ENERGY RESOLUTION AND NON-PROPORTIONALITY IN SCINTILLATORS

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γ-ray spectrometry with scintillation detectors

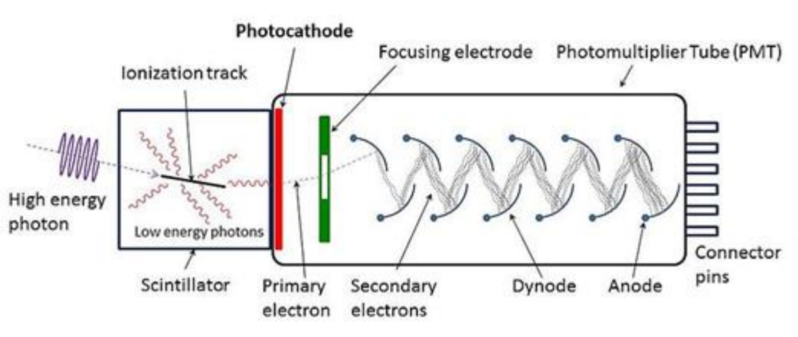
- γ-ray spectrometry with scintillation detectors is one of the most important methods in research and different applications of nuclear science

    - High-energy physics
    - Nuclear physics
    - Nuclear medicine
    - Environmental studies
    - Homeland security

γ-ray spectrometry with scintillation detectors

Detection process of γ-rays with a scintillator coupled to a photomultiplier (PMT):

- γ-ray absorption and light emission
- Light collection at the photocathode
- Production of photoelectrons
- Collection of photoelectrons
- Multiplication of photoelectrons by PMT dynodes



γ-ray spectrometry with scintillation detectors

PMT

high multiplication ☺

fast response ☺

low noise ☺

sensitive to external M field ☹

high voltage supply ☹

delicate ☹

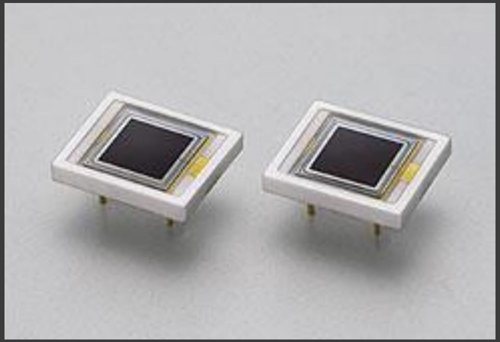
Other photoreceivers:

PIN diode, Avalanche Photodiode (APD), Silicon Photomultiplier (SiPM), Silicon Drift Detector (SDD)

Solid state devices have many practical advantages over the PMT, and this led to the PIN diode (for example) being used in applications where PMTs were too bulky or delicate, or where high voltages were not possible.

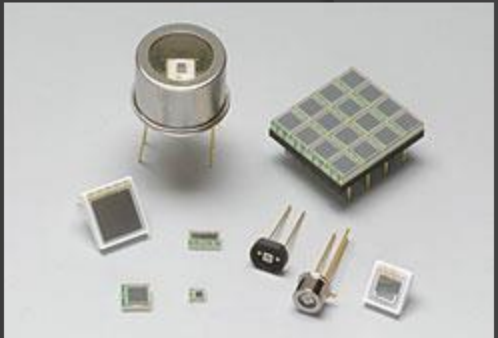
γ-ray spectrometry with scintillation detectors

high quantum efficiency ☺
moderate multiplication ☺
insensitive to external M field ☺
low voltage supply ☺
excess noise ☹
limited in size ☹



APD

SiPM



high multiplication ☺
insensitive to external M field ☺
low voltage supply ☺
moderate quantum efficiency **+/-**
dark count rate ☹
limited in size ☹

Advantages of scintillation detectors

- High detection efficiency for nuclear radiation
- Ability to measure energy spectra
- Detection at high rates up to 10^7 cps
- Superior time resolution (T-o-F, coincidence measurements)
- Ability to detect a wide assortment of radiations: γ and X-rays, charged particles, neutrons
- Great variety in size and constitution

γ-ray spectrometry with scintillation detectors

γ-ray spectrometry requires:

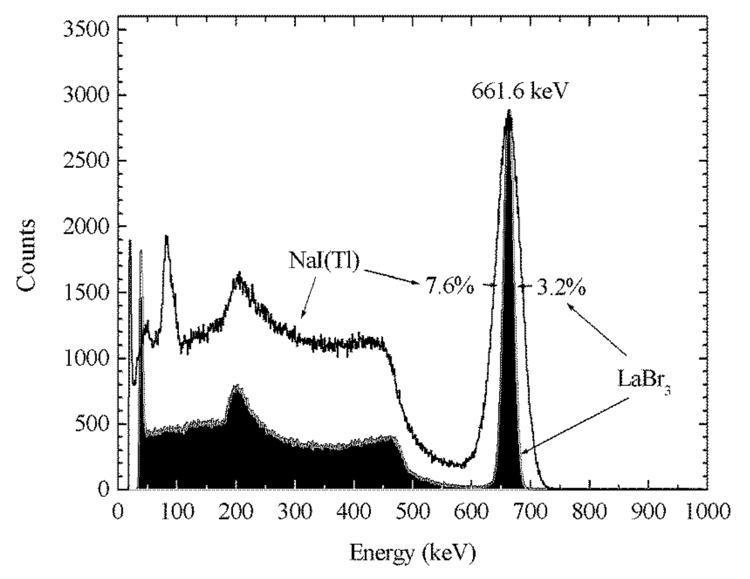
- High density of the material and a high atomic number of the major element which assure high detection efficiency of the γ-rays and a high photofraction
- High light output responsible for high statistical accuracy of the delivered signal
- Fast decay time of the light pulse reflecting the decay time of the fluorescence components of the crystal thus allowing measuring at high counting rates

γ-ray spectrometry with scintillation detectors

γ-ray spectrometry requires:

- Low contribution of scintillator to measured energy resolution, which is associated mainly with its non-proportionality characteristics.

