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## CHARACTERIZATION OF NONLINEARITIES IN THE DOSE DEPENDENCE OF THERMOLUMINESCENCE

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**Abstract**—Nonlinearities often occur in the dose dependence of thermoluminescence (TL). These include sublinearity, usually when there is an approach to saturation in the dose dependence, as well as supralinearity, also termed superlinearity in the literature. Different researchers in the field have viewed the effect of supralinearity/superlinearity from two somewhat different points of view. One point of view has to do with the rate of change with dose of the dose dependence function. 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 supra(super)linearity occurs following an initial linear dose range, or prior to such a linear range. In the present work we propose quantitative methods to characterize these nonlinearities. We suggest the use of two different nonlinearity indices, depending upon how one wishes to describe the nonlinearity. We propose use of the term “supralinearity index”,  $f(D)$ , in cases where the feature of interest is the deviation from linearity, namely, when the correction in extrapolation is the main issue. We propose the term “superlinearity index”,  $g(D)$ , in dose ranges where the growth is “more than linear” and when extrapolation is not the main issue. We mathematically define each of these indices and give examples of their use for different dose dependencies.

### 1. INTRODUCTION

PERHAPS the most significant feature of thermoluminescence (TL) when applied to radiation dosimetry and to dating of archaeological or geological samples, is the dose dependence. For these applications, the desired behavior of the material is a linear dose dependence over as broad a dose range as possible. At higher doses, the common behavior is that of an approach to saturation, which is usually referred to as a sublinear dose dependence. In many materials, however, ranges of supralinearity, also termed superlinearity, occur. Beside the linguistic ambiguity, no unique definition of supralinearity, or superlinearity, is accepted in the literature beyond the general feeling that in such a range the TL intensity (measured either as maximum peak intensity or total area) grows “more than linearly” with the dose.

Nonlinear growth of TL as a function of absorbed dose has been discussed in some detail in a book by Cameron *et al.* (1968). The discussion was based on studies published by Cameron’s group: Wagner and Cameron (1966) and Suntharalingam and Cameron (1967). In fact, a previous paper by Cameron *et al.* (1964) shows a slight “more-than-linear” growth (their Fig. 4) in the dose dependence of LiF powder; however, neither of the terms “supralinearity” or “superlinearity” were used at that time. Initial work was concentrated on the main TL dosimeter

in use during that period, namely the TLD-100 (LiF:Mg,Ti). Several other researchers (e.g. Zimmerman, 1971) reported that the nonlinear range follows an initial linear region. Zimmerman (1972) reported similar results in CaF<sub>2</sub>. The characteristic of this type of dose behavior (for  $\beta$  or  $\gamma$  irradiation) is that linearity is observed up to a certain absorbed dose (for TLD-100 this is  $\sim 100$  Gy) and this is then followed by a range of nonlinearity (i.e. increasing slope) which in turn is followed by a transition to a sublinear range and an approach to saturation (see Fig. 1). Gorbics *et al.* (1973) plotted the TL light sum response per roentgen, normalized to 1.00 at 1000 R. This plot showed the nonlinear part by having the relative response per roentgen larger than unity above 1000 R. Similar plots to those obtained by Gorbics *et al.* were given more recently by Mische and McKeever (1989) for a variety of LiF samples, doped with different amounts of Mg, and some examples are shown in Fig. 2.

A first theoretical account of this type of behavior, termed “supralinearity” by many authors (e.g. Gorbics *et al.*, 1973; Horowitz, 1984a, 1984b, 1990; McKeever, 1985; Mische and McKeever, 1989) was given by Cameron and Zimmerman (1965). They proposed a model in which additional traps were created by the radiation causing the TL response to grow more rapidly than linear. A further theoretical work, with better agreement with the experimental

