

# Dynamics of excited region created by a VUV-X photon in solids

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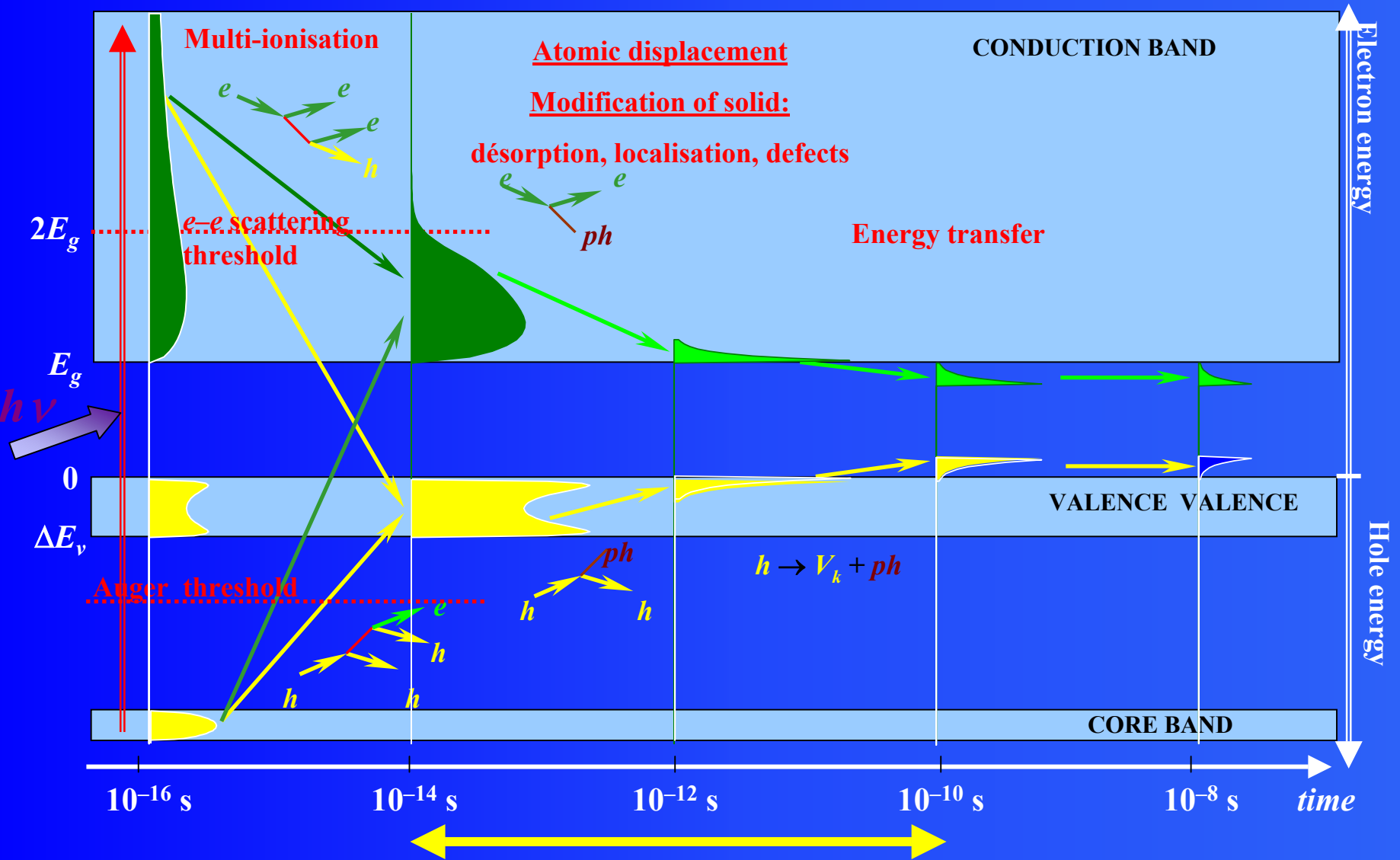
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Gory, 119992, Moscow, Russia