Measured energy resolution is a function of the light output, but it is also affected by the internal properties of the scintillator.



γ-ray spectrometry with scintillation detectors

Light output is often quantified as a number of scintillation photons produced per MeV of deposited energy. It affects both the efficiency and the resolution of the detector.

Example:		
	NaI(Tl)	40 000 photons/MeV,
	BGO	8 000 photons/MeV

Non-proportionality of scintillator's response to γ-rays: means that *relative* light output decreases (or increases) as the γ-ray energy decreases.

γ-ray spectrometry with scintillation detectors

Integral or differential non-proportionality/nonlinearity

Linear response $b \neq 0$

$$L=a*E+b$$

Proportional response $b=0$

$$L=a*E$$

Derivative of both equations

$$dL/dE = a$$

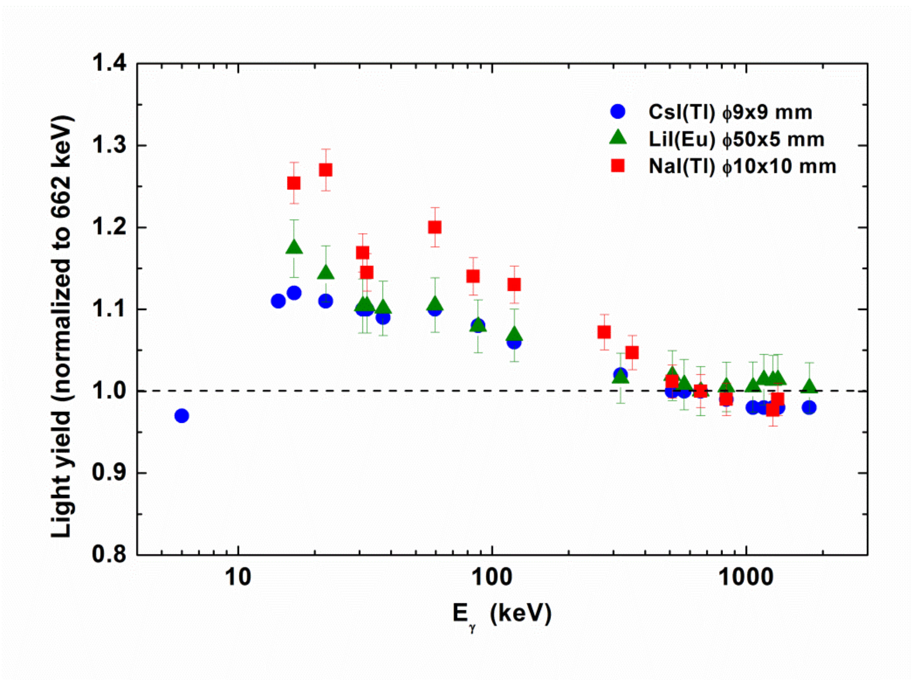
- It is not possible to directly measure this quantity (derivative) as it relates to scintillator light output ***L(E)***
- An **alternative technique** was adopted to present the non-proportionality. The technique is something between the integral (L vs. E) and differential ones.

γ-ray spectrometry with scintillation detectors

- Relationship **L(E)/E** = *a* is plotted to present this alternative approach. Deviations from the horizontal line *a* indicate light yield non-proportionality.
- In most cases the light yield per energy unit is normalized to the value measured at 662 keV from a ^{137}Cs calibration source.

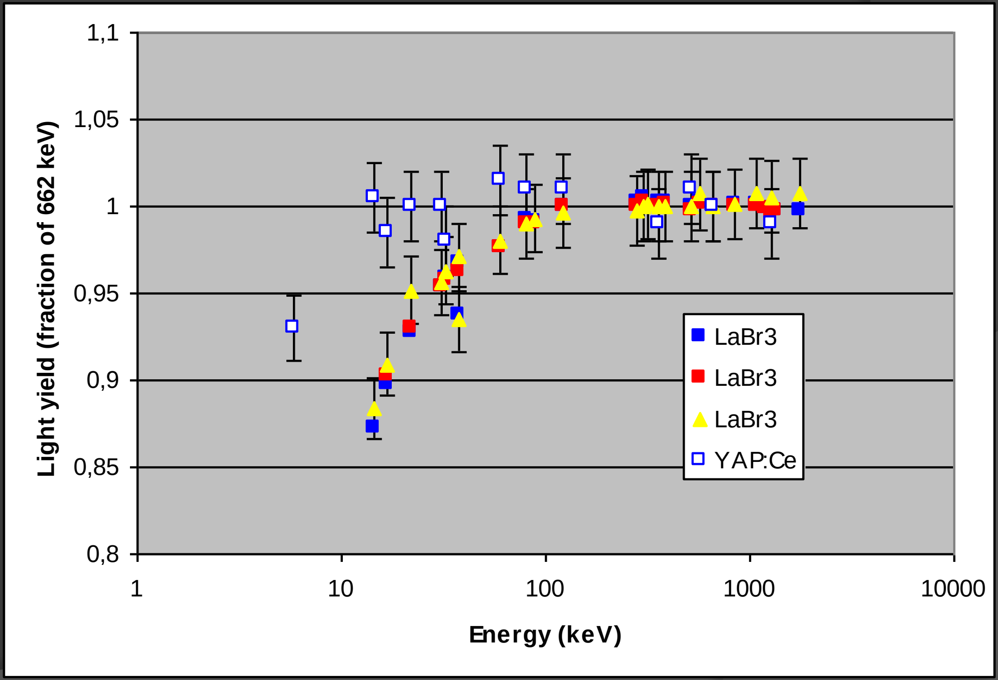
When plotting *L/E* versus *E*, it is imperative that the value of *L* be directly proportional to the scintillator light yield. If a multichannel pulse height spectrum is used for this purpose, the pulse amplitude must be corrected for the zero offset of the pulse processing instrumentation. The zero offset must be determined using specialized technique which is known to be absent of non-proportionalities.

