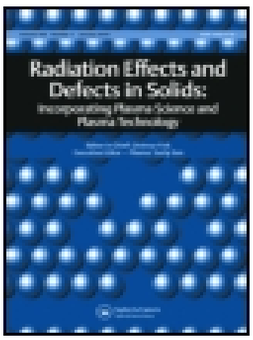

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A novel algorithm to solve the differential equation describing the non-interactive multiple-trap system model in thermoluminescence

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ABSTRACT

The analysis of glow curves in thermoluminescence requires finding a closed expression for the light emitted as a function of the temperature and the parameters characterizing trap and recombination centers. Since it is not possible to derive a closed expression from the set of coupled differential equations describing the thermoluminescence phenomenon, approximations are made even for the simplest models. All of them resort to an approximation known as the quasi-equilibrium approximation (QE), and to further approximations. In this article, an algorithm is reported that permit the derivation of a closed expression for the emitted light for the model known as the non-interactive multi-trap system (NMTS) model by resorting only to the QE approximation. It is shown that the integration of the first order differential equation related to the NMTS model can be replaced by finding the roots of an analytical expression.

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Thermoluminescence; glow curve analysis; new algorithm

1. Introduction

Thermoluminescence is a form of luminescence that is exhibited by some materials, which after having been irradiated with ionizing radiation, emit light upon heating. Different experiments have been developed to investigate the physics involved in the emission of light. The most usual experiment consists of recording the emitted light when a sample is heated with a constant heating rate, say, $T = T0 + \beta \cdot t$, where T stands for the temperature in Kelvin, t for the time and β for the heating rate. $T0$ is the temperature at which the recording of a glow curve starts. The resulting curve is called glow curve.

In order to describe the physics involved in thermoluminescence, several models have been put forward. The simplest one is that shown in Figure 1. It is known as one trap-one recombination center (OTOR model).

A way to find the values of the parameters, namely, recombination and retrapping probabilities, activation energies and frequency factors, consists of fitting a theoretical expression for the emitted light, say $I_{th}(T,\alpha)$, to the glow curve $I_{exp}(T)$ (*1*). α stands for the set of parameters. A closed expressions for $I_{th}(T,\alpha)$ cannot be found from the set of

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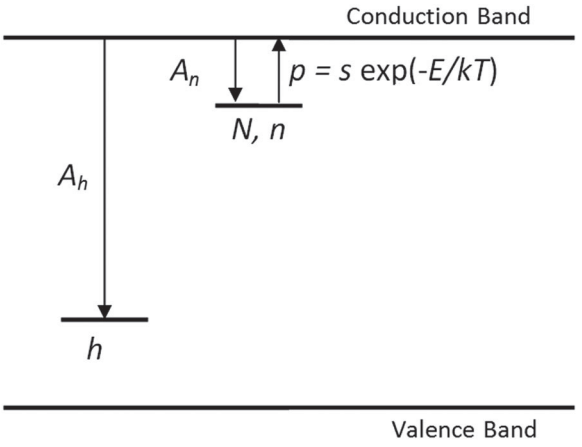

Figure 1. OTOR model. A_h is the recombination probability, h is the concentration of holes in the recombination centers, A_n is the retrapping probability, N is the concentration of traps, n is the concentration of trapped electrons, s is the frequency factor, E the activation energy, and k is the Boltzmann constant. The product $s \exp(-E/kT)$ is the escape probability of an electron from a trap.

differential equations describing the carrier traffic among the trap, the conduction band and the recombination center. Thus approximations have been made in order the get a closed expression for $I_{\text{th}}(T,\alpha)$. All the models put forward so far rely on the quasi-equilibrium approximation (QE). The QE approximation assumes that $\frac{dn_c}{dt} \cong 0$ and $n_c \ll n$. With n_c the concentration of electrons in the conduction band is indicated. If recombination prevails over retrapping, namely $(N - n(T))A_n \ll n(T)A_h$, then an equation is obtained, called first order kinetics (FO) (*2*). The light intensity is given by:

$$I(T) = n0.\frac{s}{\beta}\exp\left(\frac{-E}{kT}\right)\exp\left(-\frac{s}{\beta}\int_{T0}^{T}\exp\left(-\frac{E}{ku}\right)du\right), \tag{1}$$

$n0$ stands for the initial concentration of trapped electrons, and $T0$ for the temperature at which the recording of the glow curve starts. This kinetics was put forward in 1945 by Randall and Wilkins (*3*).

On the contrary, if retrapping prevails over recombination the light intensity is given by:

$$I(T) = n0^2 s' \exp\left(\frac{-E}{kT}\right)\left[1 + \frac{n0s'}{\beta}T\int_{T0}^{T}\exp\left(-\frac{E}{ku}\right)du\right]^2. \tag{2}$$

Garlick and Gibson put forward this kinetics, called second-order (SO) kinetics (*4*).