# Dynamics of electronic relaxation in large bandgap solids

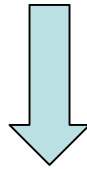
100 as 1 fs

1 ps

1 ns



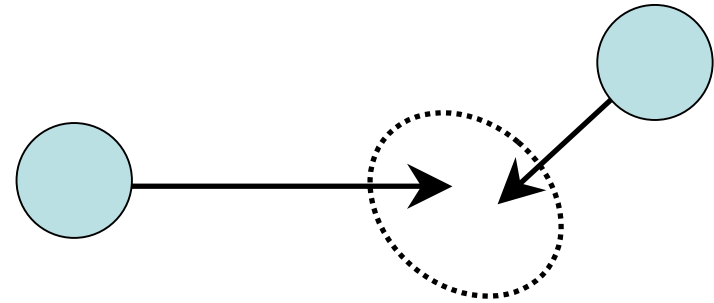
Modification of large band gap solids  
by electronic relaxation



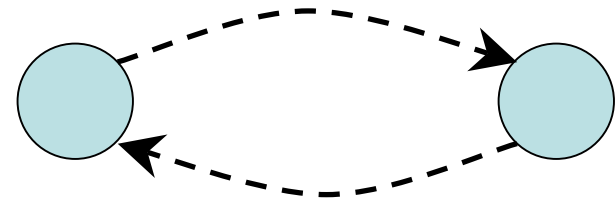
Interaction of high energy electronic excitations

# Three types of EE interaction

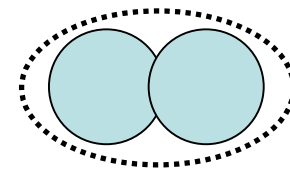
- Weak interaction by collision (mean distance between excitations is well above 100 nm)



- Interaction due to energy exchange (mean distance between excitations is about 10 nm)



- Strong interaction with modification of solid (mean distance between excitations is about 1 nm)

