

LaBr₃:Ce and new scintillators, status and perspectives

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Outline:

Old and New Scintillators.

Test, measurements and R&D:

- CeBr₃:Ce, SrI₂ and GYGAG
- CLYC
- LaBr₃:Ce

Perspective of Co-Doping.

Scintillator in SPES facility and LOIs.

Evolution of scintillators

First Generation 'old' scintillators

- NaI ⇒ acceptable energy resolution, strong non linearity in energy, bad time resolution
BaF₂ ⇒ bad energy resolution, excellent time resolution
BGO ⇒ bad energy resolution, bad time resolution, excellent efficiency
CsI ⇒ good for the measurement of light charged particles

Second Generation 'new' scintillators

- New Materials: ⇒ SrI₂:Eu, CeBr₃ ⇒ **Co Doped** CeBr₃:Ce - Ca⁺⁺
Elpasolite : ⇒ CLYC, CLLB, CLLC
Ceramic ⇒ GYGAG
Lanthanum Halide ⇒ LaBr₃:Ce, LaCl₃:Ce ⇒ **Co Doped** LaBr₃:Ce - Sr⁺⁺

Composite Detectors

- PARIS (LaBr₃:Ce + NaI) array (see Adam Maj talk)
- take the best from different scintillators

Performances of new scintillators (SrI_2 , CeBr_3 , GYGAG)

SrI_2

Slow scintillator (τ on μs scale) with self absorption but with excellent energy resolution $< 3\text{-}4\%$ @ 662 keV. It is available

CeBr_3

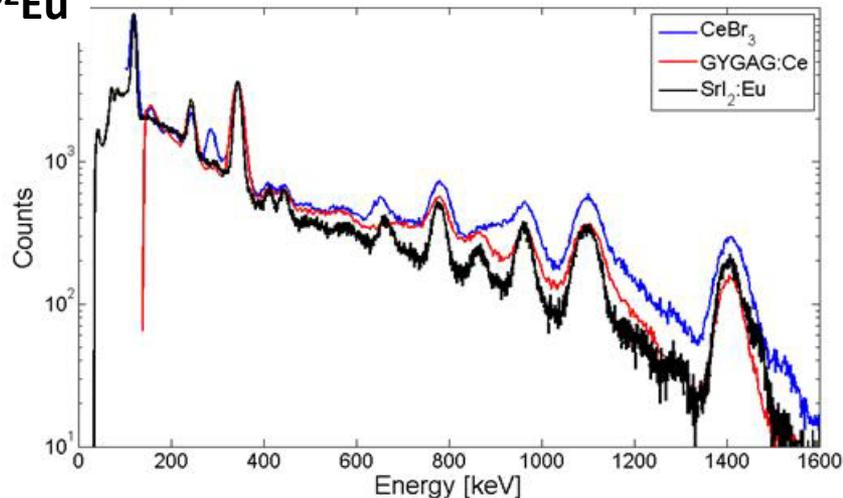
Fast scintillator (subnanosecond time resolution)
Good Energy resolution ($< 5\%$ @ 662 keV)
No internal radiation
It is available up to 3"x3" (CoDoping developed)

GYGAG

$\text{Gd}_{1.5}\text{Y}_{1.5}\text{Ga}_{2.2}\text{Al}_{1.8}\text{O}_{12}$ (Ce)
Transparent polycrystalline ceramic scintillators
Density and Z_{eff} of GYGAG are 5.8 g/cm^3 and 48,
Good Energy resolution with SDD ($< 5\%$ @ 662)
Fast scintillator (rise time $\sim 30\text{ ns}$)



^{152}Eu



Spectra Acquired in Milano with a ^{152}Eu source using :

- Cylindrical 2" x 2" SrI_2
- Cylindrical 2" x 3" CeBr_3
- Cylindrical 0.3" x 2" GYGAG

- All detectors have an energy resolution better than that of NaI

Work done together with G.Hull Detectors from Livermore and IPN Orsay

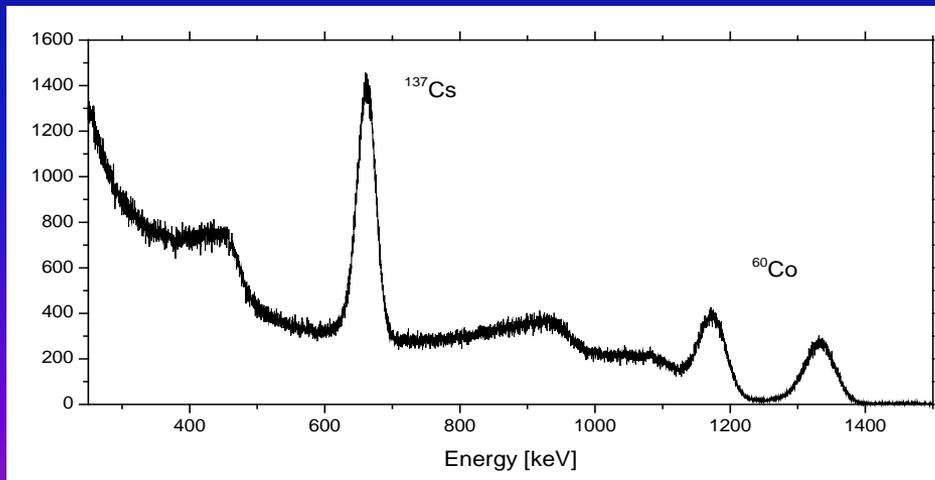
Spectra from A.Giaz

General Performances of new scintillators (CLYC)

CLYC →

Material	Cs ₂ LiYCl ₆ :Ce	Peak Scintillation Wavelength	370 nm
Melting Point	640°C	Decay Constants (CVL, Ce3+, Ce-STE).....	1 ns, 50 ns, 1000 ns
Density	3.31 g/cm ³	Scintillation Light Yield.....	20,000 ph/MeV
Crystal Structure	Cubic	X-ray Absorption Coef. at 100 KeV.....	3.97 cm ⁻¹
Cleavage planes	None	X-ray Absorption Coef. at 662 KeV.....	0.251 cm ⁻¹
Water Solubility	Hygroscopic	Radiation Length	3.42 cm
Refractive index	1.81 ± 0.037 @ 405 nm	Heat Capacity	0.379 J/(g*K)
Coefficient of Thermal Expansion	34.34x10 ⁻⁶ /°C @ 30°C	Thermal Conductivity	0.0067 W/(cm*K) at 50°C
Emission Spectral Range	275 – 450 nm		

