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Thermoluminescence Governed by Simultaneous Thermal Stimulation of Electrons and Holes

By

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The possibility of a simultaneous thermal release of electrons and holes in the thermoluminescence (TL) process was suggested in the early stages of the development of this discipline. However, most investigators utilized a model in which carriers of only one kind are thermally released from their "traps". In this model these carriers then radiatively recombine with carriers of opposite charge in "luminescence centers". This paper presents the results of a theoretical investigation into a more complex model, which allows the thermal release of electrons and holes in the same temperature range. The solution of a set of four simultaneous differential equations is required in this investigation. The equations are not amenable to analytical solution, so the equations are solved numerically for given values of the model parameters. Conclusions are drawn about the shapes of the TL curves, and the expected stability of the corresponding traps at given temperatures, and the apparent effective parameters namely, the activation energy, the frequency factor, and the order of kinetics, are obtained using the "general order" formalism. In all cases examined, there is a good three parameter fit to the TL peak generated by the examined model. This shows that even a rather general TL peak can be described quite accurately by the three parameter model. The activation energy determined using the three parameter analysis is found to agree rather well with the input activation energy of trapped electrons which is used in the original peak generation for the particular cases examined. In cases where the activation energy for holes is small, the glow peaks occur at relatively low temperatures, and are characterized by a very high effective frequency factor in the three parameter analysis. Furthermore, several cases are studied in which the calculated kinetics order is found to be less than 1.0.

La possibilité d'une liberation thermique simultanée des electrons at des trous dans le processus de thermoluminescence avait été suggérée dans les premières étapes du développement de cette discipline; mais la plupart des chercheurs ont préféré utiliser le modèle plus restrictif où un seul type de porteur de charge est libéré thermiquement des pièges pour ensuite se recombiner avec des porteurs de signe opposé dans les "centres". Dans cet article, nous traitons le cas plus complexe où les electrons et les trous peuvent être libérés dans le même domaine de température. Cela appelle la solution d'un systême de quatre equations differéntielles simultanées, ce qu'on ne sait bièn faire analytiquement. Au lieu de cela, c'est une methode numériqué que nous utilisons pour résoudre ce systême d'equations pour des valeurs données des paramètres. Nous sommes ainsi parvenu à des conclusions, concernant la forme des courbes de TL obtenues, la stabilité des pièges correspondants à attendre à des températures données, et les valeurs effectives des paramètres qu on pourrait déduire d une approche par la methode de "l'ordre général"; savoir, l'energie d'activation, le fac-

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teur de frequence et l'ordre de la cinétique. Dans tous les cas traités numériquement, un très bon accord a été trouve entre la courbe calculée à partir du système différentiel et la courbe a trois paramètres, ce qui nous montre qu'un pic de TL même d'un type fort général se laisse décrire assez exactement par le modèle à trois paramètres. Nous avons trouvé aussi que l'energie d'activation obtenue dans l'analyse a trois paramètres est fort voisine de l'énergie d'activation réelle des electrons. Dans les cas où l'energie d'activation des trous a été faible, le pics sont apparu à des températures relativement basses, cas caracterisé par un facteur de fréquence effectif de valeur élevée dans l'analyse à trois paramètres. En outre, plusieurs cas ont été appris où l'ordre de la cinétique calculée a été trouvé moins que 1,0.

1. Introduction

The usual model for a single thermoluminescence (TL) peak is that of an electron trapping state situated energetically close enough to the conduction band so that electrons can be released thermally in the temperature range of interest, and subsequently recombine with holes trapped in a "recombination center". The recombination center is usually assumed to be energetically far from the valence band so that no holes are released thermally into the valence band. Thus, only one channel of electron–hole recombination is taken into account. Sometimes, the possibility that thermally excited electrons can retrap into a trapping state of the same kind is included in the analysis. The analogous inverted process in which holes become thermally excited into the valence band and then recombine with electrons in recombination centers, may be as likely to occur as the previous one.

