

Seminar at LNGS, July 23, 2015

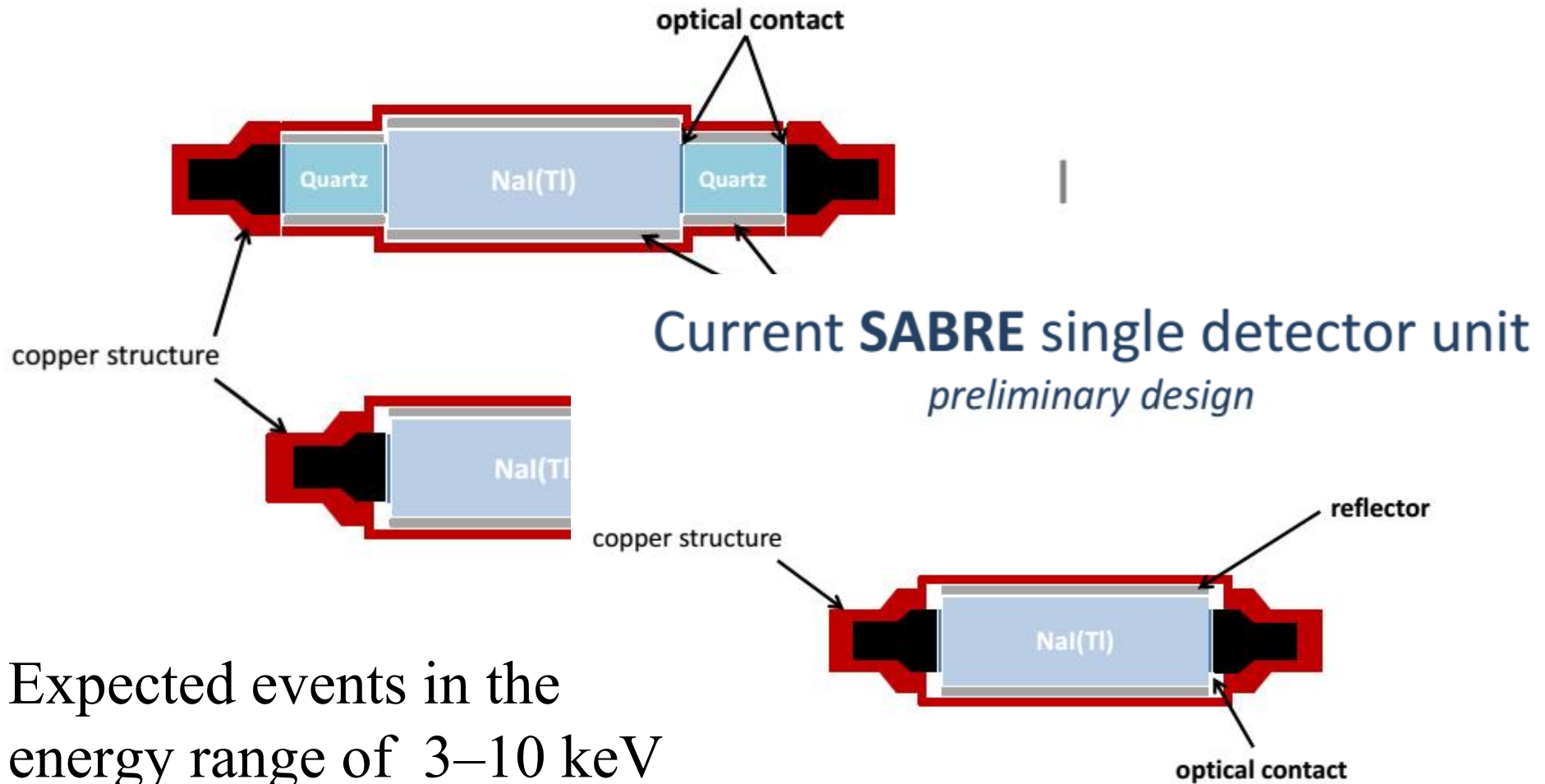
**The NaI:Tl and CsI:Tl crystals for effective  
detection of X-rays and low energy charged  
particles**

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July 22, Assergi, Italy

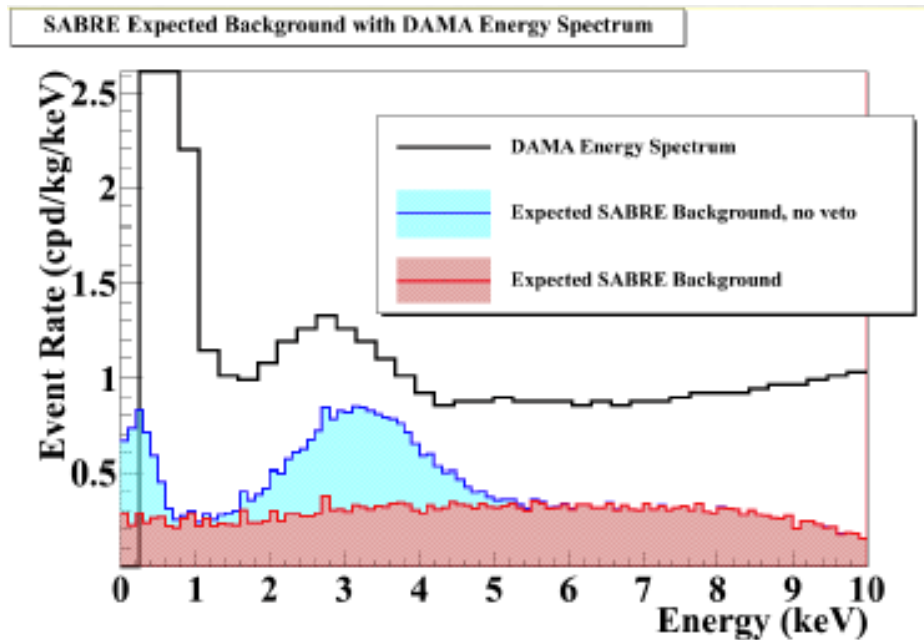
# DAMA single detector unit



Expected events in the energy range of 3–10 keV

**Design like DAMA II stage....  
Why not think about how to improve it?**

# In DAMA/SABRE experiments expected energy range is 2-4 keV



See Talk by E. Shields at TAUP2013

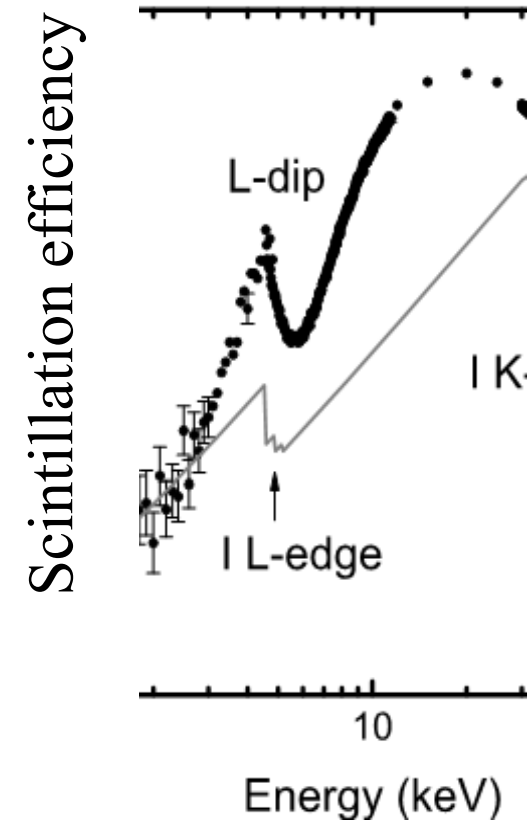
## Internal Source: 0.84 keV

*McCann M.F. and Smith K.M.*

On the Detection of 1 keV Events in NaI:Tl  
NIM, 65 (1968) 173.

