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Sublinear dose dependence of thermoluminescence as a result of competition between electron and hole trapping centers

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HIGHLIGHTS

- A kinetic model of sublinearity of TL dose response is proposed.
- The sublinearity is due to the competition between electron and hole traps.
- Sublinearity occurs when the trap occupancy was far from saturation.
- The dependence of the linearity index on the model parameters is studied.

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ABSTRACT

This paper presents a model of sublinearity of thermoluminescence (TL) dose response, based on the competitive interaction between active electron trapping centers and deep hole centers during excitation and heating of a phosphor. It is shown by analytical and numerical calculations that the sublinear response is observed at low doses, when the trap occupancy is far from saturation. It is found that the linearity index of TL dose response depends on the concentration of the deep hole traps, and on the coefficient of non-radiative electron recombination. The TL dose response is derived in parametric form. A new analytical expression is derived for the threshold doses, at which the TL response changes from a sublinear growth to an almost linear behavior. This phenomenon of a sublinear TL response changing into a linear behavior, is explained by the deep traps filling up to saturation. The dependence of the linearity index on the model parameters is found by numerical methods.

1. Introduction

Dose dependence of luminescence yield is one of the important characteristics of materials used in thermoluminescence (TL) dosimetry of ionizing radiation. A linear TL dose response is a desirable property of dosimetric materials, since it provides a maximally simple calibration of a TL reader. However, non-linear TL dose response is observed in many luminescence dosimetry materials. In such non-linear dose response cases, the non-linearity can be quantitatively characterized by the numerical value and sign of the second derivative of the function of luminescence intensity on dose $I(D)$. If the second derivative is positive then superlinearity is observed, while for a negative second derivative sublinearity occurs (Chen and McKeever, 1994). If the TL dose dependence is described by relationship:

$$I \sim \alpha D^k \tag{1}$$

where α is a proportionality coefficient, then k is a constant known usually as the linearity index. Superlinearity occurs when $k > 1$, while for $k < 1$ sublinearity is observed. Clearly a linear TL dose response occurs when $k = 1$ (Halperin and Chen, 1966).

Sublinear TL dose response is not a critical obstacle to using the material in TL dosimetry, but it requires more complex calibration of the measurement device. In sublinearity cases, it is important to obtain an understanding of the underlying mechanisms, since this gives researchers the opportunity to manage the TL properties of the detector material, in order to improve the TL parameters and to optimize the dosimetric operation of the material.

Several examples of sublinear TL dose response are known from

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the literature in different materials. The most typical situation for phosphors is the one when the TL grows linearly with the irradiation dose in a rather long initial part of dose dependence. For higher doses, a sublinear region occurs and then the TL signal passes into saturation. Thus, the results of the study of TL in $K_2Ca_2(SO_4)_3$:Cu nanophosphor showed that the TL response of the sample annealed at 700 °C for 2 h and irradiated with different gamma doses shows linear behavior from 0.01 up to 300 Gy and becomes sublinear in the range of 300 Gy–1 kGy before it saturates with further increase in the dose (Mandlik et al., 2014). In some materials, the region of superlinearity can be a precursor to the sublinear part of dose dependence. For example, for α-Al_2O_3:C,Mg, the dose response of the main peak at 161 °C (heating rate is 1 °C/s) is superlinear in the range 0.1–30 Gy of beta dose, and then it becomes sublinear up to 100 Gy (Kalita and Chithambo, 2017). The linearity index analysis for Nigerian quartz showed that the TL responses of peaks at 170 and 226 °C (heating rate is 2 °C/s) were linear at low doses, grew supralinear as the dose was increased and then became sublinear at high doses (Ogundare et al., 2006). Most often, the sublinear part of the dose dependence observed at high doses is explained by the saturation of trapped charge carriers' concentration as the irradiation dose is increased. Another explanation commonly used is the onset of radiation damage processes.

In some cases, sublinearity can be observed on the initial part of the dose dependence. Thus, the X-ray dose response of LiF:Mg,Cu,P TL foils appears to be strongly sublinear below doses of ~0.1 Gy (Kopec et al., 2013). In kunzite ($LiAlSi_2O_6$) the dose response of peak at 338 K is highly sublinear at low doses as well (Ogundare et al., 2016). Such sublinear behavior can be caused by the effect of background emission in the TL reader (Kopec et al., 2013).

It is known that in some materials a large sublinear region occurs in a wide absorbed dose range which is located far enough from saturation. Thus, sublinear TL response is observed in Tm-doped silica over the whole analyzed dose range of 0.1–10 Gy for both 21 MeV electrons and 10 MV photons (Alawiah et al., 2015). Quantitative analysis of the dose response shows that the 573 K annealed sample of calcite showed sublinear dose response from 10 mGy to 1 Gy (Kalita and Wary, 2016). Sublinear TL dose responses were also observed in Al_2O_3:C single crystals and ultrafine anion-defective ceramics of aluminum and magnesium oxides irradiated by a high dose (1–800 kGy) of a pulsed electron beam (130 keV) (Nikiforov et al., 2014, 2016; Nikiforov and Kortov, 2014a).

