

Superlinear growth of thermoluminescence due to competition during irradiation

Abstract

The superlinear dependence of thermoluminescence intensity on the dose of excitation which often occurs under β and γ irradiations is theoretically investigated, the superlinearity being due to competition during irradiation of carriers falling into TL traps and into deeper traps. The conditions for various growth behaviours are analytically investigated, namely, the relations between the trapping parameters leading to superlinearity, linearity or sublinearity. Also given are the conditions for having superlinearity starting at low doses versus those leading to a linear-superlinear-saturation behaviour. Computer calculations have been performed to show various dependences on dose for chosen sets of parameters.

Introduction

The growth of thermoluminescence (TL) with excitation dose is frequently found to exhibit superlinearity. In general terms, this means that the glow intensity grows faster than the exciting dose. More precisely, if we denote the TL glow intensity by S and the dose by D , this means that dS/dD increases over a certain dose range, or in other words, that $d^2S/dD^2 > 0$ in this particular range. It should be noted here that S can be either the maximum intensity or the total area under the curve since it has been shown by Kristianpoller et al. (1974) that to a good approximation, these two quantities are proportional, even in complex cases.

Superlinear behaviour can be roughly divided into two categories, one in which superlinearity starts at the lowest measurable level (Halperin and Chen, 1966) and the other, which seems to be more common, in particular when the excitation is by nuclear irradiation in which the growth starts at a linear rate and then becomes superlinear (for example, Cameron et al., 1968). In both cases, the growth curve goes to saturation at high doses. In the former case, superlinearity can be described by $S \propto D^k$ with $k > 1$ (Chen and Halperin, 1966), however, such a definition is not applicable to the latter; only the definition $d^2S/dD^2 > 0$ is general.

Various workers have suggested explanations for the observed forms of superlinearity. Halperin and Chen (1966) explained the superlinear TL growth in UV irradiated diamonds as due to the multistage transition of electrons from the valence to the conduction band prior to being trapped in electron luminescence centres. This accounted for the observed results of superlinearity starting at the lowest doses such that $S \propto D^k$ (D being the UV light dose). The value of k depended on the excitation wavelength, its maximum value was found to be 3.

Another approach was taken by Cameron and Zimmerman (1966) and Cameron et al. (1968) who ascribed the superlinear response of LiF to the creation by the ionising radiation of additional traps or centres in the crystal.

Rodine and Land (1971) suggested a model in which the TL intensity is proportional to both the initial concentration of electrons in traps and holes in centres due to the presence of an additional postulated trap, the retrapping into which is more efficient during heating, than both retrapping into the initial trap, and recombination. If this is the case and if both initial concentrations are linearly dose dependent, one should expect a D^2 behaviour starting at the lowest doses and up to a point where saturation begins. This model was further investigated by Kristianpoller et al. (1974) who found the conditions leading to superlinearity under these assumptions.

An alternative way of explaining superlinearity is the track interaction model (Claffy et al., 1968; Cameron et al., 1968; Dobson and Midkiff, 1970; Attix, 1975; Larsson and Katz, 1976). A track is taken to be the path of the primary excitation (e.g. energetic electron) and the associated ionization. According to the model, at low doses all tracks are considered to be well separated, therefore the TL response arises from the recombination of electrons and holes within the same track, the result is linear growth of TL with dose. For higher doses, the tracks overlap and a hole released from one track during heating may interact with centres in nearby tracks, this increases recombination probability and hence gives rise to superlinearity.

Suntharalingam and Cameron (1969) suggested a different and seemingly more satisfactory model for superlinearity. They postulate that the filling of the trap (or equivalently, the centre) 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 traps. In order that superlinearity be apparent in the TL response, the superlinearly filled trap must be of lower population than the corresponding centre. 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. It is to be noted that such competition-superlinearity in TL bears some resemblance to superlinear luminescence (Nail et al., 1949) and superlinearity in photoconductivity (Duboc, 1955; Cardon and Bube, 1964; Dussel and Bube, 1966), the difference being mainly that in luminescence and photoconductivity, the superlinear behaviour is of intensity vs. excitation intensity (rather than vs. dose).

A number of investigations dealt further with this possibility of competition during excitation (Aitken et al., 1968; Savikhin, 1972; Zavt and Savikhin, 1974; Nakajima, 1976). A slightly different version was suggested by Israeli et al. (1972), who dealt with the creation of point defects in alkali halides in which competition during excitation took place. In this case, the attenuation of the exciting UV beam was also taken into account.

Finally, a recent paper by Fain and Monnin (1977) which explains superlinearity as being due to spatial correlation between charged sites, should be mentioned.

The main purpose of the present paper is to further investigate, both analytically and numerically, the model suggested by Suntharalingam and Cameron (1968). The conditions under which superlinear behaviour would be expected are investigated as well as the specific conditions for subcategories of superlinearity, namely, that preceded by linearity and that beginning from zero dose.

Analytical approach

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 also be used to denote the concentrations of these traps, n_1 and n_2 represent the concentration of electrons in these traps and m represents the concentration of holes in luminescence centres. We assume that at the end of the irradiation, $n_1 < m$ and therefore, the peak intensity S (or I_m) is proportional to n_1 . We are thus interested in the dependence of n_1 on the dose.

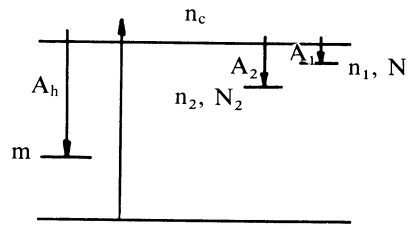


Fig. 1. Energy level scheme with the TL trap N_1 , competing trap N_2 and TL centre m .