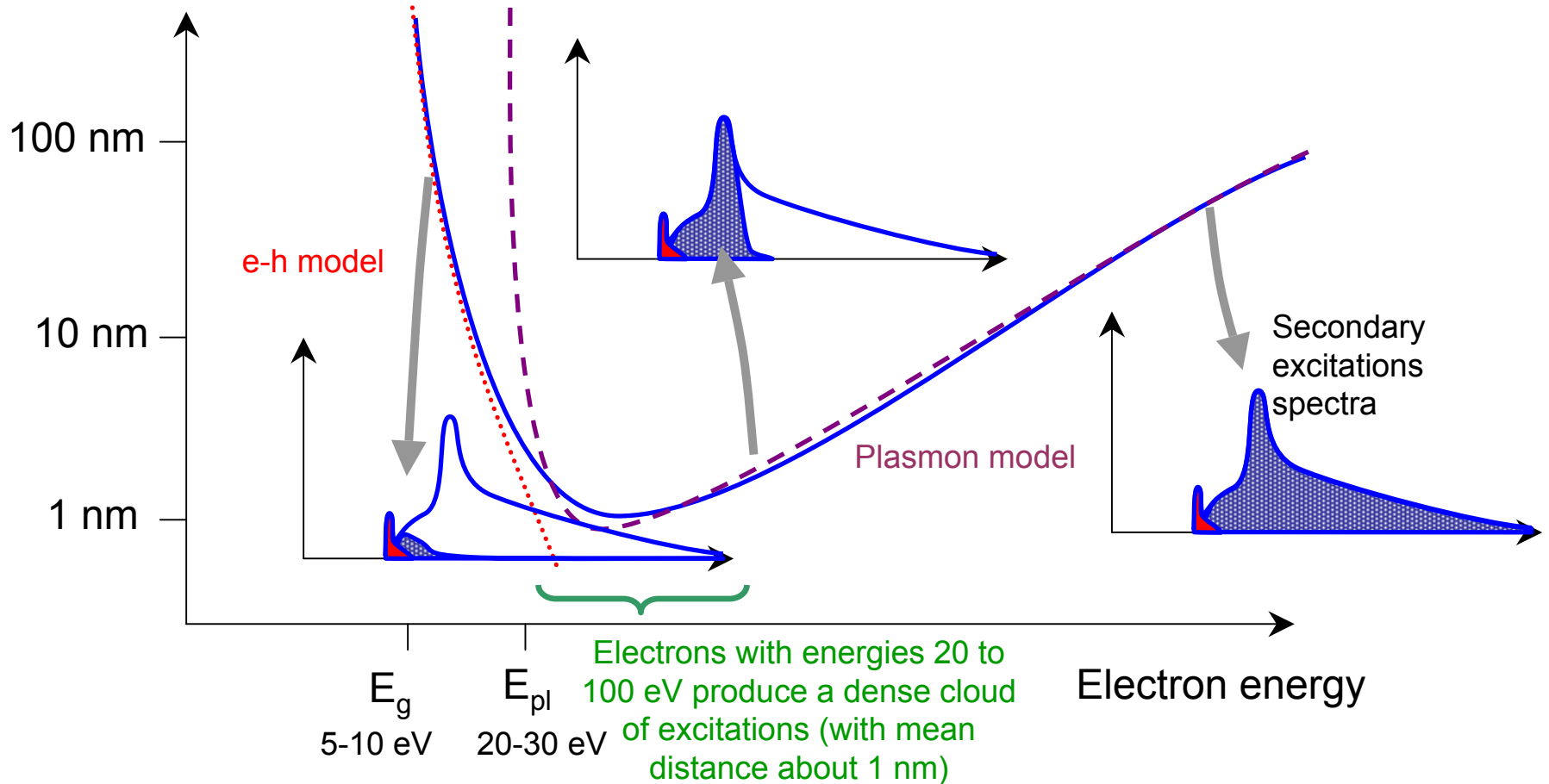


At dense excitation → superposition of all types

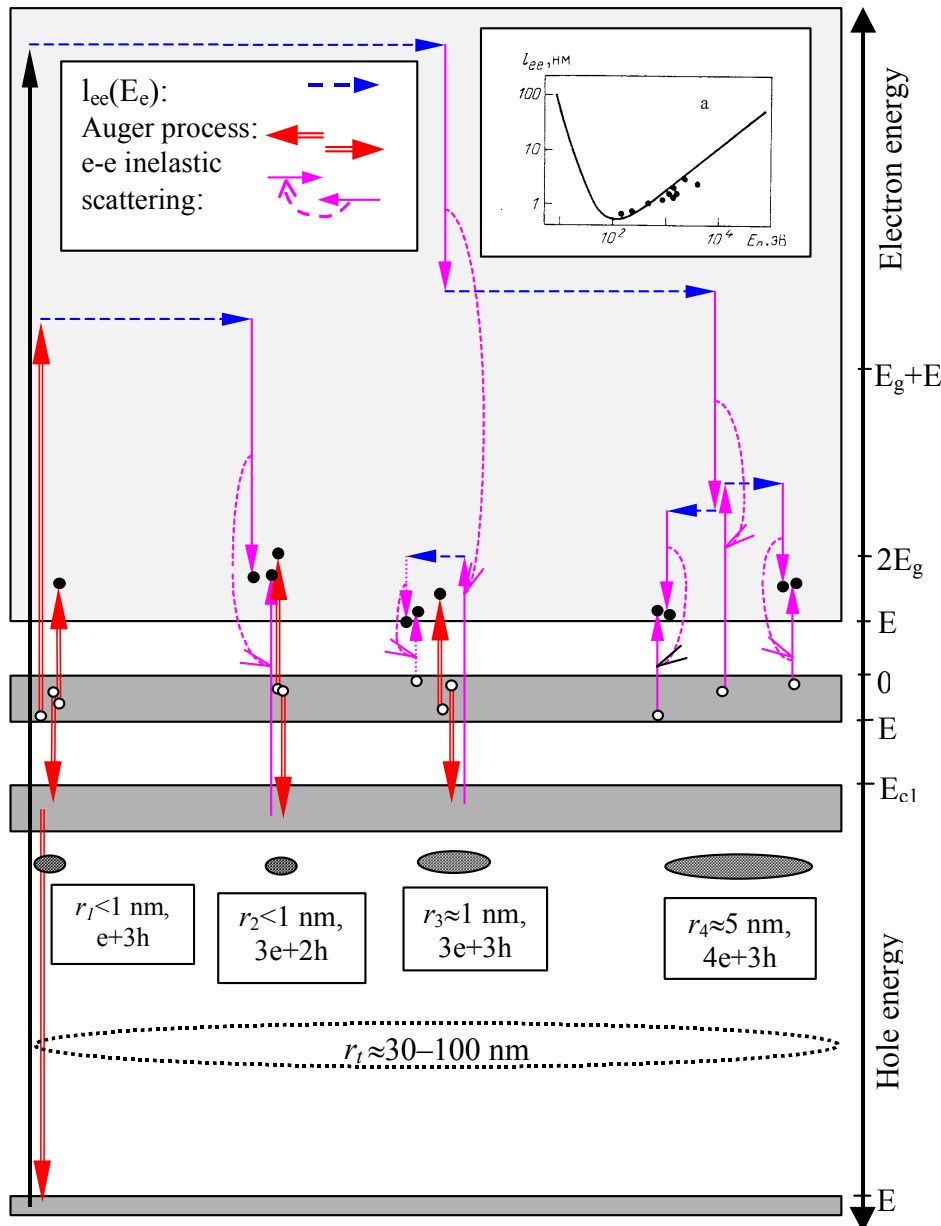
For investigation → VUV-X spectroscopy

# Creation of secondary electronic excitation area by hot electrons

Electron mean free path : e-e scattering



# Excited region in solids created by VUV-X photon



The spatial distribution of electronic excitations is controlled by energy of VUV photon:

*Electrons with energies 10 to 100 eV produce excited region with mean distance about between the excitations 1 to 100 nm*

*Auger decay of a deep core hole results in creation of multiple charged ions – several excitations in one elementary cell*

# *Investigation of fundamental mechanisms of electronic relaxation in large bandgap solids*

*Experimental approach :*

*Excitation by VUV-X photon with selected energy*

*from 10 eV to 1 KeV*

*Detection of luminescence kinetics*

