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## Study of the stability of the TL and OSL signals

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## HIGHLIGHTS

- Stability and fading of TL and OSL during very long periods of time.
- Monte-Carlo simulations.
- Exponential and non-exponential decay behavior.
- Possibility of unexpected stability.

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## ABSTRACT

In the study of thermoluminescence (TL) and optically stimulated luminescence (OSL), and in particular in the applications of archaeological and geological dating as well as dosimetry, the issue of stability of the signal at ambient temperature following excitation is of paramount importance. In many cases, one determines the activation energy ( $E$ ) and frequency factor ( $s$ ) of a TL peak, and tries to evaluate the lifetime of the excited signal. This is meaningful if the process is of pure first order, and may not be so in non-first-order situations. In the present work, we study this matter for both first-order and the more general one-trap-one-recombination-center (OTOR) cases using numerical simulations. The conventional numerical solution of the relevant set of coupled differential equations may not work when the traps are deep and the length of time is, say, thousands of years or more, and we therefore resort to a Monte-Carlo approach. It is obvious that in instances of dominating recombination, the long-time decay is exponential, and the decay constant is as expected from the first-order behavior and the  $E$  and  $s$  values. However, in cases of substantial retrapping, the fading is slower, sometimes very significantly, and is not exponential. Thus, one may deduce from the evaluated  $E$  and  $s$  shorter decay times than occur in fact. This may lead to an apparent effect of unexpected stability, namely, that a signal is stable much longer than expected from the evaluated trapping parameters. Possible implications concerning applications in archaeological and geological dating are obvious.

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## 1. Introduction

The main applications of thermoluminescence (TL) and optically stimulated luminescence (OSL) are in dating of archaeological and geological specimens and in dosimetry. A crucial property to be considered in this respect is the stability of the signal following excitation, when the sample is held at ambient temperature. Obviously, the parameters related to the relevant trapping states of electrons and holes determine this property of stability. In the simplest possible case of first-order kinetics, the situation

concerning stability is very easy to follow. As first given by [Randall and Wilkins \(1945\)](#), the equation governing this process is

$$I(T) = -\frac{dn}{dt} = s \cdot n \cdot \exp(-E/kT), \quad (1)$$

where  $n$  ( $\text{cm}^{-3}$ ) is the concentration of electrons in traps,  $E$  (eV) the activation energy for their release into the conduction band,  $s$  ( $\text{s}^{-1}$ ) the frequency factor,  $T$  (K) the temperature,  $t$  (s) the time,  $k$  ( $\text{eV K}^{-1}$ ) the Boltzmann constant and  $I$  ( $\text{cm}^{-3} \text{s}^{-1}$ ) the TL intensity. Eq. (1) governs the rate of depletion of electrons from the trap at a constant temperature  $T$ . Thus it is associated with the stability of the signal which is directly related to the remaining concentration of electrons after a certain period of time following excitation and

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prior to the heating of the sample in TL or its optical read-out in OSL. When the temperature at which the sample is held is significantly lower than that of the expected TL peak, the isothermal emitted light  $I(T)$  may be very weak and usually not measurable, but after a long period of time, the remaining value of  $n$  may be significantly smaller than right after the irradiation, which is expressed as thermal fading of the TL or OSL signal. According to Eq. (1), the time dependence of the concentration of electrons in traps at a given temperature  $T$  is

$$n(t) = n_0 \exp [(-s \exp(-E/kT))t]. \quad (2)$$

This is an exponentially decreasing function, and its “lifetime”, namely, the time it takes for the concentration of trapped electrons to reduce to  $1/e$  of its initial value is

$$\tau = s^{-1} \exp(E/kT). \quad (3)$$

Obviously, under these circumstances, if the sample is held at a constant temperature (usually “room temperature”, RT), the expected TL signal following a time  $t$  will be reduced at the same rate as  $n(t)$  and therefore, with the same lifetime.

