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A model for explaining the concentration quenching of thermoluminescence

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ABSTRACT

The effect of concentration quenching (CQ) of luminescence has been reported in the literature. The luminescence efficiency dependence on the concentration of a specific impurity was found to reach a maximum intensity for a certain concentration, and decline at higher concentrations. A formula has been developed for the dependence of the efficiency on the concentration, assuming that only activators not adjacent to other activators can emit luminescence. Curve fitting of the CQ experimental curves to the theoretical function resulted in very large values of the parameter z, the number of nearest neighbors, of up to 4000, which is not feasible. A similar effect was found in TL of some materials, and the same formula for explaining the effect was used. Medlin has described the TL properties of calcite and dolomite. For a 300 K peak in Pb^{++} doped calcite, he used the same function and found $z = 700$, and for a 410 K peak, he got $z = 150$; the maxima occurred at different concentrations.

In the present work, we propose a possible, alternative model to explain the QC of thermoluminescence (TL). The model includes 3 trapping states and one recombination center (3T1C model). We assume that the 3 traps have a constant concentration, and the variable concentration is that of the recombination center M. An important assumption made is that the initial occupancy of M is not zero, and we assume that $m(0) = 0.1M$. The results yield the concentration dependence of the area under two simulated peaks reached by solving numerically the relevant set of six simultaneous rate equations. The maximum intensities of the two peaks occur at different concentrations, similarly to experimental results in Pb^{++} doped calcite and Mn^{++} doped dolomite. Approximate analytical derivations support these results.

1. Introduction

All luminescence effects including thermoluminescence (TL) and optically stimulated luminescence (OSL) are directly related to the occurrence of imperfections, namely impurities and defects in the host crystal. Normally, one would expect that when a certain imperfection is responsible for the appearance of TL or OSL, higher impurity levels would mean more emitted luminescence for a given excitation. However, several experiments have shown that a non-monotonic dependence on the impurity concentration may take place. The sensitivity to a given excitation irradiation may increase with the impurity concentration up to a maximum, and then decline for higher concentrations. This effect has been termed concentration quenching (CQ).

The effect of concentration quenching of luminescence has been first described by Johnson and Williams (1950a, 1950b). Experimental results of the dependence of the luminescence efficiency on the concentration of Mn in ZnF_2 showed an increase with the concentration up to a certain fraction of the activator, a maximum at this concentration and a decline of the efficiency at higher concentrations. These authors assume a random distribution of the activators (luminescence centers). They also assume that only activators not adjacent to other activators are capable of luminescence. They show that the efficiency of a phosphor can be quantitatively related to the activator concentration and to the capture cross sections for the exciting energy of luminescent and non-luminescent activators and the host lattice. With these assumptions, the authors developed the following equation for the efficiency η,

$$\eta = \frac{c(1-c)^z}{c + (\sigma/\sigma')(1-c)}, \tag{1}$$

where c is the total mole fraction of the activator and where σ and σ' are the capture cross sections of radiative and non-radiative centers and z the number of nearest cation neighbors. Equation (1) is

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a peak-shaped function which yields a maximum at a certain concentration, followed by a rather slow decrease as seen in several experimental works (Johnson and Williams, 1950a,b, 1953; Ewles and Lee, 1953; Schulman et al., 1957; Van Uitert, 1960, 1967, 1968; Van Uitert et al., 1962, 1967; Van Uitert and Johnson, 1966). While trying to fit this equation to experimental results, most authors considered z and $\gamma = \sigma/\sigma'$ as adjustable parameters. Johnson and Williams (1953) and (1950b) reported values of $z = 70$ for 305 nm fluorescent emission from KCl:Tl and $z = 4000$ for the fluorescent emission of ZnS:Cu. This value seems to be exceedingly large to represent the number of "nearest neighbors", and the authors suggest that in this case, the process involves transitions through the conduction band.

Ewles and Lee (1953) developed another formula, based on the same assumptions and taking into account the effect of absorption,

$$E = \frac{K}{1 + \alpha c^{-1} \exp(nc)}, \tag{2}$$

where E is the measured quantity, proportional to η, K is a normalization constant and $n = 1/c_{max}$ where c_{max} is the dopant concentration corresponding to η_{max}.

