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Nonlinear dose dependence and dose-rate dependence of optically stimulated luminescence and thermoluminescence

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Abstract

When thermoluminescence (TL) and optically stimulated luminescence (OSL) are utilized for dosimetry and for dating of archaeological and geological samples, one hopes that the dependence of the measured signal on the dose is linear, and that no dose-rate effects occur. In TL measurements, however, several cases of superlinear dose dependence have been reported and also some dose-rate effects have been found. It has been shown theoretically that such superlinearity can result from a simple model of trapping states and recombination centers, provided that a disconnected competing trap or center is involved. Similar circumstances were shown to cause a dose-rate dependence of the measured TL. More recently, some results of OSL superlinearity have been reported. The present work provides a theoretical account of this effect. A distinction is made between OSL due to relatively short pulses of stimulating light and the integral over a long illumination. It is shown that in the former, one can expect a quadratic dose dependence of the effect provided one starts with empty trapping states and recombination centers. In the latter, superlinearity can be found only in the presence of competitors, in a similar way to the TL behavior. Also, the possibility of dose-rate dependence of OSL, which has not been reported in the literature is predicted and should be checked in future OSL measurements. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: Optically stimulated luminescence; Thermoluminescence; Nonlinearity; Dose-rate effect

1. Introduction

The utilization of thermoluminescence (TL) and optically stimulated luminescence (OSL) for the evaluation of irradiation dose either for dosimetry or for the dating of archaeological or geological specimens is based on the possibility of comparing the results following the unknown dose to those resulting from a calibrated radiation source. In order to get reliable results, the nature of the trapping levels in the dosimetric material should be such that the interpretation of the measured luminescence signal should be as simple as possible. Thus, in an ideal dosimetric material one hopes that the dose dependence of the signal is linear with the excitation dose, which makes the interpolation or extrapolation performed while evaluating the unknown dose very simple. Also, if it is the total dose that is to be evaluated, the question should be asked whether the measured signal is independent of the dose rate. Many of the works in the literature in this field either assumed linear dose dependence and dose-rate independence or, in cases where nonlinearities occurred, some corrections were offered so that the deduced dose (or the date derived thereof) is as accurate as possible. A quick look at the rate equations governing the kinetics of the process taking place during the excitation and the heating of the sample (in TL), or the exposure to stimulating light (in OSL) reveals that they are not linear in the relevant quantities such as the occupation of trapping states and luminescence centers. Linearity of the measured luminescence signal with the excitation dose is expected in a strictly first order kinetics, but it is recognized that such a simple situation is rather a rare occurrence. In other cases,

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the question of dose-dependence linearity should be more thoroughly considered.

Different kinds of nonlinear TL dose dependence were described in the literature. Cameron et al. (1968), summing up results by their group, described the dose dependence of TL in LiF (TLD-100) which is linear at doses up to $\sim$10 Gy, goes superlinear at higher doses and then approaches saturation at $\sim 10^4$ Gy. Zimmerman and Cameron (1968) presented a model that had to do with filling of existing traps in the presence of a disconnected trap (or nonradiative center) which is active during the excitation. Aitken (1974) gave a summary of dose dependence of TL from pottery quartz, found by the Oxford Group. This is superlinear up to $\sim$2.5 Gy and continues linearly up to at least 6 Gy of β irradiation. Rodine and Land (1971) reported a dose dependence of approximately D^2 in one peak out of a series of TL peaks in UV irradiated ThO_2. They suggested a model which was related to the occurrence of competition during the heating phase. This model was further developed by Kristianpoller et al. (1974) and the same model was later utilized by Chen et al. (1988) to explain their finding of strong superlinearity (up to approximately D^3) in the 110°C peak in synthetic quartz under β irradiation. Chen and Fogel (1993) argued that in every situation where a competitor is involved, the processes both during excitation and heating should be considered. For further discussion on this subject see Chen and McKeever (1997). It should be noted that similar considerations have been made independently by Jaek et al. (1974).