One should note that the main assumption leading to the first-order situation is that recombination is significantly stronger than retrapping, which is a rather limiting assertion. The situation gets much more complex in the more realistic framework, in which retrapping of free electrons is considered, as first studied by Halperin and Braner (1960). Assuming a system with one trapping state and one recombination center, they presented the following set of simultaneous differential equations,

$$\frac{dn}{dt} = A_n(N - n)n_c - s \cdot n \cdot \exp(-E/kT), \quad (4)$$

$$I(T) = -\frac{dm}{dt} = A_m m n_c, \quad (5)$$

$$\frac{dn_c}{dt} = s \cdot n \cdot \exp(-E/kT) - A_n(N - n)n_c - A_m m n_c, \quad (6)$$

where  $N$  ( $\text{cm}^{-3}$ ) is the total concentration of traps,  $m$  ( $\text{cm}^{-3}$ ) is the instantaneous concentration of holes in centers,  $n_c$  ( $\text{cm}^{-3}$ ) is the concentration of free electrons in the conduction band and  $A_n$ ,  $A_m$  ( $\text{cm}^3 \text{s}^{-1}$ ) are, respectively, the retrapping and recombination probability coefficients. Halperin and Braner have made a simplification of these equations by making the “quasi-equilibrium” assumptions, namely, that  $n_c \ll n$ ,  $m$  and  $dn_c/dt \approx 0$ . This resulted in the approximate single equation

$$I_{app}(T) = -\frac{dm}{dt} = s \cdot n \cdot \exp(-E/kT) \frac{A_m m}{A_m m + A_n(N - n)}. \quad (7)$$

In the restricted case of only one trapping state and one kind of recombination center, one has  $m = n$  following the excitation, and  $m = n + n_c \approx n$  along the TL process. This situation is termed the one trap-one recombination center (OTOR) model. Note also that the occurrence of dominating recombination,  $A_m m \gg A_n(N - n)$  leads to the Randall-Wilkins first-order Eq. (1). On the other hand, the inverse condition  $A_m m \ll A_n(N - n)$  may lead to a second-order situation if, again,  $n = m$  and  $N \gg n$ . The resulting second-order equation is

$$I(T) = -\frac{dn}{dt} = \frac{sA_m}{NA_n} \cdot n^2 \cdot \exp(-E/kT). \quad (8)$$

A similar equation governs the process when the retrapping and recombination probability coefficients are equal,  $A_n = A_m$  (Garlick

and Gibson, 1948).

In the present paper we consider the decay functions of trapped carriers at relatively low temperatures so that the TL or OSL signals may survive for very long periods of time. Under these circumstances, trying to solve Eqs. (4)–(6) using the MATLAB ode solvers resulted in exceedingly long computation times, and a Monte-Carlo method is shown to be a viable option.

## 2. The Monte-Carlo method

The use of the Monte-Carlo algorithms for the study of TL has been first 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 carriers in a sample is large and the kinetic equations (e.g. Eqs. (4)–(6)) 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 individually, 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 use for the track-interaction model which may explain the supralinearity in the TL response. Bailey (2004) has used the Monte-Carlo approach to follow 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 the present work, we demonstrate the use of the Monte-Carlo method in simulating the very slow thermal fading of TL signals when the sample is held at a temperature significantly lower than that of the TL peak. Here, we consider the electrons one at a time. The interesting cases here are those where retrapping is not negligibly small. The results reported below have important bearing on the stability of the TL signal.

Let us consider first the first-order case, described in Eq. (1). Assume that electrons can be released thermally into the conduction band. As long as we assume dominating recombination, and once the electrons are in the conduction band, they recombine practically immediately with a trapped hole in center (Randall and Wilkins, 1945). If the sample is held at a temperature  $T$  and if the activation energy is  $E$  and the frequency factor  $s$ , 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  $\Delta t$ . 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. The considerations for choosing a reasonable compromise value of  $\Delta t$  in specific cases will be discussed below. Once we choose a certain initial number of trapped electrons (typically  $10^6$ ), 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 undergone recombination, and therefore, it is reduced from the number of electrons in the trap and one hole is reduced from the center. Once all the electrons have been tested, we register the final number of remaining electrons in traps, go to the next time interval  $\Delta t$  and repeat the procedure with the remaining electrons as the new initial value.