Characteristic X-rays

Figure from: *I. Khodyuk and  
P. Dorenbos, IEEE TNS, 2012*

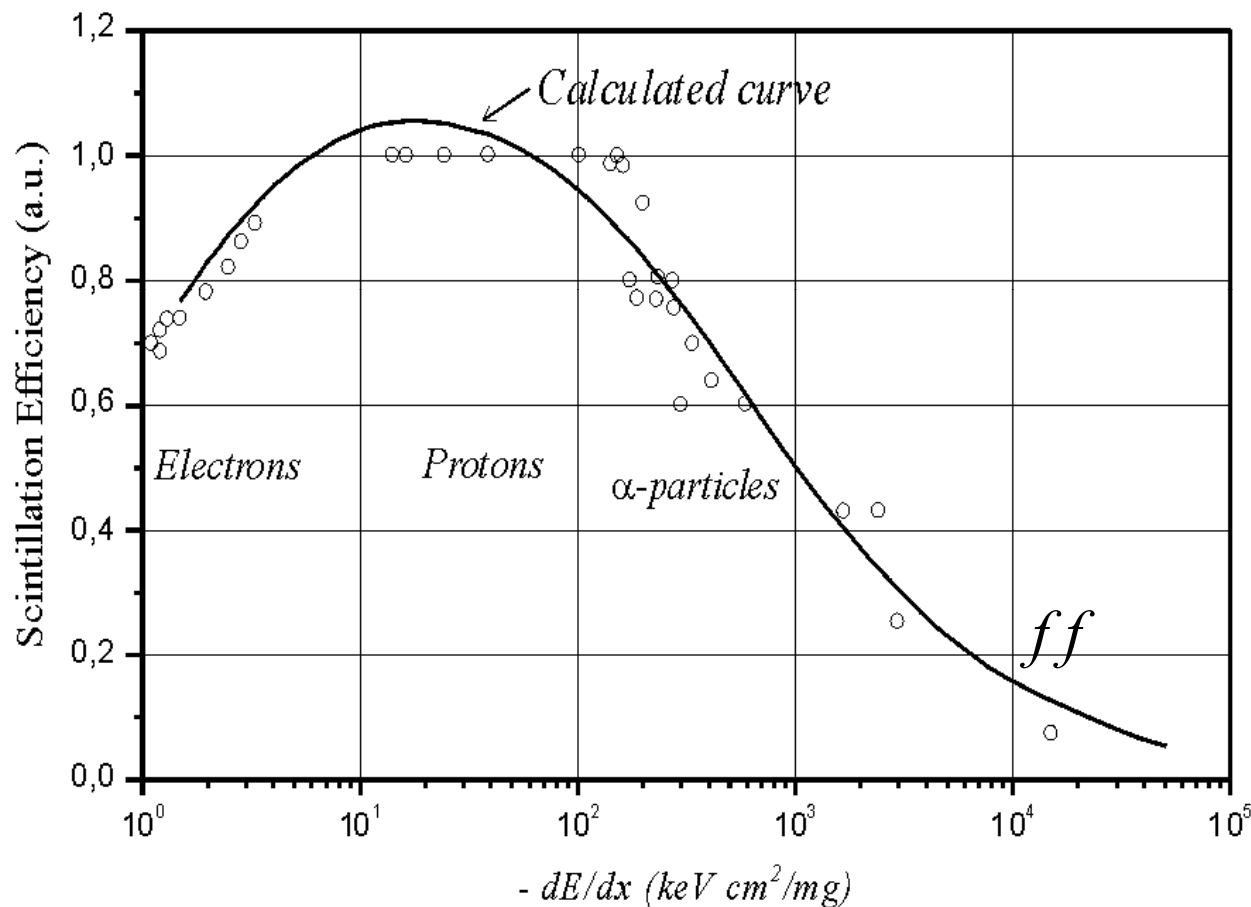


NaI:Tl crystal  
External source of  
synchrotron radiation

# Problems of low energy particle detection

Energy resolution and  
non-proportionality of response

Light yield depends on  $dE/dx$

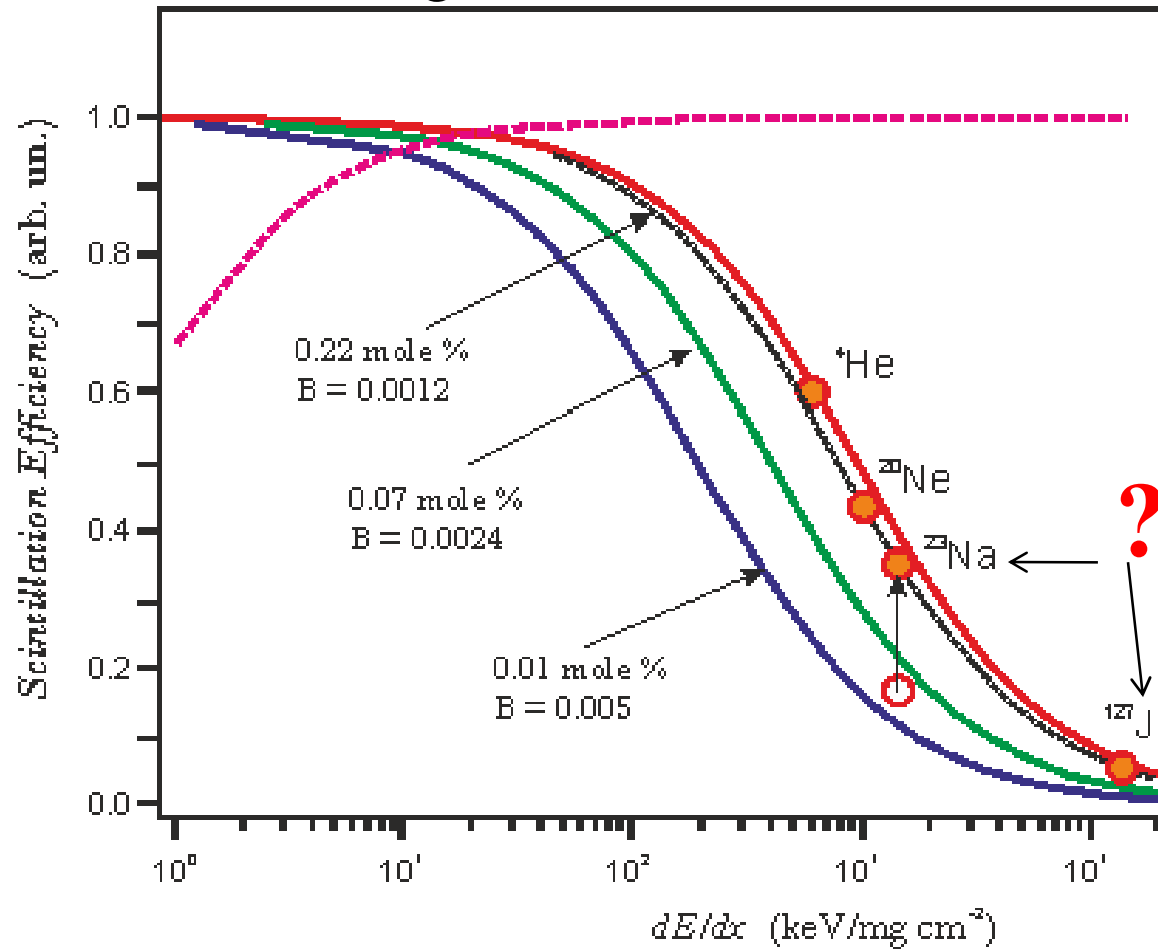


$QF$  – quenching  
factor

**Fission fragments ( $ff$ )  
have only 10% of  
maximum light yield,  
 $QF \sim 0.1$**

# Dark matter search with NaI and CsI

According to Birks model

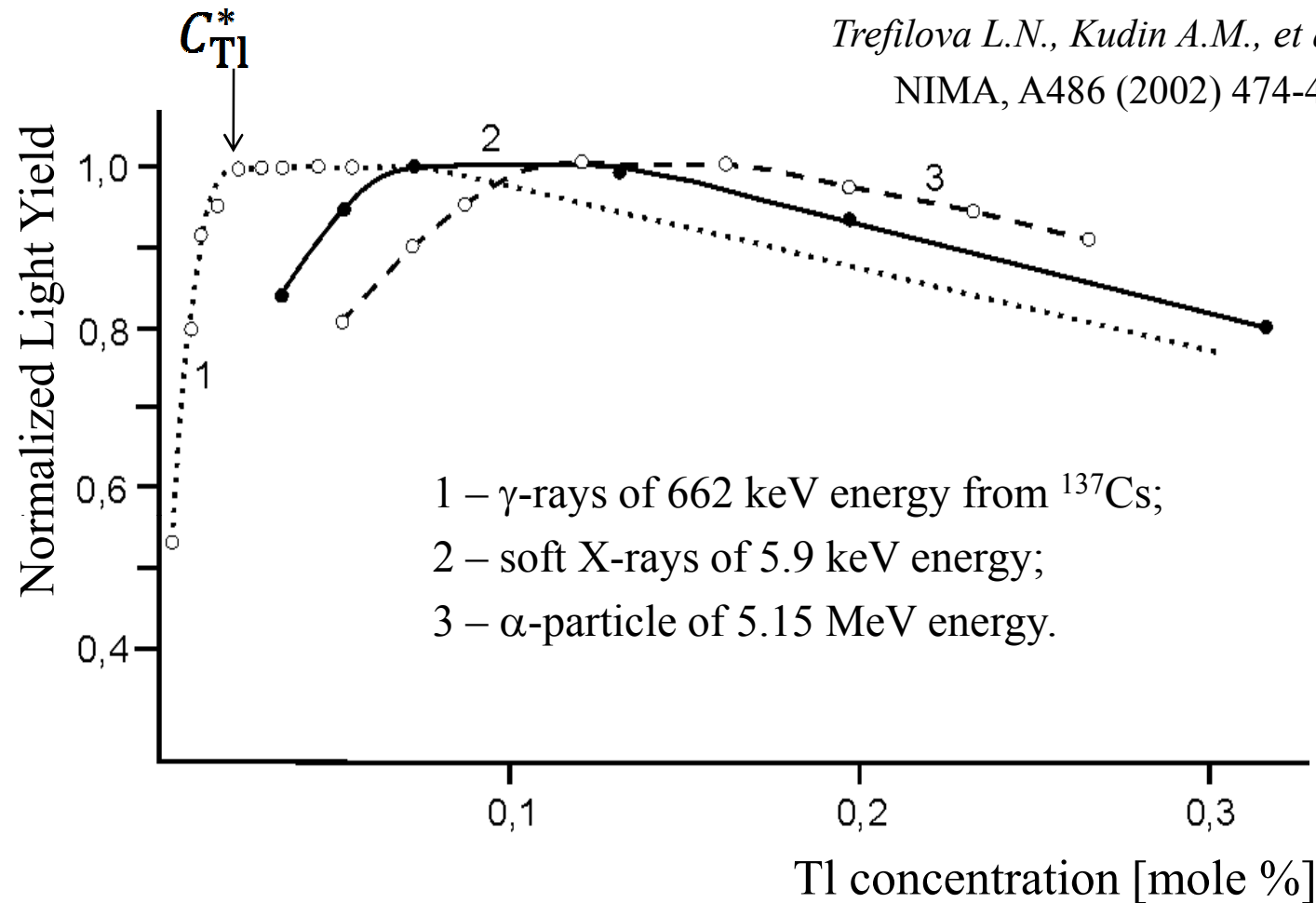


Quenching factor  
for <sup>23</sup>Na and <sup>127</sup>I

Protons	100 %
Alphas	60 %
<sup>20</sup> Ne	42 %
it can be estimated	
<sup>23</sup> Na	~ 35%
<sup>127</sup> I	~ 8 %

Quenching factor depends on Tl concentration

# Optimum Tl concentration ( $C_{Tl}^*$ ) in NaI:Tl



$$C_{Tl}^* = 2.2 \cdot 10^{-2} \% \text{ for } \gamma\text{-rays};$$

$$C_{Tl}^* = 7.3 \cdot 10^{-2} \% \text{ for soft X-rays of 5.9 keV};$$