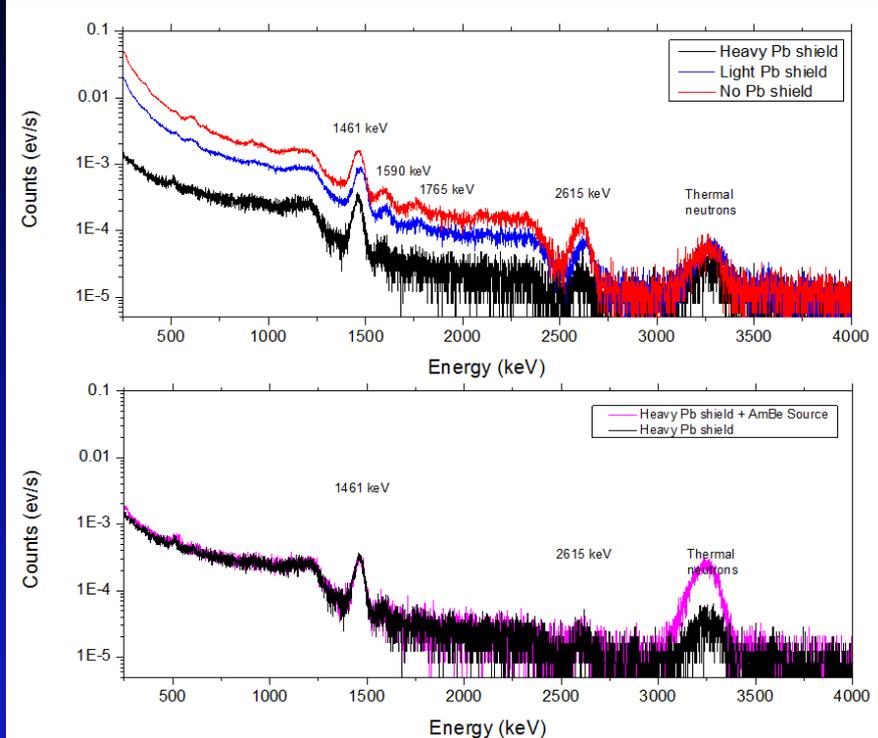
- Fast Risetime (for γ -rays only) but long fall time
 - Few ns (the CVL lighth is, as for BaF₂, in the UV region)
- Good energy resolution
 - FWHM < 5% at 662 keV



Energy spectrum from a ¹³⁷Cs and ⁶⁰Co Source measured in Milano



CLYC and Internal Radiation



Measurements performed in Milano using a 95% enriched ${}^6\text{Li}$ 1"x1" CLYC scintillator

Measurement of the internal radiation

- Red spectrum -> no shield
- Blue spectrum -> light Pb shield
- Black spectrum -> heavy Pb shield

Internal radiation is not be affected by any kind of shield

Thermal Neutrons are weakly affected by Pb shield

- Black spectrum -> heavy Pb shield
- Purple spectrum -> as black but with a AmBe source at few meter distance

The internal radiation is practically absent in CLYC

We have measured that the internal radiation is weaker than 0.02 events/cm^3

CLYC and Thermal Neutrons

If ${}^6\text{Li}$ enriched CLYC has very high sensitivity to thermal neutrons

- ${}^6\text{Li}(n,\alpha){}^3\text{H}$
- Cross Section for thermal neutrons is 980 Barns
- The sensitivity to thermal neutrons of CLYC is higher than that of ${}^3\text{He}$

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CLYC:Ce and PMT Properties



$\text{Cs}_2\text{LiYCl}_6:\text{Ce}$ (95% ${}^6\text{Li}$ enrichment)

Main Emission:

- $\text{Ce}^{3+}/\text{STE}$ located between 350 to 450 nm

Secondary Emission:

- CVL located between 250 and 350 nm

Decay Constant

- (CVL, Ce^{3+} , Ce-STE): 1 ns, 50 ns, 1000 ns

Scintillation Light Yield: 20 ph/keV



PMT R2059

Material Window:

- Quartz (160 nm – 650 nm)

Rise Time: 1.3 ns @ 2500V

Blue Sensitivity Index: 9.8

Timing PMT with Quartz Window



PMT R6231-100 mod

Material Window:

- Quartz (160 nm – 650 nm)

Rise Time: 8.5 ns @ 1000V

Blue Sensitivity Index: 14

Spectroscopy PMT with Quartz Window



PMT H6533

Material Window:

- Borosilicate Glass (300 nm – 650 nm)

Rise Time: 0.7 ns @ -2250V

Blue Sensitivity Index: 10.4

Timing PMT with Borosilicate Glass Window



PMT R6233

Material Window:

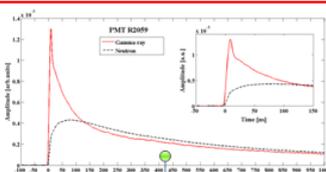
- Borosilicate Glass (300 nm – 650 nm)

Rise Time: 9.5 ns @ 1000V

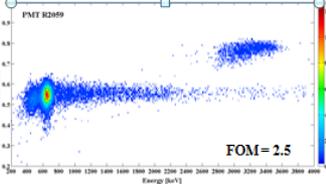
Blue Sensitivity Index: 11.7

Spectroscopy PMT with Borosilicate Glass Window

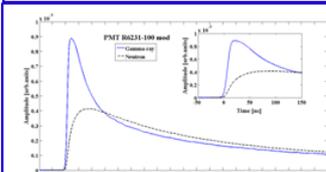
PMT R2059 – HV -2000V



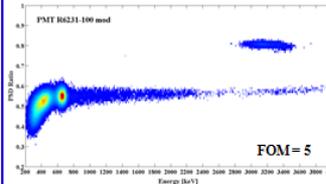
Gamma-ray - Rise time= 8 ns Fall Time= 792 ns
Neutron - Rise time= 33.5 ns Fall Time= 3169 ns



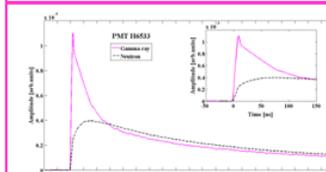
PMT R6231-100 mod – HV +900V



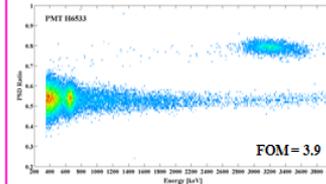
Gamma-ray - Rise time= 13 ns Fall Time= 1221 ns
Neutron - Rise time= 41 ns Fall Time= 3432 ns



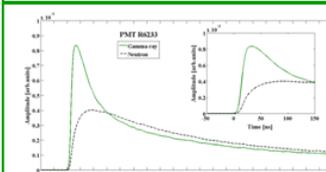
PMT H6533 – HV -1800V



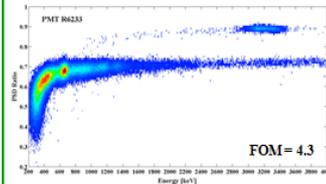
Gamma-ray - Rise time= 8 ns Fall Time= 991 ns
Neutron - Rise time= 35.5 ns Fall Time= 3684 ns



PMT R6233 – HV +900V



Gamma-ray - Rise time= 14.5 ns Fall Time= 1264 ns
Neutron - Rise time= 41.5 ns Fall Time= 3333 ns



These tests have been done in Milano using a 95% ${}^6\text{Li}$ enriched CLYC and a close AmBe source

CLYC and Thermal Neutrons

If ${}^6\text{Li}$ enriched CLYC has very high sensitivity to thermal neutrons

- ${}^6\text{Li}(n,\alpha){}^3\text{H}$
- Cross Section for
- The sensitivity to



$\text{Cs}_2\text{LiYCl}_6:\text{Ce}$ (95% ${}^6\text{Li}$ enrichment)

Main Emission:

- $\text{Ce}^{3+}/\text{STE}$ located between 350 to 450 nm

Secondary Emission:

- CVL located between 250 and 350 nm

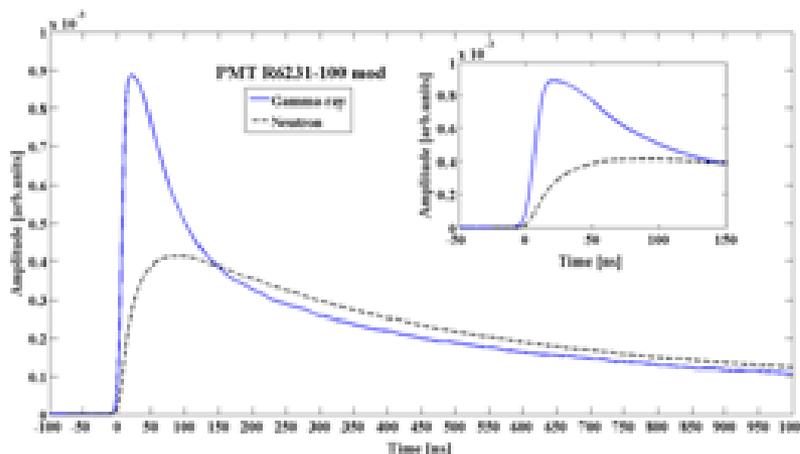
Decay Constant

- (CVL, Ce^{3+} , Ce-STE): 1 ns, 50 ns, 1000 ns

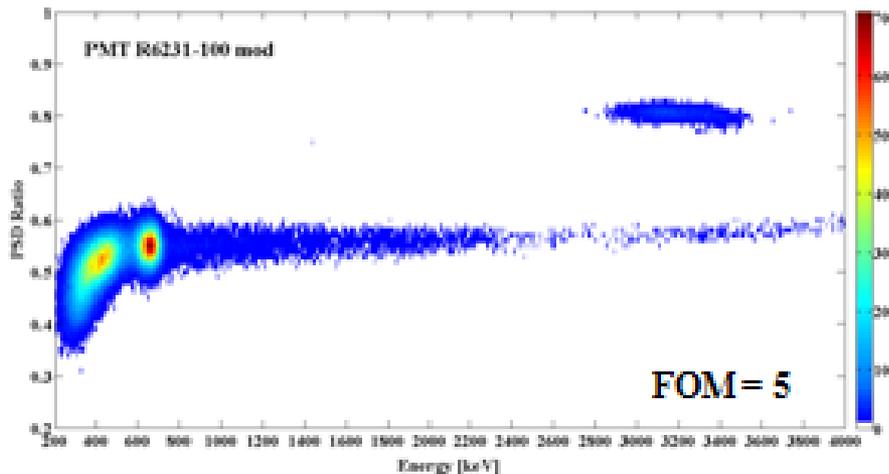
Scintillation Light Yield: 20 ph/keV

These tests have been done in Milano using a 95% ${}^6\text{Li}$ enriched CLYC and a close AmBe source

PMT R6231-100 mod – HV +900V



Gamma-ray - Rise time= 13 ns Fall Time= 1221 ns
Neutron - Rise time= 41 ns Fall Time= 3432 ns



${}^3\text{He}$



33

n – 650 nm)

V

osilicate Glass



PMT R6233

Material Window:

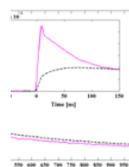
- Borosilicate Glass (300 nm – 650 nm)

Rise Time: 9.5 ns @ 1000V

Blue Sensitivity Index: 11.7

Spectroscopy PMT with Borosilicate Glass Window

V-1800V

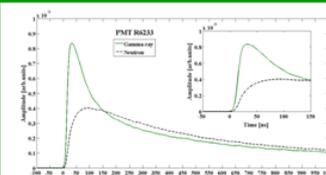


Fall Time= 991 ns
all Time= 3684 ns

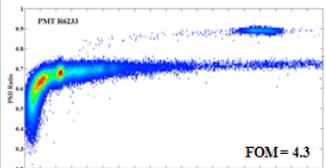


FOM= 3.9

PMT R6233 – HV +900V



Gamma-ray - Rise time= 14.5 ns Fall Time= 1264 ns
Neutron - Rise time= 41.5 ns Fall Time= 3333 ns



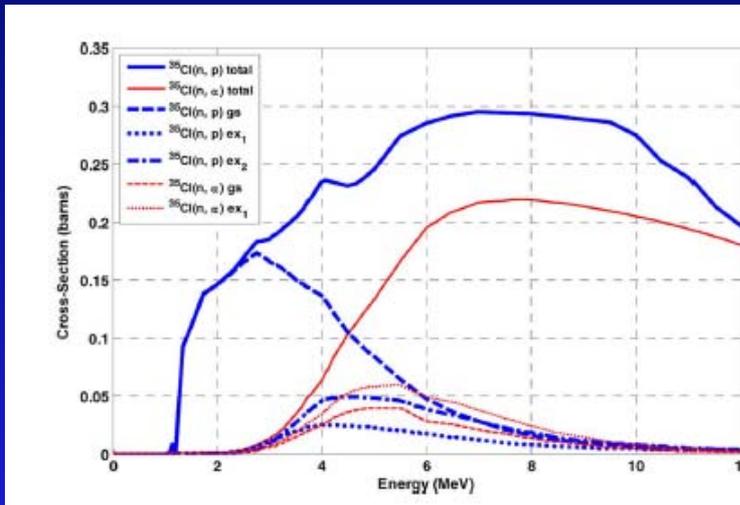
FOM= 4.3

CLYC and Fast Neutrons

It has been found that the proton energy released in the $^{35}\text{Cl} + n \Rightarrow ^{35}\text{S} + p$ reaction varies linearly with the energy of the incoming neutron, enabling fast-neutron spectroscopy via well defined peaks in the pulse-height spectrum. The kinetic energy of the neutron can therefore be measured

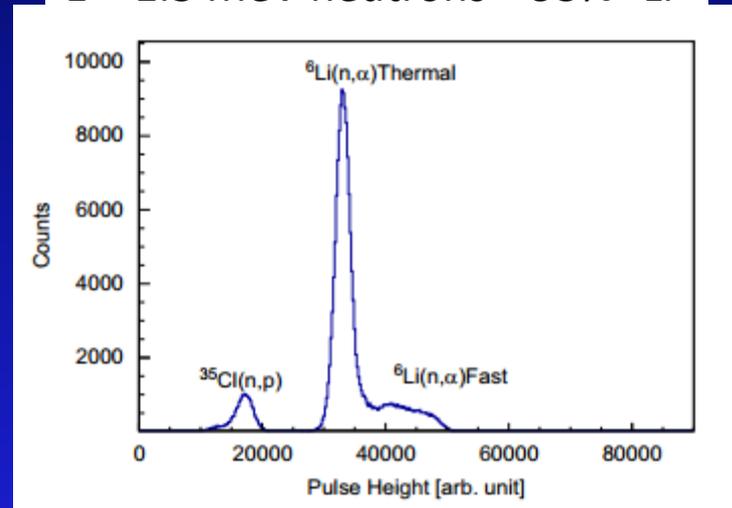
- Via the time signal using Time of Flight (FWHM < 1 ns)
- Via the energy signal

