Radiative relaxation of electron excitations in CsI:X (X=Tl, In). Temperature and concentration dependences.

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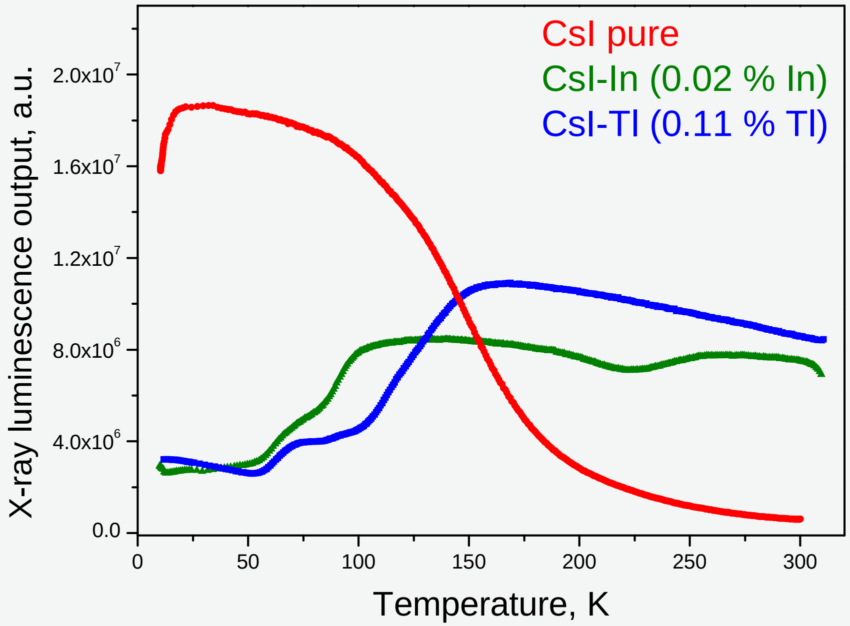
Motivation

Yield of STE emission vs the yield of activated CsI



Scintillator	LY Exp, ph/MeV	<E>/photon, Eg
CsI 77K	**115,000**[1]	1.5
CsI:Tl 300K	**56,000**	2.9

[1][Moszynski et al, 2005]

High efficiency of STE emission in CsI - self-trapping of holes

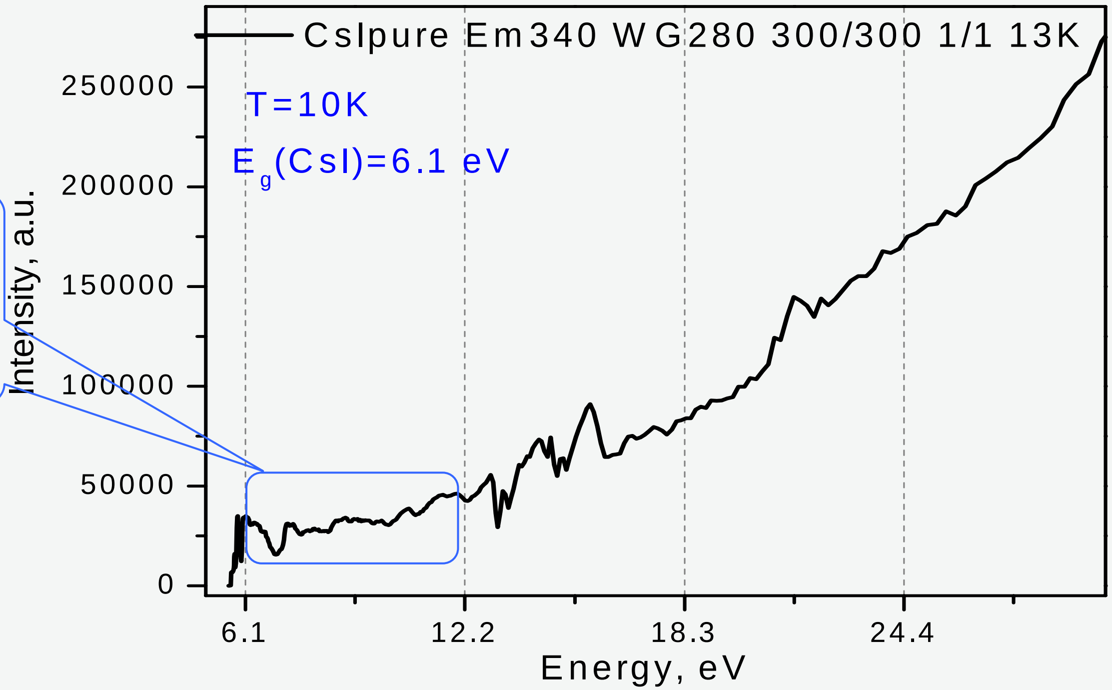
CsI:Tl scintillator gives only ½ of the CsI potential
- high quality CsI pure
- "genetic" e-h recombination in CsI is highly probable
- efficient recombination of "non-genetic" e-h pairs in pure CsI

What is the origin of the energy loss?

Excitation of STE emission in CsI pure

Efficient excitation in E_g-$2E_g$

Separated e-h pairs bound into STE state



$E_v < E_g$

$$\varepsilon = E_g e^{2C}\left[1+\int_0^\infty \frac{(v-1)e^v+1}{v^2}\left(1-e^{2Ei(-v)}\right)dv\right]^{-1} = 1{,}38\,E_g.$$

- no migration loss

A. Vasil'ev, V. Mihailin. *Role of phonon relaxation in the process of cascade multiplication X-ray induced electron excitations*_1986

Outline

- Synthesized In- and Tl-doped CsI scintillation crystals with activator concentration 10^{-4} – 10^{-1} mol %

