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Institute of Physics and Technology

FORMATION OF THERMOLUMINESCENCE DOSE RESPONSE NONLINEARITY IN CLUSTER MODEL WITH DEEP HOLE TRAPS

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Tomsk, 2020

Introduction

Thermoluminescence (TL) is widely used in dosimetry and study of defect properties in wide-gap insulators. Study of formation of complex TL dose dependence such as non-linear dose dependence is of great interest. It is known that the competition processes associated with the presence of deep hole traps in phosphors can cause superlinearity [1] and sublinearity [2] of the TL dose dependence.

Also non-monotonic TL dose dependence is observed in number of materials, for example in LiF [3, 4]. Usually presence of non-monotonic TL dose dependence is explained by radiation damage of trapping centers [4, 5]. At the same time competition processes which involve deep hole traps was used to explain non-monotonic TL dose dependence [5].

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2. S.V. Nikiforov, A.S. Merezhnikov, V. Pagonis, Radiat. Meas.105, 54-61 (2017).
3. G. C. Crittenden, P. D. Townsend, J. Gilkest, M. C. Wintersgill. J. Phys. D: Appl. Phys. 7, 197, 2410-2421 (1974)
4. V. K. Jain, S. P. Kathuria and A. K. Ganguly. J. Phys. C: Solid State Phys. 8, 2191-2197 (1975)
5. J. L. Lawless, R. Chen, D. Lo, V. Pagonis. J. Phys.: Condens. Matter. 17, 737-753 (2005)

Introduction

It is known that models with cluster defects are used for describing TL kinetics in polycrystalline and nanostructured phosphors, as well as phosphors irradiated with high doses [6]. In such models, transitions between different localized levels in the cluster are taken into account along with delocalized transitions. It is known that presence of localized transitions can lead to anomalous heating rate effect which was observed in $YPO_4:Ce^{3+}$, Sm^{3+} [7].

Unresolved problems

Models with deep hole traps in the cluster have not been investigated earlier. We expect that presence of deep hole traps in cluster model can lead to new features of TL dose dependence.

6. A. Mandowski, J. Swiatek, Radiat. Meas., vol. 29, pp. 415-419, 1998.

7. A.J.J. Bos, N.R.J. Poolton, J. Wallinga, A. Bessiere, P. Dorenbos. Radiat. Meas. 45, 343-346 (2010)

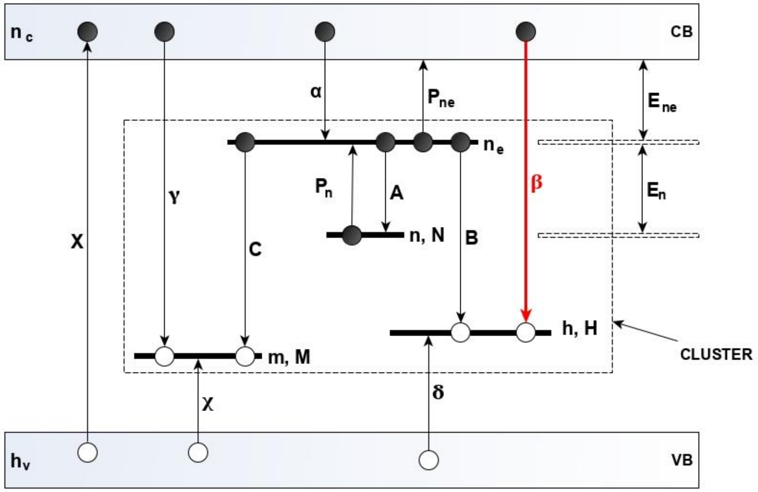
Aim of the research:

To Analyze TL dose dependences in cluster model with deep hole traps and study influence of localized transition on features of TL dose dependences.

OUTLINE

1. Cluster model description
2. Simulation process
3. Calculation method and algorithm
4. Description of analytical approach of TL output estimation
5. No deep hole traps case
6. Clusters with deep hole trap
7. Conclusions

Cluster Model



Cluster capture centers:

Electron Trap (N),

Luminescence Center (H),

Deep Hole Trap (M)

N: number of the main (TL-active) traps per cluster

M: number of deep traps per cluster

H: number of luminescence centers per cluster

N = H + M

Delocalized transitions

α: electron capture coefficient to the excited state of the main trap from conduction band

β: probability coefficient of radiative recombination

γ: recombination coefficient with hole in deep trap

δ: probability coefficient of hole capture to luminescence center

χ: probability coefficient of hole capture to hole trap

Localized transitions

A: probability coefficient of local trapping to the main trap

B: probability coefficient of local recombination to luminescence center

C: probability coefficient of local recombination to deep trap

X: efficiency of electron-hole pair's creation

E_n: energetic distance from the ground state of the main trap to the excited state of the main trap (activation energy)

E_{ne}: energetic distance from the excited state of the main trap to the conduction band (activation energy)

