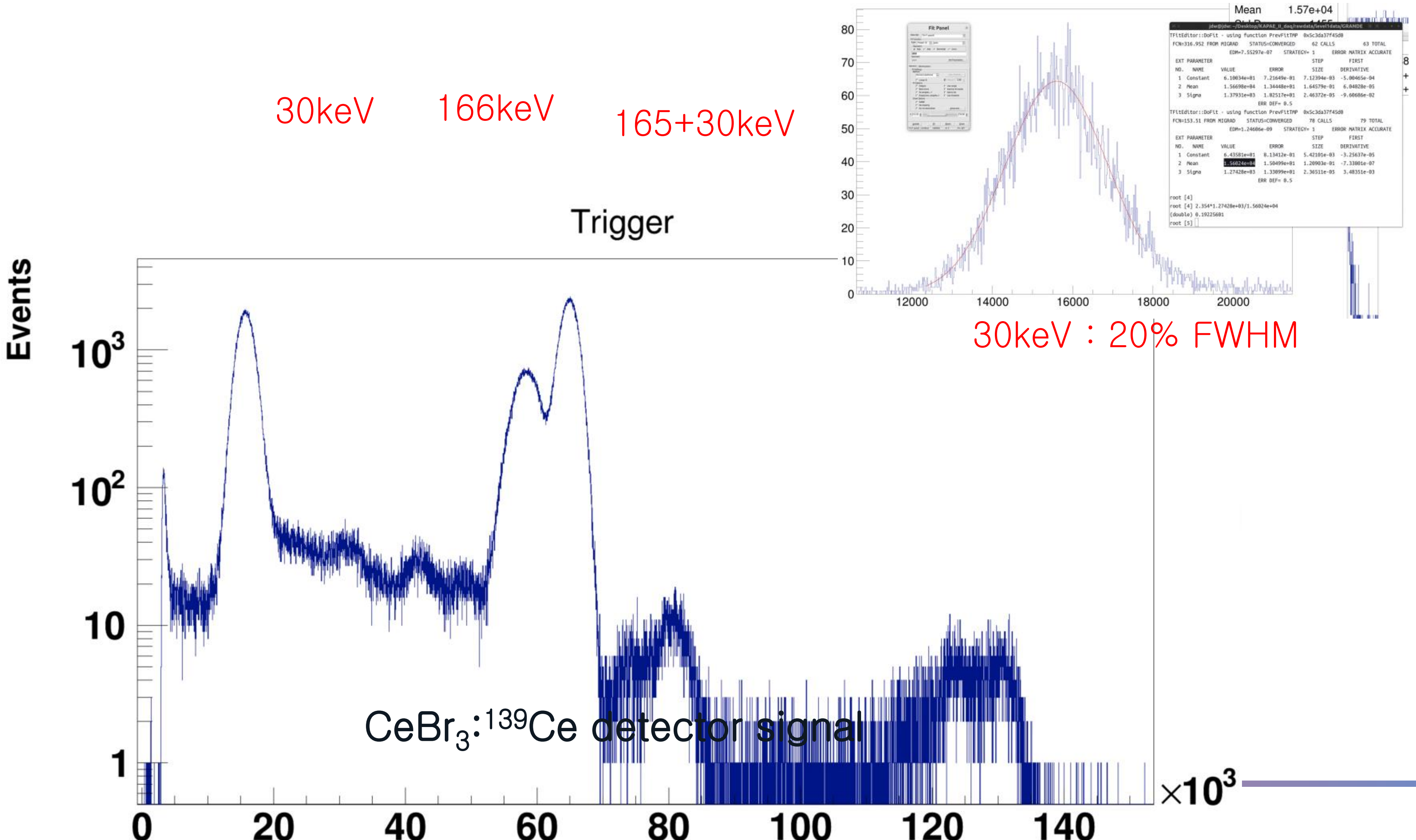
^{139}Ce experiment with KAPAE-II Setup



