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Theoretical Investigation of Thermoluminescence in the Presence of Thermally Disconnected Deep Traps

Nebiyu Gemechu[1], Tamirat Abebe[1], Kebede Shogile[1], Solomon H/Mariam[1], Shimelis Addisu[2]

[1]Jimma University, Department of Physics, P. O. Box 378, Jimma, Ethiopia. E-mail: nafsif@gmail.com

[2]Jimma University, Department of Chemistry, P. O. Box 378, Jimma, Ethiopia

ABSTRACT: Thermoluminescence (TL) phenomenon is theoretically investigated using two active electron traps and one recombination center. The presence of one and two thermally disconnected deep traps (TDDTs) is taken in to account. The rate equations corresponding to this model are clearly formulated and the results are obtained using MTHEMATICA code. In particular, the variation of concentration of electrons in the active and thermally disconnected deep traps and the TL intensity are generated for various TL kinetic parameters such as activation energy and the frequency factor. The results show that the concentration of electrons in the active electron traps decrease with temperature while that in the TDDTs increase with temperature and approaches a constant. The generated TL glow curve (for $E_1 = 0.9\ eV$ and $E_2 = 1\ eV$) has two maxima. However, one broad glow curve with only one maximum is obtained when E_1 and E_2 are closer to each other. The profiles of the TL glow curves and the variation of concentration of electrons in the conduction band are the same except for their intensity.

Keywords: Thermally Disconnected Deep Traps, Thermoluminscence, Rate Equations, Active Electron Traps, Activation Energy, and Frequency Factor.

INTRODUCTION

Thermoluminescence (TL), which has important applications in various fields, is the thermally stimulated emission of light following the previous absorption of energy from radiation. TL is also called radiation-induced thermally stimulated luminescence or simply thermally stimulated luminescence (TSL) (Belayneh Mesfin and Teshome Senbeta, 2019; Sunta C., 2014; McKeever S., 1988; Pagonis V. et al., 2006; Sadek M. et al., 2014).

TL mechanisms are usually described using the energy band theory of solids. Irradiation of a solid sample results in trapping of electrons and holes in the trap centers of the sample. These trap centers are associated with impurity or defect created by imperfections. The energy absorbed during excitation liberates electrons and holes that are free to migrate in the conduction and valence bands, respectively, and then to be trapped in the electron and hole allowed states. If the energy levels of the traps are sufficiently close to the bands, the trapped charge carriers will be thermally released into their respective bands during heating (Sanga S. et al., 2018; Parganiha Y. et al., 2015; Kumar V., et al., 2015; Parganiha Y. et al., 2015; Ortega F. et al., 2017). However, charge carriers trapped to very deep traps will not be released into their respective bands regardless of the amount of heat applied during heating. Such traps are called thermally disconnected deep traps (TDDTs). However, such trapping states are most likely to result in recombination of a mobile carrier and trapped charges of opposite sign, and these are usually referred to as recombination centers. Thus, either electrons or holes can recombine with charges of opposite sign, and a certain fraction of the previously absorbed energy is released as photons (Bos J., 2006; Bos J., 2017; Chen R. and McKeever S., 1997).

The common models of TL implemented in theoretical approaches include the one-trap-one-recombination center (OTOR) and the interactive-multi-trap system (IMTS). The OTOR model is the

*Author to whom correspondence should be addressed.

simplest possible model that has been used to describe the process by which materials emit light when heated. It consists of two localized levels: an isolated electron trap and a recombination center. This approach is commonly called one-trap-one-recombination center (OTOR) model (Belayneh Mesfin and Teshome Senbeta, 2019; Sunta C., 2014). Moreover, the interactive multi-trap system (IMTS) model assumes; one active electron trap, one thermally disconnected deep trap (TDDT) that cannot be thermally activated, and one recombination center (McKeever S., 1988; Bos J., 2017).

However, these models are very simple in their nature that they cannot fully describe a real TL material. Therefore, making TL more efficient for practical applications without deviation from reality should be given more attention in theoretical approaches. Real TL materials involve several impurities or defect levels within the band gap which act as localized energy levels. This results in complex rate equations governing the TL kinetics which cannot be solved analytically in general. Real TL materials contain several traps and recombination centers and hence the OTOR and IMTS models are insufficient to describe the TL process. Moreover, a given electron or hole may be trapped several times before recombination, increasing the order of kinetics. Therefore, in this work, TL phenomenon is discussed in detail by using two active electron traps and one recombination center. Moreover, the presence of one and two TDDTs is taken in to account.

Two Active Electron Traps, One TDDT, and One Recombination Center Model of TL

Figure 1 shows two active traps, one thermally disconnected deep trap (TDDT) and one recombination center model of TL. The two active electron traps are located at the trap depths of E_1 and E_2 beneath the edge of the conduction band.

