SCINT 2013
April 15-19, 2013
Shanghai, China

Energy relaxation channels in Tl- and In-doped CsI crystals

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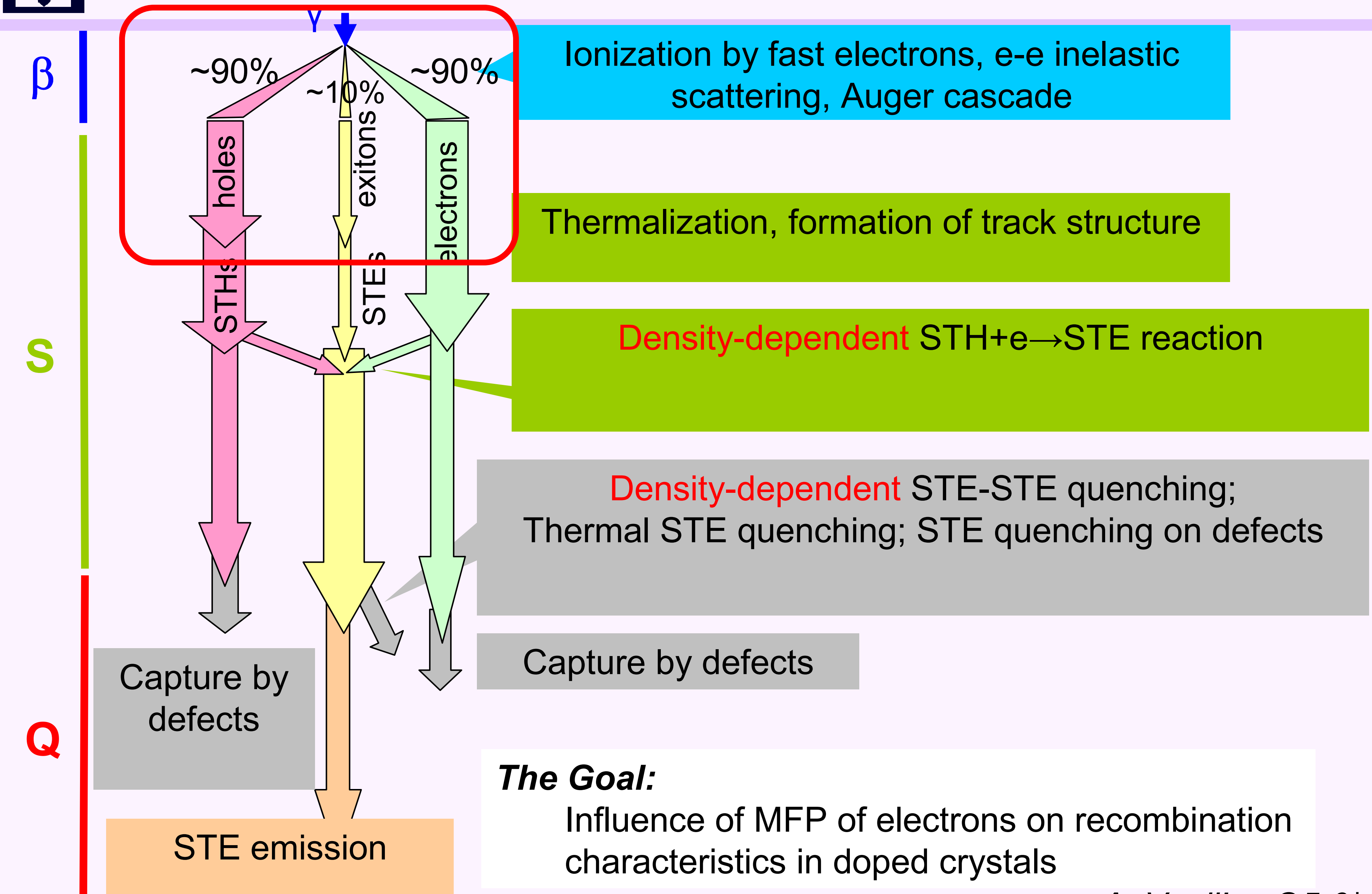
Motivation



The Goal:
Influence of MFP of electrons on recombination characteristics in doped crystals

*A. Vasil'ev O5.6**

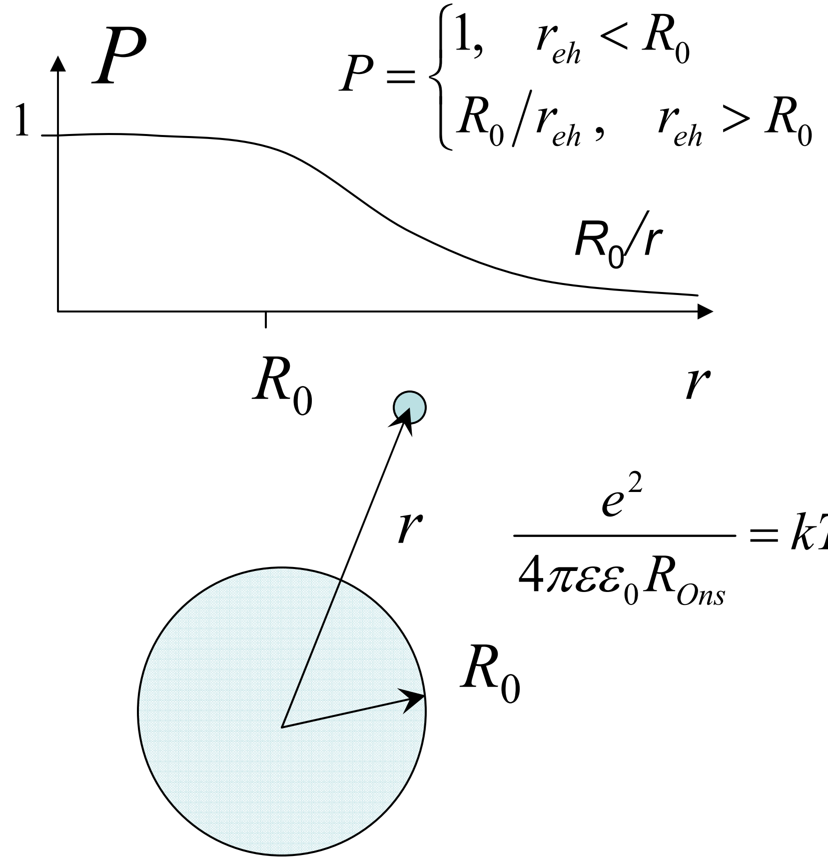
Outline

- **Theoretical approach**
  - MFP of electrons
  - *genetic* and *stochastic* recombination
  - influence of activator
- **Experiment**
  - CsI:X – modeling material
  - *excitonic* and *activator* relaxation channels
  - temperature and concentration dependences of yield
- **Experiment vs Theory**
  - comparison with modeling results

