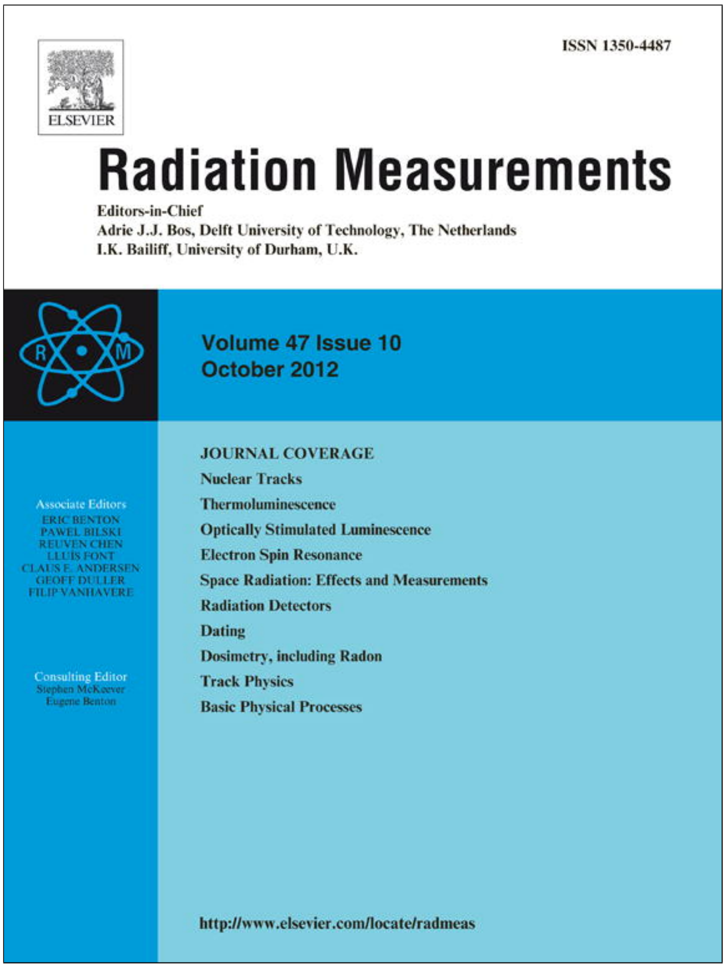

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Modelling the optical bleaching of the thermoluminescence of K_2YF_5:Pr^{3+}

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HIGHLIGHTS

- ► The optical bleaching of the thermoluminescence of K2YF5:Pr^{3+} has been studied.
- ► A model accounting for the optical bleaching has been put forward.
- ► Thermoluminescence occurs via delocalized transitions.
- ► Localized transitions occur during optical stimulation.

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ABSTRACT

Optical bleaching of the thermoluminescence (TL) curve of K_2YF_5:Pr^{3+} has been observed after optically stimulated luminescence (OSL) readout of pre-irradiated crystals. The traps being responsible for the TL signal are not emptied completely by the optical stimulation. Furthermore, if the illumination time is increased a constant intensity level of the residual TL glow curve is eventually achieved. On the other hand, if the low temperature peak of the glow curve is thermally cleaned, no subsequent OSL is measured. This behavior has been successfully explained by assuming that part of the electrons in the trap being responsible for the low temperature glow peak of K_2YF_5:Pr^{3+} recombine with holes via localized transitions during optical stimulation. During TL all trapped electrons recombine via delocalized transitions. Simulations have been carried out in order to demonstrate the feasibility of the model.

1. Introduction

Thermoluminescence (TL) of dosimetric materials is a complex process involving several stages of energy and charge transfer between the different defects of the material. The TL experiment has as one of its prime objectives to extract data from an experimental glow curve and to use them to obtain information about the parameters associated with the charge transfer process. These parameters (trap depths, frequency factors, capture cross-sections and densities of traps and recombination centers (RC)) reveal information about the processes of retrapping and recombination of charge carriers in the material under study. The knowledge of the values of these parameters is an important step to arrive at an acceptable level of understanding of the radiation-induced physical processes taking place in the material (McKeever, 1985).

