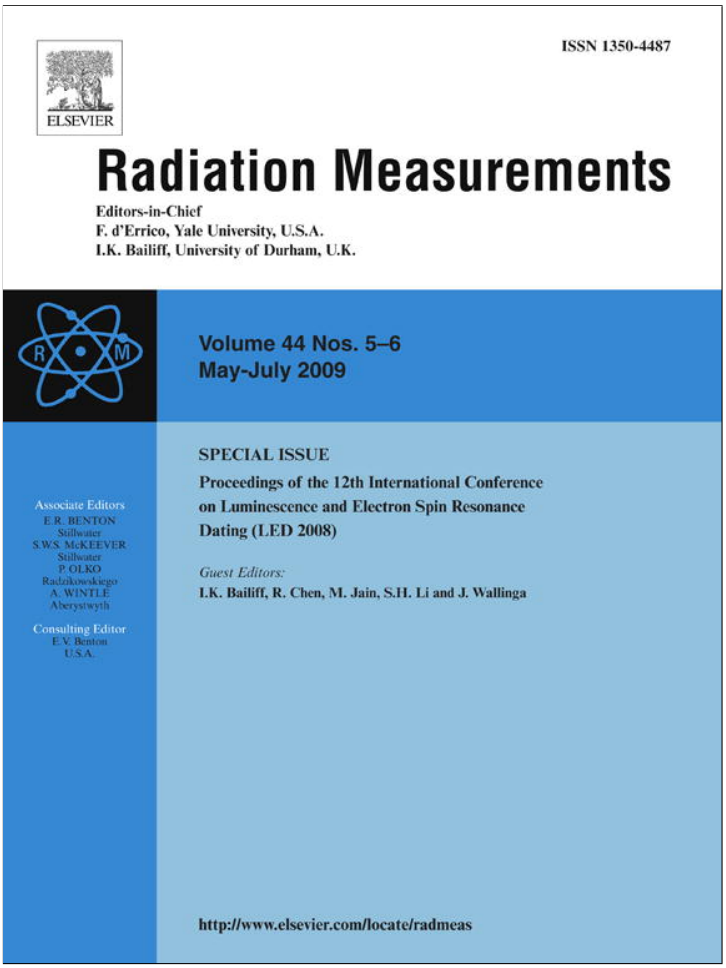

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Sublinear dose dependence of thermoluminescence and optically stimulated luminescence prior to the approach to saturation level

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ABSTRACT

In the use of thermoluminescence (TL) and optically stimulated luminescence (OSL) for dosimetry and for geological and archaeological dating, the nature of the dose dependence of the luminescence signal is of great importance. Non-linear dependence has been shown to result either from non-linear filling of the relevant traps or recombination centers during excitation, or by a combined effect of the linear filling of traps and centers due to processes taking place during the read-out stage. Sublinearity, which had been found in several materials, was usually attributed to saturation effects during excitation of either the relevant traps or centers. Sometimes, the competition effects during the excitation between traps result in superlinearity of some TL peaks and sublinearity of others. In the present work, we show that sublinear dose dependence may take place even in the simplest possible case of one trap–one recombination center (OTOR), even when the traps and centers are far from saturation. Analytical derivations as well as simulations consisting of the numerical solution of the relevant sets of coupled differential equations show the occurrence of the sublinear dose dependence under these circumstances. The filling of the traps is shown to behave like $D^{1/2}$ where D is the excitation dose, for an appropriate choice of the trapping parameters. This, in turn, may result in a similar dose dependence of the TL and OSL signals.

