Inorganic Scintillators for the detection of ionizing radiation

P. Dorenbos

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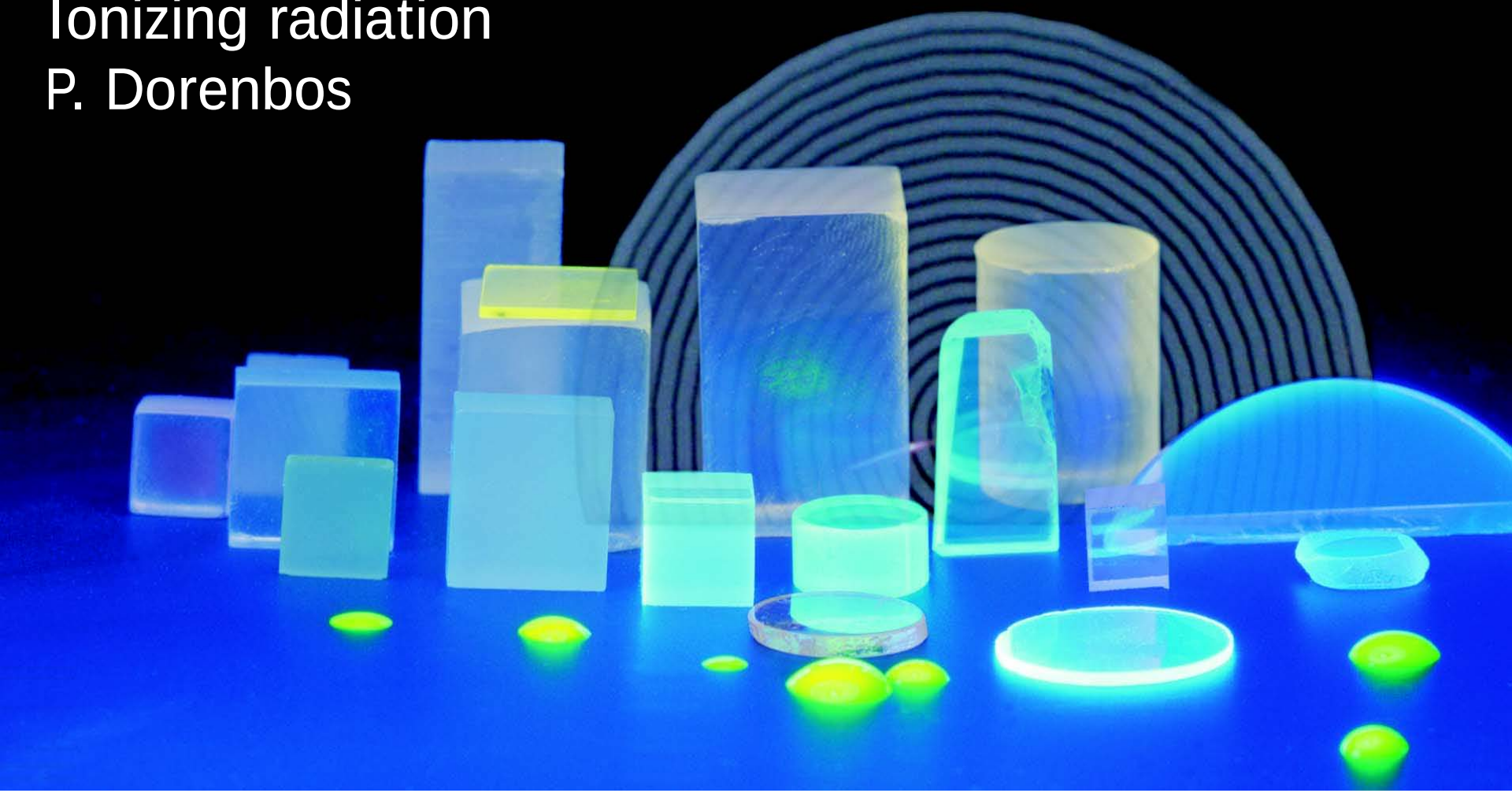
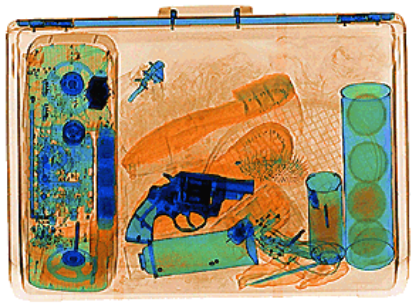
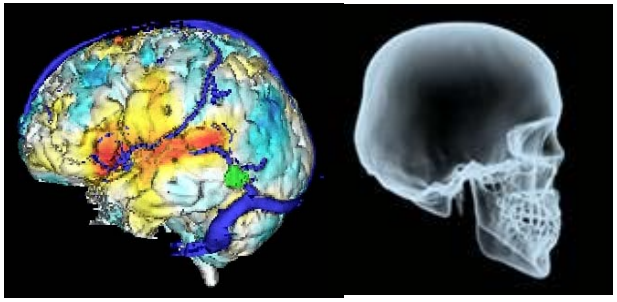
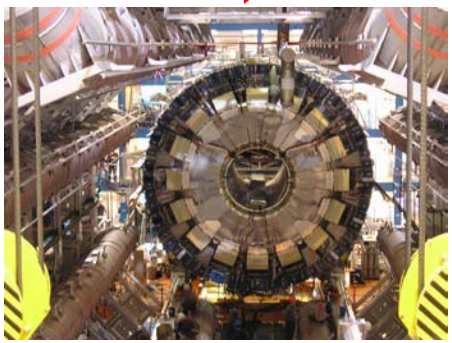


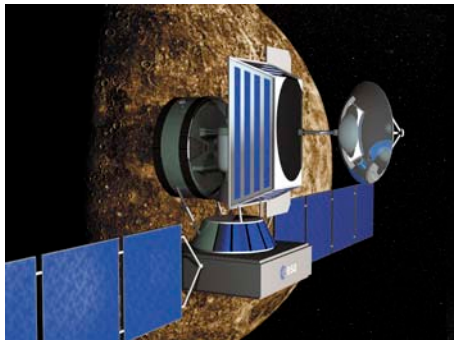


Scintillator applications

- security
- medical diagnostics
- industry + science
- space explorations
- high energy physics



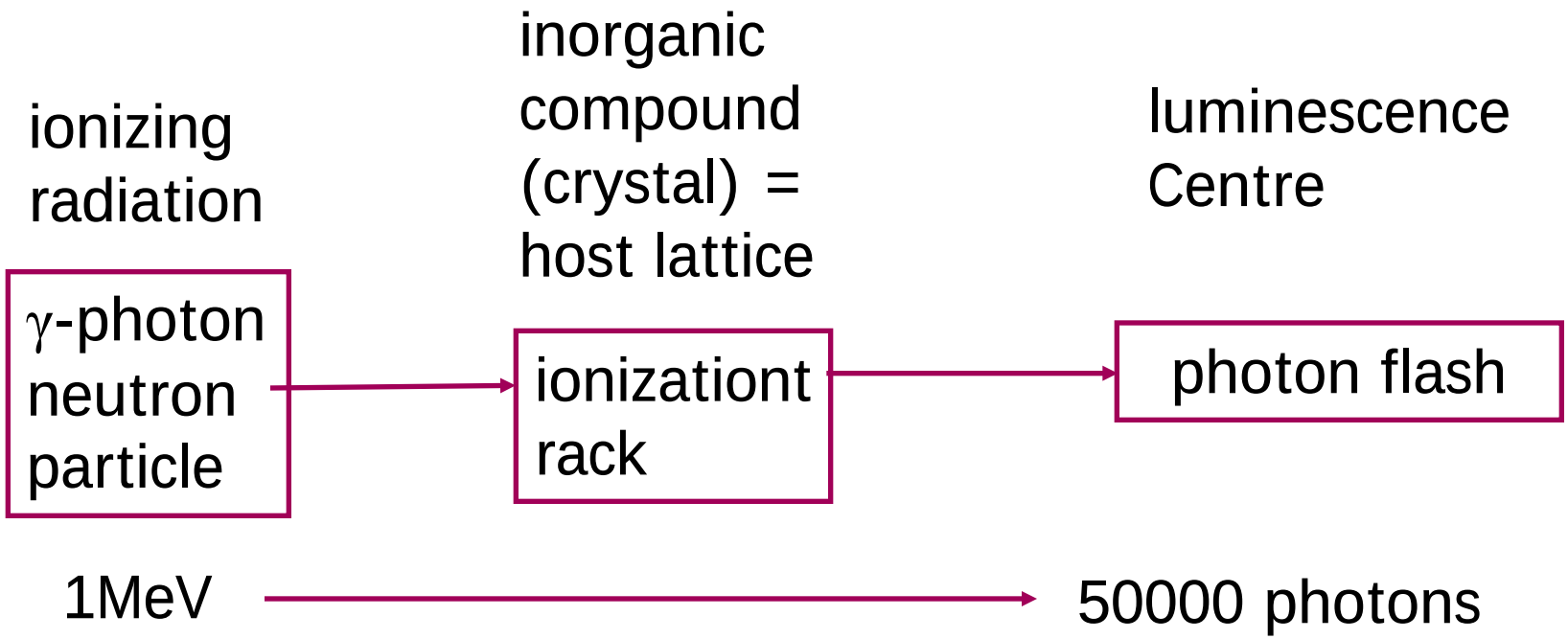




Topics

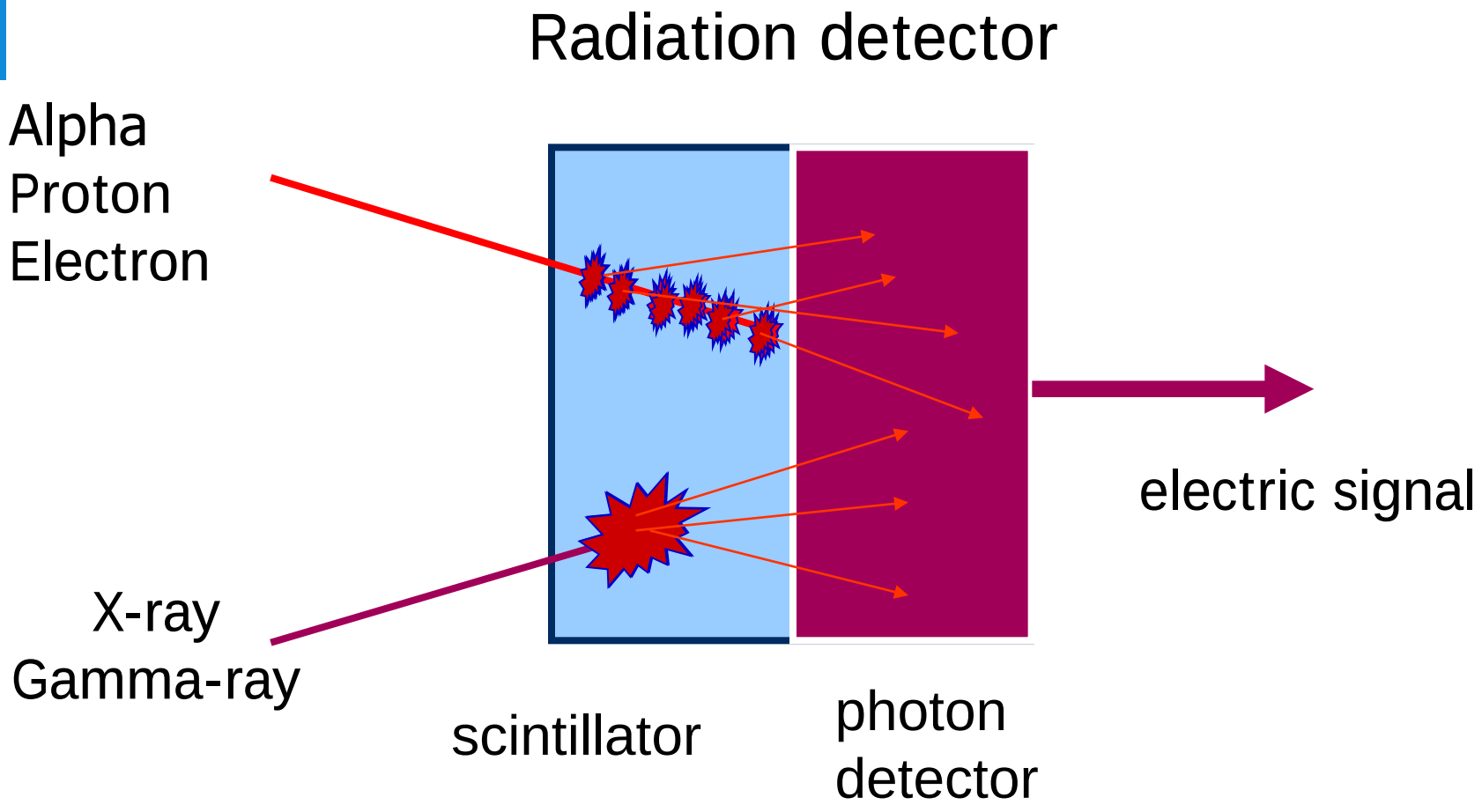
- How it works?
  - The light output — How many photons are emitted?
  - The luminescence — What is color and speed of emission?
  - The scintillation mechanism — What happens on an atomic scale?
- How it performs?
  - Energy resolution — What is the status and are the limits?
  - Time resolution — What is the status and are the limits?
- How it is used?
  - Current developments — What are the hot topics today?

The scintillation process



in between a lot is happening!

Schematic of a radiation detector



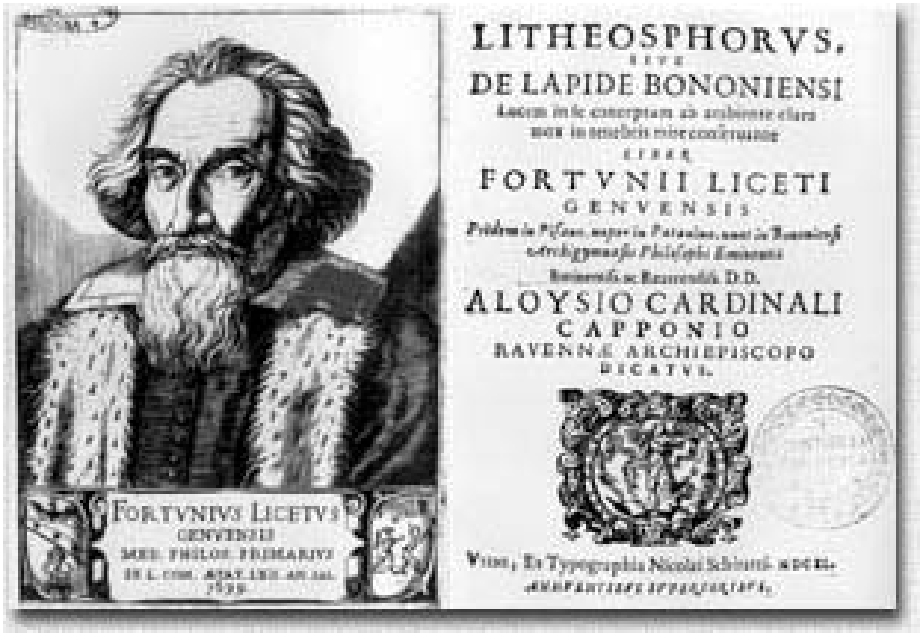
Scintillators

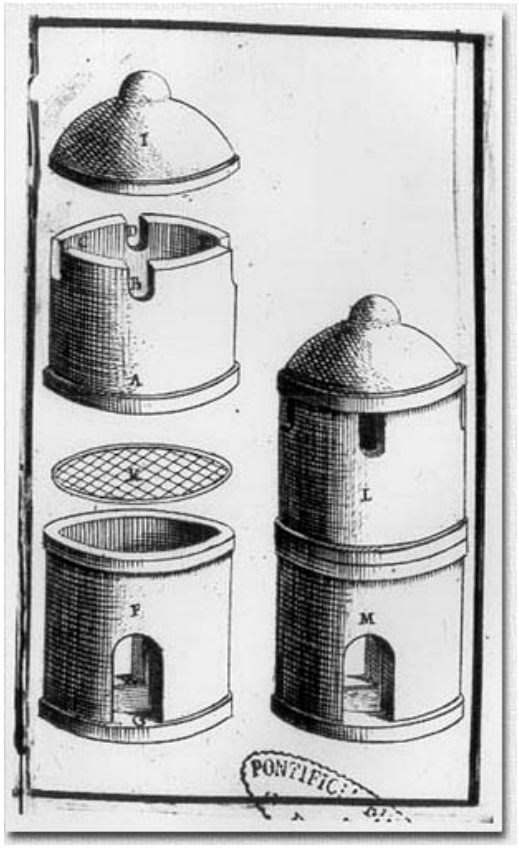
inorganic crystals/powder
organic plastics/crystals
glass
liquid
gas

Light sensors

Human eye
Photomultiplier tubes
silicon photo diodes
Si-PMTs
CCDs
gas-filled detectors

The Bolognian stone




I
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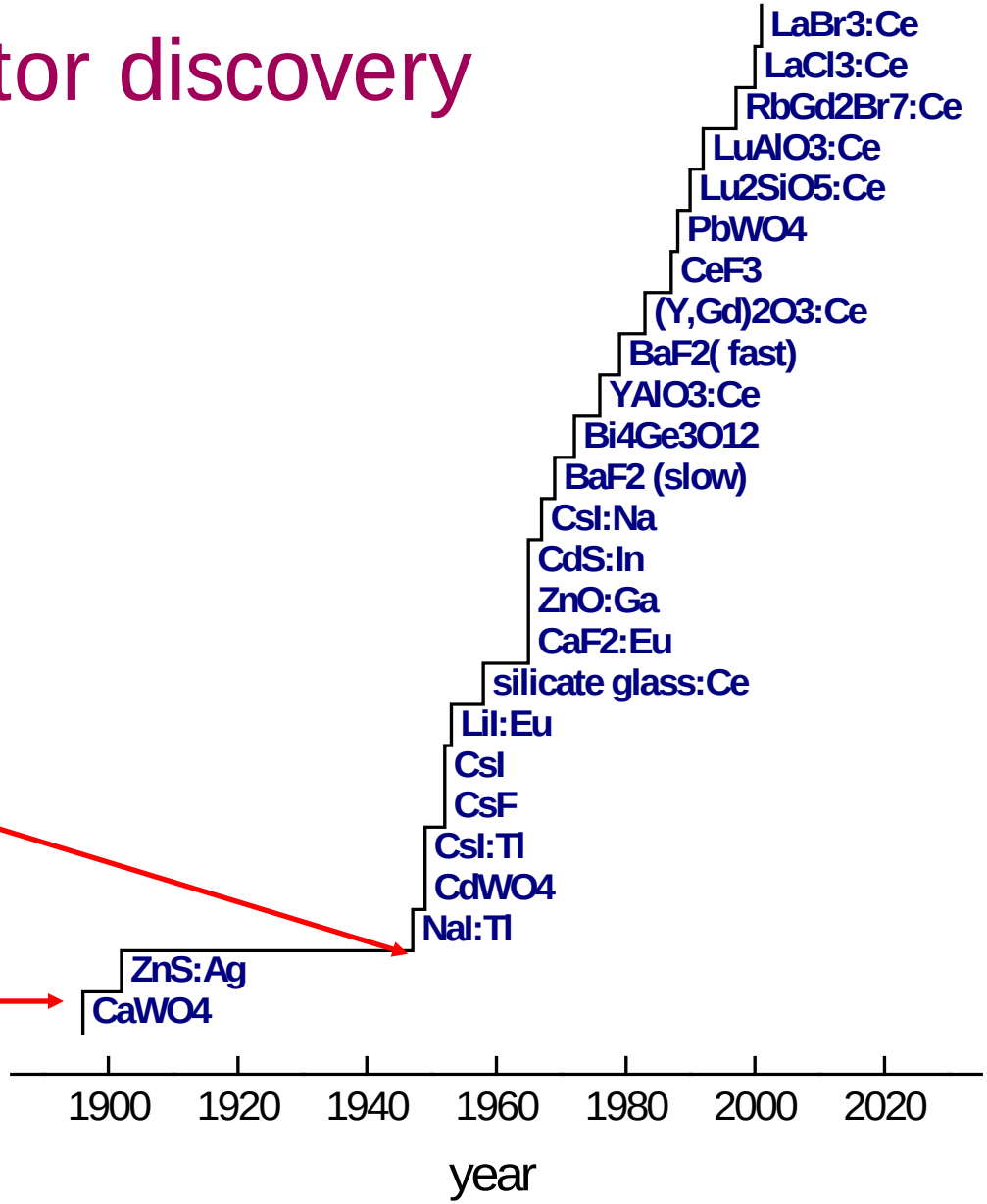
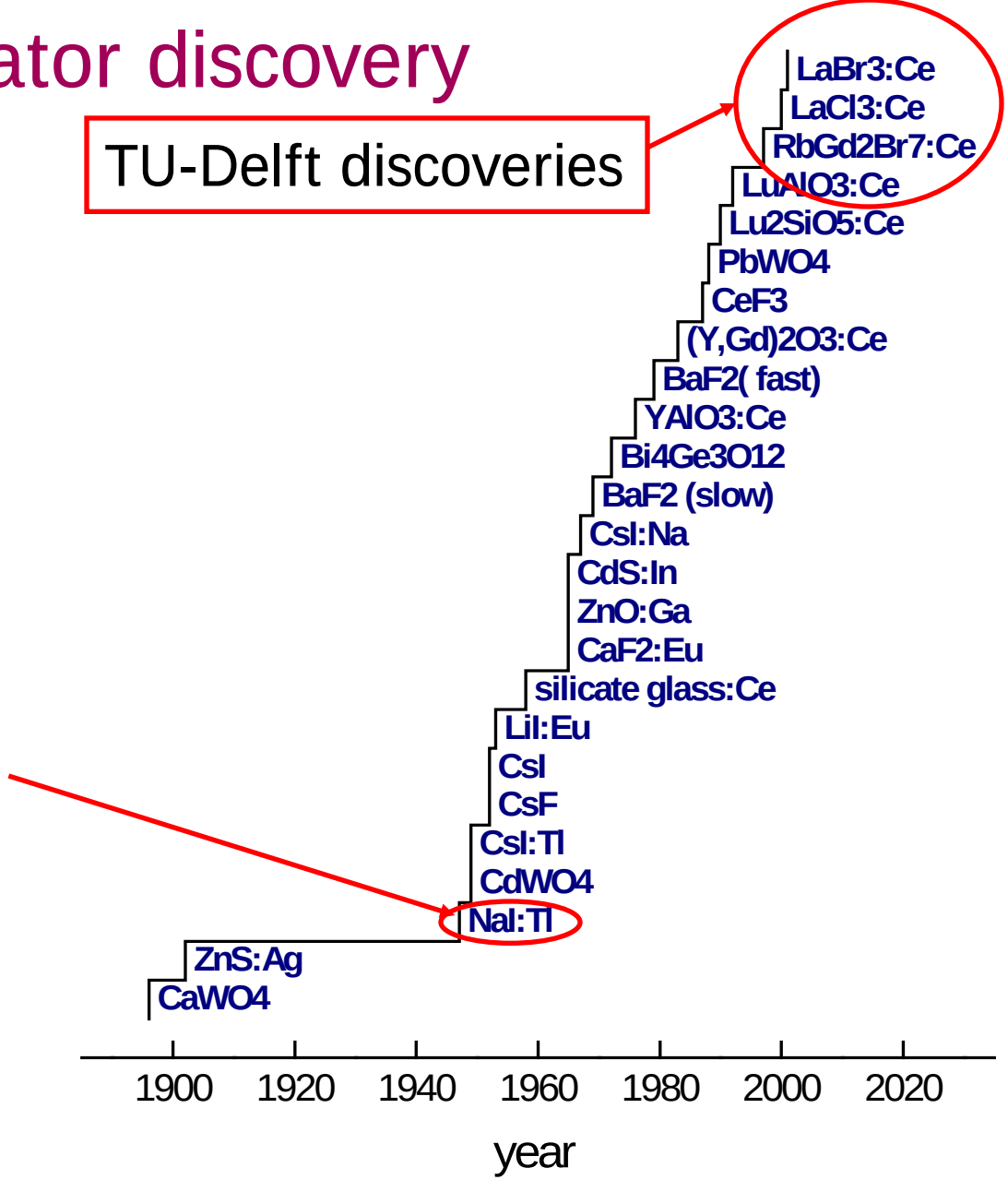
History of scintillator discovery

Invention of Photomultiplier tube

Human eye



History of scintillator discovery



Main drivers of scintillation research

- Homeland security
  - High resolution scintillators for radioisotope identification
  - Shortage of ^{3}He→thermal neutron scintillators
- Space explorations
  - High resolution, low intrinsic count rate scintillators
- Medical diagnostics
  - TOF-PET very fast risetime scintillators
  - SPECT
- High energy physics
  - High density and high radiation hardness
  - Large volumes at low cost

Important gamma ray scintillator parameters

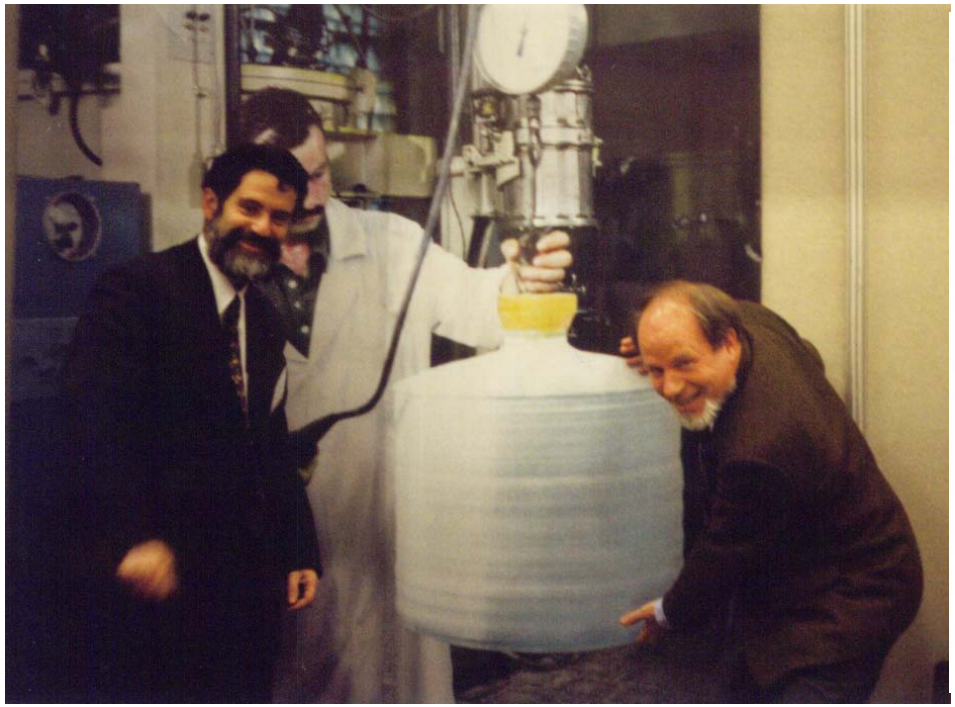
- High light output — Y (photons/MeV)
- fast scintillation speed — τ_s (ns)
- low energy resolution — R_{FWHM} (%)
- high density for γ detection — ρ (g/cm^3)
- large size of crystal — 10-100-1000 cm^3
- low cost per cm^3
- low afterglow (low phosphorescence)
- low background count rate (low intrinsic activity)
  - absence of radioactive isotopes

