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THE ROLE OF RETRAPPING IN DOSE DEPENDENCE OF PULSED OPTICALLY STIMULATED LUMINESCENCE

R. Chen
School of Physics and Astronomy
Raymond and Beverly Sackler Faculty of Exact Sciences
Tel-Aviv University, Tel-Aviv 69978, Israel

Abstract — Superlinear behaviour has been observed in dose dependence measurements of optically stimulated luminescence (OSL). Previous theoretical work has shown that superlinearity of the integral under the OSL decay curve may occur, resulting from competition with either radiationless luminescence centres or disconnected trapping states. Also, it has recently been shown that if OSL is measured during a relatively short pulse of stimulating light, quadratic dose dependence may take place even when only one trapping state and one kind of recombination centre take part. Here, the conditions for superlinear dose dependence, quadratic and more, to occur are considered. Also, the dose dependence of 'pulsed OSL' is discussed. In this procedure, the luminescence is only detected after the end of the stimulating light pulse; it is shown that when retrapping is relatively strong and initially the traps and centres are empty, superlinear dose dependence is expected.

INTRODUCTION

Optically stimulated luminescence (OSL) has become a very important method for radiation dosimetry. In many laboratories, it is replacing the well-established method of thermoluminescence (TL). The main advantages of OSL are that the sample is not heated during the readout, and therefore blackbody radiation is avoided. Also, possible thermal quenching of luminescence is prevented.

In many reports on OSL it is assumed and sometimes shown[1] that the dose dependence of OSL is initially linear, followed by an approach to saturation; the utilisation of this method for dosimetry and dating is basically dependent on this premise. There are, however, some reports on superlinear dose dependence of OSL in quartz and mixed feldspars from sediments. Godfrey-Smith[2] reported a linear dose dependence of unheated samples, but 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*[3] 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 applied dose and a, b and c are positive magnitudes. Bøtter-Jensen *et al*[4] also reported a superlinearity of the OSL signal in quartz samples extracted from bricks while performing retrospective dosimetry. They pointed out that linearity takes place in unannealed samples whereas superlinearity occurs in annealed specimens. These authors presented a model with three trapping states and two recombination centres, performed a numerical simulation of the relevant coupled differential equations and found as a result a superlinear dose dependence of OSL. Further work along the same lines was reported by Banerjee[5]. He also found superlinear dose dependence of OSL in annealed quartz in the range of 0–5 Gy, and explained it using arguments previously employed by Kristianpoller *et al*[6] for TL superlinearity. This work explained the possible occurrence of superlinearity of TL due to competition either between trapping states or between recombination centres during the readout stage. Banerjee[5] suggested that the OSL dose response is entirely analogous to that of TL, thus explaining the superlinear dose dependence of OSL. Chen and Leung[7], using numerical simulation with a model including only one trapping state and one kind of recombination centre, theoretically studied the response to dose of OSL during short pulses of stimulating light. They found that when non-first-order conditions prevail, an initial quadratic dose dependence can be expected, provided that, before the irradiation, the relevant traps and centres are empty or nearly empty, which could be expected in annealed samples.

This kind of OSL in response to short pulses of stimulating light is quite common in present use. In particular, a variant termed 'pulsed optically stimulated luminescence (POSL)' has been described by McKeever and Akselrod[1], in which the luminescence is only detected after the end of the stimulating light pulse.

In the present work, the nonlinear dose dependence of the OSL signal in response to stimulating light *pulses* is further studied. Using simulations as well as an approximate analytical approach, a quadratic and more than quadratic dose dependence is predicted for the initial dose range, mainly in annealed samples. The results of both the response to dose during the stimulating pulse and following it are considered. The conditions under which linear dose dependence can be expected are also discussed, within the simplest possible model, which includes only one trapping state and one kind of recombination centre. The main point here is

Contact author E-mail: chenr@ccsg.tau.ac.il

the effect of retrapping of optically freed carriers on the dose dependence.

THEORETICAL CONSIDERATIONS

As pointed out previously[7], the set of simultaneous differential equations governing the process of excitation by irradiation when one trapping state and one kind of recombination centre are involved (see Figure 1) is

$$dn_v/dt = x - B(M - m)n_v \quad (1)$$

$$dm/dt = - A_m m n_c + B(M - m)n_v \quad (2)$$

$$dn/dt = A_n(N - n)n_c \quad (3)$$

$$dn_c/dt = dm/dt + dn_v/dt - dn/dt \quad (4)$$

where N (m^{-3}) is the concentration of electron traps and M (m^{-3}) the concentration of hole centres. x ($m^{-3}.s^{-1}$) is the rate per unit volume of production by the irradiation of electrons and holes in the conduction and valence band, respectively. Assuming a homogeneous excitation of the sample, x is proportional to the excitation intensity. The instantaneous concentrations of electrons in the conduction band and holes in the valence band are, respectively n_c (m^{-3}) and n_v (m^{-3}). The free electrons in the conduction band can be trapped with a retrapping coefficient A_n ($m^3.s^{-1}$) whereas the free holes from the valence band can be trapped in the centres with a probability coefficient of B ($m^3.s^{-1}$). The instantaneous occupancy of electrons in traps and holes in centres during excitation and also during the optical stimulation are denoted by n (m^{-3}) and m (m^{-3}), respectively. After the excitation is finished, a relaxation period is allowed for; 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 centres, respectively. Once holes are accumulated in the centre, recombination may take place between these holes and free electrons with a recombination coefficient of A_m ($m^3.s^{-1}$).