Several attempts were made earlier in the literature to explain the presence of the sublinear part of TL dose response in a wide dose range, when the trap occupancy is far from saturation. It was found that sublinear TL dose response can be explained in terms of a simple two-level model (Lawless et al., 2009; Chen et al., 2010). The reason for the sublinearity effects in this simple model was found to be the competition between the trap and the recombination center for free electrons during irradiation. However, the simple two-level kinetic model is not suitable for the description of TL mechanisms in most phosphors which, as a rule, are characterized by the presence of several trapping and emission centers with different nature and energy depth.

A new mechanism of sublinear TL dose response was proposed by Nikiforov and Kortov (2014b). This mechanism was based on a kinetic model of competitive centers (Chen et al., 1996), and on competitive interaction between electron TL active traps and deep hole traps during irradiation and the heating of the samples. It was shown that the sublinearity can be observed when the occupancy of TL active traps is far from saturation. The preliminary results of numerical solution of the system of differential kinetic equations which describe this model during irradiation and heating stages showed that TL intensity can be proportional to $D^{0.40-0.62}$ for certain model parameters.

The main disadvantage of the numerical approach to model testing is that the calculation occurs for certain sets of parameters. This causes difficulties in the formulation of general conclusions from the simulation results, since other parameter values may yield different conclusions (Chen and Pagonis, 2014). In this case, it is recommended to use analytical methods of calculations alongside with the numerical ones, in order to improve the validity of the general findings from the model.

In this paper we apply the analytical approach to the sublinearity model proposed by Nikiforov and Kortov (2014b), for the first time. This is a more detailed study of the effects of various parameters in the model. In particular, we study in detail the effect of deep trap concentration and of the probability density of non-radiative recombination, on the degree of sublinearity characterized by the linearity index k.

The overall aim of this paper is the calculation of sublinear TL dose response using the model of competitive electron and hole traps. The calculations are carried out with the simultaneous use of the numerical and analytical approaches.

2. The model

The energy band diagram used in the present paper is identical to the one described in the recently published Nikiforov and Kortov (2014b) (Fig. 1). In this figure the electron and hole transitions during the excitation stage are shown with solid lines. The transitions during the heating stage are shown with dashed lines. During irradiation, electron-hole pairs are created (transition X). Holes in the valence band can be captured by either the emission center H (transition γ_2), or by the competitive center shown by the deep hole trap M (transition δ). There are three channels for electron relaxation for electrons in the conduction band: capture into the main TL active electron trap N (transition α), radiative recombination in the emission center (transition γ_1), and non-radiative recombination with a hole in trap M (transition β).

During heating, the electrons are released from trap N into the conduction band (transition P). Subsequently, they can be captured by the recombination center H followed by photon emission (transition γ_1), or they can recombine non-radiatively with holes in trap M, when this trap is previously occupied (transition β). Levels M and H are supposed to be thermally stable in the whole temperature range of TL measurements of the main trap N.

The systems of differential equations describing the model in the excitation (equations (2)–(6)) and heating (equations (7)–(11))

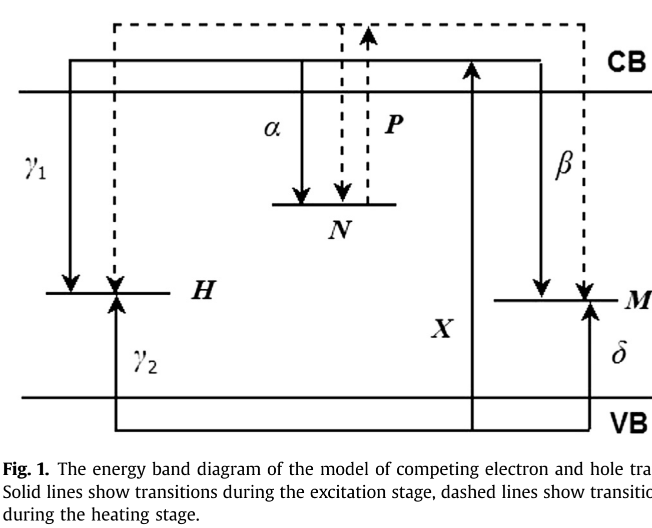

Fig. 1. The energy band diagram of the model of competing electron and hole traps. Solid lines show transitions during the excitation stage, dashed lines show transitions during the heating stage.

stages are presented below.

$$\frac{dn}{dt} = \alpha(N-n)n_c \tag{2}$$

$$\frac{dm}{dt} = \delta(M-m)n_v - \beta m n_c \tag{3}$$

$$\frac{dh}{dt} = \gamma_2(H-h)n_v - \gamma_1 h n_c \tag{4}$$

$$\frac{dn_c}{dt} = X - \alpha(N-n)n_c - \beta m n_c - \gamma_1 h n_c \tag{5}$$

$$\frac{dn_v}{dt} = X - \gamma_2(H-h)n_v - \delta(M-m)n_v \tag{6}$$

$$\frac{dn}{dt} = -Pn + \alpha(N-n)n_c \tag{7}$$

$$\frac{dm}{dt} = -\beta m n_c \tag{8}$$

$$\frac{dh}{dt} = -\gamma_1 h n_c \tag{9}$$

$$\frac{dn_c}{dt} = Pn - \alpha(N-n)n_c - \beta m n_c - \gamma_1 h n_c \tag{10}$$

