Analytical solutions for stimulated luminescence emission from tunneling recombination in random distributions of defects

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ABSTRACT

Recently a new kinetic model was presented in the literature, which describes localized electronic recombination in donor–acceptor pairs of luminescent materials. Within this model, recombination is assumed to take place via the excited state of the donor, and nearest-neighbor recombinations take place within a random distribution of centers. Two versions of the model were presented which were found to be in good agreement with each other, namely an exact model that evolves both in space and in time, and an approximate semi-analytical model evolving only in time. The model simulated successfully both thermally stimulated luminescence (TL) and optically stimulated luminescence (OSL), and also demonstrated the power law behavior for simulated OSL signals. This paper shows that the system of simultaneous differential equations in the semi-analytical model can be approximated to an excellent precision by a single differential equation. Furthermore, analytical solutions are obtained for this single differential equation, and for four different experimental modes of stimulation: TL, OSL, linearly modulated OSL (LM-OSL) and isothermal TL processes. The exact form of the power law for the model is found in analytical form for both OSL and isothermal TL processes. The analytical equations are tested by successfully fitting typical infrared stimulated luminescence (IRSL) signals, as well as experimental TL glow curves from feldspar samples. The dimensionless number density of acceptors in the model is estimated from fitting the experimental IRSL and TL data. The analytical expressions derived in this paper apply also to stimulated emission via the excited state of the donor–acceptor system. However, the same analytical expression, with different numerical values for its constants, can also be applied in the case of ground state tunneling, with important implications for luminescence dating.

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1. Introduction

Localized transition recombination models (LTMs) have been used for almost half a century since the pioneer work of Halperin and Braner [1], to describe a variety of behaviors of luminescence signals and for a wide variety of materials. Many variations of these models have been developed, which are based on the assumption of a fixed recombination probability between the excited state of a trap and a recombination center [2–7]. For reviews of experimental and modeling studies involving these various versions of LTM models and their application for a variety of natural and synthetic materials, the reader is referred to the luminescence books by Bøtter-Jensen et al. [8], Chen and McKeever [9], and Chen and Pagonis [10].

In addition to these models which assume a fixed recombination probability, recent experimental and modeling studies support the notion that the tunneling/recombination probability in many materials may vary with time. Specific attention has been paid to "anomalous fading" of luminescence signals in feldspar samples, due to the importance of this phenomenon in dating studies ([11–14]; and references therein). Several of these studies have suggested that anomalous fading is due to quantum mechanical tunneling from the ground state of the trap [15–21]. Furthermore, it has been shown that this ground state tunneling process in various materials can be described by power-law decay [22–24]. The experimental and modeling work by Poolton et al. [15,16,25] and more recently by Jain and Ankjærgaard [26] and Ankjærgaard et al. [27], provided valuable information on the origin of infrared stimulated luminescence (IRSL) from feldspars, and supported the existence of tunneling processes involving localized recombinations with tunneling taking place from the excited state of the trap, as well as charge migration through the conduction band-tail states. In a recent paper Pagonis et al. [28]

presented a new empirical kinetic model based on localized electronic transitions between the ground and the excited state, in an attempt to describe such tunneling via the excited state in feldspars. Results from this model were compared successfully with experimental CW-IRSL data, and were consistent with the reported power law decay of luminescence from feldspars.

In a very recent important development in this research area, Jain et al. [29] presented a new general kinetic model which quantifies localized electronic recombination of donor–acceptor pairs in luminescent materials. Recombination is assumed to take place via the excited state of the donor, and to take place between nearest-neighbors within a random distribution of centers. Two versions of the model were presented, an exact model that evolves in both space and time, and an approximate semi-analytical model evolving only in time. These authors found good agreement between the two models, and simulated successfully both thermally stimulated luminescence (TL) and optically stimulated luminescence (OSL). The model also demonstrated the power law behavior for OSL signals simulated within the model.

The goals of the present paper are:

(a) To show that the system of simultaneous differential equations developed by Jain et al. [29] can be approximated to an excellent precision by a single differential equation describing stimulated luminescence emission in this physical system.
(b) To obtain analytical solutions of this single differential equation, for several possible modes of stimulation, namely TL, OSL, linearly modulated OSL (LM-OSL) and isothermal TL (ITL).
(c) To derive and study the exact analytical form for the power law behavior in this system.
(d) To demonstrate how typical infrared stimulated luminescence (IRSL) and TL signals can be analyzed using the derived analytical equations, and what physical information can be extracted from such experimental data.

The analytical expression derived in this paper applies to stimulated emission via the excited state of the donor–acceptor system. However, it can also be applied in the case of ground state tunneling, a result of interest in luminescence dating.

