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DOSE-RATE DEPENDENCE OF THERMOLUMINESCENCE RESPONSE

S.W.S. McKEEVER, R. CHEN *, P.J. GROOM ** and S.A. DURRANI
Department of Physics, University of Birmingham, P.O. Box 363, Birmingham B15 2TT, England

The previously observed dose-rate effect of thermoluminescence in quartz at high dose-rates is given a theoretical formulation. Computer calculations simulating the experimental conditions yield similar results to the experimental ones.

Most research workers in the fields of TL dating and dosimetry assume that the TL response is independent of the dose-rate [1,2]. Justification for this assumption stems predominantly from the experimental work of Karzmark et al. [3] and Tochilin and Goldstein [4] who observed a dose-rate independent TL response from LiF for dose-rates ranging from 5×10^2 to 1.7×10^{10} Rad s^{-1}. On the other hand, Groom et al. [5] reported a significant decrease of TL with increasing dose-rate in powdered samples of Brazilian crystalline quartz when irradiated with ^{60}Co γ-rays at dose-rates ranging from 0.14 to 330 Rad s^{-1}. A smaller effect was reported by Hsu and Weng [6].

A simple model of the excitation of TL in which electrons are raised, by the irradiation, from the valence into the conduction band and then accumulate in a trap, with the remaining holes in the valence band accumulating in a centre, would result in dose-rate independence. The purpose of the present work is to show that if we allow for recombination during excitation, an appreciable dose-rate effect may result. The amount of the effect depends on the relative magnitudes of the trapping parameters involved and on the total dose imparted.

The simple energy-level scheme used is depicted in fig. 1. One electron trapping state and one hole centre are assumed, having, respectively, concentrations N and N_h (cm^{-3}); the instantaneous populations of electrons and holes in these states are denoted by n and n_h (cm^{-3}), respectively. n_c and n_v (cm^{-3}) represent the instantaneous concentrations of electrons in the conduction band and holes in the valence band, respectively. A ($cm^3\ s^{-1}$) is the trapping probability of electrons from the conduction band; A_h ($cm^3\ s^{-1}$) the probability of trapping of holes in the centre; and A_R ($cm^3\ s^{-1}$) the probability of recombination of an electron in the conduction band with a hole trapped already in the centre. Instead of speaking in terms of dose-rate imparted r (Rad s^{-1}), we consider the 'electron–hole generation rate' f ($cm^{-3}\ s^{-1}$). The two are related by $f = kr$, where, e.g. in quartz, k was reported [7] to be 6×10^{12} cm^{-3} Rad^{-1}. Similarly, we consider D, the 'total electron–hole generation' (cm^{-3}) which is related to the total dose in Rad by the same constant k. Assuming f to be constant over the irradiation time, the total generation during t seconds of irradiation is $D = ft$. The system thus involves the parameters N, N_h, A, A_h, A_R, f and D, and the unknown functions $n(t)$, $n_h(t)$, $n_v(t)$ and $n_c(t)$. The four coupled equations governing the process are

$$dn_c/dt = f - n_c A_R n_h - n_c(N - n) A \ ;$$

$$dn/dt = n_c(N - n) A \ ;$$

$$dn_v/dt = f - n_v(N_h - n_h) A_h \ ;$$

$$dn_h/dt = n_v(N_h - n_h) A_h - n_c n_h A_R \ .$$

For a given set of the parameters, these simultaneous equations can be numerically solved. The sixth order

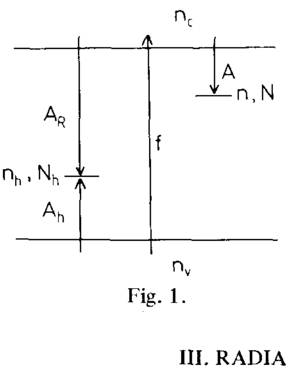

Fig. 1.

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* Permanent address: Department of Physics and Astronomy, Tel-Aviv University, Ramat-Aviv, Israel.

** Now at British Nuclear Fuels Limited, Risley, Warrington, England.

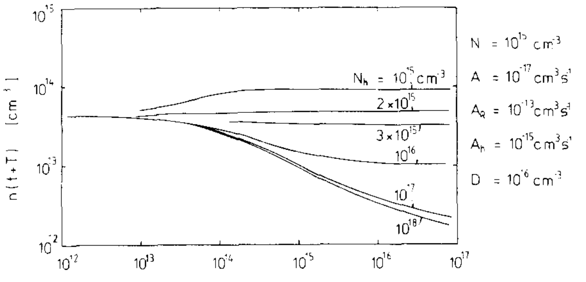

Fig. 2.

Predictor–Corrector method has been used for the solution. An important point considered is that, since the dose-rates involved are rather large, the concentrations, at the end of irradiation, of electrons in the conduction band, $n_c(t)$, and of holes in the valence band, $n_v(t)$, may be quite substantial. If one wants to simulate the experimental conditions, the addition of carriers to the trapping states *after* the end of irradiation (and before read-out starts) is to be accounted for. This has been done by continuing the computation for a further time T at the end of which both n_c and n_v have decayed to negligible values. In a series of computations, a certain value of D has been chosen, and the generation rate f is changed, along with irradiation time t, in such a way that $ft = D$ is constant. It is assumed that the final value $n(t + T)$ represents the measurable TL intensity; this is usually the case as long as $n \leqslant n_h$. [In the present framework, we have $n(t + T) \cong n_h(t + T)$.] An example of some of the results is shown in fig. 2. The parameters chosen are indicated on the graph. It is seen that at low dose-rates the curves merge, as the result becomes dose-rate independent. The result is very sensitive to the value of N_h. As N_h decreases, with the other parameters kept constant, the effect can change from one of decreasing $n(t + T)$ with increasing f, to one which increases with f. The maximum decrease calculated is by a factor of ~20; this may be compared with a factor of ~5 reported by Groom et al. [5] in quartz. For a given total dose, the effect is strong in the high dose-rate range, and is relatively dose-rate independent at low dose-rates. The results indicate that the curves would level off when the dose-rate is further increased.

For the sake of simplicity, we have neglected thermal drainage during irradiation. This seems reasonable for the reported case of quartz, since irradiation was at room temperature and the measured glow peaks were at ~300°C.

The model investigated is probably an over-simplification of the real physical situation. We have, however, demonstrated that even in this simple case, a dependence upon dose-rate is possible.

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