Since a closed expression cannot be derived when recombination does not prevail over retrapping, or retrapping over recombination, May and Partridge put forward a heuristic expression with the aim of describing kinetics comprised between first and second order (*5*):

$$I(T) = n0s''\exp\left(-\frac{E}{kT}\right)\left[1 + (b-1)\frac{s''}{\beta}\int_{T0}^{T}\exp\left(-\frac{E}{ku}\right)du\right]^{-b/(b-1)}. \tag{3}$$

In this equation s'' stands for $(sA_n n_0^{b-1}/NA_h)$, and b is a parameter loosely related to the retrapping probability. When $b \to 1$ Equation (3) converges to FO kinetics, and when $b = 2$,

to SO kinetics. This kinetics is known as the general order (GO) kinetics, and it is nowadays the most employed kinetics for analyzing glow curves. This kinetics may fail to yield the correct parameters (*6*, *7*). For this reason, recently a closed expression for the light intensity has been derived from the system of coupled differential equations describing the OTOR model in terms of the two real branches of the transcendental Lambert function (*8*). The closed expression is obtained by resorting to the QE approximation. An expression for the light intensity has been derived for a more general model described in the next section, known as non-interactive multi-trap system (NMTS), also in terms of the Lambert function (*9*). A further assumption, reported below, was made in addition to the QE approximation. Which branch of the Lambert function should be considered depends on the ratio $R = A_n/A_h$. Since R is unknown the analysis of a glow curve becomes difficult. For this reason, we report a new expression for the light intensity, which overcomes this difficulty. Furthermore, the new expression resort only to the QE approximation, and not to an additional approximation as the expression reported in reference (*9*).

2. The non-interactive multi-trap system

A model, known as the interactive multi-trap system (IMTS), is depicted in Figure 2. It is based on the band model of crystals, and localized levels in the band gap. This model includes a thermally disconnected trap, namely, a center that can capture electrons, but cannot release electrons for the temperature interval the glow curve is recorded. In fact, the glow curve will change from measurement to measurement, except that the thermally disconnected trap is fully occupied. When the thermally disconnected trap is fully occupied, the model is known as the NMTS. The concentration of thermally disconnected traps is indicated with M. The rest of the parameters shown in Figure 2 have the same meaning as those shown in Figure 1.

The equations describing the carrier traffic are:

$$\frac{dn(t)}{dt} = -n(t) \cdot s \cdot \exp\left(-\frac{E}{kT(t)}\right) + A_n \cdot [N - n(t)] \cdot n_c(t), \tag{4}$$

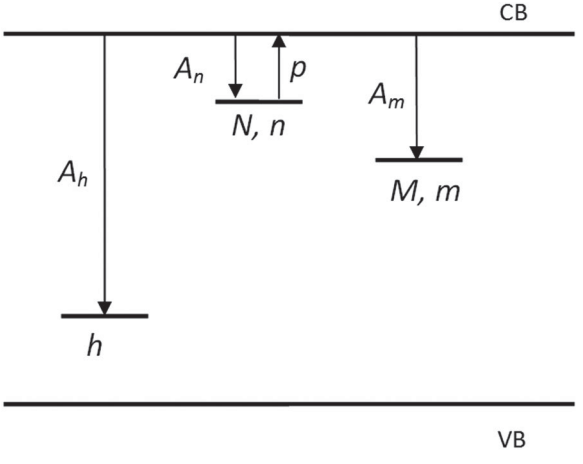

Figure 2. Sketch of the IMTS model.

$$\frac{\mathrm{d}h(t)}{\mathrm{d}t} = -A_h \cdot n_c(t) \cdot h(t), \tag{5}$$

$$h(t) = n(t) + n_c(t) + M, \tag{6}$$

$$I(t) = -\frac{\mathrm{d}h(t)}{\mathrm{d}t}, \tag{7}$$

In the equations, n_c stands for the concentration of electrons in the conduction band. Equation (7) is the thermoluminescent light, and Equation (6) represents charge conservation. If a probe is heated with a constant heating rate β, namely, $T(t) = T0 + \beta t$, then Equations (4–7) read:

$$\frac{\mathrm{d}n(T)}{\mathrm{d}T} = -n(T) \cdot \frac{s}{\beta} \cdot \exp\left(-\frac{E}{kT}\right) + \frac{A_n}{\beta} \cdot [N - n(T)] \cdot n_c(T), \tag{8}$$

$$\frac{\mathrm{d}h(T)}{\mathrm{d}T} = -\frac{A_h}{\beta} \cdot n_c(T) \cdot h(T), \tag{9}$$

$$h(T) = n(T) + n_c(T) + M, \tag{10}$$

$$I(T) = -\beta \frac{\mathrm{d}h(T)}{\mathrm{d}T}, \tag{11}$$

$T0$ stands for the temperature at which the recording of a glow curve starts.