2. Analytical solutions of the model by Jain et al. [29]

In this section we show that the system of equations in the semi-analytical model can be replaced with a *single* differential equation, which provides an excellent approximation to the original set of equations. Furthermore, an analytical solution is obtained for this single differential equation. The equations in the model of Jain et al. [29] are:

$$\frac{dn_g}{dt} = -An_g + Bn_e, \tag{1}$$

$$\frac{dn_e}{dt} = An_g - Bn_e - \frac{3n_e\rho'^{1/3}}{\tau_c}\left(\ln\frac{n_o}{n}\right)^{2/3}z, \tag{2}$$

$$L(t) = -\frac{dm}{dt} = \frac{3n_e\rho'^{1/3}}{\tau_c}\left(\ln\frac{n_o}{n}\right)^{2/3}z, \tag{3}$$

$$\frac{dT}{dt} = \beta \quad \text{(for TL)}$$
$$\frac{dT}{dt} = 0 \quad \text{(for isothermal TL, CW-OSL and LM-OSL)}, \tag{4}$$

$$\tau_c = s^{-1}\exp\left[\left(\frac{1}{\rho'}\ln\frac{n_o}{n}\right)^{1/3}\right] \tag{5}$$

The following parameters and symbols are used in the model: n_g and n_e are the instantaneous concentrations of electrons in the ground state and in the excited state correspondingly. m is the instantaneous concentration of acceptors (holes), n is the instantaneous concentration of all the donors, and N represents the instantaneous concentration of electrons in thermally disconnected states, such that $m = n + N = (n_g + n_e) + N$. The parameter A represents the excitation rate from the ground to the excited state, and is equal to $A = s\exp(-E/kT)$ and $A = \sigma(\lambda)I$ for the cases of thermal and optical excitation correspondingly, and β is the linear heating rate. Here $E = $ thermal activation energy, s is the frequency factor, λ is the optical stimulation wavelength, $\sigma(\lambda)$ is the optical absorption cross-section and I is the light intensity $(cm^{-2}\,s^{-1})$. Additional parameters are the dimensionless number density of acceptors ρ', the critical tunneling lifetime τ_c and $z = 1.8$ a dimensionless constant introduced in the model. B is the relaxation rate from the excited into the ground state, and $L(t)$ is the instantaneous tunneling luminescence from recombination via the excited state. If the detailed balance principle is assumed to be valid, one also has $B = s$. However, this is not a necessary condition for obtaining the analytical solutions in this paper.

Jain et al. ([29], their figure 4b) showed that for typical numerical values of the parameters in the model, n_e is many orders of magnitude smaller than n_g. Furthermore, excited states relax quite rapidly compared to the time scales of TL and OSL experiments, and therefore one can model the excited state in the quasi-steady approximation. Specifically the time scale for electronic relaxation processes involving the excited states (term dn_e/dt in Eq. (2)) can be of the order of ns or μs, while the time scale for TL processes can be of the order of ms or s (luminescence term $3\rho'^{1/3}/\tau_c(\ln n_o/n)^{2/3}z$ in Eq. (2)). On the basis of this observation (which is verified by the numerical simulations in this paper), one can use the approximation

$$\frac{1}{n_e}\frac{dn_e}{dt} << \left[B + \frac{3\rho'^{1/3}}{\tau_c}\left(\ln\frac{n_o}{n}\right)^{2/3}z\right] \tag{6}$$

With typical values of B in the range of 10^6–$10^{12}\,s^{-1}$ or higher, this is likely a very accurate assumption. Applying the condition (6) to Eq. (2), it then follows that the term on the left hand side can be ignored, and this equation can be solved for n_e to yield:

$$n_e = \frac{An_g}{B + \left(3\rho'^{1/3}/\tau_c\right)\ln(n_o/n)^{2/3}z} \tag{7}$$

It is noted that a similar approximating method was used in a recent paper by Chen et al. [30], to obtain semi-analytical solutions for a system of differential equations in a two stage model of TL. Substituting Eq. (7) into (1) we obtain:

$$\frac{dn_g}{dt} = -An_g + B\frac{An_g}{B + \left(3\rho'^{1/3}/\tau_c\right)\ln(n_o/n)^{2/3}z} = \frac{-An_g\left(3\rho'^{1/3}/B\tau_c\right)\ln(n_o/n)^{2/3}z}{1 + \left(3\rho'^{1/3}/B\tau_c\right)\ln(n_o/n)^{2/3}z} \tag{8}$$

For typical values of the parameters, the numerical value of the quantity $3\rho'^{1/3}/B\tau_c(\ln n_o/n)^{2/3}z$ in the denominator of this equation is much smaller than 1. By inspection of Eq. (2), it is seen that this is equivalent to the retrapping term Bn_e being much smaller that the term $3n_e\rho'^{1/3}/\tau_c(\ln n_o/n)^{2/3}z$ representing the luminescence pathway. Hence we can approximate

$$\frac{dn_g}{dt} = -An_g\frac{3\rho'^{1/3}}{B\tau_c}\left(\ln\frac{n_o}{n}\right)^{2/3}z \tag{9}$$

It is noted that the derivative dn_g/dt in this equation also represents the luminescence intensity given in Eq. (3) as $L(t) = -dm/dt \approx -dn_g/dt$, since $m = (n_g + n_e) + N$ and $n_e << n_g$, as discussed above. Substituting Eq. (5) into (9) and using

