# Recent advancements in inorganic scintillator detectors

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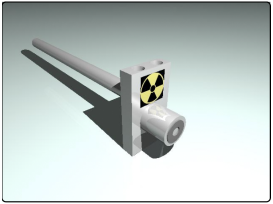
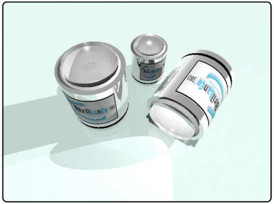
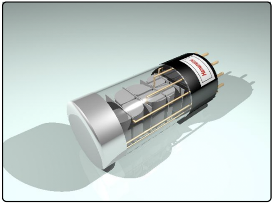
RIKEN, Japan

# Outline

# Triptic

## Excitation sources

- Gamma-ray
- Neutrons
- Alpha...

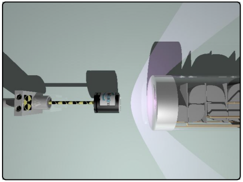
## Samples

- Single crystal
- Ceramic
- Composite...

## Detectors

- PMT
- APD
- HybridPMT...

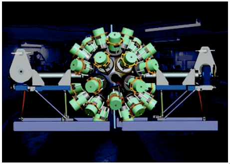
# Various and Numerous



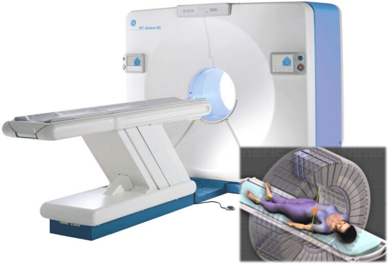
## Common goal

Convert the high energy of the particles or $\gamma$-ray into detectable light

## $\gamma$-ray spectroscopy



## Medical imaging



# Scintillator property

## Ideal scintillator

Ideal scintillator should have all these properties:

1. **Performance**
   - Absorption ⇒ **Density**
   - Efficiency ⇒ **Light output**
   - Speed ⇒ **Decay time, Rise time**
   - Accuracy ⇒ **Energy resolution**
2. **Cost**
   - Cheap ⇒ **Crystal growth yield**

**Ideal scintillator does not exist**

Application dependent requirements: different for High energy physics, homeland security or medical imaging applications

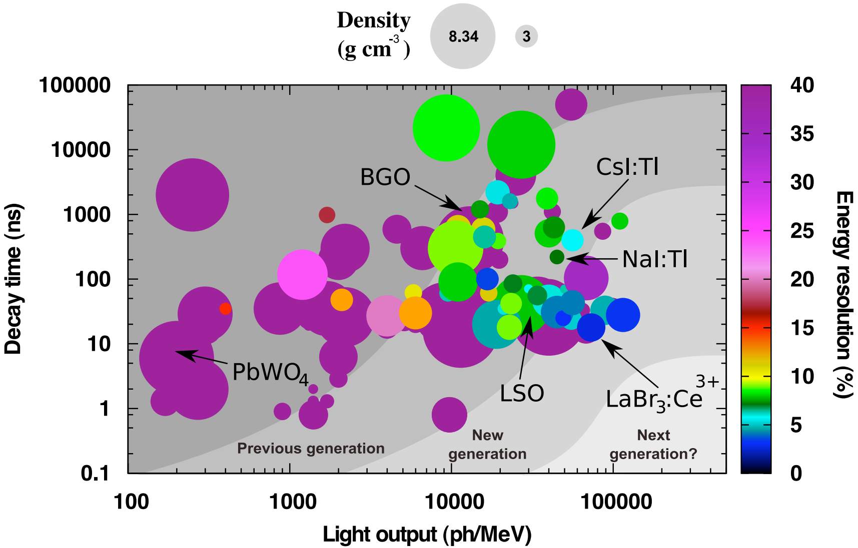
# Requirements

A next generation, Scintillator-based High-resOlution Gamma spectrometer for Unstable Nuclei (SHOGUN) is proposed that is ideally suited for in-beam experiments with fast beams of rare isotopes at the RIBF with velocities of 5060% of the speed of light. The array will be based on the novel scintillator $LaBr_3(Ce)$, which has exceptional properties making it the ideal material for such a spectrometer. Its very high light output and the very short decay time of the scintillation light results in an unprecedented energy resolution for a scintillation counter and a time resolution that is much faster than that of NaI(Tl) or HPGe detectors. Furthermore, due to the large attenuation coefficient for $\gamma$-rays, a compact array can achieve a very high full-energy peak efficiency.

