Radiation Measurements 132 (2020) 106257

Contents lists available at ScienceDirect

Radiation Measurements

journal homepage: http://www.elsevier.com/locate/radmeas

A Monte-Carlo study of the fading of TL and OSL signals in the presence of deep-level competitors

R. Chen [a,*], V. Pagonis [b]

[a] Raymond and Beverly Sackler School of Physics and Astronomy, Tel-Aviv University, Tel-Aviv, 69978, Israel
[b] Physics Department, McDaniel College, Westminster, MD21157, USA

ARTICLE INFO

Keywords:
Optically stimulated luminescence
Thermoluminescence
Exponential and non-exponential decay
Monte-Carlo method
Fading

ABSTRACT

In a previous paper, the issue of the evaluated lifetimes of thermoluminescence (TL) and optically stimulated luminescence (OSL) has been studied for the one-trap-one-recombination-center (OTOR) case, using the Monte-Carlo simulation. It was shown that under these circumstances, the decay curve of the electron occupancy along many thousands of years may not be exponential. Therefore, a lifetime determined from the results at short periods of time may not apply at longer periods of time. The decay at longer times was found to be slower than exponential and thus, one may observe longer lifetimes than predicted by the evaluated trapping parameters. In the present work we demonstrate that with a more complex model, namely, when an additional deeper trap is involved, the probability of getting an exponential decay of the signal is much larger. We study the fading of OSL and TL signals with different times elapsing between excitation and read-out under these circumstances, using a Monte-Carlo procedure, and show that with a significant deep-trap competitor, the extrapolation leading to the evaluation of the long-term stability of the signal is more viable. The results are compatible with previously existing evidence that the chances of having a TL peak with first-order characteristics are significantly larger in cases where a large deep trap, acting as a competitor, is present.

1. Introduction

The use of the Monte-Carlo algorithms for the study of TL has first been proposed in a series of papers by Mandowski and Świątek (1992, 1996; 2000), Mandowski (2001a,b; 2006; 2008) and Mandowski et al. (2010). As pointed out by these authors, usually, the number of free carriers in a sample is large and the kinetic equations describe the system properly. However, in some cases, in microcrystalline or two-dimensional solids, where each grain or plane can be considered as a separate system, one should consider each carrier separately, and the continuous differential equations may not be used. Mandowski et al. have proposed the use of the Monte-Carlo simulation for these cases. Another reason for the use of the Monte-Carlo method for the study of TL has been given by Kulkarni (1994) who stated that if the lifetime of electrons in the conduction band is very small, the conventional numerical calculation will take very long time and the use of the Monte-Carlo procedure may bypass this problem. Yet another use of the Monte-Carlo approach has been suggested by Rodríguez-Villafuerte (1999) who described its applicability for the track-interaction model which may explain the supralinearity in the TL response. Bailey (2004) has used the Monte-Carlo approach to monitor the evaluated equivalent dose in quartz in a complex model including 12 electron- and hole-trapping levels. Monte-Carlo simulations of optically stimulated luminescence (OSL) have been discussed by Thompson (2007) who has simulated the single aliquot regeneration OSL dosimetry measurements.

In a more recent paper, Chen and Pagonis (2015) have reported the results of Monte-Carlo simulations which follow the fading of thermoluminescence (TL) and optically stimulated luminescence (OSL) signals in cases where the sample is held at relatively low temperature, e.g., room temperature (RT). The Monte-Carlo method has been used since at the temperature at which the sample is held, the average number of electrons in the conduction band is significantly smaller than unity and the correct way to deal with the released electrons is one at a time, according to the relevant probability of their thermal release. Once the electron is determined to have reached the conduction band, according to the one-trap-one-recombination center (OTOR) model discussed by Chen and Pagonis (2015), it can either retrap or perform recombination with a hole in the center. This second stage of retrapping or recombination was also monitored by the Monte-Carlo procedure. The results showed an exponential decay only in cases of dominating

* Corresponding author.
E-mail address: chenr@tauex.tau.ac.il (R. Chen).

https://doi.org/10.1016/j.radmeas.2020.106257
Received 20 October 2019; Received in revised form 17 January 2020; Accepted 29 January 2020
Available online 31 January 2020

recombination. In cases where retrapping was significant, approximately exponential decay was seen only in portions of the time periods of fading, and different fading functions were found in other time ranges. For instance, in one example, the decrease in the number of remaining electrons in traps was found to be approximately exponential during the first 500 years of fading, but continued with a slower decay at longer times; see Fig. 3 in Chen and Pagonis (2015). Note that this behavior can be considered as "anomalous stability". From the initial decline, one expects the line to continue in an exponential decay, but actually, at longer periods of time, the decay is significantly slower. It should be noted that decay times of hundreds and thousands years are relevant to the TL and OSL study of small grains (e.g. of quartz) in archaeological and geological samples.

