## Scintillators for new HEP calorimeters: what properties are not well understood?

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## Outline

- Intrinsic energy resolution, scintillator non-proportionality and dependence on the particle type: track structure
- Linear and non-linear processes in energy transfer in scintillators
- Linear energy losses ionization/excitation and Cherenkov
- Cascade of strongly inelastic collisions and thermalization
- Different types of electron-phonon interaction, mobility in crystals and glasses; trapping in glasses
- Role of Coulomb interactions in recombinations in the track region
- Exciton creation due to hot recombination
- Recombination in clusters of excitations: triplets and effects due to electric fields
- Granularity, absorption and energy resolution



Scheme of relaxation of electronic excitations in crystals with "simple" energy structure



Studies and applications of new scintillating material

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#### Light Output per keV of Electron Energy for Several Scintillators

From W. Mengesha, T. Taulbee, B. Rooney and J. Valentine, "Light yield nonproportionality of CsI(Tl), CsI(Na), and YAP," *IEEE Trans Nucl Sci* 45, pp. 456-461, 1998.



#### Ideal Scintillator Would Be Horizontal Line

are not well understood?

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# What are the emission centers in scintillators?

## A. Neutral excitations:

- Excited state of an activator (rare earth: Ce, Pr, Eu,..., d-element: Tl, Bi, ...)
- Excitons (CdWO<sub>4</sub>, BaF<sub>2</sub> slow, ...)
- Defects (PbWO<sub>4</sub>, ...)
- Complex centers (pure CsI ?)
- B. Charged excitations:
- Interband transitions (BaF<sub>2</sub> fast, pure CsI ?)



# What is the last but one stage in energy transfer (case A)?

- Exciton or Excitonic transfer to the activator (rate is linear function on excitation density)
- Sequential recombination of an electron and a hole pair at activator or recombination with production of the exciton (rate is quadratic or bilinear function on excitation dencity)



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#### ENERGY CONVERSION

Scintillation efficiency (A. Lempicki, A.J. Wojtowicz, E. Berman, NIM A 333 (1993) 304):

η = β*SQ* 

where  $\beta$  is the conversion efficiency of absorbed energy into energy of e-h pairs and excitons, S < 1 is the transfer efficiency of e-h pair and exciton energies to the luminescence centre, and Q < 1 is the quantum yield of luminescence center

$$I = N_{eh} S Q$$
  

$$N_{eh} = \beta E / E_g = E / E_{eh}$$
  

$$E_{eh} = (2 \div 4) E_g$$



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Ionization by fast electrons, e-e inelastic scattering, Auger cascade

S

 $\eta$  $E_{eh}$ 



Ionization by fast electrons, e-e inelastic scattering, Auger cascade

Thermalization, formation of track structure

η







Low density



.



Low density

Medium density













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## Modification of kinetics in scintillators





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#### Interaction of excitations in the regions with high excitation concentration

$$I_{\textit{lum}}(t) \equiv I_{\textit{lum}}(t, n_0(\mathbf{r}))$$

#### STE+STE $\rightarrow$ STE (e.g. CdWO<sub>4</sub>):





Dipole-dipole transfer:

$$I_{lum}(t, n_0(\mathbf{r})) = \frac{n_0(\mathbf{r})}{\tau_r} \frac{\exp(-t/\tau_r)}{1 + \frac{2\pi^2}{3} n_0(\mathbf{r}) R_{d-d}^3} \operatorname{erf}\left(\sqrt{t/\tau_r}\right)$$



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#### Interaction of excitations in the regions with high excitation concentration

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#### STE+STE $\rightarrow$ STE (e.g. CdWO<sub>4</sub>):





Dipole-dipole transfer:

$$U_{lum}(t, n_0(\mathbf{r})) = \frac{n_0(\mathbf{r})}{\tau_r} \frac{\exp(-t/\tau_r)}{1 + \frac{2\pi^2}{3} n_0(\mathbf{r}) R_{d-d}^3} \operatorname{erf}\left(\sqrt{t/\tau_r}\right)$$

$$R_{t} = 2.1 \text{ nm}$$

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M. Kirm et al, PRB 79, 233103 (2009)