$$C_{Tl}^* = 1.3 \cdot 10^{-1} \% \text{ for } \alpha\text{-particles}.$$

## Requirements to scintillation material for charged particles and light ions detection:

- maximum scintillation efficiency to photons detection;
- high transparency to rich maximum light collection coefficient;
- homogeneity of thallium distribution and other dopands to rich best value of energy resolution;

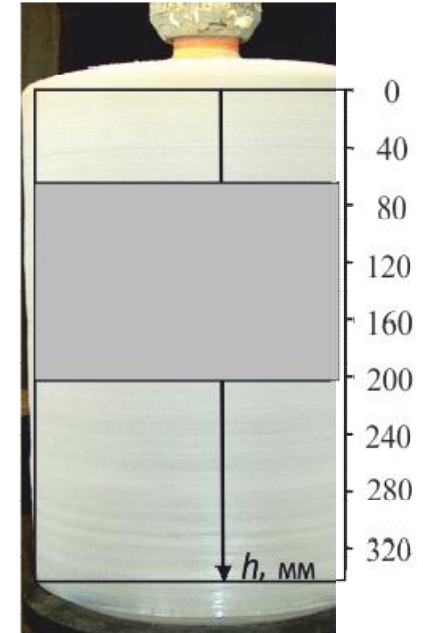
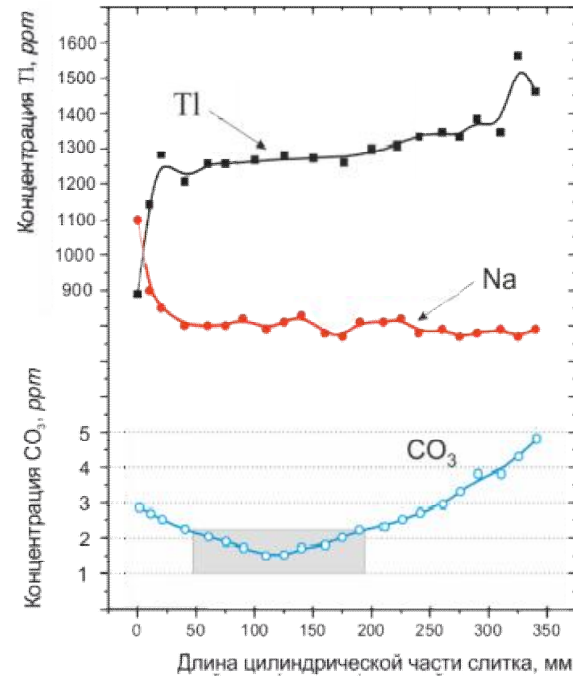
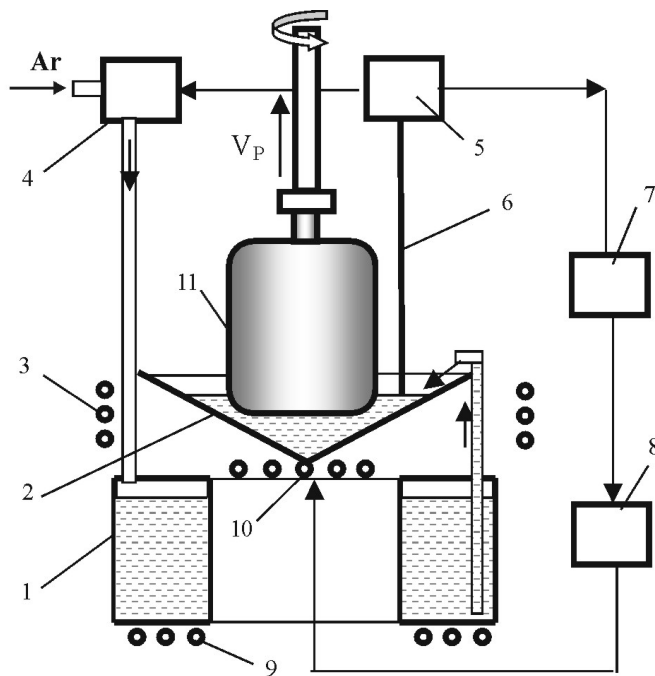
### **Additional:**

- increased Tl concentration to rich best scintillation efficiency for charged particles detection;
- stability of surface state;
- absence of dead layer.

$C_{Tl}^* > 0.15 \%$  for ion detection in NaI:Tl

# NaI:Tl crystal for particle detection: homogeneity

Furnace for crystal growth with conical crucible "Crystal-400"



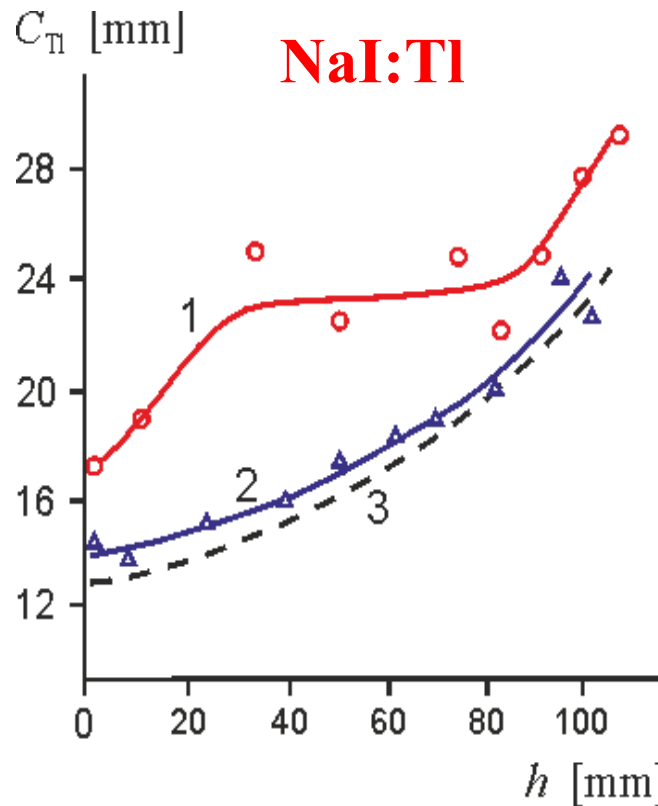
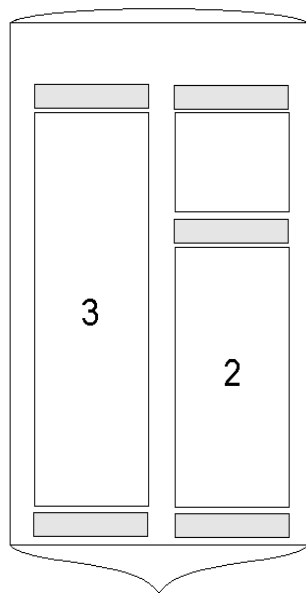
Distribution of Tl and other co-activators in scintillation material "CsI:Tl,Na,CO<sub>3</sub>" along height.