In the present work, we go beyond the OTOR model and add to the energy-level model a deeper trap, N_d, which may compete for free electrons with both the active trap and the recombination center. The motivation for this is that when a similar model is used for the study of TL peaks, the addition of a significant deep trap has usually resulted in the occurrence of first-order kinetics (Lewandowski and McKeever, 1991; Sunta et al., 1999, 2001; 2005; Pagonis and Kitis, 2012). Chen and Pagonis (2013) have shown that in the case of the occurrence of a series of TL peaks associated with a series of traps, chances are that all the peaks except for the last in the series, will have the features of a first-order curve. Intuitively, one may expect that the long-term fading of the TL and OSL signals be exponential in the presence of deep competing traps. Due to the nature of the very slow decay at relatively low temperature (usually room temperature, RT), this process has been followed using Monte-Carlo simulation. Note that due to the more complex nature of the model, one cannot choose arbitrarily the concentrations of the trapped carriers at the beginning of the decay process. Instead, one has to run the relevant set of coupled differential equations governing the excitation stage for a certain period of excitation time t_D using the relevant trapping parameters, and use the final distribution of the electrons between the two traps and the concentration of holes in centers as an initial condition for the next stage. The same is true for the use of the final occupancies of traps and centers at the end of the long fading as initial values for the pulsed-OSL or TL stage.

2. The model

Fig. 1 shows a schematic energy-level model including an active trap N, a deep competing trap N_d and a hole center M with occupancies of n, n_d and m, respectively. The meaning of all the relevant parameters is given in the caption to Fig. 1. Note that the single deep trap N_d may represent a number of competing deep traps. All the transitions taking place during the excitation, the long decay and the OSL read-out are shown. In the previously described OTOR model (Chen and Pagonis, 2015), we considered the situation where, following excitation, the traps and centers are equally populated and their decay during long time was examined by a Monte-Carlo procedure. Since we are dealing here with two traps and one center, the situation is different. At the end of excitation, the sum of concentrations of electrons in the two kinds of traps must be equal to the concentration of holes in centers. However, one cannot determine arbitrarily the distribution of electrons between the two traps. Instead, we have solved numerically the relevant set of simultaneous differential equations for a given set of trapping parameters. The concentrations of electrons in traps and holes in the center at the end of the excitation are used as initial values for the next stage of long-time decay which is performed by a Monte-Carlo procedure. The final values of the carriers' concentrations were used in the next stage of simulation of the OSL pulse read-out. Note that at this stage also we allow the optical release of an electron from the active trap into the conduction band, followed by either radiative recombination with a trapped hole, trapping in the competitor or retrapping into the active trap. It should also be noted that we are mainly interested in the dependence of the sensitivity of the sample to the stimulating light pulse following different decay times. Therefore, although the concentration of carriers varies during the exposure to stimulating light, we do not consider this variation while proceeding to longer decay times. In other words, we are simulating here the multiple aliquot technique.

The relevant set of equations governing the processes is:

$$\frac{dn}{dt} = A_n(N-n)n_c - s\cdot n\cdot\exp(-E/kT) - f\cdot n, \tag{1}$$

$$\frac{dn_d}{dt} = A_{nd}(N_d - n_d)n_c, \tag{2}$$

$$\frac{dn_v}{dt} = X - B\cdot(M-m)\cdot n_v, \tag{3}$$

$$\frac{dm}{dt} = B\cdot(M-m)\cdot n_v - A_m\cdot m\cdot n_c, \tag{4}$$

$$\frac{dn_c}{dt} = \frac{dm}{dt} + \frac{dn_v}{dt} - \frac{dn}{dt} - \frac{dn_d}{dt}. \tag{5}$$

Note that this set of equations can be adapted to the different stages of the process. During excitation, we set $f = 0$ and $X > 0$, proportional to the intensity of the irradiation. During OSL read-out, we set $X = 0$ and f is proportional to the stimulating-light intensity. During TL read-out as well as during isothermal decay of the relevant phenomena, both X and f are assumed to be nil. In TL, the temperature varies during read-out, usually with a linear heating function $T=T_0+\beta t$ where T(K) and T_0(K)

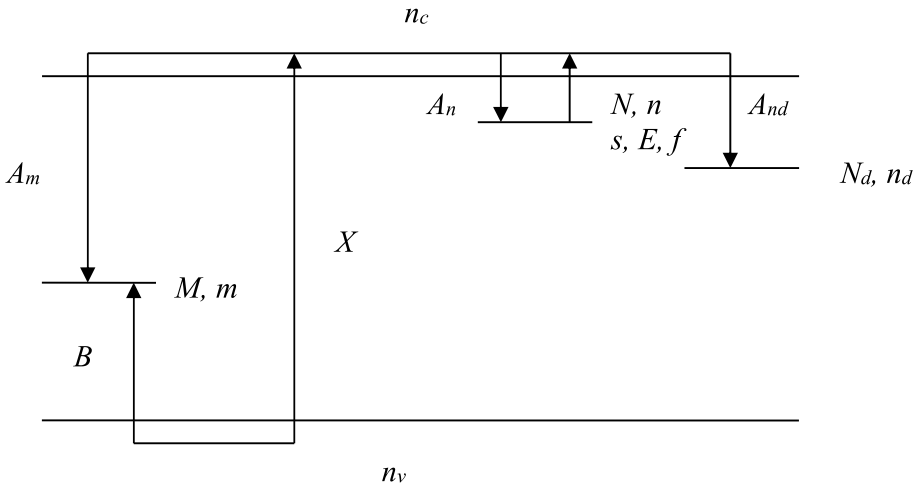

Fig. 1. Energy level diagram including one hole center M (cm^{-3}) with occupancy m (cm^{-3}) and two electron trapping states, N (cm^{-3}), an active trap with instantaneous occupancy n (cm^{-3}), and a deeper, competing trap N_d (cm^{-3}) with instantaneous occupancy of n_d (cm^{-3}). X ($cm^{-3}s^{-1}$) is the rate of production of electron and hole pairs during excitation which is proportional to the dose-rate. A_m, A_n and A_{nd} (cm^3s^{-1}) are the probability coefficients for the recombination, retrapping and trapping in deep traps, respectively and B (cm^3s^{-1}) is the trapping coefficient of free holes in centers during excitation. E (eV) is the activation energy of the trap N and s (s^{-1}) its frequency factor. f (s^{-1}) is the factor associated with the optical release of trapped electrons which is proportional to the intensity of the stimulating light in OSL.