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### Two equation model with quenching: analytically solvable case

$$\frac{\partial n_{ex}(t)}{\partial t} = -a_{ex} n_{ex}(t) - b_{ex-ex} n_{ex}^2(t) + g_{eh} n_e(t) n_h(t)$$

Excitons and geminate electron-hole pairs

$$\frac{\partial n_e(t)}{\partial t} = \frac{e + h \rightarrow 0}{-(b_{eh} + g_{eh})n_e(t)n_h(t)}$$
$$n_e(t) = n_h(t)$$

NB! All Auger type processes are neglected!

Non-geminate electrons and holes

Initial conditions

$$n_{ex}(0) = n^0 f_{ex}$$
$$n_h(0) = n_e(0) = n^0 (1 - f_{ex})$$

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## Two equation model with quenching: analytically solvable case

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Excitons and geminate electron-hole pairs

$$\frac{\partial n_{e}(t)}{\partial t} = -(b_{eh} + g_{eh})n_{e}(t)n_{h}(t)$$
$$n_{e}(t) = n_{h}(t)$$

NB! All Auger type processes are neglected!

G. Bizarri, W.W. Moses, J. Singh, A.N. Vasil'ev, and R.T. Williams, J. Lumin., 129 (2009) 1790–1793





WAKE FOREST

Non-geminate electrons and holes

Initial conditions

$$n_{ex}(0) = n^0 f_{ex}$$
$$n_h(0) = n_e(0) = n^0 (1 - f_{ex})$$

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#### Yield as a function of the excitation density (uniform distribution of excitations): sensitivity to the shaping time T



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#### Yield as a function of the excitation density (uniform distribution of excitations): sensitivity to the shaping time T



 $a_{ex}=1, b_{ex-ex}=1, b_{eh}=0.5, g_{eh}=0.5$ 

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## General: The structure of energy losses by fast particle

From classical electrodynamics: the force acting on a charge *ze* which is moving with velocity *v* through the media with complex dielectric permittivity  $\epsilon(\omega, \mathbf{q})$  equals to

$$F = \frac{1}{(2\pi)^{3}} \bigvee_{-\infty}^{\infty} d\omega \int_{0}^{\infty} q^{2} dq \int_{-1}^{1} dx \frac{8\pi^{2} e^{2} z^{2} i}{\omega} \left\{ \frac{\omega^{2}}{q^{2} \varepsilon(\omega, \mathbf{q})} + \frac{q^{2} v^{2} - \omega^{2}}{q^{2} \left[ \varepsilon(\omega, \mathbf{q}) - c^{2} q^{2} / \omega^{2} \right]} \right\} \delta(\omega - qvx)$$
Longitudinal (Coulomb)
field – pure ionization
Transversal field –
Cherenkov + ionization + X

Energy losses of the moving charge:

$$-\frac{dE}{dx} = \frac{2e^2z^2}{\pi v^2} \int_{0}^{E/\hbar} \omega \, d\omega \int_{Q_{\min}}^{Q_{\max}} \frac{dQ}{Q} \operatorname{Im} \left[ -\frac{1}{\varepsilon(\omega, Q)} - \frac{v^2Q^2/\omega^2 - 1}{\varepsilon(\omega, Q) - c^2Q^2/\omega^2} \right]$$

$$Q_{\max}_{\min} = \frac{1}{\hbar c} \left( \sqrt{E(E + 2mc^2)} \pm \sqrt{(E - \hbar \omega)(E - \hbar \omega + 2mc^2)} \right)$$