Path of electrons. Idea of Onsager radius.
Genetic and stochastic recombination

Recombination probability

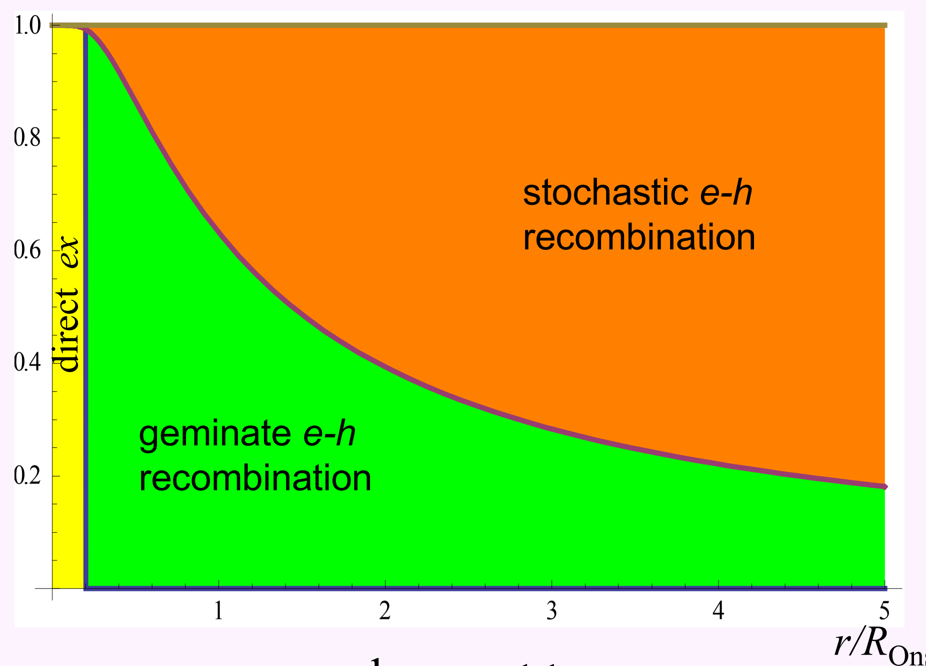
$$P = \begin{cases} 1, & r_{eh} < R_0 \\ R_0/r_{eh}, & r_{eh} > R_0 \end{cases}$$



$$\frac{e^2}{4\pi\varepsilon\varepsilon_0 R_{Ons}} = kT$$

Probability of geminate recombination

$$1 - e^{-R_{Ons}/r}$$



low $n_A \ll n$

♦ *Distance **r** between e^- and h^+ by the end of thermalization stage (respective to R_0) is the key parameter determining future recombination or capture of e^-*

Onsager, L. Initial recombination of ions. Phys. Rev. 54 (8), 554–557, 1938

Influence of e⁻ free path on recombination in presence of impurity A

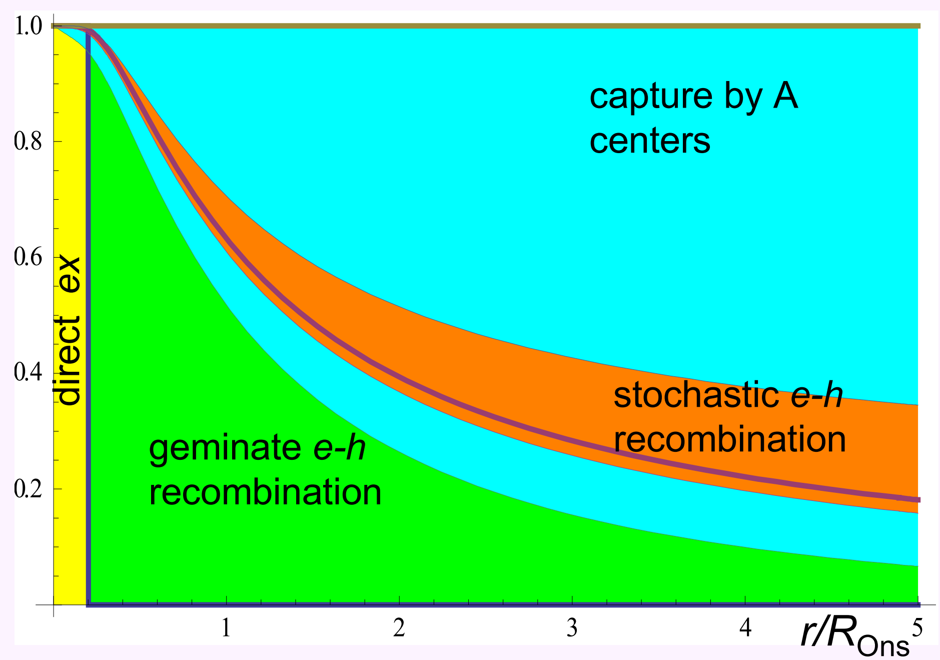
In presence of impurity (activator A)
apart recombination there is capture by centers A

Decrease of genetic recombination fraction [1]

$$\exp\left(-r\sqrt{4\pi R_{e+A} n_A}\right)$$

stochastic recombination : capture by A centers

$$4\pi R_{Ons}\left(D_e + D_h\right)n_h : 4\pi R_{e+A} D_e n_A$$



high $n_A >> n$

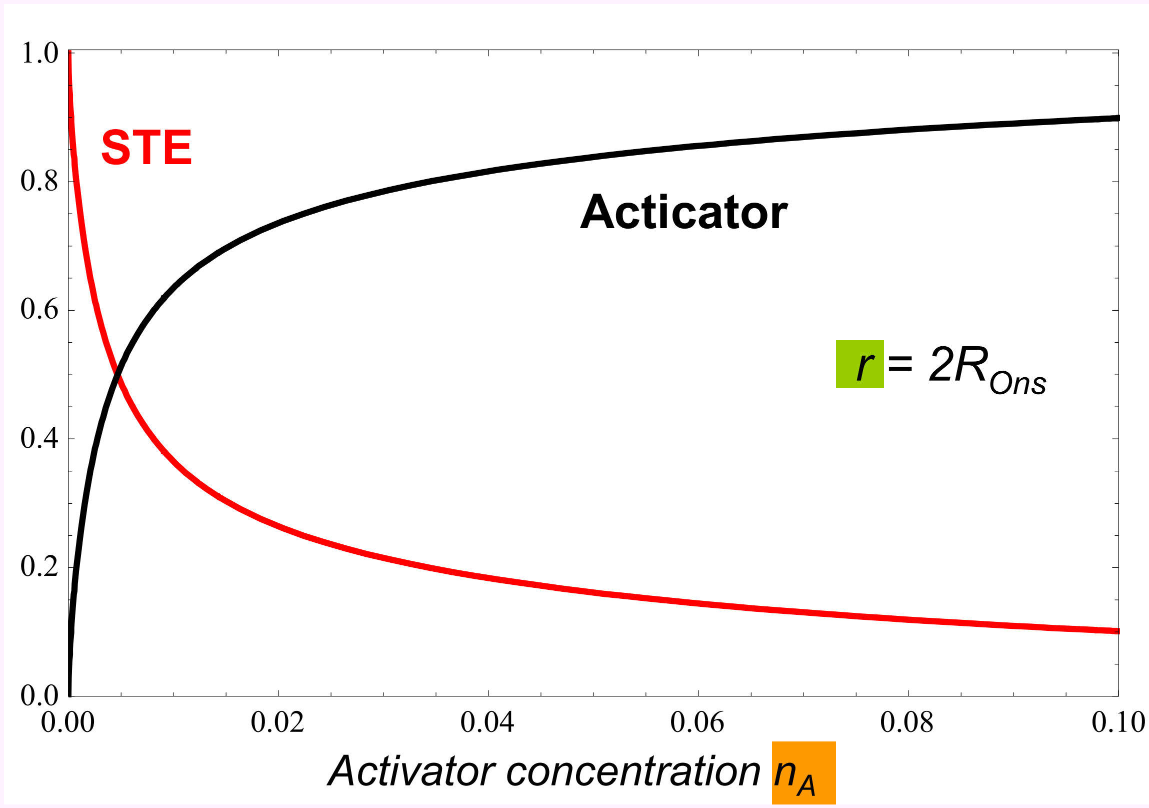
- Impurity centers capture excitations mainly form the stochastic part