$$I = \gamma_1 h n_c \tag{11}$$

Here X ($cm^{-3}s^{-1}$) is the efficiency of electron-hole pair's creation during irradiation, and its value is proportional to the dose-rate. N and M (cm^{-3}) are the total concentrations of main electron and deep hole traps, respectively. H (cm^{-3}) is the concentration of recombination centers; n, m, h are the occupancies of N, M, H levels, which are functions of time. n_c, n_v are the charge carrier concentrations in the conduction and valence bands. The coefficients α, β, γ_1, γ_2, δ (cm^3s^{-1}) characterize the probability densities of the corresponding transitions in Fig. 1. The parameter P=Sexp(-E/kT) is the rate of thermal emptying of traps N, while E (eV) is the energy depth and S (s^{-1}) is the frequency factor characterizing these traps. I ($cm^{-3}s^{-1}$) is the TL intensity.

The simulations using this model presented in Chen et al. (1996) showed that the TL response grows superlinearly with the dose, and depends on the concentration of holes in recombination centers (h). In this case superlinearity arises mainly due to the competition in the capture of holes between centers H and M during excitation. In the present paper, as well as in Nikiforov and Kortov (2014b), the model (Fig. 1) was studied at a non-zero initial occupancy of level H. Under these conditions, the changes of h values and the probability of γ_2 transition remain negligible for both excitation and heating stages. This minimizes the competitive interaction between centers H and M during irradiation. As a result, the electron processes associated with transitions β, α and γ_1, are going to play the main role in the formation of a non-linear TL dose response. Specifically the non-radiative transition β will be shown to cause sublinearity of the TL dose dependence.

In this paper we explain the sublinearity mechanism in some detail; the dose dependences of charge carriers' concentrations in traps, and the TL response are simulated by using two approaches: analytical and numerical. The cycle of calculation contains three successive stages of TL response formation, namely the excitation, relaxation and heating stages. The simulations were carried out repeatedly for increasing values of the irradiation time (t), which is equivalent to an increasing absorbed dose $D = Xt$. Every cycle uses the same initial conditions. The initial concentration of electrons in TL active traps and that of holes in deep traps were equal to zero ($n_0 = 0$; $m_0 = 0$). By comparison, in the related work by Nikiforov and Kortov (2014b), we did not take into account the hole accumulation in deep traps M from cycle to cycle, in order to simplify analytical calculations. In this previous study it was assumed that after each irradiation cycle the traps M were emptied, for example, as a result of high-temperature treatment. Moreover, the initial hole concentration in emission centers was taken to be non-zero in this study, and it was equal to $h_0 = 10^{14}$ cm^{-3}, as was also assumed earlier in Nikiforov and Kortov (2014b).

3. Analytical approach

First, we consider the analytical method of solution of equation systems (2)–(6) and (7)–(11). The method is based on some assumptions which simplify the mathematical description of the model. Assuming physically realistic rate constants and not exceedingly large dose rates, we can make the usual quasi-equilibrium approximation: $\frac{dn_c}{dt} \simeq \frac{dn_v}{dt} \simeq 0$ and n_c, $n_v \ll n, m, h$ (Chen and Pagonis, 2014). As earlier in Nikiforov and Kortov (2014b), we assume that the absorption dose in material is small enough that the trap occupancies are far from their maximal values ($m \ll M$, $n \ll N$). We also assume that the concentration of electron traps is relatively small in comparison with the concentration of hole centers ($N \ll H, M$) and H and M have the same order of magnitude. We limit ourselves to first order TL kinetics, when the probability of retrapping of electrons by trap N is relatively small. Moreover, we assume the initial occupancy of the luminescence centers (h_0) is not equal to zero (Nikiforov and Kortov, 2014b).

Let us consider the excitation stage. Taking into account quasi-equilibrium conditions and equating the left parts in equations (5) and (6) to zero, we obtain:

$$n_c = \frac{X}{\alpha(N-n) + \beta m + \gamma_1 h} \tag{12}$$

$$n_v = \frac{X}{\gamma_2(H-h) + \delta(M-m)} \tag{13}$$

With this, equations (2)–(4) can be rewritten as

$$\frac{dn}{dt} = \frac{\alpha(N-n)X}{\alpha(N-n) + \beta m + \gamma_1 h} \tag{14}$$

$$\frac{dm}{dt} = \frac{\delta(M-m)X}{\gamma_2(H-h) + \delta(M-m)} - \frac{\beta m X}{\alpha(N-n) + \beta m + \gamma_1 h} \tag{15}$$

$$\frac{dh}{dt} = \frac{\gamma_2(H-h)X}{\gamma_2(H-h) + \delta(M-m)} - \frac{\gamma_1 h X}{\alpha(N-n) + \beta m + \gamma_1 h} \tag{16}$$