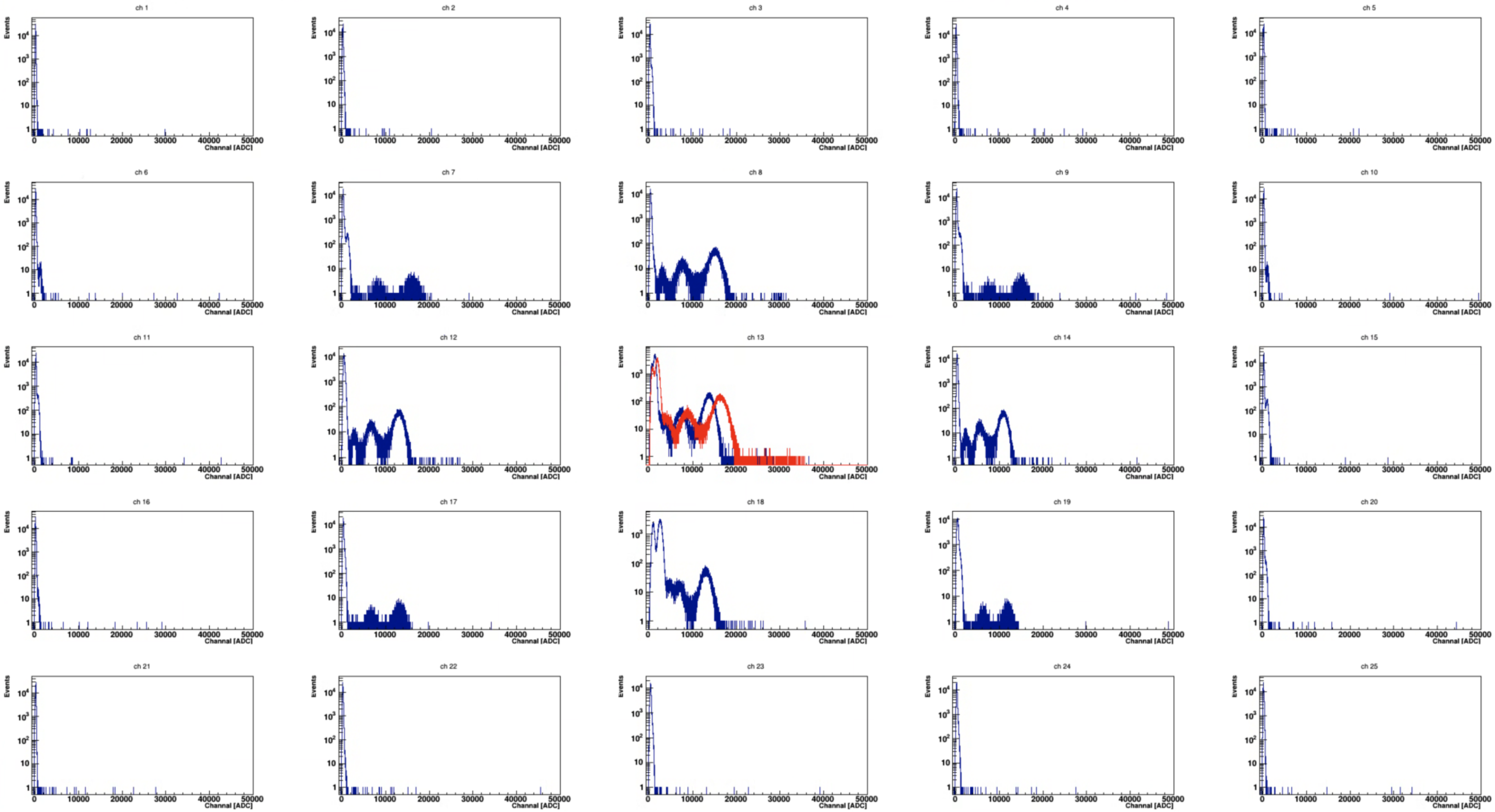
$CeBr_3$:^{139}Ce Crystal with SiPM connection

