



On the verification of the simple trap model by simultaneous TL/TSC measurements

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Abstract

Simultaneous detection of thermoluminescence (TL) and thermally stimulated conductivity (TSC) allows acquiring a lot of information concerning basic trapping processes in dielectrics. The values of activation energies and other trap parameters determined this way noticeably depend on the kinetic model applied. This paper shows that assuming some general conditions the TL/TSC ratio should yield a monotone decreasing function. This simple property holds for the standard model consisting of an arbitrary number of trap levels and recombination centres including also continuous distribution of traps. Counter examples considering simultaneous release of electrons and holes and temperature-dependent recombination probability are given. The later case, in some situations, could be ruled out by plotting a special test function. Thus, the TL/TSC ratio provides a test for a broad class of one-carrier trap models that are used for theoretical description of trapping and recombination phenomena.

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1. Introduction

Thermally stimulated relaxation phenomena are very important tools in characterising high-resistivity materials. Using thermoluminescence (TL) and

thermally stimulated conductivity (TSC) one gets information concerning the trap structure of dielectric materials. TL is the most important technique for modern dosimetry. It is extensively used also for archaeological/geological dating (for review see Ref. [1]). However, with all the wide range of applications, the physical basis involved in the kinetic mechanisms of the processes is still not well known.

TL and TSC could be observed when a sample under study is excited at appropriately low

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temperature T_0 by a high-energy radiation (e.g. UV or X-rays). Then, the excitation is removed and the sample is heated, usually with a constant rate β , i.e. $T(t) = T_0 + \beta t$. During heating, luminescence and/or conductivity of the sample is measured. A lot of effort has been put into extracting trap parameters from TL and TSC curves, for detailed review see Ref. [1]. Simultaneous TL/TSC measurement gives new possibilities in determining trap parameters based solely on the simple trap model (STM) [2–5] and its quasi-equilibrium approximation [5–8]. Many approximations of the STM are utilized for the analysis of TL in various applications and basic research. However, some old [3] and newer analyses [5] based on TL/TSC measurements, demonstrate inappropriateness of the model for proper description of trapping and recombination kinetics. Until now, no clear explanation of the discrepancy has been offered. Recently, considering the STM it was found that TL/TSC ratio should be an always-decreasing function. This property served for the determination of the range of validity of one trap STM and subsequent estimation of trap level activation energies in α -alumina (Al_2O_3) [5]. Nevertheless, it could be done merely for a very narrow range of temperatures. Experimentally determined TL/TSC ratios usually have a complex, non-monotone behaviour. In this paper, we examine a broad range of the basic kinetic models used for theoretical description of trapping and recombination kinetics. These include general multi-level case of STM and continuous distribution of activation energies of traps and recombination centres. For these models TL/TSC ratio provides a test which allows their falsification in some cases.

2. Theory

2.1. One trap model

Usually, one assumes that within a given temperature range only one trap level and one type of recombination centres are ‘active’. For the sake of simplicity, we will assume that active charge carriers are electrons. The situation is

schematically depicted in Fig. 1 with allowed transitions denoted by solid lines. In this case, termed the simple trap model (STM) charge carriers kinetics during thermal stimulation is governed by the following set of equations:

$$-\dot{n} = nv \exp\left(\frac{-E}{kT}\right) - n_c A(N - n), \quad (1a)$$

$$-\dot{h} = Bhn_c, \quad (1b)$$

$$h = n + n_c + M, \quad (1c)$$

where E stands for the activation energy and N , n and h denote the concentrations of trap states, electrons trapped in ‘active’ traps and holes trapped in recombination centres (RC), respectively. M stands for the number of electrons in the thermally disconnected traps (deep traps), i.e., traps that are not emptied during the experiment. A and B stand for the trapping and recombination probabilities, respectively, v the frequency factor and k the Boltzmann constant. The set of Eq. (1) has no analytical solutions in general case. However, some exact analytical relations were found when considering simultaneous TL/TSC measurement [2–4]. Now, let us define the ratio