Cherenkovandionization
$$\hbar \omega < E_g$$
 $\hbar \omega > E_g$ 

 $E_g$  – forbidden gap of the scintillator (fundamental absorption edge)



$$=\frac{2e^{2}z^{2}}{\pi v^{2}}\int_{E_{g}/\hbar}^{E/\hbar}\omega d\omega \int_{Q_{\min}}^{Q_{\max}}\frac{dQ}{Q} \operatorname{Im}\left[-\frac{1}{\varepsilon(\omega,Q)}-\frac{v^{2}Q^{2}/\omega^{2}-1}{\varepsilon(\omega,Q)-c^{2}Q^{2}/\omega^{2}}\right]$$
$$+\frac{e^{2}z^{2}}{v^{2}}\int_{0}^{E_{g}/\hbar}\theta\left(v\sqrt{\varepsilon(\omega,0)}-c\right)\left(\frac{v^{2}}{c^{2}}-\frac{1}{\varepsilon(\omega,0)}\right)\omega d\omega$$



# Example: NaI energy loss function in GOS relativistic approximation using EPDL97 database



Energy loss function  $\operatorname{Im}(-\varepsilon^{-1}(\omega, q))$ multiplied by the energy losses  $\hbar\omega$ for different values of  $q(E_q = \hbar^2 q^2/2m)$ .

• Low-angle scattering  $(E_q << E)$ 

Large-angle scattering (E<sub>q</sub>~E) (δ-electron production) due to Bethe ridge (Mott scattering)
– production of electron-hole pair with large electron momentum,

$$\hbar\omega \approx E_q = \hbar^2 q^2 / 2m$$

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#### Stopping power for NaI in GOS relativistic approximation





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#### Excitation density along the track



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#### Excitation density along the track







# Vield as a function of the excitation density (Gaussian distribution of excitations over the cross-section)



## BaF<sub>2</sub> Crossluminescence

- R. Novotny, Proc. Int. Conf. On Inorganic Scintillators and Their Applications, SCINT95, Delft University Press, The Netherlands, 1996, pp. 70-73.
- R. Novotny, et al., IEEE Trans. Nucl. Sci. NS-43 (1996) 1260.



Fig. 1. Scatter plot of the fast scintillation component versus the total light output of a  $BaF_2$ -detector. The correlation pattern is shown for the neutral and the charged events.

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# BaF<sub>2</sub> particle discrimination



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# BaF<sub>2</sub> crossluminescence excitation spectrum (simulation and experiment, SCINT2005)



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## Electron Energy Deposit is Non-Uniform! (W.W.Moses, Scintillator Non-Proportionality, SCINT-2007)



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#### NaI energy loss function in GOS approximation using EPDL97 database relativistic



 $\operatorname{Im}(-\varepsilon^{-1}(\omega, q))$ Energy loss function multiplied by the energy losses  $\hbar\omega$ for different values of  $q(E_a = \hbar^2 q^2/2m)$ .

Low-angle scattering ( $E_q << E$ )

Large-angle scattering  $(E_{q} \sim E)$ ( $\delta$ -electron production) due to Bethe ridge (Rutherford scattering) - production of electron-hole pair with large electron momentum,

$$\hbar\omega \approx E_q = \hbar^2 q^2 / 2m$$





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### Track spatial and temporal structure

The sizes of the excited clusters of excitations just after initial recombination and thermalization of electrons and holes (ps time domain) depend on the thermalization length, which is determined by electron-phonon interaction. For ionic crystals it can be estimated to be about 2-4 nm, for piezoelectric crystals it can be even shorter, and for covalent crystals longer.

Initial electron track structure in Nal is defined by isolated and overlapped clusters of excitations with initial size of about 3 nm. Distances between clusters are about 1 to 100 nm. The shown track is straitened along the path of the primary electron.



## Why the exciton yield is so high?

The simulation of Ce-activated and intrinsic scintillators shows that yield can be as high as 80%

R<sub>track</sub>~3 nm ???