Dose-rate dependence of TL has also been reported in the literature. Groom et al. (1978) reported a decrease of TL by up to a factor of 5 while the dose rate increased in powdered samples of Brazilian quartz irradiated by ^{60}Co γ-rays ranging from 1.4×10^{-3} to 3.3 Gy s^{-1}. A smaller effect of the same sort was reported by Hsu and Weng (1980). An opposite effect of higher TL for larger dose rate was reported by Kvasnička (1983) who found the effect in Brazilian and milky quartz excited by ^{60}Co γ-ray, using dose-rates from 2×10^{-5} to 2×10^{-2} Gy s^{-1}. The apparent discrepancy between the two sets of results is probably due to the different ranges of applied dose. Valladas and Ferreira (1980) distinguished between three components in the emission of TL in quartz, namely, UV, blue and green, and found different behaviors for the three components.

Theoretical work by McKeever et al. (1980) and Chen et al. (1981) demonstrated that using rather simple models of trapping states and recombination centers, one can obtain a dose-rate dependency of trap filling. Chen et al. (1981) also showed that, as far as the center filling is concerned, when two centers exist in the same material, it is possible for the population of one of them to increase and that of the other to decrease with the dose rate for a given total dose. In a recent work, Chen and Leung (2000) took into consideration also the heating stage which follows the excitation. For a model with one trapping state and two kinds of recombination centers, they solved the relevant sets of simultaneous differential equations for the three stages of excitation, relaxation and heating. They found different dose-rate dependencies for two TL spectral components, in accord with the experimental results by Valladas and Ferreira (1980).

In recent years, optically stimulated luminescence has started to replace TL in some of the dosimetry and dating applications. This began with a pioneering work by Huntley et al. (1985) on the optical dating of sediments. The method has more recently also been utilized for archaeological dating and dosimetry. The advantages of OSL over TL are rather obvious. There is no need to heat the sample, thus avoiding the black-body radiation occurring at relatively high temperature. Also, possible thermal quenching of luminescence is avoided.

In nearly all the reports on OSL it is assumed, and sometimes shown (see e.g. McKeever and Akselrod, 1999) that the initial dose dependence is linear, followed by an approach to saturation. It is also assumed that there are no dose-rate effects and therefore, one can calibrate the sample at high dose-rates and deduce the archaeological dose imparted at a much lower rate. There are, however, some reports in the literature on superlinear dose dependence of OSL. In the study of OSL of quartz and mixed feldspars from sediments, Godfrey-Smith (1994) found linear dependence on the dose of the unheated samples. However, following a preheat at 225°C, the samples showed a clear superlinearity of the OSL signal at low excitation doses of γ irradiation. Roberts et al. (1994) have also found superlinearity of quartz OSL in several samples. For samples preheated at 160°C, they reported a quadratic equation, $y = aD^2 + bD + c$, which describes the dose dependence where D is the dose and a, b and c are positive numbers.

Banerjee (2001) has also seen superlinear dose dependence of OSL on the excitation dose in annealed quartz in the dose range of 0–5 Gy and explained it using arguments previously employed by Kristianpoller et al. (1974), and by Chen and McKeever (1997) for accounting for different kinds of TL superlinearity. The possibility of using arguments applicable for TL in explaining OSL results will be elaborated upon below. In the present work, we discuss the similarities and dissimilarities between TL and OSL as far as the dose dependence and dose-rate behavior are concerned. Also, the distinction is made between short pulse OSL and integral OSL taken over a long illumination of a previously irradiated sample. It is to be noted in this respect that in his book, Aitken (1998) considers the use of the "total light sum" in certain samples or, alternatively, a "short shine" for bright samples. He adds that this can be made short enough for the depletion of trapped electrons to be negligibly small (e.g. a fraction of 1%), allowing repeated measurements to be made on the same aliquot. We are going to argue here that using the total light sum and the "short shine" may not yield the same results since the dose dependencies of these two measured quantities are not necessarily the same.

The comparison between TL and OSL as far as their dose dependence and dose-rate dependence are concerned is for the simplest situation of one-trap one-center situation as well as for the cases in which a disconnected trapping state or a radiationless recombination center take part in the process.

2. The model

The considerations made by Banerjee (2001) are, on first sight, somewhat questionable due to the following reasons. In the first place, he takes the arguments previously made for TL and adopts them to OSL. One may ask whether the use of this analogy is fully justified. Also, the considerations made by Kristianpoller et al. (1974) include a large number of simplifying assumptions. The previously published work on TL showed, using computations and without simplifying assumptions, that at least for certain choices of sets of parameters, the expected TL superlinearity indeed resulted. It is of interest to compare the TL results to the analogous case in OSL. In particular, the question of whether the important magnitude to be measured is the area under the relevant curve or an instantaneous intensity, say in the maximum of the TL peak or somewhere along the OSL decay curve, is of great importance both for TL and OSL. With the use of new numerical simulations, we would like to answer some of these questions.