Optically stimulated luminescence (OSL), like TL, involves several stages of charge transfer between different defects, but not always the traps responsible for TL are the same involved in OSL. In many materials the TL intensity decreases after optical stimulation, which does not necessarily mean that the traps responsible for TL are emptying during light stimulation. This effect, dubbed optical bleaching, has been observed during TL/OSL experiments with geological samples of quartz and feldspar and other dosimetric phosphors (Chruścińska, 2006; Chen and Pagonis, 2011). In order to explain this kind of behavior Chen et al. (1990) considered a one-trap/one-RC model including electron retrapping in the OSL trap and the possibility that electrons from the RC can also be excited into the conduction band during photo-stimulation. According to this model a constant residual TL curve is observed once equilibrium between the excitation rates of electrons from RC and traps has been achieved. This residue does not depend on the initial trap occupation. Besides, illumination could produce a TL signal in

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a previously unirradiated sample (McKeever, 2001). McKeever (1991) and Chruścińska (2006) put forward a model, which considers that the traps involved in OSL are thermally disconnected and TL traps are not directly emptied during optical stimulation. Besides, two kinds of RC are considered, namely, radiative and non-radiative hole trap centers. In this case the optical bleaching is the result of hole depopulation of RC during optical stimulation. This model predicts that the residual TL yield depends on the dose given for trap filling. In both Chen and McKeever/Chruścińska's models, the residual TL measured after the OSL readout reaches a stable behavior, and cannot be bleached by any longer optical stimulation. It is important to emphasize that both models rely on the assumption that traps can interact and consider only transitions via delocalized bands.

In the last years, several articles have been published demonstrating the feasibility of using hydrothermally grown rare-earth doped yttrium fluorides as radiation detectors (Faria et al., 2004; Silva et al., 2007; Marcazzó et al., 2011; Molina et al., 2011). In particular the efficient TL response of $K_2YF_5:Pr^{3+}$ has been reported (Marcazzó et al., 2004). The TL yield of $K_2YF_5:Pr^{3+}$ 0.5 at.% under ^{60}Co gamma irradiation is three times higher than that of commercial TLD-700. In order to understand the underlying physical process giving rise to TL in this compound its glow curve was analyzed (Marcazzó et al., 2008). It was found that the behavior of the glow peaks in samples irradiated under different dose levels can be correctly described only if interaction among traps is considered (Marcazzó et al., 2008, 2007).

Recently, the OSL properties of $K_2YF_5:Pr^{3+}$ has been also reported (Marcazzó et al., 2009). In particular, the OSL intensity of $K_2YF_5:Pr^{3+}$ 0.5 at.% under ^{90}Sr beta irradiation is of the same order of magnitude as that of commercial Al_2O_3:C, which makes this compound very interesting from the point of view of its application in dosimetry. In the same work the effects of optical bleaching are observed in this compound. The optical stimulation during OSL readout reduces the intensity of both of the glow peaks of the residual glow curve. However, their intensities reach a stable behavior when the duration of the optical stimulation in the particular experimental conditions is longer than 125 s.

In this paper we have addressed the detailed study of the OSL/TL correlation in order to develop a model describing the traffic of charge carriers between traps and RC during irradiation and optical/thermal stimulation. The model considers only one type of RC and two electron traps corresponding to each of the observed glow peaks of $K_2YF_5:Pr^{3+}$. Interaction among traps via delocalized transitions during TL readout have been assumed. However, localized transitions have been addressed to be responsible for the OSL signal. The model correctly describes not only the bleaching effect observed in $K_2YF_5:Pr^{3+}$, but also other observed experimental findings.

2. Samples and experimental methods

Crystals of K_2YF_5 doped with 0.5 at.% Pr^{3+} grown under hydrothermal conditions (Dubinskii et al., 1990) were used in this work. A complete description of the synthesis, structure type, stoichiometry and phase purity of synthesized samples can be found in (Marcazzó et al., 2009).

Irradiation of the samples was carried out by placing them 1 cm away from a 3.7 × 108 Bq ophthalmic ^{90}Sr beta-source rendering a dose rate equivalent to 0.024 Gy min^{-1} in tissue at the sample location. In all cases the TL and OSL measurements were performed after 24 h of storage of the irradiated sample in dark at room temperature (RT) in order to allow depletion of shallow traps.

TL glow curves from the samples were recorded from RT up to 650 K with a constant heating rate of 1.0 K s^{-1} by using a Harshaw-Bicron 3500 TL reader featuring a Hamamatsu R6094 photomultiplier tube.