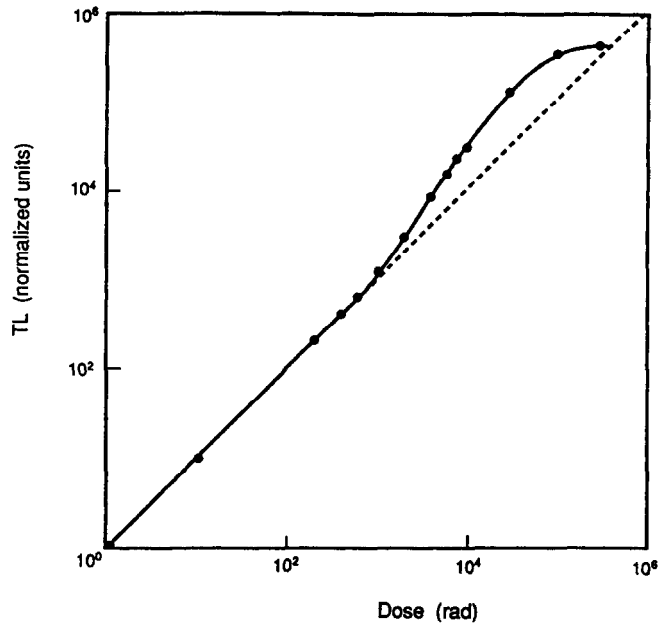


FIG. 1. Supralinearity and superlinearity in the TLD-100 (after Zimmerman, 1971). In the low dose, linear region  $g(D) = f(D) = 1$  (see text for definitions); in the nonlinear, high dose region, both  $g(D)$  and  $f(D)$  start off greater than 1. At the highest doses, however,  $g(D)$  becomes  $\leq 1$ , but still  $f(D) > 1$ . Thus, a dose response may be supralinear, but not superlinear, over certain dose ranges.

results, was given by Zimmerman and Cameron (1968). These authors presented a model which had to do with filling existing traps only (i.e. no trap creation), in the presence of a nonradiative competitor which is active during the excitation. Further

calculations along the same lines were later given by Chen and Bowman (1978).

Another kind of nonlinear growth was reported by Halperin and Chen (1966). These authors used the term "superlinearity" to describe the dose dependence of TL in semiconducting diamonds irradiated by UV light. In this case, for a range of wavelengths between 300 and 400 nm, the dose dependence was strongly nonlinear as of the lowest measurable doses  $D$  of UV irradiation without any linear region being observed, and could be described as being proportional to  $D^k$ . The index  $k$  was found to be between 2 and 3 (depending on the excitation wavelength) up to a point where saturation effects occurred. The authors plotted their results on a log-log scale and superlinearity was revealed by having, in a certain dose range, a straight line with a slope larger than unity (Fig. 3). The authors explained their results to be due to a multistage transition of charge carriers during the excitation period.

Further results of the same nature were later found by Rodine and Land (1971) who reported a dose dependence of  $\sim D^2$  in one peak out of a series of TL peaks in UV-irradiated  $\text{ThO}_2$ . They suggested a model which was related to the occurrence of competition during the heating phase. This model was later developed by Kristianpoller *et al.* (1974) and the same theory was later utilized by Chen *et al.* (1988) to explain their findings of strong superlinearity (up to  $D^3$ ) in the  $110^\circ\text{C}$  peak in synthetic quartz under  $\beta$  irradiation (see also Chen and Fogel (1993)). Continuing this line of discussion, Mische and McKeever (1989) demonstrated that the previously discussed

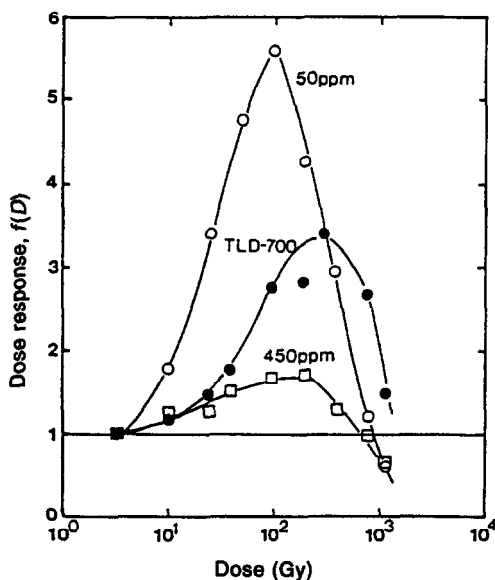


FIG. 2. Plot of  $f(D)$  vs  $D$  for various samples of LiF:Mg (after Mische and McKeever, 1989).  $f(D)$  is termed the "dose response" here, but was defined by Mische and McKeever according to equation (8). The samples used for these data were TLD-700 and LiF:Mg, doped with either 50 or 450 ppm Mg.

