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

Alexander M. Kudin

Institute for Scintillation Materials NAS of Ukraine
National University of Civil Protection of Ukraine

July 22, L'Aquila, Italy

What is our typical view on NaI(Tl) crystal?

Dimensions	from diam.10×10 mm to diam. 500×500 mm
Energy resolution	5.6% @ 662 keV (^{137}Cs)
Typical energy range	from 17 keV to more than 10 MeV
Light yield	about 40 000 ph/MeV (gamma)

What will change if we are looking for low energy events?

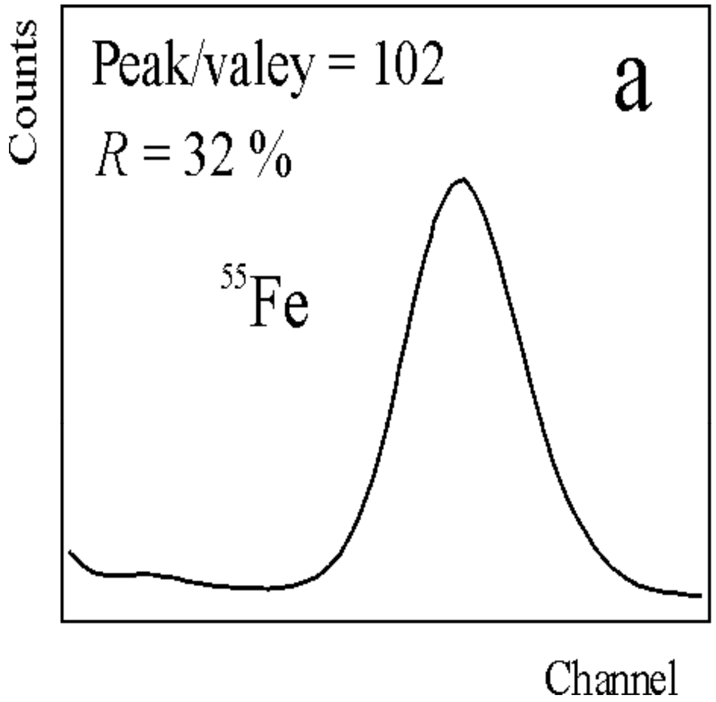
The best energy resolution – 32% @ 5.9 keV of ^{55}Fe

Improved non-proportionality at low energy

Different quenching factor for different particle type

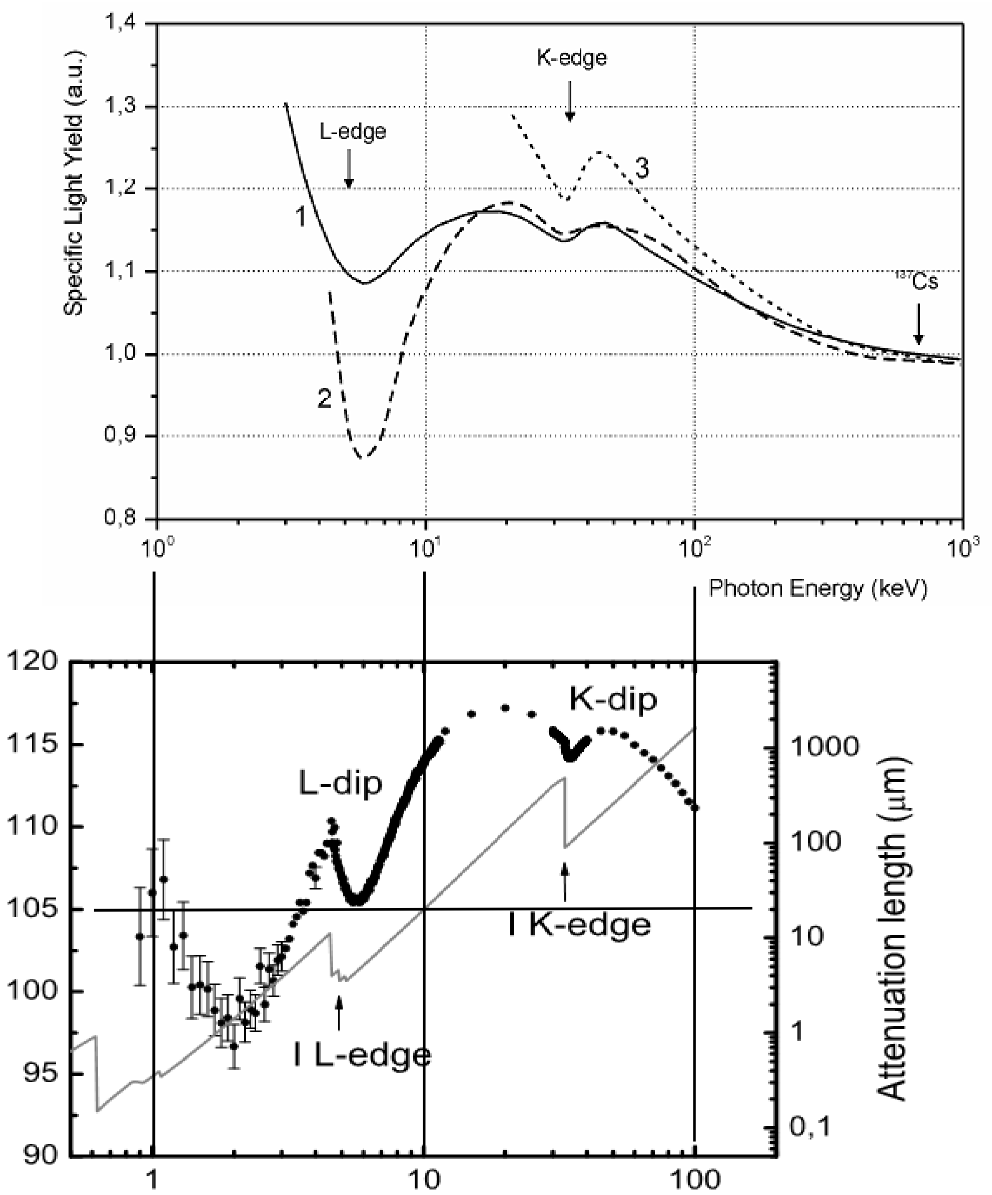
Detection of Soft X-rays by NaI:Tl Crystal

(*Persik D.E., Moi T.E.*, IEEE Trans. Nucl. Sci. – 1978. - Vol. NS-25, 615)



Pulse height spectrum of NaI:Tl crystal at excitation by ^{55}Fe isotope (a) with 5.9 keV energy. Peak at 1.5 keV on part *b* arises from interaction of X-rays in collimator.

Non-proportionality of Response



Aitken D.W., et al. IEEE TNS, NS-14, 1967.

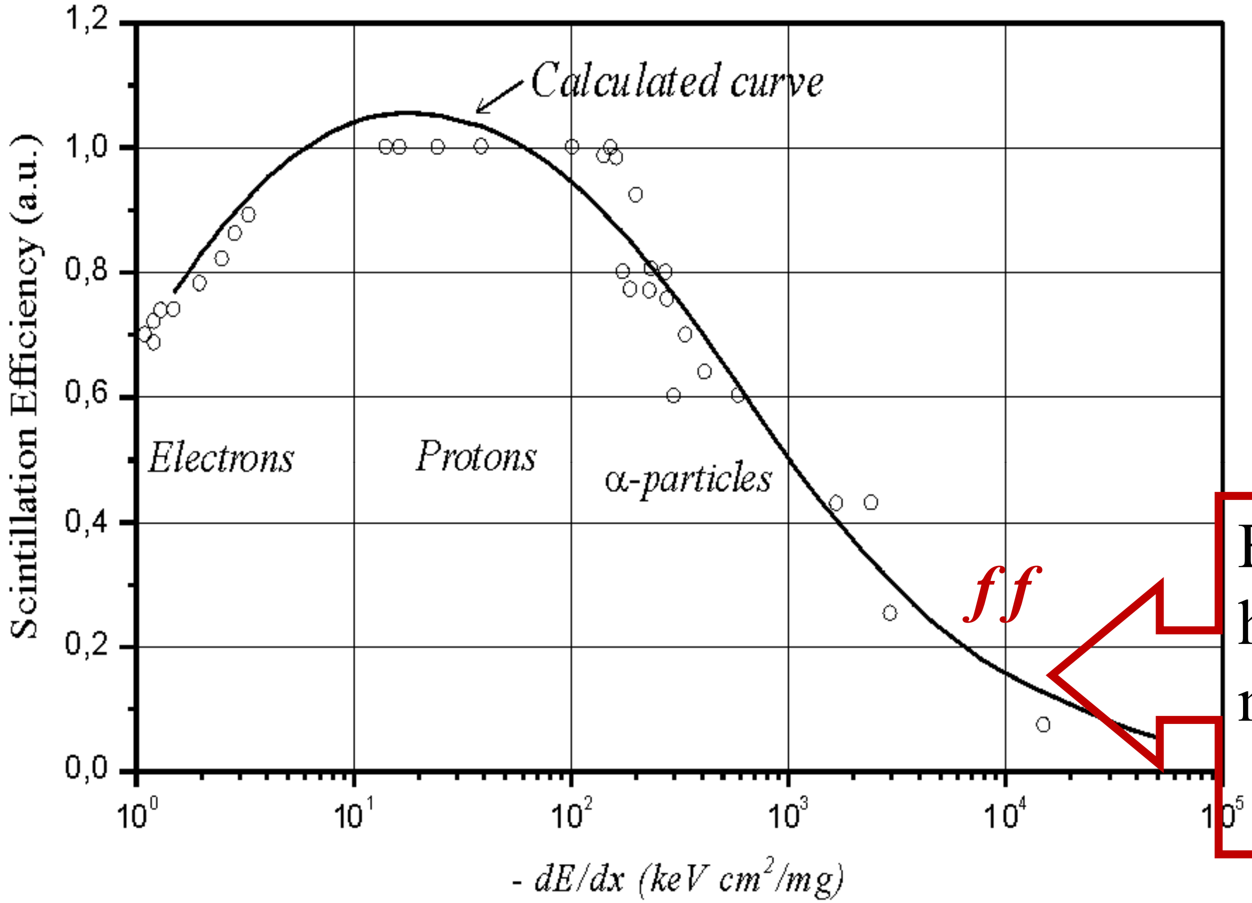
1 – NaI:Tl;

2 – CsI:Tl;

3 – CsI:Na.