The experimental setup employed for the OSL experiments has been described elsewhere (Marcazzó et al., 2009). In particular, optical stimulation of irradiated samples was achieved by means of a Luxeon V Star green LED (maximum emission at 530 nm). In all of the OSL experiments, the LED was driven at 500 mA yielding an effective luminous flux of 128 lm at the sample position. The light emitted by the LED was filtered before reaching the sample by means of two 3 mm thick Schott OG530 long-pass filters. The OG530 filter features a maximum transmission of 91% at wavelengths higher than the cutoff wavelength (530 nm) and lower than 10^{-6} at shorter wavelengths.

In order to get rid of the stimulation light, two 3 mm thick Hoya B-390 band-pass filters were interposed between the sample and the light detector. The B-390 filter has non-zero transmission between 320 and 500 nm and maximum transmission (77%) at 400 nm. Optically stimulated luminescence was measured by means of the aforementioned P25PC-02 photon counting head. In all measurements both irradiation and optical stimulation were applied to the same face of the sample from which the emitted light was detected. All the OSL measurements were performed at RT. In the case of correlated measurements the delay between the OSL readout and the recording of the residual TL was never longer than 1 min.

3. Experimental

3.1. Results

In Fig. 1 the OSL curves corresponding to a crystal irradiated with 2.40 and 0.24 Gy are shown. The signal background has also been included, which is made up of the dark current of the light detector plus the small amount of stimulation light passing through the set of Hoya B-390 band-pass filters. The latter contribution starts when the stimulation green LED is switched on 10 s after the beginning of the measurement. The OSL signal rises quickly up to a maximum value from which decay is observed. It is apparent from the figure that the OSL curve cannot be fitted by means of a single exponential component. It can be also seen in the figure that in the case of the crystal irradiated with 0.24 Gy the OSL intensity almost reaches the background level (implying net OSL signal equal to zero) if the optical stimulation is longer than 125 s, as reported by Marcazzó et al. (2009).

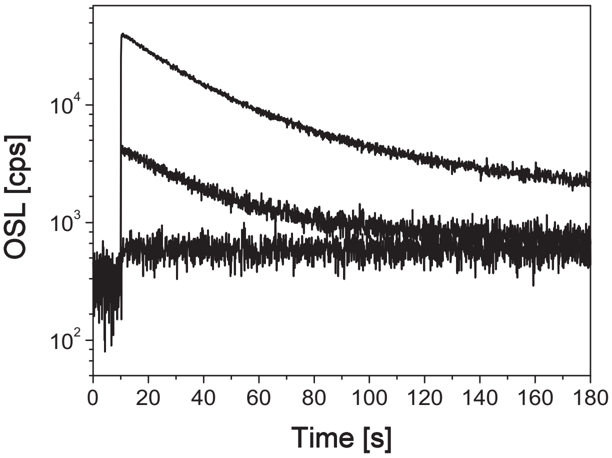

Fig. 1. OSL signal of a $K_2YF_5:Pr^{3+}$ crystal irradiated with 2.40 and 0.24 Gy (upper and intermediate curves, respectively). The bottom curve corresponds to the background as explained in the text.

In Fig. 2(a) the glow curve of a $K_2YF_5:Pr^{3+}$ crystal irradiated with 0.24 Gy is shown (solid). As reported by Marcazzó et al. (2004) two peaks are observed at 400 and 495 K that will be dubbed peak#1 and #2 respectively. Both peaks feature a rather symmetric shape, which in principle could be regarded as an indication of charge retrapping during thermal stimulation (McKeever, 1985). An irradiated crystal (0.24 Gy) was annealed at 420 K during 10 min and the residual glow has been immediately recorded (Fig. 2(a), dashed). As expected, the annealing completely depletes the traps responsible for peak #1. No appreciable effects are observed on the height of peak #2. On the other hand, if the OSL experiment is performed immediately after the thermal cleaning at 420 K, no OSL emission is observed. Further, if the residual glow curve is subsequently recorded, the same signal shown in Fig. 2(a) (dashed) is obtained.