1. Introduction

Non-linear dose dependence of thermoluminescence (TL) and optically stimulated luminescence (OSL) has been reported in most luminescent materials. The most common behavior of this sort is the exponential approach to saturation at high doses which takes place in practically all materials. This usually means that the available traps or recombination centers are close to being filled to capacity, and further irradiation cannot increase the number of trapped carriers, hence the subsequently measured luminescence (be it TL or OSL) reaches saturation. Superlinear dose dependence of TL, namely, a range of doses in which the slope of the TL intensity vs. excitation dose is increasing or, in other words, that the second derivative of the signal with respect to the dose, d^2S/dD^2 is positive (see Chen and McKeever, 1994), was found in a number of materials. These include LiF (see, e.g., Cameron et al., 1964; Suntharalingam and Cameron, 1969; Horowitz, 1990), and quartz (see, e.g., Chen et al., 1988).

Cases of sublinear dose dependence of TL and OSL at relatively low doses, namely, ranges where $d^2S/dD^2 < 0$, have also been reported. Rodine and Land (1971) reported on the dose dependence of TL in ThO_2. Different peaks showed different dependencies on the dose, some were superlinear whereas others were sublinear. These authors explained the results in terms of competition over carriers during the excitation and heating stages. Boustead and Charlesby (1970) studied TL in squalane and reported superlinearity in some peaks and sublinearity in others. Goldstein (1966, 1967), Durand et al. (1969), Farge (1969) and Israeli et al. (1972) studied the TL resulting from the production of point defects in UV-irradiated KI, LiF and other alkali-halides. The situation here is different from the mentioned case of filling of existing traps, however, a close analogy can be found between the sets of simultaneous differential equations governing the two processes. These authors showed that a $D^{1/2}$ dependence can be expected under certain conditions.

A material in which sublinearity was found as of very low doses is $CaSO_4$:Dy. Caldas and Mayhugh (1976) reported on photo-stimulated TL (also termed photo-transferred TL; PTTL) which goes like $D^{0.55}$ where the dose varies by 5 orders of magnitude. Lakshmanan et al. (1981) presented results of sublinear γ-ray dose response of

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the high temperature peaks in $CaSO_4$:Dy. Pagonis et al. (2008) discuss a previously suggested model by Bailey (2001), which includes several traps and recombination centers for TL and OSL that explains the sublinear dose dependence of OSL and thermally stimulated OSL (TT-OSL) in quartz.

In the present work, we concentrate on the simplest model of one trap–one recombination center (OTOR) and show, both analytically and by numerical simulation, that the filling of traps far from saturation may be sublinear, and that under certain conditions, it may go like $D^{1/2}$. The resulting TL or OSL may depend in a similar way on the dose.

2. The model

The model used includes one trap and one kind of recombination center, which is the minimum required for explanation of TL and OSL. This model is shown in Fig. 1. Electrons are raised by the irradiation at a rate X ($cm^{-3}\,s^{-1}$) from the valence band, leaving free holes behind, with the instantaneous concentration n_v (cm^{-3}). In this simple model, the holes can be trapped in the centers only, with a probability coefficient B ($cm^3\,s^{-1}$). The instantaneous concentration of electrons in the conduction band is denoted by n_c (cm^{-3}). A_n ($cm^3\,s^{-1}$) is the retrapping probability coefficient and A_m ($cm^3\,s^{-1}$) the recombination probability coefficient. Xt (cm^{-3}) is the total concentration of produced electrons and holes, which is proportional to the absorbed dose, and in the present work we consider it as being the dose.