The next step is to consider the more complex situation where retrapping is allowed. If we look at the last two terms in Eq. (6), it is quite obvious that once an electron has been raised into the conduction band, its probabilities of performing recombination and retrapping are proportional to  $A_m m$  and  $A_n(N-n)$ , respectively. Thus, the probability for recombination is  $A_m m / [A_m m + A_n(N-n)]$  and the probability for retrapping is  $A_n(N-n) / [A_m m + A_n(N-n)]$ . In the 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  $A_m m / [A_m m + A_n(N-n)]$  is larger 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 be back in its box and the procedure continues to the next electron. Note that in this relatively simple OTOR situation,  $m = n$  all along.

### 3. Results of the simulations

Fig. 1 depicts three simulated TL peaks, whose stability will be studied. The set of three simultaneous differential Eq. (4)–(6) has been solved numerically using the ode15 s Matlab solver. The parameters chosen were  $N = 10^{13} \text{ cm}^{-3}$ ;  $n_0 = m_0 = 10^{12} \text{ cm}^{-3}$ ;  $E = 1.3 \text{ eV}$ ;  $s = 10^{12} \text{ s}^{-1}$ ;  $\beta = 1 \text{ K s}^{-1}$  and  $A_m = 10^{-8} \text{ cm}^3 \text{ s}^{-1}$ . The values of the retrapping probability coefficient  $A_n$  were (a)  $10^{-10} \text{ cm}^3 \text{ s}^{-1}$  (solid line); (b)  $10^{-8} \text{ cm}^3 \text{ s}^{-1}$  (dashed line) and (c)  $10^{-6} \text{ cm}^3 \text{ s}^{-1}$  (dotted line).

In curve (a), the maximum intensity occurs at  $T_m = 496.4 \text{ K}$ ; the effective activation energy was found by the full width peak-shape method as  $E_{\text{eff}} = 1.339 \text{ eV}$  and the effective frequency factor  $s_{\text{eff}} = 2.46 \times 10^{12} \text{ s}^{-1}$ ; the symmetry factor is  $\mu_g = 0.435$ , typical of a first-order peak. (For a detailed discussion of the peak-shape

methods and symmetry factor see, e.g., Chen, 1969). This is reasonable since the recombination probability coefficient here is two orders of magnitude larger than the retrapping probability coefficient, so dominating recombination may be expected. This point will be further discussed below. The lifetime here, as calculated by Eq. (3) is  $\tau_f = 1.25 \times 10^{10} \text{ s} \sim 398 \text{ years}$ .

In curve (b),  $E_{\text{eff}} = 1.296 \text{ eV}$ ;  $s_{\text{eff}} = 9.7 \times 10^{10} \text{ s}^{-1}$ ;  $T_m = 532.7 \text{ K}$  and  $\mu_g = 0.519$ , typical of second-order peaks. Second-order is indeed expected when the retrapping and recombination coefficients are equal. The lifetime here as calculated using the effective activation energy and frequency factor is  $\tau_f = 6.13 \times 10^{10} \text{ s} \sim 1943 \text{ years}$ .

In curve (c),  $E_{\text{eff}} = 1.29 \text{ eV}$ ;  $s_{\text{eff}} = 1.04 \times 10^9 \text{ s}^{-1}$ ;  $T_m = 556.8 \text{ K}$  and  $\mu_g = 0.526$ , also typical of second-order peaks. The apparent lifetime here is  $\tau_f = 5.2 \times 10^{12} \text{ s} \sim 165,000 \text{ year}$ . In the case of dominating retrapping, second-order kinetics is expected. The meaning of  $s_{\text{eff}}$  in the second-order case is discussed below. Note that the areas under peaks (a), (b) and (c) are the same since in all three, the area is equal to  $m_0$ .

