# MATHEMATICAL ASPECTS OF GROUND STATE TUNNELING MODELS IN LUMINESCENCE MATERIALS

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# Mathematical aspects of ground state tunneling models in luminescence materials

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ABSTRACT

Luminescence signals from a variety of natural materials have been known to decrease with storage time at room temperature due to quantum tunneling, a phenomenon known as anomalous fading. This paper is a study of several mathematical aspects of two previously published luminescence models which describe tunneling phenomena from the ground state of a donor–acceptor system. It is shown that both models are described by the same type of integral equation, and two new analytical equations are presented. The first new analytical equation describes the effect of anomalous fading on the dose response curves (DRCs) of naturally irradiated samples. The DRCs in the model were previously expressed in the form of integral equations requiring numerical integration, while the new analytical equation can be used immediately as a tool for analyzing experimental data. The second analytical equation presented in this paper describes the anomalous fading rate (*g*-Value per decade) as a function of the charge density in the model. This new analytical expression for the *g*-Value is tested using experimental anomalous fading data for several apatite crystals which exhibit high rate of anomalous fading. The two new analytical results can be useful tools for analyzing anomalous fading data from luminescence materials. In addition to the two new analytical equations, an explanation is provided for the numerical value of a constant previously introduced in the models.



## 1. Introduction

During the past decade significant progress has been made both experimentally and theoretically in understanding the behavior of luminescence signals from feldspars, apatites and other natural materials. Quantum mechanical tunneling and the associated phenomenon of "anomalous fading" of these luminescence signals are now well established as dominant mechanisms in these materials [1–13]. Two types of tunneling processes have been investigated experimentally and by the development of appropriate models. The first type of quantum mechanical tunneling is considered to take place directly from the ground state of the trap [14–20], and has been shown to be associated with a power-law type of decay of the luminescence signal [21–24].

The second type of quantum mechanical tunneling is considered to take place via the excited state of the system of electron–hole pairs, and is commonly associated with the description of continuous-wave infrared stimulated luminescence signals (CW-IRSL) from feldspars. Experimental and modeling work by several researchers has provided valuable information on the origin of these IRSL signals, and supports the existence of tunneling processes involving localized recombinations taking place from the excited state of the trap, as well as charge migration through the conduction band-tail states. [9,10,22,25–37].

There have been several efforts during the past 40 years to develop models for luminescence signals originating in a random distribution of donor–acceptor pairs, especially in connection with kinetics of chemical reactions [28–30,11]; and references therein).

This paper is concerned with two previously proposed relatively simple models for luminescence signals in feldspars, namely the two ground state tunneling models by Huntley [24] and by Li and Li [17]. These two models are shown schematically in Fig. 1a and b respectively. The model by Jain et al. [11] is also shown for comparison purposes in Fig. 1c, and is considered in the Discussion section of this paper. Huntley [24] presented a model for quantum tunneling shown in Fig. 1a, which explained the power-law decay of luminescence and also proposed using an analytical equation expressing the concentration of charge carriers in the ground state $n(t)$ during geological time scales. The model by Huntley [24] introduced an numerical constant with a value of 1.8 in order to describe accurately the time development of $n(t)$. However, the value of this constant has not been explained previously, and one

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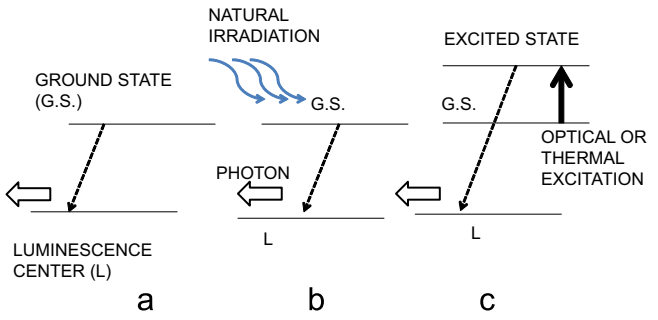


**Fig. 1.** Schematic depiction of the three luminescence models considered in this paper: (a) The ground state tunneling model of Huntley [24]. (b) The more general ground state tunneling model of Li and Li [17], in which anomalous fading and natural irradiation are taking place simultaneously. (c) The excited state tunneling model of Jain et al. [11].

of the goals of this paper is to provide an analytical explanation for its value.

Huntley and Lian [31] suggested an extension of the model by Huntley shown schematically in Fig. 1b, which uses a differential equation to describe simultaneous natural irradiation and anomalous fading effects on the luminescence of feldspars. Kars et al. [19] applied the model by Huntley [24] and Huntley and Lian [31] to construct unfaded and natural dose response curves (DRCs) of IRSL signals. On the basis of their modeling and experimental results, Kars et al. [19] proposed a new correction method for anomalous fading effects, and their model predicted the anomalous fading rate for samples which had reached field saturation.

Li and Li [17] applied the model of Huntley and Lian [31] in an extensive experimental and modeling study of both laboratory-irradiated and naturally irradiated feldspars. These authors studied several aspects of the model: the decay of IRSL signals, the effects of anomalous fading on the shape of dose response curves (DRCs), the probability distribution of trap-to-center distance and the dependence of anomalous fading on the parameters of the model. Both the experimental data and the model showed that the fading rates depend strongly on the size of radiation dose previously received by the samples. The modeling results of Li and Li were expressed in terms of integral equations which require numerical integration over the donor–acceptor distances in the model. One of the main goals of this paper is to show that the integral equations for DRCs in the model of Li and Li [17] can be replaced with analytical equations.

