CsI(Tl) and ZnSe(Te) columnar films

A. Lebedynskiy
Institute for Scintillation Materials NAS of Ukraine

Motivation

- ***General***

  - application of scintillation films (ionizing radiation imaging systems)

- ***Current meeting topics***

  - different properties of material in bulk and film form ⟶ *structures and types of defects*

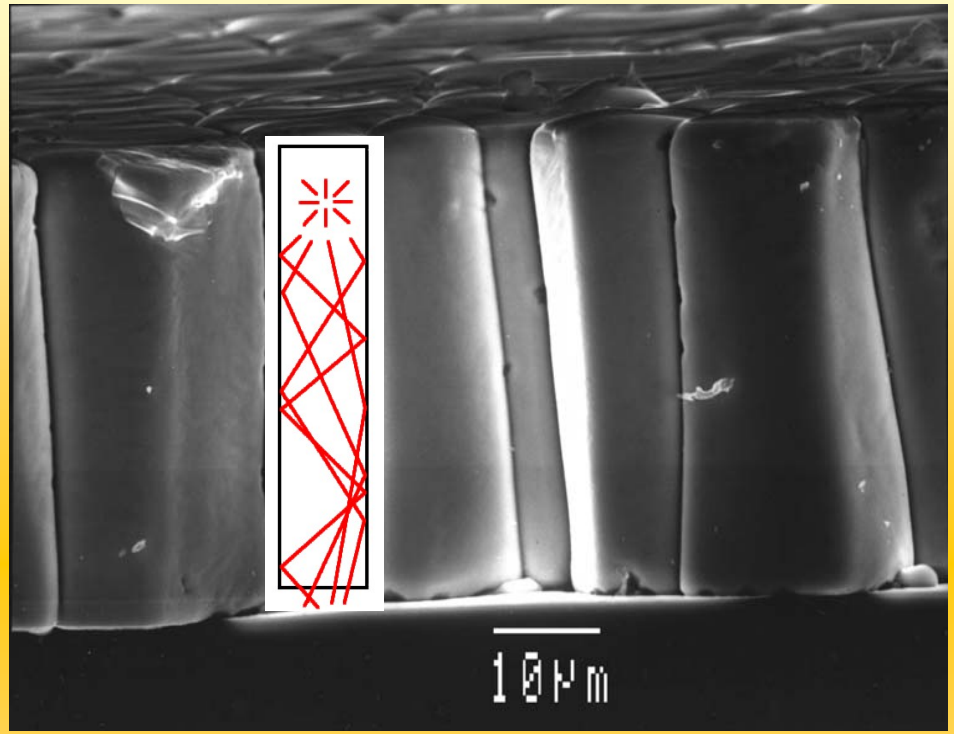
  - “nanophysics”

Motivation

- ***General***

  - application of scintillation films (ionizing radiation imaging systems)

Why do we need scintillation films of columnar morphology?



A column, suppressing the lateral light spread of scintillations due to the total internal reflection, acts as a light guide

The following scintillation materials have been deposited in the columnar morphology:

CsI(Tl),

CsI(Na),

ZnSe(Te),

ZnTe(O)

$LaBr_3$(Ce),

$LaCl_3$(Ce),

LiI(Eu) etc.

Institute for Scintillation Materials has a great experience in the research and production of scintillation crystals based on alkali-halide and chalcogenide compounds.

CsI, CsI(Tl), CsI(Na), NaI(Tl) and so on

The first studies of thin (about 5-10 mkm) films of CsI doped with Tl and Na were begun in early 70's of the last century

The deposition and investigation of thick columnar films of CsI(Tl) were started in 2002.

ZnSe(Te), ZnTe(O)

The deposition and investigation of ZnSe(Te) and ZnTe(O) was started in 2004

CsI(Tl) columnar films

Thick (50 – 1000 mkm) CsI(Tl) columnar films were obtained by Physical Vapour Deposition (PVD) technique on different substrates materials.

Substrate materials:

cleavage plane (100) of LiF, NaF single crystals; *(to investigate the possibility of obtaining a single crystalline structure of layers on the orienting substrate)*

- ***glass of different thickness (from 180 mkm up to 1300 mkm);***
- ***graphite plates;*** *(for possible application)*
- ***silicon.***

Substrate temperature - 296–773K.

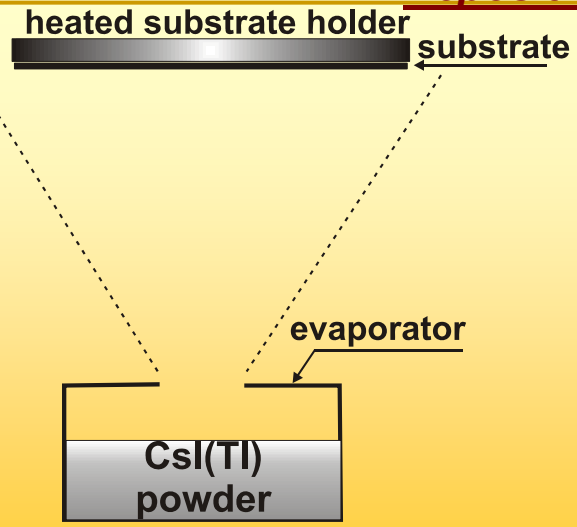
The thickness of CsI(Tl) films was in a range from 40 to hundreds micrometers.

Concentration of Tl in a source material

from $9 \cdot 10^{-2}$ mass% (0.11 mol%) up to $1.26 \cdot 10^{-1}$ mass% (0.16 mol%).
(These values correspond to the "plateau" on the chart of the scintillations light output versus the activator concentration)