As aforementioned, from the set of coupled differential equations, it is not possible to derive a close expression for $I_{\mathrm{th}}(T,\alpha)$. Resorting to the QE approximation in reference (*1*) it is shown that the derivative of the trapped electrons with respect to the temperature is given by:

$$\frac{\mathrm{d}n}{\mathrm{d}T} = -\frac{n(T) \cdot \frac{s}{\beta} \exp(-E/kT) h(T) A_h}{(N - n(T))A_n + h(T)A_h}, \tag{12}$$

Since $ncn(T)$. Because of the QE approximation $h(T) = n(T) + M$. Thus

$$\frac{\mathrm{d}h(T)}{\mathrm{d}T} = \frac{\mathrm{d}n(T)}{\mathrm{d}T}$$

because M is constant in the NMTS model. Since $I(T) = -\frac{\mathrm{d}h(t)}{\mathrm{d}t} = -\beta \frac{\mathrm{d}h(T)}{\mathrm{d}T}$ we have:

$$I(T) = \frac{n(T).\frac{s}{\beta} \exp\left(-\frac{E}{kT}\right)(n(T) + M)}{(N - n(T))R + n(T) + M}, \tag{13}$$

R, as before, stands for A_n/A_h.

In order to obtain an analytical expression for the light intensity equation 12 should be integrated.

3. Expression for the emitted light in terms of the Lambert function

If recombination does not prevail on retrapping, or retrapping on recombination, A. M. Sadek et al. derived an equation for $I_{th}(T,\alpha)$ by assuming that the expression

$$\phi(T) = \frac{n(T)}{h(T)}, \tag{14}$$

can be replaced by an effective value ϕ_{eff} independent of the temperature (9). This is a strong assumption if $M(T) > > n(T)$. Because of the QE approximation $h(T) = n(T) + M$. Thus $\phi(T) = \frac{n(T)}{n(T)+M}$. If $M \approx 0$ the approximation holds ($\phi(T) \approx 1$ for all T), but if $M(T) > > n(T)$, then $\phi(T) \approx \frac{n(T)}{M}$. Thus $\phi(T) = n(T)/(n(T) + M)$. If $M \approx 0$ the approximation holds ($\phi(T) \approx 1$for all T), but if $M(T) > > n(T)$, then $\phi(T) \approx n(T)/M$. Thus $\phi(T)$ will change between $n0/M$ for $T = T0$ and ≈ 0 for the highest temperature the glow curve was recorded.

The light intensity for $R < 1$ is given by:

$$I(t) = \frac{N \cdot R \cdot \phi_{eff}}{\beta(1 - \phi_{eff} \cdot R)^2} \cdot \frac{s \exp(-E/kT)}{W_0[\exp(z_1)] + W[\exp(z_1)]^2}, \tag{15}$$

$$z_1 = \frac{1}{\varepsilon} + \ln(\varepsilon) + \frac{\phi_{eff} \cdot s}{1 - \phi_{eff} \cdot R} \int_{T0}^{T} \exp\left(-\frac{E}{k \cdot u}\right) du, \tag{16}$$

$$\varepsilon = \frac{h0 \cdot (1 - \phi_{eff} \cdot R)}{N \cdot R}, \tag{17}$$

$W_0(\exp(z))$ is the principal branch of the Lambert-W function. and for $R > 1$

$$I(t) = \frac{N \cdot R \cdot \phi_{eff}}{\beta(1 - \phi_{eff} \cdot R)^2} \cdot \frac{s \exp(-E/kT)}{W_0[-1, \exp(-z_2)] + W[-1, \exp(z_2)]^2}, \tag{18}$$

$$z_2 = \frac{1}{|\varepsilon|} + \ln(|\varepsilon|) + \frac{\phi_{eff} \cdot s}{1 - \phi_{eff} \cdot R} \int_{T0}^{T} \exp\left(-\frac{E}{k \cdot u}\right) du, \tag{19}$$

$W_0(-1, -\exp(z))$ is the second branch of the Lambert-W function.

Since the parameter R is unknown, an initial step is required in order to determine which one of the two expressions shall be used. This makes the calculations cumbersome.

In the next section, an algorithm is reported, which allows the integration of Equation (12) without resorting to approximations, such as that of a φ_{eff}, and has an unique expression for the emitted light whatever the value of R.

4. New algorithm

The fraction of the occupation of a trap, indicated with $x(T)$, is given by

$$x(T) = \frac{n(T)}{N} \tag{20}$$

Then Equation (12) becomes:

$$\frac{dx(T)}{dT} = -\frac{s/\beta \exp(-E/kT)x(T) \cdot (x(T) + M')}{(1 - x(T))R + x(T) + M'}, \tag{21}$$

$$M' = \frac{M}{N}.$$

Since the right side of Equation (21) is always negative, $x(T)$ is a steady decreasing function of temperature. This result will be employed below.

Equation (21) can be written:

$$\frac{[(1 - x(T))R + x(T) + M']}{x(T) \cdot (x(T) + M')} \mathrm{d}x = -\frac{s}{\beta} \exp\left(-\frac{E}{kT}\right) \mathrm{d}T. \tag{22}$$

Integration of Equation (22) gives:

$$\left(\frac{R}{M'} + 1\right) \ln\left(\frac{x(T)}{x0}\right) - \frac{R}{M'}(1 + M') \ln\left(\frac{x(T) + M'}{x0 + M'}\right) = -\frac{s}{\beta} \int_{T0}^{T} \exp\left(-\frac{E}{k \cdot T'}\right) \mathrm{d}T'. \tag{23}$$

The parameter $f(T)$ is defined as $f(T) = \frac{x(T)}{x0}$. $f(T)$ varies between 1 and 0 as the temperature increases. $x0 = x(T_R)$, and T_R is the room temperature.