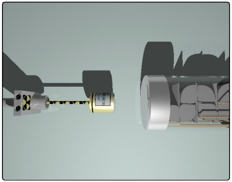
## Ideal scintillator

$LaBr_3$ scintillator?
Can we do better?

# Where did we stand



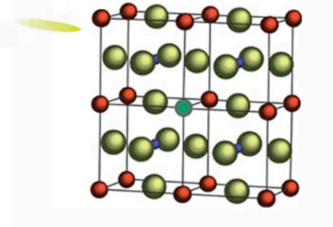
# Step 1/5



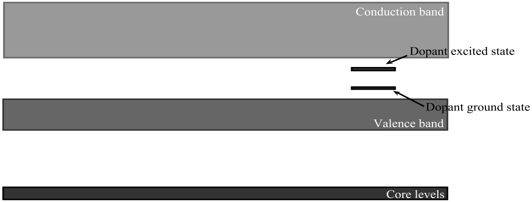
## Absorption mechanism

$\gamma$-ray interacts with the lattice only

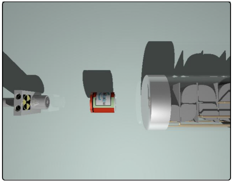
## Structural scheme



## Energy scheme



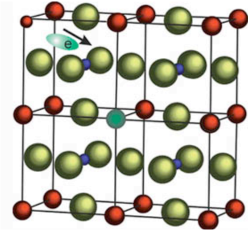
# Step 2/5



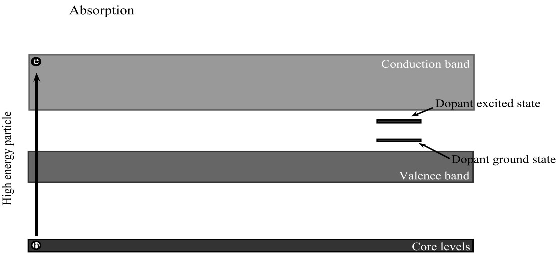
## Absorption mechanism

$\gamma$-ray is converted into a high energy electron

## Structural scheme



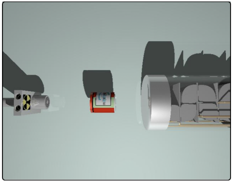

## Energy scheme



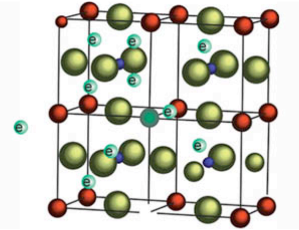
## Step 3/5

### Multiplication mechanism

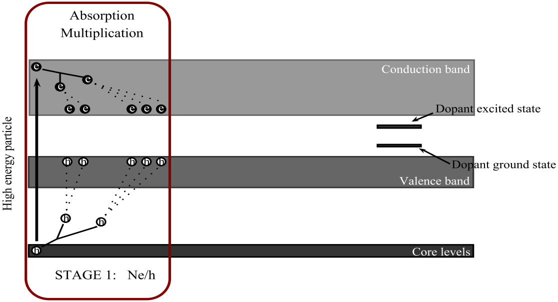
High energy electron creates e/h pairs loosing its energy