The same is thru for NaI:Tl



# Impurity distribution in crystal grown by Bridgman-Stockbarger technique

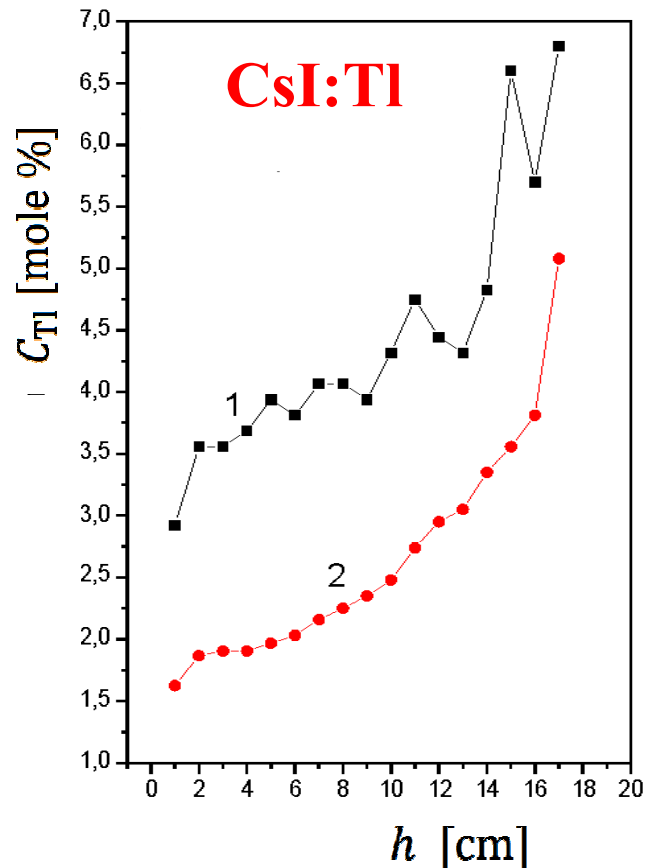
$$C_{\text{Tl}} = C_0 k_0 \left(1 - \frac{V}{V_0}\right)^{k_0 - 1} \quad k_0 - \text{equilibrium segregation coefficient}$$



- 1 – crystal growth in vacuum;
- 2 – crystal growth in oxygen;
- 3 – calculated curve for  $k_0 = 0.25$   
 $C_0 = 0.36 \%$

In heavy doped crystal activator is distributed non-uniformly

# Non-homogeneous distribution of activator in CsI:Tl (microscopic)



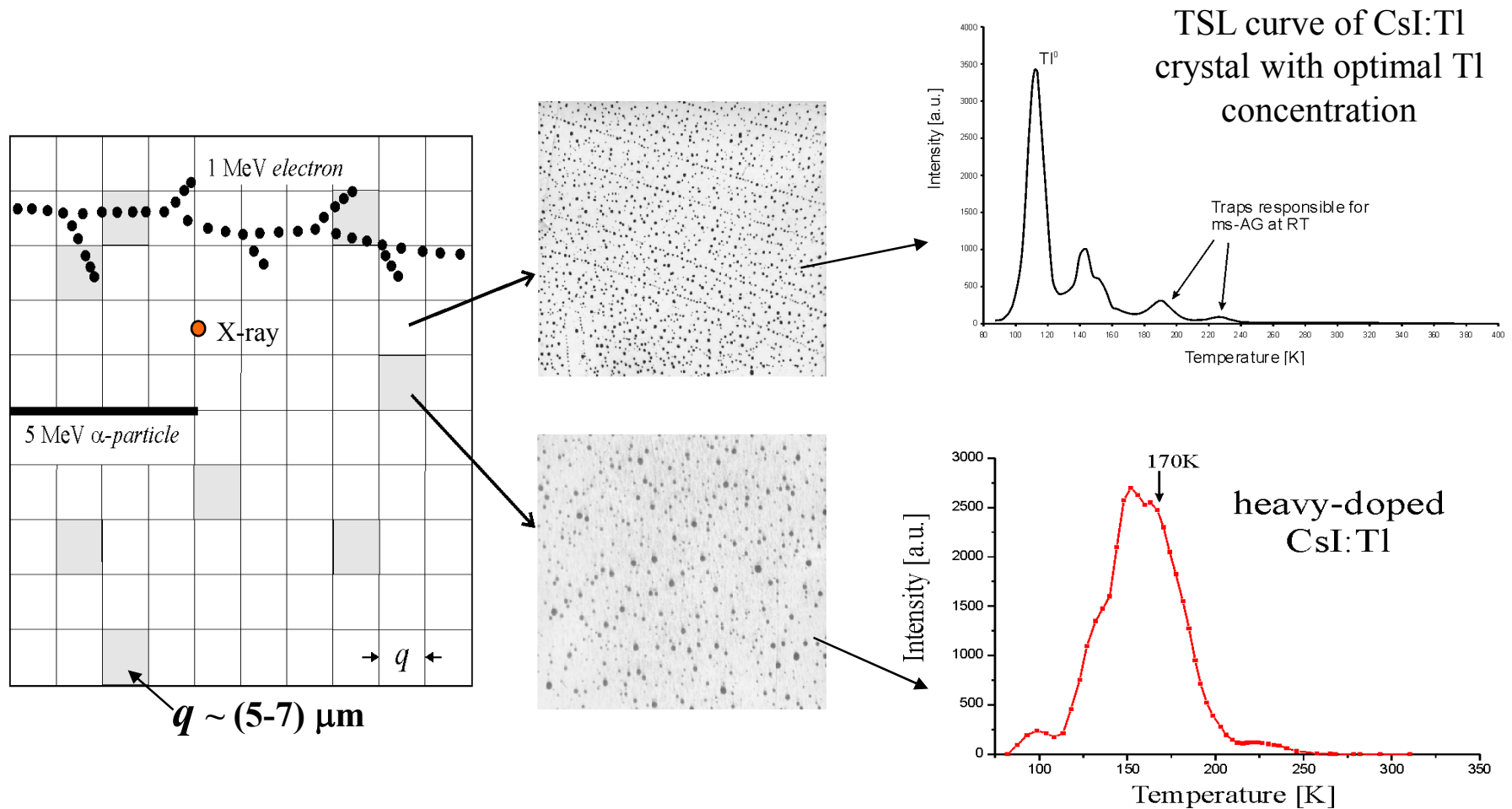
**Table: PIXE analysis results**

Brand	CsI n.	Nominal Tl conc.(ppm)	Measured Tl conc. (ppm)	Type of measure
GB	1	4000	6400±200	Face A, av
GB	1	4000	9300±300	Face A, point
GB	1	4000	5400±200	Face A, point
GB	1	4000	6100±200	Face B, av
GB	1	4000	4610±180	Side, av
GB	2	3000	2950±110	Face A, av
GB	2	3000	4900±200	Face B, point
GB	2	3000	3030±120	Face B, av
St. Gobain	3	500	440±50	Face A, av
St. Gobain	4	200	280±30	Face A, av
Marketech	5	700	520±30	Face A, av
Scionix	6	2500	5220±160	Face A, av

**FAZIA collaboration results**

In  $C_{Tl} > 0.2\%$  the activator is not homogeneous distributed both macroscopically and microscopically

# Nature of concentration quenching



**Schematic image of microscopic distribution of Tl<sup>+</sup> center in CsI crystal at high Tl concentration. Photo represents the character of decoration of the cleavage plane in two different places. (Electron microscope;  $\times 16\ 000$ ; decoration by gold). Black squares correspond to places of increased Tl content (so-called spinodal decay of solid solution)**

# Uniformity of spectrometric parameters

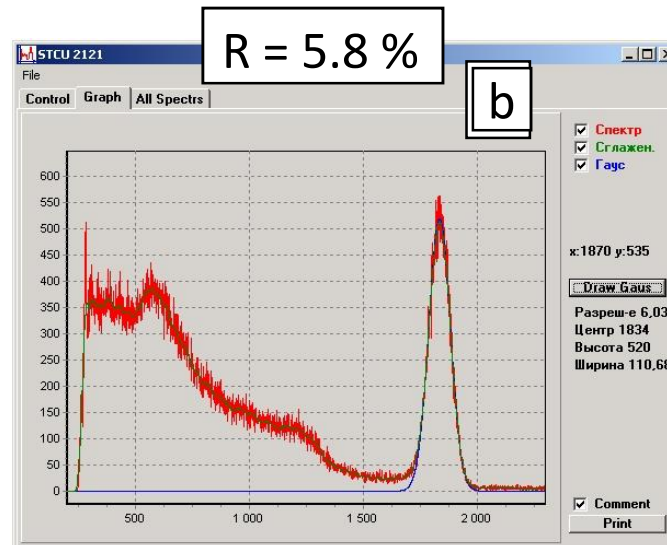
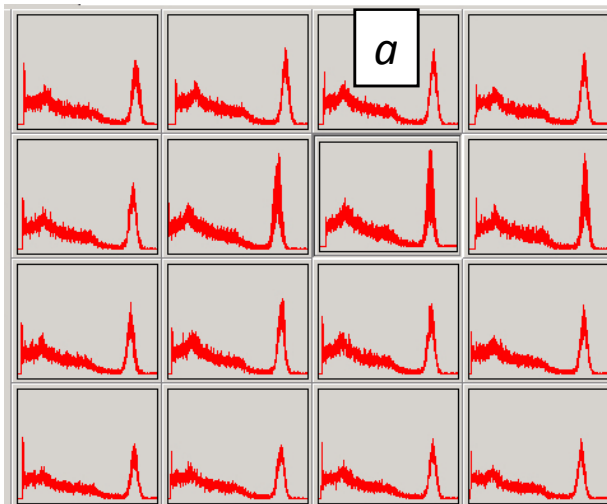
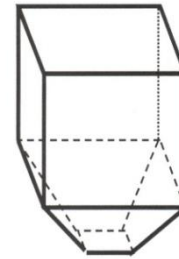
Photodiode scintillator of 200 cm<sup>3</sup> volume.