[1]Vasil'ev A.N., Mikhailin V.V., Ovchinnikova I.V. Bulletin AS USSR. Phys. Ser., v. 49, No. 10, pp. 2044, 1985

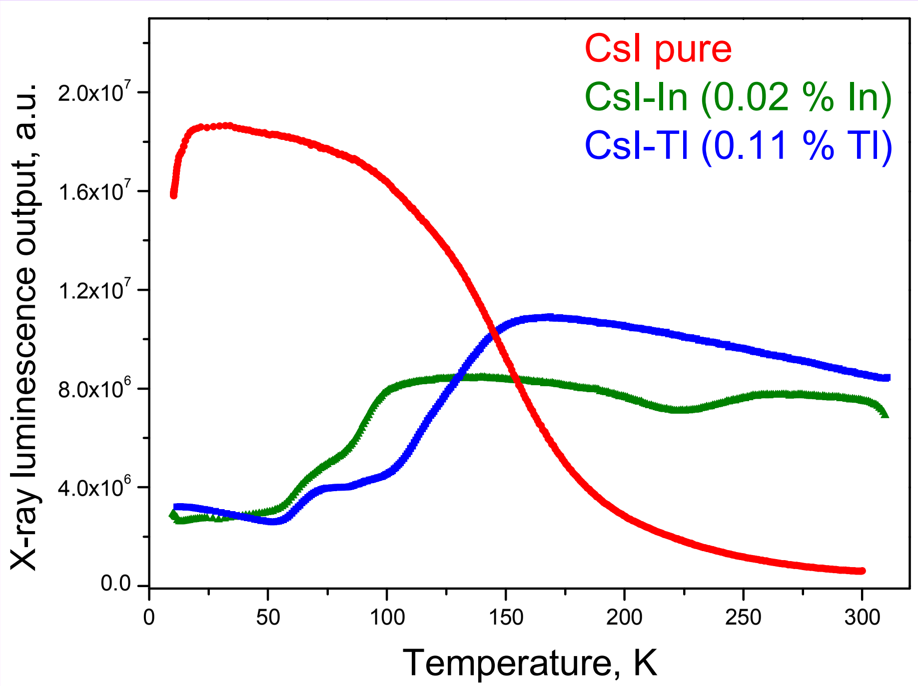
Concentration dependence of the fration of **recombination** and **activator** channels of relaxation

Fraction of **excitonic** channel of relaxation can by estimated as

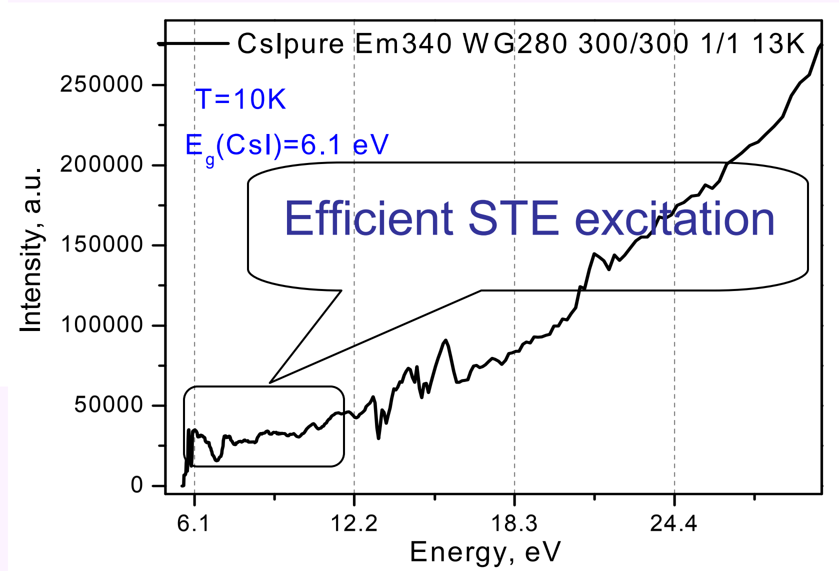
$$\left(1-e^{-R_{Ons}/r}\right)\exp\left(-r\sqrt{4\pi R_{e+A} n_A}\right) + \left(1-\left(1-e^{-R_{Ons}/r}\right)\exp\left(-r\sqrt{4\pi R_{e+A} n_A}\right)\right)\frac{4\pi R_{Ons}\left(D_e+D_h\right)n_h}{4\pi R_{Ons}\left(D_e+D_h\right)n_h+4\pi R_{e+A} D_e n_A}$$



CsI based scintillators as modeling material



Scintillator	LY_{Exp}, ph/MeV	<E>/photon, E_g
CsI **77K**	**107,000**[1]	1.5
CsI:Tl 300K	**60,000**[2]	2.9

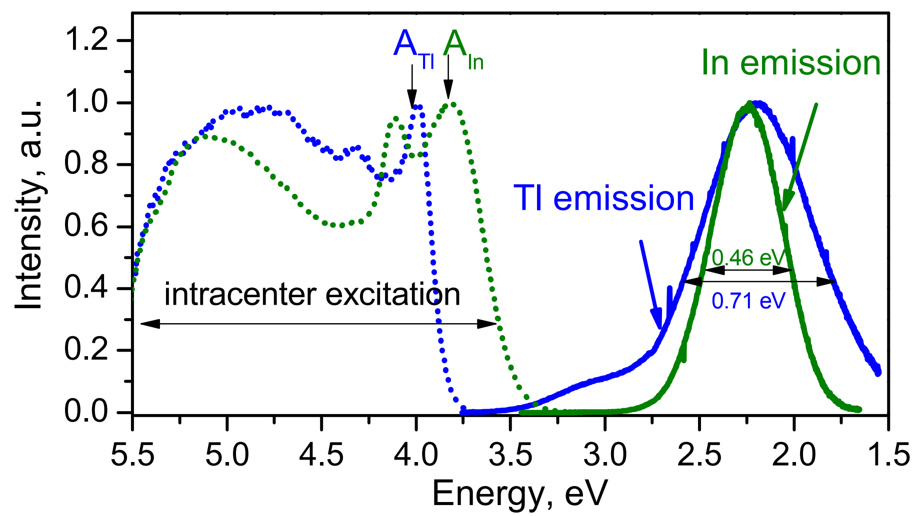


- Two channels of radiative relaxation
- High yield of STE emission in pure CsI
- *CsI:Tl* at 300K – half of *CsI pure* at 77K