Then Equation (23) turns into

$$\left(\frac{R}{M'} + 1\right) \ln(f(T)) - \frac{R}{M'}(1 + M') \ln\left(\frac{x0 \cdot f(T) + M'}{x0 + M'}\right) + \frac{s}{\beta} \int_{T0}^{T} \exp\left(-\frac{E}{k \cdot u}\right) \mathrm{d}u = 0. \tag{24}$$

We define the following functions:

$$G(f, x0, R, M) = \left(\frac{R}{M'} + 1\right) \ln(f) - \frac{R}{M'}(1 + M') \ln\left(\frac{x0 \cdot f + M'}{x0 + M'}\right), \tag{25}$$

and

$$U(T, s, \beta, E) = \frac{-s}{\beta} \int_{T0}^{T} \exp\left(-\frac{E}{k \cdot u}\right) \mathrm{d}u. \tag{26}$$

Thus, Equation (24) can be written:

$$G(f, x0, R, M') = U(T, s, \beta, E). \tag{27}$$

The equation $G(f, x0, R, M)$ takes the value 0 for $f = 1$, and $-\infty$ for $f = 0$. Therefore $G(f, x0, R, M)$ varies between 0 and $-\infty$ as f varies from 1 to 0. Since $U(T, s, \beta, E)$ is always negative, there is always a solution for Equation (27), and it is unique.

Figure 3 shows the function $G(f,x0,R,M')$ for the following values of the parameters: $x0 = 0.01$, $M' = 100$, $A_n = 4 \times 10^{-7}$ cm^3/s, $N = 10^{10}$ 1/cm^3, and $R = 4$ ($A_n = 1 \times 10^{-7}$ cm^3/s).

For instance, the value of the function $U(T; 10^{12}1/s, 1\text{ K}\,1/s, 1\text{ eV})$ for $T = 397$ K is -3. Thus taking $G(f,0.01,4,100) = -3$ it results follows that $f = 0.053$.

The value of f for a given set of parameters can be computed for any temperature by resorting to a root finder, as shown in the next section.

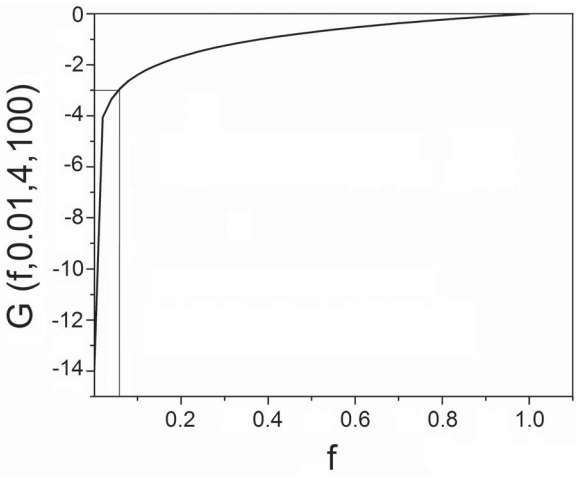

Figure 3. $G(f,0.01,4,100)$ versus f. For $T = 397$ K the value of $U(397\,\text{K}, 10^{12}\,\text{1/s}, 1\,\text{K/s}, 1\,\text{eV}) = -3$ is shown in the figure along with the corresponding value of $f = 0.053$.

5. Computing *f* by means of a root finder

For the parameters employed for computing Figure 3 Equation (27) becomes:

$$G(f, 0.01, 4, 100) = 1.04 \ln(f) - 4.04 \ln\left(\frac{0.01f + 100}{100.01}\right) = -10^{12} \int_{300}^{T} \exp\left(-\frac{1\text{eV}}{k \cdot T'}\right) \mathrm{d}T'.$$

For a given temperature T the right-hand side will give a number. For instance, for $T = 400$ K the result is 3.853. Thus we have:

$$1.04 \ln(f) - 4.04 \ln\left(\frac{0.01f + 100}{100.01}\right) + 3.853 = 0.$$

The value of f can be found by finding the root of the equation above. One of the most efficient algorithms is the Brent algorithm (*10*). This algorithm requires indicating the interval where the root lies. Since the values of f lies between 0 and 1, this interval should be chosen. The Brent's method relies mainly on the bisection method. This algorithm repeatedly bisects an interval and then selects a subinterval in which a root lies for further processing. It is a very simple and robust method, but it is also relatively slow. Thus, the Brent's algorithm tries to use the potentially fast converging secant method or inverse quadratic interpolation if possible, but it falls back to the more robust bisection method if necessary.