CsI(Tl) columnar films

Deposition and scintillation properties



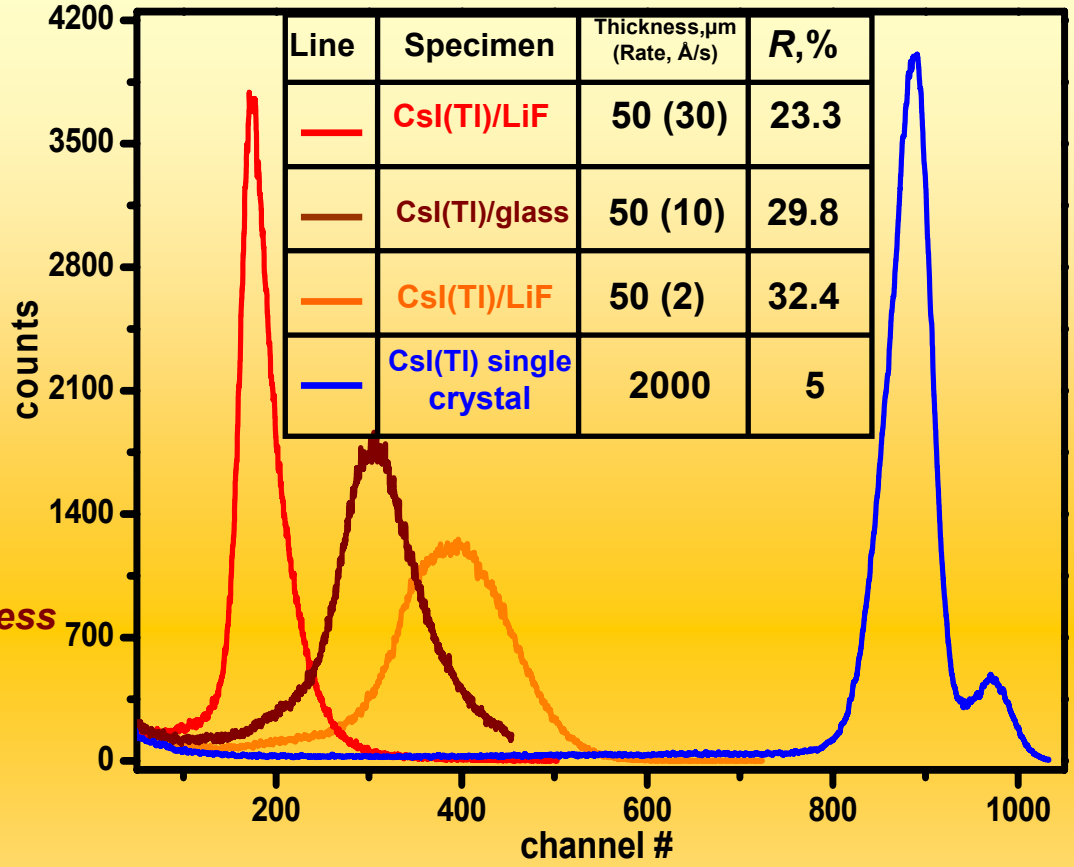
The difference in partial vapour pressures of CsI and TlI depletes the activator from source material during the deposition process (initial C_{Tl} - $1.6 \cdot 10^{-1}$ mol %, after deposition - $2.03 \cdot 10^{-5}$ mol %).

The consequences of such a depletion:

reduction of light yield (LY);

LY depends on deposition rate;

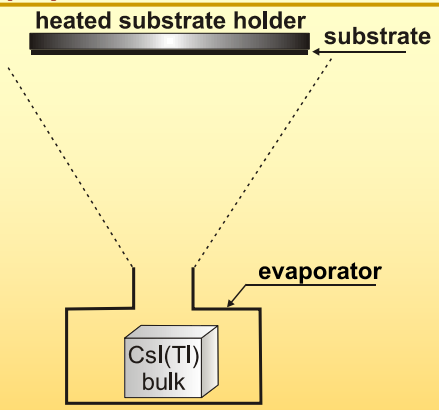
nonuniform distribution of Tl in film volume, which degrades energy resolution (R)



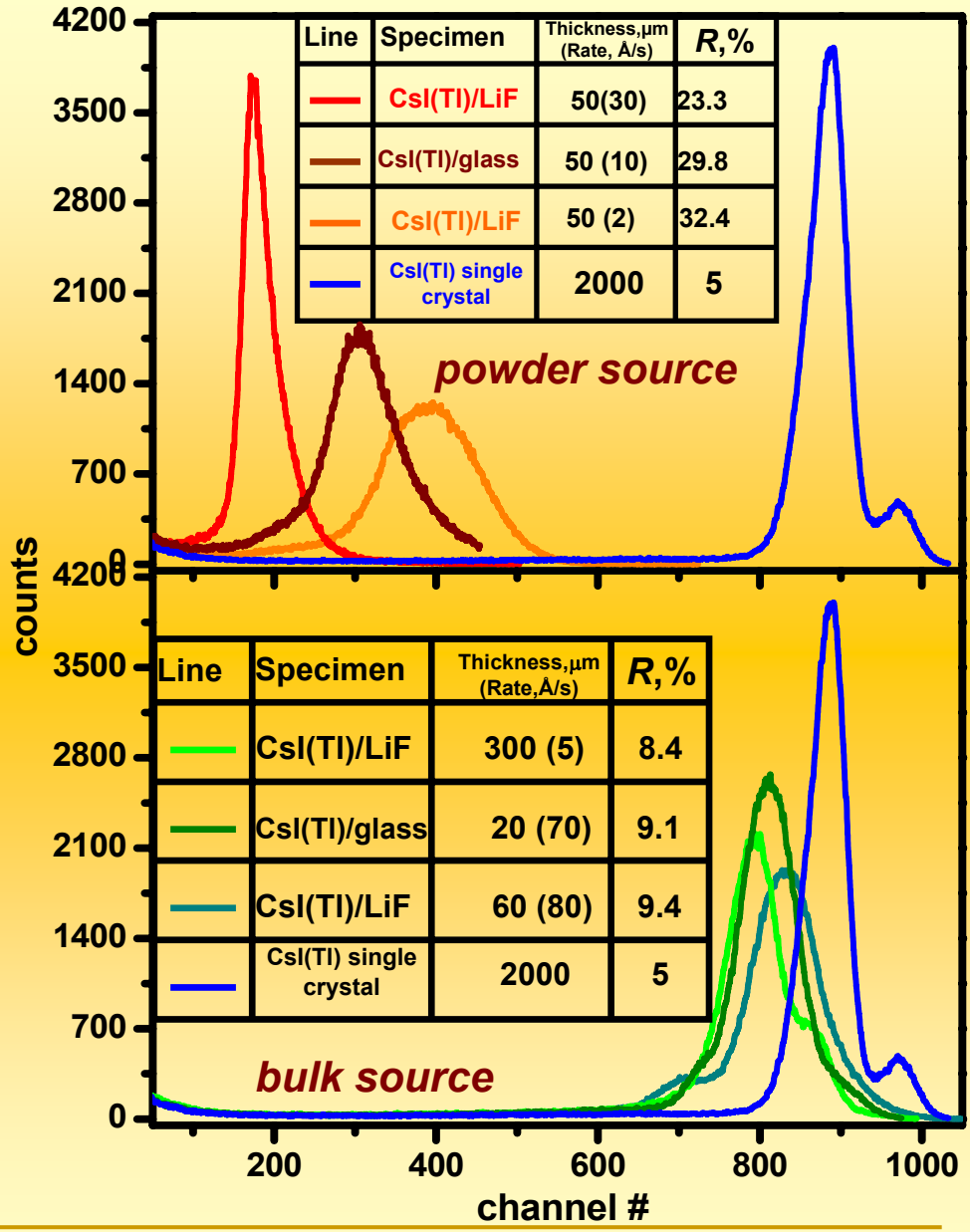
All the spectra were obtained under α-irradiation (^{239}Pu)

CsI(Tl) columnar films

Deposition and scintillation properties



Unlike powder CsI(Tl), the sublimation of bulk CsI(Tl) allows suppressing the Tl diffusion in source material, which ensures the uniform distribution and reproducible concentration of dopant in the film. The same amount of Tl in the film and in the initial source material was ascertained by atomic absorption analysis. A sublimation of bulk CsI(Tl) crystal leads to reproduction of the required concentration and uniform distribution of the dopant in growing films.