The main purpose of this paper is to examine several mathematical aspects of the two models by Huntley [24] and Li and Li [17] and to develop new analytical equations.

The specific goals of this paper are:

(a) To examine the mathematical similarities between the two models. It is shown that anomalous fading phenomena in both models are described by the same type of integral equation.
(b) To develop new analytical expressions for the DRCs in the model of Li and Li [17]. These authors presented DRCs in the form of integral equations, while the new equations developed in this paper are in analytical form; the new DRC equations in this paper are useful tools for data analysis.
(c) To provide an analytical explanation for the numerical value of 1.8 for a constant introduced in the model of Huntley [24]. To the best of our knowledge, no previous analytical explanation has been provided in the literature for the value of this constant.
(d) To develop a new analytical equation describing the anomalous fading rate (*g*-Value per decade) as a function of the charge density in the models, under specific assumptions.
(e) To test the equations developed in part (d) by fitting experimental data for several types of apatites which are known to exhibit high anomalous fading effects.

## 2. The two models of anomalous fading in feldspars

The purpose of this section is to summarize the two models shown in Fig. 1, to point out their mathematical similarities, and to develop new analytical equations when possible.

### 2.1. The model of Huntley [24]

Huntley [24] discussed the tunneling mechanism for a random distribution of holes and electrons in feldspars, based on transitions taking place directly from the ground state of the system. The instantaneous concentration of electrons in the ground state is denoted by $n(r',t)$ and depends both on the elapsed time $t$ and on the dimensionless separation distance parameter $r'$ between donor and acceptor. This distance parameter is defined by $r'=(4\pi\rho/3)^{1/3}r$, where $r$ represents the actual donor–acceptor separation distance and $\rho$ represents the actual number density of acceptors per unit volume. The dimensionless number density of acceptors parameter $\rho'$ is defined by $\rho'=(4\pi\rho/3)\alpha^{-3}$, where $\alpha$ is the potential barrier penetration constant Huntley, [24]. The tunneling lifetime $\tau$ in this model is given by:

$$\tau = s^{-1}\exp\left(\frac{r'}{(\rho')^{1/3}}\right), \tag{1}$$

where $s$ is the frequency factor characterizing the tunneling process taking place from the ground state of the system.

The distribution of electrons in the ground state $n(r',t)$ varies with the distance parameter $r'$ and with time $t$ according to:

$$\frac{n(r',t)}{n_o} = 3(r')^2\exp\left[-(r')^3\right]\exp[-t/\tau] \tag{2}$$

where $n_0$ is the total concentration of carriers at time $t=0$. By combining Eqs. (1) and (2):

$$\frac{n(r',t)}{n_o} = 3(r')^2\exp\left[-(r')^3\right]\exp\left[-st\exp\left(-(\rho')^{-1/3}r'\right)\right] \tag{3}$$

Fig. 2(a) shows an example of the distribution $n(r',t)$ obtained using Eq. (3) as a function of the dimensionless distance $r'$, and at a time $t=1000$ s after the start of the tunneling process (dashed line). The solid line in Fig. 2(a) indicates the initial peak-shaped symmetric distribution $P(r')=n(r',0)=3(r')^2\exp[-(r')^3]$ at time $t=0$. The values of the parameters used in Fig. 2 are typical for feldspars, $\rho'=3\times10^{-6}$ and $s=3\times10^{15}$ s$^{-1}$. The sharply rising part of the dashed line in Fig. 2(a) represents the "moving tunneling front" in the tunneling process. The characteristic shape of this tunneling front is the product of the two functions appearing in Eq. (3), namely of the sharply rising double exponential function $\exp[-st\exp(-(\rho')^{-1/3}r')]$ and of the symmetric distribution $3(r')^2\exp[-(r')^3]$. These two functions are shown in Fig. 2(b) for the data in Fig. 2(a).

The instantaneous total concentration of remaining carriers $n(t)$ is calculated by integrating Eq. (3) over all possible distances $r'$:

$$\frac{n(t)}{n_o} = \int_0^\infty 3(r')^2\exp\left[-(r')^3\right]\exp[-t/\tau]dr'. \tag{4}$$

In the approximate semi-analytical version of the model of Huntley [24], one introduces a critical lifetime $\tau_c$ and a corresponding critical radius $r_c'$, which describes the behavior of the physical system. From a physical point of view, this approximation means that for times $t<\tau_c$ no tunneling has occurred, while for