$$\bar{R}(t, T(t)) = \frac{\mathcal{L}}{\mathcal{I}}, \quad (2)$$

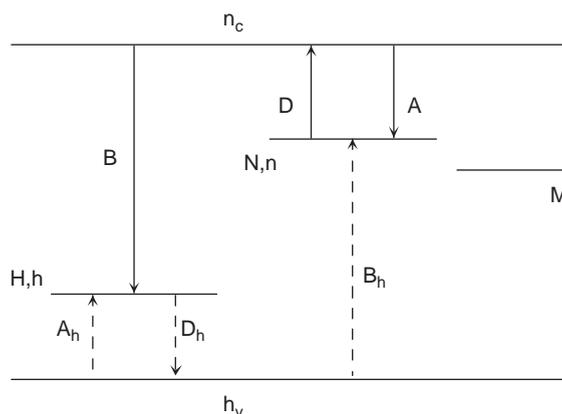


Fig. 1. Energy diagram for the simple trap model (STM—only solid lines) and simultaneous release of electrons and holes (solid and dashed lines). Solid lines denote electron transitions (A, D and B). Dashed lines denote hole transitions (A_h , D_h and B_h). Meaning of the symbols: A, A_h —trapping, D, D_h —detrapping, B, B_h —recombination.

where \mathcal{L} and \mathcal{J} are experimental TL and TSC intensities, respectively. We will assume that these intensities could be corrected due to temperature variation of charge carriers' mobility and the radiative efficiency. Therefore, these can be written as

$$\mathcal{L} = -\gamma_L \dot{h} = \gamma_L B h n_c, \quad (3)$$

$$\mathcal{J} = \gamma_c n_c, \quad (4)$$

where γ_L and γ_c are temperature-dependent functions: $\gamma_L = \gamma_L(T)$ and $\gamma_c = \gamma_c(T)$. Using Eq. (1b) we get

$$\begin{aligned} \bar{R}(t, T) &= \frac{\gamma_L(T)}{\gamma_c(T)} B h(t) \\ &= \gamma(T) B h_0 \exp \left[-B \int_0^t n_c(t') dt' \right], \end{aligned} \quad (5)$$

where $\gamma = \gamma_L/\gamma_c$ and h_0 is the initial concentration of holes in RCs. $\bar{R}(t, T)$ is an uncorrected TL/TSC ratio. The corrected one is defined as $R(t) = \bar{R}(t, T)/\gamma(T)$. For a given temperature dependence $T(t)$ we will simplify the notation as $\bar{R}(t, T(t)) \equiv \bar{R}(t)$ and $\bar{R}(t, T(t)) \equiv R(t)$. Therefore

$$R(t) = B h(t) = B h_0 \exp \left[-B \int_0^t n_c(t') dt' \right]. \quad (6)$$

Experimentally, it is possible to determine $R(t)$ with an unknown proportionality factor. It was shown earlier by Gasiot and Fillard [3] that $\dot{R}(t) \propto -J_{TL}$. J_{TL} and J_{TSC} are normalized TL and TSC intensities, respectively. From Eq. (6) it results also that $R(t)$ must be a monotone decreasing function. This is a weaker condition. Recently, this property was utilized for the determination of the range of validity of one trap STM in α -alumina (Al_2O_3) [5]. An example of $R(t)$ function calculated for numerically generated TL and TSC curves is shown in Fig. 2. $R(t)$ exhibits two plateau regions at the beginning and at the end of TL/TSC peaks with constant values R_0 and R_{end} , respectively. Using Eq. (1c) one gets (see appendix)

$$\frac{R_0}{R_{\text{end}}} = 1 + \frac{n_0}{M}. \quad (7)$$

This ratio can be determined without any proportionality factor; therefore, it allows estimation of the relative density of deep traps M/n_0 . The