As to the effect of the optical stimulation on the residual glow curve, say, the TL glow curve recorded after the OSL readout, Marcazzó et al. (2009) reported that the intensity of the residual TL decreases monotonically as the duration of the optical stimulation increases. Eventually the residual TL reaches a stationary, no null intensity level once the OSL signal previously measured has decreased to zero. In the case of a $K_2YF_5:Pr^{3+}$ crystal irradiated with 0.24 Gy the effect of the optical bleaching reaches a stationary level for stimulation times longer than 125 s. Of course, this characteristic time depends on the dose given to the crystal and the absolute intensity of the stimulation light in the particular experimental setup. It is also worth mentioning that no TL is observed after optically stimulating an unirradiated sample.

In Fig. 2(b) the glow curve recorded after irradiation (solid) is compared with the residual TL measured after 180 s of optical stimulation (dotted). The sample received a dose of 0.24 Gy. Like the original glow curve the residual glow curve is made up of two peaks having maxima at 400 and 495 K respectively. It is apparent from the figure that the OSL measurement have effect on both peaks of the glow curve and not only on peak #1, as could be expected by taking into account the result shown in Fig. 2(a). However, the bleaching effect is stronger for peak #1. In fact the intensity of the low temperature peak is reduced by 60% whereas in the case of peak #2 the decrease of its intensity is of 30%. Summarizing, the structure of the residual glow curve is similar to that of the original glow curve. In particular, none of the peaks disappears and no new peaks appear in the residual glow curve.

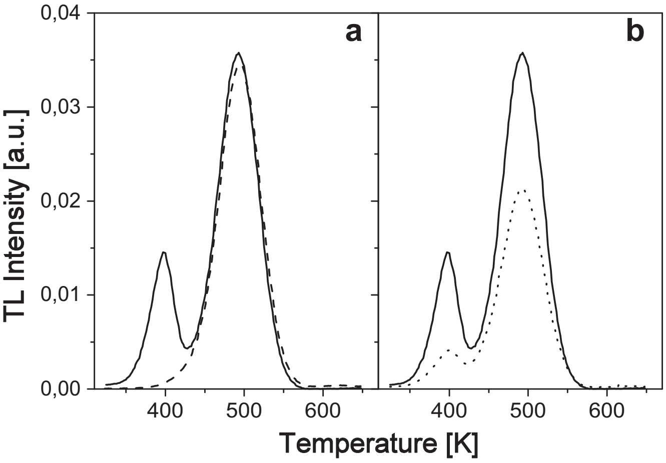

Fig. 2. Glow curve of a $K_2YF_5:Pr^{3+}$ crystal irradiated with 0.24 Gy (solid curve in both subfigures). This curve is compared with a) the residual TL (dash) measured after thermally cleaning the low temperature peak (annealing at 420 K) and performing an OSL reading, and b) the residual TL measured after 180 s of optical bleaching (dot).

It has been observed that if a $K_2YF_5:Pr^{3+}$ crystal is irradiated successively with the same dose (0.24 Gy), and, in each cycle we measure the OSL signal (stimulation time 180 s), the OSL curves obtained are exactly the same. This result is observed regardless whether the residual TL is measured or not in each cycle. Moreover, if at the end of several irradiation/OSL readout cycles the residual TL is measured, its area is just the product of the number of cycles and the area of the residual TL as measured after a single irradiation/OSL cycle. In other words, the area of the residual TL is proportional to dose.

3.2. *Discussion of experimental results*

A detailed analysis of the OSL signal of $K_2YF_5:Pr^{3+}$ can be found in (Marcazzó et al., 2008), where the authors show that at least two exponential components are necessary to fit the OSL curve of this compound. In principle this result could be taken as an evidence that at least two first order kinetics traps are emptied during optical stimulation (Chen and Pagonis, 2011). However, as shown by McKeever (2001), a single trap could render a non single-exponential OSL curve if retrapping is present during optical stimulation. For the sake of simplicity, we will assume that only one kind of trap is responsible for OSL. This trap will be named OSL trap in what follows.

As to the emission spectrum Marcazzó et al. (2004, 2009) reported that not only TL but also OSL spectra can be assigned to characteristic Pr^{3+} emission in K_2YF_5. In this context, trivalent Pr cations could play the role of RC during the TL/OSL process in this compound. This behavior is not unexpected if we consider that Pr^{3+} like other lanthanides (Ce^{3+} and Tb^{3+}) can trap holes during irradiation to form Pr^{4+} in wide bandgap isolators (Dorenbos and Bos, 2008). During either thermal or optical stimulation electrons released from electron traps recombine with Pr^{4+} leading to Pr^{3+} characteristic emission (Dorenbos et al., 2011).