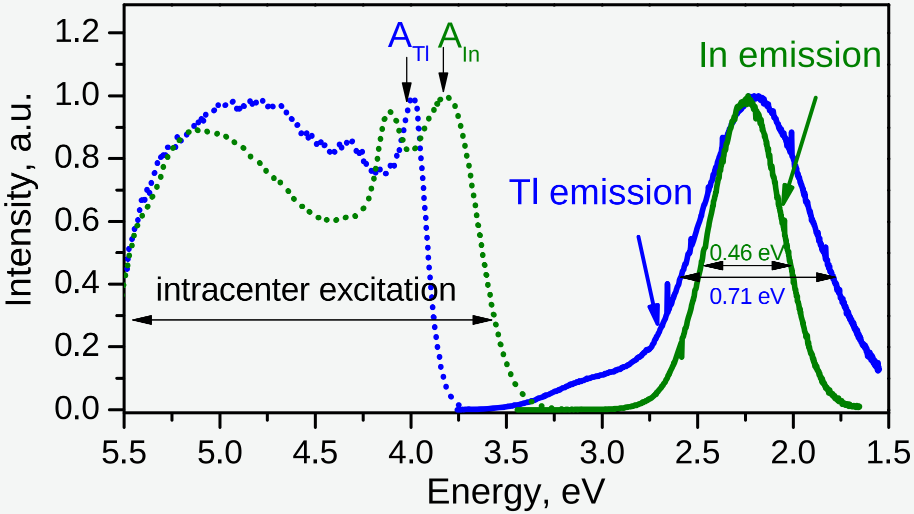
- Investigated
  - general luminescent properties
  - temperature dependence of X-luminescence output (LHT - RT)
  - traps of charge carriers (TSL)

- Comparison with theoretical simulations of scintillation in CsI- based crystals

General luminescent properties of CsI:In and CsI:Tl

Activator-related emission in In- and Tl-doped CsI scintillators

Excitation and Emission spectra of CsI:Tl and CsI:In at 300K



Similar:
main emission band 550 nm
for intracenter and high energy excitation
•same emission centers

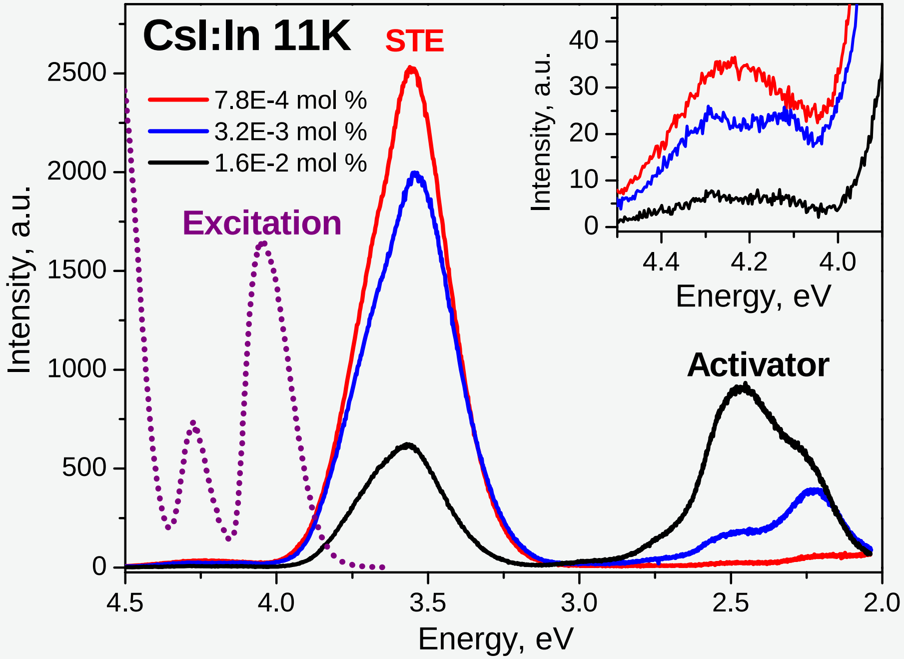
Different:
A-absorption band of In is shifted
to lower energies
•parameters of excitation localization may differ

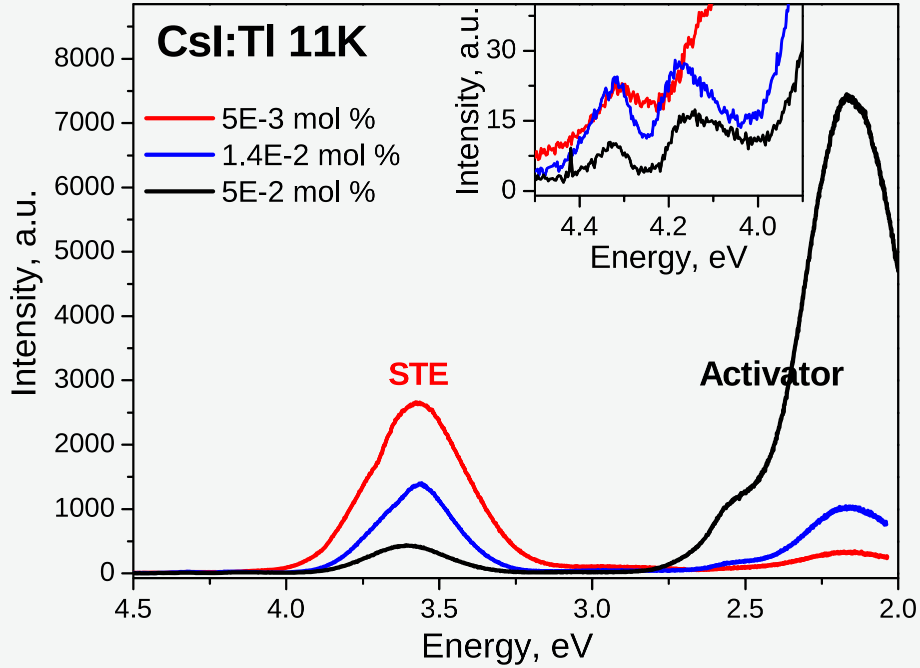
Ion of dopant	Ionic radius, Å	Segregation coefficient	A absorption band, nm	Emission max. at RT, nm	Decay, nces
Tl+	1.59	0.2-0.3	299	550-560	620
In+	~1,35	~0.15	310	545	1900

- CsI:In shows similar luminescent properties with CsI:Tl
- similar energy transfer mechanism for both scintillators can be expected

STE and activator emission in CsI:X at LHT

X-ray luminescenec spectra of CsI:X at LHT for different activator concentrations



