Connection of electron and phonon structure of scintillators with spatial structure of excited regions and efficiency of bulk and nano scintillators

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Cascade

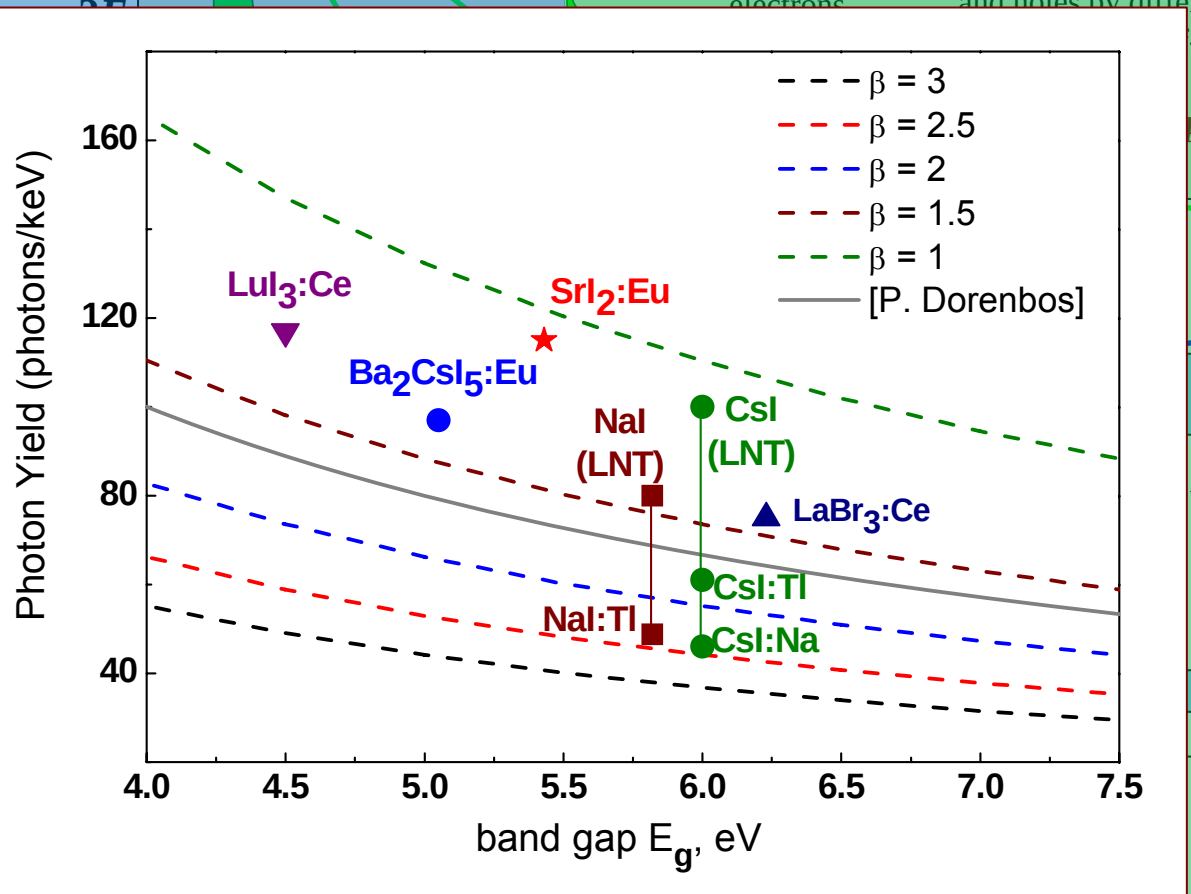
Transport

Center

$(\beta E_g)^{-1}$

S

Q



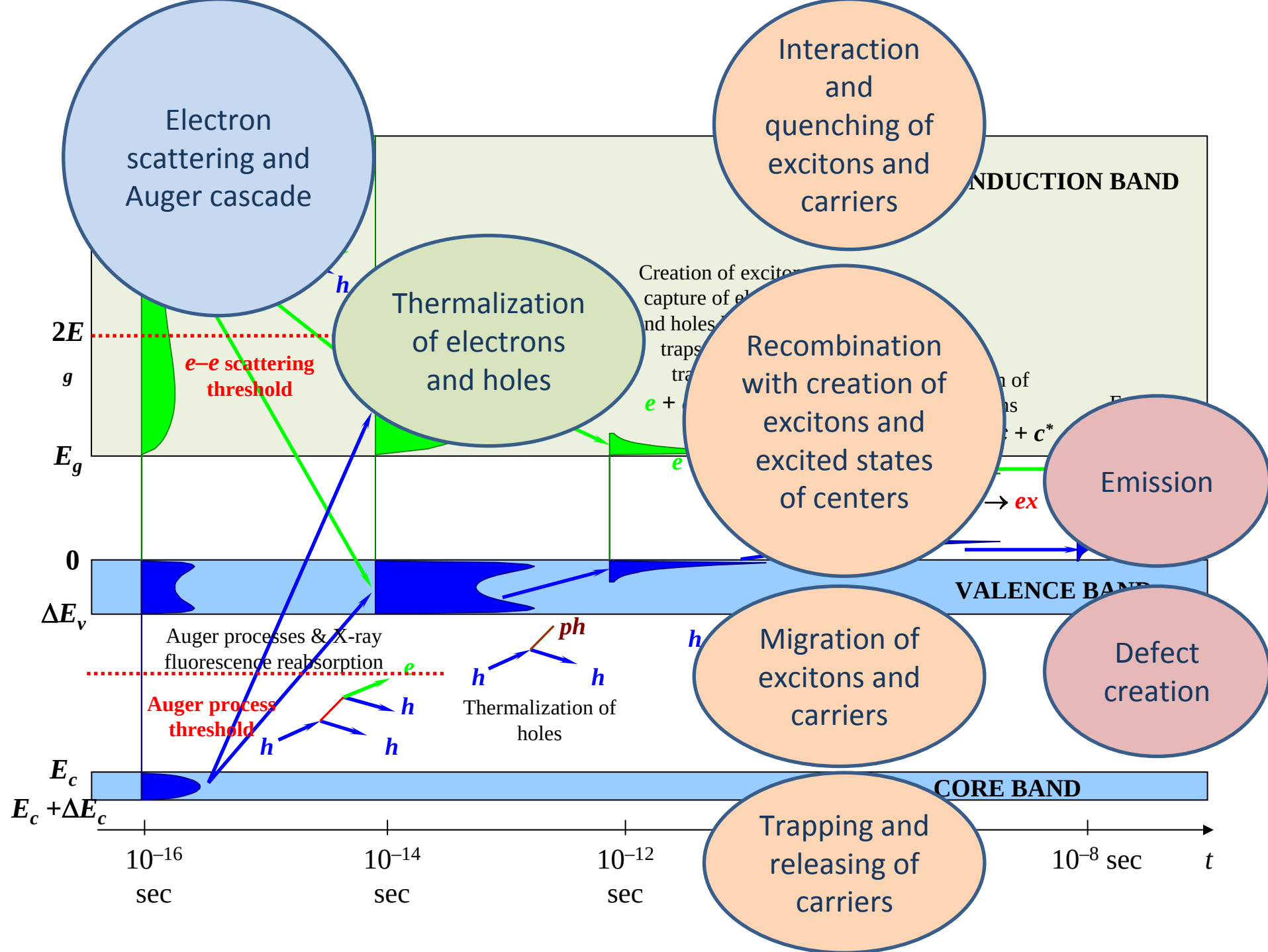
Applications and material engineering request quantitative predictions of properties of new scintillating materials

Simple estimation of scintillation efficiency as βSQ (cascade-transport-center) is only qualitative

~~New theory of scintillations?~~

Re-estimation of the role of different stages of energy relaxation in crystals on the basis of deeper experimental investigation of new materials

Contemporary model: Scintillation as collective interconnected processes of spatial and temporal evolution of strongly non-equilibrium excited region in media


Electron scattering and Auger cascade
Interaction and quenching of excitons and carriers
Thermalization of electrons and holes
Recombination with creation of excitons and excited states of centers
Emission
Migration of excitons and carriers
Defect creation
Trapping and releasing of carriers
NDUCTION BAND
2E
g
E
g
0
ΔE
v
E
c
E
c
+ΔE
c
e–e scattering threshold
Auger processes & X-ray fluorescence reabsorption
Auger process threshold
Thermalization of holes
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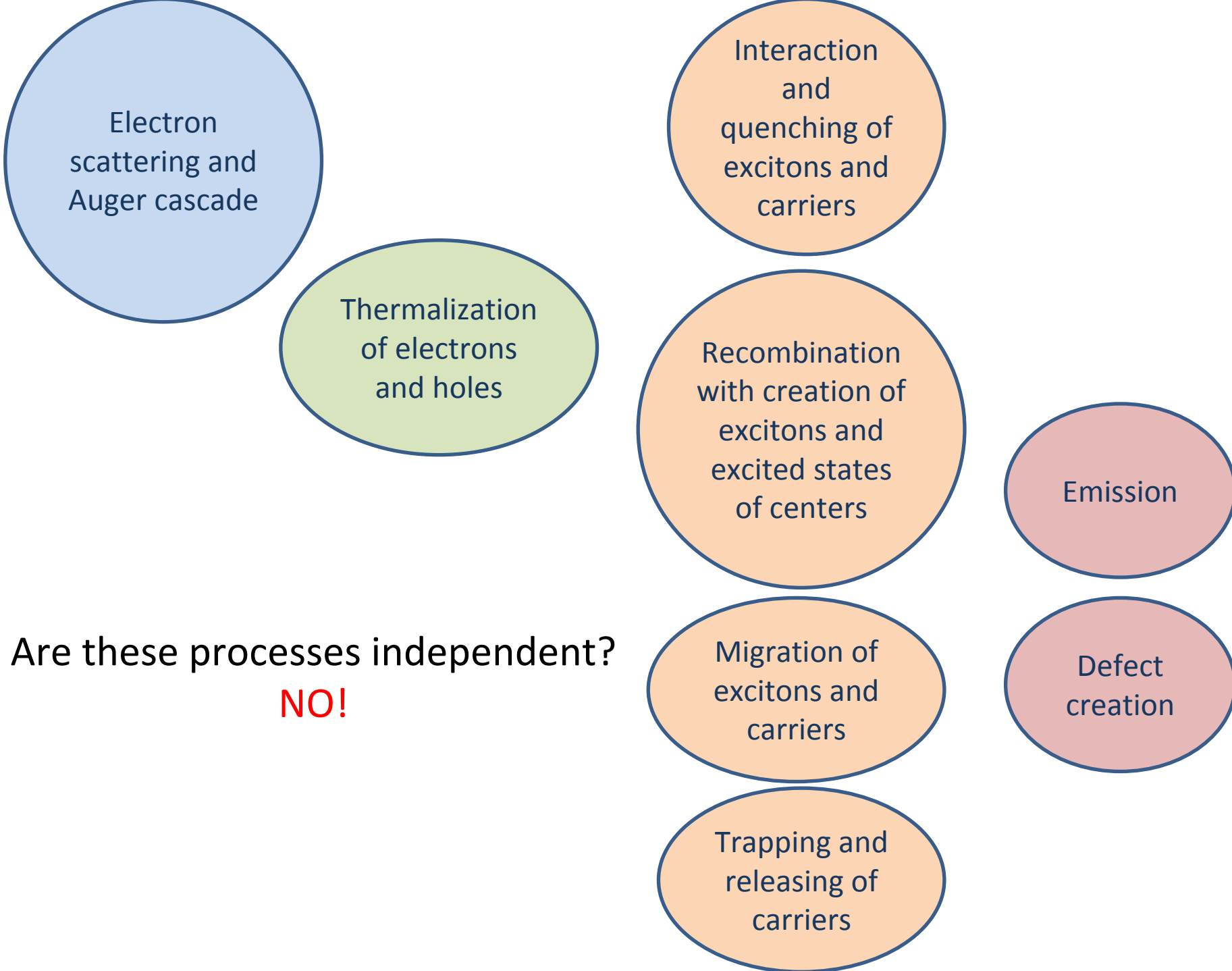
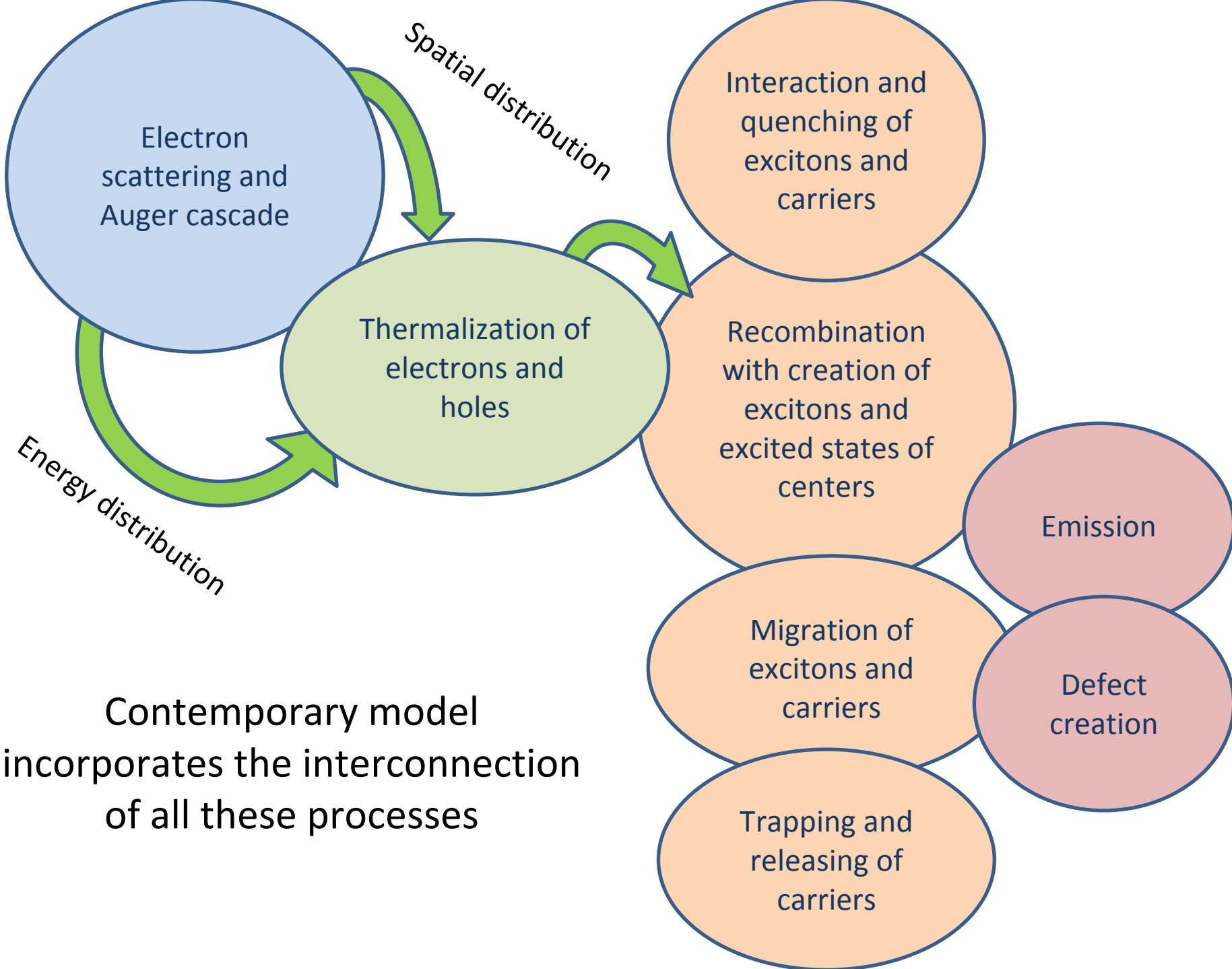
VALENCE BAND
CORE BAND
10–16 sec
10–14 sec
10–12 sec
10–8 sec
t

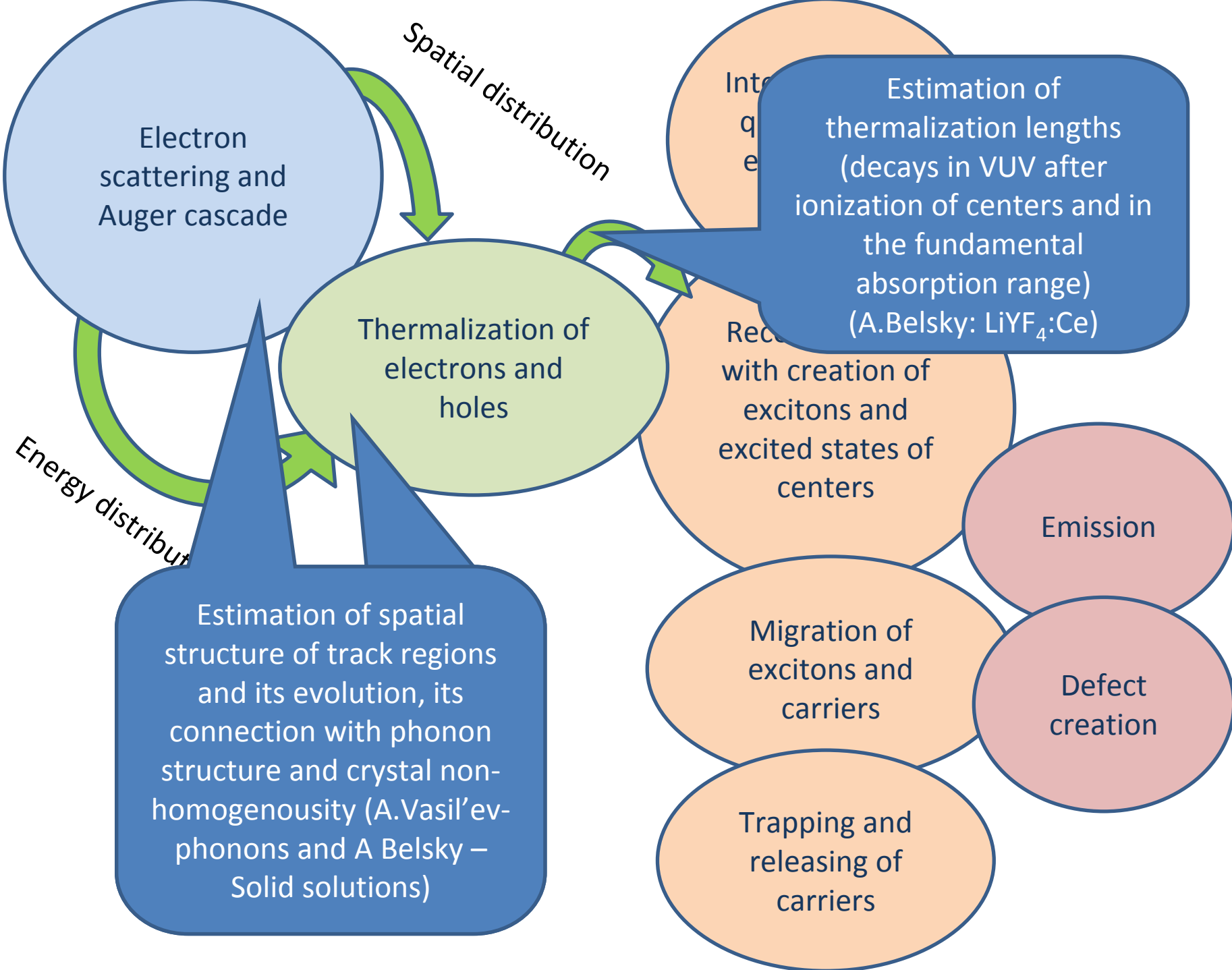


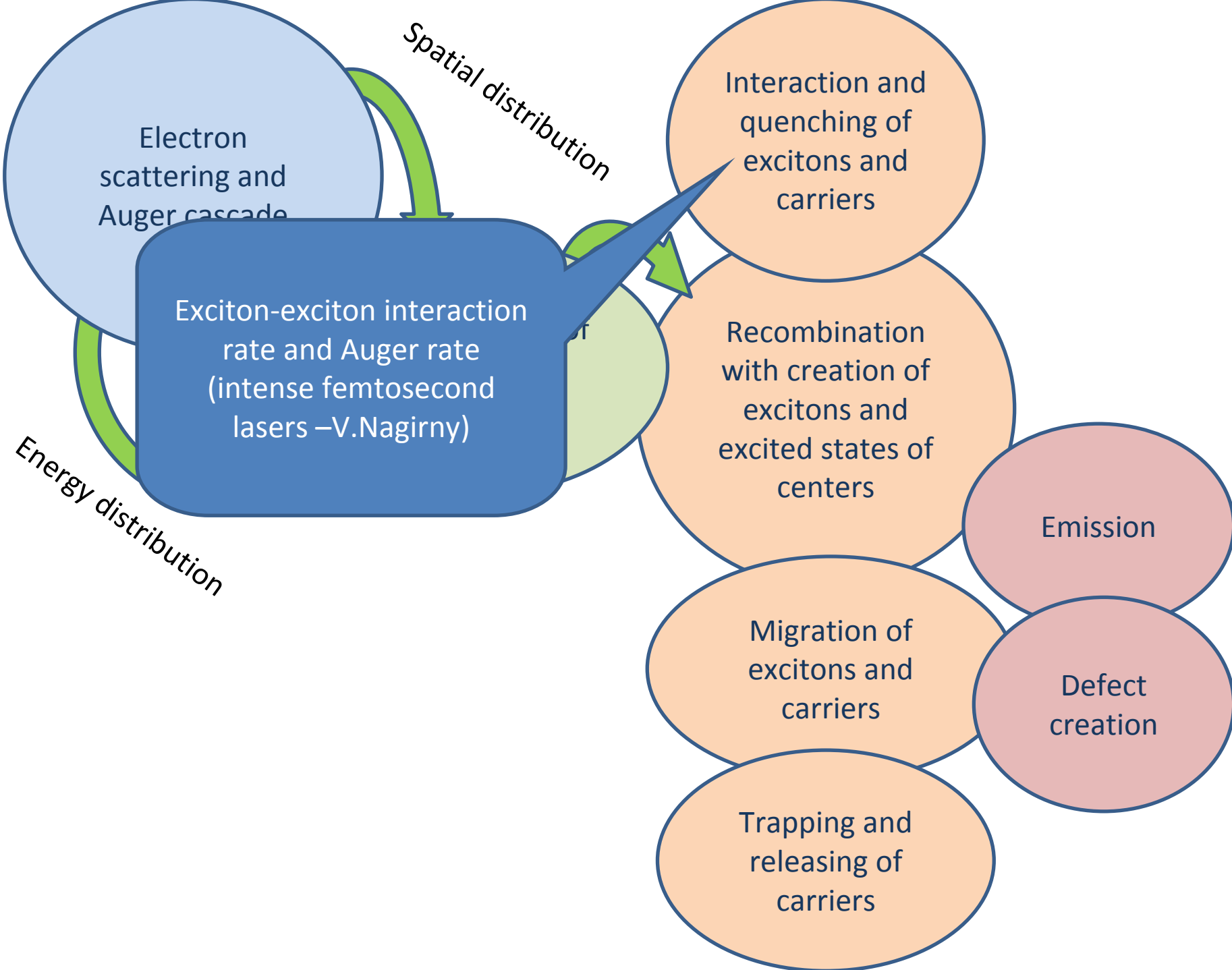
Are these processes independent?