The coupled simultaneous rate equations governing the process of excitation is

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

$$\frac{dn_c}{dt} = X - A_n(N-n)n_c - A_m m n_c, \tag{2}$$

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

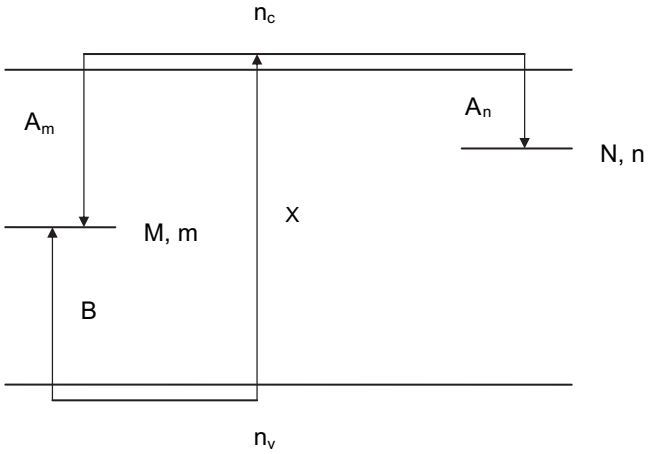

Fig. 1. Energy level scheme of the one trap–one recombination center (OTOR) model. N and M (cm^{-3}) are the concentrations of the electron traps and hole recombination centers, and n and m (cm^{-3}) their occupancies, respectively. X ($cm^{-3}\,s^{-1}$) is the rate of production of free electrons and 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.

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

Assuming physically realistic rate constants and not exceedingly large dose rates, we can make the usual quasi-steady assumption $dn_c/dt \approx dn_v/dt \approx 0$ which results in

$$n_v = \frac{X}{B(M-m)} \tag{5}$$

and

$$n_c = \frac{X}{A_n(N-n) + A_m m}. \tag{6}$$

Substituting Eqs. (5) and (6) into Eq. (1) yields

$$\frac{dm}{dt} = \frac{dn}{dt} = \frac{A_n(N-n)}{A_n(N-n) + A_m m}X. \tag{7}$$

Under the present assumptions of one trap–one recombination center, and assuming that n_c and n_v are significantly smaller than n and m, we have

$$m = n. \tag{8}$$

With this, Eq. (7) can be rewritten as

$$\frac{dn}{dt} = \frac{A_n(N-n)}{A_n(N-n) + A_m n}X, \tag{9}$$

which can be rearranged

$$X\,dt = \frac{A_n(N-n) + A_m n}{A_n(N-n)}\,dn = \left(1 + \frac{A_m n}{A_n(N-n)}\right)dn, \tag{10}$$

and integrated

$$Xt = n + \frac{A_m}{A_n}\left[-N\ln\left(1-\frac{n}{N}\right) - n\right]. \tag{11}$$

For a small argument, $x \ll 1$, one can use the first two terms in the relevant series expansion,

$$\ln(1-x) = -x - \frac{1}{2}x^2, \tag{12}$$

which yields

$$Xt = n + \frac{A_m}{2A_n}\frac{n^2}{N}, \tag{13}$$

which, in turn, results in

$$A_n N n + A_m n^2/2 = A_n N X t. \tag{14}$$

Expression (14) is a quadratic equation in n which actually gives a relation between n and t (or the dose $D = Xt$). The solution for n yields

$$n = \frac{-A_n N \pm \sqrt{(A_n N)^2 + 2A_m A_n N X t}}{A_m}. \tag{15}$$

Obviously, in order to get positive value for n, we have to choose the plus sign, which results in

$$n = \frac{A_n N}{A_m}\left(\sqrt{1 + \frac{2A_m}{A_n N}Xt} - 1\right). \tag{16}$$

If following the excitation we allow for relaxation, i.e., that the free electrons and holes get either recombined or trapped following

the excitation, we end up with $n_c = n_v = 0$. We will also choose the parameters in such a way that n_c and n_v are much smaller than n and m, respectively, and therefore, the additional contribution to n and m during relaxation is relatively small. Equation (16) can be considered as a good approximation to the dependence of n on t or on D under the mentioned assumptions. The square-root sign shows that the expression is sublinear.

We can consider two limiting cases:

1. Small doses
   For $Xt \ll A_n N/(2A_m)$, using binomial series expansion, Eq. (16) simplifies to

$$n \approx Xt. \tag{17}$$

   This means that, during excitation and for small doses, the trap captures the available free electrons with 100% efficiency.

2. Large doses
   For $Xt \gg A_n N/(2A_m)$, Eq. (16) simplifies to

$$n \approx \sqrt{\frac{2A_n N}{A_m} Xt}. \tag{18}$$

   This indicates that as doses grow larger, the trap captures an ever smaller fraction of the available free electrons. This is because m grows with dose and, as it grows, the recombination rate $A_m m n_c$ becomes stronger relative to the rate for capture by traps, $A_n N n_c$. This explains the sublinearity of n.