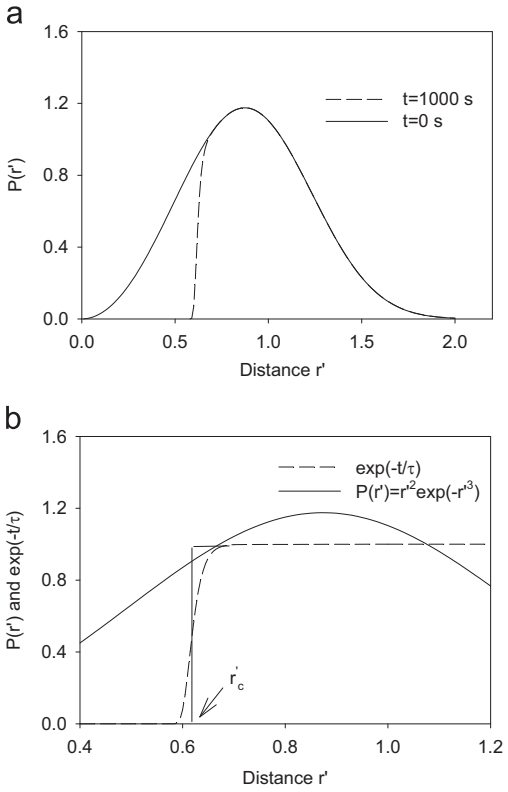


**Fig. 2.** (a) An example of the distribution $n(r',t)$ obtained using Eq. (3) as a function of the dimensionless distance $r'$, and at a time $t=1000$ s after the start of the tunneling process (dashed line). The solid line indicates the initial peak-shaped symmetric distribution of distances at time $t=0$. The values of the parameters are $\rho'=3\times10^{-6}$, $s=3\times10^{15}$ s$^{-1}$. (b) The sharply rising dashed line in (a) is the "moving tunneling front", obtained as the product of the two functions appearing in Eq. (3), as discussed in the text. The critical radius $r_c'$ corresponds to the approximation of replacing the dashed line by a vertical line.

times $t>\tau_c$ all electrons have tunneled out. Geometrically, this approximation corresponds to replacing the dashed lines in Fig. 2 (b) by a vertical line. Mathematically the value of the critical distance can be estimated from the *inflection point* or from the *half-way point* of the double exponential function $\exp[-t/\tau]$ in Eq. (3) [28–30,33]. By taking the second derivative of $\exp[-t/\tau]$ in Eq. (3) with respect to $r'$and setting it equal to zero, one obtains easily the critical radius

$$r_c' = (\rho')^{1/3}\ln(st) \quad . \tag{5}$$

This is the same value as the one obtained in Huntley [24] and by other authors by using physical arguments. The same equation can be obtained by considering the half-way or 50% reduction point of the luminescence signal. Using this value of $r_c'$ one can now approximate the integral in Eq. (4), by replacing the exponential $\exp[-t/\tau]$ with a value of 0 for $t<\tau$ and with a value of 1 for $t\geq\tau$, and by carrying out the integration:

$$\frac{n(t)}{n_o} = \int_0^\infty 3(r')^2\exp\left[-(r')^3\right]\exp[-t/\tau]dr' \simeq \int_{r_c'}^\infty 3(r')^2\exp\left[-(r')^3\right]dr' = \exp\left[-(r')^3\right]\Big|_{r_c'}^\infty \tag{6}$$

and using Eq. (5):

$$\frac{n(t)}{n_o} = \exp\left[-\rho'\ln(st)^3\right]. \tag{7}$$

Huntley [24] pointed out that this expression is a good approximation for large values of the parameter $st$, but a much better numerical approximation is obtained at all values of $st$ by

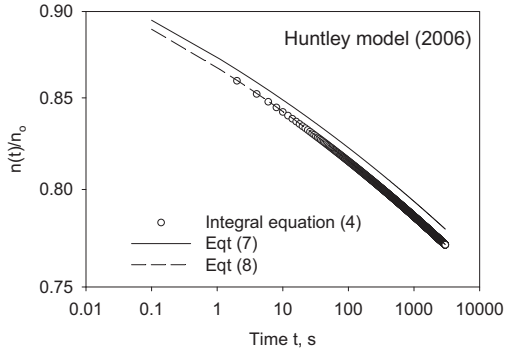


**Fig. 3.** Comparison of the concentration $n(t)$ obtained by numerically integrating Eq. (4) over the possible range of values of distances $r'$, with the analytical expressions in Eqs. (7) and (8). Eq. (7) (solid line) overestimates the result of the numerical integration, while Eq. (8) provides an accurate description of the numerical integration (dashed line).

using the following slightly modified version of Eq. (7):

$$\frac{n(t)}{n_o} = \exp\left[-\rho'\ln(zst)^3\right] \tag{8}$$

where $z=1.8$ is a correction factor which was introduced arbitrarily by Huntley [24], in order to describe accurately the function $n(t)$ obtained by numerically integrating Eq. (4). The value of this constant $z=1.8$ has been used by several researchers [11, 19, 24]. Jain et al. (2012, 2015) provided a physical meaning for this factor $z$, as representing the time derivative of the critical lifetime $\tau_c$, i.e. $z\approx d\tau_c/dt$. However, its numerical value of 1.8 has not been explained previously by using analytical considerations. Such an analytical explanation for the value $z=1.8$ is presented in Appendix A.

Fig. 3 shows a comparison of the concentration $n(t)$ obtained by numerically integrating Eq. (4) over the possible range of values of distances $r'$, with the analytical expressions in Eqs. (7) and (8). The simulation in Fig. 3 shows clearly that Eq. (7) (solid curve) overestimates the result of the numerical integration from Eq. (4) (open circles), while Eq. (8) provides an accurate description of the numerical integration (dashed curve).

### 2.2. The model of Li and Li (2008)

Li and Li [17] extended the model by Huntley [24] and investigated the effects of anomalous fading on the shape of dose response curve (DRCs) under natural irradiation. The results of Li and Li [17] are expressed in terms of equations which require numerical integration over the distances $r'$ in the model. In this section new analytical equations are presented for the model of Li and Li [17].