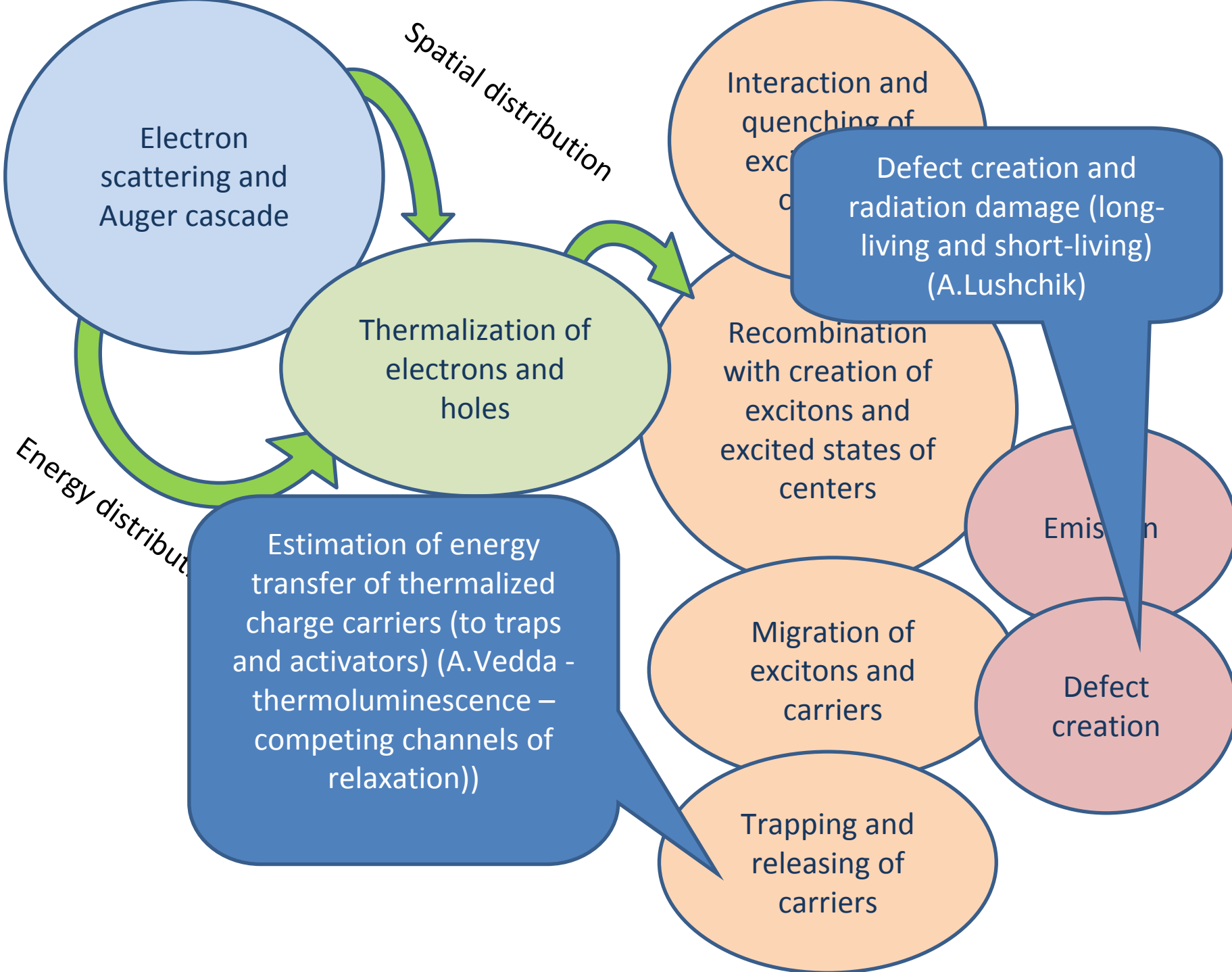
NO!

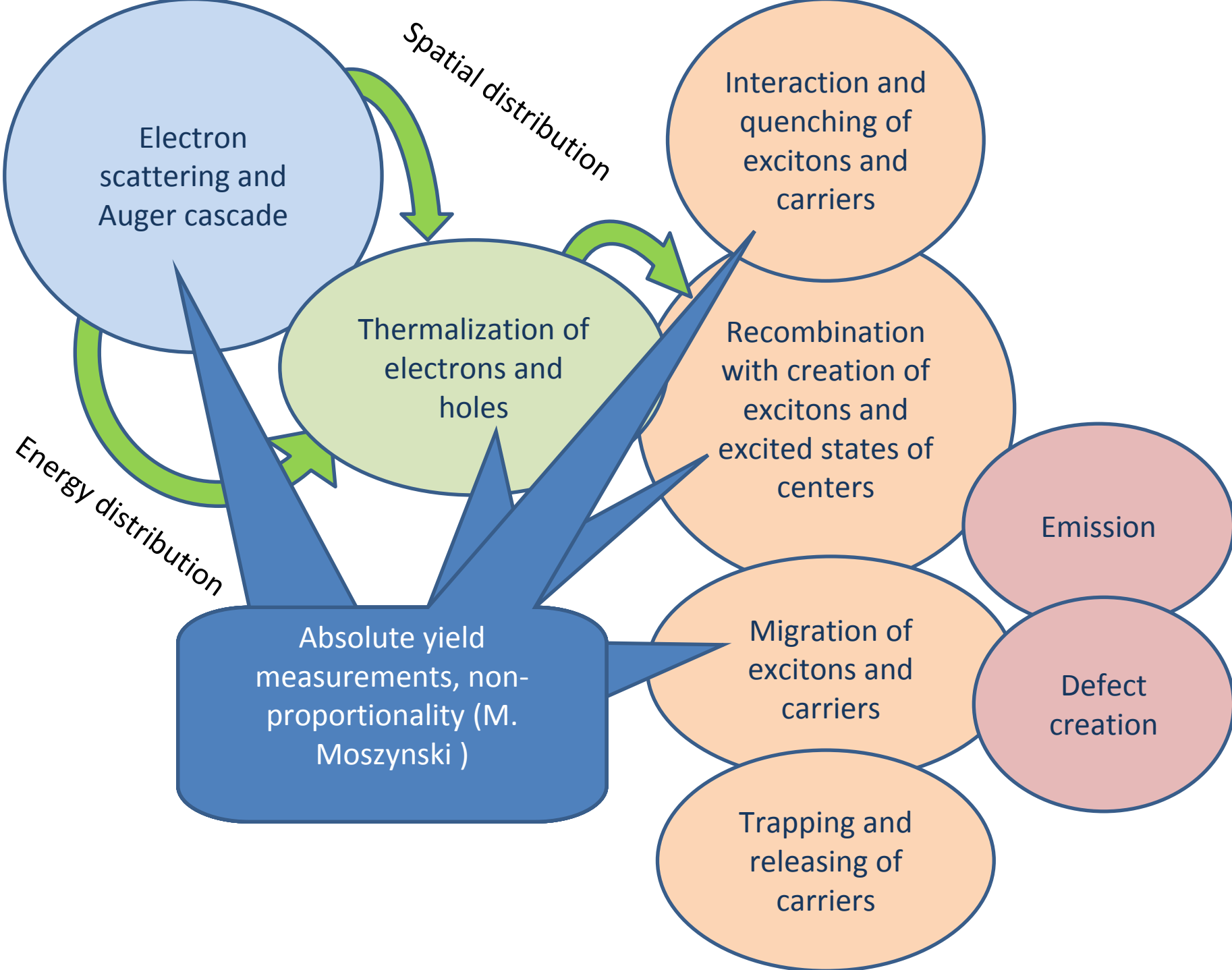


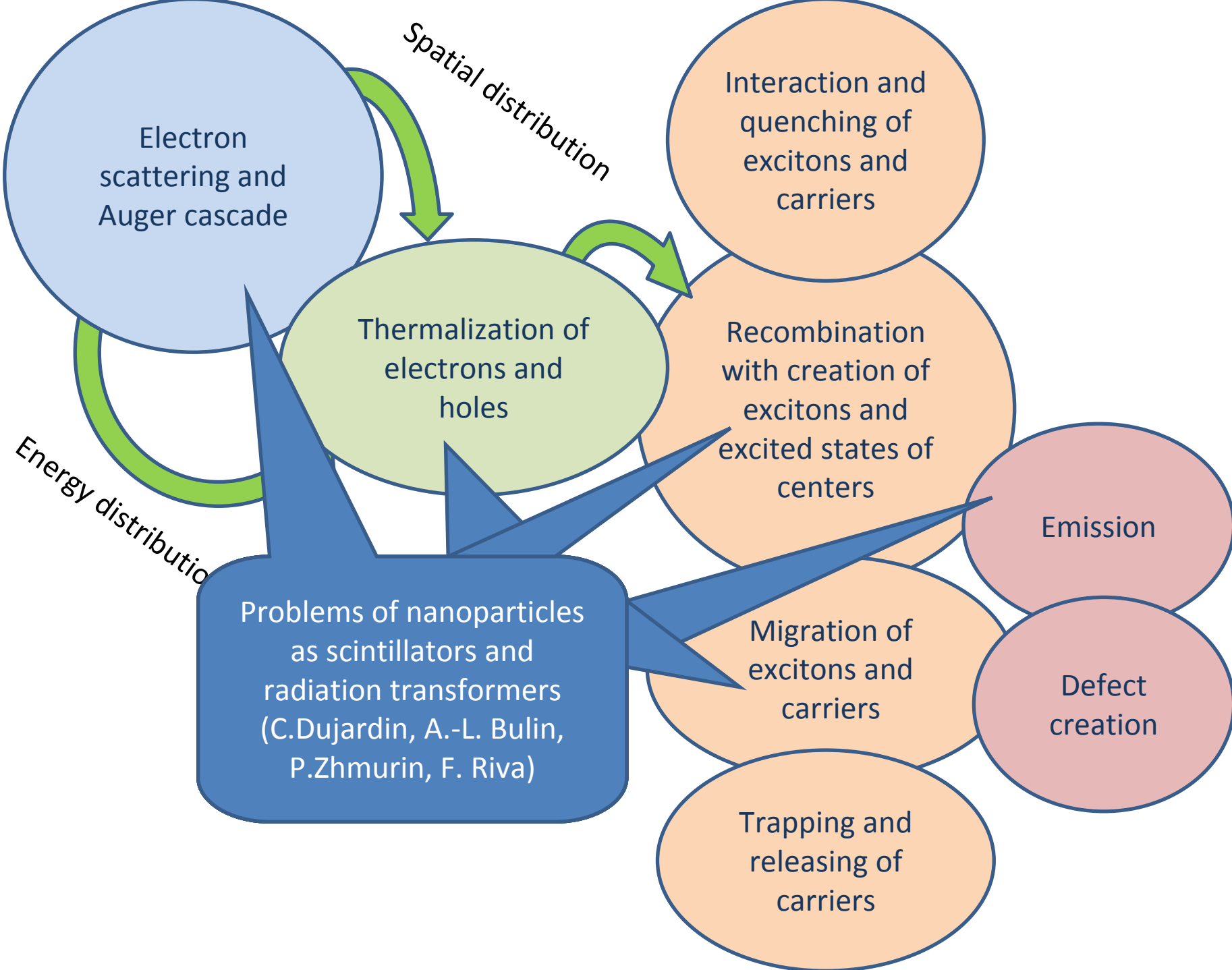
Contemporary model incorporates the interconnection of all these processes


Electron scattering and Auger cascade
Spatial distribution
Int
q
e
Estimation of thermalization lengths (decays in VUV after ionization of centers and in the fundamental absorption range) (A.Belsky: LiYF4:Ce)
Thermalization of electrons and holes
Rec
with creation of excitons and excited states of centers
Energy distribut
Emission
Migration of excitons and carriers
Defect creation
Estimation of spatial structure of track regions and its evolution, its connection with phonon structure and crystal non-homogenousity (A.Vasil'ev-phonons and A Belsky – Solid solutions)
Trapping and releasing of carriers


Electron scattering and Auger cascade
Spatial distribution
Interaction and quenching of excitons and carriers
Exciton-exciton interaction rate and Auger rate (intense femtosecond lasers –V.Nagirny)
Recombination with creation of excitons and excited states of centers
Energy distribution
Emission
Migration of excitons and carriers
Defect creation
Trapping and releasing of carriers


Electron scattering and Auger cascade
Spatial distribution
Interaction and quenching of exci
Defect creation and radiation damage (long-living and short-living) (A.Lushchik)
Thermalization of electrons and holes
Recombination with creation of excitons and excited states of centers
Energy distribut
Emis n
Estimation of energy transfer of thermalized charge carriers (to traps and activators) (A.Vedda - thermoluminescence – competing channels of relaxation))
Migration of excitons and carriers
Defect creation
Trapping and releasing of carriers

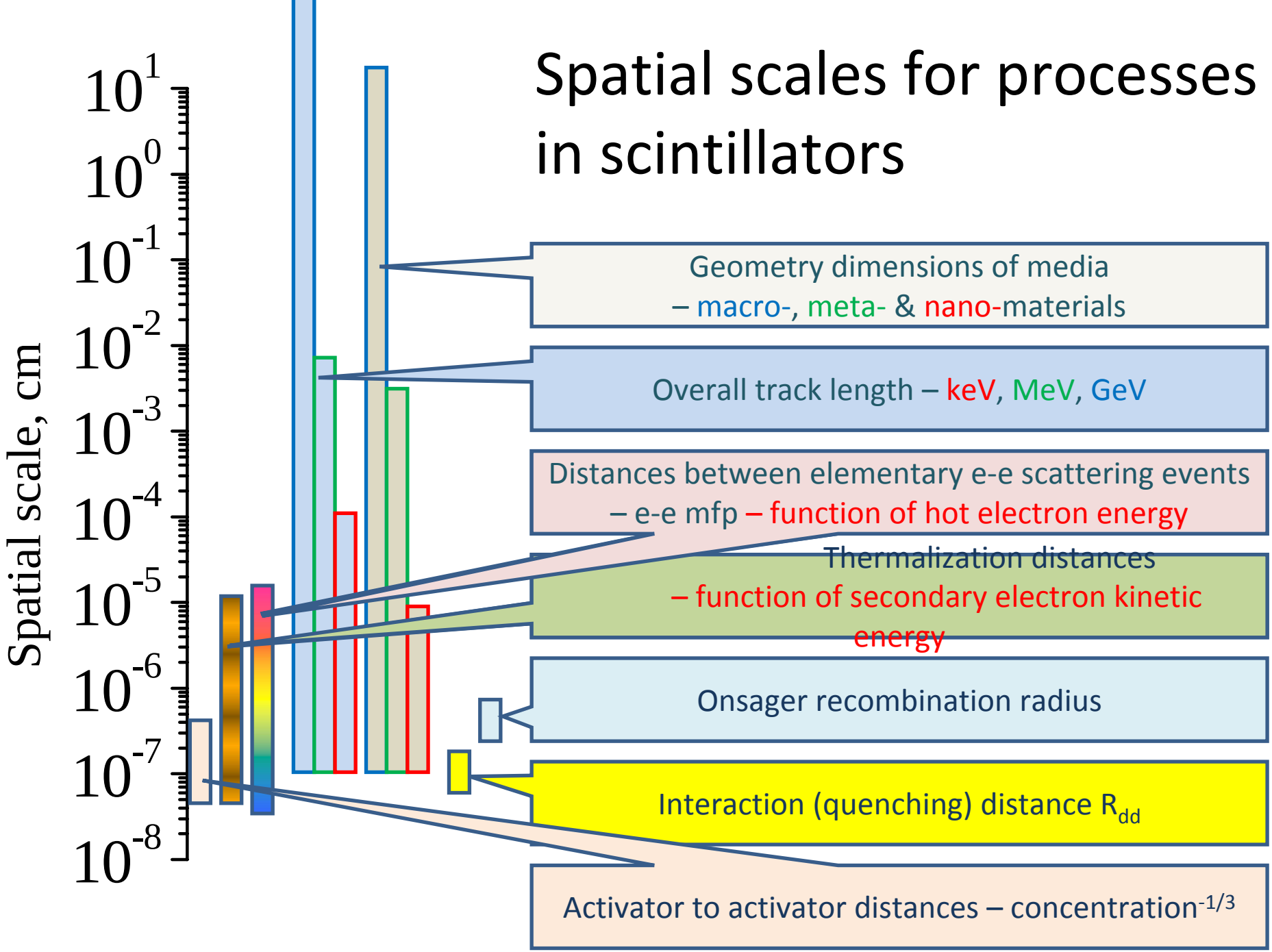

Electron scattering and Auger cascade
Spatial distribution
Interaction and quenching of excitons and carriers
Thermalization of electrons and holes
Recombination with creation of excitons and excited states of centers
Energy distribution
Emission
Absolute yield measurements, non-proportionality (M. Moszynski)
Migration of excitons and carriers
Defect creation
Trapping and releasing of carriers


Electron scattering and Auger cascade
Spatial distribution
Interaction and quenching of excitons and carriers
Thermalization of electrons and holes
Recombination with creation of excitons and excited states of centers
Energy distributio
Emission
Problems of nanoparticles as scintillators and radiation transformers (C.Dujardin, A.-L. Bulin, P.Zhmurin, F. Riva)
Migration of excitons and carriers
Defect creation
Trapping and releasing of carriers

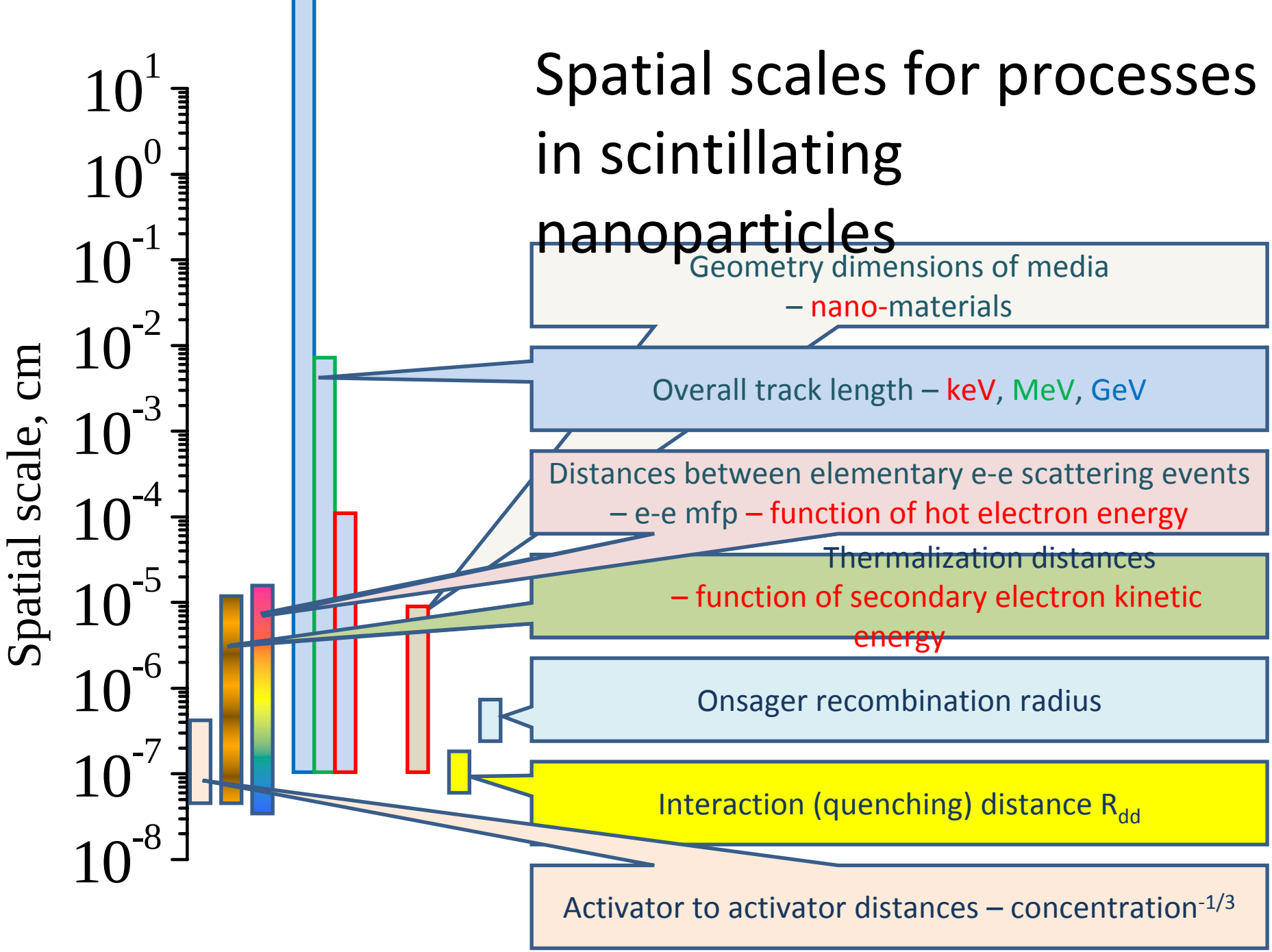
Outline

- Spatial scales for processes in scintillators
- Nanoparticles as scintillators
- Cascade, thermalization and recombination
- Different types of mobilities
- Thermalization length for different types of crystals
- Interconnection of cascade, thermalization and recombination stages in binary iodides
- Why cascade is so effective in CsI?
- Thermalization length and impurities
- Concluding remarks