### Structural scheme



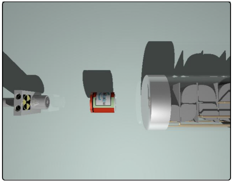
### Energy scheme



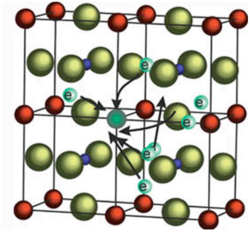
# Step 4/5

## Migration mechanism

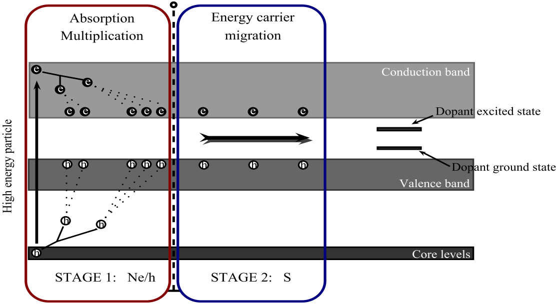
e/h pairs migrate toward the dopant ions



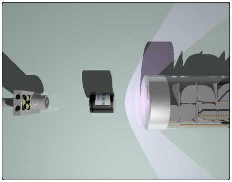
## Structural scheme




## Energy scheme



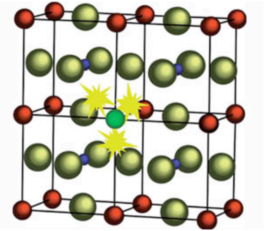
# Step 5/5



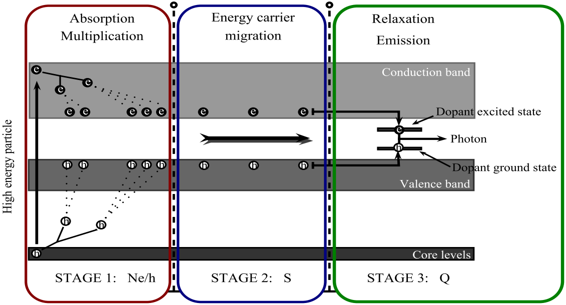
## Emission mechanism

e/h pairs recombine on the dopant ions

## Structural scheme



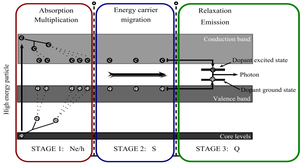
## Energy scheme



# Light output (1/3)

## Formula

- $LO = \frac{1{,}000{,}000}{\beta E_g} SQ$

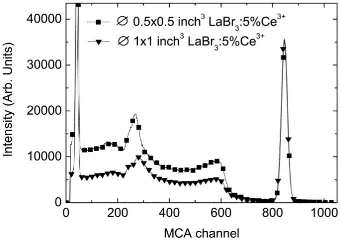



## Estimation for $LaBr_3$

- $E_g$=6.2 eV
- $\beta \sim 2$

$\Rightarrow$ **$LO_{estimated}$ = 81,000 ph/MeV**

$\Rightarrow$ **$LO_{measured}$ = 60,000 ph/MeV**



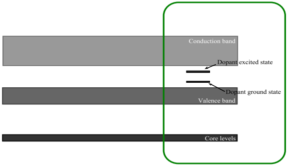

**Ideal scintillator: No**

Energy loss as large as 20% - **S or/and Q < 1**

# Light output (2/3)

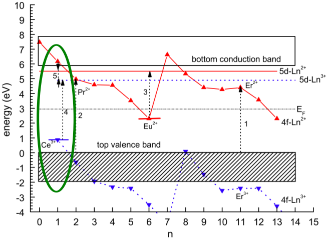
## Quantum efficiency, $Q$

- $Ce^{3+}$ energy levels vs Valence and Conduction bands




## $LaBr_3$ energy level positions

- Spectroscopy
- First principle calculation




$\Rightarrow$ **4f/VB $\sim$ 0.5 eV**

$\Rightarrow$ **5d/CB $\sim$ 1 eV**

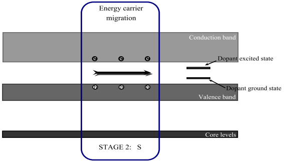
**Ideal scintillator: No**

Energy loss due to energy transfer - $S < 1$, $Q = 1$

# Light output (3/3)

## Transfer efficiency, $S$

- Self Trapped Exciton (STE) to $Ce^{3+}$ transfer

