## **Key Trends in Scintillation Physics**

A. Gektin

Institute for Scintillation Materials 60 Nauka ave., 61001, Kharkov, Ukraine Where we are with knowledge and experience?

Is this knowledge satisfy practice?

What we have improve as the first priority?

1. Are so many scintillator choices have a chance for application?

2. Different modality status and trends

3. Optimal solution for different applications. New results need and cost. (Criteria of an optimal engineering)

#### Last main reviews of scintillators development and applications

NEEDS, TRENDS and ADVANCES IN INORGANIC SCINTILLATORS C.Dujardin, E.Auffray, E.Bourret, P.Dorenbos, P.Lecoq, M.Nikl, A.N.Vasil'ev, A.Yoshikawa, R.Zhu

Reent development in X-ray imaging technology

**Robert G. Lanier** 

Development of new scintillators for medical applications

Paul Lecoq

Review of X-ray Detectors for Medical Imaging Martin Hoheisel (Siemens)

Current Trends in Scintillator Detectors and Materials William W. Moses

Recent R&D trends in inorganic single-crystal scintillator materials for radiation detection,

Martin Nikl and Akira Yoshikawa

Takayuki Yanagida, Inorganic scintillating materials and scintillation detectors. end other !

## Scintillator market and main driving forces

## **Key properties for main applications**

	Light yield	Resol.	Decay Time	Rad. damage	Back ground	Afterglow	Cost
Nuclear Medicine (SPECT, PET)		+	+				+
Health physics : Dose (particles, photons, neutrons)	+		+				+
Anatomic imaging (X-ray CT)			+			+	+
Security (non-prolifiration, scannin, z-analysis, spectroscopy		+			+		+
High energy phyics: particles, photons			+	++			+
Geology: density, well logging, finding U, Th via spectroscopy	+	+					+
Space (gamma) spectroscopy, neutrons		+			+		
Industry (density, speccopy)	+	+					+



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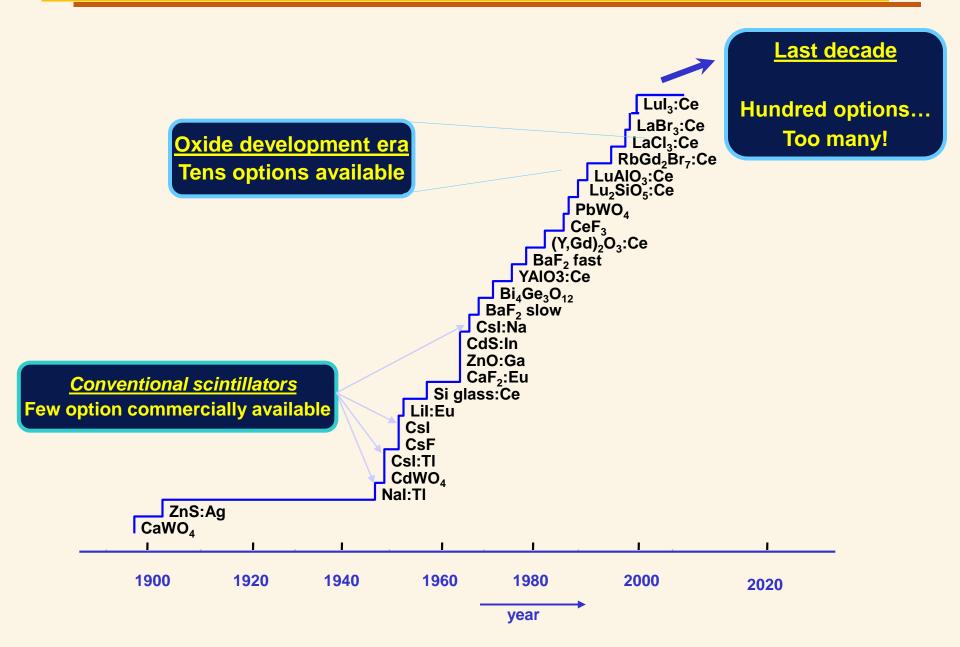


## **Key properties for main applications**

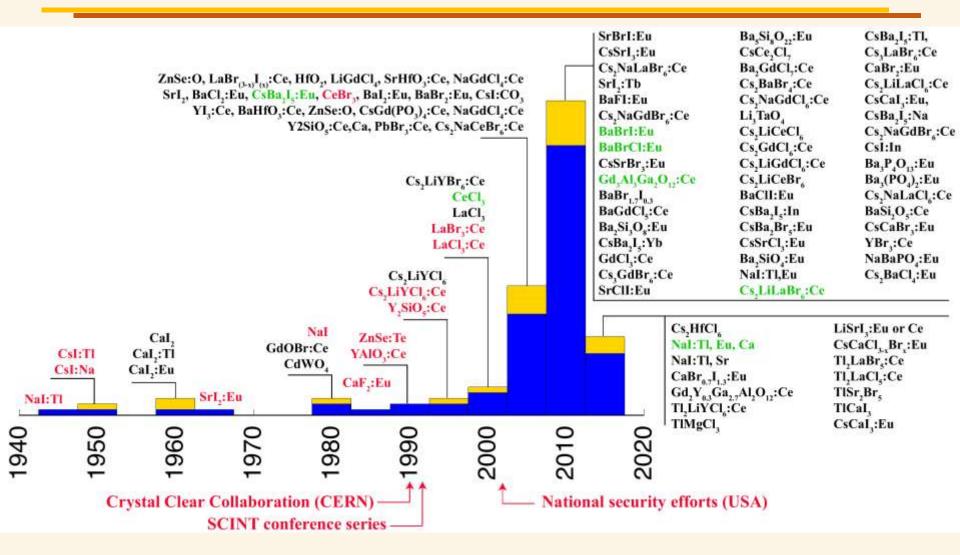
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Industry (density, speccopy)	+	+						+	