At the next stage of optical stimulation, it is assumed that the applied light releases electrons from the trapping state at a rate of fn ($m^{-3}.s^{-1}$) where f (s^{-1}) is the optical stimulation rate. Again, homogeneous illumination of the sample is assumed. fn is analogous to x ($m^{-3}.s^{-1}$) in the excitation stage, except that x is constant whereas fn varies with n. The final values of the functions n, m, n_c and n_v at the end of the relaxation as initial values for the optical stimulation can now be taken. Three simultaneous differential equations are considered

$$-dm/dt = A_m m n_c \quad (5)$$

$$dn/dt = - fn + A_n(N - n)n_c \quad (6)$$

$$dn_c/dt = dm/dt - dn/dt \quad (7)$$

Since the intensity of the OSL signal is associated with the recombination rate, the OSL intensity I can be written as

$$I = - dm/dt \quad (8)$$

In the following, the OSL measured *during* a stimulating light pulse and following such a pulse is discussed separately. In the case where the simulation is of luminescence *following* the light pulse, Equations 5–7 will be solved for a further length of time with f = 0.

The set of Equations 5–7 is very similar to that governing the heating stage in TL, but instead of f (s^{-1}), the optical stimulation rate, there is in TL s exp(–E/kT), the thermal excitation rate where s (s^{-1}) is the frequency factor, E (eV) the activation energy, k ($eV.K^{-1}$) the Boltzmann factor and T (K) the temperature. Making here, as in TL, the quasi-equilibrium assumptions[6],

$$|dn_c/dt| \ll |dn/dt|, |dm/dt|\ n_c \ll n, m \quad (9)$$

then for OSL, in full analogy with TL, the simplified equation

$$I = \frac{f A_m m n}{A_m m + A_n(N - n)} \quad (10)$$

is obtained. First consider the restricted case of pure second order, where some of the conclusions can be reached in an analytical manner. The additional simplifying assumptions N ≫ n (trapping states are far from saturation), n = m and $A_m m \ll A_n N$ (retrapping dominates) are made. Equation 10 reduces to

$$I = - \frac{dn}{dt} = \frac{f A_m}{N A_n} n^2 \quad (11)$$

Denoting the constant $A_m/(A_n N)$ by k, solving this differential equation for a given f, and re-inserting n into Equation (11) produces

$$I = \frac{f k n_o^2}{(1 + f k n_o t)^2} \quad (12)$$

For small values of t, $t \ll 1/(f k n_o)$,

$$I = f k n_o^2 \quad (13)$$

As long as $n_o \propto D$, which is indeed the case for this simple situation of one trapping state and one kind of

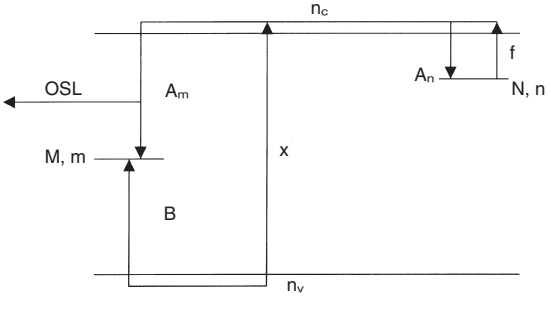

Figure 1. Energy level diagram for excitation and stimulation of OSL. A trapping state N and a centre M are involved.

centre in the initial range where $n \ll N$, we get $I \propto D^2$. This is the situation for an instantaneous OSL intensity at a given time point in the short time region. If one integrates over a certain time period within the short time range, the dependence on the dose will still be quadratic. It is now shown that the total area under the curve depends linearly on the excitation dose by integrating I in Equation 12 from 0 to infinity,

$$\int_0^\infty Idt = \int_0^\infty \frac{fkn_o^2}{(1 + fkn_o t)^2}\, dt = \left[\frac{-n_o}{1 + fkn_o t}\right]_o^\infty = n_o \tag{14}$$

As long as n_o depends linearly on the dose, the area under the whole OSL curve is also linear with the dose.