$n = n_e + n_g \approx n_g$:

$$\frac{dn_g}{dt} = -\frac{As3\rho'^{1/3}}{B} \frac{n_g}{\exp\left[\left((1/\rho')\ln(n_o/n_g)^{1/3}\right)\ln(n_o/n_g)^{2/3}z\right]}$$
$$= -R\frac{n_g}{\exp\left[\left((1/\rho')\ln(n_o/n_g)^{1/3}\right)\ln(n_o/n_g)^{2/3}\right]}, \quad (10)$$

where the quantity R is defined as:

$$R = \frac{As3\rho'^{1/3}z}{B} = A3\rho'^{1/3}z, \quad (11)$$

using $B=s$ from the detailed balance principle. Eq. (10) is the desired single differential equation for $L(t) = -\frac{dn_g}{dt}$ which provides an approximation to the solution of the system of Eqs. (1)–(4). In this equation A can be a function of time, depending on the type of stimulation mode used during the experiment. Eq. (10) is solved analytically in the next subsection.

2.1. Analytical solution of Eq. (10)

Due to the complex nature of the algebra involved, the derivation of several algebraic expressions in the rest of this paper was checked using the symbolic manipulation capability of the program *Mathematica*.

Rearranging Eq. (10) one obtains:

$$\exp\left[\frac{\left((1/\rho')\ln(n_o/n_g)\right)^{1/3}}{\ln(n_o/n_g)^{2/3}}\right]\frac{dn_g}{n_g} = -Rdt \quad (12)$$

Integrating:

$$\int_{n_o}^{n_g}\left(\ln\frac{n_o}{n'_g}\right)^{-2/3}\exp\left[\left(\frac{1}{\rho'}\ln\frac{n_o}{n'_g}\right)^{1/3}\right]\frac{dn'_g}{n'_g} = -\int_0^t Rdt', \quad (13)$$

where n_g represents the concentration of electrons in the ground state at time t, and n'_g is a dummy integration variable. The integral can be evaluated using the substitution $y = \ln(n_o/n'_g)$:

$$\int_0^{\ln(n_o/n_g)}(y)^{-2/3}\exp\left[\left(\frac{1}{\rho'}y\right)^{1/3}\right]dy = -\int_0^t Rdt, \quad (14)$$

which by integration and substitution of the value of R from Eq. (11), yields:

$$\frac{-3+3e^{-((1/\rho')\ln(n_o/n_g)^{1/3}}}{(1/\rho')^{1/3}} = -\int_0^t Rdt' = -3\rho'^{1/3}z\int_0^t Adt' \quad (15)$$

The integral $\int_0^t Adt'$ on the right hand side of this equation can be evaluated for each mode of excitation used in typical luminescence experiments. Specifically we will consider TL, isothermal TL, OSL and linearly modulated OSL (LM-OSL).

By rearranging and simplifying Eq. (15):

$$e^{-\left((1/\rho')\ln(n_o/n_g)^{1/3}\right)} = 1+z\int_0^t Adt' \quad (16)$$

After taking logarithms and some simple algebra:

$$n_g(t) = n_oe^{-\rho'\left[\ln\left(1+z\int_0^t Adt'\right)\right]^3} = n_oe^{-\rho'[F(t)]^3} \quad (17)$$

with the quantity $F(t)$ defined by:

$$F(t) = \ln\left(1+z\int_0^t Adt'\right) \quad (18)$$

At this point one can proceed by differentiating Eq. (17), and evaluating the time derivative $F'(t)$ of the time dependent function $F(t)$ given in Eq. (18) from:

$$F'(t) = zA\left[1+z\int_0^t Adt'\right]^{-1} = zAe^{-F(t)} \quad (19)$$

By substituting (18) and (20) into (10),

$$L(t) = -\frac{dn_g}{dt} = 3n_o\rho'F(t)^2zAe^{-F(t)}e^{-\rho'[F(t)]^3} \quad (20)$$

As an alternative derivation of Eq. (20), one can substitute expression (18) in Eq. (10) and carry out some algebra, with the same result.

Eqs. (17), (18) and (20) are the desired general analytical equations, which express the concentration of electrons $n_g(t)$ in the ground state and the luminescence intensity $L(t)$ as a function of time t. These equations are valid for all possible excitation modes used in typical TL and OSL experiments. By evaluating $F(t)$ for the different excitation modes, one obtains the luminescence intensity $L(t)$ as an analytical function of time, as shown in the next subsections.

