Dose dependence and dose-rate dependence of the optically stimulated luminescence signal

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The expected behavior of the dose dependence of the optically stimulated luminescence (OSL) signal has been studied using numerical simulation. A simple model of one trapping state and one kind of recombination center is presented, and the sequence of sets of simultaneous differential equations governing the processes during excitation, relaxation, and light exposure are numerically solved. With the choice of reasonable trapping and recombination parameters, it has been shown that a quadratic dose dependence of this effect results from the model when the irradiation stage starts with empty trapping states. This may explain reports in the literature of an initial supralinear dose dependence of OSL. It is also shown that within the same model, one can get an initial linear dose dependence of OSL if one starts with partly filled traps. Also has been studied the influence of dose rate on the measured OSL signal for a constant total dose, and some effect has been seen for a certain dose-rate range. The similarities and dissimilarities of OSL as compared to thermoluminescence with respect to these phenomena are discussed. [DOI: 10.1063/1.1330555]

I. INTRODUCTION

The phenomenon of optically stimulated luminescence (OSL) is the emission of light by a solid sample during its stimulation by a different wavelength light, and following a higher energy excitation. The OSL effect resembles thermoluminescence (TL), the effect observed following the excitation of a solid sample, usually by ionizing radiation during which time, energy is absorbed in the sample. In TL, the next step is heating the sample up which results in the emission of the stored energy in the form of light, measurable by an appropriate detector, e.g., a photomultiplier.

In most cases, TL was found to be linear or nearly linear with the dose of excitation. This helped a lot in the applications of TL, namely, in dosimetry of different kinds of irradiation as well as in the TL dating of archeological samples such as pottery. For a detailed explanation of TL and related phenomena see Chen and McKeever.[1] In a number of cases, however, TL intensity was found to be supralinear with the excitation dose, and sometimes very strong supralinearity was reported.[2] The explanation to the effect was given in terms of competition with radiationless centers during the heating stage,[3] the excitation stage,[4] or both.[5]

Some accounts on dose-rate effects in TL have been given in the literature. Valladas and Ferreira[6] reported on two spectral components of light emission from quartz, one increasing and the other decreasing with the dose rate where the total dose of excitation remained unchanged. Explanation of the dose-rate dependence of TL has been given by Chen *et al.*[7] who used a model with competition between traps or luminescence centers to explain the phenomenon.

In recent years, OSL has started to replace TL in some of these applications in dosimetry and dating. This started with a work by Huntley *et al.*[8] on the optical dating of sediments; the method has also been utilized more recently for archeological dating and for dosimetry. The advantages of OSL over TL are rather obvious in the applications. There is no need to heat the sample, thus avoiding the blackbody radiation occurring at relatively high temperatures. Also, possible thermal quenching of luminescence is avoided. Finally, for dosimetry, the use of plastic materials that cannot be heated to high temperature can be considered for OSL dosimetry.

In nearly all the reports on OSL and its applications it is assumed, and sometimes shown,[9] that the initial dose dependence at low doses is linear followed by an approach to saturation at high doses.[10] 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 archeological dose imparted at a much lower rate.

There are a few reports in the literature on supralinear dose dependence of OSL. In the study of OSL of quartz and mixed feldspars from sediments, Godfrey-Smith[11] found linear dependence on the dose of the unheated samples. However, following a preheat at 225 °C, the samples showed a clear supralinearity of the OSL signal at low excitation doses of γ irradiation. Roberts *et al.*[12] have also found supralinearity of quartz OSL in several samples. For samples preheated at 160 °C, they reported a quadratic equation describing the dose dependence. There is no mention in the literature of a dose-rate effect of OSL.

In the present work, we deal with the problems of dose dependence and dose-rate dependence of OSL theoretically, by solving numerically the sets of simultaneous differential rate equations, which are believed to govern the processes involved. The approach is similar to that by McKeever *et al.*[13] except that they mainly followed the shape of the