I. Khodyuk, P. Dorenbos, IEEE TNS, 2012

Problems of low energy particle detection



Fusion fragments have only 20% of maximum light yield

How to solve all these problems in framework of DM particles search?

our experimental goals

expected events in the energy range of 3–10 keV
As low as possible energy threshold
As high as possible energy resolution
As high as possible light yield proportionality

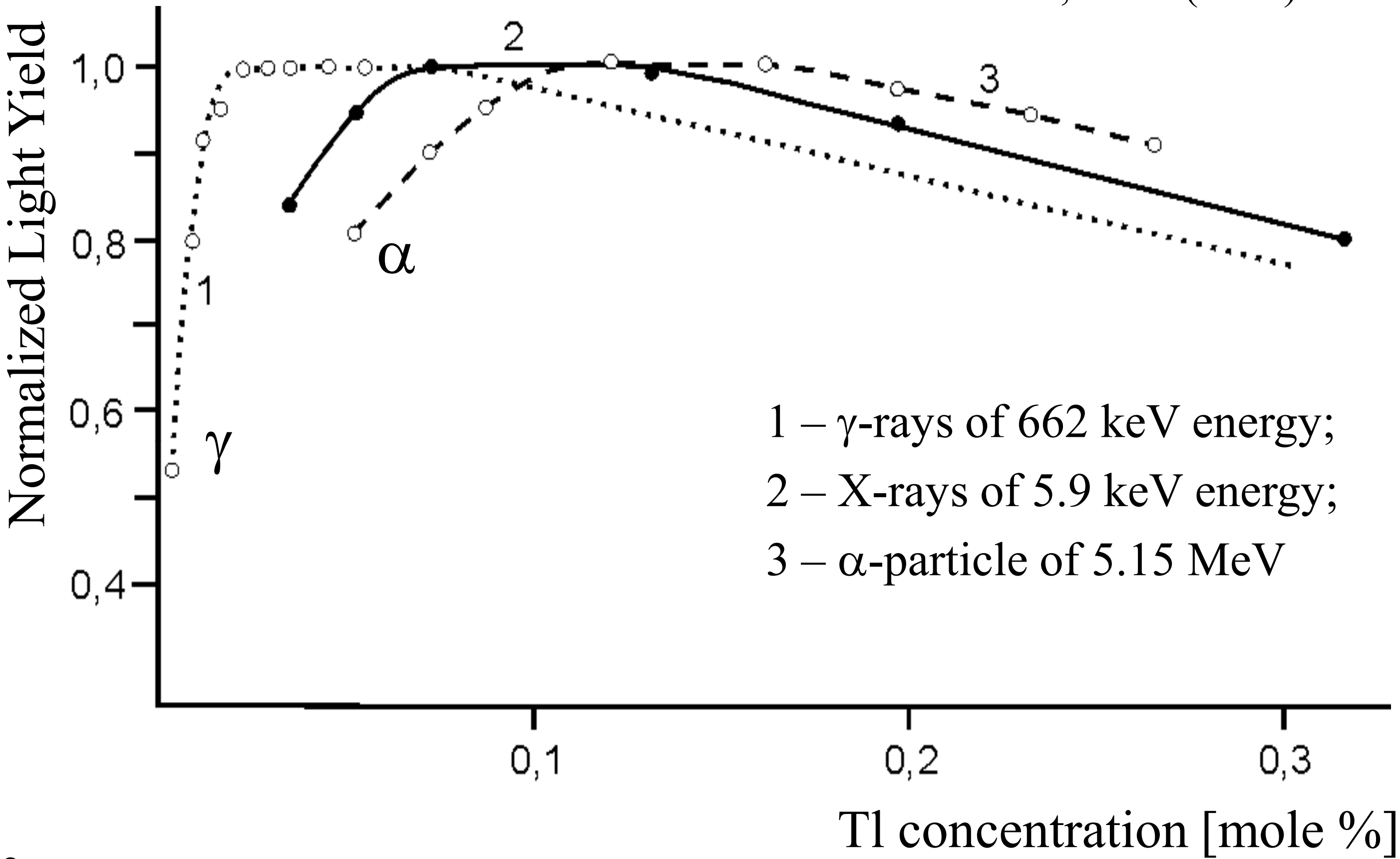
All these requirements could be satisfied
Only by crystals grown in dedicated conditions
Different from conditions for common crystal application

One of possible solution

Find optimum Tl concentration for detection of heavy charged particles

Optimum Tl concentration (C^{*}_{Tl}) in NaI:Tl

Trefilova L.N., Kudin A.M., et all.,
NIMA, A486 (2002) 474-481.



$C^{*}_{\mathrm{Tl}} = 2.2\cdot10^{-2}$ % for γ-rays;
$C^{*}_{\mathrm{Tl}} = 7.3\cdot10^{-2}$ % for soft X-rays of 5.9 keV;
$C^{*}_{\mathrm{Tl}} = 1.3\cdot10^{-1}$ % for α-particles.

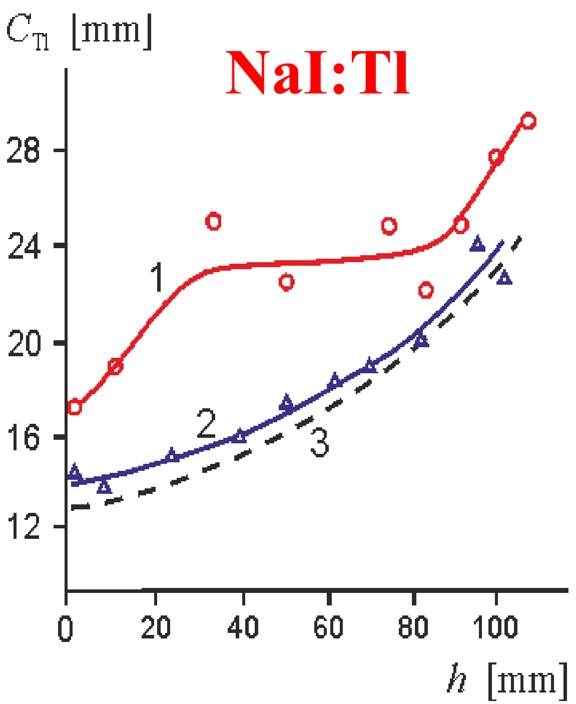
$C^{*}_{\mathrm{Tl}} > 0.15$ % for ion detection

Impurity distribution in crystal grown by Bridgman-Stockbarger technique

$$C_{Tl} = C_0 k_0 \left(1 - \frac{V}{V_0}\right)^{k_0 - 1}$$

k_o – equilibrium segregation coefficient

C_0 – initial concentration



1 – crystal growth in vacuum;

2 - crystal growth in oxygen;

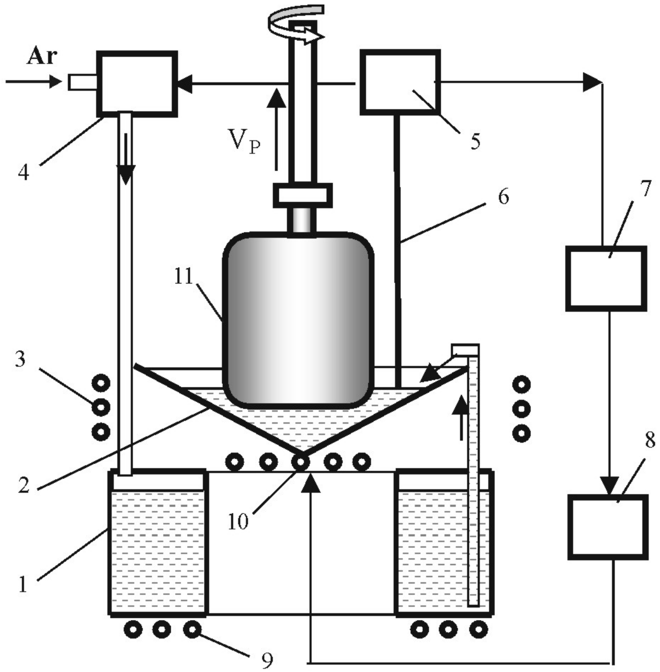
3 – calculated curve for $k_o = 0.25$, $C_o = 0.36$ %

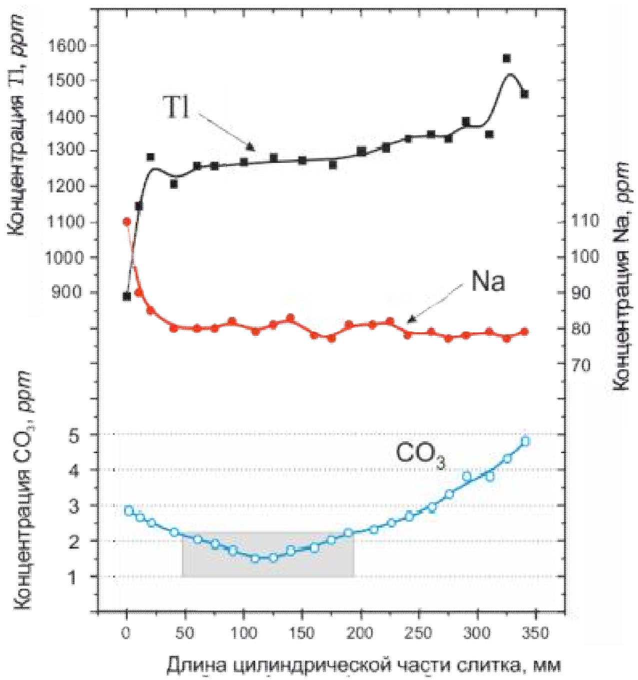
In heavily doped crystal activator is distributed non-uniformly

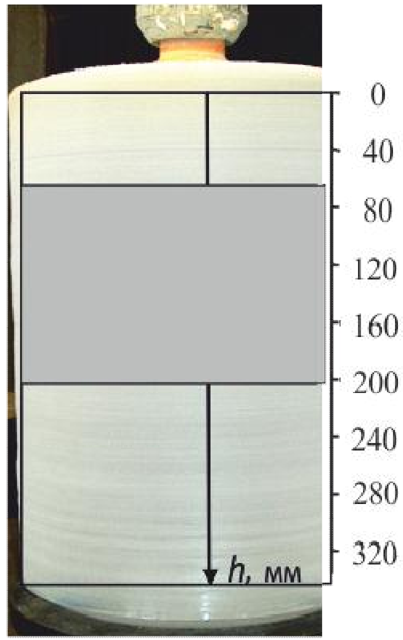
It can not be described by Pfann rule

NaI:Tl crystal for particle detection: homogeneity

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







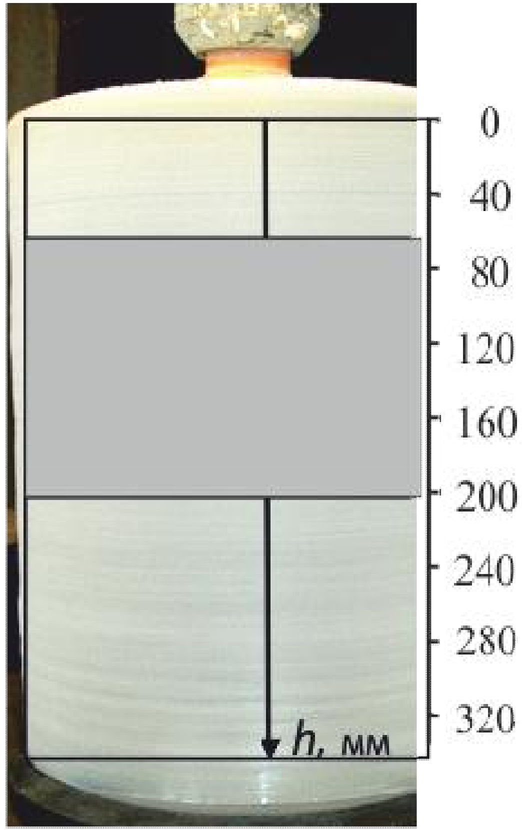
Distribution of Tl in scintillation material CsI:Tl along height

Uniformity of spectrometric parameters

CsI:Tl photodiode scintillators of 1 cm^3.

12 sample from selected region.