2.2. Isothermal TL experiments

In the case of isothermal TL experiments (ITL), the quantity $A = s\exp(-E/kT_o) = $ constant, where T_o is the temperature at which the isothermal experiment is carried out. From Eq. (18) one finds:

$$F_{ITL} = \ln(1+szt\exp(-E/kT_o)) \quad (21)$$

For typical values of the parameters, the quantity $szt\exp(-E/kT_o) >> 1$ and therefore

$$e^{-F(t)} \approx e^{-\ln(zs\exp(-E/kT_o)t)} = \frac{1}{zs\exp(-E/kT_o)t} \quad (22)$$

With this approximation Eq. (20) yields:

$$L_{ITL} = 3n_o\rho'F(t)^2\frac{1}{t}e^{-\rho'[F(t)]^3} \quad (23)$$

Eqs. (21) and (23) are the analytical equations for isothermal luminescence intensity L_{ITL} at temperature T_o. At large times t, the function $F(t)^2e^{-\rho'[F(t)]^3}$ in the numerator of Eq. (23) varies slowly with time, and therefore the isothermal intensity follows an approximate $1/t$ power law. This result is in general agreement with detailed isothermal experiments carried out in feldspars (Baril [24]), who showed that at large times the isothermal signals follow a $1/t^k$ power law, with the coefficient k very close to 1.

Fig. 1 shows a comparison of the analytical Eqs. (23) and (21) with the solution of the system of Eqs. (1)–(4), for the case of isothermal TL signals. Fig. 1(a) shows three simulated isothermal TL signals for temperatures $T=573$, 623 and 733 K, while Fig. 1(b) shows the corresponding concentration of electrons in the ground state $n_g(t)$. The solid lines in this figure represent the analytical solution from Eqs. (23) and (21), and excellent agreement is seen between the analytical solution and the solution of the system of differential equations. Fig. 1(c) shows the simulated data in Fig. 1(a) on a log–log scale; for large times t the data follows indeed the power law of luminescence with a power law coefficient $k=(0.930\pm0.001)$.

2.3. CW-OSL, IRSL and LM-OSL experiments

The case of CW-OSL or IRSL excitation mode is similar to the isothermal TL experiments, with $A=\lambda=$ constant representing the optical excitation probability. In this case:

$$F_{CW-OSL} = \ln(1+z\lambda t) \quad (24)$$

and the modified power law in Eq. (23) is valid also in this case, with $F(t)$ given by Eq. (24).

In the case of linearly modulated OSL (LM-OSL) excitation mode, the probability of optical excitation is varied linearly with time in the form $A=bt/T$, where $T=$ total excitation period and b is
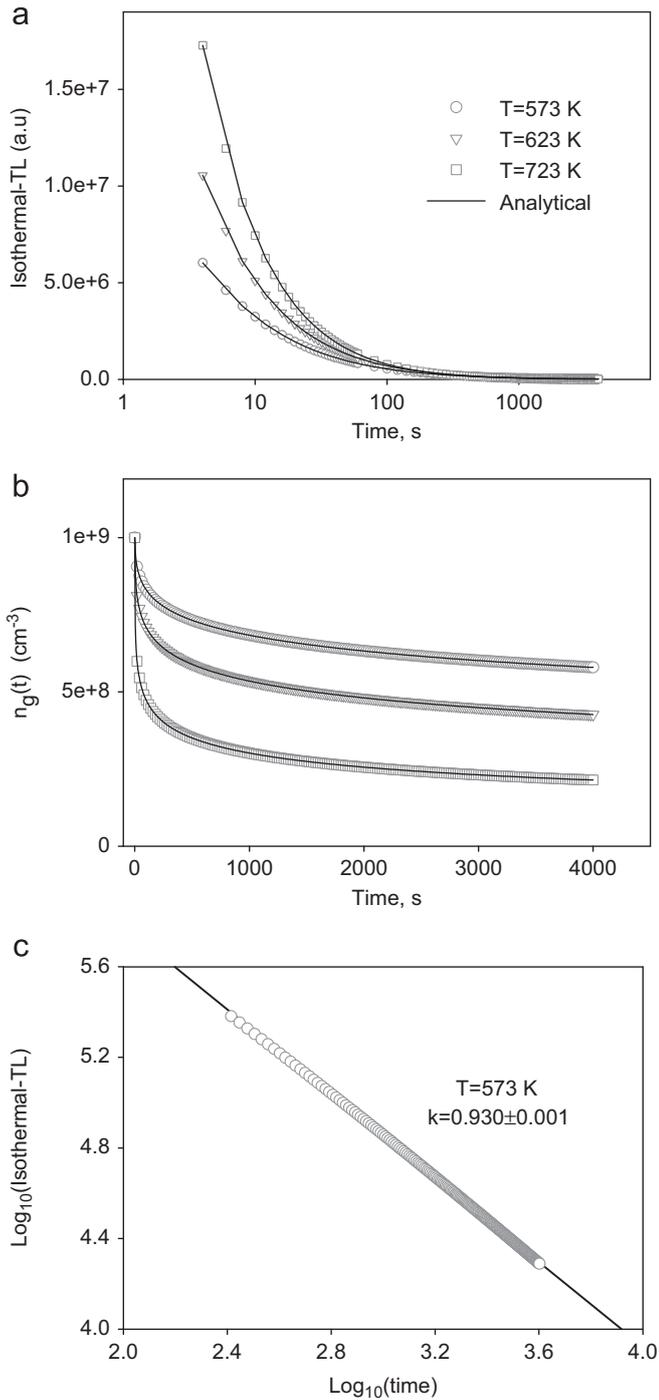
a

b

c

Fig. 1. Comparison of the analytical Eqs. (23) and (21) with the solution of the system of Eqs. (1)–(4), for the case of isothermal TL. (a) Three simulated isothermal TL signals are shown at temperatures $T=573$, 623 and 733 K, (b) the corresponding concentration of electrons in the ground state $n_g(t)$, and (c) the data in (a) is shown on a log–log scale for large times t, demonstrating the power law of luminescence.

a constant. In this case Eq. (18) becomes:

$$F_{LM-OSL} = \ln\left(1 + \frac{zbt^2}{2T}\right) \tag{25}$$

and Eq. (23) is valid, with $F(t)$ given by Eq. (25).