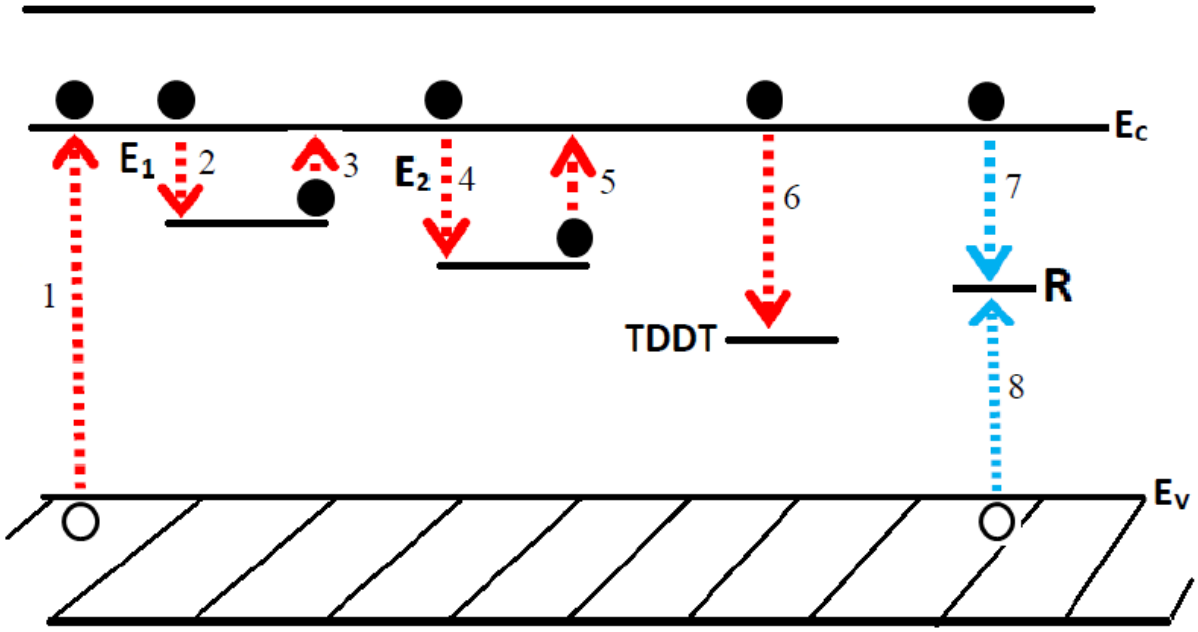

Fig. 1: Two active traps, one thermally disconnected deep trap (TDDT) and one recombination center model of TL. Allowed transitions: (1) ionization; (2), (4), (6), (8) trapping; (3), (5) de trapping; (7) radiative recombination and emission of light.

The rate equations corresponding to this model are (Pagonis V., et al., 2006; Asamin Regassa and Belayneh Mesfin, 2018; Mady F., et al., 2006; Kitis G. and Pagonis V., 2023):

$$\frac{dn_1}{dt} = -s_1 n_1 \exp\left\{-\frac{E_1}{kT}\right\} + cA_1(N_1 - n_1), \qquad (1)$$

$$\frac{dn_2}{dt} = -s_2 n_2 \exp\left\{-\frac{E_2}{kT}\right\} + cA_2(N_2 - n_2), \qquad (2)$$

$$\frac{dm}{dt} = cA_m(M - m), \qquad (3)$$

$$\frac{dc}{dt} = s_1 n_1 \exp\left\{-\frac{E_1}{kT}\right\} + s_2 n_2 \exp\left\{-\frac{E_2}{kT}\right\} - c[A_1(N_1 - n_1) + A_2(N_2 - n_2) + A_m(M - m) + A_r n_h]. \qquad (4)$$

Here $n_i\ (i = 1,2)$ is the concentration of trapped electrons (cm^{-3}) on the levels $E_i(eV)$, $N_i\ (i = 1,2)$ is the total concentration of traps (cm^{-3}) on the levels $E_i(eV)$, k is the Boltzmann constant (JK^{-1}); $s_i(i = 1,2)$ is the probability of electron to escape from the i −th trap (s^{-1}), $A_i(i = 1,2)$ is the re-trapping probability on the i −th energy level $(cm^3 s^{-1})$; A_r is the radiative recombination probability $(cm^3 s^{-1})$, c and n_h are the concentration of electrons in the conduction band and in the recombination centers, respectively (cm^{-3}), m is the instantaneous concentration of electrons in the TDDT (cm^{-3}), A_m is the probability coefficient of electron retrapping to the TDDT $(cm^3 s^{-1})$, and M is the total concentration of traps in the TDDT (cm^{-3}). It is assumed that $N_1 = N_2 = N_3 = N$. The term s is commonly called the 'frequency factor', although when applied to TL it is often called 'attempt-to-escape frequency'. The usual interpretation of s is that it represents the number of times per second that a bound electron interacts with lattice phonons times transition probability (Parganiha Y., et al., 2015). The maximum value expected for s is therefore the lattice vibration frequency, namely $10^{12} - 10^{14} s^{-1}$. The system (1-4) is closed by the quasi-neutrality condition

$$n_1(t) + n_2(t) + c(t) + m_1(t) = n_h(t) \qquad (5)$$

Here $n_h(t)$ is the instantaneous concentration of holes in hole trap. The temperature profile can be expresses as

$$T(t) = T_0 + \beta t, \qquad (6)$$

where β is the heating rate. In our case, $N = 10^{10}$, $M = 10^{10}$, $A_1 = A_2 = A_m = 10^{-5}$, $\beta = 1\text{degree}/s$ are used in all the results. Fixing these parameters is based on the most common values reported in literature (McKeever S., 1988; Pagonis V., et al., 2006).

Moreover, the initial concentration of filled traps is taken to be $n_1(0) = n_2(0) = m(0) = 10^9$.