### Scintillation discovery key points



#### New scintillator search and development



## Scintillation parameters for new scintillators

Scintillator	Melting point, <sup>o</sup> C	Density, g/cm <sup>3</sup>	Z <sub>eff</sub>	LY, ph/MeV	Scintillation decay, ns	∆E at 662keV, %
LaBr <sub>3</sub> :Ce 5%	743	5.1	48.3	~65 000	16	3.2
Srl <sub>2</sub> :Eu	538	4.55	50.3	~100 000	1 000	2.6
Nal:Tl	651	3.7	хх	~40 000	230	6-7
BaFI:Eu 5%	893	5.45	хх	55 000	хх	8.5
BaClBr:Eu 5%	890	4.5	xx	52 000	хх	3.55
BaCll:Eu 5%	887	4.6	xx	54 000	хх	9
BaBrI:Eu 8%	885	5.2	xx	97 000	хх	3.4
KSr <sub>2</sub> I <sub>5</sub> :Eu 4%	476	4.3	51.4	~94 000	990 (~89%)	2.4
KSr <sub>2</sub> Br <sub>5</sub> :Eu 5%	575	4.0	хх	~75,000	1100 (80%)	3.5
KBa <sub>2</sub> I <sub>5</sub> :Eu	566	4.52	53.2	~84 000	910 (~81%)	2.6
K <sub>2</sub> Bal <sub>4</sub> :Eu	579	4.01	51.2	~57 000	720 (~67%)	2.9
KCal <sub>3</sub> :Eu 3%	524	3.81	50.6	~70 000	1 000 (~90%)	3.8
CsSrl <sub>3</sub> :Eu 7%	640	4.29	51.1	~73 000	3 000 (90%)	3.7
CsSrBr <sub>3</sub> :Eu 5%	760	3.8	44.2	~40 000	3 000 (90%)	3.8
CsCal <sub>3</sub> :Eu 3%	690	4.06	52.6	~39 000	1 000 (~90%)	3.8
Cs <sub>2</sub> NaYBr <sub>3</sub> I <sub>3</sub> :Ce	508	4.03	48.6	~41 000	56 (~47%)	3.5
Cs <sub>2</sub> NaLaBr <sub>3</sub> I <sub>3</sub> :Ce	508	4.05	50.2	~58 000	78 (~25%)	3.3

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### **Oxide scintillators for new application**

Some parameters of oxide scintillators used for low-energy  $\gamma$ -quanta detection

Crystal	Density g/cm <sup>3</sup>	Ce <sup>3+</sup> (Pr <sup>3+</sup> ) 5d-4f emission, nm	Decay time, ns	Max. LY, ph/MeV <sup>-1</sup>	Energy res., [%] @662 keV
YAG:Ce	4.56	550	90–100	28000	6–7
LuAG:Ce	6.67	525	55–65	24000	6–7
GGAG:Ce	6.2	540	90–170	58000	4.2–5.2
LuAG:Pr	6.67	308	20–22	20000	4.6–5
LuYAG:Pr	6.2–6.5	310	20–22	33000	4.4–6
YAP:Ce	5.35	365	19–25	20000	4.5–5.5
YAP:Pr	5.35	247	8–10	12000	11–13
LYSO:Ce, Ca	7.2	400	30–35	32000	8–9
(Gd,La)PS :Ce	5.4–5.7	365–370	45–50	41000	5–6

### Scintillation parameters of main elparolites for neutron detection

	CLYB	CLYC	CLLC	CLLB	CLLBC
Light yield, gamma,					
ph/MeV neutron, n/MeV	24 000 90 000	20 000 70 000	35 000 110 000	45 000 150 000	45 000 150 000
ER, %@662 keV	4.1	4.0	3.4	2.9	3
Emission, nm	410	370	380	410	410

There are many choices even in the same material type !

Which is the best? Compare with existed options ?

## Important commercially available scintilators and their applications are :

L(Y)SO	PET (medical)
PWO	HEP (CMS)
LaBr <sub>3</sub> (Ce)	High resolution (HR) gamma spectroscopy
CLYC(Ce)	Neutron detection
Srl <sub>2</sub> (Eu)	HR gamma spectroscopy
GAGG	Position sensitive detectors (not hygroscopic
	crystal)

### Important factors for Industrial use:

- Can larger crystals be grown?
- What is the cost ?
- Is the cost/performance rate corresponds to industry claim?

### Market data

The global radiation detection, monitoring, and safety market is expected to reach USD 2.26 billion by 2022 from USD 1.71 billion in 2017

## Scintillator market - ~200mln USD

- \* market structure
- \* estimations details
- \* complimentary markets (screens, storage etc.)

## Photo receiver market 436 mln USD

- (to 2020 up to 520 mln USD)
  - PMT 277 mln (-2.1% trend)
  - SiPMT 95 mln (+16% trend)
  - other 63 mln

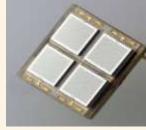
M&M data

### NOTES:

- Raw material cost can reach up to 70%
- Photoreceiver is significant part of detector cost
- Electronic cost can exceed scintillatr cost itself
- Phosphor screens comparable with crystal production



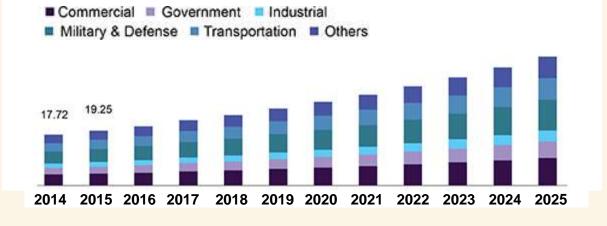




## Few examples of market trends

## Global security market size will rise to 167 bln USD till 2025

#### U.S. security market, by end-use, 2014 - 2025 (USD Billion)



- \* Secure Cities
- \* Radiation Portal Monitoring
- \* Material Protection, Control, and Accountability
- \* Mega ports
- \* Container Security Initiative
- \* Second Line of DefenseSecure Cities







### From: DNDO

DNDO is the primary entity in the U.S. government for implementing domestic nuclear detection efforts



Started at15 April 2005. 563,8 mln \$ annual budget

To: CWMD June 2018

The mission of the Countering Weapons of Mass Destruction (CWMD) Office is to counter attempts by terrorists or other threat actors to carry out an attack against the United States or its interests using a weapon of mass destruction.

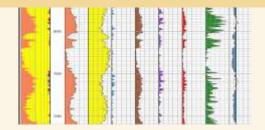
## Oil explore market

## Well logging and scintillators place

### Negative trends. Scintillators for minerals and oil explore



## Oil price down terminate search and development in this application !





**Schlumberger Doll Research** 

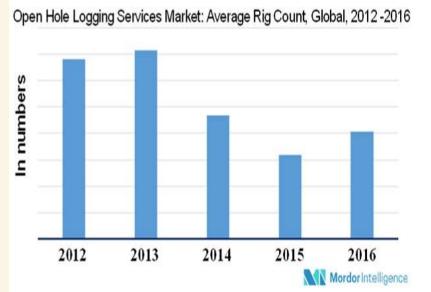
### Oil market as diver for scintillator development and production

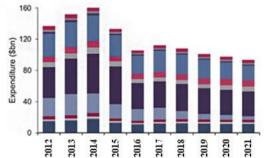


\* Oil market is follows oil market price

## \* Drilling market follows general oil market

Global drilling and well services and oilfield equipment expenditure by market segment (double counting adjusted), 2012-2021.