This non-monotonic effect has been reported in several materials in addition to ZnF$_2$:Mn mentioned above. Schulman et al. (1957) report on such a curve of luminescence efficiency with a maximum at ∼0.1 mol percent in KCl:Tl. Ewles and Lee (1953) describe this effect in yellow emission and UV emission in CaO:Bi and CaO:Pb. Van Uitert (1960) describes the effect in CaWO$_4$:Tb and in CaWO$_4$:Eu. In the latter, the maximum occurs at different concentrations for different luminescence emission wavelengths. More results of the non-monotonic effect associated with concentration quenching in various materials have been given in a series of papers by Van Uitert et al. (1962), Van Uitert (1967, 1968) and Van Uitert and Johnson (1966).

Similar effects of non-monotonic dependence of TL on the concentration of the relevant impurity in various materials have been reported by several authors. Medlin (1959) described the TL properties of calcite and reported on concentration quenching of the 350 K TL peak due to Mn^{++} in calcite. He used Formula (1), utilized before for fluorescence efficiency and by curve-fitting got values of $z = 75$ and $\gamma = 0.001$. For a 300 K peak in Pb^{++} doped calcite, he got $z = 700$ and $\gamma = 0.0001$ and for a peak at 410 K, he got $z = 150$ and $\gamma = 0.0001$. The former peak had a maximum at ∼0.0003 mol fraction of Pb^{++} and the latter at ∼0.001 mol fraction Pb^{++}. In another work, Medlin (1961) reported on CQ results in Mn^{++} doped Dolomite. The effect occurred for four peaks, at 330, 380, 500 and 600 K at different concentrations between 0.001 and 0.003 mol fraction of Mn^{++}. Rossiter et al. (1971) reported on concentration dependence of peak 5, at 210 °C, in LiF:Ti. They found a peak-shaped dependence on the Ti concentration with a maximum efficiency at ∼8 ppm Ti. Nambi et al. (1974) and Nambi (1979) described the concentration quenching effect of CaSO$_4$ with Dy and Tm impurities. In both cases, the maximum efficiency occurred at ∼0.1 weight percent of the dopant. The authors fitted their experimental results to the expression (2). Mulla and Pawar (1979) reported on the CQ effect in CaSO$_4$:Dy. Wachter (1982) studied the dependence of the sensitivity of LiF:Mg,Ti (TLD-100) on the ratio of the two dopants, Mg/Ti, and found a peak-shaped dependence with a maximum at a ratio of ∼0.32. Lai et al. (2006) investigated the TL of ZrO$_2$ doped with Yb$_2$O$_3$ and found concentration quenching with a maximum at 5 mol%, followed by a decrease at higher concentrations. Fitting the results to Eq. (1) yielded values of the adjustable parameters of $z = 1$ and $\gamma = 0.003$. Tajika and Hashimoto (2006) studied the blue TL in synthetic quartz with aluminum impurity. The characteristic concentration quenching behavior is observed with a maximum

blue TL intensity at ∼10 ppm of aluminum. Vij et al. (2009, 2010) reported on the TL of UV-irradiated Ce doped SrS nanostructures. The concentration quenching curve shows a maximum at a dopant concentration of ∼0.5 mol%. Sharma et al. (2009) describe similar results in the TL of CaS:Ce nanophosphors which show a maximum intensity of TL with ∼0.4 mol% of cerium.

In the present work, we propose a model which may explain the concentration quenching effect of two TL peaks in a given material, with the maximum sensitivity occurring at different concentrations of the relevant impurity.

2. The model

As pointed out above, the curve fitting of the experimental results of CQ, both of luminescence efficiency and TL intensity yield sometimes exceedingly large values of the parameter z which is supposed to be the number of nearest cation neighbors to a luminescence center. Furthermore, it is not clear that the theory developed for fluorescence necessarily applies for the CQ effect of thermoluminescence. In the present work, we propose an alternative model, based on the transitions of electrons and holes between trapping states, the conduction band and the valence band. This model is meant to demonstrate the possibility of explaining the CQ effect for two TL peaks, where the maximum intensity occurs at two different concentrations, in similarity to the experimental results by Medlin (1959) in Pb^{++} doped calcite. In Medlin's work, the maxima for four peaks in Mn^{++} doped dolomite occurred at different concentrations; one may expect that assuming more trapping states may yield the CQ effect for more than two peaks.