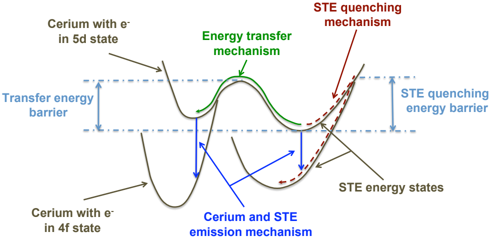



## STE properties dependence

- 2 different STEs
- 1 does not transfer entirely

⇒ **Transfer is efficient**

⇒ **But not perfect**




**Ideal scintillator: No**

Light output can be improved: better $S$

# Time response

## Formula

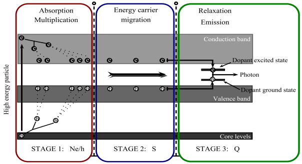
- $$\frac{1}{\tau_{Scintillation}} = \frac{1}{\tau_{Transfer}} + \frac{1}{\tau_{Dopant}}$$

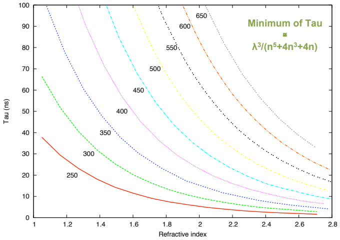
## $\tau_{Dopant}$ estimation for $LaBr_3$

- $\tau_{Ce} \sim \frac{\lambda^3}{n^5+4n^3+4n}$
- $n \sim 1.8$

$\Rightarrow \tau_{estimated} = 20$ **ns**

$\Rightarrow \tau_{measured} = 18$ **ns**




**Ideal scintillator: Yes**

No delay due to the transfer. Ideal time response

# Energy resolution (1/2)

## Formula

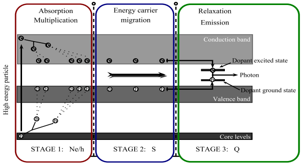
- $$E.R. = \frac{2.35}{\sqrt{Number_{photoelectron}}}$$

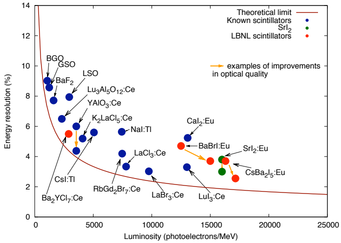
## E.R. estimation for $LaBr_3$

- $Number_{photoelectron} \sim 9{,}500$

$\Rightarrow$ **$E.R._{Estimated} = 2.4\%$**

$\Rightarrow$ **$E.R._{measured} = 2.8\%$**




**Ideal scintillator: Yes**

Ideal energy resolution at 662 keV

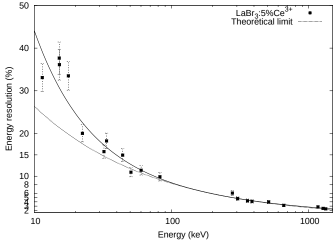
# Energy resolution (2/2)

## E.R. vs Energy

- Degradation of E.R. under 50 keV

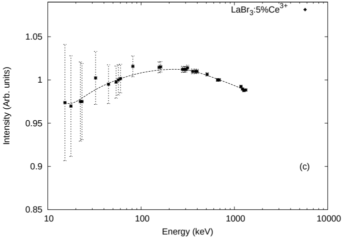
$$\Rightarrow \mathbf{E.R.}^{662keV}_{measured} = \mathbf{E.R.}^{662keV}_{theoretical}$$

$$\Rightarrow \mathbf{E.R.}^{10keV}_{measured} = \mathbf{1.5\ E.R.}^{10keV}_{theoretical}$$