Resorting to the algorithm described above $f(T)$ can be computed for a given set of parameters E, s, R, M and $x0$: T is given a value, and $f(T)$is the root of Equation (24). If the left side of Equation (24) is indicated with $F(f,R,M',s,E,x0,T)$, formally $f(T)$ can be written as:

$$f(T) = \text{root}[F(f, R, M', s, E, x0, T), f, 0, 1]. \tag{28}$$

Equation (28) should be interpreted as follows: $F(f,R,M',s,E,x0,T)$ is the function, the root of which should be found for f in the interval 0–1 after T is given a value.

6. Expression for the light intensity

From Equations (13), (20) and (21) the following expression for the light intensity can be derived:

$$I(T) = \frac{s/\beta \ \exp(E/kT)x0f(T) \cdot (x0f(T) + M')}{(1 - x0f(T))R + x0f(T) + M'}. \tag{29}$$

This equation, along equation with Equation (28), permits computing the theoretical glow curve. The next step consists in finding out the set of parameters such as the theoretical glow curve coincides with the experimental one. This is usually done by employing the Levenberg–Marquardt algorithm (*1*).

7. Comparison of both algorithms

In terms of the Lambert function, for $R > 1$ the concentration of holes in the recombination center is given according to reference (*9*) by:

$$h(T) = \frac{-h0}{\varepsilon \cdot W(-1, -\exp(z_2)}, \tag{30}$$

ε is given by Equation (17), and z_2 by Equation (19).

Thus, the concentration of trapped electrons is:

$$n(T) = \frac{-h0}{\varepsilon \cdot W(-1, -\exp(z_2))} - M. \tag{31}$$

For the algorithm put forward in this article $n(T)$ is given by:

$$n(T) = f(T) \cdot x0 \cdot N. \tag{32}$$

The set of couple differential Equations (4–6) was solved for the following set of parameters: $s = 10^{12}$ 1/s, $E = 1$ eV, $A_m = 4{\cdot}10^{12}$ cm^3/s, $A_n = 10^{12}$ cm^3/s, $N = 10^{12}$ 1/cm^3 and $M = 10^{12}$ 1/cm^3.

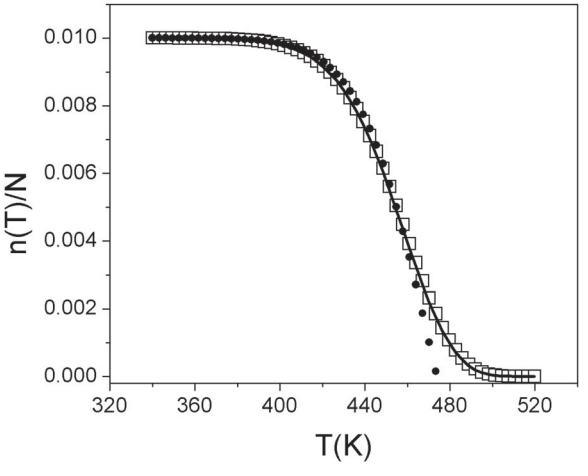

Figure 4. $n(T)/N$ computed from the set of differential equations (open square), Equation (31) (solid circle), and Equation (32) (solid line).

According to the set of parameters ϕ_{eff} varies between 0.5 and 0.

Figure 4 shows $n(T)$ computed from the set of Equations (4–6), and Equations (31) and (32). ϕ_{eff} was taken to be 0.5 because it yields the best fit of $n(T)/N$ computed with Equation (31) to that obtained from the set of differential equations.

As expected, the curve resulting from Equation (32) coincides with the concentration of trapped electrons computed from the set of differential equations, while the concentration of trapped electrons given by Equation (31) differs from the true value as from 460 K.

8. Fitting procedure

In order to find the parameters characterizing traps and recombination centers the first step consists in choosing an expression for the light intensity (Equations 1, 2, 3 or 29). Once an expression has been chosen the following function is constructed:

$$S(\alpha) = \sum_{m=1}^{L} (I_{\text{exp}}(T_m) - I_{\text{theor}}(T_m, \alpha))^2,$$

where $I_{\text{exp}}(T)$ stands for the experimental glow curve measured at L temperatures T_m ($m = 1,2,..,L$), $I_{\text{theor}}(T,\alpha)$ for the theoretical expression for the light intensity and α for the set of parameters characterizing the chosen kinetics. For example, if expression (29) is chosen for the light intensity, α stands for the set of parameters E, s, R, M' and $x0$.

The fitting proceeds by choosing a set of guess parameters α, and employing the Levenberg–Marquardt algorithm, as explained in detail in reference (*2*), section 6.3. This algorithm is iterative, namely, starting with a set of guess values the algorithm computes a new set which decreases $S(\alpha)$. The process is repeated until a minimum is reached. When a minimum is reached, the FOM should be calculated. If it is lower than 5% the fitting is considered acceptable. If the FOM is higher than 5%, a new set of guess parameters should be chosen.

