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Nonlinear dose dependence of TL and LM-OSL within the one trap-one center model

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ABSTRACT

In a recent paper, it has been shown that strong sub-linearity of the occupancy n_0 of trapping states far from saturation can be explained by the simplest model of one trap-one recombination center (OTOR). In the present work we report on results of numerical simulation of dose dependence of the TL maximum under similar conditions. In some cases, the TL maximum is found to be strictly proportional to the filling of the traps, but this is not always the case. Different sublinear dose-dependence functions of the trap occupancy and the maximum TL are demonstrated. With the same sets of parameters, curves of LM-OSL have also been simulated; superlinear as well as sublinear dependencies on the excitation dose have been found.

1. Introduction

In the applications of dosimetry and archaeological and geological dating, a linear dose dependence of thermoluminescence (TL) and optically stimulated luminescence (OSL) is very desirable. In many cases, however, nonlinear dose dependencies of these phenomena are very common. The most prevalent behavior of this kind is the approach to saturation, be it exponential or not, which takes place in all luminescent materials.

Superlinear dose dependence of TL has been reported in several materials. Goldstein (1967) and Israeli et al. (1972) studied the TL resulting from the production of point defects in UV-irradiated alkali-halides. This is a different situation than that of filling existing trapping states, however, as suggested by Lawless et al. (2009), a close analogy exists between the relevant sets of coupled simultaneous equations governing the two processes. Another material in which sub-linearity occurred is $CaSO_4$:Dy. Caldas and Mayhugh (1976) reported on PTTL, photo-transferred TL, which goes like $D^{0.55}$ where the excitation dose varies by 5 orders of magnitude.

In the mentioned paper, Lawless et al. (2009) studied the simple model of one trap, one recombination center (OTOR) both analytically and by numerical simulation. They show that the occupancy of trapping states far from saturation is sublinear, and that subject to certain choices of the sets of trapping parameters, it may behave like $D^{1/2}$. They also suggest that the resulting TL or OSL may depend in a similar way on the dose.

Another luminescence measurement utilized for dosimetry and dating is OSL. Bulur (1996) suggested the use of stimulating light which increases linearly with time, thus getting a peak-shaped linearly-modulated OSL (LM-OSL) curve. Chen and Pagonis (2008) showed that whereas for first-order kinetics the maximum intensity of the LM-OSL is proportional to the initial filling n_0, for second-order peaks, the maximum intensity is expected to behave like $n_0^{3/2}$. If n_0 is linearly dependent on the dose, this obviously means a superlinear dose dependence of LM-OSL. Chen et al. (2009) have demonstrated that the OTOR model may lead to first-order, second-order and intermediate kinetics and the dependence of the signal on n_0 may be the mentioned linear, 3/2 power or intermediate behaviors. On the other hand, since the filling of the trapping states may be sublinear with the dose, the LM-OSL maximum may be super- or sublinear, depending on the set of parameters used.

In the present work, we extend the simulations to include both the excitation and read-out stages in TL. We can thus get the simulated maximum TL intensity as a function of the excitation dose. For the LM-OSL signal, we use the same kind of simulations during excitation and read-out to see the possible dose dependence.

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2. The model

The model shown in Fig. 1 is the same as that given by Lawless et al. (2009), but transitions occurring during the read-out stage are included in addition to the transitions taking place during the excitation by irradiation. The meaning of the given magnitudes is explained in the caption. The solid lines depict transitions taking place during excitation whereas dashed lines are associated with those occurring during read-out. We start by repeating the previously mentioned (Lawless et al., 2009) set of 4 simultaneous differential equations governing the excitation process.