From the condition of first order TL kinetics and assuming relatively small concentration of traps, it follows that $\alpha N \ll \gamma_1 h$, which results in the simplified equations:

$$\frac{dn}{dt} = \frac{\alpha N X}{\beta m + \gamma_1 h} \tag{17}$$

$$\frac{dm}{dt} = \frac{\delta M X}{\gamma_2(H-h) + \delta M} - \frac{\beta m X}{\beta m + \gamma_1 h} \tag{18}$$

$$\frac{dh}{dt} = \frac{\gamma_2(H-h)X}{\gamma_2(H-h)+\delta M} - \frac{\gamma_1 hX}{\beta m + \gamma_1 h} \quad (19)$$

When we add the left and right parts of equations (18) and (19), we obtain:

$$\frac{dm}{dt} = -\frac{dh}{dt} \quad (20)$$

The integration of both parts of this equation gives:

$$m - m_0 = h_0 - h \quad (21)$$

With the initial condition $m_0 = 0$ we have

$$h = h_0 - m \quad (22)$$

The m value is far from saturation ($m \ll M$), and when M and H have the same order of magnitude, we have $m \ll H$. Let the initial occupancy of the luminescence centers $h_0 = \eta H$. By assuming the η value to be significantly less than 1.0 ($\eta = 0.1$ according to Nikiforov and Kortov, 2014b), we have $H{-}h = H - h_0 + m \simeq H - h_0 = \text{const}$.

By using equation (22), equations (17) and (18) can be rewritten as:

$$\frac{dn}{dt} = \frac{\alpha NX}{\beta m + \gamma_1(h_0 - m)} \quad (23)$$

$$\frac{dm}{dt} = \frac{\delta MX}{\gamma_2(H-h_0)+\delta M} - \frac{\beta mX}{\beta m + \gamma_1(h_0 - m)} \quad (24)$$

Equation (24) can be integrated by using standard methods of solving first order non-linear differential equations. Due to the complexity of the algebra involved, the results from integrating equation (24) were checked using the software package *Mathematica.* By integrating equation (24), we obtain the following implicit function for the hole concentration in traps (m) versus the dose Xt:

$$B\left[m + \frac{D_1}{D_2}\ln\left(\frac{D_2 m}{A\gamma_1 h_0} + 1\right)\right] = Xt \quad (25)$$

where A, B, D_1 and D_2 are the constants, determined by equations:

$$A = \frac{\delta M}{\gamma_2(H-h_0)+\delta M} \quad (26)$$

$$B = \frac{\beta - \gamma_1}{D_2} \quad (27)$$

$$D_1 = \frac{\beta\gamma_1 h_0}{\gamma_1 - \beta} \quad (28)$$

$$D_2 = A\beta - A\gamma_1 - \beta \quad (29)$$

In order to find the dose dependence of the trapped electron concentration (n), we multiply both parts of equation (23) by $\frac{dt}{dm}$:

$$\frac{dn}{dt}\frac{dt}{dm} = \frac{\alpha NX}{\beta m + \gamma_1(h_0 - m)}\frac{dt}{dm} \quad (30)$$

Substituting $\frac{dm}{dt}$ from equation (24) into the right hand side of equation (30), we obtain:

$$\frac{dn}{dm} = \frac{\alpha N(\gamma_2(H-h_0)+\delta M)}{\delta M\gamma_1(h_0 - m) - \gamma_2(H-h_0)\beta m} \quad (31)$$

By integrating, we obtain:

$$n = \frac{\alpha N}{D_2}\ln\left(\frac{D_2 m}{A\gamma_1 h_0} + 1\right) \quad (32)$$

Equations (25) and (32) are the desired results from the analytical solution of the system of differential kinetic equations which describe the charge transfer processes in the model (Fig. 1) during the excitation stage. The dependences of the carrier concentrations in traps N and M on the dose ($D = Xt$) are determined by equations (32) and (25), respectively. Specifically, the dependence $m(D)$ is expressed in an implicit form according to equation (25), and is used to calculate the dependence $n(D)$ by using equation (32).

It is assumed and is verified by simulation, that the change of captured electron and hole concentrations during relaxation stage is negligibly small. Let us now consider the analytical description of charge transfer processes during the heating stage. We obtain for the integrated TL intensity:

$$Is = \frac{\gamma_1 h_1}{\beta m_1 + \gamma_1 h_1} n_1 \quad (33)$$

In this equation Is value presents a TL output (lightsum) in the peak of TL-active trap N. Equation (33) can be obtained directly as follows, by using the method of calculating the integrated TL in Chen et al. (2011). These authors used the same assumptions as in the present paper, i.e. the quasi-equilibrium approximation and the trap occupancy which is far from saturation There are two possible pathways for the electrons released from trap N during the TL measurement. The fraction of electrons which radiatively recombine in centers H is proportional to $\gamma_1 h_1$ (for transition γ_1). The fraction of electrons which recombine non-radiatively with holes in deep traps M is fraction βm_1 (for transition β). The integrated TL intensity in equation (33) is then determined approximately by the ratio of $\gamma_1 h_1$ over the sum $(\gamma_1 h_1 + \beta m_1)$, multiplied by the concentration of electrons n_1 released during the TL measurement.