As mentioned in the previous subsection two peaks are observed in the glow curve of $K_2YF_5:Pr^{3+}$ (see Fig. 2). This result could be regarded as an indication that at least two kinds of traps are responsible for TL in the temperature range of the TL experiment. As previously mentioned, their symmetric shape is incompatible with a first order assumption for their kinectics, say, interactions between both types of traps should be expected. These traps, which will be named TL trap #1 and #2 in what follows, will be regarded as electron traps.

The OSL trap is clearly not thermally disconnected, since OSL can be erased by annealing an irradiated sample at 420 K. This fact precludes the direct use of the models by McKeever (1991) and Chruścińska (2006) for describing the TL/OSL process in this compound, since these models assume that the OSL trap is thermally disconnected. On the other hand, since no TL is observed after illuminating an unirradiated sample, the model by Chen et al. (1990) is also not valid in this context.

If the OSL trap is involved in TL it should contribute to develope a TL peak in the same temperature range where peak #1 is observed. However, no changes in the shape of peak #1 are observed in the residual glow curve (see Fig. 2(b)). In fact, only the intensity of peak #1 is affected by previous illumination. This experimental finding suggests that the OSL trap and the TL trap #1 are actually of the same type, at least from the point of view of their contribution to the TL signal.

At this stage it is possible to put forward a model explaining the optical bleaching of $K_2YF_5:Pr^{3+}$ TL if we assume that a) as usual electrons from both trap #1 and #2 recombine via the conduction band (CB) during thermal stimulation, but b) during optical stimulation only part of the electrons in trap #1 recombine with holes in the RC via some kind of localized transition leading to the OSL emission. In other words, the OSL traps are type-#1 traps, which are spatially close to an RC. According to the previous discussion an interactive scheme should be assumed, say, electrons excited into the CB during the TL experiment might either recombine with holes trapped in an RC or be retrapped by any of the two electron traps. By means of these assumptions all the observed results can be qualitatively described with ease. Indeed, it can be predicted that no OSL signal is expected after annealing an irradiated K_2YF_5:Pr^{3+} crystal at 420 K, since all electrons in trap #1 have been thermally released. On the other hand, the optical stimulation partially depletes trap #1 resulting in a decrease of the intensity of the low temperature peak in the residual glow curve. The high temperature peak is also less intense, since less electrons from trap # 1 are available to be trapped by trap #2 during the recording of the residual TL curve.

For this model to be valid it is required that part of traps #1 be spatially close to RCs in order to permit the localized recombination leading to OSL. This kind of situation has been already observed in other materials and is likely to occur also in the case of K_2YF_5:Pr^{3+}. For instance, Bos et al. (2010) reported that the Sm^{3+} cations in YPO_4:Ce^{3+}, Sm^{3+} trap electrons during irradiation to form Sm^{2+}. At the same time Ce^{3+} becomes Ce^{4+} by trapping a hole from the valence band. During optical stimulation Sm^{2+} releases an electron, which can recombine with holes trapped in Ce^{4+} via either the conduction band or localized transition (tunnelling) with different probabilities depending on temperature and wavelength of the stimulation light. The assumption of this kind of semi-localized model has been also successful to explain the unusual increase of the intensity of the high temperature glow peak of YPO_4:Ce^{3+}, Sm^{3+} as function of the heating rate in different heating rate experiments (Mandowski and Bos, 2011). A similar model involving both localized and delocalized pathways for the electron–hole recombination process during stimulation has been put forward to explain some particular characteristics of the infrared stimulated luminescence in feldespars (Jain and Ankjærgaard, 2011). This mechanism has been found to describe accurately the photo-stimulated emission in BaFCl:Eu^{2+} and BaFBr:Eu^{2+} (Hui et al., 1999; von Seggern et al., 1988).