It is the only detector capable of that



Smith, Martin B et al IEEE TNS , VOL. 60 pg. 855 (2012)

E = 1.3 MeV neutrons 95% ^6Li



N. D'Olympia et al NIM A 714 (2013) 121-127

If ^7Li enriched CLYC has 10 times less sensitivity to thermal neutrons

If ^7Li enriched CLYC has an excellent sensitivity to fast neutrons

- Thermal neutrons produce a less intense background between 3.0-3.5 MeV

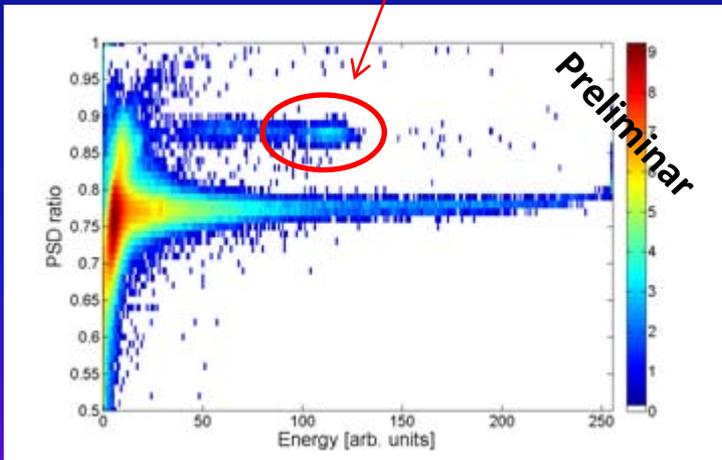
CLYC and Fast Neutrons

In Milano we could test

- 1"x1" detector with 95% ^6Li enrichment
- 1"x1" detector with ^7Li enrichment

- In april 2014 the detectors have been tested in Frascati using a monochromatic beam of 14.1 MeV neutrons and a beam of ≈ 2.5 MeV neutrons

2.5 MeV neutrons



CLYC scintillator with ^7Li enrichment

From web of science

- 2008-2009 5 papers
- 2010-2011 5 papers
- 2012 9 papers
- 2013 14 papers

Other elpasolide crystal like CLLB or CLLC
Seems to show simal properties for neutrons but
better energy resolution. However commercially
available now

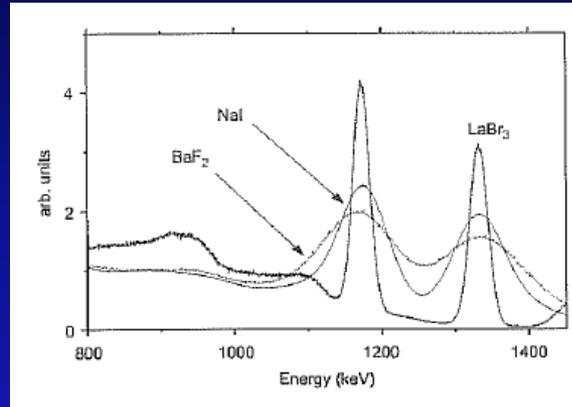
Lanthanum Halide: $\text{LaBr}_3:\text{Ce}$ Detectors

It was discovered in 2001 in Delft

It is now a well known scintillator detector

- it is in Knoll book !

Figure 8.14 Comparison of the ^{60}Co pulse height spectrum measured with 1-inch \times 1-inch LaBr_3 , NaI , and BaF_2 (From Nicolini et al.²¹⁵).



We have performed an Intense R&D activity on $\text{LaBr}_3:\text{Ce}$ detectors

- Rise time
- Pulse line-shape
- Count Rate
- Pulse distortion with γ -rays energy
- Linearity in energy
- Energy resolution and NON homogeneity
- High energy gamma rays
- Efficiency

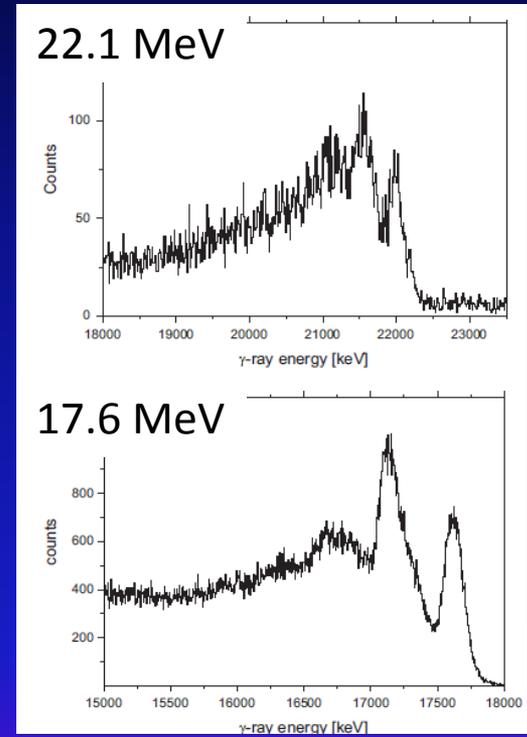
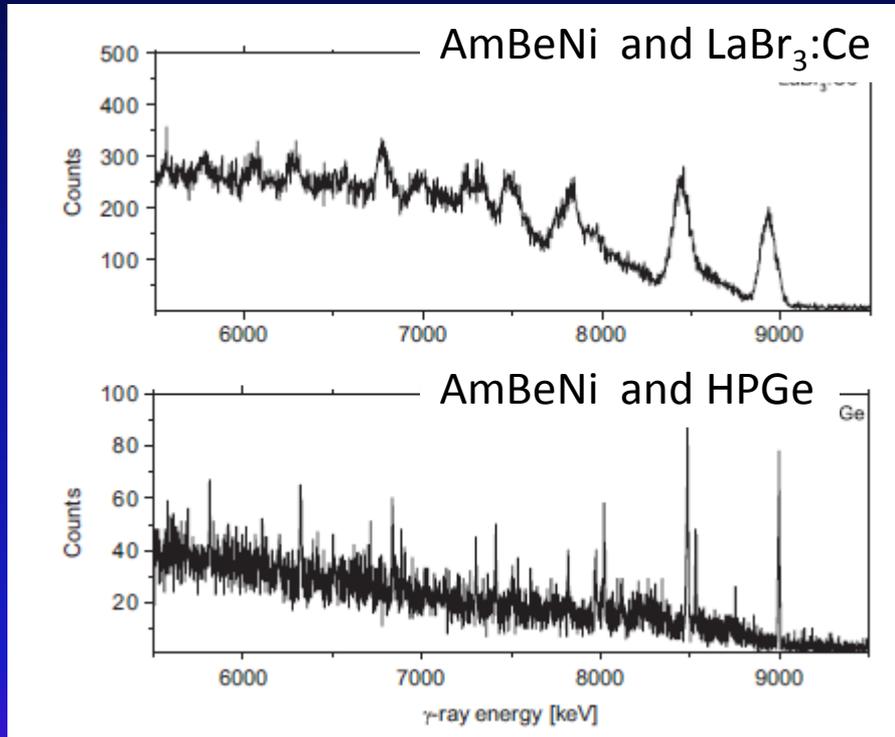


