Recent Developments in the Inorganic Scintillator Field

C.W.E. van Eijk, M.D. Birowosuto, G. Bizarri
P. Dorenbos, J.T.M. de Haas, E. van der Kolk
H. Güdel, K. Krämer

IWORID 7 – Grenoble – July 4-7 – 2005

FNSNF
Fonds national suisse
Schweizerischer Nationalfonds
Fondo nazionale svizzero
Swiss National Science Foundation

Universität Bern

Saint-Gobain Crystals & Detectors

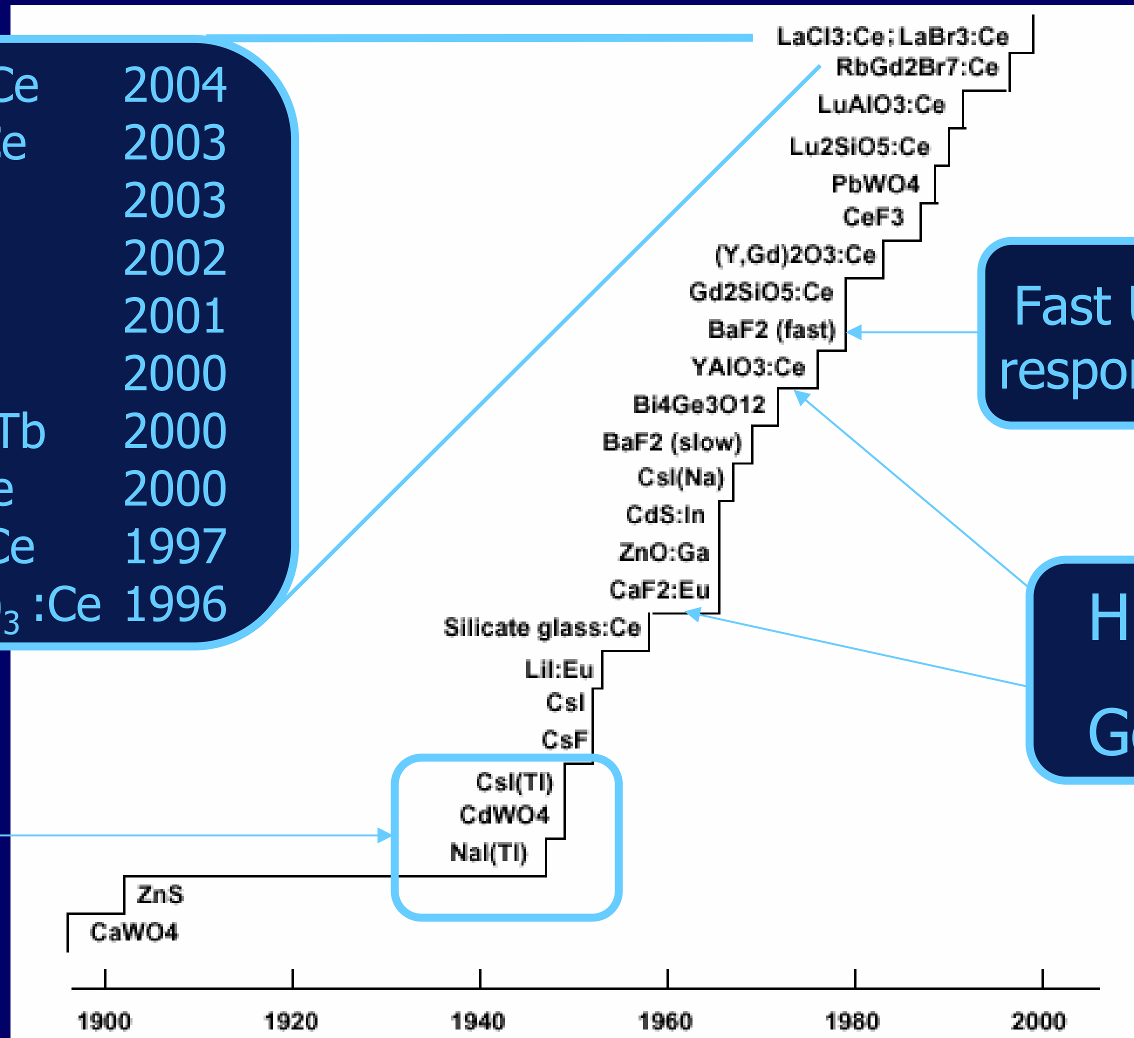
TU Delft
Delft University of Technology

History of scintillators

M. J. Weber

J. Lumin. 100 (2002) 35

Rb_2LiYBr_6:Ce	2004
Cs_2LiYCl_6:Ce	2003
LuI_3:Ce	2003
K_2LaI_5:Ce	2002
$LaBr_3$:Ce	2001
$LaCl_3$:Ce	2000
Lu_2O_3:Eu, Tb	2000
$Lu_2Si_2O_7$:Ce	2000
$RbGd_2Br_7$:Ce	1997
$^6Li_6Gd(BO_3)_3$:Ce	1996



Fast UV response

HPGe

Ge:Li

Invention of the photomultiplier tube

TU Delft

Many reviews:

M.J. Weber, *Inorganic scintillators: today and tomorrow*
J. Lumin. 100 (2002) 35-45

C.W.E. Van Eijk, *Inorganic scintillators in medical imaging detectors*
Nucl. Instr and Meth. A509 (2003) 17-25

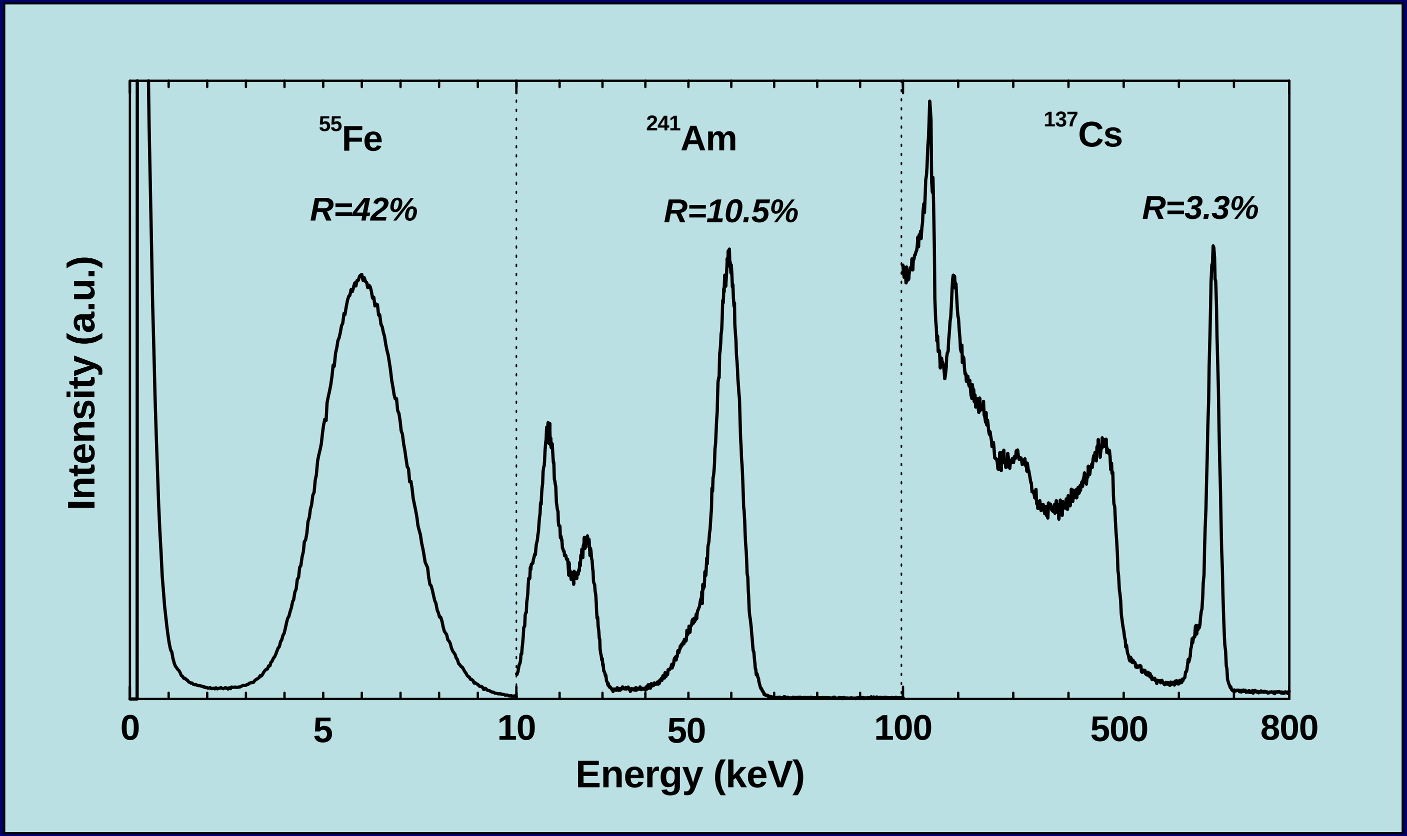
C.W.E. Van Eijk *et al*, *Inorganic thermal-neutron scintillators*
Nucl. Instr and Meth. A529 (2004) 260-267

C.L. Melcher, *Perspectives of the future development of new scintillators*
Nucl. Instr and Meth. A537 (2005) 6-14

P. Lecoq, *Ten years of lead tungstate development*
Nucl. Instr and Meth. A537 (2005) 15-21

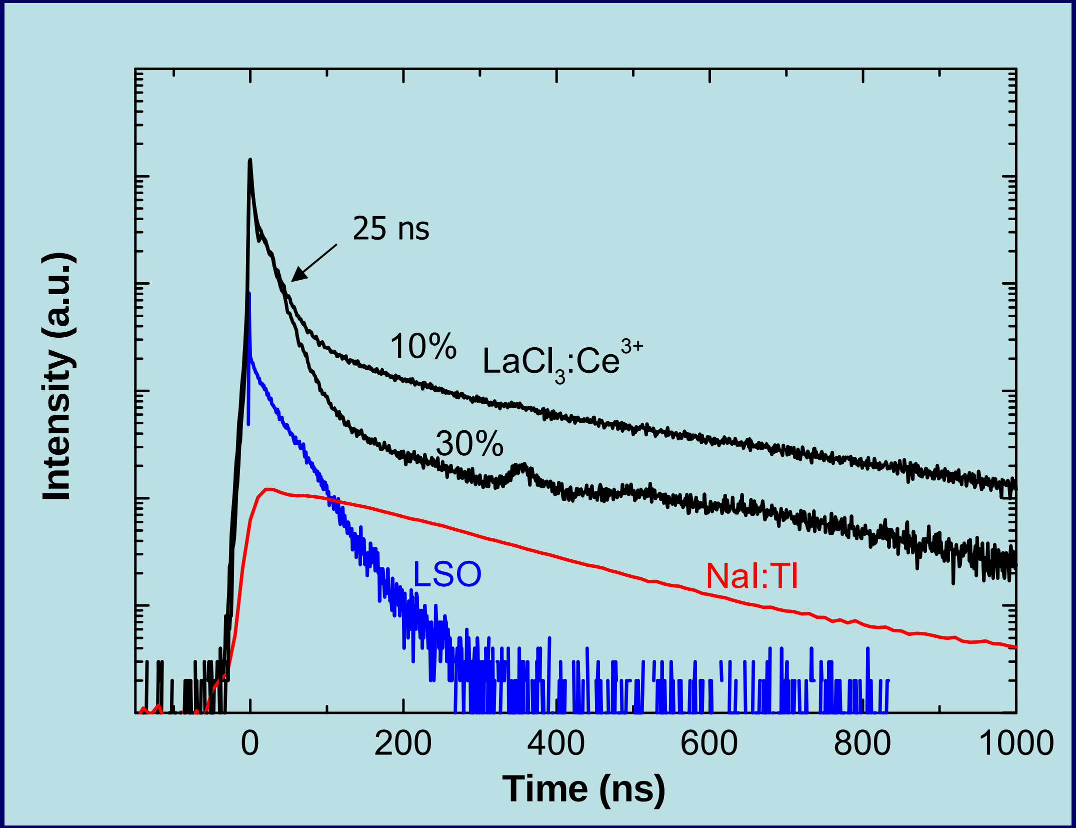
The New Scintillators

$LaCl_3$:Ce energy resolution



E.V.D. van Loef, P. Dorenbos, C.W.E. van Eijk, K.W. Krämer, H.U. Güdel
Appl. Phys. Lett. 77 (2000) 1467

