

## Determination of the activation energy of a thermoluminescence peak obeying mixed order kinetics

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**Abstract** : A method for the determination of the activation energy  $E$  of a thermoluminescence (TL) peak obeying mixed order kinetics by using a set of expressions, is presented. The method has been applied to the numerically generated mixed order and general order TL peaks and experimental TL peak of BeO.

**Keywords** : Thermoluminescence, mixed order kinetics, activation energy

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### 1. Introduction

Thermoluminescence (TL) is often used for the spectroscopic studies of trap levels, in particular, for the evaluation of the trap depth (or activation energy) [1]. The study remains active because of its application in dating and dosimetry [2,3]. A large number of TL peaks can be explained in terms of the three parameters (activation energy  $E$ , frequency factor  $s$  and the order of kinetics  $b$ ) formalism. In order to study TL peaks obeying general order (GO) kinetics whose shape factor  $\mu_g$  lies between 0.42 and 0.52, Chen [4] used the empirical relation given by May and Partridge [5]

$$I(t) = -dn/dt = s'n^b \exp(-E/(kT)), \quad (1)$$

where  $I(t)$  is the intensity of emission at time  $t$ ,  $E$  (eV) is the activation energy,  $s'$  the pre-exponential factor having a dimension of  $\text{cm}^{3(b-1)} \text{sec}^{-1}$ ,  $k$  (eV  $k^{-1}$ ) the Boltzmann constant,  $n$  ( $\text{cm}^{-3}$ ) the concentration of trapped electrons at time  $t$  and  $T$  the absolute temperature.

In spite of the extensive use and application of GO kinetics by a number of research workers it, however, lacks the physical basis and a more physical mixed order (MO) kinetics has been developed by Chen *et al* [6] from the set of three differential equations [7]. The first order equation [8] and the second order equation [9] can be derived from these differential equations. Yossian and Horowitz [10] have successfully applied MO kinetics both to the synthetic TL glow peaks and to isolated peak 5 in LiF : Mg, Ti (TLD-100) following post irradiation annealing at 165°C and commented that MO kinetics is a viable alternative to GO kinetics in the intermediate range ( $1 < b < 2$ ). Chen *et al* [6] have also presented a graphical picture of variation of  $\mu_g$  with  $\alpha$  ( $\alpha = n_0/(n_0 + c)$ ), where  $n_0$  is the initial concentration of trapped electrons and  $c$  is the concentration of trapped electrons or holes not taking part in TL process in the temperature range being considered) by choosing a certain value of  $u_m$  ( $u_m = E/(kT_m)$ , where  $T_m$  is the temperature at peak intensity  $I_m$ ) and reported that  $\mu_g$  is relatively a strong function of  $\alpha$  ( $0 \leq \alpha \leq 1$ ) and a very weak function of  $u_m$ . They have also applied the half intensity peak shape formula [4] for determination of activation energy  $E$  to synthetic glow peaks generated using MO kinetics equation and shown that the values of  $E$  are within 3% of the given values.

In this paper, the dependence of  $\mu_g$  on  $u_m$  is taken into account in obtaining the value of  $\alpha$  from  $\mu_g$  vs  $\alpha$  curve by taking average of  $\mu_g$  for different values of  $u_m$  ( $20 \leq u_m \leq 40$ ). We also present a set of peak shape formulae involving the parameter  $\alpha$  for the determination of activation energy  $E$  for TL peaks obeying MO kinetics unlike those of Chen [4] used by [6] which do not require the information of  $\alpha$ . The set of formulae has been applied to numerically generated MO TL peaks to obtain  $E$  and found to be in good agreement with the input energy. Recently, Sakurai and Gartia [11] used the exact solutions of the basic differential equations involving thermally disconnected traps to fit the experimental peak of BeO to determine the five important intrinsic trapping parameters, namely, activation energy, frequency factor, retrapping probability, recombination probability and concentration of the disconnected traps. In the present paper the applicability of MO kinetics is discussed as an alternative model to analyse the experimental TL peak of BeO irradiated with X-ray (5 minutes).