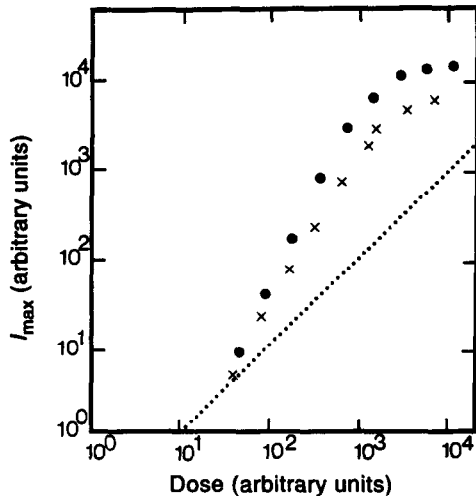


FIG. 3. Superlinearity of the 260 K peak in two samples of semiconducting diamond (redrawn from Halperin and Chen (1966)).  $I_{\max}$  is the height of the 260 K peak and the dotted line shows the line of linearity. There is no obvious linear region for the curves and therefore  $g(D) > 1$ , but  $f(D)$  cannot be defined. Thus, a dose response may be superlinear, but not supralinear.

nonlinear dose dependence of the LiF TLD-100 (see Fig. 1) was also due to competition during the heating stage, with the linear region preceding the nonlinear range being the result of spatial correlation between the relevant traps and centers (see also McKeever, 1990a). The principle of competition during heating is also central to the development of track interaction theory as a means of understanding the nonlinearities in the dose response of materials irradiated with heavy charged particles (Moscovitch and Horowitz, 1988; Horowitz, 1981, 1990; Horowitz and Rosenkrantz, 1990).

Yet another kind of nonlinearity was reported for the first time by Tite (1966) (see also the book by Aitken (1974)) in pottery samples, consisting mainly of quartz. Here, in the range of up to  $\sim 2$  Gy, the dose dependence was nonlinear and this behavior was followed by a broad range of linear dependence on the absorbed  $\beta$  or  $\gamma$  dose. The amount of supralinearity was defined by the measured parameter  $\Delta$  (see Fig. 4) which represents the so-called "supralinearity correction", i.e. the intercept of the linear portion of the curve with the dose axis.

It is clear from the above discussion that nonlinearities in TL growth curves manifest themselves in a variety of ways. However, there appears to be no agreed-upon, formalized procedure for describing these nonlinearities. Attempts to quantify the effects observed include calculations of TL/unit dose (e.g. Gorbics *et al.*, 1973), taking the slope of a log TL versus log  $D$  plot (e.g. Halperin and Chen, 1966; Rodine and Land, 1971), calculations of the factor by which the TL is enhanced at high dose compared to what one would get from an extrapolation of the low dose behavior (e.g. Mische and McKeever, 1989), and

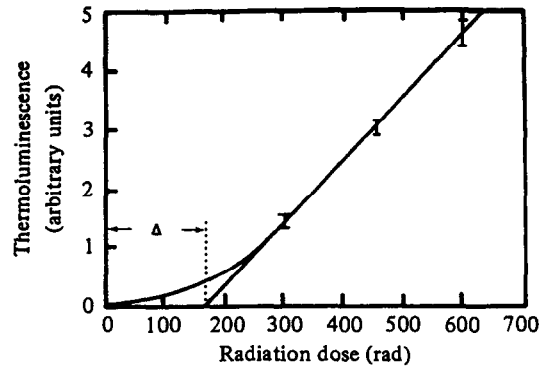


FIG. 4. Supralinearity and superlinearity from pottery quartz (after Aitken (1974)). Here the nonlinear region is observed at low doses (in contrast to Fig. 1). Using the new definition for  $f(D)$  (equation (9)) values of  $f(D)$  may be determined. Thus, both  $f(D)$  and  $g(D)$  are greater than 1 in the low dose region, but  $g(D) = f(D) = 1$  in the high dose region.

estimating the intercept on the dose axis from a backwards extrapolation of the linear dose region (Tite, 1966). Clearly, the field would benefit from a degree of uniformity in the definitions used to describe the nonlinear behavior.