The intensity of the TL is given by

$$I(T) = A_r n_h(T) c(T) \qquad (7)$$

The results of equations (1-7) are generated using MATHEMATICA 11 and discussed in detail in the next section.

RESULTS AND DISCUSSION

Figure 2 shows the variation of concentration of electrons in the first trap located at depth $E_1 = 0.9\ eV$ beneath the edge of the conduction band for various values of the attempt to escape frequency. At a given temperature, the de-trapping probability of the charge carriers from the defect centers increases with an increase in the attempt to escape frequency. Therefore, there is a high number of electrons de-trapped back to the conduction band increasing the recombination rate. This is the reason the peaks showing the variation of concentration of electrons in the trap shift to the low temperature region with increase in the value of s.

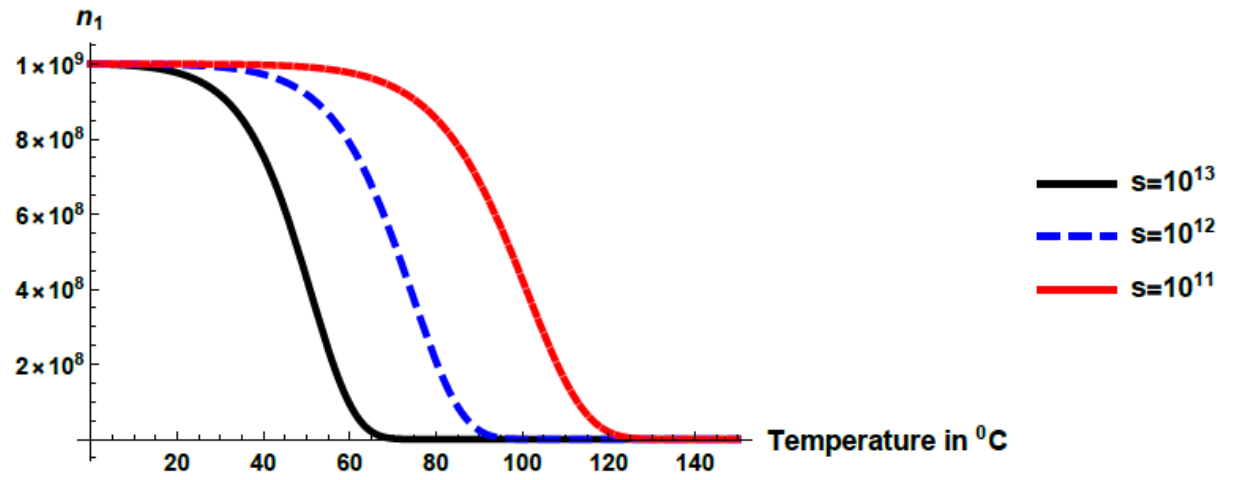

Fig. 2: Variation of concentration of electrons in the first active electron trap for various values of the frequency factor.

Moreover, it can be seen that for a given value of the frequency factor, there is a minimum temperature for which the free charge carriers start de-trapping from the defect center. This temperature is in such a way that the minimum kinetic energy of the charge carries is equal to the trap depth, which is also called the activation energy.

Figure 3 shows the variation of concentration of electrons in the second trap located at depth $E_2 = 1\,eV$ beneath the edge of the conduction band for various values of the attempt to escape frequency s. The peaks showing the variation of concentration of electrons in the second trap with $E_2 = 1\,eV$ is shifted to higher temperature region as compared to Fig. 2. This is because of the fact that the second trap is deeper as compared to the first and hence relatively high temperature is required to depopulate it. However, the general behavior of the peaks is the same as that shown in Fig. 2. It is interesting that the results shown in Figs. 1 and 2 can only be predicted from theoretical computations and cannot be generated experimentally.

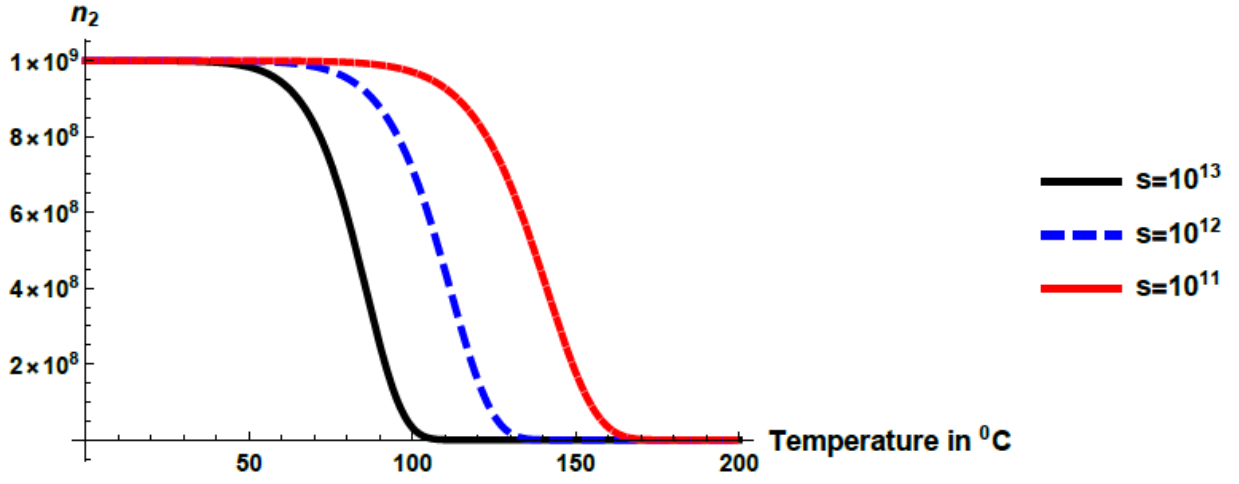

Fig. 3: Variation of concentration of electrons in the second active electron trap for various values of the frequency factor.