In this context it is worth mentioning that the role played by rare-earth cations doped into K_2YF_5 have been studied by electron paramagnetic resonance (EPR) experiments. In particular, Zverev et al. (2011) identified at least three radiation-induced paramagnetic centers in K_2YF_5:Tb^{3+}, which are Tb-related, say, their EPR signal does not show up in nominally pure K_2YF_5. They also found that Tb^{3+} doped in K_2YF_5 captures a hole during irradiation to form Tb^{4+}. Zverev et al. (2011) concluded that some of these hole traps could be associated with a nearby unknown defect compensating the effective charge $+e$ of Tb^{4+} at an Y^{3+} site. By taking into account the results reported by Dorenbos et al. (2011) the same behavior could be expected for Pr^{3+} doped into K_2YF_5. Indeed, it is not unlikely that similar kinds of defect complexes like those observed in K_2YF_5:Tb^{3+} could also be present in K_2YF_5:Pr^{3+} thus providing a pathway for the localized transitions that are responsible of the OSL emission, as considered in the suggested model.

More evidence linking OSL to localized transitions could be obtained by means of correlated OSL/photo-conductivity experiments (Whitley and McKeever, 2000). In the same sense, thermally stimulated conductivity experiments could be of help in order to experimentally determine the role of delocalized transitions in TL. In this context, the results shown in this paper could be taken as a step forward in understanding the charge transfer process leading to stimulated luminescence in irradiated K_2YF_5:Pr^{3+}, which should be enriched by future experimental work.

4. Simulations

The goal of this section is to analyze whether the main conclusions of the previous section, i.e., localized (delocalized) transitions for the OSL (TL) process, make possible to describe the most relevant aspects of the experimental findings.

4.1. Model

By considering the previous discussion we have assumed a model consisting of two interactive traps (trap #1 and #2), one recombination center (CR) and one thermally and optically disconnected trap. The different transitions and energy levels are shown in Fig. 3.

For a correct analysis of the whole irradiation-induced process, the irradiation and relaxation stages prior to the processes of thermally/optically stimulated light emission have been also simulated.

4.1.1. Irradiation and relaxation

The set of rate equations describing the irradiation stage are:

$$\frac{dn_1}{dt} = \beta_1(N_1 - n_1)n_c, \tag{1}$$

$$\frac{dn_2}{dt} = \beta_2(N_2 - n_2)n_c, \tag{2}$$

$$\frac{dn_c}{dt} = X - \gamma n_c h - \beta_1(N_1 - n_1)n_c - \beta_2(N_2 - n_2)n_c, \tag{3}$$

$$\frac{dh}{dt} = -\gamma n_c h + C(H - h)n_v, \tag{4}$$

$$\frac{dn_v}{dt} = X - C(H - h)n_v, \tag{5}$$

$$h + n_v = n_1 + n_2 + n_c + M, \tag{6}$$

where n_1 and n_2 are the concentration of electrons in traps #1 and #2 respectively; N_1 and N_2 are the corresponding trap concentrations; h is the concentration of holes in the RC. H stands for the concentration of RC and M for the concentration of thermally and optically disconnected deep traps. These traps are considered already filled before irradiation accounting for the fact that the dose response of the material is constant. This kind of traps could appear in the material during its fabrication, for instance, as the result of charge compensation processes. n_c is the concentration of electrons in the CB and n_v the concentration of holes in the valence band (VB), t is the time. X is the electron–hole pair production rate due to ionising radiation. In this model, an electron in the CB has

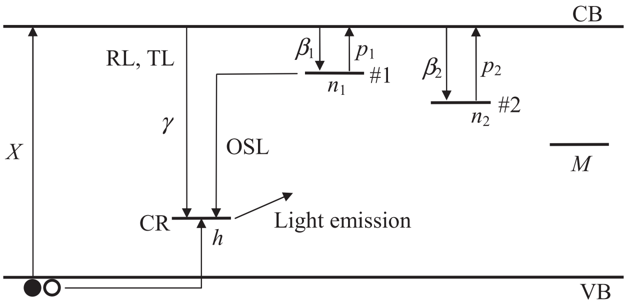

Fig. 3. Energy levels and transitions involved in the model considered in this paper. β_i are the trapping probabilities corresponding to each trap and $p_i = s_i\exp(-E_i/kT)$ the probabilities of a trapped electron being thermally released. γ is the recombination probability, M the concentration of trapped electrons in the thermally and optically disconnected traps, h the concentration of holes in the RC and X the electron–hole pair production rate induced by ionizing radiation. Further details can be found in the text.

a probability γ of recombining with a hole trapped in an RC and a probability β_i of being trapped by trap #*i*. Similarly, a hole in the valence band can be trapped by an RC with a probability *C*.