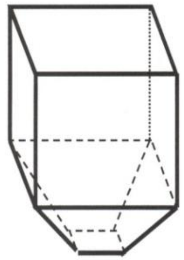
#	^{137}Cs @ 662 keV		^{241}Am @ 59.6 keV	
	L	***R, %***	***L***	***R, %***
1	3020	5.92	307	37.58
2	3036	**5.97**	302	**38.2**
3	3075	5.9	308	37.46
4	3047	5.87	309	37.34
5	3105	5.61	311	37.1
6	3000	5.8	302	38.2
7	3090	5.49	309	37.34
8	3079	5.5	308	37.46
9	3082	5.5	308	37.46
10	3070	5.83	309	37.34
11	3069	5.83	310	37.22
12	3078	5.81	311	37.1



Uniformity of spectrometric parameters

Photodiode scintillator of 200 cm^3.

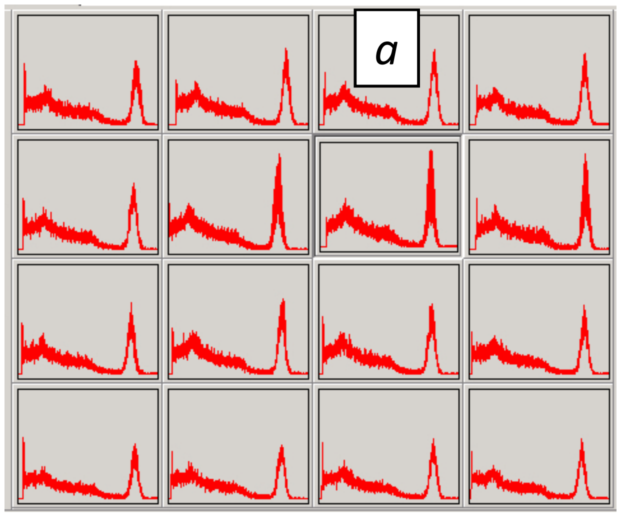
16 sample from selected region.

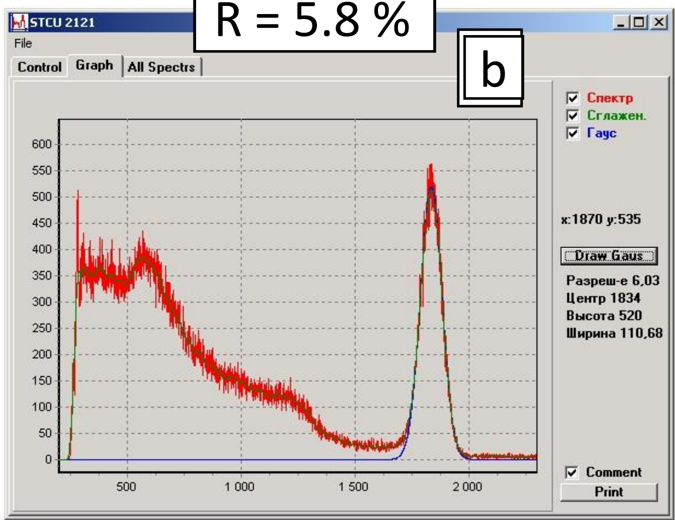


a

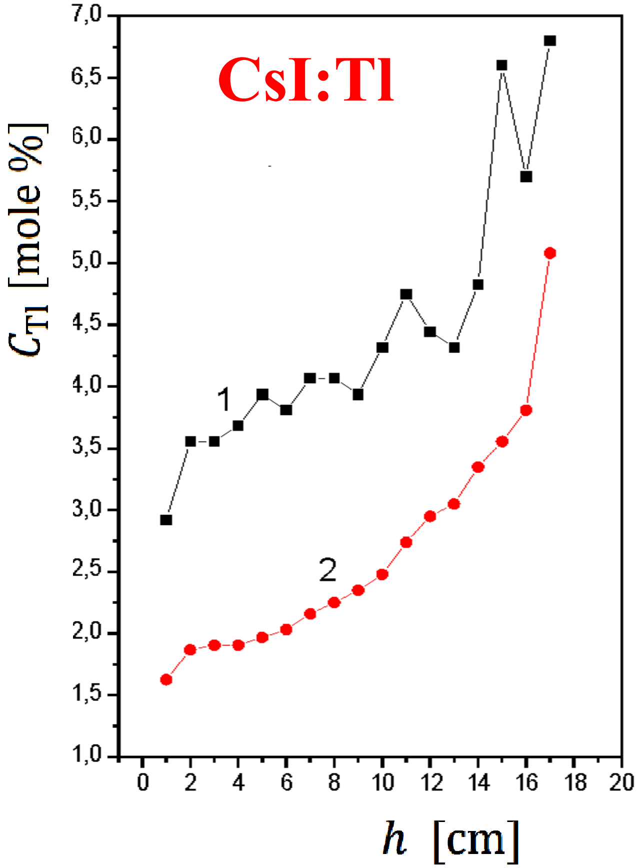
Pulse height spectra for each element (left) and summarized spectrum of whole block (right).

V = 216 см^3.





Non-homogeneous microscopic distribution of Tl in heavily activated CsI:Tl crystal



PIXE analysis results

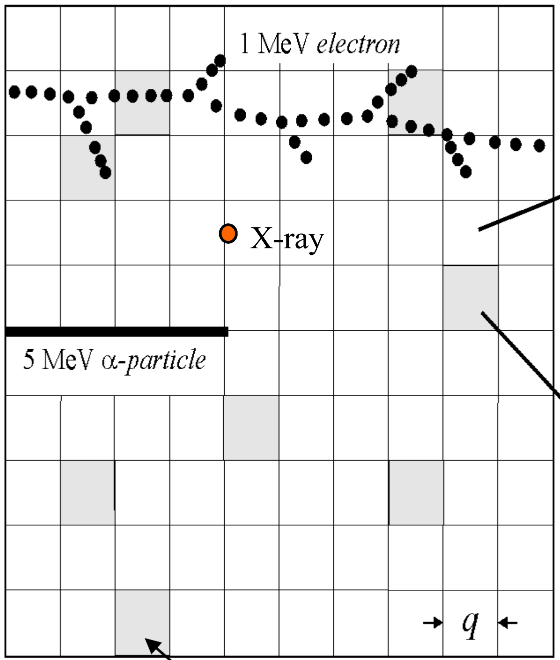
Brand	CsI n.	Nominal Tl conc.(ppm)	Measured Tl conc. (ppm)	Type of measure
GB	1	4000	6400±200	Face A, av
GB	1	4000	9300±300	Face A, point
GB	1	4000	5400±200	Face A, point
GB	1	4000	6100±200	Face B, av
GB	1	4000	4610±180	Side, av
GB	2	3000	2950±110	Face A, av
GB	2	3000	4900±200	Face B, point
GB	2	3000	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

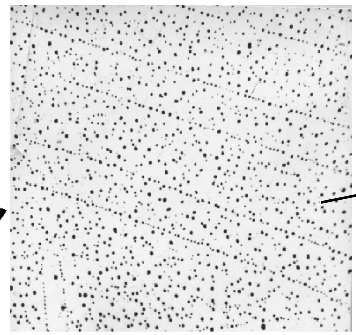
In C_{Tl} > 0.15% the activator is not homogeneous distributed both macroscopically and microscopically

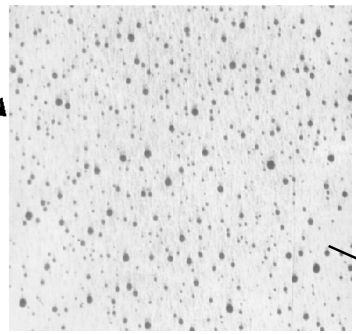
Nature of concentration quenching (CQ)

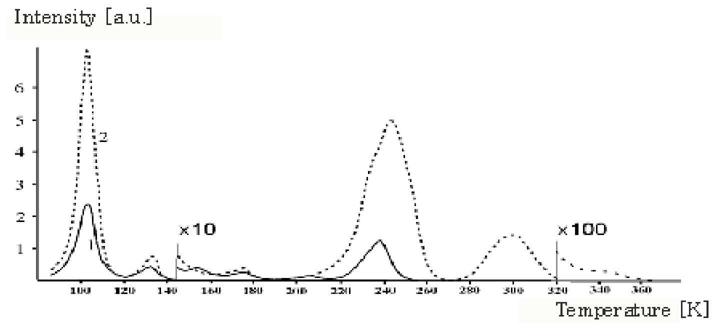
Heavily activated NaI:Tl crystal corresponds to region of CQ

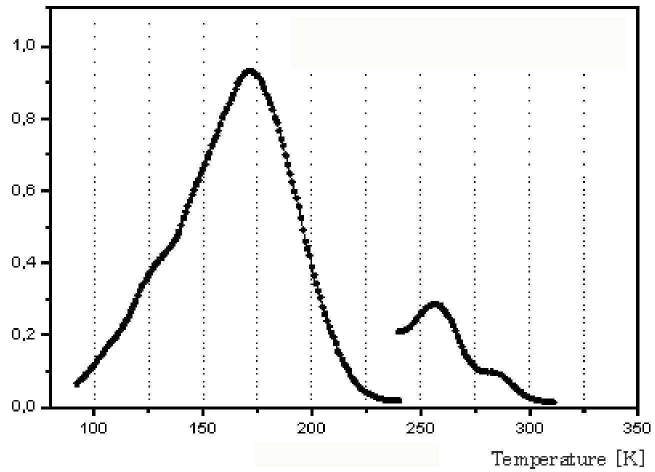


q ~ (5-7) mm
places of increased Tl content
(spinodal decay of solid solution)



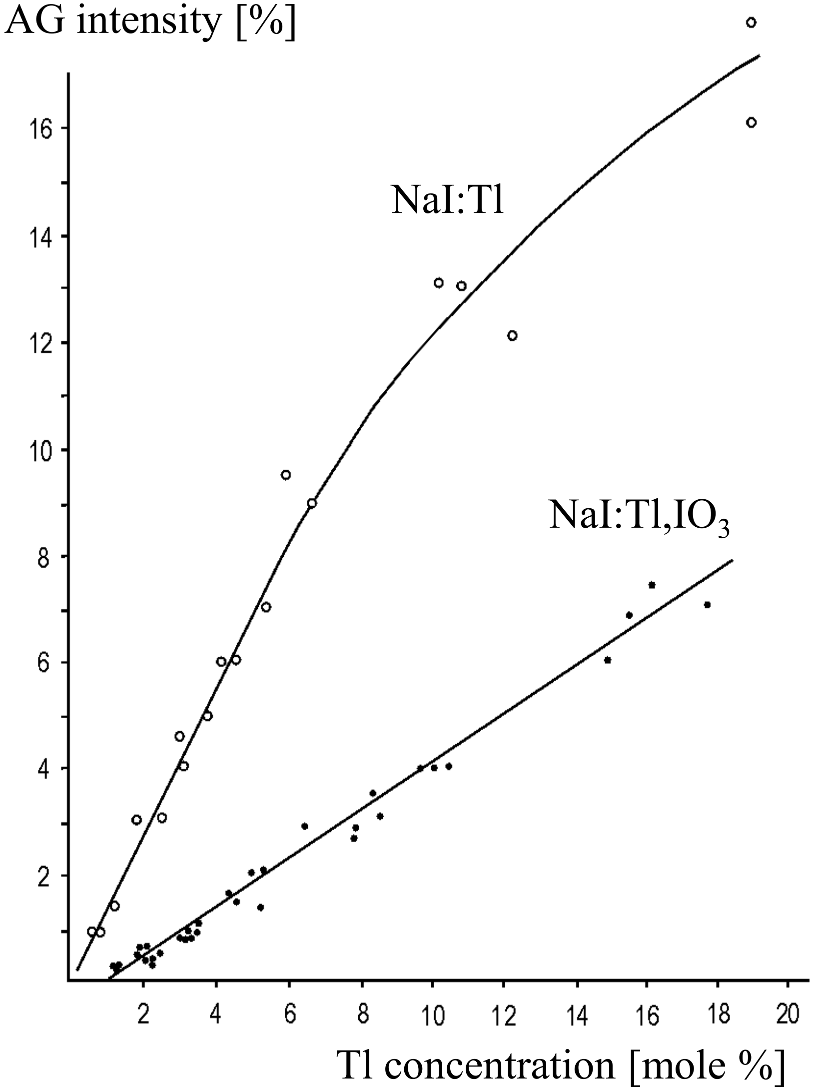




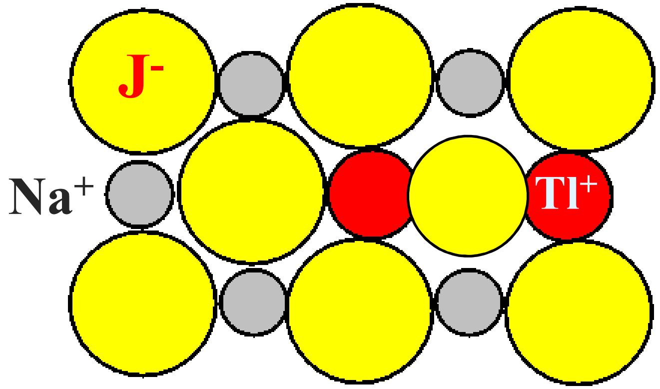


Schematic image of microscopic distribution of Tl^{+} center in CsI crystal at high Tl concentration.
Photo represents the character of decoration of the cleavage plane in two different places.