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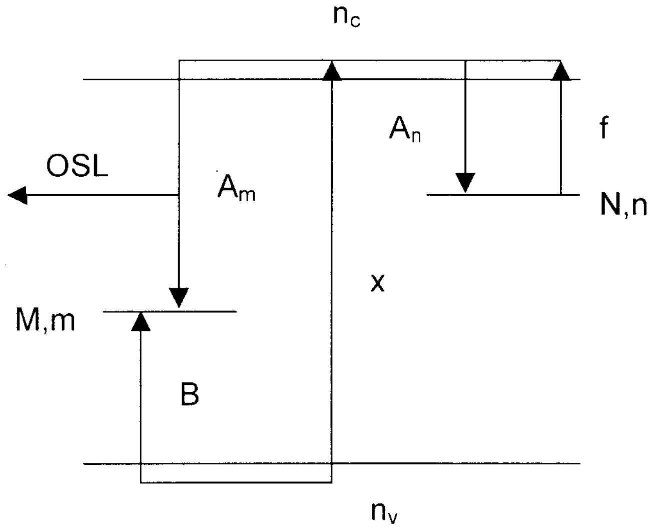

FIG. 1. Energy levels involved in the OSL phenomenon. A trapping state with a concentration N (m^{-3}) and instantaneous occupancy n (m^{-3}) as well as a recombination center M (m^{-3}) with instantaneous occupancy of m (m^{-3}) are shown. B ($m^3\,s^{-1}$) is the probability coefficient for holes in the valence band to be trapped in M. The retrapping and recombination coefficients of electrons respectively are A_n ($m^3\,s^{-1}$) and A_m ($m^3\,s^{-1}$). x ($m^{-3}\,s^{-1}$) is the intensity (dose rate) of radiation excitation of electrons from the valence to the conduction band. f (s^{-1}) is the intensity of the stimulating light. n_v (m^{-3}) and n_c (m^{-3}) are the instantaneous concentrations of free holes and electrons, respectively.

OSL decay curve with time whereas we are interested in the dose and dose-rate dependencies. Also, they include shallow traps that may cause temperature dependence of the OSL signal whereas we exclude this possibility in the present work.

II. THE MODEL

The energy level scheme shown in Fig. 1 is a simple one trap-one center model. One trapping state with a concentration of N (m^{-3}) capable of trapping electrons is assumed to be active as well as one hole center state with a concentration of M (m^{-3}). Electrons are assumed to be excited by the irradiation, the intensity of which is denoted by x($m^{-3}\,s^{-1}$). The total dose imparted during a period of time t_D (s) is given in these units as $D = xt_D$ (m^{-3}) (regarding the units of dose and dose rate, see later). The irradiation produces holes in the valence band and electrons in the conduction band, the instantaneous concentrations of which are denoted by n_v (m^{-3}) and n_c (m^{-3}), respectively. The free electrons in the conduction band may be trapped in N with a probability coefficient A_n ($m^3\,s^{-1}$) whereas the free holes from the valence band can be trapped in the centers with a probability coefficient B ($m^3\,s^{-1}$). The instantaneous filling of electrons in traps and holes in centers during the excitation (and later, during the optical stimulation as well) are denoted by n and m, respectively. After the excitation is finished, a relaxation period is allowed for. This means that the excitation intensity is set to zero, $x = 0$, and the carriers remaining in the conduction and valence bands relax to the trapping states and centers, respectively. It is to be noted that once holes are accumulated in the center, recombination may take place between these trapped holes and electrons in the conduction band with a recombination coefficient of A_m ($m^3\,s^{-1}$).

It is to be noted here that using the terms ''trapping probability'' or ''recombination probability'' for A_n, B, and A_m (with units of $m^3\,s^{-1}$), although rather common in TL theory, is not very accurate. In fact, what is meant is that products like $A_m m$ or $A_n(N-n)$, having units of s^{-1}, are probabilities per second. Another way to look at these coefficients is stating that, say, $A_m = \sigma_m v$ where σ_m is the cross section for recombination (in m^2) and v(m/s) is the thermal velocity of the relevant free carriers. We have therefore used here the terms ''probability coefficient'' or ''recombination coefficient'' and ''trapping coefficient.''

At the next stage of optical stimulation, we assume that the applied light releases electrons from the trapping state at the rate of fn($m^{-3}\,s^{-1}$), where the light intensity is represented by f(s^{-1}). The released electrons may retrap in empty trapping states with the same coefficient of trapping mentioned earlier for the excitation phase, namely, A_n ($m^3\,s^{-1}$), but they can, of course, also recombine with trapped holes with the mentioned recombination coefficient A_m. The OSL signal is assumed to result from this recombination as shown in Fig. 1.