The condition to be considered regarding superlinearity is $d^2n_1/dD^2 > 0$; this will be analytically studied. As shown below, we shall derive an expression with D as a function of n_1 , therefore, it is easier to investigate d^2D/dn_1^2 . Since

$$d^2n_1/dD^2 = -(d^2D/dn_1^2) / (dD/dn_1)^3, \quad (1)$$

the condition $d^2n_1/dD^2 > 0$ can now be written as $d^2D/dn_1^2 < 0$ (since $dD/dn_1 > 0$). This will be investigated for various cases.

We shall start with the case in which electrons are raised by the irradiation from the valence band into the conduction band, and fall into either N_1 or N_2 . We thus neglect the possibility of band-to-band or band-to-centre recombination during excitation. The equations governing the process are (see Paige, 1957),

$$dn_1/dt = A_1 (N_1 - n_1) n_c \quad (2)$$

$$dn_2/dt = A_2 (N_2 - n_2) n_c \quad (3)$$

$$dn_c/dt = X - dn_1/dt - dn_2/dt \quad (4)$$

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. It is assumed here that all the created holes are accumulated in the recombination centre m .

Eliminating n_c from (2) and (3) we get

$$\frac{1}{A_1} \frac{dn_1/dt}{N_1 - n_1} = \frac{1}{A_2} \frac{dn_2/dt}{N_2 - n_2} \quad (5)$$

the solution of which yields

$$n_2 = (N_2 - n_{20}) [(N_1 - n_1)/(N_1 - n_{10})]^{A_2/A_1}. \quad (6)$$

From the usual assumption (Halperin and Braner, 1960) that the rate of change of electron concentration in the conduction band is much smaller than the rate of change of concentration in traps, i.e., $|dn_c/dt| \ll |dn_1/dt|$ and $|dn_c/dt| \ll |dn_2/dt|$ eq. (4) becomes

$$X = dn_1/dt + dn_2/dt \quad (4')$$

and by integration

$$D = \int_0^t X dt = n_1 + n_2 - n_{10} - n_{20} \quad (7)$$

where n_{10} and n_{20} are the initial concentrations of electrons in n_1 and n_2 respectively. In the usual case where X is held constant during the excitation, $D = X.t$. Substituting n_2 from (6) to (7) we get

$$D = n_1 - n_{10} + N_2 - n_{20} - (N_2 - n_{20}) [(N_1 - n_1)/(N_1 - n_{10})]^{A_2/A_1} \quad (8)$$

The superlinearity feature can be shown by differentiating eq. (8) twice which gives

$$d^2D/dn_1^2 = -(A_2/A_1) (A_2/A_1 - 1) \frac{(N_2 - n_{20})}{(N_1 - n_{10})^2} \left(\frac{N_1 - n_1}{N_1 - n_{10}} \right)^{A_2/A_1 - 2} \quad (9)$$

The condition for this to be negative is

$$\frac{A_2}{A_1} - 1 > 0, \text{ i.e.} \\ A_2 > A_1 \quad (10)$$

This condition is explicitly that which has been previously stated in words, i.e. that the retrapping probability of the competitor should be larger than that of the trap directly involved in the TL process. We would like to mention here that the filling of n_2 follows the same equation (8) with the subscripts 1 and 2 interchanged and the same is true for eq. (9). This shows immediately that for $A_2 > A_1$, n_2 grows sublinearly, or, more specifically in this case, n_2 saturates.

It is to be noted that eq. (8) is expected to give superlinear dependence for $A_2 > A_1$ in the whole range of $0 \leq n_1 \leq N_1$, whereas in practice, a linear region of growth may be observed and certainly, at high doses saturation will begin. The point is that eq. (9) shows that d^2D/dn_1^2 is negative, but it does not show explicitly the values of d^2D/dn_1^2 . Moreover, since we are interested in $d^2n_1/dD^2 = -(d^2D/dn_1^2)/(dD/dn_1)^3$, we have to take into account the value of the first derivative as well.

The qualitative description of the model given in the introduction shows how linearity, superlinearity, linearity and ultimately saturation in turn can be expected in the growth of n_1 with dose. Numerically, regions of linearity and superlinearity also result from computer calculations on eq. (8). The fact that no saturation is predicted by this equation is related to the assumption that recombination during the excitation is negligible. This may well be the case at the low dose range including the superlinearity region but it probably ceases to be so at higher doses, while saturation is approached. This addition to the model will be described now.

Aitken et al. (1968) considered this model in which eq. (4) is replaced by

$$dn_c/dt = X - dn_1/dt - dn_2/dt - A_k n_c p \quad (11)$$

where p is the concentration of holes in the valence band and A_k the band to band transition probability. In addition, neutrality has to be fulfilled, namely,

$$p = n_1 + n_2 + n_c \quad (12)$$

which implies the possibility of accumulating holes in the valence band. Such an accumulation can be checked in cases where thermally stimulated conductivity (TSC) can be measured, i.e., when one is dealing with crystals rather than with powder samples. In general, however, it seems rather unlikely that such an accumulation occurs. This picture can be slightly modified if we denote by p the concentration of holes in the centres (previously denoted by m) and assume that A_k is the recombination probability, valence band to centre. Equation (12) will read now

$$p + p_v = n_1 + n_2 + n_c \quad (12')$$

where p_v is the concentration of holes in the valence band. Both p_v and n_c can be neglected as compared to the other terms in (12') (Halperin and Braner, 1960). 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 (Pilkuhn, 1976).