Results:

increase of LY to the level of single crystal;

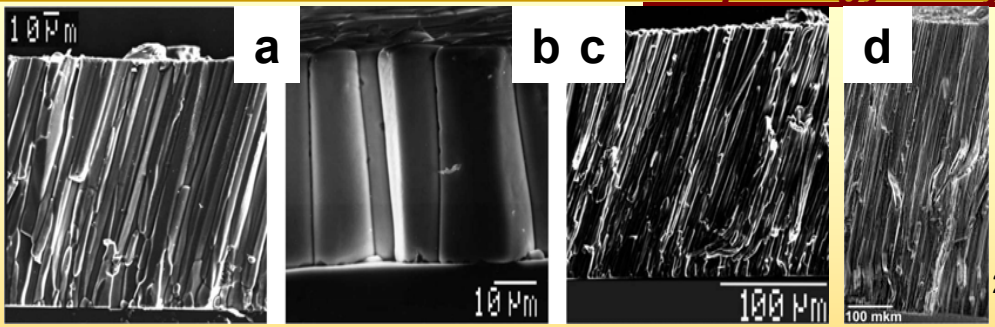
LY doesn't depend on deposition rate;

R value of CsI(Tl) films can be compared with single crystal.

All data presented in figure were measured under α-irradiation (^{239}Pu)

CsI(Tl) columnar films

Morphology & crystalline structure

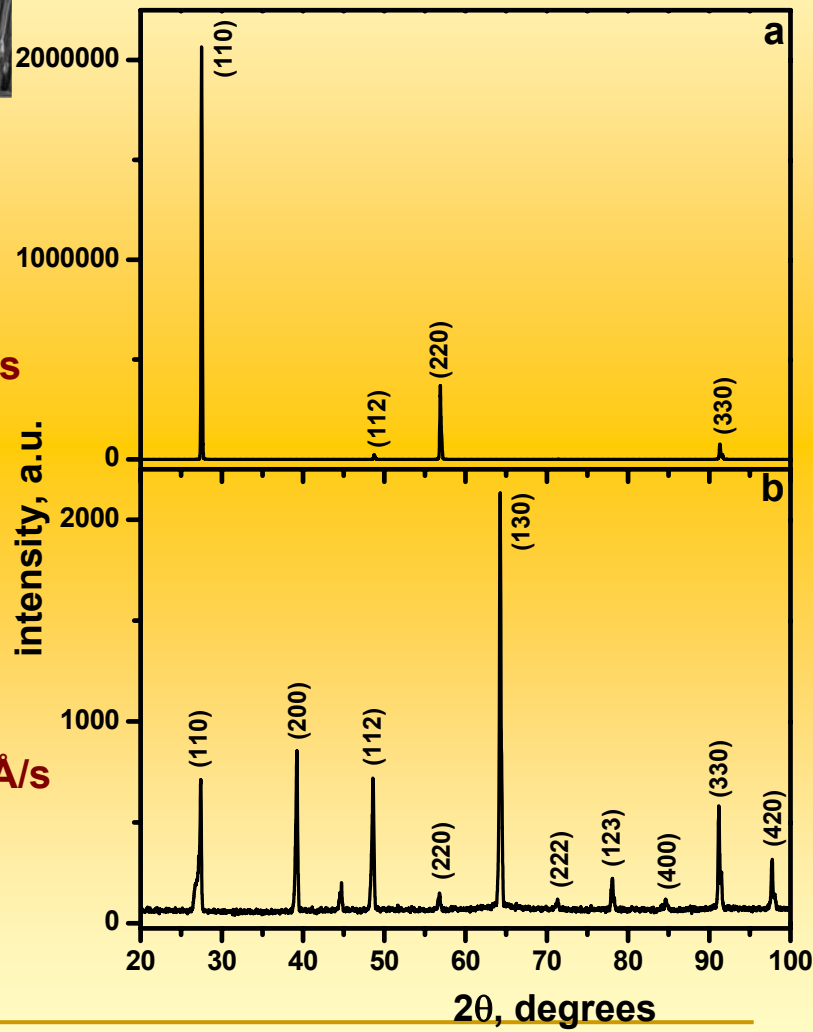


SEM image of CsI(Tl) film cross-sections:

a) – CsI(Tl)/(100) LiF (T_{sub} = 573K; deposition rate 85 Å/s);
b) – CsI(Tl)/(100) NaF (T_{sub} = 573K; deposition rate 8.2 Å/s);
c) – CsI(Tl)/glass (T_{sub} = 573K; deposition rate 120 Å/s);
d) – CsI(Tl)/glass (T_{sub} = 573K; deposition rate 50 Å/s)

CsI(Tl)/(100) LiF
Deposition rate – 5 Å/s
T_{sub}=573K

CsI(Tl)/(100) LiF
Deposition rate – 117 Å/s
T_{sub}=433K

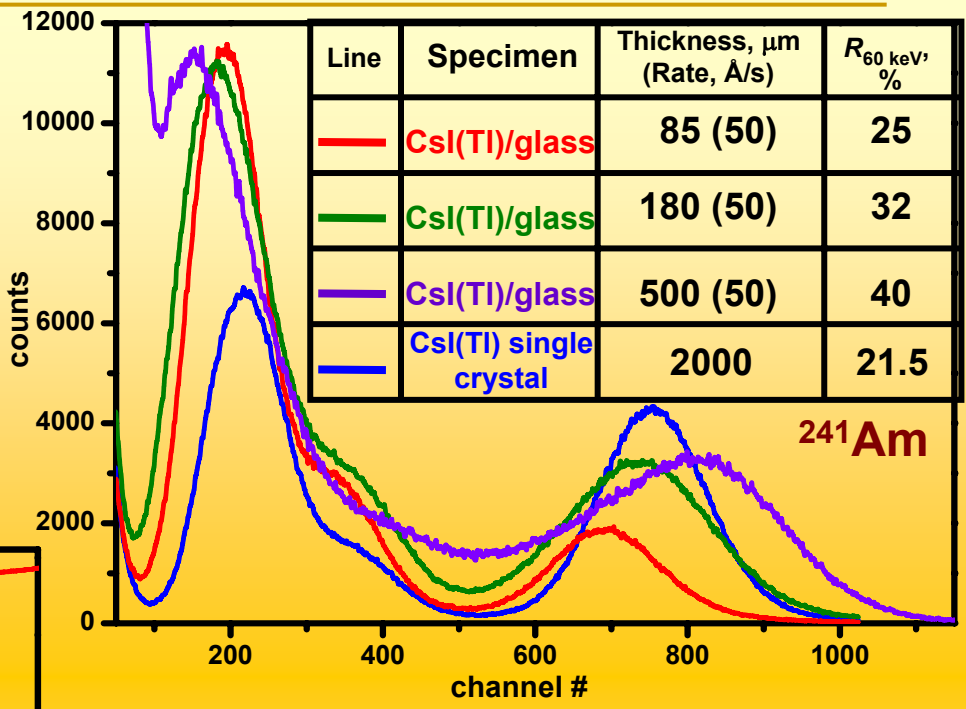


X-ray diffraction patterns of CsI(Tl) films. Miller indices of reflections are denoted.