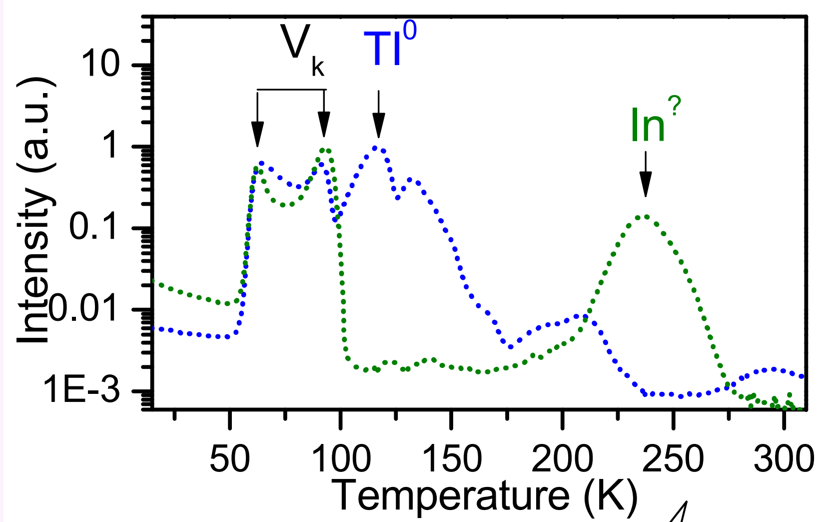
[1] Moszynski et al. Application of large area avalanche photodiodes to study scintillators. NIM 504 (2003) 307
[2] Moszynski et al. Absolute Light Output of Scintillators. IEEE TNS 44 (1997) 1052

Luminescent properties of CsI:In и CsI:Tl

Excitation and luminescence spectra of CsI:Tl и CsI:In at 300K



Glow curves of CsI:Tl and CsI:In after X-irradiation



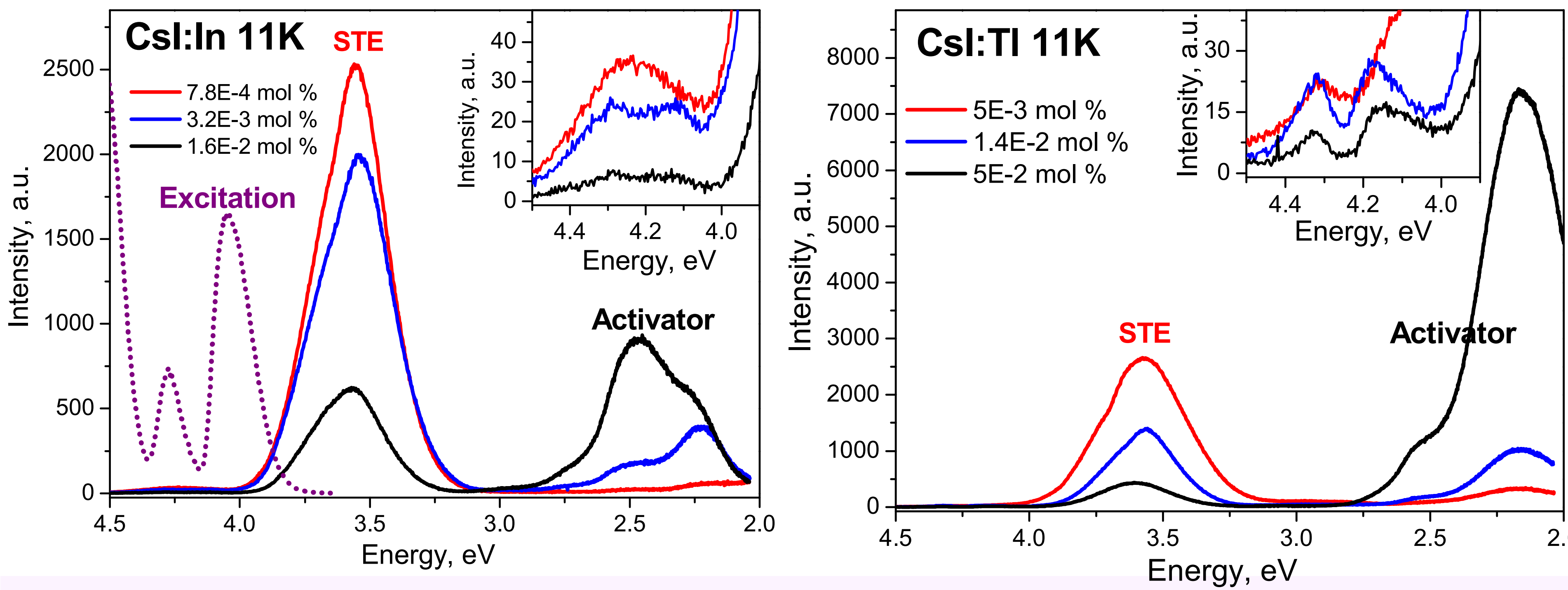
Dopant	Ionic radius, Å	Segregation coefficient	*A* absorption band, nm	Emission max. at RT, nm	Decay, ns
Tl+	1.59	0.2-0.3	299	550-560	620
In+	~1,35	~0.15	310	545	1900

Similar luminescent properties

Different parameters of charge carriers capture

Excitonic and activator channels of radiative relaxation in CsI:X

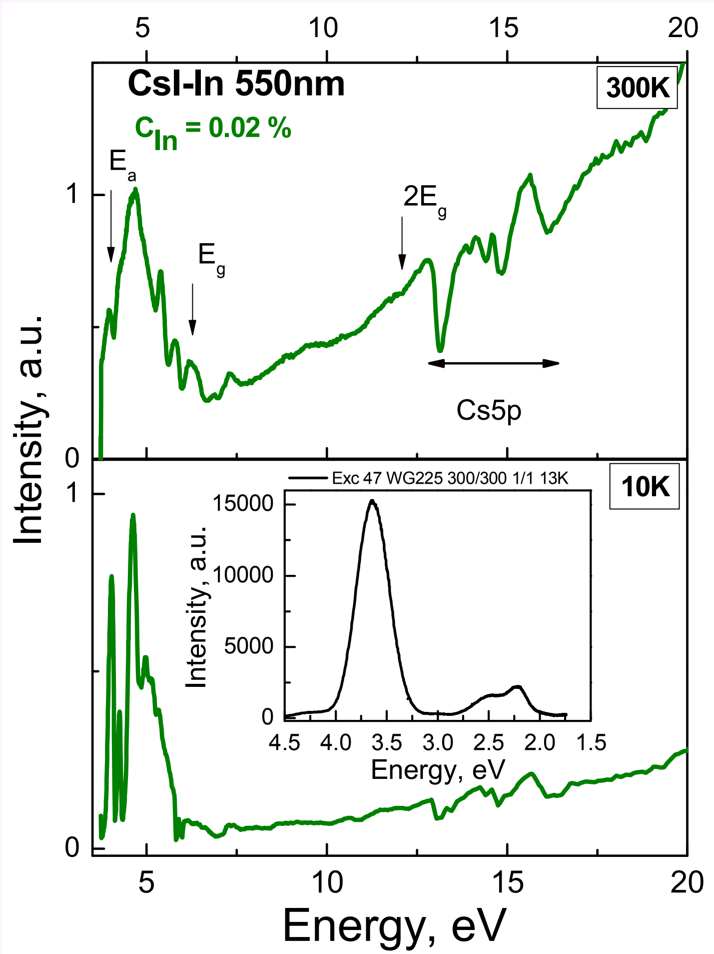
X-ray luminescence spectra of CsI:In и CsI:Tl at LHeT