$LaCl_3$:Ce scintillation decay



$LaCl_3$:Ce

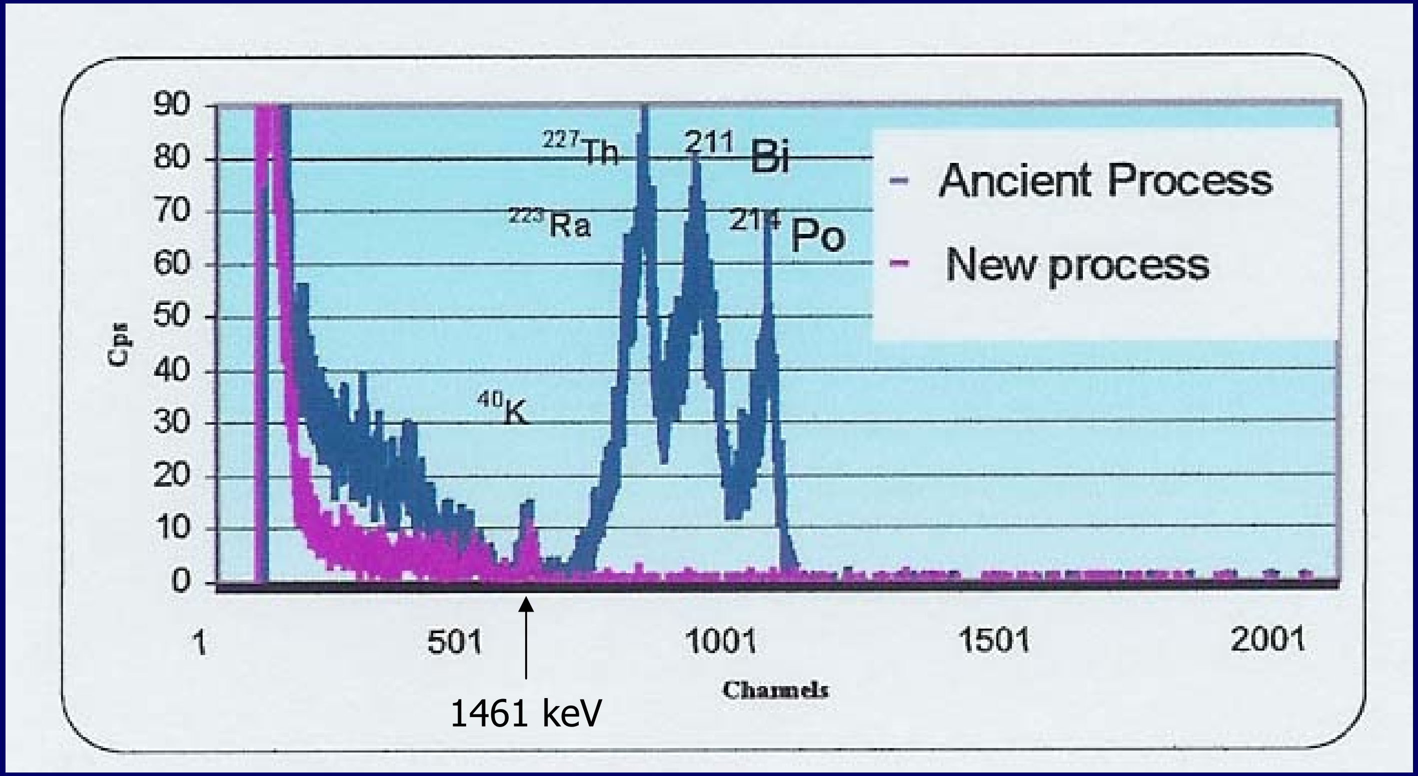
- 3″ x 3″
- 1″ x 1″

courtesy Saint-Gobain Crystals & Detectors

$LaCl_3$:Ce

Background of 1″x 1″crystals

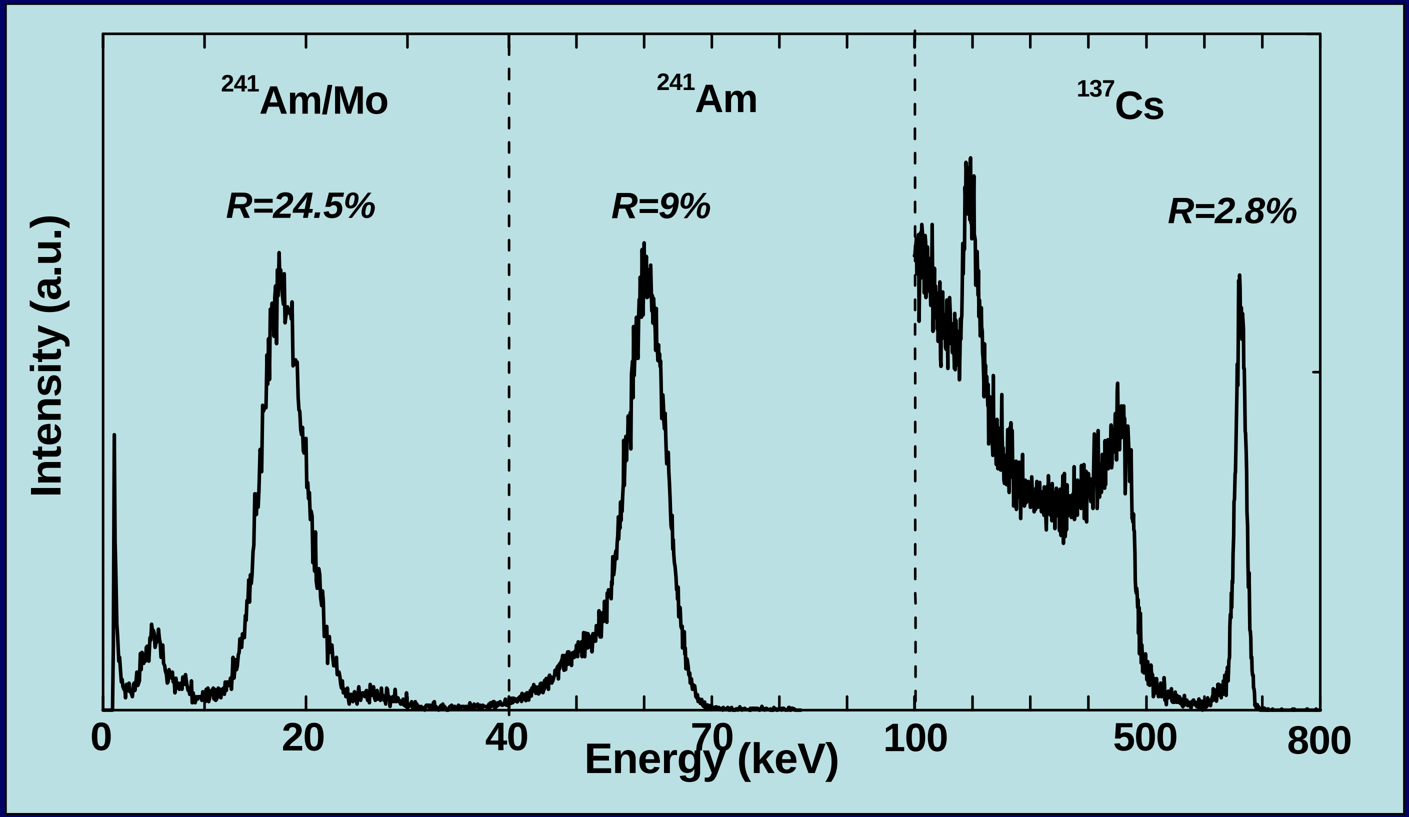
Technology from Ultra-Low Background NaI:Tl



0.05 Bq/cm^3

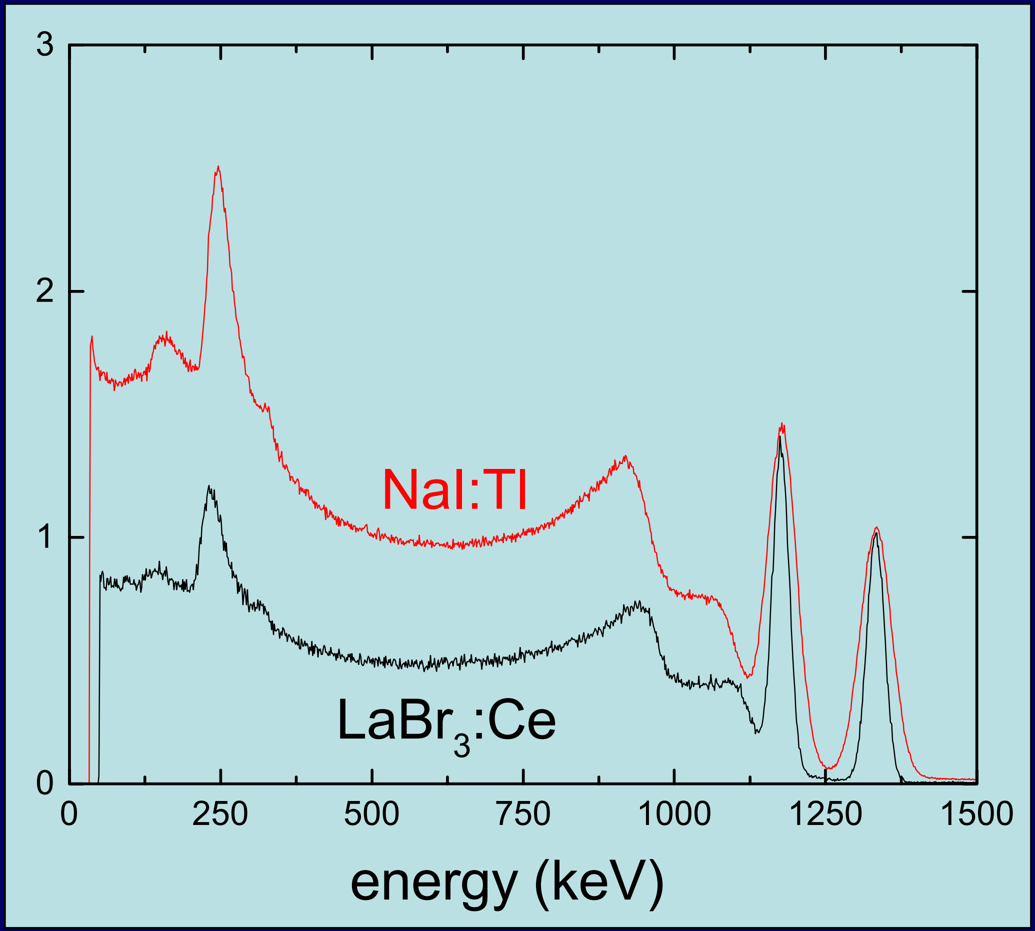
courtesy Saint-Gobain Crystals & Detectors

$LaBr_3$:0.5 % Ce^{3+} Energy Resolution

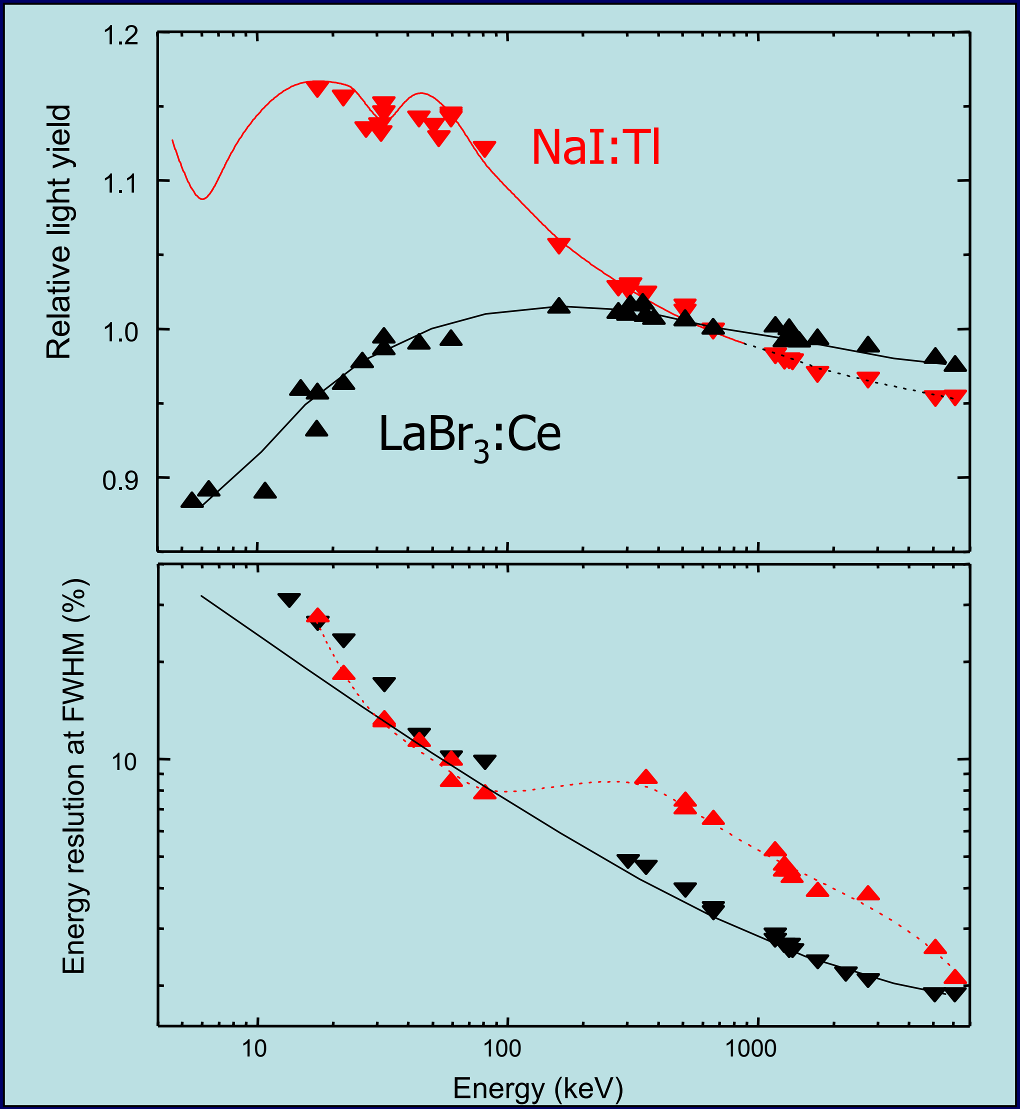


E.V.D. van Loef, P. Dorenbos, C.W.E. van Eijk, K.W. Krämer, H.U. Güdel
Appl. Phys. Lett. 79 (2001) 1573

^{60}Co spectrum measured with prototype ∅**19x19 mm^3** **$LaBr_3$:0.5 % Ce** scintillator



Non-Proportionality and Energy Resolution



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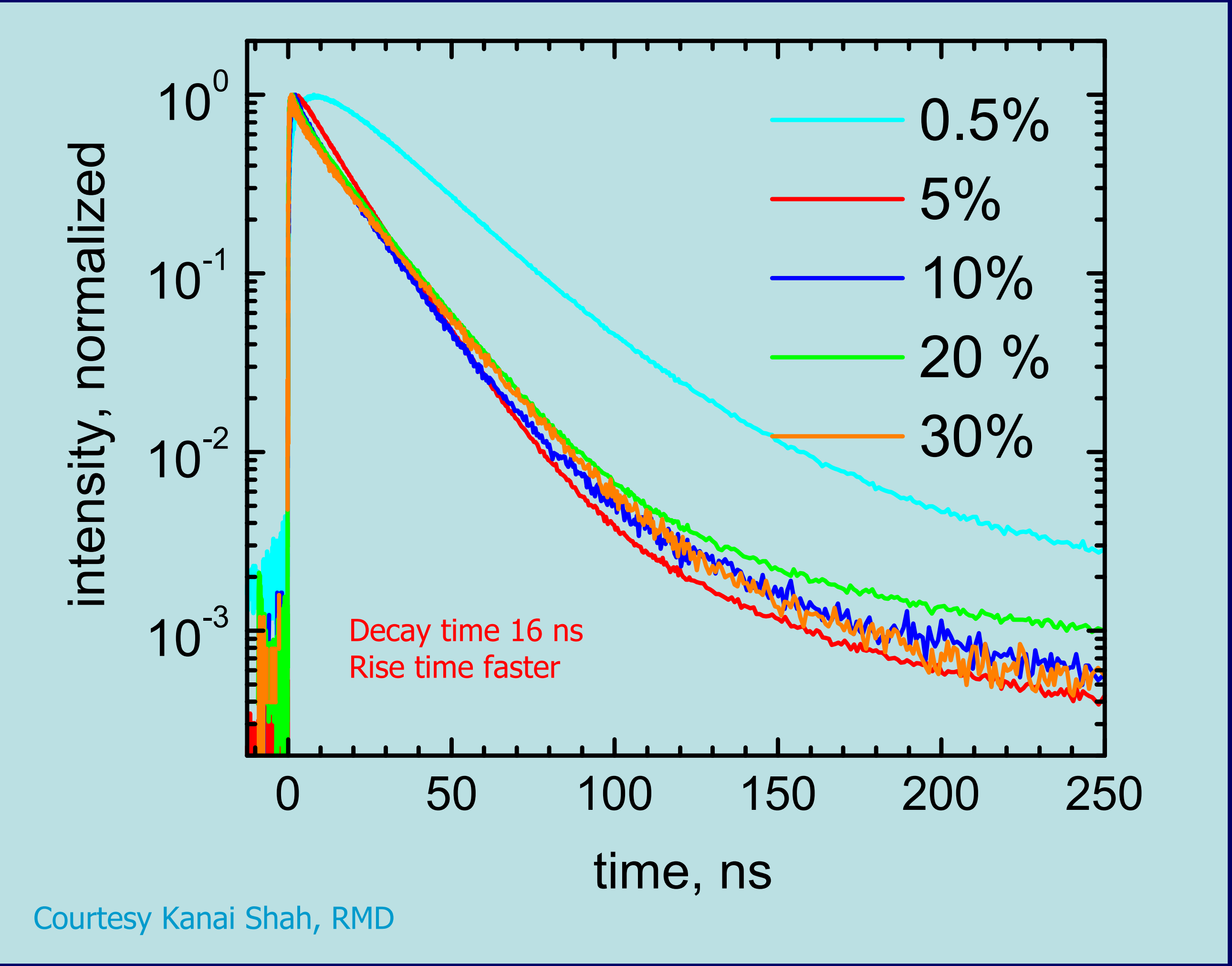
Inorganic Scintillators

Decay Time for $LaBr_3$:Ce & Time Resolution



511 keV - 511 keV

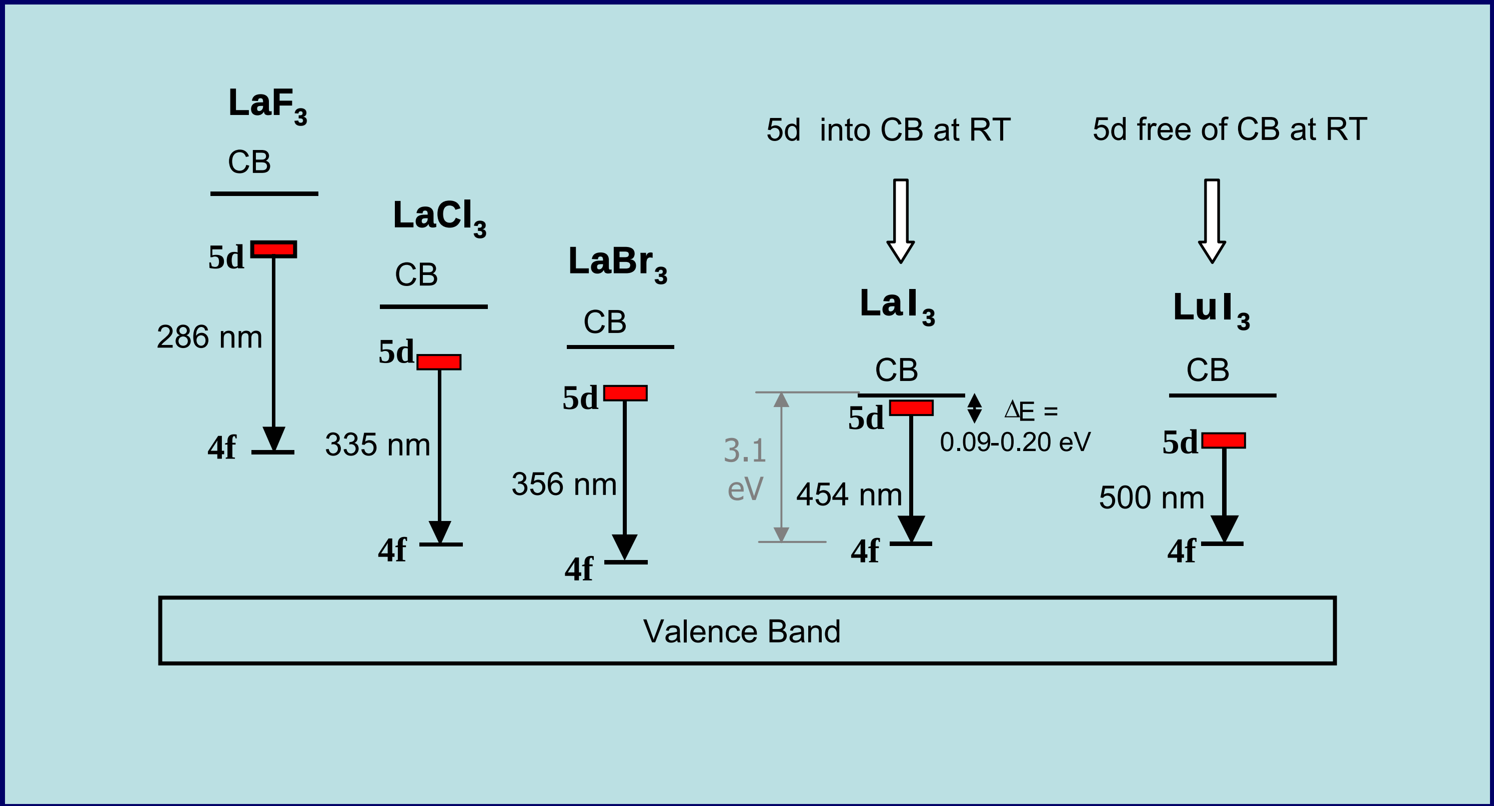
< 300ps

Inorganic Scintillators

Scintillator	Size (mm^3)	ρ (g/cm^3)	$Z_{eff.}$	$\lambda_{max.}$ (nm)	N (ph/MeV)	R (%)	τ (ns)	ref.
$LaBr_3$: 0.5% Ce	∅3x10	5.29	46.9	358	61,000	2.9	18 (90%)	[1]
$LaCl_3$:10% Ce	∅8x5	3.86	49.7	335	49,000	3.1	25 (41%)	[2]
$RbGd_2Br_7$:9.8% Ce	15x5x1	4.79	50.6	420	56,000	4.1	43 (56%)	[3]
NaI:Tl	∅25x12.5	3.67	50.8	415	40,000	6.5	230	[4]
CsI:Tl	∅3x5	4.51	53.7	540	64,800	4.3	600-800	[5]
$YAlO_3$:Ce	3x3x20	5.5	33.6	350	21,400	4.4	25	[6]