## 2. Theory

Following Chen *et al* [6] the intensity of a TL peak obeying MO kinetics can be written as

$$I(t) = -dn/dt = s''n(n+c)\exp(-E/(kT)) \quad (2)$$

with  $s'' = sA_m/NA_n$ , (3)

where  $s$  is the frequency factor ( $s^{-1}$ ),  $N$  the total concentration of traps,  $A_m$  and  $A_n$  are respectively the probabilities ( $s^{-1}$ ) of recombination and retrapping. The solution of eq. (2) for a linear heating rate  $\beta$  is given by

$$I(T) = \frac{s''c^2 \alpha \exp\left[(cs''/\beta) \int_{T_0}^T \exp(-E/(kT'))dT'\right] \exp(-E/(kT))}{\left\{ \exp\left[(cs''/\beta) \int_{T_0}^T \exp(-E/(kT'))dT'\right] - \alpha \right\}^2}, \quad (4)$$

where  $T_0$  is the initial temperature when  $n = n_0$ .

The condition for maximum intensity is given by

$$\left[ \frac{E}{kT_m^2} + (cs''/\beta) \exp(-E/(kT_m)) \right] \left[ \exp\left\{ (cs''/\beta) \int_{T_0}^{T_m} \exp(-E/(kT'))dT' \right\} - \alpha \right] = 2(cs''/\beta) \exp(-E/(kT_m)) \exp\left\{ (cs''/\beta) \int_{T_0}^{T_m} \exp(-E/(kT'))dT' \right\}. \quad (5)$$

Defining dimensionless quantities  $u = E/kT$ ,  $u_0 = E/kT_0$ , and  $u_m = E/kT_m$  and expressing the temperature integral in terms of second exponential integral [12], one can write eqs. (4) and (5) as

$$I = \frac{s''c^2 \alpha \exp\left[(cs''E/(\beta k)) (E_2(u)/u - E_2(u_0)/u_0)\right] \exp(-u)}{\left\{ \exp\left[(cs''E/(\beta k)) (E_2(u)/u - E_2(u_0)/u_0)\right] - \alpha \right\}^2} \quad (6)$$

and

$$\left\{ \frac{k}{E} u_m^2 + \frac{cs''}{\beta} \exp(-u_m) \right\} \left[ \exp\left\{ \left( \frac{cs''E}{\beta k} \right) \left( \frac{E_2(u_m)}{u_m} - \frac{E_2(u_0)}{u_0} \right) \right\} - \alpha \right] = 2 \frac{cs''}{\beta} \exp(-u_m) \exp\left\{ \left( \frac{cs''E}{\beta k} \right) \left( \frac{E_2(u_m)}{u_m} - \frac{E_2(u_0)}{u_0} \right) \right\}. \quad (7)$$

Now, the fractional intensity  $x$  ( $x = I/I_m$ ) can be expressed as

$$x = B \frac{\exp\left[(cs''E/(\beta k)) (E_2(u)/u - E_2(u_0)/u_0)\right]}{\left\{ \exp\left[(cs''E/(\beta k)) (E_2(u)/u - E_2(u_0)/u_0)\right] - \alpha \right\}^2} \quad (8)$$

with

$$B = 4(cs''/\beta)^2 \alpha \left[ (k/E)^2 U_m^4 \exp(2u_m) - (cs''/\beta)^2 \right]^{-1}. \quad (9)$$

Using Newton-Raphson method [13], we can calculate  $T_m$  from eq. (7) and temperatures  $T_x^-$  on the rising side ( $T_x^- < T_m$ ) and  $T_x^+$  on the falling side ( $T_x^+ > T_m$ ) of TL peaks at fractional intensity  $x$  from eqs. (8-9). A plot of the pairs of variables  $\{u_m, u_x^-/(u_x^- - u_m)\}$ ,  $\{u_m, u_x^+/(u_m - u_x^+)\}$  and  $\{u_m, u_x^- u_x^+ / u_m (u_x^- - u_x^+)\}$ , where  $u_x^- = E/(kT_x^-)$  and  $u_x^+ = E/(kT_x^+)$  for values of  $u_m$  in the range  $10 \leq u_m \leq 40$ , are found to be linear so that we can write

$$u_m = C_1 (u_x^- / (u_x^- - u_m)) + D_1, \quad (10)$$

$$u_m = C_2(u_x^+ / (u_m - u_x^+)) + D_2, \tag{11}$$

and 
$$u_m = C_3(u_x^- u_x^+ / u_m (u_x^- - u_x^+)) + D_3. \tag{12}$$

A linear plot of  $u_m$  as a function of  $u_x^- / (u_x^- - u_m)$  for  $\alpha = 0.5$  and  $x = 0.5$  are shown in Figure 1. Similar results have been obtained for other pairs, namely  $[u_m, u_x^+ / (u_m - u_x^+)]$  and  $[u_m, u_x^- u_x^+ / u_m (u_x^- - u_x^+)]$  and different values of  $\alpha$ . The linear plot has been obtained for  $x = 0.2$  and  $x = 0.8$  also.