16 sample from selected region of ingot.

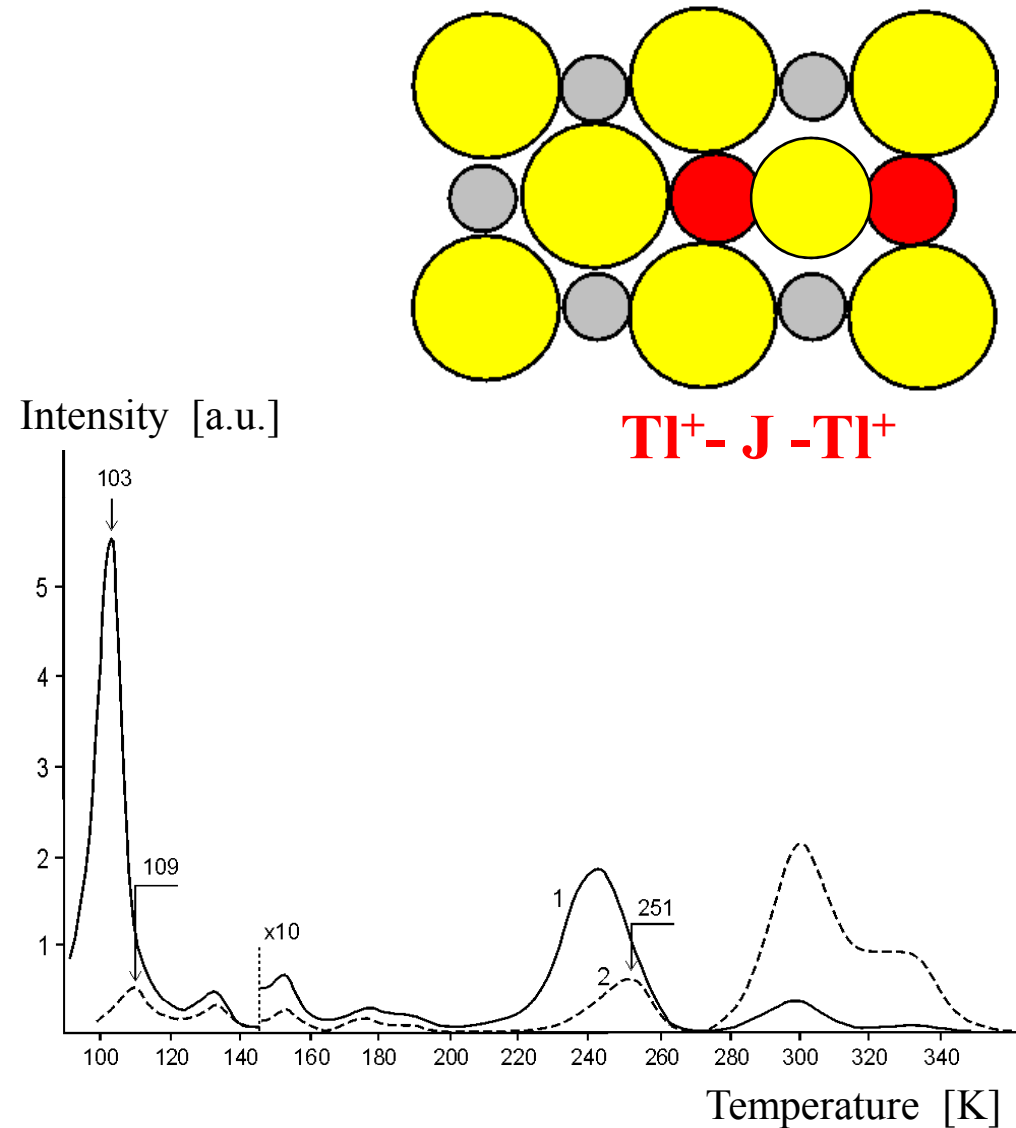
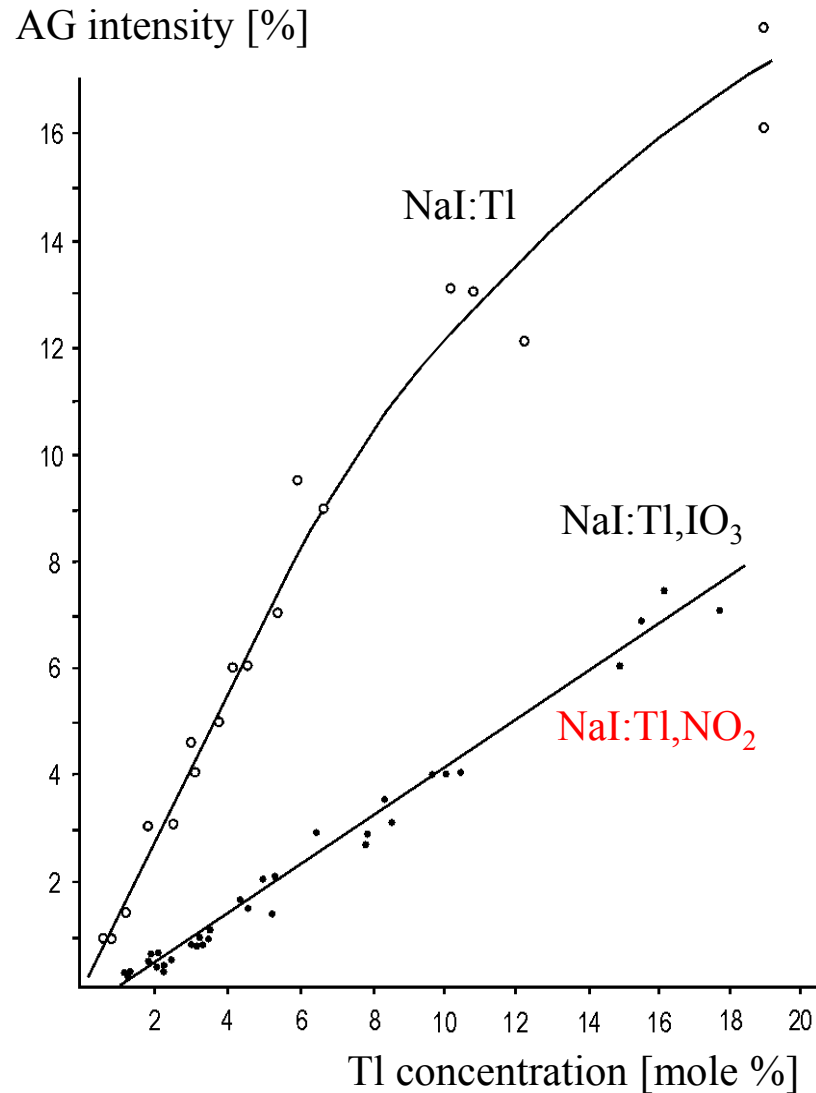
CsI:Tl ingot of 240 mm dia. and 360 mm  
height

Pulse height spectra for each element (left)  
and summarized spectrum of whole block  
(right).

