Performance of Novel Oxide Scintillator $Ce:(La,Gd)_2Si_2O_7$ with a High Energy Resolution

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[3] Institute of Physics, AS CR

Contents:

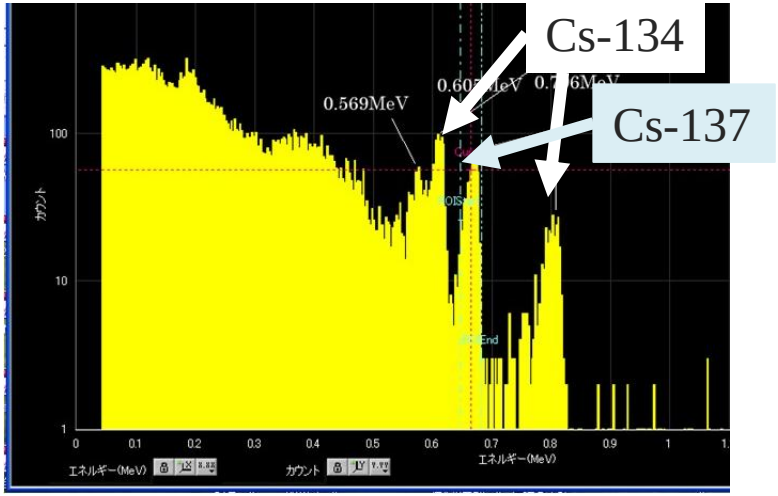
Vienna Conference on Instrumentation 2013

Introduction : Food Monitoring in Fukushima

Fukushima Problem

http://photo.tepco.co.jp/date/2011/201103-j/110321-01j.html

Ibaraki Newspaper: http://www.ifarc.metro.tokyo.jp/images/content/3685/takabe.jpg



http://www.shse.u-hyogo.ac.jp/kumagai/eac/4_20.htm

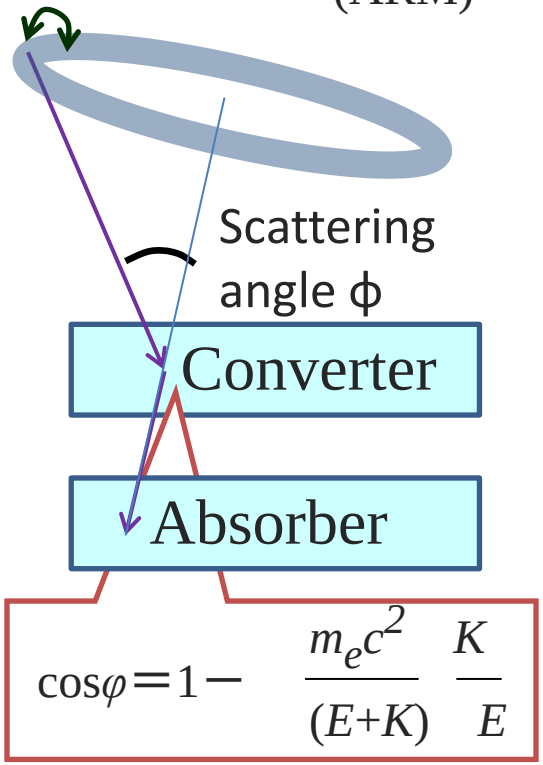
All foods in Fukushima or surrounding area are checked
[Foods < 100 Bq/kg, water < 10 Bq/kg]

Application Requirements

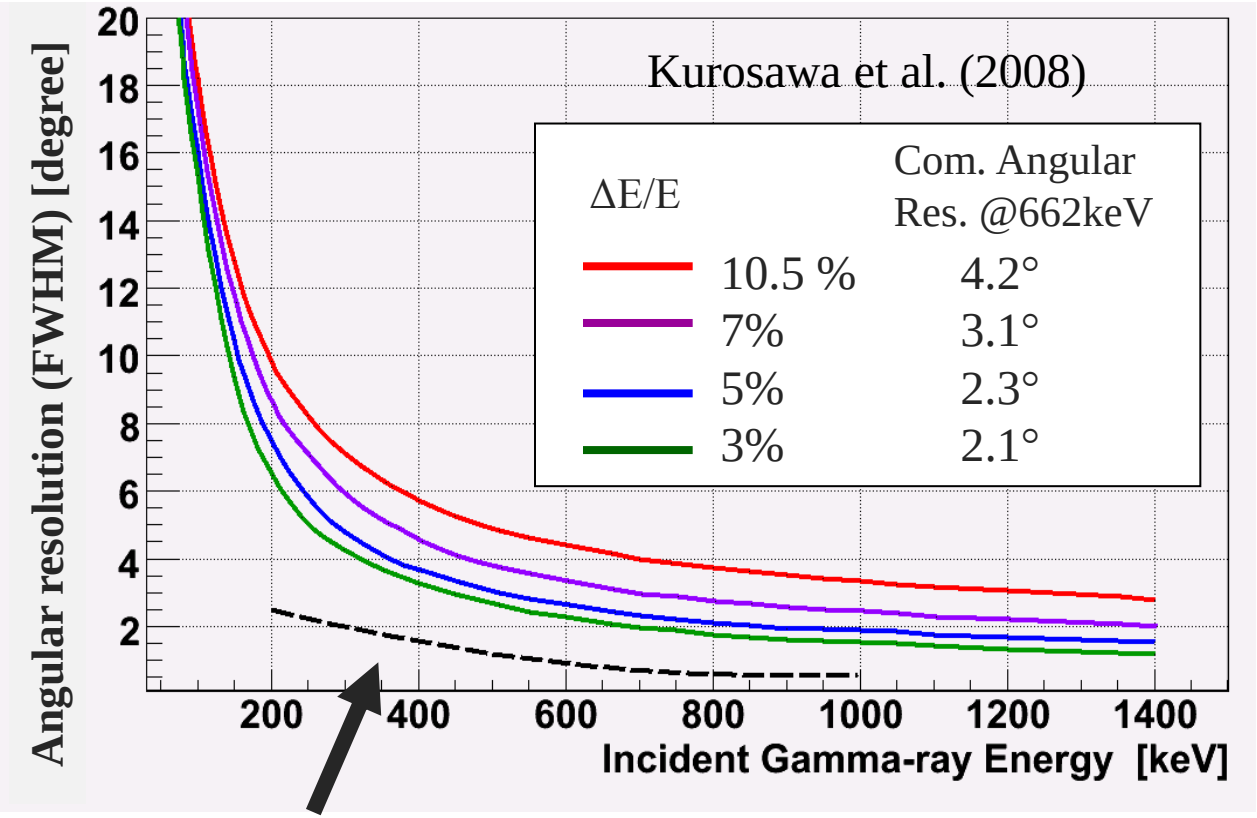
- Energy resolution [$\Delta E/E$ < 5-6%]
- low intrinsic background
- high detection efficiency

Introduction : Compton Camera

Δφ Angular resolution (ARM)



$$\cos\varphi = 1 - \frac{m_e c^2}{(E+K)} \frac{K}{E}$$



Doppler broadening (Ar) Zoglauer *et al.* (2003)

ΔE/E: energy resolution of absorber (scintillator) @ 662 keV, FWHM

Application Requirements

- Energy resolution [ΔE/E <5 %]
- low intrinsic background
- high detection efficiency

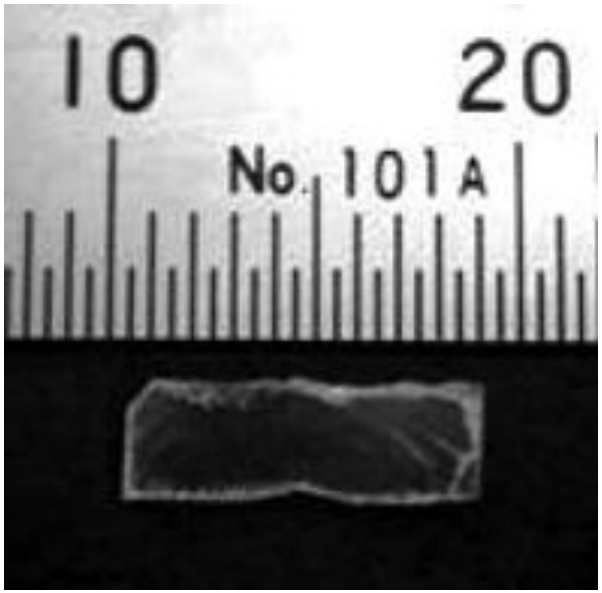
Introduction: Various Scintillators

	Density (g/cm³)	attenuation length[mm] @511keV	Decay time constant (ns)	Light output (Relative)	Hydroscopic nature	Radiation Hardness	energy res. @662 keV [%]
Tl:NaI	3.67	2.6	230	1	**Strong**	**very weak**	7
Tl:CsI	4.53	18.6	1050	0.85	**Weak**	**very weak**	7
BGO	7.13	large 1.1	300	0.07-0.12	**No**	**weak**	7
Ce:LSO	7.4	large 0.87	fast 40	0.4	**No**	**strong**	8
Ce:GSO	6.71	1.3	30-60	0.18	**No**	**very strong**	8
Ce:$LaBr_3$	5.3	2.2	20	1.6	**Very strong**	**strong**	3