Several working or almost ready arrays have been designed

- HECTOR⁺
- PARIS
- FATIMA
- $\text{LaBr}_3:\text{Ce}$ in Bucharest
- Darmstadt $\text{LaBr}_3:\text{Ce}$ array
- H γ S array

LaBr₃:Ce Detectors have excellent energy resolution

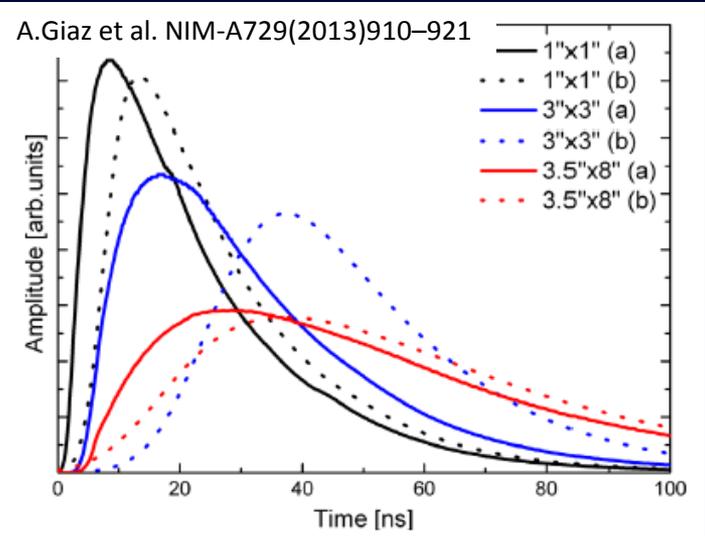
LaBr₃:Ce is, at the moment, the only scintillator which is capable to measure efficiently and separate the First Escape Peak and the Full Energy Peak in case of 15-20 MeV γ -rays



However non homogeneity in large volume crystals may limit the energy resolution to 0.5-1%

LaBr₃:Ce Detectors have excellent Time resolution

Time Resolution is however related to the size of the crystal and to the bandwidth of the coupled PMT. In general, the larger is the PMT the 'slower' is its time response.

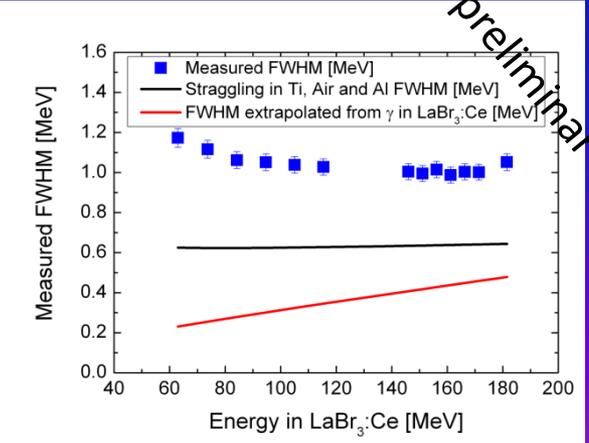


Continuous black, blue and red lines: Pulse lineshape for different LaBr₃:Ce crystals measured with Hamamatsu H6533 PMT (0.7 ns risetime)

- LaBr₃:Ce (whatever is the size) has a time resolution < 1 ns
- TOF neutron discrimination/measurement/tag
- good efficiency to detect fast neutrons (≈70%)

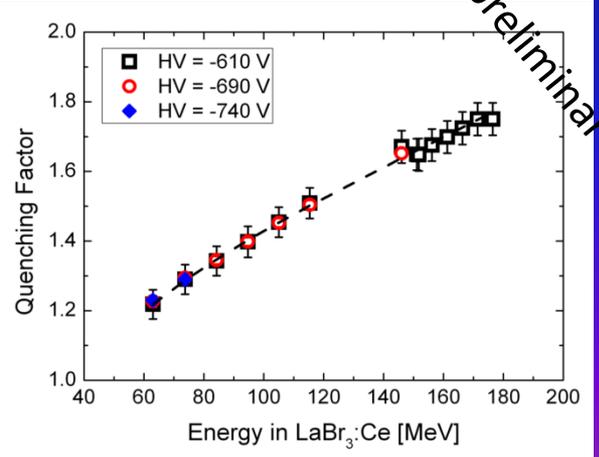
Extremely fast timing (FWHM < 300 ps) might be difficult to achieve with large volume (i.e. 3" x 3") LaBr₃:Ce

LaBr₃:Ce Detectors can measure charged particles



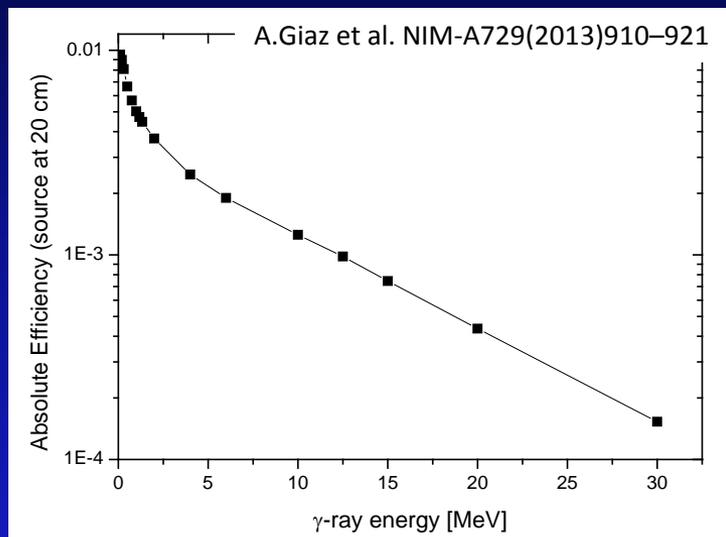
Measurement performed in Cracow using a LaBr₃:Ce 3"x3" and the proton beam from the new Cyclotron Center Bronowice at IFJ PAN Krakow.

Linearity, resolution, quenching and pulse lineshape have been measured.