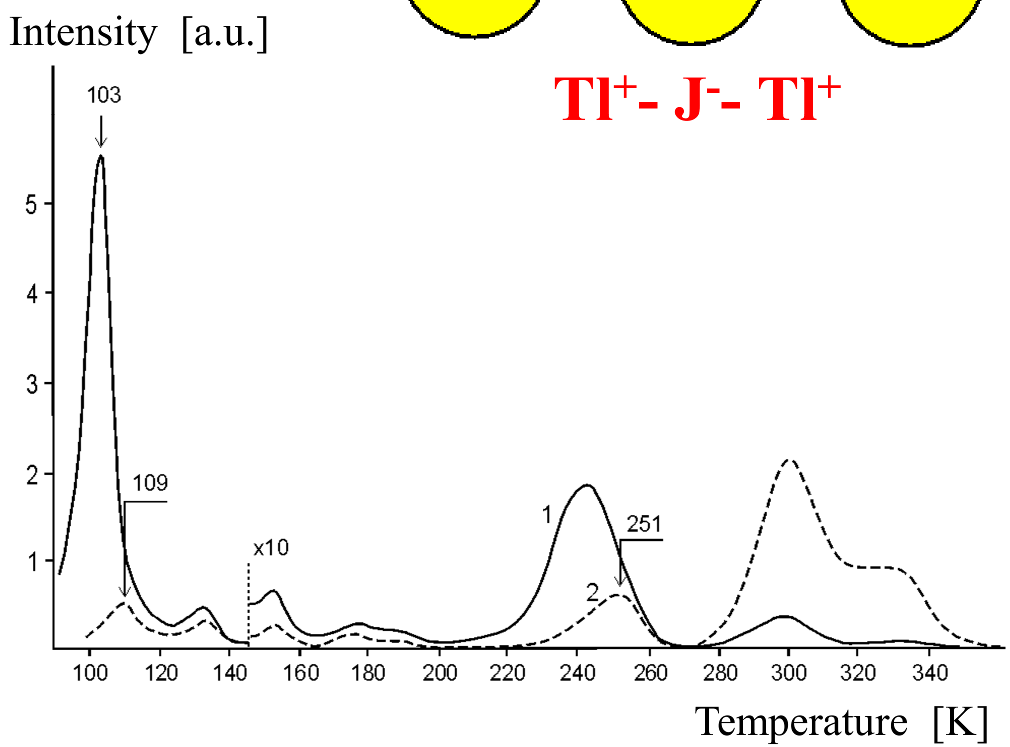
Nature of millisecond afterglow



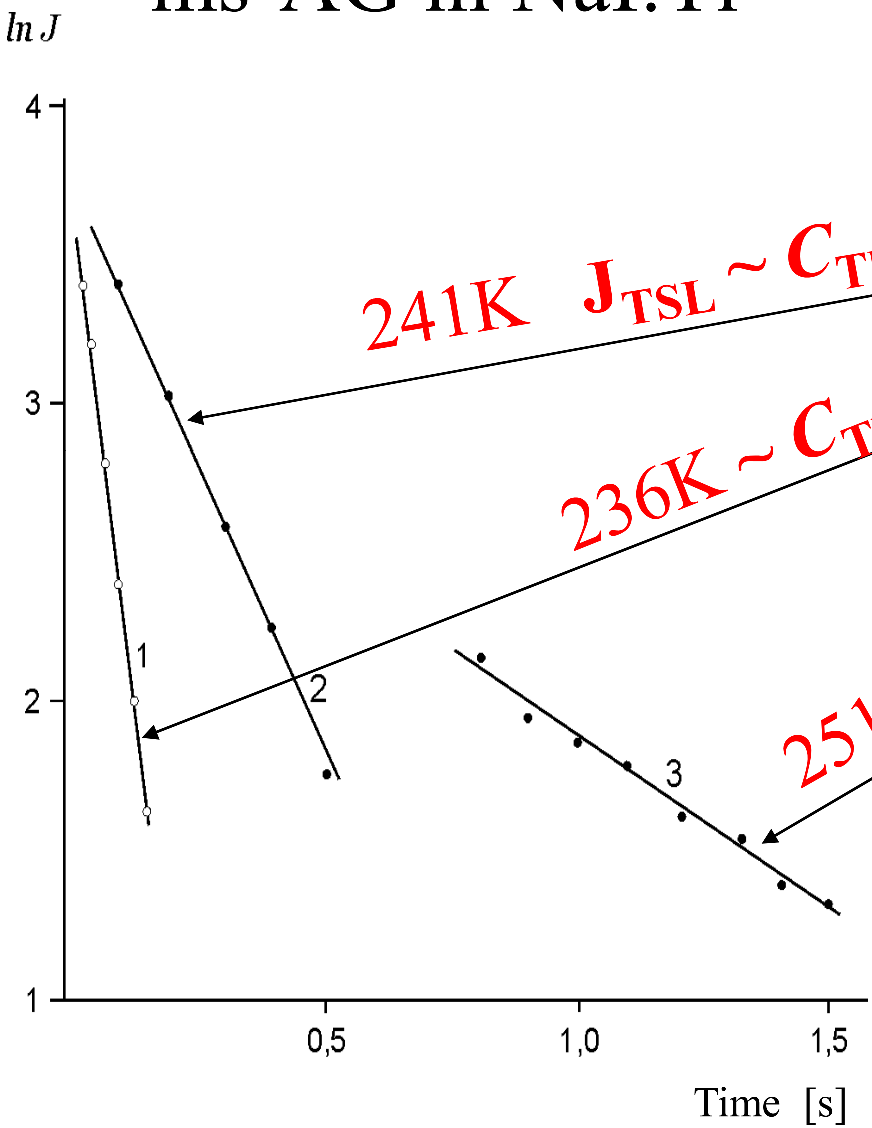
Model of trap



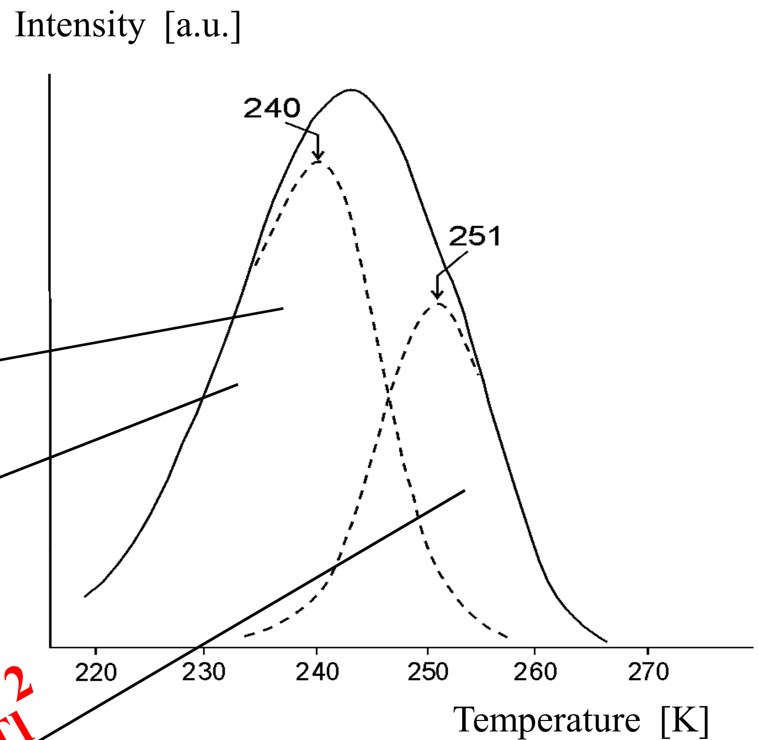
Tl^+- J^-- Tl^+



Two components of ms-AG in NaI:Tl

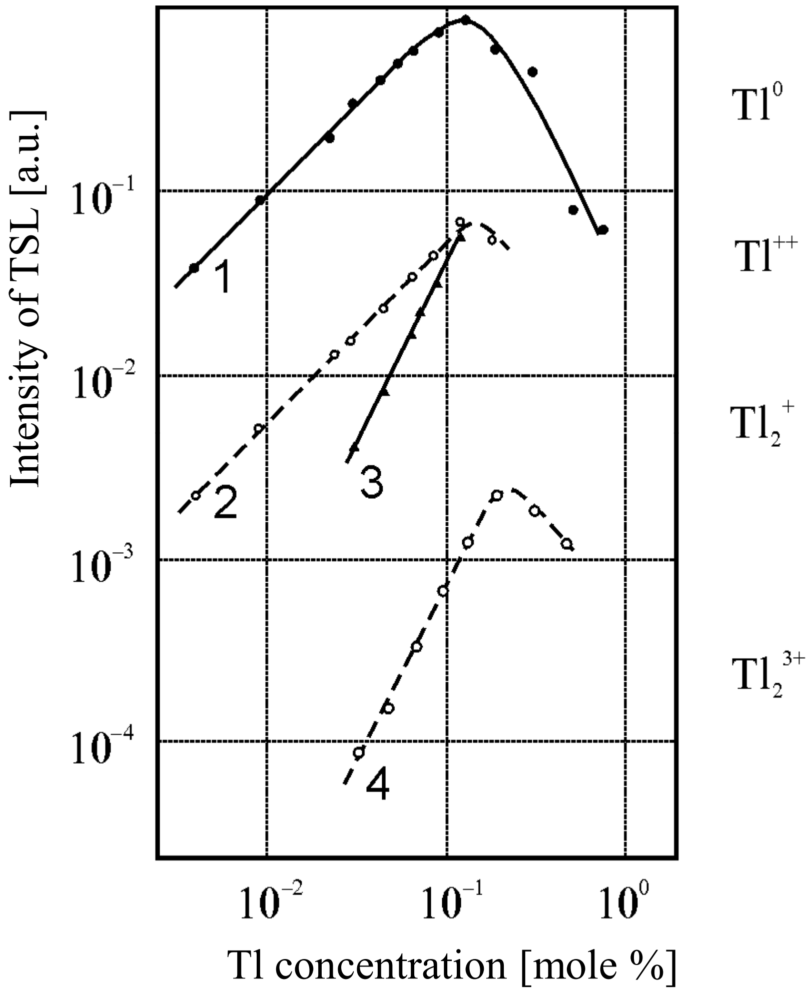


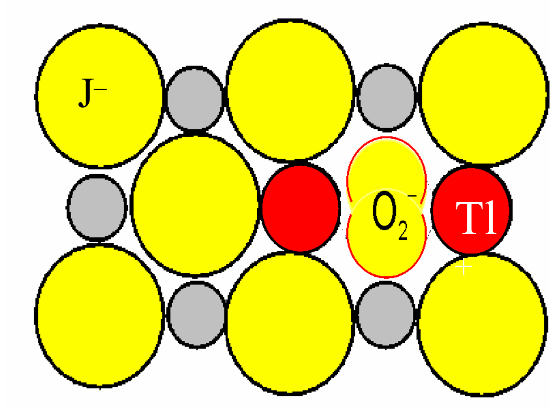
Two peaks of TSL



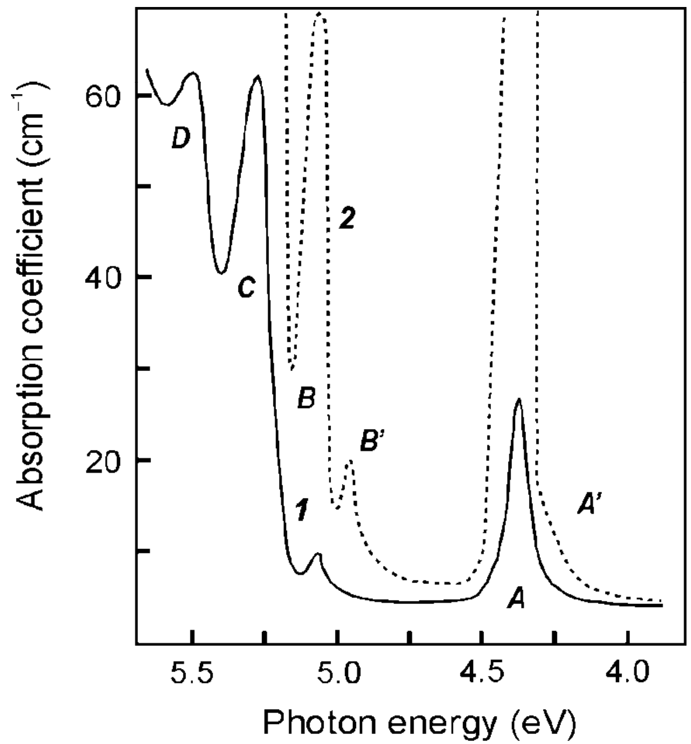
Main reason of ms-AG is an existence of stable $(Tl^{+})_2$ centers

Nature of millisecond afterglow and mechanism of its suppressing



