



# Intrinsic superlinear dose dependence of thermoluminescence and optically stimulated luminescence at high excitation dose rates



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## HIGHLIGHTS

- Intrinsic superlinear dose dependence of TL and OSL with high excitation dose.
- Theoretical model with no competition, using the one trap one center (OTOR) model.
- Analytical expressions and numerical results.
- Expected dose-rate effect within the OTOR model.

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## ABSTRACT

Superlinear dose dependence of thermoluminescence (TL) and optically stimulated luminescence (OSL) has been reported for many materials. The theoretical explanation has been ascribed to competition of either traps or recombination centers, during the excitation stage or during the read-out phase. There has been an account in the literature on superlinearity of OSL associated with merely one trapping state and one kind of recombination center. This had to do with the process taking place during the read-out stage, namely the optical stimulation. In the present work, we report on a model of one trapping state and one kind of recombination center which results in a superlinear filling of the center. Thus, one can expect a superlinear dose dependence of the area under the resulting TL glow peak as well as the OSL signal. We follow this situation by writing the simultaneous nonlinear rate equations for the one-trap-one-recombination-center (OTOR) model and study the expected results by numerical simulation consisting of solving the equations with sets of the trapping parameters. We also present analytical results based on simplifying assumptions, and compare the analytical and numerical results. The effect is significant at relatively high dose rates. The main implication is that when one tries to evaluate by TL dosimetry a dose applied at a high rate, calibration of the TL dosimeter using much smaller dose rates may result in inaccurate results.

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## 1. Introduction

In many cases, thermoluminescence (TL) and optically stimulated luminescence (OSL) intensities have been found to be linear or nearly linear with the dose. This helped a lot in the applications of TL and OSL in dosimetry as well as in dating of archaeological and geological samples. In a number of cases, however, the TL intensity was found to be superlinear with the excitation dose, and sometimes, very strong

superlinearity was reported (see, e.g., [Chen et al., 1998](#)). Note that in the literature, the terms superlinearity and supralinearity are used to describe a dose dependence which is “more than linear”. [Chen and McKeever \(1994\)](#) have made a distinction between two different though related properties. One point of view has to do with the rate of change with dose of the dose dependence function. The authors term this property “superlinearity” which actually checks whether  $d^2S/dD^2$ , the second derivative of the measured signal with respect to the dose, is positive. The other approach is related more to the applications of TL in dosimetry and archaeological and geological dating, and basically has to do with the correction to be made in extrapolation in cases where “supralinearity” occurs following an

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initial linear dose range, or prior to such a linear range. These authors define two indices, the “superlinearity index”  $g(D)$  and the “supra-linearity index”  $f(D)$  which quantify these two properties.

The explanation to the super(supra)linear effect was given in terms of competition with traps or centers during the excitation stage (Chen and Bowman, 1978), the heating stage (Kristianpoller et al., 1974) or both (Chen and Fogel, 1993). Superlinearity of OSL has also been reported. Superlinear dose dependence has been reported by Godfrey-Smith (1994) who found the effect in a study of quartz and mixed feldspars from sediments following preheat at 225 °C. 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 describing the dose dependence. Schembri and Heijman (2007) reported on superlinear dose dependence of OSL in  $\text{Al}_2\text{O}_3\text{:C}$ . Chen and Leung (2001a) described the superlinearity of TL and OSL in terms of competition, both during excitation and read-out with a competing trap. Furthermore, Chen and Leung (2001b) explained the superlinearity of OSL using a model of one trapping state and one recombination center, namely, without any competitors. The effect could be demonstrated using numerical simulation when the response to short pulses was considered, and not the total area under an OSL decay curve. Also, the effect was seen when the initial occupancy of the relevant center was zero or close to zero, and the dependence of the pulsed OSL was closer to be linear with the dose if the center had considerable initial concentration of holes. Qualitatively, the effect was explained in terms of processes taking place during the read-out stage. These authors also studied the possible dose-rate dependence under the same condition. More details on the different kinds of super(supra)linearity of TL and OSL and the physical situations leading to it can be found in McKeever (1997, Chapter 4), Chen and Pagonis (2011, Chapter 8) and Pagonis et al. (2012).