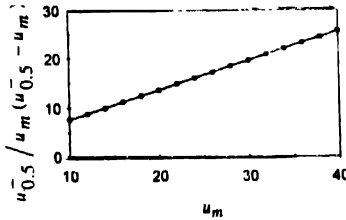


Figure 1. Variation of  $u_{0.5}^- / u_m (u_{0.5}^- - u_m)$  against  $u_m$  for  $\alpha = 0.5$ .

Eqs. (10–12) can be recast in terms of activation energy and temperatures as

$$E_1 = C_1 k T_m^2 / (T_m - T_x^-) + D_1 k T_m, \tag{13}$$

$$E_2 = C_2 k T_m^2 / (T_x^+ - T_m) + D_2 k T_m, \tag{14}$$

and 
$$E_3 = C_3 k T_m^2 / (T_x^+ - T_x^-) + D_3 k T_m. \tag{15}$$

The coefficients  $C_j$  and  $D_j$  ( $j = 1-3$ ) for a particular value of  $x$  occurring in eqs. (10–15) depend on  $\alpha$ . By using the method of non-linear least square regression [14], each of the coefficients  $C_j$  and  $D_j$  can be expressed as a quadratic function of  $\alpha$  ( $0 \leq \alpha \leq 1$ ) as

$$C_j = C_{j0} + C_{j1} \alpha + C_{j2} \alpha^2, \tag{16}$$

and 
$$D_j = D_{j0} + D_{j1} \alpha + D_{j2} \alpha^2. \tag{17}$$

The coefficients  $C_{jk}$  and  $D_{jk}$  ( $j = 1-3, k = 0-2$ ) occurring in eqs. (16–17) for  $x = 0.2, 0.5, 0.8$ , are presented in Table 1.

Table 1. Coefficients  $C_{jk}$  and  $D_{jk}$  ( $j = 1-3, k = 0-2$ ) occurring in eqs. (16–17) for  $x = 0.2, 0.5$  and  $0.8$  respectively.

$x$	$j$	$C_{j0}$	$C_{j1}$	$C_{j2}$	$D_{j0}$	$D_{j1}$	$D_{j2}$
0.2	1	2.5063	0.6373	-0.2518	-3.0022	-1.1847	0.3214
	2	1.4640	-0.1729	1.6549	-0.0261	2.0608	-1.8059
	3	3.9569	0.4113	1.4255	-1.7553	0.5179	-0.1439
0.5	1	1.4411	0.5256	-0.1947	-2.0763	1.1869	0.2053
	2	0.9861	0.3234	0.5607	-0.3134	1.7857	-2.3585
	3	2.4246	0.8274	0.3824	-1.3170	0.4891	-0.9844
0.8	1	0.7368	0.3503	-0.1139	-1.4845	-1.0331	-0.0315
	2	0.5858	0.3376	0.0868	-0.5091	0.7503	-1.7033
	3	1.3222	0.6839	0.0237	-1.0390	-0.0522	-0.8698

Eqs. (13–15) can be used to determine the activation energy of a TL glow curve.

3. Results and discussion

The activation energy of a TL peak can be calculated using eqs. (13–15) not only at a point where  $x = 0.5$  but at any other suitable points where  $x = 0.2$  and  $0.8$  also, using the values of  $C_{jk}$  and  $D_{jk}$  ( $j = 1-3, k = 0-2$ ) from Table 1 and eqs. (16–17) to obtain  $C_j$  and  $D_j$  ( $j = 1-3$ ).

Mixed order TL peaks have been generated numerically with  $E = 1.0$  eV,  $s = 10^{10} s^{-1}$ ,  $N = n_0 = 10^{10} cm^{-3}$ ,  $A_m = A_n = 10^{-7} s^{-1}$  and  $\beta = 1.0$  °C  $s^{-1}$  and different values of  $\alpha$ . Using eqs. (7–9), we have calculated  $T_m$ ,  $T_{0.5}^-$  and  $T_{0.5}^+$ . The values of  $\delta = T_{0.5}^+ - T_m$  and  $\omega = T_{0.5}^+ - T_{0.5}^-$  are presented in Table 2. The values of  $E_1, E_2$  and  $E_3$  obtained by using the present set of expressions (13–15) for  $x = 0.5$ , have been applied to the generated peaks and are presented in Table 2 along with the values of  $E$  and  $E_{IR}$  calculated using Chen's peak shape formulae [4] and initial rise method [9]. From Table 2, it is seen that the values