are the variable temperature and initial temperature, respectively, and β (Ks^{-1}) the constant heating rate. Note that in the two latter cases, $n_v \equiv 0$ and therefore Eq. (3) is superfluous in these stages. Note also that, as explained by Chen and Pagonis (2015) and as pointed out above and elaborated below, in the case of very slow decay, one would choose to follow a Monte-Carlo procedure rather than solving the equations numerically.

3. Theoretical considerations

The rate of release of electrons from active traps during the long period of time between excitation and read-out follows the first-order equation

$$\frac{dn}{dt} = -s \cdot n \cdot \exp(-E/kT). \quad (6)$$

However, when we consider the remaining concentration of electrons in traps following a certain fading time, we are interested only in the fraction of these electrons that do not perform retrapping, and therefore, the net equation is

$$\frac{dn}{dt} = -s \cdot n \cdot \exp(-E/kT) \frac{A_m m + A_{nd}(N_d - n_d)}{A_m m + A_n(N-n) + A_{nd}(N_d - n_d)}. \quad (7)$$

Let us denote the expression on the right-hand side by F, namely,

$$F = \frac{A_m m + A_{nd}(N_d - n_d)}{A_m m + A_n(N-n) + A_{nd}(N_d - n_d)}, \quad (8)$$

and then Eq. (7) reads

$$\frac{dn}{dt} = -s \cdot n \cdot \exp(-E/kT) \cdot F. \quad (9)$$

It is obvious that if the deep competitor is small, $N_d \sim 0$, or if it has a very small trapping probability, $A_{nd} \sim 0$, the last term in the numerator and in the denominator of Eqs. (7) and (8) is negligible, and we are back to the OTOR situation which usually yields non-exponential decrease of the concentration of active traps, n, as shown by Chen and Pagonis (2015). In other cases, one has to consider whether with a given set of the parameters, the function F is approximately constant and if so, is its value close to unity. For example, if both the active trap and the competitor are in saturation, the only remaining term in the numerator and denominator is $A_m m$ which therefore cancels out. The remaining expression is the simple first-order equation which thus governs a simple exponential decay of the concentration n which, in turn, leads to an exponential fading of the OSL signal.

Other situations in which the function F is approximately constant can be considered. If the competitor is far from saturation, namely, $n_d \ll N_d$ and $A_{nd}N_d >> A_m m$ and $A_{nd}N_d >> A_n(N\text{-}n)$ and if m changes very slowly, F is approximately constant and therefore, the concentration n decays approximately exponentially along the years following excitation. It is quite obvious from this discussion that the question of whether the decay is expected to be exponential or not depends on all the trapping parameters as well as on the filling of traps and centers, but with the existence of a strong competitor, the chances of having an exponential decay are significantly larger.

It should be noted that so far, we have considered only the time dependence of the concentration of trapped electrons n, whereas our main interest is in the fading of the OSL signal. When a stimulating-light is applied, the electrons raised into the conduction band may perform recombination, retrapping or trapping into the competing trap. However, as long as the stimulating light pulse is short, the variation of the relevant concentrations is not large and therefore, the number of recombinations is proportional to n, and therefore, it is expected to decay exponentially. Nevertheless, the numerical procedure described below does not rely on this hand-waving argument and as stated above, the OSL intensity is found by solving numerically the relevant set of coupled differential equations.

4. The numerical procedure

As pointed out above, the first stage consists simply of choosing a certain feasible set of trapping parameters and solving the set of differential equation at a constant temperature with a certain value of the excitation dose rate X and $f = 0$, for a certain period of time t_D, using the ode15s Matlab solver. The total dose is given by $D = X \cdot t_D$. The final concentrations of trapped electrons and holes, n, n_d and m are used as initial values for the next stage.

The second stage involves the thermal release of the electrons from traps as explained by Chen and Pagonis (2015). If the sample is held at a temperature T and if the activation energy is E and the frequency factor is s, the probability of release of an electron *per second* is $s \cdot \exp(-E/kT)$. If we consider an infinitesimal time interval dt, the probability for the release is the dimensionless quantity $s \cdot \exp(-E/kT) \cdot dt$. Obviously, the Monte-Carlo procedure cannot work with infinitesimal time intervals and dt should be approximated by a finite time interval Δt. This interval should be short enough so that the magnitude $s \cdot \exp(-E/kT) \cdot \Delta t$ be significantly smaller than unity, and thus be a good enough approximation to the aggregate of the infinitesimal probabilities $s \cdot \exp(-E/kT) \cdot dt$. At the same time, it should be long enough so that coverage of time ranges of, say, thousands of years, will not take a prohibitively long computer time. Chen and Pagonis (2015) chose Δt to be one year; the considerations made for taking this period of time are valid also in the present case. Once we have a certain initial concentration of trapped electrons, we consider each of them separately. We draw a Matlab-generated pseudo-random number between zero and 1 for each electron and if this random number is smaller than $s \cdot \exp(-E/kT) \cdot \Delta t$, this electron is determined to have been raised into the conduction band, and therefore, it is reduced from the number of electrons in the trap. Once all the electrons have been tested, we register the final number of remaining electrons in traps, go to the next time interval Δt and repeat the procedure with the remaining electrons as the new initial value.