In the present work, we consider the dose dependence of both TL and OSL when high dose rates are being used. The study involves numerical simulation of the relevant set of simultaneous differential equations as well as an analytical treatment using plausible approximations. Here we will show that due to effects taking place during the excitation, the accumulation of electrons in traps and of holes in centers may be superlinear with the dose at high dose rates, and therefore, the area under the TL curve or the OSL curve can also be expected to be superlinear with the dose. The dose rates we use in the simulations are of the order of magnitude of 1 Gy/s, equivalent to a rate of production of electron–hole pairs of  $\sim 1.7 \times 10^{15} \text{ cm}^{-3} \text{ s}^{-1}$  (see e.g., Pagonis et al., 2006; Chen and Pagonis, 2011, p. 237). Note that transformation from Gy/s to electron–hole pairs per  $\text{cm}^3$  per second is based on the data concerning  $\text{Al}_2\text{O}_3$  with density of  $\rho = 4 \text{ g cm}^{-3}$  and under the assumption that the average energy deposited per electron–hole pair created is  $\sim 1.5E_g$  where  $E_g$  is the band gap. Note also that a different value of the conversion factor of  $\sim 4.4 \times 10^{14} \text{ cm}^{-3} \text{ s}^{-1}$  is given for LiF on p. 229 of the book by Chen and Pagonis (2011). The ratio of  $\sim 3.86$  between the two factors has to do with the different density of  $2.6 \text{ g cm}^{-3}$  of LiF and the assumption, based on Avila et al. (1999), that an average of  $\sim 34 \text{ eV}$  of  $\gamma$  rays is required for producing an electron–hole pair in LiF. The use of dose rates of this order of magnitude has been reported by Sato et al. (2004). Note that even significantly higher dose rates have been reported in the literature. Tillman et al. (1997) describe an X-ray source yielding dose rates up to  $10^9 \text{ Gy/s}$ . Niroomand-Rad et al. (1998) discuss dose rates of  $^{60}\text{Co}$  radiation up to  $10^{12} \text{ Gy/s}$ . The relevance of these works to the present case is that the total dose has been evaluated using TL dosimeters. These were certainly calibrated at much lower doses, so one may suspect that if the dose dependence is not linear at

these high dose rates, some inaccuracy may be introduced. Both the numerical simulation and the approximations show that a superlinear dose dependence of the occupancy of traps and centers occurs only if the initial occupancy of the centers is non-zero. This situation has previously been discussed by Chen and Leung (2001a), Yukihiro et al. (2004), Pagonis et al. (2009) and Chen et al. (2011). It has been pointed out by Carter (1970) that the circumstance that the center is partially filled by electrons may occur if the energy of the center is near the Fermi level. We can write  $m_0 = \alpha M$  where  $M$  is the total concentration of the center,  $0 < \alpha < 1$  and  $m_0$  the initial occupancy of the centers prior to excitation by irradiation.  $\alpha$  is expected to be determined by the Fermi statistics.

## 2. The model

The OTOR model governing the filling of traps and centers is shown in Fig. 1. The magnitudes shown are, respectively,  $N$  and  $M$ , the concentrations of traps and centers ( $\text{cm}^{-3}$ ),  $n$  and  $m$  their instantaneous occupancies ( $\text{cm}^{-3}$ ),  $n_c$  and  $n_v$ , the concentrations of free electrons and holes during excitation ( $\text{cm}^{-3}$ ),  $A_m$  and  $A_n$  are, respectively, the recombination and retrapping probability coefficients for electrons ( $\text{cm}^3 \text{ s}^{-1}$ ),  $B$  is the trapping probability coefficient of free holes in centers ( $\text{cm}^3 \text{ s}^{-1}$ ) and  $X$  is the rate of production of free electrons and holes ( $\text{cm}^{-3} \text{ s}^{-1}$ ), which is proportional to the dose rate of excitation. The differential rate equations governing the process during the excitation are

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

$$\frac{dm}{dt} = B(M - m)n_v - A_m m n_c, \quad (2)$$

$$\frac{dn_v}{dt} = X - B(M - m)n_v, \quad (3)$$

$$\frac{dn_c}{dt} = X - A_n(N - n)n_c - A_m m n_c. \quad (4)$$

If the process of irradiation takes place for a time  $t$ (s), the total concentration of electrons and holes produced during excitation is given by  $D = X \cdot t$ , where  $D$  denotes the dose, or rather, the total concentration of electron–hole pairs produced by the irradiation, which is proportional to the imparted dose.