In order to simulate the relaxation stage, say, the traffic of charges leading to the emptying of the CB and the VB once irradiation has ceased, the set of Equations (1)–(6) have been considered with the assumption of no irradiation ($X = 0$). Thermally activated transitions from traps to the CB have been considered negligible at room temperature at these stages.

4.1.2. Residual TL

For the reasons previously presented, we have assumed that when an irradiated K_2YF_5:Pr^{3+} crystal is illuminated part of the charges populating trap #1 directly recombine with holes trapped in the RC without going through the CB. In particular we have considered that a concentration $n_{1,OSL} = fn_1$ $(0 < f < 1)$ of occupied traps #1 are exhausted by optically stimulating the sample for a time long enough. For the simulation of the residual TL the values of n_1 and n_2 obtained after optically stimulating an irradiated crystal have been employed as initial values for simulating the TL experiment.

The equations describing the emission of light during thermal stimulation are:

$$\frac{dn_1}{dT} = \frac{-s_1 n_1}{q}\exp(-E_1/kT) + \frac{\beta_1}{q}(N_1 - n_1)n_c, \tag{7}$$

$$\frac{dn_2}{dT} = \frac{-s_2 n_2}{q}\exp(-E_2/kT) + \frac{\beta_2}{q}(N_2 - n_2)n_c, \tag{8}$$

$$\begin{aligned}\frac{dn_c}{dT} = {} & -\frac{\gamma}{q}n_c h + \frac{s_1 n_1}{q}\exp(-E_1/kT) - \frac{\beta_1}{q}(N_1 - n_1)\, n_c \\ & + \frac{s_2 n_2}{q}\exp(-E_2/kT) - \frac{\beta_2}{q}(N_2 - n_2)n_c,\end{aligned} \tag{9}$$

$$\frac{dh}{dT} = -\frac{\gamma}{q}n_c h, \tag{10}$$

$$I_{TL} = -c\frac{dh}{dT}, \tag{11}$$

$$h = n_1 + n_2 + n_c + M, \tag{12}$$

An electron in trap #*i* has a probability $p_i = s_i\exp(-E_i/kT)$ of being thermally released from the corresponding trap (E_i and s_i are the activation energy and the frequency factor of the thermally assisted process respectively). *k* stands for the Boltzmann constant. We have assumed that the temperature is increased linearly from room temperature T_0 according to $T = T_0 + qt$. Constant *c* depends on the detection system and $I_{TL} = I_{TL}(T)$ is the glow curve. As usually found in the literature, the intensity of the thermally stimulated emission has been expressed as function of temperature rather than as a function of time.

4.2. Simulation results and discussion

The different sets of equations corresponding to each stage of the physical process have been solved by means of the Mathcad differential equation solver, Rkadapt (4th-order Runge Kutta method, with adaptive step-size). Starting from an initial set of values for the different parameters final estimates have been obtained manually by means of step-by-step changes until achieving an acceptable matching between the experimental curves and the simulated ones. It is important to emphazise that no automatic optimization algorithm has been employed, since the main goal of this simulation was to assess the capability of the model to describe qualitatively the experiments.

Table 1
Parameter values employed in the simulations.

Parameter	Value
E_1 [eV]	0.82
s_1 [s^{-1}]	2.8×10^9
β_1 [$cm^3\ s^{-1}$]	9.0×10^{-13}
N_1 [cm^{-3}]	1.0×10^{13}
E_2 [eV]	0.99
s_2 [s^{-1}]	1.7×10^9
β_2 [$cm^3\ s^{-1}$]	3.5×10^{-13}
N_2 [cm^{-3}]	$1.5 \times N_1$
γ [$cm^3\ s^{-1}$]	1.0×10^{-13}
M [cm^{-3}]	$1.45 \times N_1$
C [$cm^3\ s^{-1}$]	1.0×10^{-10}

By following Pagonis et al. (2006, Appendix A) the electron–hole pair production rate in our case has been estimated according to the expression: $X = \rho\dot{D}/W$, where $\rho = 3.1$ g cm^{-3} is the density of K_2YF_5 (Dorenbos et al., 1993), $\dot{D}$ is the dose rate employed for the experiments and *W* is the energy deposited per electron–hole pair created. We have considered $\dot{D} = 0.024$ Gy min^{-1} although it must be noted that this value was actually measured in tissue equivalent conditions (see Section 2). A value $W = 1.5 \times E_g$ has been employed for the energy per electron–hole pair, $E_g = 10$ eV being an estimate of the bandgap energy of K_2YF_5 (Kollia et al., 1998). Under these assumptions the value $X = 5 \times 10^{11}$ cm^{-3} s^{-1} has been employed for the simulations.