Importance depends on application

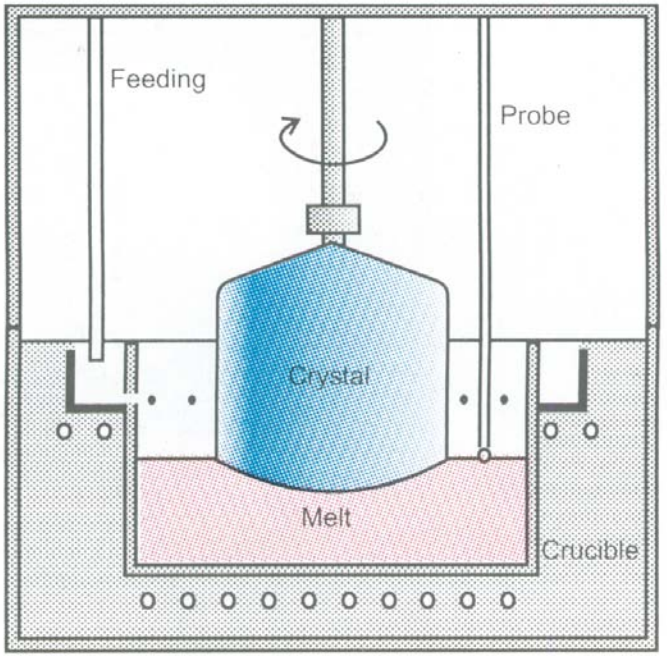
Properties of inorganic scintillators

scintillator	ρ	n	τ	λ_{max}	N_{phot} / MeV
NaI (*at 80* K)	3.67	1.75	60	303	76000
NaI(Tl)	3.67	1.75	230	415	38000
CsI(Tl)	4.51	1.75	3340	540	65000
BaF_2	4.89	1.5	630	310	9500
(*valence* e^- *to core*)			0.6	220	1400
$Bi_4Ge_3O_{12}$ (BGO)	7.13	2.15	300	480	8200
$PbWO_4$	8.28	2.20	10	470	100
Lu_2SiO_5:Ce (LSO)	7.4	1.8	47	420	25000
$YAlO_3$:Ce (YAP)	5.37	1.95	27	370	18000
$LaCl_3$:Ce	3.7	1.8	35	350	50000
$LaBr_3$:Ce	5.1	2.1	17	380	70000

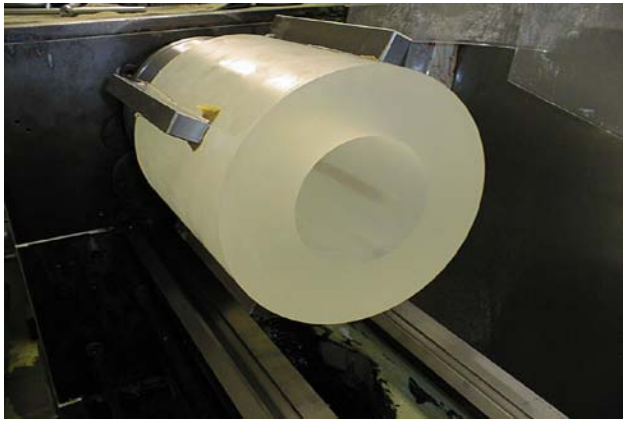
The growth of NaI:Tl scintillation crystals



dr. A. Gektin and prof. C.W.E. van Eijk with a NaI:Tl ingot



Examples or bare scintillation crystals



NaI(Tl)

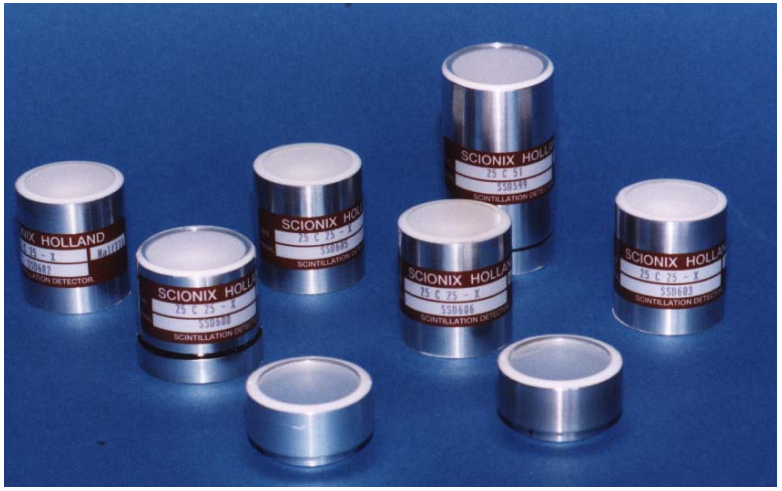
CsI(Tl)

$CdWO_4$

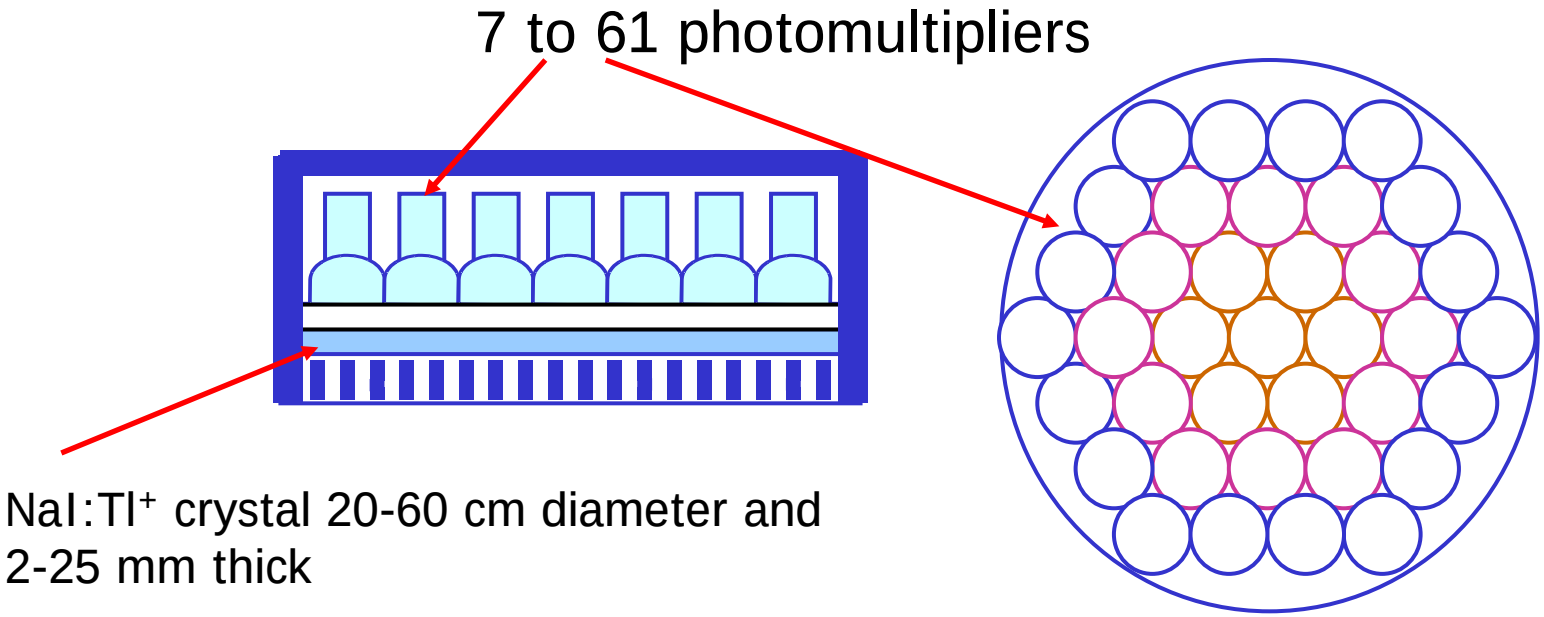
BGO

Examples of packed crystals

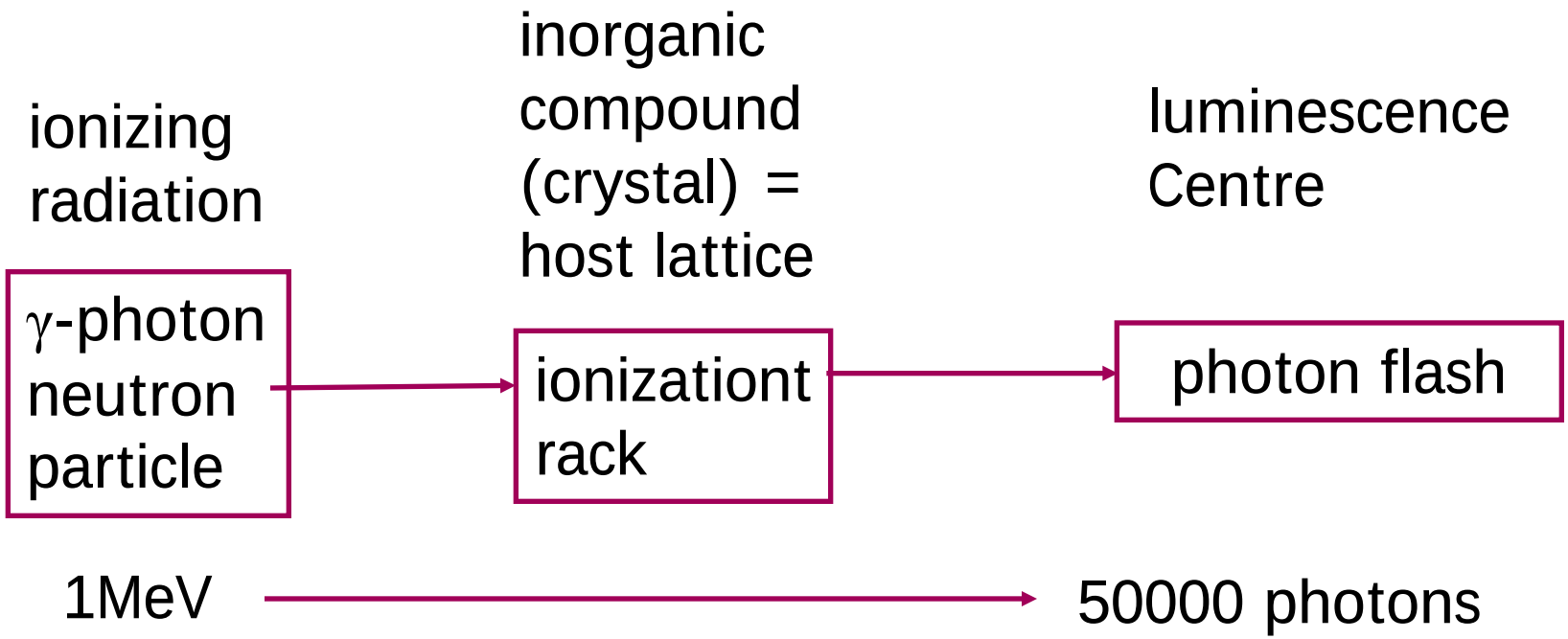
Gamma camera crystal



NaI(Tl) crystals



The scintillation process

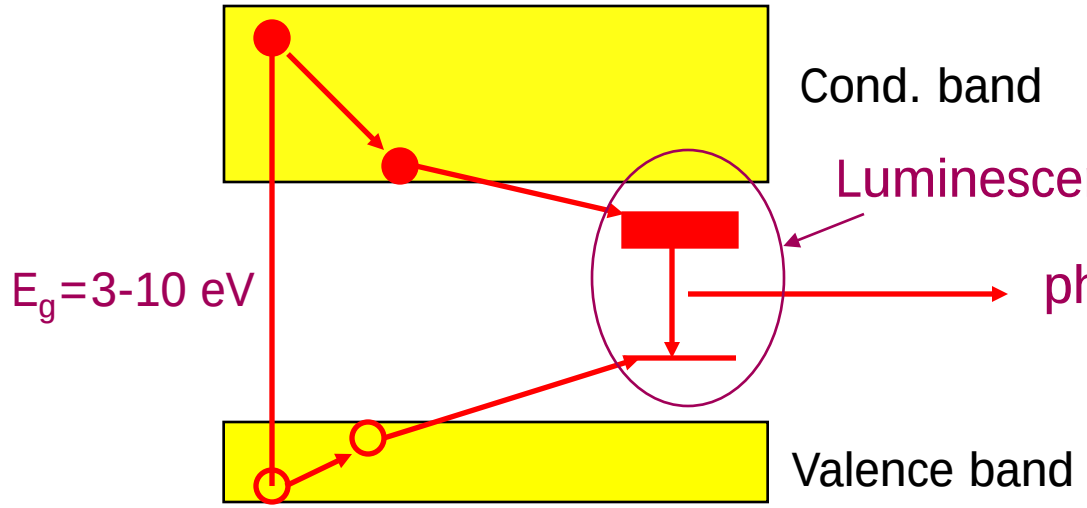


What is happening in between? A lot!

The scintillation process

Three phases

1. The interaction phase + thermalization phase (ps)
2. The charge carrier and energy migration phase (ps-ns-ms)
3. The luminescence phase (ns-µs)

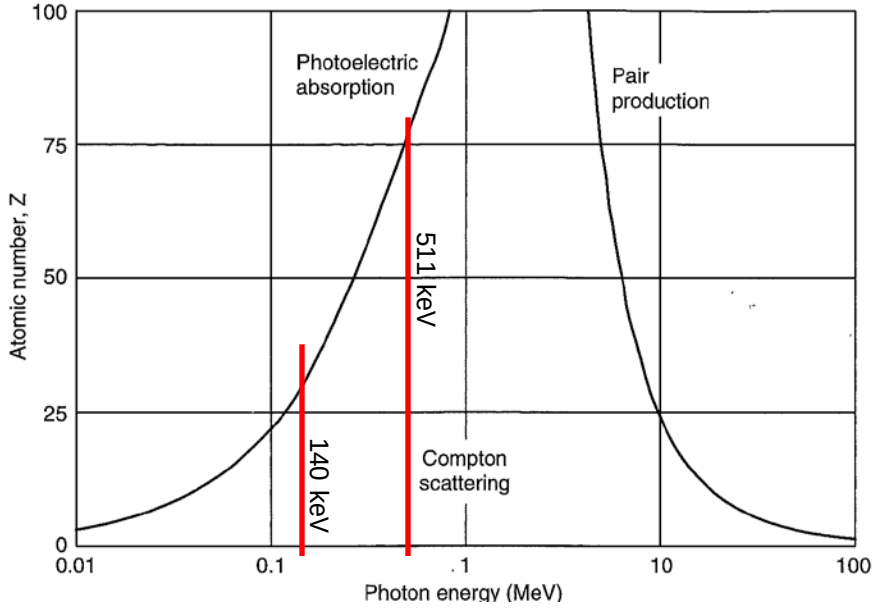


VIS	**400 - 800 nm**	**3.1 - 1.6 eV**
UV	**180 - 400 nm**	**6.9 - 3.1 eV**
VUV	**< 180 nm**	**> 6.9 eV**

Phase I: The interaction phase

- Gamma ray interaction → energetic electron
  - The photo-electric effect
  - The Compton scattering
  - Pair production

Z_{eff}^{3-4} (dominant <100 keV)
ρ (dominant around 1MeV)
E_γ >1.02 MeV



Z(I)	=53
Z(Cs)	=55
Z(La)	=57
Z(Gd)	=64
Z(Lu)	=71
Z(Pb)	=82
Z(Bi)	=83

The ionization track in scintillators

- 10 keV – 10 MeV X-ray or gamma-rays produce fast primary electrons

The Bethe formula

$$-\frac{dE}{dx} = \frac{4\pi e^4 z^2}{m_0 v^2} NZ \left[\ln \frac{2m_0 v^2}{I} - \ln\left(1 - \frac{v^2}{c^2}\right) - \frac{v^2}{c^2} \right]$$

ez = particle charge

v = particle velocity

N,Z = number density and atomic number atoms

I = average ionization potential atoms

Smaller energy → smaller v→ higher dE/dx→more ionizations/volume

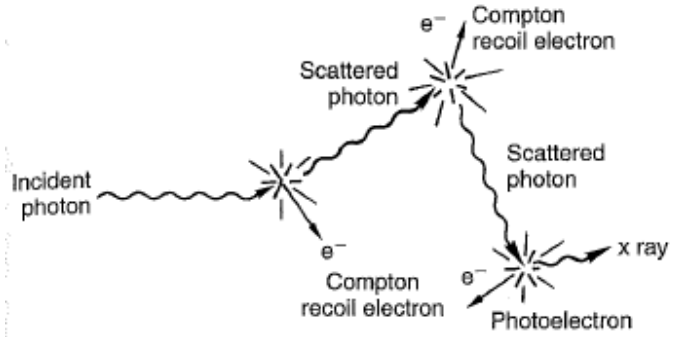


Figure 6–15. Multiple interactions of a photon passing through matter. Energy is transferred to electrons in a sequence of photon-energy degrading interactions.

- Multiple interactions
  - One scintillation event can produce separate ionization tracks in the scintillator
  - Everything takes place in the 1-10 ps time scale
  - Separate flashes add to one scintillation flash
- High energy physics GeV-TeV→ shower of events

Energy needed for electron-hole pair generation

Semiconductor	ρ (g/cm^3)	Z	E_g (eV)	E_{ion} (e
Si	2.33	14	1.12	3.6
Ge	5.33	32	0.67	2.9
GaSe	4.55	31, 34	2.03	4.5
InP	4.78	49, 15	1.30	4.2
CdS	4.84	48, 16	2.60	7.3
GaAs	5.32	31, 33	1.43	4.3
InSb	5.77	49, 51	0.20	0.6
CdSe	5.80	48, 34	1.73	5.5
CdTe	6.20	48, 52	1.44	4.7
PbI_2	6.20	82, 53	2.55	7.7
HgI_2	6.40	80, 53	2.13	4.2
TlBr	7.56	81, 35	2.68	6.5

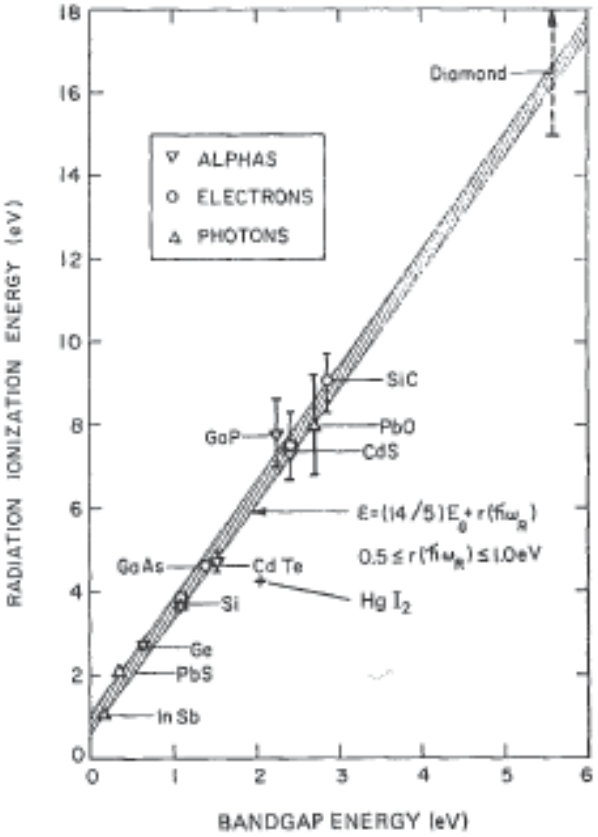
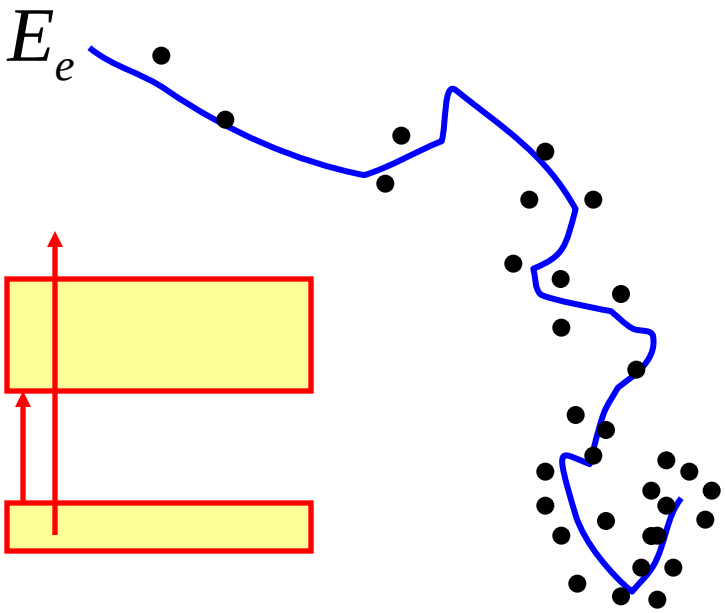


Figure 13.21 The average energy required to form one electron–hole pair (ϵ) versus bandgap energy for a number of semiconductor materials. (From Klein.[45])

Linear relationship between E_{ion} and E_g (In figure β=2.8)

The ionization track and electron-hole creation



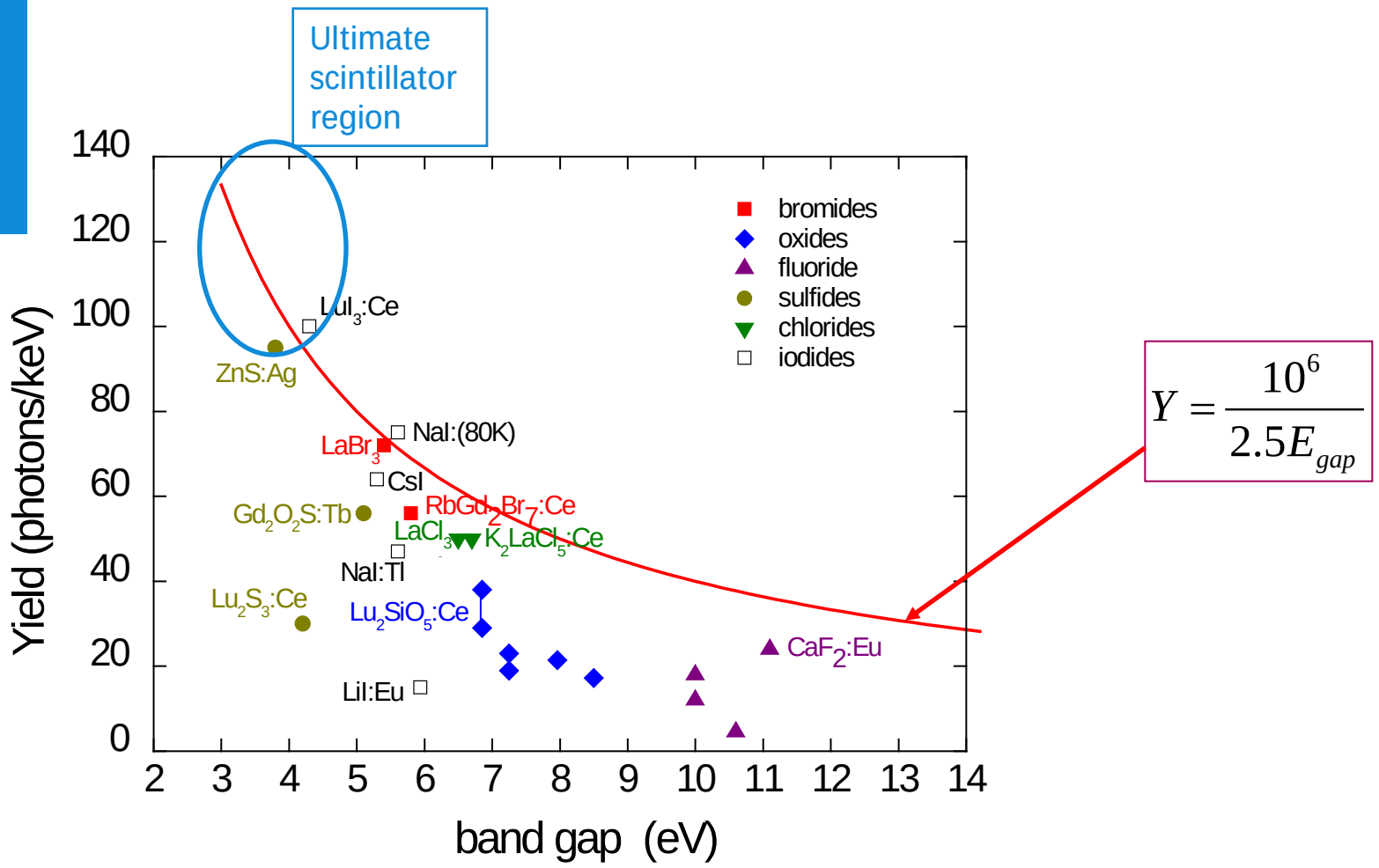
$$N_{eh} = \frac{E_\gamma}{E_{ion}} = \frac{E_\gamma}{\beta E_g}$$

$$\left.\begin{aligned} \beta &\approx 2.5 \\ E_g &= 8 \text{ eV} \end{aligned}\right\} \Rightarrow N_{eh} = 50.000 \text{ /MeV}$$