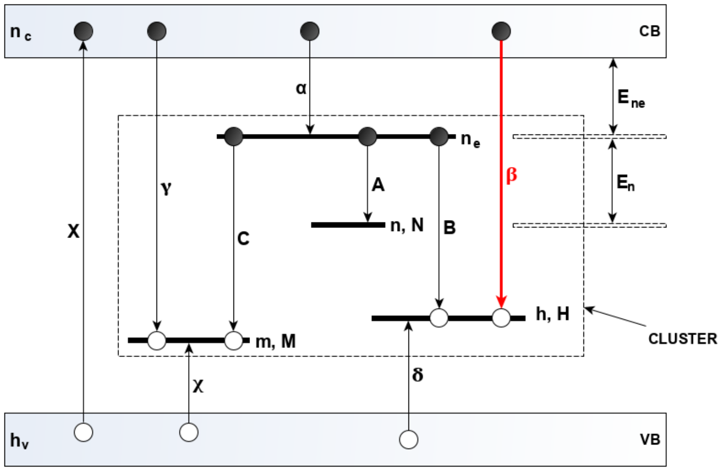
Simulation stages

Irradiation -> Relaxation -> Heating

Irradiation/Relaxation stage:

- electron-hole pair formation (transition X) (Irradiation stage only)
- electron trapping by clusters (transition α)
- recombination of conduction band electrons and holes in luminescence centers H (transition β) or in traps M (transition γ)
- holes trapping by luminescence centers (transition δ) and hole traps (transition χ)
- electrons in the excited states n_e moving to the ground state (transition A)
- electrons in the excited states n_e recombination with holes in the corresponding hole centers (transitions B, C)

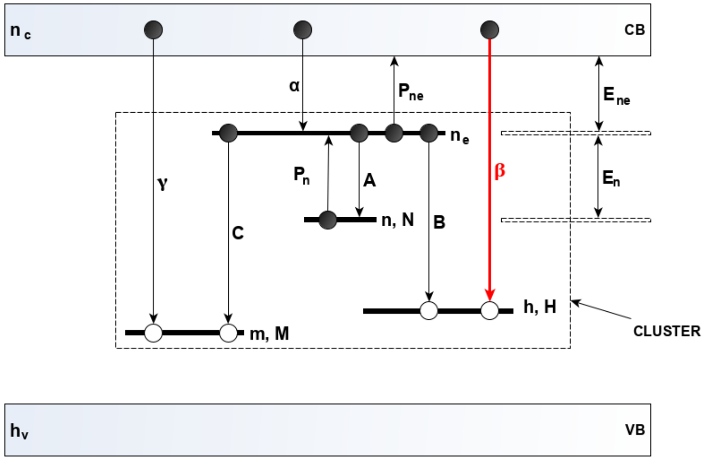
Irradiation/Relaxation (X = 0)



Heating stage:

- Thermal stimulation of electrons (transitions Pne, Pn)
- electron retrapping by clusters (transition α)
- recombination of conduction band electrons and holes in luminescence centers H (transition β) or in traps M (transition γ)
- electrons in the excited states n_e moving to the ground state (transition A)
- electrons in the excited states n_e recombination with holes in the corresponding hole centers (transitions B, C)

Heating



Monte-Carlo method

Simulation consists of many iterations at each stage.

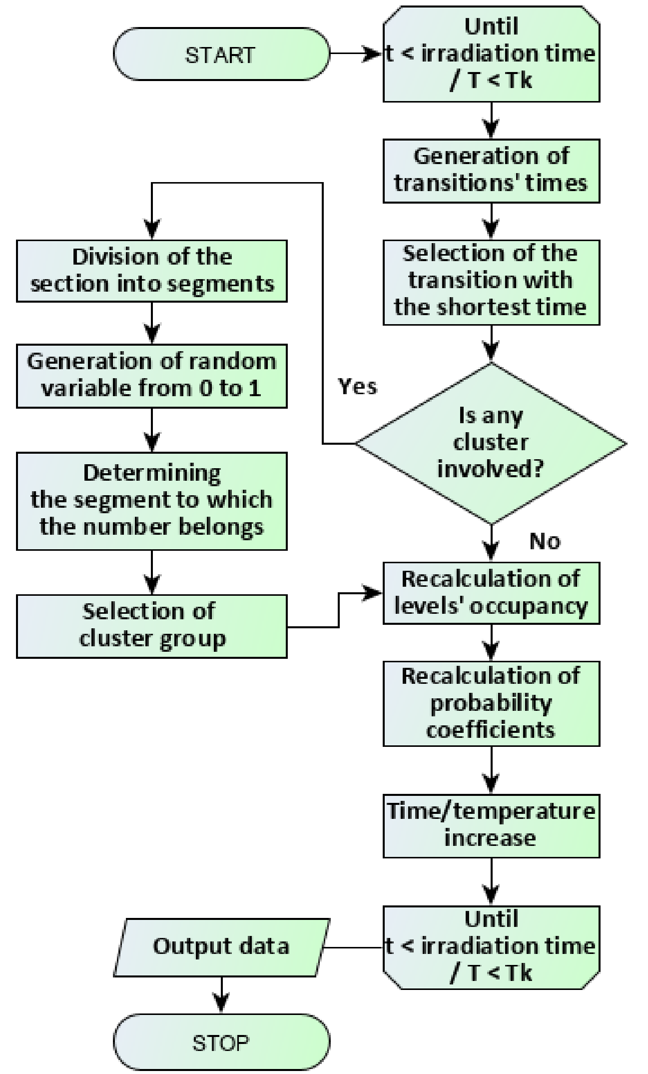
1. On every iteration for each transitions random time is calculated.
2. Among the obtained times the shortest one stands for occurred transition.
3. According to the transitions occupancies of levels are recalculated

