Thermoluminescence kinetics for multipeak glow curves produced by the release of electrons and holes

Thermoluminescence kinetics for multipeak glow curves produced by the release of electrons and holes

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Abstract. A model is described for the calculation of multipeak glow curves. A set of $(k_m + j_m + 2)$ equations are presented which describe the flow of electrons and holes between k_m different electron trapping levels, j_m different hole trapping levels and the conduction and valence bands. The model can be applied to complex cases where glow curves result from both electron and hole transport. A computer program has been written to solve the system of equations numerically. Calculated glow curves for a number of interesting cases are presented and some other applications of the model are briefly discussed.

1. Introduction

A knowledge of thermoluminescence (TL) kinetics is essential in determining the TL glow curve arising from a given set of defects and trapped charges. A number of simplified approaches to TL kinetic processes have been made (Randall and Wilkins 1945, Garlick and Gibson 1948). These studies focused on cases where only one glow peak is produced. The multipeak case has received little attention until recent work by Levy (1982, 1984a, b). These investigations of the multipeak case were based on an extension of a single-peak formula incorporating various simplifying assumptions as is discussed below. The present work treats the multipeak case starting from the fundamental equations describing charge flows between different defect levels and the conduction and valence bands.

Calculations of the kinetics of the thermoluminescence process usually start from a consideration of a simplified energy level scheme consisting of one type of electron trapping level and one type of hole-trapping level. During heating to obtain a thermo-luminescence glow curve electrons are released thermally from traps into the conduction band, from whence they may be retrapped or may recombine with trapped holes with the emission of light. The retrapping and recombination processes are characterised by coefficients A_t and B_r respectively which are equal to the products of the appropriate electron velocity and microscopic cross sections. The 'traffic' of electrons between traps, conduction band and hole centres may be described by three equations given by Halperin

and Braner (1960)

$$I(t) = -dm/dt = B_r m n_c \tag{1}$$

$$-dn/dt = sn \exp(-E/kT) - A_t n_c (N - n) \tag{2}$$

$$dn_c/dt = dm/dt - dn/dt \tag{3}$$

where n_c, n and m are the instantaneous values of the densities of conduction band electrons, trapped electrons and trapped holes respectively. E and s are the activation energy and the frequency factor for the electron trap, N is the total density of these traps and k is Boltzmann's constant. The simplifying assumption is usually made that electrons do not accumulate in the conduction band, i.e.

$$\left| \frac{dn_c}{dt} \right| \ll \left| \frac{dn}{dt} \right| \text{ and } n_c \ll n. \tag{4}$$

From these assumptions we have, using equation (3)

$$\frac{dm}{dt} = \frac{dn}{dt}$$

and so the right-hand sides of equations (1) and (2) are equal, yielding

$$n_c = \frac{sn \exp(-E/kT)}{B_r m + A_t(N - n)}$$

and upon substituting this expression into equation (1) we obtain a single equation for the TL glow intensity

$$I(t) = -\frac{dm}{dt} = sn \exp\left(-\frac{E}{kT}\right) \frac{B_r m}{B_r m + A_t(N - n)} \tag{5}$$

from which the familiar first and second-order glow curves may be obtained by making assumptions about the relative strengths of the retrapping and recombination transitions. Clearly equation (5) is capable of generating glow curves which correspond to neither the first nor the second-order cases. All of the equations given so far would be identical in form if the electrons were taken to be fixed and the holes to be thermally released to recombine with them. However, so far we have not considered the case where both electrons and holes are thermally released in the same temperature range. That this could occur was first suggested by Schön (1956, 1958). A set of equations describing such a situation was first given by Bräunlich and Scharmann (1966). These equations (in our notation) are as follows

$$\frac{dn}{dt} = -ns_e \exp\left(-\frac{E}{kT}\right) + A_t n_c (N - n) - A_r n m_v \tag{6}$$

$$\frac{dn_c}{dt} = ns_e \exp\left(-\frac{E}{kT}\right) - A_t n_c (N - n) - B_r n_c m \tag{7}$$

$$\frac{dm_v}{dt} = ms_h \exp\left(-\frac{H}{kT}\right) - B_t m_v (M - m) - A_r m_v n \tag{8}$$

$$\frac{dm}{dt} = -ms_h \exp\left(-\frac{H}{kT}\right) + B_t m_v (M - m) - B_r m n_c \tag{9}$$

where the densities of holes in the valence band and in the traps are m_v and m respectively. The density, activation energy, frequency factor, retrapping and recombination coefficients for the hole traps are denoted M, H, s_h, B_t, B_r respectively. The notations for the electron trap frequency factor and recombination coefficient are now s_e and A_r, otherwise the notation is as before. Numerical solutions to these equations have been presented by Chen *et al* (1984) and by McKeever *et al* (1985).