$$\frac{dn}{dt} = A_n(N-n)n_c, \tag{1}$$

$$\frac{dn_c}{dt} = X - \frac{dn}{dt} - A_m m n_c, \tag{2}$$

$$\frac{dm}{dt} = Bn_v(M-m) - A_m m n_c, \tag{3}$$

$$\frac{dn_v}{dt} = X - Bn_v(M-m) \tag{4}$$

As pointed out in the previous paper, we are mainly interested here in dose dependence far from saturation, and therefore choose the parameters so as to have $n << N$ and $m << M$. Note that electrons are raised by the irradiation at a rate X ($cm^{-3}\,s^{-1}$) which is proportional to the dose rate. $D = X \cdot t$ (cm^{-3}) is the total concentration of produced electrons and holes, which is proportional to the total applied dose. Lawless et al. (2009) reached the following expression for the dependence of the concentration of trapped electrons and holes

$$n = m = \frac{A_n N}{A_m}\left(\sqrt{1+\frac{2A_m}{A_n N}Xt} - 1\right), \tag{5}$$

which may be sublinear with the dose D. More specifically, they showed that for small doses, i.e., for $Xt << A_nN/(2A_m)$, this is approximately linear, $n \approx Xt$. For high doses (but far from saturation), $Xt >> A_nN/(2A_m)$, expression (5) reduces to

$$n = m \approx \sqrt{\frac{2A_n N}{A_m}Xt}, \tag{6}$$

which is a $D^{1/2}$ behavior.

In order to follow the process taking place during the heating of the sample, we have to solve numerically the relevant set of coupled rate equations

$$\frac{dn}{dt} = A_n(N-n)n_c - s \cdot n \cdot \exp(-E/kT), \tag{7}$$

$$\frac{dn_c}{dt} = -\frac{dn}{dt} - A_m m n_c, \tag{8}$$

$$\frac{dm}{dt} = -A_m m n_c, \tag{9}$$

where E(eV) is the activation energy and s(s^{-1}) the frequency factor. Using a chosen heating function $T(t)$, the linear heating function $T = T_0 + \beta t$ being a popular choice, we can write

$$I(T) = -\frac{dm}{dt} = A_m m n_c, \tag{10}$$

where $I(T)$ denotes the TL emitted light intensity.

As for the simulation of LM-OSL, exactly the same Eqs. (1)–(4) are relevant for the excitation stage whereas for the read-out phase, Eqs. (7)–(10) hold with f (s^{-1}) proportional to the stimulating light intensity replacing $s \cdot exp(-E/kT)$ in Eq. (7). If we denote by F ($cm^{-2}\,s^{-1}$) the stimulating light intensity and by a (cm^2) the optical cross section, then $f = Fa$. For f being linearly dependent on time, we take $f = f_0 \cdot t$ where f_0 (s^{-2}) is constant.

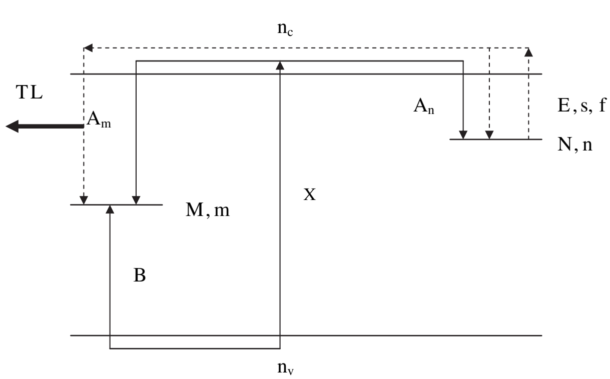

Fig. 1. Energy level scheme of the OTOR model. N and M (cm^{-3}) are the concentrations of the electron trapping states and hole recombination centers, and n and m (cm^{-3}) their occupancies, respectively. X ($cm^{-3}\,s^{-1}$) is the rate of production of free electrons and holes in the conduction and valence bands, respectively. n_c and n_v (cm^{-3}) are the concentrations of free electrons and holes in the respective bands. A_n and B ($cm^3\,s^{-1}$) are the trapping-probability coefficients of electrons in traps and holes in centers, respectively, and A_m ($cm^3\,s^{-1}$) is the recombination probability coefficient of free electrons with trapped holes in the centers.

3. Numerical simulation of TL and LM-OSL

In order to show the dose dependence of the trapped electrons, we chose sets of trapping parameters, and solved numerically Eqs. (1)–(4) using the Matlab ode23s solver for a certain period of time t. This was followed by a relaxation period where $X = 0$. In the second stage of the simulation, the temperature has been raised at a constant rate of 1 °C/s, and Eqs. (7)–(9) were solved. When the maximum intensity was reached, it was registered as the TL signal.