The exponential probability distribution is used for Monte-Carlo method [8]. The algorithm was optimized using method in the paper [9]. The method was changed in order to apply it to the model. The program was developed in C++.

$$P(t) = 1 - \exp\left[-\int_0^t \lambda(\tau)\, d\tau\right]$$

$$P(t) = 1 - \exp[-\lambda t], \quad \lambda = \text{const}$$

$$t = -\frac{\ln(1-P)}{\lambda}, \qquad P\epsilon[0,1)$$



8. Mandowski and J. Świaltek. Philos. Mag. Part B, 65, 729-732 (1992)

9. A.S. Merezhnikov , S.V. Nikiforov, V. Pagonis. Radiat. Meas. 125, 78-84 (2019)

Transition equations

1. $\lambda(\alpha) = \alpha \cdot (N - n - n_e) \cdot n_c$
2. $\lambda(\beta) = \beta \cdot h \cdot n_c$
3. $\lambda(\delta) = \delta \cdot (H - h) \cdot h_\nu$
4. $\lambda(\gamma) = \gamma \cdot h \cdot n_c$
5. $\lambda(\chi) = \chi \cdot (M - m) \cdot h_\nu$
6. $\lambda(A) = A \cdot (N - n) \cdot n_e$
7. $\lambda(B) = B \cdot h \cdot n_e$
8. $\lambda(C) = C \cdot m \cdot n_e$
9. $\lambda(\mathrm{p}) = s\, exp\left(-\frac{E}{kT}\right) n$
10. $\lambda(P_{ne}) = s_{ne} exp\left(-\frac{E_{ne}}{kT}\right) ne$

Analytical approximation was used for transitions P_n and P_{ne} in order to improve performance:

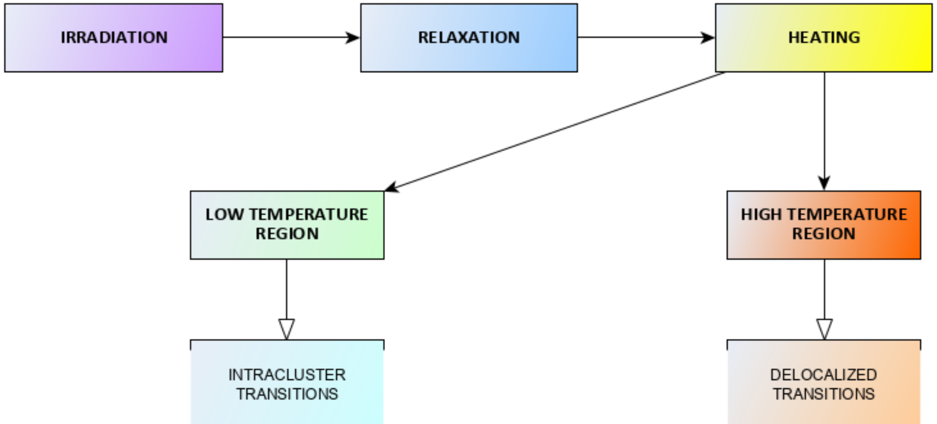
$$\ln(1-P) \cdot \beta = \frac{k \cdot s \cdot T0^2}{E}\left[\exp\left\{\frac{-E}{k(T0)}\right\} - \exp\left\{\frac{-E}{k(T0 + \Delta T)}\right\}\right]$$

10. A.S. Merezhnikov , S.V. Nikiforov, V. Pagonis. Radiat. Meas. 125, 78-84 (2019)

Model parameters

	Description	Value
Z	Total number of clusters	10^5
s	The frequency factor for the electron in the ground state of the main trap	$10^{10}\ s^{-1}$
s_{ne}	The frequency factor for the electron in the excited state of the main trap	$10^{10}\ s^{-1}$
E	The energetic distance from the ground state of the main trap to the excited state of the main trap (activation energy)	0.7 eV
E_{ne}	The energetic distance from the excited state of the main trap to the conduction band (activation energy)	0.85 eV
α	The electron capture coefficient to the excited state of the main trap from conduction band	$2 \cdot 10^{-3}\ s^{-1}$
β	Probability coefficient of radiative recombination	$10^{-3}\ s^{-1}$
γ	The recombination coefficient with hole in deep trap	$10^{-3}\ s^{-1}$
δ	Probability coefficient of hole capture to luminescence center	$10^{-3}\ s^{-1}$
χ	Probability coefficient of hole capture to hole trap	$10^{-1}\ s^{-1}$
A	Probability coefficient of local trapping to the main trap	$1\ s^{-1}$
B	Probability coefficient of local recombination to luminescence center	$0.1\ s^{-1}$
C	Probability coefficient of local recombination to deep trap	$0.1\ s^{-1}$
X	The efficiency of electron-hole pair's creation	$100\ s^{-1}$