The traffic of carriers between the trapping state, conduction (or valence) band and recombination center was previously shown [1, 2] to be governed by a set of three simultaneous differential equations. The principal conclusion, drawn on the basis of simplifying assumptions, is that the order of kinetics depends on the relative effectiveness of the recombination and retrapping processes [1, 3]. The simplifying assumptions may, however, introduce inaccuracies which can be serious. Therefore, numerical solutions of the set of equations were obtained [4 to 6] without the use of simplifying assumptions, for specific sets of the trapping parameters, and a given heating function. The concentration of carriers in the band was also found as a function of temperature, which has an important bearing on the phenomenon of thermally stimulated conductivity.

In an attempt to describe most of the experimental TL peaks, the "general order kinetics" [7 to 9] was proposed, which is governed by the equation

$$I = -\frac{dn}{dt} = s'n^b \exp\left(-\frac{E}{kT}\right), \tag{1}$$

where I is the TL intensity, n the concentration of trapped carriers (cm^{-3}), s' the preexponential factor, E the the activation energy (eV), k the Boltzmann constant (eV K^{-1}), T the absolute temperature (K), and b the kinetics order. For first order kinetics [10], $b = 1$, for second order, $b = 2$. The solution of (1) was shown [9] to be

$$I = sn_0 \exp\left(-\frac{E}{kT}\right)\left[\frac{(b-1)\,s}{\beta}\int_{T_0}^{T}\exp\left(-\frac{E}{kT'}\right)dT' + 1\right]^{-b/(b-1)} \tag{2}$$

for $b \neq 1$, where $s = s'n_0^{b-1}$ has dimensions of s^{-1} and n_0 is the initial concentration of trapped carriers.

Rather early in the history of TL, it was suggested by Schön [11, 12] that electrons and holes can be thermally excited to the conduction and valence bands respectively in the same temperature range. This model complicates the physical picture and no

less than four differential equations can adequately describe it. Bräunlich and Schar-mann [13] studied these equations and gave approximate solutions. They classified the TL into two cases. In one case only electron transitions or only hole transitions contribute to the TL emission, the other being non-radiative or in another part of the spectrum. In the other case both recombinations contribute to the emission. In the former instance, on the basis of assumptions similar to those leading to first order kinetics in the "one channel" classical model, Bräunlich and Scharmann found an expression of the form

$$I(T) = \exp\left(-\frac{E}{kT}\right)\exp\left[\left(-\frac{s}{\beta}\right)\int_{T_0}^{T}\exp\left(-\frac{E_A}{kT'}\right)\mathrm{d}T'\right] \qquad (3)$$

which resembles an expression for the first order, except the activation energy for electrons, E, appears in the first exponent and that for holes, E_A, in the second.

The simultaneous thermal release of electrons and holes was further investigated by Gasiot and Fillard [14] and Mathur and Brown [15] in the context of simultaneous measurements of thermoluminescence (TL) and thermally stimulated conductivity (TSC). These authors reported a method for extracting information on hole emission by measuring the ratio of the TSC and TL signals.

In the investigation reported here, the set of four coupled differential equations governing the process of simultaneous electron and hole emission is solved numerically for the first time without simplifying assumptions. This enables the validity of the assumptions and approximations made in previous studies to be checked, and further-more, thermally stimulated conductivity (TSC) which often accompanies the TL phenomenon can be investigated in this framework.

The present paper, however, is limited to testing the validity of applying the con-ventional "three-parameter" equation ((1), the three parameters being E, s, b) to describe the glow peaks produced from this model. A TL peak is simulated using the examined model and analyzed by the conventional, somewhat oversimplified, three-parameter equation. The goodness of fit is determined graphically and the effective values of the parameters extracted from the analysis are compared with the known parameters introduced into the original set of equations.

The results obtained indicate that, in each of the cases, the three-parameter equa-tion can be used with confidence to describe the glow peaks produced, and the activa-tion energy so-calculated agrees well with the original value of the electron trap depth. Occasionally unusually high values of the effective frequency factor and values of kinetic order of less than 1.0 were obtained.