Returning to Equation (10) without additional simplifying assumptions, in the strict case of one trapping state and one kind of recombination centre, the condition $n = m$ can be assumed to hold true since $n_c \ll n$. In the presence of additional trapping states and/or centres, the condition $n \neq m$ is usual. It has been shown[5] that when competition takes place, the dependence of the area under the OSL curve may be superlinear. This may certainly influence the dose dependence of the pulsed signal discussed here. However, situations in which the effect of competition with other traps or centres is minimal and yet $n \neq m$ can be considered. Even in this case, the conclusions reached in the pure second order case do not change significantly. In the case of dominating recombination, $A_m m \gg A_n(N-n)$, one has first order behaviour,

$$I \cong fn \tag{15}$$

and as long as n grows linearly with the dose, the response of OSL to a pulse of stimulating light (as well as the area under the decaying OSL curve) is linear. If, however, retrapping dominates, $A_n(N-n) \gg A_m m$, Equation (10) reduces to

$$I \cong \frac{fA_m mn}{A_n(N - n)} \tag{16}$$

If, in addition, the trapping state is far from saturation, $N \gg n$, this reduces to

$$I \cong \frac{fA_m mn}{A_n N} \tag{17}$$

and as long as both m and n grow linearly with the excitation dose, the response of the OSL to a pulse of stimulating light will be quadratic with the dose. If, however, the occupancy of the trapping states, n, approaches saturation, there may be situations in which a faster than quadratic dose dependence may take place. Here, N–n appearing in Equations 10 and 16 is a decreasing function of the dose. If both m and n are nearly linear with the dose, the additional function N–n appearing in the denominator causes the whole expression to have a more-than-quadratic dose dependence.

For the response to a light pulse during the stimulation, Chen and Leung[7] showed that starting with a nearly filled trapping state (e.g. $n_o = 0.9$ N), the dose dependence of OSL could be more or less linear. This agreed with the experimental fact that, at least for quartz samples, linearity was often observed in the unannealed samples and superlinearity in the annealed ones. It appears that the former has to do with $n_o \neq 0$ and the latter with $n_o \approx 0$. Some qualitative conclusions are now considered that can be drawn from Equations 10, 16 and 17 in the different conditions.

As pointed out above, the case of dominating retrapping is of main interest since the situation of dominating recombination (Equation 15) leads to the simple first order case in which linearity with the dose is expected. Equation 17 results from the assumption $N \gg n$ which does not agree with $n_o = 0.9$ N taken in the mentioned work[7]. As for Equation 16, taking $m_o = 0$ and $n_o = 0.9$ N may bring about a situation in which the variation in n with irradiation is relatively small, and therefore the dose dependence of OSL intensity is that of m. This, in turn, may be linear with the dose. The inverse possibility also exists, namely $n_o = 0$ and $m_o \neq 0$, say, $m_o = 0.9$ M. Here, the variation of m with the dose may be small and the dose dependence of I is the same as that of n, which may be linear. As mentioned already, the situation changes at very high doses when n approaches N.

NUMERICAL RESULTS

The simultaneous differential equations 5–7 are nonlinear and therefore cannot be solved analytically. The approach that was followed so far depended on making simplifying assumptions which enabled us to reach general conclusions in an analytical form. The drawback is that the approximations made may have an influence on the final results. The alternative is to choose a set of reasonable trapping parameters and solve the relevant equations numerically. Thus, the results are accurate but limited since they are valid only for the chosen set of parameters and therefore are basically a demonstration that certain dose dependence is possible within the simple model. However, if there is an agreement between the two routes taken, an amount of credibility is added to the theoretical results.

In order to follow the experimental procedure, the set of Equations 1–4 has first been solved for a certain period of time t_D for a given value of x, the rate of production of electrons and holes per unit volume by the irradiation. The total dose is represented by $D = x\, t_D$ which has dimensions of m^{-3} and actually is the total concentration of produced electrons and holes. This was followed by continuing the solution procedure during the relaxation stage as described in the Introduction, and finally, Equations 5–7 were numerically solved with the given optical stimulation rate f. Two different routes were taken which follow the two different experimental procedures in use. One is the response to a short optical

stimulating pulse *during* the exposure and the other is the light emission *following* the exposure for a short period of the decay.

An example of the calculated dose dependence of these two versions is shown in Figure 2 on a log–log scale. The parameters chosen are $A_m = 10^{-10}$ ($m^3.s^{-1}$), $B = 10^{-15}$ ($m^3.s^{-1}$), $A_n = 10^{-9}$ ($m^3.s^{-1}$), $N = 10^{13}$ (m^{-3}), $M = 10^{13}$ (m^{-3}), $t_D = 0.1$(s), $f = 10$ (s^{-1}), x varies from 10^{12} to 10^{16} ($m^{-3}.s^{-1}$) and $n_o = m_o = n_{co} = n_{vo} = 0$. The points denoted by (×) show the results calculated in a short period of time *during* the light stimulation and those by (o) the emission intensity in a short period of time *following* the stimulating pulse. The general appearance of the two curves is the same with slight differences in details. The dose dependence starts quadratic with the dose, continues faster than quadratic in a narrow dose range, then goes more or less linear before approaching saturation.