9. Analysis of glow curves with the new algorithm

Recently Sadek et al. reported that for $R > 1$ and $x0 = 1$ (saturated trap) the General Order kinetics (GO) fails (*11*). A similar result is reported by M. Karmakar et al. (*12*). They investigated the suitability of two methods known as the modified peak shape method (MPS) and the Kirsh method by considering numerically computed thermoluminescence peaks. For the NMTS model they found that for a saturated trap and $A_n > A_h$, and for $R = 100$, both methods yield quite inaccurate activation energies. For $E = 1$ eV, $s = 10^{12}$ 1/s, $N = 10^{10}$ 1/cm^3, $M = 10^{12}$ 1/cm^3, $A_n = 10^{-7}$ cm^3 1/s and $A_h = 10^{-9}$ cm^3 1/s the computed activation energies with the MPS method and Kirsh method are 0.515 and 0.543 eV respectively (see Table 2, line 5 of reference (*12*)). We analyzed the peak with Equation (28) and found the following values: $E = 1.0$ eV, $s = 1.1 \times 10^{12}$ 1/s, $R = 100$, $x0 = 1$ and $M = 1 \times 10^{12}$ 1/cm^3. These values agree quite well with the input values of the differential equations. The FOM is 2%.

Figure 5 shows the glow curve along with both the fitted curve and the residual, and Figure 6 the electron concentration in the trap and conduction band. As can be seen, the quasi-equilibrium holds. Since this is the only approximation made in deriving Equation (28), the NMTS model yields reliable parameters.

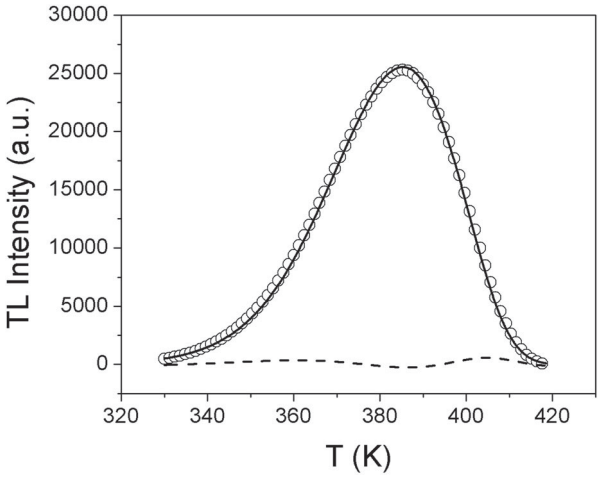

Figure 5. Computed glow curve for the following parameters: $E = 1$ eV, $s = 10^{12}$ 1/s, $N = 10^{10}$ 1/cm^3, $M = 10^{12}$ 1/cm^3, $A_n = 10^{-7}$ cm^3 1/s, and $A_h = 10^{-9}$ cm^3 1/s (open circle), fitted curve (solid line), and residual (dash line). The FOM is 2%.

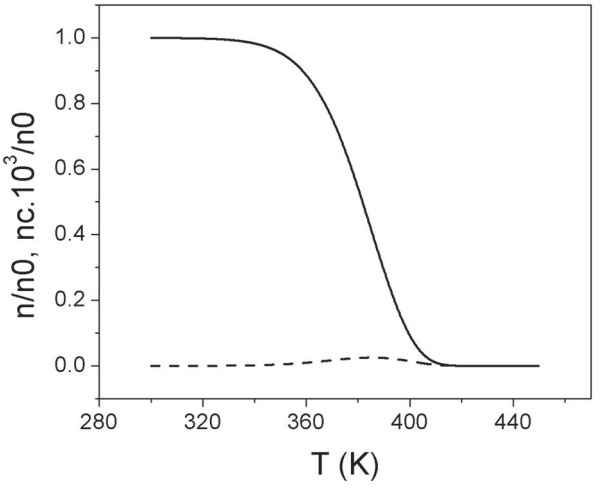

Figure 6. Normalized concentration $n/n0$ of electrons in the trap (solid line), and normalized concentration $nc/n0$ of electrons in the conduction band (dash line). Since nc is negligible against n the figure shows $(n/n0).10^3$.

We have analyzed the computed glow curve shown in Figure 5 with both versions of the GO kinetics, namely, the original and most used version (*13*) and the version due to Rasheedy (*14*). The parameters are shown in Table 1.

The results show that the old version yields parameters that differ from those employed for generating the glow curves, while the version due to Rasheedy gives parameters close to the true ones. The fact that the GO kinetics due to Rasheedy yields better results than the original version has been reported previously (*6*).

Table 1. Parameter values obtained after fitting the computed glow curve shown in Figure 5 by resorting to the original GOK model (*13*) and to the version due to Rasheedy (*14*).