   In the section below we will show how results of the numerical simulation compare with Eqs. (16) and (18) with certain sets of parameters.

3. Numerical results

In order to show the dose dependence of the trapped electrons and holes, we chose sets of trapping parameters, and solved numerically Eqs. (1)–(4) using the Matlab ode23s solver for a certain period of time t. The simulated excitation dose was therefore proportional to $D = Xt$. In order to simulate the experimental procedure properly, the solution was continued for a further period of time with $X = 0$, so that free electrons and holes relax into traps and centers, respectively. It should be noted that since the materials in question are insulators, even under irradiation, the concentrations of free electrons and holes are expected to be only a small fraction of the concentrations of trapped electrons and holes. We have monitored the simulated concentrations of trapped and free carriers, and if the latter were not significantly smaller than the former for a certain choice of parameters, we increased the values of the recombination and trapping probability coefficients A_m, A_n and B. The condition for the free carriers concentrations being small was somewhat hard to achieve in the interesting range of high dose rates X, but increasing the probability coefficients made it possible to get reliable results in this sense. The results of the simulations are compared to those of Eqs. (16) and (18) so that the validity of the approximations made while developing these equations can be checked.

Fig. 2 shows the simulation results of the time of excitation dependence of the trapped carriers for the set of parameters given in the caption, for times of excitation between 2 and 100 s, for values of X varying by order of magnitude from one curve to the next, between 10^4 and 10^8 cm^{-3} s^{-1}. For each value of the dose rate X, the change of irradiation time, t, means, in fact, changing the dose. With the mentioned times of excitation, the dose varies from $2X$ to $100X$. A short discussion on the choice of the recombination and retrapping probability coefficients is given in the Appendix. In order to accommodate the results for values of X varying by an order of magnitude from one curve to the next in the same figure, the numerical values have been multiplied by the factors shown on the upper left-hand side of the figure. It is readily seen that the lines for low values of X are close to be straight in agreement with Eq. (17). As X grows, the sublinear property is exhibited. The (+) signs show the values found by Eq. (16), and the agreement with curve (e) is very good. The (×) signs indicate the results of Eq. (18) and these results do not agree very well with those of the simulation. The reason is that the values of Xt are not large enough for the approximation to be good. In order to check the approximation of square-root dependence on the dose, larger values of X should be used. However, as pointed out above, this brings about the occurrence of relatively large values of n_c and n_v. Another set of trapping parameters has been chosen in which the values of the recombination and trapping probability coefficients are larger.