## Non proportionality

- E.R. increases with increasing non proportionality




**Ideal scintillator: Yes**

Deviation of theoretical energy resolution under 50 keV

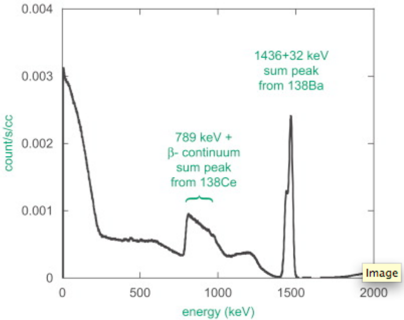
# Intrinsic activity

## $^{138}$La $\gamma$-ray background

⇒ **Lanthanum $\in$ 0.09% $^{138}$La**

⇒ **Half-life of 1.05E$^{11}$ years**

⇒ **1.5 Bq/cc in $LaBr_3$**




## Energy spectrum

- $^{138}$Ba by electron capture: Gamma-ray at 1436 keV + 32 keV Ba x-ray
- $^{138}$Ce by $\beta$-emission: Gamma-ray at 789 keV

**Ideal scintillator: No**

Application relevancy

# Cost

## Structural parameter

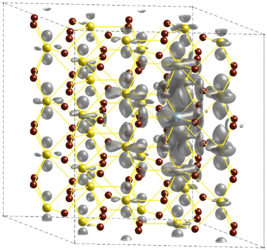
⇒ Density [g/cm3] = 5.08

⇒ Melting point [K] = 1116

⇒ Thermal expansion coefficient $[10^{-6}/C] = 8$ along C-axis

⇒ Cleavage plane: <100>

⇒ Hygroscopic: High



**Ideal scintillator: No**

Crystal growth yield is low.

# $LaBr_3$ summary

## Pro

- **Time response:** 18 ns and Time resolution: 0.450 ns
- **Energy resolution:** 2.8% at 662 keV
- **Light output:** 60,000 ph/MeV

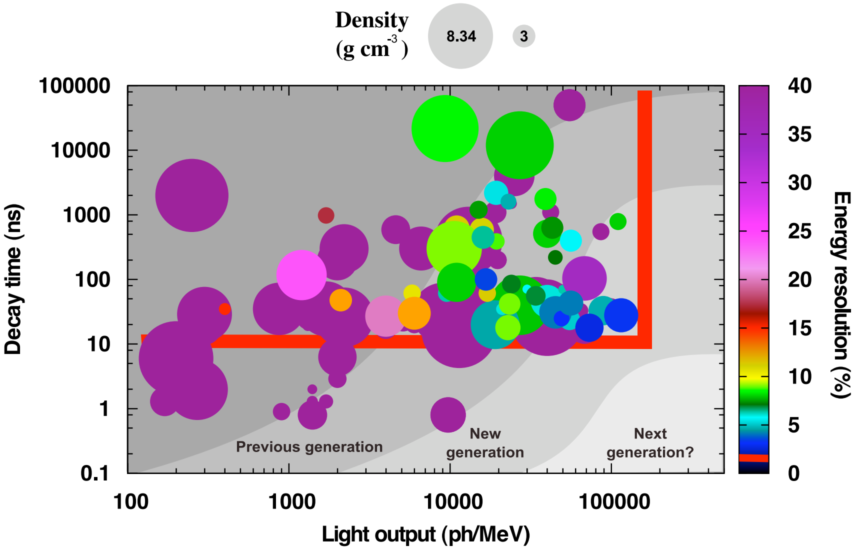
## Con

- **Cost:** thermal anisotropy leading to low growth yield
- **Light output:** substantial energy loss
- **Energy resolution:** Degradation in the low energy range
- **Hygroscopicity**: High
- **Intrinsic radioactivity**: $^{138}$La $\gamma$-ray background