The whole set of parameters eventually employed for the simulations are listed in Table 1. The selected values are within the expected ranges found in the literature (Marcazzó et al., 2007). It has been assumed that during optical stimulation 70% of the electrons occupying trap #1 recombine with RC via localized transitions, say, $f = 0.7$.

The experimental signals shown in Fig. 2 have been simulated by taking into account the experimental conditions in each case, the resulting curves being depicted in Fig. 4. By comparing both figures it is apparent that the mathematical model presented before and the set of parameters listed in Table 1 predict acceptably the main behavior of the glow curve, the residual glow curve, and the

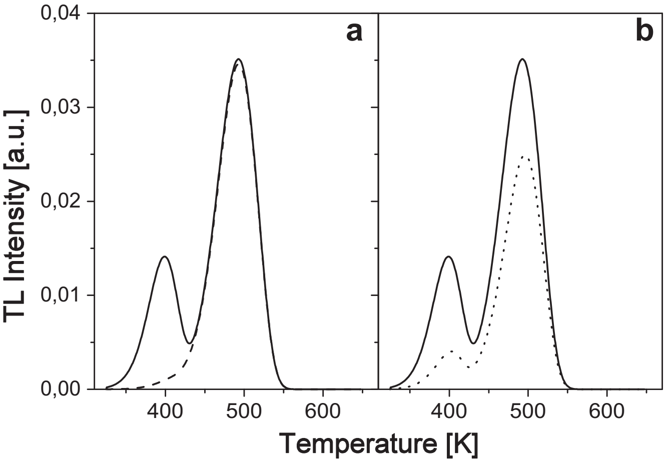

Fig. 4. Simulated glow curve of a K_2YF_5:Pr^{3+} crystal irradiated with 0.24 Gy (solid curve in both subfigures). This curve is compared with a) the simulated residual TL (dash) measured after thermally cleaning the low temperature peak (annealing at 420 K) and performing an OSL reading, and b) the residual TL measured after 180 s of optical bleaching (dot). These curves should be compared with the experimental curves shown in Fig. 2.

glow curve recorded after annealing at 420 K. In particular, the position and width of peak #1 and #2 in the simulated glow curves correspond very well with those in the experimental ones. As can be seen in Table 1 strong interaction among traps is expected by taking into account the comparable value of the retrapping and recombination parameters β_i and γ.

The activation energies rendering best results in the present simulations ($E_1 = 0.82$ eV, $E_2 = 0.99$ eV) are close to those found by Marcazzó et al. (2008), who deconvolved the glow curve of K_2YF_5:Pr^{3+} considering a model rather similar to that proposed in the present work but assuming some simplifying assumptions. In particular, Marcazzó et al. (2008) arrived to the values: $E_1 = 0.91$ eV and $E_2 = 1.05$ eV respectively.

Although the strategy for tunning up the parameter values has not been optimized, the final set of parameters (Table 1) describe acceptably the main trends of the optical bleaching of the TL in K_2YF_5:Pr^{3+}. The results of the present work are not conclusive about the nature of the defects and kind of transitions involved in the physical process but provide a clue for further investigations.

5. Conclusions

In this article a physical model has been put forward, which accounts for several experimental observations regarding the optical bleaching of irradiated K_2YF_5:Pr^{3+} crystals. The model considers two interactive electron traps, which are responsible for TL. It is assumed that during thermal stimulation both traps recombine with holes in the recombination centers via delocalized transitions. When an irradiated sample is illuminated part of the electrons in the trap being responsible for the low temperature glow peak recombine with trapped holes in the recombination centers via localized transitions. The last assumption is central to explain the facts that no OSL is observed in an irradiated K_2YF_5:Pr^{3+} crystal, whose low temperature peak has been thermally cleaned, and, that both glow peaks reduce their intensity if the irradiated crystal is illuminated beforehand.

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