Spatial scales for processes in scintillators

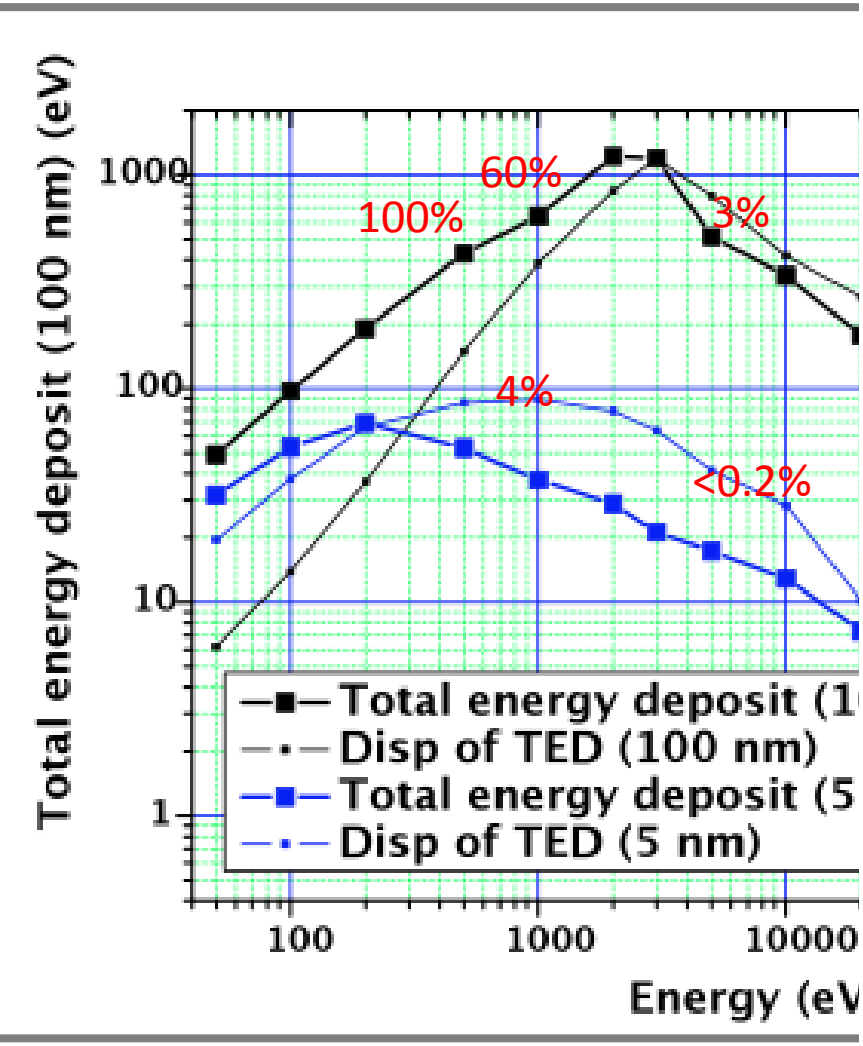


Spatial scales for processes in scintillating nanoparticles



Energy deposited within nanoparticles

Total energy deposited as a function of the energy of the primary electron



Vistovskyy *et al.* J. Appl. Phys. **112**, 024325 (2012)

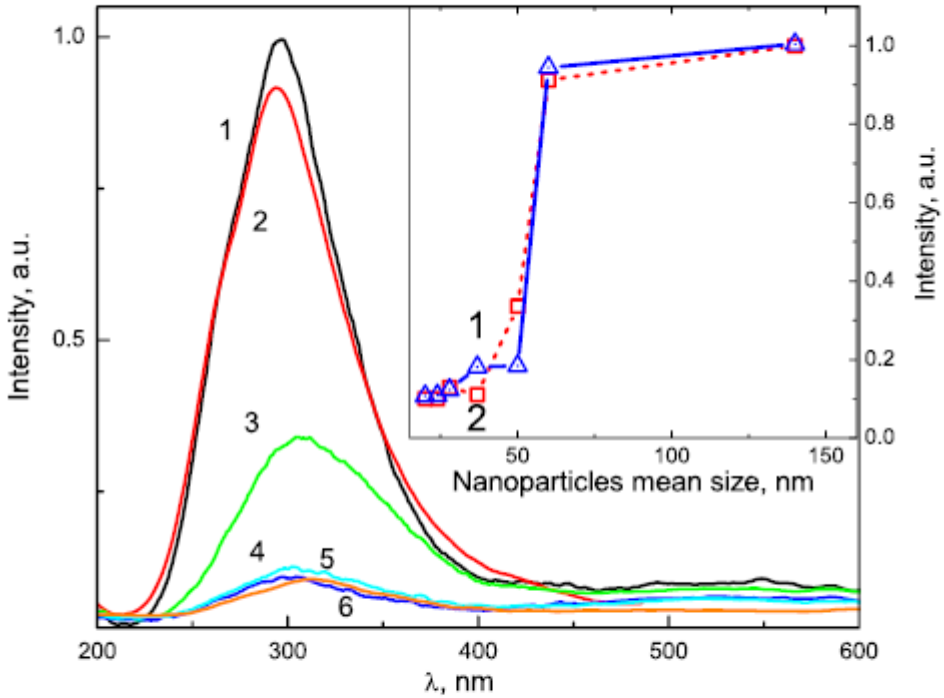


FIG. 6. X-ray excited luminescence spectra of CaF_2 nanoparticles of various size at 300 K. Curves: 1–140; 2–60; 3–50; 4–37; 5–28; 6–20 nm. The dependence of normalized luminescence intensity on the nanoparticle size is shown on inset: curve 1—luminescence intensity upon the excitation by quanta with energy $h\nu_{exc} = 16$ eV, curve 2—X-ray excited luminescence intensity.

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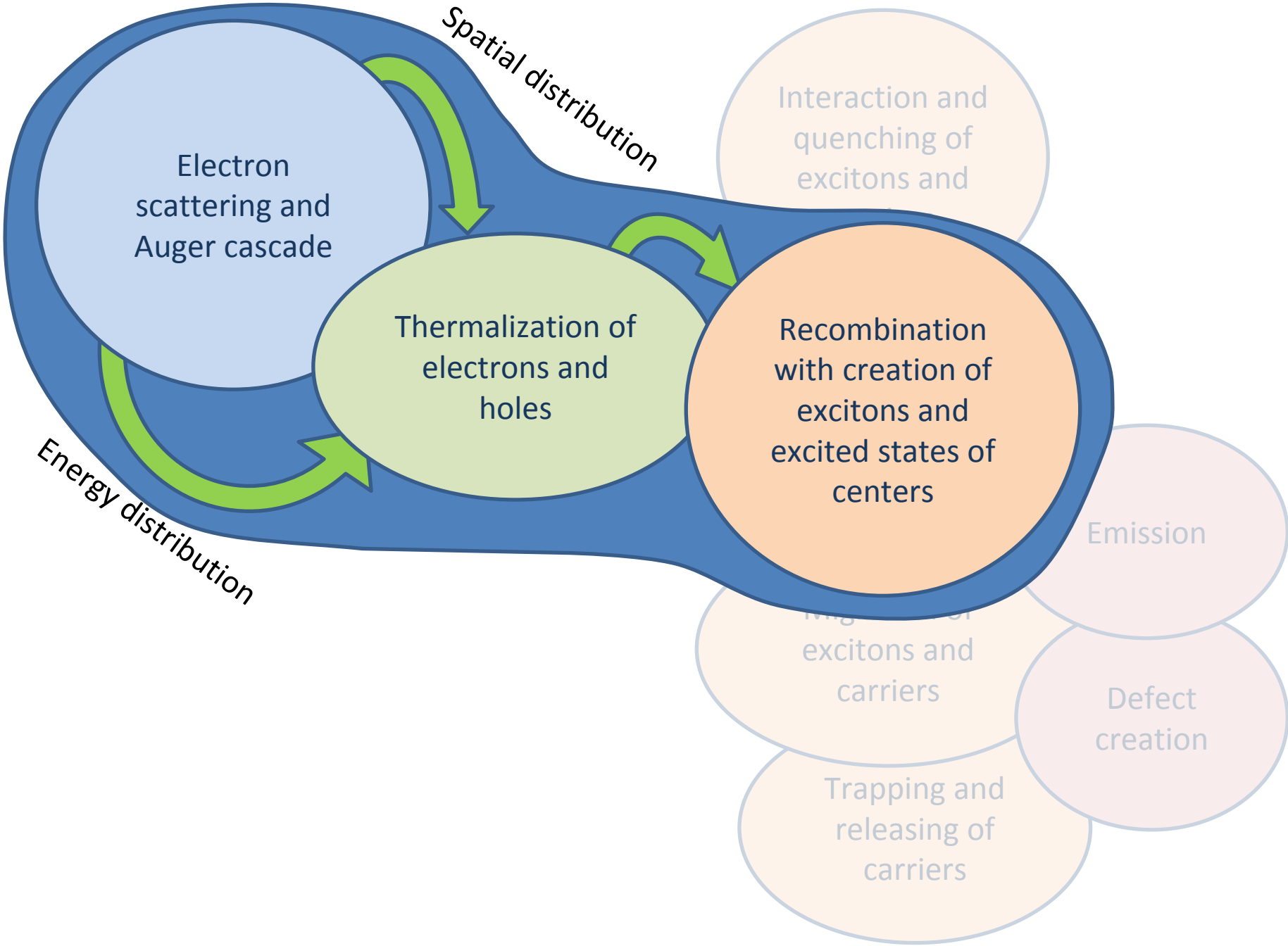
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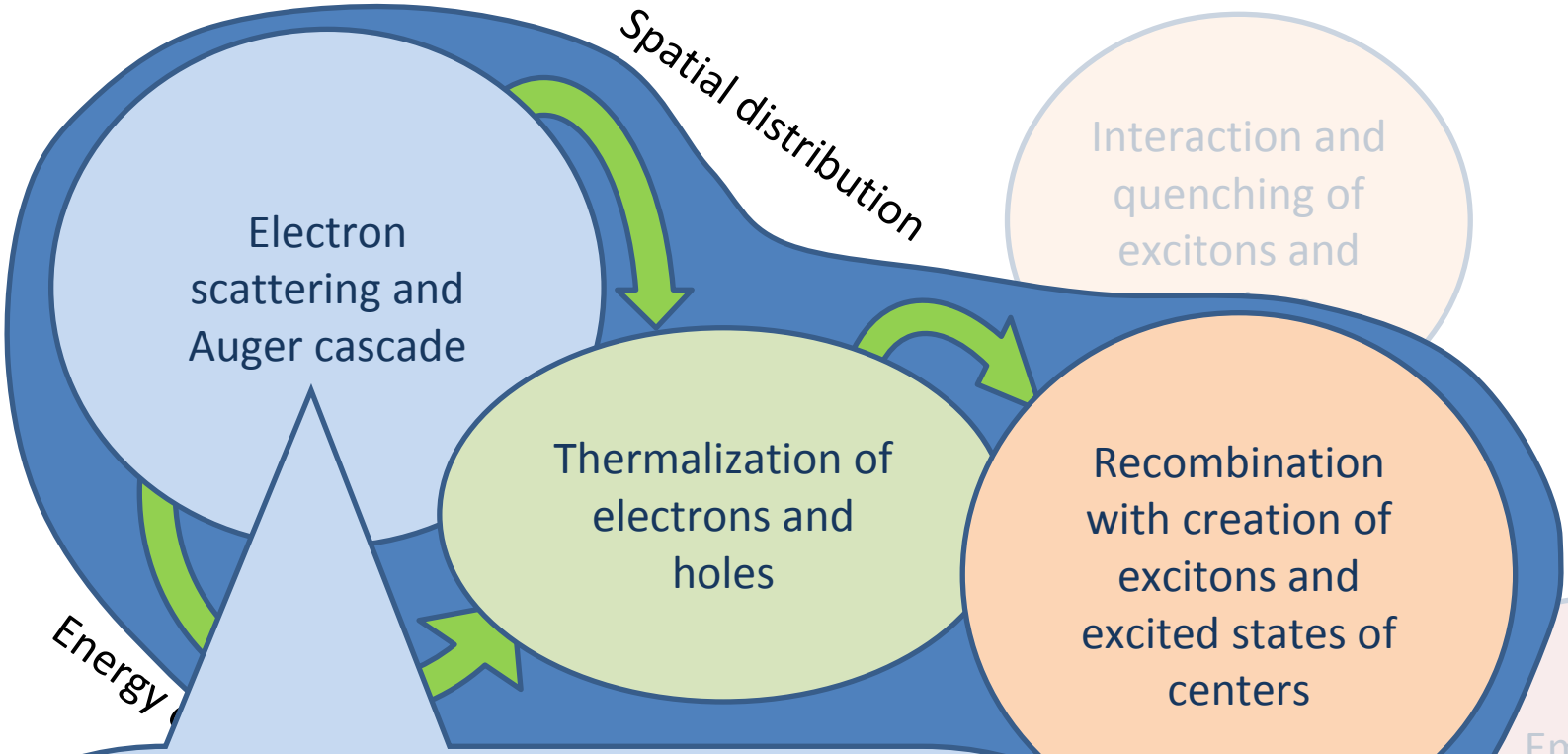


Electrons and holes after cascade of inelastic scatterings are distributed in energy (with kinetic energies below E_g, i.e. the threshold of inelastic e-e scattering) and in space (with characteristic distances equal to e-e mean free paths)

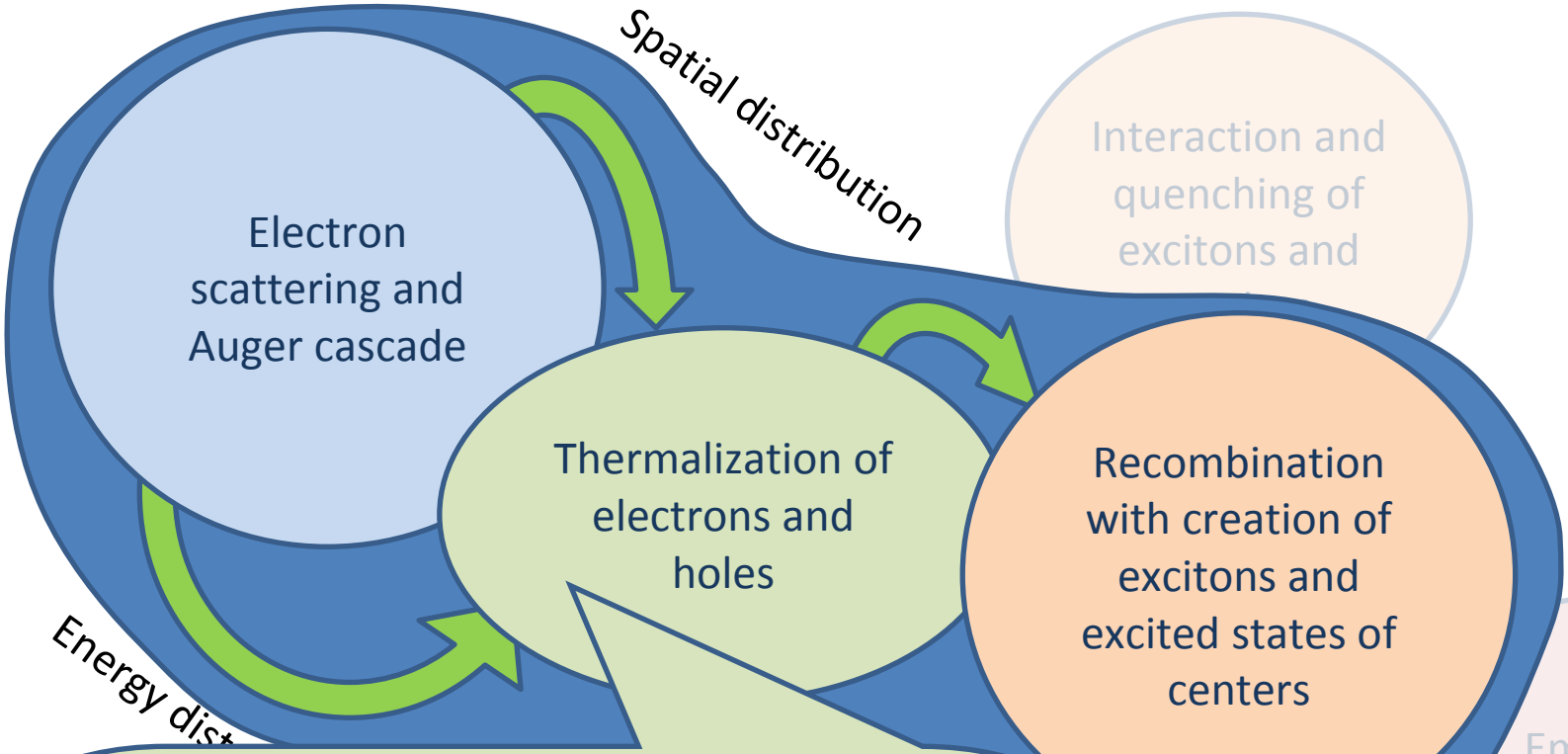
Interaction and quenching of excitons and