[1] E.V.D. van Loef, P. Dorenbos, C.W.E. van Eijk, K. W. Krämer, H.U. Güdel, Appl. Phys. Lett. 79 (10) (2001) 1573.
[2] E.V.D. van Loef, P. Dorenbos, C.W.E. van Eijk, K. W. Krämer, H.U. Güdel, Appl. Phys. Lett. 77 (10) (2000) 1467.
[3] O. Guillot-Noël, J.C. van't Spijker, J.T.M. de Haas, P. Dorenbos, C.W.E. van Eijk, K.W. Krämer, H.U. Güdel, IEEE Trans. Nucl. Sci. 46 (5) (1999) 1274.
[4] D.R. Kinoch, W. Novak, P. Raby, I. Toepke, IEEE Trans. Nucl. Sci. 41 (1994) 752.
[5] C. Fiorini, F. Perotti, Nucl. Instr. Meth. A 401 (1997) 104
[6] M. Kapsuta, M. Blacerzyk, M. Moszynski, J. Pawelke, Nucl. Instr. Meth. Phys. Res. A 421 (1999) 610.

Ce energy levels in the gap of the host



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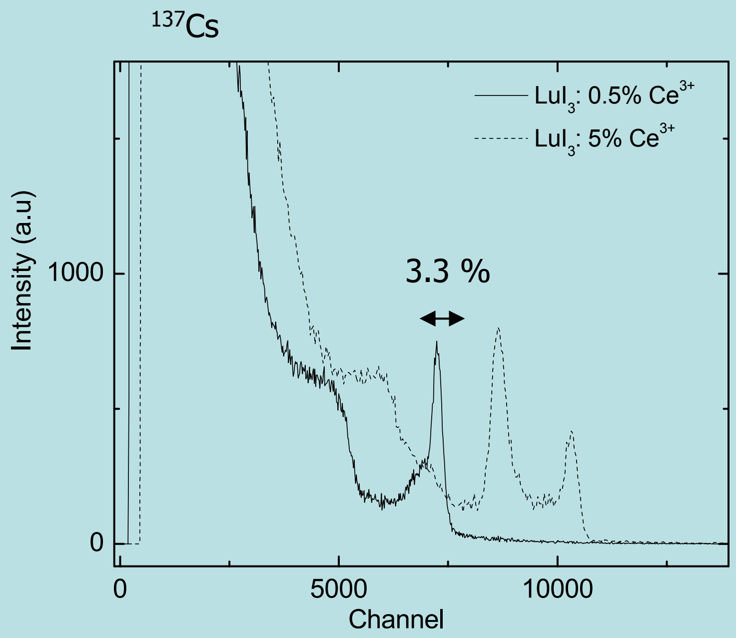
$LuI_3: Ce^{3+}$ Gamma-ray spectroscopy with APD

Photonic 630-70-73-500 APD, HV =1600 V
T = 278 K, Crystal size = 8 x 6 x 2 mm^3



Shaping time = 500 ns
Crystal size = 8 x 6 x 2 mm

^{137}Cs

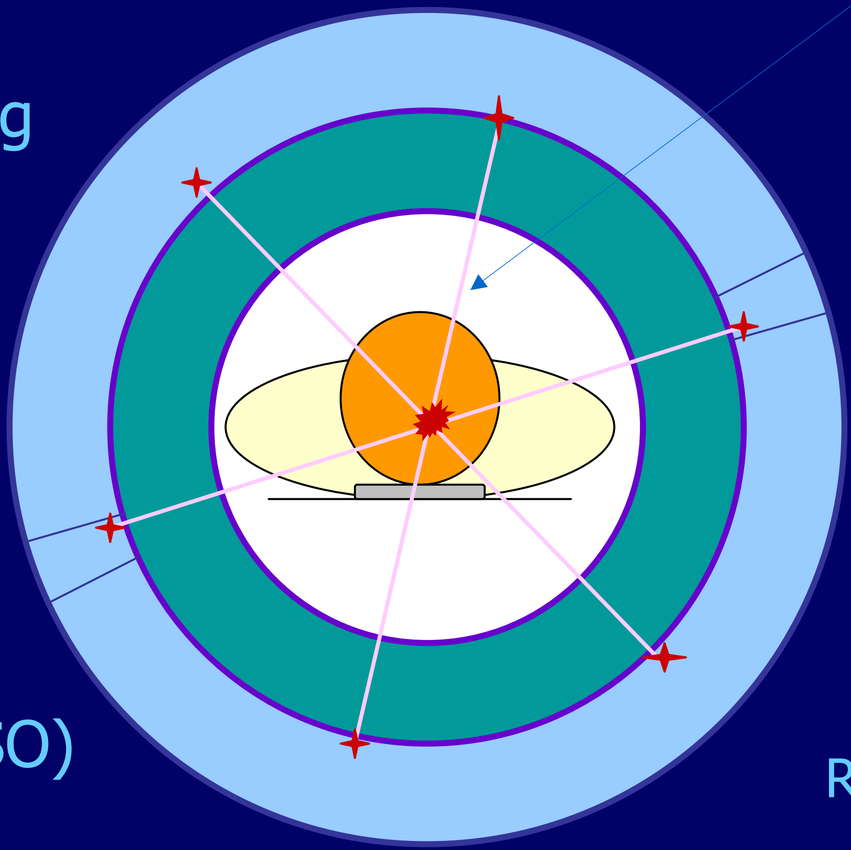
Compound	Electron hole pairs (10^3 e-h pairs/MeV)		Energy Resolution R (%)
	0.5 μs	10 μs	
LuI_3: 0.5% Ce^{3+}	50 ± 5	65 ± 6	3.3 ± 0.3
LuI_3: 2% Ce^{3+} (Two photopeaks)	58 ± 5 65 ± 6	73 ± 7 82 ± 8	- -
LuI_3: 5% Ce^{3+} (Two photopeaks)	60 ± 6 72 ± 6	83 ± 8 92 ± 9	- -

Time Resolution better than with $LaBr_3$:Ce

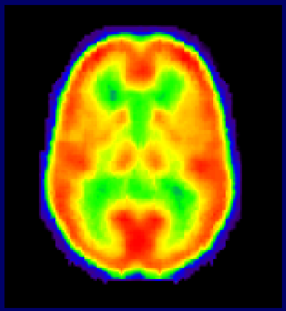
Scintillators in Positron Emission Tomography (PET)

PET basics - Imaging

Detector ring



Collinearly emitted annihilation quanta detected in coincidence



Detectors:
Scintillator
(BGO, LSO, GSO)
+ PMT

Radiopharmaceutical
positron emitter

PET Scintillators

	ρ (g/cm³)	1/μ 511 keV (mm)/PE (%)	light yield (photons/MeV)	τ (ns)	λ (nm)
$Bi_4Ge_3O_{12}$ (BGO)	7.1	11.6 / 44	9,000	300	480
Gd_2SiO_5:Ce (GSO)	6.7	15 / 26	8,000	60	440
Lu_2SiO_5:Ce (LSO + LYSO)	7.4	12.3 / 34	26,000	40	420
$Lu_xY_{1-x}AlO_3$:Ce (LuYAP)	8.3	11.0 / 32	11,000	18	365
$Lu_2Si_2O_7$:Ce (LPS)	6.2	14.5 / 29	20,000	30	380

C.L. Melcher and J.S. Schweitzer, IEEE Trans. Nucl. Sci. 39(1992) 502

B.I. Minkov, Functional Materials 1(1994)103, W.W. Moses et al IEEE Trans. Nucl. Sci. 42((1995)275,
A. Lempicki et al IEEE Trans. Nucl. Sci. 42((1995)280

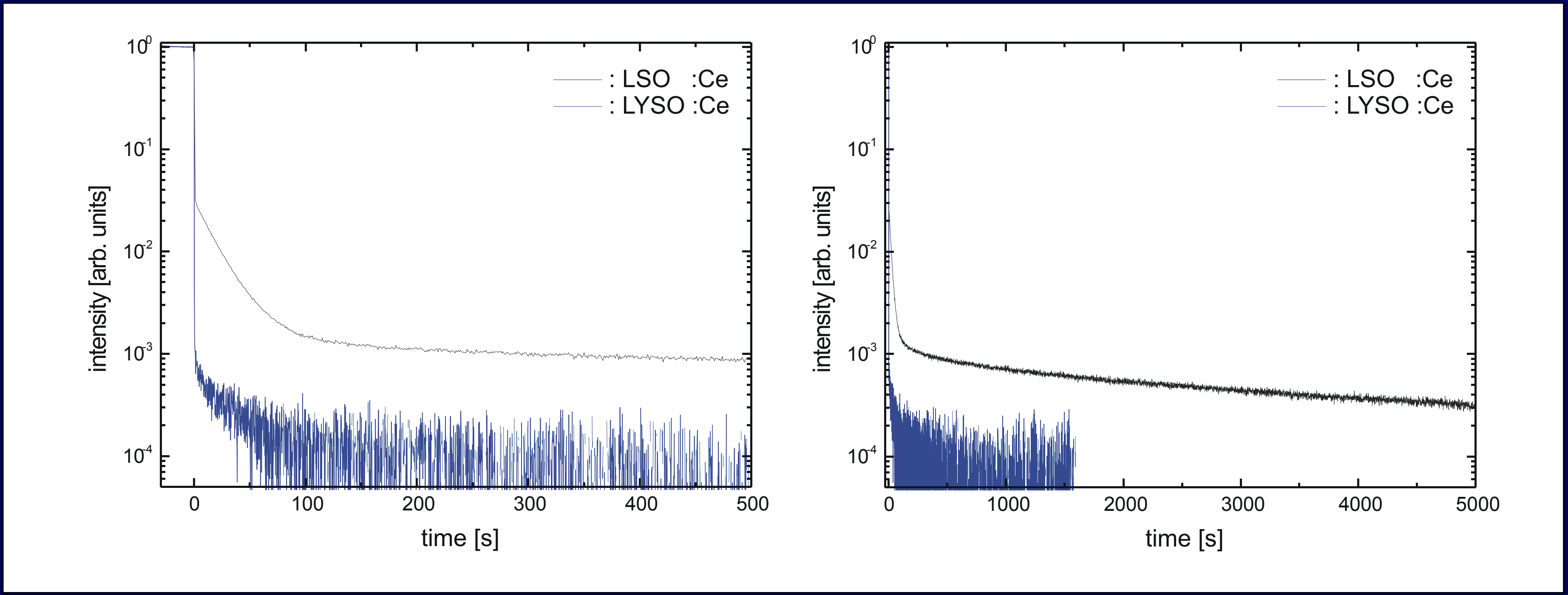
D. Pauwels et al Proc. SCINT 99, Moscow 2000, 511