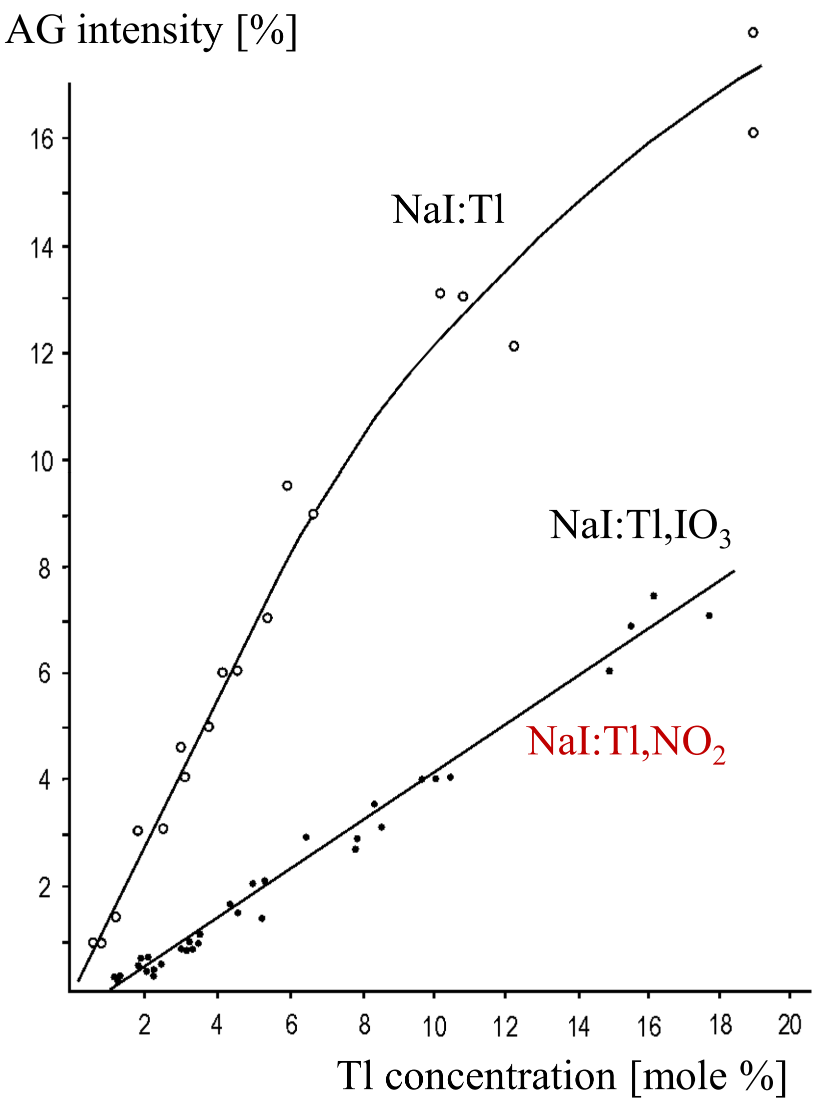


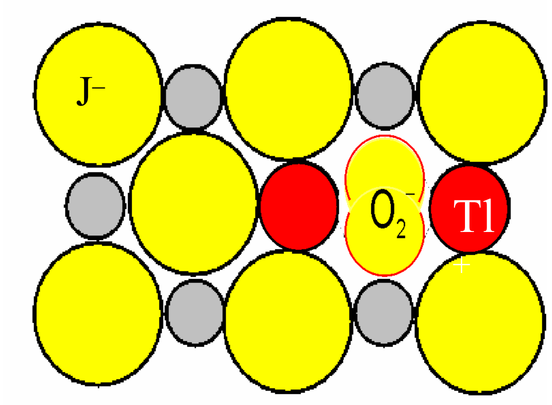
Tl^+- O_2^- -Tl^+



Oxygen suppress AG and LY

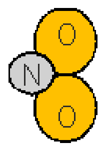
Reason of afterglow suppression by O_2^- and NO_2^-





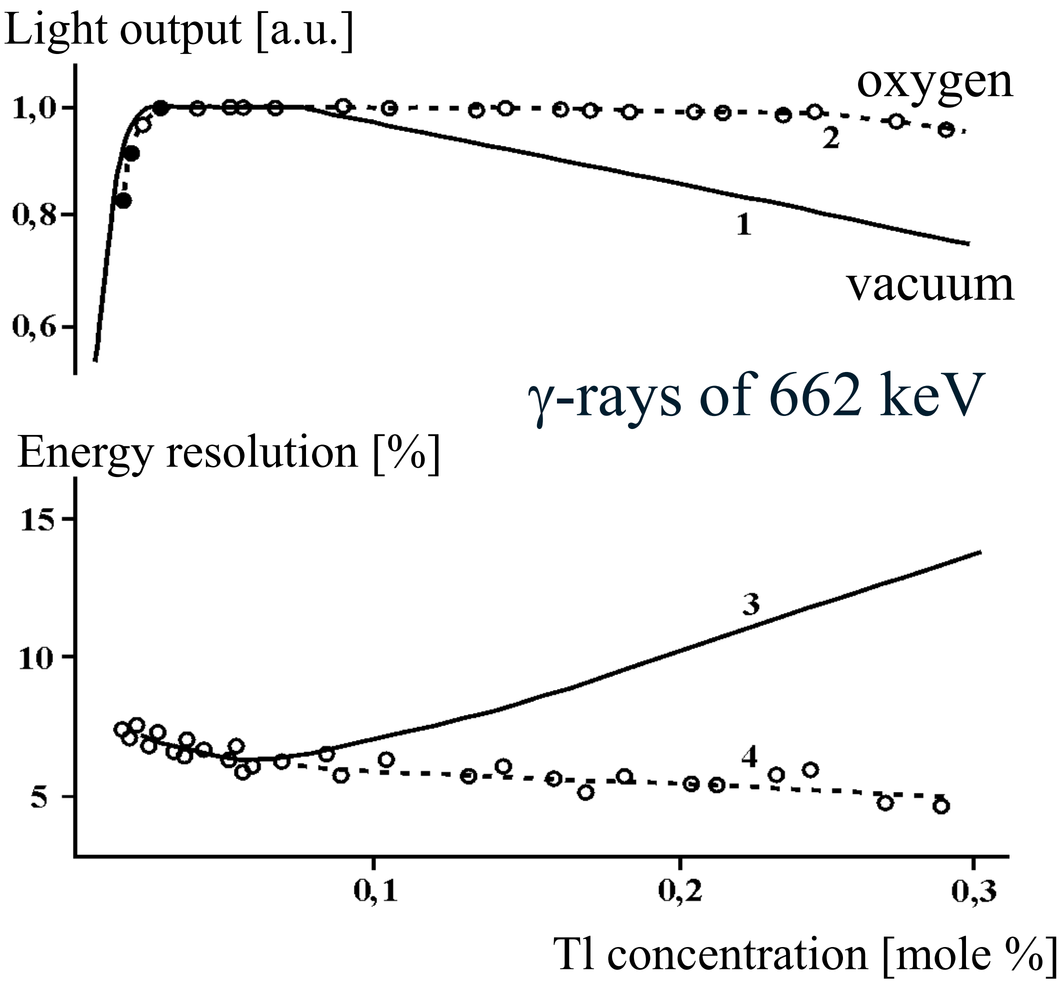
Tl^+- O_2^- -Tl^+

Model of non-luminescence center



The same for nitrate ion NO_2^-

NaI:Tl,IO_3 crystals without CQ

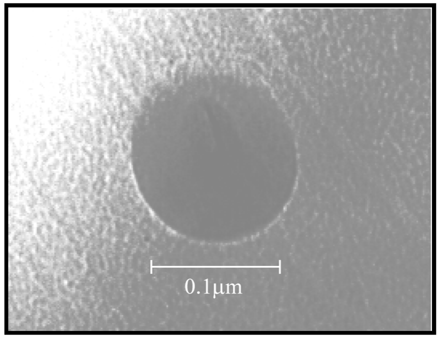


Crystal are grown by Bridgman-Stockbarger technique in oxygen atmosphere:

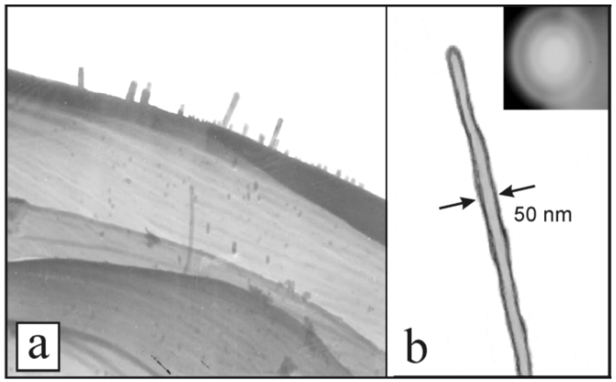
$NaIO_3 = NaI + 3/2\ O_2$

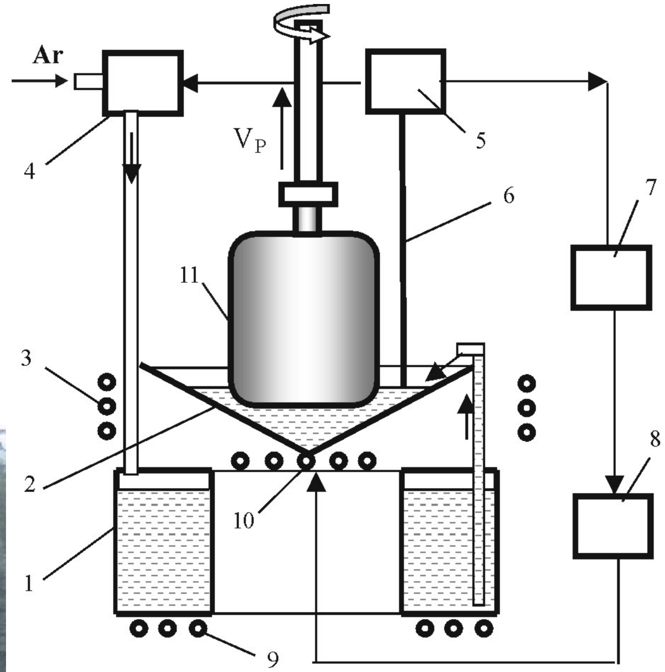
- good stirring of the melt;
- good transparency;
- low level of ms-AG;
- min-AG is absent;
- light scattering and gas bubbles are absent.

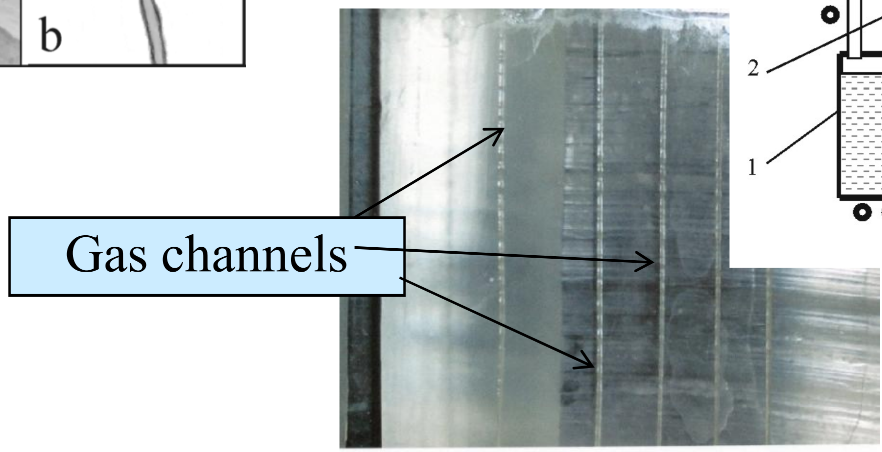
Disadvantages of NaI:Tl,IO_3 crystals



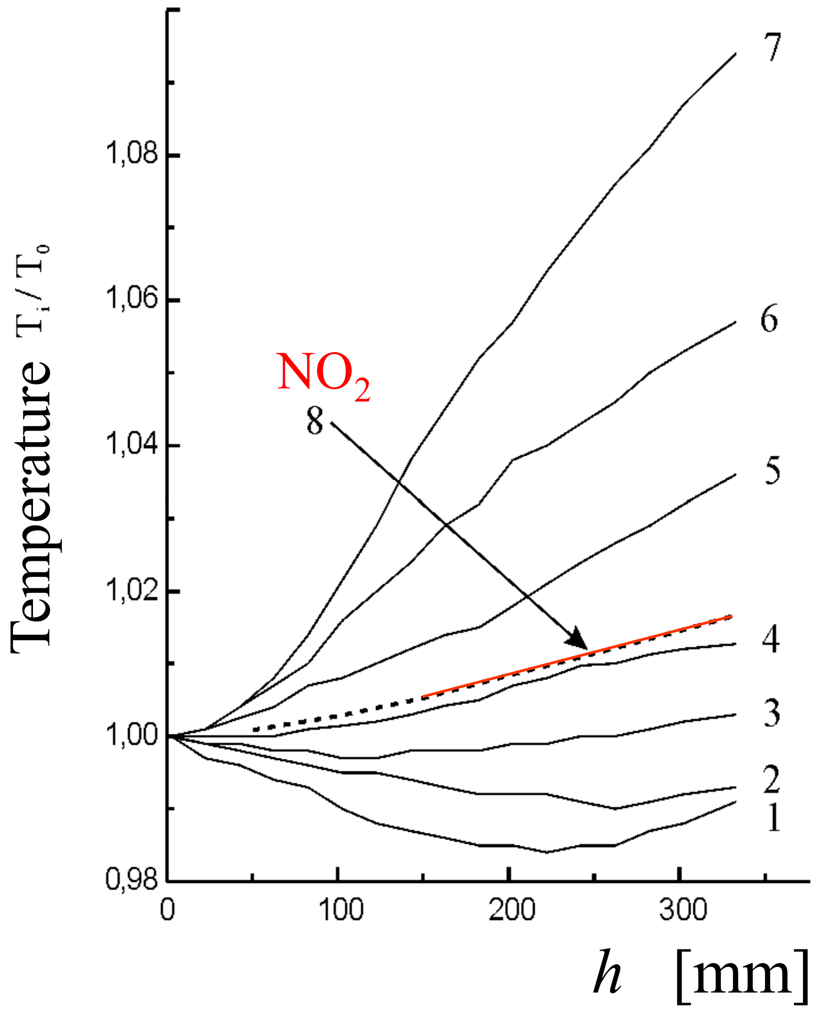
Crystals are grown by Kyropolos technique contain light scattering centers and gas bubbles.





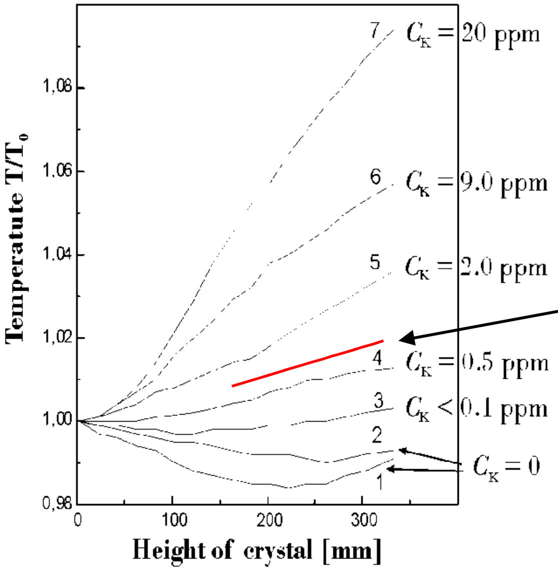


Crystal growth of uniform and heavy-activated ingot



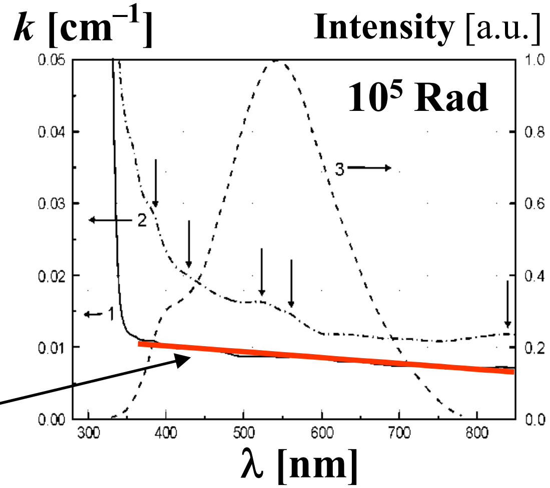
Scintillation material: NaI:Tl,CO_3 or CsI:Tl,CO_3

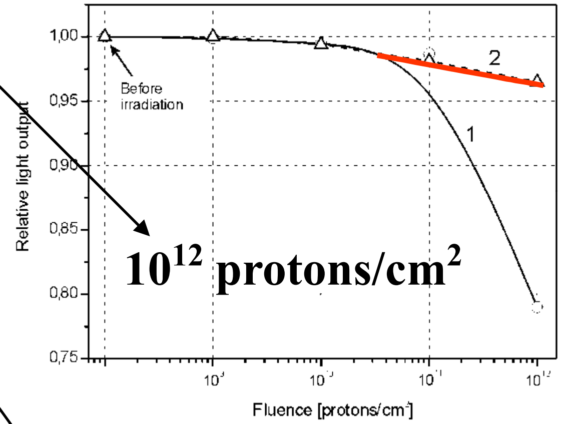
$CsI:Tl, NO_2$

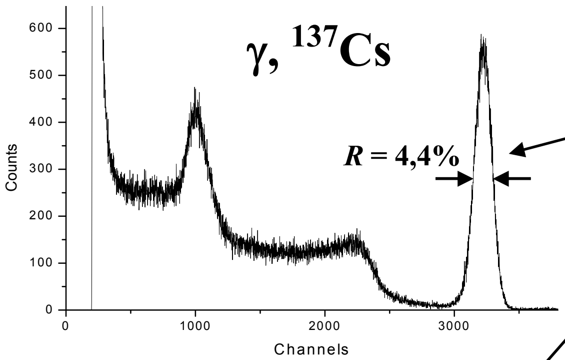


Growth conditions

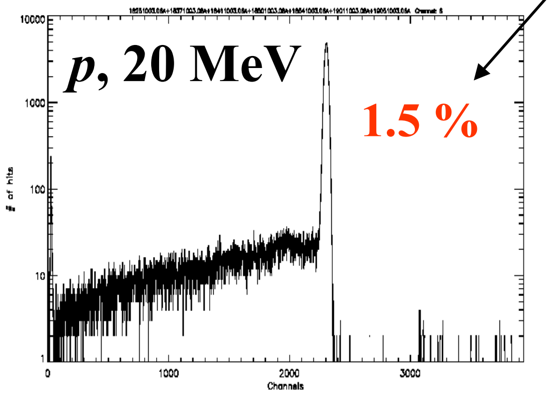
R a d. H a r d n e s s







Resolution



Afterglow

Material	AG (100 ms)
CsI:Tl	0,87…1,11%
$CsI:Tl,NO_2$	**0.42…0.57%**

Prospect

Limit of Tl concentration

0,5 %

Summary

The same results as for **CsI(Tl,NO_2)** can be achieved also for heavily doped **NaI(Tl,NO_2)** single crystals (**Tl concentration > 0.15 mol.%**)

1. Big volume crystals (diameter more than 100 mm)
2. High optical transmittance
3. High uniformity of dopant distribution
4. High energy resolution, both for 662 keV and 5.9 keV gammas
5. Optimal quenching factor for heavy charged particle
6. Without concentration quenching for gamma rays
7. Low millisecond/minute scale afterglow