4. Numerical approach

The systems of differential equations (2)–(11) were solved by the second order Gear method (Gear, 1971). During the solution of the system (2)–(6) for the excitation stage, the absorbed dose was determined by varying the irradiation time t with a constant value of X. The concentrations of free and captured electrons and holes at the end of the irradiation process were calculated. During the subsequent relaxation stage, the system of equations (2)–(6) was solved again with $X = 0$ in equations (5) and (6). The initial values of concentrations n, m, h, n_c, n_v for the relaxation stage were determined by the results of calculation for the excitation stage. The relaxation time was increased until n_c and n_v values became close to zero. The concentration values of charge carriers at the end of the relaxation period were used as the initial values for solving the system of equations (7)–(11) for the heating stage. The glow curve $I(T)$ was calculated for the temperature change from 500 to 800 K with a heating rate of 2 K/s (Nikiforov and Kortov, 2014b).

5. Discussion

We used the following parameter values for analytical and numerical simulations of dose dependences of the captured electron and hole concentrations after the irradiation and TL response

(Nikiforov and Kortov, 2014b): $X = 5\cdot10^{11}$ cm^{-3}s^{-1}, $E = 1.95$ eV, $S = 10^{13}$ s^{-1}, $\alpha = 10^{-14}$ cm^{3}s^{-1}, $\gamma_1 = 5\cdot10^{-13}$ cm^{3}s^{-1}, $\gamma_2 = 5\cdot10^{-13}$ cm^{3}s^{-1}, $H = 10^{15}$ cm^{-3}, $N = 5\cdot10^{11}$ cm^{-3}s^{-1}, $h_0 = 10^{14}$ cm^{-3}. The deep trap concentration M and the probability of non-radiative recombination β in traps M were varied, in order to study their effect on the sublinearity of the TL dose response. The M values were increased by an order of magnitude in comparison with the paper (Nikiforov and Kortov, 2014b), in order to satisfy more accurately the condition $m \ll M$. In this study, the transition constant δ was reduced to 10^{-12} cm^{3}s^{-1} in comparison with $\delta = 10^{-11}$ cm^{3}s^{-1} in Nikiforov and Kortov (2014b).

Fig. 2 shows the simulation results obtained by the analytical method (lines), and also by the numerical solution of the system of equations (symbols). The *Xt* value which characterizes the absorbed dose, is given on the X axis. The calculations were made for $M = 10^{15}$ cm^{-3} and $\beta = 10^{-11}$ cm^{3}s^{-1}. Very good agreement is found between the numerical and analytical calculations.

Curve 1 in Fig. 2 shows that the hole concentration m in deep traps M increases with the irradiation dose during the excitation stage. As the concentration of holes m increases, the probability of electron capture from the conduction band into the deep trap M also increases (transition β). As a result, the probability of electron capture by active traps (transition α) decreases. Curve 2 in Fig. 2 shows that the electron concentration in traps N after the irradiation stage behaves sublinearly for small doses, and this causes the sublinear behavior of TL response (curve 3). The TL response has a more pronounced sublinearity, because the slope of curve 2 is slightly higher than the slope of curve 3. This is due to the fact that the decrease of the number of radiative recombinations in center H (transition γ_1) during the heating stage, gives an additional contribution to the sublinearity of TL response due to the increasing probability of transition β.

Curves 2 and 3 in Fig. 2 also show that at certain irradiation dose the dose dependences of n and of the integrated TL signal become steeper, indicating a higher linearity index. When the dose reaches certain a threshold value ($D_{\rm thr}$), the parameter k stops changing and becomes close enough to 1.0 (dashed line in Fig. 2). Moreover, it is seen that when the dose is equal to $D_{\rm thr}$, the hole concentration in deep traps (m) reaches saturation (curve 1).

In Fig. 2 it is clearly seen that the saturation of deep hole trap occupancy m is observed when $m \ll M$. This is a rather peculiar behavior which be interpreted as follows. The saturation of m value is associated with the equilibrium between the processes of increasing hole concentration during the irradiation stage (transition δ), and a decrease of the value of m as a result of non-radiative recombination (transition β). At a dose of $D \sim D_{\rm thr}$ shown in Fig. 2, competition between these two opposite effects becomes weaker after, and as a consequence the slopes of the dose dependences of n and TL increase. These dependences *n(D)* and *TL(D)* become almost linear at $D > D_{\rm thr}$. Such growth of linearity index of TL response at high doses (more than 100 kGy) after the sublinear region was observed earlier experimentally for the dosimetric TL peak at 450 K in Al_2O_3:C single crystals irradiated by high current pulsed electron beam (Kortov et al., 2014; Nikiforov and Kortov, 2014a). Taking into account the presence of a high number of deep electron or hole traps in such crystals (Yukihara et al., 2003; Nikiforov et al., 2014), it is proposed that the sublinearity mechanism proposed in the present paper is observed in this material. Within the model presented in this paper, the saturation of deep traps occupancy is one of the reasons of linear growth of TL response at $D > 100$ kGy. The creation of new trapping and recombination centers during high-dose irradiation can be another factor which causes sharp growth of TL output (Kortov et al., 2014). The anomalous increase in TL yield is registered after the irradiation of Al_2O_3:C crystals by high dose of pulsed electron beam (130 keV) at the dose interval from 80 to 800 kGy. Simultaneously the intensive band appears in the green spectrum region in the photoluminescence spectrum of the crystals under study, associated with aggregate F_2-type centers. The role of aggregate defects in the increase of TL yield in Al_2O_3:C crystals under high-dose irradiation is discussed by Kortov et al. (2014).