In the present work we would like to discuss the characteristics of the "more than linear" growth described above in its different forms, and to suggest a distinction in the way that these nonlinearities are described which would indicate different features of the nonlinear property. We further propose universal "indices" to mathematically describe all forms of nonlinearity. Specifically, we suggest the use of a "supralinearity index"  $g(D)$  to give an indication of change in the slope of the dose response (in all cases), and a "superlinearity index"  $f(D)$  (previously used by others) to signify the size of the correction term required during extrapolation of the linear dose region (in cases when such a region is present). We expand on the use of these terms in the sections to follow.

## 2. SUPERLINEARITY

As discussed by Chen and Bowman (1978), we suggest that the term "superlinearity" be reserved to refer to the increase in the derivative of the dose dependence function. Let us denote by  $S$  the measured TL signal—i.e. the maximum intensity, or the total area of a peak—and  $D$  the absorbed dose. The derivative of this function at a point  $D$  is  $dS/dD$  (i.e.  $S'(D)$ ), and an increase of the derivative at a certain point is expressed by stating that  $d^2S/dD^2$  (i.e.  $S''(D)$ ) is positive. Thus,  $d^2S/dD^2 > 0$  is defined as representing ranges of superlinearity;  $d^2S/dD^2 < 0$  characterizes ranges of sublinearity; and, of course,  $d^2S/dD^2 = 0$  means a range of linearity. The difficulty with this definition is that it is merely qualitative since it has to do with the sign of the second derivative

only. We would like to propose here a normalized quantity that would not only describe the qualitative feature of being super- or sub-linear, but which would also be a quantitative measure of this property. Moreover, one of the ways to detect superlinearity is by plotting the results on a log-log scale. In this presentation of a function that has the form  $S \propto D^k$ , the slope is found on this scale to be  $k$  and is a direct measure of the degree of superlinearity (or of sublinearity). We think that if one defines a magnitude which evaluates numerically the amount of superlinearity, it would be of great help if this magnitude is indeed  $k$  in the special case in which  $S \propto D^k$ .

We propose here the function:

$$g(D) = \left[ \frac{DS''(D)}{S'(D)} \right] + 1 \quad (1)$$

called the "superlinearity index". As long as we are dealing with a range of an increasing dose dependence, i.e.  $S'(D) > 0$ , it is obvious that  $g(D) > 1$  indicates superlinearity (since  $S''(D) > 0$ ). Furthermore,  $g(D) = 1$  means a range of linearity, and  $g(D) < 1$  signifies sublinearity. It can readily be seen that  $DS''(D)/S'(D)$  is a dimensionless number, which makes  $g(D)$  a dimensionless function, and makes legal the addition of unity. It is quite obvious that taking  $S(D) = \alpha D^k$  results in  $g(D) = k$ , as initially requested. In fact, even for  $S(D) = \alpha D^k + \beta$  we get  $g(D) = k$ .

In order to understand better the significance of this new quantity  $g(D)$ , let us consider some particular cases.

(1) While plotting  $\log S$  vs  $\log D$ , ranges of slopes larger than unity occur (see Halperin and Chen, 1966; Chen *et al.*, 1988) in which the slope varies with the dose in a certain range, say, from 2 to 3. Let us consider the function:

$$S(D) = \alpha D^2 + \beta D^3. \quad (2)$$

One readily gets:

$$g(D) = (2\alpha D + 6\beta D^2)/(2\alpha D + 3\beta D^2) + 1. \quad (3)$$

This yields  $g(D) = 2$  for low doses and  $g(D) = 3$  for high doses, and, of course, a transition from 2 to 3 in between.