The solution as given by Aitken et al. is

$$D = N_2 \frac{A_k - A_2}{A_2} \left[\left(1 - \frac{n_1}{N_1} \right)^{A_2/A_1} - 1 \right] + \frac{A_1 - A_k}{A_1} n_1 - N_1 \frac{A_k}{A_1} \left(\frac{N_2}{N_1} + 1 \right) \ln \left(1 - \frac{n_1}{N_1} \right). \quad (13)$$

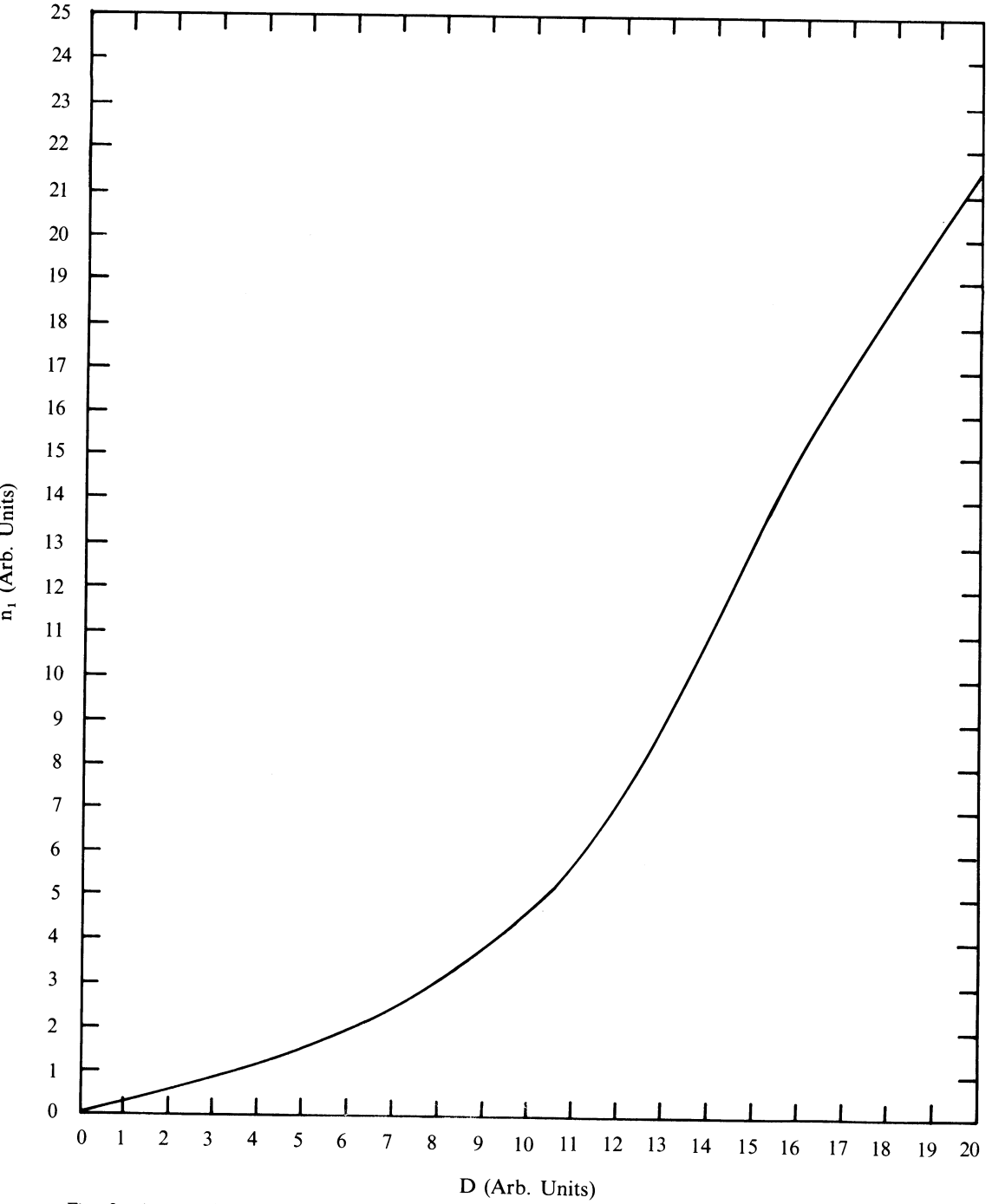


Fig. 2. A typical superlinear dose dependence as calculated by eq. 8. The parameter values are indicated in the text.

For the sake of simplicity, it has now been assumed that $n_{10} = n_{20} = 0$, otherwise n_1 has to be replaced by $n_1 - n_{10}$, N_1 by $N_1 - n_{10}$ and N_2 by $N_2 - n_{20}$.

The first derivative of (13) is

$$\frac{dD}{dn_1} = \frac{-N_2}{N_1} \frac{A_k - A_2}{A_1} \left(1 - \frac{n_1}{N_1}\right)^{A_2/A_1 - 1} + \frac{A_1 - A_k}{A_1} + \frac{A_k}{A_1} \left(\frac{N_2}{N_1} + 1\right) \left(\frac{N_1}{N_1 - n_1}\right). \quad (14)$$

For high values of n_1 , $n_1 \approx N_1$; although the first term in (13) goes to zero (provided that $A_2 > A_1$), the third term tends to infinity, and so does the sum. Therefore, $dn_1/dD \rightarrow 0$ which demonstrates the required saturation behaviour. The second derivative will now be

$$\frac{d^2D}{dn_1^2} = \frac{N_2}{N_1^2} \frac{A_k - A_2}{A_1} \left(\frac{A_2}{A_1} - 1\right) \left(1 - \frac{n_1}{N_1}\right)^{A_2/A_1 - 2} + \frac{A_k}{A_1} \left(\frac{N_2}{N_1} + 1\right) \frac{N_1}{(N_1 - n_1)^2}. \quad (15)$$

Having illustrated that saturation occurs at high doses, we now consider the low dose range for which

$$\left. \frac{d^2D}{dn_1^2} \right|_{n_1 \approx 0} \approx \frac{1}{A_1 N_1} [A_k N_2 A_2 / (A_1 N_1) + N_2 A_2 / N_1 + A_k - N_2 A_2^2 / (A_1 N_1)]. \quad (16)$$

For $A_k = 0$, this reduces to

$$\left. \frac{d^2D}{dn_1^2} \right|_{n_1 \approx 0} \approx N_2 A_2 (A_1 - A_2) / (A_1^2 N_1^2) \quad (17)$$

which will be negative if $A_2 > A_1$; this is the condition (10) again.