Let us consider now the more complex situation where retrapping and trapping into a deeper, competing trap are allowed. Once an electron has been raised into the conduction band, its probabilities of performing recombination, retrapping and deep trap capture are proportional to $A_m m$, $A_n(N\text{-}n)$ and $A_{nd}(N_d\text{-}n_d)$, respectively. Thus, the probability for recombination is $A_m m/[A_m m + A_n(N\text{-}n) + A_{nd}(N_d\text{-}n_d)]$, the probability for retrapping is $A_n(N\text{-}n)/[A_m m + A_n(N\text{-}n) + A_{nd}(N_d\text{-}n_d)]$ and that for deep trap capture is $A_{nd}(N_d\text{-}n_d)/[A_m m + A_n(N\text{-}n) + A_{nd}(N_d\text{-}n_d)]$. In the Monte-Carlo simulation, we start each step as in the previous case of pure first-order kinetics. Once an electron has been determined to be in the conduction band, we draw another pseudo-random number between zero and one; if $A_m m//[A_m m + A_n(N\text{-}n) + A_{nd}(N_d\text{-}n_d)]$ is smaller than this number, we determine that recombination has taken place, and the electron is subtracted from the trapped electron "box". Otherwise, the electron is considered to either retrap or be trapped in the deeper competitor. We therefore have to consider the conditional probability that an electron that has not performed recombination be trapped in the competitor. We draw now a third pseudo-random number between zero and one. If $A_n(N\text{-}n)/[A_n(N\text{-}n) + A_{nd}(N_d\text{-}n_d)]$ is smaller than this number, we determine that the electron has been retrapped and else, it has been trapped in the deep competitor. The process is repeated for the current time interval for all the electrons in the trap. When this is finished, the new number of electrons in traps n and n_d and holes in centers m is registered as a function of the total time elapsed from the end of the excitation period. We proceed by evaluating the response to a pulse of stimulating light. For a chosen value for f, we solve numerically the set of equations (1)–(5) with the current values of n, n_d and m, again using the Matlab ode15s solver, for a short length of time and, of course, with $X = 0$. Obviously, during the time in which the pulse is given, the concentrations of trapped electrons and holes change, however, if we wish to study the fading of the OSL pulse intensity as a function of the period

of time elapsed from the excitation, we should not take into account the change of the concentrations during this exposure to stimulating light. We therefore keep in memory the concentrations following K periods of time (years) of fading, evaluate the response to the light pulse, register it and then go to the fading in the (K+1)th period of time using the Monte-Carlo method starting with the concentrations prior to the latest short light-stimulated read-out. For simulating TL, we use the same procedure, but solve the equations with $f = 0$ and varying temperature, and record the TL peak or evaluate the area under the curve.

5. Results of the simulations

Fig. 2 shows the results of the OSL simulation using the sequence of excitation simulated by the solution of the relevant equations by the mentioned Matlab solver, followed by the slow fading simulated by the Monte-Carlo procedure. Finally, the response to the OSL light pulse is followed by using the same Matlab solver. The set of chosen parameters is given in the caption. Note that here, the trapping probability coefficient into the competing trap is $A_{nd} = 10^{-13}\text{cm}^3\text{s}^{-1}$, very low as compared to the retrapping and recombination probability coefficients. As a result, the outcome is very similar to that from the OTOR model, namely with the same set of parameters but with $A_{nd} = 0$. Similarly to the results shown by Chen and Pagonis (2015), the curve of fading is seen to be concave on the semi-log scale, representing a slower than exponential decay. Note the difference between the results here and the previous work. There, the remaining concentration of trapped electrons following the very slow thermal fading was shown whereas here, we present the simulated OSL response to a light pulse following the thermal decay. In Fig. 2, curve (a) is the original simulated decay curve. Line (b) shows (on the semilog scale) the exponential decay resulting from the s and E entered into the simulations. Curve (c) is the straight line which coincides with curve (a) in the first ~200 years. This emphasizes the "anomalous stability" mentioned in the paper. If one assumes that the decay is exponential as determined in the first 200 years, one would conclude that it takes ~900 years to reach −1 on the y-axis whereas the "real" simulated line does not reach this level even after 1400 years.

In order to examine the situation with a stronger competitor, the simulation has been repeated with the same set of parameters, except