Figure 4 shows the variation of concentration of electrons in the thermally disconnected deep trap (TDDT) for various values of the frequency factor. The peaks shift to the low temperature region with increase in the frequency factor, indicating that, for a given temperature, the filling rate of the TDDT increases with increase in the value of s. At high temperature, all the peaks approach a constant value which is equal to the total concentration of traps in the TDDT.

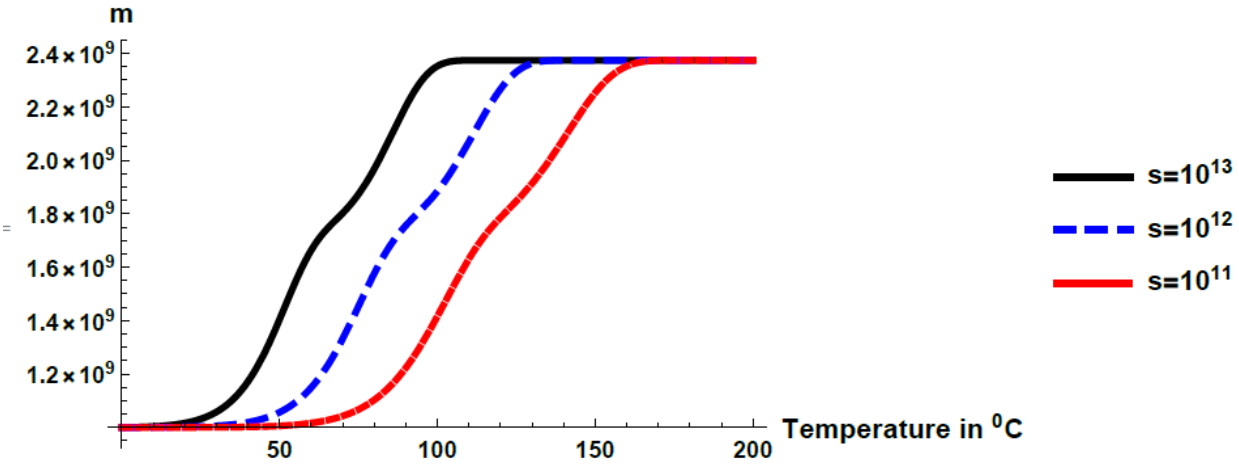

Fig. 4: Variation of concentration of electrons in the thermally disconnected deep trap (TDDT).

The variation of concentration of electrons in the conduction band is depicted in Fig. 5 for various values of the frequency factor. For each value of s, all the peaks have two maxima. The first maximum of each peak corresponds to the de-trapped electrons from the first trap and the second maximum corresponds to the de-trapped electrons from the second trap. Moreover, it can be seen that the concentration of electrons in the conduction band remains very small at all temperatures as compared to the initial concentration of electrons in traps which is of the order of 10^{10}. This is the reason c is sometimes taken as a constant in theoretical calculations.

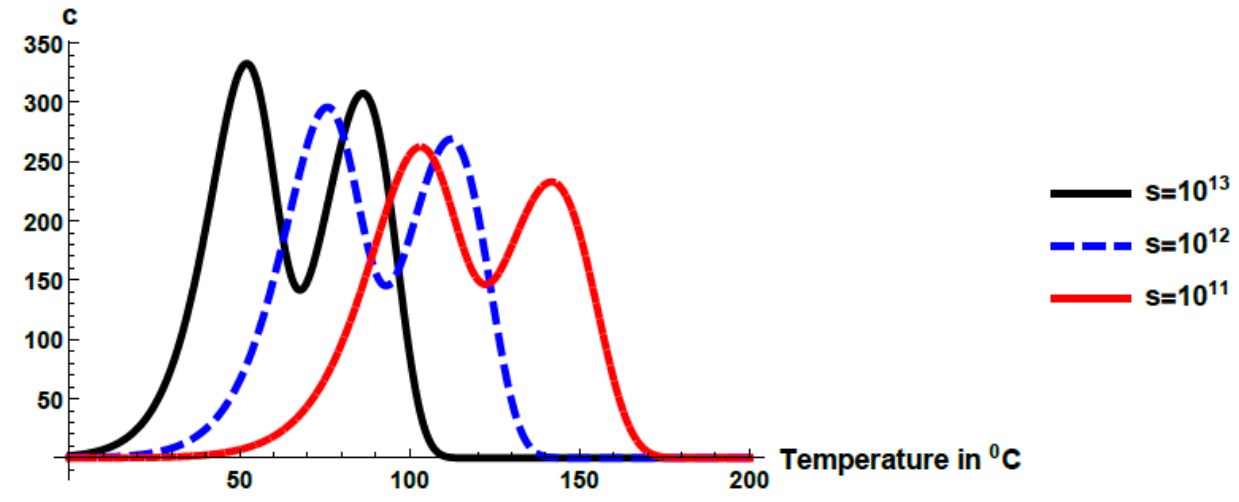

Fig. 5: Variation of concentration of electrons in the conduction for various values of the frequency factor.

