Journal of Luminescence 137 (2013) 109–115

Contents lists available at SciVerse ScienceDirect

Journal of Luminescence

journal homepage: www.elsevier.com/locate/jlumin

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

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ARTICLE INFO

Article history:
Received 1 October 2012
Received in revised form
5 December 2012
Accepted 27 December 2012
Available online 4 January 2013

Keywords:
Stimulated luminescence emission
Tunneling
Thermoluminescence
Optically stimulated luminescence
Luminescence dating
Kinetic model

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.

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]

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http://dx.doi.org/10.1016/j.jlumin.2012.12.042

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 ($\mathrm{cm}^{-2}\,\mathrm{s}^{-1}$). 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–$10^{12}\ \mathrm{s}^{-1}$ 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