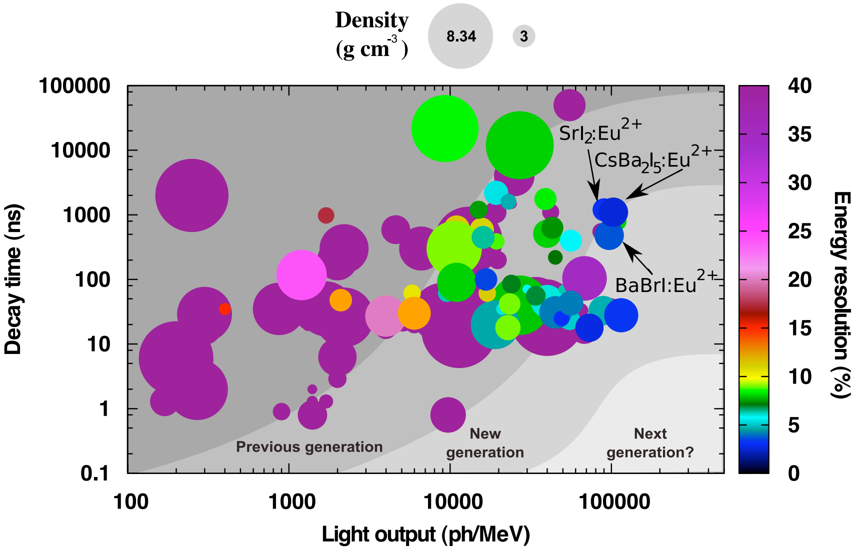
**Yes but...**

Can we do better?

# Yes, we can


Density
(g cm$^{-3}$)
8.34
3
100000
10000
1000
100
10
1
0.1
Decay time (ns)
100
1000
10000
100000
Light output (ph/MeV)
Previous generation
New generation
Next generation?
40
35
30
25
20
15
10
5
0
Energy resolution (%)

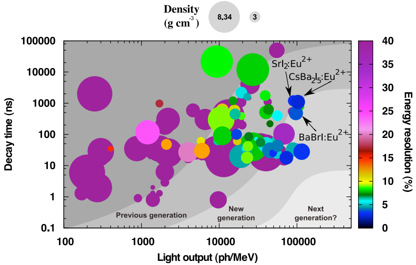
# Where do we stand



# New scintillators

## Recent development

- CLLB
- $SrI_2$
- $CsBa_2I_5$ and BaBrI



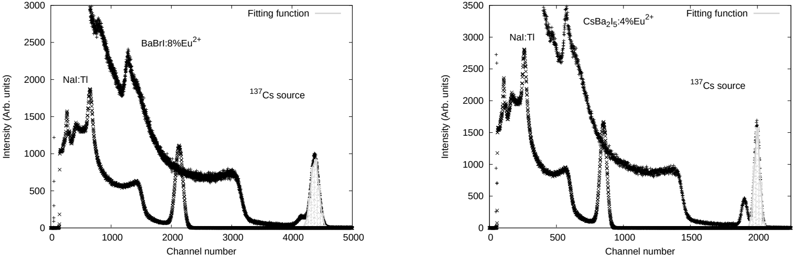

## Comparatif

| Scintillator | NaI:Tl | $LaBr_3:Ce^{3+}$ | $CLLB:Ce^{3+}$ | $SrI_2:Eu^{2+}$ | $CsBa_2I_5:Eu^{2+}$ | $BaBrI:Eu^{2+}$ |
|---|---|---|---|---|---|---|
| Development (year) | 60 | 10 | 4? | 4? | 1 | 1 |
| Structure | Cubic | Hexa | Cubic | Ortho | Mono | Ortho |
| Band gap (est. in eV) | 5.9 | 6.2 | 5.8 | 5.4 | 5.1 | 5.3 |
| Density (g $cm^{-3}$) | 3.67 | 5.1 | 4.2 | 4.5 | 5.0 | 5.0 |
| Decay time (ns) | 230 | 17 | 55 | 1,200 | 1,400 | 500 |
| Luminosity (ph/MeV) | 42,000 | 60,000 | 50,000 | 100,000 | 103,000 | 91,000 |
| Energy resolution (%) | 6.5 | 2.8 | 3.0 | 3.0 | 2.55 | 3.3 |
| Detection efficiency (%) | 30 | 26 | - | 31 | 36 | 35 |
| Hygroscopicity | High | High | - | High | Low | Low |