The TL intensity is depicted in Fig. 6 for various values of the frequency factor. Again, for each value of s, all the peaks have two maxima. The first maximum of each peak is due to recombination of electrons de-trapped back to the conduction band from the first trap and the second maximum is due to recombination of electrons de-trapped back to the conduction band from the second trap. Comparing Figs. 5 and 6, it can also be seen that the TL intensity is directly proportional to the concentration of electrons in the conduction band. Moreover, with the increase in the frequency factor s, the TL glow curves shift to low temperature region.

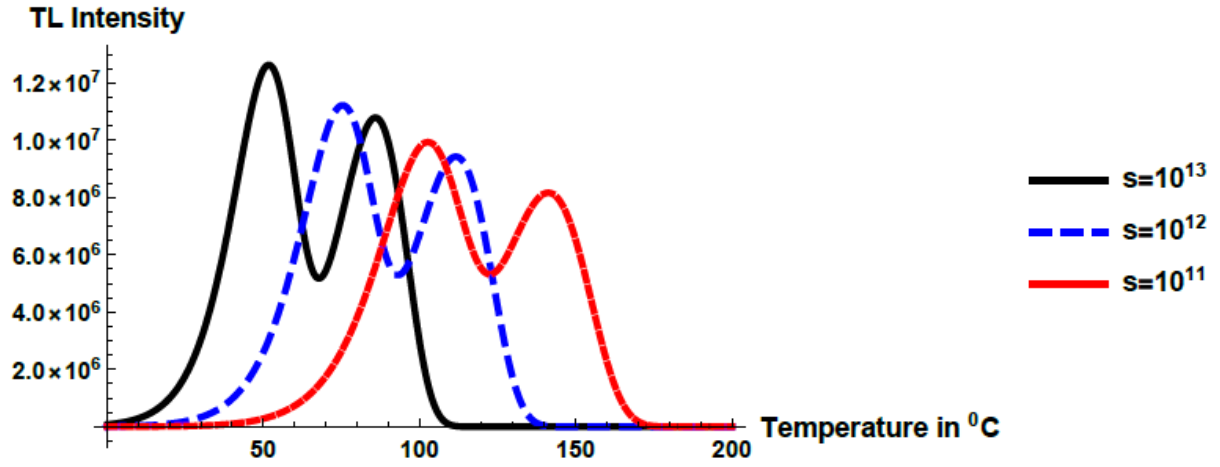

Fig. 6: The generated TL glow curves for various values of the frequency factor.

It is interesting to study the behavior of the TL glow curves and the variation of concentration of electrons in the active or thermally disconnected deep traps when the trap centers are closer to each other. For brief illustration, the variation of the concentration of electrons in the TDDT and the behavior of the TL glow curves are presented here for $E_1 = 0.95\ eV$ and $E_2 = 1\ eV$ (see Fig. 7 and Fig. 8). It is important to note that the graph profile of Fig. 7 (generated for $E_1 = 0.95\ eV$ and $E_2 = 1\ eV$) is slightly different from the graph profile in Fig. 4 (generated for $E_1 = 0.9\ eV$ and $E_2 = 1\ eV$). Figures 7 and 8 explain a scenario when E_1 and E_2 are closer to each other. In this scenario, the results are much similar to those generated for a single active electron trap taking part in the TL process. Though there are two active electron traps being involved in the TL process, only one broad glow curve is obtained (for each value of s) and the variation of concentration of electrons in the TDDT (for each value of s) looks like that obtained from a single active electron trap. Therefore, in experimental situations in particular, one glow peak doesn't necessarily mean the involvement of only one active electron trap. This is where the concept of glow curve deconvolution comes in. This means that a single broad glow curve is deconvoluted and the positions of the active defect states are determined. The process is usually assisted by Computerized Glow Curve Deconvolution software.

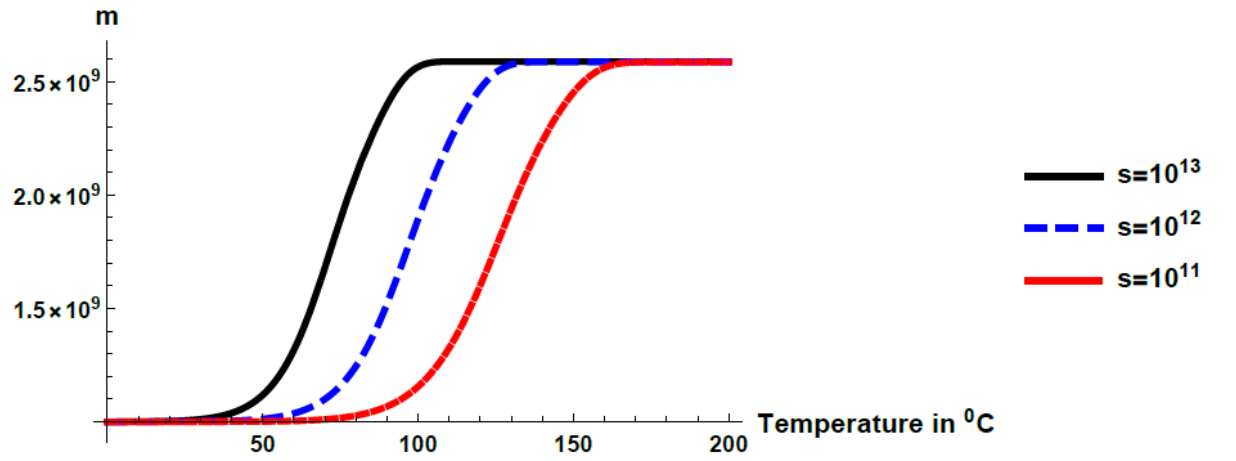

Fig. 7: Variation of concentration of electrons in the thermally disconnected deep trap (TDDT) for various values of the frequency factor when the active electron traps are closer to each other.