Light yield non-proportionality of scintillator



Alkali halides
NaI(Tl), CsI(Tl), LiI(Eu)

$LaBr_3$:Ce, YAP:Ce



A scintillator coupled to a photodetector

Energy resolution, $\Delta E/E$, at a given energy is expressed as:

$$(\Delta E/E)^2 = (\delta_{sc})^2 + (\delta_{st})^2 + (\delta_p)^2 + (\delta_n)^2$$

δ_{sc} is intrinsic resolution of scintillator
δ_{st} is statistical contribution
δ_p is transfer resolution (*negligible*)
δ_n is noise contribution

γ-ray spectrometry with scintillation detectors

Intrinsic resolution δ_{sc} of a scintillator is affected by:

- Non-uniformity in the crystal
- Non-proportional response of the crystal
  - **γ-rays absorption:** secondary electrons due to different processes, as photoeffect and secondary X-rays or Auger electrons, Compton scattering, etc.
  - **scattering of secondary electrons (δ-rays)**
- Experimentally determined intrinsic resolution is also affected by inhomogeneities in the scintillator (local variations of L.O.) and the non-uniform reflectivity of the reflecting cover of the crystal

γ-ray spectrometry with scintillation detectors

The statistical uncertainty of the signal from the PMT

$$\delta_{st} = 2.35 \times 1/N^{1/2} \times (1 + \varepsilon)^{1/2}$$

where N – number of photoelectrons
ε – wariance of the PMT gain (0.1÷0.2)

The transfer component δ_p is described by variance associated with the probability that a photon from the scintillator results in the arrival of a photoelectron at the first dynode and then is fully multiplied by the PMT.

δ_p depends on:

- quality of the optical coupling of the crystal and PMT
- homogeneity of the quantum efficiency of the photocathode
- efficiency of photoelectron collection at the first dynode

γ-ray spectrometry with scintillation detectors

Intrinsic resolution of a scintillator **is the effect of its non-proportional response** to γ-rays

Landau's fluctuations

Statistical variations in dE/dx
for a slowing down electron =>
fluctuations in the energy deposition

δ-electrons

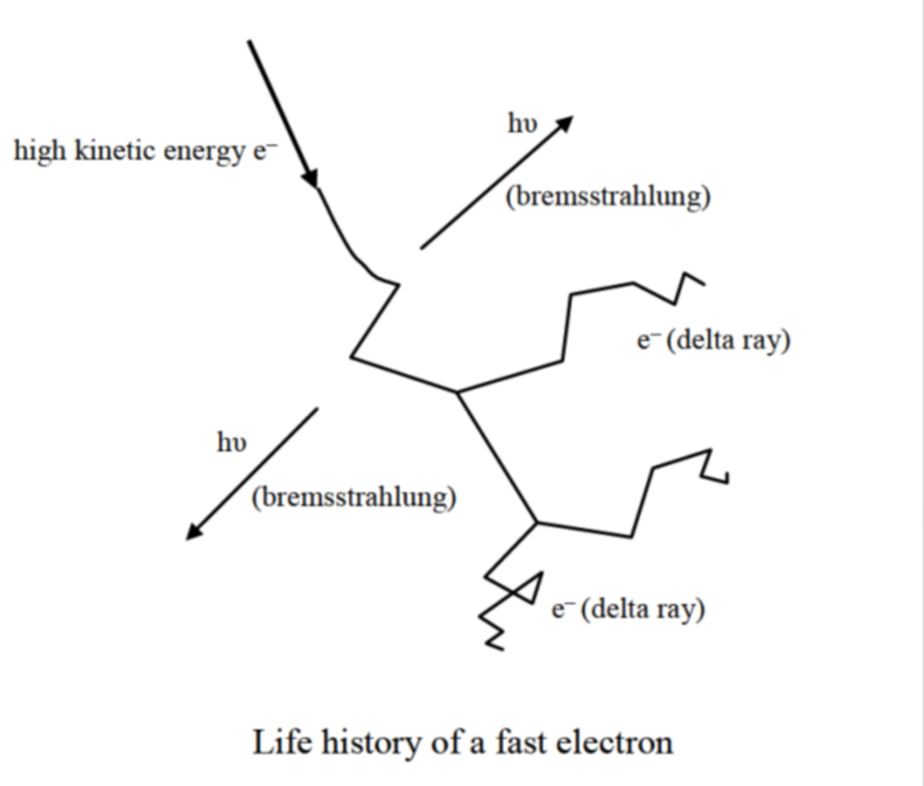
(secondary electrons)

Is it possible to affect non-proprtionality, thereby improving intrinsic resolution of scintillating materials ?

γ-ray spectrometry with scintillation detectors

Contribution of secondary electrons to the intrinsic resolution of scintillators (*Lukasz Swiderski, NCBJ*)

Primary electrons in matter often undergo large angle deflections loosing large part of energy and produce δ-rays



Life history of a fast electron

Important facts:

- primary electrons in scintillators usually have kinetic energy in the range:
1 keV < E < 1 MeV

- scattering plays a major role in the slowing-down process of primary electrons

γ-ray spectrometry with scintillation detectors

Correlation between NP and intrinsic resolution (at 662 keV)