From eq. (16), a necessary and sufficient condition for superlinearity at low doses when $A_k > 0$ is

$$N_2 A_2^2 / (A_1 N_1) > A_k + N_2 A_2 / N_1 + A_k N_2 A_2 / (A_1 N_1). \quad (18)$$

Looking at each term on the right hand side separately we see that necessary conditions for superlinearity are

$$\begin{aligned} A_2 &> A_k \\ N_2 A_2^2 / (N_1 A_1) &> A_k \\ A_2 &> A_1 \end{aligned} \quad (19)$$

The first two are automatically fulfilled for very small values of A_k and the third is condition (10) again which is a necessary and sufficient condition when A_k is zero. It is obvious that when the condition (18) is reversed, which can easily occur for a high value of A_k , the dependence of n_1 on the dose will be sublinear starting at the lowest doses.

It is to be noted that the analysis would be identical if one assumes a filling under competition conditions of a recombination centre, provided that the concentration of holes in the luminescence centre is smaller than the concentration of electrons in the trap associated with the TL process, i.e. in the case where TL intensity depends on the centre population. The two "mirror image" cases

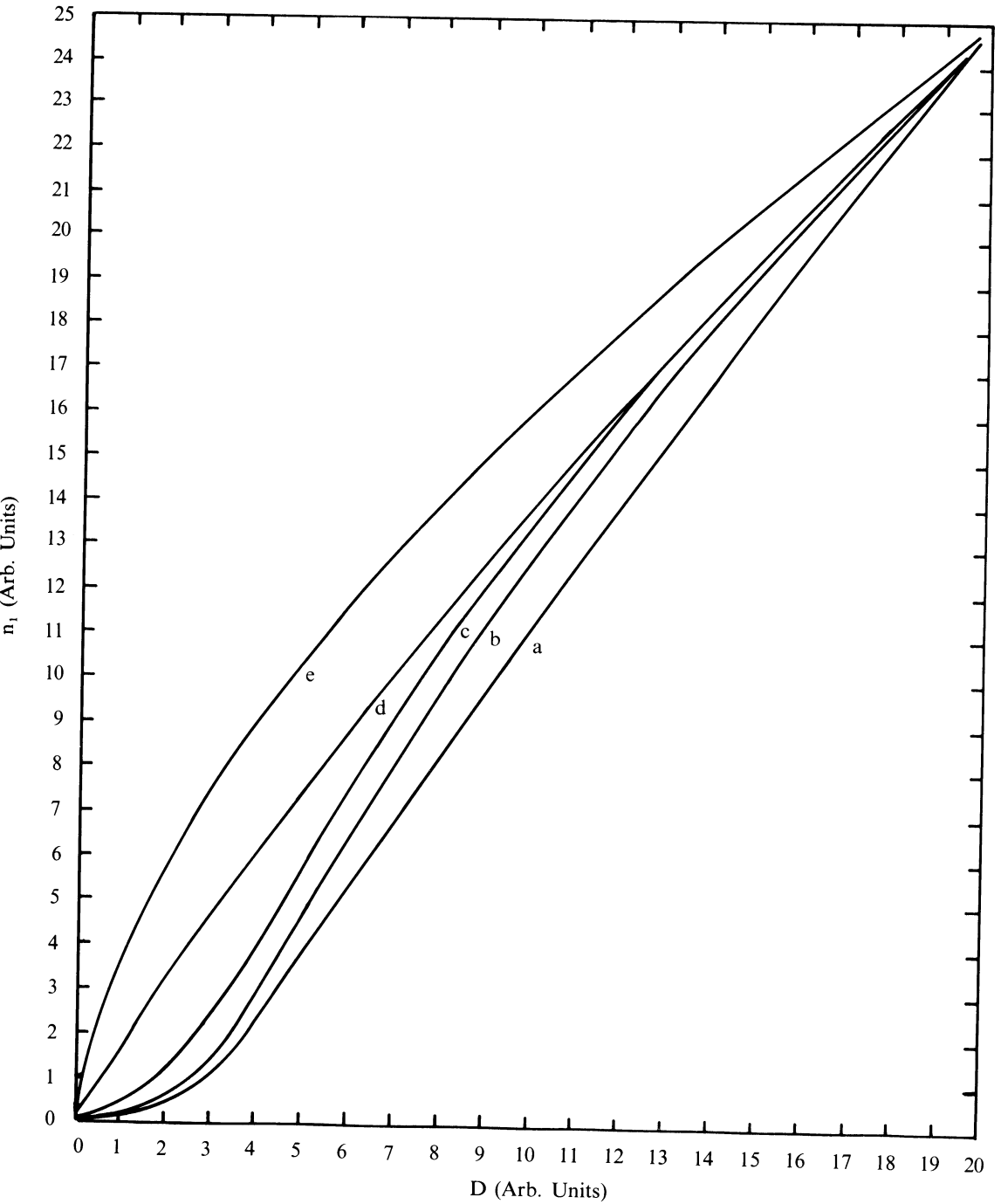


Fig. 3. Dose dependence as calculated from eq. 13 for different values of A_k . The gradual change from superlinearity to sublinearity is shown.

are also possible, namely, the cases dealing with hole traps and electron centres and their filling by irradiation.

