



## Localized transition models in luminescence: A reappraisal

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### ABSTRACT

Localized energy levels within the forbidden energy band are the source of various stimulated luminescence phenomena. The present study deals with the case of electrons stimulated from localized levels and recombining with a hole at a luminescence center, without the mediation of the conduction band. Previous research was based on three different assumptions as follows. Firstly, it was assumed that the recombination rate is independent of the concentration of recombination centers, and is proportional only to the concentration of electrons in the excited state of the trap. Secondly, it is assumed that the principle of detailed balance holds for these localized transitions. A third common assumption is that the system is in quasi-equilibrium condition. When these three conditions are applied to the system of differential equations describing the localized transitions, it was shown that the resultant thermoluminescence (TL) signals follow first order kinetics. This paper examines the assumptions used in these previous studies, and extensive simulations are carried out for a wide range of parameters in the localized transitions model. The results of the simulations show that the TL peaks in the localized model have very similar characteristics with TL peaks derived from delocalized models, including non-first order kinetic characteristics. The differential equations describing the localized transition model are solved analytically using the Lambert *W* function, and the resulting analytical master equation can describe a variety of optically and thermally stimulated phenomena.

### 1. Introduction

The models used to describe stimulated luminescence (SL) phenomena are phenomenological models based on the energy band theory of solids. Localized energy levels within the forbidden band are the source of several stimulated luminescence phenomena [1,2]. Three types of phenomenological models have been used extensively in the literature: delocalized models based on transitions involving the conduction and valence bands, localized models usually involving different energy levels of the traps/centers [1,3–5], and semilocalized models which are based on a combination of localized and delocalized energy levels [6,7]. For comprehensive historical summaries of these three types of luminescence models, the reader is referred to the relevant textbooks [1,2,8] and the recent review papers by Kumar et al. [9], Horowitz et al. [10], and Pagonis et al. [11].

There have been several types of localized models in the literature. In the original work by Halperin and Braner [3], during the thermal excitation stage electrons are raised into the excited state of the electron trap, from which they can either recombine at the recombination center, or they can be de-excited into the ground state. In this model, the conduction and valence bands participate in the kinetics only

during the irradiation stage, and direct transitions from the conduction band into the ground or excited state of the trap are not allowed. Most importantly for the purposes of this paper, the rate of recombination in this model was assumed to depend on *both* the concentrations of electrons in the excited state of the trap, and on the concentration of holes in the center. Chen and Kirsh [1] suggested that each electron can only recombine with its closest neighbor in the crystal, and modified the differential equation of Halperin and Braner [3], so that the rate of recombination is proportional *only* to the concentration of electrons in the excited state. This assumption when combined with the assumption of a quasi-equilibrium state, leads to an analytical first order kinetics equation. Bull [4] used the same assumptions as Chen and Kirsh [1] and the same system of differential equations, to simulate characteristic TL glow curves in this localized model. The model parameters used by Bull [4] resulted in TL peaks with peak maximum temperatures ranging from 350 K to 2000 K. Bull [4] also noted some deviations from pure first order kinetics in their results, and reported symmetry factors as high as 0.48 for some parameters of their model.

A different type of localized tunneling recombination model has been proposed by Jain et al. [12], in which the distances between electron and hole traps are described by a specific spatial distribution.

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This model was solved analytically by Kitis and Pagonis [13], who obtained a master equation describing stimulated luminescence phenomena based on quantum tunneling. Using the resulting analytical equations these authors were able to analyze a great number of experimental data [14–17].

During the past decade there has been extensive experimental and simulation work on luminescence from nanodosimetric materials. Recent Monte Carlo simulations have shown that in materials with very high density of defects, the nearest neighbor approximation is not valid, and therefore each electron may interact with several of its neighbors (Pagonis and Truong [18]). In such cases of localized high densities of electrons and holes, one might expect that the original model by Halperin and Braner [3] would be a better approximation than the later model by Chen and Kirsh [1].

All previous modeling research on localized transitions has been based on the following three different assumptions:

- The recombination rate is independent of the concentration of recombination centers, and is proportional to the concentration for electrons in the excited state of the trap.
- The principle of detailed balance is assumed to hold for these localized transitions.
- The system is in quasi-equilibrium condition.

When these three conditions are applied to the system of differential equations describing the localized transitions, it was shown that the resultant thermoluminescence (TL) signals follow exclusively first order kinetics [8].

The aims of the present work are:

- To examine critically the assumptions used in these previous studies.
- To simulate the localized transition model using the original version of the model by Halperin and Braner [3], without the simplifications leading to first order kinetics.
- To examine if existing analytical expressions previously developed for delocalized models, can also describe luminescence signals simulated within localized transition models.
- To evaluate in detail the peak shape characteristics of the TL peaks, for a wide range of values of the parameters in the model.
- To derive new analytical expressions by solving the system of differential equations in the localized transition model, by following the method used by Kitis and Vlachos [26] for delocalized models, and by Kitis and Pagonis [13] for localized models.
- To investigate the role of the principle of detailed balance in the localized transition models.
- To investigate whether the localized transition model can describe additional stimulated luminescence phenomena, such as Linearly modulated optically stimulated luminescence (LM-OSL), continuous wave OSL (CW-OSL), isothermal TL (ITL).