2. The Model and Solution

The model considered here includes one electron and one hole trapping state, as shown in Fig. 1. The four inter-related time (or temperature) dependent functions are the following concentrations (in cm^{-3}):

  n concentration of trapped electrons,
  m concentration of trapped holes,
  n_c concentration of electrons in the conduction band,
  p concentration of holes in the valence band.

The parameters involved are N, M concentrations (cm^{-3}) of electrons and holes trapping states, respectively; E_1, E_2 activation energies (eV) for the release of electrons and holes respectively; s_1, s_2 frequency factors (s^{-1}) for the release of electrons and holes, respectively; A_{re}, A_{rh} recombination probabilities ($cm^3\,s^{-1}$) for electrons and
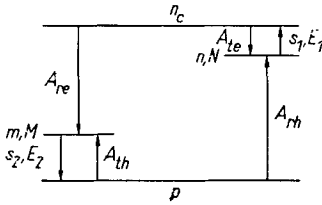
Fig. 1. Energy levels, concentrations and transition probabilities in the model

holes, respectively; A_{te}, A_{th} trapping probabilities (cm^3 s^{-1}) for electrons and holes, respectively.

The four simultaneous differential equations as first given by Bräunlich and Scharmann [13] (in a slightly different notation) are:

$$\frac{dn}{dt} = - s_1 \exp\left(-\frac{E_1}{kT}\right) n + A_{te} n_c (N - n) - A_{rh} n p \, , \tag{4}$$

$$\frac{dn_c}{dt} = s_1 \exp\left(-\frac{E_1}{kT}\right) n - A_{te} n_c (N - n) - A_{re} n_c m \, , \tag{5}$$

$$\frac{dp}{dt} = s_2 \exp\left(-\frac{E_2}{kT}\right) m - A_{rh} n p - A_{th} p (M - m) \, , \tag{6}$$

$$\frac{dm}{dt} = -s_2 \exp\left(-\frac{E_2}{kT}\right) m - A_{re} n_c m + A_{th} p (M - m) \, , \tag{7}$$

where k is the Boltzmann constant (eV K^{-1}) and T is the absolute temperature. It is quite obvious that this set of equations incorporates, necessarily, the neutrality condition

$$\frac{dn}{dt} + \frac{dn_c}{dt} = \frac{dm}{dt} + \frac{dp}{dt} \, . \tag{8}$$

The set of differential equations was solved numerically without approximations by utilizing a Runge-Kutta sixth-order predictor–corrector program that had been previously successfully employed [16] for evaluating the dose and dose rate dependences of TL. The main difference is that in the study presented here, two of the coefficients, exp $(-E_1/kT)$ and exp $(-E_2/kT)$, in the rate equations are very strongly temperature dependent. It was found that for a heating rate of 1 K/s, a temperature update every 0.1 s (i.e. 0.1 K) gives very good results. The integration interval was smaller, usually 0.001 s. In fact, the program is built in a dynamic manner such that the integration interval varies to ensure a pre-determined accuracy in the results. The program was run on a CDC 6500 computer. A typical run time for producing a glow peak consisting of a 50 K range was about 7 minutes. The resulting curves were analysed using a curve fitting program for the three parameters of (2) [3, 17].

3. Results

The number of possible choices for the sets of parameters that one can assume is, obviously, enormous. In particular, due to the rather preliminary nature of the present work, discussion is limited to a small number of cases chosen to exhibit some features that could not be accounted for by the conventional "one channel" model. Emphasis is placed on the cases where the TL is due to only one of the recombinations (e.g., electrons in the conduction band recombine with holes in centers) the other recombi-

Table 1

Parameters inserted into the simultaneous differential equations simulating TL of electrons recombination in the presence of a thermal release of holes. In all cases, no retrapping of holes is allowed ($A_{th} = 0$) and therefore no particular value is to be assigned to M