Table 2. Activation energies  $E_1$  (eV),  $E_2$  (eV) and  $E_3$  (eV) of numerically generated MO TL peaks ( $E_{in} = 1.0$  eV,  $s = 10^{10} s^{-1}$ ,  $A_m = A_n = 10^{-7} s^{-1}$ ,  $N = n_0 = 10^{10} cm^{-3}$ ,  $\beta = 1.0$  °C  $s^{-1}$  and different values of  $\alpha$ , using the present set of expressions (13–15) and Chen's formula [4].  $E_{IR}$  is the activation energies obtained by using initial rise method.

$E_{in}$ (eV)	$\alpha$	$T_m$ (°C)	$\delta$ (°C)	$\omega$ (°C)	$E_1$ (eV) $E_2$ (eV) $E_3$ (eV)			$E_1$ (eV) $E_2$ (eV) $E_3$ (eV) $E_{IR}$ (eV)				
					Present			Chen				
1.0	0.1	141.4272	15.1250	35.6503	0.4243	0.9973	0.9964	0.9968	0.9839	0.9854	0.9928	0.9890
1.0	0.2	151.4806	16.6453	38.6441	0.4307	1.0015	0.9992	1.0008	0.9696	0.9835	0.9833	0.9877
1.0	0.3	157.6403	18.0741	41.2074	0.4386	1.0027	1.0026	1.0032	0.9586	0.9831	0.9767	1.0000
1.0	0.4	162.1039	19.4828	43.6937	0.4483	1.0020	1.0046	1.0037	0.9519	0.9848	0.9736	0.9856
1.0	0.5	165.5575	21.2601	46.2173	0.4600	1.0002	1.0037	1.0023	0.9503	0.9885	0.9744	0.9860
1.0	0.6	168.2938	23.1268	48.8105	0.4738	0.9981	0.9934	0.9951	0.9547	0.9932	0.9790	0.9860
1.0	0.7	170.4603	25.1581	51.4298	0.4892	0.9981	0.9934	0.9951	0.9649	0.9975	0.9790	0.9861
1.0	0.8	172.1469	27.1922	53.9005	0.5045	0.9981	0.9981	0.9951	0.9790	1.0002	0.9946	0.9858
1.0	0.9	173.4261	28.8256	55.8208	0.5164	1.0028	1.0077	1.0059	0.9919	1.0007	1.0008	0.9857

of  $E$  obtained by using the present set of expressions are more accurate than those obtained by using Chen's formula and initial rise method. But the present expressions require the prior knowledge of  $\alpha$ . To find  $\alpha$  for an experimental peak, one has to calculate the shape factor  $\mu_g$  and derive  $\alpha$  from the  $\mu_g$  vs  $\alpha$  curve [6]. Chen *et al* [6] have presented  $\mu_g$  vs  $\alpha$  curve and observed that the curve is modified slightly with the change in  $E$  and  $s''$  i.e. with  $u_m$ , since  $T_m$  can be found for a particular  $E$  if  $s''$  is known. In Figure 2, we have presented the variation of  $\mu_g$  versus  $\alpha$  for  $u_m = 20$  and  $u_m = 40$ . The value of  $\mu_g$  decreases slightly by around 3% when  $u_m$  changes from 20 to 40 in agreement with the observation of Chen *et al* [6]. But it is observed that  $\alpha$  increases appreciably by around 0.1 to 0.2 when  $u_m$  changes from 20 to 40 for a particular value of  $\mu_g$ . For a TL peak since we do not have prior knowledge of  $u_m$  to find the accurate value of  $\alpha$  from  $\mu_g$  vs  $\alpha$  curve, the average values of  $\mu_g$  for  $u_m = 20$  and 40 are obtained for different values of  $\alpha$  and are plotted as a function of

$\alpha$  (solid line in Figure 2). The average  $\mu_R$  vs  $\alpha$  curve can now be used as a preliminary estimation of  $\alpha$  to find  $E$ . Taking into account of the error in the estimation of  $\alpha$  by this method, we have found that the possible error in the evaluation of the activation energy  $E$  does not exceed 3%. It is to be noted that for many experimental TL peaks,  $u_m$  lies between 20 to 40 except for a limited number of peaks.