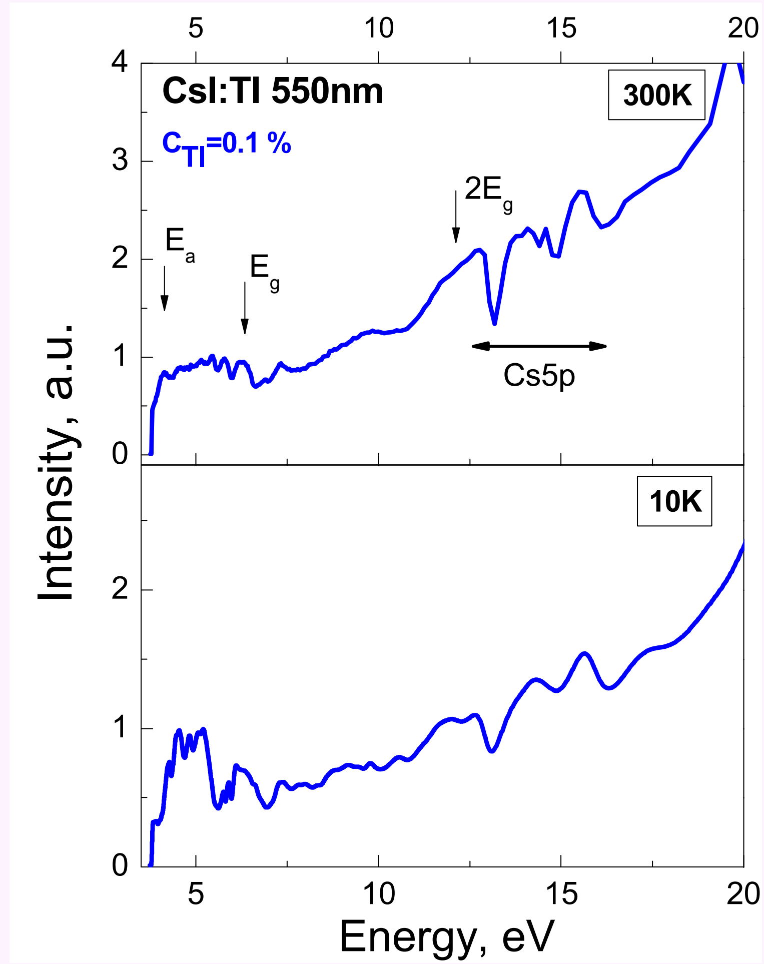


- *No overlap between excitonic and activator bands*
- *Insignificant overlap of STE band with activator absorption*

Energy transfer to activator centers

Excitation spectra of **In-** and **Tl-**related emission in CsI:X

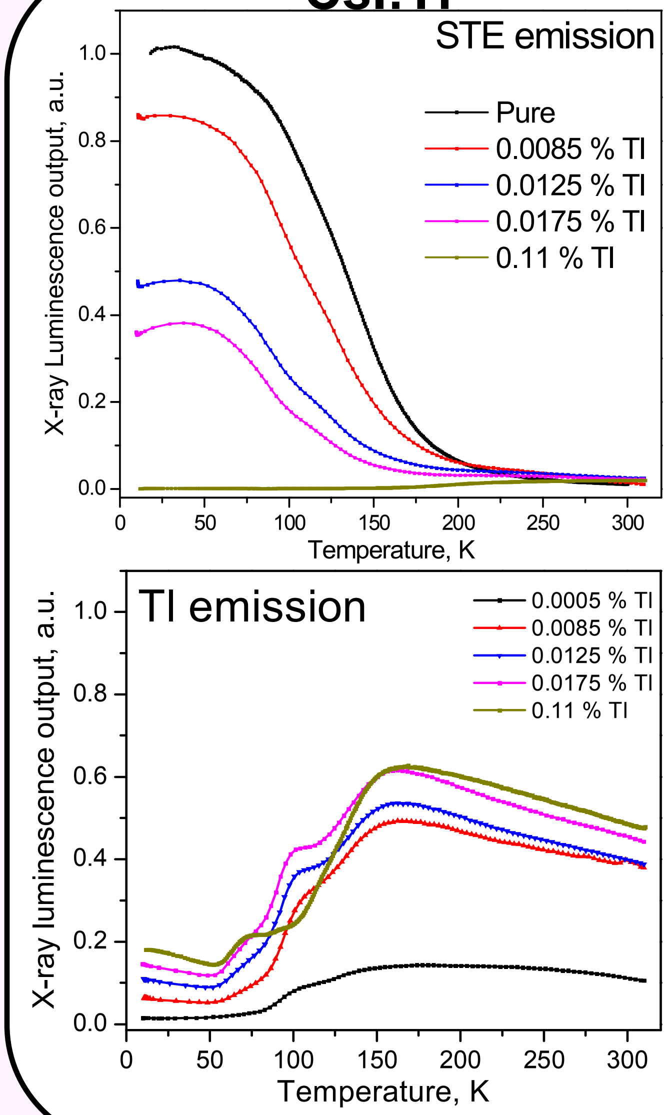




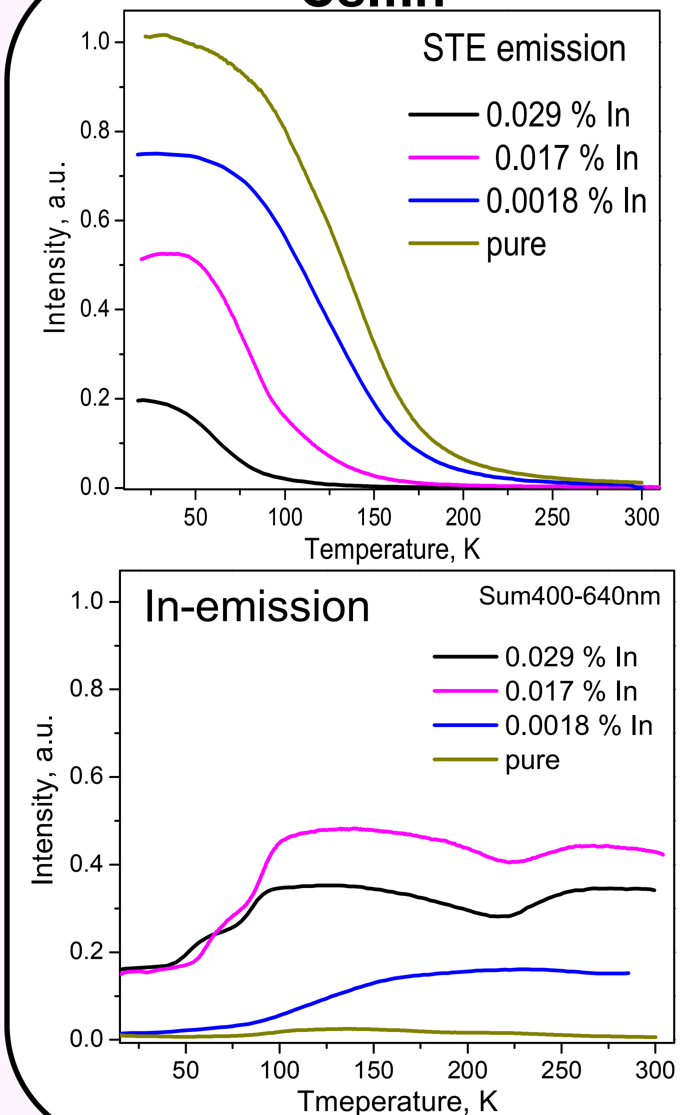
- *Recombination mechanism of e-h transfer*
- *Lower efficiency at 10K is due to STE*