Heating stage

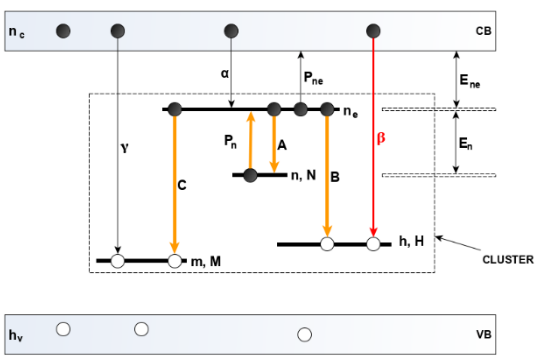


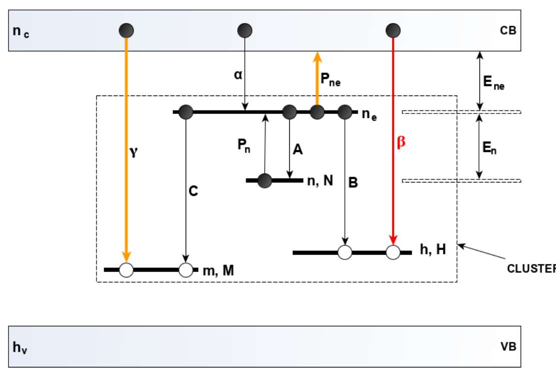
Let us consider the following case:

1. Large values of E_{ne}
2. Small heating rate

It that case probability of transition P_{ne} is very small comparing to probability of transition P_n.

It is expected that under such conditions in low temperature region only localized transitions (P_n, A, B,C) occur.





Thus at low temperatures electrons will stay inside cluster and recombine only with holes in the same cluster.

At high temperatures clusters which contain only electrons or only holes will be left.

Let us also consider a case with relatively small values of cluster trapping coefficient α.
Since all intracluster electron relaxations are occurred at low temperatures, only delocalized transitions dominate at high temperatures .

No deep hole traps

Z(n_0, m_0) - number of cluster with n_0 filled electron traps and h_0 filled luminescence centers before heating

"FREE" ELECTRONS

Z(n_0=0, h_0) -> **0**

Z(n_0=1, h_0=0) -> **1**

Z(n_0=1, h_0=1) -> 1 – 1 = **0**

Total number of "free" electrons:

$N_\Sigma =$ Z(n_0=1, h_0=0)

Total number of localized recombinations:

$R_\Sigma =$ Z(n_0=1, h_0=1)

LUMINESCENCE CENTER HOLES

Z(n_0, h_0=0) -> **0**

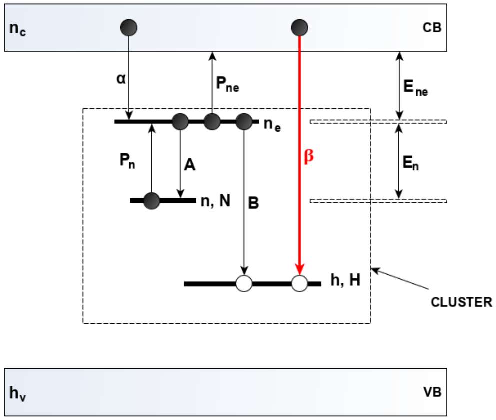
Z(n_0=0, h_0=1) -> **1**

Z(n_0=1, h_0=1) -> 1 – 1 = **0**

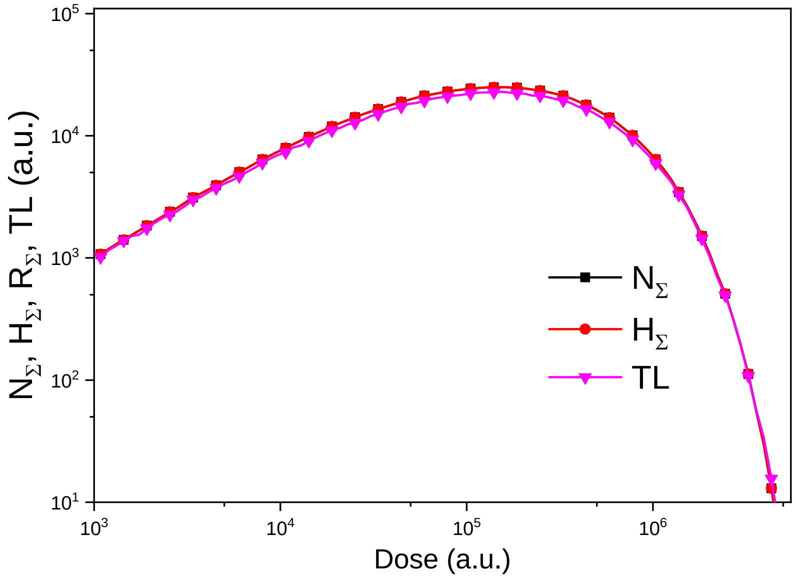
Total number of holes in luminescence centers which will recombine with electrons radiatively:

$H_\Sigma =$ Z(n_0=0, h_0=1)

	Description	Value
N	The number of the main (TL-active) traps per cluster	1
M	The number of deep traps per cluster	0
H	The number of emission centers per cluster	1
E_{ne}	The energetic distance from the excited state of the main trap to the conduction band (activation energy)	1.3 eV
	Heating rate	1 K/s



No deep hole traps



TL output fits with number of “free” electrons and occupied luminescence centers after low temperature region

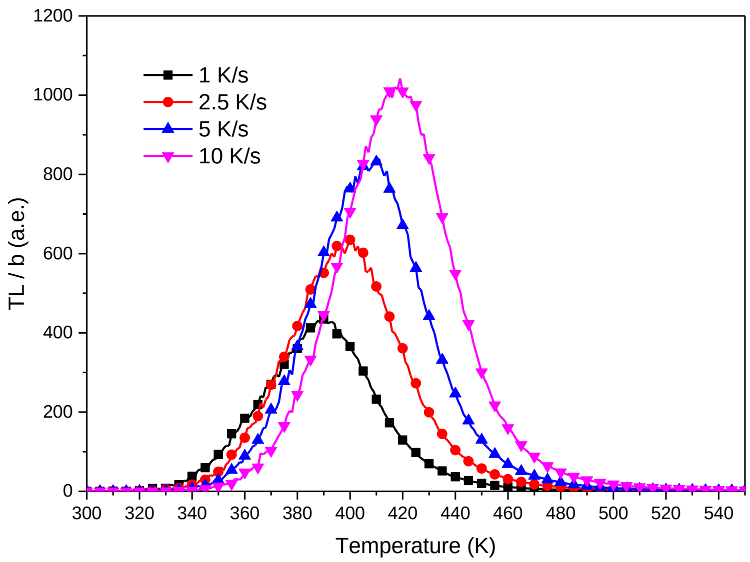
TL dose dependence and total number of electrons/holes.

N_Σ - total number of holes in deep traps which will recombine with conduction band electrons

H_Σ - total number of holes in luminescence centers which will recombine with electrons radiatively

TL output on heating rate

	Description	Value
N	The number of the main (TL-active) traps per cluster	1
M	The number of deep traps per cluster	0
H	The number of emission centers per cluster	1
E_{ne}	The energetic distance from the excited state of the main trap to the conduction band (activation energy)	0.85 eV



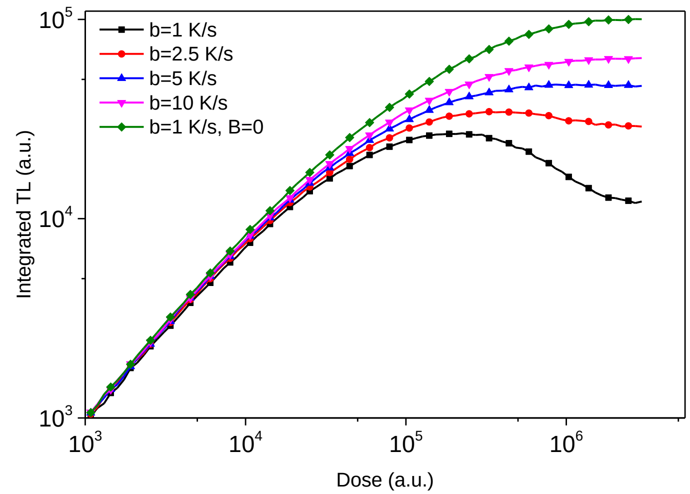
TL curves calculated with different heating rates b

The TL output increases with increasing of heating rate:

1. TL curves shift towards higher temperature region as heating rate increases
2. At high temperatures probability of electron leaving cluster is higher
3. More electrons reach conduction band and radiatively recombine with holes in luminescence centers

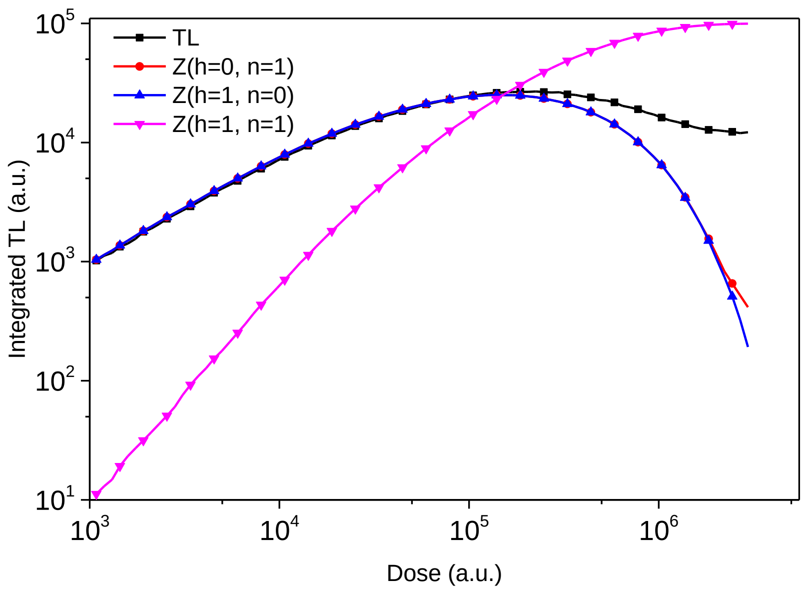
Dose dependences

TL output increases with increasing heating rate. As probability of transition B is equal to zero TL output is maximized and TL dose dependence becomes monotonic