Fig. 2 shows a comparison of the analytical Eqs. (23) and (25) with the solution of the system of Eqs. (1)–(4), for the case of LM-OSL signals. Fig. 2(a) shows three simulated LM-OSL signals for different values of the dimensionless parameter ρ', while
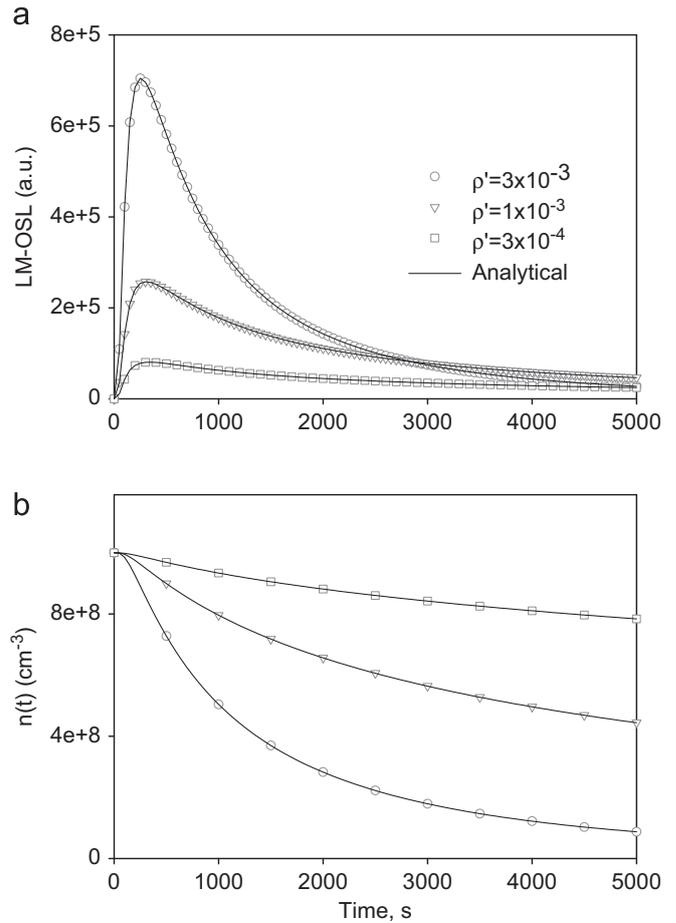
a

b

Fig. 2. Comparison of the analytical Eqs. (23) and (24) with the solution of the system of Eqs. (1)–(4), for the case of LM-OSL. (a) Three simulated LM-OSL signals are shown for different values of the dimensionless parameter ρ'. (b) The corresponding concentration of electrons in the ground state $n_g(t)$.

Fig. 2(b) shows the corresponding concentration of electrons in the ground state $n_g(t)$. The solid lines represent the analytical solution from Eqs. (23) and (25). Excellent agreement is seen between the analytical solution and the solution of the system of differential equations.

2.4. Thermal stimulation with a linear heating rate

In this case the integral appearing in Eq. (18) becomes:

$$\int_0^t A dt' = \int_0^t s \exp(-E/kT) dt' = \int_0^t s \exp\left[-\frac{E}{k(T_o + \beta t)}\right] dt' \tag{26}$$

This expression is the well-known exponential integral of TL theory, which can be approximated rather accurately by the first two terms of its series (see for example the book by Chen and Pagonis [10], chapter 15). If desired, one can of course add more series terms in the following expressions for increased accuracy.

$$\int_0^t A dt' = \frac{skT^2}{\beta E} e^{-\frac{E}{kT}}\left(1 - \frac{2kT}{E}\right) \tag{27}$$

The desired equations in this case are after some algebra:

$$n_g(t) = n_o e^{-\rho'[F(t)]^3} \tag{28}$$

$$L_{TL} = -\frac{dn_f}{dt} = -\frac{dn_f}{dT}\frac{dT}{dt} = -\frac{dn_f}{dT}\beta$$

$$= \frac{3n_o e^{-\rho'[F_{TL}(t)]^3} s\left(E^2 - 6k^2T^2\right) z\rho'[F_{TL}(t)]^2}{EksT^2z - 2k^2sT^3z + e^{E/kT}E\beta}\beta \qquad (29)$$

where

$$F_{TL}(t) = \ln\left(1 + \frac{zskT^2}{\beta E}e^{-E/kT}\left(1 - \frac{2kT}{E}\right)\right) \qquad (30)$$