*from 100 fs to 100 ps*

# Electron excitation correlation and luminescence decay kinetics

Case of excitation ( $n$ ) quenching on correlated centers ( $c$ )

$$\left\{ \begin{array}{l} \frac{\partial n}{\partial t} - D_n \frac{\partial^2 n}{\partial \mathbf{r}^2} = -\frac{n}{\tau_R} - \frac{nc}{\tau_R} \int d^3 r' K(|\mathbf{r} - \mathbf{r}'|) G_{nc}(\mathbf{r}, \mathbf{r}', t), \\ \frac{\partial c}{\partial t} - D_c \frac{\partial^2 c}{\partial \mathbf{r}^2} = -\frac{c}{\tau_c} - \frac{nc}{\tau_R} \int d^3 r' K(|\mathbf{r} - \mathbf{r}'|) G_{nc}(\mathbf{r}, \mathbf{r}', t), \\ \frac{\partial G_{nc}}{\partial t} - D_n \frac{\partial^2 G_{nc}}{\partial \mathbf{r}_n^2} - D_c \frac{\partial^2 G_{nc}}{\partial \mathbf{r}_c^2} = -\frac{1}{\tau_R} K(|\mathbf{r} - \mathbf{r}'|) G_{nc}(\mathbf{r}, \mathbf{r}', t), \end{array} \right. \quad \begin{array}{l} \text{Dipole-dipole transfer:} \\ K(r) = (R_0/r)^6 \end{array}$$