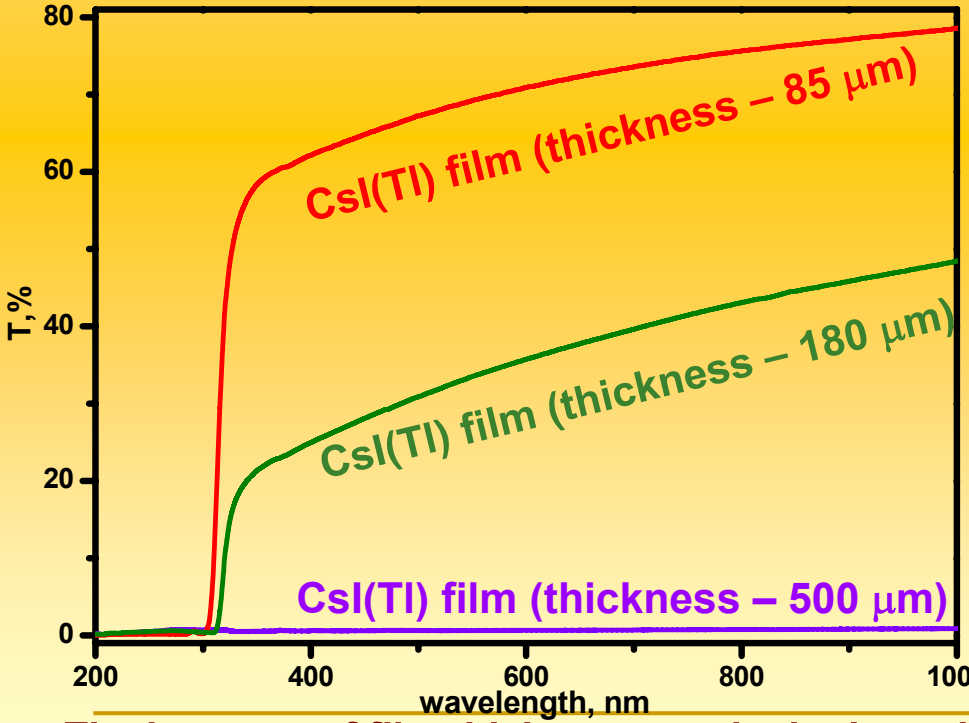
CsI(Tl) columnar films

Scintillation properties

The increase of the CsI(Tl) columnar films thickness deteriorates their transmission due to rise of light scattering centers number in the columns volume.



Line	Specimen	Thickness, μm (Rate, Å/s)	$R_{60\ keV}$, %
	CsI(Tl)/glass	85 (50)	25
	CsI(Tl)/glass	180 (50)	32
	CsI(Tl)/glass	500 (50)	40
	CsI(Tl) single crystal	2000	21.5



Consequences

degradation of energy resolution (R);

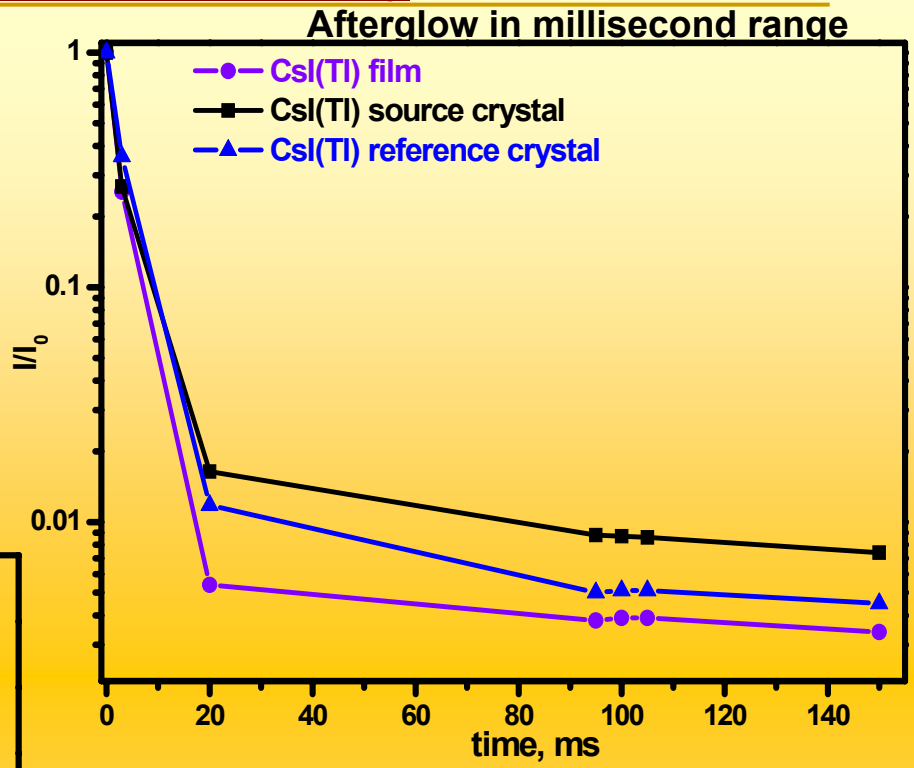
suppressing of an effective light channeling leads to decrease of LY in low energy range;

the number of light scattering centers goes up leading to a higher light output under high energy of γ-radiation

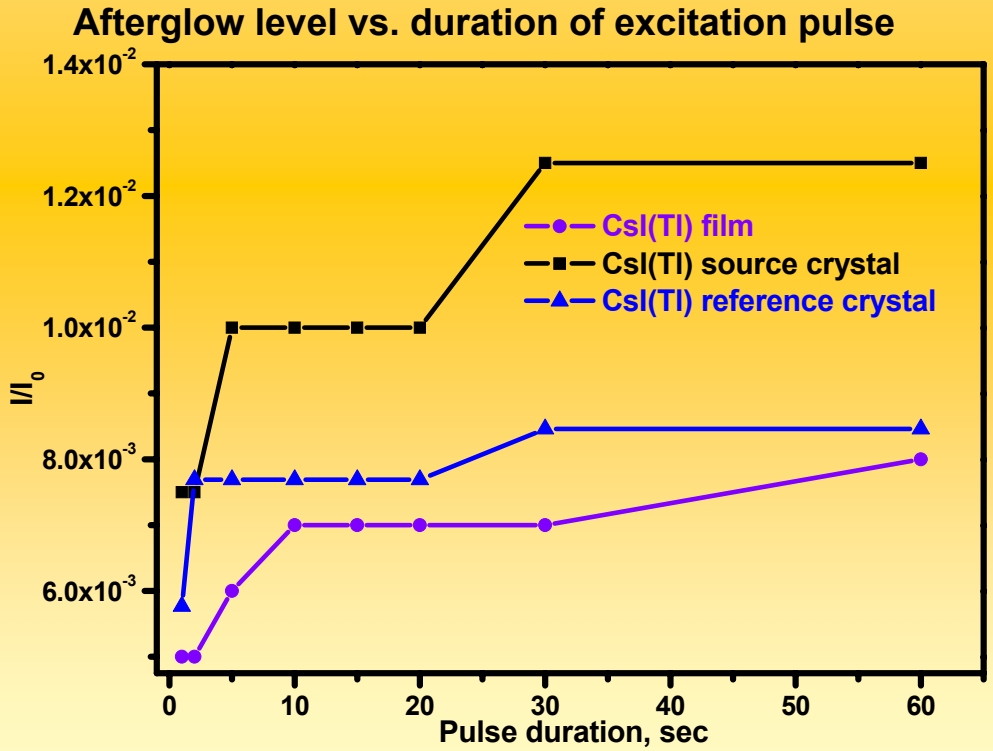
The increase of film thickness results in degradation of energy resolution R (for energy 59.5 keV) and decrease of light yield in low energy range