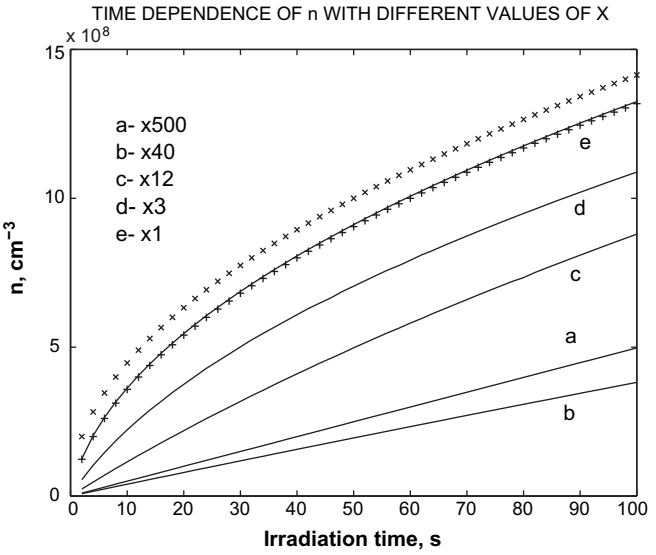

Fig. 2. Results of the dependence of the concentration of trapped electrons following excitation and relaxation on the time of excitation. The parameters chosen are $A_m = 10^{-7}$ cm^3 s^{-1}; $A_n = 10^{-13}$ cm^3 s^{-1}; $B = 10^{-14}$ cm^3 s^{-1}; $N = 10^{14}$ cm^{-3}; $M = 10^{15}$ cm^{-3}. The dose rate X is 10^4, 10^5, 10^6, 10^7, and 10^8 cm^{-3} s^{-1} in curves (a), (b), (c), (d) and (e), respectively; the results of the simulations are shown by the solid lines. The results of curves (a–d) have been multiplied by the factors shown on the upper left-hand side. For $X = 10^8$ cm^{-3} s^{-1}, the results of the two approximations are also shown (see text).

Fig. 3 depicts the results of the concentration of electrons in traps, n, as a function of the excitation dose. The value of the intensity, X, is high enough so that, along with the other chosen parameters (given in the caption), the condition for Eq. (18) to be a good approximation is fulfilled. Note that the recombination and retrapping probability coefficients are relatively large so that n_c and n_v are significantly smaller than n and m even for the high value of X used. The solid line shows the results of the simulation whereas (+) indicates the results of Eq. (16) with the same parameters and (×) the results of Eq. (18). Here, the agreement between the three is excellent, showing that the concentration depends on the dose like $D^{1/2}$.

An important point to consider is whether the effect mentioned is of sublinear dose dependence or a dose-rate effect. Eqs. (11) and (16) and their approximations for small and large dose (17) and (18)

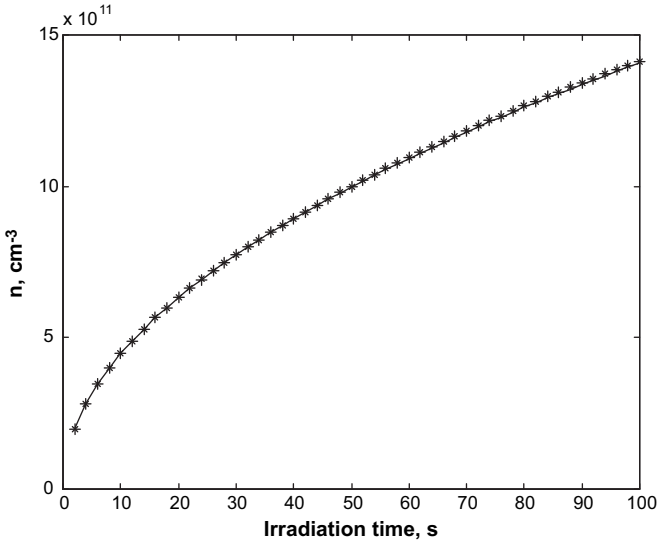

Fig. 3. Same as curve (e) in Fig. 2, but with $A_m = 10^{-5}$ cm^3 s^{-1}; $A_n = 10^{-11}$ cm^3 s^{-1}; $B = 10^{-10}$ cm^3 s^{-1}; $N = 10^{14}$ cm^{-3}; $M = 10^{15}$ cm^{-3} and $X = 10^{14}$ cm^{-3} s^{-1}. Here too, the (+) signs indicate the results of Eq. (16) and the (×) signs those of Eq. (18).

present the connection between the trap occupancy following excitation and of the total dose, and no distinction is made between changes of the dose by varying the dose rate or by the time of excitation. It should be noted, however, that these equations were developed using the quasi-equilibrium assumption. This may mean that some dose-rate effect may be expected at very high values of the dose rate X. In order to check the possibility that such effects may take place within the range of dose rates discussed here, we repeated the simulations by holding, for the same set of parameters, the excitation time constant and varying the dose rate. This was done for dose rate varying in the same range mentioned before, namely $X = 10^4$–10^8 cm^{-3} s^{-1}. The results (not shown here) reveal that within the range of doses and dose rates discussed, there is no noticeable dose-rate effect.