These authors considered a feldspar sample which undergoes two physical processes opposing each other; the sample is naturally irradiated in nature over geological times, while at the same time tunneling is taking place over this irradiation period. These opposing processes can be expressed by a single differential equation whose solution for a constant distance $r'$ is given by:

$$\frac{n(r')}{N(r')} = \frac{\dot{D}\tau}{\dot{D}\tau + D_0}\left(1 - e^{-\frac{\dot{D}\tau + D_0}{D_0\tau}t}\right), \tag{9}$$

where $\dot{D}$ is the natural irradiation rate, $D_0$ is the characteristic unfaded dose, $N(r')$ is the maximum possible concentration of carriers corresponding to the total number of trapping sites for a given distance $r'$, and the tunneling lifetime $\tau(r')$ is found from Eq. (2) as before. Using $t=D_n/\dot{D}$ where $D_n$ is the paleodose and by

defining an effective characteristic dose $D'_0$:

$$D'_0 = \frac{\dot{D}\tau D_0}{\dot{D}\tau + D_0}, \tag{10}$$

Eq. (9) becomes

$$\frac{n(r')}{N(r')} = \frac{D'_0}{D_0}\left(1 - e^{-\frac{D_n}{D'_0}}\right). \tag{11}$$

This is the saturating exponential function derived by Li and Li [17], which expresses how $n(r')$ fills up with the paleodose $D_n = \dot{D}t$. It is noted that the lifetime $\tau(r')$ and the effective characteristic dose constant $D'_0$ depend on the distance $r'$. The probability distribution of distances $r'$ for the system as a function of the paleodose $D_n = \dot{D}t$ is found from the equation:

$$P_n(r') = \frac{P(r')n(r')}{N(r')} = P(r')\frac{D'_0}{D_0}\left(1 - e^{-\frac{D_n}{D'_0}}\right) \tag{12}$$

where $P(r') = 3(r')^2 e^{-(r')^3}$ is the unfaded probability distribution function (PDF) of charge carriers (Eq. (15) in Li and Li, [17]).

Fig. 4a shows an example of the probability distribution $P_n(r')$ from Eq. (12), with the numerical values $\rho' = 3\times10^{-6}$, $s = 3\times10^{15}\ s^{-1}$, $D_n = 500$ Gy and $D_0 = 538$ Gy. The solid curve in Fig. 4a represents the symmetric unfaded PDF $P(r') = 3(r')^2 e^{-(r')^3}$, and the dashed curve represents the faded distribution $P_n(r')$. Physically this $P_n(r')$ function represents a "tunneling front" which is mathematically similar to the front shown in Fig 1a for ground state tunneling.

Li and Li [17] in their Fig. 3 showed that the effective dose constant $D'_0$ is a very sharp rising function of the distance $r'$. The function $P_n(r')$ shown in Fig 4a is the product of two functions, of the symmetric function $P(r') = 3(r')^2 e^{-(r')^3}$ and of this sharply rising function $D'_0/D_0$. These two functions are shown in Fig. 4b, and are similar to the two functions shown in Fig. 2b.

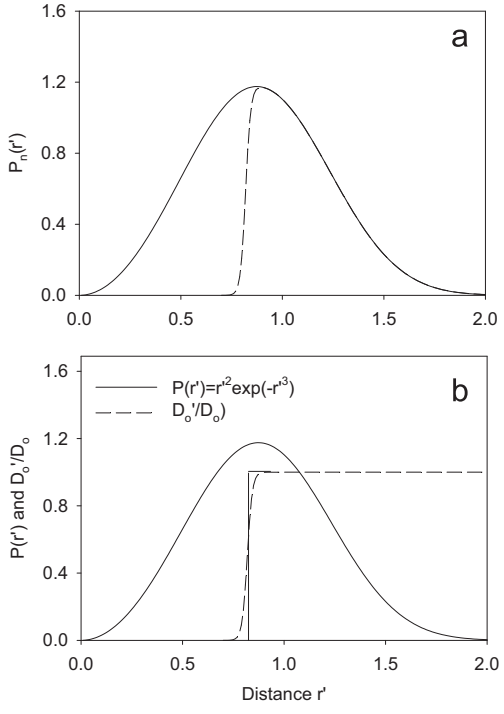


**Fig. 4.** (a) An example of the probability distribution $P_n(r')$ from Eq. (12), with the numerical values $\rho' = 3\times10^{-6}$, $s = 3\times10^{15}\ s^{-1}$, $D_n = 500$ Gy and $D_0 = 538$ Gy. The solid line represents the symmetric unfaded PDF $P(r')$, and the dashed line represents the faded distribution $P_n(r')$, which is mathematically similar to the tunneling front shown in Fig. 2a for ground state tunneling. (b) The function $P_n(r')$ shown in (a) is the product of the two functions $P(r')$ and $D'_0/D_0$ shown here. These two functions are similar to the two functions shown in Fig. 2b.

The mathematical similarity between the sharply rising functions in Figs. 2b and 4b leads us to consider a critical radius *rc* for the model in Fig. 4b. This critical radius will not represent the fading front of Fig. 2b, but rather is a mathematical analog between the two models, which allows us to obtain an analytical expression for the DRCs in tis section.