Let us now consider the effect of the model parameters β and M, on the dose dependences of captured electron and hole concentrations, as well as on the TL response in the region where $D < D_{\rm thr}$. The obtained results of calculations are shown in Fig. 3 – 5, and the variable model parameters (β and M) are presented in Table 1. Curves 1 in Figs. 3–5 correspond to the initial model parameter values which were used to obtain the data of Fig. 2. Good agreement of analytical and numerical methods for the calculated curves in Figs. 3–5 is noted. We denote by k_1 the linearity index of the TL dose response for low doses ($D = Xt = 5\cdot10^{12}$–$5\cdot10^{13}$ a.u.). The value of k_1 is calculated by the fitting of the TL dose dependences (Fig. 5) in the above mentioned region by formula (1).

When the value of the transition coefficient β is reduced (curves 2 in Figs. 3–5), the probability of competitive non-radiative

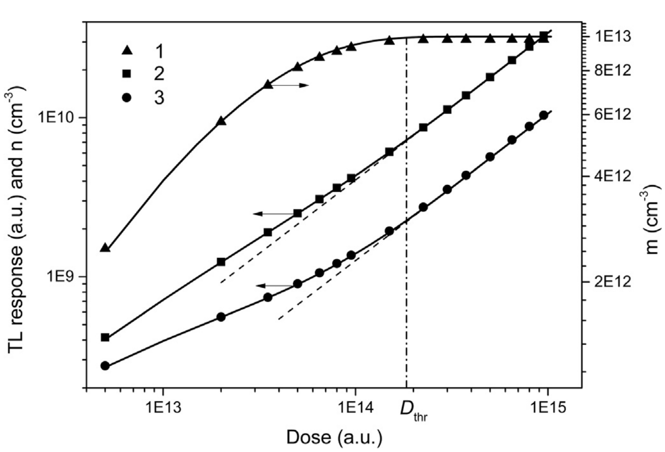

Fig. 2. The dose dependences of carriers' concentration of hole traps m (Curve 1), electron traps n (Curve 2), and TL response (Curve 3). The model parameters are presented in the text. Solid lines show the results of analytical approach, symbols show the results of numerical approach by solving the system of differential equations. D_{thr} is the threshold dose at which the linearity index becomes close enough to 1.0.

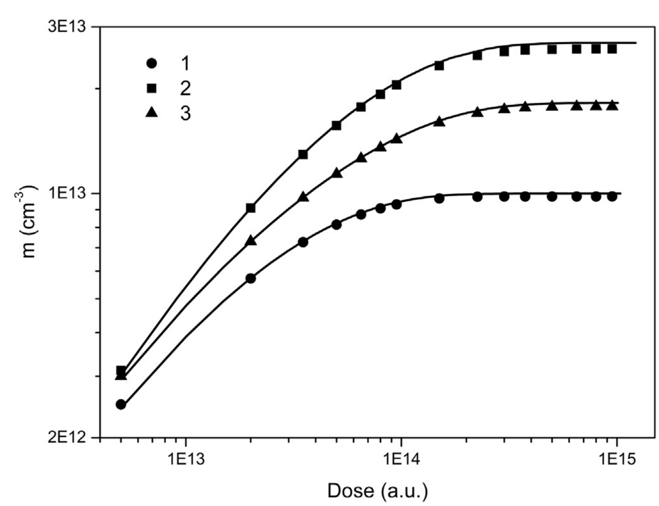

Fig. 3. The dependence of hole concentration m on the irradiation dose. The curve numbers correspond to those given in Table 1. Solid lines show the results of analytical approach, symbols show the results of numerical approach.

Table 1
The results of parameters calculation of TL dose response.

Number of curve in Fig. 3–5	β, cm^{3}s^{-1}	M, cm^{-3}	Linearity index in the initial region of irradiation dose, k_1	Linearity index at the $D > D_{thr}$, k_2	m_{max}, cm^{-3}	The graphically determined threshold dose D_{gr}, a.u.	The analytically determined threshold dose D_{an}, a.u.
1	10^{-11}	10^{15}	0.48	0.92	$1\cdot10^{13}$	$1.8\cdot10^{14}$	$1.7\cdot10^{14}$
2	$3\cdot10^{-12}$	10^{15}	0.55	0.88	$2.7\cdot10^{13}$	$3.5\cdot10^{14}$	$3.7\cdot10^{14}$
3	10^{-11}	$2\cdot10^{15}$	0.36	0.86	$1.8\cdot10^{13}$	$3.8\cdot10^{14}$	$3.8\cdot10^{14}$