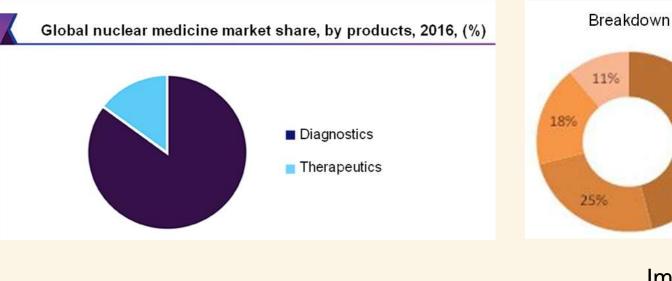


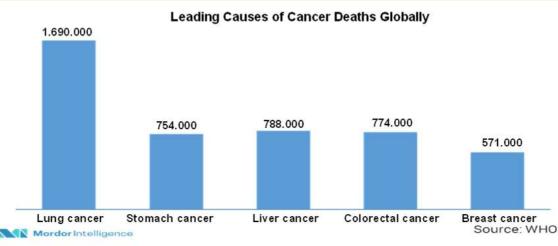


Medical scintillator market.

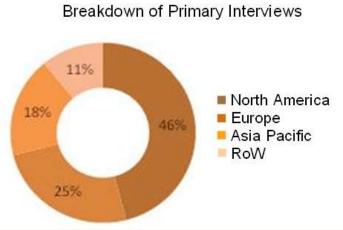
X-Ray materials and designs

### Nuclear medicine still is the main market and driving force for scintillator market





## Nuclear Medicine Market Size Worth \$15.2 Billion By 2025



Imaging systems play the dominant role in detector design and development

#### but

dominant trends are permanently changed

### NM market pluses and minuses

Diagnostic equipment reach 35% of the medical market

"Big Three" does not obviously claim for the cheap solution.

Optimal does not mean the cheap!



Medical market is the most stable for detector (4-5% growth).

The last trends are moved to semiconductor and ceramic detectors

### General trends in medical imaging (Siemens note)

- C All images become digital
- C 3D methods are gaining preference over 2D
- C **Combination** of different modalities
- c Functional imaging
- c Imaging for therapy
- C Connectivity

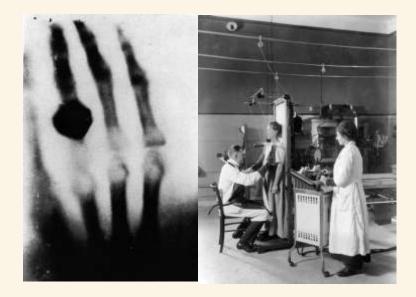
#### Aims

- better diagnosis
- targeted therapy
- cost optimization
- prevention
- B Availability of images throughout the whole health care system
- B Tele-medicine
- B Electronic patient record
- C Computer-Assisted Diagnosis (CAD)

### **Dominant option for X-ray CT detectors**

- Amorphous Selenium (a-Se) flat panel technology with active pixel technology.
- Structured CsI (Cesium Iodide) scintillators
- TFT (thin-film transistor) and CMOS (complementary metal–oxide– semiconductor) technology
- CZT (cadmium-zinc telluride) arrays and their associated electronics.
  - Ceramic scintillators

#### 90 years ago and today





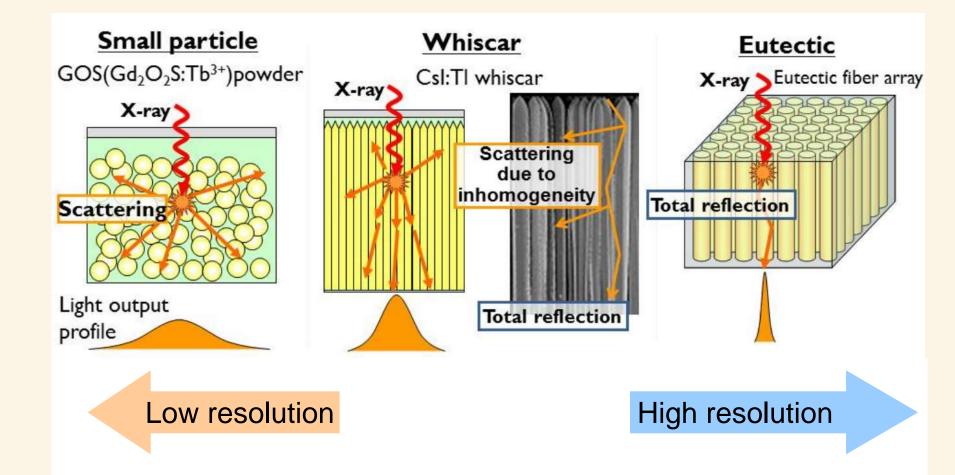
#### **CT** - There are many manufacturers and strong competitive landscape for detectors

1.	Flash Pad - GE	16. The Solid-State X-Ray Image Intensifier (SSXII) –
2.	PHOTON 100 - Bruker AXS	SUNY Researchers
3.	XinRay Systems - Siemens.	17. RadEye <sup>™</sup> X-ray Sensor Modules - Teledyne Rad-icon
4.	Digital Silicon Photomultipliers – Phillips	Imaging
5.	CdZnTe (CZT) Detectors- Redlen	18. The INTEGRAL Soft Gamma-Ray Imager (ISGRI) –
6.	Lensfree Optical Tomography – UCLA Research	ACRORAD
7.	<b>D-SPECT System</b> - Spectrum Dynamics	19. CCD 485 with Fiber Optic Faceplate – Fairchild
8.	XDAS detector boards V3 – Sens	Imaging
Tech		20. CMOS Linear Arrays– Fairchild Imaging
9.	Ultrafast Ceramic Scintillator (UFC) - Siemens.	21. Selenium-based Flat panel X-ray Detector – Toshiba
10.	Advanced X-Ray Detectors— DxRay Inc	22. SAPHIRE (scintillator avalanche photoconductor
11.	CCD Detector Development – MPI Halbleiterlabor,	with high-resolution emitter readout) Detector 23.
12.	<b>DEPFET Detectors</b> - MPI Halbleiterlabor	(DEXI) diffraction-enhanced x-ray imaging instrument –
13.	HICAM Gamma Camera – HICAM Collaboration	Nesch, LLC
14.	LuAG Scintillator Array – Japan Research	24. Linear scanning sensors with gas-based detector
15.	Strip Detectors – Baltic Scientific Instruments	modules for X-ray imaging – Korean Collaboration
		25 Socurity Dotoctore: High on orgy V roy Distforms