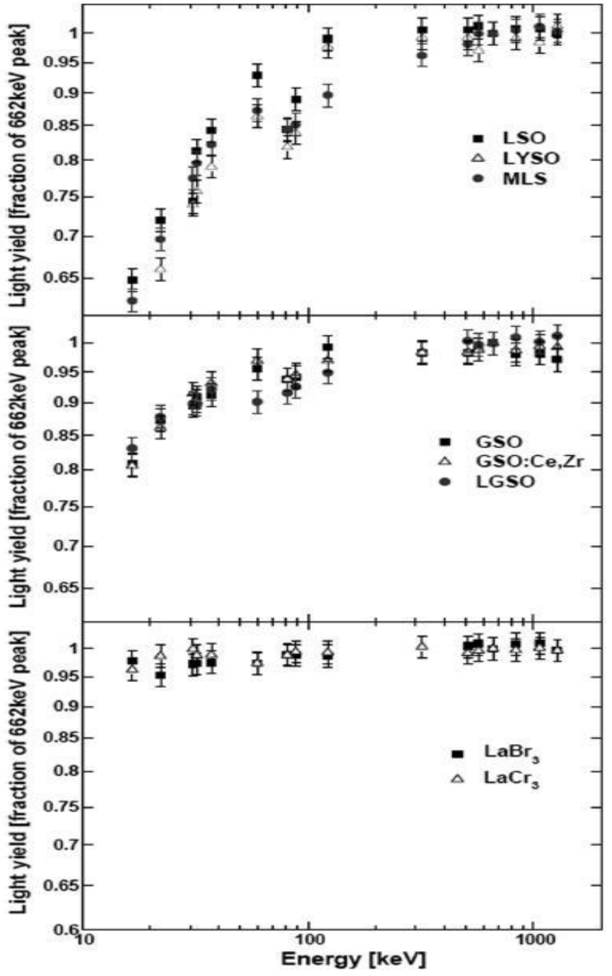


Fig. 1. Light yield as a function of the photon energy of the studied scintillators.

LSO, LYSO: 6.8 – 8%

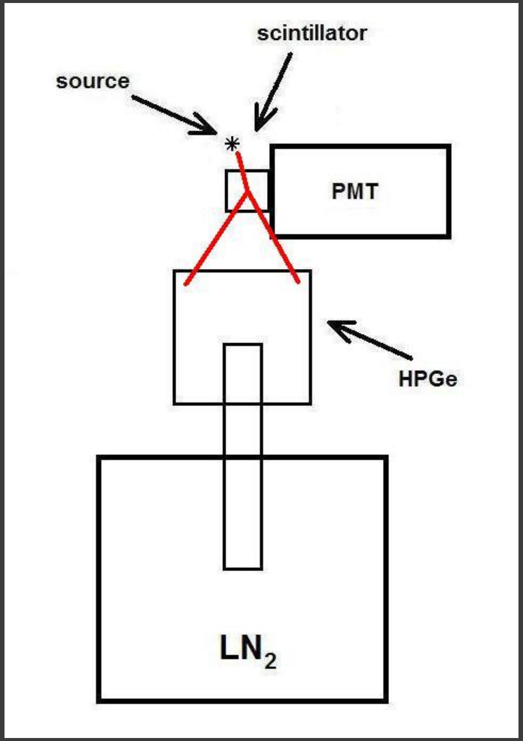
GSO, LGSO: 4.5 – 5.5%

$LaBr_3$, $LaCl_3$: 1.8 – 2.6%

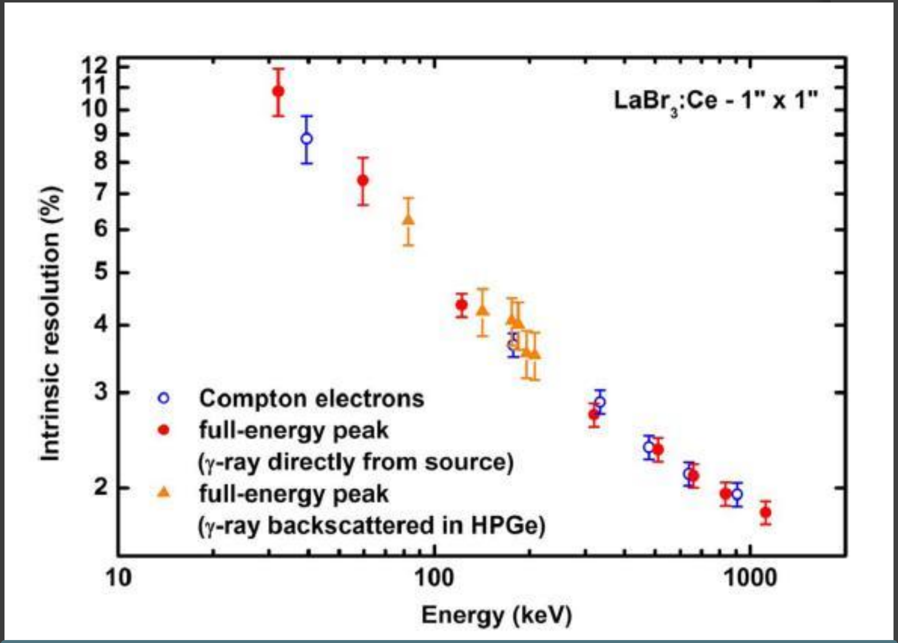
TNS 54 (2007) 3 – A. Nassalski et al.

γ-ray spectrometry with scintillation detectors

Wide Angle Compton Coincidence (WACC) technique developed in NCBJ



close geometry: ~ 1- 6 cm
large solid angle: ≤ 90º
weak sources: ~ 10 - 30 uCi



TNS 57 (2010) 1697 – L. Swiderski et al.

Intrinsic resolution data measured for Compton e^- (single interaction) **overlap** with gamma-rays (complex interaction) data