The analytical expressions (28)–(30) give the instantaneous concentration n_g of electrons in the ground state and the corresponding TL intensity $L(t)$ at temperature T, during a TL experiment carried out with a constant heating rate β. Even though Eq. (29) may appear to be cumbersome, it can be easily programmed into a least squares fitting function in order to analyze experimental TL data. The fitting parameters would be the energy E, proportionality constant n_o, frequency s, and dimensionless concentration ρ'. It is important to note that Eq. (29) can be used as is to fit experimental data, while the original set of equations needs to be solved numerically before fitting of data is attempted. The authors were able to adapt these equations into least square fitting routines based on the Levenberg–Marquardt algorithm, using several commercially available software packages.

Fig. 3 shows a comparison of the analytical Eqs. (28)–(30) with the solution of the system of Eqs. (1)–(4), for the case of TL signals. Fig. 3(a) shows three simulated TL signals for different values of the dimensionless parameter ρ', while Fig. 3(b) shows the corresponding concentration of electrons in the ground state $n_g(t)$. The solid lines represent the analytical solution from Eqs.
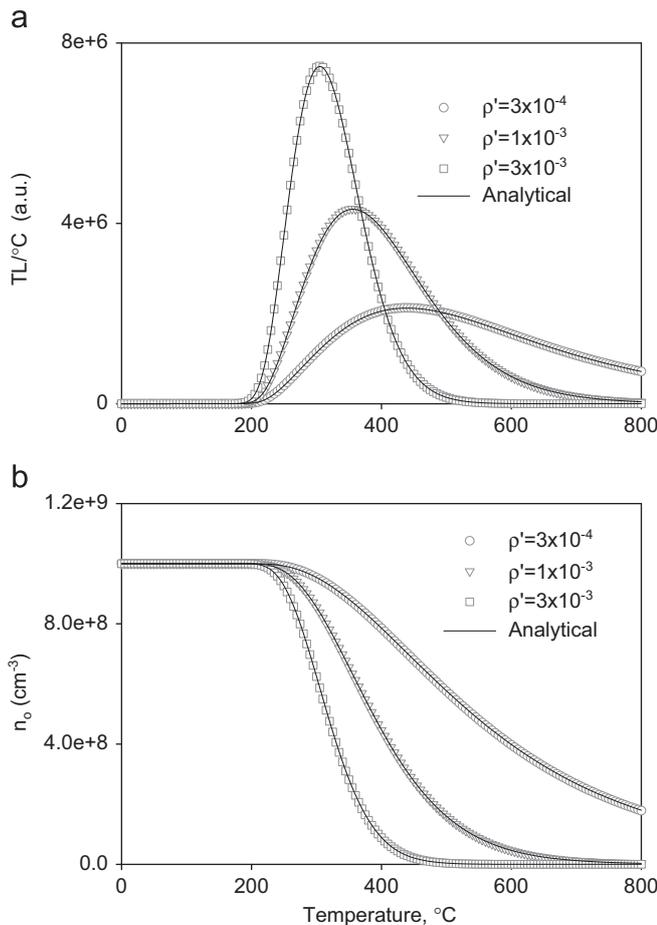
Fig. 3. Comparison of the analytical Eqs. (30) and (31) with the solution of the system of Eqs. (1)–(4), for the case of TL. (a) Three simulated TL signals are shown for different values of the dimensionless parameter ρ'. (b) The corresponding concentration of electrons in the ground state $n_g(t)$.

(28)–(30). Once more excellent agreement is seen between the analytical solution and the solution of the system of differential equations. The analytical equations derived in this paper can be easily adopted in a least squares fitting routine, which would allow the extraction of physical parameters from experimental TL, OSL, LM-OSL and isothermal TL data.

3. Comparison of analytical equations with experimental data

In this section we give two examples of fitting experimental IRSL and TL data for feldspars using the analytical equations in this paper.

3.1. Comparison with continuous-wave IRSL data in feldspars

The continuous-wave IRSL signals (CW-IRSL, also known as "infrared shine down" curves) from feldspars, are known to have a remarkably stable shape which changes little with the irradiation dose and preheat temperature, while it is sensitive to changes in the intensity of stimulating IR light [31,32]. These signals from feldspars are known to decay in a non-exponential manner and the exact mathematical shape of these curves is an open research question. Bailiff and Poolton [33] and Baril [24] showed that the IRSL decay follows a power law, while Poolton et al., [15,16,25] explained IRSL in feldspars using a donor–acceptor model, in which electron tunneling occurs from the excited state and the band tail states of the IRSL trap at about 1.4 eV. Thomsen et al. [31] suggested that in this model, the beginning of the IRSL decay curve originates with the luminescence emitted from close donor–acceptor pairs, while the end of the IRSL curve most likely represents the tunneling of distant pairs. There is an abundance of experimental data as well as significant modeling work in the literature which suggests that the dominant process in anomalous fading in feldspars is tunneling from the ground state or via the excited state, or from both.

