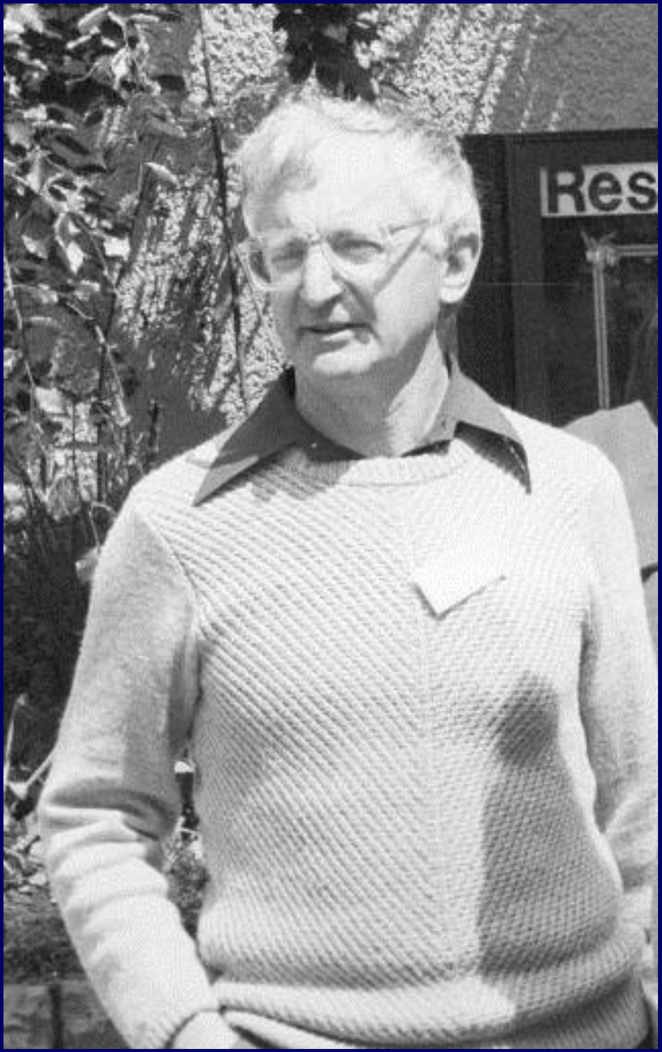
Inorganic Scintillator R&D

Elkofest 80 symposium

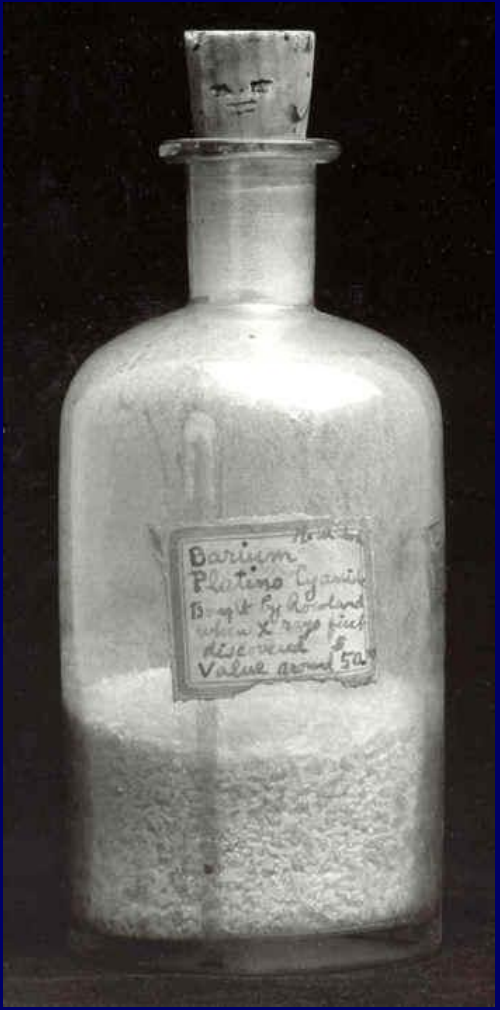
Carel W.E. van Eijk

Ljubljana
13 September 2012



Delft University of Technology

End of the 1800s



$BaPt(CN)_4$

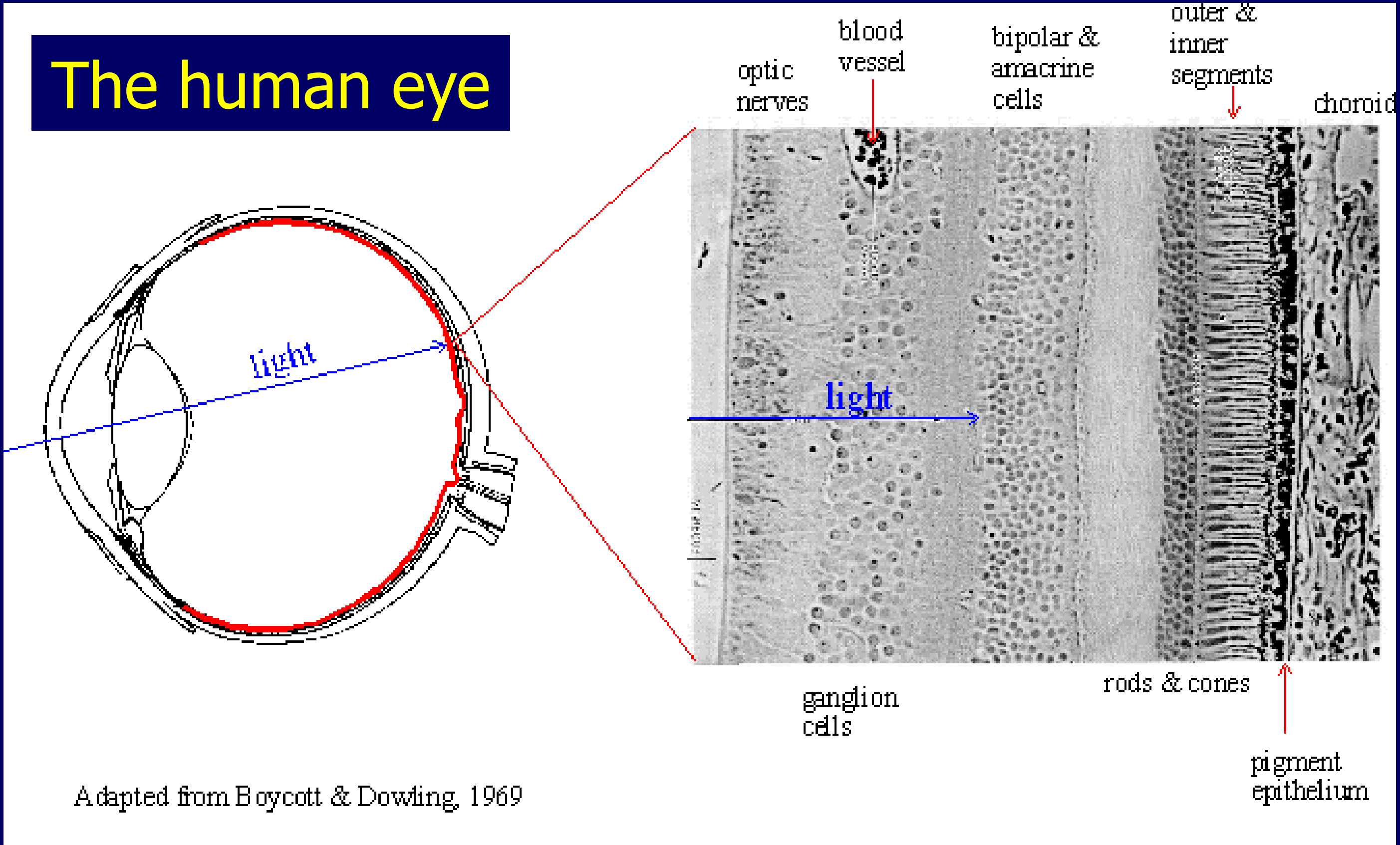
Barium platino cyanide

When Roentgen discovered x-rays (1895), his first clue that his cathode-ray tubes had produced a new type of radiation was the **faint glow of a barium platinocyanide screen** lying on a nearby table more than a meter away.

Roentgen knew that cathode rays traveled only a few cm in air; the black cardboard covering the tube eliminated ultraviolet and visible light; he knew that he had discovered something new.

The first inorganic-scintillator radiation detector

The most sensitive scintillation-light detector



Adapted from Boycott & Dowling, 1969

and there is …… the photographic plate

Barium platino cyanide grains spread out on a photographic plate

Barium ore is contaminated with measurable quantities of radium chemically very similar

Roentgen could have discovered radioactivity as well in 1895!

1. Inorganic scintillator as radiation detector mainly of gamma rays
2. Light emission
3. Light sensor

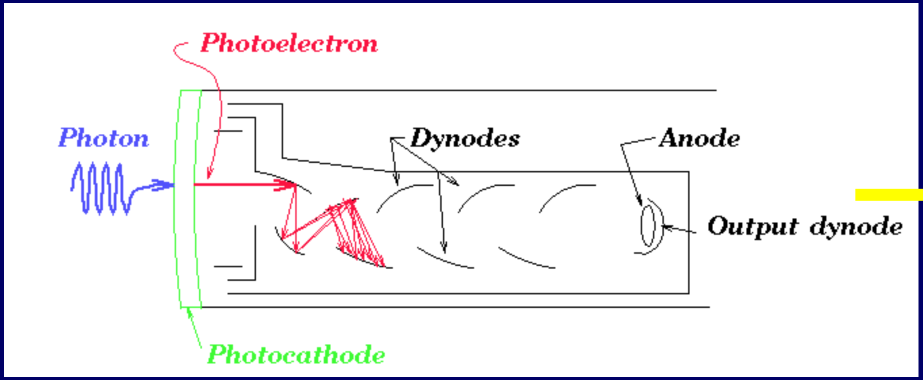
In the 1900s extensive R&D on these topics

Many applications

Light sensor

Invention and development of photomultiplier tube – PMT from 1934 on

Leonid A. Kubetsky & RCA



→ Measurable signal

~ 15 cm

A boost for scintillator R&D

Inorganic scintillator with high light yield

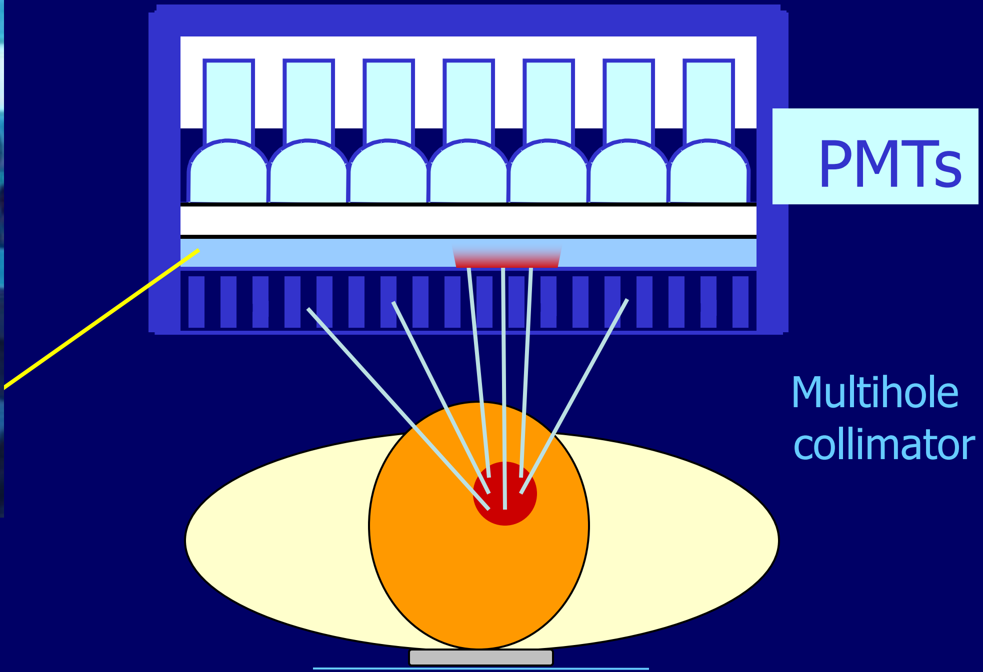
Discovery of **NaI:Tl**

Robert Hofstadter 1948

Boost for many fields e.g. medical imaging

NaI:Tl mono crystal 50 x 60 cm^2

Gamma camera



Radioisotope imaging: Planar scintigraphy

Phosphonate tagged with ^{99m}Tc,
injected into the blood stream,
is mainly transported to bones,
producing a view of the
skeletal system
The method is e.g. used to determine
whether or not cancer has metastized

From: National Geographic 171/1(1987)2-41

Why research on/ Search for New scintillators?

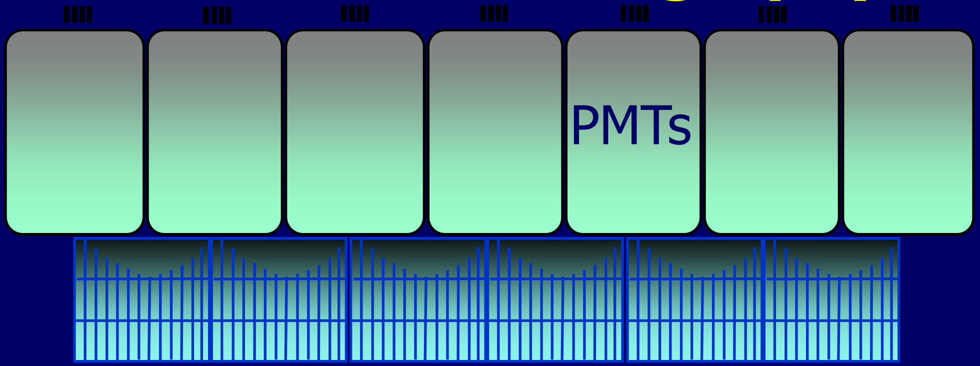
Specs of existing scintillators
do not meet the requirements
as for
Detection efficiency, Energy resolution
Time resolution, Count rate,
Light yield

We want more & better information!