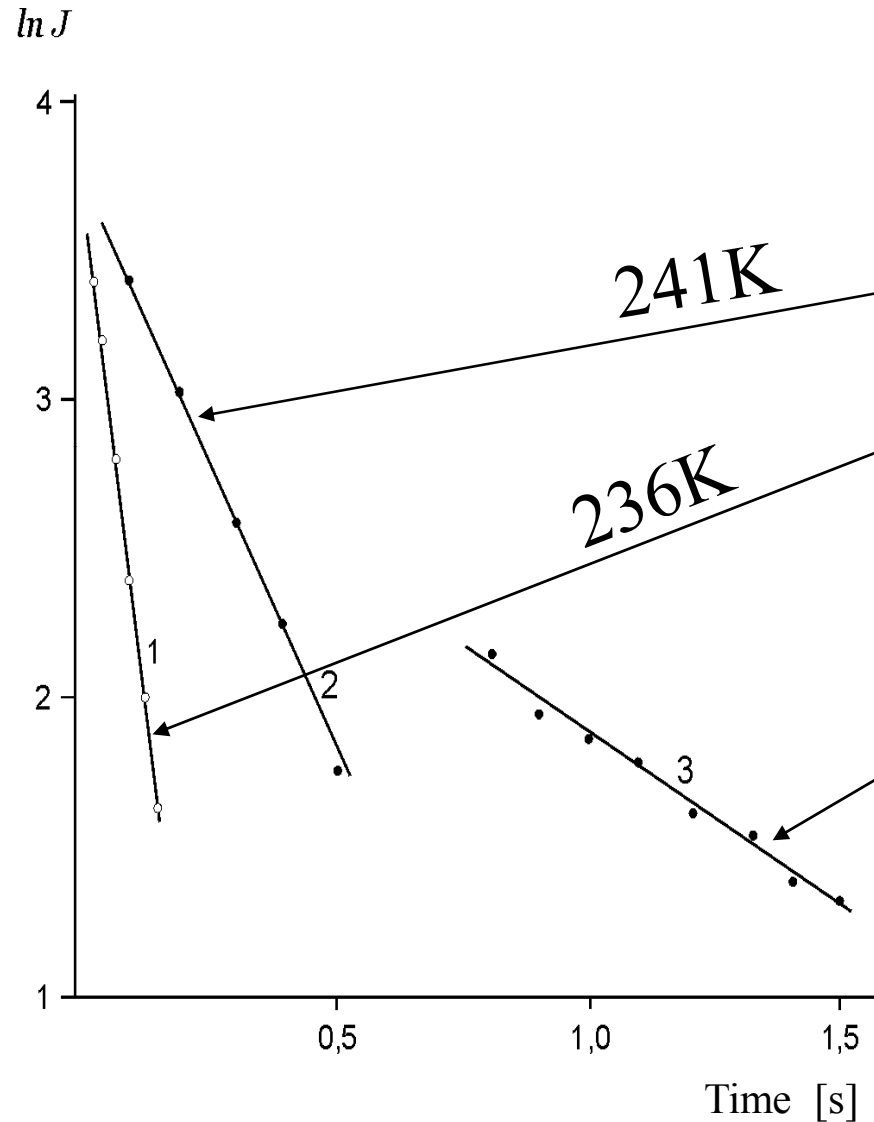
$$V = 216 \text{ cm}^3.$$



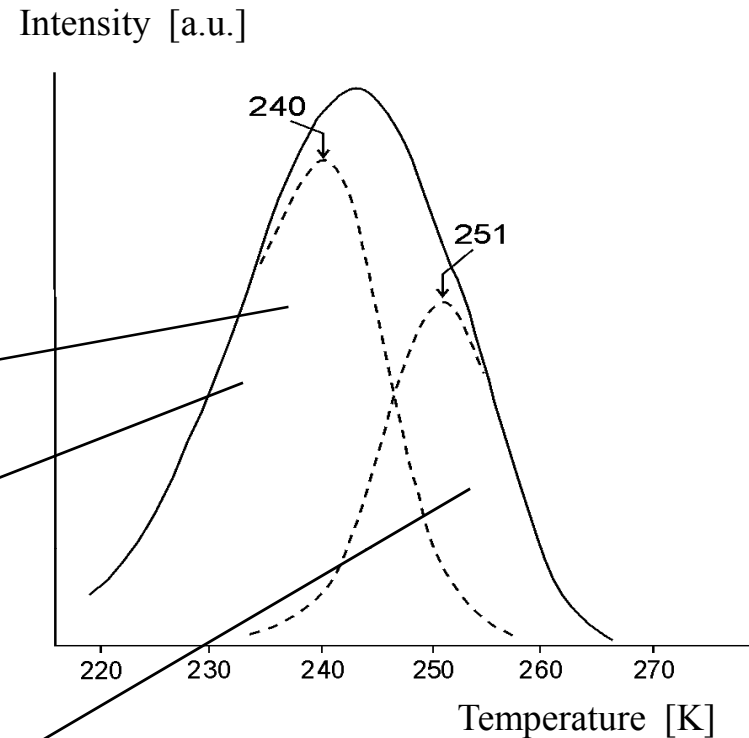
# Nature of millisecond afterglow



# Two components of ms-AG

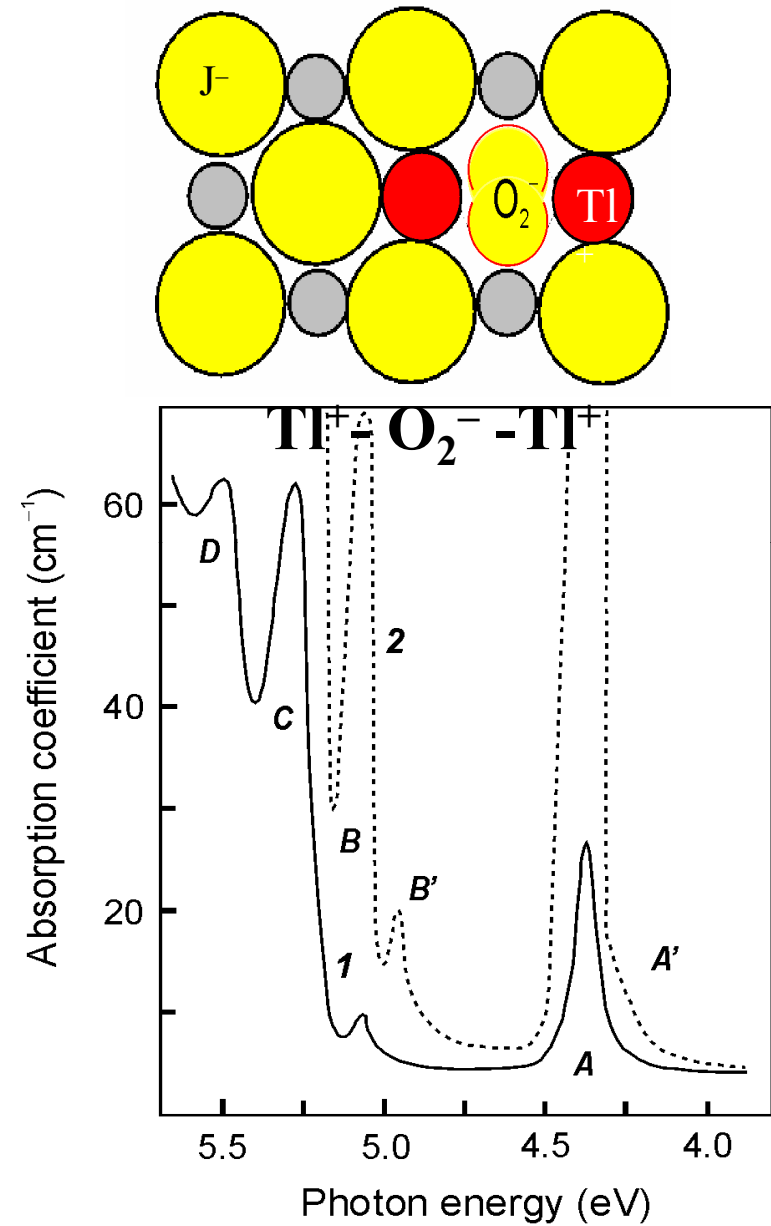
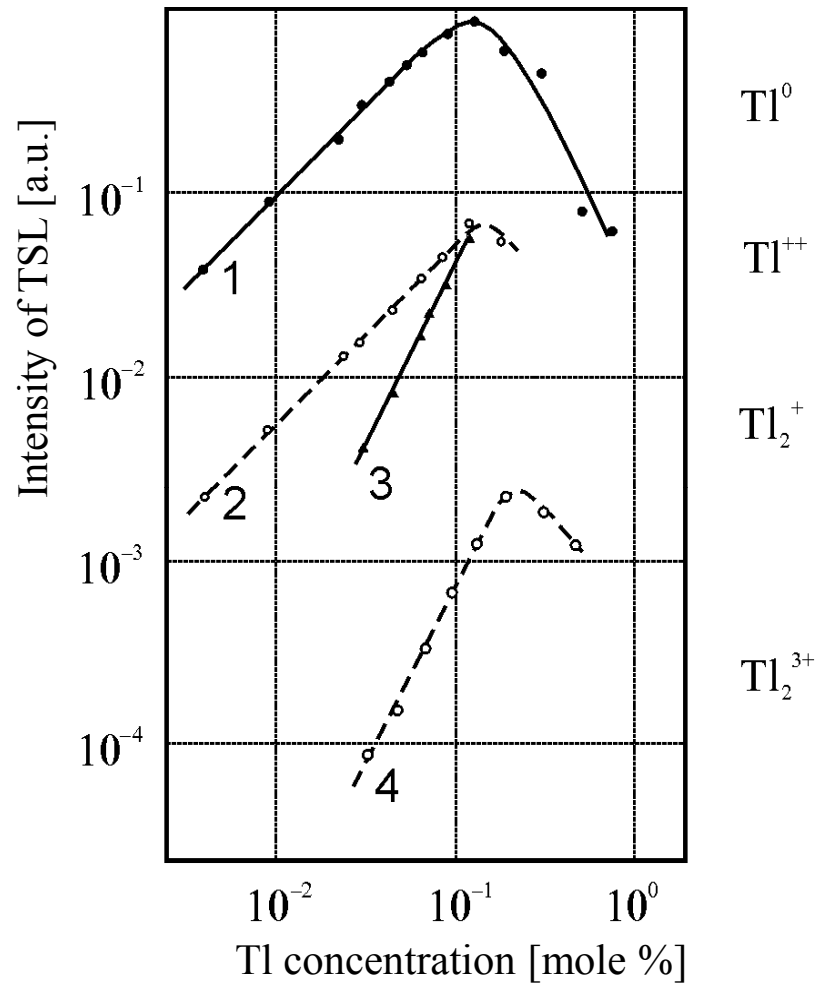