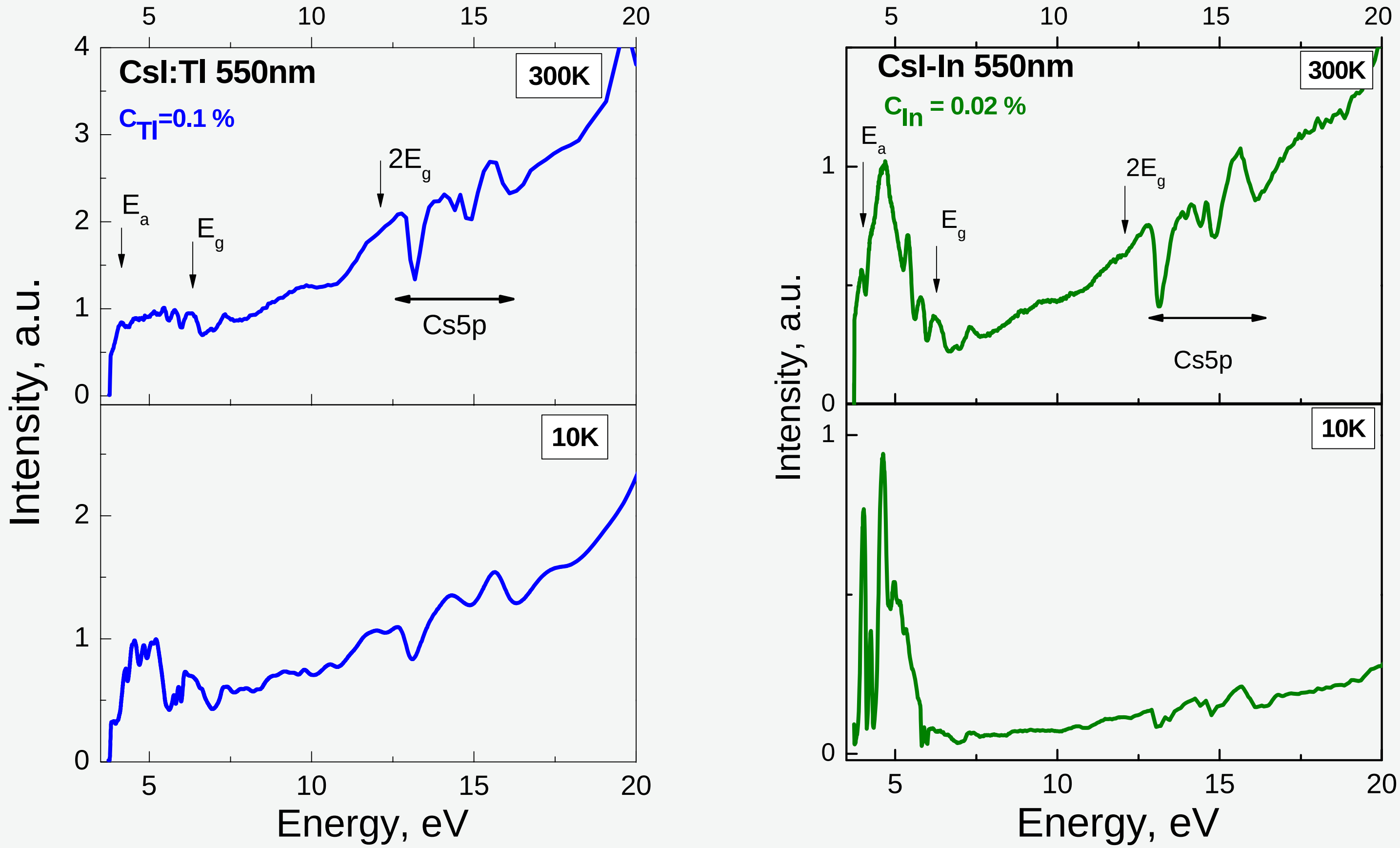


- at least 2 activator-related bands under X-ray
- respective intensity of these bands depends on the concentration

• insignificant reabsorption of the 290 nm band STE band

Proportionality of Yield with excitation energy. Energy transfer

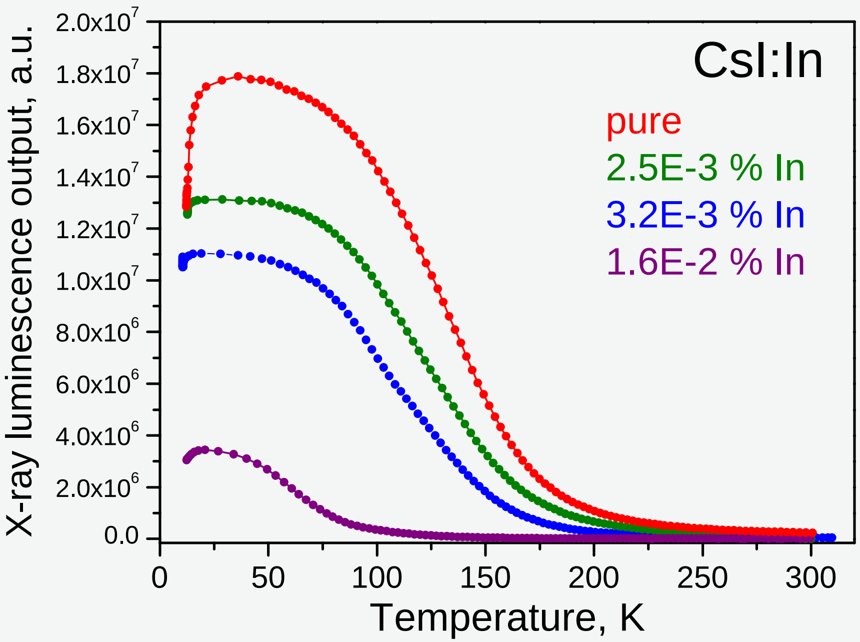
Excitation spectra of In- and Tl-related emission in CsI:X (SUPERLUMI)