We shall conclude this mathematical section by commenting on the usual way of plotting the TL intensity as a function of the dose when the behaviour is superlinear. In the cases where the superlinearity starts at the low doses, the behaviour can be approximated as $S \propto D^k$ for a certain dose range and a log-log scale should yield a straight line the slope of which is k . If, on the other hand, the dependence is different, for example, if it is linear first and becomes superlinear at higher doses, the log-log plot does not yield any useful information. If, in general, we write $S = f(D)$, we have

$$d \log f(D) / d \log D = D f'(D) / f(D). \quad (20)$$

For $f(D) \cong D^k$ this gives the desired (effective) slope of k , but even for a simple combined function such as

$$S = aD + bD^2 \quad (21)$$

we get the slope

$$d \log f(D) / d \log D = (a + 2bD) / (a + bD) \quad (22)$$

which is dose-dependent unless $a \ll bD$ and, therefore, is not very useful in depicting the behaviour. Even in a simpler case of an additive constant

$$S = \alpha + \beta D^2 \quad (23)$$

we have

$$d \log f(D) / d \log D = 2\beta D^2 / (\alpha + \beta D^2) \quad (24)$$

and again the slope on the log-log scale is dose-dependent unless $\alpha \ll \beta D^2$.

Numerical Method and Results

Equations (8) and (13) could be solved for a given set of parameters by choosing values of n_1 and directly calculating the corresponding value of D . Since one is interested in n_1 as a function of D for an appropriate dose range, it is more convenient to solve the transcendental equation in n_1 for a given value of D , change D in suitable steps and repeat the calculation for each value of D . The solution has been carried out using the Newton-Raphson method; some of the results are shown in the following figures. Since the superlinear feature is related to the competition between N_1 and N_2 , we start with a solution of eq. (8) and introduce the "interfering" effects of $A_k > 0$ only later through the solution of eq. (13). Figure 2 depicts the results of eq. (8) for $A_2/A_1 = 100$, $N_1 - n_{10} \cong 10^{18} \text{ cm}^{-3}$ and $N_2 - n_{20} \cong 10^{17} \text{ cm}^{-3}$. It is clearly seen that the curve starts linearly, becomes superlinear and then enters a second linear range before showing a slight tendency to saturate.

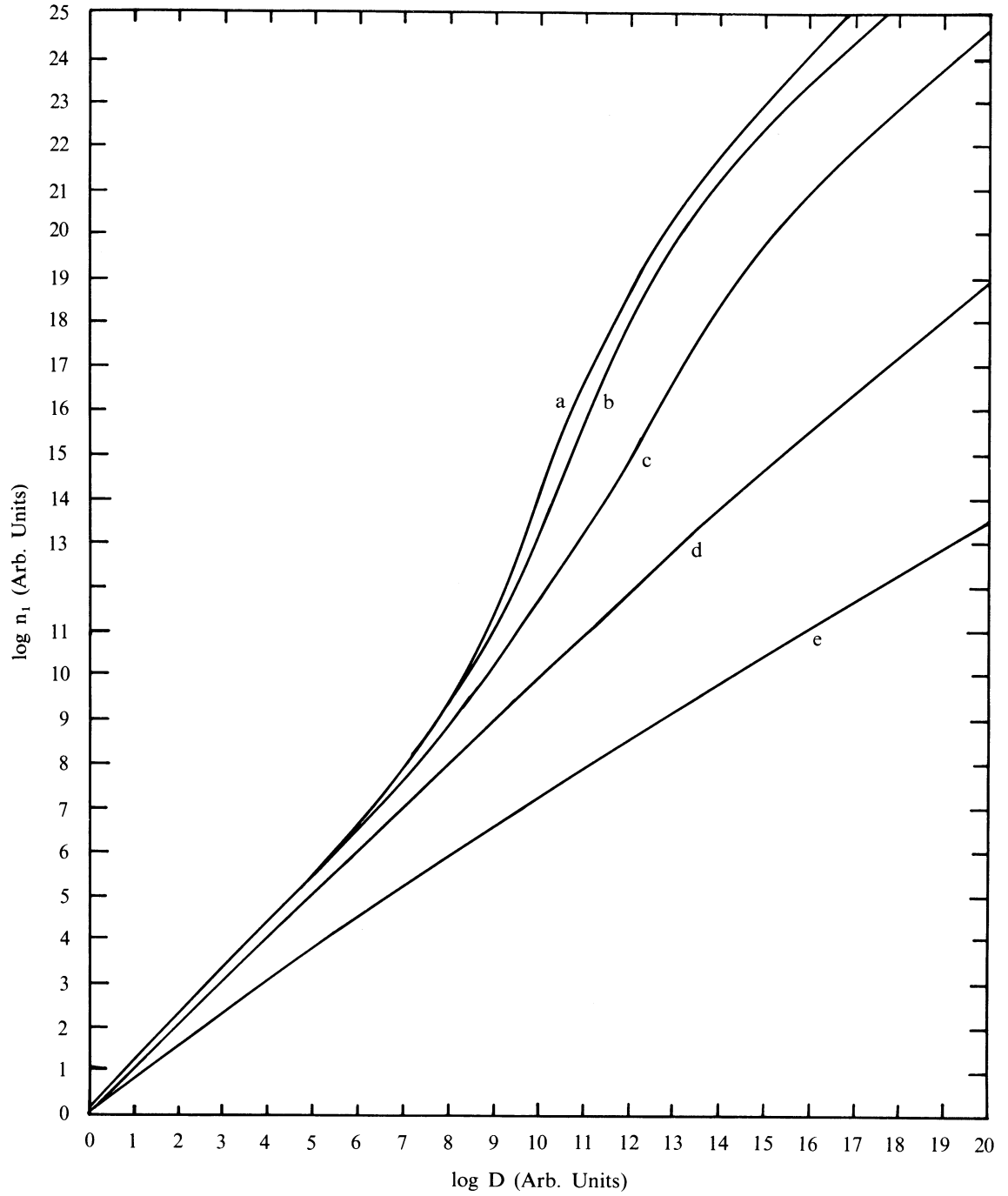


Fig. 4. The results of fig. 3 plotted on a log-log scale.

