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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.

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 "supralinearity 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 Al_2O_3: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 $\sim 1.7 \times 10^{15}$ cm^{-3} 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 cm^3 per second is based on the data concerning Al_2O_3 with density of $\rho = 4$ 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 $\sim 4.4 \times 10^{14}$ cm^{-3} s^{-1} is given for LiF on p. 229 of the book by Chen and Pagonis (2011). The ratio of ~ 3.86 between the two factors has to do with the different density of 2.6gcm^{-3} of LiF and the assumption, based on Avila et al. (1999), that an average of ~ 34 eV of γ 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 Gy/s. Niroomand-Rad et al. (1998) discuss dose rates of ^{60}Co radiation up to 10^{12} 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), Yukihara 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. α 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 (cm^{-3}), n and m their instantaneous occupancies (cm^{-3}), n_c and n_v, the concentrations of free electrons and holes during excitation (cm^{-3}), A_m and A_n are, respectively, the recombination and retrapping probability coefficients for electrons (cm^3 s^{-1}), B is the trapping probability coefficient of free holes in centers (cm^3 s^{-1}) and X is the rate of production of free electrons and holes (cm^{-3} s^{-1}), which is proportional to the dose rate of excitation. The differential rate equations governing the process during the excitation are

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

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

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

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

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.

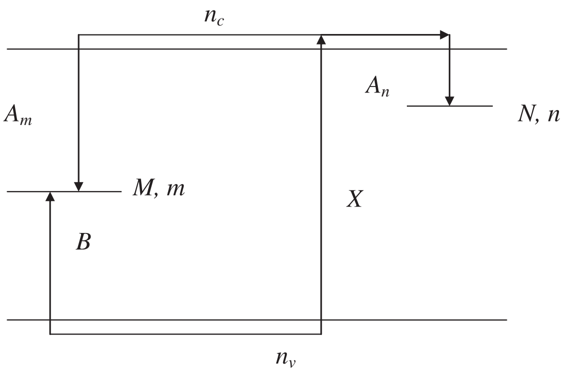

Fig. 1. The one-trap, one-center model of TL and OSL. The meaning of the parameters is given in the text.