γ-ray spectrometry with scintillation detectors

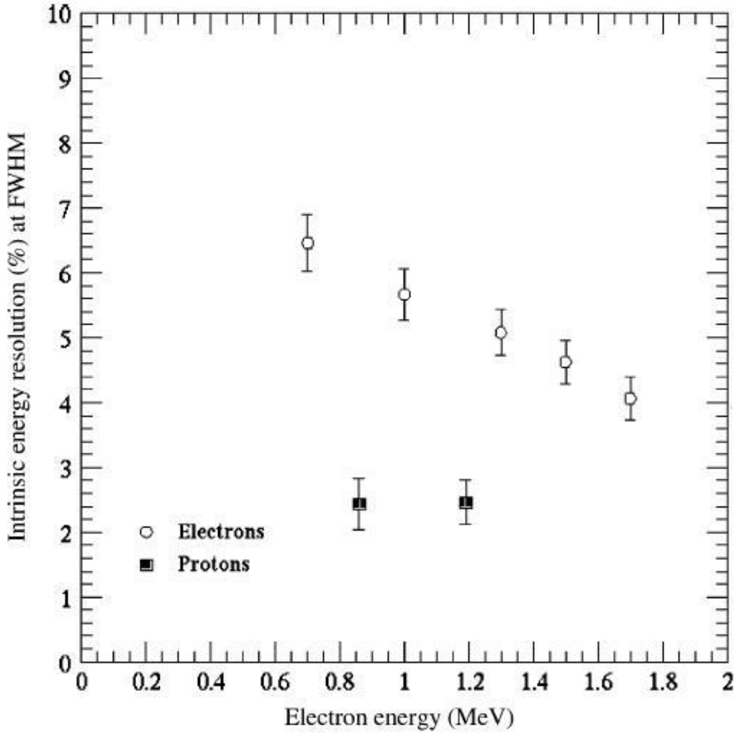


Fig. 14. Intrinsic energy resolution (%) at FWHM for electrons (open data points) and protons (filled data points) versus the electron energy (MeV). Proton energies of 2.8 MeV and 3.4 MeV are plotted on the equivalent electron energy scale.

TNS 55 (2008) 3717 – H.H. Vo et al.

Intrinsic resolution for protons considerably better than for electrons

⇓

Large impact of δ-rays generation on intrinsic resolution

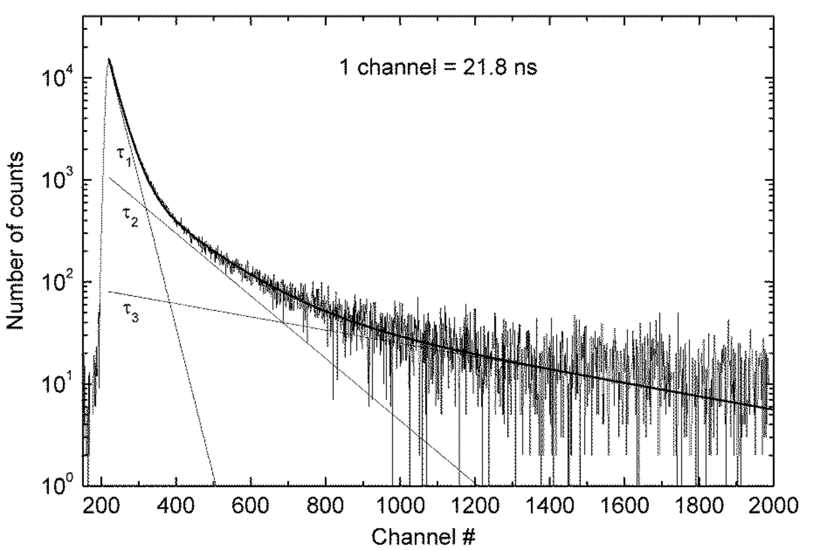
electrons: from magnetic spectrometer (≤1.8% at 1 MeV)

micro-beam of protons (≤1% at 1 MeV)

Time profile of the scintillation light

Studies on the non-proportionality and energy resolution of alkali halide scintillators showed an unexpected effect

=> the energy resolution is **affected by the slow component** of the light pulse.