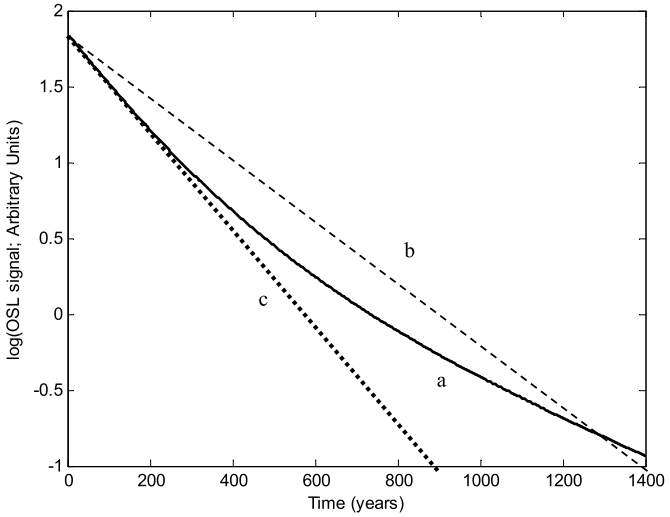

Fig. 2. The OSL signal (solid line, (a)) as a function of the time between excitation and readout within the given model. The parameters used were $N = 10^7\text{cm}^{-3}$; $N_d = 10^9\text{cm}^{-3}$; $M = 10^9\text{cm}^{-3}$; $X = 10^9\text{cm}^{-3}\text{s}^{-1}$; $A_m = 10^{-8}\text{cm}^3\text{s}^{-1}$; $A_n = 10^{-10}\text{cm}^3\text{s}^{-1}$; $A_{nd} = 10^{-13}\text{cm}^3\text{s}^{-1}$; $B = 10^{-7}\text{cm}^3\text{s}^{-1}$; $E = 1.3\text{eV}$; $s = 10^{12}\text{s}^{-1}$; $T = 300$ K. Since the trapping probability is relatively very small, the results are very similar to those of the OTOR model, with $A_{nd} = 0$. Curve (b) (dashed line) shows the exponential decay with the same E and s values. Curve (c) (dotted line) is the line extrapolated from the first 200 years.

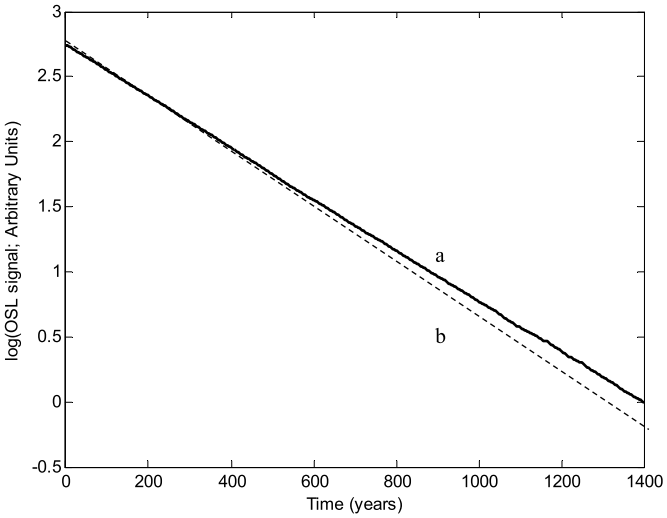

Fig. 3. The OSL signal as a function of the time between excitation and readout in the presence of a deep-trap competitor. The parameters are the same as in Fig. 2, except that the trapping recombination probability coefficient of the deep trap is significantly larger, namely $A_{nd} = 10^{-8}\text{cm}^3\text{s}^{-1}$. Line (a) shows the simulated results and line (b) the exponent due to the given E and s values.

that the trapping probability coefficient of the competitor has been significantly larger, namely, $A_{nd} = 10^{-8}\text{cm}^3\text{s}^{-1}$. Obviously, this set has been used in the three stages of excitation, long-time fading and the OSL response to a pulse of stimulating light. The results shown in Fig. 3 depict a straight line on the semi-log scale. This shows that within this model of two traps and one center, namely, a model in which, in addition to the levels involved in the OTOR system, a significant trapping competitor takes part in the process, the possibility of an exponential fading appears to be more feasible. It should be noted that under these circumstances, the slope in the semi-log scale in Fig. 3 and taking into account the factor needed to transfer from decimal to natural logarithms is nearly equal to $s{\cdot}exp(-E/kT)$, as could be expected. Note that under the present conditions, $A_n(N\text{-}n) \sim 10^{-3}$, $A_{nd}(N_d\text{-}n_d) \sim 10^{-4}$ and $A_m m \sim 0.6$, and therefore, $F \sim 1$. Thus, line (b) of the exponential decay based on the E and s values is nearly the same as the simulated results.

Fig. 4 depicts another exponential decay reached by the simulation when some of the parameters were changed, namely, $N_d = 10^8\text{cm}^{-3}$; $X = 10^6\text{cm}^{-3}\text{s}^{-1}$; $A_n = 10^{-8}\text{cm}^3\text{s}^{-1}$; $A_{nd} = 10^{-9}\text{cm}^3\text{s}^{-1}$. The results are seen to yield a straight line on the semilog scale, namely they represent an exponential decay. The slope, however, is ~1.9 times smaller than in Fig. 3, meaning that the magnitude F is 1/1.9~0.53. This is in good agreement with Eq. (8) which yields $F \sim 0.52$ by inserting the constant parameters as well as the nearly constant value of $m \sim 9 \times 10^5\text{cm}^{-3}$ and the values of n and n_d which were found to be significantly smaller than N and N_d, respectively. The significant deviation of F from unity is reflected by the large difference between curve (a) which shows the simulated decay of OSL and curve (b) which is determined by the E and s values.