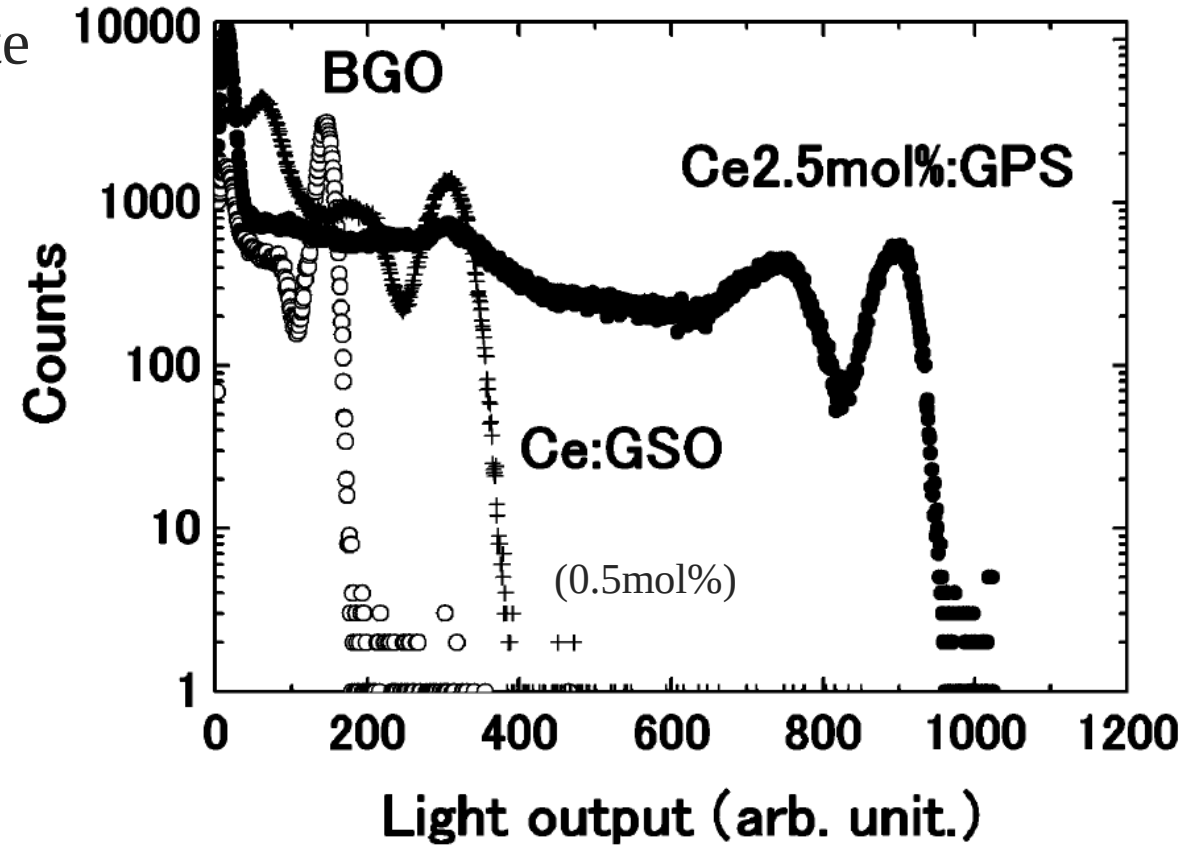
Although halide scintillators have good properties, they are strongly hydroscopic.

Introduction: $Gd_2Si_2O_7$ (GPS) Scintillator

Gadolinium Pyro-Silicate
Ce:$Gd_2Si_2O_7$ (Ce:GPS)



10 mol% Ce-doped GPS crystals grown by Floating Zone method [1].



Pulse height spectra excited (^{137}Cs irradiation). [2]

Light output: 24000 photons/MeV

Stabilizing to grow the single crystal

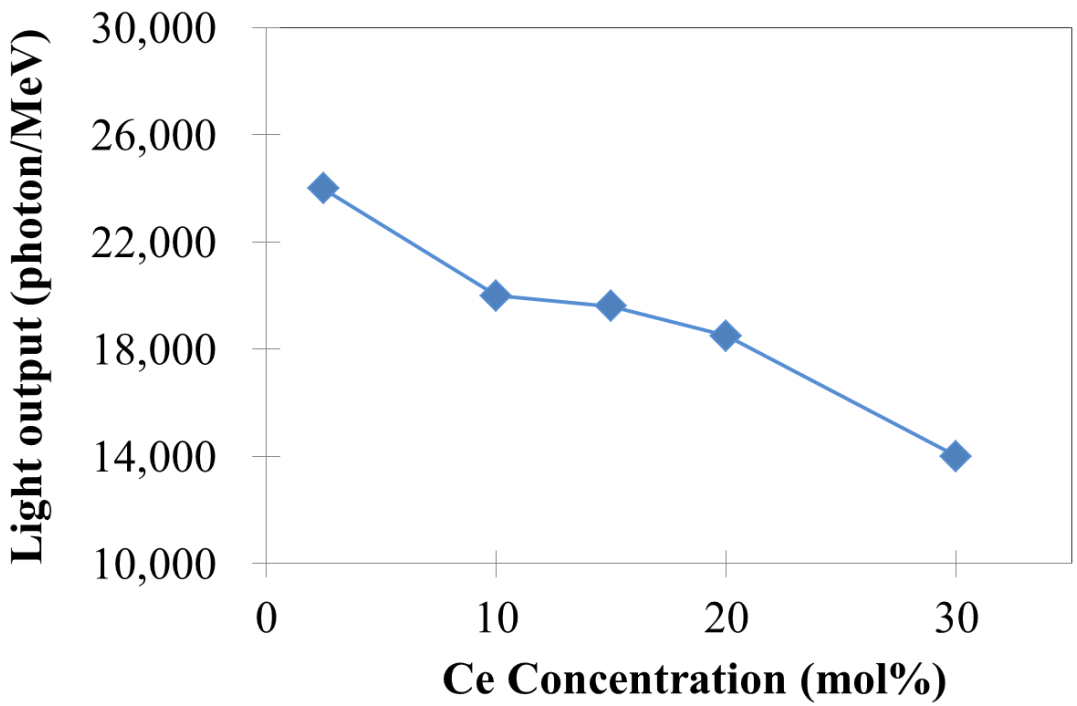
▽

Heavy Ce-doping

Reference:
[1] S. Kawamura et al.: IEEE Trans. Nucl. Sci. 54 (2007) 1383.
[2] S. Kawamura et al.: IEEE Trans. Nucl. Sci. 56 (2009) 328.

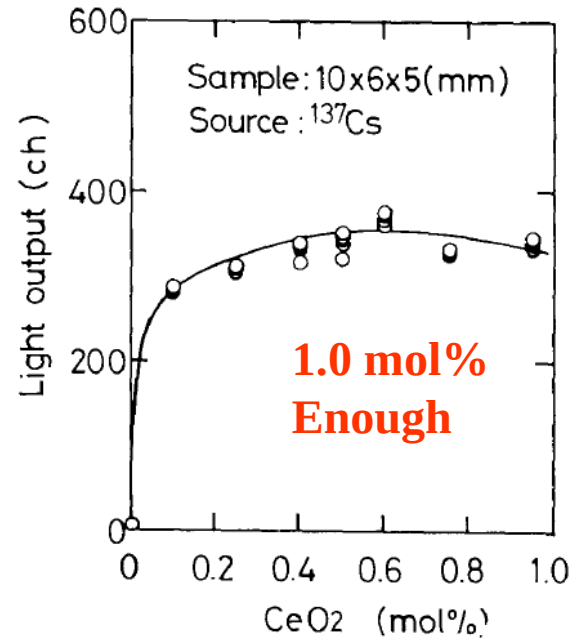
Introduction:Quenched Luminescence in Ce:GPS

Problem: Concentration Quenching → Much Higher Performance



Dependency of light output on
Ce concentration in Ce:GPS scintillator [1]

Case : Ce:GSO [2]



We challenged to develop low Ce-doped GPS.