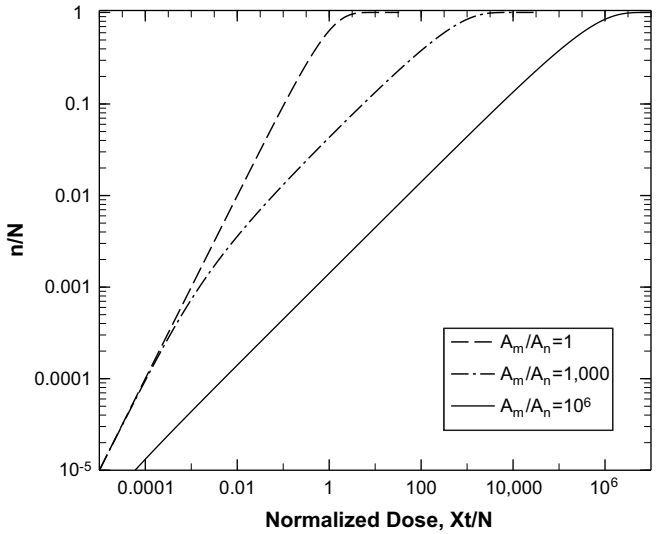

Fig. 4. Dose dependence of the trap occupancy calculated by solving Eq. (11) numerically for different values of the dose Xt. The solid line depicts the results for $A_m/A_n = 10^6$, the dashed–dotted line for $A_m/A_n = 10^3$ and the dashed line for $A_m/A_n = 1$. Note the different scale of the two axes.

One may be interested in the dose dependence all the way to true saturation. Fig. 4 gives an example based on the numerical solution of the implicit function given in Eq. (11). As shown in the figure, three dose dependencies are shown on a log–log scale of n/N as a function of the normalized dose Xt/N for different values of the relative recombination to retrapping probabilities. The solid line depicts the results for $A_m/A_n = 10^6$, the dashed–dotted line for $A_m/A_n = 10^3$ and the dashed line for $A_m/A_n = 1$. Note that the scales of the x- and y-axis are not the same and, in fact, the solid line starts with a slope of 0.5, meaning a dose dependence of $D^{1/2}$ nearly all the way to saturation. The other two curves start with a slope of unity, but the dashed one goes linear for a long way before approaching saturation whereas the dashed–dotted line changes from linearity to an approximate $D^{1/2}$ dependence before going to saturation (see Discussion below).

4. Discussion

In this work we have used numerical simulation based on the solution of the relevant set of simultaneous differential equations, as well as plausible approximations, and showed that the dose dependence of trapped carriers may be significantly sublinear for a one trap–one recombination center (OTOR) model, even when the traps are far from saturation. It is shown, both by plausible approximations and numerical simulation that at relatively low doses, the dose dependence tends to be linear and at higher doses it is sublinear. The reason for this sublinearity is competition between the trap and the recombination center for free electrons during irradiation. We have shown that with an appropriate choice of the parameters and large enough dose of excitation, a square-root dependence on the dose can be reached. To some extent, these results resemble those by Goldstein (1966, 1967), Durand et al. (1969), Farge (1969) and Israeli et al. (1972) mentioned above, although these authors dealt with the production of new F-centers rather than filling of existing traps. The similarity between the two situations takes place in spite of the fact that in the present case there is a limitation of the number of existing traps which is not present in the case of production of defects.