25. Security Detectors; High-energy X-ray Platforms – Varian

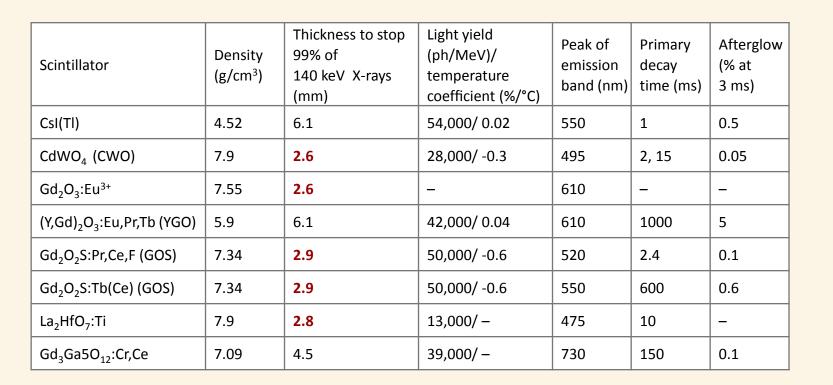
### Big Three (GE, Siemans and Philips) practically fully occupy SPECT and PET markets

### Granularity or pixelesation for spatial high reolution



### **Properties of scintillators used in X-ray CT imaging**

- The are many similar candidates
- Electronics compensate some differences between crystals
- Market value as the limit for some updates





# What are the mutual trends for all scintillators development?

Which claims are mutual?

Can we unify material claims to minimize list of scintillators?

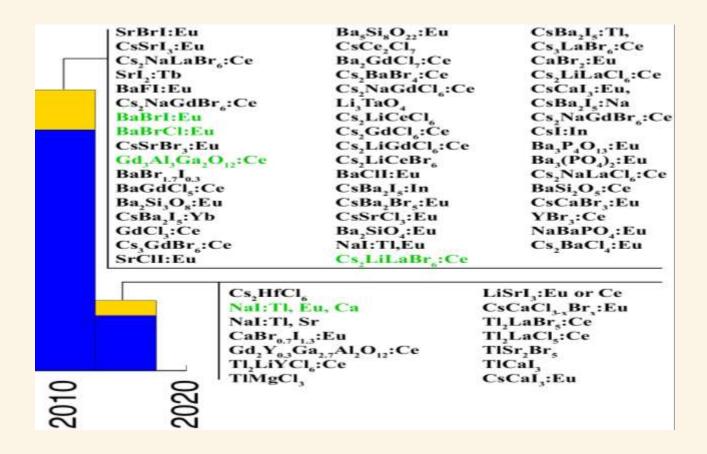
## What we can propose as scintillator improvement

\* New material search (???) or conventional scintillators improvement?

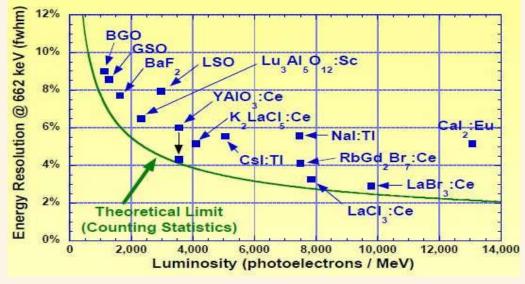
**\*\*** Co-doping as scintillator improvement method

**\*\*\*** Crystal treatment of data processing improvement

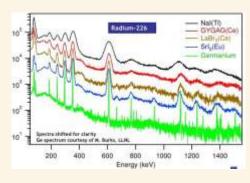
### New scintillator search and development



### Crystal performance spread... Energy resolution



#### **P.Dorenbos**



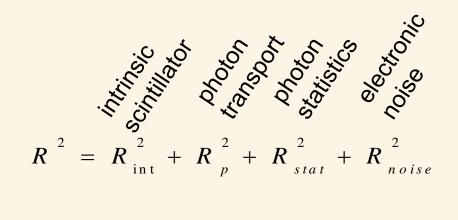
Isotopes separation

\* Theoretical energy resolution spread is not so wide as experimental one

- \*\* What are the reasons for such spread?
- \*\*\* Is resolution really significantly depends on the material

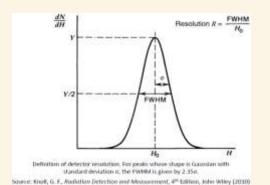
\*\*\*\*Can we manage the energy resolution and how we can do it?

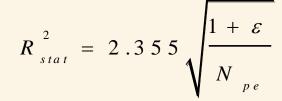
## **Energy resolution of a scintillator detector**

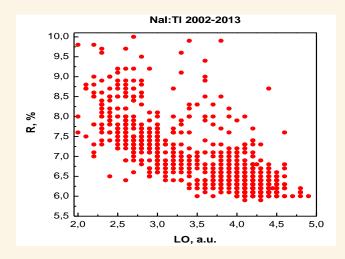


$$R_{\text{int}}^2 = R_{\text{in hom}}^2 + R_{\text{nonprop}}^2$$

#### M.Moszynski et al





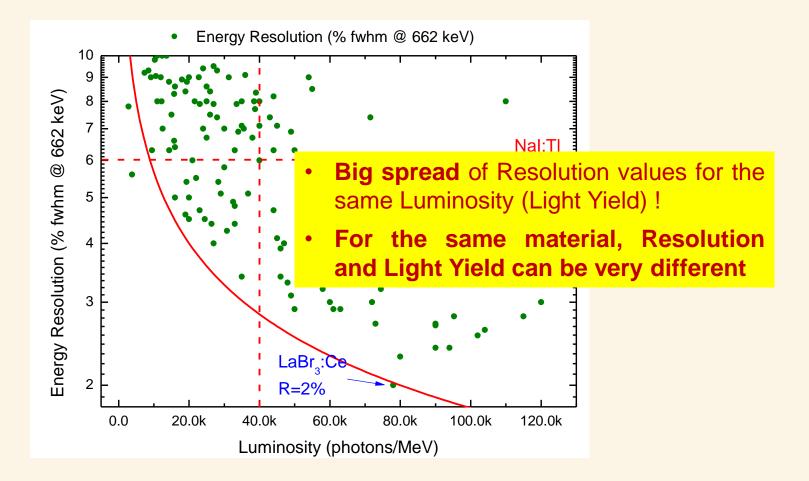


## There no direct correlation between light yield and energy resolution !!!

- Non-proportionality and resolution. Is any direct correlation?
- Contrary to definition energy resolution looks like structure sensitive phenomenon
  - dependence from crystal purity (undoped crystals)
  - dependence from peaking time
  - finally dependence from the luminescence type
- Definitions and theory