We shall discuss here the models of OSL dose dependence with and without a competing disconnected trapping state or a nonradiative center. Fig. 1 depicts an energy level model with one recombination center M and two trapping states, N_1 and N_2. At first, we will consider the situation where the competitor N_2 does not exist, and will refer to N_1, the remaining trapping state as N (N being also its concentration in m^{-3}); its occupancy will be denoted here by n (m^{-3}). M is the concentration (m^{-3}) of hole centers and m (m^{-3}) is its instantaneous occupancy. B ($\mathrm{m}^3\ \mathrm{s}^{-1}$) is the probability coefficient for free holes to be captured in the recombination center during excitation. A_{n1}, A_{n2} ($\mathrm{m}^3\ \mathrm{s}^{-1}$) are the probability coefficients for retrapping into N_1 and N_2, respectively (again, as long as we deal with one trapping state only, N, we denote the retrapping coefficient by A_n). A_m ($\mathrm{m}^3\ \mathrm{s}^{-1}$) is the recombination coefficient. n_c (m^{-3}) and n_v (m^{-3}) are the concentrations of free electrons in the conduction band and free holes in the valence band, respectively, x and f represent, respectively, the intensity of excitation by the irradiation and the intensity of the light stimulation. The difference between the meaning of these two magnitudes will be explained below.

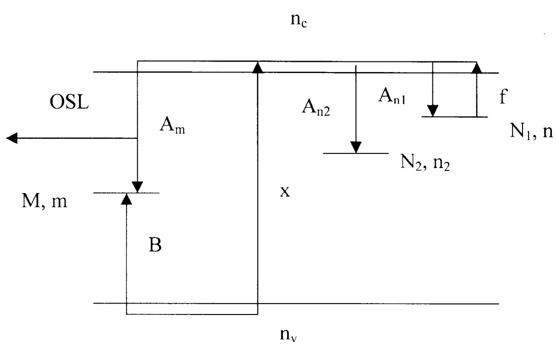

Fig. 1. Energy level scheme for excitation and stimulation of OSL. An active level, N_1 and a center, M, are involved, as well as a disconnected competing trapping state, N_2. For the case of only one trapping state, N replaces N_1.

The set of simultaneous differential equations governing the process during excitation is given by

$$\mathrm{d}n_v/\mathrm{d}t = x - B(M - m)n_v, \tag{1}$$

$$\mathrm{d}m/\mathrm{d}t = -A_m m n_c + B(M - m)n_v, \tag{2}$$

$$\mathrm{d}n/\mathrm{d}t = A_n(N - n)n_c, \tag{3}$$

$$\mathrm{d}n_c/\mathrm{d}t = \mathrm{d}m/\mathrm{d}t + \mathrm{d}n_v/\mathrm{d}t - \mathrm{d}n/\mathrm{d}t. \tag{4}$$

x appearing in Eq. (1) has the dimensions of $\mathrm{m}^{-3}\ \mathrm{s}^{-1}$ and is the rate of production of electron–hole pairs per second per m^3, by the excitation irradiation. The intensity of irradiation in the appropriate units (say, Gy s^{-1}) is proportional to x.

In order to simulate the experimental conditions properly, we take the final values of n, m, n_c and n_v at the end of the excitation stage as initial values for the next stage of relaxation. This is done by setting x to zero and solving the same set of equations for a further period of time until both n_c and n_v get negligibly small.

For the next stage of light stimulation we take the final values of the functions n, m, n_c and n_v at the end of relaxation and solve the following equations:

$$-\mathrm{d}m/\mathrm{d}t = A_m m n_c, \tag{5}$$

$$\mathrm{d}n/\mathrm{d}t = -fn + A_n(N - n)n_c, \tag{6}$$

$$\mathrm{d}n_c/\mathrm{d}t = \mathrm{d}m/\mathrm{d}t - \mathrm{d}n/\mathrm{d}t. \tag{7}$$

It is quite obvious that the dimension of f here is per second. In this sense, fn is analogous to x in Eq. (1). However, whereas x is a constant, fn is not. For constant stimulation intensity, f is a constant which is proportional to the stimulating light intensity. We associate the intensity of the OSL signal with the recombination rate, and therefore can write the OSL intensity as

$$I = -\mathrm{d}m/\mathrm{d}t. \tag{8}$$

In fact, a dimensional proportionality factor should have been inserted into the right-hand side of Eq. (8), which would determine the dimensions of the intensity I. Usually this is not done, and therefore the intensity in Eq. (8) is in "arbitrary units". It is to be noted here that the I appearing on the left-hand side is an instantaneous intensity. In the treatment below we will distinguish between this intensity in response to a relatively short pulse of light and an integral of the intensity over a long period of time, preferably all the way to the depletion of all the traps and/or centers involved. Some computational results are given in the next section.