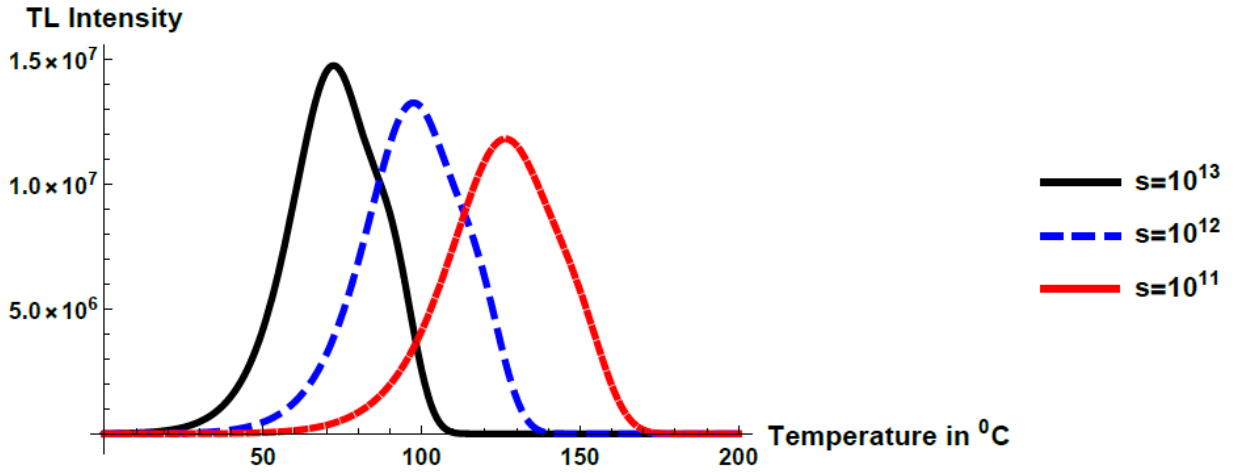

Fig. 8: The generated TL glow curves for various values of the frequency factor when the active electron traps are closer to each other.

It has been reported that the concentration of electrons in the active electron traps decreases with an increase in temperature; the concentration of electrons in the TDDT increases with an increase in temperature and one broad TL peak will be obtained when the active electron traps are very close to one another (Belayneh Mesfin and Teshome Senbeta, 2019; Sunta C., 2014; McKeever S., 1988; Pagonis V. et al., 2006; Pagonis V., et al., 2006). Therefore, the above results are highly similar to those reported in literature.

Two Active Electron Traps, Two TDDTs and One Recombination Center Model of TL

Generally, the above explanation is based on the involvement of two active electron traps and one TDDT in the TL process. However, one may ask the question: What if the number of TDDTs is more than one? Suppose there is one more TDDT in our problem. In this case, we investigate the model of TL in view of two active electron traps, two TDDTs, and one recombination center. The rate equations become (Pagonis V., et al., 2006; Asamin Regassa and Belayneh Mesfin,

2018; Mady F., et al., 2006; Kitis G. and Pagonis V., 2023):

$$\frac{dn_1}{dt} = -s_1 n_1 \exp\left\{-\frac{E_1}{kT}\right\} + cA_1(N_1 - n_1), \quad (8)$$

$$\frac{dn_2}{dt} = -s_2 n_2 \exp\left\{-\frac{E_2}{kT}\right\} + cA_2(N_2 - n_2), \quad (9)$$

$$\frac{dm_1}{dt} = cA_3(M_1 - m_1), \quad (10)$$

$$\frac{dm_2}{dt} = cA_4(M_2 - m_2), \quad (11)$$

$$\frac{dc}{dt} = s_1 n_1 \exp\left\{-\frac{E_1}{kT}\right\} + s_2 n_2 \exp\left\{-\frac{E_2}{kT}\right\} - c[A_1(N_1 - n_1) + A_2(N_2 - n_2) + A_3(M_1 - m_1) + A_4(M_2 - m_2) + A_r n_h]. \quad (12)$$

Here m_1 and m_2 are the concentration of trapped electrons (cm^{-3}) on the first and second TDDT; M_1 and M_2 are the total concentration of traps (cm^{-3}) on the first and second TDDT; A_3 and A_4 are the re-trapping probabilities of the electrons to the first and second TDDT $(cm^3 s^{-1})$, respectively.

All other parameters are as defined before. The system (8-12) is closed by the quasi-neutrality condition:

$$n_1(t) + n_2(t) + c(t) + m_1(t) + m_2(t) = n_h(t) \quad (13)$$

The generated results (using $M_1 = M_2 = 10^{10}, A_3 = 10^{-5}, A_4 = 10^{-7}, m_1(0) = m_2(0) = 10^9, \beta = 1 \text{degree/s}$) show that the variation of concentration of electrons in the active electron traps is unchanged (similar to Figs. 2 and 3). However, the temperature-dependent concentration of electrons in the conduction band and the TL intensity are slightly decreased as shown in Figs. 9 and 10. This is because of re-trapping of more electrons to the TDDTs.