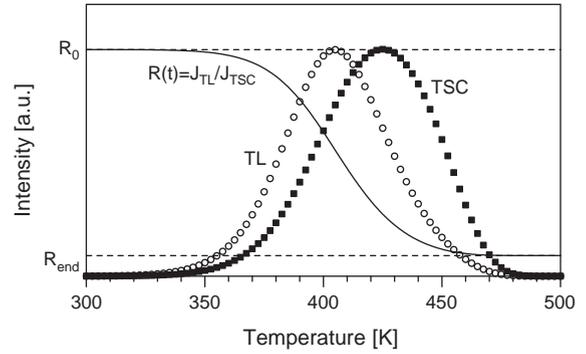


Fig. 2. Normalized TL, TSC curves and the ratio $R(t) = J_{TL}/J_{TSC}$ calculated in the framework of the simple trap model defined by Eqs. (1). The vertical axis has a linear scale. The spectra were calculated using the following parameters: $E = 0.9$ eV, $\nu = 10^{10}$ s $^{-1}$, $A/B = 1$, $M/N = 0.1$, $n_0/N = 1$, and the heating rate $\beta = 1$ K/s.

possibility of determining M/n_0 from TL/TSC data was widely discussed in some recent papers [4–8]. The uncorrected TL/TSC ratio $\bar{R}(t, T)$ is much easier to be measured than $R(t)$. In most cases, for testing the validity of the STM, it is sufficient to measure only $\bar{R}(t, T)$. Typically, if $R(t)$ decreases $\bar{R}(t)$ will be decreasing too. This is because thermal quenching of luminescence increases with temperature and the mobility of carriers increases also. As a result, the $\gamma(T)$ function is decreasing. Another way of verifying STM is to measure $\bar{R}(t)$ at a constant temperature by recording simultaneously current and luminescence decay.

2.2. Multiple traps and recombination centres

2.2.1. Spectrally resolved measurements

STM can be easily extended for an arbitrary number of discrete trap levels and RCs. Again we will assume that only one type of charge carriers is ‘active’. Considering p active trap levels and q recombination centres, the set of kinetic equations has the form

$$\begin{aligned} -\dot{n}_i &= n_i v_i \exp \left(\frac{-E_i}{kT} \right) - n_c A (N_i - n_i), \\ i &= 1, \dots, p, \end{aligned} \quad (8a)$$

$$-\dot{h}_j = B_j h_j n_c, \quad j = 1, \dots, q, \quad (8b)$$

$$\sum_{j=1}^q h_j = \sum_{i=1}^p n_i + n_c + M. \quad (8c)$$

The symbols above have the same meaning as in Eq. (1). Additionally, the subscripts i and j number trap levels and RCs, respectively. Each RC contributes to the total TL at specific range of wavelengths. Using the apparatus for spectrally resolved measurements (e.g. CCD camera with a spectrograph [9,10]) one is able to determine various emission bands relating to different types of RCs. Therefore, for each RC we define

$$\mathcal{L}_j = -\gamma_{L_j} \dot{h}_j, \quad (9)$$

where γ_{L_j} is a temperature-dependent function: $\gamma_{L_j} = \gamma_{L_j}(T)$. Experimentally, \mathcal{L}_j could be determined by integrating luminescence intensity over the whole emission band. TSC intensity is still defined by the same Eq. (4). Consequently, we define q ratios $\bar{R}_j(t, T)$:

$$\begin{aligned} \bar{R}_j(t, T(t)) &= \frac{\mathcal{L}_j}{\mathcal{I}} = \frac{\gamma_{L_j}(T)}{\gamma_c(T)} B_j h_j(t) \\ &= \gamma_j(T) B_j h_{0j} \exp \left[-B_j \int_0^t n_c(t') dt' \right], \end{aligned} \quad (10)$$

where $\gamma_j = \gamma_{L_j}/\gamma_c$. Respective corrected values $R_j(t) = \bar{R}_j(t, T)/\gamma_j(T)$ are given by

$$R_j(t) = B_j h_j(t) = B_j h_{0j} \exp \left[-B_j \int_0^t n_c(t') dt' \right]. \quad (11)$$

Again, we get monotone decreasing functions. Considering that for typical materials $\gamma_j(T)$ are also decreasing, both functions $\bar{R}_j(t, T(t))$ and $R_j(t)$ have to be decreasing with respect to time (or temperature). An example of this kind is presented in Fig. 3. Moreover, there is a strict dependence between various functions $R_j(t)$. Calculating the logarithm on both sides of Eq. (11) we can see that the logarithms $\ln[R_j(t)]$ are linearly dependent. This is an additional check for the model. Details of an algorithm based on this property will be given in a separate paper.