$\beta = 1$ K/s $E_1 = 1$ eV $s_1 = 10^{12}$ s^{-1} $s_2 = 10^{12}$ s^{-1}

$A_{re} = 10^{-13}$ cm^3 s^{-1} $A_{te} = 10^{-12}$ cm^3 s^{-1} $A_{rh} = 10^{-12}$ cm^3 s^{-1}

$N = 10^{15}$ cm^{-3} $m_0 = 10^{14}$ cm^{-3} $n_{c0} = 10^6$ cm^{-3} $p_0 = 10^6$ cm^{-3}

$n_0 = 10^{13}$ cm^{-3}

No.	E_2 (eV)	T_m (K)	E (eV)	b	s (s^{-1})
1	0.7	251	0.96	0.63	2.8×10^{19}
2	0.8	285	0.98	0.62	3.2×10^{16}
3	0.9	319	1.01	0.61	9.5×10^{16}
4	1.0	353	1.015	0.59	3.1×10^{13}
5	1.1	386	1.02	0.56	1.6×10^{12}
6	1.2	418	0.98	0.53	5.0×10^{10}
7	1.3	440	0.98	0.67	1.0×10^{10}
8	1.5	448	0.98	1.03	5.3×10^{9}

$n_0 = 10^{14}$ cm^{-3}

No.	E_2 (eV)	T_m (K)	E (eV)	b	s (s^{-1})
9	0.7	269	0.90	1.12	8.9×10^{15}
10	0.8	305	0.94	1.08	3.5×10^{14}
11	0.9	341	0.93	0.98	5.4×10^{12}
12	1.0	376	0.99	1.01	1.7×10^{12}
13	1.1	410	1.00	1.01	1.8×10^{11}
14	1.2	434	0.95	1.18	6.8×10^{9}
15	1.3	443	0.96	1.76	4.4×10^{9}
16	1.5	443	0.99	2.07	9.7×10^{9}

nation is assumed to be either radiationless or not detected. Of course, even in this case, the occurrence of the second recombination is of great importance since it may influence substantially the balance of carriers and therefore, the rate of the recombination which yields TL. In many of these calculations hole retrapping was ignored (i.e. $A_{th} = 0$) and therefore the total number of hole traps, M, was unimpotant. Thus the number of parameters was reduced by two.

This was done simply to reduce the number of parameters involved and future work will include hole retrapping. In the present work emphasis is put on the apparently dominant parameters which are expected to have maximum impact on the results. In the present situation the electron and hole activation energies E_1 and E_2 are expected to play the dominant role. Of particular importance is the relative magnitude of E_1 and E_2. In the computational work, therefore, $E_1 = 1.0$ eV is kept constant and E_2 is varied.

Table 1 summarizes some of the computational results obtained. The parameters which were not varied are listed in the caption. The table is divided into two groups of results. In the first, the peak follows practically first order kinetics when there is a very low hole emission ($E_2 = 1.5$ eV). In the second, the peak is very close to second order kinetics under the same circumstances. The difference between the two cases
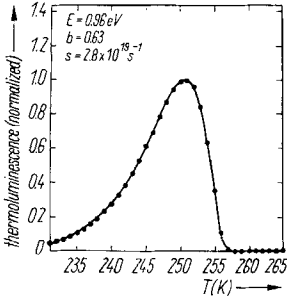
Fig. 2

Fig. 3

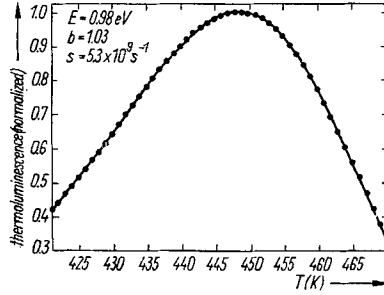Fig. 2. First order conditions with strong hole emission; the resulting peak has an effective order of 0.63

Fig. 3. First order conditions with very small hole emission; the resulting peak has an effective order of 1.03

is that for $m_0 = 10^{14}$ cm^{-3}, the first group has $n_0 = 10^{13}$ cm^{-3} whereas in the second, $n_0 = 10^{14}$ cm^{-3}. The table gives the values of E_2 and T_m, the peak temperature, as well as the resulting "three parameters", E, s' and b (order of kinetics) found by curve fitting.

