



# A model explaining the anomalous heating-rate effect in thermoluminescence as an inverse thermal quenching based on simultaneous thermal release of electrons and holes



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## HIGHLIGHTS

- The anomalous heating rate effect of thermoluminescence (TL) is explained.
- A model with one electron trap, a hole center and a hole-reservoir center is used.
- With a small change in the model, the effect of thermal quenching can be explained.
- A recently found case in which one TL peak decreases with heating rate and another peak increases can also be interpreted.

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## ABSTRACT

A model is presented which explains the anomalous heating-rate effect of thermoluminescence (TL) in which the peak area increases with increasing heating rate. In a similar way to the Schön-Klasens model, the present model is based on delocalized transitions only. In addition to the occurrence of an electron trapping state and a hole recombination center, we assume the participation of a hole reservoir which competes with the other levels and participates in the process during both the excitation and the read-out stages. Moreover, we assume that the reservoir is close enough to the valence band so that holes may be thermally released in the same temperature range in which electrons are thermally raised into the conduction band. Simulations with this model show that, with certain sets of trapping parameters, an increase of the heating rates results in an increase in the area under the normalized TL curve. Inverting the roles of the recombination center and the reservoir so that the recombination of a free electron with a hole in the reservoir is assumed to be radiative and the other recombination is radiationless yields opposite results. Increasing the heating rates causes a significant decrease in the area under the TL curve which is a demonstration of the well-known thermal quenching heating-rate effect of TL. An intuitive qualitative explanation of these two effects within the proposed model is given. A recently discovered case in which two consecutive TL peaks respond to the heating rate change in opposing directions, one decreases and the other increases with increasing heating rate can also be explained by this model.

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## 1. Introduction

An important experimental parameter in the study of thermoluminescence (TL) is the heating rate, usually constant, denoted by  $\beta$  ( $\text{Ks}^{-1}$ ). In practically all known cases, the TL glow peak shifts to higher temperature with increasing heating rate (see e.g., Booth, 1954; Bohun, 1954; Parfianovich, 1954; Hoogenstraaten, 1958).

This can be easily shown for the simple case of first-order kinetics. The equation for the maximum in this case is

$$\beta = (sk/E)T_m^2 \exp(-E/kT_m), \quad (1)$$

where  $E$  (eV) is the activation energy,  $s$  ( $\text{s}^{-1}$ ) the frequency factor,  $k$  ( $\text{eV}\cdot\text{K}^{-1}$ ) Boltzmann's constant and  $T_m$  (K) the temperature at the maximum. When  $\beta$  increases, the right-hand side must increase by the same amount. However, since  $T_m^2 \exp(-E/kT_m)$  is an increasing function of  $T_m$ , the increase in its value implies that  $T_m$  must

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increase. It has been shown (e.g., [Chen and Winer, 1970](#)) that although in more complex cases of TL kinetics Eq. (1) does not hold precisely, it can serve in many cases as an approximation. Anyway, the shift of a TL peak to higher temperatures with increasing heating rates seems to be a general property.

A distinction should be made between two alternative presentations of TL. As an example, let us consider first the simplest example of first-order kinetics. The governing equation is

$$I(t) = -\frac{dn}{dt} = s \cdot n \cdot \exp(-E/kT), \quad (2)$$

where  $n$  ( $\text{cm}^{-3}$ ) is the concentration of trapped electrons,  $T$  (K) is the absolute temperature and  $t$  (s) is time. In fact, a proportionality factor is missing between  $I(t)$  and  $-dn/dt$ , which is set here arbitrarily to unity. As is, the units of  $I(t)$  are  $\text{cm}^{-3}\text{s}^{-1}$  whereas the real intensity is given in photons per second or emitted energy per second. As shown before (e.g. [Kumar et al., 2006, 2010](#)), when increasing the heating rate, the maximum intensity increases nearly proportionally. The area under the curve must remain the same for all heating rates and the simple explanation for the increased intensity is that when heating is faster, the peak gets much narrower on the time scale. An alternative, quite common presentation is reached by normalizing the intensity as defined in Eq. (2) by dividing it by the heating rate  $\beta$ ; unfortunately, this is also usually termed in the literature “intensity”. This magnitude is usually plotted as a function of temperature rather than time. Thus, the area under the curve remains constant with different heating rates ([Kumar et al., 2010](#)), and this normalized intensity, which is  $I(T) = -dn/dT$  ( $\text{cm}^{-3}\text{K}^{-1}$ ) has a slightly decreasing maximum value associated with a slight broadening of the peak with increasing heating rate. It should be noted that this property of a decrease of the normalized intensity with the heating rate is not limited to the first-order case and is seen in many more complicated situations.