So, there is a disbalance between light collection (channeling) and scattering

- **Current consortium application**
  - different properties of material in bulk and film form

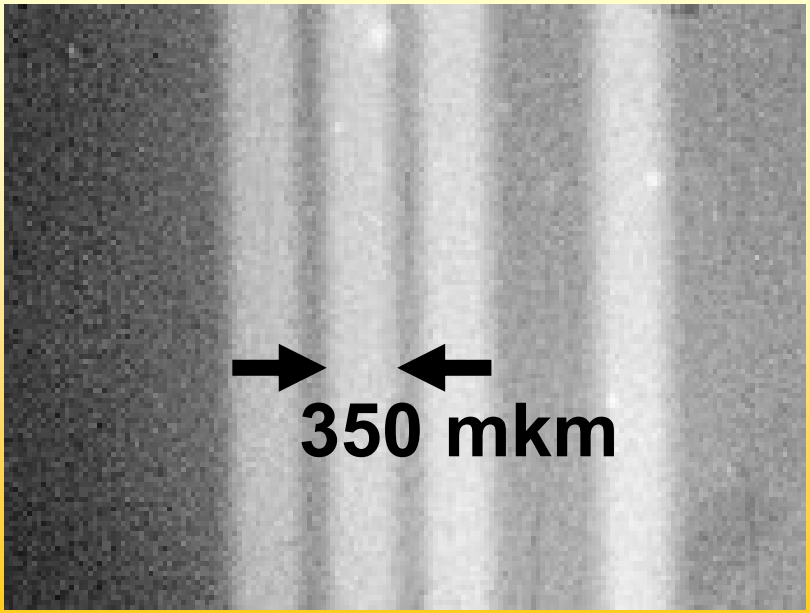


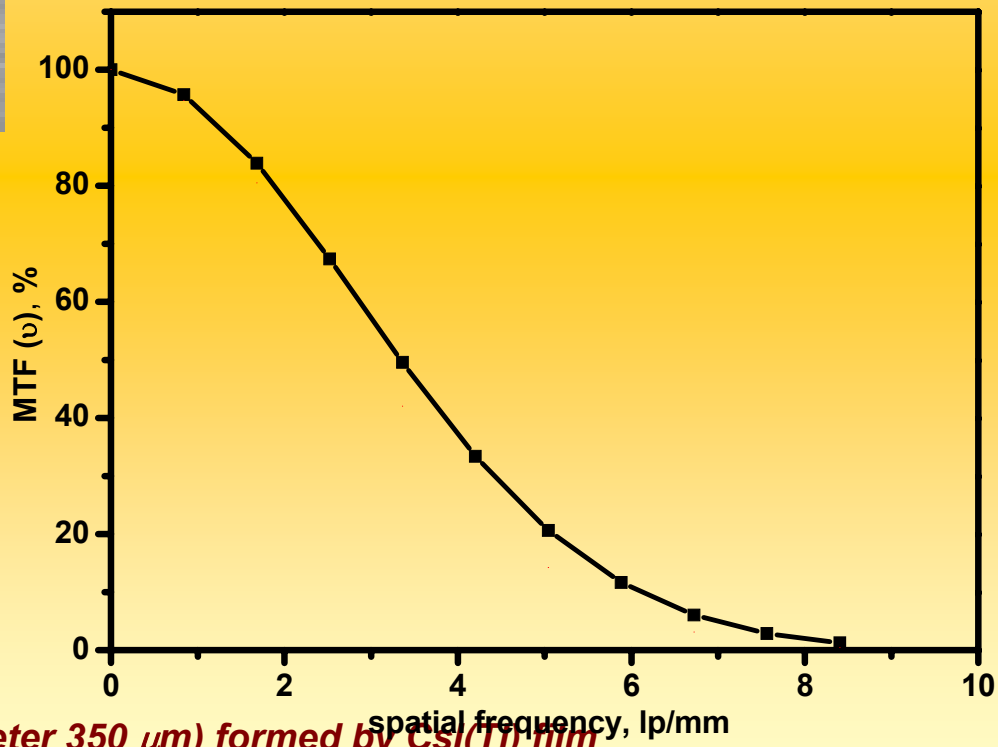
The afterglow level of CsI(Tl) films is about 1.5 times lower than that in source CsI:Tl crystal in millisecond range .



This effect remains the same even when the radiation dose is 60 times higher. This demonstrates a better radiation stability of the obtained films.

Afterglow level was measured after X-ray (140 keV, pulse duration 6 seconds) irradiation.





Tantalum (Ta) wires (diameter 350 μm) formed by CsI(Tl) film in γ - radiation (source ^{241}Am)

Motivation

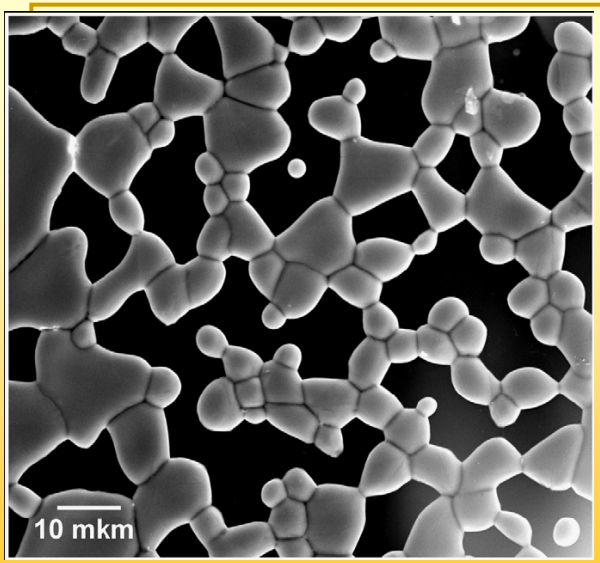
- ***General***

  - application of scintillation films (ionizing radiation imaging systems)

- ***Current meeting topics***

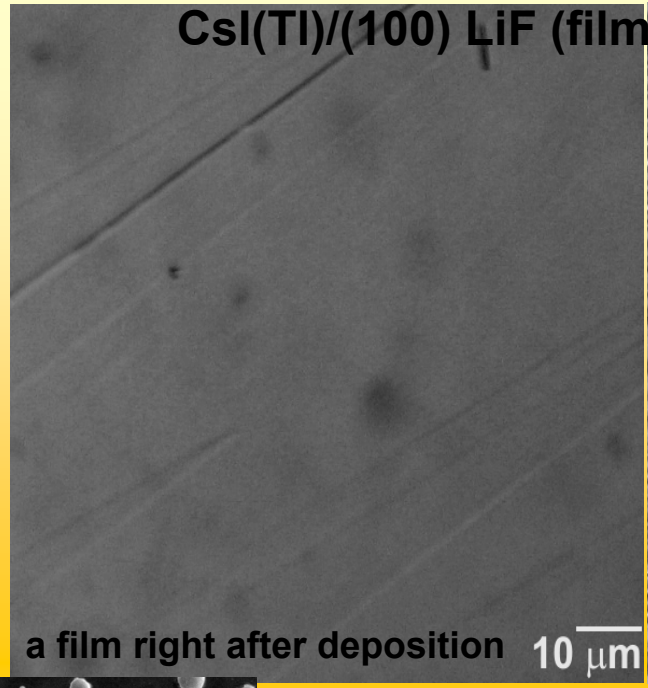
  - different properties of material in bulk and films form ⟶ *structure and types of defects*
  - "nanophysics"