HECTOR⁺ Array

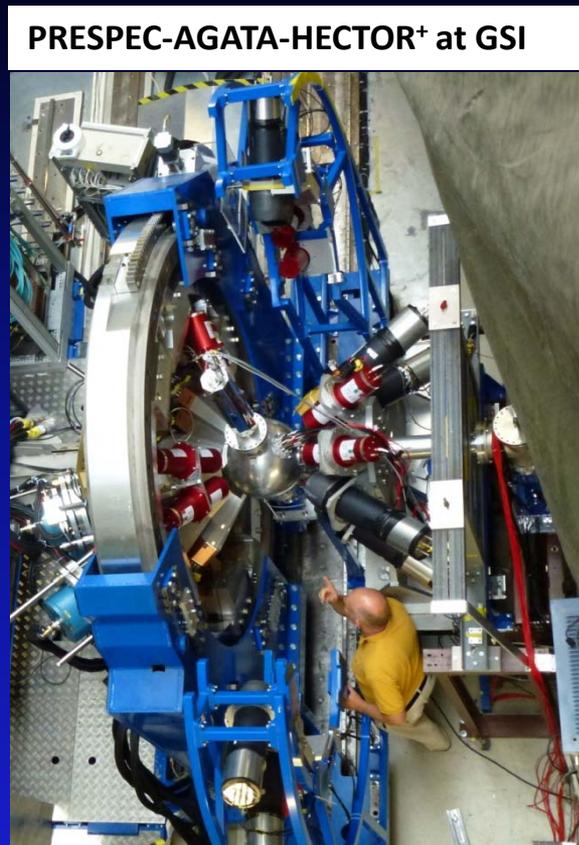
- High efficiency portable scintillator detector array
- 10 large Volume LaBr₃:Ce detectors (9 x 20 cm)



GEANT Simulations for **1 detector**
Source at 20 cm

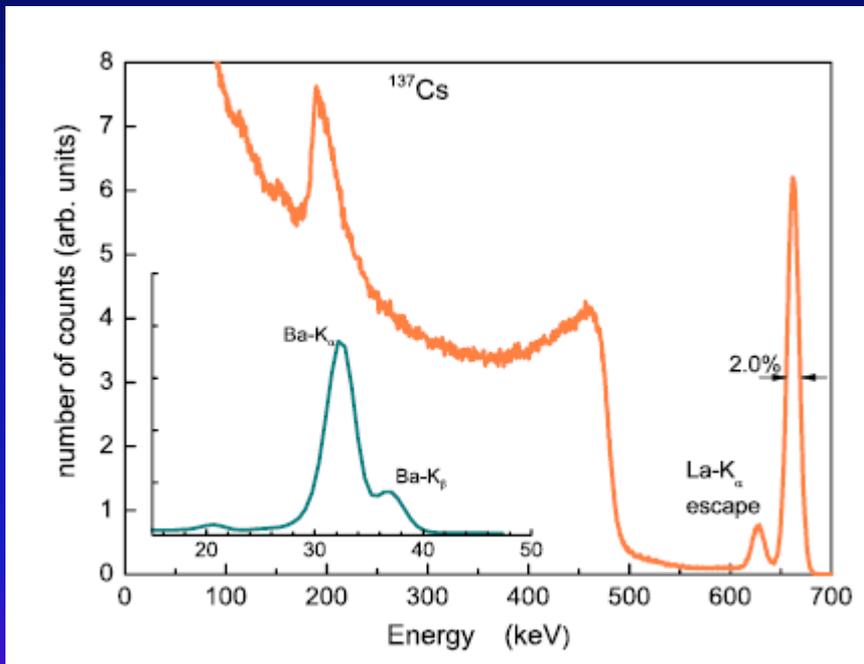
One large volume 3.5"×8" LaBr₃:Ce detector at 20 cm from target has ~10% relative full energy peak efficiency for 10 MeV γ-rays.

A 10 detector large volume 3.5"×8" LaBr₃:Ce array placed 20 cm from the target has 1% absolute full energy peak efficiency for 10 MeV γ-rays.



Co-Doping breakthrough in $\text{LaBr}_3:\text{Ce}$ and $\text{CeBr}_3:\text{Ce}$

- Low energy electrons non linear response and scintillator non constant light yield for small values of deposited energy affect the overall performances of a scintillator
- Ionization densities and electron-hole mobility characterize the detector performances
- Very recently this problem was understood and a solution was identified in co-doping



APPLIED PHYSICS LETTERS 102, 161915 (2013)

In this paper an energy resolution of 13 keV at 662 keV was reported for Sr^{++} co doped $\text{LaBr}_3:\text{Ce}$

At IEEE-2013 in Seoul an energy resolution of 20 keV at 662 keV was reported for Ca^{++} co doped CeBr_3

Such crystals are not available for tests at the moment

PMT non idealities can constitute an issue for the standard use of such a high performing detector.

We are also measuring large volume $\text{LaBr}_3:\text{Ce}$ position sensitivity for Doppler Broadening correction with excellent results

Scintillators are getting more and more performing thanks to the R&D work done in companies, universities and laboratories

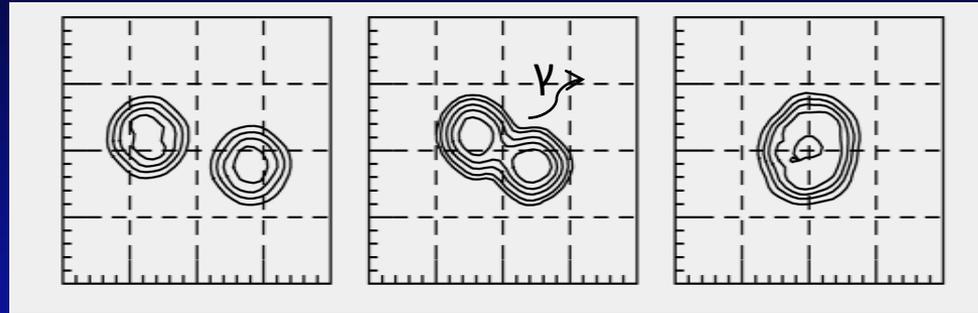
Scintillator cannot compete with HPGe detectors in terms of energy resolution but ..

- Energy Resolution might not be a key factor if the density of transitions is small enough.
- Scintillators have sub-nanosecond time resolution for
 - Ultra fast timing (see for example Fatima, Bucharest $\text{LaBr}_3\text{:Ce}$ array).
 - Background rejection using TOF.
 - Neutron discrimination using TOF.
- Scintillators (if in large volume) have a high efficiency for high energy γ -rays.
- Scintillators are easy to maintain and versatile (no cooling, no radiation damage).
- Scintillators might be cheap.
- New scintillators (CLYC, CLLB, CLLC) can measure neutron kinetic energy.

A scintillator array (i.e. $\text{LaBr}_3\text{:Ce}$, CLYC, PARIS, ...) is capable to work in a standalone configuration but, if coupled to a second radiation detection system (HPGe, Active Target, β decay station, ...) makes much more powerful the physics program.

SPES LOIs on Prompt Dipole and Dynamical Dipole and Hot GDR measurements

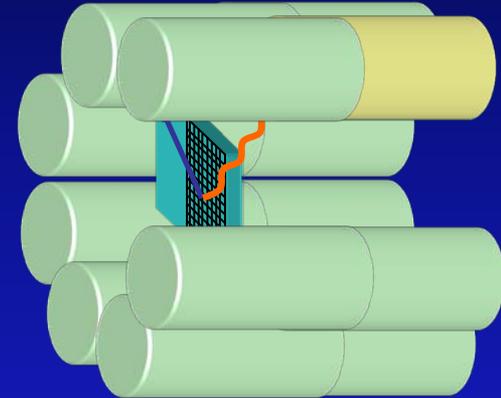
- Measurement of high energy (8-22 MeV) γ -rays emission from compound nuclei in very asymmetric (in term of N/Z) reactions