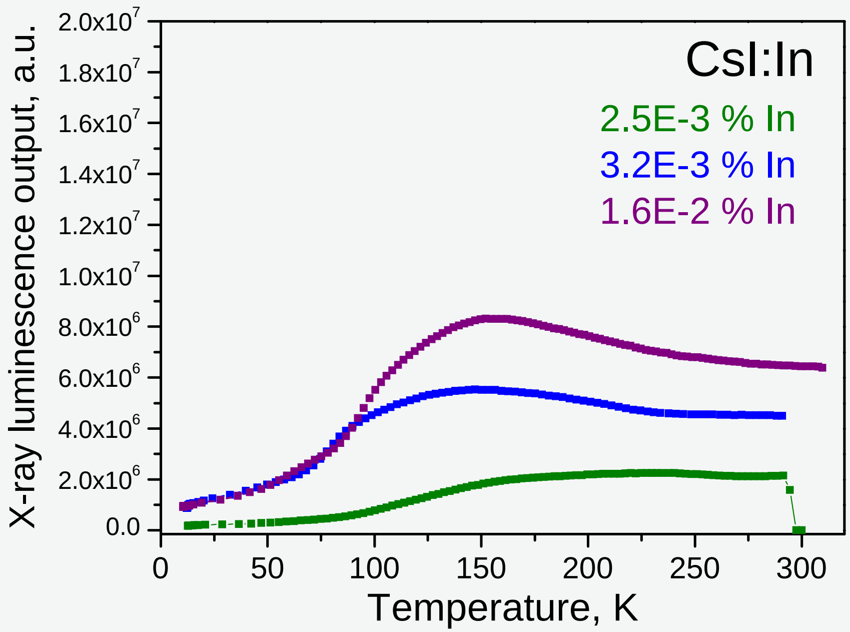


Luminescence intensity in the region E_g – E_g+E_a characterizes the efficiency of emission due to sequential capture of an electron and a hole (or vise versa).

- in both CsI:In and CsI:Tl e-h transfer mechanism is quite efficient
- at low temperature is less efficient than at RT

Concentration dependence of X-excited emission of CsI:X

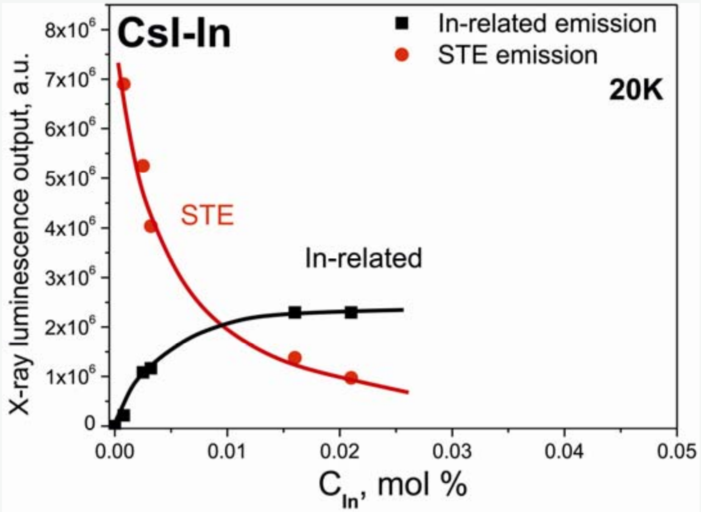


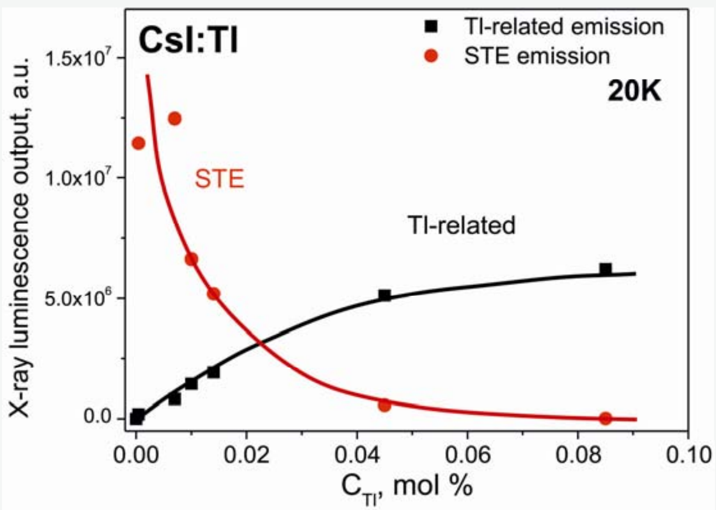


- STE temperature quenching in pure CsI is due to delocalization of holes
- when we start introduce an activator the STE yield at low temperatures falls down rapidly, which is caused by e^- capture by activator
- when temperature goes up STH delocalize and activator emission increases

Concentration dependence of X-luminescence output at **low temperature**. Experiment VS theory

Experiment





Theory

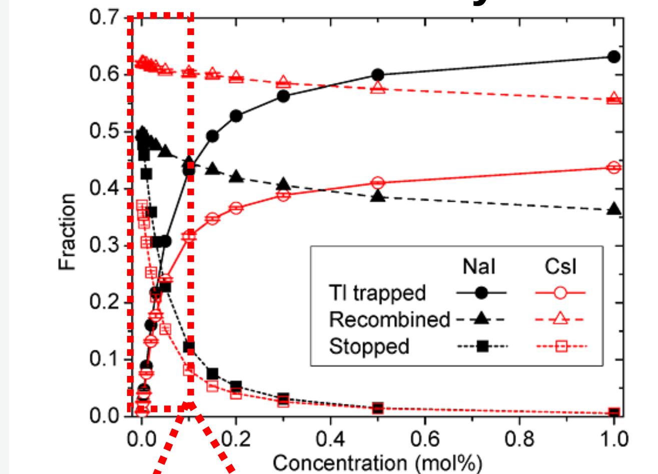
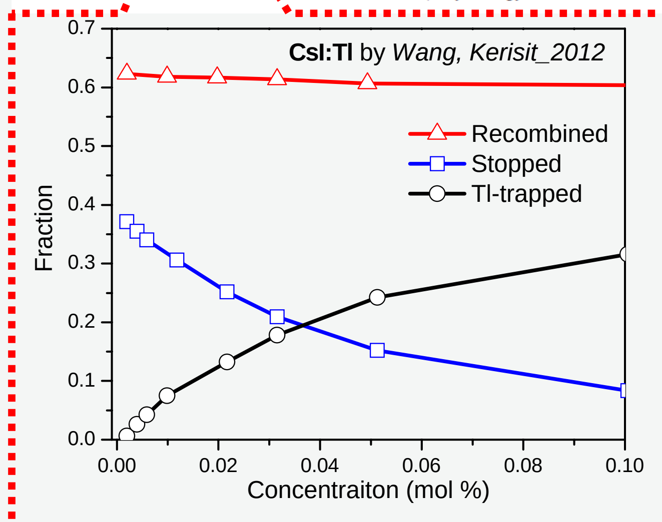