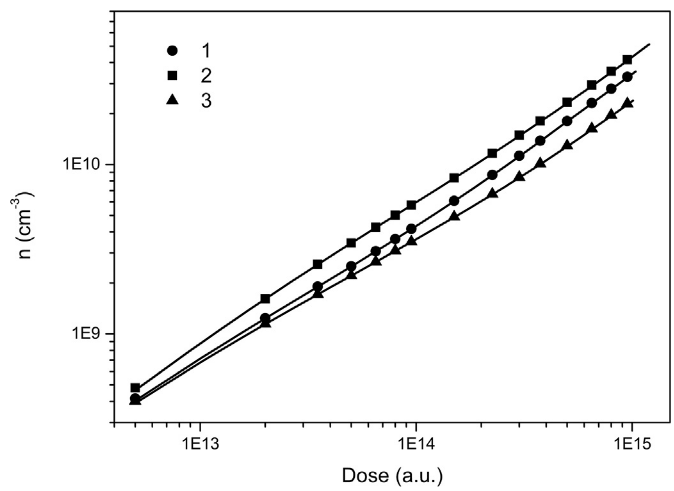

Fig. 4. The dependence of electron concentration n on the irradiation dose. The curve numbers correspond to those given in Table 1. Solid lines show the results of analytical approach, symbols show the results of numerical approach.

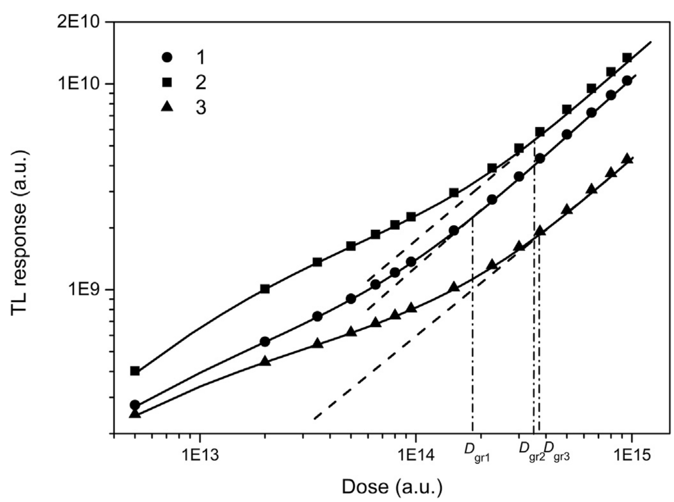

Fig. 5. The dose dependences of TL response. The curve numbers correspond to those given in Table 1. Solid lines show the results of analytical approach, symbols show the results of numerical approach. The D_{gr1}, D_{gr2} and D_{gr3} are the threshold doses for curve 1, 2 and 3 respectively, at which the linearity index becomes close enough to 1.0.

recombination of electrons with holes in trap M decreases. In this case the deep hole occupancy m and n increases during the excitation stage (Figs. 3 and 4). As a result, the TL response grows and the slope of the TL dose response becomes steeper (Fig. 5). This corresponds to higher linearity index k_1 which shows a value changing from 0.48 (curve 1) to 0.55 (curve 2) (Fig. 5 and Table 1).

When parameter β decreases significantly (and becomes much less than transition coefficient γ_1), competition for electron capture by traps M becomes negligible. In this case, we obtain from the formulas (23) and (24):

$$\frac{dn}{dt} = \frac{\alpha NX}{\gamma_1(h_0 - m)} \quad (34)$$

$$\frac{dm}{dt} = \frac{\delta MX}{\gamma_2(H - h_0) + \delta M} \quad (35)$$

Since $\frac{dm}{dt} = const$, the m value increases linearly and the denominator in expression (34) decreases with time, which causes an increased value of $\frac{dn}{dt}$, and the occupancy of TL active traps (n) changes slightly superlinearly and the TL dose response has a linearity index close to 1 (k_1~1). The dependence of linearity index of TL dose response k_1 on the coefficient of non-radiative recombination β is shown in Fig. 6 (for a fixed value of $M = 10^{15}$ cm^{-3}). It can be seen that k_1 increases from 0.48 to 1.0 when the β value changes from 10^{-11} to 10^{-14} cm^{3}s^{-1}.

Let us also consider the effect of deep hole trap concentration M on the TL dose response (curves 1 and 3 in Figs. 3–5). When the parameter M increases, the capture probability of the holes in the valence band by these traps grows during the excitation stage and their occupancy increases (Fig. 3). The increase of M has a stronger competitive effect on electron transfer in the conduction band, than the increase of the transition coefficient β. As a result, the occupancy of electron traps (n) drops (Fig. 4, curves 1 and 3). The slope of the TL dose response decreases on the initial part (Fig. 5) and the linearity index k_1 changes from 0.48 to 0.36 (Table 1). The dependence of linearity index k_1 on deep hole traps concentration M is presented in Fig. 7 (for a constant value of $\beta = 10^{-11}$ cm^{3}s^{-1}). It is seen that the k_1 value decreases from 0.62 to 0.31 when the M value changes from $5\cdot10^{14}$ to $3\cdot10^{15}$ cm^{-3}.