Figure 3 shows the change of behaviour for a given set of parameters when the magnitude of the recombination during irradiation, A_k , is changing from curve to curve. Since we are mainly interested in superlinearity, we only take the low dose region, namely far enough from saturation, in Fig. 3 (as well as in Figs. 4 and 5); the high-dose saturation will be shown in Fig. 6. The parameters chosen in Fig. 3 are $N_1 = 10^{18} \text{ cm}^{-3}$, $N_2 = 10^{17} \text{ cm}^{-3}$, $A_1 = 1 \text{ cm}^3 \text{ sec}^{-1}$ and $A_2 = 100 \text{ cm}^3 \text{ sec}^{-1}$. In curve a, $A_k = 0.1$; the superlinear behaviour is seen at low doses, the curve being practically linear thereafter. With increasing values of A_k in b, c..., the initial superlinearity becomes less distinct until at $A_k = 100$ (curve d), the low-dose behaviour is linear with some tendency to saturation at higher doses. At an even bigger recombination, $A_k = 1000 \text{ cm}^3 \text{ sec}^{-1}$ (curve e) the sublinearity starts at the lowest dose. In Fig. 4, the same results are shown on a log-log scale. The sublinear feature of curve e and the linearity of a are evident. The fact that a, b and c are only slightly superlinear at low doses proves that, although it is not clearly seen in Fig. 3, there is an almost linear narrow range at the beginning of the superlinear region. Curve d is seen to be nearly of slope 1.

In Fig. 5 are shown results where the superlinearity is seen to prevail from the very lowest doses. For this we chose $N_1 = 10^{18} \text{ cm}^{-3}$, $N_2 = 10^{16} \text{ cm}^{-3}$, $A_1 = 1 \text{ cm}^3 \text{ sec}^{-1}$, $A_2 = 300 \text{ cm}^3 \text{ sec}^{-1}$. On the log-log scale we see (curve a) that for $A_k = 0.1$, n_1 is proportional to $D^{1.58}$ whereas for $A_k = 100 \text{ cm}^3 \text{ sec}^{-1}$, $n_1 \propto D^{1.46}$. At higher doses, the growth is closer to linear in curve a and tends to saturation in curve b. A higher initial slope on the log-log scale can easily be found by taking bigger A_2/A_1 ratios and keeping A_k very small. Thus we could get $k = 2$ for $A_2/A_1 = 500$ and $A_k = 0$.

In curve a of fig. 6, the whole range of possibilities is shown, linearity-superlinearity, a second linear range and tendency toward total saturation. This was computed for $N_1 = N_2 = 10^{17} \text{ cm}^{-3}$, $A_1 = A_k = 1 \text{ cm}^3 \text{ sec}^{-1}$, $A_2 = 30 \text{ cm}^3 \text{ sec}^{-1}$ and $n_{10} = n_{20} = 0$. Curve b shows the same results on a log-log scale. The first linear region is seen much more clearly but, as discussed above, the slope on the log-log scale does not yield the effective power of dependence on dose. The second linear region is not seen either and it looks as if the response goes from superlinearity directly to saturation.

A dose dependence in which there is a long linear growth before the superlinear region can be obtained by choosing N_2 much bigger in the above examples ($N_2 > N_1$, for example). An appropriate choice of the parameters can also bring about a linear-superlinear-saturation behaviour, i.e., that the second linear region is very narrow.

Discussion

Following Aitken et al. (1968) we have investigated various dependences of TL intensity when the TL traps are filled under competition conditions. Defining generally a region of superlinearity as one in which $d^2S/dD^2 > 0$ where S is the TL intensity, we have shown analytically that if recombination into the centres