Example

Medical Diagnostics

Functional Imaging - Positron Emission Tomography

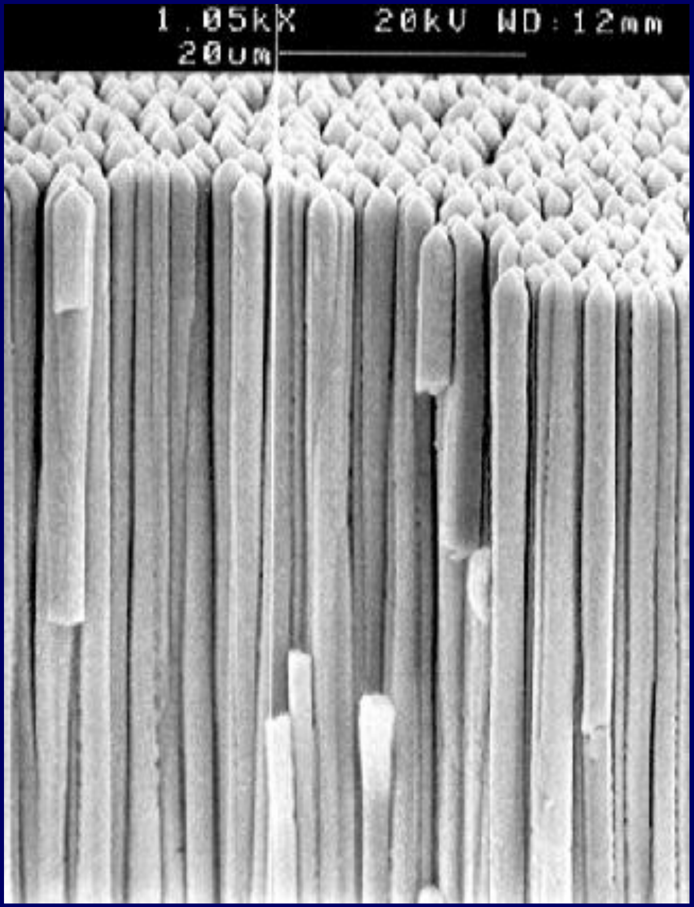


Siemens HRRT: **LSO scintillators** (7.5 x 2.1 x 2.1 mm^3)

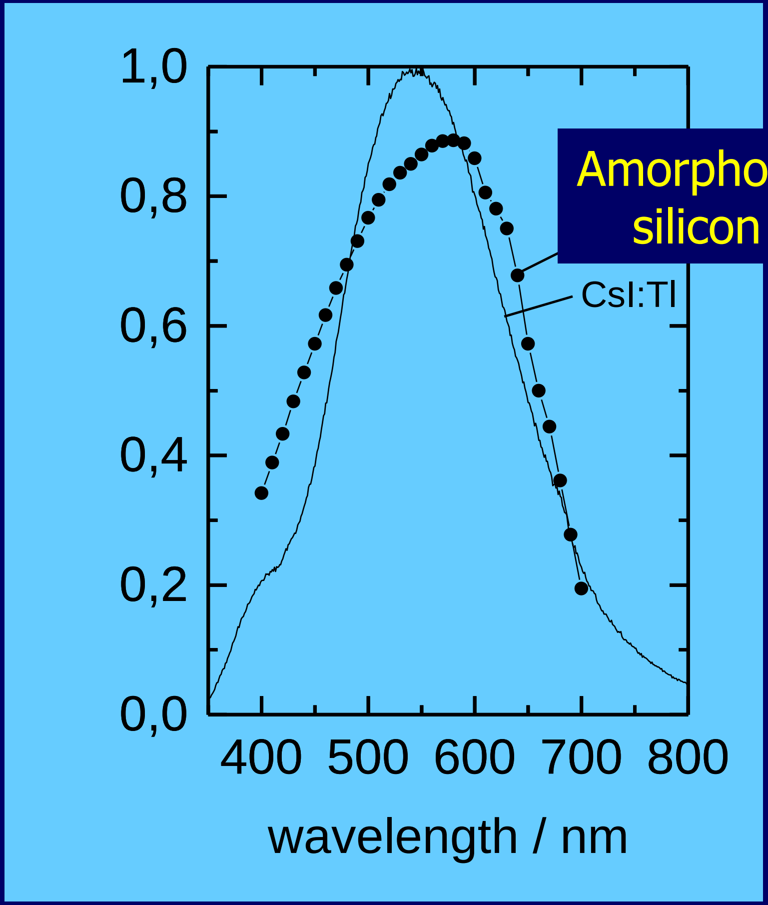
Efficient & fast response

X-ray imaging

CsI:Tl



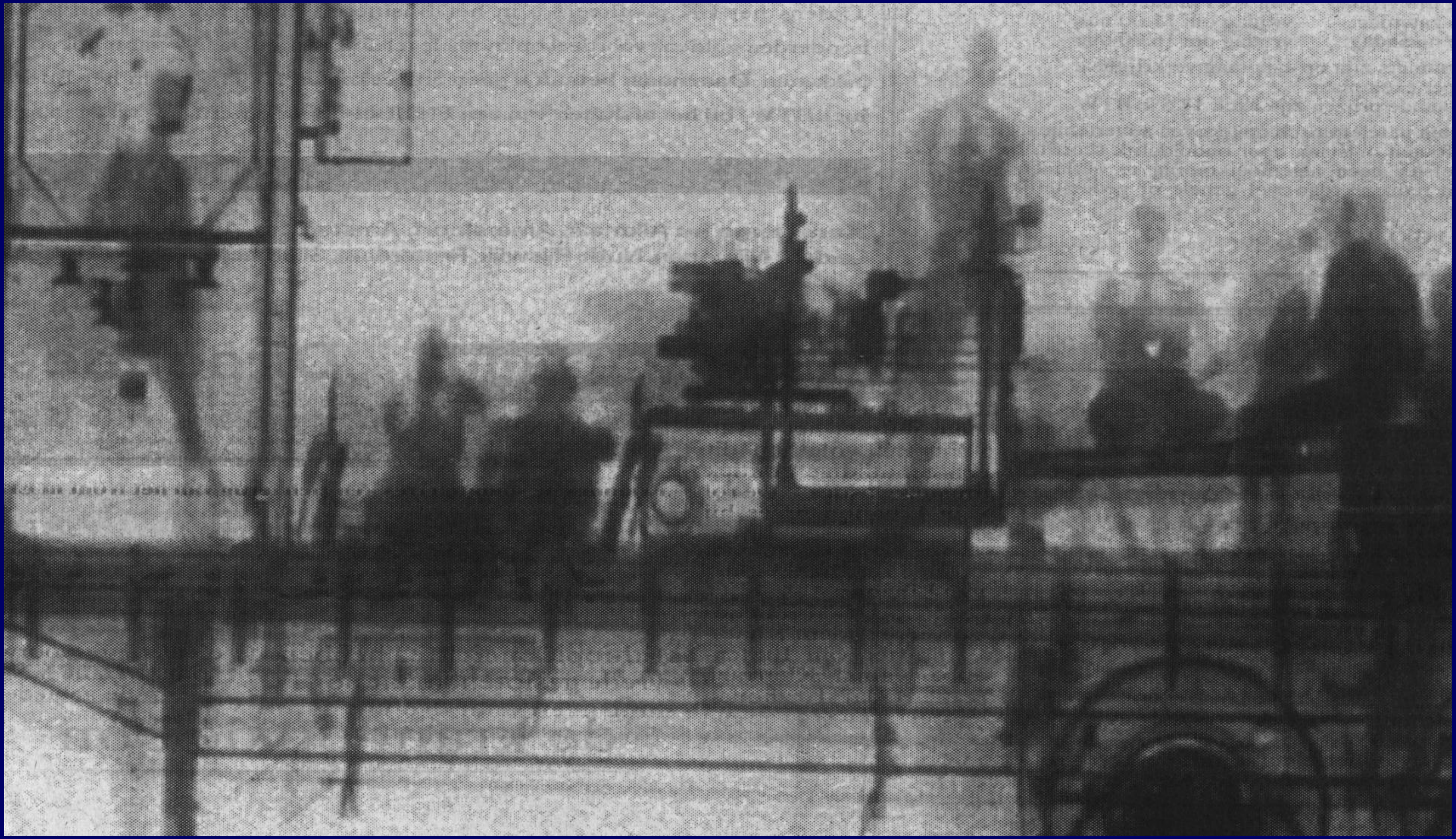
columnar growth



spectral sensitivity

From: Philips Aachen

Gamma-ray imaging: Security

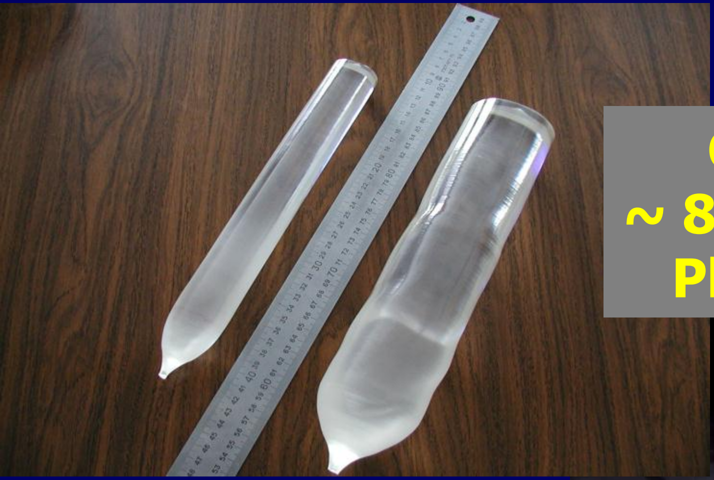


CsI:Tl

efficient at higher energies

Example

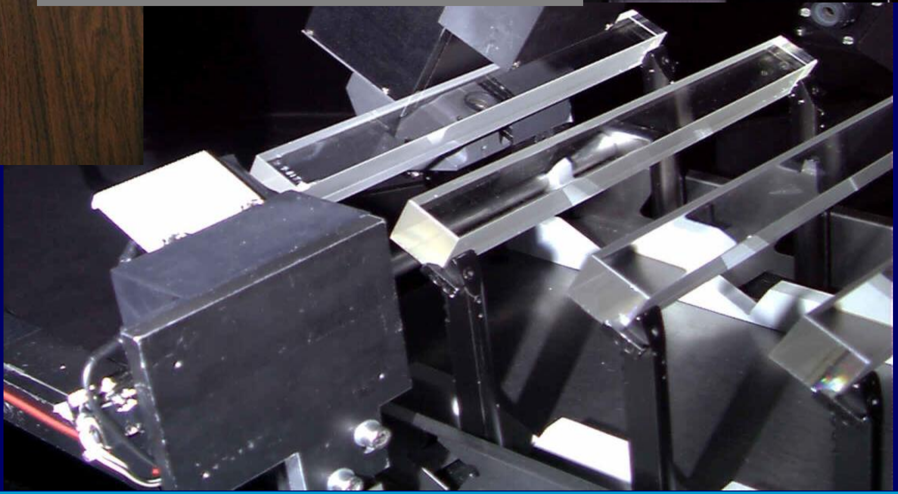
CERN
Electromagnetic Calorimeter



65mm diameter ingots
from Bogoroditsk

CMS + ALICE
~ 80,000 + 18,000
$PbWO_4$ crystals

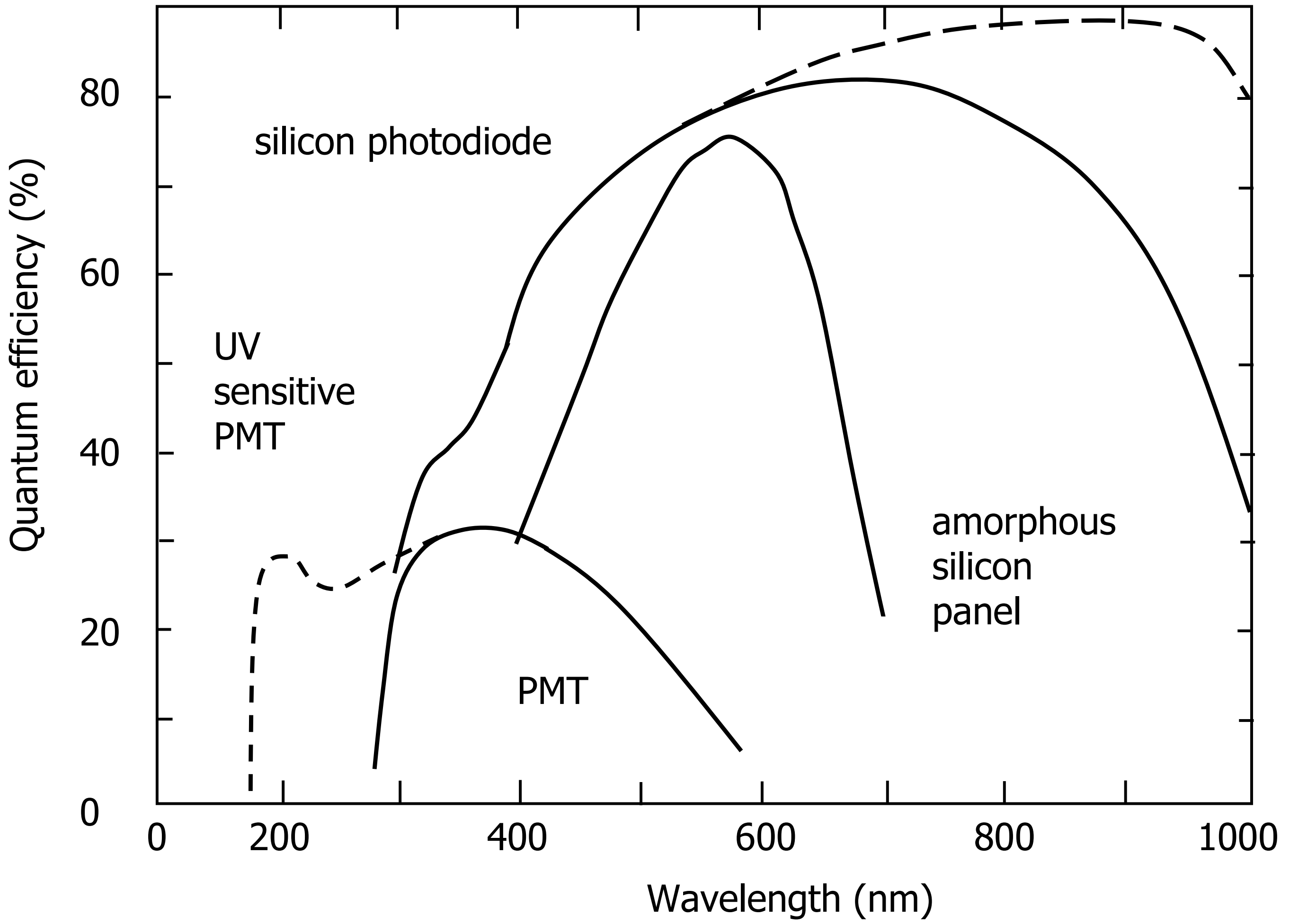

Quality control CMS



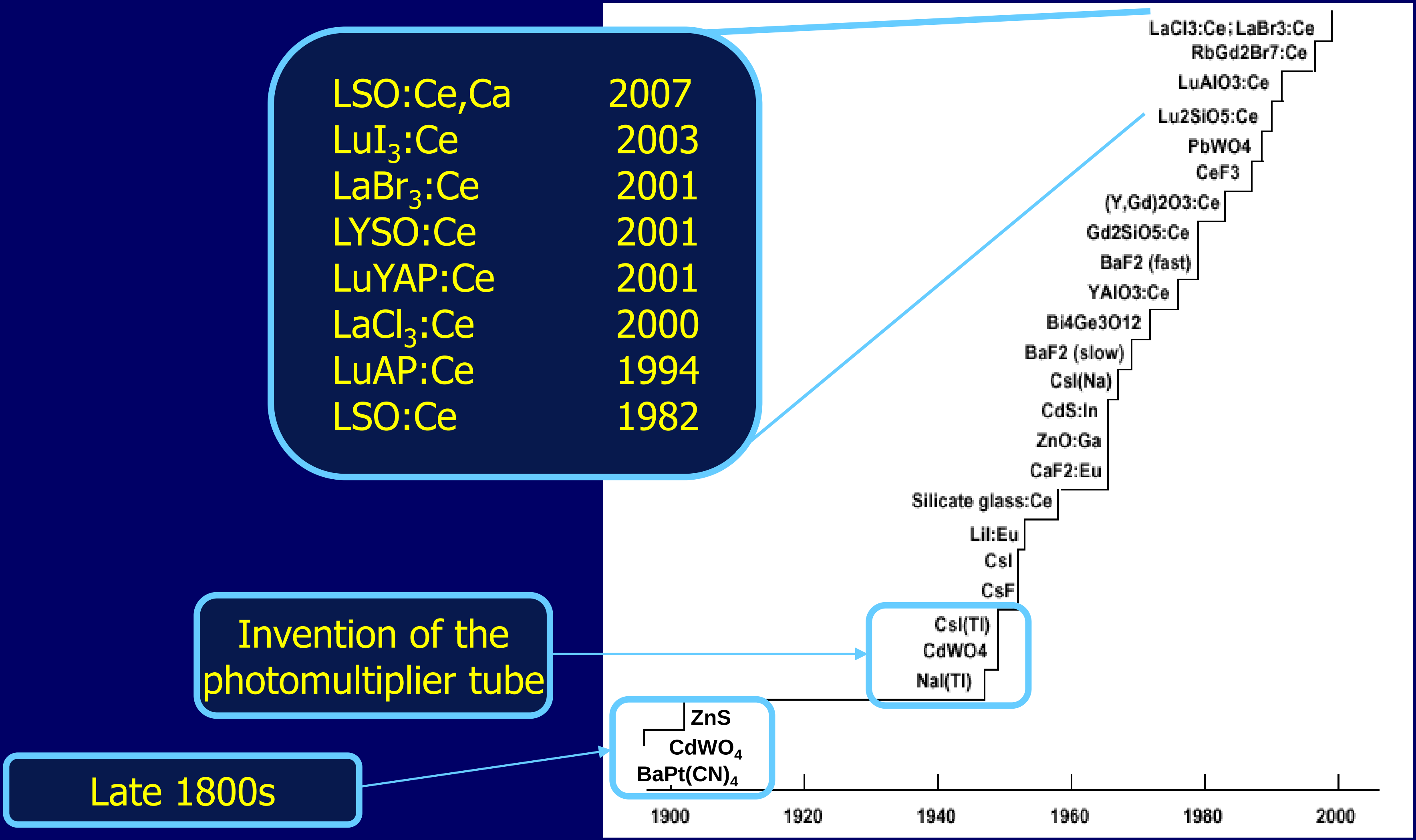
radiation length & fast

Silicon Avalanche Photodiodes

Inorganic scintillator R&D



History of scintillators



LSO:Ce,Ca	2007
LuI_3:Ce	2003
$LaBr_3$:Ce	2001
LYSO:Ce	2001
LuYAP:Ce	2001
$LaCl_3$:Ce	2000
LuAP:Ce	1994
LSO:Ce	1982