CsI(Tl) – three scintillation components

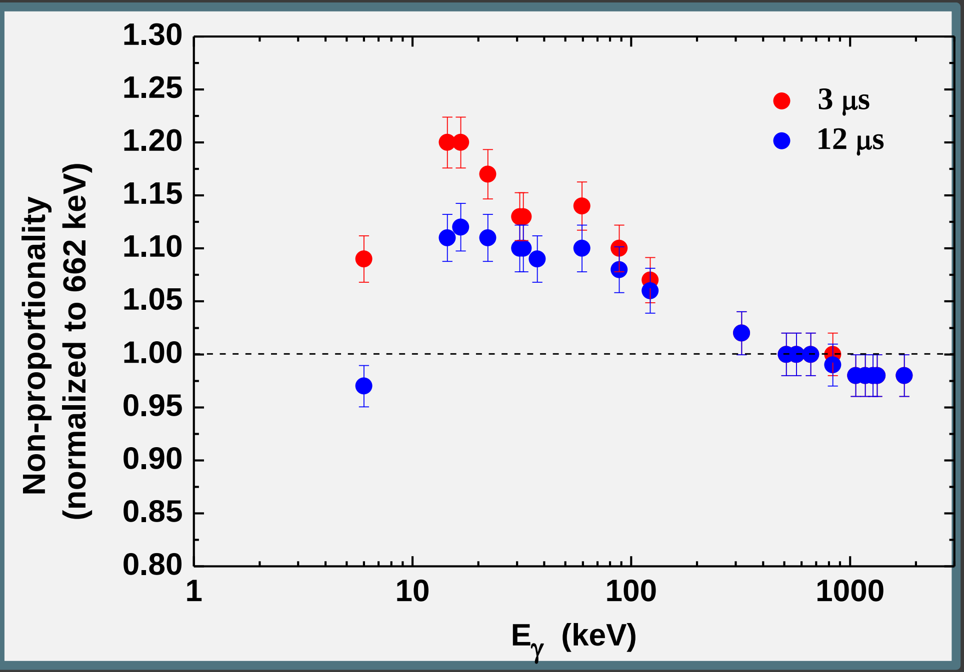
τ_1 – fast ~ 0.8 μs

τ_2 – slow ~ 4 μs

τ_3 – „tail” ~ 18 μs

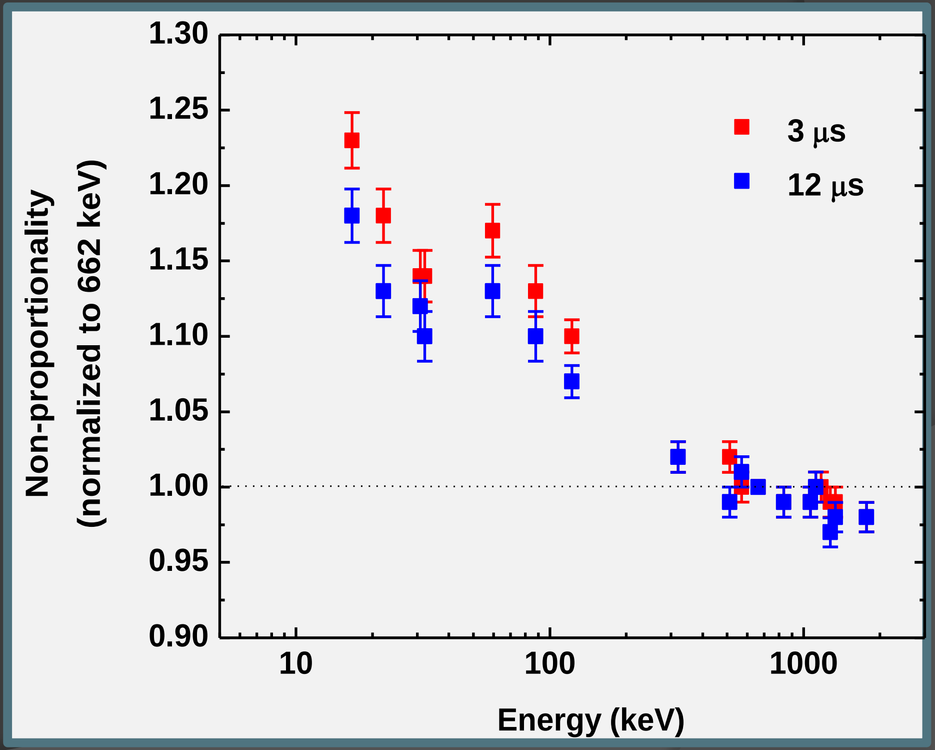
γ-ray spectrometry with scintillation detectors

Non-proportionality measured at two different integration time at the amplifier: 3 μs and 12 μs



CsI(Tl)
ϕ 9mm × 9mm

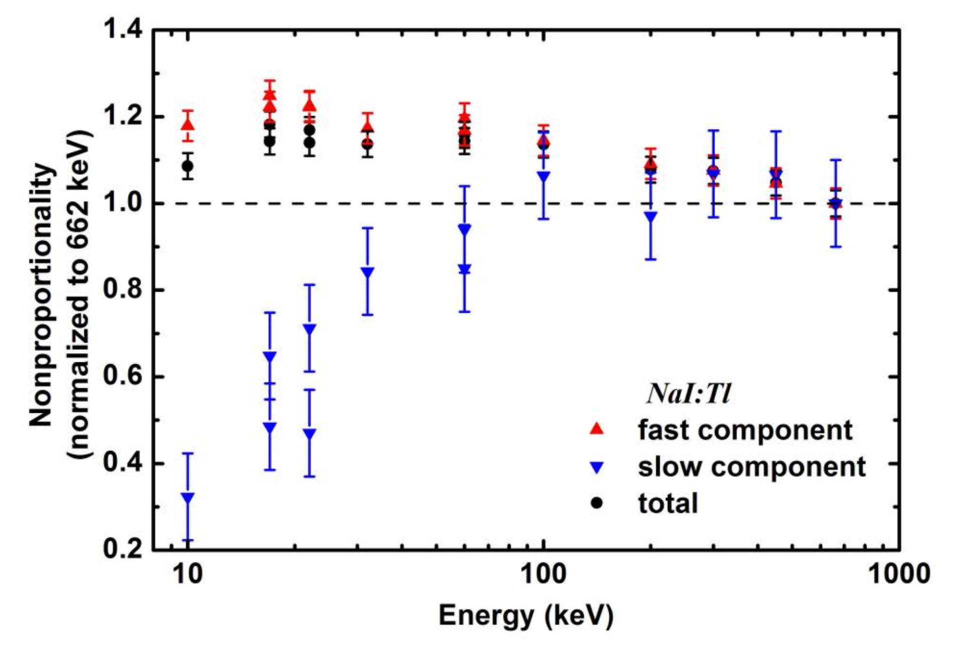
CsI(Na)
ϕ 1” × 1”



γ-ray spectrometry with scintillation detectors

Significant progress in understanding the observed effects in alkali halide crystals was achieved due to measurements of **non-proportionality of separate components of the light pulse.**

NaI(Tl) – two scintillation components



L. Swiderski et al. NIM A 749 (2014) 68

$I_{fast} \approx 90\%$ $\tau_{fast} \approx 0.23\ \mu s$

$I_{slow} \approx 10\%$ $\tau_{slow} \approx 1\ \mu s$

$$I_{tot} = A_1 * \tau_1 + A_2 * \tau_2$$

Non-proportionality defined as:

I_{tot} per energy unit for a given energy normalized to I_{tot} per energy unit obtained for 662 keV

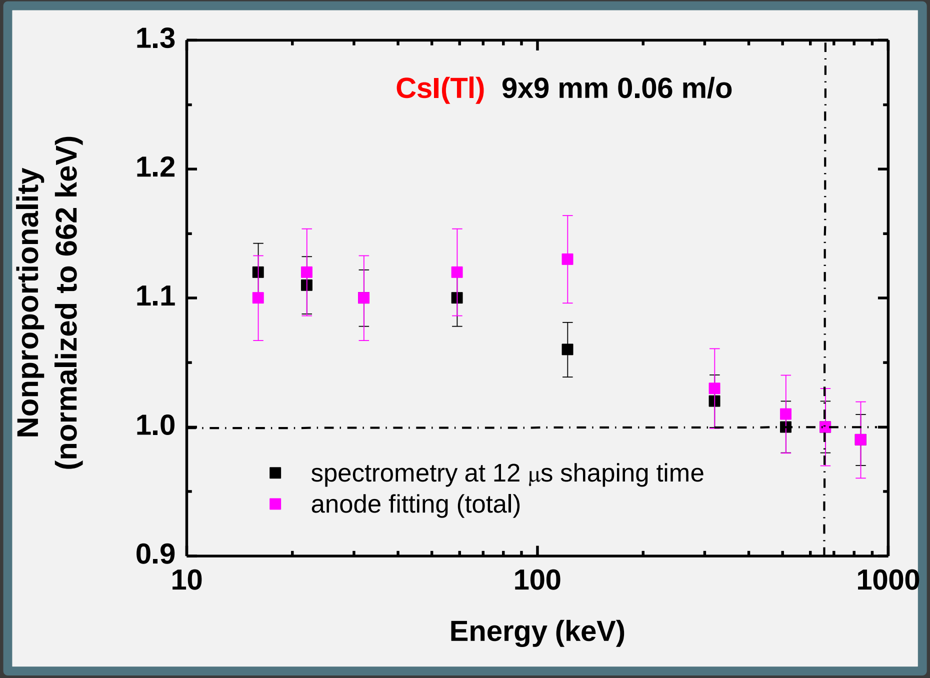
γ-ray spectrometry with scintillation detectors