(2) Another interesting point relates to the dose ranges in which there is an approach to saturation. One example of such behavior is the function:

$$S = S_0(1 - e^{-\gamma D}), \quad (4)$$

for  $\gamma > 0$ . Using equation (1), one gets:

$$g(D) = 1 - \gamma D \quad (5)$$

which may be approximately unity, positive but less than unity, zero or negative, depending on the dose  $D$ . Of course,  $g(D) < 1$  always indicates sublinearity.

(3) Let us consider the hypothetical case of strong superlinearity, i.e.

$$S(D) = S_0 e^{\gamma D}. \quad (6)$$

This yields:

$$g(D) = \gamma D + 1. \quad (7)$$

The main feature of this is that the superlinearity starts as being  $\sim 1$  at low doses, which represents a nearly linear behavior at low doses, where  $e^{\gamma D} \cong 1 + \gamma D$ . At higher doses, the amount of superlinearity  $g(D)$  grows steadily as more terms of higher order are to be included in the series expansion of  $e^{\gamma D}$ . In other words, the dependence of  $g(D)$  (linearly) on  $D$  demonstrates the very rapid increase of  $S(D) = S_0 e^{\gamma D}$  and its derivatives.

### 3. SUPRALINEARITY

As pointed out in the Introduction, a number of materials have the characteristic property depicted in Fig. 1, namely, an initial linear range which is followed by a nonlinear (i.e. superlinear) region before saturation effects set in. Along with previous authors we describe this property as "supralinearity" and, as shown above in Fig. 2, past authors have presented the amount of supralinearity as response per unit of dose, normalized to a dose in the initial linear range. Some authors (e.g. Horowitz, 1981; Mische and McKeever, 1989) utilized explicitly the dimensionless function  $f(D)$ :

$$f(D) = [S(D)/D]/[S(D_1)/D_1] \quad (8)$$

termed by these authors as either the "supralinearity index", or the "dose response function". Here  $D_1$  is the normalization dose in the linear range and graphical examples of the function  $f(D)$  are shown in Fig. 2. (In Fig. 2,  $f(D)$  is called the "dose response function" but is defined according to equation (8).)

However, we propose here a modified definition of  $f(D)$ , namely:

$$f(D) = \left[ \frac{S(D) - S_0}{D} \right] / \left[ \frac{S(D_1) - S_0}{D_1} \right] \quad (9)$$

where  $S_0$  is the intercept on the TL axis of the extrapolation of the linear region. Note that  $f(D)$  cannot be defined at  $D = 0$ .

For cases such as those shown in Fig. 1, it is clear that  $S_0 = 0$ , and thus the new, more general definition of  $f(D)$  is identical numerically to the older definition of equation (8). However, the advantage of the new definition is that it can be applied to cases such as that illustrated in Fig. 4 in which the superlinear region precedes the linear region. In this case it is impossible to define  $f(D)$  according to equation (8); however, equation (9) may be used by noting that for a dose response of this type we have  $S_0 < 0$ . A negative value for  $S_0$  is valid since it has no physical meaning, but now we see that  $f(D) > 1$  in the low dose region (i.e. that part of the dose dependence in which the TL intensity is greater than that inferred from a backwards extrapolation of the linear region).

With this new definition of  $f(D)$  we see that as long as the function  $S(D)$  is proportional to the dose then

obviously  $f(D) = 1$  and in this case we see that  $g(D) = 1$  and  $f(D) = 1$  mean the same thing. (However, as we will see below, for other cases  $g(D) = 1$  does not necessarily mean  $f(D) = 1$ , and vice-versa.) Values of  $S(D)$  above the extrapolated linear portion (e.g. in either Fig. 1 or Fig. 4) cause  $f(D)$  to be larger than 1. It is obvious that in those dose ranges where the function  $S(D)$  "takes off" above linearity, the dose dependence is both superlinear and supralinear by our definitions. At higher doses, however, when saturation effects start to occur, it may very well happen that the curve is not superlinear any more ( $d^2S/dD^2 \leq 0$ , or  $g(D) \leq 1$ ) but is still above the extrapolated range and therefore  $f(D) > 1$ , i.e. supralinearity occurs. Of course,  $f(D)$  monitors exactly the property it is supposed to monitor, namely the amount of deviation from linearity, that is, the amount to be considered for extrapolation purposes.