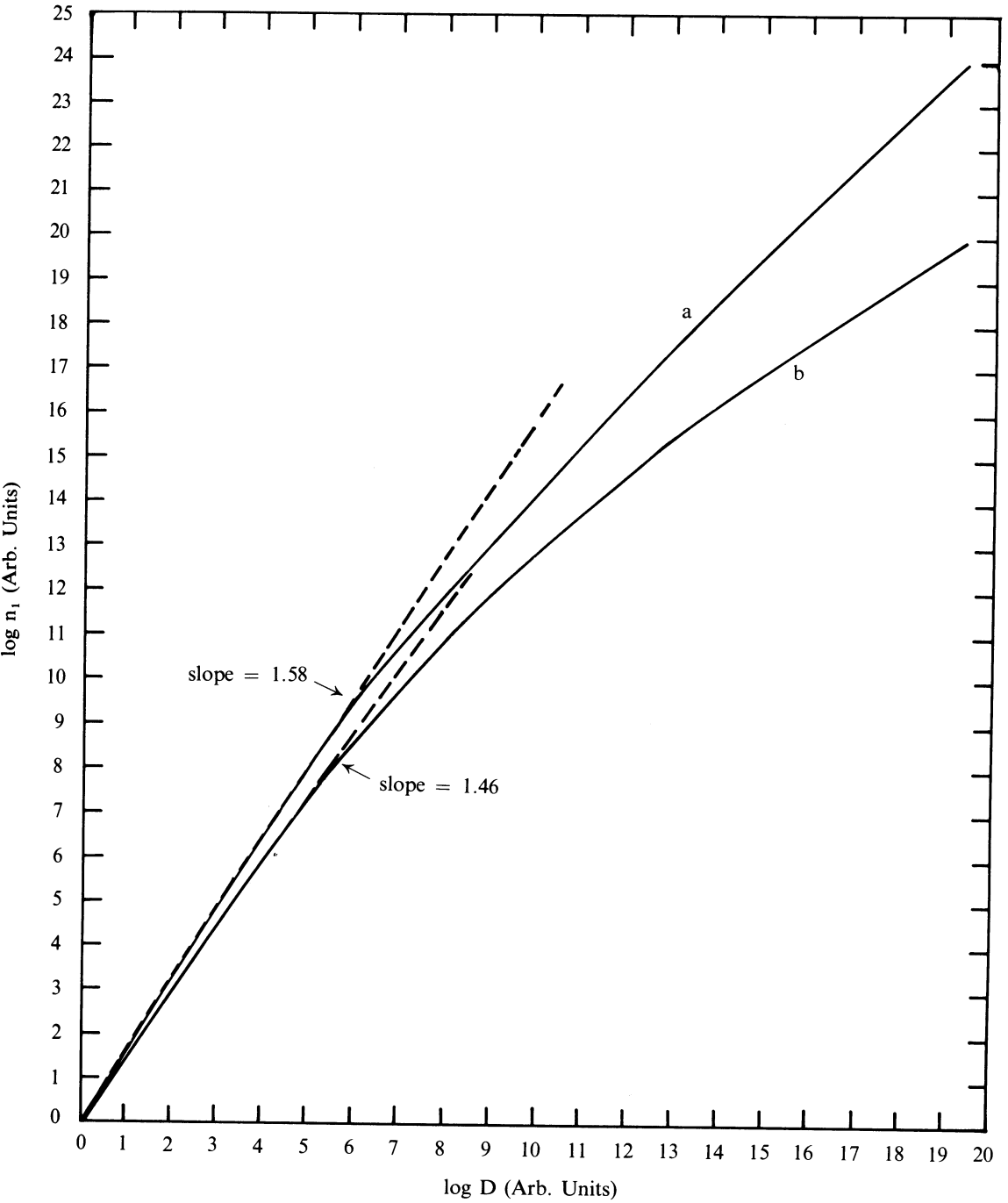


Fig. 5. Superlinear dependence starting at the lowest doses as shown on a log-log scale. The parameters are indicated in the text.

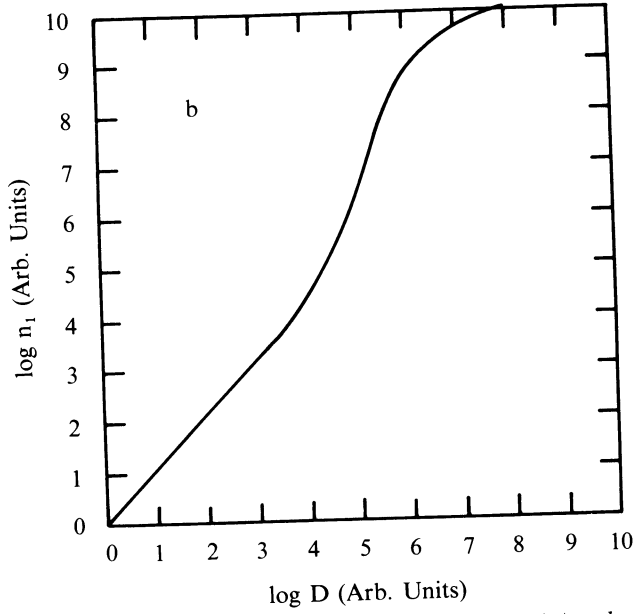
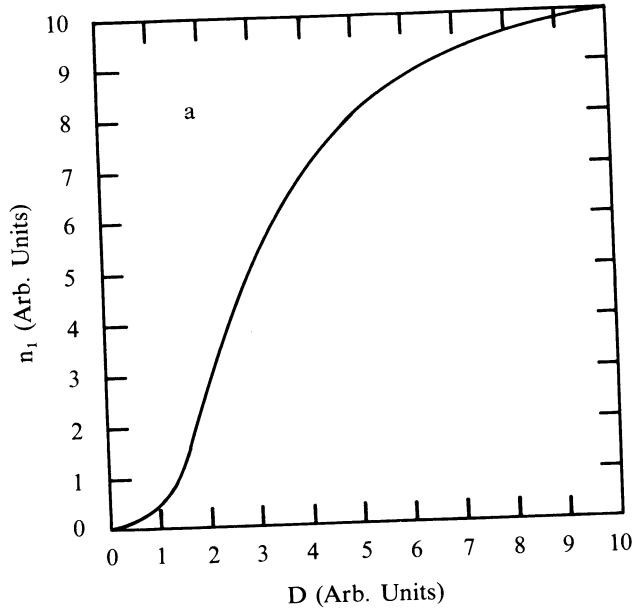


Fig. 6. The linear-superlinear-linear-saturation dependence on a linear (a) and a log-log (b) scale. The parameters are given in the text.

during excitation is negligible ($A_k \approx 0$), then superlinearity exists throughout the growth curve when $A_2 > A_1$. The superlinearity is such, however, that at the lower and higher doses, the dependence is practically linear, with a superlinear range in between.

Although we have assumed a very small recombination probability during irradiation whereas, obviously, during read-out this probability is of great importance, it must be remembered that during heating, the competitor is highly populated, therefore retrapping into it is negligible. It is implicit in the model that N_2 is not emptied prior to N_1 , otherwise its population, n_1 , would not be described by equation 8 or 13. High retrapping by N_1 (relative to recombination) is allowed and is the explanation for the occurrence of a second order peak (Halperin and Braner, 1960).