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

$$dn_v/dt = x - B(M-m)n_v. \tag{1}$$

$$dm/dt = -A_m m n_c + B(M-m)n_v, \tag{2}$$

$$dn/dt = A_n(N-n)n_c, \tag{3}$$

$$dn_c/dt = dm/dt + dn_v/dt - dn/dt. \tag{4}$$

As pointed out earlier, the following stage of relaxation is simulated by taking the final values of n, m, n_c, and n_v at the end of the excitation stage as initial values for the relaxation stage; setting x to zero and solving the 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 the relaxation as initial values, keep $x = 0$ and add the term associated with the optical stimulation. The set of equations to be solved now is

$$-dm/dt = A_m m n_c, \tag{5}$$

$$dn/dt = -fn + A_n(N-n)n_c, \tag{6}$$

$$dn_c/dt = dm/dt - dn/dt. \tag{7}$$

Since, as stated earlier, we associate the intensity of the OSL signal with the recombination rate, we can write the OSL, intensity I as

$$I = -dm/dt. \tag{8}$$

In fact, a proportionality factor should have preceded the right-hand side of Eq. (8), however, omitting it merely means that we measure the emission intensity in some different units. It is to be mentioned at this point that usually, the OSL intensity is sampled with a stimulating light pulse of, say, 1 s or less. This has been simulated here by solving the set of Eqs. (5)–(8) for 1 s and recording the final value of $I=-dm/dt$ as the OSL signal. This bypasses the question of what is happening during the first fraction of a second when the stimulating light is turned on. It also disregards the time dependence of the OSL during a long period of light stimulation, during which the emitted light intensity may be decaying with time when n and/or m are depleted.

A note should be made here concerning the meaning of x and f and the reason why they do not have the same dimensions in the given sets of rate equations. The dose rate and total dose are given here in units of $m^{-3}\,s^{-1}$ and m^{-3}, respectively. These are, in fact, the rate of electron-hole pair production and the total concentration of electrons and holes produced, respectively. Let us consider the ranges of dose rates and doses in $Gy\,s^{-1}$ and Gy, respectively, associated with the x values chosen. Let us take as an example LiF for which, according to Avila *et al.*,[14] an average of 36 eV is required for heavy charged particles to produce an electron-hole pair and ~34 eV is required for γ rays (this is about three times the width of the band gap). LiF has a specific gravity of 2.6 or a density of 2600 kg/m^3. Since 1 Gy equals 1 J/kg, and 1 J equals 6×10^{18} eV, the number of pairs produced per kg is about 1.7×10^{17}. Therefore, the number of pairs produced in m^3 is 4.4×10^{20}. Thus, for example, $x=10^{17}\,m^{-3}\,s^{-1}$ (see e.g., the second point from the left in Fig. 3 later) is equivalent to 2.3×10^{-4} Gy/s whereas $x=10^{20}\,m^{-3}\,s^{-1}$ is equivalent to 0.23 Gy/s.

We should also briefly discuss the dimensions of x and f. Basically, since both represent irradiation intensity, they should have the same dimensions. A closer look (see the sets of equations earlier) shows however, that the equivalent of x in Eq. (1) is fn in Eq. (6), both having units of $m^{-3}\,s^{-1}$. However, whereas x is constant, fn varies when n varies, whereas f is constant. A slightly different way to present this is to write $x=N\cdot\mathbf{x}$ where N is the (constant) concentration of valence band electrons and $\mathbf{x}$ is a coefficient proportional to the light intensity with units of s^{-1}. Had we done it this way, f and $\mathbf{x}$ would have had the same dimension of s^{-1}.

The same set of equations was solved in the same sequence for studying the dose-rate effect. Here, the excitation intensity and the length of excitation were changed inversely, so as to keep a constant excitation dose. The details are given in Sec. IV.