Fig. 2 shows the results for row 1 in Table 1. The dots are the glow peak points simulated by the simultaneous differential equations whereas the solid line is the best fit three parameter curve to the simulated glow peak. The fit is remarkably good. The effective activation energy was found to be 0.96 eV, reasonably close to the value of 1.0 eV inserted into the equations. The quite rapid fall-off of the peak at high temperatures deserves some attention. This is directly related to the resulting order of kinetics. Orders of kinetics smaller than 1 have been reported in other thermally stimulated processes such as thermogravimetry (e.g. see p. 89 in [3]). May and Partridge [8] reported an order of kinetics of 0.7 ± 0.1 for the TL of NaCl, KCl and KBr. Qualitatively, the rapid fall on the high temperature side is related to the thermal exhaustion of holes from the traps. The fact that a peak with relatively high activation energy occurs at such a low temperature is accounted for, in the three parameter presentation, by an unusually high effective frequency factor, 2.8×10^{19} s^{-1}.

Fig. 3 is generated from the data in row 8 of Table 1. Since the activation energy for holes is very high, its influence on the peak shape is minor as compared to the case of no thermal hole emission. The best fit yields values for the effective activation energy of 0.98 eV, order of kinetics of 1.03 and frequency factor of 5.3×10^9 s^{-1}.

Fig. 4 shows the curve for row 9 in the table. Although the situation without hole emission is basically that of second order, with hole emission (and $E_2 = 0.7$ eV) the shape is close to that of first order kinetics, the effective order being 1.12. The effective activation energy in this case is 0.9 eV, 10% below the original electron activation energy. In the cases checked, the initial rise analysis revealed that the increase of the intensity function is dominated by the electron activation energy, 1.0 eV. This is the value which would have been obtained with the initial rise method in the very low temperature range. The effective frequency factor is very high here too, due to the combination of a rather high activation energy with a relatively low temperature of the maximum.

Fig. 5 is generated from the data in row 16. Due to the very high activation energy for the release of holes, 1.5 eV as compared to the 1.0 eV for the electrons, the peak appears to be close to second order (2.07), an order expected with practically no
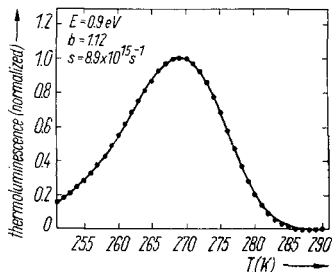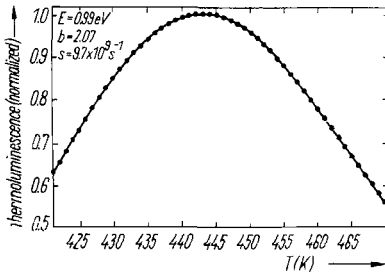
Fig. 4

Fig. 5

Fig. 4. Second order conditions with strong hole emission; the resulting peak has an effective order of 1.12

Fig. 5. Second order conditions with very small hole emission; the resulting peak has an effective order of 2.07

thermal release of holes. The effective activation energy was found to be 0.99 eV, in very good agreement with the inserted electron activation energy.

Another interesting simulation can be performed using the same program. This involves the variation of the emitted light and the trapping concentrations of holes with time while at a constant temperature. In the present program, the phosphorescence behavior is studied by setting the heating rate $\beta = 0$.

Fig. 6 depicts the decay of phosphoresence at 450 K for the trapping parameters in row 14. Curve 1 shows the decay of phosphorescence whereas curve 2 shows the decay of the concentration of holes in recombination centers. No simple analytical function is offered for these decay curves.