We will describe here the changes made in the relevant sets of differential equations in the case where a trapping competitor exists, as shown in Fig. 1. Eqs. (1) and (2) remain the same, but instead of Eqs. (3) and (4) we now have for the excitation phase

$$dn_1/dt = A_{n1}(N_1 - n_1)n_c, \tag{9}$$

$$dn_2/dt = A_{n2}(N_2 - n_2)n_c, \tag{10}$$

$$dn_c/dt = dm/dt + dn_v/dt - dn_1/dt - dn_2/dt. \tag{11}$$

As for the stimulation stage, we assume that the stimulating light raises electrons only from N_1, so that N_2 acts only as a competitor. Eqs. (5) and (8) remain the same, and Eqs. (6) and (7) are replaced by

$$dn_1/dt = -fn_1 + A_{n1}(N_1 - n_1)n_c, \tag{12}$$

$$dn_2/dt = A_{n2}(N_2 - n_2)n_c, \tag{13}$$

$$dn_c/dt = dm/dt - dn_1/dt - dn_2/dt. \tag{14}$$

Here too, the computational results are given in the following section.

As pointed out above, in the study of TL dose dependence, a significant difference has been found between a model including a competition with a disconnected trap and one where competition takes place with a nonradiative center. As compared with Fig. 1, the model here has one trapping state N with occupancy n, a retrapping coefficient A_n, and two recombination centers, M_1 and M_2 with occupancies m_1 and m_2, and recombination coefficients A_{m1} and A_{m2}, respectively. Also, the two coefficients B_1 and B_2 for trapping holes in M_1 and M_2 are to be considered. We assume here that the measured OSL results from transitions into M_1 whereas the transitions into M_2 are radiationless. The governing set of equations for the excitation stage here is

$$dn_v/dt = x - B_1(M_1 - m_1)n_v - B_2(M_2 - m_2)n_v, \tag{15}$$

$$dm_1/dt = -A_{m1}m_1n_c + B_1(M_1 - m_1)n_v, \tag{16}$$

$$dm_2/dt = -A_{m2}m_2n_c + B_2(M_2 - m_2)n_v, \tag{17}$$

$$dn/dt = A_n(N - n)n_c, \tag{18}$$

$$dn_c/dt = dm_1/dt + dm_2/dt + dn_v/dt - dn/dt. \tag{19}$$

Finally, the set of equations for the stimulation stage under these circumstances can be written as

$$I = -dm_1/dt = A_{m1}m_1n_c, \tag{20}$$

$$-dm_2/dt = A_{m2}m_2n_c, \tag{21}$$

$$dn/dt = -fn + A_n(N - n)n_c, \tag{22}$$

$$dn_c/dt = dm_1/dt + dm_2/dt - dn/dt. \tag{23}$$

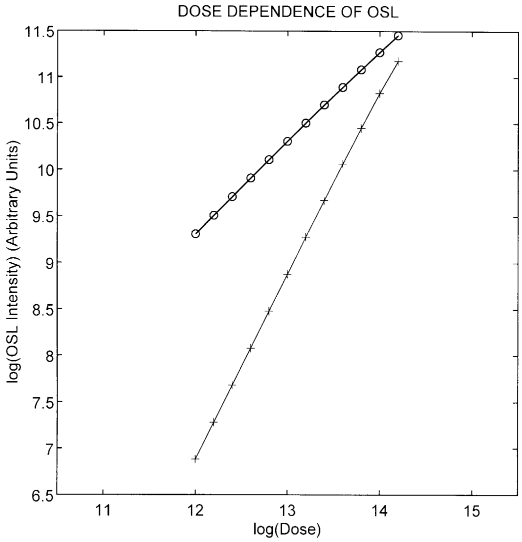

Fig. 2. Dose dependence of pulse OSL when only one trapping state and one kind of recombination center are involved. (+) represents the results when the excitation starts with empty traps and (○) the results when $n_0 = 0.9N$. The values of the chosen parameters are given in the text.