Fig. 2 depicts the values of $m = n$ as the dashed line, and the TL maximum intensity as the solid line for the set of parameters given in the caption. The concentration curve is the same as given by Lawless et al. (2009) for the same set of parameters. These authors have shown that with this set of parameters, the dose dependence is very closely a square-root function as shown in Eq. (6). The temperature of the TL peak was constant for these results, $T_m = 449$ K. Note that whereas the values of $n = m$ are given as received from the simulations with the given parameters. The calculated values of the TL curves were multiplied by a factor of 45, which made the two curves to practically coincide, and than were shifted by a constant value so that the two curves are seen separately.

Fig. 3 shows the results of the maximum TL and $n = m$ for the set of parameters given in the caption. The trapping parameters are the same as in Fig. 2 except that the retrapping coefficient probability is 3 orders of magnitude smaller. The intensity of excitation $X = 10^4$ cm^{-3} s^{-1} is significantly lower here, by 10 orders of magnitude, than in the case of Fig. 2. Obviously, in this case, the traps and centers are very far from saturation. The concentration $n = m$ and maximum TL curves are both sublinear. The values on the TL curve have been multiplied by a factor of ~80 so that the two curves can be easily seen on the same graph. The maximum TL

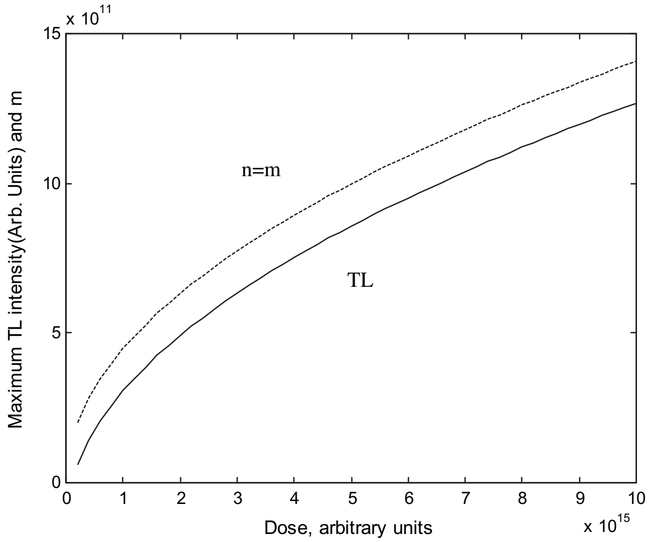

Fig. 2. Maximum TL intensity (solid line) and concentration of electrons in traps and holes in centers ($n = m$) (dashed line) as simulated by solving the mentioned sets of coupled differential equations for $A_m = 10^{-5}$ cm^3 s^{-1}; $A_n = 10^{-11}$ cm^3 s^{-1}; $B = 10^{-10}$ cm^3 s^{-1}; $N = 10^{14}$ cm^{-3}; $M = 10^{15}$ cm^{-3}; $X = 10^{14}$ cm^{-3} s^{-1}. The irradiation time varies between 2 and 100 s. For the two lines to be seen separately, the dashed line is shifted upward.

temperature varied with the dose, changing gradually from 479 K to 452 K as the dose increases. One should note that such large variations in the dose rate of excitation are not uncommon in the study of TL dating, when one compares the natural and laboratory dose rates. Simulations with the same trapping parameters and higher dose rates have also been performed. With $X = 10^6$ cm^{-3} s^{-1}, the two (normalized) curves were sublinear and came closer together. With $X = 10^8$ cm^{-3} s^{-1}, the normalized curves were even closer and with $X = 10^{10}$cm^{-3} s^{-1}, they practically coincided. With such high values of X (and therefore of D), the dose dependence of both was practically a square-root function, which is consistent with the above mentioned transition from Eqs. (5) and (6).