^{139}Ce experiment with KAPAE-II Setup : CeBr3 signal

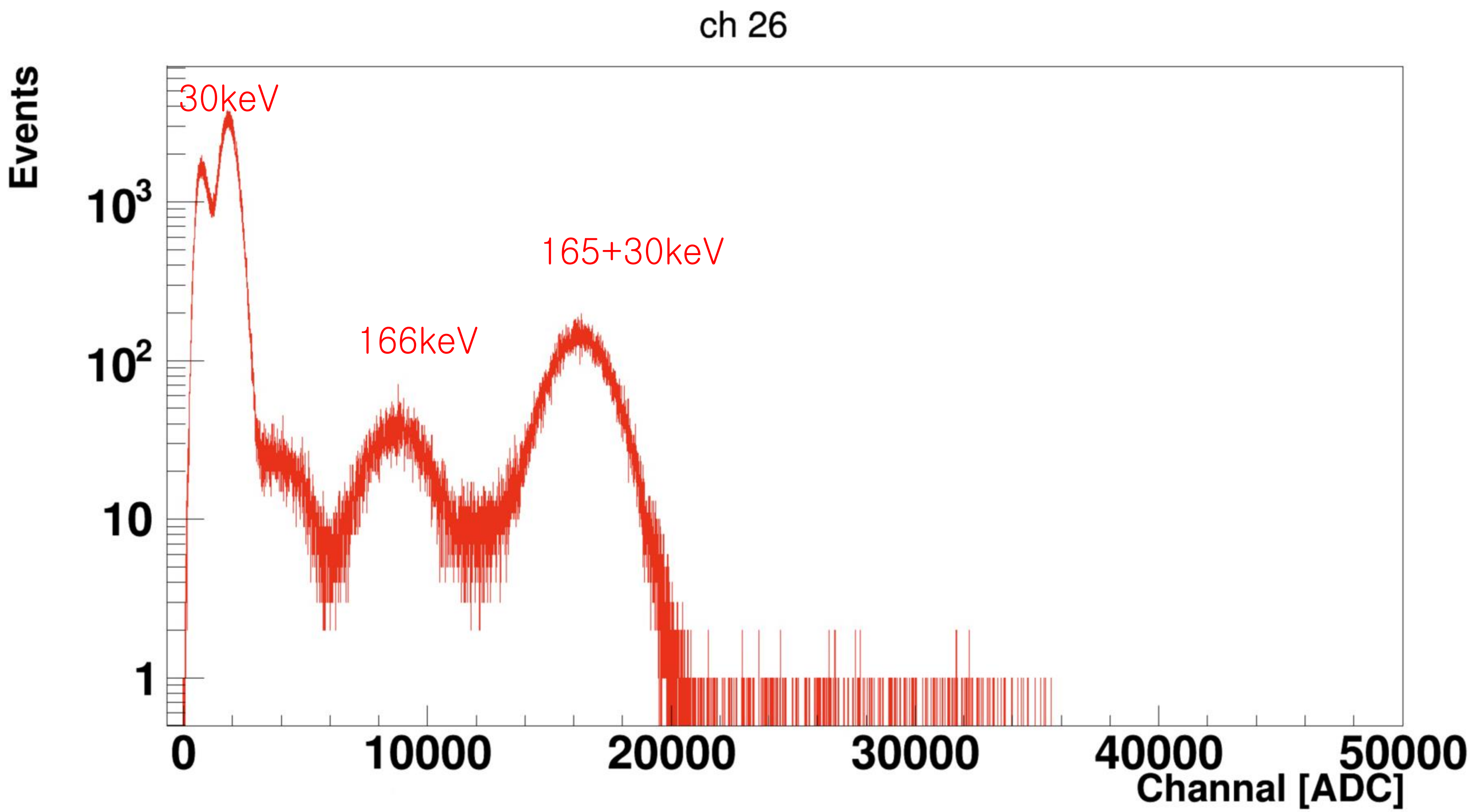
Data just started Oct 17, 2025 and will be continue for 3 month
Trigger rate is 3352 Hz that we expect to have 10^{10} events