III. NUMERICAL RESULTS OF OSL DOSE DEPENDENCE

It has been quite easy to find sets of trapping parameters within this basic energy level model that yielded quadratic dependence on the dose of excitation while starting with empty traps and centers. As an example, we give the following set: $A_m=10^{-17}\,m^3\,s^{-1}$; $B=10^{-18}\,m^3\,s^{-1}$; $N=10^{17}\,m^{-3}$; $M=10^{19}\,m^{-3}$, 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 the set of 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 for relaxation, and finally Eqs. (5)–(7) with the given value of f. The (+) points in Fig. 2 show, on a log–log scale, the dependence of the simulated

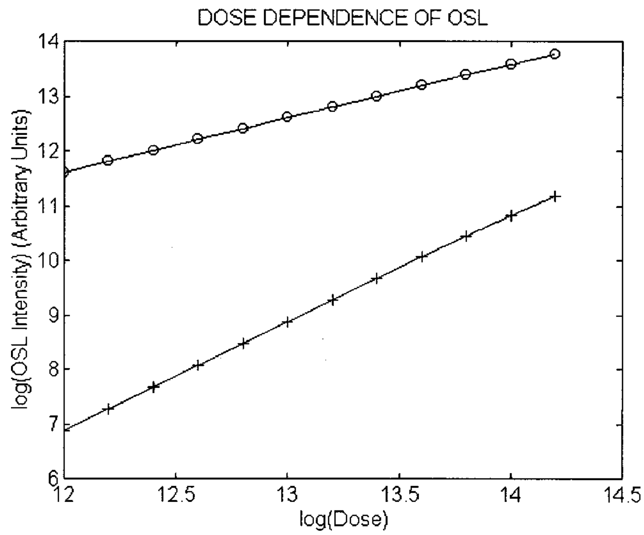

FIG. 2. Dose dependence of OSL on a log–log scale as calculated using the model; the parameters utilized are $A_m=10^{-17}\,m^3\,s^{-1}$; $B=10^{-18}\,m^3\,s^{-1}$; $N=10^{17}\,m^{-3}$; $M=10^{19}\,m^{-3}$; $A_n=10^{-19}\,m^3\,s^{-1}$; $f=1\,s^{-1}$, and x varied between 10^{12} and $10^{14}\,m^{-3}\,s^{-1}$. The (+) symbols are used in the case where $n(0)=0$ at the beginning of the excitation whereas the (○) symbols are used for the case with $n(0)=0.9N$. Note that the (logarithmic) scales are not the same, and therefore, the straight line formed by the circles has, in fact, a slope of 1, indicating linear dependence of the emitted intensity on the excitation dose. The straight line formed by the (+) points has a slope of 2, indicating a quadratic dependence of the emitted light intensity on the excitation dose.

OSL signal on the total dose, changed here by changing the intensity of excitation x in the mentioned range and keeping the length of excitation time at 1 s. Note that the x and y (logarithmic) scales are significantly different, and the straight line seen has, in fact, a slope of 2 meaning a quadratic dose dependence. It is to be noted that very similar results are found if the same parameters and the same initial conditions of empty traps and centers are maintained, but the excitation dose is varied by changing the excitation time and keeping the excitation intensity constant.

The results so far, an example of which just given, suggest that when one is dealing with empty traps and centers at the outset, usually associated with properly annealed samples, the quadratic dose dependence comes out as a natural result. The question may arise why most works assumed (sometimes implicitly) that the OSL signal is linear with the dose. It is obvious that once we increase the applied dose further than, say, what was shown in Fig. 2, an approach to saturation will occur and, on the way, there will be a range, broad or narrow depending on the parameters, of approximately linear dependence. An alternative which we pursue in the present work is assuming that one of the trapping states involved, either the electron trap or the hole center, is nearly full of carriers to begin with. From the theoretical point of view, if the quadratic behavior is associated with the product of the concentrations of electrons in traps and holes in centers, each of which being filled linearly, having one of these practically constant should bring about a linear dependence of the OSL intensity on the dose. From the experimental results, the fact that supralinearity occurs at annealed samples whereas a linear dose dependence is observed in ''as is'' samples,[11] points to the same direction. In order to simulate this situation we have taken $n_0 = 9\times10^{16}\,\mathrm{m}^{-3}$ which means that 90% of the traps are full with electrons at the beginning. We keep all the parameters the same, and repeat the same simulated cycles of excitation, relaxation and optical stimulation. The results are shown as the points given as circles (○) in Fig. 2. Note again that the x and y logarithmic scales are not the same and, in fact, the straight line seen here has a slope of unity. This, of course, indicates that the dose dependence under these circumstances is linear. It is to be noted that the intensities of the points in the latter case of linear dependence are significantly higher (3–5 orders of magnitude) than those calculated with the same parameters and doses when the trapping states start empty.