By using $P_n(r')$ from Eq. (12) one can find the faded luminescence signal by integrating over the distance $r'$

$$L_n(D_n) = \int_0^\infty MP_n(r')dr' = \int_0^\infty M\frac{P(r')D'_0}{D_0}\left(1 - e^{-\frac{D_n}{D'_0}}\right)dr' \tag{13}$$

where $M$=total number of traps such that $N(r') = MP(r')dr'$ (see Li and Li (2008), their Eq. (10)). The unfaded luminescence signal is found from:

$$L_{unfaded} = \int_0^\infty MP(r')\left(1 - e^{-\frac{D_n}{D_0}}\right)dr' \tag{14}$$

The goal of this subsection is to obtain analytical expressions for the two integral Eqs. (13) and (14). Just as in the approximate version of the model of Huntley [24] in Section 2.1, one introduces a critical radius $r'_c$ which can be used to approximate the mathematical behavior of the sharp rising function in Fig. 4b. The value of the critical distance can once more be estimated from the inflection point of the probability function $P_n(r')$ in Eq. (12). By taking the second derivative of Eq. (12):

$$\frac{d^2P_n(r')}{dr'^2} = -\frac{s\dot{D}D_0 e^{r'(\rho')^{-1/3}}\left(\dot{D}e^{r'(\rho')^{-1/3}} - D_0 s\right)}{\left(\dot{D}e^{r'(\rho')^{-1/3}} + D_0 s\right)^3(\rho')^{2/3}}. \tag{15}$$

Setting the term in parenthesis in the numerator equal to zero one obtains the critical radius

$$r'_c = (\rho')^{1/3}\ln\left(\frac{D_o s}{\dot{D}}\right). \tag{16}$$

This equation expresses the critical radius $r'_c$ as a function of the density $\rho'$, and of the additional parameters $D_0, s, \dot{D}$ in the model by Li and Li [17]. The values of $r'_c$ calculated from Eq. (16) agree with the numerical examples shown by Li and Li [17] in their Fig. 3. The result in Eq. (16) can also be obtained by considering the half-way point of the probability function $P_n(r')$ in Eq. (12).

Using this value of $r_c'$ from Eq. (16), one can now approximate the integral in Eq. (13), by replacing the sharp rising term $D'_0/D_0$ with a value of for $r' < r'_c$ and with a value of for $r' \geq r'_c$, and by carrying out the integration from $r' = r'_c$ to infinity, Eq. (13) becomes

$$\begin{aligned} L_n(D_n) &= \int_0^\infty M\frac{P(r')D'_0}{D_0}\left(1 - e^{-\frac{D_n}{D'_0}}\right)dr' \\ &\simeq \int_{r'_c}^\infty MP(r')\left(1 - e^{-\frac{D_n}{D_0}}\right)dr' \\ &\simeq M\left(1 - e^{-\frac{D_n}{D_0}}\right)\int_{r'_c}^\infty P(r')dr' \end{aligned} \tag{17}$$

Integrating by using Eq. (16) we get

$$L_n(D_n) = \left(1 - e^{-\frac{D_n}{D_0}}\right)Me^{-\rho'\ln\left[\frac{D_0 s}{\dot{D}}\right]^3} \tag{18}$$

This is the desired analytical expression for the luminescence $L_n$ as a function the natural dose $D_n$.

For the unfaded signal the corresponding expression becomes

$$L_{unfaded} = \int_0^{\infty} MP(r')\left(1 - e^{-\frac{D_n}{D_0}}\right)dr' = M\left(1 - e^{-\frac{D_n}{D_0}}\right) \tag{19}$$

Eqs. (18) and (19) are the derived analytical equations which can be compared with the exact numerical results from Eqs. (13) and (14). Mathematically the faded and unfaded signals $L_n$ and $L_{unfaded}$ in Eqs. (18) and (19) are both simple saturating exponentials of the natural dose $D_n$. As expected, the saturation level reached by the unfaded signal in Eq. (19) is equal to the total number of traps $M$. However, the saturation level reached by the faded signal in Eq. (18) is equal to $Me^{-\rho' \ln\left[\frac{D_0 s}{D}\right]^3}$. The ratio of the faded over the unfaded signal is then equal to $e^{-\rho' \ln\left[\frac{D_0 s}{D}\right]^3}$, and will depend on the parameters $\rho'$, $s$, $D_0$ and $D$ in the model. By using the values $\rho' = 3 \times 10^{-6}$, $s = 3 \times 10^{15}\ s^{-1}$, $D_0 = 538$ Gy and $D = 3$ Gy ka$^{-1}$ we obtain the fading ratio $e^{-\rho' \ln\left[\frac{D_0 s}{D}\right]^3} = 0.438 = 43.8\%$ of the original signal remaining in the sample.

Experimentally one usually measures a sensitivity corrected signal, by using the response of the material to a test dose. For a small test dose of $D_{test}$ the corresponding unfaded test dose signal $T_x$ from Eq. (19) is

$$T = M\left(1 - e^{-\frac{D_{test}}{D_0}}\right) \tag{20}$$

The sensitivity corrected signal $L/T$ is then found from the ratio of Eqs. (18) and (20), to yield the analytical expression:

$$\frac{L_n}{T_n} = \frac{\left(1 - e^{-\frac{D_n}{D_0}}\right)e^{-\rho' \ln\left[\frac{D_0 s}{D}\right]^3}}{\left(1 - e^{-\frac{D_{test}}{D_0}}\right)} \tag{21}$$

Fig. 5a and b compares the results from the DRC analytical Eqs. (18) and (21) with the corresponding results obtained by the numerical integration of Eq. (13). Very good agreement is seen at all values of the natural dose $D_n$.