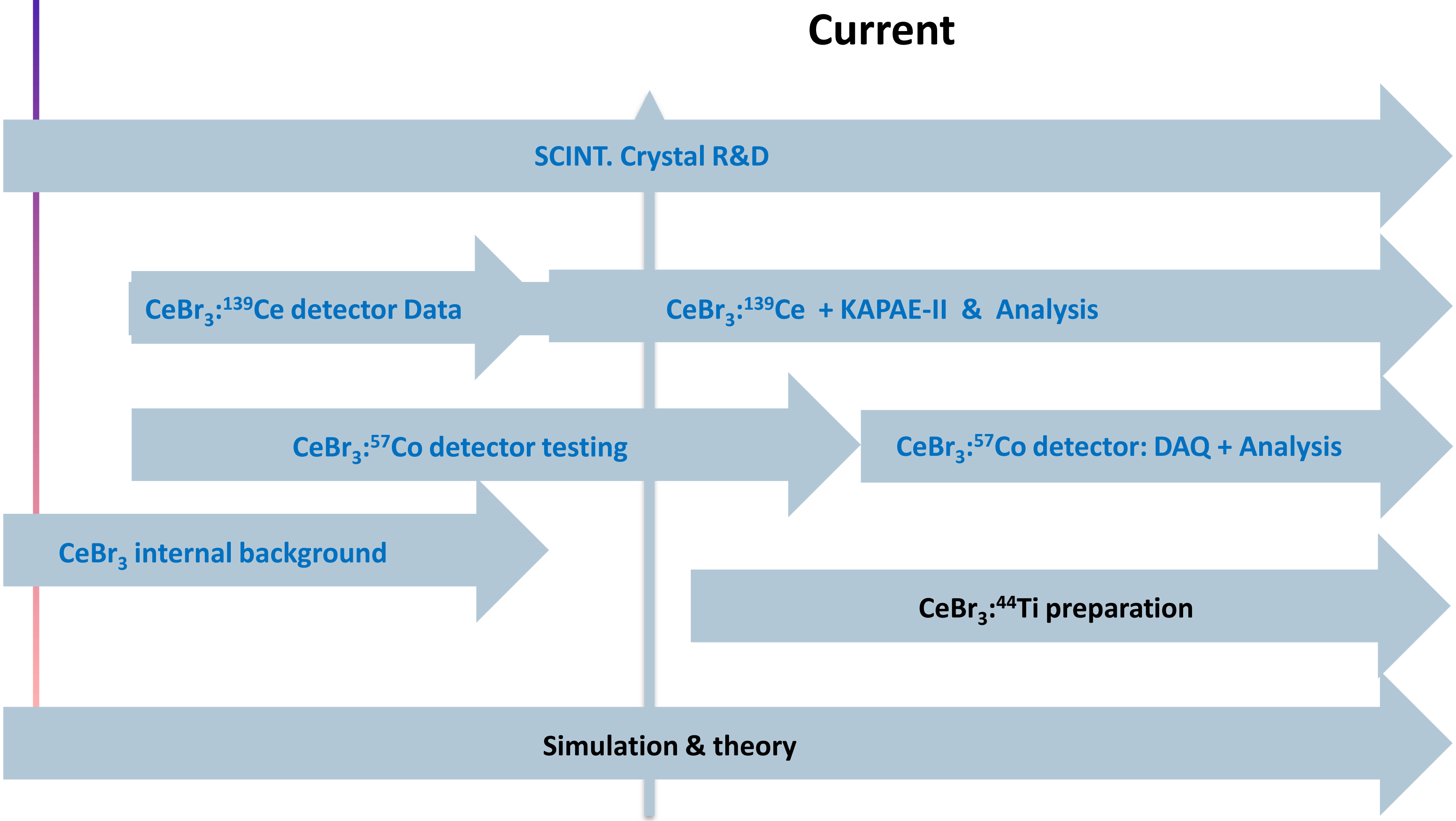
^{139}Ce experiment with KAPAE-II Setup :BGO signal



^{139}Ce experiment with KAPAE-II Setup : BGO @ Endcap



PLAN



Current
SCINT. Crystal R&D
$CeBr_3$:^{139}Ce detector Data
$CeBr_3$:^{139}Ce + KAPAE-II & Analysis
$CeBr_3$:^{57}Co detector testing
$CeBr_3$:^{57}Co detector: DAQ + Analysis
$CeBr_3$ internal background
$CeBr_3$:^{44}Ti preparation
Simulation & theory

Summary and Prospect

- Radioactive sources are good for the Tabletop experiments wit low budget.
- Search for the exotic dark matter search and extremely rare nuclear EC transition measurement will be performed with GRANDE.
- Source-Detector technique is used with $CeBr_3$ doped with radio-active source.
- To reduced external radio-active background, 4p BGO veto is used and experiment is on going at Yemi underground laboratory in Cu and Pb shielding.
- Missing gamma study with ^{139}Ce, ^{57}Co and ^{44}Ti of EC M1/E1/E2 transition, exotic dark matter search of ALP, dark photon and anapole will be performed in GRANDE.
- Are there any new theoretical motivation at E < 1MeV for new particle searches related with invisible particles such as new bosonic particle?

18th International Conference on
Scintillating Materials and their Applications

SCINT 2026
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SCINT 2026
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Main Topics
- Mechanisms and theory of scintillation
- Characterization of scintillators
- Crystal growth and structural control
- Optical ceramics and glasses
- Nano- and metamaterials, hybrids, organic, and liquid scintillators
- Scintillators for fast-timing detection and imaging
- Scintillators for neutron detection and imaging
- Applications of scintillators in fundamental research, health, environment, energy, metrology, and industrial controls

Thank You !

Collaboration Welcome!