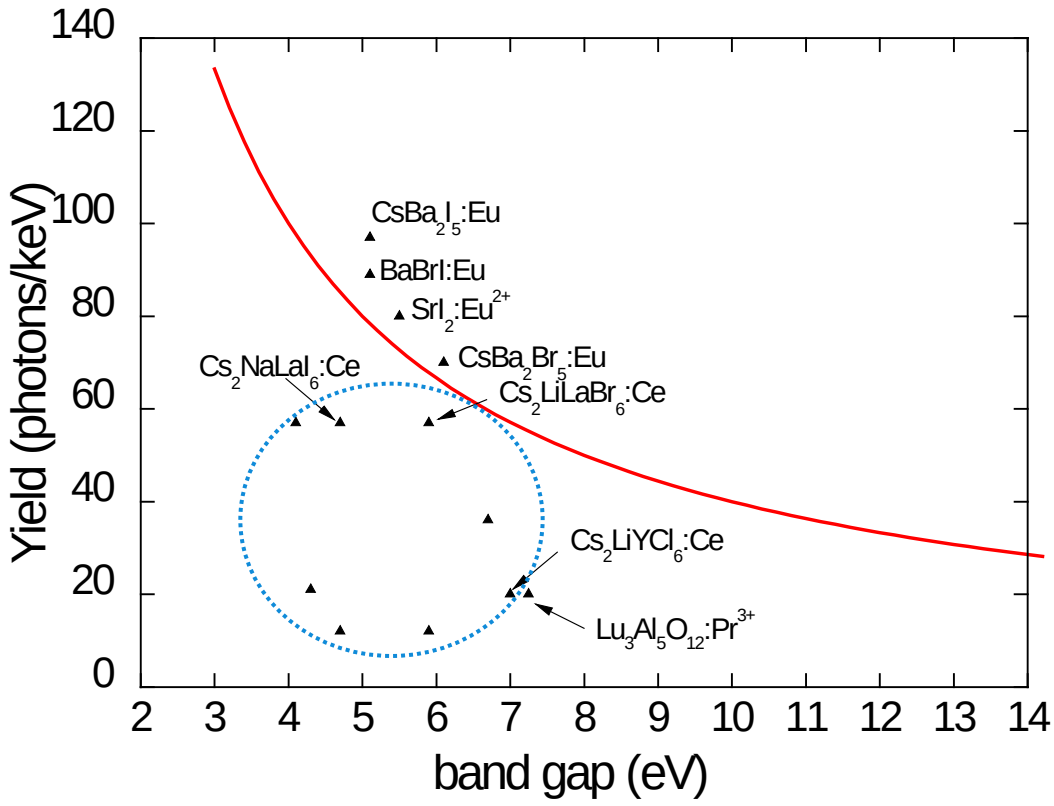
$$\mathrm{N_{ph}} = SQN_{eh}$$

- S is the transfer efficiency from track to luminescence center
- Q is quantum efficiency of luminescence center

The ideal scintillator has S=Q=1



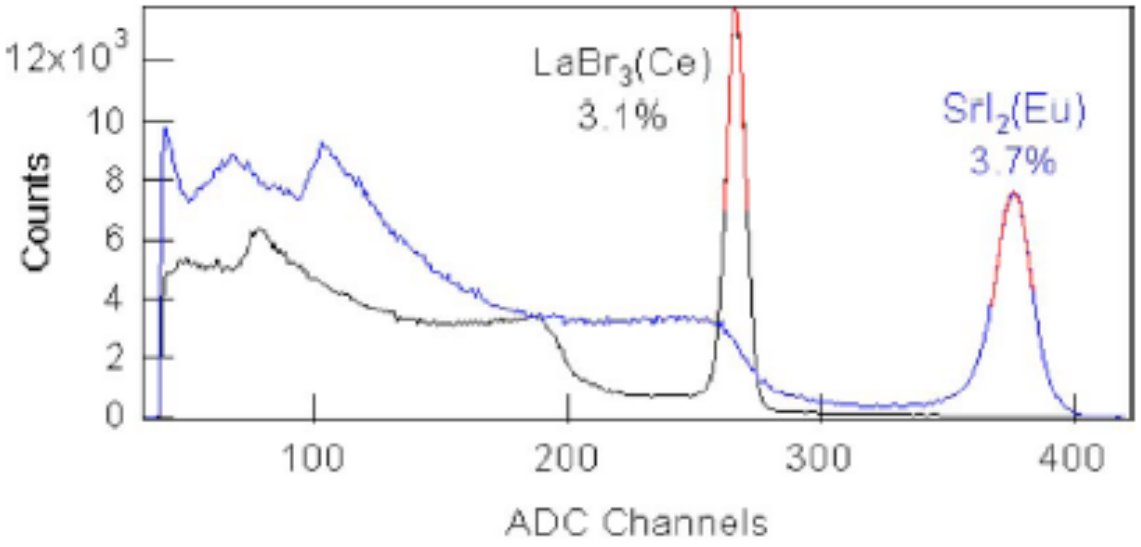
Latest developments in scintillation research



Latest materials developments

- Elpasolites (Radiation Monitoring Devices, dr. K. Shah)
- Eu^{2+} doped halides (University of Berkeley, prof. Derenzo, prof. Moses)
- $Lu_3Al_5O_{12}:Pr^{3+}$ (Tohuku Univ, prof. Yoshikawa, Inst. of Phys. Prague, dr. M. Nikl)

Current developments SrI_2:Eu



Cherepy et al. Appl. Phys. Lett. 92 (2008) 083508

Current developments $Ba_2CsI_5:Eu^{2+}$

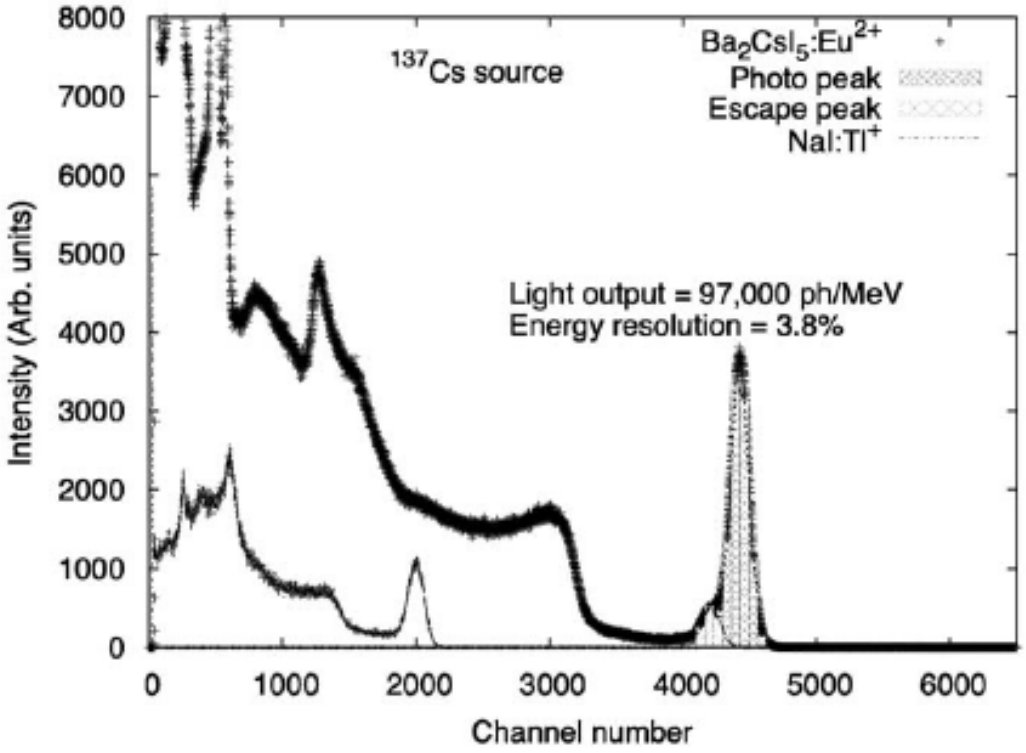


Fig. 3. Pulse height spectra of $Ba_2CsI_5:Eu^{2+}$ and NaI:Tl measured under ^{137}Cs gamma-ray excitation.

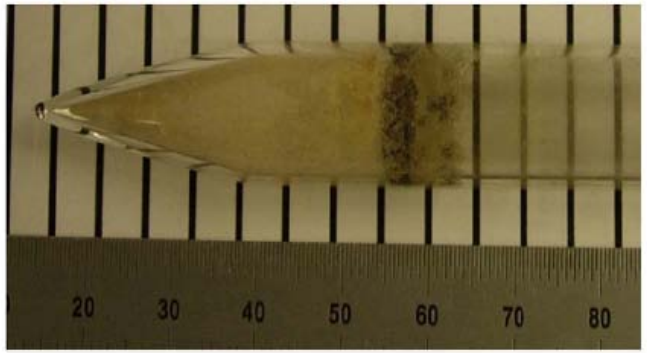


Fig. 1. A 1 cm in diameter $Ba_2CsI_5:Eu^{2+}$ shown as-grown in a sealed quartz ampoule.

Bourret-Courchesne et al. Nucl. Instr. Meth. A612 (2009) 138

Current developments $BaBrI_3:Eu^{2+}$

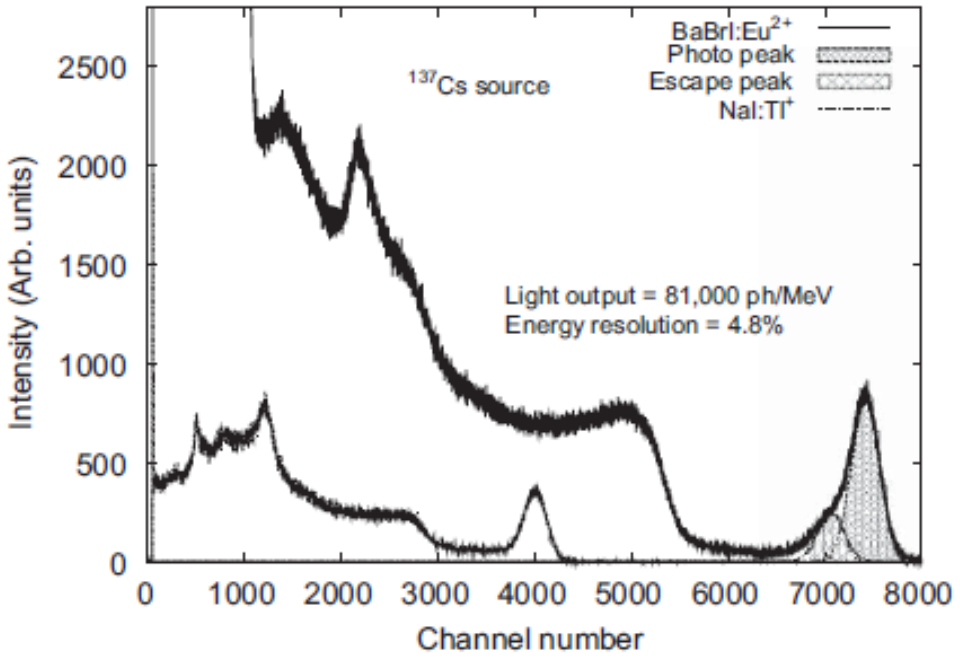


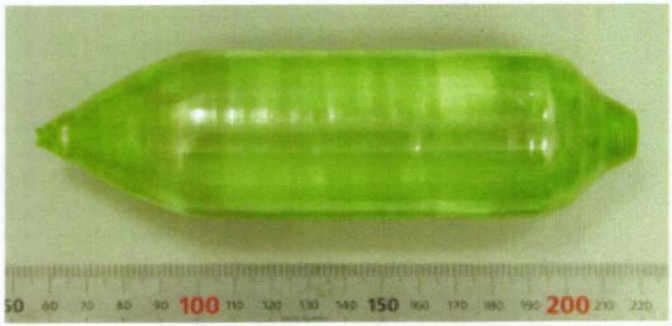
Fig. 2. Pulse height spectra of $BaBrI:Eu^{2+}$ and NaI:Tl measured under ^{137}Cs gamma-ray excitation.

Fig. 1. 1 cm in diameter $BaBrI:Eu^{2+}$ shown as-grown in a sealed quartz ampoule.

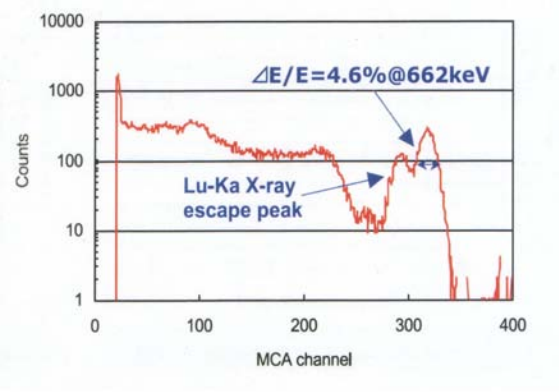
Bourret-Courchesne et al. Nucl. Instr. Meth. A613 (2010) 95

Recently more materials were discovered. All have their pros and cons.

Scintillators	Pr:LuAG ($Lu_3Al_5O_{12}$)
Density (g/cm^3)	6.7
Light Yield (BGO=100)	330
Decay Time (ns)	<25
Peak emission (nm)	310
Energy Resolution (%@662keV)	5
Hygroscopicity	No
Cleavage	No
Melting Point (℃)	1970



2 inch ϕ L uAG single crystal grown by Cz method



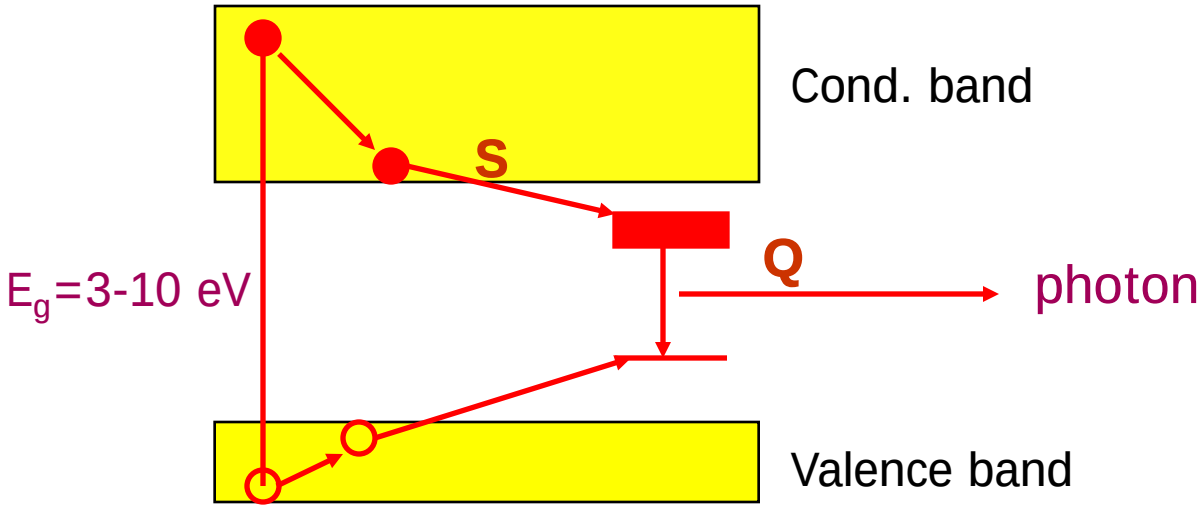
Fast and good energy resolution

But high intrinsic background (Lu)

The scintillation process; phase III

Three phases

1. The interaction phase + thermalization (ps)
2. The charge carrier and energy migration phase (ns-ms)
3. The luminescence phase (ns-μs)

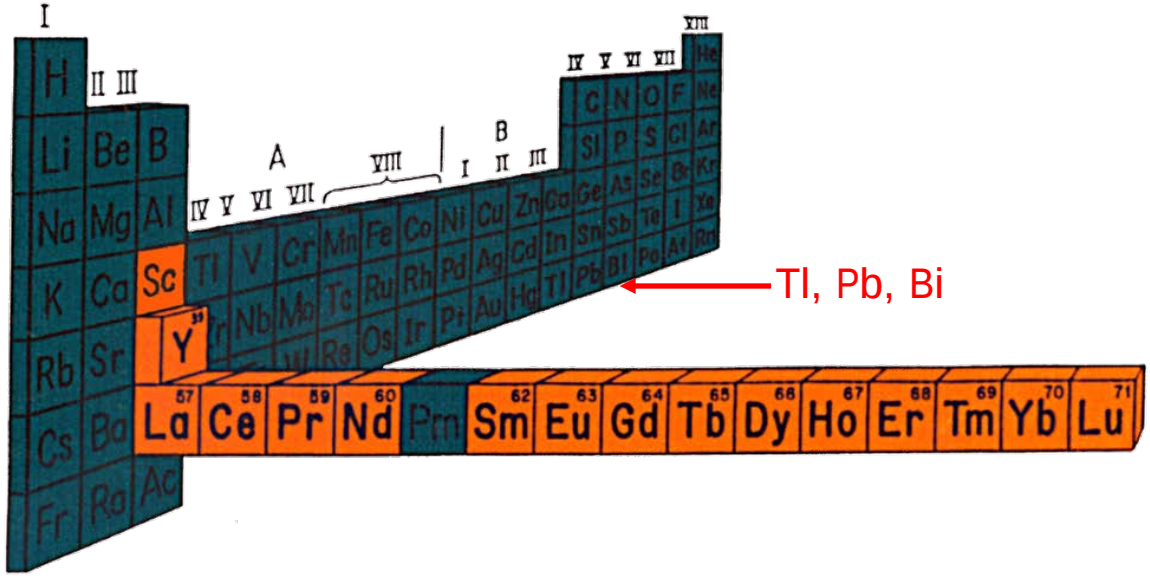


Ideal scintillator
S=Q=1

The luminescence phase

The most popular activator ions

- The $6s^2$-ions Tl^+, Pb^{2+}, Bi^{3+}
  - NaI:Tl, CsI:Tl, $Bi_4Ge_3O_{12}$, $PbWO_4$
- The lanthanide ions Ce^{3+}, Pr^{3+}, Eu^{2+}
  - $YAlO_3$:Ce, $Y_3Al_5O_{12}$:Ce, Lu_2SiO_5:Ce, $LuAlO_3$:Ce, $LaCl_3$:Ce, $LaBr_3$:Ce, LiI:Eu, $Lu_3Al_5O_{12}$:Pr^{3+}



$6s^2$-ions Tl^+, Pb^{2+}, Bi^{3+}

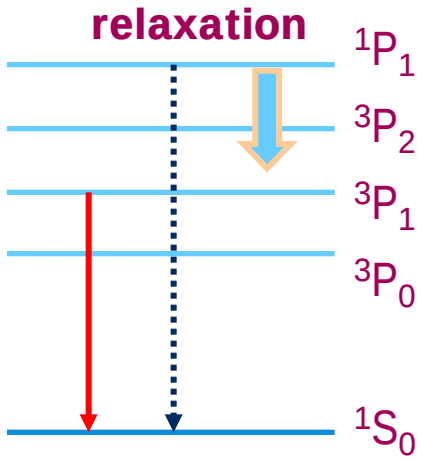
		$^{2S+1}L_J$	
ground state configuration	$6s^2$	$^1S_0$	
excited state configuration	6s6p	$^1P_1$	singlet
		$^3P_2$	
		$^3P_1$	triplet
		$^3P_0$	

Selection rules Allowed transitions

$\Delta J = 0, \pm 1$ (not $J = 0 \rightarrow J' = 0$), $\Delta S = 0$

Singlet $\rightarrow$ singlet allowed, fast

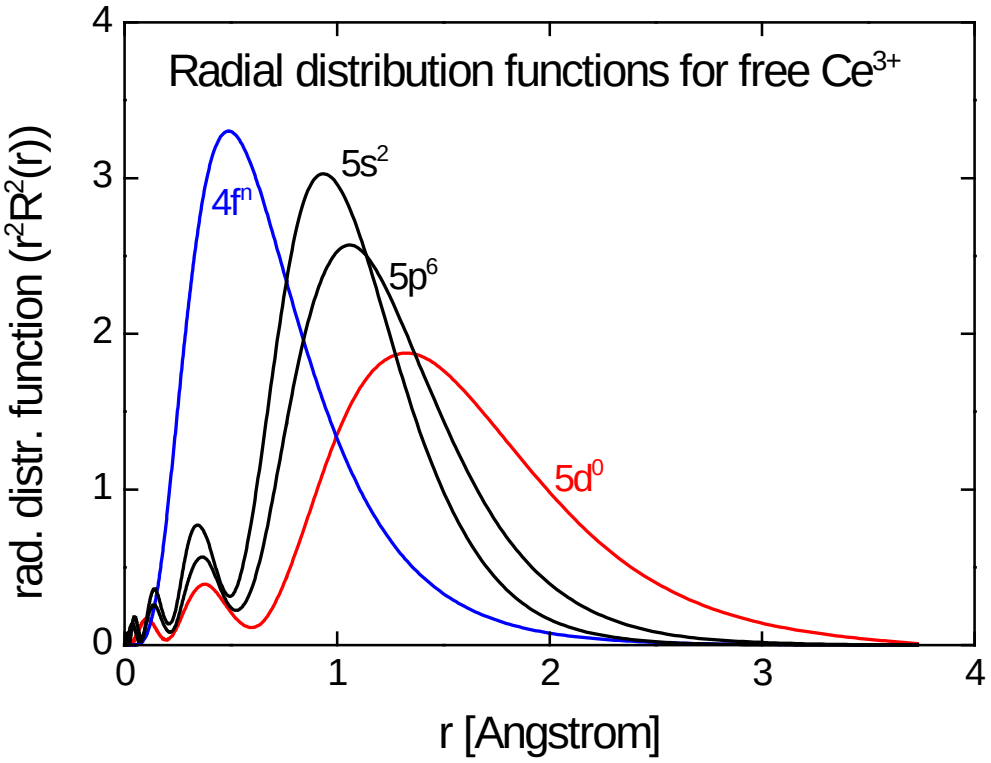
Triplet $\rightarrow$ singlet forbidden, slow



NaI:Tl	τ=230 ns
CsI:Tl	τ=3.3 μs

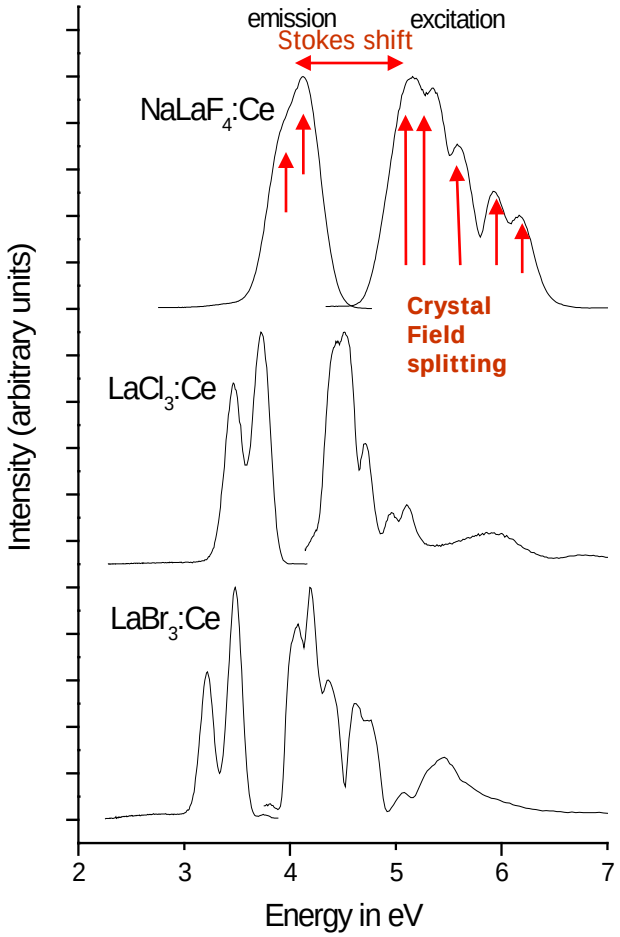
Lanthanide luminescence

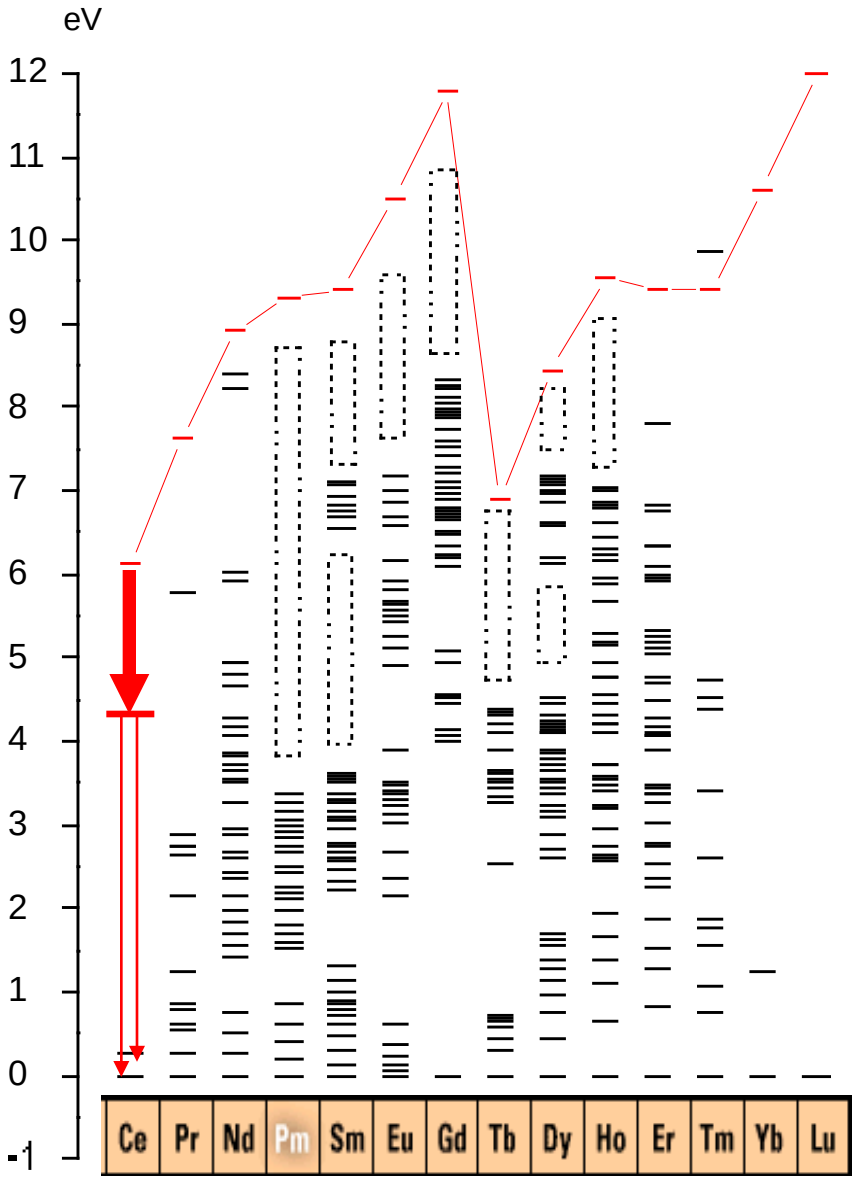
La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu



- electron configuration $[Xe]4f^n5d^0$
- 4f electrons are shielded from the crystalline field. Level energies independent on host lattice
- 5d electron strong interaction with crystal field. Level energies depend on host crystal

The Dieke diagram of free ion trivalent lanthanide $4f^n$ and 5d levels

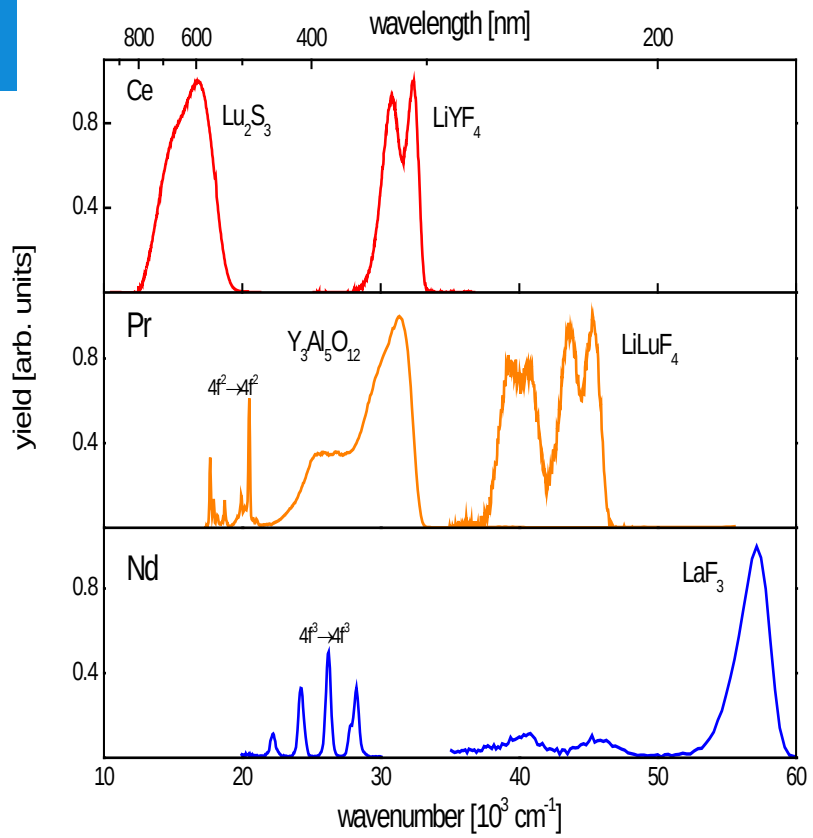


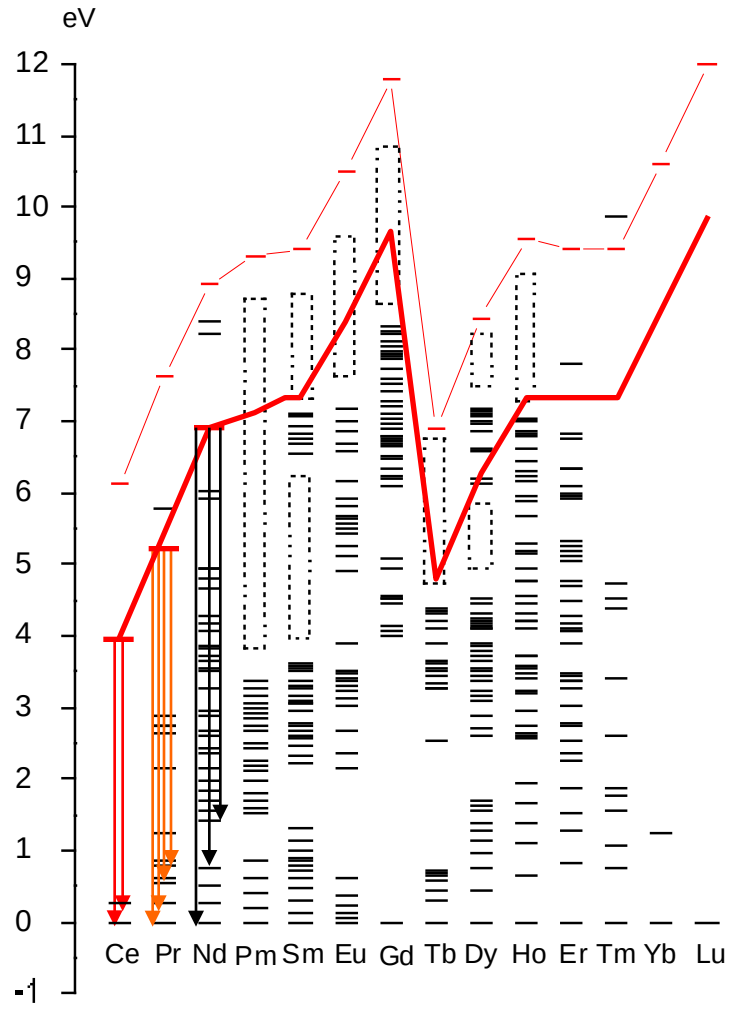


The emission wavelength

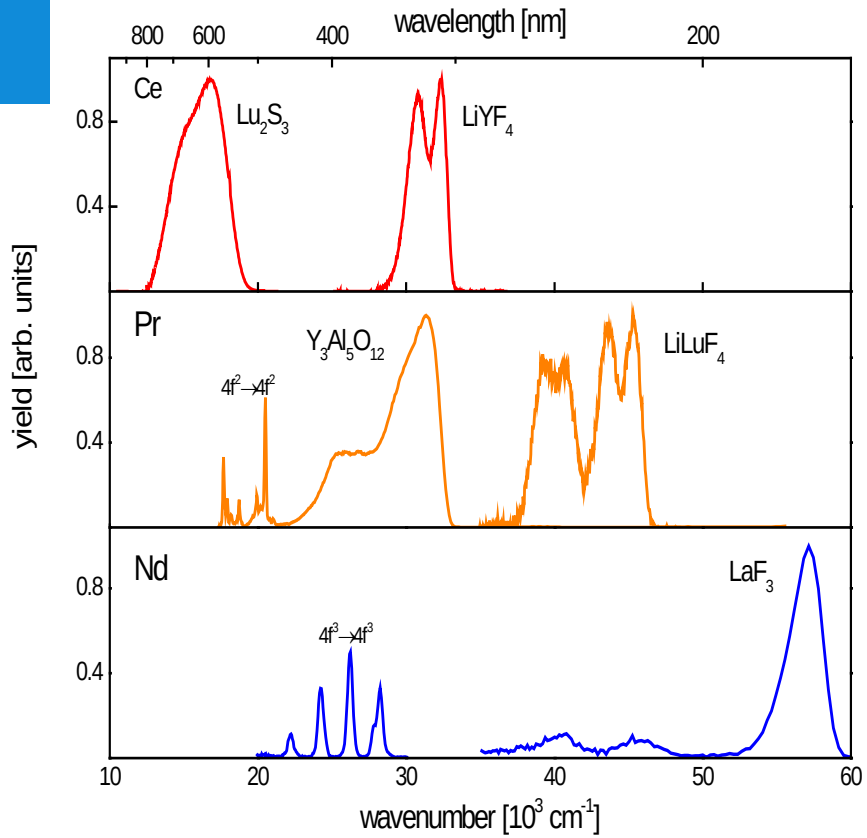
- Electrons either in the ground state or excited state of a luminescence center interact with the surrounding atoms in a compound
  - 4f electrons have negligible interaction
  - 5d electrons (also 6s and 6p electrons of $6s^2$-ions) have strong interaction
- Interaction depends on
  - Crystal structure,
    - coordination number, bond lengths etc.
  - The chemical properties of the anions
    - F^-, Cl^-, Br^-, I^- and O^{2-}, S^{2-}
- The emission of Ce^{3+} may vary from 300 nm in fluoride compounds up to 600 nm in sulfide compounds

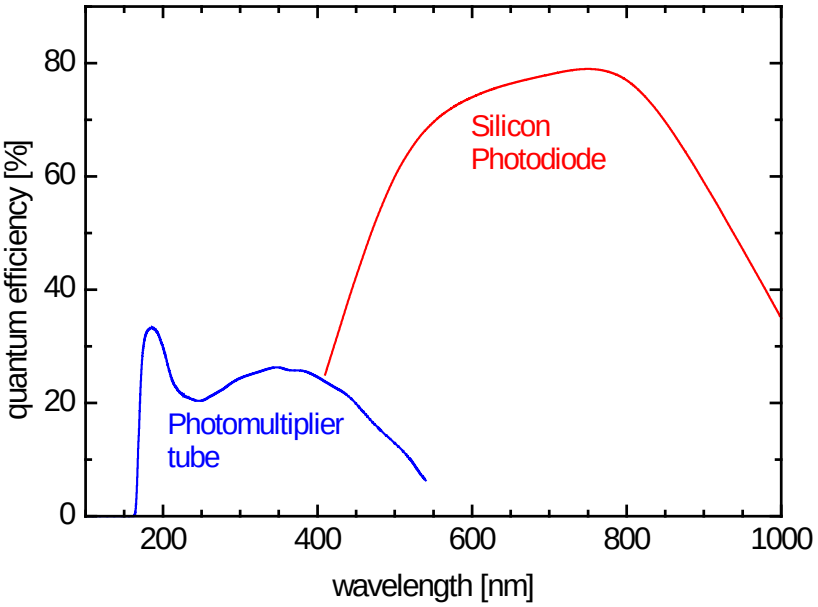
Characteristic emission spectra Ce, Pr, Nd





Emission and quantum efficiency





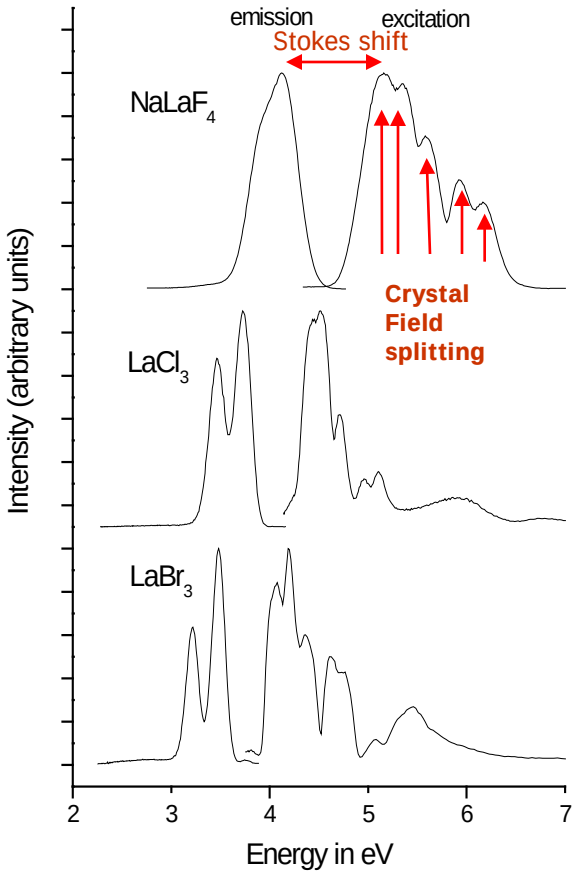
Ce^{3+} doped lanthanide compounds are ideal for gamma ray scintillators

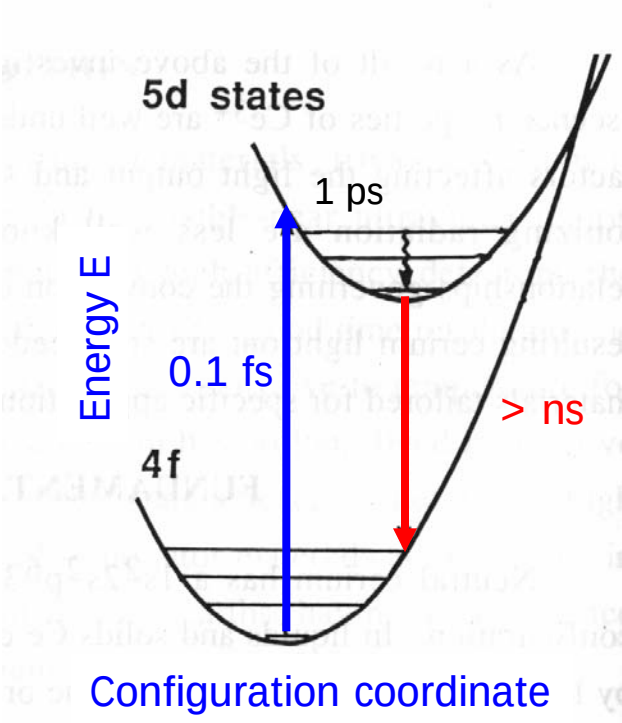
- Fast dipole and spin allowed 5d-4f emission (15-60 ns)
- Emission wavelength matches sensitivity of photomultiplier tubes
- High thermal stability of emission
- Absence of slow 4f-4f emission
- Ce^{3+} is a good hole trap which is regarded as a first important step in the scintillation process
- Lanthanide (La, Gd, Lu) based host lattices are dense providing efficient gamma ray detection materials
  - $LaBr_3$:Ce, Gd_2SiO_5:Ce, Lu_2SiO_5:Ce

Stokes shift and self-absorption

- A scintillator crystal needs to be transparent to its own scintillation light
- A photon emitted by a luminescence center can be re-absorbed by another luminescence center of the same type
- Probability increases with
  - Larger activator concentration
  - Higher temperature (broadening of emission and absorption bands)
  - Larger crystal size
  - Smaller value for the Stokes shift
    - Stokes shift = energy difference between emission and first absorption band of luminescence center

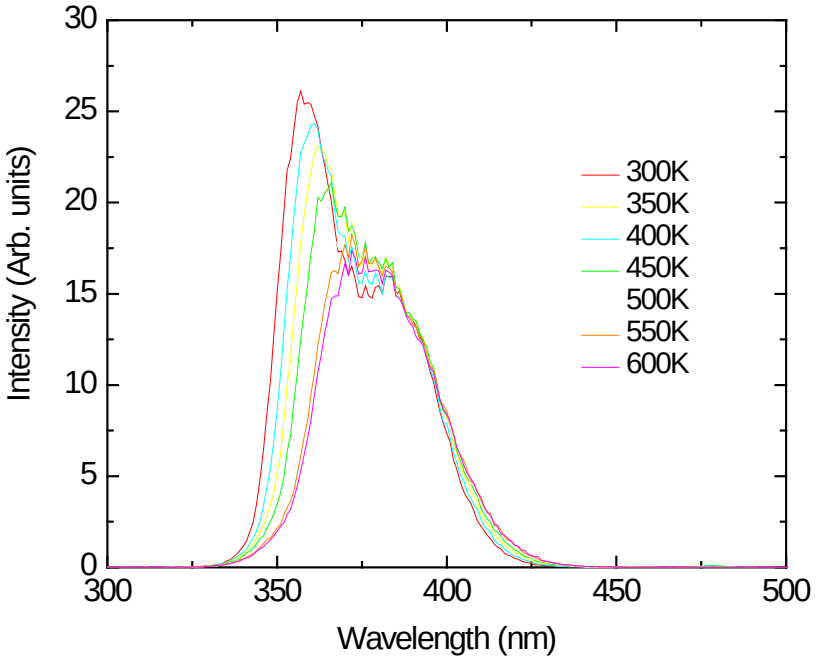
Stokes shift and self-absorption

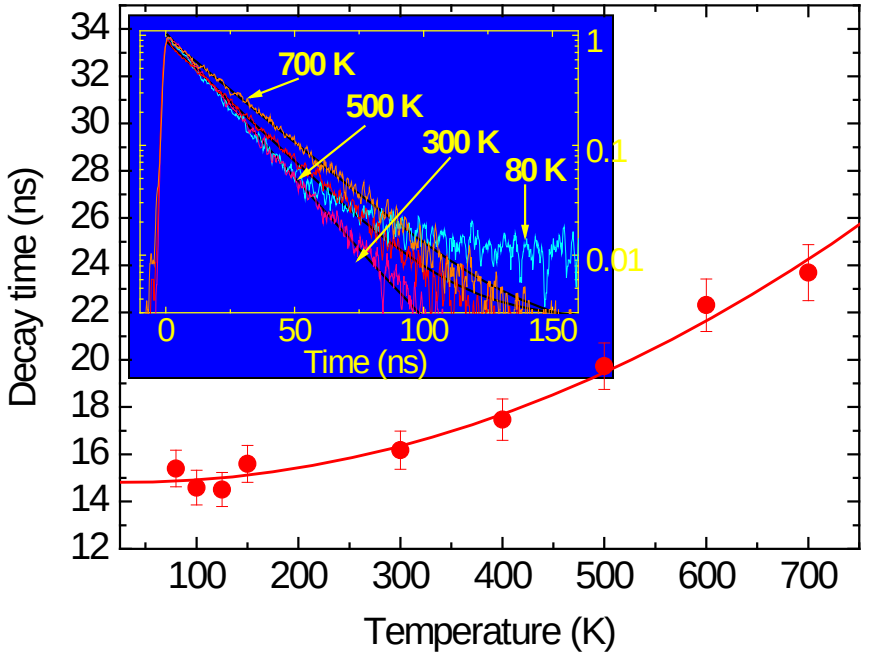




Stokes shift is the energy lost due to lattice relaxation

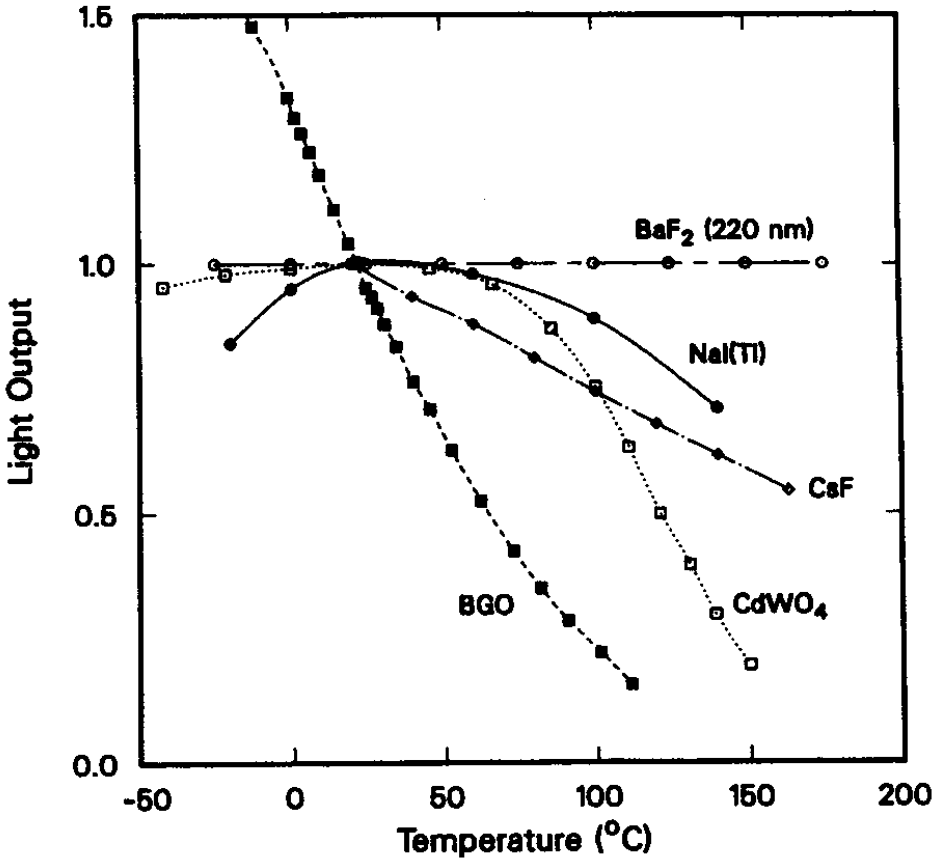
Aspects of self-absorption in $LaBr_3$:5% Ce





- High energy part of emission re-absorbed by Ce^{3+}
- Re-emission by Ce^{3+}
  - emission profile changes
  - scintillation decay time lengthening
  - scintillation light losses (because Q<1)

Thermal quenching



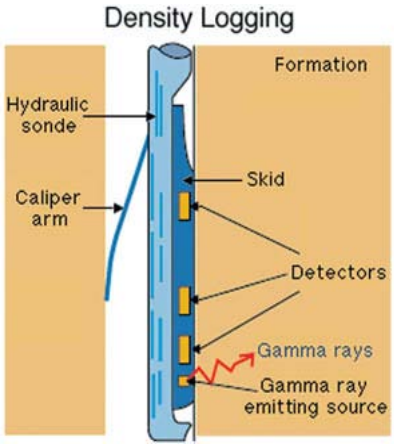
Important for:

- luminescence efficiency Q
- stable scintillator operation

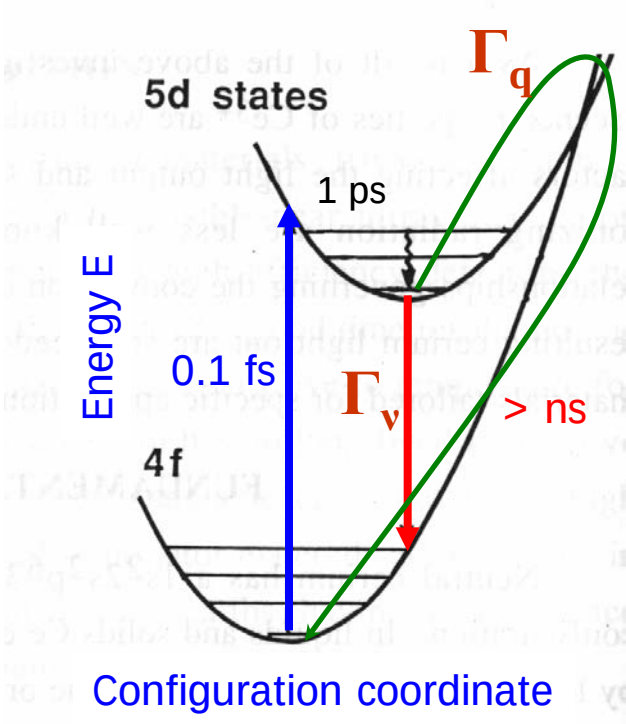
against temperature fluctuations

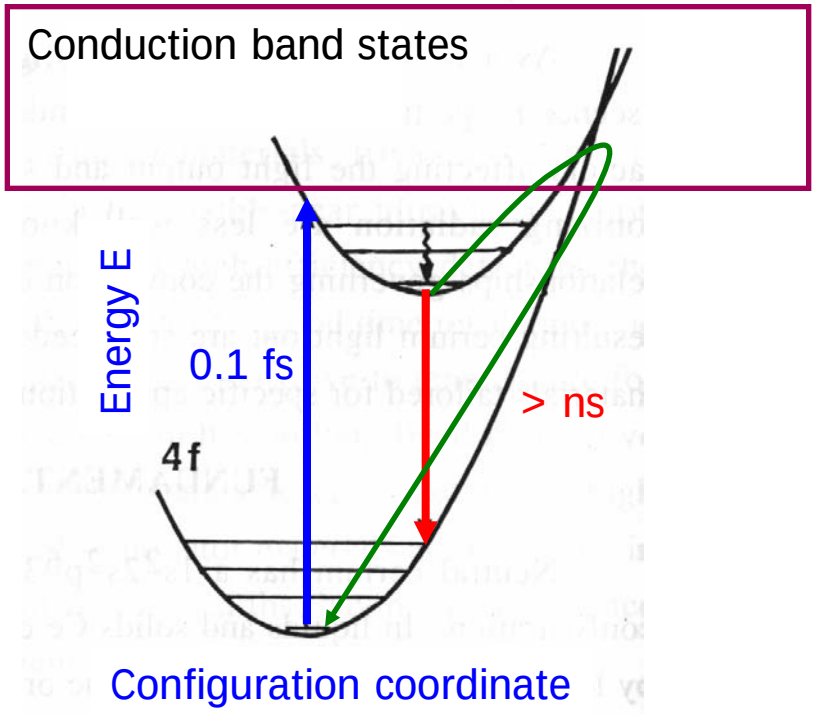
- high temperature applications

e.g. oil-well logging



Thermal quenching of luminescence

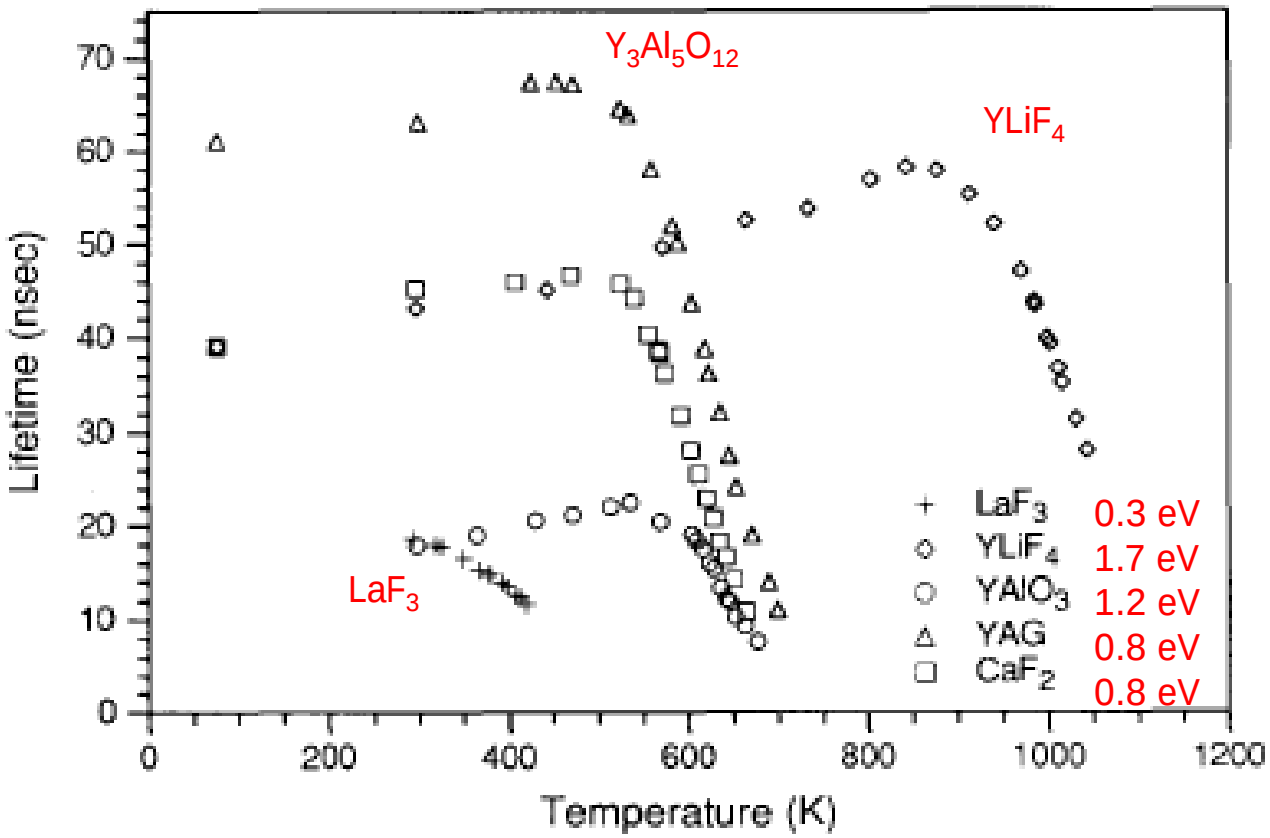




$$Q(T) = \frac{\Gamma_v}{\Gamma_v + \Gamma_q} = \frac{\tau(T)}{\tau_v} = \frac{\tau_v^{-1}}{\tau_v^{-1} + s_0 \exp\frac{-\Delta E_q}{k_B T}}$$

