SUPERLINEAR FILLING OF TRAPS IN CRYSTALS DUE TO COMPETITION DURING IRRADIATION

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The superlinear dose dependence of the filling of traps, as measured by thermoluminescence (TL), which often occurs under β and γ irradiations, has been theoretically investigated. Assuming competition during irradiation of carriers falling into TL traps and into deeper traps, one gets a linear–superlinear–linear–saturation behavior.

1. Introduction

In a number of growth curves of thermoluminescence (TL) intensity as a function of the excitation dose, a superlinear behavior is observed, i.e. if we denote the TL glow intensity by S and the dose by D, this means that dS/dD is growing in a certain dose range, namely, that $d^2S/dD^2 > 0$. Superlinearity has been found in two main forms.

1) Starting from the very low doses of excitation [1].

2) The growth curve starts linearly with dose, becomes superlinear at higher doses and then goes to saturation [2], possibly through a second linear range.

Several explanations to the superlinear effects were given. Chen and Halperin [1] explained the superlinear growth in uv irradiated semiconducting diamonds to be due to the multistage transition of electrons from the valence to the conduction band. Another approach by Cameron and Zimmerman [3] ascribed the superlinear response of LiF to the creation by the ionising irradiation of additional traps or centers in the crystal. Rodine and Land [4] suggested a model which was further investigated [5]. According to this, TL intensity is proportional to both the initial concentration of electrons in traps and holes in centers (rather than the usual case where it is proportional to the smaller of them), due to the effect of an additional trap. Two more works explaining certain kinds of superlinearity are the track interaction model [6] and another [7] which is based on the spatial correlation between charged sites.

Suntharalingam and Cameron [8] suggested a different and apparently more satisfactory way to explain superlinearity. They postulate that the filling of the

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trap giving rise to TL is superlinear due to competition during irradiation by another trap which is of lower concentration but higher trapping probability than the TL trap. Qualitatively, at low doses, the excitation fills both traps linearly. At a certain dose, however, the competing trap comes to saturation, hence more electrons are available to the trap of interest. This causes a faster though linear filling of this trap, the transition region from one linear range to the other would, however, be superlinear. Such competition superlinearity in TL bears some resemblance to superlinear photoluminescence [9] and superlinearity in photoconductivity [10].

2. Mathematical analysis

We assume an energy level diagram as depicted in fig. 1. N_1 is the trap giving rise to TL and N_2 is the competitor; N_1 and N_2 will be used to denote the concentrations of these traps. At time t, n_1 and n_2 represent the concentrations of electrons in these traps and m, the concentration of holes in luminescence centers. We assume that at the end of the irradiation, $n_1 < m$ and are interested in the dependence of n_1 on the dose.

As shown below, we shall derive an expression with D as a function of n_1, therefore it is easier to study the sign of d^2D/dn_1^2 than that of d^2n_1/dD^2. Since, however,

$$d^2n_1/dD^2 = -(d^2D/dn_1^2)/(dD/dn_1)^3 \tag{1}$$

and since $dD/dn_1 > 0$, the desired condition for superlinearity $d^2n_1/dD^2 > 0$ can be written as $d^2D/dn_1^2 < 0$.

We shall start with the case of electrons raised by the irradiation from the valence to the conduction band and fall into either N_1 or N_2. The equations governing the process are

$$dn_1/dt = A_1(N_1 - n_1)n_c, \tag{2}$$

$$dn_2/dt = A_2(N_2 - n_2)n_c, \tag{3}$$

$$dn_c/dt = X - dn_1/dt - dn_2/dt, \tag{4}$$

where n_c is the concentration of electrons in the conduction band. A_1 and A_2 are the transition probabilities into N_1 and N_2, respectively, and X is the rate of creation of electron–hole pairs. By eliminating n_c from eq. (2) and (3), a direct relation is found between $n_2(t)$ and $n_1(t)$. Substituting in eq. (4) and assuming
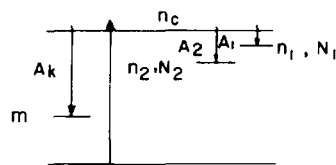

Fig. 1. Energy levels in the forbidden gap.