TL dose dependences calculated with different heating rates b

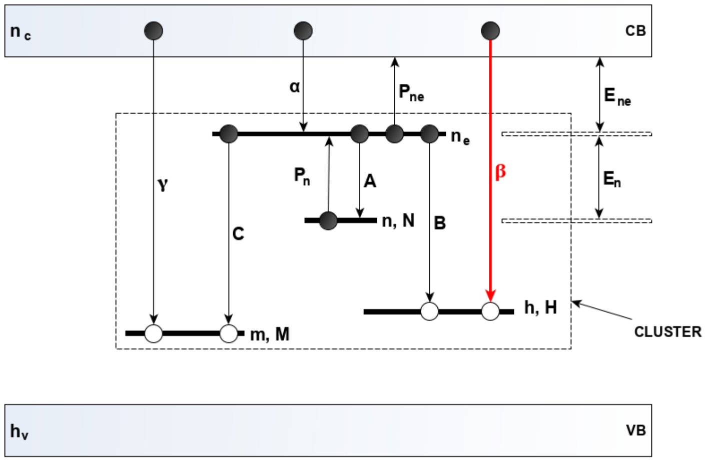
TL output does not fit with number of “free” electron and occupied luminescence centers after low temperature region completely. But it can be seen that the derivative of the TL dose dependence changes sign near intersection of curves Z(h=1, n=1) and Z(h=1, n=0), where the number of completely occupied clusters becomes greater than one of cluster with electron or hole only.



TL dose dependence and numbers of cluster. b=1 K/s, Z (h, n) – number of clusters with h occupied luminescence centers and n occupied electron traps

Deep hole trap presence

	Description	Value
N	The number of the main (TL-active) traps per cluster	2
M	The number of deep traps per cluster	1
H	The number of emission centers per cluster	1
E_{ne}	The energetic distance from the excited state of the main trap to the conduction band (activation energy)	1.3 eV
	Heating rate	1 K/s



Presence of deep hole traps make analytical consideration of TL output on heating stage slightly more complex. Influence of localized competition between deep hole traps and luminescence centers on TL dose dependence is expected

“FREE” ELECTRONS

$Z(n_0, m_0, h_0)$ - number of cluster with n_0 filled electron traps, m_0 filled hole traps and h_0 filled luminescence centers before heating

$Z(n_0=0, m_0, h_0)$ -> **0**

$Z(n_0=1, m_0=0, h_0=0)$ -> **1**

$Z(n_0=2, m_0=0, h_0=0)$ -> **2**

$Z(n_0=2, m_0=0, h_0=1)$ -> 2 – 1 = **1**

$Z(n_0=2, m_0=1, h_0=0)$ -> 2 – 1 = **1**

$Z(n_0=1, m_0=1, h_0=0)$ -> **0**

$Z(n_0=1, m_0=0, h_0=1)$ -> 1 – 1 = **0**

$Z(n_0=1, m_0=1, h_0=1)$ -> 1 – 1 = **0**

$Z(n_0=2, m_0=1, h_0=1)$ -> 2 – 1 – 1 = **0**

Total number of “free” electrons:

$$N_\Sigma = 2 * Z(n_0 = 2, m_0 = 0, h_0 = 0) + \\ Z(n_0 = 1, m_0 = 0, h_0 = 0) + \\ Z(n_0 = 2, m_0 = 0, h_0 = 1) + \\ Z(n_0 = 2, m_0 = 1, h_0 = 0)$$

LUMINESCENCE CENTER HOLES

$Z(n_0, m_0, h_0)$ - number of cluster with n_0 filled electron traps, m_0 filled hole traps and h_0 filled luminescence centers before heating

$Z(n_0, m_0, h_0=0)$ -> **0**

$Z(n_0=2, m_0=0, h_0=1)$ -> 1 – 1 = **0**

$Z(n_0=1, m_0=0, h_0=1)$ -> 1 – 1 = **0**

$Z(n_0=2, m_0=1, h_0=1)$ -> 1 – 1 = **0**

$Z(n_0=0, m_0=0, h_0=1)$ -> **1**

$Z(n_0=0, m_0=1, h_0=1)$ -> **1**

$Z(n_0=1, m_0=1, h_0=1)$ -> **C/(B+C)**

Total number of holes in luminescence centers which will recombine with electrons radiatively :

$$H_{\Sigma} = Z(n_0 = 0, m_0 = 0, h_0 = 1) + Z(n_0 = 0, m_0 = 1, h_0 = 1) + \frac{C}{B + C} * Z(n_0 = 1, m_0 = 1, h_0 = 1)$$

DEEP TRAP HOLES

$Z(n_0, m_0, h_0)$ - number of cluster with n_0 filled electron traps, m_0 filled hole traps and h_0 filled luminescence centers before heating