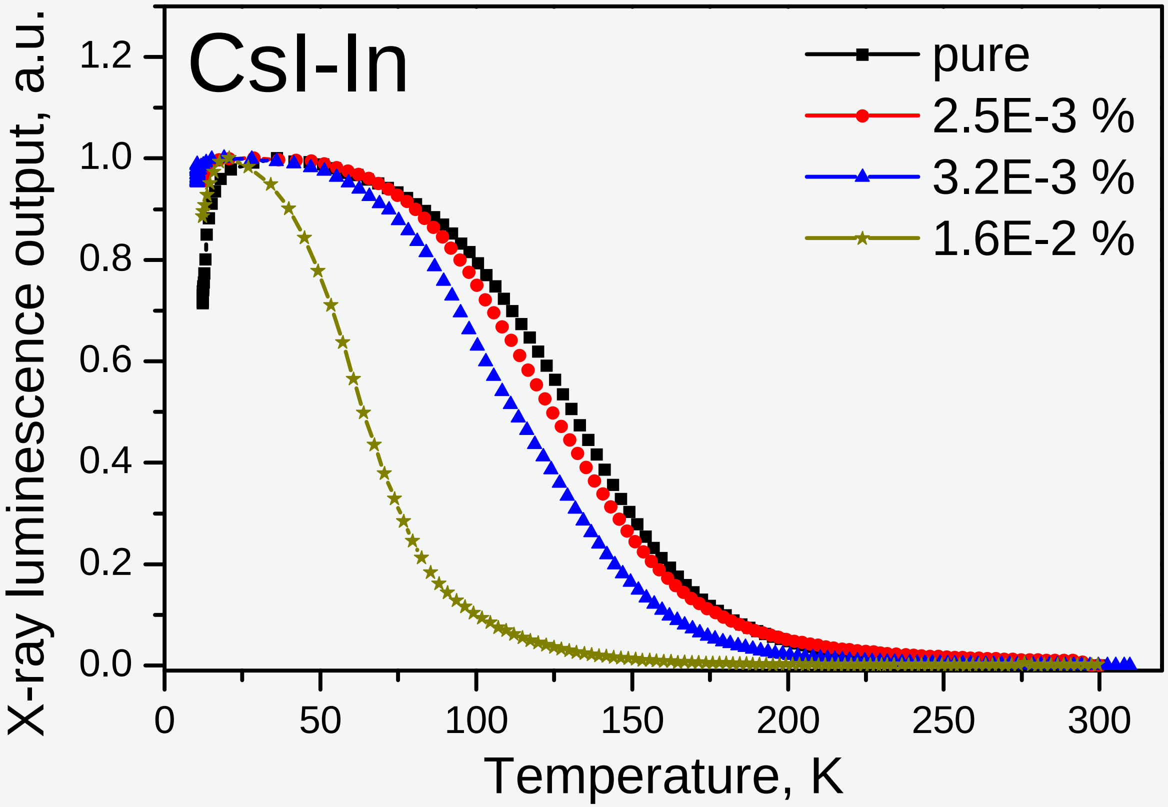
FIG. 6. Fraction of Tl-trapped electrons and recombined electron-hole pairs as a function of Tl concentration for an incident γ-ray energy of 2 keV.



[Wang et al., 2012]

- holes are assumed to become self-trapped immediately (**T < 100K**)
- “Tl-trapped” electron doesn’t imply a hole will be captured (may not result in light emission)

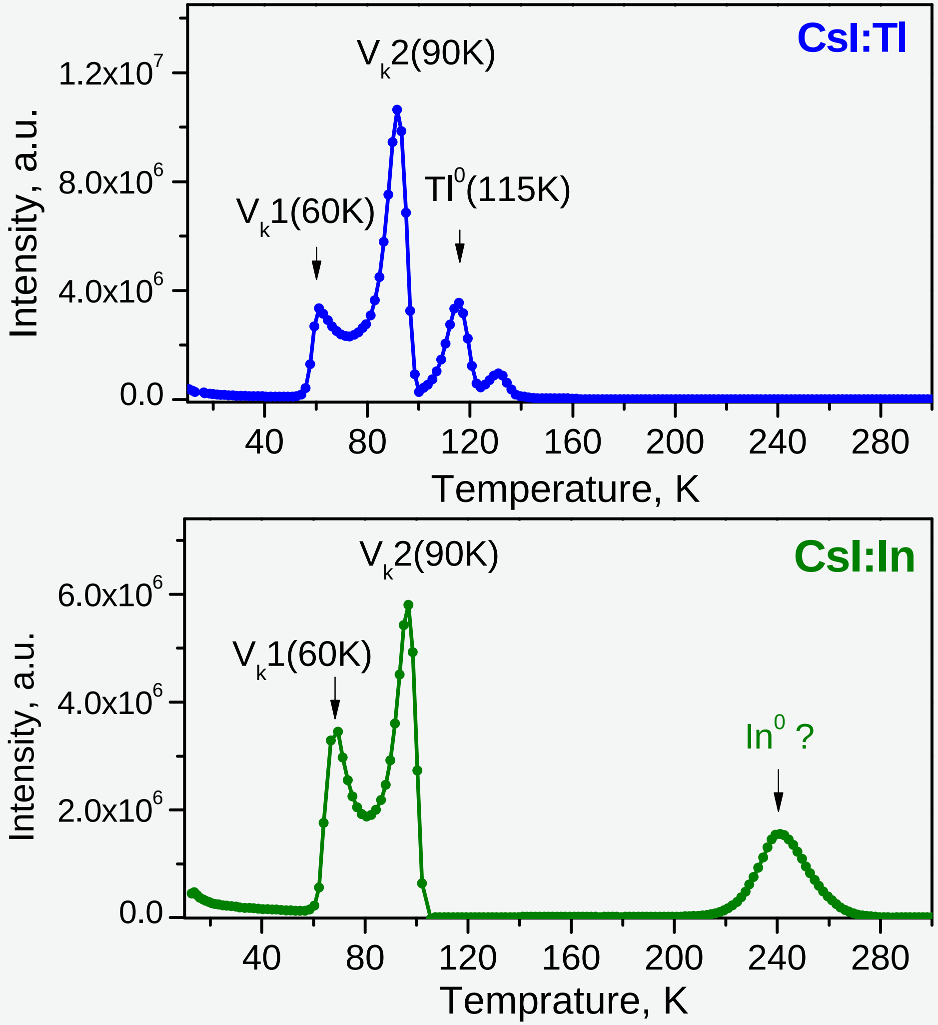
Temperature stability of STE emission



Not only does the intensity of STE emission decrease, but also the thermal stability