Fig. 2 shows the results of simulations of the reduction in the number of trapped electrons when the sample is held at  $300 \text{ K}$  ( $27^\circ \text{C}$ ), in the pure first-order case. The parameters chosen are the same as above, namely,  $E = 1.3 \text{ eV}$ ;  $s = 10^{12} \text{ s}^{-1}$ ;  $n_0 = 10^6$  and  $\Delta t = 3.15 \times 10^7 \text{ s} = 1 \text{ year}$ . One may wonder whether 1 year may be considered as a rough period of time. Note that with the present values of  $E$  and  $s$ , the relevant probability is, as mentioned,  $s \cdot \exp(-E/kT) \cdot \Delta t = 0.00455$ , significantly smaller than unity. This means that in each period  $\Delta t$ , less than half a percent of the remaining electrons are released to the conduction band and subsequently recombine. The dotted line depicts the results of the simulation whereas the solid line represents the results of direct substitution into Eq. (2). The agreement is seen to be very good. So far, this is merely a demonstration of the performance of the Monte-Carlo procedure.

Figs. 3–5 show the results of the Monte-Carlo simulation of the emptying of the traps leading to the three peaks in Fig. 1. The  $E$  and  $s$  parameters are the same as in Fig. 2, and so are the  $n_0$ ,  $\Delta t$  and  $T = 300 \text{ K}$  used. The simulated results go to 14,000 years, and are given on a semi-log scale. In these three figures, we used the

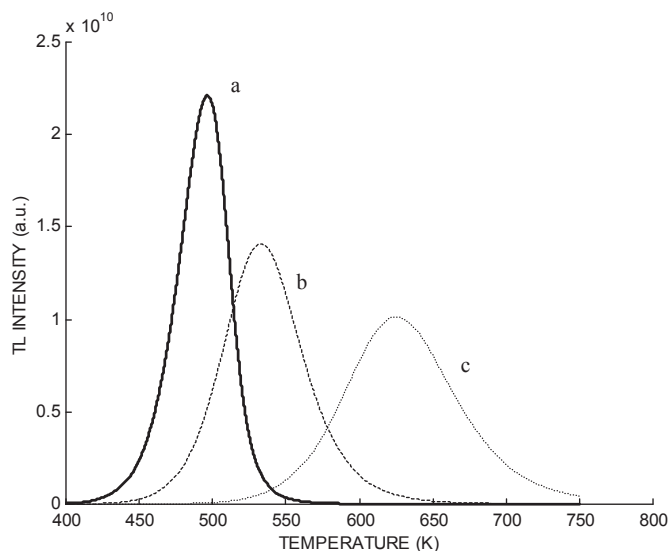


Fig. 1. Three TL peaks simulated by solving numerically the set of Eqs. (4)–(6). The parameters used were:  $N = 10^{13} \text{ cm}^{-3}$ ;  $n_0 = m_0 = 10^{12} \text{ cm}^{-3}$ ;  $E = 1.3 \text{ eV}$ ;  $s = 10^{12} \text{ s}^{-1}$ ;  $\beta = 1 \text{ K s}^{-1}$ ;  $A_m = 10^{-8} \text{ cm}^3 \text{ s}^{-1}$ . (a) (solid line):  $A_n = 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ . (b) (dashed line):  $A_n = 10^{-8} \text{ cm}^3 \text{ s}^{-1}$ . (c) (dotted line):  $A_n = 10^{-6} \text{ cm}^3 \text{ s}^{-1}$ .