$Z(n_0, m_0=0, h_0)$ -> **0**

$Z(n_0=2, m_0=1, h_0=0)$ -> 1 – 1 = **0**

$Z(n_0=1, m_0=1, h_0=0)$ -> 1 – 1 = **0**

$Z(n_0=2, m_0=1, h_0=1)$ -> 1 – 1 = **0**

$Z(n_0=0, m_0=1, h_0=0)$ -> **1**

$Z(n_0=0, m_0=1, h_0=1)$ -> **1**

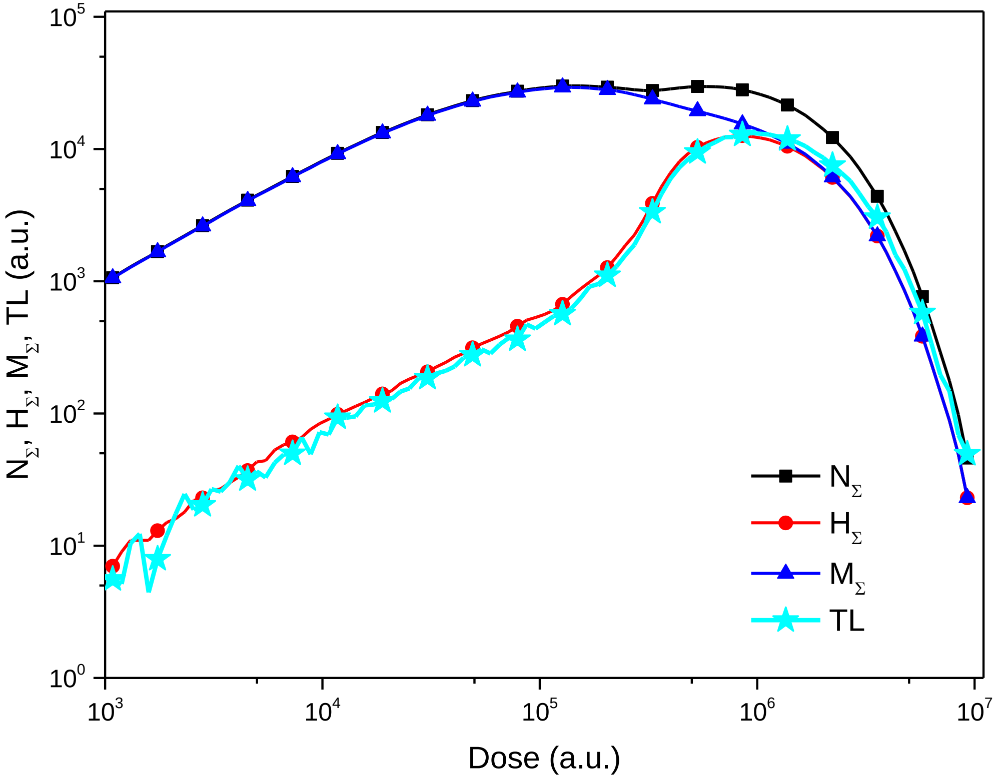
$Z(n_0=1, m_0=1, h_0=1)$ -> **B/(B+C)**

Total number of holes in deep traps which will recombine with conduction band electrons:

$$M_{\Sigma} = Z(n_0 = 0, m_0 = 1, h_0 = 0) + Z(n_0 = 0, m_0 = 1, h_0 = 1) + \frac{B}{B+C} * Z(n_0 = 1, m_0 = 1, h_0 = 1)$$

Deep hole trap presence

$$N_\Sigma = H_\Sigma + M_\Sigma$$



TL dose dependence and total number of electrons/holes.

N_Σ - total number of holes in deep traps which will recombine with conduction band electrons

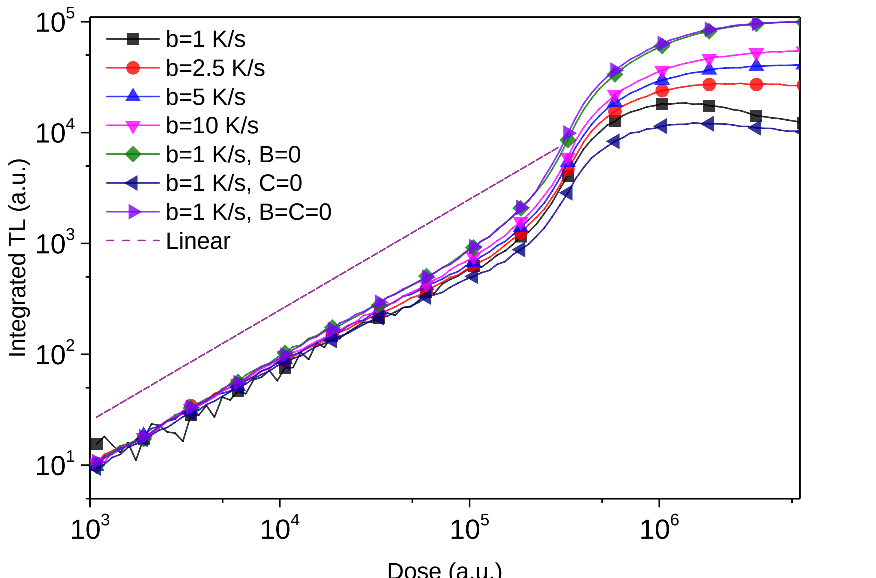
H_Σ - total number of holes in luminescence centers which will recombine with electrons radiatively