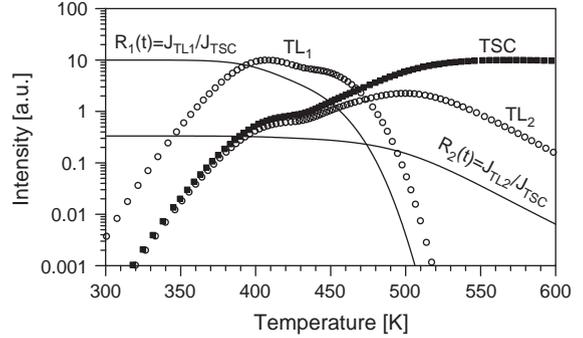


Fig. 3. TL/TSC calculated for two traps and two recombination centres. Trap and RC parameters: $E_1 = 0.9$ eV, $E_2 = 1.0$ eV, $v_1 = v_2 = 10^{10}$ s $^{-1}$, $A_1/B_1 = 1$, $A_2/B_2 = 10$, $A_1 = A_2$, $n_{01}/N_1 = n_{02}/N_2 = 1$, $N_1 = N_2$, $h_{01}/h_{02} = 3$. Heating rate $\beta = 1$ K/s.

2.2.2. Spectrally integrated and band segment measurements

Applying conventional measurement techniques (e.g. photomultiplier) one integrates luminescence intensity over a certain region of wavelengths. Thus, the measured value \mathcal{L}_{tot} is a weighted sum of \mathcal{L}_j , i.e. $\mathcal{L}_{\text{tot}} = \sum_{j=1}^q w_j \mathcal{L}_j$, where w_j are usually unknown constants ($w_j \geq 0$) dependent on detector sensitivity curve. \mathcal{L}_j are given by Eq. (9). Straightforward calculations yield for the TL/TSC ratio \bar{R}_f

$$\begin{aligned} \bar{R}_f(t, T(t)) &= \frac{\sum_{i=1}^q w_j \mathcal{L}_j}{\mathcal{I}} \\ &= \sum_{j=1}^q w_j \gamma_j(T) B_j h_{0j} \exp \left[-B_j \int_0^t n_c(t') dt' \right]. \end{aligned} \quad (12)$$

As a linear combination of monotone decreasing functions $\bar{R}_f(t, T(t))$ is also decreasing. There is no possibility to define here a corrected value $R_f(t)$.

A frequent modification of this type of measurements is an application of narrow band pass filters in front of the photomultiplier to separate various emission bands. In view of the fact that typical emission bands are very broad (usually more than 100 nm—e.g. Ref. [10]) and overlapping, such a

procedure does not guarantee that one records really a single emission band. For that reason Eq. (12) is still valid in this case, considering that coefficients w_j characterize also transmission properties of the band pass filter.

2.3. Continuous distribution of traps

In amorphous solids, activation energies of traps and RC's have continuous distribution. Considering that traps occupy energies between E_{r1} and E_{r2} , and RC's occupy energies between E_{r1} and E_{r2} , we can write the following set of kinetic equations [11–14]:

$$-\frac{\partial n(E, t)}{\partial t} = n(E, t)D(E, t) - n_c(t)A(E) \times [N(E) - n(E, t)], \quad (13a)$$

$$-\frac{\partial h(E, t)}{\partial t} = B(E)h(E, t)n_c(t), \quad (13b)$$

$$\int_{E_{r1}}^{E_{r2}} h(E, t) dE = \int_{E_{r1}}^{E_{r2}} n(E, t) dE + n_c(t) + M, \quad (13c)$$

where

$$D(E, t) = v(E) \exp\left[\frac{-E}{kT(t)}\right]. \quad (14)$$

In the above equations it was still assumed that one-carrier approximation holds, i.e. charge carriers of one type (here electrons) are active within considered temperature range. TSC intensity could be still expressed by Eq. (4). A contribution to luminescence intensity $\mathcal{L}(E, t)$ coming from recombination of free electrons with holes at the level E is proportional to $[-\partial h(E, t)/\partial t]$. Solving Eq. (13b) it is easy to show that

$$\mathcal{L}(E, t) \propto B(E)h_0(E)n_c(t) \exp\left[-B(E) \int_0^t n_c(t') dt'\right]. \quad (15)$$