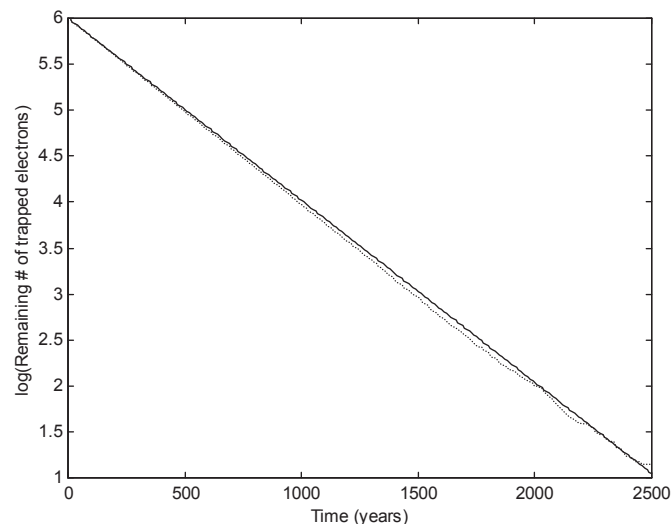
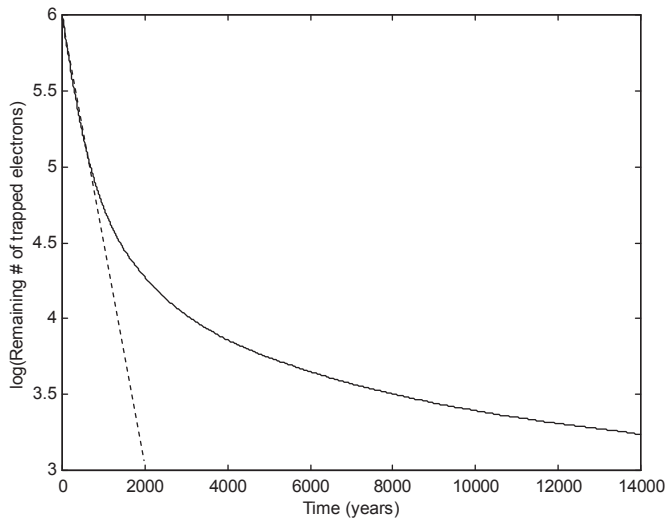


Fig. 2. Time dependence of the remaining concentration of trapped electrons following excitation for the pure first-order case, shown on a semi-log scale. The parameters used are  $n_0 = 10^6$ ;  $E = 1.3 \text{ eV}$ ;  $s = 10^{12} \text{ s}^{-1}$ ;  $T = 300 \text{ K}$ . The dotted line depicts the results of the Monte-Carlo simulation and the solid line shows the results from Eq. (2).

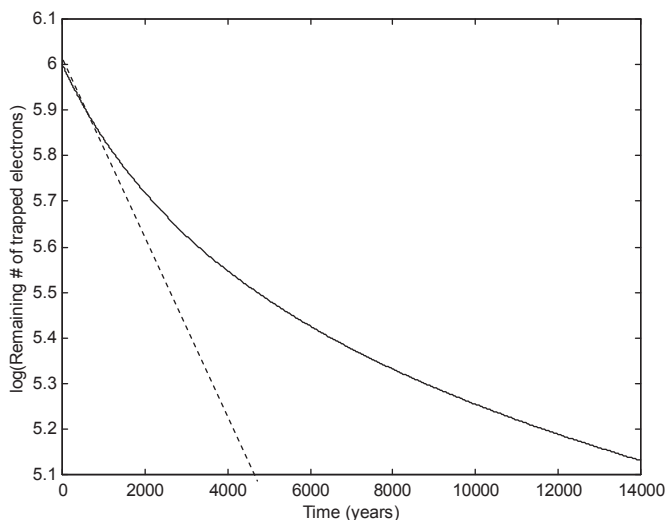


**Fig. 3.** Time dependence of the remaining concentration of electrons in traps, simulated by the Monte-Carlo method, given on a semi-log scale, in the case of small retrapping probability coefficient. The parameters are the same as in Fig. 2, and  $N = 10^7$ ,  $A_m = 10^{-8} \text{ cm}^3 \text{ s}^{-1}$ ;  $A_n = 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ . The straight, dashed line represents the exponential decay at the rate determined by the initial  $E$  and  $s$  values.