All of the foregoing equations refer to cases leading to glow curves having only one peak. Multipeak glow curves can, of course, be generated in a simple-minded way by just using the first- or second-order equations k times for k sets of E, s, N_0, N values thus obtaining k glow peaks. Inherent in this approach is the assumption that the electron transport leading to each peak is non-interactive, i.e. electrons released from trapping level 1 cannot be retrapped by one of the other trapping levels.

A calculation of multipeak glow curves using interactive kinetics has been carried out by Levy (1982, 1984a, b) using equations which are essentially, generalisations of equation (5). This approach has yielded a number of interesting results; nevertheless it necessarily incorporates simplifying assumptions similar to those given in equation (4). In addition, this method does not deal with the simultaneous release of electrons and holes and so cannot be used to examine the case where electron and hole peaks are present together in the same glow curve. Such glow curves seem to occur for some phosphors (see, for example, Summers (1984) for a discussion of the case of α-Al_2O_3). (Kemmey *et al* (1967) have constructed a general model for TL kinetics but do not seem to have applied it to the multipeak case or to the case of mixed electron and hole transport).

In order to overcome these limitations a model describing multipeak, interactive kinetics for the case of simultaneous electron and hole release has been constructed by generalising equations (6–9). This model is described in the following section.

2. The model and numerical analysis

A schematic energy level diagram for an insulator having several electron and hole trapping levels is shown in figure 1. Electrons released from any trap may (depending on the values assigned to the different retrapping coefficients) be retrapped at any of the electron trapping levels or may (depending on the values assigned to the different
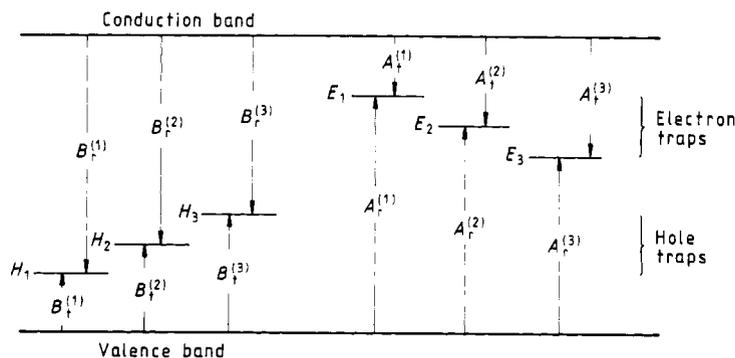

Figure 1. Schematic diagram of the energy levels associated with the transitions considered in the model.

recombination coefficients) recombine at any of the hole trapping levels. An exactly analogous situation exists for thermally released holes†.

Associated with the kth electron level we have electron densities, trap densities, activation energies, frequency factors, retrapping coefficients and recombination coefficients $n_k, N_k, E_k, s_e^{(k)}, A_t^{(k)}, A_r^{(k)}$ and similarly for the jth hole level we have hole densities etc denoted by $m_j, M_j, H_j, s_h^{(j)}, B_t^{(j)}, B_r^{(j)}$.

With this notation equations for the flow of charge carriers between the different bands and localised levels can be written. For k_m electron levels and j_m hole levels we have k_m equations

$$\frac{\mathrm{d}n_k}{\mathrm{d}t} = -n_k s_e^{(k)} \exp\left(-\frac{E_k}{kT}\right) + n_c A_t^{(k)}(N_k - n_k) - m_r A_r^{(k)} n_k \tag{10}$$

for $k = 1, k_m$ and j_m equations

$$\frac{\mathrm{d}m_j}{\mathrm{d}t} = -m_j s_h^{(j)} \exp\left(-\frac{H_j}{kT}\right) + m_v B_t^{(j)}(M_j - m_j) - n_c B_r^{(j)} m_j \tag{11}$$

for $j = 1, j_m$.

There are also two equations for the densities of conduction band electrons and valence band holes i.e.

$$\frac{\mathrm{d}n_c}{\mathrm{d}t} = \sum_{k=1}^{k_m} n_k s_e^{(k)} \exp\left(-\frac{E_k}{kT}\right) - \sum_{k=1}^{k_m} n_c A_t^{(k)}(N_k - n_k) - \sum_{j=1}^{j_m} n_c B_r^{(j)} m_j \tag{12}$$

and

$$\frac{\mathrm{d}m_v}{\mathrm{d}t} = \sum_{j=1}^{j_m} m_j s_h^{(j)} \exp\left(-\frac{H_j}{kT}\right) - \sum_{j=1}^{j_m} m_v B_t^{(j)}(M_j - m_j) - \sum_{k=1}^{k_m} m_v A_r^{(k)} n_k. \tag{13}$$

Thus we have to solve a system of $(k_m + j_m + 2)$ simultaneous, first order differential equations. These equations incorporate the charge balance condition

$$\frac{\mathrm{d}n_c}{\mathrm{d}t} + \sum_{k=1}^{k_m} \frac{\mathrm{d}n_k}{\mathrm{d}t} = \frac{\mathrm{d}m_v}{\mathrm{d}t} + \sum_{j=1}^{j_m} \frac{\mathrm{d}m_j}{\mathrm{d}t}. \tag{14}$$