Invention of the photomultiplier tube

Late 1800s

M. J. Weber, J. Lumin. 100 (2002) 35

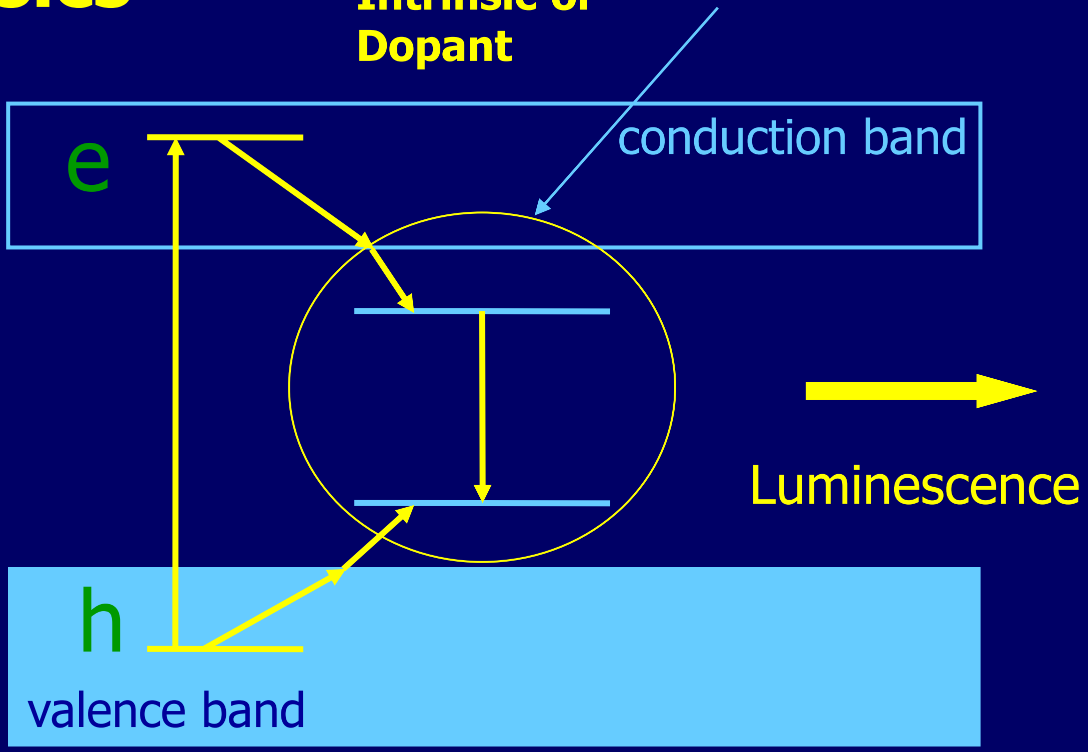
Scintillator Basics

Luminescence Centre – LC

Intrinsic or
Dopant

Ionic crystal
Host material

$E_{gap} > \sim 3 - 12$ eV



VIS	400 - 800 nm	3.1 - 1.6 eV
UV	180 - 400 nm	6.9 - 3.1 eV
VUV	< 180 nm	> 6.9 eV

absorption in air

thermalization
1 - 100 ps

Scintillation Light Yield Gamma-ray detection

$$\boxed{N_{photon} = \frac{E_{\gamma}}{E_{e-h}} \; S \; Q}$$

E_{γ} gamma-ray energy

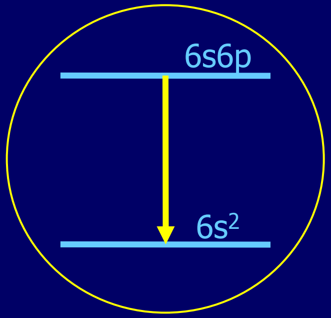
E_{e-h} = ~ 2.5 E_{gap}

$\Rightarrow$ # e-h pairs at bandgap

S transport/transfer efficiency to LC

Q quantum efficiency of LC

NaI:Tl Tl dopant is LC



Tl^+ ions ground state $6s^2$
excited state 6s6p

Quantum mechanics

Selection rules Allowed transitions

Radiationless relaxation

Singlet → singlet allowed, fast

Triplet → singlet forbidden, slow

NaI:Tl 230 ns **slow**

Gamma-rays ⟹ [

Interaction with host material

- high density
- high Z
- many scintillation photons → small Egap
- accomodates dopant

LC intrinsic or dopant

- efficient luminescence
- fast response

Find / Study **S = 1**

Q = 1

History of scintillators

Many with Ce doping

LSO:Ce,Ca	2007
LuI_3:Ce	2003
$LaBr_3$:Ce	2001
LYSO:Ce	2001
LuYAP:Ce	2001
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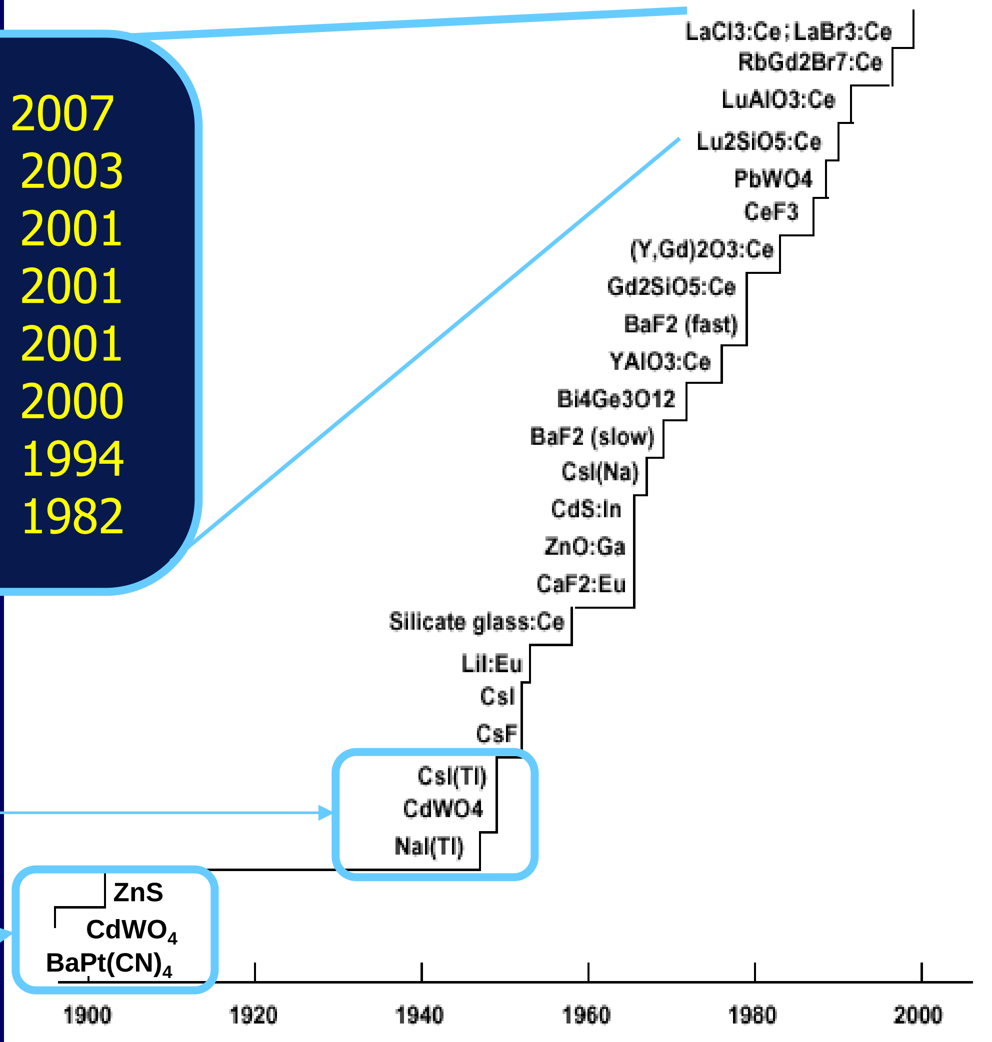


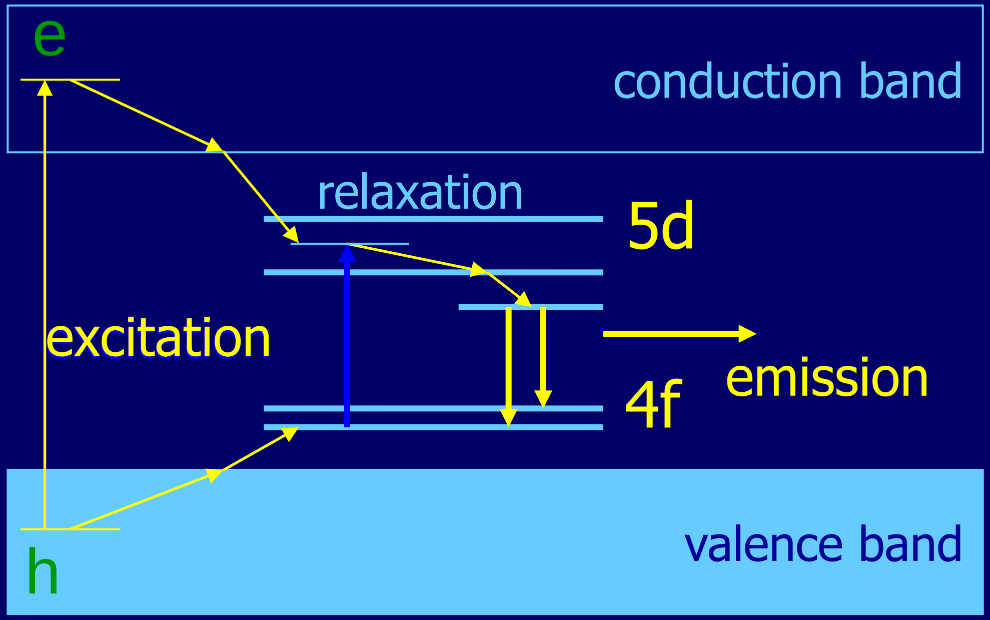
Invention of the photomultiplier tube

Late 1800s

M. J. Weber, J. Lumin. 100 (2002) 35

Why Ce?

