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How to detect trap cluster systems?

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Abstract

Spatially correlated traps and recombination centres (trap-recombination centre pairs and larger clusters) are responsible for many anomalous phenomena that are difficult to explain in the framework of both classical models, i.e. model of localized transitions (LT) and the simple trap model (STM), even with a number of discrete energy levels. However, these 'anomalous' effects may provide a good platform for identifying trap cluster systems. This paper considers selected cluster-type effects, mainly relating to an anomalous dependence of TL on absorbed dose in the system of isolated clusters (ICs). Some consequences for interacting cluster (IAC) systems, involving both localized and delocalized transitions occurring simultaneously, are also discussed.
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1. Introduction

Classical models for trapping and recombination of charge carriers in dielectrics relate to two analytical cases. The first one relates to uniform distribution of traps and recombination centres (RCs), where all transitions of charge carriers from traps to RCs go through delocalized transport bands. This is the simple trap model (STM—cf. Chen and McKeever, 1997). The second one relates to pairs of traps and RCs (T–RC) located close to each other. The model of localized transitions (LT), for the first time formulated by Halperin and Braner (1960) and then modified by Land (1969), assumes that recombination of trapped carriers proceeds through a local excited level. This system of T–RC pairs may be considered as a system of smallest clusters. Results of many experiments and theoretical considerations gave evidence that thermoluminescence (TL) kinetic processes proceed in large-scale defects rather than in point-like traps (e.g. the paper of Townsend and Rowlands, 1999). None of the standard TL kinetic models is able to describe this type of kinetics properly. All the types of non-standard kinetics we will term the spatially correlated systems (SCSs).

The studies of TL kinetics in various SCSs were started some years ago by Mandowski and Świątek (1992). For this purpose they used several Monte Carlo algorithms modelling TL, e.g. in 1-D and 3-D systems under different external conditions (e.g. Mandowski and Świątek, 1997, 1998). It was found that TL in SCSs shows many unexpected features that cannot be explained within the framework of LT and STM models. Examples include apparently composite structure of monoenergetic peaks, additional 'displacement' peaks and the dependence of TL on the external electric field. For review, see Mandowski (1998, 2001, 2006a). Recently, an analytical model was proposed for the isolated clusters (ICs) model, which is a special case of SCSs (Mandowski, 2002, 2006b). The theory is based on two trap structural functions (TSFs)—Γ_n and Γ_h, for electrons and holes, respectively. Extensive numerical calculations gave the evidence that these functions depend only on structural properties of the solid. TSFs do not depend e.g. on the heating rate and the activation energy of traps.

In this paper, the properties of IC systems are studied with respect to dose dependence. Using Monte Carlo algorithms TSFs were calculated for various initial fillings of traps. It will be shown that TSFs depend on dose in a very specific way, quite different from that found in both classical models—STM and LT. In this way it is possible to distinguish between classical and cluster-type kinetics of charge carrier recombination.

Other properties relate to SCSs with both localized and delocalized transitions occurring simultaneously. The simplest case of this kind, relating to T–RC pairs, was recently formulated analytically as the model of semi-localized transitions (SLT)

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(Mandowski, 2005, 2007). The model is able to explain many unexpected features observed in typical solid state detectors, e.g. extremely high-frequency factors (Mandowski, 2006a). It seems that these and some other features relate also to much complex systems consisting of large interacting clusters. Qualitative discussion of the dose dependence in interactive cluster (IAC) systems will be provided.

2. TL kinetics of isolated clusters

Commonly accepted explanation of long-lasting phosphorescence and TL phenomena is based on the assumption of metastable levels (traps and RCs) situated within the energy gap. Although direct transition from trap to a recombination centre is possible, most of the transitions take place through excited states. In the model of ICs all trapping and recombination processes occur only within the clusters, i.e. no interaction between clusters is possible. TL is produced in the same way as in the LT model—a trapped charge carrier is thermally excited to a local excited level and then it may be retrapped or may recombine with an opposite charge carrier trapped at the RC level. For the simplest case of a single type of active traps, deep traps and RCs the following set of kinetic equations was proposed (Mandowski, 2002):