In this context, it is interesting to note the results shown in Fig. 4. As long as the condition leading to Eq. (11) is valid, namely, that the quasi-equilibrium condition holds, the figure shows the dose dependence all the way to saturation with different relative ratios of the recombination to retrapping probabilities. Taking into consideration the different scales of the x- and y-axis, the solid line with $A_m/A_n = 10^6$ has a slope of 0.5. Let us consider the cases of small and large doses, leading to Eqs. (17) and (18) respectively. The condition bringing about Eq. (18), $Xt \gg A_n N/(2A_m)$, is much easier to be fulfilled for this large ratio of 10^6, and one can expect a broader range of a $D^{1/2}$ dependence which, in fact is seen in the figure almost all the way to saturation. As for the opposite condition $Xt \ll A_n N/(2A_m)$, this can be fulfilled in a broad range of doses for a much smaller ratio of recombination to retrapping probabilities, and for the example given of $A_m/A_n = 1$ (dashed line in Fig. 4), a linear dependence on the dose is seen, in agreement with Eq. (17). In the intermediate case (dashed–dotted line in Fig. 4), with $A_m/A_n = 10^3$, the behavior is more complex. The dose dependence starts being linear, it then goes sublinear at a relatively low dose, far from saturation, remains approximately $\propto D^{1/2}$ for a few orders of magnitude change of the dose, and finally goes to saturation. In conclusion, a long region of $D^{1/2}$ occurs if the ratio A_m/A_n is large and becomes longer as this ratio increases. By contrast, if the ratio is not large, then no such region occurs regardless of the values chosen for N and M. Changing N has no effect on the data once it is normalized as in Fig. 4. It should be noted that the model requires $M > N$ in order to avoid the possibility of accumulation of electrons

in the conduction band with very large doses. As long as this condition is obeyed and quasi-equilibrium is valid, the value of M has no effect on the dose-scaling results at all.

Finally, it should be noted that the TL and OSL often depend linearly but sometimes non-linearly on the concentration following the irradiation. These dependencies should be combined with the presently discussed dependence of the carriers' concentration on the dose, to yield the total dependence of the luminescence intensities on the dose of excitation.

Appendix

We chose the largest electron recombination probability coefficient to be $A_m = 10^{-5}\ \mathrm{cm}^3\ \mathrm{s}^{-1}$. Values of this size are observed when a carrier, an electron in this case, and a trap or center have opposite charges. For such coulomb attractive combinations, experimentally measured probability coefficients are typically in the range 10^{-5}–$10^{-8}\ \mathrm{cm}^3\ \mathrm{s}^{-1}$ (Lax, 1960; McKeever, 1985). Smaller probability coefficients, typically 10^{-8}–$10^{-11}\ \mathrm{cm}^3\ \mathrm{s}^{-1}$, are observed for neutral traps (Lax, 1960; McKeever, 1985). Traps with the same charge sign as the free particles cause mutual repulsion and consequently have still smaller rate constants below $10^{-11}\ \mathrm{cm}^3\ \mathrm{s}^{-1}$, and could be as low as $10^{-15}\ \mathrm{cm}^3\ \mathrm{s}^{-1}$ (Lax, 1960). The values of B and A_n chosen for the simulations leading to Fig. 2 were in the range typical of the repulsive type and A_m is of the attractive type. As for the parameters taken for Fig. 3, A_m was again of the attractive type whereas B and A_n are consistent with the neutral type. Note that assuming that A_m is of the attractive type agrees with the fact that recombination takes place only after a hole has been captured in the center.

An additional test of the reasonableness of the assumed A_n value is provided by detailed balance which relates the capture rate constant A_n to the trap's pre-exponential factor s (see McKeever, 1985; Chen and McKeever, 1997),

$$\frac{s}{A_n} = \frac{\left(2\pi m_e^* kT\right)^{3/2}}{h^3}\frac{2g_0}{g_1}, \tag{A1}$$

where m_e^* is the effective mass of electrons in the conduction band, g_0 is the degeneracy of an empty trap, g_1 is the degeneracy of a filled trap, k is Boltzmann's constant and h is the Planck constant. A typical value for the right-hand side is about $10^{19}\ \mathrm{cm}^{-3}$. Thus by Eq. (A1), the value $A_n = 10^{-11}\ \mathrm{cm}^3\ \mathrm{s}^{-1}$ corresponds to a pre-exponential factor s in the neighborhood of $10^8\ \mathrm{s}^{-1}$ which is well within the range of s that is commonly observed in experiments, confirming the reasonableness of the chosen A_n (see Chen and McKeever, 1997).

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