Quenching starts at lower temperature with the increase of activator concentration

Electron and hole traps in CsI:X



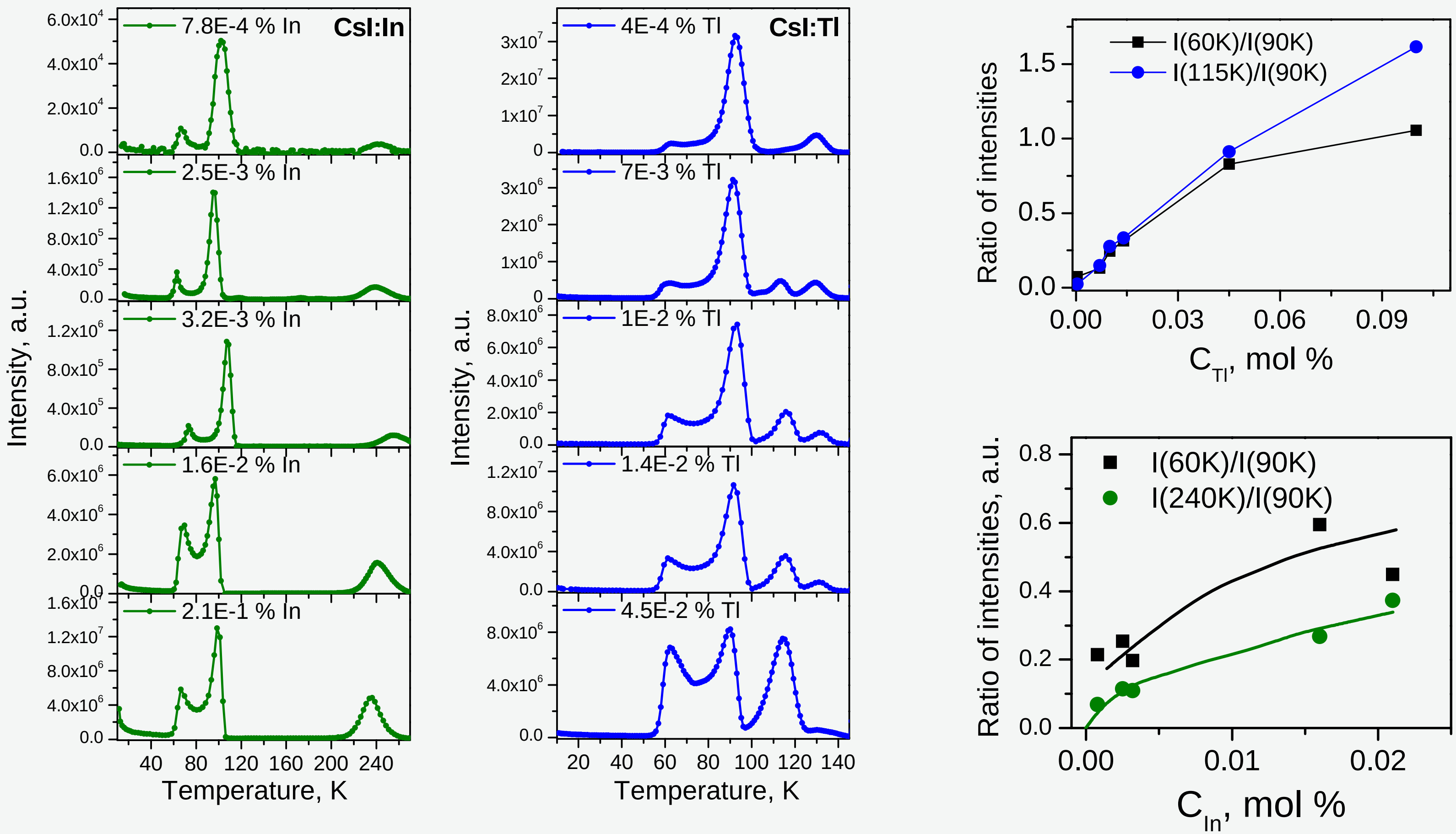
V_k1 (60K) – jump diffusion of holes
V_k2 (90K) – delocalization of holes
Tl^0 (115K) – e^- delocalization

[P. Martinez et al., 1964]
[V. Babin, K. Kalder, A. Krasnikov, S. Zazubovich, 2002]

Can the 240K peak in CsI:In be attributed to In^0 ?

- very low mobility of holes at low temperatures in CsI
- h^+ transport to activator centers is limited at T<100K

Concentration dependence of TSL glow peaks



• two temperature-dependent peaks with similar behavior in both CsI:In and CsI:Tl

Summary

- luminescent properties of CsI:In scintillator were investigated in comparison with CsI:Tl in wide concentration and temperature range

- In^{+} may be a good activator for alkali-halide scintillators. Scintillation yield of CsI:In is close to CsI:Tl

- In both of investigated scintillators energy transfer to emission centers is realized by sequential capture of e^{-} and h^{+}

- Migration loss doesn't allow reaching the maximum yield

Possible ways to improve the efficiency

Localization of charge carriers

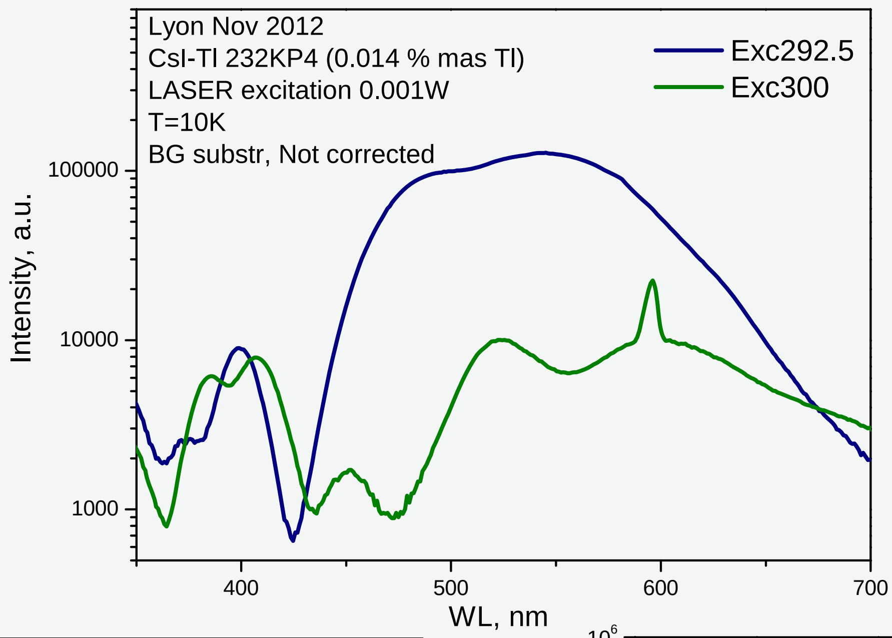
Increase of thermalization rate
(density of electron states in CB)