Energy resolution poor

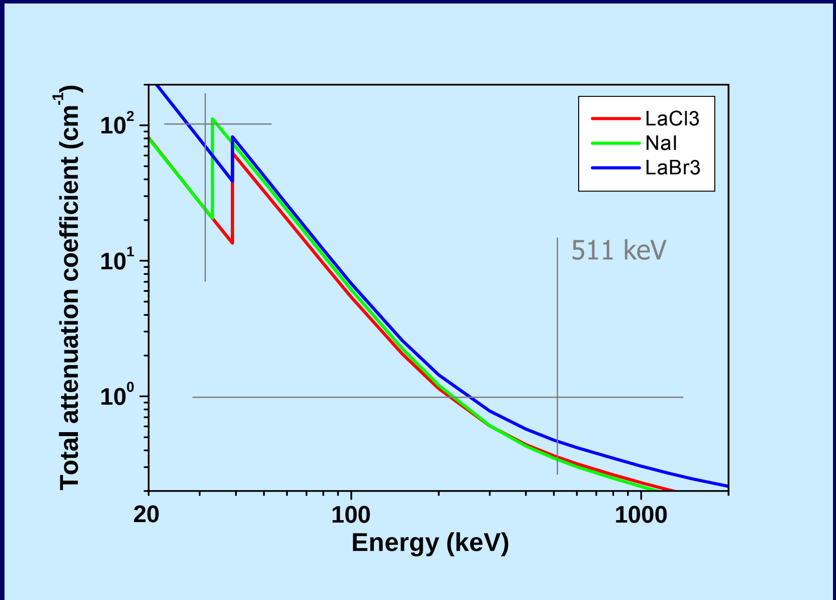
Lu_2SiO_5:Ce

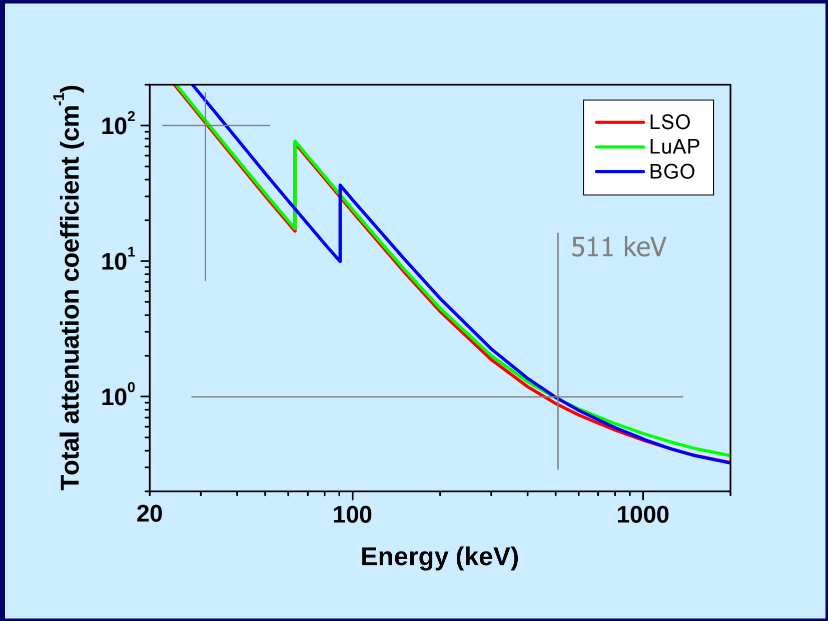
$Lu_{2(1-x)}Y_{2x}SiO_5$:Ce

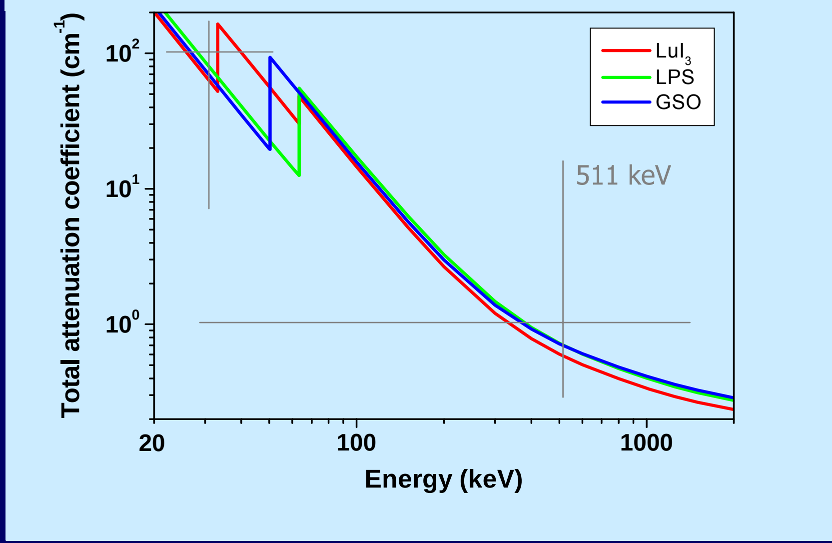
Less Afterglow



Inorganic Scintillators

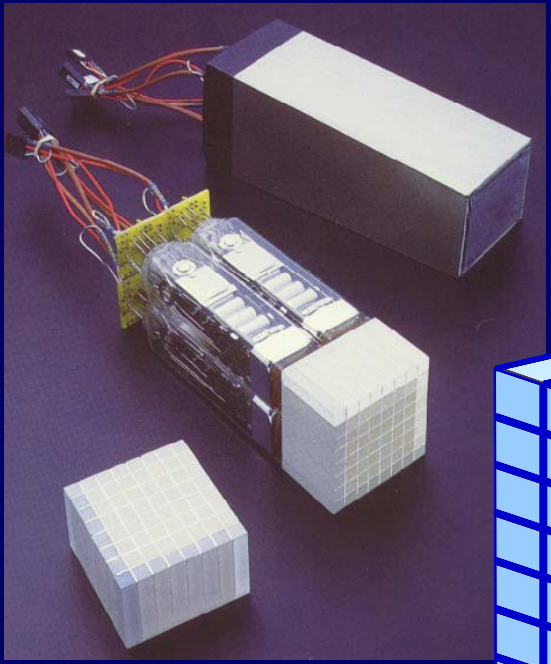


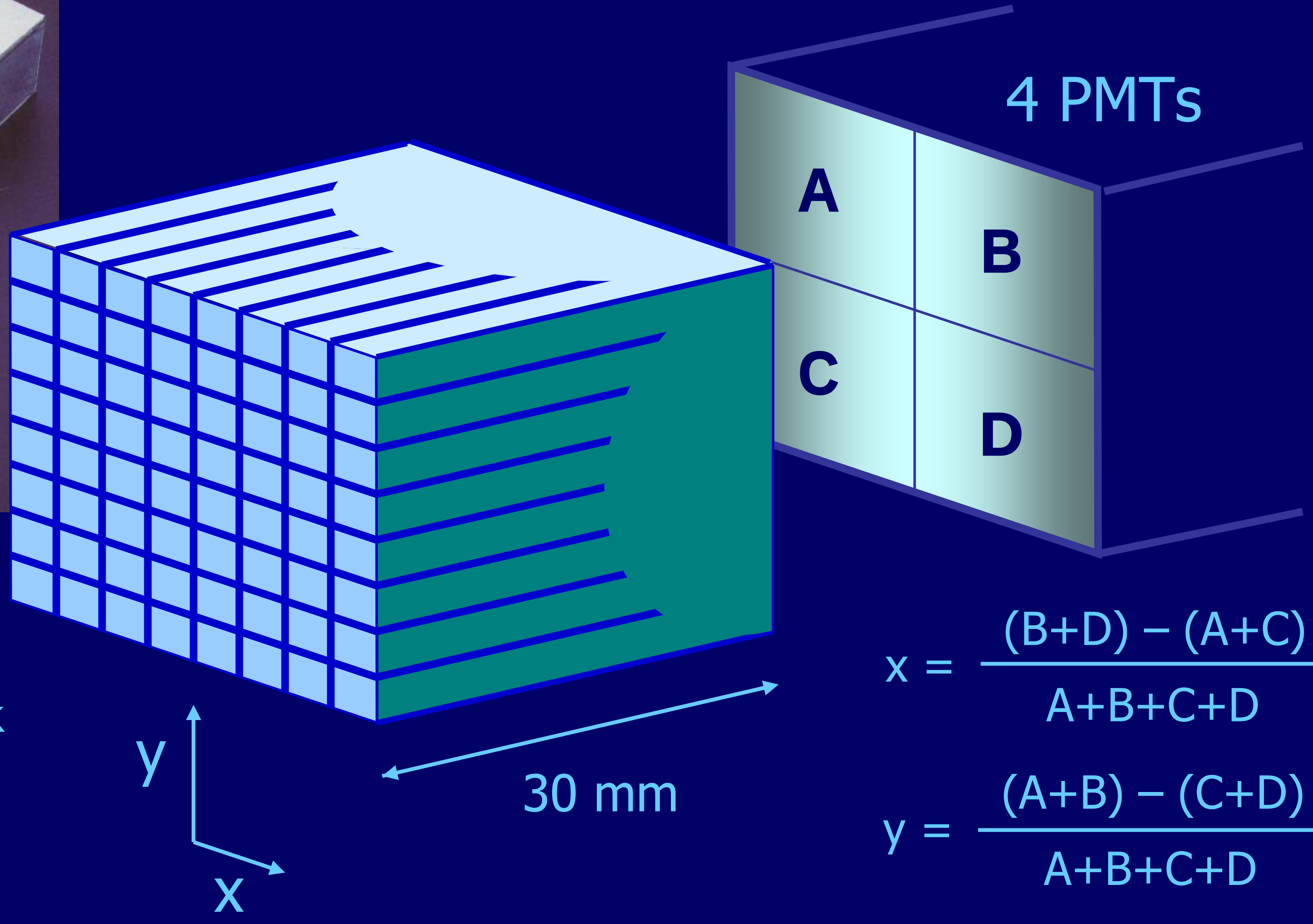




attenuation coefficients
of a number of
inorganic scintillators

PET Detector Block





BGO detector block
8 x 8 columns
of 6 x 6 x 30 mm^3

$$x = \frac{(B+D) - (A+C)}{A+B+C+D}$$

$$y = \frac{(A+B) - (C+D)}{A+B+C+D}$$

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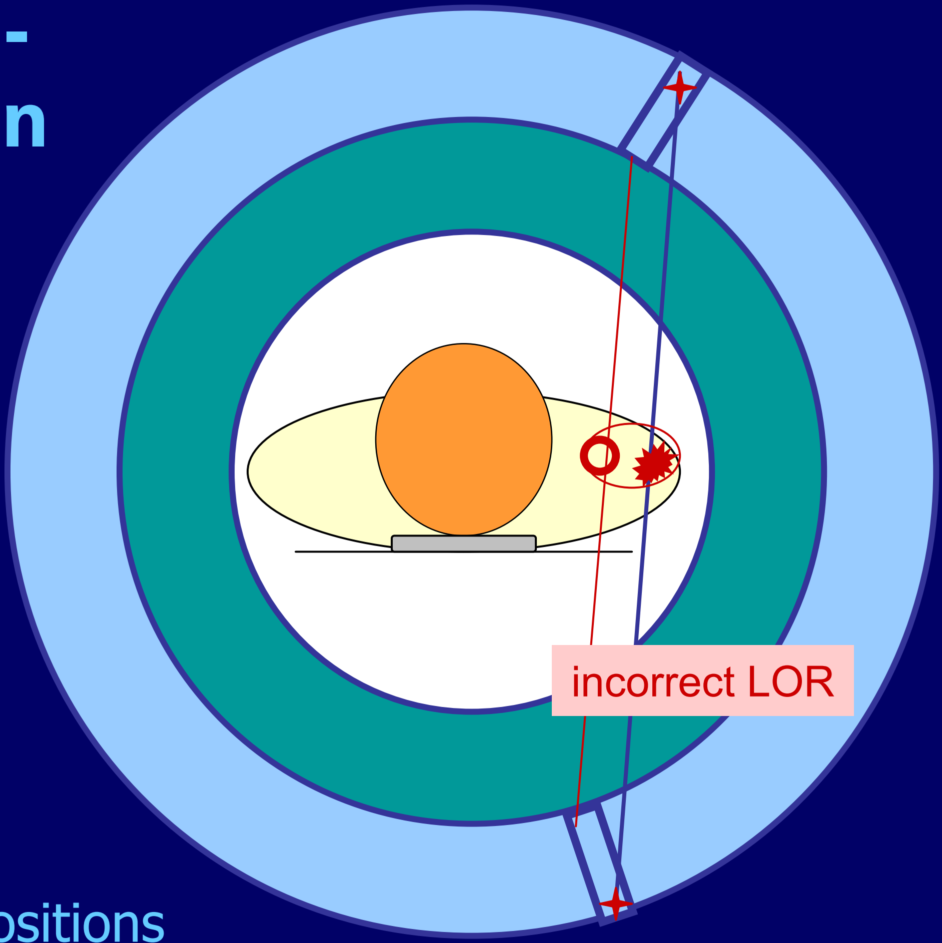
Spatial Resolution - Depth-of-interaction

DOI

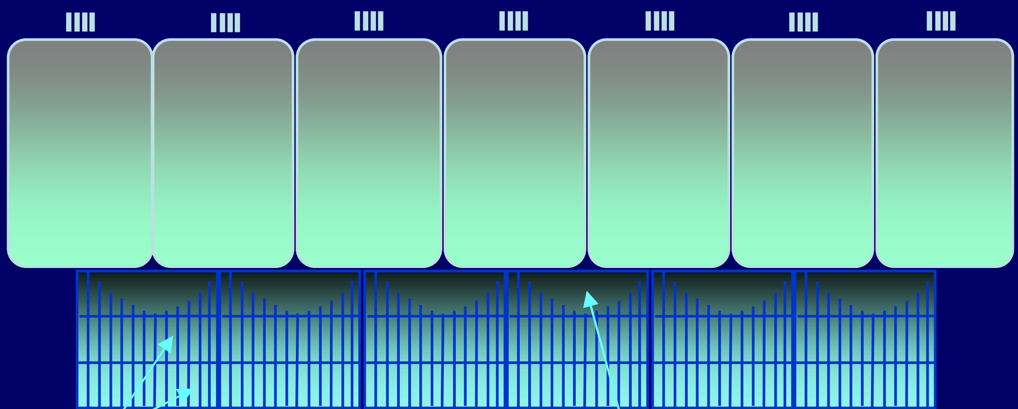


Depth-of-interaction (DOI) information is needed to maintain good resolution at off-centre positions