	E (eV)	*S* (1/s)	*b*	FOM (%)
Original version	0.85	7.5×10^9	1.5	3.1
Rasheedy	0.94	1.3×10^{11}	1.2	4.2

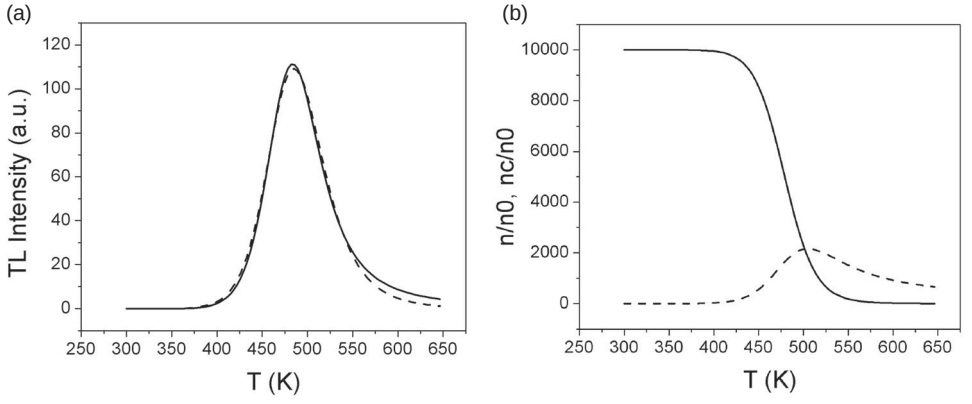

Figure 7. (a) computed glow curve (solid line), and glow curve obtained by fitting (dash line). FOM = 6%. (b) normalized concentration of electrons in the trap (solid line) and in the conduction band (dash line).

The difference between both versions of the GO kinetics and the NMTS model is that the latter provides physically meaningful parameters, such as *R* and *M*, while both GO kinetics give the *b* parameter, which has no physical significance.

When the QE approximation does not hold, the parameters resulting by employing Equation (29) are not accurate.

The glow curve for the following parameters was computed: $E = 1.04$ eV, $s = 1 \times 10^{12}$ 1/s, $N0 = 10^{10}$ 1/cm^3, $n0 = 10^8$ 1/cm^3, $A_n = 4 \times 10^{-9}$ cm^3/s, $A_h = 1 \times 10^{-9}$ cm^3/s ($R = 4$) and $M = 0$. The parameters obtained by fitting Equation (29) to the glow curve are: $E = 0.90$ eV, $s = 1.7 \times 10^{11}$ 1/s, $n0 = 9.6 \times 10^7$ 1/cm^3 and $R = 14.8$. The computed energy differs from the true one by 14%, and the value of *R* is about 3.7 times larger than the correct one. The computed glow curve and that obtained by fitting Equation (29) are shown in Figure 7(a). Figure 7(b) shows the normalized concentration of electrons in the trap and in the conduction band. It is clear from the figure that the QE approximation does not hold.

The wide of a peak depends on both the retrapping factor *R* (the higher the retrapping, the wider the peak), and the concentration of electron n_c in the conduction band (the higher n_c the wider the peak). Since Equation (29) was derived under the assumption that n_c is negligible against the concentration *n* of electrons in the trap, the widening of the peak employing Equation (29) is reflected in the retrapping factor $R = 14.8$, which is larger than the correct one ($R = 4$).

Glow curves generated with different retrapping factors *R*, lower and higher than 1, have been analyzed, and the results coincide with those employed for computing the glow curves as long as the QE equilibrium approximation holds. As illustration Figure 8 shows the glow curve computed for $A_h = 4 \times 10^{-7}$ cm^3/s, $A_n = 10^{-7}$ cm^3 /s,

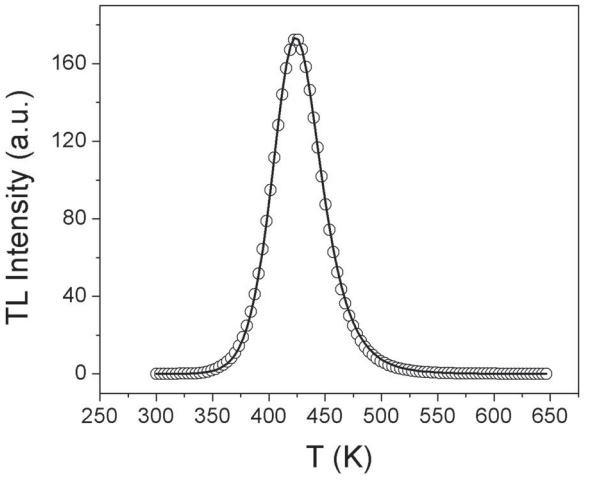

Figure 8. Computed glow curve for $R = 0.25$ (open circle) and fitted glow curve (solid line). FOM = 1.9%.

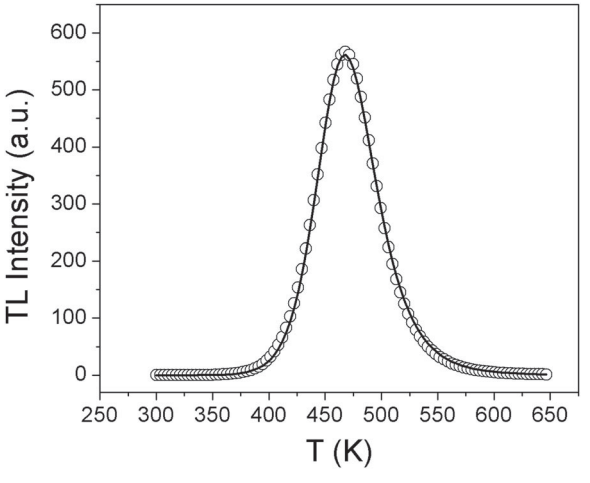

Figure 9. Computed glow curve for $R = 4$ (open circle) and fitted glow curve (solid line). FOM = 2.1 %.