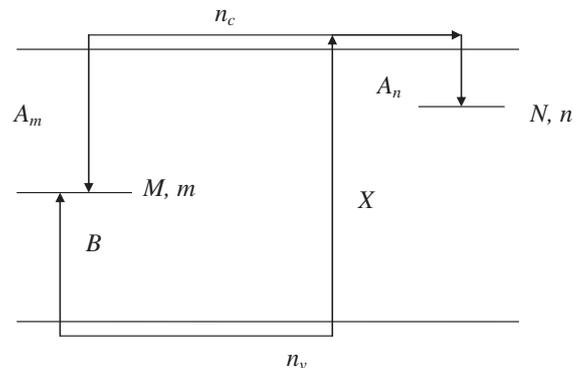


Fig. 1. The one-trap, one-center model of TL and OSL. The meaning of the parameters is given in the text.

$$\begin{aligned} \frac{d^2n}{dt^2} &= \frac{[A_n(N-n) + A_m m]A_n(-dn/dt) - A_n(N-n)[A_n(-dn/dt) + A_m \cdot dm/dt]}{[A_n(N-n) + A_m m]^2} X = \frac{(A_m m)A_n(-dn/dt) - A_n(N-n)(A_m \cdot dm/dt)}{[A_n(N-n) + A_m m]^2} X \\ &= -X \frac{A_m m \cdot A_n(N-n)}{[A_n(N-n) + A_m m]^2} \left( \frac{1}{N-n} \frac{dn}{dt} + \frac{1}{m} \frac{dm}{dt} \right). \end{aligned} \quad (13)$$

### 3. Analytical considerations

Let us consider the initial conditions,

$$n_c(0) = n_v(0) = n(0) = 0, \quad (5)$$

and

$$m(0) = m_0 \neq 0. \quad (6)$$

Making the usual quasi-equilibrium assumption,

$$\left| \frac{dn_c}{dt} \right| \ll A_n(N-n)n_c + A_m m n_c. \quad (7)$$

and using inequality (7) in Eq. (4), we find the quasi-steady value for the free electron density,

$$n_c = \frac{X}{A_n(N-n) + A_m m}. \quad (8)$$

Substituting Eq. (8) into Eqs. (1) and (2) yields

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

$$\frac{dm}{dt} = B n_v(M-m) - \frac{A_m m}{A_n(N-n) + A_m m} X. \quad (10)$$

Using the initial conditions (5) and (6), and assuming that the parameter  $B$  is relatively small so that the first term in Eq. (10) is initially small as compared to the second, we can evaluate from Eqs. (9) and (10)

$$\left. \frac{dn}{dt} \right|_{t=0} = \frac{A_n N}{A_n N + A_m m_0} X, \quad (11)$$

$$\left. \frac{dm}{dt} \right|_{t=0} = -\frac{A_m m_0}{A_n N + A_m m_0} X. \quad (12)$$

Note that the trap occupancy,  $n$ , has a positive initial slope reflecting the capture of free electrons from the conduction band into the trap. The occupancy of the center,  $m$ , however, has initially a negative slope because some free electrons from the conduction band recombine at the center, reducing its population. The population of the center,  $m$ , will eventually be replenished by the capture of free holes from the valence band into the center. However, initially,  $n_v = 0$ , so this will happen only later.

Let us consider the second derivative of  $n$  with respect to time by differentiating Eq. (9),

To form a Taylor series, we need to evaluate the second derivative in Eq. (13) at  $t = 0$ ,

$$\begin{aligned} \left. \frac{d^2n}{dt^2} \right|_{t=0} &= -X \frac{A_m m_0 A_n N}{(A_n N + A_m m_0)^2} \left( \left. \frac{1}{N} \frac{dn}{dt} \right|_{t=0} + \left. \frac{1}{m_0} \frac{dm}{dt} \right|_{t=0} \right) \\ &= X^2 \frac{A_m m_0 A_n N}{(A_n N + A_m m_0)^3} (A_m - A_n), \end{aligned} \quad (14)$$

where we have used Eqs. (5), (11) and (12).