# Light output

## Criteria

- Band gap
  $BaBrI:Eu^{2+} \sim 5.3$ eV, $CsBa_2I_5:Eu^{2+} \sim 5.1$ eV
- Estimated Light output
  $BaBrI:Eu^{2+} \sim 95{,}000$ ph/MeV, $CsBa_2I_5:Eu^{2+} \sim 100{,}000$ ph/MeV
- Measured Light output
  $BaBrI:Eu^{2+} \sim 91{,}000$ ph/MeV, $CsBa_2I_5:Eu^{2+} \sim 100{,}000$ ph/MeV




**$LaBr_3$ vs BBI, CBI**

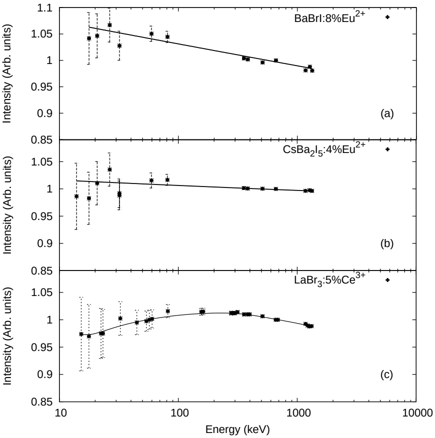
Smaller band gap and better transfer efficiency

# Proportionality

## Gamma-ray response

## Benchmark

- $\sigma_{np} = \sqrt{\frac{1}{N}\sum_{i=1}^{N}\left(\frac{Y(E_i)}{Y(^{137}Cs)} - 1\right)^2}$




$\Rightarrow \sigma_{np}(BaBrI) = \mathbf{0.024}$

$\Rightarrow \sigma_{np}(CsBa2I5) = \mathbf{0.009}$

$\Rightarrow \sigma_{np}(LaBr3) = \mathbf{0.013}$

**$LaBr_3$ vs BBI, CBI**

Better response for CBI

# Energy resolution

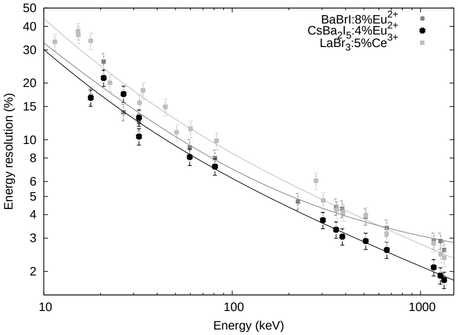
## Energy resolution at 662 keV

$\Rightarrow$ **E.R.**$^{662keV}_{BaBrI}$ **= 3.3%**

$\Rightarrow$ **E.R.**$^{662keV}_{CsBa2I5}$ **= 2.55%**

## Energy resolution vs Energy

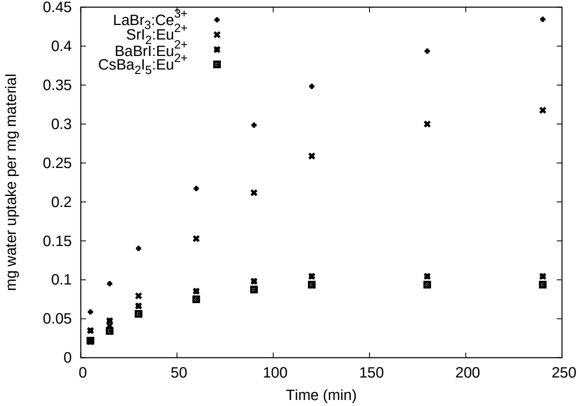
- Both scintillators give better E.R. under 50 keV