However, due to spectral broadening this 'partial' luminescence could not be directly measured even in spectrally resolved TL experiments. Let us assume that $W(E, \lambda)$ describes the shape of $\mathcal{L}(E, t)$ normalized to unit intensity. Hence, the intensity recorded at the wavelength λ is

given by

$$\begin{aligned} \mathcal{L}(t, \lambda) &= \gamma_L(T, \lambda) \int_{E_{r1}}^{E_{r2}} L(E, t)W(E, \lambda) dE \\ &= \gamma_L(T, \lambda)n_c(t) \int_{E_{r1}}^{E_{r2}} B(E)h_0(E)W(E, \lambda) \\ &\quad \times \exp\left[-B(E) \int_0^t n_c(t') dt'\right] dE. \end{aligned} \quad (16)$$

Similarly as in previous cases γ_L represent a function dependent on spectral sensitivity of the detection system and luminous efficiency of the sample. Defining respective uncorrected $\bar{R}(t, T, \lambda)$ and corrected $R(t, T, \lambda)$ TL/TSC ratios we get

$$\begin{aligned} \bar{R}(t, T(t), \lambda) &= \frac{\mathcal{L}(t, \lambda)}{\mathcal{J}} \\ &= \gamma_L(T, \lambda) \int_{E_{r1}}^{E_{r2}} B(E)h_0(E)W(E, \lambda) \\ &\quad \times \exp\left[-B(E) \int_0^t n_c(t') dt'\right] dE, \end{aligned} \quad (17)$$

$$\begin{aligned} R(t, T(t), \lambda) &= \frac{\bar{R}(t, T(t), \lambda)}{\gamma_L(T, \lambda)} \\ &= \int_{E_{r1}}^{E_{r2}} B(E)h_0(E)W(E, \lambda) \\ &\quad \times \exp\left[-B(E) \int_0^t n_c(t') dt'\right] dE. \end{aligned} \quad (18)$$

All the functions under the integrals are positive and the exponential function decreases with time. For some reasons discussed earlier γ_L is also decreasing. So, obviously, \bar{R} and R have to be decreasing with respect to time.

2.4. Counter example I—temperature-dependent trapping and recombination probabilities

In all cases considered above the $R(t)$ function was proved to be monotone decreasing. However, one can expect a different behaviour when the trapping and recombination probabilities A and B are temperature dependent. This case was considered by Chen and Fleming [15] who showed that under specific conditions TL and TSC peaks occur in reversed order, i.e. TL peak occurs at higher temperatures than TSC peak. In this situation $R(t)$ obviously is no longer decreasing.

For the STM model the TL/TSC is expressed by

$$R(t) = B(t)h_0 \exp \left[- \int_0^t B(t')n_c(t') dt' \right]. \quad (19)$$

The sign of the derivative $\dot{R}(t)$ depends on $(\dot{B}(t) - B^2(t)n_c(t))$. The expression is particularly clear when applied to both sides of the TSC peak when $n_c \approx 0$. Here, $\dot{R}(t) \propto \dot{B}(t)$. Even slight increase in $B(t)$ results in increase in $R(t)$. An example of this kind is presented in Fig. 4. It was assumed that $A = A_0T^{0.3}$ and $B = B_0T^{0.3}$, where A_0 and B_0 are constants. Here TL and TSC peaks still occur in normal order; however, the ‘monotone decreasing’ property of $R(t)$ is violated. When the temperature dependence of $B(t)$ is known from other measurements, it is possible to correct $R(t)$ for the contribution defining $\dot{R}_T(t) \equiv R(t)/B(t)$ that restores again the decreasing feature. The same way of reasoning could be applied to the models of multiple traps and recombination centres and the continuous distribution of traps rearranging Eqs. (11), (12) and (18) in an analogous manner.