Thin CsI films

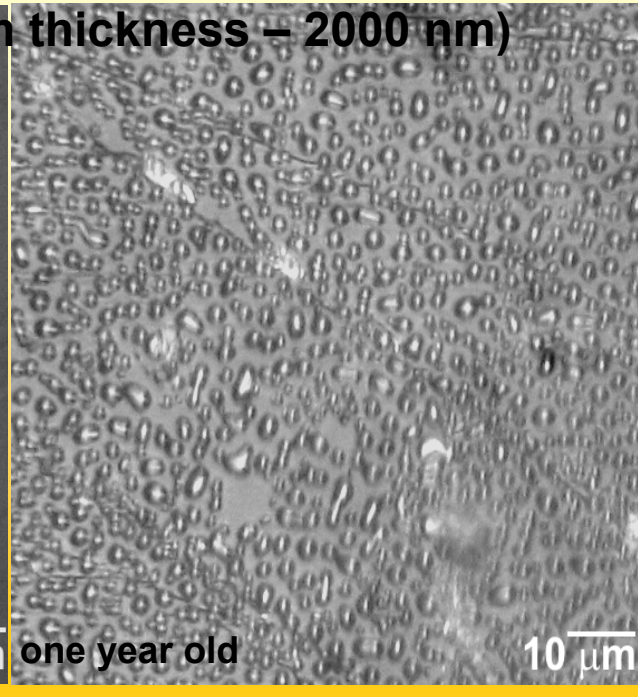


The SEM image of CsI(Tl) film on the LiF substrate

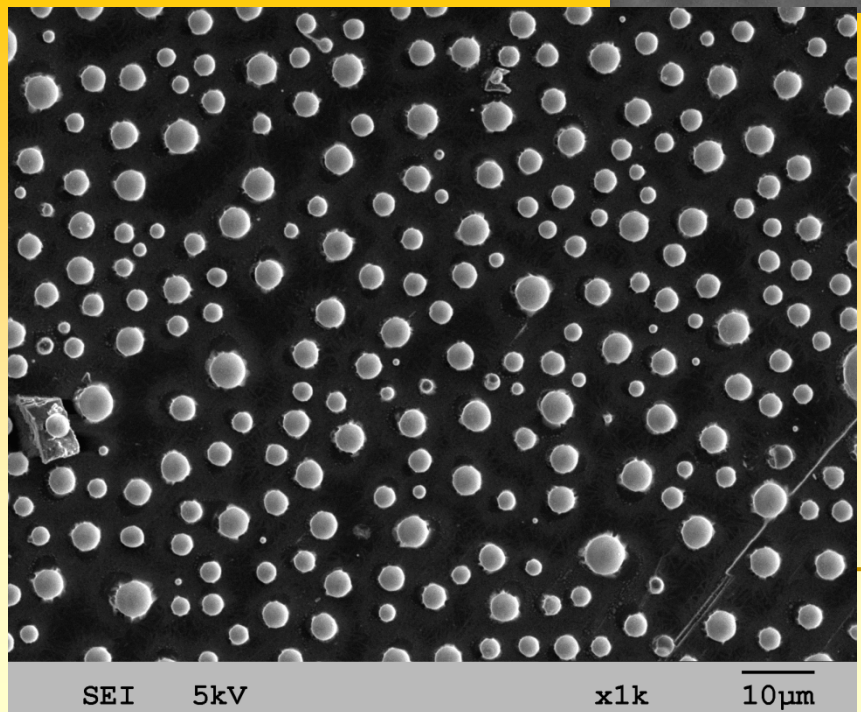
CsI(Tl)/(100) LiF (film thickness – 2000 nm)



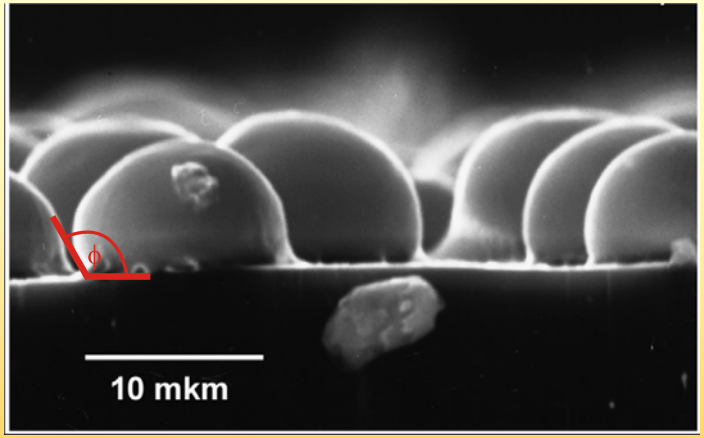
a film right after deposition



one year old



Thin CsI films



SEM image of cross section of CsI(Tl) film deposited on (100) LiF

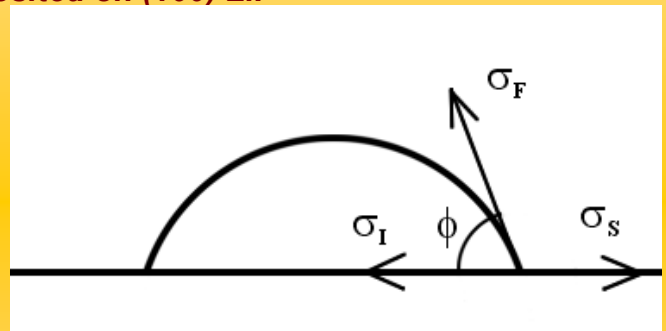
Young's equation $\sigma_F \cos\phi + \sigma_I = \sigma_S$

where σ_F **- the surface energy of the film,**

σ_I **- the interface energy,**

σ_S **- the substrate surface energy,**

ϕ **- the contact angle.**



$$\sigma(hkl) = n_0(hkl)\frac{Me^2}{4\pi\varepsilon_0 r_0}\left(1-\frac{1}{n}\right)\sum_{j=0}^{\infty}\left(\frac{M_j}{M}-1\right)$$

(zeroth approximation)

The surface energies calculated in zeroth approximation:

LiF (100) σ_S ***= - 0.765 J/m²***

CsI (110) σ_F ***= - 0.208 J/m²***

the contact angle (ϕ) of CsI particles is about 110°

$$\sigma_I = -0.836\,J/m^2$$

ZnSe(Te) columnar films

Thick (up to 500 mkm) ZnSe(Te) columnar films were prepared by Physical Vapour Deposition by hot wall technique.