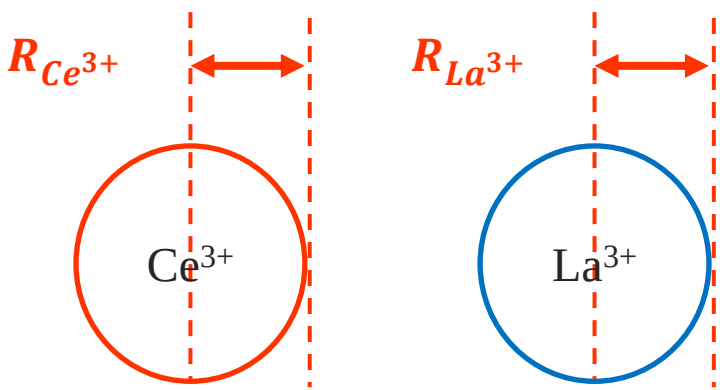
Reference [1] S. Kawamura et al.: IEEE Trans. Nucl. Sci. 56 (2009) 328.
[2] H. Ishibashi et al.: IEEE Trans. Nucl. Sci. 36 (1989) 170.

Introduction: $(La, Gd)_2Si_2O_7$ (La-GPS)

Ce:$(La, Gd)_2Si_2O_7$ (Ce:La-GPS)

Conventional GPS	Novel La-GPS
10 mol% Ce	1 mol% Ce for emission center **9 mol% La for stabilizing of crystal**

Material Design

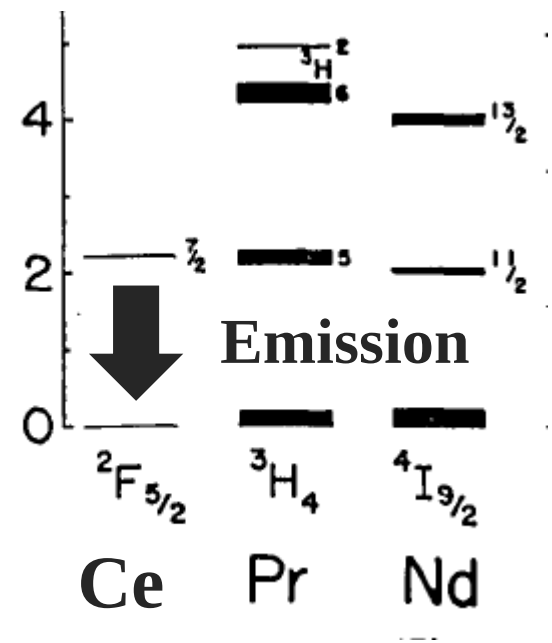


Comparable Ionic Radius

$$R_{Ce^{3+}} \sim R_{La^{3+}}$$

No emission

La



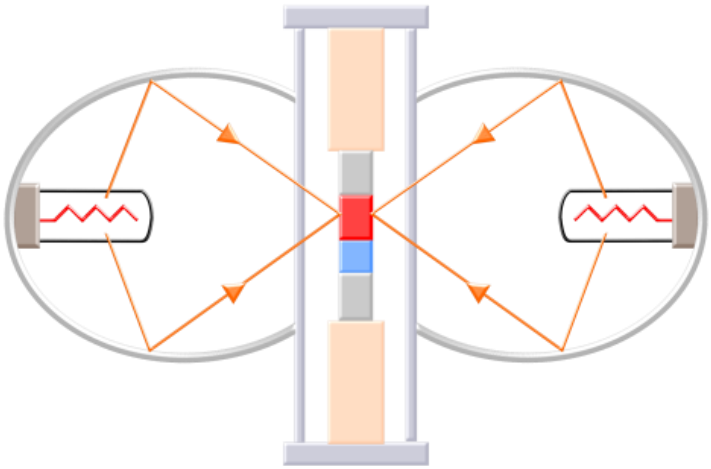
Dieke diagram

(G. H. Dieke and H. M. Crosswhite: Appl. Opt. 2 (1963) 675.)