This value allows to explain the excitation density quenching at low energy part of nonproportionality curve, but physical sense of this radius can be still discussed



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- CSI, μ<sub>e</sub>=8 cm<sup>2</sup> /V s at room temperature (B. P. Aduev, E. D. Aluker, G. M. Belokurov, and V. N. Shvayko, Phys. Status Solidi B 208, 137 (1998))
- $D_e = 0.2 \text{ cm}^2/\text{s}$
- $\Delta r_e = (D_e \Delta t)^{1/2}$
- $\Delta t=1 \text{ ps} \rightarrow \Delta r_e=4.5 \text{ nm}$
- $\Delta t=1 \text{ ns} \rightarrow \Delta r_e=140 \text{ nm}$



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# 3D diffusion-controlled recombination



Black sphere		$P = \begin{cases} 1, & r_{eh} < R_0 \\ R_0 / r_{eh}, & r_{eh} > R_0 \end{cases}$	
Coulomb		$P = 1 - \exp(-R_{Ons}/r_{eh})$ $\frac{e^2}{\varepsilon R_{Ons}} = k_B T$	
<i>ɛ</i> =5.7	<i>T</i> =300K	<i>R<sub>Ons</sub></i> =10 nm	
	<i>T</i> =77K	<i>R<sub>Ons</sub></i> =38 nm	
	<i>T</i> =10K	<i>R<sub>Ons</sub></i> =300 nm ???	
alized excitations P		Ir cc1	

For thermalized excitations  $R_{Ons}/r_{eh}$  << 1 – exciton yield after thermalization is low



## Phonon scattering mechanisms

Polarizational optical phonons

$$\tau_{LO}^{-1}(E_e) = \frac{e^2 \Omega_{LO} \sqrt{m_e^*}}{\widetilde{\varepsilon} \hbar \sqrt{2E_e}} [(n_{LO} + 1)\Lambda_- + n_{LO}\Lambda_+]$$
$$\Lambda_{\pm} = \ln \left( \left| \frac{\sqrt{E_e} + \sqrt{E_e \pm \hbar \Omega_{LO}}}{\sqrt{E_e} - \sqrt{E_e \pm \hbar \Omega_{LO}}} \right| \right)$$

Deformational acoustical phonons

$$\tau_{LA}^{-1}(E_{e}) = \frac{2^{1/2} \sigma_{s}^{2} m^{*3/2} k_{B} T \sqrt{E_{e}}}{\pi \rho \hbar^{4} C_{LA}^{2}}$$

Energy losses rate

$$\frac{d\overline{E_e}}{dt} = -S_{e,LA}\left(\overline{E_e}\right)$$

$$S_{LO}(E_e) = \frac{e^2 \Omega_{LO}^2 \sqrt{m_e^*}}{\widetilde{\varepsilon} \sqrt{2E_e}} \ln\left(\frac{4E_e}{\hbar\Omega_{LO}}\right)$$

picosecond duration of relaxation

nanosecond duration of relaxation (excitons in solid Kr are created in few ns after the pulse, Kirm, M., Kisand, V., Sombrowsky, E., Steeg, B., Vielhauer, S., and Zimmerer, G., Low Temperature Physics, v.29, 1081–1092 (2003))

 $S_{LA}(E) = \frac{2^{3/2} \sigma_s^2 m^{*5/2} E^{3/2}}{\pi \sigma_s^4}$ 

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are not well understood



![](_page_48_Figure_0.jpeg)

![](_page_48_Picture_1.jpeg)

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# Recombination of non-thermalized geminate e-h pairs

![](_page_49_Figure_1.jpeg)

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![](_page_49_Picture_4.jpeg)

# Recombination of non-thermalized geminate e-h pairs

![](_page_50_Figure_1.jpeg)

![](_page_50_Picture_2.jpeg)

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# Recombination and interaction of excitations in clusters of excitations (spurs)

![](_page_51_Figure_1.jpeg)

If energy transfer in a scattering is relatively low, only one excitation (exciton or electron-hole pair) is created. For higher energy transfers clusters consist of several excitations.