There are k_m components to the TL glow curve of the form

$$I_k(t) = \alpha_k m_v A_r^{(k)} n_k \tag{15}$$

and j_m components of the form

$$I_j(t) = \beta_j n_c B_r^{(j)} m_j \tag{16}$$

where the α_k, β_j represent the luminescent efficiencies of the transitions and may take values between 0 and 1. The total TL output is thus

$$I_{\text{TOT}}(t) = \sum_{k=1}^{k_m} I_k(t) + \sum_{j=1}^{j_m} I_j(t). \tag{17}$$

It is not possible to solve equations (10–13) analytically unless a number of restrictive assumptions are made. However in this paper the equations are solved numerically

† Note that throughout this discussion we refer to electron traps and hole traps rather than traps and centres since the model is symmetric with respect to electrons and holes and indeed a given level may take on the role of both trap and centre.

using a fourth-order Runge–Kutta method. The temperature step width used for the calculation was usually 0.002 K. For a three peak glow curve covering a temperature range from 293 to 700 K a CPU time of about 100 s is required using the IBM 3081 K Harwell Central Computer. At large carrier densities (>10^{17} m^{-3}) or for extremely high retrapping or recombination numerical instabilities occur. These could be overcome by reducing the step width (the accuracy of a fourth-order method improves as the fifth power of the inverse of the step width) or by using a higher order of numerical solution. Both alternatives lead to increased computing times. Nevertheless, the present program is capable of obtaining solutions for a wide range of interesting cases.

The program calculates TL glow curves and also the total integrated TL output. It can also provide the following information.

(i) The electron and hole densities are calculated at each temperature throughout the glow curve. The densities of electrons and holes in conduction and valence bands respectively are of particular interest since they allow the calculation of the thermally stimulated conductivity $\sigma(T)$ from the equation

$$\sigma(T) = n_c(T)e\mu_e + m_v(T)e\mu_h \tag{18}$$

where e is the electronic charge and μ_e and μ_h are the electron and hole mobilities respectively.

(ii) In addition to the total TL output, the emission produced by each individual transition is recorded as a function of temperature. For models involving three electron levels and three hole levels a total of six different transitions are possible, although usually only a few of these contribute significantly to the total TL output. If emission bands could be assigned to these different transitions it would be possible to model changes in TL spectra with glow curve temperature.

(iii) The program allows the heating rate to be set to zero so that the effects of heating at a constant temperature can be studied. The trapped charge densities at the end of this period can then be used as input for a second calculation (using a finite heating rate) in order to obtain the glow curve resulting from a period of thermal fading.

These latter features of the program will not be explored in this paper in which attention is focused on the calculation of TL glow curves and integrated TL output.

3. Results

3.1. TL glow curves

Case 1: First-order type
Figure 2 shows a computed glow curve for the case of three electron levels and three hole levels and a set of input parameters (see table 1) intended to approximate first-order conditions i.e. when recombination greatly exceeds retrapping. The hole traps are all thermally disconnected, i.e. $H_j \gg kT$ throughout the glow curve. The resulting glow curve has three peaks each having the usually asymmetric 'first-order' shape. For comparison, glow curves obtained with the same E and s values using the simple first-order kinetics equation (Randall and Wilkins 1945) are shown. The agreement is good, as it should be in this case, giving some confidence in the model and in the calculations. Note that both this model and the simple first-order case yield lower, broader peaks at higher temperatures although the initial electron density in each of the three trapping levels was the same.
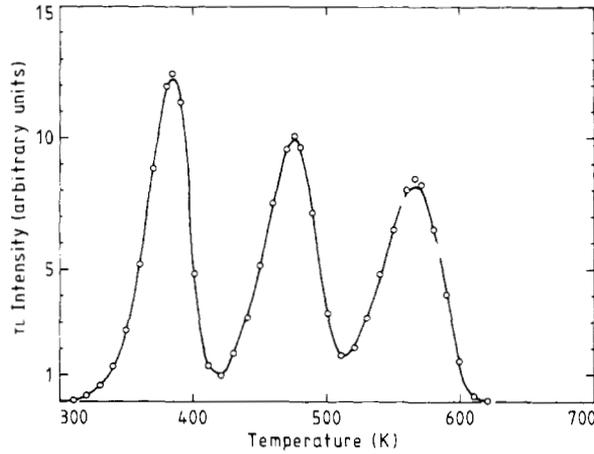
Figure 2. Calculated glow curve showing three electron peaks of first-order character. Also shown is the ($\bigcirc$) curve calculated using the simple first-order equations for three trapping levels with the same E and s values as the electron traps considered in the interactive kinetics calculations.