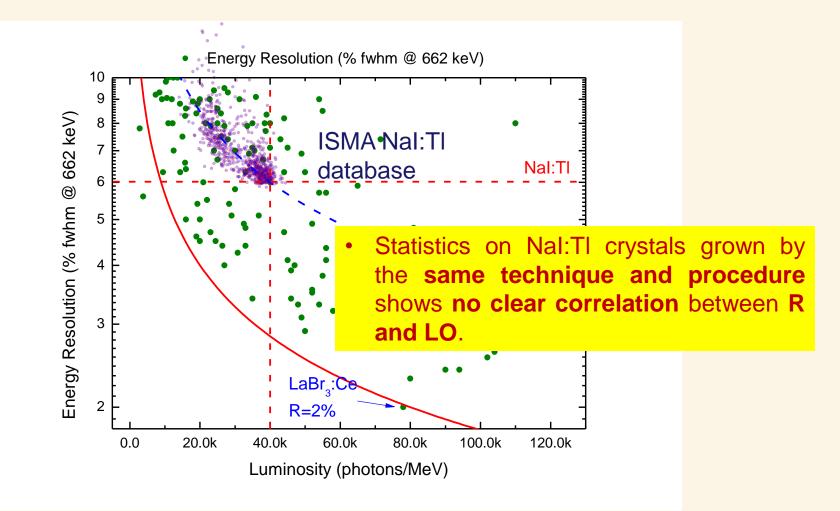
## **Classic theory does not properly describe scintillator !**

## Energy resolution vs Luminosity



Compiled by S. Vasyukov from S. Derenzo et al LBNL scintillator database

## Energy resolution vs Luminosity

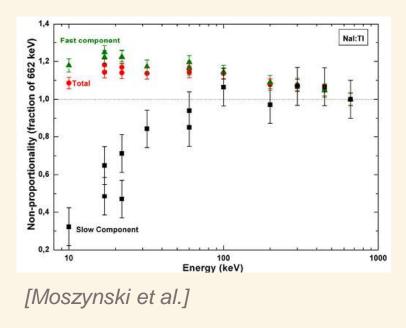


Compiled by V.Vasil'ev, S. Vasyukov from S. Derenzo et al LBNL scintillator database

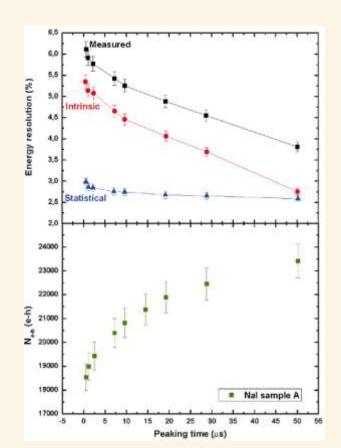
What we have to keep in mind when try to understand resolution phenomenon? What we have to keep in mind when try to understand resolution phenomenon?

### Non-proportionality depends on peaking time

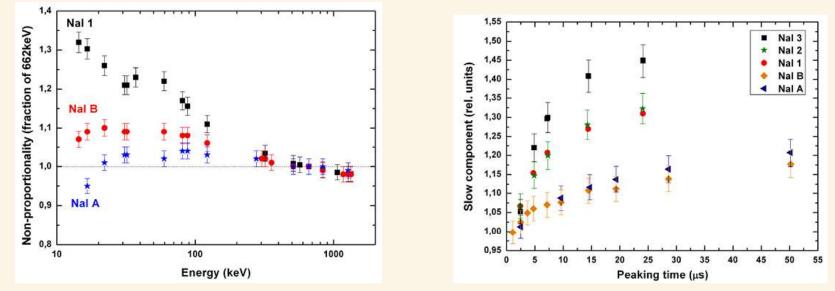
- Peaking (integration) time is an important for exact resolution measurement
- Peaking time measurements reflects contributions of different types of luminescence



• There is no direct correlation Between non-proportionality and energy resolution



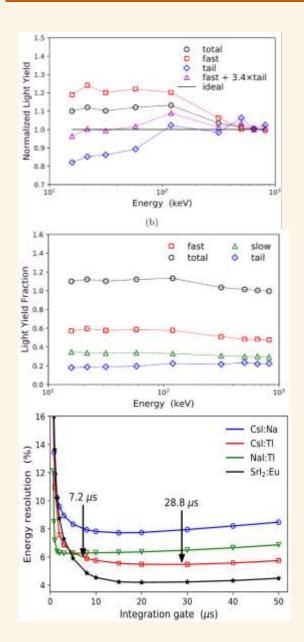




[Moszynski et al.]

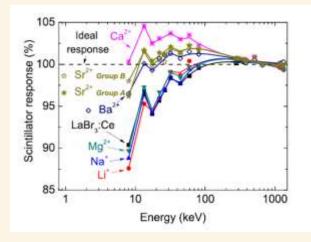
- Pure crystals are nominally pure only
- Nal with linear proportionality does not possess with better resolution
- The main difference of purity connects with long decay components

What we have to keep in mind when try to understand resolution phenomenon?



- Non proportionality (NP) and light yield for different decay components are different
- •The main difference of purity connects with long decay
- It is possible to modify NP and energy resolution by crystal purification or codoping

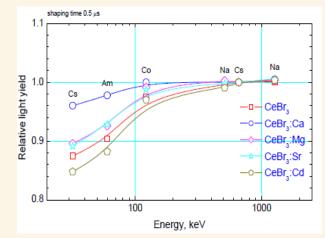
## **Co-doming (impurity) can change non-proportionality**



Non-proportionality of LaBr<sub>3</sub>:Ce scintillators with different co-dopants: Ca<sup>2+</sup>, Sr<sup>2+</sup>, Ba<sup>2+</sup>, Mg<sup>2+</sup>, Na<sup>+</sup>, and Li<sup>+</sup>.

### **Co-doping. Impurity** strongly affects the scintillator response.

[Alekhin et al.]



Non-proportionality of CeBr<sub>3</sub> scintillators with different co-dopants: Ca, Mg, Sr, and Cd.