Similar simulations were performed for TL. The excitation and fading parts are exactly the same as before. Instead of evaluating the response to a light pulse in OSL, we perform here TL runs following different fading times. Obviously, we repeat the excitation and fading for different periods of fading time before simulating the TL curves. Fig. 5 shows the results of the simulated TL curves with the parameters given in the caption and fading times of 1, 10, 100, 200, 300 and 400 years. Note that here, the trapping coefficient A_{nd} is relatively large, $10^{-8}\text{cm}^3\text{s}^{-1}$, and as mentioned above (see also Chen and Pagonis, 2013), one may expect a first-order-like TL peak. Indeed, the resulting curves in Fig. 5 have the asymmetry of first-order peaks and also, with the fading, the peak reduces but is not distorted. The situation is different in Fig. 6. Here, A_{nd} is 7 orders of magnitude smaller and therefore, there is

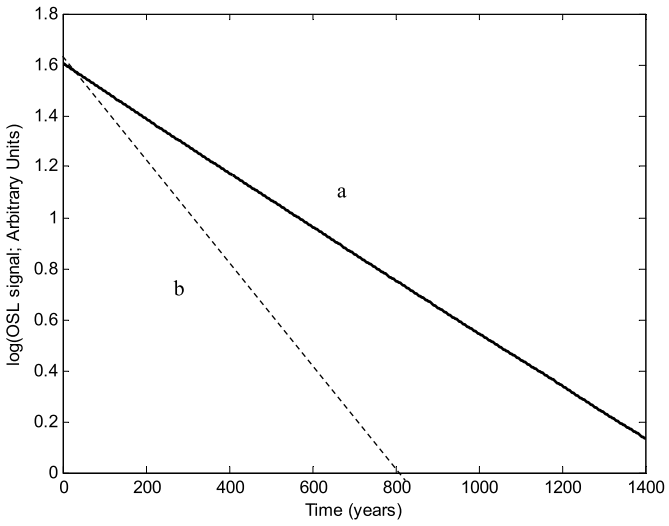

Fig. 4. The OSL signal as a function of time with the set of parameters: $N = 10^7 cm^{-3}$; $M = 10^9 cm^{-3}$; $N_d = 10^8 cm^{-3}$; $X = 10^6 cm^{-3} s^{-1}$; $A_m = 10^{-8} cm^3 s^{-1}$; $A_n = 10^{-8} cm^3 s^{-1}$; $A_{nd} = 10^{-9} cm^3 s^{-1}$; $B = 10^{-7} cm^3 s^{-1}$. Line (a) shows the simulated results and line (b) the simple exponential decay with the same E and s values.

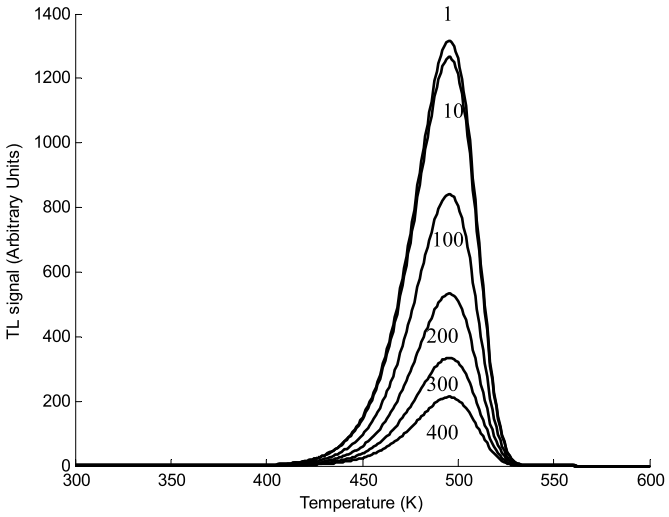

Fig. 5. TL curve following excitation and fading times of 1, 10, 100, 200, 300 and 400 years. The set of parameters used was: $N = 10^7 cm^{-3}$; $M = 10^9 cm^{-3}$; $N_d = 10^9 cm^{-3}$; $A_m = 10^{-8} cm^3 s^{-1}$; $A_n = 10^{-10} cm^3 s^{-1}$; $A_{nd} = 10^{-8} cm^3 s^{-1}$; $B = 10^{-7} cm^3 s^{-1}$; $E = 1.3$eV; $s = 10^{12} s^{-1}$; $t_D = 1$s; $X = 10^9 cm^{-3} s^{-1}$; $\beta = 1 Ks^{-1}$.

practically no competitive trapping. The TL peaks have an entirely different symmetry, definitely not first-order-like. With different fading times, the shape of the peak changes and the maximum peak temperature shifts to higher temperatures with fading time.

In Fig. 7, the simulated fading of OSL and TL with the same parameters as in Figs. 5 and 6, are shown. In OSL, the signal is the response to a short stimulating light pulse whereas in TL, it is the area under the glow curve from 400 to 700 K. Note that the competitor here is negligibly small, similar to the results in Fig. 2. The two decreasing curves have a generally similar appearance, but they are not identical. The OSL curve shows a faster decrease with time than that of TL. This may be attributed to the change in shape of the TL peak with the fading time which may have resulted in inclusion of a different portion of the total area within the integrated area. We have repeated the TL simulations to higher temperatures and then, the two curves looked indeed parallel. We have left, however, the curves in the present form since in real life, the