Thermal quenching of Ce^{3+} emission

Luy *et al.* J. Lumin. 48/49 (1991) 251



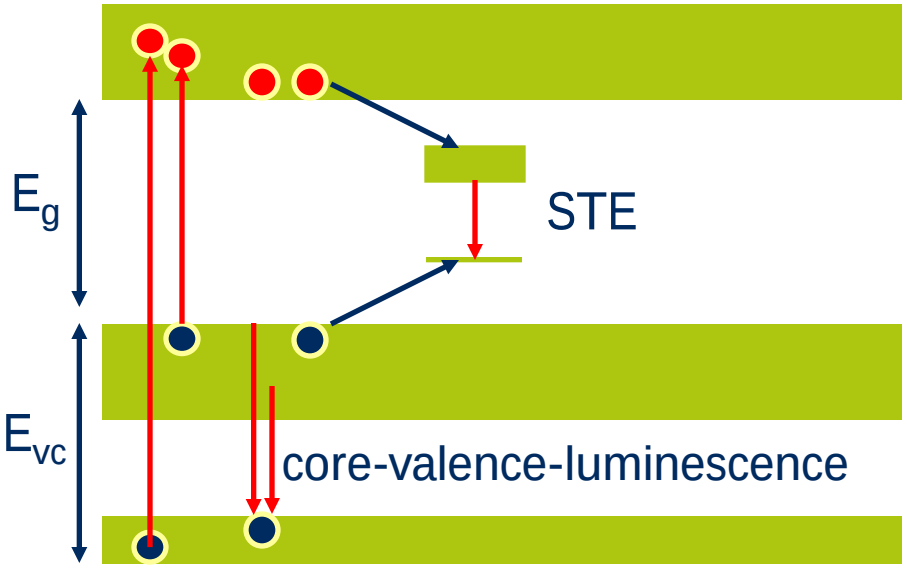
If quenching via CB-states than 5d-level location important

So far we have treated:

- Phase I: Interaction of a gamma photon with scintillators
  - Creation of the ionization track
  - Relation between number of ionizations and band gap
- Phase III: The luminescence centers
  - s^2-elements (Tl, Pb, Bi) and lanthanides (Ce^{3+}, Pr^{3+})
  - Wavelength of emission, self-absorption, Stokes shift
  - Thermal stability and thermal quenching

- Next, Phase II: scintillation mechanisms
  - Intrinsic scintillators
    - Self trapped exciton and core valence luminescence
  - Activated scintillators
    - energy migration from ionization track to luminescence centers
    - $LaCl_3$:Ce and $LaBr_3$:Ce as an example

Core valence luminescence



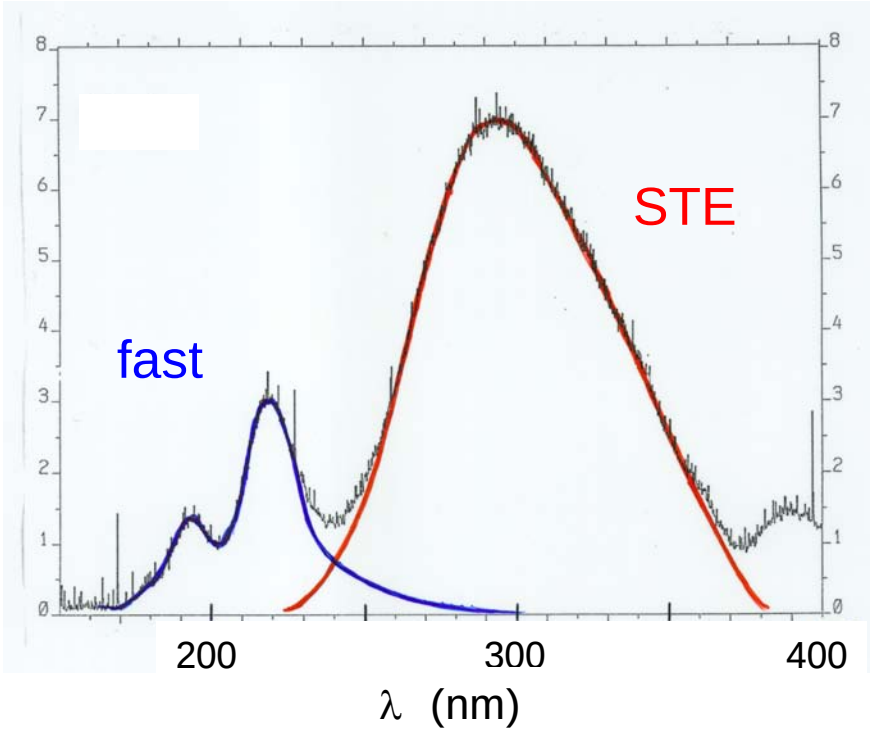
cation cond. band

anion valence band

cation valence band

condition for CVL: $E_{vc} < E_g$

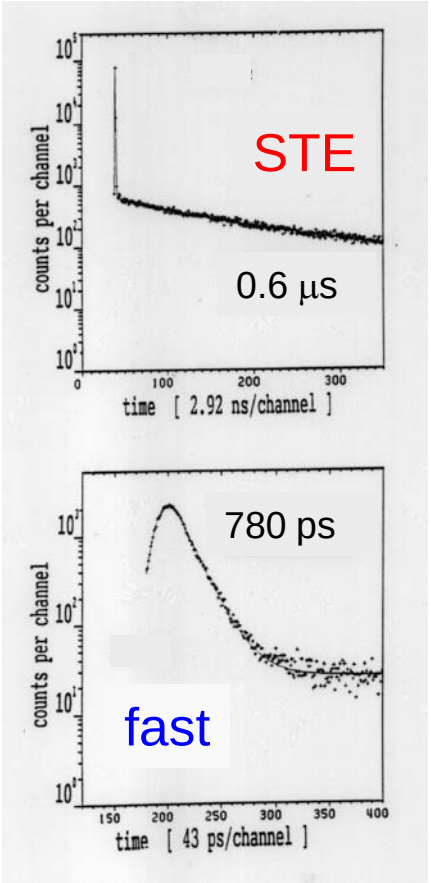
Self trapped exciton and Core valence luminescence in BaF_2



CVL: 1400 ph/MeV 800 ps

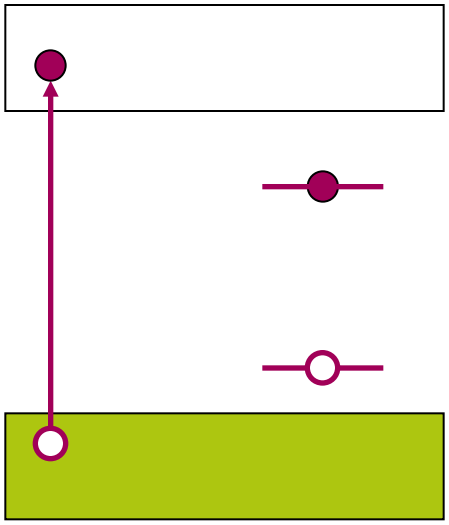
STE: 10^4 ph/MeV 600 ns

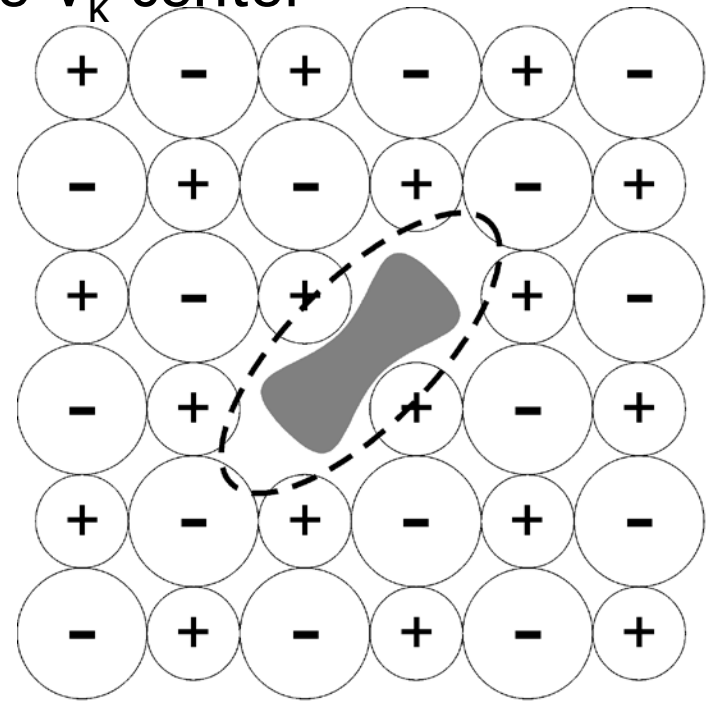
N.N. Ershov, N.G. Zakharov, P.A. Rodnyi
Opt. Spektrosk. 53(1982)89-93



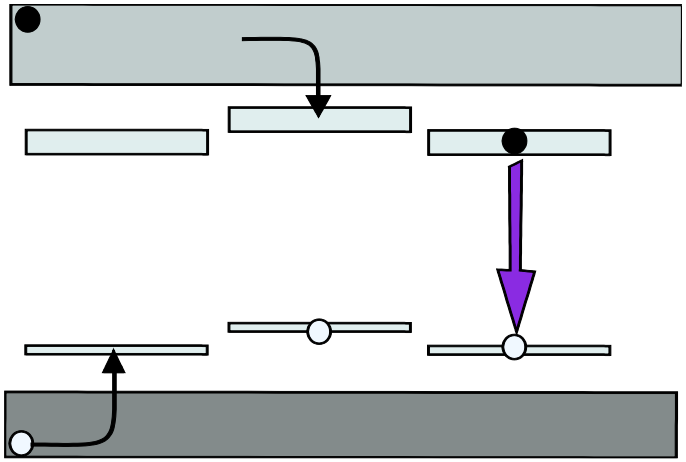
What is a self trapped exciton?

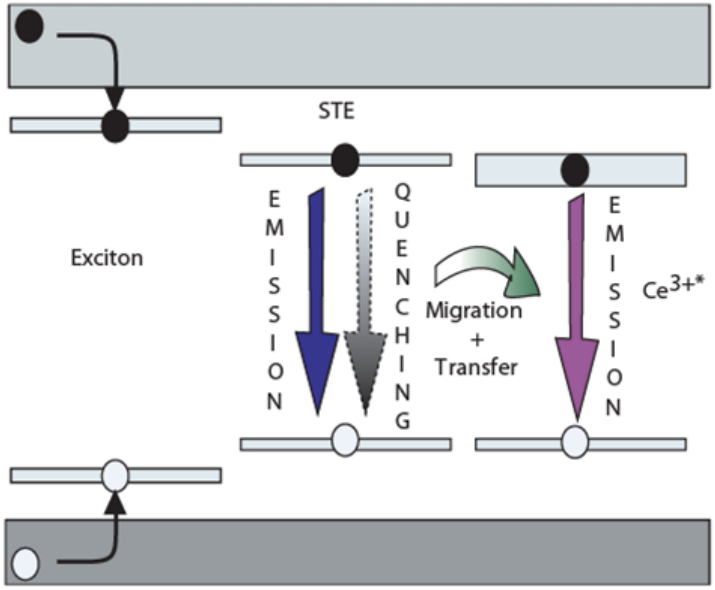
- hole in valence band is shared between two anions forming an X_2^- molecule like defect (V_k center)
- electron in an orbit around the V_k center





Scintillation mechanism in $LaCl_3$ and $LaBr_3$



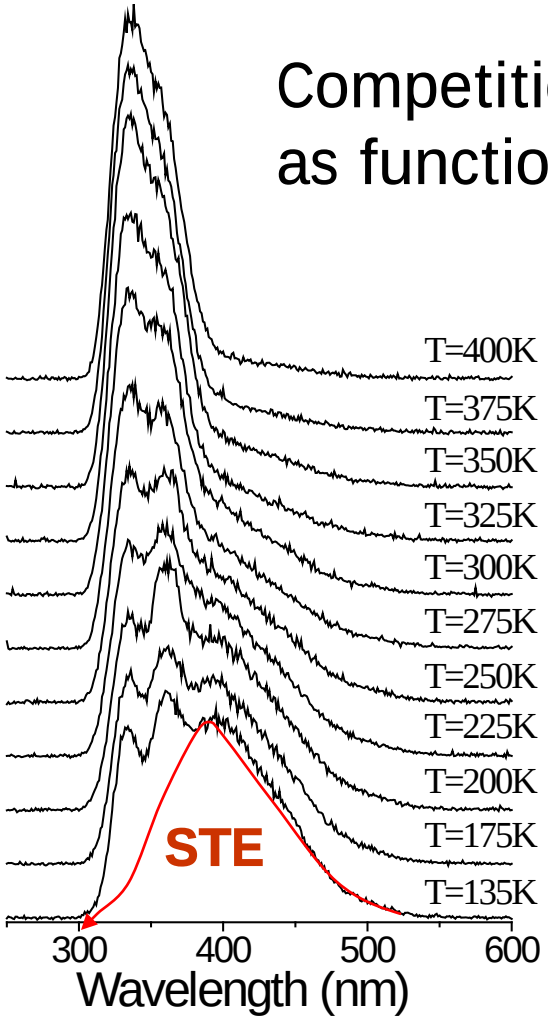


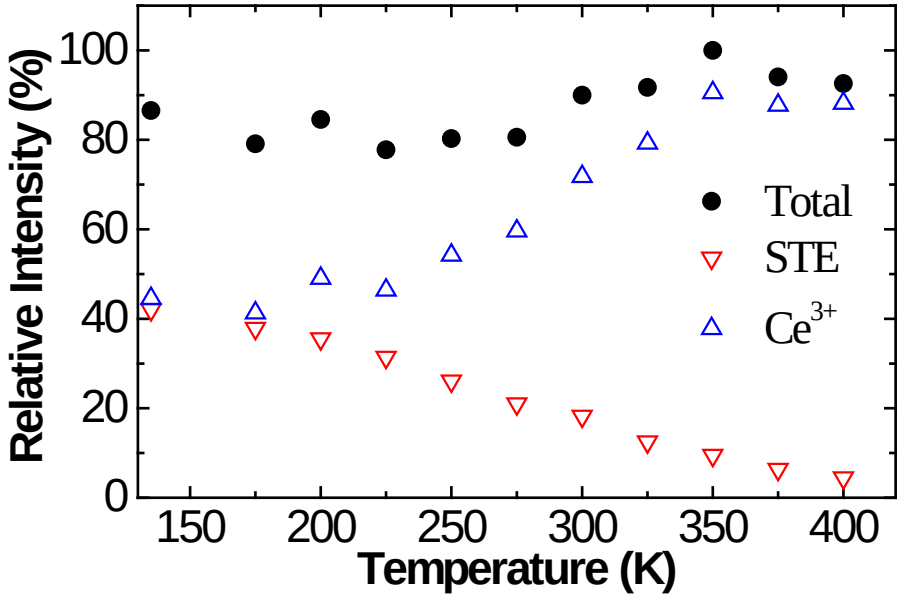
Ce^{3+} + h → Ce^{4+}
Ce^{4+} + e → $(Ce^{3+})^*$ → photon
17 ns fast scintillation component

e + h → exciton→ STE
STE→ STE emission (or loss)
µs slow scintillation component
STE→Ce transfer

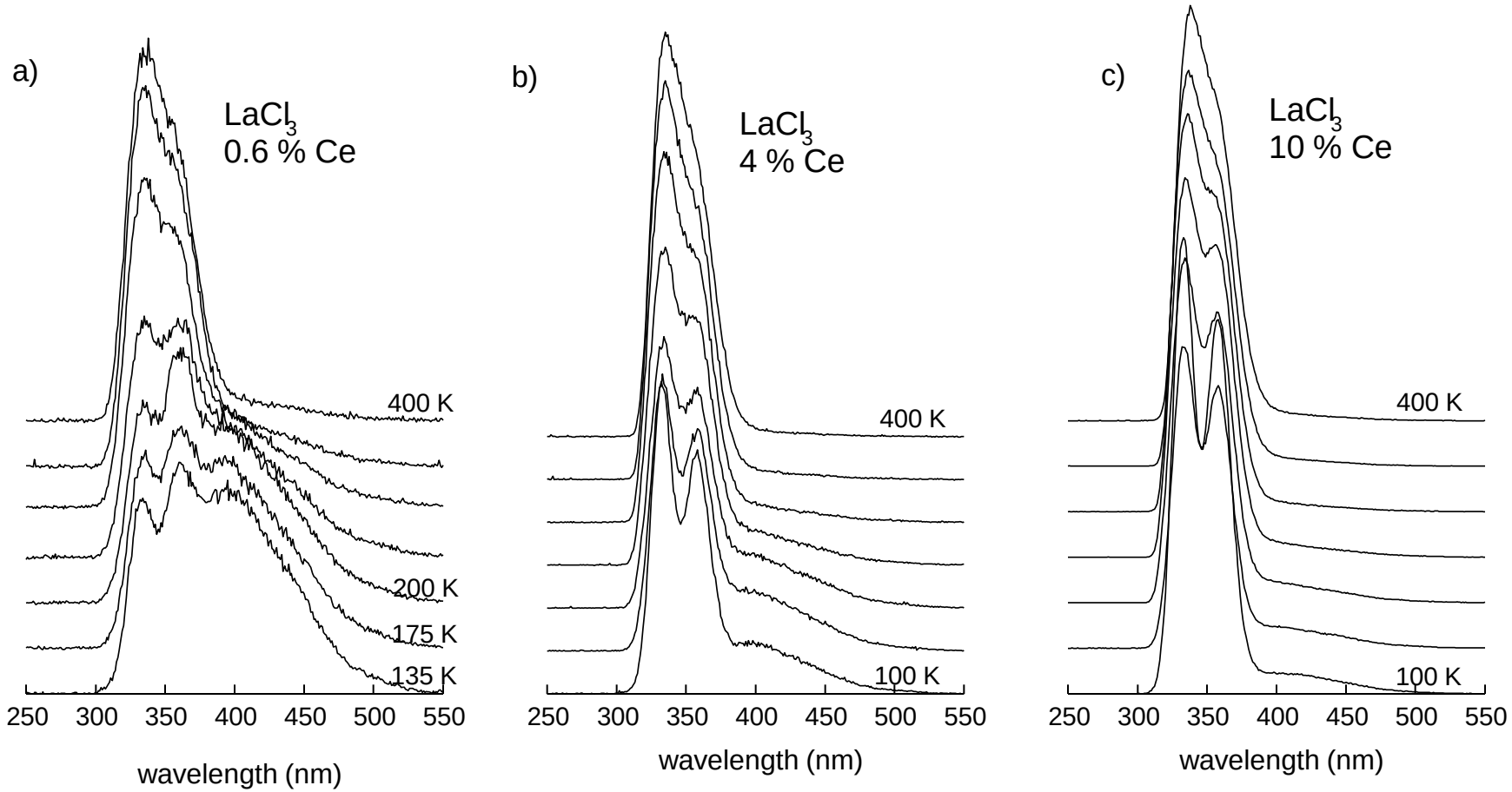
X-ray excited luminescence $LaCl_3$:0.6% Ce^{3+}

Competition STE emission and Ce emission as function of temperature





Competition STE emission and Ce emission as function of temperature and concentration



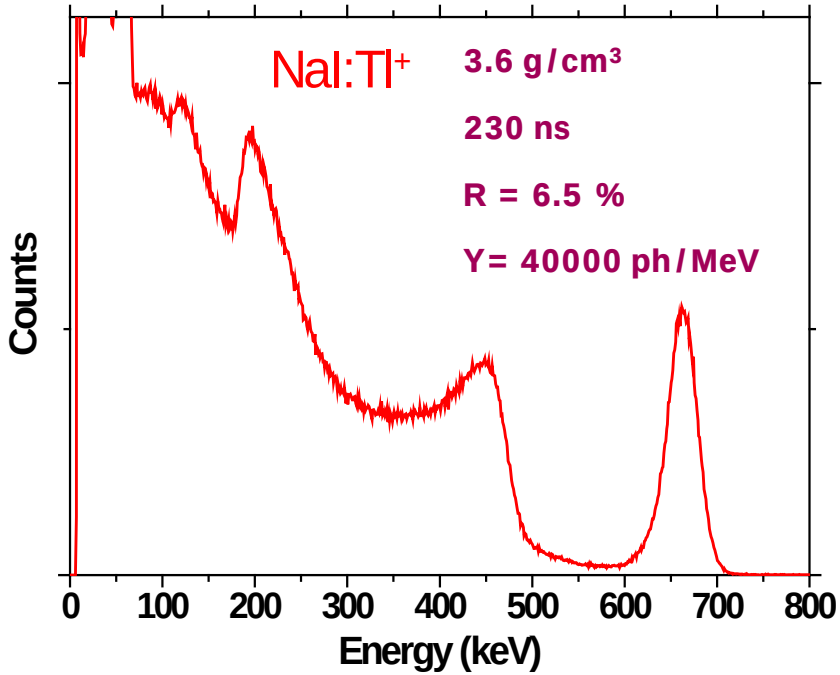
Summary scintillation processes in La-halides

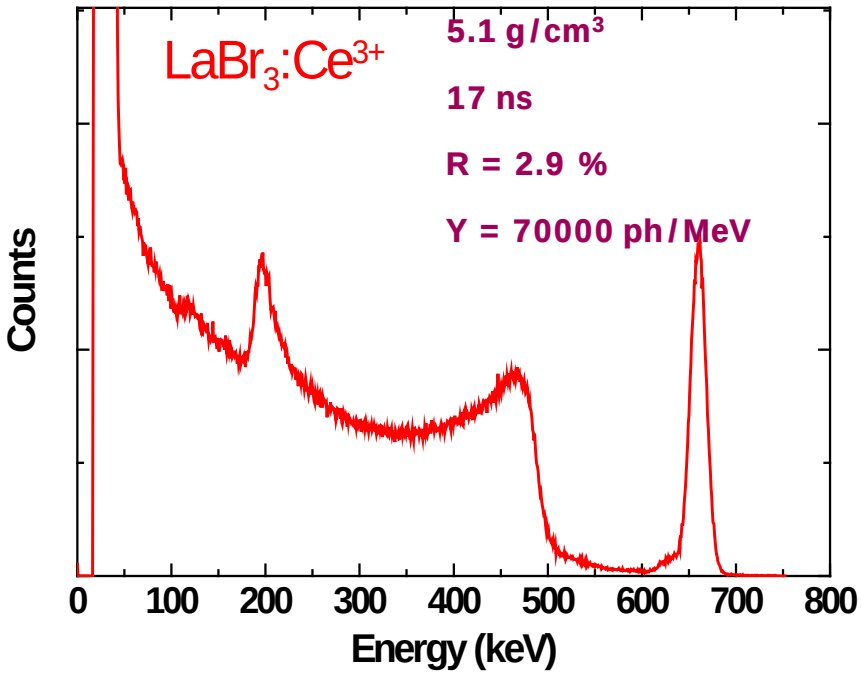
- Direct capture of electron and holes
  - Fast Ce^{3+} emission decay component of 16 ns
- Creation of self trapped excitons
  - Slow STE emission
  - Thermally activated STE migration
- Energy transfer from STE to Ce^{3+}
  - Slow Ce^{3+} scintillation components

Different competing scintillation processes depending on
- Ce concentration
- on temperatures

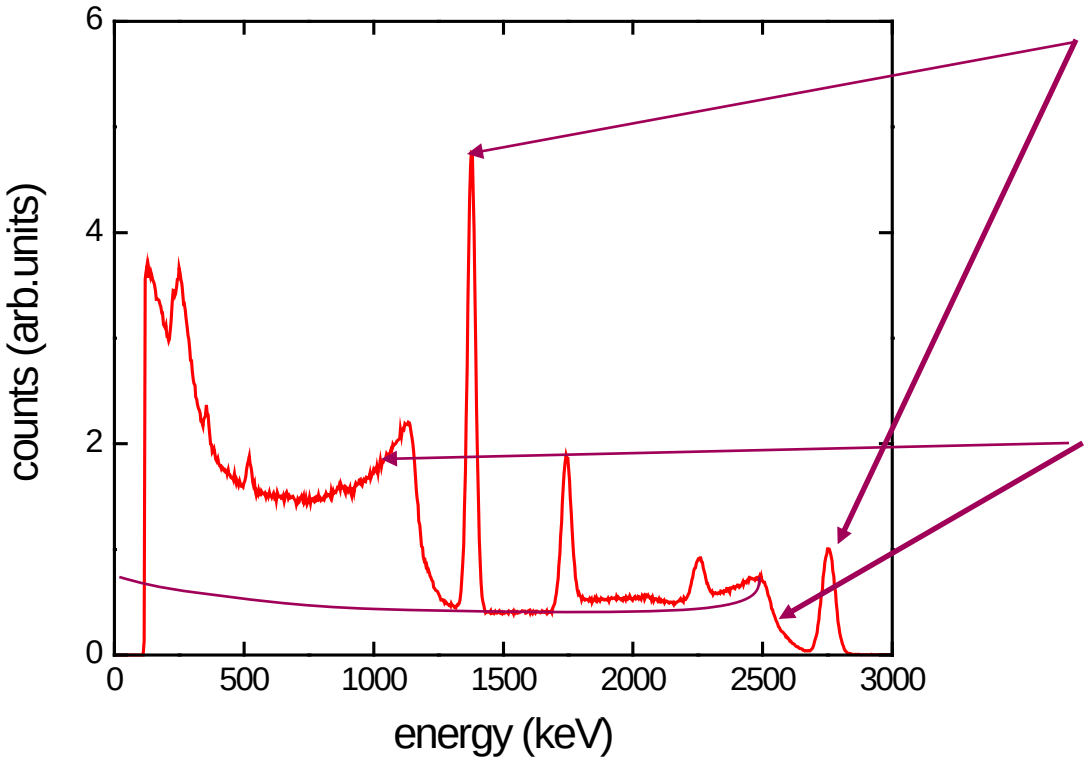
Further reading: G. Bizarri, P. Dorenbos, Phys. Rev. B 75, 184302 2000