Emission

excitons and carriers

Defect creation

Trapping and releasing of carriers



Interaction and quenching of excitons and

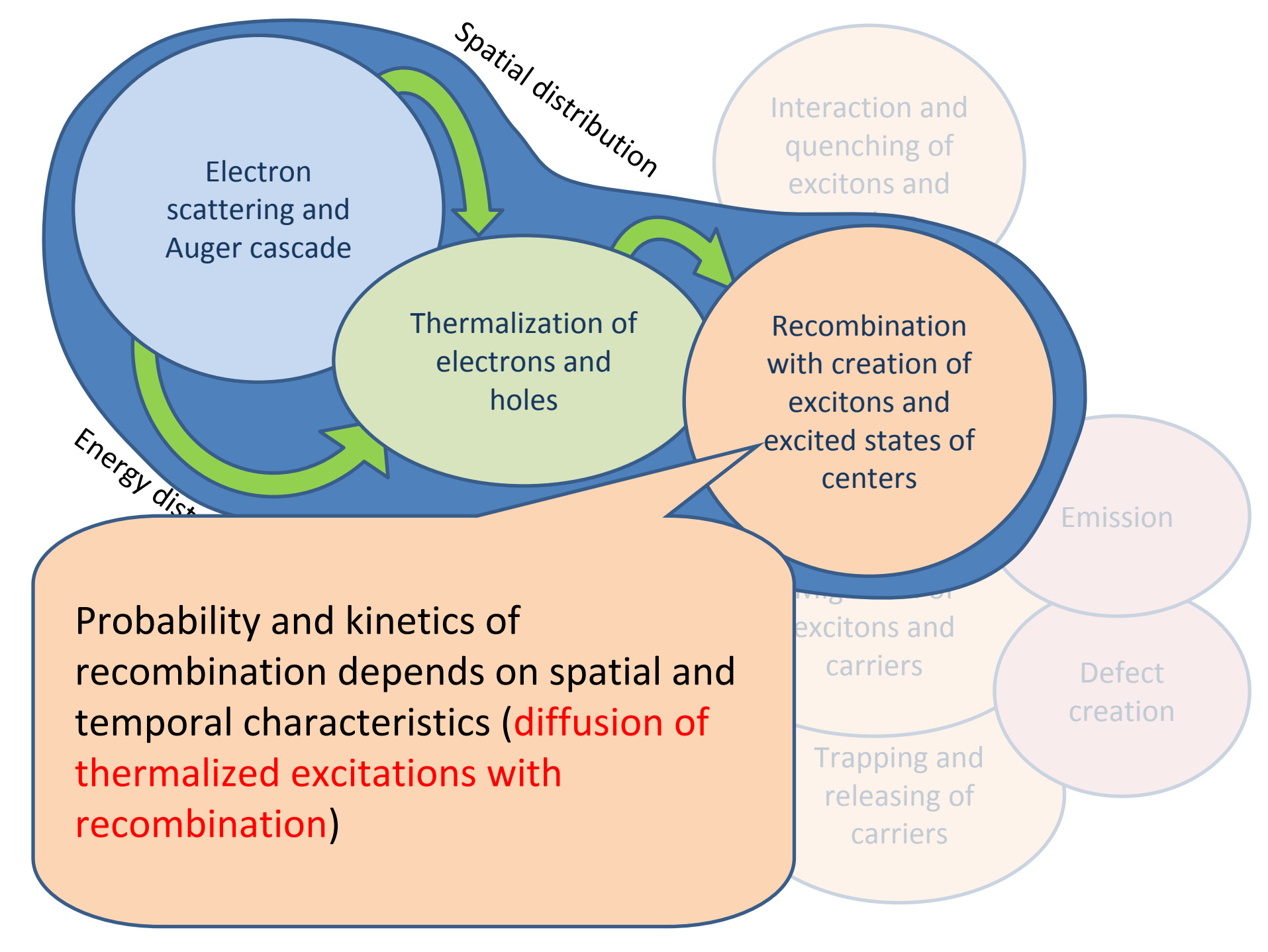
Emission

excitons and carriers

Defect creation

Trapping and releasing of carriers

Electrons and holes are produced as *e-h* pairs; each geminate *e* and *h* are created in the same physical point and then go away from the birthplace during thermalization (thermalization distance, which depends on initial kinetic energy)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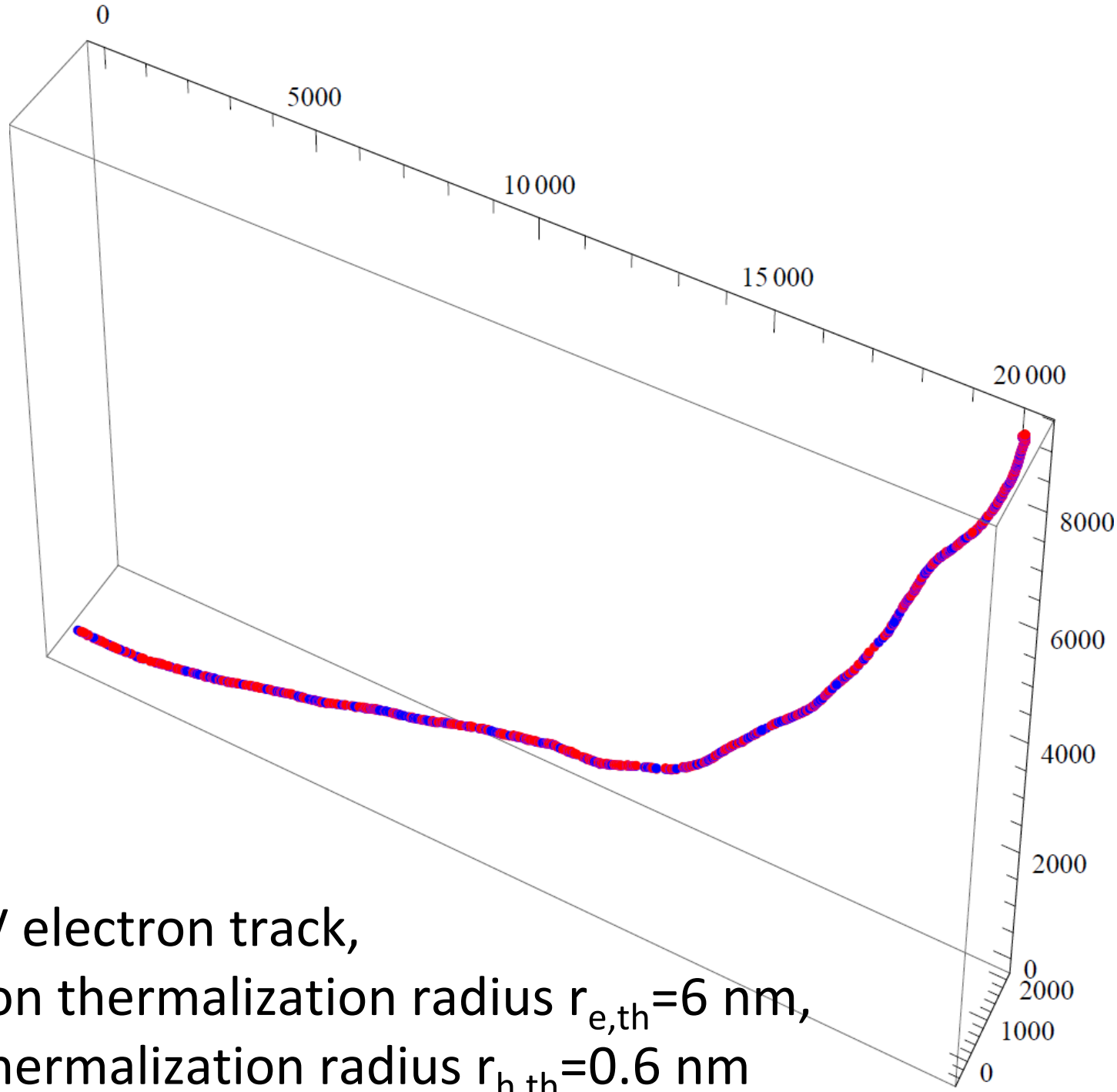
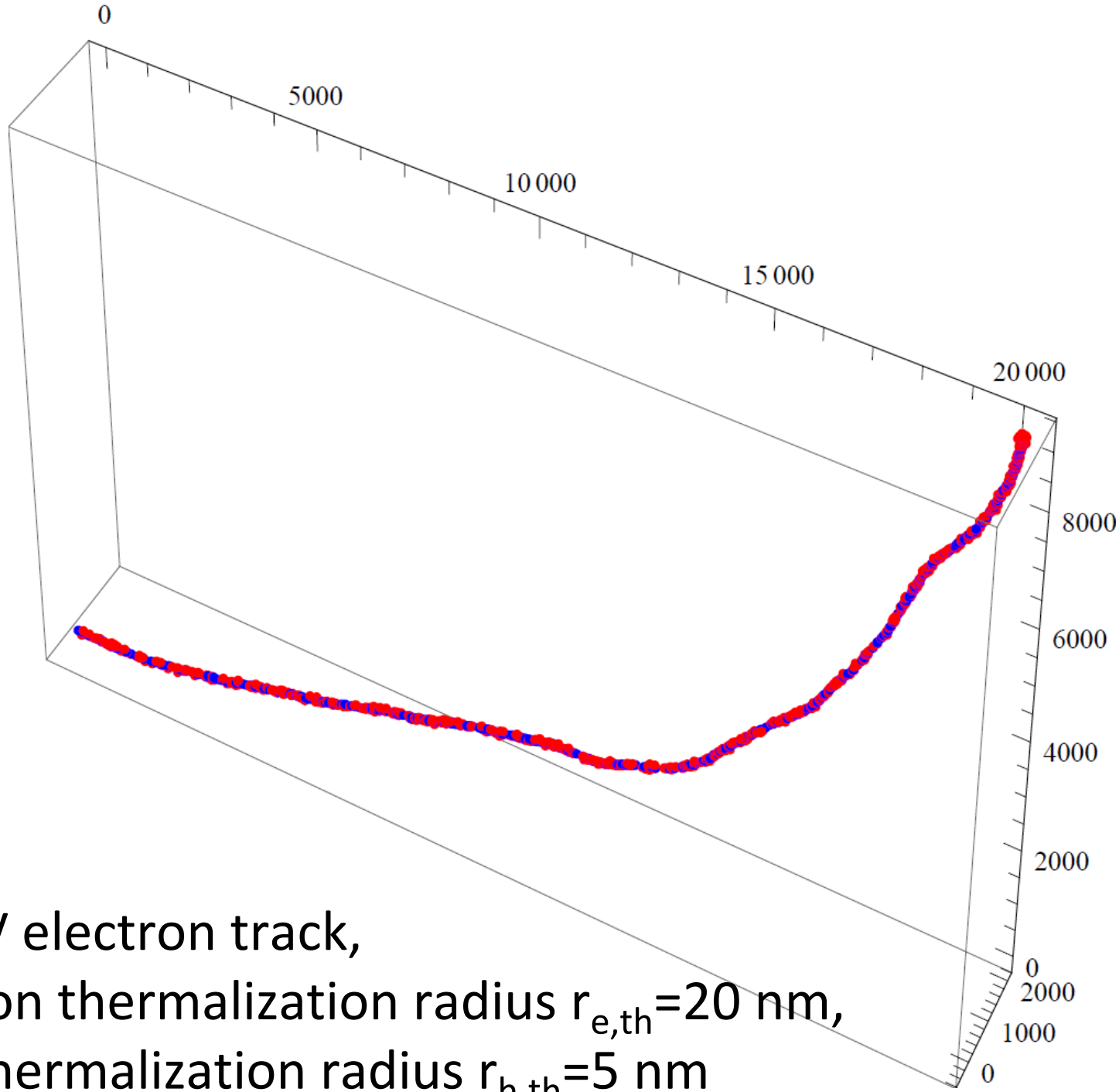
Probability and kinetics of recombination depends on spatial and temporal characteristics (diffusion of thermalized excitations with recombination)

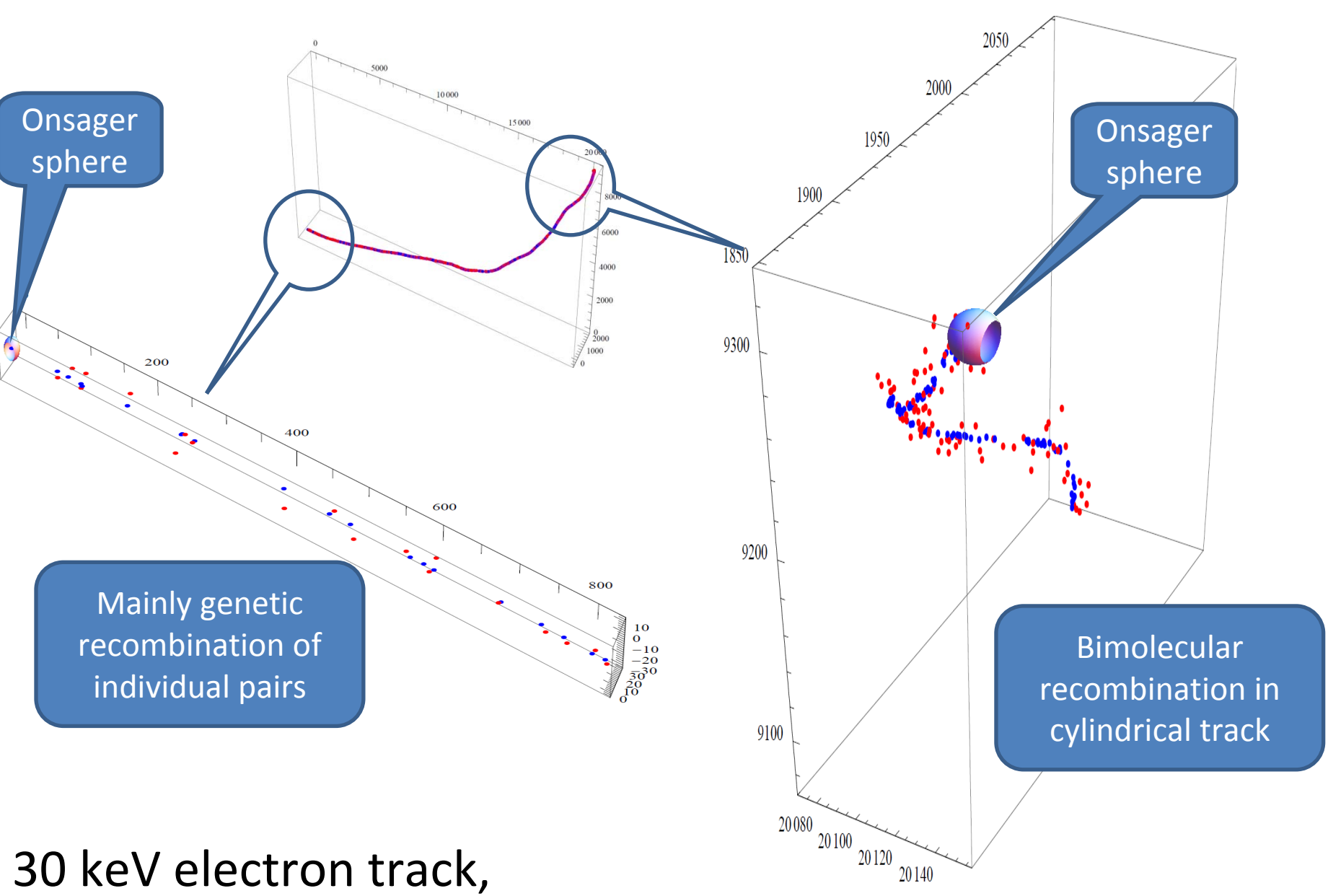
Example of structure of excited region after 30 keV electron passage



30 keV electron track,
electron thermalization radius $r_{e,th}$=6 nm,
hole thermalization radius $r_{h,th}$=0.6 nm



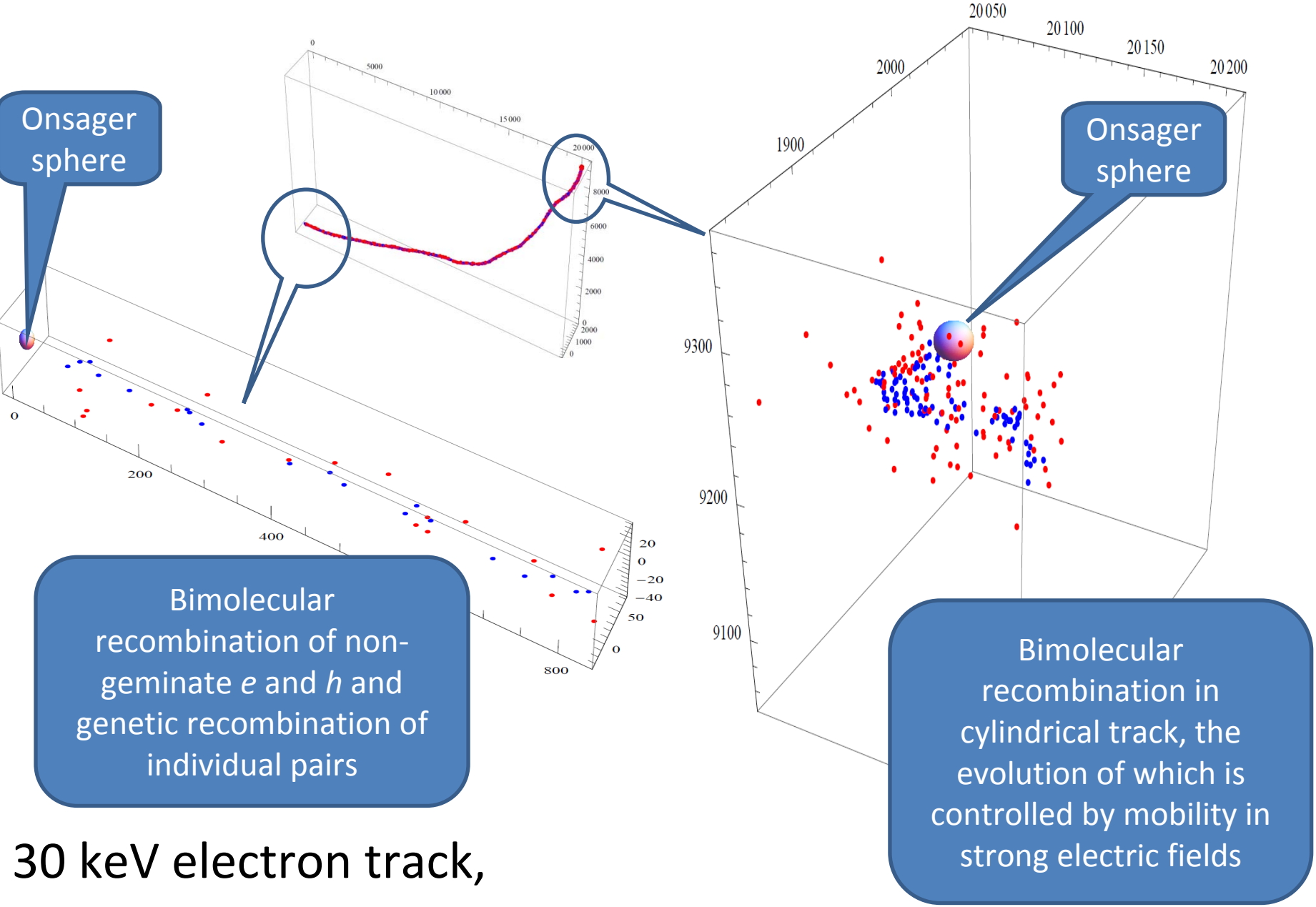
30 keV electron track,
electron thermalization radius $r_{e,th}$=20 nm,
hole thermalization radius $r_{h,th}$=5 nm



30 keV electron track,

$r_{e,th}$=6 nm,

$r_{h,th}$=0.6 nm (red=e, blue=h)



30 keV electron track,

$r_{e,th}$=20 nm,

$r_{h,th}$=5 nm (red=e, blue=h)

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Non-proportionality and mobility

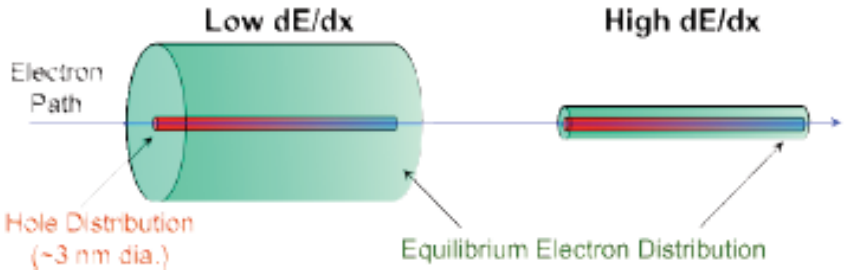

Figure 4. Difference between equilibrium diameters of the electrons (assuming immobile holes) for two different ionizations. Low density is on the left, high density is on the right.

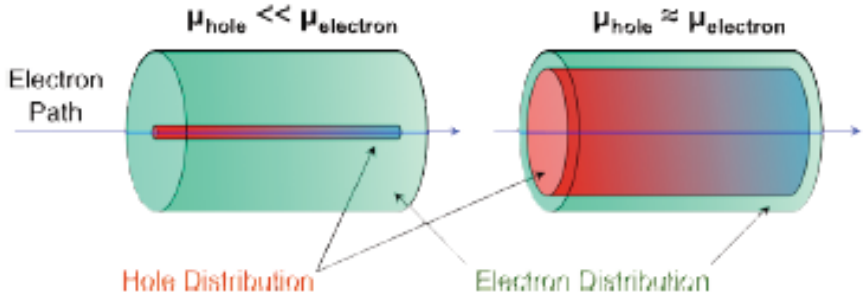

Figure 5. Relative sizes of electron and hole diameters as a function of relative hole mobility. The left side illustrates the distributions when the electron mobility is significantly higher than then hole mobility, while the right side illustrates the distributions when they have similar mobilities.

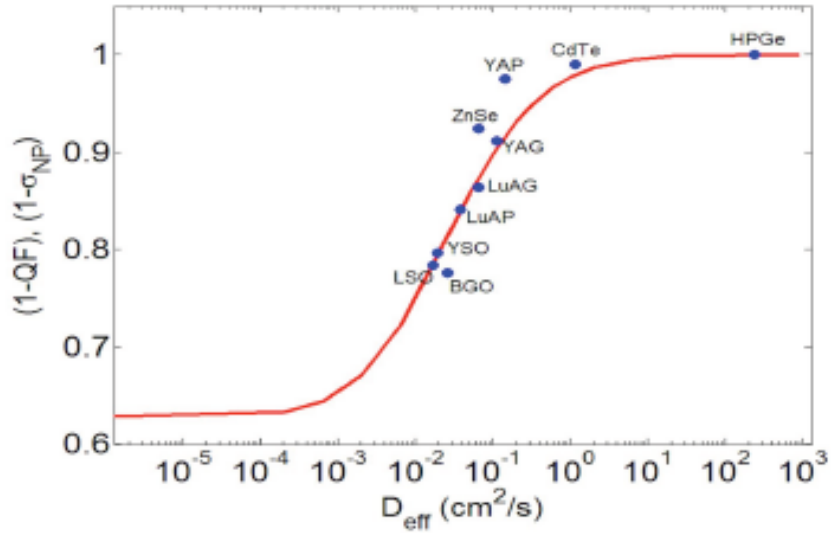

Figure 6. Measured relative light yield at low electron energy for a number of scintillators (solid points) and predicted luminosity (solid curve) as a function of diffusion coefficient. Reprinted with permission from [61]. See text for definitions.

W. W. Moses, G. A. Bizarri, R. T. Williams, S. A. Payne, A. N. Vasil'ev, J. Singh, Q. Li, J. Q. Grim, and W–S. Choong, *The Origins of Scintillator Non-Proportionality*, IEEE Transactions on Nuclear Science, vol. 59, issue 5, pp. 2038-2044 (2012)

Spatial distribution of electrons, holes and excitons due to mobility in e-e passive energy domain

- Two types of carrier mobilities: thermalization length (mobility of hot electrons and holes) and mobility of thermalized excitations (electrons, holes & excitons).
- High-energy part of ionization track – individual electron-hole pairs and small non-overlapping clusters of excitations. Negative role of mobility: the higher the thermalization length (in comparison with Onsager radius), the lower the recombination yield (HPGe – the limiting case of high mobility w/o any luminescence).
- Low-energy part of ionization track – overlapping clusters of excitations. Mean distance between interacting excitations increases with increase of the mobility of excitons. Positive role of mobility: the higher the mobility, the lower the quenching of excitation due to high EE density.
- "Ideal" scintillator: Low hot mobility (high yield of excitons) and high thermalized mobility (low interaction).