[Wintle \(1974, 1975\)](#) reported on thermal quenching of TL in quartz which is the decrease in luminescence efficiency with the rise in temperature. In fact, this effect had been known before for other luminescence effects ([Curie, 1963](#); [Morehead, 1966](#)). Two models have been suggested for the explanation of the effect. According to the theory of Mott and Seitz ([Seitz, 1939](#); [Mott and Gurney, 1948](#)) radiative and non-radiative transitions compete within the confines of the luminescence centers as is the case for KCl(Tl). The luminescence efficiency decreases with increasing temperature due to a reduction in the quantum efficiency of the luminescence centers (see e.g. [Yukihara and McKeever, 2011](#)).

An alternative explanation of the thermal quenching has to do with the Schön-Klasens model. [Schön \(1942\)](#) and [Klasens \(1946\)](#) suggested a model in which the holes in centers may be thermally released into the valence band, thus decreasing the number of holes available for recombination with thermally stimulated electrons (see [Bräunlich and Scharmann, 1966](#); [McKeever et al., 1985](#); [Yukihara and McKeever, 2011](#)). A similar model with traps and centers releasing electrons and holes, respectively, simultaneously have been discussed by [Chen et al. \(2008\)](#) and [Lawless et al. \(2009\)](#) and explained the possibility of “duplicitous” TL peaks. Another model of thermal quenching, which takes into account thermal ionization of excited states of F-centers has been given by [Nikiforov et al. \(2001\)](#) (see also [Chen and Pagonis, 2011](#)).

As reported by several authors ([Nanjundaswamy et al., 2002](#); [Rasheedy and Zahran, 2006](#); [Kafadar, 2011](#); [Kalita and Wary, 2012](#); [Subedi et al., 2010a, 2010b](#)), the decrease of the maximum of normalized TL with the heating rate was significantly faster than the slight decrease described above. The common explanation has been that since the peak shifts to higher temperature with the heating rate, its intensity decreases due to the thermal quenching.

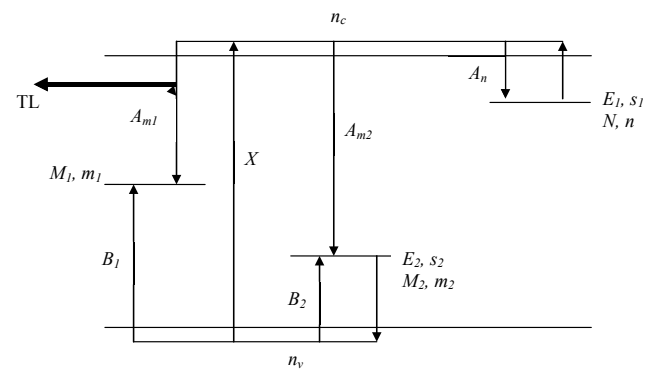
Thus, the area under the normalized curve is not constant but rather, it decreases with increasing heating rate.

In recent years, a number of reports on an inverse, anomalous heating-rates effect have been published. [Kitis et al. \(2006\)](#) have reported on a heating-rate effect in fluorapatite in which the maximum normalized TL intensity and the area under the curve increased with the heating rate. [Pradhan et al. \(2008\)](#) have described the effect in LiF:Mg,Cu,Si. [Bos et al. \(2010\)](#) described a similar effect in  $\text{YPO}_4:\text{Ce}^{3+}, \text{Sm}^{3+}$ . [Delice et al. \(2014\)](#) reported the effect in  $\text{Ti}_2\text{GaInS}_4$  and [Benabdesselam et al. \(2014\)](#) found it in Ge-doped silica-based optical fiber (GDF). [Nsengiyumva et al. \(2014\)](#) have reported on the normal heating-rate effect in synthetic quartz and the inverse effect in similar quartz previously implanted by 60 keV  $\text{N}^+$  ions. [Delice et al. \(2015\)](#) have recently described the results of TL in GaS. Two peaks were reported for different heating rates. Whereas the lower-temperature normalized peak decreased with the heating rate, the higher-temperature one increased with  $\beta$ . [Mandowski and Bos \(2011\)](#) explained the effect in  $\text{YPO}_4:\text{Ce}^{3+}, \text{Sm}^{3+}$  by a semi-localized-transitions model. [Pagonis et al. \(2013\)](#) used a simplified semi-localized-transition model to explain the anomalous heating-rate effect.