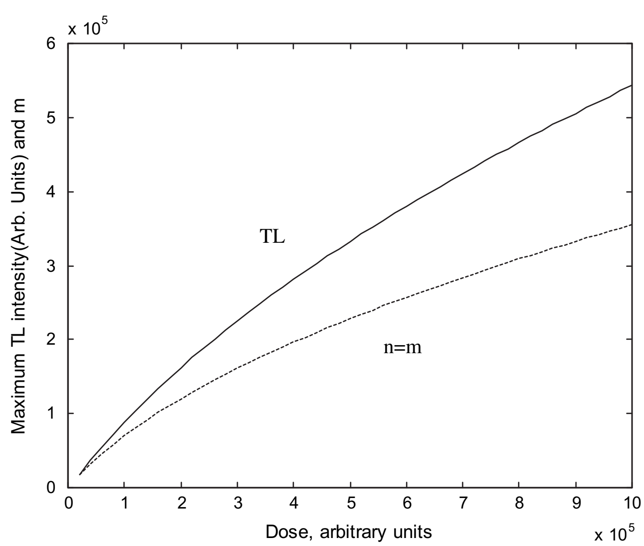

Fig. 3. Maximum TL intensity (solid line) and concentration of electrons in traps and holes in centers ($n = m$) (dashed line) as simulated by solving the mentioned sets of coupled differential equations for $A_m = 10^{-5}$ cm^3 s^{-1}; $A_n = 10^{-14}$cm^3 s^{-1}; $B = 10^{-10}$ cm^3 s^{-1}; $N = 10^{14}$ cm^{-3}; $M = 10^{15}$ cm^{-3}; $X = 10^4$ cm^{-3} s^{-1}. The irradiation time varies between 2 and 100 s. The TL intensities were normalized so that the two curves coincide in the lowest dose.

Fig. 4 shows the LM-OSL curves with the same parameters as in Fig. 3. The excitation and relaxation procedures are exactly the same as in TL, whereas the read-out is simulated using Eqs. (7)–(9) with the amendment mentioned above of replacing $s \cdot exp(-E/kT)$ in Eq. (7) by f, and using the linear increase of the stimulating light, simulated by $f = f_0 \cdot t$. Like in the TL case shown in Fig. 3, the traps and centers here are very much below saturation.

4. Discussion

The condition for a TL peak to be of first order is $mA_m >> A_n(N-n)$ along with $n = m$ (see e.g., p. 29, Chen and McKeever, 1997). For getting second order, one has to have $mA_m << A_n(N-n)$ along with $n = m$ and $n << N$, i.e., the trap is far from saturation. Of course, intermediate cases of neither first nor second order may occur very frequently. With the basic model of one trapping state and one kind of recombination center, the condition $n = m$ always holds at the end of the irradiation.

The results depicted in Fig. 2 show sublinear dose dependence of the concentrations n and m as well as the maximum intensity. As pointed out by Lawless et al. (2009), the dependence of the trapped electrons concentrations is exactly of $D^{1/2}$ and as shown here, this is also the case for I_m. In the results, $n = m$ goes between 2×10^{11} and 1.4×10^{12} cm^{-3} and this yields $mA_m = (2 \times 10^6–1.4 \times 10^7)$ s^{-1}. We also get $A_n(N-n) \approx 10^{-11} \times 10^{14} = 10^3$ s^{-1}. It is obvious that $mA_m >> A_n(N-n)$ and the first-order condition prevails. This agrees very well with the fact that I_m is proportional to n and the fact that the maximum temperature does not vary with the excitation dose. With $X = 10^{14}$ cm^{-3} s^{-1}, the dose varies between 2×10^{14} and 10^{16} cm^{-3}. With these parameters, $A_nN/(2A_m) = 5 \times 10^7$ cm^{-3} and obviously, $D >> A_nN/(2A_m)$ which indicates a $D^{1/2}$ dose dependence of the trap occupancy at the end of excitation, as indeed is seen in the graph.

Fig. 3 shows also a situation where the trap occupancy and the maximum TL intensity depend sublinearly on the dose, but not in the same way. The dose D varies between 2×10^4 cm^{-3} and 10^6 cm^{-3}. With the parameters quoted in the caption, $A_nN/(2A_m) = 5 \times 10^4$ and therefore, one has $D < A_nN/(2A_m)$ at low doses, but $D > A_nN/(2A_m)$ at higher doses. Obviously, one has here a transition from one behavior to another as the dose grows. Neither of the two sublinear curves behaves like $D^{1/2}$ with the dose. The values of $m = n$ are seen to be between 2×10^4 and 3.5×10^5 cm^{-3}.