Case 2: Electron and hole peaks

Figure 3 shows a glow curve for a set of parameters almost identical to those used for figure 2 except that the second shallowest level is now a hole trap (see table 1) so that the final glow curve consists of a hole peak between electron peaks. Again the simple first-order curves are plotted for comparison. The first peak is unchanged but peaks 2

Table 1. Trap parameters used in the calculations.

		n_0 and m_0 (m^{-3})	N and M (m^{-3})	s_e and s_h (s^{-1})	E and H (eV)	A_t and B_t (m^3 s^{-1})	A_r and B_r (m^3 s^{-1})
Case 1							
Electron Trap							
Parameters	E_1	1×10^{15}	1×10^{15}	1×10^{10}	0.8	1×10^{-21}	1×10^{-13}
	E_2	1×10^{15}	1×10^{15}	1×10^{10}	1.0	1×10^{-21}	1×10^{-13}
	E_3	1×10^{15}	1×10^{15}	1×10^{10}	1.2	1×10^{-21}	1×10^{-13}
Hole Trap							
Parameters	H_1	1×10^{15}	1×10^{15}	1×10^{10}	2.0	1×10^{-21}	1×10^{-13}
	H_2	1×10^{15}	1×10^{15}	1×10^{10}	2.5	1×10^{-21}	1×10^{-13}
	H_3	1×10^{15}	1×10^{15}	1×10^{10}	3.0	1×10^{-21}	1×10^{-13}
Case 2 As 1 except							
	E_2	1×10^{15}	1×10^{15}	1×10^{10}	1.2	1×10^{-21}	1×10^{-13}
	E_3	1×10^{15}	1×10^{15}	1×10^{10}	2.0	1×10^{-21}	1×10^{-13}
	H_1	1×10^{15}	1×10^{15}	1×10^{10}	1.0	1×10^{-21}	1×10^{-13}
Case 3 Curve A							
	E_1	1×10^{15}	1×10^{16}	1×10^{10}	0.8	1×10^{-15}	1×10^{-13}
	E_2	1×10^{15}	1×10^{16}	1×10^{10}	1.0	1×10^{-15}	1×10^{-13}
	E_3	1×10^{15}	1×10^{16}	1×10^{10}	1.2	1×10^{-15}	1×10^{-13}
	H_1	1×10^{15}	1×10^{16}	1×10^{10}	2.0	1×10^{-15}	1×10^{-13}
	H_2	1×10^{15}	1×10^{16}	1×10^{10}	2.5	1×10^{-15}	1×10^{-13}
	H_3	1×10^{15}	1×10^{16}	1×10^{10}	3.0	1×10^{-15}	1×10^{-13}

Table 1. cont.
Curve B. As for Curve A except all A_t, $B_t = 1 \times 10^{-17}$ and all A_r, $B_r = 1 \times 10^{-15}$
Curve C. As for Curve A except all A_t, $B_t = 1 \times 10^{-18}$ and all A_r, $B_r = 1 \times 10^{-16}$
Curve D. As for Curve A except all A_t, $B_t = 1 \times 10^{-19}$ and all A_r, $B_r = 1 \times 10^{-17}$

Case 4

E_1	1×10^{16}	1×10^{16}	1×10^{10}	0.8	1×10^{-14}	1×10^{-14}
E_2	1×10^{16}	1×10^{16}	1×10^{10}	1.0	1×10^{-14}	1×10^{-14}
E_3	1×10^{16}	1×10^{16}	1×10^{10}	1·2	1×10^{-14}	1×10^{-14}
H_1	1×10^{16}	1×10^{16}	1×10^{10}	2.0	1×10^{-14}	1×10^{-14}
H_2	1×10^{16}	1×10^{16}	1×10^{10}	2.5	1×10^{-14}	1×10^{-14}
H_3	1×10^{16}	1×10^{16}	1×10^{10}	3.0	1×10^{-14}	1×10^{-14}

Case 4a as 4 except all n_0, N, m_0, $M = 1 \times 10^{17}$

Case 4b as 4a except all n_0, $m_0 = 3 \times 10^{16}$

Case 4c as 4a except all n_0, $m_0 = 1 \times 10^{16}$

Case 5

E_1	1×10^{16}	1×10^{16}	1×10^{10}	0.8	1×10^{-16}	1×10^{-13}
E_2	1×10^{16}	1×10^{17}	1×10^{10}	1.0	1×10^{-14}	1×10^{-13}
E_3	1×10^{16}	1×10^{16}	1×10^{10}	1.2	1×10^{-16}	1×10^{-13}
H_1	1×10^{16}	1×10^{16}	1×10^{10}	2.0	1×10^{-15}	1×10^{-14}
H_2	1×10^{16}	1×10^{16}	1×10^{10}	2.5	1×10^{-15}	1×10^{-14}
H_3	1×10^{16}	1×10^{16}	1×10^{10}	3.0	1×10^{-15}	1×10^{-14}