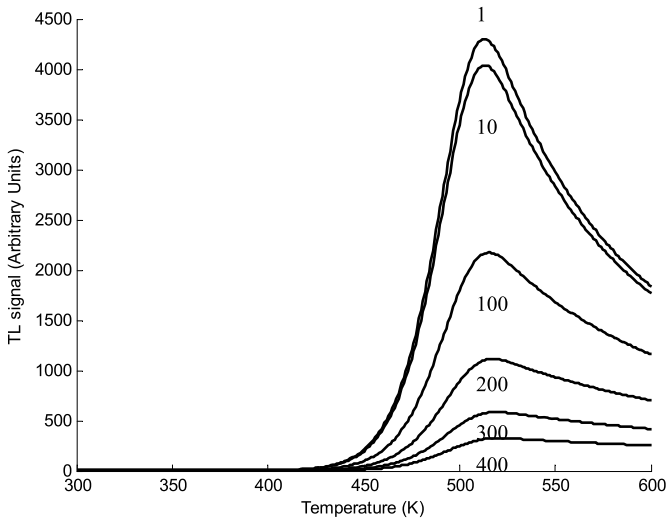

Fig. 6. Same as in Fig. 5 except that the probability coefficient of the competitor is orders of magnitude smaller, $A_{nd} = 10^{-15} cm^3 s^{-1}$.

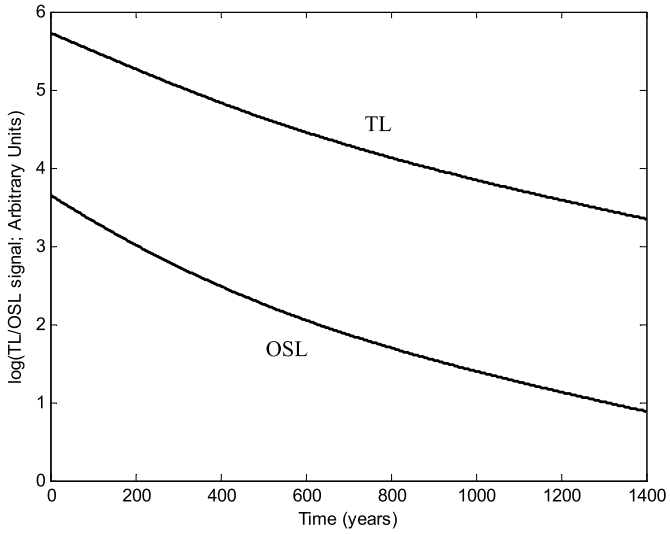

Fig. 7. The simulated fading of a short pulsed OSL and the area under the TL, integrated from 400 to 700 K. The parameters are the same as in Fig. 6.

maximum temperature is rather limited, and we wish to mimic the expected experimental results.

6. Discussion

In the present work, we have considered the expected fading of OSL and TL signals, following an appropriate excitation when the sample is held for a very long period of time at a relatively low temperature. This work is a follow-up to a previous work by Chen and Pagonis (2015) which showed that within the one-trap-one-center (OTOR) model, one should usually expect a slower than exponential fading with time of the concentration of trapped electrons. As pointed out in the previous work, this conclusion may be alarming in particular since it has been shown that within the OTOR framework, the beginning of the decay curve may look exponential but will deviate from the exponential (i.e. from linearity on the semi-log scale) in such a way that an effect of anomalous stability may be observed. It is worth mentioning that in the OTOR case it was sufficient to simulate by the Monte-Carlo method the dependence of the remaining concentration of carriers in traps and due to the simple energy scheme, it was clear that the OSL or TL signal will behave in the

same way as a function of the elapsed time. In the present case, due to the competition with a deeper trap through all the three stages of excitation, slow decay and OSL read-out, the simulation was performed in the three stages sequentially as explained above.

One should remember, however, that it is quite rare to find in real materials a pure OTOR system. In most cases, the situation is more complex and several traps and centers usually occur in a real crystal. In order to go one step forward, we have added to the model one deeper trap which may represent a number of deep traps. The result, shown by theoretical considerations as well as numerical simulation, is that if the competitor is large enough has a strong enough probability coefficient and is not too close to saturation, the possibility of getting an exponential decay is significantly stronger. Thus the chances of getting anomalous stability in real materials seem to be smaller. This, however, is not the only possibility for having $F \sim$ const. The main contribution of the competitor to the approximately exponential fading is that in the presence of such strong competitor, much more electrons are being trapped in it rather than performing recombination. Thus, the concentration m varies only slightly during the period of time between excitation and read-out. Inspecting Eq. (8), one can deduce that if N and N_d are far from saturation and m varies slowly, F is nearly constant.

We would like to mention that while considering the fading curve of the OSL/TL signal, the slope on the ln(signal) vs. time graph should yield a value of $s \cdot exp(-E/kT)$ in cases where the function F in Eq. (8) is nearly unity. This has been shown in the example in Fig. 3. On the other hand, if this term is significantly smaller than unity and the decay line is still linear on the semilog scale, one would interpret the results to be associated with first-order kinetics with the correct activation energy but with an effective frequency factor, smaller than s by the same factor $s' = Fs$ where s' is the apparent frequency factor. An example of this situation has been shown in Fig. 4.

It should be noted that, as pointed out in the Introduction, the idea that with the occurrence of a strong competitor, a first-order behavior may be expected, emerged from previous work (Lewandowski and McKeever, 1991; Sunta et al., 1999, 2001; 2005; Pagonis and Kitis, 2012; Chen and Pagonis, 2013), in which the occurrence of a deep competitor was shown to result in an abundance of first-order TL peaks in the presence of a competitor. The analogy between the two cases is not obvious a priori; in the TL case the first-order feature is expressed in the symmetry of the peak whereas in the present case it has also to do with the exponential fading of the OSL or TL signal. Rather similar arguments have been made by Bailey et al. (1997) to explain the possible occurrence of an exponential decay of OSL in quartz *during* light stimulation. Anyway, the closeness of the fading function to being exponential depends on all the relevant magnitudes in Eq. (8), but the chance of the expression F to be constant, leading to the exponential decay is much larger in the presence of the competitor.