Temperature dependences of X-ray luminescence yield for different activator concentrations

CsI:Tl

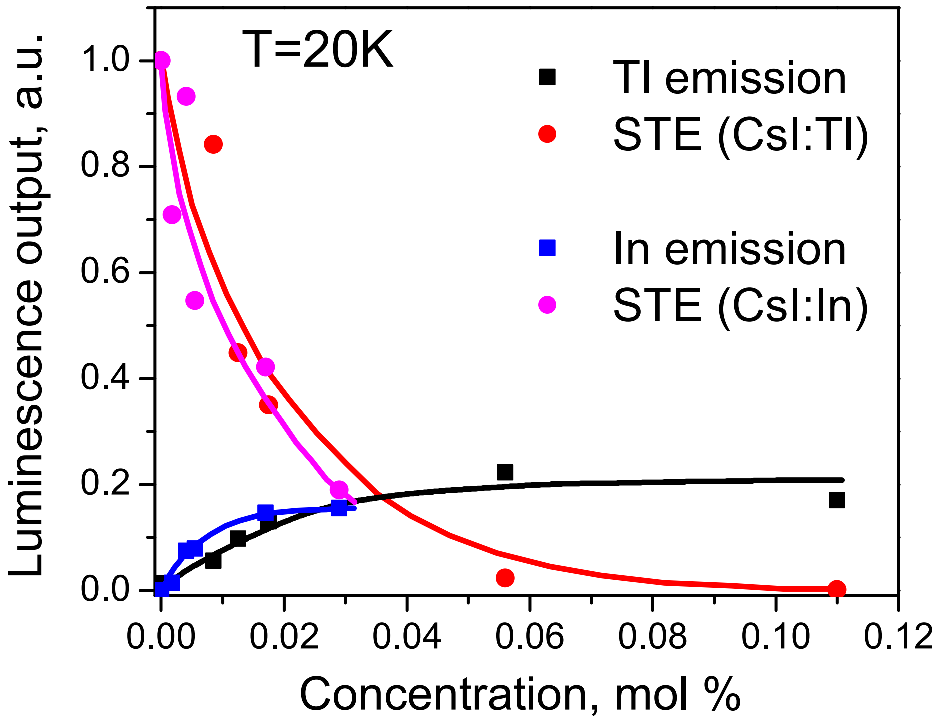


CsI:In



Concentration depencence of **STE** and **activator** luminescence yield. Experiment vs Theory

Experiment



Theory [1]

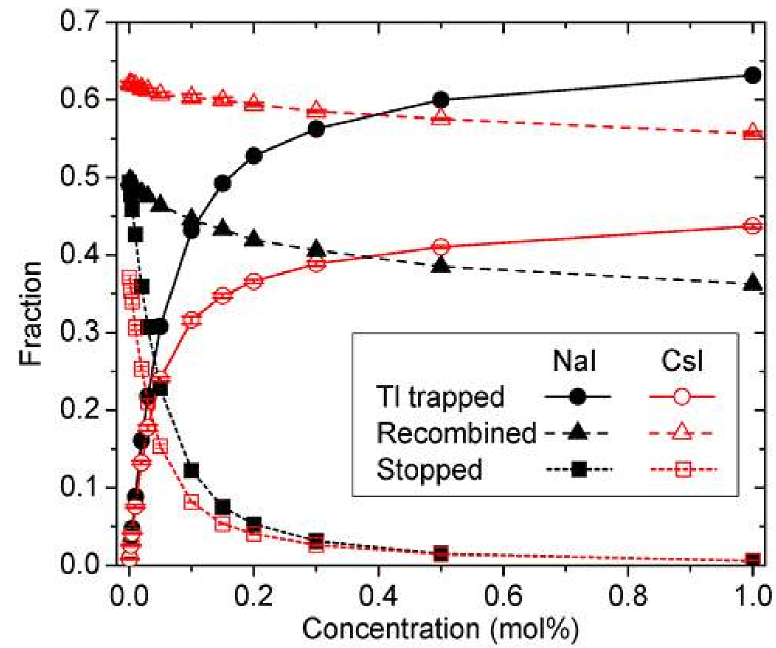
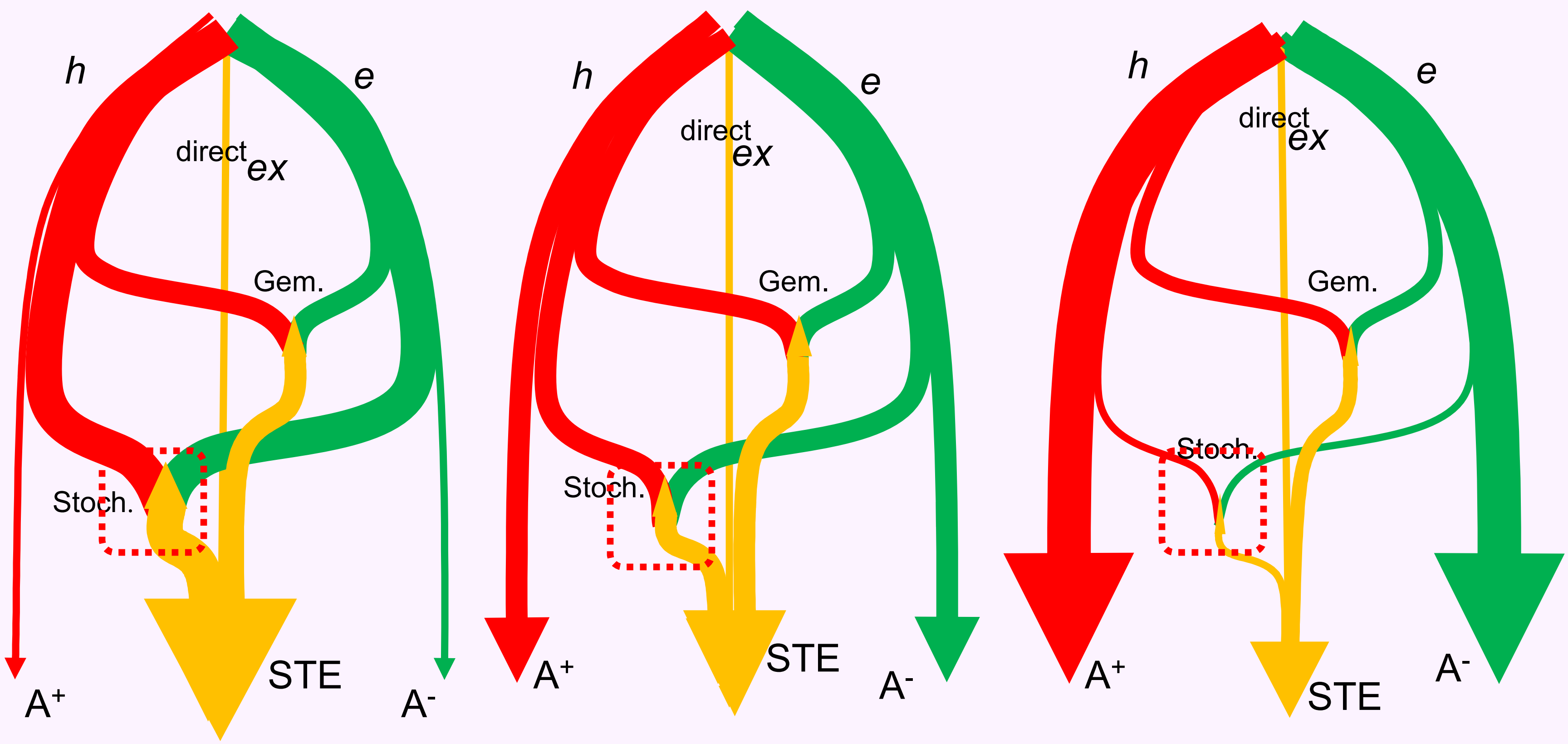