3. Numerical results

In order to demonstrate the dose behavior of OSL with a short pulse of stimulation, the following set of parameters has been chosen. $A_m = 10^{-17}$ m^3 s^{-1}; $B = 10^{-18}$ m^3 s^{-1}; $N = 10^{17}$ m^{-3}; $M = 10^{19}$m^{-3} and $A_n = 10^{-19}$ m^3 s^{-1}. The value of f was taken as 1 s^{-1} and x was varied between 10^{12} and 10^{14} m^{3}s^{-1}. The standard ode23 solver in the Matlab package has been used to solve Eqs. (1)–(4) with the given value of x, then the same set has been solved with $x=0$ for a further period of time to simulate relaxation. Finally, Eqs. (5)–(7) were solved with the given value of f. The results are shown in Fig. 2, on a log–log scale. The (+) points show the dependence of the simulated OSL signal on the total dose, changed here by changing the intensity of excitation x in the mentioned range, starting each simulation with empty traps and centers and keeping the length of excitation time at 1 s. The length of light stimulation was also chosen as 1 s. It has been found that with this set of chosen parameters, this indeed was a "short shine" in the sense that only a small fraction of the trapped electrons and holes was depleted by the light during the stimulation time. On this log–log scale, the results are seen as a straight line with a slope of 2, meaning quadratic dependence of the pulse OSL on the excitation dose. The (○) points show the dose dependence where $n_0 = 0.9N$, and the straight line with slope $\cong 1$ represents a linear dose dependence.

Following this study of pulse OSL, we have now moved to the solution of the equations of integral OSL in the case of one trapping state and one kind of recombination center.

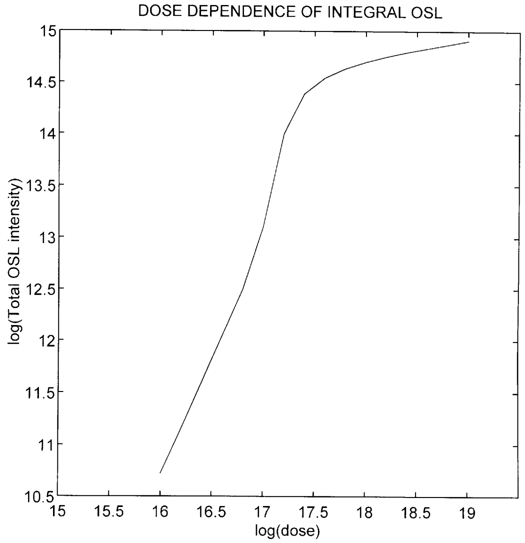

Fig. 3. Simulated dose dependence of integrated OSL for the case of two trapping states and one kind of recombination center. The parameters are given in the text.

We performed the integration on the OSL curve up to a point where < 1% of the carriers remained in the traps. Under the present circumstances, the area under the curve grew linearly up to a point where it started to approach saturation. The comparison between this result and the response to a short light pulse described above will be discussed below.

The next step has been to solve the sets of equations for the case of competition with a disconnected trapping state. The sets of equations numerically solved have been (1), (2), (9)–(11) for the excitation and relaxation stages, and (6), (7), (12)–(14) for the optical stimulation stage. The set of parameters chosen here was: $A_m = 10^{-17}$ m^3 s^{-1}; $B = 10^{-16}$ m^3 s^{-1}; $N_1 = 10^{17}$ m^{-3}; $N_2 = 10^{16}$ m^{-3}; $A_1 = 10^{-16}$ m^3 s^{-1}; $A_2 = 10^{-15}$ m^3 s^{-1}; $M = 10^{18}$ m^{-3}; $f = 1$ s^{-1} and x varied from 10^{16} to 10^{19} m^{-3} s^{-1}. The results are shown on a log–log scale in Fig. 3. The initial slope of 2 indicates the initial quadratic dose dependence. At a higher dose, the slope gets larger indicating more than quadratic dose dependence, and at higher doses, a rather fast decline of the slope is seen when the simulated OSL approaches saturation. The significance of this kind of behavior will be discussed below.