Following the analysis done by Chen and Fleming [15] $B(t)$ has the form $B(t) = B_0T^a(t)$, where $-\frac{7}{2} \leq a \leq \frac{1}{2}$. Thus, if one deals with unknown

$B(t)$ dependence and $R(t)$ reveals an increasing or peak-shape behaviour, we suggest to plot

$$R_T(t) = R(t)/\sqrt{T(t)} \quad (20)$$

to check whether temperature dependence of recombination probability could be responsible for that unusual behaviour. An example of this kind is presented in Fig. 4 by dashed line. Another possibility is to perform isothermal decay measurements. Eq. (6) holds for an arbitrary heating function $T(t)$. For $T = const.$ the $R(t)$ function is decreasing. The same applies to Eqs. (11), (12) and (18).

2.5. Counter example II—simultaneous release of electrons and holes

Now let us consider the case of simultaneous release of electrons and holes. For this purpose, we will use the simplest model of one active electron trap level and one active hole trap level shown in Fig. 1 with allowed transitions drawn by solid and dashed lines. The kinetic equations are the following [16]:

$$\dot{n} = An_c(N - n) - nD(t) - B_hh_vn, \quad (21a)$$

$$\dot{n}_c = -An_c(N - n) + nD(t) - Bn_ch, \quad (21b)$$

$$\dot{h} = A_hh_v(H - h) - hD_h(t) - Bn_ch, \quad (21c)$$

$$\dot{h}_v = -A_hh_v(H - h) + hD_h(t) - B_hh_vn. \quad (21d)$$

Here h_v denotes the concentration of free holes in the valence band, H denotes the concentration of hole traps, and A_h and B_h stand for the probability of trapping and recombination of free holes, respectively. $D_h(t)$ describes the probability of thermal detrapping of holes which is given by

$$D_h(t) = v_h \exp \left[\frac{-E_h}{kT(t)} \right], \quad (22)$$

where E_h and v_h denote the activation energy and the frequency factor for this transition. Other symbols were already defined. Here TSC intensity is defined by

$$\mathcal{I} = \gamma_cn_c + \gamma_hh_v, \quad (22)$$

where γ_h is a temperature-dependent function: $\gamma_h = \gamma_h(T)$. Luminescence may be due to B and B_h

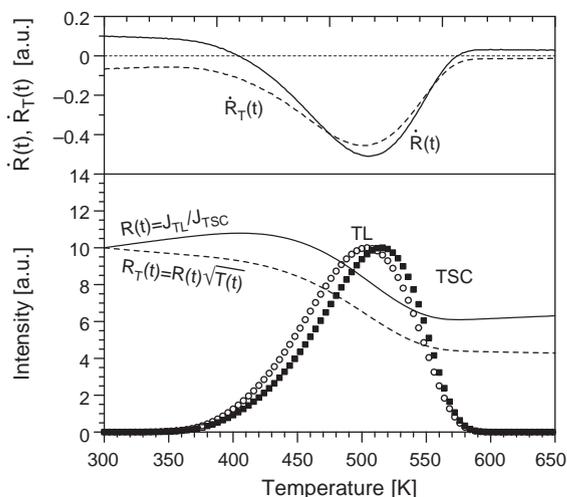


Fig. 4. TL/TSC ratio $R(t)$ calculated for the STM model with temperature-dependent trapping and recombination probabilities $A = A_0T^{0.3}$ and $B = B_0T^{0.3}$, where $A_0/B_0 = 10^2$. Other parameters: $E = 1.0\text{eV}$, $\nu = 10^{10}\text{s}^{-1}$, $M/N = 1$, $n_0/N = 1$ and the heating rate $\beta = 1\text{K/s}$. The testing function $R_T(t) = R(t)/\sqrt{T(t)}$ is also plotted. The upper diagram shows time derivatives of $R(t)$ and $R_T(t)$.