In the present work we concentrate on a version of the non-localized Schön-Klasens model which can explain the anomalous inverse heating-rate effect of TL, using a model which includes an additional reservoir center. The concept of such a center has been used successfully in the past to explain the sensitization effect in quartz ([Zimmerman, 1971](#)). With a different version of the same model we demonstrate by numerical simulation the cases in which there is a strong decrease of the TL intensity with increasing heating rates, i.e., the normal thermal quenching of TL.

## 2. The model

The model including one electron trap  $N$ , one recombination center  $M_1$  and one reservoir  $M_2$  is shown in [Fig. 1](#). We assume that the electron trap is close enough to the conduction band so that electrons are thermally raised into the conduction band during the



**Fig. 1.** Schematic energy level diagram including an electron trap  $N$  ( $\text{cm}^{-3}$ ) with instantaneous occupancy of  $n$  ( $\text{cm}^{-3}$ ), a hole center  $M_1$  ( $\text{cm}^{-3}$ ) with instantaneous hole occupancy of  $m_1$  ( $\text{cm}^{-3}$ ), and another hole center  $M_2$  ( $\text{cm}^{-3}$ ) with instantaneous occupancy of  $m_2$  ( $\text{cm}^{-3}$ ). The instantaneous concentrations of free electrons and holes are  $n_c$  ( $\text{cm}^{-3}$ ) and  $n_v$  ( $\text{cm}^{-3}$ ), respectively. The activation energy of trapped electrons is  $E_1$  (eV) and the frequency factor is  $s_1$  ( $\text{s}^{-1}$ ). The center  $M_2$  is considered to be close enough to the valence band and holes can be released thermally into the valence band with an activation energy of  $E_2$  (eV) and frequency factor  $s_2$  ( $\text{s}^{-1}$ ). The trapping probability coefficient of electrons is  $A_n$  ( $\text{cm}^2\text{s}^{-1}$ ). The trapping probability coefficients of holes into  $M_1$  and  $M_2$  are  $B_1$  ( $\text{cm}^3\text{s}^{-1}$ ) and  $B_2$  ( $\text{cm}^3\text{s}^{-1}$ ), respectively. The recombination probability coefficients into  $M_1$  and  $M_2$  are  $A_{m1}$  ( $\text{cm}^3\text{s}^{-1}$ ) and  $A_{m2}$  ( $\text{cm}^3\text{s}^{-1}$ ), respectively. The rate of production of electron and hole pairs, proportional to the dose rate is denoted by  $X$  ( $\text{cm}^{-3}\text{s}^{-1}$ ).

heating stage. We also assume that  $M_1$ , the recombination center is far enough from the valence band so that no holes are thermally released from it into the valence band during heating. As for the reservoir  $M_2$ , we assume that it may capture holes from the valence band during excitation and that electrons from the conduction band may recombine with the holes both during the excitation and the heating stages. We also assume that the reservoir is close enough to the valence band so that holes may be thermally released from it into the valence band which may be either retrapped in this center or be trapped in the center  $M_1$  during the heating stage. In a sense, this is a Schön-Klasens model with an additional reservoir center. The meanings of the different parameters and functions (carriers concentrations) shown in Fig. 1 are given in the caption. Note that the rate of production of electron and hole pairs in the conduction and valence bands, respectively,  $X$  ( $\text{cm}^{-3}\text{s}^{-1}$ ) is proportional to the dose rate of excitation. If the excitation takes place for  $t_D$  seconds, the amount  $D=X \times t_D$  is the total number of electron-hole pairs produced by the irradiation, which is proportional to the total dose applied.