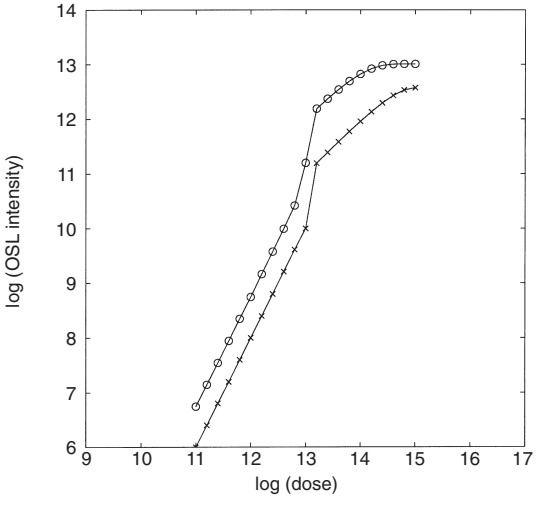

Figure 2. Calculated dose dependence of pulsed OSL. (×) represents the OSL intensity in a short period of time *during* the stimulation. (o) depicts the simulated light intensity in a short period of time *following* a short stimulating pulse. The values of the parameters are given in the text.

CONCLUSION

The possible superlinear dose dependence of pulsed OSL is further studied theoretically, using the simplest possible energy level model of one trapping state and one kind of recombination centre. Results are reached by using the quasi-equilibrium assumption as well as by solving numerically the relevant differential equations with no simplifying assumptions. Two existing experimental procedures are followed, namely, the measurement of the emitted light *during* a short pulse of the stimulating light and the measurement of the emitted light *following* such a pulse. In both cases, quadratic followed by more than quadratic dose dependence, and then, linear, followed by sublinear dependence is observed. In other results not shown here, if one starts with large m_o or n_o, say, $n_o = 0.9$ N or $m_o = 0.9$ M, the initial dose dependence is nearly linear, followed by somewhat superlinear dependence before the approach to saturation. The apparent lack of similarity in the behaviour of pulsed OSL and TL can be explained as follows. Had the total area under the OSL decay curve been taken, obviously in this simple case of a one trap-one centre, linear dependence could be expected. Also, in the case of dominating recombination, the situation would be similar to a first-order TL peak in which each part of the curve grows linearly with the dose. This is not the case, however, when retrapping dominates. The analogue of the pulsed OSL is the intensity of TL in the initial-rise range. As shown by Chen *et al*[8], in a second order TL peak, the dose dependence is expected to be quadratic in the initial-rise range. The case shown here of dominating retrapping is broader than the relatively simple second order, but the main points of analogy hold.

REFERENCES

1. McKeever, S. W. S. and Akselrod, M. S. *Radiation Dosimetry using Pulsed Optically Stimulated Luminescence of Al_2O_3:C*. Radiat. Prot. Dosim. **84**, 317–320 (1999).
2. Godfrey-Smith, D. I. *Thermal Effects in the Optically Stimulated Luminescence of Quartz and Mixed Feldspars from Sediments*. J. Phys. D: Appl. Phys. **27**, 1737–1746 (1994).
3. Roberts, R. G., Spooner, N. A. and Questiaux, D. G. *Palaeodose Underestimates Caused by Extended Duration Preheats in the Optical Dating of Quartz*. Radiat. Meas. **23**, 647–653 (1994).
4. Bøtter-Jensen, L., Agersap Larsen, N., Mejdahl, V., Poolton, N. R., Morris, M. F. and McKeever, S. W. S. *Luminescence Sensitivity Changes in Quartz as a Result of Annealing*. Radiat. Meas. **24**, 535–541 (1995).
5. Banerjee, D. *Supralinearity and Sensitivity Changes in Optically Stimulated Luminescence of Annealed Quartz*. Radiat. Meas. **33**, 47–57 (2001).
6. Kristianpoller, N., Chen, R. and Israeli, M. *Dose Dependence of Thermoluminescence Peaks*. J. Phys. D: Appl. Phys. **7**, 1063–1072 (1974).
7. Chen, R. and Leung, P. L. *Dose Dependence and Dose-rate Dependence of the Optically Stimulated Luminescence (OSL) Signal*. J. Appl. Phys. **89**, 259–263 (2001).
8. Chen, R., Huntley, D. J. and Berger, G. W. *Analysis of Thermoluminescence Dominated by Second-order Kinetics*. Phys. Status Solidi A **79**, 251–261 (1983).