Fig. 7 gives the decay curve at 270 K with the trapping parameters of row 9 in Table 1. In this case the plot of both the phosphorescence and the concentration $m(t)$ are straight lines on the semi-log scale, indicating an exponential decay, though with
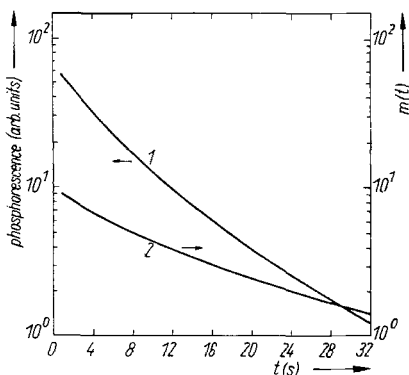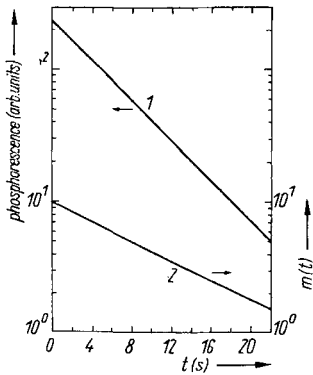

Fig. 6

Fig. 7

Fig. 6. Decay curves of phosphorescence (1) and concentration of carriers in centers (2); the parameters are these of row 14 in Table 1, and the decay curve is calculated for 450 K

Fig. 7. Decay curves of phosphorescence (1) and concentration of carriers in centers (2); the parameters are those of row 9 in Table 1, and the decay curve is calculated for 270 K

different slopes in curves 1 and 2. This is opposite to that of the simple first order case in which both the phosphorescence and the concentration of carriers decay at the same rate.

4. Discussion

A model for the thermoluminescence process governed by simultaneous thermal stimulation of electrons and holes is investigated here by numerically solving four coupled rate equations without simplifying assumptions. When the thermal hole emission was substantial, mainly due to low activation energy for holes, the calculated peak occurred at relatively low temperatures, and had an effective cut-off on the high temperature side. In all the cases examined it was possible to fit the calculated glow peak to a three parameter function. This yielded an effective activation energy, frequency factor and order of kinetics. In those peaks which displayed a rapid cut-off on the high temperature side the order of kinetics was much lower than otherwise expected (down to an effective order of 0.53 for the case of row 6 of Table 1). This result suggests an explanation for the experimental result by Partridge and May [8] who reported an effective order of kinetics of 0.7 ± 0.1 for some alkali halides. The present model cannot be utilized in its current form for the case of alkali halides, however. The actual energy band diagram in these materials is almost certainly much more complex than the simple picture described here, and in fact the TL production mechanism may even involve localized transitions between energy levels rather than transitions into and out of the delocalized bands. However, the results do highlight the feasibility of obtaining a kinetic order of less than 1.0.

Regarding the cut-off of TL peaks discribed above it is to be noted that a similar cut-off has been discussed in a theoretical paper by Halperin and Braner [2]. These authors discuss qualitatively the cut-off of different peaks due to a shortage of carriers in the luminescence centers. The shortage they discuss is due, however, to a low initial concentration in the centers rather than to a thermal release of these carriers as discussed in the present work.

Fig. 6 and 7 deserve comment. As seen in Table 1, and discussed above, the effective activation energy turned out to be rather close to the original value of the activation energy for trapped electrons, the largest discrepancy being 10% in the case of row 9 in Table 1. Fig. 7 shows the isothermal decay with time of both $m(t)$ and the phosphorescence intensity for this case. The time constant for the decay of $m(t)$ is governed primarily by the activation energy for the release of holes (0.7 eV) whereas the time constant for the phosphorescence decay is related to the effective activation energy for luminescence (viz. 0.9 eV). Thus, one might expect that the phosphorescence decay would be *slower* than the decay of $m(t)$. However, it can be seen in Fig. 7 that the phosphorescence dacay is *faster* than the decay of $m(t)$. At first sight this might seem "anomalous" in that the luminescence is decaying faster than it should (eg. see Visocekas and Geoffroy [18]). However, it must be remembered that the *effective* frequency factor is unusually high resulting in a TL peak which appears at quite a low temperature (269 K). Thus, the decay rate for phosphorescence is entirely consistent with the TL peak position, and in this respect is not anomalous at all.