Research Sample

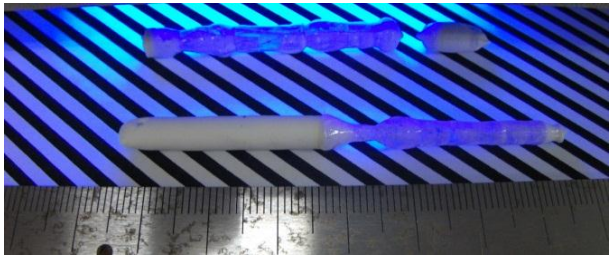
Material Preparation

Starting Material:
Gd_2O_3, La_2O_3
SiO_2, Ce_2O
99.99% Purity Powder

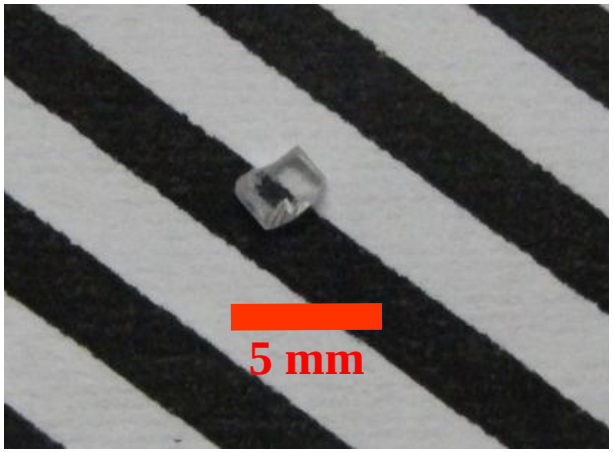


Floating Zone (FZ)
Method

As-grown Crystal

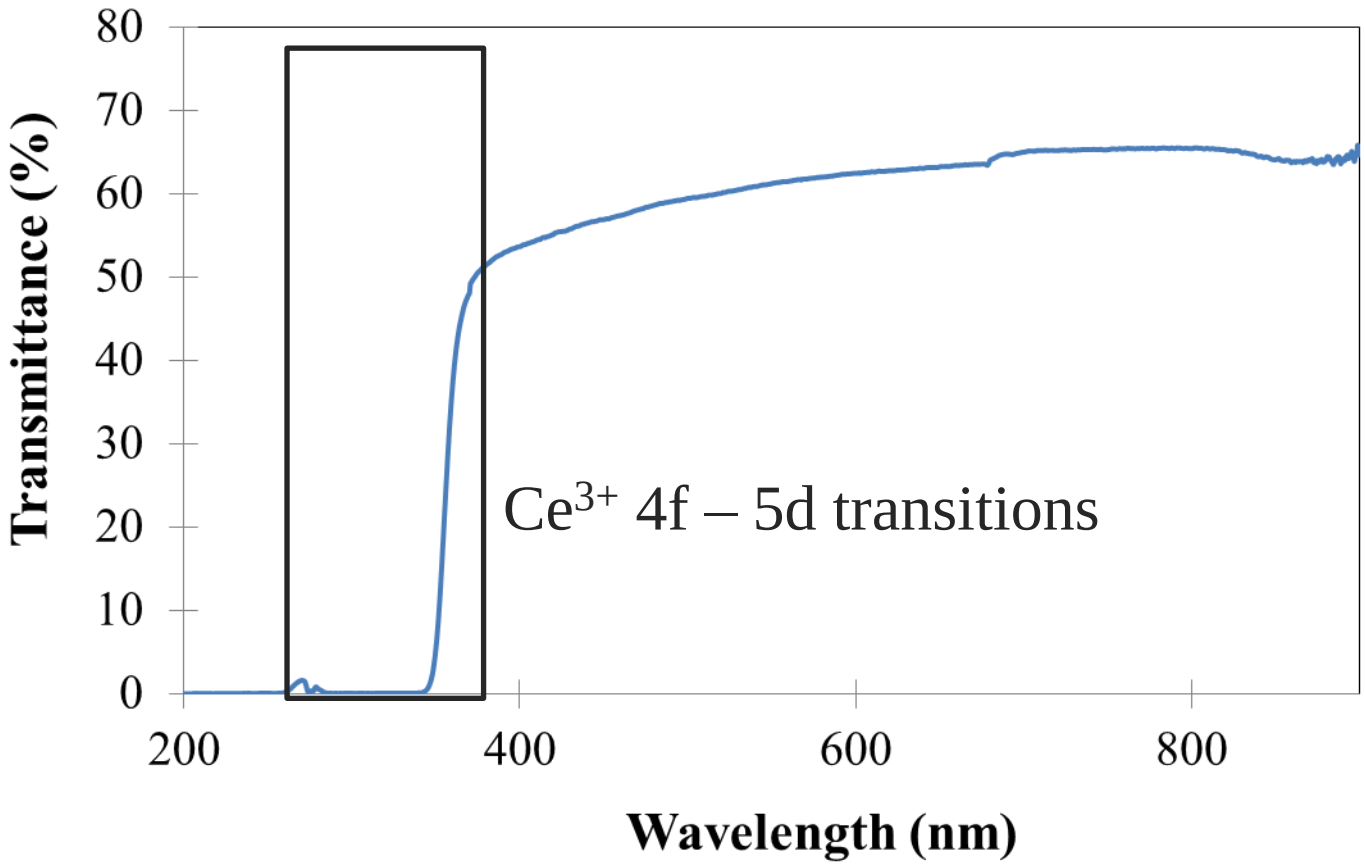


Under UV Lamp



Ce 1mol% doped
La-GPS Crystal

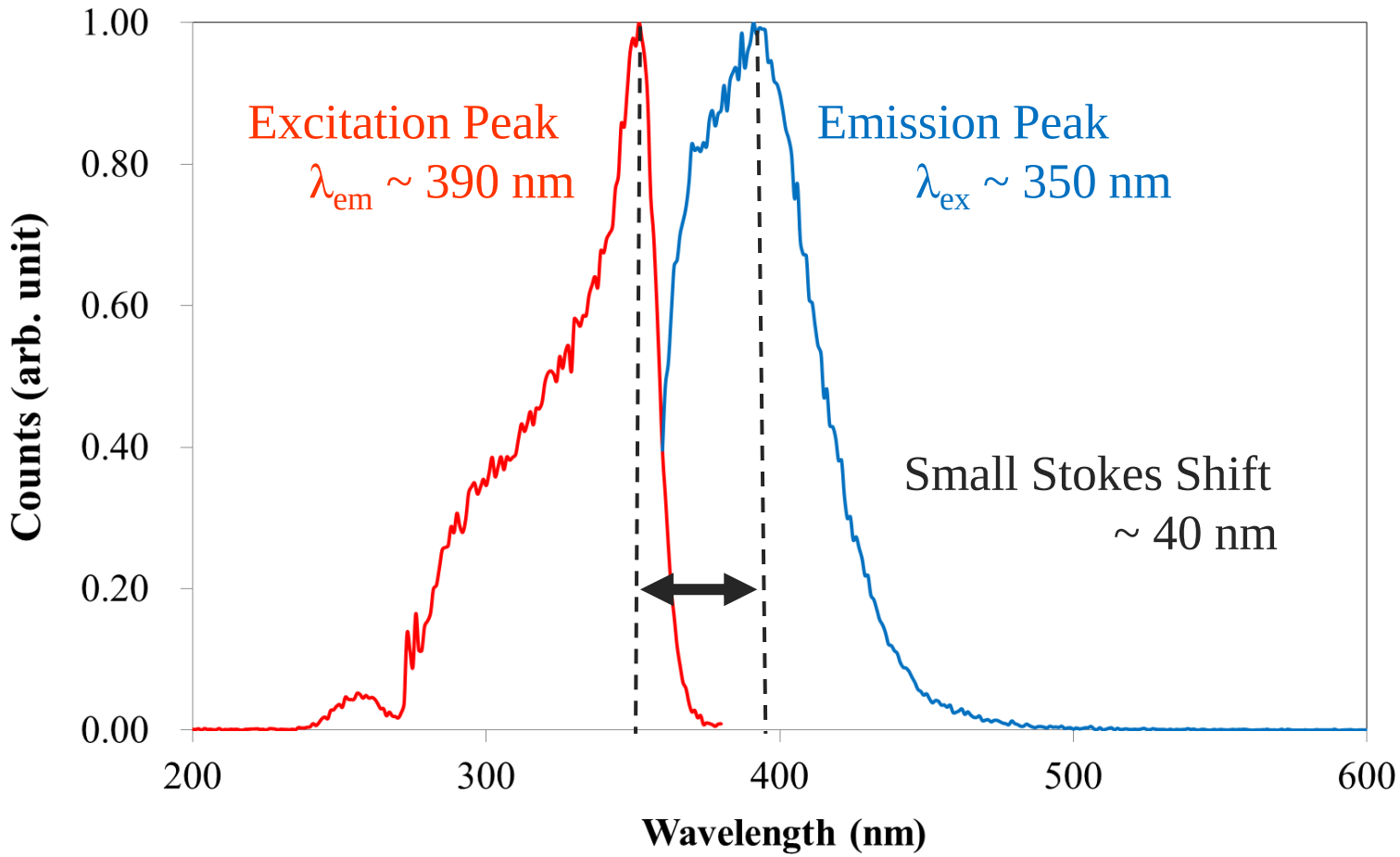
Transmittance



Measurement Instruments:

Spectrometer	V-530 (JASCO)
Excitation Source	Deutrium lamp (200 – 360 nm)
	Xe lamp (360 – 900 nm)

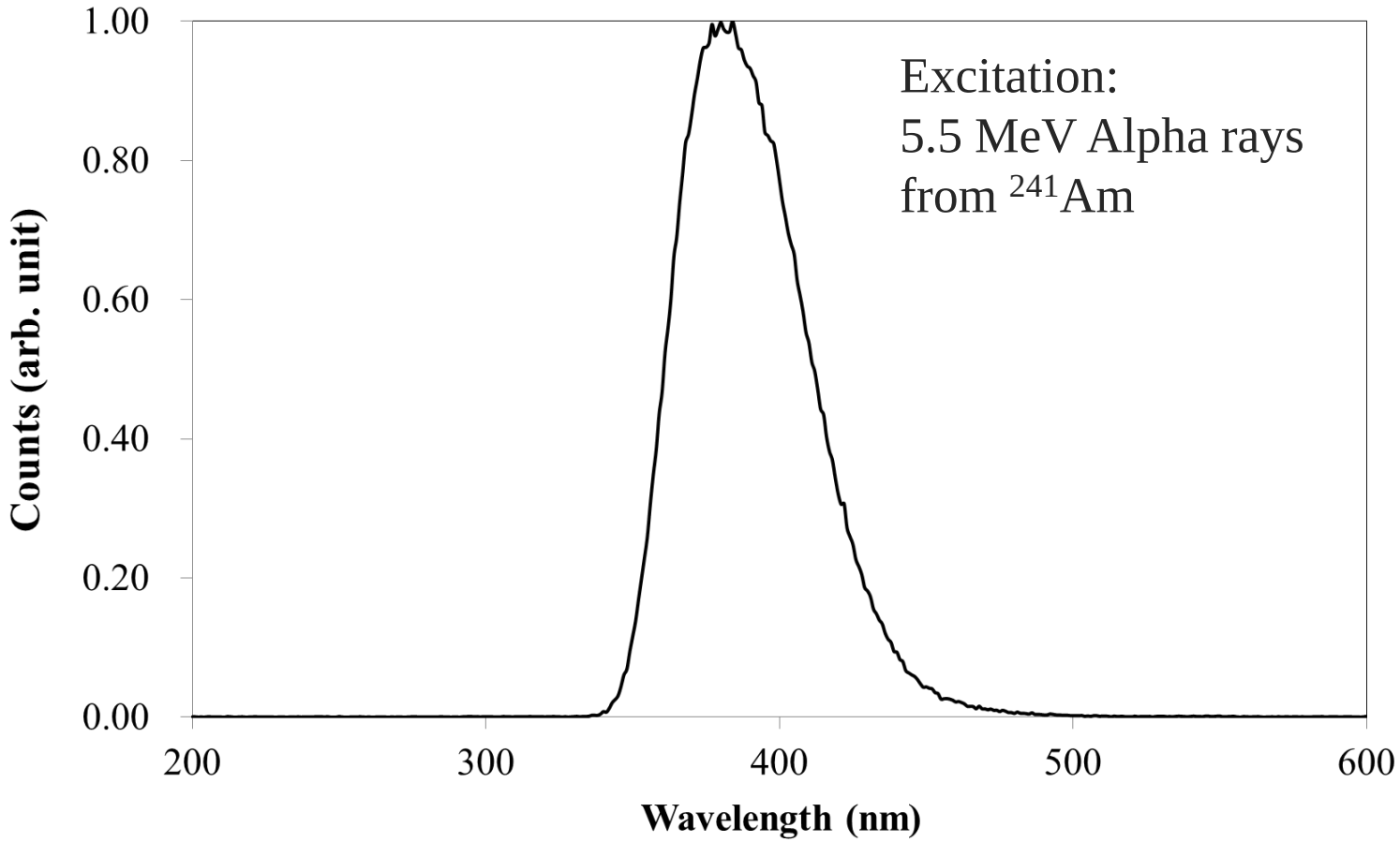
Photoluminescence



Measurement Instruments:

Spectrofluorometer	FLS920 (Edinburgh Instruments)
Excitation Source	Xe lamp Xe-900 (Edinburgh Instruments)

Scintillation Luminescence



Measurement Instruments:

Spectrofluorometer	FLS920 (Edinburgh Instruments)
Excitation Source	^{241}Am Alpha ray Source

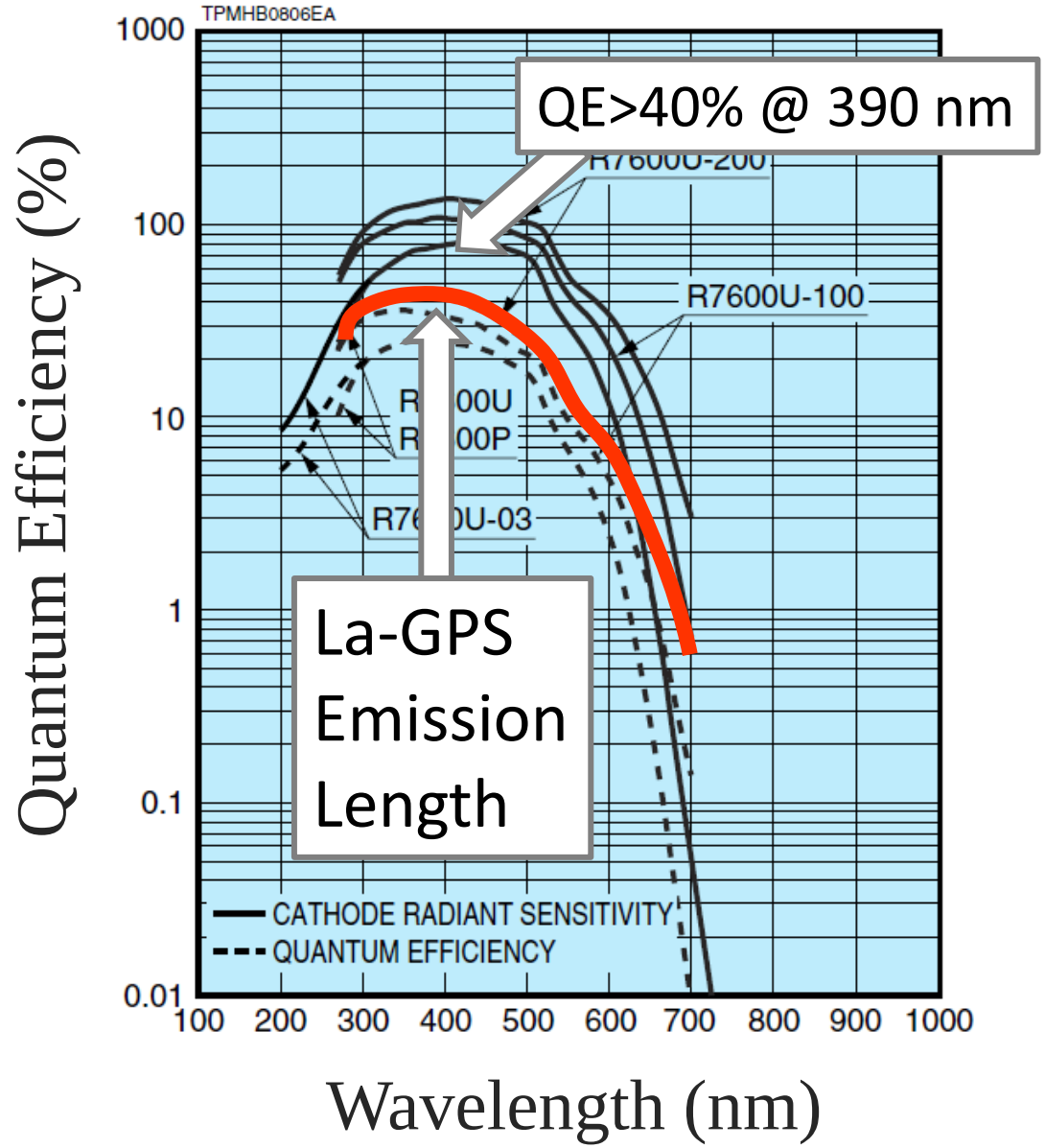
Photo-detector (I)