## 2. The model

Lawless et al. [19] demonstrated that in order for a simulation to describe properly experimental results, it must contain all three stages of irradiation, relaxation and heating. The irradiation stage must always be followed by a relaxation stage, because carriers can build up to significant levels in the conduction and valence bands at the end of the irradiation stage. The relaxation stage provides the necessary time for the concentrations of charge carriers to decay to zero before the heating stage. Sadek and Kitis [20] showed that inclusion of the three stages is also a critical requirement for simulations. These authors examined critically the TL results of Kelly et al. [21] and Opanowitz [22], and found that most of the parameters used by these authors are not compatible with the irradiation stage, and therefore their pessimistic point view on the analysis of TL glow curves is not warranted.

It must be noticed, however, that one could in principle carry out

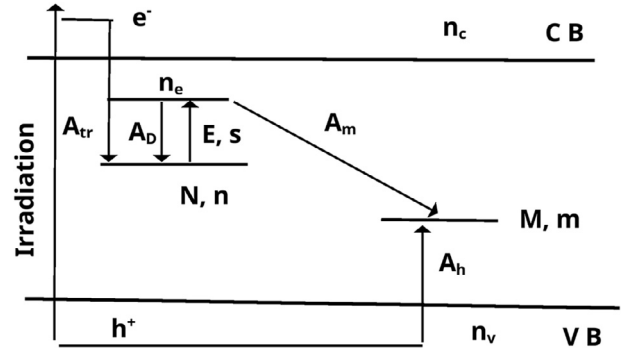


Fig. 1. Energy band diagram for the localized transition model.

the simulations in this paper by assuming equal initial concentrations of electrons and holes at the start of the heating stage; nevertheless, we have preferred to include all three stages for completeness.

In the present work we examine critically the original model of Halperin and Braner [3], by including the differential equations for the irradiation, relaxation and heating stages. More importantly, our study uses the original model without the later modifications by Chen and Kirsh [1] and Bull [4], which lead to first order kinetics.

The energy band diagram for the localized transition model is shown in Fig. 1. During irradiation electrons and holes are created in the conduction and valence bands, and are subsequently trapped into the ground state of the trap and the recombination center correspondingly. These transitions are indicated by  $A_{tr}$  and  $A_h$  in Fig. 1. During the heating stage electrons are raised into the excited state of electron trap, from which they can either recombine at the recombination center (transition  $A_m$ ), or they can be de-excited into the ground state (transition  $A_D$ ). As mentioned above, in this model the conduction and valence bands participate in the kinetics only during the irradiation stage.

The differential equations governing the traffic of free carriers during both the irradiation and heating stage are:

$$\frac{dT}{dt} = \beta, \quad (1)$$

$$\frac{dn}{dt} = -np(t) + A_D n_e + A_{tr}(N-n)n_c, \quad (2)$$

$$\frac{dm}{dt} = A_h(M-m)n_v - A_m m n_e, \quad (3)$$

$$\frac{dn_e}{dt} = np(t) - A_D n_e - A_m m n_e, \quad (4)$$

$$\frac{dn_v}{dt} = X - A_h(M-m)n_v, \quad (5)$$

$$\frac{dn_c}{dt} = X - A_{tr}(N-n)n_c. \quad (6)$$

The symbols on the diagram are:  $N$  ( $\text{cm}^{-3}$ ) is the concentration of available electron traps,  $n$  ( $\text{cm}^{-3}$ ) the concentration of trapped electrons,  $M$  ( $\text{cm}^{-3}$ ) is the concentration of available luminescence centers,  $m$  ( $\text{cm}^{-3}$ ) concentration of trapped holes.  $n_c$  ( $\text{cm}^{-3}$ ) and  $n_v$  ( $\text{cm}^{-3}$ ) are the concentration of electrons in the conduction and holes in the valence band.  $A_{tr}$  ( $\text{cm}^3 \text{s}^{-1}$ ) is the trapping coefficient in electron traps and  $A_h$  ( $\text{cm}^3 \text{s}^{-1}$ ) is the trapping coefficient of holes in luminescence centers.  $E$  (eV) is the thermal activation energy,  $s$  ( $\text{s}^{-1}$ ) is the frequency factor and  $k$  the Boltzmann constant,  $\beta$  ( $\text{K/s}$ ) is the heating rate and  $X$  ( $\text{cm}^{-3} \text{s}^{-1}$ ) is the rate of production of ion pairs per second and per  $\text{cm}^{-3}$ . This quantity is proportional to the dose rate. If the time of excitation is  $t_e$ , the total concentration of produced electron-hole pairs is  $X \cdot t_e$  (*i. p.*  $\text{cm}^{-3}$ ), a quantity which is proportional to the total dose applied.

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