For the case of row 14, Table 1, the decay rate for $m(t)$ is expected to be less than that for the phosphorescence, as is observed in Fig. 6.

It is useful to recall that high values for the frequency factor have been reported on several occasions (e.g. [19]). Although it would be unwise at this stage to claim that simultaneous hole and electron release is the cause of these high values (other mechanisms must also be considered (e.g. [20]), it is important to emphasize that high frequency factor values can emerge from applying the simplified three-parameter

analysis to this more complicated model. The important question is, can the three-parameter analysis be used to predict the thermal stability of the luminescence despite the existence of a more-complex TL model? The results of Fig. 6 and 7 appear to indicate that it can. Future work is planned on the isothermal decay properties of phosphorescence and the stability of TL peaks in order to test this point further.

Another limitation allowed in this study, besides the neglect of hole retrapping mentioned earlier, was the fact that only the recombination of free electrons with holes in centers was considered to yield TL. This may have excluded interesting results. In fact, it has been suggested [13] in the past that either this recombination, or the recombination of free holes with trapped electrons, or the sum of the two (and possibly some weighted sum) could yield the TL. Finally, it is to be noted that the functions $n_c(t)$ and $p(t)$, namely concentrations of free electrons and holes in the conduction and valence bands respectively, can be obtained from the present computation. The relation of these functions to the measurable thermally stimulated conductivity is to be explored in the future.

References

[1] E. ADIROVITCH, J. Phys. Radium 17, 705 (1956).
[2] A. HALPERIN and A. A. BRANER, Phys. Rev. 117, 408 (1960).
[3] R. CHEN and Y. KIRSH, Analysis of Thermally Stimulated Processes, Pergamon Press, Oxford 1981.
[4] P. KELLY, M. J. LAUBITZ, and P. BRÄUNLICH, Phys. Rev. B 4, 1960 (1971).
[5] D. SHENKER and R. CHEN, J. Computer Phys. 10, 272 (1972).
[6] S. HARIDOSS, J. Computer Phys. 26, 232 (1978).
[7] C. E. MAY and J. A. PARTRIDGE, J. chem. Phys. 40, 1401 (1964).
[8] J. A. PARTRIDGE and C. E. MAY, J. chem. Phys. 42, 797 (1965).
[9] R. CHEN, J. Electrochem. Soc. 116, 1254 (1969).
[10] J. T. RANDALL and M. H. F. WILKINS, Proc. Roy. Soc. A184, 347 (1945).
[11] M. SCHÖN, J. Phys. Radium 17, 689 (1956).
[12] M. SCHÖN, Tech.-wiss. Abh., Osram-Studienges. 7, 175 (1958).
[13] P. BRÄUNLICH and A. SCHARMANN, phys. stat. sol. 18, 307 (1966).
[14] J. GASIOT and J. P. FILLARD, J. appl. Phys. 48, 3171 (1977).
[15] V. K. MATHUR and M. D. BROWN, J. appl. Phys. 54, 5485 (1983).
[16] R. CHEN, S. W. MCKEEVER, and S. A. DURRANI, Phys. Rev. B 1, 24, 4931 (1981).
[17] D. SHENKER and R. CHEN, J. Computer Phys. 10, 272 (1972).
[18] R. VISOCEKAS and A. GEOFFROY, phys. stat. sol. (a) 41, 499 (1977).
[19] S. P. KATHURIA and C. M. SUNTA, J. Phys. D 12, 1573 (1979).
[20] P. D. TOWNSEND, G. C. TAYLOR, and M. C. WINTERSGILL, Radiat. Eff. 41, 11 (1979).