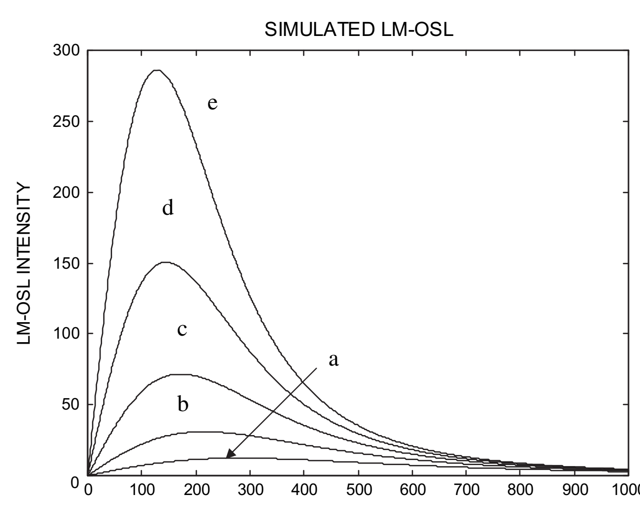

Fig. 4. Simulated LM-OSL curves for the same set of parameters as in Fig. 3. Excitation times are 1, 2, 4, 8, 16 s in curves a, b, c, d and e, respectively.

Therefore, mA_m varies from 0.2 to 3.5 s^{-1} whereas $A_n(N-n) = 1$ s^{-1}. The kinetics here is obviously intermediate in some sense between first and second order. It should be noted that the TL results shown in Fig. 3 are the combination of the sublinear dose dependence of the electron occupancy at the end of the irradiation, and a somewhat superlinear dependence of I_m on this occupancy. Comparison of the two curves shows that $I_m \propto n_0^{1.1}$.

Concerning Fig. 4, showing LM-OSL, similarly to the TL shown in Fig. 3, the kinetics is intermediate, mA_m varies from 0.2 to 3.5 s^{-1} whereas $A_n(N-n) = 1$ s^{-1} and the kinetics order is intermediate between first and second order. Comparing the maximum intensity of the simulated LM-OSL for curves (d) and (e) shows that for a factor of 2 in the dose, the maximum intensity changes by 2.51. This is superlinear, but less than $2^{3/2} = 2.83$ characterizing the second-order kinetics. As for the maxima of curves (a) and (b) their ratio is 1.90 for a dose ratio of 2, which means that the dose dependence is sublinear, but the ratio is more than $\sqrt{2}$ described in Eq. (6). t_m varies here with the dose, but much more moderately than in the pure second-order case.

5. Conclusion

In this work, we have demonstrated that within the simple one trap-one recombination center (OTOR) model different behaviors of the carrier occupancies and the maximum TL intensity as a function of the excitation dose may take place. Sublinear dose dependence, sometimes behaving like $D^{1/2}$, of the trap and center occupancies has been reported before. With certain sets of parameters, the maximum TL intensity was strictly proportional to the concentration and both behaved like the square root of the dose; here, first-order features are observed. With other sets, both the occupancies and the TL signal were sublinear, but the dose-dependence functions were different. As far as the kinetics order is concerned, these were intermediate situations. In each of these cases, the relations between the parameters leading to first- and second-order as well as intermediate kinetics, and the conditions for linear and square-root dose dependence have been considered.

The simulations of LM-OSL are also interesting and, for the same sets of parameters are commensurate with the TL results. Sublinear dose dependence of the maximum signal, behaving like $D^{1/2}$ can be found with sets of parameters leading to first-order kinetics and to Eq. (6), and superlinear dependence going like $D^{3/2}$ with sets of parameters leading to second-order kinetics. Intermediate cases can also be reached, in which the dose dependence may change from superlinear to sublinear as the dose increases, even when the relevant trapping states are far from saturation.

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