Case 6a

E_1	1×10^{16}	1×10^{16}	1×10^{10}	1.0	1×10^{-18}	1×10^{-13}
E_2	1×10^{16}	1×10^{16}	1×10^{10}	1.2	1×10^{-18}	1×10^{-13}
E_3	1×10^{16}	1×10^{16}	1×10^{10}	2.0	1×10^{-18}	1×10^{-13}
H_1	3×10^{16}	1×10^{17}	1×10^{10}	0.8	1×10^{-18}	1×10^{-13}

Case 6b As 6a except

E_1	1×10^{16}	1×10^{16}	1×10^{10}	0.8	1×10^{-18}	1×10^{-13}
E_2	1×10^{16}	1×10^{16}	1×10^{10}	1.2	1×10^{-18}	1×10^{-13}
H_1	3×10^{16}	1×10^{17}	1×10^{10}	1.0	1×10^{-18}	1×10^{-13}

Case 6c As 6a except

E_2	1×10^{16}	1×10^{16}	1×10^{10}	1.0	1×10^{-18}	1×10^{-13}
H_1	3×10^{16}	1×10^{17}	1×10^{10}	1.2	1×10^{-18}	1×10^{-13}

Case 6d As 6c except

E_3	1×10^{16}	1×10^{16}	1×10^{10}	1.2	1×10^{-18}	1×10^{-13}
H_1	3×10^{16}	1×10^{17}	1×10^{10}	2.0	1×10^{-18}	1×10^{-13}

Case 6e

E_1	1×10^{16}	1×10^{16}	1×10^{10}	1.0	1×10^{-18}	1×10^{-13}
E_2	1×10^{16}	1×10^{16}	1×10^{10}	1.2	1×10^{-18}	1×10^{-14}
E_3	1×10^{16}	1×10^{16}	1×10^{10}	2.0	1×10^{-18}	1×10^{-15}
H_1	3×10^{16}	1×10^{17}	1×10^{10}	0.8	1×10^{-18}	1×10^{-13}

Case 7

E_1	1×10^{16}	1×10^{16}	1×10^{10}	0.8	1×10^{-20}	1×10^{-13}
E_2	1×10^{16}	1×10^{16}	1×10^{10}	1.2	1×10^{-20}	1×10^{-13}
E_3	1×10^{16}	1×10^{16}	1×10^{10}	2.0	1×10^{-20}	1×10^{-13}
H_1	1×10^{16}	1×10^{16}	1×10^{10}	1.0	1×10^{-20}	1×10^{-13}
H_2	1×10^{16}	1×10^{16}	1×10^{10}	2.5	1×10^{-20}	1×10^{-13}
H_3	1×10^{16}	1×10^{16}	1×10^{10}	3.0	1×10^{-20}	1×10^{-13}

Notes: Heating rate 5 K s^{-1} in all cases.
  All transitions taken to be radiative.
  E_1, E_2, etc refer to electron trapping levels.
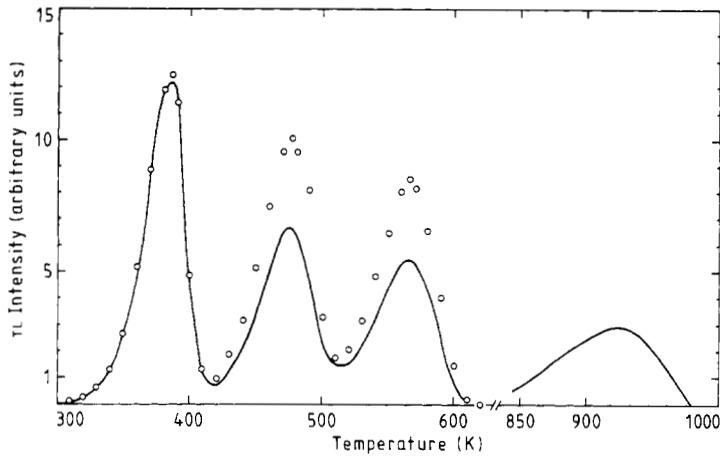 H_1, H_2, etc refer to hole trapping levels.

Figure 3. Calculated glow curve for the case where the second peak is a hole peak. Note the break in the temperature scale and the presence of a high temperature peak. The simple first-order curve is shown for comparison (○).

and 3 are now reduced in height. The reason for this is as follows. As electron level 1 empties to give peak 1, one-third of the electrons recombine with holes in the thermally connected hole trap (since recombination coefficients for each of the hole levels are equal). When this hole trap empties to give peak 2 the peak is reduced in area by one-third. Half of the released holes recombine with electrons situated in the next electron level so that when it empties to give peak 3 this peak is also reduced in area. The ratios of peak areas to the corresponding peaks in case 1 are $1, 2/3$ and $2/3$. Another interesting feature is that, at the end of peak 3, trapped charge remains in the deepest electron trap and the two deepest hole traps. The TL deficit from peaks 2 and 3 ultimately appears in a fourth peak at a temperature of $\simeq 920$ K (although such a high temperature peak would not be observed in practice). That the fourth peak occurs at such a high temperature is simply a result of the depth (2.0 eV) used for the third electron trap.