As pointed out above, the main condition for getting the non-exponential decay is that there is no significant trapping competitor. This may take place in a carefully prepared clean material with only two kinds of imperfections causing a single electron trap and one hole center. The chances of getting such a material in nature is rather small, so a cautious conclusion is that usually, an exponential decay is prevalent.

A final remark has to do with the comparison between the fading of the OSL and the TL signals for the same set of parameters. As can be seen in Figs. 5 and 6, it may not be feasible to integrate the TL area to "infinity", namely to a high enough temperature such that the "signal" will comprise of all the emitted TL light. It is quite common to integrate over a limited range of temperature, as has been done with the results in Fig. 7. When the parameters were such that the OSL and TL results were first-order like (Figs. 3–5) the OSL and TL (not shown here) fading curves looked the same (up to a constant), and with the same slope on the semi-log scale, even when only part of the TL range was used. As is quite obvious from Figs. 6 and 7, this was not the case when the competitor transition probability was negligibly small, and the shapes of the curves were twisted with different fading times. When the area under a limited range of the TL curve was taken as the signal, its decay with fading time was not exactly the same as in pulsed OSL under the same circumstances, as shown in Fig. 7.

References

Bailey, R.M., 2004. Paper I: simulation of dose absorption in quartz over geological timescales and its implications for the precision and accuracy of optical dating. Radiat. Meas. 38, 299–310.

Bailey, R.M., Smith, B.W., Rhodes, E.J., 1997. Partial bleaching and the decay form characteristics of quartz OSL. Radiat. Meas. 27, 123–136.

Chen, R., Pagonis, V., 2013. On the expected order of kinetics in a series of thermoluminescence (TL) and thermally stimulated conductivity peaks. Nucl. Instrum. Methods Phys. Res. B 312, 60–69.

Chen, R., Pagonis, V., 2015. Study of the stability of the TL and OSL signals. Radiat. Meas. 81, 192–197.

Kulkarni, R.N., 1994. The development of the Monte Carlo method for the calculation of the thermoluminescence intensity and the thermally stimulated conductivity. Radiat. Protect. Dosim. 51, 95–105.

Lewandowski, A.C., McKeever, S.W.S., 1991. Generalized description of thermally stimulated processes without quasiequilibrium approximation. Phys. Rev. B43, 8163–8178.

Mandowski, A., 2001a. Modelling of charge carriers' transport and trapping phenomena in one-dimensional structures during thermal stimulation. J. Electrost. 51–51, 585–589.

Mandowski, A., 2001b. One-dimensional thermoluminescence kinetics. Radiat. Meas. 33, 745–749.

Mandowski, A., 2006. Calculation and properties of trap structural functions for various spatially correlated systems. Radiat. Protect. Dosim. 119, 85–88.

Mandowski, A., 2008. How to detect trap cluster systems? Radiat. Meas. 43, 167–170.

Mandowski, A., Świątek, J., 1992. Monte Carlo simulation of thermally stimulated relaxation kinetics of carrier trapping in microcrystalline and two-dimensional solids. Phil. Mag. B 65, 729–732.

Mandowski, A., Świątek, J., 1996. On the influence of spatial correlation on the kinetic order of TL. Radiat. Protect. Dosim. 65, 25–28.

Mandowski, A., Świątek, J., 2000. The kinetics of trapping and recombination in low dimensional structures. Synth. Met. 109, 203–206.

Mandowski, A., Bos, A.J.J., Mandowska, E., Orzechowski, J., 2010. Monte-Carlo method for determining the quenching function from variable heating rate measurements. Radiat. Meas. 45, 284–287.

Pagonis, V., Kitis, G., 2012. Prevalence of first-order kinetics in thermoluminescence materials: an explanation based on multiple competition processes. Phys. Status Solidi 249, 1590–1601.

Rodríguez-Villafuerte, M., 1999. A Monte Carlo approach to the track interaction model to explain supralinearity in the thermoluminescence response. Nucl. Instrum. Methods Phys. Res. B 152, 105–114.

Sunta, C.M., Ayta, W.E.F., Piters, T.M., Watanabe, S., 1999. Limitation of peak fitting and peak-shape methods for determining of activation energy of thermoluminescence glow peaks. Radiat. Meas. 30, 197–201.

Sunta, C.M., Ayta, W.E.F., Chubaci, J.F.D., Watanabe, S., 2001. A critical look at the kinetic models of thermoluminescence: I. First-order kinetics. J. Phys. D Appl. Phys. 34, 2690–2698.

Sunta, C.M., Ayta, W.E.F., Chubaci, J.F.D., Watanabe, S., 2005. A critical look at the kinetic models of thermoluminescence: II. Non-first-order kinetics. J. Phys. D Appl. Phys. 38, 95–102.

Thompson, J.W., 2007. Accuracy, precision, and irradiation time for Monte Carlo simulations of single aliquot regeneration (SAR) optically stimulated luminescence (OSL) dosimetry measurements. Radiat. Meas. 42, 1637–1646.