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Coupled processes of thermalization and spatial diffusion

Four main functions which characterized spatial diffusion and thermalization:

(1) rate of electron-phonon scattering (inverse lifetime) $\tau^{-1}\left(E_e^{kin}\right)$

(2) mean free path $\lambda\left(E_e^{kin}\right)=v\left(E_e^{kin}\right)\tau\left(E_e^{kin}\right)$

(3) spatial diffusion coefficient $D^R\left(E_e^{kin}\right)=\frac{1}{3}v^2\left(E_e^{kin}\right)\tau\left(E_e^{kin}\right)$ and equation for Brownian motion

$$\frac{d<r^2>}{dt}=6D^R\left(E_e^{kin}\right)$$

(4) energy relaxation rate $S\left(E_e^{kin}\right)=D^E\left(E_e^{kin}\right)/k_BT$ and energy relaxation equation

$$\frac{dE_e^{kin}}{dt}=-S\left(E_e^{kin}\right)$$

$$\frac{d<r^2>}{dE_e^{kin}}=-6\frac{D^R\left(E_e^{kin}\right)}{S\left(E_e^{kin}\right)}$$

Coupled processes of thermalization and spatial diffusion

Mean square of the thermalization distance $$< r^2 >_{E_{e0} \to E_e^{kin}} = 6 \int_{E_e^{kin}}^{E_{e0}} \frac{D^R(E')}{S(E')} dE'$$

Spatial distribution function $$f\left(r, l_e(E_{e0})\right) = \frac{3\sqrt{6}\, r^2}{\sqrt{\pi}\, l_e^3(E_{e0})} \exp\left(- \frac{3r^2}{2 l_e^2(E_{e0})} \right)$$

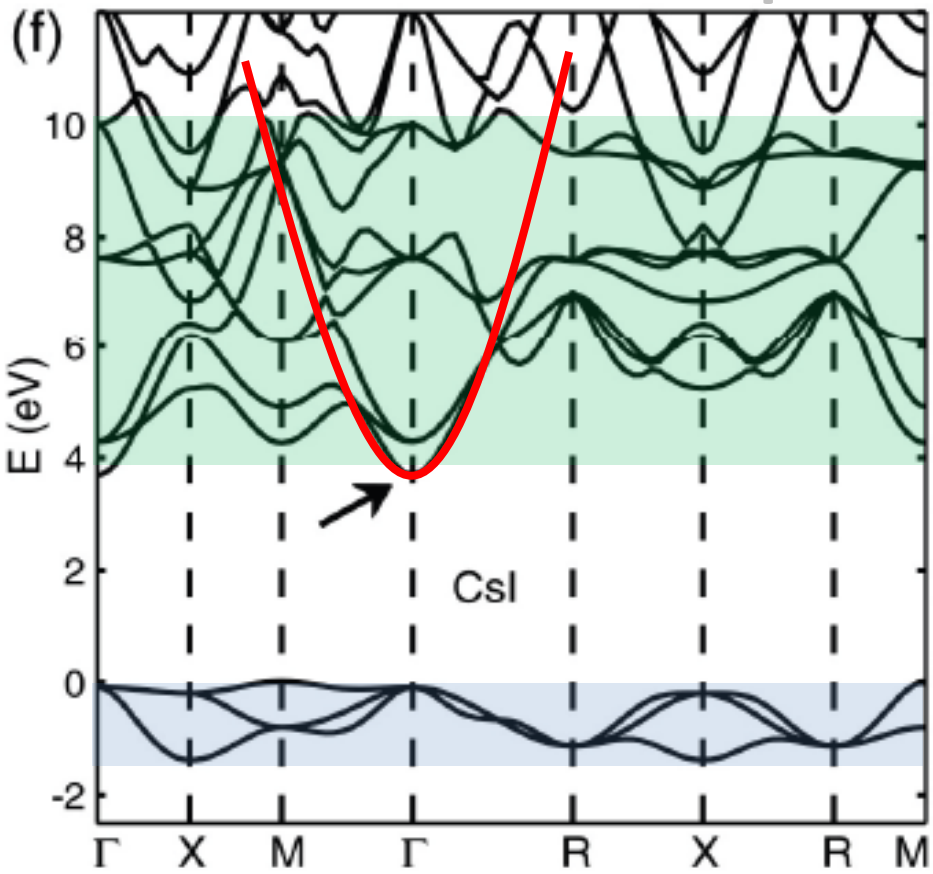
where thermalization length is $$l_e(E_{e0}) = \sqrt{< r^2 >_{E_{e0} \to k_B T}}$$

Thermalization length for one LO phonon branch

$$l_{e,LO}^2(E_{e0}) = \frac{8}{3} a_B^2 \left(\frac{\tilde{\varepsilon}}{m_e^*/m_0} \right)^2 \tanh\left(\frac{\hbar\Omega_{LO}}{2k_B T} \right) \int_{\hbar\Omega_{LO}}^{E_{e0}} \left(\frac{E'}{\hbar\Omega_{LO}} \right)^2 \frac{1}{\ln\left(4E'/\hbar\Omega_{LO}\right)} \frac{dE'}{\hbar\Omega_{LO}}$$

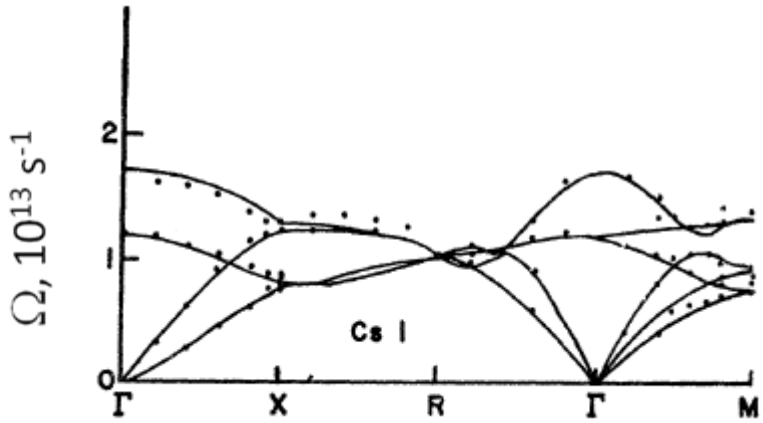
$$= \frac{1}{24} a_B^2 \left(\frac{\tilde{\varepsilon}}{m_e^*/m_0} \right)^2 \tanh\left(\frac{\hbar\Omega_{LO}}{2k_B T} \right) \mathrm{Ei}\left(3\ln\left(\frac{4E_{e0}}{\hbar\Omega_{LO}} \right) \right),$$

CsI band structure and phonon dispersion



Band structure calculations

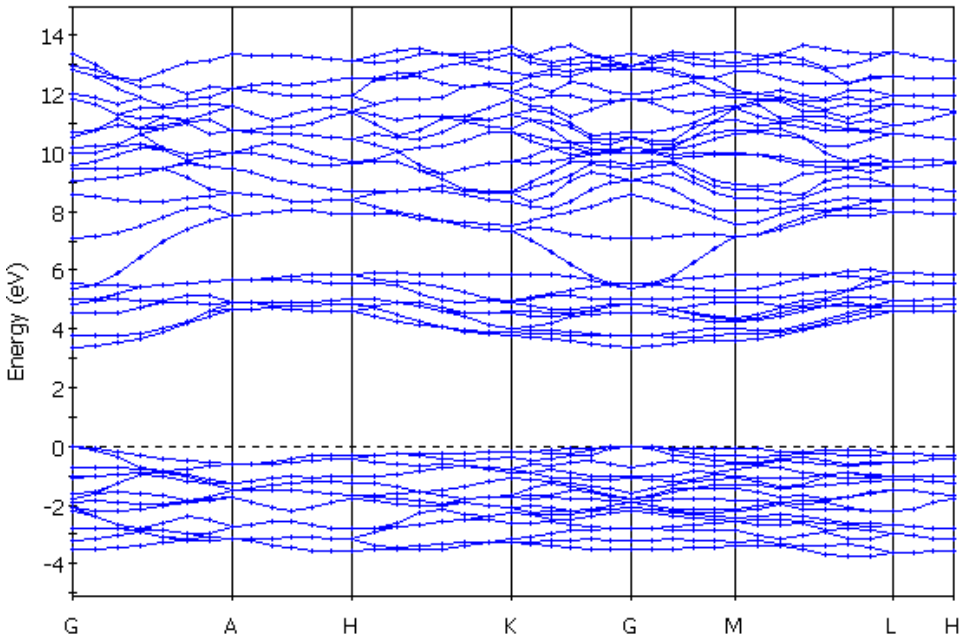
from W. Setyawan, R. M. Gaume et al. *IEEE TNS*, 2009



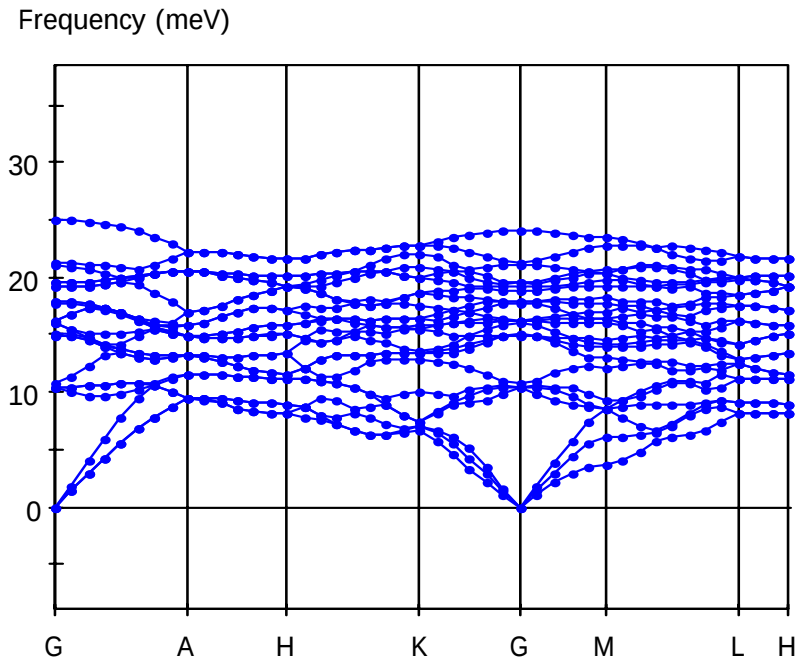
J.F. Vetelino, K. V. Namjoshi and S. S. Mitra, Phys. Rev. B 7, 4001–4004 (1973)

$LaBr_3$ band structure (w/o La4f) and phonon dispersion

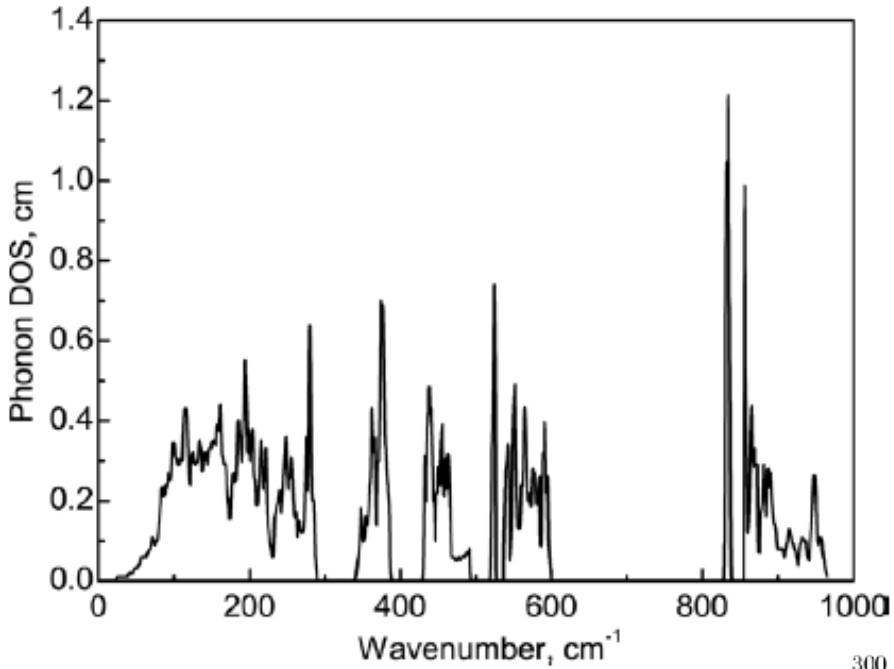
I.Iskandarova, private communication

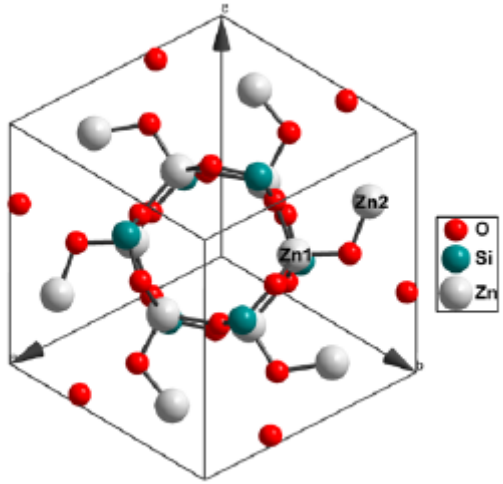


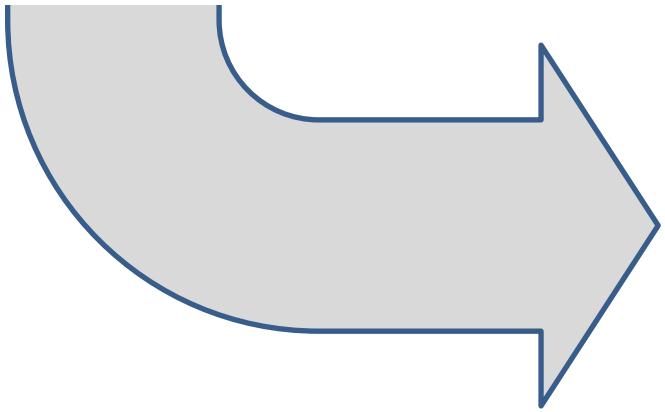
Band structure for $LaBr_3$ (without La f states). Energy scale is shifted to the top of the valence band.

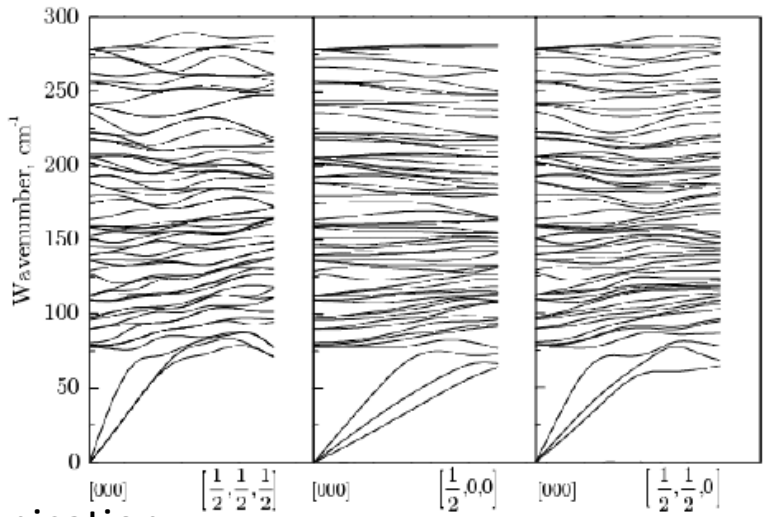


Zn_2SiO_4 (42 atoms per unit cell)









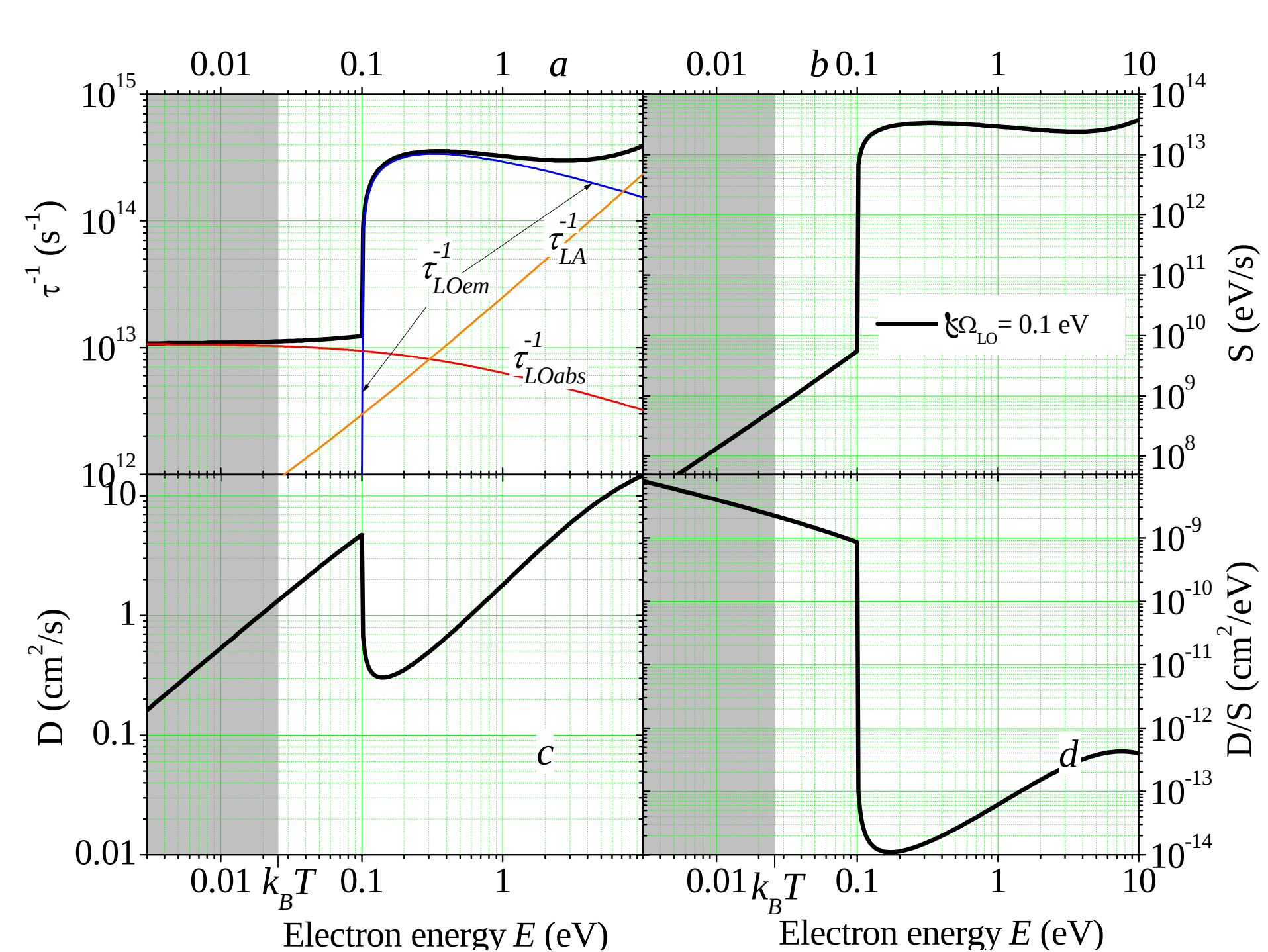
M.Kretov, A.Sherbinin, private communication

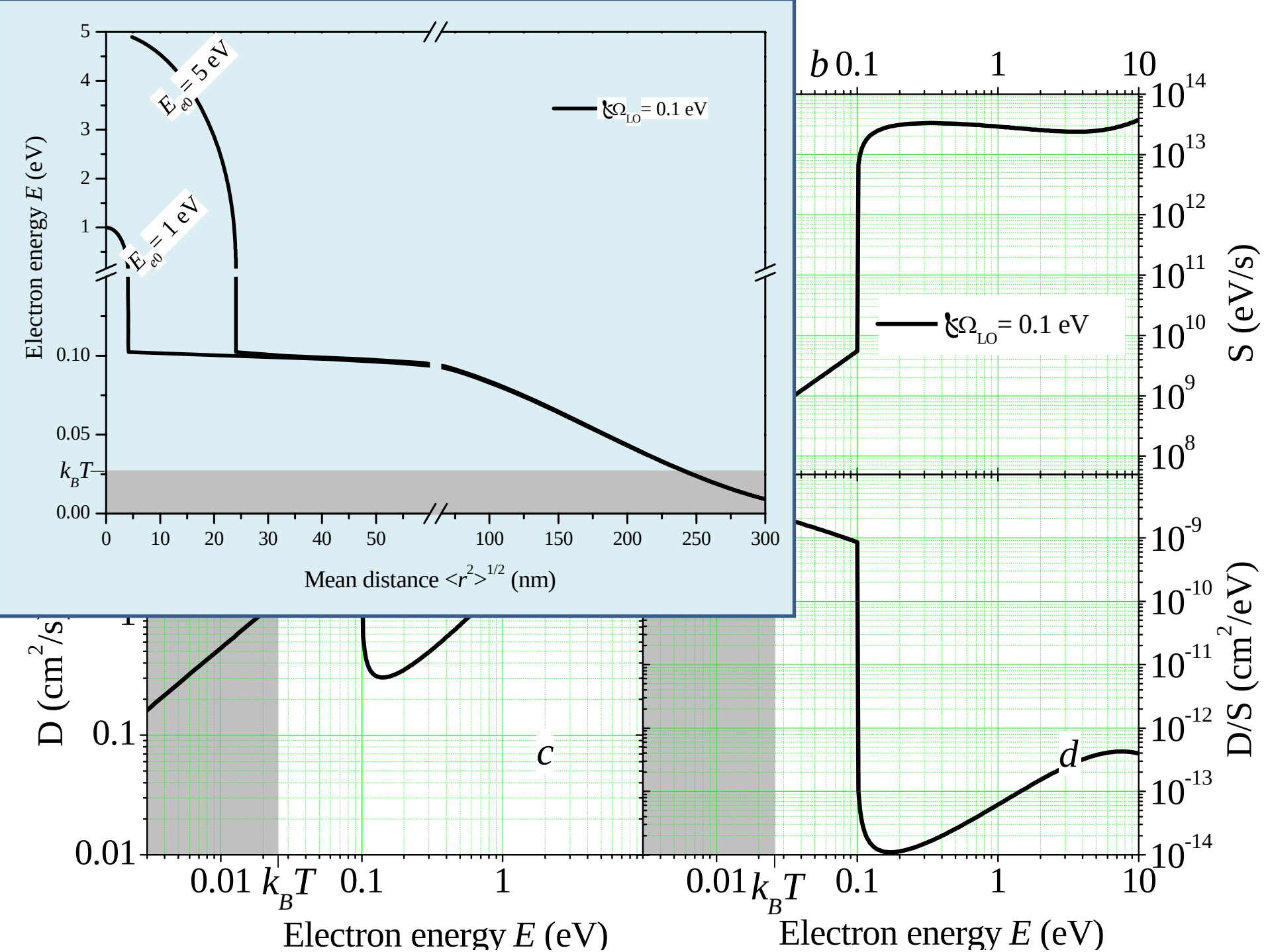
Different cases to discuss

R.Kirkin, V.V. Mikhailin, and A.N. Vasil'ev, *Recombination of correlated electron-hole pairs with account of hot capture with emission of optical phonons*, IEEE Transactions on Nuclear Science, vol. 59, issue 5, pp. 2057-2064 (2012)

- Simple oxide or fluoride (one LO branch)

$$\hbar\Omega_{LO} = 0.1\,eV > k_B T = 300K = 0.026\,eV$$


a
b
c
d
τ^{-1} (s^{-1})
τ^{-1}_{LOem}
τ^{-1}_{LA}
τ^{-1}_{LOabs}
$\hbar\Omega_{LO}$ = 0.1 eV
S (eV/s)
D (cm^2/s)
D/S (cm^2/eV)
k_BT
Electron energy E (eV)
Electron energy E (eV)


Electron energy E (eV)
E_e0 = 5 eV
E_e0 = 1 eV
ħΩ_LO = 0.1 eV
k_BT
Mean distance <r^2>^1/2 (nm)
b
S (eV/s)
c
D (cm^2/s)
d
D/S (cm^2/eV)
Electron energy E (eV)
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Different cases to discuss

R.Kirkin, V.V. Mikhailin, and A.N. Vasil'ev, *Recombination of correlated electron-hole pairs with account of hot capture with emission of optical phonons*, IEEE Transactions on Nuclear Science, vol. 59, issue 5, pp. 2057-2064 (2012)