Ce^{3+} LC



Ce^{3+}

Core +
1 electron in
4f state

5d → 4f allowed dipole transition

fast response

$\tau \sim 20$ ns

Interaction with host material

Gamma-rays

high density
high Z
many photons → small Egap
accomodates dopant

host material with

3+ ion

Y, La, Gd, Lu

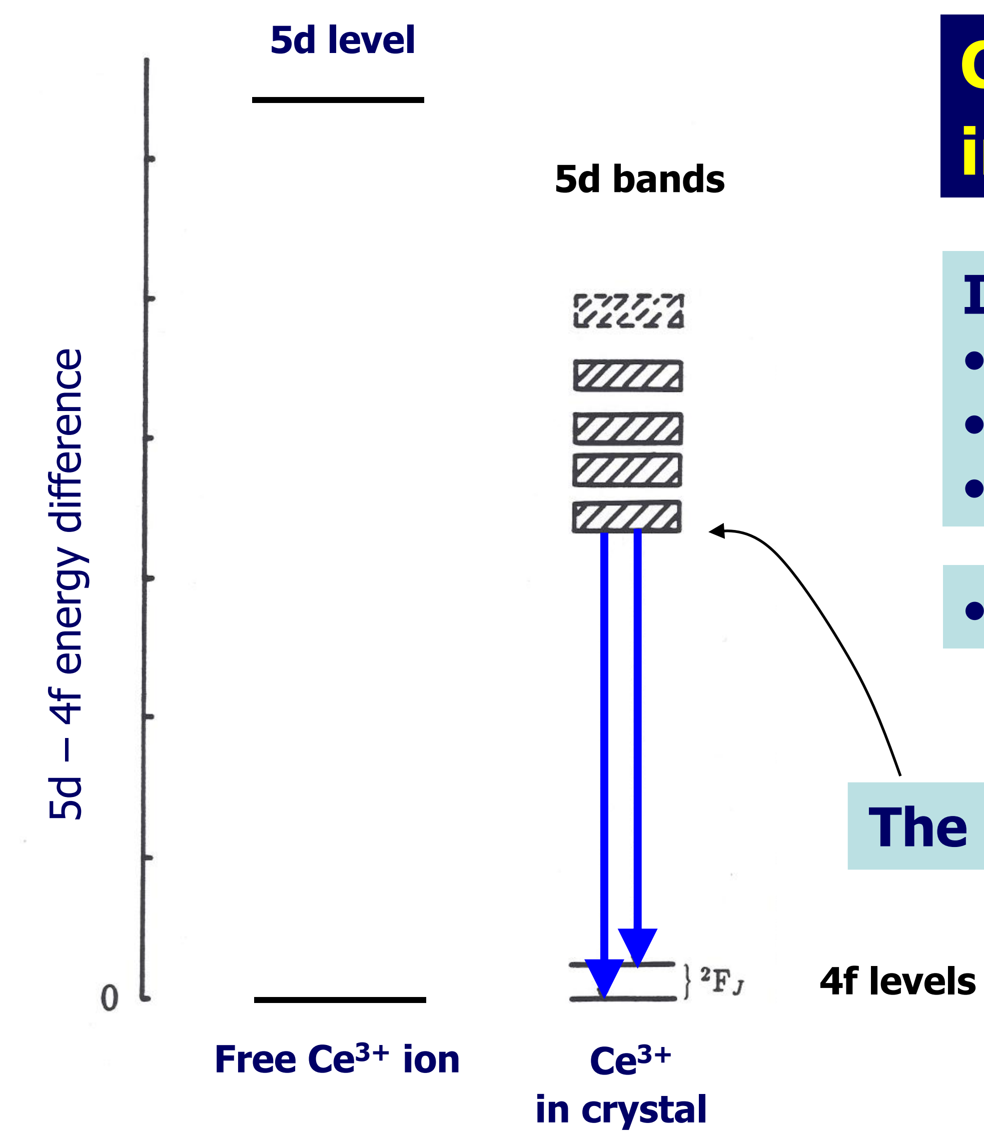
LC dopant

efficient luminescence
fast response

⇒ **Ce**$^{3+}$ ions

Find / Study
S = 1
Q = 1

Ce^{3+} 5d-4f level distance in host material



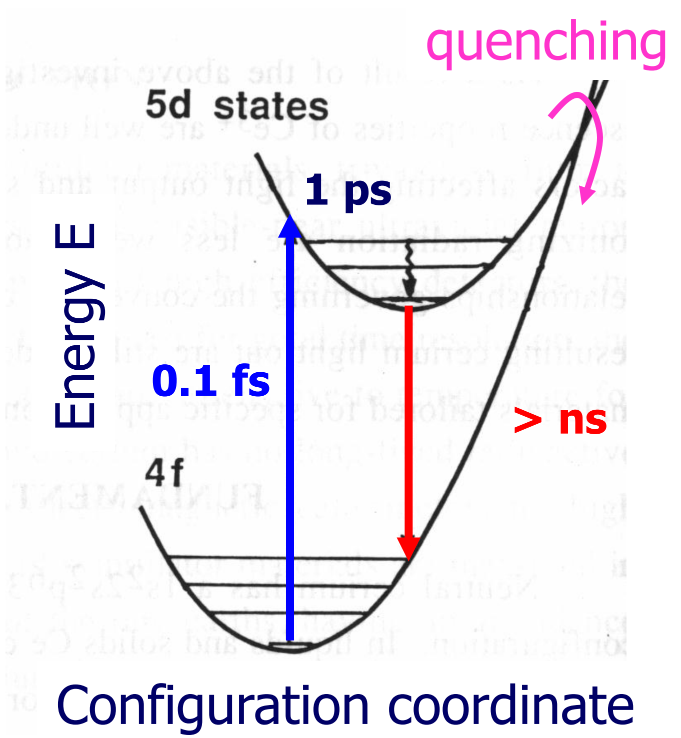
In crystal

- 5d level shifts down
- bands
- split by crystal field

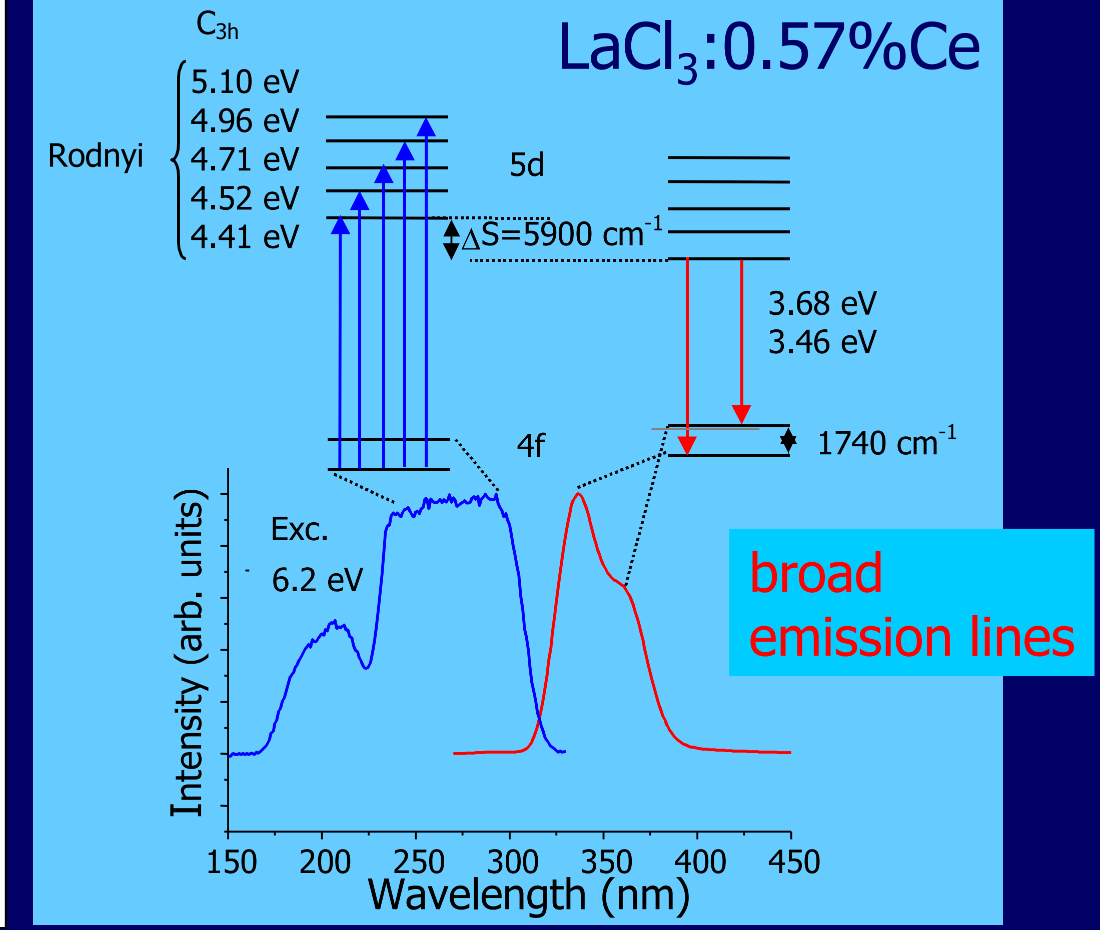
- 4f levels hardly affected

The lowest 5d-band edge matters

Ce^{3+} Relaxation ⟹ Stokes shift



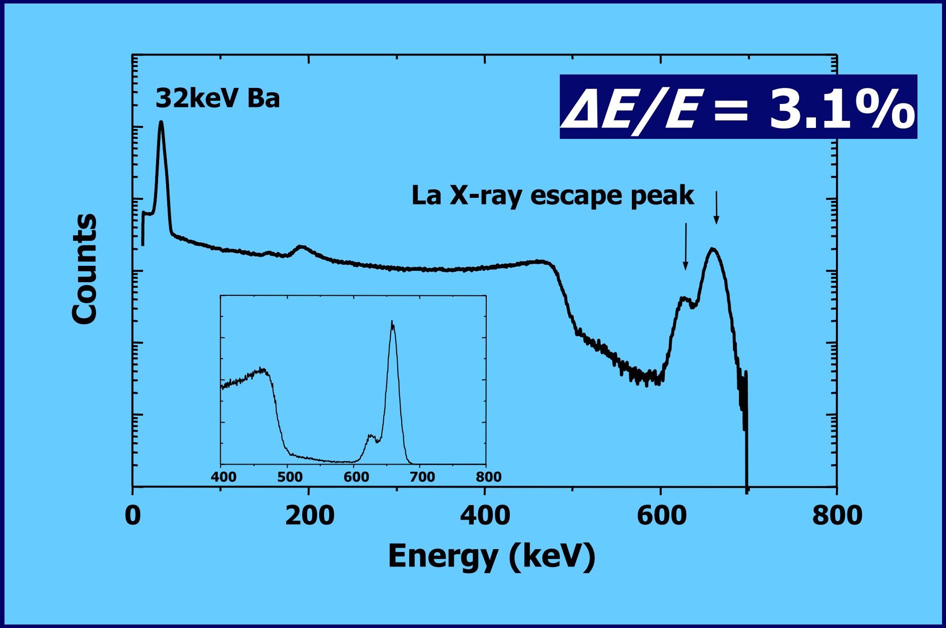
4f levels shielded
no line broadening



$LaCl_3$:0.57%Ce

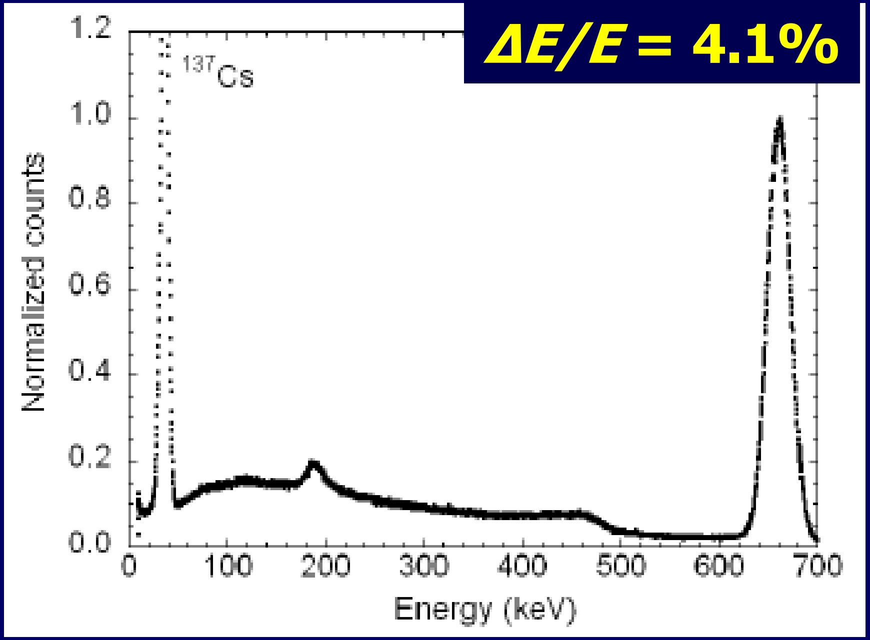
broad emission lines

Inorganic scintillator R&D



ΔE/E **= 3.1%**

$LaCl_3$:Ce
4″ x 6″



ΔE/E **= 4.1%**

$LaCl_3$:Ce
4 mm x 6 mm

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.

$LaCl_3$:Ce

4″ x 6″

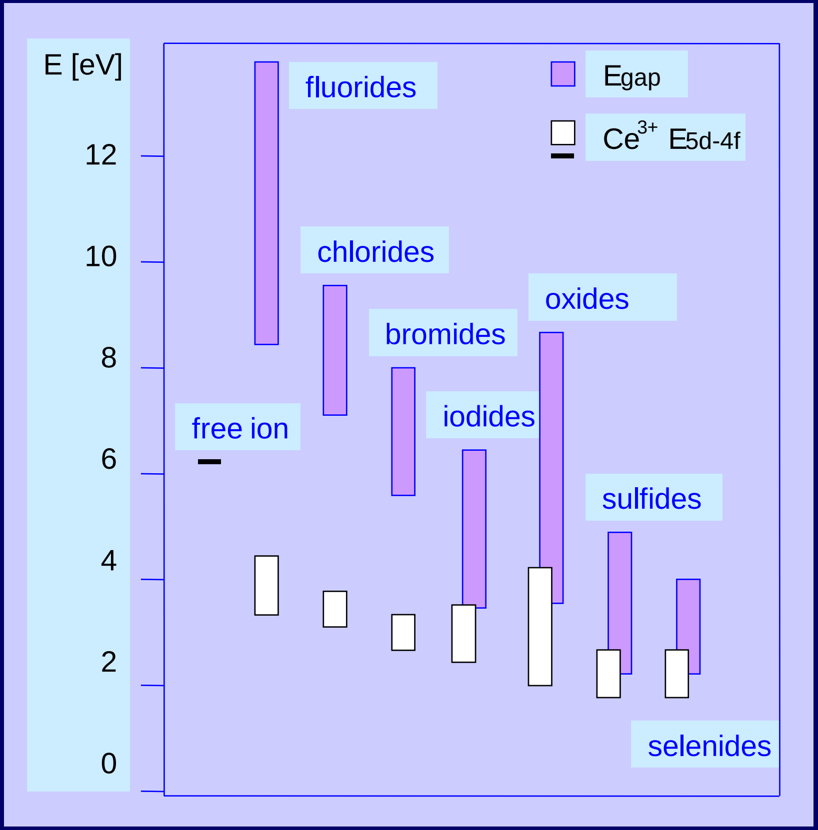
BrilLanCe™ 350

It's what's Inside that Counts!

Scintillation Material

Fast scintillation mechanism

Ce^{3+} levels and E$_{gap}$



Ce^{3+} 5d → 4f

τ ~ 20 ns

in favourable host

$$N_{photon} = \frac{E_{gamma}}{2.5\,E_{gap}}\ S\ Q$$

Egap ⇩ LY ⇧

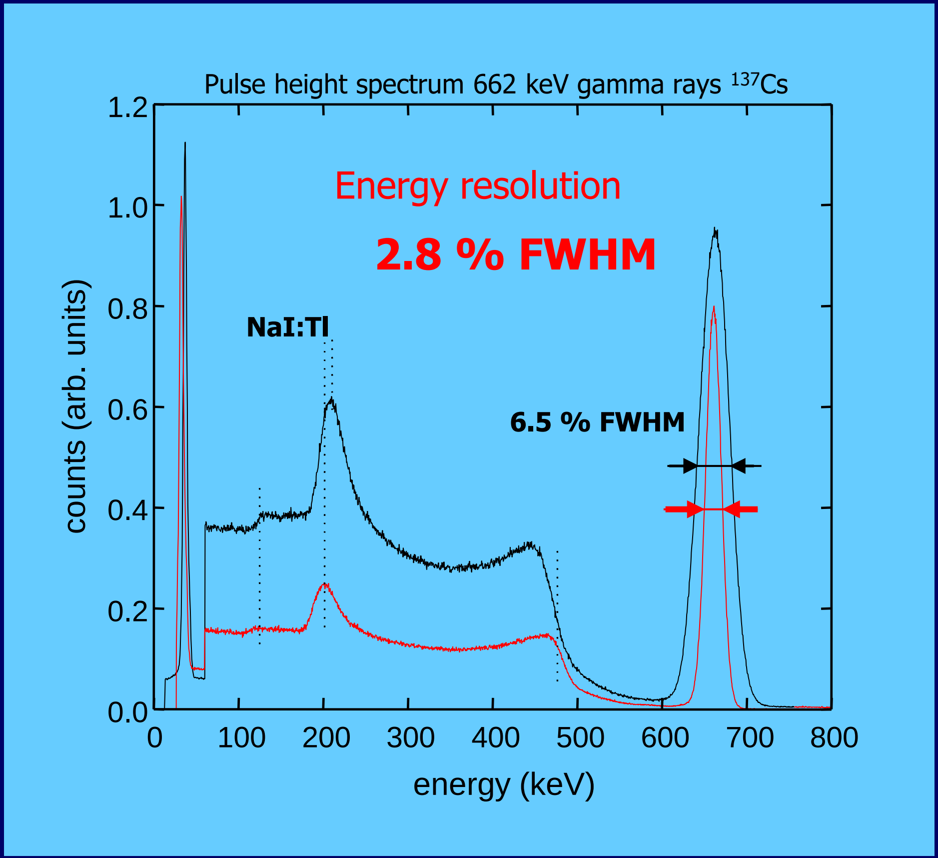
Egap ⇩ λ ⇧

Matching light sensor

$LaBr_3$: 5%Ce^{3+}

BrilLanCe™ 380

Scintillation Material



Light yield

70,000 photons/MeV

(NaI:Tl 40,000 ph/MeV)