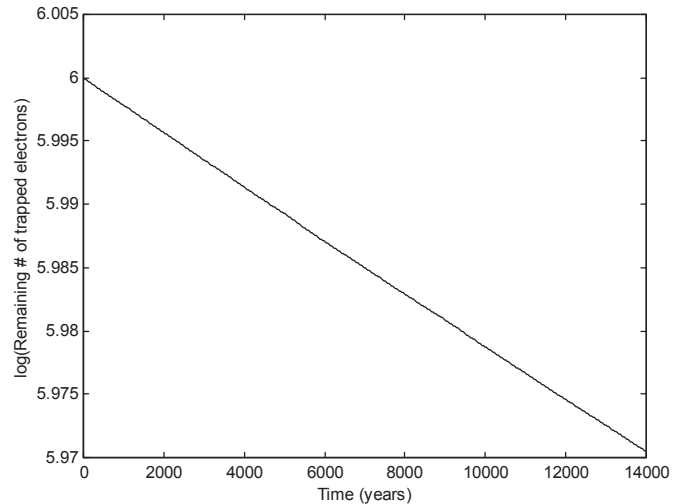
recombination probability coefficient  $A_m = 10^{-8} \text{ cm}^3 \text{ s}^{-1}$ . In Fig. 3, the retrapping probability coefficient is smaller,  $A_n = 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ . The results up to ~500 years form a nearly straight line, meaning that the decay is nearly exponential, but due to the increasing relative magnitude of retrapping, from 500 years on, the line curves very significantly. The decay is by nearly 3 orders of magnitude in 14,000 years. These results will be discussed below.

Fig. 4 shows the decay of the remaining number of trapped electrons for the case of  $A_n = A_m = 10^{-8} \text{ cm}^3 \text{ s}^{-1}$ , and all the other parameters are as before. In this pure second-order case, one does not expect an exponential decay, and indeed, the line on the semi-log scale is curved as of the beginning. Obviously, due to the stronger retrapping, the decay is much slower than in the previous case, namely, less than one order of magnitude in 14,000 years.

Fig. 5 shows the simulated decay curve for  $A_n = 10^{-6} \text{ cm}^3 \text{ s}^{-1}$  where all the other parameters are kept the same. The TL in



**Fig. 4.** Same as Fig. 3, but with equal recombination and retrapping probability coefficients,  $A_n = 10^{-8} \text{ cm}^3 \text{ s}^{-1}$ .



**Fig. 5.** Same as Fig. 3, but with stronger retrapping probability coefficient,  $A_n = 10^{-6} \text{ cm}^3 \text{ s}^{-1}$ .

question has been found to be of second order. As expected, the fading is very significantly slower than in the previous cases due to the stronger retrapping. Within 14,000 years, the number of remaining electrons in traps decreases by only ~6.9%. It is rather surprising that in this second-order case, the decay in the given time range appears like a straight line on the semi-log scale, namely, the time dependence is exponential. However, in Fig. 6 the continuation of the decay curve up to 1.4 Myr is shown, and it is seen that at longer times, the decay deviates very significantly from the exponential. This point will be discussed below.

#### 4. Discussion

In the present work, we have studied the thermal decay of the number of trapped carriers at a constant temperature (e.g. RT) in cases where the trapping parameters are such that the fading is very slow. Under these circumstances, the solution of the relevant set of simultaneous equations may be problematic since the lifetime of electrons in the conduction band is usually of the order of a fraction of a second (see e.g. Rose, 1955). Therefore, at the slow rate at which the electrons are released into the conduction band, at the relatively low temperature at which the sample is held, the electrons are raised one at a time, and there is no real meaning to the function  $n_c$  appearing in Eqs. (4)–(6). As mentioned above, the probability for releasing an electron within the interval  $\Delta t$  (chosen as 1 year) is  $s \cdot \exp(-E/kT) \cdot \Delta t$  and with the parameters chosen this is 0.00455. For a trap with the initial occupancy of  $10^6$  electrons, this means that only 4550 electrons be raised thermally per year, or about 1 electron every 2 h and fewer when the number of trapped electrons diminishes at longer times.