### 2.3. Analytical expression for the g-value at the 50% reduction point of the luminescence signal

In this subsection an analytical expression is derived for the well-known $g$-value used to describe the tunneling rate in luminescence materials exhibiting anomalous fading (AF), under certain conditions. Fig. 6 shows a plot of Eq. (8), this plot is also shown in Fig. 1 in the model by Huntley [24]. Our purpose is to obtain the $g$-value as a function of the dimensionless density $\rho'$ by analyzing the properties of the function $n(t)$ in Fig. 6. In the traditional description of anomalous fading effects, the decay of luminescence signal after the end of irradiation is written as (Li and Li, [17]):

$$\frac{L}{L_c} = 1 - \frac{g}{100}\log_{10}\left(\frac{t}{t_c}\right) \tag{22}$$

where the $g$ factor describes the percentage loss of luminescence signal per decade of time, and $t_c$ is the time when the first measurement $L_c$ takes place. In practical situations one plots the remnant luminescence signal $L$ as a function of $\log_{10}(t)$, and the slope of the linear part of this graph is the $g$-factor. According to this definition, the $g$-value depends on the exact point in time ($t_c$) in which the first measurement is carried out, as well as on the choice of the linear part of the experimental data (See Li and Li [17], for a more detailed discussion).

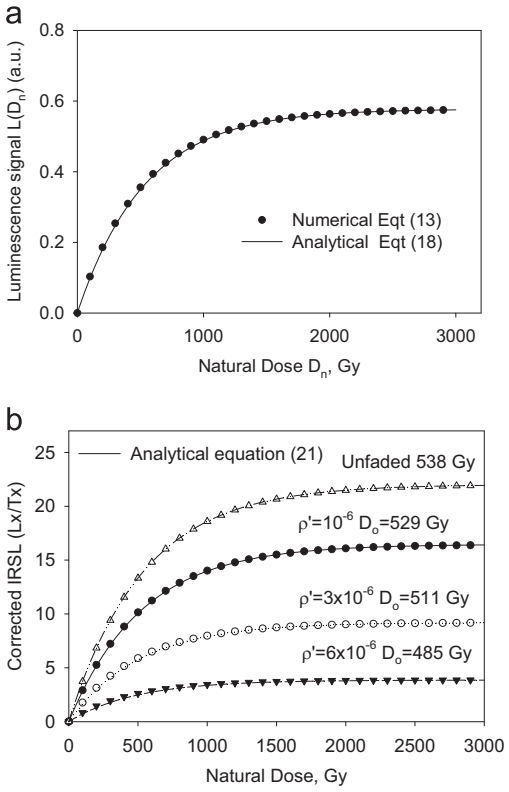


**Fig. 5.** Comparison of the DRC analytical Eq. (18) with the results obtained by the numerical integration of Eq. (13). (b) Comparison of the DRC analytical Eq. (21) with the results obtained by the numerical integration of Eq. (13).

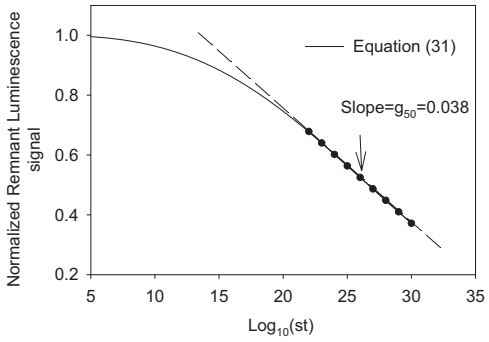


**Fig. 6.** Plot of Eq. (4) for typical feldspar values $\rho' = 3 \times 10^{-6}$, $s = 3 \times 10^{15}\ s^{-1}$ and for ground state tunneling. The slope of the linear part is $g_{50} = 0.038 = 3.8\%$ per decade.

In this subsection the analytical value of $g$ as a function of the charge density $\rho'$ is evaluated at the 50% reduction point of the luminescence signal, and this value is denoted in the rest of the paper by $g_{50}$. Fig. 6 is a plot of Eq. (4) for typical values of parameters $\rho' = 3 \times 10^{-6}$, $s = 3 \times 10^{15}\ s^{-1}$ for ground state tunneling.

In converting to a base-10 logarithm $\ln(st) = 2.3 \log_{10}(st)$, Eq. (4) can be written as:

$$n(t) = n_o e^{-\rho'\left[2.30 \log_{10}(st)\right]^3} = n_o e^{-12.167\rho'\left[\log_{10}(st)\right]^3}. \tag{23}$$

The tangent straight line in Fig. 6 shows that the linear part of the graph extends between $\log_{10}(st) = 22$ (which corresponds to a time $t \sim 1$ month after irradiation) and $\log_{10}(st) = 30$ (which corresponds to $t \sim 1$ million years after irradiation). The slope of the linear part of this graph is defined as the $g$-Value for the fading process, and is estimated graphically to be 0.0384 (i.e. $g = 3.84\%$ per decade).

For the rest of the calculation in this subsection we set

$$u = \log_{10}(st), \tag{24}$$

and Eq. (23) becomes

$$n_g(t)/n_o = e^{-12.167\rho' u^3}. \tag{25}$$

The middle point of the interval between $u = \log_{10}(t) = 22$ and $u = \log_{10}(t) = 30$ is the inflection point of the function in Eq. (25). This inflection point is almost identical (within 1%) to the point of 50% reduction of the luminescence signal, and can be found by setting the second derivative of $n(t)$ equal to zero:

$$\frac{d^2}{du^2}\left(e^{-12.167\rho' u^3}\right) = e^{-12.167\rho' u^3}u\rho'\left[-73.002 + 1332.32\rho' u^3\right] = 0 \tag{26}$$