FIG. 6. Fraction of Tl-trapped electrons and recombined electron-hole pairs as a function of Tl concentration for an incident γ-ray energy of 2 keV.

- ***Energy loss, related to STE quenching***
  - ***Recombination channel is underestimated***
  - ***possible overestimation of Activator channel***
  - ***Energy loss mechanisms are not considered***

[1] Wang et al. *Monte Carlo simulations of electron thermalization in alkali iodide and alkaline-earth fluoride scintillators*. J. App. Phys. 112, 2012

Excitonic and activator channels on n_A



low $n_A \ll n$ medium $n_A \sim n$ high $n_A \gg n$

Increase of Activator channel with the increase of activator content

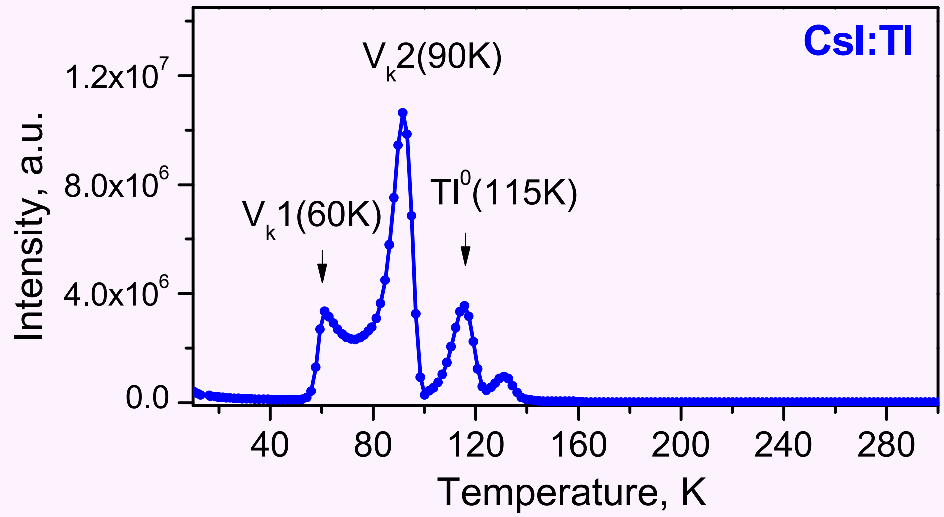
Activator channel is mostly "fed" by *stochastic* part

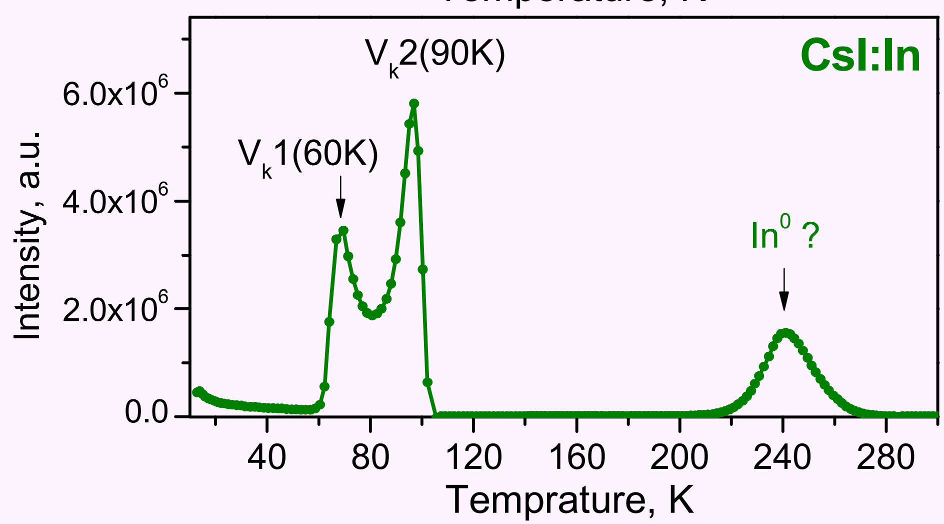
Summary

- Similar behavior of CsI:In and CsI:Tl regardless the differences in charge capture peculiarities
- STE quenching connected with the activator
- Need to take into account energy loss mechanisms

Thank you for attention

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]