Case 3: Effects of changes in magnitudes of retrapping and recombination coefficients
Figure 4 shows a set of glow curves for the case where the ratio of retrapping to recombination coefficients is 10^{-2}. Also only one-tenth of each of the trapping levels is filled initially. Retrapping though not dominant, is no longer negligible. When the retrapping and recombination coefficients are large (curve A) the agreement with the simple first-order curve is still reasonable although a high temperature tail is evident on the third peak. However, the glow curve shape alters markedly (curves B–D) when the retrapping and recombination coefficients are decreased (but are still in the ratio 10^{-2}). The peaks become lower and broader and are shifted to higher temperature. Also the high temperature tail is enhanced. The reason for this behaviour is that, as the various retrapping and recombination coefficients decrease, the lifetimes for charge carriers in the conduction and valence bands increase and 'pile-up' of carriers in these bands can occur. For example, at the third TL peak the electron density in the conduction band is 1.9×10^{12} m^{-3} for curve A and 1.7×10^{15} m^{-3} for curve D. The slow relaxation of this electron population (mainly via recombination with trapped holes) leads to the long high temperature tail in the glow curve. These results emphasise one of the differences between the model described here and that due to Levy (1982, 1984a, b). As pointed
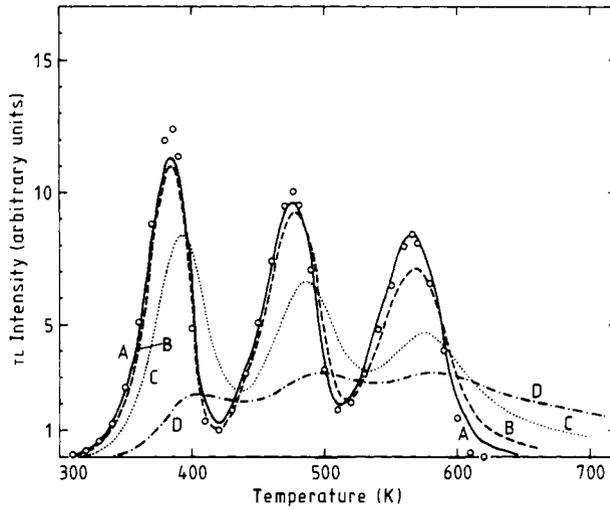
Figure 4. Calculated glow curves for cases where the ratio of retrapping to recombination coefficients is 10^{-2} but the magnitudes of the coefficients decrease from curve A to curve D. (○) The simple first-order curve is shown for comparison.

out above, the latter model incorporates the assumption that free charge accumulation is negligible and results in glow curves with shapes determined only by the ratio of retrapping to recombination coefficients (if all other parameters are unchanged).

Case 4: Second-order types
Second-order glow curves can be obtained from equation (5) either by setting $A_t(N - m) \gg B_r m$ and $n \ll N$ or by setting $B_r = A_t$ and $m = n$ throughout the glow curve. Figure 5 shows a glow curve computed for the case where retrapping and recombination
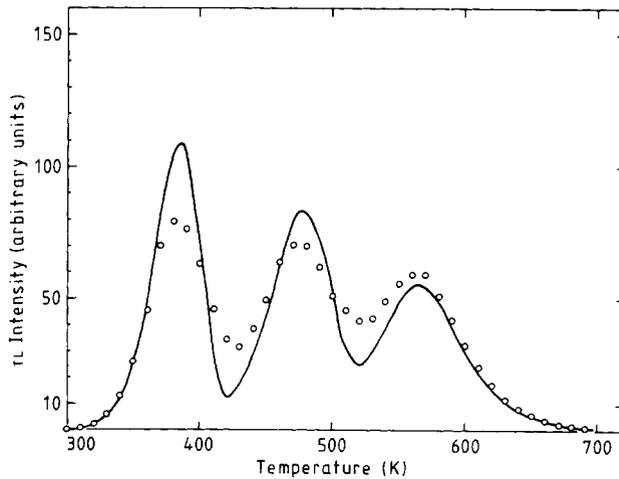
Figure 5. Calculated glow curve for a set of levels with equal retrapping and recombination coefficients. (○) The curve obtained using the simple second-order equations is shown for comparison.

coefficients are equal (see table 1). This condition is insufficient to ensure a second-order glow curve as the additional condition $m = n$ is not met for the first two peaks. As the first electron level begins to empty the n_0 electrons which are thermally connected 'see' $3\,m_0$ ($=3\,n_0$) available trapped holes and recombination is enhanced. By the time the last electron level empties, however, the n_0 released electrons have only m_0 holes available for recombination and second-order conditions are now operative. A simple second-order glow curve is shown in figure 5 for comparison. As might be expected from the foregoing discussion, this curve does not provide a perfect fit to the computed glow curve. The enhanced recombination results in the first two peaks having the more steeply falling characteristics of first-order peaks, leading to a poor correspondence with the second-order curve; however, the high temperature side of peak 3 gives a good fit to the second-order curve in agreement with the discussion above.