By setting the term inside the bracket in Eq. (26) equal to zero and solving for $u$ we find after some simple algebra the inflection point:

$$u_{\text{inflection}} = \frac{0.3798}{(\rho')^{1/3}} \tag{27}$$

The $g_{50}$-value will be equal to the value of the first derivative $dn/dt$at the inflection point:

$$g_{50} = \left[\frac{dn}{dt}\right]_{u=u_{\text{inflection}}} = \left[-36.5e^{-12.167\rho' u^3}u^2\rho'\right]_{u=0.3798/(\rho')^{1/3}} \tag{28}$$

which simplifies to:

$$g_{50} = 2.7035(\rho')^{1/3} \tag{29}$$

This is the new analytical expression for the $g$-Value at the 50% reduction point of the luminescence signal, as a function of the concentration of charge carriers $\rho'$ in the material. Using a value of $\rho' = 3 \times 10^{-6}$ Eq. (29) gives $g = 0.039 = 3.9\%$ per decade, in agreement with Fig. 6.

Eq. (29) can be expected to be accurate at the inflection point or at the 50% reduction point of the function $n(t)$, and in the linear region around this point. In practical experimental situations successful use of Eq. (29) requires the following:

(1) A single tunneling component in the experimental data.
(2) Accurate experimental data near the 50% reduction point of the faded luminescence signal and
(3) A material exhibiting high AF rate ( > 15%), so that the AF effects can be measured within a reasonable experimental time frame.

In general, if the experimental data for $n(t)$ versus $\log_{10}(t)$ is not linear, one should fit $n(t)$ with the full Eq. (23). Furthermore, it is noted that this equation will not be useful for feldspars, which typically show AF rates of the order of 3–6% per decade.

The TL, OSL and TA-OSL signals monitored during anomalous fading experiments are the remnant TL/OSL, representing the signals remaining after various times have elapsed from the end of irradiation. These remnant signals are defined as the ratio $r$ of the luminescence signal remaining after storage time $t$, to the corresponding signal measured at a given time $t_0$ after the end of the irradiation.

Fig. 7a shows three examples of previously published experimental data for apatite samples which exhibit high anomalous fading. Fig. 7b shows the same data with the horizontal axis on a logarithmic scale $\log_{10}(t)$. The experimental data shown are for the remnant blue-OSL signals from Durango apatite (Polymeris et al. [34] their Figs. 3 and 7), and for a green apatite sample from Ontario, Canada (Polymeris et al. [35]). Additional samples for which the $g$-Values were previously published in Polymeris et al. [35] were analyzed in the same manner. The values of $g_{50}$ for these apatite samples were between 15% and 35%.

In all cases studied Eq. (25) provides an excellent fit to the experimental data, and the experimental $g_{50}$-Values were calculated directly from the linear part of the graphs in Fig. 7b. Once the data is fitted with Eq. (25), the fitted values of the charge densities $\rho'$ are used to calculate the $g_{50}$ values using Eq. (29). The numerical values for the calculated and the experimental values of $g_{50}$ are plotted against each other in Fig. 8. An alternative analysis using the linear region near the 50% point yielded essentially the same results, with the error bars being slightly smaller. Since the data in Fig. 8 lies reasonably close to the 1:1 line, it is concluded that there is in general good agreement between experiment and Eq. (25) for all studied samples.

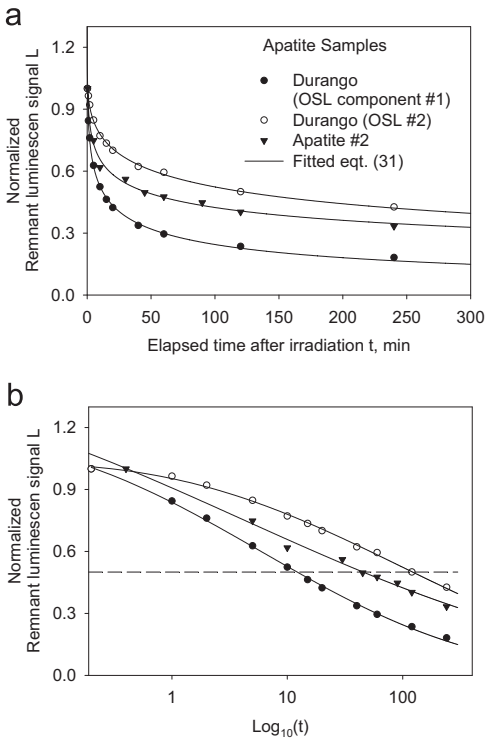


**Fig. 7.** (a)Three examples of previously published experimental data for apatite samples which exhibit high anomalous fading. In all cases studied equation (23) provides an excellent fit to the experimental data. (b) The same data as in (a), shown with the horizontal axis on a logarithmic scale $\log_{10}(t)$. The experimental $g_{50}$-Values are calculated from the fittings with equation (23), and from the linear part of such graphs.

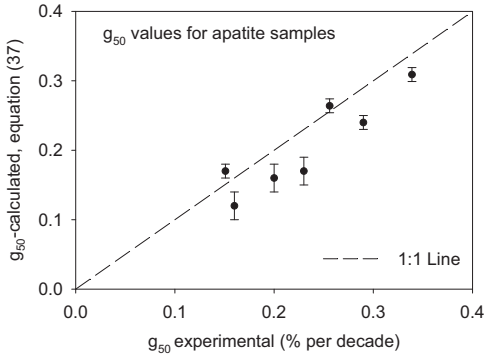


**Fig. 8.** The numerical values for the calculated and the experimental values of $g_{50}$ for several different apatites are plotted against each other. The values of $g_{50}$ for these apatite samples are between 15% and 35% and are reasonably close to the 1:1 line, showing general good agreement between experiment and equation (23) for all studied samples.