# Two peaks of TSL



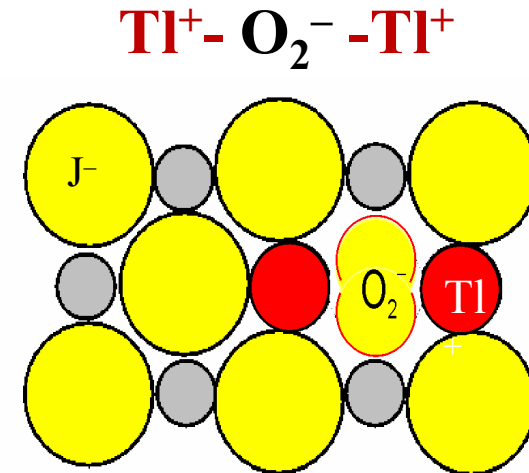
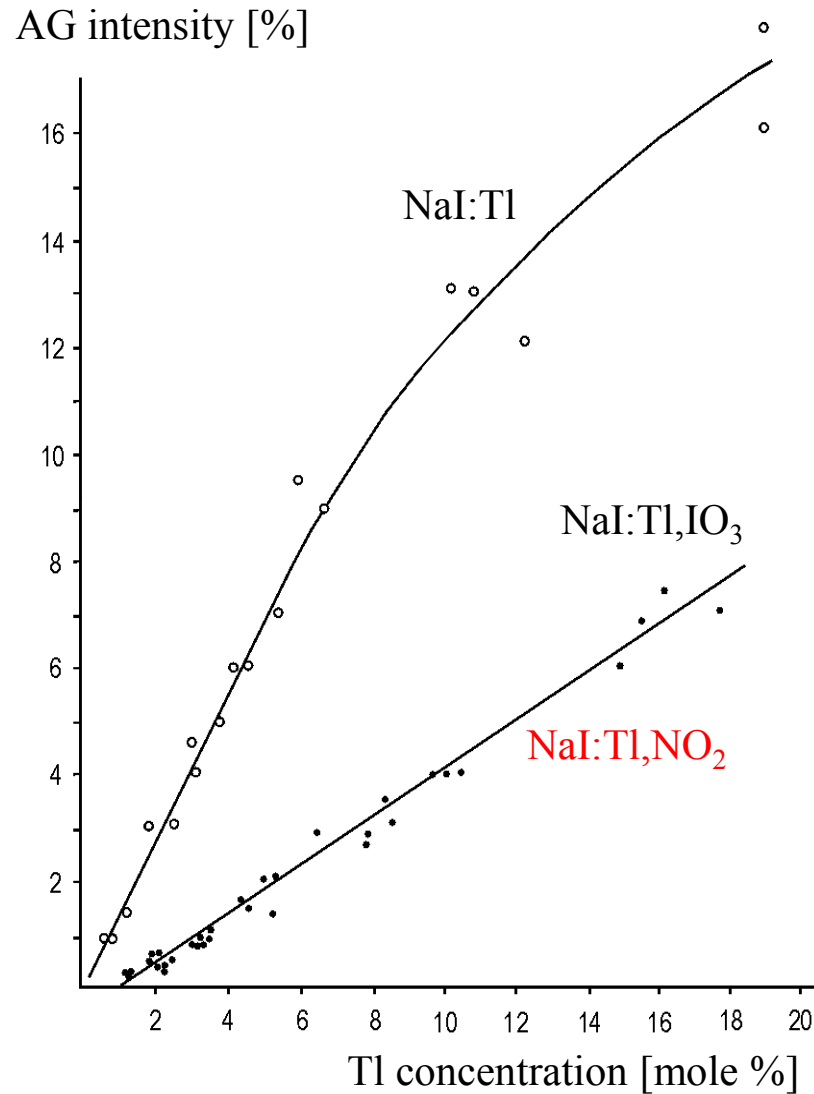
Main reason of millisecond AG in NaI:Tl is an existence of stable  $(Tl^+)_2$  centers (activator dimers)

# Nature of millisecond afterglow



Oxygen suppress AG and LY

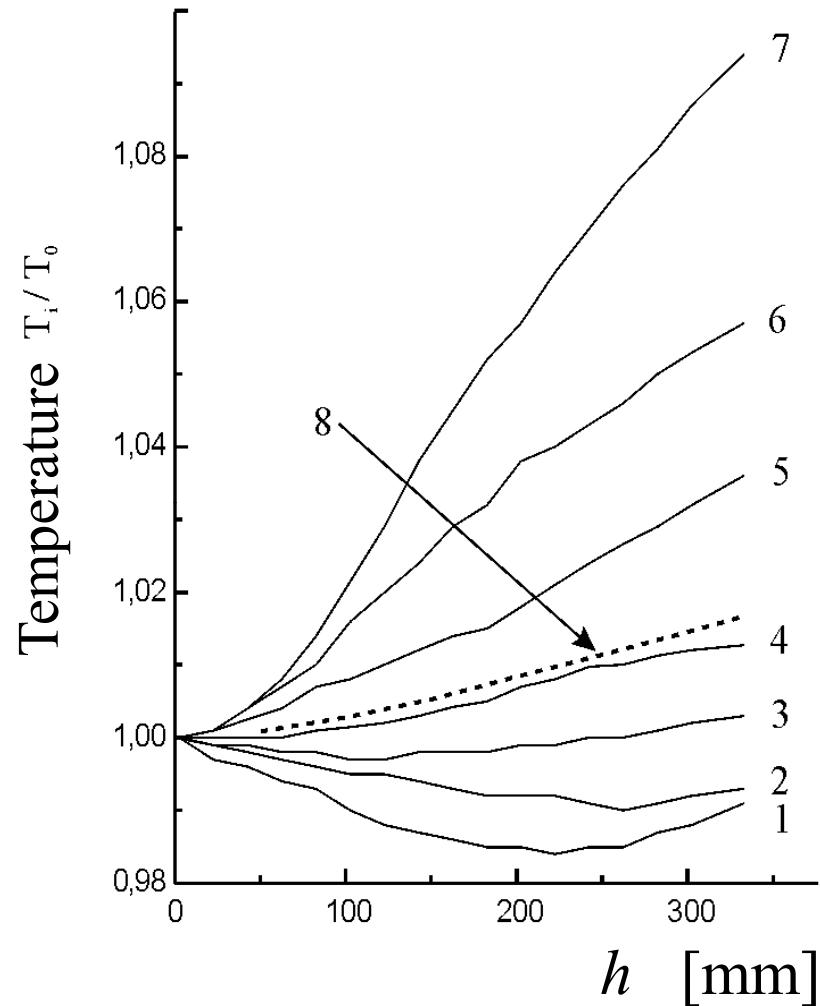
# Nature of millisecond afterglow and its suppressing



Model of electron trap  
which forms a  
quenching center for  
recombination  
luminescence