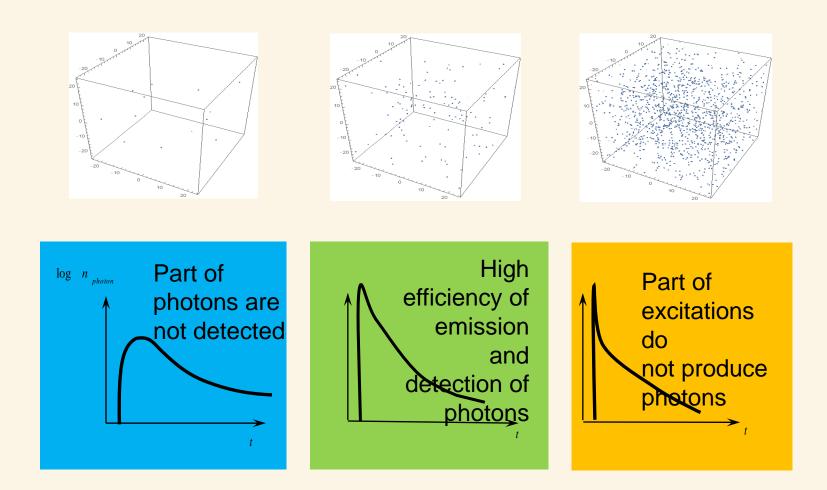
### Co-doping. Impurity can improve linearity, or make scintillator less proportional.

[Schotanus et al.]

# Can we assume the reasons of specific behavior of scintillators?

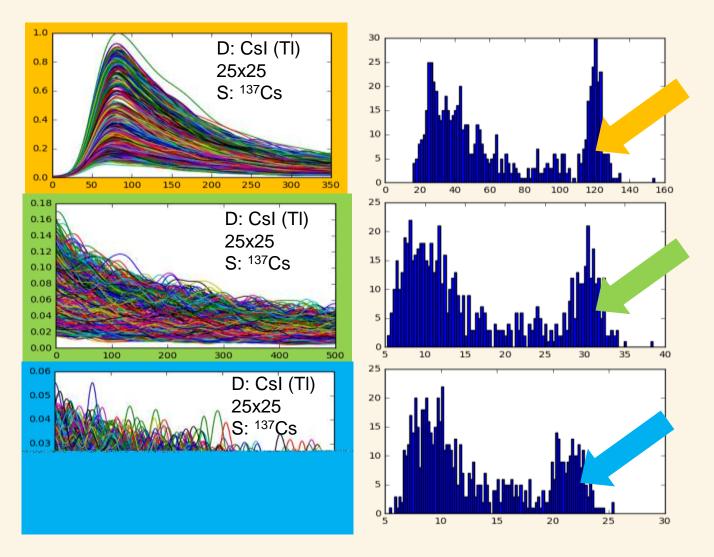
**Can we find new approaches for statistic description?** 

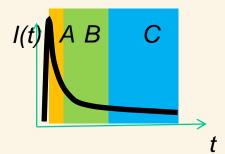
## **Excitation concentration and kinetics**



A.Vasil'ev, Fluctuation of track structure in terms of distribution of excitations and fractal dimensions, CCC Meeting, CERN, 2017

## **Decomposition of pulses by time**

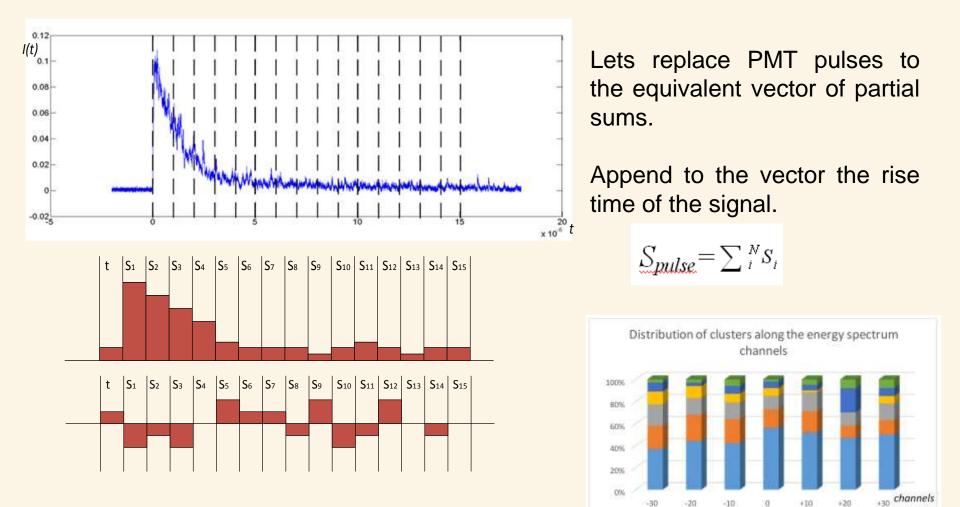




The shape of complete absorption peak is changes

A.Sobolev, LUMDETR 2018

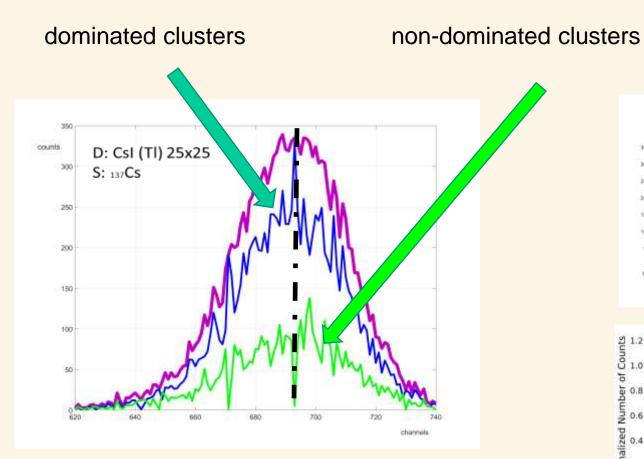
## Research idea: What do we do?