Inorganic Scintillators

PET HRRT

High resolution research tomograph



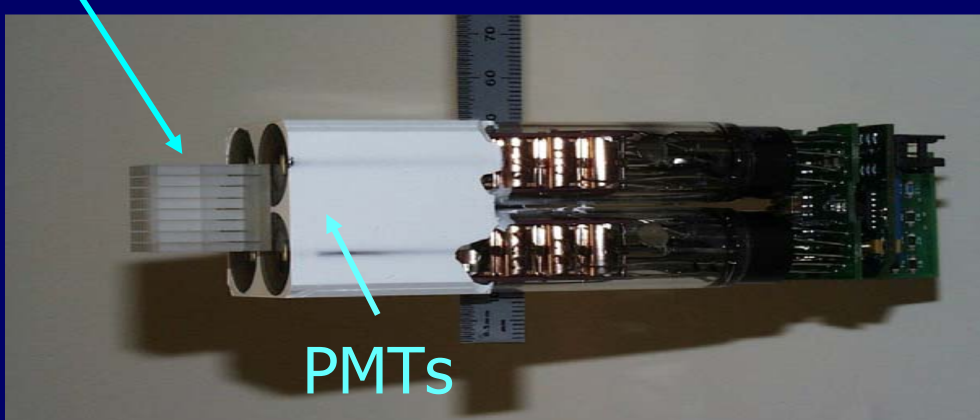
PMTs

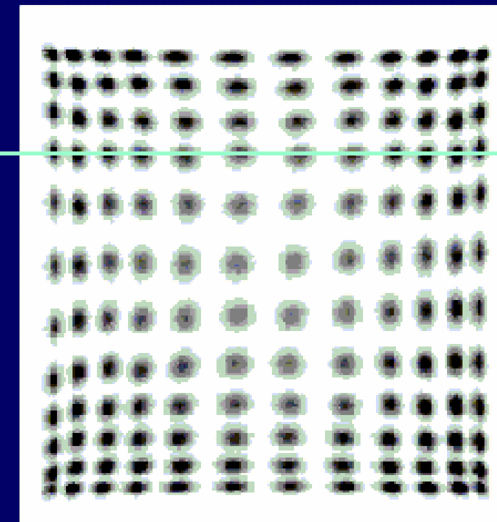
DOI

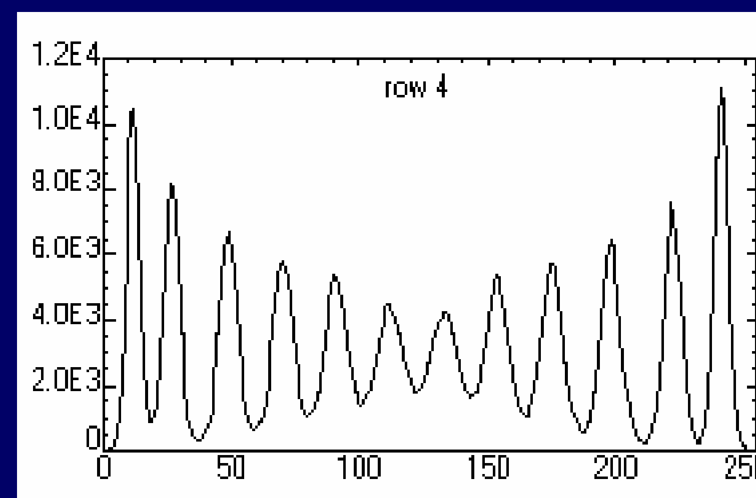
LSO scintillators

7.5 x 2.1 x 2.1 mm^3

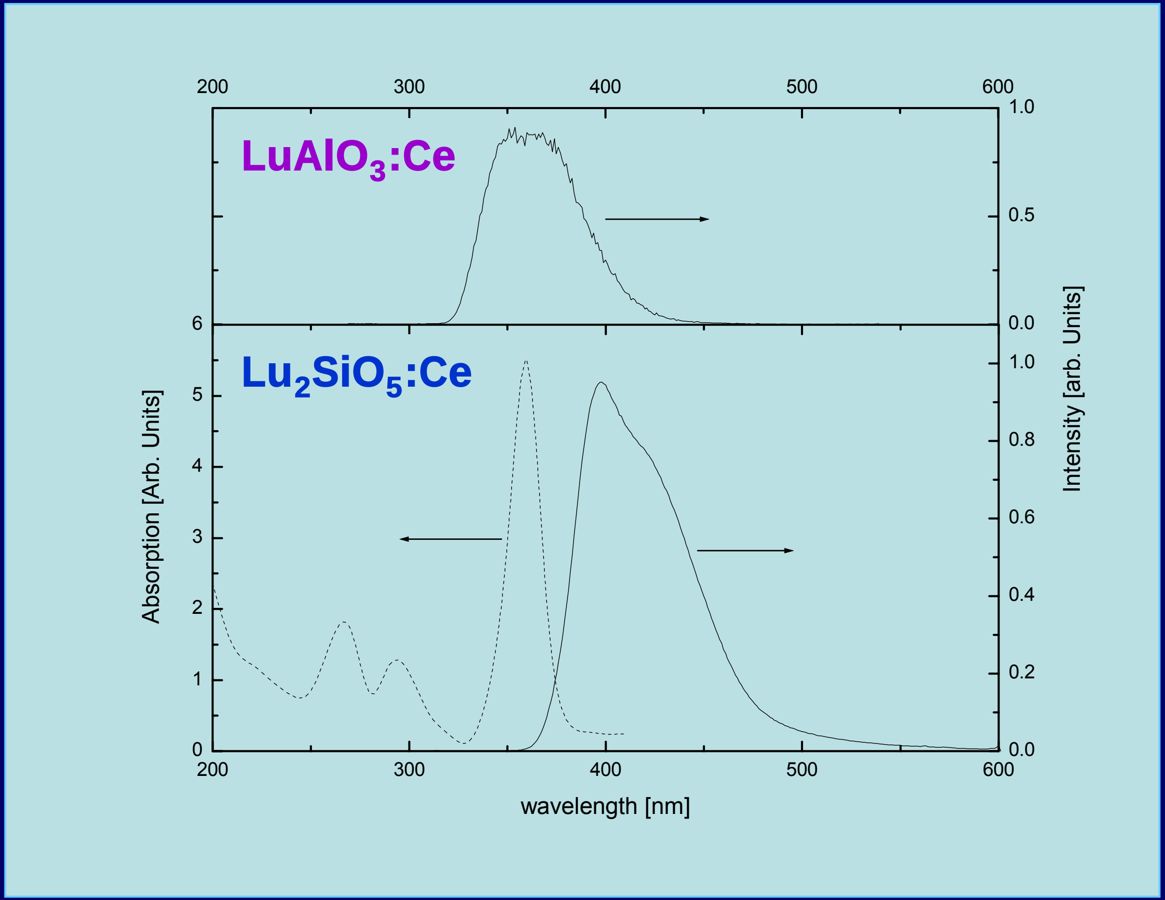
Light guide

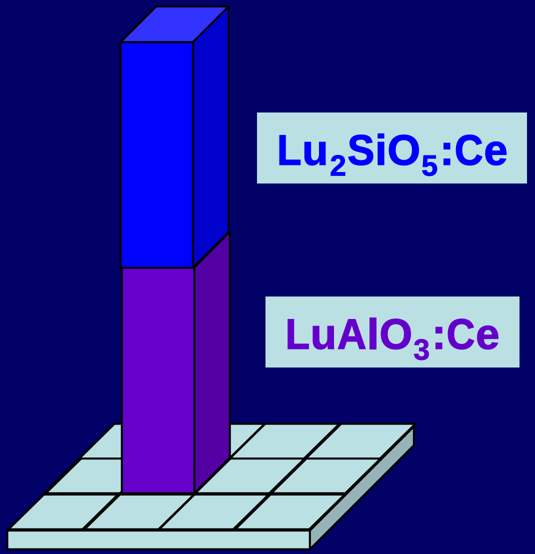






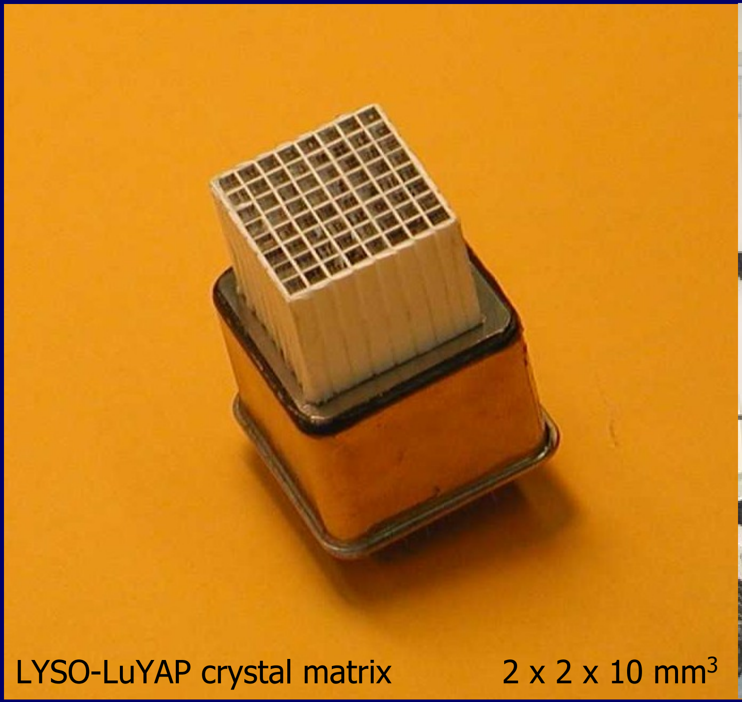
PET - depth of interaction - DOI

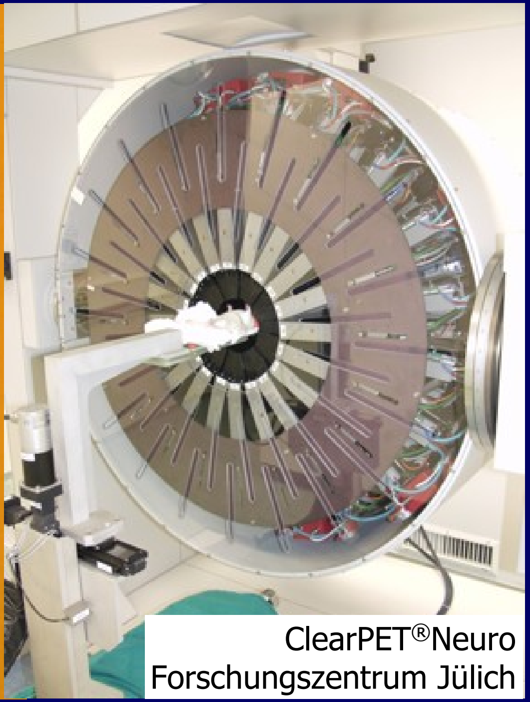




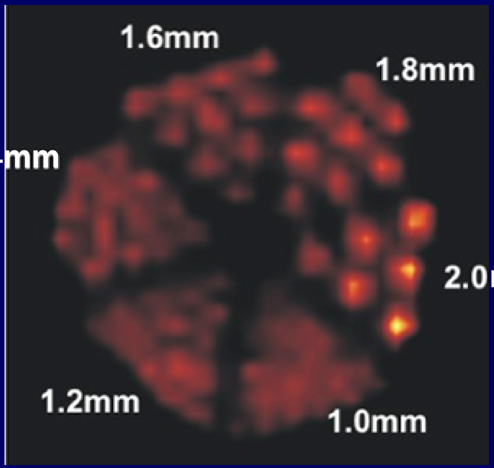
APD array
or
multi-anode PMT

ClearPET®





Derenzo phantom



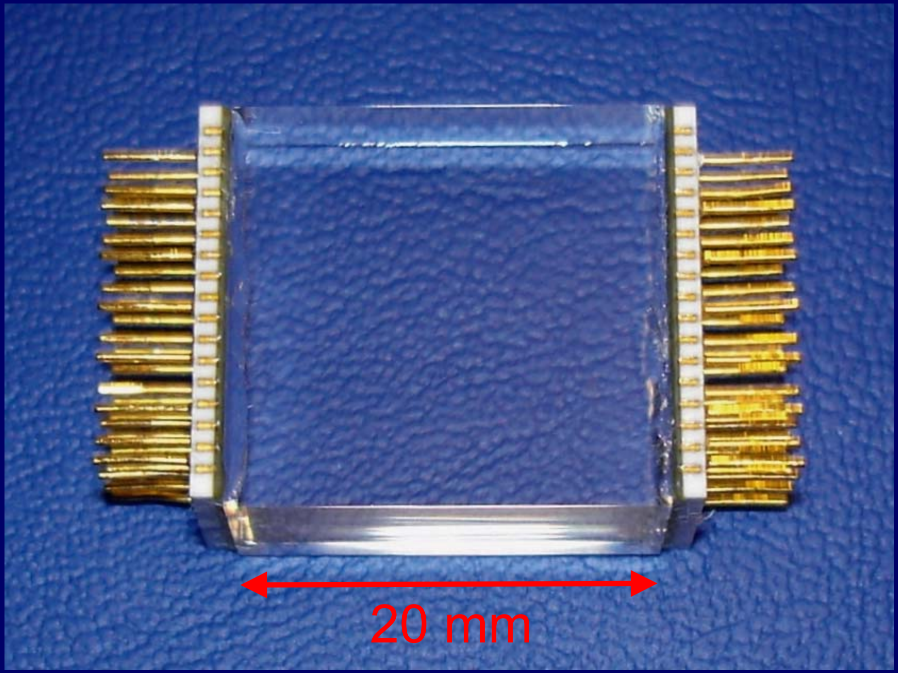
The phantom was filled with 0.5 mCi ^{18}F and scanned for 6 minutes.

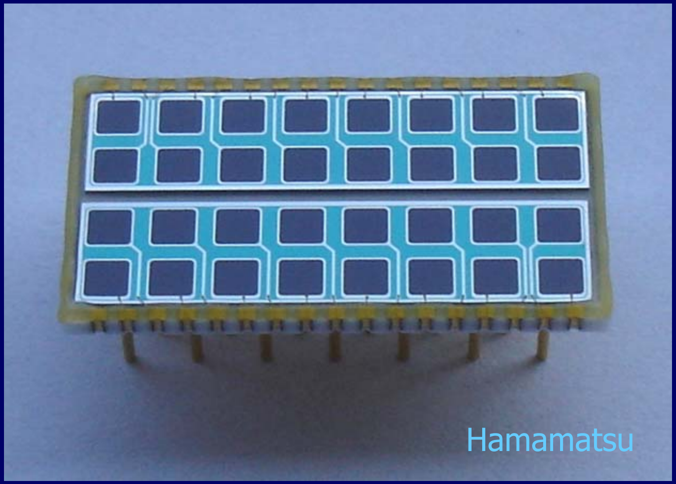
CRYSTAL CLEAR Collaboration

CERN (Geneva), Forschungszentrum Jülich, Institute of Nuclear Problems (Minsk), Institute of Physik (Ashtarak, Armenia), LIP (Lisbon), Sungkyunkwan University School of Medicine (Seoul), Université Claude Bernard (Lyon), Université de Lausanne and the Vrije Universiteit Brussel

Raytest GmbH

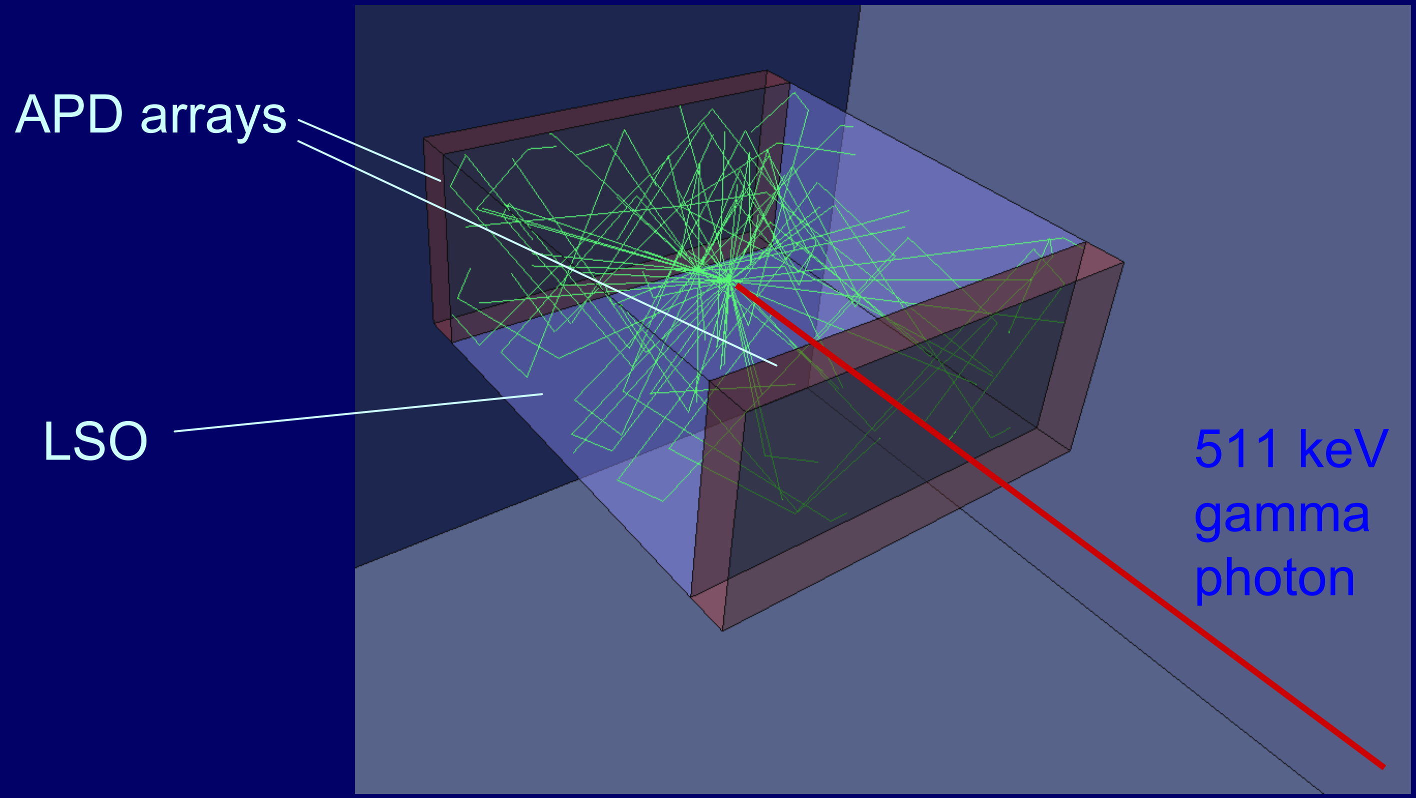
Monolithic scintillation detectors





- Monolithic crystal block LSO
- One or two APD arrays
- 3D interaction position derived from light distribution on APDs

Monolithic scintillation detectors



GEANT4 Monte Carlo simulation of an LSO block read out by two APD arrays. A small fraction of the optical photons produced by the absorption of a 511 keV annihilation photon is shown.

Spatial Resolution

Experimental data

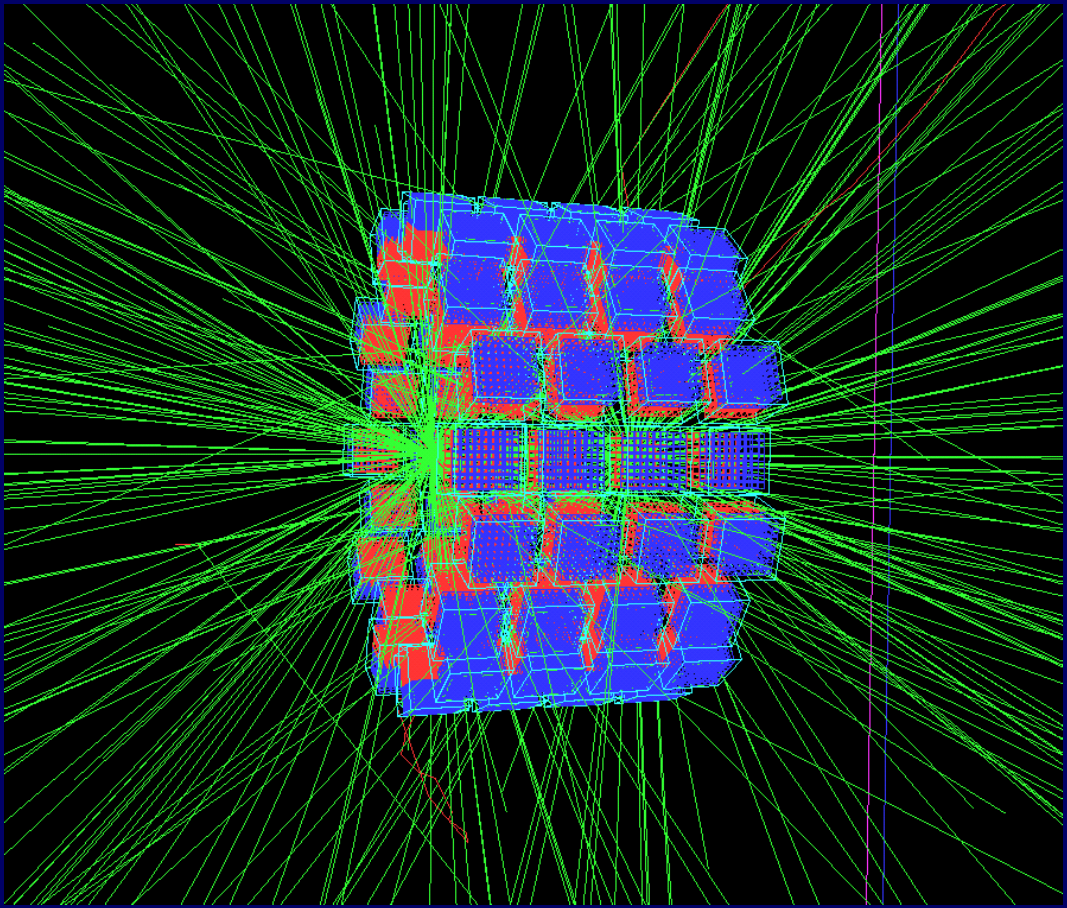
- *20x10x10 mm³ LYSO, polished, back side readout.*
- *Same number of events per position in both the training and the test set, for all LLD settings (1000 in each set).*

- LLD on test data causes some improvement, especially in FWTM
- LLD on training data causes only limited improvement

Training set	Test set	Spatial Resolution (FWHM mm)	Spatial Resolution (FWTM mm)
All E	All E	2.19	5.27
>250 keV	All E	2.20	5.27
>350 keV	All E	2.20	5.37
>415 keV	All E	2.15	5.25

Training set	Test set	Spatial Resolution (FWHM mm)	Spatial Resolution (FWTM mm)
all E	All E	2.19	5.27
all E	>250 keV	2.14	4.87
all E	>350 keV	2.13	4.81
all E	>415 keV	2.13	4.67