$$-\dot{n} = n\nu \exp\left(\frac{-E}{kT(t)}\right) - \Gamma_n(n)n_e, \tag{1a}$$

$$-\dot{h} = \Gamma_h(h)n_e, \tag{1b}$$

$$h = n + n_e + M, \tag{1c}$$

where E stands for the activation energy and ν is the frequency factor for active traps. n, n_e and h denote the total concentrations of electrons trapped in active traps, electrons in the excited levels and holes trapped in RCs. M stands for the concentration of electrons in the thermally disconnected traps (deep traps), i.e. traps that are not emptied during the experiment. $\Gamma_n(n)$ and $\Gamma_h(h)$ denote two TSFs for trapping and recombination, respectively. It is interesting that the set of Eqs. (1) is valid also for both classical cases: LT and STM. For these two limiting cases one has to assume appropriate form of TSFs

$$\Gamma_n^{(\mathrm{LT})} = \bar{A}, \tag{2a}$$

$$\Gamma_h^{(\mathrm{LT})} = \bar{B} \tag{2b}$$

and

$$\Gamma_n^{(\mathrm{STM})} = A(N - n), \tag{3a}$$

$$\Gamma_h^{(\mathrm{STM})} = Bh, \tag{3b}$$

where A, B, $\bar{A}$ and $\bar{B}$ are constants and N is the total concentration of traps (in the case of STM, n_e has the meaning of the concentration of carriers in the conduction band).

3. Dose dependence of TSFs

To calculate TSFs for an arbitrary IC system one has to perform Monte Carlo calculations. The algorithm is performed by considering elementary transitions—$\Im_D$, $\Im_T$ and $\Im_R$ for detrapping, trapping and recombination, respectively:

$$\Im_D(t) = \nu \exp\left[\frac{-E}{kT(t)}\right], \tag{4a}$$

$$\Im_T(t) = \bar{A}[\bar{N} - \bar{n}(t)], \tag{4b}$$

$$\Im_R(t) = \bar{B}\bar{h}(t). \tag{4c}$$

Here, the dashed values denote variables and parameters relating to a single cluster of the system. $\bar{N}$ denotes the number of trap levels, $\bar{n}_e$ is the number of electrons in the local excited level, $\bar{n}$ is the number of electrons in traps and $\bar{h}$ denotes the number of holes in RCs. $\bar{A}$ and $\bar{B}$ denote coefficients for trapping and recombination, respectively. A detailed method of the simulation as well as the scaling properties defining the relationship between the microscopic and the macroscopic parameters was given in some previous papers (e.g. Mandowski and Świątek, 1992, 1997).

For a given IC system TSFs can be calculated by performing a two-stage Monte Carlo simulation. In the first stage the clusters are randomly populated with electrons and holes. It is assumed that each hole–electron pair is trapped within the same cluster. Then, the TL process is simulated according to Eq. (4). Finally, Γ_n and Γ_h functions are calculated from Eqs. (1a) and (1b), respectively, i.e.

$$\Gamma_h(h) = \frac{-\dot{h}}{n_e}, \tag{5a}$$

$$\Gamma_n(n) = \frac{1}{n_{\mathrm{e}}}\left[n\nu \exp\left(\frac{-E}{kT(t)}\right) + \dot{n}\right]. \tag{5b}$$

Now, it is interesting to study what is the dependence of these functions on dose. First of all let us look at both standard models where TSFs are given by analytical equations (2a), (2b) and (3a), (3b). These functions are plotted for various initial concentrations of trapped charge carriers $\eta_0 \equiv n_0/N = 0.1$, 0.5 and 1.0 in Fig. 1. The common feature of the two models is that TSFs for partially filled systems ($\eta_0 = 0.1$ and $\eta_0 = 0.5$) reproduce perfectly a part of the whole TSFs calculated for full initial filling ($\eta_0 = 1$). Such behaviour seems to be quite obvious. Surprisingly, for all other cases—including clusters of various sizes, the dependence is quite different. As an example, we calculated TL kinetics in two cluster systems consisting of clusters containing $\bar{N} = 2$ traps and RCs as well as $\bar{N} = 5$ traps and RCs. These systems were initially populated to the relative concentration of η_0=0.1, 0.5 and 1.0. Results of this simulations are shown in Fig. 2A with respect to Γ_h functions. It is clearly seen that the calculated functions do not overlap for various initial populations. Another interesting puzzle is that the shape of these curves is somewhat similar (within preciseness of the Monte Carlo calculations) and applying simple scaling on the horizontal axis, according to the equations

$$\Gamma_n\left(\frac{n}{N}\right) \to \Gamma_n\left(\frac{n}{N}\chi\right), \tag{6a}$$

$$\Gamma_h\left(\frac{h}{h_0}\right) \to \Gamma_h\left(\frac{h}{h_0}\chi\right), \tag{6b}$$

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