PMT

R7600U-200
Hamamatsu

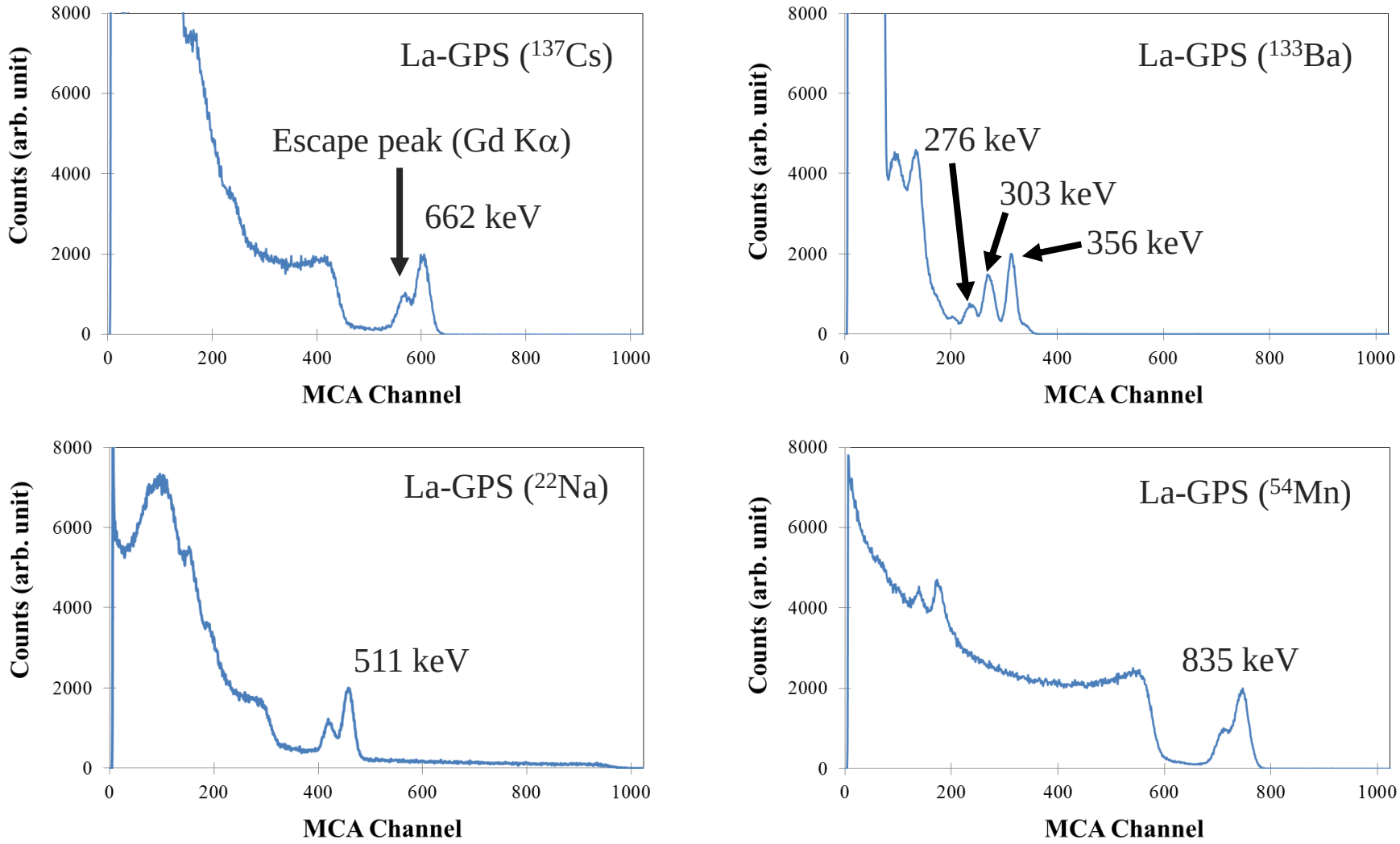
Ultra bi-alkali photocathode

Effective area: 18 x 18 mm



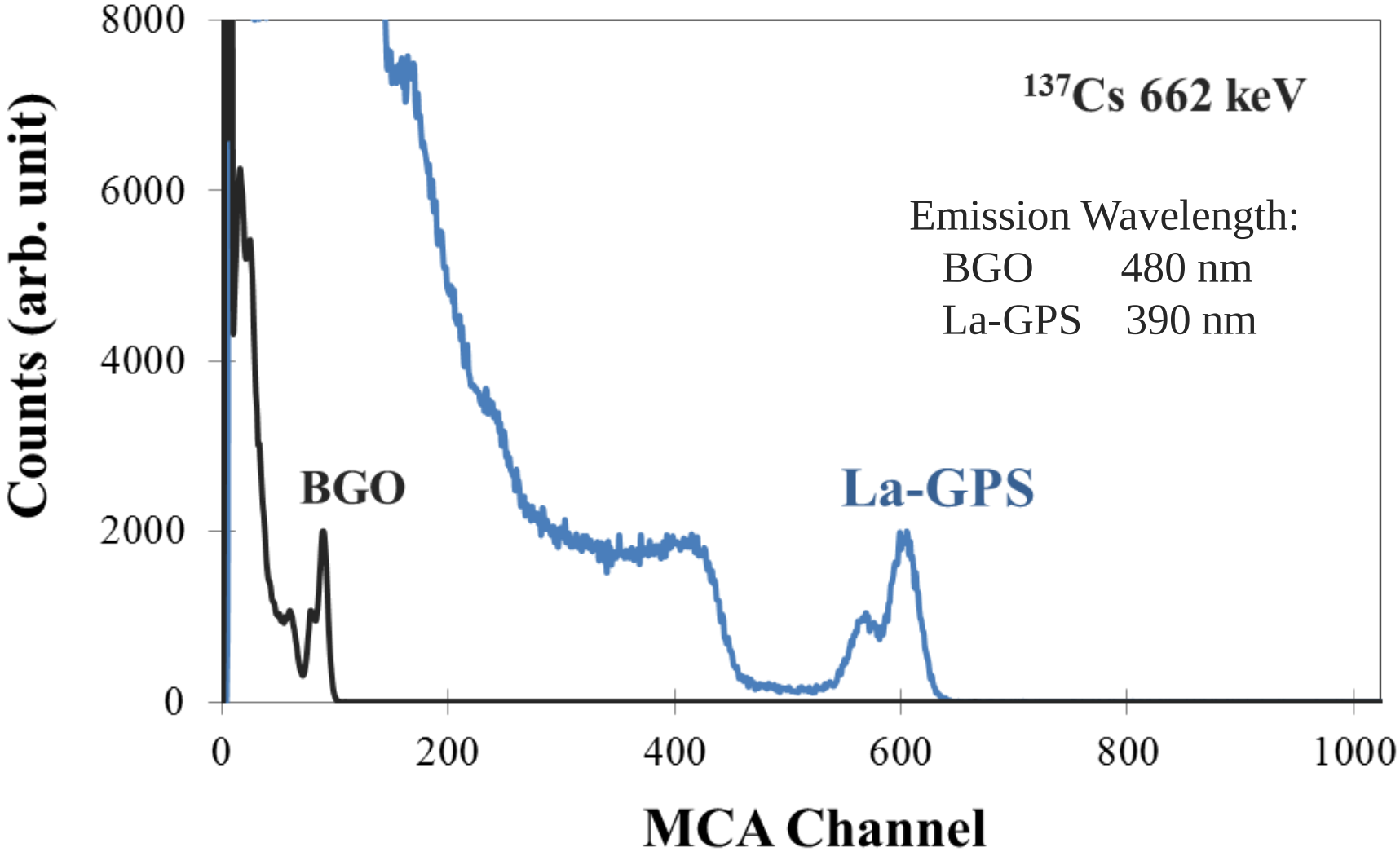
Reference: R7600U Data Sheet (HPK)

Pulse Height Spectra with PMT



Temperature: Constantly 26.0 ± 0.1 ℃ in all the measurements.
Preamplifier: ORTEC MODEL 113
Shaper: ORTEC MODEL 572A, Shaping Time: 1 us