It is usually convenient to plot  $n$  as a function of the dose  $D$  rather than of time. Consequently, let us use  $dD = Xdt$  and re-write Eqs. (11) and (14) as

$$\left. \frac{dn}{dD} \right|_{t=0} = \frac{A_n N}{A_n N + A_m m_0}, \quad (15)$$

$$\left. \frac{d^2n}{dD^2} \right|_{t=0} = \frac{A_m m_0 A_n N}{(A_n N + A_m m_0)^3} (A_m - A_n). \quad (16)$$

The Taylor series which yields the initial dose dependence is

$$n = \left. \frac{dn}{dD} \right|_{t=0} D + \frac{1}{2} \left. \frac{d^2n}{dD^2} \right|_{t=0} D^2 + O(D^3), \quad (17)$$

where  $O(D^3)$  means a term of the order of  $D^3$ . This term may be negligible at the low-dose range.

Substituting Eqs. (15) and (16) into (17) yields

$$n = aD + bD^2 + O(D^3), \quad (18)$$

where

$$a = \frac{A_n N}{A_n N + A_m m_0}, \quad (19)$$

$$b = \frac{1}{2} \frac{A_m m_0 A_n N}{(A_n N + A_m m_0)^3} (A_m - A_n). \quad (20)$$

The physical meaning of  $a$  is the initial fraction of free electrons which are captured by the trap rather than recombining with a hole in the center. The coefficient  $b$  governs whether the initial response is superlinear, linear or sublinear. According to Eq. (20), the behavior is superlinear ( $b > 0$ ) if

$$m_0 > 0 \text{ and } A_m > A_n. \quad (21)$$

The initial dose dependence is expected to be linear if  $m_0 = 0$  or, if  $m_0 > 0$ ,  $A_m = A_n$ . The results of simulations can be compared to those reached by Eq. (18). At the low-dose range, the two can be expected to coincide. At higher doses, the  $D^3$  and higher terms become important and, on the other hand, saturation effects that have not been considered in this analytical treatment may set in.

### 4. Numerical results

The numerical simulation consisted of solving the set of Eqs. (1)–(4) for a chosen set of trapping parameters. The solution was performed using the Matlab ode15s program. In the following examples, the simulated occupancy of the traps following excitation is shown as a function of the dose. The dotted line (I) gives the results from the numerical solution of the differential equations whereas the dashed lines (II) shows the quadratic approximation from Eq. (18) with  $a$  and  $b$  calculated with the aid of Eqs. (19) and (20). Note that the values of the trap occupancy as a function of dose represent the total area either under a single TL peak or under an OSL curve. This is the case because we are dealing with a simple one trap-one center situation in which, since  $m_0 > 0$ ,  $n < m$  all along.

Fig. 2 shows the dose dependence simulated using the following parameters:  $M = 10^{16} \text{ cm}^{-3}$ ;  $m_0 = 10^{15} \text{ cm}^{-3}$ ;  $N = 10^{15} \text{ cm}^{-3}$ ;  $A_n = 2 \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$ ;  $A_m = 10^{-8} \text{ cm}^3 \text{ s}^{-1}$ ;  $B = 10^{-16} \text{ cm}^3 \text{ s}^{-1}$ . As shown, the simulated results from the numerical solution of the differential equations and the quadratic approximation are very close to each other, and the superlinear dose dependence is clearly seen in the given dose range of up to  $3 \times 10^{15} \text{ cm}^{-3}$ . Note that the excitation dose rate has been here up to  $3 \times 10^{15} \text{ cm}^{-3} \text{ s}^{-1}$  and the excitation time for each simulation  $t = 1 \text{ s}$ .

Fig. 3 depicts the dose dependence with the same parameters and the same doses, but the dose rates are 10 times higher and the time of excitation shorter by the same factor of 10, namely,  $t = 0.1 \text{ s}$ . As seen, the results of the simulations and approximation coincide only up to a dose of  $\sim 7 \times 10^{14} \text{ cm}^{-3}$ , and at higher doses, the results of the simulations increase with the dose much faster than those of the approximation, until it goes abruptly to saturation at  $n = 10^{15} \text{ cm}^{-3}$ , which obviously means that  $n = N$ .

Fig. 4 shows the results of simulations, again with the same parameters and range of doses, but at dose rates lower by a factor of 10, and longer excitation time of  $t = 10 \text{ s}$ . Here, the simulated curve and the approximation results coincide only at very low doses, up to  $\sim 3 \times 10^{14} \text{ cm}^{-3}$ . The approximate results behave, of course, quadratically, but the simulated results start nearly linearly and then continue slightly sublinearly.