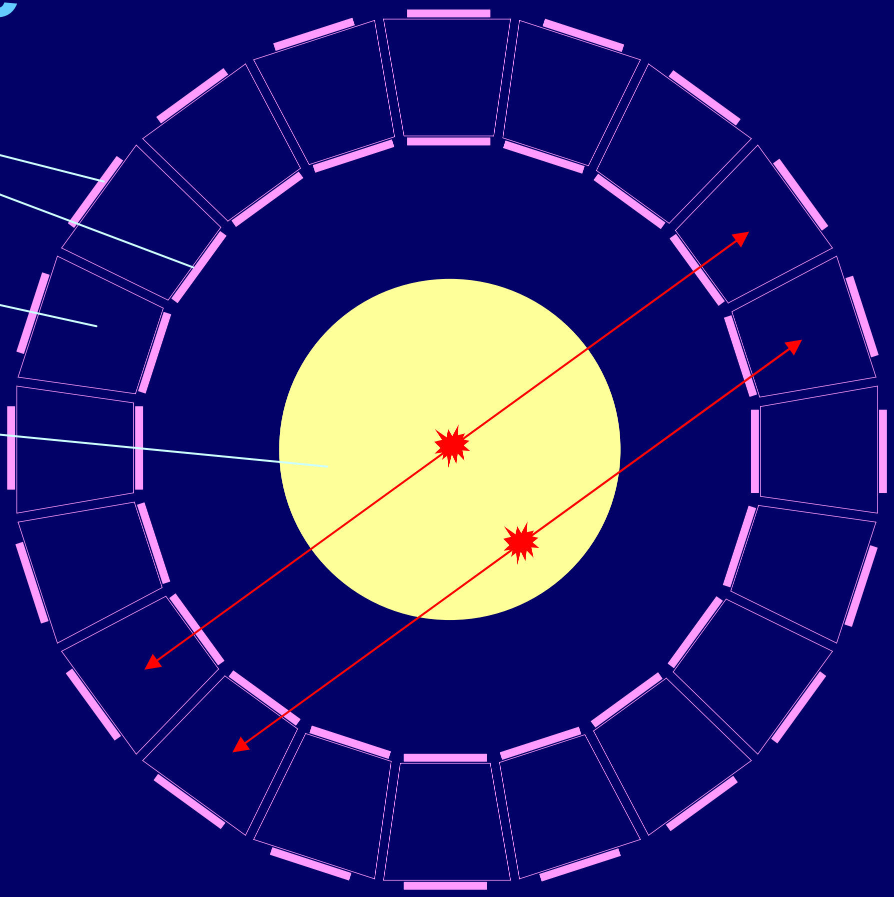
Training set	Test set	Spatial Resolution (FWHM mm)	Spatial Resolution (FWTM mm)
All E	All E	2.19	5.27
>250 keV	>250 keV	2.12	4.82
>350 keV	>350 keV	2.08	4.81
>415 keV	>415 keV	2.08	4.81



Small animal PET

GEANT4 simulation of a scanner with dead space between the scintillator pixels and between the detector modules. Note the "leakage" of radiation, reducing the overall detection efficiency.

Angle of incidence
DOI



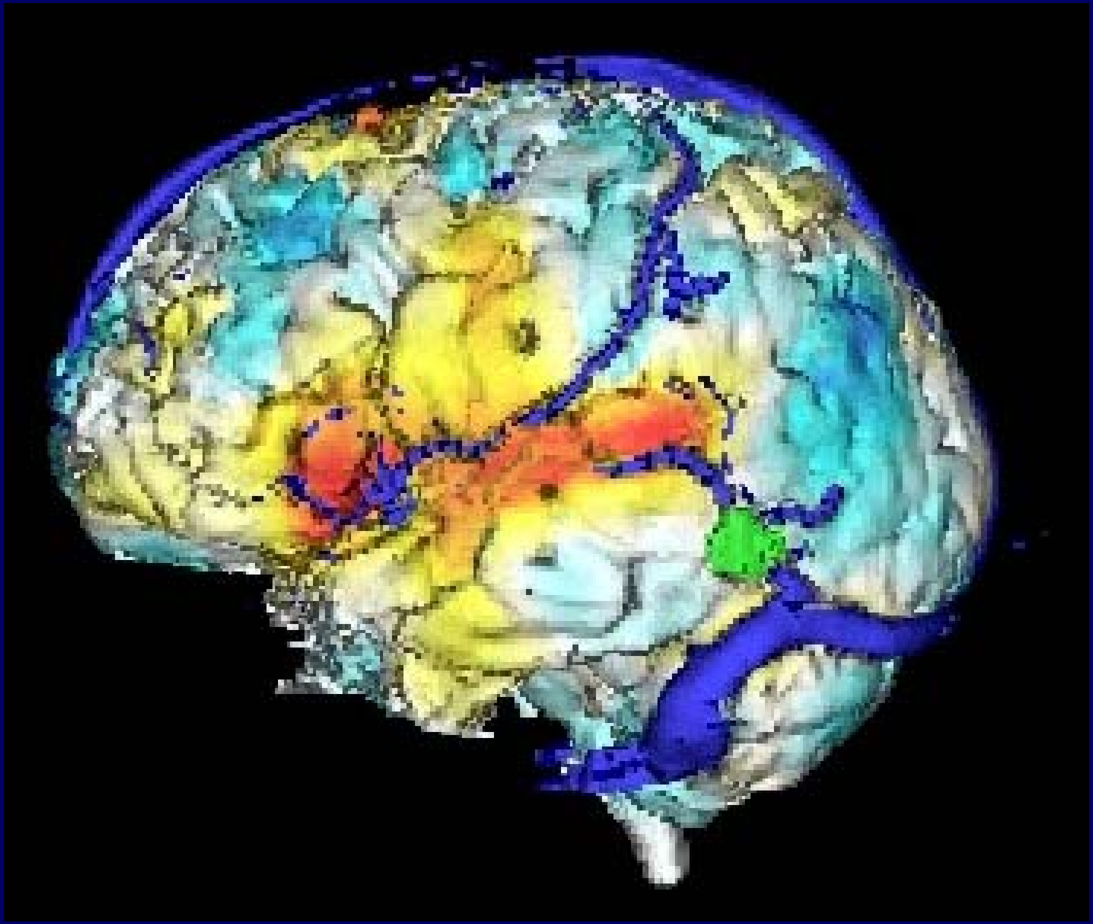
Solid angle coveragage

Efficiency gain > 2

SCINT+ APD integration of

PET + MRI ?

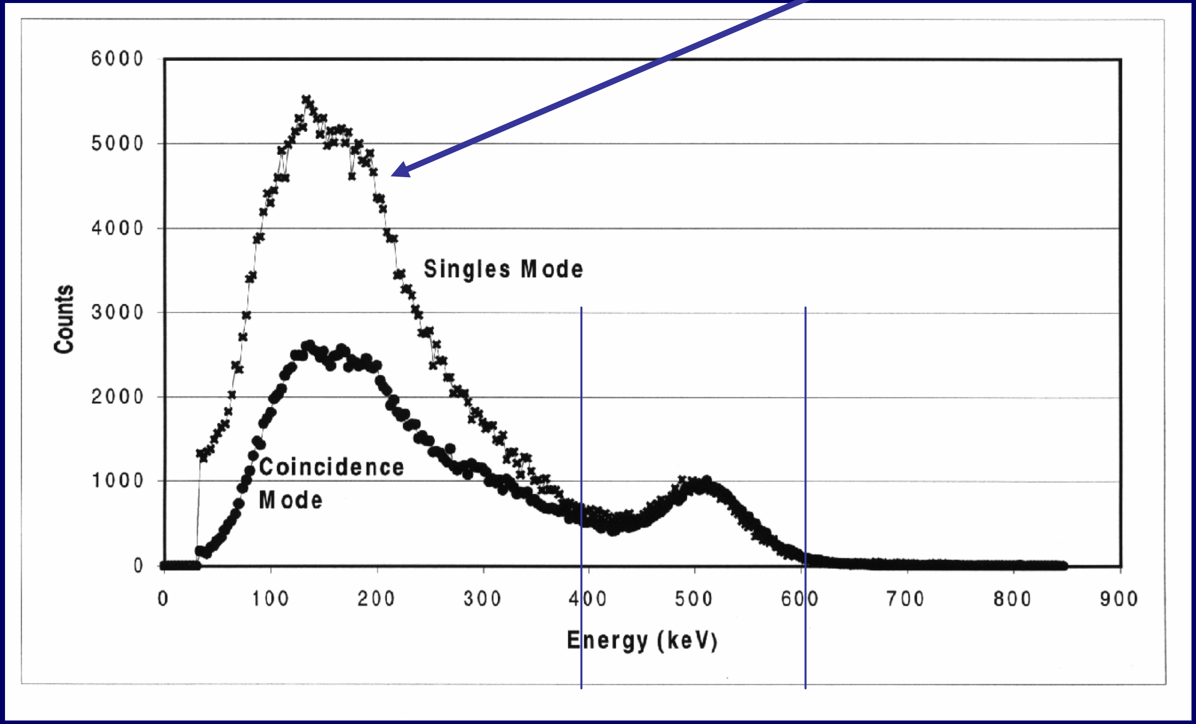
Blood flow changes under speech activation (red)
Tumor (green)



courtesy Klaus Wienhard, MPI für Neurologische Forschung, Köln

$LaBr_3$:Ce and PET

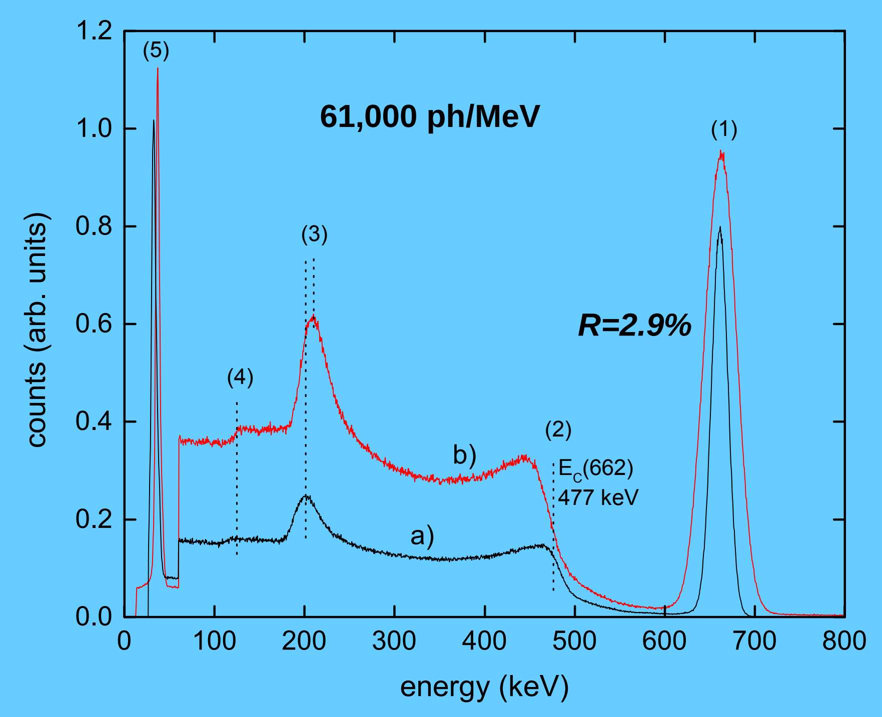
Random coincidences ~ $N^2_{singles}T$



Energy Resolution
&
Time Resolution

TOF

Energy resolution



$LaBr_3$: 0.5% Ce^{3+}

- ✓ Dimension, ∅ 3mm x 10mm
- ✓ Observed resolution, R = 2.9%
- ✓ Scintillator resolution, R_s = 1.5%

Time resolution 300 ps - TOF

4.5 cm

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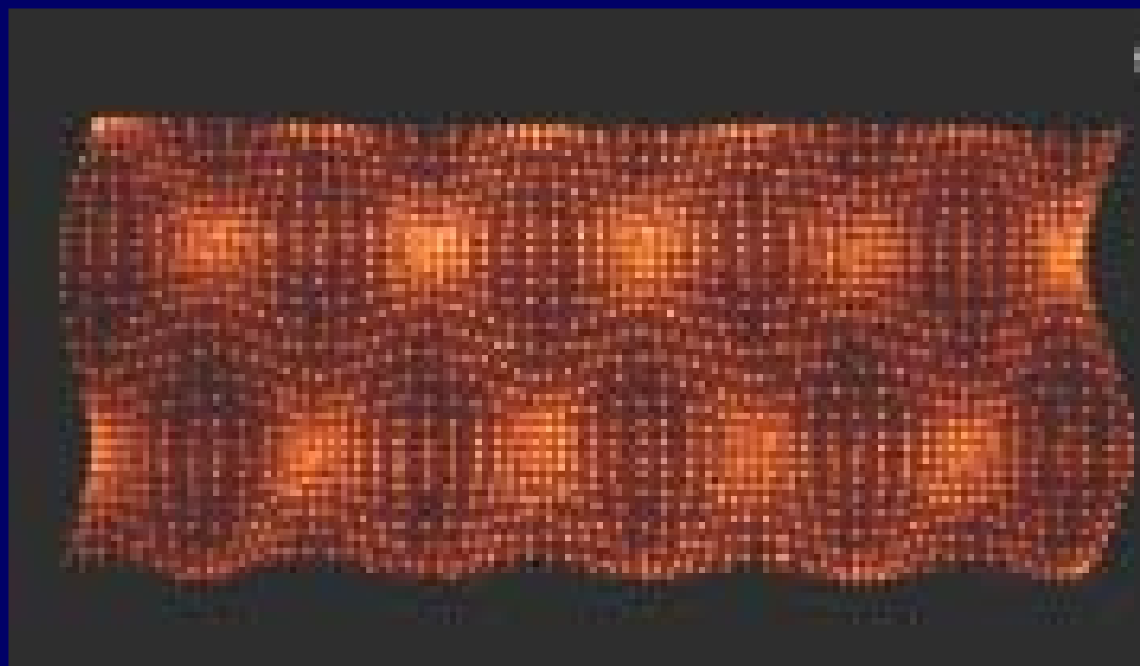
Inorganic Scintillators

Full Module
1620 (60x27) 4mm x 4mm x 30mm $LaBr_3$:Ce crystals
Raw Signals



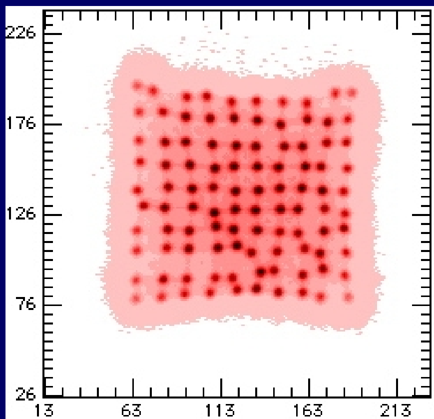
courtesy Philips Research Laboratories

Inorganic Scintillators

0.5% Ce-doped 100 Crystal Array

2D Position Flood Map @ 511 keV

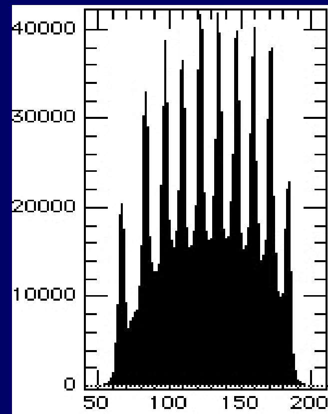
Y-Position (a.u.)



X-Position (a.u.)

Histogram for center row of crystals

Counts



X-Position (a.u.)

Performance Measurements

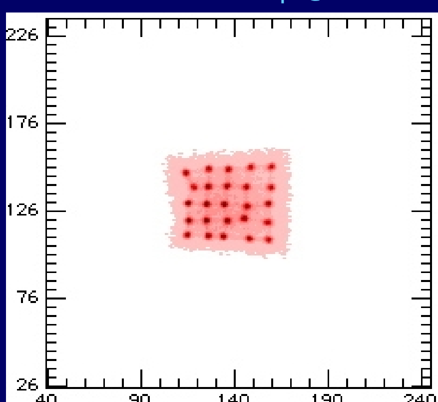
- Average Energy Resolution
  DE/Eavg 5.31% ± 0.44%
- Standard Deviation in Light Output 3%
- Average Peak-to-Valley Ratio
  P/Vavg 2.9

Array coupled to 2.1 cm light guide and 7 XP20Y0 PMT's (each 50 dia.)