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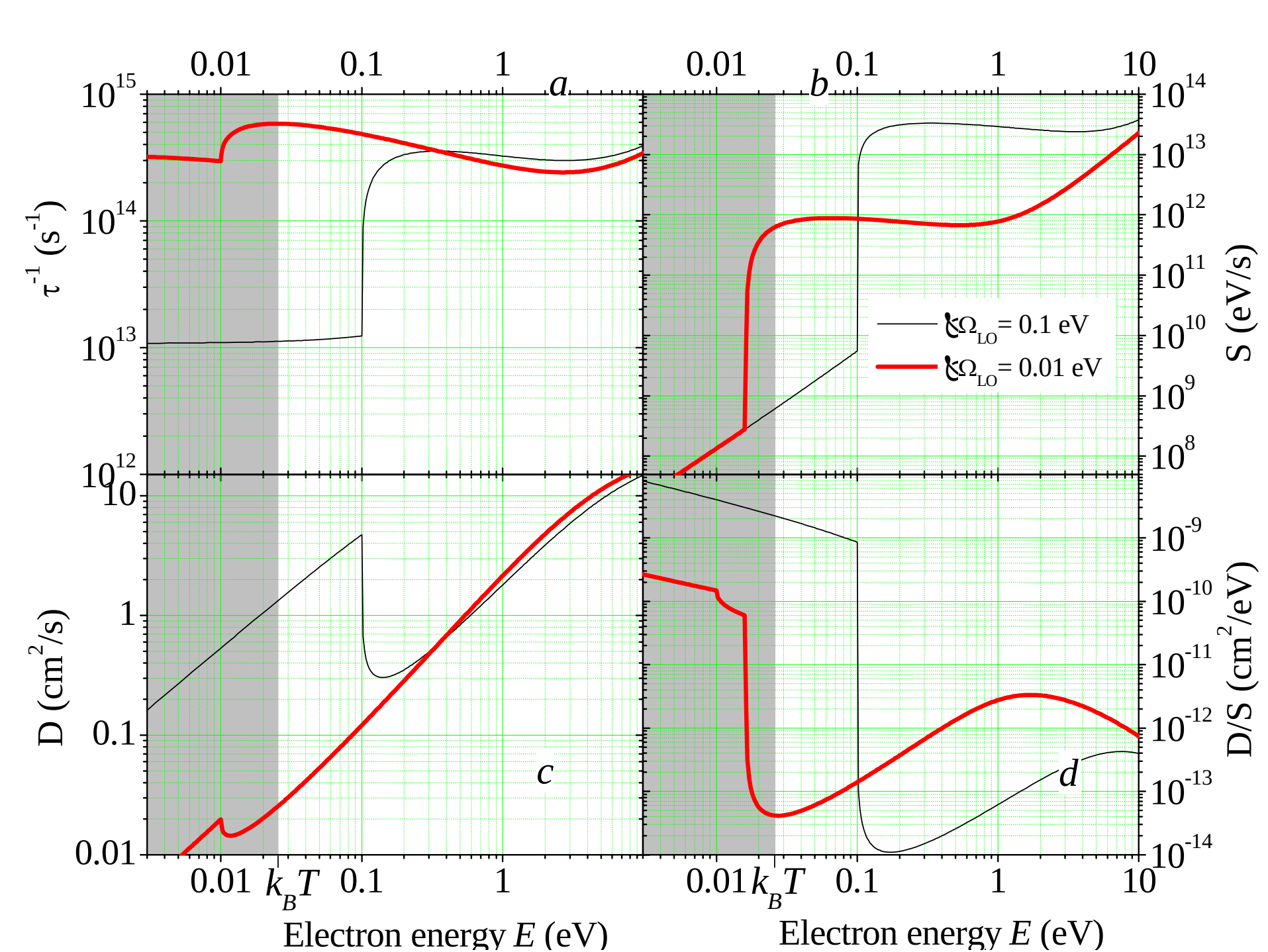
$$\hbar\Omega_{LO} = 0.01\,eV < k_B T$$


a
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d
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D (cm^2/s)
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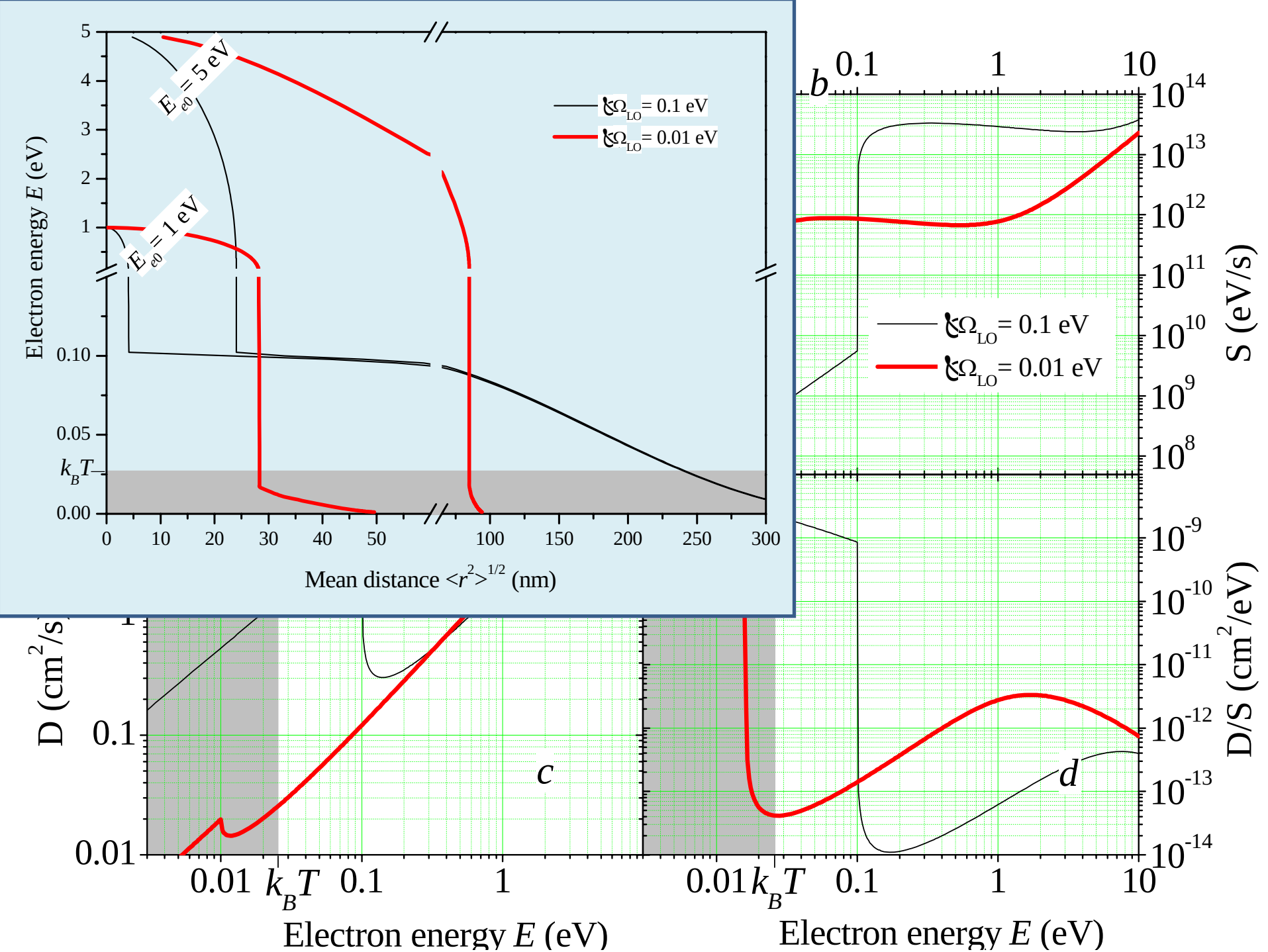
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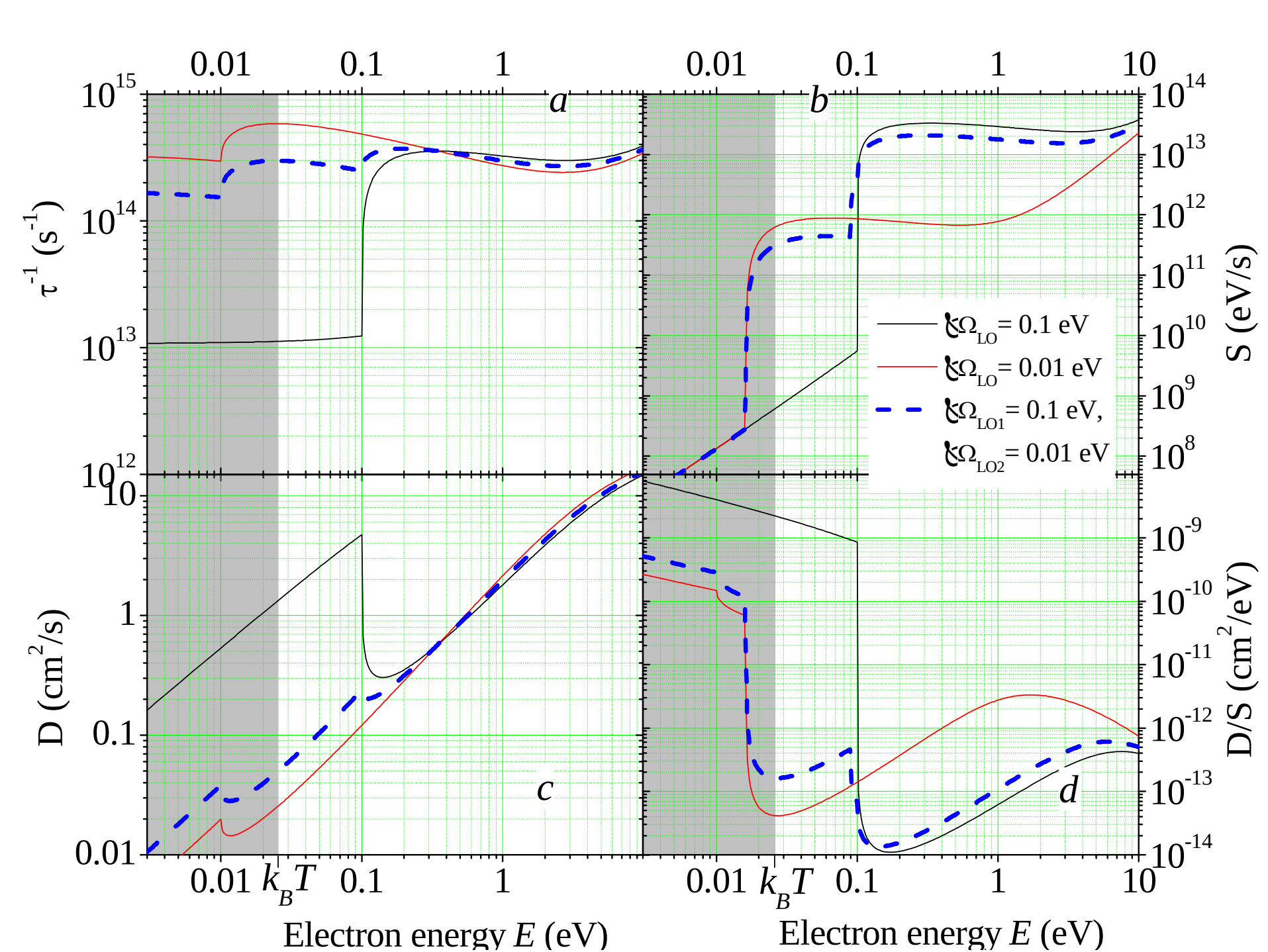
$$\hbar\Omega_{LO1} = 0.1\,eV,\ \hbar\Omega_{LO2} = 0.01\,eV$$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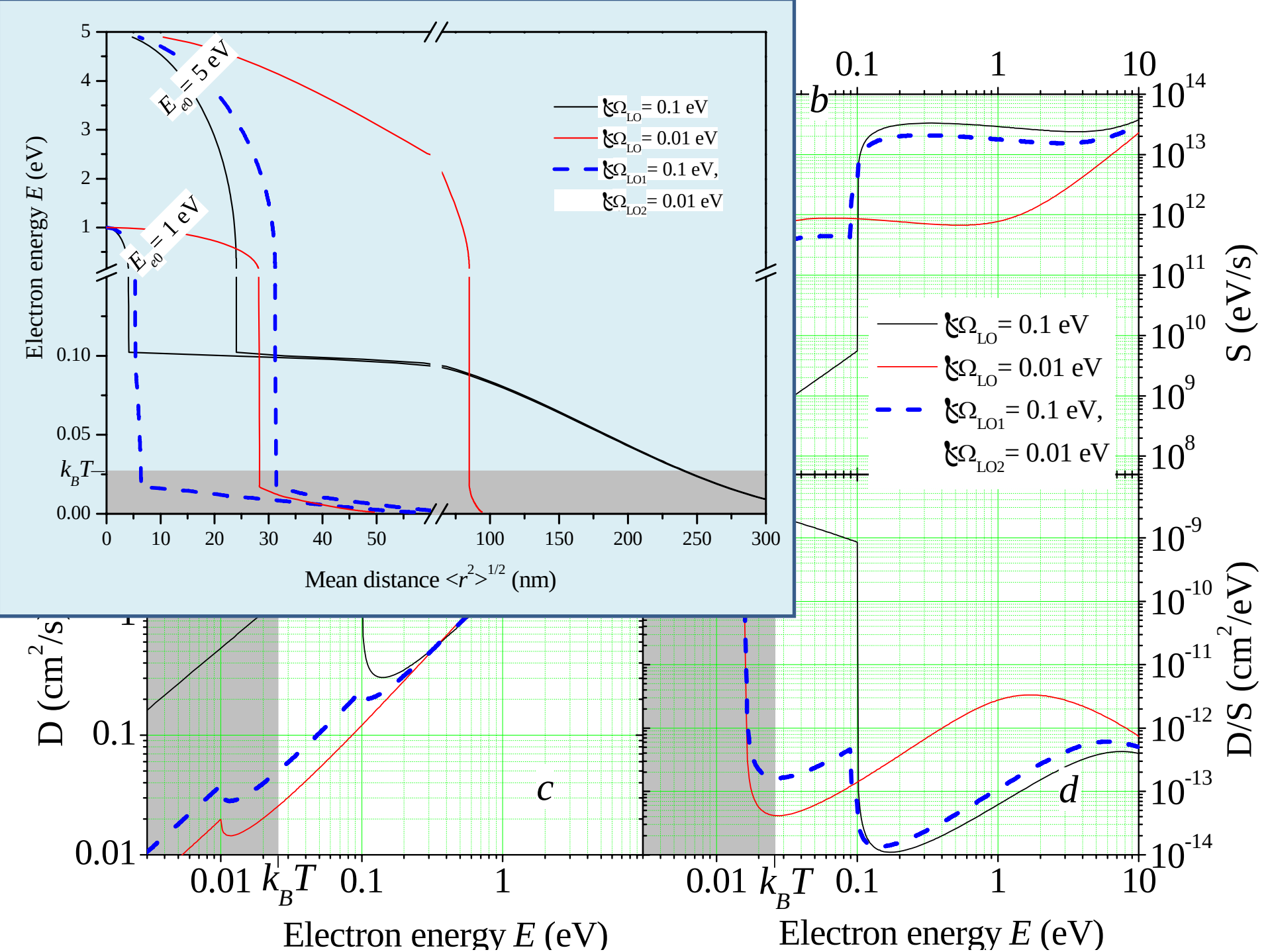
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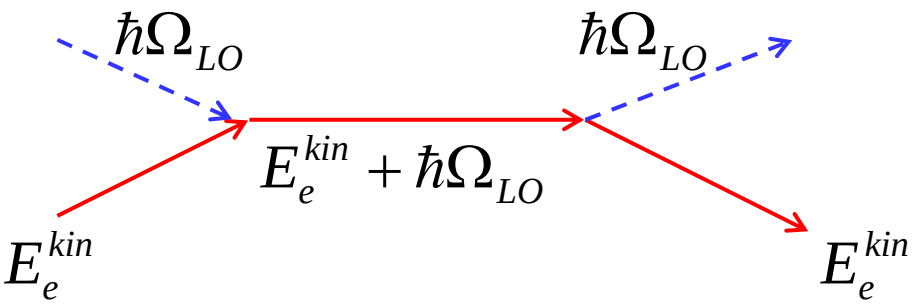
$$\hbar\Omega_{LO1} = 0.1\,eV,\ \hbar\Omega_{LO2} = 0.01\,eV$$

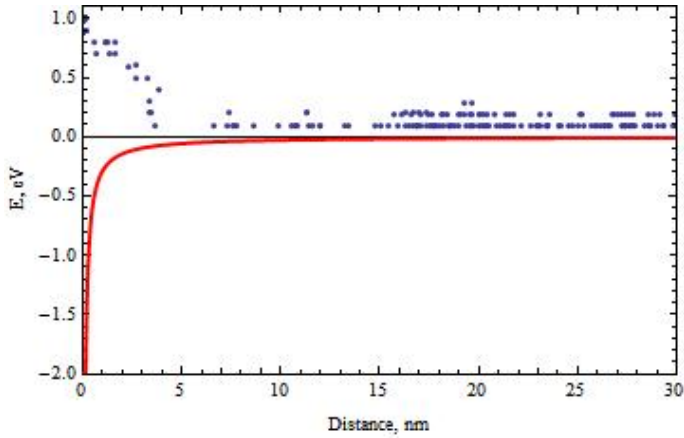
- 2 LO branches with close energies

$$\hbar\Omega_{LO1} = 0.1\,eV,\ \hbar\Omega_{LO2} = 0.08\,eV$$

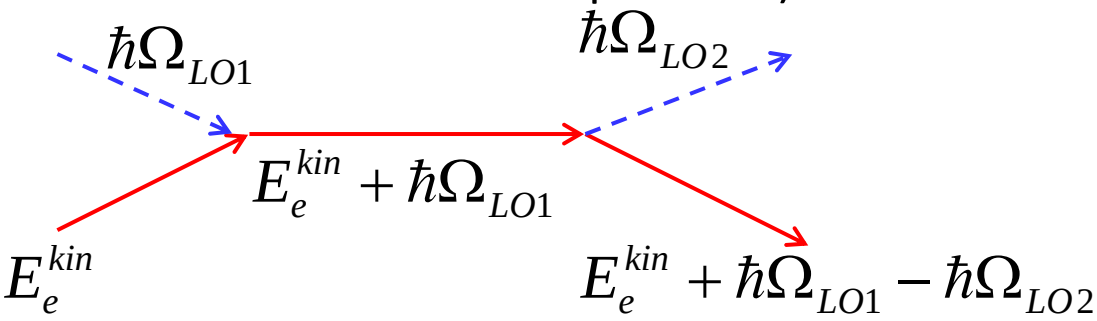
Interaction with LO phonons in LO-passive region

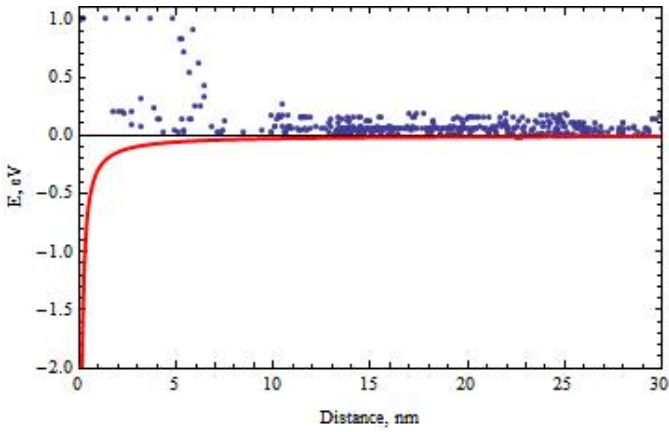
- One LO branch (2 atoms/unit cell) – Spatial diffusion is due to LO phonons, and energy relaxation is due to LA phonons)

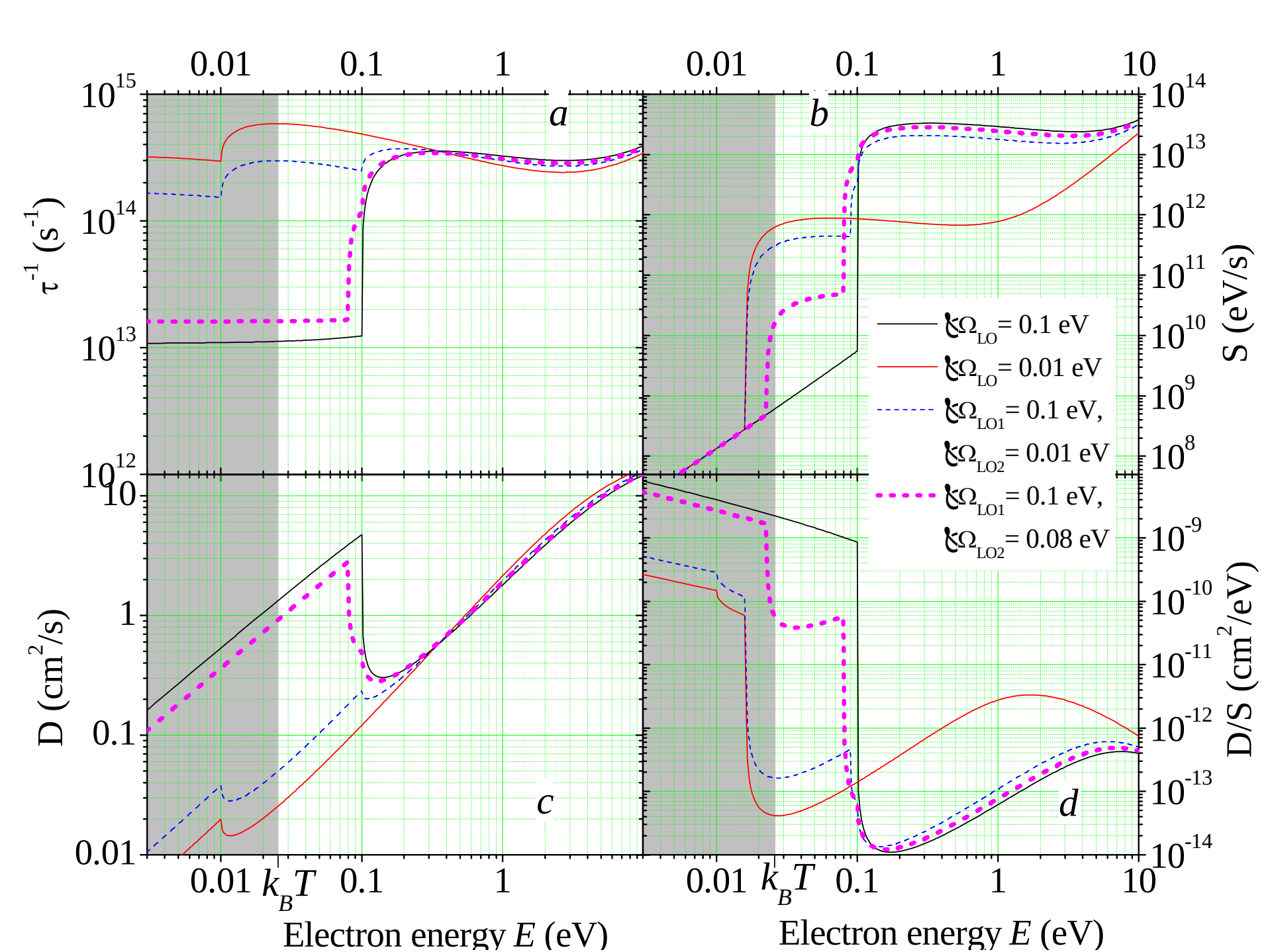




Few LO branches (N atoms/cell → (N-1) PLO) (Both spatial diffusion and relaxation are due to LO phonons)