$LaBr_3:Ce^{3+}$; record low energy resolution at 662 keV





Anatomy of a pulse height spectrum from ^{24}Na-source measured with $LaBr_3$:5%Ce



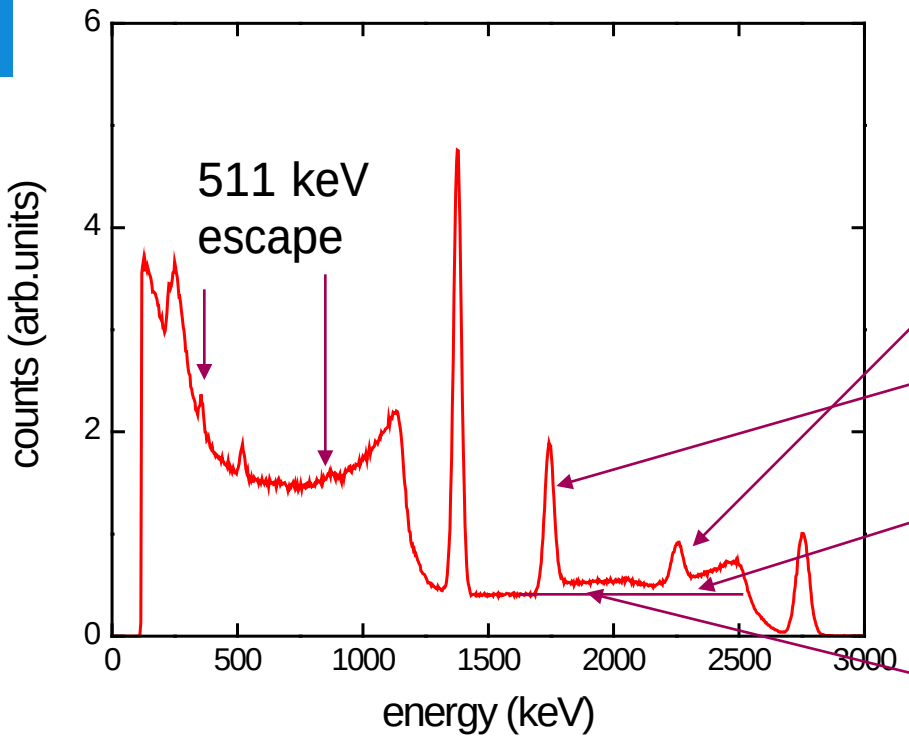
spectrum due to 1.37 MeV + 2.75 MeV gamma source

Photoelectric interaction
→ Total absorption peaks

Compton scattering
→Compton edges
→Compton continua

$$E_C = \frac{2E_\gamma^2}{511 + 2E_\gamma} = 2.52 \text{ and } 1.16 \text{ MeV}$$

Anatomy of a pulse height spectrum from ^{24}Na-source measured with $LaBr_3$:5%Ce



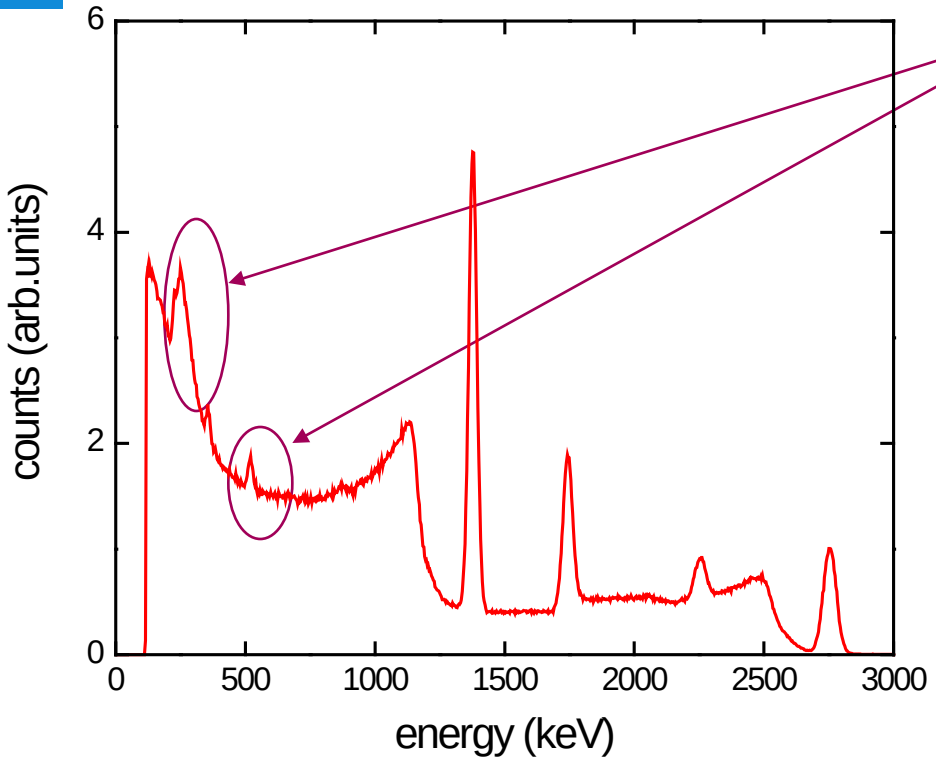
Pair creation

- $e^- + e^+ + E_{kin}$ (=Eγ -1.02 MeV)
- $e^+ + e^- \rightarrow$ 2 x 511 keV γ photons

- 511 keV escape peak at 2.24 MeV
- double escape at 1.73 MeV

Compton scattering of annihilation quanta with escape of scattered gamma photons

Anatomy of a pulse height spectrum from ^{24}Na-source measured with LaBr3:5%Ce



Two remaining features at 511 keV and around 250 keV

Back scatter peaks due to gamma rays scattered from materials outside the scintillator

- Compton scattered gamma's
- annihilation gamma 511 keV

BrilLanCe™ trademark of La-halide scintillators

BrilLanCe™ 380

It's what's Inside that Counts!

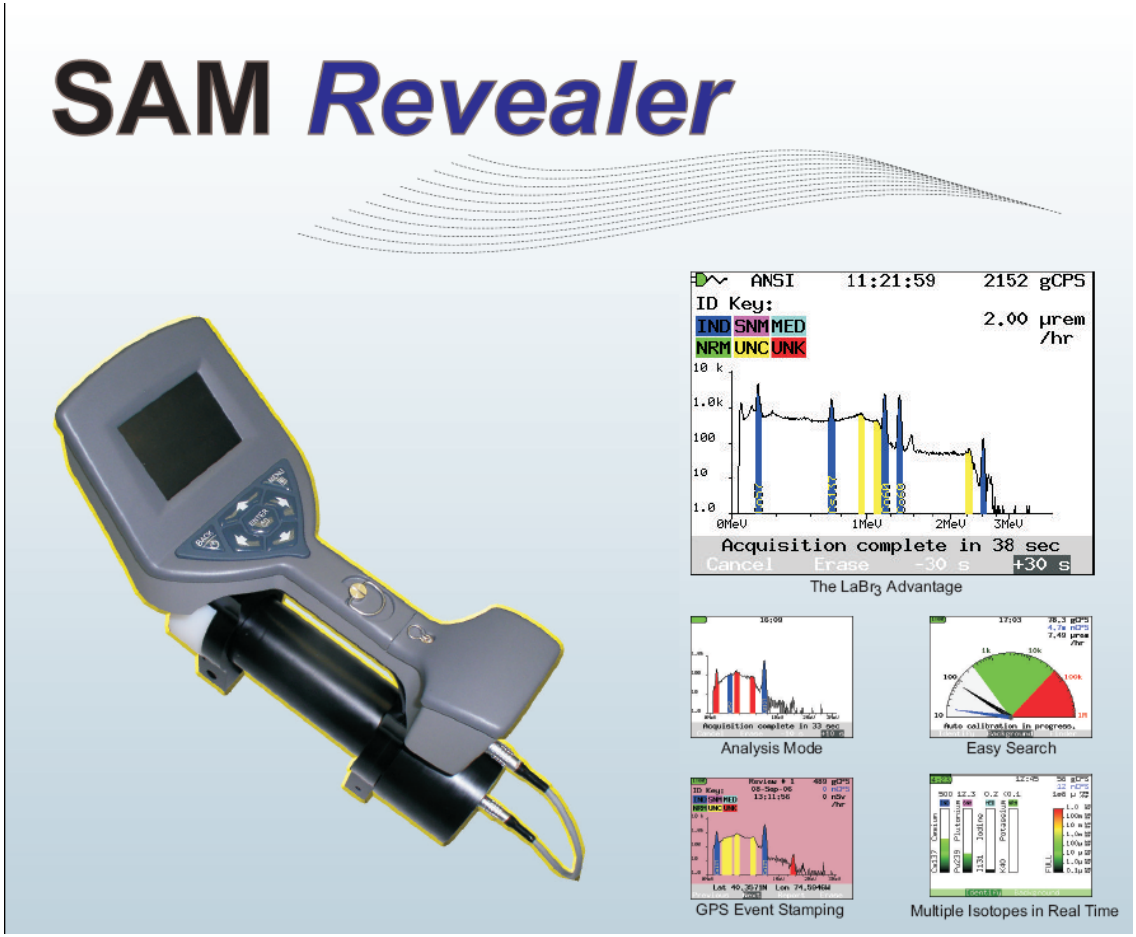
Discovered by TU-Delft

$LaCl_3$:Ce crystal growth progress
From labsize to 4”x6” scintillators crystals

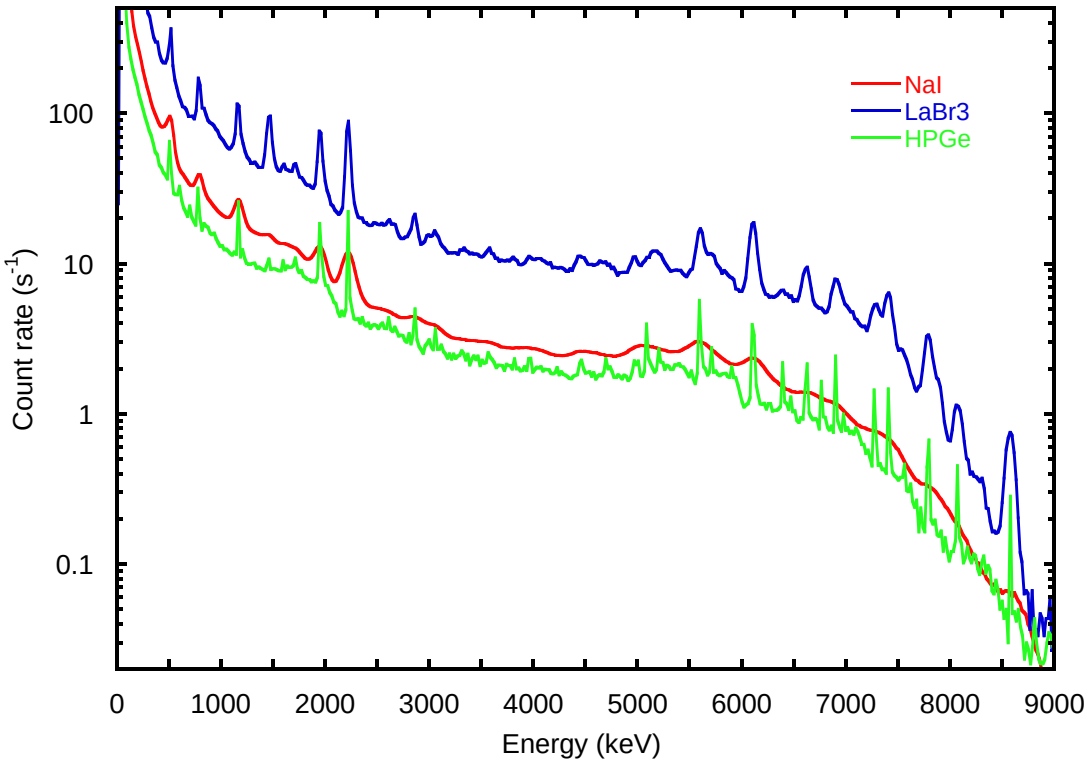
BrilLanCe™ 350
It's what's Inside that Counts!

Scintillation Material

$LaBr_3$:Ce for nuclide identification



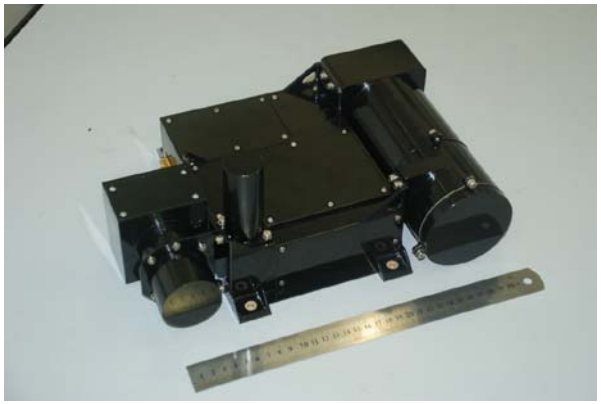
Comparison NaI:Tl, $LaBr_3$:Ce, HP-Ge



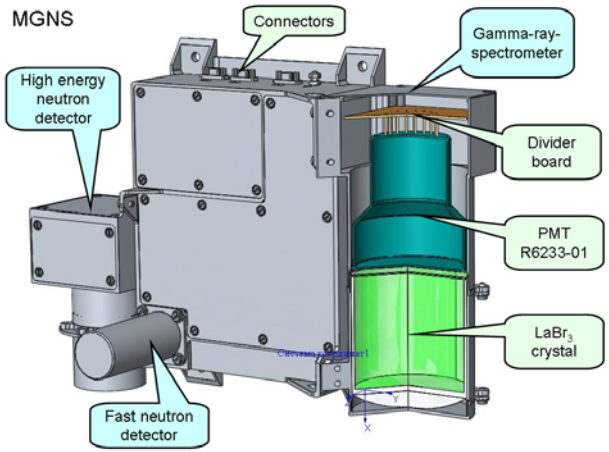
Real energy [keV]	Element
511	anihilation peak
518	Cl
583.1	208Tl
	Ge
	Ge
788	Cl
911.1	228Ac
1165	Cl
1201	H d
1460	40K decay
1600	Cl f
1686	Na s
1712	H s
1778	Al f
1953	Cl d
1957	Cl f
2223	H
2611	208Tl
2622	Na d
2676	Cl f
2843	Si d
2864	Cl f
2977	F s
2997	Cl f
3017	F f
3062	Cl f

$LaBr_3$:Ce in 2011 on mission to Phobos (Mars) and in 2014 on mission to Mercury

$LaBr_3$ detector for the Phobos-Grunt mission



$LaBr_3$ detector for the BepiColombo mission



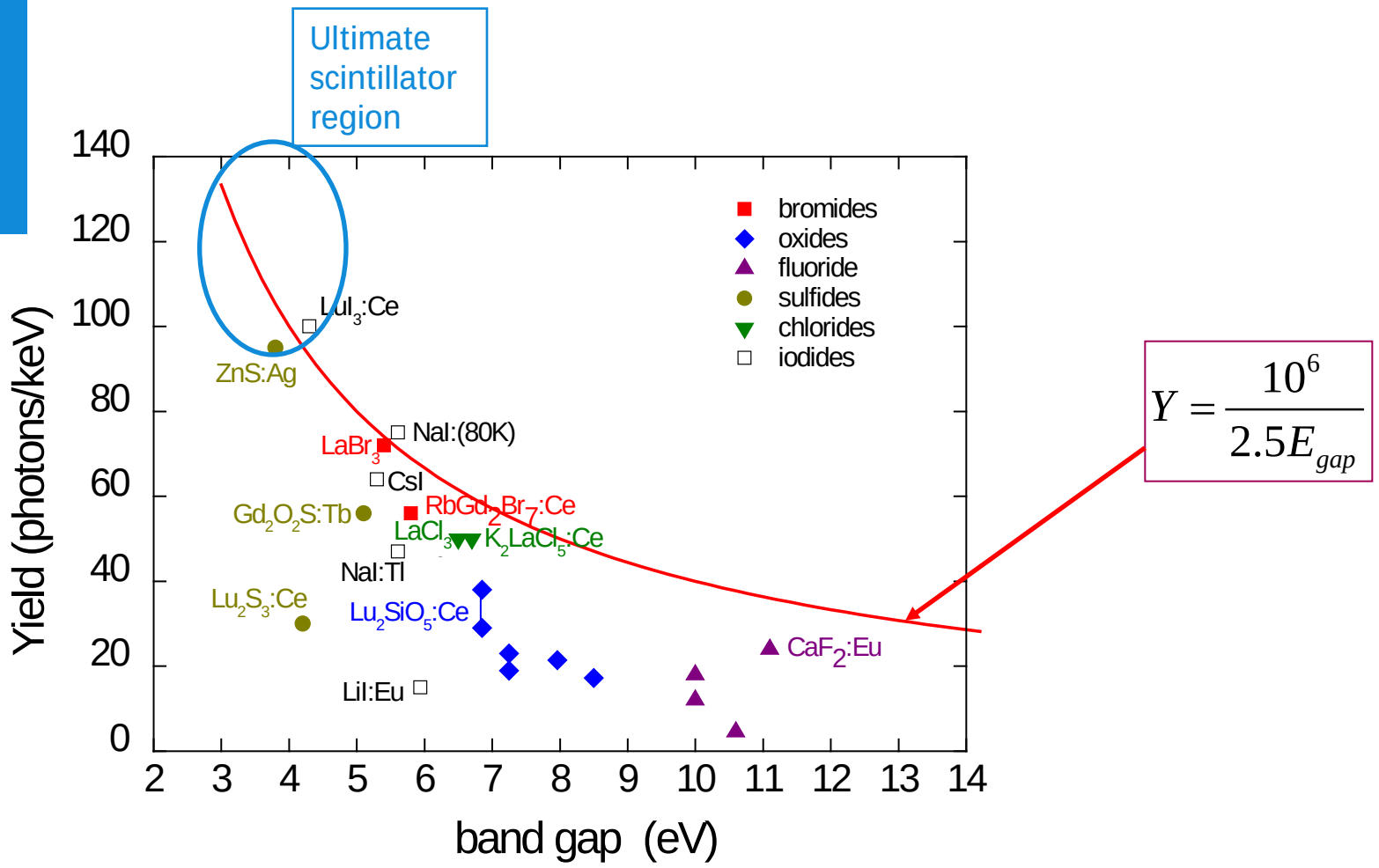
Aspects that determine resolution

$$R^2 = R_{\text{stat}}^2 + R_{\text{inhom}}^2 + R_{\text{nonprop}}^2$$

$$R_{\text{stat}}^{FWHM}(\%) = 235\sqrt{\frac{1+v_{PMT}}{N_{dph}}} \qquad v_{PMT} \approx 0.2$$

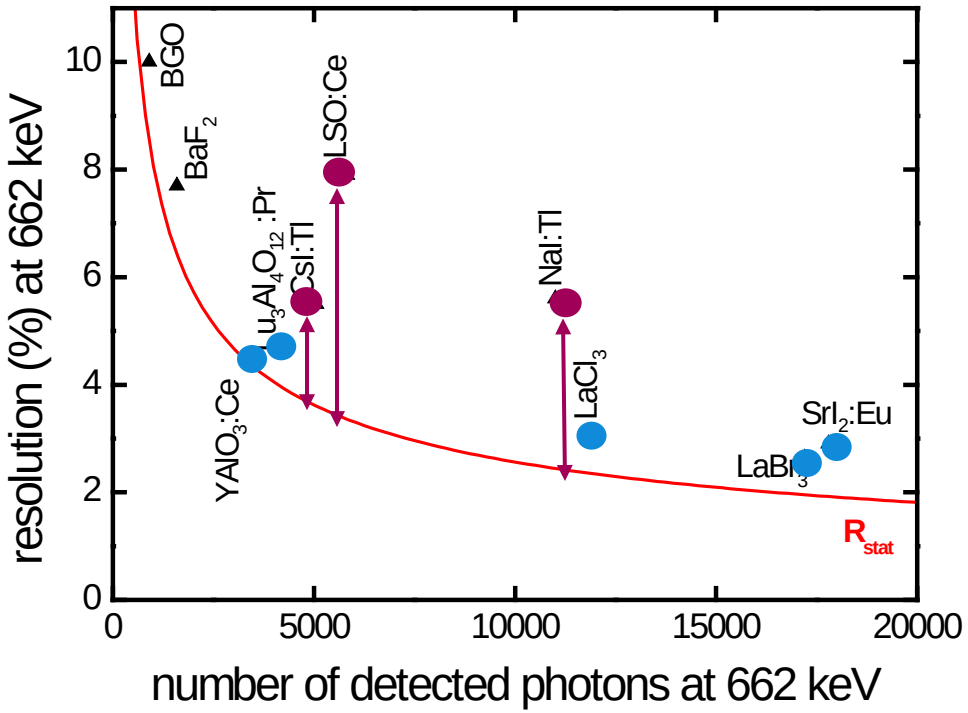
- the fundamental limit is governed by statistics
- you need to detect as many photons as possible
  - → bright scintillator
  - → efficient low noise photon detector

The ideal scintillator has S=Q=1



Energy resolution @ 662 keV

$$R_{\text{stat}} = 2.35\sqrt{\frac{1+0.2}{N_{dph}}}$$



- $YAlO_3$:Ce, $Lu_3Al_5O_{12}$:Pr, $LaCl_3$:Ce, $LaBr_3$:Ce, SrI_2:Eu are reasonably close to fundamental limit.
- Lu_2SiO_5, NaI:Tl, CsI:Tl, strong deviation

What causes the strong deviation?

Aspects that determine resolution

$$R^2 = R_{\text{stat}}^2 + R_{\text{inhom}}^2 + R_{\text{nonprop}}^2$$

$$R_{\text{stat}}^{FWHM}(\%) = 235\sqrt{\frac{1+v_{PMT}}{N_{dph}}} \qquad v_{PMT} \approx 0.2$$

- when the number of created photons is not proportional with the energy of the primary electron
  → additional contribution to resolution (R_{nonprop})
- topic of current interest
  → for study, we need a tunable β-source inside scintillator!