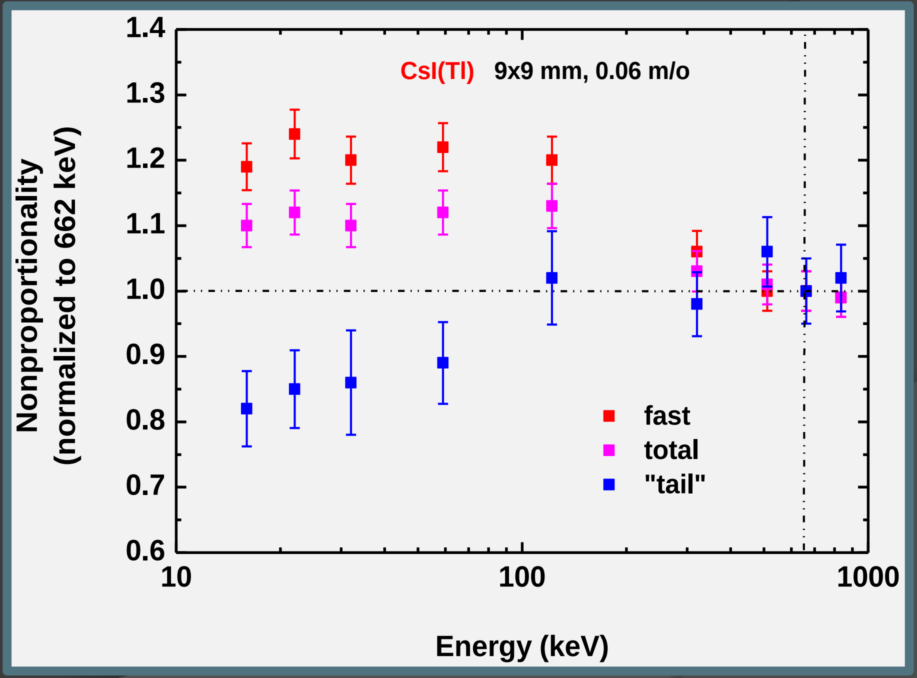
CsI(Tl) – three scintillation components

τ_1 – fast ~ 0.8 μs *48%*

τ_2 – slow ~ 4 μs *30%*

τ_3 – „tail" ~ 18 μs *22%*





γ-ray spectrometry with scintillation detectors

- **In contrast**, some other crystals such as LuAG:Pr, CsI(In) and different samples of undoped NaI (at liquid nitrogen temperature) showed a deterioration of the energy resolution at longer integration of scintillation. This was particularly pronounced in the case of the undoped NaI crystals.

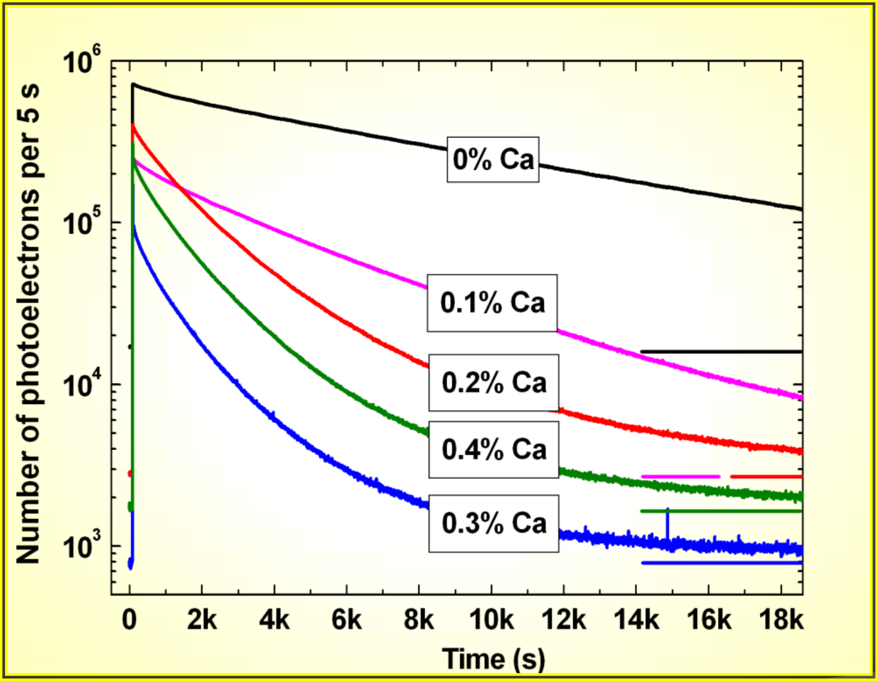
- Intense slow components of the light pulses of undoped NaI seriously affect the intrinsic resolution and are a cause of the limitation of energy resolution