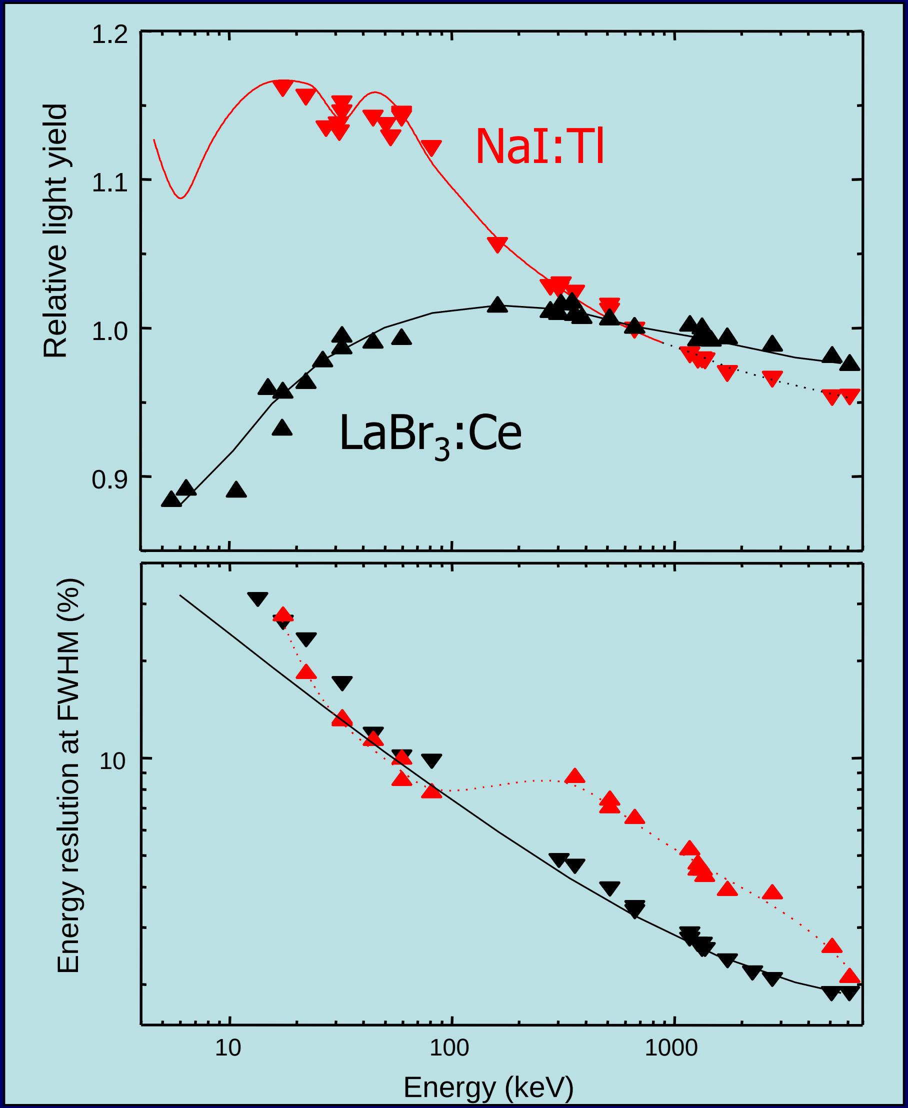
Decay time

16 ns

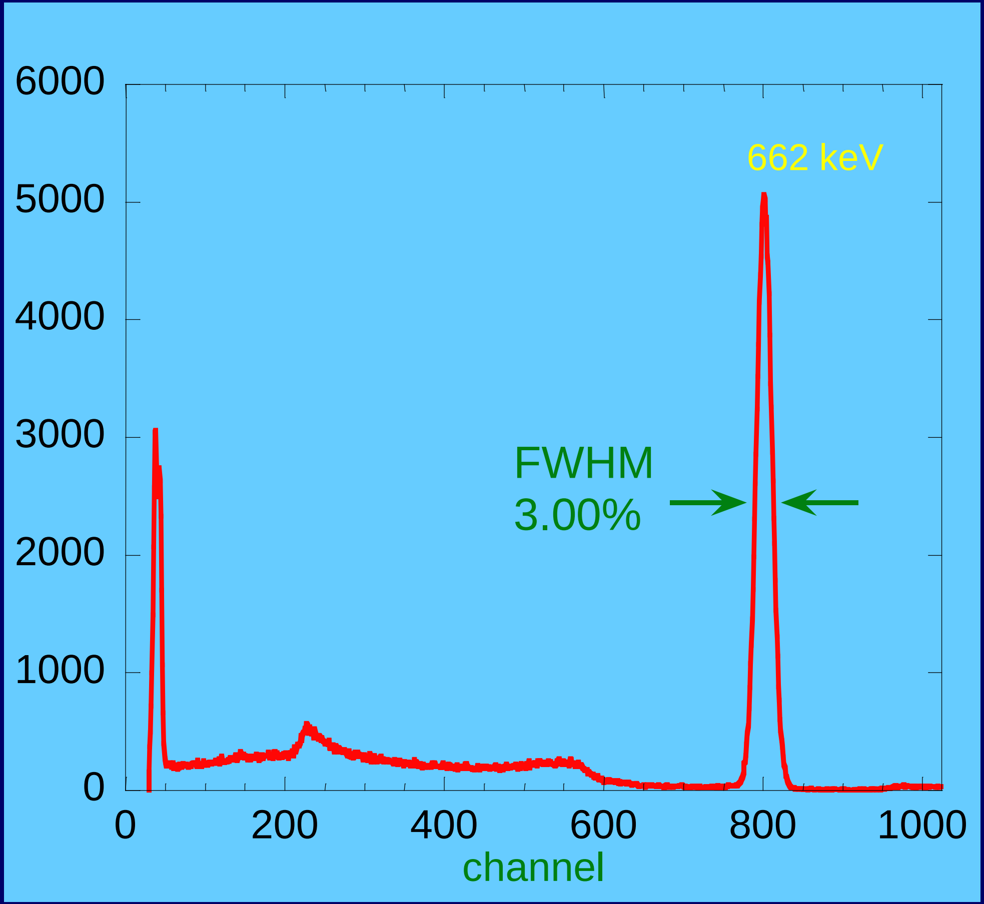
(NaI:Tl 230 ns)