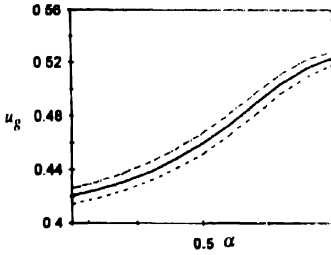


Figure 2. Variation of  $\mu_R$  (at  $x = 0.5$ ) as a function of  $\alpha$ . (a) - - - for  $u_m = 20$ , (b) - . - for  $u_m = 40$ , (c) ——— correspond to average of the values of  $\mu_R$  at  $u_m = 20$  and  $u_m = 40$ .

To show that MO kinetics model is a viable alternative to the GO kinetics model, we have generated numerically GO TL peaks with  $E = 1.0$  eV,  $s = 10^{13} \text{ s}^{-1}$ ,  $\beta = 1.0 \text{ }^\circ\text{C s}^{-1}$  and different values of  $b$  ( $1 < b < 2$ ) and computed the values of  $T_m$ ,  $T_{0.5}^-$ ,  $T_{0.5}^+$  and  $\mu_R$  (at  $x = 0.5$ ) using the expressions of Gartia *et al* [15] and also used the present set of expressions to obtain  $E_1$ ,  $E_2$  and  $E_3$  (Table 3). The values of  $\delta$  ( $\delta = T_{0.5}^+ - T_m$ ) and  $\omega$  ( $\omega = T_{0.5}^+ - T_{0.5}^-$ ) of the numerically generated GO TL peaks are given in Table 3. In computing the values of  $E$ , we have used Figure 2 (solid line) to obtain the values of  $\alpha$  from

Table 3. Activation energies  $E_1$  (eV),  $E_2$  (eV) and  $E_3$  (eV) of numerically generated GO TL peaks ( $E = 1.0$  eV,  $s = 10^{13} \text{ s}^{-1}$  and different values of  $b$ , calculated using present set of expressions (13–15))

$E$ (eV)	$s$ ( $\text{s}^{-1}$ )	$b$	$\delta$ ( $^\circ\text{C}$ )	$\omega$ ( $^\circ\text{C}$ )	$\mu_R$	$E_1$ (eV)	$E_2$ (eV)	$E_3$ (eV)
1.0	$10^{13}$	1.1	11.8	27.3	0.433	1.0398	1.0230	1.0323
1.0	$10^{13}$	1.2	12.7	28.5	0.446	1.0530	1.0345	1.0445
1.0	$10^{13}$	1.3	13.5	29.6	0.457	1.0559	1.0403	1.0485
1.0	$10^{13}$	1.4	14.4	30.7	0.468	1.0523	1.0374	1.0450
1.0	$10^{13}$	1.5	15.2	31.8	0.478	1.0457	1.0296	1.0376
1.0	$10^{13}$	1.6	16.0	32.9	0.487	1.0370	1.0157	1.0261
1.0	$10^{13}$	1.7	16.8	33.9	0.495	1.0285	1.0055	1.0165
1.0	$10^{13}$	1.8	17.6	34.9	0.503	1.0202	0.9985	1.0088
1.0	$10^{13}$	1.9	18.3	35.9	0.511	1.0120	0.9940	1.0025

the values of  $\mu_R$  ( $x = 0.5$ ). The values of  $E_1$ ,  $E_2$  and  $E_3$  agrees well with the input value  $E$ . Hence our present set of expressions can be used as an alternative method for finding the value of activation energy.

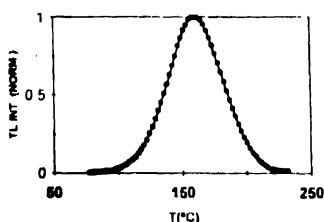
Finally, the applicability of MO kinetics model and the present set of expressions for determining the activation energy, is discussed by taking the well-studied experimental TL peak of BeO ( $T_m = 160.1^\circ\text{C}$ ) [11] irradiated with X-ray (5 minutes). Sakurai and Gartia [11]

fitted the peak with their numerically generated peak and obtained the activation energy  $E_n$  as 1.09 eV (Table 4). Now the present method for the determination of the activation energy

**Table 4.** The values of activation energies  $E_1$ ,  $E_2$  and  $E_3$  (eV) of the experimental TL peak of BeO.  $E_{mofc}$  and  $E_n$  are respectively the activation energies of the peak obtained by curve fitting with MO kinetics and numerical method [11].