5.0% Ce-doped 25 Crystal Array

2D Position Flood Map @ 511 keV

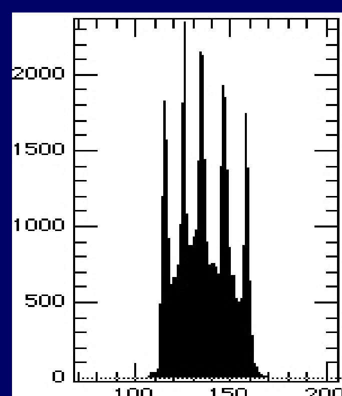
Y-Position (a.u.)



X-Position (a.u.)

Histogram for center row of crystals

Counts



X-Position (a.u.)

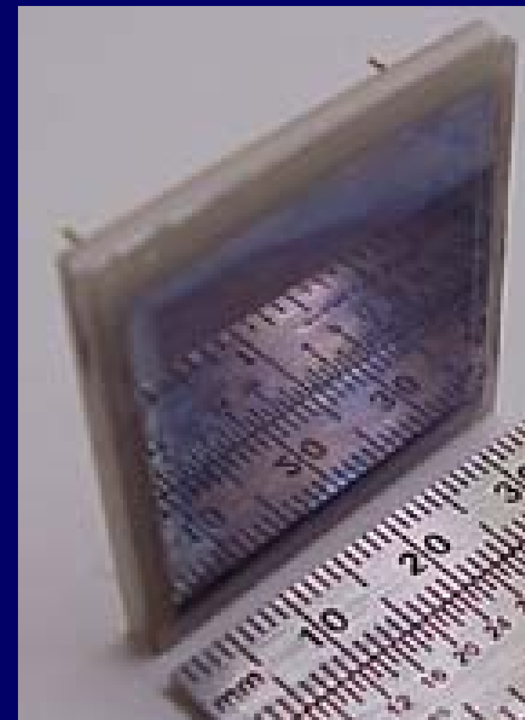
- Average Energy Resolution
  DE/E_{avg} 5.65% ± 0.59%
- Standard Deviation in Light Output 5%
- Average Peak-to-Valley Ratio
  P/V_{avg} 3.4

Array coupled to 2.1 cm light guide and 7 XP20Y0 PMT's (each 50 dia.)

courtesy Philips Research Laboratories

PSAPD-$LaBr_3$:Ce Gamma Ray Imaging Module

8x8 element $LaBr_3$ Array (2x2x5 mm pixels)



28 x 28 mm^2 PSAPD

Courtesy Kanai Shah, RMD

Moon

Mercury

Inorganic Scintillators

Space Research - Planetology

1. <u>Surface composition</u> provides information on the planet bulk composition. Bulk composition helps to understand where and how the planet forms.

   → **Solar System / Planets Origin**

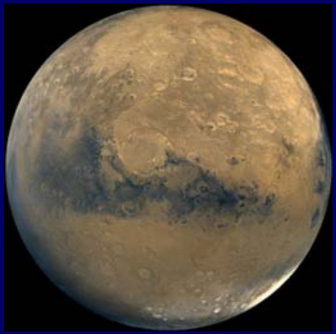
- <u>Surface composition</u> provides information about how a planet has evolved since its formation.

   → **Solar System / Planets Evolution**

- <u>Comparative studies</u> helps us to understand how planets differ from each other.

   → **Comparative Planetology**

Mars

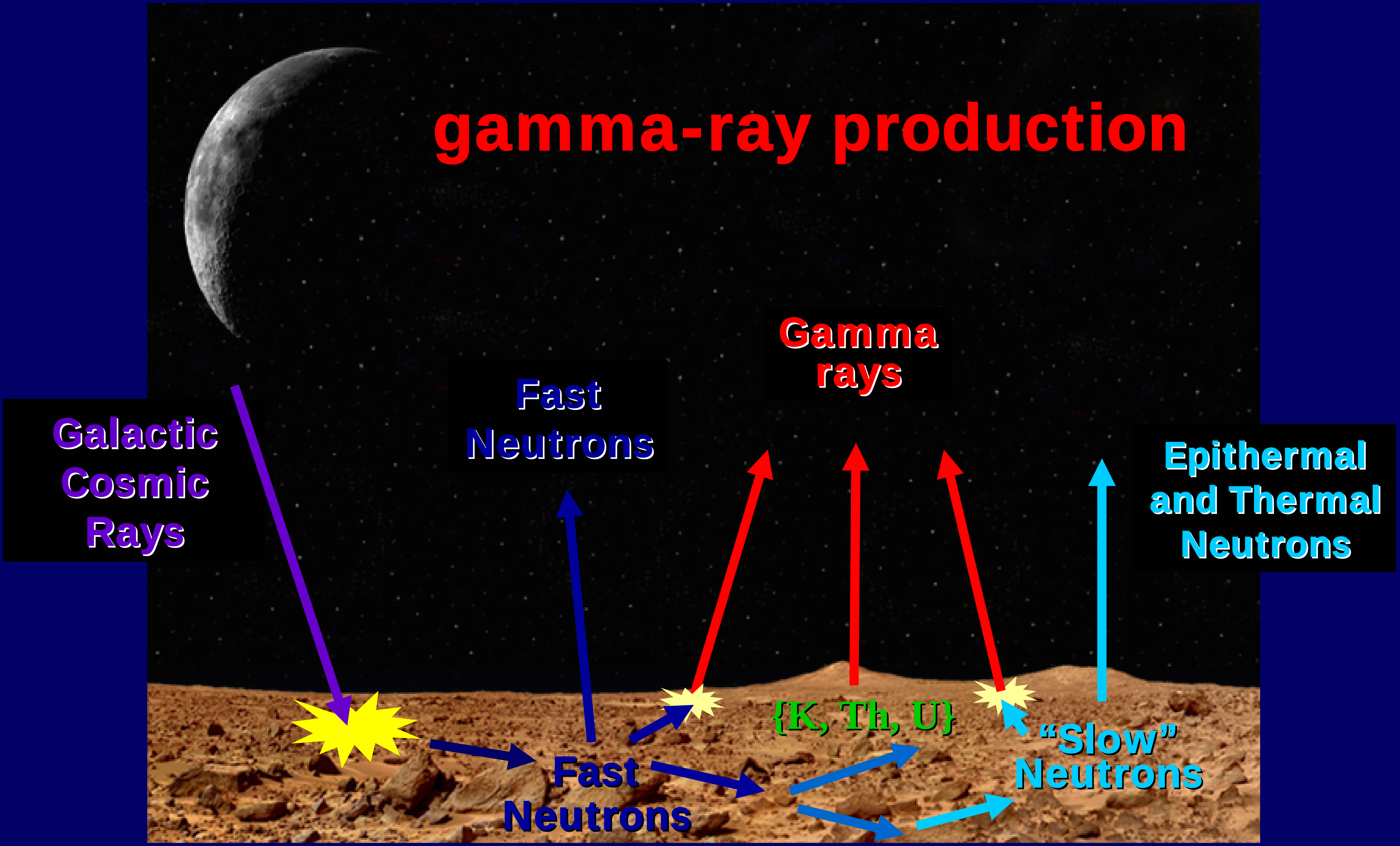


Asteroid

Remote sensing & ground truth

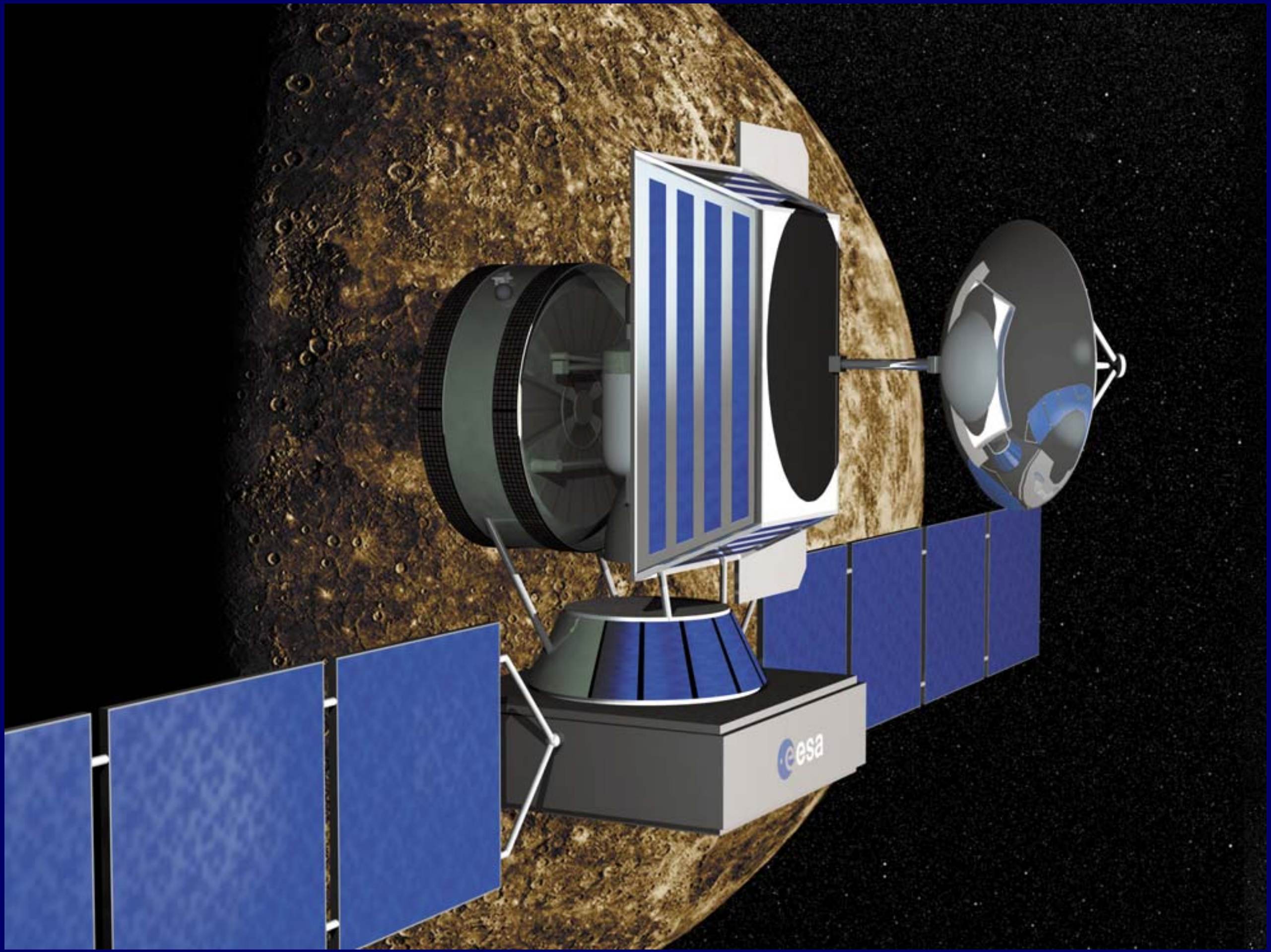
Courtesy Alan Owens, ESTEC

TU Delft



Courtesy Alan Owens, ESTEC

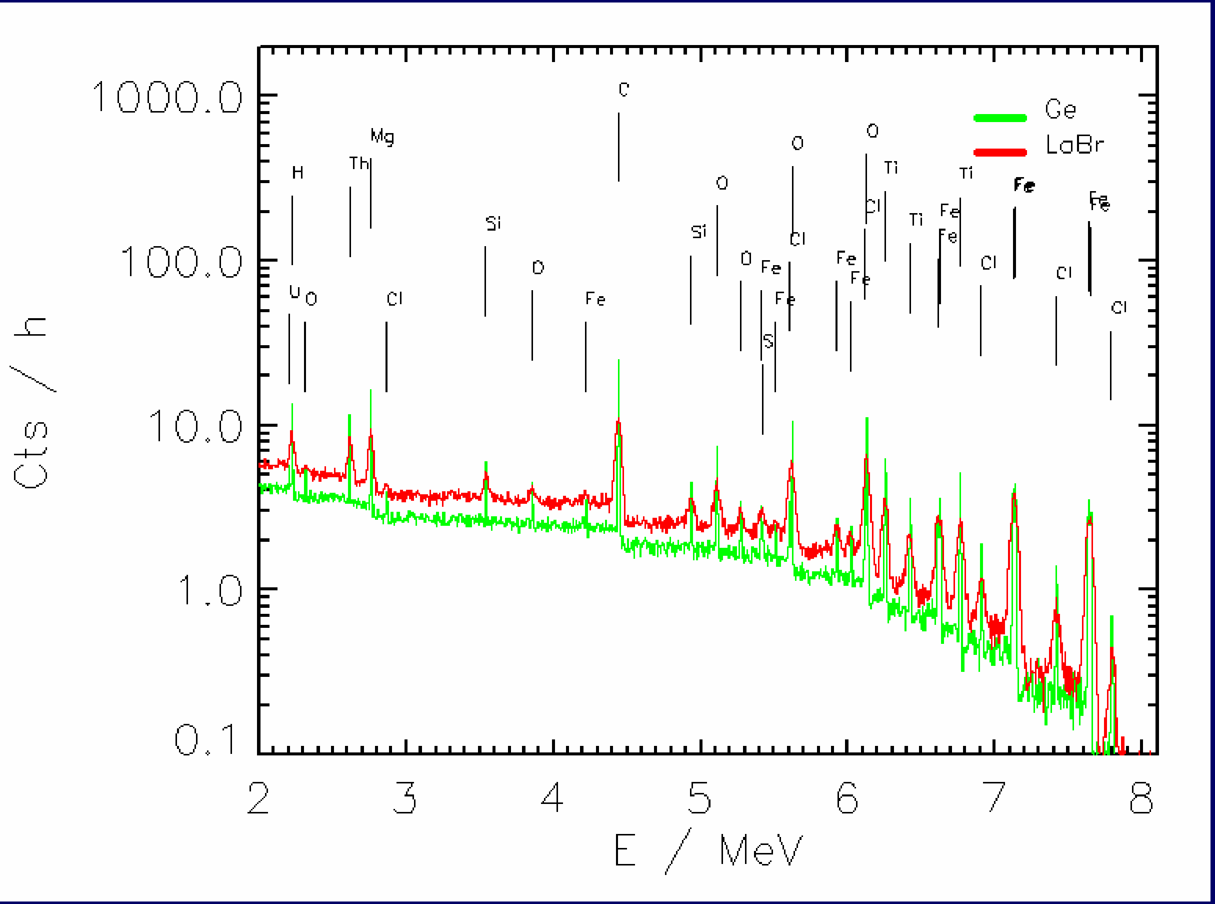
BepiColombo - An interdisciplinary mission to the planet Mercury



Courtesy Alan Owens, ESTEC

Inorganic Scintillators

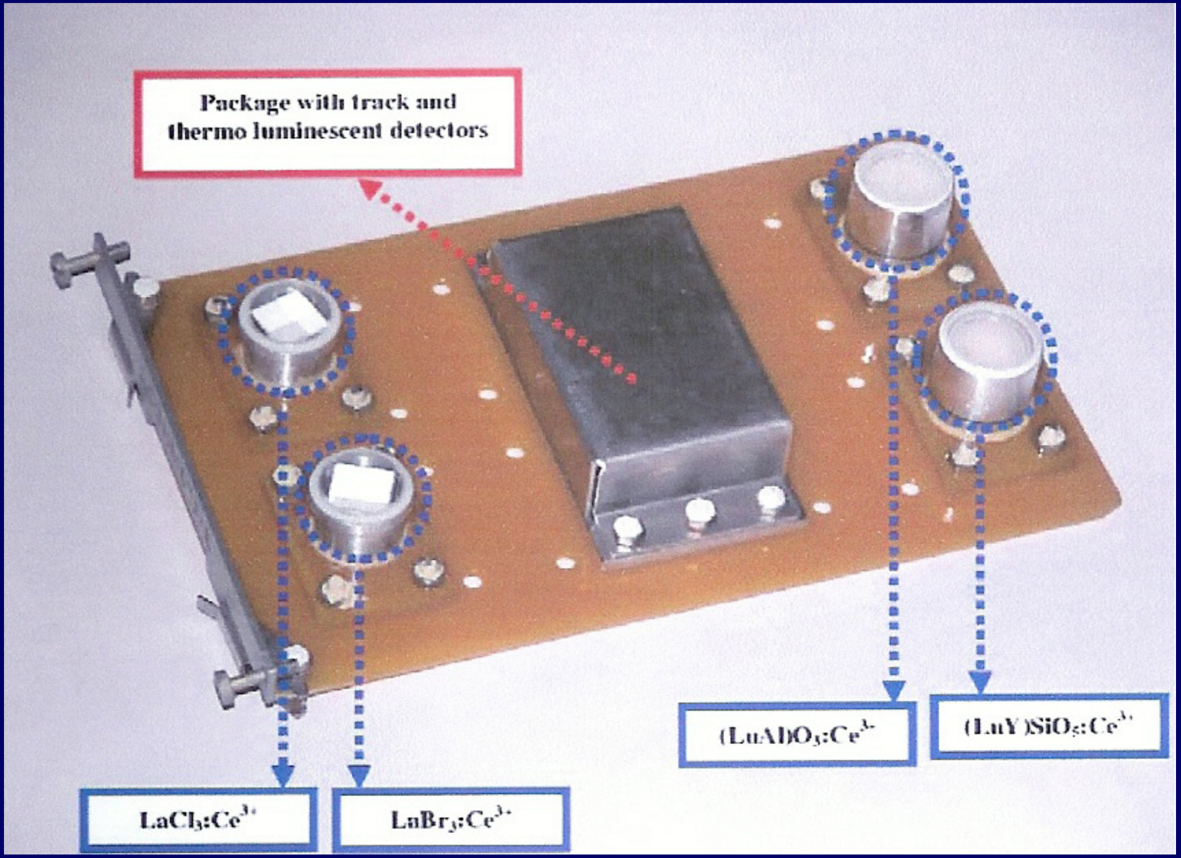
Simulated in orbit spectra measured by a 6.5 cm × 6.5 cm diameter Ge crystal and an 8 cm diameter LaBr detector.