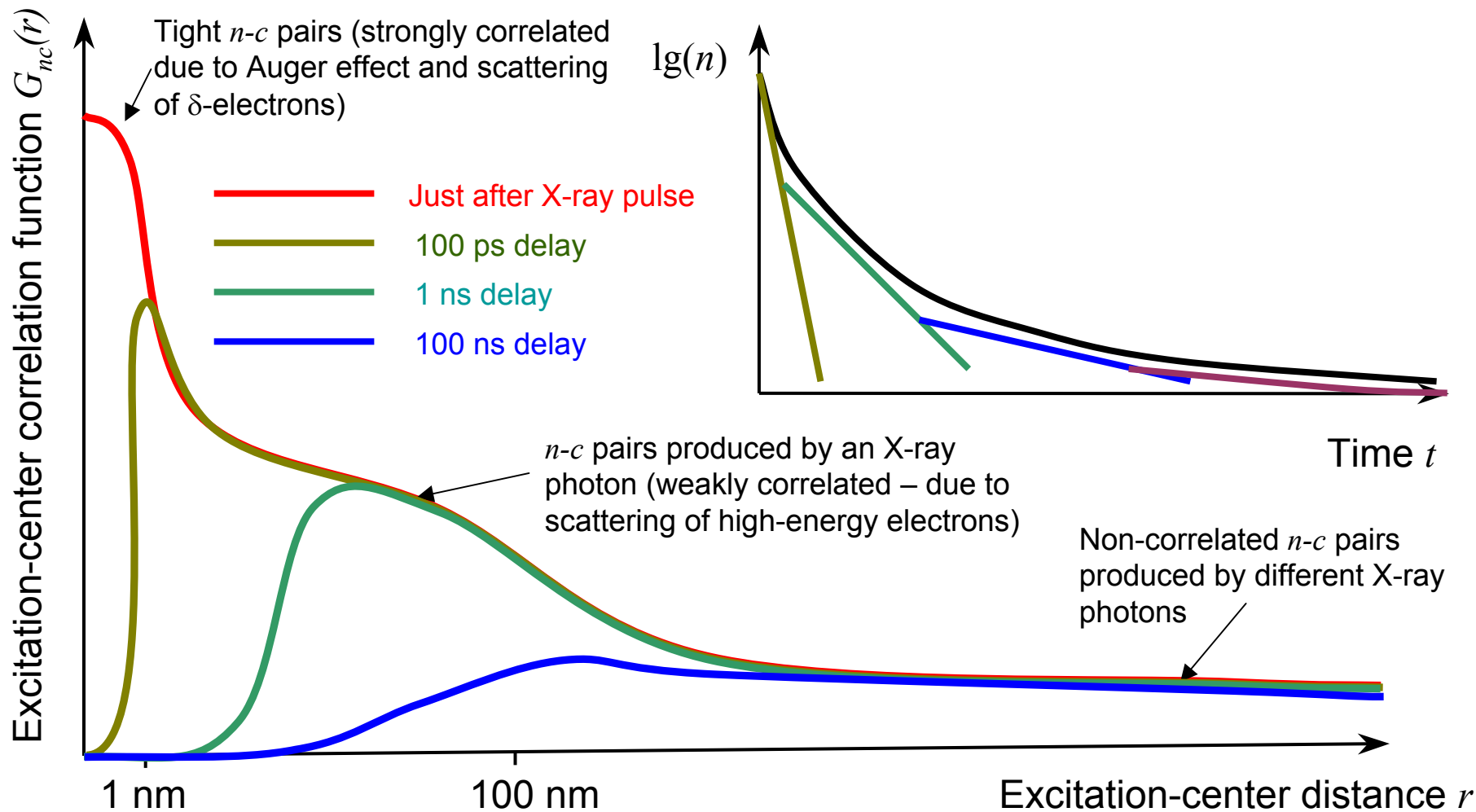
$$n(t) = n(0) \exp(-t/\tau_R) Q(t)$$

Quenching factor:

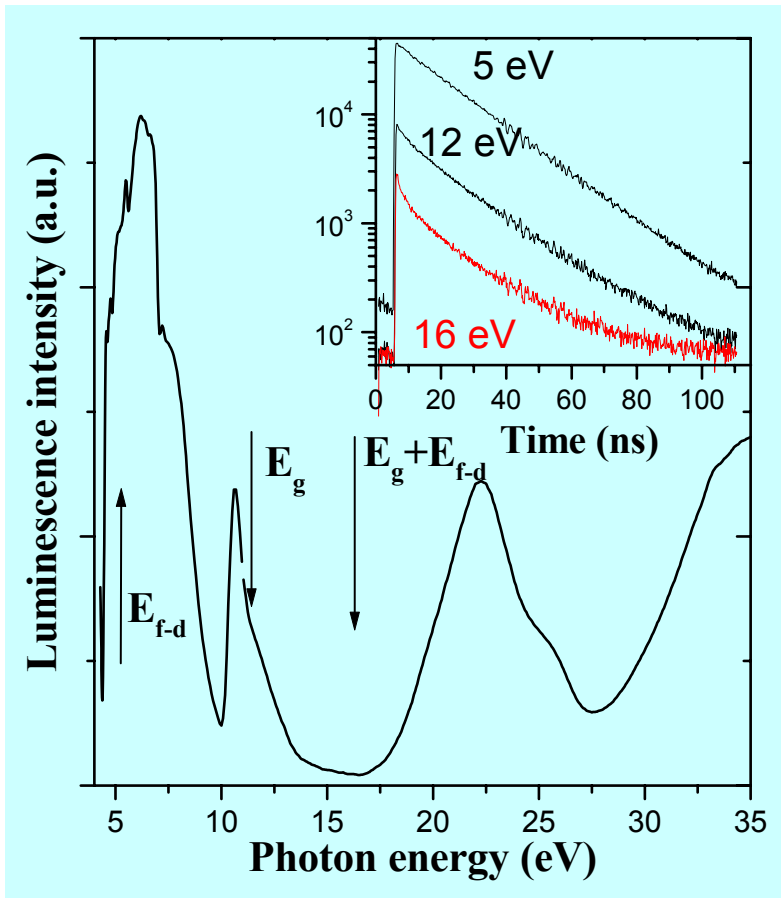
$$Q(t) = \exp \left( -\beta \int d^3 r \frac{K(r)}{K(r) + \tau_R/\tau_c} G(r, 0) \left( 1 - \exp \left( -\frac{t}{\tau_c} \left( 1 + \frac{\tau_c K(r)}{\tau_R} \right) \right) \right) \right)$$

# Electron excitation correlation and luminescence decay kinetics

## Non-exponential decay due to fast non-radiative transfer in tight excitation-center pairs



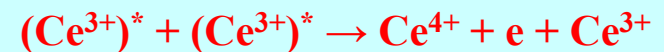
*Interaction of cerium excitons in CeF<sub>3</sub> (energy transfer)  
The elementary region of high local density of excitations  
(SuperACO)*



At high energy excitation luminescence decay of CeF<sub>3</sub> is non exponential and show a strong acceleration

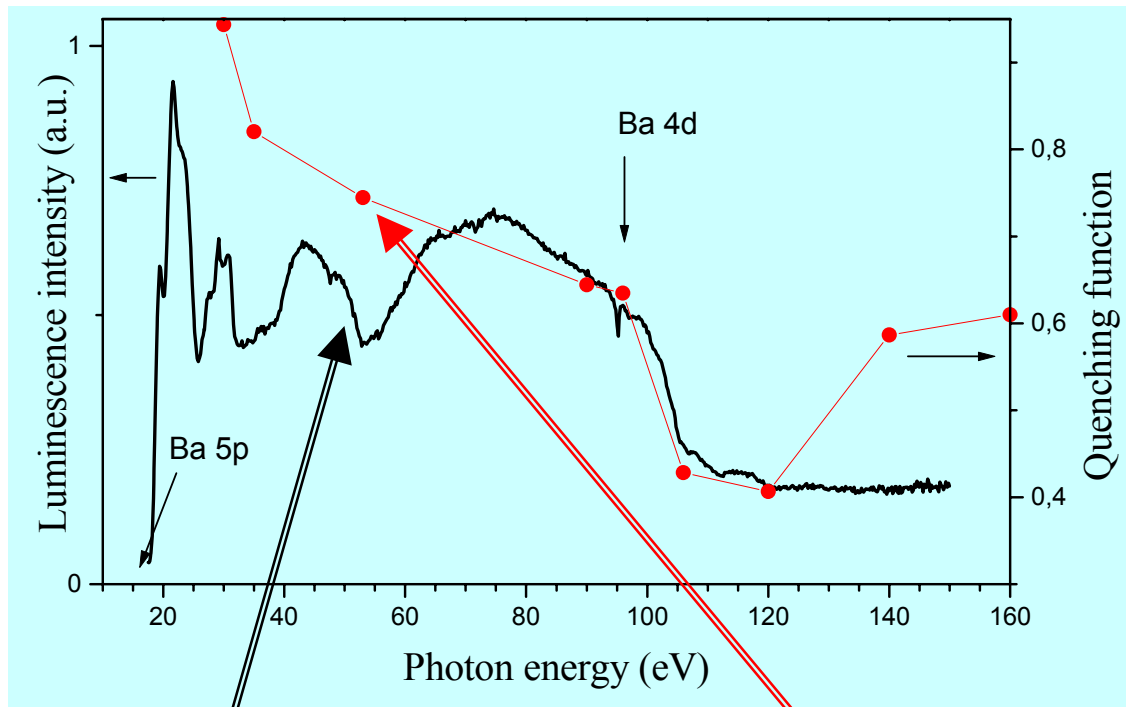
Energy threshold of decay acceleration in CeF<sub>3</sub> is at 16 eV