Fig. 1 presents the proposed energy level model which includes three electron trapping states, trap 1, trap 2 and trap 3, with concentrations of N_1, N_2 and N_3 and instantaneous occupancies of n_1, n_2 and n_3, respectively. The activation energies are E_1, E_2 and E_3 and the frequency factors are s_1, s_2 and s_3, respectively and the retrapping-probability coefficients are A_1, A_2 and A_3. Also is shown the recombination center with concentration M and instantaneous occupancy m of holes. The probability coefficient for holes to be trapped in the center is B and the electron recombination probability coefficient is A_m. n_c and n_v denote the concentrations of free electrons and holes, respectively. X denotes the rate of production of electron–hole pairs by the excitation dose, which is proportional to the dose rate. The concentration which is varied between different samples is assumed to be that of the recombination center, M, whereas the other concentrations of the traps are assumed to remain constant. Also, the dose of excitation is kept the same when simulating the excitation of the differently doped samples.

The set of simultaneous differential equations governing the process during excitation is,

$$\frac{dn_1}{dt} = A_1(N_1 - n_1)n_c, \tag{3}$$

$$\frac{dn_2}{dt} = A_2(N_2 - n_2)n_c, \tag{4}$$

$$\frac{dn_3}{dt} = A_3(N_3 - n_3)n_c, \tag{5}$$

$$\frac{dm}{dt} = B(M - m)n_v - A_m m n_c, \tag{6}$$

$$\frac{dn_v}{dt} = X - B(M - m)n_v, \tag{7}$$

$$\frac{dn_c}{dt} = \frac{dm}{dt} + \frac{dn_v}{dt} - \frac{dn_1}{dt} - \frac{dn_2}{dt} - \frac{dn_3}{dt}. \tag{8}$$
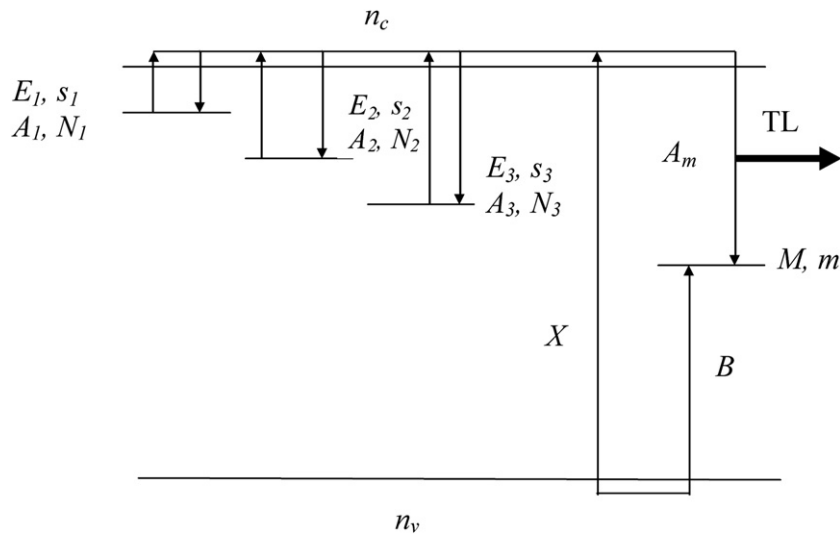
Fig. 1. Energy level diagram of the three-trap one recombination center model.