The Compton Coincidence Technique

- Developed by Valentine and Rooney
  - Nucl. Instr. and Meth A353 (1994) 37; IEEE Trans.Nucl.Sci.43(1996)1271
- Provides electron response curves

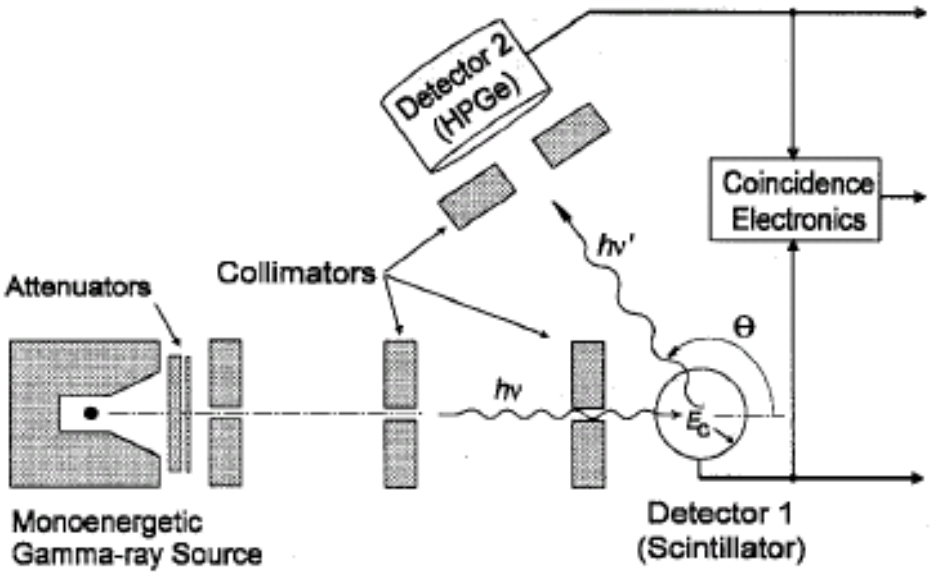


Fig. 1. Block diagram of Compton Coincidence Technique (CCT).

$$h\nu' = \frac{h\nu}{1+\left(h\nu/m_0c^2\right)\left(1-\cos\vartheta\right)}$$

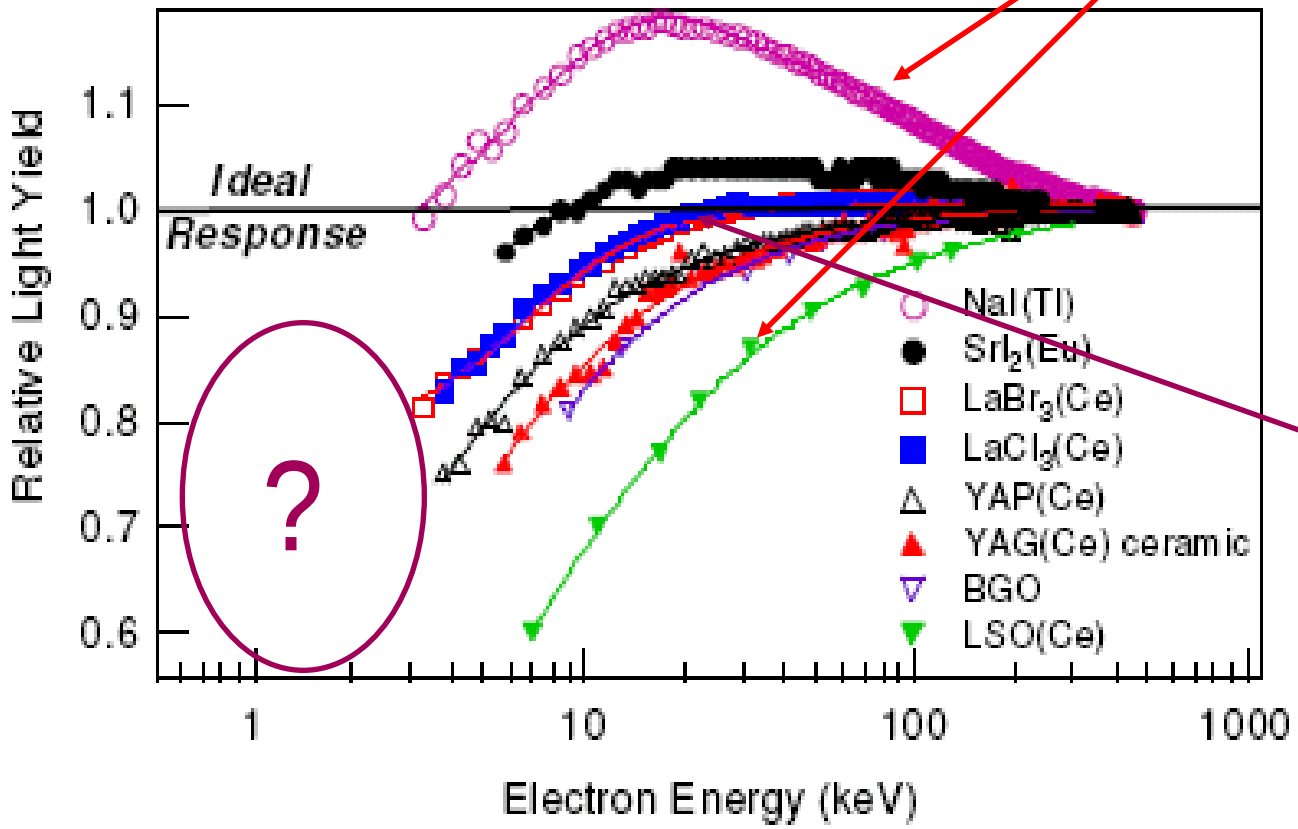
$$E_C = h\nu - h\nu'$$

Advantage
- intrinsic tunable β-source
- energy range 6-400 keV

Scintillator response to electrons

NaI:Tl. LSO:Ce
Strongly
Non-proportional

Cherepy et al. IEEE Trans. Nucl. Sci. 56(3) (2009) 873

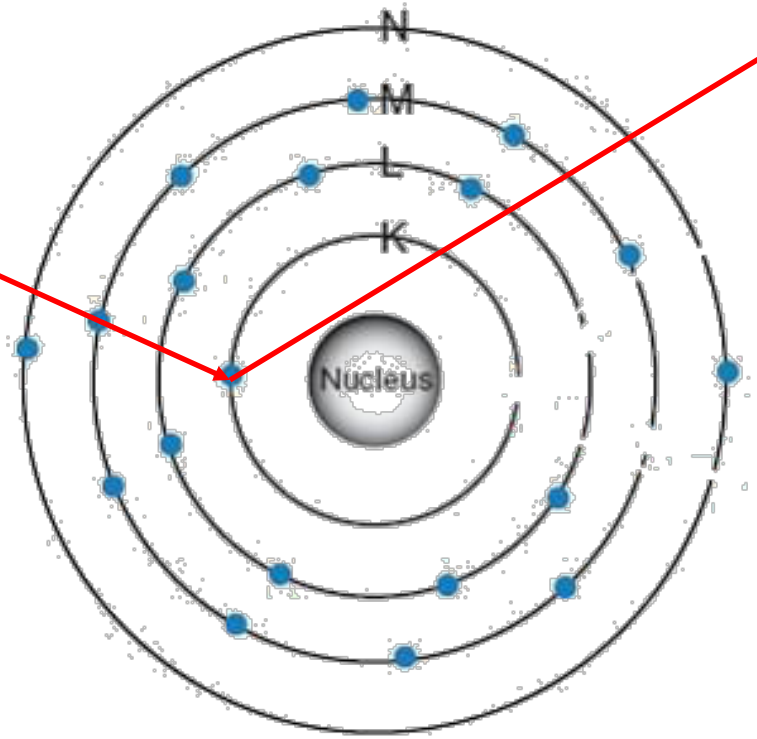


$LaBr_3$:Ce
$LaCl_3$:Ce
$YAlO_3$:Ce
proportional

K-dip spectroscopy method

$E_{phe}=E_X-E_K=\Delta E$

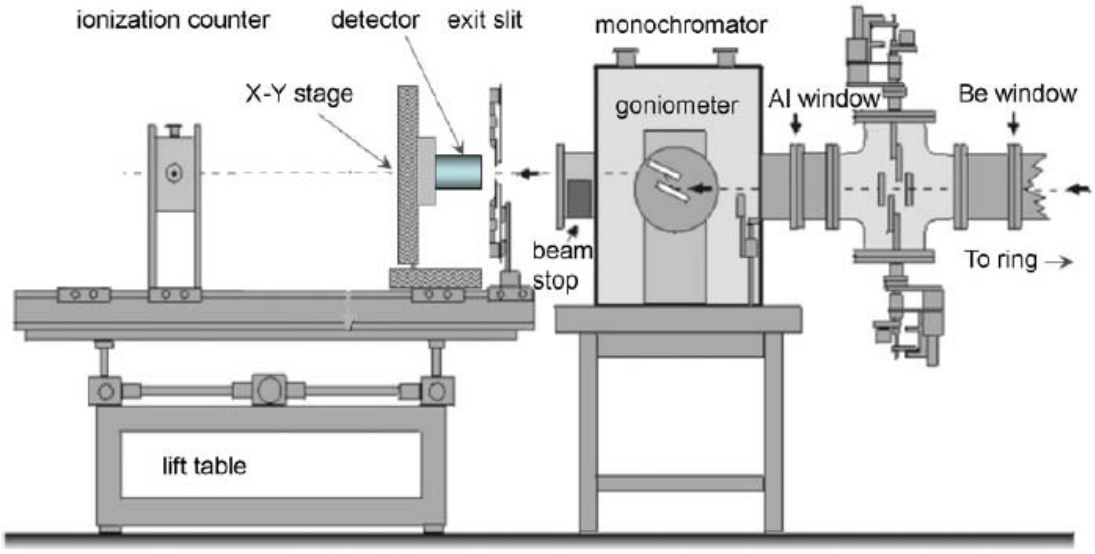
$E_X=E_K+\Delta E$



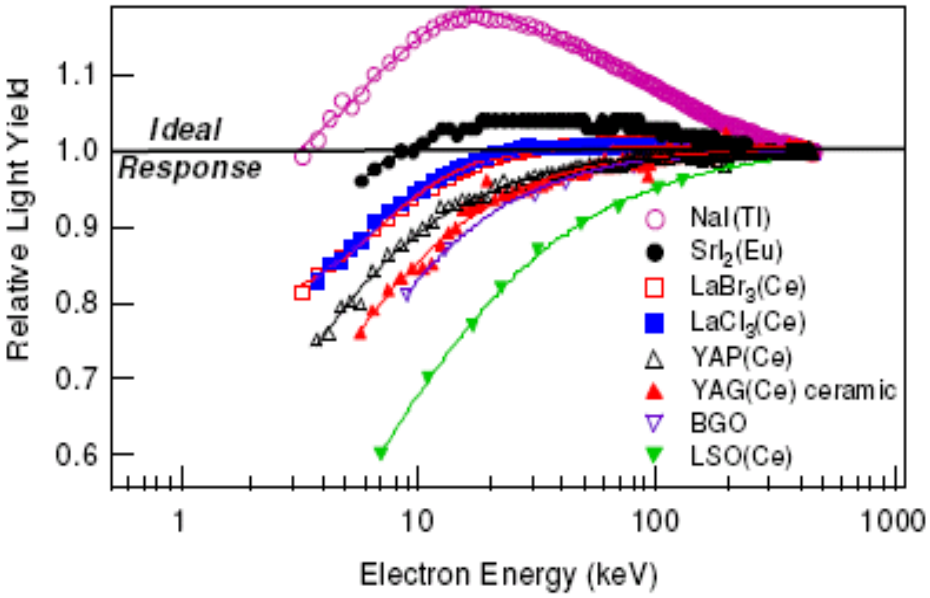
Energy photo-electron = E_X-E keV
Yield (photo-electron)= total yield – yield@E_K keV
→We have created a tunable internal β-source
→Electron response from ~0.1 to (100-E_K) keV

Tunable synchrotron X-ray source

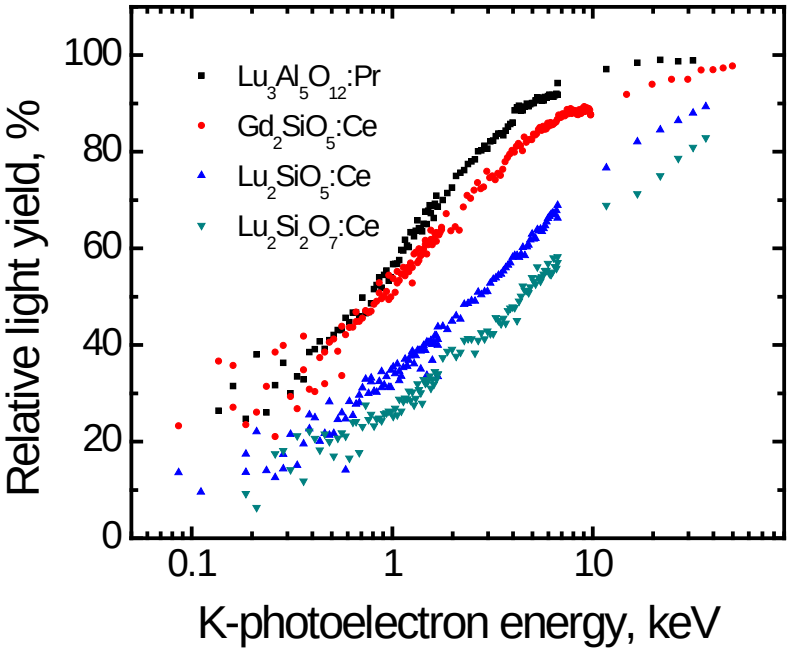
- Synchrotron X-ray beamline HASYLAB, DESY, Hamburg
- 9-100 keV monochromatic and tunable
  - 5-15 eV energy resolution
- Pencil beam
  - → we always excite the same volume element of the scintillator
  - → minimum contribution to resolution from scintillator inhomogeneity



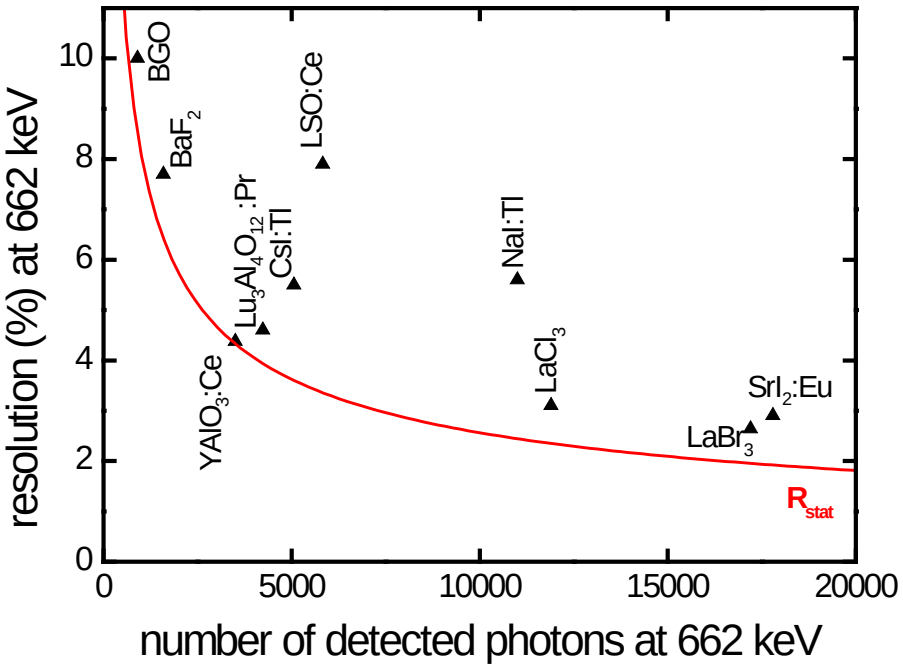
Compton-electron spectroscopy



K-electron spectroscopy



Joining K-dip with SLYNCI data
→100eV – 400keV
→Electron response curves
→20 different scintillators



- Can we go below 2% energy resolution at 662 keV?
  - We need bright scintillators combined with low noise photon detectors
  - We need proportional scintillators

Time resolution

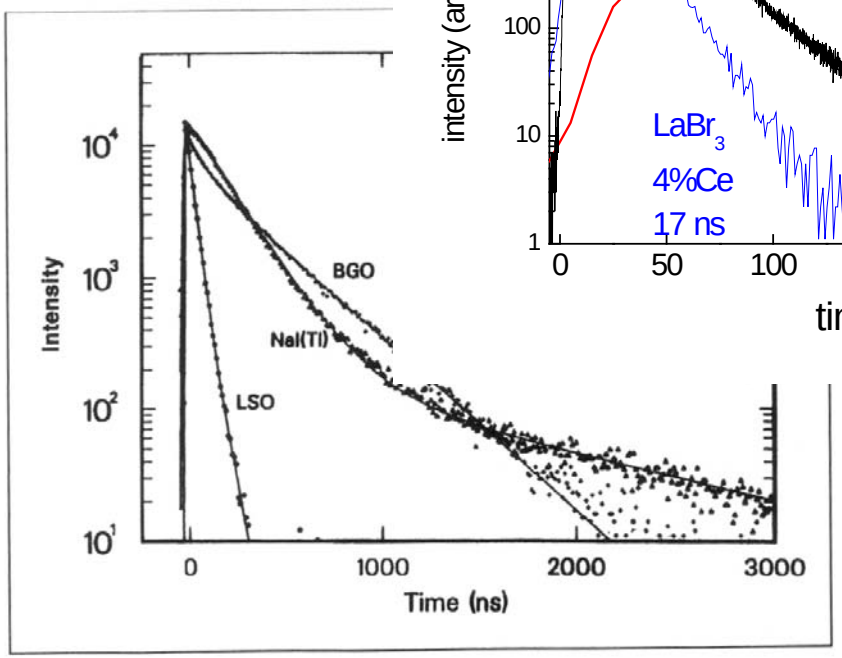
Time resolution = accuracy with which the moment of interaction can be determined

It depends:

- scintillation speed (decay time and rise time)
  - more ph/MeV
- photon detector time resolution
- electronics

Timing resolution

BaF_2



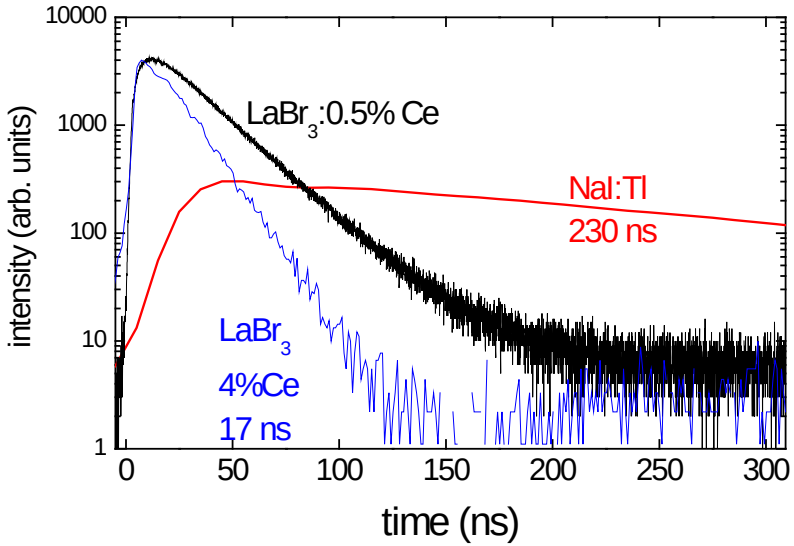
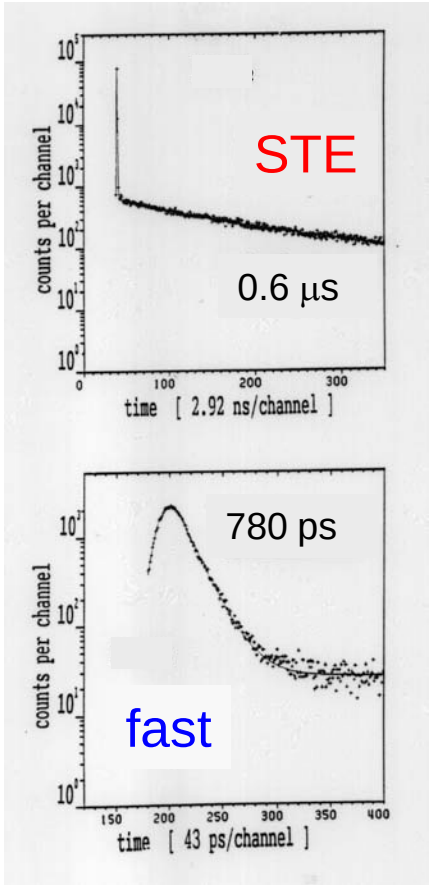
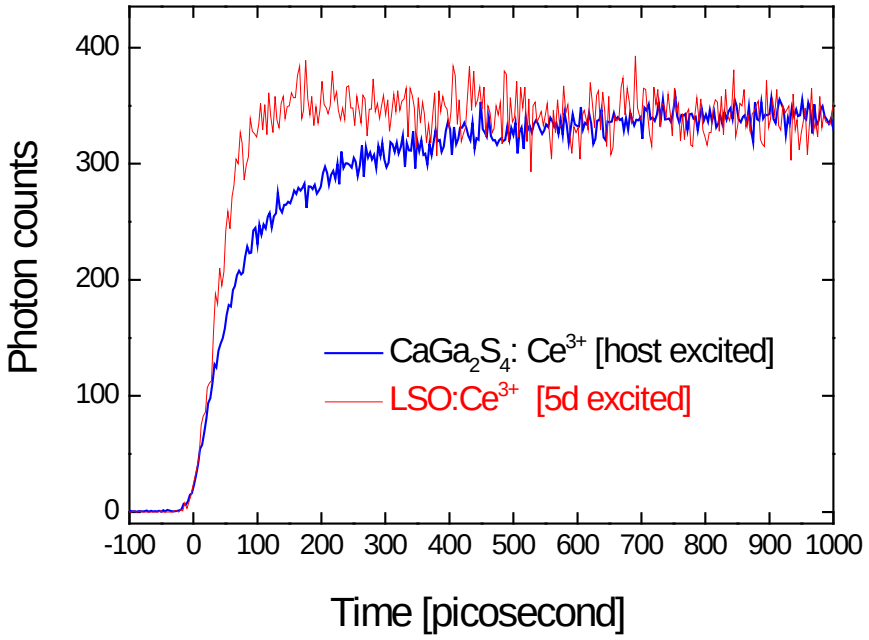


FIGURE 3. Decay of the scintillation emission of NaI(Tl), BGO, and LSO(Ce), after excitation by γ-rays.

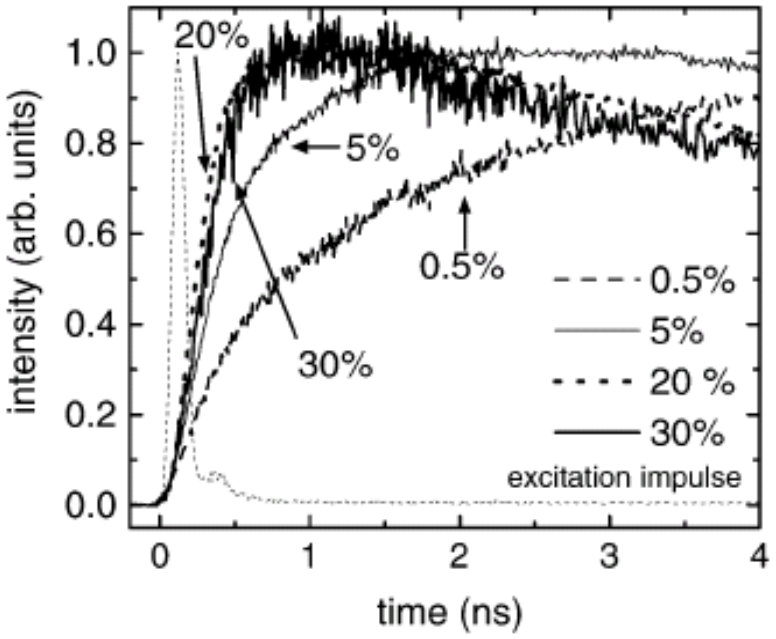


The steepness of the rising pulse is important

Scintillator risetime studies



fs laser excitation



Pulsed X-ray excitation
$LaBr_3$:Ce

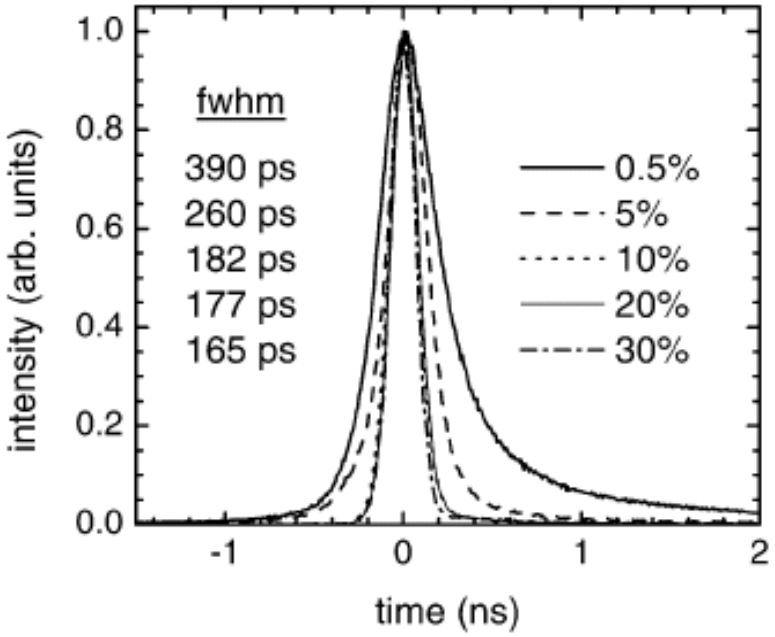
Fast timing is important for

- positron emission tomography
- positron life time studies
- time of flight applications

Typical numbers for time resolution with PMTs and 1 MeV gamma's :

NaI(Tl)	:	1 ns
BGO	:	2-4 ns
BaF_2	:	200 ps
Plastics	:	150 ps

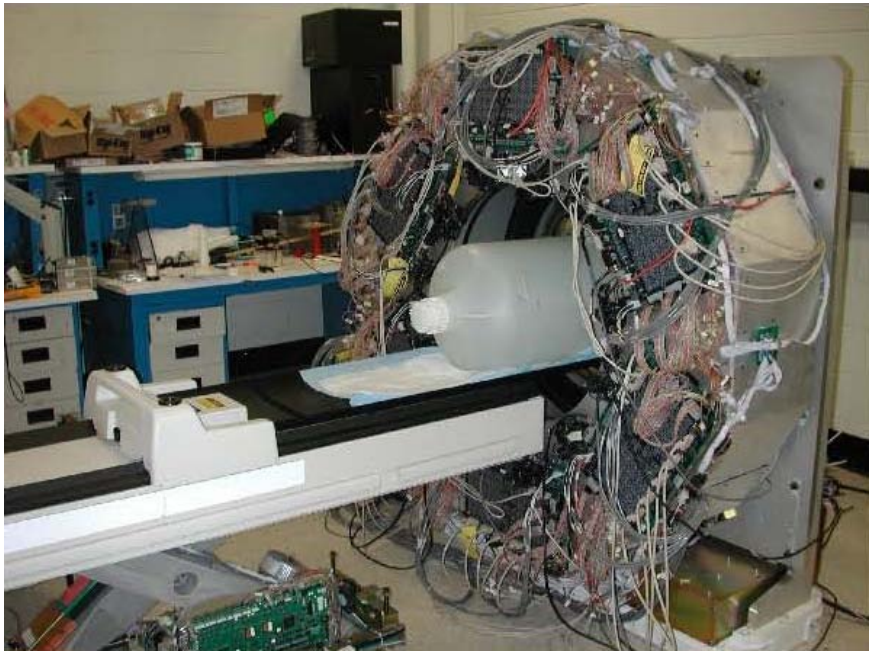
Time resolution $LaBr_3$:Ce at 511 keV with PMTs



$LaBr_3$ TOF-PET scanner

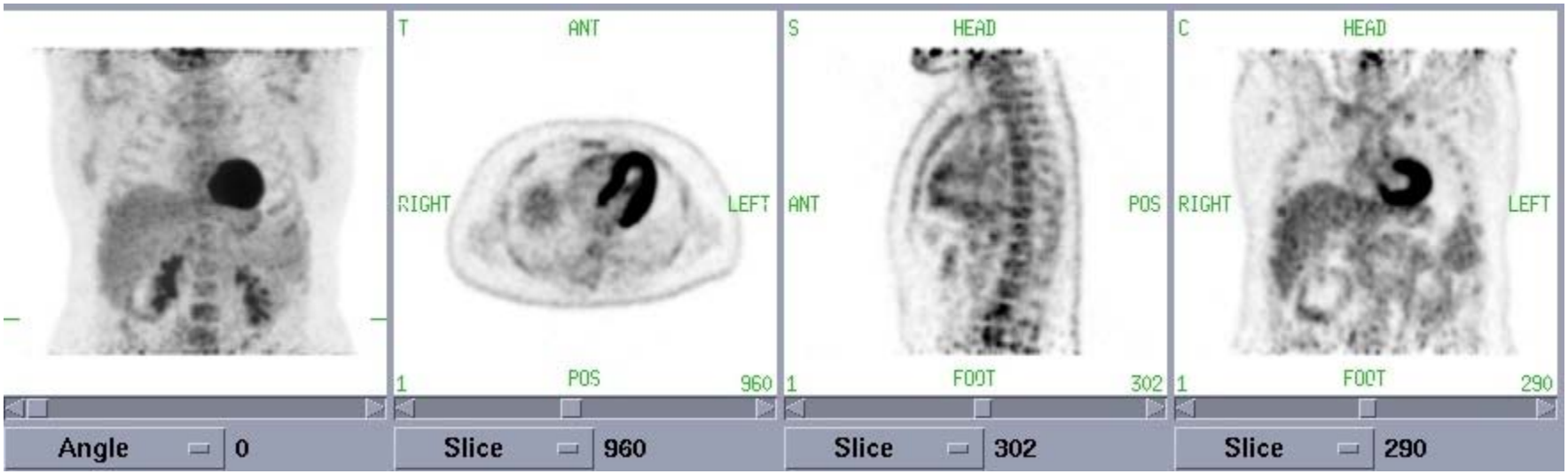
Time-of flight positron
Emission tomography scanner
- 511 keV annihilation photons
- coincident detection
- time resolution 200 ps