$N = 10^{10}$ 1/cm^3, $n0 = 10^8$ 1/cm^3 and $M = 0$ ($R = 0.25$), and Figure 9 for $A_h = 1 \times 10^{-7}$ cm^3 /s, $A_n = 4 \times 10^{-7}$ cm/s, $N = 10^{10}$ 1/cm^3, $n0 = 10^8$ 1/cm^3 and $M = 0$ ($R = 4$). In both cases, the QE approximation holds, a requirement for the validity of Equation (29).

Since the GO model fails for $R \approx 100$, for the parameters $E = 1.00$ eV, $s = 1 \times 10^{12}$ 1/s, $N0 = 10^{10}$ 1/cm^3, $n0 = 10^8$ 1/cm^3, $A_n = 4 \times 10^{-9}$ cm^3/s, $A_h = 1 \times 10^{-7}$ cm^3/s ($R = 100$) and $M = 10^{12}$ 1/cm^3 the glow curve was computed and fitted with Equation (29). All the parameters resulting from the fitting coincide with those employed for generating the glow curve (the FOM is 3%). It is worth mentioning that the QE approximation holds.

As to the possibility of applying the new algorithm derived for the NMTS model with the QE approximation we investigated whether a criterion, which has been put forward for assessing whether the QE holds when the GO kinetics is employed (*6*), is also valid for

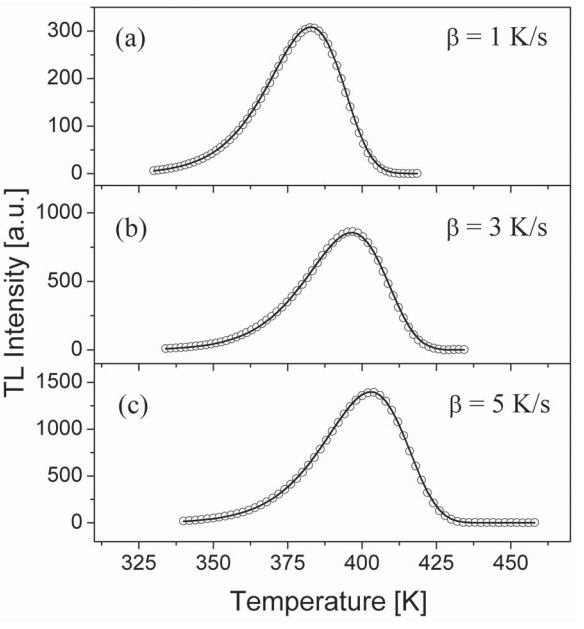

Figure 10. Computed glow curves for different heating rates (open circle) and fitted glow curves (solid line).

Equation (29). Glow curves should be recorded with three different heating rates, say 1, 3 and 5 K/s and the set of parameters for each heating rate computed. If the computed sets of parameters for each heating rate coincide, or differ little, except the frequency factor, which might change an order of magnitude, the QE approximation holds, and the computed parameters are reliable. The rationale behind the criterion is that an increase in the heating rate will produce and increase in the concentration n_c of electrons in the conduction band. If n_c is negligible for the three heating rates the QE approximation holds.

As an example, glow curves were computed for $E = 1$ eV, $s = 10^{12}$ 1/s, $A_h = 4 \times 10^{-7}$ cm^3/s, $A_n = 10^{-7}$ cm^3/s, $N = 10^{10}$ 1/cm^3, $n0 = 10^8$ 1/cm^3, $M = 10^{12}$ 1/cm^3 ($M' = 100$), and three heating rates, say 1, 3 and 5 K/s. Figure 10 shows the three glow curves, and the fitted curves. The concentration of electrons in the conduction band is negligible against the concentration of trapped electrons for the three heating rates. The resulting parameters coincide for the three heating rates: $E = 0.96$ eV, $s = 3 \times 10^{12}$ 1/s, $x0 = 0.01$, $R = 4$ and $M' = 100$. The FOM's for the three heating rates are lower than 1.6%.

10. Summary

A closed equation for the emitted light was derived for the NMTS model by resorting only to the QE approximation, which can be computed with a root finder. It offers the following advantages when compared to Equations (15) and (18): (1) no approximation is made, such as that done for deriving Equations (15) and (18) (φ_{eff}), and (2) since the value of R is not known in advance (it is a parameter that should be found by the fitting of the theoretical expression for the light intensity to the experimental glow curve), the use of Equation (15) and (18) is difficult. On the contrary, Equation (28) is valid whatever the value of R.

Finally a criterion is given to assess the reliability of the computed parameters found with the NMTS model.

Disclosure statement

No potential conflict of interest was reported by the authors.

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