The set of coupled differential equations governing the relevant processes is

$$\frac{dn}{dt} = A_n(N - n)n_c - s_1 n \exp(-E_1/kT), \quad (3)$$

$$\frac{dm_1}{dt} = B_1 n_v(M_1 - m_1) - A_{m1} m_1 n_c, \quad (4)$$

$$\frac{dm_2}{dt} = B_2 n_v(M_2 - m_2) - A_{m2} m_2 n_c - s_2 m_2 \exp(-E_2/kT), \quad (5)$$

$$\frac{dn_c}{dt} = X - A_n(N - n)n_c - A_{m1} m_1 n_c - A_{m2} m_2 n_c, \quad (6)$$

$$\frac{dn_v}{dt} = \frac{dn}{dt} + \frac{dn_c}{dt} - \frac{dm_1}{dt} - \frac{dm_2}{dt}. \quad (7)$$

The process of TL consists of three stages. The first one is the excitation. For this, we solve the set of equations with the given value of  $X$  for  $t_D$  seconds. In order to follow the experimental procedure, we perform next the relaxation stage. Before the beginning of heating, one holds the sample at ambient temperature for a certain period of time in which most free electrons and holes are trapped in traps/centers or perform recombination. To simulate this stage, we use the final values of all the concentration functions reached in the first stage and solve the same set of equations with  $X = 0$  for a time long enough so that  $n_c$  and  $n_v$  become negligibly small. In the next stage of heating, we solve the same set of equations (with  $X = 0$ ), using the final values in the second stage as initial values for the third stage. Here, we use a linear heating function  $T=T_0+\beta t$  where  $t$  is the time,  $T_0$  the initial temperature,  $T$  the varying temperature and  $\beta$  the heating rate which is constant for a certain run and is varied from one curve to another. We assume here that the recombination of an electron from the conduction band with a hole in  $M_1$  is radiative whereas the recombination with a hole in  $M_2$  is non-radiative. The emitted light intensity in this case is given by

$$I(t) = A_{m1} m_1 n_c, \quad (8)$$

and the mentioned normalized intensity is

$$I(T) = A_{m1} m_1 n_c / \beta. \quad (9)$$

It should be noted that in more elementary TL models, where valence-band holes are not involved in the read-out process, the

simple way to present the TL intensity is  $I(t) = -dm_1/dt$ , but since here the concentration  $m_1$  varies by two channels, trapping of holes from the valence band and recombination with electrons from the conduction band, Eqs. (8) and (9) are the correct way to present the emission of light due to recombination. In order to see the variations with the heating rate we repeat the whole procedure with a different value of  $\beta$ . With this simulation procedure and the chosen set of parameters we have been able to show the possible occurrence of the anomalous heating-rate effect within this model that does not include localized transitions, as discussed below.

We could also show, with a slight change in the argument that a strong decrease of the normalized TL intensity with the heating rate, similar to that usually explained as the result of thermal quenching, can be reached with nearly the same model. In fact, we use the same set of equations and the same sequence of three stages. The one different assumption we make here is that now, the transition into  $M_2$  is considered to be radiative whereas the transition into  $M_1$  is considered to be non-radiative and therefore, this center acts only as a competitor. Obviously, the TL intensity here is defined as

$$I(t) = A_{m2} m_2 n_c, \quad (10)$$

and the normalized intensity is

$$I(T) = A_{m2} m_2 n_c / \beta. \quad (11)$$

### 3. Numerical results

Fig. 2 shows the simulated normalized TL peaks reached by solving numerically Eqs. (3)–(8) in the sequence described above; the parameters used are given in the caption. The sets of equations have been solved numerically, in the three stages of excitation, relaxation and heating, using the Matlab ode15s solver, designed to deal with stiff sets of equations. The shift of the peak to higher