The alternative route to second-order kinetics is to have retrapping dominant over recombination. Figures 6(a–c) show a set of computed curves for which retrapping coefficients are still equal to the recombination coefficients but the fraction of each level initially occupied by trapped charge varies from 1.0 to 0.1 (see cases 4a, b, c, table 1). In this case no close correspondence exists with the simple second-order curves. As the fraction of occupied traps decreases and retrapping becomes more important, the peaks shift to higher temperature and the third glow peak begins to grow at the expense of the lower temperature peaks. Electrons released during the early part of the glow curve are often retrapped. Those retrapped at shallow electron traps are readily released again but those retrapped into the deepest levels will remain there. When the deepest level
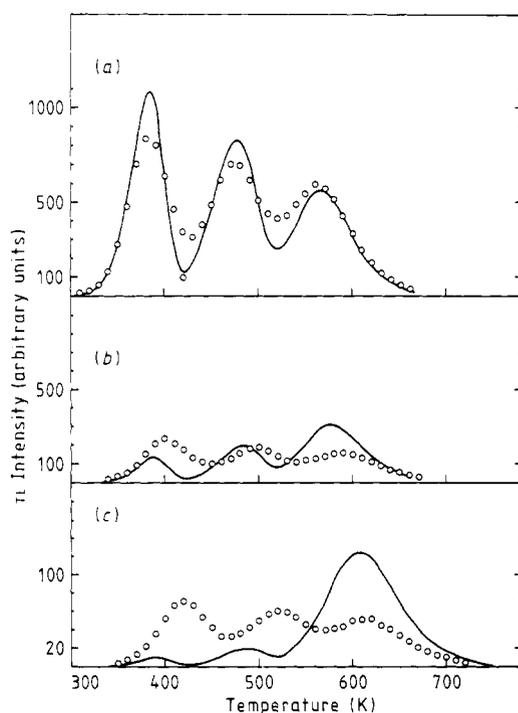

Figure 6. Calculated glow curves for cases where the retrapping and recombination coefficients are equal but the fraction of initially filled traps decreases from 1.0 (a), 0.3 (b) to 0.1 (c). Second-order curves are shown for comparison ($\bigcirc$).

empties to yield peak 3 it has acquired many electrons released from earlier peaks. This effect was also obtained by Levy (1982, 1984a) using his simpler interactive model.

Figures 6(*a–c*) clearly show that peak height is not proportional to initial trapped charge density in these cases.

Case 5: Mixed 'first' and 'second-order' peaks

Figure 7 shows a glow curve consisting of three electron peaks. The recombination
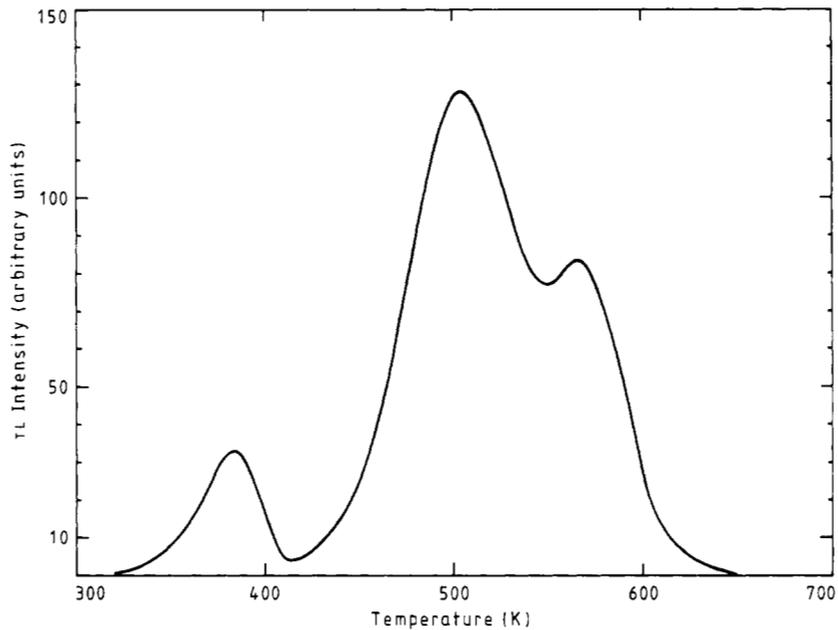

Figure 7. Calculated glow curve for a level with high retrapping characteristics situated between two first-order type peaks.

coefficients for all three hole levels are quite high (see table 1). The retrapping to the first and third electron levels is weak whereas that to the middle electron level is strong (note that equation (5) indicates that, for electron peaks, it is the retrapping to the electron levels and the recombination to the hole levels which determines peak shape). Thus a peak which, though not strictly second-order, has high retrapping lies between two peaks of first-order type. This middle trapping level tends to capture electrons released during the first peak and therefore grows at the expense of the first peak.