## 3. Discussion

It is also interesting to discuss the results of this paper within the context of the excited state tunneling model by Jain et al. [11,37]. These authors presented two versions of a new kinetic model which quantifies localized electronic recombination of donor–acceptor pairs in luminescent materials, with recombination taking place via the excited state of the system, and only for nearest-neighbors within a random distribution of centers. The semi-analytical version of the model by Jain et al. [11] was examined by Kitis and Pagonis [13], who obtained analytical solutions for several experimental modes of stimulation. In additional work Pagonis et al. [32] examined the exact version of the model by Jain et al. [11] and showed that these equations for excited state tunneling are a direct generalization of the equations previously derived by Huntley [24] for the case of ground state tunneling. In the approximate semi-analytical model of Jain et al. [11], the arbitrary time scaling factor $z=1.8$ previously introduced by Huntley (2006), was given a physical meaning as representing the time derivative of the critical lifetime $\tau_c$, i.e. $z \approx d\tau_c/dt$. The new analytical explanation for the numerical value $z=1.8$ was presented in Appendix A.

One must also keep in mind the limitations of the models presented in this paper. These models as well as the model by Jain et al. [11] have been shown to be useful for explaining anomalous fading phenomena. However, there are limitations of applying these models for tunneling taking place from the excited state. Specifically it has been suggested that band-tail states exist in feldspars and that they play an important role in the production of TL and IRSL from feldspar. The models described in this paper ignore the transition from the excited states into the band-tail states, from where the electrons can recombine with holes via thermal hopping or tunneling [5,36–39]. For example, Li and Li [37] provided an alternative model by considering the direct transition into the band-tail states, and found that their model describes the experimental data equally well. Incorporating the band-tail states into the current models is more complex and beyond the scope of this paper, but this is a limitation that needs to be addressed in future work.

## 4. Conclusions

In this paper an overview was presented of the three commonly used models in Fig. 1, and it was shown that the models are described by the same type of integral equation. In general this equation needs to be integrated numerically, but in many cases it was shown here that it is possible to obtain an accurate analytical approximation of the integrals involved.

Eqs. (18), (19) and (21) in this paper can be used directly as analytical tools to fit experimental data for dose response curves in materials showing anomalous fading over geological or laboratory time scales.

Eq. (29) can be used to obtain a quick estimate of the dimensionless concentration of charge carriers $\rho'$ in a material by using the experimentally determined $g$-Value at the 50% reduction point of the luminescence signal. Conversely, this equation provides an estimate of the expected $g_{50}$ value for the material, using the experimentally determined value of $\rho'$ from fitting CW-IRSL curves. However, one must be aware of the assumptions under which Eq. (29) was derived, namely that only a single tunneling component in present in the experimental data, and also that accurate experimental data are available near the 50% reduction point of the faded luminescence signal.

## Appendix A

Explanation of the numerical $z=1.8$ constant appearing in the model

*The empirical Eq. (8)*

By expanding the term $\ln (zst)^3$ appearing in the empirical Eq. (8) we obtain

$$\ln (1.8st)^3 = [\ln(1.8) + \ln(st)]^3 = [0.59 + \ln(st)]^3 = \ln(st)^3 + 1.763 \ln^2 + 1.03 \ln(st) + 0.20 \quad \text{(A1)}$$

For large values of $\ln(st)$ the last two terms in this expression are negligible and one can approximate:

$$\ln (1.8st)^3 = [\ln(1.8) + \ln(st)]^3 \simeq \ln(st)^3 + 1.763 \ln(st)^2 \quad \text{(A2)}$$

Therefore in the Huntley (2006) model the concentration $n(t)$ is calculated using the *empirical* relationship:

$$n(t) = n_o e^{-\rho' [\ln(zst)]^3} = n_o e^{-\rho' \left[ \ln(st)^3 + 1.763 \ln(st)^2 \right]}. \quad \text{(A3)}$$

*The analytical equation of Tachiya and Mozumder [28]*

Tachiya and Mozumder [28] showed that a more accurate expression of the concentration $n(t)$ is obtained from the numerical integration in Eq. (4)

$$n(t) = n_o e^{-\rho' [g(st)]} \quad \text{(A4)}$$

where the function $g(st)$ is given by the following series expression in Eq. (18) of the paper by Tachiya and Mozumder [28]

$$g(st) = \ln (st)^3 + 1.7316 \ln (st)^2 + 5.9343 \ln(st) + 5.4449 \quad \text{(A5)}$$

For large values of $\ln(st)$ one can again neglect the last two terms in expression (A5)

$$g(st) \simeq \ln (st)^3 + 1.7316 \ln (st)^2 \quad \text{(A6)}$$

Therefore in the detailed analysis carried out by Tachiya and Mozumder [28], the concentration $n(t)$ is calculated using the *analytical* relationship

$$n(t) = n_o e^{-\rho' [g(st)]} = n_o e^{-\rho' \left[ \ln (st)^3 + 1.7316 \ln (st)^2 \right]} \quad \text{(A7)}$$

Comparison of the right hand side of the empirical expression (A3) and the analytical expression (A7) shows that they are equal within an accuracy of 1% or better, and therefore the value of $z=1.8$ provides an accurate description of $n(t)$.

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