Production of heavily doped **NaI(Tl,NO_2)** single crystals (**Tl concentration > 0.15 mol.%**) required dedicated growth technology and knowledge

Small volume heavily doped **NaI(Tl,NO_2)** single crystals are available for testing

Large volume heavily doped **NaI(Tl,NO_2)** single crystals should be grown

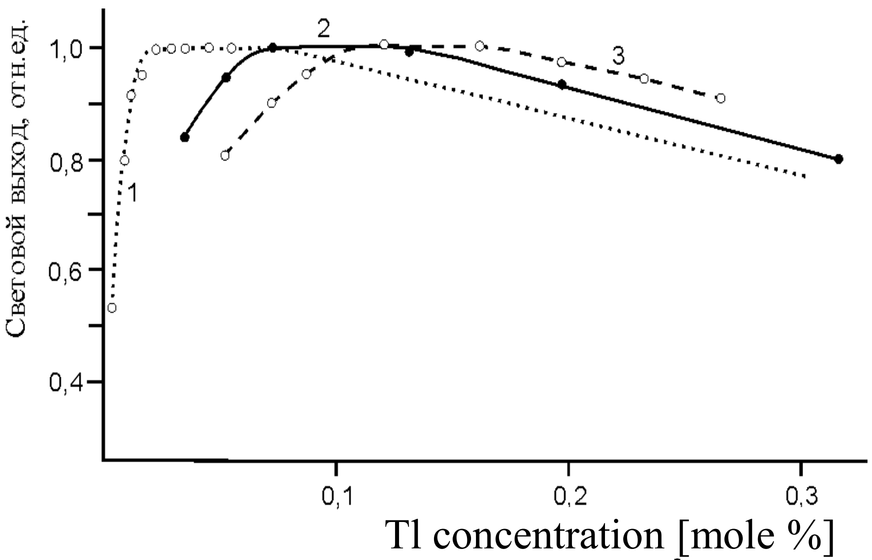

Thank you for attention!

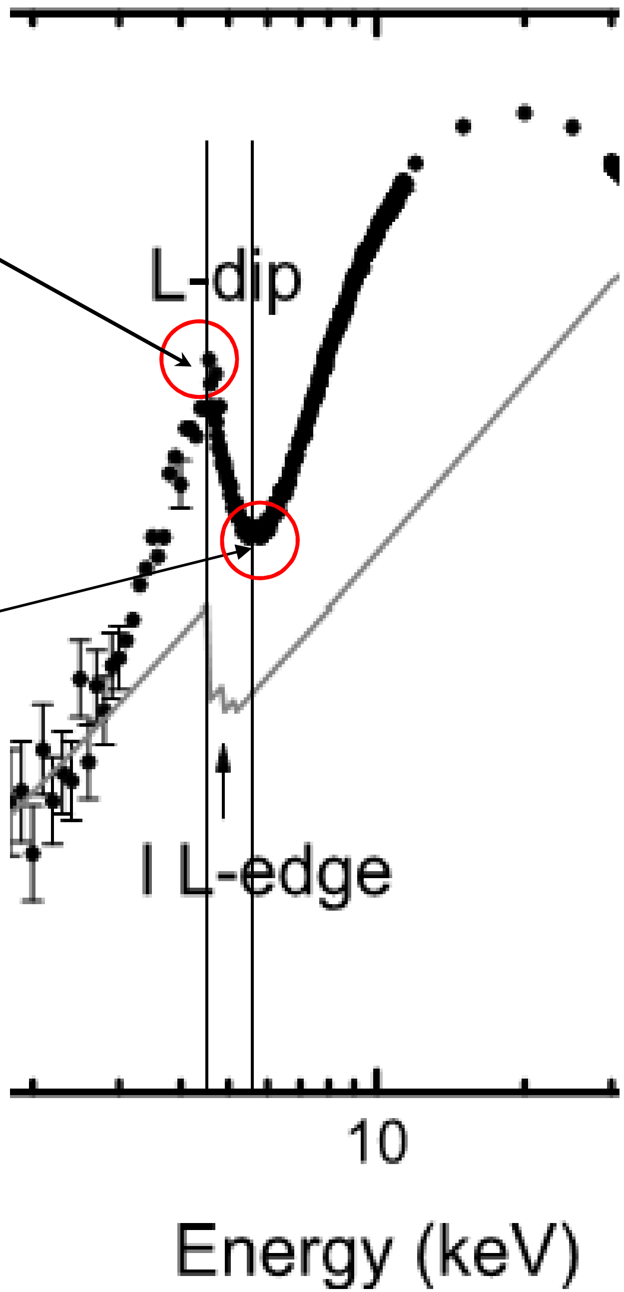
Conclusions

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

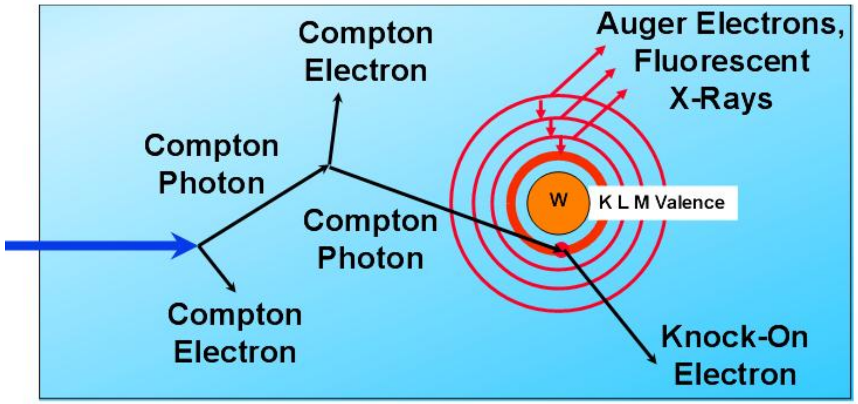
Cascade only (Auger electrons and X-ray) do not increase *dE/dx*

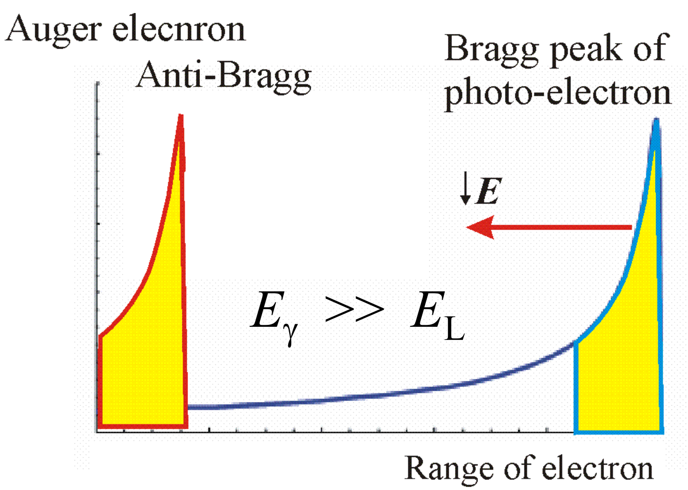
Cascade + Photoelectron of ~ 1 keV energy increase *dE/dx*