IV. DOSE-RATE DEPENDENCE OF OSL

As mentioned in the introduction, dose-rate effects of TL have been reported in the literature (e.g., Valladas and Ferreira).[6] No such reports have been given of OSL results. It should be pointed out, however, that for such complex processes taking place during the excitation and optical stimulation, there is no reason *a priori* to assume that only the total dose is the determining factor. We therefore tried to check whether within the framework of the present model, dose-rate effect could be seen. We have taken, as an example, the following set of parameters: $A_n = 3\times10^{-17}\,\mathrm{m}^3\,\mathrm{s}^{-1}$, $N = 10^{18}\,\mathrm{m}^{-3}$, $M = 10^{19}\,\mathrm{m}^{-3}$, $A_m = 10^{-17}\,\mathrm{m}^3\,\mathrm{s}^{-1}$, and $B = 10^{-17}\,\mathrm{m}^3\,\mathrm{s}^{-1}$. The dose-rate x was changed from 10^{16} to $10^{23}\,\mathrm{m}^{-3}\,\mathrm{s}^{-1}$ whereas the excitation time was changed inversely, from 10^2 to 10^{-5} s, so as to keep a constant dose. The stimulating intensity has been set at $f = 10^{-5}\,\mathrm{s}^{-1}$ and the stimulation time was taken as 1 s. The same sets of coupled differential equations have been solved numerically by the same ode23 solver and in the same order as described before for the dose dependence case. The results are shown as the circled points in Fig. 3, on a semilog scale. The OSL simulated intensity is seen to decrease by ~20%, while varying the dose rate from 10^{17} to $10^{20}\,\mathrm{m}^{-3}\,\mathrm{s}^{-1}$.

We also tried to look for sets of parameters for which the increase of the dose rate will bring about an increase in the OSL readings. The motivation has been that in those cases in which TL was found to be dose-rate dependent, both an increase and a decrease of the emitted intensity with the dose rate for a constant total dose have been seen. It turned out that by changing just one of the parameters in the set given earlier, namely, if B was taken to be $10^{-18}\,\mathrm{m}^3\,\mathrm{s}^{-1}$ rather than $10^{-17}\,\mathrm{m}^3\,\mathrm{s}^{-1}$, such an increase was seen. The sets of simultaneous differential equations have also been solved in the same way as before, and the results are shown by the (+) symbols in Fig. 3. In the same range of x varying between 10^{17} and $10^{20}\,\mathrm{m}^{-3}\,\mathrm{s}^{-1}$, the resulting OSL increased by ~19%. It should be mentioned here that the results shown in Fig. 3 are related to a situation in which initially, the trapping states and recombination centers are empty. This is associated with the supralinear dose dependence. The simulation was repeated when the initial filling of the trap was 0.9N,

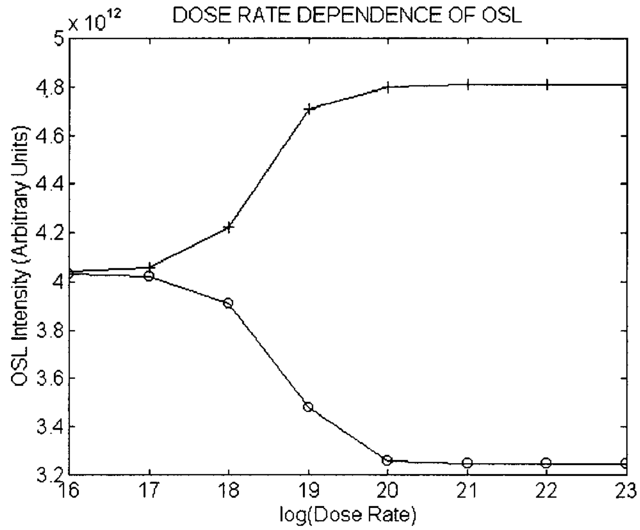

FIG. 3. Calculated dose-rate dependence of the emitted OSL signal with the total dose kept constant. The relevant parameters are $A_n = 3\times10^{-17}\,\mathrm{m}^3\,\mathrm{s}^{-1}$; $N = 10^{18}\,\mathrm{m}^{-3}$; $M = 10^{19}\,\mathrm{m}^{-3}$; $A_m = 10^{-17}\,\mathrm{m}^3\,\mathrm{s}^{-1}$; $B = 10^{-17}\,\mathrm{m}^3\,\mathrm{s}^{-1}$. x was changed from 10^{16} to $10^{23}\,\mathrm{m}^{-3}\,\mathrm{s}^{-1}$ and t_D was varied inversely, from 10^2 to 10^{-5} s. As is shown, in a range of 3–4 orders of magnitude in the dose rate of excitation, the intensity goes down by ~20% with one set of parameters, and goes up by ~19% with another.