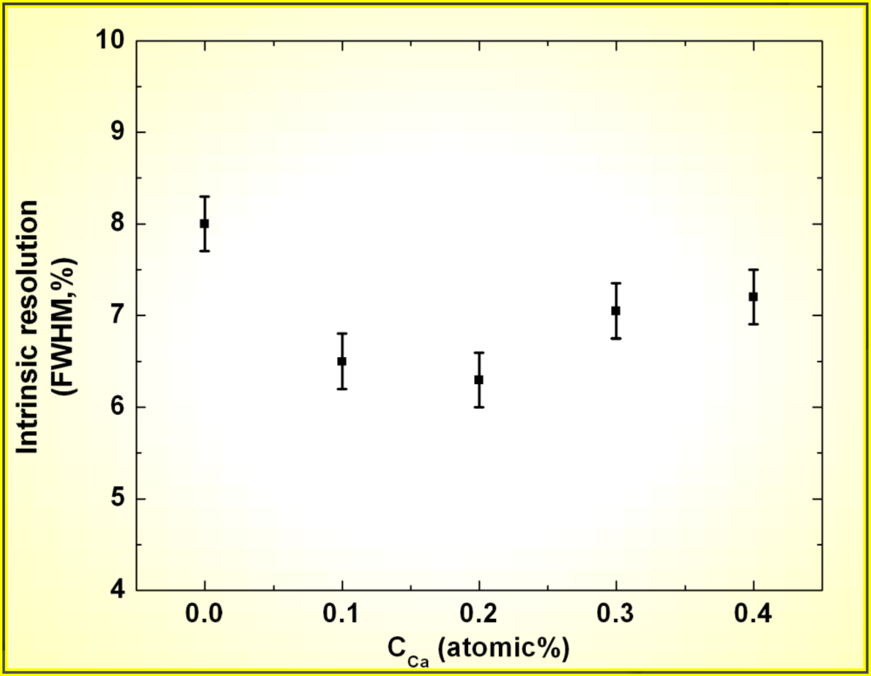
- Afterglow-like slow components ?

γ-ray spectrometry with scintillation detectors

LSO:Ce (0.1%) co-doped with **Ca^{2+}** ions

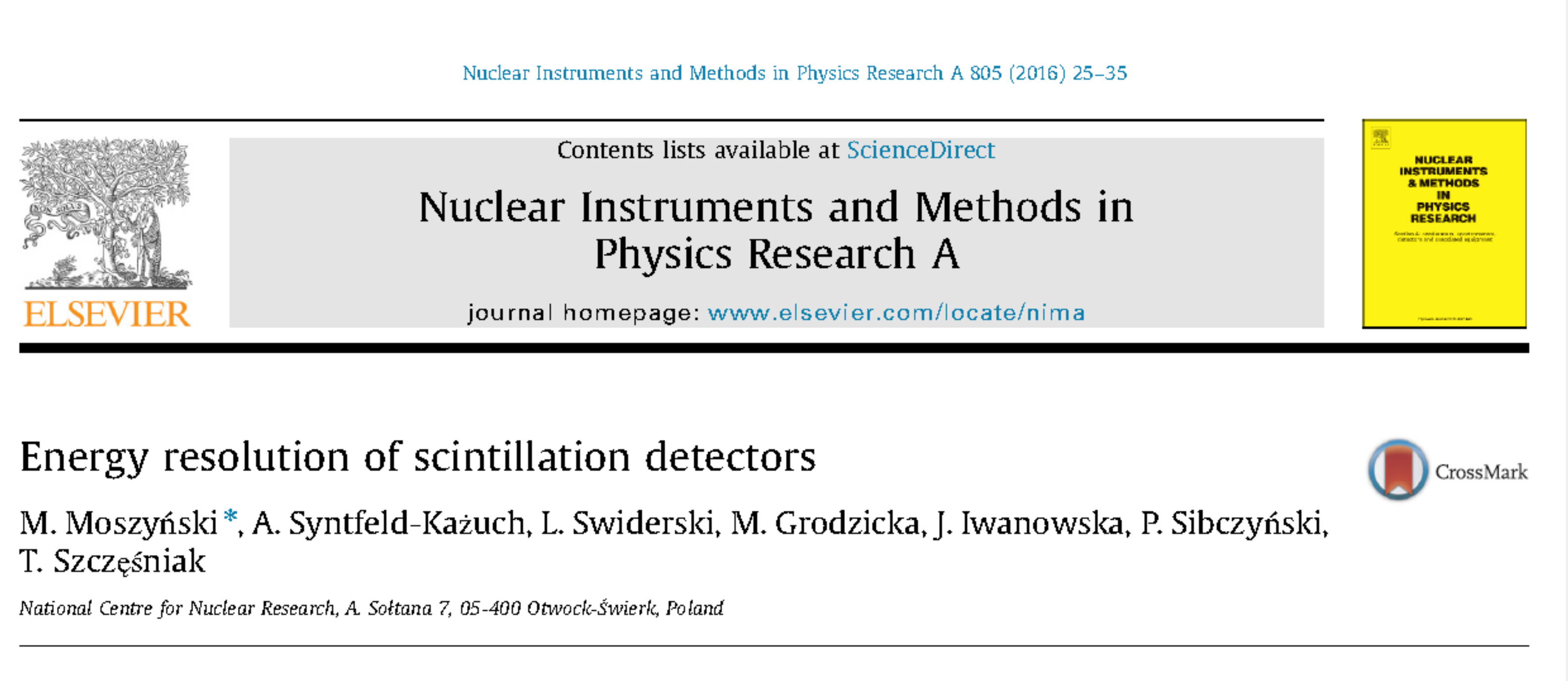
- increase of light output by about 30% for co-doped samples
- substantial improvement of energy resolution for co-doped samples
- Ca co-dopant reduces intensity of afterglow and shortens the effective decay time. No change in NP shape.





CONCLUSIONS

- In spite of substantial progress in the development of new scintillators, a complete understanding of finite energy resolution measured with different crystals has not been achieved yet.
- Non-proportionality of undoped oxide crystals such as BGO, CaWO and CWO represents the fundamental properties of scintillator materials and limits the energy resolution.
- In the case of doped crystals, particularly for alkali halides, the non-proportionality depends on the doping agents.



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Energy resolution of scintillation detectors

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Thank you for your attention!