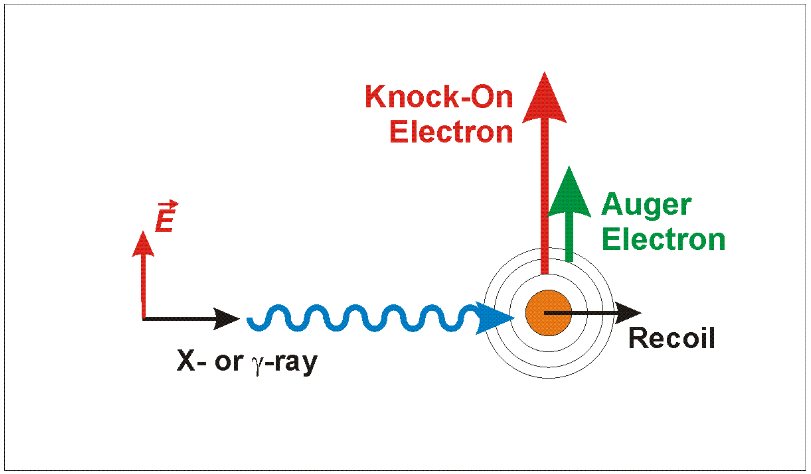


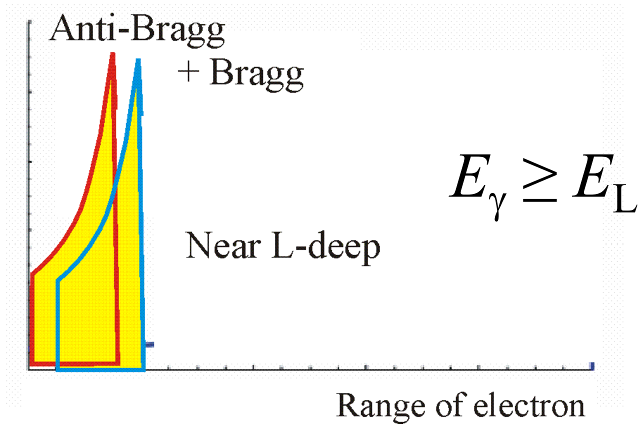


Anti-Bragg peak at place of photoelectron birth









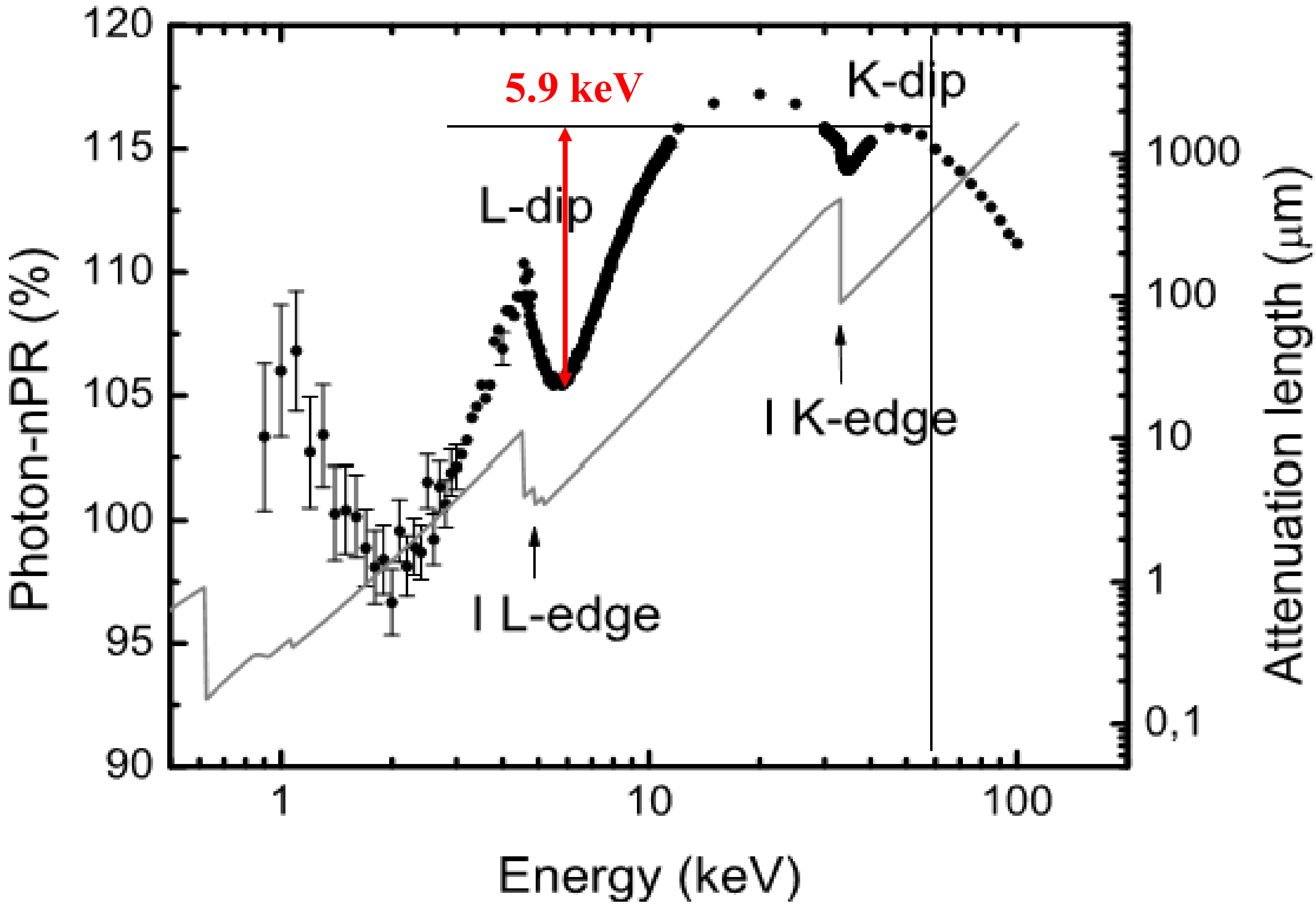
Nucleus receives recoil pulse

Direction of electron coincides with $\vec{E}$

Energy loss *dE/dx* is increased

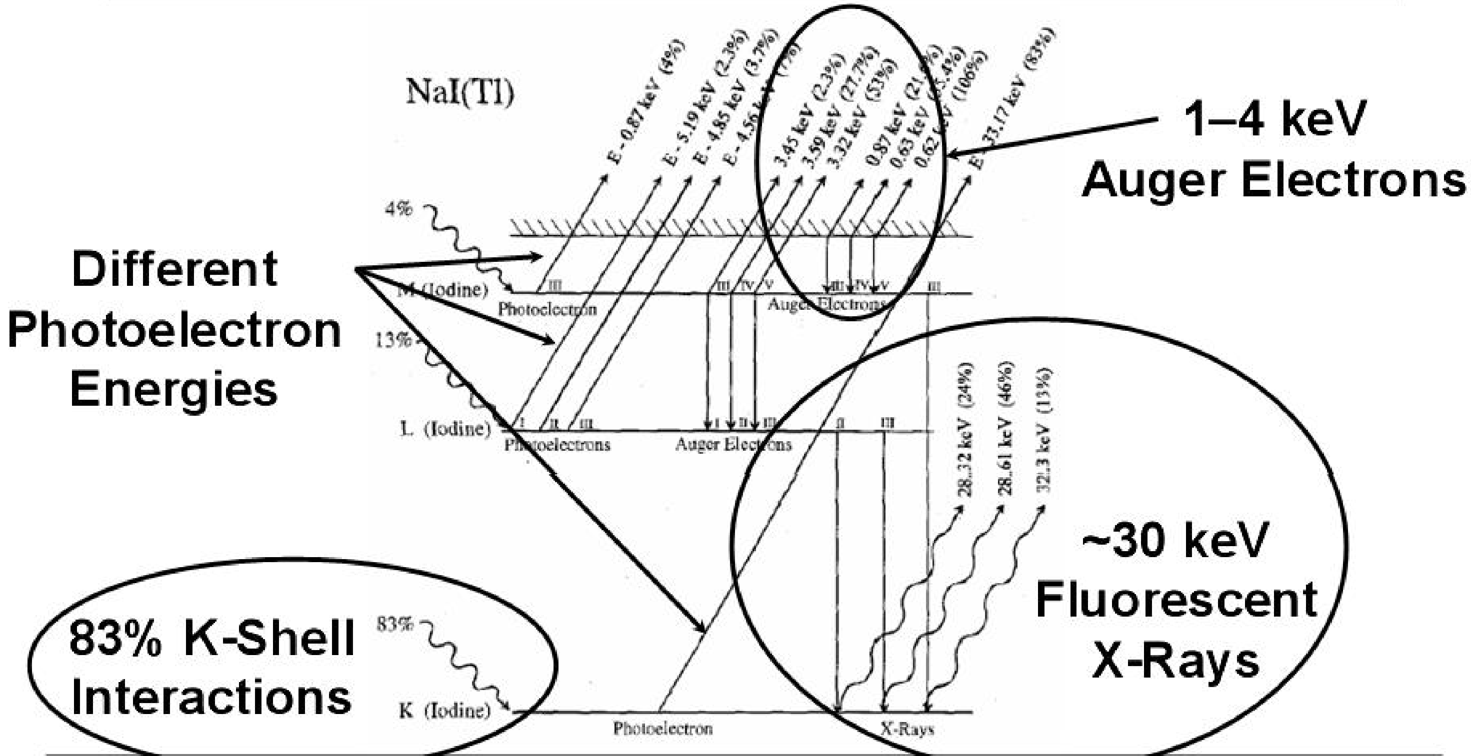
~ twice near L-edge

figure: *I. Khodyuk, P. Dorenbos, IEEE TNS, 2012*



A point 5,9 keV can be lifted from 106 to 116% if $C_{Tl} > 7.3 \cdot 10^{-2}$ mole%

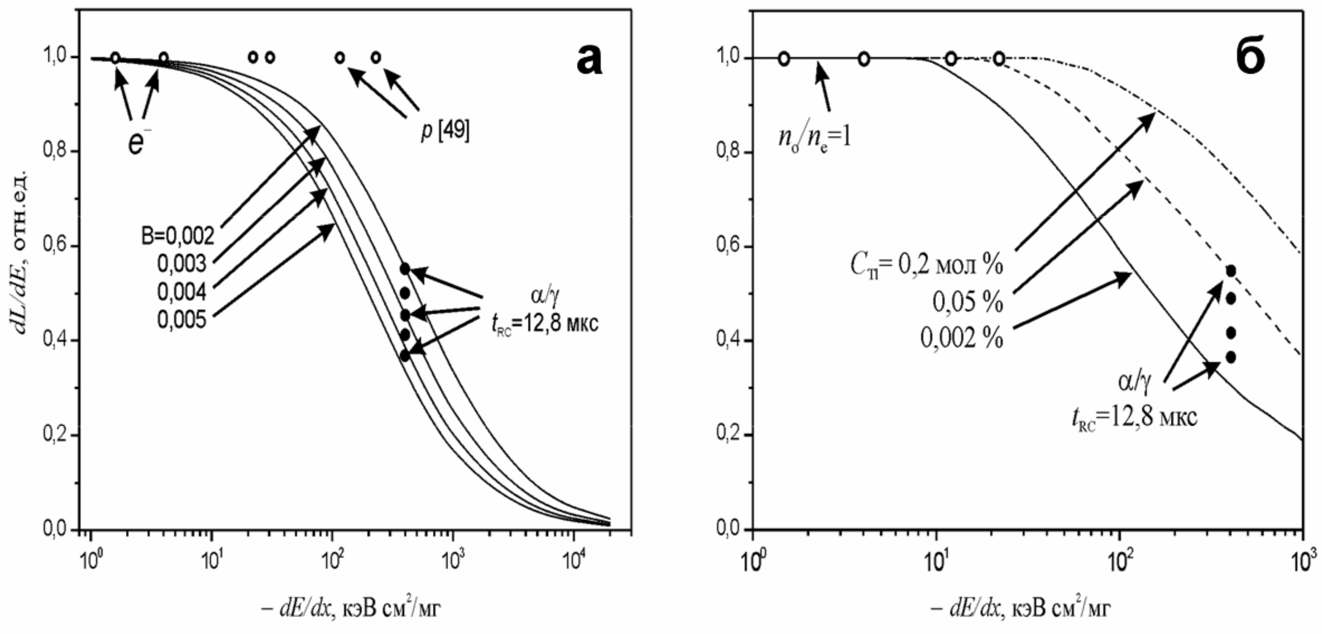
Simplified Cascade Diagram for NaI



- Many Energetic (>1 keV) Particles Created
- Fluorescent X-Rays & Auger Electrons

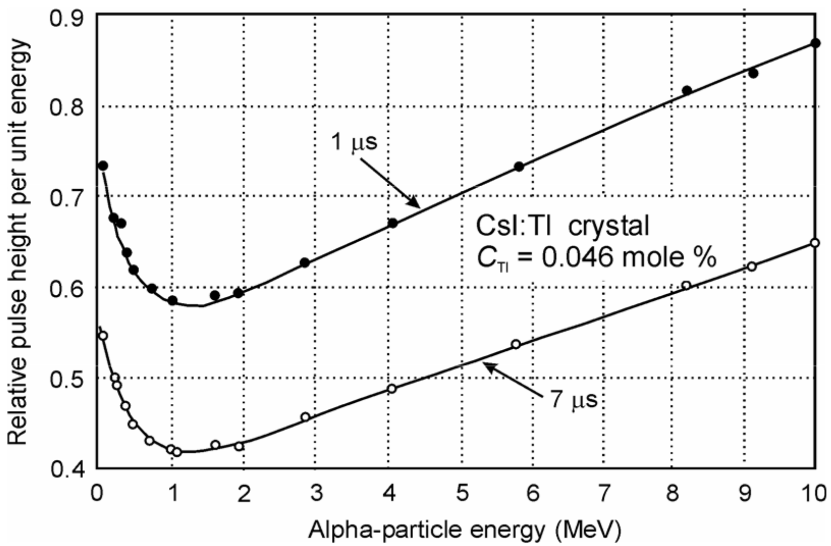
Figure from B.D. Rooney & J.D. Valentine, IEEE Trans. Nucl. Sci. 44, pp. 509–516, 1997

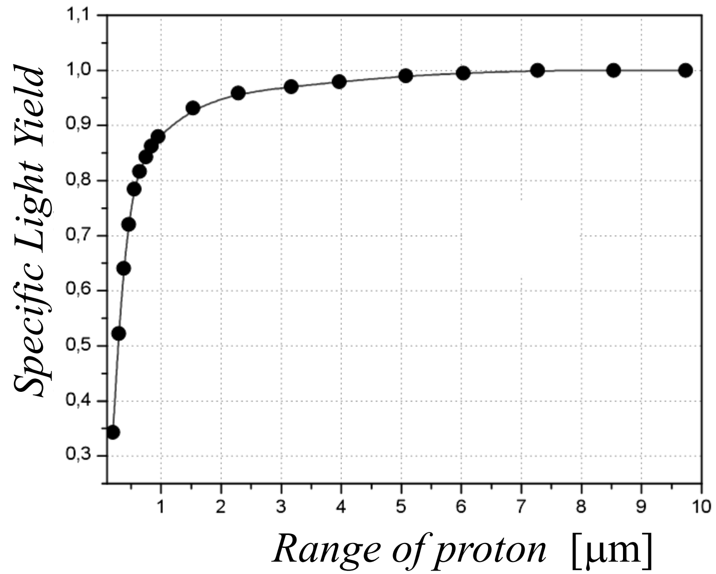
Application of Living Layer to Theory Verification



A.M.Kudin, Dr.Sci.
Thesis, 2007

Usikov, Vyday, et al. 1983





Gwin, Murray, 1963