In order to follow the experimental procedure, one has to solve this set of equations for a certain period of time t, and thus, the dose applied is proportional to X_t. The next stage of the simulation is relaxation, an additional period of time when the excitation is switched off, in which the free electrons and holes remaining in the conduction and valence bands, respectively, decay into the respective trapping states, thus contributing to the final concentrations. Next, a third stage of TL read-out starts in which the sample is heated up using a given heating function $T = T(t)$, and the emitted light denoted in Fig. 1 by a thick arrow is recorded as a function of temperature. The governing equations for this stage are,

$$\frac{dn_1}{dt} = A_1(N_1 - n_1)n_c - s_1 n_1 \exp(-E_1/kT), \tag{9}$$

$$\frac{dn_2}{dt} = A_2(N_2 - n_2)n_c - s_2 n_2 \exp(-E_2/kT), \tag{10}$$

$$\frac{dn_3}{dt} = A_3(N_3 - n_3)n_c - s_3 n_3 \exp(-E_3/kT), \tag{11}$$

$$\frac{dm}{dt} = -A_m m n_c, \tag{12}$$

$$\frac{dn_c}{dt} = \frac{dm}{dt} - \frac{dn_1}{dt} - \frac{dn_2}{dt} - \frac{dn_3}{dt}. \tag{13}$$

As seen in Fig. 1, the TL emission is associated with the recombination of free electrons with holes in centers, thus, when the heating function $T(t)$ is known, the TL intensity can be written as,

$$I(T) = -\frac{dm}{dt} = A_m m n_c. \tag{14}$$

3. Numerical results

An example of the numerical results reached in the simulation through the three mentioned stages is shown in Fig. 2. The parameters used are given in the caption. It should be noted that if we start the excitation stage with empty centers, the non-monotonic concentration quenching effect is not seen. Instead, we assume that the initial occupancy is a given fraction of the

concentration of the center, and we choose $m(t = 0) = m_0 = 0.1M$. When the heating stage is simulated, the area under each of the two peaks is recorded and then plotted against the varying concentration of the centers M. The results show that the two simulated glow peaks areas are non-monotonic with the center concentration M, and that the maxima occur at different concentrations, which, as pointed out above, resembles the experimental results by Medlin (1959, 1961). In another simulation, the details of which are not given here, we found that the CQ effect for a single TL peak, an effect found in several materials as described above, can be simulated by a simpler model of two traps and one recombination center (2T1C).

4. Analytical considerations

We would like to present here an approximate analytical way of demonstrating the CQ effect within the models described here. Let us start with the 2T1C model. We consider N_1 and N_2 to be fixed and assume that the recombination center M varies. For the initial conditions we assume $n_1(0) = n_2(0) = 0$. As explained above, we assert that $m(0) = \alpha M$. Let us assume that excitation dose used to monitor the TL sensitivity is relatively low so that the trapping
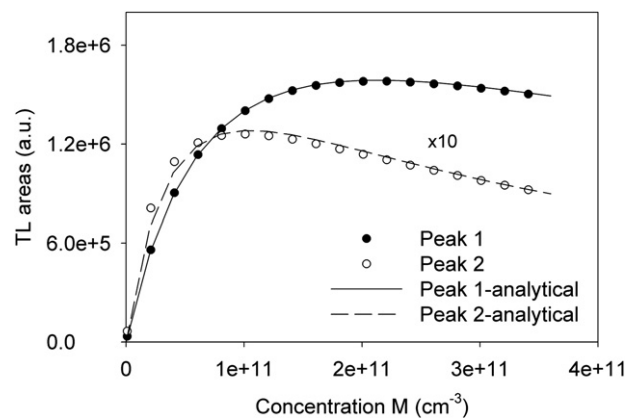

Fig. 2. Simulated results of the concentration dependence of the areas under the two peaks, at ∼91^{0}C and ∼220^{0}C with: $N_1 = 3 \times 10^8$ cm^{-3}; $N_2 = 10^7$ cm^{-3}; $N_3 = 10^9$ cm^{-3}; $A_1 = 10^{-8}$ cm^3 s^{-1}; $A_2 = 10^{-8}$ cm^3 s^{-1}; $A_3 = 10^{-9}$ cm^3 s^{-1}; $B = 10^{-9}$ cm^3 s^{-1}; $E_1 = 0.97$ eV; $E_2 = 1.55$ eV; $E_3 = 1.8$ eV; $D = 5 \times 10^6$ cm^{-3}; $s_1 = 5 \times 10^{12}$ s^{-1}; $s_2 = 5 \times 10^{12}$ s^{-1}; $s_3 = 5 \times 10^{13}$ s^{-1}.