**LaBr$_3$ vs BBI, CBI**

Better energy resolution for CBI and BBI under 50 keV

# Hygroscopicity



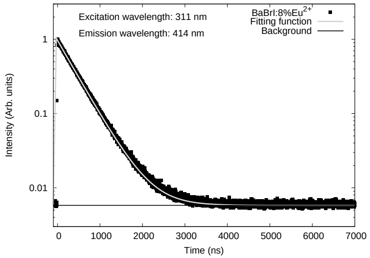

## $LaBr_3$ vs BBI, CBI

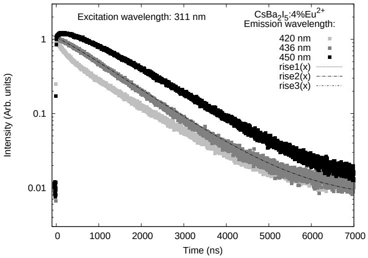
Hygroscopicity is less in both cases

# Time response

## Criteria

- Time response
  $BaBrI:Eu^{2+}$ ~ 450 ns, $CsBa_2I_5:Eu^{2+}$ ~ 1,200 ns
- Self absorption
  $BaBrI:Eu^{2+}$ ~ Not detected, $CsBa_2I_5:Eu^{2+}$ ~ Medium

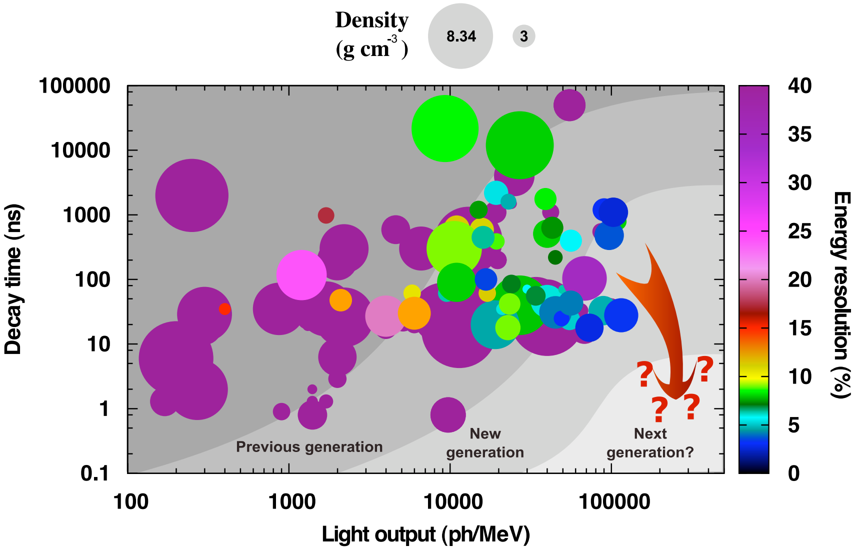



### $LaBr_3$ vs BBI, CBI

Slower than $LaBr_3$ but excellent for $Eu^{2+}$ doped compound

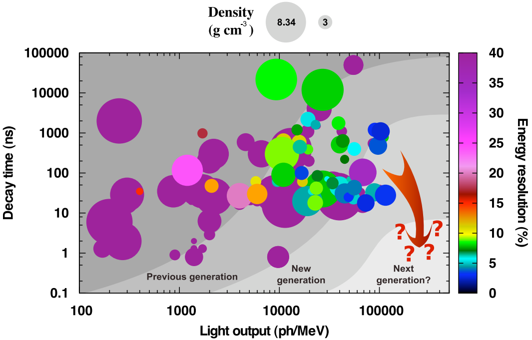
# New grounds (1/2)



# New grounds (2/2)

## Problems

- Limits are intrinsic to doped inorganic insulator
- Dopant intrinsic time response limits the decay time
- Dopant energy levels limit the band gap range available




## Approaches

| Description | Application | Advantages | Draw back |
|---|---|---|---|
| Semi conductor scintillators | Cryogeny | Fast | Thermal stability |
| Nano and composite scintillators | Any | Fast, low cost | How to absorb energy? |

## Fast moving research field

- **Large improvement within the past 5/10 year**
  - Understanding the physics of scintillators
  - Application via the improvement and discovery of high performance scintillators
  - Exploration of new concepts in solid state detectors

## Multidisciplinary collaboration

- Computational science
- Chemistry
- Physics
- Crystal growth
- Instrumentation...

# Collaborators

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