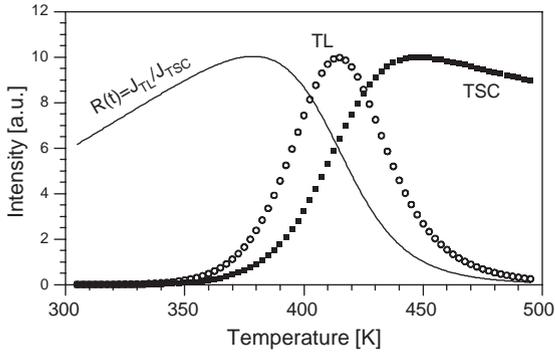


Fig. 5. Simultaneous release of electrons and holes—one electron trap and one hole trap shown in Fig. 1. Kinetic parameters: $E = 1.0\text{ eV}$, $E_h = 0.9\text{ eV}$, $\nu = \nu_h = 10^{10}\text{ s}^{-1}$, $A/B = A_h/B_h = 0.1$, $A = A_h$, $n_0/N = h_0/H = 0.1$, $N = H$ and $M = 0$. Heating rate $\beta = 1\text{ K/s}$.

transitions (Fig. 1). For simplicity we will assume that B_h transition is radiationless. Therefore

$$\mathcal{L} = \gamma_L B h n_c. \quad (23)$$

In Fig. 5 TL/TSC ratio is calculated for the following parameters: $E = 1.0\text{ eV}$, $E_h = 0.9\text{ eV}$, $\nu = \nu_h = 10^{10}\text{ s}^{-1}$, $A/B = A_h/B_h = 0.1$, $A = A_h$, $n_0/N = h_0/H = 0.1$, $N = H$, $M = 0$ and the heating rate $\beta = 1\text{ K/s}$. Here $R(t)$ has peak-shaped behaviour. Although TL and TSC occur in normal order, the ‘monotone condition’ of $R(t)$ is violated.

3. Conclusions

Numerous cases considered in this paper indicate that TL/TSC ratio $R(t)$ must be decreasing (or at least constant) function with respect to time (or temperature). This point was directly proved for a variety of models including STM, multiple traps and recombination centres and continuous distribution of traps. In all cases, it was assumed that the kinetics is governed by only one type of active charge carriers (i.e. electrons or holes). It was the only one significant assumption that was made during the calculations. Many experimentally determined TL/TSC data reveal a complex peak-shape behaviour of $\bar{R}(t)$ (or $\bar{R}(T)$ for linearly varying temperature) [5]. The analysis presented here demonstrates that the results could not be explained by straightforward generalization of the

STM to multiple (even continuous) trap levels and recombination centres as well as overlapping emission bands. Temperature-dependent recombination probability may produce non-decreasing TL/TSC curves. However, these may be corrected by dividing the $R(t)$ function by the recombination probability $B(t)$, which should be known from other measurements. When the dependence is not known, it is still possible to eliminate the cases that could not be concerned with the STM with temperature-dependent recombination probability. To test this assumption one may study the function $R(t)/\sqrt{T(t)}$ (20), which for all basic one-carrier models should be monotone and decreasing. However, the easiest way is to perform isothermal decay measurements. In this case $R(t)$ should be decreasing for any one-carrier model considered in this paper. Consequently, in many cases the TL/TSC ratio provides a simple test that allow straightforward falsification of the basic kinetic models that are ‘routinely’ used for the description of these phenomena. Mathematically, the ‘monotone property’ of $\bar{R}(t)$, $R(t)$ and $R_T(t)$ expresses the necessary condition for STM to hold. Naturally, this is not the sufficient condition. Therefore, much precise verification of the STM should be carried out by referring directly to the basic set of differential equations governing charge carrier’s kinetics (e.g. Eqs. (1), (8) or (13)).

Finally, it was shown that the general rule presented here does not hold for simultaneous release of electrons and holes. For that reason, each case not obeying the simple ‘decreasing’ $R(t)$ property, could be suspected as a ‘two carrier’ system with simultaneous release of electrons and holes. Another possibility is a system with strong spatial correlation between traps and recombination centres. At this point, it is noteworthy to bring to mind an old supposition of Fields and Moran [17] who suggested that theoretical dependence between TL and TSC, defined by Eqs. (3) and (4), may not be fulfilled in a system with traps and recombination centres not homogeneously distributed in space. It refers to a wide class of spatially correlated systems that were not considered here. Detailed analysis of this hypothesis requires a different theoretical approach [18,19] and will be given in a separate paper.