states are far from saturation, $n_1 \ll N_1$ and $n_2 \ll N_2$ and also, n_1 and n_2 are much smaller than αM. Let us also make the quasi-steady approximation for free electrons, namely, $n_c \ll n_1$, n_2 and $|dn_c/dt| \ll |dn_1/dt|$, $|dn_2/dt|$. The population of the active trap, n_1, after irradiation by dose $D = X_t$ is

$$n_1 = \frac{A_1 N_1}{A_1 N_1 + A_2 N_2 + A_m \alpha M} D. \tag{15}$$

This equation states that the concentration of this trap after irradiation is approximately the number of electron–hole pairs created times the fraction of electrons which go into the first trap as opposed to going into the second trap or recombining at the center. Going to the next stage, there is competition during read-out. The integrated TL intensity is determined approximately by the fraction of trapped electrons n_1 which radiatively recombine as opposed to getting caught by the deep trap,

$$I_{TL,int} = \frac{A_1 N_1}{A_1 N_1 + A_2 N_2 + A_m \alpha M} \times \frac{A_m \alpha M}{A_2 N_2 + A_m \alpha M} D. \tag{16}$$

Notice that the numerator scales with M. Consequently, for small M, namely for $A_m \alpha M \ll A_2 N_2$, the TL intensity grows linearly with M. For large M, namely, $A_m \alpha M \gg A_1 N_1 + A_2 N_2$, the denominator grows as the square of M and consequently, the TL intensity declines like $1/M$. Thus, the integral intensity initially grows as M increases, then it levels off, and then declines with further increases in M, in a similar manner to any of the graphs in Fig. 2.

Let us extend the approximate derivation to the 3T1C case. In full analogy with the previous case and under the same assumptions, we get for the concentrations of trapped electrons at the end of excitation,

$$n_1 = \frac{A_1 N_1}{A_1 N_1 + A_2 N_2 + A_3 N_3 + A_m \alpha M} D, \tag{17}$$

$$n_2 = \frac{A_2 N_2}{A_1 N_1 + A_2 N_2 + A_3 N_3 + A_m \alpha M} D. \tag{18}$$

For the read-out stage, let us begin with the first TL peak. The electrons from the first trap are excited into the conduction band and subsequently trapped by N_2, N_3 or M. The fraction which is trapped by M and consequently cause TL emission is $A_m \alpha M/(A_2 N_2 + A_3 N_3 + A_m \alpha M)$. Thus, the integrated intensity for the first peak is approximately,

$$
\begin{aligned}
I_{1,int} &= \frac{A_m \alpha M}{A_2 N_2 + A_3 N_3 + A_m \alpha M} n_1 \\
&= \frac{A_m \alpha M}{A_2 N_2 + A_3 N_3 + A_m \alpha M} \\
&\quad \times \frac{A_1 N_1}{A_1 N_1 + A_2 N_2 + A_3 N_3 + A_m \alpha M} D.
\end{aligned} \tag{19}
$$

Equation (19) is similar in form to (16), and therefore, the dependence on the concentration M is similar, namely, it has a peak shape like each of the graphs in Fig. 2. Similarly to the 2T1C case, $I_{1,int}$ rises linearly with M as long as M is small enough which in this case means,

$$A_m \alpha M \ll A_2 N_2 + A_3 N_3. \tag{20}$$

$I_{1,int}$ drops with the inverse of M when M is large enough, i.e.,

$$A_m \alpha M \gg A_1 N_1 + A_2 N_2 + A_3 N_3. \tag{21}$$

During the thermal excitation of n_1, some of the freed electrons become trapped by N_2. This change in n_2, which will be called Δn_2,

is the product of (a) the number of electrons released, n_1, and (b) the fraction of those electrons which are captured by N_2,

$$
\begin{aligned}
\Delta n_2 &= \frac{A_2 N_2}{A_2 N_2 + A_3 N_3 + A_m \alpha M} n_1 \\
&= \frac{A_2 N_2}{A_2 N_2 + A_3 N_3 + A_m \alpha M} \\
&\quad \times \frac{A_1 N_1}{A_1 N_1 + A_2 N_2 + A_3 N_3 + A_m \alpha M} D,
\end{aligned} \tag{22}
$$