Fig. 5 depicts the results of the simulations and the approximation with the same set of parameters and the same range of

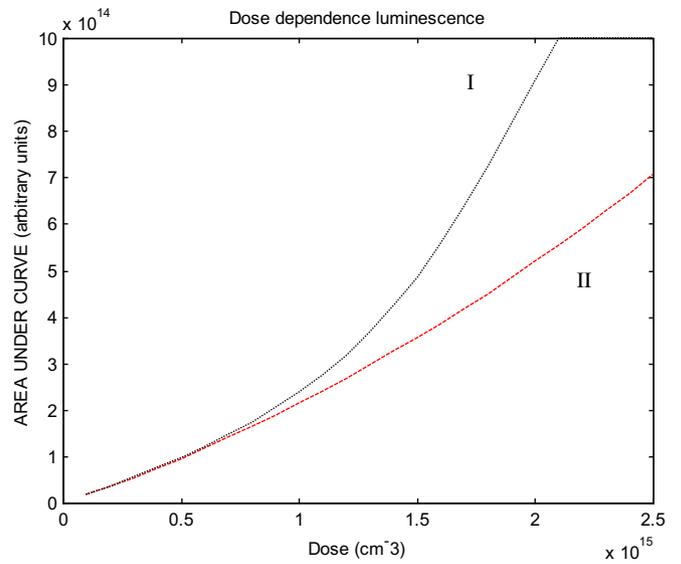


Fig. 3. Same as Fig. 2, but the dose rates are 10 times larger and  $t = 0.1 \text{ s}$ .

doses. The difference here is that we keep the dose rate constant at  $10^{16} \text{ cm}^{-3} \text{ s}^{-1}$ , and vary the time of excitation between 0.01 s and 0.3 s. The simulated and approximate curves are both superlinear. They coincide up to a dose of  $\sim 7 \times 10^{14} \text{ cm}^{-3}$ , and then the simulated curve increases faster than that of the approximate results. Here too, the simulated curve goes to saturation at  $\text{area} = 10^{15}$  rather abruptly.

Fig. 6 depicts the simulated and approximate dose-dependence curves for a case where  $A_m < A_n$ . All the parameters here, including the dose rates and the time of excitation are the same as in Fig. 3, except that  $A_m = 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ . As could be expected, the initial dependence here is not superlinear. The dose dependence is nearly linear at low doses, and then goes gradually to saturation. The simulated and approximate curves coincide up to  $\sim 6 \times 10^{14} \text{ cm}^{-3}$ .

Fig. 7 shows the possibility of dose-rate dependence under the same circumstances. The same parameters as in Fig. 2 are used. The dose rate  $X$  was varied between  $10^{13}$  and  $10^{17} \text{ cm}^{-3} \text{ s}^{-1}$ , and the time of excitation  $t$  was varied inversely so that the total dose was

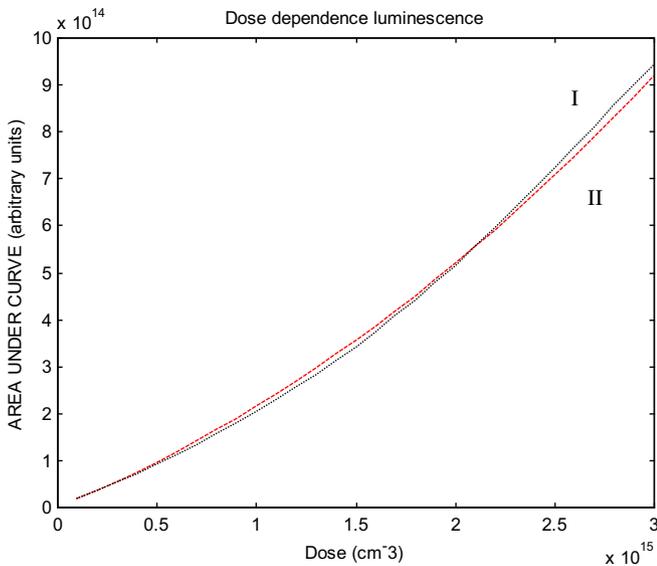


Fig. 2. Simulated dose dependence (I) and approximate dependence based on Eq. (18), and (II) with dose rates up to  $3 \times 10^{15} \text{ cm}^{-3} \text{ s}^{-1}$  and time of excitation  $t = 1 \text{ s}$ . The relevant parameters are given in the text.