- Measurement of the γ -decay of the GDR built in the compound nucleus
 - Isospin mixing measurement or Jacobi shape transition
 - In such LOIs new scintillator provides
 - Efficiency for high energy γ -rays
 - TOF discrimination of evaporated neutrons
 - measurement of pre-equilibrium neutrons
 - Measurement of populated residues alternative to other ancillaries
 - Multiplicity measurements
 - It will be better if scintillators can be placed inside scattering chamber (is it possible but not obvious)

SPES LOIs on Beta-Decay measurement at SPES

- A thin passive tape transporting system
 - Already at the first operation of SPES, in the so-called “SPES alpha phase”, many new nuclear species will be available.
 - New scintillators can provide an extremely efficient TAS total-absorption spectrometer with a good energy resolution for the single transitions, time resolution for some half-lives measurements of levels in the daughter nuclei (using fast timing techniques) and neutron tagging or spectroscopy
 - As a total absorption spectrometers TAS new scintillators can provide a measurement of the decay heat in beta decay
- It will be possible to study collective resonant states at high excitation energy states populated by β decay with an high Qvalue. In particular one can populate the Pygmy Dipole Resonance in selected physics cases, where the spin and parity of the mother nucleus do not disfavour the population of such states in the daughter nucleus.
 - In such LOIs new scintillator provides efficiency for high energy γ -rays and time resolution for background rejection



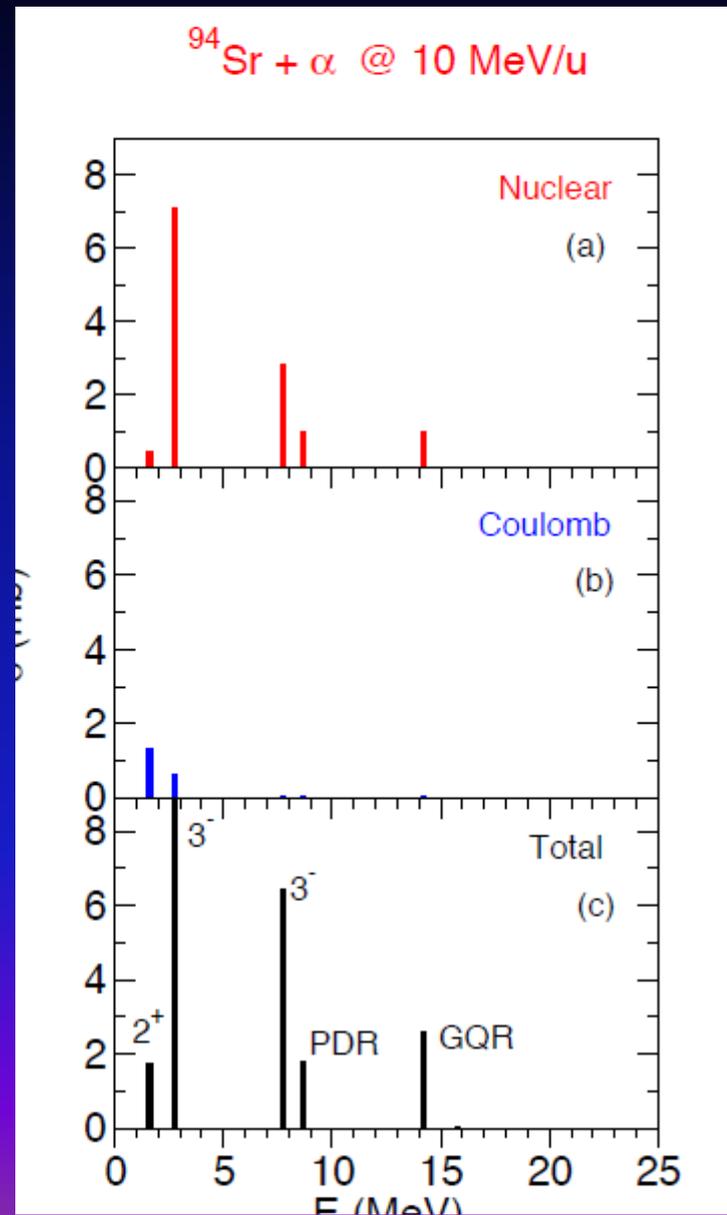
SPES LOIs on inelastic scattering experiment for PDR studies

The LOI focus on the study of low-lying dipole excitation via nuclear probes in nuclei far from stability.

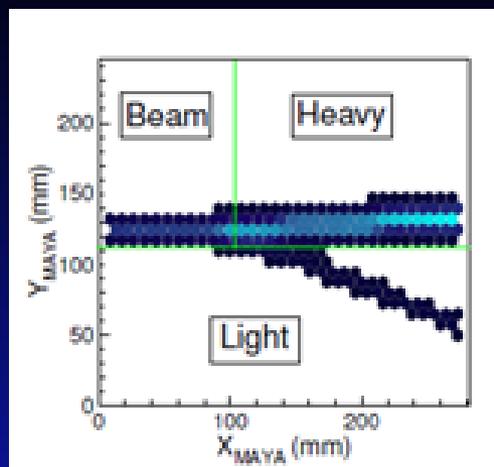
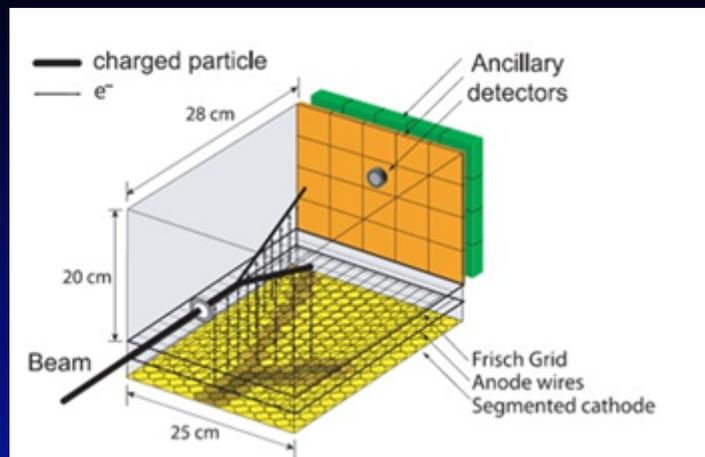
In the low-lying dipole excitations the transition densities are in phase inside the nucleus while the at the surface only the valence neutrons contribute. One can study these low-lying dipole states by using isoscalar and isovector probe

The gamma decay of the PDR requires the measurement of high energy γ -rays and fast neutrons (together with particles). In fact, the PDR states are mainly located above the neutron evaporation threshold, 7 MeV in ^{94}Sr , the measurement of neutrons can be very useful to determine the absolute cross section.