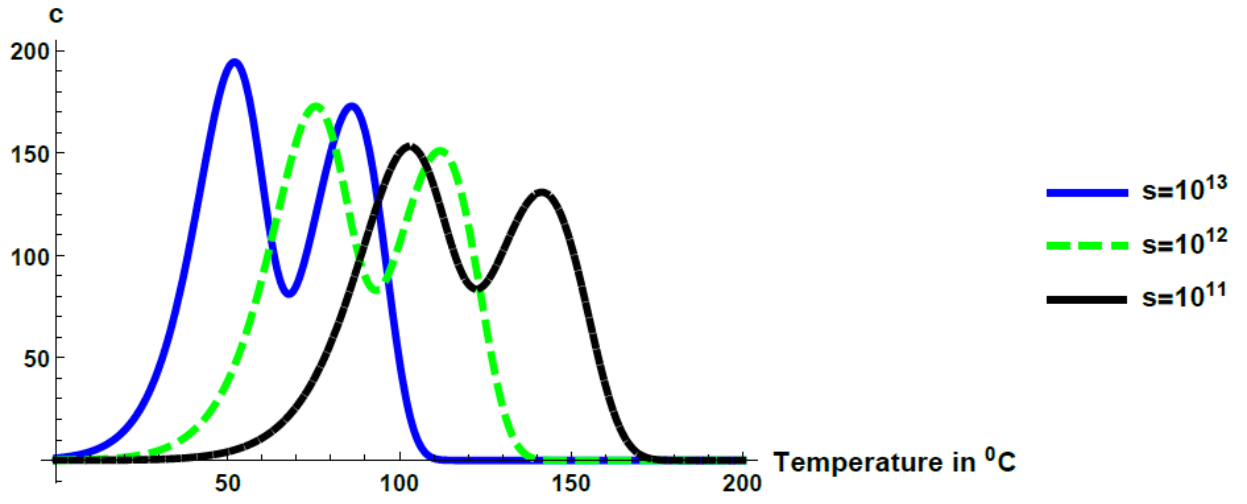

Fig. 9: Variation of concentration of electrons in the conduction band in the presence of two TDDTs.

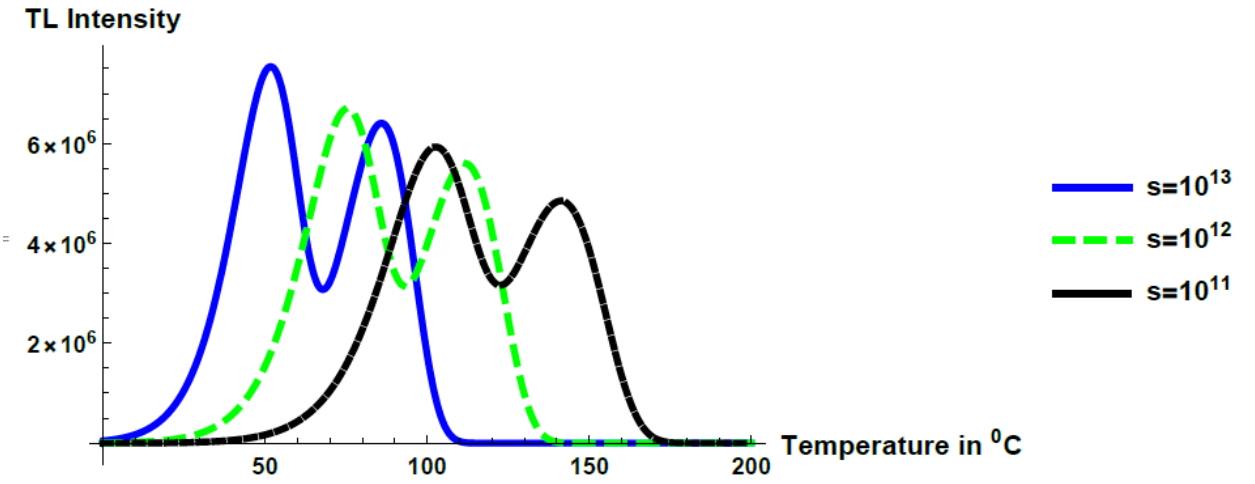

Fig. 10: The generated TL glow curves in the presence of two TDDTs.

Figures 11 and 12 depict the temperature-dependent variation of concentration of electrons in the first and second TDDT. Since A_3 and A_4 are taken to be different, the concentration of trapped electrons in the TDDTs are in general different.