To the best of our knowledge, there is no analytical equation to date in the literature which can describe the complete shape of the CW-IRSL curves. Eqs. (23) and (24) represent such analytical equations, suitable for analyzing the shape of CW-IRSL curves.

Fig. 4 shows experimental IRSL data for a feldspar sample studied by Thomsen et al. [32]. An aliquot of a coarse-grain sedimentary K-feldspar sample (laboratory code: 981014) was given a dose of 7 Gy, preheated at 280 °C for 60 s and stimulated with IR at 50 °C for 10,000 s using using full IR-stimulation power in their LED system ($\sim$135 mW/cm^2). The solid line represents the analytical solution from Eqs. (23) and (24), showing excellent agreement with the experimental data. Fig. 4(a) shows the experimental data and the fitted solid line for small times t up to 100 s, on a linear scale. Fig. 4(b) shows the full experimental IRSL data and the fitted solid line, for extended times up to 10,000 s and on a log–log scale, while Fig. 4(c) shows the same data on a log–linear scale. Fig. 4(d) shows that the IRSL data follows a $1/t^k$ power law with the power-law coefficient $k = 1.081 \pm 0.008$ very close to 1. The additional fitting parameter extracted from the fit shown in Fig. 4 is the dimensionless number density of acceptors $\rho' = 5 \times 10^{-3}$. This value is in a reasonable physical range expected for feldspars.

3.2. TL glow curves in feldspars

In this section a demonstration is given of fitting experimental TL data in a feldspar sample using the analytical expressions derived in this paper. This example is given only as a demonstration of possible analysis of the TL data, since a good fit to the experimental data does not necessarily mean that one knows the
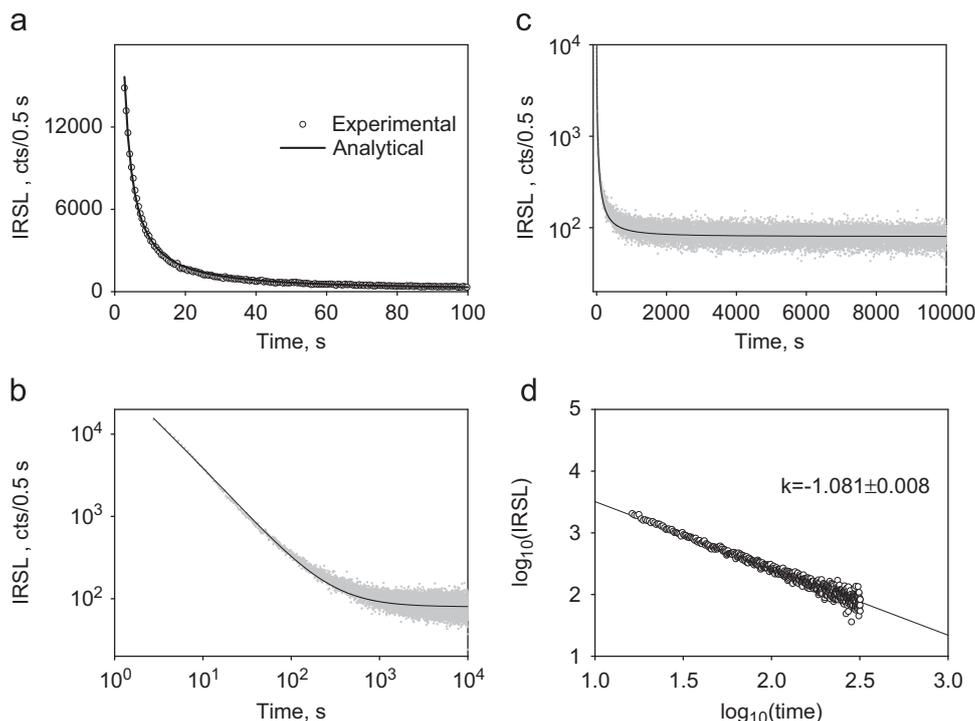
Fig. 4. Experimental IRSL data for a feldspar sample; the solid line represents the analytical solution from Eqs. (23) and (24). (a) The experimental data and fitted solid line are shown for IR excitation times up to 100 s on a linear scale for clarity. (b) The full scale of the experimental IRSL data and the fitted solid line are shown for extended times up to 5000 s on a log–log scale. (c) The same data as in (b) are shown on a log–linear scale. (d) The IRSL data follows a $1/t^k$ power law, with the power-law coefficient $k = 1.081 \pm 0.008$ very close to 1.
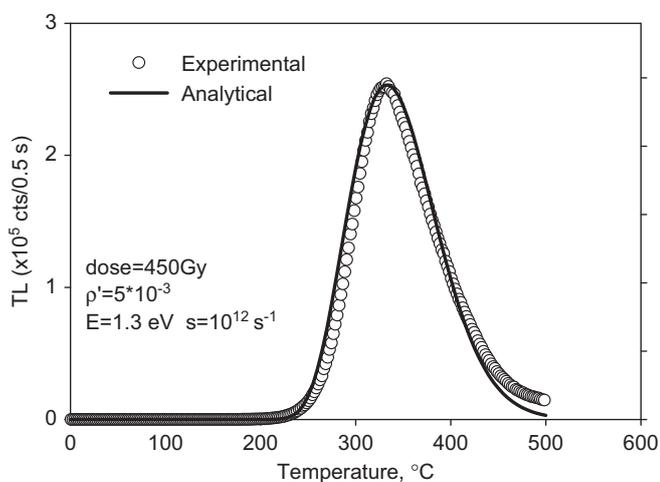