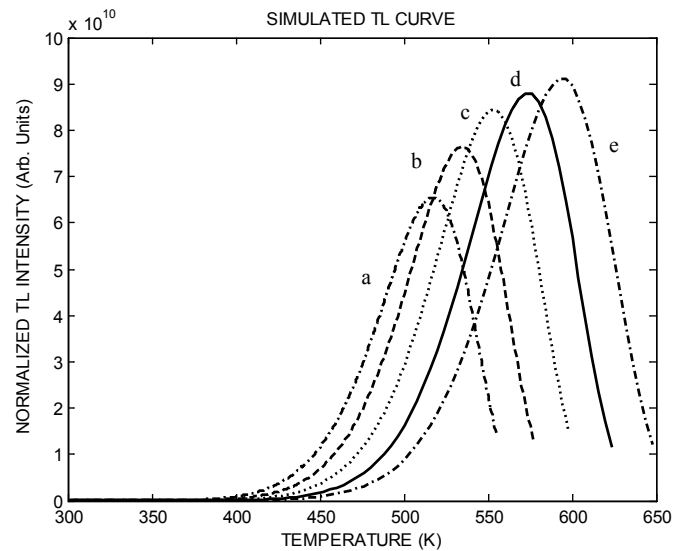


Fig. 2. Simulation of the anomalous heating-rates effect by numerical solution of the sets of equations. The parameters used are:  $E_1 = 0.9$  eV;  $s_1 = 10^{11} \text{ s}^{-1}$ ;  $E_2 = 0.7$  eV;  $s_2 = 10^{13} \text{ s}^{-1}$ ;  $N = 10^{14} \text{ cm}^{-3}$ ;  $M_1 = 10^{11} \text{ cm}^{-3}$ ;  $M_2 = 10^{14} \text{ cm}^{-3}$ ;  $A_{m1} = A_{m2} = 10^{-8} \text{ cm}^3 \text{ s}^{-1}$ ;  $B_1 = 10^{-7} \text{ cm}^3 \text{ s}^{-1}$ ;  $B_2 = 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ ;  $A_n = 10^{-7} \text{ cm}^3 \text{ s}^{-1}$ ;  $X = 10^{12} \text{ cm}^{-3} \text{ s}^{-1}$ . The excitation time was 10 s and the relaxation time between excitation and heating was 220 s. The heating rates used were: (a)  $0.5 \text{ Ks}^{-1}$ ; (b)  $1.0 \text{ Ks}^{-1}$ ; (c)  $2.0 \text{ Ks}^{-1}$ ; (d)  $4.0 \text{ Ks}^{-1}$ ; (e)  $8.0 \text{ Ks}^{-1}$ . Transition into  $M_1$  is assumed to be radiative and into  $M_2$  non-radiative. Heating-rate normalized intensities are shown.

temperature with increasing heating rate is observed. It is readily seen that the normalized maximum intensity of the TL peak as well as the normalized area under the curve increases with the heating rate as seen in a number of experimental cases mentioned above. The intensities shown are those determined by Eq. (9), associated with the transitions into the recombination center  $M_1$ , the center which is far enough from the valence band and therefore, does not release holes thermally.

Fig. 3 depicts the results of the same simulations, with the same set of parameters, except that the emission is assumed to be associated with the recombination with  $M_2$ , the center which loses holes thermally into the valence band since its activation energy is rather small. The normalized TL intensity is determined here by Eq. (11). Here too, the TL peak shifts to higher temperature with increasing heating rate. The significant decrease in the signal, both of the maximum intensity and the area under the normalized curve is evident.

Fig. 4 presents the dependence of the simulated area under the TL curve on the heating rate within the two-center one-trap model. The dash-dotted line depicts the results when  $M_1$  is assumed to be radiative and  $M_2$  non-radiative. The behavior is thermal-quenching like, namely, the normalized integrated intensity decreases quite significantly with the heating rate. The dotted line deals with the anomalous heating-rate effect. The assumption here is that  $M_2$  is radiative and  $M_1$  non-radiative and the normalized integrated area be increasing with the heating rate. The dashed line shows the sum of these intensities for each heating rate as a function of the heating rate; the sum is seen to be nearly constant.

Fig. 5 shows the variation with the heating rate of the glow curve consisting of the sum of the two emissions into  $M_1$  and  $M_2$ , assuming that both transitions are radiative and with the same efficiency. The lower temperature peak decreases with increasing heating rate whereas the higher temperature peak increases with the heating rate. These results are qualitatively similar to the experimental results in GaS crystals, given by Delice et al. (2015). The parameters used here are the same as in Figs. 2–4 except that here we use  $M_1 = 10^9 \text{ cm}^{-3}$ .

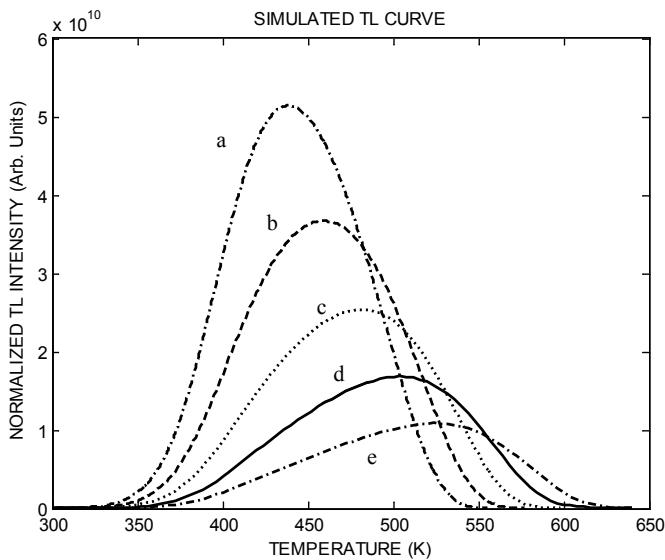


Fig. 3. Results of the same simulations with the same parameters and heating rates except that here it is assumed that recombination into  $M_2$  be radiative and into  $M_1$  is non-radiative. Normalized intensity is shown. The results show the effect of thermal quenching as described in the text.