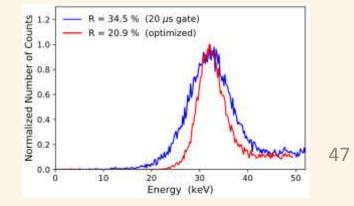
### A.Sobolev (LUMDETR 2018)

## **2-clusters decomposition**

The part of energy spectrum (complete absorption peak)

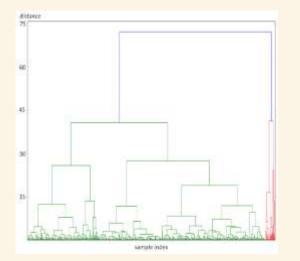


Energy spectrum 3D 300 -300



A.Sobolev, LUMDETR 2018

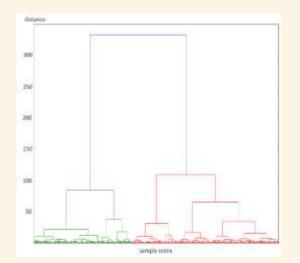
The CsI (TI) crystal with R=13,9% has large distance between a clusters and it has less number of pulses in dominant clusters



CsI:TI, 25x25 <sup>137</sup>Cs

R=6,7%

R=13,9%



#### Methodology:

- Choose norm (Euclidean, correlation, cosine, minkowski, hamming)
- Calculates distance
  matrix
- Choose a deep of clustering
- Calculates cluster diameters
- Hierarchical Clustering
- Determine a dominated clusters

A.Sobolev, ISMA, 2018

### **General factors:**

- Can larger crystals be grown at usable size?
- What is the cost ?
- Is the (superior) performance justified by the price ?

## **Specific factors:**

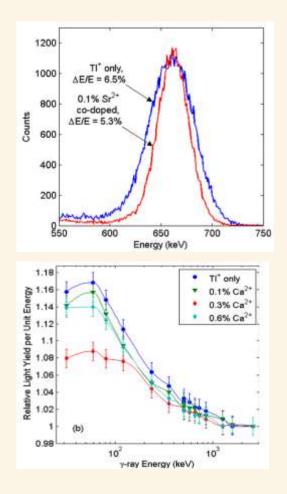
(Unique property for example)						
High speed / density	- LYSO (PET)					
Very short decaytime	- PWO(CMS)					
Very good resolution	- LaBr3:Ce, Srl2:Eu					
Price idea :	Nal(TI) detector 76 x 76 mm	: 2 k€ 5 \$ / cc				
	BGO detector 76 x 76 mm LaBr3/CeBr3 76 x 76 mm	: 5-6 k€ 20 \$ / cc : 35 k€ 100 \$ / cc				
		P.Schotanus				

- \* Fast emission Auffray E., Vasil'ev A
- \* 3D printing Sokolov P., Lobko A
- \* Meta structures (composites) Onufriev Yu
- \* n-gamma and so on separations Zhmurin P
- \* Fibers, thin films Ruziecka K
- \* SiPMT Mazzi A
- \* Ceramics Karpuh P

## Most "popular" efficient new scintillators

Crystal	ρ g/cm³	Lum λ, nm	LY ph/Mev	R, % Cs <sup>137</sup>	Decay τ, ns	Hygro- scopy	References
Cal <sub>2</sub> :Eu	3.96	467	110.000	5,2	1.000	strong	Cherepy, Moses, Derenzo, Bizarri, Bourret et
Srl <sub>2</sub> :Eu	4.55	435	115.000	2.6	1.500	strong	al.
Ba <sub>2</sub> Csl <sub>5</sub> :Eu	4.9	435	102.000	2.55	383;1.500	medium	2007 - 2012
SrCsl <sub>3</sub> :Eu	4,25	458	73.000	3.9	2.200	medium	Zhuravleva et al. 2012
BaBrl :Eu	5.2	413	97.800	3,4	500	low	Bizarri et al. 2011
Nal : TI	3.67	415	44.000	5.6	230	strong	
Csl : Tl	4.53	560	56,000	6.0	980	no	
CsI : Na		420	46,000	6.4	600	low	

## **Co-doping as the method of scintillator improvement**



[Yang et al.]

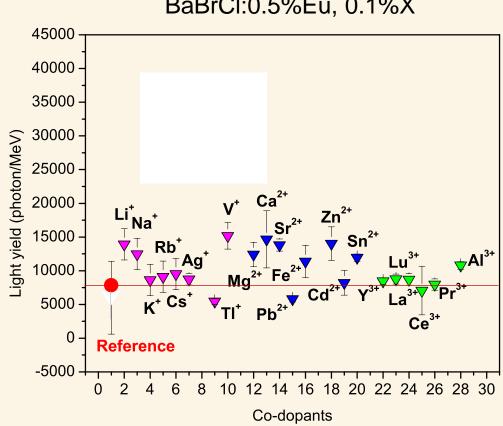
### Me<sup>2+</sup> (Ca in particular) for resolution improvement?

Nal:Tl production	R, %	Co-activators
Saint-Gobain	5.3	Eu, Sr, Ca
ISMA	5.2	Eu, Ca
LBNL	4.9	Eu, Ca
Standard	6.1	-

### Resume:

- Some performance improvement is visible
- The trend exists for some other Me<sup>2+</sup> co-doped combinations

## A Case Study: Co-doping of BaBrCl:Eu

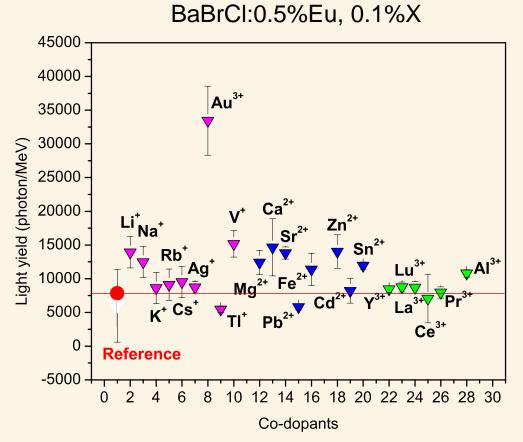


BaBrCI:0.5%Eu, 0.1%X

- Over 27 different co-dopants have ٠ been tested
- Samples obtained through non ٠ directional solidification

### E.Bourret, LUMDETR 2018

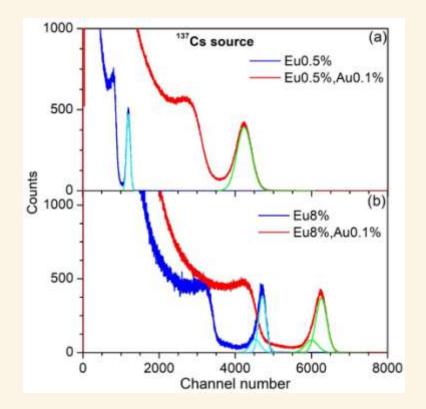
## A Case Study: Co-doping of BaBrCl:Eu



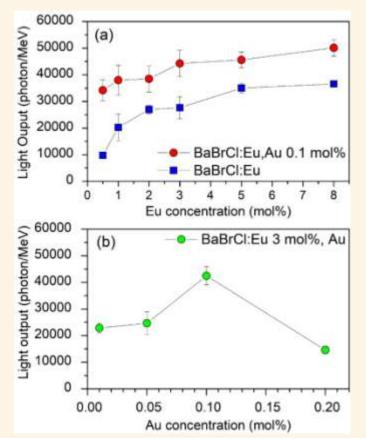
- Over 27 different co-dopants have been tested
- Samples obtained through non directional solidification
- Au co-doping stands out among all the others.