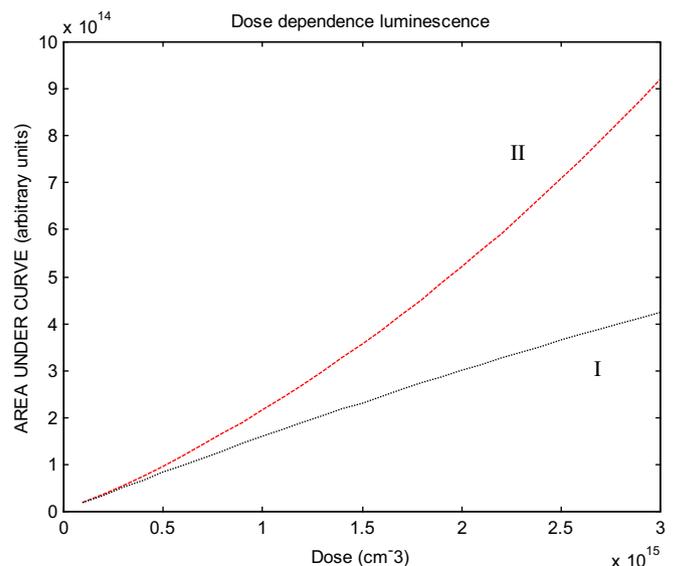
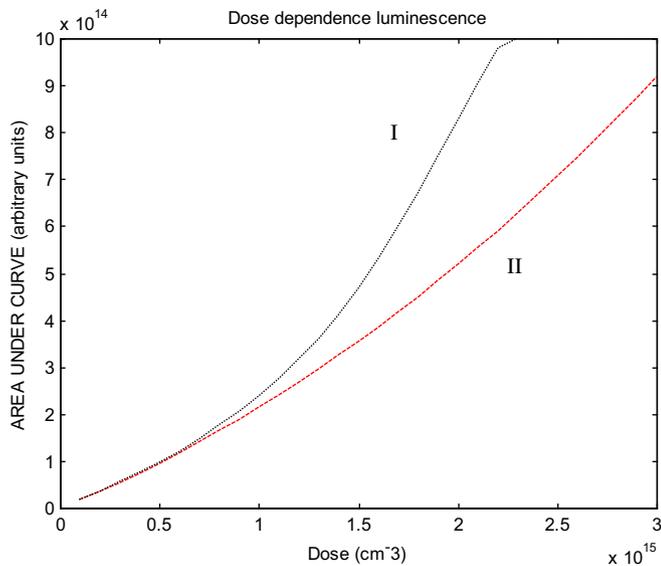


Fig. 4. Same as Fig. 2, but the dose rates are 10 times smaller and  $t = 10 \text{ s}$ .



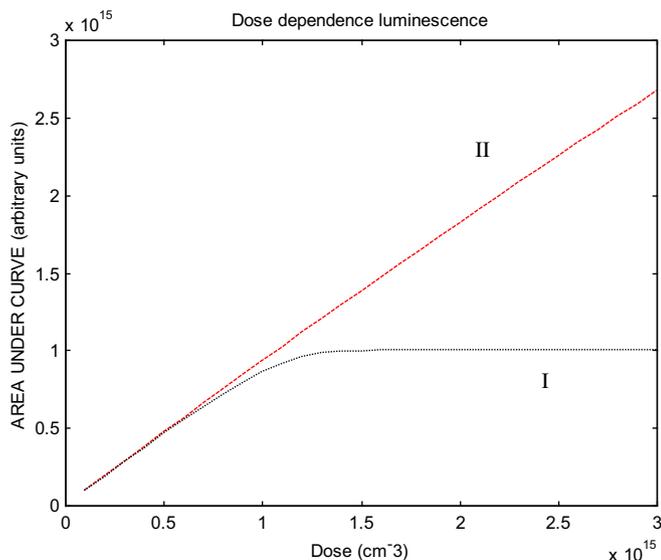
**Fig. 5.** Same as in the previous figures, but the dose rate is constant,  $X = 10^{16} \text{ cm}^{-3} \text{ s}^{-1}$  and the excitation time  $t$  varies between 0.01 s and 0.3 s. Thus, the dose range is the same as before.

kept the same,  $10^{15} \text{ cm}^{-3}$ . The X-axis is given as a 10-base logarithm.