With the same set of parameters, we have checked the possibility of having dose-rate dependence of integral OSL. Here, we choose a total dose of $xt = 10^{17}$ m^{-3} in the range of strong superlinear dose dependence, varying the dose rate between 10^{15} and 10^{19} m^{-3} s^{-1} and varying inversely the time of excitation between 100 and 0.01 s. The results are shown in Fig. 4. The integral OSL is seen to increase in this dose-rate range by a factor of > 2.

Finally, the set of equations governing the process in the case of one trapping state and two competing centers

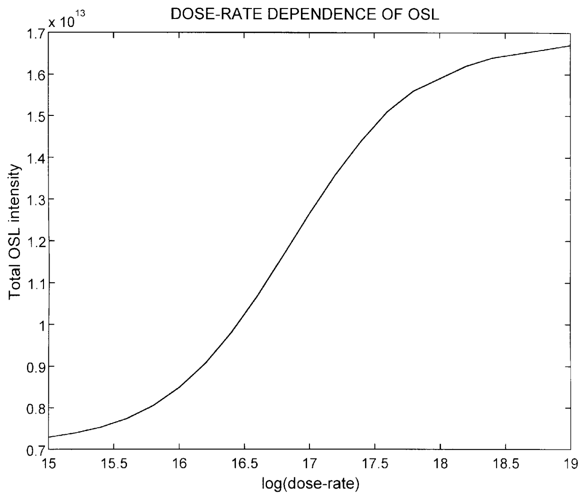

Fig. 4. Dose-rate dependence of integral OSL in the case of two trapping states and one kind of recombination center. The chosen parameters are the same as in Fig. 3.

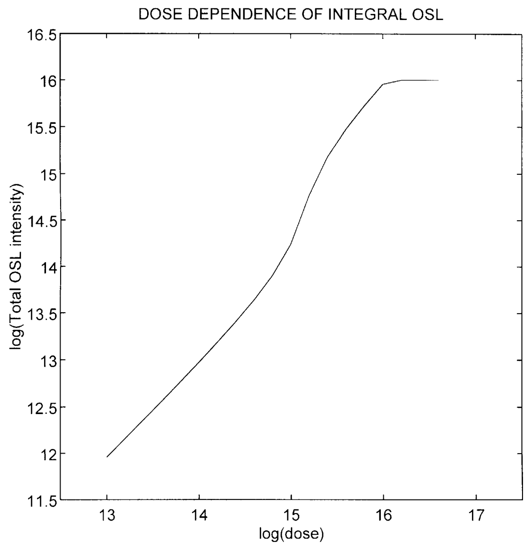

Fig. 5. Simulated dose dependence of integrated OSL on the dose in the case of two kinds of recombination center and a single trapping state. The parameters are given in the text.

has been numerically solved. These are Eqs. (15)–(19) for the excitation and relaxation stages and (20)–(23) for the stimulation stage. The parameters chosen here were: $A_{m1} = 10^{-17}$ m^3 s^{-1}; $A_{m2} = 10^{-16}$ m^3 s^{-1}; $B_1 = 10^{-14}$ m^3 s^{-1}; $B_2 = 10^{-12}$ m^3 s^{-1}; $N = 10^{20}$ m^{-3}; $M_1 = 10^{16}$ m^{-3}; $M_2 = 10^{15}$ m^{-3} and $A = 10^{-16}$ m^3 s^{-1}. f was chosen here to be 1 s^{-1} and x varied between 10^{13} and 10^{17} m^{-3} s^{-1}. The results of the integral OSL are seen in Fig. 5. The initial dependence is linear, indicated by the slope being equal to 1. At higher doses, the dependence becomes superlinear, and at still higher doses, there is an approach to saturation.

4. Discussion and conclusions

In the present work, we have demonstrated, using numerical simulation, that superlinearity in the dose dependence of the OSL signal, recently reported to occur in some materials, is in accord with rather simple energy level models, usually employed for the explanation of OSL and TL. It has been shown that, as far as short pulses of OSL are used, the simplest model of a single trapping state and one kind of recombination center can yield a quadratic dose dependence. Under the same circumstances, the total area under the OSL curve behaves linearly with dose before approaching saturation, in a similar manner to the TL signal under similar conditions. The similarities and dissimilarities between TL and OSL under these conditions should be elaborated upon. As far as the areas under the two curves are concerned, both are dependent on $\min(n_0, m_0)$ where n_0 and m_0 are the fillings of the relevant trap and center at the end of irradiation (see discussion by Chen and McKeever, 1997). As long as the occupancies n_0 and m_0 are linear with the dose, which obviously is the case in this simple model, the dependence of the total areas under the TL curve and under the OSL decay curve on the dose is expected to be linear. As far as TL is concerned, the maximum intensity of TL was usually found to be proportional to, or nearly proportional to the total area. Therefore, the maximum intensity of TL is proportional to the excitation dose in the simple case of one trapping state and one kind of recombination center.