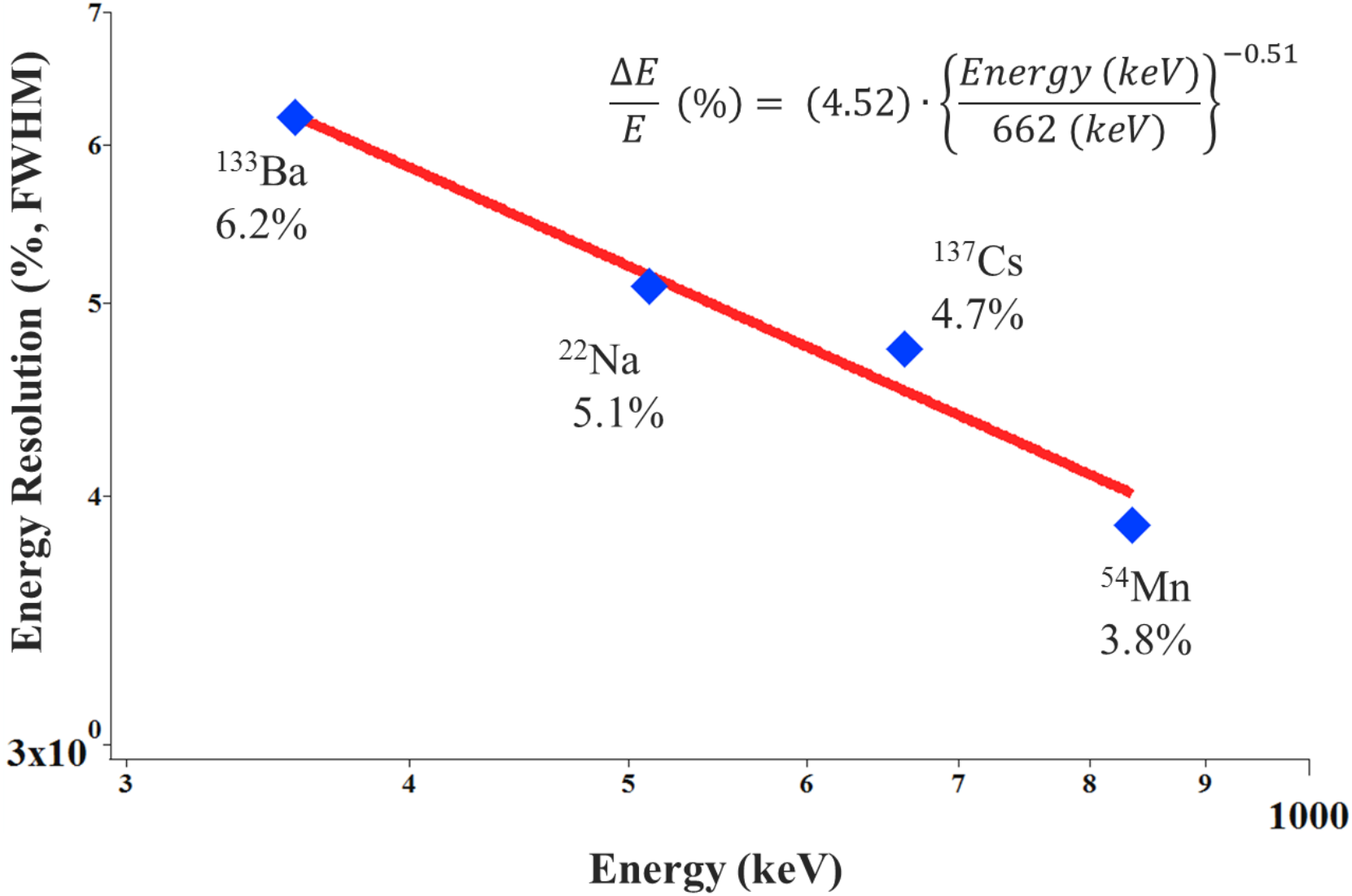
Light Output with PMT



When BGO ~ 8000 photon/MeV,

La-GPS ~ **39000** photon/MeV

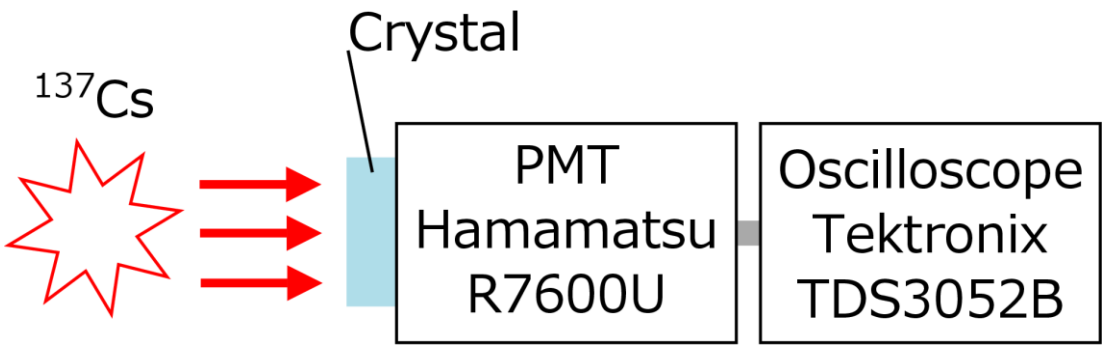
Energy Resolution with PMT



La-GPS Energy Resolution ~ **4.7**% FWHM at 662 keV

Scintillation Decay

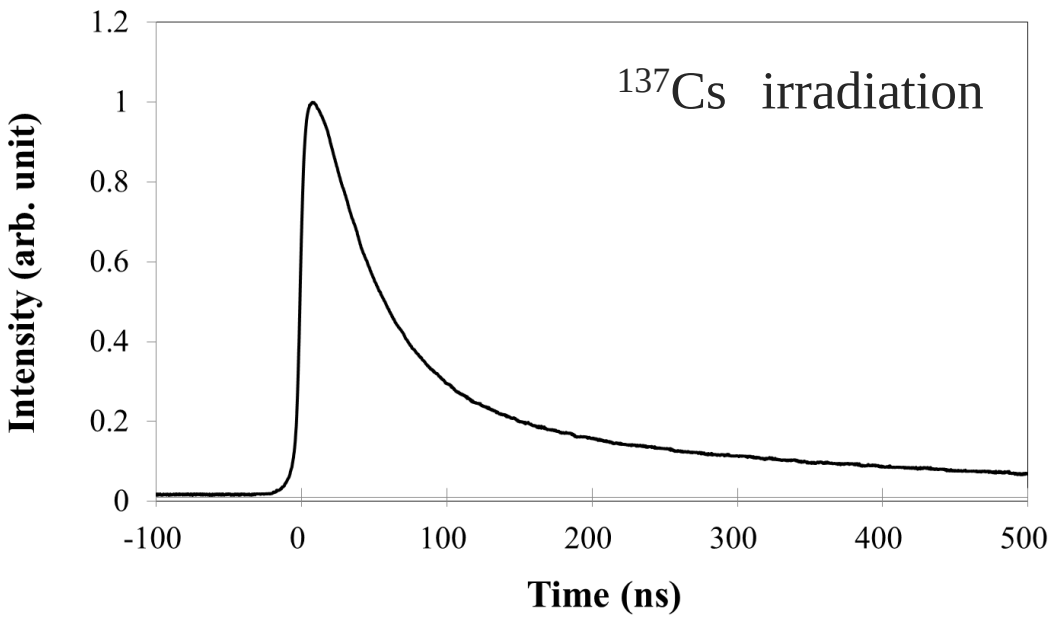
Measurement System



Exponential Fitting:

$$I = I_0 + \sum_{n=1}^{2} A_n \exp\left(-\frac{t - t_0}{\tau_n}\right)$$

I: Intensity, t: Time,
τ: Decay Constant

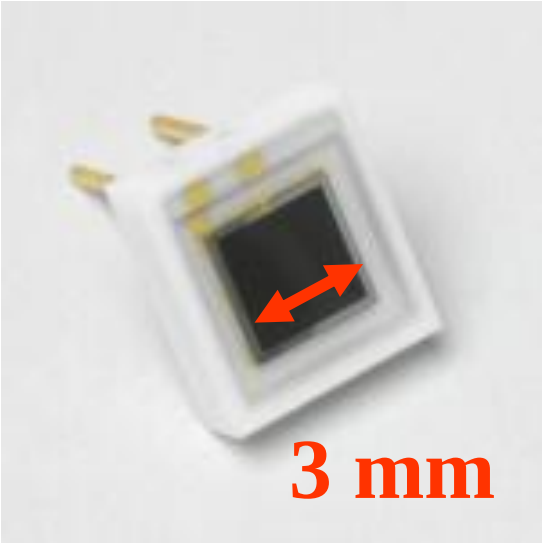


	τ_1	τ_2
Decay Time (ns)	**45.97**	345.6
Fitting Error (ns)	0.05	0.8
Ratio (%)	79	21

Photo-detector (II)