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I would like to thank Prof. Andrzej Opanowicz for many inspiring discussions.

Appendix

To derive Eq. (7) we will consider Eq. (6) in two cases $t = 0$ and $t \rightarrow \infty$:

$$R_0 \equiv R(0) = Bh_0, \quad (\text{A.1})$$

$$R_{\text{end}} \equiv R(t \rightarrow \infty) = Bh_{\text{end}}. \quad (\text{A.2})$$

Limiting values h_0 and h_{end} can be calculated from Eq. (1c):

$$h_0 = n_0 + M, \quad (\text{A.3})$$

$$h_{\text{end}} = M. \quad (\text{A.4})$$

Here, it was assumed that at the beginning and at the end of the relaxation process n_c is negligible [4]. Combining these equations we finally come to

$$\frac{R_0}{R_{\text{end}}} = \frac{h_0}{h_{\text{end}}} = 1 + \frac{n_0}{M}. \quad (\text{A.5})$$

It is noteworthy that the formula does not contain the recombination coefficient. However, considering experimentally measured TL/TSC ratio (2) we get

$$\frac{\bar{R}_0}{\bar{R}_{\text{end}}} = \frac{\gamma(T_{\text{end}})}{\gamma(T_0)} \left(1 + \frac{n_0}{M}\right). \quad (\text{A.6})$$

When the dependence $\gamma(T)$ is unknown the relative density of deep traps n_0/M can be estimated by performing the experiment at a constant temperature (isothermal decay).

References

- [1] R. Chen, S.W.S. McKeever, *Theory of Thermoluminescence and Related Phenomena*, World Scientific, Singapore, 1997.
- [2] R. Chen, *J. Appl. Phys.* 42 (1971) 5899.
- [3] J. Gasiot, J.P. Fillard, *J. Appl. Phys.* 48 (1977) 3171.
- [4] A. Mandowski, J. Świątek, *J. Phys. D* 25 (1992) 1829.
- [5] A. Mandowski, J. Świątek, P. Iaconi, R. Bindi, *Radiat. Prot. Dosimetry* 100 (2002) 187.
- [6] A. Opanowicz, *J. Appl. Phys.* 86 (1999) 6189.
- [7] A. Opanowicz, *J. Phys. D* 33 (2000) 1635.
- [8] A. Opanowicz, P. Pietrucha, *J. Appl. Phys.* 93 (2003) 957.
- [9] B.J. Luff, P.D. Townsend, *Meas. Sci. Technol.* 3 (1992) 65.
- [10] E. Mandowska, P. Bilski, E. Ochab, J. Świątek, A. Mandowski, *Rad. Prot. Dosimetry* 100 (2002) 451.
- [11] A. Mandowski, J. Świątek, in: M. Vázquez, A. Hernando (Eds.), *Nanostructured and Non-crystalline Materials*, World Scientific, Singapore, 1995, p. 227.
- [12] W. Tomaszewicz, B. Jachym, *J. Non-Cryst. Solids* 65 (1984) 193.
- [13] W. Tomaszewicz, *J. Phys.: Condens. Matter* 4 (1992) 3967.
- [14] W. Tomaszewicz, *J. Phys.: Condens. Matter* 4 (1992) 3985.
- [15] R. Chen, R.J. Fleming, *J. Appl. Phys.* 44 (1973) 1393.
- [16] S.W.S. McKeever, J.F. Rhodes, V.K. Mathur, R. Chen, M.D. Brown, R.K. Bull, *Phys. Rev. B* 32 (1985) 3835.
- [17] D.E. Fields, P.R. Moran, *Phys. Rev. B* 9 (1974) 1836.
- [18] A. Mandowski, *Radiat. Prot. Dosimetry* 100 (2002) 115.
- [19] A. Mandowski, *J. Phys. D* 38 (2005) 21.