Concerning the initial number of  $10^6$  electrons, it has taken a very long computer time to exceed this number, in particular in the cases with stronger retrapping when on the average, an electron may be released and retrapped several times before performing recombination, thus subtracted from the remaining number. These million carriers can be representative of a larger number existing in a real sample. In fact, if like in Fig. 1 we take a realistic concentration of traps of  $10^{13} \text{ cm}^{-3}$ , out of which, say, 10% are full of electrons to begin with, we start with a concentration of  $10^{12} \text{ cm}^{-3}$ . If we consider as our sample a grain of quartz which is ~100  $\mu\text{m}$  in size, its volume is ~ $10^{-6} \text{ cm}^3$  and the total number of initially trapped electrons is ~ $10^6$ . Thus, the given simulation applies directly to the

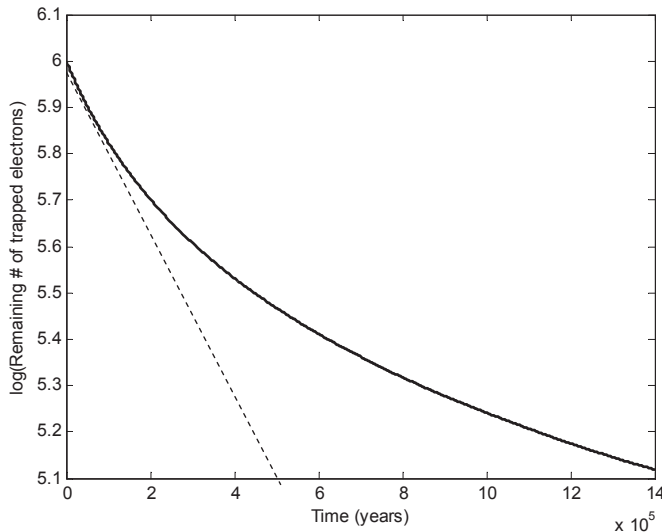


Fig. 6. Same as Fig. 5, but for a decay time 100 times longer, up to 1.4 Myr.

case of a fine grain. As for a bulk sample where the number of trapped electrons is orders of magnitude larger, dealing with  $10^6$  electrons, which is feasible with respect to computation time, is expected to be representative to the behavior of the whole electron population.

As pointed out above, the first peak with  $A_n = 0.01A_m = 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ , looks like a first-order peak, at least as far as the symmetry factor is concerned. From this, the apparent lifetime is  $\sim 398$  years. It is worth mentioning that this is longer than the first-order lifetime evaluated from the inserted  $E$  and  $s$  values which is  $\sim 219$  years and the difference has to do with the effect of retrapping. It should be noted that when the initial-rise method is used, the original activation energy of 1.3 eV has been found. However, in order to evaluate  $\tau$ , we need  $s_{\text{eff}}$ , for which we also need  $T_m$  which occurs out of the initial-rise range and where the non-first-order behavior is in effect. From the practical point of view, the initial-rise method may be problematic when the peak is not very intense and not isolated. Moreover, curve fitting or deconvolution are often used, and these utilize the same features of the peak as the peak-shape method used here. The bigger difference, however, has to do with the departure of the logarithmic curve from linearity in longer periods of time. Note that the probabilities for recombination and retrapping are proportional to  $A_m m$  and  $A_n(N-n)$ , respectively. Here, we start with the first term being  $\sim 10$  times larger, namely nearly dominating recombination leading to approximately exponential decay. However, at later times,  $m$  gets smaller and so does  $n$ , which in turn, makes  $N-n$  larger. Retrapping becomes relatively stronger which makes the fading significantly slower. We may thus have during the fading a gradual transition from a nearly first-order behavior to a nearly second-order one. Therefore, drawing a conclusion on the fading of the TL or OSL signal (which depends on the remaining number of electrons in traps) from the initial decay or from the TL-evaluated parameters may be erroneous, and an effect of unexpected stability may take place. Note that a similar effect of transition from first- to second-order behavior occurs within the TL peak shown in Fig. 1, curve a. Here, toward the end of the peak,  $A_n(N-n)$  becomes larger than  $mA_m$ , and the peak attains some second-order characteristics at this range, which is expressed by the occurrence of a longer tail.