$T_m$ (°C)	$T_{0.5}^-$ (°C)	$T_{0.5}^+$ (°C)	$\mu_g$ ( $x = 0.5$ )	$E_1$ (eV)	$E_2$ (eV)	$E_3$ (eV)	$E_{mofc}$ (eV)	$E_n$ (eV)
160.1	135.6	185.6	0.510	1.0418	1.0374	1.0397	1.0410	1.09

using the expressions [13–15] and half intensity points  $T_{0.5}^-$ ,  $T_{0.5}^+$ , has been applied to this peak. The value of  $\alpha$  of this peak used in the computation of the activation energies is observed from the Figure 2 (solid line) using the value of  $\mu_g$  (at  $x = 0.5$ ). The values of  $T_m$ ,  $T_{0.5}^-$ ,  $T_{0.5}^+$ ,  $\mu_g$  (at  $x = 0.5$ ),  $E_1$ ,  $E_2$  and  $E_3$  are given in Table 4. The experimental peak of BeO can be fitted with a MO kinetics peak using the values of  $E_{mofc} = 1.041$  eV,  $\alpha = 0.85$ ,  $s = 6.67 \times 10^{10} \text{ s}^{-1}$ ,  $A_m = A_n = 10^{-7} \text{ s}^{-1}$ ,  $N = n_0 = 10^{10} \text{ cm}^{-3}$  (Figure 3). The values of  $E_1$ ,  $E_2$  and  $E_3$  are in good agreement with the value of  $E_{mofc}$  and lies between the value of  $E_n$



**Figure 3.** Curve fitting of experimental TL peak (full circles) of BeO ( $T_m = 160$  °C) with MO kinetics (continuous line) ( $E = 1.041$  eV,  $s = 6.67 \times 10^{10} \text{ s}^{-1}$ ,  $A_m = A_n = 10^{-7} \text{ s}^{-1}$ ,  $N = n_0 = 10^{10} \text{ cm}^{-3}$  and  $\alpha = 0.85$ ).

obtained by Sakurai and Gartia and 0.98 eV obtained for the same peak by curve fitting with GO model.

#### 4. Conclusion

In the present paper, we have derived a set of expressions for the determination of activation energy of a TL peak obeying mixed order (MO) kinetics involving the important MO parameter  $\alpha$  which can be determined from the value of shape factor  $\mu_g$  at half intensity points. We have also applied the method to the experimental TL peak of BeO and obtained the activation energy. A comparison of the value of activation energy thus obtained, is made with the value obtained by using curve fitting technique with mixed order kinetics as well as general order kinetics model. It is observed that the values obtained in the present paper is comparable with the value obtained by Sakurai and Gartia [11].

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**References**

- [1] R Chen and Y Kirsh *Analysis of Thermally Stimulated Process* (Oxford : Pergamon) (1981)
- [2] M J Aitken *Thermoluminescence Dating* (New York · Academic) p 204 (1985)
- [3] K Mahesh and D R Vij *Techniques of Radiation Dosimetry* (Delhi · Wiley Eastern) (1985)
- [4] R Chen *J. Electrochem. Soc.* **116** 1254 (1969)
- [5] C E May and J A Partridge *J. Chem. Phys.* **40** 1401 (1964)
- [6] R Chen, N Kristianpoller, Z Davidson and R Visocekas *J. Lumin.* **23** 293 (1981)
- [7] A Halperin and A A Braner *Phys. Rev.* **117** 408 (1960)
- [8] J T Randall and M H F Wilkins *Proc. Roy. Soc.* **184** 366 (1945)
- [9] G F J Garlic and A F Gibson *Proc. Phys. Soc.* **60** 574 (1948)
- [10] D Yossian and Y S Horowitz *Radiation Measurement* **27** 465 (1997)
- [11] T Sakurai and R K Gartia *J. Phys* **D29** 271A (1996)
- [12] R K Gartia, S D Singh and P S Mazumdar *Phys. Stat Sol (a)* **138** 319 (1993)
- [13] W H Press, S A Teukolsky, W T Vetterling and B P Flannery *Numerical Recipes in Fortran* (Cambridge : Cambridge University Press)
- [14] E J Dudewicz and S N Misra *Modern Mathematical Statistics* (New York : Wiley) (1988)
- [15] R K Gartia, S J Singh and P S Mazumdar *Phys. Stat Sol (a)* **106** 291 (1988)