Contains 38880
$LaBr_3$ crystals



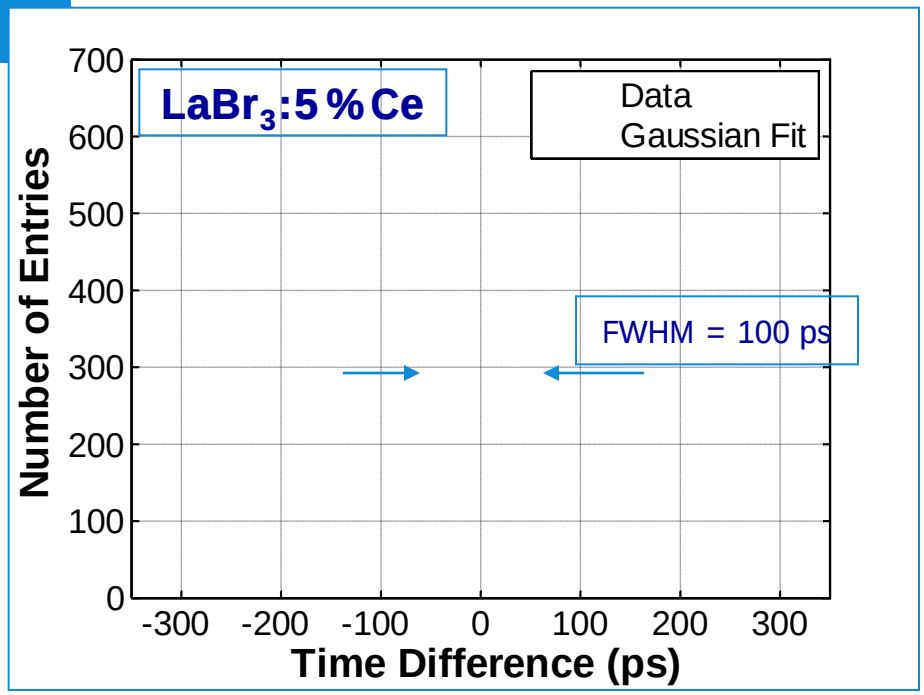
First prototype $LaBr_3$
TOF-PET-scanner (2008-2009)
(dr. J. Karp Pennsylvanian State University)
(and Philips Medical Systems)

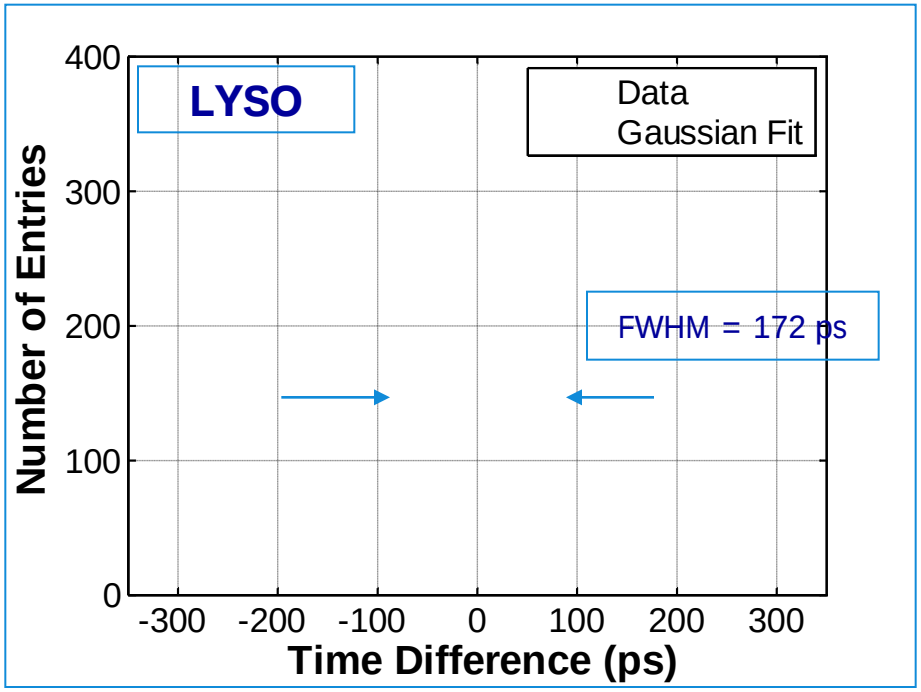
First TOF PET-image with $LaBr_3$:Ce (October 2009)



Advantage TOF-PET strong false signal reduction especially important For fat patients

Timing Spectra with SiPM – CRT at Optimum Threshold Settings





Intrinsic activity of Lu_2SiO_5:Ce

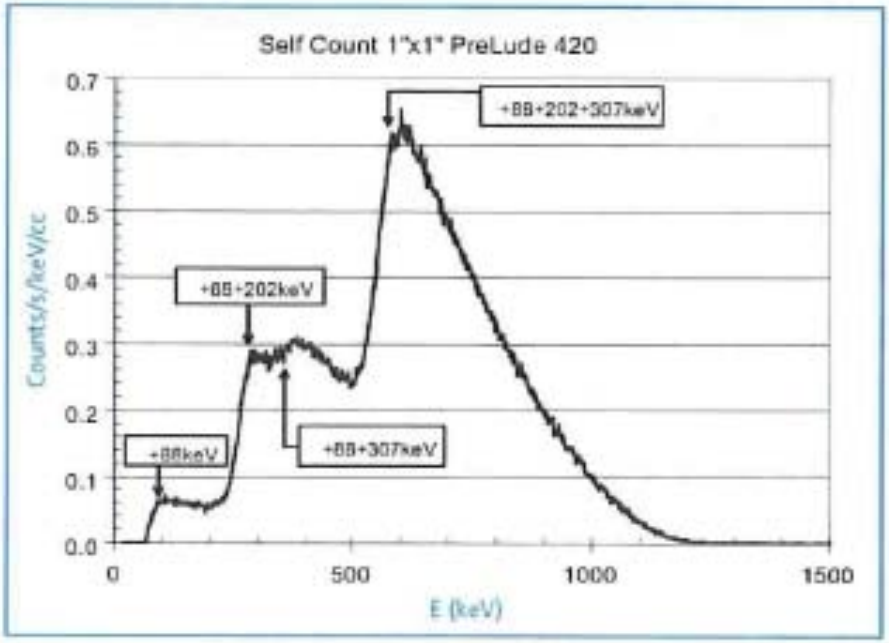


Figure 4. Beta distributions

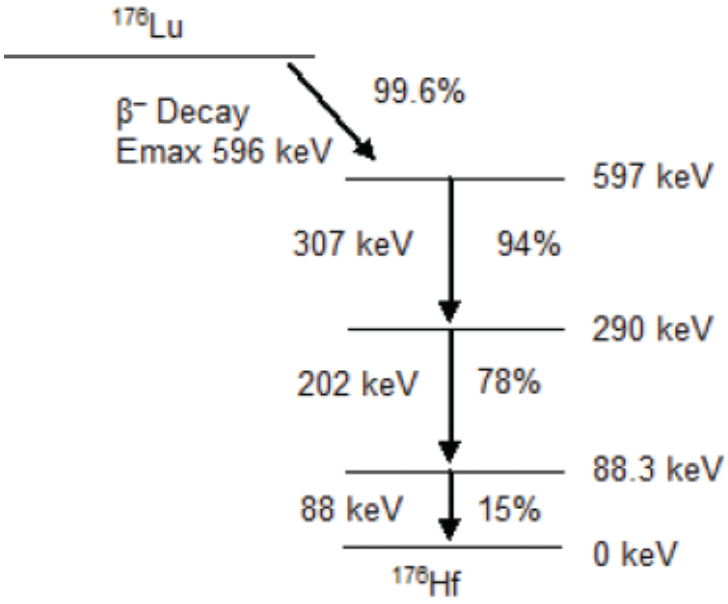
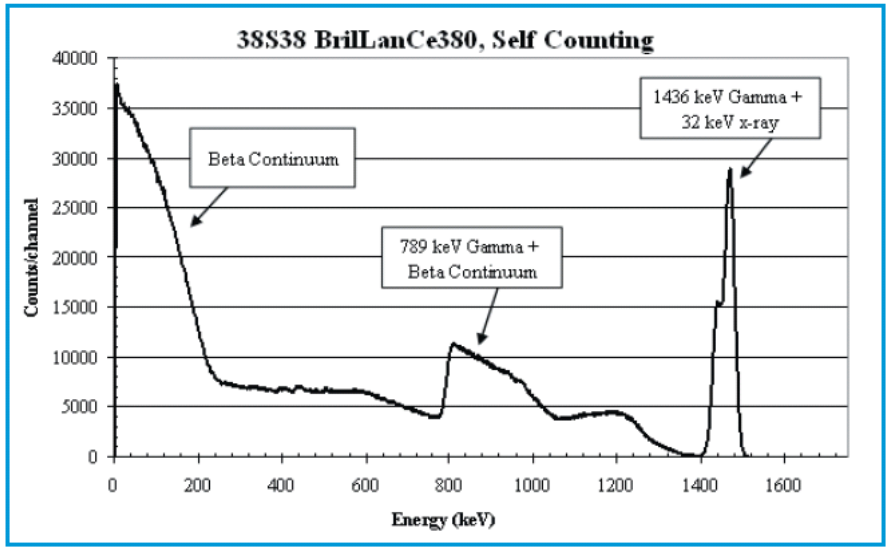


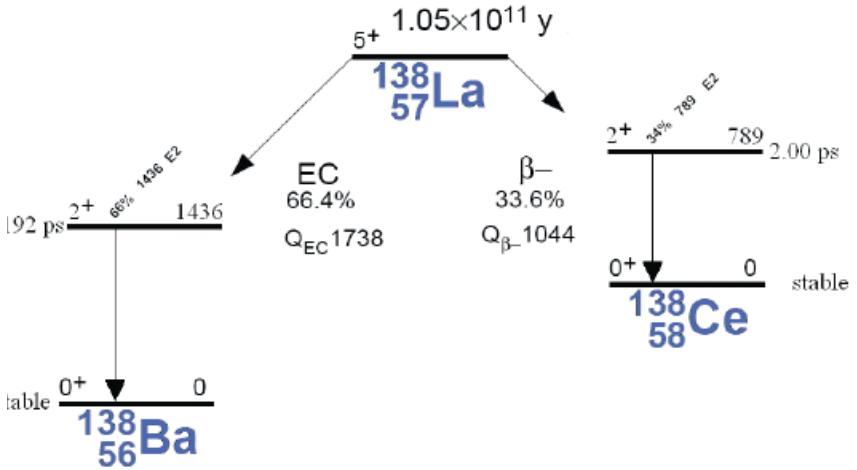
Fig.1 ^{176}Lu Decay Scheme

With abundance of 2.6% ^{176}Lu with half life of 3.8×10^{10} years an intrinsic activity of 290 cnts/cm^3 is obtained

^{176}Lu isotopes emit 89, 202, plus 307 keV γ-photons plus emission of a β^- with maximum energy of 596 keV (plus a neutrino).

Intrinsic activity of $LaBr_3$:Ce





With abundance of 0.089% ^{138}La with half life of 1.0x10^{11} years an intrinsic activity of 1.4 cnts/cm^3 is obtained

^{138}La emits either a 1.436 MeV gamma plus an 32 keV X-ray from Ba or it emits a β^- with E_{max}=255keV plus a789 keV gamma.

Final remarks

- Ideal scintillator does not exist
  - compromise based on the application
- High resolution for spectroscopy (isotope identification)
  - proportional scintillators
- extremely fast rise time for timing and TOF applications
- low intrinsic count rate for space explorations
- hight density at low cost
  - for calorimeters
- current topics
  - understanding non-proportionality
  - Eu^{2+} and Pr^{3+} activated scintillators
  - scintillator pulse rise time studies

Acknowledgements

- Some of the materials for these slides were provided by
  - Dr. P. Schotanus, company Scionix, The Netherlands
  - Dr. E. Mattmann, company Saint Gobain crystals, France
  - Dr. A. Owens and F. Quarati, European Space Agency, Netherlands
  - Dr. J Karp, Pennsylvanian State University, USA

References

- Glodo et al. IEEE TNS, 52 (2005) 1805.
- Radiation Detection and Measurements, G. Knoll, John Wiley&Sons, Inc
- Saint Gobain brochures on scintillators, http://www.detectors.saint-gobain.com/
- N. Cherepy, S.A. Payne, et al. IEEE Trans. Nucl. Sci. 56(3) (2009) 873
- I.V. Khodyuk, J.T.M. de Haas, P. Dorenbos, IEEE Trans. Nucl. Science 57(2010) 1175-1181
- G. Bizarri, P. Dorenbos, Phys. Rev. B. 75 (2007) 184302.
- P. Dorenbos, IEEE Trans. Nucl. Sci. 57 (2010) 1162-1167.
- P. Dorenbos, J.T.M. de Haas, C.W.E. van Eijk, IEEE Trans. Nucl. Sci. 51(3) (2004) 1289-1296.
- M.J. Weber, J. Lumin. 100 (2002) 35.

Thermal neutron scintillators

- Efficient thermal neutron detection
  - high capture cross section
    - suitable isotopes and isotope enrichment
  - absence of competing capture processes
- gamma ray back ground rejection
  - thin layers of low density
  - pulse height discrimination
  - pulse shape discrimination

Thermal neutron capture reactions

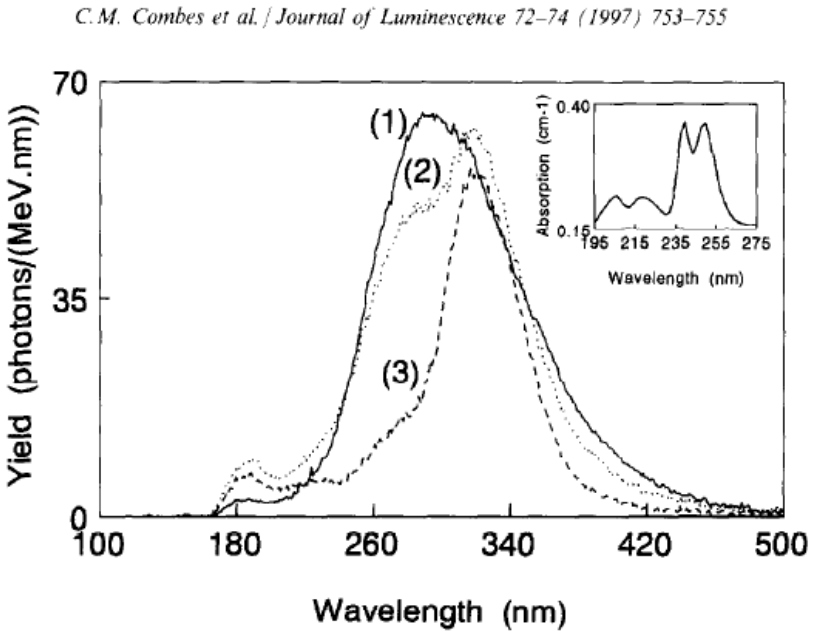
- $n + {}^{6}Li \rightarrow {}^{3}H + \alpha + 4.79$ MeV
- $n + {}^{10}B \rightarrow {}^{11}B \rightarrow {}^{7}Li + \alpha + 2.78$ MeV (7%)

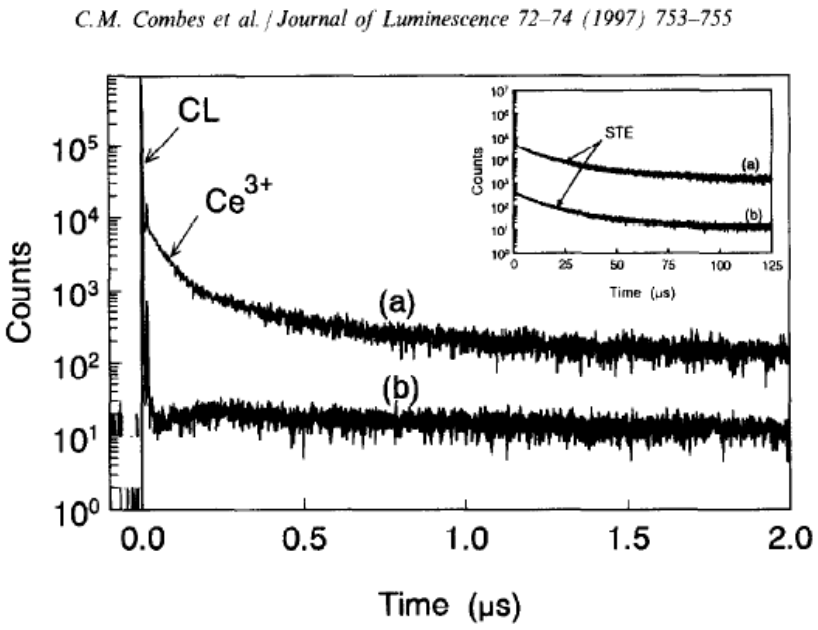
  ${}^{7}Li^{*} + \alpha + 2.30$ MeV (93%)

inorganic thermal-neutron scintillators

Scintill. host	Density ρ g/cm^3	Abs. Length at 1.8 Å mm	ρZ_{eff}^4 (x 10^6)	Dopant/ conc-mol%	λ_{em} nm	Light yield photons per neutron	Light yield photons per MeV gamma	τ ns
^{6}Li-glass	2.5	0.52		Ce	395	6.000	4.000	75
^{6}LiI	4.1	0.54	31	Eu	470	50,000	12,000	1.4 x 10^3
^{6}LiF/ZnS	2.6	0.8	1.2	Ag	450	160,000	75,000	10 x 10^3
$^6Li_6Gd(BO_3)_3$	3.5	0.35	25	Ce	385/415	40,000	20,000	200/800

Pulse shape discrimination due to core-valence luminescence in $LiBaF_3:Ce^{3+}$





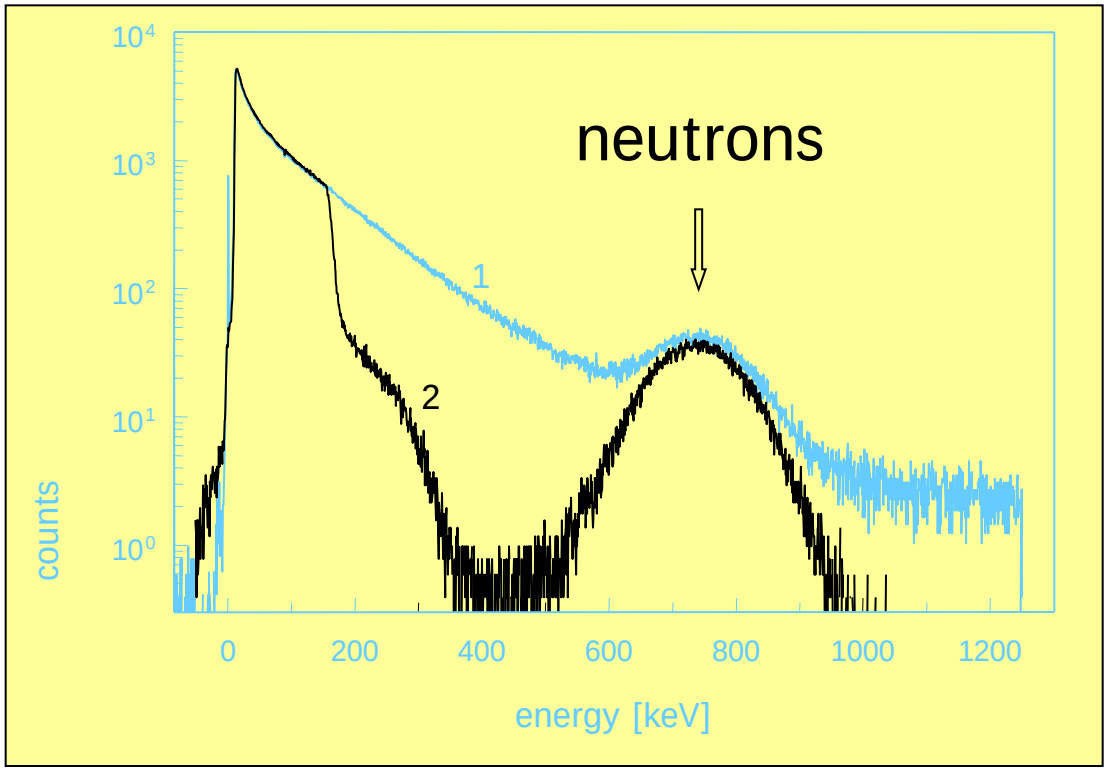
$LiBaF_3$:Ce scintillator

C.M. Combes *et al*, NIM A416(1998)364

CVL does not respond to

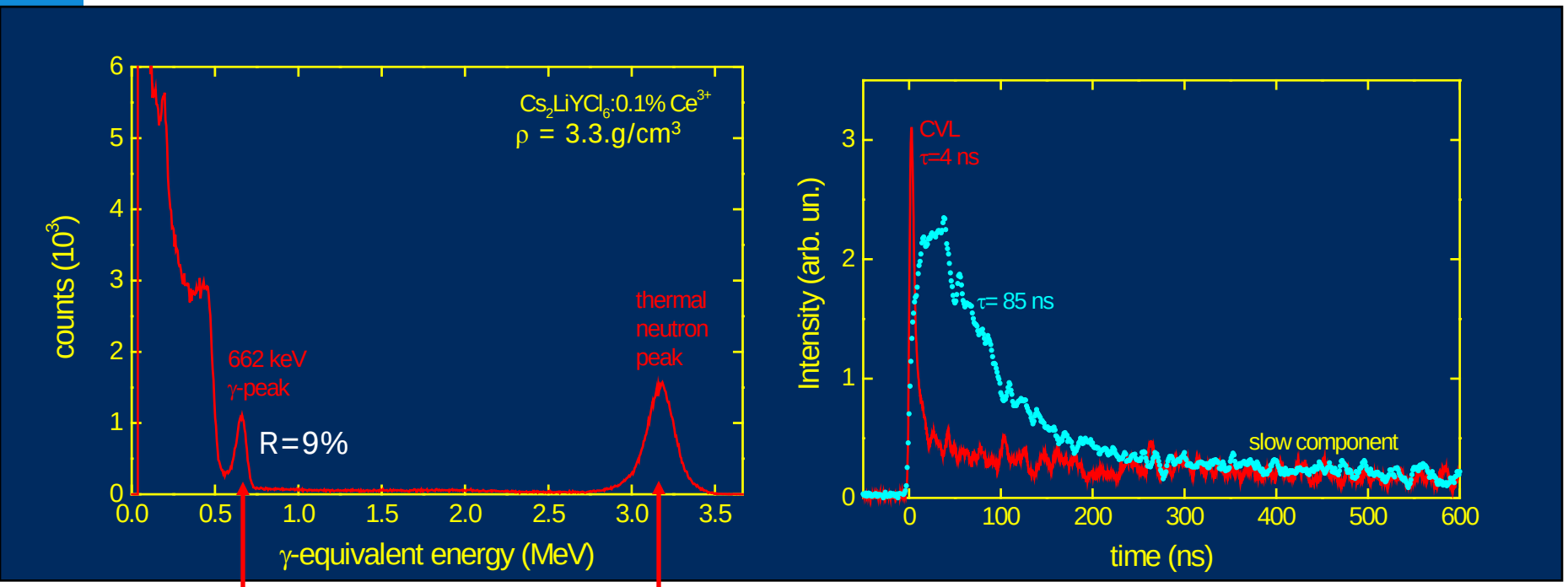
^3H (2.75 MeV) + ^4He (2.05 MeV)

Spectra of radiation emitted by a Pu-Be source shielded with paraffine and 6 cm of lead



1 is without discrimination, # 2 with discrimination. The neutron peak at 750 keV gamma-equivalent energy is almost free from background

Thermal neutron detection with Cs_2LiYCl_6:0.1% Ce^{3+}



22000 ph/MeV

70000 ph/neutron

α/β=0.66

pulse shape plus pulse height discrimination

Thermal neutron scintillators

Selection of the scintillator based on thermal neutron detection efficiency

σ (barn)	6Li - nat /%Σσ	6Li - 95enr /%Σσ	Absorption length 0.18 nm - 95enr (mm)
Cs_2LiYBr_6	71/41	895/90	3.7
Rb_2LiYBr_6	71/62	895/95	3.5
Li_2NaYBr_6	142/77	1790/98	1.7
K_2LiYBr_6	71/60	895/95	3.5
$Rb_2LiLaBr_6$	71/58	895/95	3.5
Cs_2LiLuI_6	71/29	895/84	3.9
Rb_2LiYI_6	71/65	895/96	3.4

Thermal neutron scintillation light yield and peak resolution $Rb_2LiYBr_6:Ce^{3+}$

Compounds	Photons/neutron	α/β ratio	Peak resolution %
Rb_2LiYBr_6: 0.1% Ce^{3+}	59,000 ± 5,900	0.74	3.6
Rb_2LiYBr_6: 0.5% Ce^{3+}	83,000 ± 8,300	0.75	5.4
Rb_2LiYBr_6: 1% Ce^{3+}	49,000 ± 4,900	0.79	8.5
	51,000 ± 5,100	0.82	4.2
Rb_2LiYBr_6: 5% Ce^{3+}	39,000 ± 3,900	0.74	5.8
	42,000 ± 4,200	0.80	7.8

Scintill. host	Density ρ g/cm^3	Abs. Length at 1.8 Å mm	λ_{em} nm	Light yield photons per neutron	Light yield photons per MeV gamma	α/β ratio	τ ns	Peak Resolution (%)
^{6}LiGlass:Ce	2.5	0.52	395	~6,000	~4,000	0.31	75	13-22 % 3.9 % -
$Cs_2LiYBr_6:Ce^{3+}$	4.1	3.7	389	73,000	20,000	0.76	89, 2.5 x 10^3	4.6 %
$Rb_2LiYBr_6:Ce^{3+}$	4.8	3.5	385	83,000	23,000	0.74	42,140 1.6 x 10^3	3.6 %
$Li_2NaYBr_6:Ce^{3+}$	2.9	1.7	380	39,000	12,400	0.66	30,560 2.6 x 10^3	-