$|dn_c/dt| \ll |dn/dt|$ one gets by integration

$$D = \int_0^t X \, dt' = n_1 - n_{10} + N_2 - n_{20} - (N_2 - n_{20}) \left(\frac{N_1 - n_1}{N_1 - n_{10}}\right)^{A_2/A_1}. \tag{5}$$

The second derivative of D with respect to n_1 is

$$d^2D/dn_1^2 = (-A_2/A_1)(A_2/A_1 - 1) \frac{N_2 - n_{20}}{(N_1 - n_{10})^2} \left(\frac{N_1 - n_1}{N_1 - n_{10}}\right)^{A_2/A_1 - 2} \tag{6}$$

and hence, the condition for this to be negative is $A_2 > A_1$. This condition is, explicitly, that the probability of the competitor is larger than that of the trap directly involved in the TL process. Eq. (5) is expected to yield superlinearity for $A_2 > A_1$ in the whole range of $0 \le n_1 \le N_1$ whereas actually, two linear regions of growth may be observed with superlinearity in between (and, saturation at high doses). The point is that although d^2D/dn_1^2 is negative, its absolute value may be very small, so that it is practically nil. The qualitative description of the model given in the introduction shows how linearity, superlinearity, linearity and ultimately saturation can be expected in the growth of n_1 with dose. Numerically, regions of linearity and superlinearity also result from computer calculations on eq. (5). The fact that no saturation is observed, is related to the assumption that recombination during excitation is negligible. This may well be the case at the low dose range including the suplinearity region, but it ceases to be so at higher doses. The addition to the model will be described now.

Aitken et. al. [11] considered a model in which eq. (4) is replaced by

$$dn_c/dt = X - dn_1/dt - dn_2/dt - A_k n_c p, \tag{7}$$

where p is the concentration of holes in the valence band and A_k the band to band transition probability. They assumed

$$p = n_1 + n_2 + n_c, \tag{8}$$

which implies the possibility of accumulating holes in the valence band which seems rather unlikely. This picture can slightly be modified if we denote by p the concentration of holes in the center and assume that A_k is the recombination probability conduction band to center. Eq. (8) will still hold with p_v, the concentration of holes in the valence band added to the left hand side. Both n_c and p_v can be neglected as compared to the other terms in eq. (8). The equations of Aitken et. al. remain the same with a slight change in the meaning. We still neglect the direct band to band recombination which is usually known to be small. The solution of the set of eq. (2), (3) and (7) is

$$D = N_2 \frac{A_k - A_2}{A_2} \left[\left(1 - \frac{n_1}{N_1}\right)^{A_2/A_1} - 1\right] + \frac{A_1 - A_k}{A_1} n_1 - N_1 \frac{A_k}{A_1} \left(\frac{N_2}{N_1} + 1\right) \ln\left(1 - \frac{n_1}{N_1}\right), \tag{9}$$

when for the sake of simplicity, we assumed $n_{10} = n_{20} = 0$.

By finding the first derivative one can see that dD/dn_1 goes to infinity when $n_1 \approx N_1$, which implies $dn_1/dD \approx 0$ i.e. approach to saturation. For the low dose

range, it can easily be shown that

$$d^2D/dn_1^2|_{n_1=0} \approx \frac{1}{A_1 n_1} [A_k N_2 A_2/(A_1 N_1) + N_2 A_2/N_1 + A_k - N_2 A_2^2/(A_1 N_1)]. \tag{10}$$

A necessary and sufficient condition for superlinearity at low dose is that the right hand side of eq. (10) is negative which implies the three separate necessary conditions

$$A_2 > A_1; \quad A_2 > A_k; \quad N_2 A_2^2/(N_1 A_1) > A_k. \tag{11}$$

The second and third conditions are automatically fulfilled for small enough values of A_k and the first is the one mentioned above for the more restricted case.

Eq. (5) and (9) have numerically been solved to yield $n_1 = n_1(D)$. As an illustration, fig. 2 depicts the results of n_1 as a function of D for a chosen set of parameters as calculated from eq. (9). The initial linear range, the superlinear region and the second linearity before saturation can be seen.
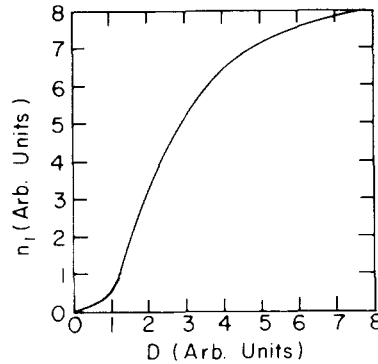

Fig. 2. Growth curve as numerically calculated for $N_1 = N_2 = 10^{17}$ cm^{-3}, $n_{10} = n_{20} = 0$, $A_1 = A_k$, $A_2/A_1 = 30$.

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