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

Non-Proportionality and **Energy Resolution**



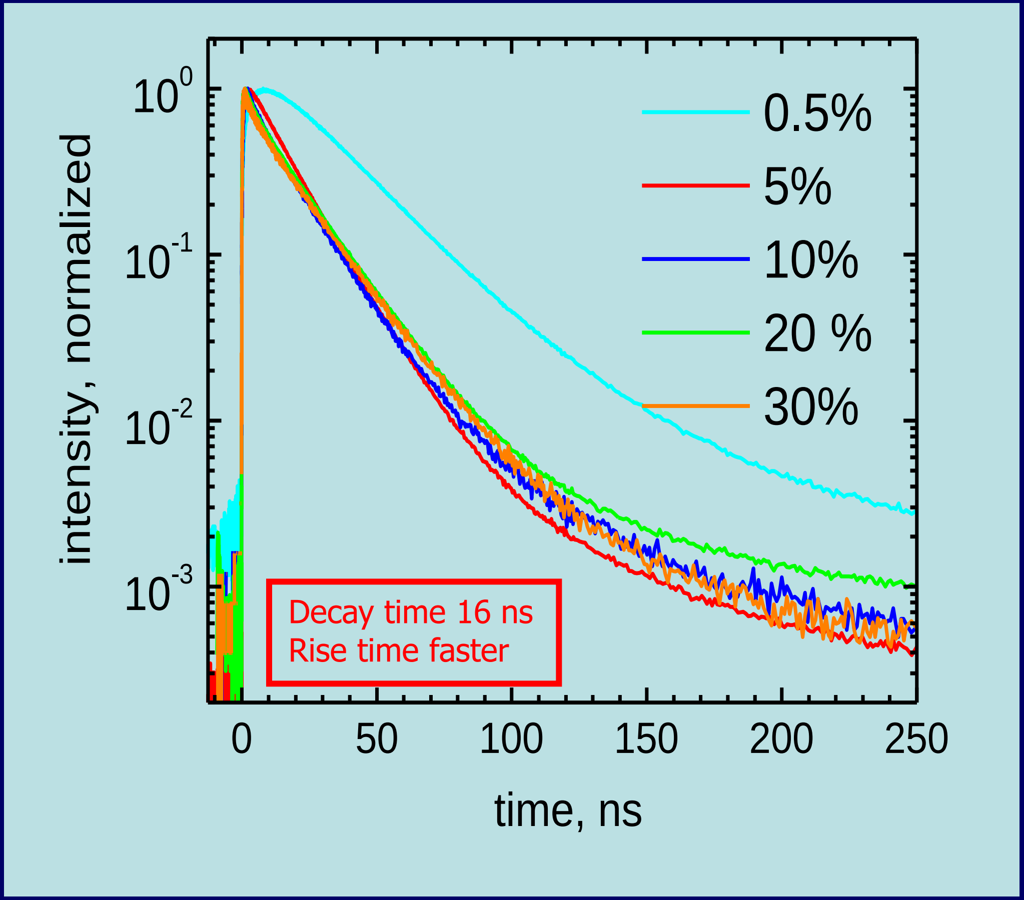
$LaBr_3$:Ce
3″ x 3″



BrilLanCe™ 380
It's what's Inside that Counts!
Scintillation Material

345 cm^3 volume

Decay Time for $LaBr_3$:Ce & Time Resolution



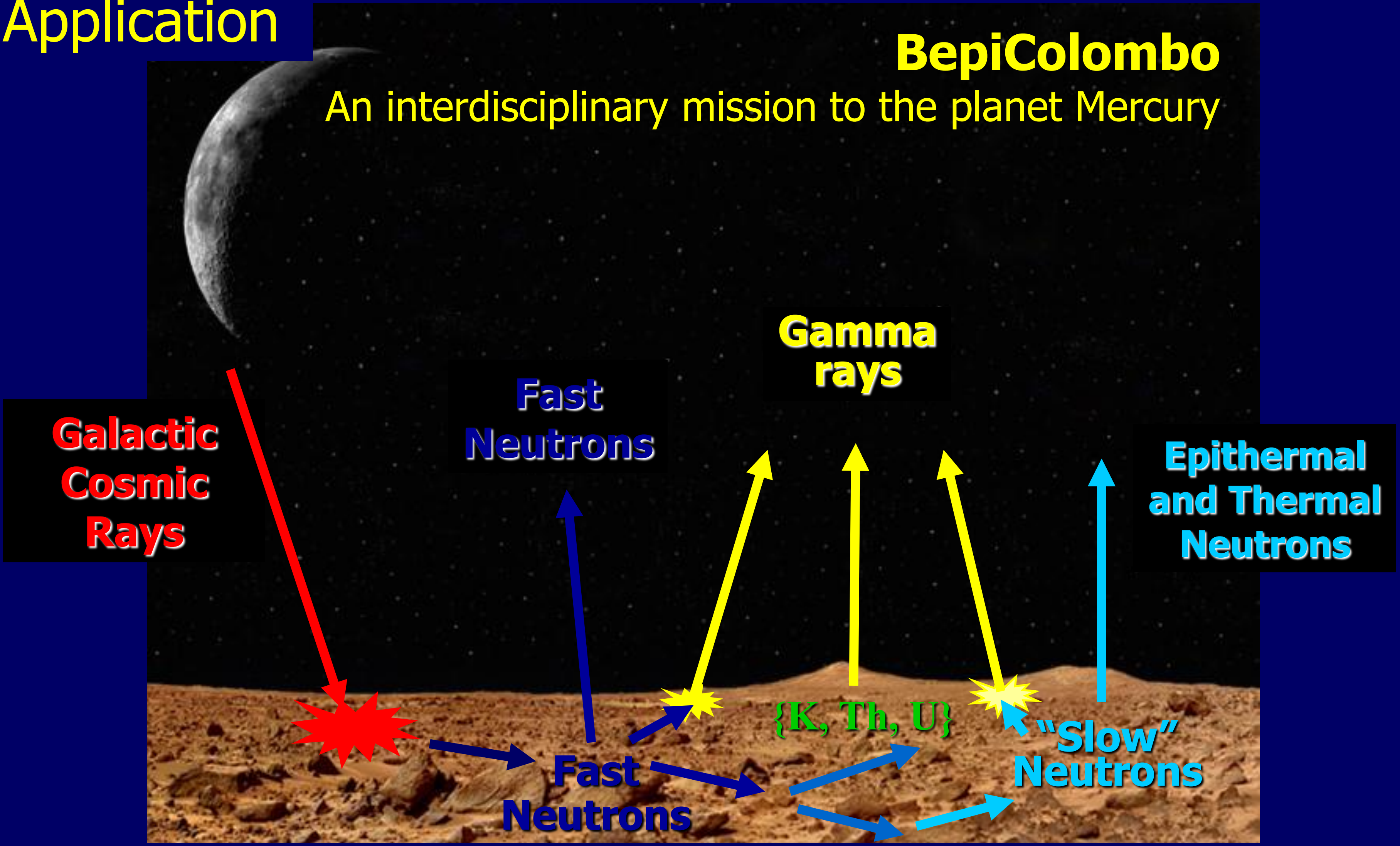
Decay time 16 ns
Rise time faster

511 keV - 511 keV

< 300ps

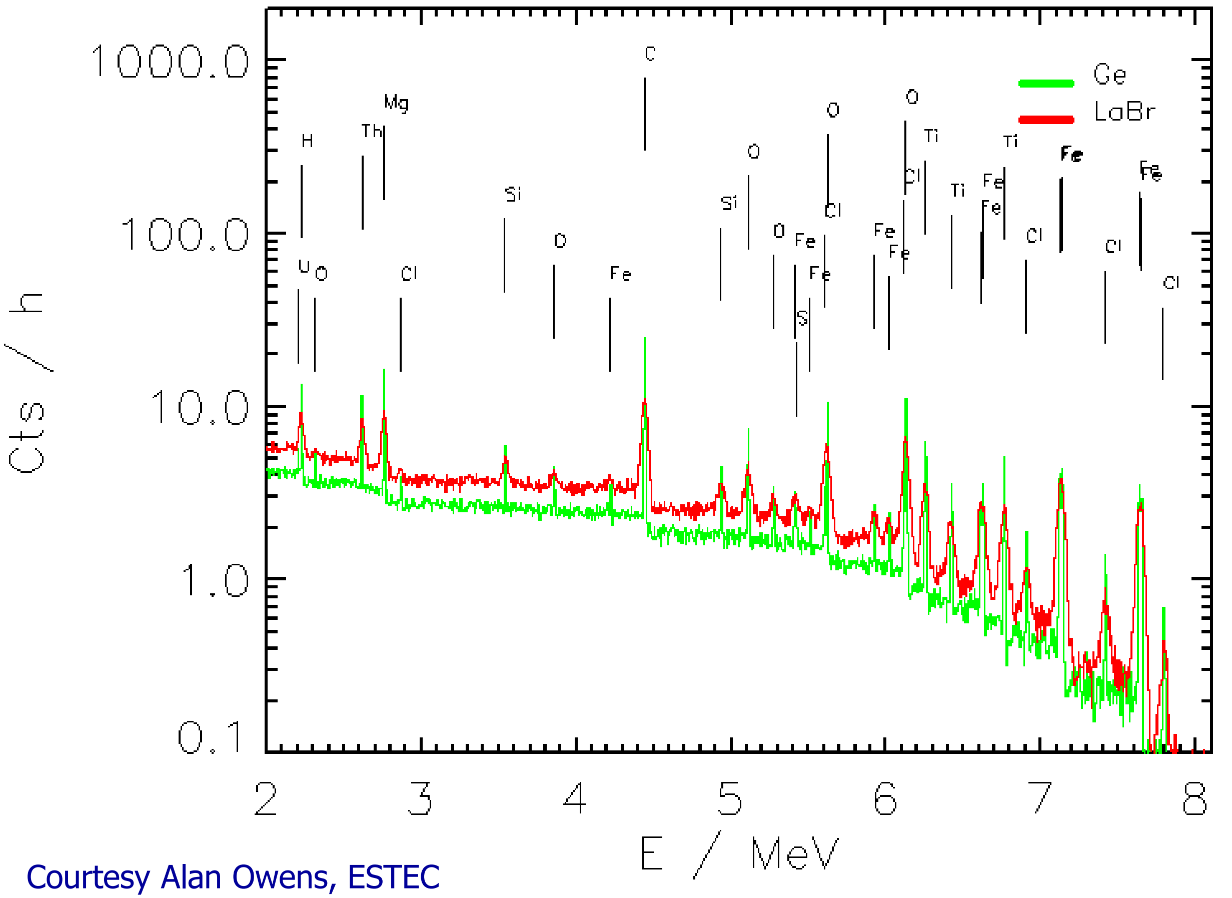
Courtesy Kanai Shah, RMD

Application



Courtesy Alan Owens, ESTEC

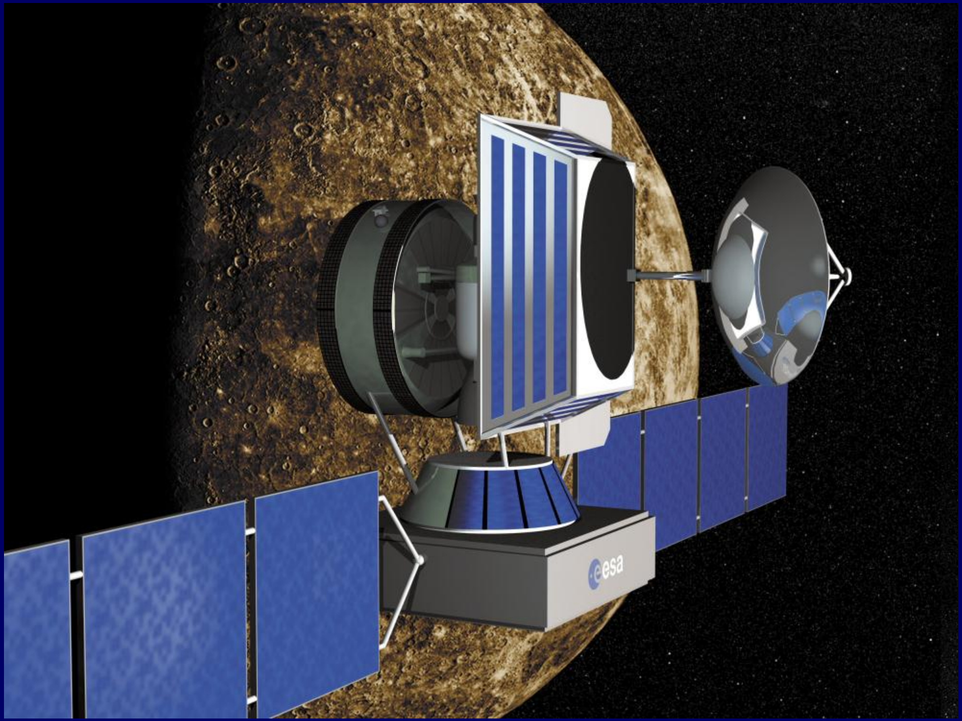
Simulated in orbit spectra measured by a 6.5 cm × 6.5 cm diameter Ge crystal and an 8 cm diameter LaBr detector



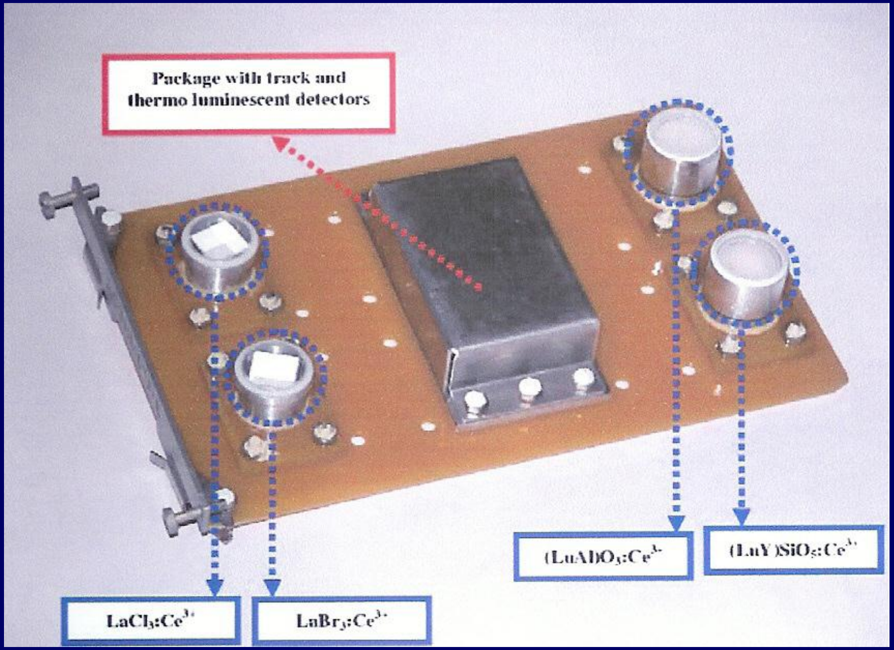
Courtesy Alan Owens, ESTEC

Mercury mission

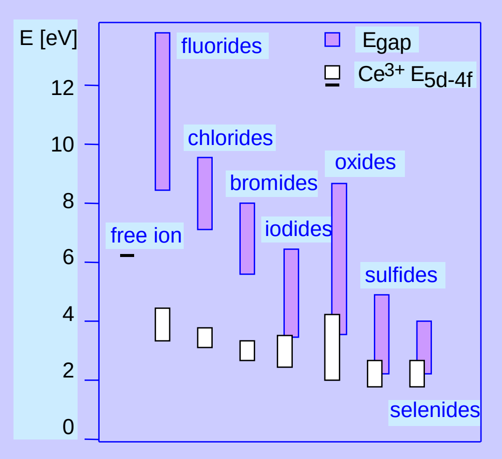
BepiColombo
An interdisciplinary mission to the planet Mercury

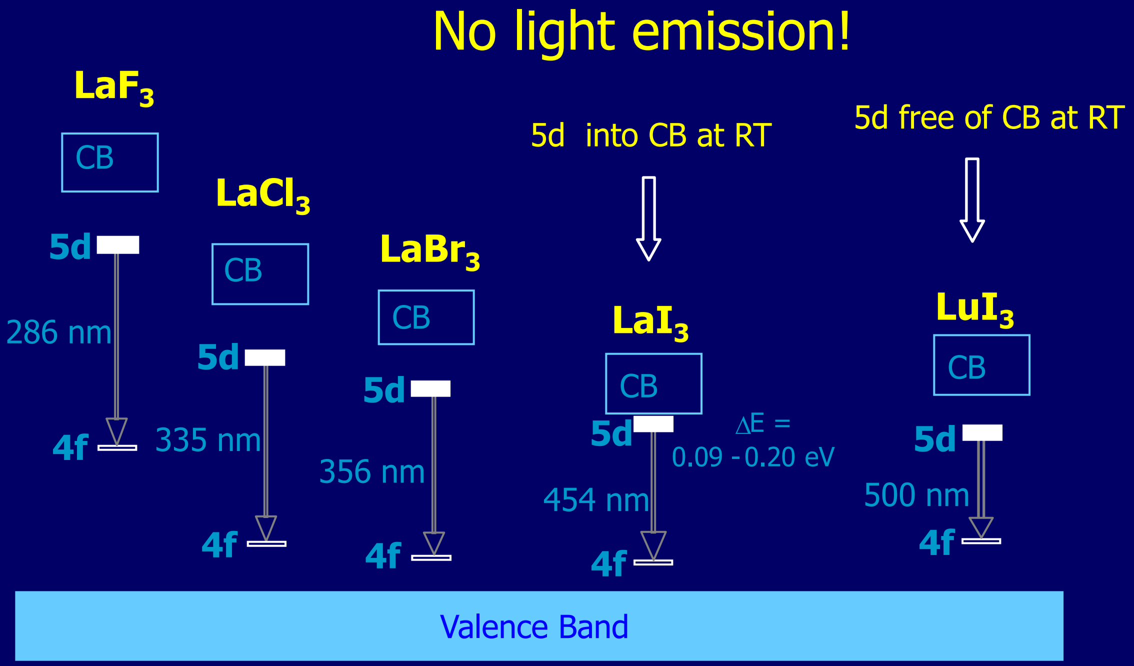


Board with scintillators is orbiting the earth
in ISS for 1 – 2 years
Test on radiation damage



Energy level shifts





No light emission!

LaF_3
CB
5d
286 nm
4f

$LaCl_3$
CB
5d
335 nm
4f

$LaBr_3$
CB
5d
356 nm
4f

5d into CB at RT

LaI_3
CB
5d
ΔE =
0.09 - 0.20 eV
454 nm
4f

5d free of CB at RT

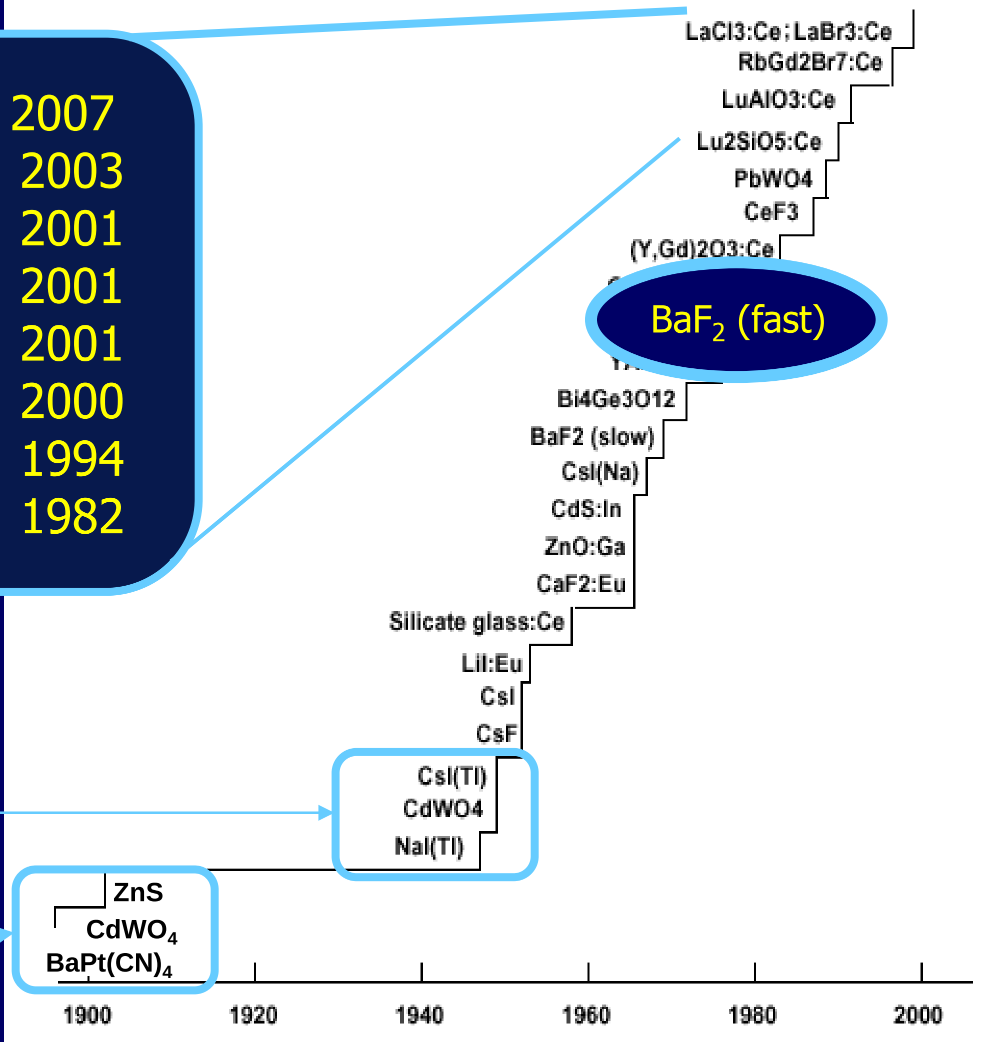
LuI_3
CB
5d
500 nm
4f

Valence Band

Summary scintillator specs

	Density g/cm^3	Attenuation length at 511 keV mm	Photoel effect %	Light yield phot/MeV	Decay time ns	Emission max nm
NaI:Tl	3.67	29.1	17	41,000	230	410
$Bi_4Ge_3O_{12}$ (BGO)	**7.1**	10.4	40	9,000	300	480
BaF_2	4.88			1500 11,000	**~0.7** 600	220 310
Lu_2SiO_5:Ce (LSO)	7.4	11.4	32	**26,000**	**40**	420
$LuAlO_3$:Ce (LuAP)	**8.3**	10.5	30	11,000	**18**	365
$LaBr_3$:Ce	5.07	22.3	13.1	**70,000**	**16**	380
LuI_3:Ce	5.6	18.2	28	**90,000**	6-140 (72%)	472, 535
$PbWO_4$ (PWO)	**8.3**	**8.7**	42	200	**15**	420

History of scintillators



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LYSO:Ce	2001
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$LaCl_3$:Ce	2000
LuAP:Ce	1994
LSO:Ce	1982

BaF_2 (fast)