### E.Bourret, LUMDETR 2018

## **Pulse Height Spectra and Light Yield**



*Representative pulse height spectra of BaBrCl:Eu with and without Au codoping for two different Eu contents* 



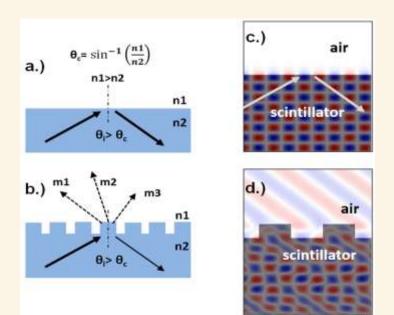
Light yield as a function of Eu and Au concentration



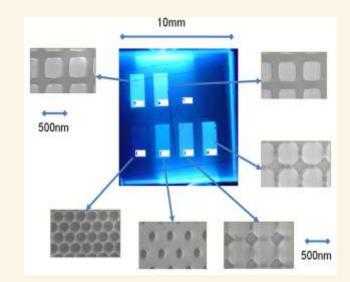
Systematic increase in light output over the entire Eu concentration range Optimum Au concentration is 0.1 mole%

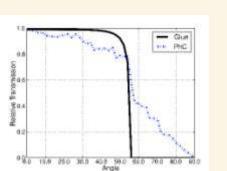
### E.Bourret, LUMDETR 2018

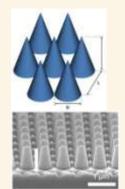
#### A.Knapitsch, E.Auffray et all



## Photonic crystals on scintillators







Surface tuning from nano to macro scale allows to modify light output distribuiton and yield

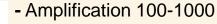
## **Photoreceivers. PMT alternatives**



#### **PMTs**

- Made of glass (fragile + K-40 background)
- Large signals, good S/N ratio, fast (ns)
- Large dimension, low price per cm<sup>2</sup>
- Sensitive to B fields
- Existing old technology (vacuum tubes)

### APDs



- rather Unstable (temp)
- rel expensive, small (max 10x10mm)

- PMTs
- (PIN) Diodes
- Avalanche Photodiodes (APDs)
- · (Drift Diodes)
- (EM) CCDs
- Si-PMs (MPPCs)

#### PIN diodes

No amplification (small signals)

- Maximum cm size
- Stable (temperature)

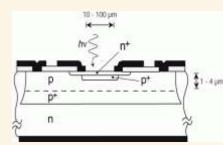
### CCDs :

- DC measurement mostly
- Imaging
- For higher radiation fields

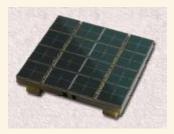
### Drift diodes (for light detection)

- small (not often used)
- still rel. expensive

#### Silicon Photomultipliers (SiPMs, MPPCs)







### Fast decay importance...

- A fast signal allows high rates (not of interest) and good timing resolution This is of interest...
- Timing resolution allows coincidence measurements to be made
- With knowledge of the decay scheme, coincidence measurements can be used to identify specific radionuclides and reject background events
- Requirements of decay scheme:
- $E\gamma > ~100$  keV, short lifetime of intermediate state and high probability of  $\gamma$ -emission (low internal conversion coefficient)

## Outlines

- 1. The market needs are the main driving force for scintillator development
- 2. Different application claim for specific development. Some materials and engineering could be very specific or even unique.
- 3. There are too many materials were invented last yeas and industry need in only few ones. These materials have to satisfy optimal cost/performance/volume rate
- 4. Main efforts in material study/development (like co-doping, resolution improvement, electronics upgrade and so on) are directed either to
  - \* advanced detector development or
  - \* cost efficient technology development
- 5. New trends in scintillation development (Fast detectors, 3D printing, Meta materials, n-gamma discrimination, SiPMT, cheap ceramics) will dominate in new detector
- 6. Conventional scintillator market is waited for improved nonproportionality and energy resolution.

## Thank you for attention!

## **GAGG** scintillator as the leader through oxides

### GAGG (Gd<sub>3</sub>Al<sub>2</sub>Ga<sub>2</sub>O<sub>12</sub>) Ce

- 6.6 g/cc

- 520 nm max emission

- 56000 photons / MeV

Proportional, not hygroscopic FIRST non hygroscopic High LO crystal

High melting point (1850 °C) → cost

Not used frequently yet SiPm readout ? Special applications ?



No new generation scintillator will be the "ideal" material = illusion

Often bottom line is COST = growing yield

(material cost is seldom the real issue)



59.54 keV

## **Co-doping in Scintillators**

40

241 Am

Aliovalent codoping: impact on light yield and energy resolution

defects

afterglow

number of counts (arb. units) 35 Sr-co-doped LaBr<sub>3</sub>:Ce 30 25 R=6.5% 20 15 standard 10 5 0 50 60 70 20 30 40 10 (keV) Energy Blahuta et al. IEEE TNS 60 (2013) 3134 Left: Reduction/ compensation of LYSO:Ce LYSO:Ce.Mg 2x10 LYSO:Ce.Ca counts) Right: Reduction of Inten  $1 \times 10^6$ 

200

100

300

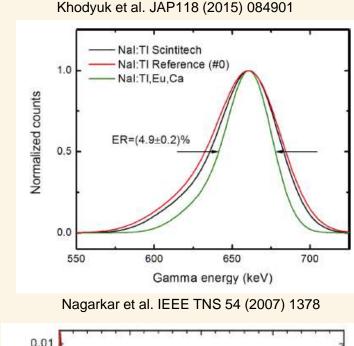
Temperature (K)

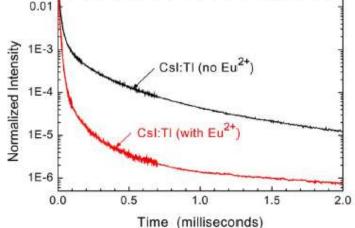
400

500

600

Alekhin et al. APL 102 (2013)161915





Photons generated at different terms of scintillation give different contribution to the energy resolution.

- Pulses clustering does refines its statistical description.
- Using of clustering and NN-models is perspective for development of new base of knowledge about functional materials.
- Digital signal processing can improve the energy resolution.