a
b
c
d
τ^{-1} (s^{-1})
S (eV/s)
D (cm^2/s)
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Electron energy E (eV)
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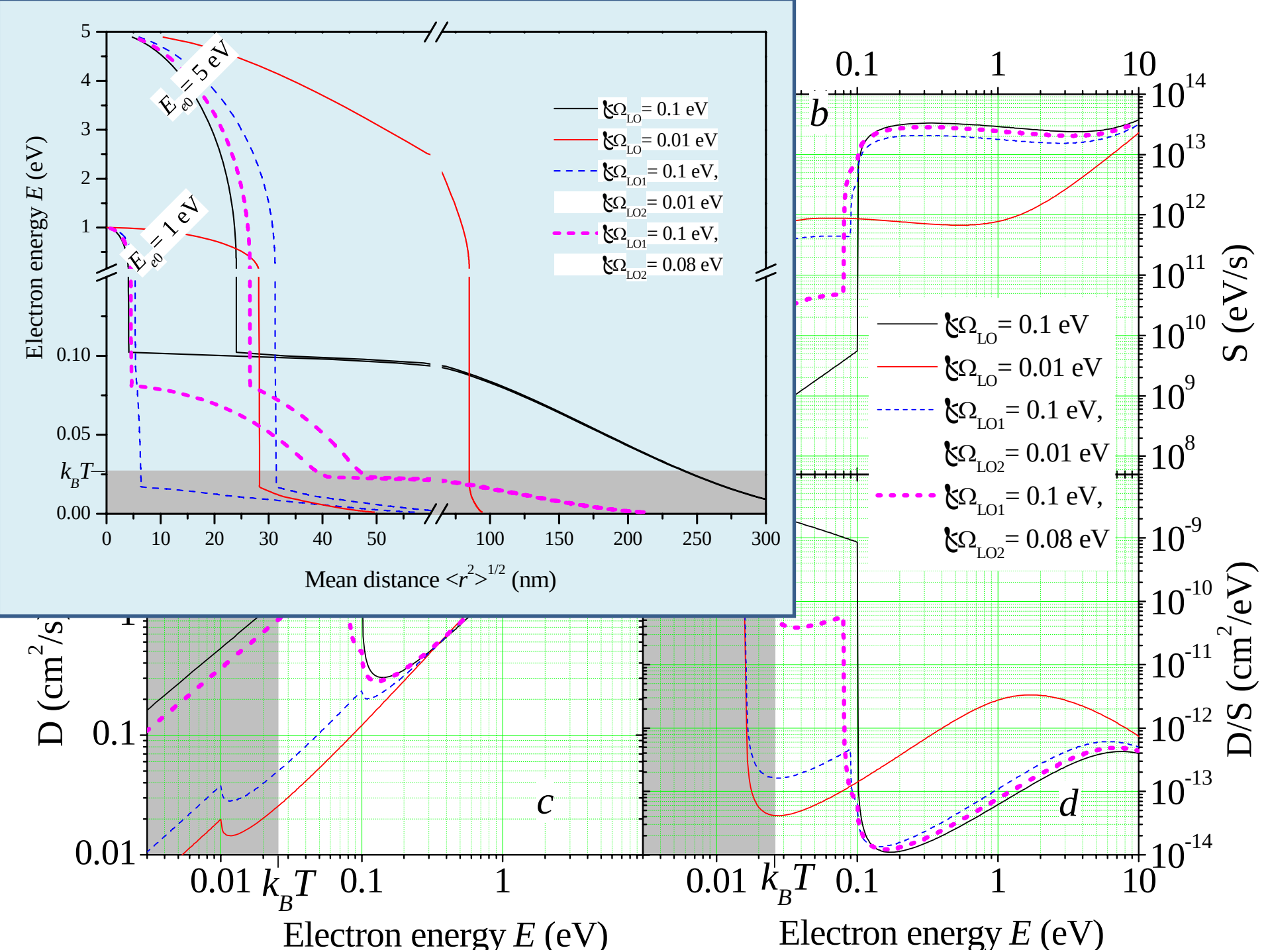
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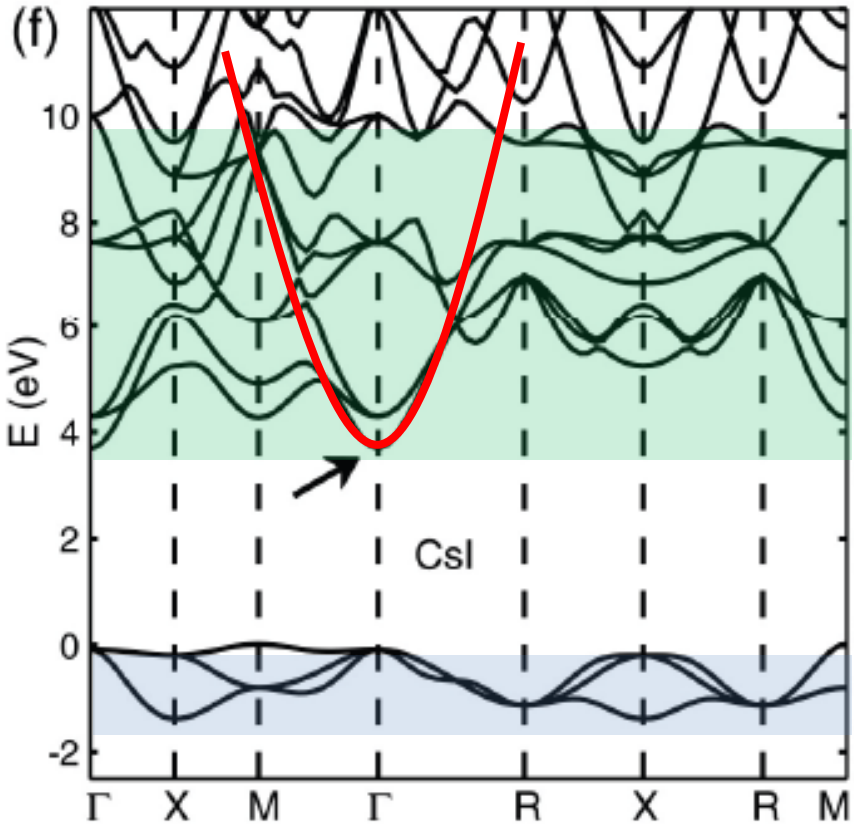
Starting states for thermalization ($E_{kin}<E_g$)

Band structure calculations

from W. Setyawan, R. M. Gaume et al. *IEEE TNS*, 2009



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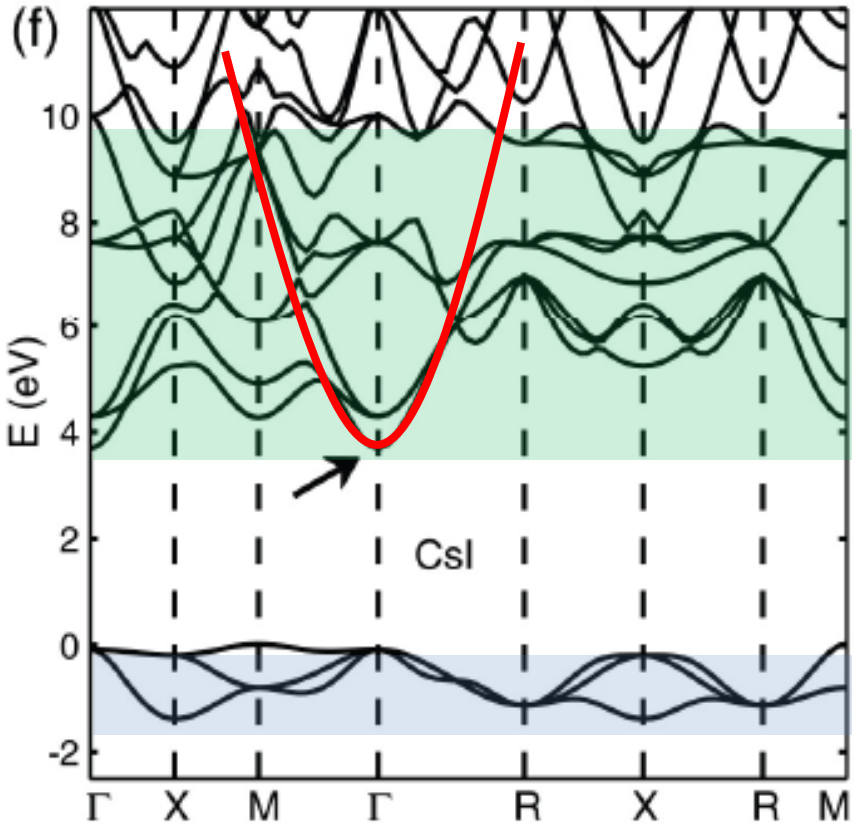
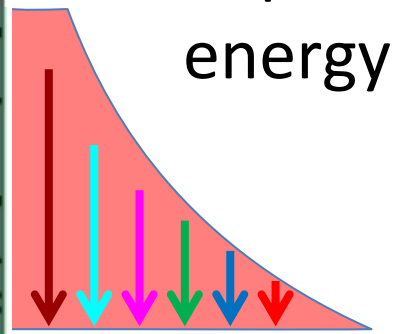
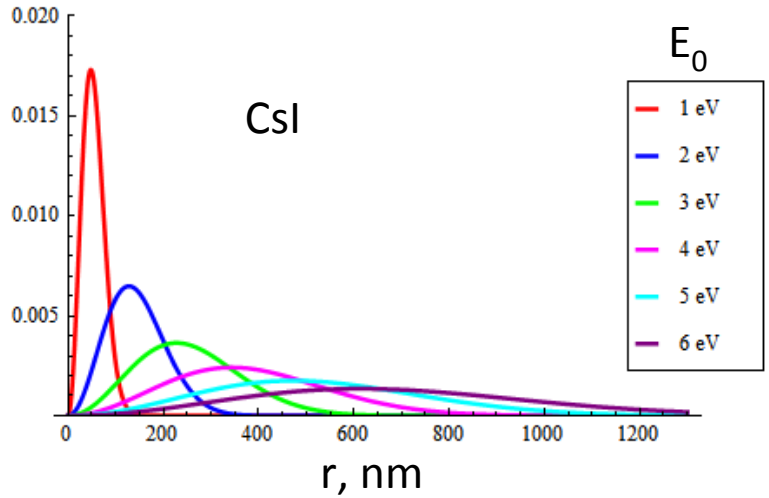
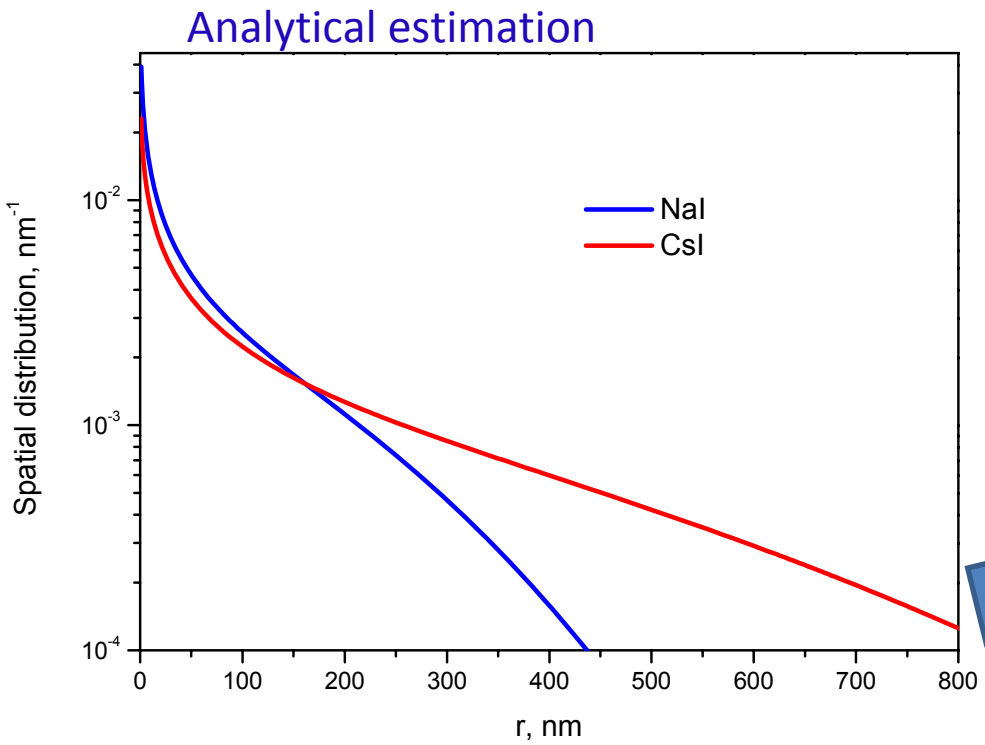




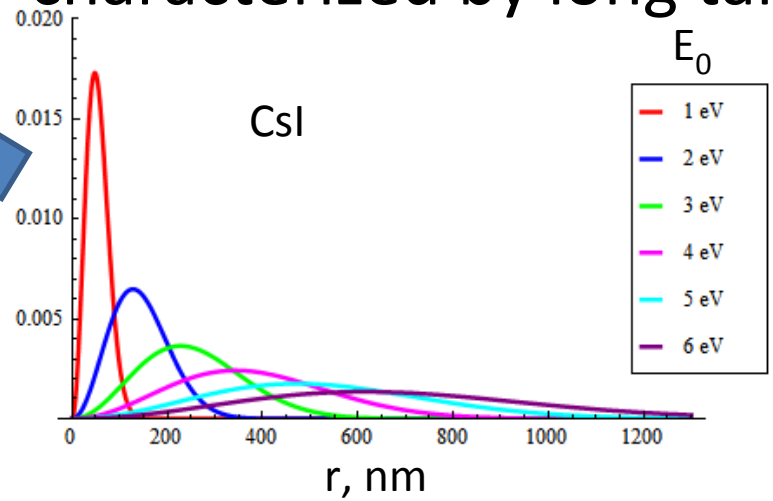
Thermalization length depends on kinetic energy as $(E_{kin})^{3/2}$



Spatial distribution of thermalized electrons

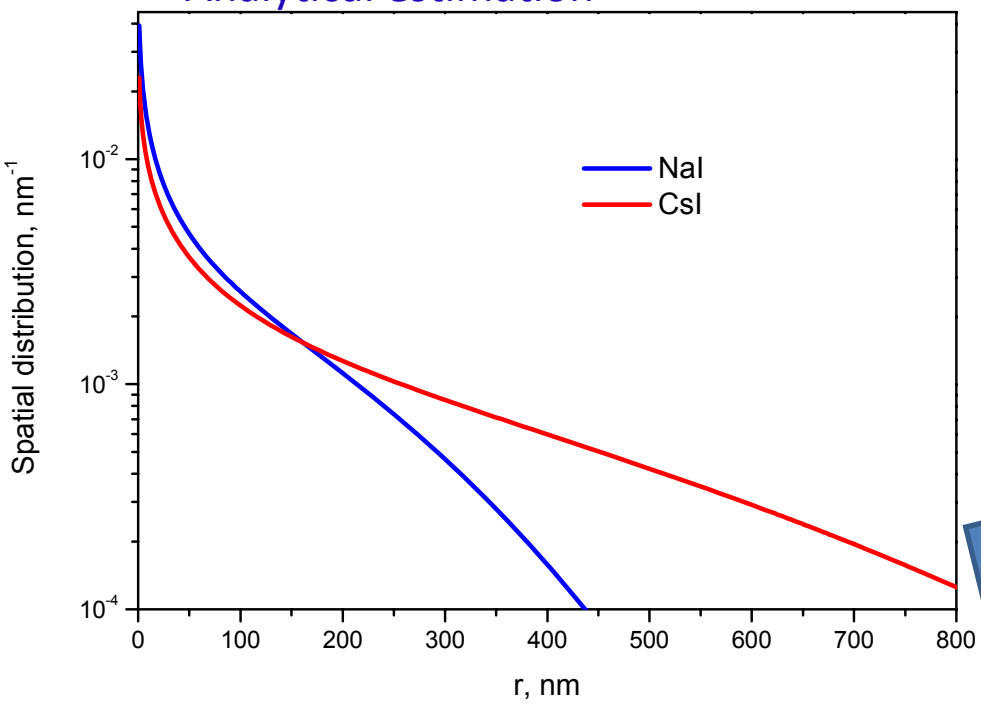


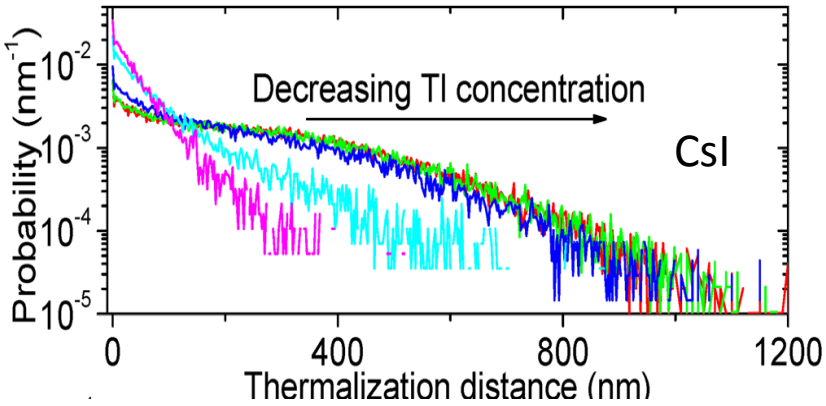
The resulting spatial distribution of excitations after thermalization is non-Gaussian and is characterized by long tail



Spatial distribution of thermalized electrons

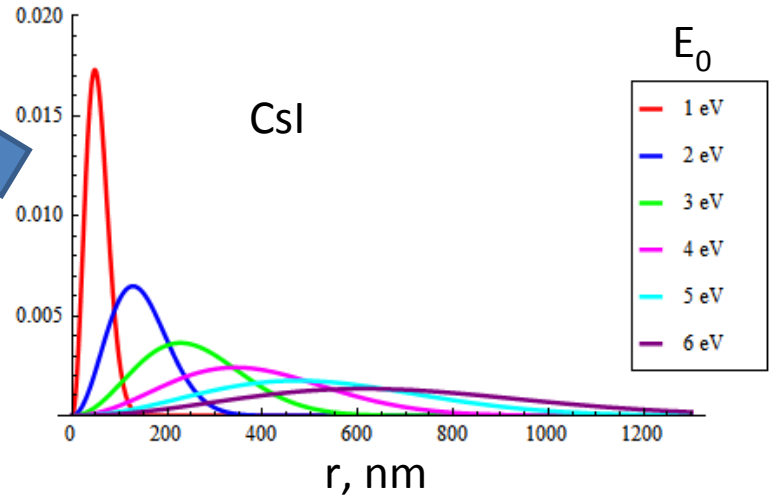
Analytical estimation





Computer simulation of electron thermalization in CsI and CsI(Tl),

Z. Wang, Y. Xie, B. D. Cannon et al. 2011



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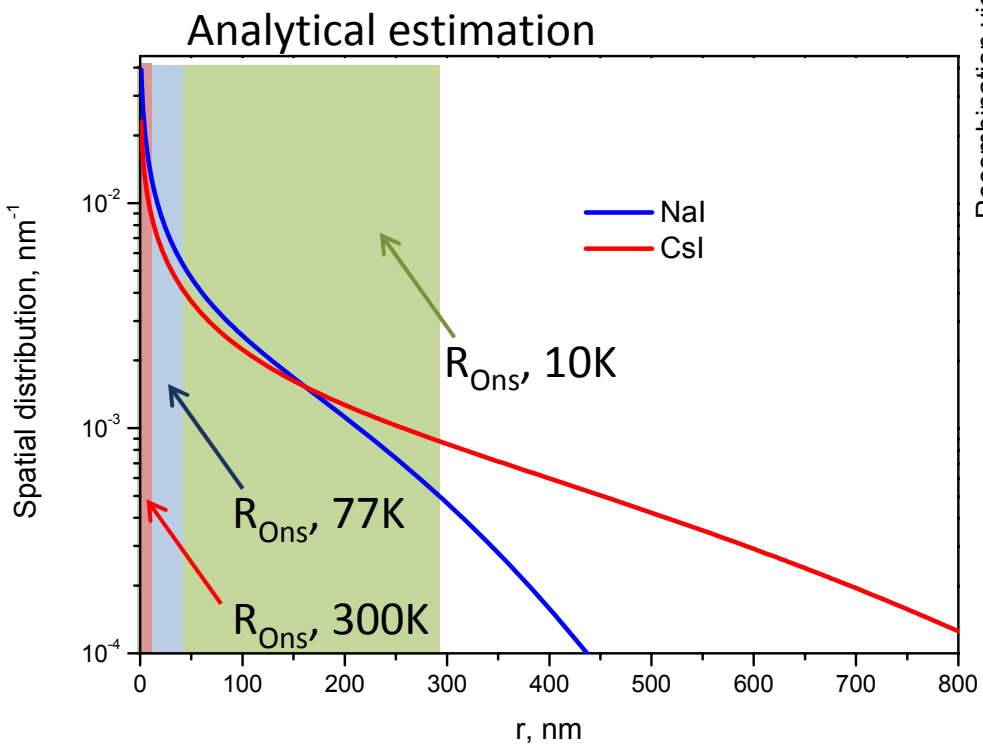

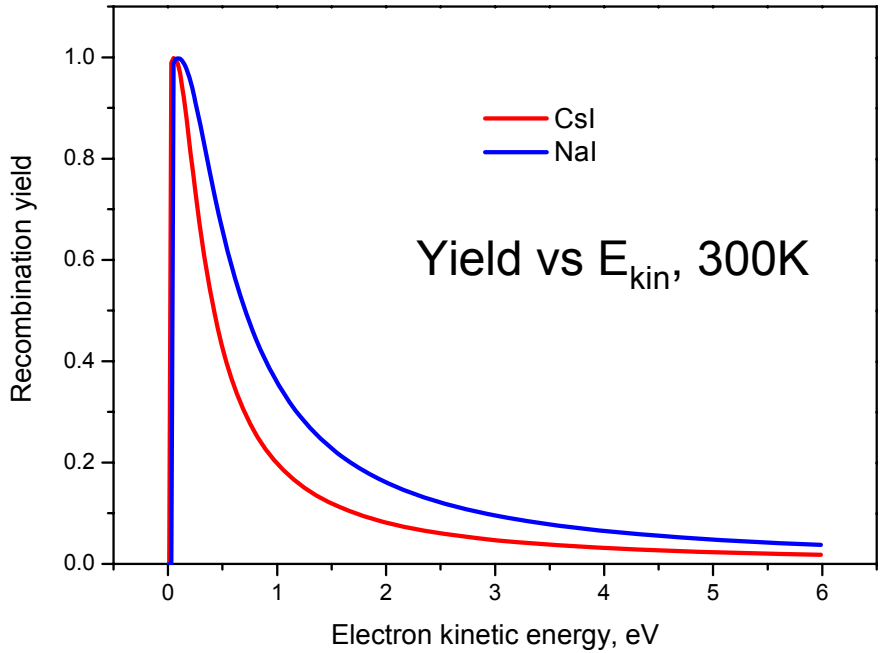

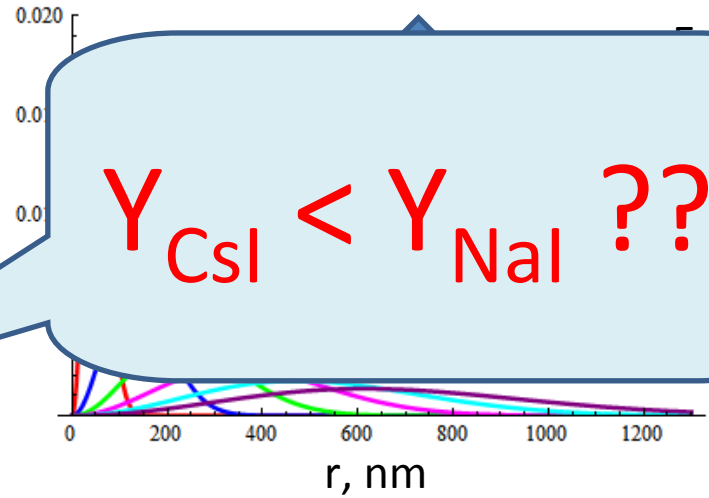

$Y_{CsI} < Y_{NaI}$???