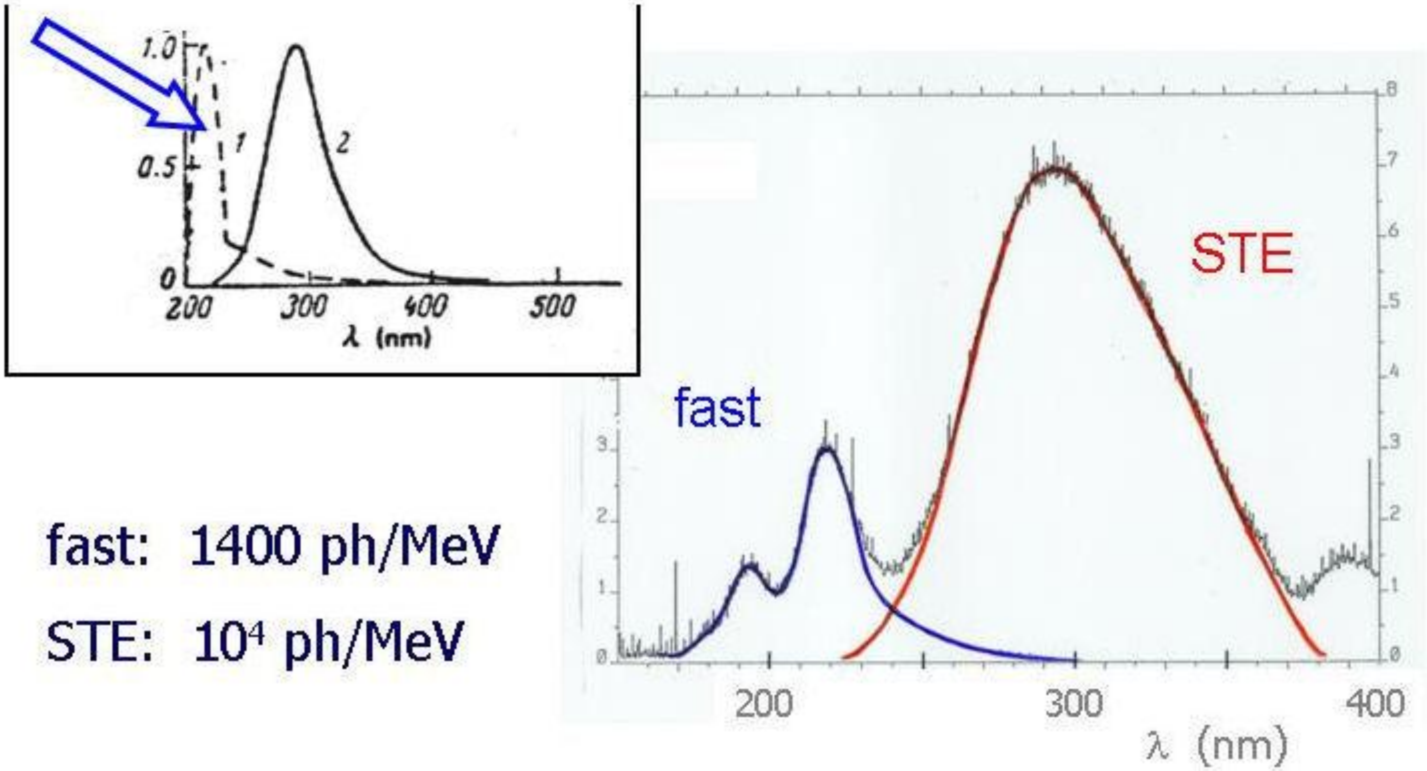
Invention of the photomultiplier tube

Late 1800s

M. J. Weber, J. Lumin. 100 (2002) 35

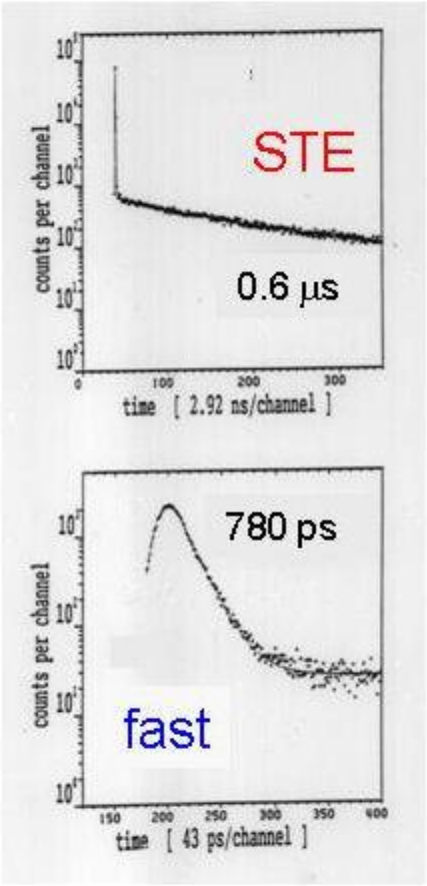
BaF_2 - fast UV luminescence

N.N. Ershov, N.G. Zakharov, P.A. Rodnyi
Experiment 1980; Publ: Opt. Spektrosk. 53(1982)89-93

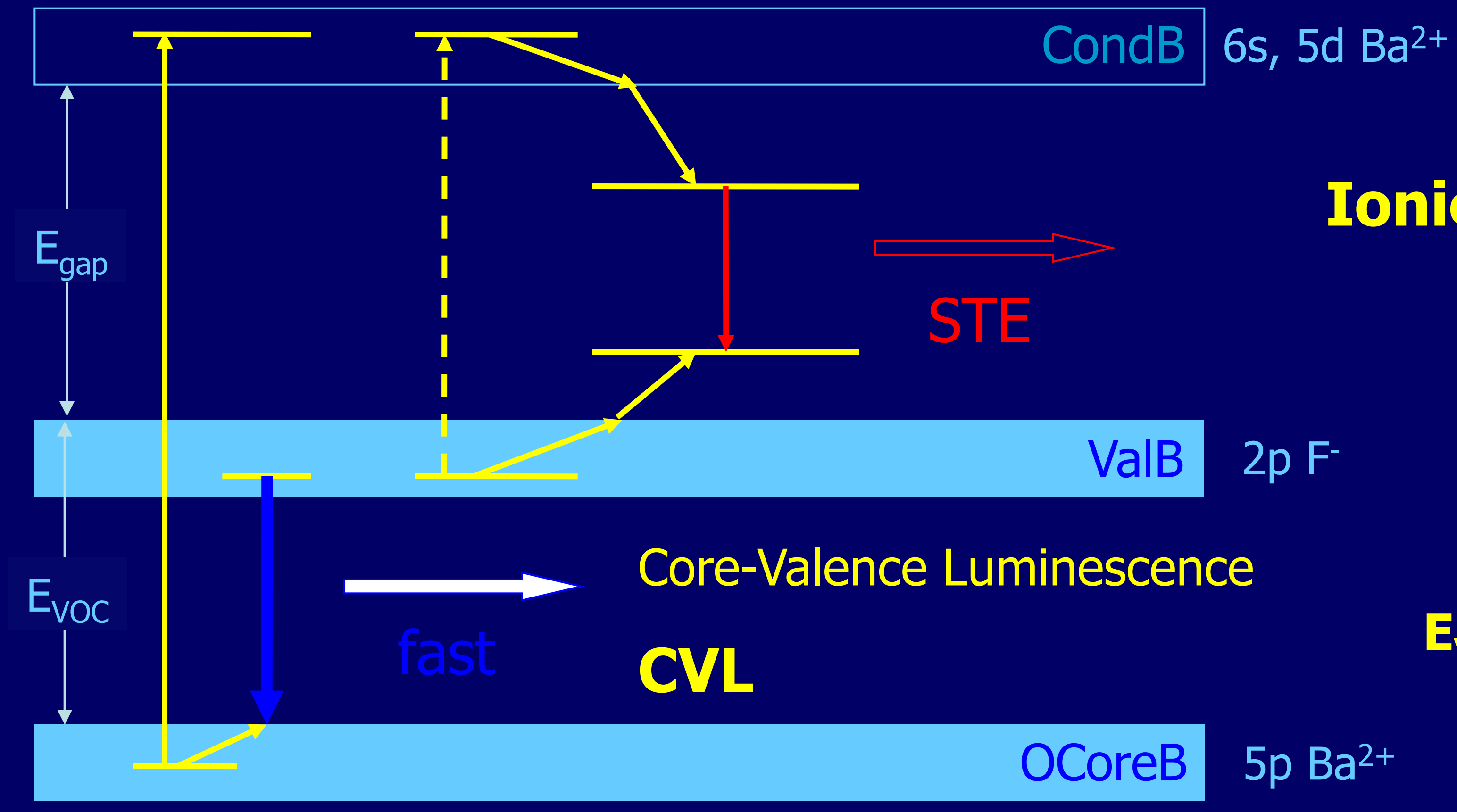


fast: 1400 ph/MeV

STE: 10^4 ph/MeV



BaF_2 fast UV luminescence - the model



Core-Valence Luminescence

CVL

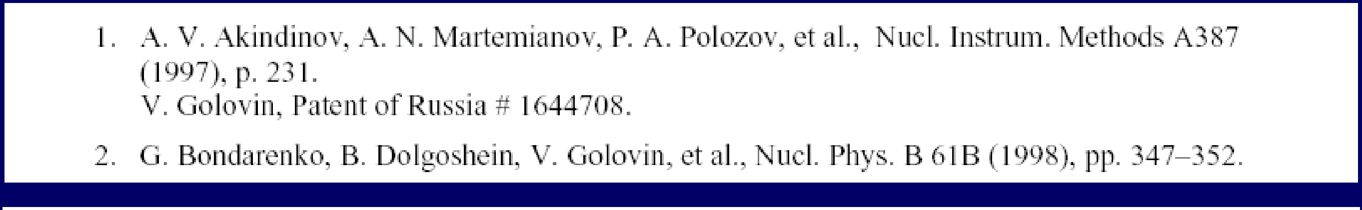
Ionic crystal

$E_{VOC} < E_{gap}$

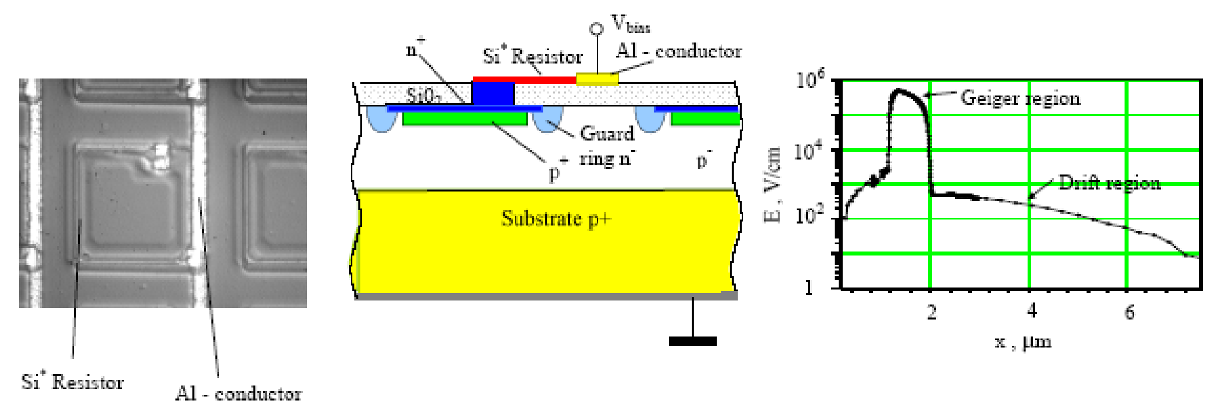
Yu.M. Aleksandrov *et al*, Sov. Phys. Sol. State 26(1984)1734

21st century

Introduction **new light sensor -** the **Silicon PM** - SiPM



1. A. V. Akindinov, A. N. Martemianov, P. A. Polozov, et al., Nucl. Instrum. Methods A387 (1997), p. 231.
   V. Golovin, Patent of Russia # 1644708.
2. G. Bondarenko, B. Dolgoshein, V. Golovin, et al., Nucl. Phys. B 61B (1998), pp. 347–352.



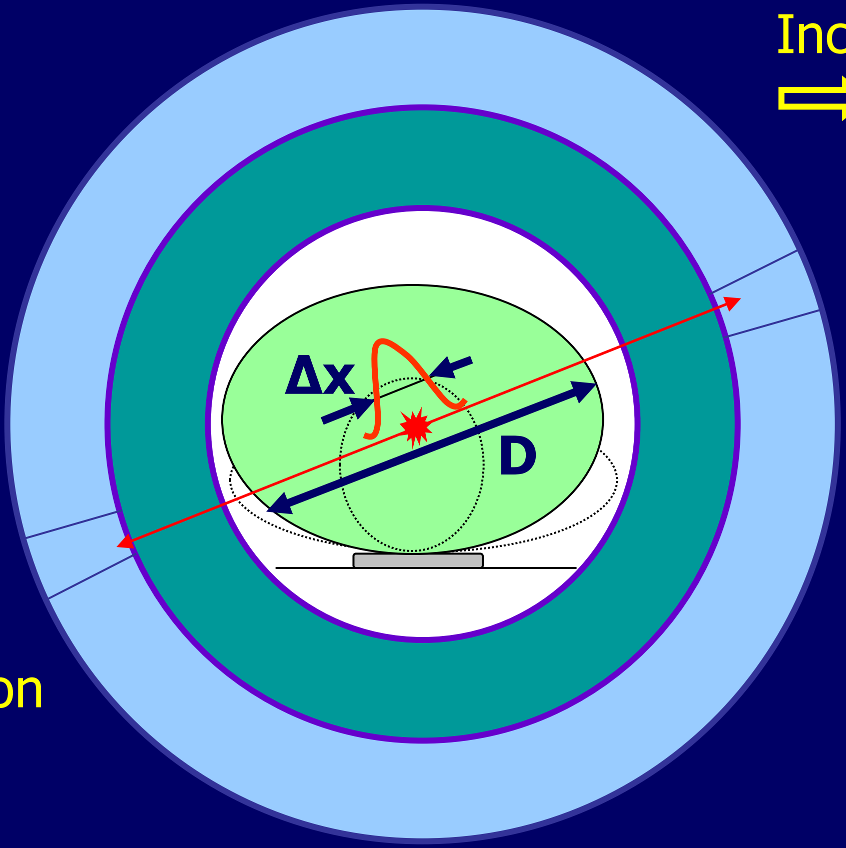
Thousands of pixels per mm^2
Light detection efficiency ~50% at 400 nm
Very fast response

PET

Time resolution

⇩

Time of Flight information



Increasing Obesity
⇨ Background
from
Scattering!