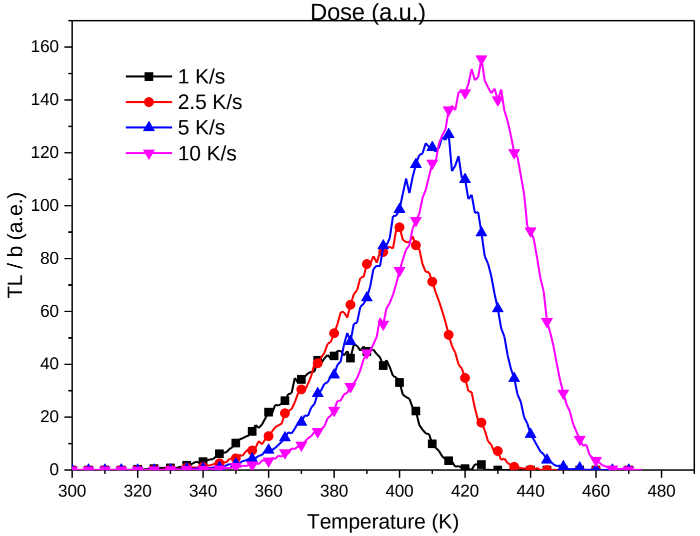
M_Σ - total number of holes in deep traps which will recombine with conduction band electrons

- TL output fits with number of occupied luminescence centers after low temperature region
- Insignificant difference between the curves can be explained by non-fulfilment of mentioned conditions and random deviations

TL curves and dose dependences

TL output increases with increasing heating rate. As probability of transition B is equal to zero TL output is maximized and TL dose dependence become monotonic. In case when B > 0 and C = 0 TL output is the lowest since localized competition between luminescence center and hole trap in the same cluster vanishes and more holes in luminescence centers recombine with electrons non-radiatively



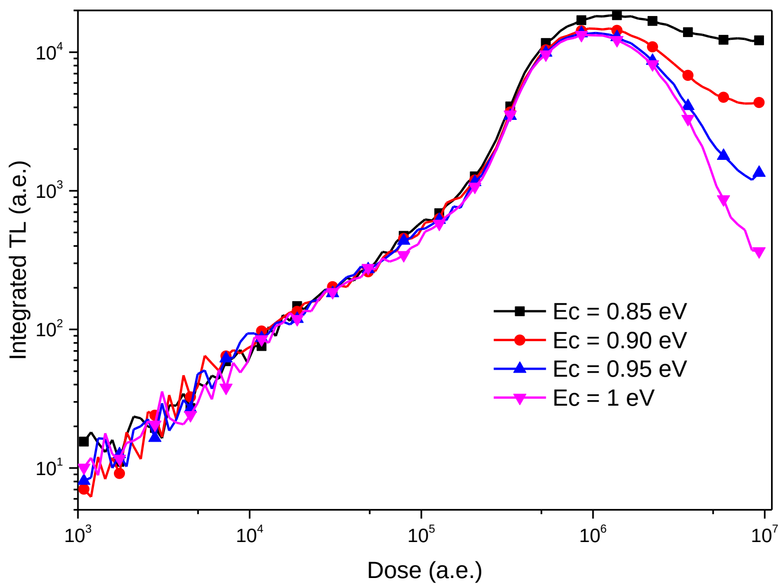


	Description	Value
N	The number of the main (TL-active) traps per cluster	2
M	The number of deep traps per cluster	1
H	The number of emission centers per cluster	1
E_{ne}	The energetic distance from the excited state of the main trap to the conduction band (activation energy)	0.85 eV

TL curves (left) and TL dose dependences (right) calculated with different heating rates b

TL dose dependences with different cluster activation energy

TL output decreases with increasing the activation energy. As activation energy increases model conditions approach extreme case when localized transitions occur only in low temperature region.



TL dose dependences calculated with cluster activation energy E_{ne}

	Description	Value
N	The number of the main (TL-active) traps per cluster	2
M	The number of deep traps per cluster	1
H	The number of emission centers per cluster	1
	Heating rate	1 K/s

Conclusions

- TL dose dependence in cluster system with deep hole traps consists of several regions with different nonlinearity index and has non-monotonic region in some cases

- Localized transitions on the one hand can lead to a non-monotonic dose dependence, on the other hand, to the formation of temperature-dependent characteristics of dose curves and TL curves.

- The presence of deep hole traps leads to temperature-dependent nonlinearity of dose dependence

- The localized competition between hole traps and luminescence centers and energy distance from the excited state of the main trap to the conduction band influence on degree of non-monotony of TL dose dependence

Acknowledgement

This work was financially supported by the Russian Science Foundation (RSF), project No. 18-72-10082.

Thank you for attention!