Regarding the results in Fig. 4, since  $A_m = A_n$ , retrapping is stronger as of the very beginning, and the decay is not exponential even at the very beginning. As mentioned above, the apparent

lifetime is 1943 years. However, the evaluation of  $\tau$  by Eq. (3) is limited, in principle, to cases of first order, and obviously, the decay here is not exponential, which explains the fact that after 14,000 years, the number of remaining electrons in traps is not even one order of magnitude smaller than the initial value.

As for the results in Fig. 5, since  $A_n = 100A_m$ , obviously the fading is significantly slower. The fact that in the first 14,000 years the graph on the semi-log scale is a straight line, indicating an approximately exponential decline, is quite surprising. However, within this range, both  $A_m m$  and  $A_n(N-n)$  vary very little so that in this period of time, the probabilities for recombination and retrapping remain practically constant, which results in a very slow exponential decay. This can be seen while re-writing Eq. (7) for the isothermal situation,

$$\frac{dn}{dt} = s \cdot n \cdot \exp(-E/kT) \frac{A_m m}{A_m m + A_n(N-n)}. \quad (9)$$

In the present case, the factor  $A_m m / [A_m m + A_n(N-n)]$  remains nearly constant and therefore, the decay here is approximately exponential. In Fig. 6, the same decay curve is continued up to 1.4 Myr, and since here the same factor varies as  $m$  and  $n$  decline, the decay curve is not exponential any more. Note that the decay curve in Fig. 6 looks very similar to that in Fig. 4, where in the former, the retrapping coefficient is 100 times larger and the time span is 100 times longer.

As for the meaning of the evaluated frequency factor and the lifetime deduced thereof in the second-order peaks, as shown by Pagonis et al. (2006), the expression for  $s$  is

$$s = \frac{\beta E}{kT_m^2} \exp(E/kT_m) \frac{1}{1 + \Delta_m}, \quad (10)$$

where  $\Delta_m = 2kT_m/E$ , i.e. rather similar to the first-order expression. However, in the second-order case, this  $s$  is actually  $s' \cdot n_0(s^{-1})$  (see e.g. Chen, 1969) where  $s'$  is a constant, and thus,  $s$  is not constant. Moreover, in all non-first-order cases,  $\tau$  as in Eq. (3) can be considered at best as a rough estimate since the decay is not exponential.

Finally, although the regular solvers of sets of simultaneous ordinary differential equations did not work well for the present problem, the use of “stiffness switching” in Mathematica, enabled the numerical solution of the relevant set of equations and produced similar results to those of the Monte-Carlo simulation. The problem here was that with the used trapping parameters and the very slow fading, the calculated concentration of electrons in the conduction band turned out to be a small fraction of an electron. Therefore, the use of the Monte-Carlo method which deals with the electrons one by one seems to be more appropriate. In fact, both methods can be considered as approximations of the real situation, but the fact that both yield practically the same results is very encouraging.

In conclusion, using the Monte-Carlo method, we could follow the decay of the number of trapped carriers for very long periods of time in cases where the conventional numerical solutions of sets of differential equations are problematic. The work has been limited to the relatively simple case of one-trap-one-recombination-center (OTOR) with different degrees of retrapping. Under certain circumstances, it appears that the fading, which may represent the measured intensities of TL or OSL in the read-out stage, is significantly slower than warranted by the evaluated parameters of TL peaks. In a sense, this is a piece of good news since, when one may expect certain fading from the effective parameters, actually, the fading is significantly slower. Such a behavior may be considered as unexpected stability of the signal (for a different possible

explanation of anomalous stability, see [Chen et al., 2012](#)). In future research, one may wish to consider the fading characteristics of TL and OSL in cases beyond the given OTO model, e.g., when different degree of closeness to saturation of the initial occupation of traps is assumed or when additional disconnected traps are involved.

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