![](_page_51_Picture_3.jpeg)

![](_page_52_Figure_1.jpeg)

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![](_page_52_Picture_4.jpeg)

![](_page_53_Figure_1.jpeg)

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![](_page_54_Figure_1.jpeg)

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![](_page_55_Figure_1.jpeg)

## NaCl STE excitation spectra

![](_page_56_Figure_1.jpeg)

M. Yanagihara, Y. Kondo, H. Kanzaki: J. Phys. Soc. Jpn. 52, 4397 (1983)

![](_page_56_Picture_3.jpeg)

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## Structure of clusters of e-h pairs

![](_page_57_Figure_1.jpeg)

### Evolution of the cluster (role of electric fields)

$$\frac{\partial f_{e}(\mathbf{r}_{e},t)}{\partial t} = \frac{\partial}{\partial \mathbf{r}_{e}} \left( D_{e} \frac{\partial f_{e}(\mathbf{r}_{e},t)}{\partial \mathbf{r}_{e}} + \mu_{e} \mathbf{E}(\mathbf{r}_{e},t) f_{e}(\mathbf{r}_{e},t) \right) - \int \mathcal{K}_{eh \to ex}(\mathbf{r}_{e} - \mathbf{r}_{h}) g_{eh}(\mathbf{r}_{e},\mathbf{r}_{h},t) d\mathbf{r}_{h}$$

$$\frac{\partial f_{h}(\mathbf{r}_{h},t)}{\partial t} = \frac{\partial}{\partial \mathbf{r}_{h}} \left( D_{h} \frac{\partial f_{h}(\mathbf{r}_{h},t)}{\partial \mathbf{r}_{h}} - \mu_{h} \mathbf{E}(\mathbf{r}_{h},t) f_{h}(\mathbf{r}_{h},t) \right) - \int \mathcal{K}_{eh \to ex}(\mathbf{r}_{e} - \mathbf{r}_{h}) g_{eh}(\mathbf{r}_{e},\mathbf{r}_{h},t) d\mathbf{r}_{e}$$

$$\mathbf{E}(\mathbf{r}_{e},t) = \underbrace{e}_{e} \sum_{i \neq i} \frac{\mathbf{r}_{ei} - \mathbf{r}_{ei}}{|\mathbf{r}_{ei} - \mathbf{r}_{ei}|^{3}} - \sum_{j} \frac{\mathbf{r}_{ei} - \mathbf{r}_{hj}}{|\mathbf{r}_{ei} - \mathbf{r}_{hj}|^{3}} \right)$$
Dimensionless time
Einstein–Smoluchowski relation:
$$\mu_{e} = \underbrace{e}_{k_{B}T} D_{e}$$

$$D_{e} t / \frac{P_{Ons}^{2}}{P_{Ons}} = \frac{e^{2}}{\varepsilon k_{B}T}$$

$$P_{Ons} = \frac{e^{2}}{\varepsilon k_{B}T} + R_{Ons} \left( \sum_{i \neq i} \frac{\mathbf{r}_{ei} - \mathbf{r}_{ei}}{|\mathbf{r}_{ei} - \mathbf{r}_{ei}|^{3}} - \sum_{i} \frac{\mathbf{r}_{ei} - \mathbf{r}_{hj}}{|\mathbf{r}_{ei} - \mathbf{r}_{hj}|^{3}} \right) f_{e}(\mathbf{r}_{e}, t) - \dots$$

![](_page_58_Picture_2.jpeg)

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## Structure of clusters of e-h pairs

![](_page_59_Figure_1.jpeg)

![](_page_60_Figure_0.jpeg)

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### Kinetics of exciton creation in clusters

![](_page_61_Figure_1.jpeg)

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![](_page_61_Picture_3.jpeg)

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## Structure of clusters of e-h pairs

![](_page_62_Figure_1.jpeg)

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![](_page_62_Picture_4.jpeg)

### Kinetics of exciton creation in clusters

![](_page_63_Figure_1.jpeg)

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 Exciton creation kinetics within clusters depends on the ratio of cluster diameter to Onsager radius and on the number of excitations in the cluster. The higher the number of electron-hole pairs, (1) the shorter the raising time, (2) higher the yield of excitons, and (3) higher the fraction of triplets created due to the e-h recombination.