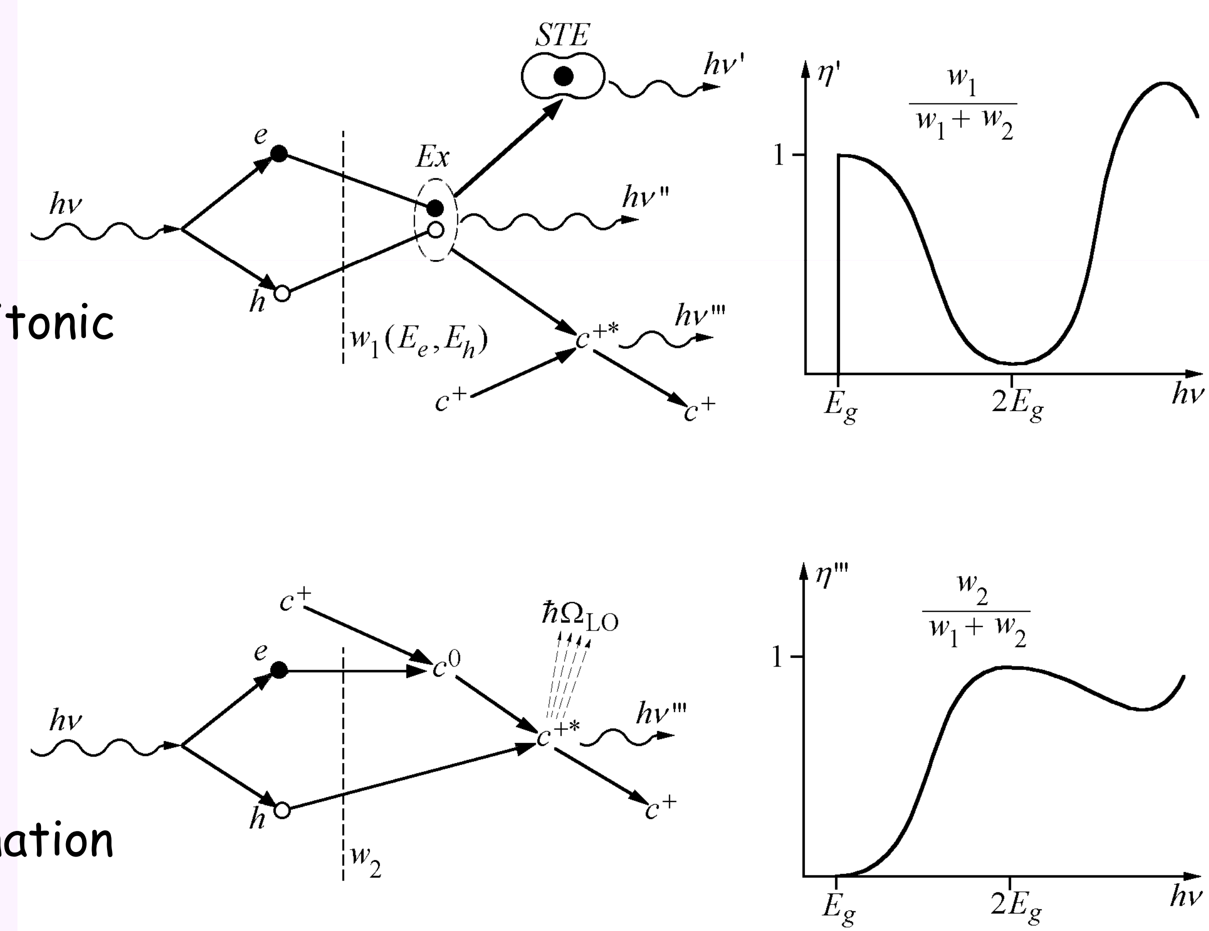
Can the 240K peak in CsI:In be attributed to In^0 ?

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

Appendix A. Energy transfer

excitonic

recombination



Typical energy dependence of the quantum luminescence yield of these channels

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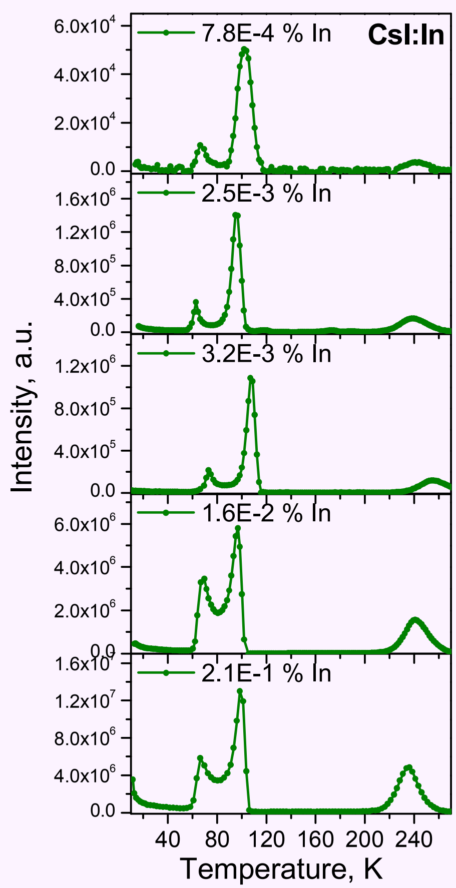
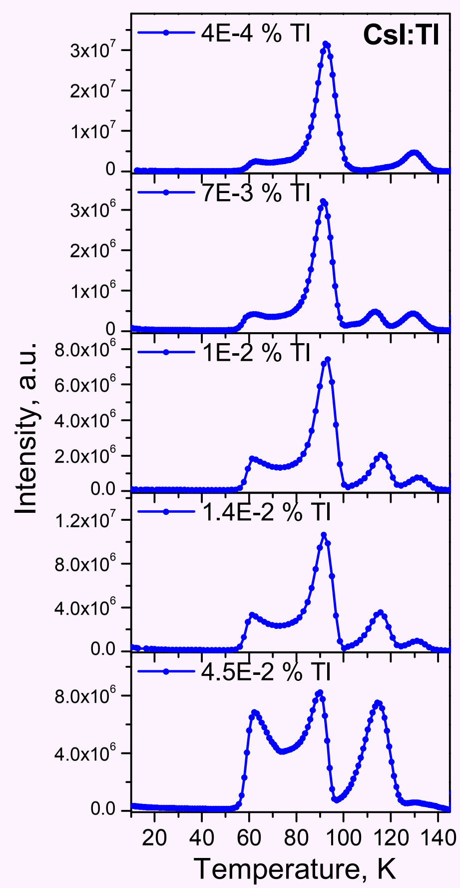
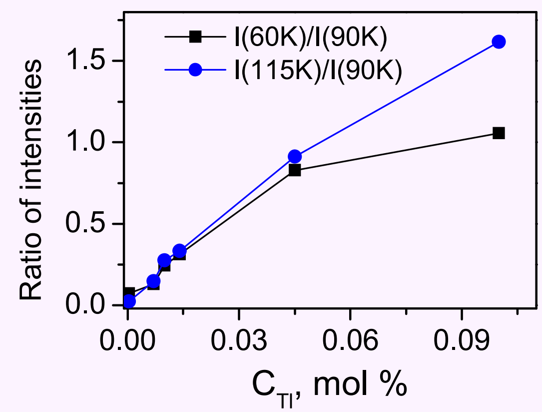
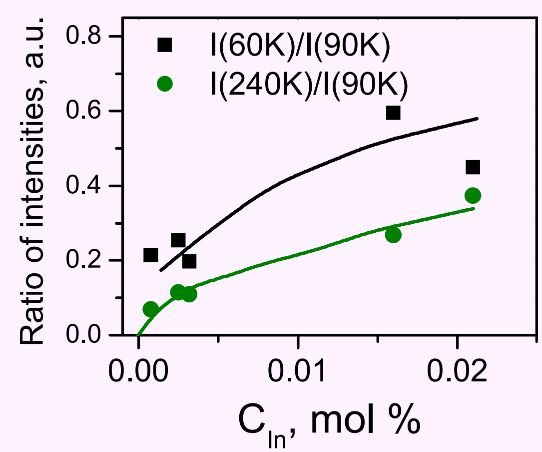
Appendix B. Concentration dependence of glow peaks intensity









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