Substrate materials:

graphite plates *(graphite is rather transparent for γ- and X-ray radiation, has the same temperature expansion coefficient as ZnSe(Te) and prevents films from cracking and peeling*

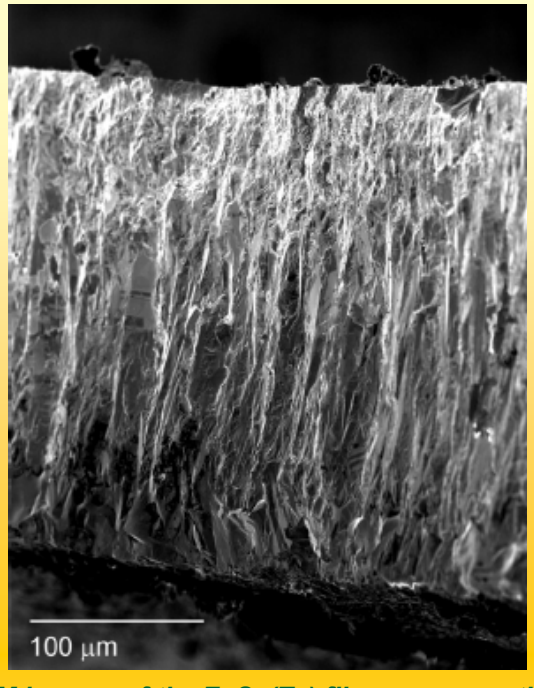
ZnSe(Te) films were deposited in a range from 200 to 600 micrometers

Source temperature varied in a range 970 − 1100 K.

This temperature range provides the sublimation of the source material. The equivalent concentration of Te (0.4 wt.%) both in the source material (crushed crystals) and in the deposited film was confirmed by the electron probe microanalysis and atomic absorption analysis.

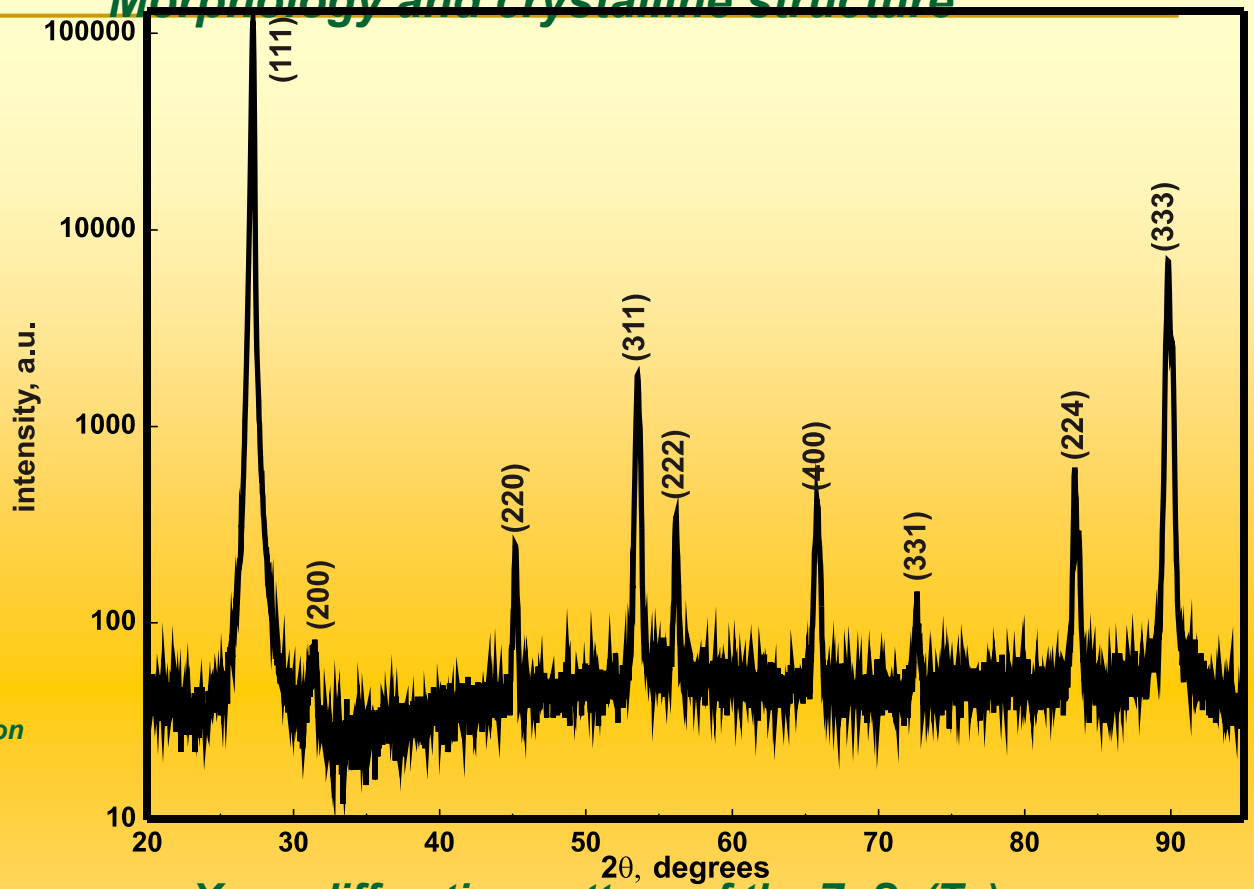
As this temperature range is below the wurtzite-sphalerite phase transition, the wurtzite phase inclusions do not occur in the films.

ZnSe(Te) columnar films



SEM images of the ZnSe(Te) films cross section

Morphology and crystalline structure

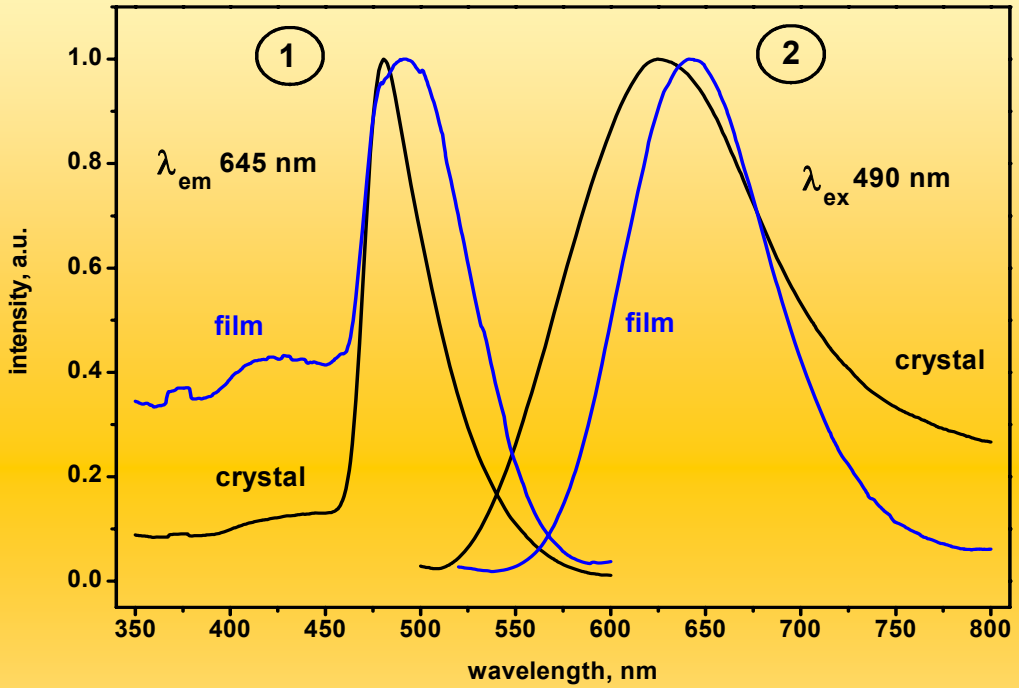


X-ray diffraction pattern of the ZnSe(Te) vacuum deposited film. Miller indices of reflections are denoted