The situation appears to be different with OSL as seen in Fig. 2 above. The first point to remember in this concern is that in OSL, there is no obvious analog to the maximum of TL. The short pulse response of OSL is more similar to the intensity of TL in the initial rise region. Here we can discuss two extreme cases. If the kinetic order of the process is first, the intensity at each temperature in the TL curve is proportional to the initial filling of the traps, n_0. Therefore, if the dose dependence is examined at any temperature along the curve, it is expected to be linear. This is not the case in second order kinetics. As pointed out by Chen et al. (1983), in a second order kinetics TL peak, the dose dependence is expected to be quadratic within the initial rise range. Although the exact conditions for such a quadratic behavior for cases beyond the strict second order kinetics have not been specified, it is certainly possible that the dose dependence in the initial rise range is superlinear. It seems that there is a close analogy between the initial-rise range of TL and short pulse OSL since in both we sample the specimen, depleting only a negligible fraction of the carriers in traps and centers. It seems quite obvious that the quadratic dose dependence of pulse OSL seen in Fig. 2 is the correct analog of the quadratic dose dependence of the mentioned TL in the initial rise range.

It can be noted that the main condition for second order kinetics is that the retrapping is relatively strong, namely,

$$A_m m \ll A_n(N - n). \tag{24}$$

Taking a look at the parameters chosen above, it is evident that, at least at relatively low doses, where m and n are small, this condition is fulfilled. It can even be argued that strict first order kinetics, a case in which TL is proportional to the dose at any given temperature point, is almost impossible to occur if irradiation starts with empty traps and centers. Strict first order will take place when A_n is very small. However, if it is that small, the trapping state can hardly capture carriers during the excitation stage. In other words, in practically all cases, there is an "element" of second order-like situation due to which the quadratic behavior of the pulse OSL dose dependence may be expected to happen quite often. As pointed out already, this is more likely to occur in annealed samples, and is less likely to take place when we start with a nearly full trapping state or center. A quick look at condition (24) reveals that if n_0 or m_0 (or both) is large, the direction of the inequality may easily be reversed, which explains the linearity of the upper curve in Fig. 2.

The question may arise as to what happens when one uses, under these conditions, a relatively long exposure which is not long enough to cover the whole decaying OSL curve. Roberts et al. (1994) describe their OSL signal which is "integrated over the first 500 mJ of laser exposure", which fits the $y = aD^2 + bD + c$ equation mentioned above. This may be a good approximation to an intermediate case of a long pulse which does not cover the whole area under the decaying curve.

The situation concerning dose dependence and dose-rate dependence is different when the total area under the decaying OSL curve is considered. We have shown here that under these circumstances, superlinearity is possible provided a competitor participates in the process. Fig. 3 shows the expected dose dependence in the presence of a disconnected trap. Similar to the dose dependence of the maximum of TL (see Chen and McKeever, 1997, p. 179), the dependence starts being quadratic and goes more strongly superlinear before going to saturation. In Fig. 5, it is shown that in the presence of a nonradiative competitor, the dose dependence is linear at low doses, becomes superlinear at high doses and finally reaches saturation. This behavior is very similar to that of the TL maximum under similar conditions (see Chen and McKeever, 1997, p. 180). This point has been suggested by Banerjee (2001) who made the analogy with TL using the considerations by Kristianpoller et al. (1974) and further by Chen and McKeever (1997) made for TL. However, two points are to be borne in mind. The first is that the *maximum* of TL is compared with the area under the OSL curve. This should not pose a severe problem since it has been pointed out that in practically all cases, the TL maximum is proportional to the area under the whole TL curve to a very good approximation. The second point is that in Banerjee's experimental results, the OSL intensity is defined as the integrated OSL signal between 0 and 1 s, and it is not clear whether this is closer to the pulse case or to the total area case discussed here. It appears, however, that Banerjee did have the correct intuition concerning the analogy between TL and integral OSL.