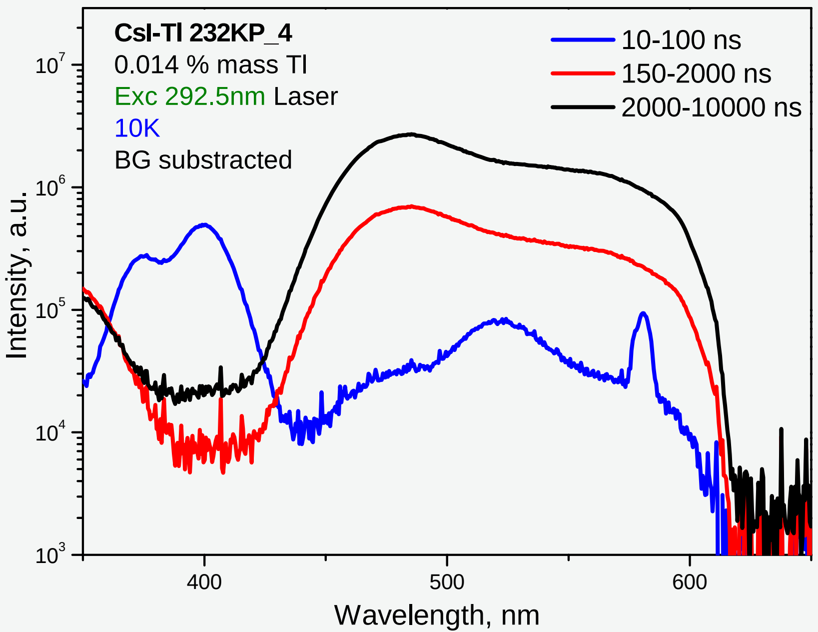
CsI:In
CsIBr:Tl
CsIBr:In

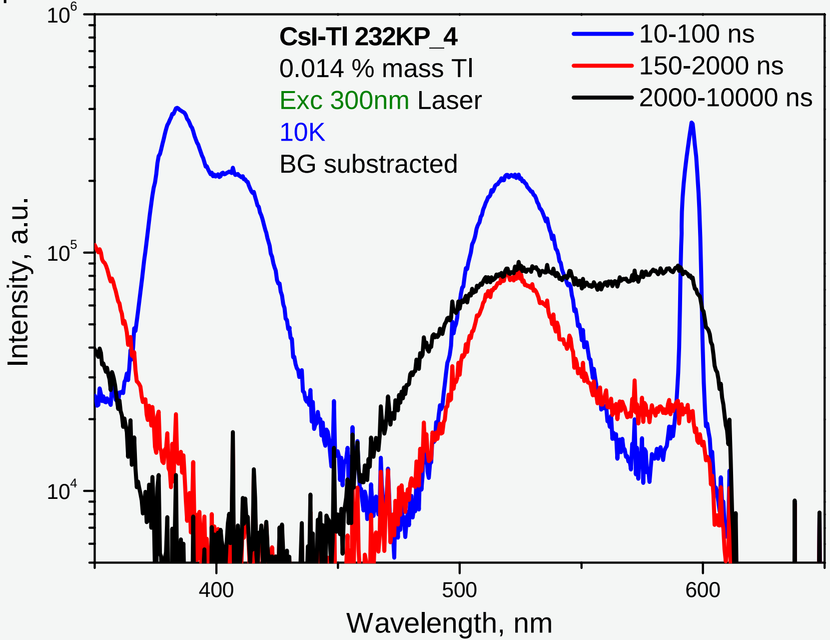
use of co-dopants and co-activators

$CsBa_2I_5$:Tl/In

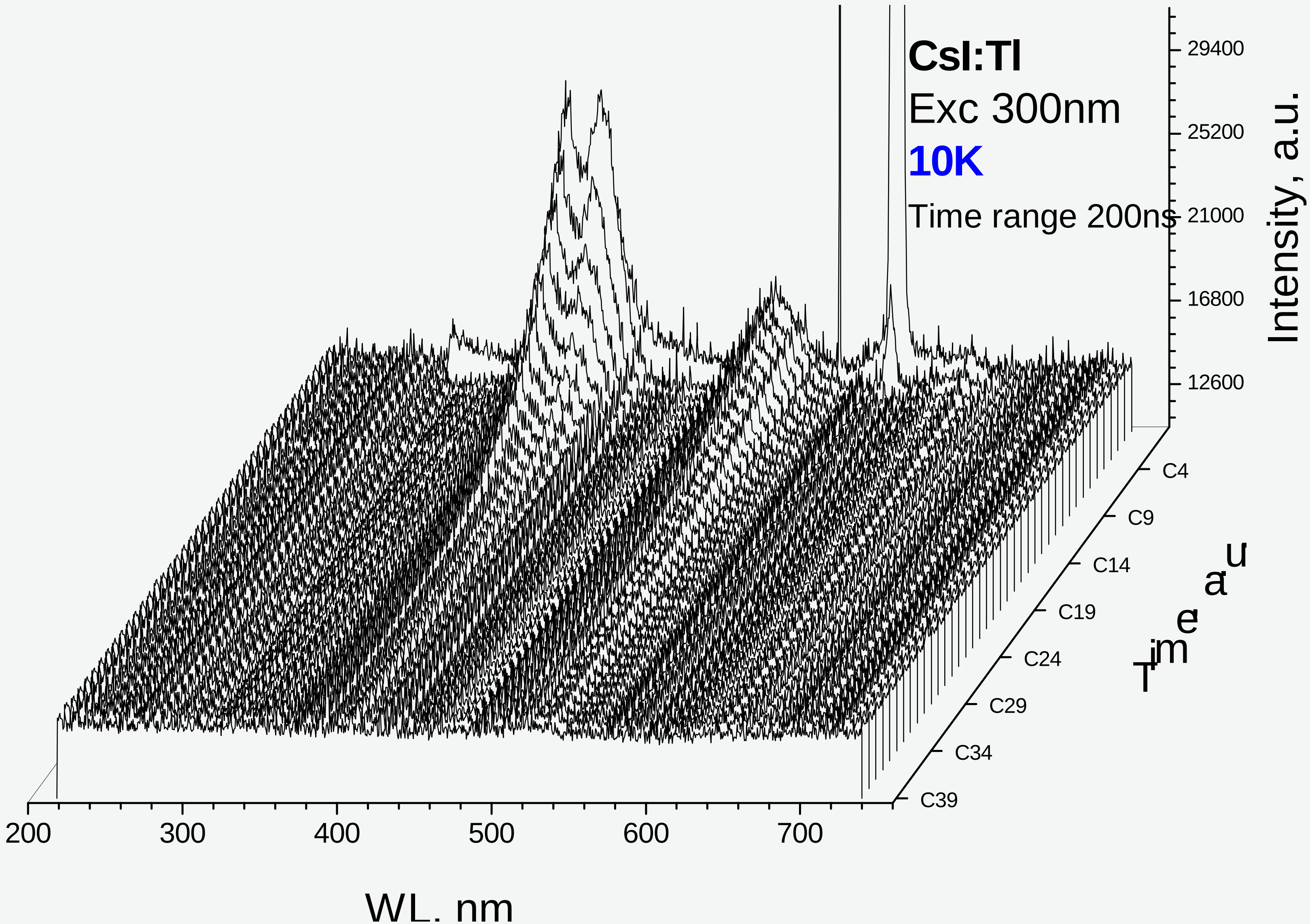
Time resolved spectroscopy
Complex band structure of **CsI:Tl**