Allowing $A_k > 0$ reduces the amount of superlinearity and for very high A_k , reduces it to be linear and even sublinear. At high doses, the introduction of A_k , causes a rather early saturation effect (i.e. when n_1 is still smaller than N_1). The expected behaviour is therefore linear-superlinear-linear-saturation. Any of these ranges can, however, shrink or disappear by the appropriate choice of parameters. Thus, superlinear-linear-saturation behaviour which has been reported before (Aitken et al., 1974) as well as linear-superlinear-saturation dependence can be accounted for. For relatively low values of A_k , the dose dependence is shown analytically (and found numerically) to be sublinear starting at the very low doses. Such behaviour has been observed experimentally.

As mentioned above, the superlinear behaviour starts at the lowest doses if $N_2 - n_{20}$ is small enough. This is the case if the concentration of the competitor trap (N_2) is small or if for a relatively large N_2 , many of the traps are filled (by n_{20}) so that $N_2 - n_{20}$ is small. Actually, it is only $N_2 - n_{20}$ which appears in eq. (8) and that is why only this combined parameter was used in the computation. This means that the first linear portion should not be seen if the competitor is rather close to saturation. The situation is similar concerning eq. (13), where we assumed $n_{10} = n_{20} = 0$.

REFERENCES

- AITKEN, M. J., THOMPSON, J., and FLEMING, S. J., 1968, in *Proc. 2nd Int. Cong. Lumin. Dosimetry*, Gatlinburg, Tenn., 364, U.S. Atomic Energy Comm. CONF-680920.
- AITKEN, M. J., HUXTABLE, J., WINTLE, A. G., and BOWMAN, S. G. E., 1974, in *IVth Int. Conf. Lumin. Dosim.*, p. 105.
- ATTIX, F. H., 1975, in *J. Appl. Phys.*, 46, p. 81.
- BOWMAN, S. G. E., 1975, in *Archaeometry*, 17, p. 129.
- CAMERON, J. R., SUNTHARALINGAM, N., and KENNEY, G. N., 1968, *Thermoluminescent Dosimetry (TLD)*. The University of Wisconsin Press, Madison, Milwaukee and London.
- CAMERON, J. R., SUNTHARALINGAM, N., WILSON, C. R., and WATANABE, S., 1968, in *Proc. 2nd Int. Conf. Lumin. Dosimetry*, 332, Gatlinburg, Tenn., U.S. Atomic Energy Comm. CONF 680920.
- CAMERON, J. R., and ZIMMERMAN, D. W., 1966, USAEC Rep. COO-1105-113.

- CARDON, F., and BUBE, R. H., 1964, in *J. Appl. Phys.*, 35, p. 3344.
- CHEN, R., and HALPERIN, A., 1966, in *Proc. Int. Conf. Lumin.*, Budapest, p. 1414.
- CLAFFY, E. W., KLUICK, C. C., and ATTIX, F. H., 1968, in *Proc. 2nd Int. Conf. Lumin. Dosimetry*, U.S. Atomic Energy Comm. CONF-680920, Gatlinburg, Tenn., p. 302.
- DOBSON, Jr. P. N., and MIDKIFF, A. A., 1970, in *Health Phys.*, 18, p. 573.
- DUBOC, C. A., 1955, in *Brit. J. Appl. Phys.*, 6, Suppl. 4, S 107.
- DUSSEL, G. A., and BUBE, R. H., 1966, in *J. Appl. Phys.*, 37, p. 13.
- FAÏN, J., and MONNIN, M., 1977, in *J. Electrostatics*, 3, p. 289.
- FLEMING, S. J., 1975, in *Archaeometry*, 17, p. 2.
- HALPERIN, A., and CHEN, R., 1966, in *Phys. Rev.*, 148, p. 839.
- ISRAELI, M., KRISTIANPOLLER, N., and CHEN, R., 1972, in *Phys. Rev.*, 6, p. 4861.
- KRISTIANPOLLER, N., CHEN, R., and ISRAELI, M., 1974, in *J. Phys. D. Appl. Phys.*, 7, p. 1063.
- LARSSON, L., and KATZ, R., 1976, in *Nuclear Inst. and Methods*, 138, p. 631.
- NAIL, N. R., URBACH, R., and PERLMAN, D. D., 1949, in *J. Opt. Soc. Amer.*, 39, p. 690.
- NAKAJIMA, T., 1976, in *Japan. J. Appl. Phys.*, 15, p. 1179.
- PAIGE, E. G. S., 1957, in *Phil. Mag.*, 2, p. 864.
- PILKUHN, M. H., 1976, in *Proc. 13th Int. Conf. Phys.*, B4, Semicond. P.61.
- RODINE, E. T., and LAND, P. L., 1971, in *Phys. Rev.*, B4, p. 2701.
- SAVIKHIN, F. A., 1972, in *J. Appl. Spect.*, 17, p. 889.
- SUNTHARALINGAM, N., and CAMERON, J. R., 1969, in *Phys. Med. Biol.*, 14, p. 397.
- ZAVT, G. S., and SAVIKHIN, F. A., 1974, in *Izvest. Akad. Nauk. SSR, Ser. Fiz.*, 38, p. 190.
- ZIMMERMAN, D. W., 1971, in *Archaeometry*, 13, p. 29.

R. CHEN* and S. G. E. BOWMAN**

Research Laboratory for Archaeology and the History of Art,
University of Oxford,
6, Keble Road, Oxford

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

** Present address : Research Laboratory, The British Museum, London.