where Eq. (17) was used. The total concentration, n_2', after thermal excitation of n_1, is the sum of Eqs. (18) and (22). This yields immediately,

$$
\begin{aligned}
n_2' &= \frac{A_2 N_2}{A_1 N_1 + A_2 N_2 + A_3 N_3 + A_m \alpha M} \\
&\quad \times \left(1 + \frac{A_1 N_1}{A_2 N_2 + A_3 N_3 + A_m \alpha M}\right) D = \frac{A_2 N_2}{A_2 N_2 + A_3 N_3 + A_m \alpha M} D.
\end{aligned}
$$

The concentrations n_3 and m both also change during this step, but under the low-dose assumption, we have no need to know the amount.

As temperature continues to rise during the read-out, the electrons are freed from trap 2. The total quantity of these electrons is given by Eq. (23). These electrons are ultimately captured by either N_3 or M. The integrated intensity of the second peak is proportional to those trapped by M, namely,

$$
\begin{aligned}
I_{2,int} &= \frac{A_m \alpha M}{A_3 N_3 + A_m \alpha M} n_2' \\
&= \frac{A_m \alpha M}{A_3 N_3 + A_m \alpha M} \times \frac{A_2 N_2}{A_2 N_2 + A_3 N_3 + A_m \alpha M} D.
\end{aligned} \tag{24}
$$

Note that Eq. (24) is equivalent in form to Eqs. (16) and (19). $I_{2,int}$ rises linearly with M as long as M is small enough that,

$$A_m \alpha M \ll A_3 N_3. \tag{25}$$

$I_{2,int}$ drops with the inverse of M when M is large enough that,

$$A_m \alpha M \gg A_2 N_2 + A_3 N_3. \tag{26}$$

Comparing the relation (20) with (25) and (21) with (26) indicates that $I_{1,int}$ and $I_{2,int}$ will peak at different values of M. As M increases, the second peak reaches its maximum before the first one. How far before is controlled by the sizes of $A_1 N_1$ and $A_2 N_2$.

One should note that the underlying reason for the non-monotonic dependence of the TL sensitivity on the concentration, as demonstrated in the simulation and Eqs. (19) and (24) is the competition in trapping, both during excitation and read-out, between trap 1, trap 2 and trap 3.

5. Discussion

In the present work, we propose a simple energy-band model which may explain the concentration quenching effect of TL for two glow peaks. We have used a model of three electron trapping states and one kind of recombination center (3T1C). We have shown that performing a simulation based on solving sequentially the relevant sets of coupled differential equations in the three stages of excitation, relaxation and heating read-out yielded the concentration quenching effect while changing the center concentration. The two simulated TL peaks reached their maxima at two different concentrations, in similarity to the experimental results by Medlin (1959, 1961). It is also reported that a simpler model of two trapping states and one kind of recombination center (2T1C) can explain the CQ effect for one TL peak. In future work,

one may check the assertion that the concentration quenching effect of 4 TL peaks, occurring at different concentrations, as described by Medlin (1961) for Mn^{++} doped dolomite, can be simulated by a 5T1C model. It should be noted that the assumption made here concerning the initial occupancy of the center not being zero has been made previously in relation to thermoluminescence and optically stimulated luminescence (OSL) by Chen and Leung (2001), Yukihara et al. (2004) and Pagonis et al. (2009). For example, as pointed out before (see e.g. Carter, 1970), if the energy of the center is near the Fermi level, the center will be partially filled with electrons (and partially with holes), so that if we write $m_0 = \alpha M$, α has some value determined by Fermi statistics. α can be assumed to be constant if the total center concentration M changes. The results do not seem to depend significantly on the strict constancy of α. The location of the Fermi level may change slightly with M. In order to check the sensitivity of the results to the values of α, we have run the simulation with a constant initial occupancy, $m_0 = 10^{11}$ cm^{-3}. The results were not the same as before, but the non-monotonic CQ effect was still clearly seen for the two simulated TL peaks with the maxima occurring at different concentrations. The main point in this respect remains that the initial occupancy of M should not be zero.

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