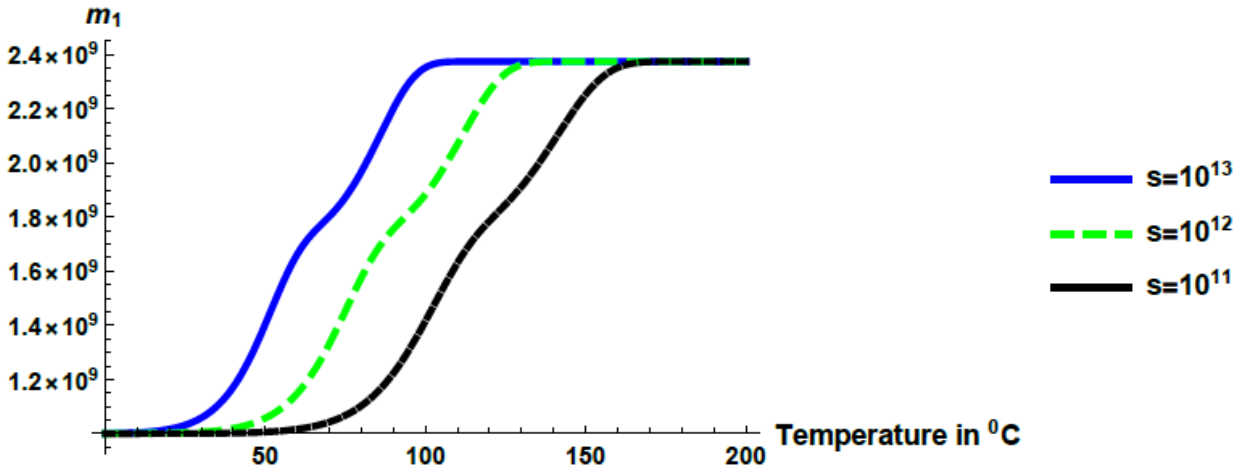

Fig. 11:Variation of concentration of electrons in the first thermally disconnected deep trap (TDDT).

The temperature dependent variation of concentration of electrons in the TDDTs depends on the re-trapping probability of the electrons to the first and second TDDT $(\mathrm{cm}^3\mathrm{s}^{-1})$. As discussed before, these re-trapping probabilities to the first and second TDDT are denoted by A_3 and A_4, respectively. The higher the re-trapping probability to the TDDT, the higher the concentration of trapped electrons in the TDDT. In the generation of Figs. 11 and 12, $\mathrm{A}_3 = 10^{-5}\ and\ A_4 = 10^{-7}$ were used. Since $A_3$and A_4 are taken to be different, the concentration of trapped electrons in the TDDTs are not the same. In particular, since, $\mathrm{A}_3 > \mathrm{A}_4$, the concentration of trapped electrons in the first TDDT (Fig. 11) is greater than that in the second TDDT (Fig. 12) as expected.

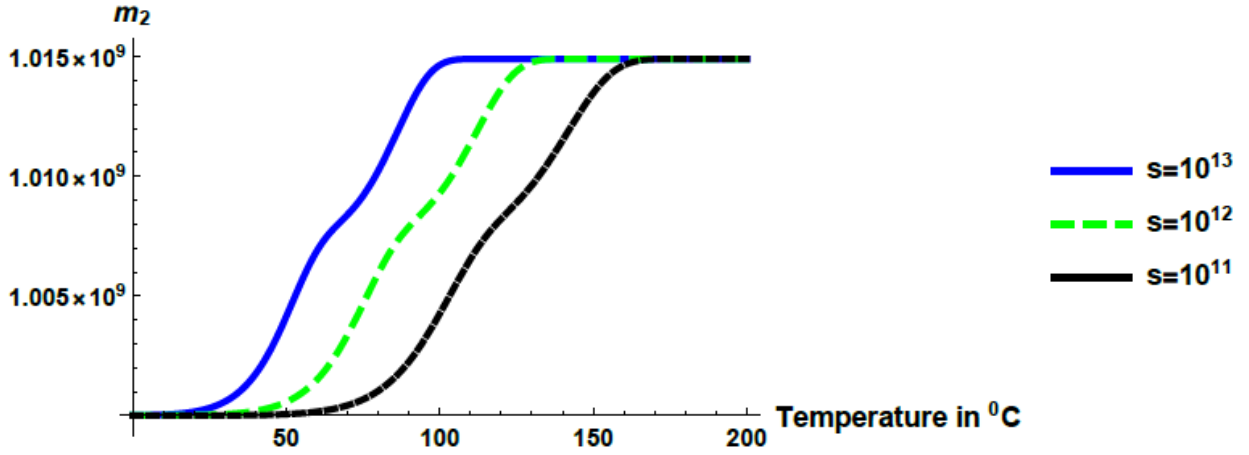

Fig. 12: Variation of concentration of electrons in the second thermally disconnected deep trap (TDDT).

The above results are also highly similar to those reported in literature (Belayneh Mesfin and Teshome Senbeta, 2019; Sunta C., 2014; McKeever S., 1988; Pagonis V. et al., 2006; Pagonis V., et al., 2006).

CONCLUSION

The rate equations corresponding to Thermoluminescence (TL) model consisting of two active electron traps, one thermally disconnected deep trap (TDDT), and one recombination center are clearly formulated and the results are discussed in detail. The presence of one and two thermally disconnected deep traps (TDDTs) is taken in account. The variation of concentration of electrons in conduction band can approximately be taken as a constant. Moreover, in experimental or theoretical results, one glow peak doesn't necessarily mean the involvement of only one active electron trap in the TL process. However, in such cases, the presence of more than one active defect center should be further investigated by using Computerized Glow Curve Deconvolution software. Moreover, the TL intensity is directly proportional to the concentration of electrons in the conduction band. The presence of multiple TDDTs does not affect the variation of concentration of electrons in the active electron traps. The theoretical investigation also fills the experimental gap where the concentration of the charge carriers in the active or thermally disconnected deep traps cannot be obtained.

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