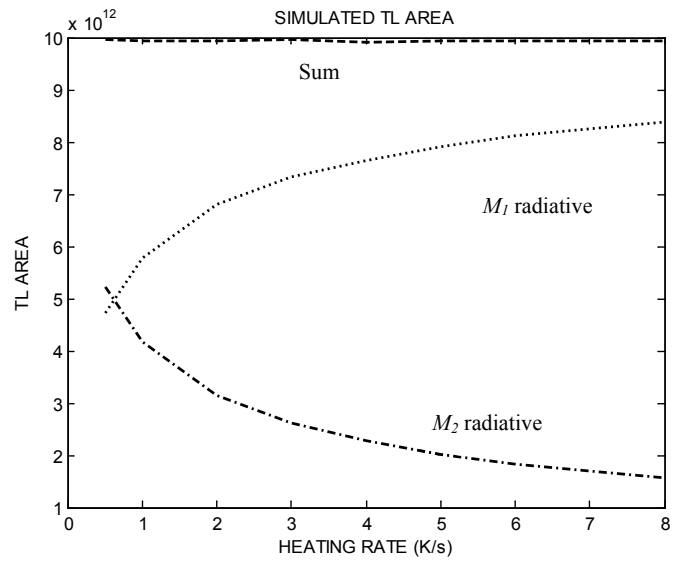


Fig. 4. Dependence of the areas under the TL peak on the heating rate  $\beta$ . “ $M_2$  radiative” shows the thermal-quenching like results whereas “ $M_1$  radiative” depicts the results of the anomalous heating rate effect. “Sum” gives the sum of the two transitions.

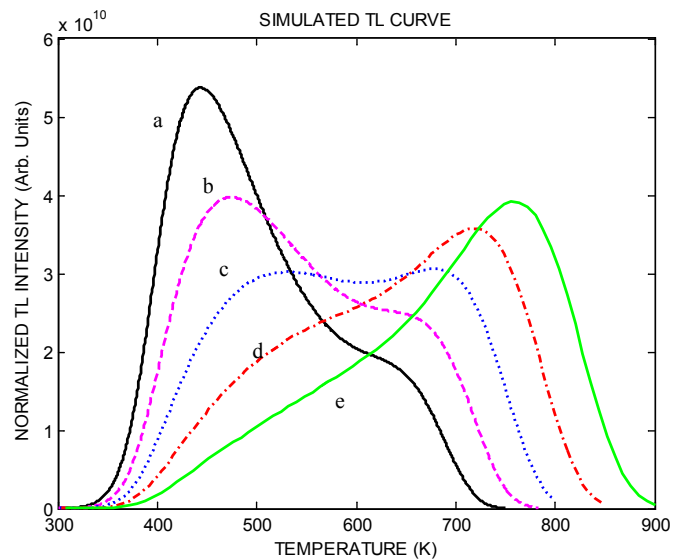


Fig. 5. Simulated TL curves assuming that both transitions into  $M_1$  and  $M_2$  are radiative. The parameters are the same as above except that here  $M_1 = 10^9 \text{ cm}^{-3}$ . The heating rates are between 0.5 and  $8 \text{ K}^{-1}$  as in Fig. 2.

#### 4. Discussion

In this work, we have dealt with two energy-level models explaining two different possible behaviors of TL peaks with the change in heating rate. One is the well known significant decrease in the normalized intensity of TL with an increase of the heating rate usually associated with thermal quenching ascribed to the decrease in the luminescence probability with increasing temperature. The other is the opposite “anomalous” effect, recently discovered in a number of materials, in which an increase in the heating rate results in an increase in the normalized TL intensity. The models we propose that can explain the effect are associated with the Schön-Klasens assertion that under the appropriate conditions, holes in centers may be thermally released into the valence



band simultaneously with the thermal release of electrons from traps into the conduction band. Both models are associated with the energy-level diagram shown in Fig. 1. In both cases, in addition to the transition of electrons from the  $N$  trap to the recombination center  $M_1$  through the conduction band (and including retrapping), we consider the traffic of carriers through  $M_2$  and allow for thermal release of holes from  $M_2$  into the valence band. The intuitive explanations of the two opposing results in the two versions of the model are as follows.