ESA, Saint-Gobain, TUD and Cosine

Inorganic Scintillators

Board with scintillators will orbit the earth in ISS for 1 – 2 years
Test on radiation damage



ESA, Saint-Gobain, TUD and Cosine

Scintillators
for
Thermal Neutron Detection

Thermal neutron detection

reaction $^{6}Li + n \rightarrow {}^{3}T + {}^{4}He$

kinetic energy 4.8 MeV

in scintillator

charged particle response < electron (gamma) response

"α/β ratio" < 1

Inorganic thermal – neutron scintillators

^{6}Li based thermal-neutron scintillators

Host	Dopant (conc mol%)	Light yield photons per neutron	Light yield photons per MeV gamma	α/β ratio	τ ns	Abs. Length at 1.8Å mm	
^{6}Li-glass	Ce	~6,000	~4,000	0.3	75	0.52	
^{6}LiI	Eu	50,000	12,000	0.87	1,400	0.54	hygr
^{6}LiF/ZnS	Ag	160,000	75,000	0.44	> 1,000	0.8	opaque
$^6Li_6Gd(BO_3)_3$	Ce	40,000	25,000	0.59	200/800	0.35	

Inorganic thermal-neutron scintillators
C.W.E. van Eijk, A. Bessière, P. Dorenbos
Nucl. Instr. Meth. A **529**(2004)260-267

Inorganic thermal – neutron scintillators

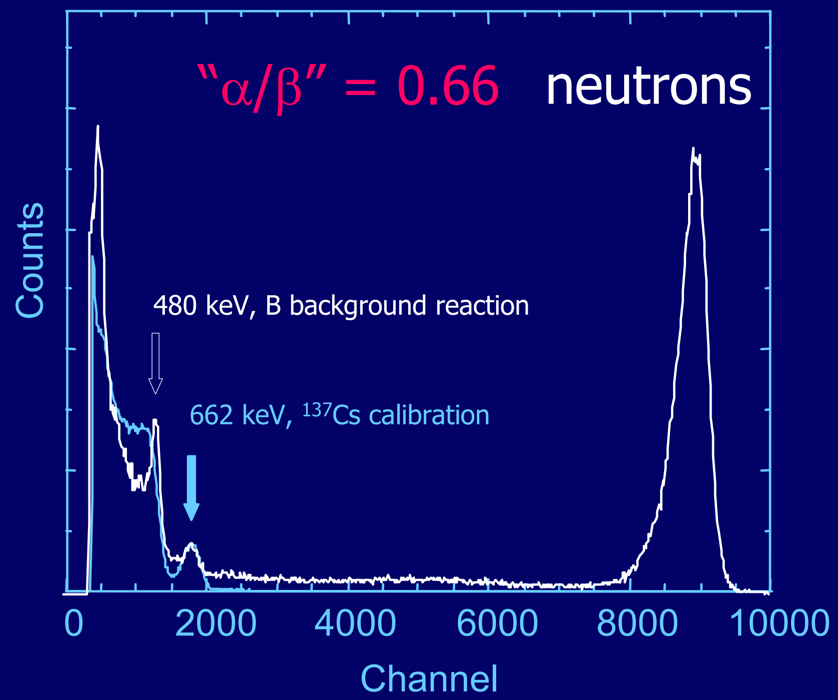
Cs_2LiYCl_6: 0.1 % Ce

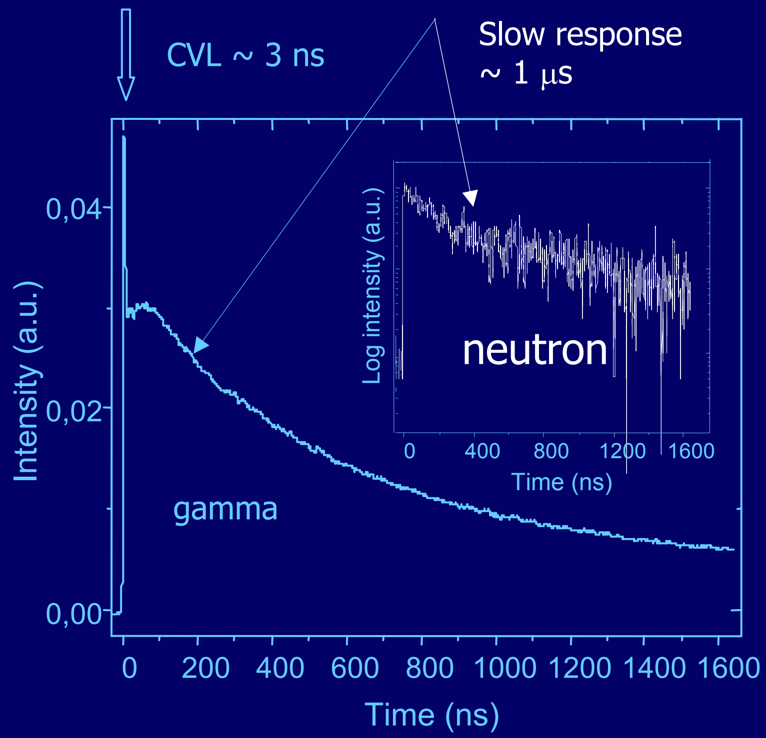
All samples studied: natural ^6Li

pulse shape &

pulse height discrimination

no CVL





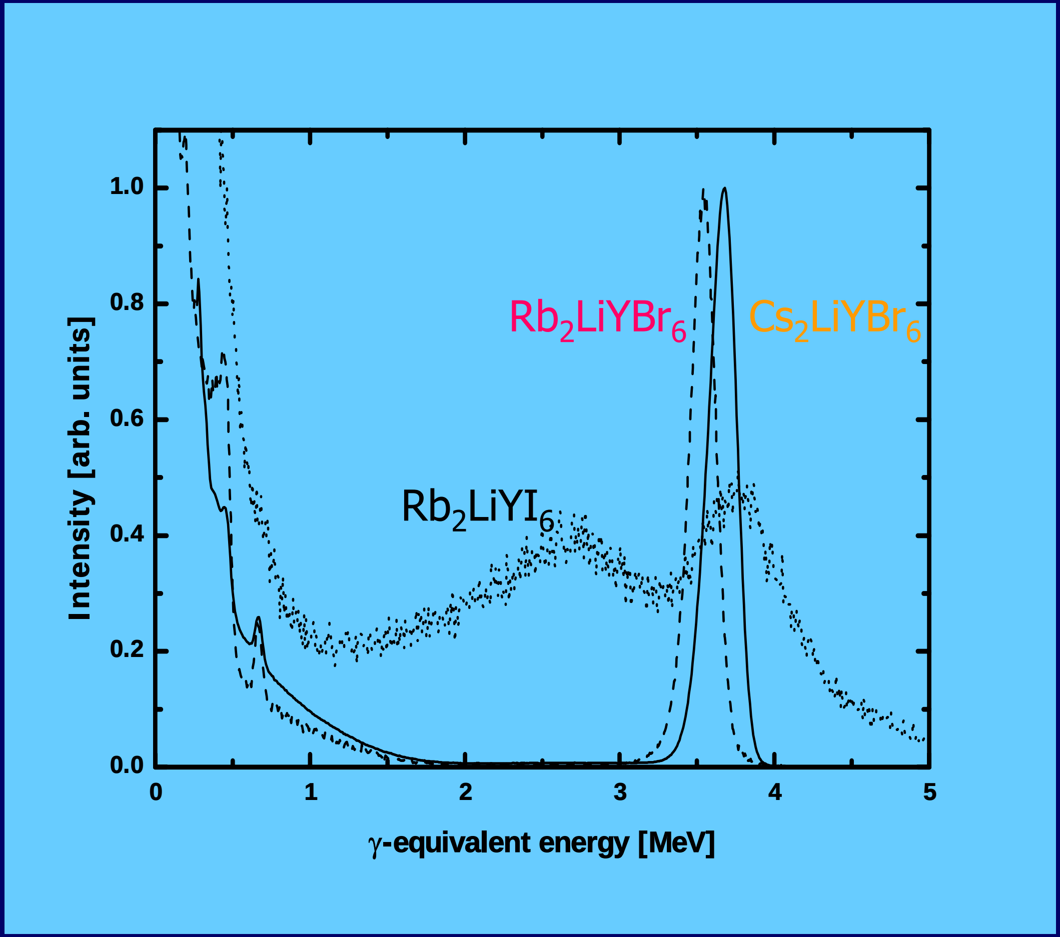
Luminescence and scintillation properties of $Cs_2LiYCl_6 : Ce^{3+}$ for γ and neutron detection
A. Bessière, P. Dorenbos, C.W.E. van Eijk, K.W. Krämer, H.U. Güdel
Nucl. Instr. Meth. A 537 (2004) 242-246

New Thermal Neutron Scintillators: $Cs_2LiYCl_6 : Ce^{3+}$ and $Cs_2LiYBr_6 : Ce^{3+}$
A. Bessière, P. Dorenbos, C.W.E. van Eijk, K.W. Krämer, H.U. Güdel
IEEE Trans Nucl Sci **51-5** (2004) *October*

Inorganic thermal-neutron scintillators

host	Ce conc. (mol%)	grain size (mm^3)	ρ (g/cm^3)	$\rho Z_{eff}^4 \times 10^{-6}$	neut. abs. length at 1.8 Å 95% ^6Li enriched (mm)	abs. in ^6Li (%)
Cs_2LiYCl_6	0.1		3.3		2.5	78
$Cs_2LiLaCl_6$	1					77
$Cs_2LiLuCl_6$						73
Cs_2LiYBr_6	0.3					
	1		4.14		3.4	90
	3					
Cs_2LiYI_6	0.5		4.36			90
Cs_2LiLuI_6	0.5		4.76			84
Rb_2LiYBr_6	0.5	~2.5 x 2.5 x 2.5	3.8		3.5	95
Rb_2LiYI_6	0.5		4.0			96

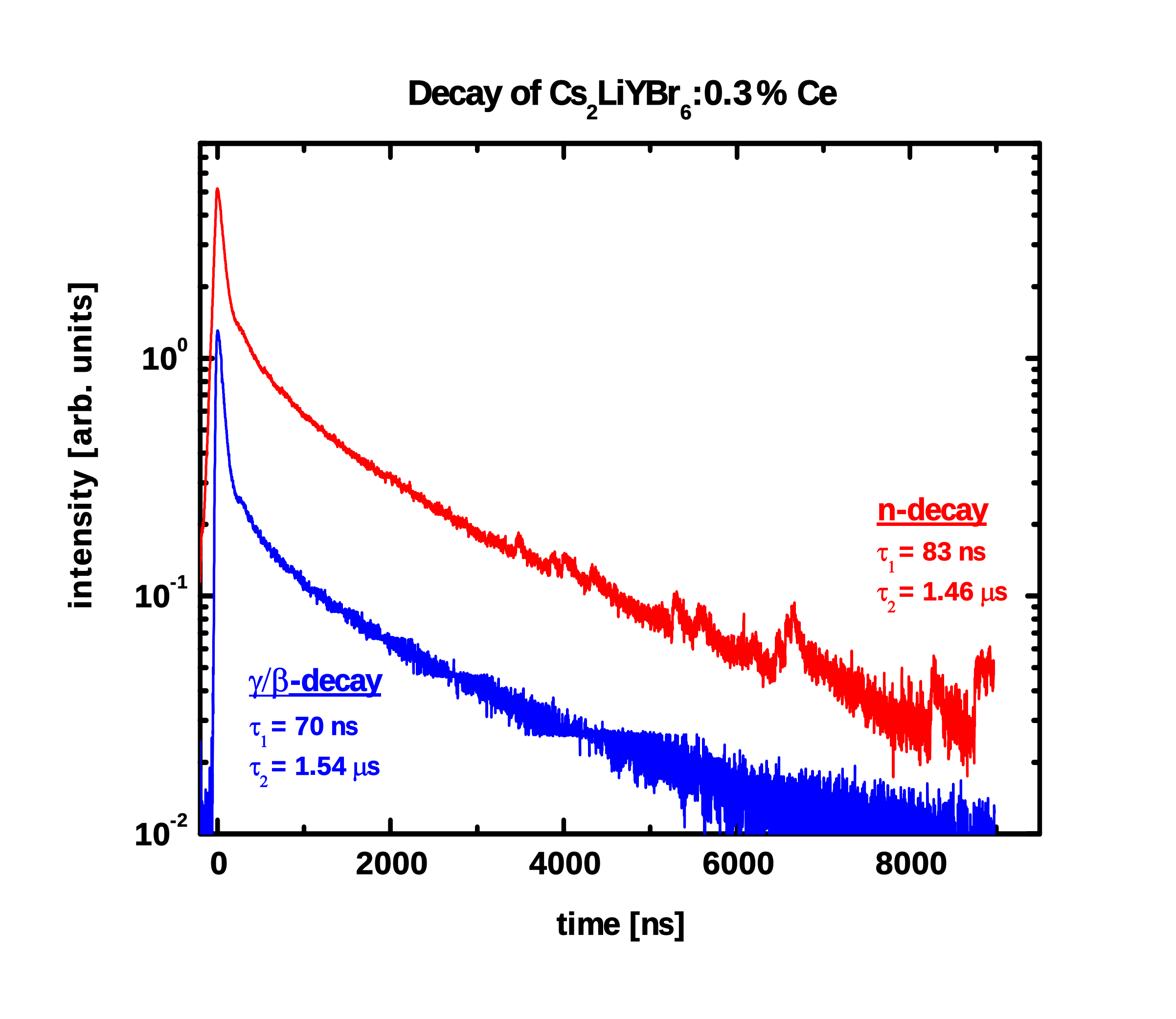
Inorganic thermal-neutron scintillators



Different light yields!

TUDelft

Inorganic thermal-neutron scintillators



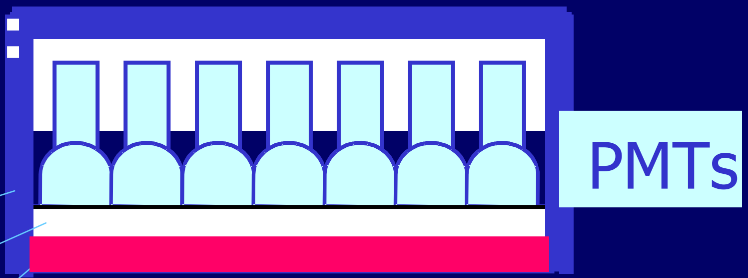
TU Delft

Inorganic thermal-neutron scintillators

	Ce conc (mol%)	em.wavel (nm)	γ-ray LY (ph/MeV)	decay (ns)	FWHM (%) at 662 keV	neutron LY (ph/n)	decay (ns)	α/β
Cs_2LiYCl_6	0.1	380	18,000	$\sim 10^3$	9	56,000	$\sim 10^3$	0.66
		255-470	700	3 (CVL)		-	-	
$Cs_2LiLaCl_6$	1	375,410	28,000	115, $\sim 10^3$	20			
$Cs_2LiLuCl_6$								
Cs_2LiYBr_6	0.3	389,423	20,000	~70, 1.5×10^3	4.6	73,000	~83, 1.5×10^3	0.76
	1		18,000	89, 2.5×10^3	8	67,000		0.77
	3		14,000		15	64,000		0.9
Cs_2LiYI_6	0.5	bad sample						
Cs_2LiLuI_6	0.5	bad sample						
Rb_2LiYBr_6	0.5	390,420	18,000	130(30), 1.7×10^3	5	65,000		0.75
Rb_2LiYI_6	0.5	425,475	7,000	80, 355		26,000		~0.8

Radioisotope imaging:

Gamma camera



Lead shield

Light guide

NaI : Tl scintillator
6 - 25 mm

spatial resolution 4 mm
at 140 keV

6000 ph

$Cs_2{}^6LiYBr_6$ neutrons → 73,000 ph
7 mm

→ ≈ 1 mm