	R_{Ons}, 300K	Yield, 300K	Yield, 77K
CsI	9.87 nm	0.24	0.44
NaI	9.05 nm	0.34	0.58

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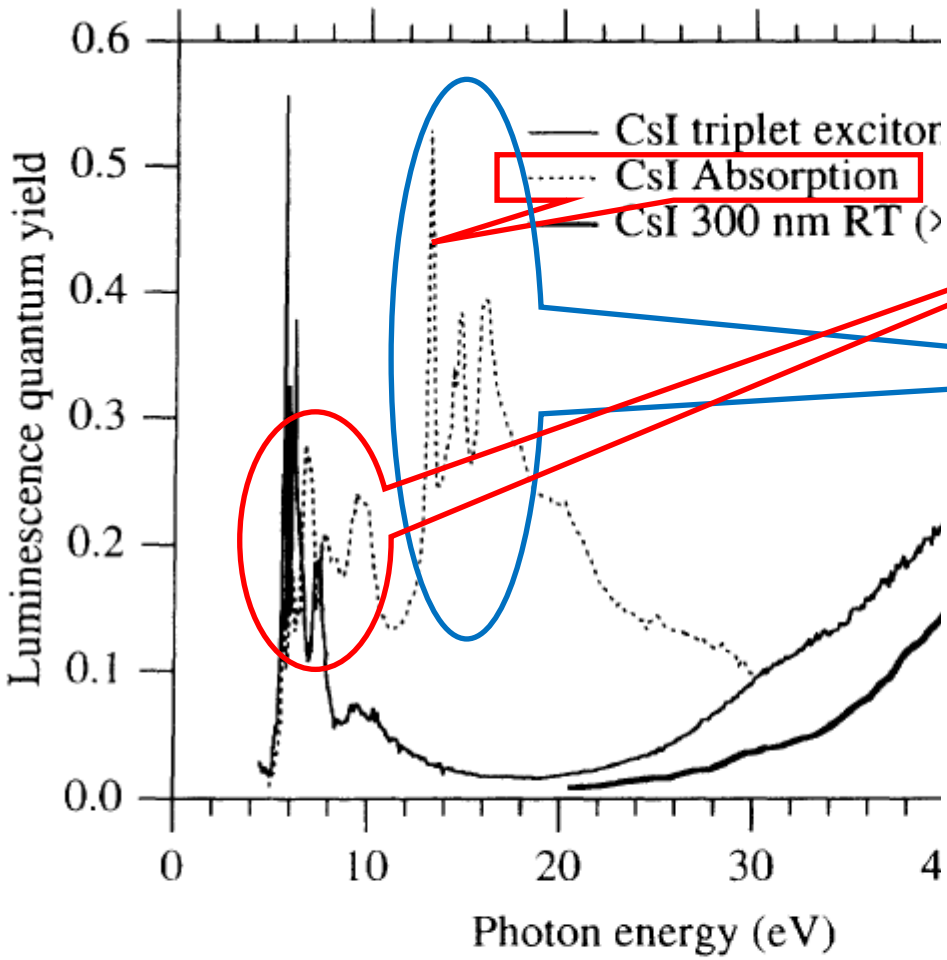
Transitions from 5pCs core levels



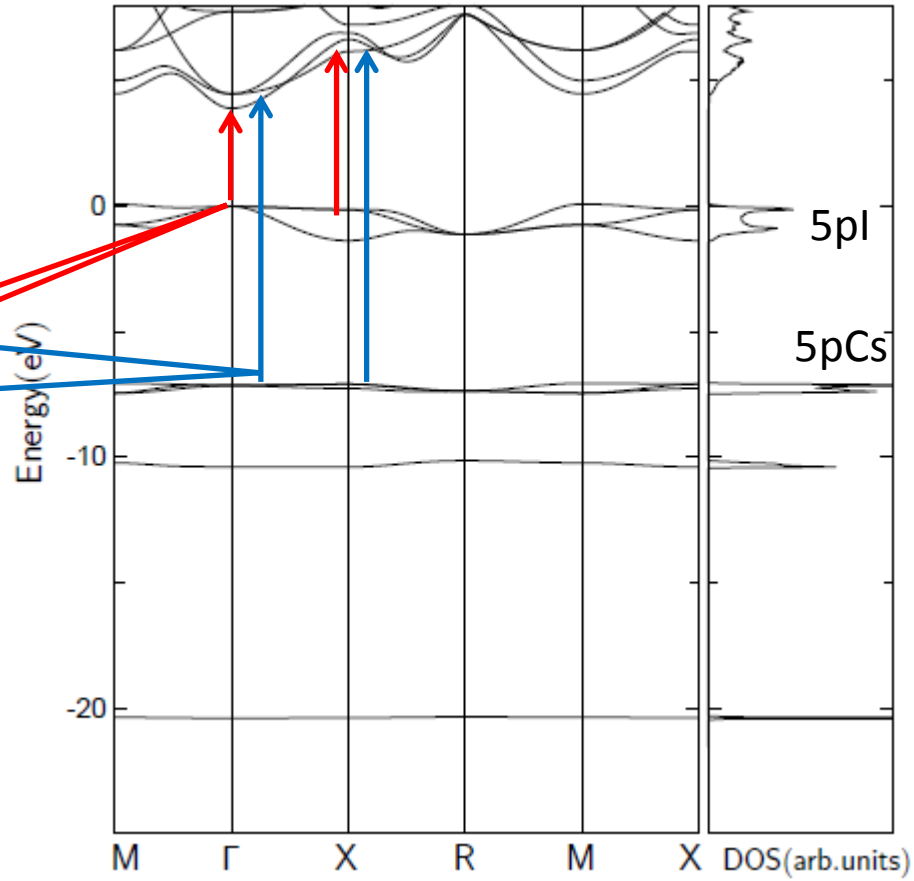
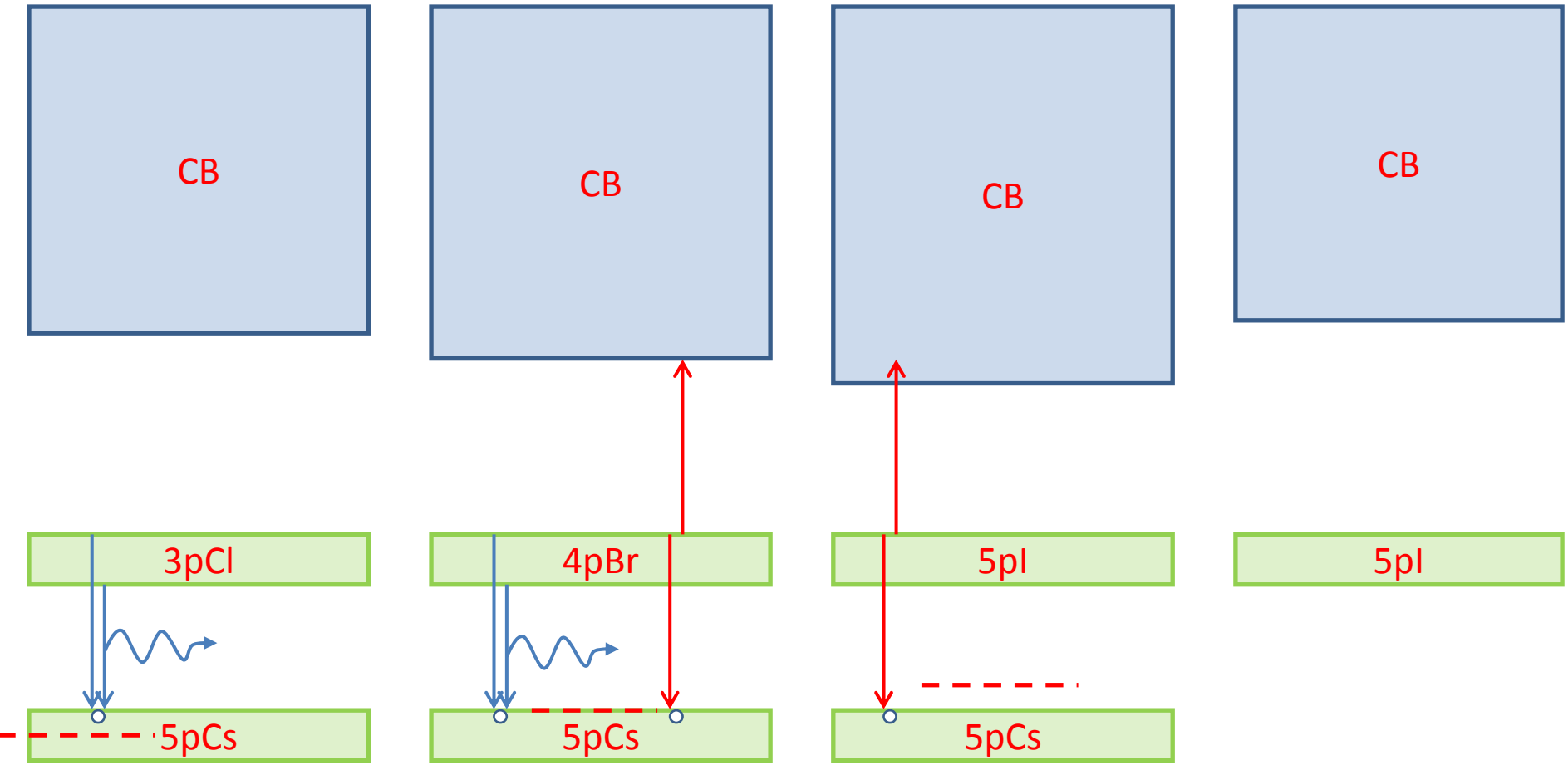


FIG. 2. Excitation spectra of CsI luminescence: the FIL (300 K) (thick) and of triplet exciton (100 K) (thin) compared with CsI absorption (dashed).

From core-valence transitions to Auger process in cesium halides



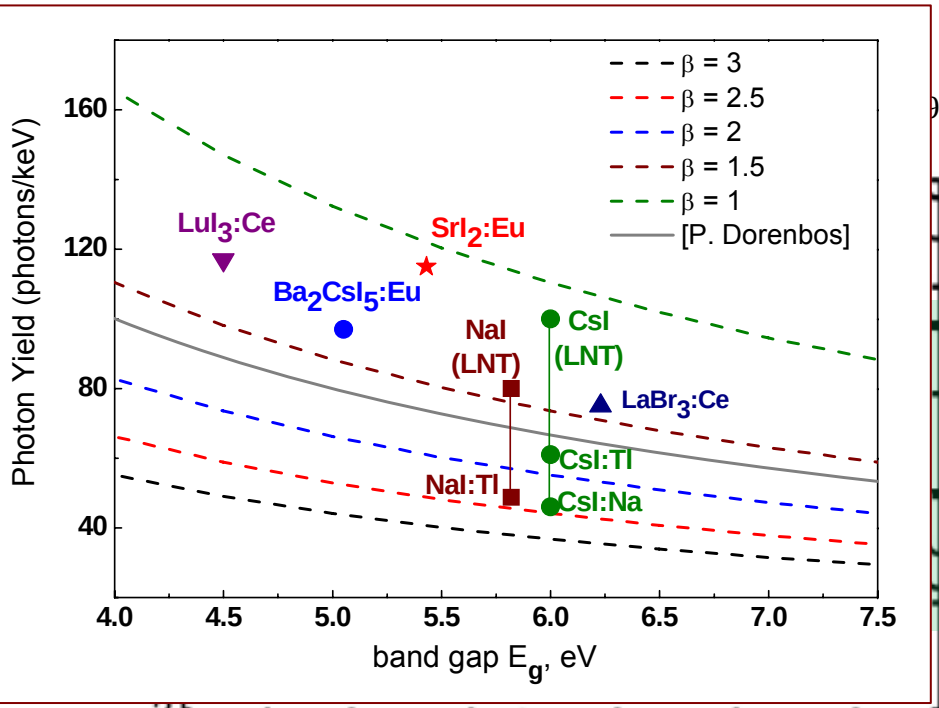
CsCl
Radiative decay of 5pI core hole

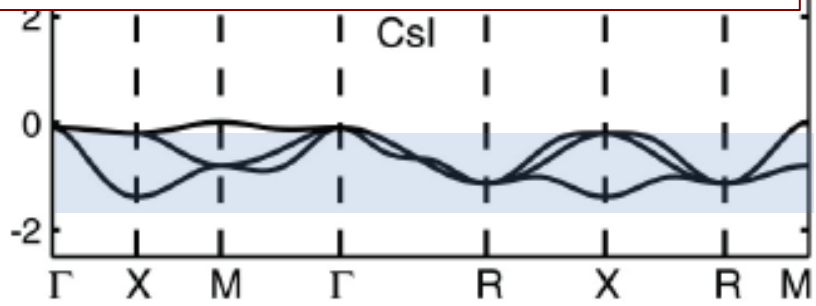
CsBr
Radiative and Auger decay of 5pI core hole

CsI
Auger decay of 5pI core hole with creation of exciton or low-energy e-h pair (E_{kin} < 1 eV)

NaI
No uppermost core levels

Starting states for thermalization ($E_{kin}<E_g$)





e-e passive region in CB after all e-e scattering events is filled mostly in low energy part with account for Auger relaxation of 5pI holes

Estimation of β decreases significantly! Creation of each 5pI hole with threshold energy 13 eV produces 2 e-h pairs, one electron and both holes of which has low kinetic energy!

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Elastic scattering on impurities and carriers

$$\frac{1}{\tau} = \nu\sigma N = \sqrt{\frac{2E}{m^*}}\sigma N$$

Charged impurities/carriers: Conwell & Weisskopf (Phys. Rev. 77, 388-390, 1950)

$$\sigma_c^{CW} = 2\pi\left(\frac{Ze^2}{4\pi\varepsilon_0\varepsilon_{st}2E}\right)^2 \ln\left(1+4\left(\frac{E}{E_m}\right)^2\right), \quad E_m = \frac{Ze^2}{4\pi\varepsilon_0\varepsilon_{st}r_m}, \quad r_m = \frac{1}{2}n_c^{-1/3}$$

$$\sigma_c^{CW} = \frac{1}{2}\pi R_{Ons}^2\left(\frac{k_BT}{E}\right)^2 \ln\left(1+\left(\frac{E}{k_BTx_m}\right)^2\right), \quad x_m = R_{Ons}n_c^{1/3}$$

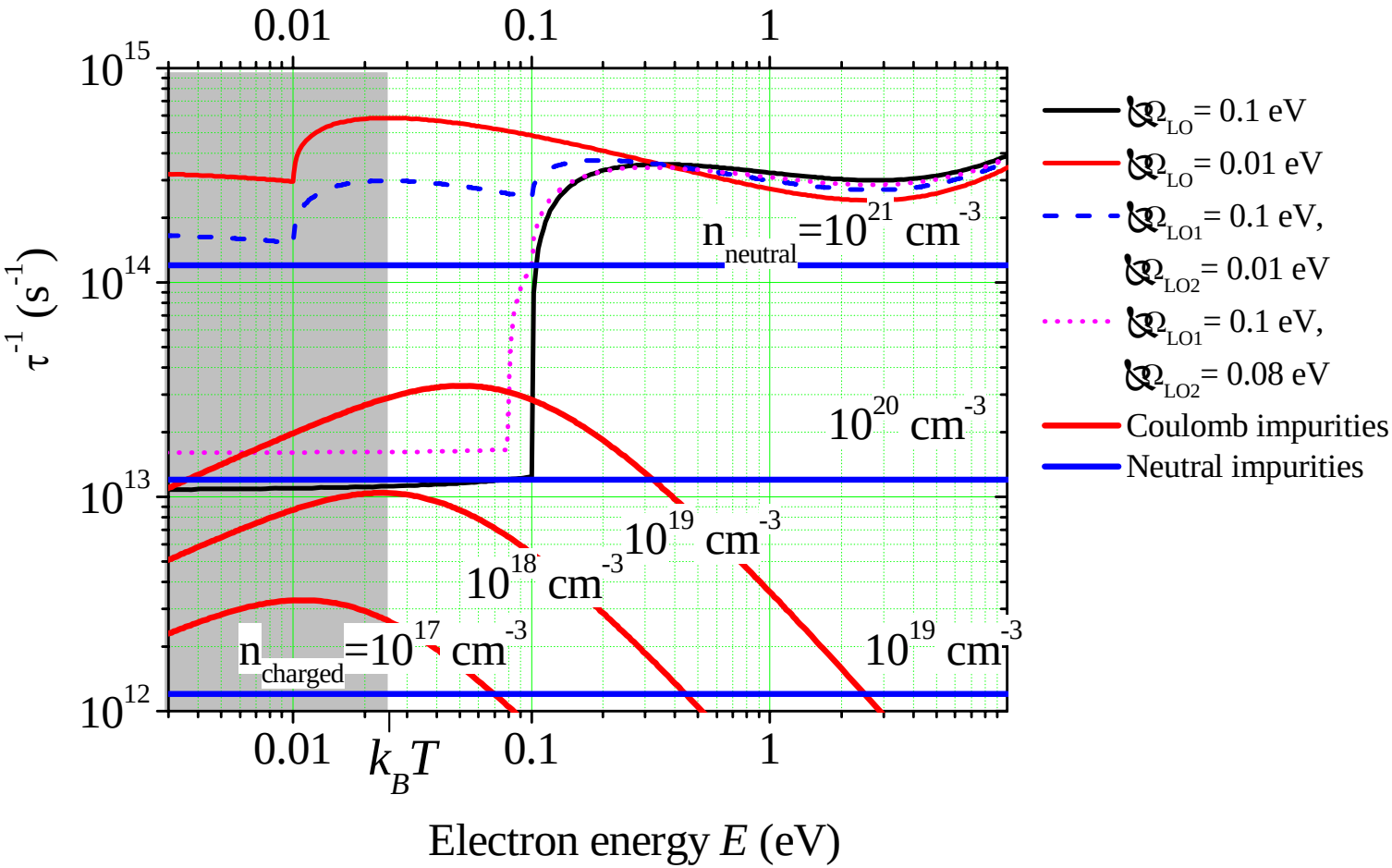
$$\frac{1}{\tau} = \sqrt{\frac{2k_BT}{m^*}}\frac{1}{2}\pi R_{Ons}^2\left(\frac{k_BT}{E}\right)^{3/2} \ln\left(1+\left(\frac{E}{k_BTx_m}\right)^2\right)n_c = 1.5\times10^{-5}\left(\frac{k_BT}{E}\right)^{3/2} \ln\left(1+\left(\frac{E}{k_BTx_m}\right)^2\right)n_c\left[\mathrm{cm}^{-3}\right]\frac{1}{\mathrm{s}}$$

Neutral impurities

$$\sigma \approx 20\frac{ka^*}{k^2}, \quad k = \frac{1}{\hbar}\sqrt{2m^*E}, \quad a^* - \text{impurity Bohr radius}$$

$$\sigma \approx \frac{20a^*\hbar}{\sqrt{2m^*E}}, \quad \frac{1}{\tau} = \frac{20a^*\hbar}{m^*}n = 1.2\times10^{-7}n[\mathrm{cm}^{-3}]\frac{1}{\mathrm{s}} = 1.2\times10^{13}n[\mathrm{mol.\%}]\frac{1}{\mathrm{s}} \quad (a^* = 1\mathrm{nm}, m^* = m_e)$$

Elastic scattering on impurities and carriers



Scattering on impurities is the limiting factor for thermalization length only for crystals with high LO phonon energies in LO-passive region for high concentrations of neutral (>0.5%) impurities and for high concentration of carriers (>10^{18} cm^{-1})

Conclusions

The development of comprehensive model of scintillator based on multi-particle consideration of multi-scale evolution of strongly non-equilibrium excited region on the basis of deep directional experimental investigations allows to

- Make a progress in fundamental physics
- Obtain new results in applied physics – e.g. by justification that mixed crystals is a way to improve scintillator properties
- Be useful in pragmatic sense, because it is a background for new material development (industrial applications)

Thank you for your attention and cooperation!