Finally, in Fig. 4 we have shown that under these conditions of measuring the total area under the OSL curve and in the presence of a competitor, a dose-rate effect can be expected. This effect has never been reported to the best of our knowledge, and it is recommended that its possible occurrence will be checked in the future.

References

Aitken, M.J., 1974. Physics and Archaeology, 2nd Edition. Oxford University Press, Oxford.

Aitken, M.J., 1998. An Introduction to Optical Dating. Oxford University Press, Oxford, NY, Tokyo.

Banerjee, D., 2001. Supralinearity and sensitivity changes in optically stimulated luminescence of annealed quartz. Radiat. Meas. 33, 47–51.

Cameron, J.R., Suntharalingam, N., Kenney, G.N., 1968. Thermoluminescence Dosimetry. University of Wisconsin Press, Madison, WI, USA.

Chen, R., Fogel, M., 1993. Superlinearity in thermoluminescence revisited. Radiat. Prot. Dosim. 47, 23–26.

Chen, R., Leung, P.L., 2000. A model for dose-rate dependence of thermoluminescence intensity. J. Phys. D 33, 846–850.

Chen, R., McKeever, S.W.S., Durrani, S.A., 1981. Solution of the kinetics equations governing trap filling. Consequences concerning the dose dependence and dose-rate effects. Phys. Rev. B 24, 4931–4944.

Chen, R., Huntley, D.J., Berger, G.W., 1983. Analysis of thermoluminescence data dominated by second-order kinetics. Phys. Stat. Sol. A 79, 251–261.

Chen, R., Yang, X.H., McKeever, S.W.S., 1988. The strongly superlinear dose dependence of thermoluminescence in synthetic quartz. J. Phys. D 21, 499–506.

Chen, R., McKeever, S.W.S., 1997. Theory of Thermoluminescence and Related Phenomena. World Scientific, Singapore.

Godfrey-Smith, D.I., 1994. Thermal effects in the optically stimulated luminescence of quartz and mixed feldspars from sediments. J. Phys. D 27, 1737–1746.

Groom, P.J., Durrani, S.A., Khazal, K.A.R., McKeever, S.W.S., 1978. The dose-rate dependence of thermoluminescence and sensitivity in quartz. Eur. PACT J. 2, 200–210.

Hsu, C., Weng, P.S., 1980. Reaffirmation on the low exposure rate dependence of the $CaSO_4$: Dy thermoluminescent dosimeter. Nucl. Instr. and Meth. 174, 73–76.

Huntley, D.J., Godfrey-Smith, D.I., Thewalt, M.L.W., 1985. Optical dating of sediments. Nature 313, 105–107.

Jaek, I.V., Savikhin, F.A., Käämbre, H., 1974. Proceedings of the Fourth Conference Lumin. Dosim. Krakow, pp. 565–573.

Kristianpoller, N., Chen, R., Israeli, M., 1974. Dose dependence of thermoluminescence peaks. J. Phys. D 7, 1063–1072.

Kvasnička, J., 1983. TL response dependence on the dose-rate and its consequences. Int. Appl. Radiat. Isot. 38, 713–715.

McKeever, S.W.S., Chen, R., Groom, P.J., Durrani, S.A., 1980. Dose-rate dependence of thermoluminescence response. Nucl. Instr. and Meth. 175, 43–44.

McKeever, S.W.S., Akselrod, M.S., 1999. Radiation dosimetry using pulsed optically stimulated luminescence of $Al_2O_3:C$. Radiat. Prot. Dosim. 84, 317–320.

Roberts, R.G., Spooner, N.A., Questiaux, D.G., 1994. Palaeodose underestimates caused by extended-duration preheats in the optical dating of quartz. Radiat. Meas. 23, 647–653.

Rodine, E.T., Land, P.L., 1971. Electronic defect structure of single crystal ThO_2 by thermoluminescence. Phys. Rev. B 4, 2701–2714.

Valladas, G., Ferreira, J., 1980. On the dose-rate dependence of the thermoluminescence response of quartz. Nucl. Instr. and Meth. 175, 216–218.

Zimmerman, D.W., Cameron, J.R., 1968. Supralinearity of thermoluminescence of LiF vs. dose. In: McDougall, D.J. (Ed.), Thermoluminescence and Geological Materials. Proc. Adv. Res. Inst. Spoleto, Italy. Academic Press, London, pp. 485–491.