Δx = uncertainty in position along LOR

= c · Δt/2

TOF 'Sensitivity' gain ~ D/Δx

Δt = 300 ps ⇨ Δx = 4.5 cm

⇨ > 5 x better Signal to Noise ratio

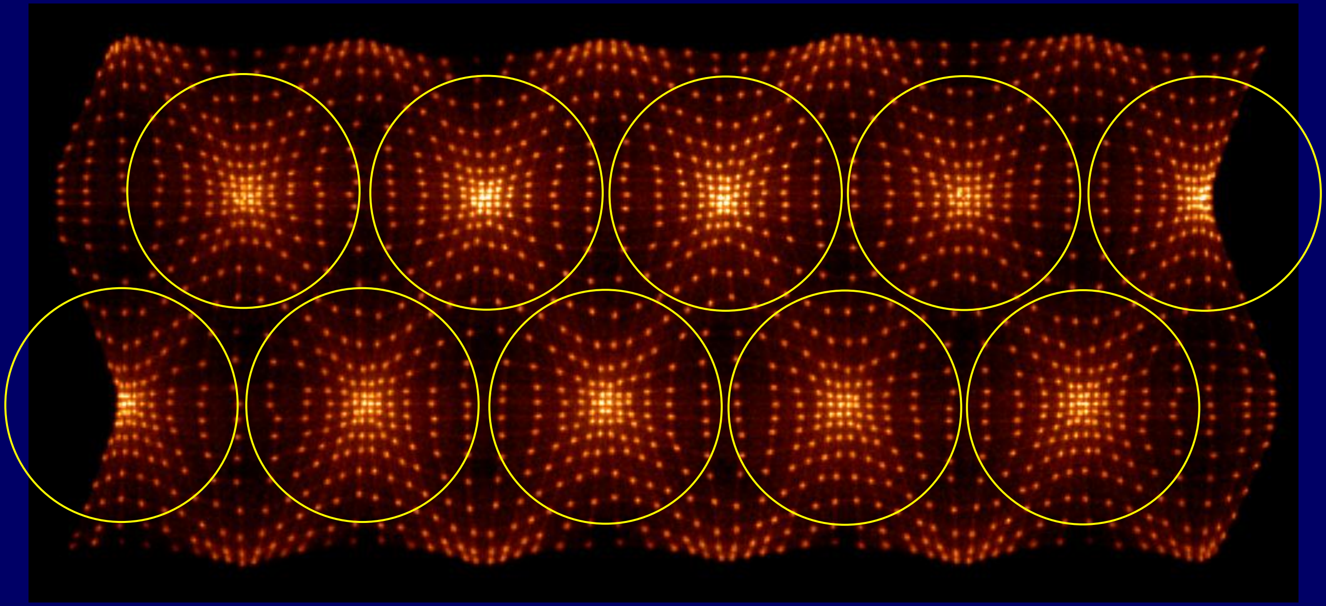
$LaBr_3$:Ce

Positron Emission Tomography

Full Module

1620 (60x27) 4mm x 4mm x 30mm $LaBr_3$:Ce crystals

10 PMTs - 50 mm diam - Raw Signals

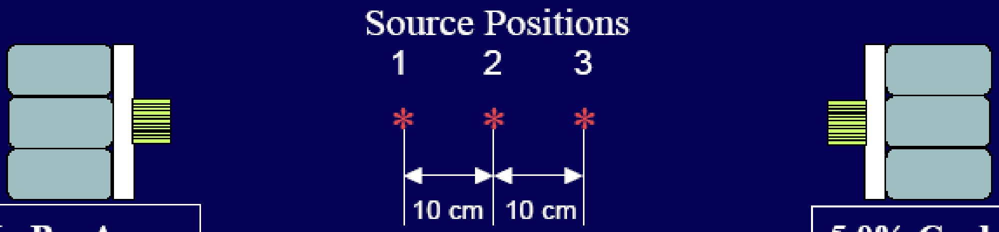


courtesy Philips Research Laboratories

PET

LaBr timing performance

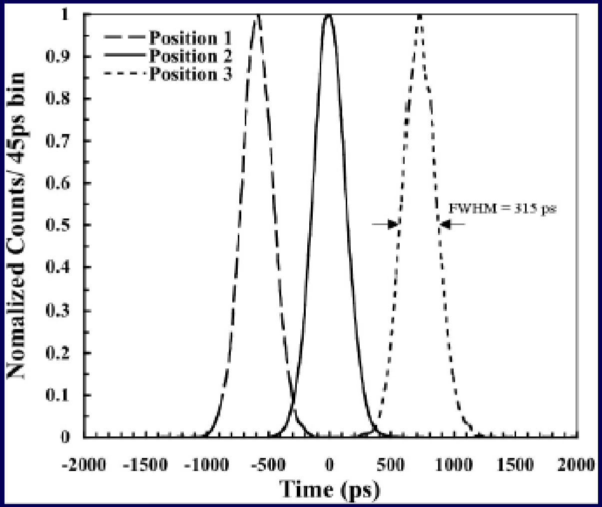
2 arrays in coincidence



5.0% Ce-doped LaBr$_3$ Array & 7 PMT cluster

5.0% Ce-doped LaBr$_3$ Array & 7 PMT cluster

Measured time spectra for three source positions

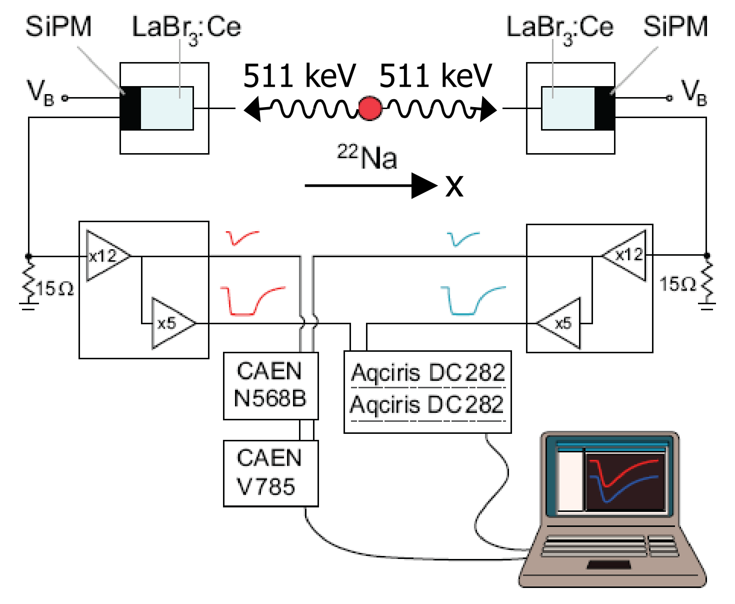


Coincidence timing resolution = 315 ps ⟷ **5 cm position resolution**

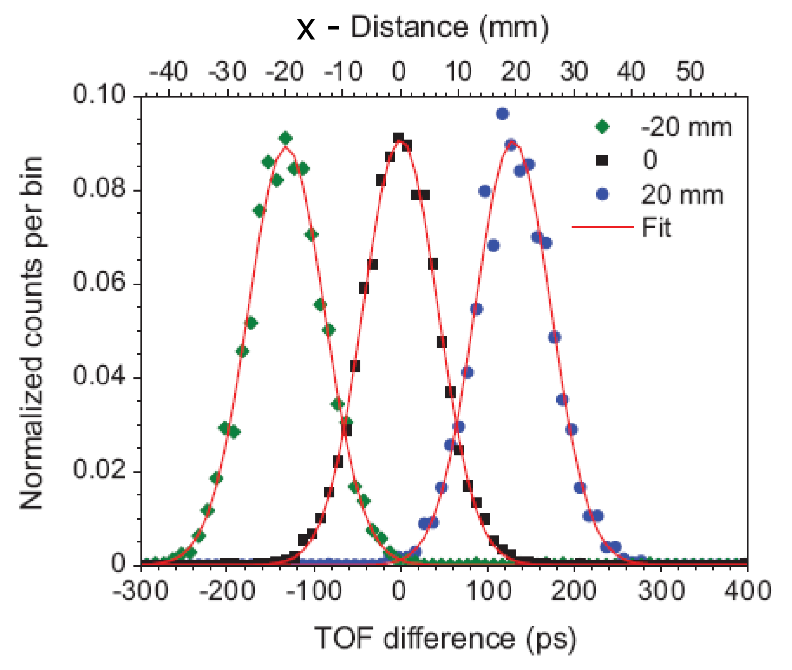
courtesy Philips Research Laboratories

LaBr3:Ce + SiPM

Time resolution 100 ps



D.R. Schaart et al



1.5 cm pos. resol.

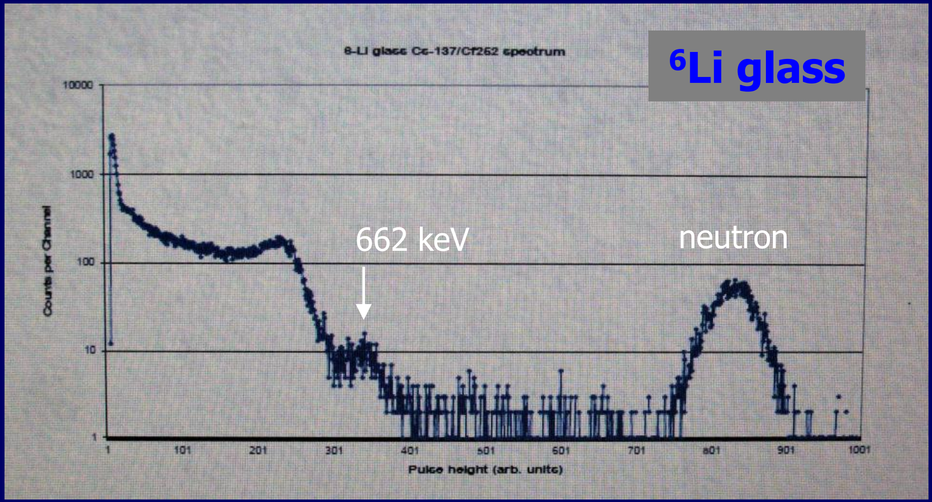
BaF2 + SiPM ?
No efficiency at 200 nm!

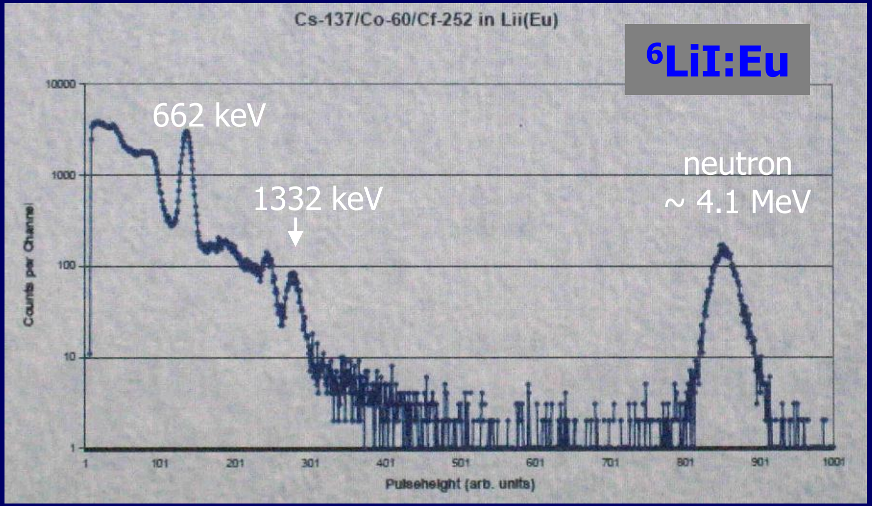
Thermal neutron detection

capture reactions of ^{6}Li

$$n + {}^6\mathrm{Li}\ (7.5\%^*) \rightarrow {}^3\mathrm{H} + {}^4\mathrm{He} \quad (4.8\ \mathrm{MeV})$$

$$\sigma = 941\ \mathrm{barn}\ @\ 1.8\ \text{Å}$$





$\alpha/\beta = 0.35$

$\alpha/\beta = 0.87$

By courtesy of Paul Schotanus,
Scionix

By courtesy of Paul Schotanus, Scionix
A.Syntfeld *et al*, IEEE Trans Nucl Sci
vol 52, no 6, 3151-3156, 2005

Potential thermal-neutron scintillators

Elpasolites $Cs_2LiYCl_6:Ce^{3+}$ (CLYC)

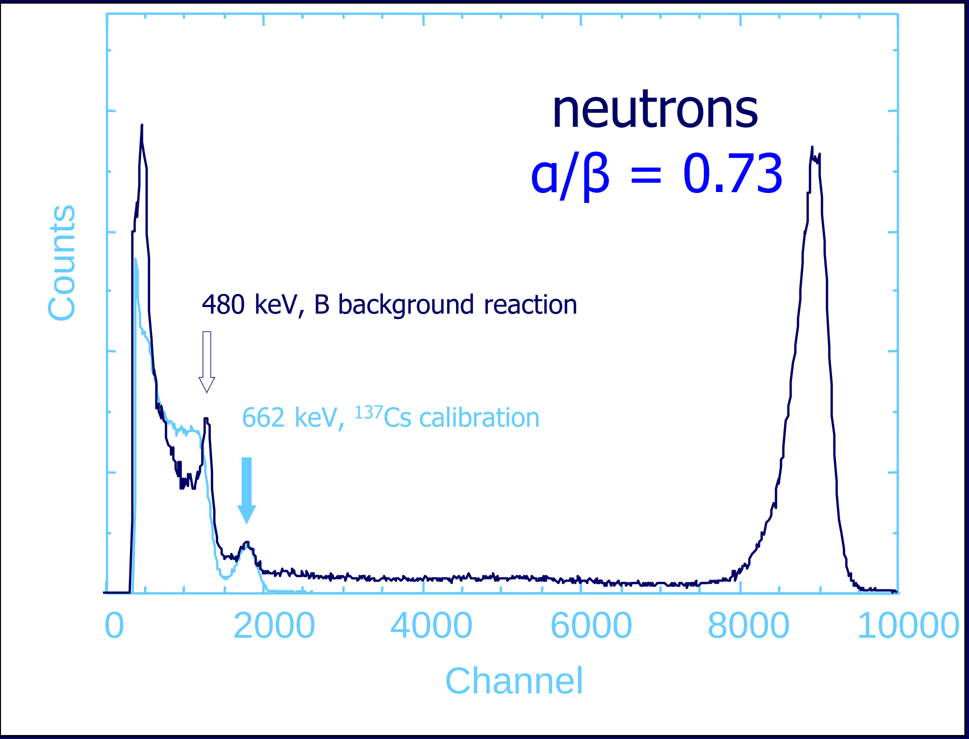
Gamma response 1999

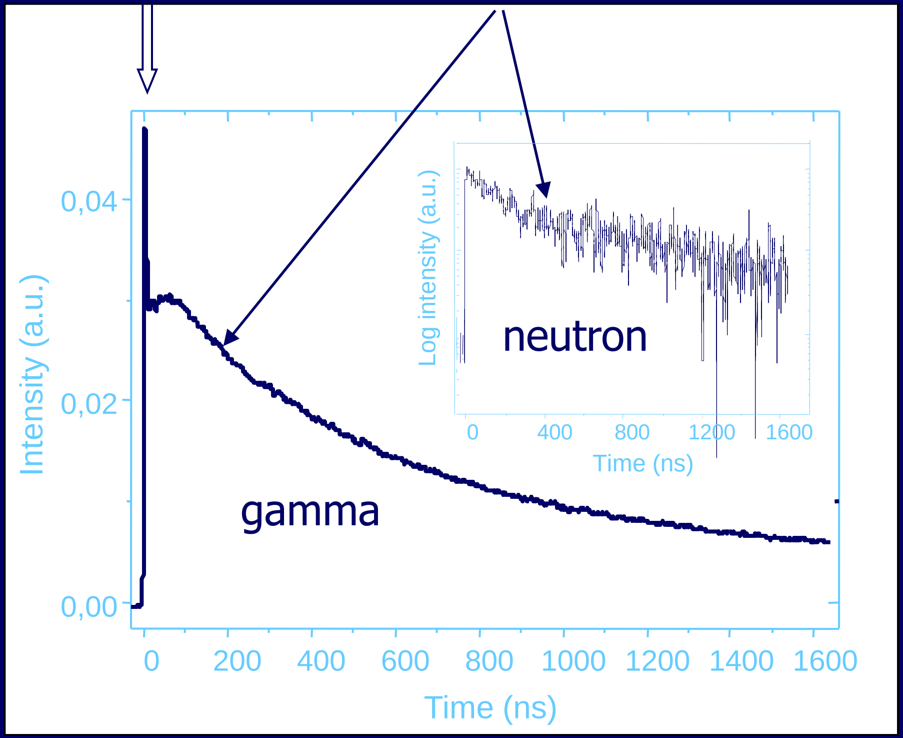
Thermal neutron response 2004

Pulse height & pulse shape n/γ discrimination

CVL ~ 3 ns

Slow response ~ 1 µs





C.M. Combes *et al*, *J. Lumin.*, vol. 82, pp. 299-305, 1999

A. Bessiere *et al*, *IEEE Trans. Nucl. Sci.*, vol. 51, no. 5, pp. 2970-2972, 2004

Continuing struggle to develop the

Better light sensor

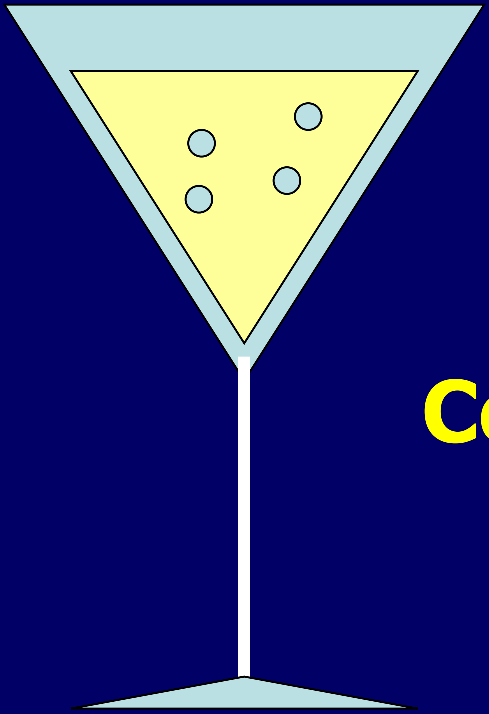
matching the

Better scintillator

and vice versa

Many challenges for the younger generations!

Thank you



Congratulations!