Above 16 eV inelastic scattering of the primary photoelectron can create two excited ions (Ce<sup>3+</sup>)<sup>\*</sup> in close vicinity. The result of their interaction can be written as



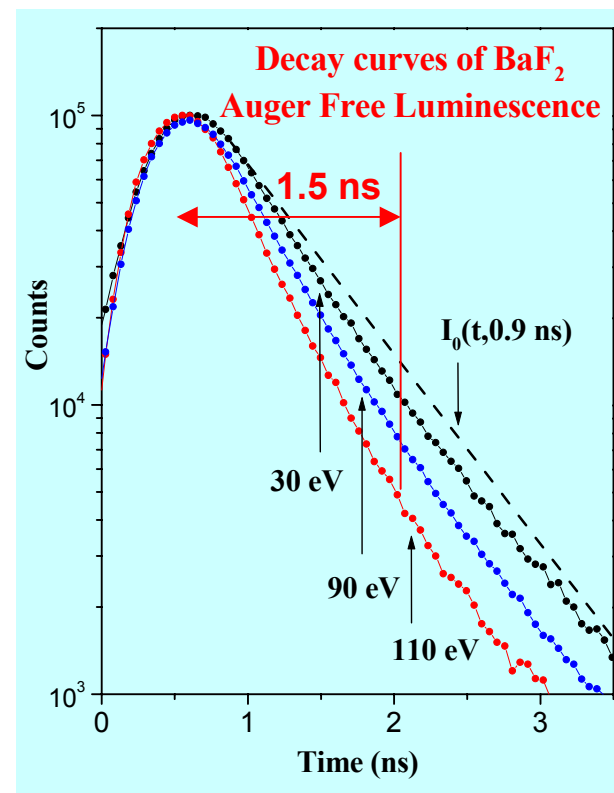
From simulation of interaction the acceleration of decay is from picosecond range

# 5p core hole interaction with excitons and conduction electrons in BaF<sub>2</sub> (SuperACO)



Crossluminescence excitation spectrum

Quenching factor at 1.5 ns after excitation pulse maximum



CL decay curves at different excitation energies

# Investigation of the strong EE interaction and atomic displacement dynamics

## Experimental

Time range

100 fs – 100 ps

Photon energy range

10 eV – 1 KeV

### VUV-X excitation:

Energy resolution  $\sim 10^3$  at 100 eV, pulse length  $\sim 100$  fs

### Luminescence detection:

1. Frequency mixing method (T.Shimizu et al. PRL 2003)

2. dissector (stroboscopic photomultiplier):

Time resolution  $\sim 20$  ps – a good experience with Synchrotron radiation (MHz)

Time resolution  $< 1$  ps (stroboscopic rate  $\sim$  GHz) in development by  
CELIA & FEMTOSCAN (Bordeaux)

INP (Novosibirsk)

## Conclusions

*Fundamental mechanisms of electronic relaxation in large bandgap solids can be studied by analysis of luminescence kinetics excited by VUV-X photon (Time-Resolved Luminescence VUV Spectroscopy)*

*The study of mechanisms which control the strong interaction between electronic excitations needs the sub-picosecond VUV-X radiation*

*The main experimental technique of sub-picosecond luminescence detection is based on frequency mixing and GHz stroboscopic methods*