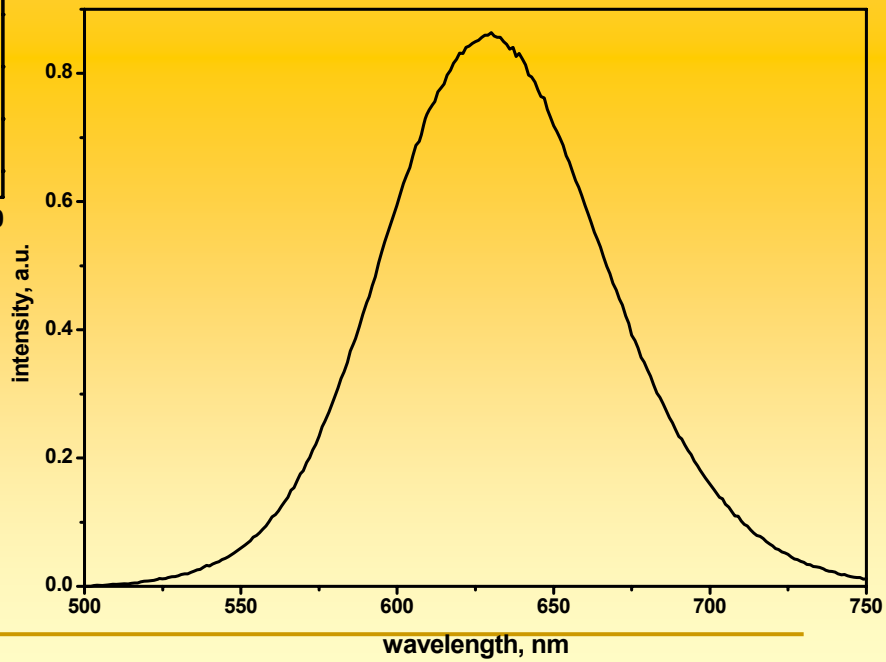
Luminescent properties

ZnSe(Te) films right after deposition **have no luminescence**

Films were annealed in Zn vapour according to the previously developed technology for ZnSe(Te) crystals.

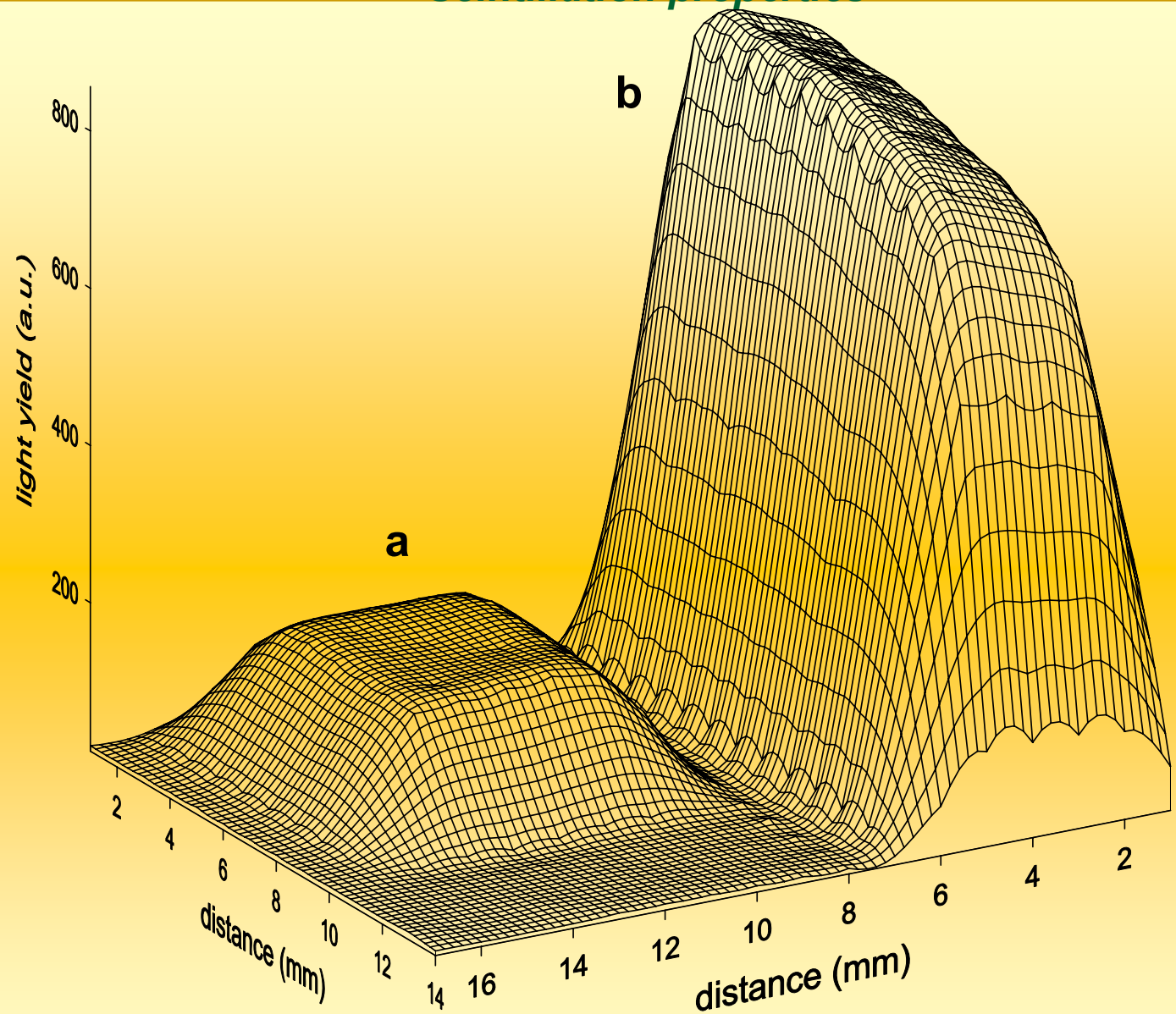


Excitation (1) and luminescent (2) spectra of ZnSe(Te) layer (*blue*) and crystal (*black*)



Radioluminescent spectra of ZnSe(Te) film (excitation by X-ray tube at 25 kV)

Scintillation properties



Relative light yield of ZnSe(Te) film (380 μm thick) on the graphite substrate(a) and 1 mm thick ZnSe(Te) bulk crystal (b). Excitation with X-ray tube at 90 kV.

Conclusions

General

- *CsI(Tl) and ZnSe(Te) columnar films with thickness ranging from 50 to 1000 mkm have been successfully grown in a one-stage process.*
- *The sublimation of bulk source material was the key of deposition mode, which allows reproducing the composition of the source material in film.*
- *Obtained thick columnar films demonstrate scintillation properties compared with the bulk crystal.*

Current meeting topics

- *Materials in film form - new objects for study.*
- *The rupture of thin films due to dewetting behavior can be employed for nanosize particles production*