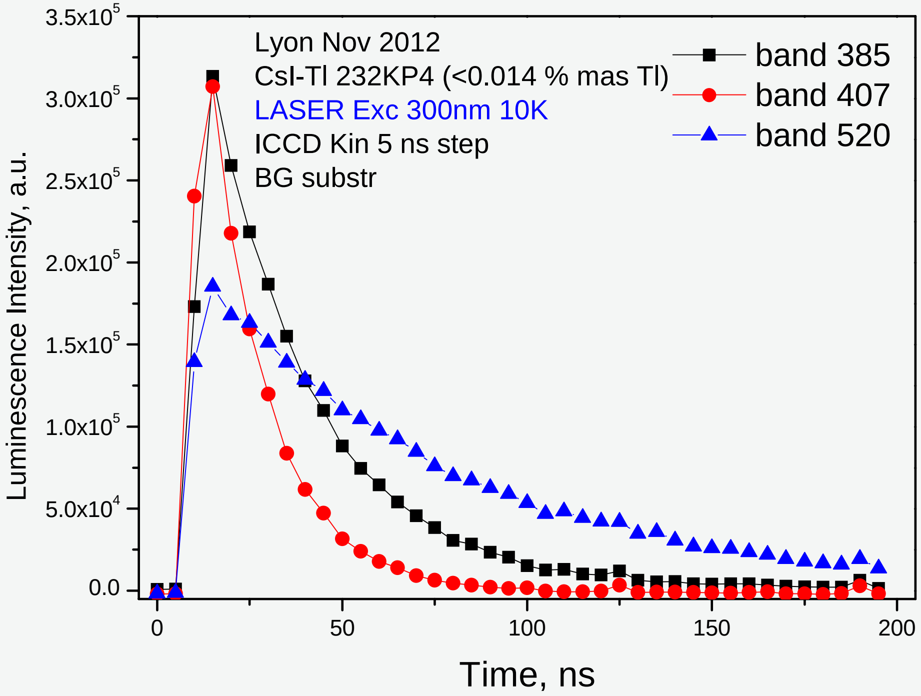


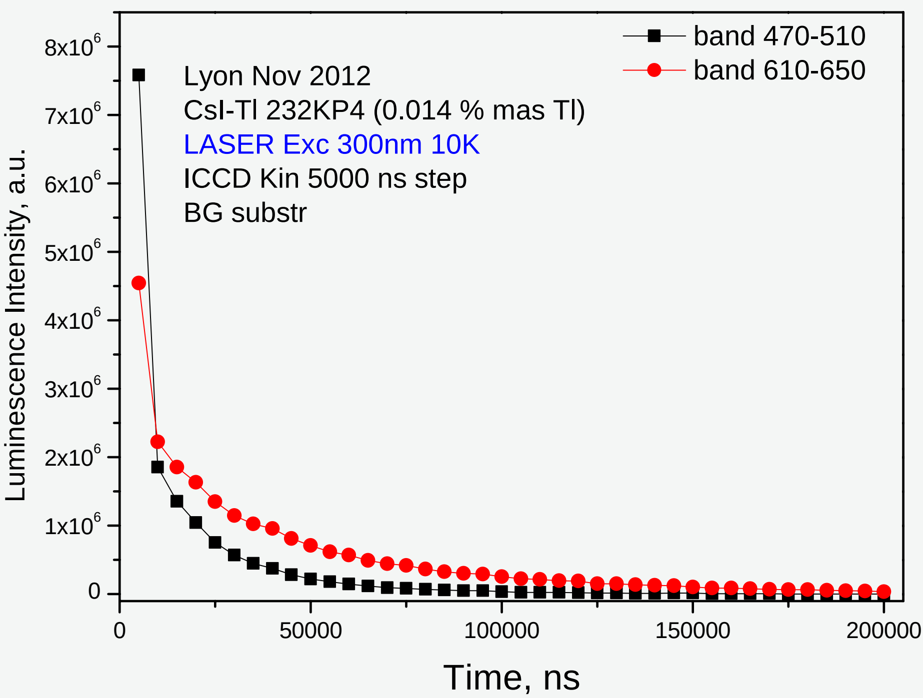


Time resolved spectroscopy
Complex band structure of **CsI:Tl**



Decay times





The fast component of luminescence in CsI:Tl in the visible region is due to a different band peaking at 520 nm

Thank you for attention