which brought about linearity in the dose dependence. In this situation, practically no dose-rate dependence was seen in the simulated results. It should be emphasized that no general statement can be made concerning this dose-rate independence. It is possible that with other sets of parameters, dose dependence will be linear whereas still some dose-rate effect can take place.

V. DISCUSSION

Following some reports in the literature of a supralinearity of the dose dependence of OSL signals, and due to the general resemblance between OSL and TL, a theoretical study has been conducted to follow the possible dose dependence and the dose-rate dependence of this effect. A simple energy level model of a single trapping state and a single kind of recombination center has been studied. It turned out to be rather easy to find reasonable sets of the trapping parameters that result in supralinear (quadratic) dose dependence or with dose-rate dependence. These results are to be considered in view of the fact that OSL is utilized for archeological and geological dating of samples, as well as in dosimetry. The question of whether the dose dependence is linear in a given sample is of great importance for the applications. The same is true for the calibration of the OSL signal by applying a laboratory irradiation in order to compare the measured intensities to those resulting from the dose received in antiquity. If, for example, a variation of ~20% occurs in materials to be dated with the dose rate changing by 3–4 orders of magnitude, as found in the simulation, this has a significant bearing on the conclusions concerning the age found by OSL.

As pointed out before, supralinearity as well as dose-rate effects were found before in TL measurements. The explanations were usually related to the existence of competitors that play an important part in the excitation stage, the readout stage or both. In the present case of OSL, it appears that these effects are prone to occur even more generally since both effects can be found in the simulation while starting with the simple model of only one electron trapping state and one kind of recombination center.

This is the place to point out a basic difference between the processes of TL and OSL. In TL, one usually measures the area under an extended curve which is normally dependent on $\min(n_0,m_0)$ where n_0 and m_0 are the concentrations of traps and centers respectively at the beginning of the heating stage (see, e.g., Chen and McKeever).[1] Here, if both n_0 and m_0 are linearly dependent on the dose, so is the TL signal. In a first order peak, different doses produce different intensities of the peak, but its shape remains unchanged. In a second order peak, however, although the total area of the peak is proportional to the dose, increasing the dose distorts the peak to some extent and causes its maximum point to shift to lower temperatures. Moreover, if one checks the dose dependence of a certain temperature point in the initial-rise region of a given second-order peak, a quadratic dose dependence is found.[15] The OSL process of illuminating the previously irradiated sample for, say, 1 s seems to resemble such sampling of a TL curve at the initial rise range.

Another point of importance is that in order to get a ''pure'' first order behavior, one has to assume no retrapping at all $(A_n=0)$. However, if this extreme situation takes place, neither TL nor OSL can be observed since during the excitation stage, no electrons can be trapped in the trapping states. Since this is obviously not the case in the measurable circumstances, some features of the second-order behavior must exist in any TL and OSL signal. Therefore, it is possible that the supralinearity and dose-rate effect seen in the present simulation have to do with the kinetics not being of pure first-order kinetics.

In TL, in order to demonstrate supralinear dose dependence and dose-rate dependence, one has to assume a competitor that may take part during the excitation stage, the heating stage or both.[1] Such competition may bring about a supralinearity in the filling of the trap or the center but, which is more important to our present case, it may also cause the signal to be proportional to $n_0 m_0$ rather than to $\min(n_0,m_0)$. The main question that may arise here is what can replace the competitor that is not assumed here to exist. It is possible that the partly empty trapping state can take the role of the competitor as well. An indication that this may be the case is that once we start with a nearly full trapping state (90% in the example given), the dose dependence is practically linear, and no dose-rate effect is seen in the simulated results.

It seems to us that in view of the present theoretical results, more experimental effort should be made to see whether supralinearity of OSL at low doses is a more common effect than thought so far, and if dose-rate effects of OSL indeed occur.

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