In cases (1) and (3) of the previous section,  $f(D)$  cannot be defined since there is no linear region. In case (2),  $f(D) = 1$  at low doses, and becomes less than 1 as saturation is approached.

Figure 5 depicts a hypothetical dose dependence curve consisting of two straight segments intersecting at the point  $D^*$  (79 dose units in the example shown). Obviously, no superlinearity occurs (excluding the point at  $D = D^*$  at which  $S(D)$  is not differentiable). The value of the function in equation (9), however, depends upon the choice of "linear" region. If region I is chosen as the linear part, then  $S_0 = 0$  and  $f(D) = 1$  up to  $D^*$ , and is larger than 1 for  $D > D^*$ . However, if region II is chosen as the linear region, then  $S_0 < 0$  (in this example  $S_0 = -158$  TL units) and thus  $f(D) > 1$  for  $D < D^*$  and  $f(D) = 1$  for  $D \geq D^*$ . In either case, supralinearity is established in a curve consisting of two linear ranges, for each of which  $g(D) = 1$ . How one would calculate  $f(D)$  in a case like this is really a matter of choice, depending upon

whether one wishes to infer TL values at high doses by extrapolation of the low dose region, or vice versa. The behavior of  $f(D)$  in the case illustrated is shown in Fig. 6.

#### 4. FURTHER DISCUSSION

Our purpose in this paper has been to devise methods by which the extent and type of nonlinearity in a TL growth curve can be quantitatively defined and can be applied to all possible cases of non-linear growth. It is not our purpose to discuss the relative merits of the various models forwarded to explain the origin of growth curve nonlinearities—a discussion which is beyond the scope of the present work, and, in any case, has been dealt with in many previous publications (e.g. books by Chen and Kirsch, 1981; McKeever, 1985; Horowitz, 1984a; and articles by Horowitz, 1984b; McKeever, 1990a, b). Instead we wish merely to propose a universal procedure for quantifying the nonlinearity that is observed which, we feel, is more exact than a qualitative—and vague—description of the growth curve as being simply "superlinear" or "supralinear".

There are several situations where one may wish to have a universally agreed-upon method to describe growth curve nonlinearities. For example, in TL dosimetry, defining  $f(D)$  and/or  $g(D)$  allows potential users of TLD materials to assess the degree of difficulty that will be posed by the nonlinear property in the particular application being considered. Thus,  $f(D)$  and/or  $g(D)$ , as defined in this paper, may be quantified and used as a TLD parameter in much the same way that "sensitivity", or "lowest level of detection", are quantified and used now. For example, one might describe  $f(D)$  at the dose  $D_{max}$  at which the maximum value of this function occurs. Thus, by reference to Fig. 2 and to equation (9), one

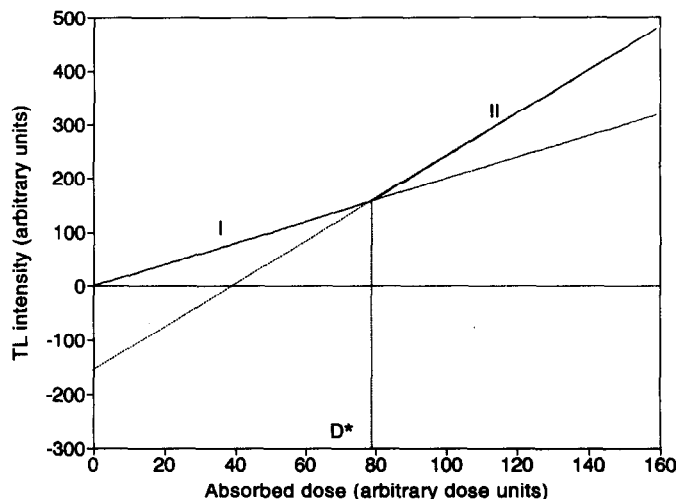


FIG. 5. Hypothetical dose dependence curve consisting of two linear segments (I and II) with different slopes.  $D^*$  (see text) is 79 arbitrary dose units.  $S_0$  for region I is 0, while  $S_0$  for region II is  $-158$ , both in arbitrary TL units.