![](_page_64_Picture_2.jpeg)

## Can excitons do not interact with each other and with STHs?

![](_page_65_Figure_1.jpeg)

### Cherenkov, ionization and bremsstrahlung for electron (integrated over range)

![](_page_66_Figure_1.jpeg)

#### Effect of absorption in the transparency region on Cherenkov spectrum

![](_page_67_Figure_1.jpeg)

## Limitations from absorption

- Energy resolution of 0.01 for both (Cherenkov and scintillator) channels for total absorption calorimeter demands high transparency of the crystal: exp(-k(w)d)>0.99, therefore k(w) should be better than 0.01 cm<sup>-1</sup> for 1 cm crystal.
- Cherenkov spectrum is wide spectral filtering is required (narrow band filters?)
- Scintillators with intrinsic type of emission is desirable - no activator absorption bands
- Probably it is possible to use red Cherenkov no Urbach tail effect and better radiation hardness (in IR extremely high transparency can be achieved for glasses)

![](_page_68_Picture_5.jpeg)

### How can we obtain scintillating yield below 1 photon/MeV? Are there any limitations to achieve even less yields for crystals with excellent transparency?

Scintillators with intrinsic emission – low yield can be achieved only due to quenching of the intrinsic emission

1) If this quenching is temperature one (like in case of PWO), the result is the strong temperature dependence of the yield, and the temperature of the detector should be stabilized over the whole detector (the lower yield, the higher the degree of temperature dependence).

2) If the quenching is due to the energy transfer from e.g. excitons to some quenchers, the absorption band of the quencher should strongly decrease the transparency of the crystal.

![](_page_69_Picture_4.jpeg)

### How can we obtain scintillating yield below 1 photon/MeV? Are there any limitations to achieve even less yields for crystals with excellent transparency?

Scintillators with activators:

Low scintillation yield and high transparency means very low concentration of activators.

This low activator concentration should make the energy resolution worse:

- 1) due to spatial fluctuations in activator distribution
- 2) due to long migration path for excitons from the track region to activators
- 3) etc. this problem is not clear now

![](_page_70_Picture_7.jpeg)

## How can we obtain scintillating yield below 1 photon/MeV?

## Are there any limitations to achieve even less yields for crystals with excellent transparency?

Some additional channels of energy relaxation can become important for scintillators with extremely low yield – e.g. hot intraband luminescence (with yield  $10^{-5}-10^{-4}$ )

![](_page_71_Figure_3.jpeg)

Competition between phonon-assisted electron (hole) relaxation and intraband radiation process – cannot be avoided

 $w_{phonon} \sim 10^{12} - 10^{14} \, \text{s}^{-1}$  $w_{rad} \sim 10^8 - 10^9 \, \text{s}^{-1}$ 

 $\eta = w_{rad} / (w_{phonon} + w_{rad}) \sim 10^{-5} - 10^{-4}$ 

Picosecond hot luminescence – can be mixed with Cherenkov radiation, but originate from ionization channel

![](_page_71_Picture_8.jpeg)
## Intrinsic picosecond luminescecne of CsI



## How can we separate Cherenkov emission from scintillation in case of compatible yields of these channels?

Spectral separation: Since Cherenkov emission is well overlapped spectrally with any kind of scintillation emission, the spectral separation using filters demands additional treatment of the signals.

Time separation:

Cherenkov emission is subnanosecond for 10 cm crystals. Therefore the separation can be based on the long (hundreds of nanosecond) scintillator emissions. But the low yield scintillators with intrinsic type of emission should shorten their emission time due to the quenching.