MPPC

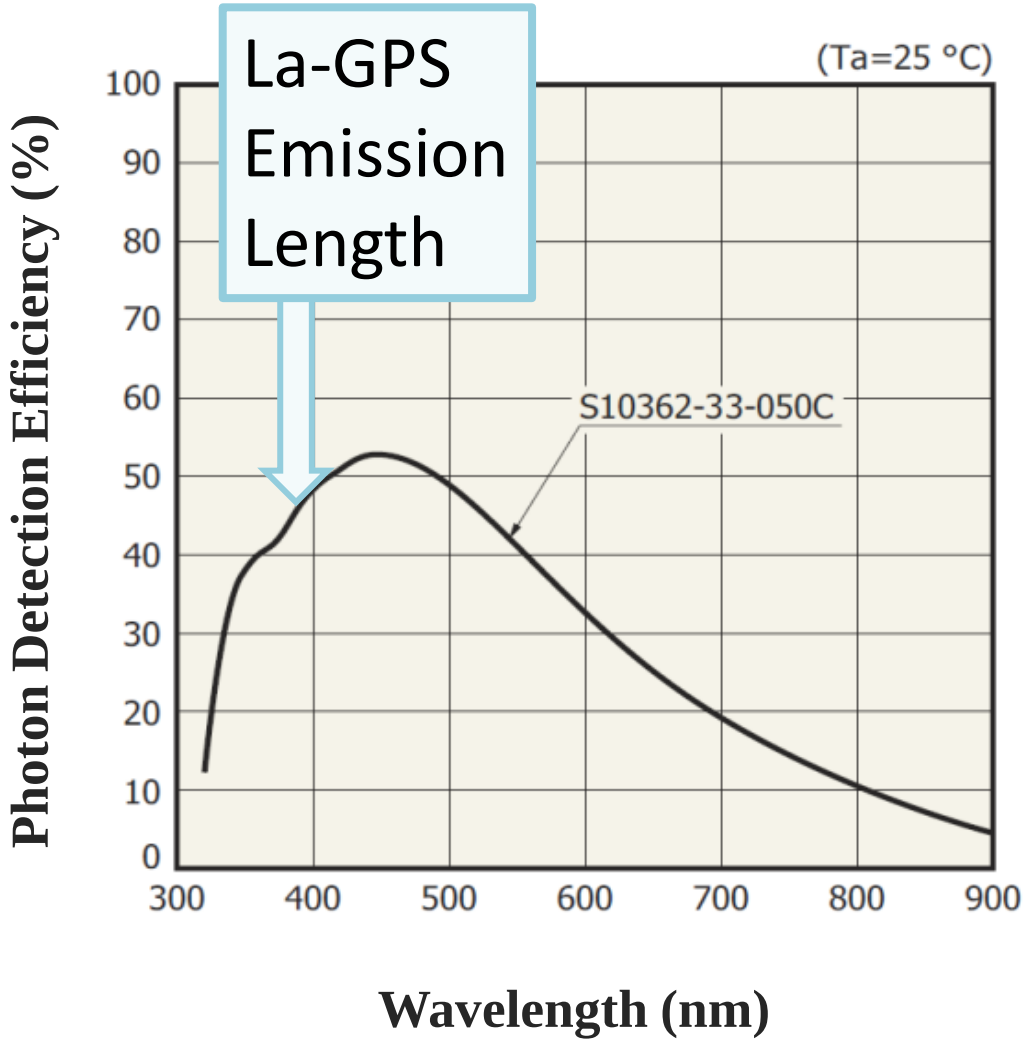
S10362-33-050C
Hamamatsu



Pixel Size: 50 x 50 um

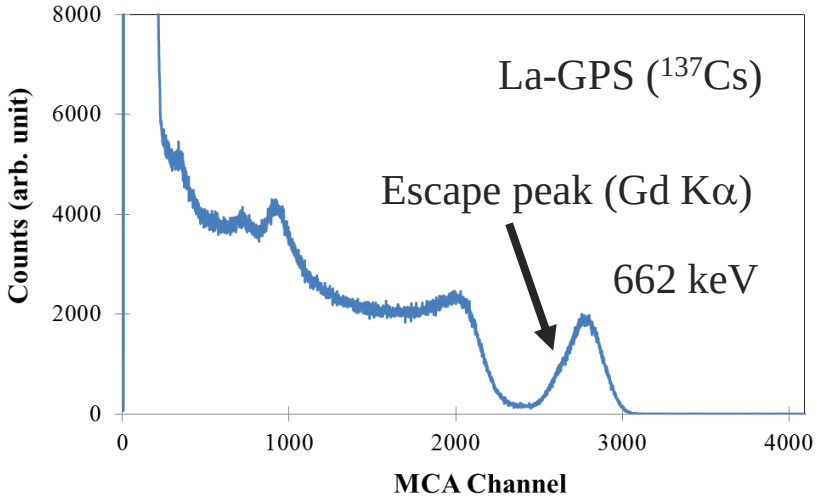
Number of Pixels: 3600

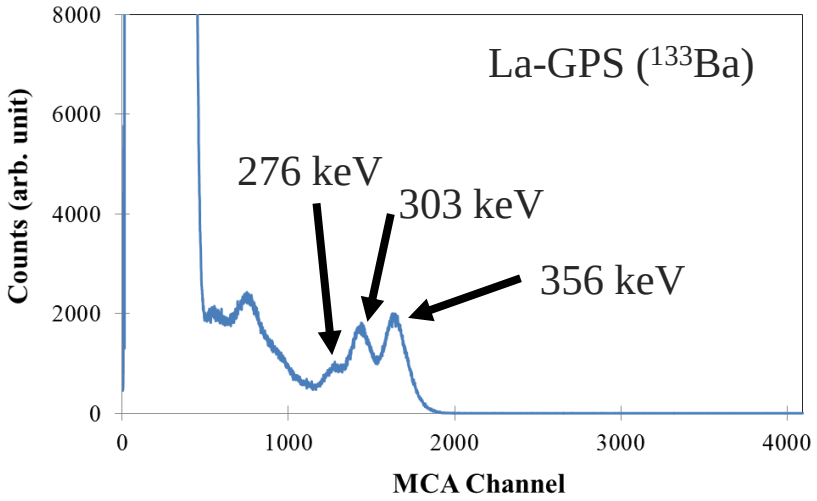
Effective area: 3 x 3 mm

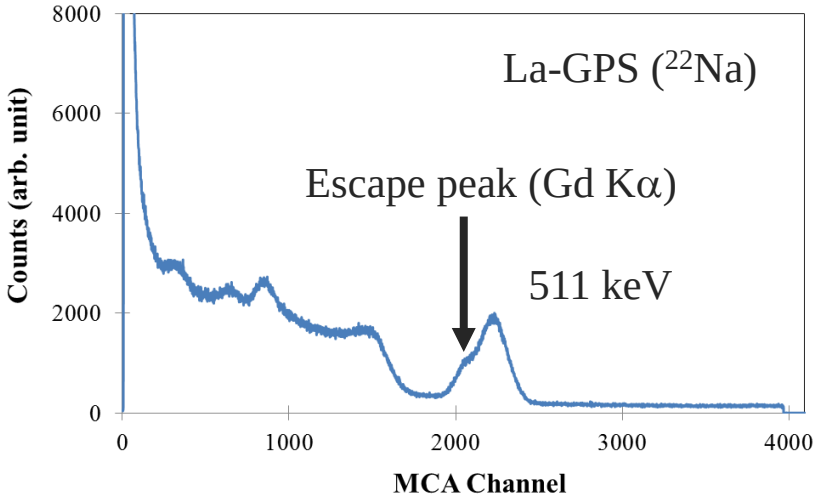


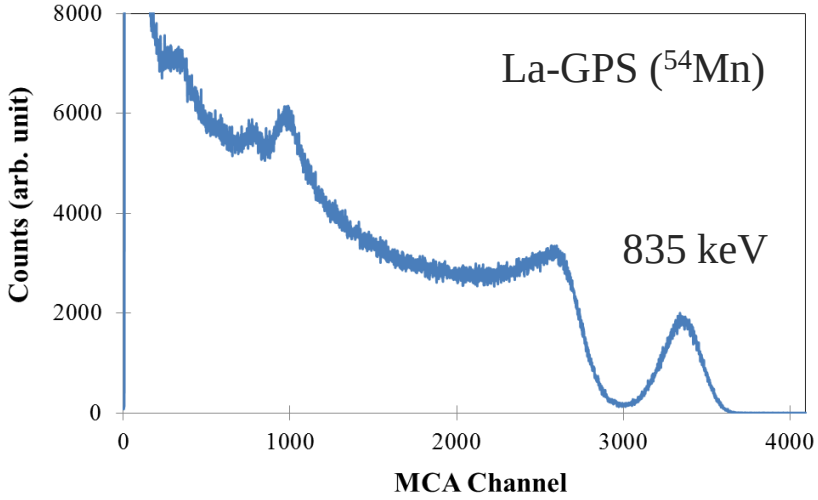
Reference: S10362-33-050C Data Sheet (HPK)

Pulse Height Spectra with MPPC



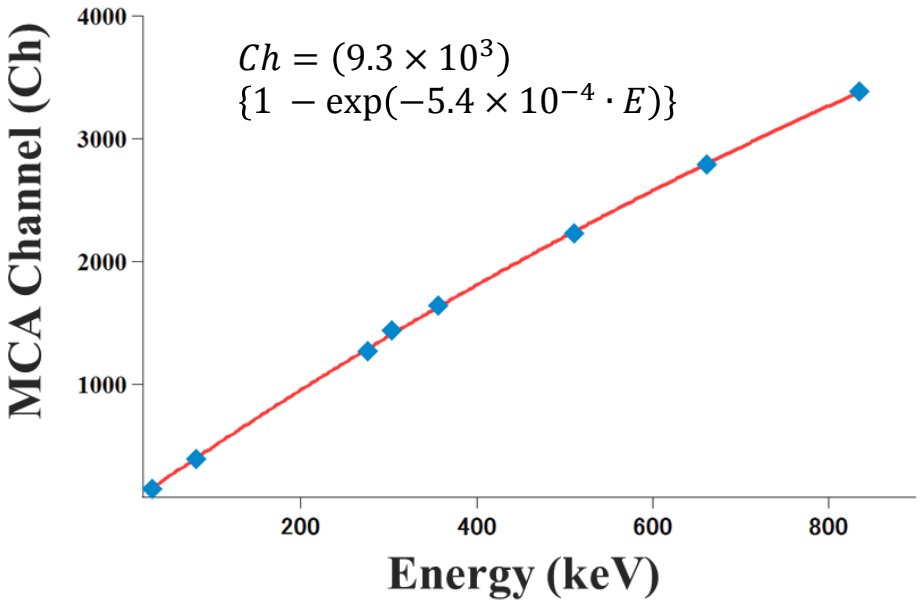






Temperature: Constantly 26.0 ± 0.1 ℃ in all the measurements.
Preamplifier: Time Constant 5.4 us
Shaper: ORTEC MODEL 572A, Shaping Time: 1 us

Energy Resolution with MPPC



Fitting Function

$$Ch = \beta\left\{1 - \exp\left(-\frac{N_{photon} \cdot E \cdot PDE}{N_{Pixel}}\right)\right\}$$

N_{photon} : Number of photons which traveled into MPPC
E : Energy of gamma rays
PDE : Photon detection efficiency
N_{Pixel} : Number of pixels
β : Constant

Energy (keV)	Radiation Source	Energy Resolution (%, FWHM)	
		MPPC	PMT
835	^{54}Mn	8.5	3.8
662	^{137}Cs	9.5	4.7
511	^{22}Na	10.2	5.1
356	^{133}Ba	10.7	6.2

Comparison of La-GPS with Other Scintillators

	Density (g/cm^3)	attenuation length[mm] @511keV	Decay time constant (ns)	Light output (Relative)	Hydroscopic nature	Radiation Hardness	energy res. @662 keV [%]
Tl:NaI	3.67	2.6	230	1	Strong	very weak	7
Tl:CsI	4.53	18.6	1050	0.85	Weak	very weak	7
BGO	7.13	large 1.1	300	0.07-0.12	No	weak	7
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Ce:GSO	6.71	1.3	30-60	0.18	No	very strong	8
Ce:$LaBr_3$	5.3	2.2	20	1.6	Very strong	strong	3
Ce:La-GPS	**---**	**---**	**46**	**0.6**	**No**	**---**	**4.7**

Summary

- **Novel Scintillator La-GPS was successfully developed.**
- **Although La-GPS is a oxide scintillator, it has a high light output and good energy resolution (FWHM) comparable with halide scintillators.**

Emission Wavelength (nm)	Light Output (photon/MeV)	Energy Resolution (%, FWHM at 662 keV)		Scintillation Decay Time (ns)	Hygroscopic Nature
		PMT	MPPC		
390	39000	4.7	9.5	45.97 (79%) 345.6 (21%)	None

Future Plan

- **Measurement of time resolution for La-GPS scintillator with a MPPC**