Case 6: Three electron traps—one hole trap

In the series of cases considered here all of the holes are contained in one trapping level and there are three electron trapping levels. Recombination dominates in all cases. Changes in glow curve structure are examined for cases where the hole trap is the first, second, third or fourth most unstable level (see table 1). The resulting glow curves are shown in figure 8.

When the hole trapping level is the least stable (curve A) there is just one low temperature peak produced when the hole traps empty, leading to recombinations with all trapped electrons.
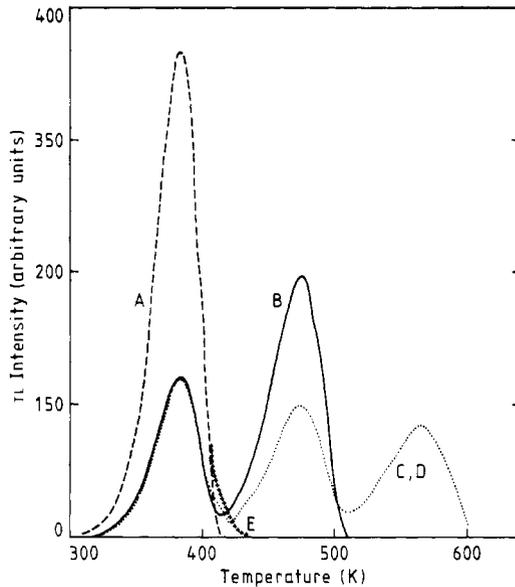
Figure 8. Calculated glow curves for three electron levels and one hole level. Curve A case 6a; Curve B case 6b; Curve C case 6c; Curve D case 6d; show the changes in glow curve structure as the hole trap is changed from the least stable to the most stable trap. Curve E is the same as Curve A except that the three electron levels now have different recombination coefficients.

Curve B shows the case where an electron level is least stable. This releases its charge to give a low temperature peak. The remaining holes are then thermally released to recombine with all remaining electrons yielding a second, more prominent peak.

In the case of curves C and D the hole trap is the third and fourth least stable trapping level respectively. Both of these cases lead to identical three-peak glow curves (see figure 8). In the case of curve C the two shallowest electron traps empty sequentially leading to recombinations with trapped holes and then the remaining holes are released to recombine at the deepest electron traps. In the case of curve D all three electron traps empty sequentially and the resulting electrons recombine with trapped holes.

Finally curve E shows an identical case to curve A except that the recombination coefficients of the three electron traps are not equal, but decrease with increasing trap depth. A single peak glow curve results as with curve A: however, a high temperature tail is produced because the reduced recombination coefficients increase the hole lifetime in the valence band.

This set of results shows how quite different glow curves can result from a broadly similar array of trapping levels depending on the order of trap stabilities.

3.2. Integrated TL

Table 2 shows the total TL emission integrated over the whole glow curve for the above mentioned cases. The integrated TL is proportional to the initial trapped charge density. Chen and Kirsh (1981) have shown that the total TL glow is proportional to the lowest of the initial charge densities n_0 and m_0. Since, for all cases examined here, $\Sigma_k n_{0k} = \Sigma_j m_{0j}$

($\Sigma_k n_{0k}$ denotes the sum over the initial trapped electron densities and $\Sigma_j m_{0j}$ the sum over the initial trapped hole densities) this statement is equivalent to the observed relationship that integrated TL is proportional to the initial trapped charge density.

Table 2. Integrated TL emission.

Case	Initial trapped electron density[†] (m^{-3})	Total TL (arb. units)
1	3×10^{15}	1.46
2	3×10^{15}	1.46
3	3×10^{15}	1.44
4a	3×10^{17}	145.0
4b	9×10^{16}	44.2
4c	3×10^{16}	14.5
5	3×10^{16}	14.5
6a	3×10^{16}	14.5

[†] Equal to the initial number of trapped holes.

4. Conclusions

The model provides a description of the kinetic processes occurring in complex glow curves, including cases where electron and hole peaks exist together.

The results described here show that for high ratios of recombination to retrapping coefficients glow curves are obtained which are closely fitted by simple first-order curves. As retrapping becomes more important, more complex behaviour is observed with the highest temperature peak growing at the expense of lower temperature ones. Multipeak glow curves are not, in general, well fitted by second-order curves except over limited regions. The presence of hole peaks amongst electron peaks can alter the relative peak heights and even introduce new peaks, even though the total number of trapping levels remains the same. Levels for which retrapping is large tend to capture charge at the expense of shallower levels for which recombination dominates.

The program described here can also be used to study TL fading and phosphorescence by setting the heating rate to zero. The various electron densities are also generated as a function of temperature and these can be used to investigate phenomena such as thermally stimulated conductivity.

A computer model describing the creation and filling of traps during irradiation is now in the process of completion and will be described in a subsequent publication. It is intended that this model be used in conjunction with the TL readout program described here in order to carry out a more complete investigation of TL phenomena.

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