Fig. 5. Experimental TL data for a feldspar sample measured with a heating rate of 5 °C/s. The solid line represents the analytical solution from Eqs. (28)–(30). The sample was given a dose of 450 Gy and was subsequently preheated to 250 °C, in order to erase two lower temperature peaks in the TL glow curve. The shape of the TL peak remained the same at doses between 4.5 Gy and 900 Gy.

underlying luminescence mechanism [34]. Previous experimental and modeling work has shown that it is also possible to obtain good fits to such experimental data using linear combinations of TL peaks following second order kinetics [35].

Some studies have suggested that in K-feldspars the IRSL signal originates from a single trap corresponding to the thermo-luminescence peak at ~400 °C (as measured with a heating rate of 5 °C/s) [35,36]. Murray et al. [36] established that the low temperature TL peaks in their samples did not contribute to IRSL signal, despite their significant IR sensitivity. This finding was explained as due to changes in luminescence recombination probability during IR stimulation.

Fig. 5 shows experimental TL data for a feldspar sample. The luminescence signals of this K-rich sediment extract (laboratory code 951020), were recently studied by Jain and Ankjaergaard [26]. In the example shown in Fig. 5, the sample was given a dose of 450 Gy and was subsequently preheated to 250 °C before measurement of the TL glow curve, in order to erase two lower temperature peaks in the TL glow curve. The heating rate used was 5 °C/s. The shape of the TL glow curve was found experimentally to be independent of the dose, and only the peak height changed appreciably when the doses were varied in a wide range between 4.5 Gy and 900 Gy. The solid line represents the analytical solution from Eqs. (28) to (30) with the following best fit parameters: activation energy $E = 1.3$ eV, frequency factor $s = 10^{12}$ s^{-1}, and the dimensionless number density of acceptors $\rho' = 5 \times 10^{-3}$, the same value as the one obtained from fitting the CW-IRSL data shown in Fig. 4.

4. Discussion—implications for luminescence dating

Jain et al. [29] suggested that their model can also be used to describe ground state tunneling, according to the equation:

$$\frac{dn_g}{dt} = -n_g \frac{3\rho'^{1/3}}{\tau_c} \left(\ln \frac{n_o}{n_g} \right)^{2/3} \frac{d\tau_c}{dt} \qquad (31)$$

where n_g represents the concentration of electrons in the ground state, and the numerical values of the parameters ρ' and τ_c in this differential equation will be different for the ground state tunneling process. This equation is almost identical to Eq. (10) in this paper apart from a constant multiplication factor, and therefore the analytical solution derived in this paper for isothermal TL will still be valid for ground state tunneling. As discussed in Jain et al. [29], Eq. (31) is a mathematically robust way to describe tunneling from the ground state, and is an alternative form of the equation derived by Huntley [23]. It is concluded that the analytical equations derived here could in

principle be used for correcting for anomalous fading of lumines-cence signals due to tunneling.

Furthermore, the analytical expressions in this paper provide a means for testing the assumed tunneling processes; by extracting the physical parameters from the fitting process, one can in principle obtain valuable insight into the underlying processes and changes taking place in the material. The authors of this paper have also fitted successfully several experimental CW-IRSL data obtained for different samples and at different doses, as well as several Post-IR IRSL experimental curves. The latter curves are believed to represent more thermally stable luminescence signals and have been the subject of several recent studies (Jain and Ankjærgaard, [29]). The analytical expressions in this paper provide a possible means for quantifying these signals, and therefore could potentially lead to improvements in luminescence dating techniques using IRSL and/or Post-IRSL signals.

5. Conclusions

In this paper it was shown that the system of simultaneous differential equations in the semi-analytical model of Jain et al. [29] can be approximated to an excellent precision by a single differ-ential equation. Analytical solutions were obtained for this single differential equation, and for four different experimental modes of stimulation: TL, CW-OSL, LM-OSL and isothermal TL processes. The analytical expressions for OSL and isothermal TL were shown to be a modified power law, in agreement with experimental results. The analytical equations are tested by successfully fitting typical infra-red stimulated luminescence (CW-IRSL) signals, as well as experi-mental TL glow curves from feldspar samples. The dimensionless number density of acceptors in the model is estimated from fitting both experimental IRSL and TL data, and the extracted values were very similar. The analytical expressions provide a robust analytical tool which can help analyze experimental data from the four different modes of excitation, and hence provide valuable insights into the underlying luminescence processes.

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