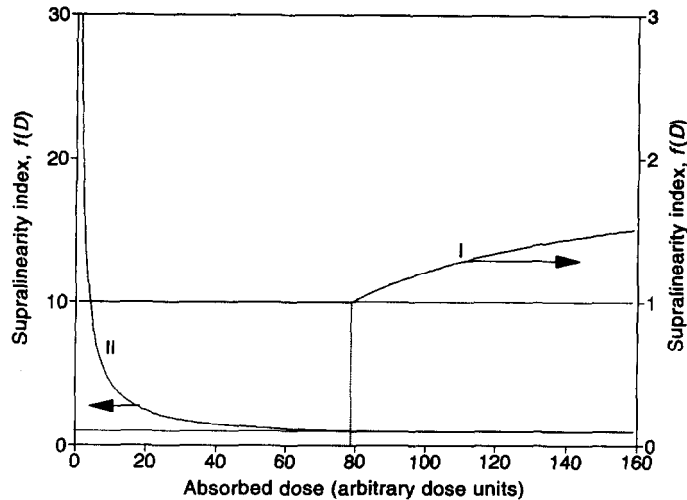


Fig. 6.  $f(D)$  vs  $D$  as determined from the hypothetical case depicted in Fig. 5. Two calculations are shown, depending upon the choice of linear region. If region I from Fig. 5 is chosen as the linear part (i.e.  $S_0 = 0$ ) then curve I (right-hand axis) is calculated from equation (9), using a value of  $D_1$  of 40 arbitrary dose units. If region II from Fig. 5 is chosen, then  $S_0 = -158$  arbitrary TL units and curve II (left-hand axis) is calculated from equation (9), with  $D_1 = 110$  arbitrary dose units.

would define  $f(D_{\max})$  for the 50 ppm sample as “5.6 at  $10^2$  Gy, with  $S_0 = 0$ ”. The complete function  $f(D)$  would give the precise correction needed to calculate the dose, assuming the dosimeter has been normalized in the linear dose region (as is normal practice).

From the figures presented in this paper we see examples in which a growth curve can be supralinear, but not superlinear (for example, Fig. 1 near  $\sim 10^5$  Gy); or superlinear, but not supralinear (for example, Fig. 3 for all doses). For other doses of Fig. 1 we see regions in which the growth curve is both superlinear and supralinear, and regions in which it is neither. The definitions of  $f(D)$  and  $g(D)$  presented herein allow precise quantitative descriptions for all of these regions.

## 5. SUMMARY

In the present paper we have tried to sort and categorize the different aspects of dose dependencies which are “faster than linear”. We propose a somewhat new nomenclature to distinguish between the different features in question, and attach more specific terms to different situations. We have suggested the use of the term “superlinearity” for all cases in which  $d^2S/dD^2 > 0$ . We have defined a new, dimensionless function, called the “superlinearity index”,  $g(D)$ , which makes the property of superlinearity a quantitative measure, and discussed its values in different situations. We have also discussed the properties of the previously defined dimensionless function, the “supralinearity index”,  $f(D)$ , and suggested a modified definition. The new definition is a quantitative measure of the departure from linearity over particular dose ranges.

Examples have been given of dose dependence functions which are superlinear but not supralinear over given ranges, and vice versa. The advantages of the new functions  $g(D)$  and  $f(D)$  is that they allow one to quantify the extent of the nonlinearity, and, at the same time, distinguish between the type of nonlinearity observed. Most experimentally observed nonlinearities can now be quantified using these standard definitions.

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