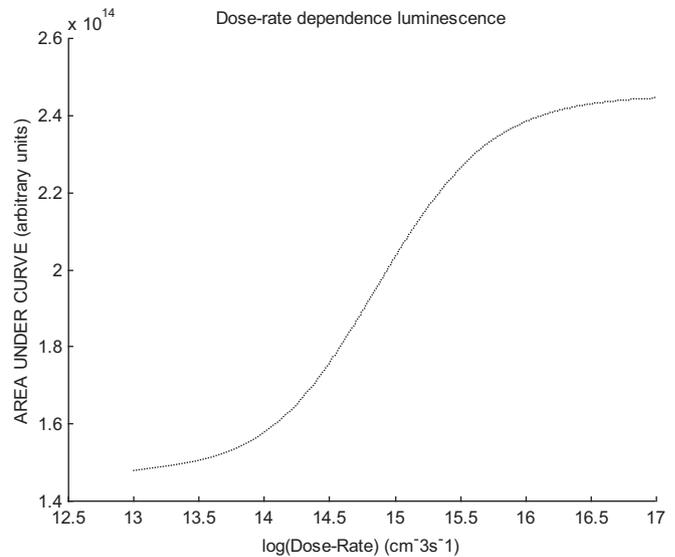
## 5. Discussion

We have demonstrated in this work that within the simplest model of one trapping state and one kind of recombination center, the filling of traps may be superlinear with the dose of excitation. As a result, the area under a thermoluminescence peak or an OSL decay curve may be superlinear with the dose. The conditions leading to this behavior are studied. Numerical results as well as an analytical treatment are presented, the latter being a good approximation at relatively low doses.

A necessary condition for the occurrence of this effect is that initially, prior to excitation, the occupancy of the centers is non-zero ( $m_0 > 0$ ). This situation has been dealt with by previous



**Fig. 6.** Same as in Fig. 2, but the recombination probability coefficient is taken to be smaller,  $A_m = 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ , so that  $A_m < A_n$ .



**Fig. 7.** Simulated dose-rate dependence of the accumulated carriers in traps. The parameters are the same as in Fig. 2. The time of excitation  $t$  is varied inversely so that the total dose is kept constant,  $D = 10^{15} \text{ cm}^{-3}$ . The X-axis is given in a 10-base logarithmic scale.

researchers as mentioned above. Another important condition is that the recombination probability coefficient be larger than the retrapping probability coefficient ( $A_m > A_n$ ), as is obvious from Eq. (20). Yet another crucial condition is that the excitation dose rate be rather high. The numerical results in Figs. 2–4 show how the superlinearity depends on the dose rate. When the dose rate is low (Fig. 4), no superlinearity occurs, as seen in curve I, but at higher dose rates, it sets in and gets stronger as shown in Figs. 2 and 3. In all these cases, the dose is changed by using a variable dose rate and keeping the time of excitation constant (but varying from one case to another). In Fig. 5, it is shown that the superlinearity also occurs when a constant dose rate is used, and the time of excitation varies, provided that the dose rate is high enough. The opposite case of  $A_m < A_n$  is tested in Fig. 6. From Eq. (20), one expects  $b$  to be negative which means sublinearity as of the lowest doses. However, in this case, from Eq. (19), one gets  $a \approx 1$  and from Eq. (20) it is obvious that  $b$  is significantly smaller for this case. One therefore expects an initial nearly linear dose dependence, which is really seen in Fig. 6.

The role of high dose rates for getting this kind of superlinear dose dependence has been mentioned. Fig. 7 shows the transition from “low” to “high” dose rates by checking the dose-rate dependence for a constant total dose. With the given parameters, a change of  $\sim 70\%$  is seen while going from  $X = 10^{13}$  to  $X = 10^{17} \text{ cm}^{-3} \text{ s}^{-1}$ .

The main practical aspect of this work has to do with the dose evaluation by TL dosimeters (TLDs) in cases where very high dose rates are used. Niroomand-Rad et al. (1998) mention the use of TLDs for evaluation of the dose when the dose rate may be as high as  $10^{12} \text{ Gy/s}$ . Obviously, the calibration of the dosimeter has been performed at dose rates many orders of magnitude smaller. The concern arising from the results in the present work is that due to the mentioned superlinearity, the dose thus evaluated may not be accurate.

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