- In such LOIs new scintillators can/might provide:
 - Efficiency for low and high energy γ -rays
 - TOF background rejection
 - neutron spectroscopy
 - n, γ coincidences



SPES LOIs with Active Target



Active target in SPES
- see talk of G. Grinyer

- new way to tag nuclear
reactions with

Surrounding the active target with high performing scintillators permits to tag the reaction channel (with gamma or neutron emission) and in case very thin layer are used in front of the detector to additionally measure light charged particles emitted in the reaction

SPES LOIs

- Heavy-ion binary reactions (Multi Nucleon Transfer)
- Search for deformed oblate structures in ^{96}Y
- γ -ray spectroscopy in Ac and Fr
- Entry distributions for fragments produced in deep-inelastic collisions
- Coulomb excitation tagged by beta decay

- Scintillators as ancillary to increase efficiency for high energy γ rays
- Scintillators for sum energy and multiplicity

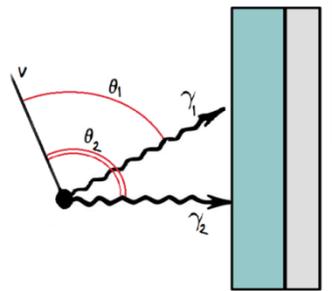
Conclusion and Perspective

- An impressive development in scintillator technology took place in these last years. This 'revolution' is not yet finished and newly designed high performing scintillator detector appeared and will appear.
- Scintillators cannot compete with HPGe detectors in terms of energy resolution. They could be, however, easy to handle, cheap, with excellent time resolution and efficiency for high energy γ -rays.
 - The scintillator $\text{LaBr}_3:\text{Ce}$ was discovered in 2001, now 2 liter crystal are available and several arrays are developed (i.e. HECTOR⁺, PARIS, ...)
 - $\text{LaBr}_3:\text{Ce}$ is now a 'mature' high performing scintillator that can/is necessary to fulfill several the experimental task in SPES:
 - Measurement of Dynamic Dipole and of hot GDR
 - A total absorption spectrometer in a beta decay station
 - Measurement of the gamma decay of the Pygmy Dipole Resonance
 - Measurement with active target
 -
 - Several other 'new' scintillator are almost ready to be used and can/will enforce several physics cases in SPES (i.e. CLYC or Co-Doped scintillators)

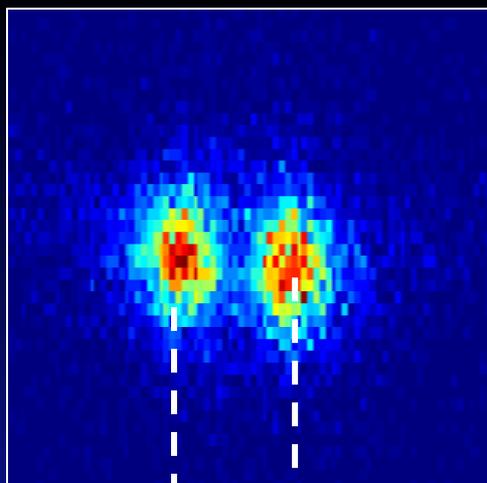
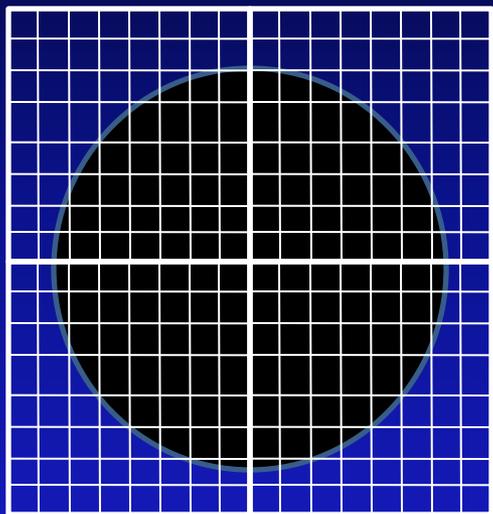
A scintillator array (i.e. $\text{LaBr}_3:\text{Ce}$, CLYC, PARIS, ...) is capable to work in a standalone configuration but, if coupled to a second radiation detection system (HPGe, Active Target, β decay station, ...) makes much more powerful the physics program.

Thanks for the attention

SPARE

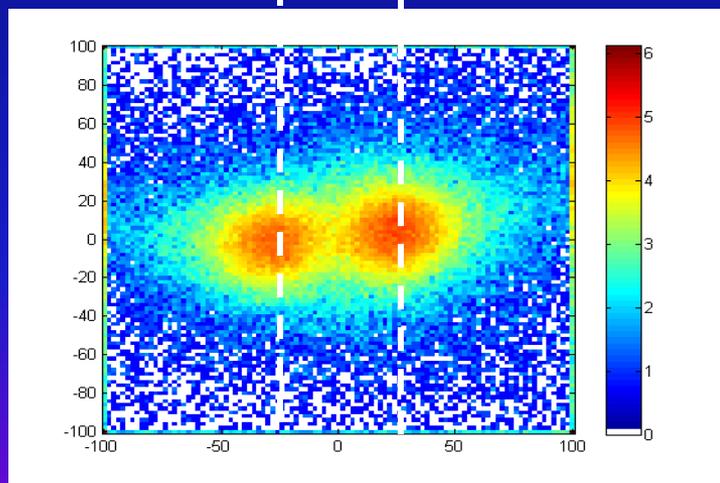
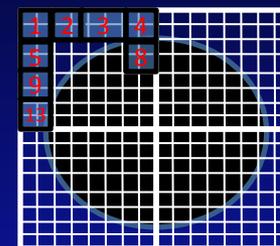


3"x3"
LaBr₃:Ce



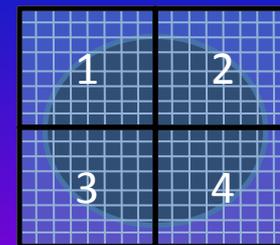
E gamma = 662 keV

64 integral signals
Instead of 256



E gamma = 662 keV

4 integral signals
Instead of 256



4 cm



Properties of new scintillators (SrI₂, CeBr₃, GYGAG)

SrI₂



Material	SrI ₂ :Eu	Emission Spectral Range	400 – 480 nm
Melting Point	538°C	Peak Scintillation Wavelength	~ 435 nm
Density	4.59 g/cm ³	Decay Constants (Eu2+).....	1 – 5 μs *
Z _{eff}	49	Scintillation Light Yield.....	80,000 ph/MeV
Crystal Structure	Orthorhombic	X-ray Absorption Coef. at 100 KeV	2.88 cm ⁻¹
Water Solubility	Hygroscopic	X-ray Absorption Coef. at 662 KeV	0.13 cm ⁻¹
Refractive Index	1.85	Radiation Length	1.95 cm
Coefficient of Thermal Expansion	2.164 x 10 ⁻⁵ /°C (lattice b)		* Depending on sample size

Slow scintillator
with self absorption
but with excellent
energy resolution
< 3-4% @ 662 keV.
It is available

CeBr₃



It can be seen as a 100% doped LaBr₃:Ce
Fast scintillator (subnanosecond time resolution as LaBr₃:Ce)
Good Energy resolution (< 5% @ 662 keV but worse than LaBr₃:Ce)
No internal radiation
It is available up to 3"x3"
CoDoping developed in prototypes

GYGAG



Gd_{1.5}Y_{1.5}Ga_{2.2}Al_{1.8}O₁₂ (Ce) - Transparent polycrystalline ceramic scintillators
It is a Ceramic Matherial (no crystal grow)
Density and effective Z of GYGAG are 5.8 g/cm³ and 48,
Very few samples available (probably one)
Good Energy resolution with SDD (< 5% @ 662 keV but worse than LaBr₃:Ce)
Fast scintillator (rise time ~ 30 ns)