In the first version of the model, we assume that the radiative transition occurs due to recombination of free electrons with  $M_1$ . Therefore, the area under the TL peak is not proportional to the initial occupancy of this center because during the heating, more holes are added to  $M_1$  which also contribute to the emitted light. While the heating rate increases, the peak shifts to higher temperature, and more holes move through the valence band to  $M_1$  and are then involved in the radiative process. Therefore, with higher heating rate, the area under the normalized TL curve and the maximum intensity increase with the heating rate. This is a possible explanation of the anomalous, inverse heating-rate effect of TL.

In the second version, we assume that the transition into  $M_2$  is radiative and to  $M_1$  is non-radiative. Here, if holes are thermally released from  $M_2$  and some of them are trapped in  $M_1$ , the total number of holes contributing to TL is reduced during heating. Since with an increase of the heating rate the peak shifts to higher temperature, more holes may be lost at higher heating rates, hence the thermal-quenching-like effect of a decrease of the area under the normalized peak and the maximum intensity with increasing heating rates.

The numerical results in Fig. 4 sum up the heating-rate dependence of the two possible versions of the model namely, where only  $M_1$  or only  $M_2$  is radiative. The decrease and increase in the two versions with heating rate, respectively, are clearly seen and the two curves look like a mirror image of each other. The “sum” line is nearly constant with the heating rate. As for Fig. 5, it demonstrates that if we assume that both transitions into  $M_1$  and  $M_2$  are radiative, the experimental results given by Delice et al. (2015) for GaS, namely that the lower-temperature peak decreases and the higher temperature peak increases with the heating rate, can be qualitatively explained. It should be mentioned that Delice et al. did not report on the emission spectra of the two peaks in question. If, as the present model suggests, the two peaks result from transitions into different centers, their emission spectrum is expected to be different. It would be interesting to check this point experimentally.

A close look at Fig. 2 reveals that the curves look approximately like first-order peaks. The peaks are asymmetric with shape factors  $\mu_g$  between 0.39 and 0.42, averaging 0.406 for the five heating rates studied. This is slightly smaller than the “classical” value of 0.42 for first-order peaks (see e.g. Chen, 1969). The activation energies evaluated by the full-width shape method vary between 0.71 eV and 0.84 eV, within the range between  $E_2 = 0.7$  eV and  $E_1 = 0.9$  eV of the activation energies used in the simulations. As for the peaks shown in Fig. 3, they look broader than “normal”, and the shape varies with the heating rate. The analysis of the peak shape shows symmetry factors going from  $\mu_g = 0.54$  for the heating rate  $\beta = 0.5$  K/s gradually to  $\mu_g = 0.39$  with  $\beta = 8$  K/s. As for the evaluated activation energies by the same full-width shape method, it varied from  $E_{eff} = 0.54$  eV at  $\beta = 0.5$  K/s to  $E_{eff} = 0.27$  eV at  $\beta = 8$  K/s. These relatively low values are directly connected to the fact that the peaks shown in Fig. 3 are very broad. One should not be too surprised to see that the evaluated energies are far from the inserted ones and that the symmetry properties as well as the effective activation energies vary with the heating rates. The original conclusions that the symmetry factor is practically

independent of the heating rate and that the developed formulae for the activation energies are valid for all heating rates were based on the assumption that there is only one channel of changing the concentration of holes in centers during heating, namely, the recombination of conduction-band electrons with holes in centers. This is not the situation here since holes are depleted thermally from the centers and go to the valence band simultaneously with the process of recombination. As seen in Figs. 2 and 3, with the given set of trapping parameters, this has a smaller effect on the TL associated with transitions into  $M_1$  and much more significant effect on transitions into  $M_2$ .

It should be emphasized that the present models do not contradict the previously published explanations of the normal heating-rate effect associated with thermal quenching and the anomalous effect ascribed to localized transitions. The proposed models are merely a possible alternative which may apply in some cases where these effects take place.

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