Let us also analyze the features of the TL dose response in the region when the slope of curves changes, at $D > D_{thr}$. Let us denote

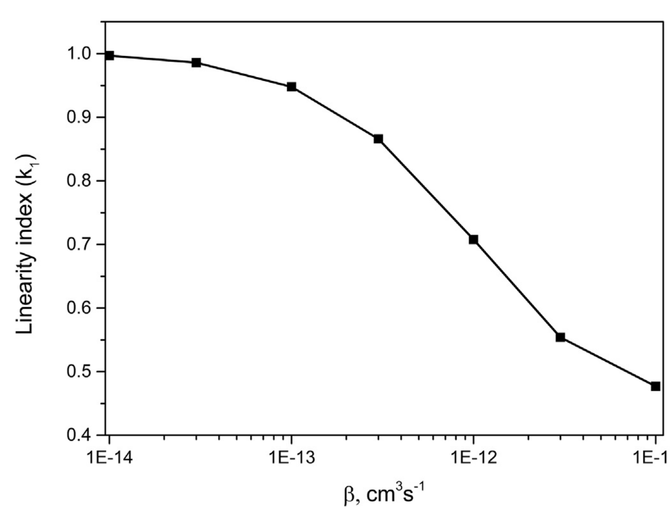

Fig. 6. The dependence of the linearity index in the initial region of irradiation dose, on the non-radiative recombination coefficient β.

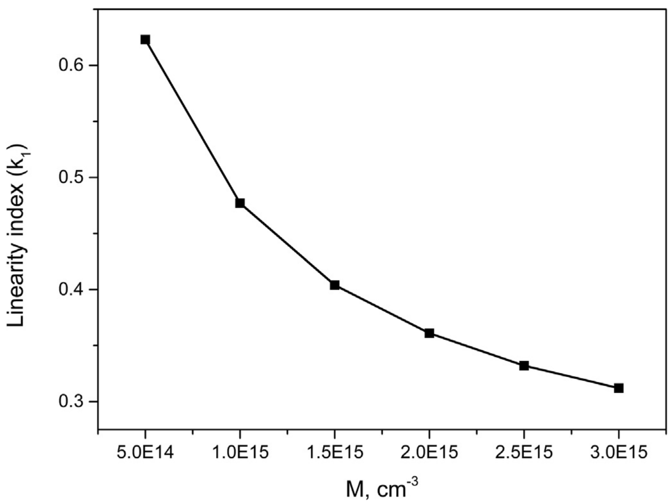

Fig. 7. The dependence of the linearity index in the initial region of irradiation dose, on the concentration of deep hole traps *M*.

the linearity index at $D > D_{thr}$ as k_2. The value of k_2 is calculated by the fitting of the TL dose dependences (Fig. 5) in the region where $D > D_{thr}$ by formula (1). The calculation results show that the linearity index k_2 changes insignificantly, and the TL dose response is close to linear (k_2~0.9) (Table 1). From the data of Fig. 5, the threshold doses D_{gr} for curves 1–3 were estimated graphically and their values are shown in Table 1.

The analytical approach used in the present paper for simulation allows us to estimate these threshold doses D_{thr} using the values of the parameter in the model. We can calculate the maximum hole concentration in traps M (m_{max}), by equating $\frac{dm}{dt}$ in (24) to zero:

$$m_{max} = \frac{\delta M \gamma_1 h_0}{\beta \gamma_2 (H - h_0) + \gamma_1 \delta M} \quad (36)$$

The calculated m_{max} values for different model parameters are present in Table 1. It is noteworthy that they are very close to the m values in the saturation region of curves 1–3 in Fig. 3. In this case, it should be taken into account that $m = m_{max}$ value is a limiting value of m, when $D \to \infty$. Let us assume that the threshold dose corresponds to $m = 0.99m_{max}$. Substituting these m values in formula (25), which represent the implicit function of m value on dose, we can analytically calculate the threshold doses (D_{an}) which correspond to curves 1–3 in Fig. 5. The obtained D_{an} values are shown in Table 1. Comparison between the graphical and analytical calculation results of threshold doses (D_{gr} and D_{an}) shows a good agreement of the values obtained from the two methods. Thus, the use of analytical approach to simulation, which was implement for the first time for the model under study in the present paper, allows estimating the threshold doses at which significant improvement of linearity of TL dose response is observed. In this case the calculation is based on the model parameters, and plotting of the TL dose dependences is not required. The obtained results show that it is possible to manage the linearity index of TL dose response by changing the trap concentration and transition coefficients, in order to optimize the dosimetric properties of materials used for ionizing radiation measurements.

6. Conclusions

In this paper the mechanism of sublinear TL dose response is explained by analytical methods and by numerical solutions of the system of differential kinetic equations. The sublinearity is due to competitive interaction between TL active electron traps and deep hole traps during the excitation and heating stages. An important feature of the model is the fact that it explains sublinear growth of TL response when the trap occupancy is far from saturation. It was found that the linearity index depends on the coefficient of non-radiative recombination of electrons in deep hole traps (β), and on their total concentration *(M)*. It was shown that an analytical method can be used to find the threshold doses, at which the sublinear growth of TL response becomes almost linear. Good agreement of analytical and numerical results is obtained. This confirms that the proposed model explains the relationship between the concentration of charge carriers and the sublinear TL dose response. The proposed model is universal and can be applied to explain sublinear dose dependencies of TL peaks in different materials. A necessary condition for its applicability is the presence of thermally disconnected deep traps in the material under study which is a very common phenomenon in dosimetric and dating materials (Al_2O_3:C; LiF: Mg,Cu,P; SiO_2; CaF_2:Dy et al.) (McKeever et al., 1995).

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