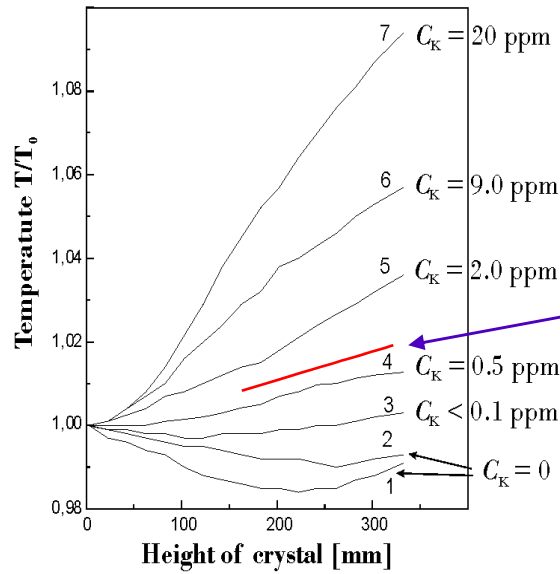


# Crystal growth of uniform and heavy-activated ingot

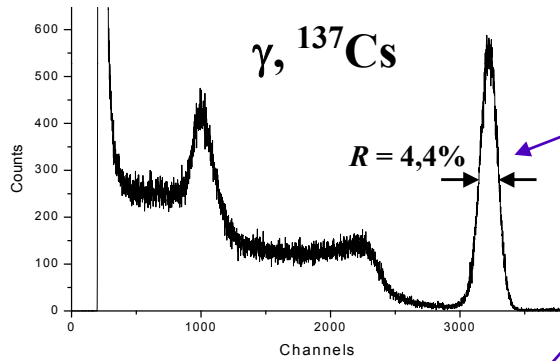


Scintillation materials: CsI:Tl,CO<sub>3</sub> or NaI:Tl,CO<sub>3</sub>

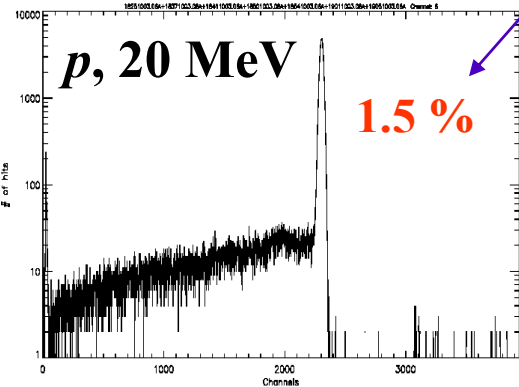
# CsI:Tl, NO<sub>2</sub>



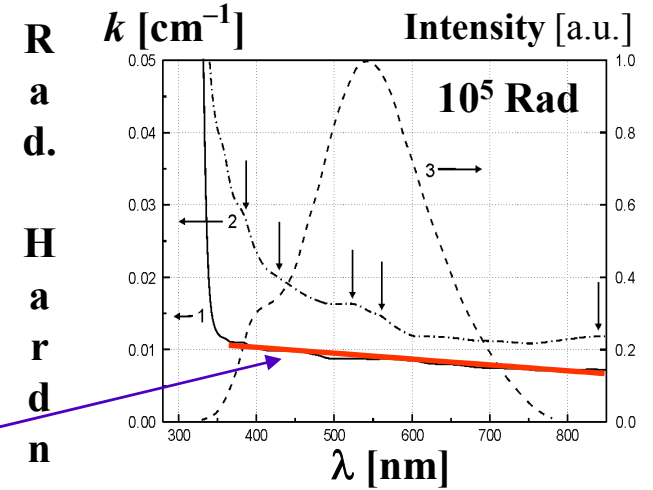
**Growth conditions**



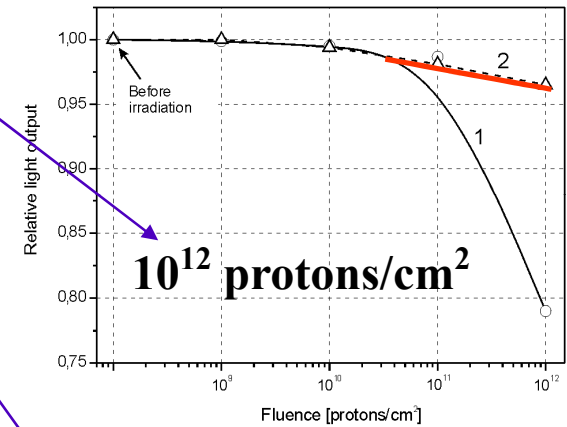
**Resolution**



**1.5 %**



**Rad. Hardness**



**Prospect**

**Afterglow**

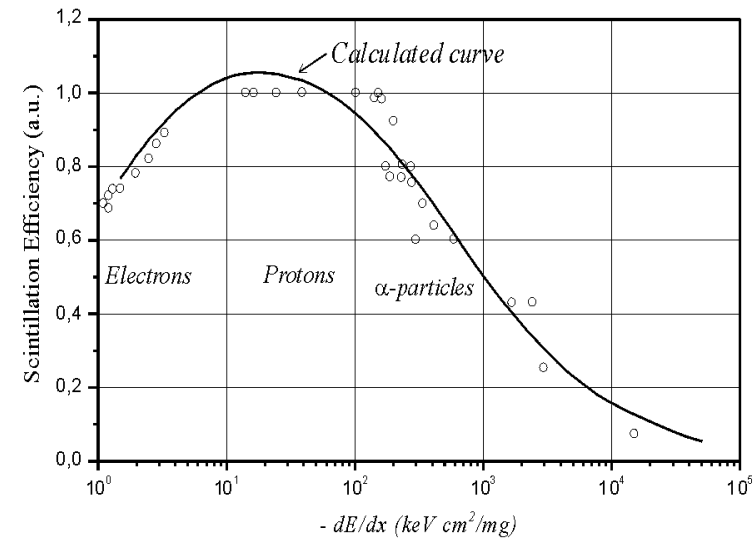
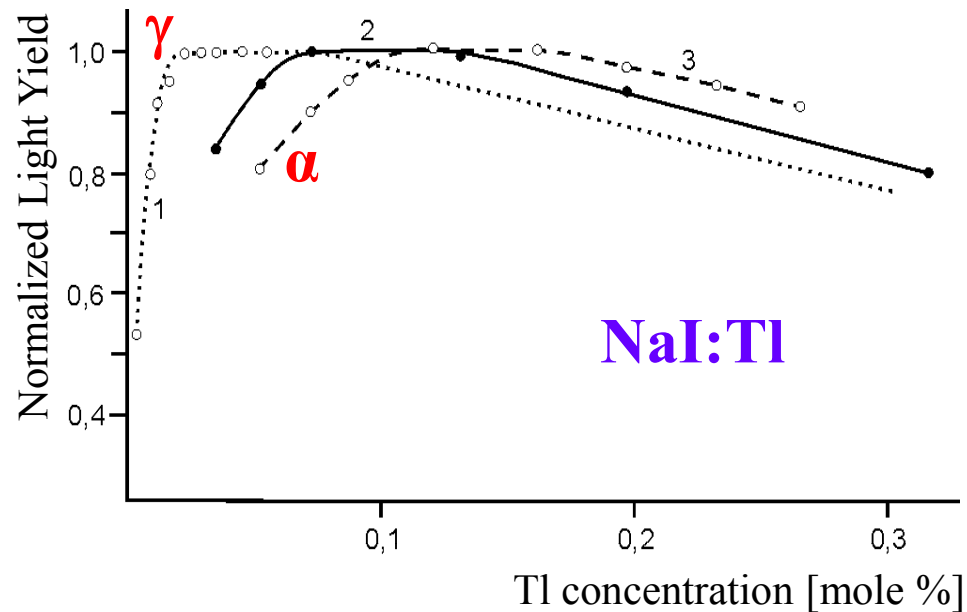
Material	AG (100 ms)
CsI:Tl	0,87...1,11%
CsI:Tl,NO <sub>2</sub>	<b>0.42...0.57%</b>

Limit of Tl concentration  
**0,5 %**

# Conclusion

- for Dark Matter search the heavy doped NaI:Tl crystals are needed;
- characterization of crystal quality should be done using alpha-particles and fission fragments;
- it has been shown that NaI:Tl crystals with  $C_{Tl} \sim 0.3 \%$  are available (so called NaI:Tl,IO<sub>3</sub> crystals). CsI:Tl,IO<sub>3</sub> crystals with  $C_{Tl} \sim 0.5 \%$  can be grown by Stockbarger technique;
- uniformity of NaI:Tl,IO<sub>3</sub> and CsI:Tl,IO<sub>3</sub> ingots is bad due to used crystallization technique;
- to obtain large uniform NaI:Tl crystal the modified Kyropoulos technique should be used;
- to obtain large heavy doped NaI:Tl crystal we recommend NaI:Tl,NO<sub>2</sub> scintillation material for crystal growth.

# Concentration dependences of light yield



Density of  $e-h$  pair in electron track:

$$7.3/2.2 \approx 3 \quad \left\{ \begin{array}{l} 2.2 \cdot 10^{18} \text{ cm}^{-3} \text{ for } 662 \text{ keV } (1, \gamma) \\ 7.3 \cdot 10^{18} \text{ cm}^{-3} \text{ for } 5.9 \text{ keV } (2) \\ 1,3 \cdot 10^{19} \text{ cm}^{-3} \text{ for } \alpha (5,15 \text{ MeV}) \end{array} \right.$$

**Volume density  $dE/dx^3$  is increased  
3 times in L-deep**

## Conclusions

- Dep.  $L$  vs  $C_{Tl}$
- Decay time
- Resolution

Cascade only  
(Auger electrons  
and X-ray)  
do not increase  
 $dE/dx$

Cascade +  
Photoelectron of  
 $\sim 1$  keV energy  
increase  $dE/dx$

